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ICELAND SPAR AND ITS INFLUENCE ON THE DEVELOPMENT OF SCIENCE & TECHNOLOGY IN THE PERIOD 1780–1930

ICELAND SPAR AND ITS INFLUENCE ON THE DEVELOPMENT OF SCIENCE AND TECHNOLOGY IN THE PERIOD 1780 – 1930: NOTES AND REFERENCES

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Summary

In 1668, King Frederik III of Denmark and Iceland ordered arrangements to be made for the collection of large transparent crystals which had been noted at a site on the Reydarfjördur fjord in Eastern Iceland. In the following year, R. Bartholin in Copenhagen published a 60-page booklet describing their properties.

This booklet is generally acknowledged as a major milestone in the emergence of both crystallography and optics as scientific disciplines. Among the properties of Bartholin's material was the appearance of a double image when objects were viewed through it. A few decades later, this double refraction of the Icelandic crystals was discussed in treatises on light by C. Huygens (who considered light to be a wave motion in a very tenuous "aether") and I. Newton (who described it as a stream of emitted particles). Both realized that their theories had to be modified to account for the behavior of light in this material (a variety of calcite which came to be known as Iceland spar) and in other doubly refracting crystals such as quartz.

Not much happened in this field of optics during most of the 18th century. From about 1760 Iceland spar and other crystal forms of calcite played an increasingly important role in the development of crystallography (T. Bergman, R.J. Hauy, and many others). It also generated much interest among mineralogists, regarding for instance concepts like di- and isomorphism (E. Mitscherlich).

Following sporadic publications after 1760 (such as by E. Malm, B. Martin, and G. Monge), Iceland spar revived general interest in theories of light around the turn of the century (e.g. R.J. Hauy, W. Wollaston, T. Young). E. Malus' observation in 1808 that light became "polarized" by reflection as well as by passage through Iceland spar, led to much new research on optical phenomena by brilliant scientists like J.B. Biot, F. Arago, D. Brewster and A. Fresnel. The realization by Young in 1817 that light was a transverse wave opened up a new dimension in optics. Interference effects in polarized light, as well as optical activity (rotation of the plane of polarization during the passage of light through various compounds) sparked additional interest. In the first half of the 19th century, observations on the polarization of radiated heat by Iceland spar and other means (J.E. Bérard, M. Melloni, J.D. Forbes) contributed to its acceptance as being of the same kind as visible light.

In 1820-50, much progress was made in the theory of elastic wave motion in matter (S.D. Poisson, C. Navier, A. Cauchy, G. Green, F.E. Neumann, J. MacCullagh, G.G. Stokes and others) but a reasonably satisfactory theory for propagation of purely transverse waves in the aether within material media was not developed until after 1880. Measurements of the refracting and reflecting properties of Iceland spar were made with increasing accuracy through the 19th century (e.g. by J. Grailich, A. Cornu, and R.T. Glazebrook), in order to test theoretical predictions by Huygens, Fresnel and others.

In optical research it was important to separate light vibrations distinctly into its orthogonal components. Attempts to do this included specially cut wedges of Iceland spar and quartz crystals (Rochon prisms, etc.), dichroic materials (chiefly tournaline), and inclined piles of glass plates, but the winner was the compound Iceland spar prism invented by W. Nicol of Edinburgh in 1829. Nicol prisms in various modifications were subsequently incorporated into specialized optical instruments, the most common types being polariscopes or polarimeters (for analysis of elliptically polarized light, from about 1840), petrographic microscopes, and photometers, see below. These instruments were widely used by physicists and others studying fundamental questions of optics, interactions between light and matter, luminous phenomena, vision, etc.

Research on the optical activity of organic liquids and solutions which had been initiated by J.B. Biot before 1820, led to much new knowledge about these compounds, especially sugars and processes involving them (hydrolysis, fermentation, etc.). Further work in that field especially by L. Pasteur from 1848, later opened up a whole new branch of chemistry, stereochemistry. Polarimetric measurements continued to be of great value in studying the structure of many types of molecules, through the 19th and 20th centuries.

Around 1850, Iceland spar was important to the experimental and theoretical development of various aspects of anisotropy (i.e. directional dependence), which characterizes properties like hardness, thermal and electric conduction, electric and magnetic susceptibility, and elastic parameters of crystals. Here, the names of E. Mitscherlich, H. Senarmont, M. Faraday, H. Knoblauch, W. Thomson, W. Voigt and many other scientists may be mentioned.

Microscopical studies on crystals and thin sections of rocks, which utilized polarized light more and more after 1860 (H.C. Sorby, F. Zirkel, A. Des Cloizeaux, H. Rosenbusch, A. Michel-Lévy) revolutionized the fields of crystallography, mineralogy and petrology. Apart from Nicol prisms (in many modifications), Iceland spar was used in various accessories in this research, such as dichroscope prisms, Savart plates, Calderón plates, and later rotatable compensators. Several manufacturers of optical equipment developed standard petrographic microscopes as well as more specialized devices for mineralogy, incorporating these components. These microscopes also found a variety of uses in amateur science (from the 1840s), chemistry and chemical industries, agriculture, textiles, food science, biological research especially in histology (G. Valentin, T. Engelmann, W.J. Schmidt), and so on. Reflected-light microscopes, introduced after 1900 (by J. Königsberger, F.E. Wright and others, with later technical innovations by e.g. H. Schneiderhöhn and M. Berek) for studying properties of ore minerals and metals with the aid of Iceland spar prisms and accessories, became increasingly popular in the 1920s. Knowledge of crystal structures deduced from microscope observations has been central to many technical advances, e.g. in telecommunications.

Faraday's discovery in 1845 that magnetic fields could affect the state of polarization of light in materials, demonstrated a link between magnetism and light. This was probably a factor influencing J.C. Maxwell in his development of an electromagnetic theory of light (early 1860's), and the acceptance of this theory by physicists may have been hastened by the discovery by J. Kerr of other electrical and magnetic effects on light in the mid-1870s. Polarization also was a key concept in research on scattering of light by particles, e.g. with respect to the blue color of the sky (J. Tyndall, Lord Rayleigh). Calcite prisms and lenses played a part in the development of atomic spectroscopy in the late 1800's, especially in the ultraviolet (W. Huggins, A. Cornu and others). Around the turn of the century, measurements of light polarization were important in the study of additional electro- and magneto-optic effects. These included the Zeeman effect (1896), which indicated that electrons were the chief emitters of light. This effect was later strongly involved in the application of quantum theory to atoms and molecules, as well as giving new insight into astrophysics. Several other polarization phenomena (found e.g. by A. Cotton and by J. Stark) also were useful in studies of atomic and molecular structure. After 1865 the artificial generation of twinning and slip planes in Iceland spar by mechanical pressure, also increased the understanding of crystal physics. In crystallographic work on the various forms of calcite, however, specimens from other localities were studied in much more detail than Icelandic ones. Research on the etching effects of various fluids on surfaces of Iceland spar and other minerals (mostly after 1880), helped in research on symmetry of crystals.

Iceland spar was used considerably in the 19th and early 20th century as a source material in atomic weight determinations for calcium and in other chemical research. In investigations of the properties of X-rays, as well as in the use of X-rays to study crystal structure from 1913 to at least 1950, Iceland spar crystals (from Iceland and elsewhere) were used by many (such as M. Siegbahn, A.H. Compton) as a reference material for wavelengths because of their unusual purity and regularity. This research contributed to our knowledge of atomic structure and to the development of various aspects of modern physics such as quantum mechanics and relativity. Iceland spar crystals were also involved in the precise evaluation of fundamental quantities of physics including the electron charge and Planck's constant.

Among still other fields benefitting greatly from polarized light were studies of mechanical stress and strain (photoelasticity), of suspensions and colloids (nephelometry, flow birefringence), of liquid crystals, and of metal surfaces and thin films (ellipsometry). Observations on optical rotatory dispersion of various compounds, along with infra-red reflectance measurements (Rest-strahlen method of H. Rubens) contributed to the understanding of molecular oscillations. High-speed light shutters incorporating Kerr cells and Nicol prisms were used for wireless transmission of pictures, precise measurements of the speed of light (from 1925), etc.

Nicol prisms also were an essential component of various photometers (for measuring light intensity) including spectrophotometers, from around 1860 to at least 1930. These instruments were developed by P. Glan, H. Wild, F.F. Martens and others. They were used in astronomy (F. Zöllner, E.C. Pickering) and in many fields of optics research, e.g. regarding emission and absorption of light (by E. Becquerel and others), fluorescence, and scattering. Later applications included in particular chemical analysis. Modified types of these instruments were useful in the science and technology of colors, photography and dyes, for measurements of high temperatures, and in studies of visual perception (A. König).

For many decades, polarimeters were standard equipment in chemical, biochemical and medical laboratories, being especially important in the sugar industry. Among major advances in chemistry around 1900 were E. Fischer's synthesis of sugars, amino acids and other organic molecules which brought new understanding of biochemical processes, and A. Werner's work in inorganic chemistry, especially on so-called coordination compounds.

Almost all calcite used for optical research and instrumentation (and many of the crystals used in physical property studies) well into the 20th century seems to have been obtained from the single quarry at Helgustadir in Eastern Iceland. The cause of the exceptional size, lack of defects, and chemical purity of these crystals is connected with the presence of a Late Tertiary volcanic center in the area (mapped by G.P.L. Walker in the late 1950's). Probably, crystals were mostly picked from the soil or surface outcrops until the 1850's, when organized mining and exporting started. The heyday of activities was in 1863-72, when the quarry was leased by C.D. Tulinius, a merchant in Eskifjördur. The site was fully owned by the State from 1879. Extraction of crystals was only intermittent from 1873 until 1914, causing (after 1880) "famine" conditions for many scientists and instrument makers in need of the material. In Europe, much effort was spent on ways to economize in the use of Iceland spar and to discover or invent substitutes.

Apparently, new sources of optical crystals in commercial quantities were located in South Africa in the early 1920's, soon after the Icelandic government had re-opened the Helgustadir

quarry at considerable expense. After 1925, not much optical-quality material seems to have been extracted there, but inferior calcite material from the tailings of the quarry was used extensively in a surface finish for buildings between the 1930's and early 1960's. A smaller calcite deposit at Hoffell in SE-Iceland was worked intermittently in 1911-39. The Helgustadir quarry and its immediate surroundings were declared a protected area by the Government of Iceland in 1975, including a ban on removal of mineral specimens.

"Polaroid" dichroic materials patented in the U.S. around 1933 and later improved, greatly reduced the optical industry's dependence on calcite but the supply of optical-quality material was still insufficient for decades. The demand for the calcite crystals increased again after 1960, due among other things to the advent of lasers in optics research. Many deposits of Iceland spar are now being exploited commercially, e.g. in Russia, Southern Africa, Mexico, the U.S. and Brazil.

1 Introduction

Motto: "...Iceland spar, this incomparable mineral... the cornerstone of our knowledge of crystal physics..." (Tschermak 1881, p. 101)

1.1 On this compilation

At the farm Helgustadir north of the fjord Reydarfjördur in East Iceland there lies a small disused quarry. The word "quarry" seems appropriate here, as "mine" generally refers to underground workings. The quarry was for at least two and a half centuries the world's chief source of a certain variety of crystals of calcite (CaCO₃). By the late 18^{th} century they were well known among mineralogists and collectors by the name Iceland spar (spath d'Islande), while Icelanders called them silfurberg (silvery rock).

Although the quarry has been protected by law since 1975, no one is taking care of it. In the recent geographical, historical and tourist literature about the region, it is generally only stated in brief terms, that the crystals were exported for use in optical equipment or for displaying at mineralogical museums. Various less accurate statements on these uses may also be found in the above type of literature (section 40.4). Sporadic papers have described the nature of the quarry and of these crystals, for instance some by Th. Thoroddsen around 1900, a couple in the Journal of the Society of Engineers in Iceland (Eiriksson 1922) and in the natural history journal Nátturufraedingurinn (Thordarson 1945), etc. None of these publications quoted any other papers describing applications of the material.

I began in 1995 to study the history of the Helgustadir quarry and the crystals it produced. This came about while I stayed for a few months in Madison, Wisconsin where my wife had obtained a temporary academic position. The University of Wisconsin did not have facilities for the geophysical research that I have been carrying out here in Iceland. Instead, I decided to look up in the University's excellent libraries some information regarding past connections of Iceland with the history of the natural sciences, such as foreign scientific expeditions to the country and studies on its minerals. I soon came across references to Iceland spar in various 19th century journals in these libraries, and I have continued to collect them. The first brief paper describing my findings appeared in a symposium volume of the Icelandic Physical Society (Kristjansson 1996), and the first Icelandic edition of this report was issued by the Science Institute of the University of Iceland five years later (Kristjansson 2001a). A second edition of more than twice its size was prepared in 2007, and papers on various aspects of the Iceland spar story have been published in the meantime (e.g. Kristjansson 2001b, 2002, 2003). This English version of my report is essentially a transla-

tion of the second Icelandic edition but it contains additional information, illustrations and references within most of its sections.

My knowledge on Iceland spar has been acquired in various ways: i) by photocopying papers (or parts of papers) in journals accessible in libraries. Again the University of Wisconsin libraries deserve special mention, as I have visited Madison several times since 1995. Other university libraries which I have found useful include those in Copenhagen, in Lund, and at the M.I.T. ii) by printing out papers that have been scanned and made available on the Internet. My prime source of this type has been the website gallica.bnf.fr operated by the French National Library. It has also been very helpful to be able to look up and download material from journals that their publishers or others have made freely available. iii) by ordering photocopies through inter-library loan services, mostly at the University of Iceland. iv) by purchasing books from antiquarian shops and dealers.

The period covered in this compilation is essentially from 1780 to 1930. My emphasis has been on the applications of Iceland spar in experiments and in optical instrumentation, and on the impact that these applications often had on the development of different fields within science and technology. Obtaining information on quarrying operations at Helgustadir and the recovery of Iceland spar crystals elsewhere has not been a priority. However, some notes on these aspects (based mostly on government documents accessible in the National Archive of Iceland, old newspaper items, and biographical accounts) appear in sections of the report. I have not looked for documentation on for instance the exporting of Iceland spar, the trade in these crystals, or the manufacture of instruments containing them. This awaits the appearance of historians interested in such subjects.

I wish here to express my thanks to Geirfinnur Jonsson and to my son Kristjan for help in scanning pictures from various books and preparing them for my reports and papers. Kristjan and his wife Hildigunnur Sverrisdottir provided valuable assistance in the production of some of these pictures in A1 poster format, for a small exhibition held in 2005. Kristjan also did the layout work for the second edition of the report in 2007. My wife Elin Olafsdottir has provided much information regarding in particular aspects of organic chemistry, biochemistry and medicine. The costs of these investigations have largely been carried by by myself and the Science Institute of the University of Iceland, but contributions from various quarters towards for instance travel expenses have also been very helpful.

1.2 The impact of Iceland spar on the natural sciences

One main question to be answered by my compilation efforts, concerns the uses to which Iceland spar crystals, and equipment incorporating these, were put by scientists. Then, I try to assess the contribution that the resulting publications made to various fields of science. As mentioned above, the period covered is from 1780 to the 1920s, but many developments occurring in the preceding and succeeding decades are also mentioned. During this time, enormous progress has been made across a wide frontier of natural science and technology, and it has profoundly influenced the activities and environment of mankind.

Iceland spar crystals appear in surprisingly many different roles in this story, and were often essential to the success of experimental studies. As the term "Iceland spar" commonly has been used to designate large transparent crystals of calcite regardless of provenance, it is not always certain whether the crystals being described in particular instances came from Iceland. However, it seems that those from Iceland were of such superior quality, that scientists carrying out critical research would have been wasting their time using material from elsewhere. Sometimes it is even emphasised in secondary references, such as the chapter reviewing properties of calcite in the major mineralogical work by Hintze (1930), that Icelandic crystals were being studied even if the primary publications have omitted to mention it.

The many roles of Iceland spar just mentioned may perhaps be split into three categories:

Firstly, a great many experiments were made on, or with the aid of, Iceland spar crystals by themselves. Throughout the 19th century and longer, some of these experiments created much interest and discussion, which directed the attention of scientists towards fundamental aspects of optics, crystallography, mineralogy, physics of the solid state, and various other disciplines.

Second, calcite and Iceland spar in particular became a "type example" in theoretical work and education on important properties of crystals such as their anisotropy, as well as on the properties and propagation of light.

Third, prisms from Iceland spar were a vital component of many types of optical instruments. Beginning around 1840, many thousands of these instruments were manufactured and purchased for the scientific study of various natural phenomena, as well as for practical applications in industry and technology.

With respect to these matters and particularly the third type of roles, it needs pointing out that physicists collect evidence on the nature of the material world by many different methods. They investigate how materials respond to applied pressure, temperature changes, electrical and magnetic fields, sound waves, chemical effects of other materials, and so on. However, the various uses of light may collectively represent the most efficient modern way of probing materials. The physicist can for this purpose change e.g. the intensity of the light or the length of a light pulse, the wavelength (color) of the light, or its angle of incidence on a material surface, in addition to study-ing light emitted by the material under various conditions. After 1810 the polarization properties of light, which were most conveniently investigated with the aid of Iceland spar prisms (especially Nicol prisms from around 1840) turned out to be very sensitive to interactions with matter. These prisms have been used extensively in physics and other fields of the natural sciences since then. Additionally, Iceland spar prisms were employed in devices that measured among other things light intensity and temperature.

The raw material for the construction of these prisms and various other related components in optical equipment seems to have mostly originated at Helgustadir at least until 1900 (Appendix 1). It also seems that no single area became a larger supplier of large optical-quality calcite until after 1920.

Arguably, three naturally occurring minerals have been the most valuable ones in physics research during the last couple of centuries. These are diamond (due to its hardness and its optical and thermal properties), quartz (due also to its hardness, optical, elastic and electrical properties), and calcite (in particular Iceland spar) as will be explained in this report. Excluding special cases such as ice, the list of other minerals of particular value to physics may include graphite, common salt, zinc sulfide, and mica.

1.3 On the references and the reference list

The list of references in this report is set up in a less formal or rigorous way than in most scientific journals. The report, although based on thousands of references, is in some ways incomplete and not historically precise. Many of these references I have come across by chance when looking for other references or just browsing, and they may not be the first or the most complete account of the topic referred to in the report. However, they always contain some pertinent information, and the reference list has been augmented by many notes on where supplementary material can be found. When one is living in Iceland, obtaining copies of papers from many old foreign publications can be a slow and expensive process. I keep very long lists of papers and books which might provide additional information about the many uses of Iceland spar, but that information is likely to be to a large extent "more of the same". One is also deterred somewhat by experience: many of the publications with promising titles which I have purchased in the hope of finding valuable new data or illustrative material regarding Iceland spar, have in fact turned out to contain little of relevance to my studies.

An enormous number of papers and books concerning the topics dealt with in this report (polarized light, double refraction, crystallography, etc., etc.) appeared in the period covered by the report. The reference list therefore only constitutes a very small subsample of all the publications available on these topics, but it hopefully conveys an idea of their great breadth. Many aspects of the story deserve closer attention by historians of science. Later accounts such as reviews or biographies do not always agree upon who was the first to introduce an idea, an instrument, or a method, on on what occasion this happened. I have not had the opportunity to follow up on most of such conflicting accounts, but advice from readers regarding corrections or additional material would be most appreciated.

In the 19th century it was quite common for the same scientific results to be reported through two or more different channels. Thus, an abbreviated version of a paper in Annales de Chimie et de Physique might appear in the Comptes Rendus of the Academy of Sciences, with translations following in Annalen der Physik, the Philosophical Magazine, the Edinburgh (New) Philosophical Journal, and so on. These accounts were sometimes published more or less simultaneously, while in other cases there were lag times of up to years. Years might also pass between the submission or reading of a paper at an academy or scientific society, and its publication. More abbreviated accounts (Referate, etc.) of the contents of papers in other journals commonly appeared for instance in Neues Jahrbuch für Mineralogie, Zeitschrift für Kristallographie, Naturwissenschaftliche Rundschau, and Bulletin de la Société Chimique. Some journals issued special abstract volumes, including the Journal of the Chemical Society and Annalen der Physik (Beiblätter). As I have sometimes only had access to these secondary publications, the titles of articles are not always given in the original language in the present list of references. For a more complete list of the various appearances of the same paper, the reader is referred to the great work "The Royal Society Catalogue of Scientific Papers 1800-1900".

The list of references also reflects to some extent a variable accessibility to journals. Thus, in general it has been easier to obtain articles published in the U.K., France, Germany and the U.S. than in for instance Italy or Russia. Furthermore, publications that only had a limited circulation will obviously be preserved only at few libraries and not be readily available for browsing. That is especially the case with some old journals catering to specific branches of science or of industry (meteorology, sugar production, photography, illumination, strength of materials, glassmaking, microscopy, brewing, physiology,...), equipment catalogs, journals published by amateur societies, Festschrifts, and doctoral theses.

In spite of these shortcomings, I hope that the present work will give a reasonable insight into the enormously varied and often important applications of Iceland spar.

1.4 Expectations

Ever since I began presenting results from my studies on the valuable functions of Iceland spar, I have hoped that these results would spark related actions by others to terminate the decades-long neglect and ignorance in Iceland of the Helgustadir quarry. It has become more evident to me year by year, that the quarry is probably the world's single most important mineral locality, and that it has had a greater impact on human activities in the last two centuries than anything else happening in this country. For instance, the history of the quarry and of Iceland spar might be given some attention in the national educational system, in the tourist industry, as the basis of a local dedicated museum and permanent exhibition, as the subject of documentary films for international distribution, as a focus for geological and geochemical research in the surrounding area, and as a source of inspiration in design. The site is very accessible, being a short walk from a country road 10 km east of the town of Eskifjördur. It is not impossible that new occurrences of large calcite crystals remain to be discovered in areas of similar geological character; one such locality near the village of Breiddalsvik in East Iceland has been mentioned in newspapers a few years ago.

Unfortunately, most of the parties who might have the means of promoting such actions or who might benefit from them, have so far not shown any interest in the matter. The quarry has therefore languished as before. The only change at the site in the last decade has consisted in improvements of the local gravel road by the state Road Administration, with an accompanying expansion of a car-parking area and the erection of descriptive billboards.

2 On limestone, calcite, Iceland spar, and the Reydarfjördur volcanic center

2.1 Nomenclature

Rocks in the earth's crust generally consist of crystals of various minerals. Each type of rock, such as basalt, contains a characteristic set of minerals. Some of these minerals are primary, i.e. originating with the rock. Others may have been formed at a later date, during secondary alteration processes. A pure mineral can be composed of a single element (e.g. diamond) but most are chemical compounds of two to several elements. Natural minerals generally contain admixtures and impurities which may for instance change their color. Around 4000 minerals are known but the great majority occur very rarely: about 10 make up altogether 95% of the Earth's crust.

In the choice of a name for a mineral in the 18th century and earlier, the appearance and other properties of the respective crystals often played a part. The old expression "spar" or "spath" seems to have referred to minerals whose crystals could be easily cleaved, were light in color and lustrous. In the great French Encyclopaedia (published in 1751-80) the Swedish mineralogist Wallerius is said to have defined nine types of spar. Most minerals with -spar names are metal carbonates, but exceptions include fluorspar and feldspar.

Among the commonest crystalline materials in the crust are silica (silicon oxide, SiO2) which includes quartz in its various modifications such as rock crystal, and calcium carbonate (CaCO3) which crystallizes in two different ways (see section 5.3). The better-known of these types was early on called carbonate of lime or calcareous spar, often abbreviated to calc-spar. In German the corresponding names were kohlensäure Kalkerde and Kalkspath (later: Kalkspat), in French chaux carbonatée or spath calcaire. The shorter name of calcite which was given to a rare mineral in 1836 (see Des Cloizeaux 1843) gradually became attached to this one. Limestone and chalk are names used in a wider sense to cover all kinds of rock formations from calcium carbonate, such as sediments composed of shells of marine organisms.

Silfurberg is the Icelandic name for calcium carbonate which in volcanic formations is generally formed as a secondary mineral during hydrothermal alteration. Large clear crystals of this mineral were called Iceland crystal in the 17th and 18th centuries, later Iceland spar (isländischer Doppelspat, spath d'Islande). In the late 20th century, the alternative term "optical calcite" has also often been used.

The characteristic crystalline form of Iceland spar is a rhombohedron, with sides in the shape of a parallelogram. The angles between the sides are 105°05' or 74°55' while the angles of each parallelogram are 101°55' and 78°05'. The density of pure calcite is 2.71029 g/cc according to the review by Birge (1941).

2.2 The Reydarfjördur central volcano, and its Iceland spar deposit

The volcanic geology of Iceland (as now exposed above sea level), spans about 15 million years. Eruptions of these volcanics chiefly took place in so-called central volcano complexes. These are similar in character to those in the older Tertiary areas of the British Isles but smaller in size, of order 5-10 km. Each center may have been active for around half a million years. The geologist G.P.L. Walker (1959) who mapped the geological formations around Reydarfjördur, made the important observation that such volcanic centers occur in East Iceland. This had not been stated clearly before, and in fact Walker only mentions explicitly in a paper in the following year, that the eroded remains of one of these volcanoes are situated in Reydarfjördur. It is elongated north-south and spans the outer part of the fjord. A dike swarm belonging to this volcano can be traced to the nearby fjords Mjoifjördur to the north and Faskrudsfjördur in the south. Many sill-type intrusives are also noted within the Reydarfjördur volcano, being most dense in an area about 6 km across (Walker 1974).

The doctoral thesis (Gibson 1963) of one of Walker's graduate students deals with the Reydarfjördur volcano, but it does not cover the secondary alteration of its strata in any detail. Gibson only shows that the area of greatest alteration (propylitized zone) reaches the shore both at Helgustadir north of the fjord and on the south side of the fjord. Evidence of a caldera collapse, observed in many of the central volcanoes of Iceland, does not seem to have been noted in Reydarfjördur. The age of that central volcano may be estimated at some 11 million years.

In and around central volcanoes, advanced hydrothermal alteration has been caused by circulating hot water which dissolves some of the constituents of the rock and deposits them in cracks and vesicles elsewhere at lower temperature. The best-known minerals formed in this way in Iceland include zeolites and silica. The former belong to a large class of hydrated compounds of sodium, potassium and other metals, while the silica is usually in the form of opal, chalcedony and jasper rather than rock crystal. Calcite is also a common alteration mineral in Iceland, but its crystals are rarely pure or more than a couple of cm in size. Their growth has probably in most cases been irregular due to changes occurring in the chemistry, temperature and pressure of the geothermal fluids. However, in at least one locality in Reydarfjördur conditions have for a long period of time been stable enough for large, pure and regular crystals to form in cavities. They are often accompanied by the zeolite stilbite, also called desmin. The crystal deposit reached the surface in a small spot at 90-100 m above sea level east of the farm Helgustadir. Then a small brook gradually broke up this exposure and carried crystals with it down towards the sea shore.

3 Iceland spar attracts attention – developments to 1700 A.D.

3.1 Collection of minerals in past centuries – Ole Worm and Icelanders

In the 16th to 18th centuries it was popular among scholars as well as noble and wealthy persons, to collect beautiful works of art or handicraft, objects from distant lands, antique relics, and specimens from Nature. The last category included minerals and fossils, see the extensive compilation by Wilson (1994). Such collections were named "Kunstkammer" or "Naturalienkabinet". One, probably the largest in Europe, belonged to the Danish polymath Ole Worm who died in 1654. Among other things, Worm had contacts with several Icelanders who sent him collectible items (see Benediktsson 1948, Schepelern 1965-68, 1971), and much of their correspondence has survived. One of the Icelanders was Gisli (nicknamed "the Wise") Magnusson who was sheriff of the counties of central Eastern Iceland from 1649. Gisli had studied Natural History and stayed in Denmark, Leiden in the Netherlands, and England. He was very interested in various aspects of the geology of Iceland, such as the mining of sulfur (which was exported for use in gunpowder). It is quite possible that Gisli or others sent Ole Worm or his son Willum pieces of Iceland spar, but nothing definite about this can be gleaned from the above correspondence. It is also not mentioned specifically in other letters from Gisli that have been preserved, for instance to his son in Copenhagen 1669-70 (Benediktsson 1939). Worm had family connections with the Bartholins, whose generations of notable scholars included Rasmus Bartholin. As described in section 5.2, Rasmus Bartholin had lived in Leiden like Gisli the Wise. After the death of Ole Worm, his collection of minerals and other objects was acquired by King Frederik III who also had a keen interest in such matters.

By chance, one of the trading posts of the Danish monopoly trade with Iceland (instituted in 1602) was situated at the inlet Stóra-Breidavik in Reydarfjördur. The site, now occupied by the farm Utstekkur, is close to the Iceland spar outcrop of Helgustadir. It is then almost inevitable that local farmers and other persons crossing the brook below the outcrop would pick up crystal specimens and bring them to the attention of staff at the post. In the period 1662-83 one of the two Copenhagen merchants licenced to trade in Reydarfjördur was Christopher Hansen, who until 1664 also was one of the four mayors of that city. He was also the father-in-law of of the famous medical doctor and anatomist Thomas Bartholin, brother of Rasmus.

Contemporary accounts of how Iceland spar was transported abroad in the period 1600-1800 may exist in archives in Denmark and elsewhere, but I have not attempted searching for these. Paragraph 11 of a decree by Frederik III in 1662 which sets out conditions for the monopoly trade, states that: "...if Our envoys to Iceland recover sulfur, saltpeter, minerals or other ores which may be found there, the licencees are obliged to load these materials on to their vessels and transport them hither at moderate cost". A similar condition may be found in a 1684 decree by Frederik's successor Christian V, when modifying the monopoly regulations: "...Minerals and other rari-

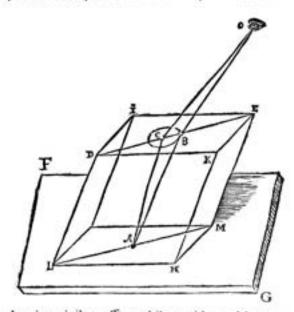


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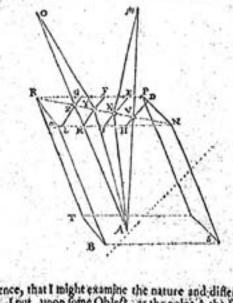
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Prederik den Tredie &c. V. S. G. T. Voris nandigste Villie og Befaling er, at I udi sex Manneders Tid til den islandske Krystal at udbryde antager og bestiller en Stenhuggersvend, og samme til maanedlig Besolding imidlertid lader bekomme 12 Rigsdaler; item en Mand til hannem om Maaneden 8 Rigsdaler; nok som vil behöves udi adskillige Maader til Arbeidet at fortsette, Penge 80 Rigsdaler; disligeste 6 Maaneders Proviant, nemlig en Skipper-Kost for Stenhuggeren selv og en Baadsmands-Kost imidlertid for den som hannem medfölger. Dermed &c. Hafn, 11. April 1668.

remains unmoved. Wherefore in this Transparent Ablitances the Insign which appears first, may proceed according to the ordinary Laws of Usual Refraction 5 but that, which moveth, and is carried about according to the motion of the Diaphanons Body. while the Object remains un film d, Ocoveth an Us agent kind of Refraction, hitherto un observed by Diap, trictant.



bus imaginibus effe mobilem : id qvod hac ratione indagare licet. In fchemate præcedentes, C 3 ma-



Hence, that I might examine the nature and difference of both, I put, upon forme Object, as the point A, the Prime of my Double-retracting Chiyfial N, P, R, Q, T, B, S; and the Eye M, being perpendicularly pointed over the upper plain of the Prime N, P, R, Q, I noted, whether there was any Refraction of the point A, (for the nore Laws of Refraction teach, that there is none.). But the Perpendicu-

Figure 3-1. Top left: The front page of R. Bartholin's (1669) essay. Right: The text of a royal decree by Frederik III in 1668, on sending a stone mason to Iceland to recover crystals. Bottom left: A drawing from Bartholin (1669) showing how B, one image of the dot A, moves when an Iceland spar crystal is rotated about a vertical axis. Right: A description of Bartholin's (1671) experiments in the Philosophical Transactions.

ties... which may be found in the country, shall be ...collected by them on our behalf and transported hither at moderate cost".

3.2 The first records of Iceland spar – Rasmus Bartholin and his 1669 essay

The first document known with certainty to mention the Iceland spar deposit, is a decree by Frederik III in the Spring of 1668 (Fig. 3-1). This document, the text of which was later (1853) included in the first volume of a collection of older legislation concerning Iceland, states that a stonemason and his assistant shall be sent for six months to collect "the Icelandic crystals". An accompanying letter quoted by Garboe (1918; Buchwald and Pedersen 1991) concerns procurement of tools for the stonemason. As indicated in the previous section, this matter must have had some events leading up to it.

Then the very important essay of Rasmus Bartholin (1669, Fig. 3-1 and 5-2) on the Iceland spar crystals was published in Copenhagen. Here he described his various observations on these, and especially on their effects upon light rays. Bartholin had become professor of geometry in 1656 and of medicine in 1657. In the entry on him in the Danish Biographical Lexicon it is stated that his researches on the Icelandic crystals (apart from their optical properties) are important enough to place him alongside his countryman Steno as a founder of crystallography. This refers to the discovery by Steno (Niels Stensen), also made in 1669, that the angles between the crystal surfaces in hematite and quartz were constant during the growth of these crystals (see e.g. Flint 1971). Lang (1868) similarly argues that the origin of proper crystallographic research is tied to the discovery of Iceland spar.

At the time of these developments, some laws regarding the refraction of a light ray at a plane material surface were already known. Among these was the fact that both the refracted and the reflected ray lie in the plane of incidence, defined by the incoming ray and a normal to the surface. Snell's law which was first published in 1637, states that for a variable angle of incidence i of a ray from empty space, the angle of refraction b obeys the relation sin $b = \sin i /n$. The constant n, known as the refractive index of that material, is about 1.33 for water and 1.5 for ordinary glass. n is also dependent on the color of the light, changing for instance in one type of glass from 1.508 for red light (700 nm wavelength) to 1.525 for violet light (400 nm). This property, called dispersion, greatly puzzled scientists for more than two centuries.

The behavior of Iceland spar was at variance with the known laws, especially in that when a rhombohedron of this crystal was placed on a sheet of white paper with a black dot on it, two dots were seen (Fig. 3-1). Both appeared to lie within the crystal as happens in the case of water or glass, but one was higher than the other. When Bartholin rotated the crystal about a vertical axis, the upper image was fixed but the lower one rotated with the crystal. This was investigated by Bartholin in clever experiments. He showed that the path of light from the fixed dot agreed with Snell's law. A ray incident on the crystal thus was split in two, and the refraction of one of these was determined by the fundamental structure of the crystal (rather than, for instance, by its lateral dimensions or by reflections from some internal planes). This "double refraction" was a totally new phenomenon.

3.3 Research by Huygens, 1670-90

Bartholin's essay reached the Royal Society of London, and an extract was published in the Society's Philosophical Transactions (Bartholin 1671, Fig. 3-1) as well as elsewhere (see Romé Del-

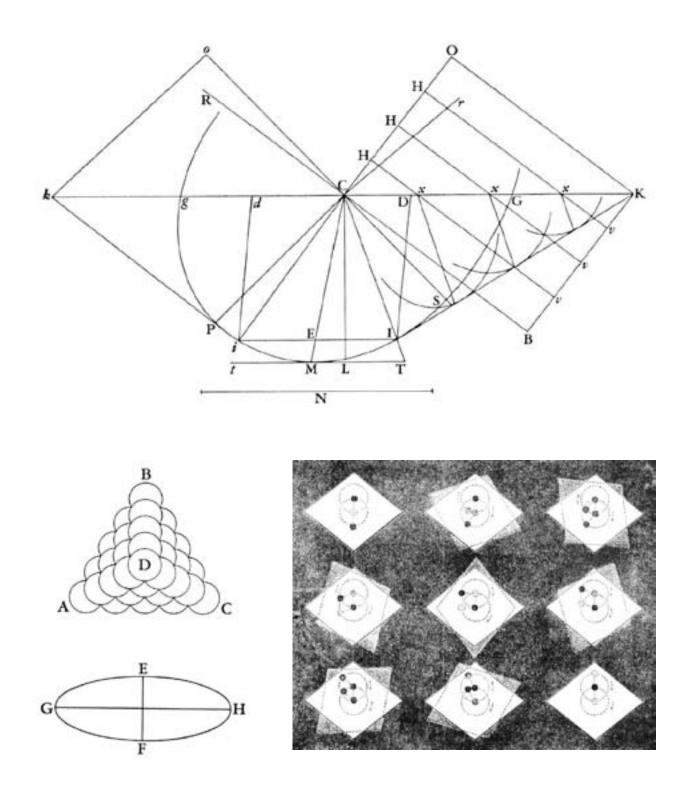


Figure 3-2. Top: A drawing by Huygens (1690) of a narrow light ray RC incident on the surface kCK of Iceland spar. It gives rise to an ellipsoidal wave inside the material, and so do also the rays Hx, Hx, Hx and OK from the same wave front. The ellipsoidal waves combine to form the (extraordinary) wave front SK which is their common tangent. Bottom left: Huygens' drawing of the composition of Iceland spar from particles. Here they are shown as spheres but he in fact considered them to be ellipsoidal, formed by rotation about the diameter EF. He also draws attention to the threefold symmetry of an Iceland spar crystal about one of its corners. These diagrams are from a 1952 edition of an English translation of Huygens' book. Right: A view of a dark dot on paper lying underneath two Iceland spar rhombs. In the first drawing, the rhombs have the same orientation. When Huygens rotated the upper one a half-turn in eight steps, he saw four or two dots of varying gray shades, and finally a single dark dot. From Paulsen (1893).

isle 1772). In a short note in another journal in 1671 (see Garboe 1949), Bartholin pointed out the necessity for further research on the properties of Iceland spar. From his stay in Leiden, Rasmus Bartholin was acquainted with the Dutch physicist C. Huygens (Fig. 5-2), and sent Iceland spar crystals to him and to some other scholars (Garboe 1949). Huygens made (around 1677, according to Garboe 1959, p. 86 and Ziggelaar 1980) remarkable measurements on these crystals which are described in detail in one of the six chapters of his book on light. Huygens presented his findings to the French Academy of Sciences in 1679 (Bruhl-Metzger 1914) but the book (Huygens 1690) was not published until more than a decade later. Shapiro (1973) and Dijksterhuis (2004) who have studied the diaries and letters of Huygens, consider the double refraction to be the central theme of the book, and even a main reason for its being published.

Huygens imagined light to be a wave motion transmitted through matter as well as empty space, in a tenuous elastic substance called aether. Inside a body of matter, the properties of the aether were to some extent governed by that of the material. It was known that the speed of light was not infinite, after observations on the motion of Jupiter's moons made by O. Rømer (assistant and later son-in-law of R. Bartholin) were published in 1676. Every particle inside a material excited by light sends out a spherical aether wave. The combined emission of such waves by many particles can form a narrow ray of light or a wide wavefront, depending on circumstances. If a particle emitting a wave is at the plane interface between two media, two hemispherical waves will emanate from it. It is easy to deduce from Snell's law that the speed of light in a medium, and hence the radius of the hemispheres, varies as the inverse of the refractive index. Light therefore behaves differently from sound in that its speed is generally slower in dense materials than in materials of lower density. Another difference lies in the fact that sound waves are not dispersed appreciably.

In his 1690 book, Huygens argued in clear terms for the very important hypothesis that in Iceland spar each excited particle emitted two waves. One of these was spherical and the other one was in the shape of an ellipsoid. This is a surface generated by rotation of an ellipse around one of its two axes. The ellipsoid would be symmetrical about a particular direction in the crystal, namely that of the line (later called the optical axis) equidistant from all the edges of the crystal where they meet at 102° angles. According to modern measurements, the spherical wave corresponds to the refractive index $n_o = 1.658$ (o stands for ordinary; the value applies to yellow light) while the other wave has a refractive index n_e (e for extraordinary) which varies from 1.486 to 1.658 depending on direction.

One thing noted by Huygens concerning light passing through a piece of Iceland spar, was that the crystal did not just separate the light ray into two fainter rays; a change in the character of the light also occurred. This could be appreciated by placing one piece of Iceland spar on top of another one with a dot underneath, and rotating the upper piece. The expected pattern of four still fainter but equal dots was not seen, cf. Fig. 3-2: instead the intensity of all the dots varied.

Various other important observations and ideas concerning Iceland spar appear in the writings of Huygens (1690, and his correspondence), for instance his hypothesis that it is composed of small ellipsoidal particles (Fig. 3-2). Some of these ideas were not followed up until the 19th century.

3.4 Newton, around 1700

Isaac Newton in England had also carried out important research on light, his results originally appearing in his book "Opticks" in 1704. He considered light to consist of particles that were emitted in straight lines away from any luminous body. He contemplated the presence of an aether

which was different from that of Huygens, but his ideas on that subject mostly went unpublished. Newton (1706, and in later editions) discusses the properties of "Island Crystal". It is not entirely clear from the text whether he himself had acquired specimens, but it is very likely that he obtained some from Huygens who visited England in 1689. During that visit, they both addressed a meeting at the Royal Society (Hall 1993, p. 131).

In Opticks, Newton (1706) puts forward some numbered queries about the Icelandic crystals. One of these contains the suggestion that light particles may have two "sides" (cf. the two poles of a magnet) that are pulled in different measure to the sides of these crystals as they pass through. According to Newton, the speed of light-particles in a material increases in proportion to its refractive index. Thus, direct measurements of this speed in transparent materials could have decided between his ideas and those of Huygens, but techniques for making such measurements were not available until 1850.

Huygens and Newton were both aware of the fact that double refraction occurs in rock crystal. However, the difference between the ordinary and the minimum extraordinary refractive index in quartz is small, only one-nineteenth of that in Iceland spar (i.e. less than 0.01 as compared to 0.17). This also applies to many other crystalline minerals. If the property had not been discovered in Bartholin's crystals, it might not have been noticed until much later. Barnard (1863) says in a review article on optics that double refraction "…is exceedingly common…it is only in Iceland spar, however, that it manifests itself in a degree remarkable enough to attract attention…, and in most cases it can only be detected by special arrangements.". Barnard also mentions the size and transparency of the spar crystals in this context.

4 Crystallography and optics, 1700-80

4.1 Not much happened regarding double refraction

During all of the 18th century and even into the 19th, most scholars did not question Newton's corpuscular theory of light, which is understandable in view of his stature in this and other fields of science. His Opticks was translated into French in 1787 by J.-P. Marat, better known for his participation in the Revolution. Marat himself wrote a book on the subject published in the following year, but it does not mention double refraction.

Apart from Newton's work, I have only come across one published essay from the first half of the 18th century which discusses Iceland spar. This essay by la Hire (1710, see Buchwald 1980) describes its properties in comparison with a type of rock found in the Paris region, and recounts the observations of Bartholin and Huygens. In an obituary of C.-F. de C. Dufay in the Mémoires of the French Academy for 1739 (p. 71-83), Dufay is said to have studied the refraction of light in cristal d'Islande, rock crystal and other transparent minerals. He was unable to complete these studies, but he had found that all crystals having 90° angles do not exhibit double refraction while others do.

It is quite likely that the behavior of light in Iceland spar was by many considered an odd curiosity without a wider significance. Various other more general properties of light called for explanations, such as its dispersion, the absorption of different colors in materials, vision, the reflection of light from surfaces, light emission by hot bodies, etc. Perhaps the only well-known European scientist of the 18th century who favored the wave theory and Huygens' aether, was L. Euler (in his 1746 book Nova Theoria Lucis et Colorum, quoted by Born and Wolf 1999, possibly again in 1769). He did not attract any followers (see Brougham 1803a) and he did not address the problem of double refraction. I have also noticed recently (in Physics Today, Oct. 2003) that Benjamin Franklin supported the wave theory.

The thesis of E. Malm (1761, Fig. 4-1) at Uppsala describes double refraction, but it is brief and to me does not appear to contain new discoveries. At a similar time G. Beccaria (1762) wrote among other things about double refraction in quartz, and he was familiar with the works of Huygens and Newton on the subject.

The next one to write about double refraction was the versatile instrument dealer B. Martin in London. He invented and constructed various optical devices, in addition to much writing and lecturing about science. See Williams (1863) and Moe (1990, in particular p. 103-110). Martin (1774) published a leaflet containing two essays on the remarkable optical properties of "Island Crystal", cf. Fig. 4-1. The essays describe his experiments on these crystals, including the making of a lens with two focal points. Martin mentions that calcite crystals are commonly found in the county of Derbyshire (but their forms are different from those in Iceland, see Naumann 1885).

I. N. J. DISSERTATIO GRADUALIS, DE REFRAC' TION CRYSTA OUAM . CONSENT. AMPLISS, FACULT, PHILOSOPH. IN REGIA ACADEMIA UPSALIENSI, MODERANTE MAG. $\mathbf{L}\mathbf{E}$ PROFESS. REG. ET ORD. REG. ACAD. SCHENT. HOLM. MEMP. PUBLICE VENTILANDAM EXHIBET, ALUMNUS REGIUS, ERICUS MALM, SUDERMANNUS.

IN AUDITORIO GUSTAVIANO DIE X. JUNIL ANNI MDCCLXI.

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§. 1.

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NATURE and wonderful PROPERTIES O F

Island CRYSTAL.

ESSAY I.

CONTAINING,

All that has been hitherto delivered in regard to its Form, Dimenfions, and double Refraction of Light; with the Addition of feveral New Experiments with polifhed PA-RALLELOPIPEDS and PRISMS made of this Subflance, which differer a various, multiple, and colorific REFRACTION; and exhibit a Variety of IMAGES of one and the fame Object.

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VARIÆ CRYSTALLORUM FORMÆ,

A SPATHO ORTÆ, EXPLICATÆ

TORBERNO BERGMAN,

CHEM. ET METALLURG. PROF. EQVIT. REG. ORD. DE WASA.

Cryftalli falinæ, lapideæ, pyritaceæ & metallicæ corporum geometricorum tantam primo intuitu offerunt varietatem, ut forma, modis fere infinitis multiplicata, nobiscum ludere videatur Natura. Hifce vero fingulis fedulo confideratis compt-

Spathum calcareum, uti notum eft, teffera conftat feu parallelepipedo obliquo, cojus omnia plana rhombeam habent figuram ejus indolis, ut anguli obtufi gradibus 101¹/₂, acuti vero finguli 78¹/₂ fint æquales. Difpiciamus jam, quomodo ex hac, fola parallelogrammorum fimilium idonea accumulatione, cryftalli generari poffint divertiffimæ.

Figure 4-1. Pages from four publications on Iceland spar and optics in 1760-80. Top: The front pages of Malm's (1761) thesis and Martin's (1774) two essays on Iceland spar. Bottom left: A page from Silberschlag's (1787) paper. Right: Front page and a paragraph from Bergman's (1773) article on Iceland spar.

Martin's work on Iceland spar does not seem to have attracted much attention among his contemporaries, according to Brewster (1822-23).

Soon after Martin's essays appeared, O. Olavius travelled around Iceland to observe the state of its economy, visiting the Helgustadir crystal site in 1776. In his report (Olavius 1780) which was for instance published in German in 1787 and 1805, he states that people have often attempted excavating this mineral, but no pieces weighing more than 10 pounds have been recovered. With the help of others he managed to collect one of 42 pounds. Olavius recounts some earlier ideas about using Icelandic decorative stones in altar pieces, and suggests that Iceland spar might be embedded in the walls of royal palaces. Another traveller on a similar mission, N. Mohr, inspected the site in 1781.

In the manuscripts of the famous chemist A. Lavoisier it is mentioned that cristal d'Islande, also called spath parallélogrammatique, was one of the compounds that he attempted to heat using sunlight and a converging lens in 1772-73. In a description of geological phenomena in the Pyrenees published in 1786, Baron de Dietrich refers to an older pamphlet which states that spath d'Islande has been found there. Finally, Count Buffon's (1786) book on the natural history of minerals contains a section on Bartholin's studies and some observations made by Buffon himself on cristal d'Islande. Buchwald (1980) states that Buffon did not grasp the significance of Huygens' research on its double refraction, and a hypothesis on the subject which Buffon put forward, was soon forgotten (Bruhl-Metzger 1914).

4.2 Crystallography, to 1780

Following the 1669 publications by Stensen and Bartholin on what Lima-de-Faria (1990) calls "the first important experimental discoveries in crystallography", new ideas about chemical compounds, crystals, minerals and rocks gradually emerged. These developments are dealt with in various books on the history of these fields in science, but here I am only going to mention a few points.

One of the main tasks of research in the natural sciences, is the classification of related subjects according to some rational system. By the middle of the 18th century C. von Linné had created such schemes for the animal and vegetable kingdoms, and attempts to set up analogous ones for the inanimate worlds of elements, compounds, crystals, minerals, fossils etc. could be expected. Linné himself (around 1735, according to Jong and Stradner 1954-56) divided crystals into classes depending on their external forms, constructed wooden models of them, and published accurate drawings of crystals. His works influenced many later scholars. A short description of Doppelspat is included in J.F. Gmelin's 1777 augmented German translation of the 12th edition of Linné's book in Latin on the mineral kingdom.

Westfeld (1767) and Bergman (1773, based on observations by J.S. Gahn; Fig. 4-1) put forward hypotheses on the structure of calcite crystals. They envisaged these to be composed of layers of rhomboeders around a small nucleus, even if the crystal itself had a different form (scalenohedral, see later). Wyckoff (1922, p. 1) considers Bergman to have "developed what might be called the first geometrical theory of crystal structure". It is very likely that they had access to specimens of Iceland spar, at least Gahn (Cuvier 1829, p. clvi). Bergman was also acquainted with it, as writings by him on its electrical properties (in Bergman 1763) have survived. A chemical analysis by Bergman in 1774 (quoted by Hauy 1801 and Hintze 1930) indicated that calcite contains 11% of water which is of course incorrect; his results may refer for instance to sinter from carbonate-rich springs which are well known in the Snaefellsnes peninsula of Western Iceland (cf. correspondence printed in U. von Troil's contemporary travel account from Iceland).

Among other mineralogists describing the Icelandic type of calcite before 1785, are Cronstedt (1758) who calls it Dubbelsten or Spatum Islandicum, and Romé Delisle (1772) who states that it is commonly known as cristal d'Islande. Werner (1774) discusses the optical properties of this Doppelspat at some length. ". A sentence in the chapter on Calx in Bergman's (1782) book on minerals begins "Rarissime omne desideratur martiale, quo etiam contaminatum clarissimum spathum Islandicum...". Around 1780 a major step forward in crystallographic science was taken when A. Carangeot constructed an instrument for measuring angles between crystal faces. This "contact goniometer" had arms which could be aligned with the faces. On the basis of Carangeot's measurements, Romé Delisle (1783) suggested that Stensen's 1669 hypothesis on the constancy of angles in quartz could be extended to all crystals.

5 Crystallographic research by Abbé Hauy and others, 1780-1830

5.1 Introduction

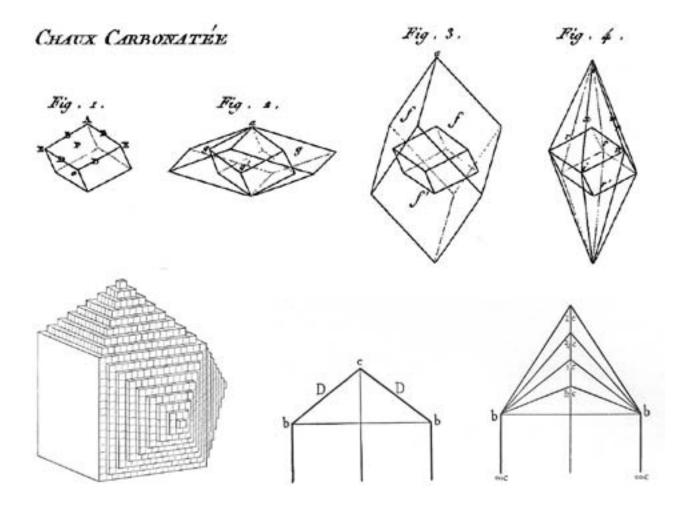
Around 1800 a gradual change was taking place in attitudes to fossils, crystals and minerals (Wilson 1994), as emphasis moved from individual large-scale collectors to scientists and to public museums and exhibits. At that point in time also, the first societies and learned journals dedicated to minerals were born. The latter part of the 18th century saw a great increase in the number of mineral purveyors living in the vicinity of rich occurrences. Mineral dealers also became numerous, for instance in cities like Vienna.

Chemists such as M.H. Klaproth and C.F. Bucholz in Germany, L.N. Vauquelin in France and J.J. Berzelius in Sweden had by 1800 perfected methods for analyzing minerals and rocks, which led to the discovery of many new elements. Analyses on calcite were published by Klaproth (1788) and by others soon after 1800, see Appendix 2. By that time people were presumably realizing that Iceland spar was composed of pure calcium carbonate.

5.2 Research by Hauy and others on crystals, 1780-1805

The beginning of a period of major progress in crystallography and optics may be marked by a single chance event, similar to the fall of Newton's proverbial apple. The abbot R.J. Hauy (Fig. 5-2) who was a keen collector of minerals, was one day in 1779 handling a cluster of calcite crystals at his friend's house. He accidentally dropped the cluster on the floor and it broke into pieces. Hauy then noticed that the angles between surfaces of these pieces were similar to those of Iceland spar, and cried: "Tout est trouvé !" (Cuvier 1829, Fig. 5-1). The simple characteristic form of the Icelandic crystals (which was rarely seen in calcite from elsewhere), had to be the fundamental one for all calcite crystals, in spite of their very diverse appearances (Fig. 5-1, top). This story indicates that Hauy was already familiar with Iceland spar. Measurements of the angles of the above-mentioned fragments, as well as on pieces from other crystals that he smashed deliberately, confirmed his conclusion (see Hauy 1784, 1798). From then on his work was mainly concerned with crystals and minerals.

Among the many scientific publications of Hauy, at least two deal specifically with Iceland spar. In one of these (Hauy 1782) which treats its crystallographic properties, he uses the expression "spath d'Islande". Other papers by Hauy (1788, 1792, 1800) on Iceland spar describe aspects of its double refraction. A lengthy section on Iceland spar may also be found in vol. 2 of Hauy's (1801) great work on mineralogy. T. Young (1802, p. 45, 1809) states that Hauy's refraction experiments on Iceland spar supported the wave theory of Huygens rather than Newton's corpuscu-



Un monde nouveau semble à l'instant s'ouvrir pour lui. Il rentre dans son cabinet, prend un spath cristallisé en pyramide hexaèdre, ce que l'on appelait *dent de cochon;* il essaie de le casser, et il en voit encore sortir ce rhomboïde, ce spath d'Islande; les éclats qu'il en fait tomber sont euxmêmes de petits rhomboïdes; il casse un troisième spath, celui que l'on nommait *lenticulaire;* c'est encore un rhomboïde qui se montre dans le centre, et des rhomboïdes plus petits qui s'en détachent.

Tout est trouvé! s'écrie-t-il; les molécules du spath calcaire n'ont qu'une seule et même forme : c'est en se groupant di-

Figure 5-1. Top: Part of a series of diagrams on calcite crystals, from Hauy's (1801) book. Center: Hauy's suggestion that a crystal grows in steps. The slope of a side in a cubical crystal can only have a value of a or a(n/m) where n and m are (usually small) integers. In two of the diagrams the slope of the pyramidal sides is 1 but in the right-hand one the possible sides with slopes 1/2, 3/2 and 2 are also shown. Bottom: A famous anecdote published in the obituary of Hauy by Cuvier (1829). It describes his reactions when he accidentally broke a group of calcite crystals and noted that the pieces had the same form as the one prevailing in Iceland spar.

lar theory of light, but not all of Hauy's results were correct (Buchwald 1989, p. 9-18). His writings on the subject were also unclear (see Gillispie 1997) and his refraction research did not lead to new discoveries (American Mineralogist 1918, p. 117). Hauy (1801) claimed to have found double refraction in 20 minerals (quoted by Brewster 1823; see also Hauy 1793). His textbook on physics (Hauy 1803) which was no doubt read by many, also contains sections on crystals and on the refraction of light in Iceland spar.

Hauy experienced his share of hardships in the troubled times following the Revolution of 1789. Later, the authorities became generous to scientists and the French made great progress in various fields of the natural sciences. Many honours were bestowed upon Hauy, as may be seen in the biographical literature, for instance Cuvier (1829; American Mineralogist 1918). He is often called the "father of crystallography" and he had a huge collection of mineral specimens.

It has been debated by historians of the subject whether Hauy was aware of the previous work of T. Bergman (Dictionary of Scientific Biography 1980-90), but Hintze (1930, p. 2897) considers that "with his studies of calcite, Hauy founded mathematical crystallography". One of his main discoveries (Hauy 1799, 1801) concerns what has been called "the law of rational proportions". It implies that the surfaces of crystals are either completely plane (i.e. cleavage surfaces) or that they are formed by microscopic steps. These steps are built up from cells (molécules integrantes) arranged in a regular fashion, for instance one step up for one step along (as on the cubical kernel in Fig. 5-1), or one step up for two steps along, etc. Another way of expressing this law was the "Zonengesetz" put forward by Weiss (1804).

The oldest use of the word Doppelspath that I have come across, is in Gerhard's (1782) book on minerals. In the following year, a piece of Icelandic Doppelspath of 12" by 5" size (in a collection in London) is mentioned in vol. 10 of J. Bernoulli's compilation of travel accounts. I have also seen a paper on Iceland spar (Silberschlag 1787) where it is called Isländischer Crystall (Fig. 4-1) and Isländischer Spath. The author has clearly been able to carry out his own experiments on specimens, and among other things he concludes that the double refraction is not caused by internal reflections.

5.3 More on crystallography: Hauy and others 1805-22

Early on, Hauy (1785) began studying the electrification of crystals and other substances caused by friction, pressure, and heating. Later he found (Hauy 1817) that Iceland spar (already investigated with respect to these properties by T. Bergman and others) acquired by rubbing and compression a greater electrical charge than most other materials, and kept it for longer periods. These results are probably discussed in the second (1822) edition of his treatise on mineralogy, which I have not had the opportunity to consult. Hauy's research was continued (from 1820 or earlier) by a young scientist, A.C. Becquerel (1823, 1827) who investigated the electrical behavior of various substances including Iceland spar (see Dictionary of Scientific Biography 1980-90). He made significant discoveries in this field, but Iceland spar turned out to be much less interesting in this regard than for instance quartz. See also section 9.2.

J.J. Bernhardi (1807) wrote a paper on double refraction in many materials, criticizing some points made by Hauy. He says that Iceland spar is well known and very suitable for research on this phenomenon, whereas the available specimens of other crystal types tend to be too small, irregular, or insufficiently transparent. He stresses the role of a direction of symmetry which he calls "Lichtaxe" and which later acquired the name "optical axis" in English. Bernhardi (1808b) also discusses the surfaces and cleavage directions of calcite, including his "purest pieces of Icelandic Doppelspath". Bernhardi (1808a) was the first to divide crystals into six groups (now



Figure 5-2. Four of the first and most distinguished scientists who carried out research on Iceland spar and polarized light. Top left: R. Bartholin (1625-1698). Right: C. Huygens (1629-1695). Bottom left: R.J. Hauy (1743-1822). Right: E.L. Malus (1775-1812). The portrait of Bartholin is taken from the 1991 facsimile edition by the Copenhagen University Library of his Iceland spar essay, but others in this report are mostly obtained from collections on Internet websites.

called systems) depending on their geometrical properties, but Weiss (1815) preferred splitting one of these groups in two. They are still in use, but each system was later subdivided into two to seven so-called symmetry classes or point groups, a total of 32. One of the system is the cubic or regular one, and Bernhardi states that all minerals belonging to it which he knows do not exhibit double refraction (as was in fact also observed by C. Dufay before 1739) nor do they have an optical axis. Weiss (1811), Cauchy (1813) and Hauy (1815) also realized that geometrical symmetry constituted an important aspect of the structure of crystals. Seeber (1824) further emphasized that repetition or periodicity was another major factor in that structure. Seeber used calcite crystals as a particular example in his paper, which has later been acknowledged to be one of the milestones in the development of geometrical crystallography (Lima-de-Faria 1990, p. 47).

Hauy (1801) had expressed clearly in his mineralogy a rule to the effect that each crystal species is distinguished from all others by the unique form of its fundamental cells. The regular polyhedra such as the cube were an exception from the rule. However, later findings showed Hauy to have erred here on two counts, and in both cases Iceland spar was involved in these findings.

First, chemists (e.g. Fourcroy and Vauquelin 1804) demonstrated shortly after Hauy's proclamation, that the common mineral aragonite had the same chemical composition as pure calcite, namely CaCO₃. Yet their crystal structures are quite different, and so are their refractive indices, densities and other physical properties. This caused great puzzlement until around 1820 (see a comprehensive review by Arzruni 1893, p. 22-36). Some people (e.g. Stromeyer 1813, 1814) suggested that the differences were caused by the aragonite containing some strontium or water, but that explanation was discounted (Thenard and Biot 1807, see Rose 1864, and Gay-Lussac 1816). The story continues in the next two sections.

5.4 Dimorphous compounds, 1823-40 and later

E. Mitscherlich (1823, see Rose 1864) then observed that substances other than calcium carbonate, even pure elements like sulfur, could crystallize in two ways. The name "dimorphism" was coined to describe this behavior. Calcium carbonate was in fact later found to exist in a third modification called vaterite, but it is quite rare. Mitscherlich (1831) pointed out that aragonite becomes unstable on heating and is converted to calcite. Subsequent research confirmed that this happens at around 400°C.

Various experiments were carried out on dimorphous materials, in order to ascertain the causes of the phenomenon. In the case of calcium carbonate, it was found that the ambient temperature, pressure of CO_2 and other conditions determined whether calcite or aragonite crystallized from a solution, see for instance Rose (1837) and Credner (1870). According to further experimental work, these parameters also decide the form of the crystals in the case of calcite (Kostov 1968, Fig. 5-3 bottom; Kostov and Kostov 1999). Investigators studied thoroughly the influence of other compounds (Lösungsgenossen) in the carbonate solution, for instance Credner (1870) and Vater (1893, 1899) both of whom used Iceland spar in some of their experiments. Hintze (1930, p. 2819) quotes research indicating that the carbonate has to be very pure for the Icelandic form (Spaltungsrhomboeder) to be precipitated. In the 20th century it was established that the size of the metal ion relative to the CO_3 -ion decides to which crystal system the carbonate will belong. When the former is small, the carbonate crystallizes in the same system as calcite; when it is large the carbonate chooses to crystallize like aragonite. The calcium ion is a boundary case, which according to Kostov (1968) also causes calcite to exhibit a greater variety of crystal forms (Fig. 5-3, top) than any other mineral.

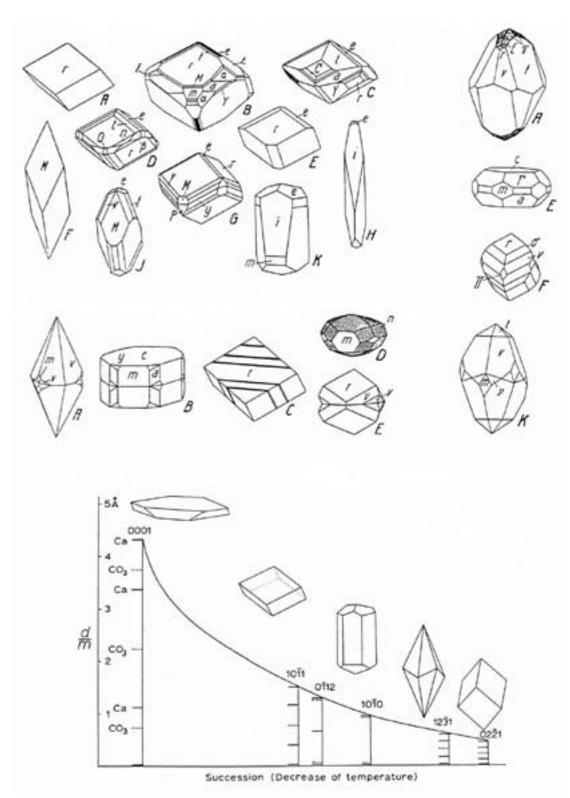


Figure 5-3. Some types of calcite crystals. Top left: Examples of what crystallographers call rhombohedral forms. Irby (1878) stated that 50 different forms of this type had been confirmed to occur in calcite. A is the fundamental cleavage rhombohedron. According to Tschermak (1894) and others, this form was common at Helgustaðir but it was probably rarely seen in calcite specimens from other locations before 1900. G is from Iceland. Right: Examples of so-called scalenohedral forms, from the U.S. and Europe. Irby (1878) states that 155 of these are known. Bottom left: Various types of twin crystals. Twin lamellae as in C can be produced artificially by pressure, see section 21.3. The diagrams are from P. Niggli's Lehrbuch der Mineralogie, 2nd ed. Vol. II, 1926. Bottom: Kostov's (1968) suggestion on how the different crystal forms of calcite reflect lowering of the ambient temperature during its deposition. 1011 is the fundamental rhombohedron, 1231 is a scalenohedron. The quantity on the vertical axis relates to a characteristic distance (grating space) between crystalline layers involved in the respective forms.

5.5 Isomorphous minerals, around 1820 and later

The other side of Hauy's (1801) assertion on a one-to-one correspondence between the unique molecular and crystalline shapes of compounds, also turned out to be debatable. Calcite again was a player in this debate, because the shape of its fundamental rhombohedron was very similar to that of the metal carbonates (Ca,Mg)CO₃ (dolomite), MgCO₃ (magnesite), FeCO₃ (siderite), ZnCO₃ and MnCO₃. At first the crystal angles seemed to be identical, but Wollaston (1812) came to Hauy's rescue by finding with a novel accurate goniometer that the acute angles of Iceland spar, dolomite and magnesite were respectively 74°55', 73°45' and 73°0' (see Partington 1952, p. 114).

Mitscherlich (1818-19, see Rose 1864) then pointed out that in various related or even unrelated compounds, the differences between crystal angles and other properties could be very small; his first examples included certain phosphates and arsenates. A new name had to be found for this phenomenon, and such groups are said to be "isomorphous".

The similarity in shape of the calcite series of metal carbonate crystals gave rise to many kinds of comparative studies, intended to throw light on causes of systematic variations in their physical properties. Thus, Neumann (1831) showed that their specific heats vary inversely with molecular mass, which turned out to be a special case of a general empirical law. Much material on such studies may be found in Arzruni (1893), Tutton (1922, part IV), Linck (1923, chapter IV) and Groth (1926). Related research using X-rays will be described in chapter 36 below. In addition to the above metal carbonates, sodium nitrate (Chile saltpeter, NaNO₃) is isomorphous with calcite. Frankenheim (1836) made interesting observations on the deposition of saltpeter crystals from solution on an Iceland spar substrate, which could be shown to govern the crystal growth under certain conditions. Tschermak (1881) carried out a thorough comparison between properties of saltpeter, Iceland spar and the other isomorphous carbonates. The saltpeter will be returned to later, for instance in section 33.3.

6 An epoch of great progress in optics, 1800-15

6.1 Events in optics etc. in Britain 1800-10

Around 1800, important discoveries were made in England, in fields relevant to the history of Iceland spar. In particular, T. Young (1802) had investigated the behavior of light incident on opaque screens with narrow slits. His results strongly indicated that light had to be a wave motion in an aether substance. They were also supported by the colors of thin plates. Young (1802) referred to Huygens' observations on Iceland spar but did not himself study double refraction. His conclusions did not please a certain H. Brougham, who had previously published two papers on optics (Brougham 1796, 1797). In the latter of these, Brougham had repeated some of the experiments of Martin (1774) on Icelandic crystals. He trusted Newton's ideas of light, and published polemical reviews (Brougham 1803a,b) discounting Young's work. Brougham's arguments do not appear convincing today, but he may have delayed for a while the general acceptance of the wave theory. All of Brougham writings on a wide variety of subjects had in fact "but little permanent value" according to Chambers's Biographical Dictionary of 1938.

In his paper on the wave nature of light, Young (1802) pointed out recent experimental results by W. Herschel which suggested that the heat radiation from the Sun had in many respects the same character as visible light. Young had then found out that wavelengths of the visible spectrum ranged from 425 nm in the extreme violet to extreme red at 675 nm, so that the heat rays had a longer wavelength than red light. Indications that the total spectrum of Sun's radiation also reached beyond violet were first noted on the Continent by J.W. Ritter (1803), and soon confirmed by Young.

Wollaston (1802a) constructed a new instrument for making accurate measurements on the refractive indices of solids and liquids for light of different colors. He also made (Wollaston 1802b, see Fig. 6-1) measurements on the refraction of the extraordinary ray in Iceland spar at various angles of incidence. The measurements agreed, at least to the second decimal place, with calculations by Hauy based on Huygens' (1890) suggestion of elliptical wavefronts. This could be considered, in Wollaston's words, "to be highly favourable to the Huygenian theory". In a review of Wollaston's paper, Brougham (1803b) praised his experimental work but was obliged to lambast the "untenable and useless hypothesis" of light being a wave.

Wollaston (1809) then described a novel instrument for measuring angles between the faces of crystals, a reflection goniometer (top of Fig. 6-1). Here the crystal was placed at the center of a rotatable circular platform, and a horizontal light ray was reflected from its sides in turn. In the first material that Wollaston measured, namely Iceland spar, he confirmed his own previous result that the angle between the symmetry axis and any of the three sides of the crystal is slightly greater than the 45° assumed by Huygens. The new instrument was more precise than that of Carangeot mentioned above, and it could also measure small or broken crystals. Its accuracy was improved further by the addition of a telescope with crosshairs (by E. Malus, probably 1809-10) and later by employing a collimator to produce parallel light, see Ann. Phys. 2, 83-89, 1824.

Wollaston also was the first to describe (in an article on the element palladium, in 1804) crystals whose colors depended on the direction from which they were viewed. These were named "dichroic" minerals, the best known being tournaline. Its properties were described in more detail by T.J. Seebeck (1813), Biot (1815d) and Brewster (1820). It turned out that such crystals were doubly refracting but absorbed one of the two rays much more strongly than the other one. Throughout the 19th century, thin plates of tournaline were a useful tool for studying double refraction in other materials, and related optical phenomena. However, finding crystals of tournaline suitable for making such plates could be difficult (see Herschel 1820, p. 97 and Groth 1885, p. 149). It is not always clear from the text of publications early in the century whether Iceland spar, tournaline plates, or other techniques were being used in various experiments within this field of optics.

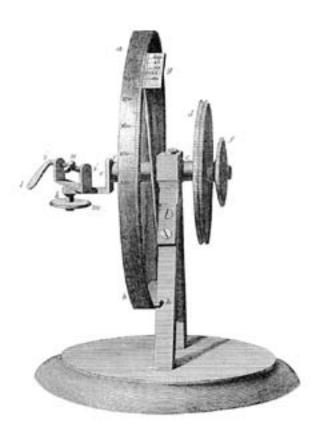
6.2 Great times for science coming up in France around 1800

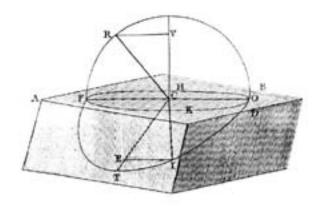
In France, large strides were made in the natural sciences and engineering around and soon after the turn of the century. This progress was in part connected with the country's famous Academy of Sciences (named Institut de France from 1795). In a few decades, a remarkable number of important discoveries were made in for instance mathematical analysis, mechanics, thermodynamics, electromagnetism, the properties of solids and fluids, geodesy, and engineering design (see Timoshenko 1953, Grattan-Guinness 1990, and other historical accounts). Including the development of optics to be described below, it may be said that in the years 1795-1825 physics was becoming solidly established as an independent field of the sciences. The rapidity of this progress is even more remarkable because the careers of scientists were often adversely affected by the political turmoil and wars following the Revolution of 1789; some also suffered poor health.

Many of the foremost natural scientists of France were either teachers or past students at the engineering school École Polytechnique, established in 1794. One of the founders of that school, G. Monge (1789?), wrote an article describing a particular effect of the double refraction in Iceland spar, but he is best known for his researches in mathematics. In addition to Hauy who has already been mentioned, some others in France and Germany were writing about double refraction and related fields of optics around 1800, for instance Link (1797) but I have not acquired copies of their publications.

6.3 News of Wollaston reaches France: consequences

Communications between England and France at the time of the Napoleonic wars (1799-1815) must have been rather sporadic, and some historians have claimed that French scientists did not learn about Wollaston's (1802b) research on the double refraction of Iceland spar until 1807 (see Gillispie 1997, p. 211-212, and the entry on Laplace in Dictionary of Scientific Biography 1980-90). Buchwald (1980) however says that both of Wollaston's 1802 papers were published in France in 1803. In 1805-06 P.S. de Laplace, J.B. Biot and F. Arago were investigating matters connected with the refraction of light, and Buchwald expects that Laplace wanted to test whether Newton's corpuscular theory of light could explain double refraction. Laplace himself had much faith in that theory, and used it in the interpretation of experiments. He was very influential in





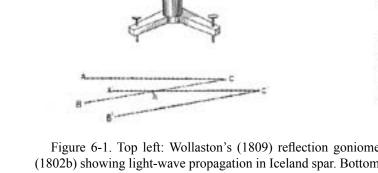
Sur une propriété de la lumière réfléchie

PAR M. MALUS.

Lonsqu'un rayon solaire est réfléchi ouréfracté, il conserve en général ses propriétés physiques; et, soumis à de nouvelles épreuves, il se comporte comme s'il émanoit directement du corps lumineux : le prisme, en dispersant les rayons colorés, ne fait que changer leur direction respective, sans altérer leur nature. Il y a cependant des circoustances où l'influence de certains corps imprime aux rayons qu'ils réfléchissent, ou qu'ils réfractent, des caractères et des propriétés qu'ils transportent avec eux, et qui les distinguent essentiellement de la lumière directe.

La propriété de la lumière que je vais décrire est une modification de ce genre. Elle avoit déja été apperçue dans une circonstance particulière de la duplication des images, offerte par le spath calcaire; mais le phénomène qui en résultoit étant attribué aux propriétés de ce cristal,

Figure 6-1. Top left: Wollaston's (1809) reflection goniometer for crystals. Right: A diagram from Wollaston (1802b) showing light-wave propagation in Iceland spar. Bottom left: Drawing (Wood 1905) of Malus' 1808 method for measuring precisely the refraction of light in natural and cut pieces of Iceland spar. The lower part shows what was seen in the telescope with the crystal in place. Right: The initial paragraphs of the paper by Malus (1809) on his fundamental discovery regarding the polarization of light by reflection. He uses the term "spath calcaire" here, but "spath d'Islande" later in the paper.



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the scientific institutions of France, having been for decades one of the nation's most prolific and famous researchers in fields including mathematics, astronomical sciences, and theoretical physics. Probably at his initiative, the Academy announced in December 1807 a prize competition on double refraction, calling both for a theory to explain it and an experimental test of that theory. The competition would close at the end of 1809.

6.4 On the role of Iceland spar around 1800, and on beam-splitting prisms 1777-1820

To 1820 or later, Iceland spar rhombohedrons were probably the main tool for studying double refraction in other materials, because of their large size, transparency and regular shape in addition to the fact that the difference between the two refractive indices of Iceland spar is unusually large. Observers then watched rays passing through the materials, checking whether their intensities changed on rotation (cf. Fig. 3-2).

The two rays coming through the Iceland spar are parallel and not far apart (just over 2 mm for 2 cm thickness, see Fig. 8-3), so that the images of objects overlap. At some point in time it was realized that such observations would be easier if the rays were separated in direction. This could be effected by a wedge-shaped prism. In order to preserve the colors of the images (i.e. reduce dispersion), two wedges of spar with different optical axes could be cemented together into a rectangular shape as in Fig. 12-1. The earliest available description of such a prism is by A. Rochon (1777, see also Torelli de Narci 1803 and references e.g. in Partington 1953).

Rochon's original prisms however, were made of calcite or quartz and glass, and for a quite different purpose. He intended to use the constant small angular distance between the two images of a small or distant light source, as a standard in measurements of small angles. Rochon (1811, 1812) also mounted this sort of prisms in telescopes, intended as range-finding devices in for instance naval warfare. They would be simpler to use than glass prisms invented by L. Euler in 1783. More on this in section 12.1.

6.5 A big leap forward in optics: the discovery by Malus in 1808

A young engineer, E.L. Malus (Fig. 5-2) who had carried out research in optics (Malus 1807a,b) that was of interest to Laplace, took part in the competition mentioned above. He designed a new and very accurate instrument for measuring the refraction in natural or cut pieces of Iceland spar. It employed a circular goniometer invented earlier by J.C. Borda, and two scales engraved in a metal plate, meeting at an angle of arctan(0.1) (Fig. 6-1). Buchwald (1989, p. 31-36) describes Malus' procedures, which were exemplary. First, he bypassed certain problems with internal reflections that had affected Wollaston's results; second, his measurements and the presentation of results including error estimates, were executed with thoroughness and clarity; third, processing of the results and comparisons with Huygens' hypothesis on the extraordinary wave surface, were carried out by computations which was a more complicated but also a more exact method than the previous one of drawing geometrical constructions.

Malus confirmed the theoretical predictions of Huygens within 1%, but he did not himself favor the undulation hypothesis (see e.g. Born and Wolf 1999, p. xxvii) and neither did Wollaston (see Gillispie 1997, p. 212). Malus (1809) also tested Huygens' theory in a less comprehensive way on some other crystallized compounds like lead sulfate, quartz and sulfur.

A big leap forward in the understanding of the nature of light occurred by a chance event dur-

ing Malus' researches. According to a biographical account by F. Arago, Malus was one evening inspecting a doubly refracting crystal (most probably Iceland spar) in his rooms at the Rue d'Enfer near the Luxembourg Palace in Paris. He happened to look at the reflection of the setting sun from windows of the palace, and expected to see two equally bright images. They were however different, their intensities varying as he rotated the crystal. He subsequently made the same observation with light reflected from other smooth non-metallic surfaces, and he also noted that the relative intensity of the reflected images varied with the angle of incidence. It is inevitable that many people had looked at reflected light through Iceland spar before Malus, but he was the one to grasp the fundamental significance of his observations. In brief, he had found that *the change in the character of light which occurs on its passage through a doubly refracting crystal (as noted by Huygens), also occurs to some extent when it is reflected and transmitted at interfaces of ordinary materials.*

Malus made further experiments on the refraction of light in Iceland spar and on its reflection, deducing among other things certain "cosine laws" (named after him) relating to both processes. The light that had been modified by these processes was by him said to be "polarized", this property having a direction perpendicular to the direction of propagation. The expression has persisted, although it refers to Newton's tentative suggestion that his hypothetical light particles might have two poles like magnets. Malus informed the Academy of his findings, and they were deemed important enough to be published with haste, i.e. long before the competition deadline. Another essay (Kramp 1811) which was submitted to the Academy, received honorable mention. However, Kramp concentrated on the crystallography of Iceland spar and his results were of much less general importance than those of Malus.

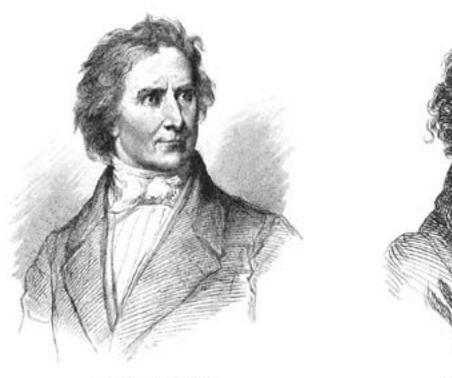
6.6 Publication of Malus' results, around 1810

Daumas (1987, p. 81) says about the results obtained by Malus, that "a new branch of optics was born", which was no exaggeration. A new and sensitive property of light had now been added to those (like its intensity, color, or angle of incidence on a surface) that could be measured and changed in order to study the propagation of light and its interactions with matter. Laplace (1808a,b, 1809a,b) published a description of Malus' results and explained them, in fact ahead of Malus (1808a,b, 1809, 1810a,b, and others) himself. The prize essay of Malus (1811) is an extended compilation of his results on double refraction and polarization. He died in February of 1812.

Laplace thought that Newton's theory of light was able to explain the behavior of the extraordinary ray in crystals like Iceland spar, by assuming that forces from the material particles of the crystal upon the light corpuscles varied in a certain way depending on their direction of motion. Here he applied the principle of least action, due to P.-L. Maupertuis in 1744.

Young (1809, 1810) agreed that the research by Malus on the refraction of light in Iceland spar and on its polarization was "the most important and interesting that has been made in France, concerning the properties of light, at least since the time of Huygens". He however criticized Laplace's attempts to keep the unnecessary corpuscular theory alive. Young made the valuable point that the elastic modulus and hence the speed of sound in certain substances such as wood vary with direction. The directional dependence of the extraordinary index of refraction in crystals might perhaps have a similar cause.

During the next decade, Young and other supporters had no explanation of the polarization property of light, as the wave motion in light was expected to be longitudinal (i.e. back and forth in the direction of propagation) like in sound. Furthermore, they could neither explain the disper-



FRANÇOIS ARAGO

JEAN-BAPTISTE BIOT



Figure 6-2. Four notable scientists who studied Iceland spar and polarized light. Top left: F. Arago (1786-1853). Right: J.B. Biot (1774-1862). Bottom left: D. Brewster (1781-1868), taken from Morrison-Low and Christie (1984). Right: A. Fresnel (1788-1827).

sion of light in materials, nor the chemical effects of light (such as the darkening of silver halides, which later became the basis of photography).

6.7 On some of the wide-ranging consequences of Malus' discovery, after 1810

During the next years and decades, scientists studied the new aspect of the nature of light, and made one major discovery after another. Many books have been written about this revolution in optics and the lives of its chief participants. To quote again from Daumas (1987): "The phenomena of polarization were the center of preoccupation among the physicists. Malus had made optics fashionable". The French were the leaders in the field until at least 1826. In most of this research, Iceland spar crystals were an indispensable tool. This section and the next deal mostly with writings by F. Arago and J.B. Biot (Fig. 6-2); among others studying polarized light soon after 1810 were A.M. Ampère (1813-15) and J.E. Bérard, see section 10.5.

Arago (1811) by chance observed the light from the clear sky through a thin plate of mica and an Iceland spar rhomb. The sky light is partially polarized, as will be discussed for instance in section 27.1. This produced beautiful colors in the plate, similar to those seen in oil films on water in ordinary light. Arago made use of the phenomenon by constructing a device to inspect qualitatively whether a particular source of light was polarized. This "polariscope" was a telescope containing a thin piece of mica or clear selenite (gypsum) and a spar rhomb that could be rotated around the line of sight. An observer saw in the telescope two spots of light with different colors dependent on the state of polarization of the light (Verdet-Exner 1887, section 256). In another instrument illustrated in his "Astronomie Populaire" and in Daumas (1987) he found for instance that the polarization of reflected light varies between the different regions of the Moon.

Biot (1811, 1812, and later) made a very extensive series of experiments on the colors of thin crystalline plates in polarized light. He advanced an explanation of this "polarisation chromatique" by allowing Newton's luminous particles to rotate or oscillate in a certain fashion inside the materials. Young (1814) soon pointed out the correct cause of the colors in terms of the wave hypothesis, but his explanation took a while to be accepted by Biot or others (Kipnis 1991). The colors were due to the combination (known as interference) of the two waves of light passing through the crystalline plates. As the difference in the two velocities was changing progressively with the frequency of the light, the waves emerged from such a plate in the same phase for some frequencies and out of phase for others. Many scientists wrote subsequently about aspects of this chromatic polarization, including Mayer (1812, 1815) who however did not produce significant new results. The phenomenon attained practical importance when applied in polarizing microscopes for mineralogical and petrographic studies after 1860. See for instance Billet (1858-59, Chapter X), Bertin (1879) and sections 18.2 and 22.4 below.

Biot wrote many papers on the various interactions between light and matter in a period of over 40 years, frequently mentioning Iceland spar (e.g. Biot 1816a). One of these (Biot 1815b) treats in particular the variation in its refractive indices with wavelength of the light, i.e. its dispersion. Biot (1814) also found that in some minerals including quartz the index of the ordinary ray (n_o) was less than that of the extraordinary ray (n_o) . This was opposite to the situation in Iceland spar and many other crystals, and he realized that in the quartz group the ellipsoid of Huygens would be elongated (prolate), rather than flattened (oblate) as in Fig. 6-1. According to the corpuscular theory however, one might assume that the light particles were attracted to the symmetry axis of the crystal (its optical axis, called axe de réfraction by Malus) in the quartz group and repelled from it in the calcite group. Since then, the former minerals have often been called positive uniaxial, and the others are called negative uniaxial ones.

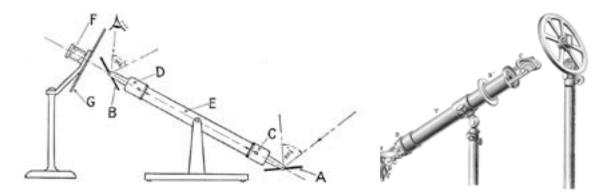


Figure 6-3. Two illustrations of the polariscope of J.B. Biot. The left-hand one is from Cheshire (1923), based on a description in Biot's 1816 textbook on Physics. A is a glass-plate polarizer. One can either look from above at the glass mirror B (eye) or remove this mirror and look from the left through the Iceland spar rhomb F which may be rotated about the axis through the instrument. G is a graduated circle seen edge-on. The right-hand illustration (Mascart 1891) is based on Biot's papers from 1812-14. M is the glass-plate polarizer. Crystal plates could be placed on the platform C for inspection; the observer looks through the Iceland spar rhomb attached to the graduated circle on the right.

In the experiments of Biot (e.g. 1812, 1818 and later) a "polarizer" first produced pure polarized light from ordinary light. The state of polarization of the light was then observed in an "analyzer" after having for instance passed through a material specimen. Both could be rotated about the direction of the ray path, and adjusted so that the analyzer blocked all the light passed by the polarizer. As a polarizer, Biot used a reflecting glass plate tilted at an optimum angle (see sections 7.3 and 8.1) determined initially by the experiments of Malus. His analyzer was another glass plate, a rhombohedron of Iceland spar, or a Rochon prism also of Iceland spar (see Mascart 1891 and Cheshire 1923, Fig. 6-3, also Bates et al. 1942, and Appelquist 1987). This type of polariscope later developed into the most common form of instruments called polarimeters, see section 13.2 and others below. I have not found much information on the contributions of others to stages in that development before 1840, except for one German description of such an instrument (Schulze-Montanus 1817).

6.8 Optical activity, 1811-15

Another phenomenon first noticed by Arago (1811) and studied in detail by Biot (1812, 1817 and many other papers) was a peculiar action of quartz plates upon polarized light, in addition to the double refraction exhibited by that mineral. If a plate had been cut in a direction at right angles to the optical axis of a quartz crystal, the characteristic direction of polarization in a light ray *ro-tated* progressively on its way straight through the plate. Some crystals of quartz rotated light to the right, others to the right by an equal amount. This was a puzzle for decades, see section 16.2 on L. Pasteur. The rotation amounts to 21.7° per mm of thickness for yellow sodium light, but it increases rapidly from red to violet. This property was called by Biot "polarisation circulaire" or "pouvoir rotatoire"; in English the terms rotatory power or optical activity are used.

Other instances of optically active crystals were not discovered until around 1855, see section 18.2. According to Biot's ideas, the corpuscles of light were supposed to rotate about the optical axis of quartz on their way. Soon however, yet another unexpected phenomenon demonstrated that the cause of the rotation of light in a material need not be connected to a crystalline structure. Biot (1815a, 1817 and later) namely announced that optical activity was also present in organic liquids such as essential oils from plants and aqueous sugar solutions. This important story will be resumed in section 12.4.

7 Continuing progress, 1816-20

7.1 The wave theory gains ground: Fresnel, transverse waves etc. from 1816

In the spring of 1817, the French Academy of Sciences again announced a prize competition in optics. This time the object was to explain the bending of light around obstacles, called diffraction. The members of the Academy involved were hoping that this would help in getting the corpuscular theory firmly established. The competition was the occasion for the first scientific publications of another young engineer, A. Fresnel (1816 and later, see also Fresnel 1822b). Fresnel (Fig. 6-2) had been taught in the École Polytechnique that light was a stream of particles, but his own observations convinced him of the superiority of the wave hypothesis. He used it to explain thoroughly the phenomenon of diffraction, and won the competition in 1819. His prize essay did not appear in print until 1826, but his analysis had along with other evidence converted most scientists to the wave hypothesis by 1830 (Silliman 1975, p. 157; Buchwald 1989, p. 291-297).

Fresnel went on to make other major contributions in the field of optics. Arago had also come to believe in the wave theory of light, and together they investigated in 1815-19 the interference of polarized light. Among other things they demonstrated that rays polarized "en sens contraire", i.e. at right angles, only gave rise to an interference pattern under certain conditions (Arago and Fresnel 1819, see Wild 1856 and Verdet 1869). T. Young heard of their results, and pointed out in letters to Arago in 1817-18 that the movement in the light wave had to be transverse to its direction of propagation. This was particularly important in the continuing development of optical theories, and soon also in ideas about heat radiation (see Silliman 1975). Lloyd (1857, p. 136) and Verdet (1869, p. 444-445) are of the opinion that Fresnel admitted the possibility of transverse waves before Young who might even have thought that the transverse component of the wave was small. It should also be noted that R. Hooke had suggested that light waves were transverse, in a 1672 publication.

7.2 Polarization of light - explanations

The admission of transverse vibrations in light calls for explanations and definitions. Let the z-axis of a rectangular system of coordinates be aligned with the direction of propagation of a monochromatic light ray. Then the actual vibration of the "light vector" (whatever its nature) may be considered to consist of two components: one is a sinusoidal wave in a quantity pointing the x-direction and the other a sinusoidal wave of a quantity pointing in the y-direction. See Fig. 7-1.

If the amplitudes of these component waves are X and Y respectively and they are completely

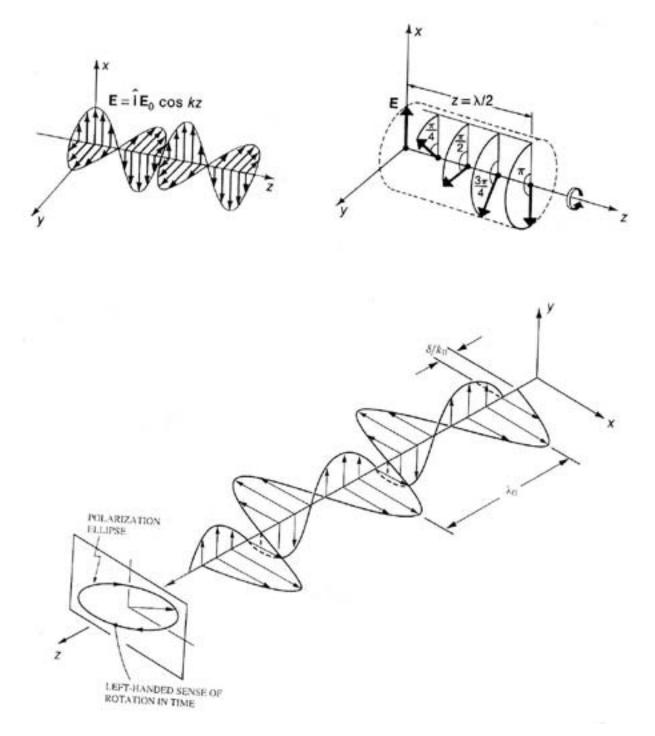


Figure 7-1. Three different states of polarization of light, partly from the viewpoint of electromagnetic theory. Top left: Two similar waves (i.e. light rays) travelling along the z-axis. The electric field of one of these is in the x-direction, that of the other is in the y-direction. Their combination is a single linearly polarized wave with its field inclined 45° from both the x- and y- directions. Right: By passage through a mica sheet of appropriate thickness, one wave has been delayed ¼ of an oscillation relative to the other. They now combine to form a circularly polarized wave. Bottom: A general case, where the waves are of unequal amplitudes and have an arbitrary phase difference less than ¼ of oscillation. They now form an elliptically polarized wave. Simplified from J.B. Marion and W.F. Hornyak: Physics, 1982.

in step, then their resultant will always lie in a plane that makes an angle $\arctan(Y/X)$ with the xz-plane. This is said to be a case of "linear" or "plane" polarization, and so is the case when the x- and y-waves are half a vibration out of step. If the components are of equal amplitude and one is a quarter of a vibration ahead of the other, the end of the wave vector will appear (from an observer looking along the z-axis) to go around in a circle. In fact it performs a helical motion, because the wave is advancing. This is called circular polarization, right- or left-handed depending on conditions and conventions. If the difference in time between the waves is some other arbitrary fraction of a whole cycle, we speak like Fresnel of elliptical polarization because the projection of the wave vector on the xy-plane will describe an ellipse. Instead of referring to a time difference, one generally refers to the difference in phase between the two components: 0° or 180° for linear polarization, 90° or 270° for circular polarization, and so on. Ordinary light travelling in the z-direction, for instance from the Sun or from a lamp, is composed of a multitude of such waves with random directions of vibrations and random phase differences from each other (in addition to being of course of many frequencies). It is therefore said to be unpolarized.

7.3 Brewster: Brewster's angle, biaxial crystals and more, 1810-20

We now move to Scotland where D. Brewster (Fig. 6-2) worked independently on a wide range of experiments in optics, crystallography, mineralogy and related fields. The list of his publications includes over 1200 titles in 1800-69 (Morrison-Low and Christie 1984), from small newspaper items to books. He was also active in societies and as a journal editor. Brewster seems never to have taken a definite stand in the particle vs. wave debate on light, but his research greatly advanced optical science.

One of his results (Brewster 1815a) from measurements made around 1811 on many materials, was this: when light with "parallel polarization" falls from air on to the smooth surface of a solid or a liquid, none of it is reflected if the angle of incidence (φ in Fig. 7-2, top left) is equal to arctan(n) where n is the index of refraction of the material. This "Brewster's angle" is for instance 56.3° for glass with an index of 1.5. More complex laws apply to doubly refracting crystals, as will be mentioned later.

W.H. Wollaston told Brewster in 1814 that if an Iceland spar plate cut at right angles to the optical axis, was illuminated with convergent light, concentric colored rings with a dark cross could be seen. Brewster, who had himself noted this in a few precious stones (Brewster 1813a, see also Brewster 1860), then carried out similar observations on many minerals (Fig. 7-2 and 7-3). He was thus able to confirm (Brewster 1818b, 1819-20) and extend a result that J.J. Bernhardi had previously obtained from more limited evidence, namely that certain symmetry aspects of the geometry of crystals also appear in their optical properties. Minerals forming cubical-shaped crystals do not exhibit double refraction, crystals from some of the other systems (see section 9.1) have one optical axis along which the speeds of the ordinary and extraordinary waves are equal, and still other crystals have two such optical axes. This major discovery later became, like the thin-plate colors of Arago and Biot, an important tool in crystallographic and mineralogical research.

One more discovery by Brewster (1815c, 1816b) concerned the fact that objects from glass, wax, jelly or fluorspar which normally are not doubly refracting, acquire that property in they are subjected to non-hydrostatic pressure. This phenomenon was for a long while called "accidental" or "artificial" double refraction, later "photo-elasticity". Both Brewster (1814, 1816a), T.J. Seebeck (1813, 1814) and Biot (1815c) studied also, with glass mirrors or Iceland spar, double refraction, in rapidly cooled glass which has large internal stresses. This attracted considerable attention, see Biot (1816b), and Brewster and Seebeck were awarded a prize by the French Academy for

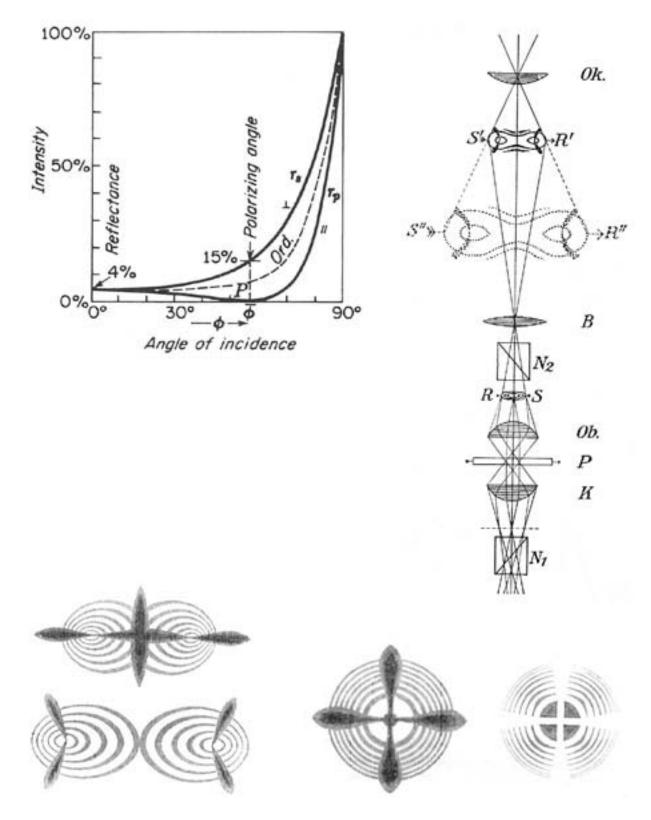


Figure 7-2. On two of the many discoveries of D. Brewster. Top left: Graph of the relative intensity of light reflected from a glass surface, for different angles of incidence. The upper and lower curves apply to light polarized respectively at right angles to the plane of incidence and parallel to it. The center curve is for unpolarized light. At the Brewster angle arctan(n) which is 56-60° for common types of glass, none of the parallel-polarized light is reflected. From Jenkins and White (1957). Right: Equipment for viewing a thin plate P of a crystal in convergent polarized light. The Nicol prisms N correspond to P and A in the next Figure. From Weinschenk (1925). Bottom: Patterns seen in the above equipment; those on the right appear (under different conditions) when the crystal has one optical axis, those on the right appear when it has two such axes. From Lloyd (1857).

their discovery. See chapter 11 and section 15.3 on further work in this field. It may be mentioned here that J.W. Goethe was very interested in the science of light and colors, and he had published a large work on the subject in 1810, "Zur Farbenlehre" which was influential for a long time (Dove 1853). Malus and others criticized Goethe's theories, as he for instance considered white light not to be composed of other colors. This led to correspondence between Seebeck and Goethe about Seebeck's research and previous discoveries by Malus, Biot and Arago regarding polarized light. In late 1812, Seebeck sent a prism of Iceland spar to Goethe. In a follow-up volume by Goethe, "Ergänzungen zur Farbenlehre" published in 1820, there are sections on Iceland spar, colors of thin crystalline plates, and related matters.

Brewster (1816c) demonstrated that the crystalline lens in the eyes of animals is doubly refracting, and he investigated various topics regarding the optics of eyes and vision during the following decades. Those investigations, according to Morrison-Low and Christie (1984, p. 101-103) "may be truly said to lay the foundations for modern biophysics".

Brewster also made some studies on Iceland spar (e.g. Brewster 1815b) and used natural samples or prisms from it in much of his research. He usually calls it "calcareous spar" but also "Iceland spar" (at least in Brewster 1813b, 1814, 1815b). In one of his papers (Brewster 1818a) he had recently acquired "some very perfect specimens of calcareous spar". He rarely states where these came from (see Appendix 4); however, once in an 1819 article he mentions the Faeroes, and Ireland in another instance.

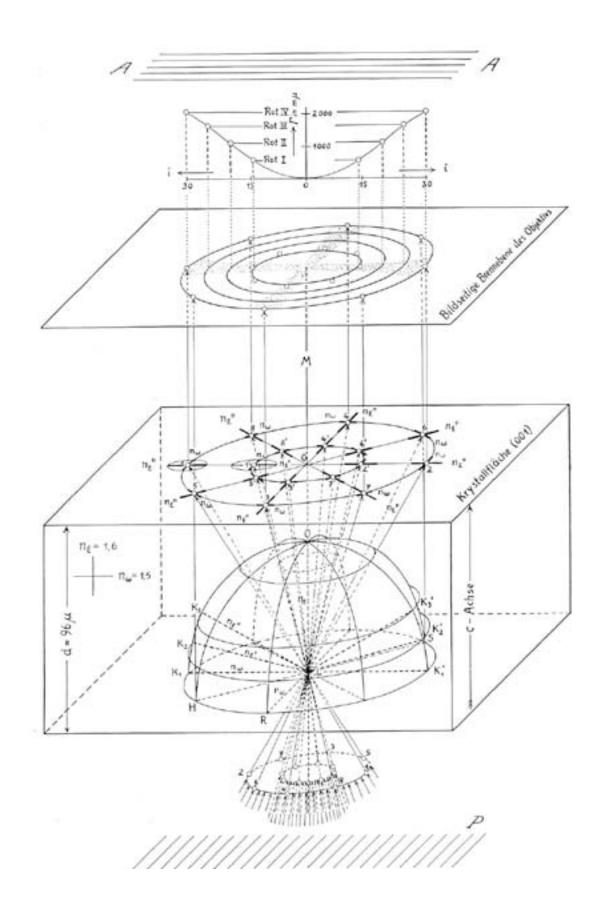


Figure 7-3. A diagram explaining how the circles and the shadow-cross in the previous Figure originate. A conical beam of light enters from below through the polarizer P and a plate of uniaxial crystal. The circles are formed by interference of the ordinary and extraordinary rays. The cross appears where the net light vibration is in a direction not transmitted by the analyzer A. From Houben-Weyl (1955).

8 Further major contributions by Fresnel, 1817-1827

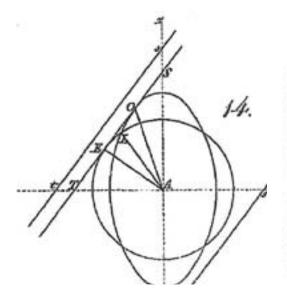
In the last decade of his short life, A. Fresnel was very productive in research and writing. Lloyd (1857, p. 136) says that Fresnel in his research on light as a transverse wave "…reared…the noblest fabric which has ever adorned the domain of physical science, - Newton's system of the universe alone excepted." (In a similar statement referred to by D. Brewster in the Report of the British Association of 1832, the names of several other scientists are also mentioned). At the beginning of a 150-p. translation of Fresnel's (1822b) review chapter on light, the editor of Annalen der Physik anticipates that his research will have great impact in various fields of physics. He also considers that optics has hiterto been given far too little attention in Germany. As the publication history of Fresnel's papers, and their interrelationships, are unusually complicated, I shall merely present some of his most important contributions connected to polarized light. More detailed accounts may be found e.g. in Buchwald (1989) or Grattan-Guinness (1990).

F. Arago supported Fresnel's work in various ways and publicized it, for instance in a major article in the Encyclopaedia Britannica in 1824 (Arago 1858a). Some of Arago's own contributions to the study of polarized light and photometry are mentioned elsewhere in this account; however, many of his writings were only published in the posthumous collection of his complete works (Arago 1858b,c) except for short abstracts (e.g. Arago 1850).

8.1 Theoretical and experimental work by Fresnel on light in ordinary materials

Fresnel (e.g. 1817) embarked on a study of the question how a light wave is transmitted through ordinary (uncrystallized) materials like glass. By his incredible insight, he was able to predict various aspects of the behavior of the light in such environments. For instance he obtained precise equations for the relative amount of light that is reflected when incident on a surface of a solid or a liquid, as a function of its refractive index and the angle of incidence. It turned out that this amount is also to a considerable extent dependent on whether the light is polarized in the plane of incidence or at right angles to it. Brewster's experimental result (section 7.3) that no light in the former state of polarization is reflected at the angle arctan(n), corresponds to a simple special case of Fresnel's equations. Within a range of angles of incidence in the vicinity of Brewster's angle, very little of that component is reflected (Fig. 7-2). This was the circumstance that had led to Malus' discovery of polarization by reflection in 1808.

No accurate equipment for measuring light intensity was available at this time, so that Fresnel's reflection formulas were not tested directly until 1874 (according to Winkelmann 1906, p. 1249). However, his general results were confirmed in many other ways, and the formulas are still an essential item in University textbooks on optics. Fresnel himself carried out ingenious experi-



DE LA DOUBLE RÉFRACTION ET DE LA POLARISATION

57. — Lorsqu'on fait tomber un faisceau lumineux sur une des faces naturelles d'un rhomboïde de spath calcaire, il se divise dans son intérieur en deux autres faisceaux, qui suivent des routes différentes, et présentent ainsi deux images des objets vus au travers du rhomboïde. On a donné le nom de double réfraction à ce phénomène, ainsi qu'à tous ceux du même genre que produisent beaucoup d'autres cristaux, quand on les taille en prismes pour rendre plus sensible la séparation des deux images.

58. → Mais cette bifurcation de la lumière n'est pas le seul fait remarquable qu'offre la double réfraction : chacun des faisceaux dans lesquels se divisent les rayons incidents jouit de propriétés singulières, qui établissent des différences entre ses côtés. Pour décrire avec précision les phénomènes qu'elles présentent, il est nécessaire d'employer et de faire connaître les expressions usitées.

Dans les cristaux où les lois de la double réfraction sont

Figure 8-1. Left: One of the best-known illustrations from Fresnel (1827), dealing with the propagation of a light wave in a biaxial crystal. The intersection of the two elliptical surfaces causes a peculiar phenomenon known as conical refraction. Right: In this text from a re-publication of Fresnel's (1822a) review chapter on light, it is seen that (like many others) he used calcite as the type example of uniaxial crystals.

ments related to his theoretical derivations, using among other things "un rhomboide de spath d'Islande". A trapezoidal glass prism named after him (Fresnel 1822a), which converts linearly polarized (or: plane-polarized) light into circularly polarized light or the other way around, is illustrated in Fig. 8-2. These experiments furnished convincing support for the wave theory of light (cf. for instance Biot 1817, p. 133-136).

Fresnel did not publish his fundamental assumptions or the derivations of his formulas on the propagation of light. He must have realized that the aether concept was in certain respects not realistic, and his computations were also very complicated. Fresnel's essay on these matters only appeared in the collection of his complete works (Fresnel 1866-70). The speed of a sound wave in an elastic material is generally equal to the square root of a modulus of elasticity divided by a density. In Fresnel's theory the elastic modulus of the aether in different materials was the same, while its density varied between them. This led to the conclusion that the transverse motion of the aether was at right angles to what Malus and others had called the "plane of polarization" of a linearly polarized light ray.

8.2 Fresnel's hypothesis on double refraction in crystallized materials

Fresnel (1821) studied the reasons for the colors and figures observed in thin plates of mica, gypsum and other minerals in polarized light, explaining them on the basis of Huygens' theory. Thanks to Arago, efforts by Biot (who still adhered to Newton's ideas) to stop the Academy from publishing Fresnel's paper were unsuccessful. Iceland spar was much involved in these studies, as recalled by Billet (1862): "...the celebrated experiment of crossed Iceland spar prisms by Fresnel, designed to counter objections raised against his theory of colors in crystalline plates...".

Fresnel (e.g. 1821-22, 1822b, 1827) also investigated the wave motion of the aether in crystals, both uniaxial (such as Iceland spar, Fig. 8-3) and biaxial. Instead of considering waves emanating from a single point, he studied the propagation of plane waves passing in all directions through a point. He defined new expressions that were also used by subsequent researchers, among them

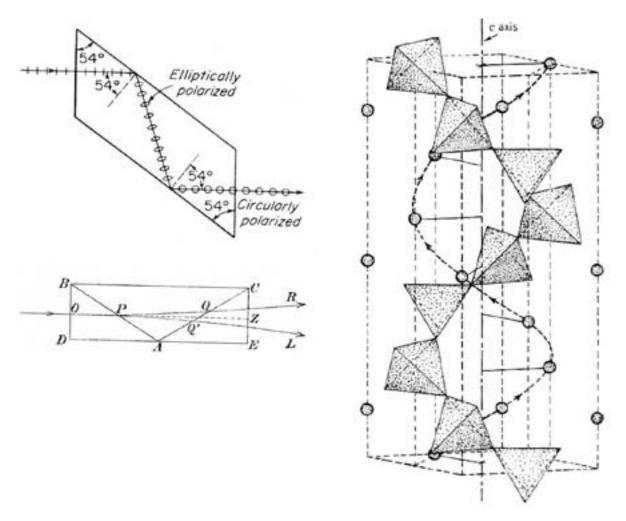


Figure 8-2. Top left: Fresnel's glass prism, which converts linearly polarized light into circularly polarized light by two total internal reflections. From Jenkins and White (1957). Bottom left: Fresnel's composite prism made from right-handed and left-handed quartz. A linearly polarized light beam enters from the left and is split into two circularly polarized beams, one rotating to the left and the other to the right. This agreed with Fresnel's theoretical prediction. Right: The spiral structure of quartz, suggested by Fresnel in order to explain its optical activity and confirmed by X-ray studies more than a century later. From Wahlstrom (1969).

"ellipsoid of elasticity" and "Fresnel's wave surface". From his assumptions he deduced that in a uniaxial crystal the luminous oscillations would indeed be propagated as two waves, agreeing precisely with Huygens' (1690) construction for the ordinary and extraordinary rays. In the latter, the aether oscillations would lie in the plane determined by the direction of propagation and the optical axis. In the ordinary wave, the oscillations were at right angles to these and also at right angles to its own direction of propagation (Verdet 1869; Wood 1977, p. 81-82).

Here we can introduce the concept of a "principal section" in a uniaxial crystal (Fig. 8-3); Huygens (1690) had already appreciated its importance. In calcite there are three principal sections, each of which is a plane containing the optical axis and a normal to a cleavage plane. Such a plane cuts the crystal rhombohedron in a parallelogram having angles of 71°/109°. Fig. 8-3 shows a light ray falling on a surface of Iceland spar: a natural specimen is on the left. The area HBDF emphasized in bold, is one of the principal sections and it is perpendicular to the surfaces EFGH and ABCD. Imagine light coming through the side EFGH at an angle of incidence i. The righthand diagram shows how the ordinary ray o (polarized at right angles to the section) is refracted according to Snell's law. The extraordinary ray e is refracted in a different direction, away from

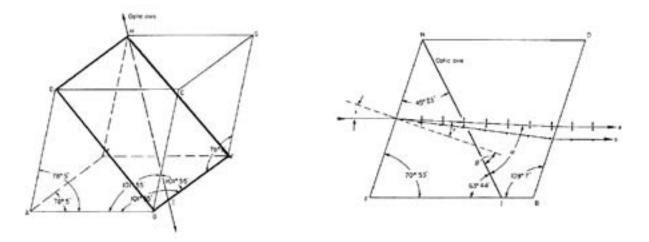


Figure 8-3. On the left is an Iceland spar rhomb, with one of its principal planes outlined in bold. On the right a light ray falls on the back side of the rhomb, the angle of incidence being i in that principal plane. In the extraordinary (e-) ray, the direction of polarization lies in the principal plane as shown by up-down arrows, while in the ordinary (o-) ray it is at right angles to that plane as shown by dots. The angles α and β can be derived from i and properties of the crystal. From Bennett and Bennett (1978).

the optical axis. Huygens' hypothesis leads to the speed v of the e-wave normal being found from the equation $v^2/c^2 = \sin^2\phi/n_e^2 + \cos^2\phi/n_o^2$. Here, ϕ is the angle between the optical axis and the wave normal, not shown in Fig. 8-3.

In biaxial crystals, light is propagated as two extraordinary rays, and there is no ordinary ray obeying Snell's law. In these crystals the wave surface of Huygens is an ellipsoid with three unequal axes (giving three characteristic refractive indices). According to the laws of geometry, there are always two planes through the center that cut the ellipsoid in a circle (Fig. 8-1). The optical axes are perpendicular to these rings, and the angle between them differs from one mineral to another. See discussion in section 10.4 of an important effect connected to this.

Fresnel (1822a) also investigated the optical activity of quartz. He considered this property to be caused by slightly unequal speeds of waves with left-hand or right-hand polarization within the material. He tested his idea in an ingenious way using a composite quartz prism (Fig. 8-2). Fresnel himself suggested that the reason for optical activity in crystalline quartz was related to a spiral arrangement of its constituent particles (molécules integrantes). This was eventually confirmed (see Wahlstrom 1969, and Fig. 8-2).

9 On crystallography and the role of calcite in its development in the first half of the 19th century

9.1 On crystallography and light, 1815-30

Around 1815 scientists had realized (section 5.3) that each crystalline substance belonged to one of six or seven so-called systems, based on their geometry. In the order of decreasing symmetry and using their modern names, they are the following:

-The *cubic* or "regular" system, based on a three-dimensional network of cubical cells. Examples include diamond, halides like common salt (NaCl), and various metal sulfides. As already stated, they do not exhibit double refraction, but certain exceptions will be mentioned in section 18.2.

-Crystals with one optical axis belong to the *tetragonal* and *hexagonal* systems. As the names imply, they have four- and six-fold symmetry around that axis. A seventh system with trigonal symmetry was often counted as a separate one, but especially after the advent of X-ray spectroscopy (1913) it has been considered part of the hexagonal system. Calcite belongs to a rhombohedral class within the trigonal system. Quartz is also trigonal.

-Biaxial crystals are divided into three systems. The one with most symmetry is the *ortho-rhombic* system. Examples of orthorhombic minerals include olivine and aragonite (CaCO₃). The others are the *monoclinic* and the *triclinic* systems; among minerals belonging to these systems are augite and the plagioclase feldspars which are common in Icelandic basalts. In the feldspars as well as in olivine and various other minerals, the content of certain metal ions can have a wide range of values. These variations in composition are reflected in variations in the refractive indices and in the angle between the optical axes. That angle is also dependent on color as first noted by Herschel (1820), and on temperature.

Two papers on the important topic of how the crystal systems can be subdivided into classes on the basis of their symmetry, appeared around 1830. These papers by M.L. Frankenheim and by J.F.C. v. Hessel were however overlooked by others for some decades (see for instance Beckenkamp 1923 and Lima-de-Faria 1990, p. 46). Many numbering and naming conventions have been used for these classes.

9.2 On calcite in particular, and rock crystal (quartz)

All along, calcite was a big player in the development of crystallography and mineralogy, as is evident from the following quotes: "The calcite modification of CaCO₃ is one of the most extensively studied of all crystals" (Wyckoff 1924, p. 356) and "[Calcite is] in a scientific context the most important mineral species...the story of calcite is the story of mineralogy." (Tschermak-Becke 1915, p. 529).

The prominent position of calcite may have two main causes. On one hand its crystal structure is sufficiently asymmetrical to demonstrate important features which distinguish crystals in general from those of the cubic system, but it is not too asymmetrical. See for instance Barlow (1883), and Kreutz (1908) remarks: "The high degree of symmetry in calcite allows one to note at once all irregularities in its appearance, which is much more difficult in less symmetric crystals". On the other hand calcite crystals appear in a large number of forms (cf. Fig. 5-3), first catalogued in the work of Bournon (1808). "...the most developed of all crystal groups is the rhombohedral calcite group" according to Hochstetter (1854, extract). Irby (1878) enumerates over 200 simple forms, and many hundreds of composite forms. According to Bauer (1904, p. 578) about 280 simple forms are known, and "Calcite displays perhaps a greater variety of forms than any other mineral". Similar expressions may be seen in Brauns (1903a, p. 386) and other sources.

Quartz was also studied intensively: "Quartz and calcite are probably the most important minerals, because of their wide distribution, their abilities to crystallize in a variety of ways, and their physical properties" (Beckenkamp 1912, p. 62). Quartz clearly possessed less symmetry than calcite, so that some of its most valuable properties are very different from those of calcite, for instance its optical activity. Quartz had a wide range of applications in physics because of its hardness, transparency, low conductivity for heat and electricity, and excellent elastic properties of quartz fibers. The pressure-induced voltages (piezoelectricity) exhibited by quartz attracted increasing attention in the late 19th century, whereas old claims by Hauy and others regarding such effects in calcite were found to be quite erroneous (e.g. Curie and Curie 1880).

9.3 Properties of the crystals from Iceland

Around 1850 or even earlier, the following must have been known about the Iceland spar from Iceland, in relation to other calcite crystals:

1. It was composed of much purer calcite than other such crystals (see chapter 26 and Appendix 2). Calcite crystals from various locations on the European continent often contained impurities amounting to 0.3 to 5% (Doelter 1912, p. 275-276, Hintze 1930) while samples of the Icelandic material might contain less than 0.05%. Boeke (1915, p. 9) mentions that "In a few important cases, Nature is providing us with minerals which are even purer than the best products of technology", citing Iceland spar as an example.

2. The crystals could reach very large dimensions, the largest one on record being described by Des Cloizeaux (1846-47), about 6 by 3 m in situ.

3. The Icelandic crystals could be easily cleaved along three main directions, whereas such rhombohedrons were not commonly obtained from other calcite localities (cf. Naumann 1885, Hintze 1930). The cleavage planes were very flat and clean, so that polishing was unnecessary. This was an advantage in various experiments. However, it is not clear to me what proportion of the crystals from Iceland had the simple form of the fundamental rhombohedron when in their natural state.

4. In calcite crystals from all localities it was common to observe vesicles (sometimes containing liquid), cloudiness, inclusions, cracks, and twinning (i.e. when a crystal grows in two different mirror-image directions from the same plane). Other irregularities called "optical anomalies" were also noted, such as the presence of two optical axes instead of one (e.g. Madelung 1883). These problems were also known in the material from Helgustadir, but that locality was the only one which also produced a quantity of large flawless crystals. "Particularly beautiful, large and transparent calcite crystals which are found in Iceland, are therefore preferred in experiments on double refraction" says Lummer (1909, p. 835).

In research employing X-rays around 1920 (see chapter 36) it was realized that the crystal structure of some individual samples of Iceland spar was closer to being perfectly regular than that in almost any other natural material. Partington (1952, p. 93 and 148) also mentions the occurrence of near-perfect crystals of diamond and quartz, while others are suspicious of the quartz, preferring for instance to test selected samples of common salt or zinc blende for comparison with theoretical predictions. It was often important for scientists to be able to make use of this quality in their studies. Here we may quote Cornu's (1874a, p. 2-3) introduction to results from his research on double refraction: "These experiments have been made mostly with prisms of Iceland spar, a substance which is especially suited for this type of studies, by its homogenity, its transparence, the geometrical perfection of its cleavage, and its strong double refraction.". Cornu continues on "...the constancy of its optical properties...so that a large number of physicists obtain almost identical results from it". He also states that other non-cubic crystals such as quartz, topaz, barium sulfate, saltpeter, sulfur and aragonite are much inferior materials for optical studies, for one reason or another. The main disadvantage of Iceland spar according to Cornu, lies in its softness and difficulties of polishing flat surfaces in directions other than those of the cleavage planes.

10 Fresnel's successors, 1820-40

10.1 Elastic waves

Around 1820, scientists began attempting a comprehensive analysis of how elastic waves (sound) are transmitted through solid materials. It is clear that this development was closely connected to the successes of A. Fresnel and others in optics. For instance, Cornu (1899) stresses that in transverse waves, Fresnel had introduced an entirely new way in which nature transfers energy from one place to another. "The possible existence of waves which are propagated without change of density, has profoundly modified the mathematical theory of elasticity. Geometers...learned... from Fresnel the most general constitution of elastic media, of which they had not dreamed". And in his well-known historical treatise, Whittaker (1951) states: "...under the stimulus of Fresnel's discoveries, the best intellects of the age were attracted to the subject". Among the first of these were members of the École Polytechnique, because of the relevance of material waves to engineering.

Various complex mathematical problems (cf. Fig. 10-1) were solved in this analysis of elastic waves, and its success has been of great value ever since. Papers by three pioneers in the field may be mentioned here, C. Navier (1824, submitted 1821), A. Cauchy (1823, Fig. 10-2) and S.D. Poisson (1829, 1830, see Lloyd 1834). These papers considered ordinary solid materials like glass or metals. These materials turned out to transmit two types of waves: longitudinal and transverse, the latter travelling at a slower speed. The next step was to set up and solve the wave equations for crystalline elastic materials, where Fresnel had considered the elasticity of the aether to be dependent on direction, "…an unexpected condition and one of extreme importance, which has transformed the fundamental bases of molecular mechanics" (Cornu 1899). Cauchy (e.g. 1828) solved that problem under certain conditions. A point source in a uniaxial crystal was found to generate two waves, a spherical and an ellipsoidal one just like Huygens had predicted for light in Iceland spar. Poisson (1842) also began a study of waves in crystals but was unable to complete it.

Among other French investigators contributing to the science of mechanical stresses, elastic waves and sound in materials after 1830, are P.A. Laurent and G. Lamé. Both also wrote papers on optics, including Lamé (1834). His works turned out to be useful later (see Timoshenko 1953, Gårding 1989), for instance where polarized light was employed in research on the elastic properties of materials (see Delanghe 1928).

10.2 Theoretical approaches to the aether problem on the continent, to 1840

The aether and its elastic wave motion inside materials were contemplated by hundreds, or even thousands, of scientists until after 1890. The theoretical equations about that motion had to agree with the observations that had been made by Fresnel and others regarding the behavior of light at plane interfaces, the shape of the wave surface in doubly refracting materials, and so on. Around 1830 it was clear that if a transverse aether wave encountered a boundary between two different non-absorbing materials (such as glass and salt) it was impossible to satisfy simultaneously reasonable conditions regarding for instance continuity and conservation of energy, using only a refracted and a transmitted transverse ray: a ray performing longitudinal vibrations also had to arise. The theoreticians reacted by for instance giving this last ray an infinite speed.

In A. Cauchy's first solution of the propagation of light waves around 1830 (e.g., Cauchy 1831; see a review by Grailich 1856, p. 74-75) he assumed the motion of the aether to take place in the plane of polarization, rather than at right angles to it as suggested by Fresnel. At a similar time F.E. Neumann (1832a) published a very comprehensive monograph reaching the same conclusion as Cauchy. A fundamental aspect of Neumann's work is that the elastic constant of the aether is dependent on the material but its density is the same everywhere. Neumann (1832b) also presented a general theoretical derivation of wave propagation in uniaxial crystals like calcite. Cauchy (1836) however found a new solution to the wave problem, where the motion of the aether is at right angles to the plane of polarization. He considered this solution to be more realistic than his previous one. Cauchy (1839a, see Fig. 10-1), Plücker (1839) and others also addressed the issue. Detailed theoretical accounts of many aspects of polarized light are given in Radicke's (1839) book.

10.3 Theoretical approaches to the aether problem in the U.K., 1820-40

Around 1820, research in the natural sciences was expanding in the U.K. (including Ireland), to a considerable extent due to the progress of mathematics and physics in France (Timoshenko 1953, p. 222). The emphasis was on theoretical aspects, and optics soon came to be studied. Among those who introduced the works of Fresnel and Cauchy in that field to their countrymen, were MacCullagh (1831), Powell (1835), and Smith (1839). MacCullagh (1836, 1837), Sylvester (1837) and Kelland (1838) wrote about the propagation of light in crystallized materials and at interfaces, utilizing a similar approach as Neumann. Green (1842a) who showed that Cauchy's above-mentioned second method of solution ran into difficulties with boundary conditions, suggested that the aether might have certain jelly-like properties. However, others considered that an awkward way out. MacCullagh also published papers on the theoretical derivation of light reflection from metallic surfaces in 1836-44. This topic had been studied previously by Brewster (1830) and Neumann (1832b), and Cauchy (see section 17.1) was also attacking it around 1840.

MacCullagh (1839) introduced a new type of aether, where energy was stored in rotational movements rather than in compression and tension as in ordinary elastic materials. The consequences of that model agreed quite well with known facts about the behavior of light in materials, but it received little attention for the next forty years. During these forty years, it was much debated whether the oscillations of the aether took place in the plane of polarization or at right angles to it. Scores of papers discussed the theoretical aspects of that problem, some suggesting experimental ways of deciding between the two possibilities. Among later reviews of these issues we may mention those by Thomson (1887-88) and Glazebrook (1885, 1903).

In order to fully describe sound waves in solids, Green (1842b) argued that two independent elastic parameters were needed in simple materials like glass, their number increasing to 21 in the

Man setze ferner in den Werthen von l, n für sin ϕ , cos ϕ , wo sie allein vorkommen, die Werthe

$$\sin \phi = \sin \phi' \cos (\phi - \phi') + \cos \phi' \sin (\phi - \phi')$$

$$\cos \phi = \cos \phi' \cos (\phi - \phi') - \sin \phi' \sin (\phi - \phi'),$$

so erhält man

 $I = \mathcal{A}^{\tau} \cos(\phi + \phi') \{\sin^{\tau} \omega \sin \phi' \cos(\phi - \phi') + \cos \phi' \sin(\phi - \phi')\} + \sin \phi' M$ $n = \cos(\phi + \phi') \{\mathcal{A}^{\tau} \sin^{\tau} \omega \cos \phi' \cos(\phi - \phi') - (C^{\tau} + \mathcal{A}^{\tau} \sin^{\tau} \omega) \sin \phi' \sin(\phi - \phi')\} + \cos \phi' M.$ Es sei jetzt

$$C + A \cos \omega \tan \varphi^* = \frac{I + m' \cos \omega}{n + p' \cos \omega}$$
$$A - C \cos \omega \tan \varphi^* = \frac{I' + m'' \cos \omega}{n + p' \cos \omega},$$

so wird

$$l = Cn + A \cos^{*} \omega m \quad m' = Cp + Al$$

$$l' = An + C \cos^{*} \omega m \quad m^{*} = Ap - Cl$$

oder wenn man die zuletzt angegebenen Werthe von l, m, n, p substituirt:

$$I = C \cos(\phi + \phi') \{A^{2} \sin^{2} \omega \cos \phi' \cos(\phi - \phi') - \sin \phi' \sin(\phi - \phi')\} + C \cos \phi' M$$

$$m' = A \cos(\phi + \phi') \{A^{2} \sin^{2} \omega \sin \phi' \cos(\phi - \phi') + \cos \phi' \sin(\phi - \phi')\} + A \sin \phi' M$$

$$I' = A \sin^{2} \omega \cos(\phi + \phi') \{A^{2} \cos \phi' \cos(\phi - \phi') - \sin \phi' \sin(\phi - \phi')\} + A \cos \phi' M$$

$$m'' = -A^{4} C \sin^{2} \omega \sin \phi' \cos(\phi + \phi') \cos(\phi - \phi') - C \sin \phi' M.$$

$$\mathbf{n} = (\mathbf{R} + \mathbf{G})\mathbf{a}^{*} + (\mathbf{P} + \mathbf{H})\mathbf{v}^{*} + (\mathbf{N} - \mathbf{a}\frac{\mathbf{PQ}}{\mathbf{R}} + \mathbf{I})\mathbf{w}^{*} + \mathbf{etc.}$$

Enfin, si l'on pose

$${}^{(5_1)} \begin{cases} 3 = (\mathbf{L} - 2\frac{\mathbf{Q}\mathbf{R}}{\mathbf{P}} + \mathbf{G})a^* + (\mathbf{R} + \mathbf{H})b^* + (\mathbf{Q} + \mathbf{I})c^*, \\ \mathbf{G} = (\mathbf{R} + \mathbf{G})a^* + (\mathbf{M} - 2\frac{\mathbf{R}\mathbf{P}}{\mathbf{Q}} + \mathbf{H})b^* + (\mathbf{P} + \mathbf{I})c^*, \\ \mathbf{C} = (\mathbf{Q} + \mathbf{G})a^* + (\mathbf{P} + \mathbf{H})b^* + (\mathbf{N} - 2\frac{\mathbf{P}\mathbf{Q}}{\mathbf{K}} + \mathbf{I})c^*, \end{cases}$$

Dans la deuxième série, on a 9 = 90°, et, par suite.

$$T_{i} = \sin i - \frac{a \sin i \cos i}{b \sqrt{-A + a^{i} A \sin^{2} i}},$$

$$T_{i} = -\sin i + \frac{a \sin i \cos i}{b \sqrt{-A + a^{i} A \sin^{2} i}},$$

$$T_{i} - T_{i} = 2 \sin i \left(1 - \frac{a \cos i}{b \sqrt{-A + a^{i} A \sin^{2} i}}\right),$$

$$T_{i} + T_{i} = 0,$$

Ici les effets de gauche et de droite sont symétriques et l'on n'a pas de transport pour i = 0.

Dans la troisième série, on a

$$\begin{split} \varphi &= 45, \quad \cos \varphi = \sin \varphi = \frac{1}{\sqrt{2}}, \\ T_i &= \sin i + \frac{1}{2} \cos i \left[\frac{(1 - a^2 A) \sin i}{A \, a b \, \sqrt{-A + \frac{1}{2} (A \, a^2 - 1) \sin^2 i}} + \sqrt{2} \frac{B}{A} \right], \\ T_i &= -\sin i + \frac{1}{2} \cos i \left[\frac{-(1 - a^2 A) \sin i}{A \, a b \, \sqrt{-A + \frac{1}{2} (A \, a^2 - 1) \sin^2 i}} + \sqrt{2} \frac{B}{A} \right], \end{split}$$

or $u^* \sin^* i = a^* (\cos - \sin \psi + \sin \beta \sin \phi)^*$ + $b^{\alpha}(\sin = \sin \psi + \cos \beta \sin \phi)^{\beta}$(6), the relation between ϕ and ϕ . Putting $\phi - \phi = \theta$, and taking account of (1), (6) becomes $2u^{i} \sin^{i} i + \{a^{i} \cos^{i} a + b^{i} \sin^{i} a\} \{1 - \cos (D + i + \theta)\}$ * $\{a^{i} \sin^{i} \beta + b^{i} \cos^{i} \beta\} \{1 - \cos (D + i - \theta)\}$ + 2 ($a^i \cos a \sin \beta + b^i \sin a \cos \beta$) {cos θ - cos (D + i)}, $F\cos\theta + G\sin\theta + H = 0.....(7),$ 0r where $F = a^{i} \{(\cos^{i} a + \sin^{i} \beta) \cos (D + i) - 2 \cos a \sin \beta\}$ $+ b^{2} \{(\sin^{2} + \cos^{2} \beta) \cos (D + i) - 2 \sin + \cos \beta\},\$ $G = (a^i - b^i) (\sin^i \beta - \cos^i a) \sin (D + i),$ $H = 2a^{4} \sin^{4} i = a^{4} (\cos^{4} a + \sin^{4} \beta - 2 \cos a \sin \beta \cos (D + i))$ $= \delta^{i} \{ \sin^{i} a + \cos^{i} \beta = 2 \sin a \cos \beta \cos (D + i) \}.$ Now when D_r regarded as a function of θ , is a maximum or minimum $\frac{dD}{d\theta} = 0$, whence from (7) = $F \sin \theta + G \cos \theta = 0$;

and eliminating θ from this equation and (7), we have $F^{a} + G^{a} = H^{a}$.

Putting for F, G and H their values, and reducing, this equation becomes

$$\begin{split} & \sin^4 \left(D+i\right) \, a^4 b^4 - \left(\cos^4 a + \sin^4 \beta - 2 \cos \left(D+i\right) \cos s + \sin \beta\right) \, u^2 a^4 \\ & - \left(\sin^4 a + \cos^4 \beta - 2 \cos \left(D+i\right) \sin a \, \cos \beta\right) \, u^2 b^4 + \sin^4 i \, u^4 = 0, \ , (\delta). \end{split}$$

This equation will be rendered more convenient for numerical calculation by replacing products and powers of sines and cosines by sums and differences. Treated in this manner, the equation becomes

 $\begin{array}{l} \operatorname{versin}\ 2\left(D+i\right)a^{i}b^{i}-\left(A+B\right)u^{i}a^{i}-\left(A-B\right)u^{i}b^{i}\\ +\operatorname{versin}\ 2ia^{i}=0,\ldots,9\}, \end{array}$

where $A = \operatorname{versin} D + \operatorname{versin} (D + 2i)$, $B = \cos 2a - \cos 2\beta - \cos (D + 2s) + \cos (D + 2\beta)$.

If the principal plane AOC of the crystal bisects the angle between the refracting faces, we have $s = \frac{i}{2}$, $\beta = \frac{\pi}{2} \cdot \frac{i}{2}$, whence from (8), putting $D + i = \Delta$,

$$\left(u^{t}\sin^{t}\frac{\Delta}{2}-u^{t}\sin^{t}\frac{i}{2}\right)\left(b^{t}\cos^{t}\frac{\Delta}{2}-u^{t}\cos^{t}\frac{i}{2}\right)=0$$

Figure 10-1. Mathematical derivations in theories of double refraction could become quite complex. In some papers in 19th century journals they fill pages, and in doctoral theses even tens of pages. The formulas at the top are from Neumann (1835). Below, those on the left are from Cauchy (1839) and Billet (1859), while those on the right are from Stokes (1846).



Figure 10-2. Two scientists who published important papers on polarized light after 1830; however, they are now chiefly known for their contributions to other fields of science. Left: G.B. Airy (1801-1892), astronomer. Right: A.L. Cauchy (1789-1857), mathematician.

least symmetric crystal classes. Earlier, Navier and Cauchy had considered 15 parameters to be sufficient in the latter case, and some used a single parameter for glass. This led to a long-standing controversy, which increased interest in experimental research on elasticity (Timoshenko 1953). See for instance in section 15.3 below.

Airy (1833c, Fig. 10-2) published a valuable essay on the connections between double refraction and optical activity in quartz. MacCullagh made an extensive theoretical study of that matter after 1835, and his conclusions were confirmed in measurements by J. Jamin around 1850 (Verdet-Exner 1887).

10.4 Experiments in 1830-40

In the experimental field, two events in the U.K. strengthened the case for Fresnel's hypotheses on the propagation of light. One was an investigation by Airy (1833b) of the polarization of Newton's rings (which are seen when the surfaces of two objects, one or both being transparent, are almost in contact). His results agreed with the theoretical predictions. The other event concerned the case when a light wave in a biaxial crystal propagates in the direction of the dimple point L in Fig. 8-1. Soon after it was pointed out that such a wave should give rise to a peculiar refraction effect, this "conical refraction" was observed by H. Lloyd (1833). It attracted considerable attention and is still covered in textbooks on optics. However, people continued being aware of the underlying flaws in Fresnel's derivations (Lloyd 1834).

Seebeck (1831, 1836) and Neumann (1837a,b) in Germany made measurements of the light reflected from natural and polished surfaces of calcite, also agreeing with theory. Liebisch (1891, p. 440) states that "The careful observations by A. Seebeck on the complete polarization of light by reflection from calcite....were of prime importance to the field of crystal optics" because they tested certain theoretical derivations by Neumann (1835, cf. Fig. 10-1). Rudberg (1829) measured

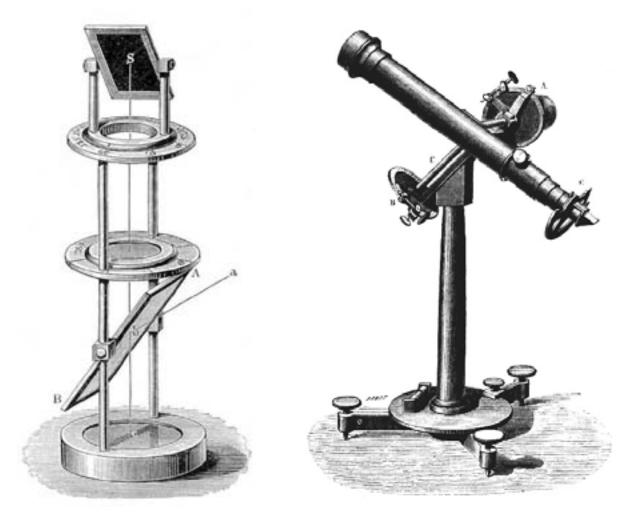


Figure 10-3. Left: A device employing a glass mirror to polarize light. It is often attributed to J. Nörrenberg but it was in fact first described by J.T. Mayer (1812). These instruments were used in optics instruction, simple testing, etc. Thin crystal plates could be placed on the central platform, for inspection in the polarized light from the lower mirror. The picture is from Drude (1900a). Right: An instrument employed by Wartmann (1851) to investigate polarization of the celestial thermal radiation. Picture from a catalog issued by the instrument maker J. Duboscq (1885).

the refraction of light in prisms cut from the biaxial minerals topaz and aragonite, and reported that his results were consistent with Fresnel's hypotheses.

According to Cheshire (1923), the above-mentioned G.B. Airy constructed an instrument with glass mirrors for use in his experiments on polarized light around 1830. Various people later copied this instrument, in some cases replacing the mirrors with Nicol prisms (to be described in section 12.2), but Airy's role in its invention was forgotten. It probably resembled that of J. Nörrenberg (Fig. 10-3) which was also designed around 1830 (although not demonstrated until much later, see section 18.2), but the first one to describe such a device in print may have been J.T. Mayer (1812), cf. an account in Die Naturwissenschaften 14, 511-512, 1926.

10.5 Infra-red radiation, dispersion, and other matters 1812-50

In the 1810s, J.E. Bérard (e.g. 1812) carried out some experiments on the invisible radiation emitted by hot objects and fire. His results indicated among other things that this radiation could be polarized in the same way as visible light. Bérard also used an Iceland spar rhomb to demonstrate polarization of the heat rays from the Sun by reflection. His findings met with some scepti-

cism (Berthollet et al. 1813, Powell 1830-31). According to Barr (1962), A.M. Ampère around 1832 became the first scientist to be convinced that heat radiation was light which the human eye could not sense, see for instance Ampère (1835). Soon thereafter, M. Melloni (1836, 1837, and more) made extensive measurements on heat rays, see Barr (1962). With the aid of a very sensitive electric thermometer, he showed that they had properties similar to those of visible light regarding for instance reflection, refraction, and polarization, including their optical rotation in quartz (Biot and Melloni 1836). Melloni and J.D. Forbes (e.g. 1836) who obtained similar results, mostly used tourmaline and tilted sheets of glass or mica in their polarization studies.

The first observations on the polarization of ultraviolet rays that have come to my attention, were made by Sutherland (1841) using Iceland spar rhombs and tilted mica sheets. Melloni (1842) emphasized that the only difference between infra-red, visible, and ultra-violet light was in their wavelengths. Later, Fizeau and Foucault (1847), Knoblauch (1848, 1852), and Provostaye and Desains (1849) made comprehensive experiments on the polarization of infra-red rays, partly with Iceland spar rhombs and Nicol prisms (see section 12.2), confirming that it behaved just like light. Wartmann (1847) observed qualitatively using mica sheets as polarizers, that heat radiation showed a Faraday effect in a magnetic field (section 16.1) as light did. He later (Wartmann 1851) devised a sort of telescope containing a large Nicol prism (7.5 cm long) and a thermometer to investigate the polarization of sky light (Fig. 10-3). He found it to depend on direction in the same way as visible light. By replacing the thermometer with a photographic plate, the polarization of the ultraviolet sky-light could be demonstrated in the same way.

Until after 1870 the wave theory of light could not provide any physical explanation of the dispersion of light, i.e. the change of refractive index with wavelength, even in simple materials like air, water or glass. Rudberg (1831) made extensive measurements of this property in various crystals including Iceland spar. In the next decade or so A. Cauchy, B. Powell and others published several theoretical papers where they among other things attempted to simultaneously account for dispersion and double refraction (see Chapter 21 of Verdet-Exner 1887).

11 Indications of anisotropy, 1820-35

Many bulk properties of material objects, such as their temperature or density, may be described by a single number and its associated units of measurement. However, there are also material properties which vary with direction within an object; these properties are connected to vector quantities. The view that such directional dependence may occur in crystals, was first expressed by Weiss (1804); see also the remark by Young (1809) referred to in section 6.6. This phenomenon, called anisotropy, has turned out to be a very important aspect of crystal physics. It is a fundamental property of all the crystal classes except the cubic one.

In the summer of 1823, E. Mitscherlich was carrying out research with the famous chemist J.J. Berzelius. Among other things he measured the angle between the sides of an Iceland spar crystal twice in the same day (see obituary by G. Rose, 1864), but in between the measurements the laboratory room had been warmed by sunlight. As a result, the angle had changed; not much but measurably because Mitscherlich's goniometer was quite accurate and the cleavage planes of the crystal were very flat. This was most unexpected, so Mitscherlich started measuring the angles between the faces of Iceland spar and of some other crystals at many temperatures. The Iceland spar was found to expand with temperature in the direction of the optical axis, but contract in directions at right angles to it. This caused the above-mentioned angles to change by about 1 minute of arc per 10°C (Fig. 11-1). The results (Mitscherlich 1825) generated much interest: the thermal expansion of solids had of course been measured before, but generally by heating of long multicrystalline rods. No one had looked for directional variations or changes of shape.

Such thermal changes were studied in many other crystalline materials during the next several years. Rudberg (1828, 1831) and others measured the expansion of Iceland spar and various other crystals with improved accuracy over a wider temperature interval, and attempted to correlate it with changes in refractive indices. Corresponding measurements on crystals with two optical axes (such as gypsum) gave complicated results regarding for instance the direction of an "axis of expansion" relative to their optical axes (e.g. Neumann 1833). Neumann (1834) also initiated a study of the directional dependence of elastic properties in crystals, but his theoretical approach was later found to be inadequate.

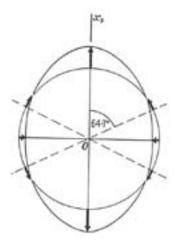
Frankenheim (1829) made a thorough survey of the hardness of crystals, finding it to be generally dependent on direction. Savart (1829) studied the oscillations of circular plates made from crystals (mostly quartz, but also calcite and some other minerals), concluding among other things that the elastic constant (Young's modulus) along the optical axis of calcite was different from that at right angles to it.

In section 7.3 it was described how Brewster (1816a,b) and others investigated the occurrence of stresses and double refraction (caused by pressure or unequal heating) in otherwise isotropic

Ich habe dann den Kalkspath auf die oben erwähnte Weise in heifsem Quecksilber gemessen.

T. 8° R. 105° 3' 591/

72		104	57	234	also	für	64°	eine	Veränderung	von	0	° 6'	36"
				321				-		-			
127			52	0		-	119	-					
131	*		51	25		-	123			-	•	12	341
						-	380°		-	-		38	37*
					-	-	80°			-		8	8"



Der spitze Winkel gab ein vollkommen hiermit übereinstimmendes Resultat.

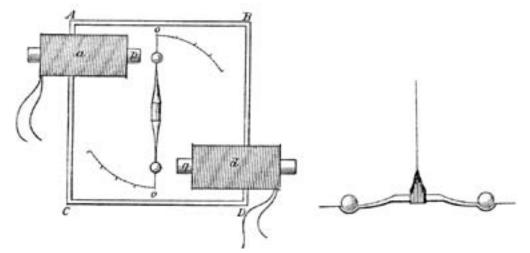


Figure 11-1. Experiments on anisotropic properties of Iceland spar. Top left: Data recorded by Mitscherlich (1825) from measurements of the angles between the sides of an Iceland spar crystal at various temperatures. Right: Let us slice an Iceland spar crystal along a plane containing its optical axis (x3) and paint a circle on that plane. If we then heat the crystal, the circle will change into an ellipse. The picture (from J.F. Nye: Physical Properties of Crystals, 1957) is greatly exaggerated. Below: In the experiment by J. Tyndall (1851) the pole of a magnet pushed an Iceland spar sphere away from itself with a 10% greater force, when the optical axis was parallel to the field as compared to when the axis was perpendicular to the field. On the left the view is from the top, on the right it is from the side. Drawing from Liebisch (1891).

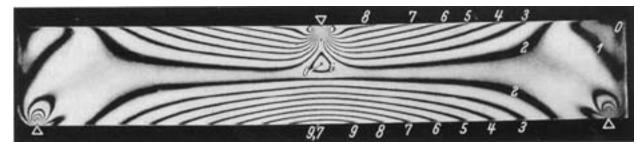


Figure 11-2. Neumann (1843) followed up on the discovery by Seebeck (1814) and Brewster (1816a,b) that glass becomes anisotropic and exhibits double refraction if it is under stress from externally applied forces or unequal heating. After 1850, G. Wertheim and J.C. Maxwell carried out further research on this aspect. The diagram which is of a more recent date (Föppl and Mönch 1959) shows the interference pattern appearing in a bent rectangular glass rod, illuminated by polarized light and viewed through an analyzer.

materials like jelly or glass. F.E. Neumann (1843) published a major theoretical study analysing these effects in greater detail than had been done previously. He also tested his conclusions experimentally in a few simple cases, using glass models observed through calcite prisms. As an example, a glass rod of rectangular cross section which is bent by a load, exhibits in monochromatic polarized light a pattern of light and dark stripes (Fig. 11-2). These stripes reflect in a certain way the state of stress within the glass. If we understand the physics of their relationship, then we have at our disposal a straightforward but powerful method for studying the deformations caused by forces applied to solid objects, and hence their mechanical strength. To quote Timoshenko (1953, p. 249): "The most important contributions made by Neumann to the theory of elasticity are included in his great memoir dealing with double refraction.".

Duhamel (1832) wrote a theoretical paper on the conduction of heat in crystals, where he anticipated that their conduction coefficients would be dependent on direction. Few people paid any notice to his work, and the matter was dormant until around 1850, see section 15.2. With the benefit of hindsight, it is now evident why this property and various others should be anisotropic in crystals: the very fact that their cohesion (i.e. resistance to cleavage) varies a great deal with direction indicates that the forces between their constituent particles have directional preferences. See a remark made by J. Tyndall (1855, footnote p. 5).

12 On instrumentation, 1820-40

12.1 Beam-splitting prisms

Wollaston (1820) invented a prism that makes the two polarized components of a light beam diverge more from each other than Rochon's prism of the same dimensions does. However, Wollaston's prism has a greater dispersion, which can be a disadvantage. These prisms are made by gluing together two wedges of a doubly refracting material, with different orientations of their optical axes (Fig. 12-1). The angle between the two rays can be 15-20° if calcite is used (Wright 1919a), while it is less than 1° with quartz which was chosen by Wollaston for the purpose of measuring small angles. Arago (1847) measured the angular diameters of the planets with devices containing Rochon-type prisms from quartz. Prisms of this type are mentioned occasionally below (see Fig. 13-7). They are still being manufactured for use in various optical instruments.

12.2 The Nicol prism, 1829: an invention of great importance

As mentioned in previous sections, a ray or a wide beam of light is separated into two polarized ones on its way through a doubly refracting material such as Iceland spar. In research where polarized light is being generated or analyzed, it is often only necessary to make use of one of these beams, and the intrusion of the other one into the observer's field of view can be disturbing. Rochon- and Wollaston-prisms solved this problem partially by deflecting it sideways. In natural calcite rhombs it was possible to roughen a spot on the face closest to the observer's eye, to stop the unwanted beam. Brewster (1819) pointed out that by coating the roughened spot with a liquid of suitable refractive index, one beam would pass through it relatively unharmed while the other one would be diffused at the surface and hardly visible. I have not seen any subsequent papers indicating explicitly that the writers were familiar with this simple technique, but it may well have been widely used.

W. Nicol in Edinburgh realized that the best solution to the overlap problem would be to make one of the two polarized rays disappear altogether. He accomplished this by slicing an oblong rhomb of Iceland spar diagonally (Fig. 12-1) and gluing the pieces together with wood resin (Canada balsam). Its refractive index was such that the extraordinary ray in the spar passes through it while the ordinary one suffers total reflection at the glue. The latter ray could then be for instance absorbed by a coat of black paint or graphite on the lateral surface of the crystal. Any directional component of the incident light could be selected for transmission, by rotating the prism around its axis or by making corresponding provisions at the source of the light. Nicol's (1829) modest

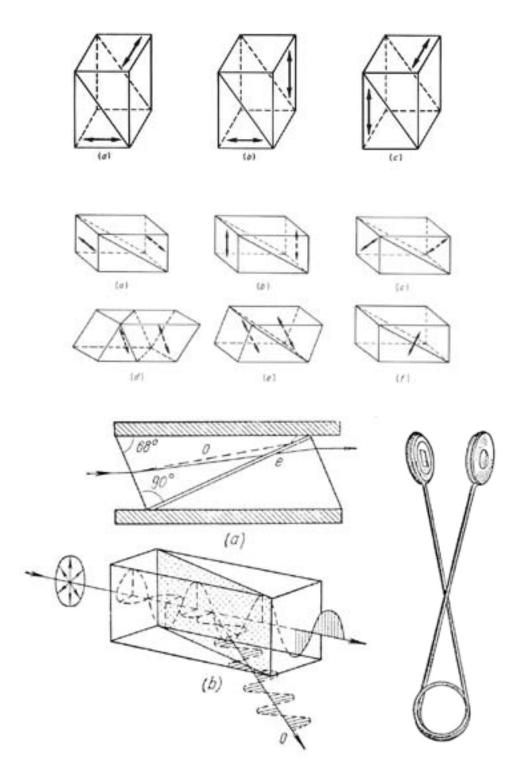


Figure 12-1. Top: Schematic diagrams of three common types of beam-splitting birefringent prisms, usually made from Iceland spar or quartz. If a beam of unpolarized light is incident on such a prism horizontally from the left, two polarized rays diverge from it towards the right. (a) Rochon prism, (b) Senarmont prism and (c) Wollaston prism. Arrows indicate the directions of optical axes. Sometimes, one half was of glass.

Center: Schematic diagrams of six common types of polarizing prisms. Arrows indicate optical axes. (a-c) are of Glan-type, i.e. a Glan-Thompson (or Glazebrook) prism, a Lippich prism, and a Frank-Ritter prism. (d-f) are of Nicol-type, i.e. an original Nicol prism, a Halle prism and a Hartnack-Prazmowski prism. In some cases, the two halves are separated by an air gap instead of a resin cement or oil. From Bennett and Bennett (1978).

Bottom left: (a) A light beam enters a Nicol prism from the left. The prism divides it into two rays o and e, each linearly polarized. The o-ray is reflected sideways from the Canada-balsam cement and is discarded, but the e-ray continues through the other half of the prism. (b) Here the prism has been rotated 90° about the direction of the light ray. Right: Tourmaline tongs, a simple device for observing double refraction in minerals. From Wood (1977).

note describing this invention has probably had a greater impact than most other scientific papers of comparable length. These devices, which were named after Nicol and gained popularity within a few years, were often employed in pairs: one as a polarizer of ordinary light and the other (sometimes smaller) one along with accessories as an analyzer of that light after some interaction with matter.

The production of prisms from Iceland spar is not an easy task. It had to be relatively free from defects, which could occur even in the best material from Iceland (see Goldschmidt 1907, Hughes 1937). Iceland spar is quite soft, but it cracks or splits easily along its three main cleavage directions. Instead of cutting a single natural rhomb into two diagonal wedges, two rhombs were often ground down to the wedge shape to reduce the risk of breakage. The hardness of the spar crystals varies with direction, and flakes could get detached during grinding (Halle 1921). The precise adjustment of two Nicol prisms along the same axis was difficult (Glazebrook 1880b) and careful handling was needed. For instance, exposure to too much light could cause heating and damage to the glue (Laurent 1879, Sleeman 1884, Schulz 1925, Schumann and Piller 1950).

12.3 Other means of producing and analyzing polarized light

Two other methods were at this time available for producing polarized light and analyzing it. One was based on the fact that light reflected from glass at the Brewster angle is linearly polarized. This method, sometimes attributed to J. Nörrenberg, has always been used to some extent in instruments (Fig. 10-3, Fig. 13-1) especially if a wide field of view was required (Dove 1860). It has however some drawbacks: one is that only about 15% of incident unpolarized light is reflected, while a Nicol prism delivers about 40%. Light that has passed through a pile of 10-15 tilting glass plates is also almost linearly polarized. See section 33.1.

The other method was based on the phenomenon of dichroism, already mentioned. It is due to the fact that doubly refracting substances absorb the two polarized light components in different amounts. In calcite this difference is so small as to be of no consequence, but one needs only a 1-2 mm thick plate of some minerals to stop one ray almost completely. Such plates, however, also have drawbacks, including the absorption of both rays generally being color-dependent (Talbot 1834a; Rosenbusch 1885, p. 109-111). The mineral tourmaline has been most used for this purpose from around 1820, but other drawbacks include its brittleness (Barnard 1863). The main application of this method has been in simple devices, such as so-called "tourmaline tongs" (Fig. 12-1) for quick testing of the optical properties of gemstones.

12.4 Optical activity, continued – Biot 1832-40

Here we have to start with a discussion of the term "sugar", which can apply to many substances having certain similar properties. Fundamental aspects of their structure and chemical behavior were not clarified until after 1900. Solutions of most sugar compounds are optically active, but in a variable degree and either to the right or left. They may be divided into mono-, diand trisaccharides, plus polysaccharides such as starch from grain or potatoes. All belong to the class of carbohydrates, having a general formula (CH2O)n. The most common monosaccharides contain a chain of 5 carbon atoms (pentoses) or 6 (hexoses). They may then be divided into aldose and ketose sugars depending on the presence of an aldehyde or ketone group at one end of the chain. The best-known monosaccharides are the aldohexoses glucose (grape sugar) and galactose, and the ketohexose fructose. Of these, glucose may be considered the most important molecule in the living world. The most familiar disaccharide is sucrose (saccharose, ordinary sugar) with 12 carbon atoms. Its original source as a foodstuff was sugar cane which was chiefly cultivated around the Caribbean. However, it was known already before 1800 that sugar could also be obtained from a species of beet. Due to a blockade of ports in the Napoleonic wars, production of beet sugar in Europe increased greatly in 1806-15 but it then fell off and was not resumed until around 1840. In 1811 it was discovered that glucose could be made from starch by the action of acids (hydrolysis).

J.B. Biot who had discovered optical activity in organic liquids and solutions (section 6.8), realized that measurements of this property could be a valuable aid in research on such compounds. After having worked in unrelated fields of science during 1820-32, he pointed out (Biot 1833) the usefulness of the method in estimating the sugar content in juices from fruits or other parts of plants. He also began a study of the effects of acids upon sugars and starch (Biot and Persoz 1833). Among these effects is a change in optical activity, in some cases from left-handed to right-handed rotation. Biot's (1835) term describing this process, "inversion", is still used. So is the name "dextrine" given by Biot and Persoz to a water-soluble viscous material obtained from starch with acids, or by heating it to more than 120°C. The name reflects the fact that it rotates polarized light strongly to the right. The dextrin (which is not a pure compound, but a mixture of many carbohydrates of various molecular sizes) soon acquired practical importance as an ingredient in bakery products, drinks, sweets, and glue. The increased knowledge of the character of starch compounds prompted further studies on them, Payen and Persoz (1833, 1834) becoming the first to isolate a biological catalyst, i.e. an enzyme. This material which they called "diastase" (now: β-amylase) transformed starch in a similar way as heating or acid treatment. More on sugar compounds will be presented in sections 13.2 and 18.1, including the processes involved in hydrolysis and fermentation.

Biot (1817) had found with a rather inaccurate type of polariscope, that any optically active organic liquid exhibited approximately the same amount of this activity per unit mass, whether it was measured in pure form, dissolved in an inactive liquid, or even as a gas. The gas part of these results was however only obtained from a single measurement with turpentine vapor in a 30-m long glass tube just before it exploded. That experiment was not repeated for almost half a century (Gernez 1864). Biot (1835) hence defined the term "specific rotatory power" of a compound. The term is still in use, but with improved instrumentation it was found that this property was to some extent dependent on the solvent used, the concentration of the solution, and the temperature, see for instance in Landolt (1898) and Partington (1953).

Biot had discovered early on, that the optical activity of both quartz and organic liquids varied approximately as the inverse second power of wavelength of the light used. Solutions of tartaric acid and its derivatives constituted a significant exception from this general relation, and Biot (1838) carried out a very thorough investigation of their optical properties. This behavior which was called "anomalous rotatory dispersion" was to have considerable impact upon the chemical sciences, as will be presented in section 16.2 and later.

13 Improvements to Nicol prisms; polarimeters and measurements of optical activity from 1840

13.1 Introduction

Polarized light had become a well-known subject in science by 1830, as may be seen e.g. from an advertisement by J.W. Albert (1828). He was selling a variety of prisms, plates, etc. of Iceland spar and other crystals for experimentation. I have not found much information on the initial reception of W. Nicol's polarizing prism in the years after its invention in 1828-29. Talbot (1834a) states that he did not hear of it until he read the 1833 German translation of Nicol's paper. Dove (1835, p. 607) however mentions an instrument maker in Berlin "whose Nicol prisms are already in the hands of several scientists". In the late 1830's it was noted that the performance of the prism could be improved by widening its field of view, reducing the lateral shift of the transmitted ray, and increasing transmission through the glue layer and end faces. Nicol (1839) himself suggested some such improvements, also Sang (1837 and 1891), Spassky (1838) and Radicke (1840). Some scientists however continued using Iceland spar rhombs as polarizers in various situations, for instance A. Becquerel (1849), Jamin (1850a), Billet (1858-59, I, p. 345), Cornu (1867, 1889), Kerr (1894), J. Becquerel (1910), Nichols and Howes (1914), and some of those investigating the Zeeman effect (see chapter 30) after 1897.

In addition to the inclusion of Nicol prisms in various types of instruments to be described later in this compilation, they were used on their own in many experiments. As an example we may consider a simple but fundamental question regarding a narrow ray of monochromatic light: is the light-wave oscillation in it completely regular for a long time, or is the ray perhaps composed of short successive "strands" of light that are out of step with each other ? This could be checked by splitting a ray in two and delaying one of these before they were brought together again to form an interference pattern. Fizeau and Foucault (1845, Fig. 13-2) sent sunlight through thin plates of Iceland spar and quartz between two Nicol prisms, and then through a spectroscope. By observing the presence of certain dark lines in the spectrum, it could be shown that the oscillations in light were continuous for at least seven thousand wavelengths. Stefan (1864b) used a similar method, remarking that a better one was yet to be found, and doubled this limit. Mascart (1872, p. 196) managed to demonstrate interference in beams of sodium light differing in phase by 105 thousand wavelengths "... in spite of the enormous crystal thicknesses (36 cm) required. This was because Iceland spar is perhaps the most homogeneous of all available transparent substances..."; he also states that its cleavage planes are more even than polished glass surfaces. Around 1900, people had succeeded in obtaining interference between light waves with a difference of two million wavelengths by other methods (see Winkelmann 1906, p. 1135). The reasons for the finite length of light pulses were much clarified after the advent of quantum theory in the early 1900s.

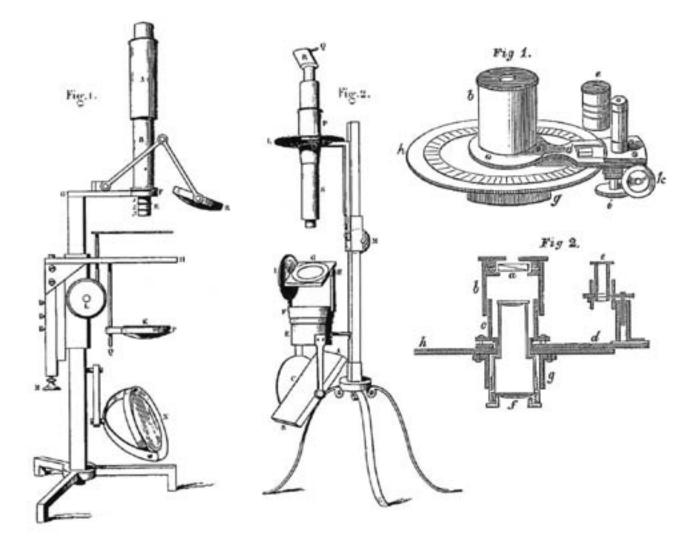


Figure 13-1. Left: Polarizing microscope (Fig. 1) and polariscope (Fig. 2) described by Amici (1844). R at the top of Fig. 2 is a rhomb of Iceland spar, two such rhombs are in the microscope but not shown. Both these instruments are among the first of their type for which contemporary drawings are available. Right: External appearance and cross-section of the instrument designed by Leeson (1845) for studies of refraction in crystals. The Rochon prism at the top is made from Iceland spar and glass.

The first one to equip a microscope with Nicol prisms was probably H. Fox Talbot, who later became a pioneer in photography. Talbot (1834a, 1837) describes various observations with this equipment, and some developments in the field took place in Britain in the next couple of decades (see notes in the reference list, and the first paragraph of section 18.2). However, polarizing microscopes probably did not become widely known by amateur microscopists until the mid-1840s, or by geologists until the 1860s. According to a short note on p. 414 of Comptes Rendus 8, J.B. and H. Soleil presented in 1839 an instrument for measuring the axial angle in biaxial crystals, and it probably contained Nicol prisms. Leeson (1845, Fig. 13-1) described a device with an Iceland spar prism for studying the refractive indices of crystals. Lima-de-Faria (1990) considers the first polarizing microscope to have been designed by Amici (1844, Fig. 1 in Fig. 13-1); it has a glass-mirror polarizer and an Iceland spar rhomb analyzer. A description and a drawing of this instrument may in fact be found in an earlier book by R. Gerbi: Corso Elementare di Fisica, vol. 3 p. 229-231, Pisa 1832. Brewster (1848a) pointed out that bothersome reflections in microscopes may be reduced by the use of polarized light, and Brücke (1848) used a microscope with two Ni-

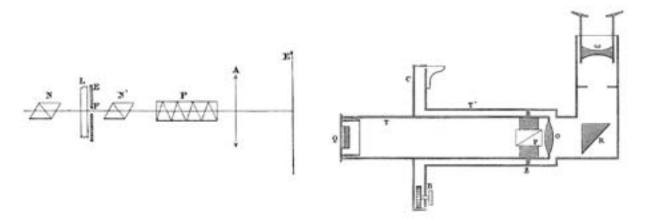


Figure 13-2. Left: Fizeau and Foucault (1845, 1847) showed that a so-called "channeled spectrum" with dark stripes appears on the screen E' due to interference between two components of polarized light emerging from the birefringent crystal plate L. P is a dispersing prism and A is a lens. Many others later employed similar techniques in optical research. Right: A simple instrument (horloge polaire, by J.B. Soleil) for finding the direction to the Sun from the polarization of skylight. The first description of such a device is due to Wheatstone (1848). The diagram is from Billet (1858-59).

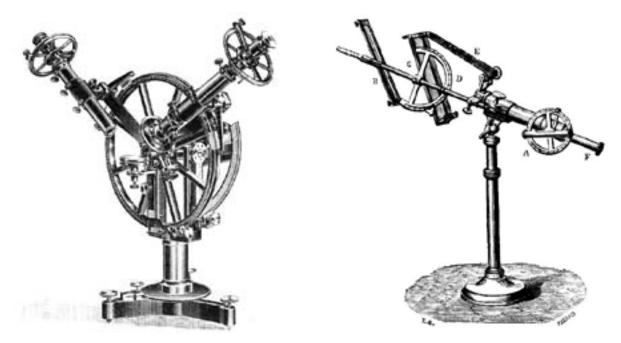


Figure 13-3. Left: Jamin's (1850a) circle equipped with Iceland spar prisms and quartz wedges, for analysing elliptically polarized light. The figure is from a Société Genevoise catalog issued in 1900. Right: F. Arago's polariscope for observing polarization of skylight, described in 1841. From the catalog of J. Duboscq (1885).

col prisms in work on colored rings in thin crystalline plates. Microscopes and related equipment will be discussed further in sections 18.2, 18.3 and later.

13.2 Rapid development of polarimeters, 1840-50

When people began making fairly accurate measurements with polariscopes, they were increasingly often called polarimeters in Europe and later in the U.S., though not always in a consistent way. In the section on J.B. Biot in Dictionary of Scientific Biography (1980-90) and other sources, his improvements on older types of polariscopes around 1840 are said to have resulted in the first polarimeter (e.g. Biot 1840a, Fig. 13-5). They developed rapidly when Nicol prisms replaced other methods of polarizing and analyzing light. See publications on the use of these devices by Dove (1835), Babinet (1837), Biot (1840a, 1845a,b), Brewster (1841), Pereira (1846) and Duboscq and Soleil (1850, Fig. 13-5). They had three main fields of application, not always explained fully in the publications involved: (A) the measurement of the rotation of plane-polarized light in a sample; this type was called polaristrobometer by Wild (1870) and Landolt (1898). (B) The analysis of the principal parameters of elliptically polarized light, which might for instance be produced from plane-polarized light on reflection from a surface. The chief difference between these two lies in the provision of a phase compensator (section 13.5) in the latter type (Zehnder 1908a) which was later named ellipsometer. (C) Detection and measurement of plane-polarized light in the presence of unpolarized light. According to Verdet-Exner (1887, section 486) a new polariscope by Arago (Fig. 13-3) demonstrated to the French Academy in 1841 was of this type.

Ventzke (1842, 1843) improved Biot's polarimeter (in the A-category above). His instrument which is illustrated in his paper, contained two Nicol prisms. He, as well as for instance Wagenmann (1842) suggested and tested various uses for it in the sugar industry. The earliest practical polarimeter of this type, however, was constructed by E. Mitscherlich in connection with his research (Mitscherlich 1841, see D.S.B.) on fermentation and digestion. It also incorporated improvements on Biot's device, and its description appeared in Mitscherlich's (1844, p. 360-366) textbook of chemistry. Biot (1845c) immediately praised the virtues of this new model. A less known instrument for similar use is due to Leeson (1843-45) who investigated the optical activity of various essential oils from plants.

Biot (1840b) pointed out that polarimeters provided a rapid and simple method of measuring the concentration of sugar solutions. They would therefore be very suitable for quality control in the production, trade, and industrial consumption of sugar; see for instance Soubeiran (1842). However, it was soon found that in the fluids being processed at various stages in the beet-sugar industry, there are other optically active compounds which can distort the interpretation of results. Biot (e.g. 1843a) therefore made an extensive series of experiments where he measured the activity of sugar solutions before and after treatment with acids. This treatment affects the specific rotatory power of sugar considerably in a known way, but does not significantly change the activity of other compounds. Clerget (1846, 1849, Fig. 13-1) continued Biot's work, eventually presenting precise recipes for reliable measurement of sugar content. Some of these recipes with modifications by Herzfeld (1890) and others, were in use into the 20th century (Rolfe 1905, Saillard 1913), improved versions even until the 1940s (Hinton 1930, Bates et al. 1942, p. 126-132).

Dubrunfaut (1847) argued on the basis of polarimetric measurements, that a type of sugar which he called malt-glucose and later maltose, was produced in the fermentation of starch. This discovery went unheeded for a long while, but it was confirmed by O'Sullivan (1874) who also depended to a considerable extent on measurements of optical activity. The maltose turned out to be a disaccharide composed of two glucose molecules (minus water), and it is the chief building block both of starch from the vegetable kingdom and of related polysaccharides such as glycogen (section 18.1) from the animal kingdom. Dubrunfaut (1849) also found by detailed research, that the "inversion" of optical activity of sucrose brought about by acids or fermentation, is due to the splitting of the dextro-rotatory sucrose molecule. Its constituent parts are glucose (also often called dextrose) and fructose (also called levulose) with stronger specific rotation. This result was a major step in the understanding of the nature of sugar compounds; for instance, Dubrunfaut himself showed with the aid of polarimeters and other methods, that processes of this kind took place in honey and in raisins. More on his achievements in sections 18.1 and 27.7.

In diabetic illness, glucose is excreted in the urine, and Biot (1840c, 1845a, 1848) proposed the use of polarimeters for rapid measurements of its concentration in the diagnosis and treatment of this condition. A. Bouchardat (1841), who had been studying diabetes, made such measurements

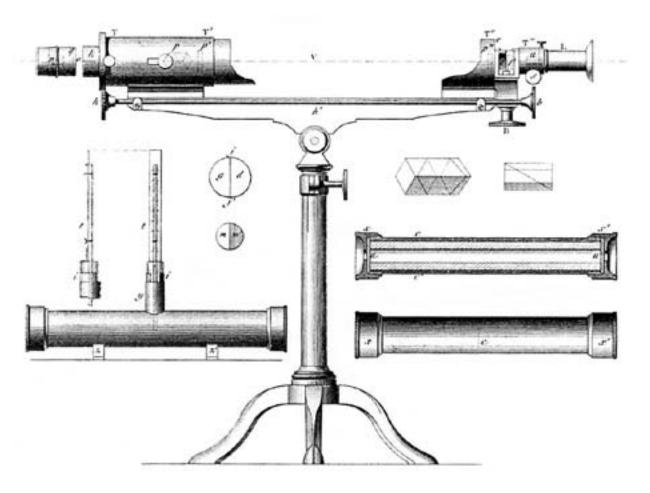


Figure 13-4. Part of an illustration in Clerget (1849) showing his Soleil-made polarimeter for measurements on sugar solutions. Components visible here are the Nicol prisms p and a, quartz wedges, a sample tube, and a thermometer. A light source is to the left.

with Biot's instrument. He (Bouchardat 1842) confirmed Biot's conclusion that albumin and other proteins which occur widely in organisms, are optically active. Bouchardat (1844) also noted the presence of optical activity in various so-called glycosides, a class of chemicals including for instance salicin which was known to reduce fever and inflammation. Bouchardat and Sandras (1845) showed that a diastase (i.e. an enzyme, later called β -amylase) similar to the compound from the plant kingdom mentioned in section 12.4, is produced in the digestive system of animals. It is likely that this finding was to some extent based on polarimetric measurements. Pasteur (1851) found that two amino acids occurring in asparagus, were optically active; they were soon also shown to be among the active materials that (as mentioned earlier in this section) are present in sugar beet.

By 1850, polarimetric methods were sufficiently established to be treated at some length in textbooks of chemistry (e.g. Mitscherlich 1844, Pelouze and Fremy 1850). More on developments in polarimetry will appear in section 18.1, for instance on saccharimeters, a type of polarimeter used especially in the sugar industry. A thesis by Ward (1980) deals with various aspects of the 19th century history of polarimetry.

After 1820, many optical phenomena in the environment were observed through polarimeters. The light from the clear sky, which is polarized to a variable extent depending on direction relative to the observer and the Sun, was studied by Arago as mentioned above, Babinet (1840), Brewster (1842) and later Bernard (1854, 1856) and many others. Wheatstone (1848) designed an instrument with a glass reflector, a gypsum plate and Nicol prism which he called a "polar clock". With this instrument, the direction to the Sun could be found if it was hidden but the sky was at least



Figure 13-5. Some of the first generation of polarimeters in common use. Top left: A polarimeter produced by the Soleil workshop from 1846. The picture is from a Duboscq catalog. Right: A picture from Lummer (1909) of Biot's (1840a) polarimeter with a glass-mirror polarizer and an Iceland spar prism. d is a sample tube. Bottom left: Biot's polarimeter as improved by E. Mitscherlich in 1844, with two Nicol prisms. From Landolt (1898). Right: The polarimeter of Robiquet (1856), containing two Nicol prisms and a double quartz plate f. From Landolt (1898).

partly clear. This device was later improved by others (Fig. 13-2, see also Spottiswoode 1874b) and sold for instance by Duboscq (1885). A century after Wheatstone's invention, a similar instrument was built at the U.S. Bureau of Standards for use in aircraft on polar routes (Rev. Sci. Instr. 20, 1949, p. 460). From around 1968, a discussion has been going on regarding the nature of a "sunstone" mentioned in a couple of old Nordic texts. Claims that such a device may have been a navigational aid based on polarization effects, remain very speculative.

People also investigated the polarization of the light from the aurora borealis, rainbows, halos around the Sun, comets, planets, the zodiacal light, the Sun's corona at eclipses, etc. These measurements were mostly qualitative, of the type C above. Some of these luminous phenomena are complex and not easy to interpret in terms of existing knowledge of that period. Their spectra were also not analyzed until around 1860.

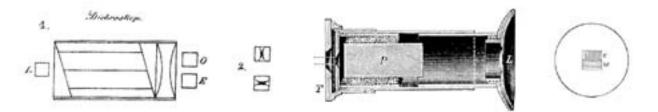


Figure 13-6. Left: Cross-section of Haidinger's (1845) dichroscope. Right: A dichroscope from the Fuess workshop (Johannsen 1914). P is a calcite rhomb, L is a lens. A piece of an unknown crystal is brought to a hole in the plate T on the left-hand side of the dichroscope, while the observer is looking at a light source through it. He then sees two images of the hole, often in different colors. Various manufacturers produced dichroscopes, and they are still in use for rapid inspection of gemstones.

The state of polarization of light reflected from surfaces of various materials was studied thoroughly, e.g. by Senarmont (1840), Powell (1843), Jamin (1845, 1847, 1850a), and Dove (1847b), using method B above. In particular, measurements on metallic reflection were said (Lloyd 1857, p. 156, Partington 1953) to agree reasonably with the theories of Cauchy and Fresnel. An instrument constructed by Jamin (1850a, Fig. 13-3) for such work was later produced commercially and used for decades in similar research, e.g. by Kent (1919).

Polarized light was also used in investigations of how the human eye perceives light and colors (e.g. Wartmann 1843, Dove 1847a, 1853, Foucault and Regnauld 1956, Maxwell 1856). A visual effect called "Haidinger's brushes" generated much attention. These are figures resembling sheaves of grain that are seen (or appear in the eye) in plane-polarized light (Haidinger 1844 and later, Silbermann 1846, and many others). From then on, people used polarized light in various ways to study vision and its physiology, see section 29.7. As mentioned in section 7.3, D. Brewster had already entered that field long before.

13.3 Quarter-wave plates

In addition to Nicol prisms, scientists employed a variety of thin plates and wedges from doubly refracting materials in research involving polarized light. Some of these aided in the analysis of the polarization state of light, others changed that state from for instance circular to linear polarization. Some were made from Iceland spar, others from quartz, mica (muscovite) or clear gypsum (selenite). See section 13.5 for more on certain applications of such devices.

One of the simplest tools of this kind is the so-called quarter-wave plate, often attributed to Airy (1833a) although for instance Mason (1982) considers A. Fresnel to have invented it earlier. It is usually made from quartz or mica (Senarmont 1840); calcite is too difficult to polish to the right thickness. The optical axis is in the plane of the plate. In a light beam passing through the plate, the phase of one vibration component (parallel or perpendicular to the optical axis) will be delayed by 90° relative to the other one. By rotating the plate or the polarizing prism, the phase difference can be varied. See e.g. Fig. 29-4. This is just like in Fresnel's glass rhomb of section 8.1, and both have been used extensively. The plate takes up less space than the prism, but its delay angle is much more wavelength-dependent which can be a disadvantage. In a simple instrument designed around 1850 by Wheatstone (1871) the quarter-wave plate was replaced by reflection from a silvered mirror at a particular angle of incidence, but this technique does not seem to have been much used.

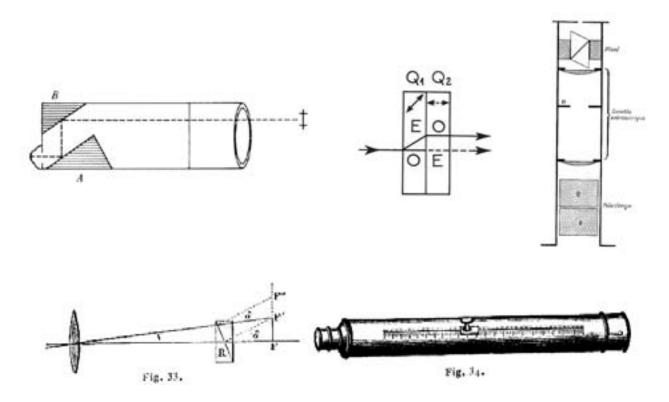


Figure 13-7. Top left: A tube with glass mirrors for observations on the polarization of light, made by the schoolboy J.C. Maxwell in 1848. The diagram is taken from one of the volumes of his collected works. Top center: Savart's (1840) plate, which is made from two specially cut plates of quartz or Iceland spar. It has been used in many instruments for investigating polarization of light. The small arrows indicate optical axes. Diagram from Françon (1963). Right: The telescope (Bouasse 1925) is an example of the use of Savart's plate to detect a polarized component mixed with ordinary light. Bottom: A telemeter with a Rochon prism (probably of quartz), for measuring small angles or estimating distances to objects of known size. From Pellin's (1899) catalog.

13.4 Dichroscopes

So-called dichroscopes (Fig. 13-6) first introduced by Haidinger (1844, 1845), are still in use by mineral collectors to recognize specimens and by jewellers in appraising gemstones. With these, comparison is made of the color and intensity of two polarized light beams coming through the specimen and a calcite rhomb. These beams may have been absorbed differently in the specimen, and the human eye can more easily distinguish such differences than the attributes of a single ray. Powell (1842) had shortly before this used a similar technique in a polariscope, which made it simpler to use than Biot's original version. Comparison methods of this kind later became a standard in polarimeters and photometers, cf. the next section.

13.5 Phase compensators and other analyzing devices

In many optical experiments on matter where linearly polarized light becomes elliptically polarized (Fig. 7-1), the observer needs to find the shape of the ellipse. One parameter determining this shape is the phase difference between x- and y- oscillations of the light. Thin plates which can be applied to change this phase difference continuously or in steps to convert the light back to linear polarization, are generally called compensators. They have mostly been made from quartz, and are employed in conjunction with Nicol prisms. The first compensator was suggested by J. Babinet before or around 1840, but its operation was described by Jamin (1850a). It is made from two thin calibrated quartz wedges that are moved against each other (see Fig. 22-1) until the observer can extinguish the light with his analyzing Nicol prism. J.B. Soleil (1845) described a similar type of compensator containing an additional quartz plate. This makes the measurements more convenient and more accurate, but it is more difficult to construct than the Babinet device. Both are still being produced. Compensators reaching to several wavelengths were introduced later in polarizing microscopes for transmitted light, in order to measure differences between the refractive indices of a crystal specimen.

Somewhat analogous methods were also made use of in the early polarimeters, as described for instance in the papers of Clerget (1846, 1849, Fig. 13-4) on his comprehensive methodology for sugar analysis by polarimetry. Here, the accuracy of rotation readings in a new instrument by J.B. Soleil is improved by the introduction of a split rotatory compensator from quartz. Robiquet (1856, see Fig. 13-5) similarly increased the accuracy of Mitscherlich's polarimeter by adding Soleil's bi-quartz plate into the light path. Then the observer's circular field of view is divided into two halves of different tints, one becoming more bluish and the other more reddish on adjustment. A reading is taken when both were of the same color. Many variations on this technique were soon developed, and such half-shade instruments (polarimètre à penombre, Fig. 18-1) became dominant in polarimetry from the 1870's until the advent of photo-electric sensors in the 20th century. See Landolt (1898), Pellin (1903) and Bates et al. (1942) on these historical aspects.

The so-called Savart's (1840, Fig. 13-7) plate is composed of two calcite or quartz plates, cut in a particular way relative to the optical axis. By the use of this plate between two Nicol prisms, it could be ascertained whether a source of light contained a plane-polarized component, even if it only amounted to 1% of the total light intensity (cf. Wood 1905, Wright 1919b). The application of Savart's plate in some optical instruments will be described in later sections.

14 Speculations regarding Iceland spar exports to 1850

It is clear that a great number of Iceland spar crystals was required for research purposes until 1850, and that most of these came from Iceland. The demand for Nicol prisms must for instance have increased rapidly after 1840 particularly for use in polarimeters, but details regarding the commercial production of both the prisms and the polarimeters remain to be explored. The trading post at Stóra-Breidavík was situated only 2 km from the site at Helgustadir, and it is likely that much of the exported material passed through Stóra-Breidavík on its way to Copenhagen. Soon after the partial liberalization of the monopoly trade in 1786, the economic situation of that post deteriorated and it was finally abandoned around 1806, while new merchants began trading at Eskifjördur about 10 km away from the calcite locality in the other direction (see vol. 5 of the book "Eskja" by Einar Bragi, 1986).

It is quite possible that sailors on the many foreign fishing boats that came here every year, did pick or buy crystals while lying in Reydarfjördur, in order to sell them at home to collectors and scientists. This might even to a greater degree apply to the larger vessels accompanying these fleets, such as transport ships (cf. memoirs of J. Ólafsson, which appeared in the periodical Idunn 1915-16) and military vessels. The number of French boats in Icelandic waters in 1765-92 and 1815-50 averaged over 50, according to E. Pálmadóttir's book "Fransí biskví", published in 1989. This speculation is supported by the existence of a French nautical map no. 1480 from 1855 of a part of the Reydarfjördur coast (printed with another map of the anchorage at Eskifjördur, in scale approx. 1:12,000): its title is "Crocquis des Mouillages du Spath et de Svartas Kiær", i.e. sketch of the anchorages at the Iceland spar site and the Black Skerry.

The foreign travellers who passed the Helgustadir site may also have collected crystals there. Among scientists known to have visited there are F.A. Thienemann in 1820-21 and Jónas Hallgrímsson in 1842, see Fig. 37-2. Other scientists may have established contacts in Iceland for acquiring Iceland spar, such as G.S. Mackenzie who travelled in this country in 1810. For instance, D. Brewster (1815b) mentions that he obtained some Iceland spar crystals from Mackenzie. In a letter to H.C. Ørsted in 1825 (printed in Ørsted's collection of letters, published in 1920) Brewster states that two men recently went from Edinburgh to Iceland, and in the following year it is noted in the Edinburgh New Philosophical Journal (vol. 1, p. 179) that spar from Iceland collected by Mr. Rose and Mr. Brown had been exhibited at a meeting of the Wernerian Natural History Society in Edinburgh. The latter gentleman also brought a mineral specimen from Iceland which Brewster described in an article and gave the name oxahverit. In a letter to Ørsted in 1826, Brewster thanks him for a parcel from Count Vargas Bedemar, who was curator of a large mineral collection belonging to the Crown Prince of Denmark and had written papers about Icelandic minerals. A letter from A. Fresnel in 1825 reveals that Ørsted himself appears to have procured some Iceland spar for him, and Melloni (1836, 1837) acknowledges that a 92-mm piece that he used in his experiments also came from Ørsted.

A notable direct source of information about a crystal-collection effort at Helgustadir may be found in letters from P. Gaimard (1836) on behalf of his expedition on the vessel "La Recherche". Here it is stated that they sent to France a large trunk (caisse) of this "precious substance", much in demand by French and foreign scientists. However, in the published expedition reports (Robert 2007) it appears that its members were quite disappointed by the Helgustadir locality. Dumas (1842) used Iceland spar crystals from E. Robert in his chemical analyses (see chapter 25); these probably were recovered in the 1836 expedition.

In one of his papers in early 1845, J.B. Biot noted that opticians had insufficient stocks of Iceland spar to meet increasing demand for use in constructing the polarimeters he was championing. Soon after, he mentions that a supply may be on its way: this was clearly because he had arranged for the authorities to send a young scientist, A. Des Cloizeaux to the Iceland spar deposit. Des Cloizeaux (1846-47), who had in fact an illustrious career in mineralogy ahead of him, visited the site again in the following year. He took a quantity of crystals home with him on both occasions (see e.g. Senarmont 1845).

15 More on anisotropy and double refraction, from 1845

15.1 Anisotropic magnetization, 1845-55

Michael Faraday, who was active in research from about 1815 to 1860, was one of Britain's most productive scientists. His interests to begin with were mostly in the field of chemistry, where he made many discoveries, but his greatest work is the series of articles "Experimental Researches in Electricity". He discovered electromagnetic induction in 1831, and among other things he had a large electromagnet in his laboratory. With this magnet, he discovered diamagnetism and investigated it thoroughly in 1845-49. In an inhomogeneous magnetic field, an object made from a diamagnetic material experiences a weak force pushing it out of the field. Elongated objects suspended in a homogeneous magnetic field experience a torque tending to align them at right angles to the field.

These findings by Faraday regarding magnetic fields generated much discussion and thinking about the nature of matter, although probably less than his results to be described in section 16.1. Many others invested in electromagnets for experimentation, discussing their results with Faraday. One of these was J. Plücker, who investigated the forces exerted by magnetic fields on various crystals, including Iceland spar. He was surprised to find (Plücker 1847) that the spar and other diamagnetic crystalline materials were reluctant to behave according to Faraday's principles. His main conclusion was: "When one puts a uniaxial crystal between the poles of a magnet, the optical axis is repelled by both of the poles". In modern terms, this means that a torque tends to turn the axis into a direction at right angles to the field. If a crystal specimen is not very elongated in a direction perpendicular to the optical axis, this torque may dominate the torque caused to its shape. Plücker checked his results specifically with calcite, concluding (Plücker and Beer 1850) that "Iceland spar shows a very definite axial effect".

Faraday had himself been studying this odd behavior, for instance in bismuth which shows the greatest diamagnetic effect (per mass) of all materials, and he called it "magne-crystalline action" (Faraday 1848). J. Tyndall (1894) who worked with Faraday in his experiments and became his successor, says in his biographical notes on Faraday: "An eminent example of magne-crystalline action adduced by Plücker, and experimented on by Faraday, was Iceland spar". In a discussion of Plücker's and his own results, Faraday writes in 1848 (Proc. Royal Soc. 5, p. 783): "How rapidly the knowledge of molecular forces grows upon us, and how strikingly every investigation tends to develop more and more their importance and their extreme attraction as an object of study ! A few years ago magnetism was an occult power affecting only a few bodies; now it is found to influence all bodies, and to possess the most intimate relations with electricity, heat, chemical action, light, crystallization, and through it, the forces concerned in cohesion.".

Around 1848, Tyndall went to Germany and continued magnetic measurements on crystals for three years, first with H. Knoblauch in Marburg (e.g. Tyndall and Knoblauch 1850) and then in Berlin. From his measurements, he computed the ratio of susceptibilities of Iceland spar for its magnetization parallel and perpendicular to the optical axis. He and Knoblauch questioned some generalizations by Plücker regarding correlations between magnetic and optical properties of crystals. Variable results from their measurements may partly have been caused by the presence of ferromagnetic impurities, whose reactions to magnetic fields can easily mask the weak diamagnetic forces on the bulk substance of a crystal (Plücker and Beer 1850).

Faraday had in 1838 also attempted to measure whether cubes of Iceland spar where the optical axis was parallel to one of the three sets of edges, had different susceptibilities for electric fields directed along these. His results were not conclusive. W. Thomson who had previously written various papers on mechanics, electricity and thermodynamics, began contemplating how materials respond to electrical and magnetic fields, after he heard of Faraday's discovery (section 16.1) regarding the effects of magnetic fields on light (Thomson 1847). One of the resulting publications (Thomson 1851) included theoretical equations describing the anisotropic magnetic properties of crystals. This work was based on a draft theory proposed by S.D. Poisson around 1824. Here, Thomson suggests experiments to test the theoretical predictions, by measuring forces and torques acting on spheres from calcite and other materials in magnetic fields. Tyndall (1851, Fig. 11-1, 1855, 1856) and Faraday (1856) accordingly made magnetic measurements on spheres, cubes and cylinders from Iceland spar, bismuth and other anisotropic substances such as wood. Referring to these and perhaps also to subsequent studies (see sections 15.2 and 28.3), Tutton (1922, p. 1402) states that "Measurements carried out with spheres of calcite agreed perfectly with Thomson's theory".

15.2 Theories on the behavior of crystals in fields – general importance

From the few relevant papers from the mid-nineteenth century that I have read, it seems likely that interest in the geometrical and physical properties of Iceland spar and other crystals promoted the development of some very important areas within physical science. These properties were also connected with mechanical concepts like moments of inertia, and novel ideas in mathematics (quadratic forms, matrices). In physics the term "field" means that to any point of space you can tie a value of some defined quantity with its associated units of measurement. This quantity can often be represented by a single number, for instance when it represents density or temperature. These are called scalar fields. However, there are also many vector fields where the quantity has both a magnitude and a direction, such as the velocity of a fluid, an electrical field, or a flow of heat. In materials, one vector field often causes another one, and the microscopic version of Ohm's law is a good example of this. It says that an electrical field at a point in a material gives rise to a current density in the at that point, and the constant ratio between these is the electrical conductivity of the material. In this simple case, the latter (resulting) field is always parallel to the former (causative) field, and the ratio is not dependent on direction.

Around 1850 it was becoming apparent that in crystals (other than those of the cubical system), material coefficients such as the conductivity just mentioned depended on the direction relative to the crystal axes. Thus Senarmont (1847b, 1848) showed that this applied to thermal conductivity, by heating wax-coated samples of Iceland spar (from Des Cloizeaux' 1845 visit to Iceland). His results prompted G.G. Stokes (1851b) to write a notable theoretical paper about crystalline thermal conduction in general. C. Neumann (1864) later dealt with the theory of heat conduction in biaxial crystals. Observations on the electrical conduction of various minerals (Senarmont 1849,

and others such as C. Matteucci shortly after) and of their electrical susceptibility (Wiedemann 1849, Knoblauch 1851, both including Doppelspath) also showed that these properties were direction-dependent. The same applied to the hardness of Iceland spar and other crystals (Franz 1850, cf. results from later work in Fig. 28-1), and their thermal radiation (Ångström 1853). Grailich and Pekárek (1854) constructed an instrument which they called a sklerometer, using it to carry out detailed hardness measurements on for instance surfaces of "several pieces of the Icelandic spar". The hardness could even vary with the direction by a factor of three, which was considered quite remarkable cf. a description of their experiments in E. Mallard's book on crystallography published three decades later.

The preceding section mentioned W. Thomson's theory on the anisotropic behavior of magnetism in crystals, and its confirmation by experiment. The entry on J.C. Maxwell in Dictionary of Scientific Biography (1980-90) refers to his lengthy correspondence with Thomson on these matters. Maxwell then put forward in 1856 some ideas about two different magnetic vectors H and B in materials, where B is caused by H in the same way as a pressure gradient causes fluid flow in a porous material, or a temperature gradient causes flow of heat. This very much strengthened the foundations of Thomson's theories regarding Faraday's magne-crystalline action. Liebisch (1891, p. 119-120) claims that Thomson and Maxwell were the prime discoverers of the close mathematical analogies between the various vector fields in crystal physics. Other sources also consider Maxwell's writings also to have opened the eyes of many physicists to the importance of vector fields. It might be interesting to trace that story in more detail.

15.3 More on the effects of pressure on optical properties of materials, to 1880

While J.C. Maxwell was still a schoolboy in Edinburgh in 1847, his uncle took him to meet W. Nicol who showed him his composite Iceland spar prisms (section 12.2), see Dictionary of Scientific Biography (1980-90). Nicol later gave him a pair of such prisms. Maxwell constructed a viewer with glass mirrors (Fig. 13-7) and made with it some remarkable experiments concerning the effects of pressure on refraction in a gelatinous substance. In a large paper by Maxwell (1853) on these observations, important principles regarding the pressure effects were put forward, and (according to D.S.B.) his theoretical interpretation was an improvement upon that of Neumann (1843) quoted in chapter 11 above. Maxwell's biographers point out that here he acquired valuable experience of material physics, and also of polarized light cf. his 1856 paper mentioned in section 13.2. More on him later, but in the next several years his research interests were in other fields such as mathematics and thermodynamics.

The most notable experimental study on double refraction in transparent isotropic materials in the mid-nineteenth century was carried out by G. Wertheim (1851, 1854). He showed that the optical effects of compression and dilatation are of equal magnitude and approximately proportional to the deformation of the material. He also investigated how these effects depended on the wavelength of the light. No major steps forward in this field occurred before 1880, but A. Bravais (1855), A. Kundt, E. Mach and others (cited by Verdet-Exner 1887 and by Kerr 1888, 1894) made some contributions to it. See section 27.3 for important practical applications of this technique in the early 20th century.

16 Two major discoveries involving optical activity, 1845-50

16.1 Faraday: Magnetic effects on light

In another very important research project by Faraday (Fig. 16-1) in 1845, Iceland spar also played an even more significant role than in the anisotropy studies discussed above. Among other things he was looking for some relations between magnetic fields and light, and for this purpose he sent rays of linearly polarized light (from a glass mirror) through various materials including a piece of dense lead-glass which he had prepared for use in other experiments (see Tyndall 1894, chapters 10 and 11). If the glass was in a strong magnetic field parallel to the light beam (see Fig. 16-1) he saw with the aid of a Nicol prism that the direction of the plane of polarization rotated. Faraday (1845) soon found similar effects occurring in other solids and liquids. They turned out to be proportional to the intensity of the field and to the distance covered in the magnetized material. This Faraday effect, as it is now known, was the first indication that magnetic fields had something to do with light. It generated immediate interest among physicists: some attempted to explain the effect with models of the structure of matter, while others set up experiments to improve and extend Faraday's results. See for instance Biot (1846) who writes that the instrument maker Ruhmkorff designed new electromagnets for this purpose, Matteucci (1848), Bertin (1848) and Wiedemann (1851). Faraday and others attempted to measure the magneto-optical effect in Iceland spar; however, these attempts did not succeed until much later (see section 28.1). Faraday also looked for some effects of electrical fields on polarized light in materials, but such effects were not detected until 1875 (see chapter 23).

16.2 Pasteur 1848: Asymmetric crystals and molecules

As noted already, J.B. Biot was carrying out research on optical activity for much of the period from 1812 to 1860. His results and those of others had led to various hypotheses regarding the structure of matter and of molecules. Although Biot (e.g. 1844) repeatedly pointed out the potential of the method in experimental chemistry (cf. Vallery-Radot 1900), few chemists had yet followed his example when a young scientist, L. Pasteur (Fig. 16-1) became interested in studying the activity of tartaric acid and its salts. Biot (cf. section 12.4), F. de la Provostaye and E. Mitscherlich had written papers on certain puzzling aspects (noted in part already by Berzelius) of these compounds. Further historical details on events leading up to Pasteur's project are presented by Mauskopf (1976). During his work, Pasteur (1848, 1850, 1851, 1853c) discovered that in aqueous solution, one of the crystal mirror images in Fig. 16-1 gave a left-handed rotation while the



MICHEL PARADAY

LOUIS PASTEUR

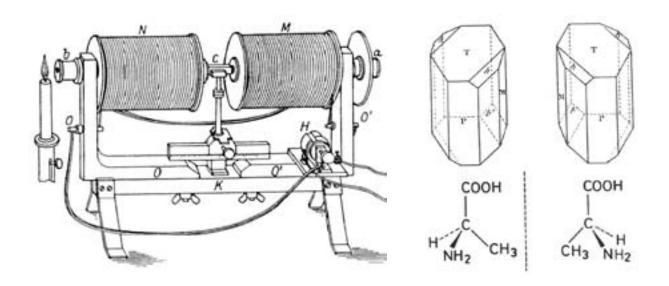


Figure 16-1. M. Faraday (1791-1867), L. Pasteur (1822-1895), and two of their important discoveries. Left: Equipment for studying the Faraday (1845) magneto-optic effect, i.e. rotation of the plane of polarization of light in materials placed in a magnetic field. The drawing shows a reconstruction of Faraday's experiment by Biot (1846) and is taken here from Handbuch der Physik, vol. 16/I, 1936. Light enters from the left through a fixed Nicol prism in b, a sample (glass rod, or tube containing liquid) at c, and another rotatable Nicol prism at a. Right: It was known that some compounds crystallized in two mirror-image modifications. Pasteur (1848) demonstrated that aqueous solutions of tartrate crystals (upper diagram) rotated the polarization of light respectively to the left and to the right. Subsequently, J.H. van't Hoff and J.A. le Bel suggested in 1874 that three-dimensional chemical bonds of carbon could form mirror-image molecules. In this case (lower diagram) it is the amino acid alanine. The broken line indicates a mirror.

other had a right-handed rotation. People soon realized the related fact that a similar mirror-image symmetry in quartz crystals corresponded directly to their having left-handed or right-handed optical activity. This had been noted earlier by Herschel (1822) and Weiss (1836) but not followed up. Many aspects of the phenomenon were still unexplained, for instance why quartz should be the only known optically active crystal, its activity even disappearing both upon melting and upon dissolving in strong alkali.

Pasteur clarified various points regarding the crystallization of tartaric acid and other compounds of this type (cf. Mason 1982, section 1.3). If for instance an aqueous solution contains an inactive (racemic) mixture of left-handed and right-handed molecules, it is sometimes possible to achieve their precipitation in such a way that some of the resultant crystals are right-handed and others left-handed. His results caused much stir among chemists, and many started investigating the optical activity of crystals (section 18.2) and organic liquids with polarimeters. Already by 1860, textbooks of chemistry were covering the phenomenon, and the coming decades saw the publication of hundreds of papers on the behavior of optical activity in many compounds under varying conditions.

Another method discovered by Pasteur (1853a) for separating left-handed and right-handed molecules of the same compound in solution, was to let it combine with another purely right-handed or left-handed compound. The left-right molecules of such a combination often have different propertes, such as solubility, than right-right and left-left molecules do. After isolation of one type, the original compound could be recovered from it. In the next few decades, this method was mostly applied to acids and bases. See more in section 27.4.

Pasteur continued his work on the optical activity of chemicals until around 1860, again making history when he was studying the effects of microorganisms on salts of tartaric acid. He found (Pasteur 1858, 1860) that they were able to distinguish between, and separate, left-handed and right-handed versions of the same compound. This was a fundamental discovery in biology. It greatly accelerated research on this aspect of organic materials, and gave rise to much discussion for instance about the origin of life on the Earth. In the case of Pasteur himself, this discovery in turn led to (Vallery-Radot 1900, p. 73-74) the research on fermentation and later on germs for which he is most famous. It may be mentioned here that some consider Pasteur's ideas about those matters to have been partially based on experimental results published by A. Béchamp. Béchamp (e.g. 1855) had studied with a polarimeter and other apparatus, how sugar solutions containing various salts changed with time due to moulds.

The processes involved in fermentation were at this time a long way from being understood. Pasteur for instance held the opinion that a certain acid which is formed during the fermentation of sucrose by yeast, took part in the task of splitting its molecules into the two monosaccharides. However, Berthelot (1860) demonstrated by polarimetric measurements and other means that the yeast managed this without the aid of the acid.

J.H. van't Hoff (1874) and J.A. Le Bel (1874) independently suggested a general explanation of Pasteur's (1848) discovery of the connection between the optical activity of organic carbon compounds in solution and the mirror-image symmetry of their crystals. The ideas of van't Hoff partly originated when reading recent papers on lactic acid. J. Wislicenus (e.g. 1873) had carried out research on this acid which is optically active when derived from animal tissues, whereas an apparently identical species derived from sour milk is inactive. Wislicenus considered it probable that the acid molecules could have two different shapes. The hypothesis of van't Hoff and Le Bel is based on the view that the four valency bonds of the carbon atom are not in the same plane as previously thought, but are three-dimensional. Therefore, a "handedness" appears when a carbon atom is connected to four different atoms or atomic groups. In their original arguments, optical activity played a larger role in Le Bel's writings than in those of van't Hoff, who however soon

recognized its value in this context (Hoff 1877). Their hypotheses, at first opposed or ignored by some but supported vigorously by Wislicenus (1888), turned out to be correct. They became the foundation of stereochemistry, a field of chemistry which deals specifically with three-dimensional aspects of molecules and their interactions. According to Lowry (1913a, p. 545): "...no firmer basis for theories of molecular structure has yet been found than that which rests upon the use of the polarimeter to detect rotatory polarization.". This discovery also improved understanding of the question to which crystal classes optically active compounds belong (see e.g. Becke 1889).

17 Development of physical optics, 1840-80

17.1 Hypotheses and their experimental verification

On the continent of Europe, much discussion took place among theoreticians on the propagation of light in doubly refracting materials, reflection from these, and total internal reflection (Broch 1842, Senarmont 1843, 1856, Cauchy 1850b, Lang 1861, Stefan 1864c, Cornu 1867, Briot 1867, Lommel 1878 and others). I have not checked in any detail whether these were just reviews of older papers or if new ideas were being presented. At least though, Dictionary of Scientific Biography (1980-90) considers the theoretical research of Briot in optics to have led to progress in elliptical functions and other areas of mathematics. Cornu (1872, 1874a) presented a general derivation of the path of a light ray through a prism of doubly refracting material, and tested it in detail on Iceland spar. Ohm (1855) explained the interference of light in plates of uniaxial crystals.

Cauchy (1839b, and others) investigated theoretically and by comparison with experiments, the polarization of light reflected from metal surfaces. In section 13.2 it was mentioned that J. Jamin (e.g. 1847) carried out detailed measurements in such cases. Formulas deduced by Cauchy in 1849 relating the polarization to properties of the metal, have been "of great value in the technical field" according to R.W. Pohl's textbook: Optik und Atomphysik, 1954. This also applies to papers by Beer (1854) and Quincke (1863) on the effect of metallic reflection on polarized light.

In Britain, much theoretical work in optics was also going on. Among papers on light propagation in crystals in 1840-50 we may mention those by Green (1842b), Kelland (1844), Stokes (1846, see Fig. 10-1) and Challis (1847), later for instance Strutt (1871c). The papers by Stokes and Challis are connected to what has been called the golden age of theoretical physics research at Cambridge University (see Timoshenko 1953): this research involved among other things various aspects of optics and elastic waves in matter. Other works by Stokes on optics include an important mathematical description of the properties of polarized light (Stokes 1852a); "Stokes parameters" which were introduced in this paper, are still being used.

For the paragraph below, a digression is needed to explain the concept of a diffraction grating. These are made by scratching a very large number of closely spaced parallel lines on plates of glass or metal. For instance, Lloyd (1857) mentions a grating with 30,000 lines per inch. Part of a light ray hitting the grating is deflected by an angle whose magnitude is a simple but accurate function of the wavelength of the light and the line spacing. By means of gratings, a spectrum can be spread out a lot more than is possible with a prism, but the intensity of the light in each wavelength interval is low.

In 1840-80, the discussion on the properties of the luminiferous aether continued, especially on whether its oscillatory movement took place in the plane of polarization of light or at right angles

to it. Among the many papers contributing to this discussion are those by Schmid (1842), Cauchy (1849, 1850a), Ettingshausen (1856), Bartlett (1859), Jamin (1860), Lorenz (1860 and later), Fizeau (1861), Quincke (1862 and later), Lippich (1863), Mascart (1866), Boussinesq (1867), and Ketteler (1877). Rankine (1851, 1853), Haidinger (1855a), Stokes (1859) and Verdet (1865) supported the views of Fresnel, and they also considered that his hypothesis regarding a material-dependent density could be extended to crystals having a direction-dependent density of the aether. Stokes and others contemplated into the 1880s whether measurements of the polarization state of light that has passed an obstacle (such as a diffraction grating) could resolve the question of the vibration direction of the aether. However, Poincaré (1892) and Winkelmann (1906, p. 1110-13) claim that the question was never definitely resolved. In a large paper reviewing the state of research on double refraction, Stokes (1862) suggested a new and precise method for testing this matter, see the following section. Another historical review of optical theories is by Saint-Venant (1872).

The many facets of polarization and double refraction had by 1850 become a sizable part of the contents of texts on physical optics (or wave optics, as distinct from geometrical optics). Among such books were those by Lloyd (1857) and larger works by Billet (1858-59) and Verdet (1869), Fig. 17-1. University-level textbooks on physics dealt with these topics at some length (e.g. Baumgartner 1845, Christie 1864-65), and they were explained to the literate public in general science journals (e.g. Spottiswoode 1874b) as well as in booklets (Woodward 1851) and monographs (Dove 1853, 1854 edition of Pereira 1846). Among polarization phenomena presented in student experiments and demonstrations, were Brewster's angle, the colors of thin crystalline plates, optical activity, and interference in crystal plates in convergent light. See for instance the advertisements by Griffin (1855) and Steeg (1857), a paper by Billet (1862), and catalogs from Lerebours & Secretan (1853) and Soleil (1867) which offer crystals of Iceland spar and other minerals, Nicol prisms, and related educational apparatus. Several French firms displayed such apparatus as well as research instruments at the World Exhibition in Paris (Österreichische Central-Comité 1867). The instrument-maker firms Franz Schmidt & Haensch and R. Fuess that were established in 1864 and 1865 respectively, later grew into major suppliers of optical instruments containing Nicol prisms.

17.2 Experiments on Iceland spar

In previous sections it has been mentioned that certain consequences of Huygens' hypothesis of an ellipsoidal shape of the extraordinary ray in uniaxial crystals like Iceland spar were tested by Hauy (1788), then with greater accuracy by Wollaston (1802b) and then with still greater accuracy by Malus around 1809. In the decades to follow, Fresnel and others extended the hypothesis of Huygens cf. section 10.1, and experimental techniques in optics were improved. The time eventually came for renewed testing of Huygens' construction in crystal specimens. Once more Iceland spar tended to be chosen because of its strong double refraction, homogeneity and other desirable properties. This was indeed a good choice, in view of the much later conclusion from X-ray research that Iceland spar was among the most perfect crystals found in nature (Compton 1916).

Billet (1859) describes a method due to F. Bernard, where the lateral shift of a ray travelling through a plate of Iceland spar was measured for different angles of incidence, giving good agreement with the theory. Desains (1862) proposed a similar method for the measurement of refraction in crystal plates, and Lang (1858) presented another one using specially cut prisms, but I have not discovered whether such measurements were actually performed. Senarmont (1847a) and Cornu (1866, 1867) investigated the reflection of polarized light from the surface of calcite crystals, find-

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LONDON: LOTONAN, BROWN, GEREN, LOTONANS, AND DORESTS. 1857

Figure 17-1. Iceland spar greatly advanced progress in physical optics. This is seen for instance in 19th century textbooks and monographs, both on the subject in general and on specific topics such as wave theory and crystal optics. The two books at the top are written around the time when A. Bertin stated (in Rev. Sci. 4, p. 136, 1867) that "the science of light is totally French". These works were published in two volumes each; if sections connected with Iceland spar and the polarization of light are removed, one volume would would suffice in either case. The lower two books are a reminder of the many contributions to optics by Irishmen in the English-speaking countries and of Austrians in German-speaking regions.

ing that the plane of polarization rotated by up to a few degrees, as predicted by theory. Stokes (1872) measured refraction in Iceland spar prisms with a method previously devised by himself, concluding that the hypothesis of an anisotropic density of the aether was untenable. Abria (1874, 1875) carried out a series of painstaking measurements on refraction and total reflection in Iceland spar. Accurate measurements on the refraction of the ordinary ray in Iceland spar were also made (Swan 1849), in order to test a suggestion by J. MacCullagh that it might be dependent on direction (cf. Brewster 1843).

Grailich (1853, 1856) made an extensive theoretical study of the refraction and reflection of light at a twinning plane in a uniaxial crystal, with computation of numerical results for Doppel-spath and a description of samples which he inspected. It may have caught his attention, that when one looks at a white light through an Iceland spar crystal containing such a plane, the white light is seen in the middle with a colorful image on each side. Some other crystals such as potassium chlorate also show peculiar color effects (iridescence) due to multiple twinning. These phenomena were investigated more fully by others (cf. Pockels 1906, p. 203-208), including later the Rayleigh father and son, and C.V. Raman.

The hypotheses by Huygens and Fresnel were not tested to any great extent on minerals other than Iceland spar in the period covered here. I have seen observations by Jamin (1850b) on the refractive indices of quartz, later improved by Lang (1870). Abria (1874, 1875) measured angles of refraction and reflection in quartz. G. Kirchhhoff in 1859 (see Stokes 1862, p. 272) and Glazebrook (1878) similarly measured the biaxial mineral aragonite, and Cornu (1874a, p. 43-46) made a limited study of sodium nitrate. Nicol prisms were an essential tool in much of the above work. See more in section 28.2.

17.3 On the radiation of light

P. Desains, H. Knoblauch (see section 10.5) and others continued research on infrared radiation in 1850-70. Among other things it was found (Provostaye and Desains 1851) that the light emitted by the surfaces of hot liquid and solid objects was to some extent polarized. The polarization increased with angular distance from the normal to a surface. Kirchhoff (1860) pointed out that this result was a natural consequence of a law recently formulated by him (Kirchhoff 1859): "...for radiation of the same wavelength at the same temperature, the emissivity [of a surface] equals the absorptivity". If light from the outside was reflected to a variable degree from the surface of a body depending on its polarization and its angle of incidence, the same would apply to light coming from the inside. This was supported by qualitative experiments by Magnus (1866), but Arago (1852, 1858c) had in fact carried out similar observations before 1824 ! Arago's results on the polarization of thermal radiation had (according to Barnard 1863) helped to settle arguments as to whether the Sun's light-emitting surface was a solid, liquid or a gas. Since the light received neartangentially from hot solids and liquids was partially polarized while that from a hot gas or a flame or the Sun was not, it was evident that the Sun was gaseous. Studies of the polarization of oblique heat radiation were resumed by e.g. Violle (1887), and by Millikan (1895) who also made similar observations on fluorescence light. Both used a "photopolarimeter" invented by Cornu (1882).

Provostaye and Desains (1854) made quantitative measurements on the radiation emitted from hot objects, finding e.g. that blackened metal surfaces emitted much more light than polished surfaces. They employed a photometer with a Nicol prism and an Iceland spar rhomb (see section 18.4). Although others had no doubt made similar observations with thermometers, this result was quite important and agreed with Kirchhoff's (1859) law. Cotton (1899) says in a review of this law: "A relation between the absorption and the emission of what is still called "radiant heat"

had long been known, and de la Provostaye and Desains had verified it with great care". Among other aspects of radiation from hot objects considered by Kirchhoff, was the question whether the surface of a dichroic material would preferentially emit (along a normal) light polarized in the direction which is absorbed (when incident on the material). He placed a sample of tourmaline in an oven to test this, and the result agreed with his expectations but was rather inaccurate. Confirmation was however soon provided by Stewart (1860), and later by others. This may have been one stage in Kirchhoff's path towards the very important concept of "black-body radiation" which he proposed in 1862.

E. Becquerel (1863, 1867 p. 61-97) used a thermocouple, a photometer with Nicol prisms, and other apparatus in research on light emitted from hot surfaces. He also quotes measurements by F. Zöllner (1860) on glowing filaments using a photometer with a glass mirror and one Nicol prism. Becquerel concluded that the intensity of emitted light increased as a particular exponential function of temperature. In a paper in the Physical Review, vol. 13, 1901, E.L. Nichols writes about Becquerel's observations: "…his work is especially noteworthy in that he employed many of the methods to which, in the hands of later investigators, our knowledge of the laws of incandescence is due. He established the fact of the direct proportionality of the logarithm of the intensity of radiation to the temperature and pointed out the possibility of optical pyrometry".

The ability of various inorganic and biological substances to emit light without being heated, presented a puzzle to science for centuries. Depending on conditions, the phenomenon had names like luminescence, fluorescence or phosphorescence. Calcite was among these materials, giving off its own light (often orange in color) if illuminated. The first observations of the luminescence of Iceland spar by a scientist that I am aware of, were made by Dessaignes (1809). From the 1850's onwards, G.G. Stokes and others carried out comprehensive studies on luminescence, including its polarization. They (e.g. Stokes 1852b) found that the wavelength of light that was excited by other (monochromatic) light, always had a longer wavelength. This rule could not be explained before the advent of A. Einstein's quantum theory in 1905. In his research, Stokes found that quartz transmitted ultraviolet light better than other materials, and Miller (1862) noted that Iceland spar also performed quite well in that respect. See section 29.8 on applications of that property. E. Becquerel (1857, 1861, 1867) investigated luminescence for a long time (see section 18.4), Iceland spar being one of the materials that he gave special attention. Among other things, he showed with the aid of his Nicol-prism photometer, that the intensity of radiation given off by a luminescent sample is proportional to that of the exciting light.

18 Developments in instrumentation 1850-70

In the decades 1850-70, various new types of polarizing prisms from Iceland spar were described, for instance by Foucault (1857), Potter (1857), Jellett (1858-61), Hasert (1861), Dove (1864, beam-splitting prism), Hartnack and Prazmowski (1866), and Jamin (1869). Senarmont's (1857, top of Fig. 12-1) prism which resembles those of Rochon and Wollaston, is still in production. This chapter will present developments within three main classes of optical instruments in which Iceland-spar prisms were a key component, namely polarimeters, microscopes and photometers. In addition, Iceland spar played a role in some other types of equipment. One example is the sensitive interferometer with two spar plates invented by Jamin (1868). This device could among other things detect very small variations in the refractive indices of materials, but it was probably more difficult to use than Jamin's popular glass-plate interferometer of earlier date. See section 39.5 for much later applications of this concept.

18.1 On new polarimeters and research conducted with them, 1850-70

Among those writing about improvements in polarimeters were Senarmont (1850), Stokes (1851a), and Pohl (1856). Some of the improvements had lasting value, such as Bravais' (1855) bi-quartz plate which made it possible to use a half-shade technique in measurements on elliptically polarized light. In an advertisement by the workshop of Lerebours and Secretan (1853) which includes various apparatus containing Nicol prisms, the firm is said to have produced one hundred Soleil polarimeters for the Ministry of Finance.

A new type of polarimeters by Listing (1855), Wild (1863, 1864, 1870) and Bakhuyzen (1872) was among other things intended for measurements of glucose in diabetic urine. In this instrument a Savart plate from Iceland spar or quartz produces many parallel fringes seen by the observer, and they disappear at the right setting (Fig. 18-1). The Wild polarimeter, as it was generally called, was produced commercially from about 1867 (Österreichische Central-Comité 1867, p. 121). Others also tested urine with polarimeters, for instance Robiquet (1856) and Bence Jones (1861). The latter compared results obtained polarimetrically and by chemical analyses, and in addition he concluded that healthy people excrete a minor amount of glucose. This point was debated until the 1930s, because of disturbing influences on all the numerous methods developed for this purpose, but it was finally established that there is no glucose in normal urine.

Polarimeters where the concentration of sugar was read directly off a scale, were called saccharimeters. They were manufactured for decades, with many improvements. In these measurements it is possible to use a circular scale as in ordinary polarimeters. However, it was probably more common to keep the analyzing Nicol prism in a fixed position, letting a quartz plate (with its opti-

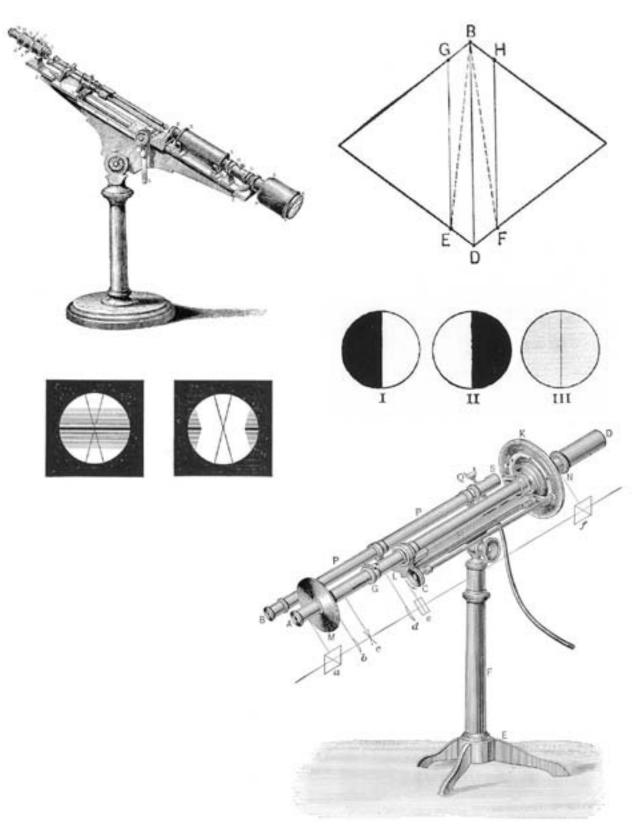


Figure 18-1. Upper part: The polarimeter of Jellett (1861-64, 1875), containing a special polarizing prism. This prism was constructed by removing the wedge BEDF from an Iceland spar rhomb and gluing the pieces together. An experimenter looking from the left saw a divided circular area, and he rotated the analyzing nicol until its two halves were equally bright as in III on the left. such "half-shade" techniques gradually became dominant both in polarimeters and photometers. Lower part: Instead of the half-shade, the polarimeter of Wild (1864, 1870) as well as his photometer employed interference of light caused by a Savart plate. The plate is marked e in the diagram, taken from Browne (1912). The observer rotates the wheel C until the stripes disappear, and then reads a scale with the small telescope B.

cal axis at right angles to the plate) rotate the plane of polarization in an opposite direction to the rotation within the fluid. The plate was in fact made of two wedges of respectively left-handed and right-handed quartz, which were moved against each other. Rolfe (1905, p. 33) states that "About 1850, Duboscq was the first to make a practical quartz-wedge saccharimeter". Here, he may be referring to the previously mentioned paper by Clerget (1849) or to Duboscq and Soleil (1850). An important advantage of the quartz wedge was that a source of white light could be used, instead of the more bothersome sodium flame. The optical activity of quartz had to be known precisely; one determination at six wavelengths was carried out by O.J. Broch (1846), employing a method like that of Fizeau and Foucault (1845) from Fig. 13-2. Broch's calibration was improved by Stefan (1864a) and by Soret and Sarasin (1875, 1882). Empirical formulas for the variation of the rotation angle of quartz with wavelength (i.e. its rotatory dispersion) were proposed by Boltzmann (1874) and others.

As already mentioned, the accuracy of polarimeter readings could be increased significantly by providing a split (half-shade) field of view. The instrument designed by Jellett (1861-64, Fig. 18-1, top) was the first one where this was accomplished with a specially cut double Iceland spar prism, instead of Soleil's biquartz plate. The Jellett prism (Fig. 18-1) soon became popular and was used for decades (Schulz and Gleichen 1919, p. 66; Struers 1925; Kessler 1926, p. 1403-1404) even though it was difficult to make (Salomons 1893, p. 214) and faced competition from other precise half-shade arrangements.

Many applications for polarimeters were developed in Europe during the 1850s (Pohl 1856, Buignet 1861), and they no doubt aided in the resurgence of the beet-sugar industry that took place there from around 1840. Thus, the polarimeters took part in the isolation of new sugars, e.g. by Pelouze (1852, sorbose) and by Berthelot (1859, trehalose), of organic acids (e.g. Wurtz 1857) and other optically active chemicals. They also were involved in revealing that various essential oils derived from plant constituents such as flowers, fruits, seeds, and pine needles were composed of two or more hydrocarbons (e.g. Jeanjean 1856). The same applied to turpentine (Berthelot 1853), and turpentine as well as camphor from different tree species turned out to have different optical properties (e.g. Chautard 1863). Here one may also mention again A.P. Dubrunfaut who was working for more than half a century to promote the sugar and fermentation industries in France, earning the title of "the father of beet sugar" from his contemporaries. He (Dubrunfaut 1869) demonstrated with a polarimeter, that if raw sugar on its way to refineries is contaminated by impurities, microorganisms will ferment part of it and cause a deterioration in quality. In the U.S., some people had taken notice of these new instruments around 1850 (for instance Riddell 1853, see Warner 2007) but their use did not become widespread there until after 1880. More on research with polarimeters in the sugar industry and on related fronts in section 22.2.

There is no doubt that polarimeters played a part in many discoveries in chemistry, medicine and other fields in the latter part of the 19th century. One example is the thorough investigation by L. Wilhelmy (1850) on the rate of the "inversion" reaction occurring when acid is added to a sugar solution. He measured the sugar concentration with a polarimeter from the Soleil workshop. According to modern sources, this study was an important milestone in the origin of chemical kinetics. Rolfe (1905, p. 251) says that being the first to propose a mathematical model of this chemical reaction, Wilhelmy "…opened up a field of research which has had enormous influence on the science of chemistry". Some historical accounts (e.g. Heys 1952, p. 222-225) state that Wilhelmy's results were used as an argument in favor of the "law of mass action" by its originators, P. Guldberg and C.M. Waage in 1863-67. However, I have not seen a direct reference to Wilhelmy's work in those papers by Guldberg and Waage (e.g. 1864) that I have consulted.

J.H. Jellett (1875) who was mentioned above, applied his polarimeter to a series of investigations on chemical equilibria and on the properties of sugars and alkaloids (more on them at the end

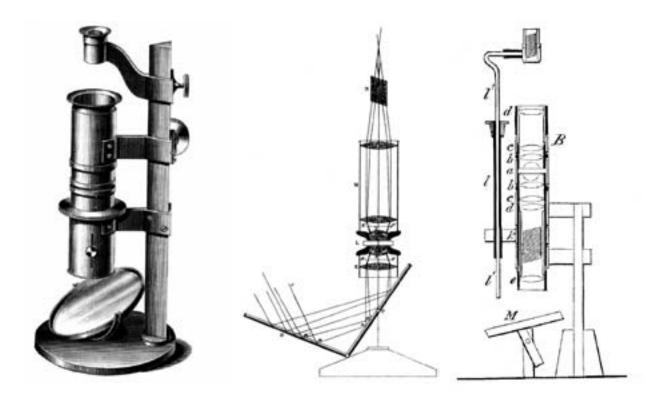


Figure 18-2. Three polarizing microscopes for studying thin crystal plates. Left: J. Nörrenberg's apparatus from 1858 (cf. Bertin 1863). A glass mirror acts as polarizer while the Nicol prism N is the analyzer. A. Des Cloizeaux employed a somewhat similar device in his extensive work on minerals. Center: A Nörrenberg microscope as improved by V. v. Lang (1871). The illustration is taken from a catalog issued by the equipment supplier A. Stendicke some time after 1890. There are Nicol prisms inside both the main tube (which has a slot for insertion of specimens) and the small ocular tube at the top. Right: The microscope described by Grailich (1858) which is similar to that of Lang.

of this section). It seems that his results did not attract much attention at the time, but in a preface to the German edition of Jellett's paper in 1908, W. Nernst points out that they contain some notable extensions to those of Guldberg and Waage. Voit (1867; also F. Hoppe-Seyler around that time) studied the diffusion of sugar in an aqueous solution; it may be of interest that A. Einstein made use of data from further research by others on this phenomenon in his doctoral thesis published in 1905. With respect to the chemistry of sugars, the major steps forward were taken in E. Fischer's work which began in the late 1880s, see section 27.4.

During the time interval being considered here, chemists finally began sharing J.B. Biot's conviction that research on optical activity might provide important information on molecular structure. Krecke (1872) remarks in a review article, that "…hardly any branch of science is better equipped than the measurement of optical rotations, to guide chemists in their research on the structure of molecules". One of those who became interested in optical activity, was Arndtsen (1858) who for instance studied the anomalous rotatory dispersion of tartaric acid and suggested an explanation of it (see section 12.4). This matter will be treated more fully in section 22.3.

Becquerel (1849) and Hoppe (1857) describe how they made use of polarimeters in measurements of albumin in serum, milk, urine and other bodily fluids when diagnosing illness. A. Béchamp completed a doctoral thesis in medicine 1856 on albumin and related proteins, and according to the Dictionary of Scientific Biography (1980-90) he employed a polarimeter to discover various properties of these that had not been resolved by older methods. He later published valuable papers on these proteins, again with polarimetric observations playing an important role (e.g. Béchamp 1873, 1884). However, these methods do not seem to have been used much in medical or physiological work on proteins after 1880. A. Bouchardat who appeared in section 13.2 above, wrote among other things about optical activity in alkaloids (Biot 1843b; Bouchardat 1843, Bouchardat and Boudet 1853), a class of substances which have through the ages had various functions as medicines, addictive drugs, or poisons. They include quinine and many related compounds, atropine, codeine, strychnine, and morphine. Pasteur (1853b), Buignet (1861) and others also investigated the optical activity of alkaloids.

18.2 Microscopes, Nicol prisms and thin sections of rocks 1850-70

Turner (1989) describes many old microscopes preserved in a London museum, including some from the mid-19th century with fixed or detachable Nicol prisms. Among them are four from the makers Powell & Lealand delivered in 1840-53, one from Smith & Beck around 1850, one from A. Ross around 1853 and one from Negretti & Zambra around 1855. Bryson (1850) also mentions microscopes with Nicol prisms. In France, Dujardin (1843) and Lerebours (1846) are familiar with microscopes fitted with Nicol prisms and other polarizing arrangements, and A. Nachet (1979) offers a microscope attachment with two Nicol prisms in his 1856 catalog. In the United States polarizing microscopes were advertised by J. & W. Grunow (1856). Books (Pritchard 1845, Quekett 1848, Hogg 1854) and papers (Legg 1849, Bird 1853) appearing in London in this time interval described the advantages of using polarized light for observations on a variety of crystals and biological materials. The technique seems to have been popular with amateurs who often were organized in clubs and published magazines, but I have not come across many papers by scientists in the U.K., France or the U.S. before 1855 on comprehensive studies involving polarization microscopy. In that year, another book (by H. Schacht, in Berlin) even asserts that "Polarizing outfits on microscopes are better suited for playing with beautiful color effects than for learning anything scientific".

Marbach (1854) used a polarizing microscope when he recorded for the first time optical activity in the cubic crystal system, namely in sodium chlorate. In another paper, Marbach (1855) discussed the presence of double refraction in cubic crystals and related peculiar properties of various minerals (in fact noted earlier by D. Brewster). These properties have since then been called "optical anomalies" (see e.g. Mallard 1876), and they were thought to be caused by phenomena such as twinning or internal tensions. Marbach did not describe his microscope or mention where it came from. However, it may be the same one as his teacher M.L. Frankenheim (1854, 1860) used in research on the growth and dissolution of crystals. Frankenheim's instrument had two Nicol prisms.

Kobell (1855) designed a microscope which he called a stauroscope, in order to identify minerals with the aid of the interference patterns seen in Fig. 7-2. It contained a glass-mirror polarizer and a tourmaline-plate analyzer. J. Nörrenberg demonstrated in 1858 his mineralogical microscope with a double glass-mirror polarizer and a Nicol-prism analyzer (Fig. 18-2), see Bertin (1863) and Groth (1926, p. 246).

A. Des Cloizeaux began studying plates of transparent minerals with Amici's microscope around 1855 (Des Cloizeaux 1855, 1857a) and with Nörrenberg's microscope around 1860, cf. his book on mineralogy (Des Cloizeaux 1862) and Bertin (1863). He investigated some 500 minerals (according to D.S.B.) and was the first of his countrymen to employ optical methods to any extent in mineralogical research (Lacroix 1893). See for instance Des Cloizeaux (1861) where a

microscope with at least one Nicol prism is described. Grailich (1858) wrote a book (Fig. 17-1) on his studies of many minerals and man-made crystals; these were carried out to begin with in Kobell's stauroscope and a microscope of Amici's type (Fig. 18-2), later in a much better Nörrenberg instrument for crystal plates with two Nicol prisms. Rath (1860) and Dove (1861) were also familiar with such microscopes containing Nicol prisms, but their use was not common among geologists before 1865; see literature quoted by Young (2003).

A comprehensive article by Des Cloizeaux (1864) contains illustrations of two microscopes, too small and fuzzy to be reproduced here. The polarizer is either glass plates (the method of Amici and Nörrenberg) or a Nicol prism, and the analyzer is a Nicol prism. He was the first to take photographs through such a microscope (see Des Cloizeaux 1855, and an obituary by A. Lacroix in Bull. Soc. Minéral. Fr. 20, 1897). Des Cloizeaux could measure the angle between the optical axes in biaxial minerals, and he (Des Cloizeaux 1866) also installed a heated stage in a microscope to test the effect of elevated temperatures on minerals. V. v. Lang similarly constructed a specialized instrument to measure axial angles in 1862, and in a paper on a new model of this instrument he says (Lang 1867) that the "…studies of the optical constants are becoming of more and more importance in the identification of minerals". Lang (1871) also improved Nörrenberg's microscope (Fig. 18-2).

Des Cloizeaux (1857b) found that the mineral cinnabar (HgS, trigonal) showed strong optical activity, and soon such behavior was noted in some other uniaxial crystals. This and Marbach's (1854) discovery of an active cubic crystal metioned above, generated new discussions on the symmetry of crystals (e.g. Lang 1869). More on this in section 22.3.

Kristjansson (2000) has reviewed some stages in the emergence of methods for the microscopic examination of thin sections of rocks. The main pioneers in this field were A. Oschatz and H.C. Sorby from the early 1850s (especially Sorby 1858), but F. Zirkel developed it further in the 1860s and drew attention to them in his textbooks (e.g. Zirkel 1866, 1873). In Zirkel's (1863) first paper on microscopic studies of thin sections, some of these were made from rocks which he had collected in Iceland. From the publications that I have consulted, it is clear that Zirkel and his collaborators had started using polarized light by 1864. For instance, H. Vogelsang (1867) states in paper containing descriptions and colored micrographs of some thin sections: "Excellent support is provided by the use of the polarizing microscope, not only in distinguishing between mineral species such as quartz and feldspar, or triclinic feldspar with its twinning from monoclinic, or leucite from rounded feldspar grains and so on. In particular, it allows you to make decisions about the nature of those constituent parts of the rock which we usually designate in somewhat loose terms as groundmass. We can also ascertain whether it is in a glassy, semi-crystallized or crystallized state.". Similar statements appear in Forbes (1867).

In addition to these quotes from Vogelsang, the advantages of mineralogical analysis by examination of thin sections (of standard thickness) in a polarizing microscope include:

-The method is convenient, and is non-destructive unlike chemical analysis.

-In polarized white light with crossed Nicol prisms, each mineral grain has characteristic colors which are not seen in ordinary light. From these a trained observer can estimate the difference between its refractive indices, a useful diagnostic tool for grain types of similar appearance.

-Twinning which is a very useful characteristic, is clearly seen, and also variations in composition within grains (zoning); both are indistinct or invisible in ordinary light.

-Measurements may be made on very small grains or parts of these, not readily accessible for analyzing by chemical methods.

One of those who was applying polarizing microscopes to the study of thin sections already by 1870, was G. Tschermak (1869, 1870) in his research on the composition of various common

minerals such as the plagioclase feldspars. These feldspars form a continuous range of isomorphous crystals from albite to anorthite, with calcium and aluminium gradually replacing sodium and silicon. His studies resolved long-standing series of erroneous notions about these feldspars, where even Icelandic rocks played a role under names like baulite, krablite and havnefjordite. Rath (1868) found a mineral where the chemical compound SiO_2 crystallizes in a different class from quartz, calling it tridymite. Subsequent industrial applications are mentioned in section 22.3. Maskelyne (1870) describes his observations on minerals in meteorites, where he employed a polarizing microscope. Kile (2003) recalls the large volume "Microscopical Petrography" by Zirkel (1876) which was part of a comprehensive survey of the geology of the United States along the fortieth parallel. It contained 44 chromolithographs of thin sections with detailed explanations. According to Kile, this work "established petrography as a fundamental science", and geological institutions in the U.S. soon began hiring scientists to carry out research in that field.

18.3 Polarizing microscopes in biological research

It may be mentioned here in passing, that W. Nicol published in the 1830s some observations on thin sections from wood (including fossilized material), however without employing the prisms he had invented. I have only gathered limited information on how biologists made use of polarized light in microscopy in 1850-70, but it was known by 1840 that for instance bone and muscle tissue, seashells, and starch particles exhibited polarization interference colors in a similar way as crystals did. Erlach (1847) describes a microscope with two Nicol prisms with which he observed various plant and animal tissues, and he published drawings of these. The microbiologist C.G. Ehrenberg (1848, 1849) refers to previous work by others and also lists many examples where in his own experience polarized light helps in the microscopic observation of biological specimens. It allows the observer to distinguish between organic and inorganic objects, as well as between crystallized and uncrystallized material. In those of his papers that I have seen, it is not stated what sort of polarizing apparatus he was employing. Mohl (1858, 1859) pointed out that Ehrenberg and other pioneers in the field did have various incorrect ideas, but he agreed that a polarizing microscope reveals features not seen in ordinary light, for instance in his studies of cell membranes. He found that crystals were present in much greater variety and numbers in the vegetable kingdom than previously known. In Mohl's opinion Nicol prisms are far superior to other means of polarizing light. See also White (1858), Brücke (1858) who published detailed observations of muscles from various animals accompanied by illustrations in color, and Klebs (1864) who investigated nerves in muscular tissue.

The well-known physiologist G. Valentin investigated in the period 1858-81 tissues from plants and animals, both with a Nörrenberg microscope and a similar-looking setup designed by H. Wild. See for instance Valentin (1858) on investigations of the crystalline lenses of the eyes of many animals. His book (Valentin 1861) contains a detailed account of the properties of polarized light and appropriate microscopic equipment, as well as short descriptions of many types of tissues. Nägeli (e.g. 1862) who studied plant cells in polarized light, considered starch grains, cellulose membranes and much other organic material to be composed of crystallized molecular groups which he called micelles, separated by thin films of water. "The crystalline nature of the micelles is especially obvious in polarized light" he states in an 1879 publication. Not everyone agreed with his interpretations (cf. D.S.B.) but according to Zsigmondy (1925, p. 39, see also Muralt and Edsall 1930, p. 352) Nägeli was far ahead of his time in his research on this state of organic matter. Harley (1865), a professor of physiology and histology, introduced to German readers a microscope which was already in wide use in England. Dareste (1871) described particles in egg yolk which in polarized light exhibited crystalline properties like starch. These particles were composed among other things of lecithin which later turned out to belong to so-called liquid crystals (section 27.2).

18.4 Iceland spar in measurements of light intensity, 1850-70 (with some prehistory)

Photometers are devices for the measurement of the intensity of light. The first photometers appeared already in the 18th century, cf. the chapter on P. Bouguer in D.S.B. Photometry was not a prominent branch of optics in the first half of the 19th century, but it has since then become an important field both in science and technology. Until around 1920, photometric measurements were always relative, an observer viewing two sources of light simultaneously and adjusting one of them until both appeared equal. One source was of unknown intensity and the other was supposed to be known; various methods were developed in order to attenuate either of them by a measurable factor.

One of the several methods employed in such attenuation, is based on a law proposed by Malus around 1810 (section 6.5) regarding a light beam that is reflected successively from two glass plates as in a Nörrenberg instrument (Fig. 10-3). Its intensity should change as cos²s where s is the azimuth angle between the beam's planes of incidence on the mirrors. Photometric apparatus of this type was tested by Arago (1835, 1841, 1858c) and later for instance by Jamin (1847), Babinet (1853, see Dahlander 1902), Wild (1856) and Crookes (1868) who in some cases added a Nicol prism or a beam-splitting prism. This method does not seem to have been used much subsequently, because of some practical problems. However, Wild (1888, 1890) described a new version of his photometer, and the workshops of Duboscq (1885) and Pellin (1899) advertise improved models of Babinet's instrument.

Arago (1833, 1835 and later, see Lloyd 1834, p. 384; Wild 1863) confirmed the other law named after Malus (see Fresnel 1822b) concerning a linearly polarized light ray that falls on an Iceland spar crystal. Then the intensity of one of the rays passing through the crystal is proportional to cos²a and that of the other one is proportional to sin²a, where a is the angle between the plane of incidence and the principal section of the crystal. In this way the intensity of a light beam could be changed continuously by known amounts. Arago (1833) used the method to estimate the intensity of light from the Moon and from the satellites of Jupiter. Talbot (1834b) also pointed out the possibility of making a Nicol-prism photometer.

One of those who later constructed such photometers was Beer (1851, 1852). In the latter paper he measured the attenuation of light in aqueous solutions of several compounds at varying concentrations. He found a simple law governing light transmission in such solutions. The fraction of the light intensity remaining after traversing a distance in the solution is a negative exponential function of the product of its concentration, the distance, and an "extinction coefficient" depending on the solution and the wavelength. This law was an extension of another one proposed by J.H. Lambert in the 18th century, and is often called Beer's absorption law or the Lambert-Beer law. It has been of great practical importance, for instance in chemical analysis. Roudolf (1852) used a photometer of Beer's type to make a thorough evaluation of the illumination provided by various types of lamps and fuel.

Later, photometers containing Nicol prisms and/or Iceland spar rhombs were designed and used in a variety of research situations by Bernard (1852, 1853, Fig. 18-3), Provostaye and Desains (1854), Hagen (1859), Dove (1861), Wild (cf. for instance Wild 1863, Fig. 18-3), E. Becquerel (1861, 1867, Fig. 18-3), Glan (1870, Fig. 18-3). Lallemand (1871), and others. Bernard considers

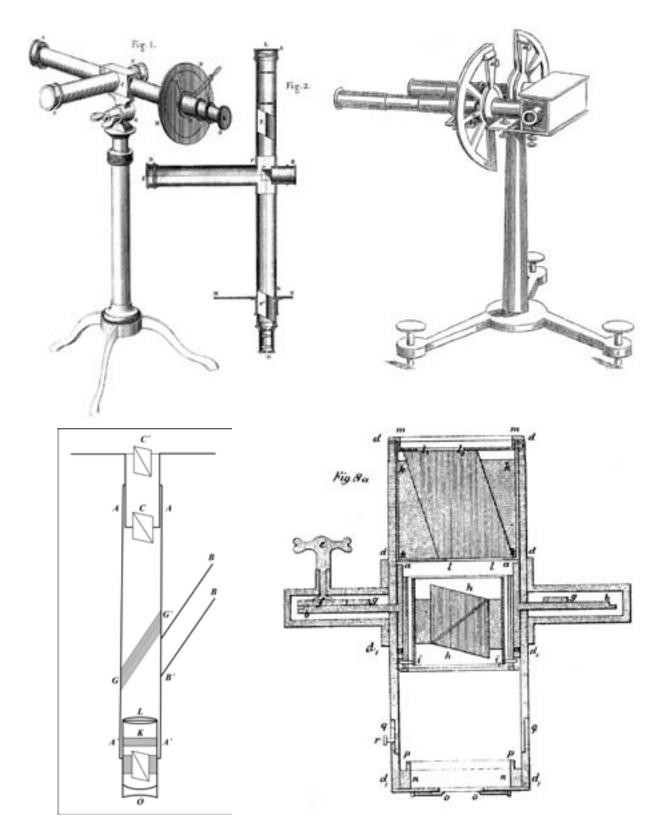


Figure 18-3. Older types of photometers. Top left: The instrument used by E. Becquerel (1861) in his research on heat radiation and luminescence. P and P' are Nicol prisms. Top right: The photometer of F. Bernard (1852). Light from two sources to be compared arrives from left through the tubes; in each tube there are two Nicol prisms. Right-angle glass prisms deflect both light beams towards the eyepiece. Rotating one of the wheels will attenuate the respective beam in a known proportion. Bottom left: A diagram redrawn from Glan (1870), illustrating his first photometer. It contains three Nicol prisms, a crystal plate K, a pile of glass plates GG' and a tube BB'. The observer looks upwards. Bottom right: The main part of Wild's (1863) photometer, with a Nicol prism of Foucault type and an Iceland spar rhomb. The remaining part is more or less Wild's polarimeter described earlier.

this to be the most accurate photometric method available. According to Becker (1903), Hagen was the first to study the absorption of light by crystals as a function of wavelength. Glan (1874) confirmed with his instrument the validity of Fresnel's laws regarding the amount of light reflected from glass at various angles of incidence. Becquerel investigated luminescence in many materials including Iceland spar and uranium salts, in addition to his work on the radiation of hot objects already mentioned. Some of these photometers were developed further and produced commercially, see sections 29.4 and 29.5.

Zöllner (1861, 1865, 1866) who had previously used Nicol prisms in research on heat radiation, designed three types of instruments containing such prisms for measurements of the magnitudes of stars and planets. With the addition of a quartz plate (Zöllner 1861, 1868), one of these was employed to a minor extent in the classification of 37 stars by their color. See Sterken and Staubermann (2000) and section 29.6 below.

In optical experiments it was often convenient to have a means of arbitrarily varying the intensity of a light beam, without necessarily measuring this intensity. Rotating a Nicol prism was one of these means; the earliest account that I have seen is in Hankel's (1862) paper on the absorption of ultraviolet rays in various materials.

19 Mining of crystals at Helgustadir, 1850-79

The main printed sources of information on the Helgustadir calcite quarry in the second half of the 19th century are papers and books by the Icelandic geologist Thorvaldur Thoroddsen (1889, 1890, 1892-1904, 1958-60). I have also inspected briefly documents in two small filing boxes at the National Archive of Iceland: Journal 16 nr. 368 from the Department for Iceland in Copenhagen and Diary 7 from the Government's II. Office, file 385. These cover respectively the time intervals 1855-84 and 1885-1908. The handwriting on many of these documents is not easy for untrained people to read, and they need to be looked at more closely. It is quite likely that more written material may be found elsewhere in the National Archive, at the manuscript department of the National Library, or in other Icelandic and Danish archives.

According to Thoroddsen, the first attempt at exploitation of the crystal site for export was made around 1850 by T.F. Thomsen, a merchant from the village of Seydisfjördur some distance to the north. From 1854 a merchant by the name of Svendsen (sources do not agree on his first name and nationality) in Eskifjördur leased the site from the Reverend T. Erlendsson who owned ³/₄ of the farm Helgustadir, at least for the period 1855-60 at a fixed annual rate. The remaining ¹/₄ was owned by the State (perhaps having been confiscated by the King from a local monastery at the Reformation in 1550), which also received a smaller annual sum by Svendsen at least in the years 1855-57. An Inspector of the property was appointed by the authorities.

In a letter from a Mr. Th. Petursson of a farmstead annexed to Helgustadir, dated 15 March 1860, there is an account of Svendsen's mining activities and his efforts to sell the crystals abroad. These efforts were not too successful to begin with (in 1855-56) but on the whole he did not lose money on the enterprise. At that time a good deal of Iceland spar must have already been available in Europe cf. a paper by Volger (1856, and a preface to it by W. Haidinger) who tells of attempts in Switzerland to polish "a piece of the purest and most transparent Doppelspath from Iceland". Judging from the paper, he had a choice of several such pieces. At a similar time J.J. Griffin (1855) in London and the optician W. Steeg (1857) in Bad Homburg advertise that they sell Nicol prisms (the latter in all sizes) as well as other Iceland spar components, see Appendix 1B. Petursson also states that little was done at the site in 1857 and 1858, but in 1859 70 crates of spar totalling 5600 pounds weight were exported. Albert (1859) in Frankfurt is advertising "excellent Nicols", projectors with "quite large calcite prisms" and other such wares. According to a letter from the local sheriff in the National Archive dated 27 Dec 1860, a ship carrying 20 "commerce læster" (= 40 tons ?) left at some time that year, another one took 866 pounds and a third 900 pounds. The Nordanfari newspaper reports in July of 1862 that a ship had been wrecked in a gale in the Spring of that year, while being loaded with Iceland spar at Helgustadir. It should be noted that most of the material recovered there may have been of inferior quality, including crystals with disfiguring "oblique colored stripes" as explained by Petursson (probably twinning, cf. section 17.2). Documents relating to activities around 1860 are difficult to read, but in the Fall of 1862 a trader named E. Iversen is trying to obtain a lease on the Helgustadir site. According to the above-mentioned newspaper, his men were working hard at the site in the following winter months.

C.D. Tulinius who was born in the border region between Denmark and Germany, became manager at Örum & Wulff's store in Eskifjördur around 1860 and bought that business around 1864. He made a deal with the Rev. Erlendsson, by then his father-in-law, to exploit the Helgustadir outcrop (jointly with Iversen to start with) for the period 1863-72 and pay the State an annual fee. The first contract with the State ran for five years with a sum of 100 rigsdaler being paid each year. The contract was then extended for two years at 20 rigsdaler per year, and extended again through 1871. It is not clear to me whether Tulinius was obliged to file any reports on the quantities of crystals that he sold, or on his earnings, to the authorities. However, contemporary export records in economic bulletins (Landshagskýrslur, published by the Icelandic Literary Society) sometimes list Iceland spar: for instance, 32 thousand pounds in 1866 but nothing in 1867-71. Hessenberg (1866) says that thousands of crystals from Iceland have been delivered to workshops and scientific laboratories, see Appendix 1b. In addition to advertisements and catalogs already mentioned, Lutz (1872) advertises optical equipment including rhombs and Nicol prisms from Iceland spar.

Around 1870 the authorities wanted to find out whether Mr. Tulinius was carrying out his operations responsibly, and whether the State might be able to increase its revenue from the site. F. Johnstrup, a professor and head of the mineralogical museum of the University of Copenhagen, was commissioned to investigate the matter. He visited the locality in 1871, on his way to evaluate sulfur mines. At that moment Tulinius himself was in Britain selling the crystals, but his men worked day and night during the summer "blasting the rock with gunpowder so that much more progress is made than ever before" according to the Nordanfari newspaper. Prof. Johnstrup's report on Tulinius' activities is dated 28 Nov of that year. Among other things he was worried that at the current rate of exploitation the Iceland spar might be fetching too low a price, and also that the site might be depleted quickly. Johnstrup suggested a cessation of the mining for some years. He had also heard that foreigners (mostly French) had been picking crystals there, which could damage the deposit.

However, it was decided to lease the mining rights to Tulinius for one more year, i.e. 1872, but the local sheriff and the Inspector were told to keep an eye on the place to make sure that he was not over-exploiting it. This caused some complaints by Tulinius, cf. letters from him in the National Archive dated 2 March and 12 Aug 1872. He also demanded compensation for being unable to fulfill conditions in his rent deal with Rev. Erlendsson on the ³/₄ of the property which by then had been extended to 1877.

The government arranged for the outcrop, by now a veritable quarry, to be covered by gravel in late 1872. C. Tulinius bought the ³/₄ part of Helgustadir from his father-in-law in 1872 or 1873. Johnstrup wrote a supplementary report dated 19 June 1873, emphasizing that the crystal site was a "unique natural wonder" which should be treated with care. He also discussed the merits of various forms of ownership, mining operations, and trade in the products, considering himself that the State should not become involved in the two latter aspects. In official records of parliamentary debates in 1879, Johnstrup is said to have recommended that the extracting of crystals would take place only every sixth year. G. Aragon who was travelling around Iceland on a vessel patrolling the French fishing fleet (probably) in 1875, says that the quarry was closed and only one caretaker around, but he and his mates collected there a "quite considerable number of fragments of spar" (Revue des deux Mondes 11, p. 774).

In 1875 the government introduced a Bill in the Icelandic Parliament, proposing to sell the State's ¹/₄ stake in Helgustadir. After some discussion, the Bill was rejected. A motion recom-

mending purchase of the other ³/₄ was also rejected. In the same session, a member tabled a Bill to "ban all export of limestone, Iceland spar and raw materials for cement, as well as bones". This member, a medical man in fact, wanted all Iceland spar to be made into lime in order to provide the population with dwellings from more robust and healthy materials than turf and stones. He thought this to be preferable to Mr. Tulinius' "exporting it to England for the production of carbonated drinks". In the ensuing debate, the proposer claimed it was well known that the Helgusta-dir quarry had been pillaged by foreigners who had without permission collected there many tons or even shiploads of the rarest and best type of calcite. Also that "the French had once taken there 30-40 tons of the Iceland spar calcite, and distributed it to museums all over Europe". The Bill was not voted upon, but in Parliamentary debates in 1879 the same member again mentioned this or another French raid on the site which was supposed to have happened in 1832 according to his source, a mineral dealer in Copenhagen. These statements are probably exaggerated cf. the last paragraph of chapter 14 above, but É. Jardin says about the crystal outcrop in his 1866 travelogue from Iceland (published in 1874) that "…the scientific commission of La Recherche exploited it on a very large scale".

20 J.C. Maxwell's electromagnetic theory of light in the 1860's

J.C. Maxwell who had been acquainted with Nicol prisms and polarized light since his school days (section 15.3), studied at the Universities of Edinburgh and Cambridge where optics researchers like P. Kelland, J.D. Forbes and G.G. Stokes were teaching at the time. It may be assumed that he followed the discussions going on about the aether and its problems. Interest in these topics was heightened by the results of Faraday (1845, cf. section 16.1) which demonstrated the influence of magnetic fields on light propagation in matter. Faraday's experiments were repeated by Verdet (1854, 1856 and later) and others with improved precision, and Maxwell refers to these investigations in his works. Among other things it was found that paramagnetic materials such as iron salts rotated polarized light in a direction opposite to that of diamagnetic substances.

Maxwell published in 1861-62 four papers with the common title "On physical lines of force" where he introduced new hypotheses about electrical and magnetic fields. In the third of these papers he points out that the a quantity obtained by W. Weber and R. Kohlrausch in 1857 from recent measurements of stationary fields had the dimensions of a velocity and a numerical value very similar to the speed of light. In that paper Maxwell also mentions Iceland spar as an example of a material having anisotropic electrical susceptibility, and derives a formula for the torque acting on a spar sphere in an electrical field. In the fourth paper (Maxwell 1862) which deals with the Faraday effect, he develops important ideas about electrical current loops in molecules which were originally proposed by Thomson (1856).

Shortly thereafter, Maxwell (e.g. 1864) presented a revolutionary theory, treating light as a propagating wave of electrical and magnetic fields, rather than an elastic wave in the aether. Both the fields were transverse to the propagating direction (and at right angles to each other), which automatically explained the polarization property of light. Maxwell also derived Fresnel's equations for light waves in crystals, and their double refraction. In his description of certain phenomena relating to electrical charges and currents, he refers to his own measurements on viscous fluids (section 27.3) where he had probably used Nicol prisms. Maxwell's theory was however not greeted with much enthusiasm for the first 15-20 years. It was partially put forward in a rather complex mathematical form, and it was no more successful than the elastic-aether theory at explaining some puzzling effects of light, cf. for instance the book by Poincaré (1892).

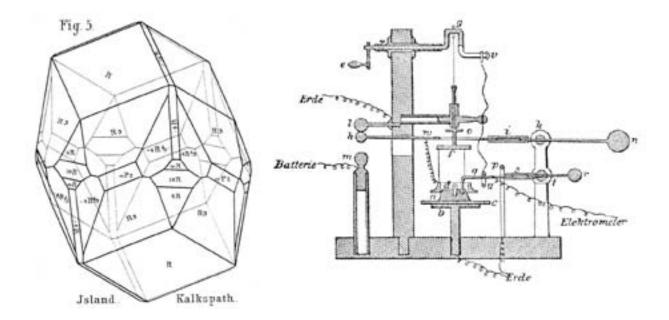


TABLE X.—Theoretical results for the line C.

	θ.	From Theory.	From Experiment.	Excess of Experiment.
I.	\$ 20 42	1.65368	1-65367	- 1
Î.	2 31 33	1.65393	1.65393	0
I.	1 31 16	1.65422	1.65416	- 6
I.	0 20 35	1.65435	1.65438	+ 3
I.	0 59 31	1.65430	1.65431	+ 1
I.	2 28 17	1.65399	1.65395	- 4
I.	4 5 7	1.65335	1.65335	0
Ι.	5 48 58	1.65231	1.65223	- 8
I.	7 39 25	1.65082	1.65078	- 4
I.	9 35 35	1.64883	1.64873	-10
I.	11 34 51	1.64635	1.64627	- 8
I.	11 37 2	1.64631	1.64623	- 8
I.	13 36 26	1.64340	1.64335	- 5
I.	15 32 41	1.64018	1.64021	+ 3
I.	17 23 14	1.63678	1.63684	+ 6
I.	19 6 50	1.63332	1.63341	+ 9
Ι.	20 42 48	1:62989	1.62991	+ 2
I.	22 9 42	1.62660	1.62669	+ 9
п.	22 35 37	1.62560	1.62580	+20
I.	23 27 13	1.62355	1.62356	+ 1
п.	23 34 31	1.62326	1.62352	+26
I.	24 33 38	1.62085	1.62095	+10

Figure 21-1. Research on Iceland spar. Top left: Hessenberg's (1874) drawing of a crystal from Iceland, with indices based on precise measurement of angles. Top right: Experiment by Rowland and Nichols (1881) on plates c of calcite and other crystals, to test certain predictions of J.C. Maxwell's theories regarding the electrical properties of insulators. Below: A table by Glazebrook (1880a) on the index of refraction of the extraordinary ray in Iceland spar. Measurements were made on four prisms I-IV at three wavelengths A, B and C from the spectrum of hydrogen, at various angles of incidence. The results agreed very well with the theories of Huygens and Fresnel.

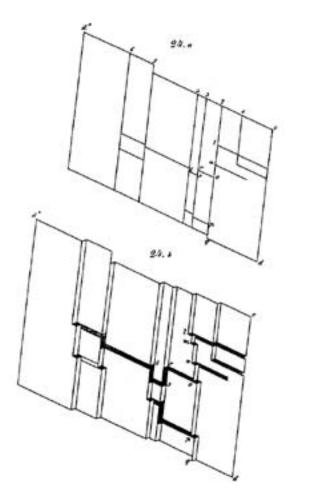
21 Continuing research on Iceland spar, 1850-1900

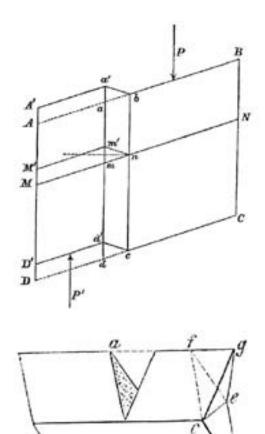
In the general development of crystallography, things were happening in the latter half of the 19th century. Regarding geometrical crystallography, A. Bravais (e.g. 1851) published three major papers with various theoretical results on the symmetry of crystals and their division into classes. It is said that C. Jordan made use of these results when composing a fundamental work on one field within group theory in mathematics, appearing in 1868 (see D.S.B.). Many advances were also made in physical and chemical crystallography. These matters are beyond our present scope, but historical reviews may be found e.g. in Arzruni (1893), Jong and Stradner (1954-56) and Lima-de-Faria (1990).

The following four sections describe some research areas where calcite crystals played a part; in many cases they probably came from Iceland although their origin is not specified in the relevant publications. A specialized experiment not belonging in any of the sections concerns the origin of gravitational forces: it was thought by some that this force might have a kind of directionality, causing the pull of gravity from a crystalline material to vary depending on angle relative to its optical axis. This was tested carefully by Mackenzie (1895) with spheres of "optically perfact calc-spar", later by J.H. Poynting with quartz balls around 1900 and P.R. Heyl with various materials in 1924. No such effect was found, within experimental errors.

21.1 The crystal forms of calcite; fluid inclusions

The simple fundamental form of Iceland spar was important to scientists, which may have been one reason why crystals from Helgustadir were rarely included in studies on other crystalline forms of calcite. Perhaps also, C.D.Tulinius and other operators had the crystals trimmed before packing them for export. A great multitude of forms has been described from other notable calcite-crystal localities, such as Andreasberg in the Harz mountains of Germany (see Sansoni 1885), whereas Hessenberg (1866) says of the Icelandic material that "...of its actual crystal forms, we know next to nothing". Thus, only a few Icelandic cases are included in Zippe's (1852) comprehensive list of known forms of calcite, and none of its 93 illustrations show a specimen from here. The same probably applies to the subsequent compilations by Hochstetter (1854), Des Cloizeaux (1874), Irby (1878), Jeremejew (1898) and Goldschmidt (1913), but I have only seen abstracts of these works or references to them. The main detailed descriptions of Iceland spar crystals that I know of, are by Hessenberg (1866, 1872, 1874, Fig. 21-1). According to Rath (1867) and Purgold (1881) the Danish exhibit at an exposition in Paris 1867 included a giant crystal of Iceland spar, about two feet in size and one foot in thickness. Rath describes it briefly, but adds a comment on Iceland: "As is known, opportunities for seeing a well-developed crystal from there have been







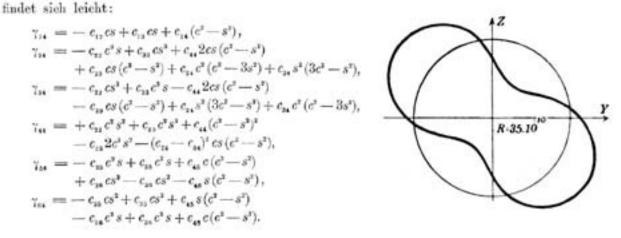


Figure 21-2. On the deformation of Iceland spar. Top left: Drawings from Rose (1868) where he shows twin lamellae on a cleavage surface of an Icelandic specimen. Elongated holes through the crystal occur where two of these lamellae intersect. Top right: A diagram from Reusch (1867) illustrating the formation of glide planes and twinning due to shear produced by the applied pair of forces PP'. Voigt (1910, p. 951) calls this "one of the most remarkable discoveries in the entire field of crystal physics". Center right: Another well-known picture, indicating how Baumhauer (1878-79) created twinning in an Iceland spar crystal by simply pushing down on its edge at a with knife blade. Bottom: Formulas and a graph presented by Voigt (1889) when processing experimental data of G. Baumgarten on the direction-dependent constants of elasticity in Iceland spar.

rare.". A comprehensive review of calcite crystal forms by Whitlock (1915) only refers to one of Hessenberg's specimens.

Scientists and collectors have from early on noticed that bubbles containing liquids are often present in the crystals of various minerals. In recent decades they have become the objects of detailed chemical studies, giving clues regarding conditions prevailing at the formation of the crystals. Such knowledge may also be of economic value, e.g. in prospecting for ores. Fluid inclusions do appear in Iceland spar (Brewster 1848b, Sang 1872-75) but I am not aware of any attempts to analyze them.

21.2 Electrical and thermal properties; dispersion

Fizeau (1862, 1866) used a recently developed interferometric method to measure very accurately the changes of shape that occurred in Iceland spar and other materials on heating. He, as well as Müller (1885), discussed possible relations between these changes and those of the refractive indices. Pfaff (1861), Jannettaz (1873) and Tuchschmid (1883) improved upon Senarmont's measurements of the thermal conductivity of Iceland spar and other crystals; Jannettaz confirmed in a subsequent paper that this property also varied with direction in schistose metamorphic rocks. Le Chatelier (1893) measured the heat released when Iceland spar and aragonite were dissolved in acid, in order to find the difference in internal energy between them, but his results were inaccurate (Bäckström 1925).

Scientists were also interested in measuring the optical dispersion of materials. This increase of refractive index with frequency of the light follows a smooth curve which has fairly similar shape for all materials, and it was hoped that a simple physical model or mathematical formula could be found to fit the curve. For this purpose the relationship of the dispersion to double refraction was investigated thoroughly, see for instance measurements on (Icelandic) calcite carried out by Esselbach (1856), Mascart (1864) and Willigen (1869), and theoretical considerations by Challis (1863) and Carvallo (1890). Soundly based physical models of the dispersion only appeared after C. Christiansen had discovered so-called "anomalous dispersion" in the dye fuchsin in 1870, and they still attempted to take double refraction into account (Sellmeier 1872, Ketteler 1874). These models assumed that the aether caused the constituent particles of materials to oscillate with it, which in fact Young (1802, p. 32-33) had suggested long before. They did not become fully realistic until after 1885-90, with recognition of Maxwell's electromagnetic theory.

Maxwell became professor of experimental physics in Cambridge 1871 and established there the renowned Cavendish Laboratory; previously, physics research at that University had mostly been of a theoretical nature. Dictionary of Scientific Biography (1980-90) notes that in this laboratory, great emphasis was placed on the quality and accuracy of measurements. One of four projects from its early days that are mentioned explicitly in D.S.B., concerned a repetition of the study by Stokes (1872, section 17.2 above) on the refraction of the extraordinary ray in Iceland spar. Glazebrook (1880a) measured the refractive index to five decimal places in four 60°- prisms for many different angles between the ray and the optical axis, and compared the results to the predictions of Huygens' hypothesis. See the lower part of Fig. 21-1.

Romich and Nowak (1874), Root (1876), and Rowland and Nichols (1881) made detailed series of measurements on the electrical properties of calcite with new equipment (Fig. 21-1). These experiments were intended to test the theories of Maxwell and others on how these materials responded to time-varying electrical fields. Both Romich and Nowak (who worked in L. Boltzmann's laboratory), and Root also tested how these electrical properties depend on direction, and Boltzmann himself had just before this published a similar study on anisotropic sulfur crystals.

Boltzmann claims (in Note 64, appended to his German translation of Maxwell's "Physical lines of force", 2nd edition, 1898) that the results increased significantly the credibility of Maxwell's electromagnetic theory of light. Boltzmann and Root, however, were aware of the fact that the effective electrical susceptibilities of materials may have much lower values in the case of light than in low-frequency electric fields. J. Curie (1889) made some measurements on the electrical properties of calcite in connection with his more extensive work on quartz. See more details in Chapter 6 of Liebisch (1891).

E. Becquerel (1885, 1886) resumed investigations on the luminescence of Iceland spar and other substances that he had been carrying out in the 1860, cf. section 17.3. He concluded from his experiments that it could be due to trace impurities of manganese, and this has been confirmed by modern techniques (see at the end of Appendix 2). It is of interest that Becquerel's son Henri carried on with similar studies in the 1890's (although not on Iceland spar), discovering radioactivity in 1896 when he was looking for connections between luminescence phenomena and W. Röntgen's X-rays.

21.3 Iceland spar and the permanent deformation and metamorphism of crystals

Brewster (e.g. 1848b) drew attention to spots or circles of light sometimes seen in Iceland spar and other crystals. The appearance of such spots had in fact been known for a long time, under the name of asterisms. Some papers on this topic were published after 1850 (Volger 1856, Kobell 1862, Haushofer 1865, Baumhauer 1869, and others) but they do not seem to have led to any notable findings. The asterisms may in many cases be related to so-called glide planes, cf. below.

Twinned crystals and thin twinned layers often present in minerals were originally thought to have been created only during the growth of the crystals from a solution. Around 1860, H.W. Dove and F. Pfaff suggested that twinning events might have taken place at subsequent times, due to the effects of pressure. Reusch (1866-67, 1867) demonstrated that it was easy to produce abundant twinning in Iceland spar, by squeezing plates of it in a vise (Fig. 21-2). This discovery generated much interest, and it must have influenced discussion on twinning in other materials. For instance, H. Laspeyres says in a paper on antimony crystals in 1870, that twins in it "...are totally reminescent of twins in calcite from Iceland, Auerbach etc." On the discovery by Reusch, Groth (1905, p. 245) states: "By far the most interesting and theoretically important phenomena are presented by calcite...". Baumhauer (1878-79, 1883) had by then made a surprising variation on Reusch's demonstration, simply producing a twinning plane by pushing on the edge of an Iceland spar crystal with a sharp knife (Fig. 21-2). Additional research on processes of this kind was carried out by e.g. Brezina (1880), Linck (1883), and Thomson (1888-89), using among other things etching techniques (cf. section 28.1). Slip-planes (Gleitflächen) had also been described for the first time by Reusch in his samples of Iceland spar and others that he had hammered on with a pointed chisel (see Scharff 1870). Rose (1868) investigated samples of Icelandic calcite which contained long hollow channels; their presence had already been noted by D. Brewster and others. Rose confirmed (Fig. 21-2) that they occurred where two glide planes intersected. These planes which may have directions different from those of easy cleavage planes or twin planes, were investigated in much detail in Iceland spar and other forms of calcite. See for instance Gümbel (1880), Purgold (1881), Mügge (1883), Lehmann (1886a), Cesaro (1890), Kenngott (1892), Voigt (1899), Wernadsky (1899), Friedel (1902) and papers referred to by them. Many other crystals were also studied; the appearance of deformation twins in metallic tin was demonstrated in 1916.

People gradually realized that the various types of twinning and permanent deformation of crystals can be classified on the basis of geometrical symmetry considerations. Thus, Liebisch

(1887) pointed out that the strain effected in the earlier experiments of Reusch and Baumhauer, corresponds to what is known in elasticity theory as "simple shear". This field of research has later risen to great influence in materials science in general, especially in metallurgy. In the paper referred to above, Voigt (1899) states that if calcite is not too different from other materials "...then we may conclude that in all deformations of solid bodies, large intermolecular displacements are taking place". In geology, these discoveries improved understanding of the changes in structure and shape of crystals occurring for instance in the tectonic folding of strata (see e.g. Adams and Nicolson 1901, Brauns 1903a, p. 388-389; Bauer 1904) or in glacier ice (cf. Klocke 1879, McConnel and Kidd 1888, Tutton 1908). More information on these matters may be found in the book by Groth (1905); a review paper by Johnsen (1913) covers additional experiments on the deformation of Iceland spar and other crystals.

Becker (1886) carried out heating experiments on Iceland spar, aragonite and other forms of calcium carbonate under pressure, in order to study their mutual transformations. From his own results and those of previous researchers like G. Rose and H. Debray in the 1860s, he drew various conclusions regarding the metamorphism of limestone.

21.4 Elastic properties and Iceland spar, 1870-90 and some earlier events

As mentioned in section 10.3 above, theoretical physicists already in the 1830s began debating how many independent elastic constants (Young's modulus, shear modulus, torsion modulus,....) were needed in each symmetry class of crystals, in order to fully describe their response in wave motion such as sound or light. Navier, Cauchy and Poisson had assumed in their analyses of the problem, that forces between neighboring material particles always were directed straight from one to the other. F.E. Neumann (1834, and later) however became convinced that this was not sufficient: one would also have to take oblique forces into account. He suggested experimental methods to test this matter, and such experiments were carried out by W. Voigt and other students.

Baumgarten (1874) made very detailed measurements of the anisotropic elastic behavior of rods, cut accurately from crystals of Icelandic calcite with respect to its optical axis. Voigt (1889, see Fig. 21-2) who processed and interpreted Baumgarten's data, states that calcite was the first crystalline material to be subjected to such studies, but rock salt and other minerals soon followed. In a paper by Voigt on these matters in 1898, the word "tensor" appears for the first time in its current meaning as a very important concept in mathematical physics.

According to Timoshenko (1953, p. 248-249) and others, Voigt (1887) correctly concluded that with certain simplifying assumptions 2 elastic constants are needed in glass, 3 in cubic crystals and an increasing number up to 21 in the least symmetric classes as Green (1842b) had indeed proposed. Iceland spar has 6 such constants, and quartz 7.

22 New instrumentation and new research, c. 1870-90

22.1 Prisms 1870-90

The development of Nicol prisms and apparatus containing them (and sometimes additional components from Iceland spar or other minerals) continued in the period 1870-90. Papers presenting novelties include those by Bertin (1870), Cornu (1870, improvements on Jellett's half-shade prism), Adams (1875), Bosanquet (1876), Lang (1881, dichroscope), Glazebrook (1883a), Bertrand (1884b), Ahrens (1885), Sorby (1885), Madan (1885) and Thompson (1886). See Fig. 12-1. Especially worth mentioning is the description by P. Glan (1880) of the improved Nicol prism which (in various versions, Fig. 12-1) has been very popular in the last several decades. In the Glan prism the ray that is going to be used travels straight through the prism, whereas it suffered a lateral shift in the original Nicol prism. In the Glan prism, the ray is also closer to being completely polarized all over the field of view, less of the light is reflected away, and the prism is shorter than the original Nicol prism for the same field of view.

The main types of polarizing prisms from Iceland spar are reviewed by Feussner (1884) and by Grosse (1890). The former paper also describes prisms intended to economize as much as possible on the spar material, cf. chapter 33 below.

22.2 Polarimeters in research and industry, c. 1870-1900

In 1870-1900, a very large amount of data was collected on the optical activity of organic compounds in various solvents. These compounds included acids, alcohols, esters, oils, sugar- and starch- products, bile constituents, proteins, and derivatives of some of these. The book by Landolt (1898; its first edition appeared in 1879) lists over 700 compounds, with detailed results on many of these. Books on polarimetry and other applications of polarized light were also published (e.g. Sidersky 1895, Pellat 1896). Presumably, optical activity measurements were of considerable use in physical chemistry, organic chemistry, and general biochemistry, as well as in chemical industries where active compounds were produced from raw materials, synthesized, or used in manufacturing processes (see e.g. Jeancard and Satie 1901 on essential oils). Below, some areas where optical activity played a part in progress, will be highlighted.

One example concerns so-called alkaloids from plants that were mentioned in section 18.1; polarimeters found various applications in research on these. A.C. Oudemans, O. Hesse and others measured the optical activity of a great number of alkaloids in the above period (e.g. Hesse 1876, Oudemans 1878, see Landolt 1898). Grimaux (1881) confirmed by polarimetric measurements

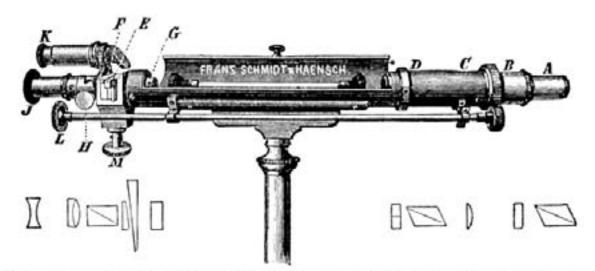


FIG. 22. - SOLEIL-VENTZKE-SCHEIBLER TRANSITION-TINT SACCHARIMETER.

- Shows position of Nicol of tint producer.
- B. Position of quartz plate of tint producer.
- C. Position of polarizer.
- D. Position of transition-tint plate,

F, E, G. Quartz-wedge compensator.

- H. Adjustment device of analyzer.
 - 7. Eyepiece.
 - K. Reading-glass of scale.
 - L. Pinion for setting sensitive-tint producer.
 - M. Pinion for moving wedge.

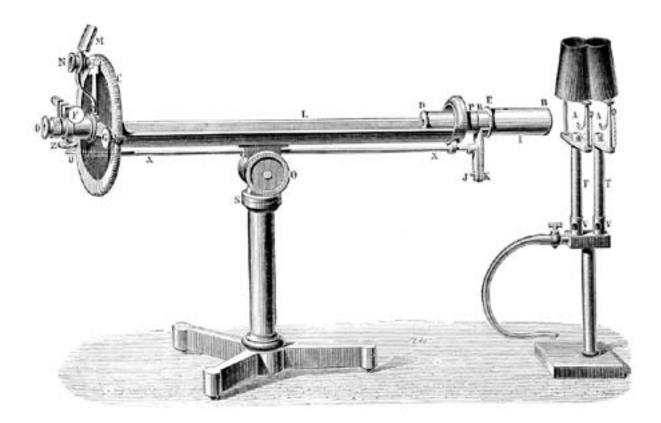


Figure 22-1. Top: A type of saccharimeter improved by C. Scheibler around 1870, with three Nicol prisms. The liquid specimen is in a glass tube underneath the manufacturer's name. Instead of rotating the analysing prism, the observer moves the large quartz wedge in or out to compensate the rotation imparted to the light beam by the liquid. From Rolfe (1905). Bottom: The half-shade polarimeter of Laurent (1874, 1879) for monochromatic light, which was popular in the sugar industry for decades. From Sidersky (1895).

and other means, that codeine which he had prepared from morphine, was identical to the natural product. This became a key point in the manufacturing of codeine and related chemicals. Antrick (1887) described a useful method to test the purity of cocaine samples. The first alkaloid for which a complete laboratory synthesis was effected, was the poison coniine from hemlock (Ladenburg 1886), also verified by polarimetry. Will and Bredig (1888) used a polarimeter to follow the transformation of hydroscamine to atropine in alkaline solution.

Another example relates to research on the terpene and camphor (or: alicyclic) group of compounds. Among these are very numerous substances from flowers, fruits, vegetables, seeds, roots, pine needles, wood resin etc., some being well known from perfumes, soaps and other cosmetics, sweets (peppermint), and spices. "The camphor series is one of the most interesting ones in organic chemistry", says the noted chemist M. Berthelot in an 1891 paper. Among those contributing to the field were G. Bouchardat, E. Beckmann (e.g. 1889), H. Landolt, A. Haller (e.g. 1892) and F. Tiemann (1897, among many); they also studied other groups of chemicals. Some of these compounds, or their derivatives, are optically active. Producers, traders, scientists and industrial staff could then use polarimeters for instance to test their purity, cf. section 27.9. It was often found that compounds of different origins which had been given different names (even up to 20 !) were in fact identical. It was especially the research of O. Wallach and his collaborators (e.g. Wallach 1888, Wallach and Conrady 1889, Wallach 1891) that increased our knowledge of the constitution of the terpenes and camphors, see Appendix 5. In the early 20th century, this knowledge became the foundation of factory production of such compounds, on a much larger scale and at much lower cost than was previously possible by isolating them from their natural sources. The composition of the factory products could be controlled, while for instance that of plant oils varies depending on soils, weather conditions and collection methods (Landolt 1898, p. 578-583) and they may contain impurities which spoil their desired effects (cf. O. Wallach's Nobel lecture, 1910).

One more example from a related field concerns the synthetic production of vanillin, $C_8H_8O_3$. This aldehyde (which is not optically active) gives vanilla, which is found in certain plant pods, most of its characteristic smell and flavor. The above- mentioned F. Tiemann and W. Haarmann (1874) found that by oxidation, the optically active chemical coniferin which can be obtained in large quantities from wood, becomes an active combination of vanillic acid and glucose. After removal of the glucose, the acid could be converted to vanillin. Haarmann patented the process and set up a factory in his home town to synthesize vanillin. It may be assumed that polarimeters were involved at some stages in this discovery and in the production process. Later, Haarmann began producing additional essences, and the firm of Haarmann & Reimer is still the largest manufacturer of such materials in Germany.

The method employed by L. Pasteur, letting microorganisms separate right-handed and lefthanded tartaric acid from a solution of both, was not tested again for the next two decades. However, interest in the method increased when it had been applied to amyl alcohol (Le Bel 1878) and to amino acids (Schulze and Bosshard 1886, Schulze and Likiernik 1893). McKenzie and Harden (1903) and Werner (1904, p. 63-64) list more than 30 compounds that had been separated in this way, using for instance the mould penicillium glaucum. Left-handed and right-handed compounds in some cases had different effects upon animals that ingested them. Many aspects of this matter were investigated, but it turned out to be quite complex (see e.g. Fischer 1898-99, a review in Stewart 1919, p. 243-256, and Cushny 1926) and is still a subject of discussion.

In connection with Pasteur's method, we may recall the case of lactic acid which played a key role in the history of ideas about three-dimensional carbon bonds and is important in various industrial processes. Although it is most common for microbes to create a racemic (50:50) mixture of this acid from hydrocarbons, there are some which only produce a right-handed or a left-handed acid (Schardinger 1890). Purdie and Walker (1892) described a chemical method to separate the

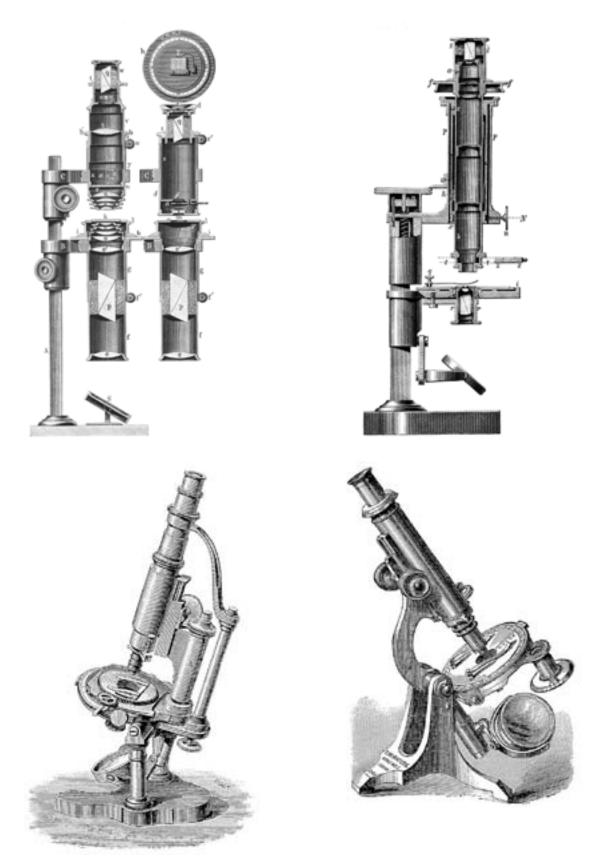


Figure 22-2. Some of the first commercially produced polarizing microscopes containing Nicol prisms. Top left: Groth's (1871) microscope which could be used as a konoscope for studying grains in thin sections (tube on the left) or a stauroscope with low magnification for studying crystal plates (tube on the right). Top right: The microscope of Rosenbusch (1876). Both were made by R. Fuess; the illustrations are from Groth (1885). Bottom left: A microscope probably made by A. Nachet, described by Fouqué (1876). Right: A new microscope from W. Watson, praised by Rutley (1879).

right-handed and left-handed parts of a racemic mixture, which was in use for a few decades in research on this acid. See Currie (1911) for a brief review of the subject.

Camphor is a general name for a resin from certain species of trees. In particular, two tropical trees produce slightly different varieties (Borneo- and Japan- camphor). Both may be dissolved in organic solvents and are then optically active. Four alternative suggestions for the structure of the camphor molecule were put forward in 1893-98. Within a few years, it was determined which of them was the correct one (e.g. Aschan 1901), partly with the aid of polarimetric measurements on camphor derivatives. Camphor and related compounds were mostly marketed as raw materials for various medical products, but after 1875 it acquired a new role as one of the two major constituents of celluloid. Into the early 20th century, celluloid was incorporated into many household objects and toys, replacing for instance ivory, mother-of-pearl, hardwood, and horn. Celluloid also was used in varnishes, photographic films (invented by G. Eastman in 1888), and all movie films after the movie industry took off around 1895. Celluloid's main advantage there, compared to the older glass-plate techniques, was of course the facility with which it could be wound on reels. In the production of celluloid "...the simple determination of camphor in solution by means of its optical activity...was of considerable importance" according to Landolt (1898, p. 452), as this was for a long time the main method for such measurements (e.g. Förster 1890). When the 1904-05 war between Russia and Japan created a shortage of camphor, it was to some extent produced synthetically from the optically active pinene, which is the chief constituent of turpentine from pine trees and is therefore inexpensive. A few different methods were available for this synthesis, based on a knowledge of chemistry which had been partly acquired through polarimetric measurements (e.g. Bertram and Walbaum 1894). The factory production, however, did not become profitable until after 1910, and soon thereafter other substances began replacing the camphor. Celluloid also lost out to new man-made materials, largely because it had a disagreeable odor and was quite flammable.

Although it now sounds odd, a discussion was taking place until around 1900 as to whether crude petroleum originated from organic remains or was generated inorganically, for instance from carbide compounds in the Earth's crust (Walden 1900, p. 198; Neuberg 1908; Stewart 1919, p. 244, and many others). The matter was settled when it was found that petroleum from some locations had a slight optical activity, as all known optically active chemicals were organic. On the other hand, L. Pasteur had expected that optical activity was exclusively a property of the living world, and that no such materials could be totally synthesized in the laboratory. However, Jungfleisch (1873) succeeded in making an active tartaric acid, and this discovery probably encouraged other chemists to take up research in that field (see e.g. Landolt 1898, p. 110, and the review paper by Walden 1900).

As regards the development of experimental methods, we may mention that a saccharimeter based on J.B. Biot's design which had been improved by Ventzke (1842, 1843) and by J.B. So-leil (1847), was improved further by C. Scheibler around 1870 (according to Sidersky 1895, p. 147, and other sources). With later additions from the workshops of Schmidt & Haensch (Fig. 22-1) it became one of the most common types in use by 1890. A more complex version appears in Glan (1891). Laurent (1874, 1877b, 1879) developed the half-shadow method of Jellett and Cornu (from Fig. 18-1) in polaristrobometers. Duboscq (1885) also claims having constructed a half-shadow saccharimeter in 1872, whereas Pellin (1899) and Bruhat (1930, p. 109) consider Prazmowski (1873) to have been the first to describe such an instrument for white light. After detailed tests (e.g. Luynes and Girard 1875), laws and regulations were issued making the use of selected polarimeter designs mandatory in the French sugar industry and Customs, including seven official sugar-testing stations for the domestic productions and five at import harbors (Girard 1877). These designs were on the market for a long time, and Brenni (1996) has established that

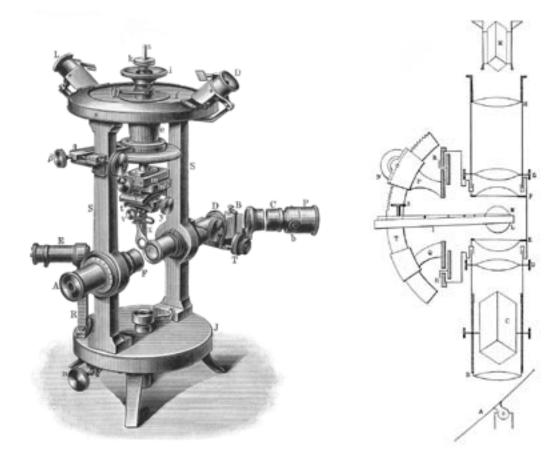


Figure 22-3. Instruments for measuring the angle between the optical axes in single crystals. Left: Apparatus with a dispersing prism B, allowing measurements to be made at different wavelengths. Nicol prisms are at P and A. This type, designed by Liebisch (1885) and manufactured for sale by R. Fuess, is similar in appearance to another one described by Lang (1867). The illustration is from Lummer (1909). Right: Cross-section of a simpler apparatus by Adams (1879).

Laurent sold 1250 units in the period 1875-86. Poynting (1880) suggested a simplification of the Laurent apparatus, and the manufacturers introduced various novelties such as concentric half-shade circles instead of two semi-circles (cf. Pellin 1903). In addition to the sugar industry (see e.g. Dubrunfaut 1870), saccharimetry was important in brewing (Heron 1888) and winemaking (Bauer 1891), processing of starch (Brown et al. 1897), and research in the food- and pharmaceutical industries (Rolfe 1905). In the British Empire, the use of polarimeters in the sugar industry began later than on the Continent, but according to Lock et al. (1882, p. 550) the determination of sucrose (crystallizable sugar) "is now universally made by means of the polarising saccharometer". The most common glass sample tubes were 20 cm in length, but specialized instruments could accommodate other containers, from 10 cm (for colored solutions) up to 1 m long or more. Polarimeters dedicated to measuring diabetic urine also appeared (e.g. Yvon and Duboscq 1880, Fleischl 1885, Duboscq 1885, Pellin 1899).

Lippich (1882, 1885; Landolt 1883) invented a new half-shadow arrangement by placing a small extra Nicol prism in the light path on one side of polarimeters; see more on this version in section 29.2 below. Wild's polarimeter which employed interference fringes rather than a half-shade technique (see section 18.1), was produced commercially from around 1870 (see Wild 1870; Landolt 1873, 1876), but the half-shade instruments had almost completely taken over the field around 1895 (Landolt 1898, cf. Sidersky 1895 and Rolfe 1905).

Other equipment introduced in that period includes devices containing Nicol and Wollaston prisms for measuring the proportion of polarized light to unpolarized (Pickering 1873-74, Cornu 1882, 1890), for instance in a reflected ray or light-- from the sky. Cornu also suggested that his "photo-polarimeter" might be used in observations of the state of the atmosphere, to aid in making weather forecasts. Pickering (1886) who will be reappearing in section 29.6, invented another polarization meter with an Iceland spar plate for use in meteorological and astronomical research, but it was forgotten for half a century. The workshop of Pellin (1899) who was producing Cornu's instrument for sale around the turn of the century, also constructed in 1900 a spectrometer with an Iceland spar prism for A. Paulsen of the Danish Meteorological Office (Boutaric 1916); Paulsen was in charge of an expedition to Akureyri in North Iceland in 1899-1900 to study the aurora borealis.

Measurements on glucose in urine were continued, see for instance Minkowski (1884) and W. Müller (1885). The former demonstrated, partly with the aid of a polarimeter, that an optically active derivative of butyric acid is excreted in diabetes. It was one of the materials that had been causing discrepancies between the results of polarimetry and other methods for estimating urinary sugar content. In research on metabolism, as well as in the medical treatment of a range of health problems, it is also important to measure the concentration of glucose in blood, but it is very low and other substances in the blood interfered with the various methods employed. Polarimetry was for a long while one of these methods, and the interpretation of results caused much debate for decades (see e.g. discussion on the conclusions by C. Bernard and others in J. Pharm. 29, 500-506, 1879; Oppler 1910). As an example, it was suggested by some that maltose sugar was present in blood (Lépine 1911); according to other suggestions, the glucose in blood and the digestive system might be chemically bound to other compounds (Michaelis and Rona 1908), or it might be in a very reactive state (e.g. Creveld 1923). Analogous problems were encountered in the measurement of sugar solutions where more than one type of sugar and/or other optically active materials might be present, for instance in liqueurs, fruit juices, cakes, condensed milk, and chocolate (Rathgen 1888, Raczkowski 1896, Fogelberg 1904, Yoder 1911, Grossfeld 1918, Auerbach and Krüger 1923b, etc.). In those cases, various means were employed to separate the interfering constituents, such as precipitation, heating, inversion by acids, fermentation, or treatment with alkalis (see e.g. Chapters X-XI of Browne 1912).

In addition to the glucose measurements just mentioned, polarimeters were around 1870 becoming useful tools in other physiological and medical research. The observations by A. Béchamp on albumins which have already been referred to in section 18.1, were continued later by e.g. Hopkins (1900). Another example is when Thierfelder (1890) established by means of polarimetric measurements, that a monosaccharide found in brain tissue was galactose. Hoppe-Seyler and Araki (1895) investigated the formation of lactic acid in animals by laboratory tests. The digestion of sugars and starch was also studied (e.g. Musculus and Mering 1878-79, Miura 1895, Weinland 1899, Krüger 1899).

Knowledge of polysaccharides from the vegetable kingdom, acquired since J.B. Biot initiated his pioneering work on these in 1832 (section 12.4), no doubt helped in research on glycogen after C. Bernard described its important role in animal metabolism in 1857. Glycogen which is primarily found in the liver, is composed of glucose units like starch (section 13.2). See for instance Bernard (1872) and Külz (1881) on comparisons between the optical properties of glycogen, starch, and some other substances. Salkowski (1894) suggested on the basis of polarimetry and other evidence, that glycogen is a constituent of yeast; this was confirmed later.

In the large-scale production and transportation of sugar compounds, starch and related substances, polarimeters once more played a role. For instance, the work by A. Béchamp on detrimental effects by microbes on raw materials in the sugar industry (mentioned in section 16.2) was continued by Gayon (1877, 1882) who suggested ways of minimizing these effects. It is clear from his papers that the industry was increasingly aware of the necessity to know the character of the materials being used, and to keep them pure. The first U.S. book on sugar production which includes sections on polarimetry (Tucker 1881) was reprinted several times to 1912. Another book by Wiechmann (1890) emphasizes the use of polarimeters at all stages in the whole production process. Research on the formation of sugars from starch by acid treatment was of course also continued (Allihn 1880, Salomon 1883, Johnson 1898), as this was already an important area within the food- and brewing industries.

The pentoses arabinose and xylose may be recovered from various gummy and gelatinous vegetable materials as well as from plant stalks, roots, bark, husks, straw etc. They are optically active, which was very useful in research on them and their derivatives (e.g. Scheibler 1873, Kiliani 1886, Wheeler and Tollens 1889, Steiger and Schulze 1890, Grünhut 1901). Another example of a sugar is the trisaccharide raffinose which was isolated by Loiseau (1876) from sugar refinery products. It is present in very small quantities (< 0.02%) in sugar beet, but at the end of the sugar extraction process may constitute up to 10% or more of some of the residual syrups (molasses). As stated already by Loiseau, raffinose has higher optical activity than sucrose, which explained why these syrups had for years appeared to contain more than the expected amount of sucrose. Scheibler (1885) and Tollens (1886) showed with the aid of polarimeters, that raffinose was was the same substance as two others, which had been derived respectively from the resin of eucalyptus trees and from cotton seeds. It was later found widely in the vegetable kingdom, and also stachyose (Planta and Schulze 1890), a tetrasaccharide which was studied at an early stage with polarimetric measurements. With improved knowledge of compounds like these, more valuable applications could sometimes be found for agricultural materials that had previously been used as animal feed, fertilizer, or fuel, or even been thrown away.

It should be noted that polarimetry was not the only method available for measurements on sugar products in industry, see section 35.3.

22.3 Optical activity in studies of molecular structure, 1880-1900

The above-mentioned J.A. Le Bel (e.g. 1891), as well as Ph. A. Guye and collaborators (Guye 1891, Guye and Chavanne 1896, etc.), P.F. Frankland, J.H. van't Hoff, A.C. Oudemans, C.S. Hudson, T.S. Patterson, H.G. Rule, H. Rupe, P.A. Levene and others attempted until 1930 to connect the optical activity of organic liquids and solutions quantitatively with the expected shape of their molecules. An important parameter here was the asymmetry of the molecular mass distribution. The main hypothesis in these efforts, referred to as optical superposition, prompted much new research in the field according to Landolt (1898, section G). Rolfe (1905, p. 249) and Lowry (1935, p. 274-277) even consider it to have been "of vital importance". The monographs on sugars by Pringsheim (1925, p. 146-150) and Bates et al. (1942, p. 428-435) also state that certain rules proposed by Hudson on the basis of these hypotheses, were very useful in the synthesis of new compounds. The superposition idea however did have its limitations, as the distribution of electrical charge within the molecules, and the nature of the bonds between their constituent atoms, may along with the mass distribution influence the optical activity (see Walden 1900, p. 171; Wittig 1930, p. 111-114; Haworth and Hirst 1930; Freudenberg and Kuhn 1931; Lowry 1935, p. 278).

Various aspects related to these studies were investigated extensively, some in fact going back to discoveries by J.B. Biot and L. Pasteur before 1860. One aspect concerned the effects of inactive solvents on the optical activity of compounds dissolved in them (Winther 1907, and many other authors). Another one was a large increase (up to a hundredfold) in rotatory power occur-

ring in many active compounds when combined with others containing boron or certain heavy metals (Gernez 1887, Walden 1897). The specific activity of some compounds such as malic and lactic acids turned out to be highly dependent on their concentration in aqueous solution or on the temperature, indicating that they existed in two or more different forms (cf. the mutarotation phenomenon of section 22.7). Discussions on the most suitable way of writing structural formulas for organic molecules, partly connected to the supposed causes of their optical activity, lasted for decades (see chapter 9 of Ramsay 1981).

The measurements by Guye and others were carried out with yellow sodium light only, but in order to obtain more significant information on the molecular structures, it was necessary to know how the activity changed with wavelength (optical rotatory dispersion). As mentioned before, J.B. Biot had found that it increased rapidly towards the violet in most cases, but occasionally (e.g. in tartaric acid, as Biot noted in 1838) the change is somewhat irregular. This phenomenon (anomalous rotatory dispersion) was studied further around the turn of the century, especially by A. Cotton (1895b, 1896) who used an instrument with Nicol prisms and a Fresnel quarter-wave glass prism (section 8.1). Cotton pointed out that the anomalous rotatory dispersion had close similarity to the strange behavior of refractive indices (anomalous dispersion) in solutions of certain dyes which C. Christiansen was the first to notice in 1870 (section 21.2). Among those studying rotatory dispersion in great detail later was T.M. Lowry (1913b) with various collaborators (e.g. Lowry and Richards 1924).

Cotton (1895a, 1896) also found that various optically active solids and liquids absorbed more of right-handed than left-handed circularly polarized light, or vice versa. This effect, sometimes named after Cotton but also called "circular dichroism" is related to the optical rotatory dispersion and yields in many cases more detailed information about the molecules involved. See Kuhn and Braun (1930) and section 39.1 on important applications of these effects in the 20th century.

W.H. Perkin the elder, who had made important discoveries in the chemistry of dyes, began in the early 1880s to perform independent research on the Faraday effect in organic chemicals. Little work had been done on these previously, except by De la Rive (1871). Perkin (e.g. 1884, 1896) published several articles on his measurements which for instance revealed a simple relationship between the number of CH_2 -groups in a molecule and its rotational (Faraday) effect upon polarized monochromatic light in a magnetic field. He also contemplated the interpretation of these results in terms of molecular structure, and one colleague wrote in a letter to him in 1906: "You created a new branch of science" (according to the entry on Perkin in D.S.B.). Among others carrying out research on this phenomenon in liquids were Joubin (1889) and Schönrock (1893); see also section 30.2.

Much was written in the latter part of the 19th century about the reasons for optical activity of crystals, cf. for instance Sohncke (1876) and Barlow (1897). Many of these papers quote an experiment by Reusch (1870) who laid a large number of thin mica sheets on top of each other. The optical axis of each sheet (which lies approximately in the plane of the sheet) made 60° angles with those above and below. Such a stack was found to rotate the plane of polarized light in a similar way as a quartz crystal; this supported Fresnel's suggestion (section 8.2) that the optical activity of quartz is caused by some kind of a spiral structure of its particles.

Throughout the 19th century, no optical activity was detected in biaxial crystals, and some scientists provided theoretical arguments for its non-existence in these (see Rev. Gén. Sci. 14, 1018-1019, 1903) while others blamed technical problems or "optical anomalies" (Pockels 1906, p. 301-303). It however supported the possibility of such crystals being eventually found, that quartz became biaxial under pressure but kept its optical activity (Mach and Merten 1876). At last Pocklington (1901) succeeded in demonstrating that sucrose crystals (monoclinic) and Seignette salt (also known as Rochelle salt, an orthorhombic Na-K tartrate) are optically active. The prop-

erty was soon found in additional biaxial crystals. It was later realized that out of the 32 symmetry classes of crystals, 15 (from all the 7 systems) should exhibit optical activity, i.e. those 11 that have no plane of symmetry (see Tutton 1922, p. 1272) and 4 others. Lowry (1935, p. 340) states that all compounds that are optically active in solution, must also form active crystals. However (according to Handbuch der Physik vol. 25/I, p. 83), optical activity had been experimentally observed in only 7 symmetry classes by 1960.

In continuation of G. v. Rath's discovery of the tridymite modification of silicon oxide (section 18.2) he found another one in 1887 and called it cristobalite. Le Chatelier (1889) investigated properties of quartz while it was being heated, and noted that they changed in a reversible way at 570°C. These properties included its optical activity and refractive indices, as measured with the method of Fizeau and Foucault (1845) of section 13.1. It turned out that at this temperature the crystal structure of quartz changes from what is now called α -quartz to β -quartz; tridymite and cristobalite are formed at much higher temperatures. These were important steps in the acquisition of knowledge about the physical state of silicon oxides at high temperatures and pressures. Such knowledge is valuable in geology (Fenner 1912) and also in industry: thus, tridymite and cristobalite are the main ingredients of "silica bricks" used widely in the internal lining of furnaces for the manufacture of for instance glass and steel. Also, quartz later became one of the raw materials in a high-temperature porcelain for use e.g. in the spark plugs of aircraft engines.

Before 1890, J.A. Le Bel and others had began speculating whether any molecules with a mirror-image symmetry (being therefore optically active) could be based on atoms of other elements than carbon. In particular, the behavior of nitrogen in its organic compounds suggested that its chemical bonds might be three-dimensional. See section 27.5 on the development of that topic during the following decades.

Great progress was made in geometrical crystallography around 1890, partly based on theoretical work by C. Jordan mentioned at the beginning of chapter 21. Among the contributors to this progress were L. Sohncke (1879) and W. Barlow, both of whom appeared earlier in this section, A. Schoenflies (1888), and E.v. Fedorow (who also contributed to advances in polarizing microscopes, see section 29.1). This progress increased scientists' understanding of the physical properties of crystals (see e.g. Liebisch 1891, Voigt 1910), and paved the way for technical applications of these properties. As an example, the absence of a certain symmetry element is the prerequisite for a crystal to exhibit the piezoelectric effect, cf. the case of optical activity: the Seignette salt that was just mentioned, was used in microphones and record-player pickups because of this property. The crystals in question were studied in detail with polarized light and other methods (e.g. Valasek 1922). W.G. Cady found around 1922 that by utilizing the piezoelectric effect in specially cut quartz plates, it was possible to design electronic circuits with very narrow resonance-frequency peaks. His work played a major part in rapid development of radio communication and broadcasting in the following decades. In patent descriptions of that period it may also be seen that people became interested in using quartz crystals in various ways in television technology, sometimes along with Nicol prisms. I have not checked what impact such patents may have had in practice.

22.4 Microscopes and other petrographical instruments, c. 1870-90

Groth (1871) describes a microscope that was constructed for him by R. Fuess in Berlin for the purpose of petrographical studies in polarized light (Fig. 22-2). Fuess made another (possibly simpler) one in 1870 for H. Rosenbusch, later on display at the Deutsches Museum. An improved model (Rosenbusch 1876), was in commercial production around 1876 (see also Girard 1875, Rosenbusch 1892, 1904). E. Bertrand (1880) describes a petrographical microscope that

was made for him around 1870, see Lima-de-Faria (1990, p. 68-69). The microscope maker E. Hartnack in Paris advertises polarizing attachments for his instruments in an 1872 catalog, and the geologists F. Fouqué and A. Michel-Lévy were publishing papers on the examination of thin sections by the methods of Zirkel and collaborators from 1874-75. They and A. Lacroix subsequently wrote much on mineralogy and on the petrography of igneous rocks. One more microscope with Nicol prisms appears in Lasaulx (1878) but in an instrument produced by Nodot (1877) a glassplate mirror is used as a polarizer. Other workshops had started producing microscopes containing Nicol prisms by 1880 (Fouqué 1876, Rutley 1879, Fig. 22-2), including the well-known firm of A. Nachet in Paris (Fig. 22-4). In 1880 W. Bulloch was the first one to make such microscopes in the U.S. (Winchell 1889), and others soon followed (see Williams 1888, Kile 2003, Fig. 22-4). Important new techniques were introduced, especially for viewing individual grains in convergent light. See for instance Bertrand (1878 and later), Lasaulx (1878), Michel-Lévy (1877, 1883, Fig. 22-5), Mallard (1882) and the horizontal microscope of Dufet (1886). E. Abbe (1884), the famous designer of optical instruments, and others tried mounting a special Iceland spar analyzer prism between the lenses of a microscope ocular, but this arrangement turned out to have some disadvantages (cf. Calderón 1878, Köhler 1926, p. 914-915, 940-941, Ambronn and Frey 1926, p. 102). The above developments are discussed in for instance a review by Fouqué (1879), in the book by Rosenbusch referred to, and in the comprehensive volume by Johannsen (1914).

In addition to the ordinary type of polarizing microscopes, other instruments with Nicol prisms for petrographic research appeared in 1870-90. Stauroscopes (i.e. microscopes with moderate magnification, for establishing axial directions in crystal plates) have already been mentioned; they were improved by Brezina (1867), Groth (1871, Fig. 22-2), Calderón (1878) and Laspeyres (1882) by the use of a specially cut Iceland spar plate. Stauroscopes were produced at least to 1920, see Tutton (1922, p. 974 and 1159) who states that Groth's stauroscope was "the most efficient form of polariscope for the measurements of extinction angles". These are angles that are measured between the optical axis of a crystal and the direction of polarization in a polarized ray passing through it, under certain conditions. The magnitudes of extinction angles turned out to depend on the composition of biaxial crystals, and so did the angle between the two optical axes. Sophisticated instruments for axial-angle measurements were designed by Groth (1871), Adams (1879, Fig. 22-3), Becke (1879), Fuess (see Bauer 1882), Liebisch (1885, Fig. 22-3), and Mülheims (1888). Devices for measuring refractive indices with great accuracy included so-called total-reflection meters described by F. Kohlrausch (1878, see also W. Kohlrausch 1879), Bauer (1882), Pulfrich (1887) and Abbe (see Leiss 1898a). Rollett (1881) made a spectro-polarizer (Fig. 22-4) to allow microscopic measurements at various wavelengths. Some of these instruments were produced commercially, the last-named by the Zeiss company according to Rollett (1891) and Zeiss catalogs to at least 1902.

By making use of comparisons of optical properties and chemical analyses on large crystals, improved models of polarizing microscopes (see e.g. new types from R. Fuess in Fig. 22-6) could to an increasing degree be employed in estimating the chemical composition of small crystals in thin sections. This applies especially to the plagioclase feldspars mentioned in section 18.2 (see e.g. Des Cloizeaux 1875, Schuster 1881, Becke 1906) but also to the pyroxene and amphibole groups. In the microscope work, it was also found that minerals which previously had been known from a few localities, were in fact quite common in the Earth's crust. Thus, Grubenmann (1887) writes that petrographers had been "…very surprised to encounter these accessory components of rock in unexpected quantity and variety. Apatite, magnetite, zircon, rutile, tourmaline, hauyne, perovskite etc. etc. have emerged as ever more widely distributed minerals". This view is seconded by Tschermak (1894, p. 5). People also found that particular minerals often accompanied each other in rocks, and they could track the changes occurring on alteration. This among other

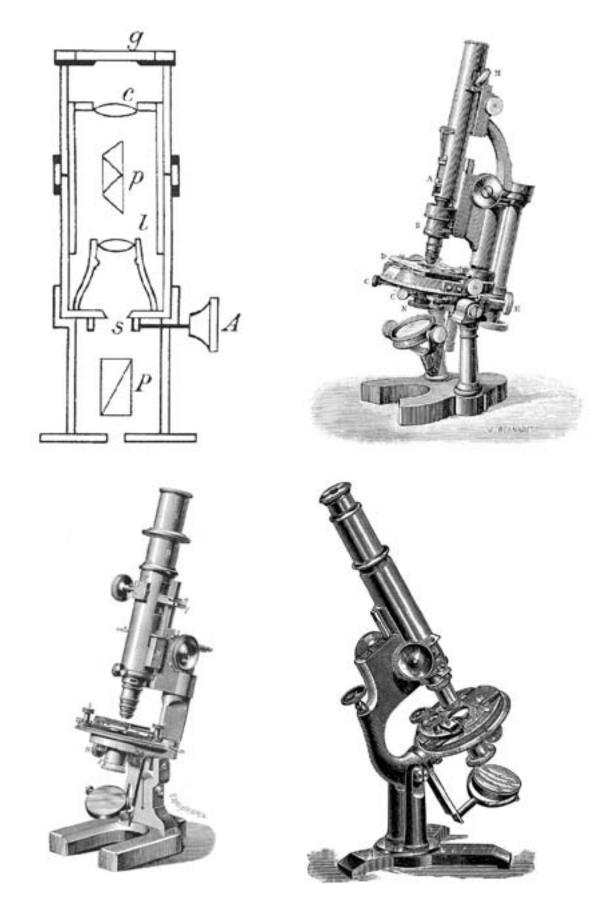


Figure 22-4. Polarizing microscopes in development. Top left: The spectro-polarizer of Rollett (1881) for viewing microscopic specimens in different colors. From Becker (1903). Right: A. Nachet's (1979) "grand modèle", from his 1881 catalog. Bottom left: Rosenbusch's (1885) microscope, incorporating improvements from his design of 1876. Right: The microscope of Williams (1888).

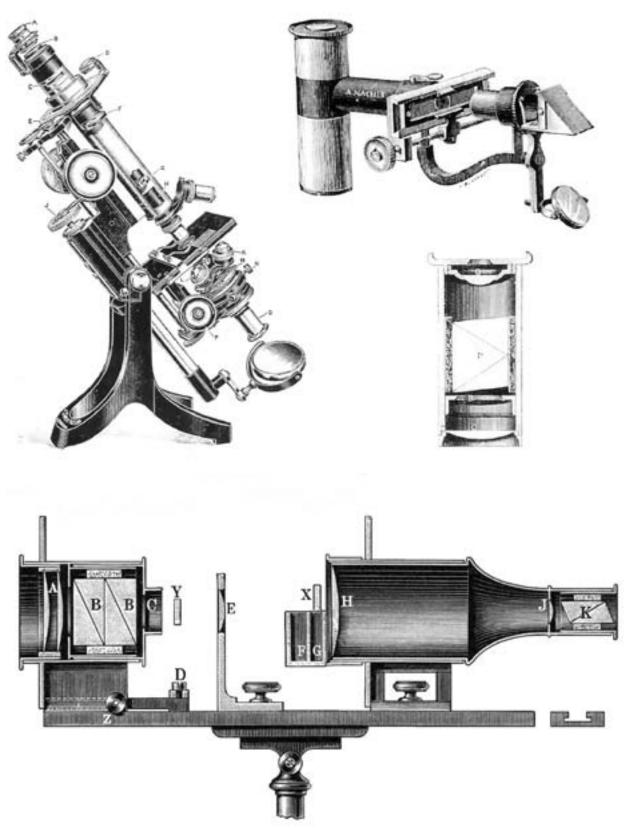


Figure 22-5. Various innovations. Top left: The petrographic microscope of Dick (1889), as improved and manufactured by J. Swift. From Johannsen (1914). Right: The upper object is the Michel-Lévy (1883) comparator for measuring birefringences in thin sections. It incorporates two Nicol prisms and a quartz plate. From A. Nachet's (1979) catalog of 1892. The lower object is the special analyzing prism installed by Abbe (1884) in a microscope ocular. Bottom: A projector with large Senarmont and Nicol prisms, first made by J. Duboscq (Bertin 1875) but later improved by V. v. Lang. Light enters from the left, crystal plates e.g. for demonstrations are placed at X or Y. From Verdet-Exner (1887).

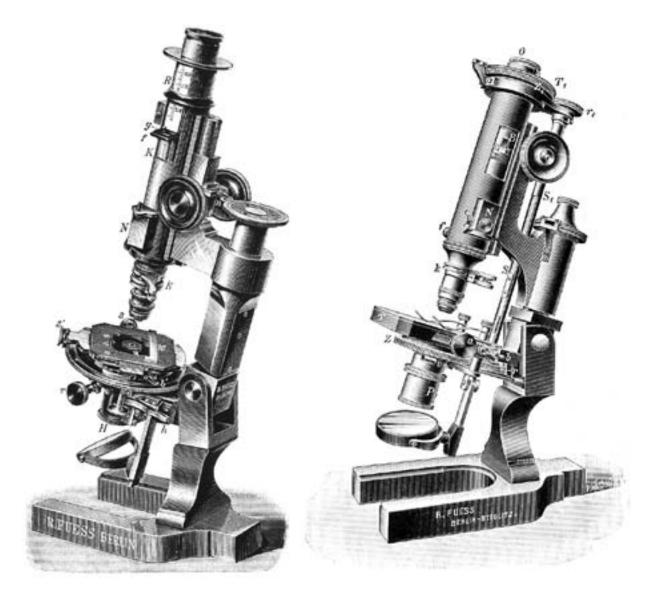


Figure 22-6. Two microscopes from the R. Fuess workshop. The one on the left was presented by Fuess (1891). The other one which is probably somewhat younger, is illustrated in the book by Duparc and Pearce (1907).

things led to some revisions in the classification of rock types. P. Hautefeuille (1880), F. Fouqué (1882), A. Michel-Lévy, C. Friedel and others produced around 1880 in the laboratory various minerals, gemstones (see Fremy 1891), rocks such as basalt, and even meteorites. Their work was continued after 1890 by scientists like J. Morozewicz (1898) and H. Moissan (Nobel prize in chemistry 1906) who in his electrical furnaces succeeded in making both diamonds and the new abrasive compound carborundum.

C. Ahrens who was mentioned in section 22.1, and perhaps some others as well, constructed quite large Nicol prisms (up to 7-9 cm wide) and beam-splitting prisms while large crystals of Iceland spar were available. Most of these were probably used in equipment for demonstrations in optical crystallography and mineralogy, see Cheshire (1906-07) and Tutton (1922, p. 848-860). A versatile projector from J. Duboscq with large Nicol prisms (Bertin 1875) was later improved by V. v. Lang (Verdet-Exner 1887, p. 132; Lummer 1909, p. 854), see Fig. 22-5. Mach (1875) and Govi (1880) built projectors for demonstrating polarization effects in crystals, where the analyzing prism was rotated by a motor; these were manufactured for sale (e.g. Pellin 1899).

Increased interest in microscopical studies in geology was reflected in the publication of scholarly journals. G. Tschermak began in 1871 issuing Mineralogische Mittheilungen, which became Mineralogische und Petrographische Mittheilungen in 1878. The Mineralogical Magazine was founded in 1876, and P. Groth launched the Zeitschrift für Kristallographie und Mineralogie in 1877. The Bulletin de la Société Minéralogique de France appeared in 1878, and the long-established Neues Jahrbuch für Mineralogie expanded considerably from 1880. These journals contained annually tens of papers on mineralogy and petrology, in addition to a much larger number of extracts of papers from other journals and serials, doctoral theses, and books.

Examples of the use of polarizing microscopes in biology in 1870-90 include the research by Ebner (1874, 1882 and more) on the structure of bones and teeth, as well as of carbonate skeletons in sponges and corals. Engelmann (1873, 1875, and later) investigated with a polarizing microscope important features of the action of muscle fibers. Modern textbooks also indicate that such microscopes found continued applications in other studies of muscles, connective tissue, substances like cholesterol (which is a liquid crystal, see section 27.2), and crystals from blood (Ewald 1886). Around 1880 various scientists were beginning to investigate chlorophyll, mostly because of its biological importance but also because of its fluorescence. This research had some problems as chlorophyll is a rather unstable compound, it crystallizes only with difficulty, and it is generally accompanied by related colored compounds like carotene and xanthophyll. Borodin (1883) was the first to show, using a polarizing microscope, that some of these compounds could form doubly refracting crystals.

23 Effects of electric fields and magnetic materials on polarized light, 1875-1900

J. Kerr (1875, 1879, 1880, see Fig. 23-1 and a diagram in Kristjansson 1996) demonstrated that when linearly polarized light passes through some transparent materials like glass or fluids in a strong transverse electric field, then these materials show evidence of double refraction. This effect of the field, which is generally very modest, is variable depending on the materials, and is proportional to the square of the field intensity, was named the Kerr (electro-optical) effect. Its discovery generated considerable interest, but its existence was not confirmed until a few years later, by W.C. Röntgen (1880) and G. Quincke. The Kerr effect was then investigated in more detail by Quincke (1883), Blondlot (1888), Kerr himself (1888, 1894) and others. The effect is most noticeable in substances where the molecules are quite asymmetric, i.e. have a large electric dipole moment. Among those showing the most pronounced Kerr effect were the liquids nitrobenzene ($C_6H_5NO_2$) and nitro-toluene. Later, nitro-benzene and CS_2 were used in various technical applications of the effect, see chapter 38.

A related optical effect due to electric fields, which however increases in proportion to the field intensity, was probably first noted in quartz by Röntgen (1883) and A. Kundt. Among later investigators were Beulard (1893) and in particular Pockels (1891, 1894, 1906), who had previously studied in detail the effects of pressure on optical properties of quartz and fluorspar. This electro-optic effect which only appears in crystal classes exhibiting the piezo-electric effect, was therefore named after Pockels. It causes a change in the state of polarization of light on passing through an a crystal (superposed on the changes which happen anyway). See section 39.6.

Kerr (1877, 1878) also discovered a new magneto-optical effect when he let a linearly polarized light ray be reflected from a polished iron surface at the core of an electro-magnet. The light then became elliptically polarized, to an increasing degree as the magnetization of the iron increased. This discovery created even more stir than Kerr's electric effect, and others looked e.g. for connections between this effect and the Hall effect (i.e. appearance of a transverse electric field in a current-carrying conductor in a magnetic field) which was found at a similar time. The Kerr magneto-optical effect was investigated by Hall (1881), Kundt (e.g. 1884, thin films), Righi (1885, 1886, 1887, Fig. 23-1), H. du Bois, P. Zeeman, D. Goldhammer, and others around 1890 (see Drude 1900a, p. 412-419). It was found that this effect resembled a very strong Faraday effect: in passing through iron carrying its saturation magnetization, the plane of polarization rotated of the order of 20,000° /mm.

In the time interval given above, research on the Faraday effect was continued, for instance by H. Becquerel (1877, 1880, Fig. 23-2) in scores of solids, liquids and gases; cf. also the paragraph on W.H. Perkin in section 22.3. Sohncke (1886) demonstrated that the magnetic field caused

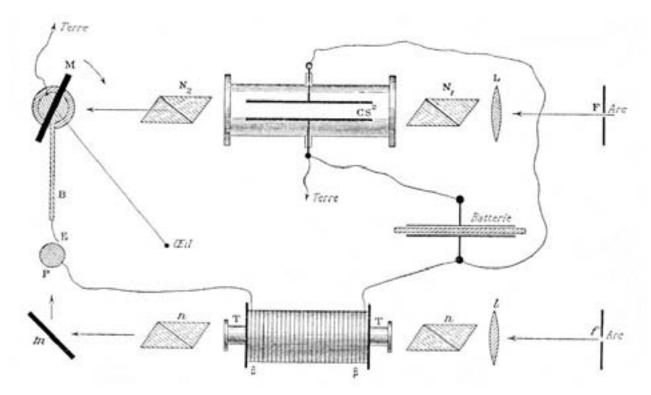


Figure 23-1. Left: The upper part of the drawing shows an experimental setup for the Kerr (1875) electro-optic effect. Light arrives from the right through the Nicol prism N1 and a glass vessel containing CS2, but it stops in N2 which is perpendicular to N1. When a high-tension voltage source has been connected to the capacitor plates, part of the light can reach the mirror M. Blondlot (1888) is testing here whether this effect has a shorter time constant than Faraday's magneto-optic effect (in the lower part of the drawing). Right: One of many experiments of Righi (1885, 1886) on aspects of Kerr's (1877, 1878) magneto-optic effects. Three Nicol prisms, the quarter-wave plate m, and the glass plate V are used in measuring the rotation of the plane of polarization in a beam of light upon reflection from the polished iron pole of the electro-magnet R_2 .

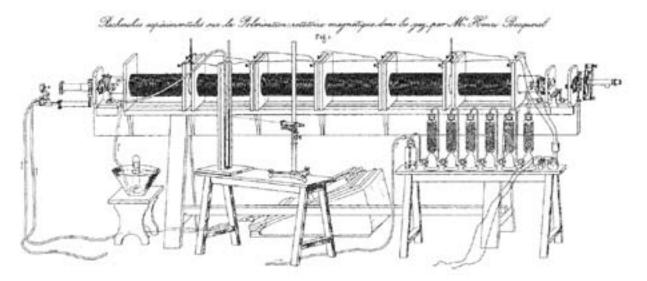


Figure 23-2. Becquerel's (1880) instrumentation for measuring the Faraday effect in gases. On the left we see a gas-flame light source and "un très beau prisme de Nicol", 4.5 cm square. The optical rotation is measured with another Nicol prism on the right, after nine trips along the tube which is 3 m long and tightly wound with current-carrying wire.

materials to act upon light in a way very similar to the optical activity in quartz. These electric and magnetic effects no doubt encouraged new discussions on the nature of light, but I have not investigated whether they generally increased support for Maxwell's electromagnetic theory. This must however be considered a likely consequence, as is indicated by the fact that the first scientific paper of G.F. FitzGerald (1877) concerned the Kerr magnetic effect; FitzGerald soon became one of the main proponents of Maxwell's theory. See more on this in section 28.4.

24 The Iceland spar quarry 1879-95

In 1875-76, the authorities in Iceland negotiated unsuccessfully with C.D. Tulinius on the continuation of his lease of the Helgustadir quarry (cf. a letter to the Governor of Iceland printed in the Stjornartidindi B Gazette, 16 Dec 1876). Not much seems to have happened in the following years, until the Parliament voted in 1879 to permit the purchase of the ³/₄ parts of the Helgustadir farm for up to 16,000 kr. Legislation to this effect was issued in September. The price was paid directly to a wholesaler in Copenhagen, to whom Tulinius owed money. Tulinius made the Government an offer dated 7 Oct of the same year, that he would operate the quarry and organize the sales of its products for a certain fee, but this fee was considered far too high.

One more evaluation by F. Johnstrup of matters regarding the Helgustadir site is dated 18 May 1881. He is of the opinion that Tulinius removed "quite extraordinarily large quantities" of optical crystals from the mine, especially in the final year of his lease (i.e. 1872), and that his stockpile had been sufficient to supply the market since then. This view is echoed for instance by Merrill (1905). Johnstrup states that no one knows the size of Tulinius' remaining stock, and neither could he find out how much of it was being sold annually. He suggested that the quarry should be operated every fifth year. During the summer of 1881, it was visited by the Norwegian geologist A. Helland (1884) who described it briefly, with a drawing, in a paper on the geology of Iceland.

It was decided that Iceland's first geologist, Thorvaldur Thoroddsen, should travel to East Iceland in 1882 to resume mining operations. These plans were mostly dropped (cf. report from Thoroddsen dated 8 Nov 1882, in the National Achives), both on advice from Johnstrup and because of difficult conditions such as a measles epidemic and very adverse weather. However, Thoroddsen arranged for the recovery of a quantity of crystals which were exported in 13 boxes and sold by Johnstrup to the optician F. Schmidt in Berlin in late 1883. It seems likely that this Schmidt is the one who in 1864 had become a partner in the firm Franz Schmidt & Haensch. The proceeds did not cover the costs of this test operation, and Thoroddsen in fact states in his report that he does not believe that the quarry can be run profitably. Thoroddsen quotes Tulinius telling him that he had himself exported in total 280 tons and his predecessor Svendsen 50 tons. I guess that much of this material may have been of inferior quality.

In the year 1885 another mining attempt was made on behalf of the government. Various documents relating to it survive in the National Archive, including lists of tools and of wages paid to local laborers (Kristjansson 2003). T. Gunnarsson, a noted entrepreneur and politician, directed these efforts (cf. vol. IV of his biography by B. Jonsson, published in 1990). The material recovered was sold in commission through the laboratory-supplies firm of H. Struers in Copenhagen (which had bought 20 kg in 1884, through Johnstrup), mostly in the years 1886-90. Garboe (1961, p. 340) states that Struers for a long time was offering Iceland spar from a small stock he kept, even from 1871 (although his firm was not founded until 1875).

It is likely that other parties than the State collected some crystals from the Helgustadir quarry in the period 1879-90. Thus, Sède (1884) mentions that in the year 1883 a good piece of 8 x 6 x 2 cm size was offered for 4 kr. at the site. According to Eiriksson (1930), some transparent crystals weighing 2-300 kg were recovered and sold for tens of thousands of kronur in 1886-91. Labonne (1887) who passed through in 1887 was of the opinion (as was Helland in 1881) that plenty of crystals still remained there. According to his account, C.D. Tulinius was leasing the quarry at this time: I have not seen contemporary evidence supporting this, but the newspaper Thjodolfur on 29 Sept. 1910 indicates that Tulinius and his son Thorarinn did lease it for some unspecified time interval between 1880 and 1910. Finally it may be added that Bréon (1882) in the year 1880 took a look at the calcite outcrops in Djupidalur in NW-Iceland which O. Olavius had inspected a century before. Bréon writes that the calcite in the Djupidalur outcrops is generally opaque, but if that turns out to be caused by weathering then these outcrops may perhaps replace the Helgustadir site when depleted.

Haensch (1889) quotes the above-mentioned equipment dealer H. Struers in Copenhagen as saying that Danish authorities intend to send an expedition to Iceland in 1891 to collect a large quantity of spar. I have no further information on these plans. In the Parliament of Iceland in 1891 a Government bill was tabled, authorizing the sale of the quarry (as an entity separated from the Helgustadir farm). In explanatory notes appended to the bill it is revealed that the operations in 1882 and 1885 returned very little profit, and that Prof. Johnstrup advises asking the major constructors of optical instruments for bids on leasing the site. This bill was rejected.

25 The "spar famine" in 1884-1920, cf. quotes in Appendix 1

Past 1880, the supply of Iceland spar appears to have satisfied demand, although for instance Ladd (1874) and Laurent (1877a) who needed large prisms from it, had to manage with smaller pieces glued together. Thus, the well-known instrument maker A. Hilger advertises in the astronomical journal The Observatory in 1880-82 that he has a selection of prisms (probably triangular ones) for sale (Fig. 25-1). After 1882 he does not advertise any Iceland spar in that journal, and around 1884 complaints begin to appear regarding a shortage of spar for the purposes of research and equipment manufacture. Sentences from various sources of such complaints have been assembled in Appendix 1A. The president of the Royal Society of London, G.G. Stokes who himself had made important discoveries in optics as noted above, wrote a letter to the Minister for Iceland, dated in July of 1886. The letter, preserved in the National Archive, is reproduced in Appendix 1C. Here he points out that Iceland spar used to be easily available in Britain, but a dearth of the material in recent years is causing great problems. Stokes implores the government to re-open the quarry; perhaps pieces previously rejected might be found around the mine and sold to offset initial costs. The letter was accompanied by a translation made by his friend Eirikur Magnusson in Cambridge. In the Parliament session of 1887, a motion to allow Eirikur to recover up to 500 pounds of crystals at Helgustadir was tabled but not discussed or voted upon.

Labonne (1887) writes that in France there are great worries about the limited availability of Iceland spar. Towards the end of that year, German earth-science journals (cf. Zeitschr. Deutsch. Geol. Gesellsch. 1888) report that the office of the German Chancellor is making enquiries to authorities in Iceland and in the U.S. about Iceland spar at the request of instrument manufacturers. According to the journals, optical-quality crystals of calcite are very seldom found outside of Iceland. In response to these enquiries, T. Thoroddsen (1889, 1890) published articles that appeared at least in Danish and German. Here as well as in his later books, Thoroddsen pins his hopes on Djupidalur as a possible new source of spar crystals.

H. Haensch (1889), addressing a meeting of German manufacturers, informed them that the previous enquiries made by the Chancellor's office had not resulted in any new Iceland spar appearing on the market. The situation was becoming serious for the workshops involved, as well as for the sugar industry and others. A motion was passed at the meeting, strongly urging their government to intervene in the matter. The participants guaranteed that they would buy a considerable amount of crystals if material of sufficient quality could be procured.

J. L. Hoskyns-Abrahall (1892) from Britain made a detour in his journey to Iceland in 1890 to inspect the quarry, at the request of H.G. Madan (mentioned above) who was concerned about "the difficulty of obtaining large pieces of clear calcspar for optical purposes". He writes that Thor E. Tulinius (a son of C.D. Tulinius) who resides in Copenhagen, is still selling crystals from

ICELAND-SPAR PRISMS

SPECTROSCOPES COMPLETELY MOUNTED WITH QUARTZ LENSES AND PRISMS OF ICELAND-SPAR.

ADAM HILGER,

SCREWER DE LEMERANY MALIE TO THE ROYAL COMPERANCE AN GRANWICH AND RECEPTER, THE COMPERANCE OF MELSOCREE, STAWY, POINTAR, AD. &C. 193 TOTTENHAM COURT ROAD, W.

SPECTROSCOPES

WITH ICELAND-SPAR OR QUARTZ PRISMS, QUARTZ LENSES, AND FLUORESCENT EYEPIECES,

CONSTRUCTED FOR THE OBSERVATORY OR LABORATORY. A large stock of unmounted Iceland-Sper Prisms and Quartz Lenses of various sizes on hand.

> List of Prices post-free on application, ADAM HILDER, Astronomical and Optical Inframent Maker, 192 Terremona Cores Ross, Leocar, W.

Figure 25-1. These advertisements for Iceland spar prisms, from the well-known instrument maker A. Hilger, appeared in the astronomy journal The Observatory in 1880 and 1882 respectively. At that time he was also offering triangular prisms of up to 5" size, and spectroscopes with six prisms. Soon after 1882 however, the supply of Iceland spar declined sharply.

his father's stock. On the other hand, Fr. Johnstrup in an 1890 letter preserved in the National Archive, appears to believe that the last of the stock was disposed of in 1889.

It is likely that during this time of shortage, instrument makers acquired Iceland spar by for instance purchasing spare specimens from museums (Beckenkamp 1915). Garboe (1961, p. 503) mentions that the Mineralogical Museum in Copenhagen was one of those approached, but he does not provide additional details. It was also fairly common for large private collections of minerals to be auctioned off, say in case of death or bankruptcy of the owners. Advertisements of such events might appear in journals like the Neues Jahrbuch für Mineralogie or in newspapers; thus, auctions including optical apparatus, Nicol prisms and Iceland spar were announced in The Times e.g. on 2 July 1885 and 25 Nov 1887.

Grosse (1890, see Appendix 1A) states that Iceland spar prisms have become expensive, as 95% of the raw material is wasted in making them. Beckenkamp (1915) and others iterate that large clear spar crystals have only been obtained from Iceland, while specimens from other localities may be used for making small Nicol prisms. These localities possibly included some in Germany such as Auerbach an der Bergstrasse; in 1895 R. Scheibe exhibited two Nicol prisms made of Auerbach calcite (not of sufficient quality for precision instruments) at a meeting of the German Geological Society. Reasonably good pieces were also found in the Crimea region of Russia shortly before the turn of the century. More on these matters in chapter 32 and in Appendix 1A, 1B.

26 Iceland spar and chemical research, especially in 1840-1915

Chemists have been steadily improving their analytical methods, and in these it is of course imperative to know precisely the atomic weights of the various elements. Measurements of atomic weights could be inaccurate if the compounds used were derived from impure raw materials; for instance Dumas (1842, p. 546) proclaims that in such determinations one has to "make analyses or syntheses on a large scale, operating with simple reactions of very pure compounds.". A similar view is expressed by Marchand and Scheerer (1850).

As regards calcium, results of measurements of its atomic weight were somewhat variable in the period 1840-1900, fluctuating between 39.96 and 40.22 (on the basis of oxygen = 16). Out of these attempts, those made by Dumas (1842), Erdmann and Marchand (1842, 1844, 1850) and probably some others employed Icelandic calcite as their source of calcium. I have not read the works of J.-S. Stas who was most productive in atomic weight determinations during the second half of the 19th century, except for one paper published posthumously (Stas 1892). In chapters V-VI of this paper it is revealed that he did make use of Iceland spar to a considerable extent when preparing samples for spectral analysis.

In an 1898 report of a committee on atomic weights, organized at the initiative of the German Chemical Society, the weight of calcium was merely listed as 40 (Ber. Deutsch. Chem. Gesell-sch. 31, 2761-2768). At that time, debate was still going on regarding the validity of W. Prout's hypothesis dating from 1815, which postulated that all atomic weights were integer multiples of that for hydrogen.

In a special investigation of the atomic weight of calcium, Hinrichsen (1901) emphasizes that in his analyses he only employed some crushed pieces of Iceland spar. From these and other published investigations (see Appendix 2) it is clear that the Icelandic material is very pure calcium carbonate, containing less than 0.1% of other elements. In particular it was quite free of crystal water, which caused problems in the use of most other calcium minerals from nature or of manmade compounds for this purpose. The result obtained by Hinrichsen (1901) was 40.142. At a similar time, T.W. Richards (1902) announced a preliminary value of 40.126 for calcium from marble. The committee mentioned in the previous paragraph (which in the meantime had been raised to international status) therefore changed its listed value to 40.1 (Ber. Deutsch. Chem. Gesellsch. 34, p. 4384, 1901). Shortly thereafter, Hinrichsen (1902) repeated his measurements using Iceland spar from the Crimean peninsula. Now he obtained 40.136 but he appears to have had more faith in his earlier result. An atomic weight value of 40.09 for Ca was approved by the international committee in 1909 (see Rev. Gén. Sci. 24, p. 102-103) and 40.07 in 1912 (see J. Prakt. Chemie 85, p. 93) in agreement with a new determination by Richards on calcium salts. Oechsner de Coninck (1911) obtained 40.06 from a measurement using Icelandic material. The committee finally settled on 40.08.

From bibliographies on the Internet and other sources such as Hughes (1937) and Fries (1948, p. 131) it is clear that Iceland spar found wide use among chemists from around 1860 (cf. Zeitschr. Anal. Chemie 41, p. 145, 1902) until 1960 or even later. Much of this material may have been left-overs from the production of Nicol prisms. Iceland spar did play some role in measurements of the atomic weight of fluorine (Louyet 1849), chlorine and boron (Laurent 1849) and, indirectly, magnesium (Marchand and Scheerer 1850). In papers on new or modified procedures for the analysis of carbonates it is sometimes stated that the authors (e.g. Jäger and Krüss 1888) used samples of Iceland spar for the testing of their methods.

Besides, Iceland spar was often useful in the standardization of solutions in inorganic analysis, physical chemistry, and biochemistry. Bornträger (1892) calls it "a prime titration substance for the control of nitric or hydrochloric acid". D.O. Masson (1900) advises chemists to measure the concentration of for instance hydrochloric acid by the amount of Iceland spar ("probably as pure a substance as any that the chemist deals with") it dissolves. According to Masson's 1938 obituary, the method was in common use; it is also described by Thiele and Richter (1900). I do not know where Masson or others obtained their material. Other examples where Iceland spar played a role in chemical research include work by Wagner (1870, test of new method for determining gaseous carbon dioxide), Crookes (1885, notable research on the spectra of rare-earth metals), Lemberg (1887, demonstration of a microchemical procedure), Heidenhain (1896), Brunck (1906), Blanc and Novotný (1906, production of caustic soda), Rose (1909), J.I.O. Masson (1911), Dodge (1915, standardization of solutions), Winkler (1916, measurement of the calcium content of potable water), Foote and Bradley (1926), and Dittler and Hueber (1931, testing of methods to measure carbonic acid in rock).

Knowledge of the chemical equilibria between calcium oxide, carbon dioxide, calcite and/or water under different conditions is important for instance in various industrial processes, as well as in agriculture, hydrology, oceanography, and in research on the geochemistry of limestone strata. Many scientists have written on their experiments in these fields and on the interpretation of the results in the light of thermodynamic principles. Among those who specify that Iceland spar was employed in their experiments, are Raoult (1881), Kendall (1912), Prideaux (1915), Smyth and Adams (1923), and Miller and Witt (1929). It is quite possible that this was also the case in other comparable projects where the origin of calcite samples is not stated.

Around 1890, chemists were discussing whether the well-known ability of strong electrolytes to dissolve other substances depended on some water being present. One experiment for testing this matter was carried out by Hughes and Wilson (1892) who exposed Iceland spar to very dry hydrogen chloride gas at 100°C for long periods of time. The gas then turned out to be rather ineffective.

27 Various research subjects involving Nicol prisms, around 1900

In the decades around the turn of the century, optics was a popular field within the natural sciences. Polarized light was a key component in many well-known books published before 1900 on optics in general (e.g. Beer 1882, Verdet-Exner 1887, Basset 1892, Mascart 1889-93, Dufet 1898-1900) as well as on specialized topics like crystal optics (e.g. Pellat 1896) and optical activity (Landolt 1898, Fig. 27-6, and others). Optical research also made great strides; several important fields where Nicol prisms were involved, will be described briefly in this chapter.

27.1 Scattered light, and the color of the sky

From F. Arago's discovery of the polarization of light from the clear sky in 1809 and until the 1860s, many scientists studied it with a variety of polariscopes. The blue color of the sky and the polarization (which varies depending on the Sun's altitude and on the viewing direction) were even called "the two great standing enigmas of meteorology". Some made very comprehensive surveys of the polarization, such as Rubenson (1864). Others proposed theoretical explanations and connections with the intensity of the sky light, including R. Clausius (1847, and later) who is better known for his contributions to thermodynamics.

J. Tyndall (Fig. 27-1) who has already been mentioned in sections 16.1 and 17.1, was keen on finding explanations for various physical phenomena, anywhere between heaven and Earth. Among other things he published a lecture that he had given on Icelandic hot springs (Tyndall 1853); as I am not aware that he visited this country himself, the lecture probably is based on observations by people like G.S. Mackenzie, R. Bunsen and H. Holland. It may be pointed out here that Tyndall describes the water in the great Geysir's bowl as being "the purest azure" in color; in fact the hot springs probably had inspired Bunsen (1849) to be the first scientist to study the color of water. One topic of interest to Tyndall (1869a,b, 1870) concerned the interaction of light with the molecules of the atmosphere, including those of water vapor and other minor constituents. He produced vapor clouds from various compounds (including butyl nitrite, isopropyl iodide and other organic chemicals) as well as smoke from incense, gunpowder, tobacco, etc., in a glass tube illuminated with a beam of white light. If the particles of the vapor or smoke were sufficiently small, the light emanating sideways from them was beautifully blue like "the purest Italian sky". Using Nicol prisms and other apparatus he also made some unexpected observations, such as finding that the light from these clouds was in some cases linearly polarized (Fig. 27-1) even if the incoming beam was unpolarized. Govi (1860a) had in fact pointed this out earlier. When the particles increased in size, both the bluishness and the polarization gradually disappeared (cf. ordinary clouds). The same happened with fine dust suspended in water, but Tyndall (1869b, 1870)

does not refer to the Icelandic hot springs in that context and some (Busch and Jensen 1911, p. 109) claim that his experiments had been prompted by observations of smoke from peat fires in Ireland. The polarization was later called "the most interesting property of turbid media" (Verdet-Exner 1887, p. 404), and it is generally acknowledged (see Rev. Gén. Sci. 13, 1902, p. 59-60) that Tyndall's experiments on this aspect provided the key to solving the blue-sky mystery.

From his experiments, Tyndall concluded that the blue light from the sky was sunlight reflected off particles smaller than the wavelength of the light. J.W. Strutt (1871a; he became Lord Rayleigh in 1873, see Fig. 27-1) pointed out that this phenomenon was not comparable to ordinary reflection, and it was eventually given a special name: Rayleigh scattering. Strutt (1871a,b) made a new theoretical analysis of the probable response of such small particles to the effects of the passing light wave, obtaining the important result that the relative intensity of the scattered light increased inversely as the fourth power of its wavelength. Even though the aether theory still held the ground at that time, Strutt's result is still a valid approximation for the interaction of light with very small particles. He could also explain the polarization of the light scattered sideways, and he concluded that Fresnel's hypothesis on the vibration direction of the aether was preferable to that of Neumann. In addition to Tyndall and Strutt, J.L. Soret, A. Lallemand (1871) and E. Hagenbach were among those reporting investigations on the color of air and water around 1870. Other related phenomena were also studied: for instance, Soret (1874) demonstrated the existence of soot particles in a gas flame by means of the polarization of its light, and Tyndall himself applied his experience of scattered light to valuable work on particulate matter (such as bacteria) in the atmosphere in the 1870s. Rayleigh (1881a, 1899) showed that the polarization of light scattered from small particles could also be explained by Maxwell's electromagnetic theory. Various studies of the intensity, color and polarization of sky light were continued, e.g. by Wild (1876, Fig. 27-2) and Crova (1890), both of whom employed photometers with Nicol prisms. Hurion (1896) measured the attenuation and polarization of light in a turbid liquid with Cornu's photo-polarimeter, and Compan (1899) made similar measurements of attenuation with Crova's photometer. Compan's results agreed with the fourth-power law of Rayleigh for very small particles; for those of sizes comparable to the wavelength, a different relation proposed by R. Clausius around 1850 gave a better fit to the observations.

In the years around and soon after 1900, many scientists investigated scattered light and its polarization by laboratory measurements on gases (e.g. Cabannes 1915, who was the first to show that light was scattered by pure air), liquids (Spring 1899, Müller 1907, and others), metal particles in liquids (Ehrenhaft 1903, with Cornu's meter; Steubing 1908), and transparent solids (e.g. Siedentopf and Zsigmondy 1903, Rayleigh jr. 1919). Theoretical studies were also carried out, for instance by Mie (1908, metal particles). Many people such as Pernter (1901), Busch and Jensen (1911) and Müller (1912) measured the spectrum, polarization and attenuation of sunlight in the Earth's atmosphere. Additionally, the state of polarization of light from the Sun's corona was observed (e.g. by Wood 1900, Salet 1910) during many eclipses to 1920, in order to find out if this light was due to reflection, scattering, or fluorescence. These observations often employed Savart plates (from quartz or calcite) in addition to Nicol prisms.

The study by Cabannes just referred to, was related to suggestions in the 1910s (see Martin 1920) that scattering of light in a transparent substance such as a gas was not only caused by foreign particles (dust, etc.) or other inhomogeneities, but also by the molecules of the substance itself. It was shown that the attenuation of light due to the scattering could be used to estimate the number of molecules in a mole (i.e. Avogadro's number, see Bruhat 1942, p. 287; Partington 1953, p. 242). Some such estimates were made, with varying success, both before R.A. Millikan's measurements of the electronic charge in 1910-17 (section 36.4), and later (e.g. Cabannes 1921).

Research on scattered light also became the basis of an important method, nephelometry, for

On the Colour of Water. By Professor BUNSEN.

The hot springs which occur in many parts of Iceland, and are especially remarkable at Reykir, are, says that excellent observer Bunsen, characterised by extreme beauty. In the depths of the clear unruffled blue waters of these basins, from which rises a light vapour, the dark outlines of what once formed the mouth of a Geyser may be faintly traced amid the fantastic forms of the white stalactic walls. Nowhere can the beautiful greenish-blue tint of water be seen in greater purity than in these springs.

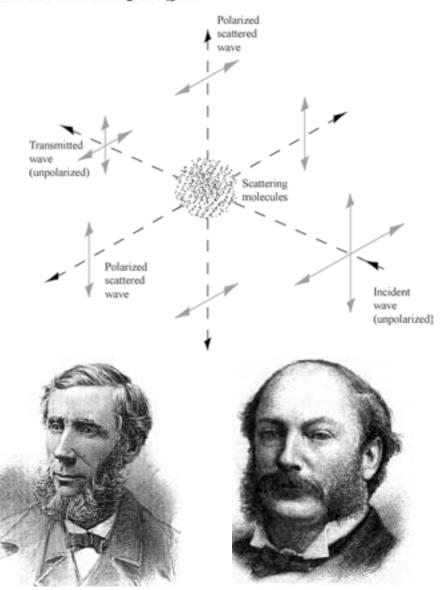


Figure 27-1. The top item is the beginning of a paper by Bunsen (1849) which among other things discusses the blue color of water in Icelandic hot springs. J. Tyndall (1820-1893, bottom left) demonstrated in 1869 that light scattered to the side from smoke or fine dust, is bluish and linearly polarized. See the central explanatory diagram which is redrawn from G.R. Fowles: Introduction to Modern Optics, 1975. The diagram is greatly simplified: the light is in fact scattered towards all possible directions. J.W. Strutt (later Lord Rayleigh, 1842-1919, bottom right) published important theoretical results on the scattering of light by small particles in 1871 and later.

studying suspensions. Here the scattered light is measured with photometers, which in a few cases contained Nicol prisms. See for instance Mecklenburg's (1914, 1915) papers on a "Tyndallmeter" with three such prisms, Wilke and Handovsky (1913) on a meter with four prisms, Weinberg (1921), and the book by Walsh (1958). This method is still used extensively in biology, chemistry, and biochemistry as well as in technology (brewing, purification of water, etc.)

Various interesting aspects of further developments regarding scattered light are treated in section 36.7.

27.2 Liquid crystals, from 1870

It should not be forgotten that natural minerals are only a small fraction of all the compounds that can crystallize. Polarized light is a convenient and efficient way of studying man-made crystals. However, only a single example of such studies is described briefly in this section, and a few others will be considered in the latter part of section 29.1.

Around 1870, scientists began to look into the physical properties of certain organic substances which do not change between liquid and solid states at a fixed temperature, but stay in an intermediate configuration over a temperature interval of the order of 10°C or more. Among these are various soapy, waxy or greasy compounds such as "soft soap" produced for household use in past times. A special field of research concerned with these materials (Houben-Weyl 1955) is often said to have been born when the physical chemist O. Lehmann (1889) became acquainted with observations on them by the botanist F. Reinitzer. For studying this class of materials, it was appropriate to use a microscope with Nicol prisms and a heated stage, which Lehmann had invented in 1877. He had also fitted such a microscope with equipment for photography and for projection on a screen (Lehmann 1886b). This research had its ups and downs to start with, and it encountered disbelief as the behavior of the materials was very odd and the microscopic specimens sometimes resembled living cells. However, Lehmann (e.g. 1900, 1908, 1911-13, Fig. 27-3) did not give up but published tens of papers on his observations up to around 1920. In the 1900 paper for instance, there are 169 color drawings of drops as they appeared in Lehmann's polarizing microscope under varying conditions; later, he took movies of them. He gradually convinced others (e.g. Schenck 1899) that he was dealing with a significant physical phenomenon, and Rinne (1922b) calls the discovery and preparation of liquid crystals "one of the finest achievements of science".

The intermediate-state materials which were given the name "liquid crystals" consist of large elongated molecules and are very anisotropic. Thousands were known already by 1940, and they have now been classified in various ways according to their internal structure. Polarized light continued to be very useful in this work (Gaubert 1905; Vorländer 1908; Wien-Harms 1928, and many others), and it still is "the most powerful tool for investigating liquid crystals, all of which exhibit characteristic optical properties" (Van Nostrand 1989). These properties include extremely strong optical activity (with rotations even exceeding 50,000°/mm!) and circular dichroism. Some liquid crystals become doubly refracting in an electric field (e.g. Björnståhl 1918) which from around 1970 has been a basis for their utilization in displays for digital watches and instruments, later in screens for television and computers.

Polarizing microscopes with a heated stage, as mentioned above, also found wide use in studying crystallization processes in general. On some such stages (Fig. 27-3), specimens could reach temperatures well above 1000°C. Microscopes of this type are e.g. described in the book by Groth (1885, p. 658-660) who considered them to be "indispensable for investigating transformations taking place in the various phases of materials".

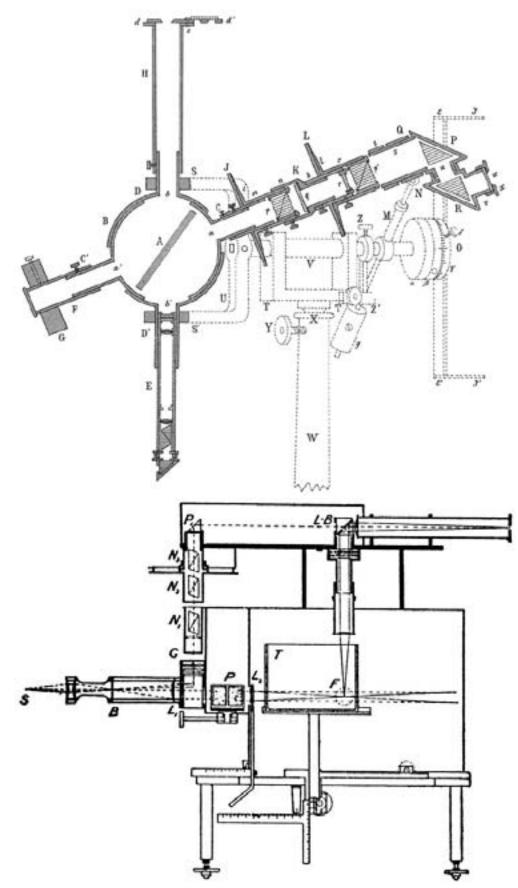


Figure 27-2. Equipment for studying scattered light. Top: The "uranophotometer" device constructed by Wild (1876) for measurements of the properties of sky light. It contains three Nicol prisms and a Savart plate of Iceland spar. Bottom: Mecklenburg's (1914) instrument with three Nicol prisms, for measuring the light scattered from colloidal matter in water in the trough T.

27.3 Photo-elasticity 1880-1920 and later; flow birefringence in liquids

The double refraction effects that can be brought about in isotropic solids by pressure, do not seem to have been investigated much for 2-3 decades following the work of Wertheim, Maxwell and others in the 1850s (section 15.3). The topic also was not given much attention in books on optics. Lepinay (1880) confirmed Wertheim's (1851) conclusion that the effect of pressure on light velocity in some (but not all) types of glass was almost independent of the wavelength of the light. Kerr (1888), Carus-Wilson (1891), Marston (1893), Pockels (1902), and W. König (1903) also carried out some work in this field. Pockels found that some of Neumann's (1843) theoretical results on the subject needed revision. Pockels (1889, 1903) also analyzed the first measurements made on refraction in crystals (quartz and Iceland spar) under pressure, using theoretical formulas for their elastic parameters. König (1901) investigated double refraction in a vibrating glass plate, for comparison with older theoretical and experimental studies. Ewell (1903) applied a twisting torque to cylinders of jelly, and studied the optical activity it produced. In this kind of research, it was eventually found that it was easier to make test objects from new transparent isotropic materials such as celluloid (from 1906) and bakelite (from about 1920) than from glass. Adams and Williamson (1919, 1920) greatly improved the annealing techniques employed in the production of strain-free optical glass.

Around 1910, engineers began to realize the potential of polarized light in studies of stress in components of buildings and machinery, using two-dimensional transparent scale models. The method came to be called photo-elasticity. As has been touched on in section 15.3, white linearly polarized light from a Nicol prism was sent through such a model and then through another Nicol prism (as in a petrographic microscope). To an observer, a pattern of two different kinds of fringes was then seen in the model. One set was composed of black-and-white "isoclinics", passing through all points where principal stresses have parallel directions. The other set was colored "isochromatics", connecting points in the material with the same maximum shear stress. The delay of one polarized component of the light relative to the other is constant along an isochromatic. If circularly polarized light is used, the former fringes disappear.

Mesnager (1913) constructed a glass model of a reinforced-concrete bridge to be built across the Rhône river, in order to visualize stresses in it by means of polarized light. This generated considerable attention (see Skinner 1923). Coker (1910, 1912, 1916, etc.) and Filon (1907, 1912, etc.) used photoelastic techniques to test general theoretical hypotheses relating to the strength of materials. They also investigated many specific cases of internal stresses that were important in engineering design, see e.g. Fig. 27-5. Their comprehensive book (Coker and Filon 1931) did much to increase the interest of scientists and engineers in the method. As stated in the book's preface: "The practical importance of photo-elasticity to the engineer can hardly be overrated. It provides him, as no other existing method does, with an immediate practical solution of fundamental problems concerning the stresses in the elements of structures and machines, which cannot be otherwise directly observed and which are usually beyond the reach of calculation.".

Some large laboratories, operated e.g. by the General Electric and Westinghouse companies as well as by universities like M.I.T. started carrying out applied research in photo-elasticity in the early 1920's (Kimball 1921, Fig. 27-5, Heymans 1922, Delanghe 1928). Delanghe describes examples of the optical technique being used in observing tensions in plates with rivet holes, in fasteners for cables, and in cogwheel teeth. In the preface to the two-volume monograph by Frocht (1941, 1948, Fig. 27-5) it is reiterated that this research has been of enormous practical value, because calculations of stress fields in objects are very complicated and people previously could only estimate the mean stress within sizable areas in these. The objects however will break at

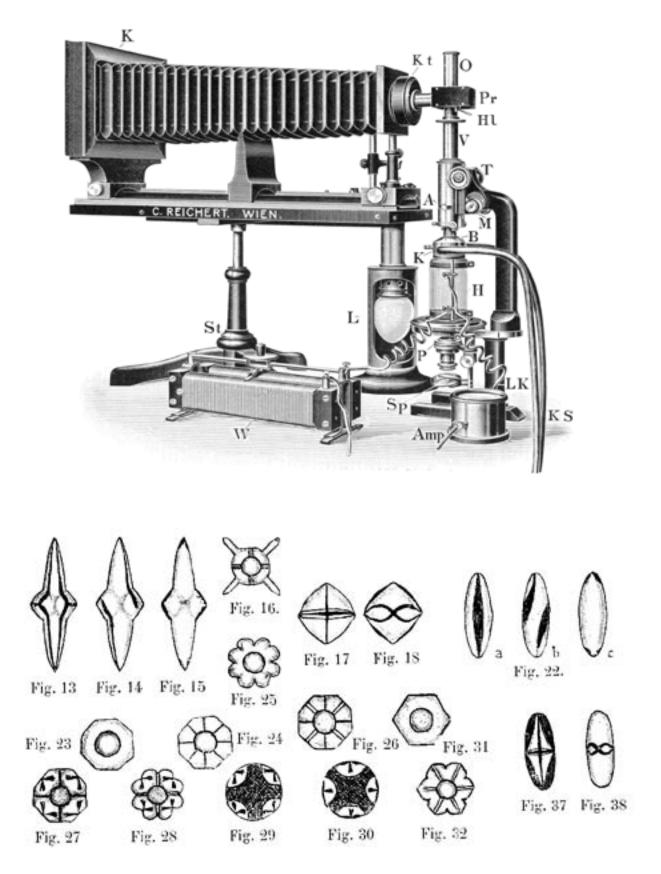


Figure 27-3. Top part: Research on the optical properties of natural and man-made minerals, liquid crystals and other materials required facilities for observing them at high and low temperatures. The occurrence of phase changes was of particular interest. Around 1900, descriptions appeared of several polarizing microscopes where the stage could be heated by a gas flame or an electrical current. The one shown here was designed by Doelter (1909), K is a camera. From Weinschenk (1925). Bottom part: Strange patterns exhibited by liquid crystal specimens, when viewed in a polarizing microscope under varying conditions. From Lehmann (1911-13), magnification not stated.

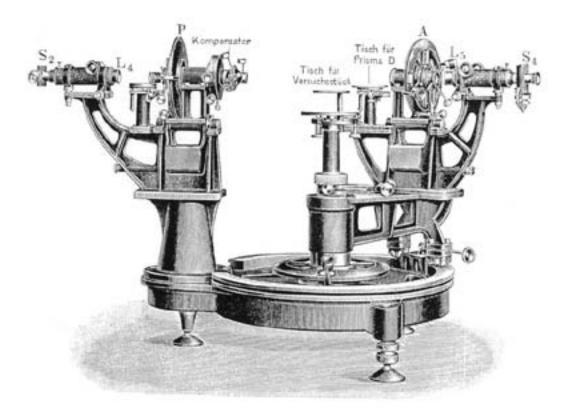


Figure 27-4. Polarized light has for more than a century had important applications in the inspection of internal stresses in glass. One instance concerns the lenses and prisms intended for use in precise instrumentation, as well as the end-plates of polarimeter sample tubes. The figure shows the testing apparatus of Schulz (1912).

points where the actual stress, not the averaged stress around them, reaches a critical value. Frocht presents an overview of the very varied research going on in photo-elasticity, and of the apparatus employed. At the time of writing of his first volume, monochromatic light was being generally preferred to white light; Nicol-prisms seem still to be widely used, but glass reflectors (Fig. 27-5) and Polaroid sheets (from 1935 or thereabout, see section 35.2) often replaced them when large pieces were being tested, because of the limited field of view of the prisms.

After 1960 or so, digital computers made it possible to calculate stress values accurately in ever more complex situations, and other equipment like resistive strain gages was brought in to check the calculations. In these cases the role of photoelastic observations on transparent models was less important than before, but new applications of the technique were also emerging. Such applications included research with movie cameras and stroboscopes on rotating or vibrating pieces, observations of shock waves, crack formation, plastic deformation, and so on (Föppl and Mönch 1959).

It was known since 1814 (see section 7.3) that when glass is cooled rapidly or unevenly from a molten state, internals stresses arise which make the glass doubly refracting. This was investigated by various scientists in the 19th century (e.g. Airy 1833a, Neumann 1843, Czapski 1891c, Rayleigh 1901) and applied in the production of so-called tempered glass (Luynes 1876). Polarized light may be used in observing these stresses or others imparted during the production of objects from raw glass (Preston 1930). As pointed out already by Brewster (1816c, p. 113-114) the presence of stress will increase the brittleness of the glass; this may be remedied by annealing, i.e. re-heating the objects to a temperature near the melting point, followed by slow cooling. Internal stresses also degrade the optical performance of glass in the lenses and prisms of microscopes, telescopes, cameras, projectors, and range finders, as well as in containers for samples that are to be analyzed by means of light, etc. Schulz (1912, Fig. 27-4), Zschimmer and Schulz (1913), Halle

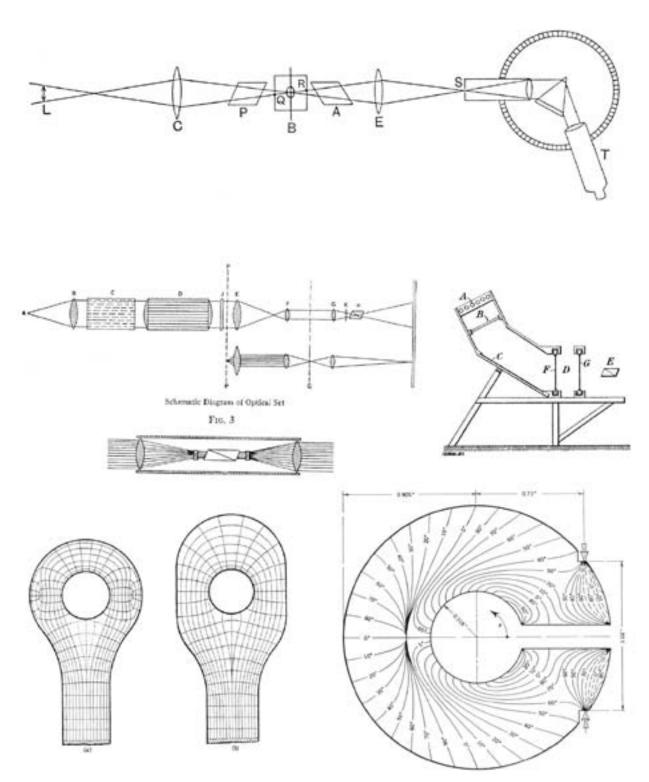


Figure 27-5. Top: A typical experimental situation from around 1905 (Coker and Filon 1931) for using monochromatic polarized light to study tensions within a small glass model B under stress. The light source is to the left, observations are made through the telescope T. Center left: In order to inspect photoelastically a large area of an object P, much light had to pass the polarizing prism which is inside a tube D, cf. inset below. In order to prevent overheating of the prism, the light beam was first filtered in the water bath C (Kimball 1921). Right: A glass mirror is preferred here to a Nicol prism for polarizing the light from the lamps A. A transparent piece for stress testing is then placed in the space D between the quarter-wave plates F and G (Coker and Thompson 1912). Bottom: The distribution of stress found from transparent models. On the left-hand side, there are results from two different eye bars for the anchoring of bridge cables. In the one on the right the stress is more even, reducing the danger of failure (Coker 1912). The right-hand diagram shows stresses (which are high where the contours are close together) in a circular washer with a slot, when squeezed together (Frocht 1941).

(1921), Wright (1921), Skinner (1923, p. 749-750), Tardy (1929) and Grabau (1938) mention that the manufacturers and users of optical equipment carry out appropriate tests on their glass components with polarized light.

The effect of mechanical stress on materials like glass is to impart a directional bias to its molecules which normally have random orientations. Each small region within these materials therefore acts upon light as a uniaxial crystal. In the year 1866 Maxwell (1873-74) was one of the first to study an analogous phenomenon in flowing or rotating liquids, called flow birefringence or the Maxwell effect. Here, shearing movements in the flow cause the liquid molecules, which are seldom spherical in shape, to align with each other to some extent. Bouasse (1925, p. 411-418, cf. Gehrcke I, 2, 1927, p. 879-880) points out that in experiments on viscous fluids with polarized light, it is easy to observe the time lag of their movement relative to the forces causing that movement. In a notable paper on the nature of gases which Maxwell also published in 1866, he introduces new concepts like "relaxation time", and mentions that the response of some materials to applied pressure or shear may show characteristics of both elastic and viscous deformation. It is possible that some of these general ideas may have originated in Maxwell's experiments on liguids with polarized light. The Maxwell effect was later investigated by Kundt (1881), his students and others, for instance Hill (1899), Metz (1902), and Zakrzewski and Kraft (1905). It turned out to be quite a complex phenomenon in many situations (Coker and Filon 1931, p. 286-288); see section 36.8 on developments after 1920.

27.4 E. Fischer's research on sugars and other compounds, and some related discoveries

The branch of organic chemistry concerned with the three-dimensional structure of molecules (in particular those forming mirror-image pairs), grew rapidly as more and more scientists agreed with the views first expressed by J.H. van't Hoff and J.A. Le Bel in 1874. At least ten books on this topic which was given the name "stereochemistry", appeared before or just after 1900 (Fig. 27-6).

One of the most productive pioneers in this field was Emil Fischer. He created a chemical foundation for biochemistry by his research on sugar compounds, enzymes, proteins and purines; to the last group belong for instance adenine and guanine which are constituents of nucleic acids in cells. Only two aldohexose sugars were well known from nature when Fischer began his studies on sugars around 1887, namely glucose and galactose, cf. sections 12.4 and 13.2. Both rotated polarized light to the right. He realized that there must be eight different left/right pairs of aldohexoses. With an ingenious series of experiments which were partly based on prior findings by others, he managed to determine the structure of the above two sugars and to synthesize them as well as most of the other fourteen (Fig. 27-7). Polarimetric measurements were essential to his success, see for instance Fischer (1890a,b,c,d,1894a). As an example, one of his new sugars which he called mannose and separated into its two stereo-isomeric forms by means of micro-organisms, was later found in juices and seeds from various plants. He also synthesized the ketohexoses fructose and sorbose, some disaccharides (the first one being isomaltose, see Fischer 1895), other types of sugars and many compounds derived from these. Research by other chemists along similar lines concerned among other things the effects of acids and bases on sugars. Thus, Bruyn and Ekenstein (1895, and later) showed that in an alkaline solution, glucose may be partly transformed into fructose and mannose; such investigations were extended around the turn of the century by J.U. Nef (1907, and later), O. Ruff, and C. Neuberg, to name a few.

Fischer studied the relations between the hexoses and pentoses (i.e. sugars with five C-atoms),

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Dr. H. LANDOLT

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Dr. O. SCHÖNROCK, Dr. P. LINDXER, Dr. F. SCHÜTT, Dr. L. REENDT, Dr. T. POSNER

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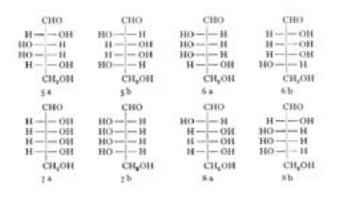
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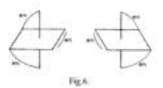
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Jena. Verlag von Gustav Flacher. 1904.

Figure 27-6. Intensive research on optical activity of organic substances around 1900 is reflected in the book by Landolt (1898) which contains detailed data on some 700 pure compounds. Many books on stereochemistry in general were also published, e.g. by Werner (1904).





In this case, the asymmetrical structure of the molecules is thus wholly dictated by the spatial arrangement of the groups, and is entirely independenof the nature of the groups linked together. It therefore appeared to be particularly important for our conceptions of the origin of the optical activity of chemical molecules to establish whether such asymmetrical molecular structure also results in optical activity. This is in fact the case. In four different metals we succeeded in resolving compounds of this kind into optical isomers, i.e. in the case of cobalt, chromium, rhodium, and iron. For the first three metals we obtained in optically active form the triethylene-diamine compounds:

[Coen,] X,[Cren,] X,[Rhen,] X,

Figure 27-7. Left: Four out of eight mirror-image pairs of aldohexose sugars. E. Fischer was awarded the 1902 Nobel prize in Chemistry for his fundamental research on the structure of carbohydrate molecules. Right: A paragraph from the speech by A. Werner on the occasion of receiving the 1913 prize (www.nobelprize.org), where he describes the importance of optical activity measurements in the confirmation of his theories about chemical bonds.

and he prepared in 1891 new pentoses in addition to the two (xylose and arabinose) that had previously been obtained from natural sources. One of the new sugars was called ribose (Fischer and Piloty 1891). A. Kossel then found in 1893 that the nucleic acids of cells contained a carbohydrate which he considered to be a pentose sugar. See more on this important matter towards the end of section 29.2; other people continued Fischer's research on the various properties of the pentoses (e.g. Wohl 1899).

Glycosides are a class of chemicals consisting of a monosaccharide (commonly glucose) and some other group of atoms. Many are optically active, which has often aided researchers. Of these chemicals, salicin and coniferin have already been mentioned; other glycosides from plants include phloridzin which can induce diabetes, the heart drug digitoxin, and amygdalin. A large number of known and unknown glycosides were synthesized (see e.g. Koenigs and Knorr 1901), beginning in fact before 1880. E. Fischer developed new powerful methods for that purpose, and made several such compounds with his associates (Fischer and Raske 1909, etc.). Willstätter (1915) investigated anthocyanin pigments in plants which belong to this class. Among properties of glycosides that were studied with the aid of polarimeters, was their decomposition by acids and by enzymes (Walker and Krieble 1909, Bourquelot and Bridel 1913, and many others). Later, W.N. Haworth and his collaborators (cf. section 29.2 and Fig. 35-1) made extensive use of glycoside syntheses in their efforts to elucidate the configuration of mono-, di- and polysaccharides, see Appendix 5.

Fischer (1894b) drew an important and much-quoted conclusion from his research on the varying efficiency of enzymes from different yeast strains in the fermentation of sugar: "As an illustration, I would say that an enzyme and a glucoside must fit together like a lock and a key, in order to exert a chemical effect on each other.". A similar assertion was made by Fischer and Thierfelder (1894), stating "...that the fermentability is closely connected to the geometrical structure of molecules, and can thus be considered to be a stereochemical question.". As Fischer (1898-99) also noted, three-dimensional chemistry is a cornerstone of all biological activity. See Appendix 5 on Fischer.

In addition to the importance of E. Fischer's sugar research to biochemistry, it was of enormous value to industrial sectors connected with sugars, starch, cellulose and chemicals of that kind (see Pringsheim 1925). In the context of industry, we may also point out attempts to transform cellulose to di- and monosaccharides by means of acids (e.g. by Ost and Wilkening 1910, Willstätter and Zechmeister 1913, Zemplén 1913, Cunningham 1918, Irvine and Hirst 1922, Sherrard and Froehler 1923); such transformations could be easily followed with a polarimeter. The work of O. Wallach and collaborators mentioned in section 22.2 was another example of an industrially important research effort in a field of organic chemistry: they published at least a hundred papers on the structure of the terpene and camphor groups (alicyclic compounds, many of which are optically active, see Fig. 27-8) in the time interval from 1885 to the 1910s.

Scientists had investigated the optical activity of natural proteins since the mid-19th century, and also the 20 or so amino acids which they are composed of (e.g. Schulze and Likiernik 1893). The amino acids are all optically active (cf. the example in Fig. 16-1, bottom right) except one, i.e. lycine. Some of them (designated by d- or +) rotate light to the right, and others to the left (l- or -); however, the handedness of the molecules themselves can also be defined, a property designated as D or L. All amino acids in natural proteins turned out to be of the L-type. Fischer (e.g. 1899) made new discoveries in this field, and wrote over 30 papers on the synthesis of amino acids and proteins in 1903-09. Initially he made use of Pasteur's method of separating optically inactive mixtures of a d- and an l- acid into two active components by combining them with alkaloids. He also employed a peculiar and unexpected class of reactions discovered by P. Walden (1896, 1919; see D.S.B. and Fig. 27-8). This "Walden transformation" as it was named by Fischer (1906) makes it possible to convert in a simple way a left-handed compound into the same right-handed

compound, or vice versa. These reactions have been used and studied intensively by chemists ever since (Ramsay 1981).

In the early decades of the 20th century, polarimetric measurements often played a part in research on amino acids and proteins, regarding e.g. their sources in nature, their relationships, compounds derived from them, their breakdown products, methods of synthesis, and so on (cf. Edlbacher 1927). Only a few randomly selected examples of such research will be reported here. Ehrlich (1904) studied the properties of leucin and isoleucin (which he isolated from molasses) in detail; Locquin (1907) subsequently showed that an amino acid synthesized by himself and L. Bouveault possessed the same optical activity and other properties as Ehrlich's isoleucin. Levene and van Slyke (1909) concluded from their optical measurements of casein and other substances: "An exact analysis of the important leucin fraction of protein is thus rendered possible for the first time.". Such results no doubt were very useful in the acquisition of further knowledge regarding the role of these compounds in biochemical processes, pharmaceutics, agriculture, food science and so on. Osborne and Harris (1903) measured carefully the optical activity of at least a dozen proteins such as gliadin from plants, and in a paper on the optical activity of gliadin proteins in wheat Dill and Alsberg (1925) claim that "The properties of flour depend largely upon the proteins it contains". In a discussion of E. Fischer's research on the common amino acid cystine, Toennies and Lavine (1930) state that "The only practical quantitative criterion for the purity of l-cystine is its optical rotation.". Freudenberg and Rhino (1924) demonstrated, using a polarimeter among other techniques, that the configuration of alanine (Fig. 16-1) is identical to that of lactic acid, with an NH₂-group replacing OH. Similarly, aspartic acid was found to be analogous to malic acid (see Freudenberg and Lux 1928, Fig. 35-1; Wittig 1930, p. 66), etc.

In the so-called "racemization reaction" which commonly occurs in amino acids and related compounds (Kossel and Weiss 1909), a sample of pure d- or l- acid in solution gradually turns into an inactive mixture of both. Much research was carried out on the effects of e.g. temperature and pH upon this process, and on ways to resolve such mixtures into their active components. It may be added that after 1970, racemization has become the basis of a method to estimate the age of organic remains in geological strata.

In sections 16.2 and 22.2, three methods suggested by L. Pasteur for the separation of righthanded and left-handed molecules in racemic mixtures were described. A fourth method (Marckwald and McKenzie 1899) which depended on the difference in reaction rates between a righthanded acid and right- or left-handed bases was for a long time of theoretical interest only (Wittig 1930, p. 35). After 1900 it was shown by Neuberg (1903), Erlenmeyer and Arnold (1904) and others how dibasic acids could be utilized in the resolution of mixtures of left- and right-handed alcohols. Marckwald (1904) was the first to synthesize an optically active compound from inactive components, with the aid of an active intermediary material. Soon after, it was found that catalysts (other than enzymes) containing mirror-symmetric atoms were useful agents for such resolutions (Bredig and Fiske 1912). These methods and more recent ones have been of increasing importance in chemical and pharmaceutical industries, as manifested e.g. by the awarding of a Nobel prize in that field of chemistry in 2001.

27.5 Research on inorganic compounds by A. Werner and others

For several decades, carbon was the only element known to form mirror-image molecules. However, it appeared possible that other elements having a valency of 4 or higher could also do so. Among those looking for such molecules was a young chemist, A. Werner. Hantzsch and Werner (1890) attempted to explain certain properties of so-called azo- and oxime- compounds from the viewpoint of nitrogen being at the center of a mirror-image molecular group. Their paper was a landmark achievement regarding the three-dimensional chemistry of nitrogen.

Werner wrote a well-known book on stereochemistry (Werner 1904, Fig. 27-6) and made great strides in research on this topic in the first two decades of the 20th century. In 1893 he advanced a hypothesis on what he called "coordination" chemical bonds, and he synthesized a large number of compounds around atoms e.g. of cobalt, chromium and platinum (Werner 1893, 1906). Some of these molecules could exist in either symmetrical or mirror-image configurations. Polarimetric measurements were therefore an indispensable tool in Werner's research from 1897 (see e.g. Douglas 1994, and Fig. 27-7), as in the case of E. Fischer in the preceding section. To begin with, not all scientists took his ideas seriously (see Stewart 1919, chapter XV) but Lowry (1935, p. 72) claims that they "received immediate and almost universal recognition, as the result of the preparation of an optically active salt, the dissymmetry of which could not have been foreseen apart from the hypothesis of coordination". This was a complex cobalt compound (Werner 1911) and Werner wrote many additional papers on such substances in the next several years. Among them were chemicals which were optically active while containing no carbon. In his first paper on these, Werner (1914) states that "... the investigation encountered many difficulties, and the only feature lightening our task was the enormously large optical activity [of the material]". Some of his most important papers have been published by G.B. Kauffman (1968) in English translation; in the preface of that book, Kauffman recalls that the last-named paper "...silenced even Werner's most sceptical opponents... The last brick in the crumbling wall of separation between inorganic and organic chemistry had been razed.". See also Kauffman (1975).

Werner's work completely changed people's views of chemical bonding. "The implications of his research have been of inestimable value in biochemistry, and in analytical organic and physical chemistry, as well as in mineralogy and crystallography" (Dictionary of Scientific Biography 1980-90, abbreviated here). See Appendix 5. Examples of coordination compounds include the heme group in the blood of vertebrates and hemocyanin in invertebrates, vitamin B₁₂, chlorophyll, metal silicates in rocks, various pharmaceutical products, dyes, and catalysts. An important class called chelation compounds is used widely in chemical industries, agriculture, food production, water purification, medicine, etc. Various aspects of Werner's theories were debated into the 1930s, and these debates were often resolved by observations of optical activity in relevant compounds (see Ramsay 1981, and the entry on W.H. Mills in D.S.B.).

The first molecule that was synthesized and was proved to contain an N-atom with mirror-image symmetry, was an ammonium compound (Pope and Peachey 1899). Other chemicals having this property were soon made around N-atoms, but they all contained carbon and for a while it was thought that the carbon had some controlling influence on the symmetry aspect. Pope, his collaborators and others (see Scholtz 1907) went on to synthesize before 1910 many optically active chemicals around atoms of for instance tin, selenium, sulfur (Smiles 1900) and silicon. Phosphorus joined this group in 1911. Lowry (1935, p. 54) considers that new methods introduced by these researchers "were immediately fruitful in disclosing a new type of molecular dissymmetry, the first really new experimental development since the work of Pasteur half a century before". As regards silicon, C. Friedel and others had shown around 1865 that it is in many respects analogous to carbon, as it is tetravalent and can replace carbon in some molecules. In continuation of their studies, F.S. Kipping began research on active carbon-silicon compounds around the turn of the century (see e.g. Challenger and Kipping 1910). He and his co-workers published some fifty papers on such substances in J. Chem. Soc. to 1935. This research led later to the synthesis of so-called silicones; their structure resembles that of quartz (SiO₂) but with e.g. methyl groups partly replacing the oxygen. In a review, Kipping (1937; see also D.S.B.) did not envisage any practical uses for his siliconcontaining compounds, but important industrial applications in fact emerged by the early 1940s.

27.6 Reaction kinetics and equilibria

After the emergence of the above-mentioned "Law of mass action" on chemical equilibria, many chemists studied the rate of chemical reactions as a function of time, when the temperature or other conditions were varied. The results of such observations may reveal whether for instance a reaction is taking place in one step or in a succession of steps each at its characteristic pace. Several methods of monitoring the progress of a reaction were tested, for instance by following the color changes of the reacting mixture, but polarimetric measurements were also very suitable where some of the components were optically active. Thus, Ostwald (1884) used a Laurent polarimeter to repeat and extend Wilhelmy's (1850) observations on sugar hydrolysis (i.e. inversion) by acid (section 18.1). This was part of an important series of experiments by Ostwald in this field, which demonstrated that the catalytic effect of acids on reactions depends on the number of ions in the solution. See Appendix 5. Others carrying out similar work included Urech (1880 and later), Arrhenius (1889), and H.E. Armstrong's group (e.g. Armstrong et al. 1908, Worley 1912). Rolfe (1905, p. 254) and Dictionary of Scientific Biography (1980-90, on Arrhenius) state that in these papers equations describing the effects of temperature on reaction rates were derived. Interesting conclusions on the law of mass action were also obtained in the work of Will and Bredig (1888) on alkaloids which was mentioned in section 22.2.

The most significant process being studied from this point of view was the splitting of sucrose by the yeast enzyme invertase. Gubbe (1885) and Ost (1891) confirmed Dubrunfaut's (1849) result from section 13.2 above, that the products of the inversion consisted of glucose and fructose. They also investigated the effect of acids on the process. O'Sullivan and Tompson (1890) wrote a major and widely noted paper on the subject. Methods in research of this type were revised (e.g. by Henri 1901, Brown 1902, Armstrong 1904, Hudson 1908) after E. Buchner demonstrated in 1897-98 that the yeast cells themselves did not have to be present for the inversion to take place. See the comprehensive review paper by Caldwell (1906) on inversion experiments (where the dominant role of polarimetry is acknowledged) and related theories.

Until around 1900 the only known function of enzymes was the transformation of larger molecules to smaller ones, accompanied by uptake of water (hydrolysis). Many attempted to reverse such transformations, and an example was finally found by Hill (1898, 1903) with the aid of a polarimeter. This discovery which was confirmed by further work using polarimetry (e.g. Bayliss 1912) and other methods, was later said to have "entirely changed the conceptions of the physiology of metabolism" (cf. Kohler 1973). After 1900 various studies on how organisms utilize the energy content of glucose were also initiated, and for instance A. Harden and W.J. Young showed in 1908 that phosphate played an important part here. Like E. Buchner just mentioned and many others investigating fermentation, they mostly made use of volumetric measurements of the carbon dioxide evolved in their experiments. However, I have found papers where they apply polarimeters to some extent (e.g. Young 1909, Harden and Young 1913, Harden and Zilva 1914). See Appendix 5.

A very important step was taken with the comprehensive paper by Michaelis and Menten (1913) on fermentation of sugar, using measurements of optical activity (Fig. 27-9). They made several improvements upon earlier experimental procedures and proposed a general model describing the rate of this catalyzed reaction, in the form of a differential equation. In his book, Fundamentals of Enzyme Kinetics (1995 edition), A. Cornish-Bowden states that Michaelis and Menten may be "…regarded as the founders of modern enzymology" due to this single contribution. However, according to D.S.B. it did not gain much recognition until around 1930. More sophisticated theoretical models on catalyzed reactions appeared later, and precise measurements were often needed

in order to find out which of them was most applicable: "... due to the rapidity and great precision of the polarimetric method, it is superior to any other in following the hydrolysis of sucrose" says for instance R. Kuhn (1924, p. 252) in his treatise on fermentation processes.

As people acquired more and more experience in the production, purification, and storage of enzymes, as well as in the control of their activity, they became very important in chemical and biochemical industries. As far as sugar was concerned, the invertase enzyme mentioned above was around 1910 beginning to replace hydrochloric acid in Clerget's method of estimating its concentration in solutions (Hudson 1910a, Browne 1912). In the production of beet sugar, it was also sometimes necessary to find out how much of the optically active trisaccharide raffinose was present in samples of sucrose solutions. This was conventionally done by measuring the optical activity of the samples before and after hydrolysis by hydrochloric acid (Herzfeld 1890). Paine and Balch (1925, 1927) showed that a more accurate estimate would be obtained in three measurements, if invertase first split both these in two and another specialized enzyme then split the disaccharide of the raffinose into two monosaccharides.

27.7 The mutarotation phenomenon

Dubrunfaut (1846) noticed that changes took place with time in the optical activity of glucose solutions, without any apparent cause. Measurements showed that in aqueous solutions that were freshly prepared (or produced by inversion) the specific rotatory power of the glucose was about +110°, but it decreased over a period of hours to +52° (Fig. 27-9). Pasteur (1856) and others found that a similar change also occurred in some other sugars (e.g. Parcus and Tollens 1890, Tanret 1896). It was also observed in related compounds, all of which contained an aldehyde group. Some researchers thought that a hydrate was being formed in these reactions. However, people realized later in the century that the molecules of the above sugars etc. could in solution have two (or three) different shapes, with slightly different potential energies (see e.g. Brown and Pickering 1897). The molecules were constantly changing from one shape to another in collisions: at the start of the dissolution process all the molecules were alike, but a dynamic equilibrium was finally established when equal numbers changed from one configuration to another and v.v. per second.

Much research was carried out for decades on the kinetic properties of these processes and the effects of temperature and acidity upon them; this work was also connected to other studies on the structure of the respective molecules and on their interactions with the solvents. Polarimeters were particularly suitable for following such slow reactions, as their measurements were accurate and non-interfering. As regards glucose, Tanret (1895) discovered a new crystalline modification which had a specific rotatory power of +22.5° just after dissolution, increasing gradually to +52° (Fig. 27-9). Subsequent research showed that most of the glucose in aqueous solution is present in two ring-shaped forms (gluco-pyranose): at equilibrium, 64% is in the so-called β -form discovered by Tanret, while 36% is in the well-known other (α -)form. The temporal changes in optical activity were originally given names like multirotation or birotation, but the common term mutarotation, proposed by Lowry (1899) in an important paper on nitrocamphor, was eventually adopted. See more details in Lowry (1935, Chapter XXII). Subsequent research showed for instance, that sucrose is a combination of α -glucose and β -fructose. Much research of this type was also carried out on the mutarotation of lactose (e.g. Trey 1903), with later practical applications.

Hudson (1910b) writes of the work by Dubrunfaut and Tanret in a short review paper, that the world's chemists owe much to French science "...because more fruitful single discoveries in the chemistry of the carbohydrates have hardly been made". He concludes that "...it is even now apparent that the unfolding chemistry of the polysaccharides is to be largely a development of the

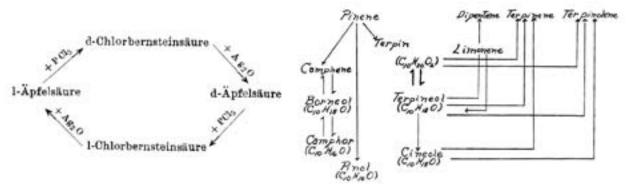


Figure 27-8. Left: One example (Walden 1919) of the Walden transformation between left-handed and righthanded versions of the same molecule. Right: A scheme indicating relations between various compounds of the terpene and camphor groups. This scheme, first published by O. Wallach in 1891, is here reproduced from a paper in J. Chemical Education 24, 1947. Several of the compounds are optically active.

mutarotation reaction.". This is so e.g. because it was found that starch is composed of glucose units in the α -configuration while cellulose consists of glucose units in the β -configuration. Two of the former can join to make maltose, while two of the latter make another disaccharide. Sk-raup and König (1901) who were among the first to study its properties by polarimetry and other means, gave it the name cellobiose.

Among chemists studying mutarotation in great detail around 1920 were Hudson (1917), Nelson and Beegle (1919), Euler et al. (1925), and Worley and Andrews (1927, 1928). Kuhn and Jacob (1924) and Kuhn (1925) investigated this process in connection with the action of β -amylase on starch. From the results of the latter paper, which is considered a classic piece of research, it could be concluded how the enzyme severs certain chemical bonds in the starch. See Appendix 5.

Bruhat (1930, p. 207) states that the mutarotation phenomenon "…has permitted us to discover a number of slow reactions which would not have been noticed otherwise, and to determine their rates and constants of equilibrium: it has played a fundamental role in establishing experimentally the laws of the chemical mechanics of solutions.". Here one may also refer to L. Onsager's speech on receiving the 1968 Nobel Prize in chemistry for discovering new laws of thermodynamics in irreversible processes. He mentions that his research in that field had in part been prompted by the precise measurements of C.N. Riiber and collaborators in the 1920s (e.g. Riiber and Minsaas 1926) on the mutarotation and concurrent changes in other properties of sugar solutions.

27.8 The photoelectic effect and related phenomena

Light affects the electrical properties of some metals and semiconductors, such as the resistivity of selenium (first described in 1873) and various metal sulfides. H. Hertz discovered in 1887 in connection with his reseach on electromagnetic waves (section 28.4) that if you illuminate a metal surface with ultraviolet light, the air in its vicinity conducts electricity better than before. This turned out to be due to the fact that the incident light ejected electrons from the metal. This "photoelectric effect", especially in the alkali metals, was widely known by 1890. Polarized light was to some extent involved in the early experiments on photoelectricity (Elster and Geitel 1894, 1895) as well as in further research on the nature of photo-sensitive materials (e.g. Drude 1898, Edmunds 1904, Kunz 1909, Elster and Geitel 1909, Duncan and Duncan 1912, Brown 1914, Frehafer 1920). This research demonstrated among other things a direct relationship between the amount of light absorbed and the photoelectric current (i.e. the number of electrons which the light removes from the metal). As will be discussed in section 36.5, the photoelectric effect was one of the phenomena which Einstein's theory of light particles could explain; however, these particles retained some of the properties of transverse waves. Thus, Elster and Geitel found that the photoelectric current could be much greater when the electrical field vector in the light (see section 28.4) was perpendicular to the surface than when parallel to it (Fig. 27-10). In the former case (called the selective photoelectric effect) one can imagine the electric field pulling the electrons out of the surface. This aspect and its relation to the presence of gases around the metal surfaces, was tested in more detail by Elster and Geitel and by others (e.g. Hughes 1916) with the aid of Nicol prisms, cf. Fig. 27-10.

Additionally, Nicol prisms were often employed in varying the light intensity when photoelectric elements were being tested and adjusted (Elster and Geitel 1912, 1913, Schulz 1913, Pohl and Pringsheim 1914 and references therein, Nathanson 1916, Kunz 1917, Ives 1924). As knowledge of these complicated electrical effects of light accumulated, such elements began to be used for astronomical measurements around 1913, but they still needed calibrating by means of Nicol prisms (Rosenberg 1921b). Photoelectric sensors soon found further practical applications, for instance in photography and printing.

27.9 On the role of polarimeters in quality control and standardization

In France, the development of polarimeters in the mid-19th century was supported by the government and by industrial associations, in order to improve the quality of raw materials and reduce adulteration of food and medicines (Billet 1858-59, II, p. 291-297). Such practices were very common, and often detrimental to the consumers' health. Legislation against them was enacted in some European countries before or around 1880, and also in some states of the U.S. Large books containing advice in the fight against adulteration were published (e.g. Chevallier-Baudrimont 1882). Polarimeters could in various cases aid in this fight, giving rapid and definite results. The sugary syrup that was produced by hydrolysis of starch from potatoes etc. in Europe from around 1820, was legitimately used in various wares such as bread or distilled spirits. However, it was considered fraud when the starch syrup was added surreptiously to the sugar syrups of commerce (Soubeiran 1850), or when sugar or starch syrup was added to honey or glycerine (Pohl 1861, König and Karsch 1895, Lehmann and Stadlinger 1907). Neubauer (1876) writes a detailed account on unwanted admixtures of starch sugars to wine and fruit juices; diluted juices and other fruit products were studied by Juckenack and Pasternack (1904). Tony-Garcin (1887) emphasizes that "The falsification of wines by sugar, glucose and dextrine... is most widespread". See also Sidersky (1895, p. 124-126). Steeg and Reuter (1888) advertise a type of polarimeter dedicated to the testing of wine, but the problem persisted for quite a while yet (Fresenius and Grünhut 1921).

Polarimeters could also detect watering-down of milk (Vernois and Becquerel 1853, p. 133-137). Costly plant oils for use in perfumes and other cosmetics, or linseed oil for the manufacture of paints, were sometimes diluted by cheaper oils (Gladstone 1864, Sidersky 1895, Piesse 1903) and even by turpentine (Buignet 1861, Maier 1865, Gerber 1896, Rolfe 1905). All this affected the optical activity of the products. One more field where polarimeters aided in quality control, concerned the purity of quinine (Wolff 1881, Koppeschaar 1885) and other alkaloids (Rolfe 1905) for pharmaceutical purposes.

Polarimeters had some connections with the development of the starch industry in Europe in the 19th century, as noted in section 12.4. In the U.S. it was found that corn (from maize) was very well suited for the production of sugar compounds with the aid of acids, and later enzymes. This industry had taken off on a large scale by the early 1870s, and the main products that were obtained

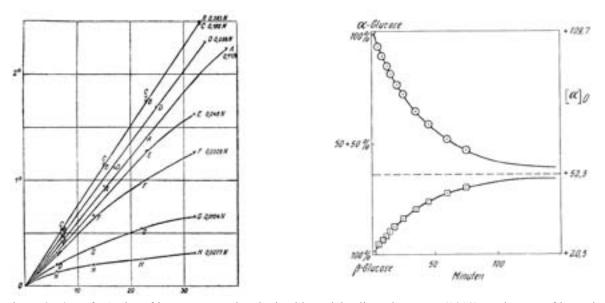


Figure 27-9. Left: A plot of important results obtained by Michaelis and Menten (1913) on the rate of inversion of sugar by enzymes. The different curves relate to different concentrations of sugar in aqueous solution. The x-axis shows time in minutes while the y-axis shows the decrease in optical rotations relative to initial values, in degrees. The diagram is taken from Kuhn (1924). Right: A graph of mutarotation, in this case the changing optical activity of glucose solutions with time after their preparation. The upper curve applies to α -glucose, i.e. the type originally known, the lower one applies to β -glucose which C. Tanret was the first to prepare in pure form in 1895. The diagram (Houben-Weyl 1955) is based on numerical data published in 1908.

from the starch were called "glucose" and "grape sugar". Both however were in fact mixtures of glucose, maltose, dextrin and water. They were not as sweet as sucrose, but cheaper and probably less unhealthy in the long term. These products soon became popular with swindlers (Higel 1883), being mixed in with household sugar, preserved fruit, jams, honey and such expensive items, often at enormous profits. Lock et al. (1882) and Browne (1912) recommend a method due to P. de P. Ricketts (1879) and C.F. Chandler for detecting admixture of grape sugar in cane sugar and various sugar products. The method employed measurements of the optical activity of a solution of the material after inversion by acids. Federal legislation on the quality of foodstuffs (The Pure Food and Drugs Act) was not passed until 1906 after a long struggle. As an example of the need for such measures, it was pointed out in the national press in 1905, that of all the "maple syrup" sold in the U.S. hardly more than 5% was actual maple syrup; the remainder tended to be corn syrup dressed up with imitation colors and flavors. The chief pioneer in this struggle was H.W. Wiley, who also happened to be one of the first Americans to become acquainted with polarimeters and to use them in research on sugar products (Wiley 1880, 1896a, and others). The above-mentioned legislation, largely drafted by him, provided for the establishment of the U.S. Food and Drug Administration. The importance of accurate measurements on sugar products was also one of the main reasons for the setting up of the National Bureau of Standards in 1901. Successes of the N.B.S. in that field during its first few years greatly enhanced its reputation (Singerman 2007).

Steady quality control and breeding experimentation on beets was an important factor in sugar production, and the effects of different growth conditions, harvesting and processing techniques also needed to be looked into in order to maximize yields in the competition with cane sugar. Growers gradually succeeded in increasing the sugar content of the beets from about 7% to 18%. Special polarimetric methods were developed (e.g. Hiltner and Thatcher 1901) in order to estimate rapidly the sugar content of the beets, even special types of saccharimeters such as that by K. Stammer from 1887, described in Landolt (1898) and Browne (1912, p. 143-145). See Fig. 29-9. The world production of beet sugar overtook that of cane sugar in 1887 (H.W. Wiley, U.S. Dept.

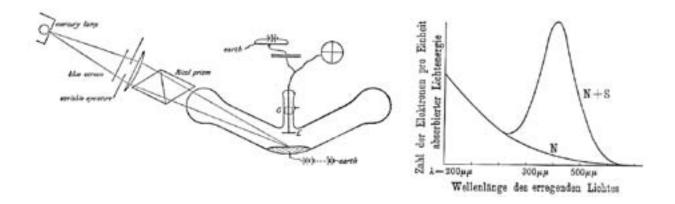


Figure 27-10. Left: A typical experiment (Hughes 1916) to examine how the magnitude of the photoelectric effect depends on the polarization of the incident light. A liquid metal (for instance, a mixture of potassium and sodium) is in the V-shaped glass bulb, and the electrical current between the metal surface and the electrode E is being measured. Right: Results from a similar experiment, for different wavelengths of the light (Pohl and Pringsheim 1914). The curve N shows the current when the electrical vector of the light is parallel to the metal surface, but an additional current S is generated when the light also has a component which is perpendicular to that surface.

Agric., Div. Chemistry, Bull. 27, 1890). The importance of polarimeters is reflected in a statement by a spokesman for the sugar industry in England, quoted in The Times on 8 Feb. 1916 as new domestically produced instruments were launched on the market: "...that the present highly-developed state of the sugar industry was to be ascribed more to the control exercised in that industry by the polarimeter than to any other cause.".

28 More research on Iceland spar, 1880-1900

28.1 Etch figures in Iceland spar and other crystals

Etching is a method in crystallographic research, first described by J.F. Daniell in 1816. Acidic or alkalic fluid is left on a polished surface of a crystal for some time, and the surface is then inspected in a microscope after the fluid has been removed. It turns out that the surfaces are not dissolved evenly in layers; rather, the fluid often leaves small pits. Such "etch figures" can be very regular in shape (Fig. 28-1), reflecting the symmetry elements of the crystal. This may help in assigning a crystal specimen to a symmetry class, which is sometimes difficult to do from its appearance, goniometric results etc. Etch studies were also connected with measurements of the hardness of crystals, and new techniques of hardness determinations were developed, see Pfaff (1883) and Fig. 28-1.

Iceland spar from Iceland did play a part in the history of these methods. Among those who wrote about etching experiments on Iceland spar before 1900 were Baumhauer (1869), Meyer (1883), Ebner (1884, 1885) and Hamberg (1890). A related field of research was concerned with the rate of etching under various conditions (e.g. Cesaro 1889, Spring 1888, 1890). Others investigated the form that an Iceland spar sphere might assume (Lösungskörper) after being immersed in an etching fluid (Lavizzari 1865, Gaubert 1901). The light spots mentioned in section 21.3 above, also investigated by Schmidt (1888), may in some cases have been related to dissolution in progress where the crystals were found.

28.2 Additional measurements of refraction in Iceland spar and other minerals, for comparison with Huygens' and Fresnel's theories

In the opinion of Hastings (1888) in the U.S., the experiments by Glazebrook (1880a, see section 21.2) confirmed the hypothesis of Huygens regarding the propagation of the extraordinary ray in Iceland spar to an accuracy of 1/10,000. Hastings himself improved Glazebrook's results in a new series of measurements on spar prisms, bringing disagreement with theory down to 1/500,000. It may be mentioned in passing that Americans were for most of the 19th century well behind Europeans in research on optics, mineralogy, and various other branches of science discussed above. In the manufacture of optical instruments they did not get much done before 1916, but at around that time they overtook Europe in matters regarding for instance photometry and illumination.

Glazebrook (1882), Basso (1883), Schrauf (1885-86), Conroy (1886), Nörrenberg (1888),

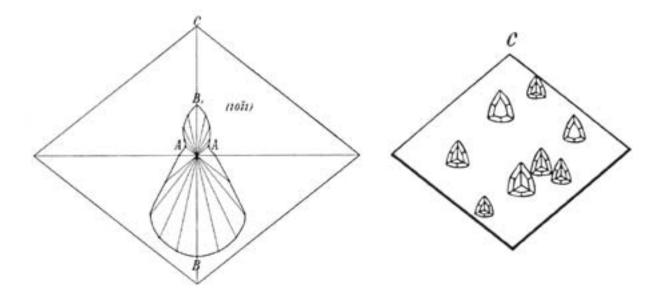


Figure 28-1. Left: Measurements of hardness on a cleavage surface of Iceland spar, made with a sklerometer. The length of each straight line is proportional to the force needed to scratch the crystal in that direction with a diamond point. The diagram is probably taken from a doctoral thesis by E. Müller in 1906 (Linck 1923). Right: A surface of Iceland spar after etching with hydrochloric acid. C is the obtuse angle of the crystal (Linck 1923).

Schmidt (1889) and Verschaffelt (1896) made measurements on the refraction and reflection of light at surfaces of Iceland spar. They improved or extended the experimental procedures formerly applied by Brewster (1819), Seebeck (1831) and Neumann (1835, 1837), and also the theoretical interpretations of Malus, Biot, and Abria (1873). P. Drude who had been investigating the reflection of light from calcite, rock salt and other crystals (Drude 1889a), disagreed with Schmidt about the effects of polishing upon the results of such measurements (Drude 1889b, see also Spurge 1887).

W. Kohlrausch (1879), Danker (1886), Pulfrich (1887) and Mülheims (1888) measured the refractive indices in Iceland spar, selenite and other crystals using total reflection. Their results agreed with formulas that Senarmont, Ketteler, and Liebisch (e.g. 1885) had derived from Fresnel's theories. Carvallo (1890) confirmed the conclusion of Swan (1849) that the ordinary index in Iceland spar was not dependent on direction.

Few other crystalline materials were investigated in any detail after 1880 with respect to the theories of Huygens and Fresnel on angles of incidence and refraction. However, McConnel (1886) studied refraction in quartz, and Brunhes (1893) measured with thoroughness internal reflections in quartz and Iceland spar. Brunhes' results were in agreement with formulas from J. MacCullagh which have been mentioned above. Born (1887) made a limited study on dolomite and anglesite, for comparison with theoretical results by Liebisch (1886). Additional studies of this kind after 1900 are reviewed in Handbuch der Physik 20, p. 650, 1928; for instance, Nakamura (1903) found that the hypotheses of Huygens and Fresnel are valid in tourmaline in spite of its strong absorption.

As regards the reflection of polarized light from plane surfaces of isotropic insulators, Fresnel's formulas for the intensity of the reflected proportion could be considered to have been confirmed in various indirect ways long before 1890. This intensity had also been measured by Glan (1874) as mentioned in sections 18.4 and 27.8, as well as by H. Knoblauch and others in the case of infrared radiation. More precise measurements were carried out for instance by Glan (1893) with a polarizing photometer, and later with other photometers (and Nicol prisms for polarizing) by Rayleigh, Murphy (1896), see Fig. 28-2, and Lummer (1909).

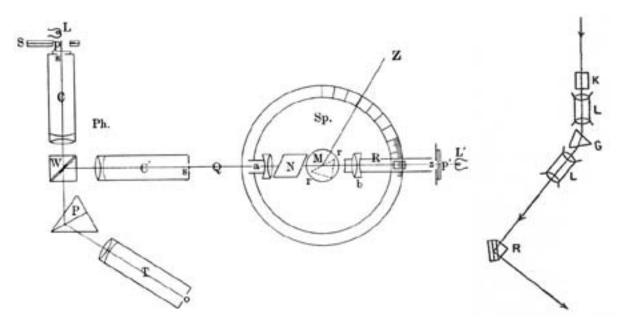


Figure 28-2. Left: An experimental test of Fresnel's laws of reflection from a glass surface M, made by Murphy (1896). N is a Nicol prism. The intensity of the light is measured on the left with a Lummer-Brodhun photometer, a chief rival of polarization photometers. Right: Part of Wiener's (1890) setup to study the reflection of light from a gelatine-coated metal surface at R. It provided solid confirmation of one aspect of J.C. Maxwell's electromagnetic theory of light. K is a calcite rhomb.

28.3 Dispersion in Iceland spar, and other properties

Sarasin (1882) measured the refractive indices of Iceland spar for visible and ultraviolet light. So did also Dufet (1891, 1893) who pointed out error sources in previous investigations, and compared the dispersion with various theoretical formulas. Their work was repeated by Carvallo (1900) over a broad range of wavelengths. Gifford (1902) measured the refraction in Iceland spar and other materials at various wavelengths to 7 decimal places; however, probably only 5 are significant (Hintze 1930). Vogel (1885), Offret (1890) and Reed (1898) charted the changes in the indices in Iceland spar as functions of temperature. Benoit (1888) made measurements of the expansion coefficients of Iceland spar over a wide temperature interval with much better precision than in the original interferometric work of Fizeau (section 21.2); these results were very useful later when Iceland spar was being employed in X-ray spectrometry (sections 36.3-36.5).

As interest in research on and with infrared radiation increased, it became necessary to measure its attenuation in Iceland spar. See papers by Nichols and Snow (1892), Merritt (1894-95), Königsberger (1897), and Stewart (1897) who used accurate measurements on this attenuation to differentiate between three theories on light absorption in crystals in general. Ultraviolet light will be treated in sections 29.8 and 34.2. The fluorescence of Iceland spar was studied by Lommel (1884), Sohncke (1897) and Schincaglia (1899), among others. Chauvin (1889) was the first to measure the Faraday effect in Iceland spar.

In chapter 15 it was mentioned that Plücker and Beer as well as Tyndall and Knoblauch had shown experimentally around 1850 that the magnetic properties of Iceland spar and some other crystals were anisotropic. Their results however only gave the ratio of the magnetic coefficients in question, but not their absolute values. Rowland and Jacques (1879) attempted measuring these values in many diamagnetic materials, but because of ferromagnetic impurities their attempts succeeded only in the case of bismuth and calcite. Voigt and Kinoshita (1907; Voigt 1910, p. 494) say that due to other flaws in the study of Rowland and Jacques, the first significant measurement of

the diamagnetic coefficients of any crystal was performed by Stenger (1883) on spar from Iceland. Knowledge of these parameters, however, did not help in understanding the structure of crystals until after 1930.

28.4 The discussion on the aether and Maxwell's theories, to the 1890s

J.C. Maxwell died in 1879, but physicists took his theories more and more into account in optics as supporting evidence accumulated. See Kirchhoff (1876), Lorentz (1877, 1878), J.J. Thomson (1880), FitzGerald (1881), Rayleigh (1881a), Glazebrook (1882, 1885, 1888), Gibbs (1882, 1888, 1889) and Ketteler (1893). FitzGerald demonstrated that from the electromagnetic equations one could reach the same conclusions on reflection and refraction at boundaries, both of isotropic and anisotropic materials, as from the elastic-aether theories; this had not been done by Maxwell himself. FitzGerald also considered that the Kerr electro-optic effect could be understood on the basis of Maxwell's theories. His paper concludes: " ... the foundation has certainly been laid of a very great addition to our knowledge, and if it induced us to emancipate our minds from the thraldom of a material aether it might possibly lead to most important results in the theoretical explanation of nature.". Not everyone was willing to discard the aether, however, for instance W. Thomson (1888, 1890; he became Lord Kelvin in 1892). He improved upon old suggestions of J. Mac-Cullagh about it possessing certain rotational properties, thus eliminating one problem that had plagued applications of Fresnel's hypotheses. Glazebrook (1888, 1889) applied Thomson's ideas to double refraction and various other optical phenomena, without mentioning those of Maxwell. Larmor (1894) considered Maxwell's theories to be still somewhat defective.

The experiments of H. Hertz (1888) on electrical waves (as they were called) convinced many scientists of the validity of Maxwell's views. Hertz' waves had much greater wavelengths than light, of the order of meters, but he could show that they had comparable properties with respect to reflection, refraction in insulating materials, and polarization. Further observations in the following years, often involving polarized waves, provided additional support for the electromagnetic character of light. In particular, experiments by Wiener (1890) attracted much attention. In these, monochromatic light was reflected (both normally and at 45° angle) from a metal surface covered by a thin layer of gel containing photosensitive silver halide (Fig. 28-2). Observations with polarized light (produced with an Iceland spar rhomb) were an important part of these experiments. Wiener could demonstrate among other things that the electrical field of a light wave oscillated in the direction which Fresnel had considered to be the vibration direction of the aether several decades previously. The light formed standing waves in the gel, and dark stripes appeared in it where the amplitude of the electrical field was greatest: it is therefore the electrical field of the light wave, rather than its magnetic field, which is mostly responsible for its chemical action. Wiener also showed that the electical field component parallel to the metal surface was zero, in agreement with Maxwell's theories and with the observations by Hertz on low-frequency waves. According to Winkelmann (1906, p. 1025), that type of interference had not been observed before with light waves. Drude and Nernst (1891) repeated Wiener's experiments using a 3-cm plate of Doppelspat and emulsions containing fluorescent compounds instead of his photographic ones, obtaining similar results. This work encouraged a variety of other research on varying electrical fields; see for instance the experiments of Righi (1893) on the polarization of electromagnetic waves. The above and other discoveries also provided a basis for the first practical method of color photography, for which G. Lippmann was awarded the Nobel prize in physics in 1908.

A good overview of the state of theoretical optics appeared in the book by H. Poincaré (1889, 1892). A large part of that book is dedicated to polarized light, and Poincaré proposed a new

method of describing its state of polarization. This "Poincaré sphere" is still in use. Around 1890, Hertz, Lorentz, O. Heaviside and others wrote many papers explaining Maxwell's electromagnetic theory in more understandable terms than Maxwell had done. The discussion on the behavior of light in crystals and other materials continued, see e.g. Rayleigh (1888, reflection from twinning planes), Fletcher (1891), Voigt (1891), Basset (1892) and Ketteler (1895). As the theory gained ground, it was applied to new situations such as the propagation of waves subject to various boundary conditions, diffraction at sharp edges (Sommerfeld 1896), dispersion and absorption (Helmholtz 1892), and so on.

Finally it may be recalled that in his work on light in biaxial crystals, Fresnel (1822c) derived equations for what he called "the wave surface". It turned out to have various interesting geometrical properties, which were studied by tens of physicists and mathematicians in the 19th century. Among those that have not been already mentioned above, are well-known names like A. Cayley, W.R. Hamilton, and S. Kowalewski. These theoretical contributions were reviewed by Wölffing (1902).

29 On the main types of instruments employing polarized light, ca. 1890-1930

At this point in the story, a wide range of scientific fields had begun to make use of equipment with Nicol prisms. Comprehensive knowledge of these applications can be obtained through perusal of a large number of scientific journals and books from the period 1890-1950 or so. However, few of these are available in Iceland. In the following sections it will be attempted to illustrate the variety of instruments available, and in the next chapter some new research topics will be introduced. One field of research of which I have little information but where it was undoubtedly very advantageous to employ polarized light, was the determination of pressure-dependent phase diagrams and equations of state for minerals and other crystalline materials. See an example regarding some bromine compounds in Wahl (1903), where references to previous work may also be found. Another research topic concerns the crystallization of water under various conditions, for instance in glaciers (e.g. Hagenbach 1890, Quincke 1905). A field which is only mentioned briefly in the next section, is the overall development of earth science and exploitation of mineral resources; a thorough survey of the role played by polarizing microscopes and specialized petrographic devices within this field alone in the above period might well fill an entire book.

29.1 Petrographic microscopes

Many scholarly books on mineralogy and crystallography appeared in 1890-1930. Among these were works by Michel-Lévy and Lacroix (1888), Groth (1905), Wallerant (1909), Rinne (1922a,b) and Friedel (1926). New editions appeared, even repeatedly, of popular textbooks or monographs by for instance H. Rosenbusch, G. Tschermak, C.F. Naumann, F. Rutley and E.S. Dana. Books describing the petrography of individual countries or regions were also published, e.g. by Teall (1888) on British rocks. These works reflect rapid developments in petrographic microscopes, techniques of observation and measurement, and consequent accumulation of knowledge, see Figs. 29-1, 29-2 and 29-3. Weinschenk (1901 and later), Duparc and Pearce (1907), Wright (1911b), Johannsen (1914), Larsen (1921) and others wrote handbooks on the use of the microscopes. Weinschenk claims in an introduction to his book, that the enormous progress made in the science of petrography (Gesteinskunde) during the last one-third of the 19th century was solely due to the application of microscopic methods. Similarly, Bonney (1892) writes that the microscopic observations clarified many aspects of the genesis of rock formations worldwide, and of their subsequent alteration. These had previously been afflicted with various incorrect interpretations: "The microscope has dispelled many an illusion, and reduced a chaos to order.". Morozewicz (1898) estimated that at least a third of the currently appearing literature in geology concerns microscopical petrography,

which had progressed very rapidly. However, the classification and naming of rock types still varied a great deal between countries at the turn of the century (Hobbs 1900). In the preface to his book on a practical application of such knowledge, Berg (1915) states that "...today, hardly any major effort in ore prospecting takes place without the broad involvement of microscopical petrography, which is particularly important in determining the genesis of the deposit.".

Among steps of progress in technique was the construction (Dick 1889, Fig. 22-5) of a microscope where the polarizer and analyzer are rotated in unison. This method (which was more practical than rotating the stage with the thin-section specimen) was soon improved upon by others such as Sommerfeldt (1904, 1905, Fig. 29-1) and Leiss (1910). Fedorow (1891) invented the so-called universal- or theodolite-stage which could be rotated about any axis; this was also improved later by F.E. Wright and others (see e.g. Leiss 1912; Fig. 29-2). As an example of the use of this stage, the directions of the optical axes in many quartz grains in a thin section could be charted; their alignment might be due to tectonic deformation of the rock. Another clue to its value is provided by C.W. Correns in a 1957 centenary Festschrift for the instrument firm of R. Winkel: "… the study of characteristic minerals which were of importance for the oil industry was much advanced by the development of the universal stage.".

Fuess (1891, 1896), Leiss (1897b, 1898a,b, 1899b, 1908 and more, Figs. 29-3, 29-5), Hirschwald (1904) and Wright (1910, 1911a, Fig. 29-2) presented many novel instruments from Fuess' workshop. Czapski (1891a, Fig. 29-1) and Lincio (1907, Fig. 29-5) introduced new microscopes from Zeiss and Leitz respectively. In order to reduce costs, some attempts were made at using glassplate reflectors (Fig. 33-2) as polarizers instead of Nicol prisms, but Leiss (1897c) and Cheshire (1923) consider this to be unacceptable in professional microscopes. Halle (1896, 1908) described improved methods in the construction of the various types of Nicol prisms, and Thompson (1905) published a comprehensive review of new or modified types; another new microscope prism patented in 1910 was the Frank-Ritter prism (see Wülfing 1918). More on the development of polarizing prisms may be found in Bennett and Bennett (1978).

Various improvements were made on Bravais' (1855) half-shade technique for measuring elliptically polarized light, for instance by Nakamura (1905) and Lummer and Kynast (1907). Berek (1913b, Fig. 29-10) described a new type of compensator for petrographic microscopes, where a plate of Iceland spar (about 0.1 mm thick) or another suitable crystal may be rotated in order to change the difference in phase between two polarized rays continuously over wide limits. This type gained much popularity (cf. Ehringhaus 1940, Schumann and Piller 1950, Partington 1953, and a review in Handbuch der Physik, vol. 25/I, 1961) and it is still in use. Berek (1915-20), Ehringhaus (e.g. 1920) and others introduced further improvements in polarizing microscopes. Many scientists devised stage heaters in order to be able to observe melting points and other changes in minerals and industrial raw materials, even at temperatures up to 1500°C (e.g. Sommerfeldt 1904, Doelter 1909, Fig. 27-3, Wright 1913a, Endell 1921).

Development of specialized instruments for the measurement of refractive indices and axial angles of crystals continued, see e.g. Czapski (1891b, Fig. 29-5), Friedel (1893), Viola (1897, Fig. 29-5), Wallerant (1897, Fig. 29-3), Wülfing (1898), Stöber (1898), Leiss (1899a,b), Klein (1900, 1902, Fig. 29-5) and Wright (1908a, Fig. 29-7). New demonstration equipment (such as projectors) for mineralogical education was also constructed, by Pellin (1897), Leiss (1897-98, 1903, Fig. 29-6), Brauns (1903b) and later by G. Halle (1910), Berek (1913a, Fig. 29-6) and Tutton (1922, Fig. 29-6), among others. Some manufacturers offered simple types of microscopes for student use (Fig. 29-7). Spectrometers allowing polarization analysis by means of Nicol prisms were produced (Leiss 1905, Fuess ca. 1910, Fig. 29-4), as well as apparatus for microscopical examination of crystals in monochromatic light (Fig. 29-6).



Figure 29-1. Polarizing microscopes around 1900. Top left: The microscope of Sommerfeldt (1905), one of the first where the polarizer and the analyzer were connected with a bar. The observer could then turn them both by the same amount, which was often an advantage. Right: A microscope designed by V. de Souza-Brandao in 1903 and manufactured by R. Fuess from 1910. The figure is taken from Johannsen (1914). Bottom left: A microscope produced by Société Genevoise (Duparc and Pearce 1907). Right: A microscope from C. Zeiss (Czapski 1891a).

Phase changes and chemical equilibria at high temperatures between the main components of common minerals were charted by the use of heated microscope stages (e.g. Fenner 1912, Andersen 1915, Rankin and Wright 1915, Rankin and Merwin 1918, Ferguson and Merwin 1919). It should also be kept in mind here that scientists are interested in many crystalline materials in addition to the minerals found in nature. Thus, the making of cement for building purposes was known by the ancient Romans, but then largely forgotten until the mid-18th century. Late in the 19th century, H. Le Chatelier (in a doctoral thesis in 1887) and A.E. Törnebohm were among the first to investigate its properties. I have not been able to consult their original writings, but the journal Moniteur Scientifique (vol. 18(i), p. 294, 1904) refers to an 1895 source stating that Le Chatelier employed a polarizing microscope to distinguish the two chief constituents of cement clinker. In 1906, an extensive study of crystallization processes and equilibria in composites of calcium-, aluminium-, and silicon oxides was initiated (Shepherd and Rankin 1909, and other papers), leading in particular to improved understanding of the properties of Portland cement. See a relatively simple stability-field diagram for two mineral constituents in Fig. 29-7. Much of the knowledge which was acquired in such studies by means of polarizing microscopes, would not have been obtained by any other experimental methods available at that time (Rankin 1916, Wright 1916). For a list of references on subsequent research in this field, see the paper by Tippmann (1931) who himself used Icelandic calcite as raw material in his cement mixtures.

Peck (1919), Endell (1921), Fisk (1934), Bayley (1937) and Hartshorne and Stuart (1950, p. 434-439) state that in the 1920s and 1930s important research similar to the above, was carried out on the composition and thermodynamics of metal oxides and carbonates in ceramics, industrial slags and the like. In the same way, knowledge was improved of the manufacturing processes of glass (e.g. Bowen 1918, Wright 1921), tiles, clays, and refractory materials such as silicon and zircon oxides used for instance in the lining of furnaces and in high-tension insulators. Milligan (1927) has this to say on abrasive materials, which are used in a wide range of activities from dental repairs to the automotive industry: "The petrographic microscope and microscopic methods in general have proved invaluable tools for the study of abrasives and grinding wheels.". Such abrasive materials include aluminium oxide (i.e. emery and other modifications) originally recovered by mining but later man-made, and the much harder carborundum (SiC) which was produced in electrical furnaces from around 1890. After 1930 or so, X-ray methods were also employed in the analysis of these compounds.

The above comments on cements, abrasives, etc. may apply equally to zeolites, as microscope research on their structure has contributed to the creation of many new compounds in that class not found in nature. They have found a wide range of industrial applications.

Polarizing microscopes were directly or indirectly connected with progress in various other sectors of materials science in the early 20th century. Liquid crystals have already been mentioned in section 27.2; see a lengthy review of further research on these by Friedel (1922). Two other examples of materials with special properties may be mentioned here; in both cases, investigations on these properties began before 1940, and they became very important in electronics and communications technology in the second half of the century. One case concerns ferrite magnets which were discovered during investigations of crystals related to the mineral spinel (MgAl₂O₄) and to metal silicates of the garnet group. The other case where polarizing microscopes were involved, concerns the development of new crystalline materials exhibiting large piezoelectric effects along with electrical properties analogous to ferromagnetism. These ferroelectric materials (such as BaTiO₃) are for instance used in electrical capacitors and in computer memories.

29.2 Polarimeters 1900-30 and later

Many books and papers were written about polarimetric practice in the above period, e.g. by Rolfe (1905), Bates (1908), Zehnder (1908a), Schulz and Gleichen (1919), Skinner (1923), and Bruhat (1930). In a review of Rolfe's book in the Journal of the American Chemical Society it is stated that: "At the present day no laboratory for ordinary research, commercial business, or teaching is considered complete without a good polariscope.". The more advanced types of polarimeters for analyzing elliptically polarized light (Fig. 29-4), contained wedges or plates from quartz or mica (see Brace 1904a, Wright 1908b, Tool 1910, Szivessy 1921, Skinner 1925). At the other end of the price scale were simple polarimeters for teaching purposes, made e.g. by W. Wilson (see figure in Houstoun 1927, p. 218; Kristjansson 1996).

In section 22.2 a half-shade technique was mentioned, dating from 1885 and involving a small extra Nicol prism (Fig. 29-8). The firm of Schmidt & Haensch became one of the largest producer of polarimeters and saccharimeters of that type towards the end of the century, and made various alterations in their design. For instance, precision instruments were offered from around 1894 where a second extra prism caused the observer's circular field of view to be split into three parts (Lippich 1896). There was even a version with a third extra prism (Lummer 1896). The firm also initiated production of saccharimeters with a double compensating quartz wedge which were easier to use than previous models (Landolt 1898, Martens 1900b). Similar saccharimeters designed by J. Peters (1894) and produced by a company of the same name, were shown at the 1904 World Exhibition in St. Louis and sold in some quantity in the U.S. around that time (Rolfe 1905, Browne 1912, Fig. 29-9). In the foreword to his book on the optical activity of organic compounds, Landolt (1898) is of the opinion that "...polarimeters are now among the most precise physical measuring instruments.". The quality of the measurement results, as well as the possibility of measuring the optical activity at discrete wavelengths other than that of the yellow sodium lines, was however limited by various factors such as access to suitable light sources. Only after the turn of the century did practical mercury vapor lamps for polarimeters become available (Armstrong et al. 1908) and later other types (Lowry 1913b), gradually replacing sodium lamps.

As already stated, saccharimeters were often calibrated for direct reading in terms of the concentration of an aqueous sugar solution, using wedges and plates from quartz. International committees dealt with the standardization of such calibrations and of other sugar measurements in the years 1894-1912 (Schulz and Gleichen 1919, Bruhat 1930, p. 279-281, Bates et al. 1942, p. 767-779). Increasing demands were made regarding the precision of the results (see e.g. Pellat 1901), because large amounts of money were involved in the production, trade, and taxation of sugar. Competent official institutions in each country were responsible for issuing calibration certificates for the quartz components (Brodhun 1913), but it was quite difficult to find natural crystals of sufficient size and quality for use in saccharimeters (Soleil 1866, Gumlich 1896, Stanley 1919). Bruhat (1930, p. 111) remarks that perhaps one out of every fifty perfectly transparent quartz specimens actually turned out to be adequate as raw material for high-quality polarimeter components.

At the time when the U.S. entered World War I in 1916, almost all polarimeters in that country were of German or Austro-Hungarian origin (Browne 1918). Replacements and spares could not be obtained from workshops in these countries, and due to the military situation manufacturers in France were unable to supply any polarimeters, "...the shortage of which is becoming at present a serious detriment in many industries.". Browne calls for the initiation of polarimeter production in the U.S. as soon as possible. Skinner (1923) reveals that the Bureau of Standards did in fact undertake the maintenance of existing instruments in the late war years; without this service "it is

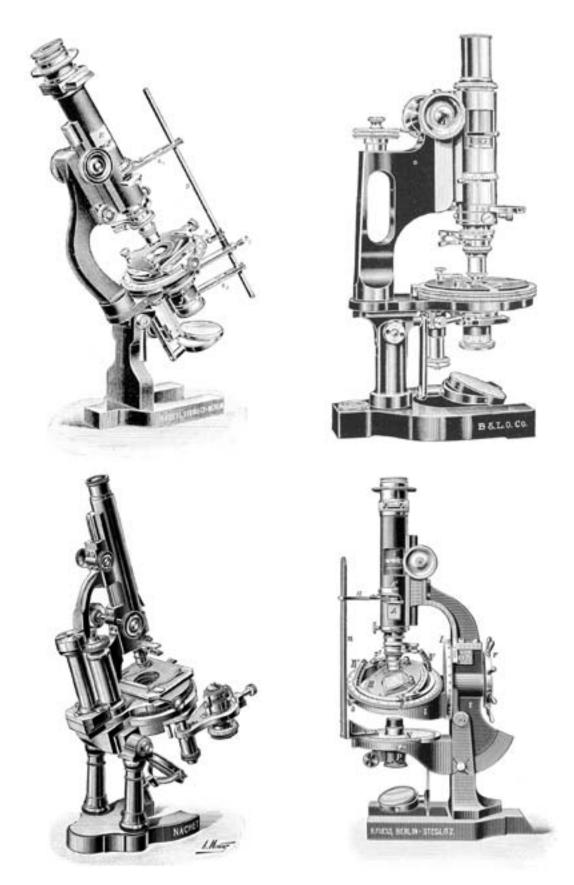


Figure 29-2. Microscopes available around 1900 or soon after. Top left: A microscope designed by Wright (1911a) and manufactured by R. Fuess. Right: A Bausch & Lomb microscope, the figure is from Johannsen (1914). Bottom left: A "grand modèle" offered by A. Nachet's workshop from around 1898. The figure is from their catalog (Nachet 1979). Right: Microscope with a universal stage, invented by Fedorow (1891) and produced by R. Fuess from about 1912 (Weinschenk 1925).

difficult to estimate what would have been the consequences of the resultant paralysis to the [U.S. sugar] industry.".

By leafing through the index pages of general chemistry journals such as that of the Chemical Society of London in the first quarter of the 20th century, it may be estimated that up to a tenth of the papers published deal with some studies on optically active compounds. The great impact of research on the three-dimensional structure of organic and later also inorganic molecules where polarimeters played a part, is evident for instance in the book by Stewart (1919) and in the two review papers by Walden (1925a,b). In concluding the former paper, Walden points out that optically active natural compounds such as terpenes and camphors, alkaloids, proteins, sugars, starch, polyamyloses, cellulose, chlorophyll, tannins, etc. are "at the focus of current interest in organic and biological chemistry, and provide major challenges in the ongoing very lively and admirable chemical research activities". These materials were obtained largely from parts of plants and trees, but also from e.g. algae, fungi, insects, or organs of higher animals.

Concerning the sugars mentioned by Walden, one can in particular point to the many scores of papers published by W.N. Haworth and his collaborators. In their work, polarimeters were valuable in ascertaining the atomic configuration of sugars and related hydrocarbons, see Haworth and Law (1916), Haworth (1920, 1932), Haworth et al. (1923 on raffinose), Haworth and Hirst (1921 on fructose, 1926 on cellobiose, 1930 on sugars in general) etc. Among these compounds was ascorbic acid (Vitamin C, $C_6H_8O_6$); optical activity was one of the properties monitored when the acid was being isolated from plants and animal tissues (Waugh and King 1932, Svirbely and Szent-György 1933) and subsequently synthesized from simpler molecules (Ault et al. 1933). See Appendix 5.

As in the earlier cases of M. Berthelot, E. Fischer and others, polarimeters aided in the investigation of novel species of sugars and related biological compounds, as well as when previously described sugars were discovered in new species of plants or animals (e.g. Wiley 1891, Forge and Hudson 1917). Attempts aided by polarimetry around 1912 to produce glucose from cellulose have also been mentioned (section 27.4). With regard to the food industry, one may quote Baumann and Grossfeld (1917): "Polarimetry provides the easiest and safest determination of starch, the most important and widely distributed carbohydrate from the vegetable kingdom in the nutrition of humans and animals.". Some of the methods developed for that purpose (e.g. by Lintner 1907, Ewers 1908, Mannich and Lenz 1920) are still in use, see for instance the regulations of the European Union (1999) pertaining to measurements of the starch content in various types of animal feedstuffs. Polarimeters could also be applied to the determination of organic acids in fruit products (Willaman 1918, Auerbach and Krüger 1923a,b).

Many scientists employed polarimeters in studies on the fermentation of sugar, in continuation of those mentioned in section 27.6. Their studies involved among other things the influence of various conditions (such as the concentrations of materials used, temperature, acidity, and the presence of heavy metal ions, salts or other admixtures), upon the reactions. The scientists carrying out this work included H. Euler (for at least 15 years, e.g. Euler and Ugglas 1910, Euler and Laurin 1920, Euler and Myrbäck 1923) and R. Willstätter, see Appendix 5, as well as J.H. Aberson, E. Schulze, F. Musculus, H. Colin, J.M. Nelson, C.S. Hudson (1910a, etc.), O. Meyerhof, and H.C. Sherman, along with numerous collaborators. In addition to important investigations on sucrose (see e.g. Willstätter and Kuhn 1923), polarimeters were useful in investigations of the breakdown of starch (Pringsheim 1924, see Fig. 35-1) and the fermentation of maltose (Willstätter et al. 1920). It may be assumed that they also played a part in the research on the metabolism of sugar and its role in biological energetics, which was accelerating rapidly after 1930.

Nicotine is one example of the alkaloids mentioned by Walden. It was first synthesized by Pictet and Rotschy (1904) in both left-handed and right-handed versions; these turned out to have some-

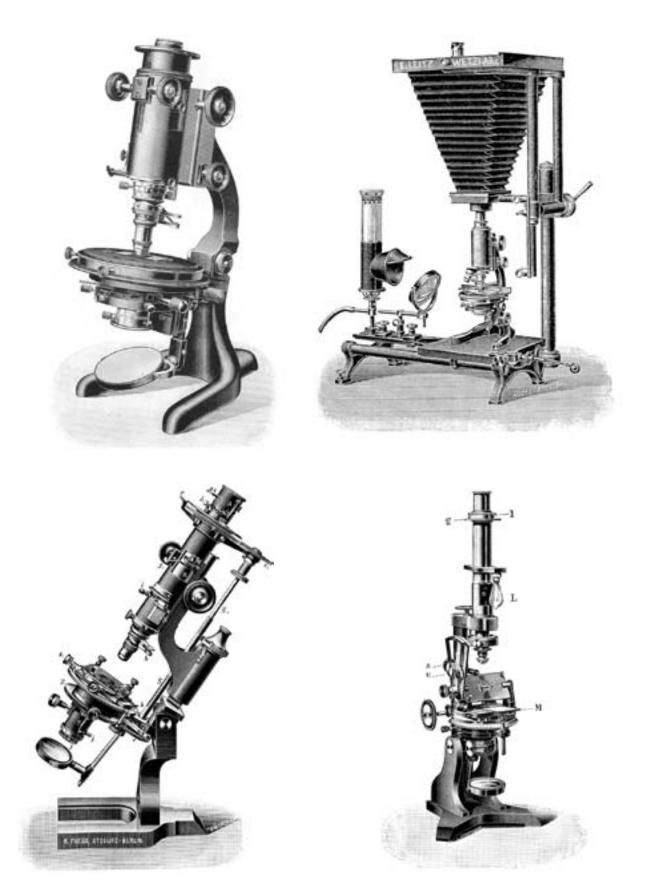


Figure 29-3. More microscopes, after 1900. Top left: E. Leitz' instrument intended among other things for photography of thin sections. From Duparc and Pearce (1907). Right: The same microscope attached to camera bellows (Lincio 1907). Bottom left: A microscope from the Fuess workshop (Lummer 1909). Right: A Société Genevoise microscope with an attachment designed by F. Wallerant (1897) for measurements of the refractive indices of individual crystal grains. From Duparc and Pearce (1907).

what different biological effects. Another alkaloid is quinine, produced from the bark of certain trees and used as an anti-malaria drug. When Rabe and Kindler (1918) succeeded in converting a related poisonous compound into quinine, they depended in part on polarimetric measurements. Man-made quinine, however, did not become competitive in price with the natural product until decades later. Polarimeters also played an important role in the first syntheses of ephedrine (Spåth and Göhring 1920) which is used in various medicines as well as a somewhat controversial food supplement, and of cocaine (Willstätter et al. 1923).

Yet another example of the optically active natural products listed by Walden above, is a chemical discovered by Tanret (1890) in deposits appearing on rye plants due to a fungal infection. He noted that it resembled cholesterol from animals, and gave it the name ergostérine (now ergosterol). It was soon found also in other fungi and yeast. Polarimeter measurements and observations in a polarizing microscope were quite helpful in the purification and characterization of this compound (Tanret 1908). In the early 1920s it was noted that Vitamin D which protects children against the bone-softening disease rickets, could be produced from various fatty substances by irradiation with ultraviolet light. This discovery sparked extensive research where polarimeters again were involved (see e.g. Bills and Honeywell 1928, and the Nobel lecture of A. Windaus in 1928). For a while it was thought that the ultraviolet rays were transforming cholesterol into Vitamin D. However, it was eventually found that the source of one modification of the vitamin (called D₂ or ergo-calciferol) was ergosterol which tends to occur in cholesterol as a tiny impurity. The light also transforms a related compound from animal tissues into another modification of the vitamin (D₂ or chole-calciferol).

Research in human biochemistry, clinical medicine, and pathology often made use of polarimeters (Lecher 1917); some of those on the market were spectro-polarimeters (Fig. 29-4 top, Fig. 29-13, Twyman 1906). Glycogen (e.g. Pflüger 1906), glucuronic acid, maltose (Blake 1916), lactose (e.g. Wiley 1896b, Pellin 1899), alkaloids (Ratz 1905, Annett and Bose 1923), pentoses in urine (Zerner and Waltuch 1914), bile acids and pigments (Vahlen 1895-96, H. Fischer 1911, Wieland and Sorge 1916, see Appendix 5) and albumins (Schütz and Huppert 1900, Young 1922) were among the substances investigated. Polarimeters also aided (Rosenheim and Tebb 1908) in research on "protagon", a substance of controversial properties reported to occur in various animal organs. Black (1908; Benedict and Joslin 1910) described a polarimeters in research on diabetes continued to at least 1930 (Kuriyama 1916, Winter and Smith 1923, Lundsgaard and Holböll 1924, Paul 1925, and many others; Struers 1925). This included observations on the effects of insulin after it was discovered in 1921-22.

A peculiar story concerns certain hypotheses and experimental results of E. Abderhalden, who was well known from other achievements and was even nominated for Nobel prizes more than once. From around 1909 he claimed (e.g. Abderhalden 1913) that human and animal bodies respond to events like infections, cancer and pregnancy by forming "defence enzymes" (Abwehrfermente). This was for a while considered a remarkable discovery. The activity of the enzymes in blood was supposed to be measurable with polarimeters and other methods, and a pregnancy test based on it (Abderhalden and Kiutsi 1912) was in some use for at least 20 years. However, the test turned out to be very difficult to perform and interpret, and others demonstrated already in 1914-16 (see Nature 393, p. 109-111, 1998) that the defence-enzyme hypothesis had no foundation at all. Another suggestion by Abderhalden may have been more reasonable, namely to estimate the total amount of blood in a living animal by injecting a known quantity of an inert compound with strong optical activity into an artery, and then measuring its concentration when it had mixed uniformily with the blood (Abderhalden and Schmid 1910). Analogous methods for finding the amount of fluids or their rates of flow, based e.g. on the use of dyes or isotope tracers, later became

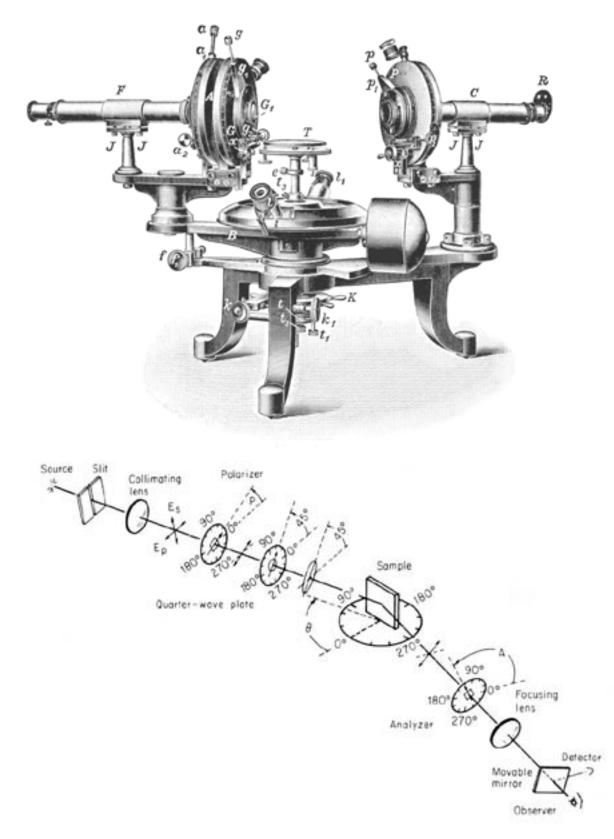


Figure 29-4. Top: A sophisticated instrument for precise measurements of the state of polarization of light at various wavelengths, improved by Leiss (1905) from an original design by T. Liebisch. Light is incident from the right, a prism or a diffraction grating may be placed on the platform T. From the catalog of Fuess (ca. 1910). Bottom: Diagram of a similar device for testing the change in polarization of light (coming from the left) upon reflection from for instance metallic surfaces and thin films of various materials. Such instruments, which were used by P. Drude and many others from around 1890, later incorporated photoelectric sensors and became known as ellipsometers. They are still important in research on semiconductors. From Bennett and Bennett (1978).

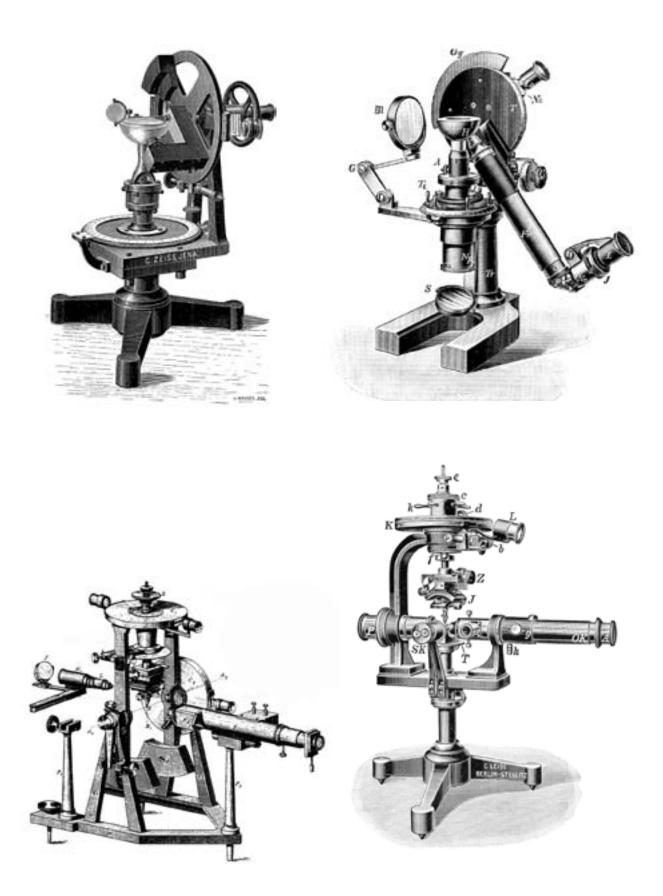


Figure 29-5. Top left: Apparatus for measurement of refractive indices of crystals, from C. Zeiss (Czapski 1891b). Right: Similar equipment designed by C. Klein (1902) and produced by R. Fuess (Duparc and Pearce 1907). Nicol prisms are at Np and Na. Bottom left: A versatile crystallographic instrument described by Viola (1897). Right: The axial-angle apparatus of Wülfing (1898) with Nicol prisms at P and A, produced by Leiss (1925).

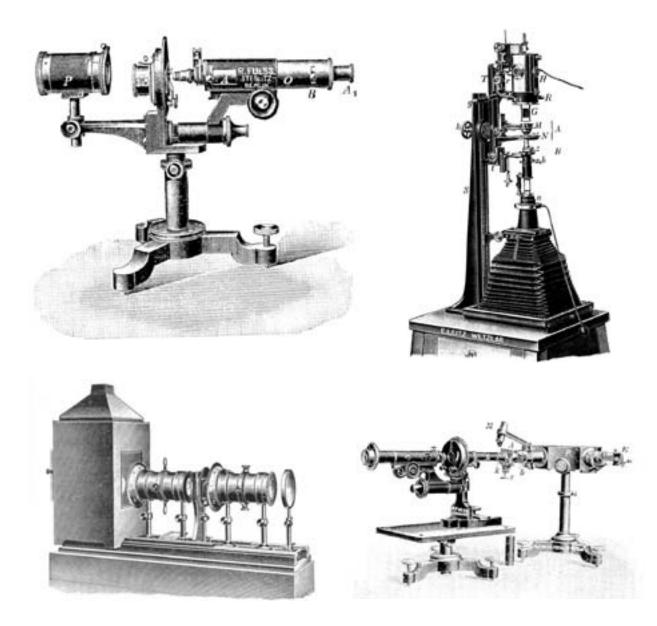


Figure 29-6. Various instruments for research and teaching in crystallography and mineralogy. Top left: A projector, coupled to a horizontally mounted microscope from R. Fuess (Leiss 1903). Right: A Leitz enlarger for the demonstrating, photographing and drawing of microscope images (Berek 1913a). Bottom left: A projector with polarizing prisms for viewing large thin sections. The diagram is from the book by Tutton (1922), and the projector is probably his own design. Right: On the very right we have E.A. Wülfing's monochromator light source for the polarizing microscope seen on the left, both manufactured by R. Fuess. The illustration is from Rinne (1922a).

common in many situations. Various other research projects by Abderhalden and co-workers on animal metabolism, not described here, also employed polarimeters.

As regards plant metabolism, various extensive studies were carried out on the genesis and storage of sugar in plants, the sugar beet and vine being of particular interest. Polarimeters were probably involved in some such studies, but papers about these largely appeared in journals which I have not had the opportunity to consult. The concentrations of sucrose, other sugar compounds and starch as well as their changes with time and conditions, were investigated in other plants (Brown and Morris 1893, Schulze and Frankfurt 1895, Gast 1917) and in seaweeds (Kylin 1915).

Gamgee and Hill (1903) and Gamgee and Jones (1903, 1904) measured hemoglobin and some nucleic acid proteins with a polarimeter, remarking that such measurements had not been carried

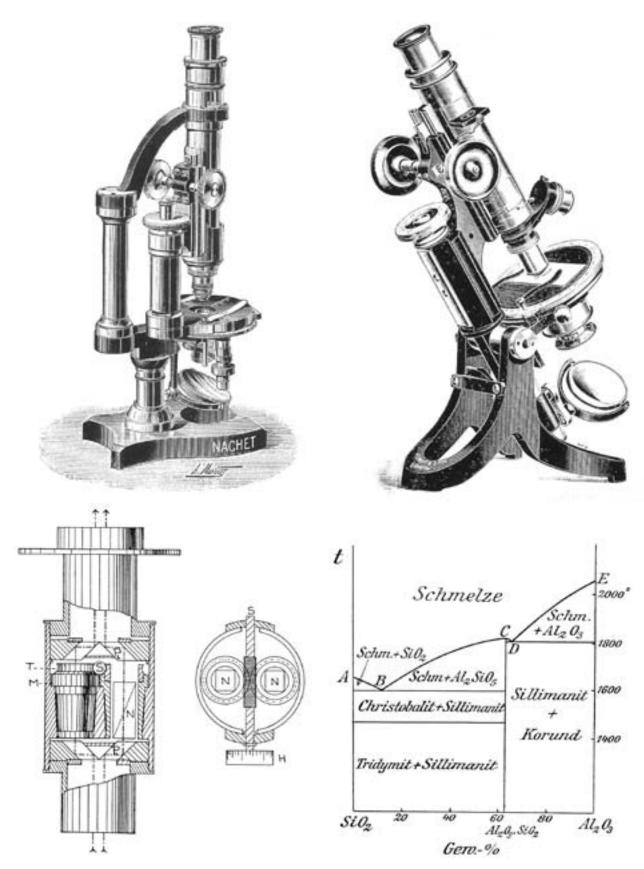


Figure 29-7. Top: Two microscopes for student use. The one on the left is from A. Nachet's (1979) 1910 catalog and the one on the right is from J. Swift & Son (displayed in J. Royal Microsc. Soc., 1907). Bottom left: Wright's (1908a) bi-nicol ocular for measuring extinction angles of minerals. Right: A stability diagram for mixtures of silica and aluminium oxides, obtained by Shepherd and Rankin (1909) with the aid of Wright's optical observations. From Boeke (1915).

out previously on these important compounds. Among other things they found strong optical activity in nucleic acid material from cells of the thymus gland, and Jones (1908) used polarimetric data to confirm that the same nucleic acid was present in other organs. At the beginning of his paper, Jones states that "A study of the nucleic acids is …one of the most puzzling tasks that confront the student of physiological chemistry and…the literature on the subject reveals a mass of contradictions, corrections and inconsistencies which it would seem almost impossible to reduce to any satisfactory scientific order.".

Neuberg (1902) studied in pancreatic tissue a pentose sugar which A. Kossel had found in 1893 (section 27.4) to be a constituent of nucleic acids. Neuberg concluded that it was xylose, and Wohlgemuth (1902-03) found a sugar with the same optical activity in liver. However, Levene and Jacobs (1909) succeeded with the aid of polarimetric measurements, to exclude the possibility of this sugar being xylose. On the other hand, both its melting point and optical activity agreed with those of ribose which A. van Ekenstein had recently synthesized. This was later amply confirmed; one type of nucleic acids, RNA, is in fact named after this sugar. In the nucleic acids called DNA, a hydrogen atom has replaced an OH-group in one of the C-atoms of the ribose; see for instance Levene et al. (1930) who substantiated this by measurements of optical activity and other properties. Adenosine triphosphate (ATP) which has a key role in the energy conversion in cells, contains a ribose.

Optical activity measurements also promoted realization of the fact that quite similar nucleic acids were widespread in the living world (e.g. Schulze and Trier 1910-11). Pighini (1910-11), Amberg and Jones (1911) and Levene and Medigreceanu (1911) studied the enzymatic decomposition of nucleic acids under varying conditions. Levene (1920, etc.) and others investigated the activity of various structural units (nucleotides) in these acids, and of their derivatives. In addition to the work of E. Fischer covered in section 27.4, these and other polarimetric measurements (section 22.3) probably aided in unravelling those main characteristics and functions of the nucleic acids which are now well known. Later (cf. section 36.8), polarized light was used in other research on nucleic acids, for instance in studies on scattering (cf. section 27.1) and on the Maxwell effect (cf. section 27.3).

The rapid development of stereochemistry in the 1920s is described in books like those of Wittig (1930), Freudenberg (1933) and Lowry (1935). In connection with Lowry himself, one may here recall that he proposed in 1923 an important extension of the acid-base concept of physical chemistry. According to the Dictionary of National Biography (supplement volume for 1931-40) this may be traced to his research on the optical activity of organic compounds. J.N. Brönsted who independently made a similar proposal at the same time, also investigated optical activity, but I have not checked where his ideas on the matter originated.

29.3 Reflected-light microscopy, ca. 1900-20

The early history of this field has been decribed by Orcel (1972). As is well known, many useful metals such as iron, lead, copper and zinc are present in the earth's crust in the form of oxides, sulfides etc. rather than pure elements. The grains of these "ore minerals" are generally semiconductors and therefore opaque in thin section. Several of their properties aided geologists in distinguishing them in hand samples or under the microscope: density, hardness, color, magnetism, effects of acids, and so on. Chemical analyses were time-consuming, and it might be difficult to isolate fine-grained ores from their silicate matrix. Additional rapid and dependable analyzing methods were therefore needed, especially in prospecting, mapping of mineral bodies, and mining operations. The first attempts to employ light reflected from polished surfaces in microscopic

studies of ore minerals were possibly made by H. Baumhauer around 1885. Similar studies on metals and alloys began a decade later, according to a note in La Nature 26(I), 139-140, 1898. Charpy (1896) introduced various techniques for this purpose, and they were advanced further by W. Campbell around 1907 (Orcel 1972).

The state of polarization of light reflected normally from a polished ore-mineral surface depends on the mineral, also varying with the angle of the surface relative to crystal axes if the crystal is anisotropic, and with the wavelength. This can be utilized in two ways, as in transmitted-light microscopes. On one hand a polished rock section containing different ore minerals may be viewed through Nicol prisms (and usually other accessories) to discern variations in brightness and color between grains. For instance, in igneous rocks magnetite (Fe₃O₄) and ilmenite (FeTiO₃) often occur side by side or even as intergrowths, and in polarized light they are easier to distinguish than in ordinary light because magnetite is cubic while ilmenite is trigonal and therefore anisotropic. Short's (1931) handbook on the microscopic determination of ore minerals where the various methods of identification are described, stresses that "The most useful test that can be applied to determine an unknown mineral is the test for anisotropism.". One the other hand, measurements on the intensity and polarization of the light reflected from individual grains may be employed for quantitative estimates of their composition.

A major step forward in this field was taken with J. Königsberger's (1901a, 1908) microscope designs (Fig. 29-10). In his first model, the light incident on a polished specimen was unpolarized, and a Nicol prism, a tilted glass plate and a Savart plate (section 13.5) were used for analyzing the reflected light. In a later model (Königsberger 1909, cf. Schlossmacher 1924) the incident light was first made linearly polarized by a Nicol prism, and the rotation of its plane of polarization upon reflection was measured with another Nicol prism and a quartz plate (Fig. 29-10). Königsberger points out in his papers that for instance the quality of coal may be estimated from the anisotropic reflectivity of polished samples; this technique was later used extensively.

In papers in 1908-10, Königsberger quotes some cases where useful information on metallic specimens may be obtained by microscope examination in reflected light; additional examples are given by Endell and Hanemann (1913). Johannsen (1914) mentions at least two manufacturers of metallurgical microscopes but does not state whether they use polarized light. After World War I, ore microscopes developed rapidly: F.E. Wright replaced Königsberger's quartz plate with an Iceland spar rhomb (see figure in Kristjansson 2000), later introducing a new type of quartz wedge (Wright 1919b, Fig. 29-10). Arrangements for varying the illumination of the specimens by means of Nicol prisms also appeared (Fig. 29-10). More information on this topic will be presented in section 39.1.

29.4 Photometers based on Iceland spar prisms, 1870-1930

Photometers have for a long time been used widely in research and industry to measure the amount of light that materials or objects emit, reflect, absorb etc. They have also been used to investigate how we perceive light. As mentioned in section 18.4, measurement of light intensity were (until the early 20th century) made by comparison of a light source of "unknown" intensity with a "known" or standard source. Both of these sources were made to illuminate comparable areas within the observer's field of view, and he then attenuated one of them in a measureable way until they appeared equal. The five main methods of attenuation were i) moving one source away by a known distance ii) interposing a calibrated wedge of smoke-colored glass iii) varying the size of apertures iv) cutting a light beam off intermittently with a rapidly rotating sectored disc, and v) interposing Iceland spar rhombs, Nicol prisms and/or Wollaston prisms at variable

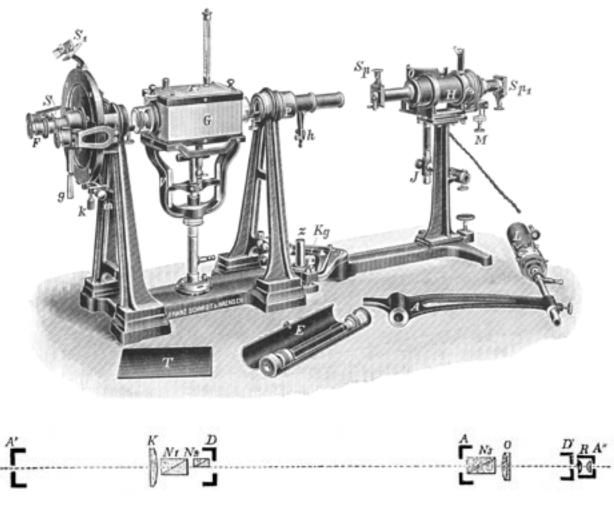


Figure 29-8. Lippich (1885) found a new way of creating a half-shade field in polarimeters, by putting a small Nicol prism (N2 in the bottom diagram) behind the polarizer: the light source is to the left. Different variants appeared later from H. Landolt and others, mostly to be used in high-quality types of polarimeters for research such as the above instrument. H is a spectroscope, G is a bath for keeping the sample tube at constant temperature. One such tube lies at E in front of the polarimeter, along with a lamp L to be mounted at the right-hand end of the frame. From Wien-Harms (1928).

angles and using Malus' cosine law to calculate the resulting attenuation. Wright (1919a) says that photometers of the last-named type are "widely used and are sensitive and reliable". In scientific journals from the period 1860-1940, a great many examples are encountered where they are being employed directly for measuring light intensities, for calibrating other photometers or light filters, or for checking the stability of light sources.

Möller (1885a,b) who improved Wild's (1863) photometer, claims that "...it is known that only polarization photometers are suitable for precise photometric studies". Pellin (1899) advertises a photometer of E. Becquerel's design from the 1860's. Martens (1900a, 1903, Fig. 29-12 top) describes two new photometers (one being easily portable) for white light containing an Iceland spar Wollaston prism and a Nicol prism. The firm of Schmidt & Haensch where Martens was working, manufactured these and other polarization photometers for at least 60 years, see e.g. Glan (1877) and section 39.1.

Godard (1886), Grosse (1887, Fig. 29-11), Weber (1891, Fig. 29-11), Salomons (1893), Königsberger (1901b, Fig. 29-12) and Nutting (1903a, Fig. 29-15) also designed polarization photometers. Grosse's instrument is advertised in a catalog from A. Krüss' workshop in Hamburg in 1899,

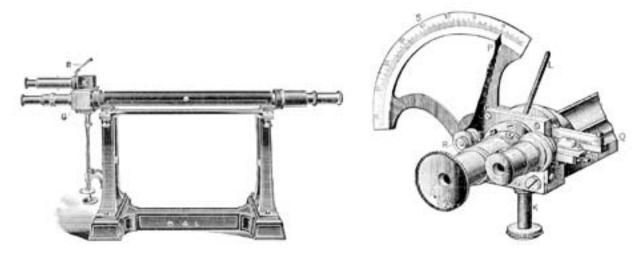


Figure 29-9. Left: A saccharimeter from Bausch & Lomb (Rolfe 1905), of conventional appearance but containing a double quartz wedge (Peters 1894) for increased accuracy. Right: The observer's end of a saccharimeter from 1887, intended for rapid estimation of the sugar concentration in beet. From Browne (1912); some polarimeters for measuring optical activity in wine (Steeg & Reuter 1888) or in diabetic urine had similar scales.

along with some types of spectrophotometers. Weber's photometer, as well as an earlier version without Nicol prisms, was produced commercially by Schmidt & Haensch (cf. Struers 1925), and used among other things to measure the illuminating power of lamps. See also section 36.1. The Weber meter could also provide an estimate of the state of polarization of the light. This was used by Pochettino (1905) when observing luminescence in crystals, and by Busch and Jensen (1911) who with their co-workers investigated the light from the sky (Fig. 29-11, and section 27.1). Blondel (1920) introduced new photometers with Nicol prisms, intended for studies of the absorption of light in gases and liquids.

One popular version of the Martens photometer (see Boas 1899) was dedicated to measurements of the darkening of areas of photographic plates (Fig. 29-11), and used e.g. in fundamental work in that field at the Eastman Kodak laboratories (Wightman et al. 1923). Another instrument developed for this purpose was a photometer with Nicol prisms and a split 60° glass prism (Wallace 1907), based on an older instrument by D.B. Brace. If an empirical calibration formula for the darkening as a function of light intensity, exposure time etc. was available (see the final paragraph of section 29.5), the darkening could be used in estimating the intensity of very faint light, such as from individual stars. The Martens instrument also found applications in spectrometric and Xray (Glocker and Traub 1921) work. Danjon (1926) similarly constructed a small photometer of the type originated by Wild (1863), in order to measure darkening of photographic films and to calibrate glass absorption wedges. Santon (1928) made further modifications to this type of photometer, as Wild (1883, 1888, 1890) himself had also done much earlier (Fig. 29-14). In case of fine details (especially spectral lines) in photographs, different photometer types were preferred.

Kieser (1919) adapted a Martens-type photometer for the measurement of gloss on photographic paper (Fig. 29-17). Other specially designed apparatus with Nicol prisms, produced commercially, was also intended to measure the surface characteristics of paper, paint, enamel etc. (Glanzmesser, glarimeter; Ingersoll 1921, Wien-Harms 1928, p. 517-519, Boffey and Derrett-Smith 1931).

In so-called "flicker photometers", the light beams from the two sources to be compared were made to alternate between the two adjacent spots (or the same spot) in the observer's field of view many times per second. One version of these meters contained a Nicol prism which rotated at constant rate in front of a Rochon- or Wollaston prism (Maisel 1908, Ives 1917).

The polarization photometers for white light do not seem to have been much used in fundamental research on thermal radiation, apart from the examples mentioned in section 17.3. However, Möller (1885a) made measurements with an instrument of the Wild type which confirmed Lambert's emission law. According to this law, the intensity of the light emitted in a particular direction is proportional to the cosine of the angle from the normal to the emitting surface. Later, Laue and Martens (1907) carried out analogous experiments on hot platinum, with Martens' polarization photometer. They computed the optical properties of the metal from the state of polarization of the emitted light, and criticized some of the procedures of Möller and other previous investigators.

As mentioned at the end of section 18.4, a simple setup with two Nicol prisms in tandem was often used when people desired to dim a light by a known amount during experimental work. One such case is in the comparisons by Crova and Lagarde (1881) between light from the sun and from a standard lamp. J. Fröhlich who continued studies by G.G. Stokes on the polarization of diffracted light (see section 17.1), also measured the intensity of that light by means of Nicol prisms (Fröhlich 1878, 1882). R.W. Wood (1905, p. 175) similarly studied the properties of diffraction gratings by comparing the intensities of light received directly from a sodium source and light from that source deflected by the grating. It may be recalled in this context that the use of diffraction gratings in spectral analysis increased much after ca. 1882, but the first generations of gratings tended to give rise to spurious extra spectral lines, called ghosts. According to later literature, Wood contributed to the banishing of these ghosts by improvements in grating production. Camichel (1905) used a pair of Nicol prisms in studies on light absorption in fluorescent substances, and so did Küch and Retschinsky (1906, 1907) when measuring the light intensity from a new type of mercury lamp, as well as Lépine (1915) in an investigation of luminescence (Fig. 36-5). Merton and Nicholson (1918) and Fabry and Buisson (1921) attenuated their light sources in the same way when calibrating the sensitivity of photographic plates in their research on spectra. Dietrich (1881), Brotherus (1912), Voss (1918), Gerlach and Brezina (1924) and others utilized Nicol prisms when measuring the ratio of intensities of adjacent spectral lines from hot metal vapors. Gerlach later wrote widely-read books on chemical analysis of metals by spectroscopy of light emitted from high-tension discharges. His methods were in part based on such intensity comparisons, made in most cases with different types of photometric equipment.

Polarization photometers were no doubt applied to some extent in measurements of light intensity from street lamps, of illumination in schools and workplaces, of reflection from walls, and so on. Accounts of such practical studies may however mostly have appeared in hard-to-find industrial journals and bulletins. One of the pioneers in illumination research (Priest 1913) described a photometer for this purpose, with three Nicol prisms instead of the usual two.

29.5 Spectrophotometers and related equipment with Nicol prisms, 1870-1930

Two different types of photometers may be used when light intensity is only being measured in a limited (perhaps variable) range of the visible spectrum. One of their most important roles was in analytical work in chemistry, biochemistry and medicine. The light might be given off by a substance in a flame or an electric spark, or it might be absorbed (especially in an aqueous solution). In the latter case, the interpretation of the measurements is based on Beer's law which was introduced in section 18.4 above and which had been verified satisfactorily around 1870 (Krüss and Krüss 1909, p. 101-106). The less expensive photometer type is called colorimeters, measuring light in a rather wide (and often fixed) band of wavelengths. They are convenient to use in routine work on solutions where a single colored compound is present in much greater concentration than

others. In a colorimeter, one of two beams from a lamp traverses an unknown solution, while the other one is sent through a standard solution (or a colored glass rod). The observer makes adjustments in the path lengths or the relative intensities of the incident beams, until both the emerging beams appear equal. In the more expensive type, spectrophotometers, comparisons are made between two light sources. A narrow wavelength interval to be used is selected by means of a prism or a grating. The observer may even scan the whole visible spectrum in this way. The method may be used in estimating the concentration of one compound in a complex mixture, if it is the only one that emits or absorbs light in a particular wavelength range. Iceland spar prisms were used for light attenuation in some models of both these photometers.

Beginning with colorimeters, it appears from e.g. Journal of Biological Chemistry 1905-25 and general catalogs like Struers (1925) that only a small fraction of these instruments were based on polarization techniques. However, Krüss (1892) designed one containing a Nicol prism, two Iceland spar rhombs and a quartz plate, and he claims that it is "very superior" to other colorimeters. Krüss and Krüss (1909, Fig. 29-16) present examples of measurements of the concentration of metal salts in aqueous solution to an accuracy better than 1%. They point out that it may be useful in quality control of drinking water and beer, industrial dyes, and even steel. The Krüss colorimeters were produced by a family firm of the same name which was mentioned in the previous section and is still in business in similar fields. Pellin (1899, 1913) advertised colorimeters with Nicol prisms, and Meisling (1904, Fig. 29-16) designed a model intended for measurements of hemoglobin in blood. Colorimetric measurements on blood and other fluids often tended to be much simpler and faster than spectrophotometric measurements (see e.g. a paper by W.C. Stadie in J. Biol. Chem. 41, 1920).

The concept of a spectrophotometer was suggested by Govi (1860a), but K. Vierordt was the first to demonstrate a working instrument a decade later. Soon after, Nicol prisms were incorporated in such apparatus. Encyclopaedia Britannica (11th ed., 1910-11, entry on Photometry), Weigert (1927), Walsh (1958) and other sources refer to polarization spectrophotometers by for instance H. Trannin (1876), P. Glan (1877, Fig. 29-13), W.v. Zahn (1878), A. Crova (1880, 1883, Fig. 29-13), G. Hüfner (1877, 1889, Fig. 29-15), improved by Houstoun (1908) with a pentagonal Iceland spar prism, A. König (1894), G. Melander (1897, advertised by Pellin 1899), J. Königsberger (1903), J.R. Milne (1904), P.G. Nutting (1911, etc., see Fig. 29-12 and 29-15), and H.B. Lemon (1914, Fig. 39-4). However, the sources just mentioned also point out some problems with the operation of this general type of photometers and spectrophotometers.

The König meter was used e.g. by Köttgen and Abelsdorff (1895) in research on pigments in vertebrate's eyes but it was soon superseded by the König-Martens polarization spectrophotometer (Fig. 29-15). This very successful device was an improvement by Martens and Grünbaum (1903) upon the König instrument, with among other things an Iceland spar Wollaston prism replacing a quartz Rochon prism. The König-Martens meters did much service in the research on scattered light which was introduced in section 27.1 (see e.g. Aufsess 1904, Steubing 1908, Stamm and Svedberg 1925) and which will be dealt with further in section 36.7 (Fürth 1915). To give a few examples, they were also useful in measurements of light absorption by dissolved salts (Grünbaum 1903, Müller 1906), and of the concentration of chlorophyll in plant tissues (Malarski and Marchlewski 1910), in studies of chemical equilibria in solutions (Hildebrand 1908), of the rate of photochemical reactions (Luther and Forbes 1909), of light emission from hot metal surfaces (Stubbs and Prideaux 1912), and of the absorption spectra of photographic sensitizers (Wise and Adams 1918). Stuchtey and Wegener (1911) measured the albedo of the Earth and of clouds in six balloon ascents, using a specially constructed König-Martens instrument.

The Hüfner spectrophotometer was employed in a similarly wide range of projects, including observations of diffusive mixing of fluids (Wroblewski 1881), light transmission by water (Hüfner

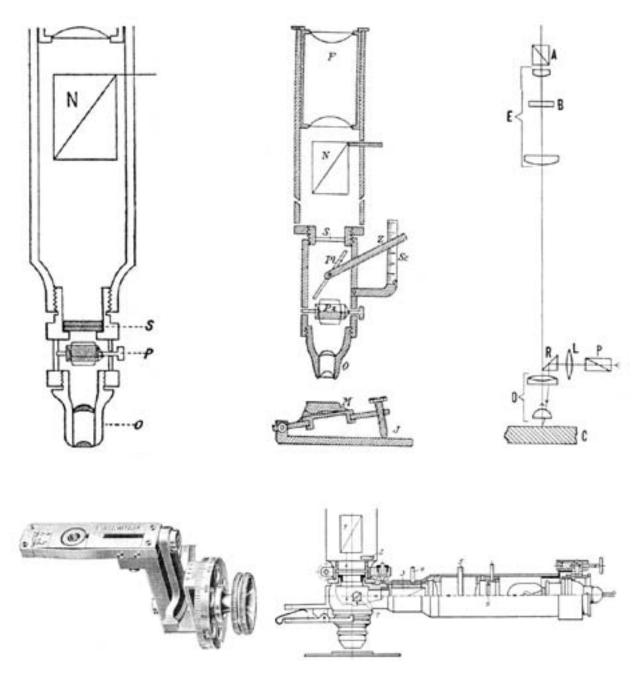


Figure 29-10. Development of reflected-light techniques in microscopy, for studying ore minerals (and later, metals). Top left and center: Schematic diagrams of microscopes made by Königsberger (1901a, 1908). N is a Nicol prism, and S is a Savart-plate compensator from Iceland spar. The 90° glass prism P reflects light to the specimen M. Right: In the later design of Wright (1919b) a quartz wedge B invented by Bates (1908) has replaced the Savart plate. Bottom left: Berek's (1913b) compensator for measuring the phase angle of elliptically polarized light in microscopes (from Rosenbusch 1924). The wheel makes a 0.1 mm thick Iceland spar plate rotate in the round hole. Right: Illumination arrangement for reflected-light microscopy, from the workshop of C. Reichert. Nos. 1 and 3 are Nicol prisms. A specimen with a highly polished surface is on the plate below the microscope objective.

and Albrecht 1889), acid-base reactions (Lellmann and Gross 1890), light absorption by aqueous solutions of metal salts (Magnanini and Bentivoglio 1893), and emission spectra of lamps (Leder 1907). The Hüfner meter, with improvements by Twyman (1906) was produced for a decade or so by the firm of A. Hilger. After 1918 however, Hilger instead began selling a Nutting polarization photometer attachment for their own quartz-prism spectroscope. This equipment also found many applications (Getman 1922, French and Lowry 1924, both studying dissolved copper salts), and it

was even in occasional use into the 1940s in spite of increasing competition from non-polarizing spectrophotometers after 1920 (section 39.1).

Ketteler (1881) and Pulfrich (1881; Ketteler and Pulfrich 1882) employed a Glan spectrophotometer for testing of theories on the relation between refractive indices and absorption in colored materials. It may also be noted here that Ketteler had used Mascart's (1864) measurements of the dispersion of the ordinary ray in Iceland spar, when formulating these theories. Similar studies on organic dyes were made by Plaats (1915). Zsigmondy (1901) at the famous O. Schott glass-works measured light absorption in glasses colored by admixtures of metal salts; he states that knowledge in that field is "of the greatest importance for the glass industry". The Glan meter (improved by H.C. Vogel, see section 29.6) was also used in measurements by G. Müller (1912) of the attenuation of light in the atmosphere at Tenerife; the results agreed well with theoretical derivations.

Spectrophotometers with Nicol prisms were used in distinguishing between coloring agents e.g. in paints (Pitcher 1888) or food (Gibson et al. 1922), raw materials in the sugar industry (Hinton 1930) and in industrial goods (Nichols 1884, Holmes 1923), in measuring the light intensity from gas lamps (Wild 1888) and other lighting (Vogel 1877, Nichols and Franklin 1889) for streets and factories, studying diffuse reflection from powders (Wright 1900), painted walls (Nutting 1912) and other matt surfaces for practical purposes (Messerschmitt 1888, Návrat 1912, Woronkoff and Pokrowski 1923), investigating luminescence caused by radioactivity or other agents (Hauer and Kowalski 1914, Fig. 29-15, Wawilow 1925, Smith 1926), measuring light absorption in various types of glass and in dye solutions (Königsberger 1901c, Houstoun 1906, Königsberger and Kilchling 1909, Wright 1921), and so on. Umow (1912) describes a spectroscope with Nicol prisms and a Savart plate, for studying light absorption in materials and the nature of dyes. According to the Dictionary of Scientific Biography (1980-90) instruments of the Umow type were used widely both in laboratories and in testing of colors in textile industries. Fuess (ca. 1910) advertised three models of pocket spectrophotometers (Fig. 29-12), one of which was based on the design by Nutting (1906). They were intended e.g. for work in botany and forestry but I have not located any papers describing their application.

Gouy (1876, Fig. 29-13) introduced a new polarization spectrophotometer containing Nicol prisms and an Iceland spar rhomb, for measuring light intensity from a gas flame into which solutions of various metal salts were injected. An improved version is described in a comprehensive paper on the results of his experiments (Gouy 1879). Cotton (1899) states that they provided interesting insight into the general laws governing emission and absorption of light. Camichel (1895) made use of a modified Gouy instrument in research on absorption of light by crystals and by bromine. It is advertised in the catalog of Pellin (1899) as being suitable for research on spectra from flames and metallic vapors.

I have not investigated when large-scale application of spectrophotometers in routine analytical work began. Hüfner (1877), Glazebrook (1883b) and Crova (1883) discuss this possibility when describing their meters, referring also to previously published papers. Sabatier (e.g. 1887) used the Crova meter in a study of the absorption of chromium- and bromine compounds, and the catalog of Pellin (1899) claims that it can provide quantitative analytical results. Camichel and Bayrac (1901) investigated phenol dyes with this type of instrument, and Vaillant (1903) studied solutions of salts and organic compounds. Houstoun and Brown (1911) carried out similar work with Houstoun's spectrophotometer mentioned above.

Spectrophotometers with Nicol prisms clearly played a role in the development of absorptionanalytical techniques for blood samples. Thus, Noorden (1880), Hüfner and Külz (1883) and Otto (1885) studied the properties of hemoglobin by means of Hüfner's meter, later also Krüger (1888), Dreser (1891, carbon monoxide poisoning), Saint-Martin (1900), Butterfield (1909), Bürker (1911, calibration of a new type of colorimeter for blood), Hári (1917) and Welker and Williamson (1920). According to Ray et al. (1932), tables regarding hemoglobin absorption which Hüfner published before 1900, were still in use. For a while it appeared that hemoglobin underwent some changes during illness but Butterfield showed that this was not the case. Hüfner improved his spectrophotometer around 1907 and it was used at first by Butterfield who then switched to a König-Martens meter. Schertz (1923) employed the latter instrument in his research on important plant pigments related to hemoglobin. Kennedy (1927) found that results from a Bausch & Lomb König-Martens spectrophotometer compared favorably with new analytical methods which were being developed at that time. É. Branly (1882, Fig. 29-14) and Kraus et al. (1899) used a Glantype meter in studies on hemoglobin, and Branly concluded that polarization spectrophotometers had great advantages over other instruments for such research.

Soret (1883) used Iceland spar prisms in his spectrophotometer (Fig. 29-21) when investigating how blood and its various components absorb ultraviolet light. He found strong absorption at just over 400 nm. This was due to porphyrins which are characteristic for so-called hemoproteins, and the wavelength interval in question was subsequently named "Soret's band". In some proteins he also found an absorption peak around 275 nm; it may for instance be responsible for some of the damage to animal tissues caused by ultraviolet light (UVB-rays). Chardonnet (1883) also explored with the aid of Iceland spar prisms how eyes absorb ultraviolet light. In 1896 scientists realized for the first time that the chemical compositions of chlorophyll and hemoglobin are very similar. In that context, Iceland spar prisms had at least a small role in continuing research on the absorption spectra of porphyrins (e.g. Schunck 1898, 1899, Marchlewski and Schunck 1900) and plant pigments (Schunck 1903).

As stated above, colorimeters were probably more popular than spectrophotometers in research on hemoglobin and related compounds in the early 20th century. However, Ray et al. (1932) and Drabkin and Austin (1932) note that with appropriate procedures, the photometers provide accurate and rapid determinations. The latter authors were using a König-Martens instrument.

Instruments with Nicol prisms were not involved in the measurements on which J. Stefan in 1879 based his hypothesis that the total light intensity emitted by a surface increases with fourth power of its absolute temperature. Violle (1881) measured such thermal radiations with a polarization spectrophotometer which he had evolved from older types (Fig. 29-14). He considered that a more complex formula of his own fitted his results better than Stefan's. Unfortunately for him however, L. Boltzmann demonstrated in 1884 that the fourth-power law could be deduced theoretically from the basic principles of thermodynamics. Garbe (1887) who measured the light intensity from various lamps with a Crova instrument (cf. above), had even less luck than Violle as he had more faith in an old formula proposed by Becquerel (1863, see section 17.3) than in the Stefan-Boltzmann law for use in his interpretation. These happenings may for a while have led to diminished interest in the use of polarization spectrophotometers in research on thermal emissions. At any rate, up to the end of the century they for instance only played a minor role (Crova and Houdaille 1890) in studies on the solar radiation.

In connection with the research on thermal radiation, Nicol prisms took part in attempts to solve the question of how the radiated energy is distributed by wavelength at different temperatures. Preliminary measurements were made by Nichols (1879, 1880) with a leucoscope (see section 29.7) and by Lecher (1882) using a polarization photometer of the Glan type. Important series of measurements on the spectral distribution of black-body radiation (Paschen and Wanner 1899, Wanner 1900, Paschen 1901) were performed in part with König's spectrophotometer which contained Rochon- and Nicol prisms as already mentioned. These measurements are referred to in a paper by M. Planck (1900) on such radiation, and they aided in the verification of W. Wien's hypothesis that the wavelength of maximum intensity is inversely proportional to temperature. The important distribution formula published by Planck in 1901 which marks the beginning of quantum theory, was however deduced to explain results on the thermal radiation which had been acquired by electrical sensors (bolometers, etc.)

For a sensible interpretation of fundamental and practical measurements of light intensity, it is necessary that all investigators have access to standardized light sources. Many types of candles, oil-, gas- or electrical lamps were employed for this purpose after being thoroughly tested and optimized for consistency of output. Violle (1884) suggested that the light emitted per area by pure platinum at its melting point should be used as a universal standard. In estimating this quantity, he used both his spectrophotometer mentioned earlier in this section, and a modified version. Violle's proposal which may have been the first one for a scientifically defined reference in the field of luminosity, was favorably received by many (Winkelmann 1906, p. 755). Unfortunately there were serious technical problems with its implementation (see a committee evaluation in the British Association Report for 1888; Liebenthal 1907, p. 134, 147) and it was little used until 1930 or later. A proposal for a standard similar to that of Violle was ratified internationally in 1948.

In continuation of the work on thermal radiation, increased efforts took place to utilize it in the estimation of high temperatures. As an example, J. Violle made measurements with his spectrophotometer in the early stages of development of H. Moissan's electrical furnaces, and he assisted in the research for which Moissan was awarded the Nobel prize (see section 22.4). H. Wanner whose papers were referred to above, adapted the instrument of A. König for the specific purpose of measuring the temperature of glowing objects (at 900-4000°C; Wanner 1902, Liebenthal 1907, p. 421, Burgess and Le Chatelier 1912, Ribaud 1931, p. 282-289). The Wanner optical pyrometer was produced commercially for some 15-20 years (Fig. 29-17, top) and was said to be "of great precision and convenience" in a review paper on high-temperature measurements (Phys. Rev. 19, 1904). It clearly found many uses in industry, contributing to improved products and energy savings. Among these uses one may mention the melting of various metals (Nernst and Wartenberg 1906, Greenwood 1909), production of glass, ceramics and cement, operation of carbon-arc lights (Waidner and Burgess 1904), conversion of coal to coke and gas (Jüptner 1908), as well as production of fire-resistant materials (Goodwin and Mailey 1906, Ruff and Schmidt 1921, Matignon 1923), carbides and other abrasives (Lampen 1906, Pring 1908, Gillett 1911), heavy metals (Kalmus 1914, Lely and Hamburger 1915), various silicates (Stein 1907), steel (Greenwood 1918), and filaments for incandescent electrical bulbs (Pirani 1910). O. Serpek invented a new procedure for the synthesis of aluminium nitride which was among other things useful as a refractory material for furnace linings and as an intermediate step in ammonia production. In one of his many patents (Serpek 1909) it is emphasized that the process requires "precise monitoring of the temperature by means of a Wanner pyrometer". This type of pyrometer also aided in a study of a different important method of nitrogen fixation (Thompson and Lombard 1910). Other users of Wanner instruments included Nernst (e.g. 1906) who studied black-body radiation, Jellinek (1906) who investigated the thermodynamics of decomposition of nitrous oxide, Pring and Fairlie (1912) who followed the generation of methane from carbon and hydrogen, and King (1913, and more) in a thorough survey of the effects of temperature on the emission spectra of several metals, among other things for comparison with stellar spectra.

One more area of application of photometry with the aid of Nicol prisms may have been in studies of the various materials involved in photography. For instance, the darkening of photographic emulsions had to be known as a function of light intensity, exposure time and wavelength. Here one may point to papers by Messerschmitt (1885) who charted with a Glan-Vogel spectrophotometer the absorption of so-called sensitizers that improved the performance of photographic plates, Mees and Sheppard (1904, Sheppard 1909, etc.) who made extensive researches in this field with Hüfner- and Hilger- instruments, Kron (1913), Helmick (1918, with a Lemon meter), and Schaum (1924, König-Martens meter). While many other types of photometers were used in photographic research, W. Meidinger claims in the handbook by Gehrcke (1927, vol. II, p. 39) that "polarizing photometers are better suited to measurements of photographic darkening than any others". Weigert (1920 and many others) investigated in detail the influence of polarized light on a number of photo-sensitive substances; his papers have been widely quoted in later works on light perception by organisms and on photochemistry in general.

29.6 Nicol prisms in astronomy, from 1875

Iceland spar has had a surprising variety of roles in observational astronomy. Elsewhere in this compilation, there are accounts of the measurements by F. Zöllner on the light intensity received from stars and planets (section 18.4, Fig. 29-11), the dispersion prisms used by W. Huggins and others (section 29.8), research on the magnetic field of the Sun (section 30.1), and the remarkable instruments invented by B. Lyot (section 39.2). Salet (1910) remarks that because of the polarization of sky light, stars can be seen more clearly in the morning and evening through a Nicol prism than with the naked eye, but I have not seen other references to similar applications. Some important astronomical projects featuring Iceland spar (mostly in the form of Nicol prisms) will be described below.

"After spectral analysis, photometry is the most important field of astrophysics" (Newcomb-Engelmann 1921). An improved version of one of Zöllner's astrophotometers, shown at the 1867 Paris Exhibition, was installed in several observatories in Europe and beyond from 1870. With some further improvements (e.g. Müller 1901), they were used much until 1910 or so, sporadically even to 1930 although photographic techniques and photoelectric sensors had by then mostly replaced visual photometry. The greatest contribution of the Zöllner meters was the measurement in 1886-1905 at Potsdam (Fig. 29-11) of the magnitudes of all stars in the northern celestial hemisphere to M= 7.5. Their total number was about 19,000 (see notes at Müller and Kempf 1894, 1899 in the list of references below).

A Zöllner instrument was tested for a few years at the Harvard College Observatory (Peirce 1878). Then E.C. Pickering who was its director from 1876, developed different photometers containing a Nicol prism and either a beam-splitting prism from Iceland spar and glass, or an Iceland spar plate (Pickering 1879, 1882, Pickering et al. 1884, Fig. 29-18 bottom). In these devices the observer saw side by side a star of known magnitude (in the role played by a lamp flame in the original Zöllner instruments) and the star to be measured. Extensive systematic measurements of star magnitudes began at Harvard in 1879. Pickering and his collaborators made around a million observations on 45,000 stars in the whole sky until 1906, in part if not mostly with the polarizing photometers (see Pickering 1895, King 1955, p. 296, Sterken and Staubermann 2000). In the entry on G. Müller, Dictionary of Scientific Biography (1980-90) states that these large efforts by the Potsdam and Harvard groups "furnished the most exact photometric information on stars then available" and that their published records are still "an indispensable standard work.". Instead of the Nicol prisms, wedges of smoked glass were sometimes used for attenuation, but there is evidence in the writings of Pickering, C. Pritchard and others that these were calibrated with the aid of Nicol prisms.

As mentioned in section 18.4, Zöllner (1868) had made additions to his astrophotometer which enabled him to express the colors of stars by numerical values. He may have hoped that this method would replace subjective color classifications initiated earlier by J. Schmidt, H. Klein and others. A few astronomers did carry out systematic star-color surveys by Zöllner's method (e.g. Rosén 1870, Peirce 1878) and a few others made occasional observations; however, most of those interested in the subject such as G. Müller, P. Kempff and H. Osthoff continued using the unaided

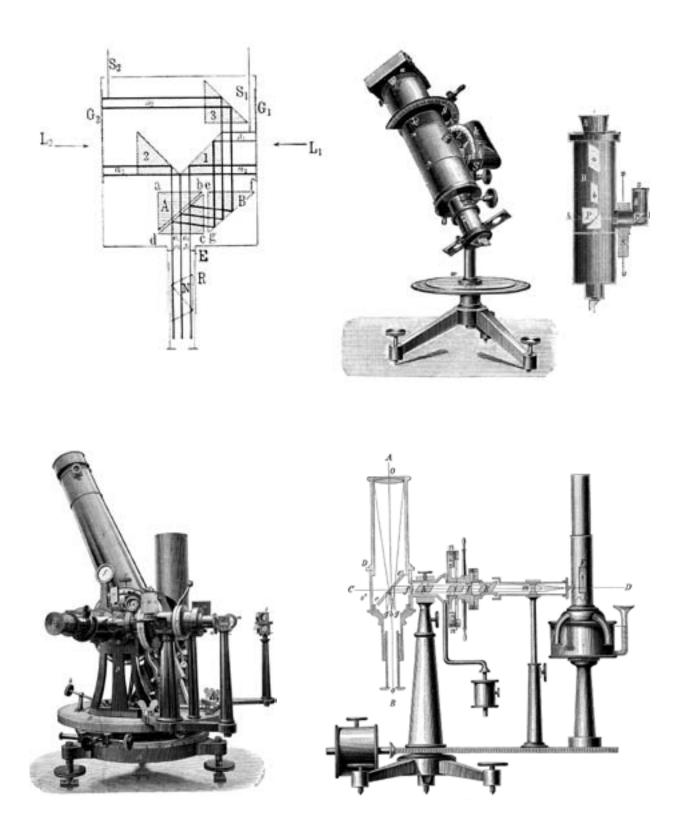


Figure 29-11. Photometers with Nicol prisms. Top left: The "mixing photometer" of Grosse (1887) which compares the intensities of light falling on the matt glass plates G1 and G2. In addition to the Nicol prism N, the instrument contains a Dove (1864) prism of Iceland spar. From Liebenthal (1907). Right: Weber's (1891) photometer, from Busch and Jensen (1910) who used it for observations of sky light. Bottom: Zöllner's (1861, and later) photometer mounted in an astronomical telescope in Potsdam. A cross-section is shown on the right; an observer at B attenuates the light from a standard lamp in the chimney with Nicol prisms, until it appears equal in brightness to a star in the telescope. The third Nicol prism and a quartz plate could be used in estimating (or compensating for) the different colors of the stars being observed. From Newcomb-Engelmann (1921).

eye and possibly colored glass plates until after 1900. Their methods were for instance of insufficient resolution to notice that the colors of so-called cepheid variables changed along with their magnitudes. Around the turn of the century, new methods of color classification emerged. One of these (named after K. Schwarzschild) depends on the fact that the human eye (and therefore also the Zöllner- and Pickering- photometers) is most sensitive to light of around 560 nm wavelength while certain standard photographic plates experience maximum darkening at 440 nm. These methods therefore yielded unequal magnitudes for a star, the difference depending on how its radiation was distributed in the spectrum. This difference (called color index) allowed stars to be divided into some 20 classes (Newcomb-Engelmann 1921, p. 550) and some astronomers attempted to relate it to temperature. Still other classification schemes were developed early in the 20th century (including the OBAFGKMN-scheme at the Harvard Observatory), soon replacing the above ones.

In addition to the very large sky surveys, the Zöllner- and Pickering- photometers were used in the detailed monitoring of a great many variable stars such as novae, cepheids, and eclipsing binaries, see Fig. 29-19. These measurements were carried out at various observatories as well as at Potsdam and Harvard, for instance by Roberts (1906), Wendell (1909), Dugan (1911), McDiarmid (1924) and V.K. Ceraski who also measured star clusters (Ceraski 1902). A very extensive review of all available observations on variable stars to 1915 was prepared by Müller and Hartwig (1918, 1920, 1922). According to the Dictionary of Scientific Biography (1980-90), polarization photometers provided e.g. a considerable part of the experimental basis for understanding the behavior of binary stars. In that work, the measurements of Shapley (1915) with Pickering photometers were the most important. Shapley's interpretation also led to his valuable conclusion at around the same time, that the cepheids were not binaries. He subsequently followed up (e.g. Shapley 1918) E. Hertzsprung's 1913 suggestion that in cepheids a close correlation existed between their average luminosity and period of oscillation. This was a large step in the construction of a scale for the size of the universe. A part of the data set processed by Hertzsprung and Shapley for this purpose seems to have been acquired with Iceland spar photometers.

In 1691, I. Newton had asked the fundamental question whether light of all colors travelled at the same speed through the emptiness of space. Much later, it became a central point in the theories of Maxwell (and still later, of Einstein) that all electromagnetic waves had the same speed in space. In the mid-19th century F. Arago suggested a means of answering this question, by checking whether the variations in brightness of stars like Algol (an eclipsing binary) were in step for red and blue light. Nordmann (1909a) made such observations on some stars of the Algol type, using his own photometer (Fig. 29-19) which had two Nicol prisms like that of Zöllner. Nordmann placed different color filters (aqueous solutions of dyes) in turn in front of its aperture. His methods and conclusions met with some criticism, but Shapley (1923) later made a similar analysis of the magnitude observations made (on photographs with different color sensitivities) by S.I. Bailey and by himself. Shapley concluded that the difference in speed between blue and yellow light was definitely no more than one part in 2.10¹⁰. This result which may be considered a satisfactory confirmation of the theories of Maxwell and Einstein, could not have been easily obtained by any terrestrial means, as the accuracy of light-speed measurements was at that time around one part in 10⁵. Another question of fundamental significance to the above theories is whether the motion of a celestial light source affects the speed of electromagnetic waves emanating from it. This question was also settled through analysis of light from eclipsing stars (e.g. Zurhellen 1914).

As already noted, spectrophotometers with Nicol prisms were used to some extent in research on the radiation from very hot solids (see e.g. Crova 1878, 1880) and gases at varying pressures (Lagarde 1885). The results may have hastened various discoveries in that field. Crova (1878, p. 981) discussed the possibility of estimating the surface temperatures of stars by comparison with

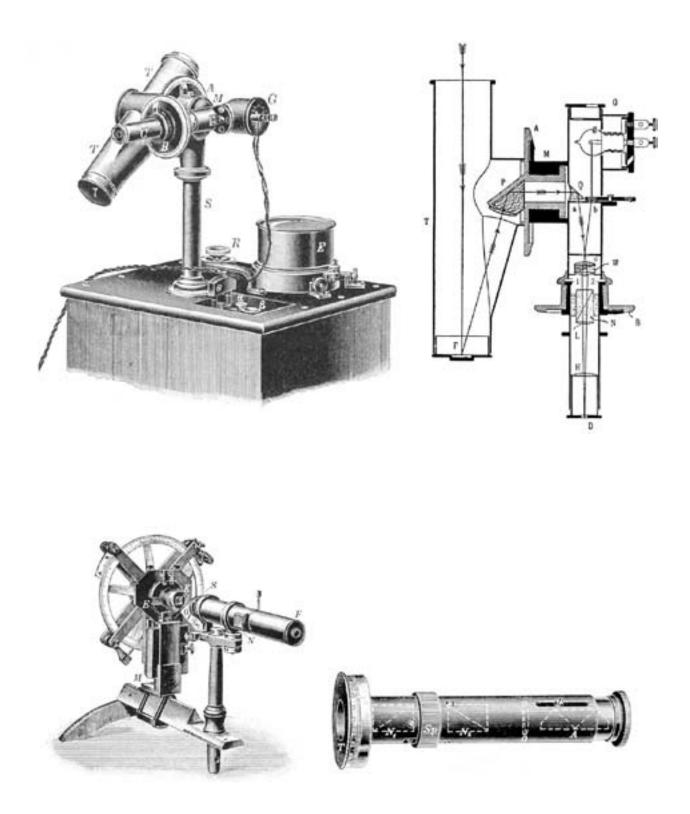


Figure 29-12. Photometers. Top left: Martens' (1900a) popular photometer for white light. Right: A cross-section of this device. The light to be measured enters the telescope T and is reflected from the matt-glass plate F through the hole a. An observer at D also sees the adjacent hole b, which is illuminated by the standard incandescent bulb g through another matt-glass plate. W is a calcite Wollaston prism. From Liebenthal (1907). Bottom left: Königsberger's (1901b) microphotometer for measuring light from very small areas. It contains a Nicol prism, a calcite rhomb and a Savart-plate of quartz, mounted within a horizontal microscope. From Duparc and Pearce (1907). Right: A handy spectrophotometer from Fuess (ca. 1910), based on an idea from Nutting (1906).

the spectra of hot objects and flames. H.C. Vogel (1877, Fig. 29-13) modified the Glan spectrophotometer and incorporated it in a telescope, in order to make important measurements on the solar radiation (see Hellerich 1937, p. 638-639) as well as on the absorption of light both in its outermost layers and in the terrestrial atmosphere (Müller 1882). He also measured the radiation from several stars at different wavelengths (Vogel 1880).

Among the scientists who had by 1900 started dividing bright stars into classes according to their temperature, was N. Lockyer (1904). He had a spectroscopic camera made, containing an Iceland spar dispersing prism and quartz lenses, in order to estimate ultraviolet stellar radiation. Harkányi (1902) pointed out that the data of Vogel (1880) could be used to find the actual star temperatures, which he did himself for some stars and the sun. Nordmann (1909b) published such estimates which he had deduced for 14 stars (Fig. 29-19) from data obtained with his color-filter photometer recently mentioned. Next, Wilsing and Scheiner (1910) used a Nicol-prism spectrophotometer in order to similarly estimate the temperatures of 109 stars assumed to radiate as black bodies. They added at least the same number in the 1910s (see Handbuch der Physik 19, p. 185-187, 1928). The temperature values obtained by these pioneers obtained turned later out to be somewhat too high because stars do not behave like black bodies. Baillaud (1924) recorded the spectra of a few stars down to 350 nm wavelengths using a large Iceland spar dispersing prism, and Graff (1931) made extensive observations on star colors with the aid of Nicol prisms and colored glass wedges, but it seems uncertain whether his data were processed.

The planets and the moon were not excluded in this photometric activity, right from the early work of Zöllner (1868). H.C. Vogel (1880) measured the intensity of sunlight reflected from the moon with his polarization spectrophotometer, and Landerer (1889) and later Barabascheff (1927) investigated the polarization of this light using an adapted version of Cornu's (1882) polarimeter. They compared the results to the properties of light reflected from various rock types and soil. G. Müller, Pickering, Landerer and others made detailed measurements of the light from the planets, and some studies were even made on light from satellites (e.g. Guthnick 1910), comets and minor planets of the solar system with Zöllner- and Pickering-photometers. Rosenberg (1921a) designed apparatus which he called an "area photometer" containing Nicol prisms like the Zöllner instrument, in order to measure for instance the light from different regions of the moon. Yet another photometer of this type was constructed by E. Schoenberg (1917), see Fig. 29-18. Wilsing and Scheiner (1909, 1921) studied light reflected from the moon, planets and rocks with a spectrophotometer, probably containing Nicol prisms.

It was attempted to look for polarization in the light from nebulae (e.g. Meyer 1920) but interesting results were not obtained until much later.

29.7 Iceland spar prisms in research on colors and vision

An important problem in photometry was connected with the comparison of intensities between two sources having different colors. Polarization photometers could surmount this to some extent if they contained thin quartz plates cut parallel to the optic axis (Zöllner 1868, Zahn 1874, Crova 1881), and the same applied to colorimeters (Pellin 1899, Meisling 1904). Here, the interference colors discovered by Arago in 1811 and investigated by Biot (e.g. 1812, 1816c) were utilized.

By placing quartz, gypsum or mica plates of different thickness between two Nicol prisms, one was in a simple way able to produce light having almost any combination of spectral colors. Complementary color pairs (which together make white light) could also be produced, and detailed studies on these matters were carried out by Brücke (1848) with a microscope. They were useful in the observations on color perception in polarized light mentioned at the end of section

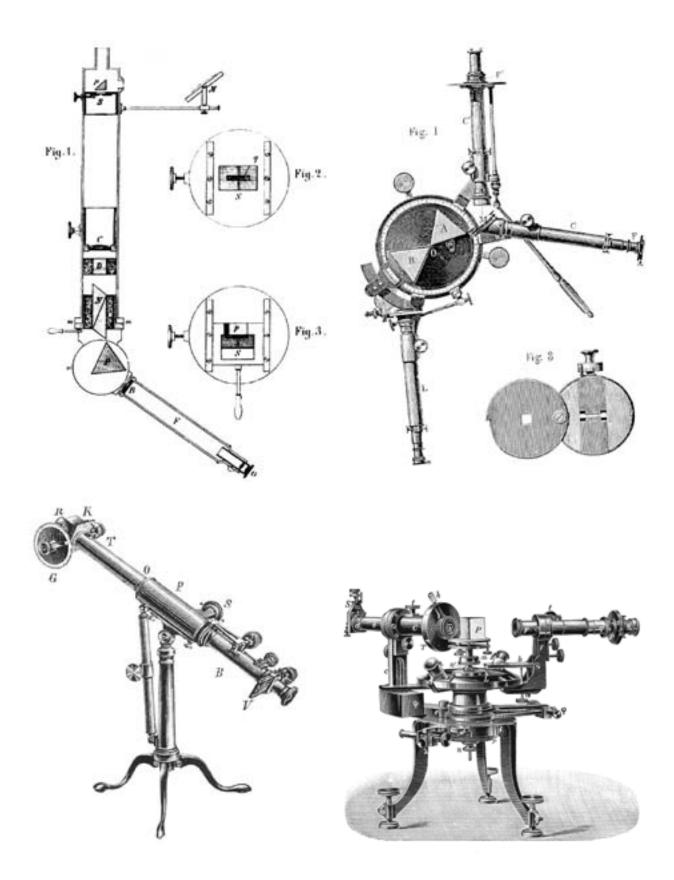


Figure 29-13. Spectrophotometers from around 1880, all incorporating Nicol prisms. Top left: The instrument of Vogel (1877) intended for solar observations. Right: The photometer of Gouy (1879) for research on colored flames. Bottom left: Crova's (1883) meter for analysis of aqueous solution by absorption (Krüss and Krüss 1909). The last two, as well as the photometers of Govi, Violle and Melander, were produced commercially by Ph. Pellin. Right: Glan's (1877, and later) instrument, here installed in one arm of a spectrometer (Liebisch 1891) to study e.g. light absorption in crystals.

13.2. Brücke's friend H. v. Helmholtz revived in the 1850s a largely forgotten theory of T. Young regarding three different color sensors in the human eye: these are the cone cells of the retina which are sensitive to light in three overlapping ranges of the spectrum (see König 1886). Brücke (1866; Ditscheiner 1871) later described a device with Iceland spar prisms which he called a schistoscope, in a book intended to teach people in art industries about colors. Some other papers on color-vision investigations with the aid of Nicol prisms which have come to my attention are by E. Rose (1863, 1865), Dobrowolsky (1872, 1876), Raehlmann (1874), Spottiswoode (1874a), Glan (1881), and Rayleigh (1881b) who continued experiments in this field carried out by J.C. Maxwell with glass prisms before 1860. In a 1906 catalog of physical instruments, the Cambridge Scientific Instrument Co. offers apparatus with Nicol prisms (possibly based on the Glazebrook spectrophotometer) and a quartz plate, for use among other things in tests for color blindness by Rayleigh's method.

Further development of equipment containing Nicol prisms and crystal plates for research on various aspects of color included the leucoscope (König 1882) and the chromoscope (Arons 1910, 1912, Fig. 29-20). These instruments were used by many scientists studying human perception of light and colors (e.g. Brodhun 1888, Schuster 1890, Hering 1890, König 1897, Exner 1902, Lummer 1905, Priest 1920), as well as for the calibration of some industrial color standards.

Around or before 1878, v. Helmholtz invented vision-testing apparatus where a researcher could by means of Nicol prisms and other components vary the intensity and color of two light beams viewed simultaneously by a subject (Fig. 29-20). An improved version was constructed at the works of Schmidt and Haensch (1893), one such instrument being presented to Helmholtz on his 70th birthday by the union of German makers of scientific equipment. König and Dieterici (1893) who describe this color-mixing apparatus, made extensive investigations on the color sensitivity of the eye. It was being produced at least until 1928 and found use into the 1950s. Other papers on its application include Tonn (1894), Kries and Nagel (1896), Allen (1902), Engelking (1925), Kravkov (1927) and Hecht and Shlaer (1936) dealing mostly with various aspects of color blindness, and Kohlrausch (1923) on sensitivity for changes in light intensity. The Helmholtz instrument was also to some extent used as a spectrophotometer, such as in Trendelenburg's (1905) studies on rhodopsin from the eyes of animals. The Young-Helmholtz theory of color perception, to which J.C. Maxwell later made important contributions, always had great influence in vision research, and it seems to be still considered valid in many respects. However, much debate took place between followers of this theory and a rival one proposed by E. Hering whose work was referred to above. The Young-Helmholtz theory may eventually have absorbed some of Hering's viewpoints, for instance regarding how the brain processes nerve signals from the eye.

Numerous other applications for color mixers with Nicol prisms and quartz plates were found, such as: comparison of light emitted by different lamp types (König 1882), choice of colors in the manufacture of for instance textiles or decorative objects (Borchardt 1913), development of light filters for the printing and photographic industries (Jones 1914), and the creation of artificial day-light (Priest 1918). One more field of application concerned the estimation of high temperatures, as in furnaces, kilns and incinerators (see Nichols 1879, 1880, Priest 1921, Fig. 29-20, Skinner 1923). A handy telescope device for this purpose was sold by the firm of E. Ducretet (1889, Fig. 29-17) for decades (cf. Struers 1925). Burgess and Le Chatelier (1912, p. 348-350) consider this Ducretet pyrometer to be quite inaccurate, while various authors (McWilliam and Longmuir 1907, Jüptner 1908, Arnold 1917-18) speak favorably of it in connection with for instance production of steel and coal gas, as well as in the ceramics industry.

Bernard (1856) and Wild (1876) modified their polarizing equipment in order to produce a color resembling that of the sky, but Arago (Fig. 13-3) had in fact initiated similar experiments with more primitive means long before them. Pellin (1899, 1913) advertises a "chromatometer"

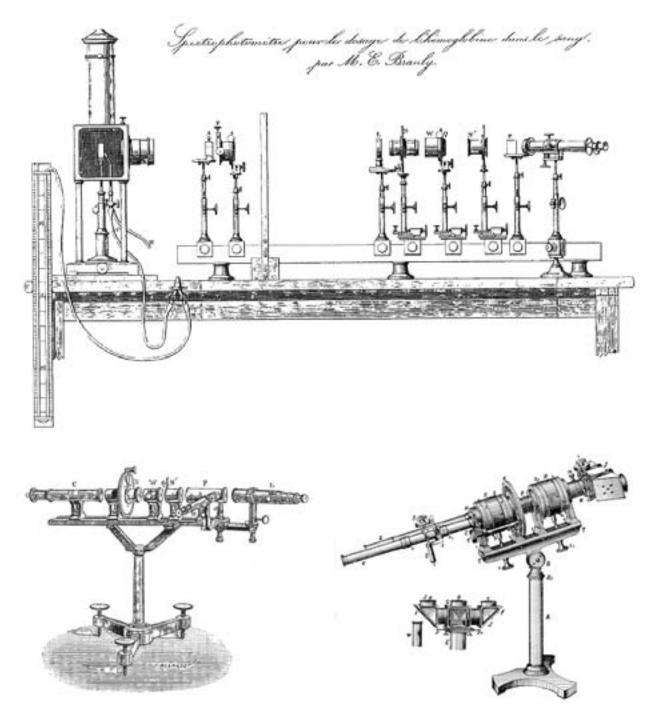


Figure 29-14. More spectrophotometers. Top: The equipment used by Branly (1882) in his work on hemoglobin. Bottom left: Violle's (1884) meter, employed in research on incandescent surfaces. Right: Wild's (1883) instrument (Krüss and Krüss 1909).

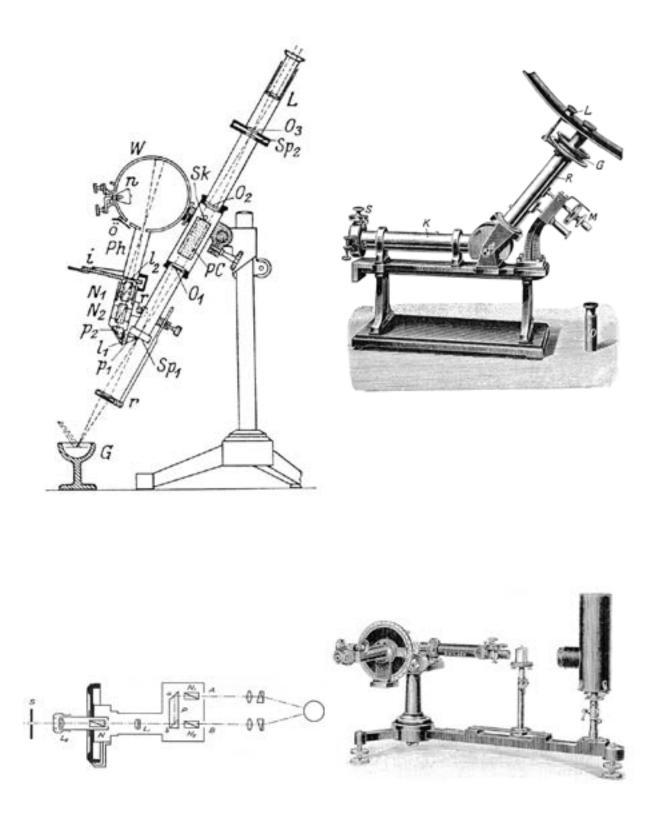


Figure 29-15. Spectrophotometers after 1890. Top left: The meter of Hauer and Kowalski (1914) in the process of measuring luminescence in a liquid. The light source causing the luminescence is omitted here. Right: König's (1894) spectrophotometer which was improved by Martens and Grünbaum (1903) and marketed for decades by Schmidt & Haensch. From Weigert (1927). Light which enters from the left is dispersed by a glass prism at the bend, and then its intensity is estimated with the aid of Wollaston and Nicol prisms in the upper tube. Bottom left: Nutting (1903 and later) invented some instruments for studying light and colors. One which was produced by A. Hilger for various industrial applications, is shown schematically here (Walsh 1958). Right: G. Hüfner (1877 and later) described some new spectrophotometers, such as this one intended for research on blood. From Mees and Sheppard (1904).

invented by L. Andrieu (1886, Fig. 29-20) which was primarily intended for detailed observations on liquid industrial dyestuffs. It could also be used for monitoring colors of food pigments, wine, urine, blood etc. Weinschenk (1925, p. 92-94) and Rosenbusch (1924, p. 592-593) describe a similar method for studies of mineral colors in thin sections under the microscope. W. Ostwald (1919) introduced a color mixer nicknamed Pomi (= Polarisations-farbenmischapparat), containing Nicol and Wollaston prisms. This was in connection with novel theories of colors on which he and others published much from around 1916 until the 1930s. A sketch of this simple device may be found in Handbuch der Physik 19, p. 683, 1928. Ostwald was a great admirer of the poet J.W. Goethe and probably adopted some of his views on color perception (see section 7.3). While Ostwald's ideas in this field were controversial (see e.g. Kohlrausch 1920), they had wide influence on the standardization of colors for industrial and commercial purposes (cf. entry on Ostwald in D.S.B. Supplement I; Pulfrich 1925, Weigert 1927, p. 276-277).

Spectra could be manipulated in more ways with quartz plates. Thus, Mascart (1874, p. 396) wanted in some of his experiments to work with only one of the two closely spaced yellow lines of the sodium spectrum. He found a clever method of eliminating the other one by destructive interference, by letting his light beam traverse a quartz plate of 3.16 cm thickness between two Nicol prisms. Fabry and Perot (1900) used the same technique in early studies on the effects of external conditions on the structure and stability of spectral lines. So did Voss (1918) when measuring the ratio of the intensities of the sodium lines, and R.W. Wood (e.g. Wood and Dunoyer 1914, Wood and Mohler 1918) applied it in important researches on the "resonance radiation" of sodium and some other elements (see section 39.3). Attempts were also made to use the wavelength-dependent rotation in quartz plates cut perpendicular to the optic axis for spectroscopic purposes (Tait 1880).

Nutting (1913) described instrumentation for measuring the intensity and color composition of light reflected from surfaces, developed at the U.S. National Bureau of Standards and sold by A. Hilger. It did not contain quartz plates; instead, four or six Nicol prisms were used to attenuate different light beams (Fig. 29-20, see Fleury 1930).

The ordinary polarization spectrophotometers like those of Glan or König-Martens often provided service in testing the sensitivity of the eye to differences in color or in intensity, either on their own or in combination with other instruments (Bohn 1874, Trannin 1876, Salomonson and Schoute 1904, Kohlrausch 1920). Bezold (1876) and König and Brodhun (1888) also carried out fundamental research in that field, employing Nicol prisms and an Iceland spar rhomb. With the projectors of Duboscq, Mach, Cheshire and Govi for polarized light which were mentioned in section 22.4, certain aspects of vision could be tested such as the permanence of images in the retina. W. Abney used an Iceland spar double-image prism in a part of his extensive research on light and color vision (Abney and Festing 1888), also Hering (1905) and Houstoun (1918). Lummer (1909, p. 401-402), Hecht and Williams (1922) and others employed a pair of Nicol prisms for attenuating light beams when studying human color perception.

Various material properties of the tissues of the eye were investigated with the aid of calcite plates (Carion 1853) and later with instruments containing Nicol prisms, for instance by Nordenson (1921). Finally it may be mentioned that ophthalmoscopes (or -meters) based on double refraction in quartz or Iceland spar prisms were invented. One such instrument with a quartz Wollaston prism (Javal and Schiötz 1881) was acclaimed as a significant achievement in research on astigmatism, as may be deduced from advertisements for similar instruments and patent applications in the following decades. See also the paper by Salomonson (1919) on an ophthalmoscope with Nicol prisms, and especially the publications of Koeppe (1921, etc.) who studied eye defects with polarized light from a lamp invented by A. Gullstrand (cf. Appendix 5). Gullstrand (1906) himself also employed Nicol prisms in some of his ophthalmological work.

29.8 Research on ultraviolet radiation

The absorption of ultraviolet light is less in quartz, fluorspar and calcite than in glass. According to Lowry (1935, p. 213), a 4-cm thick Iceland spar plate transmits rays down to 240 nm wavelength, while thinner plates can be used to 220 nm or so (Liveing and Dewar 1882, and other sources). Radiation down to 180-200 nm passes through quartz prisms, whereas light crown glass only transmits down to 360 nm and heavy flint glass stops anything shorter than 390 nm (Handbuch der Physik 19, 1928, p. 852; Houstoun 1927). Iceland spar was therefore chosen for dispersing prisms in some optical equipment operating with ultraviolet light in the late 19th century and into the 20th. One of the first instances of its use was in E. Mascart's (1864) measurements of the wavelengths of the solar absorption lines. During this work he measured the index of refraction for the ordinary ray in Iceland spar as noted in section 21.2, to 320 nm. He selected Iceland spar because "its great dispersion of the ordinary ray spectrum allows one to distinguish many more lines" than in the case of quartz prisms. Around 1867, Mascart 1869, Soret 1878). Some makers or suppliers of scientific equipment advertised Iceland spar 60°- prisms, among them Adam Hilger who was mentioned at the beginning of chapter 25, and B. Halle (ca. 1895)

A. Cornu who has also appeared before in this compilation, began studying solar radiation in the early 1870s (Cornu 1874b, 1880, etc.). He designed for this purpose a spectroscope with a dispersing prism of Iceland spar (Cornu 1879). He considered this prism to resolve adjacent spectral lines better than the gratings then available. Cornu used compound lenses of quartz and Iceland spar until 1879, and then switched to quartz/fluorspar lenses. Others who were researching ultraviolet light with Iceland spar prisms around 1880 included Monckhoven (1877, radiation from gases), Schönn (1880, radiation from metals; absorption in various materials) and Hartley (1882, ultraviolet emission spectra of the vapors of 24 metals). Hartley, however, mostly employed quartz prisms in his series of experiments.

W. Huggins who had since around 1860 investigated lines in the visible spectrum from stars, planets, nebulae and comets, was also interested in extending his observations into the ultraviolet. He succeeded with a new instrument equipped with quartz lenses (Huggins 1877, 1880a) where a 60° Iceland spar prism dispersed light from a reflector telescope on to a photographic plate (Fig. 29-21) in long exposures; photographic techniques were advancing rapidly at that time. Later, Huggins designed another spectroscope with two Iceland spar prisms in tandem, for improved resolution. He analysed light from many stars in this way to the late 1890s (see e.g. Huggins 1895, a news item in The Times 9 June of that year, and his obituary in Proc. Royal Soc. A86, 1912). Some of these spectra appeared in the Atlas of Huggins and Huggins (1899). In a review of that publication by G.E. Hale (in Astrophys. J. 12, 1900) it is indicated that Huggins' research was a milestone in endeavors to classify stars and understand their properties. This view is supported by later writers on the subject (cf. chapter 14 of King 1955). Huggins also obtained an ultraviolet spectrum from e.g. the Orion nebula with his Iceland spar prisms.

A most noteworthy discovery made by Huggins (1880a) concerned atomic hydrogen. It emits visible light of four spectral lines, and it appeared possible that their wavelengths might be expressed by a mathematical relation with a single variable. N. Lockyer had found a fifth spectral line in an experiment described in a short paper in 1880; that line was at 397 nm near the boundary between the visible and ultraviolet field. In the sun's spectrum, it coincided with a strong calcium line. J. Balmer who was one of those attempting to find an empirical formula for the hydrogen lines, originally did not know of Lockyer's result. He then learned that H.W. Vogel (1879, 1880)

had observed that line and four other ultraviolet ones in his laboratory (with glass prisms) and that these were prominent in Huggins' spectra of white stars (Fig. 29-21). Balmer (1885) subsequently published a simple formula which fitted all the lines with remarkable precision. It was $h.m^2/(m^2 - 4)$ where h = 364.5 nm and m = 3, 4, 5, ... 11. In an appendix to his paper Balmer notes that Huggins had in fact discerned five more hydrogen lines, whose wavelengths agreed with his formula for m = 12 to 16.

Huggins (1880b) and Liveing and Dewar (1882, 1889) investigated the ultraviolet spectrum of very hot water vapor with Iceland spar prisms; the latter authors also studied the absorption- and emission spectra of many metals, noble gases, carbon and its compounds, and other substances with reflection gratings and Iceland spar. From 1879, they realized that the spectra of elements recorded in their photographs and measurements (e.g. Liveing and Dewar 1883) could be separated into two groups of lines having different appearance ("sharp" and "diffuse"). They also found regular patterns in the wavelengths of various visible and ultraviolet lines from for instance the alkali metals and magnesium, analogous to the Balmer series of hydrogen. The above-mentioned A. Cornu who was at around this time studying ultraviolet radiation from metal vapors with Iceland spar prisms in his laboratory, took a new look at the hydrogen spectrum and managed to confirm the presence of almost all of the ultraviolet lines seen by Huggins (Cornu 1886). Here he used a diffraction grating, but does not mention Iceland spar. Zenger (1889) described means of improving upon Cornu's technique.

From around 1885, H. Deslandres (1888, and others) continued Cornu's research with his equipment, to some extent employing Iceland spar prisms in work on the spectra of gases. He was the first to study in detail many aspects of so-called band spectra of molecules. These spectra are caused by changes in the rotation of the molecules, internal oscillations, or electronic rearrangements. He (Deslandres 1886) found among other things a very regular pattern in the band spectrum of nitrogen, especially in ultraviolet light. Deslandres made further discoveries in the field of molecular spectra over a long period from 1888, but by then he had started using glass prisms and gratings instead of Iceland spar. He also studied the sun's corona, its chromosphere and surface phenomena. He and others like G.E. Hale and N. Lockyer confirmed the presence of Huggins' ultraviolet hydrogen lines in spectra of light from these regions of the sun in the years between 1892 and 1900, both during eclipses and at other times. Balmer's formula presumably aided in recognizing them in a myriad of other spectral lines from the sun. Deslandres (e.g. 1893) and others such as Copeland (1900) sometimes used equipment with Iceland spar prisms or lenses in these solar investigations, but different types of instrumentation had generally replaced them before 1900.

Until 1880 various scientists had tried to come up with empirical formulas or physical models for the many spectral lines emitted by the elements: in some cases these lines appeared to have some regularity, in other cases not. Often it was assumed that they corresponded to harmonics in an oscillating system (see Schuster 1882). In summary however (Kayser and Runge 1888), it was admitted that these attempts had been "complete failures" and that the matter had been dormant for a few years when Balmer (1885) "got things moving again". In 1885, Cornu had also pointed out certain analogies in the ultraviolet spectra of hydrogen and some metals. King (1955, p. 293) and other historical accounts stress that the ultraviolet studies by Huggins, Hartley, Liveing and Dewar, Cornu, and Deslandres played a role in laying the foundation for new extensive research on spectra. This refers in particular to the work of Kayser and Runge (1888 and later) who decided to carry out detailed mapping of the spectral lines of many elements with the latest techniques in gratings and photography. New relations were soon found between wavelengths, both within individual spectra and between elements (e.g. Runge 1888). The ultraviolet data were quite important in this work, because the grouping of lines into different series was most clear in that part of the

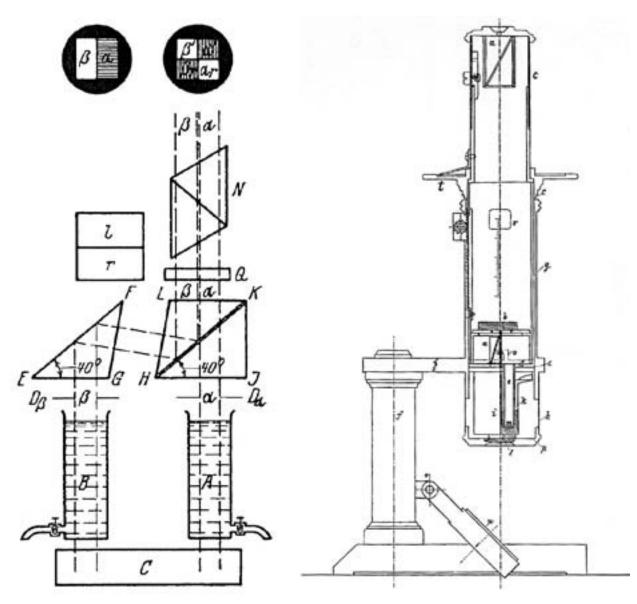


Figure 29-16. Two colorimeters. Left: G. and H. Krüss (e.g. 1909) designed and manufactured both spectrophotometers and colorimeters, some with Nicol prisms. The present example was used for estimating the concentration of metallic salts and other compounds in aqueous solutions. Diagram from Handbuch der Physik 19, 1928. Right: Meisling's (1904) colorimeter for measuring hemoglobin in solution.

spectra. The relations were generally represented by simple but accurate mathematical formulas, resembling that of Balmer. Rydberg (1890) also deduced even simpler rules from the wavelengths obtained by Liveing and Dewar, especially one stating that the sum of frequencies of two lines often equalled the frequency of a third line. A related rule said that the difference in frequencies of two lines of the same element was often equal to that between another pair (or pairs) of its lines.

These researches on spectra had a decisive influence in the development of modern physics. Rydberg's rules on the sums and differences of frequencies became easily understandable as being consequences of the quantized energy levels of N. Bohr (1913) and of the conclusion by A. Einstein (1905a) that the energy of a light quantum is proportional to its frequency. Kayser and Runge found additional examples of Rydberg's rules, and other investigators (such as W. Ritz in the years 1903-09) expanded these rules. Balmer's (1885) formula for the hydrogen spectral lines was a cornerstone of Bohr's theories; when altogether 33 lines of that series had been measured in stellar spectra, Bohr (1913) stated that "The agreement between the theoretical and observed

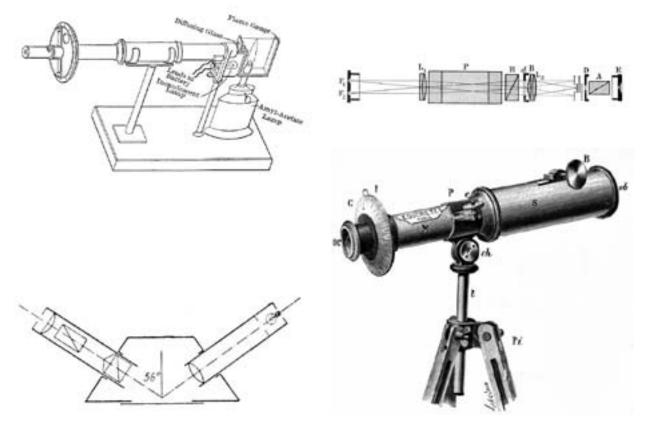


Figure 29-17. Specialized photometers. Top: Wanner's (1902) pyrometer, produced in quantity for high-temperature scientific research as well as industrial applications. P is a glass dispersing prism, R is a Rochon prism and A is a Nicol prism. The instrument is calibrated with the gas lamp. From Burgess and Le Chatelier (1912) and Ribaud (1931). Bottom left: A simplified cross-section (Wien-Harms 1928) of the gloss meter designed by Kieser (1919) for use in the paper industry. It contains Nicol and Wollaston prisms. Right: Ducretet's (1889) pyrometer telescope for furnaces, with Nicol prisms and a quartz plate. The illustration is from a 1902 catalog.

values is inside the uncertainty due to experimental errors...".

Cornu (e.g. 1878, 1881) noticed that the spectra of the sun and stars faded out completely at wavelengths shorter than 290-300 nm, and he ascertained that they were absorbed by the earth's atmosphere. Hartley (1881), Cornu (1884) and Huggins (1889) provided evidence that ozone might be responsible, which was indeed confirmed later (see section 36.7). Cornu and Huggins employed Iceland spar prisms in their observations, at least in part. Huggins and Huggins (1890) found from their starlight exposures, that the ozone also absorbed some light in the wavelength interval 320-334 nm.

Others who were studying ultraviolet light from 1874 or earlier, used gratings for its spectral decomposition rather than prisms. The gratings along with prisms and lenses from quartz and fluorspar gradually took over the role of Iceland spar in this field, for instance in the extensive work by Kayser and Runge mentioned above. However, achromatic spar-quartz lenses for research in physics and astronomy remained in some demand until 1900 at least (see e.g. Nature 56, p. 34, 1897 and J.W. Gifford in British Assoc. Report 1900, p. 630-631), although V. Schumann had declared in 1893 (quoted by Leiss 1899b, p. 63) that they offered no advantage over lenses made from quartz only. Around 1905 new types of glass (uviol) which passed wavelengths down to 300 nm, appeared on the market; however, some scientists continued using the quartz-spar lenses for a while yet, such as Lowry and Vernon (1928) in studies on the optical activity of tartrates. Attempts were made to employ lenses from spar in the ultraviolet to replace Nicol prisms, in order to avoid loss of light by absorption and reflection (see Schulz 1925 and section 33.2).

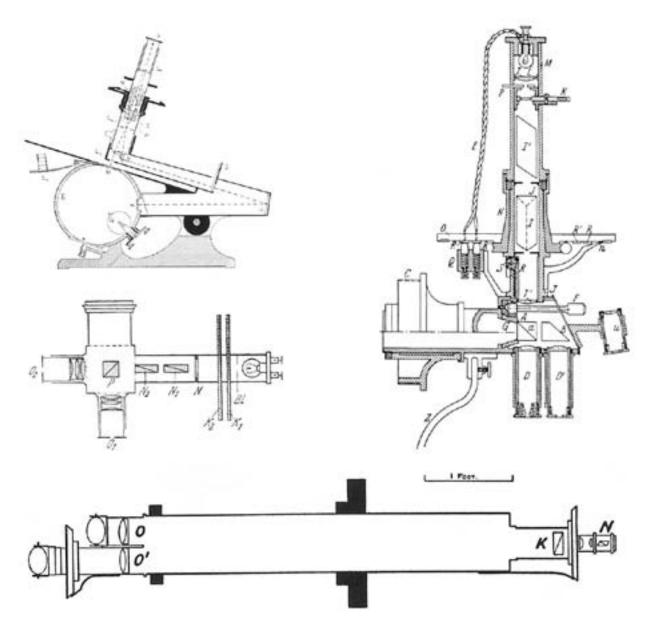
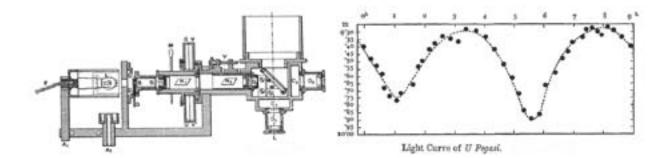


Figure 29-18. Photometers for astronomical research. Top left: The Martens (1900) photometer, here set up for measurements of the darkening of photographic plates (from e.g. astronomical telescopes or spectroscopes). The observer uses Iceland spar prisms to attenuate light received directly, until it matches the intensity of light that has been partly absorbed on passing through such a glass plate. From Weigert (1927). Center left: Rosenberg's (1921) astrophotometer. Light from the Moon or other heavenly bodies falls from above on to the Lummer-Brodhun glass prism P. An observer in O1 or O2 compares it to the light from the bulb, which may be dimmed with the Nicol prisms or the colored glass wedges K1, K2. Right: E. Schoenberg'photometer from 1917 (Hellerich 1936) for measuring light from the Moon and planets. The light beam enters from left in the lower part of the equipment, and an observer at D compares it with light from the bulb at the top. Bottom: Pickering's (1882) meridian photometer, one of several photometric instruments that he designed and used with his observatory staff for measurements on tens of thousands of stars. Light from two stars coming through O and O' is compared with the aid of the quartz/glass Rochon prism K and the Nicol prism N (Walsh 1958).

Iceland spar and/or quartz prisms continued being employed for the dispersion of ultraviolet light in various studies on refractive indices, emission or absorption of other materials. P. Pellin advertises them in a joint French presentation of scientific instruments in 1901-02, but H. Buisson and C. Fabry indicate in a 1911 paper that they are rarely needed. In addition to works already mentioned, these prisms were used by e.g. Chardonnet (1881), Schaik (1882), Soret and Sarasin (1882), Hartley (1883), Joubin (1889, Fig. 29-21), Guye (1889, optical activity of sodium



Nom de l'étoile.	$\log \frac{R}{B}$.	Température effective absolue.	Type spectral d'après	
			Lockyer.	Harvard College.
o Persée	-0,230	2980	Antarian	M <i>b</i>
ζ Céphée	0,552	4610	Aldebarian	K
ô Céphée (min.)	0,592	4950	Polarian	G
Soleil	—0,690	5990	Arcturian	G
γ Cygne	-0,710	6330	Polarian	F 8 G Péc.
ở Céphée (max.)	0,788	7940	Polarian	G

Figure 29-19. More on the photometry and colorimetry of star light. Top left: The astrophotometer of Nordmann (1909a) with Nicol prisms and liquid color filters. Light from a star coming down the telescope on the right is compared to an electric light source. Right: O.C. Wendell's results (Roberts 1906) on the magnitude of a certain Algol-type variable (i.e. eclipsing binary) with a 9-hour period. One of E.C. Pickering's polarization photometers was used for these measurements. Bottom: Part of Nordmann's (1909b) table of effective surface temperatures of various stars, based on estimates from his heterochrome photometer. Later, these temperature values were found to be rather inaccurate.

chlorate), Schunck (1901), Liveing and Dewar (1901a,b, spectra of noble gases), Gramont (1909, spectra of metal vapors in electric sparks), and Bruhat and Pauthenier (1927, Fig. 29-22). In particular, Gramont's research was an important step towards the application of emission spectra in analytical chemistry.

Nicol prisms also served in the ultraviolet, especially in research on rotatory dispersion of light. For this purpose, prisms with air spacing instead of glue were preferred, such as those named after Foucault (1857) or Glan (1880). This is because resins like the commonly used Canada balsam start absorbing at wavelengths shorter than ca. 340 or 320 nm (Cabannes 1921) while Iceland spar itself is transparent down to around 220 nm as mentioned above. Research in this field was carried out by Soret (1878), Soret and Sarasin (1875, 1882, Fig. 29-21), Guye (1889), Joubin (1889), Gumlich (1898), Lowry (1908), Descamps (1926), Bruhat and Pauthenier (1927), and others. Among those continuing A. Cotton's work (section 22.3) on rotatory dispersion was Darmois (1911) who measured it in camphor, pinene and other compounds with an ultraviolet spectroscope made entirely from Iceland spar. This property later became very useful in investigations of the structure of larger organic molecules (see Darmois 1922 and section 39.1).

In ultraviolet research from around 1900, new intense light sources were becoming available, encased in quartz (Küch and Retchinsky 1906) or uviol glass. Sparks between metal electrodes were also used, e.g. when the ultraviolet absorption of various dyestuffs and types of glass was being measured in a novel spectrophotometer with Nicol prisms (Krüss 1903). New methods of analyzing the state of polarization of ultraviolet light appeared, such as one suggested by Voigt (1901). It was applied by Minor (1902, Fig. 29-22), Nutting (1903a) and later others in studies of

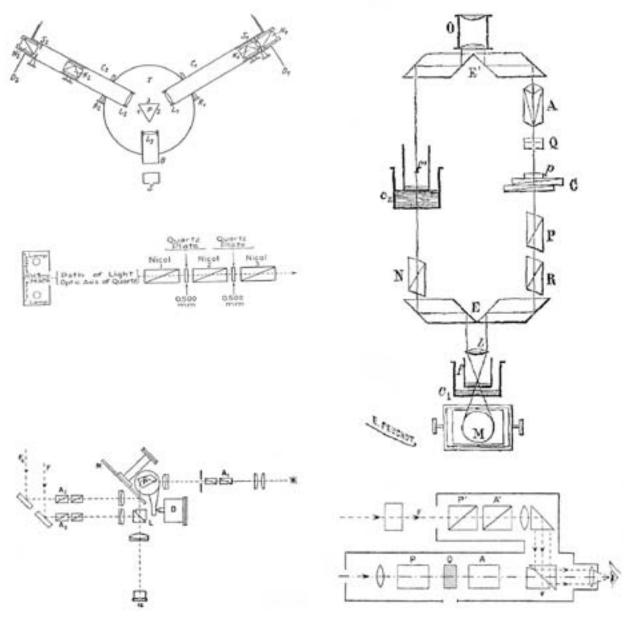


Figure 29-20. Instruments for research on colors, illumination and human vision. These might contain 2-6 Nicol prisms and sometimes also quartz plates. Top left: The color- mixing apparatus used by H.v. Helmholtz from around 1878, for testing color perception in a subject who looks through the aperture S. The apparatus was improved by A. König (Schmidt & Haensch 1893), the diagram is taken from König's chapter in Wien-Harms (1928). Center left: Schematic diagram of Priest's (1921) equipment for charting the spectral distribution of electrical illumination. Bottom left: An instrument produced by A. Hilger for measuring the colors of surfaces, based on a concept from Nutting (1913). Top right: Andrieu's (1886) equipment for the quantitative description of colors of various liquids such as wine. Bottom right: The chromoscope of Arons (1910, 1912), in which an experimenter can produce any desired color from a white light source (Fleury 1930).

light reflection from surfaces of various materials. See more on this in section 36.9.

Iceland spar sometimes also had a role in experiments with infrared light. Thus, Mouton (1877, 1879) used spar prisms in studies on the reflection of heat radiation from metallic mirrors, and two "enormous nicols of 5 cm aperture" when determining the wavelengths of this radiation by means of the method of Fizeau and Foucault (1847). Javal (1905) dispersed infrared rays with an Iceland spar prism when measuring their transmission through metal foils. Ingersoll (1906, 1910, 1917, Fig. 29-22) used a calcite beam-splitting prism when measuring the Faraday effect in water for infrared wavelengths, and in studying reflection from metals. In 1906 it was pointed out that a

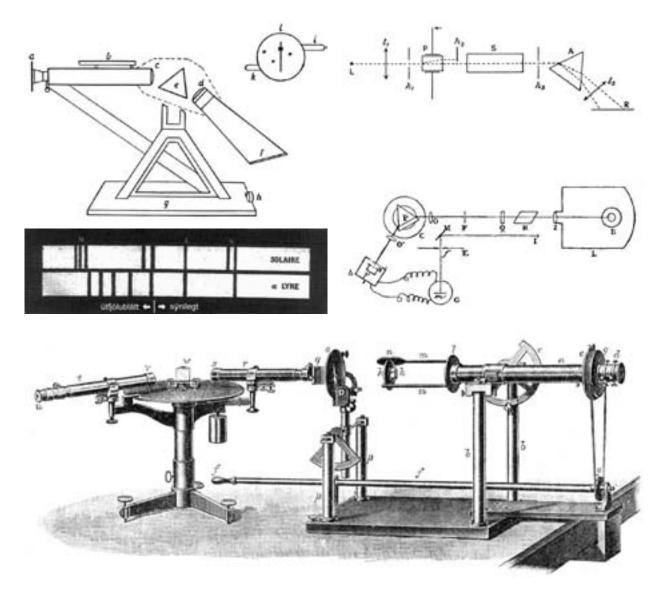


Figure 29-21. Research on ultraviolet and infrared light, before 1900. Top left: Huggins' (1880a) spectroscope with quartz lenses, for analysing ultraviolet light from young stars. The light enters through a slot in a on the left-hand side, is dispersed by the Iceland spar prism e and falls on a photographic plate at f (King 1955). Spectra obtained with this instrument (Huggins 1877) are also shown. Top right: Joubin's (1889) experiment on the Faraday effect in liquids S in ultraviolet light. P and A are Iceland spar prisms, R is a photographic plate (Lowry 1935). Below: Carvallo's (1892) setup to measure optical rotatory dispersion in the infrared. P and R are made of Iceland spar. The intensity of radiation received by a thermoelectric sensor is monitored by means of the galvanometer G. Bottom: The equipment used by Soret and Sarasin (1882) for measuring ultraviolet rotatory dispersion in quartz. The dispersion prism w in their experiments was often one made from Iceland spar. The figure is taken from Tutton (1922).

smooth selenium reflector was a better polarizer for infrared radiation than a Nicol prism, and this technique was utilized by for instance Nyswander (1909) and Schaefer and Schubert (1916). Iceland spar however continued being used where infrared light of certain well-defined wavelengths was needed ("Reststrahlen"-method, see section 34.2), as by Czerny (1924) and Krebs (1927). Lowry (1935, p. 230-232) mentions the use of triangular Iceland spar prisms in connection with observations on the optical activity of quartz up to 2000 nm, carried out by Carvallo (1892, Fig. 29-21) and shortly thereafter also by A. Hussell and by R. Dongier.

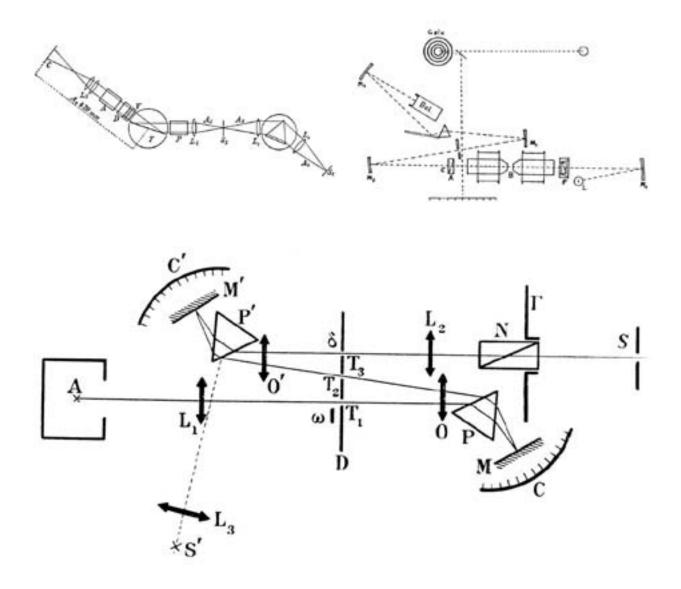


Figure 29-22. Techniques in research on ultraviolet and infrared light after 1900. Top left: Minor's (1902) measurements on the optical properties of metals. Linearly polarized monochromatic light from the Nicol prism P falls on a polished metal plate on the rotatable platform T. Its state of polarization is then analyzed with the Nicol prism A and the quartz plates V, D. Right: The experiment of Ingersoll (1917) on the Faraday effect in various materials, using infrared light from the lamp L. Samples for measuring are placed in B, P is a Nicol prism and A is a beamsplitter prism, probably from Iceland spar. Bottom: Equipment of Bruhat and Pauthenier (1927) for producing ultraviolet light of a narrow spectral interval (monochromateur). A is a mercury lamp, P and P' are dispersing prisms from quartz or Iceland spar, L1 and L2 are composite quartz-calcite lenses, and N is a Nicol prism. The light is then used in experiments on the other side of the screen S.

29.9 Biological research; microchemistry

According to Lecher (1917, p. 290) and Köhler (1926), polarizing microscopes have been of much use to biologists when probing various animal and human organs. In histology, research on muscles that had started around the middle of the century as noted in section 18.3, was continued by for instance Rollett (1891), Engelmann (1895) and Hürthle (1909). Adami and Aschoff (1906) pointed out that double refraction is a very characteristic property of myelins, an essential constituent of nerve tissue. Schmidt (1920, 1924) confirms that examination in a polarizing microscope is an important aid in ascertaining the structure of many tissues in addition to nerves and muscles:

he presents examples concerning bones, teeth (Ebner 1906), sinews, hair, horn, scales, shells and exoskeletons of invertebrates, foraminifera, corals, pearls, sponges and so on. See for instance thorough studies by Kelly (1901) and Bütschli (1908) on calcareous skeletons and other deposits in tens of animal species.

In a review of Schmidt's (1924) book (Naturwissenschaften 12, p. 745-746), polarizing microscopes are said to have been for a long time an indispensable tool in human histology and pathology. By cursory inspection of publications from that period, it is evident that this often relates to the nature of a variety of pathological conditions, such as in the digestive system (see Ebstein 1888 and Schmidt 1924 on kidney- and gallstones, Panzer 1906 on inflammations), eyes, skin, cancerous tumors (White 1909), spinal fluids (Donath 1905) and so on. Among the commonest of such conditions are deposits of liquid crystals with a clear double refraction, including cholesterol and myelin; see section 27.2, Kaiserling and Orgler (1902), Löhlein (1905) and Adami and Aschoff (1906). Anitschkow and Chalatow (1913) found that if rabbits ingest food rich in cholesterol, it accumulates in various organs where its presence can be demonstrated by a polarizing microscope. This discovery was not followed up much in the next 30-40 years with regard to a connection between cholesterol in human food and in the cardiovascular system, but it was later seen to be quite important. Microscopes were also used in the characterization of hundreds of new crystalline materials isolated or synthesized during biochemical research, e.g. nucleic acids (Kossel and Neumann 1893), glycosides (Koenigs and Knorr 1901), the bile acids studied by Wieland and Sorge (1916, Anhang), and alkaloids (Willstätter et al. 1923). A notable work in this general field was the study by Reichert and Brown (1909) on hemoglobins from a large number of vertebrates.

Turning to the vegetable kingdom, one of the pioneers in studying plant tissues by polarizing microscopes was H. Ambronn. He had towards the end of the 19th century used such microscopes when investigating colloid suspensions of matter in liquids, but in 1913-19 he carried out note-worthy research (e.g. Ambronn 1916) on the structure of cellulose. Here one had to distinguish between the conventional double refraction of crystalline substances and an analogous phenomenon caused by the shape of grains or grain parts, even if they are composed of isotropic materials. This phenomenon which was called rod- or plate- refraction depending on circumstances, had in fact been a subject of discussion in 1840-60; it was revived by Wiener (1904) as a consequence of his experiments mentioned in section 28.4. Ambronn himself revived C. Nägeli's micellar theory of cell structure (see final paragraph of section 18.3), which had been largely forgotten for twenty years. Additionally, Ambronn and others studied the optical properties of various gels (e.g. Neubert 1925).

The double refraction observed in the fibers of many industrially valuable organic materials (wool, cotton, hemp, silk, etc.) is due to their being composed of elongated sub-parallel molecules. Polarizing microscopes have been of much use in research on such fibers in the 20th century (cf. Herzog 1909, Harrison 1918, Frey et al. 1926, and Hartshorne and Stuart 1950, p. 439-452). Treatment of fiber samples with certain colored chemicals, metal salts, iodine or other compounds (dichroic staining) can enhance their double refraction, see for instance Ambronn (1896), Frey (1925), Ambronn and Frey (1926, p. 176-185), and McGraw-Hill (1992). A particular application of that technique will be introduced in section 35.2; in recent years it has also been employed in combination with liquid crystals in digital and television displays.

For some decades, biologists were not quite happy (see Ebner 1892) with the polarizing microscopes on the market, which was dominated by petrographers. Eventually, the Zeiss company offered some accessories for their benefit, upon the advice of the physiologist T. Engelmann (Siedentopf 1902). The Leitz workshop sold Nicol- prism arrangements that could be fitted to ordinary biological microscopes (Schmidt 1924, p. 19). Later, Schmidt (1925) persuaded Leitz to

produce a specially designed polarizing microscope for biologists. It was to some extent simpler than the petrographic models, but contained a new type of analyzing prism (Tubusanalysator, see Ehringhaus 1920) which improved the resolution of delicate features. Without doubt, these instruments were not only valuable when studying modern tissues, but also in microscopic paleontology. One example would be in oil exploration, where the fossilized remains of organisms in ocean sediments can reveal much about their age and economic potential.

In publications from even before 1900, many instances may be found where polarizing microscopes were essential in studies of crystallization (see sections 27.2 and 29.1). They could also aid in recognizing materials which were only available to the scientist in very small quantities. Among those contributing to the development of such "microchemical" techniques were K. v. Haushofer in the 1880s, Behrens (1891; Behrens-Kley 1915) who asserts that polarizing prisms are indispensable, and Pozzi-Escot (1900). Emich's (1911) book on the subject gives many examples of polarization techniques in the microscopic identification of compounds. Emich (1916) also points out that small-scale polarimetric observations were of great value to E. Fischer (see Fischer 1911, and Appendix 5) in his research on polypeptides. E.M. Chamot designed a simple microscope with Nicol prisms for chemists, sold by Bausch & Lomb from 1899; see Chamot's (1915) book on applications of these microscopes. Books by Johannsen (1914, chapter XL), Rosenbusch (1924, chapter on microchemical reactions p. 742-761) and Hartshorne and Stuart (1950, p. 422-433) also explain their use in chemical analysis. Wright (1916), Peck (1919), Benedict (1930) and others list cases where polarizing microscopes come in handy in the examination of various crystalline grains (or mixtures of these) in industry: sugar, dyes, alkaloids (Kley 1904), abrasives, photographic developers, amino acids, explosives, fertilizers (see Bowen 1926), raw materials for the production of paper (Dickson 1925) or of glass, and so on. The famous geochemist V.M. Goldschmidt (1920) used a polarizing microscope when inventing new titanium oxide pigments for use in paint. He patented a production process for these compounds, and the titanium white gradually replaced lead white whose properties could have adverse effects on health and the environment. The microscopes also played many roles in connection with research and quality control on food, even butter (Taylor 1885, Bömer 1908, Litterscheid 1924). Chamot (1921, p. 50) and Weinschenk (1925, preface) criticized chemists in general for not making sufficient use of polarizing microscopes.

30 New magnetic and electrical effects on light, 1896-1925

30.1 The Zeeman effect, 1896-1910

In the wake of the discoveries of magnetic effects on light made by M. Faraday and later by J. Kerr, it may be assumed that many people searched for influences of magnetic fields on radiation processes. In 1896, P. Zeeman who had in fact been making experiments on the magnetic Kerr effect, looked carefully at the spectral lines of light from hot sodium vapor (i.e. a flame charged with sodium salt) in a strong magnetic field. Using a high-quality diffraction grating from H.A. Rowland, he noted (Zeeman 1897a) a slight widening of the lines which "...made it likely that a real magneto-optical effect was being observed. This became certain when polarization features were seen to be connected with the widening ... " (Zeeman and Bruin 1927). Here Zeeman is referring to the fact that his compatriot H.A. Lorentz had then recently put forward some considerations on the nature of electrical charges in matter, based on Maxwell's theories. According to Lorentz' ideas, the spectral lines of light emitted in a magnetic field would be split into two or three components respectively, depending on whether the light was being observed along the magnetic field lines or at right angles to the field. It the former case they would be circularly polarized, in the latter linearly polarized. Zeeman (1897a,b) was able to confirm this to a certain extent. The same changes occurred in the absorption spectrum of sodium vapor. In a review by Bohr (1922) of the history of these investigations, he recalls that the sense of rotation in the circularly polarized components showed that the light emanated from negatively charged particles, presumably electrons.

The changes in frequency caused by the magnetic field were proportional to its intensity. From Lorentz' (1897) formulas and Zeeman's measurements on line spectra, a further very important conclusion could be drawn: in the radiation of light from hot atoms, the particles involved had in most cases a charge-to-mass ratio, e/m, of a similar magnitude as those which conducted electricity in rarefied gases. In some instances (the so-called normal Zeeman effect), e/m even agreed reasonably with the value that J.J. Thomson found in 1897 for electrons in deflection experiments on cathode rays. This soon led to much new research activity in related subjects. See the book by Drude (1900a, p. 410) who also points out that the e/m ratio for hydrogen ions is of the order of 2000 times less than for electrons.

Becquerel (1897) proposed a hypothesis on certain relations between the Faraday effect and the Zeeman effect, and his formulas could be used for estimating e/m from the Faraday effect. (The result however tended to be lower than expected, see a paper by C.G. Darwin and W.H. Watson in Proc. Royal Soc. A114, 1927). Larmor (1900, p. 341-356) deduced general theorems on the move-

ment of charged particles in magnetic field, which went farther than Lorentz' theory in explaining the frequency changes and polarization of light in the Zeeman effect. Later, Larmor's theorems also became important as a theoretical model for diamagnetism in materials. With more powerful magnets, sensitive spectral analysis by interference techniques, and reduction of the temperature in the emitting vapors (see e.g. Becquerel and Deslandres 1898, Fig. 30-1; Michelson 1897, 1898) it was soon realized that the Zeeman effect in the minority of cases split spectral lines into the expected two or three components; they could also be four, six or nine, and often of different intensities. This was called "the anomalous Zeeman effect". Nicol prisms (Michelson 1898, Reese 1900, Baeyer and Gehrcke 1909, etc.), Iceland spar rhombs (e.g. Runge and Paschen 1902) or other polarizers (Preston 1899) were useful in ascertaining the state of polarization of the various component lines. Zeeman (1909) demonstrated with the aid of the method of Fizeau and Foucault (1845) that the orbits of electrons in atoms were very close to being of circular shape.

It must be stressed here that all this happened before E. Rutherford's 1911 demonstration of the nature of atoms in collision experiments. Bohr (1922) states that the results of Zeeman and Lorentz were "generally accepted as a most convincing proof of the electronic theory of matter, and...established beyond doubt that the origin of these spectra is to be found in the motion of electrons within atoms.". See Appendix 5.

One of the interference spectroscopes employed in the early 20th century to resolve the Zeeman multiplets, was based on total reflection of light inside a thick plate of glass or quartz (Lummer and Gehrcke 1903, Fig. 30-2). Its performance could be enhanced if the light was made linearly polarized with a Nicol prism before entering the plate, but this was not always needed. The plate and other forms of interference spectroscopes were also used in studies of the fine structure (and later hyperfine structure) of spectral lines. See for instance the analysis by Galli and Försterling (1917) of ultraviolet lines of mercury with a Lummer-Gehrcke plate and a Nicol prism, by Hansen (1925) of the Balmer lines of hydrogen, and by Stauffer (1930) of the spectrum of a chopped light beam, Fig. 38-2. The fine structure of spectra has always been very important in tests of hypotheses on the nature of atoms.

"Zeeman's discovery and further work connected to it has caused great excitement in the world of physics" according to the editors of the Physikalische Zeitschrift, in a brief introduction to a review paper by Voigt (1899-1900). One of the experiments creating much interest was that of Macaluso and Corbino (1898, Fig. 30-1) where hot sodium vapor in a magnetic field exhibited strong optical activity if the frequency of the light sent through it was close to that of sodium's two yellow lines. See more in sections 30.2 and 39.3. In another review paper, Cotton (1899) emphasizes the value of the Zeeman effect for enhanced understanding of the fundamental laws of light-matter interaction. This effect, including its polarization aspects, was very useful in the coming years, for instance when the many spectral lines of the elements were being grouped into different series (Preston 1899, Runge and Paschen 1902).

In the normal Zeeman effect which is rather rare and was first studied closely around 1907 in certain lines from helium (Paschen and Back 1912, p. 897), zink and some other atoms, the frequency changes in a magnetic field agreed precisely with the theoretical predictions of Lorentz. By this time, experimental techniques had progressed so much that the determination of e/m for electrons from the Zeeman splitting (Weiss and Cotton 1907) gave a more accurate value than experiments using J.J. Thomson's method mentioned above. Weiss and Cotton used an "analyzer", probably a Nicol prism, to improve resolution of the spectral lines, and so did also subsequent observers such as Babcock (1923) in more extensive measurements of e/m. Scientists appended various ad hoc conditions to the Lorentz theory in attempts to explain the anomalous Zeeman effect.

G.E. Hale (1908) published results from measurements on frequency shifts and (using Nicol prisms) polarization of the light emitted from sunspots. They definitely revealed the presence

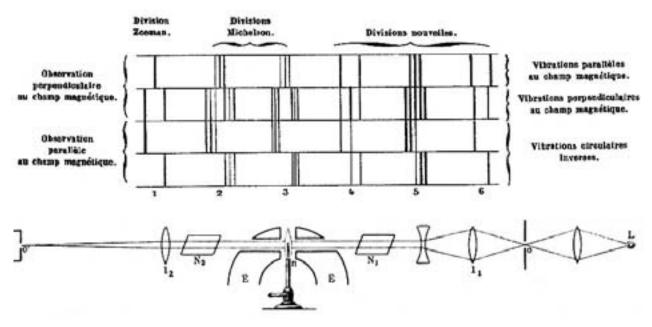


Figure 30-1. Top: Becquerel and Deslandres (1898) photographed spectral lines from incandescent vapors of iron and other metals in a magnetic field through "un beau rhomboèdre de spath d'Islande". On the left we see the simple (normal triplet) Zeeman splitting of spectral lines, while the others are more complex patterns found by A.A. Michelson and the authors. Bottom: A sodium flame in a magnetic field, for research on the Zeeman effect (Macaluso and Corbino 1898). Considerable rotation of the plane of polarization occurs in polarized sodium light on passing through the flame (here, towards the left). The diagram is from Lummer (1909).

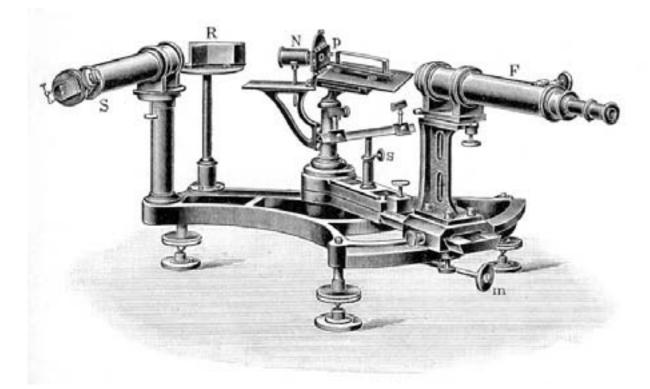


Figure 30-2. Research on the Zeeman effect called for both strong magnetic fields and improved resolution of spectra. One novel device of high dispersion consisted simply of a glass or quartz plate with precisely parallel polished surfaces, invented by Lummer and Gehrcke (1903). It is seen here to the left of p, accompanied by a glass prism R for selecting the wavelength interval (of light from the source S) to be expanded. N is a Nicol prism whose function in this case was to sharpen the spectral lines. From Lummer (1909). of strong magnetic fields around the spots, and according to Dictionary of Scientific Biography (1980-90) this was considered to be the most important step forward in our knowledge of sunspots since the 17th century. With improved instrumentation it was later found (Hale 1913) that the sun as a whole has a magnetic field which at its surface is about a hundred times stronger than that of the earth. Around 1914 Hale and his collaborators had found that changes were taking place in the magnetic field of the sunspots (see Hale's biography by H. Wright, 1966) and that they often occurred in pairs where one was a north magnetic pole and the other a south pole. In 1918-19 they established that the fields of sun spots as well as the main field of the sun changed their polarity at 11-year intervals (see e.g. Hale and Nicholson 1925). All this had great influence on research in astrophysics and solar-terrestrial relations.

In connection with research on the Zeeman effect, as well as in some other fields (such as the use of polarization spectrophotometers), properties of diffraction gratings had to be looked into. This was because a grating may alter the polarization state of light in the spectra that it resolves. See for instance the paper by Ingersoll (1920) which reviews previous work on this aspect.

30.2 More on the Zeeman effect and other magneto- and electro-optic effects connected with polarization, 1900-25

In addition to the Zeeman effect just described, a number of different effects of magnetic and electric effects upon light in materials have been discovered, investigated, and utilized. The Faraday magnetic effect, the Kerr electric and magnetic effects, and the Pockels electric effect have already been mentioned above.

Before 1890 it was known that in the Faraday effect and the Kerr electric effect it took the molecules in material samples less than 0.1 millisecond to respond to externally applied fields. Abraham and Lemoine (1900, Fig. 30-3) demonstrated that in the latter the time lag was only a small fraction of a microsecond. At the turn of the century, practical applications of these effects had also appeared. As an example, one may place a substance having a strong Faraday effect (such as CS_2 , enclosed in a tube) between two crossed Nicol prisms. A light ray cannot pass through this setup until a magnetic field is switched on around the tube. This was used e.g. in measuring the speed of artillery projectiles (Crehore and Squier 1895, Fig. 30-3) and in estimating the amplitude of an oscillating current (Pionchon 1895, Abraham and Buisson 1897). Tauern (1910, Fig. 30-3) studied the double refraction in glass due to electric fields with great precision, as it was by then possible to detect changes in the phase angle of light to $1/2000^{th}$ of a wavelength. He considered it possible that the Kerr effect might be employed in voltage measurements, and H. Becquerel had in fact suggested in 1884 utilizing the Faraday effect in measuring currents. Similar ideas have also cropped up later, but they have only been put into practice in some very specific cases. See more on the Kerr effect in chapter 38.

The Majorana (1902) effect appears as double refraction in colloidal suspensions of magnetic materials when in a magnetic field. It was investigated during the following years by Cotton and Mouton (1905, 1907a), G. Meslin, A. Schmauss, and J. Chaudier. The Cotton-Mouton effect (Cotton and Mouton 1907b, Fig. 30-4) is another magnetic phenomenon, analogous to the Kerr electro-optic effect. It causes liquids to be slightly birefringent in magnetic fields, especially if the shape of their molecules is non-spherical. Cotton and Mouton (1913) soon pointed out that the effect could be of use in studies of the nature of organic molecules. The same kind of effect in gases is named after W. Voigt.

The Stark effect is the splitting of spectral lines into several components when the emitter is in an electric field. Many futile attempts had been made at finding such an effect, when J. Stark

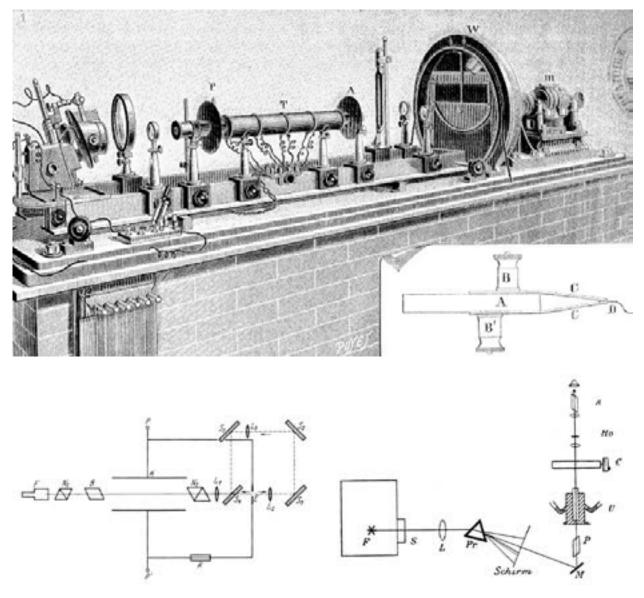


Figure 30-3. The Kerr electro-optic effect. Top: Measurements by Crehore and Squier (1895) of the speed of cannon projectiles. To begin with, no light from a lamp at the far left passes through the analyzer prism A which is perpendicular to the polarizer P. When the projectile starts moving, it switches on a high voltage across a capacitor in the tube T, causing a liquid between the capacitor plates to become doubly refracting. A narrow beam of light then traverses A and falls on a photographic plate at W. A motor m rotates this plate at a known constant speed. The projectile switches off the voltage after travelling a known distance. The illustration is from La Nature. Bottom left: The experiment of Abraham and Lemoine (1900) which showed that the Kerr effect (in a liquid between the capacitor plates K) is established in about 10-8 sec. This made it possible to utilize the effect for measuring e.g. the speed of light and very short lifetimes of fluorescence. Diagram from Born (1933). Right: Tauern's (1910) research on the Kerr effect in glass and CS2 at various light wavelengths. The samples are in the vessel U.

(1913, and A. Lo Surdo soon after) demonstrated it in the case of hydrogen and helium. Stark's equipment could produce fields of the order of 100,000 V/cm; see Appendix 5. The light has different polarization depending on the direction of observation relative to that of the field. P.S. Epstein (1916) provided an explanation of both the frequency shifts and the polarization in terms of the quantum hypothesis, concluding that the effect provides a "convincing proof of the correctness of Bohr's model of the atom". Bruhat (1942) agrees that the derivation by Epstein (and independently by K. Schwarzschild in the same year) had been "one of the first successes of the quantum theories". The Stark effect has generally been given less attention by physicists than

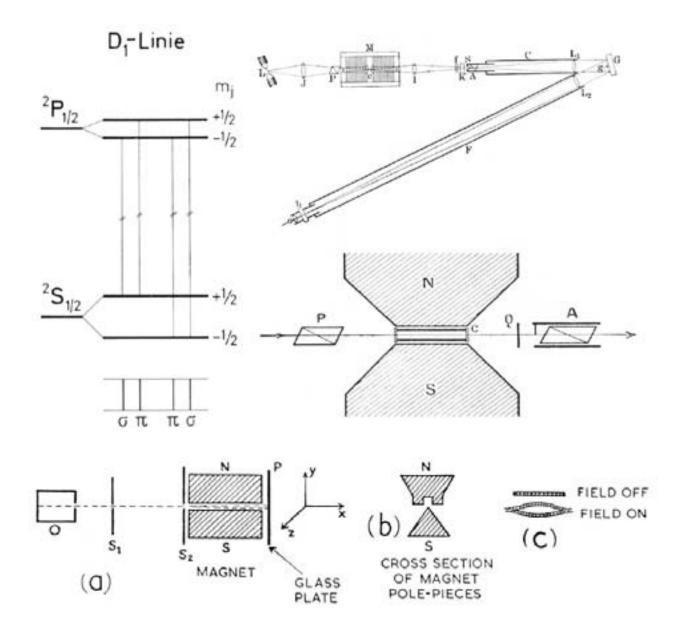


Figure 30-4. On the Zeeman effect and other interactions between magnetic fields and matter. Left: This schematic diagram shows how two of the permitted energy levels of a free sodium atom are split when it is in a magnetic field. At the bottom of the diagram it is indicated that two spectral lines (emitted in transitions between the split levels) are seen when one looks at light-emitting sodium along field lines (π) and another two are seen at right angles to the field lines (σ). From Haken-Wolf: Atom- und Quantenphysik, 1990. Right: The diagram at the top illustrates one of the many experiments which tested various theories about the Zeeman effect (Geiger 1907). G is a diffraction grating for detailed spectral analysis of the polarized light which has passed through a metal vapor in a magnetic field at c. Below is the setup of Cotton and Mouton (1905, 1907a,b) for measuring the double refraction of certain liquids in a magnetic field. It includes a compensator Q and a half-shadow prism A. Bottom: The experiment of Gerlach and Stern (1922), prompted by considerations of the Zeeman effect. Silver atoms, emitted from an oven (left), pass through an elongated evacuated volume where an inhomogeneous magnetic field pulls them to one side or the other. The fact that they form distinct stripes on the glass plate P, is evidence for the quantization of their magnetic moments. From Yarwood (1963).

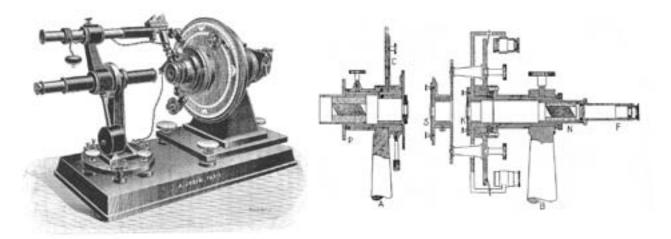


Figure 30-5. Left: The specially designed equipment of Chaumont (1915), containing Nicol prisms and thin plates of quartz and mica, for investigations of the Kerr electro-optic effect. Right: A part of Bergholm's (1916) instrumentation for studying the temperature dependence of the Kerr effect in various liquids. Light arrives from the left through a sample at S, between the Nicol prisms P and N.

the Zeeman effect, and G. Herzberg's 1937 book on atomic spectra even claims that it is "of no particular value as a help in the analysis of a spectrum". However, it has played a notable role in modern research on the topics like the genesis of molecules, widening of spectral lines, and electrical susceptibility of materials.

Many aspects of the Zeeman effect were studied up to the 1920s, by A. Righi, C. Runge, R.W. Wood, and J. Becquerel, to name a few. Examples include the research of L. Geiger (1907, Fig. 30-4) and R. Ladenburg (1912; see D.S.B.) on the Faraday effect in incandescent vapors of elements. These experiments continued the work of Macaluso and Corbino (1898) on the Zeeman effect, mentioned in the preceding section. Their purpose was to discriminate between different theories on the motions of electrons in atoms, although the interpretation of the results was somewhat uncertain. A new stage in understanding of the Zeeman effect was reached when Paschen and Back (1912) demonstrated that the anomalous effect resembled the normal effect more and more as the intensity of the magnetic field at the light emitter was increased. A further stage was reached with papers by A. Landé in 1921-23, where he among other things connects the polarization observed in the Zeeman effect, to those transitions between energy levels that are allowed by the rules of the quantum theory (Fig. 30-4). To quote Born (1933, p. 459): "Landé was the first to base a quantum-theoretical description of the Zeeman splitting on experimental facts. This provided one of the most important building blocks for the framework of quantum mechanics and for its application to atomic structure.". See also the review paper by Zeeman and Bruin (1927).

In 1886, E. Goldstein discovered so-called "canal rays", i.e. streams of positively charged atoms that could be generated in evacuated glass vessels with perforated high-tension electrodes. These particles emitted light when colliding, and the Stark effect mentioned above was in fact originally detected in light of that kind. The canal-ray radiation was subject to considerable research in the early 20th century, including its polarization under various conditions (e.g. Stark and Lunelund 1914, Hertel 1927), and the spectral distribution of the radiation was measured with a König-Martens polarization spectrophotometer (Stark and Steubing 1908). Some of this research was considered to be "an important contribution to the problem of the nature of light" (Tomaschek 1934, p. 223-224) but it will not be dealt with here; the part played by canal rays in the development of atomic physics does not seem to have been given much prominence in historical accounts. The Faraday and Kerr effects were also studied further in 1900-25, both experimentally and theoretically. For instance, Lowry (1913b and later) improved upon the methods used by Perkins (section 22.3) in his measurements on organic compounds, especially by using more than one wavelength in the visible spectrum, and Ingersoll (1917) extended them to the infrared. J. Becquerel (1909, and many other papers) made noteworthy discoveries regarding the Faraday effect and associated absorption of light at very low temperatures. Chaumont (1915) and Bergholm (1916) investigated the Kerr electro-optic effect in detail with new instrumentation (Fig. 30-5) and Barker (1916) studied the magnetic saturation of many ferromagnetic materials by means of the Kerr magneto-optic effect. All these effects have been useful in research on the structure of atoms, molecules, liquids and/or solids; in particular Tomaschek (1934, p. 362) claims that the Kerr electro-optic effect is a "very important tool" for such research.

One more major step towards full understanding of the laws of quantum theory was taken by the experiments of Gerlach and Stern (1922, etc.) which were prompted by the Zeeman effect (cf. e.g. Yarwood 1963). These authors showed that the magnetic moments of atoms in silver vapor had discrete values, see the bottom part of Fig. 30-4 and Appendix 5. The moments could be presumed to belong to silver's single valency electron. We shall return to these matters in section 39.3.

31 Motion relative to the aether, ca. 1860-1915

The earth's speed in its orbit around the sun is about 30 km/sec, whereas the speed of a point on the equator due to the earth's rotation is less than 0.5 km/sec. The former is 1/10,000 of the speed of light, and causes the direction to the pole star to vary a little with the season. This effect, called aberration, was known since around 1730.

During the time when scientists still believed in a material aether that pervaded the universe, they were keen to find out whether the speed of any point on the earth with respect to the aether could be measured in the laboratory. A. Fresnel had predicted that a moving object "dragged" the aether along with it, and H. Fizeau confirmed this prediction in 1851 by measuring the speed of light in flowing water. Fizeau (1860) then carried out extensive measurements to test whether the earth's orbital motion affected the direction of the plane of polarization of light travelling through a pile of tilted glass plates. Fizeau's results, while not conclusive, seemed to indicate some motion relative to the aether. In the year 1870, the French Academy of Sciences announced a prize for new optical discoveries concerning this subject. The prize was awarded to E. Mascart for a series of detailed experiments (Mascart 1872, 1874, see C.R. 79, p. 1531-1534, 1874). One of the experiments tested whether the earth's motion affected the double refraction in Iceland spar, and another one (Fig. 31-1) tested its effects on the optical activity of quartz. Mascart however concluded from his observations that it was impossible to find the speed of the earth through the aether. At a similar time, E. Ketteler (1872) published theoretical papers on the effect of the earth's speed on double refraction and other optical phenomena. These were expanded in a book (Ketteler 1873) which included a description of the work of Fizeau and of Mascart on these matters. Ketteler himself also made two experiments to measure the aether speed. In one of these he employed a train of four specially constructed 90° totally-reflecting Iceland spar prisms, yielding a result for the speed of less than $1/18^{h}$ of the expected value. In Ketteler's other experiment where six such spar prisms were used, it was anticipated that certain images observed in a small telescope would move laterally when the setup was rotated through 180° at noon, but not the slightest translation was perceived.

In the following decades various further attempts were made at estimating motion with respect to the aether, using mechanical methods, electric or magnetic fields, or radiation. All gave negative or unsatisfactory results and they have been forgotten, with a single exception published in 1887. This very comprehensive experiment which was carried out by A.A. Michelson and E.W. Morley, was based on an original suggestion by J. C. Maxwell (see entry on him in D.S.B.) and employed Michelson's very sensitive interferometer. The back-and-forth speed of light in vacuum in two perpendicular directions was compared, and found to be invariant. This result caused much discussion for many years (see e.g. Preston 1895, and the entry on G.F. FitzGerald in D.S.B.), and additional measurements of that type were carried out by for instance Morley and D.C. Miller in 1904.

At the turn of the century, considerations by FitzGerald, H.A. Lorentz and others (see van Nostrand 1989) of the laws of mechanics in the light of Maxwell's electromagnetic theory, made it clear that moving objects were bound to contract in the direction of motion. In this way, Lorentz could among other things explain why Mascart's experiments had not indicated any earth-aether motion. Subsequently, experiments were set up by Rayleigh (1902b) and by Brace (1904b) to find out whether such a contraction caused double refraction in transparent materials, analogous to that caused by pressure (section 27.3). It is estimated that the latter's apparatus (Fig. 31-1) was 300 times more sensitive than that of Michelson and Morley, but no effects from the earth's orbital motion were detected.

Rayleigh (1902a) and Brace (1905b) also carried out other experiments on the aether motion, employing polarized light. These tested whether the motion had an effect on optical activity in quartz or in oil. Rayleigh's experiment, which resembled one of Mascart's (1872, 1874), made use of thick quartz blocks which rotated the plane of polarization of yellow light by more than 5000°. Brace (1905c) then repeated the experiment of Fizeau mentioned early in this section, with much improved techniques. The results continued to be negative: the properties of a light ray in space were quite independent of its direction of propagation. These experiments were discussed by many physicists, e.g. Larmor (1904).

At this point, Albert Einstein entered the scene. It may be pointed out here that his earliest surviving scientific essay, written in 1895 while he was a college student, deals with light transmission through magnetized matter: here he mentions double refraction and polarized waves. The essay is reproduced in vol. 1 of Princeton University's edition of Einstein's collected works, 1987. In an 1899 letter, Einstein mentions that he has read the paper by W. Wien (1898) which lists some 13 aether-speed experiments including the polarized-light ones of Fizeau and Mascart.

In Einstein's (1905b) famous first paper on his theory of relativity, he does not quote any previous papers, but he considers that "...the lack of success in attempts to establish a motion of the earth relative to the "light medium", leads to the assumption that the concept of absolute rest...does not fit the observations". It is quite possible that Einstein was here referring for instance to the very recent experiments by Rayleigh and by Brace on polarized light, along with the older observations of Michelson and Morley which used unpolarized light. However, in those later articles by Einstein and interviews with him which discuss the origins of his relativity theory, he only mentions the Michelson-Morley experiment. As it was later put by P.A.M. Dirac, Einstein's theory "destroyed the aether" but it took a long while to gain acceptance, and various aether-drift experiments were conducted up to 1930 at least. Among those where polarized light played a part, was a further improvement by Strasser (1907) of the Fizeau-Brace observations. Kennedy (1922) set up an optical experiment with a Wollaston prism and four Nicol prisms for the same purpose, and he later carried out one with simplified apparatus (Kennedy 1928), however still requiring polarized light.

Sagnac (1913) made noteworthy measurements of the angular velocity of a rotating object. With an interferometer, he observed how much faster a light beam going against the rotation travelled around a circular path inside the object than a beam going with the rotation. This experiment (Fig. 31-1) where a Nicol prism plays a minor role, was for a long time involved in discussions of the aether and of the relativity theory, but in fact its outcome had no bearing on that theory. The "Sagnac effect" has had important uses in positioning techniques in recent years.

One more instance where the paths of Albert Einstein and Iceland spar happened to cross, was at Princeton University where R.A. Beth (1936) designed a very sensitive experiment to measure the angular momentum of circularly polarized light. In his apparatus, a light ray polarized by a large Nicol prism impinged on thin quartz plates suspended in a very fine fiber. In a footnote, Beth thanks Einstein for their several interesting discussions on the equipment, and for assistance in correcting an error in an older theoretical paper on the matter. According to later accounts, Beth's

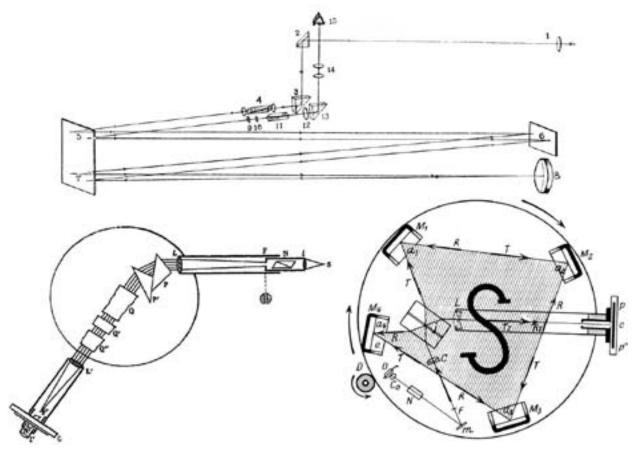


Figure 31-1. Three experiments concerning movement of the Earth relative to the luminiferous aether. Top: Brace (1904b) sent a light beam back and forth in a 4-m long water-filled trough. He attempted to find evidence of double refraction in the water, caused by the contraction (in the direction of the Earth's velocity) suggested by H.A. Lorentz and G.F. FitzGerald. Objects no. 4 and 11 are Nicol prisms. Bottom left: One of Mascart's (1872) aether-drift experiments which were rewarded by an Academy of Sciences prize. Here he is looking for the influence of the Earth's motion on the rotation of the plane of polarization of light (emitted by s) in the quartz blocks QQ'Q''. The view is from above; there are Nicol prisms at both ends of the light path. Right: The vessel S which is viewed from above and is about 0.5 m across, rotates. Sagnac (1913) investigated whether a light ray travelling with the rotation had a higher velocity than a ray moving the other way around. The Nicol prism N is used to equalize the intensities of the rays before they enter the interferometer at M4. From Wien-Harms (1928).

results had some fundamental significance.

Finally, two points regarding the relativity theory must be stressed here. One is that the theory owes its appearance to Maxwell's electromagnetism and its general recognition by 1900. At that time it had also been realized that Maxwell's field equations were incompatible with Newton's mechanics, and the relativity theory was one of the suggested ways to resolve that problem. As has been shown in the present compilation, polarized light was a factor all through this story, beginning with the Faraday effect. The other point is that one consequence of the relativity theory is the equation $E = mc^2$, which Einstein derived in a separate paper in late 1905. At that time, scientists were giving much thought to the source of the energy released in the radioactivity of some elements, and also to the source of the energy radiated by the sun and stars. This equation might possibly have been derived without the relativity theory, for instance from measurements of atomic masses of radioactive mother and daughter elements along with the energy of emitted particles. However, this would hardly have happened until long after 1905. Einstein's mass-energy equation clearly guided scientists in their research on atomic transmutations, including nuclear fission from around 1940 onwards, and in the applications of fission for military and civilian purposes.

32 The Helgustadir quarry in 1895-1910, and the supply of Iceland spar

After the quarry at Helgustadir had been dormant for a decade, the government leased its operation to Carl D. and his son Thor E. Tulinius, initially for five years from mid-1895. They also handled the sale of all the product, through the latter's commercial company in Copenhagen. The contract was extended twice for five years, i.e. to 1910. A biography of Thor E. by Gils Gudmundsson may be found in the compilation "Their settu svip a öldina" (They made their mark on the 20th century, vol. I, p. 304-317), published in Reykjavik in 1987. Carl D. Tulinius died in 1905. The company was required to deliver balance sheets on the finances of the operation and on its stock situation at the end of each calendar year; several of these are preserved in the files of the National Archive (Fig. 32-1). The spar crystals were sorted into three categories, A+B, C, and D. I have not seen the detailed criteria that were used, but B probably covered small perfect crystals and/or large slightly defective ones, and C was "secunda". Annual sales from the A+B class amounted to 100-250 pounds (1 pound = 0.5 kg), while less was sold from the other two. Prices clearly kept rising, at least to 1908. The revenue delivered to the state, after deduction of all costs, seems to have been around 2000 kr. per year on average. This sum is also quoted by Ussing (1902) who mentions that ten workers are usually hired in the summer months. For comparison, the very first engine in an Icelandic rowing boat (in 1902) cost 900 kr. including its installment.

A number of users of Iceland spar continued complaining about its scarcity, as may be seen in Appendix 1A. However, some clearly had access to a supply, for selling or for making prisms to incorporate in optical equipment. It is probable that they had managed to build up their own stocks during the time when plenty of spar was available. For instance I have acquired an undated catalog from the optical workshop of Bernhard Halle (ca. 1895-99, Fig. 32-2) of Steglitz near Berlin. This firm offers some 30 sizes and modifications of Nicol prisms, other polarizing prisms, polished rhombohedra, spheres, lenses and plates from Iceland spar, altogether about 200 different products. The catalog of Pellin (1899) similarly lists Nicol prisms, rhombs, equilateral dispersing prisms and lenses of various shapes and sizes from Iceland spar for research and teaching. At the end of P. Groth's (1905) book on physical crystallography, Steeg & Reuter advertise a variety of Nicol prisms and Iceland spar pieces. A.B. Porter (1907) sells many types of Nicol prisms up to 4.5 cm long, lenses, dispersing prisms, beam-splitting prisms, rhombs, plates and spheres of spar. Equipment suppliers like F. Ernecke and C. Gerhardt also offered this sort of materials around 1900. Advertisements and descriptive papers on polarizing microscopes and related apparatus kept appearing from producers like R. Fuess (Leiss 1898, 1908, etc.), Voigt & Hochgesang, and Zeiss, see the relevant sections in chapter 29. In the books of Landolt (1898) and Rolfe (1905), various available polarimeters for research on organic liquids are presented.

Most microscopes, polarimeters and photometers contained two Nicol prisms, some a third one or other accessories from Iceland spar. For an average Nicol prism which I estimate to have been

Havnegade 43.

Kjøbenhavn K., den 1. Januar 08.

Thor E. Tulinius.

Telegramadresse: _Thore".

		Telefoner
Ne.	5078	Chelm.
	555	Hovedkonitor.
	5042	Dampshibsexpedition.
2	29115 y	Pakhas Asiatisk Kompagni.
	29130 4	 Ornitiangate.

Herved har jeg den Ere at fremsende Forhandlingsregning over Doppelspath, solgt 1 Aaret 1907, hvoraf Regeringens Andel er

Kr. 2751,81,

som staar til det høje Ministeriums Disposition.

Beholdningen ved Nytaar stiller sig saaledes:

I Behold 1 Jan.1908		2589,97 Pd.	390,28 Pd.	3438,14 Pd.
1907)	113,35 *	6,40 *	
Solgt 1 Løbet af)			
1907	;	2703,32 Pd.	396,68 Pd.	3498,12 Pd.
Beholdning 1 Jan.	?	A&B	с.	D.

Da det, forinden yderligere Udvinding kan ske, er nødvendigt at foretage betydelige Sprengninger og Udgravninger, har man ikke i 1907 haft Anledning til at faa noget op af Minen.

Til 1023/308 glands Ministeriums Kontor i København. Stjórnarráð Íslands II Danis / 1 19 463

Figure 32-1. A balance sheet from Thor E. Tulinius for the sales of Iceland spar in 1907 (National Archive of Iceland). It was accompanied by a detailed statement on income and costs. According to the 1895 contract between Tulinius and the Government, he was responsible for the operation of the Helgustaðir quarry and all sales of crystals. The net profit was to be split evenly between the parties. This amounted only to a very small fraction of the turnover of the Thore Company in trade and shipping at that time. In the letter, Tulinius stresses the need for new blasting efforts.

2 cm in dimensions (a size of 1.8 cm is mentioned by M. Katalinic in Ann. Phys. 83, 1927), two crystals of spar of at least 2.5 cm size may have been required, which corresponds to of the order of 0.2 kg per instrument. The amount of spar of all quality classes sold by T.E. Tulinius for the state according to his available balance sheets from 1897-1908 might therefore have been sufficient for the production of an unrealistically small number of 500 instruments per year on average, even if none of this material was wasted or used for other purposes.

The balance statements by Tulinius at least from 1897 onwards repeatedly make the point that the quality of the recovered crystals is not very satisfactory, and is becoming worse. See also quotes from Badermann (1910) and Encyclopaedia Britannica (1910-11) in Appendix 1A. In 1901, not much was done except blasting cliffs for improved access to crystal outcrops. Three miners from Norway were brought in to carry out explosive work in 1905, but already at the end of 1906 Tulinius stressed that more blasting was needed (Fig. 32-1). No significant quantities were recovered in the years 1905-08. According to the book by Dammer and Tietze (1913), "the production" of spar was similar in 1908-10 as in the first years of the century, but this may be referring to the amounts sold by Tulinius.

A letter dated 23 Oct 1906 from the firm of Carl Zeiss to H. Hafstein, Minister for Iceland, is in the National Archive, see Kristjansson (2003). Here Dr. R. Straubel on behalf of the firm offers to purchase Iceland spar directly from the owner of the quarry, as the available material is not up to standard for their uses. The firm of B. Halle which was mentioned above, corresponded with authorities in Iceland in 1907-08 and was interested in leasing the quarry. The Halle people even wanted the government to terminate the contract with T.E. Tulinius as soon as possible, as he hald told them already in 1905 that not much spar was to be found there. They had heard that samples from another locality in Iceland had reached the mainland (see also Stadthagen 1909 in Appendix 1A): it may have been at the farm Akrar in West Iceland where the local farmers had discovered around 1900 that Iceland spar was to be found. This is mentioned in the Reykjavik newspaper Fjallkonan in Oct. 1901, with a favorable comment on samples from the geologist H. Pjeturss. Two Icelanders (G. Jakobsson and M. Blöndal, according to document 218 of a collection to be mentioned in section 37.1) had acquired mining rights at Akrar by 1910. In December of that year, Fjallkonan reports that Frenchmen have recently bought the Akrar mine, but I have no further information on this locality or whether any spar was ever exported from there. Badermann (1910) writes about two spar mines, recently investigated by emissaries from German instrument makers, where some recoverable material was thought to remain. These emissaries may have been G. Angenheister and A. Ansel who made enquiries regarding the subject while travelling in Iceland; their main task however was to carry out observations connected with the 1910 appearance of Halley's comet.

In 1897 or even earlier, a locality with a variety of calcite crystals at the so-called Baidar Gate in the Crimean peninsula was becoming well known, and by the turn of the century Nicol prisms had already been made from some of these (Sokolow 1898, Zemiatcenskij 1902). A minor quantity of Iceland spar crystals "not much inferior to those from Iceland" had been found at Auerbach in Germany before 1900 (see abstract from a report by A. Leuze in Zeitschr. Kristallogr. 30, 1899). One partial explanation of the obvious gap between the supply of spar crystals from Iceland and the demand, might be that some manufacturers had access to source sites that others did not know about, say in the two areas mentioned or elsewhere, for instance in the Pyrenees (cf. section 4.1) or in Switzerland (cited in some 18th century books on mineralogy). However, this is merely a guess.



Figure 32-2. Frontispiece and one page from the catalog of B. Halle (ca. 1895). His workshop and supply firm, which is still in business, offered many types of Nicol prisms and other components for optical research instruments. Educational equipment for practical instruction and demonstrations regarding the properties of polarized light was also available.

33 Components to replace Iceland spar in Nicol prisms and elsewhere, 1885-1920

When Iceland spar became scarce after 1880, scientists naturally endeavoured to economize on its use in optical instruments, and to develop substitutes. This applied mostly to polarizers, while analyzers continued to be made from Iceland spar. Some developments in the field after 1920 will be covered in chapter 35.

33.1 Glass plates, etc.

One way of producing polarized light is by reflecting ordinary light from the surface of transparent or dark glass (cf. Malus' discovery in section 6.5, Fig. 33-1) or by transmitting it through an inclined pile of glass plates. Use of the reflection method in optical equipment has various drawbacks: only about 15% of that half of the light which is desired, actually is reflected from a single surface (Fig. 7-2; 25% from both surfaces of a glass plate); the field of view (i.e. the range of usable angles of incidence) is narrow (Bennett and Bennett 1978, p. 83), the light beam changes its direction which is inconvenient; and the light is not completely polarized, due to surface imperfections (Zehnder 1908b). This method, sometimes named after J. Nörrenberg (Fig. 10-3) found some applications as a polarizer in simple inexpensive microscopes (Leiss 1897a, see Fig. 33-2), in heated-stage microscopes for observing crystallization processes (Lehmann 1890, Rosenbusch 1924) and in magnifying viewers for quick inspection of large thin sections of coarse-grained rocks or sediments (see Schneiderhöhn 1922, p. 5), ice (Klocke 1879, McConnel and Kidd 1888) and thin sections of individual crystals (Wheatstone 1871, Bertin 1879, Gumlich 1896) as in Fig. 33-2. Furthermore, the glass-plate polarizers were employed in apparatus for demonstrations and practical exercises in schools (e.g. Thompson 1905, p. 234-235, Fig. 33-1 bottom left), photoelasticity work (Coker and Thompson 1912, Coker and Filon 1931, Fig. 27-5 and Fig. 33-1), studies of the Maxwell effect in fluids (Zocher 1925), inspection of mica sheets for electrical capacitors, some solar observations (Chevalier 1906), monitoring of strain in optical glass (Halle 1921) and in glass factories in general (Spencer and Jones 1931). The method was also used much in the case of ultraviolet and infra-red (> 2400 nm) light which is absorbed by Iceland spar (Lowry 1935, Françon 1963, Bennett and Bennett 1978).

By reflecting a light beam twice from parallel glass plates, it was shifted laterally instead of changing direction (Hall 1892, Grimsehl 1905). In this variant, use was also sometimes made of total reflection in glass prisms invented by C. Delezenne in 1834 (Barnard 1863, Mascart 1889, p. 517, Pellin 1899, Thompson 1905, see Fig. 33-1). Triple glass mirrors were also introduced, e.g.

for projectors coupled to microscopes (Metzner 1920). Polarizing prisms from glass occasionally replaced the glass plates (Schulz 1911, see also Johannsen 1914, p. 175, and Metzner 1920). However, they do not seem to have been widely used, possibly because of a narrow field of view or because of poor image quality due to strains in the glass (Glazebrook 1923, p. 498).

Around 1920 several versions appeared of the method where polarized light was produced by transmission through tilted glass plates (see Brodsky 1919, Metzner 1920, Wien-Harms 1928, p. 370-373, Fig. 33-1). However, it also has limitations such as the light being less completely polarized than predicted by theoretical formulas (Bennett and Bennett 1978, p. 88). Nelson (1919) finds this light to be unsuitable for microscopy, and Bruhat (1930, p. 6) says that glass-plate polarizers generally are inadequate for use in any measuring apparatus.

A few attempts were made at employing thin plates of tourmaline e.g. in petrographic microscopes (Nelson 1902) and in projectors for teaching about polarized light (Lummer 1909, p. 960), without much success as different samples of it have different colors (Nelson 1919, Wien-Harms 1928, p. 398; section 12.3 above). This material was therefore mostly used in simple devices for the rapid identification of minerals by means of interference patterns or dichroic properties (Halle 1921, p. 125, Rosenbusch 1924, p. 221; catalogs and many other references; Fig. 12-1), for observing the polarization of sky light (Busch and Jensen 1911, p. 293), and in teaching (Hauswaldt 1904). Lommel (1881, and earlier) described plates from a magnesium-platinum-cyanide compound that had a similar effect as tourmaline, but only on blue light.

Small or slightly defective Iceland spar crystals could be used in the dichroscopes for evaluation of gems etc. that were described in section 13.4, see Fig. 33-3. This figure also illustrates a handy device with small Nicol prisms for checking whether a thin section had been ground to proper thickness.

33.2 Calcite-saving prisms, and calcite lenses

The great diversity of Nicol-type prisms indicated in Fig. 12-1 (center) partly resulted from efforts to optimize their performance in the various optical instruments. Many different materials were also used for cementing the two prism halves, instead of the original Canada balsam.

Soon after the invention of the Nicol prism, H.F. Talbot realized that its exit half played no part in the polarizing function of the prism; it only insured that the extraordinary ray did not change its direction of propagation in the prism as a whole. Therefore, much valuable raw material could be saved by replacing this half with glass. However, experiments to test this possibility were not performed until the early 1870s (Talbot 1872, Leiss 1897c, Lommel 1898), see top of Fig. 33-1. Although it was easy to find a glass with a suitable index of refraction for say yellow light, glasses having also the same dispersion as calcite could not be procured for a long time yet (Halle 1921, p. 113, Hardy 1935). Therefore, two perpendicular spar-glass prisms would not completely extinguish a white light beam. When at last glass with the right dispersion became available (Ammann and Massey 1968) image distortion appeared, as well as problems due to the different thermal expansion of glass and calcite (Bennett and Bennett 1978, p. 39-40).

It was attempted to invert the composite glass-calcite prisms, by letting the light ray travel first through a glass wedge of high refractive index, so that the extraordinary ray would be reflected away at the interface. However, this required using a glue with a refractive index of at least 1.655, rarely encountered in such materials. Yet another spar-saving approach was to place a thin plate of Iceland spar between two wedges of glass or certain liquids like CS_2 (Jamin 1869, Feussner 1884, Bertrand 1884a, Madan 1890, Sang 1891, Coker and Filon 1931, p. 60, Fig. 33-1) but this also encountered thermal expansion problems (see Ammann and Massey 1968, Bennett and Bennett

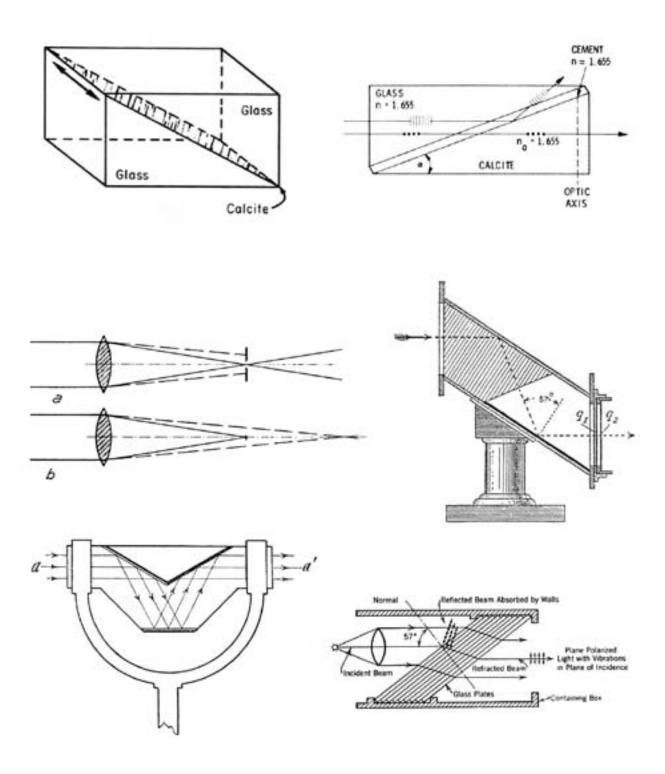


Figure 33-1. Various means of creating linearly polarized light, to economize on Iceland spar or replace it. None of these attempts gained wide application. Top left: A so-called Feussner prism, improved by Bertrand (1884). It is a sandwich of glass with a thin diagonal plate of calcite. Instead of the glass, a liquid with a high refractive index could be used, and sodium nitrate instead of calcite. From Bennett and Bennett (1978). Right: A composite glass-calcite prism as suggested e.g. by Leiss (1897c). From Ammann and Massey (1968). Center left: An calcite lens with two focal points; the optical axis is in the plane of the lens. The unbroken lines on the right indicate the ordinary ray; it is intercepted in the lower diagram while in the upper one the extraordinary ray is screened off (Schulz 1925). Right: Polarization by means of reflection, either from glass plates or a Delezenne glass prism, see Barnard (1863). q1 and q2 are mica quarter-wave plates, which make it possible to rotate the plane of polarization of the emergent light. From Thompson (1905), see also Schulz (1911). Bottom left: Polarization by two reflections from the sides of a triangular glass prism (and a polished metal surface below). From Rosenberg (1924). Right: Light being polarized by passage through a pile of tilted glass plates (Frocht 1941).

1978). These prisms also transmitted a narrower range of wavelengths than than ordinary Nicol prisms. One version designed by Brace (1903) was said to be a very good polarizer but fragile and difficult to handle. In Handbuch der Physik vol. 20, 1928 it is stated that polarizers of the calcite-plate kind have "hardly been used at all".

Around 1920 there are signs of people beginning to use beam-splitting Iceland spar prisms (section 12.1) instead of Nicol prisms for polarizers in experiments (Wood and Mohler 1918) and instruments. Grosse (1887) had in fact pointed out long before, that the right-angle prism first described by Dove (1864, Fig. 33-2) was inexpensive and also quite practical in some situations. In a short note in Nature 103, p. 239, 1919 it is mentioned that F.J. Cheshire had utilized a beam-splitting prism in a projector he constructed, needing for it only one-eighth of the spar material going into a Nicol prism of comparable performance. For a similar purpose Weigert and Käppler (1924) used a Senarmont prism, and Hardy (1935) chose a Rochon prism for his popular spectrophotometer. Schönrock (1928, p. 729-731) remarks that manufacturers some years before (i.e. about 1920?) redesigned some of their polarimeter models in order to employ Senarmont-and Dove prisms, because very decent polarizers could be obtained in that way from spar pieces previously discarded.

A clever approach to Iceland spar savings in optical instrumentation involved making lenses from it instead of prisms. This method was described e.g. by Fedorow (1901) but B. Martin (1774) originally drew attention to that possibility. If a point light source is placed sufficiently far from a converging spar lens (Fig. 33-1), two images of it appear on the other side of the lens. The ray forming the image closer to the lens may be disposed of by letting it fall on a small opaque screen, which will only slightly diminish the intensity of the other image. Such lenses were used for instance by Sagnac (1904) in testing some aspects of the propagation of light along the axis of optical instruments, and by Lowry (1908) in research on the optical activity of quartz in the ultraviolet. Several papers on the properties of calcite lenses appeared in the 1920's in connection with a patent dispute (see Schulz 1925) but they do not seem to have found any lasting applications.

33.3 Saltpeter crystals

In order to release optical research from the burden of inadequate Iceland spar supplies and some of the material's limitations, it would have been sufficient to find a comparable natural or man-made mineral. The most obvious choice and the only one seriously considered, was sodium nitrate (Chile saltpeter). It is transparent, inexpensive, trigonal and optically negative like calcite, and has an even stronger double refraction ($n_0 = 1.587$, $n_2 = 1.336$). It is also isomorphous with the calcite group of carbonates, having angles of about 106° between its faces instead of calcite's 105°. See Feussner (1884) and Bertrand (1884a) on prisms and plates from saltpeter which they hoped would find their way into general use in optical instruments. A report on years of work in growing saltpeter crystals is presented by Wulff (1895, 1897). The task turned out to be challenging, but people continued believing in this alternative for a number of years; thus, Bauer (1904, p. 269) says about Nicol prisms that "sometimes they are made from other substances (sodium saltpeter, etc.)". In an equally optimistic vein, Badermann (1910) indicates that "large pieces of saltpeter of quite good quality" are arriving on the market from Paris. Unfortunately, it is very hygroscopic, so that all polished surfaces must be coated immediately with other materials. It is also much more difficult to work with than calcite (Halle 1921), due at least in part to its softness. According to Schulz and Gleichen (1919, p. 42): "The attempts made to replace...calcite by other crystals of similar optical properties, have so far not resulted in any practical solution.".

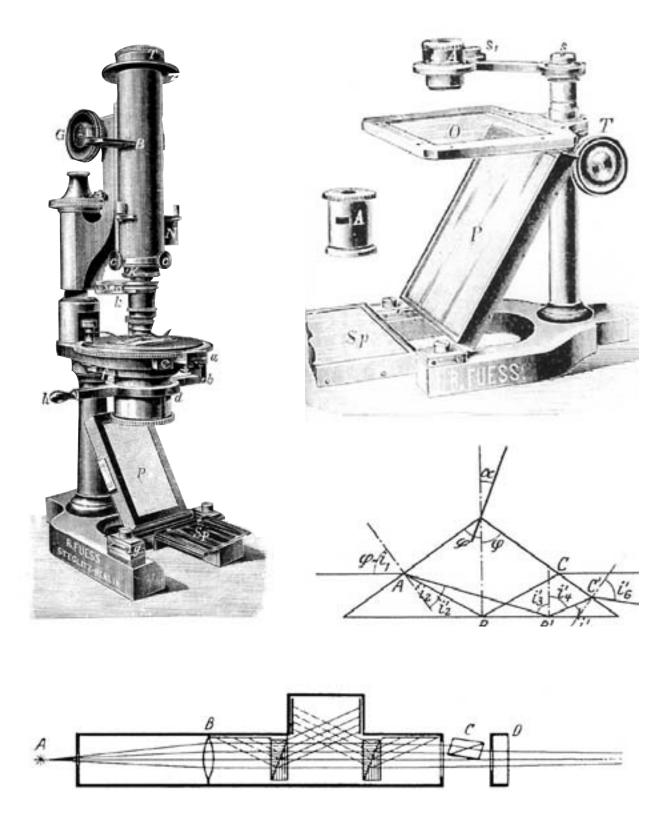


Figure 33-2. More ways of saving Iceland spar. Left: A microscope with a glass-plate polarizer P and a small analyzing Nicol prism at N, probably intended for student use (Leiss 1899). Top right: A simple device for rapid inspection of e.g. mineral fragments or large thin sections, with a glass mirror P for polarizing. An ocular magnifier with an analyzing prism is at A (Leiss 1897a). Center right: Dove's (1864) Iceland spar prism was occasionally used as a polarizer in optical equipment, for instance around 1920 when Iceland spar was hard to come by (Schönrock 1928). A ray of ordinary light entering horizontally from the right, exits as two linearly polarized rays in different directions. Bottom: A polarimeter where two Senarmont prisms, made from leftover Iceland spar pieces, substitute for a Nicol prism. Dark matt glass plates absorb unwanted rays. A half-shadow field of view is provided by the small Nicol prism C (Schönrock 1928).

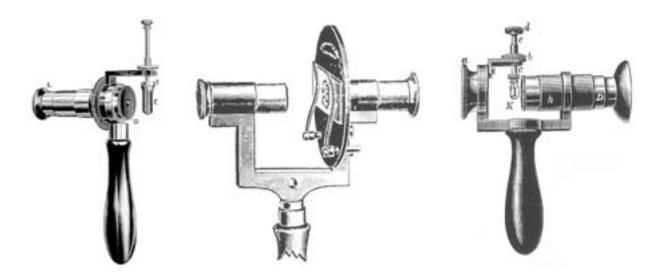


Figure 33-3. Dichroscopes for inspection of gemstones, on the left from Winkel/Zeiss (as shown in their leaflet no. 200, around 1920), on the right from R. Fuess (Leiss 1899b). In the center we have a viewer from J. Swift & Son for checking whether thin sections have reached an appropriate thickness when being ground (from J. Royal Microsc. Soc. 1907).

34.1 Physical properties of Iceland spar, etc.

Research on the properties of Iceland spar and of calcite in general continued after the turn of the century. No doubt its results were useful in some ways although not always opening up new avenues in science. In addition to optical studies on the spar (sections 28.3 and 34.2) there were investigations of its magnetic properties (Königsberger 1898, Voigt and Kinoshita 1907), cleavage directions (Friedel 1902), electrical properties (Fellinger 1902, W. Schmidt 1902, Pisani 1906, Röntgen 1907, Richardson 1915), deformation caused by pressure (Rinne 1903, Adams 1910), changes in crystal shape and refractive indices at low and high temperatures (V.M. Goldschmidt 1912, Wright 1913b, Rinne 1914), dissociation and melting at very high temperature (Brill 1905, Boeke 1912), etching and rate of dissolution (V. Goldschmidt and Wright 1903, 1904, Beilby 1909, Gaubert 1912, Honess 1918), thermal conductivity (Eucken 1911) and the transformations occurring in calcite during calcination (Rinne 1928). Bäckström (1921, 1925) carried out a thorough investigation of thermodynamic variables during the conversion of aragonite to calcite, using in part crystals presumably of Iceland spar.

In the above time interval, important books on optics and crystal properties appeared, often reviewing previous research on Iceland spar. These include Drude (1900a), Becker (1903), Wood (1905), Pockels (1906), Winkelmann (1906), Lummer (1909), Voigt (1910), and Schuster and Nicholson (1923). See Fig. 34-1. On the theoretical side, a general paper by Grünwald (1902) on the transmission of elastic and electromagnetic waves through uniaxial crystals was a notable step forward (Gårding 1989). Around 1915 a number of papers were published on light in liquids and crystals, for instance by P. Langevin, M. Born (1915), P.P. Ewald, and C.W. Oseen.

It is also worth pointing out at this stage, that doctoral theses in the 19th and 20th centuries often involved polarized light and double refraction in one way or the other. This applies to many fields of the physical sciences, but also to some extent to biological sciences and engineering. Results of these projects were in many cases presented in papers in scientific journals, or printed on their own (Fig. 34-1), even both. Thousands of young scientists thus received their basic training in research through involvement with Iceland spar, and many of these continued into illustrious careers.

34.2 Oscillating systems in matter

One major motive for detailed measurements of the frequency dispersion of both refractive indices and optical rotation on both sides of the visible band of the spectrum, was a desire to de-

scribe it with formulas that were simple, accurate, and representing physically realistic models. Before 1900, H.v. Helmholtz (1892), H.A. Lorentz and others explained the former dispersion with general formulas (which Sellmeier (1872) and Ketteler (1874) had in fact already proposed from the viewpoint of the old theories of light, see section 21.2). They were based on Maxwell's electromagnetism and the suggested presence of damped oscillators within the molecular structures, having various different resonance frequencies. Changes in the refractive index (including the phenomenon of anomalous dispersion) will be most pronounced in the vicinity of a resonance frequency. There, the absorption of the light will also reach a maximum (Fig. 34-2), as was confirmed in measurements by Pflüger (1898) who used König's polarizing spectrophotometer.

The observed dispersion in most materials indicated (e.g. Carvallo 1900, Martens 1901) that their resonance frequencies were quite far from the visible range on both the infra-red and the ultraviolet side. Drude (1900a) pointed out that a similar formula could be used in accounting for the optical rotatory dispersion in quartz, however using only the ultraviolet resonance frequency deduced from its refractive index dispersion. Drude's formula in fact fitted the rotatory dispersion in quartz, measured by Gumlich (1898) in the interval 214-2200 nm, much better than earlier empirical formulas by Boltzmann (section 18.1) and others. At that time, precise rotatory dispersion measurements had not been made for other materials over a wide wavelength interval, but the dispersion of the Faraday effect in two liquids measured by Joubin (1889) agreed with Drude's derivations. Drude (1900a, p. 383) also points out in support of his approach, that the absorption of circularly polarized light discovered by Cotton (1895a, see section 22.3) is analogous to the absorption mentioned in the previous paragraph.

It had become clear by 1900 (for instance from the Zeeman effect) that a hydrogen atom was about 1800 times heavier than an electron, and it is well known that the resonance frequency of an oscillator with a given force constant is inversely proportional to the square root of its mass. This made it likely (Drude 1904) that the ultraviolet resonance frequencies in molecules belonged to electrons while infrared ones corresponded to vibrations of single atoms, parts of molecules, or entire molecules. Nutting (1903b) measured the ultraviolet rotatory dispersion of solutions of four organic compounds, and estimated a resonance frequency for each.

H. Rubens and collaborators who investigated infra-red radiation since about 1892, found that plates of insulating materials which are transparent in visible light, absorb strongly in a small number of infra-red frequency intervals. These rays are therefore reflected (Fig. 34-2 top), and after 3-5 reflections of heat radiation from the same material it consists of a few narrow frequency bands. Rubens called them "residual rays" (Reststrahlen) from 1897; the first materials investigated in this way (see D.S.B.) were quartz which yielded rays of about 8800 nm wavelength, and fluorspar with about 24000 nm. Common salt, mica, diamond and other crystals soon followed, including calcite (Fig. 34-2) whose residual rays included one at an unusually short wavelength of 6800 nm. Some of these wavelengths agreed with the infra-red resonance frequencies which could be computed from dispersion measurements at shorter infra-red and visible wavelengths. For instance, Aschkinass (1900) showed that the frequencies were (at least in part) the same for two different modifications of calcium carbonate, namely calcite and marble; therefore it was likely that such oscillations were taking place within molecules rather than being due to movement of entire molecules or larger entities.

Subsequently, calcite was studied in more detail, both by Rubens (1911, Liebisch and Rubens 1919) and by others (Morse 1907, Nyswander 1909, Koch 1911, etc.). C. Schaefer and his coworkers (e.g. Schaefer and Schubert 1916, see Fig. 34-2, Schaefer et al. 1926) and others (such as Plyler 1929) compared the observed residual infra-red frequencies from calcite (probably Icelandic), other metal carbonates, saltpeter and further compounds to computations of their resonance frequencies. Using a selenium mirror for polarization, they could discriminate between oscil-

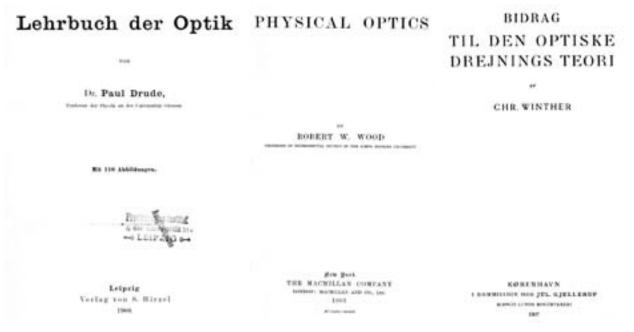


Figure 34-1. Left and center: Two of the best-known university textbooks in optics at the beginning of the 20th century. New editions of the book by Drude (1900a) appeared in 1906 and 1912 and were later reprinted; there were also two editions in English and one in French. Wood's (1905) book similarly appeared in three editions as well as reprints. Both covered a great deal of material related to polarization and double refraction. Right: A doctoral thesis in physical chemistry from Copenhagen University (Winther 1907). Research on or with polarized light constituted a significant part of many theses in fields within earth science, physics and chemistry, and also some in biology or engineering.

lations belonging to the ordinary ray and those belonging to the extraordinary ray. Some of the strongest reflections occurred at the same frequencies in all the isomorphous trigonal carbonates. The structure of these reflection peaks which presumably represented oscillations within the CO_3 -group, was the subject of special investigations (Schaefer et al. 1927). Other (lower) resonance frequencies which differed between these minerals could be due to the metal ion oscillating relative to that group as a whole. As mentioned in section 29.8, the residual radiation from calcite was also used up to the late 1920s in various investigations on other substances, such as metals. Later, the properties of residual rays played an important role in the interpretation of the Raman effect, see section 39.7.

34.3 Infra-red radiation and thermodynamics

In the 19th century, it was known that when a piece of solid matter was heated, the agitation of its constituent particles increased. However, little was known about the nature of those motions or their connection to the radiation from hot surfaces. If a piece of matter resembles a black body, the radiation takes place over a wide frequency range as is also well known. Hence, it is natural to assume that the movement of particles inside a piece of material is more or less of a random character, cf. the speed distribution of molecules in a gas. The unexpected finding that the molecular systems within crystals had only a few characteristic frequencies, became the foundation of a new hypothesis on their specific heats by Einstein (1907). For that purpose he employed the quantum theory along with the simple assumption that all the atoms in a given material are independent oscillators having the same infra-red resonance frequency. With input from a table of the residual

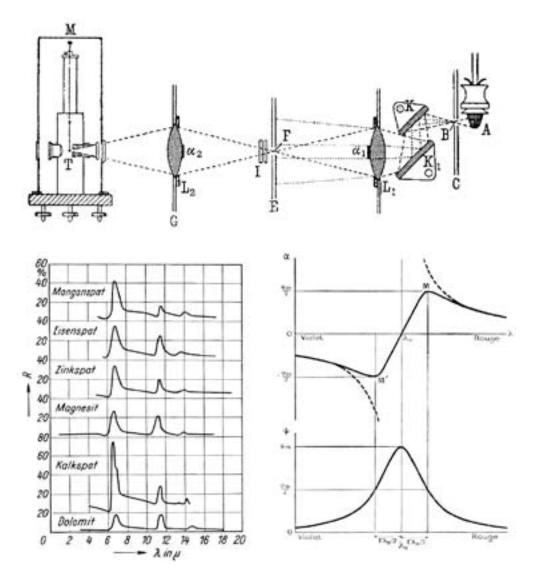


Figure 34-2. Top: The equipment used by Rubens (1911) for measuring infrared radiation (80-130 μ m wavelengths) reflected from the calcite plates K, K1. A is the lamp producing the radiation, which is then detected with the thermocouple T. Bottom left: In the spectrum of infrared light (5-15 μ m in this case) reflected from the isomorphous metal carbonates there are peaks at similar wavelengths. The vertical scale shows relative intensities in %. The peaks correspond to vibrations of atoms within the CO3-group (Schaefer and Schubert 1916). Right: Both the usual dispersion of light and its rotatory dispersion in matter are due to the fact that the light excites damped oscillations of atoms (Drude 1900a). The upper curve shows qualitatively how the index of refraction or the optical activity change as functions of the (vacuum) light wavelength around a resonance frequency of the electrons or the atoms in a material. The broken curve segments are plots of certain simple approximation formulas which are valid at frequencies far removed from the resonances. The lower curve indicates how the absorption, and the reflectivity as well, reaches a maximum at the wavelength of resonance (Bruhat 1942).

ray frequencies of calcite and four other transparent crystalline substances, he deduced formulas for the specific heat of each as a function of temperature. He also argued that because the high resonance frequency of electrons in solids, they did not contribute to the specific heat. Einstein's formulas agreed reasonably well with the rapid decrease in specific heat which experiments had shown to occur when the temperature approached absolute zero.

Einstein (1911) also considered the thermal conductivity of insulators in the light of the quantum theory, referring to measurements by Eucken (1911) of how the conductivity of calcite and some other crystals changes with temperature. The papers by Einstein attracted attention (see entry on W. Nernst in D.S.B.) but by 1911 it had become clear that his theory on specific heats needed improving. P. Debye and others soon succeeded in this, by looking at the thermal movements in a crystal as oscillations of the crystal structure as a whole, rather than of individual atoms. In A.H. Wilson's 1960 book: Thermodynamics and Statistical Mechanics, it is stated that all these researches "…had a considerable influence on the early development of the quantum theory.". They indeed pointed the way towards applications of quantum theory to the specific heat of other states of matter, other properties of materials, mechanical systems in general, and fundamental aspects of thermodynamics.

35 Polaroid and other polarizers after 1920; sugar measurements

35.1 Crystal growing and related matters

Attempts to grow large sodium nitrate crystals continued, see Stöber (1924), Kremers (1940), Bouhet and Lafont (1945) and West (1945). As remarked in section 33.3, researchers were for decades hoping that rhombs or plates of the saltpeter could substitute for Iceland spar in optical instruments. These attempts seem to have failed (Weinschenk 1925, p. 15, Rinne-Berek 1953), because the grown crystals were often cloudy inside, besides being soft and hygroscopic in spite of various coating methods applied to them. Halle (1921) also notes that they were too small and impure to be useful, a view seconded by Evans (1949) much later.

Furthermore, attempts were made to produce calcite crystals in the laboratory. I have not looked into that story in detail, but for instance Nester and Schroeder (1967) remark that "no one has been able to successfully grow crystals of even moderate size…". Similar statements may be found in Chandler (1970) and in Kinloch et al. (1974). On the other hand, large defect-free specimens of some other crystals for various uses in optics were grown from the late 1920s, such as various halides (LiF, KBr,…) which have good transmission properties in the ultraviolet and infra-red (Kremers 1940). After 1950 (Waesche 1960) perfect quartz crystals were made in large quantities, to some extent for use in optics but mostly for other purposes including in particular piezo-electric oscillators for clocks, telecommunications equipment, frequency meters and so on.

Still other avenues were explored in the search for calcite-prism replacements. One of these concerned so-called polarizing dispersion filters, probably made by crushing a uniaxial crystal like calcite to a coarse powder and immersing it in a matrix of clear jelly or resin (Bernauer 1930, Haase 1936-39; see also Schumann and Piller 1950, and Land 1951, p. 959). If the matrix has an index of refraction equal to that of the ordinary ray of the crystal, it should pass through the mixture without loss while the extraordinary ray is scattered. The idea is probably based on observations made by C. Christiansen in 1884, see Bennett and Bennett (1978). The most original suggestion in this field was to make the powder out of the calcite skeletons of echinoderms (Becher 1914). However, the dispersion approach did not reach any practical stage.

The development of glass-mirror polarizing arrangements continued where these were found to be useful, for instance in photoelasticity research (Smith 1933).

35.2 Herapathite and Polaroid sheets

It had been noticed in the mid-nineteenth century, that crystals of an iodine-quinine sulfate

named herapathite (e.g. Haidinger 1853) were a more efficient dichroic polarizer than tourmaline. Extensive searches among related and unrelated compounds for an even better material of this kind were unsuccessful (Haase 1936-39). Early on it was attempted to produce large herapathite crystals to replace tourmaline in tourmaline tongs (Haidinger 1855b) as well as Nicol prisms in microscopes (Valentin 1861, p. 106) and other equipment (see references in Pollard 1936). These attempts were also without permanent success until given attention at the Zeiss company by F. Bernauer (1930, 1935) who incidentally also wrote papers on the geology of Iceland. Zeiss began in the early 1930s to market single-crystal plates of herapathite (Grabau 1938, Schumann and Piller 1950) for use as polarizing filters in photography (possibly also in stereoscopic microscopes). However, these components which were sold under trade names like Herotar og Bernotar, were very fragile.

At this point, E.H. Land enters the stage. He became interested in optics as a youngster, when a summer-camp instructor in 1922 demonstrated with a Nicol prism the polarization of light reflected from a table top (McElheny 1998). This is reminescent of the 1808 discovery by Malus described in section 6.5. Land later studied the topic in books by Wood (1905) and others, and began in 1927 to explore ways to imbed a large number of very small aligned herapathite crystals in transparent sheets of celluloid (see Land 1951, McElheny 1998). His first patents on such products were awarded in 1933 and improvements soon followed, including crystals of complex compounds with better dichroic properties than herapathite (Pollard 1936). The sheets which Land called "Polaroid" were in some respects inferior to Nicol prisms, as they absorbed some of the desired light component; two sheets at right angles also let through a little light instead of extinguishing it completely. These drawbacks however did not matter in many less demanding applications (cf. list in section 33.1), and other advantages often outweighed them: they were inexpensive, had a wide field of view, took up little space in instruments, and did not need correction for astigmatism or color distortion as was caused by Nicol prism in microscopes. The Polaroid could be produced in any dimensions, so that for instance in photo-elasticity research large test objects could be viewed as a whole. If a quarter-wave retarder was needed, it was sometimes sufficient to use cellophane wrapping paper. At the same time, the lenses required for squeezing light through Nicol prisms (cf. Kimball's apparatus in Fig. 27-5) could be left out. The old term "polariscope" now became an appropriate one for such setups (Frocht 1941, Föppl and Mönch 1959).

One chief initial motive for the development of the polarizing sheets, was to incorporate them into the headlights and windscreens of automobiles, to avoid blinding drivers by glare (Grabau 1938, Land 1951, p. 962, McElheny 1998). However, this scheme was eventually dropped, because the appearance of the road and its surroundings became unfamiliar. Instead, car manufacturers began installing headlights with high and low beams. Polaroid on the other hand was a boon to photographers, as it for instance reduced unwanted reflections from window glass (Pollard 1936) which are always polarized to some extent. For the same reason, Polaroid sunglasses became popular with anglers, as they still are. That idea was in fact far from being original: in the first volume of Comptes Rendus (p. 405-407, 1835), F. Arago advised mariners that they can detect submerged reefs by watching the smooth sea surface through a tournaline plate. Bryson (1850) also described spectacles with Nicol prisms instead of lenses, allowing engineers to inspect the bottom of rivers or canals.

Improved versions of Polaroid sheets were in production by the early 1940s. They contained organic polymers (polyvinyl alcohol, etc.) forming elongated parallel molecules (Land 1951, McElheny 1998). By staining with iodine these molecules become dichroic, as H. Ambronn and others had found and investigated extensively in other substances much earlier (cf. section 29.9). The new sheets quickly found their way into some petrographic microscopes (Hallimond 1944, Hallimond and Taylor 1946).

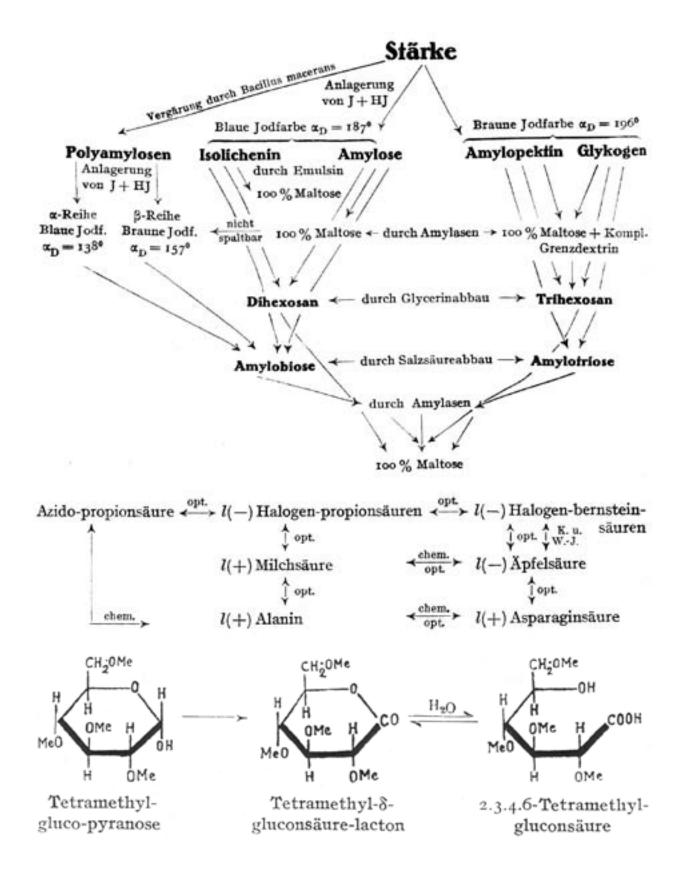


Figure 35-1. Top: A scheme from Pringsheim (1924) demonstrating the possibility of complete conversion of starch to maltose by hydrolysis and enzyme action. At that time, observations of optical activity had for 90 years been an important method in research on starch and its breakdown products. Middle: A scheme from Freudenberg and Lux (1928) showing close structural correspondence between two amino acids and other organic acids. "opt." indicates similarities deduced from polarimetric measurements. Bottom: A diagram from Haworth's (1932) review, showing how methyl groups attached to the glucose molecule aided in elucidating its configuration.

It had long been known that polarized light could be used in stereoscopy, where a three-dimensional image appears when two pictures of the same object, taken from slightly different angles, are viewed. J. Anderton was granted a British patent on such a method in 1891 (and a U.S. one in 1895). The two pictures were projected on a screen through tilted glass plates, and viewed through bulky tilted-glass spectacles. The technique was revived after 1940 or so (see Grabau 1938, Lipton 2001), and for a while around 1950 considerable production of three-dimensional pictures took place, later movies (McElheny 1998) for watching through Polaroid. However, showing the movies turned out to be a difficult and costly affair, requiring special metal-coated screens (a light beam being greatly de-polarized upon diffuse reflection from ordinary movie screens) as well as two identical synchronized projectors. This last point caused problems, as films quite commonly got torn in those days and had to be spliced.

35.3 On polarimetry, especially alternatives to it in the sugar industry

As is clear from this account so far, polarimeters were chiefly employed in two fields: one was research on optically active organic liquids and solutions, and the other was in the sugar industry. If the demand for Iceland spar decreased after 1920 as claimed by Hagalin (1970), literature from that period ought to contain evidence of increasing use of alternative methods in the commercial production and trade of sugars.

Let us start by considering how the value of raw sugar from producers was assessed before the advent of polarimeters. From the year 1861, such assessment was founded on the belief that whiteness could be equated with purity. In the U.S. for instance, glass vials containing sugars with some 25 color nuances from dark brown to white were purchased annually from the Netherlands for the estimation of sugar colors in shipments arriving from the Caribbean. These "Dutch Standard" colors also formed the legal basis of taxation, which was no small matter as sugar provided a sizable proportion of all federal U.S. import tax revenues. The parties involved were becoming quite dissatisfied with the color method by 1880, because production methods had evolved in such a way that the color of raw sugar did not always reflect its purity. Also, sellers were tempted to add artificial coloring to their produce in order to reduce its tax value (Warner 2007). Following extensive discussions, disputes in Congress and court cases on these matters, polarimeters played an increasingly important role in U.S. raw-sugar evaluation from 1882. Some historians claim that this change contributed to New York gradually taking over from Havana as the chief center of sugar trading. However, the Dutch Standard colors continued being used in quality assessment of some of the imported sugars and molasses to 1913, in Canada even into the 1930s.

In Europe, polarimetry was almost the only method in use for measuring the sucrose content of raw-sugar products in the industry, at least until around 1900 (Fresenius 1896). However, such precise measurements of optical activity in solutions of sugar raw materials, intermediates or finished products, were not quite straightforward especially if other sugar compounds were to be measured concurrently. It was necessary to eliminate colorings, colloids and other impurities by filtering and precipitation, treat the samples with acids (later enzymes) or by heating, calibrate and adjust the instruments precisely, clean all sample vessels, and carefully control the temperature during measurements. In the analysis of sugar products, the following three types of methods may gradually have become realistic alternatives to polarimetry (see e.g. Saillard 1913, p. 34-64).

1) Many chemical methods were being developed during the late 19th and early 20th centuries (see Chapter IX of Bates et al. 1942). Most of them involved precipitation of copper compounds, as first described by H. Fehling in 1849. Rolfe (1905) states that up to 30 versions of that procedure had been described, and many more appeared later. It may be concluded from Hudson's

(1910a) paper that measurements of sugar concentration using Fehling's solution are about as common in the sugar industry as polarimeter measurements. However, Cajori (1922) indicates that polarimeters are almost always used in simultaneous analysis of samples for more than one saccharide. Shaffer and Hartmann (1921) describe a convenient copper-iodine method for measuring glucose in various liquids, and Cajori (1922) introduces iodine methods for estimating other sugars. Additional examples of chemical procedures include colorimetric measurements on reaction products of sugar or other hydrocarbons and picric acid (Dehn and Hartman 1914) and a colorimetric method based on the reduction of ferric oxide (from around 1923). In tests on urine, the decrease in its density upon fermentation, or the volume of carbon dioxide produced, was often used as a measure of glucose content (e.g. Struers 1925, p. 415-416) but this method was time-consuming and inaccurate.

2) Measurements of the density of aqueous solutions of sugar, either with the aid of hydrometers (invented by Brix, Baumé, Westphal or others) or by weighing of containers of accurately known volume (pycnometers, etc.), were convenient to perform but their results were not as precise as those from the polarimeters.

3) After 1910, measurements of refractive indices increased rapidly in popularity as it had been demonstrated that they were superior to density measurements. The refractometry also required only a few drops of solution, and it was not necessary to carry out precipitations as in the polarimetric method. "When by experience there is a close correlation between sugar concentration and total content of dry matter, the refractometer is much to be preferred to the polarimeter...." (Löwe 1925). In the 1920s refractometers were also built into the walls of processing pans in sugar factories (Hinton 1930).

It is possible that by the early 1920s the above alternatives had collectively gained a wide enough application to reduce somewhat the need for polarimetry in the sugar and food industries and in the sugar trade. Thus, a partial explanation may be found for the claim in Hagalin's (1970) book mentioned in the first paragraph of the present section. However, statements like this one from the U.S.: "...every sugar-house uses the [polariscope] for routine control work of its manufacturing processes." (The Cotton Oil Press, Chemists' Section 4, p. 43, 1921) show that polarimeters still were important. As far as taxation is concerned, it is also clear that in U.S. Customs regulations on sugar tariffs issued both in 1931 and 1937, polarimeters/saccharimeters with Nicol prisms and quartz plates played a key role. Singerman (2007) even states that polarimeters of the ingenious type designed by Bates (1908) were the main instruments used by U.S. Customs for sugar measurements in the 1940s. With regard to imports and production of sugar in the U.K., one may similarly conclude from perusing The Times newspaper of London from 1920-32 that polarimetry was still being depended upon in the trade.

As far as research on the structure of optically active compounds and their reactions was concerned, polarimeters containing Iceland spar prisms continued to be indispensable in the 1920s and beyond. Thus, Fig. 35-1 shows three diagrams published in 1924-32, based largely on polarimetric measurements. One is a scheme of methods for the quantitative degradation of starch to maltose, another reveals close similarities between certain organic acids, and the third illustrates the use of glucosides in work on the detailed configuration of the glucose molecule.

36 X-rays and other research on radiation and materials, 1895-1930

36.1 Röntgen

From 1913, calcite crystals from Iceland were given challenging tasks in a new and important field of the physical sciences, namely the use of X-rays in research on the submicroscopic structure of crystals and of atoms. Their discoverer in 1895, W.C. Röntgen (see Appendix 5) was himself interested in crystallography, and he also made observations with Nicol prisms. These included studying the Kerr and Pockels electro-optic effects (see chapter 23) and demonstrating that the Faraday magneto-optic effect could take place in gases (Kundt and Röntgen 1879a,b). Röntgen (1896) mentions in his first paper on the rays named after him, that he had found Iceland spar to be transparent to them and that they caused luminescence in this material. He considered that the rays might be a form of longitudinal waves. In this paper he also reported measuring the fluorescence of another material with the aid of a Weber photometer; however, it is not clear whether this was the 1891-model of that device (containing Nicol prisms, see section 29.4) or an older version. Later, Röntgen (1907) tested whether his X-rays caused any changes in the electrical conductivity of calcite. Some other scientists attempted using Iceland spar in research on radioactivity which was discovered in 1896, but it turned out not to have any notable properties in that respect.

36.2 Scattering and diffraction of X-rays; the structure of calcite

C.G. Barkla (1904) published the first observations on scattering of X-rays (from the particles of solid matter) which showed that they had polarization properties like light, cf. the drawing in Fig. 27-1. In the following years H. Haga, R. Bassler, J. Herweg and others confirmed that they were waves; however, no means of measuring their wavelengths were available.

In 1911 it became clear that atoms were composed of very small nuclei and electrons orbiting around them. A young physicist, P.P. Ewald, had selected as his doctoral thesis project to study theoretically how the atoms in crystals could cause double refraction of light. He asked another physicist, M. v. Laue who was familiar with both wave optics and crystallography, what would happen if light waves impinging on a crystal had a wavelength of similar magnitude as the distances between its atomic nuclei. Laue then realized that the crystal would affect the X-ray waves in a similar way as a diffraction grating affects visible light. Laue and his collaborators demonstrated this experimentally in early 1912 (Fig. 36-1), attracting much attention. It should be noted here that the various expressions describing these phenomena, have not always been used in a transparent way in the literature. Thus, in the case of visible light "diffraction" usually refers to

the bending of the light around an extended obstacle assumed to be made of an opaque material continuum, whereas in X-rays both this term and "reflection" often refer to situations that might be more appropriately called "combined scattering" from many particles.

By measuring the directional change of a parallel beam of monochromatic X-rays upon transmission through, or reflection from, a crystal plate (Fig. 36-1), their wavelength could be found in terms of some characteristic interatomic distance. The interaction process was in this case much more complex than in an ordinary grating, because the deflected X-rays emanate from a three-dimensional network of particles rather than from a flat surface with parallel lines. In 1913-17 P.P. Ewald published his theoretical investigations on the matter, introducing various new concepts. With extensions from e.g. C.G. Darwin and C.W. Oseen, this came to be called "the dynamical theory". Their remarkable achievement had one main drawback: it only applied to crystals that were perfectly regular in their structure. See more on this in section 36.3. Simultaneously, experimental techniques were improved from the original methods of Laue and others, particularly by W.H. Bragg and his son W.L. Bragg. The samples used were either individual crystals or powdered ones. As almost all of these were quite far from being perfectly regular, an interpretation procedure appropriate for them was developed and used in the following decades under the name of "the kinematic(al) theory".

Calcite was one of the first crystals on which X-ray diffraction was tested, and it turned out to be particularly straightforward to estimate the main features of its structure from the deflection angles (W.L. Bragg 1913, 1914). As remarked by W.H. Bragg (1915, footnote p. 268). "...no case has yet occurred in which the opportunities for comparison are so direct as in calcite, with its different sets of equally spaced planes". Schiebold's (1919) doctoral thesis dealt with the structure of calcite, from X-ray observations on a glass-clear Icelandic specimen of about 20 mm size. He also looked into aspects like twinning and glide planes (cf. section 21.3). In the interpretation of the complex diffraction patterns in these early years, it was also of great help to have good crystals of isomorphous crystals such as iron-, zinc- and manganese carbonates for study along with those from calcite (see e.g. W.L. Bragg 1914, Jaeger and Haga 1916, Wyckoff 1920, 1922). Therefore, the precise geometrical arrangement of the atoms in calcite was known quite early (see Ewald 1923, Fig. 36-1). With that knowledge, physicists could start figuring out how this arrangement was connected to the various physical properties of calcite. Thus, W.L. Bragg (1924) suggested reasons why the relative positions of the calcium, carbon and oxygen atoms in calcite and aragonite (both having the chemical formula CaCO₃) might explain their unequal double refraction.

Later researches (see e.g. Wahlstrom 1969) have revealed that the unusual refraction properties of calcite are related to the planar shape of the CO_3 -groups. In an analogous way, the reason for the easy cleavage of Iceland spar along its three main directions is due to unusually weak forces between atoms across those planes (Beckenkamp 1923, and other authors). This fact was of considerable significance in X-ray work, because the stresses arising in cleaving or in the polishing of surfaces can cause deformation of the adjacent molecular layers. Bragg and Chapman (1924) made a reasonably successful attempt at explaining why the crystal angles vary from one trigonal metal carbonate to another.

Various studies were made on X-ray diffraction from optically active crystals like quartz and cinnabar, in order to aid theoreticians' efforts in elucidating the causes of this property (see e.g. Born 1935).

36.3 The role of Iceland spar as a yardstick for X-rays

Soon after 1913 it was realized that the laboratories investigating X-rays needed a common eas-

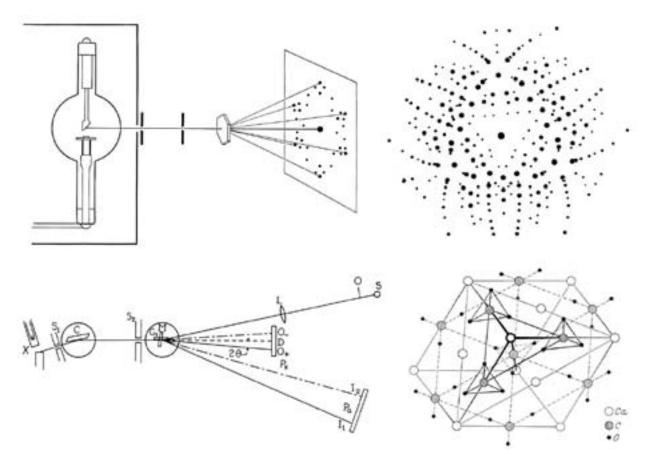


Figure 36-1. Top: In 1912 M. v. Laue sent a narrow beam of X-rays through crystal plates. On development of a photographic negative behind the plates, it was seen that the rays had been deflected into specific directions, because the crystals act like a three-dimensional diffraction grating. On the right, the Laue pattern for calcite (from Rinne 1922b) is shown; Iceland spar was probably among the first crystalline materials tested with this technique. Bottom left: The arrangement of the atoms within a crystal structure may be found by mathematical processing of X-ray patterns like those above. However, better results are obtained if the rays are reflected from a plane surface of the crystal C, and angles like Θ are measured accurately. The diagram illustrates the setup of Wadlund (1928) which gave a precise value of the spacing of atomic planes in calcite (possibly from Iceland). From that value, Avogadro's number could be derived. G is a diffraction grating for the X-rays. Right: A drawing by Ewald (1923) of the fundamental unit cell of a calcite crystal; a similar one was published already by Bragg (1914).

ily accessible and accurate reference method for measuring the X-ray wavelengths employed in crystal-structure determinations. This was also necessary in order to chart accurately the complex emission and absorption spectra of the elements, see section 36.5. At that time, their wavelengths were considered to be much too short for analysis by gratings (i.e. metal plates with parallel scratched lines) to be feasible.

In the search for such a yardstick, the first choices were simple cubic two-element crystals like NaCl (rock salt), KCl (sylvin) and ZnS (zinc blende). It is easy to compute their interatomic distances (grating space) if the crystal density and the atomic weights of the elements are known. These materials, however, did not perform as well as calcite. Thus, Compton (1916, see Fig. 36-2) speaks of "extremely perfect faces" of calcite crystals, later reiterating (Compton 1918) that "…calcite is more perfect and is less apt to contain inclusions than rock-salt". The salt gave stronger X-ray reflections than calcite, and some researchers learned to select adequate specimens of it (e.g. Siegbahn 1919, p. 605). On the other hand it had an uncomfortably high thermal expansion coefficient, and therefore calcite was preferred by many from around 1920 as a length standard for rays having relatively short wavelengths (< 0.5 nm). Siegbahn (1933) in fact became disillusioned

with rock salt when he found in his improved equipment that "it was impossible to obtain sharply defined spectral lines" with it. He adds that calcite crystals are "the most frequently used ones for precision measurements as well as for studies on the structure of spectral lines". Here he was no doubt referring largely to samples of Iceland spar (from Iceland, Montana, Spain, Argentina, etc.) that had been recovered from drawers in research institutes and museums and enlisted in the service of X-ray work.

In some important investigations of X-ray diffraction, much improved resolution could be attained by using two analyzing crystals in succession, having exactly parallel surfaces (Fig. 36-2 bottom right). In this method which was invented by A.H. Compton in 1917 and became "an invaluable tool" in the field (according to Parratt 1932), calcite crystals were mostly employed. For instance, Davis and Stempel (1921; see Hildebrandt 1995) record that they split "a clear specimen of Iceland spar" for this purpose, while Wagner and Kulenkampff (1922) state that in rock salt it is "most difficult...to attain good cleavage planes". Other relevant opinions which may be quoted here are: "The calcite possesses approximately the same dispersing power as rock salt, and good specimens of it are easily obtained" (Siegbahn 1924, p. 85), and "...calcite crystals are much more perfect than...rock salt" (Compton et al. 1925), and "...for calcite forms very perfect crystals" (Havighurst 1926). Cooksey and Cooksey (1930) were given "a nearly perfect specimen of Iceland spar" from the Yale University collections for use in a new determination of its grating space; they measured the deflection of a beam with an accuracy better than 0.1" (see also a note under Havighurst 1926 in the reference list). Some scientists such as Wyckoff (1926) considered quartz to be an alternative as a precise wavelength standard, whereas others had less faith in it due to "variation in [its] internal structure" (Tu 1932).

Calcite was therefore one of the most commonly used crystals in precision X-ray spectroscopy to 1950 or even later (see e.g. Wadlund 1928, Fig. 36-1, Bearden 1931b, 1935, Birge 1941). Bearden used for his measurements two crystals "split from a block of Icelandic calcite" and Cork (1932) claims that the most accurate values of X-ray wavelengths up to at least 5 Å (0.5 nm) "have been obtained almost entirely by the use of calcite crystals". Ievins and Straumanis (1940) recommend calcite, rock salt being "unsuitable for precision measurements because of its mosaic structure". Batuecas (1950) states that in calcite the density, grating space, and geometrical properties are known with more accuracy than in any other crystal. Yarwood (1963, p. 633) informs us that in the standardization of X-ray wavelengths after 1930 calcite was preferred to rock salt because the hygroscopic nature of the latter spoils its surface properties. Bearden (1965) expects that calcite is still the most common analyzing crystal. However, at that point in time instrumentation had reached sufficient precision to determine that grating space values varied between individual calcite specimens by amounts of the order of 1/100,000. This was inconvenient and physicists therefore searched for better reference standards, such as the use of particular very sharp X-ray spectral lines.

Until around 1932 the above-mentioned "dynamical theory" of X-ray diffraction was developed further, but few crystals were found to be of sufficient perfection to enable testing of its predictions. However, this was eventually possible "thanks almost exclusively to the best specimens of Icelandic calcite" as recalled by G. Borrmann (1988, see also in Brill 1962 and Hildebrandt 1995) although studies on for instance diamond and zincblende also provided some useful evidence. Later, Borrmann made important discoveries regarding the interference of X-rays around 1940, by their transmission through "a small piece of Icelandic calcite" (Hildebrandt 2002) which he had procured from a mineralogical collection. Borrmann in fact found that much more of the radiation passed through the crystal in a Laue-type experiment than one could expect by comparison with other crystalline materials. Due to circumstances in the World War his results were not published at that time, but Borrmann (1950) continued his work on the same spar piece and other crystals

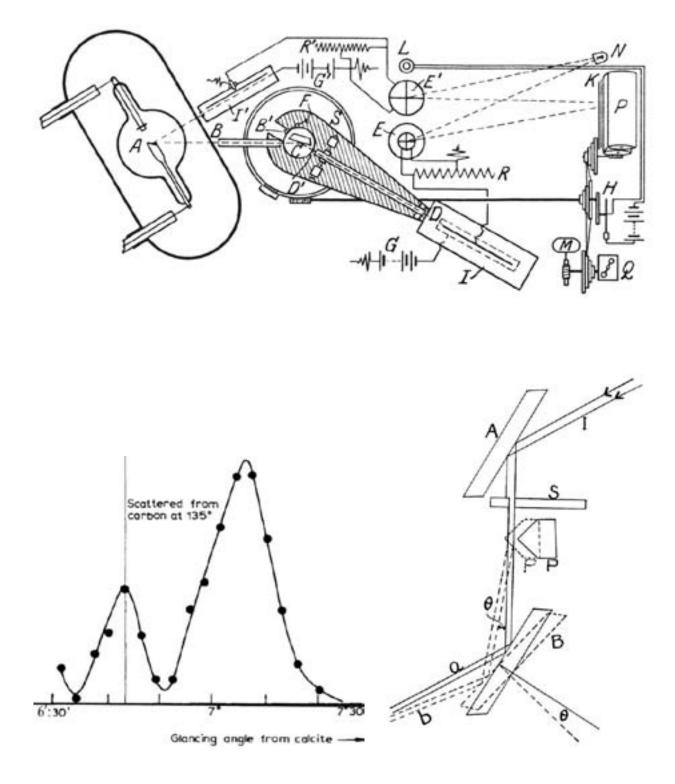
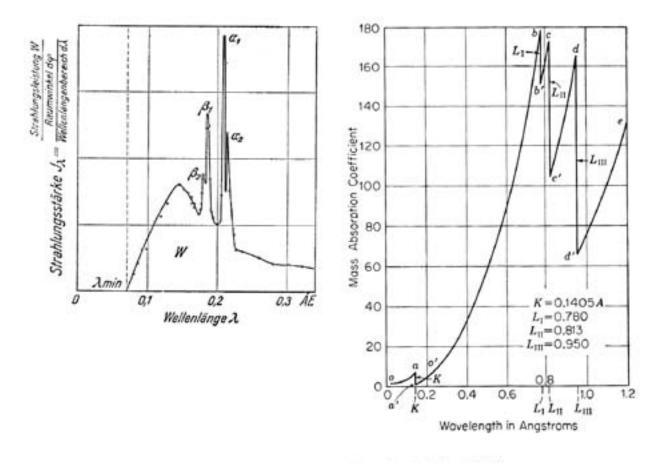


Figure 36-2. Top: The directional change of X-rays on reflection from a crystal C was often measured by means of an ionization chamber I on an arm which could be rotated around C. The figure shows the instruments of A.H. Compton (1916) who often used Iceland spar in his spectroscopic work. From Siegbahn (1924). Bottom left: Compton (1923) found that if a narrow X-ray beam passed through for instance a lump of graphite, the wavelength of a part of the beam increased (right-hand peak). This was caused by collisions between the rays and electrons. It agreed with the quantum theory of A. Einstein which predicted that individual photons of radiation carry momentum. The change in wavelength was around 1% or less, and the experiment called for good analyzing crystals like calcite. The graph is from Compton's Nobel lecture in 1927 (www.nobelprize.org). Right: Very high resolution of X-ray directions and wavelengths could be obtained by letting the rays impinge on two parallel surfaces of the same split crystal, here A and B of Iceland spar. If the width of the X-ray beam was limited by a narrow slit in the screen S, the wide aluminium wedge P could be shown to cause a "refraction" of the beam. The angular change was only a few seconds of arc.



k-Werte der L-Reihe. Emission.

	92 U	90 Th	83 13	82 Pb	81 TI	80 Hg	79 Au	78 Pt	77 Jr	76 Os	74 W	73 Ta	72 H?
1	1064,77	1112,41	1312,95	1346,62	-	1418,3	1456,54	1497,23	kanada	week	1675,05		
×2	920,14	965,24	1153,3	1183,52	1216,03	1249,7	1284,89	1321,21	1359,39	1398,2	1484,52	1529,4	1576,70
N ₁	906,33	953,42	1141,15	1172,02	1204,71	1238,5	1273,55	1310,08	1348,34	1388,16	1473,48	1518,25	1566,14
	802,9						1199.5				1417.7		

Figure 36-3. The wavelengths of characteristic features in the X-ray spectra of the elements had to be evaluated accurately, in order to test theoretical predictions regarding atomic energy levels. Top left: The spectrum of the X-rays emitted by wolfram metal when hit by very energetic electrons. From R.W. Pohl: Optik und Atomphysik, 1954. Right: Absorption spectrum for X-rays in lead. The energy of each photon decreases with increasing wavelength. K refers to the innermost orbit of electrons around a nucleus, and L refers to the next orbit. From F.K. Richtmyer et al.: Introduction to Modern Physics, 1955. Bottom: Part of a table of wavelengths of L-series peaks in the emission spectra of several metals (Siegbahn 1924). They are given in the X-units introduced by Siegbahn, which were defined in terms of the interatomic spacing of Iceland spar.

from 1948. He then made further remarkable findings regarding the dynamical theory, which others had difficulties in believing until they were confirmed around 1955 by measurements on "a 32 mm thick Icelandic calcite plate of the highest perfection." (Hildebrandt 1995). Renninger (1955) who also made experiments on Borrmann's samples to confirm predictions of the dynamical theory, states that calcite is still the most perfect crystal available.

Attempts to grow large pure crystals of for instance rock salt were not successful until around 1925 (see D.S.B. on R. Pohl), and after 1950 very pure crystals of aluminium, silicon, germanium and other materials were produced. In the 1960s such man-made specimens had reached sufficient perfection for them to be used in tests of the dynamical theory, encouraging its further development. The new materials also replaced Iceland spar in the role as an X-ray wavelength standard.

Out of these materials, the semiconductors silicon and germanium had very valuable properties, as tiny amounts of impurities can greatly influence the energy levels available for their electrons and therefore change their electrical conductivity. The knowledge which had been obtained by applying the dynamical theory to X-ray diffraction in Iceland spar and other near-perfect crystals, improved understanding of the nature of these semiconductors and thus hastened the electronics revolution in the second half of the 20th century. It may also be concluded from the literature on these subjects, that the dynamical theory played a significant role in the revolution itself, because it was used in the interpretation of data from X-ray analyses (carried out both by conventional means and by synchrotron radiation from particle accelerators) on many other perfect man-made crystals. These included semiconductors, piezoelectric crystals like quartz, ferroelectric crystals, photoelectric materials, magnetic materials, crystals for use in lasers and in light modulation, etc.

Some other important stages in X-ray studies were reached with the aid of calcite crystals. For instance, Siegbahn (1921) used calcite to confirm results which his collaborators had obtained on gypsum, indicating that refraction was taking place at the surface of these crystals. After this discovery, Davis and Terrill (1922) were among the first to measure directly refraction angles for X-rays, in an Iceland spar crystal. The results agreed with theoretical calculations on the interaction of the rays with electrons. These angles are very small indeed, as the index of refraction of all materials for X-rays is very close to 1. Actually it is less than 1, which opens up possibilities for the total reflection of X-rays; this has in recent years been utilized in various ways. Later, the directional change of X-rays in metal prisms was measured; it is only a few arc-seconds. In one such experiment (Davis and Slack 1926) the measurement was made with the above-mentioned double spectrometer of Davis and Stempel (1921) containing two halves of a recently split Iceland spar crystal; the authors conclude that the high quality of the surfaces was an important factor in their success. Still more precise results on refractive indices were obtained in a new spectrometer designed by Compton (1931) with two crystals "freshly cleaved from an optically perfect sample of Iceland spar". In the first test of the effect of low temperatures on X-ray reflection coefficients (for comparison with the quantum-mechanical theories of P. Debye and others on lattice vibrations), Nies (1926) employed "a transparent cleavage piece of Icelandic calcite".

Iceland spar makes an occasional appearance in early efforts to use X-rays in specialized chemical analyses. Thus, Glocker and Frohnmayer (1925) used Nicol prisms in apparatus to measure the darkening of photographic plates when X-rays were employed in making quantitative estimates of the content of some foreign elements in glass and minerals. Later, C.H. Eddy and his co-workers had a calcite diffracting crystal in their X-ray equipment when attempting to detect minor constituents in samples of materials (e.g. Eddy et al. 1929). They state that their method can measure concentrations of trace elements down to 0.00001%.

Around 1924 L. de Broglie suggested that material particles could behave like waves. Experiments to demonstrate the wave-like properties of electrons, by scattering from metal crystals, began yielding positive results in 1927. It is more difficult to use insulators in this sort of work because they become charged, but Kikuchi and Nishikawa (1928) were the first to detect diffraction effects in fast-moving electrons when interacting with a cleavage face of calcite.

By 1936 people were directing neutron beams towards samples of liquids and solids, and observing the scattering of the neutrons. This technique has been important in investigations of material structure. It was also found that various nuclei absorbed neutrons, some becoming radioactive, but these processes were very dependent on the speed of the neutrons. Calcite was one of the crystals employed in pioneering studies of that kind, see e.g. Rasetti (1940) who used "a perfect specimen of Iceland spar" in a part of his project. Interest in these matters greatly increased when nuclear energy was being harnessed, as neutron capture by uranium nuclei could bring about fis-

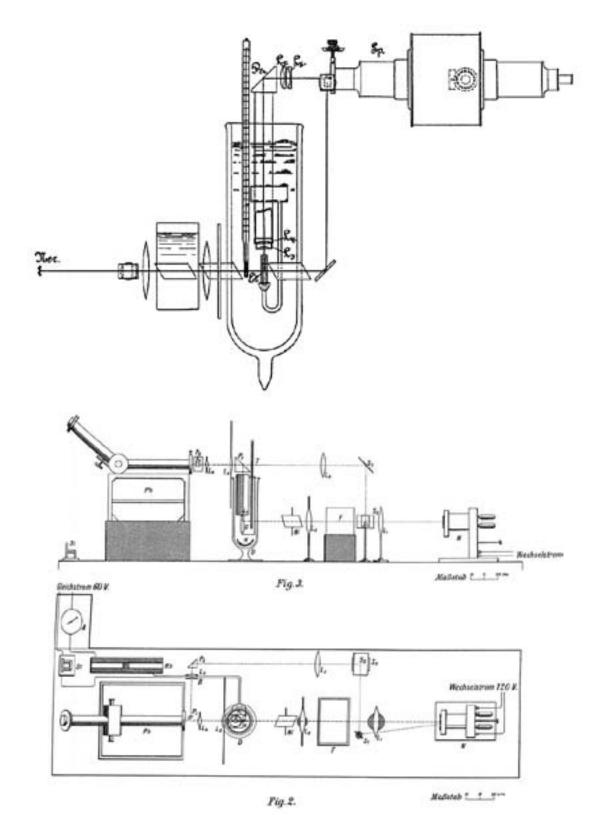


Figure 36-4. Top: The measurements of Onnes and Keesom (1908) on light scattered in a sample Et of ethylene (in a temperature-controlled bath) near its critical temperature. The light source is the lamp Ner on the left. Three Nicol prisms are used in this experiment, which is relevant to contemporary theoretical works by M. Smoluchowski and A. Einstein. Bottom: With this setup (seen here both from above and from the side), Fürth (1915) confirmed conclusions by Einstein (1910) on light scattering "mit grosser Annäherung". Light from the lamp N is polarized by the Nicol prism Ni and passes through an enclosed sample of a water-phenol mixture, submerged in the glass vessel D. The intensity of the light is measured with the König-Martens photometer Ph (which contains Nicol and Wollaston prisms of Iceland spar) as the sample temperature is varied.

sion. Thus Fermi and Marshall (1947) investigated the scattering of neutrons on their way through calcite and many other materials, and Havens et al. (1948) borrowed for similar research a large Nicol prism whose owner many decades before may have been J. Tyndall.

36.4 Distances within crystals

Computations of the fundamental interatomic distance d in all crystalline materials depend on the value of Avogadro's number, i.e. the number of atoms in one mole. This number can be obtained by dividing the amount of electrical charge in one gram-equivalent (called Faraday's constant) by the numerical value of the electron charge e. The latter was for a long while only known from measurements by R.A. Millikan, published in 1910-17. In estimating it, he made use of the velocity of oil drops falling through air, and the results only had sufficient accuracy to express d with four figures. In rock salt for instance, d was determined to be 0.2814 nm. Relative measurements in X-ray spectroscopy had however by 1924 reached six-figure accuracy (Siegbahn 1924, p. 20). For inter-laboratory comparisons, Siegbahn therefore introduced a new unit defined on the basis of the d of Iceland spar. This he called the X-unit but it was later named after himself.

In the year 1925 it was possible for the first time to measure X-ray wavelengths by reflection from a ruled metal grating under incidence angles close to 90° (see Yarwood 1963). The results did not quite agree with those expected. In a doctoral thesis in 1928, E. Bäcklin pointed out that Millikan's e-value must be too low by about 0.4%, and eventually this discrepancy was traced to an incorrect formula for the viscous resistance of air to the motion of Millikan's oil drops. This had the effect of changing e.g. the rock-salt d-value from 0.2814 to 0.28197 nm. In the following years therefore, procedures were reversed: the electron charge was computed from the properties of calcite after its d had been found by comparison with a ruled grating (see Bearden 1931a, 1935). Bollman and DuMond (1938) studied in particular on two Iceland spar specimens (provenance not given) whether the very thin surface layer involved in reflection experiments had the same properties as their bulk substance. This turned out to be the case, "...furnishing clinching justification for the validity of the well-known precise determinations of e by Bragg reflection from the surface of macroscopic pieces of calcite". According to Birge (1941, p. 119) the oil-drop method was still not yielding a sufficiently precise value of e, mostly because of uncertainties in the above-mentioned air resistance.

The electron charge was involved in the determination of some other quantities. For instance, the electron mass m could not be measured directly, whereas various experimental methods were available to find the ratio e/m accurately. In this way, Iceland spar contributed to improvements in our knowledge of the electron mass.

36.5 The fundamental constants of physics; atomic structure

Iceland spar had further important roles in connection with X-ray research. One had to do with precision measurements of the constant h which is fundamental to the quantum theory introduced by M. Planck in 1901. h is a mechanical quantity but no purely mechanical methods were available to measure it directly. Instead, the ratio h/e was obtained from measurements on electronic processes. After the appearance of Einstein's (1905a) hypothesis that light of frequency f has some properties of particles carrying energy quanta of magnitude h.f, scientists attempted to use the photoelectric effect to estimate h. However, in the beginning these experiments suffered problems due to various disturbing factors.

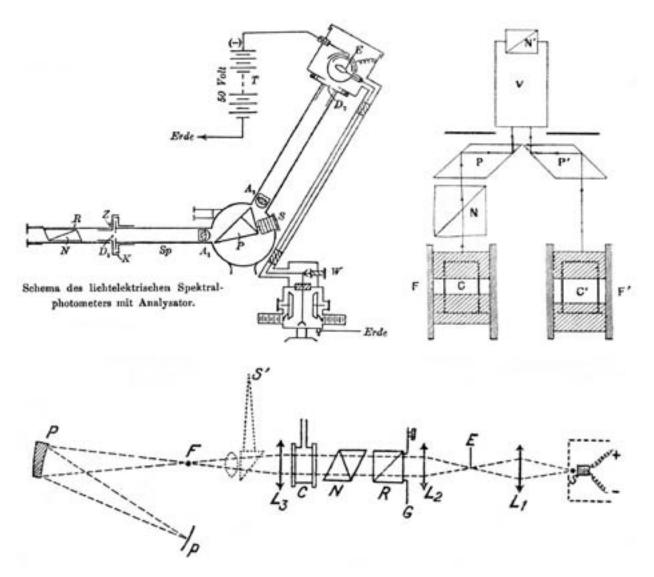


Figure 36-5. Optical instrumentation for research on gases and liquids. Top left: A photometer with elecrical recording, used by Dember and Uibe (1918) for analysis of the spectrum of sky light as well as of its polarization. The light arrives from the left through the Nicol prism N. Right: A vertical section through the equipment designed by Lépine (1915), for comparison between the fluorescence properties of dyes in different concentrations and different solvents. The experimenter looks from above through the Nicol prism N' and the telescope V, and turns N until light from fluids in the containers C and C' appears to be of equal intensity. P and P' are glass prisms. Bottom: Ribaud's (1919) measurements of the absorption of visible and ultraviolet light by bromine vapor in the vessel C at various temperatures. A lamp is to the right and a spectroscope to the right. R is a Rochon prism, N is a Nicol prism and the L's are quartz lenses.

R.A. Millikan succeeded in eliminating the main error sources in a famous series of measurements on the photoelectric effect in alkali metals, published in 1914-16. His result agreed well with that deduced by Planck from the spectrum of black-body radiation. It is not as well known today, that at a similar time very satisfactory values of h were being obtained from the "inverse photoelectric effect" of X-rays. Here, one determines the minimum voltage difference V needed for the acceleration of electrons, in order to create X-rays of a given wavelength (λ_{min} in Fig. 36-3, upper left) upon collision with a metal surface. In short, e.V = h.f = h.c/ λ_{min} where the wavelengths λ were measured with a calcite crystal in the earliest studies by Duane and Hunt (1915) and Webster (1916). As is evident e.g. in Millikan's (1917a) conclusions, the close similarity of h-values obtained by very different methods provided strong support for the new quantum approach to physics in the 1910s and 1920s. At that time the quantum theory was being developed rapidly, while some scientists (including Millikan himself, cf. his Nobel lecture in 1923) considered its foundations to be somewhat insecure.

Out of six published series of X-ray measurements of h quoted by Siegbahn (1924, p. 201), the above-mentioned two and two later studies used calcite as a wavelength standard. At that point, the oil-drop value of the electronic charge governed the accepted value of the grating space of calcite and other crystals, so that a systematic error in e could affect the experimental value of h in more than one way. All the six X-ray series and most of those in Siegbahn's list employing other methods yielded h in the range $(6.50 - 6.58) \cdot 10^{-27}$ cgs units. As pointed out e.g. by Millikan (1917a,b) and reiterated by Bollman and DuMond (1938), the electron mass m, its charge e, the speed of light c and Planck's constant h all enter into the so-called Rydberg's constant R. It has the dimension 1/length, and it can be found quite accurately by measurements on the spectra of hydrogen and helium with the aid of ordinary diffraction gratings. Therefore, the agreement between values of R_m obtained in this direct way and indirectly from the various measurements of h and other quantities, provided a further test of the quantum theory. Precision determinations of these fundamental constants gradually converged. The source of a bothersome 0.2% systematic error (unrelated to the analyzing crystals) in the X-ray spectrometry values of the quantity h/e was finally eliminated in 1941. Birge (1941) and Bruhat (1942) therefore consider the consistency of the constants to be quite satisfactory, but improvements of experimental techniques continued. Thus, Aminoff (1946) suggested a way to increase the X-ray resolution provided by Iceland spar, and Bearden and Schwarz (1950) found h/e to six figures in a new spectrograph employing an Iceland spar crystal (previously used by Bearden 1930, probably Icelandic).

As soon as physicists began studying the emission and absorption X-ray spectra of the elements, they found that these are partly composed of individual lines, partly of continuous bands that stop abruptly at a characteristic wavelength. Various rules regarding the wavelength values in these spectra were proposed, one of the first, simplest and most important being that of H.G.J. Moseley. It stipulates that the shortest X-ray wavelength λ_{min} radiated by an element (Fig. 36-3 top) with an atomic number Z is proportional to $1/(Z-1)^2$. This rule was originally found in 1913 from measurements with a potassium ferrocyanide analyzing crystal on metals having Z = 13 to 47. Moseley's rule on this so-called K-series of X-rays was subsequently confirmed by further spectrometric work on heavier elements where both calcite, rock salt, gypsum and sucrose were employed (cf. chapter IV of Siegbahn 1924). The elements emit the K-rays when electrons fall down to their innermost allowed orbits. Calcite was also used in part in the 1920s for the charting of the complex L- and M-series of X-rays from the heavy elements (Fig. 36-3 bottom), where the availability of a good analyzing standard was clearly much appreciated. Thus, Coster (1921) used a "very beautiful" calcite crystal in his equipment, and Kellström (1927) had at his disposal "two different crystal specimens of excellent quality". The absorption spectra of the elements (Fig. 36-3) which were no less interesting than their emission spectra, were also in many cases investigated with calcite crystals (e.g. Duane and Hu 1919, Lindh 1925). It was important to carry out the spectral analyses with great precision, for comparison with the quantum theories of N. Bohr, A. Sommerfeld, G. Wentzel, W. Kossel, L. Vegard, R. Kronig and others on atomic structure. In the early 1930s the above-mentioned calcite double-crystal spectrometers were improved in order to study in detail the widths and shapes of the individual X-ray spectral lines (e.g. Richtmyer et al. 1934).

In the period 1920-50, X-rays continued to be used by many laboratories in finding the relative positions of atoms in innumerable inorganic and organic crystals, and in investigating various processes involving these compounds. I have not checked whether the use of calcite in that field had any decisive effect on its development.

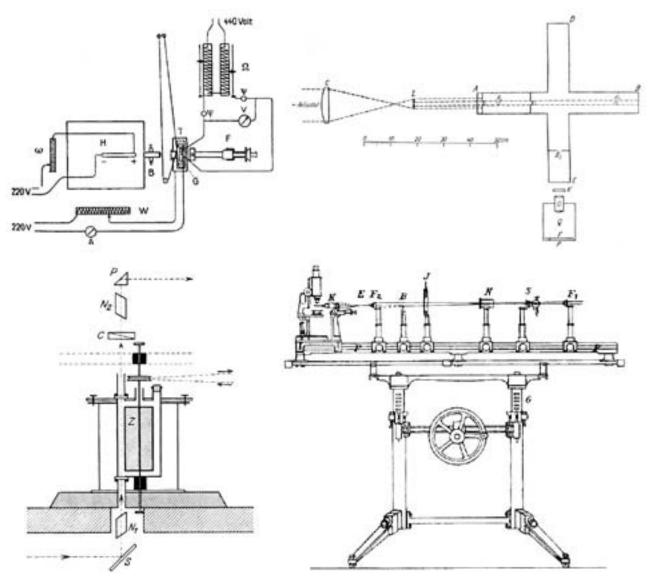


Figure 36-6. More research on gases and liquids. Top left: Björnståhl's (1918) work on the effect of electrical fields on refraction in liquid crystals. A sample in the container G is illuminated with the lamp B, and the light is then analyzed in the König-Martens spectrophotometer F. Four rheostats control the electrical field strength, the light intensity and the temperature. Right: The equipment of Gans (1921), here seen from above, contributed to improvements upon Einstein's (1910) theoretical work on light scattering. A light beam enters a cross-shaped vessel from the left, and an observer evaluates (through a window in one arm of the cross) the state of polarization of the light scattered from molecules of gas such as H2 in the vessel. K is a calcite-glass beam-splitting prism. Bottom left: Equipment for studying the Maxwell effect (flow-induced double refraction) in fluids containing rod- or plate-shaped particles. The view is from the side, the cylinder Z is rotated at constant angular speed in the fluid container. Polarized light arriving from below is analyzed with the compensator C and the Nicol prism N2. The experiment was suggested by Kundt (1881); the illustration is from Ambronn and Frey (1926). Right: The ultramicroscope of H. Siedentopf and R. Zsigmondy. Light from the right illuminates a fluid sample in a vial underneath the microscope on the left, making particles visible as bright spots. The Nicol prism N was sometimes used, especially if information on the shape of the particles was desired. The illustration is from Zsigmondy (1925), but it may have originally appeared in a C. Zeiss advertising leaflet around 1905.

36.6 The Compton effect

A.H. Compton, some of whose research has already been referred to, surmised that according to quantum theory and the principle of conservation of energy, the wavelength of an X-ray beam ought to increase after collisions with electrons. He made an effort to verify this by measuring

such changes in a beam scattered from a piece of graphite (Fig. 36-2 bottom left). They were quite small, and it was therefore advantageous to analyze the rays with a crystal which reflected them without appreciable angular broadening. In his first paper describing his experiments, Compton (1923) says: "Using a crystal of calcite, this made possible a rather high resolving power even in the first order spectrum.". Others studying these changes in the following couple of years also used calcite crystals, such as Ross (1923), Hagen (1925) and in particular W. Duane who was already well known for his achievements in the field. Duane had originally not been able to reproduce Compton's results, and was among those who considered his ideas to be unrealistic. However, he was converted to Compton's views (Allison and Duane 1925) after repeating his experiment with improved equipment and a 9-cm long calcite crystal.

The increase in X-ray wavelengths on collisions with electrons were consequently called the Compton effect, see Appendix 5. It may be concluded from its existence that light quanta carry a linear momentum in agreement with the quantum theory of Einstein (1905a); this provided new support to that theory. With improved resolution techniques it was later attempted to split the Compton peak into discrete spectral lines (e.g. Bearden 1930), analogous to a phenomenon which had by then been found in visible light (the Raman effect, section 39.7). Bearden used for this purpose "a large very perfect calcite (Iceland spar) crystal", but without success. It was also investigated whether the Compton effect was influenced by the electrons (which caused the X-ray scattering) being more or less free or bound to atoms. Again, calcite was chosen for the task, for instance by DuMond (1933) who designed an instrument with 50 small crystals, and by Ross and Kirkpatrick (1934).

36.7 Light scattering, ozone and other topics

Lord Rayleigh had been of the opinion that the blue color of lakes and ocean water came from the sky, but not everyone agreed with him. Smoluchowski (1907) presented a hypothesis to the effect that the scattering of light in fluids and fluid mixtures might be caused by internal fluctuations in their density. This hypothesis was in agreement with the kinetic theory of thermodynamics. The fluctuations will become particularly large in the immediate vicinity of the critical temperature of a fluid, and it had in fact long been known that fluids take on an opalescing milky-white appearance when in that state. Smoluchowski's views were supported by the results of experiments by Onnes and Keesom (1908; Keesom 1911) on the absorption of light in ethylene around its critical point. Einstein (1910) improved Smoluchowski's work to the extent of predicting quantitatively the amount of scattered light, and connecting it with Rayleigh's original theory of scattering. Einstein's conclusions received additional confirmation in measurements on a water-phenol mixture by Fürth (1915). In both these experimental studies, Nicol prisms were employed in photometric measurements (Fig. 36-4).

Zernike (1916), Schirmann (1919, 1926), Gans (1920 and later), Cabannes (1922) and Raman (1922) with his collaborators were among the many scientists involved in experimental and theoretical work on scattering and absorption of light in liquids around 1920. Others investigating scattered light and its polarization included King (1913), Smoluchowski (1916), Ribaud (1919, Fig. 36-5), Rayleigh jr. (1920 and earlier), Cabannes (1921) and Gans (1921), using instruments like those shown in Fig. 36-6. Raman and Rao (1923) studied similar phenomena in gases, and Rayleigh jr. (1919) in Iceland spar and other solids. Their measurements agreed fairly well with the Smoluchowski-Einstein theory mentioned above, but the light scattered at right angles to the incoming beam was not quite linearly polarized as Einstein had predicted (Partington 1953, p. 246) except in the noble gases. This so-called depolarization of the 90°-scattered light was partly caused by the fact that the "clumps" that especially form in liquids and gases near their critical temperature, are anisotropic to a variable degree as well as being of variable density. However, the depolarization was greatest in those substances which exhibited a strong Kerr electro-optic effect (section 23). Gans, Raman, Cabannes and others showed that this furnished a means to estimate the size and shape of the molecules or other particles causing the scattering. See discussions in Partington (1953, p. 255-267) and in Stuart (1931) who states in an abstract that "Measurements of the depolarization of scattered light and investigations of the Kerr effect in molecules are important, because they permit conclusions to be drawn about their structure and mutual interactions of their constituents.". Subsequent research on scattering has for instance contributed to improved understanding of stellar radiation and of the atmospheres of planets.

Björnståhl (1918, Fig. 36-6) carried out a set of somewhat similar observations on the light absorption by liquid crystals. He demonstrated with the aid of a König-Martens spectrophotometer (Fig. 29-15) that the optical properties of this class of substances may be modified by the application of electric fields; the practical importance of this discovery has been mentioned in section 27.2 above.

If a compound of very large molecular size is dissolved in a liquid and its molecules do not aggregate, it may be deduced from the hypothesis of Einstein (1910) that the amount of light scattered by it is proportional to its molecular weight. This was first confirmed experimentally by Putzeys and Brosteaux (1935) on several proteins, and Debye (1944, 1947) pointed out that aspects like the shape of the molecules or the wavelength of the light did not have much influence on the result. This knowledge was then used to some extent around 1950 in research on the molecular weights (> 10,000 a.m.u.) of for instance albumins and globulins, polymers, antibodies, and viruses. Other methods of finding these weights were also available, such as ultracentrifuging and measurements of osmotic pressure.

Yet another application of scattered light in addition to those described above and in section 27.1, was in so-called ultramicroscopes (Fig. 36-6). In these instruments, particles in colloidal suspension in liquids or in glass (Siedentopf and Zsigmondy 1903, Cotton and Mouton 1903) could be made visible even if they were smaller than the wavelength of the light used. With illumination coming from the side, the particles (such as microorganisms) appeared as luminous specks on a darker background. Ultramicroscopes were marketed for decades by the Zeiss company and others. With an optional Nicol prism in the light path, some information could be obtained about the shape of the particles, or about the presence of fluorescence. See Appendix 5.

Measurements of the spectrum, intensity and polarization of light from the sky under varying conditions were continued, including studies on the correlation of these properties with the latitude and altitude of the observer, the seasons, meteorological factors, sun-spot activity, and major volcanic eruptions. Many of the original sources on these studies are in meteorological journals, doctoral theses and other publications which I have not consulted, but an extensive review was presented by Jensen (1928). As an example of this type of work one may mention the paper by Dember and Uibe (1918, Fig. 36-5) on the state of polarization of sky light depending on its color, using a new electrically recording instrument with a Nicol prism. A second example is represented by the long-standing detailed measurements by C. Dorno (e.g. 1919) on the visible part of the sky light. He employed in part Weber's (1891) photometer from section 29.4 above, and probably also other equipment with Iceland spar prisms. Dorno later achieved fame for his research on ultraviolet radiation in the atmosphere.

A related topic which has been of much interest in recent decades, concerns the absorption of the solar ultraviolet radiation by ozone. As described in section 29.8, A. Cornu and others had found that this radiation had almost completely disappeared at wavelengths shorter than 290-300 nm, which agreed with the properties of ozone. Cornu also concluded from his measurements at

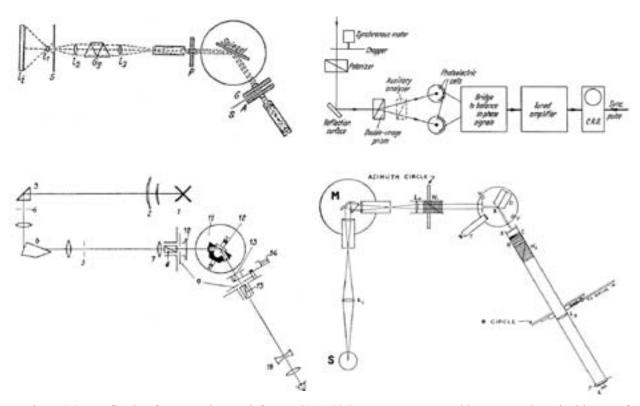


Figure 36-7. Reflection from metals. Top left: Lauch's (1924) measurements on thin vacuum-deposited layers of noble metals. Light from the lamp on the left is spectrally dispersed by the prism Gg and polarized by a Nicol prism P. After reflection from the metal, its state of polarization is analyzed with a sophisticated instrument (Leiss ca. 1910) containing a Savart plate, a Senarmont compensator and a Nicol prism. Right: Equipment similar to that of Pfestorf (1926) in research on the optical properties of several metals. In this more recent setup (Handbuch der Physik vol. 25/I, 1961), his photographic recording has been replaced by photoelectric detectors. Bottom left: Measurements by Tronstad (1933) on the surface properties of e.g. mercury. The prisms on the left select light of a narrow wavelength interval from the lamp 1. It is then polarized by the Nicol prism 8 and is reflected from the metal at 12. Information of its state of polarization is obtained with the half-shadow prism 13 and the Nicol prism 15. Right: With Weld's (1922) "crystelliptometer" which among other things contains the lamp S and two Nicol prisms, the properties of light reflected from the semiconductor surface M may be determined.

mountain sites of up to 3.7 km altitude, that when the sun was near zenith slightly more of the shortest wavelengths passed through than at sea level. However, painstaking observations by for instance Wigand (1913) who took a calcite-prism spectroscope to 9 km height in a balloon, did not reveal any significant change in the short-wave limit of the solar radiation. Therefore, the ozone layer was bound to lie even higher, and altitudes of over 40 km were deduced from further research where Iceland spar prisms still had some role (Lambert et al. 1927). More recent work has however placed most of the ozone in the range of 20-40 km. Fabry and Buisson (1921) estimated the total amount of ozone in the atmosphere, using in their study Iceland spar prisms in the calibration of photographic plates. They found that if all the ozone was brought down to sea level under standard conditions, it would form a 3-mm thick layer. This conclusion is still valid.

G.M.B. Dobson and his associates who initiated comprehensive investigations of the ozone layer around 1923, appear to have mostly made use of photo-electric sensors. However, their techniques were to a considerable extent based on the fact that thin films of silver metal are fairly transparent in a certain wavelength interval in the ultraviolet. This property which was discovered in 1883, had been studied for instance by Cornu (1889) and Minor (1902, Fig. 29-22) with the aid of Iceland spar prisms.

36.8 Luminescence; the Maxwell effect

Exposure to a brief flash of light raises the molecules of some compounds to an excited state, and for a short while thereafter they emit faint light, fluorescence. Many fluorescing substances were known by the mid-19th century, and others were discovered later. In the solid state, they have found applications e.g. in self-luminous watch dials, radar-, oscilloscope- and television screens. Substances of this kind, often dissolved in water or other liquids, include fluorescein, eosin, rhodamine and erythrosin. They may be encountered in photographic or printing materials, textiles, detergents, staining of histological specimens, and biochemical experiments. Photometers with Nicol prisms have been involved with various aspects of fluorescence, but only a few examples have been mentioned in this compilation (e.g. Becquerel 1861, Fig. 18-3, Lépine 1915, Fig. 36-5).

Fluorescence after illumination by strong polarized light is generally unpolarized, but some exceptions accumulated. The first one may have been noted by R.W. Wood in vapors of sodium, potassium and iodine in 1908. Franck and Hertz (1912) confirmed that this was the case at least to some extent, in iodine. A new phase in research on gas fluorescence began about a decade later, see section 39.3. Becquerel (1910) and Nichols and Howes (1914) investigated the polarization and other properties of fluorescence in ruby and crystals of uranium salts. Pochettino (1905, 1911) observed polarization phenomena in the luminescence produced in Iceland spar and other minerals by a current of cathode rays.

The polarization of fluorescence in liquids illuminated by polarized light was discovered by F. Weigert around 1920 (see Weigert and Käppler 1925). The phenomenon was studied in more detail e.g. by Wawilow (1925) and Hakenbeck (1929), both of whom used König-Martens spectrophotometers. The polarization decays rapidly because the molecules in the liquid are moving to and fro and colliding with each other. Such fluctuating movements of particles in a liquid were first described by the botanist R. Brown much earlier, and A. Einstein had investigated them theoretically in the case of spheres in 1905-06. Einstein however did not consider the much more complicated case when all the particles are either flattened or elongated in shape. Then their orientation will also fluctuate irregularly, but if the liquid is very viscous they have not rotated much before dropping to their normal energy level. Thus the emitted light can be polarized, for instance in the case of rhodamine B and erythrosin in glycerine. In the 1920s this motivated various studies, such as measurements of the decay times of the fluorescence polarization (with Kerr cells and other equipment) in liquids at different concentrations of the fluorescent substance, different temperatures and viscosities. Extensive theoretical analyses of the Brownian rotational motion also appeared (Perrin 1925, 1929 and later, Lewschin 1925, Hakenbeck 1929). For a while the measurements seemed to indicate that the fluorescence did not start immediately at the instant of the light flash; such a "dead time" would have posed an interesting problem to quantum theory, but Gaviola (1929) blamed that result on experimental errors.

In the early 1950s scientists began investigating the decay of polarization in the fluorescence of small molecules that had been attached to much larger ones like proteins. This was done in order to obtain information on the sizes and shapes of the latter (Weber 1952); a further development of the technique is mentioned early in section 39.2. In a recent book (Sun 2004) this fluorescence depolarization is said to be one of two classical methods of studying that aspect of diffusion in fluids which is related to molecular rotation. The other classical method according to Sun, is the Maxwell effect from section 27.3, see Fig. 36-6. This effect has provided much useful information on for instance the shape and refractive properties of particles in colloids (cf. Zocher 1925) and of large molecules (Vorländer and Walter 1925), see also Piganiol (1947) and Partington (1953,

p. 274-276). In this way Muralt and Edsall (1930) studied myosin from muscles, and Lauffer and Stanley (1938) used the Maxwell effect to demonstrate that certain viruses infecting tobacco plants are rod-shaped. The latter experiment is quoted in Stanley's lecture when he received the 1946 Nobel prize in chemistry. Around 1940 and later, important research was also carried out on the size and shape of DNA (Signer et al. 1938) and polymers with this technique.

36.9 Metal and semiconductor surfaces, and thin films

Of the various publications in the mid-19th century which dealt with the state of polarization and the relative intensity of light reflected from polished metal surfaces, those by Cauchy (1839b), Jamin (1845, 1847) and Quincke (1863) have been mentioned above. Around 1890 interest in the topic was revived, probably as a result of the confirmation of J.C. Maxwell's electromagnetic theory of light. According to the theory, the refractive index of conducting materials can be written as a complex number. Experiments involved not only metal surfaces, but also thin films of metals, semiconductor surfaces, and later thin insulating films. P. Drude (e.g. 1891, 1900a,b) may be the best-known participant in this field, and contributions were also made by A. Cornu, A. Kundt, W. Wernicke (e.g. 1885), J. Conroy, G. Meslin, H. du Bois, O. Wiener, H. Rubens, W. Voigt, and H. v. Wartenberg (1910). These and further experiments employing polarized light no doubt played a role in the formulation of theoretical works on the causes of electrical conduction, by Drude (1904, chapter XXXVI of Winkelmann 1906) and subsequently by Lorentz (1915) and others such as Kent (1919, on metals). This research also may have accelerated development of the reflected-light microscopical techniques described in section 29.3, see for instance Königsberger (1908) and Tyndall (1923).

New methods of producing thin films of metals were developed, including vacuum deposition on glass (e.g. Lauch 1924, Fig. 36-7). The analysis of polarization states in visible and ultraviolet light after reflection from metal surfaces and metallic films which was mentioned in section 29.8, was continued by Pogány (1916), Hauschild (1920), Weld (1922, Fig. 36-7) and Pfestorf (1926, Fig. 36-7), among others. It also laid the foundations for an important method (Tronstad 1933, and many others) which has been called "ellipsometry", Figs. 29-4 and 36-7. Since around 1960 it has been applied extensively in the non-destructive measurement of the thickness and physical properties of thin and very thin films used in optics and electronics. See for instance the compilation of reprinted classic papers edited by Azzam (1991). Such films are essential in the production of integrated circuits and microprocessors for computers, and Collett (1993) claims that ellipsometry is "one of the most important applications of polarized light"; according to a paper in the Physics Today magazine of May 2009, it speeded up the appearance of modern computer technology by at least 15 years. Research on reflected elliptically polarized light also greatly improved understanding of the films forming on metal surfaces due to the action of electrolytes or of weathering (Tronstad 1929, 1932). The method could be applied to films whose thickness was less than a tenth of that which other known methods were able to resolve.

37 Iceland spar production 1910-40

37.1 The Helgustadir quarry

Matters related to Iceland spar were somewhat prominent in Iceland politics in 1910-11, as may be seen from the Reykjavík newspapers, but these matters will not be covered in any detail here. A parliamentary committee of inquiry was set up in 1911 to scrutinize actions of past Minister Björn Jónsson, because of five points of alleged improprieties; one of these had to do with affairs of the Helgustadir quarry. There are references to the committee's findings in the records of Parliamentary debates for 1911, division A p. X, with supplementary documents nos. 195 to 239 which I have looked through briefly. The Iceland spar issue is also referred to in memoirs by Sveinn Björnsson (son of the above, and later president of the Republic of Iceland), published in 1957. What happened was that the Government had in 1910 put out a tender for the operation of the quarry, after the third 5-year term of the Tulinius companies ran out. Two bids were received, one from Thor E. Tulinius. The other one, which was higher, came from a Reykjavik businessman by the name of G. Jakobsson. The Government made a 10-year contract with Jakobsson, who was soon joined by another businessman, M.Th.S. Blöndal. According to memoirs of the civil servant J. Krabbe (published in 1959) and supplementary documents no. 218-219 of the above collection, the Zeiss Co. sought unsuccessfully to lease the quarry from them. Instead, the authorities permitted Jakobsson and Blöndal to transfer the quarry contract to a French bank which in turn transferred it to a French limited-liability company named Société Française d'Entreprises en Islande. This outfit had been set up by J.P. Brillouin, the French consul-general in Reykjavik.

According to a book by Thorarensen (1966), Thor E. Tulinius sold half of the stockpile remaining from the 1895-1910 mining operations for 75,000 kr, and turned the remaining half (stored at Eskifjördur and in Copenhagen) over to the authorities. However, the original agreement between the parties would have required Tulinius to first sell the whole stockpile and then keep half of the proceeds; the lawfulness of Tulinius' actions was debated, and some critics also voiced suspicions as to whether the half he sold had been of better quality than the one delivered to the state. The Government subsequently had to pay Jakobsson and Blöndal a considerable sum for selling (to Mr. Brillouin cf. Supplementary document 224) its half of the stockpile. This half amounted to about 7 tons according to the newspaper Lögretta on 15 Nov. 1911. 4500 pounds (2250 kg) were also recovered by them from the quarry in 1910. The scientific advisor of Brillouin was named in the Reykjavik press as being a Mr. Jobin, presumably from the optical-equipment workshop of that name in Paris.

In Supplementary documents 202-207 it appears that the Thor E. Tulinius' sale of materials from his part of the Iceland spar stockpile naturally reduced the demand for further supplies.

This was a disappointment to Brillouin, and it may have been a chief cause for his company not working the quarry in 1911. They did operate there in 1912, and the state received 4000 kr. in fees according to Parliamentary records for 1915 (division A, p. 37). Eiriksson (1930) states that the French recovered a total of two tons, presumably in 1912. No quarrying took place in 1913 but the State received 6114 Ffr. from the sales of Iceland spar, cf. the same records. Balance sheets for 1912 and 1913 were provided (cf. Parliamentary records, division A for 1915, p. 1735).

I have searched in Icelandic newspapers for evidence of quarrying operations in 1914, but found none. S.H. Kvaran, a medical doctor and M.P. in the area, was in that year appointed as official inspector of the Helgustadir site instead of P. Torfason. The World War prevented any further exploitation of the quarry for a while, and Brillouin's group never showed up again. Some documents relating to the quarry in the period are in storage at the National Archive but its staff inform me that these will be hard to find there. A private M.P.'s bill proposing in 1917 sale of the Helgustadir farm (without the mining rights) to the sitting tenant was not ratified by Parliament.

With Parliament's consent, the government subsequently decided to operate the Helgustadir quarry on its own, and a recent mining engineering graduate, H.H. Eiríksson, was hired in 1920 to direct activities. See Eiríksson (1920, 1922). A report by him, referred to in series A2 of the Parliamentary records for 1921, p. 1686, seems to have been lost from the archive of the Parliament. Several relevant documents including a diary of operations at Helgustadir, were found by G. Jensson in the Reykjavik Trades College (where Eiríksson was headmaster until 1954) around 1997 and delivered to the National Archive but I have not inspected these. Some details of the site operations are published in Parliamentary debates, in Eiríksson's biography (Hagalín 1970) and in a paper by him (Eiríksson 1943). His workmen made a tunnel of about 70 m length into the hillside below the quarry (Fig. 37-1), recovering there some tons of good crystals (Hagalín 1970) which were exported. Curiously, another published source (Jónsson 1988) claims that no crystals were found in the tunnel and that all the recovered material came from the quarry itself. There certainly was some activity there, as one worker was killed by a rock-fall in late October of 1923; H. Hjelm (pers. comm. 2005) informed me that this happened during a flood in the Silfurlækur brook due to heavy rain. In the book by Kuhn and Prinz (2003) a photo dated in July 1924 shows six people at work in the quarry.

Parliament approved in 1921 that an Icelandic geologist named S. Emilsson be sent at its expense to the Zeiss works in Jena to learn techniques in cleaving and sorting Iceland spar crystals. See e.g. Emilsson's obituary published in the journal Náttúrufraedingurinn in 1976. He did complete this training, but his skills do not seem to have been put to any use after he returned to Iceland. In 1922 Parliament passed an Act to the effect that the State had a monopoly on selling all Iceland spar recovered in the country; this legislation was annulled in 1933. In fiscal records, some income from sales of Iceland spar is listed on a few occasions, for instance 5300 kr. in 1929 which was of a similar order of magnitude as the annual wages of a civil engineer.

Hagalín (1970) quotes Eiríksson as having learned in 1924 that the major manufacturers of polarized-light equipment had found a less expensive and more practical material to replace Iceland spar. Furthermore, British Customs authorities were by then also supposed to have discontinued the use of Iceland spar in instruments for taxing sugar products. It now seems clear that these accounts by Hagalín are quite inaccurate. At least, no alternative to Iceland spar had been discovered, but better and/or cheaper supplies may have appeared on the market cf. the following sections. It is ironic, that while Eiríksson was closing down the Helgustadir quarry for these supposed reasons, Bouasse (1925, p. 2) wrote that large Iceland spar crystals are worth their weight in gold!

According to file indexes in the National Archive, documents from the local sheriff's office relating to affairs of the Helgustadir quarry in 1925-36 are somewhere in their vaults but archivists again have not had time to look for them. Eiríksson (1943) states that the quarry was leased to a

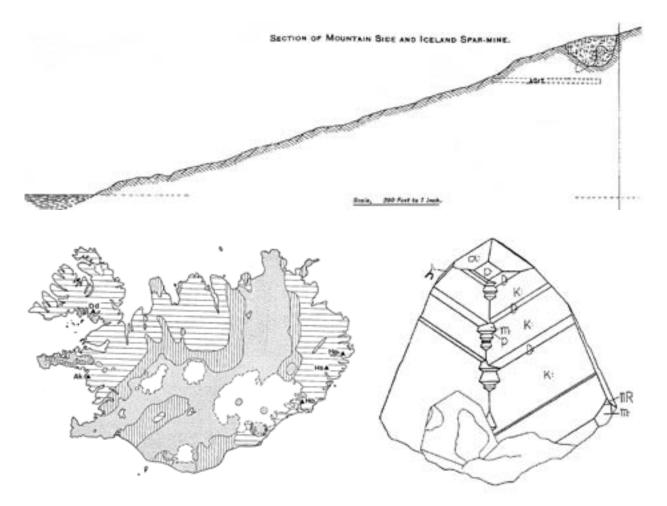


Figure 37-1. Top: Sketch by H.H. Eiriksson (1920) of the slope up to the Helgustadir quarry, including the tunnel to be dug underneath it. Bottom left: Iceland, with horizontal lines indicating basalt areas of Tertiary age. From Th. Einarsson's textbook of geology, 3rd ed., 1978. The quarries at Helgustadir (He) and Hoffell (Ho) are indicated, and also three other locations where some amounts of impure calcite have been noted. These are Djupidalur (Dd), Akrar (Ak) and most recently Hoskuldsstadasel (Hs). Right: An Iceland spar crystal from Helgustadir, described by Kaspar (1940).

K. Magnússon from Reykjavík in 1935; a track for carts or lorries up to the site may have been constructed at that time. Another gentleman, E. Sigurbergsson, leased it for a period from 1939. Presumably they were mostly collecting waste spar from the tailings of the quarry, and crushing it for use in a surface finish on buildings (along with crushed obsidian, flint, jasper, marble and even seashells). This method of coating buildings was first used in 1933 on the National Theatre (Gudmundsson 2003), and numerous other official buildings, private houses, garden walls etc. were coated in this way at least into the late 1950s. Some experimentation in setting Iceland spar crystals into exterior or interior concrete surfaces also took place, the earliest example probably being the Catholic Church in Reykjavik around 1928.

Fig. 37-2 shows a timeline for events at the Helgustadir quarry in 1780-1930, with geologists and other occasional visitors on the left and operators on the right. It may be added here that a Czechoslovakian expedition in the late 1930s published a report on the quarry, also describing some crystallographic properties of samples from there (Kaspar 1940, see Fig. 37-1).

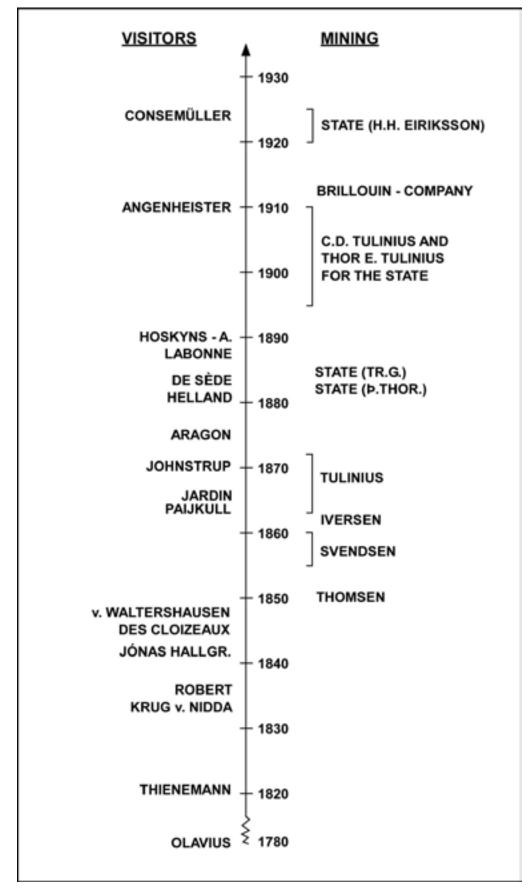


Figure 37-2. A timeline of mining activities at Helgustadir is on the right-hand side. The State owned $\frac{1}{4}$ of the farm and mining rights to 1879; since then, it has been the sole owner. The left-hand side gives the names of some travellers who visited the site in the period 1780-1930, or were otherwise interested in it.

37.2 Hoffell and other potential spar localities

G. J. Hoffell (1946, p. 39-44) has written an account of the discovery of a site yielding good Iceland spar crystals, in late 1910. The site was at 500 m altitude on a mountainside not far from his farm Hoffell in Hornafjördur fjord, Southeastern Iceland. See a map of the locality and some others in Fig. 37-1. A quantity of spar was recovered there in the following years and exported through the initiative of B. Kristjansson, a merchant and M.P. in Reykjavik. See also Arason (2004). In a Parliamentary debate of 1922, Kristjansson however does not speak very highly of the site, and neither does H.H. Eiriksson who inspected it in 1923 (Hagalín 1970, p. 172). Access to both the site itself and transportation facilities was much more difficult than at Helgustadir. The exporting of spar from Hoffell was discontinued during WW I but then resumed (from 1921 according to Eiríksson 1943), when T.E. Tulinius leased the mining rights and worked the site for three seasons. Eiríksson (1930) states that there has been no mining activity at Hoffell since 1925. According to Gudmundsson (2003) the calcite for the National Theatre exterior coating came from Hoffell around 1933, and Arason (2004) says that it is also a part of decorations inside the theatre foyer. Hoffell (1946) and other sources state that Iceland spar and aragonite were last mined at the Hoffell site in 1938-39, in order to provide materials for the main building of the University of Iceland. On the exterior of this building, spar crystals are enclosed in large whitish tiles around some windows and above the main entrance. Inside, there is a large number of Iceland spar rhombs in a small cupola above the entrance, and on the altar of a chapel used by the Faculty of Divinity.

As mentioned in chapter 24, T. Thoroddsen (1889, 1890, and other publications) considered Djupidalur on the south coast of the northwestern peninsula to be a potential spar-mining site. Occasional attempts at prospecting for optical-quality calcite were carried out here in the early 20th century (see e.g. Hagalín 1970, p. 233). According to a newspaper account in 1951 the material was too yellowish for that purpose, but some calcite was collected there in the 1940s for use in masonry. In his writings, T. Thoroddsen suggests that the Gvendarnes promontory in Fáskruds-fjördur (south of Reydarfjördur) may contain some calcite, and also the farm Akrar in central western Iceland (Fig. 37-1) already referred to in chapter 32. However, neither seems to have ever produced any optically useful crystals.

37.3 Iceland spar mining abroad, 1910-40

In the U.S. state of Montana, a minor quantity of Iceland spar was recovered before 1918, and some 600 lbs. were exported (Parsons 1918). This mining does not seem to have continued after 1918, even though Parsons considers the Montana material to have been "the nearest approach to Iceland spar yet uncovered anywhere in any part of the world". In a report of the U.S. Geological Survey, Mineral Resources of the United States 1918, this locality is said to be the only place outside Iceland where Iceland spar is being mined. Tschirwinsky (1910) mentions a new spar locality in the Crimean peninsula, but the only other reference to it in the following decades that I am aware of is in Dammer and Tietze (1913, p. 387), who say that "the Crimea has occasionally provided some quantities of clear calcite useful for technical purposes". Hughes (1937) indicates that small amounts of Iceland spar from Montana and California were used in U.S.-made optical instruments in WW I. I have also seen crystals from these places mentioned in the context of X-ray spectroscopy. However, H. Schulz (in Wien-Harms 1928, see Appendix 1A) claims that most of the U.S. production was of poor quality. There do exist U.S. reports on the supply of Iceland spar after the war (Bowles 1920, Hughes 1931) which I have not managed to get hold of. Lowry

(1943) and Wright (1950) state that a deposit at Cedarville, California, was worked in 1920-24 until depleted. Wright estimates that the annual world consumption of Iceland spar before WW II was always less than 500 lbs.

In a publication of the South African Geological Survey (1936) the following appears:

"In the Union of South Africa, Iceland spar has been found in deposits sporadically distributed over a large area in the north-western Cape Province including portions of the divisions of Kenhardt, Calvinia, and Namaqualand...nearly all the Iceland spar produced has come from scattered deposits on the farms...situated some 50 to 70 miles southwest of Kenhardt.

No accurate statistics of the Iceland spar recovered from this area are available as the output from the various deposits has been small and the production intermittent; moreover most of it has been exported in small quantities by post and and no official control on the output has been exercised. It has been estimated, however, that since exploitation first began possibly 20 years ago, between 1,500 and 2,000 lbs. of Iceland spar have been exported from the Union.

...the Iceland spar is found in association with a good deal of worthless calcite. It occurs in the form of veins, lenses and isolated crystal aggregates of irregular shapes and sizes embedded in soft decomposed doleritic rocks, which occur abundantly in that area as dykes and sills...The Iceland spar in the Kenhardt area is recovered from shallow open workings none of which are deeper than 70 feet in the decomposed doleritic rocks.".

According to H. H. Hughes (1937; see also Kelley 1940) the Kenhardt area replaced Iceland as the main source of Iceland spar for optical instruments soon after 1920. On p. 497 of the 4th edition of the above-mentioned South African handbook (published in 1959; L. Hughes, pers. comm. 2000) as well as in its 5th edition (Coetzee 1976) it is confirmed that this was the case up to WW II. After 1944 Polaroid sheets "virtually eliminated commercial interest in Iceland spar" so that no further production took place from the South African sites before the late 1960s. In some U.S. newspapers in 1939 there appeared a news item from Windhoek in Southwest Africa (at that time administered by South Africa, but now called Namibia). It stated that Iceland spar was found in the region in 1926 and that organized mining for export was being established; the recovery methods up to then appear to have been rather crude (cf. Appendix on SW-Africa in South African Geological Survey 1936).

I have received information on a site discovered around 1912 in a gold mine near the town of Tabulam in New South Wales, Australia (G. Smith 1926, referred to in a personal communication from T. Krassmann 2005). At this site large calcite crystals, partly transparent Iceland spar, occupied a cave of $70 \times 16 \times 9$ feet in size. In about 1924 an employee of the Zeiss company (probably R. Straubel) arrived and hired local people to break the crystals apart and pack them in crates for shipment to Germany.

Good-quality spar arrived on the market from Spain "within the past few years" (Hughes 1937), but the quantity was small according to Lowry (1943). Later, supplies may have come from Siberia and/or Japan (Eiríksson 1943, and a chapter by T. Tryggvason in a book on the natural history of Iceland published in 1961). Coetzee (1976) also mentions spar sites in e.g. Cornwall, the Harz mountains in Germany, Mexico, and Canada.

38 The Faraday effect and Kerr cells, mostly in 1920-40

38.1 Research on very rapid processes

Let a beam of ordinary light pass through a polarizing Nicol prism and a glass vessel of several cm length containing nitrobenzene or CS_2 between capacitor plates (a Kerr cell, see chapter 23) tilted at 45° angle to the plane of polarization. If the beam then encounters a second Nicol prism set at right angles to the first one, no light is transmitted by this arrangement. When however a high voltage is applied to the plates, so that the fluid becomes doubly refracting to a significant extent, the Kerr cell acts like a phase compensator (section 13.5) and a part of the light can pass through the second Nicol prism. The apparatus therefore constitutes a voltage-controlled light switch. Pauthenier (1920) and others showed that it could modulate light at frequencies up to at least a thousand times higher than mechanical devices (such as slotted metal plates or polygonal mirrors, rotated by electrical motors). According to Bennett and Bennett (1978) these frequencies can even be increased to more than 1 GHz with suitable electronic circuitry.

Kerr cells were used up to 1940 or so in research on very rapid processes and short-lived phenomena, including the duration of fluorescence in dissolved dyes (1-10 ns; Gottling 1923, Gaviola 1926, 1929, cf. Bruhat 1942, p. 738) and lifetimes of excited states in gas molecules (Hupfeld 1929, Griffiths 1934). Brown and Beams (1925, Fig. 38-1), Lawrence and Dunnington (1930), Hámos (1930) and others employed Kerr cells in studies on the formation of electrical sparks.

Research on the duration of continuous light pulses (i.e. without phase jumps) was mentioned in section 13.1 above. This question of the "length" of a photon became still more interesting after the advent of quantum theory. Thus, it was attempted to measure the time it took light to eject electrons from a metal surface (Lawrence and Beams 1927) but the results were probably not significant. E. Rupp (1928) investigated whether the spectrum of a monochromatic light beam which was chopped into short segments by a Kerr cell was altered in a particular way. This was in continuation of experiments that he had done in collaboration with A. Einstein to test certain aspects of quantum theory. Later however, it has become evident that an account by Rupp in 1926 on the former experiments was mostly fictive (see the compilation by A.P. French in the journal Physics in Perspective 1, 3-21, 1999), and so were papers published by him after 1930. Stauffer (1930) in fact pointed out deficiencies in Rupp's (1928) procedures and improved on his experiments (Fig. 38-2, top) but did not obtain any unexpected results regarding the spectrum of the chopped beam. A. Righi (1883) had much earlier made spectral analyses of light transmitted through rotating Nicol prisms, and such frequency modulation of light and other electromagnetic radiation became technically important in the late 20th century.

I. W. Beams and F. Allison (e.g. 1927) measured the time that it took the Faraday effect to become

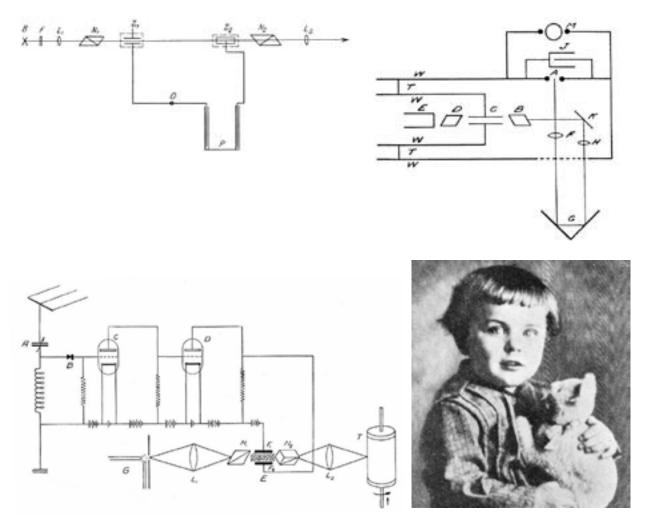


Figure 38-1. Applications of the Kerr electro-optic effect. Top left: Gutton's (1911a) comparison of the speed of light in air (horizontally at the top) and as an electromagnetic wave along wires. O is an oscillator, P is a slider for changing the length of the wires. Z1, Z2 are Kerr cells and N1, N2 are Nicol prisms (Wien-Harms 1928). Right: The setup by Brown and Beams (1925) to study the development of an electrical spark across the gap A. G is a mirror which can be moved farther away to delay the light that is analyzed by the spectrometer E. B and D are Nicol prisms, C is a Kerr cell. Bottom left: A Telefunken AG receiver circuit for the wireless transmission of pictures. A signal from the aerial on the left controls the voltage applied to the Kerr cell at E, varying the intensity of a very small spot of light from the arc A which reaches the photographic paper on the slowly rotating drum T. Right: A photograph sent in one minute using this technique. The illustrations are taken from a Danish book containing popular accounts of various inventions and discoveries: Teknikens Vidunderland, ed. V. Marstrand, 1928. A similar circuit drawing may be found in U.S. patent no. 1,730,772 granted to A. Karolus in 1929.

established in fluids upon the sudden application of a magnetic field. With his collaborators, Allison later developed from this a method of chemical analysis, which was also supposed to indicate what isotopes of each element were present in a fluid. One result announced by him around 1931 was that a heavy isotope of hydrogen existed. This prompted H.C. Urey to initiate an extensive research project to isolate this isotope and study its properties, see e.g. Urey (1934). The project yielded important results, for which Urey was awarded the Nobel prize in 1934. In that year it was also demonstrated (Phys. Rev. 47, 310-315 and 546-548, cf. a lecture by I. Langmuir reproduced in Physics Today, Oct. issue 1989, p. 36-48) that all the magneto-optic work of the Beams and Allison group was based on instrumental artifacts and its results were therefore totally meaningless !

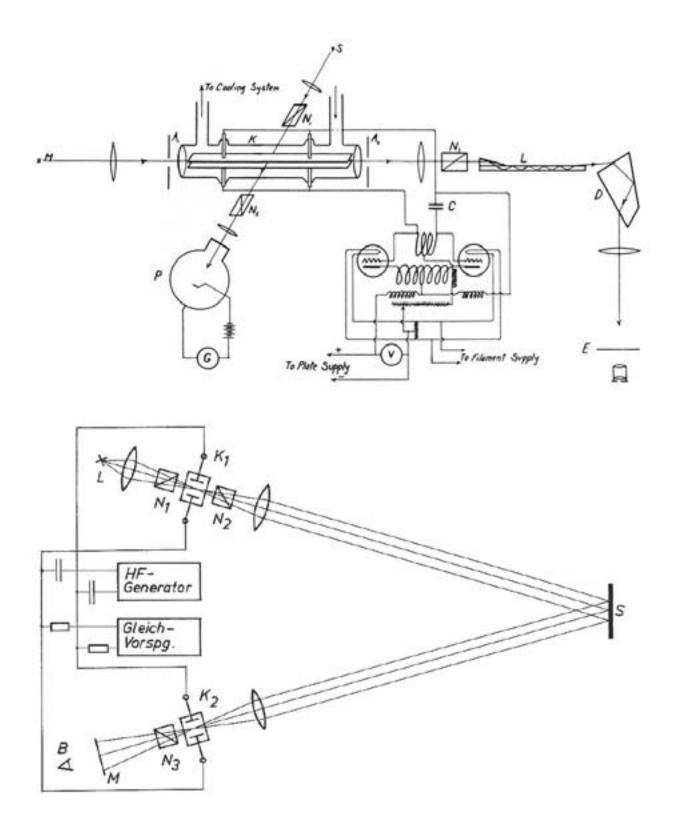


Figure 38-2. More applications of the Kerr effect. Top: Stauffer (1930) investigated how the shape of the 546.1 nm green spectral line in a light beam from the mercury lamp on the left-hand side is changed when chopped into segments by the Kerr cell K. The spectrum is dispersed by the Lummer-Gehrcke plate L whose resolving power is increased by the Nicol prism N3. Bottom: Measurement of the speed of light, made by Karolus and Mittelstaedt (1928). They employed the Kerr cells K1 and K2 whose voltages are governed by a stable MHz oscillator circuit, to measure the travel time to and from the mirror S (about 41 m each way). This replaces the toothed wheel in the 1849 measurement by H. Fizeau. Anderson (1937) and Hüttel (1940) used a similar technique, with improvements such as a photocell detector and better frequency control.

38.2 The Faraday effect and Kerr cells in communications technology

A short paper by W. Lucas (1882) was among the first appearances in print where the possibility of transmitting pictures by wire was discussed. Like many later suggestions, Lucas' idea was based on the fact that the electrical conductivity of selenium metal depends on its illumination. He envisaged varying the light intensity with a pair of Nicol prisms, one rotating relative to the other as in the sections 18.4 and 29.4 above.

Two years later a young man, P. Nipkow, was granted German patent no. 30105 on an "electrical telescope" to send a pattern of light dots in sequence between distant locations. A key element of his invention was a pair of rotating discs with a number of small perforations arranged in a spiral. The total transmission time had to be so short (0.1 second) that the dots appeared as a picture. The receiving apparatus contained Nicol prisms, and the light intensity passing through them was to be controlled by means of the Faraday effect in heavy glass or CS_2 . Sutton (1890) proposed a similar system as that of Nipkow, however employing the Kerr effect instead of the Faraday effect in his receiver.

These devices probably were not actually constructed at that time; the first one acknowledged to have been demonstrated with success was introduced by G. Rignoux (1914) in 1909. He transmitted pictures of four capital letters. At the receiving end the illumination was controlled in the same way as in Nipkow's patent, i.e. through the use of the Faraday effect in CS₂.

Nipkow put his invention away until he retired in 1919; in the meantime it had become feasible to send information by wireless. After he had improved his synchronized spiral-hole discs, his new patent on these (issued in 1924) was purchased by the firm Siemens & Halske in 1930. The German Biographic Encyclopedia states that A. Karolus had constructed a television receiver based on a Kerr cell in 1924, which indicates that it was more practical than the Faraday-effect arrangement. In the years to follow, Karolus patented several pieces of equipment containing Kerr cells, for use in communications and other fields.

In some of the first attempts at sending television pictures to be projected on large screens (by J. L. Baird in 1931-32 and by Fernseh AG in 1933 or 1934), Kerr cells were also employed. See Wright (1932), news items in The Times 6 Jan. 1931, and an account by R. Barthélemy in Revue Scientifique, 9 Dec. 1933. Scheduled transmissions began in Berlin in the Spring of 1935 and 15 screens were installed for public viewing, but the transmitting station was destroyed in a fire in the late summer. Baird seems to have made use of Kerr cells for television displays until 1937, cf. a news item in The Times 7 Jan. However, cathode-ray tubes which were being developed rapidly in the 1930s, soon replaced both the Nipkow discs and the Kerr cells in television technology.

Around 1926, the above-mentioned A. Karolus and his associates at the Siemens and the Telefunken companies had began using an improved type of Kerr cell to make copies of documents that had been scanned with photocells in distant locations and transmitted by telephone or wireless. This was quite analogous to the more modern fax technology; however, it was not at all a new concept as other methods had been in use for this purpose since 1907 (cf. a paper by A. Korn in Die Naturwissenschaften in 1925). Following extensive tests, a commercial service was inaugurated at the end of 1927 (according to The Times) to transmit photographs, bank documents, fingerprints of criminals, engineering drawings and the like between Vienna and Berlin. See the lower part of Fig. 38-1, published in 1928. A news item on similar picture transmissions between Britain and the U.S. appeared in The Times 4 Feb. 1929, and E.V. Appleton (1929) described technical improvements in this service later that year. Incidentally, Appleton was at that time investigating how radio waves were propagated through the earth's ionosphere or were reflected from it. He discovered among other things that the ionosphere acts on these waves as a doubly refracting substance, due to effects of the geomagnetic field. For his researches, Appleton was awarded the Nobel prize in physics in 1947. Kerr cells continued to be used in receivers for transmitted pictures at least until the mid-1930s (Küpfmüller 1936).

With the aid of Kerr cells, it is also a simple matter to use amplified sound oscillations to modulate a light wave. Tests of the application of this method to telephone communications were made around 1930 (e.g. West and Jones 1951) but it proved impractical as lasers and fiber optics had not yet arrived on that scene. The same technique was also being tested in recording a sound track onto movie films in the 1920s. I have not found out whether it was used on the very first talkies (beginning with The Jazz Singer in the U.S. in 1927), but Gibson (1929) considers U.S. talkie production still to be in its early stages while a company in Germany is already producing good-quality movies where a Kerr cell controls the sound. The method was in use into the 1930s according to Pauthenier (1932), Waetzmann (1932, Ch. 12) and Bruhat (1942, p. 518), and major firms (such as Westinghouse) on both sides of the Atlantic were taking out many related patents. However, the fluid most advantageous for Kerr cells is nitro-benzene which is both quite poisonous and sensitive to impurities which accumulate in it during use. Around 1950, attempts were initiated to employ the Pockels effect (see section 39.6) in ammonium phosphate crystals to make movie sound tracks (Carpenter 1953) and to replace Kerr cells in various other applications.

38.3 Kerr cells and the speed of light

The first suggestions to use the Kerr effect in light-speed measurements appeared in 1899, and Brunhes (1900) showed with the aid of a CS₂ Kerr cell that the speed of X-rays was of similar order of magnitude as that of light. Brace (1905a) proposed an experiment involving Kerr cells, to estimate the earth's speed relative to the aether. Gutton (1911a, Fig. 38-1) compared the speed of light in air with that of electromagnetic waves propagating along wires, and found a difference of less than 1%. Gutton (1911b) also used Kerr cells to confirm certain theories regarding the socalled group velocity of light waves in dispersive media. Around 1925, A. Karolus of the last section, Gaviola (1926) and others discussed the possibility of measuring the speed of light directly with this technique. Results of such determinations were published by Karolus and Mittelstaedt (1928) and by Mittelstaedt (1929). Here, a Kerr cell activated by high-frequency high-voltage oscillations (3-7 MHz, 6000 V) was in the role of a light gate, and a calibrated vacuum-tube oscillator served as a clock to measure the time it took a light beam to travel a known distance of some 330 m (i.e. four times back and forth between mirrors in the lower part of Fig. 38-2). Later, speed-of-light measurements were made with Kerr cells by Anderson (1937) and Hüttel (1940). In their work, Nicol prisms were employed, but in determinations published by Anderson in 1941 and by E. Bergstrand in 1950 (see West and Jones 1951, Jenkins and White 1957) the prisms were replaced by Polaroid polarizing sheets.

Whatever the reason, the Kerr-cell determinations of light speed performed in 1928-41 yielded values that were too high by 15 km/s or more. Measurements made by A.A. Michelson in 1924-27 on a 35-km path in air with a rotating-mirror octagon turned out to give better results, although this may have been due to chance (cf. Birge 1941). By 1948 however, the light speed was known so accurately that it was possible to turn the problem on its head: very stable high-frequency oscillators and Kerr cells were used for distance measurements in geodetic surveys during the next couple of decades (Rinner and Benz 1966).

39 On new applications of Iceland spar from around 1930

After the early 1920s, there is a decreasing probability that Nicol prisms or other components in new instruments and experiments are made of Iceland spar from the Helgustadir or Hoffell sites. In some cases however, suppliers or equipment producers may have been using up stocks originating from Iceland. Also, scientists who needed a prism or two for temporary use, are likely to have borrowed older prisms from others (e.g. Rayleigh jr. 1921) or removed them from decommissioned equipment. The following sections are meant to give an impression of various selected fields of science and technology where Nicol prisms played a part from the 1920s onwards.

39.1 The production of instruments containing Nicol prims from around 1920

New models of Iceland spar polarizing prisms which were introduced in 1928-60 include the half-shade prism of Schönrock (1928, p. 750), a small prism by Cotton (1931) of the Dove type, Nicol prisms with a "roof" (Dowell 1931), an asymmetric calcite/glass Rochon prism (Hardy 1935), Foster's (1938) prism for reflected-light microscopy, the Glan-Taylor prism (Archard and Taylor 1948), the prism of Bouhet and Lafont (1949), the Marple-Hess prism from around 1960, a double Frank-Ritter prism, and so on. These developments continued later, as may be seen e.g. from Bennett and Bennett (1978). For novelties in phase compensators, see Jerrard (1948).

Polarimeters of the Wild type were for a long time sold by J.G. Hoffman in Paris, Hermann & Pfister in Berne, H. Heele and Schmidt & Haensch in Berlin, but production had ceased by around 1920. This was because of their insufficient accuracy (Browne 1912, Schulz and Gleichen 1919, Kessler 1926) and eye strain that they caused for the observer (cf. Cornu 1870). Among European firms producing polarimeters after 1920 were A. Hilger and Bellingham & Stanley in London, C. Reichert in Vienna, Schmidt & Haensch and C.P. Goerz in Berlin, J. & J. Fric in Prague, Ph. Pellin and Jobin & Yvon in Paris, and R. Winkel in Göttingen (according to Tutton 1922, p. 1022, Struers 1925, Bruhat 1930, Bates et al. 1942, and manufacturers' catalogs). In the U.S., Bausch & Lomb in Rochester, N.Y. and the Gaertner Scientific Corp. in Chicago were probably the main producers around 1940. Typical instruments are illustrated in Figs. 39-1 and 39-2. After 1960 the polarizers and analyzers in less expensive models were probably made from improved types of Polaroid sheets, but Houben-Weyl (1955) and Moenke and Moenke-Blankenburg (1965) describe high-quality polarimeters and spectro-polarimeters with Iceland spar prisms from manufacturers in several countries.

Among important new research topics in the field of optical activity were measurements of ultraviolet rotatory dispersion (mostly in organic compounds), which a few groups of scientists

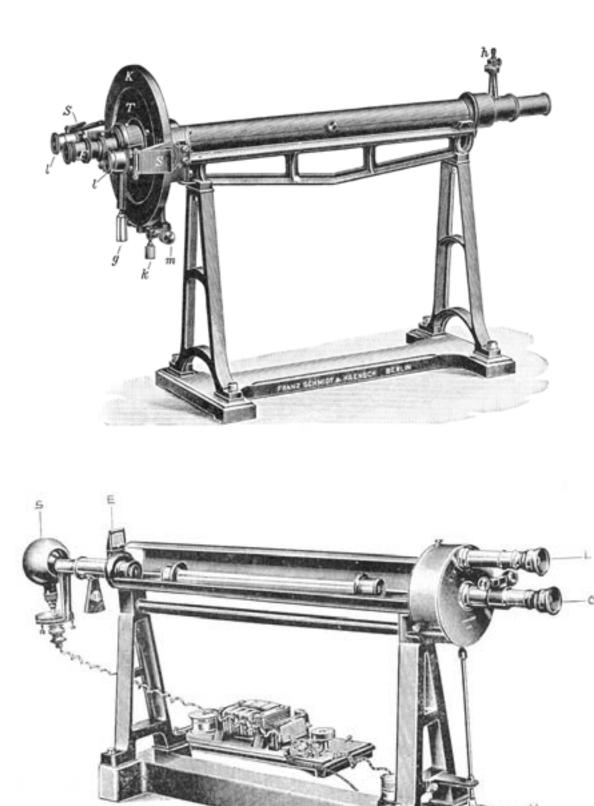


Figure 39-1. Top: A polarimeter from Schmidt & Haensch, with Lippich's half-shade prism (from Struers 1925). Bottom: A saccharimeter, probably of French origin. S is an electrical lamp. The observer turns the knob H until the two parts of a half-shade field seen through O are of equal brightness, then reads a scale with the eyepiece L (Bruhat 1942).

had initiated around 1930 (Lowry and Vernon 1928, Descamps 1930, Kuhn and Braun 1930; see Lowry 1935, chapter XVII). This technique later became valuable in investigations of the structure of proteins and nucleic acids. Optical activity was even being measured in those parts of the ultraviolet and infrared spectrum where calcite is not transparent; there, new polarizing prisms made from materials like quartz, titanium oxide (rutile) and magnesium fluoride appeared, as well as new polarizing mirrors. Research on the Cotton effect (section 22.3) for instance aided in improving understanding of the photosynthesis process, as chlorophyll is one of the substances exhibiting this effect (see Mathieu 1956). Measurements of the Cotton effect and of optical rotatory dispersion were for a long time too time-consuming for routine application. This situation changed in 1953 with the appearance of automatically recording spectropolarimeters from the firm of H. Rudolph and Sons (see Douglas 1994). As an example, these instruments played an important role in investigations of the structure of steroids. "Currently, no other physical tool can approach rotatory dispersion in its utility for settling stereochemical problems in the steroid field" says Djerassi (1960, p. 51), a modern pioneer in that field. Around 1960, new automatically recording spectropolarimeters used the Faraday effect (instead of quartz wedges or a rotatable analyzing prism) to measure the rotation of the plane of polarization of light in a fluid sample.

During the first decade or two after the appearance of Polaroid sheets, their optical properties did not warrant their use in petrographic microscopes for detailed inspection and measurements. Cotton and Manigault (1946) even prefer using glass mirrors as both polarizers and analyzers in these microscopes. In the 1940s, interest in the use of polarizing microscopes in biology was also increasing (Vickers 1956), and here Polaroid was also inadequate for demanding work. With improved types of the polarizing sheets, Waesche (1960) considers them to have "virtually eliminated demand for optical calcite", although some still prefer Nicol prisms in their equipment. Zirkl (1961) also says in 1959 that the sheets "are in almost all cases a worthy substitute for calcite prisms". According to Moenke and Moenke-Blankenburg (1965) most producers of petrographic microscopes had incorporated sheet polarizers in their models by the early 1960s.

The enormous scientific and economic importance of microscopic examination of minerals and rocks during the 20th century should be evident to the readers. The books by Johannsen (1914, p. 199) and Rosenbusch (1924) list the major producers of petrographic microscopes in western countries. Among the pioneers in that field were R. & J. Beck and J. Swift in London, A. Nachet in Paris, R. Fuess in Steglitz, Voigt & Hochgesang in Göttingen, and Steeg & Reuter in Homburg. Others include E. Leitz and W. & H. Seibert in Wetzlar, C. Reichert in Vienna, C. Zeiss in Jena, R. Winkel in Göttingen (e.g. Wülfing 1918, Fig. 39-3), Société Genevoise in Geneva, C. Leiss in Steglitz (see his universal stage microscope in Fig. 39-3, and other instruments in Leiss 1925), and Bausch & Lomb in Rochester. The strong position of German firms in this field is no doubt in part connected to the vigorous research and development of glass- and lensmaking in Jena (see Moe 1990).

The firms of C. Reichert and E. Leitz also became known for their reflected-light microscopes. The latter originally produced the versatile instrument designed by Scheffer (1919, Fig. 39-3) where ore microscopy was one of the available options. Following the appearance of H. Schneiderhöhn's (1922) text on the use of reflected light and the marketing by Leitz of their large Erzmikroskop MOP in 1923, "whirlwind development" of this field within petrography took place (according to Schneiderhöhn in a 1941 Leitz Festschrift). Glaser (1924) describes various design features in reflected-light microscopy, intended for instance to study internal tensions in metal alloys. It appears to me from a cursory look at the literature, that while polarizing arrangements are fairly widespread in present-day microscopical metallography, they were not much used before 1930. Their application in metal research also has been relatively less than in work on ore minerals. This is perhaps due to the fact that many common metals and alloys belong to the cubic crystal

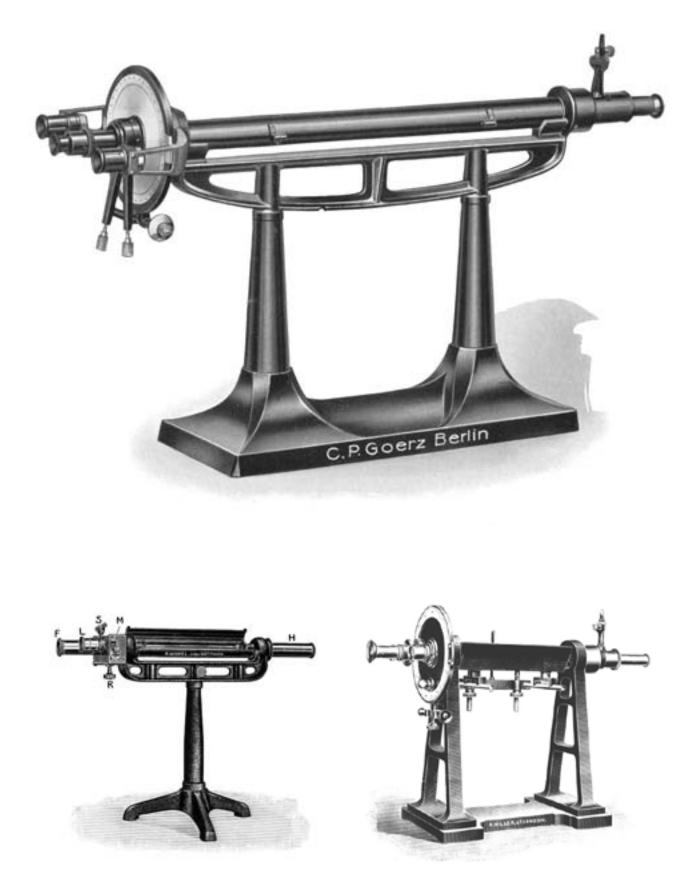


Figure 39-2. More polarimeters. Top: A medium circular-scale instrument from a 1924 C.P. Goerz catalog. Bottom left: A polarimeter included in R. Winkel's 1925 price list. Right: An A. Hilger instrument dating from around 1913, illustrated in a Cenco catalog.

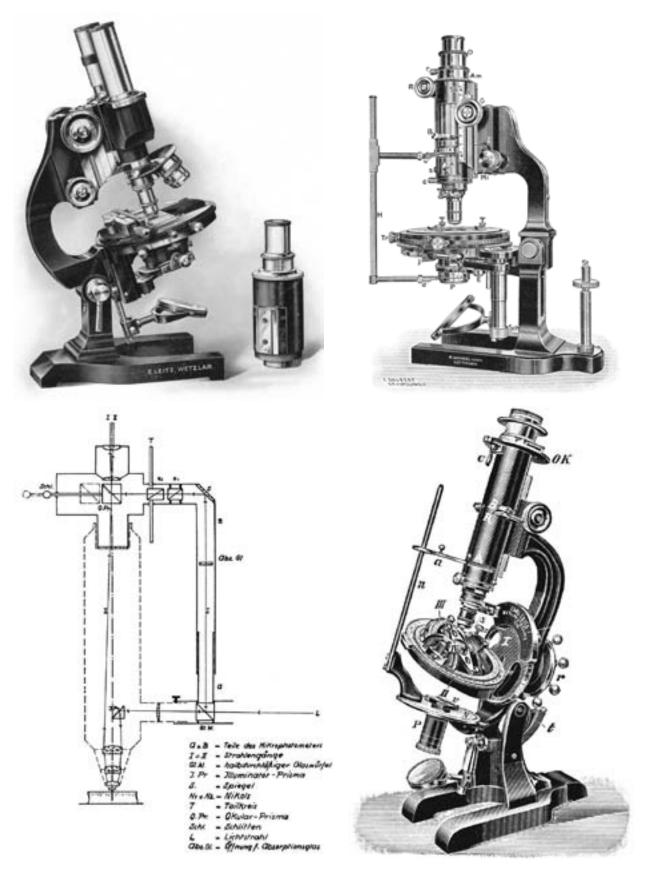


Figure 39-3. New microscopes. Top left: Scheffer's (1919) versatile microscope from E. Leitz, which among other things allowed observations in reflected polarized light. The picture is from Schneiderhöhn (1922). Right: A sophisticated instrument designed by Wülfing (1918) and produced by R. Winkel (from Leiss 1925). Bottom left: Frick's (1930) polarization photometer for Leitz reflected-light microscopes. Right: A universal-stage microscope offered by Leiss (1925), incorporating various improvements on older models.

system, and in others the reflections are not very dependent on direction (Dayton 1935). In a review paper, Mott and Haines (1951) state that uranium (orthorhombic) and beryllium (hexagonal) are the metals most often investigated in polarizing microscopes, which is connected to their role in the harnessing of nuclear energy.

Weigert (1927, p. 125) considers polarization instruments to be the most widely used kind of photometers. I have not looked much into the fortunes of such photometers for white light after 1930, but the photometric attachment of Frick (1930, Fig. 39-3) for reflection microscopes may be mentioned here. Berek (1931) published an important paper on precise analyses of ore minerals in such microscopes, where Nicol prisms were used both in the measurement of polarization and intensity of the reflected light. This was possible due to Berek's invention of a new glass prism which altered the direction of a light beam by 90° without affecting its state of polarization (German patent 630359, awarded in 1936). These microscope photometers were produced to 1960 at least. Sauer (1934) of the Zeiss company developed a new polarizing photometer, and Beck (1943) described improvements on the Martens photometer in the middle of WW II. An illustration of the latter may be found in a widely read textbook (Bergmann-Schaefer 1959), accompanied by the assertion that Nicol prisms are "particularly well suited for photometric work". The book also contains an illustrated description of the polarizing photometer Leifo which E. Leitz began producing in 1933. Leifo was for a long time a popular instrument for routine colorimetric work, for instance in chemical analyses of metals and alloys, in biochemistry and medicine, and in environmental science. An updated version containing a photoelectric sensor is displayed in the book of Moenke and Moenke-Blankenburg (1965).

I.G. Priest who was mentioned at the end of section 29.4 and in section 29.7, later added two quartz plates to his three-prism photometer, in order to estimate quantitatively the color of a light source (Priest 1923). A somewhat similar instrument with five Nicol prisms was later invented by Dowell (1931) and sold commercially. See both in Fig. 39-4.

Of course the development of various types of spectrophotometers which attenuated light beams by means other than Nicol prisms (see at top of section 29.4), continued from 1870 and into the 20th century. However, the polarizing types including new versions e.g. by Gehlhoff and Schering (1920) did survive to some extent up to 1950 at least. Skinner (1923, p. 736-737, Optical Society of America (1925) and Houstoun (1927) discuss them briefly, and Houstoun states (p. 324) that they are "much the most popular" of spectrophotometers. E. Brodhun claims in a review in Ergebn. Exakten Naturwiss. 1927 that the König-Martens meter continues to be the most commonly used visual spectrophotometer both in Germany and abroad. The Lemon (1914) instrument was produced commercially (Fig. 39-4), a new versatile one was introduced by the firm of Jobin (1922), and another one by Bellingham & Stanley (Schoen 1927). In a chapter on photochemistry in Abderhalden's Handbook of Biological Laboratory Methods published in 1926, T. Swensson informs readers that "the best spectrophotometers depend on the polarization principle". He mentions in particular the Hüfner and the König-Martens instruments, which may be used in the whole range of visible light. Weigert (1927, p. 193) also praises the König-Martens meter. In a paper on methods of light-absorption measurements, Laszlo (1928) states that most of these methods depend on Nicol prisms. He cites as examples the new Bellingham & Stanley spectrophotometer and the König-Martens meter. The latter was still on the market in the U.S. around 1940, along with at least three other polarizing spectrophotometers (Bates et al. 1942, p. 304-310), and it was widely in use in Europe as late as 1955.

Some of the available flicker photometers mentioned in section 29.4 were of the polarizing kind. Photoelectric sensing and recording of the light intensity was well suited to the flicker instruments, because an alternating voltage is easier to amplify (see e.g. Tardy 1928, Bruhat and Chatelain 1933, Hardy 1935). One such spectrophotometer with Rochon-, Nicol- and Wollaston

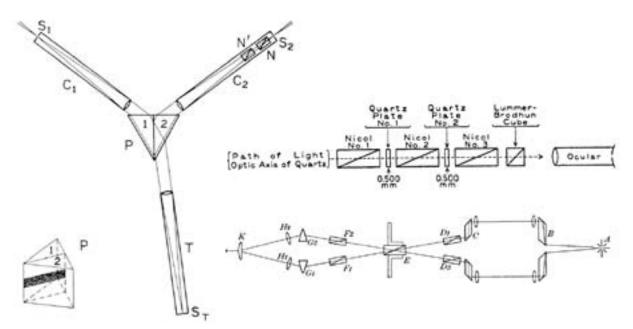


Figure 39-4. Photometers after 1920. Left: Lemon's (1914) photometer, later produced by Gaertner Scientific Corp. In the telescope T, the experimenter compares light from C1 and a more intense light from C2; images of these two appear side by side in the bisected glass prism P. By rotating one Nicol prism, he attenuates the light in C2. The diagram is from a committee review (Optical Society of America 1925). Top right: Priest's (1923) device for comparing the unknown brightness and color of an area to the left, with a standard light source below the Lummer-Brodhun prism. The experimenter is looking from the right. Bottom: The polarizer part of a photometer suggested by Dowell (1931). It has four fixed Nicol prisms and one (E) to be rotated. A is the light source. For instance, a vial with a fluid to be analyzed can be inserted into one of the two horizontal light beams. E is then rotated until the light spots seen from K appear to be of equal brightness.

prisms was designed by Hardy (1935, 1938) and mass produced by General Electric Co. It soon served in a variety of roles including calibration of reference samples for colorimeters, in the metal industries, in the construction of the first atomic bombs (according to the book Analytical Chemistry of the Manhattan Project, published in 1950), and in the production of dyes for color photography.

Around 1920 or earlier, chemists' interest in the use of ultraviolet spectrophotometry (especially for measuring light absorption by chemicals) had become sufficiently widespread for manufacturers to start offering such equipment. For instance Lowry and French (1924) use a new ultraviolet spectrophotometer (similar to the Nutting meter of section 29.5) that probably contained Iceland spar prisms, in their research on camphor compounds. According to Weigert (1927, p. 125-130, 156-157, 199-200) an ultraviolet meter of the König-Martens type is available from Schmidt & Haensch, and he also describes special versions of that instrument for measurements on thermal and other infrared radiation.

39.2 Astrophysical observations by B. Lyot and others

B. Lyot (1926, Fig. 39-7) constructed a very sensitive polarimeter containing a thin plate of Iceland spar, in order to measure very small (to 0.1%) proportions of linearly polarized light mixed with unpolarized light. He pointed this instrument at e.g. the moon and five planets (Lyot 1923, 1929, etc.) and compared the results with light reflected from various surfaces. One of his conclusions was that the surface of the moon, and even also those of Mars and Mercury, might consist of rock dust or cindery material. Wright (1927a,b, 1934) similarly analyzed the moonlight with a

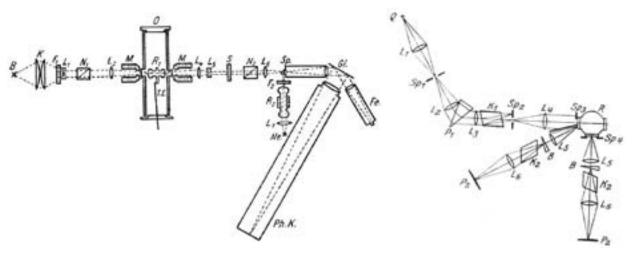


Figure 39-5. Left: Minkowski's (1921) test of quantum-mechanical theories concerning the Faraday effect in sodium vapor. Polarized light comes from the left through a glass vessel R containing the vapor in the oven O in a magnetic field. It is then analyzed by means of the Savart plate S, a Nicol prism, a diffraction grating Gi and the camera Ph.K. Right: Hanle's (1926) investigations of the effects of electric and magnetic fields on the polarization of resonance radiation from mercury vapor in the glass bulb R. Ultraviolet light from Q excites radiation which is then measured with the equipment below (shown in two positions). K1, K2 are calcite rhombs or Nicol prisms, B is a phase compensator, P2 is a photographic plate.

polarimeter and polarizing photometers, considering it to be reflected from a powdery substance rather than solid rock. Lyot's concept later led to the development of equipment (Harrington et al. 1956) to measure the polarization of fluorescence, which yielded information on certain properties of large molecules such as proteins.

Öhman (1933) who was making measurements on the polarization of starlight around 1930, used an Iceland spar rhomb to aid in the determination of star magnitudes from photographs. His method may not have been entirely original, as Leavitt (1914) had employed two Iceland spar plates in a similar way in her survey of circumpolar stars from 1909. Öhman carried out various studies on astronomical polarization up to around 1950.

Let a large object emit light of a wide range of wavelengths, and let us want to look at the whole object simultaneously to see how the emission of light of a chosen wavelength interval is distributed across the object's surface. If this interval is wide, it can be done easily with ordinary color filters. If the desired interval is very narrow (for instance containing only the red hydrogen spectral line), neither such filters nor spectroscopes turn out to be adequate. One solution is to use so-called interference filters, which are most commonly based on multiple reflections in very thin transparent films. A type invented by B. Lyot (1933) employs interference of polarized light (cf. Fabry and Perot 1900). For each individual filter, thin plates of quartz or Iceland spar are needed in addition to a large number of good-quality polarizing prisms. Lyot could not procure these until 1939 when he (Lyot 1944, 1945) was given a "beautiful rhombohedron of Iceland spar". A less satisfactory version with Polaroid sheets was in use from 1938, and in fact the above-mentioned Öhman (1938) had independently developed the same concept. The Lyot filters soon became very useful for studies of the solar surface, and Lyot also took with them movies of transient phenomena in the corona. According to Evans (1949), quartz had mostly replaced calcite in Lyot filters by 1950.

The third Iceland spar device invented by B. Lyot for astrophysical research (after 1940) contained a rotatable thin plate of Iceland spar. With this device mounted in a telescope, very small angles (for instance between double stars) could be measured in a similar way as was done with Rochon and Wollaston prisms long before (section 12.1). The Lyot micrometers are still being used.

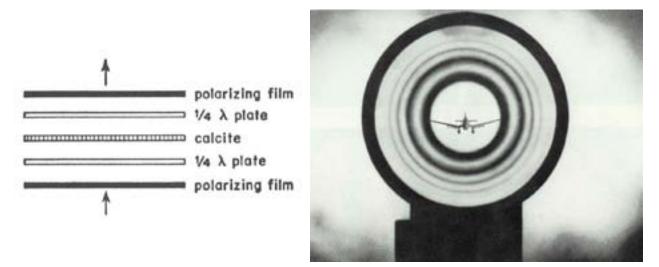


Figure 39-6. Polaroid Corp.'s Optical Ring Sight from World War II. Left: A drawing of its composition. Right: Taking aim at an aircraft. The rings followed the eye of the gunner so that it did not have to be directly over the center of the sight (Wood 1977). The diameter of the device was 3-4 cm and its thickness about 0.5 cm (L. Hughes, pers. comm. 2001).

39.3 Light and magnetic fields, from around 1925

In a 1944 textbook on quantum chemistry, H. Eyring and his co-authors stated that "…investigations of the Zeemen effect provide one of the most powerful tools for the determination of the states involved in atomic spectra". In the wake of the Stern-Gerlach experiment and theoretical contributions by W. Pauli on A. Landé's rules of spectral relationships (section 30.2), G.E. Uhlenbeck and S. Goudsmit suggested in 1925 that electrons had a quantized property called "spin". This provided an explanation both of the anomalous Zeeman effect and of the low values of the ratio e/m for electrons obtained from the Faraday effect. In turn it led to new theoretical work by Rosenfeld (1928) on the reasons for optical activity in liquids and gases, which was the first such investigation based on quantum mechanics.

A. Cotton who has been mentioned previously, began in 1912 to seek funding for a very large research electromagnet. The various effects of magnetism on optical processes were among the chief motivation factors in these efforts, but reasons given by Cotton (1914) also include interest in measurement of magnetic properties in materials like Iceland spar. After some delays due to the World War, Cotton and others managed to install a 100-ton 100-kW electromagnet in Paris in 1928. Various related equipment was added later, such as a large fixed magnet, spectroscopes, and low-temperature facilities. Many important discoveries were made with this apparatus in the following decades, both regarding magnetic effects on light and in other areas such as nuclear physics. Cotton himself carried out observations on one class of the Cotton effect which only occurs in magnetic fields (magnetic circular dichroism); this phenomenon has proved to be quite useful in research on all kinds of magnetic materials. The interaction of magnetism and light in matter no doubt prompted others to improve their experimental equipment within that field; for instance, Kapitza and Skinner (1925) studied the Zeeman effect in radiation emitted inside current coils while a powerful accumulator was momentarily short-circuited through them.

J. Becquerel who appeared in section 30.2, continued his interesting research on the Faraday effect and related phenomena (especially in paramagnetic compounds at low temperatures) from

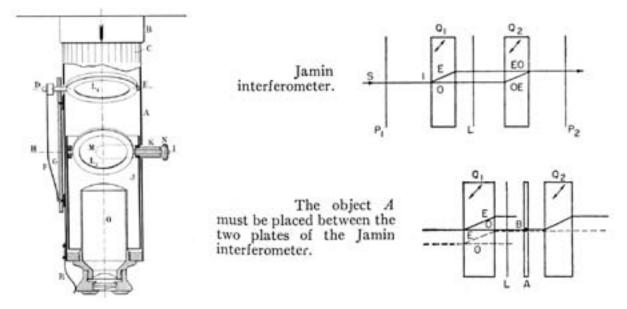


Figure 39-7. Two very sensitive interferometers based on Iceland spar. The principles of their operation will not be explained here. Left: Lyot's (1926) instrument which detects a small polarized proportion in a light beam, received for instance from a planet. The light enters from above through the glass plates L1, L2 which can be tilted, to the eyepiece O. At the base of O there are two identical Iceland spar plates and a beam-splitting prism of Iceland spar and glass. Right: A schematic diagram of Lebedeff's (1930) interferometric method to display small changes in the thickness or index of refraction of the specimen A (such as a histological thin section) as changes in brightness when viewed from the right. Q1, Q2 are two identical thin Iceland spar plates, P1, P2 are Nicol prisms and L is a quarter-wave plate (Françon 1957).

1925 (Onnes et al. 1925) and into the 1930s. The Kerr magneto-optic effect which was mentioned in chapter 23, is in fact a manifestation of the Faraday effect in ferromagnetic materials. These effects provided valuable insight into the nature of magnetism, which would have been difficult to obtain by other available methods. See e.g. Ingersoll and Vinney (1925) who investigated the magnetic properties of very thin layers of nickel. The Kerr effect has also been utilized in the late 20th century, in for instance magneto-optical disks for large computer memories and storing of images.

An important aspect of the development of quantum theory concerned its ability to predict the probabilities of an electron to move from one of its energy levels in an atom to another level. In its original form, quantum theory seemed to have nothing to say about such transition probabilities, but Einstein (1917) was one of the first to apply the theory to this aspect. The transitions are a key element in the interaction between light and atoms, governing among other things the dispersion behavior of materials. Experimental tests of the predictions of the quantum theory in this case included for instance the measurement of the relative intensities of neighboring spectral lines under given conditions (as discussed at the end of section 29.4). The relative intensities of the subordinate lines into which a spectral line was split in the Zeeman and Stark effects, could also throw light on transition probabilities. These intensity measurements were from 1920 or so mostly made on photographs, and photometers with Nicol prisms (especially the Martens photometer) may have played some role there, but other instruments were employed in the many examples which I have looked up. On the other hand, it was found that the transition probabilities in for instance hot metal vapors could be obtained by measurements of dispersion, absorption and Faraday-effect rotations in the vicinity of their spectral lines. Some data on such processes were already available (Senftleben 1915) and more were acquired by Minkowski (1921, Fig. 39-5), Ladenburg and Reiche (1923), Ladenburg (1926), Weiler (1929) and others. Iceland spar prisms were used in many of these studies.

Polarization of light also was involved in the theoretical interpretation of absorption of light by matter in magnetic fields, of band spectra and other phenomena. They were, like those of the last paragraph, connected to the emergence of the new and powerful version of quantum theory based on papers by W. Heisenberg and E. Schrödinger in 1925-26. Thus, Wood and Ellett (1923, 1924) found an unexpected effect by magnetic fields on the polarization state of so-called resonance radiation from low-pressure mercury vapor. This radiation is a type of fluorescence, characterized by the emitted light having the same frequency as the incident light. It occurs when an excited electron only has a single energy level which it is permitted to drop down to. The best known cases are an ultraviolet line of mercury (253.67 nm) and the two yellow sodium lines (near 589.3 nm) which R.W. Wood had in fact started investigating earlier (Wood and Hackett 1909, Wood and Mohler 1918). The phenomenon was studied in more detail by for instance Rayleigh jr. (1922), Gaviola and Pringsheim (1924), Hanle (1924, 1926, Fig. 39-5), Keussler (1927), Foote (1927), and Schein (1928), and its theoretical basis in terms of quantum postulates was discussed by W. Hanle, V. Weisskopf, N. Bohr, W. Heisenberg, and G. Breit.

One of those who became interested in resonance radiation was a young physicist by the name of E. Fermi. He wrote his first scientific paper (Fermi and Rasetti 1925) on a study of the polarization of this radiation from mercury vapor in high-frequency magnetic fields, using two calcite crystals. This paper marks the beginning of research in the "double resonance" method of spectral analysis of electromagnetic radiation. A. Kastler (1931) carried out a special experiment on the polarization of sodium resonance radiation in a magnetic field, to test the fundamental question of whether photons carry angular momentum. In his subsequent research on the Zeeman effect and resonance radiation, Kastler and his coworkers developed around 1950 a technique called "optical pumping" based on the double resonance concept. In this technique, metal vapor in a magnetic field which is absorbing polarized light at its resonance frequency, is simultaneously made to absorb a low-frequency electromagnetic radiation. The magnetic field splits the excited energy level of the vapor into two, and temporarily (due to the low-frequency radiation absorbed) more electrons end up occupying the upper one than the lower one. See Appendix 5 on Fermi and Kastler. Budker et al. (2002) claim that all the above research on polarized resonance radiation has its origin in the paper of Macaluso and Corbino (1898) from section 30.1. It has provided the foundation for a large new field of physics, leading to the development of important technologies such as lasers, very sensitive magnetometers, and atomic clocks which are essential to for instance space travel and the operation of accurate navigation systems.

39.4 The "Optical Ring Sight" and large-scale calcite mining in the U.S. and Mexico

When the United States entered WW II, their production of large and small gun sights increased greatly. Military officials were not quite happy about the traditional sights, which were too bulky and inconvenient in use. The above-mentioned founder of the Polaroid Corporation, E.H. Land, then invented a new and simple sighting device based on interference of convergent light in thin uniaxial crystal plates (Fig. 7-3). Land replaced the prisms which are used when such interference figures are observed in microscopes, by Polaroid sheets to make the device very compact. In this gunsight, only concentric circles are seen, as the black cross of Fig. 7-3 could be eliminated by placing quarter-wave plates on each side of the central plate (Fig. 39-6) of Iceland spar. This was by no means a new idea, as such a situation had already been demonstrated or analyzed theoretically by N.N. (Nicol 1829 in the list of references), Airy (1833a), Dove (1835, p. 607, and papers

by J. Müller in Ann. Phys. 1833-35), Wilde (1853), Soleil (1855b), Bertin (1859, 1884), Preston (1895), and Pellin (1899, Fasc. IV). Land (1947) was nevertheless granted a patent on this optical ring sight, as it was called.

Now, Land's company needed a large quantity of Iceland spar, preferably in rhombs like the Icelandic material, at least 3 cm in size and relatively free of defects and twinning although not necessarily of Nicol-prism quality. H. Berman who was advising the Polaroid Corp., sent a message to Iceland in March 1942 enquiring about the matter (L. Hughes, pers. comm. 2003) and the U.S. military again made enquiries in 1944, but this does not seem to have had much effect. A sizable amount of Iceland spar turned out to be present at a locality in the Taos county of New Mexico, discovered in 1931 but not yet exploited (Johnson 1940, Kelley 1940, and a news item in the Albuquerque Journal 2 Feb. 1942). However, DeMille (1947) claims that production had started there in 1939. It was considered that this mine was not far behind Helgustadir in total quantity and in the size of crystals: according to Blazek (1976), two crystals of more than 25 tons each had been found there. In the beginning it appeared that this source would be able to supply both the needs of the ring sight production and the U.S. Optics industry for a long while; however, according to minutes of the appropriations committees of the U.S. Congress which are accessible on the Internet, it had possibly been depleted already by the end of 1943.

Good Iceland spar was found in Tertiary volcanics in several places in Northern Mexico (Fries 1948), mostly in the vicinity of Chihuahua. About 9 tons were mined here in total in 1943 and 1944, although only a small proportion was first class. King (1946, 1947) describes various aspects of the efforts in the search for Iceland spar, its recovery, and the grading of the crystals. Some Iceland spar was obtained from Montana (cf. above), about 4 tons in 1942-44 (Stoll and Armstrong 1958), and 3 tons from California in the same period (Wright 1950). South- and Southwest Africa, Spain, and Colombia also delivered minor quantities.

The mining of Iceland spar in both the U.S. and Mexico ceased in the fall of 1944, when Land's Polaroid Corp. succeeded in growing saltpeter crystals (see West 1945 and Waesche 1960) to substitute for it in the optical ring sight. The sight was mostly intended for use in anti-aircraft and anti-tank guns (Stoll and Armstrong 1958, p. 431, McElheny 1998, p. 132). It appears to have been primarily installed on Navy supply ships and on merchant freighters, but L. Hughes (pers. comm. 2001) who has researched the history of the ring sight, concludes that its appearance on the war scene was too late to have a significant effect. The surplus optical-quality spar from the procurement efforts found its way to optical instrument makers and other users in the United States, "greatly exceeding their immediate needs" (King 1947).

39.5 Iceland spar in interferometry

At the beginning of chapter 18, two instruments invented by J. Jamin were described. They were intended for measurements of very small distances or very small variations in refractive indices, with the aid of interference of light. One of them which employed silver-coated glass plates and unpolarized light, became popular and is still in use. Occasionally, Nicol prisms and a phase compensator were also made use of, for instance by Clark (1906) in research on the optical properties of thin carbon films. Jamin's other type of instrument (Jamin 1868) which was based on interference between polarized rays, was largely forgotten (see however Kerr 1888) until revived by Skinner and Tuckerman (1911), Sagnac (1911) and others. They pointed out that through a certain kind of interference effects, the half-shade arrangement of Brace (1904a) could make the path difference of two rays visible as a change in light intensity even if it was less than 0.01% of

a wavelength. The method required high-quality polarizing prisms and was difficult to use, but Lebedeff (1930) found a practical way of employing it in microscopes, Fig. 39-7.

From around 1950, rapid technical development of this method took place. Production began of interference microscopes with Nicol or Wollaston prisms, for examination of biological materials where the refractive index varied very little across a sample slide (Smith 1955, Huxley 1957). These microscopes resembled to some extent the simpler and better known phase-contrast microscopes for biologists, which edged them off the market after 1970 (according to Dunn 2005). Living cells could be studied with the interference microscopes (e.g. Davies et al. 1954), which is said to have led to notable discoveries concerning the structure of cells and their movements. Analogous techniques in polarized light were also applied to the study of small deformations of surfaces, of air currents in wind tunnels, and of other phenomena involving very small variations in the phase of a light wave (Françon 1957, 1963).

It may also be mentioned here that interference methods were tested earlier in investigations of the flatness of cleavage surfaces on Iceland spar and other crystals (Siegbahn 1933). This was connected to evaluation of their suitability for X-ray spectroscopic work.

39.6 Pockels cells

The demand for polarizing prisms must have increased with the spread of laser devices after their invention around 1960 (cf. Nester and Schroeder 1967, Coetzee 1976). In a laser beam, considerable energy travels through a small cross-sectional area, and this can lead to rapid damage of Polaroid sheets or their successors due to the heat produced (e.g. Zirkl 1961). I have not looked much into the early history of the many applications of lasers, but one topic will be described briefly here.

Chapter 23 above mentions the effect of electrical fields on refractive indices of crystals which is named after F. Pockels (1891, and later). It occurs in those crystal classes which lack a center of symmetry (20 out of 32), i.e. the classes that exhibit piezoelectricity. This makes the effect different from the Kerr electro-optic effect which only is found in liquids and gases. Another difference between these two is that in the Pockels effect the change in refractive index is a linear function of the field strength, while the magnitude of the Kerr effect is proportional to its square. A crystal fitted with two electrodes can then function as a controllable phase compensator for light in the same way as a Kerr cell, but the light beam really needs to be quite monochromatic and parallel (see Goldstein 1986). Both these conditions are fully met by laser light. With a Nicol prism on each side of the crystal, we have a means of modulating the intensity of light beam, even shutting it off altogether.

Suggestions for technical applications of this effect in e.g. television sets had already appeared around 1930, cf. British patent no. 375856 and U.S. patent no. 1,879,138 (both granted in 1932) concerning the use of Rochelle salt to control light. In recent decades, people have mostly employed the so-called longitudinal Pockels effect, where the electrical field is parallel to the light beam. This variant of the effect primarily occurs in cubic or uniaxial piezoelectric crystals not having optical activity, which limits the choice to two crystal symmetry classes. Among suitable crystals is the compound $NH_3H_2PO_4$, i.e. ammonium dihydrogen phosphate, ADP. After WWII the Polaroid Corporation intended to base production of color television sets on the Pockels effect in this material (Land 1950), but very different approaches by competitors won that race. Before 1960, crystals of other compounds from the same symmetry class were being grown artificially for use in optics technology, including LiNbO₃ and KH₂PO₄ (KDP). Pockels cells do not require as high electrical voltages as Kerr cells, have shorter response times (less than 1 ns), and are

probably easier to handle. They have found applications in many types of apparatus, for instance in fiber-optics communications, video recording, and laser distance meters. Non-linear Pockels effects have also become important in optical technology.

39.7 The Raman effect

When a clear or turbid liquid is illuminated by an intense monochromatic source, spectroscopic examination shows the light scattered sideways to have precisely the same wavelength. C.V. Raman who was mentioned in section 36.7, succeeded with his collaborators in challenging this well-known experimental fact in 1928. If the light from the spectroscope falls on photographic paper for many hours, the photograph eventually exhibits a very faint line spectrum in the vicinity of the original spectral line. For a while this phenomenon was thought to be of the same character as the Compton effect, but it turned out not to be the case. The frequency shifts in the new lines correspond to certain resonance oscillations within the molecules of the fluid. The frequencies of these oscillations are much lower than those of visible light, as was the case in the molecular oscillations which can be excited in the liquids by infrared radiation (section 34.1). It is specifically stated in the first papers on this Raman effect (Raman 1928, Raman and Krishnan 1928) that the pronounced polarization of the frequency-shifted light (observed with Iceland spar prisms) contributed to the conclusion that it was a kind of scattered light rather than fluorescence. This is because light due to fluorescence in liquids is unpolarized in liquids if the exciting light source is unpolarized (see e.g. Sohncke 1897, p. 81-84, Zsigmondy 1925, p. 9). At a similar time the effect was also found in crystals, including calcite (Landsberg and Mandelstam 1928, Wood 1928, Cabannes 1929b) which was very well suited to such observations due to its transparency. Thus, Wood investigated in addition to quartz and various liquids, "a very clear natural rhomb measuring 4 x 5 x 2 inches" of Iceland spar. Similarly, Schaefer et al. (1930a) state that "In the largest crystal, a clear rhombohedron of Icelandic Doppelspat of 40 mm size, a few hours of exposure were sufficient". They also studied other materials, but they had to give up on some of these because their crystal samples were too small or too opaque. Physicists soon began combining the data on frequencies and polarization properties of the Raman radiation and of the residual rays of section 34.1 with quantum-theoretical models of molecular oscillations (e.g. Cabannes 1929a, Schaefer et al. 1930b, Kastler 1930, Hanle 1931, Menzies 1931). For calcite and other crystals, their structural characteristics deduced from X-ray spectroscopy provided a basis for the theoretical work. The Raman effect has ever since been very useful in chemistry, especially in research on covalent chemical bonds. See Appendix 5.

40 Iceland spar mining after 1940; concluding remarks

40.1 The Helgustadir quarry; mining abroad, applications of Iceland spar

One more attempt at mining optical crystals in the Helgustadir quarry was organized in 1946-47 by the National Research Council of Iceland, cf. an account by P. Skulason in his periodical Skjöldur, p. 18-20 of issue 3, 2002. S. Steinthorsson (pers. comm. 2001) also informs me that a small amount was recovered and that a technician in Reykjavik prepared the crystals for export. This effort was soon discontinued, perhaps in part because the responsible staff became occupied with monitoring the Hekla eruption which began in March 1947. According to official records, 27 kg of Iceland spar were exported in 1947. In a 1963 speech on the history of the research council, A. Thorsteinsson states that the crystals of 1947 had proved too small for use in instruments. A final mining attempt in 1952 with the council's participation was quite unsuccessful (according to newspaper accounts in early 1953), but about 10 tons of crushed inferior calcite were produced for use in the building industry.

The Helgustadir quarry was declared a protected "natural monument" by the Ministry of Education and Culture, in an announcement published in the government's gazette on 1 Dec. 1975. Among other things this declaration forbids collection of crystals at the quarry or in its vicinity.

The Optical Society of America awarded a medal to Karl Lambrecht, as recorded in the Journal of that society vol. 66, p. 188, 1976. After having been for some years an employee of manufacturers of optical instruments, he set up in 1933 his own company to produce polarizing prisms and other such components. In order to remedy the shortage of Iceland spar which had for years hampered the industry, he travelled in the 1950s to Mexico, South Africa, India and other countries to locate new sources. His high-quality prisms made of material from these sources were well received, and the Lambrecht company is still thriving. However, complaints of inadequate supply of Iceland spar appeared occasionally in the literature until 1960, see Appendix 1A.

Mines of optical-quality calcite which have been opened up in previously inaccessible areas, now seem to satisfy the demand, and advertisements for large amounts of Iceland spar may be found on the Internet, from e.g. Brazil, Namibia, China, and Siberia. Nice rhombs and whole crystals of other habits may be purchased at fair prices from dealers and museums. Numerous companies are producing polarizing prisms, often of the Glan type (with either glue or an air gap between the wedges), Rochon- and Wollaston beam-splitting prisms, variable compensators (as in Fig. 29-10 bottom left), and other items from defect-free Iceland spar. Many of these presumably end up as essential components or accessories in polarimeters, ellipsometers and microscopes, while others are used on their own in research on light, atomic physics, solid-state physics and so on.

40.2 Concluding comments on the quarrying activities

In Iceland, three different categories of parties were involved in the recovery and export of Iceland spar crystals through the ages.

- -individuals who collected crystals at Helgustadir, without special arrangements
- -private operators, quarrying the site commercially on lease from the owners
- -the authorities

Naturally, most of the surviving documentation relates to the part played the authorities. They were not always well aware of conditions in Reydarfjördur, but they wished to proceed carefully with the exploitation of the deposit and sought geological advice. They were also not well informed about the fate of the Iceland spar recovered, i.e. where it went, what it was used for, or what it was worth. In Parliamentary debates in 1922 for instance, it was claimed that the material was in particular demand for military purposes. The role of the authorities was in some contrast to that of the private operators (especially C.D. Tulinius) who were familiar with both the local conditions and foreign markets, who invested in mining equipment, and took the initiative to make these glassy rocks into a valuable international commodity. Critical comments on Tulinius and his son were voiced from time to time, as can be seen for instance in Appendix 1A and sections on the quarrying activities above. Thus it would probably have been more advantageous in some respects to spread the quantity recovered in 1863-72 over a longer interval of time and then to continue the quarrying activities at an even pace into the 1920s. Some of these criticisms may probably be countered with evidence from files in the National Archive and elsewhere. It is difficult today to judge the late 19th century situation in Iceland, including the effects of travel and communication being only sporadic, of national poverty, and of adverse climate, upon undertakings like the quarry operations. At any rate it is quite clear that the efforts of C.D. Tulinius and others in the exploitation of the Helgustadir calcite were of inestimable value to the natural sciences.

40.3 On optics and physics in schools

For most of the 20th century, optics represented a larger part of the physics curriculum in universities, and therefore a larger part of a physicist's knowledge base, than at present. In their first and second year, students of physics probably read optics books like those of Drude (1900a), Wood (1905), Schuster and Nicholson (1923) and Houstoun (1927) where double refraction and polarized light took up 50-70 pages. In older works such as by Verdet (1869) and Preston (1895) these topics were given even more attention. It was also not uncommon for general physics texts used in universities, high schools and self-study to contain a good deal of material on polarization. Thus, 187 pages of the university physics book by Christie (1864-65, 685 p. in total) are devoted to optics, whereof 54 treat double refraction and polarized light. The first volume of A. Paulsen's (1893) book on physics "presented in an understandable way for the general public" contains a whole chapter on polarized light (see Fig. 3-2). The 1100 pages of text in Ganot-Maneuvrier's Traité Élémentaire de Physique (24th ed. 1908) include 105 pages on optics with 13 on polarized light. In Dahlander (1902) the numbers are respectively 892, 157 and 38. It is evident that much of the Iceland spar prisms and other polarization-related items listed in catalogs like that of B. Halle (about 1895, Fig. 32-2) has been intended for practical instructions in schools and universities, see also Grimsehl (1905) and Rosenberg (1924). In the optics textbook of G.R. Noakes (expanded edition, 1940) for the advanced level of British high schools, 18 out of 355 pages deal with polarized light.

In the last half-century, new fields have squeezed coverage of older optical knowledge out of physics curricula, and other subjects have also tended to crowd out physics in the overall timetable of education below university level. In new editions of U.S. textbooks like those taught in the first year of physical sciences and engineering at the University of Iceland, on average less than 5 pages out of every 1000 specifically describe polarized light; in some cases, the authors even remark that it is merely a supplementary topic. These changes may be inevitable, but students are missing valuable insights into the nature of light and other vibrations if they for instance do not have the opportunity themselves to carry out practical exercises on polarization. A related matter concerns the presentation of currently active fields of scientific research in textbooks. Thus it is evident from the preceding chapters that polarized light played a wide range of significant roles in the early development of modern physics (i.e. quantum theory, relativity, atomic physics, etc.); yet it is completely ignored in many textbooks on that subject (such as A. Beiser's popular Perspectives of Modern Physics, published in 1969). One reason for this omission may be the authors' belief that students who are going to read their books have no prior knowledge of polarized light.

40.4 Some inaccurate statements on Iceland spar

Various statements regarding Iceland spar and the Helgustadir site which have appeared in the literature through the years, are inaccurate. Some keep cropping up even in recent Icelandic newspaper accounts or information for tourists. A few examples will be paraphrased below, with explanations.

"Most of the large specimens of calcite in foreign museums are from Iceland": calcite crystals from locations abroad exhibit much more varied forms than those in Iceland, and many of these are quite spectacular. Furthermore, large optical-quality crystals have been found since 1920 in various locations worldwide.

"Iceland spar was used in telescopes": this was generally not the case, and its uses as an accessory in telescopes were limited to specific astrophysical observations. They include photometry (mostly in 1870-1910, see references to Zöllner, Pickering and Lyot), ultraviolet spectroscopy (mostly 1870-1900, see e.g. references to Huggins and Cornu), magnetic field observations (after 1905, see references to Hale) and color filters (see section 39.2). One possible application would be in the measurement of small angles (like between double stars) by means of beamsplitting prisms. However, I have only come across one astronomer who may have attempted to make a double-image micrometer with calcite wedges, namely O. Lohse in studies of the planets before 1890. Others such as Arago (1847), J. Dollond in Phil. Trans. 1821, W. Pearson in Mem. Astron. Soc. 1822, A. Secchi in C.R. 1855, V. Wellman in Astron. Nachr. 1889, and E.C. Pickering in the Harvard Annals 33(7) in 1900, used only quartz wedges. Other types of astronomical micrometers were much more common. With respect to the above claim, it is possible that some people have considered the prisms in ordinary binoculars to be made of calcite, but they are always made of glass. Others may have confused telescopes with microscopes.

"Iceland spar was used in (microscope) lenses": such applications were extremely rare, and mostly for ultraviolet light. I have found no instance describing its use in microscope lenses. See e.g. section 29.8 and references to Fedorow and Lowry.

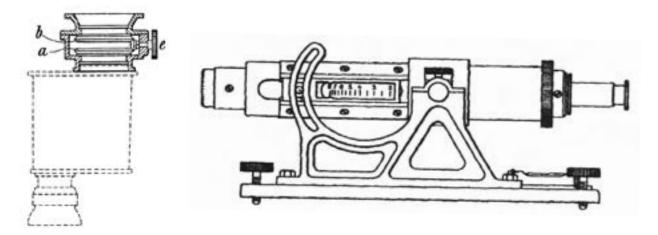


Figure 40-1. Simplified diagrams of two C. Zeiss telemeters, from patent documents. Left: A. König's invention according to a German patent no. 188992, 1906; the drawing is from U.S. patent no. 942393, granted 1909. The view is from above. With the knob e the observer moves the slim Rochon prisms a and b (from Iceland spar or quartz) laterally until two images coincide. Right: A drawing published in British patent no. 190922523, 1909 of a telescopic range finder containing a single Rochon prism.

"Iceland spar was in great demand for use in telemeters and other military equipment": this statement may be found e.g. in parliamentary debates in Iceland. Similar claims have also appeared abroad, even (in a news item on SW-Africa mentioned in section 37.3) that Iceland spar was used in the periscopes of submarines. The catalogs of Duboscq (1885) and Pellin (1899) include a small telescope with a quartz Rochon prism, to be used for estimating distances to objects of known size. This device (Fig. 13-7) may have been invented by Soleil (1855a). A more complex apparatus with a similar prism was tested by Giraud-Teulon (1875). Wulff (1897) discusses the possibility of measuring small angles with wedges of doubly refracting materials but he does not give any concrete examples. The firm of C. Zeiss obtained a German patent no. 188,992 in 1906, a U.S. one no. 942,393 in 1907 and a British one in 1910 for two different telemeters with Rochon prisms (Fig. 40-1); at least one of these employed calcite. I do not know whether they went into production, and despite considerable search efforts I have not found any solid evidence for the use of such devices prior to 1925 (e.g. in WW I). However, it is clear that Iceland spar prisms were essential in the testing of optical glass and glass objects for strains, regardless of whether they were intended for military or civilian purposes (Wright 1921).

"Dichroic materials have completely replaced Iceland spar in optical equipment": such materials (i.e. the successors of Polaroid sheets, see section 35.2) have both advantages and disadvantages compared to Nicol prisms and other Iceland spar polarizers. In many instruments and experimental setups, for instance those using intense laser beams, Iceland spar prisms are still preferred. Advertisements and catalogs from many current manufacturers of these may be found on the Internet.

"The Iceland spar crystals at the Helgustadir site were damaged by the use of explosives": contemporary accounts show that gunpowder explosions to dislodge basalt blocks had started by 1870, and continued into the 1900's, cf. Fig. 32-1. Rumors of specific parties causing damage in this way were for instance afoot in 1910-11 (Thjodolfur newspaper, 7 Oct 1911, p. 146) and have persisted. Good crystals were however recovered from the quarry up to its closure in 1925. King (1947) states that adverse effects of dynamite explosions on optical calcite in rock reach about

5 meters from the shot point. A quite different account of possible damage to the spar at Helgustadir may be found in a small book by H.N. Holmes: Strategic Materials and National Strength, published in New York in 1942. He reports that during WW I the quarry had been flooded with water in order to minimize weathering of the crystals, this operation having even worse effects on them.

"The largest Iceland spar crystal ever found came from Helgustadir": this claim possibly originates in a biographical essay on the skipper Brynjólfur Jónsson by Ásmundur Helgason, published in 1946. In the essay it is reported that Jónsson's boat carried a 600-pound crystal to Leith in 1870 in a cargo of other Iceland spar of both optical and inferior quality. This crystal found a permanent home in the British Museum and is presumably still on display there. On the Museum's home page it is said to be 60 cm in size and to have been acquired in 1876. It may have been the largest specimen recovered in one piece at Helgustadir, but there are reliable reports of much larger calcite crystals occurring both there (Des Cloizeaux 1846-47) and elsewhere (see e.g. section 39.4). In the 1990s, a 17-ton crystal was found in China (Zhaochang 1994).

Appendix 1A: Various statements regarding the Helgustadir quarry and the supply of Iceland spar, especially shortages. (Note: some of the persons cited may not have had access to accurate or up-to-date information)

-1857 Foucault, Fr... le prisme de Nicol devient dispendieux et difficile à se procurer en raison de la rareté des beaux échantillons de spath.

-1874 Ladd, U.K.: In ...1869 I constructed two Nicol's prisms of about 2.5" aperture[but now the demand for prisms of this size has increased so much that] very great difficulty is experienced in procuring pieces of spar of sufficient purity to give such a field. He therefore made such prisms by combining smaller ones.

-1877 Laurent, Fr. similarly complains about having to glue together small pieces of Iceland spar in order to make sufficiently large Nicol prisms for his new projector.

-1878 v. Zahn, D. in footnote: Wie schwer es ist Nicols von der erforderlichen Güte zu erlangen, weiss jeder, der sich mit der Zusammenstellung eines Polarisationsphotometers abgegeben hat.

-1878-79 Des Cloizeaux, Fr. obtains samples of Iceland spar from the Mineralogical Museum of Copenhagen University (according to the University's yearbook).

-1882 or later, book by Merrill, U.S. (1905) ... The chief commercial source of the mineral has for many years been Iceland... From [1872] until 1882 the mine remained idle, when operations were once more renewed, though not on an extensive scale, owing, presumably in part, to the fact that Tullinius, the last year he rented the mine, had taken out a sufficient quantity to satisfy all the needs of the market. Merrill does not mention other locations, or how long the stockpile lasted.

-1882 Bréon, Fr..... Eskifjord, localité qui seule a fourni au monde civilisé cette précieuse substance pour les instruments d'optique.

-1884 Bertrand, Fr. ... un morceau pur et assez gros de spath d'Islande, substance chère et qui devient de plus en plus rare.

-1884 Feussner, D... während die grossen, homogenen Kalkspathstücke, welche.... die Prismen sehr theuer machten...;

-1885 Madan, U.K. ...Iceland spar, a material which is becoming deplorably scarce and expensive (I question if there is in England or elsewhere a piece of spar fit to make a Nicol's prism of 5 cm. aperture).

-1886 Stokes, U.K. ... see letter in Appendix 1C below.

-1887 Labonne, Fr. ... la mine du spath d'Islande, substance dont la rareté préoccupé a juste titre les laboratoires de physique et de minéralogie... Le gite est actuellement concédé à un negociant islandais, M. Carl Tulinius, qui se contente de vendre au jour le jour le spath abattu, sans poursuivre aucun travail d'exploitation régulière...

-1887 Reichskanzleramt, D.... makes enquiries in Copenhagen, see Zeitschr. D. Geol. Gesellsch. 1888 which states that Iceland spar crystals occur "nur vereinzelt" and "als mineralogische Seltenheiten" elsewhere.

-1888 Deslandres, Fr. needed two Iceland spar prisms for an ultraviolet spectrograph and managed to procure them; however,Le second prisme a été difficile à obtenir, à cause de la rareté actuelle des beaux morceaux de spath.

-1889 Chauvin, Fr. Malheureusement, les échantillons de cristaux épais et transparents, nécessaires pour ces expériences, sont très rares....

-1889 Haensch, D, in speech: Der Doppelspath...ist in den letzten Jahren so hoch im Preise gestiegen, dass es gar nicht mehr lange dauern wird, bis wir den zehnfachen...Preis bezahlen müssen....die Hauptfundorte des Doppelspaths liegen bekanntlich in Island...Da inzwischen die Vorräthe in Doppelspath völlig erschöpft sind, so wird es in kurzer Zeit nicht mehr möglich sein, manche wissenschaftlichen Apparate und solche für industrielle Zwecke herstellen zu können.

-1890 Hoskyns-Abrahall, U.K speaks of difficulty in obtaining large clear pieces, and Madan (1890) mentions ... these spar-famine days.

-1890 Grosse, D...Leider ist der Preis des Kalkspathes ein sehr hoher und die Verluste bei der Anfertigung der Prismen so gross, das nur etwa 5% des Rohmaterials nutzbar sind, da von fünf gleich grossen Stücken nur etwa ein Stück brauchbar ist und von diesem bei Bearbeiten...wieder drei Viertel verloren geht.

-1890-91 Tait (see Sang), U.K...in view of the scarcity of this precious substance. The great dearth of Iceland spar is also mentioned by S.P. Thompson in an account of the annual meeting of the British Association (The Times, 27 Aug 1891).

-1893 Lacroix, Fr. ...Les physiciens d'alors [around 1845], et Biot en particulier, se préoccupaient déjà de la rareté du spath d'Islande, rareté qui n'a que s'accroître depuis.

-1893 Knipe, U.S. describing a new projector, in Science 22:...unfortunately it is now impossible to obtain such large crystals of spar [i.e. for making Nicol prisms 2" across]

-1894 Tschermak, A, on Helgustadir...Dies ist das einzige bisher bekannte Vorkommen von grossen, klaren Individuen und liefert demnach alles Material für optische Zwecke.

-1895 Wulff, D. Der Substanz [i.e. sodium nitrate], im Fall der Kalkspatmangel anhält, wird viel gezüchtet werden müssen....

-ca.1895-99, B. Halle catalog, D...Rohe Krystalle, vornehmlich Isl. Doppelspath und Bergkrystall, zu optischen Zwecken brauchbar, werden stets gekauft oder in Tausch angenommen.

-1896 Macnair, U.S. (in Nature 53) ... the present scarcity of Iceland spar...

-1896 Turner, U.K. [decided against acquiring a double-image prism to use in a solar-eclipse expedition to Japan, as] ...this would mean a great expense, at the present price of Iceland spar (Monthly Not. Royal Astron. Soc. 57, p. 110).

-1897 Elster and Geitel, D...die Beschaffung eines Nicol'sches Prismas von den nöthigen Dimensionen bei völliger Reinheit des Kalkspathes nicht möglich war,...

-1897c, 1899 Leiss, D.....die Schwierigkeit der Beschaffung optisch brauchbaren Kalkspathes... hat sich in den letzten Jahren immer mehr und mehr gesteigert....Das zur Herstellung grosser und reiner Prismen erforderliche Material ist jetzt entweder schon gar nicht oder nur mit sehr bedeutendem Kostenaufwand zu erhalten...., [and] ...Der sich mehr und mehr geltend machende Mangel an optisch brauchbarem Kalkspat...Nur für die Herstellung kleinerer analysierender Nicol'scher Prismen ist im allgemeinen noch genügend Material vorhanden.

-1898 v. Lommel, D. on glass-calcite prisms:...die Hälfte des immer kostbarer werdenden Kalkspathmaterials zu ersparen.

-1901 Comment on Sokolow (1898) in N. Jahrb. Mineral., D....Bei dem Seltenwerden absolut wasserklaren und fehlerfreien Doppelspathes in Island verdient die Krymsche Lagerstätte eine eingehende Untersuchung....

-1903 Becker, D. on polarizing prisms of the Feussner type:...Der wesentliche Zweck dieser neuen Polarisatoren geht auf die Ersparnis an dem immer im Preise steigenden Kalkspat hinaus. (He also states that nitrate polarizers may be used with advantage).

-1903 Brauns, D....es ist noch nicht gelungen, den immer seltener und teurer werdenden klaren isländischen Doppelspat zu ersetzen.....Zum grossen Bedauern der Optiker entspricht die Produktion von klarem Doppelspat lange nicht dem Bedarf und der Preis geht stetig in der Höhe...

-1905 Thompson, U.K....This last consideration has been forced upon designers by the increasing scarceness of the material since the closing of the mines in Iceland.

-1905 Turner, U.K. (Proc. R. S. A77, 1906) made repeated but unsuccessful attempts to obtain a replacement for a faulty Iceland spar prism, to use in solar eclipse observations.

-1906 Firma Carl Zeiss, D., letter to the Minister for Iceland ...die für gewöhnlich im Handel vorkommenden Krystalle für optische Zwecke meistens nicht brauchbar sind.

-1906-07 Cheshire, U.K.: Iceland spar has never been obtained, I believe, of optical quality and of large size, except from a single mine in Iceland, and even this supply has now been exhausted for some years.

-1907 A. Torfason, an industrial chemist in Reykjavik, advertises in the local newspapers that he wishes to buy clear crack-free Iceland spar at a high price.

-1908 B. Halle, D, in a description of improved methods of making polarizing prisms: ...der Isländische Spat ist seit 30 Jahren um das 20-fache im Preise gestiegen.

-1909 Lummer, D. ... Nicols grosser Apertur sind unerschwinglich.

-1909 Stadthagen, D... Leider findet sich dieses Material in optisch brauchbaren Stücken und in wesentlichen Mengen nur in Island...In Island gebe es zwei Fundstellen...

-1910 G. Halle, D... Bei den stetig zunehmenden Mangel an leidlich reinem Kalkspat, besonders grösserer Stücke...eine ungewöhnliche Preissteigerung [ist] eingetreten.

-1910 Badermann, D., is very critical of the Danish (sic) company which has been selling poorly sorted Iceland spar from its two (sic) quarries in Iceland to dealers: Es wurde aber...nicht die geringste Rücksicht auf die Auswahl und Güte des gewonnenen Produktes genommen...So stellte sich nach und nach im Bezuge von Doppelspath ein immer grösserer Notstand heraus... The optical workshops are at the mercy of these dealers who charge exorbitantly for the good-quality material. In the near future however, sodium nitrate may provide some competition.

-1911 Schulz, D... ist bisher vorwiegend Kalkspat zu Polarisatoren benutzt worden; jedoch ist mit zunehmender Verwendung die Erlangung brauchbaren Materials so schwierig und damit der Preis ein so hoher geworden, dass Prismen von grösserer Öffnung fast unerschwinglich geworden sind.

-1911 Busch and Jensen, D. on the Foucault type of Nicol prisms:Hierdurch kann, was natürlich bei der Seltenheit des isländischen Vorkommens sehr ins Gewicht fällt, das Prisma sehr verkürzt werden.

-1911 Encycl. Britannica, U.K.in recent years much of the material taken out has not been of sufficient transparency for optical purposes, and this, together with the very limited supply, has caused a considerable rise in price. Only very occasionally has calcite from any locality other than Iceland been used for the construction of a Nicol's prism.

-1912 Coker and Thompson, U.K....at the present time, the number of large prisms in existence is very limited, and there is a complete dearth of spar of a suitable kind, owing to the stoppage of the supply from Iceland, and the lack of any other supply of sufficiently good optical quality.

-1913 Dammer and Tietze, D.... Da die Feinmechanik fast ausschliesslich auf den isl. Kalkspat angewiesen ist und die Beschaffung dieses in ausreichender Menge und genügender Qualität oft mit Schwierigkeiten verknüpft ist.... [The quarrying in Iceland is said to have been at times "wahrer Raubbau"]....bis in der actziger Jahre konnte der Betrieb nur in sehr geringem Umfange geführt werden, da ein Unternehmer Anfang der siebziger Jahre so grosse Mengen auf den Markt geworfen hatte, dass der Bedarf lange Zeit hindurch zum grössten Teile aus diesen gedeckt werden konnte.

-1913 Lowry, U.K... But the increasing demand for the spar has not been accompanied by any corresponding increase in the supply, and large clear pieces of the mineral are becoming increasingly difficult to procure. It may indeed be doubted whether large polarising prisms such as those which have been handed down as heirlooms at the Royal Institution could now be purchased at any price, in view of the "sparfamine" which has prevailed for some years.

-1915 Beckenkamp, D..... Nun ist das zu grösseren Nicols brauchbare Material auf nur eine Fundstelle,

oder sogar einen einzigen Kristallblock auf Island beschränkt, und zur Herstellung grosser Nicols werden schon lange die seit älterer Zeit in den Sammlungen befindlichen grossen Spaltungsstücke von Island verwendet; der Preis grosser Nicols ist daher sehr hoch, während man für kleine Nicols auch das Material anderer Fundorte verwenden kann.

-1918 Parsons, U.S. ...Since the 17th century science requirements for optically perfect calcite have been supplied almost wholly from [Helgustadir]...In recent years, however, the difficulty in securing first class spar from the Iceland deposit has steadily increased, much of the material being taken out being use-less for optical purposes.

-1919 Wright, U.S. ... Wollaston prisms were not obtainable during the war... P.G. Nutting also states in J. Opt. Soc. America 1, p. 83, 1917 that calcite for making Wollaston and Nicol prisms for use in Martens photometers is not available.

-1919 Schulz and Gleichen, D...Bei dem hohen Preise und die Schwierigkeit der Beschaffung geeigneten Kalkspates....

-1919 Nelson, U.K. It is now no longer possible to add a large Nicol to one's box of microscopic apparatus, because of the greatly increased price of large pieces of calcite. (In another paper in 1917 he calls Nicol prisms "an expensive luxury" for microscopists).

-1920 Lord Rayleigh jr., U.K., mentions that he only had half a calcite Wollaston prism for experiments, and could not procure another half.

-1921 T. Crook, U.K, an expert in mineral resources, states in his book on Economic Mineralogy that: ...large transparent and flawless pieces such as are required for making polarizing prisms [have] only been found in any quantity in Iceland,...B.K. Johnson is not optimistic in his paper in J. Royal Microsc. Soc. in the same year: Iceland spar is very scarce and is becoming more scarce.

-1921 Halle, D., describes his methods....zur Ersparnis des immer kostbarer werdenden Rohmaterials... in the production of various types of Nicol prisms.

-1922 The Minister for Industries, Iceland, states in Parliament that there is great demand from many quarters for optical-quality spar.

-1922 Tutton, U.K. Large crystals are now becoming rare and exceedingly valuable ...Such large prisms can now no longer be made, the original magnificent deposit of enormous clear rhombs of Iceland spar at Eskifjördhr having become exhausted ...There is, happily, plenty of smaller calc-spar constantly being discovered, so that Nicol prisms for all ordinary crystallographic apparatus and the polarising microscope are more or less readily procured...

-1923 Glazebrook, U.K., describing construction of Nicol prisms: ...have sought the ideal mean between a prism of greatest utility and one involving small waste of material, an almost equally important consideration owing to the small supply of large flawless crystals of Iceland spar.

-1924 Stöber, D. ... Da die isländische Kalkspatgruben, die einzigen, welche für den zu optischen Zwecken brauchbaren Kalkspat in Frage kommen, schon seit längerer Zeit so gut wie erschöpft sind, so macht es sich in den optischen Werkstätten der Mangel an Kalkspat... immer mehr fühlbar.

-1924 Glaser, D, discussing Nicol-prism types for metallography...ein Umstand, der bei der heutigen Knappheit an gutem Kalkspat stark ins Gewicht fällt.

-1925 Bouasse, Fr. book... D'où l'importance historique et technique de ce cristal dont les gros échantillons se vendent au poids d'or.

-1928 Schulz, D. in Wien-Harmsein in jeder Beziehung brauchbarer Ersatz für das bis vor kurzem fast nur in Island in guter optischer Beschaffenheit geförderte Material bislang noch nicht gefunden ist.....

- 1928 Schönrock, D. Eine Zeitlang [around 1920?] schien es allerdings, als wenn bald die nötige Menge an reinem Kalkspat-Material nicht mehr geliefert werden könnte.

-1929 Attempts to purchase Iceland spar specimens from the Mineralogical Museum in Copenhagen continue, according to Garboe's 1961 book.

-1931 Coker and Filon, U.K. book ... Of late years the supply of Iceland spar has not been kept up, and of consequence new Nicol prisms are almost impossible to obtain....

-1931 Spencer and Jones, U.S.: Natural crystals, however, having a cross-section ... greater than 2 by 2 inches are so rare as to be practically unobtainable.

-1933 Smith, U.K. (at Hilger Ltd.) It is becoming increasingly difficult, for example, to obtain crystals of calcite which will enable first quality Nicol's prisms to be made with a maximum aperture of 1 in. diameter. In Smith's U.S. Patent it is also stated that...pieces of Iceland spar are no longer obtainable, at no matter what price, in sizes large enough to cut prisms of more than 50 mm. effective aperture.

-1933 Lyot, Fr. mentions in a 1941 paper that his ingenious monochromatic filter which needed 10 Nicol prisms (of Glazebrook-type, with 18-mm aperture) could not be constructed, faute du spath nécessaire pour les tailler. The filter was eventually made in 1938 with Polaroid sheets and in 1940 with Iceland spar.

-1935 Hardy, U.S. uses a specially designed calcite-glass Rochon prism instead of a Nicol prism in his new spectrophotometer, partly to economize on calcite.

-1936 Pollard, ?U.K. Usually, Nicol's prisms are much smaller than this [i.e. 4" aperture] and even moderately sized optically perfect crystals of the spar have already become scarce.

-1936 South African Geological Survey: Iceland spar has, in the past, been obtained almost exclusively from Iceland but owing to the falling off of the production of good optical grade material from that country and the increasing demand for the mineral, attention has been directed to other possible sources of supply.

-1938 Griggs, U.S. ... Iceland spar... difficult to obtain...

-1940 Kremers, U.S. Natural calcite of optical quality is scarce.....The acute shortage of calcite in the larger sizes has for several years stimulated the search for a proper substitute [he mentions sodium nitrate specifically].

-1948 Archard and Taylor, U.S. ... more recently [i.e. after 1936], owing to the shortage of calcite,...

- 1949 Evans, U.S. ... Unfortunately calcite is so difficult to obtain [in large sizes] that its general use in birefringent filters is probably impossible.

-1950 Schumann and Piller, D... Das Versiegen der natürlichen Vorkommen von optischem Kalkspat bildet seit langem eine beständige Sorge aller beteiligten Fachkreise...., [and] ...Kalkspatpolarisatoren, deren Beschaffung ständig schwieriger wird...

-1950 Hartshorne and Stuart, U.K. ... because large pieces of optical calcite are practically unobtainable nowadays, and...

-1953 Rinne-Berek, 12th ed. D... Da für optische Zwecke verwendbarer Kalkspat in den letzten Jahren immer seltener geworden ist...

-1961 Zirkl, A ... (summary of 1959 lecture) Hinzu kommt, dass die Beschaffung von Kalkspat immer schwieriger und kostspieliger wird, weil die Vorkommen in Island praktisch erschöpft sind.

-1967 Nester and Schroeder, U.S. Naturally occurring calcite of great purity has gradually become more difficult to obtain as great use has been made of this material in relatively recent years.

-1968 Ammann and Massey, U.S....large natural [calcite] crystals of good optical quality are becoming increasingly scarce.

Appendix 1B: A selection of some advertisements and other positive information on supplies of Iceland spar.

-1855 Griffin, U.K., advertisement: Iceland spar, unpolished. A variety of specimens from half an inch to three inches in diameter...Mr. Griffin has just received from Iceland a large supply of this beautiful mineral of a quality well suited for optical purposes.

-1857 Optikus Steeg, D. advertises in Annalen der Physik: Nicol'sche Prismen in allen Grössen, doppelbrechende Prismen von Doppelspath, Doppelspath-Platten, Polariskope etc. A similar advertisement from J.V. Albert appeared in 1859.

-1857 Lloyd's book, U.K. ... Iceland spar... is found in considerable masses, and in a state of great purity and transparency...

-1858 White, U.S. employs an Iceland spar prism from the calcite locality at Rossie, N.Y. in microscope observations.

-1866 Hessenberg, D.Die vielgesuchten Spaltungsstücke dieses edelsten und berühmtesten unter den Kalkspäthen haben seit langer Zeit zu Tausenden den Weg in die Werkstätten der Naturforschung gefunden... and he obtained some samples from A. Purgold. In 1872 he notes that Fr. Scharff had acquired a fresh supply, and in 1874 he obtains Iceland spar from Steeg. In an 1867 paper, E. Reusch reports having purchased crystals from the well-known W. Steeg (who later also prepared plates of calcite for the thermal conductivity studies by A. Tuchschmid 1883).

-1870 C.D. Ahrens, U.K. starts constructing large Nicol prisms, according to Cheshire 1906-07.

-1874 Baumgarten, D. obtained good samples from the gem dealer Henriques in Copenhagen.

-1874 Jardin, Fr.:...mais l'Islande seule le produit assez limpide pour que les experiences [i.e. optical experiments by physicists] puissant être faites avec succès. A similar expression is found in vol. II of Des Cloizeaux' Mineralogy in the same year, however also stating that one locality in New York State delivers cristaux quelquefois aussi limpides que du spath d'Islande.

-1876 Spottiswoode, U.K. mentions in a news item in The Times on 24 August ...that he had fortunately had the first pick of two cargoes of Iceland spar..., and that he among other things had a sphere of 5" diameter made.

-1878 Lutz, Fr. made an Iceland spar sphere of 46 mm diameter for a world exhibition in Paris, cf. a paper in J. Phys. Radium 6, 259-265, 1925.

-1880 Glazebrook, U.K.... Iceland spar, which could be obtained in large pieces with ease...; in the same year W. Huggins has triangular prisms made at the workshop of A. Hilger in London. Hilger adver-

tises in The Observatory 1880 that he possesses A large stock of perfectly pure Iceland spar, see also Fig. 25-1.

-1883 Report by Th. Thoroddsen in the National Archive: a small quantity was recovered at Helgustadir in 1882 and sold to the optician F. Schmidt in Berlin.

-1884 Feussner, D. ... mentions various types of Nicol prisms produced by Steeg & Reuter. In the following year they advertise these and calcite pieces in the 2nd edition of Groth's book on physical crystallography.

-1886 Danker, D. selects Iceland spar samples for his research from a grösserer Anzahl von Spaltungsstücken.

-1886-90 H. Struers, Dk, was selling the crystals recovered in the 1885 quarrying effort at Helgustadir, see e.g. Zeitschr. Deutsch. Geol. Gesellsch. 1888.

-1887 Pulfrich, D., uses Icelandic calcite from Steeg & Reuter.

-1890 Hoskyns-Abrahall (1892), U.K. states that Thor E. Tulinius is selling calcite for his father, who charges high prices to make his moderate stock last for a considerable time.

-1891 Dufet, Fr. discussing why measurements of the refractive indices have yielded very uniform results: ...il me paraît d'ailleurs peu vraisemblable que l'indice varie d'un échantillon à l'autre, étant donné que tout le spath utilisé pour l'optique provient du même gisement.

-1895, D At a meeting of the German Geological Society (Zeitschr. Deutsch. Geol. Gesellsch. 47, p. 223) two Nicol prisms made of calcite from limestone quarries at Auerbach were exhibited. It is said to be be found there "not too rarely" but in quality it is only comparable to second-class material from Iceland.

- Approx. 1895-99 The firm of B. Halle, D. offers a great selection of Iceland spar prisms and other objects (see Fig. 32-2). They also advertise these materials at the world exhibition in St. Louis 1904.

-1898 C.A. Niendorf, D., advertises in several issues of Deutsche Mechaniker-Zeitung that he sells Icelandic Doppelspath.

-1901 Hinrichsen, D. acquires grössere Menge von isl. Doppelspat from Schmidt & Haensch for his atomic-weight determinations.

-1901-02 The firm of A. Jobin, Fr., advertises in a Syndicat des Constructeurs - catalog, a 60° prism of Iceland spar for astronomical work, with side length 60 mm and height 80 mm.

-1903 Pockels, D. obtains calcite prisms from Steeg & Reuter for measurements of pressure effects.

-1905 4th edition of Groth's crystallography, D.: advertisement from Steeg & Reuter offering all types of polarizing prisms; R. Fuess and W. Apel also advertise Nicol prisms.

-1909 Rose, U.S.?, on chemical analyses:...Iceland spar is pure and easily obtained,....

-1921 Ingersoll, U.S. in a description of his glarimeter: ...the required optical parts – Wollaston doubleimage prism, nicol and lens – could be fairly readily obtained. A number of these glarimeters have been made up [for use] in various paper mills,...

-1922 Tutton, U.K.: There is, happily, plenty of smaller calc-spar constantly being discovered [he does not state where], so that Nicol prisms for all ordinary crystallographic apparatus and the polarising microscope are more or less readily procured....Elsewhere in his book he states that the available material is adequate for all but projection purposes.

-1925 Schütz, D. and 1927 Kellström, Sweden, use calcite from Steeg & Reuter.

-1928 Schulz, D. in Wien-Harms: Die amerikanischen Vorkommen bei Cedarville (Kalifornien) und Graycliff (Montana) liefern mit wenigen Ausnahmen nur minderwertige...Kristalle. Weitere wenigstens bedingt brauchbaren Doppelspat ergebende Fundstellen sind bekannt geworden in Südafrika, in Brasilien, sowie neuerdings im Gebiet der russischen Sowjetrepubliken.

-1936 A catalog of educational laboratory apparatus in physics from H. Struers Chemiske Laboratorium, Dk, offers Nicol prisms and calcite of various sizes on special order.

-1937 Hughes, U.S....Recent demand for Iceland spar has been supplied principally from South Africa (cf. Eiriksson 1943, who also mentions e.g. Spain and Siberia).

-1949 Dan E. Mayers in New York advertises in the journal Acta Crystallographica (vol. 2, p. 260) crystals of optical calcite, quartz and other substances for optical research.

-1950 K. Lambrecht, U.S., offers a variety of Nicol prisms in the Science journal, and later begins working new Iceland spar mines, see section 40.1. In a paper by W. Shurcliff on the polarization of light in Am. J. Physics 1962, readers are informed that Lambrecht's firm in Chicago sells many types of calcite polarizers.

-1978 Bennett and Bennett, U.S.: There is much more calcite wastage in making Glan-type prisms than in making the simpler Nicol types, so that, ...it was only following the recent discoveries of new calcite deposits that they became popular. (According to Bennett and Bennett these mines are in Mexico, Africa and Siberia).

Appendix 1C: A letter from G.G. Stokes

Lensfield Cottage, Cambridge, 6 July, 1886.

To his Excellency, the Minister for Iceland, Copenhagen.

Sir,

As I hold the office of president of the Royal Society of London, the oldest scientific society in this Kingdom, I hope your Excellency will excuse me for troubling you on a matter of scientific importance, possibly also of some slight commercial interest to your country.

This mineral known as calcite is much used by scientific men for instruments employed in investigations and lectures relating to polarised light. This mineral is common enough: but it has nowhere, to my knowledge, been found in large, clear, regularly crystallized masses except in Iceland, from which circumstance it is otherwise called Iceland spar. It is from Iceland that the supply of this material used by opticians has been exclusively derived.

Some years ago large blocks of this material used to be imported into this country from Iceland, and were freely purchased, and used by opticians. But for some time the supply has altogether ceased: the stock in hand has been used up, and opticians try in vain to get more. I feared that the mines were exhausted, but I am informed that the cause of the cessation of the supply is that the mines were purchased by Government and have not since been worked.

I imagine that they have been worked in a rather wasteful way, for the sake of quick returns, and probably at the time of the purchase there was a glut in the market: for though there is a steady demand for the material it is not used in large quantities, and the consumption is slow. But I hear on all sides from opticians and scientific men of the impossibility of procuring the material now, so there can be no doubt that there would be a demand for it if it could be supplied.

The magnificence of the blocks I used to see some years ago leads me to suspect that among the heaps of rubbish about the mine there may be many small pieces which were not thought worth collecting, nor were they perhaps when the market was glutted. But when I mention that a piece an inch or an inch and a half long and say three eighths of an inch in diameter would suffice to make a prism which when made could be sold for perhaps 8 shillings, even when there was still spar to be had in the market, it will be seen that even small pieces, comparatively speaking, have their value: and the collection of these, if such there be lying about, might help to defray the cost of re-opening the mine.

I enclose a translation of this letter into Icelandic, which my friend Mr. Magnússon has kindly undertaken to make.

I have the honour of being your Excellency's obedient servant

G.G. Stokes (sign.)

Appendix 2: A list of chemical analyses of Iceland spar from Iceland and elsewhere

Note: I have not obtained copies of all existing analyses, e.g. that of Klaproth (1788).

Fourcroy and Vauquelin (1804) Iceland spar from Iceland. No data given on impurities. Weight loss in acid was 43-44% (the correct value is 44 for CO_2).

Bucholz (1805) Transparent Doppelspath from Iceland: Calcium oxide 56.5%, carbon dioxide 43.0%, water 0.5%.

Thenard and Biot (1807) Chaux carbonatée rhombohedral: water 0.63%. They do not state the where the sample came from but Malus (1811, p. 306) and Des Cloizeaux (1874, bls. 113) indicate that it was spath d'Islande.

Stromeyer (1814) Iceland spar from Iceland: manganese oxide and a trace of iron, total 0.15%. Andreasberg in Germany: 0.36% of the same, and 0.10% water.

Dumas (1842) Icelandic Iceland spar: silica 0.013%, iron oxide 0.017%, traces of manganese and magnesium. Calcium carbonate 99.97%.

Erdmann and Marchand (1842, 1844) Iceland spar (very pure pieces): silica 0.014%, magnesium oxide 0.005%, iron oxide 0.020%, trace of potassium or sulfur. Calcium carbonate 99.96%. In the 1844 paper it is not clearly stated whether the material is from Iceland, but one sample yields 0.001% silica, and a total of 0.035% of aluminium-, iron- and manganese oxides. Another sample contained 0.049% of the same impurities.

Stas (1892) Spath d'Islande contains silica (one part in seven or eight thousand) and a trace of lithium.

Thiele and Richter (1900) Samples of Doppelspat contain a little less than 0.02% impurities.

Hinrichsen (1901, 1902) Four measurements on pieces from Icelandic material: average 0.046% iron carbonate (or 0.032% hematite). A sample from Crimea gave similar results but it also might contain a little magnesium.

E. Marbach (doctoral dissertation in Leipzig 1913, referred to by Hintze 1930) One Icelandic sample contained only small traces of MgO. A sample from Cumberland in North England gave 0.13% MgO.

G.K. Almström (J. Prakt. Chemie 99, 1919) Uses a sample of clear Icelandic calcite to test a method of measuring water and carbon dioxide in minerals. Obtains 0.26% H_2O and 43.83% CO_2 (which should be 43.94% in pure carbonate).

F. Hastings Smyth and L.H. Adams (J. Am. Chem. Soc. 45, 1923): In their research on thermodynamic properties of the system CaO-CO₂ they use "natural Iceland spar" which is "found by analysis to be free from iron and manganese and contain less than 0.05% of MgO".

Foote and Bradley (1926) A sample of "transparent cleavage crystals of Iceland spar" from an undisclosed location, contained a small amount of iron carbonate corresponding to 0.04% of hematite. In Proc. Nat. Acad. Sci. 16, p. 214, F. Zwicky says: "It is known…that Iceland spar of the purest kind contains only 0.03% Fe_2O_3 and no other chemically measurable contaminations", possibly referring to this determination.

Bearden (1931b) Two Icelandic samples. Iron oxide 0.007% and 0.006%, manganese oxide 0.008% and 0.007%. Silica, aluminium- and magnesium oxides were not detected, calcium carbonate 99.98%. Similar compositions were found in samples from Montana, Spain and Argentina.

Ievins and Straumanis (1940), two Icelandic samples. Iron carbonate 0.005% in both, manganese carbonate 0.032% and 0.011%, zinc carbonate 0.001% in both, traces of magnesium- and barium karbonate,

strontium carbonate 0.01-0.05%, calcium carbonate 99.92 and 99.95%. A sample from Cumberland had less impurities, while others from Norway, Caucasus, Graz and Andreasberg had more. In a table in Zeitschr. Phys. 125, 1948 p. 179 the average content of calcium carbonate in an undisclosed number of samples determined by Straumanis is 99.95 of a total of 99.977%.

Bearden (1965), one Icelandic sample: manganese 0.006%, strontium 0.02%, other elements each less than 0.0005%. Six selected samples of unknown origin plus one from Mexico and one from South America gave similar amounts of impurities.

D. Habermann (Mineralogy & Petrology 76, 2002) measures the Mn content of a sample from Helgustadir to be 0.02%, and Fe 0.025%.

The book by Hintze (1930, bls. 2956-) lists analyses of calcite samples from many worldwide locations, containing up to 5% or even more of other compounds.

Appendix 3: A list of some published photographs and other illustrations from the Helgustadir quarry, to 1960.

An Atlas of detailed illustrations accompanying the report by P. Gaimard's 1836 expedition contains a drawing of the Helgustadir calcite outcrop and the brook running through it. This drawing was also published in vol. 2 of the collected works of Jónas Hallgrímsson 1989 and by Kristjansson (2003).

In display cases at the Mineralogical Museum in Copenhagen, there was (in the late 1990's at least) a drawing of the Helgustadir outcrop, possibly dating from Fr. Johnstrup's visit in 1871. It may not have been published.

A. Helland: A drawing from his 1881 trip accompanied his paper on the geology of Iceland, published in Archiv Math. Naturv. 9, 1884.

Th. Thoroddsen: A drawing of Iceland spar veins made in 1882 appeared in his papers in 1890 and was copied e.g. by Merrill (1905) and Dammer and Tietze (1913). The originals of three drawings are enclosed with Thoroddsen's report dated 8 Nov 1882 in the National Archive.

H. Labonne: A photograph of two marines sitting in the quarry in 1887 was published in the book "Islande en Vue" by Æsa Sigurjónsdóttir, Reykjavik 2000. A part of it was also included in Kristjansson (2003).

F.W.W. Howell: A drawing of the mine and its immediate surroundings is reproduced on p. 50 of his book "Icelandic Drawings with Pen and Pencil" published in 1893. Howell travelled here in 1890-91 but the drawing may have been made by someone else.

C. Küchler: In his book "Wüstenritte und Vulkanbesteigungen auf Island" published in 1909, there is a photo of three miners. It also appeared in vol. 2 of the historical account "Eskja" by E.B. Sigurdsson, published at Eskifjördur in 1983, and by Kristjansson (2003). Küchler may have obtained it from earlier travellers such as D. Bruun.

H.H. Eiriksson: Drawings in his paper in Transact. Inst. Mining Eng. 1920, one reproduced in this report and by Kristjansson (2003); he also publishes drawings and three photographs in J. Soc. Eng. Iceland 1922. Better drawings in scale 1:500 dated 1921 are in the National Archive (Government Documents II, Journal 6 no. 385).

E. Consemüller: A photograph probably taken in July of 1924 is in vol. 2 of Kuhn and Prinz (2003), p. 140. It is reproduced in the 2005 yearbook of the Iceland Touring Association by H. Guttormsson, along with Labonne's photo and a photo of the entrance of the 1921 tunnel.

J.V. Kaspar's group, 1938 and 1947: Several photos appear in his 1940 report and are reproduced in its 1960 version.

S. Einarsson: In his geographical account of the southern part of the eastern fjords of Iceland in the 1955 yearbook of the Iceland Touring Association, there is an undated photo by T. Josepsson of a man standing at the end of the 1921 tunnel.

Appendix 4: A compilation of some relations between individuals named in this account

The present account mentions research by a very large number of scientists. The names of some may be familiar to say petrologists or physicists, while most of the readers have not heard of them before. One may ask: what connections were there between these scientists ? How was knowledge about Iceland spar, its properties, its use in instruments and so on disseminated ?

One particular example concerns the upsurge of interest in physical optics in France around 1807, which led to enormous progress in the field. How did it begin ? At least a partial answer to this question seems to be that the French read in scientific journals of research by W. Herschel, T. Young, W. H. Wollaston and others in England around 1800. Publications by R. Hauy may also have promoted the subject. In turn, how did Young and the others in England become interested in optics ? In Young's case it is evident from his writings that he was familiar with the work of older generations of scientists like C. Huygens and B. Martin. The reasons for Wollaston's entry into the field are less clear. Thus, Kipnis (1991, p. 73) says of him: "He had no particular interest in physical optics and came to the study of double refraction by a chance..." soon after he had invented his instrument (Wollaston 1802a) for measuring refractive indices and "came across Iceland spar". In contrast, Cheshire (1923) states that Wollaston's device was designed for the specific purpose of measuring Iceland spar. No doubt there are many similar examples where historical causes and effects might be investigated.

Another intriguing problem concerns the means by which Iceland spar found its way to various mineral dealers, museums, commercial instrument makers, and individual scientists in Europe. It would also be interesting to know whether the production of polarizing instruments was a major factor in the business of these instrument makers.

Below I list some pieces of about connections between some scientists, and connections of some of them to Iceland, that I have come across. This information is somewhat haphazard and probably not always of significance, but it is recorded here to give a glimpse of the intricate network of relations which may have aided the spread of knowledge about Iceland spar and polarized light.

F. Arago who was one of France's most notable scientists, was acquainted with T. Young and visited him in 1816 after the Napoleonic wars. Arago was also a friend of A. v. Humboldt who lived in Paris most of the time between 1804 and 1827 (for a while in the same house as Arago, in the same street as E. Malus). Along with J.B. Biot, the chemist L.J.Thenard and others they were members of a small but distinguished society of scientists, Société d'Arcueil. It is named after a small village where the astronomer and mathematician P.S. de Laplace and the chemist C.L. Berthollet were neighbors.

G. Rose studied mineralogy with C.S. Weiss who introduced Hauy's crystallography to German scientists. E. Mitscherlich who had studied chemistry with F. Stromeyer (who had written on the composition of Iceland spar) was instructed in crystallography by Rose in 1818, leading to Mitscherlich's work on isomorphism. Mitscherlich also was in contact with A. Fresnel and other French and British pioneers. Another student of Weiss was F.E. Neumann who around 1825 made important contributions to geometrical crystallography and later wrote major papers on optics (especially polarized light) and elasticity. H.W. Dove in Berlin whose publications mostly deal with meteorology (including an 1840 paper on data collected by a physician in Iceland) was also very active in crystallography and optics which he may have learned from Neumann at Königsberg in 1828-29.

v. Humboldt began his expedition to Siberia in 1829, accompanied by C.G. Ehrenberg and the above G. Rose. Some 15 years later, Ehrenberg published results on microscopic animals in Icelandic soil samples, collected by F.A.L. Thienemann (who had visited the Helgustadir quarry in 1820-21). Ehrenberg also

studied tephra from the 1845 Hekla eruption which he had obtained through F. Magnusson (an Icelandic archivist) and J.G. Forchhammer in Copenhagen as well as from R. Bunsen. Rose was the first to describe the zeolite epistilbite (in 1826, probably Icelandic specimens) and much later wrote a paper on Iceland spar (Rose 1868). P. Groth who published well-known books on crystallography, was Rose's student.

H.C. Örsted, famous for discovering the magnetic effects of electrical currents in 1820, probably did not himself write anything on Iceland spar. As noted in chapter 14 however, he helped other scientists to obtain crystals from Iceland in at least a few instances. Some correspondence between Örsted and Icelanders is preserved in Danish archives (according to a list compiled by S. Kristjansson, accessible at the National Library of Iceland) but I have not looked further into that matter. The geologist and chemist J.G. Forchhammer who was in charge of the mineralogical collection of the University of Copenhagen, may also have been an intermediary in the procurement of Iceland spar samples. He wrote papers on the geology and mineralogy of Iceland, but I have not found references to Iceland spar in his published correspondence.

T.J. Seebeck had started investigations in optics when the publications of E.L. Malus directed his interests towards polarized light around 1810, see section 7.3. He wrote papers e.g. on the effects of pressure on the optical properties of glass, but he is best known for his discovery of thermoelectricity (the Seebeck effect), later used by M. Melloni in research on infra-red radiation. His son, A. Seebeck, studied the reflection of light from Iceland spar but then turned to other fields of science.

As noted above, the Scotsman D. Brewster obtained in at least one case several samples of Iceland spar from his compatriot G.S. Mackenzie who had travelled in Iceland in 1810. They were without doubt acquainted, as Mackenzie wrote papers on various topics in the journals published by Brewster. According to Brewster's papers and a letter to H.C. Örsted, he also obtained mineral specimens from the banker T. Allan and his son R. Allan in Edinburgh. The Austrian W. Haidinger catalogued T. Allan's large mineral collection around 1825, and the Dictionary of Scientific Biography states that Haidinger's interest in mineralogy was due to his contacts with Brewster. F. Zirkel stayed for a year with Haidinger in Vienna around 1862 (according to a 1912 obituary of Zirkel), when working on his pioneering petrographic study on thin sections of rocks from Iceland and elsewhere. Among younger scientists in Vienna probably also influenced by Haidinger, were J. Grailich, V. v. Lang and G. Tschermak.

Irish scientists carried out valuable research in optics, electromagnetism and related fields in much of the 19th century. Some of them, like G.G. Stokes and W. Thomson (Lord Kelvin), moved to England or Scotland at a young age or worked there for a long while. From the names in the present list of references we may mention J.H. Jellett, J. MacCullagh, H. Lloyd, W.R. Hamilton and J. Tyndall, with later arrivals on the scene including G.F. FitzGerald, W.N. Hartley, T. Preston and J. Larmor.

J.D. Forbes who investigated thermal radiation, P. Kelland and W. Nicol were on the Edinburgh University teaching staff while J.C. Maxwell was a student there, and Maxwell also got to know G.G. Stokes during his studies in Cambridge. Nicol made a large number of Nicol prisms over a period of at least 20 years (see Swan 1849), and the instrument-makers Bryson in Edinburgh whom he knew well, also made Iceland spar prisms for J.H. Jellett and others. A. Bryson visited Iceland in 1862. W. Sang was Nicol's brother-in-law.

Among the teachers of L. Pasteur were the famous chemist J.B. Dumas, who used Iceland spar in his analytical work, and G. Delafosse who had studied with Hauy. Pasteur's interest in polarized light and optical activity may have been stimulated by his reading papers published around 1844 by E. Mitscherlich and J.B. Biot. Biot who had a high standing in French science, was just at that time arranging for the young mineralogist A. Des Cloizeaux to be sent to Iceland in 1845 to inspect the Helgustadir quarry. Des Cloizeaux also visited Iceland in the following year with R. Bunsen and others. H. Rosenbusch who was Bunsen's student, often mentions Icelandic rocks in his writings. R. Fuess who established an optics workshop in 1865, entered into the field of polarized light by producing microscopes for P. Groth around 1870, later for Rosenbusch. He became one of the major suppliers of petrographic microscopes and related optical equipment. C. Leiss who was for a long time employed by Fuess, set up his own business in 1921.

J.B. Soleil who was one of the main producers of optical instruments in France, inherited a workshop from his father who had from around 1820 manufactured large lenses for lighthouses (invented by A. Fresnel). The younger Soleil who worked for Fresnel, F. Arago, J. Babinet, F. Rudberg in Sweden, J. Nör-

renberg in Germany and others, published papers on his instruments in scientific journals to about 1850. At that time the workshop was divided, part of it being run until 1886 by J.B. Soleil's son-in-law J. Duboscq, then by Ph. Pellin. The other part was directed by H. Soleil and later by Duboscq's son-in-law L. Laurent, then by A. Jobin from 1892 and later by G. Yvon. Both workshops were very well known among scientists (see Brenni 1996), gaining their reputation in particular from polarimeters and other polarized-light equipment. One of the customers of the Soleil firms was H. de Senarmont, originally an engineer. His interest in optics was awakened by Fresnel's work, and he himself contributed various discoveries and inventions to the field.

Four generations of physicists named Becquerel wrote much about optics, including polarized light and crystal optics. The first was A.C. Becquerel, then E. Becquerel (whose brother A. Becquerel also used polarimeters in research on e.g. blood and foodstuffs, see Vernois and Becquerel 1853), H. Becquerel (whose first wife was J. Jamin's daughter) and J. Becquerel.

Among the students of F.E. Neumann who was mentioned above, were G. Kirchhoff, W. Voigt and L. Sohncke who made important contributions to crystallography and optics. His son C. Neumann who became one of Germany's better-known mathematicians, also wrote some papers on these subjects. Voigt's students included P. Drude, F. Pockels, R.A. Houstoun from the U.K. and P.G. Nutting from the U.S. Another of the many Americans doing their graduate studies in Germany, was D.B. Brace who studied optics with Kirchhoff and H. v. Helmholtz; some of Brace's students in turn continued his notable research into optical activity, such as F.J. Bates, A.Q. Tool and C.A. Skinner. One more German-educated American was F.E. Wright who studied with H. Rosenbusch and V. Goldschmidt; he travelled in Iceland in 1909.

Appendix 5: On connections between Iceland spar and Nobel prizes, 1901 to 1930 and later

These connections may be divided into five categories:

Discoveries dependent on Nicol prisms (made from Iceland spar) to a large extent:
 Emil Fischer, Chem. 1902 (synthesis of sugars and other organic compounds)
 Alfred Werner, Chem. 1913 (coordination bonds; see Kauffman 1968)

2. Discoveries where polarization played a considerable part:
-H.A. Lorentz and P. Zeeman, Phys. 1902 (Zeeman magneto-optic effect)
-J. Stark, Phys. 1919 (Stark electro-optic effect on spectral lines)
-C.V. Raman, Phys. 1930 (scattering of light, with change of wavelength).

- H. v. Euler, 1929 (fermentation and the nature of enzymes)

3. Discoveries where Nicol prisms or calcite crystals played some part:

The X-ray diffraction studies of W.H. and W.L. Bragg, Phys. 1915, M. Siegbahn, Phys. 1924, and A.H. Compton, Phys. 1927 employed calcite for accurate wavelength measurements. S. Arrhenius, Chem. 1903 and W. Ostwald, Chem. 1909, used polarimeters in some of their studies on chemical processes cited by the Nobel committee. O. Wallach, Chem. 1910, was awarded the prize for his work on terpene and camphor compounds from plants, including optical activity measurements. R. Willstätter, Chem. 1915, notes in his work on flower pigments (e.g. Willstätter 1915) that some of these exhibit high optical activity and rotatory dispersion. He also used polarimeters and other instruments with Nicol prisms in his research on cellulose, alkaloids, hydrolysis, and enzymes. The research of R.A. Zsigmondy, Chem. 1925, on colloids which included observations of light scattering and polarization, was preceded by his studies on colored glasses (see Zsigmondy 1901, Siedentopf and Zsigmondy 1903, Bachmann 1915). Zsigmondy's investigations in turn inspired investigations by J.B. Perrin, Phys. 1926, on the atomic structure of matter. H. Fischer, Chem. 1930, employed polarimeters in some of his work on haemin and chlorophyll (including specially constructed equipment to measure strongly colored solutions).

Several Nobel prizes in Chemistry have been awarded for discoveries which relied to some extent on results from previous work employing polarimeters, polarization photometers etc., such as those of A. v. Baeyer 1905, E. Buchner 1907, H.O. Wieland 1928, A. Windaus 1928, A. Harden 1929, and later W.N. Haworth 1937, R. Robinson 1947, C.F. and G.T. Cori also 1947, etc. As an example, a synthesis of the optically active compound ascorbic acid (Vitamin C), first carried out by Haworth, starts from sorbose or other sugars whose structure was clarified by E. Fischer. Fischer also studied nucleic acids and polypeptides, as mentioned in A. Kossel's lecture when receiving the 1910 Nobel prize in Physiology or Medicine for research on these compounds. The same applies to some Physics prizes, such as that of A. Kastler 1966: it was awarded for work which began around 1930 and concerned the polarization of resonance radiation, Raman-scattered light etc.

4. Nobel prize winners who published papers on work involving polarized light or calcite, but received their prizes for discoveries in different fields: prominent among these are W.C. Röntgen, Phys. 1901; H.

Becquerel, Phys. 1903; Rayleigh, Phys. 1904; J.J. Thomson, Phys. 1906; A.A. Michelson, Phys. 1907; R.A. Millikan, Phys. 1923.

Further: M. Born who received the Physics prize in 1954 wrote important papers and books on crystal optics, the Kerr effect, and optical activity (e.g. Born 1935). J.H. van't Hoff, Chem. 1901, put organic chemistry on a new foundation by his suggestion (Hoff 1874) that optical activity of asymmetric carboncontaining molecules is due to three-dimensional bonds. W. Ostwald, Chem. 1909, used the interference of polarized light in his later influential studies on color perception. W. Nernst, Chem. 1920, published a well-known paper with P. Drude on the polarization of standing light waves, and also papers where a Wanner pyrometer with Nicol prisms was employed. Both N. Bohr, Phys. 1922, J. Franck and G. Hertz, Phys. 1925, W. Heisenberg, Phys. 1932, and E. Fermi, Phys. 1938 wrote on polarized fluorescence, resonance radiation and related topics. A. Harden, Chem. 1929 investigated the resolving effect of microorganisms on optically inactive acids with A. McKenzie. R. Kuhn, Chem. 1938 did some of his prize-winning work on carotenes with a polarimeter, as well as research on mutarotation, fermentation and amygdalin derivatives in the early 1920s. He also wrote major review papers on optical activity. A. Gullstrand, Physiology or Medicine 1911, described some ophthalmological procedures where Nicol prisms played a part. O. Warburg, Physiology or Medicine 1931, began his scientific career with research on optically active organic compounds with E. Fischer. F.W. Aston, Chem. 1922, also began his career by writing on a similar topic with P.F. Frankland. P. Karrer, Chem. 1937, did his doctoral research on cobalt complexes under A. Werner and later carried out various studies (on polysaccharides, vitamins, tannins, glucosides, alkaloids etc.) where polarimeters were employed.

5. Finally it may be mentioned that some Nobel prize winners in the above or other fields had work on double refraction and polarized light done in their laboratories, perhaps in some cases without personally taking part in the execution of these projects. Thus, H. Kamerlingh Onnes, Phys. 1913, provided facilities e.g. for P. Zeeman and later J. Becquerel in their researches on magneto-optics, as well as for H. Keesom's investigations of light scattering in fluids. W. Hanle investigated polarized fluorescence under J. Franck, Phys. 1925. J. Hildebrand built a new version of the König spectrophotometer in W. Nernst's laboratory. E.O. Lawrence, Phys. 1939, assisted F.G. Dunnington and others in their work with electro-optic Kerr cells in the 1920's.

Appendix 6 in the Icelandic edition consists of translations of various expressions which appeared in the text in their original French or German. These all appear in English in the present English edition.

Appendix 7: A list of the names of some periodicals frequently quoted in the report

Am. Mineral. = American Mineralogist.

Am. J. Sci. = American Journal of Science.

Ann. Chim. = Annales de Chimie et de Physique, later split into two journals.

Ann. Phys. = Annalen der Physik (often attributed to J. Poggendorff or its other editors).

Arch. Sci. Phys. Nat. = Archives des Sciences Physiques et Naturelles, originally appearing as a Supplément to the Swiss journal Bibliothèque Universelle.

Astron. Nachr. = Astronomische Nachrichten.

Astrophys. J. = Astrophysical Journal, U.S.

Ber. Deutsch. Chem. Gesellsch. = Berichte der Deutschen Chemischen Gesellschaft zu Berlin.

British Assoc. Report = Report of the (Annual) Meeting of British Association for the Advancement of Science. Contains original articles, reviews, committee reports, etc.

Bull. Soc. Minéral. Fr. = Bulletin de la Société Minéralogique de France, later Bulletin de la Société Française de Minéralogie.

Comptes Rendus Acad. Sci. Fr. = Comptes Rendus Hebdomadaires des Séances de l'Académie Française des Sciences. Short papers; longer ones appeared as Mémoires.

C.R. Assoc. Fr. = Compte-Rendu, Session (Annuelle) de l'Association Française pour l'Avancement des Sciences.

Edinb. Philos. J. = Edinburgh Philosophical Journal (later split into the Edinburgh Journal of Science and Edinburgh New Philosophical Journal).

J. Am. Chem. Soc. = Journal of the American Chemical Society.

J. Biol. Chem. = Journal of Biological Chemistry, U.S.

J. Chem. Soc. = Journal of the Chemical Society, originally: ...of London. For some decades, its oddnumbered volumes were called Transactions and contained articles, while even-numbered volumes contained abstracts of work published elsewhere.

J. Pharm. = Journal de Pharmacie et de Chimie.

J. Prakt. Chemie = (O.L. Erdmann's) Journal für Praktische Chemie.

Liebig's Ann. Chemie = (Justus von Liebig's) Annalen der Chemie und Pharmacie.

Mineral. Mag. = Mineralogical Magazine, British.

Mineral. Petrogr. Mitt. = (G. Tschermak's) Mineralogische und Petrographische Mitteilungen, Austrian.

Naturwissensch. = Die Naturwissenschaften, preceded by the Naturwissenschaftliche Rundschau.

N. Jahrb. Mineral. = Neues Jahrbuch für Mineralogie, Geologie und Paläontologie.

Phil. Mag. = Philosophical Magazine. British, mostly on physics.

Phil. Trans. Royal Soc. = Philosophical Transactions of the Royal Society of London.

Phys. Rev. = The Physical Review, published by the American Physical Society.

Phys. Zeitschr. = Physikalische Zeitschrift.

Proc. Nat. Acad. Sci. = Proceedings of the National Academy of Sciences of the U.S.

Proc. Royal Soc. = Proceedings of the Royal Society of London. To begin with, it mostly contained abstracts of papers later appearing in the Philosophical Transactions.

Rev. Gén. Sci. = Revue Générale des Sciences Pures et Appliquées.

Rev. Sci. = Revue Scientifique (originally: Revue des Cours Scientifiques).

Sitzungsber. Akad. Wiss. Berlin = Sitzungsberichte der Königlich Preussischen Akademie der Wissenschaften zu Berlin (previously Monatsberichte, and still earlier Berichte). Large papers were published in a series of Abhandlungen.

Sitzungsber. Akad. Wiss. Wien = Sitzungsberichte der Kaiserlichen Akademie der Wissenschaften in Wien, Mathematisch-Naturwissenschaftliche Klasse. Larger publications often appeared in the Academy's Denkschriften.

Zeitschr. Krystallogr. = Zeitschrift für Krystallographie (originally: ...und Mineralogie).

Zeitschr. Phys. = Zeitschrift für Physik.

Zeitschr. Physikal. Chemie = Zeitschrift für Physikalische Chemie.

Zeitschr. Physiol. Chemie = (F. Hoppe-Seyler's) Zeitschrift für Physiologische Chemie.

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