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# Cross Sections and Rate Coefficients for Collisions of Electrons with Atomic and Molecular Fluorine

by

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RH-25-99

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2nd December 1999

## Abstract

An overview of the current status in electron collision cross sections for the excitation and ionization of molecular and atomic fluorine by electron impact is presented. The available data is used to calculate the rate coefficients for electron impact collision reactions by assuming Maxwellian electron energy distribution. The data presented will be useful in modeling of the plasma chemistry and electrical properties of low-pressure plasma discharges for materials processing.

## 1 Introduction

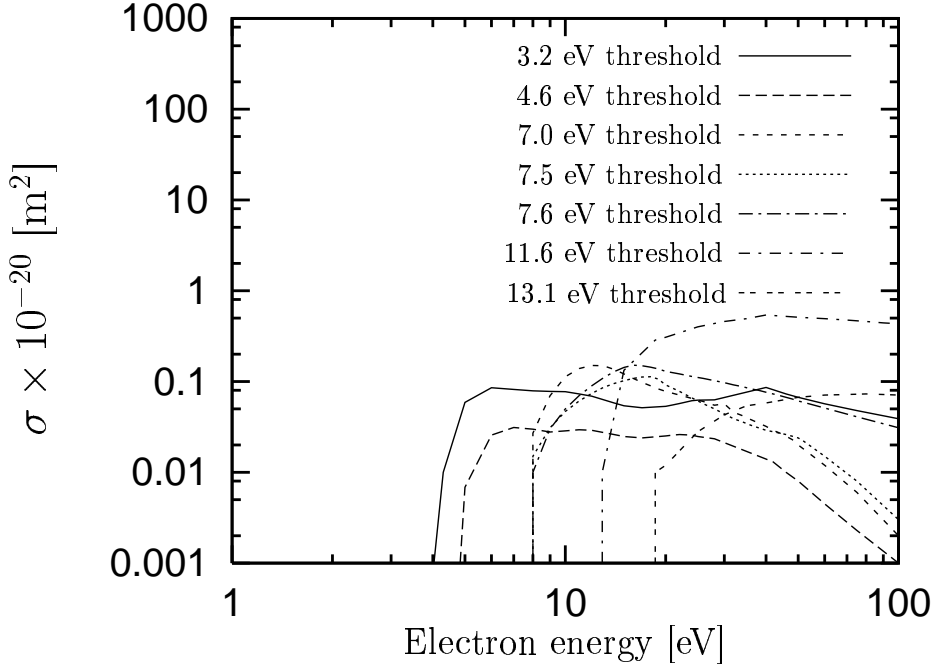
Electron-impact cross sections of fluorine gases are needed as input data for calculations of the chemical and electrical properties of processing plasmas and rare-gas fluorine lasers. Electron collisions are the driving mechanism behind the plasma processes employed in the semiconductor industry. Gas discharges are maintained by electron-neutral ionization, and energy is lost in part by collisional energy losses such as ionization, excitation and elastic scattering. In molecular gasses, electron impact dissociation is the key player in much of the chemistry; as well as being an energy-loss-mechanism. Molecular fluorine gas, and its mixtures, are used in the microelectronic industry for materials processing such as etching of polysilicon, silicon oxide and aluminum [1]. Many plasma processes use fluorine atoms as the dominant etching species. The plasma feed mixtures include  $F_2$ ,  $CF_4$ ,  $CF_4/O_2$ ,  $SiF_4$ ,  $SF_6$ ,  $SF_6/O_2$ ,  $NF_3$  and  $ClF_3$ . The gas chemistry of these discharges, however, is poorly understood. The rate coefficients for electron impact collisions are required for modeling of such discharges.

Hayashi and Nimura [2] and later Morgan [3] used a combination of measured and theoretically calculated cross sections in an attempt to assemble a cross section set for electron impact on molecular fluorine. This current work is an attempt to assemble a complete cross section set for both molecular and atomic fluorine. The high chemical activity of molecular fluorine makes it difficult to obtain reliable values of the electron-impact cross sections. Of the available cross sections for electron-impact on atomic and molecular fluorine only the electron attachment in molecular fluorine and the ionization of molecular and atomic fluorine has been measured experimentally. The electron excitation cross sections as well as elastic cross sections for both molecular and atomic fluorine are taken from theoretical estimates.

From the cross sections the rate coefficient are calculated assuming a Maxwellian electron energy distribution. Furthermore, an overview is given of the available rate coefficients for reactions of fluorine atoms, molecules and ions.

## 2 Molecular Fluorine

Because of its large attachment cross section, molecular fluorine has been the negative ion of choice in rare gas fluoride lasers [3]. Fluorine is a shell-closed molecule with a  $^1\Sigma_g^+$  ground state. The fluorine molecule is known to dissociate into neutral fluorine atoms via various electronic state excitations. Electron-impact dissociation of the fluorine molecule is considered to take place via two classes of excited electronic states, purely repulsive or strongly perturbed by Rydberg valence-ionic potential energy curve crossings [4]. The first class consist of all the 11 valence states of molecular fluorine that dissociate into two ground state fluorine atoms. The 11 valence excitation processes can be divided into one-electron molecular orbital transition that characterize the five valence states,  $^1,^3\Pi_u$ ,  $^1,^3\Pi_g$ ,  $^3\Sigma_u^+$ , and two-electron molecular orbital transitions,  $^3\Sigma_g^+$ ,  $^3\Sigma_u^-$ ,  $^1\Sigma_g^+$ ,  $^1\Sigma_u^-$ ,  $^1\Delta_g$ ,  $^3\Sigma_u^+(2)$ . The transitions in the latter group are considerably weaker than the former group. The perturbed Rydberg state is expected to dissociate into the fragments  $F^-(^1S)$  and  $F^+(^1D)$  [4].

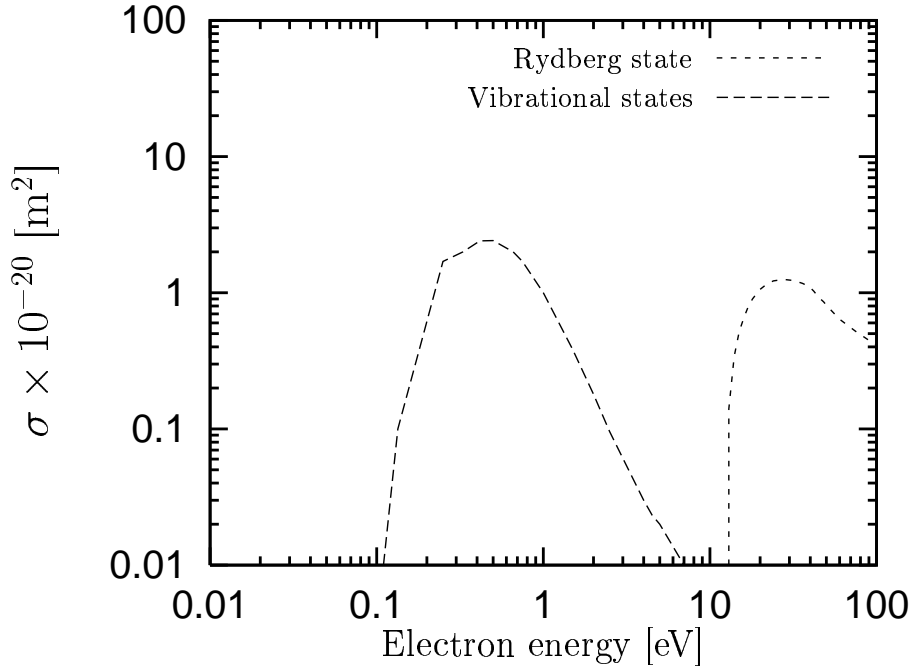


**Figure 1:** The electron-impact excitation cross section of  $F_2$  as a function of the electron impact energy. The cross section for the excitation of the one-electron states,  $^3\Pi_u$ ,  $^1\Pi_u$ ,  $^3\Pi_g$ ,  $^1\Pi_g$  and  $^3\Sigma_u^+$ , with threshold 3.32, 4.64, 7.0, 7.5 and 7.6 eV, respectively, excitation to the  $C^1\Sigma_u$  and  $H^1\Pi_u$  states, with threshold 11.56 eV and 13.08 eV respectively.

## 2.1 Electron-Impact Excitation Cross Sections

The two lowest excited states are the dissociative  $^3\Pi_u$  and  $^1\Pi_u$  states, with threshold energies 3.32 eV and 4.64 eV respectively. The values for the lowest electron excitation level  $a^3\Pi_u$  (threshold 3.16 eV) were calculated theoretically by Lengsfeld and Rescigno [5] and are shown in figure 1. The cross section is roughly 2.5 times smaller than the other theoretical value by Fliflet et al. [6]. The cross section for the dissociative level  $A^1\Pi_u$  (threshold 4.34 eV) calculated theoretically by Lengsfeld and Rescigno [5] is shown in figure 1 as well. The cross section for the excitation of the three one-electron states,  $^3\Pi_g$ ,  $^1\Pi_g$ ,  $^3\Sigma_u^+$ , with threshold 7.0, 7.5 and 7.6 eV, respectively, are taken from an estimate by Cartwright et al. [4]. Theoretical estimation of the cross sections for the electron impact excitation for the two levels  $C^1\Sigma_u$  (threshold 11.57 eV) and  $H^1\Pi_u$  (threshold 13.08 eV) is taken from Hazi [7].

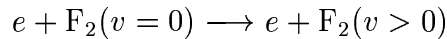
Features in the  $F_2$  electron energy loss spectra at 12.87 eV and 14.13 – 14.27 eV are proposed by Cartwright et al. [4] to be due to very strongly avoided crossing involving the Rydberg-valence-ionic electronic states of  $^1\Sigma_u^+$  symmetry. They also give an estimate of the electron-impact cross section for the process and suggest that the



**Figure 2:** The electron-impact excitation cross section of  $F_2$  as a function of the electron impact energy. The perturbed Rydberg state and vibrational states.

excitation to the perturbed Rydberg state, 12.87 eV, results in immediate dissociation into the fragments  $F^-$  and  $F^+$ .

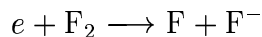
A theoretical evaluation of the vibrational excitation cross sections



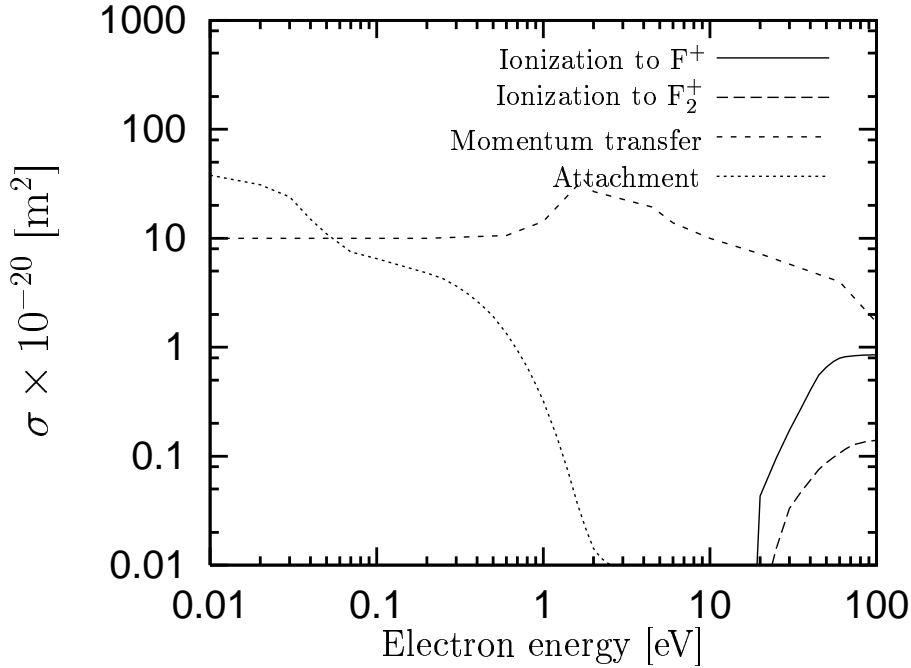
is given by Hall [8]. The total vibration cross section is shown in figure 2. The threshold energy for the vibrational excitation is 0.11 eV [2].

## 2.2 Electron Attachment in $F_2$

The beam measurements of the dissociative attachment,



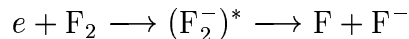
for molecular fluorine by Chantry [9] show an increasing cross section with decreasing electron energy. This is consistent with other measurements [10]. For electron energy above 0.25 eV the agreement is very good with the theoretical results of Hall [8]. More recent theoretical ab initio calculation by Hazi et al. [11] agrees reasonably with the data for electron energy above 0.15 eV. However, theoretical calculations cannot account for



**Figure 3:** The electron-impact momentum transfer cross section, the electron-impact ionization cross section for creation of  $F_2^+$  and  $F^+$  and the electron attachment cross section of molecular fluorine as a function of the electron impact energy.

the cross-section shape below 0.15 eV as shown in figure 3. The cross section for direct dissociative attachment via the  ${}^2\Sigma_u^+$   $F_2^-$  state must go to zero at zero electron energy so this process alone cannot account for the cross-section shape below 0.15 eV as shown in figure 3.

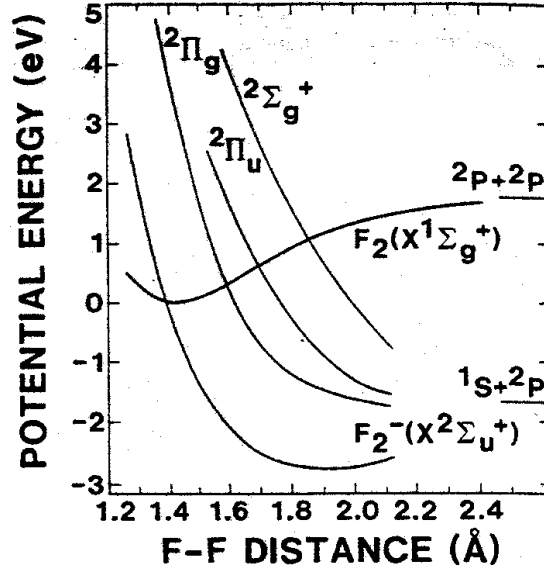
In most cases negative ion proceeds by electron capture by a neutral molecule, forming a intermediate resonant negative ion state. The dominant mechanism for producing stable negative ions from an intermediate  $(F_2^-)^*$  state is



The dissociation limit  $F^-({}^1S) + F({}^2P)$  gives rise to four molecular electronic states, the ground state of  $F_2^-$ ,  ${}^2\Sigma_u^+$ , and mostly repulsive states  ${}^2\Pi_g$ ,  ${}^2\Pi_u$  and  ${}^2\Sigma_g^+$ . Potential energy curves for the states of  $F_2$  and  $F_2^-$  are shown in figure 4.

### 2.3 Electron-Impact Momentum Transfer

The elastic transfer cross section for molecular fluorine shown in figure 3 is taken from Hayashi and Nimura [2] who base it on theoretical estimation of the elastic scattering

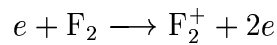


**Figure 4:** Potential energy curves for  $F_2$  and  $F_2^-$ . From A. Chutjian and S. H. Alajajian, *Physical Review A* **35** (1987) 4512.

of electron by fluorine molecule given by Schneider and Hay [12] and Rescigno et al. [13]. The more recent calculations by Morgan and Noble [14] are in good agreement with the result of Schneider and Hay [12]. Their calculations in the static exchange approximation presented here show a resonance around 2.2 eV. Hayashi and Nimura [2] make the assumption that the total momentum transfer cross section is equal to the elastic collision cross section. The values of the momentum transfer cross section for energies above 14 eV are assumed to be equal to the values for molecular nitrogen.

## 2.4 Electron-Impact Ionization Cross Section

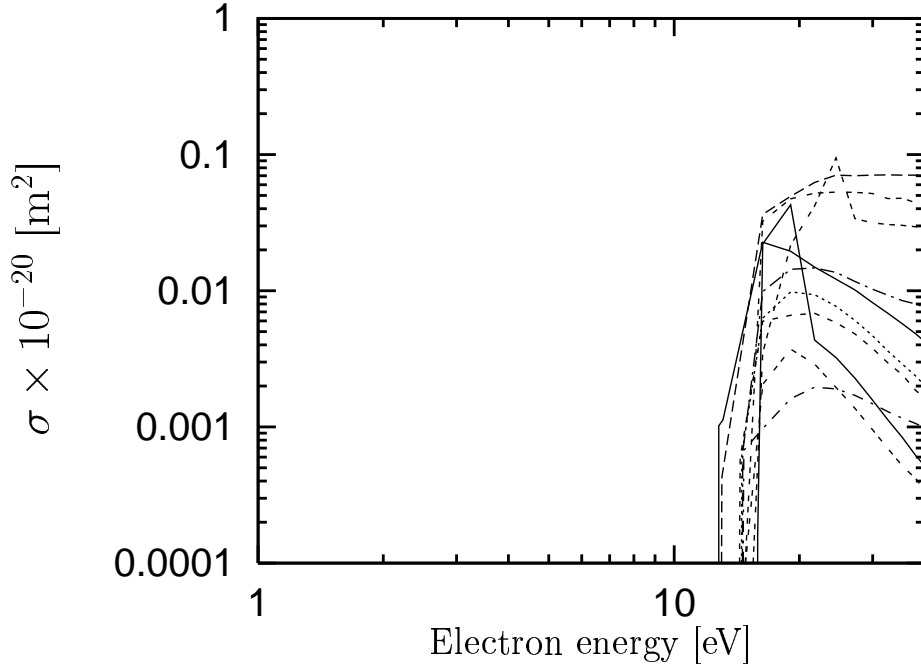
The cross section for ionization of the fluorine molecule



was measured in a crossed beam apparatus by Rao and Srivastava [15] and is shown in figure 3. They determined the ionization thresholds for  $F_2^+$  and  $F^+$  to be 15.7 eV and 15.5 eV, respectively.  $F^+$  ions are generated at lower energy than are  $F_2^+$  ions by the process of polar dissociation



through repulsive states lying between the ionization continuum of  $F_2^+$ . For simplification we assign the ionization cross section for the production of  $(F^+ + F_2^{2+})$ , from electron impact on  $F_2$  molecule, measured by Rao and Srivastav [15] to represent only the above polar dissociation and neglect the production of  $F_2^{2+}$ .



**Figure 5:** The electron-impact excitation cross sections, for the excitation to the 10 lowest excited states of atomic fluorine, calculated from the estimations of the the electron-impact collision strength.

### 3 Atomic Fluorine

The ground state of the fluorine atom  $1s^2 2s^2 2p^5 \ ^2P_{3/2}^o$  is specified to comprise two closely spaced levels  $^2P_2$  and  $^2P_1$ . The  $^2P_1$  level lies 0.05 eV above the  $^2P_2$  level [16]. The statistical weight, or the mean number of occupied states for the ground state group of levels is thus  $\bar{g}_F = 8$ .

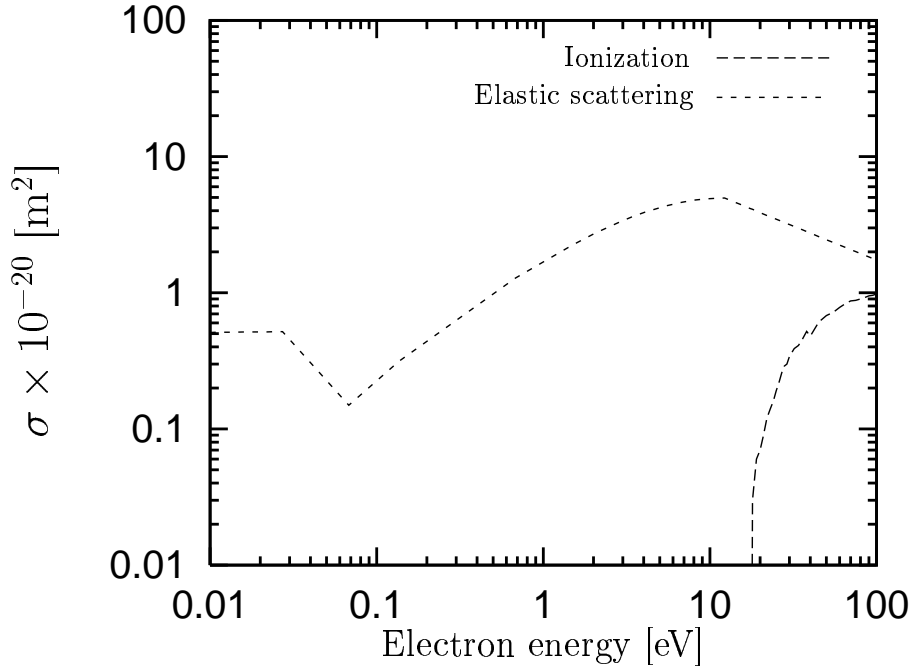
#### 3.1 Electron-Impact Excitation Cross Sections

Baliyan and Bhatia [17] give theoretical estimation of the electron-impact collision strength for the excitation of the lowest 10 excitation states of atomic fluorine for electron impact energy in the range 16.3 to 41 eV. Several other  $2p^4 3l$  states with threshold energy above 19 eV are neglected. The collision strength for the transition  $i \rightarrow f$  is given in terms of the corresponding excitation cross section  $\sigma_{if}$  by the equation

$$\Omega_{if} = g_i \mathcal{E} \sigma_{if} \quad (1)$$

where  $\mathcal{E}$  is the impact energy in Rydbergs,  $\sigma_{if}$  is the cross section in units of  $\pi a_o^2$ , and  $g_i$  the statistical weight of the lower (initial) level [18]. The electron-impact excitation





**Figure 6:** The cross section for elastic scattering of electrons by atomic fluorine and the ionization cross section of atomic fluorine as a function of the electron impact energy.

cross sections for the excitation to the 10 lowest excited states of atomic fluorine calculated from the estimations of the the electron-impact collision strength are shown in figure 5. To estimate the cross section next to the observed threshold energy the data was fitted to Thomson cross section.

### 3.2 Electron-Impact Momentum Transfer

The cross section for elastic scattering of electrons by atomic fluorine was calculated by Robb and Henry [19], in the range 2.7 – 27.2 meV. In the energy range 0.07 eV – 12.2 eV the elastic cross section is given by Robinson and Geltman [20]. It is not clear whether the minimum that appears in the curve around  $\sim 0.07$  eV is an artifact due to inconsistency in the calculation or a Ramsauer minimum. At electron impact energy above 12 eV the cross section is assumed to follow  $\sigma_{\text{elas}} \propto 1/\mathcal{E}$ . The elastic cross section for atomic fluorine is shown in figure 6.

### 3.3 Electron-Impact Ionization Cross Section

The electron-impact ionization cross section for atomic fluorine was measured by Hayes et al. [21] from threshold to 200 eV and is shown in figure 6. The absolute accuracy of

**Table 1:** Energy-loss reactions for fluorine molecule.

Reaction	Thresh. [eV]	Rate coefficient [m <sup>3</sup> /s]	Ref.
$e + \text{F}_2 \longrightarrow \text{F}_2(a^3\Pi_u) + e$	3.21	$k_{\text{exc1}} = 1.93 \times 10^{-15} \exp(-4.24/T_e)$	[4, 5]
$e + \text{F}_2 \longrightarrow \text{F}_2(A^1\Pi_u) + e$	4.34	$k_{\text{exc2}} = 7.36 \times 10^{-16} \exp(-4.71/T_e)$	[4, 5]
$e + \text{F}_2 \longrightarrow \text{F}_2(^3\Pi_g) + e$	7.0	$k_{\text{exc3}} = 3.14 \times 10^{-15} \exp(-7.30/T_e)$	[4]
$e + \text{F}_2 \longrightarrow \text{F}_2(^3\Sigma_u^+) + e$	7.5	$k_{\text{exc4}} = 2.62 \times 10^{-15} \exp(-8.10/T_e)$	[4]
$e + \text{F}_2 \longrightarrow \text{F}_2(^1\Pi_g) + e$	7.6	$k_{\text{exc5}} = 4.11 \times 10^{-15} \exp(-8.74/T_e)$	[4]
$e + \text{F}_2 \longrightarrow \text{F}_2(C^1\Sigma_u) + e$	11.57	$k_{\text{exc6}} = 1.80 \times 10^{-14} \exp(-14.12/T_e)$	[7]
$e + \text{F}_2 \longrightarrow \text{F}_2(H^1\Pi_u) + e$	13.08	$k_{\text{exc7}} = 2.13 \times 10^{-15} \exp(-18.22/T_e)$	[7]
$e + \text{F}_2 \longrightarrow \text{F}_2(\text{Rydberg}) + e$	12.87	$k_{\text{Ry}} = 4.78 \times 10^{-14} \exp(-13.37/T_e)$	[4]
$e + \text{F}_2 \longrightarrow \text{F}_2(\text{vib.}) + e$	0.11	$k_{\text{vib}} = 1.29 \times 10^{-16} \exp\left(\frac{12.73}{T_e} - \frac{17.46}{T_e^2} + \frac{8.24}{T_e^3}\right)$	[2, 8]
$e + \text{F}_2 \longrightarrow e + \text{F}_2(\text{mom.})$		$k_{\text{mom}} = 2.48 \times 10^{-13} \exp(-0.48/T_e)$	[2]

the data is  $\pm 20\%$ . The ionization potential for the fluorine atom is 17.42 eV [16].

## 4 Rate Coefficients

The rate coefficients for electron-impact collision reactions were obtained from integration of the electron collision cross section over an assumed Maxwellian electron energy distribution,

$$k_i = \left(\frac{2e}{m_e}\right)^{1/2} \int_0^\infty \sigma(\mathcal{E}) \mathcal{E} f(\mathcal{E}) d\mathcal{E} \quad (2)$$

where

$$f(\mathcal{E}) = \frac{2}{\sqrt{\pi}} \frac{1}{(eT_e)^{3/2}} \exp\left(-\frac{\mathcal{E}}{T_e}\right) \quad (3)$$

is the Maxwellian energy distribution function,  $T_e$  is the electron temperature in equivalent voltage units,  $\sigma$  is the collision cross section, and  $m_e$  is the mass of the electron. The rate coefficients are then fitted to the Arrhenius form

$$k = A \exp\left(-\frac{B}{T_e}\right) \quad (4)$$

over electron temperature range of 0.9 – 12 eV where  $A$  and  $B$  are constants.

The rate coefficients for electron impact excitation of the fluorine molecule calculated from the cross sections discussed in section 2.1 are listed in table 1. The rate coefficient for the dissociation of the fluorine molecule was calculated from the excitation cross sections for one-electron molecular orbital transitions,  $a^3\Pi_u$  (threshold 3.21 eV) [4, 5],  $A^1\Pi_u$  (threshold 4.34 eV) [4, 5],  $^3\Pi_g$  (threshold 7.0 eV) [4],  $^1\Pi_g$  (threshold 7.5

**Table 2:** Energy-loss reactions for atomic fluorine.

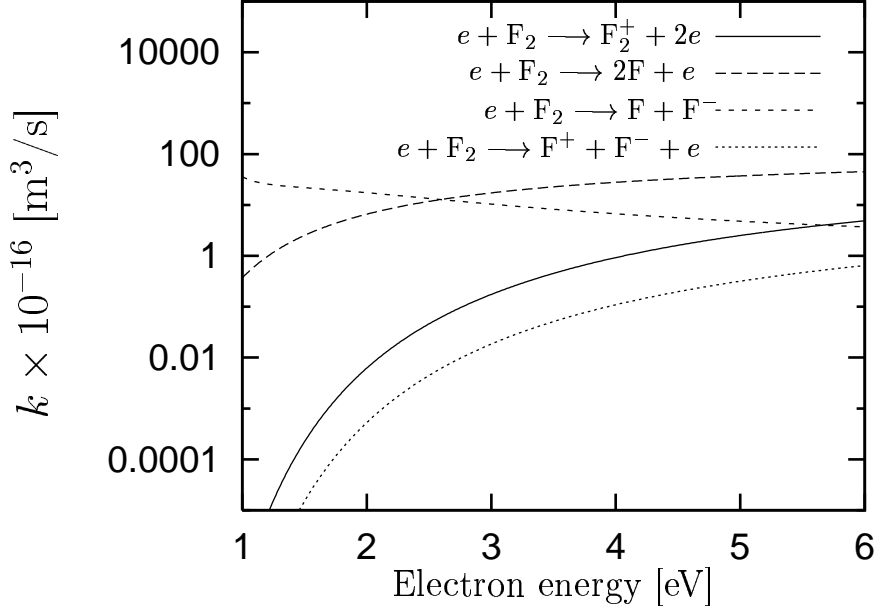
Reaction	Final state	Thresh. [eV]	Rate coefficient [m <sup>3</sup> /s]	Ref.
$e+F \rightarrow F^{1*} + e$	$2s^2 2p^4(^3P)3s^4P^e$	12.70	$k_{1*} = 6.07 \times 10^{-16} \exp(-12.87/T_e)$	[17]
$e+F \rightarrow F^{2*} + e$	$2s^2 2p^4(^3P)3s^2P^e$	12.99	$k_{2*} = 2.72 \times 10^{-15} \exp(-14.52/T_e)$	[17]
$e+F \rightarrow F^{3*} + e$	$2s^2 2p^4(^3P)3s^4P^o$	14.37	$k_{3*} = 2.28 \times 10^{-16} \exp(-13.77/T_e)$	[17]
$e+F \rightarrow F^{4*} + e$	$2s^2 2p^4(^3P)3s^4D^o$	14.51	$k_{4*} = 3.02 \times 10^{-16} \exp(-14.04/T_e)$	[17]
$e+F \rightarrow F^{5*} + e$	$2s^2 2p^4(^3P)3s^2D^o$	14.58	$k_{5*} = 5.31 \times 10^{-16} \exp(-14.32/T_e)$	[17]
$e+F \rightarrow F^{6*} + e$	$2s^2 2p^4(^3P)3s^2S^o$	14.67	$k_{6*} = 6.60 \times 10^{-17} \exp(-14.9/T_e)$	[17]
$e+F \rightarrow F^{7*} + e$	$2s^2 2p^4(^3P)3s^4S^o$	14.67	$k_{7*} = 9.07 \times 10^{-17} \exp(-13.96/T_e)$	[17]
$e+F \rightarrow F^{8*} + e$	$2s^2 2p^4(^3P)3s^2P^o$	14.74	$k_{8*} = 2.14 \times 10^{-15} \exp(-14.73/T_e)$	[17]
$e+F \rightarrow F^{9*} + e$	$2s^2 2p^4(^1D)3s^2D^e$	15.35	$k_{9*} = 1.98 \times 10^{-15} \exp(-16.67/T_e)$	[17]
$e+F \rightarrow F^{10*} + e$	$2s^2 2p^4(^3P)3s^4D^e$	15.87	$k_{10*} = 5.30 \times 10^{-16} \exp(-13.39/T_e)$	[17]
$e+F \rightarrow F+e$	momentum transfer		$k_{\text{mom}} = 1.15 \times 10^{-13} \left( \frac{-3.24}{T_e} + \frac{1.31}{T_e^2} \right)$	

eV) [4] and  $^3\Sigma_u^+$  (threshold 7.6 eV) [4]. The sum of these five excitation cross sections is assumed to correspond the cross section for dissociation of the fluorine molecule.

The rate coefficients for excitation of the fluorine atom are listed in table 2. The rate constants were calculated from the estimated collision strength given by Baliyan and Bhatia [17].

A reaction set for atomic and molecular fluorine along with rate coefficients is listed in table 3. The rate coefficients for reaction  $k_5$ , the recombination between  $F_2^+$  and  $F^-$  ions, reaction  $k_7$ , detachment of electrons from  $F^-$  ions, reaction  $k_{10}$ , associative detachment, and reaction  $k_5$ ,  $F_2^+$  ion recombination, are taken from the literature [22]. The rate coefficient for the associative detachment  $k_{10}$  was evaluated in the electron temperature range  $0.35 \leq T_e \leq 0.52$  eV. Hoffland et al. [22] measured the rate coefficient for the recombination between  $F_2^+$  and  $F^-$  ions,  $k_5$ . The rate coefficient for recombination of  $F^+$  and  $F^-$  is expected to be large and comparable to that for  $F_2^+$  and  $F^-$  so we assume  $k_6 = k_5 = 1.5 \times 10^{-14}$  m<sup>3</sup>/s. These rate coefficients have been used in simulating  $CF_4$  plasma for etching silicon [23] and modeling KrF lasers [24].

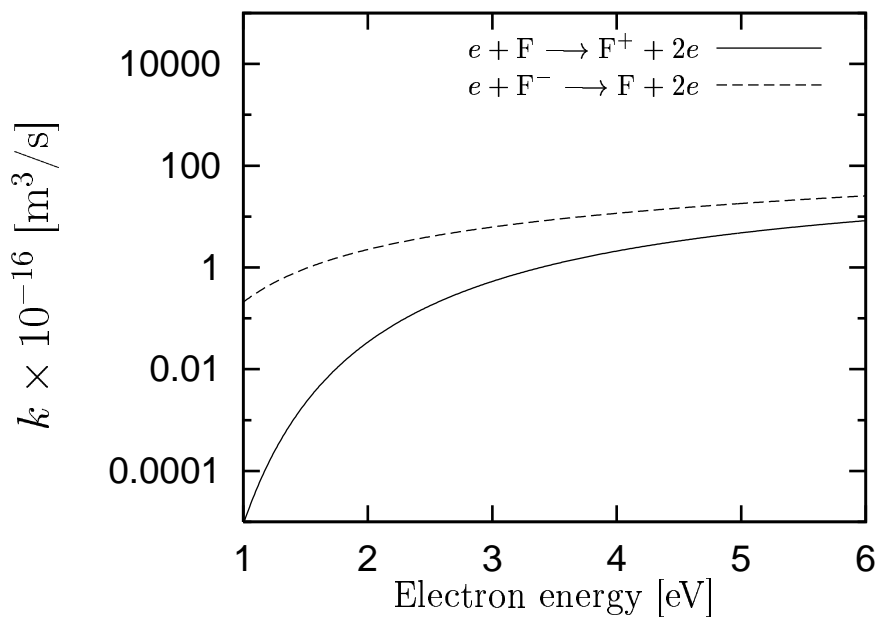
Figure 7 shows the rate coefficients for ionization to form  $F_2^+$ , dissociation, dissociative attachment and ionization to form  $F^+$  and  $F_2^{2+}$  by electron impact of the fluorine molecule. As expected for the fluorine molecule the rate coefficient for dissociative attachment is the dominant process at low electron impact energy. At around 2.5 eV dissociation of the molecular fluorine becomes the dominating process. Ionization to form the  $F_2^+$  ions dominates the production of  $F^+$  ions by electron impact of the fluorine molecule. Hansen et al. [25] show by LIF and emission measurements that



**Figure 7:** The rate coefficients for ionization to form  $F_2^+$ , dissociation, dissociative attachment and ionization to form  $F^+$  and  $F_2^{2+}$  by electron impact of molecular fluorine.

**Table 3:** A fluorine reaction set.

Reaction	Rate coefficient [ $m^3/s$ ]	Reference
$e+F_2 \longrightarrow F_2^+ + 2e$	$k_1 = 1.37 \times 10^{-14} \exp(-20.70/T_e)$	[15]
$e+F_2 \longrightarrow 2F + e$	$k_2 = 1.18 \times 10^{-14} \exp\left(\frac{-6.88}{T_e} + \frac{1.11}{T_e^2}\right)$	
$e+F_2 \longrightarrow F + F^-$	$k_3 = 6.26 \times 10^{-17} \exp\left(\frac{13.34}{T_e} - \frac{17.47}{T_e^2} + \frac{8.18}{T_e^3}\right)$	[9]
$e+F \longrightarrow F^+ + 2e$	$k_4 = 1.30 \times 10^{-14} \exp(-16.50/T_e)$	[21]
$F^- + F_2^+ \longrightarrow 3F$	$k_5 = 1.5 \times 10^{-14}$	[22]
$F^- + F^+ \longrightarrow 2F$	$k_6 = 1.5 \times 10^{-14}$	
$e+F^- \longrightarrow F + 2e$	$k_7 = 1.2 \times 10^{-16} T_e^{3/2} \left(2 + \frac{3.45}{T_e}\right) \exp(-3.45/T_e)$	[22]
$e+F_2 \longrightarrow F^+ + F^- + e$	$k_8 = 2.26 \times 10^{-15} \exp(-21.33/T_e)$	[15]
$e+F_2^+ \longrightarrow 2F$	$k_9 \leq 5 \times 10^{-14}$	[22]
$F + F^- \longrightarrow F_2 + e$	$k_{10} = 1.4 \times 10^{-16} \exp(-1.9/T_e)$	[22]



**Figure 8:** The rate coefficients for ionization of the fluorine atom by electron impact and electron impact detachment of the fluorine atom.

an excited fluorine atom,  $\text{F}^*$ , is not likely to be a significant participant in heterogeneous processes. At pressures as low as 100 mTorr  $\text{F}^*$  travels less than a mm before quenching.

## 5 Summary

An overview was given of the available cross sectional data for electron impact collisions of atomic and molecular fluorine. The available cross sections were used to calculate the rate coefficients for electron impact collisions assuming a Maxwellian electron energy distribution.

### Acknowledgements

This work was supported by the Icelandic Research Council.

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