Electron excitation rate coefficients for the nitrogen discharge

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Abstract

The rate coefficients for electron impact excitation, electron impact ionization and elastic scattering of electrons for nitrogen atom and nitrogen molecule are reviewed. We assume the electron energy distribution to be Maxwellian and the fit to the rate coefficient is valid in the range 1 – 7 eV. The rate coefficients are then used to evaluate the collisional energy loss per electron ion pair created.

1 Introduction

To describe discharge operation a noble gas such as argon is usually used Lieberman and Lichtenberg (1994). However, most process gases are molecular and electronegative. The volume averaged global model for low pressure high density discharges for noble gases (Lieberman and Gottscho, 1994) was extended to include molecular gases Lee et al. (Lee et al., 1994; Lee and Lieberman, 1995).

An important quantity used in the global model is the collisional energy loss per electron-ion pair created, $\mathcal{E}_{\rm c}(T_{\rm e})$ is defined as

$$\mathcal{E}_{\rm c} = \mathcal{E}_{\rm iz} + \sum_{\rm i} \mathcal{E}_{{\rm ex},i} \frac{k_{{\rm ex},i}}{k_{\rm iz}} + \frac{k_{\rm el}}{k_{\rm iz}} \frac{3m_{\rm e}}{m_{\rm i}} T_{\rm e} \tag{1}$$

where $\mathcal{E}_{\rm iz}$ is the ionization energy, $\mathcal{E}_{{\rm ex},i}$ is the threshold energy for the i-th excitation process, $k_{\rm iz}$ is the ionization rate coefficient, $k_{{\rm ex},i}$ is the rate coefficient for the i-th excited state and $k_{\rm el}$ is the elastic rate coefficient. Thus the terms on the right hand side account for the loss of electron energy due to ionization, excitation, and elastic (polarization) scattering against neutral atoms.

The main purpose of this work is to review the rate coefficients that are needed to evaluate the collisional energy loss per electron ion pair created for the nitrogen atom and the nitrogen molecule. Throughout the text we use the roman typeface symbol T for the voltage equivalent of the temperature.

2 Rate coefficient

The rate coefficient for an electron impact collision is obtained by integrating the cross sections over an assumed Maxwellian distribution

$$k = \langle \sigma(v) v \rangle_v = 4\pi \int_0^\infty \sigma(v) v^3 f(v) dv \tag{2}$$

where σ is the collision cross section, v the electron velocity and

$$f(v) = \left(\frac{m_{\rm e}}{2\pi e T_{\rm e}} \right)^{3/2} \exp\left(-\frac{m_{\rm e} v^2}{2eT_{\rm e}} \right) \tag{3}$$

is the Maxwellian velocity distribution, m_e is the electron mass, e is the electron charge and T_e is the electron temperature. Using the relationship for the velocity and kinetic energy of a particle

$$\mathcal{E} = \frac{m_e v^2}{2e} \tag{4}$$

we find

$$f(\mathcal{E}) = \frac{2}{\sqrt{\pi}} \frac{1}{\mathrm{T_e^{3/2}}} \exp\left(-\frac{\mathcal{E}}{\mathrm{T_e}}\right) \tag{5}$$

to be the normalized Maxwellian electron energy distribution. Thus

$$k(\mathrm{T_e}) = \int_0^\infty \sigma(\mathcal{E}) \left(\frac{8e\mathrm{T_e}}{\pi m_e}\right)^{1/2} \frac{\mathcal{E}}{\mathrm{T_e}} \exp\left(-\frac{\mathcal{E}}{\mathrm{T_e}}\right) \frac{d\mathcal{E}}{\mathrm{T_e}} \tag{6}$$

is the rate coefficient. The computed rate coefficients are fit to Arrhenius form

$$k(\mathrm{T_e}) = A T_e^B \exp(-C/\mathrm{T_e}) \tag{7}$$

in the range 1 - 7 eV using the code written by Philip (1998) where A, B and C are constants.

3 Nitrogen atom

The most important rate coefficients for electron collisions in nitrogen are electron-neutral ionization k_{iz}, electron impact excitation k_{ex}, and elastic scattering k_{el}. The ion-neutral collisional cross section is mainly due to resonant charge transfer of N^+ on N and elastic scattering and a typical value is (Book, 1990)

$$\sigma_i \approx 5 \times 10^{-19} \quad \mathrm{m}^2$$

3.1 Excitation rate coefficient

The electron impact excitation cross sections for excitation to the $(2s^2 2p^2 3s)^4$P and $(2s^2 p^4)^4$P states of the nitrogen atom were measured by Stone and Zipf (1973) and Spence and Burrow (1980). The various electron impact excitation cross sections for the nitrogen atom have been calculated by Henry et al. (1969), Ganas (1973), Tayal and Beatty (1999), Kim and Desclaux (2002) and Frost et al. (1998). The available data on excitation of atomic nitrogen is reviewed by Kato (1994). The $2s^2 2p^3$ $^2\mathrm{D^o}$ and $^2\mathrm{P^o}$ states are metastable and are 2.38 eV and 3.58 eV above the ground state, respectively. The cross section used to calculate the rate coefficients for electron impact excitation to the metastable levels $2s^2 2p^3$ $^2\mathrm{P^o}$ and $^2\mathrm{D^o}$ are taken from the theoretical estimate by Kim and Desclaux (2002). The cross section for calculating the rate coefficients for electron impact excitation to the 3s states (^{4}P and ^{4}P), the 3p states ($^2\mathrm{S^o}$, $^4\mathrm{D^o}$, $^4\mathrm{P^o}$, $^4\mathrm{S^o}$, $^2\mathrm{D^o}$, $^2\mathrm{P^o}$) and the 3d states (^{2}P, ^{4}F, ^{2}F, ^{4}D, ^{2}D and ^{4}P) are taken from the theoretical work by Ganas (1973) which uses a Born approximation to calculate the cross sections. The cross section used to calculate the rate coefficients for electron impact excitation to the $2s^2p^4$ (^{4}P) level is taken from the theoretical work of Berrington et al. (1975). The cross section for electron impact excitation to the $2s^2p^4$ (^{4}P) state was measured by Stone and Zipf (1973) and they report a peak value of 2×10^{-20} m^2 which is somewhat higher than the 7.5×10^{-21} m^2 reported by Berrington et al. (1975) for the same transition. The peak in the cross section for the electron impact excitation to the $2s^2p^3 3s$ (^{4}P) measured by Stone and Zipf (1973) is 2.5×10^{-20} m^2 and the peak value measured by Spence and Burrow (1980) is 7.1×10^{-20} m^2 which is somewhat higher than the 2.5×10^{-21} m^2 reported by Ganas (1973) for both $2s^2p^3 3s$ states. The rate coefficients for electron impact excitation of the nitrogen atom are listed in table 1.

3.2 Ionization rate coefficient

The ionization energy for nitrogen atom is $\mathcal{E}_{\mathrm{iz}} = 14.54$ V. The cross section for the ionization of the nitrogen atom has been measured by Brook et al. (1978) and agrees well with the ionization cross section calculated by Kim and Desclaux (2002). To calculate the ionization rate coefficient the ionization cross section calculated by Kim and Desclaux (2002) is used and gives

$$k_{\mathrm{iz}} = 3.87 \times 10^{-15} \mathrm{T_e^{0.86}} \exp(-14.62/\mathrm{T_e})$$

Table 1: Rate coefficients for electron impact excitation of the nitrogen atom.

Final state	Thresh. [V]	Rate coefficient [m^3/s]	Ref.
$2s^2p^3$			
$^4S^o$		ground state	
$^2D^o$	2.38	$1.85 \times 10^{-14} T_e^{0.024} \exp(-12.92/T_e)$	(Kim and Desclaux, 2002)
$^2P^o$	3.58	$8.64 \times 10^{-15} T_e^{0.59} \exp(-11.24/T_e)$	(Kim and Desclaux, 2002)
$2s^2p^33s$		$9.09 \times 10^{-15} T_e^{-0.054} \exp(-11.44/T_e)$	(Ganas, 1973)
^4P	10.33		
^2P	10.69		
$2s^2p^33p$		$2.26 \times 10^{-14} T_e^{-0.187} \exp(-12.02/T_e)$	(Ganas, 1973)
$^2S^o$	11.60		
$^4D^o$	11.76		
$^4P^o$	11.84		
$^4S^o$	11.996		
$^2D^o$	12.006		
$^2P^o$	12.125		
$2s^2p^4$		$3.45 \times 10^{-14} T_e^{-0.40} \exp(-13.42/T_e)$	(Berrington et al., 1975)
^4P	10.93		
$2s^2p^23s'$			
^2D	12.36		
$2s^2p^23d$		$9.22 \times 10^{-15} T_e^{-0.236} \exp(-13.73/T_e)$	(Ganas, 1973)
^2P	12.97		
^4F	12.98		
^2F	13.00		
^4D	13.02		
^2D	13.04		
^4P	13.00		
$2s^2p^23p'$			
$^2D^o$	13.70		
$^2P^o$	13.92		

3.3 Rate coefficient for elastic collision

The low energy electron scattering from atomic nitrogen has been measured by Neynaber et al. (1963) and Miller et al. (1970) and calculated by Ormonde et al. (1973); Thomas et al. (1974); John and Williams (1977); Berrington et al. (1975) and Ramsbottom and Kell (1994). All of the theoretically determined cross sections are somewhat higher than the cross section measured by Neynaber et al. (1963). The measurements by Miller et al. (1970) indicate an increasing cross section with decreasing electron impact energy below 1 eV. But the authors themselves claim the measured values should be treated with caution. We assemble a cross section using the measured values from Neynaber et al. (1963) in the range 2 – 10 eV and Miller et al. (1970) (actually taken from Ramsbottom and Kell (1994)) below 2 eV. The rate coefficient is estimated to be

$$k_{\mathrm{el}} = 2.18 \times 10^{-13} \mathrm{T_e}^{-0.84} \exp(-0.685/\mathrm{T_e})$$

4 Molecular nitrogen

Here we discuss the rate coefficients for electron impact excitation and ionization of the nitrogen molecule as well as the rate coefficient for elastic collision of electrons. The ion-neutral collisional cross section for N_2^+ on N has been estimated to be (Meyer et al., 1992)

$$\sigma_{\mathrm{i}} \approx 8.7 \times 10^{-19} \quad \mathrm{m}^2$$

4.1 Excitation rate coefficient

There are several reports on the integral cross section for electron impact excitation of the N_2 molecule (Cartwright, 1970; Cartwright et al., 1977; Campbell et al., 2001) and production of vibrationally excited N_2 by electron impact (Schulz, 1964; Campbell et al., 2004). The $A^3\Sigma_u^+$, $B^3\Pi_g$, $a'^1\Sigma_u^-$, and $C^3\Pi_u$ states are metastable, located 6.17, 7.35, 8.40 and 11.03 eV above the ground state, respectively. The excitation rate coefficients for the molecular nitrogen are listed in table 2. The cross sections are taken from the data set compiled by Phelps (2004) (see also Phelps and Pitchford (1985)). The vibration cross section listed in the Phelps (2004) dataset was measured by Schulz (1964) and the cross sections for electron impact excitation are taken from Cartwright et al. (1977) except for the cross section for excitation to the singlet states which is from Zipf and Mclaughlin (1978).

4.2 Ionization rate coefficient

The ionization energy of the molecular nitrogen is 15.6 eV. The rate coefficient for ionization of the nitrogen molecule was estimated from the cross section measured by Straub et al. (1996) and is

$$k_{\rm iz} = 7.76 \times 10^{-15} \mathrm{T}_{\rm e}^{0.79} \exp(-16.75/\mathrm{T}_{\rm e})$$

4.3 Rate coefficient for elastic collision

There have been quite a few reports on measurements of the cross section for elastic electron collision with the nitrogen molecules (Shyn and Carignan, 1980; Hoffman et al., 1982; Randell et al., 1994) and for the total scattering cross section (Golden, 1966) as well as a number of theoretical studies (Jain et al., 1984; Dill and Dehmer, 1977). The elastic collision cross section is assembled using the the cross section measured by Randell et al. (1994) in the range 0.01 – 0.14 eV, the cross section measured by Shyn and Carignan (1980) in the range 1.5 – 4 eV and the cross section measured by Hoffman et al. (1982) in the range 2.2 – 700 eV. A fit of the calculated rate coefficient in the range 1 – 7 eV is

$$k_{\rm el} = 1.04 \times 10^{-13} \mathrm{T}_{\rm e}^{0.43} \exp(-0.206/\mathrm{T}_{\rm e})$$

5 Collisional loss

The collisional energy loss per electron-ion pair created $\mathcal{E}_{\rm c}$ as a function of the electron temperature $\mathrm{T}_{\rm e}$ is shown in figure 1 for the nitrogen atom and the nitrogen molecule. At low electron temperature elastic transfer is the dominant contributor to the collisional energy loss. At high electron temperature the energy loss $\mathcal{E}_{\rm c}$ asymptotes to somewhat higher than $\mathcal{E}_{\rm iz}$. For

Table 2: Rate coefficients for electron impact excitation of the nitrogen molecule.

Final state	Thresh. [V]	Rate coefficient [m^3/s]	Ref.
$X^1\Sigma_u^+$	ground		
$r > 0$	0.02	$9.89 \times 10^{-15}\mathrm{T_e^{-1.47}}\exp(-2.42/\mathrm{T_e})$	
$v = 1$	0.29	$4.83 \times 10^{-14}\mathrm{T_e^{-1.46}}\exp(-2.37/\mathrm{T_e})$	
$v = 2$	0.59	$2.97 \times 10^{-14}\mathrm{T_e^{-1.47}}\exp(-2.39/\mathrm{T_e})$	
$v = 3$	0.88	$2.01 \times 10^{-14}\mathrm{T_e^{-1.48}}\exp(-2.37/\mathrm{T_e})$	
$v = 4$	1.17	$1.42 \times 10^{-14}\mathrm{T_e^{-1.49}}\exp(-2.43/\mathrm{T_e})$	
$v = 5$	1.47	$1.21 \times 10^{-14}\mathrm{T_e^{-1.49}}\exp(-2.49/\mathrm{T_e})$	
$v = 6$	1.76	$1.06 \times 10^{-14}\mathrm{T_e^{-1.49}}\exp(-2.59/\mathrm{T_e})$	
$v = 7$	2.06	$6.00 \times 10^{-15}\mathrm{T_e^{-1.49}}\exp(-2.73/\mathrm{T_e})$	
$v = 8$	2.35	$2.96 \times 10^{-15}\mathrm{T_e^{-1.49}}\exp(-2.88/\mathrm{T_e})$	
$A^3\Sigma_u^+(v = 0 - 4)$	6.17	$8.06 \times 10^{-16}\mathrm{T_e^{-0.306}}\exp(-8.87/\mathrm{T_e})$	
$A^3\Sigma_u^+(v = 5 - 9)$	7.0	$3.98 \times 10^{-15}\mathrm{T_e^{-0.35}}\exp(-9.39/\mathrm{T_e})$	
$B^3\Pi_g$	7.35	$1.56 \times 10^{-14}\mathrm{T_e^{-0.52}}\exp(-9.16/\mathrm{T_e})$	
$W^3\Delta_u$	7.36	$7.13 \times 10^{-15}\mathrm{T_e^{-0.086}}\exp(-8.83/\mathrm{T_e})$	
$A^3\Sigma_u^+(v > 10)$	7.80	$2.79 \times 10^{-15}\mathrm{T_e^{-0.18}}\exp(-9.27/\mathrm{T_e})$	
$B'^3\Sigma_u^-$	8.16	$3.79 \times 10^{-15}\mathrm{T_e^{-0.33}}\exp(-9.76/\mathrm{T_e})$	
$a'^1\Sigma_u^-$	8.40	$6.60 \times 10^{-15}\mathrm{T_e^{-0.66}}\exp(-11.05/\mathrm{T_e})$	
$a^1\Pi_g$	8.55	$2.85 \times 10^{-15}\mathrm{T_e^{0.39}}\exp(-9.40/\mathrm{T_e})$	
$w^1\Pi_u$	8.89	$6.57 \times 10^{-15}\mathrm{T_e^{-0.61}}\exp(-10.45/\mathrm{T_e})$	
$C^3\Pi_u$	11.03	$6.46 \times 10^{-15}\mathrm{T_e^{-0.74}}\exp(-12.90/\mathrm{T_e})$	
$E^3\Sigma_g^+$	11.88	$3.96 \times 10^{-15}\mathrm{T_e^{-0.38}}\exp(-10.38/\mathrm{T_e})$	
$a''^1\Sigma_g^+$	12.25	$2.21 \times 10^{-15}\mathrm{T_e^{-0.25}}\exp(-13.64/\mathrm{T_e})$	
sum of singlets	13	$1.15 \times 10^{-14}\mathrm{T_e^{0.704}}\exp(-13.33/\mathrm{T_e})$	

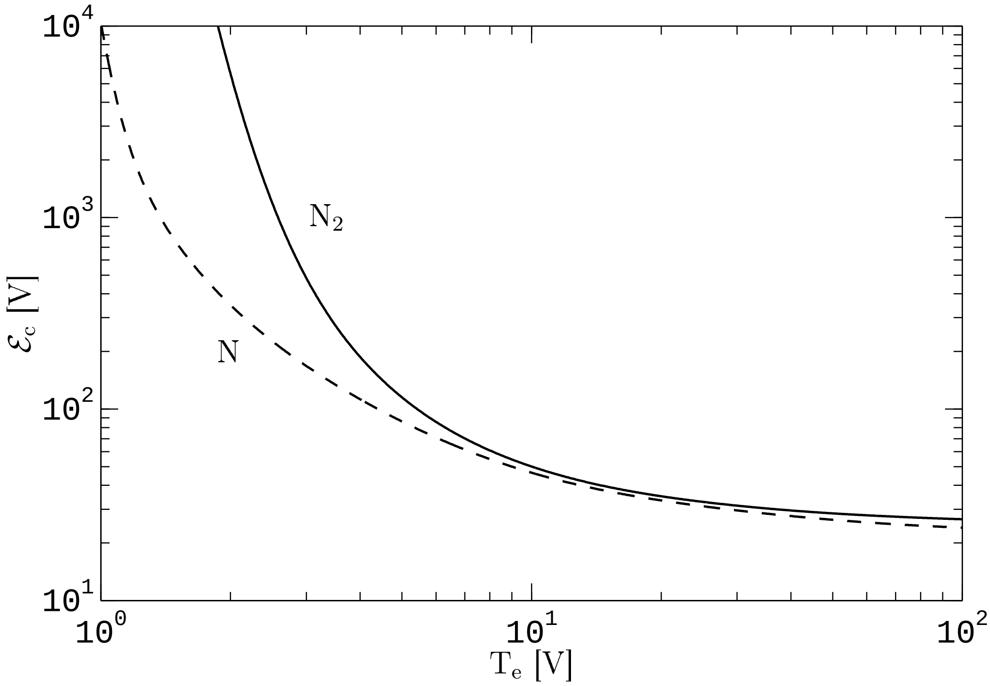

Figure 1: The collisional energy loss per electron-ion pair created $\mathcal{E}_c$ as a function of the electron temperature T_e for the nitrogen atom and the nitrogen molecule.

molecular gases such as nitrogen additional energy losses include excitation of vibrational and rotational energy levels, molecular dissociation etc. Thus in molecular gas $\mathcal{E}_c$ is significantly higher than for noble gas at a given electron temperature.

6 Conclusion

The rate coefficients used to evaluate the collisional energy loss per electron ion pair created are reviewed for the nitrogen atom and the nitrogen molecule. The electron energy distribution is assumed to be Maxwellian and the fit to the rate coefficient is valid in the range 1 – 7 eV.

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