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Electron Excitation and Ionization Rate Coefficients, and the Rate of Energy Loss to the Vibrational Levels
(Application to the Nitrogen Molecule)

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CONTENTS

Abstract	ii
Problem Status	ii
Authorization	ii
INTRODUCTION	1
ELECTRON EXCITATION AND IONIZATION RATE COEFFICIENTS	1
APPLICATION TO THE NITROGEN MOLECULE	2
Electron Excitation Rate Coefficient $X_{OC} \equiv \langle \sigma_{OC} v \rangle$	2
Electron Excitation Rate Coefficient X_{OB}	5
Ionization Rate Coefficient X_i	6
THE RATE OF ENERGY LOSS BY ELECTRONS TO THE GROUND STATE VIBRATIONAL LEVELS OF THE NITROGEN MOLECULE	7
REFERENCES	10

ABSTRACT

A general expression was given for the calculations of electron excitation or ionization rate coefficients using reliably measured cross sections. This method was then applied to the nitrogen molecule, where the cross section for the excitation of $C^3\pi\,(v = 0)$ and the ionization were used to obtain the respective rate coefficients as functions of the electron temperature. In addition, the rate of the energy loss, by electrons, to the ground state vibrational levels of the nitrogen molecule was also calculated using an expression obtained for such a loss rate.

PROBLEM STATUS

This is a final report on one phase of the theoretical calculations on the performance and limitations of high-temperature gas lasers.

AUTHORIZATION

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ELECTRON EXCITATION AND IONIZATION RATE COEFFICIENTS, AND THE RATE OF ENERGY LOSS TO THE VIBRATIONAL LEVELS (APPLICATION TO THE NITROGEN MOLECULE)

INTRODUCTION

Recently, laser actions from molecular nitrogen have been reported (1-3). One particular lasing line, the 3371-Å line, which corresponds to the transition $C^3\pi_u \rightarrow B^3\pi_g$ in the second positive band, has attracted considerable interest because of the possibility of obtaining a high power output (2,3) from such a line. A theoretical calculation of the saturated power output was first advanced by Gerry (4). An improved theory including the collisional mixing is forthcoming (5). However, to be able to calculate the power output, electron excitation rate coefficients to the zero vibrational levels of the upper laser ($C^3\pi_u$) and the lower laser ($B^3\pi_g$) levels, from the ground state ($X'\Sigma_g^+$) of the nitrogen molecule, must be obtained in at least an empirical form. In addition to the aforementioned rate coefficients, the ionization rate coefficient of the nitrogen molecule and the rate of energy loss, by electrons, to the ground state vibrational levels must be known. The motive behind the calculation of these rate coefficients is clear; however, the method which is used here is more general and can be applied to any measured cross section.

ELECTRON EXCITATION AND IONIZATION RATE COEFFICIENTS

To calculate excitation or ionization rate coefficients, one must have either analytic expressions for the cross sections or experimentally measured cross sections. If an analytic, i.e., a theoretical expression exists, then one proceeds to average the product of this expression and the electron velocity over a Maxwellian velocity distribution and thus obtains the relevant rate coefficient. However, experimentally measured cross sections are usually more often available than the theoretical expressions. Therefore, in most cases, one must have reliably measured cross sections to calculate these rate coefficients. The procedure, then, is to obtain an empirical expression for the measured cross section over the entire range of the incident electron energy. This can be done by dividing the range into a few sections and obtaining empirical expressions for the cross section, as a function of the incident electron energy, for each of the sections. Then if σ_{ij} denotes the cross section as a function of E, the incident electron energy, for either excitation or ionization of atoms, ions, or molecules, the relevant rate coefficient can be expressed as

$$X_{ij} \equiv \langle \sigma_{ij} v \rangle = \int \sigma_{ij} v\, f(\mathbf{v})\, d\mathbf{v}\,, \tag{1}$$

where $f(\mathbf{v})\, d\mathbf{v}$ is the Maxwellian velocity distribution within the velocity range of $\mathbf{v}$ and $\mathbf{v} + d\mathbf{v}$ and satisfies the condition $\int f(\mathbf{v})\, d\mathbf{v} = 1$. The distribution $f(\mathbf{v})\, d\mathbf{v}$ can be expressed as

$$f(\mathbf{v})\, d\mathbf{v} = 10.74 \times 10^{-24} \left(T_e^{3/2}\right)^{-1} v^2\, e^{-mv^2/2T_e}\, dv, \tag{2}$$

where T_e* is in units of eV. However, from Eqs. (1) and (2) it is convenient to express $vf(\mathbf{v})\, d\mathbf{v}$ in terms of the electron energy. This leads to the expression

*Hereafter, T_e will be written as T.

$$X_{ij} \equiv \langle \sigma_{ij} v \rangle = K_0 T^{-3/2} \int E \sigma_{ij}(E)\, e^{-E/T}\, dE, \tag{3}$$

where $K_0 = 6.6971 \times 10^7$.

Generally, one can express each of the sections of σ_{ij} with reasonable accuracy by one or more terms of the polynomial $C_0 + C_1 E + C_2 E^2 + C_3 E^3$, or by an exponential term such as $C_e e^{-aE}$, or by a combination of the two. Thus the rate coefficient can now be expressed in a more general fashion as

$$X_{ij} = K_0 T^{-3/2} \sum_a \int_{E_a}^{E_{(a+1)}} E(C_{0a} + C_{1a}E + C_{2a}E^2 + C_{3a}E^3)\, e^{-[\alpha_a E + (E/T)]}\, dE, \tag{4}$$

where a indicates the ath division of the cross section between E_a and $E_{(a+1)}$, and the coefficients of the polynomial, C_{0a}, C_{1a}, etc., and α_a are for the ath division. The summation, of course, is over all sections of σ.

Typical of the integrals* which arise is

$$K_0 T^{-3/2} \int_{E_a}^{E_{(a+1)}} E(C_{0a} + C_{1a}E + C_{2a}E^2 + C_{3a}E^3)\, e^{-[\alpha_a E + (E/T)]}\, dE$$

$$= \frac{K_0 T^{-1/2}}{1 + \alpha_a T} \Big\{ e^{-E(1+\alpha_a T)/T} \Big[(C_{0a}E + C_{1a}E^2 + C_{2a}E^3 + C_{3a}E^4)$$

$$+ \big(T/(1+\alpha_a T)\big)(C_{0a} + 2C_{1a}E + 3C_{2a}E^2 + 4C_{3a}E^3)$$

$$+ \big(T/(1+\alpha_a T)\big)^2 (2C_{1a} + 6C_{2a}E + 12C_{3a}E^2)$$

$$+ \big(T/(1+\alpha_a T)\big)^3 (6C_{2a} + 24C_{3a}E) + \big(T/(1+\alpha_a T)\big)^4\, 24C_{3a} \Big] \Big\}_{E_{(a+1)}}^{E_a}, \tag{5}$$

where evaluation limits have been interchanged to eliminate the minus sign.

APPLICATION TO THE NITROGEN MOLECULE

The method described in the previous section is applied here to calculate the electron excitation rate coefficient $X_{OC} \equiv \langle \sigma_{OC} v \rangle$, the electron excitation rate coefficient X_{OB}, and the ionization rate coefficient X_i.

Electron Excitation Rate Coefficient $X_{OC} \equiv \langle \sigma_{OC} v \rangle$

Figure 1 shows the measured (6) cross section of excitation by electron impact for a nitrogen molecule from the zero vibrational level of the ground state $X'\Sigma_g^+$ to the zero

*Integrals of this type are readily evaluated by the formula

$$\int P_m(x)\, e^{ax}\, dx \equiv \frac{e^{ax}}{a} \sum_{k=0}^{m} \frac{(-1)^k P^{(k)}(x)}{a^k},$$

where $P_m(x)$ is a polynomial of degree m in x and $p^{(k)}(x)$ is the kth derivative of $P_m(x)$ with respect to x.

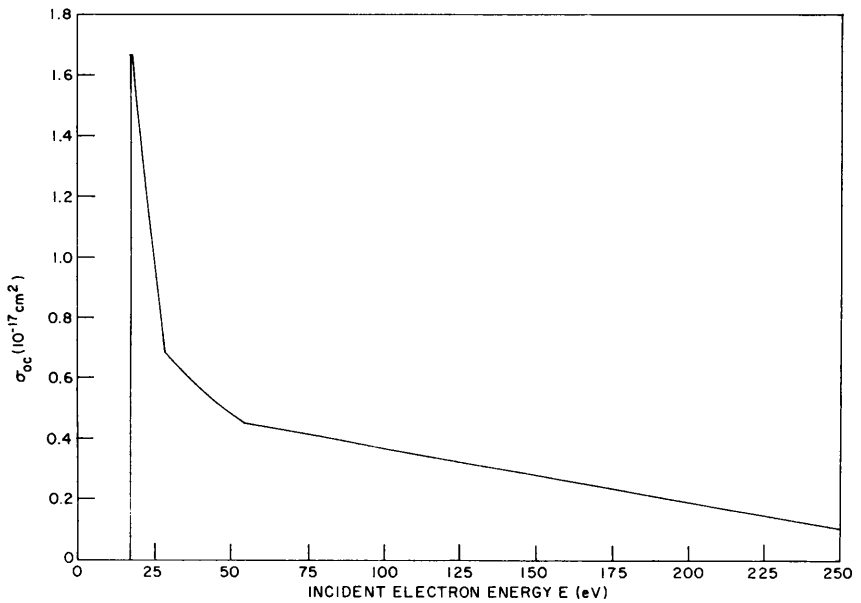

Fig. 1 - Measured electron excitation cross section σ_{0c} of Stewart and Gabathuler

vibrational level of the excited state $C^3\pi_u$ (hereafter called σ_{0c}). This cross section was divided into five regions, and an empirical expression for σ_{0c} as a function of E, the energy of the incident electron in eV, was obtained. These expressions are (a) $\sigma_{0c} = 0$ for $0 \leqq E \leqq 17.00$, (b) $\sigma_{0c} = 16.6668 \times 10^{-18}$ for $17 \leqq E \leqq 18.21$, (c) $\sigma_{0c} = 75.3954 \times 10^{-18} e^{-0.083E}$ for $18.21 \leqq E \leqq 29.01$, (d) $\sigma_{0c} = 10.5217 \times 10^{-18} e^{-0.015E}$ for $29.01 \leqq E \leqq 56.46$, and (e) $\sigma_{0c} = (5.5227 - 0.0018E) \times 10^{-18}$ for $56.46 \leqq E \leqq 307.38$. Inserting these expressions into Eq. (5) (for example in the range $18.21 \leqq E \leqq 29.01$, all coefficients are zero except C_1 and α) and regrouping terms with the same exponential, one arrives at the following expression for X_{0c} as a function of T:

$$
\begin{aligned}
X_{0c} = \frac{10^{-8}}{\sqrt{T}} \Bigg\{ & (1.8975 + 0.11162T)\, e^{-17/T} \\
& + \left[2.0322 \left(-1 + \frac{1}{1 + 0.083T} \right) + 0.11162T \left(-1 + \frac{1}{(1 + 0.083T)^2} \right) \right] e^{-18.21/T} \\
& + \left[1.3225 \left(-\frac{1}{1 + 0.083T} + \frac{1}{1 + 0.015T} \right) \right. \\
& \left. + 0.045592T \left(\frac{1}{(1 + 0.083T)^2} + \frac{1}{(1 + 0.015T)^2} \right) \right] e^{-29.01/T} \\
& + \left[\frac{1.7048}{1 + 0.015T} - \frac{0.030192T}{(1 + 0.015T)^2} + 1.7048 + 0.023398T - 0.00024066T^2 \right] e^{-56.46/T} \\
& + [0.036986T + 0.00024066T^2]\, e^{-307.38/T} \Bigg\}
\end{aligned}
\tag{6}
$$

Although Eq. (6) is long and tedious, it can be programmed easily for calculations with a computer. A computer calculation of X_{0c} as a function of T is shown in Fig. 2 and tabulated in Table 1.

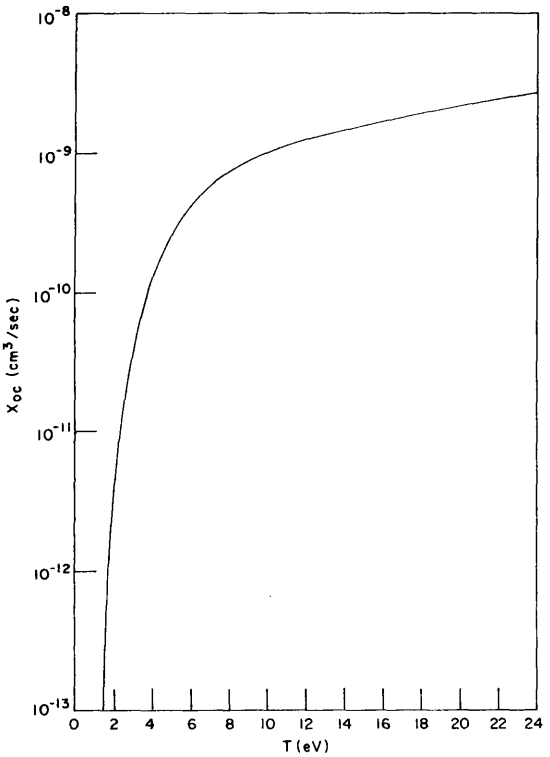

Fig. 2 - Excitation rate coefficient X_{0c}

Table 1
Excitation and Ionization Rate Coefficients as Functions of Electron Temperature

T_e	X_{0c} (cm³/sec)	X_{0B} (cm³/sec)	X_i (cm³/sec)
1	8.11(-16)*	2.51(-15)	1.15(-15)
2	2.78(-12)	1.39(-12)	4.81(-12)
3	3.77(-11)	1.04(-11)	8.99(-11)
4	1.31(-10)	2.68(-11)	4.21(-10)
5	2.69(-10)	4.61(-11)	1.12(-9)
6	4.26(-10)	6.49(-11)	2.23(-9)
7	5.84(-10)	8.18(-11)	3.73(-9)
8	7.34(-10)	9.65(-11)	5.57(-9)
9	8.74(-10)	1.09(-10)	7.72(-9)
10	1.01(-9)	1.20(-10)	1.01(-8)
11	1.13(-9)	1.29(-10)	1.27(-8)
12	1.25(-9)	1.37(-10)	1.54(-8)
13	1.36(-9)	1.43(-10)	1.83(-8)
14	1.47(-9)	1.49(-10)	2.12(-8)
16	1.70(-9)	1.58(-10)	2.72(-8)
18	1.94(-9)	1.66(-10)	3.32(-8)
20	2.18(-9)	1.71(-10)	3.92(-8)
22	2.44(-9)	1.76(-10)	4.51(-8)
24	2.72(-9)	1.80(-10)	5.09(-8)

*Numbers in parenthesis indicate powers of 10 by which the entries are to be multiplied.

Electron Excitation Rate Coefficient X_{0B}

Although no measured cross section for σ_{0B}, the excitation cross section to the zero vibrational level of the excited state $B^3\pi_g$, is available, the physical mechanism for its excitation is the same as that for $C^3\pi_u$. Therefore, it will be expected that σ_{0B} will have the same shape as σ_{0c} (Fig. 1) except that it will have a somewhat smaller amplitude and be shifted towards the lower energy by 3.66 eV. An expression for X_{0B} can be given as

$$
\begin{aligned}
X_{0B} = \frac{10^{-8}\gamma}{\sqrt{T}} \Bigg\{ & (1.4890 + 0.11162T)\, e^{-13.34/T} + \Bigg[1.6237 \left(-1 + \frac{1}{1 + 0.083T} \right) \\
& + 0.11162T \left(-1 + \frac{1}{(1 + 0.083T)^2} \right) \Bigg] e^{-14.55/T} + \Bigg[1.1555 \left(\frac{1}{1 + 0.083T} + \frac{1}{1 + 0.015T} \right) \\
& + 0.045586T \left(\frac{-1}{(1 + 0.083T)^2} + \frac{1}{(1 + 0.015T)^2} \right) \Bigg] e^{-25.35/T} \\
& + \left[\frac{-1.5929}{1 + 0.015T} - \frac{0.030260T}{(1 + 0.015T)^2} + 1.5929 + 0.023899T - 0.00024168T^2 \right] e^{-52.64/T} \\
& + [0.036621T + 0.00024168T^2]\, e^{-303.06/T} \Bigg\}, \qquad (7)
\end{aligned}
$$

where γ is a number less than 1 indicating that the cross section σ_{0B} has a smaller amplitude than σ_{0c}.

Figure 3 shows a calculation X_{0B}, given in Table 1, for $\gamma = 0.1$.

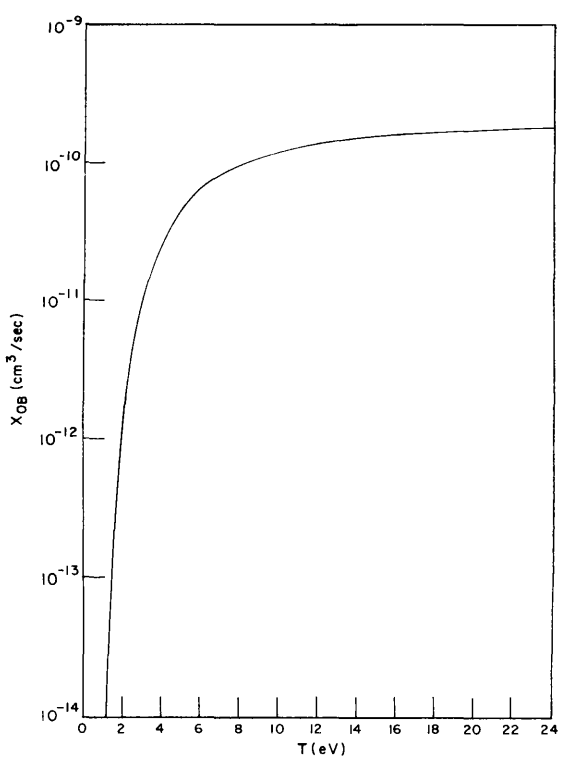

Fig. 3 - Excitation rate coefficient X_{0B}

Ionization Rate Coefficient X_i

The ionization rate coefficient $\langle\sigma_i v\rangle$ (hereafter called X_i) was obtained in the same manner as discussed earlier. The measured cross section for ionization of the nitrogen molecule as obtained by Tate and Smith (7), shown in Fig. 4, was used. The empirical

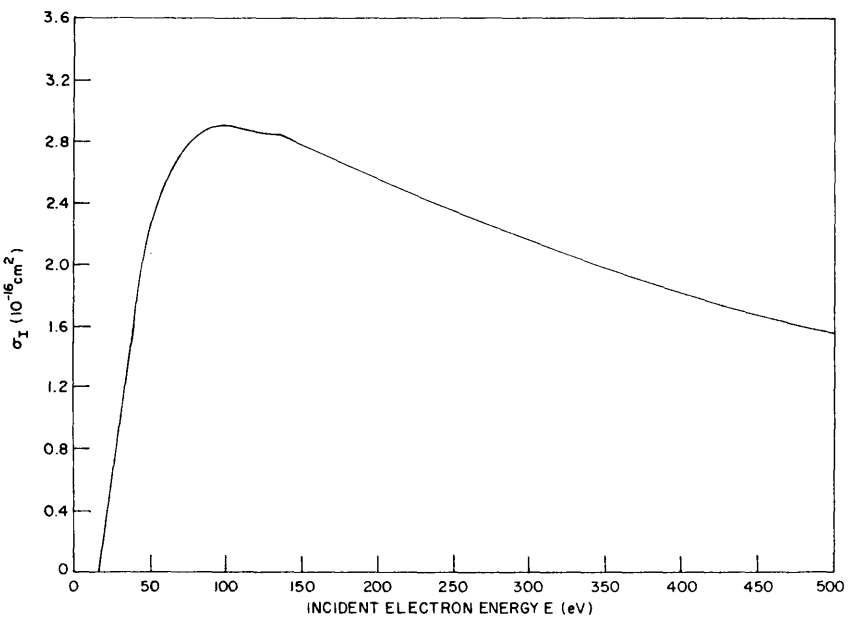

Fig. 4 - Measured nitrogen molecule ionization cross section of Tate and Smith

expressions for σ_i for several regions in which it was divided are (a) $\sigma_i = 0$ for $0 \leqq E \leqq 15.77$, (b) $\sigma_i = 0.283 \times 10^{-16}(-3.7995 + 0.24099E)$ for $15.77 \leqq E \leqq 45.81$, (c) $\sigma_i = 0.283 \times 10^{-16}(-4.5203 + 0.39369E - 0.0034374E^2 + 0.0000097687E^3)$ for $45.81 \leqq E \leqq 135.77$, (d) $\sigma_i = 0.283 \times 10^{-16} \times 12.5837e^{-0.00168E}$ for $135.77 \leqq E \leqq 465.16$, and (e) $\sigma_i = 0.283 \times 10^{-16} \times 10.065e^{-0.0012E}$ for $465.16 \leqq E \leqq 750$.

Finally, the expression for X_i as a function of electron temperature is

$$
\begin{aligned}
X_i = \frac{2.83 \times 10^{-10}}{\sqrt{T}} \Bigg\{ & [25.446T + 3.2279T^2]e^{-15.77/T} \\
& + [-30.904T - 2.6346T^2 - 0.066195T^3 + 0.0015701T^4]e^{-45.81/T} \\
& + \Bigg[-9108.1 - 67.561T - 0.99207T^2 - 0.075060T^3 - 0.0015701T^4 \\
& + \frac{9108.1}{1 + 0.00168T} + \frac{67.803T}{(1 + 0.00168T)^2}\Bigg]e^{-135.77/T} + \Bigg[17940.3\left(\frac{1}{1 + 0.0012T} - \frac{1}{1 + 0.00168T}\right) \\
& + 38.568\left(\frac{1}{(1 + 0.0012T)^2} - \frac{1}{(1 + 0.00168T)^2}\right)\Bigg] e^{-465.16/T} \\
& + \left(\frac{-25050.0}{1 + 0.0012T} - \frac{27.400T}{(1 + 0.0012T)^2}\right) e^{-750/T}\Bigg\}. \qquad (8)
\end{aligned}
$$

Figure 5 shows a calculation of X_i as a function of temperature, which is also given in Table 1.

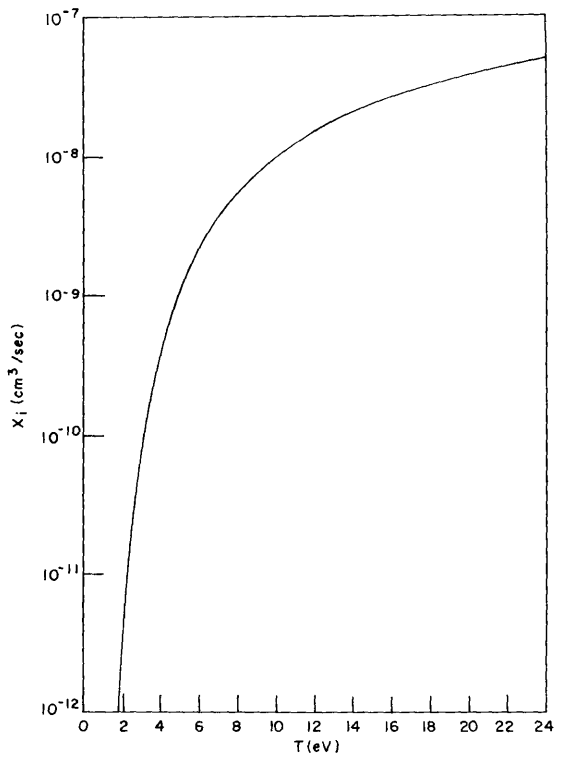

Fig. 5 - Ionization rate coefficient

THE RATE OF ENERGY LOSS BY ELECTRONS TO THE GROUND STATE VIBRATIONAL LEVELS OF THE NITROGEN MOLECULE

Haas (8) first observed a peak with a maximum at 2.3 eV in the excitation spectrum of the nitrogen molecule by electron impacts. This low-energy loss does not result from any known electronically excited level, since the first electronically excited level ($A^3\,\Sigma_u$) lies above 6.0 eV. This low-energy loss, then, is attributed to the ground state vibrational levels which are separated by approximately 0.3 eV. Schulz (9) was able to observe up to $v = 8$ ground state vibrational levels, indicating that these vibrational levels were responsible for the low-energy-loss peak in the excitation spectrum. The total cross section for these vibrational levels σ_{vib} (9) is shown in Fig. 6. Since the peak of each individual vibrational levels comes at different energies, it is more appropriate to calculate the rate of the energy loss $\langle\sigma_{vib} vE\rangle$ rather than the rate coefficient. Thus,

$$\langle\sigma_{vib} vE\rangle = K_0 T^{-3/2} \sum_a \int_{E_a}^{E_{(a+1)}} E^2 (C_{0a} + C_{1a}E + C_{2a}E^2 + C_{3a}E^3)\, e^{-[\alpha_a E + (E/T)]}\, dE\,, \tag{9}$$

and a typical integral for the range (E_a, $E_{(a+1)}$) is

$$K_0 T^{-3/2} \int_{E_a}^{E_{(a+1)}} E^2 (C_{0a} + C_{1a}E + C_{2a}E^2 + C_{3a}E^3)\, e^{-[E\alpha_a + (E/T)]} dE$$

$$= \frac{K_0 T^{-1/2}}{1+\alpha_a T} \left\{ e^{-(1+\alpha_a T)E/T} \left[(C_{0a}E^2 + C_{1a}E^3 + C_{2a}E^4 + C_{3a}E^5) \right.\right.$$

$$+ \frac{T}{1+\alpha_a T} (2C_{0a}E + 3C_{1a}E^2 + 4C_{2a}E^3 + 5C_{3a}E^4) + \left(\frac{T}{1+\alpha_a T}\right)^2 (2C_{0a} + 6C_{1a}E + 12C_{2a}E^2 + 20C_{3a}E^3)$$

$$+ \left(\frac{T}{1+\alpha_a T}\right)^3 (6C_{1a} + 24C_{2a}E + 60C_{3a}E^2) + \left(\frac{T}{1+\alpha_a T}\right)^4 (24C_{2a} + 120C_{3a}E)$$

$$\left.\left. + \left(\frac{T}{1+\alpha_a T}\right)^5 (120C_{3a}) \right] \right\}_{E_{(a+1)}}^{E_a} \tag{10}$$

where, again, the limits have been switched to eliminate the minus sign.

Figure 6 shows σ_{vib} for which the following empirical expressions were obtained: (a) $\sigma_{vib} = 0$ for $0 \leqq E \leqq 0.85$, (b) $\sigma_{vib} = 1.5 \times 10^{-16}(-1.614 + 4.518E - 4.198E^2 + 1.314E^3)$ for $0.85 \leqq E \leqq 1.52$, (c) $\sigma_{vib} = 1.5 \times 10^{-16}(-3.141 + 2.173E)$ for $1.52 \leqq E \leqq 1.85$, (d) $\sigma_{vib} = 1.5 \times 10^{-16}(-17.87 + 23.31E - 9.354E^2 + 1.207E^3)$ for $1.85 \leqq E \leqq 2.45$, (e) $\sigma_{vib} = 1.5 \times 10^{-16}(3.507 - 1.086E)$ for $2.45 \leqq E \leqq 2.84$, (f) $\sigma_{vib} = 1.5 \times 10^{-16}(3.959 - 1.917E + 0.2362E^2)$ for $2.84 \leqq E \leqq 4.0$, and (g) $\sigma_{vib} = 1.5 \times 10^{-16} \times 0.1543e^{-0.1923E}$ for $4 \leqq E \leqq \infty$.

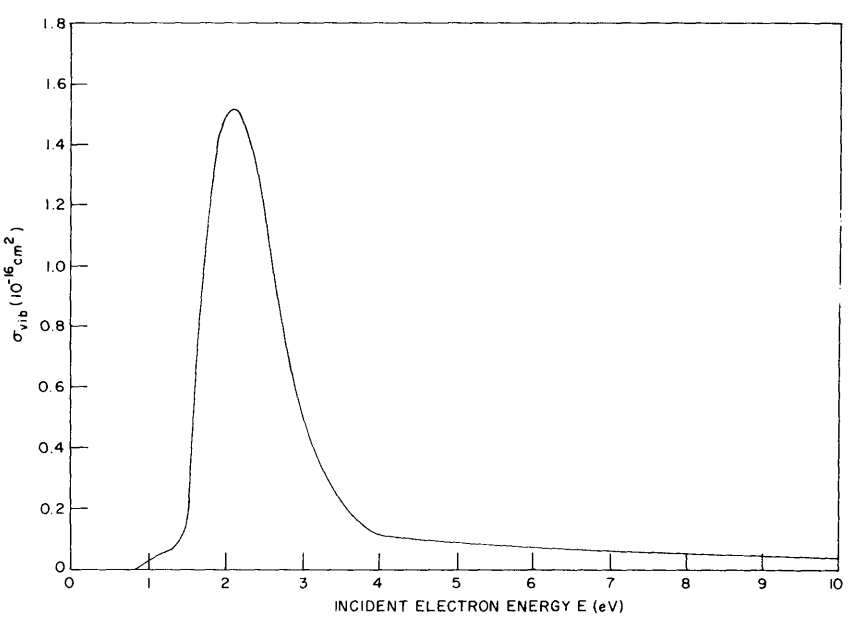

Fig. 6 - Ground state vibrational level cross section of Shulz

Inserting these expressions of σ_{vib} into Eq. (10) and regrouping terms with the same exponential, one arrives at the following expressions* for $\langle\sigma_{vib} vE\rangle$ as a function of T_e:

$$\begin{aligned}\langle\sigma_{vib} vE\rangle = \frac{1.5\times10^{-9}}{\sqrt{T}}\Bigg\{&[1.0855T - 3.0702T^2 - 10.082T^3 + 225.42T^4 + 1055.6T^5]e^{-0.85/T}\\
&+ [20.246T - 2.8376T^2 - 290.73T^3 - 932.87T^4 - 1055.6T^5]e^{-1.52/T}\\
&+ [-12.061T + 18.769T^2 - 209.33T^3 - 874.35T^4 - 970.16T^5]e^{-2.45/T}\\
&+ [-25.162T - 175.89T^2 - 271.00T^3 + 294.34T^4 + 970.16T^5]e^{-1.85/T}\\
&+ [27.716T + 64.564T^2 + 74.541T^3 + 37.968T^4]e^{-2.84/T}\\
&+ \left[7.6592\left(-1 + \frac{1}{1+0.1923T}\right) + \frac{3.8296T}{(1+0.1923T)^2} - 0.92802T\right.\\
&\left.+ T^2\left(-48.680 + \frac{0.95740}{(1+0.1923T)^3}\right) - 74.848T^3 - 37.968T^4\right]e^{-4.0/T}\Bigg\}. \qquad (11)\end{aligned}$$

Equation (11) was programmed for the computer, and that result is shown in Fig. 7 and in Table 2.

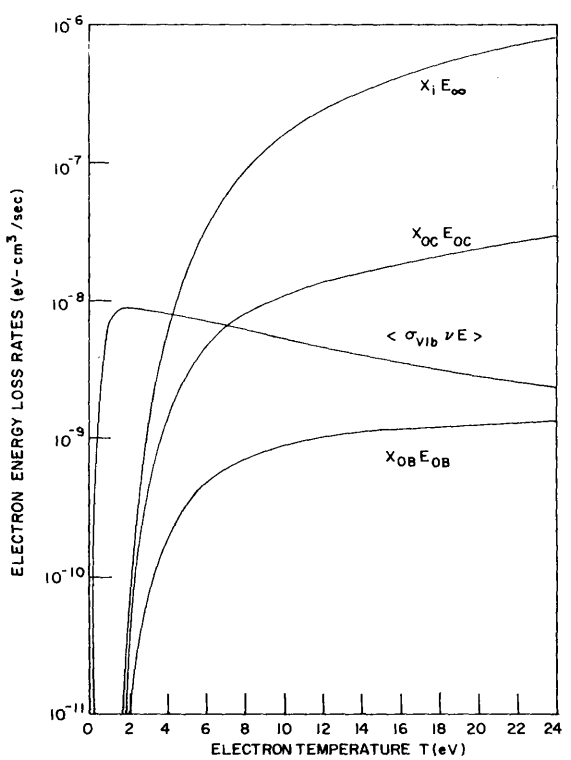

Fig. 7 - Electron energy loss rates as a function of electron temperature

*While only four or five figures are shown here, the calculations were done carrying ten digits. This was necessary because of severe cancellations in Eq. (11).

Table 2
Rate of Energy Loss to the Ground State Vibrational Levels as a Function of Electron Temperature

T_e	$\langle\sigma_{vib}Ev\rangle$ (eV-cm^3/sec)	T_e	$\langle\sigma_{vib}Ev\rangle$ (eV-cm^3/sec)
1	6.58(-9)*	11	4.93(-9)
2	8.91(-9)	12	4.63(-9)
3	8.70(-9)	13	4.34(-9)
4	8.14(-9)	14	4.10(-9)
5	7.56(-9)	16	3.64(-9)
6	7.01(-9)	18	3.30(-9)
7	6.51(-9)	20	2.91(-9)
8	6.05(-9)	22	2.60(-9)
9	5.64(-9)	24	2.30(-9)
10	5.27(-9)		

*Numbers in parenthesis indicate powers of 10 by which the entries are to be multiplied.

Finally, the rate of the electron energy loss to the vibrational levels is compared with the energy loss to the upper laser level $X_{0C}E_{0C}$, to the lower laser level $X_{0B}E_{0B}$, and to the ionization of the nitrogen molecule $X_i E_\infty$, as shown on Fig. 7.

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ELECTRON EXCITATION AND IONIZATION RATE COEFFICIENTS, AND THE RATE OF ENERGY LOSS TO THE VIBRATIONAL LEVELS (APPLICATION TO THE NITROGEN MOLECULE)

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)

This is a final report on one phase of the problem.

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13. ABSTRACT

A general expression was given for the calculations of electron excitation or ionization rate coefficients using reliably measured cross sections. This method was then applied to the nitrogen molecule, where the cross section for the excitation of $C^3\pi(v = 0)$ and the ionization were used to obtain the respective rate coefficients as functions of the electron temperature. In addition, the rate of the energy loss, by electrons, to the ground state vibrational levels of the nitrogen molecule was also calculated using an expression obtained for such a loss rate.

DD FORM 1 NOV 65 1473 (PAGE 1)

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DD FORM 1 NOV 65 1473 (BACK) GPO 927-609

(PAGE 2)

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