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Bibliography of Electron and Photon Cross Sections  
with Atoms and Molecules  
Published in the 20<sup>th</sup> Century  
– Halogen Molecules –

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# Bibliography of Electron and Photon Cross Sections

with Atoms and Molecules

Published in the 20<sup>th</sup> Century

—— Halogen Molecules ——\*

Makoto Hayashi

(Gaseous Electronics Institute)

A bibliographies of original and review reports of experiments or theories of electron and photon cross sections and also electron swarm data are presented for atomic or molecular species with specified targets. These works covered 17 atoms and 51 molecules. The present bibliography is only for halogen molecules ( $F_2$ ,  $Cl_2$ ,  $Br_2$ ,  $I_2$ ). About 190 ( $F_2$ ), 360 ( $Cl_2$ ), 140 ( $Br_2$ ) and 240 ( $I_2$ ) papers were compiled respectively. A comprehensive author indexes for each molecule are included. The bibliography covers the period 1901 through 2000 for  $F_2$ - $I_2$ . Finally, author's comments for  $F_2$ - $I_2$  electron collision cross sections are given.

Keywords :  $F_2$ ,  $Cl_2$ ,  $Br_2$ ,  $I_2$  molecules, collision cross sections, electron, elastic scattering, rotational, vibrational and electronic excitations, dissociation, ionization photon, photoabsorption, photodissociation, photoexcitation, photoionization, electron swarm, drift velocity, diffusion coefficient, ionization coefficient, excitation and ionization energies, transition probabilities, lifetimes of excited states

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## Introduction

### History

This bibliography is the result of a continuing literature survey which was begun around 1970 and originally encompassed only electron collision cross section and electron swarm data. The organization responsible for continuing this survey is Nagoya Institute of Technology, Nagoya. From 1994, the work continued to Gaseous Electronics Institute, Nagoya. In 1997, the collection of photon cross section references was begun. The search for references in both cases was retrospective and included all papers reporting measurements, theoretical calculations or reviews and data compilations of such cross sections and electron swarm data.

### Scope

This bibliography contains references to original research papers which report experiments or theoretical calculations of cross sections for electron and photon collisions with halogen molecules  $F_2$ ,  $Cl_2$ ,  $Br_2$ , and  $I_2$ . The review papers on this subject are also included. Some halogen molecule cluster papers are included. Some conference reports, company or agency reports and PhD thesis are included. Halogen molecules ion papers and positron collision papers are not included in principle.

Papers reporting the following data are included.

For electron collision cross section :

- 1) elastic scattering
- 2) rotational excitation
- 3) vibrational excitation
- 4) electronic excitation
- 5) dissociation
- 6) ionization
- 7) attachment
- 8) grand total scattering (sum of elastic and inelastic cross sections)
- 9) electron swarm parameters (drift velocity, diffusion coefficient)
- 10) excitation and ionization coefficients

For photon collision cross section :

- 1) photoabsorption
- 2) photoexcitation and fluorescence
- 3) photodissociation
- 4) photoionization

For some related data :

- 1) excitation and ionization energies
- 2) transition probabilities
- 3) lifetimes of excited states
- 4) others

The energy range for electron cross section data is usually 0 - 10 keV, but some higher electron energy papers are included. The wavelength range for photon cross section data is from microwave to X-ray. Most papers are concerned with infrared, visible and ultraviolet ray region.

The bibliography includes the papers published in the 20th century, from 1901 to 2000. Oldest paper in this list is given by E. R. Laird (1901) for Cl<sub>2</sub>. Oldest papers for other halogen molecules are shown in each molecules. So for this bibliography, published papers from 1901 to 1999 are compiled by alphabetical order of the first author's surname of the paper. And the references published in 2000 and plus some old papers found very recently after compilation are added as "Addenda of References for each Halogen Molecule". In total, about 190 for F<sub>2</sub>, 360 for Cl<sub>2</sub>, 140 for Br<sub>2</sub> and 240 for I<sub>2</sub> papers are compiled in the four halogen molecules bibliography.

### Organization

This report consists of four parts : introduction, the bibliography and its addenda, author index, and some comments on electron collision cross sections.

### Bibliography

In this section the complete citation for all references are given. At first following classifications are shown :

- E : Elastic collision
- R : Rotational excitation
- V : Vibrational excitation
- EX : electronic EXcitation
- D : Dissociation
- I : Ionization
- A : Attachment
- ME : MEtastable molecules
- S : electron Swarm
- O : Others (photon cross sections and the others)

All authors' initials and surname, journal name, volume, inclusive pages and year of publication are given as well as the title, and some additional information in the square bracket [ ]. E and T in the square bracket mean experiment and theory.

Bibliography for F<sub>2</sub>, Cl<sub>2</sub>, Br<sub>2</sub>, and I<sub>2</sub> are divided into two parts :

F<sub>2</sub>

Part 1. 1900 - 1999 p. 1 - 16

Part 2. Addenda of References some old papers p. 17

Cl<sub>2</sub>

Part 1. 1900 - 1999 p. 1 - 29

Part 2. Addenda of References published in 2000, plus some  
old papers p. 30 - 32

Br<sub>2</sub>

Part 1. 1900 - 1999 p. 1 - 14

Part 2. Addenda of References published in 2000, plus some  
old papers p. 14 - 14

I<sub>2</sub>

Part 1. 1900 - 1999 p. 1 - 23

Part 2. Addenda of References published in 2000, plus some  
old papers p. 24 - 24

#### Author Index

In this section all authors are listed alphabetically by surname. After each author's name is a list of page numbers indicating which references he or she authored or coauthored.

|                 |    |       |              |
|-----------------|----|-------|--------------|
| F <sub>2</sub>  | p. | 1 - 4 |              |
| Cl <sub>2</sub> | p. | 1 - 7 | Addenda p. 8 |
| Br <sub>2</sub> | p. | 1 - 4 |              |
| I <sub>2</sub>  | p. | 1 - 4 |              |

Each author index of four halogen molecules follows each bibliography.

Some Comments on Electron Collision Cross Sections for F<sub>2</sub>, Cl<sub>2</sub>, Br<sub>2</sub>  
and I<sub>2</sub>

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Fluorine Molecule  $F_2$

References for F<sub>2</sub> (1900 - 1999)

(Fluorine)

[Halogen]

|                             |                                    |
|-----------------------------|------------------------------------|
| E : Elastic collision.      | R : Rotational excitation.         |
| V : Vibrational excitation. | EX : Electronic excitation.        |
| D : Dissociation.           | I : Ionization.                    |
| A : Attachment.             | QT : Grand total cross section.    |
| S : Swarm.                  | $\alpha$ : Ionization coefficient. |
| O : The others.             | [ ] : Additional informations.     |
|                             | E : Exp. T : Theory.               |

The oldest paper in this list is given by H. G. Gale (1924).

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 Über eine photometrische Untersuchung der stärksten Emissionsbanden von Fluor. [E,  $h\nu$ , F<sub>2</sub>]
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 Sov. J. Plasma Phys. 6, 748-751 (1980) -  
 Electron kinetic coefficients in a weakly ionized plasma with high attachment rates. [T, F<sub>2</sub> + He]
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 On the ab initio determination of higher-order force constants at nonstationally reference geometries. [T, F<sub>2</sub>, N<sub>2</sub>, N<sub>2</sub>O, F<sub>2</sub>O]
- O D. Andrychuk : J. Chem. Phys. 18, 233-233 (1950)  
 Raman spectrum of fluorine. [E,  $h\nu$ , F<sub>2</sub>; 892 cm<sup>-1</sup>]
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 The Raman spectrum of fluorine. [E, F<sub>2</sub>; B<sub>0</sub> = 0.8828 cm<sup>-1</sup>, r<sub>0</sub> = 1.418 Å]
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 Computer calculation of neutral-radical densities in a CF<sub>4</sub> electron cyclotron resonance plasma processing system. [T, F<sub>2</sub>, CF<sub>4</sub>]
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 Electron attachment to halogens.  
 [E, F<sub>2</sub> - I<sub>2</sub>; thermal electron attachment]
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 Molecular resonance phenomena. [review, F<sub>2</sub>, Cl<sub>2</sub>, I<sub>2</sub>, H<sub>2</sub>, N<sub>2</sub>, O<sub>2</sub>, NO, HCl]
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 Dissociative attachment in HCl, DCl, and F<sub>2</sub>. [T, F<sub>2</sub>, HCl, DCl]
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 The ultra-violet absorption spectra of some gaseous alkali-metal halides  
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 Temperature dependence of the continuous ultraviolet absorption spectrum  
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 Dissociation of fluorine molecules by electron impact.  
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 The heat of dissociation of N<sub>2</sub> and the appearance potentials of some ions  
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 [review, F<sub>2</sub>, HF, COS, CS<sub>2</sub>, CO<sub>2</sub>, N<sub>2</sub>O, etc.]
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Chlorine Molecule  $\text{Cl}_2$

References for Cl<sub>2</sub> (1900 - 1999)

(Chlorine)

[Halogen]

|                             |                                    |
|-----------------------------|------------------------------------|
| E : Elastic collision,      | R : Rotational excitation,         |
| V : Vibrational excitation, | EX : Electronic excitation,        |
| D : Dissociation,           | I : Ionization,                    |
| A : Attachment,             | QT : Grand total cross section,    |
| S : Swarm,                  | $\alpha$ : Ionization coefficient, |
| O : The others,             | [ ] : Additional informations,     |
|                             | E : Exp., T : Theory.              |

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Bromine Molecule  $\text{Br}_2$

References for Br<sub>2</sub> (1900 - 1999)

(Bromine)

{Halogen}

|                             |                                    |
|-----------------------------|------------------------------------|
| E : Elastic collision,      | R : Rotational excitation,         |
| V : Vibrational excitation, | EX : Electronic excitation,        |
| D : Dissociation,           | I : Ionization,                    |
| A : Attachment,             | QT : Grand total cross section,    |
| S : Swarm,                  | $\alpha$ : Ionization coefficient, |
| O : The others,             | [ ] : Additional informations,     |
|                             | E : Exp., T : Theory.              |

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Iodine Molecule  $I_2$

References for I<sub>2</sub> (1900 - 1999)

(Iodine)

[Halogen]

|                             |                                    |
|-----------------------------|------------------------------------|
| E : Elastic collision,      | R : Rotational excitation,         |
| V : Vibrational excitation, | EX : Electronic excitation,        |
| D : Dissociation,           | I : Ionization,                    |
| A : Attachment,             | QT : Grand total cross section,    |
| S : Swarm,                  | $\alpha$ : Ionization coefficient, |
| O : The others,             | [ ] : Additional informations,     |
|                             | E : Exp., T : Theory.              |

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dimers and trimers. [T. I<sub>2</sub>, F<sub>2</sub>, Br<sub>2</sub>, O<sub>3</sub>, C<sub>60</sub>, etc.]

Addenda (1901 - 1999)

- K. Asagoe : Sci. Rep. Tokyo Bunrika Daigaku 2, 9- (1935a)
- K. Asagoe and Y. Inuzuka : Sci. Rep. Tokyo Bunrika Daigaku 2, 73- (1935b)
- O V. H. Dibeler, J. A. Walker, K. E. McCulloh and H. M. Rosenstock : Int. J. Mass  
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Effect of hot bands on the ionization threshold of some diatomic halogen  
molecules. [E, F<sub>2</sub> - I<sub>2</sub>, ICl, IBr]

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Some Comments on Electron Collision Cross Sections

for F<sub>2</sub>, Cl<sub>2</sub>, Br<sub>2</sub> and I<sub>2</sub>

The pioneer work on electron collision cross section set for F<sub>2</sub> is given by authors, M. Hayashi and T. Nimura in 1983a. Almost no exact experimental data for F<sub>2</sub> cross sections, because experiments are extremely difficult. Our cross section set for F<sub>2</sub> is still tentative value. Now author cannot improve the cross section set, in spite of twenty years are passed.

Now I do not like this F<sub>2</sub> cross section set by another reason. All molecules have the component M(g), M(r), M(v) and M<sub>n</sub> (n = 2), depend on the pressure and temperature condition of the experiment. In F<sub>2</sub>, there are some concentration of F<sub>2</sub>(g) component, and plus many F<sub>2</sub>(r) and small F<sub>2</sub>(v) components. On the other hand, in I<sub>2</sub> molecules, there are no concentration of I<sub>2</sub>(g), almost I<sub>2</sub> are I<sub>2</sub>(r) of large j values and plus I<sub>2</sub>(v) components at usual experimental conditions.

Many scientists requested the cross section set for Cl<sub>2</sub> to the author during these twenty years or so. The cross section sets for Cl<sub>2</sub> are given by G. L. Rogoff (1986), W. L. Morgan (1991), (1992), (1999), E. S. Aydil (1992), N. Pinhao (1995), and G. I. Font (1997). Recently, L. G. Christophorou and J. K. Olthoff (1999) compiled the cross sections for Cl<sub>2</sub>. The summary said that we need more experimental studies of cross sections and other processes in Cl<sub>2</sub>. Temperature dependence of electron collisions and other processes are interesting subject for this molecules.

Almost theoretical results of electron collision cross sections for molecules are for M(g). Theoretical studies involving the M(r) and M(v) are urgently required for many molecules.

I would like to present our two papers on F<sub>2</sub> and recent report on all molecules at the end of this report.

M. Hayashi and T. Nimura : J. Appl. Phys. 54, 4879-4882 (1983a)

M. Hayashi : Jpn. J. Appl. Phys. 22, L565-L566 (1983b)

M. Hayashi and Y. Nakamura : EMS-99, Tokyo 175-176 (1999)

# Calculation of electron swarm parameters in fluorine

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Swarm parameters of electrons have been calculated for fluorine by a Monte Carlo simulation and the Boltzmann equation method for the first time. Values of these parameters have been obtained for ratios of the electric field to the gas number density  $E/N$  from 100 to 3000 Td ( $E/p_0 = 35 \sim 1000$  V/cm Torr). Available experimental and theoretical cross sections were used for the calculations.

PACS numbers: 51.50.+v, 52.80.Dy, 52.25.Fi, 52.20.Fs

## I. INTRODUCTION

The role of negative ions in gas lasers is important.<sup>1</sup> Rare-gas fluoride lasers are being investigated extensively. Experimental values of electron transport parameters for fluorine are not available. The high chemical activity of  $F_2$  makes it difficult to obtain reliable values of the electron cross sections and the electron swarm parameters in  $F_2$ . In this paper, values of the electron drift velocity  $W$ , the characteristic energy  $eD_T/\mu$ , the mean electron energy  $\langle \epsilon \rangle$ , the Townsend's ionization coefficient  $\alpha$ , the attachment coefficient  $\eta$ , and the energy distribution functions  $f(\epsilon)$  calculated by the Boltzmann equation and a Monte Carlo simulation method are reported. The calculated range of the electric field to the gas number density  $E/N$  is from about 100 to 3000 Td.

## II. COLLISION CROSS SECTIONS

The cross sections used in this paper are shown in Fig. 1.

The theoretical values for elastic total cross sections  $q_e$  were given by Schneider and Hay<sup>2</sup> and Rescigno *et al.*<sup>3</sup> for the electron energy from about 0.03 to 14 eV. In Ref. 2, there are two calculated results for the values of  $q_e$ , one has a resonance at 1.8 eV ( $F_2$  core orbitals) and another one has no resonance ( $F_2^-$  core orbitals). The result of Ref. 3 has a resonance at about 2.2 eV. Here the values of  $q_e$ , which have a resonance, were chosen, and calculations using the values of  $q_e$  of nonresonance were not carried out. These values of  $q_e$  are almost the same as  $q_e$  in  $N_2$ .<sup>4</sup> It is quite a coincidence. The assumption was therefore made that the values for elastic momentum transfer cross section  $q_m$  were equal to the values for  $q_e$  for  $F_2$ , as is approximately the case with  $N_2$ . The values for  $q_m$  at energies greater than 14 eV were assumed to be equal to the values for  $q_m$  in  $N_2$ .<sup>4</sup>

There is one theoretical paper giving values for the vibrational excitation cross section  $q_v$ .<sup>5</sup> These values were used for these calculations. The threshold energy is 0.11 eV.

Values for electronic excitation cross sections  $q_e$  were calculated by Fliflet *et al.*<sup>6</sup> for the lowest excitation level  $a^3\Pi_u$  (threshold 3.16 eV), and by Hazi<sup>7</sup> for the two levels  $C^1\Sigma_u^+$  (threshold 11.57 eV) and  $H^1\Pi_u$  (threshold 13.08 eV). The values for  $q_e$  of triplet level  $a^3\Pi_u$  were connected smoothly and decreased rapidly with electron energy. The energy dependence of the values for  $q_e$  for the two singlet states are similar to those for  $q_e$  in  $H_2$ . The values for  $q_e$  for

the  $C^1\Sigma_u^+$  and  $H^1\Pi_u$  levels were extrapolated for higher energies.

The other  $q_e$  values for dissociative level  $A^1\Pi_u$  (threshold 4.34 eV) were assumed. There are no theoretical and experimental data for this energy level. There is no base for the determination of the peak value of  $q_e$ , so we chose about half peak value of  $q_e$  compared with the peak value of  $q_e$  for  $a^3\Pi_u$  level. The level  $A^1\Pi_u$  is a singlet state, so the electron energy dependence on  $q_e$  is spread widely, as it is at  $q_e$  for  $C^1\Sigma_u^+$ .

The values for the four  $q_e$ 's are small compared with the values for  $q_m$  and ionization cross section  $q_i$ , as shown in Fig. 1. Consequently, the influence of the values chosen for  $q_e$  on the swarm parameters is small. If the values for  $q_e$  are much larger than the values used, the effect would be important. However, the values for  $q_e$  are sensitive to the values for  $\alpha$  at low  $E/N$ .<sup>8</sup> Consequently, there is a possibility that the values for  $\alpha$  at low  $E/N$  are affected by the values of  $q_e$ . However, the values of  $\eta$  are much larger than the values of  $\alpha$  at low  $E/N$ . Therefore, we believe that the reasonable values of  $(\alpha - \eta)$  were obtained, without large influence by the values of  $q_e$ .

Values of  $q_e$  for other electronic excitation levels were

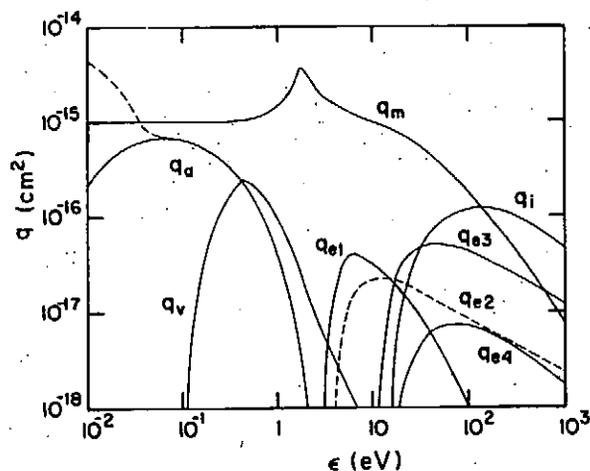


FIG. 1. Cross sections of electrons in  $F_2$  used in this paper. The dotted curve for  $q_e$  is the low energy part of the attachment cross section obtained by Chantry. The solid curve for  $q_e$  was used for this calculation. The curves for  $q_{e1}$ ,  $q_{e2}$ ,  $q_{e3}$  and  $q_{e4}$  show the excitation cross sections of  $a^3\Pi_u$ ,  $A^1\Pi_u$ ,  $C^1\Sigma_u^+$ , and  $H^1\Pi_u$  levels, respectively. The dotted curve for  $q_{e2}$  was assumed.

neglected. More detailed information about  $q_a$  is needed.<sup>9</sup>

Recently measured values of ionization cross sections  $q_i$  were used in these calculations. The data were measured by Stevie and Vasile<sup>10</sup> and covered the energy range from threshold, 15.69 eV, to about 100 eV. The values for  $q_i$  above 100 eV were extrapolated smoothly using the usual energy dependence of  $q_i$  in many gases.

There are experimental values for attachment cross section  $q_a$  given by Chantry<sup>1,11</sup> at 365 K. Calculated values for  $q_a$  are given by Hazi *et al.*<sup>11</sup> The values of Hazi *et al.* were used below about 0.1 eV. There is little effect of the two values for  $q_a$  on swarm parameters for the range of  $E/N$  which is reported in this paper. For higher energies, Chantry's values were employed. There is a maximum value for  $q_a$  and the value is  $7.0 \times 10^{-19}$  cm<sup>2</sup> at 6.2 eV. These two values for  $q_a$  are shown in Fig. 1, however, a small peak at 6.2 eV is not shown in this figure.

### III. PROCEDURES

The Monte Carlo simulation (MCS) method used in this paper has been discussed previously.<sup>12</sup>

The well-established two-term approximation was used to solve the Boltzmann equation.<sup>13</sup> The energy distribution functions of electrons  $f^0(\epsilon)$  and  $f^1(\epsilon)$  are described by the equations

$$\begin{aligned} & \frac{(\alpha - \eta)\sqrt{\epsilon}}{3} f^1(\epsilon) + \frac{eE\sqrt{\epsilon}}{3} \frac{d}{d\epsilon} [\sqrt{\epsilon} f^1(\epsilon)] \\ &= 2 \frac{m}{M} \frac{d}{d\epsilon} \left\{ \epsilon \sqrt{\epsilon} N q_m f^0(\epsilon) + kT \epsilon^2 N q_m \frac{d}{d\epsilon} \left[ \frac{1}{\sqrt{\epsilon}} f^0(\epsilon) \right] \right\} \\ &+ \sum_j [\sqrt{\epsilon + \epsilon'_j} N q'_{\alpha}(\epsilon + \epsilon'_j) f^0(\epsilon + \epsilon'_j)] \\ &+ 2 \int_{\epsilon}^{\infty} \frac{1}{\epsilon'} \sqrt{\epsilon' + \epsilon_i} N q_i(\epsilon' + \epsilon_i) f^0(\epsilon' + \epsilon_i) d\epsilon' \\ &- \sqrt{\epsilon} \left[ \sum_j N q'_{\alpha}(\epsilon) + N q_i(\epsilon) + N q_a(\epsilon) \right] f^0(\epsilon), \end{aligned}$$

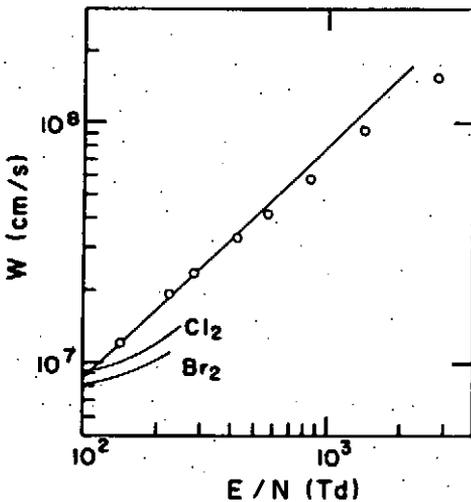


FIG. 2. Drift velocity of electrons  $W$ . Solid curve is the present calculation for  $F_2$  by  $B Eq$  and the circles are the present calculated values for  $F_2$  by MCS. Values for  $W$  in  $Cl_2$  and  $Br_2$ , taken from references, are shown for comparison.

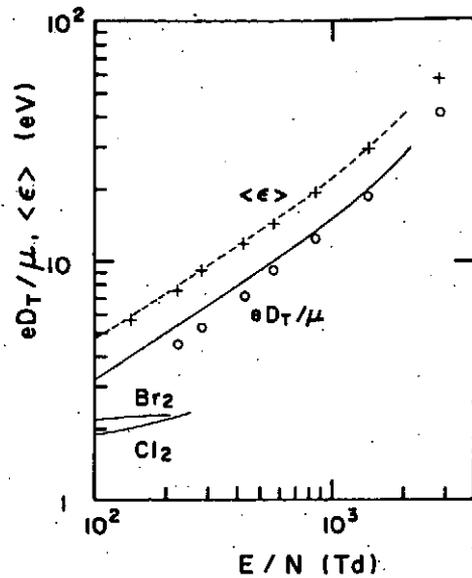


FIG. 3. Characteristic energy  $eD_T/\mu$  and mean electron energy  $\langle \epsilon \rangle$  in  $F_2$ . Solid and dotted curves are present calculated values for  $eD_T/\mu$  and  $\langle \epsilon \rangle$  by  $B Eq$  and circle and plus points are present calculated values of  $eD_T/\mu$  and  $\langle \epsilon \rangle$  by MCS, respectively. Values for  $eD_T/\mu$  in  $Cl_2$  and  $Br_2$  taken from references are also shown.

and

$$\begin{aligned} f^1(\epsilon) = & - \frac{1}{NQ(\epsilon)} \left\{ (\alpha - \eta) f^0(\epsilon) \right. \\ & \left. + eE\sqrt{\epsilon} \frac{d}{d\epsilon} \left[ \frac{1}{\sqrt{\epsilon}} f^0(\epsilon) \right] \right\}, \end{aligned}$$

where  $e$  and  $m$  are the electron charge and mass,  $M$  is the

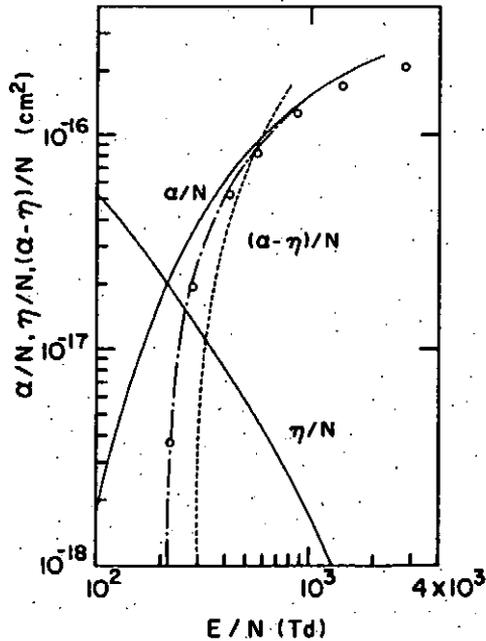


FIG. 4. Townsend's ionization coefficient  $\alpha$ , attachment coefficient  $\eta$ , and effective ionization coefficient  $(\alpha - \eta)$  in  $F_2$ . Solid curves are the present calculated values for  $\alpha$  and  $\eta$  obtained by  $B Eq$ . Dot-dashed curve is the present calculated values for  $(\alpha - \eta)$ . Circles are present calculated values for  $(\alpha - \eta)$  obtained by MCS. The dotted curve is the experimental values for  $(\alpha - \eta)$  in  $Cl_2$  and  $Br_2$  for comparison. In  $Cl_2$  and  $Br_2$ , values for  $(\alpha - \eta)$  are almost the same.

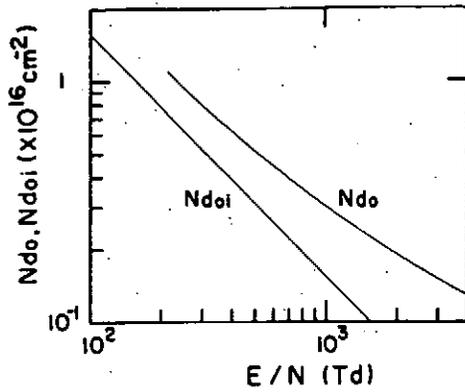


FIG. 5. Values of  $d_0$  relating to the nonequilibrium distance in  $F_2$  calculated by MCS. Straight line shows the values of  $Nd_{0i} = V_i/(E/N)$ .

mass of gas atom,  $\epsilon$  is the energy of electrons,  $k$  is the Boltzmann constant,  $T$  is the gas temperature, and  $\int_0^\infty f^0(\epsilon) d\epsilon = 1$ . The total collision cross section  $Q$  is defined by

$$Q(\epsilon) = q_m(\epsilon) + \sum q_{inel}(\epsilon),$$

where the second term on the right side denotes the summation of all the inelastic collision cross sections.  $q_{ex}$  is the excitation cross section of all kinds, including  $q_0, q_{e1}, \dots, q_{e4}$ , in this case.  $\epsilon_j^e$  is the excitation energy loss for  $j$ th kind or state of species, and  $\epsilon_i$  is the ionization energy loss.

The swarm parameters are given by

$$W = -\frac{1}{3} \sqrt{\frac{2}{m}} eE \int_0^\infty \frac{\epsilon}{NQ(\epsilon)} \frac{d}{d\epsilon} \left[ \frac{1}{\sqrt{\epsilon}} f^0(\epsilon) \right] d\epsilon,$$

$$D = \frac{1}{3} \sqrt{\frac{2}{m}} \int_0^\infty \frac{\sqrt{\epsilon}}{NQ(\epsilon)} f^0(\epsilon) d\epsilon,$$

$$\langle \epsilon \rangle = \int_0^\infty \epsilon f^0(\epsilon) d\epsilon,$$

$$v_i = \sqrt{\frac{2}{m}} \int_0^\infty \sqrt{\epsilon} f^0(\epsilon) Nq_i(\epsilon) d\epsilon,$$

$$v_a = \sqrt{\frac{2}{m}} \int_0^\infty \sqrt{\epsilon} f^0(\epsilon) Nq_a(\epsilon) d\epsilon,$$

$$\alpha = \frac{v_i}{\frac{W}{2} + \sqrt{\left(\frac{W}{2}\right)^2 - (v_i - v_a)D}}.$$

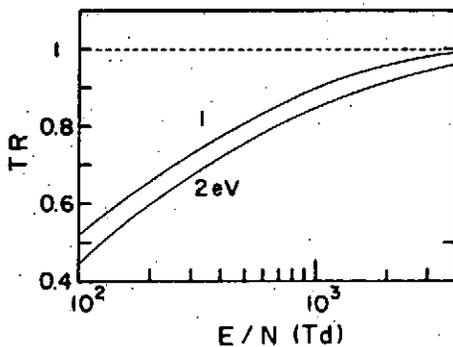


FIG. 6. Present calculated values for the transmission coefficient  $TR$  in  $F_2$  by MCS for initial energy of electrons of 1 and 2 eV. The reflection coefficient of electrons at the cathode is assumed to be 0.5.

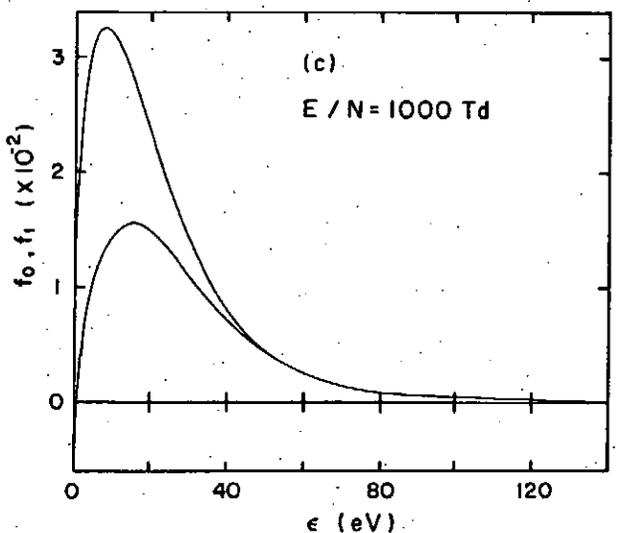
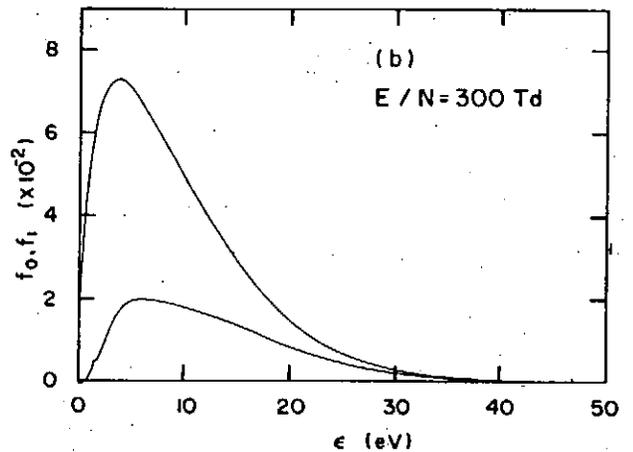
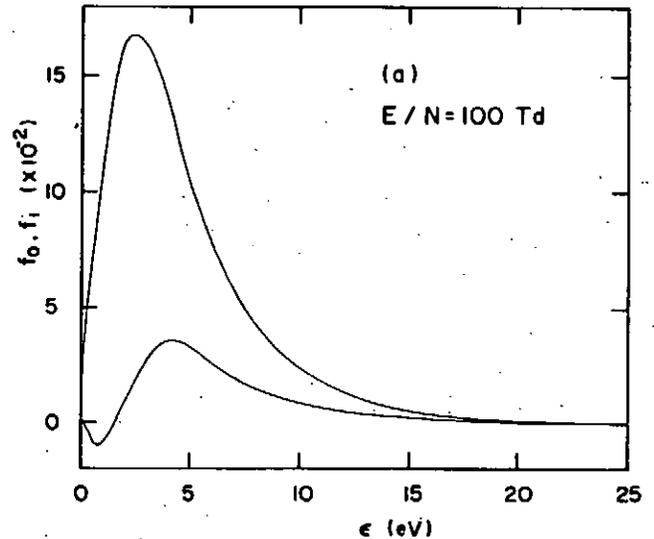


FIG. 7. Energy distribution function of electrons in  $F_2$  calculated by  $B Eq$  at  $E/N = 100, 300,$  and  $1000$  Td, respectively.

and

$$\eta = \frac{\nu_a}{\frac{W}{2} + \sqrt{\left(\frac{W}{2}\right)^2 - (\nu_i - \nu_a)D}}$$

where  $\nu_i$  and  $\nu_a$  are ionization frequency and attachment frequency, respectively.

#### IV. RESULTS

Calculated values for  $W$  are shown in Fig. 2. There are no experimental data for  $W$  in  $F_2$ . However there are some old data for  $W$  in  $Cl_2$  (Ref. 14) and  $Br_2$ .<sup>15</sup> These values are shown in Fig. 2 for comparison. The values for  $W$  at high  $E/N$  calculated by  $B Eq$  are higher than the values for  $W$  by MCS method.

The calculated values for  $eD_T/\mu$  and  $\langle \epsilon \rangle$  are shown in Fig. 3. Values for  $eD_T/\mu$  in  $Cl_2$  and  $Br_2$  are also shown in Fig. 3. The values for  $eD_T/\mu$  in  $Cl_2$  and  $Br_2$  were calculated from the equation  $eD_T/\mu = 2.48 \times 10^{-2} K$ , where  $K$  is the mean energy of agitation of an electron in terms of the mean energy of agitation of a molecule at 15 °C, given in Refs. 14 and 15.

The calculated values for  $\alpha$ ,  $\eta$ , and  $(\alpha - \eta)$  are shown in Fig. 4. Experimental values for  $(\alpha - \eta)$  in  $Cl_2$  (Ref. 16) and  $Br_2$  (Ref. 17) are also shown in Fig. 4 for comparison purposes. In  $Cl_2$  and  $Br_2$ , values for  $(\alpha - \eta)$  are almost the same.

The values  $d_0$  relating to the nonequilibrium distance<sup>18</sup> were calculated by the MCS method. The values of  $Nd_0$  as a function of  $E/N$  are shown in Fig. 5, along with the values of  $Nd_{0i} = V_i/(E/N)$ , where  $V_i$  is the voltage at which the electron runs to get the ionization energy of  $F_2$ .

The values for the transmission coefficient  $TR$  calculated by MCS are shown in Fig. 6 for the initial electron energies of 1 and 2 eV. In this case, the reflection coefficient of electrons at the cathode surface is assumed to be 0.5.

Some examples of the energy distribution functions of

electrons at an equilibrium conditions are shown in Fig. 7.

It is difficult to estimate the errors of these swarm parameters. Determination of a more comprehensive set of cross sections or experimental swarm data will provide a basis for further analysis.

#### ACKNOWLEDGMENTS

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## Calculation of Electron Attachment Rate Coefficient in Fluorine

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Electron attachment rate coefficients have been calculated for fluorine by the Boltzmann equation. The range of  $E/N$ , ratio of the electric field to the gas number density, for which the coefficients were calculated ranged from 70 to 2000 Td.

### §1. Introduction

Swarm parameters of electrons have been calculated for pure fluorine for the first time.<sup>1)</sup> In these calculations, values of the electron drift velocity  $W$  and attachment coefficient  $\eta$  were obtained. From these values, electron attachment rate coefficients  $k_a$  have been determined and compared with available experimental data in dilute fluorine as well as with calculated data. It is concluded that the present calculated values of  $k_a$  are reasonable, in spite of the fact that values of cross sections used in the calculations are incomplete.

The methods of calculation and the cross sections used in this paper are given in detail in reference 1).

### §2. Calculated Results

Calculated values of  $k_a$  are shown in Figs. 1 and 2.

Measurements of electron attachment rate coefficient studies for mixtures of gases including  $F_2$  have been made by

|                                       |                                   |
|---------------------------------------|-----------------------------------|
| Sides <i>et al.</i> <sup>2)</sup> ,   | 0.065% $F_2$ in Ar,               |
| Chen <i>et al.</i> <sup>3)</sup> ,    | 0.25% $F_2$ in $N_2$ ,            |
| Schneider and Brau <sup>4)</sup> ,    | 0.01~0.03% $F_2$ in $N_2$ and Ar, |
| Trainor and Jacob <sup>5)</sup> ,     | 0.13% $F_2$ in $N_2$ , and        |
| Nygaard <i>et al.</i> <sup>6)</sup> , | 1% $F_2$ in He.                   |

Calculated values of  $k_a$  for dilute  $F_2$  have been given by

|                                    |                            |
|------------------------------------|----------------------------|
| Hazi <i>et al.</i> <sup>7)</sup> , | $F_2$ in $N_2$ and Ar, and |
| Mitchell <sup>8)</sup> ,           | $F_2$ in He and $N_2$ .    |

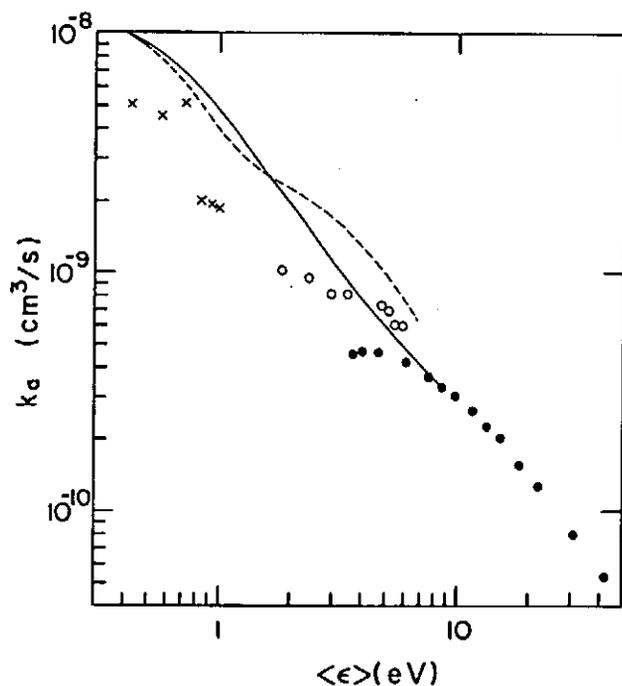


Fig. 1. Attachment rate coefficient  $k_a$  for  $F_2$  as a function of electron mean energy. ● Values in pure  $F_2$  calculated in the present work. ○ Experimental values obtained by Nygaard *et al.*<sup>6)</sup> × Experimental values given by Chen *et al.*<sup>3)</sup> —, ---- Calculated values of Mitchell.<sup>8)</sup>

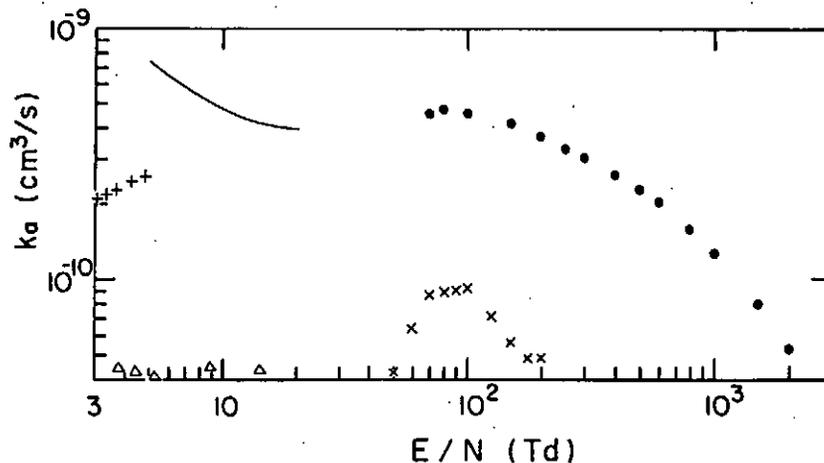


Fig. 2. Attachment rate coefficient  $k_a$  for  $F_2$  as a function of  $E/N$ . ● Present calculated values. — Nighan's experimental value. The symbols +,  $\Delta$  and  $\times$  are comparative experimental values for HCl: Christophorou *et al.*<sup>11)</sup> Sierra *et al.*<sup>12)</sup> and Davies,<sup>13)</sup> respectively.



These values have been summarized in a recent report.<sup>9)</sup> Some examples of the values of  $k_a$  given in these references are also shown in Fig. 1 ( $k_a$  versus electron mean energy  $\langle \varepsilon \rangle$ ) for comparison.

Nighan has given the only experimental determination of  $k_a$  in dilute fluorine as a function of  $E/N^{10)}$ ; the solid curve in Fig. 2 shows his results. Values of  $k_a$  for HCl measured by Christophorou *et al.*,<sup>11)</sup> Sierra *et al.*<sup>12)</sup> and Davies,<sup>13)</sup> are also shown in Fig. 2 for comparison.

From these figures, it seems that the present calculated values of  $k_a$  are reasonable. We feel that some of the cross sections used in the present calculations are uncertain. In order to get more exact values of  $k_a$ , better experimental values of cross sections are needed.

Below  $E/N=70$  Td, swarm parameters could not be calculated with our program code. It is of interest to determine whether or not electron swarms have equilibrium values of transport coefficients at low  $E/N$  in strong attaching gases.

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# May We Measure the Exact Values of Electron Collision Cross Sections for Molecules by Beam and Swarm Experiments ?

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We cannot measure the exact values of DCS for molecules intrinsically. Of course, we can measure the approximate values of DCS for molecules, but can measure the exact values of DCS for atoms. The reason is very simple.

Target molecule  $M$  in both beam and swarm experiments consists of the mixture of different states :

$$M = M(g) + M(r_j) + M(v_1) + M_N$$

where  $M(g)$ ,  $M(r_j)$ ,  $M(v_1)$  and  $M_N$  represent the completely ground state molecules, the rotationally excited molecules, the vibrationally excited molecules and the van der Waals clusters consisting of  $N$  molecules ( $N \geq 2$ ), respectively. Usually beam and swarm experiments are carried out at about 300 K. The concentration of  $M(g)$  is small compared to  $M(r_j)$  (except  $H_2$  molecules) at 300 K. Then the molecule  $M$  is always mixture of  $M(r_j)$  and  $M(v_1)$ , and the concentration of  $M(r_j)$  and  $M(v_1)$  changes with temperature. And electron collision cross section sets from elastic to inelastic collision processes for  $M(r)$  and  $M(v)$  are different each other. The target molecules  $M$  in the beam and swarm experiments are always mixtures of different molecules  $M(r_j)$  and  $M(v_1)$ . In the case of  $H_2$ , the target gas consists of  $M(g)$  and  $M(r_j)$ , especially  $M(g)$  and  $M(r_1)$  at 78 K.

Most clear change of cross sections of  $M(r_j)$  and  $M(v_1)$  will occur for triatomic molecules.  $CO_2$ ,  $N_2O$  and so on. Triatomic molecules can change from linear to bend, or vice versa easily. The authors [1] have presented the different elastic momentum transfer cross sections  $q_{mr}$  and  $q_{mv}$  for  $CO_2(r)$  and  $CO_2(v)$ , where they assumed that all other inelastic cross sections of  $CO_2(r)$  and  $CO_2(v)$  are practically the same. Then they have calculated the electron drift velocity  $W$  as a function of gas temperature  $T$ . When  $T$  increases, concentrations of  $CO_2(v)$  increases, then  $W$  decreases with  $T$  at the same  $E/N$ , the electric field over the gas number density, around 50 Td.

We have a comment to the interesting and important paper of W. Johnstone, et al. [2]. They have measured the temperature dependence of elastic DCS for  $CO_2$  at 4.0 eV. Unfortunately,  $q_{mv}$  for  $CO_2(v)$  at 4 eV is almost equal to  $q_{mr}$  for  $CO_2(r)$  [1]. Then we propose the same experiments at about 3.4 eV for  $CO_2$ , because the temperature dependence of DCS seems to be very large there.

Winstead and McKoy [3] calculated the elastic DCS for  $N_2O(g)$  at low electron energies and compare the experimental DCS data for  $N_2O(r) + N_2O(v)$  mixture at 300 K. We can see large discrepancy between them at lower than about 10 eV. We urge Winstead and McKoy to calculate the DCS

for  $N_2O(v)$  and also  $N_2O(r)$  for comparison. We can see the same discrepancy of DCS for  $CO_2$  [4] [5] at low electron energies.

$H_2$  molecules have the famous long standing controversy in the vibrational excitation cross section [6]. A possible way to solve the problem may be as follows. At first, theoreticians calculate the  $Q_{mg}$ ,  $Q_{mr}$ ,  $Q_{mv}$ ,  $Q_{rg}$ ,  $Q_{rr}$ ,  $Q_{rv}$ ,  $Q_{vg}$ ,  $Q_{vr}$ , and  $Q_{vv}$  for  $H_2(g)$ ,  $H_2(r_1)$  and  $H_2(v_1)$ , as a function of electron energies. Using these data, we calculate the electron swarm parameters. From beam experiments, we cannot determine the values of  $Q_{mg}$ ,  $Q_{mr}$ ,  $Q_{rg}$ ,  $Q_{rr}$  for  $H_2(g)$  and  $H_2(r_1)$  at the same time. It is clear that the threshold energies of  $q_r$  and  $q_v$  for  $H_2(g)$  and  $H_2(r_1)$  are different. Bhattacharyya, et al. [7] have shown that elastic integral cross sections  $q_{tr}$  are larger than  $q_{tg}$  for 20 to 200 eV for  $H_2(g)$  and  $H_2(r_1)$ . We want the elastic DCS values for  $H_2(g)$  and  $H_2(r_1)$  at low electron energies lower than 10 eV. Swarm experiments also carried out in the mixtures of  $H_2(g)$  and  $H_2(r_1)$ , except for para- $H_2(g)$  at 78 K (concentration of  $H_2(g)$  is 99.3 %). We compare the experimental and calculated swarm parameters at given conditions.

Usually, theoreticians calculate the DCS for  $M(g)$ , not for  $M(r_j)$  and  $M(v_1)$ , for most molecules. Theoretical studies involving the rotationally and vibrationally excited species are urgently required for many molecules. There is an interesting paper given by A. Jain [8].

If we have the cross section sets for excited inert gas clusters, we can calculate the electron swarm parameters of inert gases at high pressure and low temperature conditions. The concentration of the clusters for atoms and molecules are important at low temperature and high pressure conditions.

Most interesting temperature dependence occur for attachment cross sections  $q_a$  [9]-[13]. The values of non-dissociative  $q_{an}$  and dissociative  $q_{ad}$  are quite different for  $M(r)$  and  $M(v)$ , and  $M(r)$  and  $M(v)$  have the definite and individual cross sections, independent on the temperature. Apparent temperature dependence of attachment cross section is caused through different concentration of the excited components  $M(r)$  and  $M(v)$  at different temperatures.

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published in the 20th Century

| Atoms (17)                        |        | Molecules (51)                    |        |                                  |       |
|-----------------------------------|--------|-----------------------------------|--------|----------------------------------|-------|
| A + e.                            | A + hν |                                   | M + e. | M + hν.                          |       |
| He 2                              | 2170 * | 2 H <sub>2</sub> , D <sub>2</sub> | 2000   | 5 CH <sub>4</sub>                | 780   |
| Ne 10                             | 1140 * | N <sub>2</sub>                    | 2240 ○ |                                  |       |
| Ar 18                             | 1960 ○ | O <sub>2</sub>                    | 1700   | CF <sub>4</sub>                  | 390   |
| Kr 36                             | 1000   | CO                                | 1190   | CCl <sub>4</sub>                 | 210   |
| Xe 54                             | 1180 ○ | NO                                | 880    | CCl <sub>2</sub> F <sub>2</sub>  | 250   |
|                                   |        |                                   |        | CH <sub>3</sub> Cl               | 90    |
| Li 3                              | 450    | F <sub>2</sub>                    | 190 ○  |                                  |       |
| Na 11                             | 800    | Cl <sub>2</sub>                   | 360 ○  | SiH <sub>4</sub>                 | 230   |
|                                   |        | Br <sub>2</sub>                   | 140 ○  | SiF <sub>4</sub>                 | 140   |
| K 19                              | 370    | I <sub>2</sub>                    | 240 ○  | GeH <sub>4</sub>                 | 50    |
| Rb 37                             | 220    |                                   |        |                                  |       |
| Cs 55                             | 370    | HF                                | 260    | 6 C <sub>2</sub> H <sub>4</sub>  | 370   |
|                                   |        | HCl                               | 320    | CH <sub>3</sub> OH               | 350   |
| O 8                               | 390    | HBr                               | 200    |                                  |       |
|                                   |        | HI                                | 130    | 7 SF <sub>6</sub>                | 920 ○ |
| F 9                               | 90     |                                   |        |                                  |       |
| Cl 17                             | 130    | 3 CO <sub>2</sub>                 | 1240 ○ | 8 C <sub>2</sub> H <sub>6</sub>  | 260   |
|                                   |        |                                   |        | C <sub>2</sub> F <sub>6</sub>    | 150   |
| Cu 29                             | 180    | H <sub>2</sub> O                  | 930    | Si <sub>2</sub> H <sub>6</sub>   | 70    |
| Cd 48                             | 210    | H <sub>2</sub> S                  | 270    |                                  |       |
| Ba 56                             | 340    | O <sub>3</sub>                    | 480    | 9 C <sub>3</sub> H <sub>6</sub>  | 120   |
|                                   |        | N <sub>2</sub> O                  | 450    | C <sub>2</sub> H <sub>5</sub> OH | 60    |
| Hg 80                             | 600    | NO <sub>2</sub>                   | 300    |                                  |       |
|                                   |        | SO <sub>2</sub>                   | 260    |                                  |       |
|                                   |        | CS <sub>2</sub>                   | 260    |                                  |       |
|                                   |        | OCS                               | 280    | 11 C <sub>3</sub> H <sub>8</sub> | 190   |
| not final, but<br>finished mostly |        | 4 C <sub>2</sub> H <sub>2</sub>   | 390    | C <sub>3</sub> F <sub>8</sub>    | 100   |
|                                   |        |                                   |        | 12 C <sub>4</sub> F <sub>8</sub> | 100   |
| include electron<br>swarm papers  |        | NH <sub>3</sub>                   | 500    | C <sub>6</sub> H <sub>6</sub>    | 240   |
|                                   |        | NF <sub>3</sub>                   | 110    | C <sub>6</sub> F <sub>6</sub>    | 100   |
|                                   |        | BF <sub>3</sub>                   | 110    |                                  |       |
| include<br>review papers          |        | BCl <sub>3</sub>                  | 90     | 60 C <sub>6</sub> O              | 300   |
|                                   |        | PH <sub>3</sub>                   | 80     |                                  |       |
|                                   |        | H <sub>2</sub> CO                 | 180    | M <sub>r</sub> + M <sub>v</sub>  | 850   |

\* He(Ne) + e only. Not include He(Ne) + hν papers.

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