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ABSTRACT

Thomson's classical theory for the calculation of ionization cross sections in electron-atom encounters is applied for calculating the ionization cross section for atom-atom collisions. A very simple formula is obtained permitting a rapid calculation of the total ionization cross sections as a function of the kinetic energy E_a and the mass m_A of the ionizing atom, and of the binding energy E_i and the number ξ_i of equivalent electrons in the electronic shell to be ionized. The formula is in good agreement with quantum mechanical calculations and with experimental results. It can be applied to ionization from ground and from excited states. From this formula one obtains an expression for the rate coefficient for ionization in the center-of-mass system of the colliding particles. Then, the method of detailed balancing is applied to calculate the rate coefficient for ion-electron three-body collisional recombination with a neutral atom acting as the third body. This latter formula is applicable to recombination into the ground and into excited states.

INTRODUCTION

In plasma physics, only collisions of electrons with heavy particles (atoms, ions) are examined in calculating ionization and recombination as a rule. This is a limitation that is actually only permitted so long as the plasma is located in local thermal equilibrium or in the general case a high degree of ionization $x (x > 1\%)$ is attained so that reactions with neutral particles play only a subordinate role. In the case of non-thermal plasmas, however, there are many cases in which collisions of heavy particles with one another are able to influence ionization and recombination considerably. This is always the case when the reaction rates $n_a n_a \langle\sigma v\rangle_{a-a}$ become comparable or larger than the reaction rates $n_a n_e \langle\sigma v\rangle_{e-a}$. Here n_a, n_e denote the particle numbers of the heavy particles and the electrons respectively, $\langle\sigma v\rangle$ the coefficients of reaction. The case $n_a n_a \langle\sigma v\rangle_{a-a} > n_a n_e \langle\sigma v\rangle_{e-a}$ occurs when

$$\begin{pmatrix} n_a \gg n_e \\ T_a \simeq T_e \end{pmatrix} \text{ and } \begin{pmatrix} n_a \geqslant n_e \\ T_a > T_e \end{pmatrix} \text{ respectively,}$$

where T denotes the temperature. Practical cases in which these conditions are fulfilled occur, e.g., in plasma experiments with small degrees of ionization ($x < 1\%$) or in the plasma impurity [inclusion] in magnetic configurations ($T_i \cong T_a \gg T_e$).

In order to be able to examine the influence of heavy particle collisions on the state of the plasma, the corresponding effective cross sections are necessary in the region from a few electron volts to kiloelectron volts, i.e., both for the ground state and for the excited states. The heavy particle collisions have not been quantitatively examined to date at all in the calculation of non-thermal plasmas. The essential reason for this might be that no reliable effective cross sections are known. The present experimental and theoretical data are extraordinarily sparse and partially contradictory. Experiments exist for He-He, He-Ne, and Ar-Ar atom collisions as well as for N_2-N_2 molecule collisions. There is already in existence more material for the ion-atom collisions since the experiments are more easily performable.

Here we shall confine ourselves exclusively to atom-atom collisions. Quantum mechanical calculations exist only for H and Ar. To date there is no formula that makes possible a rapid estimation of the corresponding [appropriate] cross sections. Indeed, Russek and Thomas [ref. #1] have attempted to develop a purely phenomenological theory for the ionization process, however, it is to be coordinated so particularly to the atomic configuration that a general analytical expression is not possible. In this theory the ionization process is conceived as a two-step process. 1. When the two atoms approach one another, their atom shells meet one another in an electrostatic reaction. One part of the kinetic energy of the atoms is converted into internal energy of the electrons by "friction." This energy is statistically distributed among the electrons of the outer shell. 2. Then, the possibility that an electron acquires more energy E than corresponds to the ionization energy E_i of this electron in its special shell is calculated on a statistical basis. When $E > E_i$, ionization occurs.

The ionization mechanism is regarded as an "evaporation of electrons" from the atomic bond. This theory based on classic considerations was further worked out in two other works [ref. #2,3].

With respect to the excitation of electrons in atom-atom collisions, Firsov [ref. #4] has developed a similar theory in which the impulse transfer [momentum] from the colliding atom to the electron shell is calculated.

A classic theory was developed by Gryzinski [ref. #5,6] for the ionization cross section on collision of an electron with heavy particles; this theory has led to the expression of the ionization cross section in an easily plottable formula. Only a classic theory of scattering with respect to the laboratory is mentioned for atom-atom collisions [ref.#6].

In full generality, the ionization in atom-atom collisions can only be treated within the framework of quantum mechanics. Bates [ref. #7], however, expressly points out that in the practical performance of calculations so many approximations and omissions must be introduced that the accuracy of the calculations is considerably affected. In particular, one encounters difficulties that are hardly overcome in the region of small kinetic energies. These difficulties increase all the more for heavier atoms.

Considering this situation, therefore, it was attempted to obtain an analytical expression which on a simple basis makes it possible to calculate according to size the ionization cross sections for atom-atom collisions, can be used for all atoms, and leads particularly to an expression of recombination into the excited states in dense neutral gases. Since the velocity distribution in the center-of-mass system is to be used for the calculation of the rate coefficient for ionization $\langle \sigma v \rangle_{a-a}$, it is practical to relate the ionization cross sections to the center-of-mass system.

ATOM-ATOM COLLISIONS

We consider two atoms A^0 of mass m_A which can be in the ground state and when colliding lead to an ionization of *one* atom corresponding to the reaction:

$$A^0 + A^0 \rightarrow A^0 + A^+ + e \tag{1}$$

With respect to this process, quantum mechanical considerations [ref. #7, 8] lead to the following three general expressions:

(i) At very high kinetic energies E_a of the colliding particles, the effective cross section decreases in proportion to $1/E_a$.

(ii) For kinetic energies that lie at or below a certain threshold, the effective cross section is equal to zero.

(iii) The maximum of the effective cross section lies at an energy E_a which is larger than the energy E_e, where the maximum of the corresponding effective cross section of the electron-atom collision is located, by a factor from $m_A/2m_e$ to m_A/m_e, relative to the center-of-mass system.

These three expressions immediately suggest transfering the method of calculation from the ion cross section for electron-atom collisions mentioned by Thomson [ref. #9] to atom-atom collisions. If Thomson's result has already been used, then it is enough to transform his equation and the threshold energy into the center-of-mass system of heavy particle collisions.

If the kinetic energy of the electrons E_e is introduced in [into] units of the ionization energy E_i, where $U_i = E_e/E_i$, then Thomson's formula with Elwert [ref. #10] can be written as follows:

$$\sigma_{e-a} = 4\pi a_0^2 \left(\frac{E_1^H}{E_i}\right)^2 \xi_i \frac{U_i - 1}{U_i^2}, \tag{2}$$

where E_1^H = ionization energy of a hydrogen atom (13.58 eV) and ξ_i = the number of equivalent electrons in the shell to be ionized with the ionization energy E_i. For excited states, ξ_i is to be set at 1. The ionization threshold is at $E_e = E_i$. The energy dependence of σ_{e-a} is shown in Figure 1. The maximum is at $E = 2E_i$.

In the transformation from electron to atom collisions, first to be examined is the fact that the threshold is not at $E_a = E_i$ as in electron collisions, but at $E_a = (M + M)E_i/M = 2E_i$, since in collisions the energy $E_a^{labor}\, M/(M + M)$ is first transfered to the quiet atom and only the remaining amount that is then still above $2E_i$ is available for ionization. Below, the kinetic energy of the atoms with respect to the center-of-mass system is understood as E_a. If the new variable $(E_a - E_i)$ is introduced, then the threshold of the ionization cross section as a function of $(E_a - E_i)$ is at E_i. With introduction of dimensionless energy variables $W_i = (E_a - E_i)/E_i$, the threshold is put towards [along] $W_i = 1$. The variable W_i thus plays the same role as U_i in electron collisions.

For the energy transfer to the electron bound in the atom, there is only available the excess quantity $\Delta E_a/E_i$ lying above the threshold $W_i = 1$, so as in electron collisions for the ionization, only the energy excess $\Delta E_e/E_i$ that lies above the threshold $U_i = 1$ can be used for the ionization. Hence, U_i and W_i can be written as follows:

$$U_i = (E_i + \Delta E_e)/E_i \quad \text{and} \quad W_i = (2E_i - E_i + \Delta E_a)/E_i.$$

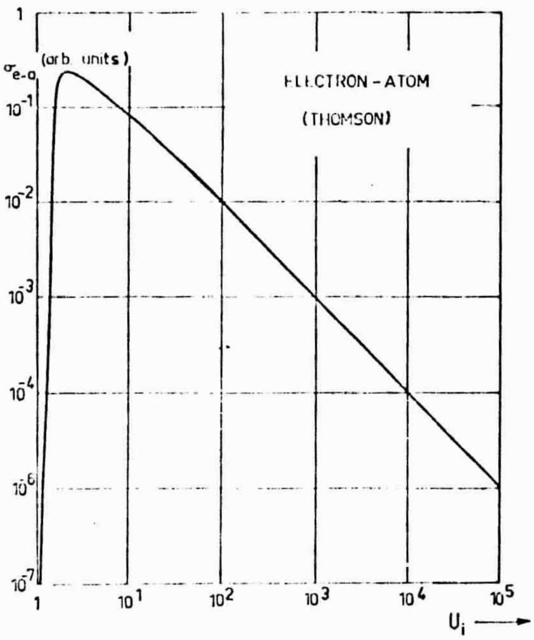

Fig. 1. Energy dependence of Thomson's effective cross section after equation (2)

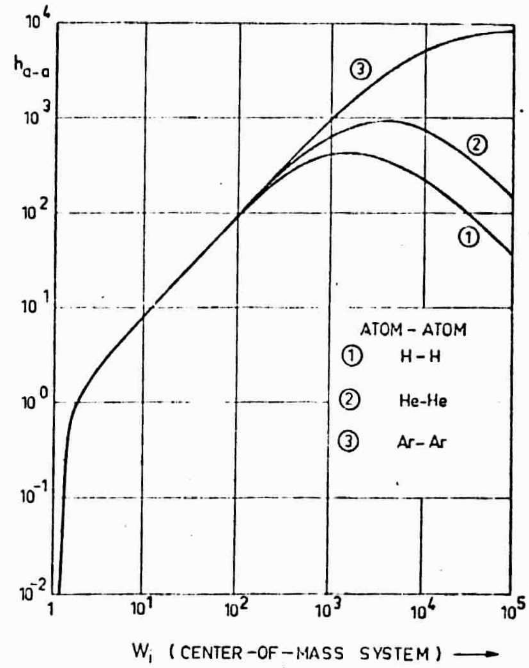

Fig. 2. Reduced effective cross sections for atom-atom collisions, equation (5)

Due to the unfavorable mass ratios $m_e/(m_A + m_e)$, U_i and W_i are identical with respect to the energy transfer to the bound electron when $\Delta E_e m_e/(m_e + m_e) = \Delta E_a m_e/(m_A + m_e)$. This leads to the following relationship between U_i and W_i:

$$U_i = 1 + \frac{2m_e}{m_A + m_e}(W_i - 1). \tag{3}$$

Insertion into equation (2) and reduction of the effective cross section to mass m_H of the hydrogen atom yields the following formula for the total ionization cross section for atom-atom collisions:

$$\boxed{\sigma_{a-a} = 4\pi a_0^2 \left(\frac{E_1^H}{E_i}\right)^2 \frac{m_A}{m_H} \cdot \xi_i \frac{2m_e}{m_A + m_e} \frac{W_i - 1}{\left[1 + \frac{2m_e}{m_A + m_e}(W_i - 1)\right]^2}}. \tag{4}$$

The function

$$h_{a-a} = \frac{W_i - 1}{\left[1 + \frac{2m_e}{m_A + m_e}(W_i - 1)\right]^2} \tag{5}$$

can be designated as the *reduced effective cross section.* The curve of this equation (3) is reproduced in Figure 2. It satisfies the three quantum mechanical expressions above. This is of course just an accident, it may be mentioned, however, that the original Thomson formula equation (2) is in contradiction to the quantum mechanical calculations inasmuch as it deals with electron-atom collisions. However, it proves to be true for atom-atom collisions (forbidden dipole transition).

According to its derivation, equation (4) should be utilizable for all atoms. We now compare the numerical values with the currently known effective cross sections.

Figure 3 shows this comparison with the values calculated from by Bates and Griffin [ref. #11] for hydrogen. Curve 3 refers to the case in which excitation of the neutral atoms still occurs in addition to the ionization. Curve 2 shows the effective cross section for ionization without change of the configuration of the neutral atom on the right side of reaction (1). The maximum deviation of the curves from one another amounts to approximately 40 per cent.

Figure 4 shows the comparison with experimental data for He. Curve 2—the measurements by Hayden and Utterback [ref. #12], curve 3—the values of Solov'ev *et al.* [ref. #13], see also [ref. #12]. The agreement of equation (4) with the experiment is surprisingly good. The maximum deviation of the curves amounts to approximately 30 per cent, generally it is less.

Figure 5 shows the effective cross section for the collision of two argon atoms. Curve 2—the quantum mechanical results by Rosen [ref. #14]; curve 3—the measurements by Rostagni [ref. #15] (see also [ref. #12]); curve 4—the experimental results by Hayden and Amme [ref. #16]. The measurements by Afrosimov *et al.* [ref. #17] are shown in curve 5. The experimental values show considerable discrepancies which are especially pointed out by Hayden and Amme. The numerical values of equation (4) (curve 1) are not in contradiction with the experiment, however, they do lie partially above and partially below the measured values.

Of course, measurements for N_2 do exist. In this case the following should be inserted into equation (4):

$$E_i = 15.6 \text{ eV}; \quad \xi_i = 2 \cdot 3 = 6; \quad m_A = 28 \cdot m_H.$$

The values calculated from equation (4) are reproduced in curve 1 of Figure 6. Curves 2 and 3 are measured values of Amme and Hayden [ref.#18].

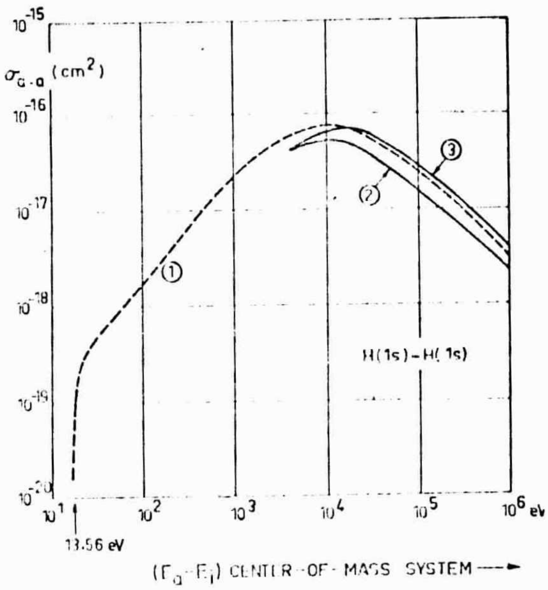

Fig.3. Effective cross section for hydrogen in ground state, curve *1* after eq. (4) with E_i = 13.58 eV, ξ_i = 1, $m_A = m_H$. Curves *2* and *3* after [ref. #11].

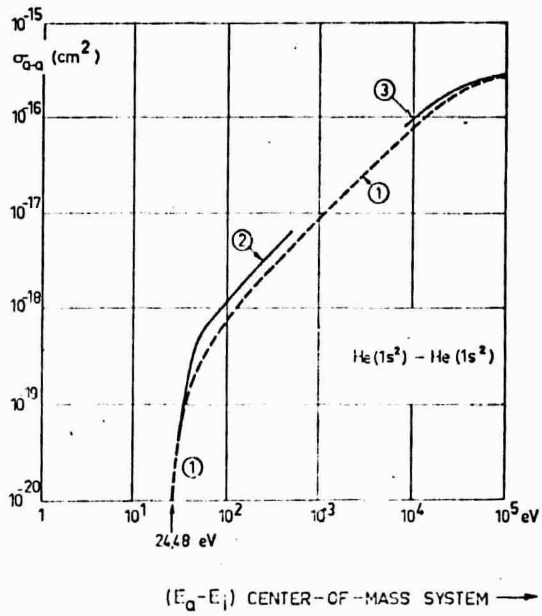

Fig. 4. Effective cross section for helium in the ground state. Curve *1* after eq. (4) with E_i = 24.48 eV, ξ_i = 2, $m_A = 4m_H$. Curves *2* and *3* experimentally after [ref.#12] and [ref. #13] respectively.

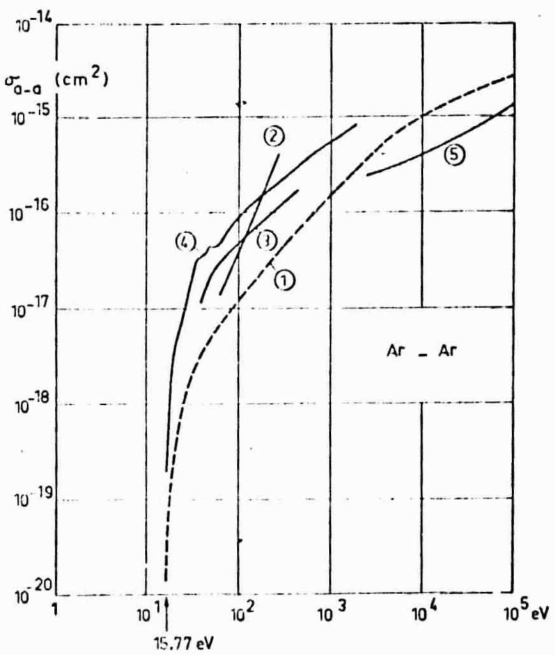

Fig. 5. Effective cross section for argon in the ground state. Curve *1* after eq. (4) with E_i = 15.77 eV, ξ_i = 6, $m_A = 40m_H$. Curves *2-5* after [ref.# 12-17].

Below 15.6 eV ionization may no longer occur. The authors explain that the measurements points of curve 3 are probably caused by rapid metastable $\vec{N}_2^*$ molecules which originate in the production of neutral particles in the charge-exchange chamber corresponding to the reaction

$$N_2^+ + N_2 \rightarrow \vec{N}_2^* + N_2^+$$
$$N_2^+ + \vec{N}_2^*$$

Curve path 3 is then to be considered as a partial ionization cross section for the excited metastable $\vec{N}_2^*$ molecules and tends toward a threshold line at lower energy values. According to these authors [ref. #18] only curve branch 2 comes into question for the ground state of the N_2 molecules. The numerical values of equation (4) agree with it.

According to the present state of our knowledge, equation (4) can be considered a useful formula for estimating the effective cross section. It is quite simple to use. If the atom to be ionized is located in a hydrogen-like state of primary quantum number n and the colliding atom in the ground state—or conversely—, then the effective cross section assumes the even simpler form

$$\boxed{\sigma_{a-a}=8\pi a_0^2 n^4 \frac{m_e}{m_H} \frac{W_i-1}{\left[1+\frac{2m_e}{m_A}(W_i-1)\right]^2}}, \qquad (6)$$

where m_e compared with m_A was omitted in the denominator.

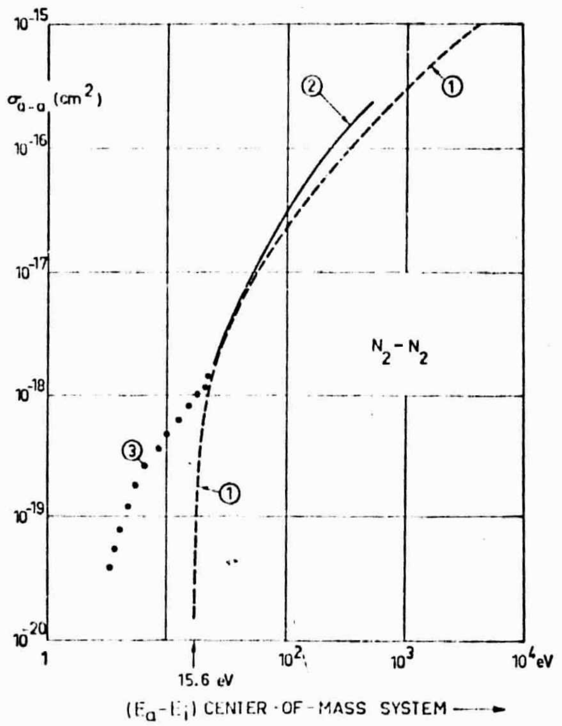

Fig. 6. Effective cross section for nitrogen molecule N_2 in the ground state. Curve *1* after eq. (4) with E_i = 15.60 eV, ξ_i = 6, m_A = 28 m_H. Curves *2* and *3* after [ref. #18].

Equations (4) and (6) might correctly reproduce especially the energy dependence at small energies, thus there where the quantum mechanical calculations and the experiments have the power to make only inaccurate data. This is, however, just the energy region important for plasma physics. One may expect no details such as, e.g., discontinuities in the effective cross section from equation (4) or (6). This is also by no means required for plasma physics and particularly for plasma spectroscopy since it must be integrated over the velocity distribution in order to arrive at the actually important value, i.e., the reaction coefficient as a function of the temperature. In this integration, all units of the effective cross section are again lost.

RATE COEFFICIENT FOR IONIZATION

In plasma physics, the rate coefficient for ionization $\langle\sigma v\rangle_{a-a}$ is needed. It is attained through integration of the effective cross section over the velocity distribution $f(v)dv$. In the center-of-mass system it holds true for like particles of mass m_A in the case of a Maxwell distribution that

$$f(v)\,dv=4\pi\left(\frac{m_A}{2\pi kT}\right)^{\frac{3}{2}} v^2 \exp\left(-\frac{m_A v^2}{4kT}\right) dv, \qquad (7)$$

and for the reaction coefficient it follows that

$$\langle \sigma v \rangle_{a-a} = \int_{v=2v_i}^{\infty} \sigma_{a-a} v f(v)\, dv,$$

where $v = 2v_i$ is to be taken as the lower boundary. This integration was performed on the IBM 360/50. The rate coefficient for ionization can then be written as follows:

$$\langle \sigma v \rangle_{a-a} = 32\pi a_0^2 \left(\frac{E_1^H}{E_i}\right)^2 \left(\frac{kT}{\pi m_A}\right)^{\frac{1}{2}} \xi_i \frac{m_e \cdot m_A}{m_H (m_A + m_e)} \Psi_{m_A}(\omega_i). \tag{8}$$

The dimensionless function $\Psi_{m_A}(\omega_i)$ is shown in the table as a function of the dimensionless variables $\omega_i = E_i/kT$ for three different masses of m_A. It can be approximated by the following expression in the region $0 \leqslant \omega_i \leqslant \infty$:

$$\Psi_{m_A}(\omega_i) = \frac{1 + \dfrac{2}{\omega_i}}{1 + \left(\dfrac{2m_e}{(m_e + m_A)\,\omega_i}\right)^2} \cdot \exp(-\omega_i). \tag{9}$$

Since eq. (4) should also be valid for excited levels, this of course occurs for the function Ψ. For highly excited levels, E_i becomes very small, and E_i/kT can fully reach the order of magnitude from 10^{-5} to 10^{-6}.

$\omega_i = E_i/kT$	$\Psi_{m_A}(\omega_i)$ $m_A = m_H$	$m_A = 4m_H$	$m_A = 10m_H$
1.0, −6	1.68, 0	2.73, 1	2.68, 3
1.0, −5	1.68, 1	2.73, 2	2.36, 4
1.0, −4	1.67, 2	2.40, 3	1.86, 4
1.0, −3	9.15, 2	1.86, 3	1.99, 3
1.0, −2	1.96, 2	1.98, 2	1.98, 2
1.0, −1	1.89, 1	1.90, 1	1.90, 1
1.0, 0	1.10, 0	1.10, 0	1.10, 0
2.0, 0	2.70, −1	2.70, −1	2.70, −1
4.0, 0	2.74, −2	2.74, −2	2.74, −2
6.0, 0	3.30, −3	3.30, −3	3.30, −3
8.0, 0	4.19, −4	4.19, −4	4.19, −4
1.0, 1	5.44, −5	5.44, −5	5.44, −5
1.2, 1	7.16, −6	7.16, −6	7.16, −6
1.4, 1	9.50, −7	9.50, −7	9.50, −7
1.6, 1	1.26, −7	1.26, −7	1.26, −7
1.8, 1	1.69, −8	1.69, −8	1.69, −8
2.0, 1	2.26, −9	2.26, −9	2.26, −9
2.5, 1	1.49, −11	1.49, −11	1.49, −11
3.0, 1	9.98, −14	9.98, −14	9.98, −14
3.5, 1	6.66, −16	6.66, −16	6.66, −16
4.0, 1	4.46, −18	4.46, −18	4.46, −18
5.0, 1	2.00, −22	2.00, −22	2.00, −22
6.0, 1	9.04, −27	9.04, −27	9.04, −27
7.0, 1	4.08, −31	4.08, −31	4.08, −31
8.0, 1	1.84, −35	1.84, −35	1.84, −35
9.0, 1	8.37, −40	8.37, −40	8.37, −40
1.0, 2	3.79, −44	3.79, −44	3.79, −44
1.2, 2	7.79, −53	7.79, −53	7.79, −53
1.4, 2	1.60, −61	1.60, −61	1.60, −61
1.5, 2	7.27, −66	7.27, −66	7.27, −66
1.6, 2	3.29, −70	3.29, −70	3.29, −70
1.7, 2	1.49, −74	1.49, −74	1.49, −74

Table. The function $\Psi_{m_A}(\omega_i)$ for different masses of m_A.

Read: 2.74, -2 = $2.74 \cdot 10^{-2}$, etc.; m_H is the mass of the H atom.

RATE COEFFICIENT FOR RECOMBINATION

The rate coefficient for ionization equation (8) now permits the calculation of the reaction rate $n_+ n_e n_a \cdot Q_{a-a}$ for the three-body collisional recombination in dense neutral gases

$$A^+ + A^0 + e \rightarrow A^0 + A^* \tag{10}$$

where A^* can denote an atom in the ground state or in an excited state. For the reaction $A^*+e+e \rightarrow A^*+e$ analogous to reaction (10), the reaction coefficient Q_{a-a} has long been known.

This is not yet the case for reaction (10). If the method of detailed balancing is applied to the two processes (1) and (10), then the following formula is obtained for the recombination of an electron into a level with ionization energy E_i and statistical weight g_i:

$$Q_{a-a} = \frac{g_i}{2g_+}\left(\frac{m_A + m_e}{m_e m_A}\right)^{3/2} \frac{h^3}{(2\pi kT)^{3/2}} \langle\sigma v\rangle_{a-a} \exp(E_i/kT), \tag{11}$$

where g^+ is the statistical weight of the ground state of ion A^+. [weight = specific density]

In equation (11) the exponential factor is made prominant owing to $\langle\sigma v\rangle_{a-a} \sim \exp(-E_i/kT)$. The term $\langle\sigma v\rangle_{a-a}$ is the expression given by equation (8). It is seen that in the case $n_a \gg n_e$ the three-body collisional recombination following reaction (10) can be more important for excited levels than the normal three-body collisional recombination in which an electron acts as a third particle. The population numbers of the highly excited levels then align themselves along the neutral particles and not along the electrons.

VALIDITY BOUNDARIES; POSSIBILITY OF MULTIPLE COLLISIONS

In a weakly ionized gas ($n_e = n_+ \ll n_a$), the mean distance ρ_0 of two neutral atoms A^0 in the ground state is given by $4\pi\rho_0^3 n_a/3 = 1$. The two-body collisional ionization (reaction (1)) of an excited atom in the n-th Bohr orbit through collision with an atom in the ground state is thus only prevalent so long as—in the classic Bohr pattern—the corresponding sphere with radius $a_0 n^2$ is still less than ρ_0. The same thing is true then for the three-body collisional recombination following reaction (10). Thus, it further follows that with respect to ionization and recombination, equations (6) and (11) are prevalent only for such levels n for which the following inequality is true:

$$n^2 < \frac{1}{a_0}\left(\frac{3}{4\pi n_a}\right)^{1/3}. \tag{12}$$

If $a_0 n^2$ becomes very much greater than ρ_0, then multiple collisions of higher order, e.g., following the reaction

$$A^0 + A^*(n) + A^0 \rightleftarrows A^0 + A^+ + e + A^0 \tag{13}$$

or more generally *cooperative effects* must gain in significance. The probability for these reactions at a given density n_a is a function of the quantum number n. These collisional probabilities have not yet been investigated to date.

In this connection it may be mentioned that Füchtbauer *et al.* (for a summary, see e.g., Drawin [ref. #19] determined the scattering cross section for electron-atom collisions at very low energies by the fact that the widening and displacement of spectral lines was investigated for which the following condition was true:

$$n^2 > \frac{1}{a_0}\left(\frac{3}{4\pi n_a}\right)^{1/3} \tag{14}$$

It would have to be possible to comprehend the effect of the reaction probability or recombination in multiple collisions in a modified experiment through measurement of the absolute intensity of a high number of terms as a function of the particle n_a, when a weak, but measurable ionization is maintained simultaneously in the gas.

I thank Miss Lombard for the programming of the function Ψ.

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