# Transport Coefficients and Energy Distributions of Electrons in Gases*

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The steady-state energy distribution of free electrons in a gas subjected to a constant electric field has been simulated stochastically. The effects of the thermal motion of the gas molecules, inelastic collisions, and the energy dependence of the collision cross sections have been included. Under appropriate limiting conditions, the stochastically generated results reduce to the analytical results of Maxwell, Davydov, and Druyvestian. Use of this technique, along with values of cross sections reported in the literature, has made possible the calculation of the drift velocities and diffusion coefficients of electrons in helium, hydrogen, and nitrogen as a function of the ratio $E/P$. The calculated results agree well with experimental values given in the literature over a range of $E/P$ from 0.002 to 0.20 V/cm mm Hg.

## I. INTRODUCTION

THE energy distribution and transport coefficients of free electrons in a gas under the influence of an external electric field have been the subject of intensive investigation. Analytical expressions for the distribution function have been obtained by Druyvestian[1] and by Morse, Allis, and Lamar[2] in the absence of inelastic collisions and thermal motion of the gas molecules, and by Davydov,[3] who included the effects of molecular motion. Approximate account has been taken of inelastic collisions by Smit,[4] and by Druyvestian and Penning,[5] and the distribution function in a high-frequency discharge has been described by Holstein.[6] In addition, Carleton and Megill[7] have solved the Boltzmann equation for the electron distribution function in crossed magnetic and electric fields.

If the electron distribution function can be found it is possible to compute transport coefficients and related average quantities which may be compared with experiment. In this manner, estimates have been made of the drift velocities, diffusion coefficients, mean energies, and Townsend ionization coefficients of electrons in a number of gases.[8–13] Furthermore, it has been shown that it is possible to use transport coefficient data to deduce cross sections which are consistant with observation.[14–16] Most of these investigations are based on the Lorentz approximation, an assumption whose validity has been questioned,[13] and in some cases make use of the Townsend energy factor $D/u$, which has also been shown to be approximate.[17] The present investigation is based on a stochastic technique similar to those employed by Yarnold[18] and Wannier[19] in the study of the motion of ions, and by Itoh and Musha[20] in the computation of the drift velocity of electrons in helium. The present method takes into account the thermal motion of the scattering molecules, the bending of the electron trajectories by the electric field, and the variation of the collision cross section with energy along an individual electron free path. This technique readily permitted the calculation of the energy distribution, drift velocity, and diffusion coefficient of electrons in a diatomic gas as a function of the experimental parameter $E/P$ over a range of energies from thermal to the threshold for vibrational excitation. In addition to providing a simple method for solving the Boltzmann equation, this stochastic approach has also made possible the computation of a number of related physical quantities without additional labor. Thus, for example, information concerning the fraction of energy absorbed by rotational states, the mean cosine of the collision angle, the mean electron kinetic energy, fluctuations in the drift velocity, and the steady-state momentum balance has also been obtained.

The system under investigation consists of a gas contained between two large uniform parallel plates across which is applied a constant electric field. It is assumed that the density of electrons is sufficiently low that electron-electron collisions can be neglected. It is well known that, since the electrons can gain large amounts of kinetic energy between collisions as a result of the accelerating action of the field, and since they can

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lose only a small fraction of their total kinetic energy in an elastic collision, the electrons may be far from thermal equilibrium with the gas.

We would like to find the steady-state electron velocity distribution function $f(\mathbf{v})$ for such a system. The form of the Boltzmann equation which describes the steady-state electron velocity distribution is[21,22]

$$\mathbf{a}\cdot\frac{\partial f}{\partial \mathbf{v}}=N\int\int\int\int\int[f(\mathbf{v}')f(\mathbf{V}')-f(\mathbf{v})f(\mathbf{V})]\times g\sigma(g,\psi)d\mathbf{V}d\Omega, \quad (1)$$

where $\mathbf{V}$ is the velocity of the gas molecules, $\mathbf{v}$ is the velocity of the electrons, $\sigma(g,\psi)$ is the differential cross section for all types of scattering events, and $N$ is the number of gas molecules per unit volume. From the electron velocity distribution function the drift velocity can be determined:

$$\mathbf{v}_D=\int\int\int \mathbf{v}f(\mathbf{v})d\mathbf{v}. \quad (2)$$

## II. CALCULATION OF ENERGY DISTRIBUTION AND DRIFT VELOCITY

Rather than attempt to solve Eq. (1) numerically for the general case, a stochastic procedure was developed by which it was possible to generate the required distribution function and drift velocity by simulating the motion of the individual electrons. In these calculations the orbit of the electron between collisions was computed classically, and the length of the orbit, the free path, was determined from elastic and inelastic cross sections reported in the literature.[16,23–25] The range of $E/P$ investigated was such that rotational excitation and deexcitation of the diatomic molecules was the only inelastic process which needed to be considered. Cross sections for these inelastic events were based on the formalism of Gerjouy and Stein,[25] using quadrupole moments of $0.49ea_0^2$ for $H_2$ and $-1.10ea_0^2$ for $N_2$.[26] Lane and Geltman[27] have shown that scattering in $H_2$ in the energy range being considered is almost entirely $s$-wave; hence the angular distribution of scattered electrons was taken as isotropic. The possibility of electron loss from the system was not considered, thus restricting the results to gases in which electron attachment is not important. Two methods were used to compute the electron energy distribution and transport coefficients with satisfactory results. In the first method (method A) the energy and direction change in an elastic collision was determined by selecting a collision partner from an isotropic Maxwellian velocity distribution, transforming to center-of-mass coordinates, selecting polar and azimuthal scattering angles from an isotropic distribution, and returning the final electron velocity to the laboratory system. The new electron trajectory was followed to the point of the next collision which was determined by computing the optical path traversed by the electron. This procedure gave satisfactory results for nitrogen, but was found to require excessive computing time for gases in which inelastic collisions did not occur or occurred only infrequently. In these instances an alternative procedure (method B) was employed which was quite useful in accelerating convergence of the stochastic program.

In the case of an elastic collision where scattering is isotropic in the center-of-mass frame of reference, and where the struck particle has a Maxwellian kinetic-energy distribution at temperature $T$ and the incident particle has a specified kinetic energy, Wigner and Wilkins[28] have calculated the Boltzmann-equation kernel, which gives the kinetic-energy distribution of the incident particle after the collision. In the special case where the mass of the incident particle (electron) is much less than the mass of the struck particle (atom or molecule), it has been shown that the energy distribution of the incident particle after numerous multiple collisions is determined by the mean energy change and the mean-square energy change of the incident particle in a single collision.[29,30] Thus, to calculate the energy change of the electron in an elastic collision using method B, an approximate form of the Wigner-Wilkins kernel which had the same mean energy change and mean-square energy change as the exact kernel was used in the computer program. Then, using standard stochastic procedures,[31] the electron energy after the collision was determined by a random number. In addition, the point of the next collision was determined by considering the collision cross section to be constant over an individual path. Itoh and Musha[20] have discussed a method which could be used as an alternative in the case of rapidly changing cross sections. Finally, it was found that for $\gamma=m/M\ll 1$ and for a fixed temperature, the drift velocity and energy distribution function depends upon the two dimensionless groups

$$A=e^2E^2[\gamma k^2T^2N^2\sigma(kT)\sigma_e(kT)]^{-1}$$

and

$$B=\sigma_i(kT)[\gamma\sigma_e(kT)]^{-1},$$

where $\gamma$ is the ratio of the mass of the diffusing charged

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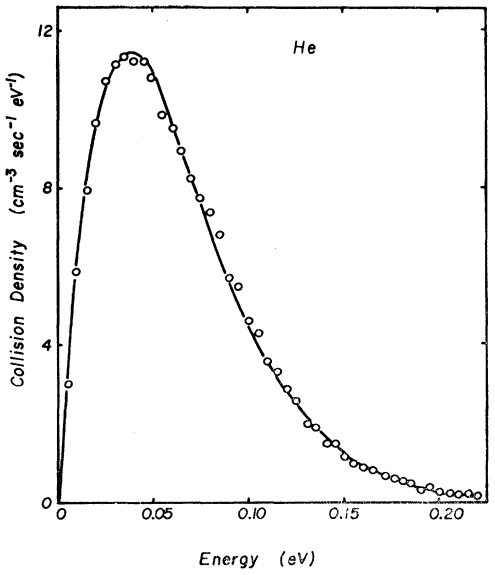


FIG. 1. Normalized collision density of electrons in He at 300°K as a function of energy for $E/P$ of 0.016 $V$/cm mm Hg. Solid curve is the analytical expression of Davydov.

particle to that of the diluent gas, and $\sigma_i$ and $\sigma_e$ are the cross sections for inelastic and elastic scattering ($\sigma_i \ll \sigma_e$), respectively. Thus, changing $\gamma$, the magnitude of the cross sections, and the field strength, but keeping $A$ and $B$ constant, did not change the drift velocity or the energy distribution function, but improved the convergence of the stochastic program. To check the procedure, energy distributions and collision densities of elastically scattered electrons were computed for various field strengths. In each case excellent agreement with the analytical expression of Davydov was obtained; a typical result is shown in Fig. 1.

The above information was used to compute stochastically the electron collision density and the drift velocity in He and $H_2$ by method B, and the diffusion coefficient for electrons in $N_2$ by method A. The electron collision density, which is the number of collisions per unit volume, time, and energy interval, is obtained directly from the stochastic computer program and may be used to calculate other quantities, such as the electron number density (which is the number of electrons per volume and energy interval, and is equal to the electron collision density divided by the electron collision frequency). The He case was primarily a test to insure that the distributions obtained in the absence of inelastic collisions agreed with the analytical expressions mentioned above. The procedure consisted of starting out a single electron with an arbitrarily assigned initial kinetic energy in a direction which was randomly selected from an isotropic angular distribution. The movement of this electron along its parabolic path in the field was followed until it has traversed a distance equal to a randomly selected free path $l$ determined from the cross-section data and a random number.[31] The energy at this point was computed and recorded in a set of equally spaced energy intervals, indicating that a collision had taken place in the interval. The distance travelled by the electron in the direction of the field and the time taken to traverse the path were also recorded. The type of collision event was determined by use of another random number and the ratios of the cross sections for the various events. If the event proved to be an elastic collision, the new energy and direction of the electron was determined by one of the two methods described above, and the entire process repeated. Runs varied from $10^4$ to $10^5$ collisions as necessary to attain the steady state.

The quantities generated by this procedure are the electron collision density and the drift velocity. A number of related quantities, such as the distribution of energy between translation and rotation, the mean cosine of the collision angle, the mean energy of the distribution, the time behavior of the drift velocity, and the momentum balance in the field direction were also recorded to provide additional information and to act as checks on the accuracy of the program.

## III. RESULTS AND DISCUSSION

The stochastic program described above was employed to compute the collision density and drift velocity of electrons in He and $H_2$ at 77 and 300°K over a range of $E/P$ from 0.002 to 0.2 V/cm mm Hg by method B. In this paper, values of $E/P$ for the stochastic results are expressed in units of $V$/cm mm Hg for an equivalent density at 273°K. Over this range of $E/P$ the collision density in He varied from very nearly a Maxwellian distribution to a Druyvestian distribution. A typical stochastically computed collision density for electrons in He at 300°K and an $E/P$ ratio of 0.016 V/cm mm Hg is shown in Fig. 1. Also shown for comparison is the analytical result of Davydov, given by the solid curve, which was obtained by numerical

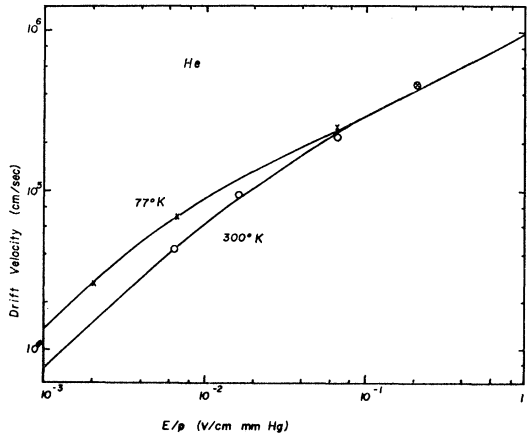


FIG. 2. Drift velocity of electrons in He as a function of $E/P$. Solid curve is taken from J. L. Pack and A. V. Phelps, Phys. Rev. **121**, 798 (1961).

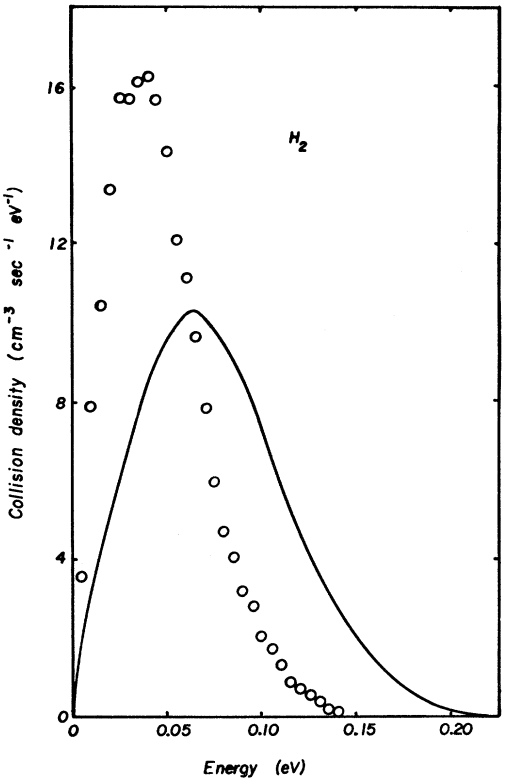


FIG. 3. Normalized collision density of electrons in $H_2$ at 77°K as a function of energy for an $E/P$ of 0.063 $V$/cm mm Hg. Solid curve is the analytical expression of Davydov, which neglects inelastic collisions.

integration using the total elastic scattering cross section of Golden and Bandel. The average energy of both the stochastic distribution and the Davydov energy distribution was computed to be 0.050 eV. The stochastic distribution was generated with $5\times10^4$ collisions using a value of $\gamma$ equal to 40 times $m_e/M_{He}$ and with the electric-field strength adjusted accordingly to keep the dimensionless parameter $A$ constant. Shown in Fig. 2 are the values of the drift velocities of the electrons in He at 77 and 300°K for a range of values of the ratio $E/P$. These drift velocities were obtained using method B with the values of the mass ratio and the square of the electric field strength increased to accelerate convergence of the stochastic program. Values of $\gamma$ that were equal to 10 to 40 times the actual value of the electron-helium atom mass ratio were used to generate the results in Fig. 2. The solid curve in Fig. 2 is taken from the experimental results reported by Pack and Phelps.[32]

Having tested the validity of the stochastic procedure by obtaining agreement with both the energy distribution and the drift velocity for He, one may now investigate these quantities in $H_2$. Shown in Fig. 3 is a typical computed collision density for electrons in $H_2$ at 77°K and $E/P$ of 0.063 V/cm mm Hg. This result was computed with $4\times10^4$ collisions and a value of $\gamma$ equal to 10 times the actual mass ratio (the cross sections and electric field strength having been adjusted to hold the dimensionless parameters $A$ and $B$ fixed). Also shown for comparison is the Davydov result, which neglects inelastic scattering. The inelastic collisions alter the collision density substantially, lowering the mean energy of the distribution to 0.039 eV from the 0.063 eV given by the Davydov expression, a result indicating the effect of inelastic collisions on an energy-dependent quantity such as the drift velocity. Figure 4 presents the stochastically generated drift velocities of electrons in $H_2$ at 77°K as a function of $E/P$. These results were obtained with a value of $\gamma$ equal to 10 times $m_e/M_{H_2}$. The computed results are again compared with the experimental data summarized by Pack and Phelps.

The electron collision density depicted in Fig. 3 is in agreement with the suggestion of Gerjouy and Stein that the large fractional energy losses observed in swarm experiments in diatomic gases are the result of rotational excitation. A further piece of information which supports this viewpoint is the fraction of energy gained from the field which is absorbed by rotational energy states; for example, this fraction has been found to be 0.5 for a value of $E/P$ of 0.020 V/cm mm Hg. The agreement of the computed drift velocity values with the experimental results indicates that the Gerjouy and Stein treatment of rotational transitions can be a useful formalism over the range of $E/P$ investigated, although investigations[14,15,26,27] over a larger range of $E/P$ have indicated that the Gerjouy and Stein cross sections may be too small by about 50%.

Similar results for the collision density and drift velocity have been obtained for electrons in nitrogen. Figure 5 presents the collision density for electrons in $N_2$ at 300°K and $E/P$ of 0.04 V/cm mm Hg. Again the shifting of the electron energy distribution toward the Maxwellian as a result of the inelastic collisions is ob-

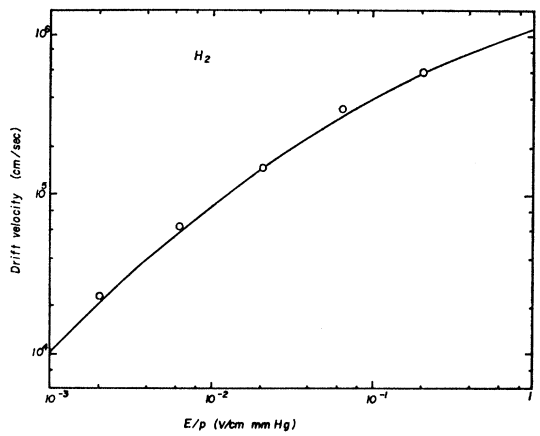


FIG. 4. Drift velocity of electrons in $H_2$ as a function of $E/P$ at 77°K. Solid curve is taken from experimental results summarized by Pack and Phelps.

[32] J. L. Pack and A. V. Phelps, Phys. Rev. **121**, 798 (1961).

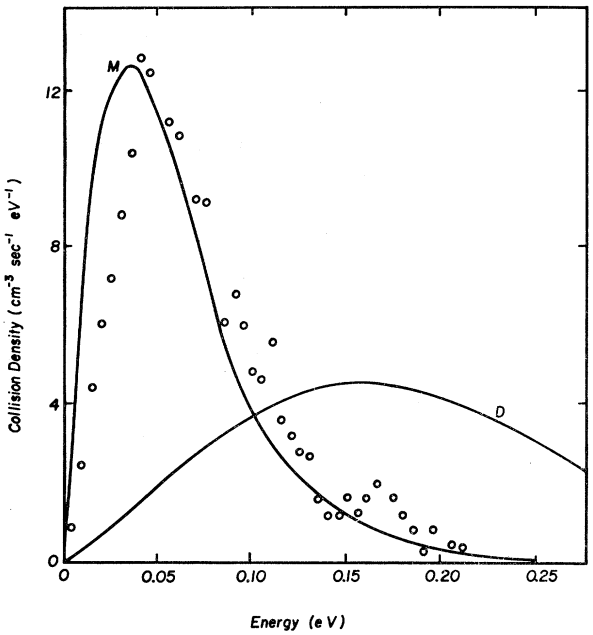


FIG. 5. Normalized collision density of electrons in $N_2$ at 300°K as a function of energy for $E/P$ of 0.04 $V$/cm mm Hg. Curve M is the Maxwell-Boltzmann distribution for 300°K and curve D is the Davydov distribution for these conditions.

served. In nitrogen more than 95% of the energy gained from the electric field is lost to rotational excitation. In addition, the diffusion coefficient in the direction perpendicular to the electric field was computed for nitrogen. This quantity was obtained by recording the mean squared distance travelled along an axis perpendicular to the electric field during a fixed time interval. From the theory of the one-dimensional random walk, one then obtains[33] $D=\langle X^2\rangle/2t$. Figure 6 presents the results of the drift-velocity computation for four values of $E/P$ in nitrogen at 300°K. The error estimates shown are the 68% confidence limits which have been obtained by assuming that the diffusion coefficient parallel to the field is equal to that perpendicular to the field and hence the variance is equal to $2D/t$. Recent experimental results indicate that this assumption may overestimate the actual error.[34]

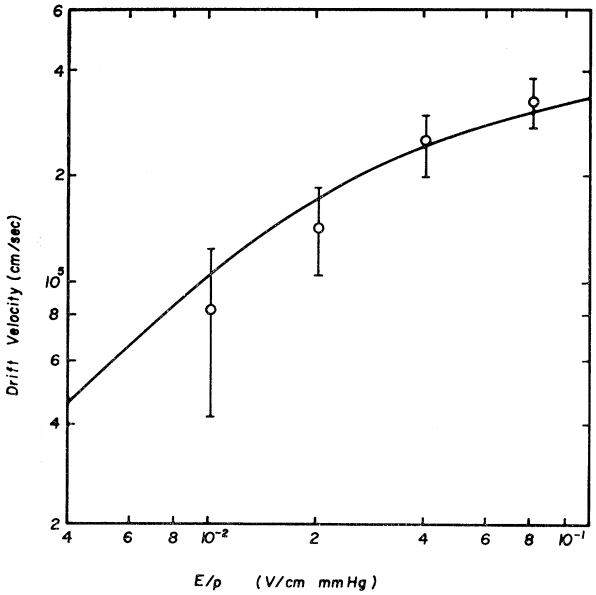


FIG. 6. Drift velocity of electrons in $N_2$ as a function of $E/P$ at 300°K. Solid curve is taken from experimental results summarized by Pack and Phelps.

The drift-velocity values shown in Figs. 2, 4, and 6 have been computed by two independent methods. The more straightforward method is the simple division of the total distance travelled by the electron parallel to the field by the time required to traverse this distance. The second involved the use of Eq. (2), which, when rewritten in terms of averages over the collision density, becomes $v_D=\langle\mu/\sigma\rangle/\langle(v\sigma)^{-1}\rangle$; here $\mu$ is the cosine of the collision angle measured with respect to the field direction. When the steady-state distribution had been obtained, the values of these two quantities differed by only a few percent, which can be attributed to stochastic fluctuations. This comparison was one of the tests used to insure that the steady state had indeed been reached. A second such test was a momentum balance in the field direction, where the net momentum gained between collisions in the direction parallel to the field was compared to the net loss of momentum along the same direction due to collisions. At steady state these two quantities differed by less than 1%.

Important additional information which may be gained from the present type of analysis concerns the influence of the electric field on the angular part of the electron velocity distribution. The individual electron paths, although randomly distributed in direction by collisions with the gas molecules, will be deflected by the electric field, and, at the time of a subsequent collision, have acquired a component of momentum parallel to the direction of the electric field. The Lorentz approximation is an attempt to represent this effect by a two-term expansion in Legendre polynomials.[2] It has been observed that this technique is invalid at extremely high $E/P$, as in the case of breakdown.[13,35] The present method provides a means of investigating the electron distribution function which does not depend on an expansion in Legendre polynomials. In fact, detailed information concerning the degree of polarization of the electron paths has been obtained in the form of the average cosine $\langle\mu\rangle$ of the angle between the direction of the force on the electron and the electron velocity at collision. This quantity for electrons in nitrogen at 300°K is given for several values of $E/P$ in Table I. The

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TABLE I. Stochastic parameters computed by method A for electrons in $N_2$ at 300°K and 55 mm Hg as a function of $E/P$.

| $E/P$ ($V$/cm mm Hg) | $v_D$ ($10^5$ cm/sec) | $D$ ($10^3$ cm$^2$/sec) | $\langle E\rangle$ (eV) | $\langle\lambda\rangle$ ($10^{-4}$ cm) | $\langle\mu\rangle$ | $f_r$ | $\langle\nu\rangle$ ($10^{10}$ sec$^{-1}$) |
|---|---|---|---|---|---|---|---|
| 0.01 | 0.84 | 3.4 | 0.038 | 9.6 | 0.002 | ... | 1.1 |
| 0.02 | 1.4 | 3.5 | 0.039 | 9.6 | 0.011 | 0.99 | 1.1 |
| 0.04 | 2.5 | 3.8 | 0.053 | 8.8 | 0.017 | 0.97 | 1.4 |
| 0.08 | 3.3 | 4.2 | 0.085 | 7.7 | 0.018 | 0.97 | 2.0 |

cosine of the collision angle is seen to be quite small in each case, indicating that a two-term expansion in $\mu$ should be adequate to represent the distribution function at these pressures and field strengths. This conclusion is also supported by the excellent agreement of the stochastic results with the Davydov distribution function for electrons in helium as demonstrated in Fig. 1, and is in agreement with earlier observation[13,35] that over the range of $E/P$ investigated the Lorentz approximation is a satisfactory assumption. The present method also provides a simple tool for extending the investigation to higher $E/P$ where the Lorentz approximation is no longer valid.

In addition to drift velocities, diffusion coefficients, and collision angles, a number of related parameters have been calculated and are also presented in Table I. These are the mean energy $\langle E\rangle$ of the stochastic distribution, the mean free path $\langle\lambda\rangle$ for the entire electron distribution, the fraction of energy $f_r$ transferred to rotational excitation, and the mean collision frequency $\langle\nu\rangle$ for the entire electron distribution.

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# Recombination of Electrons and Molecular Helium Ions

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The recombination coefficient $\alpha(N_e,T_e)$ of $He_2^+$ is measured in helium afterglow plasmas (12 Torr$\leq p\leq$20 Torr; $t\gtrsim$1 msec) as a function of electron density and gas temperature under conditions where the temperature of electrons and ions equals that of the neutral gas ($2.5\times10^{12}$ cm$^{-3}\leq N_e\leq 2\times10^{13}$ cm$^{-3}$; 900°K$\leq T\leq$2200°K). The recombination coefficient is found equal to the theoretical collisional-radiative recombination rate of $He^+$. In spite of the high vibrational excitation of the recombining $He_2^+$, corresponding to the high gas temperatures, there is no evidence for dissociative processes. In particular, the time decay of the recombination light cannot be reconciled with collisional-dissociative recombination.

## 1. INTRODUCTION

STUDIES of the recombination of electrons and molecular ions have improved the knowledge of the structure of the participating molecular particles. In most ionized gases capable of forming molecular ions the recombination has been found to proceed by the fast dissociative mechanism[1]

$$(AB)^+ + e \rightarrow A + B. \quad (1)$$

This process occurs as a result of a radiationless transition to a repulsive state of the neutral molecule, which is formed by electron capture, leading to dissociation into (possibly excited) atoms. It has been pointed out especially by Ferguson *et al.*[2] that process (1) is unlikely to play an important role in helium plasmas, mainly because the recombination rate here is much smaller, and because the afterglow light resulting from recombination shows different characteristics.

The deionization in low-pressure ($p\lesssim$1 Torr) helium afterglow plasmas containing predominantly atomic ions has already been successfully explained by the collisional-radiative recombination process[3,4]

$$He^+ + e + e \rightarrow He + e, \quad (2)$$

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[2] E. E. Ferguson, F. C. Fehsenfeld, and A. L. Schmeltekopf, Phys. Rev. 138, A381 (1965).

[3] E. Hinnov and J. G. Hirschberg, Phys. Rev. 125, 795 (1962).

[4] Reference 1, p. 253.