Collision Cross Section and Energy Loss of Slow Electrons in Hydrogen*

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Values for the collision cross section for momentum transfer and the fractional energy lost by an electron on collision with a hydrogen molecule were obtained from measurements of the microwave conductivity of a gaseous plasma. The experiments were made in the afterglow of a pulsed discharge in a cavity resonating at a wavelength of 10 cm. The mean electron energy was varied from 0.04 ev (room temperature) to 0.08 ev by heating the gas, and from 0.04 ev to 1.6 ev by microwave agitation of the electrons alone. The collision probability in molecular hydrogen at electron energies in the neighborhood of 0.04 ev is found to be $28.5(v/v_0)^{0.55}$ (cm-mm Hg)$^{-1}\pm 3\%$, where v is the electron velocity, and v_0 the most probable velocity at 300°K. Measurements in deuterium gas gave the same result. The collision probability in hydrogen increases to a peak value of 64 (cm-mm Hg)$^{-1}\pm 7\%$ at an electron energy of approximately 1.4 ev. The energy loss of electrons near room temperature was found by comparing the conductivity measurements obtained by thermal agitation with those obtained by microwave agitation of the electrons. The fraction of the excess electron energy lost on collision with the hydrogen molecule is $(3.5\pm 0.5)\times 10^{-3}$. In the higher energy range, the loss and the collision probability were computed from a comparison between microwave conductivity measurements and electron drift velocity and diffusion experiments.

INTRODUCTION

At the present time, there is some uncertainty about the value of the collision cross section of slow electrons in molecular hydrogen. At energies below 1 electron volt, techniques that employ electron beams are unreliable and various determinations of the cross section[1–5] show large discrepancies. In recent years attempts have been made to gain a better understanding of the low-energy region. Crompton and Sutton,[6] using the technique of Townsend and Bailey,[7] have measured the diffusion and mobility of electrons in dc electric fields; they calculated cross sections for electron energies from approximately 0.05 ev to 3 ev. Although microwave methods of measuring the plasma conductivity have been used with success in other gases, they have not been employed extensively in the study of the hydrogen discharge. Phelps, Fundingsland, and Brown[8] restricted their investigation to thermal electrons. Varnerin[9] continued this work, but the sensitivity of his microwave technique was insufficient to yield a reliable result for the variation of the cross section with electron energy. We have therefore repeated the measurements and extended the energy range from 0.039 ev to 1.6 ev. This paper presents the results of these investigations. In addition, the average energy loss suffered by an electron on collision with a molecule was found. The microwave method that was adopted in these investigations is similar to that used by Gould[10] and Gilardini[11] in their studies of helium and neon.

MEASUREMENTS OF PLASMA CONDUCTIVITY

The collision probability for momentum transfer is obtained from measurements of the complex microwave conductivity of a plasma. The conductivity σ is given in terms of the electron density n, the collision frequency for momentum transfer ν_m, and the radian frequency ω of the microwave field, by means of the relation

$$\sigma=\frac{ne^2}{m}\left\langle\frac{\nu_m-j\omega}{\nu_m^2+\omega^2}\right\rangle_v, \tag{1}$$

where e and m are the electronic charge and mass. Since the collision frequency is generally a function of the electron velocity v, the right-hand side of the equation must be appropriately averaged over the distribution of the electron velocities. The collision probability for momentum transfer, P_m, is defined in terms of the collision frequency by $\nu_m=P_m v p_0$, where p_0 is the gas pressure normalized to 0°C.

The measurements of the conductivity were made in a rectangular parallelepiped microwave cavity. The gas was enclosed in a small cubic quartz bottle situated at the center of the cavity in a region of nearly uniform microwave field. The gas was broken down periodically by a pulsed microwave signal that was fed into one of the three fundamental modes of the cavity. The plasma was studied in the afterglow by a microwave field in a second mode. The real and imaginary parts of the conductivity were determined from the change in the loaded Q value of the cavity and from the change of the resonant frequency of the cavity, respectively. The low-intensity probing signal produces negligible per-

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turbation of the decaying plasma. The electrons can be heated by feeding microwave power into the third mode of the cavity; the electric field strength used in heating the electrons was computed from the power input and from the Q of this mode. A detailed discussion of the measuring procedure will be found elsewhere.[10,12]

By restricting the measurements to the afterglow period, we ensured that the electrons came to a known energy equilibrium. In the absence of collisions that would lead to ionization and excitation of electronic levels, the electron distribution function can be inferred, and the averaging process in Eq. (1) can be carried out. However, the low rotational and vibrational energy states in the molecule are easily excited, even in the afterglow period. We shall find that this effect introduces an additional parameter into those measurements that rely on microwave heating of the plasma.

It will be noted from Eq. (1) that the ratio of the real to the imaginary part of the conductivity, σ_r/σ_i, is independent of the magnitude of the electron density and should, therefore, be independent of the post-discharge time at which the measurements are made. The constancy of the conductivity ratio serves as an indication of the purity of the discharge and the cleanliness of the plasma container. Before taking measurements, the following degassing procedure was carried out. The plasma container and all auxiliary equipment were baked for several days at 400°C. With the system isolated from the pumps and the gas supply by metal diaphragm valves, a holding pressure of less than 10^{-8} mm Hg was achieved. Hydrogen, obtained by decomposition of uranium hydride,[13] was then admitted into the system (deuterium was similarly obtained from uranium deuteride). Before each run a strong discharge was passed for a few hours. The spent gas was pumped out and conductivity measurements were made after admitting fresh gas. Subject to this procedure, the conductivity ratio remains constant for post-discharge times from 100 μsec to 12 msec duration.

The spectrum of the light emitted during the active discharge discloses the presence of atomic hydrogen. The relatively large cross section of atomic hydrogen (believed to be 5 to 10 times greater than that of the molecular species[14]), and its long lifetime, could lead to substantial errors in measurements in the afterglow. However, it was found that the conductivity ratio is independent of (a) the microwave breakdown power, (b) the length of the breakdown pulse, and (c) the pulse repetition rate. Thus strong support is given to the belief that the percentage of atomic hydrogen that is present is so small that it does not influence the measurements.

The experiments that were carried out can be divided conveniently into three sections. The first deals with two independent experimental methods for deriving the collision probability for low-energy electrons in the range from 0.04 to 0.08 ev. In both methods the electrons are allowed to come to thermal equilibrium with the gas molecules. In the second section we compare these results with corresponding measurements obtained by microwave agitation of the electrons alone, and obtain the average energy loss suffered by an electron when it collides with a hydrogen molecule. In the third section we extend the measurements to electron energies from approximately 0.5 to 1.6 ev and show how the data, together with Crompton and Sutton's[6] shower experiments, lead to knowledge of the collision probability and of the energy loss per collision.

EXPERIMENTAL RESULTS

1. Conductivity Ratio as a Function of Gas Pressure and Gas Temperature

The ratio of the real to the imaginary part of the microwave conductivity,[8] obtained by writing Eq. (1) in its complete form, is

$$-\frac{\sigma_r}{p_0\sigma_i}=\int_0^\infty \frac{P_m/\omega}{1+a(v)}v^4\frac{\partial f}{\partial v}dv \bigg/ \int_0^\infty \frac{1}{1+a(v)}v^3\frac{\partial f}{\partial v}dv, \quad (2)$$

where $a(v)=(P_m p_0 v/\omega)^2$, and f is the electron velocity distribution function. For gas pressures greater than 1 mm Hg, thermal equilibrium between the electrons and the hydrogen molecules is established within 100 μsec after the discharge has ceased. Henceforth, the electrons have a Maxwellian distribution, $f\sim\exp[-mv^2/2kT_g]$, which corresponds to an electron temperature, T_e, that is equal to the gas temperature, T_g.

The unknown collision probability P_m is obtained by solving the integral equation (2). However, the determination of P_m and of its velocity dependence requires more than a single measurement of the conductivity ratio. One method of attack lies in measuring this ratio

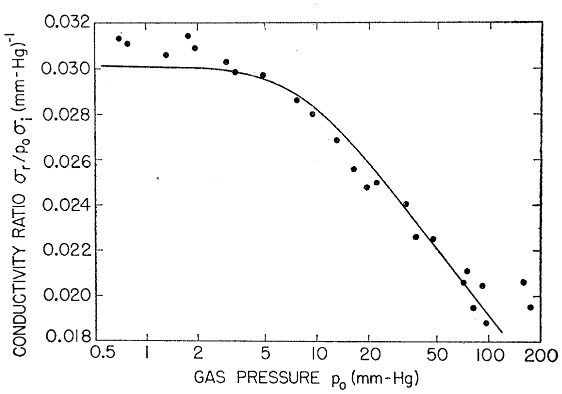

FIG. 1. Conductivity ratio as a function of the gas pressure. ● measurements; the solid line is obtained from theory for $P_m=28(v/v_0)^{0.6}$ (cm-mm Hg)$^{-1}$.

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as a function of the gas pressure at constant temperature, T_g. It can be seen from Eq. (2) that, in the limit of very low pressures $[a(v)\ll 1]$, and of very high pressures $[a(v)\gg 1]$, $\sigma_r/p_0\sigma_i$ is independent of p_0; at intermediate pressures, $\sigma_r/p_0\sigma_i$ varies with p_0 in a manner governed by the magnitude and velocity dependence of P_m. The circles in Fig. 1 represent measurements of $\sigma_r/p_0\sigma_i$ over a pressure range from approximately 1 to 200 mm Hg. Experimental difficulties prevent measurements at still higher pressures, at which $\sigma_r/p_0\sigma_i$ is expected to level off to a constant value. The collision probability P_m is obtained by fitting a theoretical curve computed from Eq. (2) to the measured values.

The integrals in Eq. (2) are difficult to evaluate unless P_m is taken to vary with v in a simple manner.[8,9,15] Therefore we assume that, in the energy range over which the integrands contribute significantly to the integral, P_m can be approximated by a power law of the form $P_m=\alpha(v/v_0)^h$, where v_0 is the most probable electron velocity at the assumed gas temperature of 300°K. The best fit with theory yields a value of $P_m=28(v/v_0)^{0.6}$ (cm-mm Hg)$^{-1}$. The result is correct only over a narrow range of electron velocities in the neighborhood of the ambient gas temperature. Similar measurements in hydrogen, carried out by Varnerin,[9] by using a wave guide in place of a cavity, gave a value of $P_m=33.6(v/v_0)^{0.6}$; but the fairly large scatter of his experimental points caused him to doubt the velocity dependence that he obtained. Phelps, Fundingsland, and Brown[8] gave a value of 46 (cm-mm Hg)$^{-1}$, which was calculated by assuming that P_m is independent of the electron energy. The very rapid disappearance of electrons reported in connection with this determination[16] suggests electron attachment to impurities that were present in the hydrogen discharge. Phelps[17] quoted the following results for the collision probability which he obtained from measurements of the electron drift velocity: $P_m=28\pm1$ at 77°K, and 29 ± 1 at 300°K.

An independent determination of P_m over the same energy range as that previously discussed can be obtained by varying the plasma temperature. This was performed by heating the cavity surrounding the plasma container to 600°K. Measurements were made in both deuterium and hydrogen, with the gas pressure kept between 1 and 3 mm Hg (see Fig. 2). By confining the measurements to low gas pressures, computations are greatly simplified; the ratio $(\nu_m/\omega)^2$ [see Eq. (1)] is then small compared with unity, and $a(v)$ in Eq. (2) can be neglected. The resulting integrals are evaluated in closed form in terms of gamma functions. Computations of the conductivity ratio were made for $P_m=28(v/v_0)^{0.6}$ by using the results of the previous experiment, and for $P_m=29(v/v_0)^{0.5}$. The former are indicated in Fig. 2 by a solid line, the latter by a dashed line. The experiments for both hydrogen and deuterium (represented as circles and triangles, respectively) agree somewhat better with the value of the collision probability indicated by the dashed line. We conclude that the collision probability for both gases is $P_m=28.5\times(v/v_0)^{0.55}$ (cm-mm Hg)$^{-1}\pm3\%$ over a limited energy range in the vicinity of 300°K.

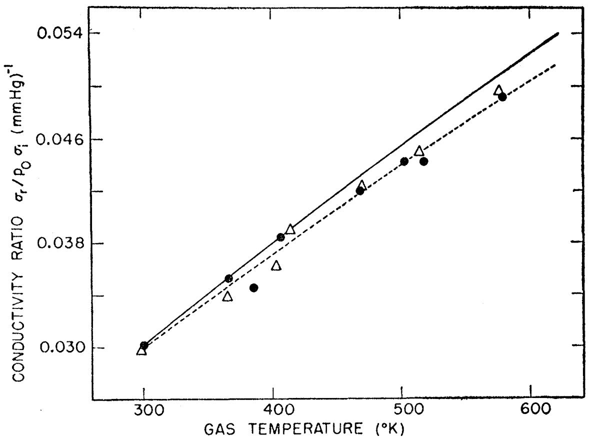

FIG. 2. Conductivity ratio as a function of the gas temperature. ● measurements in hydrogen; △ measurements in deuterium; the solid line is obtained from theory for $P_m=28(v/v_0)^{0.6}$; the dashed line is obtained from theory for $P_m=29(v/v_0)^{0.5}$ (cm-mm Hg)$^{-1}$.

2. Microwave Heating of the Plasma and Calculation of the Electron-Energy Loss

Microwave agitation of the electrons is achieved by feeding energy into a third fundamental mode of the cavity. The average equilibrium energy of the electrons depends upon the balance between the energy gained from the field and the energy-loss mechanism. We assume that the only loss present in the afterglow is the result of recoil with gas molecules and of excitation of molecular states. Under these assumptions, the velocity distribution function[18] is

$$f=C\exp\left[-\int_0^v\left\{kT_g+\frac{2e^2E^2}{3mG\omega^2[1+a(v)]}\right\}^{-1}mvdv\right]. \quad (3)$$

Here, E is the rms magnitude of the electric field used in heating the plasma, and C is a normalizing constant. $G(v)$ is the fraction of the excess energy lost, on the average, by an electron per collision with the gas molecule; for perfectly elastic collisions (such as those occurring in monatomic gases), it equals twice the ratio of the electron mass to the molecular mass $(2m/M)$. In more complex molecules, G differs from this value and may also be a function of the electron velocity. In the

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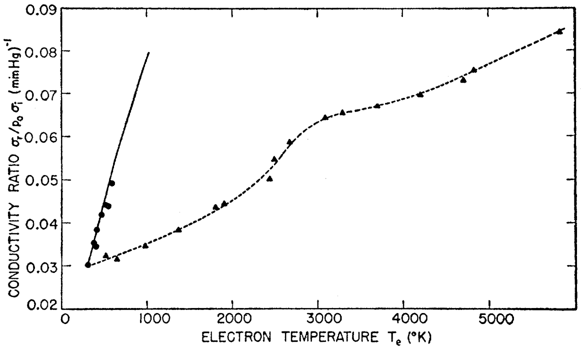

FIG. 3. Comparison between the conductivity ratio obtained by thermal agitation of the electrons (●) with that obtained by microwave agitation (--▲--▲--). The solid line is obtained from theory for $P_m=28(v/v_0)^{0.6}$ (cm-mm Hg)$^{-1}$.

latter case the variation with velocity must not be too large for Eq. (3) to apply. If G is constant and if the measurements are made at sufficiently low pressures that $(\nu_m/\omega)^2\ll 1$ (i.e., $a(v)\ll 1$), the distribution function is Maxwellian and corresponds to an electron temperature, T_e, given by

$$T_e=T_g+\frac{2e^2E^2}{3mG\omega^2k}. \tag{4}$$

Equations (3) and (4) assume implicitly that the electric field is uniform over the plasma. In the microwave cavity the electric field strength falls off approximately 10% from the center of the plasma container to its surface. We correct for this effect by averaging E^2 over the volume, V, of the container, using the relation $E^2=(1/V)\int_V E^2(V)dV$. This requires, in the present case, that E^2 which appears in Eqs. (3) and (4) be reduced by a factor 0.758, relative to the value measured at the center of the container. A much more laborious correction for the field nonuniformity is given elsewhere.[10,11] The two corrections agree within 4%.

Conductivity measurements made on a hydrogen plasma, heated by a weak microwave field, are shown in Fig. 3 by a dashed line and triangles. The electron temperature plotted along the abscissa was computed from Eq. (4) with the assumption that the G factor in hydrogen is equal to $2m/M$. That this assumption is not valid can be seen by comparing these measurements with the corresponding measurements of section 1 (shown as a solid line and circles), obtained by thermal agitation of electrons. The latter results are independent of G and thus give the correct electron temperature. The two sets of measurements can be brought into coincidence by a proper choice of G. Using Eq. (4), we find that this can be done by assigning to G the average value of $(3.5\pm0.5)\times10^{-3}$. This is to be compared with Crompton and Sutton's value of 2.5×10^{-3}. The accuracy of our determination of the excess energy loss is not good, particularly when T_e slightly exceeds T_g. This is so because G varies inversely as the difference between two nearly equal numbers, (T_e-T_g). We also assume G to be constant, independent of the electron velocity. However, the accuracy of the measurements in this energy range does not warrant a more elaborate analysis based on Eq. (3).

A quantity, more commonly used than the G factor, which is much less sensitive to temperature differences and hence less prone to experimental error, is the average energy loss per collision, λ. It is related to G by $\lambda(\bar{u}_e)=G(\bar{u}_e)[1-(\bar{u}_g/\bar{u}_e)]$; here $\bar{u}_e$ represents the average electron energy and $\bar{u}_g$ the gas energy. It will be noted that only at low electron energies does λ differ appreciably from G. A plot of λ, obtained from the results of Fig. 3, is shown in Fig. 4. Only the low-energy region [for electron velocities less than 0.35 (volt)$^{\frac{1}{2}}$] was obtained from this analysis. The remainder of the curve was computed from measurements that will be discussed in Sec. 3.

3. P_m and λ for Electron Energies from 0.5 to 1.6 Electron Volts

Conductivity measurements as a function of the microwave heating field discussed in the previous section were extended to much higher electron energies. The experimental results are shown in Fig. 5; the ratio of the conductivities is plotted against the root-mean-square value of E^2 averaged over the volume of the plasma. Throughout these measurements, the gas pressure was sufficiently low to satisfy the condition $(\nu_m/\omega)^2\ll 1$.

The collision probability was computed from Eq. (2) by using the distribution function f given by Eq. (3). We see that an additional quantity, the G factor (not present in the computations of Sec. 1), has entered into our problem. Since G is not known in the energy range in which we are now working, P_m cannot be found unless we turn to an independent experiment, which will be used in conjunction with our conductivity data.

There are two types of experiment that will serve our purpose: the dc drift-velocity measurements (v_d) of Bradbury and Nielsen[19] and of Pack and Phelps[20];

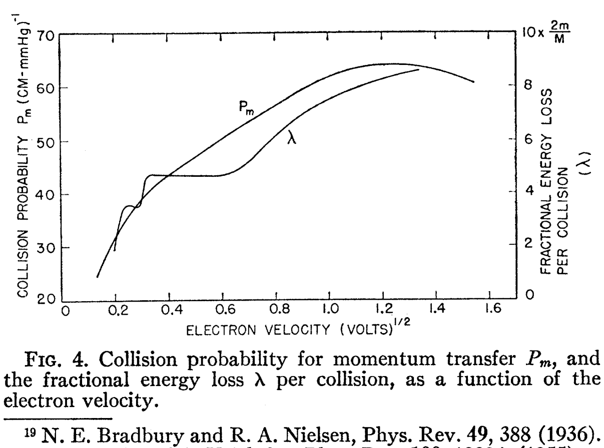

FIG. 4. Collision probability for momentum transfer P_m, and the fractional energy loss λ per collision, as a function of the electron velocity.

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[20] J. L. Pack and A. V. Phelps, Phys. Rev. 100, 1229A (1955).

and the measurements of the ratio of the diffusion coefficient to the electron mobility (DE_d/v_d, where D is the diffusion coefficient and E_d is the dc electric field) of Crompton and Sutton.[6] This ratio is, in fact, a direct measure of the average electron temperature and is exactly equal to the temperature for a Maxwellian distribution of velocities. These two experiments taken together (as Crompton and Sutton[6] did), or the latter of the two combined with the conductivity measurements (as we do here), suffice to give a complete solution for P_m and G, with no assumptions regarding the form of the electron velocity distribution function. In performing computations from these measurements we limited outselves to electron energies higher than 0.5 ev and thus ensured that $T_e \gg T_g$. We assumed that P_m and G could be represented sufficiently accurately by $P_m = \alpha v^h$ and $G = \beta v^l$ over a small range in the neighborhood of the average electron energy; α, β, h, and l are the parameters to be determined.

Subject to these conditions, the velocity distribution function given by Eq. (3), which is equally valid in the microwave and in the dc cases, takes on the simple form $f = C\exp(-Av^t)$. The coefficients A and t differ in the microwave and dc experiments. The expressions for the measured microwave conductivity, dc drift velocity, and the ratio of diffusion to mobility are:

$$\frac{-\sigma_r}{p_0\sigma_i} = \left(\frac{4+h}{3}\right)\frac{\alpha}{\omega}\int_0^\infty fv^{3+h}dv \Big/ \int_0^\infty fv^2 dv, \quad (5)$$

$$\frac{v_d p_0}{E_d} = \left(\frac{2-h}{3}\right)\frac{e}{m\alpha}\int_0^\infty f_d v^{1-h}dv \Big/ \int_0^\infty f_d v^2 dv, \quad (6)$$

$$\frac{DE_d}{v_d} = \left(\frac{1}{2-h}\right)\frac{m}{e}\int_0^\infty f_d v^{3-h}dv \Big/ \int_0^\infty f_d v^{1-h}dv, \quad (7)$$

where

$$f = \exp\left[-\frac{3\beta}{2(l+2)}\left(\frac{m\omega}{eE}\right)^2 v^{l+2}\right],$$

$$f_d = \exp\left[-\frac{3\beta}{2(l+2h+4)}\left(\frac{m\alpha p_0}{eE_d}\right)^2 v^{l+2h+4}\right].$$

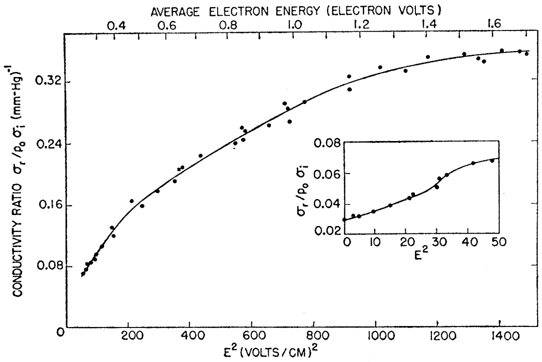

FIG. 5. Measured conductivity ratio (— ● —) as a function of the root-mean-square value of the microwave electric field used in heating the electrons. The computed average electron energy is also given.

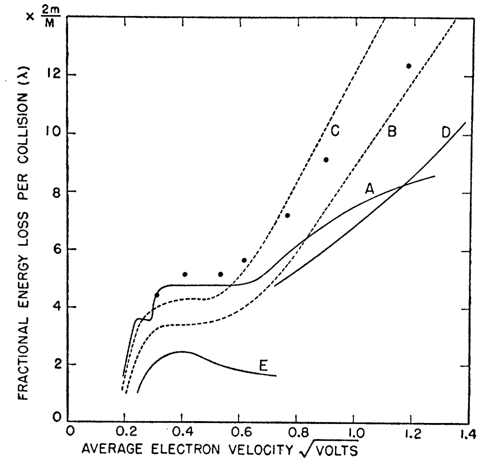

FIG. 6. The fractional energy loss of an electron per collision with a hydrogen molecule as a function of the electron velocity. Curve A, present microwave measurements; curve B, Crompton and Sutton's shower experiments for a Maxwellian distribution of electron velocities; curve C, assumed Druyvesteyn distribution; curve D, calculated with no assumptions; curve E, theoretical (Gerjuoy and Stein); the solid dots are from measurements of Townsend and Bailey.

These integrals were evaluated, and P_m and G were found by a simultaneous solution of the resulting equations. We note that Eq. (5) is given in terms of the rf electric field, while Eqs. (6) and (7) are in terms of a dc electric field. The corresponding electric fields cannot be equated by using Eq. (5) in conjunction with Eqs. (6) or (7) because the rf field causes a different energy transfer to the electrons than does the corresponding dc field. Therefore, the microwave and dc experimental results were equated at the same average electron energies ($\bar{u}$). This fact makes computation laborious, since $\bar{u}(E)$ and $\bar{u}(E_d)$ themselves depend on the electron velocity distribution function.

The method outlined above was used in calculating the average electron energy $\bar{u}$ as a function of the microwave heating field E^2. This is shown as the second abscissa of Fig. 5. Since $\bar{u}$ does not vary linearly with E^2, G must be a function of the electron energy. The collision probability P_m and the fractional energy loss λ are presented in Fig. 4 over the complete range of electron velocities that was investigated. The data for electron velocities below 0.35 (volt)$^{\frac{1}{2}}$ are taken from the measurements of Secs. 1 and 2; the values of the collision probability are accurate within $\pm 3\%$; those of λ within $\pm 7\%$. The parts of the curve which lie above 0.70 (volt)$^{\frac{1}{2}}$ represent calculations of this section. The values of P_m are good within $\pm 7\%$, and those of λ within $\pm 10\%$. The curves that lie between 0.35 and 0.70 (volt)$^{\frac{1}{2}}$ are extrapolated.

In Fig. 6, a comparison of the various determinations of the fractional energy loss λ as a function of the average electron velocity $\bar{v}$ is shown. Curve A represents our measurements. Curves B and C are Crompton and

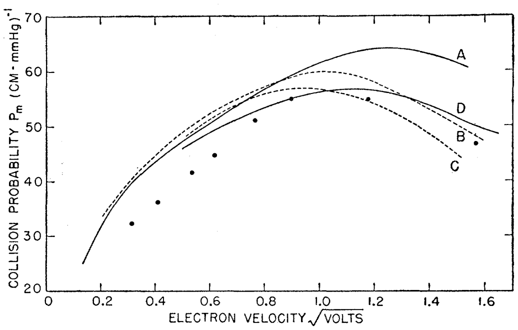

FIG. 7. Collision probability for momentum transfer as a function of the electron velocity.

Sutton's results, calculated for the case of hard-sphere electron molecule collisions ($h=0$); the former represents their calculations that were based on the assumption that the electrons have a Maxwellian distribution of velocities; the latter represents their assumption of a Druyvesteyn distribution. Curve D was calculated from Crompton and Sutton's tabulated data with the use of our Eqs. (6) and (7). This eliminates the necessity of assigning to the electrons a particular distribution function. In fact, curve D is a plot of $G(v)$ rather than of $\lambda(\bar{v})$. Curve E is theoretical[21] and based on the assumption that the electron losses arise solely from the excitation of vibrational states. In making these calculations the authors took P_m to be constant and equal to 42 (cm-mm Hg)$^{-1}$. The solid dots represent the measurements of Townsend and Bailey[7] and were calculated by using a Maxwellian distribution.

In Fig. 7 a similar comparison for the variation of the collision probability with electron velocity is shown. The lettering designating the various curves is the same as in Fig. 6. Curves A and D are plotted as a function of the electron velocity, v; the rest of the data are plotted as a function of the average velocity, $\bar{v}$. There is satisfactory agreement between our measurements and those of Crompton and Sutton. It must be stressed, however, that in the energy range above 0.7 (volt)$^{\frac{1}{2}}$, use was made of their results, and hence this agreement is better than it would have been had the calculations been made completely independently.

ACKNOWLEDGMENTS

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