Transport Collision Cross Sections from Electron Drift-Velocity Data*

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Information concerning the transport collision cross section $\sigma_t(\epsilon)$ of low-energy electrons in the noble gases was obtained from drift-velocity data. The results (with ϵ in ev) are He: $\sigma_t(\epsilon)=(28\pm1)$ cm^{-1} for $0.13\leq\epsilon\leq4$; Ne: $\sigma_t(\epsilon)=(6.05)\epsilon^{0.157}$ cm^{-1} for $0.38\leq\epsilon\leq8$; A: $\sigma_t(\epsilon)=(6.3\pm0.6)\epsilon$ cm^{-1} for $1.6\leq\epsilon\leq11$; Kr: $\sigma_t(\epsilon)=14\epsilon$ cm^{-1} for $1.6\leq\epsilon\leq3$; Xe: $\sigma_t(\epsilon)=26\epsilon$ cm^{-1} for $1\leq\epsilon\leq2.4$. All σ_t values are for 0°C and 1 mm Hg pressure. These results are compared with other experimental and theoretical data. For nitrogen the drift-velocity measurements indicate an average fractional energy loss per collision of $20(2m/M)$ in the energy range $\epsilon=0.07$ to 0.15 ev.

INTRODUCTION

REASONS for the numerous discrepancies between theoretical and experimental cross sections for very low-energy electrons are not difficult to find. On the theoretical side, simple methods of treatment are not available (the Born approximation is not valid, for example) and it is imperative that polarization and exchange effects be considered. The experimental procedures are likewise beset with difficulties which involve the production of monoenergetic electron beams and analysis of the scattered current. It is well known, however, that the difficulties inherent in beam experiments can be avoided by making measurements on electron swarms. Reference need only be made to the Townsend method[1] and to the more recent microwave techniques[2,3] as illustrative examples. In this paper, new experimental electron drift-velocity data[4] for the noble gases and nitrogen are analyzed with the objective of extracting from them conclusions concerning the transport collision cross section and the pertinent partial-wave phase shifts. A brief discussion concerning inelastic collisions involving rotational excitation of nitrogen is also presented.

METHOD

A theoretical relation between the electron drift velocity $\bar{\omega}(E)$ and the transport collision cross section $\sigma_t(\epsilon)$ is established by the following equations:

$$\begin{aligned}\bar{\omega}(E)&=(16\pi eE/m)\int_0^\infty[6mkT\epsilon\sigma_t^2(\epsilon)\\&\qquad+Me^2E^2]^{-1}\epsilon^2\sigma_t(\epsilon)f_0(\epsilon,E)d\epsilon,\\f_0(\epsilon,E)&=A\exp\left\{-6m\int_0^\epsilon[6mkT\epsilon\sigma_t^2(\epsilon)\right.\\&\qquad\left.+Me^2E^2]^{-1}\epsilon\sigma_t^2(\epsilon)d\epsilon\right\},\end{aligned}\tag{1}$$

which depend on the assumption that only elastic collisions occur. In these expressions $f_0(\epsilon,E)$ is the distribution function for electrons expressed in terms of the kinetic energy ϵ, E is the electric field strength, e and m are the electron charge and mass, M is the mass of the gas atom or molecule, T is the gas temperature, and k is the Boltzmann constant. In these equations $\sigma_t(\epsilon)$ is expressed in units of cm^{-1} at 1-mm pressure and 0°C. The function $f_0(\epsilon,E)$ is represented by the first term of a series solution to the Boltzmann transport equation.[5] Normalization is achieved by setting $4\pi\int_0^\infty v^2f_0(v)dv=1$.

The unknown functions $\sigma_t(\epsilon)$ and $f_0(\epsilon,E)$ may be regarded as solutions of the integral equations (1) in terms of the known function $\bar{\omega}(E)$. Although a general solution to this mathematical problem has not been found, particular solutions pertinent to the problem at hand are readily obtained. When $\sigma_t(\epsilon)=a\epsilon^n$, it is possible to perform the integrations indicated in Eq. (1) and thereby obtain an explicit expression for $\bar{\omega}(E)$ in terms of the unknown constants a and n, provided that the thermal motion of the gas atoms is neglected. The exponent n can be determined from the slope of the $\bar{\omega}$ *versus* E/p_0 curve at each value of E/p_0 and the constant a can then be determined from the corresponding magnitude of $\bar{\omega}$. For helium, σ_t is practically independent of ϵ, and for neon it is only slightly dependent on ϵ. For argon, krypton, and xenon $\sigma_t(\epsilon)$ is approximately proportional to ϵ except below the Ramsauer minimum.

Equations (1) cannot be applied, as they stand, to nitrogen, due to the presence of inelastic collisions which occur even at very low energy. However, if it is assumed that the average energy loss per collision is in constant ratio $\lambda(\epsilon)$ to the energy loss per elastic collision, then the factor $\lambda(\epsilon)$ can be inserted into the equations and an analysis similar to that used for the noble gases can be applied. The expression obtained in this case contains two unknown quantities, the undetermined value of $\sigma_t(\epsilon)$ and the factor $\lambda(\epsilon)$. Evaluation of the

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[1] R. W. Crompton and D. J. Sutton, Proc. Roy. Soc. (London) **A215**, 467 (1952).
[2] J. M. Anderson and L. Goldstein, Phys. Rev. **102**, 933 (1956).
[3] J. L. Hirshfield and S. C. Brown, J. Appl. Phys. **29**, 1749 (1958).
[4] J. C. Bowe, preceding paper [Phys. Rev. **117**, 1411 (1960)].
[5] S. Chapman and T. G. Cowling, *The Mathematical Theory of Nonuniform Gases* (Cambridge University Press, Cambridge, 1939), p. 347 ff.

factor $\lambda(\epsilon)$ therefore requires that $\sigma_t(\epsilon)$ be known independently.

RESULTS

Helium

If $\sigma_t(\epsilon)$ is assumed to be sensibly independent of energy and if the thermal motion of gas atoms can be neglected (i.e., $Me^2E^2 \gg 6mkT\epsilon\sigma_t^2$), then Eq. (1) takes the form $[\bar{\omega}\times(E/p_0)^{-\frac{1}{2}}]=[1/\Gamma(\frac{3}{4})](3m/M)^{\frac{1}{4}}(2\pi e/m\sigma_t)^{\frac{1}{2}}$, a constant. Experimental values of this product, which are presented in Fig. 1, show that it is indeed constant in the interval $E/p_0=0.1$ to 1.2. A least-squares analysis of the values in this interval gives $\sigma_t=28$ cm^{-1} (0°C, 1 mm) with a standard deviation of ± 1 cm^{-1}. This result is to be compared with the smaller experimental values of Anderson and Goldstein,[2] of Gould and Brown,[3,6] and of Ramsauer and Kollath as computed by Barbiere[7] (see Fig. 2). The energy region over which σ_t is constant is estimated to extend from about 0.13 ev, the average electron energy at $E/p_0=0.1$, to about 4 ev, which includes approximately 95% of the electrons at $E/p_0=1.2$. The analysis presented here yields no information for $E/p_0<0.1$, where the effects of thermal motion cannot be neglected. The experimental data were terminated at $E/p_0=1.2$ because of spurious breakdown of the gas in the chamber.

The transport cross sections are determined by the partial-wave phase shifts δ_l, i.e.,

$$\sigma_t(k)=\frac{4\pi}{k^2}\sum_{l=0}^{\infty}(l+1)\sin^2(\delta_l-\delta_{l+1})$$

(k is the wave number of the electron). Values of $\sigma_t(\epsilon)$ computed from the theoretical phase shifts δ_0 and δ_1 of Morse and Allis[8] are compared with the experimental

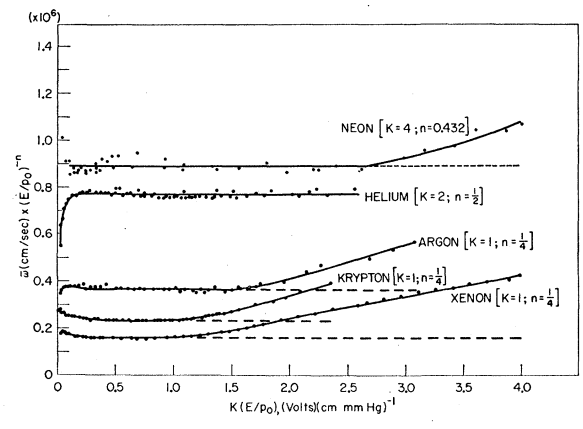

FIG. 1. These curves represent the experimental $\bar{\omega}$ data which were used to obtain $\sigma_t(\epsilon)$. When $\sigma_t(\epsilon)\approx$constant, $n=\frac{1}{2}$ and when $\sigma_t(\epsilon)/\epsilon\approx$constant, $n=\frac{1}{4}$. The dashed curves are extensions of the elastic-collision data. No interpretation was made at low values of E/p_0.

[6] L. Gould and S. C. Brown, Phys. Rev. **95**, 897 (1954).

[7] D. Barbiere, Phys. Rev. **84**, 653 (1951).

[8] P. M. Morse and W. P. Allis, Phys. Rev. **44**, 269 (1933).

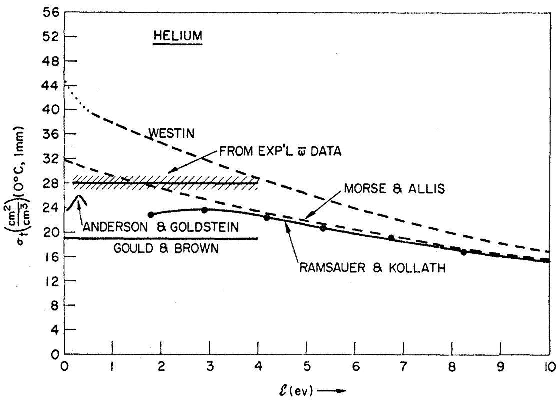

FIG. 2. The dashed curves were computed from partial-wave phase-shift data. The dotted portion of the curve labeled Westin[10] was computed by extrapolating his phase-shift data to zero energy. The Ramsauer-Kollath curve is drawn through values computed from their angular scattering measurements.[7] Gould-Brown[6] and Anderson-Goldstein[2] curves were obtained from microwave experiments. The value $\sigma_t=28\pm1$ cm^{-1} was obtained from $\bar{\omega}$ measurements.

data in Fig. 2. It is seen that the cross sections obtained from drift velocities provide the best agreement with theory yet attained at low energy. Indeed, this agreement is sufficiently good to suggest that polarization effects, which were not taken into account by Morse and Allis, are small and do not contribute significantly to $\sigma_t(\epsilon)$. Indications that polarization effects are probably small were previously noted by Moiseiwitsch,[9] but on the basis of different evidence.

A second calculation of $\sigma_t(\epsilon)$ was made using the tabulated values of δ_l which Westin[10] derived from experimental scattering data (see Fig. 2). The dotted portion of this curve ($\epsilon<0.54$ ev) was computed by extrapolating his δ_l data to the appropriate multiple of π at zero energy. Westin's δ_0 curve lies close to the theoretical curve of Morse and Allis[8] and is in exact agreement with it below 4 ev. In contrast, his δ_1 curve diverges from that of Morse and Allis below $\epsilon=50$ ev and gives much larger phase shifts at lower energy. Westin found it necessary to adopt larger values for the p-wave shift, as well as for higher order scattering, in order to account for a pronounced asymmetry in the experimental angular scattering curves. At low energy, Westin's results must be questioned on two points. First, the values of $\sigma_t(\epsilon)$ computed from his δ_l are much larger than any of the experimental values. (They are also larger than the values obtained directly from the Ramsauer-Kollath scattering data, from which his phase shifts were derived.) Second, the higher order phase shifts do not approach zero as k^2, as required by theory.[11]

[9] B. L. Moiseiwitsch, Proc. Roy. Soc. (London) **A219**, 102 (1953).

[10] S. Westin, Kgl. Norske Videnskab. Selskabs, Skrifter No. 2 (1946).

[11] Bransden, Dalgarno, John, and Seaton, Proc. Phys. Soc. (London) **71**, 877 (1958).

The discrepancy between the Morse-Allis and Westin curves in Fig. 2 is due mainly to Westin's larger values of δ_1. A substantial amount of p-wave scattering is required to obtain the minimum in the scattering amplitude at small angles which the experimental angular-distribution data of Ramsauer and Kollath exhibit. In obtaining agreement with this feature of the experimental data, however, a large amount of back-scattering is introduced, and this in turn leads to greater values of $\sigma_t(\epsilon)$. The weight of evidence contained in the experimental $\sigma_t(\epsilon)$ data rules against such large values of δ_1 and therefore questions the correctness of the small-angle scattering data of Ramsauer and Kollath.

Graham and Ruhlig[12] computed drift velocities in helium, neon, and argon using $\sigma_t(\epsilon)$ data which were derived from Westin's phase shifts. The values of $\bar{\omega}$ which they obtained in helium are about 10% to 15% lower than the author's measurements, for the reasons just given.

Neon

The transport cross section was assumed to vary with energy as $\sigma_t(\epsilon)=a\epsilon^n$ and the unknown constants a and n were determined as described previously. The values of $\bar{\omega}$ were plotted against E/p_0 on log-log graph paper and it was found that a straight line fits the data well in the interval $E/p_0=0.015$ to 0.65. The slope of the line gave a value of $n=0.157$. The value of $a=6.05$ was determined from this line and the corresponding constant value of $[\bar{\omega}\times(E/p_0)^{-0.432}]$ is shown in Fig. 1, where the experimental values of this product are also presented.

Experimental $\bar{\omega}$ values below $E/p_0=0.025$ were not interpreted, these data being of lesser reliability because of the low voltages which were used on the chamber. Above $E/p_0=0.65$, the upturn in values is brought about by an increasing $\bar{\omega}$ which is caused by inelastic collisions. An analysis of this deviation from an extrapolated "elastic-collision" curve would provide information about the inelastic-collision cross section. The electron energy at $E/p_0=0.025$ has a computed average value of 0.38 ev and at $E/p_0=0.65$ it is 8 ev. The result $\sigma_t(\epsilon)=6.05\epsilon^{0.157}$ compares favorably with the Ramsauer-Kollath and Westin curve (see Fig. 3).

The only available partial-wave phase-shift data for neon at low energy were derived by Westin[10] from experimental scattering data. These phase shifts therefore include the effects of polarization and exchange, and since they yield $\sigma_t(\epsilon)$ values which are in good agreement with the values obtained directly from the Ramsauer-Kollath[7] scattering data, it is especially interesting to determine what further information might be gained by extrapolating them to zero energy. A linear extrapolation[13] of the s- and p-wave phase shifts from $k=0.2$ to the known values at $k=0$ leads to a nearly constant $\sigma_t(k)$, as shown by the dotted curve in Fig. 3. This is in contradiction with the experimental results of Gilardini and Brown[14] and indicates a fundamental discrepancy between the two sets of data. There is no known theoretical justification which would permit extrapolation of Westin's δ_0 and δ_1 curves in a manner which would remove this difference. Therefore, the behavior of $\sigma_t(\epsilon)$ at low energy should be accepted as being uncertain until this discrepancy is resolved.

Graham and Ruhlig[12] compared their neon $\bar{\omega}$ values with the experimental results of Nielsen. The discrepancy which they observed was attributed to the existence of inelastic collisions at $E/p_0=0.25$. However, the new experimental $\bar{\omega}$ data[4] do not show evidence of inelastic collisions until about $E/p_0=0.6$, and this result is in agreement with the energy-distribution function[15] as derived from Barbiere's data. Graham and Ruhlig's $\bar{\omega}$ values are just within the upper limits of error of the new $\bar{\omega}$ measurements.

Argon

If it is assumed that $\sigma_t(\epsilon)=a\epsilon$ (which corresponds to neglect of the up-turn in σ_t below the Ramsauer

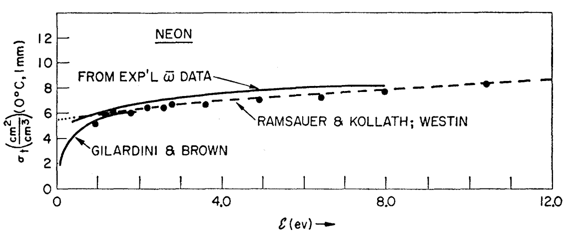

FIG. 3. The dashed curve was computed[12] from Westin's phase shifts and the points were computed[7] from the Ramsauer-Kollath scattering data. The dotted portion of the curve was obtained by linearly extrapolating Westin's s- and p-wave phase-shift curve to zero energy. The Gilardini-Brown[14] curve was obtained from microwave experiments.

minimum) and if the thermal motion of gas atoms is neglected, then Eq. (1) becomes

$$[\bar{\omega}\times(E/p_0)^{-1/4}]=0.926(m/M)^{3/8}(e/m^2a)^{1/4}$$

for all values of E/p_0. This relation is experimentally satisfied in the interval $E/p_0=0.2$ to 1.5 (see Fig. 1). A least-squares analysis of these data gave $a=6.3\pm0.6$, a value which may be assigned to the corresponding range of average energies, $\epsilon=1.6$ ev to 4.4 ev. This result falls between the Ramsauer-Kollath[7] curve and the Westin[12] curve, but definitely favors the former (see Fig. 4). The curve is arbitrarily extended to 11 ev since the effects of excitation collisions are not evident at $E/p_0=1.5$.

[12] W. J. Graham and A. J. Ruhlig, Phys. Rev. **94**, 25 (1954).

[13] If the p-wave phase shift is extrapolated to $k=0$ with zero slope as required by theory, then $\sigma_t(0)=7.7$ cm^{-1}.

[14] A. L. Gilardini and S. C. Brown, Phys. Rev. **95**, 897 (1954).

[15] Energy-distribution functions for helium, neon, and argon are tabulated in Argonne National Laboratory Report ANL-5967 (unpublished). Copies may be obtained by writing to the author.

Above $E/p_0=1.5$, the characteristic increase in $\bar{\omega}$ due to inelastic collisions is evident. Here, again, it should be possible to derive information about the inelastic cross section on the basis of the deviation from an extrapolated "elastic-collision" curve.

Graham and Ruhlig[12] computed $\bar{\omega}$ in argon over a limited range of E/p_0 extending from 0.25 to 1.0. The values which they obtained are in agreement with the author's measurements. However, their calculated energy-distribution functions indicated appreciable inelastic collisions at $E/p_0=1.0$. Experimentally, such collisions are not indicated by the $\bar{\omega}$ data until E/p_0 reaches the value of about 1.5. This discrepancy casts doubt on the correctness of Westin's phase shifts for argon. The $\bar{\omega}$ data yield no definite information about $\sigma_t(\epsilon)$ below about 1.6 ev.

Krypton and Xenon

Experimental values of $[\bar{\omega}\times(E/p_0)^{-\frac{1}{2}}]$ were found to be fairly constant for these gases over a limited range

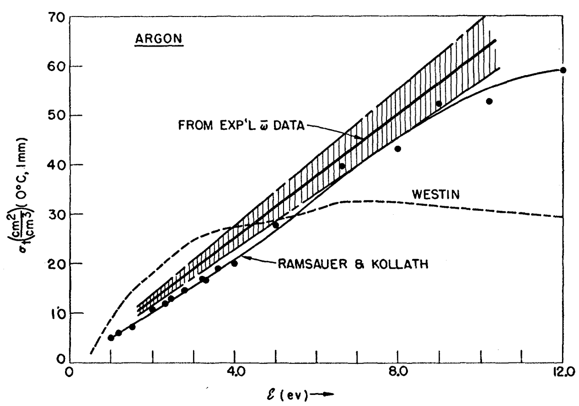

FIG. 4. The Ramsauer-Kollath curve is drawn through values computed from their scattering data.[7] The Westin curve was computed[12] from his phase shifts. The shaded area contains the result $\sigma_t(\epsilon)=(6.3\pm0.6)\epsilon$ cm^{-1} obtained from $\bar{\omega}$ measurements.

of E/p_0 (see Fig. 1) thereby indicating that the transport cross section is approximately proportional to the energy. For krypton, the average value of $[\bar{\omega}\times(E/p_0)^{-\frac{1}{2}}]$ was found to be 0.23×10^6 in the interval $E/p_0=0.3$ to 1.1. This yielded $\sigma_t=14\epsilon$ cm^{-1} over the corresponding range of average energy 1.6 to 3 ev. For xenon $[\bar{\omega}\times(E/p_0)^{-\frac{1}{2}}]$ was found to be 0.16×10^6 in the interval $E/p_0=0.2$ to 1.0. This gave $\sigma_t=26\epsilon$ cm^{-1} over the energy range 1 to 2.4 ev. Since the electrons have an energy spread about the average value, the curves in Figs. 5 and 6 were arbitrarily extended to 7 ev for krypton and 5.5 ev for xenon. These results are compared with $\sigma_t(\epsilon)$ which the author computed from the scattering data of Ramsauer and Kollath.[16] The agreement can be considered satisfactory.

On the theoretical side, Holtsmark[17] calculated values

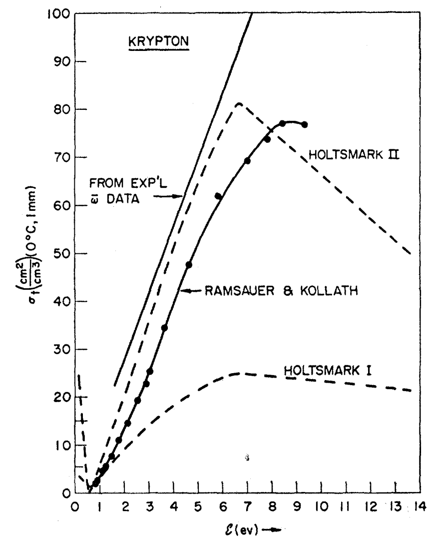

FIG. 5. The dashed curves are drawn through $\sigma_t(\epsilon)$ values computed from partial-wave phase shifts which Holtsmark[17] derived for two different polarization fields I and II. The Ramsauer-Kollath[16] curve is drawn through values computed from their scattering data. The solid curve $\sigma_t(\epsilon)=14\epsilon$ was obtained from $\bar{\omega}$ measurements.

of the total elastic collision cross section for krypton for two different polarized scattering potentials, but did not take exchange effects into account. The results which he obtained for both fields fitted the experimental total elastic cross-section data of Ramsauer and Kollath almost equally well. When the transport cross section is calculated from Holtsmark's phase shifts, it is found that only the results obtained from "atomfeld II" are consistent with the new experimental data (see Fig. 5). Unfortunately, the extent of the experimental and theoretical data presently available does not allow a closer and more detailed comparison to be made. The far greater sensitivity of the transport cross section than that of the elastic scattering cross section to the degree of polarization, however, indicates the usefulness

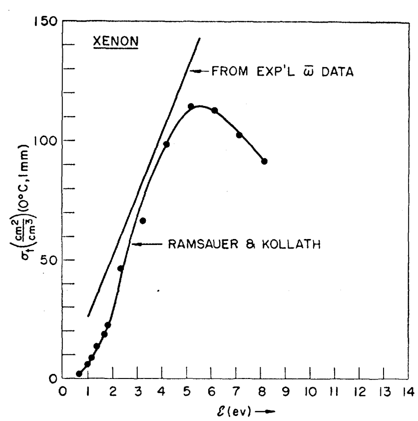

FIG. 6. The Ramsauer-Kollath[16] curve is drawn through values computed from their scattering data. The solid curve $\sigma_t(\epsilon)=26\epsilon$ was obtained from $\bar{\omega}$ measurements.

[16] C. Ramsauer and R. Kollath, Ann. Physik 12, 837 (1932).
[17] J. Holtsmark, Z. Physik 66, 24 (1930).

of $\sigma_t(\epsilon)$ to any future theoretical development of this problem. There are at present no theoretically derived phase shifts or cross sections available for xenon.

Nitrogen

Due to inelastic collisions which occur in a molecular gas, Eqs. (1) are not directly applicable in their present form. The equations can, however, be modified to include the effects of inelastic collisions by assuming that the energy losses involved in the rotational excitation of the gas molecules are comparable to the energy losses which occur when the collisions are elastic. This is accomplished by writing the energy-loss term in the energy-balance equation as $\lambda(\epsilon)(2m/M)$, where $\lambda(\epsilon)$ is a measure of the inelasticity of the impact and has the value unity when the collisions are elastic. Equations (1) are thereby modified by the appearance of the multiplying factor $\lambda(\epsilon)$ in the integrand of $f_0(\epsilon,E)$ and also in the integrand of $\bar{\omega}(E)$. If it is now assumed that $\sigma_t(\epsilon)=a\epsilon$ and that $\lambda(\epsilon)=\alpha\epsilon^{-j}$, where a, α, and j are undetermined constants, the equations can be integrated in closed form provided that the thermal motion of gas molecules is neglected. The validity of this last assumption can be judged after the computation is completed by comparing the calculated average electron energy with the thermal energy.

Integration of Eqs. (1) yields

$$\bar{\omega}(E)=(4/Mm)(2/m^3)^{-\frac{1}{2}}(\alpha)^{3/2(4-j)}\{\Gamma[3/2(4-j)]\}^{-1} \times(eE/a)^{(1-j)/4-j}[6m/(4-j)M]^{2j-5/2(4-j)}.$$

The experimental $\bar{\omega}$ *versus* E/p_0 curve has a constant slope of 0.25 in the interval $E/p_0=0.06$ to 0.3 and therefore $j=0$. Experimental values of $[\bar{\omega}\times(E/p_0)^{-\frac{1}{4}}]$ have a magnitude of $(0.54\pm0.01)10^6$ over this interval. From the measurements of Phelps *et al.*[18] and of Crompton and Sutton,[1] the value of a was estimated as 200 cm^{-1}/ev. These data therefore fix the value of α at 20 over the energy interval 0.07 ev to 0.15 ev (average energies corresponding to $E/p_0=0.06$ and 0.3, respectively). This energy range is only about 2 to 4 times the thermal energy of the gas molecules, and therefore, neglect of molecular motion is not wholly justified. On the other hand, the slope of the $\bar{\omega}$ *versus* E/p_0 curve is not unity, as it must always be at sufficiently low E/p_0 where the energy distribution of the electrons becomes Maxwellian. A more refined calculation would take molecular motion into account and would probably have to be done numerically. The additional effort involved, however, does not seem to be warranted at this time.

It is interesting to note that the value $\alpha=20$ is about twice as large as that found by Crompton and Sutton[1] and is about four times as large as that found by Anderson and Goldstein.[19] This result can be considered as additional evidence in support of the theory of Gerjuoy and Stein[20] regarding the excitation of rotational levels at energies below the vibrational threshold of 0.29 ev. In fact, $\alpha=20$ compares favorably with the average value of 25 obtained from the theoretical λ curve of Gerjuoy and Stein over the corresponding energy range.

DISCUSSION

Quantities determined from measurements on electron swarms have values which of necessity represent an effective averaging over the existing energy distribution. To this extent their energy resolution is therefore limited and not easily defined. In spite of this limitation, electron-swarm measurements provide valuable results and indeed, the only cross-section data available below about 1 ev. The theoretical data at low energy are meager and more work in this direction would be of great value. In particular, the importance of polarization and exchange needs to be assessed. Morse and Allis have already established the huge effect of electron exchange in helium. It is also well known, for example, that extrapolation to zero energy can be accomplished more easily and reliably from the phase-shift data than from experimental cross-section data.

ACKNOWLEDGMENT

The author wishes to thank Dr. Robert L. Platzman for many interesting and helpful discussions.

[18] Phelps, Fundingsland, and Brown, Phys. Rev. **84**, 559 (1951).

[19] J. M. Anderson and L. Goldstein, Phys. Rev. **102**, 388 (1956).

[20] E. Gerjuoy and S. Stein, Phys. Rev. **97**, 1671 (1955).