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TECHNICAL REPORT 20

TRANSPORT COEFFICIENTS AND CROSS SECTIONS IN ARGON AND HYDROGEN-ARGON MIXTURES

A. G. ENGELHARDT
A. V. PHELPS

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Principal Investigators: A. V. Phelps
G. J. Schulz

Physics Department
Westinghouse Research Laboratories
Pittsburgh 35, Pennsylvania

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TRANSPORT COEFFICIENTS AND CROSS SECTIONS IN ARGON AND HYDROGEN-ARGON MIXTURES*

A. G. Engelhardt and A. V. Phelps
Westinghouse Research Laboratories, Pittsburgh 35, Pennsylvania

ABSTRACT

~~Using a method described previously~~ the distribution function of electron energies for electrons in argon and mixtures of hydrogen and argon WAS ~~has been~~ obtained by a numerical solution of the Boltzmann equation which takes into account elastic and inelastic collisions. By taking appropriate averages over the distribution function, the electron drift velocity w, magnetic drift velocity w_M and characteristic energy epsilon ϵ_K are computed. A comparison of calculated and experimental values of these transport coefficients enables us, in the case of pure argon, to extend the previous work of Frost and Phelps and derive the momentum transfer cross section from 0.7 to 25.0 eV. The momentum transfer and inelastic collision cross sections derived in this paper for argon and previously for hydrogen give rise to transport coefficients in mixtures of these two gases which are consistent with most of the available experimental data.

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I. INTRODUCTION

Previous determinations[1-4] of momentum transfer and inelastic collision cross sections for electrons in inert and molecular gases provide motivation for calculations which verify the validity of these cross sections. The method which is described in this paper provides such a verification in terms of a comparison of experimental and theoretical values of transport coefficients for binary mixtures of hydrogen and argon. Our choice of argon as the inert component and hydrogen as the molecular one was suggested by the observation of a number of investigators[5-7] of a large change of electron drift velocity produced by small concentrations of H_2 in Ar, and by the availability of previous determinations[3] of the momentum transfer cross sections in Ar and of elastic and inelastic cross sections in hydrogen.[1,2] In order to be able to perform calculations for regions of higher electron energy, the determination by Frost and Phelps[3] of the momentum transfer cross section Q_m in argon for electron energies less than 0.7 eV was extended to an energy of approximately 25 eV. In these calculations we have used the argon inelastic collision cross section determined experimentally by Maier-Leibnitz.[8]

The procedure adopted is essentially the same as that discussed by Frost and Phelps[1] (hereafter called I) and Engelhardt and Phelps[2] (hereafter called II); therefore, we shall give only a brief outline here. The Boltzmann equation (i.e., Eq. (II.2))[9] is solved numerically for the distribution function f of electron energies taking into account both momentum transfer and inelastic collisions. In I and II, cross sections were obtained by comparing experimental and theoretical values of various combinations of transport coefficients--the diffusion coefficient D, the mobility μ, and the Townsend primary ionization coefficient α_i; all of these transport coefficients were obtained by taking the appropriate average over the distribution function (see Eqs. (II.6) to (II.12) inclusive). We have used this technique here with slight modifications in deducing cross sections for pure argon. In the case of mixtures of hydrogen and argon, the previously derived cross sections were checked by comparing experimental and calculated values of the drift velocity $w = \mu E$ and, in one case, the characteristic energy[2] $\epsilon_K = eD/\mu$. In this paper we also compare our calculated results with measurements[7,10] of the "magnetic" drift velocity w_M for the configuration of crossed electric and small magnetic fields. Following Townsend[10] we define w_M by

$$w_M = \frac{E}{B} \cdot \frac{w_\perp}{w_T} \qquad (1)$$

Here $w_\perp$ is the drift velocity perpendicular to both electric and magnetic fields, and w_T is the drift velocity transverse to the magnetic field but parallel to the electric field; the ratio $w_\perp / w_T$ is the tangent of the angle through which the electrons are deflected.[10] For an energy independent frequency of momentum transfer collisions, viz. $\nu_c = (2\epsilon/m)^{1/2} Q_m = \text{const.}$ where ϵ is the electron energy and m its mass, it can be shown that $w_M = w$. We shall see that in the case of pure argon the condition ν_c = const. is not fulfilled, and that the deviations from this relation are as large as a factor of four.

II. MOMENTUM TRANSFER AND INELASTIC CROSS SECTION FOR ARGON

In this section we consider the determination of the argon momentum transfer cross section in the energy range from 0.7 to approximately 25 eV; the cross section for energies less than 0.7 eV is discussed in reference 3. This cross saction is shown in Fig. 1. The dashed momentum transfer cross section, Q_{mT}, represents the original result obtained by Frost and Phelps[11] by adjusting Q_m to fit only the experimental drift velocity data.[12] Above an energy of 15 aV this cross section is identical to that reported previously by Barbiere.[13] The solid curve is our final momentum cross section, Q_{mA}, for argon. The portion of the solid curve below 0.7 eV is the result of later calculations by Frost and Phelps[3] in which the momentum transfer cross section was modified so as to give a better fit to the ϵ_K data of Warren and Parker[14] at 77^oK. The final Q_{mA} curve has been compared by Frost and Phelps with the results of electron beam experiments and recent theory, and consequently the discussion will not be repeated here. Here we will prasent results for both Q_{mT} and Q_{mA} in order to show the effect of varying Q_m. Also shown in Fig. 1 is the total effective inelastic cross section[8] with a threshold at 11.5 eV. This inelastic cross section includes processas whose energy loss is greater than 11.5 eV, e.g., ionization. However, our calculations show that the values of w, w_M, and ϵ_K do not change significantly when, instead of the single inelastic process shown, we use two cross sections having the same total effective cross section, i.e., one for excitation and one for ionization.

Our procedure has been to compare experimental and theoretical values of w and ϵ_K and to make adjustments in Q_m on this basis. In argon for ϵ_K less than approximately 7.0 eV, few electrons have energies great enough to experience inelastic collisions. In this case, one has only one adjustable parameter, $Q_m(\epsilon)$, and in principle one can obtain Q_m by fitting to a plot of either w or ϵ_K vs E/N. However, as found by Frost and Phelps,[3] values of w are rather insensitive to the choice of Q_m when w varies as slowly with E/N as in argon for $10^{-19} < E/N < 2 \times 10^{-17}$ V-cm^2. Fortunately at the lower values of E/N, one can make use of ϵ_K data to obtain more accurate Q_m values. Frost and Phelps[3] were unable to obtain a unique Q_m for energies greater than 0.7 eV because of the dominating influence of the Ramsauer minimum. As is indicated below, we believe one can overcome this difficulty by using mixtures of 10% H_2 and 90% Ar. For $\epsilon_K < 7.0$ eV we have assumed the inelastic collision cross section of Maier-Leibnitz to be correct and have, therefore, made appropriate adjustments only in Q_m. We note that because of the decreasing ϵ_K at high E/N (see Figs. 2 and 5) plots of ν_m/N and ν_u/N such as used in references 1 and 2 are not single valued and are of little use in our analysis of pure Ar at high ϵ_K.

Shown in Fig. 2 is a plot of w, w_M and ϵ_K for 77°K. The smooth solid curves represent our calculations using Q_{mA}, and the smooth dashed curves represent our calculations using Q_{mT}; the points represent the various experimental results of Warren and Parker,[14] Townsend and Bailey,[7,10] Pack and Phelps,[12] Erret,[6] Bowe,[15] Nielsen,[16] Riemann,[17] Herreng,[18] and Caren.[19] The theoretical results of Barbiere[13] and Heylen and Lewis[20] are also shown as smooth curves. The ϵ_K data of Warren and Parker from 0.06 to 1.2 eV was taken at liquid argon temperature, 87°K. Calculations[3] show that from ϵ_K = 0.06 to 0.2 eV these experimental points should be shifted downward about 10% in order to compare with the 77° calculations. In the case of the w data both Q_{mA} and Q_{mT} give reasonably good agreement with experiment up to an E/N of 10^{-16} V-cm^2. Above E/N = 10^{-16} V-cm^2 the curve showing our final values of w falls midway between the results of Erret and those of Riemann, Herreng, and Caren; in this region the w values found using Q_{mT} favor Erret's results.

That the final result Q_{mA} represents an improvement over Q_{mT} can be concluded from the ϵ_K plot where it is seen that Q_{mA} yields very acceptable agreement with the data of Warren and Parker. On the other hand by using Q_{mT}, discrepancies as large as 20% are obtained. We have been unable to obtain agreement with the ϵ_K values of Townsend and Bailey for E/N near 10^{-17} V-cm^2 using reasonable Q_m curves, and so have chosen not to force Q_m to fit this data. A similar unresolved discrepancy has been found[2] to exist over a portion of the ϵ_K data in H_2. In the case of w_M, both Q_{mA} and Q_{mT} give comparable agreement with the results of Townsend and Bailey. Both cross sections yield w_M values which change rapidly with E/N in the vicinity of E/N = 10^{-16} V-cm^2 where inelastic collisions result in an increased relative electron density in the Ramsauer minimum. The large difference between w and w_M in this region is an example of the deviation of the "magnetic deflection" factor from unity[1] which is possible when $\epsilon^{1/2}\, Q_m(\epsilon)$ varies rapidly with electron energy.

Finally we note that in the highest regime of E/N studied, the ϵ_K values of Heylen and Lewis[20] are approximately 50% greater than ours. In performing their calculations these authors have used a piecewise analytical approximation to the momentum transfer cross section which in principle should not give rise to a distribution function significantly different from our own for $E/N > 10^{-17}$ V-cm^2. Essentially the same inelastic cross sections[8] are used in both analysis. However, a perusal of Fig. 3 indicates that there are rather marked differences between the distribution function of Heylen and Lewis and our own. Since their distribution function has more particles at higher energies, it is to be expected that their value of ϵ_K will be substantially greater than our own. Moreover, we have compared our distribution function obtained with $E/N = 3.0 \times 10^{-16}$ V-cm^2 with that of Golant[21] for $E/N = 2.94 \times 10^{16}$ V-cm^2 to discover that the two are almost identical and, incidentally, not very different from our calculated $F(\epsilon)$ shown in Fig. 3. The agreement with Golant's result should not be too surprising since the elastic and inelastic cross sections used by Golant are quite similar to our own. Consequently we conclude that the method used by Heylen and Lewis to calculate the electron energy distribution function can lead to large errors in the transport coefficients as apparently is the case for Ar.

III. MIXTURES OF HYDROGEN AND ARGON

A. Comparison with Experiment

The calculation of transport coefficients for mixtures of hydrogen and argon stemmed from a desire to check the validity of previously derived momentum transfer and inelastic cross sections for both H_2 and Ar. Since argon has no inelastic processes below 11.5 eV, it is possible for sufficiently low values of ϵ_K to scrutinize the inelastic processes of rotation and vibrational excitation in H_2. The enhanced sensitivity of the transport coefficients to inelastic processes with thresholds between about 0.5 and 10 eV is due to the unusual shape of Q_{mA} which rises rapidly for $\epsilon > 0.3$ eV. Consequently the $f(\epsilon)$ in Ar drops rapidly for $\epsilon > \epsilon_K$ and leads to a narrower energy spread than in most other gases. Such a behavior is shown in Fig. 3. As a result the effects on transport coefficients of various inelastic processes are more clearly separated. Furthermore, since the onset of vibrational excitation occurs at 0.516 eV, small percentages of H_2 in Ar will result in an even larger relative number of electrons trapped in the vicinity of the Ramsauer minimum for appropriate values of E/N. In this manner we are able to investigate the validity of the rising part of the vibrational excitation cross section.

Figs. 4 and 5 exhibit a comparison of experimental and theoretical values of w, w_M, and ϵ_K for mixtures of 1%, 1.5%, 4%, and 10% H_2 in A. The H_2 momentum transfer and inelastic cross sections are the same as those shown in Fig. 1 and 7 of II. The argon momentum transfer and inelastic cross sections used are those shown in Fig. 1. As indicated above, the higher energy portion of Q_{mA} was obtained by adjusting Q_{mA} to give a fit between the calculated and measured values of w in a mixture of 10% H_2 and 90% A. The drift velocity in this mixture is sensitive to the value of Q_{mA} at the higher energies because for this case in the vicinity of the Ramsauer minimum $0.1\ Q_{mH} > .9\ Q_{mA}$, where Q_{mH} is the momentum transfer cross sections of H_2. As a result considerably fewer electrons are trapped in the Ramsauer minimum and it is easier to investigate the variations of Q_{mA} for energies greater than about 1 eV than for the case of pure Ar alone. The comparison of experimental and calculated values of w for 10% H_2 in Ar shown in Fig. 4 indicates that we have been able to obtain satisfactory agreement with the experimental data of Erret. It should be of value to repeat this evaluation of Q_{mA} using He-Ar mixtures, since the complications of inelastic collisions would not be present.

The comparison in Figs. 4 and 5 of the calculated and experimental values of w, w_M, and ϵ_K for various H_2-Ar mixtures shows generally good agreement and indicates that our assumed cross sections are reasonably consistent with experiments. However, there are several regions of some discrepancy which need to be considered. For example, we note in Fig. 4 that for 1% H_2 in A there is a discrepancy of as much as 15% with the values of w measured by Erret for E/N values above about 6×10^{-17} V-cm^2. A comparison of the w and ϵ_K curves for this mixture and for pure Ar shows that this is the same discrepancy in w values that is present in Erret's data for pure Ar for E/N above 3×10^{-17} V-cm^2. However, in pure Ar the discrepancy with respect to other recent data is much smaller or in the opposite direction. This suggests the possibility of error in Erret's data at high E/N. Since this error does not appear to be present in Erret's pure H_2 or pure N_2 data, we have assumed that the data for 10% H_2-90% Ar used in the evaluation of Q_{mA} is correct.

A second region in which we have consistently been unable to obtain as good agreement between calculated and experimental drift velocities as we might expect occurs in pure Ar and in H_2-Ar mixtures for E/N such that

$\epsilon_K \sim 0.8$ to 1.0 eV. The w data of Figs. 4 and 5 show that the maximum discrepancy in this range of ϵ_K varies from about 10% in pure Ar and in 10% H_2 in Ar to about 20% in 1.5% H_2 in Ar. This same discrepancy[22] is seen in the w_M data of Fig. 4. The presence of this discrepancy in the pure Ar data for ϵ_K near 1 eV suggests that we have not found the optimum Q_{mA} curve for energies below about 1 eV. We have not pursued this question further because of the relative smallness of the discrepancy, the lack of recent ϵ_K values in mixtures, and the repeated inconsistencies between what we consider to be reasonable cross sections and the data of Townsend and Bailey. This state of affairs emphasized the desirability of accurate measurements of w or w_M, and ϵ_K for various H_2-Ar mixtures.

In the case of 4% H_2 in Ar, we note that according to Fig. 4 the cross sections used in our analysis lead to very good agreement between the calculated and experimental values of ϵ_K. This agreement is to be compared with the existence of discrepancies of as much as 20% between the calculated and experimental values of w_M. This effect could be due to the fact that w_M is more sensitive to variations in Q_m than is ϵ_K, i.e., w depends on integrals of $1/Q_m^2$ where D depends on integrals of $1/Q_m$.

It is possible to regard the results for various concentrations as a measure of the sensitivity of w, w_M, and ϵ_K to changes in the inelastic cross sections for fixed concentration. For example, we observe that for $E/N = 10^{-17}$ V-cm^2 a decrease of 33% in the effective vibrational cross section caused by changing the mixture composition from 1.5% H_2 in Ar to 1% H_2 in Ar results in a 20% increase in ϵ_K and a 13% decrease in w.

B. Advantages of Mixture Technique

Here we would like to make use of the results of the preceding calculations to illustrate graphically the manner in which argon serves to improve the separation in energy of various inelastic collision processes. For this purpose it is convenient to employ a combination of transport coefficients which has been shown[1,2] to be a good measure of the cross section for inelastic collisions. Thus we use the energy exchange collision frequency, as defined by the relation

$$\nu_u/N = \frac{ewE/N}{\epsilon_K - kT_g} . \qquad (3)$$

For mixtures of H_2 in Ar it is most appropriate to think in terms of a hydrogen energy exchange collision frequency, $\left[\nu_u/N\right]_{H_2\text{-}Ar}$, for a mixture of H_2 and Ar which is a function not only of ϵ_K but also of the mixture composition, and which is defined by the relation,

$$\left[\frac{\nu_u}{N}\right]_{H_2\text{-}Ar} = \frac{1}{\alpha}\left[\frac{\nu_u}{N}\right]_T - \frac{1-\alpha}{\alpha}\left[\frac{\nu_u}{N}\right]_{Ar} . \qquad (4)$$

In Eq. (4) α is the fraction of H_2 in the mixture; $\left[\nu_u/N\right]_T$ is the total energy exchange collision frequency for a mixture of H_2 in Ar, and is obtained by inserting into Eq. (3) the appropriate values as shown in Figs. 4 and 5; and $\left[\nu_u/N\right]_{Ar}$ is the energy exchange collision frequency for pure Ar at the same value of ϵ_K as $\left[\nu_u/N\right]_{H_2\text{-}Ar}$ and $\left[\nu_u/N\right]_T$. To be truly meaningful

$\left[\nu_u/N \right]_{Ar}$ should be much less than $\left[\nu_u/N \right]_T$. This condition is fulfilled for the mixtures and values of ϵ_K considered.

Shown in Fig. 6 are calculated plots of $\left[\nu_u/N \right]_{H_2-Ar}$ for H_2, for 1%, 1.5%, 4% and 10% H_2-Ar, and for pure Ar. In the case of Ar we note that $\left[\nu_u/N \right]_{Ar}$ decreases up to $\epsilon_K \simeq .10$ eV because of the Ramsauer minimum. At the point where excitation first begins to assume importance, i.e., $\epsilon_K \simeq 7.4$ eV, $\left[\nu_u/N \right]_{Ar}$ starts to increase extremely rapidly, and the curve is even multi-valued for a short interval since ϵ_K decreases slightly in the region $6.0 \times 10^{-17} < E/N < 1.0 \times 10^{-16}$ V-cm^2.

For mixtures the curves of $\left[\nu_u/N \right]_{H_2-Ar}$ indicate clearly the effectiveness of Ar in separating some of the inelastic processes. In particular we wish to emphasize the improvement in the degree to which the effects of vibrational excitation are separated from electronic excitation.[23] As indicated in I and II and shown in Fig. 6, the major contribution of ν_u/N for $\epsilon_K < 0.3$ eV is due to rotational excitation. In pure H_2 the effects of vibrational excitation are seen for ϵ_K from 0.3 to about 1.5 eV where electronic excitation begins to be important. Fig. 6 shows that especially in the mixtures with low concentrations of H_2, the vibrational excitation dominates up to an ϵ_K of 5 eV. Therefore, if more and better experimental determinations of w and ϵ_K for mixtures were available, it should be possible to make an even more accurate analysis of vibrational excitation than has been previously possible.[1,2]

IV. SUMMARY AND CONCLUSIONS

In the preceding sections we have discussed the determination of the argon momentum transfer cross section from 0.7 to approximately 25 eV. This cross section has been used in conjunction with previous results for hydrogen to calculate transport coefficients for various H_2-Ar mixtures. For pure argon satisfactory agreement has been obtained between calculated and experimental values of the drift velocity w, the "magnetic" drift velocity w_M, and the characteristic energy ϵ_K. On the other hand for mixtures of H_2 and Ar the picture is less satisfactory. In some cases the discrepancy appears to be due to experimental error, whereas in others it may be due to a failure to determine the momentum transfer cross section for argon with sufficient accuracy.

Unfortunately the analysis has been hampered by insufficient data. For example, in the case of mixtures of H_2 in Ar, only for 4% H_2 in Ar is there ϵ_K data. Thus, a more complete and satisfactory analysis is predicated upon obtaining additional experimental results for pure Ar and mixtures of H_2 and Ar. As a further inducement for the procurement of such data, it has been demonstrated that improved separation is attained between processes such as vibration and electronic excitation in H_2.

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FOOTNOTES

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22. However, this correlation of discrepancies in w_M on the basis of ϵ_K values is not unique. Figures 2 and 4 show that for both pure Ar and 4% H_2 in Ar the calculated values of w_M are higher than the experimental results near $E/N = 10^{-17}$ $V\text{-}cm^2$.

23. Although the data of Fig. 6 also seems to indicate that analysis of $\left[\nu_u/N\right]_{H_2\text{-}Ar}$ should yield more accurate cross sections for rotational excitation, the increased values of the energy exchange collision frequency for $\epsilon_K < 0.15$ eV are due in part to the rapidly decreasing Q_{mA} and the resultant broadening of $f(\epsilon)$. This is in contrast to the region where vibrational excitation is dominant, and where the increasing Q_{mA} leads to a narrowing of $f(\epsilon)$.

Fig. 1 Momentum transfer and inelastic collision cross sections for electrons in argon as a function of electron energy in eV. Q_{mA} (solid line) is our final value of the momentum transfer cross section and Q_{mT} (dashed line) is a trial value.

Fig. 2 Drift velocity w, magnetic drift velocity w_M, and characteristic energy ϵ_K for electrons in argon as a function of E/N. Below $\epsilon_K = 0.5$ eV the experimental data shown is for gas temperatures T_g of 77^oK and 87^oK as discussed in the text. Above $\epsilon_K = 0.5$ eV and for $T_g \leq 300^oK$ the values of w, w_M, and ϵ_K are insensitive to T_g. The solid and dashed curves represent our calculated results using Q_{mA} and Q_{mT} respectively; the points represent the various experimental results.

Fig. 3 Energy probability function $F(\epsilon)$ for electrons in argon as a function of energy ϵ in eV. Since $F(\epsilon) = \epsilon^{1/2} f(\epsilon)$, $F(\epsilon)\, d\epsilon$ is the probability of an electron having an energy between ϵ and $\epsilon + d\epsilon$. The results of our calculations are shown as a solid line for $E/N = 4.0 \times 10^{-16}$ V-cm^2 and $\epsilon_K = 9.60$ eV. The $F(\epsilon)$ of Heylen and Lewis is plotted as a dotted line for $E/N = 4.24$ V-cm^2 and $\epsilon_K = 13.4$ eV. The difference of approximately 5% in the two values of E/N considered by itself leads to only very minor changes in $F(\epsilon)$. For illustrative purposes we display for $\epsilon_K = 9.60$ eV the Maxwellian $F(\epsilon)$ given by $F(\epsilon) = 2\ (\epsilon/\pi\epsilon_K^3)^{1/2} \exp(-\epsilon/\epsilon_K)$, where ϵ and ϵ_K are in eV.

Fig. 4 Drift velocity w, magnetic drift velocity w_M, and characteristic energy ϵ_K as a function of E/N for electrons in mixtures of 1%, 4%, and 10% H_2 in Ar. The solid, dashed, and dotted curves represent our calculated results for 1%, 4%, and 10% H_2 in Ar respectively. The various experimental data are shown as points.

Fig. 5 Drift velocity w and characteristic energy ϵ_K for electrons in a mixture of 1.5% H_2 in Ar as a function E/N. The solid lines represent our calculations for $T_g = 77^oK$ and the dashed ones for $T_g = 300^oK$. The experimental results of Pack and Phelps are shown as points.

Fig. 6 Calculated energy exchange frequency ν_u/N for Ar, H_2, and 1%, 1.5%, 4%, and 10% H_2 in Ar at $T_g = 77^oK$ as a function of ϵ_K in eV. In the case of the mixtures ν_u/N is obtained using Eq. (4). For Ar, ν_u/N is shown increased by a factor of 100.

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Momentum transfer and inelastic collision cross sections of electrons in argon

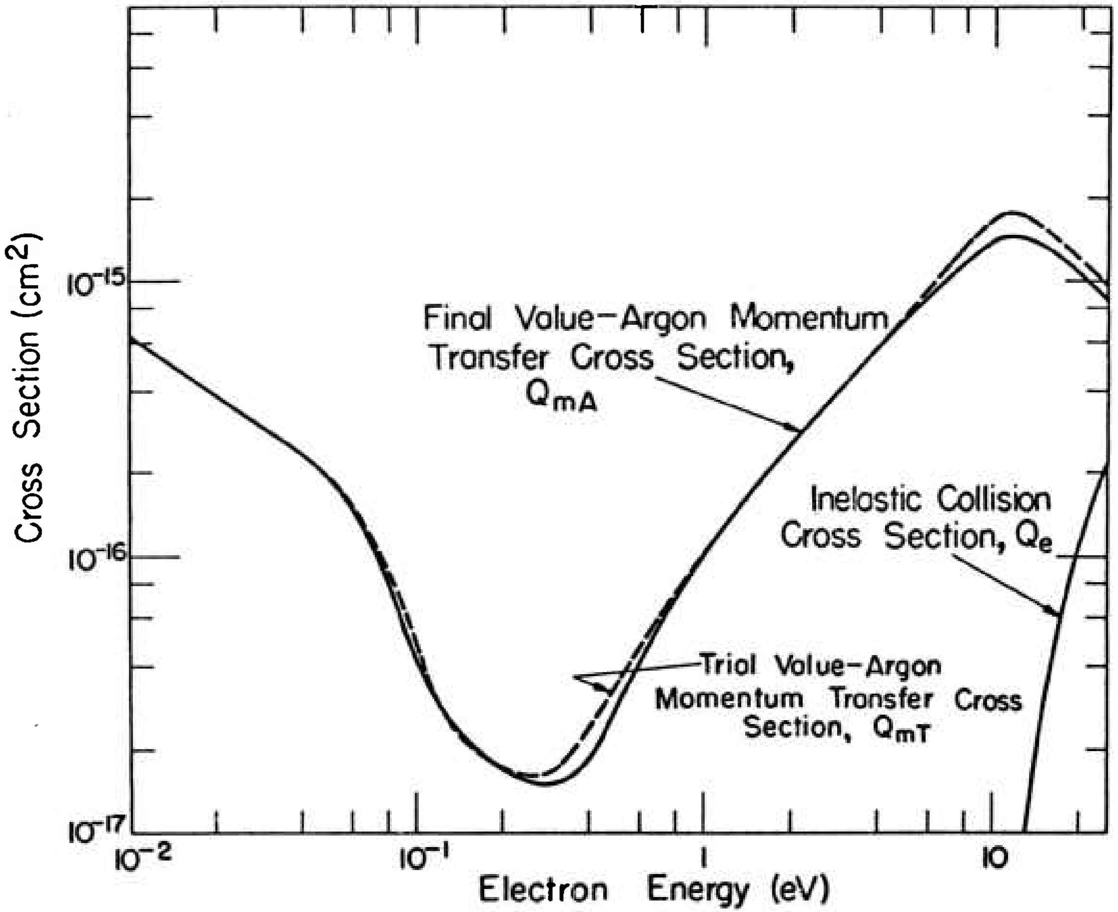

Figure 1

[For complete figure captions, see page 16]

Drift velocity w, magnetic drift velocity w_M and characteristic energy ϵ_K for argon

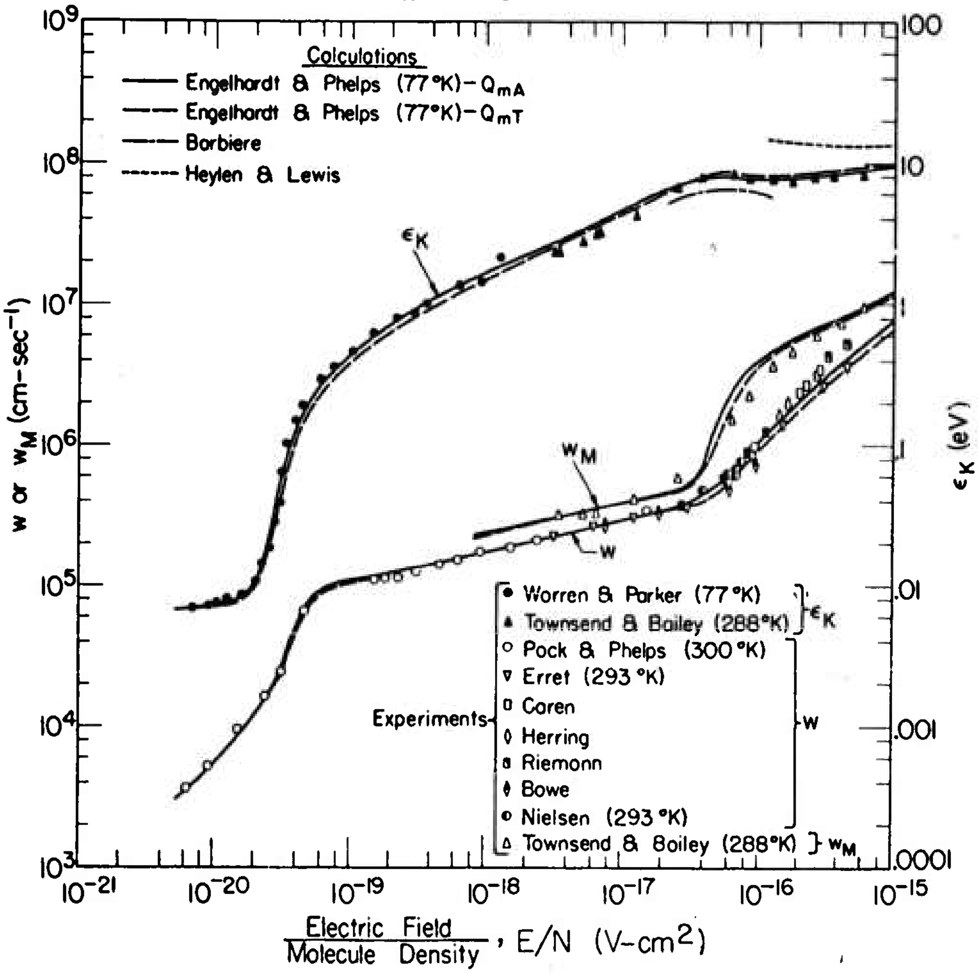

Figure 2

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Energy probability function $F(\epsilon)$ for electrons in argon

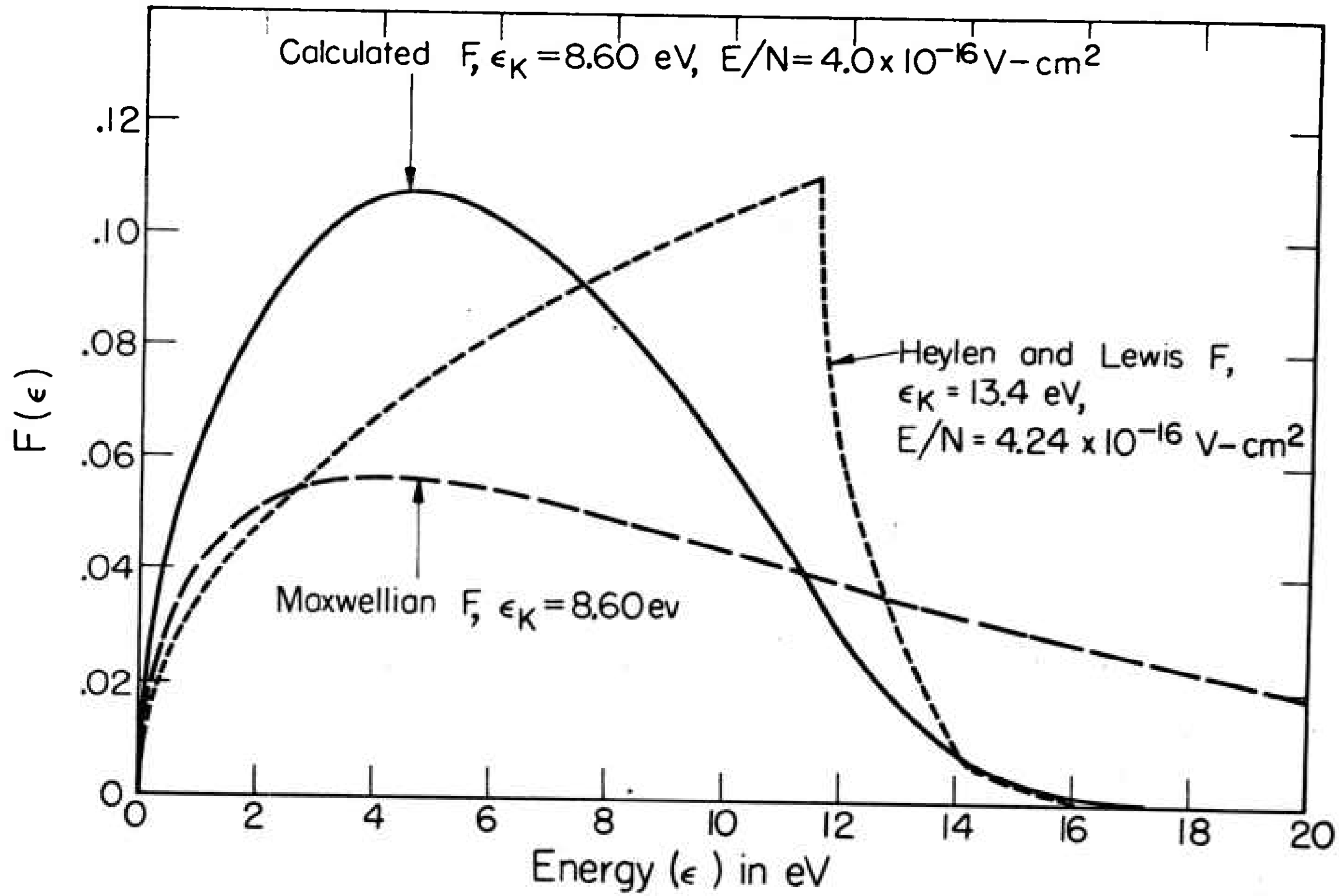

Figure 3

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Drift velocity w, magnetic drift velocity w_M and characteristic energy ϵ_K for 1%, 4%, & 10% H_2 in Ar

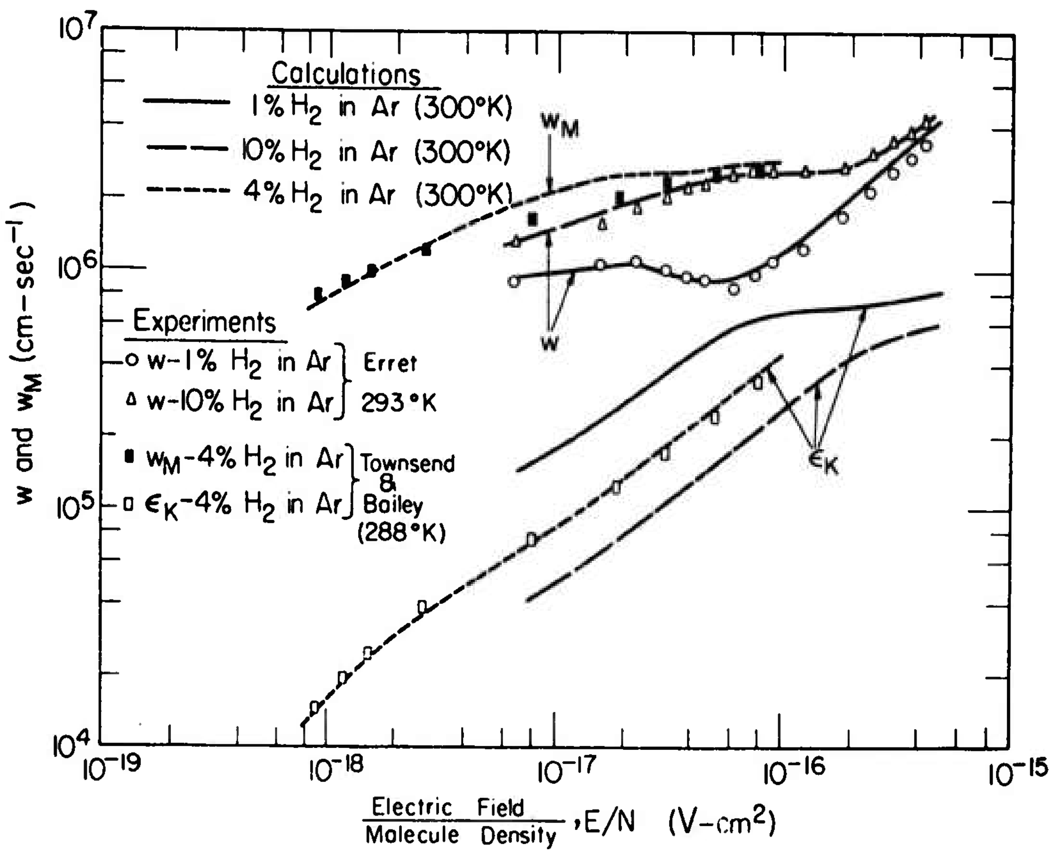

Figure 4

CURVE 566761

Drift velocity w and characteristic energy ϵ_K for 1.5% H_2 in Ar

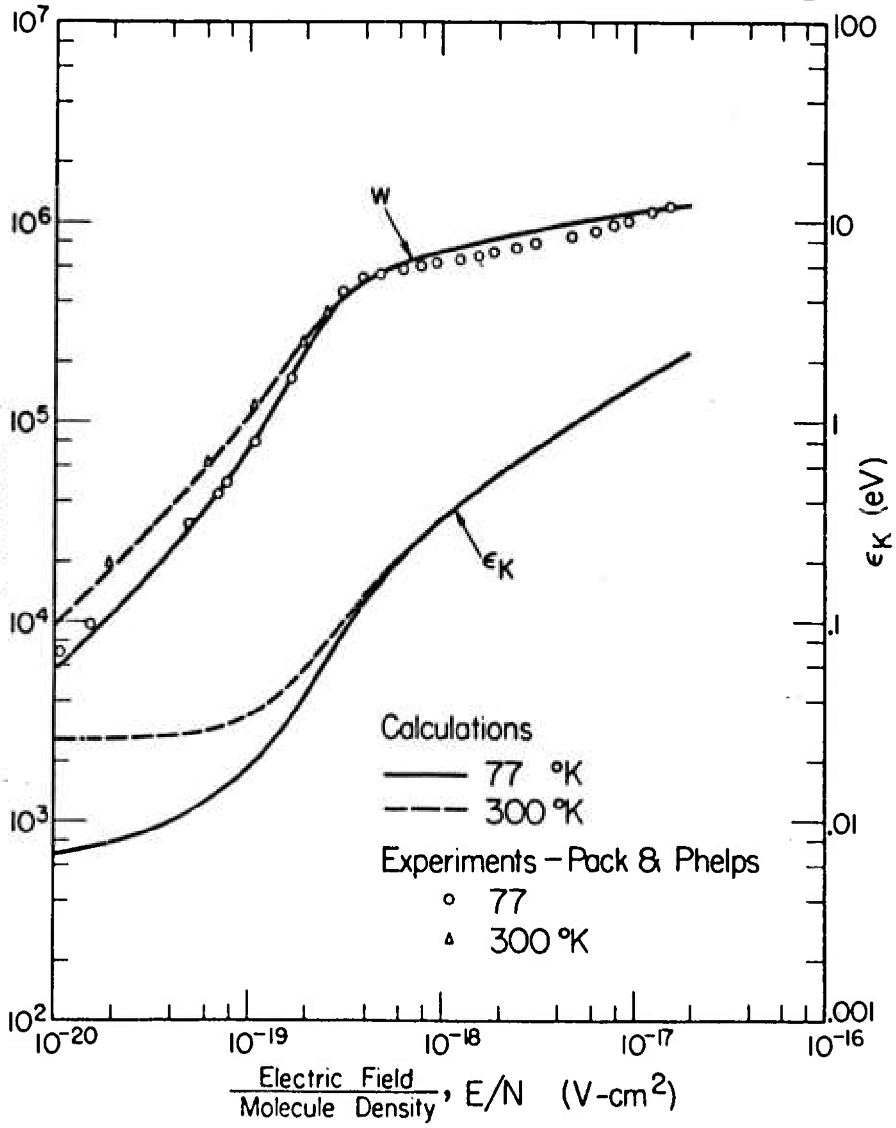

Figure 5

CURVE 566759

Energy exchange frequency (ν_u/N) for Ar, H_2, and 1% 1.5%, 4% and 10% H_2 in Ar at T = 77°K

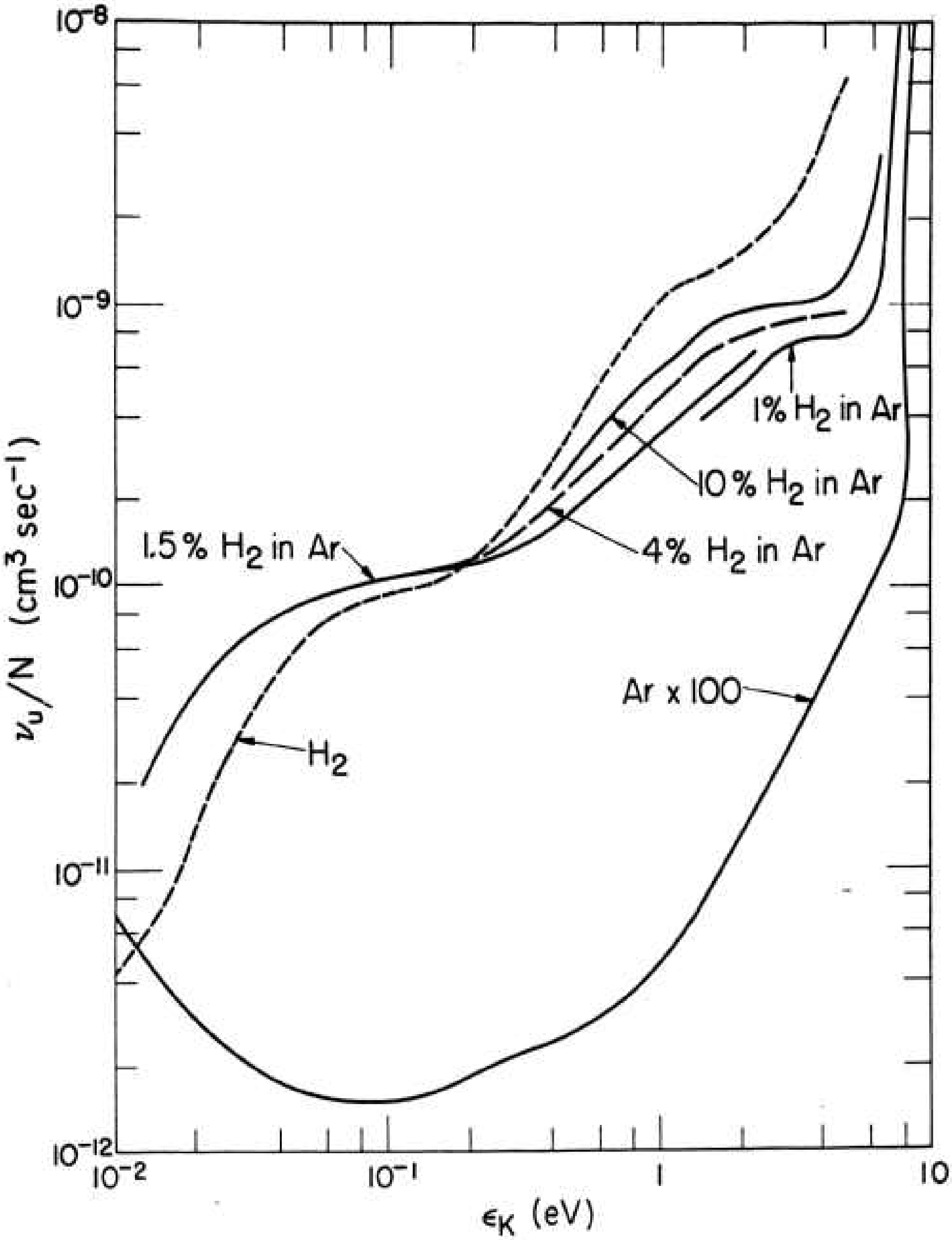

Figure 6

UNCLASSIFIED

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