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The Momentum Transfer Cross Section for Electrons in Argon in the Energy Range 0–4 eV

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Abstract

The momentum transfer cross section for electron–argon collisions in the range 0–4 eV has been derived from an analysis of recent measurements of D_T/μ as a function of E/N at 294 K (Milloy and Crompton 1977*a*) and W as a function of E/N at 90 and 293 K (Robertson 1977). Modified effective range theory was used in the fitting procedure at low energies. An investigation of the range of validity of this theory indicated that the scattering length and effective range were uniquely determined and hence the cross section could be accurately extrapolated to zero energy. It is concluded that for $\varepsilon \leqslant 0 \cdot 1$ eV the error in the cross section is less than $\pm 6\%$ and in the range $0 \cdot 4 \leqslant \varepsilon\,(\text{eV}) \leqslant 4 \cdot 0$ the error is less than $\pm 8\%$. In the range $0 \cdot 1 < \varepsilon\,(\text{eV}) < 0 \cdot 4$ the presence of the minimum makes it difficult to determine the errors in the cross section but it is estimated that they are less than -20%, $+12\%$. It is demonstrated that no other reported cross sections are compatible with the experimental results used in the present derivation.

1. Introduction

Since the pioneering work of Ramsauer (1921) there have been three experimental techniques used to determine either the total or momentum transfer cross sections for electrons in argon at energies near the minimum. The first investigation (Frost and Phelps 1964) was based on an analysis of transport coefficient data. Two years later Golden and Bandel (1966) used electron beam techniques to measure the total cross section $q_s(\varepsilon)$ in the range $0 \cdot 1$–$21 \cdot 6$ eV. These data were subsequently analysed by Golden (1966) with modified effective range theory (MERT) and the parameters derived in this analysis were used to predict the momentum transfer cross section $q_m(\varepsilon)$. The very large discrepancy that exists between Golden's results and those of Frost and Phelps (see Fig. 4 in Section 5 below) prompted the present work. More recently McPherson *et al.* (1976) have used a microwave transient response technique to determine the (normalized) collision frequency for momentum transfer in the range $0 \cdot 08$–4 eV (their momentum transfer cross section is also included in Fig. 4).

The present work is based on the swarm technique described in detail by Huxley and Crompton (1974). The advantages of this approach stem basically from the use of relatively high gas pressures (typically 1 atm, or ~ 100 kPa) in the experiments to measure the transport coefficients. This has the direct advantage that the gas number density can be measured with an absolute error of $<0 \cdot 1\%$. In addition the use of high gas pressures results in the use of high electric field strengths and the reduction of errors due to contact potential differences. In this way two of the main sources of error in a single collision type of experiment are overcome. The main disadvantage of the swarm approach for the case of argon arises from the structure of the cross section in the region of the minimum which introduces problems of uniqueness in the analysis. However, this can be overcome to a large extent if accurate experimental transport coefficient data are available.

The transport coefficients used here were room temperature measurements of the ratio of the lateral diffusion coefficient to mobility D_T/μ (Milloy and Crompton 1977, present issue p. 51) and drift velocity measurements at 90 and 293 K (Robertson 1977, present issue p. 39). As the experimental details have been considered elsewhere they need not be repeated here. However, it is necessary to briefly discuss electron transport theory. This discussion is given in Section 2 together with a summary of MERT, which was used to predict the form of the cross section at low energies. To evaluate the validity of the present and previous work with MERT, it has been necessary to discuss the energy range of validity of this theory in some detail (Sections 2 and 4). The procedure used to derive the cross section is described in Section 3 and the resulting cross section is discussed in Section 4.

2. Theoretical Summary

(*a*) *Transport Theory*

Electron motion in a gas in the presence of an electrostatic field and electron density gradients has been the subject of several papers in recent years (e.g. Skullerud 1974, and references therein). It has been shown that, irrespective of the magnitude of the density gradients and when inelastic processes can be neglected, the space and time average of the isotropic part of the distribution function for electron energies may be written (see e.g. Huxley and Crompton 1974)

$$f(\varepsilon) = A\exp\left(-\int_0^\varepsilon \left\{\left(ME^2e^2/6mN^2\varepsilon\, q_m^2(\varepsilon)\right)+kT\right\}^{-1} \mathrm{d}\varepsilon\right), \tag{1}$$

where $\varepsilon^{\frac{1}{2}} f(\varepsilon)\,\mathrm{d}\varepsilon$ is the probability that an electron has energy in the range ε to $\varepsilon+\mathrm{d}\varepsilon$, M and m are the atom and electron masses, $q_m(\varepsilon)$ is the energy-dependent momentum transfer cross section, k is the Boltzmann constant, T is the gas temperature, e is the electron charge, E is the electric field strength and N is the gas number density. The constant A is obtained from the normalizing relationship

$$\int_0^\infty \varepsilon^{\frac{1}{2}} f(\varepsilon)\,\mathrm{d}\varepsilon = 1\,.$$

Written in terms of $f(\varepsilon)$ the formulae for the drift velocity W and the lateral and longitudinal diffusion coefficients D_T and D_L become

$$W = -\frac{eE}{3N}\left(\frac{2}{m}\right)^{\frac{1}{2}} \int_0^\infty \frac{\varepsilon}{q_m(\varepsilon)}\frac{\mathrm{d}f}{\mathrm{d}\varepsilon}\,\mathrm{d}\varepsilon, \tag{2}$$

$$ND_T = \frac{1}{3}\left(\frac{2}{m}\right)^{\frac{1}{2}} \int_0^\infty \frac{\varepsilon f(\varepsilon)}{q_m(\varepsilon)}\,\mathrm{d}\varepsilon \tag{3}$$

and

$$ND_L = ND_T + \left(\frac{2}{m}\right)^{\frac{1}{2}}\frac{eE}{3N}\int_0^\infty \frac{\varepsilon}{q_m(\varepsilon)}\frac{\mathrm{d}F_1}{\mathrm{d}\varepsilon}\,\mathrm{d}\varepsilon + W\int_0^\infty \varepsilon^{\frac{1}{2}} F_1\,\mathrm{d}\varepsilon, \tag{4}$$

where

$$F_1(\varepsilon) = -\frac{f(\varepsilon)}{eE/N}\int_0^\varepsilon \frac{q_m(x)}{x\{1+kT\,\beta(x)\}f(x)}\left(\frac{x\,f(x)}{q_m(x)}+C(x)\right)\mathrm{d}x,$$

$$C(x) = \int_0^x \frac{y}{q_m(y)}\frac{\mathrm{d}f(y)}{\mathrm{d}y}\mathrm{d}y + \frac{3W(\frac{1}{2}m)^{\frac{1}{2}}}{eE/N}\int_0^x y^{\frac{1}{2}}f(y)\,\mathrm{d}y,$$

$$\beta = (6m/M)(N\,q_m(\varepsilon)/eE)^2\,\varepsilon.$$

The diffusion coefficient to mobility ratio $D_T/\mu = D_T/(W/E)$ can then be written

$$D_T/\mu = -e^{-1}\int_0^\infty \frac{\varepsilon f(\varepsilon)}{q_m(\varepsilon)}\mathrm{d}\varepsilon \Big/ \int_0^\infty \frac{\varepsilon}{q_m(\varepsilon)}\frac{\mathrm{d}f}{\mathrm{d}\varepsilon}\mathrm{d}\varepsilon. \tag{5}$$

In the following paper (Milloy and Watts 1977, p. 73) Monte Carlo techniques have been used to study the validity of the expression for the drift velocity and lateral diffusion coefficient under the conditions used in this work.

(b) Modified Effective Range Theory (MERT)

O'Malley *et al.* (1962) have derived expressions for the phase shifts of the partial waves of a scattering system comprising a charged particle plus a neutral polarizable particle. If the adjustable parameters in the expressions for the phase shifts can be determined by fitting the theoretical expressions to experimental data of suitable accuracy then MERT can be used to extrapolate data into the low and experimentally inaccessible energy range, to calculate another cross section or to identify possible inconsistencies in experimental data.

If it is assumed that the energy is small then the expressions for the s and p wave phase shifts can be written

$$k^{-1}\tan\eta_0 = -A - 0{\cdot}2839\,\alpha\varepsilon^{\frac{1}{2}} - 0{\cdot}0490\,A\alpha\varepsilon\ln\varepsilon + D\varepsilon + F\varepsilon^{3/2} + \dots, \tag{6a}$$

$$k^{-1}\tan\eta_1 = 0{\cdot}05679\,\alpha\varepsilon^{\frac{1}{2}}\{1-(\varepsilon/\varepsilon_1)^{\frac{1}{2}}\} + \dots. \tag{6b}$$

It is further assumed that the higher phase shifts can be accurately expressed using Born's approximation for the polarization potential, and thus

$$k^{-1}\tan\eta_l = 0{\cdot}8517\,\alpha\varepsilon^{\frac{1}{2}}/(2l+3)(2l+1)(2l-1) + \dots. \tag{6c}$$

In equations (6), α is the polarizability of the atom in units of a_0^3, k is the wave number in units of a_0^{-1} and ε is in units of eV. The quantities A, D, F and ε_1 are all adjustable parameters; the scattering length A is in units of a_0 and the effective range D is in units of a_0^3 while F is measured in a_0^4 and ε_1 is in eV.

The assumption is then made that the phase shifts are sufficiently small that $\tan\eta$ can be replaced by $\sin\eta$ or by η without introducing significant errors, thereby enabling the momentum transfer and total cross sections to be calculated from the well-known relations

$$q_m(\varepsilon) = 4\pi a_0^2\sum_{l=0}^{\infty}(l+1)\{\sin^2(\eta_{l+1}-\eta_l)\}/k^2, \tag{7a}$$

$$q_s(\varepsilon) = 4\pi a_0^2\sum_{l=0}^{\infty}(2l+1)(\sin^2\eta_l)/k^2. \tag{7b}$$

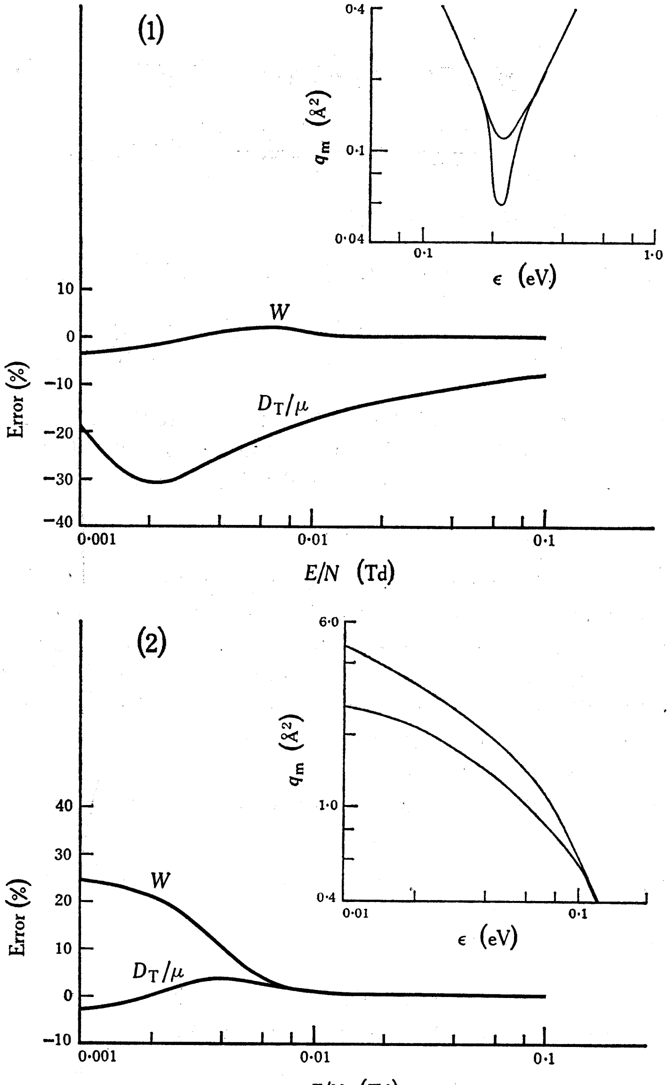

Fig. 1. Sensitivity of W and D_T/μ to changes in the cross section minimum, as demonstrated using the cross sections shown in the inset. An increase in the minimum leads to a decrease in D_T/μ.

Fig. 2. Sensitivity of W and D_T/μ to changes on the low energy side of the minimum, as demonstrated using the cross sections shown in the inset. A decrease in the cross section leads to an increase in W.

For the expansions (6) to be valid up to a given energy it is necessary to include sufficient terms in each expansion, the criterion being that the final term in each makes only a small contribution to the total phase shift. However, the inclusion of adequate terms to meet this criterion may mean that there are inadequate experimental data to determine the values of all parameters uniquely. Thus it is important to note that:

(1) once the number of adjustable parameters has been chosen the range of validity of the MERT expansions is determined, and

(2) the adequate representation of one set of experimental data by MERT does not guarantee the valid representation of another set over the same energy range unless the uniqueness of the parameters is first established.

In applying the usual form of the expansions, it is also necessary to ensure that $\tan\eta$ can be approximated by $\sin\eta$ or η up to the maximum energy within the range of validity.

3. Fitting Procedure

The minimum in the argon momentum transfer cross section can be more accurately determined by fitting to D_T/μ data than to W data of the same accuracy. This can be seen in Fig. 1 which shows the effect on W and D_T/μ of varying the depth of the minimum. The two cross sections used in the comparison are shown in the inset of Fig. 1. It can be seen that the variation in W is less than 3% while D_T/μ varies by up to 30%. Another example of the insensitivity of W to changes in the minimum was observed when an attempt was made to derive the cross section from an analysis of W data alone. It was found that the experimental data could be predicted to within the error limits on the data with a set of cross sections which varied by as much as a factor of three in the region of the minimum.

The low energy side of the minimum is more easily determined by fitting to W rather than to D_T/μ data at room temperature since the contribution to D_T/μ from the thermal motion of the gas molecules reduces the sensitivity of D_T/μ to $q_m(\varepsilon)$. Fortunately W at low E/N is very sensitive to changes in $q_m(\varepsilon)$ at low energies, as demonstrated in Fig. 2 where the two cross sections shown in the inset have been used to calculate the changes in W and D_T/μ as functions of E/N at room temperature.

The high energy side of the minimum can be determined by fitting to either W or to D_T/μ, although D_T/μ is slightly more sensitive than W to changes in $q_m(\varepsilon)$.

4. Derived Cross Section

The cross section listed in Table 1 and plotted in Fig. 4 below was derived by using MERT with four adjustable parameters in the energy range below 0·32 eV, and by empirical adjustment of the cross section at higher energies to obtain the best overall fit with the experimental data. The accuracy of the fit obtained can be seen from Fig. 3, where the differences between the experimental transport coefficients and those predicted with the derived cross section are plotted as functions of E/N. The largest deviation occurs for the D_T/μ data at low E/N, where the discrepancy of less than 3% lies just outside the estimated error bounds for the experimental data of $\pm 2\%$. The fit to the W data at 293 K has not been included in this figure. In this case the deviation was $<1\%$ at all E/N. The r.m.s. deviation between experiment and predictions is 1% for W at 90 K, 0·5% for W at 293 K and 1·5% for D_T/μ at 294 K.

(a) *Validity of MERT*

From an examination of the individual phase shifts and phase shift differences, it was found that the largest error due to the approximation $\tan\eta = \sin\eta = \eta$ was

$<0\cdot1\%$ which was therefore insignificant. The error introduced by using only four parameters up to $0\cdot32$ eV is probably more significant. At energies $\sim0\cdot3$ eV, $q_m(\varepsilon)$ is dominated by the second (i.e. $l = 1$) term in the partial wave summation and it can easily be deduced from equations (6b) and (6c) that the expansion used to calculate this term is not convergent unless the coefficients of the terms in higher

Table 1. Momentum transfer cross section for electron–argon collisions

ε (eV)	$q_m(\varepsilon)$ (Å^2)	ε (eV)	$q_m(\varepsilon)$ (Å^2)	ε (eV)	$q_m(\varepsilon)$ (Å^2)
0·014	3·88	0·130	0·348	0·320	0·188
0·017	3·56	0·140	0·284	0·325	0·206
0·020	3·28	0·150	0·233	0·400	0·317
0·025	2·89	0·170	0·161	0·500	0·504
0·030	2·57	0·180	0·135	0·650	0·792
0·035	2·29	0·190	0·115	0·800	1·05
0·040	2·05	0·200	0·101	1·00	1·37
0·050	1·662	0·210	0·092	1·20	1·66
0·060	1·357	0·220	0·086	1·50	2·05
0·070	1·114	0·230	0·085	1·70	2·33
0·080	0·916	0·240	0·087	2·00	2·70
0·090	0·754	0·250	0·091	2·50	3·43
0·100	0·621	0·260	0·098	3·00	4·20
0·110	0·511	0·280	0·120	4·00	5·70
0·120	0·420	0·300	0·151		

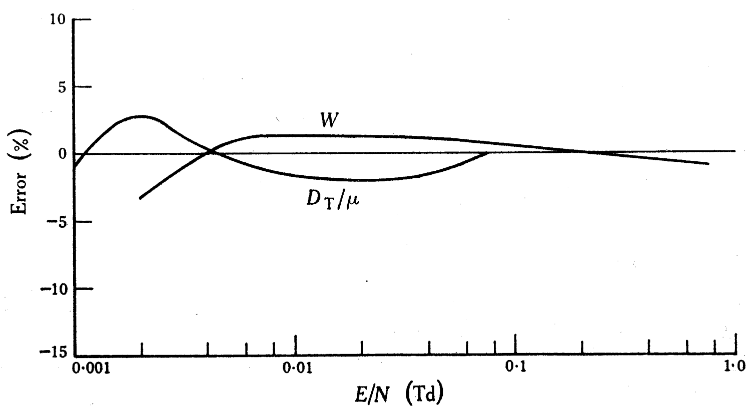

Fig. 3. Differences between the present calculated and measured values of D_T/μ at 294 K and W at 90 K, plotted as functions of E/N. The calculated W values are too low at low E/N.

powers of ε are very small. If these terms are not small ε_1 will not be uniquely determined and the parameter F cannot then be unique, since ε_1 and F together largely control the form of the cross section on the high energy side of the minimum. The parameters A and D, on the other hand, were uniquely determined. These parameters, which dominate the cross section calculations up to an energy of about $0\cdot15$ eV, could not be varied by more than 3% and 10% respectively and still give rise to a cross section compatible with the experimental data.

The values of A and D determined here are compared in Table 2 with the values obtained by Golden (1966) from a three-parameter fit to Golden and Bandel's (1966) total cross section measurements in the range $0 \cdot 1 < \varepsilon$ (eV) $\leqslant 0 \cdot 6$, and also with the values deduced by O'Malley (1963) from a three-parameter analysis of the total cross section measurements of Ramsauer and Kollath (1932). The discrepancies between the data in Table 2 must be partly due to the use of MERT at energies outside the range of validity of the expansions.

Table 2. Comparison of MERT parameters

Parameter	Present work	Golden (1966)	O'Malley (1963)
Scattering length A (a_0)	$-1 \cdot 50$	$-1 \cdot 65$	$-1 \cdot 70$
Effective range D (a_0^3)	$1 \cdot 35$	$1 \cdot 11$	$1 \cdot 23$

It is important to note that doubts about the validity of a four-parameter MERT fit in the range from $0 \cdot 15$ to $0 \cdot 32$ eV do not affect the accuracy of the derived momentum transfer cross section. These doubts do, however, affect the usefulness of MERT in predicting the total cross section at energies where the calculations are sensitive to the parameters F and ε_1.

(*b*) *Accuracy of Derived Cross Section*

If the error limits for the experimental data have been accurately assessed then it can be deduced that the derived cross section cannot be systematically in error at all energies by more than a few per cent. That is, normalization of the cross section is not a major problem. A more serious consideration is the uniqueness of the cross section in the region of the minimum. In the cases of the lighter noble gases, helium and neon, the cross section changes only slowly with energy and, unless very narrow resonances occur, the lack of uniqueness is not a significant source of error in the analysis. However, in the case of argon the width of the cross section minimum is comparable with the width of the electron energy distribution and for this reason it is more likely that the derived cross section is only one of a set which would adequately fit all the experimental data. In fact it was found that the derived cross section was not unique in the region of the minimum and an attempt was made to determine upper and lower limits for the magnitude of the cross section at the minimum. The upper limit was established in the following way. First the cross section at the minimum was increased by a few per cent. The rest of the cross section was then adjusted until a good fit was obtained. In this way it was found that if the magnitude of the minimum was greater than $0 \cdot 095$ Å^2 (i.e. increased by 12%) an adequate fit could not be obtained. The lower limit was more difficult to set since it raised the question of what is an acceptable form for the cross section at the minimum. As the cross section was made progressively smaller at the minimum, the width of the minimum had to be decreased to adequately fit the experimental data. A cross section with a minimum as low as $0 \cdot 05$ Å^2 could be found that provided an adequate fit, but its minimum was so narrow that it was regarded as physically improbable. This cross section was therefore considered to provide an overestimate of the errors in the derived cross section due to lack of uniqueness.

Uniqueness problems affect the cross section determination within the energy range from about $0 \cdot 1$ to $0 \cdot 4$ eV. At energies outside this range the error can be found by adjustment of the cross section until the predicted transport coefficients are incompatible with the measured values (Crompton *et al.* 1970). In this way it was concluded that for $\varepsilon \leqslant 0 \cdot 1$ eV the error at any energy is less than $\pm 6\%$ and for $0 \cdot 4 \leqslant \varepsilon \leqslant 4 \cdot 0$ eV the error is less than $\pm 8\%$. It is, however, necessary to stress that a systematic adjustment of the cross section of greater than 2% would not be compatible with the experimental transport data.

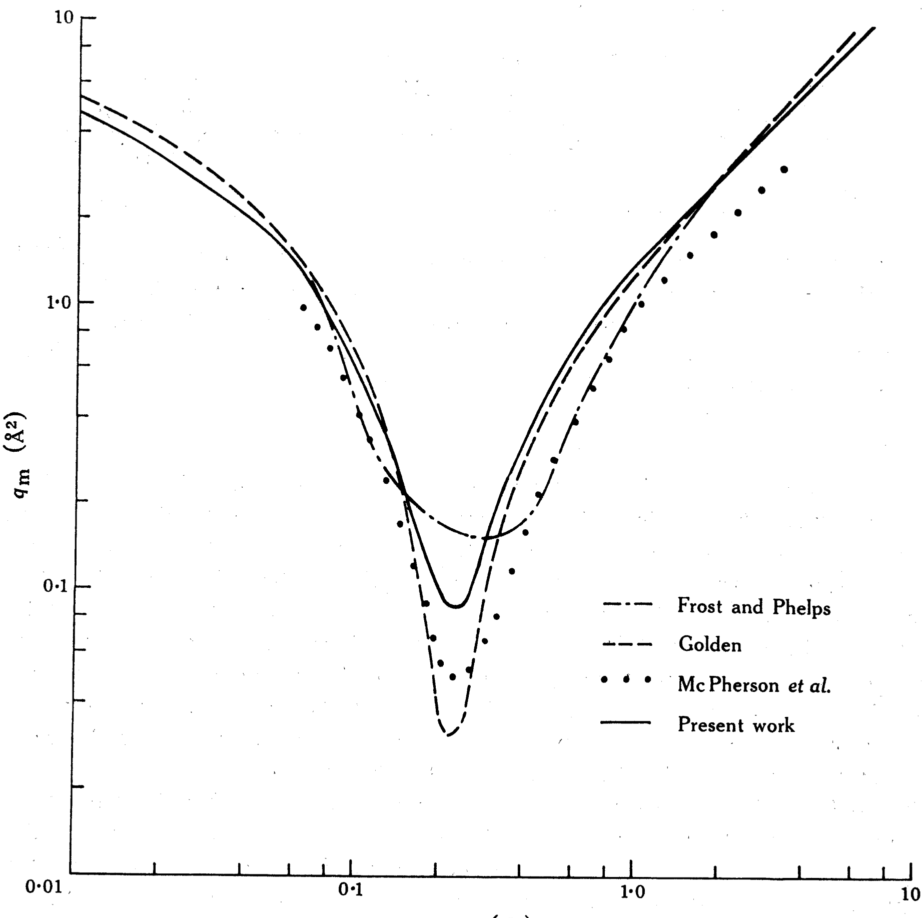

Fig. 4. Comparison of the present result for the momentum transfer cross section $q_m(\varepsilon)$ for electron–argon scattering with the previous determinations by Frost and Phelps (1964), Golden (1966) and McPherson *et al.* (1976).

5. Discussion

The cross section derived in this work is compared in Fig. 4 with the results of Frost and Phelps (1964), Golden (1966), and McPherson *et al.* (1976). For the sake of clarity the analyses based on Ramsauer and Kollath's (1932) measurements have not been included, and the reader is referred to Frost and Phelps's paper for a discussion of the earlier work.

None of the previous estimates of $q_m(\varepsilon)$ are compatible with the data used to derive the present cross section. This can be seen from Fig. 5 where the values of D_T/μ at 294 K used in this work are compared with the values predicted by the three other cross sections in Fig. 4. The results of Golden (1966) and McPherson *et al.*

(1976) predict values that are too high while Frost and Phelps's (1964) cross section gives values that are lower than experiment. The large discrepancy between our cross section and Golden's is partly due to his use of MERT at energies beyond the range of validity of the expansions involved. The reason for the discrepancy between the present work and the microwave results of McPherson *et al.* is not so easily determined. However, it seems probable that diffusion cooling effects (Rhymes and Crompton 1975) would significantly distort the electron energy distribution from the thermal Maxwell distribution assumed by McPherson *et al.* in their analysis.

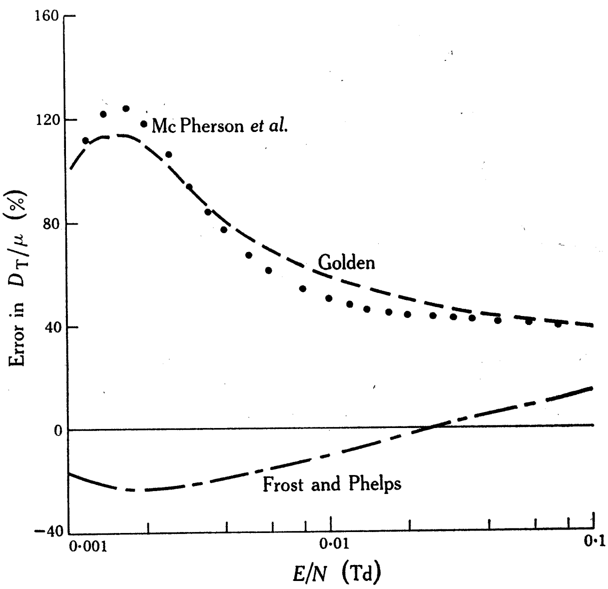

Fig. 5. Errors in the predicted dependence of D_T/μ on E/N at 294 K using the previous determinations of the electron–argon momentum transfer cross section shown in Fig. 4.

In Fig. 6 the experimental D_L/μ values of Robertson and Rees (1972) are compared with those computed using the present cross section. Since the experimental values are quoted as upper limits to the true values it can be seen that the present cross section is compatible with the Robertson and Rees results. The cross sections of Frost and Phelps (1964) and Golden (1966) were also used to compute D_L/μ values for comparison with Robertson and Rees's data. Fig. 6 shows that these cross sections give rise to D_L/μ values significantly above the experimental upper limit.

Theoretical studies of electron–argon scattering have so far been based on a variant of the polarized orbital method of Temkin (1957), usually referred to as the adiabatic exchange approximation. It can be seen from Fig. 7 that there is fair agreement between the present derivation and the theoretical treatments (Thomson 1966, 1971; Garbaty and LaBahn 1971; D. W. Walker, personal communication). However, it is important to remember that large normalization corrections have been applied to the potentials used in the calculations to ensure that the predicted polarizabilities agree with the experimental values. It is shown by Garbaty and LaBahn that without the normalization corrections the theoretical cross sections of Fig. 7 would bear little resemblance to any of the experimental work plotted in Fig. 4.

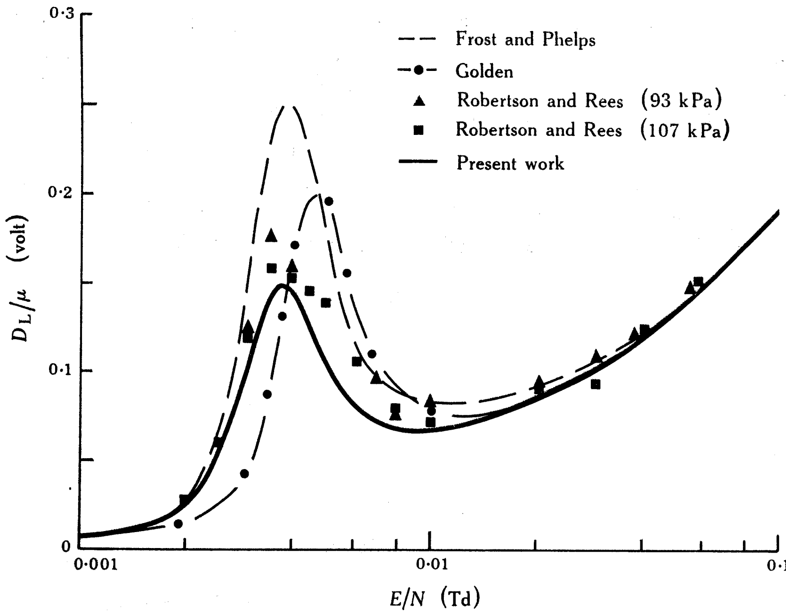

Fig. 6. Variation of measured and calculated values of D_L/μ with E/N at 90 K. The experimental values of Robertson and Rees (1972) at 93 and 107 kPa are compared with those calculated with the present cross section and the cross sections of Frost and Phelps (1964) and Golden (1966).

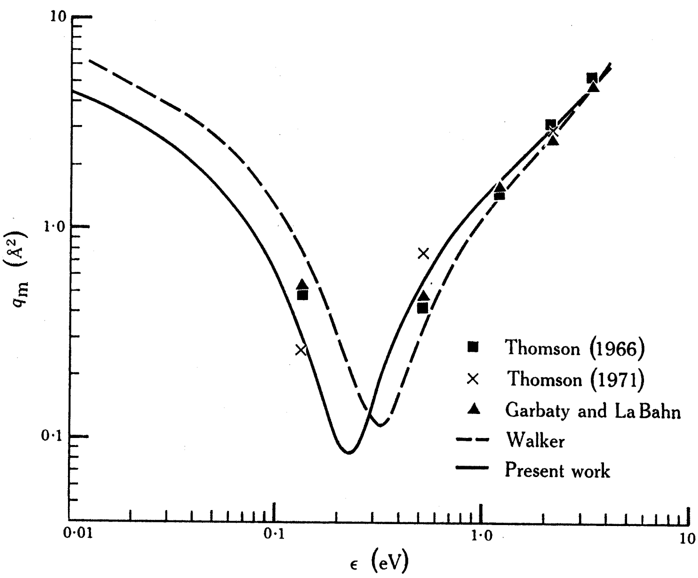

Fig. 7. Comparison of the present cross section with previous theoretical determinations by Thomson (1966, 1971), Garbaty and LaBahn (1971) and D. W. Walker (personal communication).

This is in contrast to the case of neon where the adiabatic exchange approximation without normalization results in good agreement between measured and predicted polarizabilities, and where there is good agreement with measured total and momentum transfer cross sections above $0 \cdot 2$ eV.

Collective Effects

It might be argued that our determination of $q_m(\varepsilon)$ is subject to errors due to the influence of multiple scattering effects on the experimental data taken at high gas number densities and low E/N. The possibility of errors in the D_T/μ measurements from this source has been discussed by Milloy and Crompton (1977*a*), but due to the relatively small range of pressures that could be used at the lowest E/N values they were not able to definitely eliminate collective effects as a source of error. However, the recent experimental data of Rhymes (1976) for argon–hydrogen mixtures considerably strengthen the case against significant errors having arisen from this source. Using a small proportion ($\sim 5\%$) of hydrogen to suppress the effect of diffusion cooling, Rhymes measured the diffusion coefficient for thermal electrons in argon at 293 K at pressures less than 14 kPa, that is, using densities about 200 times smaller than the densities used in the D_T/μ measurements of Milloy and Crompton and 20 times smaller than the densities used in Robertson's (1977) mobility measurements at low E/N and low temperature. The cross section obtained from the present analysis is consistent with Rhymes's results, suggesting that the data from which it was derived were not significantly affected by the large neutral densities that were used for the W and D_T/μ measurements.

6. Conclusions

The cross section derived here would appear to be the most accurate available, despite the relatively large errors inherent in the application of the swarm technique to argon. This conclusion is based on the comparison of the transport coefficient values predicted with the available cross sections (Figs 3, 5 and 6). Unless there is an error in transport theory, which in this case would have to account for more than a factor of two in the calculation of D_T/μ, neither of the cross sections determined by alternative techniques is compatible with the transport coefficients obtained from DC swarm measurements. The discrepancy between Frost and Phelps's (1964) cross section and ours, both of which were obtained using the same method, can be attributed to the development of swarm techniques in recent years. There is no evidence from other applications of the swarm technique of any fundamental source of error. On the contrary, there is excellent agreement at energies of a few electron volts between all recent *ab initio* calculations of the electron–helium momentum transfer cross section, the beam determination of Andrick and Bitsch (1975) and the swarm derivation of Milloy and Crompton (1977*b*).

The error limits on this derivation of the electron–argon momentum transfer cross section compare unfavourably with the limits placed on the corresponding derivations of the helium and neon cross sections. This is due not to any decrease in the accuracy of the experimental data for argon but to uniqueness problems associated with the Ramsauer–Townsend minimum. It is doubtful whether a significantly more accurate estimate of the cross section could be obtained from swarm data. Certainly more accurate experimental results would help in this regard but significant improvements in experimental accuracy would be difficult to achieve. In particular it would not be possible to use much higher gas number densities without a full understanding of collective phenomena and the effect of dimers on the measurements.

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