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Drift Velocities of Low Energy Electrons in Argon at 293 and 90 K

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Abstract

The drift velocity of electrons in argon has been measured at 90 K in the range $0{\cdot}002 \leqslant E/N$ (Td) $\leqslant 0{\cdot}7$ and at 293 K in the range $0{\cdot}01 \leqslant E/N$ (Td) $\leqslant 1{\cdot}0$. The high sensitivity of the drift velocities to the presence of small quantities of diatomic impurities, particularly nitrogen, was demonstrated by adding known but trace quantities of diatomic gases. In this way it was observed that the presence of 6 p.p.m. nitrogen in the argon gave errors in the drift velocity of up to 3%. As a result it was necessary to purify research grade gas before use. A full discussion is given of the errors incurred in the measurements.

1. Introduction

Over the past 50 years, since the early measurements of Ramsauer (1921) and Townsend and Bailey (1922), an extensive literature has been built up concerning the cross section for collisions between low energy electrons and argon atoms, and the electron transport properties which depend on this cross section. Much of this interest has been related to the strong energy dependence at low energies of q_s, the total elastic scattering cross section, and q_m, the momentum transfer cross section. In particular the Ramsauer–Townsend minimum has been of considerable theoretical interest. However, the most recent values available for q_m, those of Frost and Phelps (1964), derived from DC swarm data, and Golden (1966), derived by applying modified effective range theory to the q_s measurements of Golden and Bandel (1966), differ by about a factor of five in the vicinity of the minimum.

There are some formidable problems associated with investigating elastic scattering of electrons with energies close to that corresponding to the Ramsauer–Townsend minimum. This minimum is known to occur in an energy range of about 0·2–0·3 eV, which is close to the lower energy limit of single collision beam experiments (see e.g. Crompton 1969; Bederson and Kieffer 1971). In addition, the available cross sections for electron–argon atom collisions indicate that at energies at or near the minimum the cross section changes so rapidly with energy that even for an electron beam with an energy width of 10 meV the value of the cross section would vary by as much as 50% within the energy range of the beam. This nullifies the assumption, universally made in such experiments, that q_s is substantially constant over the range of energies within the electron beam.

Even for swarm experiments, in which it is usually possible to achieve mean swarm energies close to thermal energies, there are problems in reaching sufficiently low energies while still retaining adequate accuracy in the measurement of the transport

properties from which the cross section is derived. These difficulties are primarily associated with two facts. First, electrons with energies close to the minimum make relatively few collisions. Second, because of the large mass difference between electrons and argon atoms, electrons lose only a small fraction of their energy at each collision. Thus, even for very low values of the parameter E/N, the ratio of electric field strength to gas number density, the mean energy of the electron swarm is many times the thermal value. Preliminary calculations indicated that, in order to investigate the momentum transfer cross section in the region of the minimum, it is necessary to measure transport coefficients for values of $E/N < 0{\cdot}01$ Td. Only Pack and Phelps (1961) and Warren and Parker (1962) have previously reported transport measurements in argon at such low values of E/N.

In spite of the problems associated with obtaining experimental data of adequate accuracy, analysis of swarm measurements seems to offer the best available method for obtaining the value of $q_m(\varepsilon)$ for values of energy ε in the vicinity of the minimum. It is doubtful, however, whether swarm data with an accuracy of better than $\sim 10\%$ have previously been available.

The transport coefficient which can presently be measured with greatest accuracy is the drift velocity W (Elford 1971). In order to measure W at low values of E/N and still achieve a satisfactorily high accuracy, it has been the recent practice to use low gas temperatures thus allowing for increased values of N without exceeding atmospheric pressure. At the same time there is a reduction in the molecular thermal energy and thus a reduction in the lowest mean swarm energy which may be attained (e.g. Pack and Phelps 1961; Crompton *et al.* 1970*a*; Robertson 1972).

Pack and Phelps (1961) made their measurements of W at 77 K. Since this temperature is below the boiling point of argon it is not possible to use high number densities. In order to achieve sufficiently low values of E/N, they were forced to resort to very low values of E which can lead to difficulties with surface potentials (see Crompton *et al.* 1967).

Warren and Parker (1962) measured D/μ, the ratio of the transverse diffusion coefficient to the mobility, at the boiling point of liquid argon (87 K), where higher number densities are possible. However, measurements of this transport coefficient are often subject to greater error than those of the drift velocity (Crompton *et al.* 1967).

The measurements presented in this paper were undertaken primarily to provide a more accurate set of drift velocities than previously available for use as the basis for determining $q_m(\varepsilon)$ at energies about the Ramsauer–Townsend minimum. Measurements were made both at 293 K and 90 K, the boiling point of liquid oxygen.

2. Discussion of Experiment

(*a*) *Drift Tube*

The apparatus used was that described previously by Crompton *et al.* (1968), with the drift length extended to 10 cm (Crompton *et al.* 1970*a*). Details of its construction and operation have recently been given by Elford (1971). The method used was that of Bradbury and Nielsen (1936) with the procedure following the pattern established for maximum precision by Lowke (1963) and Elford (1966).

(*b*) *Temperature Determination*

Some comment is needed on the gas temperature measurement when the drift velocity apparatus was immersed in liquid oxygen contained in a stainless steel dewar.

It had previously been established (Crompton *et al.* 1970*a*) that at 77 K the thermocouples at the top and bottom of the drift regions show no detectable thermal gradient and indicate a temperature which agrees with the calculated boiling point of the bath to within 0·1 K. The temperature of the gas when the tube was immersed in liquid oxygen was therefore taken to be the value of the boiling point of the oxygen corresponding to the barometric pressure. No method was readily available to measure the impurity level in the liquid oxygen which was stated to be less than 0·4%, the chief component of which was argon. This level would introduce a change in boiling point of less than 0·1%. The thermocouples were used to confirm that thermal gradients were absent and to monitor changes in temperature which were generally extremely small (<0·1 K) and which reflected changes in atmospheric pressure. A value of 0·3% is estimated for the uncertainty of the temperature for these measurements.

At room temperature the drift tube was immersed in water. The temperature stability was better than 0·1 K per hour and the actual temperature was measured with an uncertainty of less than 0·1%.

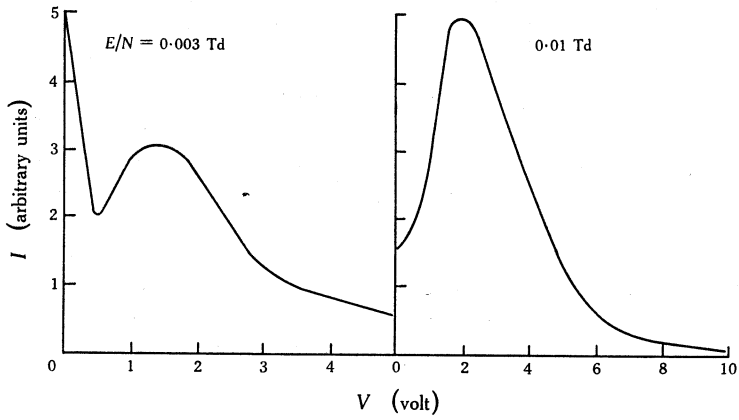

Fig. 1. Transmission characteristics of the electrical shutters, showing the transmitted current I as a function of the bias voltage V for two values of E/N.

(*c*) *Gas Number Density*

At 90 K argon departs significantly from ideal gas behaviour. It was necessary to allow for this departure in relating the number density N to the pressure p. The values of p used were calibration points of the pressure gauge. The value of the second virial coefficient $B(T)$, measured by Holborn and Otto (1925) and quoted by Hirschfelder *et al.* (1954), was used to calculate N for each value of p at the prevailing temperature. The voltage required to produce the required value of E/N was then applied. Had the correction to N not been applied, the value of E/N would have been in error by up to 0·7% at 106 kPa, the maximum pressure used. At the temperature and pressures used, corrections introduced by the third virial coefficient are negligible.

(*d*) *Shutter Characteristics*

In most gases the electrical shutters of the drift tube transmit the maximum electron current when the potential difference between adjacent wires is zero. The current

then falls as the potential difference increases from zero. It has been observed, however (Pack and Phelps 1961; Robertson 1972), that in neon and argon, where the cross section varies rapidly with energy, the electrical shutters have quite different transmission characteristics.

In Fig. 1 it can be seen that at $E/N = 0{\cdot}003$ Td, although there is the usual sharp maximum at zero bias voltage, there is a secondary maximum which is quite broad but at a higher bias voltage, whereas at $E/N = 0{\cdot}01$ Td there is a pronounced minimum in the transmitted current at zero bias voltage. The principal effect of these unusual shutter characteristics is to produce maxima in the plots of transmitted current versus frequency that are less well resolved than those when a sharply defined pulse is produced at the first shutter. Robertson and Rees (1972), in measuring longitudinal diffusion in argon, operated the shutters with a square wave rather than a sine wave signal in order to produce such a sharp pulse. In the present measurements the shutters were operated with a sine wave signal. In cases where there is a minimum in the transmitted current at zero bias the first shutter will produce a bimodal electron pulse. However, there was no abnormality in the curves of transmitted current I as a function of the frequency f of the AC signal applied to the shutters because the shapes of the current maxima were dominated by the diffusion of the electron groups during the transit time. As in previous experiments (see e.g. Elford 1971), the integrated current at the collector was in the range 10^{-13}–10^{-11} A. The frequencies corresponding to the current maxima were found to be independent of the amplitude of the AC shutter voltage, provided that the amplitude was high enough to produce adequately sharp maxima. It is clear then that the unusual shutter characteristics do not affect the measured values of the drift velocity.

In some cases it was necessary to use AC shutter voltages with peak to peak values of the order of 30% of the total voltage between the shutter planes in order to resolve the current peaks. Again it was found that the values of the frequencies corresponding to maximum transmitted current were independent of this voltage.

For values of $E/N < 0{\cdot}005$ Td it was particularly difficult to produce well-resolved current peaks. At these very low values of E/N longitudinal diffusion becomes very large causing the current peak to be poorly resolved (Robertson and Rees 1972).

3. Gas Samples and Purity

Several authors have reported measurements of drift velocities in argon to which known amounts of impurity, in particular nitrogen, were added (e.g. Klema and Allen 1950; Colli and Facchini 1952; Kirshner and Toffolo 1952). The work of these authors established that quite low levels of nitrogen have a very marked effect on the drift velocities for values of E/N above about 0·2 Td. However, the lowest level of nitrogen for which results have been reported appears to be 0·1% (Colli and Facchini 1952), which is sufficient to increase W by a factor of two at $E/N = 0{\cdot}2$ Td. There have been no reports on the effects of impurity levels of a few p.p.m. However, the fact that such levels of nitrogen produce significant changes in the measured drift velocities became apparent in preliminary experiments. In previous measurements with the inert gases helium and neon (Crompton *et al.* 1970*a*; Robertson 1972) it was found that the purity of research grade gases was adequate to avoid errors from impurities although it had been shown (Robertson 1972) that drift velocities in neon are very sensitive to nitrogen impurities at the level of a few p.p.m.

In the present investigation, initial measurements were made using research grade argon from the Matheson Gas Company and admitting the gas directly to the UHV system and drift tube. Gas was used from two cylinders, which will be designated A and B. It was immediately apparent that the gas samples gave significantly different values for the drift velocities at 293 K, the differences being as large as 3·5% for $E/N = 0{\cdot}4$ Td. However, the magnitude of the difference was strongly dependent on E/N. As a result of this discrepancy, an investigation of the effect of impurities at very low levels was undertaken and at the same time attempts were made both to measure the impurity levels and to improve the purity.

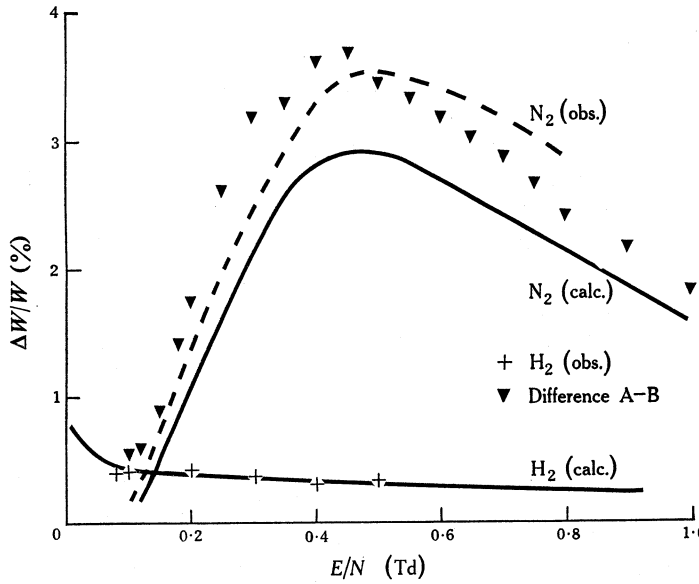

Fig. 2. Plots of the changes ΔW in W due to the addition of gaseous impurities, as functions of E/N. The continuous curves show the calculated effects of adding 6 p.p.m. of N_2 and of H_2 to samples of argon, while the dashed curve and the symbol + respectively show the corresponding observed effects. The difference between the results of samples taken from cylinders A and B (plotted as ▼) is clearly consistent with the effect of adding N_2.

(*a*) *Effect of Impurities*

By adding small measured levels of these impurities to argon samples a check was made on the effect of H_2 and N_2 impurities on the values of the drift velocity. Results are shown in Fig. 2. It is obvious that while H_2 is not such a serious contaminant, even 1 p.p.m. of N_2 would lead to significant errors. It is also apparent that the effect on W of adding N_2 to a sample of argon is consistent with the difference between the results obtained using gas from cylinders A and B.

As a check on these measurements, values of W were calculated for Ar–N_2 mixtures of varying proportions. The solution of the Boltzmann equation for the case of gaseous mixtures has been described by Lowke *et al.* (1973). The argon momentum transfer cross section used in these calculations was that reported by Milloy *et al.* (1977; present issue p. 61), the nitrogen cross sections were those of Engelhardt *et al.* (1964) and the hydrogen cross sections were those of Crompton *et al.* (1970*b*)

and Gibson (1970). It can be seen from Fig. 2 that there is good agreement between the measured and calculated effect of adding 6 p.p.m. of N_2 or 6 p.p.m. of H_2 to Ar.

The results for the two gas samples are clearly consistent with the hypothesis that cylinder A contained a few p.p.m. of N_2 more than cylinder B and demonstrate the remarkable effect of very low levels of N_2 on the electron drift velocities. The severity of this effect may not have been fully appreciated previously. For instance, Pack and Phelps (1961) reported that the argon used in their measurements had a mass spectrometric analysis showing less than 50 p.p.m. impurity. Had this impurity been N_2 then errors greater than 25% would have occurred in W for some values of E/N.

The necessity for accurate knowledge of impurity levels led Elford and Milloy (1972) to develop a method of analysing argon samples for small traces of molecular impurity. Their work showed that cylinders A and B contained $4{\cdot}8 \pm 0{\cdot}6$ and $0{\cdot}8 \pm 0{\cdot}3$ p.p.m. of N_2 respectively. These impurity levels are within the specifications of the research grade argon gas (<5 p.p.m. N_2).

A second problem with impurities was encountered with measurements at room temperature when E/N lay within the range 0·01–0·1 Td. The values of drift velocity were found to change with time, rising by up to 1% per day in some cases. The cause of this was evidently a source of impurity from within the system. Since no corresponding effect was observed in measurements at 90 K, the impurity was apparently condensible. The drift tube containing argon at 20 kPa was subjected to baking at 180°C for 24 hr. Following this treatment the values of drift velocity were found to be lower and independent of time.

(*b*) *Gas Purification*

Before final data were obtained, additional purification of the argon was obviously desirable. Therefore, for the results shown in Section 4 the gas was passed through a quartz tube heated to 850°C which contained previously outgassed titanium. Elford and Milloy (1972) were unable to detect N_2 impurity in gas treated this way and estimated the maximum level to be less than 0·3 p.p.m.

Several earlier workers (e.g. Klema and Allen 1950; Colli and Facchini 1952; Kirshner and Toffolo 1952; Bowe 1960) adopted the practice of purifying their gas samples over heated calcium or barium. There is evidence that these procedures were effective to a large extent. However, without an adequate method for measuring low levels of molecular impurities it was not possible for these authors to be sure that nitrogen contamination was insignificant.

4. Results and Discussion

(*a*) *Drift Velocities*

In the absence of broadening of the pulse by diffusion and diffusive effects at the shutters, the drift velocity would be given simply by $W = h/t$, where h is the drift distance and t the transit time. On the assumption that the diffusion equation is applicable throughout the drift space, the effects of diffusion can be allowed for by using the equation (Lowke 1962; Huxley and Crompton 1974)

$$h/t = W' = W\left(1 + \frac{C(D_L/\mu)}{V}\right) = W\left(1 + \frac{C(D_L/\mu)}{(E/N)Nh}\right), \qquad (1)$$

where D_L is the longitudinal diffusion coefficient, V the potential difference between

the shutter planes and C a constant that depends in part on the mode of operation of the shutters. Although the recent work of Skullerud (1974) shows that the use of the diffusion equation is inadmissible within the region adjacent to the boundaries (i.e. the shutters) terminating the drift space, the validity of a relation of the form of equation (1) has been demonstrated in many instances (Elford 1971), at least to the

Table 1. Electron drift velocities in argon at 293 and 89·6 K

E/N (Td)	W' (10^5 cm s^{-1}) at pressure (kPa) of:										Best est. W (10^5 cm s^{-1})
	13·4	20·7	26·9	33·1	40·3	53·7	66·1	79·6	93·0	106·4	
						(a) 293 K					
1·0		2·977	2·970								2·95
0·9		2·910	2·905	2·899							2·88
0·8		2·838	2·831	2·826							2·80
0·75		2·800	2·792	2·787	2·784						2·76
0·70		2·757	2·749	2·745	2·742						2·73
0·65		2·713	2·704	2·700	2·697						2·68
0·60		2·663	2·656	2·652	2·648						2·63
0·55		2·614	2·604	2·599	2·596	2·591					2·58
0·50		2·557	2·549	2·543	2·539	2·535					2·52
0·45			2·487	2·480	2·477	2·473	2·471				2·46
0·40			2·419	2·413	2·410	2·405	2·402				2·39
0·35				2·337	2·333	2·327	2·325				2·31
0·30				2·250	2·245	2·240	2·238				2·23
0·25				2·150	2·146	2·141	2·137				2·13
0·20						2·021	2·018	2·014		2·001	2·00
0·18						1·965	1·961	1·959		1·956	1·94(7)
0·15						1·872	1·869	1·865		1·863	1·85(3)
0·12						1·765	1·761	1·757		1·753	1·74(1)
0·10						1·680	1·677	1·673		1·669	1·65(6)
0·08							1·573	1·570		1·567	1·55(7)
0·05					1·407		1·388	1·385		1·381	1·37(0)
0·03								1·218		1·212	1·20
0·02								1·110		1·103	1·08
0·01								0·963		0·956	0·94
						(b) 89·6 K					
0·70	2·758										2·74
0·65	2·714										2·70
0·60	2·665										2·65
0·55	2·614										2·60
0·50	2·559										2·54
0·45	2·498	2·490									2·48
0·40	2·428	2·421									2·41
0·35	2·353	2·345	2·340								2·33
0·30	2·265	2·258	2·253								2·24
0·25	2·165	2·156	2·152	2·148							2·14
0·20	2·045	2·036	2·032	2·028	2·026						2·02
0·18		1·982	1·977	1·973	1·971						1·959
0·15		1·889	1·884	1·880	1·878	1·874					1·868
0·12		1·780	1·774	1·771	1·769	1·765	1·762				1·756
0·10		1·697	1·690	1·686	1·684	1·680	1·678	1·675	1·673		1·668
0·08			1·593	1·589	1·587	1·582	1·580	1·577	1·574		1·569
0·06			1·478	1·473	1·470	1·467	1·464	1·460	1·458	1·455	1·450
0·05			1·411	1·406	1·403	1·399	1·396	1·393	1·390	1·388	1·382
0·04			1·335	1·330	1·327	1·322	1·319	1·316	1·314	1·311	1·305
0·03				1·242	1·238	1·233	1·230	1·227	1·224	1·222	1·218
0·02				1·135	1·132	1·126	1·123	1·120	1·117	1·115	1·107
0·015				1·071	1·068	1·062	1·059	1·056	1·053	1·051	1·045
0·01			0·998	0·991	0·9871	0·9810	0·9779	0·9742	0·9715	0·9689	0·961
0·008						0·9338	0·9296	0·9260	0·9229	0·9205	0·906
0·006						0·8434	0·8393	0·8343	0·8307	0·8273	0·810
0·005							0·7380	0·7346	0·7278	0·7234	0·710
0·004								0·5403	0·5339	0·5283	0·520
0·0035									0·4057	0·4011	0·401
0·003									0·2883	0·2861	0·286
0·0025									0·2047	0·2029	0·204
0·002									0·1500	0·1483	0·149

extent required to make the corrections to W' necessary to obtain W, since these are usually less than 1%.

The abnormally large values of $(D_L/\mu)/(E/N)$ in argon (Milloy *et al.* 1977) result in the differences between W' and W being much larger than those observed in other gases. Thus the measured values of W' listed in Table 1 show a significant dependence on pressure, and uncertainty in the correction to be applied to W' is the major source of error in some of the best-estimate values of W.

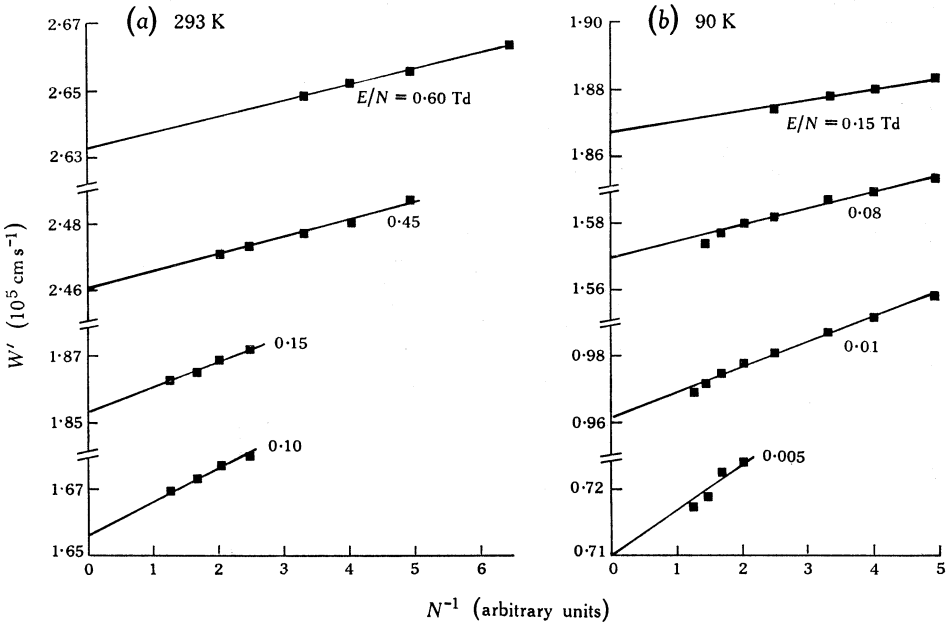

Fig. 3. Variation of the experimental drift velocity W' with N^{-1} for electrons in argon at 293 and 90 K and selected values of E/N.

In order to obtain the values of W shown in Table 1, values of W' were plotted against N^{-1} and the resultant straight lines of best fit extrapolated to $N^{-1} = 0$. Fig. 3 shows a selection of such curves, chosen to illustrate some of the features revealed. In most cases for measurements at 293 K the curves were found to be linear to within the accuracy of W'. However, at a few of the highest and lowest values of E/N there were insufficient points to allow W to be determined with the accuracy that is generally possible. In the case of the measurements at 90 K some departure from linearity in the W'–N^{-1} curves was observed as the pressure approached the highest values, i.e. as $N^{-1} \rightarrow 0$. This departure is generally not more than 0·3% and is therefore within the error limits of the data. It does, however, increase the uncertainty in the values of W and raises the possibility of the onset of some density-dependent phenomenon. Although such possibilities have been raised before (Kivel 1959; O'Malley 1963; Legler 1970), the densities used in the present measurements are not high enough to shed any new light on this phenomenon.

For several reasons the accuracy of the best-estimate values of W shown in Table 1 varies considerably with E/N. There is the uncertainty in the extrapolation of the W'–N^{-1} curves referred to above. In addition there is considerable variation in the

possible effect on the measurements of uncertainty in the experimental parameters leading to errors in E/N. A 1% error in E/N produces an error in W' which varies from less than 0·2% for $E/N \sim 0{\cdot}7$ Td to more than 2% for $E/N \sim 0{\cdot}003$ Td.

Taking all the sources of error into account the values of W in argon at 90 K are believed to be accurate to the following limits over the ranges of E/N indicated:

E/N (Td)	$\leqslant 0{\cdot}005$	0·005–0·01	0·01–0·1	$\geqslant 0{\cdot}1$
Error (%)	±4	±4 → ±1	±1 → ±2	±2

The values of W at 293 K are believed to be in error by less than ±2% for $E/N >$ 0·05 Td and ±3% for $E/N \leqslant 0{\cdot}05$ Td.

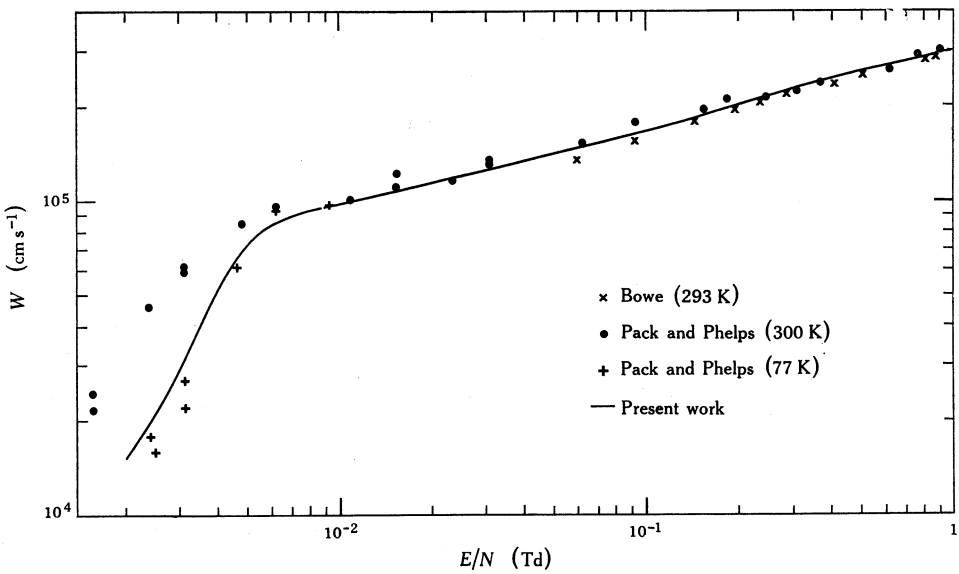

Fig. 4. Comparison of the present best-estimate data for the drift velocity W with the results of Bowe (1960) and Pack and Phelps (1961). The errors associated with the present data are discussed in the text. Bowe's results were plotted from a tabulation of his data.

(*b*) *Comparison with Other Measurements*

Fig. 4 shows a comparison between the present best-estimate data and those of Bowe (1960) and Pack and Phelps (1961). The present results at 293 and 90 K cannot be distinguished on the graphs, since at the lowest value of E/N used (0·01 Td) at 293 K the value of W is only 3% different from that at 90 K. Thus even at this low value of E/N the energy distribution of the electrons is still dominated by the electric field rather than by the thermal energy of the gas molecules. The data of Bowe at 293 K show only small scatter and lie generally somewhat below the present results. This is consistent with the pattern found in helium (Crompton *et al.* 1970*a*) and neon (Robertson 1972). Over most of the range of E/N, the results of Pack and Phelps at 300 K lie within about 10% of the present measurements, which is about the magnitude of the scatter on their data. However, for values of $E/N \lesssim 10^{-1}$ Td the present results generally lie below those of Pack and Phelps, this trend increasing as E/N decreases down to 10^{-2} Td, the lower limit of the present measurements.

There are no other data with which the results at 90 K can be compared directly. However, the results of Pack and Phelps at 77 K serve as some comparison.

Other measurements not shown in Fig. 4 include those of Nielsen (1936), Colli and Facchini (1952), Kirshner and Toffolo (1952) and Wagner *et al.* (1967), all of which agree with the present results within the accuracy of the measurements. The results of Bortner *et al.* (1957) are about 20% higher than these, while the even higher values reported by Klema and Allen (1950) and English and Hanna (1953) for $E/N > 0{\cdot}6$ Td have been attributed to impurities (see e.g. Colli and Facchini 1952; Bowe 1960). In most cases the scatter on these earlier measurements exceeds the total error limit placed on the present results. In addition, the question of impurity levels in gases used previously is unresolved. In several cases the agreement with the present measurements suggests that the N_2 levels were quite low, but since measurements were not made of the impurity levels in the samples actually used in the experiment no definite statement can be made.

Apart from the measurements of Pack and Phelps (1961), the drift velocities presented here are the only ones available which extend to sufficiently low values of E/N to be of use in trying to elucidate the momentum transfer cross section for energies below about 0·5 eV. The accuracy of the new data represents a significant improvement on the accuracy of those previously published.

(c) *Momentum Transfer Cross Section*

The drift velocities reported in this paper have been used, together with values of D_T/μ at 294 K reported in the following paper (Milloy and Crompton 1977), to derive the momentum transfer cross section for electrons in argon in the energy range 0–4 eV (Milloy *et al.* 1977; present issue p. 61).

Acknowledgments

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