

The Ratio D_T/μ for Electrons in a 0.5% Hydrogen-Krypton Mixture

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Abstract

The ratio D_T/μ for electrons in a 0.4673% H₂-99.5327% Kr mixture at 295 K has been measured by the Townsend-Huxley method in order to obtain data for use in the derivation of the momentum transfer cross section for krypton. The values of E/N and pressures used ranged from 0.025 to 1.4 Td and 13.43 to 68.16 kPa respectively. At a given value of E/N and pressure the derived values of D_T/μ were found to depend on the choice of the mathematical expression which relates the measured current ratio to D_T/μ . However, the values obtained using each of the expressions, when extrapolated to infinite pressure, agreed to within 0.5% for all E/N values ≥ 0.07 Td. The validity of the current ratio relations is discussed. The uncertainty in the best estimate values of D_T/μ is considered to be <1.5% for $E/N \geq 0.07$ Td and 2% for $E/N < 0.07$ Td.

1. Introduction

Although there have been at least eighteen published momentum transfer cross sections for electrons in krypton the cross sections differ widely, even in the case of those published in the last few years. The most common procedure has been to derive the cross section by the analysis of electron transport coefficient data for pure krypton. The earliest such derivation was that of Frost and Phelps (1964) who used the drift velocity v_{dr} data of Pack *et al.* (1962). Other derivations using transport data have been those of Hoffman and Skarsgard (1969), who used their measurements of conductivity ratios; Koizumi *et al.* (1986), who used their measurements of the ratio D_T/μ (where D_T is the transverse diffusion coefficient and μ the electron mobility); Hunter *et al.* (1988) who used their v_{dr} data; and Nakamura (1989) who used his data for v_{dr} and ND_L (where N is the gas number density and D_L the longitudinal diffusion coefficient). Derivations have also been made using hydrogen-krypton mixtures. England and Elford (1988) used their drift velocity data for mixtures of 0.47% and 1.69% hydrogen in krypton, while Mitroy (1990) used modified effective range theory (MERT) in conjunction with the v_{dr} data of both Hunter *et al.* and England and Elford.

The difficulties in obtaining accurate transport coefficient data and deriving the momentum cross section σ_m for gases which have Ramsauer-Townsend minima have been discussed in detail by both Milloy *et al.* (1977) and England and Elford

(1988). These latter authors pointed out that a significant gain in the sensitivity of the drift velocity to the cross section at energies in the vicinity of the minimum can be achieved by the use of suitable gas mixtures. The cross section which they derived not only predicted, to within their respective stated uncertainties, their drift velocity data for two hydrogen–krypton gas mixtures, but also (with some exceptions) the drift velocity data of Hunter *et al.* for electrons in pure krypton. The exceptions were data points at E/N values between about 0.02 and 0.04 Td (E is the electric field strength). Values in this E/N range appear anomalous and are also not fitted by the cross section derived by Hunter *et al.*

Mitroy (1990) criticised the cross section of England and Elford on the grounds that it was derived without constraint (except for the requirement that the calculated drift velocities fitted the experimental values) and was not well described by MERT. Mitroy derived a new cross section assuming the form given by MERT from 0 to 1 eV and an average of the cross sections of Hunter *et al.* and England and Elford at higher energies. The MERT parameters were obtained by using standard nonlinear optimisation techniques and the v_{dr} data of both Hunter *et al.* and England and Elford. This new cross section has a much deeper and narrower minimum at 0.51 eV and predicts the v_{dr} data generally to within the stated uncertainties.

It has long been recognised that values of D_{T}/μ are much more sensitive to the momentum transfer cross section in the region of the minimum than are v_{dr} values for either pure krypton or hydrogen–krypton mixtures (Milloy *et al.* 1977; England and Elford 1988). However, measurements of D_{T}/μ in pure krypton pose a number of experimental problems, including the need to use high gas pressures and the effect of trace levels of molecular impurities. It is probable that the only such data available (Koizumi *et al.* 1986) are subject to significant error (Ogawa, personal communication 1988, cited by England and Elford 1988). These experimental difficulties can be greatly reduced by the use of hydrogen–krypton mixtures as described in Section 2. This section also contains a brief description of the experimental technique used to measure D_{T}/μ . The derivation of this transport coefficient from the measured current ratios posed particular problems which are discussed in some detail in Section 3. The final results are discussed in Section 4. The procedure used to analyse these data to obtain σ_{m} is described in a companion paper (Brennan *et al.* 1993).

2. Apparatus

Measurements of D_{T}/μ were made using the Townsend–Huxley method and the variable length apparatus described in detail by Huxley and Crompton (1974) and shown schematically in Fig. 1. There is one difference between the present apparatus and that described earlier. The gaps between adjacent segments of the collector (i.e. the anode) have been significantly reduced to avoid the uncertainty introduced by the unpredictable way electrons diffuse when they enter the gap between adjacent collector segments. The gap between the central disk and the first annulus and between subsequent annuli is now 0.05 mm (previously the gaps were 0.25 mm between the central disk and first annulus, 0.25 mm between the first and second annuli and 0.5 mm between the next two sets of annuli). All measurements were made with an anode to cathode spacing h of 100.00 mm.

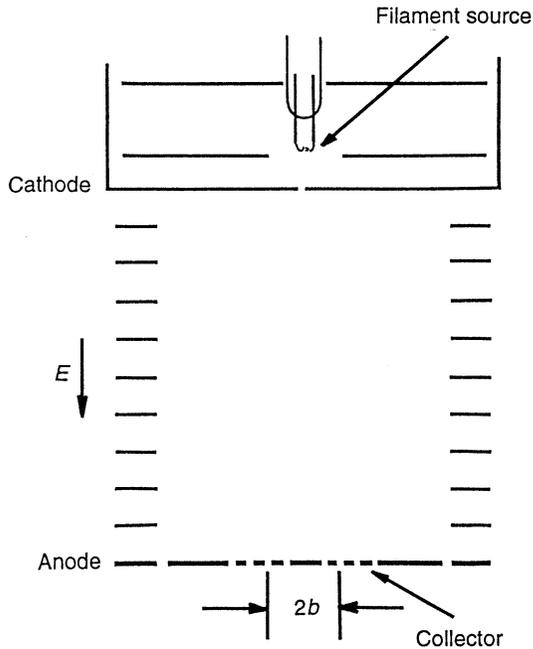


Fig. 1. Schematic diagram of the Townsend-Huxley diffusion chamber.

The electron source was a heated platinum filament, considerable care being taken to ensure that the heat dissipated had no significant effect on the measurements. The proportion of hydrogen chosen (0.467% H_2 -99.533% Kr) was a compromise between a number of factors. It is necessary for the proportion to be as small as possible to maintain a high level of sensitivity to the momentum transfer cross section for krypton at energies in the vicinity of the minimum and also to keep small the effect of the uncertainty in the momentum transfer cross section of hydrogen on the value of D_T/μ for the mixture. The proportion should, however, be sufficiently large to enable measurements of D_T/μ to be made over a wide range of experimental conditions with adequate accuracy (discussed later in this section) and to make the effect of trace levels of impurities insignificant. The mixture proportion chosen was the same as that for one of the mixtures used by England and Elford (1988).

The hydrogen was purified by passage through a heated palladium osmosis thimble and the krypton (AIRCO Research Grade) by passage through titanium pellets held at 800°C. The mixture was made by volume sharing using a mixture vessel similar to that described by Haddad (1983).

Before measurements were commenced with the mixture, a series of measurements were made using pure hydrogen in order to check the accuracy of the system. The values obtained over a wide range of pressures and E/N values differed from those of Crompton *et al.* (1986) by less than 0.5% when the central disk was used as the inner collector and 1% when the central collector consisted of the central disk plus first annulus.

In order to achieve adequate accuracy in measurements of current ratios in the mixture, the experimental conditions were chosen such that the ratio R of the

total current received by the inner collector was between approximately 0.2 and 0.8. This required the inner collector to consist of the central disk plus the first annulus, giving an inner collector of radius $b = 9.9975$ mm and a value of $b/h = 0.099975$.

Because many of the measurements were made using relatively divergent current distributions at the collector it was necessary to account for the fraction of the total current which fell outside the radius of the outermost annulus (42.00 mm). This was done using an iterative procedure when relating the current ratio to the value of D_T/μ (see below). The correction to the calculated value of D_T/μ is a function only of R and increases rapidly as R decreases below about 0.3, being 0.2% at $R = 0.30$, 1.0% at $R = 0.24$ and 2.7% at $R = 0.20$.

The measured values were stable over a period of seven days to within the experimental uncertainty, indicating that the effect of outgassing of the experimental tube was insignificant. The results were also independent of the magnitude of the electron current; increasing the current by a factor of three caused no significant change in the measured values.

All krypton produced since 1945 is contaminated to a varying degree by the radioactive isotope ^{85}Kr as a consequence of atmospheric testing of atomic weapons (The Matheson Company Inc., personal communication). The decay of this isotope (which has a half-life of 10.8 years) by the emission of 0.67 MeV β -particles gives rise to ionisation currents to the inner and outer segments of the collector which are a source of significant uncertainty in the measured current ratios at high gas pressures. These currents were annulled at each pressure before measurements commenced by generating two independent and variable displacement currents in the circuits to the electrode segments. Each current was produced by applying a ramp voltage of variable amplitude and long duration (period of one hour) to a small capacitor (20 pf), constructed to have a very high resistance and low dielectric soakage (see Crompton and Sutton 1952). Because the ionisation currents were independent of the potential difference across the diffusion chamber for potential differences greater than 10 V, and the minimum potential difference used was 30 V, it was not necessary to adjust the compensation for different values of E/N at one gas pressure. The largest compensation current was 4×10^{-14} A.

Relation between R and D_T/μ

The relation used most frequently to obtain D_T/μ from the current ratio R is that derived by Huxley (Huxley and Crompton 1974) assuming isotropic diffusion, i.e.

$$R = i_{\text{inner}}/i_{\text{total}} = 1 - (h/d)\exp[-\lambda_T(d-h)], \quad (1)$$

where i_{inner} is the current received by the inner collector, i_{total} is the total current at the collector, $\lambda_T = E/(2D_T/\mu)$ and $d = (b^2 + h^2)^{1/2}$. This relation has been shown to give values of D_T/μ from current ratios at a single value of E/N which are independent of the chamber length, the collector geometry and the gas pressure over a wide range of these experimental parameters. A particularly detailed study of the Huxley relation has been carried out for the case of hydrogen by Crompton and Jory (1962) and is discussed in detail by

Huxley and Crompton (1974). However, it should be noted that relation (1) is obtained by assuming that:

- (1) the electron number density n is a solution of a diffusion equation which is based on isotropic diffusion;
- (2) the source of electrons (i.e. the hole in the cathode) acts as a source in free space;
- (3) $n = 0$ at the anode plane (i.e. the collector surface).

Since in practice diffusion is not isotropic and it would be expected that it would be necessary to also impose a boundary condition at the cathode of $n = 0$, it is surprising that this relation has given results that are consistent with ‘asymptotic’ values (that is values obtained under conditions where the distribution of n is insensitive to anisotropy and the boundary conditions), even when anisotropy and the boundary conditions can be shown to have a significant influence.

Table 1. Relations derived from the diffusion equation for the current collected by a circular collector of radius b to the total current received

Relation	Boundary conditions		Current ratio relation
	Anode	Cathode	
<i>Anisotropic diffusion</i>			
1	$n = 0$	$n = 0$	$R = 1 - [(h/d') - (1/\lambda_L h)\{1 - (h/d')^2\}](h/d')\exp\{-\lambda_L(d' - h)\}$ (Lowke relation)
2		$n = 0$	$R = 1 - (h/d')\exp[-\lambda_L(d' - h)]$
3	$n = 0$		$R = 1 - (h/d')\exp[-\lambda_L(d' - h)]$
4			$R = 1 - 0.5\{1 + (h/d')\}\exp[-\lambda_L(d' - h)]$
<i>Isotropic diffusion</i>			
5	$n = 0$	$n = 0$	$R = 1 - [(h/d) - (1/\lambda_T h)\{1 - (h/d)^2\}](h/d)\exp\{-\lambda_T(d - h)\}$
6		$n = 0$	$R = 1 - (h/d)\exp[-\lambda_T(d - h)]$ (Huxley relation)
7	$n = 0$		$R = 1 - (h/d)\exp[-\lambda_T(d - h)]$ (Huxley relation)
8			$R = 1 - 0.5\{1 + (h/d)\}\exp[-\lambda_T(d - h)]$

A ratio relation that might be expected to relate the ratio R to D_T/μ more accurately is that obtained by Lowke (1971) by solving the diffusion equation

$$D_T \left(\frac{\partial^2 n}{\partial x^2} + \frac{\partial^2 n}{\partial y^2} \right) + D_L \frac{\partial^2 n}{\partial z^2} - v_{dr} \frac{\partial n}{\partial z} = 0, \tag{2}$$

which allows for anisotropic diffusion, and by imposing the boundary conditions $n = 0$ at both anode and cathode. The ratio relation obtained is

$$R = 1 - [(h/d') - (1/\lambda_L h)\{1 - (h/d')^2\}](h/d')\exp\{-\lambda_L(d' - h)\}, \tag{3}$$

in which $d' = (b'^2 + h^2)^{1/2}$, $b'^2 = (D_L/D_T)b^2$ and $\lambda_L = E/(2D_L/\mu)$.

Other formulae for R have been obtained using different forms of the diffusion equation (i.e. assuming isotropic or anisotropic diffusion) and different assumptions for the boundary conditions (see Table 1). Relation 4 in Table 1 was obtained by

analogy with the solution of Deslodge and Mitchell (1976) for isotropic diffusion, i.e. relation 8 (see also Crompton 1972).

Huxley and Crompton have shown that when b/h is small the Lowke relation can be accurately approximated over a wide range of experimental conditions by the relation

$$R = 1 - [1 + \{0.5 - (D_L/D_T)\}(b/d)^2](h/d)\exp[-\lambda_T(d-h)]. \quad (4)$$

The Huxley relation is a good approximation to the Lowke relation when either the quantity $(b/d)^2$ is small (as is the case in most experimental measurements) or when $D_L/D_T \sim 0.5$ (as is the case for hydrogen, for example, over a wide range of E/N values). In order to avoid uncertainties introduced by the assumption of particular boundary conditions or by the absence of accurate data for the ratio D_T/μ , it has been a common practice (see e.g. Huxley and Crompton 1974) to choose experimental conditions such that the same value of D_T/μ is obtained (to within the statistical scatter), regardless of whether the Huxley or Lowke relation is used.

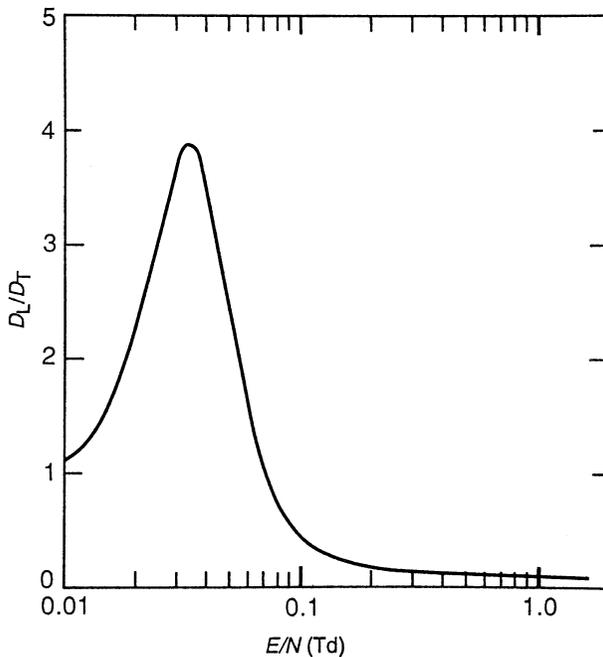


Fig. 2. Ratio D_L/D_T as a function of E/N for electrons in a 0.467% H_2 -99.533% Kr mixture at 295 K calculated from cross sections for hydrogen and krypton.

In the present work, however, significant differences were expected between the two relations because not only is the ratio b/d larger than in many other sets of measurements (in order to accurately measure current ratios for more widely divergent current distributions at the collector), but more importantly the ratio D_L/D_T was expected to be much larger than 0.5 due to the form of

the cross section. This was found to be the case (see Fig. 2) when D_L/D_T was calculated for the present mixture using the multi-term transport code of Ness and Robson (1986). In this calculation we used the momentum transfer cross section for krypton of Mitroy (1990) and the set of cross sections for hydrogen of England *et al.* (1988). Since the ratio rises rapidly to relatively large values for $E/N < 0.1$ Td, it was expected that the differences between the values of D_T/μ derived from the two ratio formulae would be greatest in this range.

Table 2. Values of D_T/μ (in V) for electrons in a 0.4673% H_2 -99.5327% Kr mixture at 295 K obtained using the Huxley relation

E/N (Td)	p (kPa)							Best estimate
	13.43	19.62	26.85	33.05	40.28	53.70	68.16	
0.025						0.0628	0.0625	0.0627
0.03						0.0971	0.0979	0.0975
0.035						0.1390	0.1383	0.138
0.04				0.1833	0.1821	0.1830	0.1827	0.1828
0.05			0.2640	0.2668	0.2651	0.2656	0.2643	0.2652
0.06		0.3416	0.3422	0.3428	0.3410	0.3412	0.3406	0.3416
0.07		0.4119	0.4113	0.4150	0.4121	0.4127	0.4108	0.4123
0.08		0.4792	0.4808	0.4813	0.4790	0.4800	0.4786	0.4798
0.10		0.6045	0.6058	0.6056	0.6045	0.6040	0.6032	0.6046
0.12		0.7213	0.7238	0.7271	0.7229	0.7215	0.7217	0.7230
0.14		0.8327	0.8355	0.8405	0.8341	0.8326	0.8349	0.8351
0.17		0.9894	0.9907	0.9922	0.9885	0.9882	0.9895	0.989
0.20		1.134	1.139	1.137	1.137	1.132	1.134	1.136
0.25		1.353	1.353	1.355	1.348	1.348	1.352	1.352
0.30		1.547	1.547	1.557	1.548	1.544	1.546	1.548
0.35		1.722	1.728	1.729	1.725	1.722	1.723	1.725
0.40		1.885	1.889	1.891	1.888	1.889		1.888
0.50	2.155	2.184	2.181	2.182	2.181	2.184		2.182
0.60	2.410	2.445	2.439	2.443	2.440			2.442
0.70	2.650	2.671	2.669	2.677				2.672
0.80	2.851	2.881	2.881	2.889				2.883
1.00	3.241	3.271	3.264					3.268
1.20	3.572	3.589						3.589
1.40	3.879	3.912						3.912

3. Results

Measurements were made at pressures from 13.43 to 68.16 kPa at 295 K and at E/N values from 0.025 to 1.4 Td. The values of D_T/μ derived using the Huxley relation are shown in Table 2. At a given pressure the lower limit to the value of E/N at which measurements were made was set by the requirement that the potential difference across the diffusion chamber should be greater than 30 V in order to avoid significant errors from contact potential differences. The upper limit to the value of E/N was set by the arbitrary limitation of the potential difference to less than about 700 V.

It is apparent from Table 2 that the values are independent of pressure (with the exception of the values at 13.43 kPa which are discussed in Section 4) to within the experimental scatter (of the order of 0.3%). The best estimate

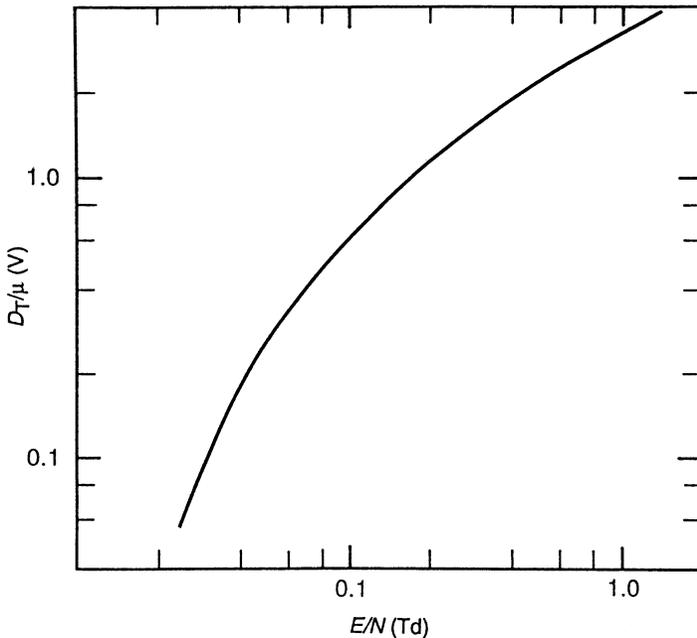


Fig. 3. 'Best estimate' values of D_T/μ as a function of E/N for the 0.467% H_2 -99.533% Kr mixture at 295 K.

values of D_T/μ were in general taken as the mean values (with the 13.43 kPa values excluded) and are plotted as a function of E/N in Fig. 3.

The values of D_T/μ derived using both the Lowke and Huxley relations are plotted as a function of $1/p$ in Fig. 4 for selected values of E/N . The Lowke relation gives values of D_T/μ which are a linear function of $1/p$, the slope depending on the value of E/N (i.e. on the ratio D_L/D_T). The largest difference observed between the D_T/μ values derived from the two relations was 7.2% at 0.04 Td and 33.05 kPa. Note that at 0.1 Td there is no significant difference between the values of D_T/μ obtained using either of the relations. This agreement is due to the fact that the ratio D_L/D_T is close to 0.5 at this E/N value (see Fig. 2 and equation 4). Since the ratio D_L/D_T continues to decrease as E/N increases beyond 0.1 Td, the values of D_T/μ derived from the Lowke relation fall below those obtained from the Huxley relation. For values of $E/N \geq 0.07$ Td the D_T/μ values derived from the Lowke relation by extrapolation to $1/p = 0$ agree with the best estimate values to within 0.5%. The sources of experimental error are listed in Table 3. The total uncertainty in D_T/μ obtained by adding the systematic errors in quadrature, the statistical error arithmetically and rounding to the nearest half per cent is $\leq 1.5\%$ for $E/N \geq 0.07$ Td and $< 2\%$ for $E/N < 0.07$ Td.

4. Discussion

The pressure dependence of the values of D_T/μ derived using the Lowke relation suggests that the model on which this relation is based is not an accurate description of the physical situation. None of the models discussed here take

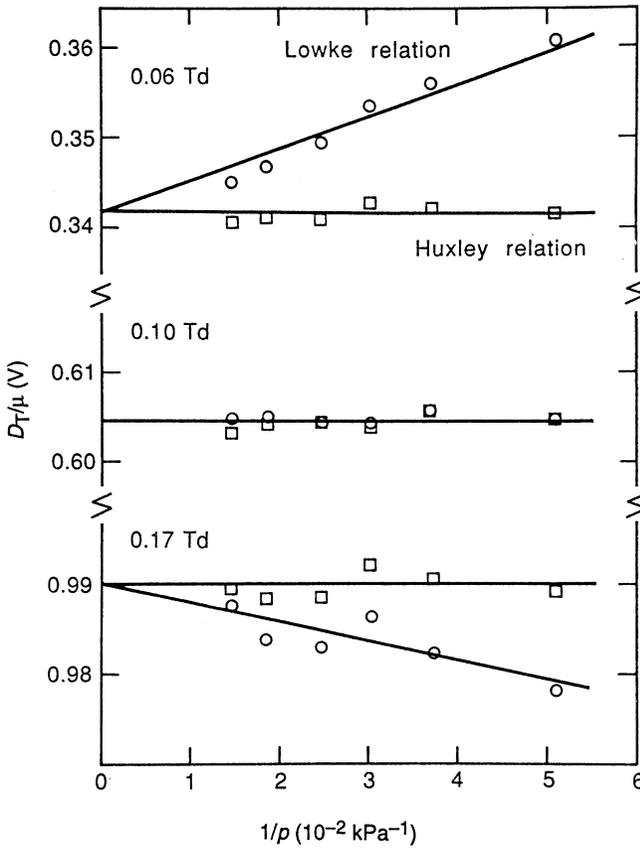


Fig. 4. Values of D_T/μ as a function of $1/p$ for 0.06, 0.10 and 0.17 Td: squares, derived using the Huxley relation; circles, derived using the Lowke relation.

into account non-hydrodynamic effects near the boundaries and it seems likely that the effect of longitudinal diffusion is overestimated. In order to study the influence of boundary conditions further, the dependence of the derived values of D_T/μ on $1/p$ has been calculated for the other ratio relations listed in Table 1 for the case of anisotropic diffusion (Fig. 5). It is apparent that the choice of ratio relation results in significantly different values of D_T/μ , although the value extrapolated to infinite pressure (where boundary effects vanish) is the same. It should be noted that the very small dependence of D_T/μ on $1/p$ for the case in which *no* boundary conditions are imposed (relation 4) is misleading since at other values of E/N the use of relation 4 gives rise to a very significant pressure dependence. The fact that the Huxley relation gives values of D_T/μ which are almost independent of pressure (the values of 13.43 kPa and E/N values ≥ 0.5 Td indicate some pressure dependence) in the presence of non-hydrodynamic effects and anisotropic diffusion is puzzling and a matter for conjecture.

There have been a number of attempts to avoid the assumption of hydrodynamic behaviour (which underlies the diffusion equation) by obtaining solutions of the Boltzmann equation with or without boundary conditions for the diffusion chamber geometry (see e.g. Huxley and Crompton 1974; Lowke *et al.* 1977; England and

Table 3. Sources of error

Source of error	Uncertainty	Est. uncertainty in D_T/μ
Pressure p	0.1%	
Temperature T	0.1%	
Chamber length d	0.1%	
Potential difference V	0.05%	
$E(V, d)/N(p, T)$	0.2%	0.6% (0.03 Td)
		0.2% (0.1 Td)
		0.1% (0.5 Td)
		0.1% (1.0 Td)
Mixture concentration		0.1%
Calculation of D_T/μ from R		
due to uncertainty in b		0.2%
due to uncertainty in h		0.2%
Calculation of extrapolation to $1/p = 0$		1.0% ($E/N < 0.04$ Td)
		0.8% ($0.04 \leq E/N < 0.07$ Td)
		0.5% ($E/N \geq 0.07$ Td)
Total systematic error		1.2% ($E/N < 0.07$ Td)
		0.7% ($E/N \geq 0.07$ Td)
Measurement of current ratio R^A		0.4%
	Total stated uncertainty	2% ($E/N < 0.07$ Td)
		1.5% ($E/N \geq 0.07$ Td)

^A The uncertainty due to the dial resolution varies with R , being 0.4% ($R = 0.2$), 0.08% ($R = 0.5$) and 0.2% ($R = 0.8$). Other sources of uncertainty are the current instability and noise.

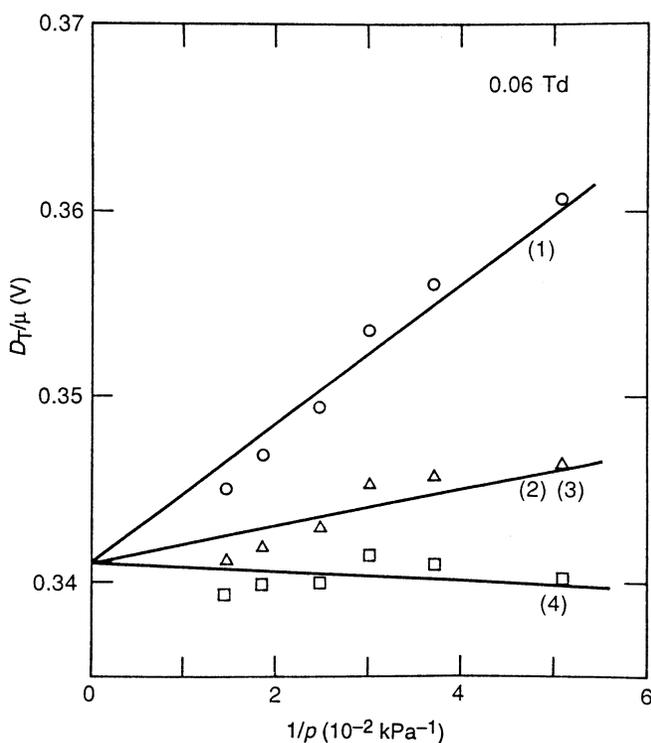


Fig. 5. Values of D_T/μ as a function of $1/p$ for ratio relations in which anisotropic diffusion is assumed, i.e. relations 1-4 of Table 1.

Skullerud, personal communication 1991). However, the treatments available are for model gases or near-thermal swarms and are not generally applicable to realistic cases.

5. Conclusion

The present measurements of D_T/μ for electrons in a 0.467% H_2 –99.53% Kr mixture provide data which are much more sensitive to the momentum transfer cross section for krypton than are drift velocity values and should therefore significantly assist in reducing the uncertainty in the derived cross section. The present values also appear to be the most rigorous test available of the adequacy of the model on which the Lowke relation is based. The failure of this model and the success of the Huxley relation raises fundamental questions concerning the treatment of boundary conditions and non-hydrodynamic effects in the theoretical description of the behaviour of electrons in gases.

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