

Drift Velocity and D_T/μ Ratio for Electrons in a 0.5% Hydrogen–Xenon Mixture at 295 K

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Abstract

The momentum transfer cross section for electrons in xenon, σ_m , has been studied using electron transport coefficient measurements for a dilute hydrogen–xenon mixture (0.47% H₂–99.53% Xe). Drift velocity measurements were made using the Bradbury–Nielsen time-of-flight method at E/N values from 0.12 to 2.50 Td, pressures from 10.3 to 94.0 kPa and a temperature of 295 K (E is the electric field strength and N the gas number density; 1 Td $\equiv 10^{-17}$ V cm²). The ratio D_T/μ (where D_T is the transverse diffusion coefficient and μ the electron mobility) was measured using a Townsend–Huxley diffusion chamber at E/N values from 0.035 to 1.70 Td, pressures from 13.4 to 40.3 kPa, and a temperature of 294 K. Three recently published σ_m values for xenon have been tested and shown to be incompatible with both the present drift velocity and D_T/μ measurements.

1. Introduction

Although the study of the momentum transfer cross section, σ_m , for electrons in xenon has attracted less interest than that for the other inert gases, there is nevertheless a substantial body of literature on this topic. The earliest derivation from electron transport coefficients was by Frost and Phelps (1964) who used the values of Pack *et al.* (1962) for the product of the thermal electron mobility and the gas number density, μN , as a function of temperature (the same data were analysed later by Hayashi 1983). Other derivations have been carried out by Chen (1963) and Hoffman and Skarsgard (1969) using microwave conductivity data, Golovanivsky and Kabilan (1981) using data from cyclotron resonance experiments, Koizumi *et al.* (1986) who used their data for the ratio D_T/μ , Hunter *et al.* (1988) who used their drift velocity (v_{dr}) data and Nakamura (1991) who used his values for v_{dr} and ND_L (where D_L is the longitudinal diffusion coefficient). There appears to be only one previous derivation using data for a xenon mixture. This was made by Schmidt (1986) who used his v_{dr} and ND_L values for a methane–xenon mixture together with his own set of cross sections for methane to derive σ_m for Xe. Measurements of differential cross sections have been used by Register *et al.* (1986) to obtain σ_m for Xe, while *ab initio* calculations of the cross section have been published by Sin Fai Lam (1982) and McEachran and Stauffer (1984, 1987).

It is clear from the study of Hunter *et al.* (1988) that there are large differences between many of these cross sections. The present work was therefore undertaken to enable more rigorous testing of published cross sections, as well as to provide additional data for analysis with the aim of reducing the uncertainty in the derived cross section.

The advantages of using transport coefficient data for mixtures with H_2 , rather than for the pure inert gas, when deriving the momentum transport cross section for those inert gases with a Ramsauer–Townsend minimum, have been discussed previously (see England and Elford 1988; Elford *et al.* 1992) and the same arguments apply to the present case of xenon. Xenon is, however, a more favourable case than krypton when H_2 is the added component, because the ratio of the σ_m for Xe and H_2 (both weighted for their relative abundances) at the Ramsauer–Townsend minimum is significantly greater than for the corresponding krypton case. This results in a smaller uncertainty in a derived xenon σ_m at energies in the vicinity of the Ramsauer–Townsend minimum, due to the uncertainty in the momentum transfer cross section for H_2 .

2. Measurements of Electron Drift Velocities

(a) Apparatus

The electron drift velocities were measured by the Bradbury–Nielsen time-of-flight method and, with one modification, by the apparatus and technique used by England and Elford (1988). This modification was the replacement of the electron source which employed α -particle ionisation by a heated platinum filament (an array of six filaments was installed to avoid frequent opening of the drift tube when a filament burnt out). The cause of the very low electron currents from the ionisation electron source, which necessitated this change, is discussed in the Appendix. The filament source was operated at the lowest possible temperature to reduce thermal gradients and the temperature difference across the drift space was monitored to ensure that significant errors were not introduced.

The xenon used (Matheson Research Grade) was purified by passage over titanium pellets at 800°C , while the hydrogen was purified by passage through a heated silver–palladium thimble. The mixtures were prepared using the mixing volume and technique employed by England and Elford (1988). Two mixtures were made: the first, containing $0.4673\% \text{H}_2$ – $99.5327\% \text{Xe}$, was prepared using calibration points of a quartz spiral pressure gauge (Texas Instruments Ltd) and a xenon filling pressure of 68.158 kPa (the maximum pressure for this gauge). Because the volume of the mixing cylinder and the drift tube were approximately the same, the pressures at which drift velocity measurements could be made were restricted to values less than about 33 kPa . At the conclusion of the measurements, the xenon was recovered from the mixture by freezing out the xenon with liquid nitrogen and pumping off the remaining hydrogen.

In order to take data at higher pressures it was necessary to prepare a second mixture using a second quartz spiral gauge covering a higher range of pressures and to interpolate between calibration points. The entire supply of purified xenon (20 atmospheric litres) available was frozen into the mixing volume using liquid

nitrogen. The mixing volume was then allowed to warm to room temperature and the xenon pressure reduced to the required value of 209.61 kPa. Because of temperature corrections the mixture proportions, 0.4660% H_2 –99.5340% Xe, differed slightly from those of the first mixture and consequently the measured drift velocities at 33.05 kPa also differed slightly (<1%). The values measured at 68.16 and 93.98 kPa at each value of E/N were therefore normalised to the values measured at 33.05 kPa using the first mixture.

Corrections were made for deviations from the perfect gas law using the values of the second virial coefficient for xenon listed by Friedman (1957). The largest correction to E/N due to this effect was 0.5% at 93.98 kPa and the corrections were only significant (i.e. >0.1%) for values of $E/N \leq 0.25$ Td.

The values were independent of the time the mixture had been held in the drift tube (up to 30 days at 93.98 kPa) to within the experimental scatter of $\pm 0.1\%$, indicating that there was no significant contamination due to outgassing.

As in the case of the measurements of electron drift velocities in hydrogen–krypton mixtures made with this drift tube (England and Elford 1988), the first-order current maxima in the arrival-time spectra observed in the present measurements were found to be slightly distorted under conditions where the diffusion broadening of the pulse at the second shutter was small compared with the width of the input pulse. This effect, which makes the first-order maximum unusable for accurate measurements, is due to the severely distorted transmission characteristics of the shutter grids. A typical plot of the current transmitted by a shutter grid as a function of the potential between adjacent wires is shown in Fig. 1. These curves, commonly known as ‘butterfly curves’, show that maximum transmission occurs not when the potential difference is zero, but at some significant value (approximately 6 V for the case shown in Fig. 1). The cause of the anomalous transmission characteristics is not known but it has been speculated that they are associated with a rapidly changing momentum transfer cross section (Robertson 1972).

The distortion of higher order current maxima due to this effect is negligible due to the effect of diffusion, which removes the distortion of the input pulse by the time the pulse reaches the second shutter. Diffusion broadening of the input pulse becomes relatively more significant with increasing order of the current maximum since the width of the input pulse is approximately inversely proportional to the order of the current maximum.

The upper limit to the range of E/N values was arbitrarily set by restricting the potential across the electrode structure to less than about 1 kV. The lower limit to E/N was determined by the loss of resolution in the arrival-time spectrum due to the combination of the transmission characteristics of the grids (the input pulses are relatively broad) and the effects of diffusion.

(b) Results

The present measurements of the drift velocity of electrons in a 0.4673% H_2 –99.5327% Xe mixture at 295 K are shown in Table 1. Corrections for the effects of diffusion and end effects, which give rise to a linear dependence of v_{dr} on $1/p$

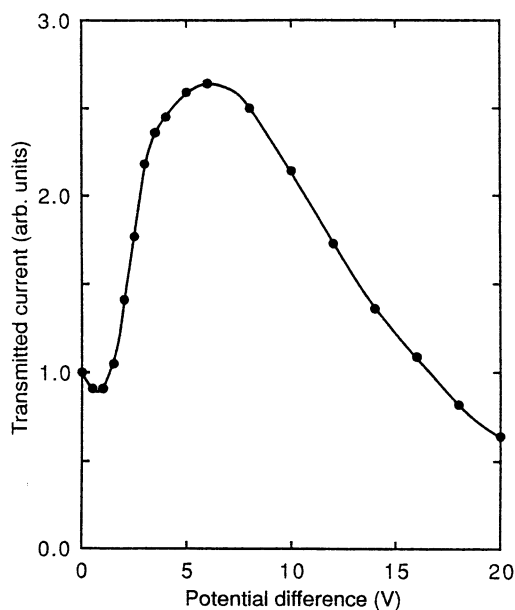


Fig. 1. Typical variation of the current transmitted by a Bradbury-Nielsen grid as a function of the potential difference between adjacent wires (0.5 Td, 13.43 kPa). The current has been normalised to that for zero potential difference. Note that maximum transmission occurs at approximately 6 V.

Table 1. Drift velocities (10^5 cm s^{-1}) of electrons in a 0.4673% H_2 -99.5327% Xe mixture at 295 K

E/N (Td)	10.33	13.43	19.62	p (kPa)			93.98	Best estimate
				26.85	33.05	68.16		
0.12				2.846	2.842	2.826	2.821	2.812
0.14				3.460	3.448	3.419	3.415	3.395
0.17			4.028	4.013	4.009	4.004	4.002	3.994
0.20			4.299	4.287	4.279	4.272	4.270	4.261
0.25		4.483	4.471	4.464	4.460	4.453	4.448	4.443
0.30	4.538	4.526	4.515	4.508	4.504	4.497	4.493	4.489
0.35	4.535	4.522	4.512	4.504	4.502	4.495		4.487
0.40	4.507	4.499	4.489	4.484	4.481	4.474		4.469
0.50	4.451	4.442	4.431	4.428	4.426			4.413
0.60	4.393	4.387	4.378	4.374	4.368			4.360
0.70	4.346	4.338	4.327	4.323	4.320			4.309
0.80	4.297	4.291	4.284	4.280	4.277			4.270
1.00	4.223	4.219	4.209	4.206				4.196
1.20	4.158	4.154	4.147					4.135
1.40	4.102	4.097						4.080
1.70	4.025	4.022						4.012
2.00	3.954							3.941
2.50	3.856							3.844

(where p is the gas pressure), were made in the manner described by England and Elford (1988). The 'best estimate' values shown in Table 1 were obtained by extrapolation to infinite pressure. The uncertainty in the best estimate values

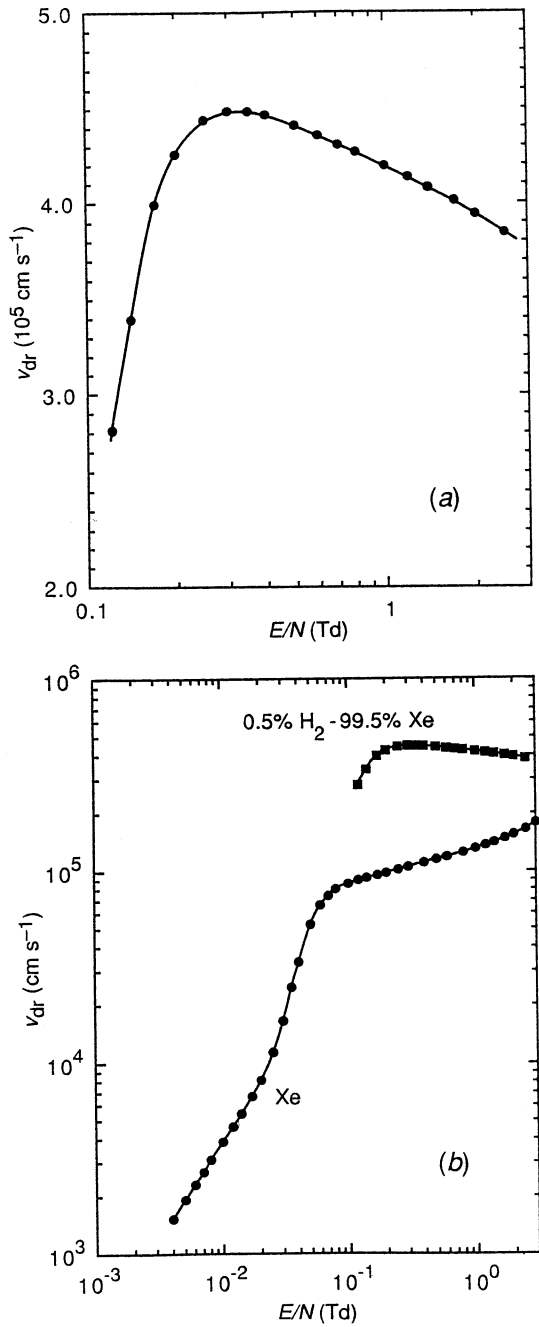


Fig. 2. (a) Best estimate values of the drift velocities of electrons in a 0.4673% H_2 -99.5327% Xe mixture at 295 K. (b) Comparison of the present values of the drift velocity for electrons in a hydrogen-xenon mixture with those for pure xenon of Hunter *et al.* (1988).

was assessed in the same manner as that used by England and Elford (1988) and is considered to be $\leq \pm 0.7\%$. The best estimate values are plotted in Fig. 2*a* and compared with the values of Hunter *et al.* (1988) for pure xenon in Fig. 2*b*.

3. Measurements of the Ratio D_T/μ

(a) Apparatus

Values of D_T/μ for electrons in a mixture of 0.4660% H_2 –99.5340% Xe were measured using the Townsend–Huxley diffusion chamber method and the variable-chamber-length apparatus described in detail by Huxley and Crompton (1974). The chamber length in the present measurements was 10.000 cm and the inner collector had a radius of 0.9975 cm (measured to the centre of the gap between adjacent electrode segments). The electron source was a heated platinum filament, particular care being taken to ensure that the measurements were not significantly affected by the heat dissipated.

Before commencing measurements in the mixture, the operation of the apparatus was checked by a series of measurements in hydrogen at 1.343 kPa. The values were found to agree to within the experimental scatter ($< \pm 0.3\%$) with similar check measurements made in 1990 (Elford *et al.* 1992).

Values of D_T/μ were derived from the measured current ratio using the Huxley empirical relation (with an appropriate iterative procedure to account for electron current which arrived at the collector outside the radius of the outer

Table 2. Values of D_T/μ for electrons in a 0.4660% H_2 –99.5340% Xe mixture at 294 K

E/N (Td)	13.43	19.62	p (kPa)			Mean
			26.85	33.05	40.28	
0.035					0.0343	0.0343
0.04				0.0399	0.0401	0.0400
0.05			0.0627	0.0624	0.0633	0.0628
0.06		0.1038	0.1049	0.1040	0.1060	0.1047
0.07		0.1583	0.1596	0.1593	0.1613	0.1596
0.08		0.2165	0.2185	0.2175	0.2195	0.2180
0.10		0.3262	0.3271	0.3260	0.3284	0.3269
0.12		0.426	0.427	0.424	0.427	0.426
0.14		0.516	0.517	0.515	0.517	0.516
0.17		0.646	0.647	0.645	0.646	0.646
0.20	0.771	0.769	0.768	0.767	0.768	0.768
0.25	0.960	0.959	0.959	0.958	0.958	0.959
0.30	1.141	1.140	1.139	1.138	1.137	1.139
0.35	1.308	1.311	1.310	1.308	1.307	1.309
0.40	1.472	1.471	1.470	1.469	1.468	1.470
0.50	1.774	1.771	1.771	1.765	1.769	1.770
0.60	2.033	2.041	2.041	2.036	2.042	2.039
0.70	2.295	2.293	2.290	2.287		2.291
0.80	2.530	2.532	2.527	2.522		2.528
1.00	2.945	2.946	2.951			2.947
1.20	3.312	3.320				3.316
1.40	3.639	3.656				3.648
1.70	4.091					4.091

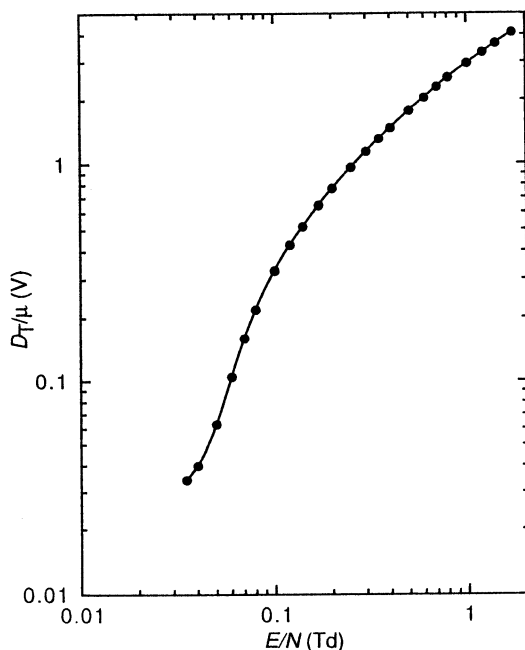


Fig. 3. Best estimate values of D_T/μ for electrons in a mixture of 0.4660% H_2 –99.5340% Xe at 294 K as a function of E/N .

collector, 4.25 cm). No dependence on pressure was observed to within the statistical scatter. The validity of the ratio formulae for the Townsend–Huxley method has been discussed in detail by Elford *et al.* (1992) for the case of a 0.4673% H_2 –99.5327% Kr mixture. Elford *et al.* showed that at sufficiently high pressures (i.e. where the boundary conditions have a negligible effect) the Lowke relation gives the same values as those given by the Huxley relation. This value may therefore be taken to be the correct value to be used in comparisons with values calculated from transport theory. The fact that the Huxley relation gives D_T/μ values which are independent of pressure indicates that this relation gives reliable values of D_T/μ at all pressures.

Corrections for deviations from perfect gas behaviour were applied to all measurements using the same procedure as used for the drift velocity measurements (Section 2a). The maximum correction to the pressure was 0.22% at 40.28 kPa.

O'Malley (1992) has shown that multiple scattering, in which electrons interact with more than one scattering centre, can cause a significant increase in the measured drift velocity and D_T/μ values at sufficiently high pressures and low E/N values. The effect increases with increasing pressure and decreases with increasing E/N . Since there is no evidence in the present data for D_T/μ of a significant dependence on pressure at E/N values greater than 0.04 Td, it is considered that the effect of multiple scattering is negligible in the present measurements of both v_{dr} and D_T/μ .

(b) Results

The values of D_T/μ for electrons in a mixture of 0.4660% H_2 -99.5340% Xe are shown in Table 2 as a function of E/N and pressure. The best estimate values (shown in Table 2 and plotted in Fig. 3) were obtained by averaging. The total uncertainty was assessed by the method used by Elford *et al.* (1992) and found to be $<\pm 2\%$.

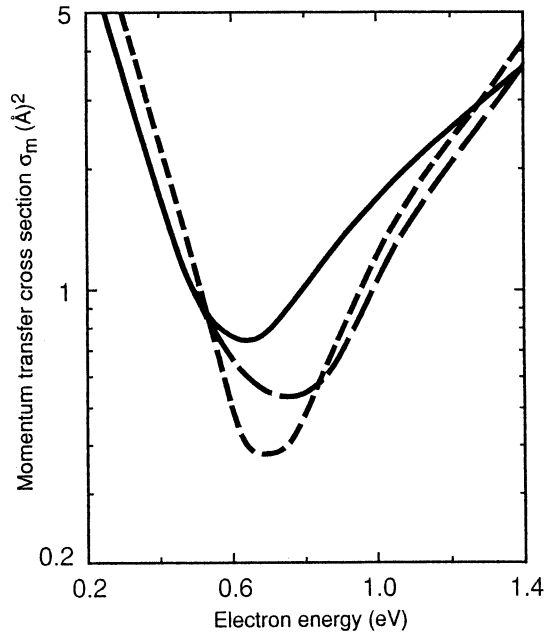


Fig. 4. Momentum transfer cross section for xenon: long-dash curve, McEachran and Stauffer (1987); solid curve, Hunter *et al.* (1988); and short-dash curve, Nakamura (1991).

4. Discussion

Three recently published momentum transfer cross sections for xenon have been studied to determine their compatibility with the present data. The cross sections chosen were those of McEachran and Stauffer (1987), Hunter *et al.* (1988) and Nakamura (1991) (Fig. 4). The cross section of Suzuki *et al.* (1992) obtained using the inversion procedure of Taniguchi *et al.* (1987) was not included as the fit obtained to the drift velocity data used was relatively poor (to within approximately 4%). The hydrogen cross sections used were those of England *et al.* (1988) and all calculations were performed using the multi-term transport code of Ness and Robson (1986). This code gave the same values of the drift velocities as the two-term transport code over the whole experimental E/N range, but the D_T/μ values were significantly higher (i.e. $> 0.1\%$) for E/N values > 0.35 Td. The difference increased with E/N and reached 2.4% at the highest E/N value used (1.7 Td).

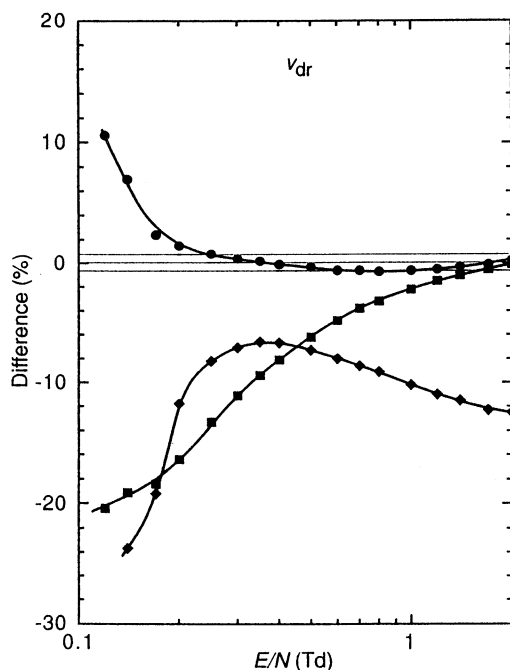


Fig. 5. Difference (%) as a function of E/N , between the calculated and present experimental values of v_{dr} . The calculated values were obtained using the set of cross sections for hydrogen of England *et al.* (1988) and the σ_m for xenon of McEachran and Stauffer (1987) (diamonds), Hunter *et al.* (1988) (squares) and Nakamura (1991) (circles). The experimental uncertainty limits ($\pm 0.7\%$) are as indicated. The difference is defined as

$$\text{difference (\%)} = [v_{dr}(\text{calc}) - v_{dr}(\text{expt})] / v_{dr}(\text{expt}) \times 100.$$

The differences between the calculated and experimental values are shown in Figs 5 and 6. In all cases the difference curves lie outside the error limits, indicated by the horizontal lines about zero, over part or whole of the E/N range for which measurements were made, the differences being up to 20% for the drift velocities and 40% for the D_T/μ values. [Note that in the case of the D_T/μ values all the difference curves must become zero at $E/N = 0$, where the swarm is in thermal equilibrium and $D_T/\mu = kTe$ (k is Boltzmann's constant and e is the electronic charge).] The importance of D_T/μ is exemplified by the difference curve (solid circles) for the cross section of Nakamura. Although his cross section gives a good fit to the present drift velocity data for values of $0.2 < E/N < 2$ Td, the differences with the present D_T/μ values are between 5 and 10 times the experimental uncertainty for most of the E/N range.

The differences shown in Figs 5 and 6 depend, of course, on the reliability of the cross-section set for hydrogen used in the calculations, and in particular on that for vibrational excitation. There has been a long-standing debate concerning this cross section (Crompton and Morrison 1993, and references therein) due to apparently irreconcilable differences between the swarm-derived cross section and those obtained theoretically and by beam techniques. In order to indicate the significance of this cross section in the present study, the theoretical vibrational excitation cross section of Morrison and Trail (1993) was used, in conjunction with

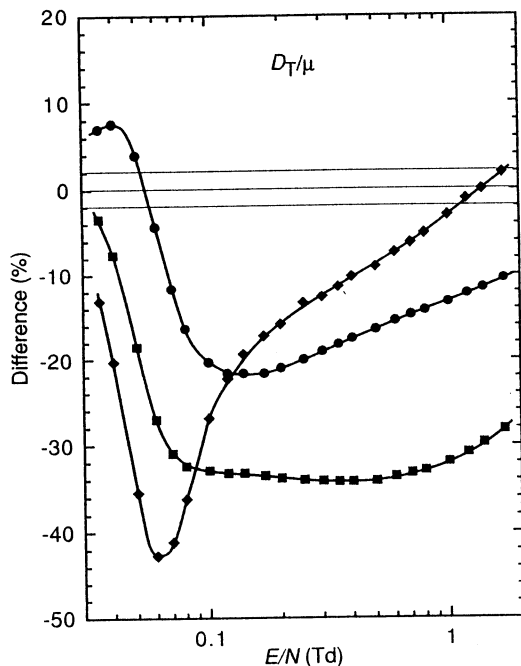


Fig. 6. Difference (%), as a function of E/N , between the calculated and present experimental values of D_T/μ . The calculated values were obtained using the set of cross sections for hydrogen of England *et al.* (1988) and σ_m for xenon of McEachran and Stauffer (1987) (diamonds), Hunter *et al.* (1988) (squares) and Nakamura (1991) (circles). The experimental uncertainty limits ($\pm 2\%$) are as indicated. The difference is defined as

$$\text{difference (\%)} = [D_T/\mu(\text{calc}) - D_T/\mu(\text{expt})]/D_T/\mu(\text{expt}) \times 100.$$

the xenon cross section of Nakamura, to predict differences with the experimental v_{dr} and D_T/μ values for the mixture. It was found that the v_{dr} differences were increased over the E/N range 0.2–2 Td by from 9 to 18%, so that there was now severe disagreement with the experimental data over the whole E/N range. The D_T/μ differences on the other hand changed by less than 1% for $E/N < 0.05$ Td, but decreased by up to 12% at higher E/N values, making the differences with our data even greater. Thus the conclusion that the three momentum transfer cross sections tested are not compatible with the present data is independent of whether the vibrational cross sections of England *et al.* or Morrison and Trail are assumed. It should be noted that the vibrational cross section of Morrison and Trail does not predict the large number of sets of transport coefficient data available for hydrogen and hydrogen mixtures, in contrast to the swarm-derived cross section of England *et al.*

We have not attempted to derive a new momentum transfer cross section for xenon from the present mixture data because of recent work by Schmidt and his collaborators (Schmidt *et al.* 1994). They have pointed out that since the transport coefficients for the mixture are very sensitive to the set of hydrogen cross sections used (an example of which has been given above) a derived xenon cross section would also be sensitive to the hydrogen cross sections assumed. It

is therefore necessary to have available the best possible set of hydrogen cross sections before commencing a derivation of the xenon cross section. Schmidt (personal communication) and his collaborators are currently analysing new, and in some cases, more accurate, transport coefficient data for hydrogen together with data for xenon–hydrogen mixtures (including the data presented in this paper) to derive such a set of cross sections for hydrogen (and in particular the cross section for vibrational excitation), as well as the momentum transfer cross section for xenon.

Acknowledgments

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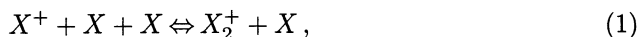
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Appendix

The source using α -particle ionisation is the electron source of choice, as there is no heat input giving rise to thermal gradients across the drift space, it has been found to produce adequate and highly stable electron currents for a wide range of gases and pressures and, of course, it is not subject to burn-out as are filament sources. However, when an α -particle ionisation source was used in pure xenon or dilute H_2 -xenon mixtures, it was found to produce only a small negative ion current. [The current was analysed using the 'inverse Loeb filter method' described by Elford (1987).] It was found possible to obtain electron current from the source only by increasing the H_2 concentration to a level much larger than that desired for the drift velocity measurements. A similar effect occurs for pure krypton and H_2 -krypton mixtures, although in this case the proportion of H_2 required for adequate electron currents was relatively small. It was therefore possible to employ an α -particle ionisation source in the measurements of drift velocities in the hydrogen-krypton mixtures used by England and Elford (1988).

The behaviour of the radioactive source in the inert gases and hydrogen-inert gas mixtures may be explained by the formation of the molecular ion species X_2^+ in three-body reactions



and by the Hornbeck-Molnar reaction

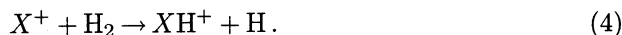


followed by dissociative recombination



where X is Kr or Xe. It is believed that reaction (3) is sufficiently fast that only a negligible number of electrons survive to be extracted from the source. There is evidence to suggest that the small negative ion current observed was caused by the interaction of metastables with oxide layers on the walls of the source enclosure.

When hydrogen is added, XH^+ ions are formed by the reaction



If the dissociative recombination rate of the XH^+ ions is much smaller than that for X_2^+ ions, then the rate of removal of electrons from the source will be significantly reduced and electrons will survive to be extracted. This explanation for the observed behaviour of the source for Xe, Kr and mixtures with H_2 is consistent with the rates for the reactions (1) to (4).