Electron transport parameters in argon and its momentum transfer cross section

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Abstract. The drift velocity and longitudinal diffusion coefficient of electrons in argon were measured over the range of E/N from 0.25 to 50 Td at room temperature by a double-shutter drift tube with variable drift distance. A momentum transfer cross section for the argon atom which was consistent with both of the present electron swarm parameters was derived over the range of electron energy from 2.5 to 15 eV.

1. Introduction

Argon gas has often been used as an excellent buffer gas in electron swarm studies of gas mixtures. For example, Nolan and Phelps (1965) determined the threshold behaviour of the lowest electronic excitation cross section of the caesium atom from electron drift velocity in Cs–Ar mixtures. Haddad and Milloy (1983) derived low-energy electron collision cross sections for carbon monoxide from the electron drift velocity in CO–Ar mixtures. Haddad (1984) and Nakamura (1987a) also determined the magnitude of the total vibrational cross sections of the nitrogen molecule from the electron drift velocity in N₂–Ar mixtures.

Those electron swarm studies in gas mixtures containing argon were very useful and, therefore, the scheme should have wider application for the purpose of determining inelastic collision cross sections of atoms or molecules (see, for example, Kurachi and Nakamura 1987).

It is well known that the momentum transfer cross section of the argon atom has a very sharp minimum in a lower energy range (the Ramsauer minimum), and that electron collisions with the atom are only elastic up to about 12 eV. Electron swarm parameters in argon are strongly influenced by introducing a small amount of atoms or molecules which have low inelastic thresholds, since the presence of the Ramsauer minimum in argon 'amplifies' (Nolan and Phelps 1965) the effect of low-energy electrons produced by inelastic collisions. Argon is the majority gas in the mixtures

and, therefore, an accurate set of collision cross sections of the atom covering not only the Ramsauer minimum but the whole energy range must be indispensable.

Cross sections of the argon atom, both elastic and inelastic, have been reported by many authors using a variety of techniques. Frost and Phelps (1964) determined a momentum transfer cross section over the energy range up to about 30 eV. Milloy et al (1977) and Haddad and O'Malley (1982) have also derived this over the energy range 0-4 eV, from previously determined electron swarm parameters. The present argument is especially focused upon the shape of the Ramsauer minimum and measurements of electron swarm parameters are mainly confined to low E/N, the ratio of the reduced electric field to the gas number density N. Recent theoretical calculations also gave elastic and momentum transfer cross sections. McEachran and Stauffer (1983) employed a polarisedorbital approximation and Fon et al (1983) and Bell et al (1984) employed an R-matrix method. Still there were substantial disagreements among these published values about the peak and the minimum of the momentum transfer cross section.

In the present paper the experimental results for the drift velocity and the longitudinal diffusion coefficients of electrons in argon are shown over the range of E/N from 0.25 to 50 Td. A momentum transfer cross section for the argon atom, which is consistent with both of the present swarm parameters, is also derived over the range of electron energy from 0.25 to 15 eV, from a Boltzmann equation analysis.

2. Experimental

The double-shutter drift tube which had been used in measurements of electron swarm parameters in nitrogen and carbon monoxide (Nakamura 1987b) was slightly modified and used in the present measurement.

The grid between the filament and the first shutter was removed and the filament was placed closer to the first shutter in order to reduce loss of electrons. The collector circuit was also changed so that the electron meter was grounded. By those improvements, the intensity and stability of the collector current were substantially improved and measurement in a wider range of E/N was realised.

The chart recorder was replaced by a microcomputer, and the time-of-flight (TOF) spectra were stored on magnetic discs. The computer was also used in the evaluation of electron swarm parameters from the measured TOF spectra.

The behaviour of electron swarms in argon gas is extremely sensitive to the presence of impurities, atoms or molecules, which have inelastic processes with lowenergy thresholds. Several measures were taken in the present experiment in order to minimise impurities in the gas. The gas used in the present experiment was of the highest purity available and its purity was 99.9999%. The gas handling system, including the pressure regulator, joints and gaskets, was made of stainless steel. The drift tube was equipped with a non-evaporable getter which has excellent sorption characteristics for the main impurity gases but does not absorb rare gases. The getter was usually activated in vacuum at 973 K for 3 min by passing current through the embedded heater. The effect of the getter was examined by measuring the rise in pressure after the drift tube was sealed off. The getter actually reduced the rate of pressure rise by two orders of magnitude and the pressure in the drift tube was still of the order of 10^{-8} kPa after about 20 hours. About an hour after activation, argon gas was introduced into the drift tube through a cold trap cooled by dry ice. The purity of argon gas used in the drift tube, therefore, can be maintained or even be improved during measurement.

The gas pressure was measured by a diaphragm manometer with digital readout. In the present experiment all measurements were carried out at room temperature.

The procedure to derive the swarm parameters from TOF spectra measured at several drift distances was the same as described in a previous paper (Nakamura 1987b), and it was very effective in the removal or reduction of the so-called end effects, such as the energy relaxation of electrons, the irregularity of the electric field near the shutters, the diffusion effect, and the width of the shutter pulse.

Possible sources of error in the present measurement were roughly analysed and maximum possible errors for the drift velocity and the longitudinal diffusion coefficient were estimated to be about 2% and 10%, respectively.

3. Summary of analysis

The theory used to analyse the drift velocity and the longitudinal diffusion coefficient of electrons was based on the works of Parker and Lowke (1969) and Lowke and Parker (1969). The equations to be solved were equations (2), (3) and (4) in the latter paper. These equations were solved successively by a backward integration procedure using the Runge–Kutta method.

The computer program was tested by computing the examples appearing in both papers, and it was confirmed to be very satisfactory in every respect. The present analysis assumed the ionisation process as one of ordinary electronic excitations and the effect of secondary electrons generated in the swarm was not included in a rigorous manner. The effect, however, was evaluated to be relatively unimportant even in the highest range of the present E/N and the electron swarm parameters derived from the present analysis should be practically valid.

4. Results and discussions

4.1. Electron swarm parameters

Measurements were carried out over the E/N range from 0.2 to 50 Td, and the gas number density N from 1×10^{17} to 7×10^{18} cm⁻³, depending on E/N.

The experimental drift velocity is shown in figure 1. The full circles show mean values of the drift velocities measured at several gas densities. The standard deviation of the measured values was very small and was typically less than 1%. The present result was compared with other results (Wagner et al 1967, Robertson 1977, Christophorou et al 1979, Kücükarpaci and Lucas 1981). The agreement with the result of Robertson (E/N < 1 Td) was especially good and the maximum deviation was about 1%. On the other hand, the results of the others, except for Christophorou et al (1979), were about 10% higher than the present result over all the present E/N. Although they also gave a higher value at low E/N (<2 Td), Christophorou et al (1979) gave a low drift velocity in a medium E/N range (5–8 Td), where the electron drift velocity shows a break against E/N. The full curve in figure 1 shows the result obtained by using the present set of cross sections, which will be explained in the next section.

The result for the longitudinal diffusion coefficient times the gas number density $(ND_{\rm L})$ is shown in figure 2. The full circles again show the result of the present measurement (mean values), and the full curve shows again the result of the present calculation. The experimental longitudinal diffusion coefficient suffered from a larger scatter but the value of $ND_{\rm L}$ showed no apparent pressure dependence. $ND_{\rm L}$ decreases slowly with increasing E/N and it sharply increases at a value of E/N where the drift velocity shows a break against E/N. The parameter was fairly constant at E/N higher

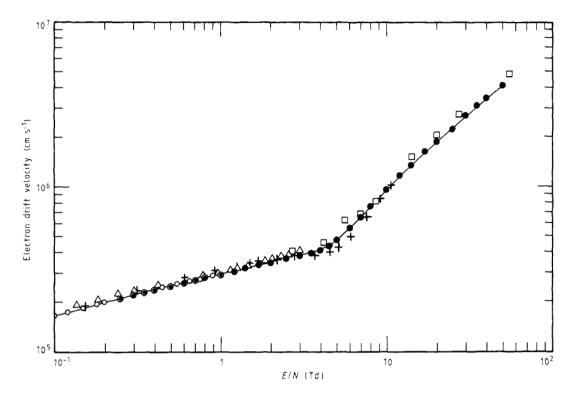


Figure 1. The electron drift velocity *W* as a function of *E/N* in argon. The full curve shows the drift velocity calculated with the present set of collision cross sections. Symbols: ●, present; △, Wagner *et al* (1967); ○, Robertson (1977); □, Kücükarpaci and Lucas (1981); +, Christophorou *et al* (1979).

than 10 Td. There have been a very limited number of measurements on this coefficient, and the two known to the authors (Wagner et al 1967, Kücükarpaci and Lucas 1981) are shown in the figure for comparison. The result of Wagner et al (1967) was overall 11-12% higher than the present result, and the result of Kücükarpaci et al (1981) was also 11-12% higher at an E/N > 8 Td. The present results for the drift velocity and the longitudinal diffusion coefficient are tabulated in table 1.

4.2. Derived momentum transfer cross section

A set of cross sections for the argon atom was derived

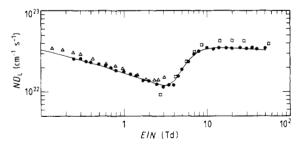


Figure 2. The value of the longitudinal diffusion coefficient times the gas number density, ND_L , as a function of E/N in argon. The full curve again shows the present calculation. Symbols: \bullet , present; \triangle , Wagner *et al* (1967); \square , Kücükarpaci and Lucas (1981).

in order that it gave electron swarm parameters consistent with the results of the present measurement. As will be shown later, the drift velocity and the longitudinal diffusion coefficient in the present E/N range were far more sensitive to the momentum transfer cross section than to inelastic cross sections and, therefore, the present investigation was focused on the former over the energy range 2.5-15 eV. The momentum transfer cross section determined by Milloy et al (1977) was used in the analysis for the range of electron energy 0-2.5 eV. Their cross section had been derived from careful analysis of the measurements of the characteristic energy (Milloy and Crompton 1977) and the electron drift velocity (Robertson 1977). An analytical momentum transfer cross section given by Fon et al (1983) was used for electron energies of over 15 eV. The momentum transfer cross section in either extreme energy range, however, had only a very small effect on the electron swarm in the present E/N. The electronic excitation cross sections derived by Ferreira and Loureiro (1983) were reasonably consistent with the established ionisation coefficient measured by Kruithof (1940) and were included in the present set of cross sections. The ionisation cross section of Rapp and Englander-Golden (1965) was also included in the present set.

The derived momentum transfer cross section is tabulated in table 2 in the energy range 2.5–15 eV and is also shown in figure 3 by the full curve. The two recent analytical momentum cross sections (Fon et al.

E/N (Td)	W (10 ⁵ cm s ⁻¹)	$ND_{\rm L}$ (10 ²² cm ⁻¹ s ⁻¹)	<i>E/N</i> (Td)	W (10 ⁵ cm s ⁻¹)	$ND_{\rm L}$ (10 ²² cm ⁻¹ s ⁻¹)	
0.25	2.13	2.58	4	4.12	1.21	
0.3	2.20	2.62	4.5	4.38	1.57	
0.35	2.29	2.39	5	4.75	1.88	
0.4	2.36	2.35	6	5.64	2.36	
0.5	2.49	2.22	7	6.54	2.96	
0.6	2.60	2.13	8	7.61	3.18	
0.7	2.71	1.99	10	9.56	3.52	
0.8	2.80	1.85	12	11.7	3.37	
1	2.93	1.77	14	13.4	3.50	
1.2	3.06	1.67	17	16.3	3.48	
1.4	3.20	1.59	20	18.8	3.45	
1.7	3.35	1.38	25	22.4	3.51	
2	3.44	1.36	30	27.1	3.41	
2.5	3.62	1.24	35	31.1	3.54	
3	3.81	1.15	40	34.4	3.38	
3.5	3.95	1.22	50	41.2	3.48	

Table 1. The drift velocity and the longitudinal diffusion coefficient of electrons in argon at room temperature.

1983, Bell et al 1984) and the result obtained by Frost and Phelps (1964) are also shown in the figure.

In order to show the effect of the momentum transfer cross section, electron swarm parameters were calculated for the two analytical cross sections and were compared with those from the present momentum cross section. Percentage deviations of the two cross sections from the present one are shown in figure 4. The cross section of Fon et al (1983) was overall larger than the present one and the deviation was about 2.5% around the peak of the cross section. The cross section of Bell et al (1984) was larger, at an energy below 5 eV, and smaller, at an energy above 5 eV, than the present one. The deviation at the peak of cross section was about -8%. The large deviation seen below 1 eV corresponded to the Ramsauer minimum and it, however, had little effect on the swarm parameters in the present E/N range. Percentage deviations of electron swarm

Table 2. The momentum transfer cross section.

Energy (eV)	Momentum cross section (10 ⁻¹⁶ cm ²)
2.5	3.4
3	4.2
4	5.7
4 5	7.6
6	9.6
7	11.5
8	13.1
9	14.6
10	15.8
11	16.2
12	16.1
13	15.6
14	15.0
15	14.2

parameters from the present one are shown in figure 5 for the drift velocity (a) and the longitudinal diffusion coefficient (b). Note the difference in the vertical scale between the two figures.

The full circles in figure 5 show the deviation of the experimental swarm parameters from those calculated

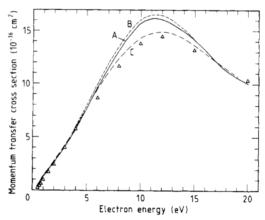


Figure 3. Momentum transfer cross sections for the argon atom. Curves: A, present; B, Fon *et al* (1983); C, Bell *et al* (1984). △, Frost and Phelps (1964).

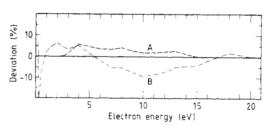
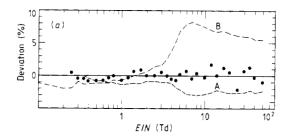


Figure 4. The percentage deviation of the momentum transfer cross sections from the present one. Curves: A, Fon *et al* (1983); B, Bell *et al* (1984).



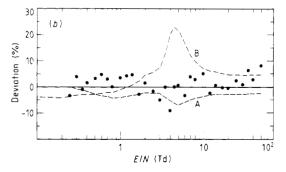


Figure 5. The percentage deviation of electron swarm parameters from the results using the present momentum transfer cross sections. Comparisons are made with analytical cross sections by Fon *et al* (1983, curve A) and by Bell *et al* (1984, curve B). The full circles show the deviation of the experimental results from the present calculation. (a) The drift velocity of electrons. (b) The longitudinal diffusion coefficient of electrons.

using the present momentum transfer cross section. Except for a few points, the deviation was less than $\pm 1\%$ for the drift velocity and less than $\pm 5\%$ for the longitudinal diffusion coefficient. With either of the two momentum transfer cross sections, the deviations of swarm parameters below 3 Td were small but entirely consistent with the behaviour of momentum transfer cross sections. Deviations, however, rapidly increased over the range 3–5 Td, and reached maxima between 4 and 7 Td. A deliberate change of the peak height of the momentum transfer cross section, in fact, revealed that the maxima of the deviations were very sensitive to the change.

Milloy (1975) estimated the onset E/N of inelastic effects on electron swarm parameters, the drift velocity and the transverse diffusion coefficient, in rare gases and it was estimated to be a few Td. The inelastic effects in the present range of E/N, however, turned out to be very small. This was easily confirmed by analysing the deviations of the swarm parameters due to a deliberate change in inelastic collision cross sections. Inelastic collision cross sections were changed by multiplying all inelastic cross sections, including the ionisation cross section, by a constant factor. When only a small fraction (a multiplying factor of 0.2) of inelastic cross sections was included, errors in the drift velocity and the longitudinal diffusion coefficient of electrons were only about 10% at most. When inelastic cross sections were reduced by 40%, errors in the swarm parameters were only comparable with the present experimental scatter. The effects of inelastic collision cross sections on the drift velocity and the longitudinal diffusion coefficient in the present range of E/N, therefore, were small, and it was reasonable to assume that only a momentum transfer cross section was responsible for both of the swarm parameters over the present E/N range. The uniqueness of the present cross section was supported by the fact that the present cross section was consistent with both the drift velocity and the longitudinal diffusion coefficient simultaneously.

The ionisation coefficient, on the other hand, was strongly influenced by the magnitudes of the inelastic cross sections. The 15% increase in electronic excitation cross sections caused about a 50% decrease in the ionisation coefficient in the present range of E/N. As was stated before, the present electronic excitation cross sections were, in total, consistent with the measured ionisation coefficient.

5. Conclusion

The drift velocity and the longitudinal diffusion coefficient of electrons in pure argon were measured over the E/N range from 0.2 to 50 Td.

Inelastic effects on electron swarm parameters were analysed and it was found that the momentum transfer cross section of the argon atom was mainly responsible for the drift velocity and the longitudinal diffusion coefficient of electrons in the present range of E/N. A momentum transfer cross section for the argon atom was derived over the range of electron energy from 2.5 to 15 eV for which the present cross section was consistent with the present experimental swarm parameters.

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