Longitudinal electron diffusion coefficients in gases: Noble gases

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Values of the ratio of the longitudinal diffusion coefficient to mobility D_L/μ for electrons in He, Ar, Kr, and Xe are derived from current waveforms obtained during earlier measurements of electron mobility. The electric field to gas density ratios E/N cover the wide range of 10^{-3} to 20 Td, thereby bridging previous experiments at low E/N to recent experiments at high E/N. Here 1 Td = 1×10^{-21} V m². The corresponding D_L/μ values range from 0.0066 eV for thermal electrons at 77 K to 10 eV. In addition to the well-known peak in D_L/μ for Ar at E/N between 0.01 and 0.1 Td caused by the Ramsauer minimum in the momentum transfer cross section, we find previously unreported low-energy peaks in D_L/μ vs E/N in Kr and Xe and previously unreported pronounced leveling-off in D_L/μ at E/N > 8 Td in Ar, Kr, and Xe. Calculations of transport coefficients using numerical solutions of the Boltzmann equation and cross section sets in the literature give good agreement with experiment from E/N producing thermal electrons up to average energies ≈ 10 eV and E/N up to 100 Td, the upper limit of our calculations. The leveling off of D_L/μ at high E/N is caused by inelastic collisions.

I. INTRODUCTION

Measurements of the ratio of the diffusion coefficient to mobility for electrons in weakly ionized gases by Wagner et al.1 using a time-of-flight technique yielded values as much as a factor of 7 smaller than values obtained using the Townsend method.^{2,3} Parker and Lowke⁴ have shown that this effect is caused by a spatial dependence of the electron energy distribution function resulting from longitudinal electron density gradients present in time-of-flight experiments. Lowke and Parker⁵ have applied the theory to calculations of the ratio of the longitudinal diffusion coefficient D_L to the mobility μ for electrons in a number of gases over a wide range of the ratio of electric field to gas density E/N. These authors find good agreement between theory and experiment over the limited range of E/N covered by the data of Wagner et al. Since these early experiments there have been measurements of D_L/μ at much higher E/N values.⁶⁻⁸

The purpose of this article is to present data extending the range of E/N over which theory and experiment can be compared, particularly for the heavier rare gases. Inclusion of inelastic cross sections allows extension of the calculations and improves the agreement with experiment from below 0.001 Td to 100 Td. Above an E/N of about 10 Td the values of D_L/μ , as well as of D_T/μ , are found to saturate.

The data were extracted from electron current waveforms obtained during drift velocity measurements. 9,10 Since the significance of D_L/μ and its variation with E/N were not understood at the time of the measurements, procedures to insure a small scatter of the widths of the current pulses were not observed. We believe that the ex-

tended range of the D_L/μ data and the tests of electron collision cross sections over a much wider range of energies and gases than previously possible justify presentation of the results in spite of their large scatter.

The experimental apparatus and the method of data analysis are described in Sec. II. The results are presented and discussed in Secs. III and IV. Formulas and sources of error are discussed in the Appendix. A table of momentum transfer cross sections is given for He, Ar, Kr, and Xe.

II. METHOD

A. Experimental apparatus

The electron current waveform records obtained in the measurements of electron drift velocity^{9,10} were used to determine D_I/μ . A pulse of UV light from a hydrogen or deuterium lamp illuminated a gold-plated cathode causing the emission of a pulse of electrons by the cathode. The electron pulse drifts through the gas toward the collector under the influence of a uniform electric field. A delayed voltage pulse applied to one of the two shutters or grids controls the passage of electrons to the collector. Depending upon the mode of operation^{9,10} the grid is made more absorbing or transmitting causing a change in collector current proportional to the electron density near the grid. Variation of the delay gives a current waveform exemplified in Fig. 1. The broadening of the waveform results from using rectangular light and grid pulses and from longitudinal diffusion.

The vacuum system was baked out for $\approx 14 \text{ h}$ at 250 °C resulting in an initial background pressure of $< 10^{-8}$ Torr. The rate of rise of 10^{-9} Torr/min limited added impurities to $\ll 0.1$ ppm in a 2 h run. Research grade noble gases used in the experiments contained 11 typically 1 to 25 ppm of some molecular gases, in particular water vapor and hydrocarbons, and 1 to 50 ppm of other noble gases. Hunter et al. 12 attribute deviations from the theoretical calculations of drift velocities near the peak in electron mobility in

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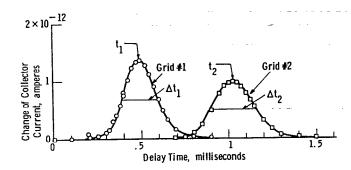


FIG. 1. Example of current waveform showing the time of arrival of the electrons t_1 and t_2 and the full widths at half maximum δt_1 and δt_2 at grid Nos. 1 and 2.

argon and krypton to trace impurities. We performed calculations of drift velocities in argon mixtures containing various amounts of nitrogen or water vapor for 0.003 < E/ N < 0.4 Td, which showed that as much as 20 ppm of nitrogen produced <1% change in drift velocity in this region of E/N. However, calculations with 1 ppm of water vapor caused a 10% change in electron drift velocity for 0.005 < E/N < 0.03 Td. Consequently, we believe that the deviations of drift velocity measurements in the rare gases from theoretical values between different laboratories are caused by trace impurities of a few ppm of water and/or hydrocarbons. Since the vapor pressure of water is negligible at 77 K, perhaps hydrocarbons are responsible for the deviations of drift velocity measurements among different laboratories at 77 K. Ice has a vapor pressure of 5.6×10^{-4} Torr at 195 K giving a possible impurity level of ≈ 1 ppm of water.

Experimental parameters were determined as described previously. Temperatures of the drift tube were controlled by immersion in liquid nitrogen at 77 K or alcohol—dry ice or acetone—dry ice mixture at 195 K and by the air conditioning system at 300 K. Use of an analog $\pm 1\%$ laboratory voltmeter gave an accuracy of $\approx 2\%$ in the E/N values. The estimated accuracy of transit times t_1 and t_2 to the first or second grid was $\approx 1\%$ and the half widths δt_1 and $\delta t_2 \approx 10\%$ resulting in scatter of $\approx 2\%$ in the drift velocity measurements and $\approx 20\%$ in the D_L/μ determinations. In some cases additional scatter of the width measurements resulted from failure to determine the background current of some current waveforms.

B. Analysis of waveforms

The method of analysis of the data is similar to that used by Wagner et al.¹ except that use of the double grid tube allowed us to check the consistency of the data as a function of drift distance. The current density J reaching either grid due to an impulse of electrons leaving the cathode and moving under the action of a uniform electric field with no absorption at the boundaries is given by Kennard¹³ as

$$J = (A/t^{1/2}) \exp[-(L - Wt)^2/4Dt], \qquad (1)$$

where A is a constant, L the distance from the cathode to grid, W the electron drift velocity, D the diffusion coefficient for diffusion in the direction of the field, hereafter designated as D_L , and time t measured from the release of electrons from the cathode. The values of W reported previously 9,10,14 were obtained from the equation

$$W = (L_2 - L_1)/(t_2 - t_1) , \qquad (2)$$

where t_1 and t_2 are the measured transit times, i.e., the times of the first and second minima (or maxima) in the current waveforms, and L_1 and L_2 the distances from the cathode to the first and second grids. End effects in time measurements subtract out to first order when using this difference method.⁹

When Eq. (1) is solved for δt , the full width at half maximum of the current waveform, the leading terms¹⁵ of the resulting expression including absorption at the cathode and at the grid can be written as

$$D_L/\mu = [1/(16 \ln 2)](\delta t_1/t_1)^2 V_1. \tag{3}$$

Here the width δt_1 , the transit time t_1 , and the distance L_1 refer to the first grid and V_1 is the voltage between the cathode and the first grid. A similar expression for the second grid is obtained by using δt_2 , t_2 , and V_2 in Eq. (3). Here we have used the relation between drift velocity and mobility, $W = \mu E$. Note that the determination of D_I/μ requires taking a difference of times and therefore is independent of end effects in t to first order. The waveforms are wider than predicted by Eq. (1) due to the rectangular gate pulses applied to the lamp modulator and grids. With no diffusion, equal light and grid pulse widths produce current waveforms triangular in shape with a full width at half maximum equal to the width of the pulses. In view of limitations in the accuracy represented by the scatter in the data, we have assumed that the resultant instrument function can be approximated by a Gaussian of width δt_i , the instrumental width as defined below. A computer convolution of rectangular gates with a Gaussian electron pulse for ratios of the gate widths to diffusion widths from 0.4 to 2.5 agree with the above approximation to within 2%. The corrected width to be used in Eq. (3) is therefore given by

$$\delta t = (\delta t_m^2 - t_i^2)^{1/2}, \tag{4}$$

where $\delta t_i = \max(\delta t_1, \delta t_g)^{16}$ and δt_m is the measured half width.

An alternate procedure for analyzing the data is to regard δt_i as an unknown and solve Eqs. (3) and (4) for D_L/μ in terms of the two measured widths. When $L_2 = 2L_1$ as in our experiments, this procedure gives

$$D_L/\mu = [2/(16 \ln 2)] V_2(\delta t_{m2}^2 - \delta t_{m_1}^2)/t_2^2$$
 (5)

and

$$\delta t_i = (2\delta t_{m1}^2 - \delta t_{m2}^2)^{1/2} \,. \tag{6}$$

The agreement of δt_i from Eq. (6) with the pulse widths used in the measurements is a measure of the consistency of the values of D_L/μ obtained using Eq. (5). Note that Eq. (5) subtracts out the instrumental width, δt_i

The data presented in Sec. III were selected from the waveform records on the basis of the following criteria:

- (a) The light and grid pulse widths, δt_i , should be ≤ 0.8 of the width of the current waveform, δt_m , or else the sensitivity to diffusion is too small. Since the widths of the light and grid pulses had little effect on the accuracy of the drift velocity measurements, some of the original data was obtained using too large a value of δt_i to resolve D_I/μ .
- (b) The ratio $\delta t_1/t_1$ should be <0.4 to limit errors introduced by boundary conditions, ^{4,16} higher-order corrections¹ to Eq. (3), and the effects of the initial electron energy distribution on the value of D_I/μ .
- (c) Finally, data are shown only when the various values of D_L/μ calculated using Eqs. (3) and (5) agree within 25%, except when $\delta t_1 < t_i$ as noted above. These criteria require consistency between the widths determined with the two grids and limits errors not covered by (a) and (b) above.

The rather large limits of acceptability were necessary in order to preserve a significant number of data points. Errors in drift velocity due to absorbing boundaries, as discussed by Duncan¹⁷ and Lowke, ¹⁷ are the order of $(\delta t/$ t)² when two grids are used.⁶ Recently, Nakamura¹⁸ reported the use of a difference technique to eliminate end effects in nitrogen, carbon monoxide, and other gases. Although a longitudinal gating field is used, the operation of his drift tube and ours are similar. The effects of pulse amplitude is discussed in the Appendix.

C. Boltzmann equation solution

The theoretical D_L/μ values were calculated using the method described by Parker and Lowke⁴ and Lowke and Parker.⁵ They developed a solution for the Boltzmann equation for electrons in a constant electric field based on a two-term spherical harmonic expansion of the electron energy distribution in velocity space and a Fourier expansion in coordinate space and time. The theoretical W and D_T/μ values were calculated using an extension of the backward prolongation technique of Frost and Phelps. 19 These theories form the basis for the computer program²⁰ used to obtain the results presented here. The cross sections used are discussed with each gas. The theoretical results were checked in argon by performing Monte Carlo (MC) calculations for 10 < E/N < 700 Td. The two calculations agree satisfactorily for E/N < 200 Td. For E/N>200 Td the MC results start to diverge from our calculations. Since the transport coefficients predicted by the two techniques are in agreement at the highest E/N value (100 Td) reported here, we expect the two-term solution to be accurate for atomic gases at lower E/N values even though the electrons produced by ionization are neglected in our version of the code. Segur and Bordage²¹ reached similar conclusions in argon for $E/N \le 1000$ Td by comparing the results of a multiterm method and the two-term method. Also, Date, Kondo, and Tagashira²² reached similar conclusions in krypton for $E/N \le 150$ Td by comparing results of an MC calculation and a two-term method.

III. RESULTS

The experimental values of D_L/μ^{23} and of drift velocity for He, Ar, Kr, and Xe are shown by points in Figs. 2 through 5, respectively. The smooth curves show the results of the Boltzmann calculations. The current waveforms for neon were not available at the time of this work; therefore, no experimental values of D_L/μ in neon were obtained.

A. Helium

Figure 2 shows the measured and calculated values of W and D_L/μ . The points show W and D_L/μ calculated from the collector current waveforms used to obtain the drift velocity data¹⁰ for electrons in helium at 77 and 300 K. Pressures ranged from 36 to 310 Torr at 77 K and from 9.8 to 693 Torr at 300 K. Drift, i.e., cathode to grid, distances were 2.5 and 5.1 cm, 3.81 and 10.2 cm, or 5.1 and 10.2 cm. Experimental D_L/μ values obtained using Eq. (5) are shown. Insignificant ionization occurred in our helium experiments.

The theoretical values shown by the curves were calculated by the method of Lowke and Parker⁵ using the cross sections for momentum transfer collisions Q_m given by Crompton et al.24 and later improved by Hayashi25 and extrapolated to 700 eV as shown in Table I. The inelastic cross sections are from Lowke et al.,26 Trajmar,27 and Vriens et al.28 At 100 Td the electron energy range used in the calculations was 0-350 eV in order for the electron energy distribution function to decrease by 106 before truncation. 19,20 There is good agreement between the theoretical calculations and experiment except for the $\approx 10\%$ discrepancy between the calculations and the D_L/μ data for 0.3 < E/N < 3 Td. Above 30 Td calculations agree with the data of Anderson, ²⁹ Phelps *et al.*, ¹⁴ and Kucukarparci *et* al.⁶ for 0.4 < E/N < 100 Td. The experimental drift velocity data are so numerous above 0.02 Td that they are not resolved on these plots but demonstrate the agreement with the theoretical calculations.

B. Argon

Figure 3 shows the data and calculated W and D/μ curves in argon at pressures between 182 and 200 Torr at 77 K and between 45.5 and 735 Torr at 300 K for E/N from 0.0006 to 10 Td. The drift distances were 2.5 and 5.1 cm. The theoretical curves were calculated, using the Q_m values given by Frost and Phelps, 30 O'Malley, 31 and Milloy et al.32 (as integrated by Hayashi25); inelastic cross sections of Tachibana; 33 and ionization cross sections of Rapp and Englander-Golden.³⁴ From 0.1 eV down to 0.01 eV the Q_m cross-section curve followed O'Malley's result, 31 below 0.01 eV Q_m was adjusted to give theoretical agreement with the 77 K drift velocity data.9

A relatively broad maxima in D_L/μ vs E/N occurs near E/N = 0.003 Td at 300 K and a narrower maxima occurs near 0.004 Td at 77 K. Our 77 K data and the 93 K data of Robertson and Rees³⁵ agree with calculations in this E/N range to within their scatter. The location and magnitudes of the maxima agree with the theoretical cal-

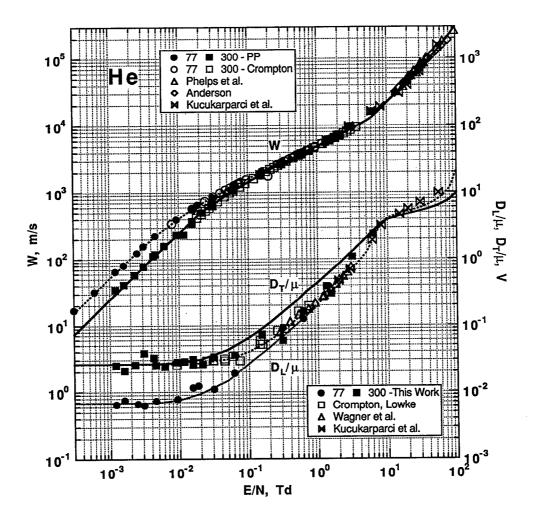


FIG. 2. Experimental and theoretical values of D_L/μ , D_T/μ , and W vs E/N at 77 and 300 K for electrons in He. The solid curves relate to 300 K calculations of W and D_T/μ . The upper dashed curve is the 77 K calculations of drift velocity. The lower dashed curves are 77 and 300 K theoretical calculations of D_L/μ . PP refers to Pack and Phelps (Ref. 9).

culations of Lowke and Parker.⁵ As shown by Parker and Lowke⁴ values of D_L/D_T greater than unity occur when the electron momentum transfer collision frequency decreases with increasing electron energy. Such a variation of the collision frequency is found³⁰ in argon at electron energies just below the Ramsauer minimum. Above 0.01 Td good agreement is obtained with Wagner *et al.*¹ over the range of their data. As noted previously,^{4,5} for 0.01 < E/N < 2 the D_L/μ values are about a factor of 7 below the D_T/μ values as the result of the rapid rise in Q_m with electron energy.

Near E/N=5 Td and the onset of excitation the calculated D_L/μ and D_T/μ rise rapidly. The D_L/μ values then level off for E/N above 10 Td as excitation limits the mean energy of the electrons. Our experimental data extend to 10 Td and, although the scatter is more than desired, fair agreement is obtained with our calculations and with the experiments of Kucukarparci and Lucas⁷ and with Nakamura and Kurachi. This second region of structure near E/N=7 Td has also been calculated by Makabe and Shimoyama. However, our experimental and calculated D_L/μ values are significantly above theirs for E/N>2 Td. The difference in calculated values is caused by differences

in cross sections used. Our calculations with mixtures show that trace impurities have little effect in this region of E/N where the momentum transfer cross section is large and inelastic losses occur.

Our experimental drift velocity data, indicated by circles in Fig. 3, agree with the calculated curve to better than 2% except around the peak in electron mobility at E/N= 0.002 Td where the maximum deviation from theory is 5% at 300 K. For more discussion, see Ref. 12. The calculated theoretical drift velocity curve agrees with the data of Jager and Otto, ³⁷ Brambring, ³⁸ Wagner, ³⁹ Errett, ⁴⁰ and Kucukarparci and Lucas⁷ for $1 < E/N \le 100$ Td, the range of overlap with their data unresolved in the figure. The constant electron mobility is consistent with the near constancy of the electron energy and the slowly varying electron collision frequency for 10 < E/N < 100 Td. Scatter in the data warrants no further adjustment of the momentum transfer cross section above 10 eV. The D_T/μ curve, shown with no experimental points for comparison since that coefficient was not measured in our experiments, agrees well with the data of Townsend and Bailey41 over the range of his measurements (0.3 < E/N < 60 Td) and with Warren

TABLE I. Momentum transfer cross sections.

Helium		Argon		Krypton		Xenon	
Energy (eV)	$Q_m \ (ext{\AA})^2$	Energy (eV)	Q_m $(\mathring{\mathrm{A}})^2$	Energy (eV)	$rac{Q_m}{(\mathring{ m A})^2}$	Energy (eV)	Q_m $(\mathring{A})^2$
0.000	4.95	0.000	10.0	0.000	39.70	0.000	176
0.0025	5.00	0.001	8.35	0.001	39.70	. 0.002	160
0.0036	5.10	0.005	6.61	0.003	35.00	0.005	139
0.010	5.27	0.010	5.60	0.005	30.00	0.007	127
0.032	5.52	0.020	4.15	0.0085	27.0	0.01	116
0.200	6.20	0.030	3.00	0.010	26.20	0.03	61.3
0.600	6.66	0.070	1.13	0.020	21.40	0.05	39.5
1.400	6.98	0.100	0.59	0.040	15.32	0.08	25.6
3.000	6.93	0.150	0.23	0.060	11.68	0.10	20.4
8.000	5.50	0.170	0.16	0.080	9.13	0.16	12.0
14.00	3.60	0.200	0.10	0.100	7.23	0.20	8.4
18.00	2.90	0.250	0.091	0.16	4.00	0.25	5.9
20.00	2.69	0.300	0.15	0.200	2.37	0.30	3.3
25.00	2.00	0.350	0.235	0.25	1.30	0.40	1.6
35.00	1.26	0.400	0.33	0.300	0.86	0.50	0.53
40.00	1.00	0.500	0.51	0.350	0.55	0.60	0.38
50.00	0.70	0.700	0.86	0.400	0.26	0.70	0.38
75.00	0.36	1.000	1.38	0.500	0.10	0.80	0.55
100.0	0.22	1.200	1.66	0.540	0.11	0.90	0.90
150.0	0.12	1.300	1.82	0.600	0.15	1.00	1.15
200.0	0.07	1.700	2.32	0.700	0.13	1.20	1.90
250.0	0.05	2.100	2.85	0.800	0.42	1.50	3.50
300.0	0.036	2.500	3.37	1.00	0.80	2.00	7.50
500.0	0.016	3.000	4.10	1.20	1.30	2.50	11.50
700.0	0.009	3.600	5.00	1.60	2.00	3.00	16.50
	0.000	4.00	6.00	2.00	3.00	4.00	24.50
		5.00	7.60	2.50	4.40	5.00	28.00
		7.00	11.00	3.00	6.00	6.00	26.00
		8.00	14.00	4.00	10.00	7.00	27.00
		12.00	15.20	5.00	14.00	8.00	26.00
		15.00	14.10	6.00	16.00	10.0	20.00
		17.00	12.00	7.00	17.00	12.0	
		20.00	9.50	8.00	16.50	15.0	13.50
		25.00	7.40	10.00	15.50	20.00	9.50
		30.00	6.00	12.00	13.50		7.00
		50.00	3.50	20.00	6.00	30.00 50.00	5.10
		75.00	2.30	50.00	1.55		3.60
		100.0	1.70	30.00	1.33	100.0	2.40

and Parker. 42 At 100 Td the normalized electron energy distribution function at 37.5 eV is $< 10^{-8}$ of that at 0 eV, in contrast to helium. Therefore, calculations neglecting energies >40 eV suffer no loss of accuracy due to the truncation of the distribution function. At 100 Td, 18% of the energy goes into ionization; however, MC calculations verify that the two-term method is good to at least 100 Td. At 10 Td, the highest E/N value used, there is no observable ionization and a negligible number of electrons having energy > 15 eV.

C. Krypton

Figure 4 shows calculated W and D/μ curves and experimental data in krypton as a function of E/N at 195 and 300 K. The pressure ranged from 5.4 to 760 Torr and the drift distances were 5.1 and 10.2 cm. The general behavior of the experimental and theoretical D_I/μ values is similar to that for argon. The momentum transfer cross sections

used in the calculations were from Hayashi²⁵ for 0.0 < energy < 0.3 eV. For 0.3 < energy < 1.2 eV we adopt the deeper minimum given by Koizumi et al.43 We used the results of Hayashi²⁵ for energies > 1.2 eV as shown in Table I. The inelastic cross sections are from Suzuki et al.44 The calculated drift velocity curve agrees with the experimental data of Hunter et al. 12 below 3 Td and that of Nakamura and Naitoh⁴⁵ above 3 Td.

Based on their measurements with as-purchased samples of krypton, Hunter et al. 12 attribute the deviations of our drift velocity data¹⁰ from theirs for 0.01 < E/N < 0.2Td to 1 ppm of trace impurities in our krypton samples. From calculations with argon mixtures one concludes that the impurities are probably water and/or hydrocarbons. We believe the slight maximum in the calculated D_L/μ and D_T/μ near 5 Td is related to the combination of decreasing Q_m with electron energy and the onset of inelastic losses. The D_L/μ curve agrees with Lowke and Parker's⁵ calcula-

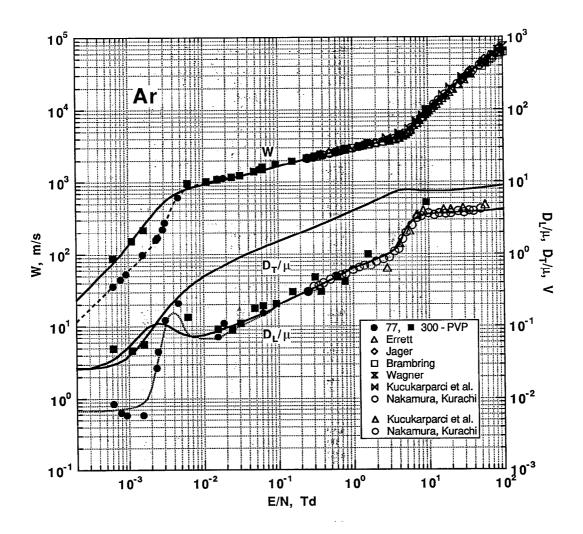


FIG. 3. Experimental and theoretical values of D_L/μ , D_T/μ , and W vs E/N at 77 and 300 K for electrons in Ar. The solid curves are theory at 300 K and the dashed curves are theory at 77 K. PVP refers to Ref. 10. The lower two references refer to D_L/μ measurements, others to W.

tions for 0.4 < E/N < 2 Td as shown by the diamonds in Fig. 4; however, our peak is much broader than theirs. Our calculated D_T/μ curve agrees with the measurements of Koizumi et al. 43 shown in part over the range (0.005 < E/N < 0.8 Td) of their data, and is about 40% above the measurements (not shown) of Al-Amin and Lucas⁴⁶ for 1.4 < E/N < 28 Td. The discrepancy of the D_T/μ calculations with the data of Al-Amin and Lucas⁴⁶ is not understood. At higher E/N our D_L/μ and W curves merge with those of Kucukarparci and Lucas⁷ to within a few percent. At 100 Td, 22% of the electron energy goes into ionization, the distribution function is $< 10^{-6}$ for energies > 32eV. At 20 Td, i.e., near the maximum E/N of our measurements, the energy going into ionization was <0.25% so that no measurable effect of ionization was present in our krypton data. Based on the MC calculation in argon we believe that the two term method is satisfactory in krypton for $E/N \le 100$ Td.

D. Xenon

Figure 5 shows the calculated W and D/μ curves and experimental data in xenon. Drift distances were 5.1 and

10.2 cm, pressures ranged from 12 to 722 Torr at 195 and 300 K. The momentum transfer and total excitation cross sections are from Hayashi⁴⁷ as modified below and Hunter et al. 12 The total ionization cross sections were from Rapp and Englander-Golden.³⁴ The D/μ and drift velocity curves have approximately the same shape as those for argon and krypton, but are displaced toward higher E/N. The drift velocity data of Brooks et al. 48 at 1 < E/N < 4 Td are about 10% higher than ours and about 5% lower than ours at E/N > 8 Td. Hunter et al. 12 claims Brooks's measurements were contaminated with halogen-containing compounds⁴⁹ accounting for his divergence from the curve below 4 Td. Since our xenon drift velocity data agree with Hunter et al. 12 within the scatter of our data around 0.1 Td he concludes that our xenon samples had insignificantly more impurities than his further purified xenon samples. As in krypton, the depth of the Ramsauer minimum was increased until the D_T/μ curve agreed with the data of Koizumi et al. 43 shown in Fig. 5. The momentum transfer cross section given in Table I is consistent with electron transport data and is close to several theoretical calculations shown by Koizumi et al.43 At 100 Td 11% of the

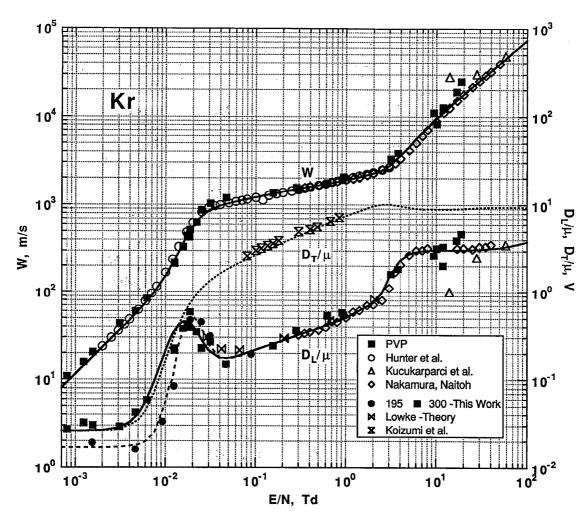


FIG. 4. Experimental and theoretical values of D_L/μ , D_T/μ , and W vs E/N at 195 and 300 K for electrons in Kr. The curves refer to the theoretical calculations. PVP refers to Ref. 10. The lower three references in the box go with the diffusion coefficients.

electron energy goes into ionization; therefore, based on the MC calculation in argon the two-term method should be satisfactory for $E/N \le 100$ Td.

IV. DISCUSSION

The experimental and calculated values of D_I/μ presented in this article proved further confirmation of the theory of longitudinal diffusion given by Parker and Lowke⁴ at low electron energies. In particular, the experimental data for Ar, Kr, and Xe show a maximum in D_L/μ at an E/N predicted by the detailed calculations of Lowke and Parker.⁵ In these gases the experimental data verify the prediction of values of D_L/D_T greater than unity in ranges of E/N and electron energy where the electron collision frequency decreases with increasing electron energy. For helium our experimental values of D_I/μ show a smooth transition from the thermal value of kT/e at low E/N to values of about one-half D_T/μ at moderate E/N. The latter ratio was predicted^{4,5} for gases in which the cross section for momentum transfer collisions varies slowly with electron energy.

We hope that this demonstration of consistency be-

tween the predictions of theory and these relatively scattered measurements of D_L/μ will serve to encourage measurements of D_L/μ with higher precision over a wide range of E/N. An important advantage of measurements of D_L/μ rather than D_T/μ using the Townsend method³ is that the D_L/μ measurements can be made at the same time as the drift velocity measurements with a suitable drift tube. Measurements of other transport coefficients, such as the Townsend ionization coefficient,³ are extremely useful for reducing the ambiguities present in the cross-section determinations. Also, experience has shown that electron transport coefficients must be measured over the widest possible range of E/N if the analyses are to yield reliable cross sections. Impurities having large inelastic losses near the Ramsauer minimum, such as water vapor,⁵⁰ in concentrations as small as 1 ppm cause appreciable errors in measurements of transport coefficients.

Tabulations of D_L/μ and drift velocity data on $5\frac{1}{4}$ " DSDD floppy disk format are available from J.L.P. by request on your disk or from the Physics Auxiliary Publication Service (PAPS), available from the American Institute of Physics.⁵¹

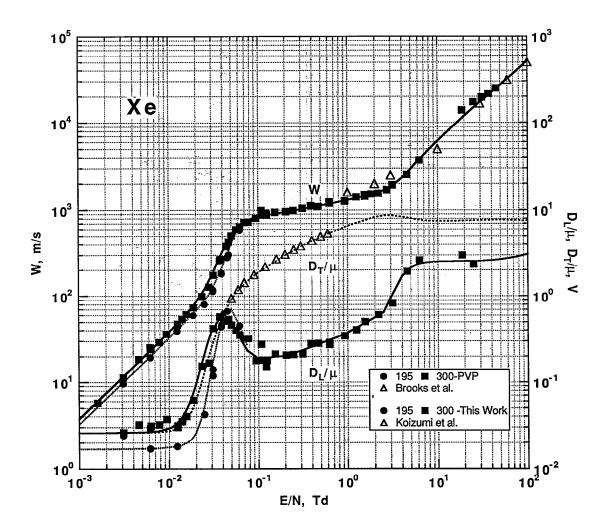


FIG. 5. Experimental and theoretical values of D_L/μ , D_T/μ , and W vs E/N at 195 and 300 K for electrons in Xe. The curves refer to the theoretical calculations. The lower two references in the box go with the diffusion coefficients.

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APPENDIX: SPACE CHARGE EFFECTS

The derivation of the criteria for negligible error in D_L/μ due to space charge effects is based on a calculation of the space charge electric field produced by the undisturbed electron pulse given in Eq. (1). Thus,

$$\frac{dE}{dz} = -\left(\frac{I}{Af}\right) \left[\epsilon_0 (4Dt)^{1/2}\right]^{-1} \exp\left(-\frac{(L-Wt)^2}{4Dt}\right),$$
(A1)

where I/Af is the charge per unit cathode area in a single electron pulse, f is the frequency of the pulses, and ϵ_0 is the

permittivity of free space. Integration from the cathode to the point of half maximum on the leading edge of the exponential factor gives a change in electric field of

$$\delta E = 1.54 I/(\epsilon_0 A f) . \tag{A2}$$

The magnitude of the error due to space charge is calculated by use of Eq. (A2). In a typical example at a very low E/N of 0.0023 Td the current was 2.5×10^{-13} A. The pulse repetition rate was 250 Hz and the area of the swarm was about 0.0011 m². This gives a change in field of 0.1 V/m compared to an applied field of 50.5 V/m or less than 0.2%. In conclusion, space charge effects were negligible in our experiments.

The use of a large instrument width has already been mentioned as a source of error, similarly the derived coefficients were seen to depend upon pulse amplitude. This dependence became small if pulse amplitudes were reduced until grid transmission during the gate was at least 10% of transmission at zero pulse amplitude as determined from a plot of grid characteristics versus bias voltage.

The difference technique represented by Eq. (2) in principle eliminates end effects and removes dependence on pulse widths.

- ¹E. B. Wagner, F. J. Davis, and G. S. Hurst, J. Chem. Phys. 47, 3138 (1967).
- ²L. B. Loeb, Basic Processes of Gaseous Electronics (University of California Press, Berkeley, 1961), Chap. 3.
- ³E. W. McDaniel, Collision Phenomena in Ionized Gases (Wiley, New York, 1964), Chap. 11.
- ⁴J. H. Parker, Jr. and J. J. Lowke, Phys. Rev. 181, 290 (1969).
- ⁵J. J. Lowke and J. H. Parker, Jr., Phys. Rev. 181, 302 (1969).
- ⁶H. N. Kucukarparci, H. T. Saelee, and J. Lucas, J. Phys. D 14, 9
- ⁷H. N. Kucukarparci and J. Lucas, J. Phys. D 14, 2001 (1981).
- ⁸Y. Nakamura and M. Kurachi, J. Phys. D 21, 719 (1988).
- ⁹J. L. Pack and A. V. Phelps, Phys. Rev. 121, 798 (1961).
- ¹⁰J. L. Pack, R. E. Voshall, and A. V. Phelps, Phys. Rev. 127, 2084 (1962).
- 11 Matheson Gas Data Book (Herst Litho, New York, 1966), pp. 23, 249, 313, and 499.
- ¹²S. R. Hunter, J. G. Carter, and L. G. Christophorou, Phys. Rev. A 38, 5539 (1988).
- ¹³E. H. Kennard, Kinetic Theory of Gases (McGraw-Hill, New York, 1938), Chap. 7.
- ¹⁴A. V. Phelps, J. L. Pack, and L. S. Frost, Phys. Rev. 117, 470 (1960). Note that the data given in this paper for E/N < 10 Td are superseded by the more accurate data of Ref. 10.
- ¹⁵Higher-order terms have been considered in Ref. 1. However, when these terms are significant, the theoretical results given by Parker and Lowke (Refs. 4 and 5) are not applicable and the effects of boundary conditions on the electron energy distribution probably become important (Ref. 4).
- ¹⁶N. Anderson and R. G. Keesing, Int. J. Electron. 31, 197 (1971).
- ¹⁷R. A. Duncan, Aust. J. Phys. 10, 54 (1957) and J. J. Lowke, Aust. J. Phys. 15, 39 (1962).
- ¹⁸Y. Nakamura, J. Phys. D 20, 933 (1987).
- ¹⁹L. S. Frost and A. V. Phelps, Phys. Rev. 127, 1621 (1962).
- ²⁰P. H. Luft, Joint Institute for Laboratory Physics Information Center Report No. 14, 1975 (unpublished). This is a detailed analysis of the 1975 version of the code used in Ref. 19 and in the present article to solve the Boltzmann equation for electrons.
- ²¹P. Segur and M. C. Bordage, XIX International Conf. on Phenomena in Ionized Gases: Invited Papers, edited by V. J. Zigman (U. of Belgrade, Belgrade, 1989), p. 86.
- ²² H. Date, K. Kondo, and H. Tagashira, J. Phys. D 23, 1384 (1990).
- ²³J. L. Pack, R. E. Voshall, A. V. Phelps, and L. E. Kline, in Nonequilibrium Effects in Ion and Electron Transport, edited by J. W. Gallagher, D. F. Hudson, E. E. Kunhardt, and R. J. Van Brunt (Plenum,

- New York, 1990), pp. 371-2.
- ²⁴R. W. Crompton, M. T. Elford, and R. L. Jory, Aust. J. Phys. 20, 369 (1967). The cross sections given in this paper are to be preferred over those of Ref. 19 for helium because they are based on more accurate electron transport data.
- ²⁵ M. Hayashi, J. Phys. D **15**, 1411 (1982).
- ²⁶J. J. Lowke, A. V. Phelps, and B. W. Irwin, J. Appl. Phys. 44, 4664 (1973).
- ²⁷S. Trajmar, Phys. Rev. A 8, 191 (1973).
- ²⁸L. Vriens, J. A. Simpson, and S. R. Mielczarek, Phys. Rev. 165, 7
- ²⁹J. H. Anderson, Phys. Fluids 7, 1517 (1964).
- ³⁰L. S. Frost and A. V. Phelps, Phys. Rev. 136, A1538 (1964).
- 31 T. F. O'Malley, Phys. Rev. 130, 1020 (1963).
- ³² H. B. Milloy, R. W. Crompton, J. A. Rees, and A. G. Robertson, Aust. J. Phys. 30, 61 (1977).
- ³³ K. Tachibana, Phys. Rev. 34, 1007 (1986).
- ³⁴E. Rapp and P. Englander-Golden, J. Chem. Phys. 43, 1464 (1965).
- ³⁵A. G. Robertson and J. A. Rees, Aust. J. Phys. 25, 637 (1972).
- ³⁶T. Makabe and M. Shimoyama, J. Phys. D 19, 2301 (1986).
- ³⁷G. Jager and W. Otto, Z. Phys. 169, 517 (1962).
- ³⁸J. Brambring, Z. Phys. **179**, 539 (1964).
- ³⁹ K. H. Wagner, Z. Phys. 178, 64 (1964).
- ⁴⁰D. D. Errett, Ph. D. Thesis, Purdue University, 1951 (unpublished).
- ⁴¹J. S. Townsend and V. A. Bailey, Philos. Mag. 43, 593 (1922).
- ⁴²R. W. Warren and J. H. Parker, Phys. Rev. 128, 2661 (1962).
- ⁴³T. Koizumi, E. Shirakawa, and I. Ogawa, J. Phys. B 19, 2331 (1986).
- ⁴⁴M. Suzuki, T. Taniguchi, and H. Tagashira, J. Phys. D 22, 1848 (1989).
- ⁴⁵Y. Nakamura and A. Naitoh (private communication). See Ref. 44.
- ⁴⁶S. A. J. Al-Amin and J. Lucas, J. Phys. D 20, 1590 (1987).
- ⁴⁷M. Hayashi, J. Phys. D 16, 581 (1983).
- ⁴⁸ H. L. Brooks, M. C. Cornell, J. Fletcher, I. M. Littlewood, and K. J. Nygaard, J. Phys. D 15, L51 (1982).
- ⁴⁹ K. J. Nygaard, H. L. Brooks, and S. R. Hunter, IEEE J. Quantum Electron. QE-15, 1216 (1979).
- ⁵⁰G. Seng and F. Linder, J. Phys. B 9, 2539 (1976).
- 51 See AIP document no. PAPS JAPIA-71-5363-8 for 8 pages of tables of data and calculations of D_I/μ . Order by PAPS number and journal reference from the American Institute of Physics, Physics Auxiliary Publication Service, 335 East 45th Street, New York, NY 10017. The price is \$1.50 for each microfiche (60 pages) or \$5.00 for photocopies of up to 30 pages, and \$0.15 for each additional page over 30 pages. Air mail additional. Make checks payable to the American Institute of Physics.