# Cross Sections for Electron-Carbon Monoxide Collisions in the Range 1-4 eV

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#### Abstract

The scattering of electrons from CO molecules has been studied over the energy range from 1 to 4 eV by analysing drift velocity data for pure CO and CO-inert gas mixtures at 294 K. The validity of using the so-called 'two term approximation' for the velocity distribution function in the solution of the Boltzmann equation to analyse *drift velocity* data for the pure gas (and thus also for the gas mixtures) has been established. The momentum transfer cross section for CO has been determined in the energy range 1–4 eV, and the measurements of the vibrational cross sections by Ehrhardt *et al.* (1968) have been renormalized. By using a solution of the Boltzmann equation which avoids the two term approximation, these cross sections have been shown to be consistent with previous measurements of the transport parameter  $D_{\perp}/\mu$  in pure CO.

### 1. Introduction

An accurate knowledge of the momentum transfer and vibrational excitation cross sections for electron-CO scattering is required for the design and understanding of CO lasers and coal-fired MHD energy converters. Many of these data in the energy range from 0 to 1 eV were derived by Hake and Phelps (1967) from an analysis of the available drift velocity and diffusion coefficient data, but due to a lack of experimental data at high E/N (E is the electric field strength and N the gas number density) the cross sections could not be accurately determined at energies greater than 1 eV. Thus Hake and Phelps were not able to discuss the normalization of Schulz's (1964) measurements of the large vibrational excitation cross sections associated with the lowest shape resonance in CO. Ehrhardt et al. (1968) also studied vibrational excitation in CO experimentally but, as in the case of Schulz's work, it was difficult to determine the *absolute* magnitude of the vibrational cross sections. The normalization of these vibrational cross sections, together with the determination of the momentum transfer cross section up to energies of a few eV, are the primary aims of this work. We shall use  $q_{\rm m}(\varepsilon)$  to denote the momentum transfer cross section,  $q_{\rm v}(\varepsilon)$  to denote the cross section for collisions in which the *j*th vibrational level is excited from the ground vibrational level and  $q_{ex}(\varepsilon)$  to denote the cross section for electronic excitation. We note that  $q_{\rm m}(\varepsilon)$  is a composite cross section representing the loss of momentum in both elastic and inelastic collisions.

The technique used was to measure and analyse drift velocity data for both pure CO and CO-inert gas mixtures. This 'mixture technique' (Engelhardt and Phelps 1964) differs slightly from the normal swarm method in which an analysis is made of two (or more) transport coefficients (usually the drift velocity  $v_{dr}$  and the ratio of the lateral diffusion coefficient to mobility  $D_{\perp}/\mu$ ) in the pure gas. By using more than one transport coefficient it is possible to separate the effects of elastic and inelastic scattering and obtain information about the cross sections for both types of process. However, as described below, the mixture technique enables the same information to be obtained from the measurement of drift velocities alone, and three factors make the use of the mixture technique particularly appropriate in the present work. Firstly, the drift velocity data for CO-inert gas mixtures are required for circuit design and plasma stability considerations in CO-laser discharges; secondly, it has been found (Hake and Phelps 1967) that the parameter  $D_{\perp}/\mu$  is difficult to measure in pure CO due to the presence of a background of negative ions in the laterally diffusing electron stream; and thirdly, the analysis of the  $D_{\perp}/\mu$  data in pure CO cannot be carried out without using a solution of the Boltzmann equation which avoids the two term approximation (see Section 4).

In the mixture technique, drift velocities are measured using mixtures of an inert gas and small concentrations (typically 2–10%) of the molecular gas under study. In mixtures of this kind the electron motion is dominated by *elastic* scattering with the inert gas atoms, and *inelastic* scattering with the molecules. If  $q_m(\varepsilon)$  for the inert gas is accurately known we can obtain an estimate of the inelastic cross sections for the molecular gas. Once these have been estimated, analysis of the measurements in the pure molecular gas gives an estimate of  $q_m(\varepsilon)$  for this species. The calculations for the mixture may then be repeated and the iterative process continued until a consistent set of cross sections for the molecular gas is obtained.

In practice this mixture technique relies firstly on an accurate knowledge of  $q_{m}(\varepsilon)$  for the inert gas and, secondly, on the drift velocity data for the mixtures being sufficiently sensitive to inelastic collisions with the molecular gas and yet sufficiently insensitive to momentum transfer to this gas. The first requirement [i.e. an accurate knowledge of  $q_m(\varepsilon)$  for the inert gas] is easily satisfied if we choose He, Ne or Ar as the inert gas. The second requirement can be investigated by making calculations of the drift velocity as a function of E/N for typical mixtures. Using the numerical solution of the Boltzmann equation due to Gibson (1970) we have found that the vibrational cross sections for CO can be more accurately determined from an analysis of the drift velocity data for CO-Ar mixtures than for CO-He mixtures, given that the accuracy of the experimental data is the same in each case. This effect has been pointed out previously by Engelhardt and Phelps (1964) who used the example of hydrogen-argon mixtures. In fact our calculations show that the drift velocity is four times more sensitive to changes in the CO vibrational cross sections in a 2%CO-Ar mixture than in pure CO. This gives yet more support to the use of the mixture method.

#### 2. Experimental Details and Results

A full description of the design and mode of operation of the drift tube and associated equipment used in this work has been given by Huxley and Crompton (1974). The overall accuracy of the apparatus was checked by repeating some earlier measurements of  $v_{dr}$  for electrons in helium at 293 K (Crompton *et al.* 1967). The largest discrepancy between the present and previous results was 0.2% which compares favourably with the error limits of  $\pm 1\%$  quoted by these authors.

<i>E/N</i> (Td)	$v_{dr}$ (10 <sup>6</sup> cm s <sup>-1</sup> )	<i>E/N</i> (Td)	$v_{\rm dr}$ (10 <sup>6</sup> cm s <sup>-1</sup> )	<i>E/N</i> (Td)	$v_{dr}$ (10 <sup>6</sup> cm s <sup>-1</sup> )
10	2.09	25	3.00	60	5.10
12	2.22	30	3.30	70	5.77
14	2.34	35	3.60	80	6.51
17	2.52	40	3.90	100	8.23
20	2.70	50	4 · 49		

Table 1. Drift velocity of electrons in carbon monoxide at 294 K

Table 2. Drift velocity of electrons in CO-Ar mixtures at 294 K

E/N	$v_{\rm dr}$ (10 <sup>6</sup> cm s <sup>-1</sup> ) for CO concentrations of					$E/N v_{\rm dr} (10^6 {\rm cm  s^{-1}})$ for CO concentrations of					
(Td)	1%	2%	5%	10%	20%	(Td)	1%	2%	5%	10%	20%
2.0	2.10					7.0	2.11	3 · 10	4.25	3.80	3.00
2.5	2.32	2.49	2.34	2.14	1.96	8.0	$2 \cdot 08$	3.02	4.42	4.14	3.24
3.0	2.43	2.78	2.60	2.33	2.07	10.0	$2 \cdot 08$	2.91	4.51	4.72	3.70
3.5	2.44	3.00	2.86	2.51	2.18	12.0	2.14	2.89	4 · 48	5.13	4.15
4·0	2.39	3.15	3.12	2.70	2.29	14.0	2.23	2.91	4.43	5.37	4.59
5.0	2.26	3.25	3 · 59	3.08	2.53	17.0		2.99	4 · 40	5.57	5.18
6.0	2.17	3 · 20	3.97	3.45	2.76	20.0		3.13	4.43	5.65	5.68

Table 3. Drift velocity of electrons in CO-He mixtures at 294 K

E/N	$v_{\rm dr}  (10^6  {\rm c}$	$m s^{-1}$ ) for (	CO concentra	ations of	E/N	$v_{\rm dr}  (10^6)$	$10^6 \mathrm{cms^{-1}}$ ) for CO concentrations of			
(Td)	2%	5%	10%	20%	(Td)	2%	5%	10%	20%	
1.7	0.761				7.0	2.12	2.19	2.10	1.99	
2.0	0.845	0.913	0·995		8.0	2.31	2.42	2.32	2.16	
2.5	0.983	1.04	1.11		10.0	2.65	2.86	2.75	2.50	
3.0	1.12	1.17	1.22		12.0	2.98	3.26	3.21	2.84	
3 · 5	1 · 26	1 · 29	1.32		14.0	3.32	3.62	3.56	3.17	
4·0	1 · 40	1.42	1 · 44	1 · 47	17.0		4.13	4.11	3.66	
5.0	1.67	1.68	1.66	1.64	20.0		4.65	4.61	4.15	
6.0	1.91	1.93	1.88	1.82						

The drift velocity results for CO, CO–Ar and CO–He mixtures are listed in Tables 1, 2 and 3 respectively. The main source of uncertainty in the tabulated data is the magnitude of the correction applied to account for diffusive effects (Lowke 1962; Huxley and Crompton 1974).

We reduced the observed values of the drift velocity according to the expression

$$v_{\rm dr} = v_{\rm dr}' \left( 1 - \frac{1 \cdot 5(D_\perp/\mu)}{V} \right),$$

where  $v_{dr}$  is the true drift velocity,  $v'_{dr}$  is the observed drift velocity, and V is the potential difference between the shutter planes in the drift tube. The values of  $D_{\perp}/\mu$  were calculated from the available cross sections; the factor of 1.5 was derived empirically from earlier investigations. The largest corrections applied to the pure CO data and the mixture data were 0.7% and 2.0% respectively. To ensure that

we have not underestimated the errors from this source in our data we have included additional uncertainties in the error analysis amounting to  $\pm 0.7\%$  for the pure CO data and  $\pm 2.0\%$  for the mixture data.

Errors also arose from the difficulty in measuring electron transit times under conditions where there was a background of negative ions [also observed by Hake and Phelps (1967) at 298 K] and broad peaks in the current-frequency spectra. Negative ions of unknown identity were found at all pressures and mixture concentrations, although the relative abundance of the ions increased with increasing absolute pressure of CO. The presence of these negative ions prevented accurate measurements being made in pure CO at pressures more than 1 kPa and E/N less than 10 Td ( $\equiv 10^{-16} \text{ V cm}^2$ ). The data were repeatable from day to day to  $\pm 0.1\%$  and care was taken to ensure that the mixture constituents had completely mixed before the final data were taken. The total estimated error in the data in Table 1 is less than  $\pm 1.5\%$  and in Tables 2 and 3 is less than  $\pm 3\%$ .

### 3. Discussion of Drift Velocity Data

### Carbon Monoxide

The only room-temperature experimental data available for comparison with the present work are those of Pack *et al.* (1962) which extend up to 25 Td. The present data (Table 1) are about 10% higher at 25 Td. Hake and Phelps (1967) made calculations of the drift velocity in CO over a wide range of E/N using a set of cross sections derived from the available transport data. They made no attempt to adjust the cross sections for energies above 1 eV and consequently used values of  $q_m(\varepsilon)$  at energies above 1 eV which were lower than ours, resulting in calculated drift velocities higher than our values over the whole range of our measurements. The discrepancy ranges from about 3% at 10 Td to about 30% at 100 Td.

## CO-Ar and CO-He Mixtures

The present data for CO-Ar mixtures are shown in Fig. 1 where they are compared with the predictions of Long *et al.* (1976).

The data for mixtures containing from 1% to 5% CO show the phenomenon of negative differential conductivity which may contribute to the instability of the discharges in some CO-laser systems. This phenomenon can occur when the combination of elastic and inelastic cross sections is such that an increase in E/N leads to an abnormally large increase in the elastic collision frequency. There is then the possibility that the enhanced randomization of the directions of the velocity vectors leads to a reduction in the drift velocity even though the average electron speed increases (Kleban and Davis 1977). Negative differential conductivity is likely to occur when the average collision frequency for momentum transfer  $\langle v_m \rangle$  increases more rapidly than E (Long *et al.* 1976) and has been shown to occur even when the elastic cross section is constant (Z. L. Petrovic, R. W. Crompton and G. N. Haddad, personal communication). In the present case the combination of the rapidly increasing elastic cross section for argon above the Ramsauer minimum and the  $CO \ 0 \rightarrow 1$  vibrational cross section produces the phenomenon when the CO relative abundance is sufficiently small.

The discrepancies between the present experimental data and the theoretical predictions of Long *et al.* (1976) are almost certainly due to errors in the Ar momentum



Fig. 1. Electron drift velocity as a function of E/N for various CO-Ar mixtures. Solid curves represent present data and broken curves represent the predictions of Long *et al.* (1976). The fractional concentration of CO is marked on each plot.

transfer cross section and the CO cross sections used by these workers. Their predictions for CO-He mixtures are in much better agreement with the experimental data (the maximum discrepancy being  $\approx 5\%$ ) due to the reduced sensitivity of the measurements in CO-He mixtures to the vibrational cross sections in CO.

### 4. Determination of CO Cross Sections

The two term approximation in Holstein's (1946) solution of the Boltzmann equation breaks down when the inelastic cross sections are sufficiently large in relation to the total scattering cross section (Reid 1979). Using this criterion as a guide one



**Fig. 2.** Differences between transport coefficients in pure CO, calculated using the two term approximation and calculated avoiding this approximation, as a function of E/N. The open symbols refer to the drift velocity  $v_{dr}$  and the closed symbols to the product of number density and lateral diffusion coefficient  $ND_{\perp}$ . The circles correspond to calculations using the solution by Lin *et al.* (1979) and the squares are the Monte Carlo calculations by G. L. Braglia (personal communication).



Fig. 3. Differences between transport coefficients in a 20% CO in argon mixture, calculated using the two term approximation and calculated avoiding this approximation, as a function of E/N. The open circles refer to  $v_{dr}$  and the closed circles to  $ND_{\perp}$ .

might therefore expect the two term approximation to break down in pure CO and some CO-Ar mixtures. We have therefore made calculations using a multiterm solution of the Boltzmann equation due to Lin *et al.* (1979) for both pure CO and CO-Ar mixtures. The results of these calculations are shown in Figs 2 and 3 respectively where they are compared with the two term results. Also shown in Fig. 2 are some calculations made by G. L. Braglia (personal communication) using a Monte Carlo technique which confirm the results of the multiterm analytical code. It can be seen that there are significant errors in the values of  $D_{\perp}/\mu$  calculated using the two term approximation. On the other hand the drift velocity is calculated sufficiently accurately with this approximation.

These calculations confirm the fact that we can analyse the *drift velocity* data in both CO and CO-Ar mixtures using methods based on the two term approximation. However, in order to calculate  $D_{\perp}/\mu$  in pure CO from an appropriate set of cross sections, we should use a solution of the Boltzmann equation which avoids the two term approximation. The following discussion of the fitting of cross sections to the transport data uses calculations made with a numerical solution of the Boltzmann equation developed by Gibson (1970)—a solution which invokes the two term approximation.

The transport data listed in Tables 1–3 are only slightly influenced by excitation and de-excitation of the rotational states of CO molecules. For this reason no attempt was made to modify the expressions for the rotational cross sections adopted by Hake and Phelps (1967) who used the formula derived by Takayanagi (1966). If the rotational cross sections are eliminated entirely it results in at most a 0.3%decrease in the calculated drift velocities for pure CO over the range of E/N from 10 to 100 Td. The values of the momentum transfer cross sections for the inert gases used in the calculations were those of Haddad and O'Malley (1982) for Ar, and Milloy and Crompton (1977) for He.



Vibrational and Electronic Excitation Cross Sections

The vibrational and electronic excitation cross sections for CO show two distinct maxima in the energy range from 0.1 to 20 eV. The sum of the vibrational cross sections reaches a value of about  $6 \text{ Å}^2$  at about 1.8 eV, while the sum of the electronic excitation cross sections has a peak of about  $1.5 \text{ Å}^2$  at around 7 eV. It follows that there is almost complete separation of the influence of vibrational and electronic excitation processes on the electron swarm, which makes it possible to define approximate E/N ranges over which the drift velocity data are primarily sensitive to one or the other type of process.

The value of  $q_{v_1}(\varepsilon)$  in the energy range less than about 1 eV is more easily determined from transport coefficient analysis than from beam experiments. Consequently, we initially chose from Hake and Phelps (1967) values of  $q_{v_1}(\varepsilon)$  in this range. Since an adequate fit to the experimental data was obtained using this cross section at energies less than 1 eV we retained the shape of the cross section in this region. For energies greater than about 1 eV we used results by Ehrhardt *et al.* (1968) for  $q_{v_j}(\varepsilon)$ for j = 1-7, and results by Boness and Schulz (1973) for j = 8-10. The only modification made (apart from a normalization factor which was applied to all the vibrational cross sections) was to blend the data by Hake and Phelps for  $q_{v_1}(\varepsilon)$  with those by Ehrhardt *et al.* as shown in Fig. 4. Data for the six electronic excitation cross sections and the ionization cross section were initially taken from Land (1979).

In the calculations used to fit the cross sections to the measured transport data, different normalizing factors were applied to the set of vibrational cross sections [apart from the  $q_{v_1}(\varepsilon)$  cross section at energies less than about 1 eV which remained unchanged throughout] and to the set of electronic excitation cross sections. These parameters were adjusted to give the best fit to the data. Transport data calculated using this cross-section set in mixtures with low concentrations of CO in Ar showed discrepancies of up to about 9% when compared with the experimental data. No changes to the normalization factors could be found which would reduce the discrepancy below this level and it was apparent that it was the electronic excitation cross sections which had to be altered in order to achieve a satisfactory fit to the experimental data.



Measurements by Swanson *et al.* (1975) show resonance structure in the  $a^3\pi$  cross section near threshold. Accordingly we have reduced the  $a^3\pi$  threshold to 6.01 eV (Krupenie 1966) and incorporated a peak in the cross section near this threshold (see Fig. 5). This modification, together with normalizing factors of 1.30 on the vibrational cross sections and 0.7 on the electronic cross sections, allows us to fit all the data to within  $\pm 4\%$ . Omitting the peak structure in the  $a^3\pi$  cross section gives a difference of about 18% between calculated and measured values for the mixture of 1% CO in argon. Some of this difference can of course be removed by adjusting the normalizing factors but we are still left with the discrepancy of about 9% between measured and calculated values for this same mixture.

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ε (eV)	$q_{\rm m}(\varepsilon)$ (Å <sup>2</sup> )	ε (eV)	$q_{\rm m}(\varepsilon)$ (Å <sup>2</sup> )	ε (eV)	$q_{\rm m}(\varepsilon)$ (Å <sup>2</sup> )	ε (eV)	$q_{\rm m}(\varepsilon)$ (Å <sup>2</sup> )
0.4	13.5	1.2	21.5	1.7	43.0	2.4	25.0
0.7	14.5	1.3	26.5	1.9	43.0	3.0	18.5
1.0	16.0	1.5	38.0	2.1	35.0	4.0	13.0





Fig. 7. Differences between the predicted and measured values of the electron drift velocity in (a) a 1% CO-Ar mixture and (b) pure CO as a function of E/N using the cross sections derived in the present work (solid curve). The results obtained using the cross sections derived by Land (1979) are shown for comparison (dashed curve).

These modifications were retained from this point for the remainder of the calculations. Naturally we make no claim as to the uniqueness of this set of cross sections but have simply obtained a fit to the experimental data by incorporating these modifications.

### Momentum Transfer Cross Section

After using the mixture data to determine our best fit values of the normalization factors for  $q_{v_j}(\varepsilon)$  and  $q_{ex}(\varepsilon)$ , the momentum transfer cross section  $q_m(\varepsilon)$  for electron-CO scattering was derived by fitting to the data for pure CO shown in Table 1. The present determination (Table 4) is thought to be in error by less than 15% at energies up to 4 eV.

#### 5. Discussion

The present determination of the momentum transfer cross section for CO is compared in Fig. 6 with the previous determination by Land (1979) and that of Hake and Phelps (1967). It can be seen that the cross section derived in the present work has the same magnitude and shape as that derived by Land but the maximum is displaced by about 0.3 eV.

The vibrational excitation cross sections derived by Land are very much larger than the cross sections derived here, except for  $q_{v_1}(\varepsilon)$  at energies less than about 1 eV. Land used the same energy dependence for  $q_{v_1}(\varepsilon)$  (from Ehrhardt *et al.* 1968) but found a best fit with a normalization factor of 1.9 (cf. our value of 1.30). Fig. 7a shows the results of using the two cross-section sets (ours and Land's) to predict the drift velocity for a 1% CO-Ar mixture. The predictions of the drift velocity for values of E/N less than about 5 Td are much more sensitive to the vibrational cross sections than to the electronic excitation cross sections whereas the opposite is true for higher E/N values. The ranges of E/N influenced by these two types of cross sections cannot, of course, be completely separated. From Fig. 7a it is clear that the normalization factor of 1.30 provides an acceptable fit to the experimental data for this mixture in the range of E/N most sensitive to the vibrational cross sections, whereas the factor of 1.9 does not.

In pure CO the differences between the predicted and experimental values of  $v_{\rm dr}$  using the two cross-section sets (ours and Land's) are not as large. In fact, as shown in Fig. 7b, the two sets predict the drift velocity in pure CO equally well. This emphasizes the greater sensitivity of the electron transport data for CO-Ar mixtures to variations in the inelastic cross sections.

The situation with respect to  $D_{\perp}/\mu$  in pure CO is summarized in Fig. 8. The experimental values found by Skinker and White (1923), which were the data used by Land in his analysis, are shown for comparison. As can be seen in Fig. 8*a*, the present set of cross sections does not predict the experimental data particularly well when calculations are made using the two term approximation (solid curve), whereas Land's cross-section set predicts the data quite well (dashed curve), as is to be expected. On the other hand, Fig. 8*b* shows that the present cross-section set used in conjunction with a solution which avoids the two term approximation predicts the data quite well whereas, if Land's cross-section set is used, the predicted values are generally substantially lower than the experimental values over this range of E/N.

In summary, the difference between Land's set of cross sections and ours can be attributed to the fact that his analysis was based on data for the lateral diffusion



**Fig. 8.** Available experimental data for  $D_{\perp}/\mu$  (Skinker and White 1923) compared with calculations using (a) the two term solution of the Boltzmann equation and (b) a solution of the Boltzmann equation which avoids the two term approximation. Solid curves correspond to points calculated using cross sections derived in the present work; in (b) the solid curve also includes some points calculated with Monte Carlo techniques by G. L. Braglia (personal communication). Dashed curves correspond to calculations using cross sections derived by Land (1979).

coefficient  $D_{\perp}$  in pure CO as well as on data for  $v_{dr}$ , and that inadequate corrections were made for errors in  $D_{\perp}$  which result from the use of the two term approximation. Corrections were applied only for E/N values greater than 110 Td, whereas Fig. 2 shows that the use of the approximation results in errors of up to 10% in the calculated values of  $D_{\perp}/\mu$  at about 70 Td. By contrast, the present analysis relies only on drift velocity data for pure CO and for CO-Ar and CO-He mixtures for which the use of the two term approximation has been shown to be adequate (although the analysis could have been carried out using a multiterm Boltzmann code had it been necessary).

#### 6. Conclusions

We have derived a momentum transfer cross section for CO over the energy range from 1 to 4 eV and a normalization factor for the vibrational excitation cross sections obtained from beam experiments.

It has been shown that values of the transport parameter  $D_{\perp}/\mu$  in pure CO or CO-Ar mixtures must be analysed using a solution of the Boltzmann equation which avoids the two term approximation.

It would obviously be desirable to have accurate measurements of  $D_{\perp}/\mu$  in pure CO which would provide an additional set of constraints on the cross sections.

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