

# Electron mobilities in low density hydrogen and carbon monoxide gases: Momentum transfer cross sections at very low energies

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Values of the momentum transfer cross sections  $\sigma_m$  of hydrogen and carbon monoxide molecules for very low energy electrons (1–300 meV, 0.2–50 zJ;  $z = \text{zepto} = 10^{-21}$ ) have been refined by electron mobility measurements in the gases over wide ranges of temperature. This is part of an ongoing study of effects of molecular shape on electron transport in fluids. Electron mobilities in hydrogen gas at temperatures 17–450 K are roughly consistent with previously reported values of  $\sigma_m$ , but do not exclude the possibility of a  $\sigma_m$  minimum of  $5.2 \times 10^{-20} \text{ m}^2$  at an electron energy  $\xi \leq 0.7 \text{ zJ}$ . Carbon monoxide molecules have a minimum  $\sigma_m$  of  $5.8 \times 10^{-20} \text{ m}^2$  at  $\xi = 5 \text{ zJ}$ . A new treatment of low energy electron scattering by CO is needed.

## I. INTRODUCTION

The scattering of very low energy ( $< 10^{-21} \text{ J}$ ,  $10^{-2} \text{ eV}$ ) electrons by diatomic and polyatomic molecules is not well understood.<sup>1–3</sup> Molecular shape and density have important effects on low energy electron scattering in gases, which have not been explained (see Refs. 1–8 in Ref. 3).

We have extended the study<sup>2</sup> by measuring electron mobilities in hydrogen and carbon monoxide gases over wider ranges of temperature than previously reported. Momentum transfer cross sections for electrons in hydrogen are available<sup>4</sup> only for electron energies  $> 3 \text{ zJ}$  ( $z = \text{zepto} = 10^{-21}$ ).<sup>5</sup> Values of electron mobilities in carbon monoxide at  $T > 195 \text{ K}$  are uncertain<sup>6</sup> because of the presence of an electron capturing impurity. Reported values of the electron momentum transfer cross section of carbon monoxide differ by up to 30%.<sup>6–8</sup>

## II. EXPERIMENT

### A. Materials

Matheson Ultrahigh Purity hydrogen (99.999%) and Matheson Research Grade carbon monoxide (99.99%) were used after further purification. (This purity of CO was an order of magnitude higher than the sample used in Ref. 7.) The gas cylinders were attached to a greaseless high vacuum line through metal bellows using Kovar seals.

The hydrogen was passed through two cold traps at 77 K followed by a 60 cm column of freshly activated metallic copper in the form of spherical pellets (kept at 773 K). The hydrogen was then passed through a liquid sodium–potassium alloy and stored over a freshly regenerated potassium mirror in a two litre Pyrex flask for at least a week before use in the experiments.

The carbon monoxide gas was passed through a column of potassium hydroxide pellets (previously degassed at  $\sim 150^\circ \text{C}$  to  $10^{-5} \text{ Pa}$ ) to reduce the  $\text{CO}_2$  level. The gas was then passed through two traps kept at liquid oxygen temperature ( $\sim 90 \text{ K}$ ) to reduce the condensable impurities.

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The initial portion of the flowing gas was discarded, to flush the system. A trace of oxygen could not be removed by storing the gas over a potassium mirror, because of a violent reaction of CO with potassium which produces a black powdery compound.<sup>9</sup>

The indication of a good gas sample in the conductance cell was the linear decay of the electron current transient (ECT) at low electric fields, after a pulse of x rays.

The conductance cells and other equipment were the same as those used for nitrogen.<sup>2</sup>

### B. Sample preparation and irradiation

The evacuated conductance cells were baked at about 520 K (or about 20 K above the maximum temperature of the experiment) at  $10^{-5} \text{ Pa}$  for about a week until no further degassing was evident (the mobility measurements in a sample were stable with time). The hot cell was rinsed with hydrogen and then reevacuated before cooling and refilling the cell at a measured temperature. This procedure was not followed in the case of CO, because hot stainless steel can react to form metal carbonyls which can capture electrons in the subsequent experiments.

The cell was immersed in a constant temperature bath (usually fresh liquid nitrogen under known barometric pressure), then opened to a purified gas reservoir. Care was taken to prevent condensation of oxygen into liquid nitrogen. After allowing sufficient time for temperature equilibration, the gas pressure was measured with a mercury manometer (isolated by a trap at 77 or 90 K in the cases of hydrogen or CO, respectively, to reduce mercury vapor contamination). The cell was sealed with a small flame at a capillary point to minimize heating of the gas sample and thus minimize the error in the calculated number density. The estimated maximum error in the density is 1%.

The radiation pulse dose was minimized to avoid space charge distortion of the electron conductance transient.<sup>2</sup> The number of ion pairs/ $\text{m}^2$  per pulse was  $\sim 3 \times 10^7$  in hydrogen and  $12 \times 10^7$  in carbon monoxide. The respective maximum space charges were equivalent to  $E/n \leq 0.03$  and  $0.11 \text{ yV m}^2/\text{molec}$  ( $y = \text{yocto} = 10^{-24}$ ),<sup>5</sup> which were negli-

gible. When signal averaging was done to improve the signal-to-noise ratio, enough time was left between pulses to remove all the positive ions from the drift space.

Measurements were made with both positive and negative applied voltages, and averaged. Usually the differences in results were less than 2%.

Overall errors in mobility measurements were up to 3%.

### III. RESULTS AND DISCUSSION

#### A. Hydrogen

The density normalized mobility  $n\mu$  was measured at a number of temperatures ranging from 16 K to about 496 K in hydrogen as a function of the density normalized field  $E/n$ , and at number densities ranging from 9.2 to 73 Ymolec/m<sup>3</sup> ( $Y = \text{yotta} = 10^{24}$ ).<sup>5</sup> Since our main objective was to obtain the zero field limit of the electron mobility, the  $E/n$  values were below 0.08 zV m<sup>2</sup>/molec (0.08 Td). The  $n\mu$  values increased with decreasing  $E/n$ , indicating that the electrons were not in thermal equilibrium with the gas molecules even at the lowest  $E/n$  that gave a good signal. The field dependence decreased with increasing temperature. Near 490 K,  $n\mu$  was essentially independent of  $E/n$  up to 0.07 Td. Thus, the energy gained by the electrons from the applied electric field was more efficiently removed by the collisions with hydrogen molecules at the higher temperatures, presumably through rotational excitation of the molecules.

Mobilities were extrapolated to zero field strength by a plot of  $(\ln n\mu)^{-1}$  against  $E/n$  to obtain  $n\mu_0$  (Fig. 1). The  $(\ln n\mu)^{-1}$  plots provided the least uncertain extrapolations to zero field, of the several types tried. The plots are justified empirically; the theory of electron scattering at very low energies is inadequate for extrapolation of  $n\mu$  to zero field.<sup>3</sup>

Plots of  $n\mu_0$  against temperature at two densities, 20.1 and 62.4 (Ymolec/m<sup>3</sup>), are shown in Fig. 2. The value of  $n\mu_0$  decreases with increasing density at low  $T$ , but the relative dependence decreases with increasing  $T$ . This behavior is attributed to the Ioffe-Regel effect.<sup>10</sup> Values of  $n\mu_0$  at several gas densities are listed in Table I.

The temperatures used at each density were not the same, so interpolated values at a given temperature were taken from the  $n\mu_0$  plots against temperature. A plot of  $n\mu_0$  against  $n$  at a given temperature was extrapolated to  $n = 0$  to obtain  $n\mu_{00}$  (Fig. 3).

The zero field, zero density values  $n\mu_{00}$  are plotted against temperature in Fig. 4. Low field, low density values of Pack and Phelps<sup>11</sup> are included for comparison. The drift velocities for electrons at different field strengths and densities at 77.6 and 293 K in hydrogen obtained by Lowke<sup>12</sup> were used to calculate the  $n\mu$  values at each temperature and number density and from these the  $n\mu_0$  and  $n\mu_{00}$  were obtained from plots similar to Figs. 1 and 3. These  $n\mu_{00}$  values are included in Fig. 4 for comparison. The value of  $n\mu_{00}$  at 76.8 K was similarly obtained from data of Robertson<sup>13</sup> and is included.

The method used to extract cross sections from the mobility measurements has been described.<sup>14,15</sup> Briefly, Eq. (1) was fitted to the experimental array of  $(n\mu_{00}, T)$  values shown in Fig. 4:

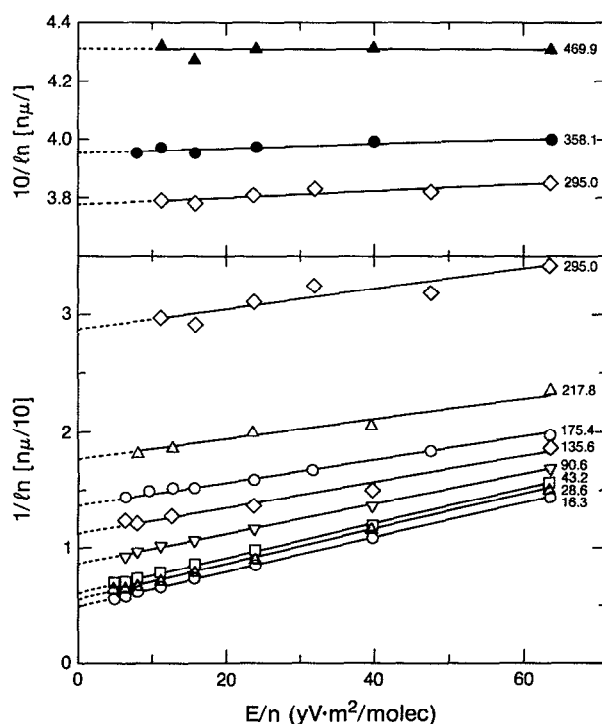


FIG. 1. Plots to extrapolate  $(\ln n\mu)^{-1}$  values of electrons in hydrogen gas to zero electric field strength  $E/n$ . yV = yoctovolt =  $10^{-24}$  V (Ref. 5); yV m<sup>2</sup>/molec = mTd. Gas density  $n = 62.4$  Ymolec/m<sup>3</sup>; Y = yotta =  $10^{24}$  (Ref. 5). The unit of  $n\mu$  is Ymolec/m V s. The values of  $T$  (K) are indicated.

$$n\mu = \frac{4e}{3(2\pi m)^{1/2}} (k_B T)^{-5/2} \int_0^\infty \frac{\xi}{\sigma_m} \exp(-\xi/k_B T) d\xi$$

$$= 1.26 \times 10^{53} T^{-2.5} \int_0^\infty \frac{\xi}{\sigma_m} \exp\left(-7.24 \times 10^{22} \frac{\xi}{T}\right) d\xi, \quad (1)$$

where  $\xi = mv^2/2$  is the electron kinetic energy and all units are SI. Equation (1) was integrated numerically, using 93 logarithmic steps between electron energies 0.02 and 80 zJ

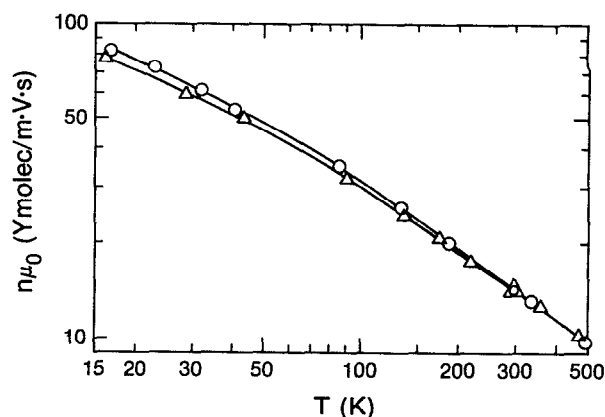
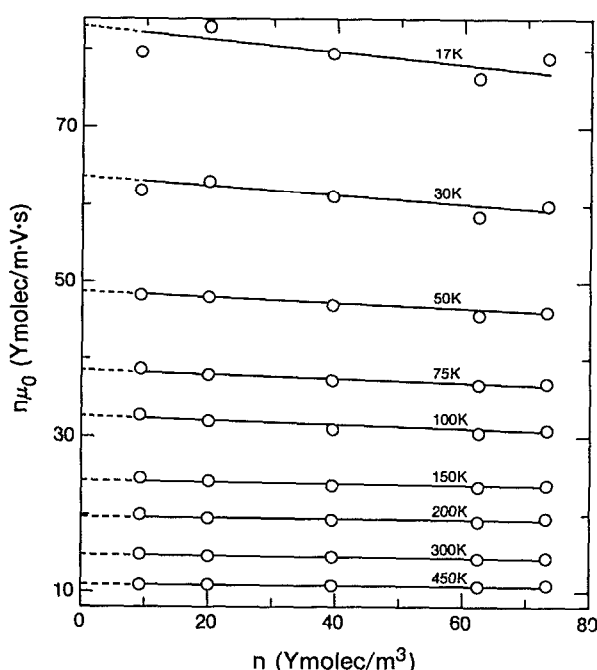


FIG. 2. Temperature dependence of zero field mobilities of electrons in hydrogen.  $n$  (Ymolec/m<sup>3</sup>): O, 20.1;  $\Delta$ , 62.4.

TABLE I. Zero field electron mobilities in hydrogen gas at various temperatures and densities.

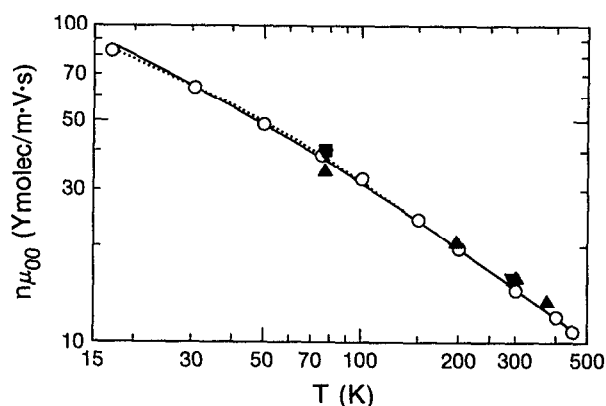
$n$ (Ymolec/m <sup>3</sup> )	$T$ (K)	$n\mu_0$ (Ymolec/m V s)	$n$ (Ymolec/m <sup>3</sup> )	$T$ (K)	$n\mu_0$ (Ymolec/m V s)
9.2	17.0	78 ± 4	62.4	16.3	77
	26.0	66 ± 2		28.6	60
	51.0	48		43.2	50
	91.7	34.9		90.6	32
	132.8	27.3		135.6	24.4
	171.9	21.7		175.4	20.9
	251.6	16.7		217.8	17.6
	294.4	15.2		295.0	14.2
	356.9	13.2		358.1	12.6
				469.9	10.2
20.1	16.8	82	73.2	16.1	82
	23.0	72.4		32.2	58
	32.0	61.6		67.7	40
	40.7	53		97.6	32.0
	86.1	35.0		129.2	25.6
	134.6	25.9		174.9	21.3
	187.5	20.0		244.3	17.2
	294.6	14.1		296.1	14.8
	336.5	13.3		327.8	13.9
	496.0	10.2		371.0	12.7
39.4	18.7	77	0	17	83 ± 3
	31.8	59		30	63.7
	57.7	43		50	48.7
	96.8	31.2		75	38.5
	126.2	25.9		100	32.5
	193.1	20.3		150	24.3
	242.4	16.4		200	19.7
	295.1	14.1		300	14.7
	357.3	12.5		400	12.0
	456.8	10.9		450	10.9

( $10^{-4}$  and 0.5 eV). The momentum transfer cross section  $\sigma_m$  is energy dependent,<sup>14</sup> so the  $(\sigma_m, \xi)$  distribution was adjusted by trial and error to optimize the agreement between the calculated and experimental arrays of  $(n\mu_{00}, T)$

FIG. 3. Variation of  $n\mu_0$  with hydrogen density at the temperatures indicated.

values. The solid line in Fig. 4 was obtained with values from the solid line in Fig. 5. The low energy ends of the curves in Fig. 5 were terminated at the energy where, at the lowest temperature, electrons with lower energies than that contributed 4% to the transport of the population. Figure 5 also shows the cross sections of Crompton<sup>4</sup> and those of Frost and Phelps<sup>14</sup> for comparison. Our values agree closely at electron energies 2–3 zJ, but are 8–10 % higher at 10–20 zJ.

It seems unlikely that a Ramsauer–Townsend minimum

FIG. 4. Variation of  $n\mu_{00}$  with temperature in hydrogen.  $\circ$ , experimental, this lab;  $\blacktriangle$ , low field averages of Pack and Phelps (Ref. 11) at low density;  $\blacktriangledown$ ,  $n\mu_{00}$  obtained from data of Lowke (Ref. 12);  $\blacksquare$ ,  $n\mu_{00}$  obtained from data of Robertson (Ref. 13). Solid and dotted lines were calculated from Eq. (1) using the corresponding cross sections from Fig. 5.

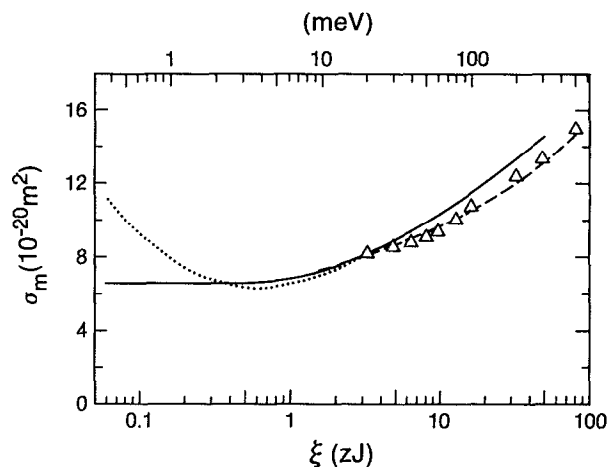


FIG. 5. Electron momentum transfer cross sections of hydrogen.—and ···, from corresponding curves in Fig. 4.—, values from Ref. 4.  $\Delta$ , cross sections from Ref. 14.  $z = \text{zepto} = 10^{-21}$  (Ref. 5).

exists in the electron momentum-transfer cross section of hydrogen molecules, but we examined the extent to which our data *exclude* the possibility that an RT minimum exists for hydrogen. The average polarizability of  $\text{H}_2$  ( $0.91 \times 10^{-40} \text{ C m}^2/\text{V}$ )<sup>16</sup> is twice that of Ne ( $0.44 \times 10^{-40} \text{ C m}^2/\text{V}$ )<sup>17</sup> and half that of Ar ( $1.83 \times 10^{-40} \text{ C m}^2/\text{V}$ );<sup>17</sup> the low energy scattering cross section of Ne is very small<sup>18</sup> and on the verge of having an RT minimum, whereas the cross section of Ar has a well defined RT minimum.<sup>19</sup> In Fig. 5 we arbitrarily drew a minimum in the cross sections at very low energies and recalculated  $n\mu_{00}$  at a series of temperatures; the  $n\mu_{00}$  curve remained the same as before. We then gradually moved the minimum to higher energies until the calculated values of  $n\mu_{00}$  were significantly different from the full curve in Fig. 4. The dotted curve in Fig. 5 generated that in Fig. 4. If an RT minimum exists for  $\text{H}_2$ , it lies at  $<0.7 \text{ zJ}$  (4 meV).

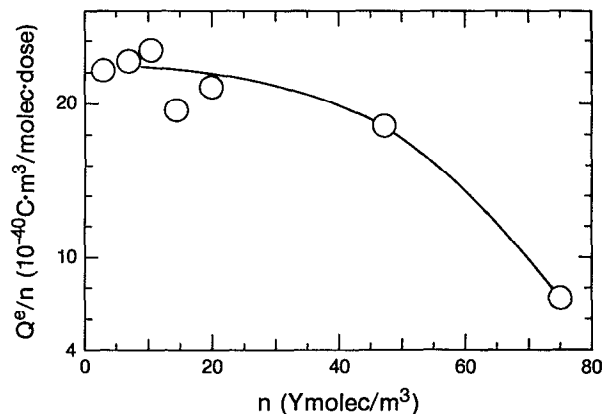


FIG. 6. Effect of electron capturing impurity produced in  $\sim 1 \text{ h}$  at 295 K by reaction of CO with metal components in the cell.  $Q^*$  = electron charge collected per unit x-ray dose at  $E/n = 0.2 \text{ zV m}^2/\text{molec}$  (0.2 Td);  $n$  = no. density of CO.

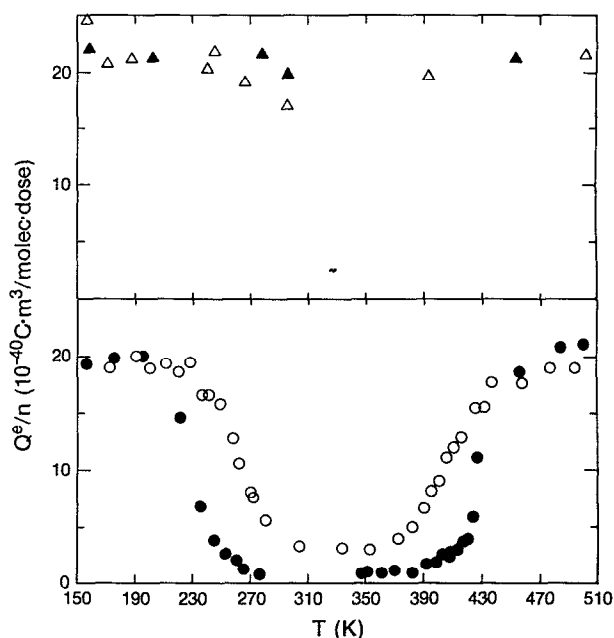


FIG. 7. Variation of  $Q^*/n$  with temperature in CO. Triangles, about 1 h after preparing sample, a few minutes after reaching the temperature indicated. Circles, after 2 (○) or 3 (●) cycles of heating to 500 K and cooling to 300 K.  $n$  (Ymolec/ $\text{m}^3$ ):  $\Delta$ , ○, 2.9;  $\blacktriangle$ , ●, 10.6.

## B. Carbon monoxide

Electron transport measurements in carbon monoxide are affected by an electron scavenging impurity that is produced by contact between carbon monoxide and metal. Pack *et al.*<sup>6</sup> found that the impurity condensed out of the gas below 200 K, so they used measurements at 77 and 195 K to estimate the momentum transfer cross sections.

Our conductance cells contain stainless steel electrodes and Kovar. The fraction of electrons captured by the impuri-

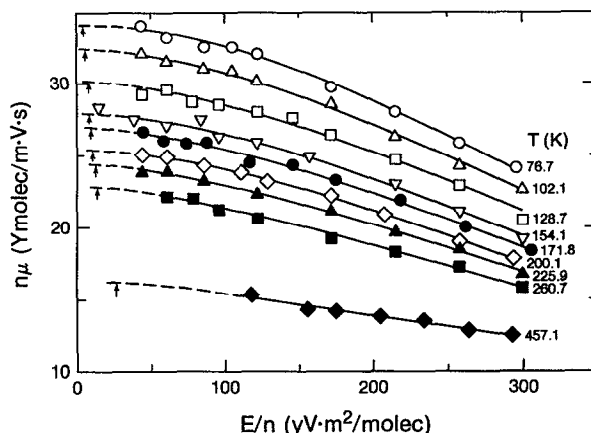


FIG. 8. Variation of  $n\mu$  of electrons with electric field strength in carbon monoxide gas at different temperatures. Each point is the average of values obtained at  $n = 6.9$  and  $47.2 \text{ Ymolec}/\text{m}^3$ . The arrows indicate  $(E/n)_{c_0}$ , the field strength at which the electron drift velocity equals the speed of sound  $c_0$  in the gas (Table II).

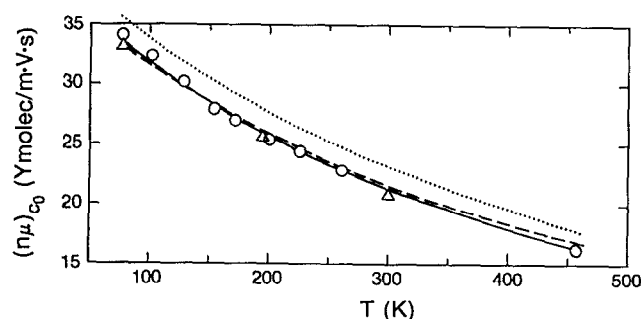


FIG. 9. Variation of  $(n\mu)_0$  with temperature in carbon monoxide gas. O, from Fig. 8.  $\Delta$ , "low field" averages at low density, from Ref. 11. Solid, dotted, and dashed lines were calculated from Eq. (1), using the corresponding cross sections from Fig. 10.

ty in a sample at 295 K increases with time after preparation and with the number density  $n$  of CO in the cell. The amount of electron charge  $Q^e$  collected per unit of x-ray dose at an applied field strength of 0.2 Td, and normalized for  $n$ , is plotted against  $n$  in Fig. 6. Each measurement was made about an hour after the sample was prepared.

The same sample could not be used for successive runs at temperatures above 200 K. Values of  $Q^e/n$  obtained from samples that had been passed through two or three cycles of heating to 500 K and cooling to 300 K are shown in Fig. 7. The electron scavenging impurity condenses out at  $T < 270$  K, and thermally decomposes at  $T > 400$  K. Successful measurements of electron mobilities were made at  $T < 270$  K and  $> 450$  K.

The impurity is probably iron pentacarbonyl, which has a vapor pressure of  $\sim 10$  Pa at 220 K,<sup>20</sup> and decomposes thermally at  $T > 420$  K.<sup>21</sup>

The values of  $n\mu$  decreased with increasing  $E/n$  and  $T$ . Within the  $\pm 1\%$  experimental scatter they were independent of  $n$  in the region 7 to 47 Ymolec/m<sup>3</sup>. The averages of the  $(n\mu, E/n)$  sets at  $n = 6.9$  and 47.2 Ymolec/m<sup>3</sup> had a scatter of  $\pm 0.6\%$  and are shown in Fig. 8.

TABLE II. Thermal electron mobilities in carbon monoxide gas.

$T$ (K)	$c_0^a$ (m/s)	$(E/n)_{th}^b$ (yV m <sup>2</sup> /molec)	$(n\mu)_{c_0}^c$ (Ymolec/m V s)
76.7	178	5	34.1
102.1	206	6	32.4
128.7	231	8	30.1
154.1	253	9	27.9
171.8	267	10	27.0
200.1	288	11	25.4
225.9	306	13	24.4
260.7	329	14	22.8
457.1	436	27	16.2

<sup>a</sup>Speed of sound in the gas, from Eq. (3).

<sup>b</sup>Approximate field strength above which electron heating occurs, Eq. (2).

<sup>c</sup>Extrapolated value of thermal electron mobility, from Fig. 8.

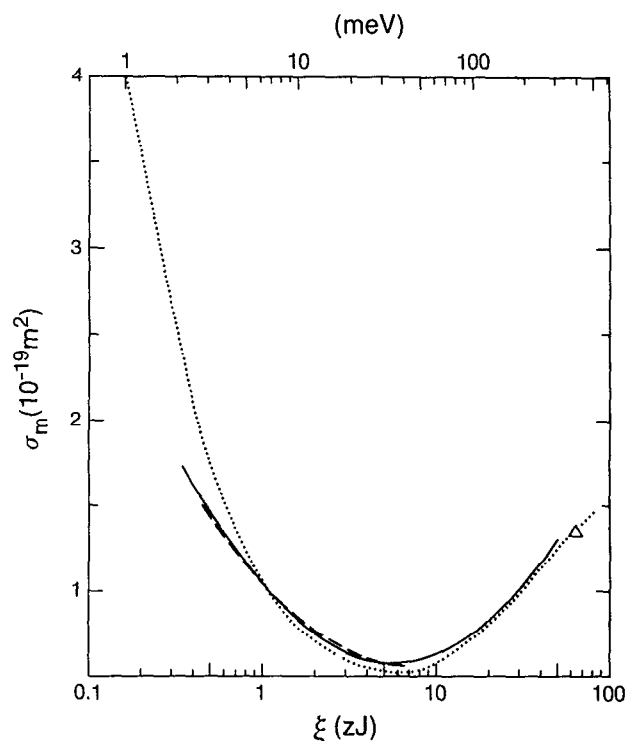


FIG. 10. Electron momentum transfer cross sections of CO. Solid line, from fitting Eq. (1) to data in Fig. 8; dotted line, from Ref. 7; dashed line, from Ref. 6;  $\Delta$ , Ref. 25.

As in nitrogen,<sup>2</sup> the thermal electron mobilities were obtained by extrapolating  $n\mu$  to the estimated threshold field strength for electron heating, where the electron drift velocity approximately equals the speed of sound:<sup>22</sup>

$$(E/n)_{th} \approx c_0/n\mu. \quad (2)$$

The speed of sound  $c_0$  in a gas is related to the average speed of the molecules. It indicates the approximate drift speed that the electrons must attain to be "hot." The value was calculated from Eq. (3):<sup>23</sup>

$$c_0 = (\gamma RT/M)^{1/2} = 20.4 T^{1/2} \text{ m/s}, \quad (3)$$

where  $\gamma = 1.40$  is the heat capacity ratio of carbon monoxide,<sup>24</sup>  $R$  is the gas constant, and  $M$  is the molar mass (0.028 kg/mol). The heat capacity ratio gives an indication of internal motions in the molecules.

Plots of  $(\ln n\mu)^{-1}$  against  $E/n$  were too curved for reliable extrapolations, so the plots in Fig. 8 were extrapolated directly. The corresponding values of  $(n\mu)_{c_0}$  are plotted in Fig. 9 and listed in Table II.

Momentum transfer cross sections were obtained by fitting Eq. (1) to the data in Fig. 9. The full line in Fig. 10 represents the cross sections so obtained. The earlier data of Phelps *et al.*<sup>6,7</sup> are included in Figs. 9 and 10 for comparison. The values of  $\sigma_m$  reported in Ref. 6 were relatively accurate at low energies, but they did not extend to energies above 6.4 zJ (40 meV). The value at the lowest energy reported by Haddad and Milloy<sup>25</sup> agrees with earlier work.

The momentum transfer cross section of CO has a minimum value of  $5.8 \times 10^{-20} \text{ m}^2$  at an electron energy of 5 zJ (Fig. 10). The minimum is partly attributable to the small

TABLE III. Molecular properties<sup>a</sup> and electron scattering minima.

	$D$ ( $10^{-30}$ C m)	$\bar{\alpha}$ ( $10^{-40}$ C m <sup>2</sup> /V)	$\alpha_{\parallel} - \alpha_{\perp}$	min $\sigma_t$ <sup>b</sup> ( $10^{-20}$ m <sup>2</sup> )	$\xi_{\min}$ <sup>c</sup> (zJ)
He	0	0.23	0	4.9 <sup>d</sup>	0
Ne	0	0.44	0	0.16 <sup>e</sup>	0
Ar	0	1.83	0	0.13 <sup>f</sup>	37
H <sub>2</sub>	0	0.91	0.35	5.2	<0.7
N <sub>2</sub>	0	1.97	0.77	0.9 <sup>d</sup>	<0.7
CO	0.37	2.20	0.59	5.8	5
H <sub>2</sub> O	6.17	1.65	0.66	20 <sup>g</sup>	400 – 800
NH <sub>3</sub>	4.90	2.47	0.32	<11 <sup>h</sup>	>160

<sup>a</sup> Dipole moment  $D$ , Ref. 26; mean polarizability  $\bar{\alpha}$  and anisotropy of polarizability ( $\alpha_{\parallel} - \alpha_{\perp}$ ), Refs. 16 and 27.

<sup>b</sup> Minimum in total scattering cross section  $\sigma_t$ .

<sup>c</sup> Electron energy where  $\sigma_t$  is minimum.

<sup>d</sup> Reference 2.

<sup>e</sup> Reference 18.

<sup>f</sup> Reference 28.

<sup>g</sup> Reference 29.

<sup>h</sup> Reference 30.

electric dipole moment of CO,<sup>26</sup> but there might also be a contribution from the Ramsauer–Townsend or another process. Minima in total scattering cross sections  $\sigma_t$  of several simple molecules are listed with values of their dipole moment,<sup>26</sup> mean polarizability and anisotropy of polarizability<sup>16,27</sup> in Table III. For the nonpolar molecules the listed values of  $\sigma_t$  correspond to  $\sigma_m$ ,<sup>2,18,28</sup> but the minima for the polar molecules CO, H<sub>2</sub>O,<sup>29</sup> and NH<sub>3</sub><sup>30</sup> include inelastic modes. The lowest rotational excitation energy of CO is 40 yJ,<sup>31</sup> which is well below the scattering minimum.

As for nitrogen,<sup>2</sup> we encourage a new theoretical approach to the interpretation of scattering of very low energy electrons by carbon monoxide molecules.

## ACKNOWLEDGMENTS

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Oct./90 to be put forward to the Conférence Générale des Poids et Mesures for adoption in Oct./91:  $10^{-21}$ , zepto, z;  $10^{21}$ , zetta, Z;  $10^{-24}$ , yocto, y;  $10^{24}$ , yotta, Y.

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