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Very low energy electron scattering in CO

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Abstract. Cross sections are reported for backward scattering of electrons by CO for electron energies in the range 2–160 meV. At very low energies, scattering is dominated by rotationally inelastic events and a set of rotationally inelastic cross sections is presented, parametrized in terms of an energy-dependent effective dipole moment for CO. A new value for the scattering length of 0.20 ± 0.03 Å is also estimated.

1. Introduction

The work presented here concerns the measurement of electron scattering cross sections for CO over the energy range 2–160 meV. There have been a large number of electron scattering studies of CO, the great majority of which, however, involve considerably higher energies. For example there have been many experimental and theoretical studies of the shape resonance at around 2 eV. The energy regime for electron scattering below 100 meV has been very little studied, due to technical problems encountered in controlling electron beams in this energy range. Earlier work on electron scattering by CO is partly reviewed in Jain and Norcross (1992), hereafter JN92, to which the reader may turn for an extensive list of references both to experiment and theory. Referring only to low-energy data, electron beam transmission studies are reported in Jordan and Burrow (1978) for energies $\geqslant 250$ meV and a crossed beam experiment is reported in Sohn *et al* (1985) at an energy of 165 meV. Swarm data, measuring momentum transfer cross sections down to energies of a few meV, are reported in Hake and Phelps (1967) and Haddad and Milloy (1984).

The high level of activity in the field of electron scattering with CO, reported in JN92, reflects its importance in a number of plasma applications, for example in electronic device technology and in plasma deposition. Electron scattering by CO is also of considerable significance in astrophysics, since CO is the most abundant molecule in molecular clouds after H_2. In astrophysical plasmas, rotational excitation of CO by photoelectrons, released from grains, and subsequent rotational emission is an important element in the energy balance in a variety of environments e.g. in molecular clouds, at cloud edges and in shocks. In these astrophysical applications, models of the environment require rotationally inelastic state-to-state cross sections as a function of electron energy in the 10 K to a few hundred K energy range (1 meV to tens of meV). Because of the interest in such data, we have sought to extract state-to-state cross sections from our experimental data. This entails a considerable

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degree of data manipulation with accompanying random and possibly systematic errors. However, the inelastic cross sections reported here represent the only experimentally based data presently available, and differ very considerably from theoretical estimates at low energies.

Our experimental results were obtained using a magnetically collimated electron-beam transmission technique which allows us to investigate very low electron kinetic energy. These are the first beam experiments to probe the energy regime in CO in which rotationally inelastic scattering constitutes a significant fraction of the scattering cross section, that is below a few tens of meV. Experiments involving more polar target molecules, e.g. CF_3Cl and CF_2Cl_2, have shown that there is very strong threshold rotational excitation in the few meV kinetic energy range (Randell *et al* 1993). Many other species, with a wide variety of dipole moments, show similar behaviour (work in preparation). These results are in accord with the predictions of long-standing theories of rotational excitation of polar molecules at low electron kinetic energy, reviewed in Shimamura and Takayanagi (1984), hereafter ST84.

2. Experimental details

The experimental system has been described in detail in Field *et al* (1984) with further modifications described in Ziesel *et al* (1993) and Randell *et al* (1994), hereafter R94. Electrons are formed by photoionization of Ar at the autoionizing resonance at 786.5 Å (Radler and Berkowitz 1979) close to the $^2P_{3/2}$ threshold, using monochromatized synchrotron radiation on beamline SA61 at SuperACO, LURE. Electrons have an energy spread equal to that of the ionizing beam, which in these experiments was maintained at $\sim$ 5 meV FWHM. The electrons are formed into a beam in a very weak extraction field and collimated in an axial magnetic field of $\sim$ 20 G. The electron beam passes through the target CO gas whose pressure is accurately measured (Baratron capacitance manometer M127A; absolute accuracy better than 0.2%). Doppler motions of the target gas make a negligible contribution to the energy resolution of the experiment at the low energies encountered here. Absolute cross sections are obtained from $\sigma_B = \ln(I_0/I)/Nz$ where I_0 and I are the unattenuated and attenuated electron currents respectively, N is the gas density and z is the effective pathlength of electrons through the gas. Since scattering takes place in the presence of an axial magnetic field, electrons which are forward scattered remain collimated along the direction of the incident beam and the measured cross section, σ_B, represents scattering into the backward hemisphere only. The value of z has been calibrated in R94 using He and has been shown to be 3.9 cm. To investigate the performance of the instrument, absolute electron energies were checked by a series of repeated measurements of the 2.3 eV resonance in N_2, using the third peak of this resonance at 2.442 ± 0.015 eV (Kennerly 1980), recognizing that this value is likely to be reduced by $\sim$ 10 meV because we sample backward scattering (Rohr 1977). For the purpose of data presentation and analysis, we assume that electron energies are dictated by the experimentally applied potentials. We return to the problem of absolute energy calibration in section 4.

Our data for CO are shown in figure 1. Experiments were repeated over the full energy range three times and each data set showed the form displayed in figure 1, which represents the average of these three data sets. In addition experiments were repeated a further five times at 100 meV, 20 meV and 4 meV. These additional checks showed that the cross section at 4 meV was $6 \pm 2\%$ higher than at 20 meV and thus that the weak dip at low energy is a genuine feature of the data, lying outside the random error. The error in the absolute values of backward-scattering cross section is $\pm 5\%$, taking into account errors of

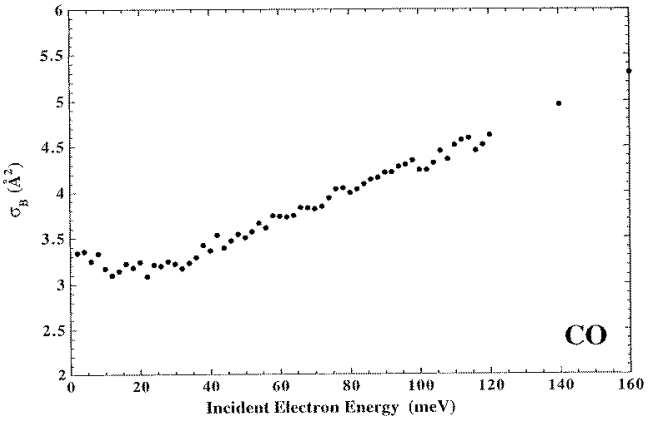

Figure 1. Variation of the backward-scattering cross section, σ_B, for CO at 293 K as a function of electron kinetic energy.

pressure measurement and errors associated with absolute cross section calibration (R94). As an additional check on absolute values, we have compared our data with calculated values of JN92, which were themselves shown to be consistent with the experimental data of Sohn *et al* (1985) at 165 meV. Using data in figure 9 of JN92, the backward-scattering cross section at this energy may be estimated to be 5.0 Å^2. Our value at 160 meV is 5.3 Å^2 and is thus in agreement within experimental error.

3. Data analysis

Application of the standard first-order Born formula (ST84), modified for backward scattering and taking into account the distribution of target molecules over rotational energy levels and the energy spread in the electron beam, shows that the Born approximation overestimates the backward rotationally inelastic scattering cross section at very low energy. For example at 4 meV electron energy, the Born formula predicts an effective rotationally inelastic scattering cross section of 4.34 Å^2 whereas the experimental value of the total backscattering cross section is 3.35 Å^2. Thus the *total* measured cross section falls below the Born values for inelastic scattering at very low energies. It is therefore clear that the Born approximation yields inaccurate values of rotationally inelastic scattering cross sections for CO at low energy. This conclusion is independent of the details of the processes of data reduction which follow below. From here on we refer to calculated cross sections as 'effective cross sections' when these represent results involving weighting over the rotational populations of the target and convolution with the spread of energy in the incident electron beam.

We note at the outset that CO is isoelectronic with N_2 and like N_2 does not show a Ramsauer–Townsend minimum. The weak dip in the backward-scattering cross section in CO between 10 and 20 meV represents the superposition of a falling elastic scattering contribution and a rising rotationally inelastic scattering contribution. As we discuss below, the latter is surprisingly weak, even given the low dipole moment of CO (0.112D (*CRC Handbook of Chemistry and Physics*)). In this connection, molecules without dipole moments such as N_2 or O_2 show cross sections which decrease monotonically with energy below a few tens of meV (R94), showing no evidence of a minimum at very low

energy.

The raw data have been subjected to a sequence of analysis which consists of the following steps. We comment further on these steps below.

(i) Absolute cross sections were obtained using the He calibration described in R94, yielding the values in figure 1.

(ii) Effective Born backward-scattering cross sections were calculated over the higher energy range 40–160 meV. Values, which are a small proportion of the elastic cross section (see below), were then subtracted from the data over this energy range. This yields the variation of the elastic backward-scattering cross section.

(iii) Modified effective range theory (MERT; O'Malley 1963) was used to fit to these data over various energy ranges between 40 and 160 meV to between 90 and 160 meV. Derived MERT parameters did not vary significantly with choice of energy range.

(iv) The MERT parameters were then used to derive the elastic backward-scattering cross sections over the entire energy range of the experiment.

(v) The elastic backward-scattering cross sections were then subtracted from the experimental data over the entire energy range to yield the rotationally inelastic backward-scattering cross section as a function of energy between 2 and 160 meV.

(vi) The predictions of the Born approximation were then compared with the inelastic data obtained in (i)–(v). The Born predictions of the effective cross sections are significantly higher than those found experimentally for the energy range below 15 meV.

(vii) An effective dipole moment was estimated, which falls as the electron energy falls in such a way that use of the Born theory yielded the experimental backward-scattering cross sections obtained above.

(viii) The analytic angular dependence of the Born theory for rotationally inelastic collisions was then used, in conjunction with the effective dipole moments from (vii), to compute the total scattering cross sections for rotationally inelastic collisions for specific state-to-state transitions.

We now consider a number of the above steps in more detail. Turning to (ii), the contributions of the Born dipole and quadrupole cross sections were obtained as follows. The Born dipole cross section in Å^2, for rotationally inelastic scattering into the backward hemisphere, σ_B, may be represented by

$$\sigma_B(J_i \rightarrow J_f) = \frac{4.9406\mu^2}{E_i} f(J_i) \ln\left[\frac{\sqrt{E_i}+\sqrt{E_f}}{\sqrt{E_i+E_f}}\right] \tag{1}$$

where E_i and E_f are the initial and final electron energies in eV, μ is the molecular target dipole moment in Debye (= 0.112 D) and $f(J_i) = (J_i+1)/(2J_i+1)$ for $J_f = J_i+1$, and $f(J_i) = J_i/(2J_i+1)$ for $J_f = J_i-1$ (ST84). The quadrupole contribution to the scattering (Gerjuoy and Stein 1955) was also included since it is more than 10% of the dipole contribution at electron energies greater than $\sim$ 20 meV and is comparable with the dipole contribution at energies above 100 meV. The kinetic energy of the electrons is sufficiently low that the more detailed theory of Dalgarno and Moffett (1963) (see also ST84) is not required. The quadrupole cross section is symmetrical about the forward and backward directions and the backward-scattering cross section may be expressed in Å^2 as

$$\sigma_Q(J_i \rightarrow J_f) = 0.234\,60 Q^2 \left(\frac{E_f}{E_i}\right)^{1/2} g(J_i) \tag{2}$$

where Q is the quadrupole moment, expressed in atomic units (= -1.44 au (Meerts *et al* 1977)) and $g(J_i) = (J_i+1)(J_i+2)/(2J_i+1)(2J_i+3)$ for $J_f = J_i+2$ and

$g(J_i) = J_i(J_i-1)/(2J_i-1)(2J_i+1)$ for $J_f = J_i - 2$. The target molecules are assumed to be distributed about the rotational levels according to the Boltzmann distribution at 293 K. The incident electron beam was assumed to be Gaussian in energy spread about the central energy, with a full width half maximum of 5 meV. The contribution of inelastic scattering at a given energy was obtained, as indicated above, by summing the population weighted cross sections for all energetically open inelastic transitions and convoluting in the effect of the spread of energy in the incident electron beam. For the very lowest energies, the Gaussian was truncated at zero energy. The resulting effective backward-scattering cross sections were then estimated for a series of energy ranges, spanning 40 to 160 meV. Subtracting these cross sections from the experimental results yields sets of values for the purely elastic contribution to the backward scattering for any chosen set of energies lying between 40 and 160 meV. It is important to note at this point that our analysis does not involve a circular argument, in which we use the Born approximation to conclude that it is incorrect to do so. At 40 meV, the contribution of inelastic scattering to the measured cross section lies around 20%. Thus while the Born approximation is not accurate, as we have seen for the lowest energy data, use of Born in the energy range down to 40 meV constitutes the introduction of an error into only a small fraction of the full backward-scattering cross section, which is clearly dominated by elastic scattering above 40 meV. This argument is still stronger if we use the energy range 90–160 meV. It turns out that the choice of energy range has no significant effect on our derived rotationally inelastic scattering cross sections.

Under (iii), the use of MERT can introduce systematic errors into our analysis. As noted for example for N_2, MERT may be inaccurate in fitting elastic data even at very low electron energy (R94). In addition, MERT does not apply to molecules with permanent dipole moments, as discussed for example in Randell *et al* (1993) for the case of electron scattering by chlorofluoromethanes. The use of MERT for molecules with dipole moments is described in Morrison (1987). In the present case, the dipole moment of CO is sufficiently small that the polarization interaction tends to dominate over the electron–dipole interaction, by more than an order of magnitude, for electron–molecule separations of $\geqslant 1$ Å. At all events, MERT yielded a good fit using only two parameters, the scattering length A and the parameter D, which multiplies k^3, where k is the electron wave vector. The relationship between the elastic backward-scattering cross section and partial wave phase shifts, η_ℓ, is given in R94 and we use the following truncated expansions for the MERT expressions for the phase shifts (Lunt *et al* 1994)

$$\tan\eta_0 = -Ak\left[1 + \left(\frac{4\alpha_d}{3}\right)k^2\ln k\right] - \left(\frac{\pi\alpha_d}{3}\right)k^2 + Dk^3 \tag{3a}$$

$$\tan\eta_1 = \left(\frac{\pi\alpha_d}{15}\right)k^2 \tag{3b}$$

$$\tan\eta_2 = \left(\frac{\pi\alpha_d}{105}\right)k^2 \tag{3c}$$

to fit the elastic data for the ranges of energy 40–160 meV to 90–160 meV. α_d, the polarizability of CO, is taken to be 198 Å^3 (Böttcher and Bordewijk 1978). The MERT parameters derived are insensitive to the choice of data set. This shows that our analysis does not depend on the arbitrary choice of energy range over which either the Born approximation or MERT are assumed to be valid. The value of the scattering length is 0.20 ± 0.03 Å, yielding a total scattering cross section at zero energy ($= 4\pi A^2$) of 0.51 ± 0.14 Å^2. The value of D is $44.1 \pm 3.9\,a_0^3$. Errors take into account both the range of values obtained by using different ranges of energy in the MERT fits and the random errors associated

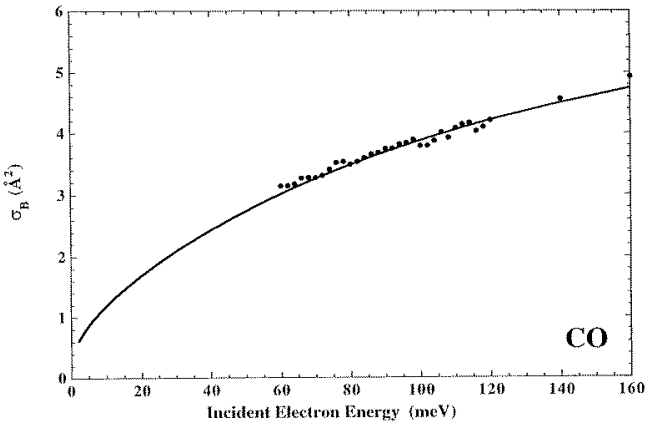

Figure 2. The modified effective range theory (MERT) fit to the elastic part of the backward-scattering cross section (see section 3).

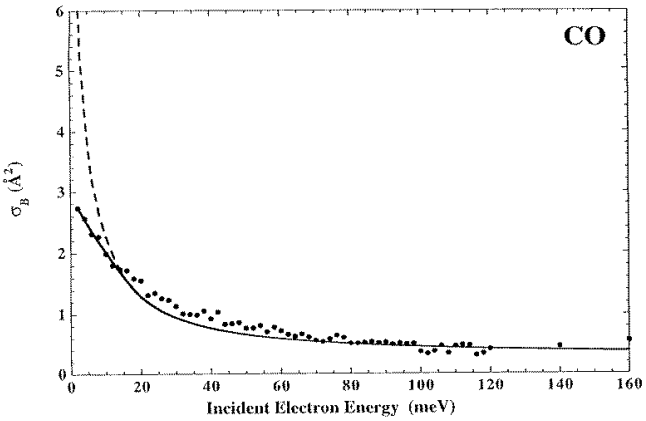

Figure 3. Variation of the backward-scattering effective cross sections for rotationally inelastic scattering of electrons by CO at 293 K. Points are the data derived from experiment. The full curve is the fit to these data obtained using the first Born dipole approximation including quadrupole contributions, and a modified dipole moment, μ_{eff}, for the target at electron energies $\leqslant$ 15 meV (see table 2). The broken curve is the prediction of Born dipole and quadrupole expressions without modification of the dipole moment.

with the MERT fits. The MERT fit to the elastic part of the cross section over the energy range 60–160 meV is shown in figure 2. We note that our value of the scattering length is approximately half that derived by Petitjean *et al* (1984) by extrapolation of theoretical values of Chandra (1977). Turning to (iv) and (v), our values for A and D were then used to estimate the elastic contribution to the backward scattering over the entire range between 2 and 160 meV and thus to give the rotationally inelastic contribution to scattering over this energy range. The variation with electron impact energy of these estimated rotationally inelastic contributions to the backward scattering is shown in figure 3.

Turning to steps (vi) and (vii) of the data analysis, figure 3 also shows Born approximation calculations of effective rotationally inelastic cross sections for comparison with experimental results. The small systematic underestimate by the Born approximation

at energies between $\sim$ 15 and 60 meV may be assumed to lie within the systematic error associated with the procedures used to obtain the experimental rotationally inelastic cross sections. The full curve shows calculated values for which the dipole moment of the target CO has been adjusted so that the Born expression fits the experimental data at very low energies and connects smoothly with the higher energy Born calculations. This adjustment of the dipole moment is most significant at energies below 10 to 15 meV for which strong divergence from Born, shown as a broken curve in figure 3, is apparent. Values of effective dipole moment, μ_{eff}, are shown in the second column of table 1. The virtue of expressing the backward-scattering cross sections in terms of a Born expression with an effective dipole moment is that integral scattering cross sections may now be derived using the analytical angular dependence of the Born cross section. Thus we write

$$\sigma_{\text{T}}(J_i \rightarrow J_f) = \frac{4.9406\mu_{\text{eff}}^2}{E_i} f(J_i) \ln\left[\frac{\sqrt{E_i}+\sqrt{E_f}}{\sqrt{E_i}-\sqrt{E_f}}\right] \tag{4}$$

where σ_{T} is in Å^2 and μ_{eff} is the effective dipole moment of the target CO in Debyes, with other symbols as defined in equation (1). In table 1 representative integral cross sections for various rotational transitions over the energy range 1–20 meV ($\sim$ 10–240 K) are shown. Equation (4) in conjunction with values of μ_{eff} may be used to generate cross sections for the appropriate electron energies and for any values of J_i and J_f.

Table 1. Values of the effective dipole moment of CO, μ_{eff} in Debye, and of integral cross sections, in Å^2, for rotationally inelastic collisions of electrons with CO calculated using equation (4). See section 4 for further details of recommended values of total cross sections.

E (meV)	μ_{eff} (D)	0–1	1–2	2–3	3–4	4–5	1–0	2–1	3–2	4–3	5–4
1	0.0674	40.88	6.23	—	—	—	17.40	16.09	14.59	13.38	12.40
2	0.0723	34.64	15.69	9.16	3.07	—	12.59	12.02	11.10	10.29	9.61
3	0.0781	31.50	15.70	10.99	7.98	5.40	11.04	10.73	10.02	9.36	8.79
4	0.0842	30.21	15.68	11.57	9.13	7.28	10.42	10.26	9.65	9.07	8.56
5	0.0899	29.41	15.63	11.84	9.67	8.09	10.06	9.99	9.46	8.92	8.45
6	0.0945	28.46	15.36	11.83	9.85	8.43	9.68	9.69	9.21	8.72	8.28
7	0.0979	27.27	14.89	11.60	9.77	8.49	9.25	9.31	8.88	8.44	8.03
8	0.1005	26.03	14.34	11.26	9.57	8.40	8.80	8.90	8.52	8.11	7.74
9	0.1027	24.87	13.80	10.90	9.33	8.25	8.39	8.52	8.19	7.81	7.46
10	0.1047	23.84	13.30	10.57	9.09	8.08	8.03	8.19	7.88	7.53	7.21
12	0.1082	22.09	12.44	9.96	8.63	7.73	7.43	7.62	7.36	7.06	6.77
14	0.1108	20.55	11.65	9.38	8.17	7.36	6.90	7.11	6.89	6.62	6.36
16	0.112	18.90	10.77	8.72	7.63	6.90	6.34	6.56	6.37	6.14	5.91
18	0.112	17.21	9.85	8.00	7.03	6.38	5.77	5.99	5.83	5.63	5.43
20	0.112	15.82	9.09	7.40	6.52	5.93	5.30	5.51	5.38	5.20	5.02

4. Discussion and conclusions

There have been three separate attempts to calculate rotationally inelastic scattering cross sections for electrons with CO: Crawford and Dalgarno (1971), Chandra (1977) and JN92. The results of Crawford and Dalgarno and of Chandra, which extend down to 5 meV, agree rather closely and in the forthcoming discussion mention of Chandra may be taken to include Crawford and Dalgarno. Those of JN92 lie 10% to 20% lower, and extend down to energies between 1 and 2 meV. In table 2 we show a comparison between our

experimentally based values and those of JN92 and of Chandra (1977). Under 'present values', cross sections for 30, 50 and 100 meV are simply those derived from the Born approximation with $\mu = 0.112$ D and are included only for comparison with other theoretical results.

Table 2. Values of integral cross sections for rotationally inelastic collisions of electrons with CO in comparison with theoretical values. Numbers in brackets are theoretical values corrected for the effective dipole moment of CO (sections 3 and 4 and table 1).

Electron energy (meV)	Rotational transition	Present values (Å^2)	Crawford and Dalgarno (1971); Chandra (1977) (Å^2)	Jain and Norcross (1992) (Å^2)
	$J = 0$–1			
2		34.6	—	76.0(31.7)
3		31.5	—	60.0(29.2)
4		30.2	—	50.0(28.3)
5		29.4	44.4(28.6)	40.1(25.8)
10		23.8	26.2(22.9)	22.6(19.8)
30		11.4	10.5	8.5
50		7.5	6.7	5.3
100		4.2	3.6	2.7
	$J = 1$–2			
2		15.7	—	33.4(13.9)
3		15.7	—	27.8(13.5)
4		15.7		24.0(13.6)
5		15.6	24.9(16.0)	20.8(13.2)
10		13.3	15.0(13.1)	12.9(11.3)
30		6.6	6.1	4.8
50		4.4	3.9	3.0
100		2.5	2.1	1.5

There are very considerable difficulties in performing scattering calculations since the CO target requires many configurations to reproduce even the correct sign of the dipole moment (see, for example, Cooper and Kirby (1987)). In the most recent work of JN92, a near Hartree–Fock limit is used for the CO wavefunction, which yields the well known result that the dipole is reversed from that determined experimentally (and is calculated to be 0.25 D). In order to correct for this error JN92 adopted the pragmatic approach of using experimental values of the dipole and quadrupole moments in calculating cross sections for rotationally inelastic scattering. In summary, we recommend that at $\leqslant 15$ meV (175 K) our values of cross section computed using Born and D_{eff} should be adopted and that above 15 meV, for $J = 0$–1 and $J = 1$–2, either the values of Chandra or of JN92 should be used. For scattering involving higher rotational states at energies above 15 meV, the Born approximation (equation (4)) should be adopted.

Theoretical results diverge strongly from our results at low energies. If, however, theoretical values are multiplied by $(\mu_{\text{eff}}/\mu)^2$, this brings values into agreement with our present data to between 10% and 20% in all cases for which comparison can be made. This simple adjustment and the agreement it yields shows that values at very low energies are effectively dominated by the first Born approximation, but one in which the target wavefunction is modified by the incoming electron to yield dipole moments which decrease with lower electron impact energies. One may speculate that the incoming electron perturbs the CO target, effectively mixing in some small fraction of excited states. If these states are

dominated by the lowest-lying singlet state $A\,^1\Pi$, which has the largest transition moment to the ground state (Kirby and Cooper 1989), then this will cause the effective dipole moment of the target CO to fall; Cooper and Kirby (1987) estimate that the dipole moment of the $A\,^1\Pi$ state for the equilibrium bond length of CO in the ground state is effectively zero. Lower-lying triplet states, $^3\Pi$ (dipole moment = 1.374 D C^+O^-), $^3\Sigma^+$ (dipole moment = 1.06 D C^-O^+), $^3\Delta$ and $^3\Sigma^-$ (Huber and Herzberg 1978), may also intervene.

Our lack of knowledge of absolute values for the electron energy introduces an additional source of uncertainty into our results. We have no firm estimate of the accuracy with which the absolute potentials applied to elements in the apparatus determine the electron energy. In related work on He, H_2, N_2 and O_2 (R94), the presence of O_2 may cause a displacement of 10 meV to higher energy, whereas data for He and H_2 show that the electron energy is indeed determined by applied potentials down to 5 and 10 meV respectively. A number of analyses have been performed with our present data introducing displacements to higher energy of between 4 and 8 meV into the electron energy scale. Such displacements modify MERT parameters somewhat, yielding a set of rotationally inelastic effective cross sections similar to those in figure 3 but shifted to higher energy. Such data would suggest that the first Born approximation strongly underestimates rotational scattering in the tens of meV energy range and would also yield poor agreement between theoretical values in table 2 and experimentally derived data. Whilst these conclusions cannot be ruled out, we have chosen not to tamper with the electron energy scale, but to assume potentials do, in fact, directly determine electron energies.

To summarize, equation (4) used in conjunction with the proposed values of $\mu_{\rm eff}$ in table 1 and combined with results in JN92, Chandra and Crawford and Dalgarno and Born calculations, form the best available data set for rotationally inelastic cross sections for electron collisions with CO in the range of energy of interest in many astrophysical problems and also for the low electron energy tail in CO containing plasmas. Noting as we have above that there may be additional systematic errors, the random error for cross sections below about 20 meV electron energy may be estimated to lie between $\pm$10 to 20%. Our results also confirm that CO shows no Ramsauer–Townsend minimum and data also yield a scattering length which is about half of the previously reported value.

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