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Absolute total cross sections for electron—CO₂ scattering at energies from 0.5 to 3000 eV

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Abstract. Employing two different experimental techniques, absolute total cross sections for e^--CO_2 scattering have been measured over a very wide energy range between 0.5 and 3000 eV. The cross section function exhibits a sharp increase below 1.5 eV and a resonant maximum around 3.8 eV. It then rises again up to a broad maximum near 30 eV and then decreases slowly. A very broad feature was observed centred at an energy of about 300 eV and extending from about 200 to 600 eV. Comparison is made with other available measurements and calculations.

1. Introduction

Electron- CO_2 interaction has been the subject of systematic studies since the early experiments of Brüche (1927). However, only since the 1960s has the number of investigations considerably increased, mainly due to the practical importance of the CO_2 molecule in processes occurring in planetary atmospheres and in energy-related technology. The CO_2 molecule also plays a role as a probe in testing new theoretical approaches aimed at explaining the physical features of electron-polyatomic-molecules scattering phenomena.

Most of the recent measurements of the electron total cross section in CO₂ have been performed using the time of flight (TOF) technique. They cover the energy range from a few hundredths of meV (Ferch et al 1981), through low energies (Hoffman et al 1982, Sueoka and Mori 1984), up to a few hundreds of eV (Kwan et al 1983, Sueoka and Mori 1984). These results are generally different in magnitude from those obtained earlier using the Ramsauer technique (Brüche 1927, Ramsauer and Kollath 1930) and from the normalised values of Szmytkowski and Zubek (1978), who employed a linear transmission method. In spite of the stated increased precision in the new measurements, there are still significant discrepancies to be found among them. In general, however, all results agree in the shape of the total cross section function. Table 1 gives

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a summary of the bibliography on CO₂ cross sections along with the energy coverage of each work.

In this paper we present the results of our electron-CO₂ total cross section measurements for incident energies varying from 0.5 to 3000 eV. These measurements were performed using two different techniques. A linear transmission method (Gdańsk) was employed for energies below 80 eV, and a modified Ramsauer technique (Trento) was used for energies above 60 eV. This work is a continuation of an earlier collaboration (Zecca et al 1986) between both laboratories.

2. Experimental

Both sets of apparatus used in the present experiments have already been used for total cross section measurements (Szmytkowski and Zubek 1978, Dalba et al 1979, 1980, Szmytkowski et al 1984, Szmytkowski and Maciąg 1986 a, b, Zecca et al 1987a, b), and since detailed descriptions of the experimental systems and procedures have been given in earlier publications (Dalba et al 1981, Szmytkowski et al 1984) only a short description will be given here.

The apparatus for low-energy measurements consists of an electron gun followed by a 127° electrostatic cylindrical deflector and electron lenses. The electron beam, with an energy spread of 80 meV, was directed into a collision chamber 30.5 mm long containing the gas under study. The electrons which escaped from the scattering volume through the exit orifice of the chamber were collected by a Faraday cup. The absolute energy scale was calibrated against the oscillatory resonant structure in the transmission current at around 2.3 eV in molecular nitrogen (Kennerly 1980). The target absolute gas pressure was measured with a capacitance manometer. Its head was kept at a constant temperature of 322 K whilst the gas cell temperature was about 10 K lower. This temperature difference was accounted for by applying the thermal transpiration correction (Knudsen 1910). To reduce the target-gas influence on the incident-electron beam intensity, the background pressure outside the scattering chamber was at the same value regardless of the presence or absence of gas in the scattering chamber.

The experimental procedure was based on the relation between electron current attenuation and the total cross section as given by the de Beer-Lambert relation (Bederson and Kieffer 1971). The final cross section at a particular energy is the weighted mean of the cross sections obtained in a series of runs. These runs were performed under different voltage settings on the electron optics and employing a range of target-gas pressures.

The Ramsauer-type apparatus, with a 200 mm electron trajectory radius, was employed for intermediate- and high-energy electron total cross section measurements. In this method, the total cross section $\sigma(E)$ at a given energy E is evaluated by the attenuation formula

$$I_{c1}/(I_{c1}+I_{s1}) = [I_{c2}/(I_{c2}+I_{s2})] \exp[-\sigma(E)L(N_1-N_2)]$$

where $I_{c1(2)}$ are the transmitted beam intensities measured with the collector and $I_{s1(2)}$ are the intensities of the scattered electrons which reach the scattering chamber walls. N_1 and N_2 are the gas densities in the interaction volume corresponding to two different pressures at which $I_{1(2)}$ currents are taken. The absolute pressure of the sample gas

was measured with a capacitance manometer. The head temperature was maintained at 292 K, while the gas chamber temperature was generally within ± 2 K of this value. The evaluated path length L of electrons in the interaction volume was 140.2 mm. The energy width (fwhm) of the electron beam was about 0.5 eV. The improved angular resolution of the present apparatus, compared with the original Ramsauer (1921) design, essentially reduced the systematic lowering of the measured cross sections. Differential pumping and the use of a diverter valve (Basta et al 1976) ensured a constant background pressure in the volume outside the scattering chamber. The experiment was controlled by a microcomputer.

Measurements at a given energy E were carried out at different sets of pressures in the scattering region and repeated several times. The final values of the total cross section are the averages of all values taken at the same energy.

The independence (within statistical uncertainties) of the measured total cross section from the gas pressure in the scattering chamber, as well as from the electron beam intensity, was established at different electron impact energies for both experiments.

As is well known, the lack of discrimination between the unscattered transmitted electrons and electrons scattered at small angles leads to an underestimation of the measured cross sections. In order to estimate this effect we used all available (Danner 1970, Bromberg 1974, Shyn and Sharp 1979, Register et al 1980, Iga et al 1984, Kochem et al 1985, Antoni et al 1986) elastic and inelastic differential cross sections for CO_2 . For the sets of apparatus used in these measurements, with angular acceptances of the detector of 2×10^{-3} sr (Gdańsk) and 3.4×10^{-4} sr (Trento), respectively, we found the small-angle contribution to be below 0.5% even for the highest energies used.

Errors in the energy scale were evaluated with respect to their effect on the magnitude of the measured cross sections. Below 1 eV the associated uncertainty did not exceed 7% and in the 3.8 eV resonance range it was about 5%, diminishing to below 0.5% for higher energies where the cross section is a very slowly varying function of electron energy.

 \cdot A relatively large uncertainty of up to 1.5% resulted from the target gas pressure measurements.

Gas samples with 99.99% purity were used directly from the tank without further purification.

Uncertainties in the sample-gas cell length, end effects and uncertainty in the gas temperature resulted in uncertainties in the total cross section of 0.3%, 0.4% and 0.4%, respectively, for both apparatuses.

Using these values for the systematic uncertainties, if the errors are considered to be uncorrelated and are added in quadrature, we obtain a total systematic error of about 8% for the lowest energies and almost 6% around the resonance, decreasing to 2% for energies above 20 eV.

The essential characteristic of the Ramsauer technique is the measurement of the electron current on the gas chamber walls. At the lowest energies (60-100 eV) attainable with the present apparatus, any random disturbances in the experimental system lead to an increase in this current and therefore to an overestimation of the cross section. This effect could have been the source of an additional systematic error in the apparatus used at Trento for this energy interval.

The statistical error was of the order of 4% below 1 eV, 3% in the resonance region, decreasing to 2% in the 6--50 eV energy range, and increasing again steadily to 6% above 1000 eV.

3. Results and discussion

The total cross sections in the energy range from 0.5 up to 3000 eV are shown graphically in figure 1 along with data from other laboratories, and the present numerical data are set out in table 1.

According to the observed features in the measured total cross section function, the whole energy range can be divided into three regions.

In the first, ranging from 0.5 to about 2 eV, the cross section increases rapidly with decreasing collision energy. This behaviour has recently been very intensively studied both experimentally (Ferch et al 1981, Kochem et al 1985) and theoretically (Morrison et al 1976, 1977, Morrison and Lane 1977, 1979, Lynch et al 1979, Collins and Morrison 1982, Morrison 1982, Whitten and Lane 1982, Estrada and Domcke 1985) and was eventually explained by the existence of an $e^- + CO_2$ virtual state. In this energy region our results are in close agreement with recent TOF measurements of Ferch et al (1981). The agreement worsens steadily with increasing energy up to 15% at 1.5 eV. Recent calculations in this energy interval are compared with present data in figure 2. Agreement with the theoretical investigations of Morrison et al (1976, 1977), Morrison and Lane (1977) and Collins and Morrison (1982) is good, while a large disagreement in magnitude with the calculations of Lynch et al (1979) is clearly visible for low energies.

The energy region between 2 and 6 eV is characterised by the well known maximum centred at 3.8 eV. It was assigned (Claydon et al 1970) to the Π_u CO₂ shape resonant

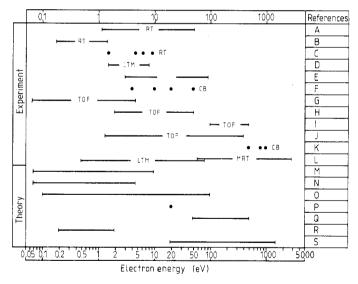


Figure 1. Historical diagram showing experimental and theoretical energy ranges covered in e⁻-CO₂ scattering. Both experiments (top of figure) and theories (bottom) are in chronological order. RT, Ramsauer technique; LTM, linear transmission method; CB, crossed beams; TOF, time of flight; MRT, modified Ramsauer technique. References: A, Brüche (1927); B, Ramsauer and Kollath (1930); C, Ramsauer and Kollath (1932); D, Szmytkowski and Zubek (1978); E, Shyn et al (1978); F, Register et al (1980); G, Ferch et al (1981); H, Hoffman et al (1982); I, Kwan et al (1983); J, Sueoka and Mori (1984); K, Iga et al (1984); L, Present results; M, Morrison et al (1977); N, Morrison and Lane (1977); O, Lynch et al (1979); P, Onda and Truhlar (1979); Q, Jain and Tayal; R, Collins and Morrison (1982); S, Botelho et al (1984).

Table 1. Present experimental results. Total absolute cross section $\sigma(10^{-20}\,\mathrm{m}^2)$ against electron energy $E(\mathrm{eV})$. The Gdańsk measurements extend from 0.5 to 80 eV, the Trento measurements from 72.2 to 2916 eV.

E	σ	Ε	σ	E	σ	Ε	σ
0.5	15.8	3.8	16.7	12	13.2	144	11.03
0.6	13.7	3.9	16.1	13.5	13.6	169	9.93
0.7	11.9	3.95	15.2	15	13.9	196	9.25
0.8	10.2	4.0	14.2	17.5	14.9	225	8.71
0.9	9.15	4.1	13.0	20	15.8	256	8.31
1.0	8.35	4.2	12.4	22.5	15.8	289	7.84
1.1	7.63	4.3	11.4	25	16.0	324	7.31
1.2	7.10	4.4	11.1	27.5	16.2	361	6.79
1.3	6.72	4.5	10.4	30	16.2	400	6.40
1.4	6.34	4.6	9.60	35	16.1	441	6.12
1.5	6.12	4.7	9.30	40	15.9	484	5.58
1.6	6.07	4.8	8.66	45	15.8	576	4.93
1.8	5.89	5.0	8.14	50	15.1	676	4.41
2.0	5.90	5.2	7.78	60	14.9	784	3.94
2.2	6.11	5.4	7.70	65	14.4	900	3.53
2.4	6.73	5.6	8.10	70	13.9	1024	3.28
2.6	7.23	5.8	8.10	80	13.4	1156	2.95
2.8	7.60	5.9	8.10			1296	2.69
3.0	8.60	6.0	8.30			1444	2.38
3.1	9.10	6.5	8.42			1600	2.13
3.2	9.60	7.0	9.01	72.2	14.40	1764	1.92
3.3	11.1	7.5	9.74	81	13.81	1936	1.79
3.4	12.8	8.0	9.83	90.2	13.39	2116	1.62
3.5	13.6	8.5	10.5	100	12.54	2304	1.50
3.6	14.8	9.0	11.1	110.2	12.14	2500	1.29
3.65	15.4	9.5	11.6	121	11.65	2704	1.27
3.7	16.0	10	12.0	132.2	11.17	2916	1.15

state. In vibrational excitation functions (Danner 1970, Boness and Schulz 1974, Čadež et al 1974, 1975), in threshold spectra (Cvejanović et al 1985) and in transmitted electron currents (Bonness and Hasted 1966, Burrow and Sanche 1972, Sanche and Schulz 1973, Szmytkowski and Zubek 1978) some oscillatory structure superimposed on the resonant maximum was distinguishable. For the CO₂ molecule these oscillations, qualitatively explained by the 'boomerang' effect (see the review by Lane 1980, and references therein), are however too small to be visible in the total cross section curve, as is the case for N₂ (Golden 1966). In the resonance region the discrepancies between different measurements are quite severe. The present results are about 7% lower at the maximum than those of Hoffman et al (1982) but are almost 20% higher than the measurements of Ferch et al (1981) and 30% higher than those of Sueoka and Mori (1984). The disagreement with the latter measurements can be probably ascribed to the better energy resolution of the Gdańsk apparatus: about 0.05 eV ғwнм, compared with 0.30 eV obtained by Sueoka and Mori (1984) and 0.25 eV obtained by Ferch et al (1981). The larger value measured by Hoffman et al cannot be explained by this argument. Their measurements are generally higher than the present ones in the overlap range. Anyway, the discrepancy is still within the combined errors (see Hoffman et al 1982, table 1). The old measurements of Brüche (1927) and Ramsauer and Kollath (1932) are not reported in figure 2 for clarity. It is worth mentioning that the data in

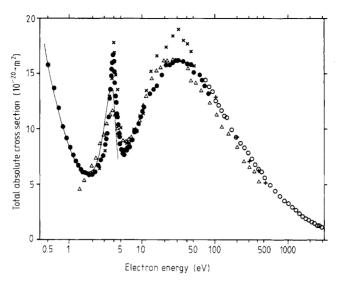


Figure 2. Experimental total cross sections for e^-CO_2 scattering: ——, Ferch *et al* (1981); ×, Hoffman *et al* (1982); +, Kwan *et al* (1983); Δ , Sueoka and Mori (1984); \bullet , \bigcirc , present results.

the latter paper are generally higher than the present ones by some 10%, while Brüche's data are some 20% lower than ours.

The theoretical calculations of Morrison et al (1976, 1977), Morrison and Lane (1977) and Lynch et al (1979) give results (see figure 2) which well reproduce the general tendency of the experimental cross section in this energy range. However, they are much higher than the experimental data due to the neglect of the nuclear vibrational motion.

In the third, the widest energy region, the total cross section first increases and reaches its maximum value near 30 eV and then slowly decreases with increasing energy. Numerous processes are energetically accessible for these energies and make detailed analysis a very difficult task. However, the calculations of Lynch et al (1979) predicted the existence of new high-energy short-lived shape resonances. Two of them, very weak ones, centred near 11 and 30 eV, were shortly after confirmed by the experiment of Tronc et al (1979). It might be the case that some change of the slope in the present cross section curve between 8 and 20 eV, similar to that observed by Hoffman et al (1982), is related to such resonances. Above threshold, ionisation phenomena also play an important role in the scattering process, and near 120 eV (around the maximum of the total ionisation cross section) they give a contribution of almost 30% (Rapp and Englander-Golden 1965) to the total cross section. This contribution rises to slightly more than 40% at 1000 eV.

In figure 3 a very broad feature centred at an energy of 300 eV is hardly visible extending from about 200 to 600 eV. The visibility can be improved by just plotting the same data with a different scale.

In order to have a better visual presentation, we used the following procedure:

- (i) a least-square polynomial fit was made on all measured points excluding those in the energy range from 150 to 800 eV; and
 - (ii) the best-fit function was subtracted from all the measured points.

The resulting plot is shown in figure 4.

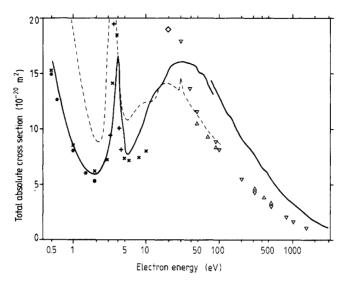


Figure 3. Comparison of calculated e^--CO_2 cross sections with the present experiments: \times , Morrison *et al* (1977), elastic; +, Morrison and Lane (1977), rotational; - -, Lynch *et al* (1979), elastic; \diamondsuit , Onda and Truhlar (1979), rotational; * , Collins and Morrison (1982), rotational; Δ , Jain and Tayal (1982), elastic; ∇ , Botelho *et al* (1984), elastic; \longrightarrow , present experimental (the line was drawn only as a guide for the eye).

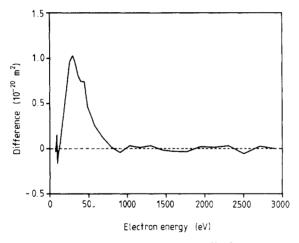


Figure 4. Difference (in absolute scale, 10^{-20} m²) between the measured cross section and the least-square polynomial fit (of the seventh degree over the first seven points and the last thirteen) against electron energy (eV). See text.

The wide peak centred at 300 eV is not dependent on the detailed choice of the points used in this fit, whereas the noise both inside and outside the 150-800 eV range changes strongly according to the detailed choice made.

The existence of this peak was checked by measuring several times up to 25 points in the energy range from 100 to 1000 eV. These points are not reported in figure 2 and table 1 since the measurements were performed in arbitrary units and not on an absolute cross section scale. All the measurements confirm the existence of the broad structure shown in figure 4. The structure is responsible for about 7% of the cross section at

300 eV. The random noise is of the order of $\pm 2\%$. Careful checks were made in order to ascertain that the peak was not produced by electron optical effects. No contaminant is known capable of producing such a peak in the cross section and indeed no physical explanation has been found until now for this structure. Below 100 eV the present results generally lie below the results of Hoffman et al (1982), agree well with the measurements of Sueoka and Mori (1984), and are more than 10% higher than the measurements of Brüche (1927). Above 100 eV there is good agreement with the TOF measurements of Kwan et al (1983) but, in turn, the results of Sueoka and Mori (1984) are a few per cent lower than the present data. The discrepancy of about 5% between the two sets of present measurements in the overlap range from 60 to 100 eV can be related to the 'low-energy' systematic error of the Ramsauer apparatus mentioned earlier. For energies above 500 eV no total cross section measurements were available for comparison. Recently, Iga et al (1984) have derived integral elastic cross sections from a crossed-beam experiment at energies of 500, 800 and 1000 eV. These cross sections could be regarded as a lower bound for the total ones. However, if one combines the results of Iga et al (1984) with the total ionisation cross sections of Rapp and Englander-Golden (1965), then one obtains quite satisfactory agreement between such summed cross sections and the present data. The elastic cross sections measured by Shyn et al (1978) are generally higher than the present total cross section below 50 eV and become lower for higher energies. Since the experimental vibrational cross section near the 3.8 eV resonance (Danner 1970, Spence et al 1972, Register et al 1980) and the experimental ionisation cross section above 50 eV (Rapp and Englander-Golden 1965) constitute at least 40% and 20% of the total, respectively, this suggests a substantial error in the normalisation procedure followed by Shyn. The rotationally summed cross section calculated by Onda and Truhlar (1979) at an electron impact energy at 20 eV (see figure 2) is much higher than the experimental data. Similar behaviour is shown by the low-energy part of the calculations of Botelho et al (1984). The results of elastic cross section calculations at higher energies (Jain and Tayal 1982, Botelho et al 1984) are more encouraging.

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