Total cross sections for electron scattering by CO_2 molecules in the energy range 400–5000 eV

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Total cross sections for electron scattering by CO_2 molecules in the energy range 400–5000 eV have been measured with experimental errors of $\sim 3\%$. The present results have been compared with available experimental and theoretical data. The dependence of the total cross sections on electron energy shows an asymptotic behavior with increasing energies, in agreement with the Born-Bethe approximation. In addition, an analytical formula is provided to extrapolate total cross sections to higher energies.

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I. INTRODUCTION

In the last few years, a large number of theoretical and experimental works devoted to the study of total cross sections (σ_T) for electron scattering by atoms and molecules has been published. To have accurate values for a wide range of energies will be useful for many scientific and technological applications. Recently, Jain and Baluja [1] have summarized the results available in the literature for an important number of molecules. As can be seen in Ref. [1], experimental data for impact energies above 500 eV are scarce. However, as pointed out by Zecca *et al.* [2] in a recent work, this energy range has a special interest to search for systematic relations between total cross sections and other molecular parameters. Moreover, theoretical approximations, required in high-energy electron-molecule collisions [1], need accurate experimental values in order to compare results.

For electron-CO_2 collisions, most of the previous measurements of σ_T [3–5] were performed at energies below 500 eV. We have only found results for energies above 500 eV in the work of Szmytkowski *et al.* [6]. These measurements were carried our for impact energies of up to 2916 eV by using a modified Ramsauer technique [7].

Concerning theoretical data, recent calculations have been made by Jain and Baluja [1] in the energy range from 10 to 5000 eV by using a complex optical model potential. More recently, Joshipura and Patel [8] have published theoretical results in the 100–1000-eV energy range, obtained by applying an addition rule to the cross sections of the constituent atoms. Discrepancies between theoretical and experimental values at high energies are still of the order of 30%. These discrepancies have prompted the present experimental work.

In this work values of total cross sections for e-CO_2 scattering in the 400–5000-eV energy range are given. The measurements have been performed by using a transmission-beam technique, and the estimated experimental errors are of approximately 3%. A detailed error source analysis has been made, paying special attention to those arising from forward scattered electrons. The dependence of σ_T on electron energy has been compared with theoretical predictions and especially with the energy dependence derived from the Born-Bethe approximation [9–11]. In addition, a simple formula to extrapolate cross-section values to higher energies has been obtained by assuming an asymptotic behavior of σ_T as a function of the energy according to the Born-Bethe theory.

II. EXPERIMENT

A. Experimental setup

The experimental setup is similar to that described in previous work [12,13] and will only be briefly mentioned here. An electron beam 1 mm in diameter is generated by an electron gun operating at typical currents of 10^{-13} A. Pressure in the gun was maintained less than 10^{-5} Torr during the measurements. The collision chamber was defined by two apertures 1 mm in diameter, separated by a distance (L) that can be changed from 70 to 127 mm according to the experimental requirements. The gas pressure in the chamber was measured with an absolute capacitance manometer (MKS Baratron 127 A). The energy of the emerging electrons from the gas cell was analyzed by an electrostatic hemispherical analyzer (see Ref. [13] for details). Transmitted electrons were finally detected by a channeltron electron multiplier operating in single-pulse mode. The energy resolution was better than 1 eV (full width at half maximum) for incident energies ranging from 400 to 5000 eV. The pressure in the region of the energy analyzer and detector was maintained less than 10^{-5} Torr during the measurements.

B. Procedure

The method is based on the measurement of the electron-beam attenuation through the gas cell. The recorded beam intensity (I) follows the law

$$I = I_0 \exp(-nL\sigma_T), \tag{1}$$

where I_0 is the intensity of the primary beam, L is the interaction region length, n is the molecular density, and σ_T is the total cross section; n was obtained from the measurement of pressure and temperature in the gas cell. Each measurement was made at gas pressures ranging from 2 to 70 mTorr.

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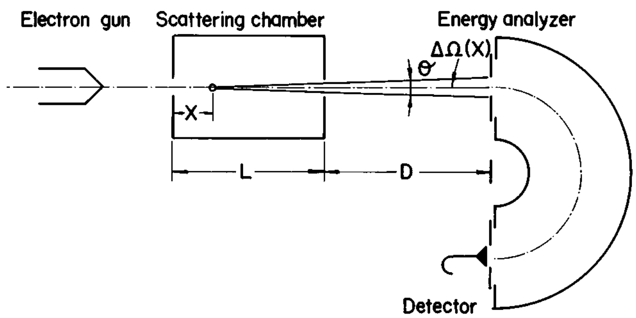

FIG. 1. A sketch of the scattering geometry of the experimental setup.

C. Error source analysis

The accuracy of the pressure measurements was assumed to be better than 1%, as stated by the manufacturer of the MKS-Baratron 127 A. To ensure that pressure gradients did not contribute to the experimental errors, the pressure was measured at several points along the cell. Measurements have been carried out at electron currents ranging from 10^{-13} to 10^{-15} A. For this current range no dependence of σ_T on the electron intensity has been found. Each measurement was repeated at least five times for the same experimental conditions, so assuring statistical uncertainties less than 2%. The length of the collision chamber was changed from 70 to 127 mm, and the measured σ_T values were found to be in agreement within the statistical uncertainties (namely, 2%). This result indicates that our measured length (L) corresponds to the actual absorption length and that possible multiscattering effects are negligible for our experimental conditions.

Special attention was paid to avoid errors arising from forward electron scattering. As has been pointed out by several authors [14,15], electrons scattered in the forward direction can be the main error source at high impact energies if they are not efficiently discriminated.

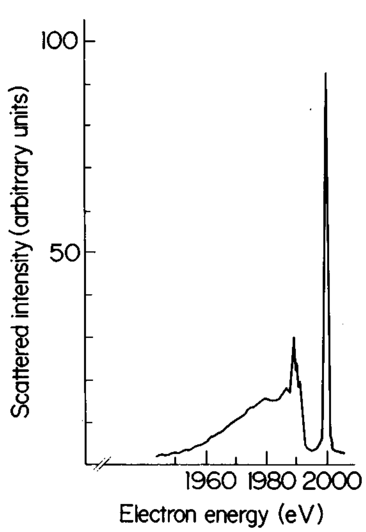

FIG. 2. Energy-loss spectrum of CO_2 for 2000-eV incident electron energy.

Equation (1) represents the ideal case, in which the beam is infinitely narrow and the solid angle subtended by the detector is zero. In Ref. [14], the small-angle scattering contribution was incorporated into Eq. (1), giving the expression:

$$I=I_0\exp\left[-nL\sigma_T+n\int_0^L dx\int_0^{\Delta\Omega(x)}\left(\frac{d\sigma}{d\Omega}\right)d\Omega\right], \qquad (2)$$

where the corrective term represents the effect of colliding electrons, at a distance X from the entrance of the gas cell, which are scattered into the solid angle subtended by the detector $[\Delta\Omega(x)]$, and $(d\sigma/d\Omega)$ represents the sum of the elastic and inelastic differential cross sections. A sketch of the scattering geometry used in the present experiment is shown in Fig. 1. In our experimental conditions, the maximum acceptance angle of the detector is defined by the distance D (150 mm) between the end of the collision chamber and the entrance aperture of the energy analyzer.

The inelastic part of the corrective term in Eq. (2) is fully suppressed by the energy analyzer, Figure 2 shows a typical energy loss spectrum for 2000-eV electrons and 75 mTorr of CO_2 in the collision chamber. As may be seen in this figure, the energy resolution of the analyzer allows one to eliminate the contribution of forward inelastic collisions.

The contribution of elastic scattering in the forward direction can be minimized by reducing the angular acceptance of the analyzer. For the energy range considered, the angular distribution of the elastically scattered electrons has a maximum for small θ angles [14]. For this reason, the elastic part of the integral expression in Eq. (2), satisfies [14] the inequality

$$\int_0^L dx\int_0^{\Delta\Omega(x)}\left(\frac{d\sigma}{d\Omega}\right)_{\text{elastic}} d\Omega\leqslant L\Delta\Omega\left(\frac{d\sigma}{d\Omega}\right)_{\substack{\text{elastic}\\ \theta=0}}. \qquad (3)$$

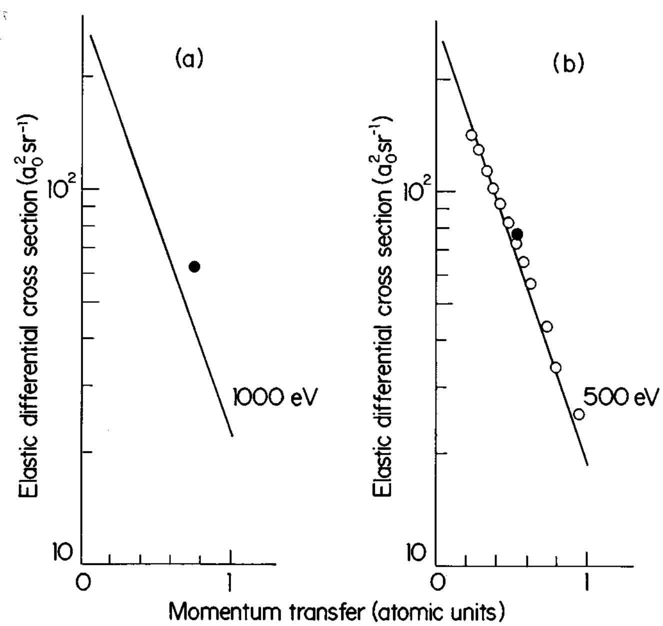

FIG. 3. Elastic differential cross sections for e-CO_2 scattering vs momentum transfer at (a) 1000 eV and (b) 500 eV. ○: experimental results of Bromberg [16]. ●: measurements of Iga, Nogueira, and Mu-Tao [17]. ——: calculations of Botelho *et al.* [18].

Differential elastic cross sections $[(d\sigma/d\Omega)_{\mathrm{elastic}}]$ for CO_2 have been measured by Bromberg [16] at 300, 400, and 500 eV for scattering angles ranging from 2° to 40°. More recent measurements were published by Iga, Nogueira, and Mu-Tao [17] for 500-, 800-, and 1000-eV impact energies. Theoretical data at these energies have been calculated by Botelho *et al.* [18]. There is good agreement between these theoretical and experimental data and they confirm the behavior observed by Bromberg [16] for small values of momentum transfer. Figure 3 is a semilogarithmic plot of the differential elastic cross section given in Refs. [16–18] against momentum transfer (ΔP) for $\Delta P \leqslant 1$. As seen from this figure, the curve shows a linear behavior in the region of small ΔP. As proposed by Bromberg [16], this means that the cross sections in that region can be described by the expression

$$\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{elastic}} = \left(\frac{d\sigma}{d\Omega}\right)_{\substack{\mathrm{elastic}\\ \Delta P=0}} \exp(-\beta\Delta P), \tag{4}$$

where $(d\sigma/d\Omega)_{\mathrm{elastic}}$ is the differential elastic cross section as a function of momentum transfer and β is the slope of the straight line shown in Fig. 3. A linear fit of the available data from Refs. [15–17] gives $\sigma_0 = 250 \pm 20(a_0^2\ \mathrm{sr}^{-1})$ and $\beta = 2.45 \pm 0.05$ (atomic units) for energies ranging from 500 to 1000 eV. Introducing this value in Eq. (3) and considering the maximum angular acceptance of the detector (3.5×10^{-5} sr), we obtain an error contribution for the elastic forward scattering of less than 0.1% at 1000-eV impact energy. The contribution at higher energies can be estimated by assuming that the Born approximation is valid for small angles and therefore $(d\sigma/d\Omega)_{\mathrm{elastic},\Delta P=0}$ must be constant.

TABLE I. Experimental and theoretical total cross sections (in units of a_0^2) for electron scattering from CO_2.

E_0			Total cross sections (a_0^2)			
(eV)	This work[a]	Ref. [4][a]	Ref. [5][a]	Ref. [6][a]	Ref. [1][b]	Ref. [8][b]
400	21.1	22.4		22.9	30.9	
403			19.0			
441				21.9		
484				19.9		
500	18.3	18.4			26.2	19.7
576				17.6		
600	16.2				22.8	
676				15.6		
700	14.5				20.1	15.5
784				14.1		
800	13.1				18.1	
900	12.2			12.6		
1000	11.3				15.0	11.9
1024				11.7		
1156				10.5		
1250	9.55					
1296				9.61		
1444				8.50		
1500	8.64					
1600	8.05			7.61		
1750	7.54					
1764				6.86		
1936				6.39		
2000	6.85				8.17	
2116				5.79		
2304				5.36		
2500	5.69			4.61		
2704				4.53		
2916				4.11		
3000	4.85				5.60	
3500	4.27					
4000	3.83				4.25	
4500	3.48					
5000	3.19				3.40	

[a]Experimental results.
[b]Theoretical values.

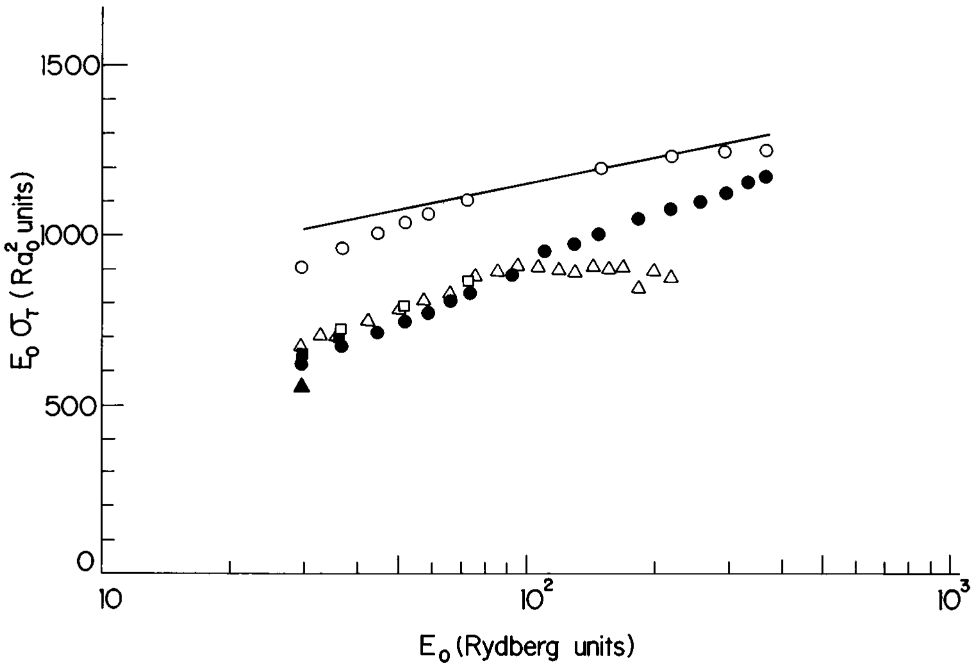

FIG. 4. $E_0\sigma_T$ in Ra_0^2 units plotted vs E_0 in R units for e-CO_2 scattering. ●: present experimental results. Δ: measurements from Ref. [6]. ■: experimental results from Ref. [4]. ▲: experimental value given in Ref. [5]. ○: theoretical data given in Ref. [1]. □: calculations from Ref. [8]. ——: Born-Bethe approximation.

Under this assumption, the maximum error contribution from forward elastic scattering at 5000 eV is less than 0.3% for our experimental conditions.

By combining the partial error components mentioned above, we have a total error of 3% for the present measurements.

III. RESULTS AND DISCUSSION

The measured total cross sections for electron scattering by CO_2 molecules in the energy range 400–5000 eV are given in Table I, together with the experimental values of Refs. [4–6] for comparison. The results of Kwan *et al.* [4] are in good agreement with those of the present work in the overlapping energy region. The value obtained by Sueoka and Mori [5] at 403 eV is 10% lower than the present one. The data from Szmytkowski *et al.* [6] show in general a good agreement with the present results for energies between 400 and 1600 eV, but are systematically less than ours for energies above 1600 eV, reaching discrepancies of about 20% at 3000 eV.

Theoretical data available in the literature are also included in Table I. Calculations of Jain and Baluja [1] give values higher than those of the present work at low energies (about 40% at 400 eV), but they agree well at higher energies (within 6% at 5000 eV). Data of Joshipura and Patel [8] show an excellent agreement with the present ones. The first Born approximation for elastic processes and the Born-Bethe theory for inelastic collisions are two significant instruments to study electron scattering from atoms and molecules at high energies [9–11]. It is worthwhile to check the suitability of these approximation for energies of up to 5000 eV, by comparing their predictions with the experimental values. As can be deduced from Refs. [9–11], a combined Born-Bethe theory gives σ_T in terms of the following equation:

$$\frac{E_0}{R}\frac{\sigma_T}{a_0^2}=\pi\left[A_{el}+B_{el}\frac{R}{E_0}+C_{el}\left(\frac{R}{E_0}\right)^2+\cdots\right]+4\pi\left[M_{\mathrm{TOT}}^2\ln\left(4C_{\mathrm{TOT}}\frac{E_0}{R}\right)+\cdots\right], \qquad (5)$$

where E_0/R is the incident energy in Rydberg units and a_0 is the Bohr radius. The constants A_{el}, B_{el}, C_{el}, M_{TOT} and C_{TOT} are related to internal dynamic properties of the targets (see Refs. [9–11]). These parameters have been calculated for some atoms in Refs. [9] and [11]. For CO_2 molecules direct calculations of these constants from molecular wave functions are not available in the literature. However, for energies high enough that the independent atom model of Mott and Massey [19] is valid, and by applying the optical theorem to the forward scattering amplitude, Joshipura and Patel [8] have proposed a simple method to obtain molecular total cross sections from the corresponding values of the constituent atoms. Accordingly, the Born-Bethe total cross sections (σ_{BB}) for electron scattering from CO_2 can be expressed from the atomic data given in Refs. [9] and [11] as

$$\frac{\sigma_{\mathrm{BB}}}{a_0^2}\frac{E_0}{R}=675.3+106.3\ \ln\frac{E_0}{R}-480.7\frac{R}{E_0}+\cdots. \qquad (6)$$

In order to study the dependence of the total cross section on the electron energy for the energy range of this experiment, we have made a Bethe plot $[(E_0\sigma_T)/(Ra_0^2)$ versus $\ln(E_0/R)]$, which includes the present results and all the experimental and theoretical values available in the literature (see Fig. 4). As may be seen the σ_T dependence on the electron energy agrees for all the experimental values given for energies less than 1600 eV. For higher energies the values given in Ref. [6] shown a clearly different behavior than those of the present work.

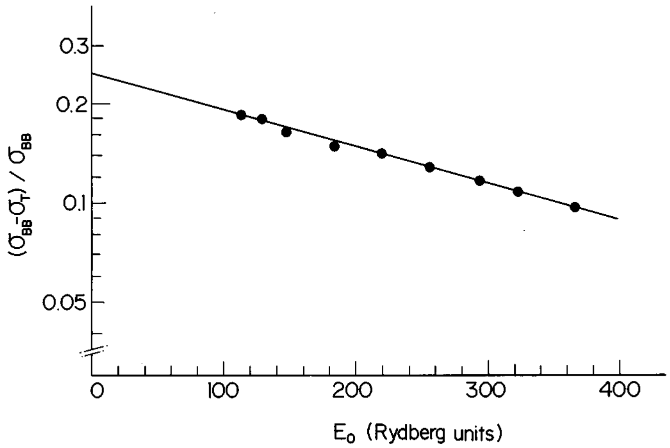

FIG. 5. Relative difference between the present total cross section (σ_T) and those predicted by the Born-Bethe approximation (σ_{BB}) vs electron incident energy (in Rydberg units).

As for the theoretical data, the calculations of Joshipura and Patel [8] predict a dependence of σ_T on the electron energy in the 500–1000-eV range, which is in good agreement with the experimental values. The values of Jain and Baluja [1], as well as those given by Eq. (6), show a shape and trend similar to the present experimental values for the whole energy range. The valid energy range of the Born-Bethe approximation for e-CO_2 collisions can be determined by studying the relative difference between σ_{BB} values and the corresponding experimental ones as a function of energy. Figure 5 is a semilogarithmic plot of $(\sigma_{BB}-\sigma_T)/\sigma_{BB}$ against E_0/R for energies above 1500 eV, obtained by using the present σ_T values. The data lie on a straight line, which may be fitted by an exponential function as

$$\frac{\sigma_{BB}-\sigma_T}{\sigma_{BB}}=0.239\exp\left[-\frac{1}{416}\frac{E_0}{R}\right]. \tag{7}$$

Thus total cross sections for high electron energies ($E_0>1500$ eV) can be expressed in terms of the following formula:

$$\sigma_T=\left[1-0.239\exp\left(-\frac{1}{416}\frac{E_0}{R}\right)\right]\sigma_{BB}, \tag{8}$$

where σ_{BB} is given by Eq. (6). This expression reproduces to a good approximation the present experimental results for electron energies ranging from 1500 to 5000 eV, and can be used to extrapolate σ_T values to higher energies.

IV. CONCLUSIONS

Total electron scattering cross sections for the CO_2 molecule have been measured in the energy range from 400 to 5000 eV. The experimental values obtained agree well, within the experimental errors, with previously published values in the energy range from 400 to 1600 eV. Above 1600 eV the only previously published measurements of Szmytkowski *et al.* [6] deviate from the present ones outside the quoted error limit, the discrepancy being larger for increasing energies. However, the present results agree well in shape and trend with the theoretically calculated values of Jain and Baluja [1] and with the predictions of the Born-Bethe approximation.

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