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Positron scattering from carbon dioxide

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We report total cross section measurements for positron scattering from carbon dioxide (CO_2). The energy range of the present measurements is 0.1–20.0 eV. The present study is undertaken to both try and resolve a discrepancy in the literature between the earlier low-energy works of Hoffman *et al.* [Phys. Rev. A **25**, 1393 (1982)] and Kimura *et al.* [J. Chem. Phys. **107**, 6616 (1997)], and to extend the available data to lower energies. We find generally good agreement with the data of Hoffman *et al.* over the common experimental energy range. A comparison of the present data with available calculations is also made, as is a comparison with corresponding electron total cross section data.

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I. INTRODUCTION

With the advent of high-resolution positron trap-based beams [1], the propensity to measure accurate integral cross sections for elastic and inelastic processes in molecules has become a reality. Under these circumstances, the need for accurate and reliable total cross section (TCS) data has become very important, because such total cross sections provide a self-consistency check for the validity of the integral cross section measurements. Such a role for total cross sections has been well known and put into practice for many years in the electron channel [2–4]. In addition, while total cross sections are perhaps not the most sensitive test for the validity of a positron-molecule scattering theory, they nonetheless still fulfill such a role.

Previous experimental measurements of total cross sections for positron (e^+) scattering from carbon dioxide (CO_2) are due to Hoffman *et al.* [5], Kwan *et al.* [6], Sueoka and Mori [7], Sueoka and Hamada [8], and Kimura *et al.* [9]. The data of Kwan *et al.* are for positron energies in the range 30–500 eV. These energies are outside those of the present study and so we do not consider this paper further. We also note that we believe the data of Kimura *et al.* supercedes that of both Sueoka and Mori and Sueoka and Hamada, and so accordingly, we restrict our discussion to the results in Kimura *et al.* It is clear from Fig. 2 of Kimura *et al.* that there are important discrepancies between their data and those of Hoffman *et al.*, and it is these discrepancies that provide one rationale for the present investigation. Theoretical results for total e^+-CO_2 cross sections are due to Gianturco and Paioletti [10], within a body fixed–fixed nuclei (BF-FN) framework, Gianturco and Mukherjee [11], within both body fixed–vibrational close coupled (BF-VCC) and body fixed-vibrational close coupled and adiabatic angular momentum coupling [BF-VCC(AAMC)] schemes, and Horbatsch and Darewych [12], who employed a single-center formalism within the fixed-nuclei approximation.

At the University of Trento, low-, intermediate-, and high-energy electron and positron total cross sections have been studied extensively [e.g., [13–15]] over many years. As a consequence, in the next section of this paper we only briefly describe the experimental apparatus and techniques used to make our measurements. Following that our results and a discussion of these results are presented. Finally, some conclusions from the present work are drawn.

II. APPARATUS AND TECHNIQUES

The positron spectrometer employed in our measurements was developed in the Trento laboratory and has already been described in a previous paper [16]. General information about the present attenuation technique can be found, for instance, in [17]. Although that paper specifically looked at electron cross sections, the two conjugated particles share most of the properties relevant to the current investigation. Here we therefore only outline those characteristics which are relevant to the present measurement.

Slow positrons are produced by a 1 μm tungsten-film moderator in front of a ^{22}Na radioactive source [18]. These positrons are transported and focused into the scattering chamber using a series of charged particle optics with appropriate applied potentials. Note that a weak magnetic field (8–10 G) is also present in the scattering region. The energy resolution of the positron beam has been evaluated to be slightly less than 0.3 eV full width at half maximum (FWHM), possibly as a result of the partial monochromatization in the deflector and in the optics [13]. In this paper we report cross section values down to energies of 0.1 eV, but values below 0.5 eV are to be regarded as indicative. Indeed, the measured values are always the convolution of the real (unknown) cross section with the spectrometer apparatus function and with the positron energy distribution. At low energies, where the width of the energy distribution becomes comparable to the energy itself, the convolution can impose sizable deformations to the measured cross section. The procedure to deconvolve the real cross section from the measured data is not straightforward, mainly because both the apparatus function and the beam energy distribution are not well characterized. On these grounds the measurements at energies lower than 0.5 eV (see Table I) should be taken with

TABLE I. The present total cross section ($\times 10^{-16}$ cm^2) data for positron scattering from carbon dioxide. The errors represent the standard deviation on the measured cross section at a given energy. See text for a discussion of the absolute error.

Energy (eV)	Total cross section ($\times 10^{-16}$ cm^2)
0.1	34.42±0.67
0.2	26.61±0.61
0.3	22.11±1.18
0.4	19.01±0.46
0.6	14.92±0.48
0.85	12.77±0.83
1.1	11.61±0.48
1.6	9.79±0.30
2.1	8.39±0.25
2.6	7.85±0.33
3.6	6.95±0.12
4.6	6.70±0.15
5.5	6.43±0.06
5.6	6.40±0.10
5.75	6.40±0.01
6.0	6.24±0.30
6.25	6.41±0.14
6.6	6.14±0.60
6.75	6.72±0.04
7.1	6.91±0.22
7.25	6.87±0.08
7.6	6.95±0.13
7.75	6.99±0.10
8.0	6.90±0.14
8.6	7.03±0.42
9.6	6.91±0.28
11.6	7.28±0.38
14.6	8.05±0.52
17.6	8.85±0.26
19.6	9.56±0.15

caution. Nevertheless, we believe they warrant publication. Using a ^{22}Na source with an activity of 8 mCi, positron beam intensities at the detector were found to vary from 8 to 40 s^{-1}, the highest value being achieved at the high-energy limit. The zero for the energy scale of the present positron measurements has been determined, in the absence of the target gas, with a retarding potential analysis of the beam. Such a measurement suggests a probable error of ±0.1 eV in our energy scale. The accurate determination of the energy scale calibration is particularly important at low energies, in all cases where the total cross section rises rapidly with decreasing energy. We note that in these instances even a small inaccuracy in the energy calibration can produce a significant uncertainty as to the true value of the TCS. Such an effect would, for example, be very misleading for theoreticians trying to describe the scattering process by reproducing experimental data. A more thorough description of these issues and our calibration technique can be found in Ref. [19].

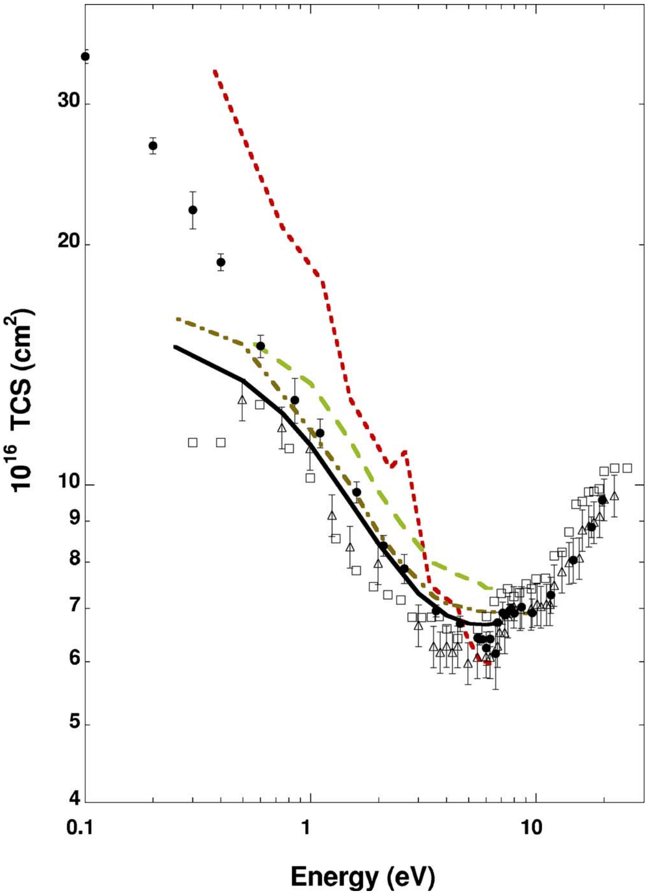

FIG. 1. (Color online) The present (•) total cross section ($\times 10^{-16}$ cm^2) data for positron scattering from CO$_2$. Also shown are the earlier TCS results from Hoffman *et al.* [5] (△) and Kimura *et al.* [9] (□) and the calculations from Horbatsch and Darewych [12] (- – - –), Gianturco and Paioletti [10] (– –), the BF-VCC-AAMC [11] (- - - - -), and the BF-VCC of Gianturco and Mukherjee [11] (—). The errors on the present data represent the standard deviation in the measured cross section at a given energy. See the text for a discussion of the absolute error.

High-purity (>99.9%) carbon dioxide was used throughout this study. The gaseous target was fed to the scattering cell with a two-way diverter valve, where the same amount of gas was diverted to the scattering cell or alternatively was injected directly into the vacuum system. In the first case attenuation of the positron beam was obtained. With such a provision we obtain that the background pressure outside the gas cell and therefore the attenuation of the beam in the path outside the gas cell are constant during the measurement cycle. The background pressure during the measurements was typically 10^{-3} of the pressure inside the gas cell.

Total cross sections were computed according to the Beer-Lambert law,

$$I_1 = I_0 \exp\left(\frac{-(P_1 - P_0)L\sigma}{kT}\right) \qquad (1)$$

where I_1 is the positron beam count rate at P_1, the pressure measured with the gas routed to the scattering cell, k is Bolt-

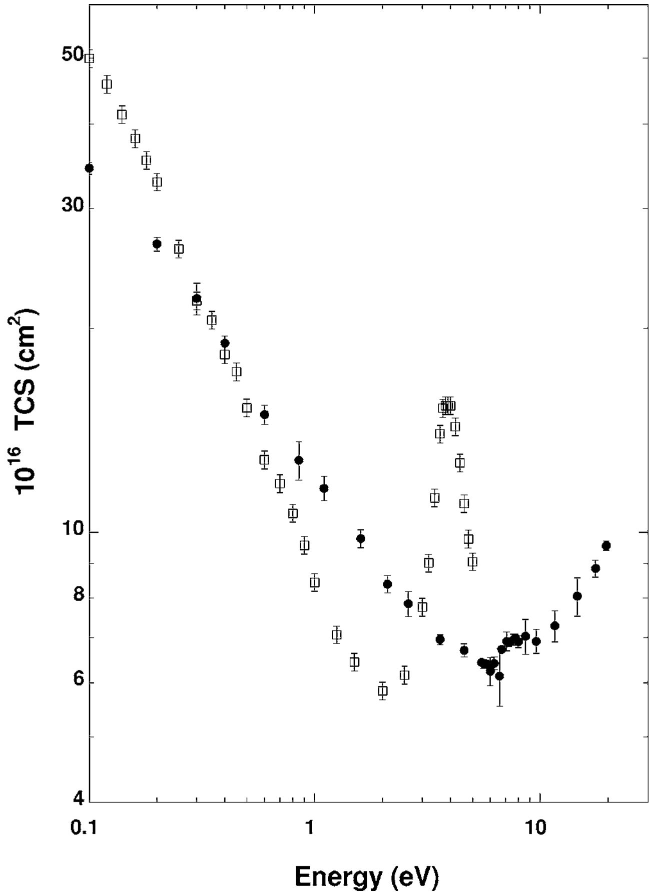

FIG. 2. Total cross sections for positron and electron scattering from CO_2. The present positron data (•) and the earlier electron data from Buckman *et al.* [24] (□) are depicted.

zmann's constant, T is the temperature of the gas (K), σ is the total cross section of interest, I_0 is the positron beam count rate at P_0, the pressure with the gas diverted to the vacuum chamber, and L is the length of the scattering region (see later). In order to minimize double scattering events and ensure the TCS is pressure independent, the ratio I_1/I_0 has been kept to values larger than 0.7. Furthermore, the standard checks on the linearity of the plots of $\log_e (I_1/I_0)$ vs gas pressure [20] were performed at selected energies. The geometrical length of the scattering region is 100 ± 0.1 mm, with apertures of 1.5 mm diameter at both the entrance and exit of the scattering chamber. End effects [13] were considered in the present study. It has been demonstrated [14,21] that the effects due to the entrance and exit apertures cancel if both the aperture diameters are equal, so that their contribution to the uncertainty in the value of L is possibly less than 0.15%. In the current application, the value of L used in Eq. (1) has been corrected to account for the path increase caused by the gyration of the positrons in the focusing magnetic field present in the scattering region (typically this correction is $\sim 6\%$). This arises because in the no-B-field configuration the positron trajectories are straight segments; however, with a field applied they are bound to move on a spiral which thus may increase the true value for L, which represents the length of the positron path in the gas-filled region. The gyration of the particles can also potentially increase the angular resolution error with respect to the no-field case [22]. However, even though absolute differential cross sections for e^+/CO_2 scattering are not currently known [1,23], we believe that the present geometry guarantees a small error ($0 < \Delta\sigma < 10\%$). The scattering cell pressure has been measured with an MKS Baratron capacitance manometer (Model 628B: 1 Torr full scale) operated at 100 °C. Since the scattering chamber was at room temperature (24 ± 2 °C), a thermal transpiration correction has been applied to the pressure readings. This correction has been calculated according to the model of Takaishi and Sensui [24] and is of the order of 10% over the entire energy range.

Measurement time was of the order of 1 h per each energy point, with each point being the average of 100 single determinations. The positron beam obtained with the present apparatus [16] was extremely stable over times of the order of one month and indeed, no influence of the target gas on the beam characteristics was noted. A new conditioning of the moderator film was made only at the beginning of the present study. The absolute errors on our measurements (not given in Table I nor Fig. 1 and 2) have been evaluated as the root of the quadratic sum of the contributing errors. A fuller discussion of the origin and of the evaluation techniques of such contributions can be found in Ref. [14] and in the references contained in that paper. At this point, however, we specifically note that the respective contributions due to the uncertainties in our thermal transpiration and B-field spirally corrections are small; they do not contribute significantly to the overall errors on our TCSs. These overall uncertainties typically amounted to ±3% at the higher energies and to ±7% at the lower energies, the dominant contribution being due to the uncertainty in the pressure determination. Note that the error quoted in Table I is the statistical error only. We further note that our lowest energy points ($E < 0.3$ eV) could bear a larger error due to the energy resolution being $\simeq 0.3$ eV.

III. RESULTS AND DISCUSSION

In Fig. 1 we plot the present TCSs for positron-CO_2 scattering along with the earlier data due to Hoffman *et al.* [5] and Kimura *et al.* [9], and various theoretical results from Gianturco and colleagues [10,11] and Horbatsch and Darewych [12]. The present TCSs are also summarized in Table I.

It is clear from Fig. 1 that the present TCS increases strongly in magnitude as the energy of the positrons is progressively decreased. As CO_2 does not have a permanent dipole moment, this behavior indicates the importance of its molecular polarizability at these lower energies. The present low-energy behavior, is in stark contrast to the data of Kimura *et al.* [9], whose TCS begins to "turn over" at 0.6 eV, and in some contrast to the TCS of Hoffman *et al.* [5], which increases in magnitude as E decreases but at a slower rate than the present. In general, however, for energies greater than about 0.8 eV, the present data are in excellent agreement, in terms of both the shape and magnitude of the TCS, with the data of Hoffman *et al.* As a consequence, our data above 0.8 eV confirm the results of Hoffman *et al.* Note that above 0.8 eV all three TCS measurements exhibit the same qualitative energy dependence and structures. In particular we confirm, both in magnitude and position, the sharp step in the TCS at around 7.8 eV which, consistent with Kimura *et al.*, we believe is due to the opening of the positronium channel. However, we also note that in some cases the magnitude of the TCS of Kimura *et al.* is ~15% larger than that of Hoffman *et al.* and the present. The discrepancy between the present data and the earlier studies at energies lower than 0.8 eV is consistent with both the earlier studies [5,9] suffering from a larger forward-angle scattering error. Alternatively, an error in their energy zero calibration could lead to a qualitatively similar disagreement. Note that a discussion of the energy zero determination in positron TCS scattering experiments can be found in [19].

If we now compare the present TCSs with the available theoretical results [10–12], worst overall agreement is observed between the current data and the BF-VCC-AAMC calculation of Gianturco and Mukherjee [11]. This calculation predicts structures in the TCS that are not observed by any of the experiments, and for energies less than 4 eV it significantly overestimates the magnitude of the TCSs. On the other hand, the shape of its lower energy cross sections exhibits a form which is very similar to that of the present results. The BF-FN calculation of Gianturco and Paioletti [10] also overestimates the magnitude of the experimental TCSs over most of the common energy range, although its global features are in qualitative accord with all the measured data. Best overall agreement between theory and the present TCSs is provided by the BF-VCC calculation of Gianturco and Mukherjee [11], and the elastic integral cross section computation of Horbatsch and Darewych [12]. It should be noted that the agreement between our data and that of Horbatsch and Darewych is a little artificial, because those authors employed semiphenomenological polarization potentials and they do not account for open inelastic channels or direct annihilation. However, neither the BF-FN or BF-VCC calculations of Gianturco and colleagues [10,11], nor the calculation of Horbatsch and Darewych, are able to reproduce the shape or magnitude of the low-energy (<1 eV) TCS. We hope that with the availability of our data, these and other theoreticians might be stimulated to revisit or consider this problem. Finally, with respect to the low-energy (now $E<2$ eV) behavior of the e^+-CO_2 total cross section, we are intrigued by the fact that to better than 3% correlation the TCS exhibits $\frac{1}{E^{1/2}}$ dependence. While it is possible this observed energy dependence is coincidental, the strong nature of the correlation makes us wonder whether there might be some interesting fundamental physics here. However, particularly in the electron channel and for the noble gases, there has been quite a lot of work [25,26,15] on partitioning the energy dependence of the total cross sections. Various energy dependencies of the TCS have been observed, including $E^{1/2}$, E^{-1}, and $E^{1/2}$ etc., with the nature of the behavior depending on the species studied and the energy regime being considered.

We believe it is also useful to compare the present positron-CO_2 TCSs to the corresponding electron (e^-)-CO_2 TCSs from Buckman *et al.* [27]. This is illustrated in Fig. 2. Here we see that both the low-energy ($E<2$ eV) e^-/TCSs and e^+/TCSs exhibit qualitatively similar behavior, namely, the magnitude of their TCSs increases sharply with decreasing energy. However, quantitatively, the respective rates of increase for the e^- and e^+ total cross sections are very different, with the rate of increase in the e^-/TCS being larger. Indeed, if we allow for a ~80 meV shift in the energy scale of the electron data (which is within its stated uncertainty), then the e^-/TCS exhibits a $1/E$ dependence at low energies. The difference in the e^- and e^+ TCSs is unlike what we found in our previous study on water [28], where the slopes of the TCSs for both particles were essentially identical (to ~10%). The present result is intuitively more understandable, as there are very different processes occurring (the absence of exchange in the positron case, while the static interaction is repulsive in the case of positrons and attractive in the case of electrons, for example) when each respective projectile interacts with carbon dioxide, so that you might expect the scattering cross sections to be different in each case. For energies greater than about 0.4 eV, the e^+/TCSs tend to be larger in magnitude than those of the electrons, until the effects of the well-known $^2\Pi_u$ resonance in e^-+CO_2 scattering become apparent.

IV. CONCLUSIONS

We have reported a total cross section measurement for positron–carbon dioxide scattering. The present study extended the available data to lower energies and resolved a discrepancy between the absolute magnitude of the earlier TCSs from Hoffman *et al.* [5] and Kimura *et al.* [9] in favor of Hoffman *et al.* [5]. The present investigation also showed that more theoretical input was needed for an accurate description of this scattering system, particularly at the lower energies. Finally, a comparison between the low-energy behavior of the e^+ and e^- total cross sections showed only a qualitative correspondence.

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