

## Low-energy positron scattering by CO<sub>2</sub>

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In this work we present results of integral (ICS) and differential (DCS) cross sections for positron-CO<sub>2</sub> scattering at low incident energies. Our ICS shows a significant improvement toward the experimental data, especially below 2 eV, and all the way up to the positronium formation threshold (7.8 eV), in comparison to our previous calculations [S. d'A. Sanchez, F. Arretche, M. T. do N. Varela, and M. A. P. Lima, *Phys. Scr.* **T110**, 276 (2004)]. Our calculated DCSs show a better resemblance in shape with the quasielastic experimental points of the Detroit group [D. A. Przybyla, W. Addo-Asah, W. E. Kaupilla, C. K. Kwan, and T. S. Stein, *Phys. Rev. A* **60**, 359 (1999)], but the agreement is still not fully satisfactory, indicating a need for further theoretical and experimental investigation.

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### I. INTRODUCTION

Positron physics plays a major role in several areas of research, ranging from medical applications like the positron emission tomography [1] to interstellar research [2]. The development of experimental and theoretical techniques have greatly improved, allowing accurate studies of positron scattering. For a good review on the subject, see [3]. This scenario motivates efforts to better understand the basic interactions of positron with matter. In this way, the study of low energy positron-atom (molecule) collisions becomes important. There are major difficulties to perform such measurements and calculations, though. On the experimental side, advances like the Surko trap allowed high-density positron beams, very important in the differential scattering studies. On the theoretical side, few methods are available to perform calculations of integral and differential cross sections for positron scattering against polyatomic molecules. Among the successful ones, obtaining cross sections for annihilation processes and real positronium formation still represents a difficult challenge [3].

The Schwinger multichannel method (SMC) for positron scattering [4] was developed to obtain data of the scattering processes, such as elastic, electronic excitation, annihilation [5], and, more recently, vibrational excitation [6] cross sections. Although it does not contain the real positronium formation channel explicitly, it has been very successful in reproducing the scattering results for several molecular systems [7]. The description of positron-molecule scattering represents a more challenging task than its more common counterpart involving electrons. The main reason lies in the fact that the polarization potential for both projectiles is attractive, while, in general, the electron experiences an attractive static potential and the positron a repulsive one. So, the overall potential experienced by the positron during the collision comes from adding the negative polarization potential to a positive static one. If both potentials are equivalently strong, this cancellation becomes very sensitive to small errors in their construction.

In a previous paper [8], we presented integral (ICS) and differential (DCS) cross sections for  $e^+$  scattering by CO<sub>2</sub>

molecule. The results we have obtained for the differential cross sections were in reasonable agreement with the experimental data of the Detroit group [9], despite some differences at low angles. Those differences occurred mainly due to a shallow minimum around 60° in their data. This sort of structure was also hard to describe even in the electron scattering case and only recently the Caltech group was able to reproduce it with a better description of the polarization [10]. Although our data did not show the minimum, we were able to get the first indication of how to deal with this problem. The major problem with our previous calculation, however, was in the integral cross section. Our results were below the experimental data even by a factor of 2 for energies below 2.0 eV. In this energy region, we found a Ramsauer-Townsend minimum around 0.5 eV in the  $A_g$  symmetry and a peak structure in the  $B_{1u}$  symmetry (at 1.25 eV) that although it looked like a resonance, the eigenphase sum indicated otherwise (our calculations were performed with the molecule in the  $D_{2h}$  symmetry group for computational reasons). Both structures occur in different impact energies, which led us to conclude that this was the reason why the minimum was strongly pronounced in our final ICS data (sum over all symmetry decompositions) and why our results were underestimated. We also stated that a better treatment of the polarization would force the minimum to the right and the peak to the left into the same energy position, increasing the value of the integral cross section and hence providing a better agreement with the experimental results. However, we were pushing our computational effort to the limit at that time and a better calculation was not feasible. Only recently, the acquisition of a bigger computer allowed us to revisit this calculation and test our previous hypothesis.

### II. THEORY

The method we used to obtain these new results was the same as before, the SMC method. It is a well established method and is extensively described in the literature [4], but a review of its major concepts is important. It is a variational method for the scattering amplitude and uses configuration state vectors ( $\chi_m$ ) to expand the scattering wave function.

Details about it will be given further on in the text. The final working form obtained for the scattering amplitude is given by

$$f_{\mathbf{k}_i, \mathbf{k}_f} = -\frac{1}{2\pi} \sum_{m,n} \langle S_{\mathbf{k}_f} | V | \chi_m \rangle (d^{-1})_{mn} \langle \chi_n | V | S_{\mathbf{k}_i} \rangle, \quad (1)$$

in which

$$d_{mn} = \langle \chi_m | A^{(+)} | \chi_n \rangle, \quad (2)$$

with

$$A^{(+)} = Q\hat{H}Q + PVP - VG_p^{(+)}V. \quad (3)$$

In the equations above,  $S_{\mathbf{k}_i(f)}$  is a solution of the unperturbed Hamiltonian (molecular Hamiltonian plus the kinetic energy operator for the incident positron);  $V$  is the interaction potential between the incident positron and the target.  $P$  and  $Q$  are, respectively, projection operators onto energetically open and closed electronic states of the target;  $\hat{H}$  is the collision energy minus the full scattering Hamiltonian; and  $G_p^{(+)}$  is the free-particle Green's function projected onto the  $P$  space. The SMC works in a fully *ab initio* framework and hence does not use model potentials of any kind. In Eq. (1), the configurations are always multiplied by short-ranged operators (either  $V$  or  $Q$ ) that vanish asymptotically. As a result, the scattering wave function may be expanded in a set of scattering basis functions ( $\chi_m$ ), constructed from Cartesian Gaussian functions, allowing the study of molecules with arbitrary geometry (the appropriate scattering boundary condition is provided by the Green's function).

As this work is concerned with the elastic scattering channel, the open space operator projects only onto the target's ground state. We performed this calculation in the static plus polarization approximation, in which correlation effects are introduced through single excitation of the  $(N+1)$ -particle compound system composing the closed space  $Q$ . This inclusion is vital when working with a low energy projectile, since the collision time is long enough to allow the electronic cloud to rearrange. In this approximation, the configurations are written as

$$|\chi_{ij}\rangle = |\Phi_i\rangle \otimes |\varphi_j\rangle, \quad (4)$$

where  $\Phi_i$  is the ground or a singly excited target state and  $\varphi_j$  is a positron scattering orbital.

### III. COMPUTATIONAL DETAILS

In our polarization calculations, the excitations from the target's occupied orbitals were performed only to polarized ones [11]. In a first calculation, we included only the valence orbitals (8 out of 11) and 33 polarized orbitals in the active space, generating a total of 35 775 configurations (hereby referred to as data set 1). To investigate the influence of the inner shell orbitals, we included them, creating a total of 42 240 configurations (data set 2).

To analyze possible numerical instability problems in our basis set, we used the strategy presented in the work of [12]. Quasilinear dependent configurations may induce the pres-

ence of small eigenvalues in the  $d_{mn}$  matrix and, since this matrix is inverted in our calculations, it would give rise to spurious structures in our curves. In [12], it was observed that if the configurations related to these eigenvalues were removed in a careful way, these structures would be eliminated in our results. To choose which one would have to be removed, the basis set born approximation (BSBA) was introduced in [12], in which the  $VG_p^{(+)}V$  term is removed from Eq. (3) and, in cases of static approximation calculations, so is the  $Q\hat{H}Q$  one. In this case,  $A^{(+)}$  reduces to the  $PVP$  term and the SMC becomes equivalent to the first-order Born approximation (FBA)—which is not able to account for physical resonances, so only the spurious ones due to basis vectors problems are present in the BSBA cross section.

In this work, two different strategies to analyze the basis set stability were used:

(i) For data set 1, configurations were removed until the ICS calculated with the BSBA approximation converges to the FBA one, i.e.,  $\sigma^{BSBA} \approx \sigma^{FBA}$ , and also, simultaneously, the annihilation parameter converges to  $Z$ , i.e.,  $Z_{eff} \approx 22$  for  $\text{CO}_2$ . The reason for this comparison is that, since the two calculations are similar (in the assumptions they made), the differences between them would arise only due to numerical problems in the basis set. With this methodology, it was noticed that all the configurations with eigenvalues lower than  $10^{-3}$  would have to be removed. This threshold was applied to the polarization calculation, and only 0.075% of the configurations were removed.

(ii) For data set 2, a different approach was used. The eigenvalues of  $\langle \chi_m | PVP + Q\bar{H}Q | \chi_n \rangle$  were analyzed directly, where  $\bar{H}$  is equal to  $\hat{H}$  for a fixed chosen energy (we have learned that this procedure is almost independent of the chosen energy). Configurations with small eigenvalues were removed in small intervals and the ICS and eigenphase sum were analyzed for each symmetry independently. A new interval was set and new configurations were removed. The new curves were compared with the ones previously obtained and the proceeding would stop when convergence was met. With this methodology, we removed only 0.035% of the configurations.

In our previous calculations [8], we have used two different basis sets. One of them was the first basis presented in the paper of Lee *et al.* [13], and the other one was the second basis shown in the paper of Kroin *et al.* [14]. Both bases provided good values for the polarizability ( $2.79 \times 10^{-24} \text{ cm}^3$  for the first one and  $2.57 \times 10^{-24} \text{ cm}^3$  for the second) and ground state energy of the molecule ( $-246.029$  and  $-246.033$  hartree, respectively). At that time, both of them had a viable size to perform such calculations, generating reasonable results when compared with the experimental data of Hoffman *et al.* [9] and the theoretical work of Gianturco and Paoletti [15]. In this work, we used the third basis presented in the paper of Lee *et al.* [13]. Although this basis provides a slightly higher ground state energy,  $-246.022$  hartree, it gives  $2.86 \times 10^{-24} \text{ cm}^3$  for the polarizability, closer to the experimental data of  $2.911 \times 10^{-24} \text{ cm}^3$  [16]. The main reason for this choice is that this basis set contains more diffuse functions, which are important to describe the long range portion of the polarization potential.

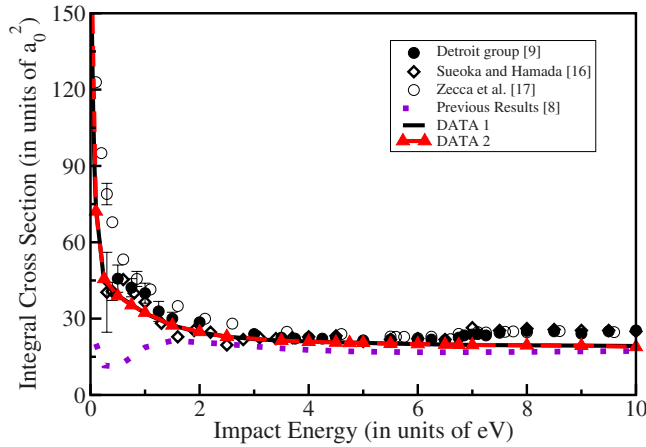


FIG. 1. (Color online) Integral cross section for positron- $\text{CO}_2$  scattering. Full circles, data of the Detroit group [9]; diamonds, measurements of Sueoka and Hamada [17]; empty circles, experimental points of Zecca *et al.* [18]; dotted line, previous results [8]; full line, data set 1; dashed line with triangles, data set 2. (Error bars are shown only when bigger than the symbols.)

#### IV. RESULTS

The improvement of the integral cross section can be seen in Fig. 1. Both data set 1 and data set 2 show very similar results, as would be expected since the inner-hole orbitals are very tightly bound to the nuclei and would bring little or even no polarization effects to the scattering process. Besides, both results are in very good agreement with the experimental results of Hoffman *et al.* [9] and Sueoka and Hamada [17] and just a little below the recent data of Zecca *et al.* [18] all the way up to the positronium formation threshold (7.8 eV).

Although not shown in the manuscript the cross sections obtained with calculations using all configurations are very similar to those obtained with data set 1 and data set 2 (in both cases they are less than 10% different from data sets 1 and 2). This difference shows that although small there existed a numerical problem which was removed with the procedures described to obtain data sets 1 and 2.

The main difference between our new curves and the one published in [8] is below 2.0 eV. This can be better explained by looking at the ICS only for the symmetries  $A_g$  and  $B_{1u}$ , which can be seen in Fig. 2 (we only show the results of data set 2 for clarity purposes, since it coincides with data set 1). We can see a shift to the right in the minimum position at data set 2 when compared to our previous calculation in the  $A_g$  symmetry, and a shift to the left for the peak structure at data set 2 for the  $B_{1u}$  symmetry. This behavior is well explained due to the improvement in our polarization treatment of the molecule. Since the peak and the minimum are now almost at the same energy position, the peak is giving the necessary background to make our curve match the experimental data. In our previous work, the peak was further to the right in energy when compared to the Ramsauer-Townsend minimum, hence explaining why it was so pronounced in our final ICS curve. This is a perfect example of how sensitive the calculation for positron scattering may be

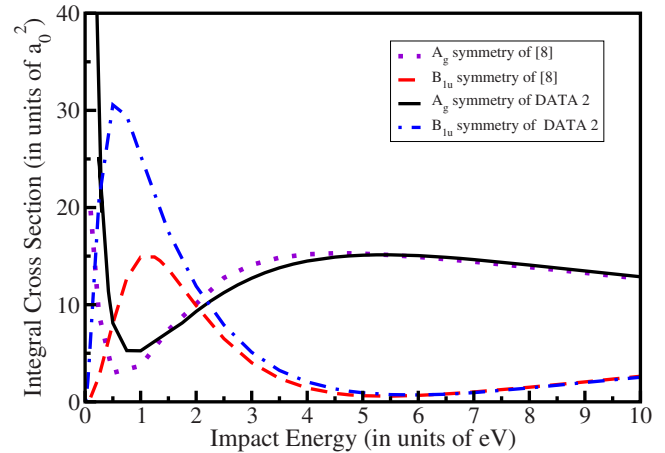


FIG. 2. (Color online) Integral cross section for positron- $\text{CO}_2$  scattering for the  $A_g$  and  $B_{1u}$  symmetries. Dotted line, previous results [8] for the  $A_g$  symmetry; dashed line, previous results [8] for the  $B_{1u}$  symmetry; full line, data set 2 for the  $A_g$  symmetry; dot-dashed line, data set 2 for the  $B_{1u}$  symmetry.

due to the difference in sign of the static and the polarization potentials.

In Fig. 3, we show the eigenphase sum for both mentioned symmetries (again, only for data set 2). Here, we can confirm that, as opposed to the electron scattering situation [19], there is no clear indication of a virtual state, although the ICS shows a tendency of it since it rapidly increases at very low impact energies. Perhaps it may still be a problem of underpolarization of the target. Also, we confirm with our new results that the ICS peak structure of the  $B_{1u}$  symmetry does not correspond to a resonant state, although its energy behavior indicates otherwise (as we further polarize the target, it moves down in energy).

The differential cross sections are now closer in shape when compared to our previous calculations [8] (Fig. 4). However, we were still not able to reproduce the data of Przybyla *et al.* [9], especially when we analyze the shallow minimum at  $60^\circ$ . We can also notice some difference between data sets 1 and 2 at 4.75 eV (Fig. 4—top panel). This

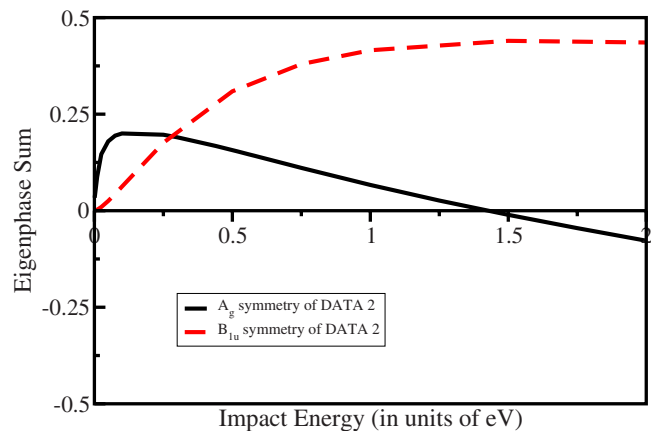


FIG. 3. (Color online) Eigenphase sum for positron- $\text{CO}_2$  scattering for the  $A_g$  and  $B_{1u}$  symmetries. Full line, data set 2 for the  $A_g$  symmetry; dot-dashed line, data set 2 for the  $B_{1u}$  symmetry.

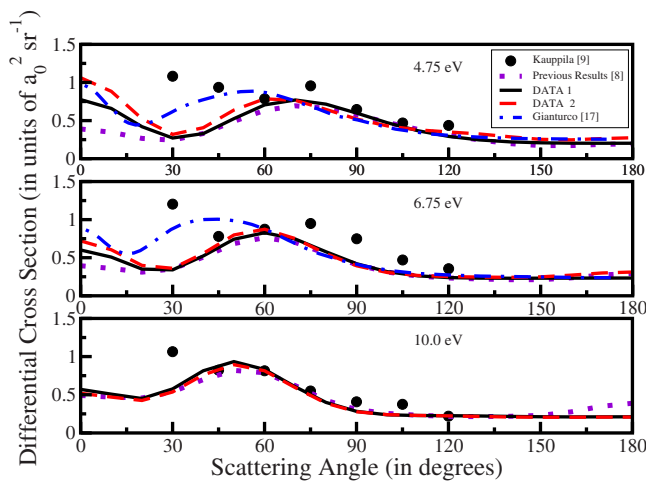


FIG. 4. (Color online) Differential cross section for positron- $\text{CO}_2$  scattering. Full circles, data of [9]; dotted line, previous results [8]; full line, data set 1; dashed line, data set 2; dot-dashed line, theoretical calculations of Gianturco and Paoletti [15].

difference is not related to the way data sets 1 and 2 were obtained in the polarization calculation, but it is due to the way we dealt with the numerical instability issue. It is still not clear which is the best procedure for dealing with the numerical instabilities of the scattering basis set. The basis set treatment in data set 1 was made by prioritizing the short range interaction and then applying it to the full polarization results, while the second one was performed taking into account also the long range potential from the start. Since high partial waves are also responsible for small eigenvalues of  $V_{mn}$  (due to the angular momentum barrier), we believe that we may have removed more configurations than needed in

data set 1, since in the static calculation the orbitals are more attached to the nuclei than in the polarization results.

## V. SUMMARY

In this work we presented our results concerning positron- $\text{CO}_2$  scattering. Our new data are in much better agreement with the experimental results of Hoffman *et al.*, Pryzbyla *et al.* [9], and Zecca *et al.* [18] than our previous one [8], especially below 2 eV. This reaffirms the necessity of a good description of the polarizability of the target and the necessary use of diffuse functions in the basis set to obtain a better description of the process. However, there are still some important differences in the differential cross section results between the experimental data and even between the two procedures data sets 1 and 2 used to calculate this process. It is very important that these, at present, *ad hoc* procedures should be more fully justified theoretically. This will require further work. It is also important, in this connection, that additional experimental results should be obtained. Finally, our results show that there is a tendency to virtual state formation and also a resonantlike structure in the cross sections that becomes more prominent as we improve our treatment of polarization. The first feature is associated with the  $A_g$  symmetry and the second with the  $B_{1u}$ . We take from this a clear invitation to carry out studies of their dependence on the nuclear geometry and vibrational excitation of  $\text{CO}_2$ .

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