Eur. Phys. J. D (2020) 74: 30
https://doi.org/10.1140/epjd/e2020-100439-5

THE EUROPEAN PHYSICAL JOURNAL D

Regular Article

Low-energy positron scattering from C_2H_2*

Wagner Tenfen[1,a], Marcos V. Barp[2], and Felipe Arretche[2]

[1] Instituto de Física e Matemática, Universidade Federal de Pelotas, 96010-900 Pelotas, Rio Grande do Sul, Brazil
[2] Departamento de Física, Universidade Federal de Santa Catarina, 88040-900 Florianópolis, Santa Catarina, Brazil

Received 9 September 2019 / Received in final form 7 January 2020
Published online 13 February 2020

Abstract. We present positron scattering cross sections with C_2H_2 molecules obtained with the Method of Continued Fractions in the static plus correlation polarization level. The differential and integral cross sections are compared with available theoretical and experimental approaches, and sensible improvement in the description of the qualitative behaviour of the differential cross sections is observed. These improvements are discussed through the evaluation of the average absolute differences between theoretical approaches and the respective normalized experimental data. Our elastic integral cross sections exhibit good agreement with model and ab initio approaches and with the available experimental data.

1 Introduction

The cross sections for positron scattering with C_2H_2 were previously studied theoretically and experimentally by several authors. From the experimental point of view, we can list the works of Sueoka and Mori [1], Kauppila et al. [2] and Zecca et al. [3], where Kauppila et al. established the experimental quasi-elastic differential cross sections for energies of 4, 6.75, 10, 20, 50 and 100 eV, and Sueoka and Mori and Zecca et al. presented the two existent sets of total cross sections for positron collisions with C_2H_2. It is important to observe that the data of Zecca et al. are uncorrected for the forward scattering effect, and this can significantly affect the measured cross sections when properly taken into account [4]. Since C_2H_2 is a nonpolar molecule, a Born-closure scheme is not necessary to the determination of the correction to the cross sections for forward scattering, so an extensive set of differential cross sections is needed in order to provide such corrections for positron scattering with C_2H_2.

On the theoretical side, we find the works of da Silva et al. [5], de Carvalho et al. [6,7], Occhigrossi and Gianturco [8], Franz et al. [9] and Zhang et al. [10] for the elastic process, and the works of Baluja and Jain [11], Raizada and Baluja [12] and Singh and Antony [13] which considered the inelastic processes through absorption potential formulations. It is noteworthy that the works of the SMC group ([5–7]), Franz et al. (R-matrix) and Zhang et al. are full ab initio calculations, and that Occhigrossi and Gianturco and Franz et al. (DFT, DPM and scaled R-matrix) employed correlation polarization models in order to obtain the scattering potential. Some of these works (particularly [7,9,10]) present their differential cross sections, which can be compared to the experimental data of Kauppila et al. [2]. Even that the comparison of the available calculated differential cross sections to the experimental ones seems to be satisfactory for higher incident positron energies (about 10 eV), we notice that further improvement is still necessary in order to describe the qualitative behaviour of the measurements. We also observe that the experimental differential cross sections were originally expressed in terms of arbitrary units, therefore a normalization of such data is necessary when comparing them with theory. In order to illustrate how the available calculations compare with the experimental data set constituted of N points, we show in Table 1 the calculated average absolute differences

$$\langle D \rangle = \frac{1}{N} \sum_{i=1}^{N} \frac{|\sigma_{\mathrm{dcs},i}^{\mathrm{teo}} - \sigma_{\mathrm{dcs},i}^{\mathrm{exp}}|}{\sigma_{\mathrm{dcs},i}^{\mathrm{exp}}}, \tag{1}$$

when normalizing the experimental data to the calculated values at 90 degrees. The choice of this angle is rather arbitrary and influences directly the comparison between theory and experiment, however this angle was chosen by Kauppila et al. when comparing their measured data to the available theories for each molecular target [2], so we chose this normalization angle in the present work as well.

The values given by equation (1) are related to the percentage differences between the theoretical differential cross sections and the normalized measured cross sections. This means that the available theoretical differential cross

* Contribution to the Topical Issue "Low-Energy Positron and Positronium Physics and Electron-Molecule Collisions and Swarms (POSMOL 2019)" edited by Michael Brunger, David Cassidy, Saša Dujko, Dragana Marić, Joan Marler, James Sullivan, Juraj Fedor.

[a] e-mail: wtenfen@gmail.com

Table 1. Average absolute differences as given by equation (1).

Reference	4.0 eV	6.75 eV	10.0 eV	20.0 eV
SMC [7]	0.316	0.299	0.217	0.321
DPM [9]	0.386	0.248	0.256	
RM [9]	0.283	0.722	0.315	
RM [10]	0.386	0.311		
This work	0.141	0.160	0.095	0.208

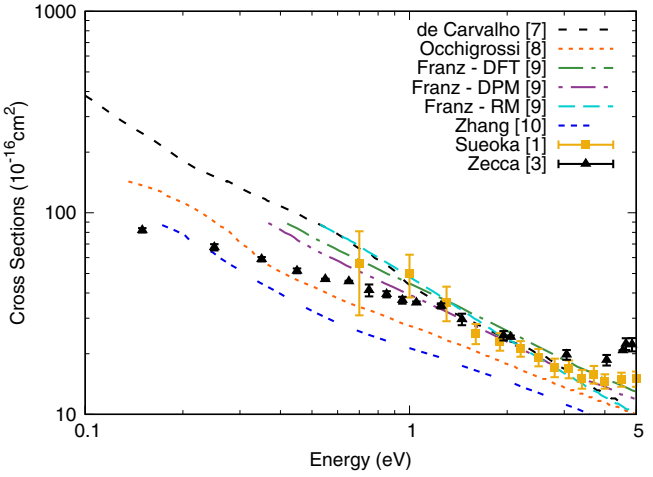

Fig. 1. Comparison between existent calculations and experimental data for positron collisions with C_2H_2 up to 5.0 eV. Theoretical elastic integral cross sections are: short dashed black line – de Carvalho et al. [7]; dotted orange line – Occhigrossi and Gianturco [8]; dashed dotted green line (DFT), dashed double dotted purple line (DPM) and long dashed turquoise line (RM) – Franz et al. [9]; triple dashed spaced blue line – Zhang et al. [10]. Experimental total cross sections are: solid yellow squares – Sueoka and Mori [1]; solid black triangles – Zecca et al. [3]. The positronium formation threshold is 4.6 eV.

sections previously reported differ at least, in average, 21.7% from the qualitative behaviour of the experimental data.

The comparison between previous theoretical integral elastic cross sections and the total cross sections up to the positronium formation threshold at 4.6 eV is satisfactory, as pointed by Zecca et al. [3], particularly for the DFT calculation of Franz et al. [9]. Also, we note that the vibrational and rotational cross sections are expected to have negligible contributions to the total cross sections, thus the comparison between theoretical elastic and total experimental cross section in this energy range is fair. Figure 1 displays the comparison between theoretical elastic and experimental total cross sections up to 5 eV. The experimental cross sections from reference [3] are not corrected for the forward scattering effects, which are particularly relevant at the lower energy range, and may be found as the main factor that justify the discrepancies between calculations and measurements toward lower energies.

Considering the arguments condensed in Table 1 and Figure 1, we present an improved description of the elastic differential cross sections using the correlation polarization potential given by the Positron Correlation Polarization (PCOP) model [14]. This paper is organized as follows: in Section 2 we introduce the numerical details concerning the scattering calculation and the target description; in Section 3 we present the results and the associated discussions about them; in Section 4 we state our conclusions.

2 Numerical details

In order to obtain the cross sections for positrons collisions with C_2H_2, we employed the Method of Continued Fractions, as described by Horáček and Sasakawa [15] and implemented by Ribeiro et al. [16]. In this methodology, the Lippmann-Schwinger equation

$$|\psi\rangle = |\phi\rangle + G_0 V |\psi\rangle \tag{2}$$

is numerically solved. In equation (2), $|\psi\rangle$ is the continuum wave function of the scattered positron, $|\phi\rangle$ is the free particle wave function, V is the reduced interaction potential and G_0 is the free particle Green's operator. In order to solve equation (2) the weakened scattering potential is defined as

$$V^{(1)} = V - \frac{V|\phi\rangle\langle\phi|V}{\langle\phi|V|\phi\rangle}, \tag{3}$$

which leads to a new scattering equation

$$|\psi_1\rangle = |\phi_1\rangle + G_0 V^{(1)} |\psi_1\rangle\,, \tag{4}$$

where

$$|\phi_1\rangle = G_0 V |\phi\rangle \tag{5}$$

and

$$|\psi_1\rangle = (1 - G_0 V^{(1)})^{-1} |\phi_1\rangle\,. \tag{6}$$

So, in order to solve the equation (6), the potential is weakened even further, which will lead to

$$|\psi_n\rangle = (1 - G_0 V^{(n)})^{-1} |\phi_n\rangle \tag{7}$$

after n repetitions of the procedure. We notice that if the scattering potential in equation (7) is negligible, then

$$|\psi_n\rangle \approx |\phi_n\rangle\,. \tag{8}$$

When this is attained in a good numerical approximation, the scattering wave function is iteratively reconstructed. The method is fast converging, so no more than 8 iterations were needed in order to obtain the converged elements of the K-matrix. More details about the methodology and the convergence criteria are available in reference [16]. The present scattering calculations were performed with partial wave expansion up to $l = 14$, where such a large expansion was not necessary for incident positron energies lower than 10.0 eV. For the low energy range, $l = 8$ was sufficient to obtain converged cross sections.

The scattering potential is obtained within the static plus correlation polarization approximation, as proposed by Jain and Gianturco [14] in which a functional of

the molecular electronic density is defined as the short range correlation interaction, and the standard polarization interaction describes the distortion of the electronic cloud for large positron distances from the molecule. The first point where these two functions cross each other is called the cutoff radius, and this position defines the transition between the correlation and the polarization interactions. For radial coordinates smaller than the cutoff radius, the correlation function is summed to the electrostatic potential, where for radial coordinates larger than the cutoff radius, the polarization potential is the one summed to the electrostatic potential. The values employed for the polarizabilities are $\alpha_0 = 28.68$ a_0^3 [8] and $\alpha_2 = 11.47$ a_0^3, this value being calculated with the GAMESS computational package [17]. These values are appreciably higher than the ones found for isoelectronic molecules like N_2 and CO, which justifies the very large cross sections in the low energy range when compared to such species.

For the determination of the static potential, a Hartree-Fock (HF) wave function for the C_2H_2 ground state was obtained with the C $(5s3p)$ and H $(3s)$ basis of Dunning [18], augmented by $s = 0.0473$; $p = 1.1233$; $p = 0.2711$; $p = 0.0697$; $d = 0.5371$ uncontracted functions at H and $s = 0.0453$; $s = 0.0157$; $s = 0.00537$; $p = 0.03237$; $p = 0.00734$; $d = 0.823$ uncontracted functions at C, employed at the experimental equilibrium geometry as determined by infrared spectroscopy [19], which yielded an energy of $E = -76.8479$ E_h. With these conditions we obtained a cutoff radius of 3.11 a_0, which is lower than the equilibrium location of the hydrogen atoms of this molecule.

3 Results and discussions

In this section, we present the calculated differential cross sections (DCS's) compared to the available experimental and previous theoretical data. In Figure 2 we show the obtained results for positron incident energy of 4 eV. As discussed before, we decided to follow the criterium of Kauppila et al. [2] and we normalized the relative quasi-elastic differential cross sections to our calculations at 90 degrees. At this point it is important to state that quasi-elastic cross sections are compositions of the elastic, vibrational and rotational cross sections, and the elastic scattering is expected to be the main part of the measured DCS's at all energies, while vibrational and rotational excitations are important at the lowest energies, as asserted by Kauppila et al. [2]. We notice that the agreement between the present results and the experimental data is remarkable up to 120 degrees, where a very small divergence is found. The calculated average absolute difference for this curve and the experimental data is $\langle D \rangle = 0.141$, which is a sensible improvement in the description of the qualitative behaviour of the calculated DCS when compared to experiment, as we can see when comparing this value with the second column of Table 1. Also, since the correlation polarization effects are more pronounced at low energies, 4 eV is a critical energy for comparison. The agreement found for this DCS suggests that the correlation polarization effects as considered here are representative of the interaction of positrons with C_2H_2 molecules.

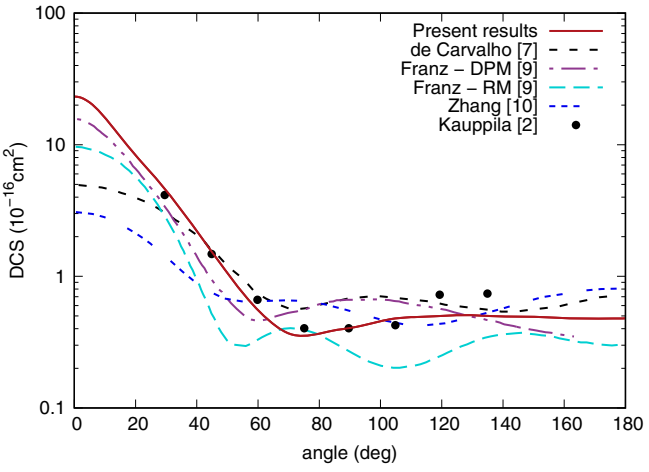

Fig. 2. Differential cross sections for positron collisions with C_2H_2 at 4.0 eV. Theories are: solid red line – present results; short dashed black line – de Carvalho et al. [7]; dashed double dotted purple line (DPM) and long dashed turquoise line (RM) – Franz et al. [9]; triple dashed spaced blue line – Zhang et al. [10]. The solid black circles are the measurements of Kauppila et al. [2] normalized to the present calculations at 90 degrees.

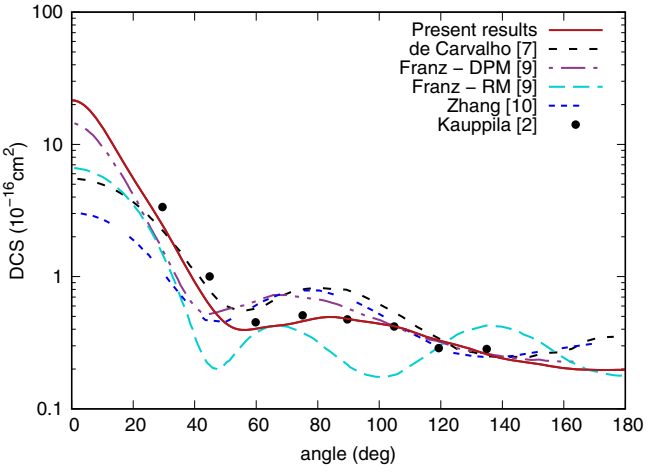

Fig. 3. Same as Figure 2, but for 6.75 eV.

In Figure 3 we present the calculated differential cross sections for positrons with incident energies of 6.75 eV, and we also compare it to other theoretical approaches and to measurements normalized at 90 degrees. The previous analysis remains valid for this incident energy, strengthening the idea that the correlation polarization potential is well described, however some differences appear to be more prominent for this curve, and this observation is sustained by the calculated value $\langle D \rangle = 0.160$, which is slightly higher than the result obtained for 4 eV. Still, this represents excellent improvement in the description of the experimental DCS.

For higher incident energies, we expect improvement in the agreement between theories and experiment, as the correlation polarization effects becomes less significant. This

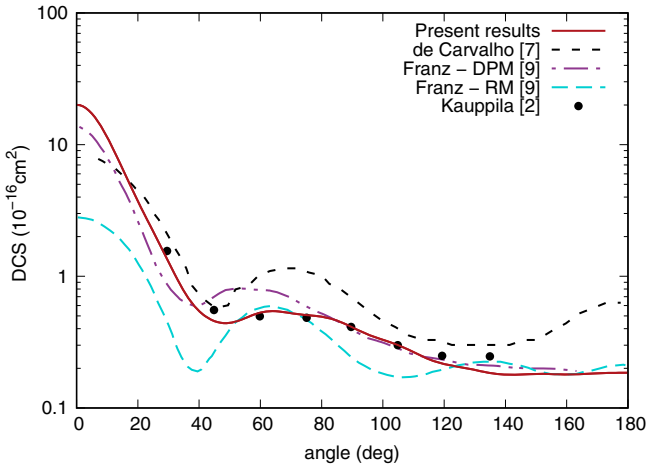

Fig. 4. Same as Figure 2, but for 10.0 eV. Calculations of Zhang et al. [10] are unavailable for this energy.

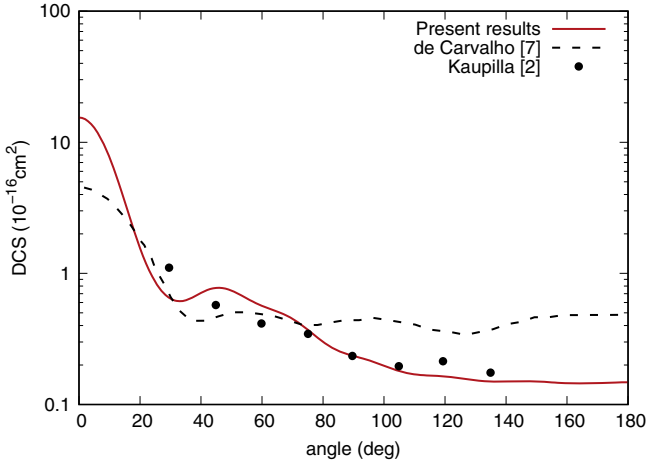

Fig. 5. Differential cross sections for positron collisions with C_2H_2 at 20.0 eV. Theories are: solid red line – present results; short dashed black line – de Carvalho et al. [7]. The solid black circles are the measurements of Kauppila et al. [2] normalized to the present calculations at 90 degrees.

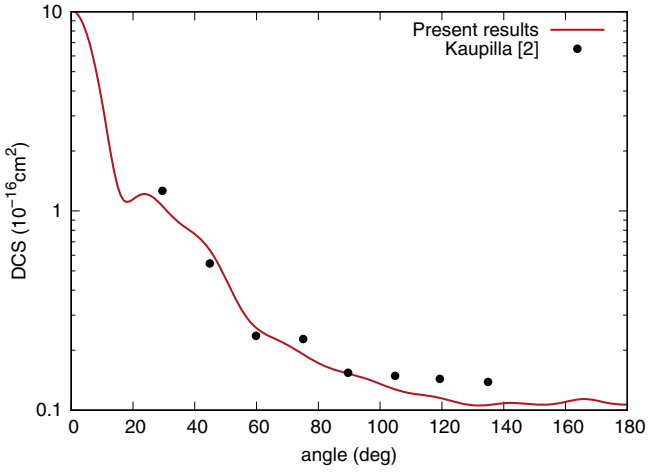

Fig. 6. Differential cross sections for positron collisions with C_2H_2 at 50.0 eV. Solid red line – present theoretical results. The solid black circles are the measurements of Kauppila et al. [2] normalized to the present calculations at 90 degrees.

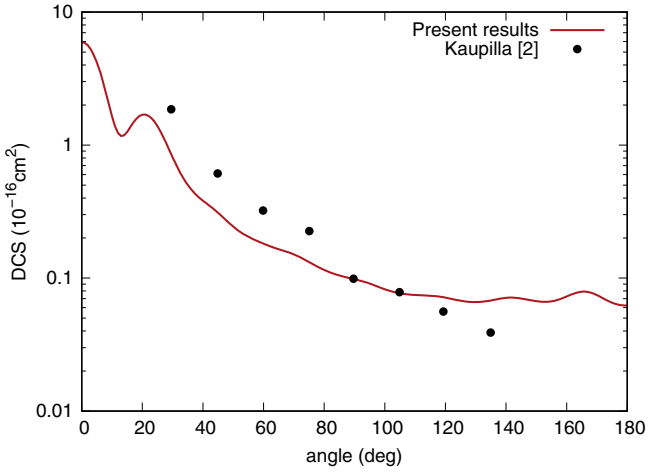

Fig. 7. Same as figure 6, but for 100.0 eV.

seems to be the case for positrons with incident energy of 10 eV for all theoretical approaches, as we can see in Table 1 and Figure 4. For this particular energy we obtained the best average absolute differences $\langle D \rangle = 0.095$, which means a very good compromise between our theoretical approach and the qualitative behaviour of the experimental data.

The increase in the incident positron energy implies the need for a better partial wave expansion of the scattering wave function. Even that our calculations were carried with $l = 14$, more partial waves are needed in order to satisfactorily describe the differential cross sections for energies higher than 10 eV, as we can see in Figures 5–7. The values of $\langle D \rangle$ for these curves are, respectively, 0.208, 0.143 and 0.130. It is expected that the inclusion of inelastic channels via model approaches will not change the comparative aspect of these curves, since the experimental data are quasi-elastic, this is, the positronium formation, ionization and electronic excitation do not contribute to the available experimental data, while the rotational and vibrational cross sections contributions are expected to be small. However, the inclusion of the inelastic processes as positronium formation, direct ionization and electronic excitation from a coupled channel formalism may produce relevant effects over the elastic scattering cross sections, which in principle could not be well evaluated from a model approach that disregards coupling effects close to the inelastic thresholds. Even that the inelastic cross sections obtained via model approaches are relevant and give valuable insight to the analysis of the scattering dynamics as we can observe in the work of Singh and Antony [13] for C_2H_2 and other small hydrocarbons, the coupling between scattering channels should be considered carefully, which may lead to improvements on the calculated cross sections lowering even more the presented $\langle D \rangle$ values. Nevertheless, such effects may be superseded by the use of an incomplete expansion of the scattering wave function or

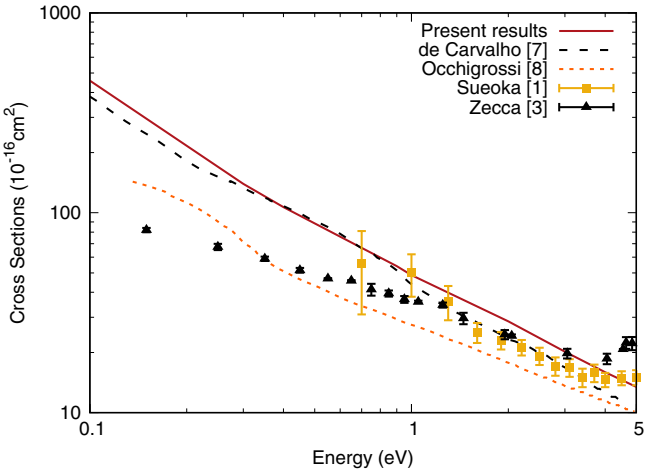

Fig. 8. Comparison between the present theoretical results, other theoretical elastic integral cross sections and experimental data for positrons collisions with C_2H_2 up to 5.0 eV. Solid red line – present theoretical elastic integral cross sections; short dashed black line – theoretical elastic integral cross sections of de Carvalho et al. [7]; dotted orange line – theoretical elastic integral cross sections of Occhigrossi and Gianturco [8]; solid yellow squares – experimental total cross sections of Sueoka and Mori [1]; solid black triangles – experimental total cross sections of Zecca et al. [3].

the employment of an inadequate polarization interaction. We expect that an improved (an ab initio formulation or the completion of the interaction with higher order polarizabilities) correlation polarization model may enrich the description of the qualitative behaviour of the differential cross sections, when combined with a more complete expansion of the scattering wave function, such as the calculations for positron scattering with O_2 molecules [20]. It is important to notice here that as the O_2 molecule is much smaller in size when compared to C_2H_2, a $l = 14$ partial wave expansion may be sufficient for energies up to 100 eV, however this is not the case for the C_2H_2 molecule even for 20 eV, as we can conclude from Figures 5–7.

Our integral elastic cross section (ICS) is shown in Figure 8. We compare it to the absolute measurements of Sueoka and Mori [1] and Zecca et al. [3] and to the calculated elastic integral cross sections of de Carvalho et al. [7] and Occhigrossi and Gianturco [8]. We omitted the other theoretical results in this figure for sake of clarity, but the comparison between theories is easily performed putting Figures 1 and 8 side by side. The comparison of our curve to the elastic cross sections at 4 eV indicates that the normalization scheme adopted to compare the relative differential cross sections may have brought those values very close to the absolute ones. However, this can only be evaluated with greater precision through an experimental effort for the determination of the absolute differential cross sections. At this energy range, our cross sections compare very well with the mentioned experimental data and the ab initio calculations of [7] and model calculations of [8].

Since we employ the same correlation polarization model, the differences between our results and the elastic integral cross sections given by Occhigrossi and Gianturco [8] may be justified by the employment of somewhat different wave functions for the target. For instance, different sets of basis functions produce different electronic densities, mainly in the valence region of the molecule where the cutoff radius is defined. Such subtlety can generate appreciable changes in the calculated cross sections. Nevertheless, we are unable to presently study these discrepancies any further. A good indication that the present results are improved in respect to those of Occhigrossi and Gianturco is the very good comparison between our calculations and the DFT results of Franz et al., which are based in a PCOP approach as well, however no further details of that calculation are available.

The comparison between the experimental TCS of Zecca et al. [3] for the very low energy range and the present theoretical results can be improved by the addition of forward scattering effects corrections, as performed by Sueoka and Mori [1] and explicitly presented by Kimura et al. [21]. For this particular molecule, the procedure of correcting the experimental TCS considering the electron elastic differential cross sections as done for positrons collisions with H_2O [21,22] may not give reasonable results, since the electron C_2H_2 cross sections present a pronounced $^2\Pi_g$ resonance around 2.5 eV. This feature dramatically affects the differential cross sections included in the spectrometer missing angle region, that may be as high as 17.5 degrees at 1 eV as mentioned by Zecca et al. [3]. Also, C_2H_2 is a nonpolar molecule, which means the very low angle differential cross sections are not dominated by the dipole interaction, as is the case with the H_2O molecule.

A good agreement between theoretical elastic integral cross sections is important in order to establish the basis for more complete calculations of positrons collisions with C_2H_2. A possible project for future works could be to implement an absorption potential [23] in order to model the total cross sections in a more complete energy range in which experimental data are available.

4 Conclusions

We calculated elastic integral and differential cross sections for positron collisions with C_2H_2 in the static plus correlation polarization approximation. The present theoretical results are in excellent qualitative agreement with the experimental data of Kauppila et al. [2] and Sueoka and Mori [1]. Comparison with the measurements of Zecca et al. [3] may be improved by the inclusion of forward scattering effects. The comparison between the theoretical and experimental differential cross sections attest some accuracy for the correlation model adopted. For future studies, we may possibly implement an absorption potential in order to calculate the total cross sections up to 400 eV for direct comparison with the available experimental data. Improvement in the partial wave expansion may be necessary, as we can notice from higher energies DCS's. Also, we intend to verify further improvements in the description of the elastic cross sections for positron collisions with C_2H_2 by the inclusion of higher order polarizabilities in

the scattering potential model, as proven very relevant for positron collisions with O_2 [20].

W. Tenfen would like to thank Universidade Federal de Pelotas for support. M.V. Barp and F. Arretche would like to thank the Programa de Pós-Graduação em Física of Universidade Federal de Santa Catarina for the support. M.V. Barp would like to thank the Conselho Nacional de Desenvolvimento Científico e Tecnológico for financial support.

Author contribution statement

All authors contributed equally to this work.

References

1. O. Sueoka, S. Mori, J. Phys. B: At., Mol. Opt. Phys. **22**, 963 (1989)
2. W.E. Kauppila, C.K. Kwan, D.A. Przybyla, T.S. Stein, Nucl. Instrum. Methods Phys. Res. B **192**, 162 (2002)
3. A. Zecca, L. Chiari, A. Sarkar, M.J. Brunger, New J. Phys. **13**, 115001 (2011)
4. J.P. Sullivan, C. Makochekanwa, A. Jones, P. Caradonna, D.S. Slaughter, J. Machacek, R.P. McEachran, D.W. Mueller, S.J. Buckman, J. Phys. B: At., Mol. Opt. Phys. **44**, 035201 (2011)
5. E.P. da Silva, J.S.E. Germano, J.L.S. Lino, C.R.C. de Carvalho, A.P.P. Natalense, M.A.P. Lima, Nucl. Instrum. Methods Phys. Res. B **143**, 140 (1998)
6. C.R.C. de Carvalho, M.T. do N. Varella, M.A.P. Lima, E.P. da Silva, J.S.E. Germano, Nucl. Instrum. Methods Phys. Res. B **171**, 33 (2000)
7. C.R.C. de Carvalho, M.T. do N. Varella, M.A.P. Lima, E.P. da Silva, Phys. Rev. A **68**, 062706 (2003)
8. A. Occhigrossi, F.A. Gianturco, J. Phys. B: At., Mol. Opt. Phys. **36**, 1383 (2003)
9. J. Franz, F.A. Gianturco, K.L. Baluja, J. Tennyson, R. Carey, R. Montuoro, R.R. Lucchese, T. Stoecklin, P. Nicholas, T.L. Gibson, Nucl. Instrum. Methods Phys. Res. B **266**, 425 (2008)
10. R. Zhang, P.G. Galiatsatos, J. Tennyson, J. Phys. B: At., Mol. Opt. Phys. **44**, 195203 (2011)
11. K.L. Baluja, A. Jain, Phys. Rev. A **45**, 7838 (1992)
12. R. Raizada, K.L. Baluja, PRAMANA **46**, 431 (1996)
13. S. Singh, B. Antony, Phys. Plasmas **25**, 053503 (2018)
14. A. Jain, F.A. Gianturco, J. Phys. B: At., Mol. Opt. Phys. **24**, 2387 (1991)
15. J. Horáček, T. Sasakawa, Phys. Rev. A **28**, 2151 (1983)
16. E.M.S. Ribeiro, L.E. Machado, M.-T. Lee, L.M. Brescansin, Comput. Phys. Commun. **136**, 117 (2001)
17. M.W. Schmidt, K.K. Baldridge, J.A. Boatz, S.T. Elbert, M.S. Gordon, J.H. Jensen, S. Koseki, N. Matsunaga, K.A. Nguyen, S. Su, T.L. Windus, M. Dupuis, J.A. Montgomery, J. Comput. Chem. **14**, 1347 (1993)
18. T.H. Dunning Jr., J. Chem. Phys. **53**, 716 (1971)
19. D.R. Lide et al., *CRC handbook of Chemistry and Physics* (CRC Press, Boca Raton, 2012)
20. W. Tenfen, M.V. Barp, F. Arretche, Phys. Rev. A **99**, 022703 (2019)
21. M. Kimura, O. Sueoka, A. Hamada, Y. Itikawa, Adv. Chem. Phys. **111**, 537 (2000)
22. C. Makochekanwa, A. Bankovic, W. Tattersall, A. Jones, P. Caradonna, D.S. Slaughter, K. Nixon, M.J. Brunger, Z. Petrovic, J.P. Sullivan, S.J. Buckman, New J. Phys. **11**, 103036 (2009)
23. D.D. Reid, W.B. Klann, J.M. Wadehra, Phys. Rev. A **70**, 062714 (2004)