Positron scattering from ethane: Elastic and inelastic scattering

Z. Cheong, D. Stevens, and J. P. Sullivan*
Positron Research Group, Research School of Physics, Australian National University, Canberra, Australian Capital Territory 2601, Australia

(Received 7 May 2024; accepted 26 July 2024; published 20 August 2024)

This paper presents measurements of scattering of positrons from ethane. Grand total, total elastic, total positronium formation, total ionization, and angular differential elastic-scattering cross sections have been observed and are shown along with comparisons to previous experimental and theoretical values. There are a number of discrepancies with previous data and some discussion of these and possible explanations are presented. This wider range of scattering measurements provides a more comprehensive picture of positron scattering from this target and then allows us to suggest pathways to resolution of the existing disagreements. However, it is clear that there is significant further work to be done before we can consider that we have a full understanding of the scattering process from this relatively simple target species.

DOI: 10.1103/PhysRevA.110.022815

I. INTRODUCTION

Experimental positron scattering is a useful tool to both probe antimatter-matter interactions and test our understanding and theoretical descriptions of low-energy charged-particle scattering more generally. When compared to electron scattering, the opposite charge and lack of exchange processes allow for elements of calculation techniques to be isolated and tested, indicating areas of potential improvement for future work. However, it must be noted that once the positronium formation channel is open, this presents a more serious challenge to theoretical description, with the lack of a clearly defined scattering center complicating the theoretical description of the scattering process. In addition to this, positron interactions with small hydrocarbons, such as ethane, have been shown to exhibit interesting phenomena, exemplified by the observation of scattering resonances in the annihilation cross section at very low energies [1]. These resonances indicate the presence of positron bound states to neutral molecules and are present in many hydrocarbon species with a large number of targets investigated [2].

In the present work we examine positron scattering from ethane over the energy range from 1 to 50 eV. Ethane is a nonpolar molecule, with a polarizability of 4.226 $Å^2$ [3] and ionization potential of 11.52 eV [3], which gives the positronium formation threshold energy of 4.72 eV. In principle, the lack of a permanent dipole moment simplifies the calculation of positron scattering processes, but recent work has shown that this is not always the case, with serious disagreement remaining for both ring [4] and linear hydrocarbon molecules [5]. Even at energies below the positronium formation threshold, discrepancies between experiment and theory persist for scattering from simple molecular targets.

Using a Surko buffer gas trap [6] combined with techniques developed to measure scattering processes in a strong magnetic field [7], we present measurements of grand total scattering cross sections (GTCSs), elastic total and differential cross sections (DCSs), and ionization and positronium formation cross sections. Measurements of electronic excitation showed that cross sections were below the precision limit of the current apparatus. The present data are compared to a range of previous experimental and theoretical work.

II. EXPERIMENTAL PROCEDURE

Experiments were performed on the positron scattering apparatus at the Australian National University. This experiment has been described in detail previously [8] and only brief details will be given here. Positrons were obtained from a ^{22}Na radioactive source, which had a strength of approximately 38 mCi for the work presented in this paper. Emitted positrons were moderated using solid neon, providing a low-energy beam confined radially using a strong magnetic field (approximately 500 G). This beam was directed into a Surko trap system [6], which trapped and cooled the positrons before releasing them as a pulsed low-energy positron beam [9], with an energy spread of 50 meV (FWHM). The beam was directed to a cell of well-defined length which contained the ethane target, with the pressure controlled using a needle valve and measured using a high-precision capacitance manometer. After passing through the target region, the beam continued to a retarding potential analyzer and microchannel plate detector system, which measured the parallel energy distribution after interaction with ethane [10]. Absolute cross sections could be obtained for a range of scattering processes by analyzing the distribution of the parallel energy, with absolute cross-section normalization provided through the value of the pressure measurement and the length of the scattering cell [7].

*Contact author: james.sullivan@anu.edu.au

TABLE I. Missing angular range for forward-scattered elastic positrons in the present measurements.

Energy (eV)	Minimum angle (deg)
1	33
2	23
5	14
10	10
20	7
50	4.5

In the case of measurements using a transmission apparatus, some portion of the scattering remains undetected, due to the inability to discriminate forward-scattered positrons from the incident beam. In the present case, this results in some of the elastic scattering being excluded from the measurements. The amount of missing scattering is characterized by the forward angle acceptance, which in turn is a function of the energy resolution of the beam in the case of strong magnetic confinement. This leads to the undercounting of scattered positrons and hence the GTCS and total elastic cross sections are only strictly lower limits of the true cross section [11]. It also provides a limit on the lowest angle able to be measured in the DCS. The missing angle limits are presented for various energies in Table I. Note that this has no effect on inelastic-scattering measurements or on positronium formation cross sections. In addition, in the current measurements vibrational and rotational excitations are included in the elastic-scattering component, which is averaged over these contributions. We anticipate that the cross sections are likely to be much smaller than the elastic scattering and thus only make a small contribution to the total cross section. Recent measurements show the magnitude of vibrational excitation cross sections for positron scattering from ethane to be on the order of 1 $Å^2$ [12].

The DCSs presented in this work are folded around 90°, due to the reflection of backscattered positrons and retransmission through the scattering cell (this also results in missing a corresponding range of backward-scattering angles in the total cross-section measurements). Systematic effects arising from this are minimized by keeping the total scattering inside the cell to less than 10% for the differential cross-section measurements. At energies above the first electronic excited state (approximately equal to 7.6 eV [13]), a magnetic-field ratio was used to separate elastic scattering from electronic excitation and ionization. This technique is explained in detail by Sullivan *et al.* [7]. It should be noted that the lowest angle presented in these measurements is somewhat higher than the stated minimum angle in Table I. This is due to the details of the measurement technique, which relies on the difference between successive transmission measurements. More detail can be found in previous papers describing the measurement procedure [7,10].

Uncertainty in the cross section comes from the pressure measurement, uncertainty in the cell length, and the statistical uncertainties in the measurement of transmitted positrons. The errors presented in this paper are a combination of all three, with the primary contribution from the statistics of the measurement.

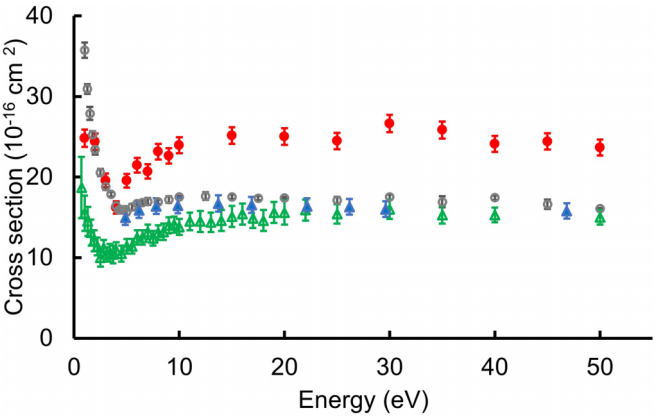

FIG. 1. Grand total cross-section measurements for positron scattering from ethane. Red closed circles denote present data; gray open circles, data from Chiari *et al.* [14]; blue closed triangles, data from Floeder *et al.* [15]; and green closed triangles, data from Sueoka and Mori [16].

III. GRAND TOTAL CROSS SECTION

The GTCS for positron scattering from ethane has been measured previously by several different groups [14–16], and the present data are compared to these measurements in Fig. 1. Tabulated values are given in Table II. At first glance, there appears to be a wide discrepancy between all the measurements, with the only area of substantial agreement being between the three previous measurements above 30 eV. The current values lie substantially above previous measurements, with the exception of the data from Chiari *et al.* and Floeder *et al.*, at impact energies below 5 eV. However, in making any comparisons, one needs to consider the different angular resolutions

TABLE II. Data for the GTCS for positron scattering from ethane.

Energy (eV)	GTCS ($Å^2$)	Error ($Å^2$)
1	24.81	1.09
2	24.39	1.01
3	19.57	0.85
4	16.22	0.75
5	19.55	0.85
6	21.45	0.91
7	20.68	0.89
8	23.14	0.97
9	22.64	0.95
10	23.96	0.99
15	25.14	1.03
20	25.03	1.03
25	24.47	1.01
30	26.64	1.08
35	25.84	1.05
40	24.10	1.00
45	24.42	1.01
50	23.65	0.98

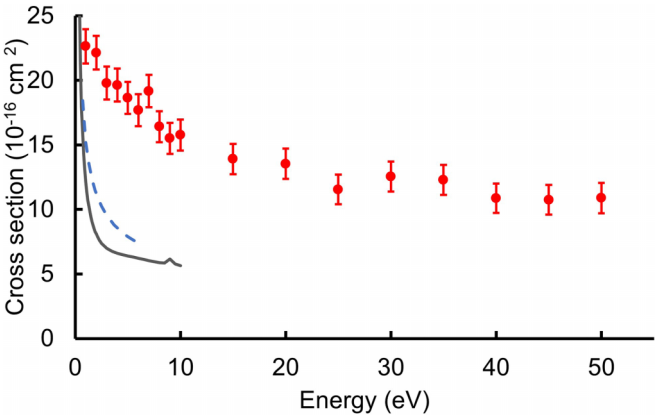

FIG. 2. Elastic total cross section for positron scattering from ethane. Red closed circles are the present experimental data, which are compared to the calculations of Chiari *et al.* [14] (gray solid line) and Occhigrossi and Gianturco [17] (blue dashed line). Both calculations are substantially lower than the measurements over the range of comparable energies.

of all the experiments, as described in the preceding section. It is not then as clear that the disagreements are as bad as they first appear. The stated angular resolutions of the two earliest measurements are substantially worse than that for the current data, which would naturally lead to the measured values from these experiments being smaller (as more of the forward angle scattering is excluded from the cross-section measurement). However, it is not possible to straightforwardly resolve the difference between the present data and that of Chiari *et al.*, who in fact claim a somewhat better angular resolution than we have in the current measurements. Nevertheless, the comparison between the two most recent data sets is consistent with previously published comparisons [5], where the data from Frighetto *et al.* lie substantially above those measured by the experiment of Chiari *et al.* at energies above the positronium formation threshold. Possible reasons for this are discussed in the next section with regard to the differential cross-section measurements.

IV. ELASTIC SCATTERING

Present measurements of total elastic scattering are shown in Fig. 2, compared to previous Schwinger multichannel (SMC) method calculations presented by Chiari *et al.* [14] and theory from Occhigrossi and Gianturco [17]. The experimental data from these measurements are presented in tabulated form in Table IV. Again, we see that the present measurements are much higher than the calculations, over the range of comparison up to 10 eV. This large discrepancy between experiment and theory was also observed in the study by Chiari *et al.* and attributed (at least in part) to the fact that experimental data included inelastic processes, including rotational, and vibrational and electronic excitations.

However, we can see here that when the elastic channel is isolated experimentally (albeit averaged over vibrational and rotational excitations), there remains a large discrepancy between theory and experiment. Low-energy vibrational excitation cross sections are on the order of 1 Å^2 or less [12] and it would be expected that rotational excitations are much the same, with the magnitude of these excitation processes decreasing as the scattering energy increases. The comparison with theory is something that is consistent with previous observations for nonpolar hydrocarbon molecules for both the SMC and *R*-matrix approaches (see, for instance, [4]), although more recent calculations seem to reduce the disparity between the SMC calculations and measurement [5]. The current measurements of elastic scattering are much closer to the GTCS results presented by Chiari *et al.*, in the region above 10 eV.

Differential cross-section measurements are presented in Figs. 3 and 4, at selected energies between 1 and 15 eV, with the tabulated data shown in Table III. The data here are

TABLE III. Data for the DCS measurements of positron scattering from ethane. Cross sections are folded around 90°, with the scattering angle given in degrees and cross-section units and errors in Å^2 sr^{-1}.

1 eV			2 eV			5 eV			10 eV			15 eV		
Angle	DCS	Error	Angle	DCS	Error	Angle	DCS	Error	Angle	DCS	Error	Angle	DCS	Error
84	0.47	0.19	85	0.01	0.26				85	0.15	0.67			
79	0.60	0.26	80	0.61	0.28	81	0.41	0.18	80	0.33	0.74	80	−0.20	−0.32
75	0.73	0.23	75	0.37	0.29	75	0.79	0.34	75	0.77	0.75	75	0.87	0.32
70	1.10	0.24	70	0.56	0.29	70	0.47	0.34	70	0.93	0.74	70	0.64	0.33
65	2.00	0.25	65	0.84	0.30	65	1.06	0.34	65	0.40	0.76	65	−0.04	−0.34
60	2.77	0.26	60	1.10	0.31	60	1.29	0.36	60	0.84	0.80	60	1.23	0.36
55	4.25	0.29	55	2.15	0.34	55	0.98	0.39	55	1.03	0.86	55	0.60	0.38
50	4.84	0.32	50	2.49	0.36	50	1.00	0.42	50	0.57	0.90	50	0.86	0.40
45	6.70	0.35	45	3.70	0.40	45	1.87	0.44	45	1.36	0.96	45	0.36	0.43
40	8.92	0.42	40	5.19	0.43	40	2.11	0.50	40	1.69	1.09	40	1.18	0.48
35	11.09	0.49	35	6.38	0.49	35	3.32	0.55	35	0.27	1.26	35	0.94	0.53
30	13.17	0.56	30	9.89	0.59	30	6.29	0.63	30	1.84	1.42	30	0.92	0.61
25	17.87	0.70	25	12.72	0.71	25	8.24	0.75	25	5.83	1.66	25	2.88	0.72
20	26.09	0.96	20	17.60	0.89	20	17.71	0.93	20	10.16	2.03	20	5.83	0.90
			15	34.34	1.37				15	19.22	2.66	15	15.16	1.23
												11	39.85	2.44

TABLE IV. Data for the partial total cross sections for positron scattering from ethane. Cross-section units and errors are given in Å^2.

Energy (eV)	Total elastic	Error	Positronium formation	Error	Ionization	Error
1	22.64	1.33	−1.44	0.83	−0.12	1.16
2	22.14	1.31	2.67	0.85	−0.57	1.11
3	19.78	1.28	−0.21	0.84	0.40	1.11
4	19.63	1.28	−2.14	0.84	0.42	1.11
5	18.64	1.25	0.89	0.84	0.09	1.11
6	17.69	1.24	4.39	0.85	−0.26	1.10
7	19.17	1.26	2.52	0.85	0.49	1.10
8	16.41	1.21	7.78	0.88	0.02	1.09
9	15.51	1.20	8.30	0.88	−0.46	1.08
10	15.77	1.20	7.76	0.90	1.44	1.12
15	13.91	1.18	10.13	0.92	1.22	1.09
20	13.53	1.17	8.88	0.88	3.04	1.09
25	11.55	1.14	8.82	0.90	4.75	1.12
30	12.55	1.16	6.98	0.88	7.91	1.14
35	12.28	1.17	7.08	0.89	7.20	1.16
40	10.88	1.14	5.68	0.87	8.46	1.15
45	10.75	1.14	4.70	0.87	8.93	1.18
50	10.88	1.17	3.86	0.87	10.62	1.19

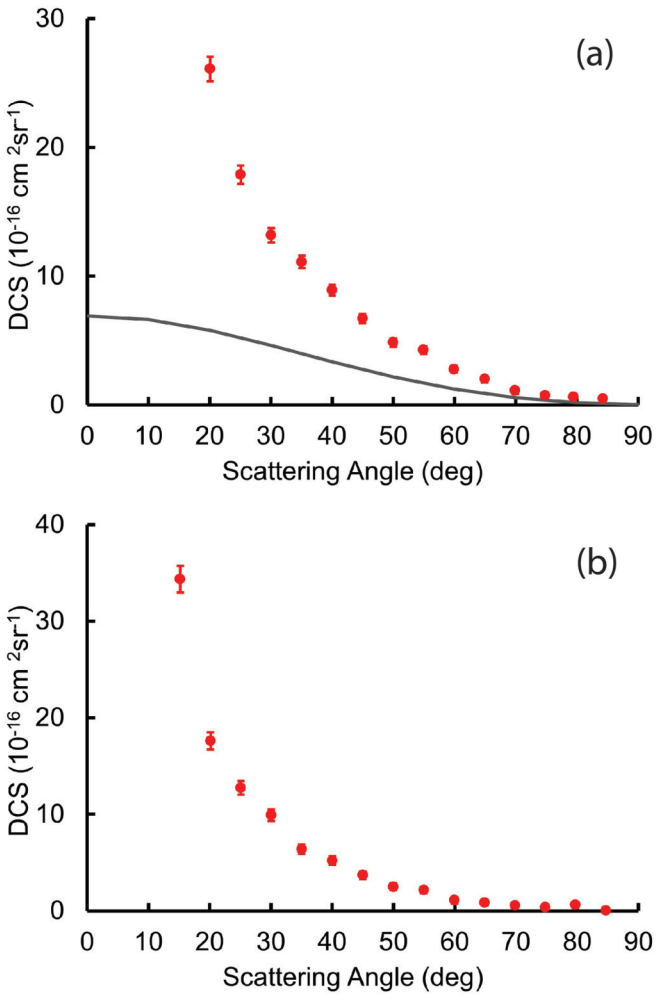

FIG. 3. DCS measurements at (a) 1 eV and (b) 2 eV. The red circles are the present data, compared to the calculations from Chiari *et al.* [14] (gray line) at 1 eV. Note that the measured data are significantly more forward peaked than the theoretical calculations.

compared to the SMC calculations at 1, 5, and 10 eV. As previously noted, the experimental data are effectively folded around 90°, and for this comparison the calculations have likewise been folded. The experimental data in Fig. 3 show the cross section to be strongly forward peaked, rising steeply at the lowest angles measured. The comparison with theory at 1 eV [Fig. 3(a)] shows that this forward angle behavior is not matched by the theory, which is flat at the most-forward-scattering angles. This suggests that the calculation is not adequately taking dipole interactions into account, which are incorporated in this case through virtual single-particle excitations into unoccupied molecular orbitals. However, it should be noted that this technique produced a reasonable estimate of the dipole polarizability of 5.023 Å^2 compared to the experimental value of 4.226 Å^2 [3]. Given that one of the features of positron interactions with ethane is the presence of scattering resonances and bound states, an alternative explanation for the discrepancy may be that there are other correlation processes in play at low energy that remain unaccounted for.

Higher-energy DCSs are presented in Fig. 4, with comparisons between experiment and theory in Figs. 4(a) and 4(b), at energies of 5 and 10 eV, respectively. In this case, at all energies, we see much the same picture, where the strongly forward peaked cross section of the experimental results is not reproduced by the calculation. We also observe that as the scattering energy increases, the DCS becomes progressively more forward peaked. It is harder to imagine that the effect of resonances and bound states is the cause of the disagreement between experiment and theory in this case, given the scale of the energy. However, this disagreement is again consistent with previous comparisons of both SMC and *R*-matrix calculations for similar targets [4]. The strongly forward peaked cross sections also suggest an explanation for the large difference between experimental measurements of the grand total cross sections. If the angular resolution of the experiment by Chiari *et al.* is only slightly worse than stated, the difference in the cross section will be profound, given the strong

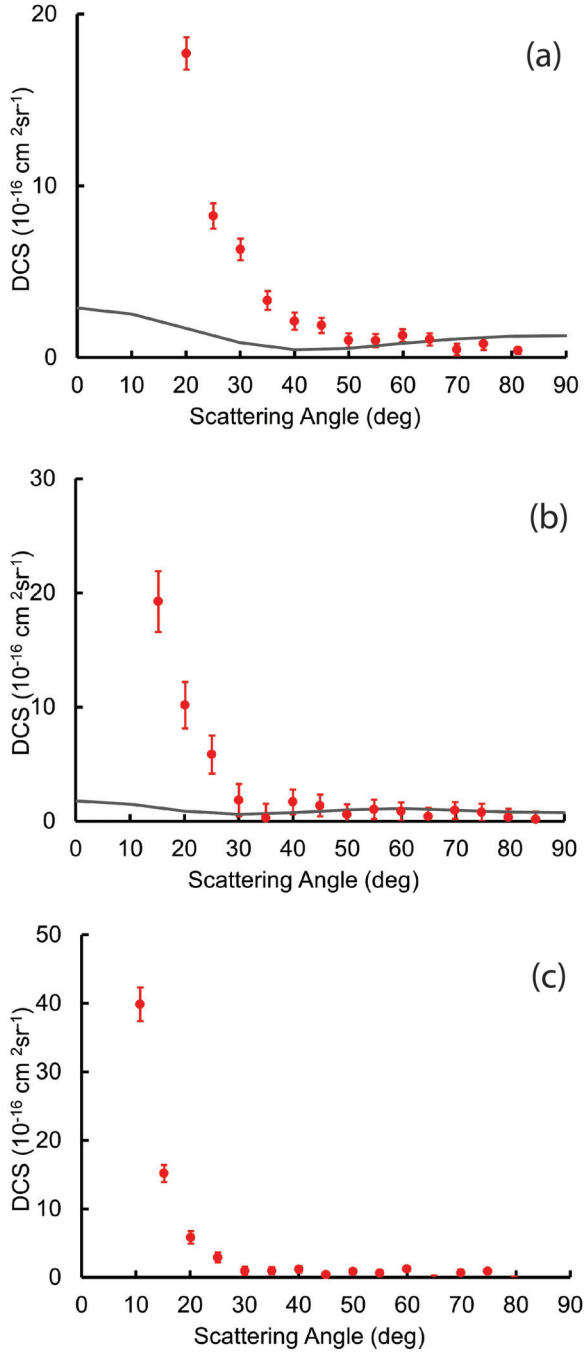

FIG. 4. DCS measurements at (a) 5 eV, (b) 10 eV, and (c) 15 eV. The red circles are the present data, compared to the calculations from Chiari *et al.* [14] (gray line) at 5 and 10 eV. Note that the measured data are significantly more forward peaked than the theoretical calculations.

contribution of forward angle scattering to the total. This would then also be consistent with the differences observed in these measurements and previous measurements from both experimental groups for positron scattering from isopentane [5]. We note that in the more recent calculations using the SMC approach [5], the forward peaked cross section is more closely reproduced by theory, although not quite to the extent of perfect agreement with experimental observation.

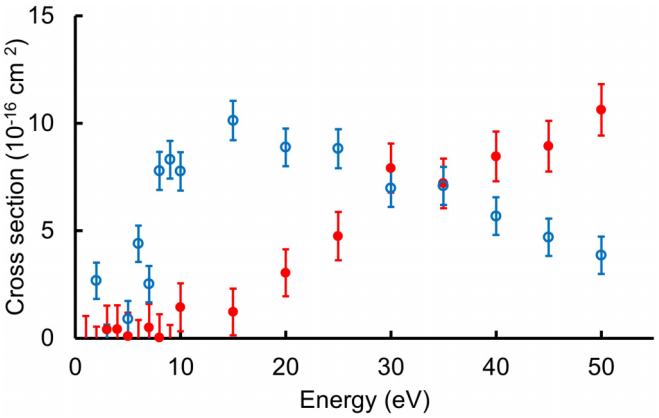

FIG. 5. Positronium formation (blue open circles) and total ionization cross sections (red closed circles) for positron impact on ethane.

V. POSITRONIUM FORMATION AND IONIZATION

Two inelastic total cross sections are presented here, but it should be noted that in the measurements, electronic excitation was also investigated. In this case, the total electronic excitation cross section was consistent with zero to within the limits of the measurements (approximately 0.5 Å^2) across the entire energy range measured.

Positronium formation and ionization total cross sections are both presented in Fig. 5, with the numerical data contained in Table IV. The positronium formation cross section presents a familiar sight, rising to a maximum value of around 10 Å^2 at an energy of 15 eV and falling off again as the scattering energy increases; this shape is universally common to positronium formation cross sections measured to date [18]. The ionization cross section rises slowly from the threshold of 11.52 eV, only becoming significantly nonzero above about 15 eV, as positronium formation declines. It should be noted that at 15 eV, the positronium formation process makes up almost half the grand total cross section, which is again consistent with previous observation. Given the magnitude of this cross section at this energy, it also lends some weight to the idea that previous measurements have underestimated the elastic-scattering component, due to a combination of the strongly forward peaked angular scattering distribution and the experimental angular resolution.

VI. CONCLUSION

The GTCS measurements presented in this paper are significantly higher than previous reported values, in particular for scattering energies above a few eV. However, differences in angular resolution will mean that different experiments report different cross-section values, depending on the shape of the angular differential cross sections and the forward angle discrimination ability of each apparatus [11]. Measurements by Chiari *et al.* [14] estimated a correction of only a few percent for energies of 5 eV and above, based on estimations from calculations of the DCS using the SMC framework. The data presented in this paper include several measurements of the DCS, which show much stronger forward peaking of the DCS than predicted by this calculation, suggesting that any error

due to missing forward angle scattering will be considerably more than this. While the reported angular resolution of the data of Chiari *et al.* is nominally better than that of the current measurements, the disagreement here is consistent with previous comparisons between the two measurement techniques (see, for instance, [5]) and suggests that the angular discrimination in the apparatus used by Chiari *et al.* is somewhat poorer than currently allowed for.

The disagreement between experiment and theory for the DCS is in line with previous comparisons for nonpolar hydrocarbon molecules for both SMC and *R*-matrix methods [4,5], although recent improvement in the SMC calculations appear to go some way to improving the situation [5]. It may be that the effect of positron bound states explains some of the difference between experiment and theory at low energies, but it is clear that an improved description of the target molecule, in particular accounting for molecular polarizability, needs to be implemented to improve the models of positron scattering from ethane and similar targets.

Positronium formation and ionization cross sections have also been measured and contribute significantly to the grand total cross section at energies above their respective thresholds. Electronic excitation is quite small, with no observation of that process at any energy over the range investigated here and it should also be noted that there are no calculations of these processes for positron scattering from ethane. In addition, at an energy of 30 eV, for example, the sum of the positronium formation and ionization cross sections comes to approximately 15 $Å^2$, which is the value of the grand total cross section measured by all previous experiments at this energy. This lends further weight to the contention that the missing forward angle scattering accounts for a much larger contribution to the GTCS than previously appreciated in the case of positron collisions with this target.

In summary, this work presents evidence that it is clear there is further work to be done to improve the description of positron scattering from small molecules, given the large disparity between experiment and theory over the range of the study presented here. In addition, improvements in experimental techniques, in particular allowing for better forward angle resolution when measuring the GTCS, DCS, and total elastic scattering, will be highly beneficial for providing more accurate cross-section values for comparison with improved theoretical calculations.

ACKNOWLEDGMENT

The authors are grateful to R. Tranter for the ongoing technical support of the experimental activities.

[1] S. J. Gilbert, L. D. Barnes, J. P. Sullivan, and C. M. Surko, Vibrational-resonance enhancement of positron annihilation in molecules, Phys. Rev. Lett. **88**, 043201 (2002).

[2] G. F. Gribakin, J. A. Young, and C. M. Surko, Positron-molecule interactions: Resonant attachment, annihilation, and bound states, Rev. Mod. Phys. **82**, 2557 (2010).

[3] NIST Computational Chemistry Comparison and Benchmark Database, edited by R. D. Johnson III (2022), http://cccbdb.nist.gov/.

[4] D. Edwards, D. Stevens, Z. Cheong, V. Graves, J. D. Gorfinkiel, F. Blanco, G. Garcia, M. J. Brunger, R. D. White, and J. P. Sullivan, Positron scattering from pyrazine, Phys. Rev. A **104**, 042807 (2021).

[5] F. F. Frighetto, L. Chiari, J. P. Sullivan, S. J. Buckman, M. H. F. Bettega, S. d'A. Sanchez, A. Souza Barbosa, and M. J. Brunger, Joint theoretical and experimental study on positron scattering by isopentane, Eur. Phys. J. D **78**, 7 (2024).

[6] T. J. Murphy and C. M. Surko, Positron trapping in an electrostatic well by inelastic collisions with nitrogen molecules, Phys. Rev. A **46**, 5696 (1992).

[7] J. P. Sullivan, S. J. Gilbert, J. P. Marler, R. G. Greaves, S. J. Buckman, and C. M. Surko, Positron scattering from atoms and molecules using a magnetized beam, Phys. Rev. A **66**, 042708 (2002).

[8] J. P. Sullivan, A. Jones, P. Caradonna, C. Makochekanwa, and S. J. Buckman, A positron trap and beam apparatus for atomic and molecular scattering experiments, Rev. Sci. Instrum. **79**, 113105 (2008).

[9] S. J. Gilbert, C. Kurz, R. G. Greaves, and C. M. Surko, Creation of a monoenergetic pulsed positron beam, Appl. Phys. Lett. **70**, 1944 (1997).

[10] S. J. Gilbert, J. Sullivan, R. G. Greaves, and C. M. Surko, Low-energy positron scattering from atoms and molecules using positron accumulation techniques, Nucl. Instrum. Methods Phys. Res. Sect. B **171**, 81 (2000).

[11] J. P. Sullivan, C. Makochekanwa, A. Jones, P. Caradonna, D. S. Slaughter, J. Machacek, R. P. McEachran, D. W. Mueller, and S. J. Buckman, Forward angle scattering effects in the measurement of total cross sections for positron scattering, J. Phys. B **44**, 035201 (2011).

[12] Z. Cheong, D. Stevens, and J. P. Sullivan, Positron resonances in vibrational excitation, Phys. Rev. A **109**, 042812 (2024).

[13] W.-Q. Xu, L.-Q. Xu, D.-G. Qi, T. Chen, Y.-W. Liu, and L.-F. Zhu, Investigations of the valence-shell excitations of molecular ethane by high-energy electron scattering, J. Chem. Phys. **148**, 144313 (2018).

[14] L. Chiari, A. Zecca, E. Trainotti, M. H. F. Bettega, S. d'A. Sanchez, M. T. d. N. Varella, M. A. P. Lima, and M. J. Brunger, Cross sections for positron scattering from ethane, Phys. Rev. A **87**, 032707 (2013).

[15] K. Floeder, D. Fromme, W. Raith, A. Schwab, and G. Sinapius, Total cross section measurements for positron and electron scattering on hydrocarbons between 5 and 400 eV, J. Phys. B **18**, 3347 (1985).

[16] O. Sueoka and S. Mori, Total cross sections for low and intermediate energy positrons and electrons colliding with CH_4, C_2H_4 and C_2H_6 molecules, J. Phys. B **19**, 4035 (1986).

[17] A. Occhigrossi and F. A. Gianturco, Low-energy positron dynamics in small hydrocarbon gases, J. Phys. B **36**, 1383 (2003).

[18] J. R. Machacek, F. Blanco, G. Garcia, S. J. Buckman, and J. P. Sullivan, Regularities in positronium formation for atoms and molecules, J. Phys. B **49**, 064003 (2016).