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Cross sections for positron scattering from ethane

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We report experimental and theoretical cross sections for positron scattering from the fundamental organic-chemistry molecule ethane (C_2H_6). The experimental total cross sections (TCSs) were obtained using a linear transmission technique, for energies in the range 0.1–70 eV and with an energy resolution of ~0.25 eV (full width at half maximum). Agreement, over the common energy range, with the earlier TCS measurements of Floeder *et al.* [J. Phys. B **18**, 3347 (1985)] is excellent, while both the present results and those of Floeder *et al.* are consistently higher in magnitude than the data of Sueoka and Mori [J. Phys. B **19**, 4035 (1986)]. The present calculations employed the Schwinger multichannel method and were performed in the static plus polarization approximation for energies up to 10 eV. Our calculated elastic integral cross sections (ICSs) indicate a Ramsauer-Townsend minimum at around 1.4 eV in the A_g scattering symmetry, and a virtual state. In addition we calculated from our scattering cross section a scattering length of $-13.83a_0$. Agreement between our measured TCS and calculated elastic ICS is found to be only qualitative, although this is perhaps not so surprising given the TCS below 10 eV in principle includes contributions from rotational, vibrational, and electronic-state excitation and positronium formation whereas the calculation does not.

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I. INTRODUCTION

The homologous series of compounds C_nH_{2n-2}, C_nH_{2n}, and C_nH_{2n+2} are fundamental in many chemical reactions in polymer chemistry and what is becoming known as "green" chemistry. We have previously reported positron scattering results for ethyne (C_2H_2, i.e., the $n = 2$ member of the C_nH_{2n-2} series) [1] and ethene (C_2H_4, i.e., the $n = 2$ member of the C_nH_{2n} series) [3]. We thus, in this manuscript, conclude our systematic investigation of these compounds by now considering positron scattering behavior for collisions from the $n = 2$ member of the C_nH_{2n+2} series, namely, ethane (C_2H_6)–see Fig. 1. Previous experimental [4,5] and theoretical [6] studies of positron scattering from C_2H_6 have been quite limited. Experimentally, there are total cross section (TCS) results from Floeder *et al.* [4], for incident positron energies in the range 5–400 eV, and Sueoka and Mori [5], for energies between 0.7 and 400 eV. Agreement between these two sets of data is marginal, particularly below about 15 eV, with this discrepancy forming one rationale for the current investigation. Another rationale, for the experimental component of this work, is to extend the available TCS data to lower energies than are currently available. This is particularly important if one is to try to benchmark theory. To the best of our knowledge, the only theoretical computation currently available in the literature is from Occhigrossi and Gianturco [6]. That computation employed a "parameter-free", symmetry-adapted, single-center-expansion approach within a fixed nuclei framework to determine elastic integral cross sections (ICSs) in the energy regime 0.7–6 eV, which were found to be in good agreement with the TCS results of Sueoka and Mori [5], at least below the threshold energy for positronium formation (E_{Ps}) in ethane. Nonetheless it is clear that further theoretical work into positron-ethane scattering is required, which the present calculations hope to address at least in part.

This paper reports experimental total cross sections and theoretical elastic differential and integral cross sections for positron scattering by ethane. The experiment was based on a linear transmission technique [1], covering the positron energies from 0.1 to 70 eV. The calculated fixed-nuclei elastic cross sections (not accounting for positronium formation) were obtained with the Schwinger Multichannel method (SMC) [7,8], with polarization effects, at very low collision energies (0.001 eV) and up to 10 eV. This extension to low energies enabled us to determine the scattering length (a) for the positron-ethane system.

The remainder of the manuscript is organized as follows. In Sec. II we provide the experimental details. Section III discusses the theoretical approach and the computational details employed in our calculations. The results are presented and discussed in Sec. IV, while some conclusions from this study are drawn in Sec. V.

II. EXPERIMENTAL DETAILS

The present measurements were carried out with a positron apparatus developed by Zecca and collaborators, which has already been described in detail in a previous paper [1]. Hence, we just note here that it is based on a linear transmission technique and that the positron beam is generated from a radioactive ^{22}Na isotope (activity of ~1.4 mCi at the time

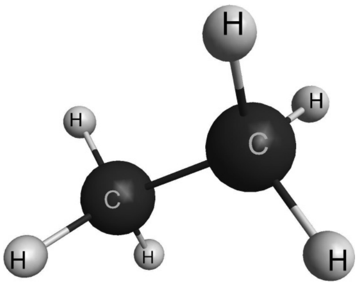

FIG. 1. Geometrical structure of C_2H_6. Generated using McMolPlt [2].

of these measurements), in conjunction with a 1-μm-thick tungsten moderator [9] and a set of electrostatic optics.

In our experiments the beam intensity is attenuated as a consequence of the incident positrons interacting (or not) with the ethane molecules, as described by the Beer-Lambert law:

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

According to Eq. (1), it is possible to determine the required TCS (σ) from measurements of the positron beam count rate with and without the ethane gas in the scattering region (I_1 and I_0, respectively). We also need to measure the pressure in the scattering cell with ethane routed to the scattering region and then measure the pressure when it is diverted into the vacuum chamber [i.e., away from the scattering chamber (P_1 and P_0, respectively)]. The temperature of the ethane gas in the scattering cell (T) is measured by a platinum (PT100) resistance thermometer in thermal contact with the scattering chamber. The length of the scattering cell in our experimental configuration is $L = 22.1 \pm 0.1$ mm, while k in Eq. (1) is Boltzmann's constant.

Several quite standard precautions need to be taken when carrying out the measurements. These include minimizing double scattering events, a condition that is fulfilled by setting the target pressure in the scattering cell such that the beam attenuation (i.e., the ratio I_1/I_0) is greater than 0.7. In addition, note that only a high-purity ethane source (99% from BOC Gases) was used throughout the current measurements. As a standard practice in our laboratory, in order to check for the validity of our techniques and procedures, before any experiment on a new target is started we make preliminary "validation" measurements using targets for which the positron scattering TCSs are now considered to be well known. Such well-characterized systems might be drawn from the noble gases [10–12] and molecular nitrogen [1].

When undertaking measurements at very low energies, such as here, it is crucial for the energy scale to be calibrated accurately. The zero for the incident positron energy scale was determined with a retarding potential analysis (RPA) of the beam, without the target gas in the vacuum chamber, as outlined in Zecca and Brunger [13]. We estimate the error on the energy scale to be ±0.05 eV in this case. The same RPA allows us to also measure the energy distribution of the beam [13] and thus its energy resolution. The energy width of the beam was found to be ~0.25 eV (full width at half maximum) for these experiments, with an uncertainty of at most ±0.05 eV. As in all spectroscopies, our measured cross sections are actually the convolution of the "real TCSs" with the beam energy distribution. This physically means that, once corrected for this effect, the "real TCSs" should be somewhat larger in magnitude than what we measure. However, this correction is expected to be significant only at very low energies (below ~0.5 eV) where the positron energy becomes comparable to the beam width itself. To try and quantify this effect a little more, we used our SMC theory integral elastic cross sections and convolved them with a beam energy-distribution characteristic of our spectrometer. We found that at 0.1 eV the correction, due to the finite energy width of the positron beam, was ~5% in this case.

The measured data also need to be corrected for some instrumental effects that inevitably affect the measurements, before they can be used in Eq. (1). For instance, the length of the scattering region (L) needs to be corrected to account for the increase in the positrons' path length due to the gyration of the particles in the focusing axial magnetic field present in the scattering region. As that magnetic field was $B \sim 11$ G in the present measurements, for positron energies between 0.1 and 30 eV, the value of L increased by 5.5%. For incident energies between 35 and 70 eV the magnetic field was decreased to $B \sim 4$ G, and hence the increase in L was just 2%. We note that the increase in L also depends in principle on the ratio of the transverse to longitudinal component of the positron energy. However, for the typical experimental conditions of our measurements, the angular divergence in our positron beam is possibly smaller than 0.05 rad between 0.1 and 50 eV. Given these conditions, charged particle optics simulations suggest that this effect on the positron path length is an increase smaller than about 0.1% and does not depend on the energy of the positron beam. In addition, the pressure measurements also need to be corrected to account for the thermal transpiration effect. Here the pressure readings were achieved with an MKS 627B capacitance manometer operating at 45 °C, whereas the ethane gas in the scattering cell was held at $T = 65 \pm 2$ °C (note that the vacuum chamber was warmed during our measurements). In this case the thermal transpiration correction was made by following the semiempirical model of Takaishi and Sensui [14] and resulted in a maximum decrease in the absolute value of the TCS of 3.1%.

As with all scattering cell-based linear transmission experiments, the present spectrometer suffers from angular discrimination limitations. They stem from the inability of the detector to distinguish between the positrons that are elastically scattered at very forward angles from those of the primary (unscattered) beam. This effect results in the scattered positron count rate being somewhat overestimated, and therefore, the measured TCSs are somewhat smaller in magnitude than their "true values". At any given energy, the extent of the forward angle scattering effect depends on the angular discrimination of the apparatus and on the nature of the elastic differential cross sections (DCSs) for the target in question in this forward angle region [1]. From the geometry of the scattering and detection regions, the angular acceptance of the Trento apparatus is estimated to be $\Delta\theta \sim 4^\circ$ [1]. This value compares favorably with that from the spectrometers at Yamaguchi University ($\Delta\theta \sim 7^\circ$) and Bielefeld University ($\Delta\theta \sim 5.7^\circ$) [4]. However, it is also known [15] that the gyration of the positrons can also potentially increase the angular discrimination error compared to the no-field case. Using some of the equations detailed in Kauppila *et al.* [16], but for the typical conditions of our experiments, the energy-dependent angular discrimination was evaluated to vary between 17.5° at 1 eV and 2.4° at 50 eV positron energy (see Table II in Zecca *et al.* [1]). Those values, as a function of the positron energy, can then in principle be used in conjunction with the appropriate elastic DCSs, at the same energy as that of the TCS, provided that these are known, to correct the measured TCSs for the forward angle scattering effect. This can be done by following the approach described, for instance, in Hamada and Sueoka [15]. In principle, such DCSs are available from our SMC-level calculations (see next section). However, given that our experimental TCS and the present computed elastic ICS agree only qualitatively, as we see later in Sec. IV, at this time employing those DCSs in the manner outlined above might be premature and so we have in general not done so. Therefore, the TCSs we present here (see Table I) are underestimated with respect to their true values. Nevertheless, to illustrate the forward angle discrimination effect, we have employed the theoretical elastic DCSs (see Sec. III) at 0.1, 0.5, 1, 5, and 10 eV (see Fig. 2 and Table II) to obtain estimates of the TCS corrections that we might expect at those energies. We find that the magnitude of the TCSs we list in Table I increases by ~44% at 0.1 eV, 20% at 0.5 eV, 14% at 1 eV, 2.7% at 5 eV, and 0.7% at 10 eV. Those corrected TCSs are also plotted in Fig. 3, where we also see that the effect is clearly bigger at the lowest energy and becomes smaller as one goes to higher energies.

TABLE I. Present measured TCSs for positron scattering from ethane. The errors given represent the statistical component of the overall uncertainty only.

Energy (eV)	TCS (10^{-20} m^2)	TCS error (10^{-20} m^2) ($\pm 1\sigma$)	Energy (eV)	TCS (10^{-20} m^2)	TCS error (10^{-20} m^2) ($\pm 1\sigma$)
0.10	121.37	5.07	5.00	15.71	0.40
0.20	89.03	4.44	5.50	16.27	0.39
0.30	73.03	1.34	6.00	16.70	0.33
0.40	60.12	1.46	6.50	16.82	0.30
0.50	52.68	0.86	7.00	16.96	0.44
0.60	47.17	1.21	8.00	16.91	0.31
0.70	44.36	1.11	9.00	17.20	0.42
0.80	40.26	0.50	10.00	17.53	0.12
0.90	37.73	1.24	12.50	17.63	0.46
1.00	35.74	0.94	15.00	17.54	0.27
1.25	30.92	0.59	17.50	17.37	0.27
1.50	27.89	0.82	20.00	17.40	0.11
1.75	25.28	0.40	25.00	17.11	0.42
2.00	23.42	0.64	30.00	17.51	0.31
2.50	20.56	0.46	35.00	16.93	0.69
3.00	18.80	0.39	40.00	17.43	0.27
3.50	17.87	0.29	45.00	16.61	0.57
4.00	16.42	0.46	50.00	16.08	0.08
4.25	15.90	0.25	60.00	15.92	0.29
4.50	15.97	0.31	70.00	15.09	0.54
4.75	15.93	0.45			

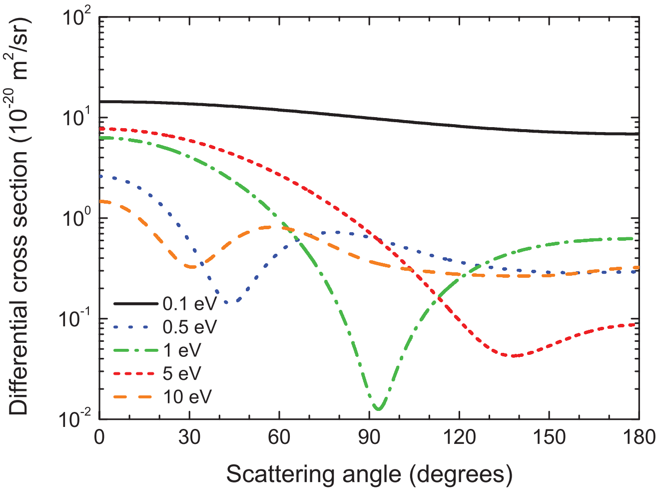

FIG. 2. (Color online) Present positron-ethane theoretical elastic differential cross sections for incident positron energies of 0.1, 0.5, 1, 5, and 10 eV. See legend in figure for further details.

The present measurements on ethane span the energy range between 0.1 and 70 eV. The statistical uncertainties of the data amount to 2.3% on average, but were found to remain between 0.5% and 5% throughout the investigated energy range. Note that the largest errors are usually found at the lowest energies, due to a decreasing positron beam intensity as you go to lower energies. The overall uncertainties of the TCSs are estimated to be within the 5–12% range. They originate from the quadrature combination of quantities like the statistical uncertainties, the uncertainty in the thermal transpiration corrections (<3%), the uncertainty in the value of the length of the scattering region and its correction for the effective positron path length (<3%), and the uncertainties in the pressure and temperature readings (<1% each).

TABLE II. Calculated elastic differential cross sections (in 10^{-20} m^2/sr) for positron scattering from C_2H_6.

Angle (°)	0.1 eV	0.5 eV	1 eV	5 eV	10 eV
0	14.37	7.71	6.30	2.60	1.46
10	14.29	7.48	6.00	2.24	1.17
20	14.05	6.84	5.19	1.41	0.61
30	13.66	5.90	4.06	0.58	0.33
40	13.15	4.81	2.86	0.17	0.47
50	12.55	3.70	1.79	0.21	0.73
60	11.89	2.69	0.97	0.46	0.81
70	11.20	1.85	0.42	0.67	0.67
80	10.51	1.19	0.13	0.72	0.49
90	9.85	0.72	0.02	0.64	0.37
100	9.23	0.40	0.04	0.53	0.32
110	8.68	0.20	0.13	0.43	0.29
120	8.19	0.10	0.25	0.37	0.28
130	7.78	0.05	0.37	0.33	0.27
140	7.45	0.04	0.47	0.30	0.27
150	7.19	0.05	0.55	0.29	0.27
160	7.01	0.07	0.59	0.29	0.28
170	6.90	0.08	0.62	0.29	0.31
180	6.86	0.09	0.62	0.29	0.32

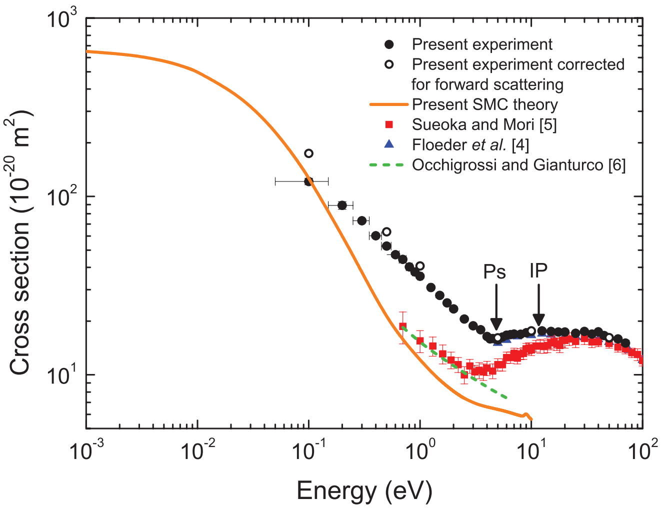

FIG. 3. (Color online) The present measured TCS (●) and calculated elastic ICS (—) for positron collisions with ethane are compared with the previous TCS measurements from Sueoka and Mori [5] and Floeder *et al.* [4] and with the earlier ICS computation by Occhigrossi and Gianturco [6]. The present experimental TCS corrected for the forward angle scattering effect (see text) is also shown at selected energies (○). See also the legend in the figure.

III. THEORY

We employed the SMC method as implemented for positron-molecule collisions to compute the elastic cross sections. This implementation of the SMC method has been described in detail elsewhere [7,8] and here we only provide the relevant points to the present calculations.

The working expression to the scattering amplitude (in the body frame) is

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

where

$$d_{mn} = \langle\chi_m|A^{(+)}|\chi_n\rangle \tag{3}$$

and

$$A^{(+)} = Q\hat{H}Q + PVP - VG_P^{(+)}V. \tag{4}$$

In the above equations, $|S_{\vec{k}_{i,f}}\rangle$ is a solution of the unperturbed Hamiltonian H_0 (the kinetic energy of the incoming positron plus the target Hamiltonian) and is a product of a target state and a plane wave, V is the interaction potential between the incident positron and the electrons and nuclei of the target, $|\chi_m\rangle$ is a set of $(N+1)$-particle configuration state functions (CSFs) used in the expansion of the trial scattering wave function, $\hat{H} = E - H$ is the total energy of the collision minus the full Hamiltonian of the system, with $H = H_0 + V$, P is a projection operator onto the open-channel space defined by the target eigenfunctions, and $G_P^{(+)}$ is the free-particle Green's function projected onto the P space. Finally $Q(= \mathbb{1} - P)$ is the projector onto the closed electronic channels of the target.

The direct space, which corresponds to the static approximation, is composed by CSFs constructed as

$$|\chi_i\rangle = |\Phi_1\rangle \otimes |\varphi_i\rangle, \tag{5}$$

where $|\Phi_1\rangle$ is the target ground state obtained at the Hartree-Fock (HF) level and $|\varphi_i\rangle$ is a single-particle function representing the incoming positron. The set of these single-particle functions is used as scattering orbitals.

To account for the target response to the positron field, polarization effects are incorporated in the calculations by augmenting the direct space by using CSFs of the closed space, which are constructed as

$$|\chi_{ij}\rangle = |\Phi_i\rangle \otimes |\varphi_j\rangle, \tag{6}$$

where $|\Phi_i\rangle$ is obtained by performing single (virtual) excitations of the target. The $|\varphi_j\rangle$ is a single-particle function.

Our calculations were carried out in the static plus polarization approximation, considering the target as belonging to the C_{2h} symmetry group. The ground-state equilibrium geometry, employed in the calculations of electron-ethane collisions [17], was also used in the present bound-state and scattering calculations. The single-particle basis set of Ref. [3] was also used in the present calculations. In Fig. 1 we present the geometrical structure of C_2H_6.

As discussed above, we included the polarization effects in the present calculations through single excitations of the target from the hole (occupied) orbitals to a set of particle (unoccupied) orbitals. Here, we considered excitations from the seven outermost occupied orbitals. To represent the particle orbitals for the A_u, B_g, and B_u symmetries, we generated a set of polarized orbitals from the occupied and virtual orbitals and from the canonical orbital energies [18]. The polarized orbitals are defined by

$$|\varphi_{i,\mu}\rangle = \sum_{j\in\text{virtuals}} \frac{\langle\varphi_j|x_\mu|\varphi_i\rangle}{E_j - E_i}|\varphi_j\rangle, \tag{7}$$

where $|\varphi_i\rangle$ is an occupied orbital, x_μ is a component of the dipole operator, and the sum runs over the HF virtual orbitals. The Schmidt procedure was used to construct an orthonormal set from the polarizing orbitals and the residual scattering orbitals. Besides the polarized orbitals, we also used nine unoccupied orbitals as particle orbitals. The polarized orbitals were used in our calculations to represent both the particle orbitals in electronic excitations and the positron scattering orbitals. All polarized orbitals and the remaining virtual orbitals were used as scattering orbitals. This provided 7682 CSFs for the A_u and B_g symmetries and 8257 CSFs for the B_u symmetry. For the A_g symmetry, we again considered excitations from the seven outermost occupied orbitals and employed improved virtual orbitals (IVOs) [19] to represent the particle and scattering orbitals. We used the lower 73 IVOs as particle orbitals and these orbitals, together with the occupied orbitals, were also used as scattering orbitals. This procedure provided 11081 CSFs for the A_g symmetry. The whole calculation employed 34702 CSFs.

We note that we computed the dipole polarizability, using the sum-over-states method [20], and obtained a value of 5.023 Å^3, in reasonable agreement with one of the experimental values available [21] of 4.226 Å^3.

IV. RESULTS AND DISCUSSION

In Table I and Fig. 3 the results for the present positron-C_2H_6 TCS measurements are given. Note that the errors listed

TABLE III. The present calculated elastic ICS (10^{-20} m^2) for positron collisions with ethane.

Energy (eV)	Elastic ICS (10^{-20} m^2)	Energy (eV)	Elastic ICS (10^{-20} m^2)
0.001	650.65	1.2	10.73
0.002	629.88	1.3	10.20
0.003	610.28	1.4	9.75
0.004	591.77	1.5	9.37
0.005	574.25	1.6	9.04
0.006	557.65	1.7	8.76
0.007	541.90	1.8	8.51
0.008	526.94	1.9	8.29
0.009	512.70	2	8.09
0.01	499.15	2.1	7.91
0.02	392.34	2.2	7.76
0.03	320.27	2.3	7.62
0.04	268.56	2.4	7.50
0.05	229.79	2.5	7.38
0.06	199.74	3	6.99
0.07	175.84	3.5	6.76
0.08	156.43	4	6.61
0.09	140.41	4.5	6.50
0.1	126.98	5	6.40
0.2	60.56	5.5	6.31
0.3	37.60	6	6.22
0.4	26.93	6.5	6.13
0.5	21.26	7	6.04
0.6	17.94	7.5	5.96
0.7	15.79	8	5.89
0.8	14.26	8.5	5.84
0.9	13.09	9	6.15
1	12.15	9.5	5.77
1.1	11.37	10	5.64

in Table I and plotted in Fig. 3 are purely statistical and are at the one standard deviation level. Also plotted in Fig. 3 (and listed in Table III) are the current SMC elastic ICS results, as are the earlier TCS measurements [4,5] and theoretical ICS results [6]. The arrows in Fig. 3 indicate, respectively, the approximate thresholds for positronium formation (Ps) and the direct (first) ionization potential (IP) in ethane. It is known that the first IP of C_2H_6 lies in the range 11.52 eV [21] to 11.65 eV [22], slightly below our estimated value from a HF calculation of 13.25 eV. The above range of IP values lead to a positronium threshold energy of $E_{\mathrm{Ps}} \sim 4.72$–$4.85$ eV as in general

$$E_{\mathrm{Ps}} = V_{\mathrm{i}} - 6.8 \text{ eV}, \tag{8}$$

where V_{i} is the ionization potential. A close examination of the present TCS in Fig. 3 suggests the existence of a quite marked change in its slope at about 4.8 ± 0.7 eV, which we believe corresponds to the opening of the positronium channel. Note that as the present elastic SMC calculations do not incorporate the positronium formation channel, an intrinsically difficult problem due to the multicenter nature of positronium, one would not expect them to compare well with the present measured TCS at energies above about 4.72 eV.

Considering Fig. 3 in more detail, we find that over the common energy range of measurements the present TCSs and those of Floeder *et al.* [4] are in very good accord. Both these experiments give TCSs that are generally systematically larger in magnitude than the results of Sueoka and Mori [5], except perhaps above 15 eV where if the total errors on all three data sets were to be considered then their level of agreement would be satisfactory. We have observed this sort of behavior, where the magnitude of the lower energy TCSs from Trento are significantly larger than those from the Yamaguichi group, in quite a few other scattering systems [1,3,23–25], and so it was not a surprise to observe it again here. We had previously explained this observation in terms of the poorer angular discrimination of the Yamaguichi spectrometer *vis à vis* the Trento spectrometer, implying that the forward angle scattering effect correction to the TCS data from Yamaguichi would be larger than that for the Trento TCS data. Indeed, this is likely to again be playing a part here. However, in this case, another factor might also contribute to the TCS discrepancy at the lower (<15 eV) energies. Namely, the TCS data of Sueoka and Mori [5] suggest a positronium formation threshold of $E_{\mathrm{Ps}} \sim 3.4$ eV which is too low to be physical [it is 1.4 eV lower than the present result of 4.8 ± 0.7 eV and we note that our result is consistent with the "known" values determined using Eq. (8) and independent IP values]. This observation suggests that the energy calibration of Sueoka and Mori [5] might be slightly incorrect, and in fact if we were to shift their TCS data by +1.4 eV in energy the agreement between the results of the two groups would improve. Our conclusion is that the data of Sueoka and Mori [5] for ethane are possibly affected by both a poorer angular discrimination compared to that at Trento and an error in their energy calibration. Perhaps the most striking feature in Fig. 3 is just how dramatically the present TCS increases in magnitude (the same is also true for our SMC elastic ICS) as one goes to lower (below E_{Ps}) incident positron energies (note the logarithmic energy and cross section axes). This behavior reflects the existence of a virtual state for ethane and the moderately strong value for the dipole polarizability ($\alpha = 30.17$ a.u. [26]) that ethane possesses. Our latter statement is also consistent with what was previously found in studies on ethene [3], methane [23], oxygen [27], formaldehyde [28], formic acid [29], and methyl 2-chloropropionate [30], although the picture is complicated a little as those last three species all have quite significant permanent dipole moments.

If we now compare the present measured TCS to our calculated SMC elastic ICS (see Fig. 3), we find only qualitative accord between them. The exception to this appears to be at the lowest common energy (0.1 eV); however that agreement is fortuitous as is apparent when we compare the experimental TCS that has been corrected for the forward angle scattering effect to our elastic SMC result. As the measured TCS includes contributions from rotational excitation, vibrational excitation, electronic-state excitation, and positronium formation (for energies less than 10 eV), as well as elastic scattering, whereas our computation is for the elastic channel only, it is natural to assume that part of this observed magnitude discrepancy is due to those additional open scattering channels in the TCS measurements. However at 1 eV this discrepancy is approximately a factor of 3 in magnitude, and so we do not believe it can entirely be due to the omission of the inelastic channels in our calculation. We thus advance three further possible factors to try and

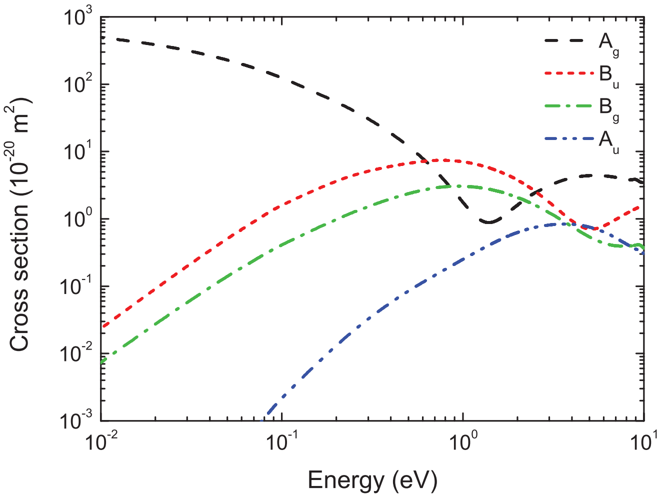

FIG. 4. (Color online) Symmetry decomposition of the elastic ICS for positron scattering from ethane, according to the C_{2h} point group. See also the legend in the figure.

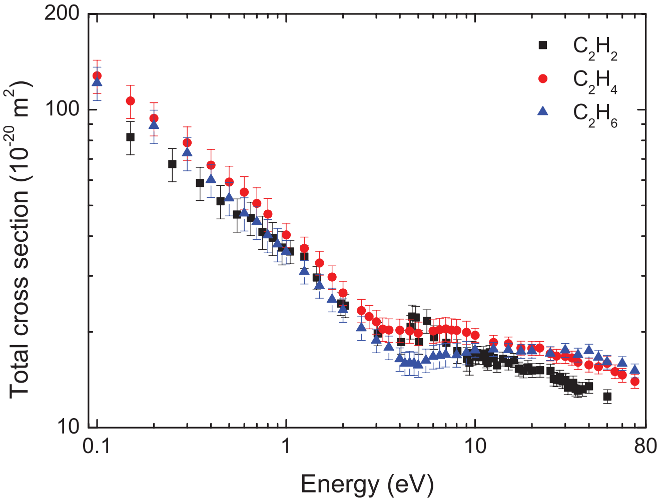

FIG. 5. (Color online) Comparison between the TCSs for positron scattering from C_2H_2 [1], C_2H_4 [3], and C_2H_6, as measured at the University of Trento. The plotted error bars represent the overall uncertainties in the data.

explain, in part, the discrepancies (see Fig. 3) in magnitude between our measurements and computations. The first is that in the experiment the ethane sample exists in a distribution of allowed rotational states, given by the Boltzmann distribution, whereas the computations are for scattering only from the ground rotational level. The second possibility is that, as a result of the radioactive decay process, the emitted positrons are highly polarized. Remarkably this degree of polarization persists after moderation [31], perhaps to as high as about 50%, for a range of metallic moderators [31]. Thus while the present experimental beam of positrons is actually quite highly polarized, in our calculations we are effectively scattering with unpolarized positrons. The third possibility for this discrepancy relates to the current SMC calculation neglecting higher-order correlation effects such as virtual positronium formation, which is known to be important in the positron scattering problem.

Figure 4 shows the symmetry decomposition of the integral cross section. The minimum seen in the cross section of the A_g symmetry corresponds to a Ramsauer-Townsend minimum in the corresponding $\ell = 0$ partial wave. Although not shown here, the s-wave eigenphase crosses zero at the energy of the minimum, indicating that the interaction potential changes from attractive to repulsive when moving from lower to higher energies. This is a common feature between ethane, methane [23], and ethene [3]. Another common feature among these molecules and ethyne [32] is the presence of a virtual state in the ICS (s wave). We employed the equations of Morrison [33] in order to estimate the scattering length for ethane. The computed scattering length for ethane is $-13.83a_0$ (a_0 is the Bohr radius and $1a_0 = 0.529\,18 \times 10^{-10}$ m). This value is greater than the computed value for methane of $-7.4a_0$ [23] and lower than the computed value for ethyne of $-229a_0$ and for ethene of $-47.76a_0$ [3]. Nishimura and Gianturco [34] calculated the scattering length for these systems and obtained $-90.07a_0$ ($-89.94a_0$) for C_2H_2, $-22.00a_0$ ($-21.45a_0$) for C_2H_4, and $-9.01a_0$ ($-8.61a_0$) for C_2H_6 using the modified effective range theory (using their calculated cross sections). Although different in magnitude from the present results, these values follow the tendency of the computed values of $|a_{C_2H_2}| > |a_{C_2H_4}| > |a_{C_2H_6}|$.

Even though there are differences in the molecular structures among C_2H_2, C_2H_4, and C_2H_6, the differences observed in their ICSs at very low energy are quite remarkable [1,3,32]. The magnitude of those differences in the ICSs cannot be easily explained by either the difference in their size or other physicochemical properties, such as the dipole polarizability. The experimental values for the average polarizabilities are in fact 3.487 Å^3 (C_2H_2), 4.188 Å^3 (C_2H_4), and 4.226 Å^3 (C_2H_6) [21]. For such systems, in fact the behavior of the ICS at very low energies is more easily explained from the virtual state.

Finally, in Fig. 5, we plot the experimental TCSs for positron scattering from ethyne [1], ethene [3], and ethane. This plot enables a transparent comparison between the TCSs for these three fundamental organic molecules to be made. Initially, we should reiterate that a physically consistent comparison of the TCSs for these three targets can only be made above ~0.5 eV. This is because the forward angle scattering corrections, which are most important at the lowest energies, may be different in each case. Similarly, the energy resolution of our positron beam might have been a little different between each of the investigations of these molecules, so that the energy-convolution effect, again only significant at the lowest energies, might not be exactly comparable in each system. Notwithstanding those caveats, to within the overall uncertainties of the measured TCSs for each molecule it appears in Fig. 5 that their TCSs are largely consistent with each other until ~4 eV (which roughly corresponds with the opening of the positronium formation channel in each species). This is quite a remarkable result given the differences in their respective physicochemical properties. Above 4 eV and until ~30 eV, no clear pattern in their TCS behavior seems to emerge. However, at energies above ~30 eV the magnitude of the TCSs seems to follow the pattern that we observed earlier in their dipole polarizabilities. Namely, $\mathrm{TCS}_{C_2H_2} < \mathrm{TCS}_{C_2H_4} < \mathrm{TCS}_{C_2H_6}$ just as $\alpha_{C_2H_2} < \alpha_{C_2H_4} < \alpha_{C_2H_6}$. We do not want to read too much into this latter observation, as it is also true that above ∼30 eV this trend in the measured TCSs is also followed by the trend in the hard sphere diameters of the three species and the total number of electrons that each molecule possesses.

V. CONCLUSIONS

We reported on total cross section measurements, which extended the available data for positron-ethane scattering to much lower energies. The present measurements were found to be in good agreement with the earlier results from Floeder *et al.* [4], over the common 5–70 eV energy range. The significant increase in magnitude of the TCS, as one went to lower incident positron energies, was attributed, at least in part, to the relatively strong (large) dipole polarizability of ethane. We also reported results from our SMC computations into positron-C_2H_6 scattering. These included the elastic integral cross section (for energies up to 10 eV) and the scattering length. We noted that only qualitative agreement was found between our calculated elastic ICS and measured TCS, with reasons for this level of accord being given earlier. Nonetheless we acknowledge that some further theoretical development is required, specifically in terms of incorporating a good model for positronium formation into our formalism that will also be computationally tractable.

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[1] A. Zecca, L. Chiari, A. Sarkar, and M. J. Brunger, New J. Phys. **13**, 115001 (2011).
[2] B. M. Bode and M. S. Gordon, J. Mol. Graphics Modell. **16**, 133 (1998).
[3] M. H. F. Bettega, S. d'A. Sanchez, M. T. do N. Varella, M. A. P. Lima, L. Chiari, A. Zecca, E. Trainotti, and M. J. Brunger, Phys. Rev. A **86**, 022709 (2012).
[4] K. Floeder, D. Fromme, W. Raith, A. Schwab, and G. Sinapius, J. Phys. B **18**, 3347 (1985).
[5] O. Sueoka and S. Mori, J. Phys. B **19**, 4035 (1986).
[6] A. Occhigrossi and F. A. Gianturco, J. Phys. B **36**, 1383 (2003).
[7] J. S. E. Germano and M. A. P. Lima, Phys. Rev. A **47**, 3976 (1993).
[8] E. P. da Silva, J. S. E. Germano, and M. A. P. Lima, Phys. Rev. A **49**, R1527 (1994).
[9] A. Zecca, L. Chiari, A. Sarkar, S. Chattopadhyay, and M. J. Brunger, Nucl. Instrum. Methods Phys. Res., Sect. B **268**, 533 (2010).
[10] A. Zecca, L. Chiari, E. Trainotti, D. V. Fursa, I. Bray, and M. J. Brunger, Eur. Phys. J. D **64**, 317 (2011).
[11] A. Zecca, L. Chiari, E. Trainotti, D. V. Fursa, I. Bray, A. Sarkar, S. Chattopadhyay, K. Ratnavelu, and M. J. Brunger, J. Phys. B **45**, 015203 (2012).
[12] A. Zecca, L. Chiari, E. Trainotti, and M. J. Brunger, J. Phys. B **45**, 085203 (2012).
[13] A. Zecca and M. J. Brunger, in *Nanoscale Interactions and Their Applications: Essays in Honour of Ian McCarthy*, edited by F. Wang and M. J. Brunger (Reserch Signpost, Trivandrum, India, 2007), p. 21.
[14] T. Takaishi and Y. Sensui, Trans. Faraday Soc. **59**, 2503 (1963).
[15] A. Hamada and O. Sueoka, J. Phys. B **27**, 5055 (1994).
[16] W. E. Kauppila, T. S. Stein, J. H. Smart, M. S. Dababneh, Y. K. Ho, J. P. Downing, and V. Pol, Phys. Rev. A **24**, 725 (1981).
[17] M. H. F. Bettega, R. F. da Costa, and M. A. P. Lima, Braz. J. Phys. **39**, 68 (2009).
[18] T. N. Rescigno, B. H. Lengsfield, and C. W. McCurdy, in *Modern Electronic Structure Theory,* edited by D. F. Yarkony (World Scientific, Singapore, 1995), Part I, p. 501.
[19] W. J. Hunt and W. A. Goddard III, Chem. Phys. Lett. **3**, 414 (1969).
[20] J. G. Fripiat, C. Barbier, V. P. Bodart, and J. Andre, J. Comput. Chem. **7**, 756 (1986).
[21] http://cccbdb.nist.gov/.
[22] G. Herzberg, *Electronic Spectra of Polyatomic Molecules* (Van Nostrand, Princeton, 1966).
[23] A. Zecca, L. Chiari, E. Trainotti, A. Sarkar, S. d'A. Sanchez, M. H. F. Bettega, M. T. do N. Varella, M. A. P. Lima, and M. J. Brunger, Phys. Rev. A **85**, 012707 (2012).
[24] A. Zecca, L. Chiari, E. Trainotti, A. Sarkar, and M. J. Brunger, PMC Phys. B **3**, 4 (2010).
[25] A. Zecca, L. Chiari, A. Sarkar, K. L. Nixon, and M. J. Brunger, Phys. Rev. A **78**, 022703 (2008).
[26] J. O. Hirschfelder, C. F. Curtiss, and R. B. Bird, *Molecular Theory of Gases and Liquids* (Wiley, New York, 1954), p. 950.
[27] L. Chiari, A. Zecca, S. Girardi, E. Trainotti, G. García, F. Blanco, R. P. McEachran, and M. J. Brunger, J. Phys. B **45**, 215206 (2012).
[28] A. Zecca, E. Trainotti, L. Chiari, G. García, F. Blanco, M. H. F. Bettega, M. T. do N. Varella, M. A. P. Lima, and M. J. Brunger, J. Phys. B **44**, 195202 (2011).
[29] A. Zecca, L. Chiari, A. Sarkar, M. A. P. Lima, M. H. F. Bettega, K. L. Nixon, and M. J. Brunger, Phys. Rev. A **78**, 042707 (2008).
[30] L. Chiari, A. Zecca, S. Girardi, A. Defant, F. Wang, X. G. Ma, M. V. Perkins, and M. J. Brunger, Phys. Rev. A **85**, 052711 (2012).
[31] J. Van House and P. W. Zitzewitz, Phys. Rev. A **29**, 96 (1984).
[32] C. R. C. de Carvalho, M. T. do N. Varella, M. A. P. Lima, and E. P. da Silva, Phys. Rev. A **68**, 062706 (2003).
[33] M. A. Morrison, Phys. Rev. A **25**, 1445 (1982).
[34] T. Nishimura and F. A. Gianturco, Phys. Rev. A **72**, 022706 (2005).