Positron scattering from the isoelectronic molecules $\text{N}_2$, CO and $\text{C}_2\text{H}_2$
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Abstract. We report our results on the experimental total cross sections (TCSs) for positron scattering from the isoelectronic molecules N$_2$, CO and C$_2$H$_2$. Where possible, for each species, comparison is made between the present results and those from earlier measurements and calculations. The agreement between the present and earlier experimental results, within the overall uncertainties on the data, is typically satisfactory for energies greater than about 8 eV, but is only marginal at lower positron impact energies. While N$_2$, CO and C$_2$H$_2$ possess 14 electrons each, we find significant differences in the magnitudes of their respective TCSs and subtle differences in the energy dependence of these TCSs. These details are discussed in depth in this paper.
1. Introduction

There has always been interest among the ATMOP community in trends in scattering cross sections between different molecular species [1]. One such group involves collisions with isoelectronic molecules, in which the main point of inquiry revolves around whether or not the common total charge on each of the species in question predominates in the scattering dynamics, over possible differences between the individual molecular physico-chemical properties, e.g. dipole moments, dipole polarizabilities and so on. An example of this for electron scattering is the work on $\text{N}_2$ and CO by Nickel et al [2], who measured directly the ratio of elastic scattering differential cross sections for these species over a wide range of energies (20–100 eV) and scattering angles (20–120°). They found that the biggest difference from unity was about 20%, with the vast majority of the ratios being within 5% of unity. This leads one to conclude that the weak dipole moment of the CO molecule ($\text{N}_2$ is non-polar) plays a minor role in the scattering process at these energies. This result was later confirmed by the independent measurements of Gibson et al [3] for CO and of Gote and Ehrhardt [4] for $\text{N}_2$. A similar conclusion, although now for vibrational excitation (0–1) in these molecules at energies between 20 and 50 eV, was reached by Middleton et al [5]. In contrast to these findings, in their recent electron impact studies on electronic state excitation in the isoelectronic molecules CO$_2$ [6] and N$_2$O [7], Kawahara et al found significant differences between the measured cross sections. For example at 50 eV, the $^1\Pi$ integral cross section (ICS) in N$_2$O is greater in magnitude than that for the $^1\Pi_u$ state in CO$_2$ by a factor of $\sim$1.5. Similarly, the $D^1\Sigma^+$ ICS in N$_2$O is greater than the ICS for the CO$_2$ $^1\Sigma_u^+$ electronic state by a factor of 3.3. As both N$_2$O and CO$_2$ are linear triatomics with similar dipole polarizabilities, Kawahara et al [7] noted that the observed differences reflect that N$_2$O has a permanent dipole moment, albeit quite weak, while CO$_2$ does not. In addition, at lower energies where shape resonances play an important role in the scattering dynamics, there is little evidence for any striking similarities in the cross section of isoelectronic species [1]. Nonetheless, the notion of trends in scattering cross sections between different molecules is still of interest to the community.

The current knowledge about positron scattering from isoelectronic molecules is much more limited. Indeed, the only work we are aware of that specifically addresses the behaviour of positron scattering from the isoelectronic molecules CO and $\text{N}_2$ and N$_2$O and CO$_2$ is the one by Kauppila et al [8]. In that study they compared measured total cross sections (TCSs) for
Table 1. A selection of the important physico-chemical properties of the isoelectronic molecules N₂, CO and acetylene (C₂H₂) [9–15].

| Property                        | N₂     | CO     | C₂H₂  |
|---------------------------------|--------|--------|-------|
| Dipole moment (µ)               | –      | 0.122 D [15] | –     |
| Dipole polarizability (α)       | 11.74 a₀^3 [12] | 13.09 a₀^3 [13] | 22.8 a₀^3 [9, 10] |
| Positronium formation energy (E_{Ps}) | 8.78 eV [11] | 7.21 eV [11] | 4.6 eV [14] |
| Structure                       | Linear diatomic | Linear diatomic | Linear polyatomic |
| Total number of electrons (Z)   | 14     | 14     | 14    |

Figure 1. Schematic diagram of the idealized structures of N₂, CO and C₂H₂.

H−C≡C−H
N≡N
C≡O

Each of the isoelectronic pairs and found that above about 5 eV, within the overall errors on their data, the TCS for N₂ and CO and those for N₂O and CO₂ were essentially identical. They also reported quasi-elastic angular distributions for these same species, with good agreement being seen between the shapes of the CO and N₂ and N₂O and CO₂ relative differential cross sections down to energies as low as about 20 eV. Here, we therefore report on an extensive series of TCS measurements for positron scattering from the isoelectronic molecules N₂, CO and C₂H₂.

The structures of each of these molecules are schematically represented in figure 1, and some of their most important physico-chemical properties [9–15] are summarized in table 1. At first glance, figure 1 suggests that all three molecules are linear and each contains a triple bond. However, the situation with respect to carbon monoxide is in fact a little more complicated as it actually possesses a double covalent bond and a further dative covalent bond. The bond length in CO is 1.128 Å [16], which is also consistent with it having a ‘partial’ triple bond. The effects of atomic formal charge and electronegativity result in CO having a small permanent dipole moment (see table 1), with its negative end based on the carbon atom. The reason for this, in spite of oxygen having greater electronegativity, is that the highest occupied molecular orbital in CO has an energy much closer to that of the p-orbitals of atomic carbon, meaning that a greater electron density is found near the carbon. In addition, carbon’s lower electronegativity creates a more diffuse electron cloud, thereby enhancing its dipole polarizability relative to say N₂ (see again table 1). This is an important point, as in some of our previous investigations [17–19] the target dipole polarizability played a significant role, particularly at the lower incident positron energies, on the scattering dynamics. In this respect the enhanced value (see table 1) of α for C₂H₂, over those of N₂ and CO, is expected to be significant.

In the next section, we present a brief description of our apparatus and measurement techniques. Following this, our TCS results are presented and discussed. In this case, we
consider sequentially our TCS results for N$_2$, CO and C$_2$H$_2$ and compare them in turn to previous experimental and theoretical data, before finishing with a comparison between the three molecules. Finally, in section 4, the conclusions drawn from the present study are presented.

2. Experimental apparatus and measurement techniques

The positron spectrometer used at the University of Trento was developed by Zecca et al and has been described previously (see e.g. [20]). However, as there has never been an ‘apparatus paper’ on this instrument, we take this opportunity to provide more details than would otherwise be the case. Here, a tungsten or nickel moderator [21] is employed in conjunction with a radioactive $^{22}$Na isotope (activity at that time $\sim 2.4 \text{ mCi}$) and some electrostatic optics to produce the positron beam. The electrostatic optics were originally designed on the basis of the tables and figures in Harting and Read’s book [22], although the practical tips on particle optics design from the unpublished (but accessible) lecture notes of Professor C E Kuyatt were also used. The Trento spectrometer consists of two functional parts: a first stage extracts positrons from the moderator and forms a positron beam at the exit plane of an electrostatic hemispherical (90°) deflector at an energy of 160 eV. From this exit plane, the positrons are subsequently transported and focused by a second stage (see figure 2) onto the scattering cell entrance aperture. As is often the case with source-based positron beams, a deflector is needed to remove those high-energy positrons that passed through the W or Ni film without being moderated. Such positrons are in our spectrometer annihilated in a ‘dump cage’ that resides inside the deflector. We will not further discuss stage 1; rather we will now concentrate on the second stage, which is responsible for the beam characteristics related to the scattering process.

The deflector output aperture acts as the object for all the subsequent stage 2 optics (see figure 2). The goal of this section of the spectrometer was to form a stable and well-focused positron beam tunable over a broad energy range. In order to achieve this the energy ‘scan’ is performed by changing the smallest number of voltages on the optic elements in the second stage. Note that no voltages in the first stage are changed as a result of changing the positron beam energy at the scattering cell entrance. In practice, the current design of stage 2 has enabled us to achieve good positron beam characteristics for beam energies in the range $\sim 0.1–150$ eV. However, in general, most of our measurements have been made at energies $\lesssim 50$ eV, as this is where we believe the positron-related physics is likely to be most interesting.
The preliminary design criteria of the spectrometer were subsequently checked and verified using the ray-tracing facility of SIMION, a commercial charged-particle optics package. Those simulations indicated that a well-collimated and focused positron beam, with good angular divergence characteristics, was formed at the entrance aperture to the scattering cell over the range of positron energies that are of interest (∼0.1–50 eV) for this study. This can clearly be seen in figure 2, where we present a result from our SIMION studies at 1 eV positron energy at the scattering cell entrance aperture. Here seven positrons are ‘flown’ from the exit plane of the hemispherical deflector (E5) to the scattering cell (SC), with elevation angles of 0°, ±0.5°, ±1° and ±1.5° with respect to the axis of symmetry being chosen. These angles seem reasonable, corresponding to results from simulations of stage 1 of the spectrometer for the maximum angular divergence of the positron beam at E5. Various positron-optic elements, for this section of the spectrometer, are labelled as E6, E7, E8, E9 and E10, with the potentials applied to these respective elements being largely equivalent to those employed in the actual experiments. Note that the confining magnetic field (see below) of the present spectrometer will, in practice, only improve the characteristics of the positron beam presented in figure 2. We further note that it is standard practice in our laboratory, as a check of the validity of our techniques and procedures, to make preliminary validation measurements using targets for which the positron scattering TCSs are considered well known. Such ‘benchmarked’ systems might be drawn from the lighter rare gases [23], for example.

The basis of all our linear transmission experiments is the Beer–Lambert law, as defined by

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

where \( I_1 \) is the positron beam count rate at \( P_1 \), the pressure measured with the species of interest routed to the scattering cell, \( k \) is Boltzmann’s constant, \( T \) is the temperature of the gas (K), \( \sigma \) is the TCS of interest, \( I_0 \) is the positron count rate at \( P_0 \), the pressure with the gas diverted into the vacuum chamber, and \( L \) is the length of the scattering region.

For a physical application of equation (1) several crucial precautions should be taken and care must be exercised during the measurements. Those considerations include minimizing double-scattering events and ensuring that the TCSs are pressure independent. In addition, only high-purity N\(_2\), CO and C\(_2\)H\(_2\) target samples were used (BOC gases) in the respective measurements. We note that while the C\(_2\)H\(_2\) sample does contain a very small (<1%) acetone contaminant, to prevent the dissociation of C\(_2\)H\(_2\), our independent TCS measurements for positron scattering from acetone [24] indicate that the effect of such a small contaminant on our C\(_2\)H\(_2\) TCS will be miniscule. Further, note that to minimize any possible impurities affecting our measurements, freeze–pump–thaw cycles were employed here.

The geometrical length of the scattering region is 22.1 ± 0.1 mm, with apertures of 1.5 mm diameter at both the entrance and exit of the scattering chamber. End effects were considered in all our studies; however, it is well known that such effects are minimized [25] if both apertures have small diameters such as in our case. As a consequence, we believe that their contribution to the uncertainty in the value of \( L \) is likely to be less than 0.2%. In our application of equation (1), the value of \( L \) used is always corrected to account for the path increase caused by the gyration of the positrons in the focusing axial magnetic field (typically ∼12 G) present in the scattering region. For our work with N\(_2\), this caused the value of \( L \) to be increased by ∼5%, while in CO and C\(_2\)H\(_2\) the increase in \( L \) was typically ∼6%. From a consideration of the
Table 2. Angular discrimination ($\theta_s$) as a function of positron energy, for the present spectrometer, as calculated using some of the analytic formulae detailed by Kauppila et al [27].

| Positron energy (eV) | $\theta_s$ (deg) |
|----------------------|------------------|
| 0.15                 | 50.8             |
| 0.3                  | 33.2             |
| 0.5                  | 25.1             |
| 1                    | 17.5             |
| 3                    | 10.0             |
| 5                    | 7.7              |
| 7                    | 6.5              |
| 10                   | 5.4              |
| 15                   | 4.4              |
| 20                   | 3.8              |
| 25                   | 3.4              |
| 30                   | 3.1              |
| 40                   | 2.7              |
| 50                   | 2.4              |

size of the entrance and exit apertures of our scattering cell, and their separation, the angular acceptance ($\Delta \theta$) of the Trento spectrometer is $\approx 4^\circ$, which compares favourably with that from the Yamaguchi spectrometer [26] ($\Delta \theta \approx 7^\circ$) and the Detroit apparatus [27] ($\Delta \theta \approx 16^\circ$). The gyration of the positrons can also potentially increase the angular resolution error compared to the no-field case [28]. This can also be corrected for, provided appropriate absolute elastic differential cross sections are available. Unfortunately, results on such differential cross sections are typically unavailable (either theory or experiment) so that the TCSs we report here represent a lower bound on the exact values. Using some of the analytic formulae detailed by Kauppila et al [27], but for the typical experimental conditions of our measurements, estimates of the present energy-dependent angular discrimination can be obtained. We found that they varied from $\sim 17.5^\circ$ at 1 eV positron energy to $\sim 5.4^\circ$ at 10 eV, with a full summary of these results being given in table 2. With these data and an appropriate elastic differential cross section, at each energy, the present TCS could in principle be corrected for the angular discrimination effect. Note that the suggested corrections in table 2 are somewhat smaller than those recently found by Makochekanwa et al [29] in their study of positron scattering from water and formic acid. A thorough, more quantitative discussion of the experimental angular discrimination and its effect on the measured TCSs can be found in [30]; for more details see [30].

It is very important for the energy scale to be calibrated accurately. The zero for the energy scale, in the absence of the target gas, was determined in all our studies with a retarding potential analysis of the beam [31]. Measurements repeated during the last two years show a surprising stability in energy zero (variance $< 0.05$ eV) when a tungsten moderator is used. We now believe that the error in our energy scale calibration is of the order of $\pm 0.1$ eV. The same measurements allow us to evaluate an energy width of the positron beam of $\sim 0.1$ eV (full-width at half-maximum (FWHM)) with a nickel moderator [21] and $\sim 0.3$ eV (FWHM) with a tungsten moderator [21]. Note that in the present study the N$_2$ measurements were made.
with a W moderator, while the CO and C₂H₂ measurements employed a Ni moderator. It is also crucial to accurately measure the scattering cell pressure, which we achieve with an MKS Baratron capacitance manometer. For the N₂ work this was a type 628B manometer with an operational temperature of 100 °C, whereas for the CO and C₂H₂ measurements, we employed a 627B manometer that operated at a temperature of 45 °C. As the manometer temperature was different from that for the target gas of interest in the scattering cell, thermal transpiration corrections to the pressure readings are made using the model of Takaishi and Sensui [32]. For the N₂ measurements this correction was a maximum of 10%, while for CO and C₂H₂ the maximum correction was about 3%. One last caveat on the data we report here should be noted. With an experimental energy resolution of ~0.1–0.3 eV (FWHM), depending on the moderator type [21], at positron energies below ~0.5 eV, the TCSs we report are actually a convolution over this energy resolution. In practice this physically implies that when corrected for this effect, our lowest energy TCSs should be somewhat higher in magnitude than what we present here. Note that the extent of this effect will depend on the actual form or shape of the TCS versus energy dependence and is therefore expected to be species specific.

Finally, we note that the data collection and analysis codes were driven by software developed at the University of Trento for application on a personal computer. The positron energy range of the present TCS measurements was usually ~0.1–50 eV, with the overall errors on our TCS typically being within the 5–12% range. All our measurements were taken under stable positron beam conditions.

3. Results and discussion

The present TCS results on positron scattering from N₂, CO and C₂H₂ are listed in tables 3–5 and plotted in figures 3–5. Also plotted in these figures are the relevant results from corresponding previous experimental and theoretical investigations, for each molecule. Finally, in figure 6, the present N₂, CO and C₂H₂ TCSs are plotted in the figure in order for us to see whether any trends in the data for this isoelectronic series are apparent.

3.1. N₂

For many years, molecular nitrogen (N₂) has served as a type of prototypical system in electron (e⁻) scattering experiments [1]. This is true for both theoretical calculations and experimental measurements, so that in a majority of cases there now appears to be a good number of available cross section data, for the various channels, that are in reasonable agreement with each other [1]. This is the case even for the excitation of the electronic states [33, 34]. As part of this database there exist very accurate e⁻–N₂ TCSs, which serve as a good first test for the validity of the scattering calculations and also as an important cross check of the self-consistency of the sum of the ICSs for the respective open channels, i.e. elastic scattering, vibrational and electronic excitation, etc. It is therefore reasonable to presume that N₂ might fulfil a similar role for positron (e⁺) scattering, particularly with the advent of Penning–Malmberg traps [11] for use in scattering experiments.

Indeed, there has been considerable theoretical effort aimed at describing e⁺–N₂ scattering, at least at the TCS and integral elastic cross-section levels [35–42]. More recently, with the advent of new experimental data [11, 43, 44], the calculations have also been extended to consider the excitation of electronic states [45, 46] and direct ionization [47]. As this paper
Table 3. The present TCSs ($\times 10^{-16}$ cm$^2$) for positron scattering from N$_2$. The errors given represent the statistical uncertainty component only of the overall error. See the text for further details.

| Energy (eV) | TCS ($10^{-16}$ cm$^2$) | TCS error ($10^{-16}$ cm$^2$ (1σ)) | Energy (eV) | TCS ($10^{-16}$ cm$^2$) | TCS error ($10^{-16}$ cm$^2$ (1σ)) |
|------------|-------------------------|-----------------------------------|------------|-------------------------|-----------------------------------|
| 0.11       | 36.85                   | 5.90                              | 8.85       | 4.29                    | 0.12                              |
| 0.13       | 38.56                   | 5.40                              | 8.95       | 4.44                    | 0.08                              |
| 0.15       | 28.84                   | 5.30                              | 9.05       | 4.57                    | 0.15                              |
| 0.18       | 24.79                   | 2.20                              | 9.15       | 4.55                    | 0.13                              |
| 0.20       | 23.98                   | 1.26                              | 9.25       | 4.67                    | 0.19                              |
| 0.22       | 21.90                   | 1.09                              | 9.35       | 4.87                    | 0.20                              |
| 0.25       | 18.92                   | 0.96                              | 9.55       | 4.87                    | 0.19                              |
| 0.35       | 14.59                   | 0.89                              | 9.65       | 4.91                    | 0.04                              |
| 0.45       | 12.45                   | 0.82                              | 9.75       | 4.94                    | 0.13                              |
| 0.55       | 8.70                    | 0.48                              | 10.05      | 5.06                    | 0.12                              |
| 0.65       | 7.83                    | 0.65                              | 10.55      | 5.21                    | 0.15                              |
| 0.75       | 7.62                    | 0.30                              | 11.05      | 5.52                    | 0.14                              |
| 0.85       | 6.36                    | 0.22                              | 11.30      | 5.65                    | 0.15                              |
| 0.95       | 6.18                    | 0.23                              | 11.60      | 5.76                    | 0.20                              |
| 1.05       | 5.61                    | 0.27                              | 12.05      | 5.91                    | 0.13                              |
| 1.35       | 5.06                    | 0.17                              | 12.30      | 5.83                    | 0.19                              |
| 1.55       | 4.73                    | 0.17                              | 12.80      | 6.06                    | 0.18                              |
| 1.65       | 4.99                    | 0.14                              | 13.05      | 6.44                    | 0.15                              |
| 1.95       | 4.66                    | 0.13                              | 13.30      | 6.43                    | 0.16                              |
| 2.05       | 4.66                    | 0.12                              | 14.05      | 6.62                    | 0.16                              |
| 2.25       | 4.45                    | 0.09                              | 15.05      | 7.23                    | 0.14                              |
| 2.55       | 4.44                    | 0.04                              | 16.05      | 7.10                    | 0.13                              |
| 2.85       | 4.40                    | 0.10                              | 17.05      | 7.48                    | 0.15                              |
| 3.05       | 4.37                    | 0.12                              | 18.05      | 7.75                    | 0.14                              |
| 3.55       | 4.47                    | 0.09                              | 19.05      | 7.63                    | 0.13                              |
| 4.05       | 4.29                    | 0.12                              | 20.05      | 8.37                    | 0.05                              |
| 5.05       | 4.11                    | 0.09                              | 21.05      | 8.10                    | 0.15                              |
| 6.05       | 4.26                    | 0.10                              | 23.05      | 8.25                    | 0.12                              |
| 6.35       | 3.99                    | 0.11                              | 25.05      | 8.30                    | 0.15                              |
| 6.65       | 4.01                    | 0.11                              | 27.05      | 8.42                    | 0.20                              |
| 7.05       | 4.17                    | 0.12                              | 27.55      | 7.96                    | 0.23                              |
| 7.30       | 4.06                    | 0.12                              | 29.05      | 8.36                    | 0.22                              |
| 7.55       | 4.23                    | 0.13                              | 31.05      | 8.14                    | 0.29                              |
| 7.80       | 4.16                    | 0.09                              | 31.30      | 7.94                    | 0.30                              |
| 8.05       | 4.27                    | 0.08                              | 33.05      | 8.16                    | 0.34                              |
| 8.30       | 4.18                    | 0.10                              | 35.05      | 8.07                    | 0.32                              |
| 8.35       | 4.27                    | 0.09                              | 37.05      | 8.11                    | 0.28                              |
| 8.55       | 4.32                    | 0.11                              | 40.05      | 7.99                    | 0.36                              |
| 8.75       | 4.29                    | 0.10                              |            |                         |                                   |
Table 4. The present TCSs ($\times 10^{-16}$ cm$^2$) for positron scattering from CO. The errors given represent the statistical uncertainty component only of the overall error. See the text for further details.

| Energy (eV) | TCS $10^{-16}$ cm$^2$ | TCS error (1σ) | Energy (eV) | TCS $10^{-16}$ cm$^2$ | TCS error (1σ) |
|------------|---------------------|----------------|------------|---------------------|----------------|
| 0.15       | 15.43               | 0.83           | 7.55       | 5.39               | 0.15           |
| 0.20       | 12.99               | 0.14           | 8.05       | 6.06               | 0.36           |
| 0.25       | 12.64               | 0.62           | 8.55       | 6.80               | 0.37           |
| 0.35       | 11.07               | 0.57           | 9.05       | 6.72               | 0.30           |
| 0.45       | 9.76                | 0.38           | 10.05      | 7.36               | 0.35           |
| 0.55       | 8.33                | 0.38           | 11.05      | 7.65               | 0.11           |
| 0.65       | 8.07                | 0.38           | 12.05      | 7.80               | 0.14           |
| 0.75       | 7.53                | 0.52           | 13.05      | 7.57               | 0.21           |
| 0.85       | 7.40                | 0.62           | 13.45      | 7.98               | 0.21           |
| 0.95       | 6.88                | 0.37           | 13.65      | 7.75               | 0.23           |
| 1.05       | 6.69                | 0.37           | 13.85      | 8.05               | 0.19           |
| 1.55       | 6.22                | 0.45           | 14.05      | 7.81               | 0.38           |
| 2.05       | 5.45                | 0.25           | 15.05      | 8.11               | 0.20           |
| 3.05       | 5.10                | 0.24           | 16.05      | 7.98               | 0.11           |
| 4.05       | 4.84                | 0.31           | 18.05      | 7.88               | 0.20           |
| 5.05       | 4.94                | 0.17           | 20.05      | 7.76               | 0.16           |
| 6.05       | 4.73                | 0.23           | 22.05      | 8.05               | 0.50           |
| 6.25       | 4.81                | 0.21           | 25.05      | 7.68               | 0.28           |
| 6.65       | 4.68                | 0.16           | 30.05      | 7.52               | 0.19           |
| 6.85       | 4.73                | 0.17           | 35.05      | 7.35               | 0.13           |
| 7.05       | 5.36                | 0.15           | 40.05      | 7.45               | 0.05           |

concerns new TCS measurements (see table 3), the discussion that follows will be largely confined to them. At this point we simply note that the level of agreement between the existing theories for the TCS, and also at the integral elastic cross section level, is only quite marginal (see figure 3). Until quite recently [11], the available e$^+$–N$_2$ cross section data were almost exclusively restricted to the TCS level [44, 48–50], although we note for completeness the pioneering relative differential cross section data of Przybyla et al [51]. Subsequent to this early work, more recent ICS data for positronium formation [11, 52], electronic state excitation [11, 43, 44] and the direct ionization process [11] have become available. Concentrating again on the TCS, the agreement between the various experimental data [44, 48–50] is only fair so that, in general, agreement between experiment and theory for e$^+$–N$_2$ scattering, even at the TCS level, is not particularly satisfactory (see figures 3(a) and (b)). As a consequence, in order to try and clarify matters, we have therefore made new TCS measurements at energies in the range 0.1–40 eV.

If we now consider figure 3(a) in more detail, we see that agreement between the present TCS and the results of a representative selection of the available calculations [35–38, 40–42] is rather poor. As none of the theories incorporate the formation of positronium into their formulations, a fair comparison can be rendered only at energies less than $\sim$8.8 eV (see
Table 5. The present TCSs ($\times 10^{-16}$ cm$^2$) for positron scattering from C$_2$H$_2$. The errors given represent the statistical uncertainty component only of the overall error. See the text for further details.

| Energy (eV) | TCS ($10^{-16}$ cm$^2$) | Energy (eV) | TCS ($10^{-16}$ cm$^2$) |
|------------|-------------------------|------------|-------------------------|
|            | TCS error ($10^{-16}$ cm$^2$) |            | TCS error ($10^{-16}$ cm$^2$) |
| 0.15       | 81.92                   | 0.15       | 71.2                     |
| 0.25       | 67.43                   | 0.25       | 56.2                     |
| 0.35       | 58.83                   | 0.35       | 48.2                     |
| 0.45       | 51.53                   | 0.45       | 41.2                     |
| 0.55       | 46.81                   | 0.55       | 36.5                     |
| 0.65       | 45.67                   | 0.65       | 35.3                     |
| 0.75       | 41.28                   | 0.75       | 30.9                     |
| 0.85       | 39.45                   | 0.85       | 29.6                     |
| 0.95       | 36.88                   | 0.95       | 26.5                     |
| 1.05       | 35.81                   | 1.05       | 23.5                     |
| 1.25       | 34.54                   | 1.25       | 20.5                     |
| 1.45       | 29.64                   | 1.45       | 15.0                     |
| 1.95       | 24.58                   | 1.95       | 10.5                     |
| 2.05       | 24.21                   | 2.05       | 9.5                      |
| 3.05       | 19.76                   | 3.05       | 15.0                     |
| 4.05       | 18.59                   | 4.05       | 11.0                     |
| 4.55       | 20.75                   | 4.55       | 8.0                      |
| 4.65       | 22.36                   | 4.65       | 5.5                      |
| 4.85       | 22.20                   | 4.85       | 4.5                      |
| 5.05       | 18.58                   | 5.05       | 3.5                      |
| 5.55       | 21.64                   | 5.55       | 2.5                      |
| 6.05       | 19.26                   | 6.05       | 1.5                      |
| 7.05       | 18.50                   | 7.05       | 1.3                      |
| 8.05       | 17.40                   | 8.05       | 1.1                      |
| 9.05       | 16.45                   | 9.05       | 1.0                      |
| 9.35       | 16.00                   | 9.35       | 0.9                      |
| 9.65       | 17.20                   | 9.65       | 0.9                      |
| 10.05      | 16.72                   | 10.05      | 0.8                      |
| 10.25      | 17.14                   | 10.25      | 0.8                      |
| 10.45      | 16.65                   | 10.45      | 0.8                      |
Figure 3. (a) The present (□) TCSs for positron scattering from N\textsubscript{2} are compared with previous theoretical results of Gillespie and Thompson [36] (⋯⋯), Darewych [35] (⋯), Elza et al [37] (— - - - - -), Danby and Tennyson [38] (—— - — — — —) and the integral elastic theory cross sections from de Carvalho et al [41] ( — — — —), del Valle et al [42] ( — — — ) and Gianturco and Mukherjee [40]: body-fixed vibrational close-coupling (BFVCC) ( - - - ) and adiabatic angular momentum coupling (AAMC) ( — · — · — ). Indicative total errors (in violet) for a few of the present TCSs are also shown. (b) The present (□) TCSs for positron scattering from N\textsubscript{2} are compared with previous experimental results from Hoffman et al [48] (⊙), Sueoka and Hamada [50] (△) and Sullivan et al [44] (◇). Indicative total errors (in violet) for a few of the present TCSs are also shown.
The present (□) TCSs for positron scattering from CO are compared with previous experimental results of Kwan et al [53] (○), Sueoka and Mori [54] (△), Sueoka and Hamada [50] (▽) and Sullivan et al [44] (◇) and the elastic ICS theories of Gianturco et al [57]: BFVCC (———) and BFVCC-AAM (———), Arretche et al [59] method of continued fractions (MCF) (- - -) and iterative Schwinger variational method (ISVM) (—— - ——); Jain [55, 56] (—— - - ——) and Tennyson and Morgan [58] (– – –). Indicative total errors (in violet) for a few of the present TCSs are also shown.

Figure 4. The present (□) TCSs for positron scattering from CO are compared with previous experimental results of Kwan et al [53] (○), Sueoka and Mori [54] (△), Sueoka and Hamada [50] (▽) and Sullivan et al [44] (◇) and the elastic ICS theories of Gianturco et al [57]: BFVCC (———) and BFVCC-AAM (———), Arretche et al [59] method of continued fractions (MCF) (- - -) and iterative Schwinger variational method (ISVM) (—— - ——); Jain [55, 56] (—— - - ——) and Tennyson and Morgan [58] (– – –). Indicative total errors (in violet) for a few of the present TCSs are also shown.

TCS might well be a reflection of the quality of the respective N\textsubscript{2} target descriptions employed in these earlier calculations [35–38, 40–42]. The situation with respect to a comparison between the present TCSs and those from previous experiments [47, 48, 50] is, however, much more promising (see figure 3(b)). Specifically, for energies above about 8 eV, there is, within the total errors on the various data sets, very good agreement between the present TCSs, in terms of both shape and magnitude, and those of the previous works by Hoffman et al [48], Sueoka and Hamada [50] and Sullivan et al [44]. At lower energies the agreement with [48] and [50] becomes progressively worse, the present TCSs being up to a third stronger in magnitude, which is well outside the combined overall errors on the respective data sets. We believe that the source of the discrepancy is due to the present apparatus having a superior angular resolution (which is energy dependent) compared to those of Hoffman et al [48] and Sueoka and Hamada [50]. As a consequence, the forward angle scattering error on the measured TCSs will be more severe in those earlier studies compared to the present.

Finally, if we take a close look at the present data and those of Hoffman et al [48] in the energy region above 10 eV, there is a suggestion of a slight error in the energy calibration of Hoffman et al. This assertion is based on the observation that if their TCS data are shifted by ~1 eV, the agreement with the present data improves quite a lot.
Figure 5. The present (□) TCSs for positron scattering from C$_2$H$_2$ are compared with previous experimental results of Sueoka and Mori [60] (◦) and theoretical elastic ICSs from de Carvalho et al [41] (— — —), Occhigrossi and Gianturco [61] (— — — — — —) and Franz et al [63] DFT ( - - ) and scaled $R$-matrix (———). Indicative total errors (in violet) for a few of the present TCSs are also shown.

3.2. CO

There have been several previous experimental [44, 50, 53, 54] positron–CO TCS measurements, and several earlier elastic scattering ICS computations [55–59]. From an experimental perspective, we note the linear transmission results of Kwan et al [53], Sueoka and Mori [54] and Sueoka and Hamada [50] and the Penning–Malmberg trap-based results of Sullivan et al [44]. Model-potential computational results have been reported by Jain [55, 56] and Gianturco et al [57], while an $R$-matrix result was due to Tennyson and Morgan [58]. The most recent calculation has employed the iterative Schwinger variational method and originates from Arretche et al [59]. All the earlier data and a selection of the theory results are plotted in figure 4, along with the present TCSs. A list of the present TCSs can also be found in table 4.

Considering first the experimental data, we find a similar picture to that which we have just described for N$_2$. Namely, to within the overall rather than statistical errors, above an energy of about 4 eV all the measurements are in good quantitative accord with one another. While the present TCSs do appear to be systematically a little lower in magnitude than those of Kwan et al [53] and Sueoka and Mori [54], we do not believe this to be physically significant when the overall errors in each case are taken into account. At energies below 4 eV, the present TCS data tend to be higher in magnitude than those of [53, 54] and Sueoka and Hamada [50] although they all exhibit the same qualitative energy dependence (shape). This once again echoes what we found in N$_2$, which we think is due to the superior angular resolution of our measurements so that the forward angular scattering effect errors are not as severe in our case.
Figure 6. The present TCSs for positron scattering from the isoelectronic molecules \( \text{N}_2 \) (\( \square \)), \( \text{CO} \) (\( \circ \)) and \( \text{C}_2\text{H}_2 \) (\( \triangle \)). The plotted errors only include the statistical uncertainty component of the overall error. Indicative total errors (in violet) for a few of the present TCSs are also shown.

when compared to others \([50, 53, 54]\). Somewhat paradoxically, all the positron–CO elastic ICS calculations \([55–59]\) appear to be giving a much more physical description of this scattering process compared to that correspondingly seen in \( \text{N}_2 \). Indeed, we do not understand why this should be the case, but the evidence in figures 3(a) and 4 clearly supports our assertion. Below the positronium formation threshold of \( \text{CO} \) (see table 1), all the theories exhibit an energy dependence that is in fair qualitative accord with the available data. Quantitatively, the present TCSs are probably best described by the \( R \)-matrix results of Tennyson and Morgan \([58]\) (see figure 4). While it is certainly unphysical for an elastic ICS to be greater in absolute value than the TCS at a given energy, if we recall that the present measured TCS will increase should a forward angle scattering correction be applied, then our TCSs’ level of agreement with the \( R \)-matrix result is actually likely to be rather good. Nonetheless, it is clear that further theoretical development is still required, even for energies below the positronium threshold, for this scattering system.

3.3. \( \text{C}_2\text{H}_2 \)

In table 5, we list the present TCSs for positron scattering from acetylene. These data are also plotted in figure 5, along with the only previous experimental determination from Sueoka and Mori \([60]\) and theoretical elastic ICSs from Occhigrossi and Gianturco \([61]\), de Carvalho \textit{et al} \([62]\) and Franz \textit{et al} \([63]\). We note again that none of these theories incorporate a model for positronium formation and hence are restricted here to energies lower than about 5 eV (see table 1). In this case, it is clear from figure 5 that the present data and those from Sueoka and Mori \([60]\) are in quite good agreement, over the common range of measured energies.
only possible exception to this general statement is near the positronium formation threshold energy, where the present TCS suggests a rather stronger opening at threshold than that found by Sueoka and Mori. This difference is possibly due to the poorer energy resolution of the Japanese apparatus. This observation also leads us to suggest that measurements by the ANU group [29] or the San Diego group [11], who can measure the positronium formation cross sections directly and with better energy resolution and signal to noise performance, might be very useful in acetylene. All of the existing theoretical calculations [61–63] do a reasonable job in reproducing the energy dependence of the experimental TCS (see figure 5). Of particular interest is the density functional theory (DFT)-based computation from Franz et al [63], which is in good accord with both their scaled $R$-matrix results and the Schwinger method calculation of de Carvalho et al [62]. Leaving aside the argument as to whether or not DFT functionals are a physical representation for ‘orbitals’, there is ample evidence from electron momentum spectroscopy studies [64] suggesting that DFT-based computations provide a very accurate description of the measured triple differential cross sections at a fraction of the usual CPU-time cost. Thus an extension of this approach, as done by Franz et al to electron and positron elastic and discrete excitation scattering processes, is a notable development.

3.4. Comparative behaviour

In figure 6, we plot our TCS data for $N_2$, CO and $C_2H_2$. This figure leaves us in no doubt that the critical factor in determining the extent of the positron scattering interaction, for a given value of $Z$, is the nature of the molecular bonding in the species rather than the number of electrons it possesses. As evidenced by its relatively larger value for $\alpha$ (see table 1), $C_2H_2$ has a more diffuse electron cloud and so its TCS is uniformly stronger in magnitude than those of CO and $N_2$. Even though the bonding in CO and in $N_2$ is quite similar, it is not until positron energies greater than $\sim 20$ eV that their TCSs become identical. Note that this observation is consistent with that made previously by Kauppila et al [8]. Below 20 eV, however, their TCSs are very different, at least in part due to the positronium formation cross section of CO being much stronger at threshold than that for $N_2$. Somewhat surprisingly, the very low energy ($<0.7$ eV) TCS for $N_2$ is stronger in magnitude than that for CO. As $N_2$ has a smaller dipole polarizability than CO and as $N_2$ has no permanent dipole moment whereas CO does (see table 1), we had not anticipated that result. However, it is possible that this observation is simply an artifact of the CO TCS needing a relatively larger forward angle scattering effect correction compared to that of $N_2$ [30]. The final comparative observation we make is that below the positronium formation threshold for $C_2H_2$, the energy dependence of the TCS for $C_2H_2$ and for CO are almost identical.

4. Conclusions

We have reported on a comprehensive series of TCS measurements for positron scattering from $N_2$, CO and $C_2H_2$. Where possible, comparison was made, in each case, to the corresponding results from previous experimental studies and theoretical investigations. With the exception of $C_2H_2$, where good agreement with Sueoka and Mori [60] was found across the common energy range, for $N_2$ and CO the agreement with previous data was limited to relatively higher positron impact energies. At the lower energies, the disagreement between the various data sets was largely thought to originate from the differing forward angle scattering corrections that need to be applied in each case. With respect to theory, while it is clear that progress is being made,
further development, including the explicit inclusion of the positronium formation channel, is required. Finally, the present work clearly highlights that the nature of bonding in the target plays a significant role in the scattering process. Indeed, in this respect, the positron is quite possibly a more sensitive probe than the electron.

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