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Positron scattering from water

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Abstract

We report results of total cross section measurements for positron scattering from the chemically, environmentally and medically important molecule water. The present measurements were conducted over the energy range of 0.1–20 eV, with the cross section found to be strongly peaked at the lower energies. We believe these are only the second independent data to have been reported in the literature for this process and we note significant discrepancies with the earlier measurements in terms of the magnitude of the total cross section.

1. Introduction

Water (H_2O) plays a very important role in diverse fields such as astrophysics, atmospheric physics, magneto-hydrodynamics and biophysics. For example, water has been detected in the atmospheres of Venus and Mars and it is also the most abundant molecule in comets [1]. Terrestrially it is an important greenhouse gas, contributing more than half of the 33 K of natural warming [2]. Water is also the major product in the combustion of hydrocarbon fuels, and thus it is one of the essential constituents of the model flue gas. Finally, water plays an essential role in life being the dominant component of the biological cell and an extremely important carrier in both chemical and biochemical liquid phase reactions [1]. Indeed it is in this latter regard that water, and in particular the interactions of positrons with water, has stimulated our interest.

To the best of our knowledge [4, 5], the only previous literature report for experimental e^+/H_2O scattering is from Sueoka and colleagues [6–8]. In their original work [6, 7], total cross sections (TCSs) for positrons with energies in the range of 1–400 eV were measured using a time of flight (TOF) technique. These data were later significantly revised by Kimura *et al* [8], in particular the magnitude of the TCSs at lower energies increased significantly in the more recent work [8] over that reported originally [6, 7]. We note,

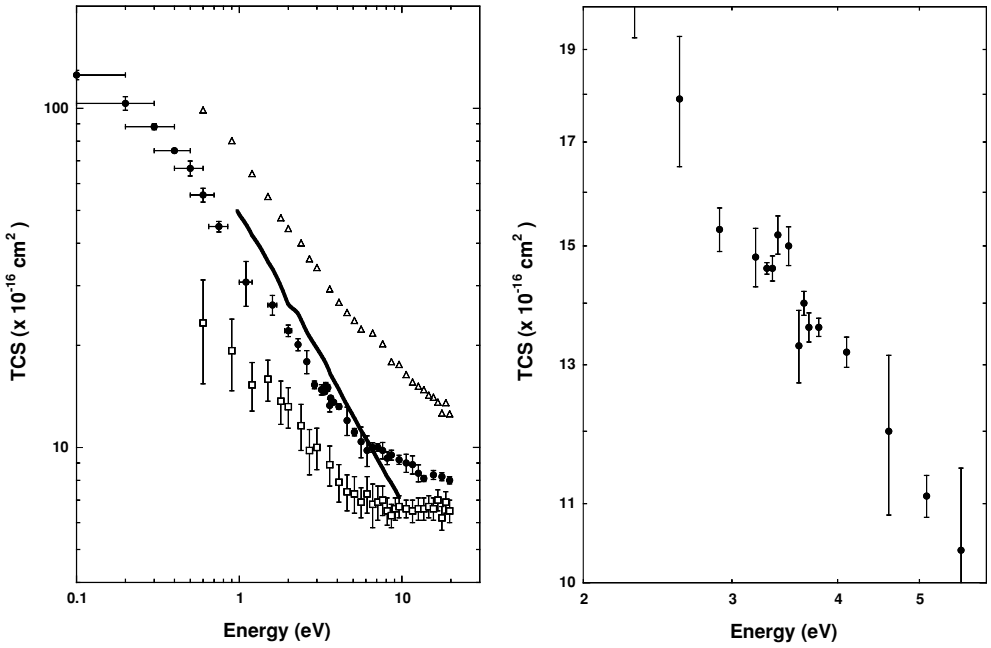

Figure 1. The present (•) total cross section ($\times 10^{-16}$cm^2) data for positron scattering from H_2O. Also shown are the earlier results from Sueoka *et al* [6, 7] (□) and Kimura *et al* [8] (△) and the calculated elastic integral cross section from Gianturco *et al* [9] (-). The errors shown in the present data represent the standard deviation in the measured cross section at a given energy. See the text for a discussion of the absolute error. An inset depicting the present data only, for the energy range of 2–6 eV, is also given.

however, that the shapes of both these TCSs were fairly consistent, being largely structureless and increasing in magnitude as you go to lower positron energies (see figure 1). Given the importance of water as a biomolecule, and given the apparent disagreement between the only two existing e^+/H_2O TCSs [6–8] from the same group, we believe that a further, independent, measurement is necessary for this system. In addition, our apparatus overcomes some of the limitations present in that of the earlier studies, and so allows us to measure positron cross sections with a greater accuracy. Hence, we report e^+/H_2O total cross sections for positrons with energies in the range of 0.1–20 eV. From a theoretical perspective, we only know of the quite recent elastic and fundamental vibrational mode integral cross sections (ICSs) for e^+/H_2O from the Rome group [9–11].

At the University of Trento, low-, intermediate- and high-energy electron and positron total cross sections have been studied extensively [e.g. 3, 12, 13] over many years. As a consequence, in the next section of this paper we only briefly describe the experimental apparatus and techniques used to make our measurements. Following that, our results and a discussion of these results are presented. Finally, some conclusions from the present work are drawn.

2. Apparatus and techniques

The positron spectrometer used in the current measurements has been developed in the Trento laboratory and has already been described in a previous paper [14]. General information

about the present attenuation technique can be found for instance in [15]. Although that paper specifically looked at electron cross sections, the two conjugated particles share most of the properties relevant to the present study. Here we therefore outline only those characteristics which are relevant to the present measurement.

Slow positrons are produced by a 1 μm tungsten-film moderator in front of a ^{22}Na radioactive source [16]. These positrons are transported and focused into the scattering chamber using a series of charged particle optics with appropriate applied potentials. Note that a weak magnetic field (8–10 Gauss) is also present in the scattering region. The energy resolution of the positron beam has been evaluated to be slightly less than 0.3 eV full width at half maximum (FWHM), possibly as a result of the partial monochromatization in the deflector and in the optics [3]. In this paper we report cross section values down to energies of 0.1 eV, but values below 0.5 eV are to be regarded as indicative. Indeed, due to the quoted energy spread, the measurements at energies lower than 0.5 eV (see table 1) should be taken as the convolution of the real (unknown) cross section with the positron energy distribution. Nevertheless, as our measurements are the first in this energy region we believe they warrant publication. Using a ^{22}Na source with an activity of 8mCi, positron beam intensities at the detector were found to vary from 10 to 130 s^{-1}, the highest value being achieved at the high-energy limit. The zero for the energy scale of the present positron measurements has been determined, in the absence of the target gas, with a retarding potential analysis of the beam. Such a measurement suggests a probable error of ± 0.1 eV in our energy scale. This determination of the energy scale calibration is particularly crucial at low energies, where the total cross section rises rapidly with decreasing energy. We note that in these instances even a small inaccuracy in the energy calibration can produce a significant uncertainty as to the true value of the TCS. Such an effect would, for example, be very misleading for theoreticians trying to describe the scattering process.

High-purity water was used throughout this study. In addition, the liquid H_2O is degassed with a freeze, pump and thaw procedure. The gaseous target was fed to the scattering cell with a two-way diverter valve, where the same amount of gas was diverted to the scattering cell or alternatively was injected directly into the vacuum system. In the first case attenuation of the positron beam was obtained. With such a provision we obtain that the background pressure outside the gas cell and therefore the attenuation of the beam in the path outside the gas cell are constant during the measurement cycle. The background pressure during the measurements was typically 10^{-3} of the pressure inside the gas chamber.

Total cross sections were computed according to the Beer–Lambert law

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

where I_1 is the positron beam count rate at P_1, the pressure measured with the gas routed to the scattering cell, k is Boltzmann's constant, T is the temperature of the gas (K), σ is the total cross section of interest, I_0 is the positron beam count rate at P_0, the pressure with the gas diverted to the vacuum chamber and L is the length of the scattering region (see later). In order to minimize double scattering events and ensure the TCS is pressure independent, the ratio $\frac{I_1}{I_0}$ has been kept to values larger than 0.7. Furthermore, the standard checks on the linearity of the plots of $\log\left(\frac{I_1}{I_0}\right)$ versus gas pressure [17] were performed at selected energies. The geometrical length of the scattering region is 100 ± 0.1 mm, with apertures of 1.5 mm diameter at both the entrance and exit of the scattering chamber. End effects [3] were considered in the present study. It has been demonstrated [12, 18] that the effects due to the entrance and exit apertures cancel if the two aperture diameters are equal, so that their contribution to the uncertainty in the value of L is possibly less than 0.15%. In the current

Table 1. The present total cross section ($\times 10^{-16}$ cm^2) data for positron scattering from water. The errors represent the standard deviation on the measured cross section at a given energy. See the text for a discussion of the absolute error.

Energy (eV)	Total cross section ($\times 10^{-16}$ cm^2)
0.1	125.4 ± 4.0
0.2	103.4 ± 4.7
0.3	88.2 ± 1.7
0.4	75.0 ± 1.0
0.5	66.5 ± 3.4
0.6	55.5 ± 2.6
0.75	44.8 ± 1.6
1.1	30.7 ± 4.6
1.6	26.3 ± 1.8
2.0	22.1 ± 0.9
2.3	20.1 ± 0.8
2.6	17.9 ± 1.4
2.9	15.3 ± 0.4
3.2	14.8 ± 0.5
3.3	14.6 ± 0.1
3.35	14.6 ± 0.2
3.4	15.2 ± 0.4
3.5	15.0 ± 0.4
3.6	13.3 ± 0.6
3.65	14.0 ± 0.2
3.7	13.6 ± 0.2
3.8	13.6 ± 0.2
4.1	13.2 ± 0.3
4.6	12.0 ± 1.2
5.1	11.1 ± 0.3
5.6	10.4 ± 1.1
6.1	9.8 ± 1.0
6.6	10.0 ± 0.3
7.1	10.0 ± 0.3
7.6	9.8 ± 0.6
8.1	9.3 ± 0.4
8.6	9.5 ± 0.3
9.6	9.2 ± 0.3
10.6	9.0 ± 0.6
11.6	8.9 ± 0.6
12.6	8.4 ± 0.5
13.6	8.1 ± 0.2
15.6	8.3 ± 0.3
17.6	8.2 ± 0.2
19.6	8.0 ± 0.2

application, the value of L used in equation (1) has been corrected to account for the path increase caused by the gyration of the positrons in the focusing magnetic field present in the scattering region (typically this correction is $\sim$6%). This arises because in the no B-field configuration the positron trajectories are straight segments; however, with a field applied they are bound to move on a spiral which thus may increase the true value for L, which represents the length of the positron path in the gas-filled region. Note again that the magnetic induction was of the order of 8–10 Gauss, depending on the positron energy under consideration.

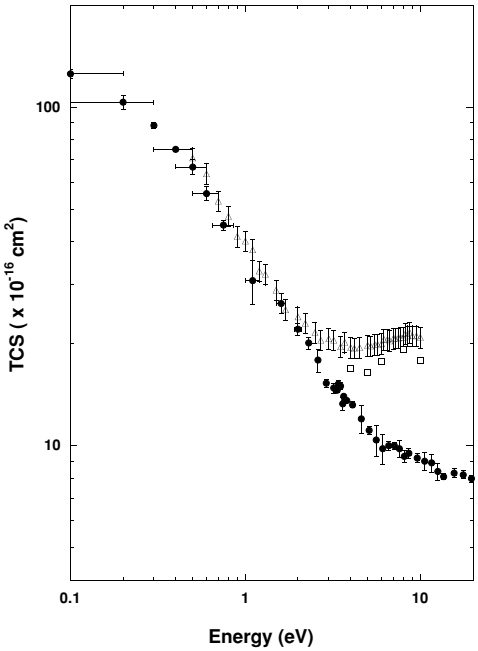

Figure 2. Total cross sections for positron and electron scattering from H_2O. The present positron data (•) and the earlier electron data of Szmytkowski [23] (Δ) and Saglam and Aktekin [24] (□) are depicted.

The gyration of the projectile particles can also potentially increase the angular resolution error with respect to the no-field case [19]. However, even though absolute differential cross sections for positrons on H_2O are not currently known [4], we believe that the present geometry guarantees a small error ($\leqslant$10%). The scattering cell pressure has been measured with an MKS Baratron capacitance manometer (Model 628B: 1 Torr full scale) operated at 100 °C. Since the scattering chamber was at room temperature (24 ± 2 °C), a thermal transpiration correction has been applied to the pressure readings. This correction has been calculated according to the model of Takaishi and Sensui [20], and is of the order of 10% over the entire energy range.

Measurement time was of the order of 1 h per each energy point, with each point being the average of 100 single determinations. The positron beam obtained with the present apparatus [14] was extremely stable over times of the order of 1 month and indeed no influence of the target gas on the beam characteristics was noted. A new conditioning of the moderator film was also not required during the present study. The absolute errors on our measurements (not given in table 1 or figures 1 and 2) have been evaluated as the root of the quadratic sum of the contributing errors. A detailed discussion of the origin and of the evaluation techniques of such contributions can be found in [12] and in the references contained in that paper. At this point, however, we specifically note that the respective contributions due to the uncertainties in our thermal transpiration and B-field spiralling corrections are small; they do not contribute significantly to the overall errors on our TCSs. These overall uncertainties typically amounted to ±3% at the higher energies and to ±7% at the lower energies, the dominant contribution being due to the uncertainty in the pressure determination. Note that the error quoted in table 1 is the statistical error only.

3. Results and discussion

In figure 1, we plot the present TCSs for positron/H_2O scattering along with earlier data due to Sueoka *et al* [6, 7] and Kimura *et al* [8] and the integral cross sections of Gianturco *et al* [9]. The data reported in Kimura *et al* are the original work of Sueoka *et al* [6], but with a correction for forward angle scattering effects applied to it. As such it represents their preferred TCS. Consequently, we restrict our discussion to a comparison between the present TCSs and those of Kimura *et al* [8], as well as the elastic theory result [9]. We note here that in the absence of any theoretical or experimental positron–water differential cross sections, we did not apply any correction for forward scattering to our data.

It is clear from figure 1 that apart from an important exception near 3.5 eV in the present data (see later), both the experimental TCSs are largely structureless over the energy range considered. In addition, both the present TCSs and those of Kimura *et al* rise significantly in magnitude as the positron energy is decreased. This indicates the importance of polarization effects (water has a dipole polarizability of ∼10 au) and/or waters permanent dipole moment (1.85D) at these lower energies. Thus at a qualitative level there are similarities between the present result and that from Kimura *et al* [8]. Quantitatively, however, the present TCSs are smaller in magnitude than those of Kimura *et al* at all energies studied. We note that this discrepancy is bigger than the combined (declared) error bars. The origin of this disagreement, at least in part, would seem to stem from the very large correction (up to 300% of the measured values) that Kimura *et al* [8] applied to the data of Sueoka *et al* [6] to account for forward angle scattering effects. We note that the present experimental geometry ensures that such a radical correction is not needed for this effect in our measurements. With regard to the elastic ICS calculation of Gianturco *et al* [9], we observe that at energies below 8 eV it overestimates the magnitude of the present TCSs.

Some insight into the energy dependence of the present e^+/TCS might be gleaned by considering the corresponding behaviour in atomic-H and the alkali atoms. In the case of H the dipole polarizability is ∼4.5 au, while for the alkali atoms they range from 141 to 338 au. According to Kwan *et al* [21], the e^+/TCS for Na and K increases strongly as the positron beam energy decreases, while Zhou *et al* [22] found that the e^+/TCS for H decreased as the positron energy decreased. Water, with a dipole polarizability of ∼10 au, is much closer in value to that of H than the alkali atoms, and yet we see its e^+/TCS increases strongly (figure 1) as the positron beam energy decreases. Thus, if the analogy holds, perhaps the behaviour we see in e^+/TCS for H_2O also reflects the fact that it has a permanent dipole moment. Further evidence in support of this conjecture comes from similar behaviour being seen in the respective TCSs for e^+-NH_3 and e^+-HCL scattering (see [8]), both these molecules also having strong permanent dipole moments.

The most unexpected result we show in figure 1 (see inset) is the suggestion of some structure in the TCS around 3.5 eV. If such a structure was simply an experimental artefact then we would have also expected to find it at this energy in tetrahydrofuran [3], but this was most certainly not the case. We note that in addition to tetrahydrofuran, we also have unpublished TCS data for e^+/Ar, N_2, CO_2 and C_6H_6 where no such feature, in the relevant energy range about 3.5 eV, is observed. These other data were taken with the same apparatus and with similar techniques to those used in the present study. In addition, this structure in the H_2O TCS persistently appeared (i.e. was statistically real) in our independent runs at the energies about 3.5 eV. We therefore believe it to be physical and peculiar to H_2O. The amplitude of the structure amounts to about 10% of the TCS at nearby energies. As it appears in figure 1, the structure is clearly above the scatter. Nevertheless it is difficult to give a measure of its FWHM: we can only state it is approximately of the order 0.35 ± 0.2 eV.

This value is compatible with our estimate of the energy resolution and therefore the width shown in figure 1 is possibly representative of a smaller natural width.

Finally, we believe it to be useful to compare the present positron/H_2O TCSs to corresponding electron/H_2O TCSs from other groups [23, 24]. This is illustrated in figure 2. Here we see that for energies below 2 eV, the shapes of both the positron and electron TCSs are very similar. In addition, the absolute magnitudes of the electron and positron TCSs are also similar (to ∼10%) for energies less than 2 eV. This is really quite an amazing result as there are very different processes occurring (for example, the absence of exchange in the positron case, while the static interaction is repulsive in the case of positrons and attractive in the case of electrons) when each respective projectile (e^+ or e^-) interacts with water, and yet the net result suggests that the effect at these lower energies is effectively the same. Note this is not what we found previously in tetrahydrofuran [3] and in several other molecules (e.g. benzene, cyclohexane, hexane, aniline, 3-hydroxy tetrahydrofuran and 3,4-dihydro-2H-pyran [25]), where the low energy e^+/TCSs were significantly larger than those for the electron channel. A detailed understanding of this latter finding, for these larger organic molecules, might well require assistance from theoretical colleagues.

4. Conclusions

We have reported a new total cross section measurement for positron–water scattering. At all common energies our data were appreciably smaller in magnitude than the earlier result from Kimura *et al* [8], and it strongly rises in magnitude at energies below 1 eV. The present data also showed that the elastic e^+/H_2O integral cross section calculation from Gianturco *et al* [9] overestimates the TCSs at energies below 8 eV. We note that a small narrow structure at around 3.5 eV was observed in the current study, although a definitive physical explanation for its origin remains elusive at this time. Finally, it is also worth recalling the very close magnitude of the positron and electron total cross sections in the energy range from 0.5 to 2 eV.

Acknowledgments

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Note added in proof. We have recently been sent a preprint of an article by Beale *et al* [26] that reports positronium and positron-water total cross sections. The positronium incident energies ranged between 10 and 100 eV, while the positron data were measured at six energies between 7 and 417 eV. Both sets of TCSs are not corrected for forward angle scattering effects. For positron-water TCSs these authors [26] found much better agreement with the results of Sueoka *et al* [6] than with Kimura *et al* [8]. On comparison with the present data, for the small energy range overlap, the TCS of Beale *et al* would appear to be a little lower in magnitude. This result is consistent with the somewhat superior angular discrimination of the Trento apparatus.

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