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Positronium- and positron–H_2O total cross sections

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Abstract

The total cross sections for positron and positronium scattering from H_2O molecules have been measured for incident energies between 7 and 417 eV, and 10 and 100 eV, respectively. The experimental system has been characterized with respect to its angular acceptance of both scattered positrons and positronium in order to correct the data for forward-scattering errors once differential cross sections become available. The present data are compared with previous results for electron and positron total cross sections.

1. Introduction

It is difficult to overestimate the importance of water in the universe. It makes up the bulk of living organisms and it provides the medium for a host of chemical reactions. Its vapour has been observed in the atmosphere of several planets within the Solar System and of the Sun itself (Wallace *et al* 1995). Within the Earth's atmosphere, water is a key greenhouse gas (Taylor 2002).

Electron collisions with water molecules have been studied for many years (see e.g. recent review by Itikawa and Mason (2005)), with total cross sections (σ_T) being reported since 1929 (Brüche 1929, Sokolov and Sokolov 1986, Sueoka *et al* 1986, Szmytkowski 1987, Zecca *et al* 1987, Jain 1988, Nishimura and Yano 1988, Sağlam and Aktekin 1990, 1991). Convergence among recent measurements of σ_T is found at high energies whilst uncertainties remain at low energies. These uncertainties stem from the difficulty in discriminating against electrons scattered at small forward angles, due to the long-range force which arises from the permanent electric dipole moment of the water molecule (e.g. Okamoto *et al* (1993), Gorfinkiel *et al* (2002)). In the case of positron projectiles, only one experimental determination of σ_T has been reported thus far in the literature (Sueoka *et al* 1986). This was later corrected for forward-scattering errors (Kimura *et al* 2000) by employing *electron* differential cross sections (DCS). No σ_T data are available for positronium projectiles.

Positronium (Ps) is the bound state of an electron (e^-) and its antimatter counterpart the positron (e^+). In contrast to conventional atoms, Ps has a zero-static interaction due to the coincidence of its centres of charge and mass. Depending on the relative spin orientation of the electron and the positron, Ps can be formed in a triplet (*ortho*-) or in a singlet (*para*-) state,

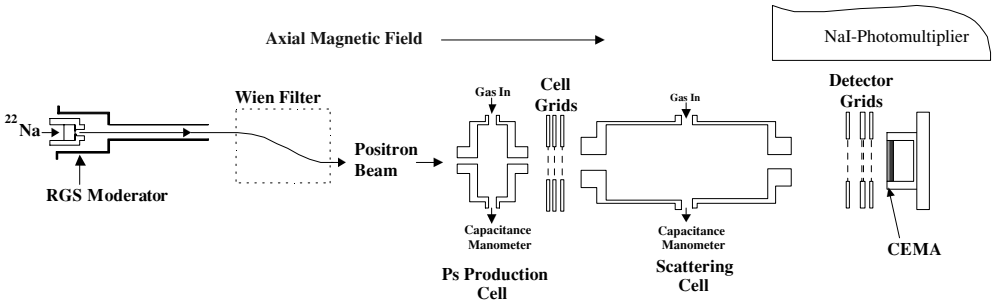

Figure 1. Schematic diagram of the beamline used for the measurement of the total cross sections for positrons and Ps atoms colliding with H_2O (not to scale).

having ground-state lifetimes of 142 ns and 125 ps, respectively. The Ps beam at UCL consists of *o*-Ps only, due to temporal suppression of the *para*-state with flight distance. The beam is produced by neutralizing a positron beam in a gaseous target (Garner *et al* 1996, Leslie *et al* 2002 and references therein) and has been used to measure total cross sections from simple atoms and molecules (Zafar *et al* 1996, Garner *et al* 1996, 1998, 2000, Leslie *et al* 2005), as well as the fragmentation of Ps in collision with He and Xe atoms (Armitage *et al* 2002, 2006).

The primary motivation for the present work was to measure positronium–H_2O total cross sections and, towards this aim, positron–H_2O total cross sections have also been measured in order to check the general performance of the system with respect to water vapour. In the next section, the experimental arrangement and method employed for the measurement of both the positron and Ps total cross sections are described. In section 3, the results are presented and discussed, whilst in section 4 conclusions are drawn.

2. Experimental details

2.1. Positron- and positronium-beamline

Figure 1 shows a schematic diagram of the beamline used for measuring positron and Ps total cross sections. ^{22}Na is the source of positrons, which are energy moderated by argon frozen on a parabolic cup (Özen *et al* 2000). The beam is guided by a magnetic field of 4–22 mT produced by a set of Helmholtz coils. Fast positrons and γ-rays emanating from the source region impinge upon a lead plug whilst the slow positron beam is deflected onto a new axis by the Wien filter. Ps is formed in the 'production cell' via the charge-exchange reaction

$$e^+ + A \rightarrow Ps + A^+, \tag{1}$$

in which the incident positron captures an electron from the production gas (A) (e.g. Zafar *et al* (1996)). For the range of energies used in this work, H_2 was introduced into the production cell in order to generate the Ps beam (Garner *et al* 1996, 1998). The gas pressure within this cell was set in the range 1.2–1.5 Pa throughout this work.

The kinetic energy of the Ps beam, E_{Ps}, is given to a first approximation (Laricchia *et al* 1992) by

$$E_{Ps} = E_+ - I + B, \tag{2}$$

where E_+ is the positron kinetic energy, I is the ionization energy of the production gas and B is the binding energy of Ps, 6.8 eV in its ground state. The positrons and Ps atoms

were detected by coincidence measurements between a NaI photomultiplier and a CEMA (channel-electron-multiplier array).

The total cross section (σ_T) was determined using the Beer–Lambert law:

$$I = I_0 \exp(-nl\sigma_T), \tag{3}$$

where I is the transmitted beam intensity, I_0 is the incident beam intensity, n is the target number density and l is the effective length of the scattering cell.

An Al scattering cell was constructed for the present measurements with H_2O. The cell body was 184 mm long, the entrance aperture had a diameter of 6 mm and the exit aperture had a diameter of 8 mm. Due to the low pressure inside the cell, a long cell was required in order to achieve reasonable attenuations. The Ps beam pencil angle was 1°.

The effective length for the scattering cell was 0.128 ± 0.003 m, as determined by measuring positron–gas (Xe, O_2 and Ne) total cross sections and normalizing these to known values (Dababneh *et al* 1980, Kauppila *et al* 1981, Dababneh *et al* 1988, respectively). The evacuation time for water vapour was measured by monitoring the time it took for the incident positron beam count rate to return to its normal level after stopping the inflow of water vapour. This evacuation time was found to be ∼10 min and thus a corresponding delay was introduced between successive measurements of the incident and transmitted beams. The pressure in the production and scattering cells was measured with Baratron gauges and adjusted by computer-controlled valves. For both positron and Ps total cross sections, the gas pressure within the scattering cell was 50 ± 5 mPa.

In order to determine positron total cross sections, the incident beam was measured with vacuum in both cells. The transmitted beam was measured with vacuum in the Ps production cell and vapour in the scattering cell. The incident and transmitted beams were measured for 20–60 s depending on the beam energy. The total data-acquisition time was between 0.6 and 1.5 h at each energy investigated.

For the determination of the Ps total cross sections, the incident beam was measured with H_2 in the Ps production cell and vacuum in the scattering cell. The transmitted beam was measured with gas in both cells and the background with vacuum in both cells. Each measurement had a duration of 7200 s each. The total data-acquisition time was between 1 and 8 days at each energy investigated. The stability of the incident positron beam was monitored for 10 s between each Ps measurement. The mean of the positron rates before and after this was used to normalize each Ps run.

The grids in front of the scattering cell were used to define the lower limit of the energy distribution of the positron beam, namely 3 eV below the maximum energy of the beam. This was done before the scattering cell in order to prevent residual positrons, from the lower (more intense) portion of the energy distribution of the beam, from entering the scattering cell and forming Ps. The grids in front of the detectors were used to discriminate against forward scattering in the positron total cross-section measurements (details are given in section 2.2). The grids were also used to stop any positrons/electrons from Ps fragmentation within the scattering cell from reaching the detector (Armitage *et al* 2002, 2006).

A freeze–pump–thaw method (Rudd *et al* 1985, Sağlam and Aktekin 1990) was used to purify the distilled water prior to its introduction into the Al scattering cell. The effectiveness of this procedure was verified by mass spectrometry on a different system (Arcidiacono *et al* 2006), which also indicated the absence of water clusters.

2.2. Angular acceptance of forward-scattered projectiles

For collisions between electrons and a polar molecule such as H_2O, the contribution of small-angle forward scattering is very large due to the long range of the interaction resulting

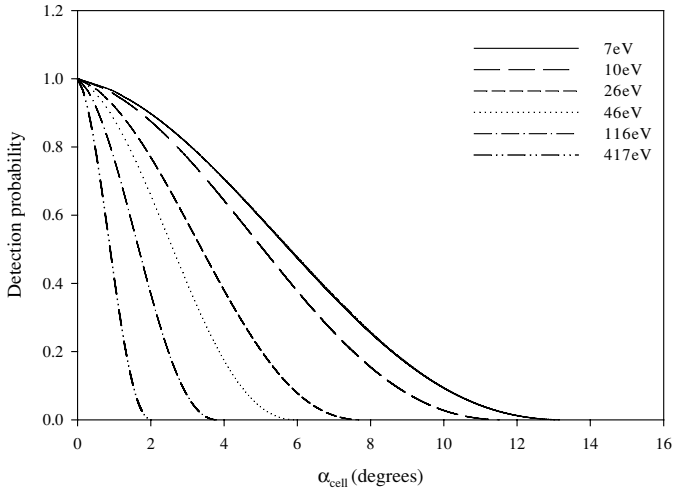

Figure 2. Dependence of the detection probability upon scattering angle at each incident positron energy investigated.

in strongly forward-peaked DCS for elastic and inelastic processes (e.g. Jung *et al* (1982), Itikawa and Mason (2005)). In this work, partial discrimination against forward scattering was implemented through a **B**-field gradient between the region where the scattering occurred and the detector region where grids were employed to analyse the longitudinal energy of the scattered projectile. In this way, ensuring that the magnetic field changes negligibly during one revolution of the helical motion of the scattered particle, the angular momentum is conserved and the pitch angle changes according to

$$\frac{B_{\text{cell}}}{B_{\text{CEMA2}}} \propto \frac{\sin^2 \alpha_{\text{cell}}}{\sin^2 \alpha_{\text{CEMA2}}}, \tag{4}$$

where B_{cell} is the magnetic field at the cells (4 mT), B_{CEMA2} is the magnetic field at the detectors (22 mT), α_{cell} is the pitch angle within the cell and α_{CEMA2} is the pitch angle at the detector.

As mentioned in section 2.1, the lower limit of the energy distribution of the incident positron beam was set by the potential (V_r) on the grids before the scattering cell, resulting in an energy distribution from eV_r to (eV_r + 3) eV. Thus, forward-scattered positrons with a longitudinal energy below eV_r were repelled by the potential V_r also applied to the grids in front of the detector. The angular variation of the detection probability of positrons forward scattered at an angle α_{cell} was computed allowing for the size of the apertures and of the beam. The results are plotted in figure 2 at various positron incident energies. The functions are approximately Gaussians with full-width-half-maxima as noted in table 1.

It is unclear what role forward-scattering effects play in the case of Ps projectiles, but it would be expected to be minor in comparison with e^- and e^+, since Ps is neutral and its detection upon scattering is limited to angles $\leqslant 1^\circ$.

3. Results and discussion

3.1. Positron total cross sections

The positron total cross sections determined in the present work are shown in figure 3 together with previous values from experiment (Sueoka *et al* 1986, Kimura *et al* 2000) and theory

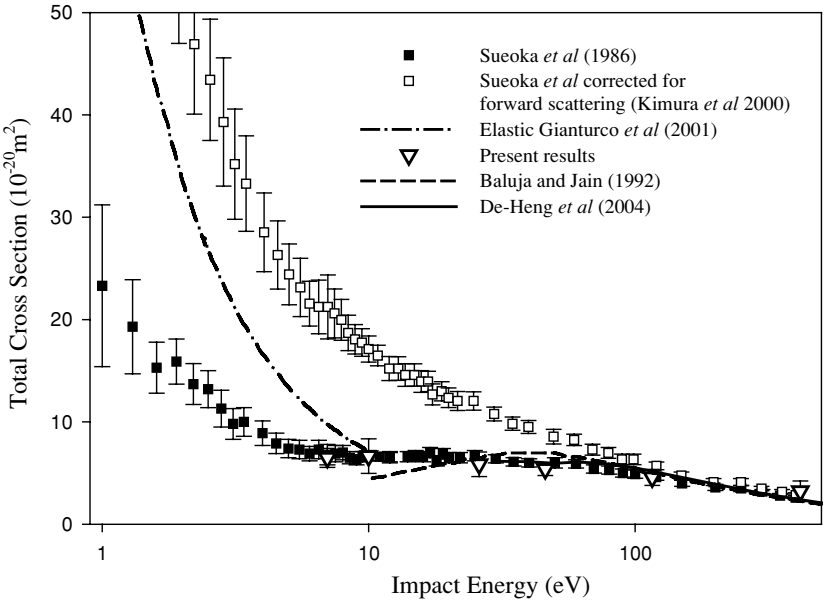

Figure 3. Total cross sections for positrons colliding with H_2O.

Table 1. FWHM of the angular distributions of the detection probability displayed in figure 2.

e^+ incident energy (eV)	FWHM (degrees)
7	5.8
10	5.1
26	3.4
46	2.5
116	1.6
417	0.9

(Gianturco *et al* 2001, Baluja and Jain 1992). As mentioned above, forward-scattering effects are considerable for electron scattering from polar molecules such as H_2O (e.g. Okamoto *et al* (1993), Yuan and Zhang (1992)) and are also expected to be important for positrons (Kimura *et al* 2000). These authors corrected the positron data of Sueoka *et al* (1986) by assuming the differential cross sections for positrons to be the same as for electrons. In the absence of explicit results for positrons, we have chosen not to apply any correction to our data and to present them as raw.

As shown in figure 3, the present data display little energy dependence over the range investigated. The values are in good agreement with those of Sueoka *et al* (1986) prior to the correction for forward scattering (Kimura *et al* 2000) which results in an increase of their cross section by up to a factor of ∼4. Both the present results and those of Sueoka *et al* (1986) are lower than the close-coupling results for the elastic cross section calculated by Gianturco *et al* (2001) using a parameter-free quantum dynamical model for the electron–positron correlations. At high energies, the present data are in reasonable agreement with the calculation of Baluja and Jain (1992) who used a spherical-complex-optical-potential method together with a molecular wavefunction. However below 50 eV, these authors state that their results may not be reliable for polar molecules. The experimental data at intermediate and high

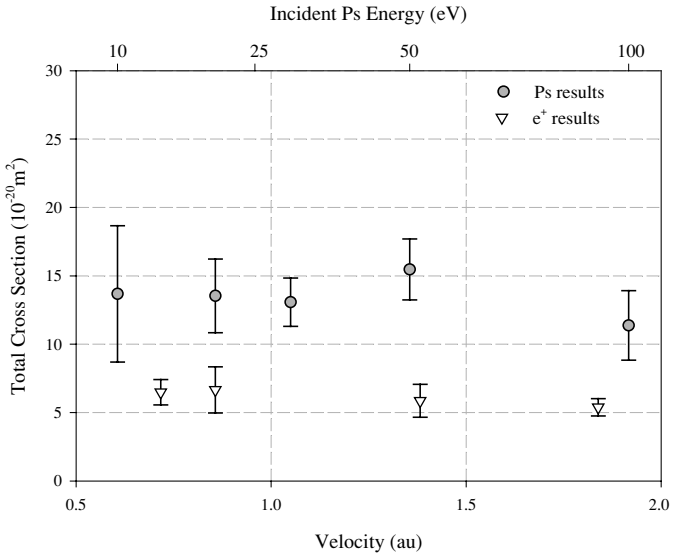

Figure 4. Total cross sections for Ps and positrons colliding with the H_2O molecule.

energies are also in agreement with the calculation of De-Heng *et al* (2004) who employed a complex-optical-potential approach and applied the additivity rule for the cross sections of the constituent atoms. In order to evaluate the contribution from forward scattering to the present measurements of σ_T, DCS for positron impact are needed to weigh the variation of the detection probability upon scattering angle presented in table 1 and figure 2.

3.2. Ps total cross sections

The results for the total cross section for Ps scattering from H_2O are shown in figure 4 together with the data obtained for positrons. As can be seen from the figure, the total cross section values determined for Ps are approximately flat within experimental errors across the velocity range investigated. This is somewhat different from previous targets for which the total cross section generally exhibits a broad peak at low energies (Garner *et al* 1996, 1998, Leslie *et al* 2005). The Ps total cross section data are a factor of 2 higher than the corresponding positron values.

The Ps and positron total cross sections are compared with experimental and theoretical data for positrons and electrons in figure 5. At higher velocities, the Ps data are comparable with the experimental electron data of Szmytkowski (1987) and Kimura *et al* (2000) and the theoretical determination of Jain (1988). Below ∼1.2 au, the Ps values fall below these data as well as the results of the *R*-matrix calculation of the electron elastic scattering cross section of Gorfinkiel *et al* (2002).

Recently, Leslie *et al* (2005) have noted a trend in the magnitude of the Ps total cross sections for simple atoms and molecules when compared with corresponding electron and positron values. Namely, between velocities of 1 and 2 au, the Ps values are closer to those for electron scattering than positron scattering, whilst at lower velocities the Ps values are found to be in general intermediate to the corresponding electron and positron cross sections. The results for H_2O would conform to this trend if considered together with the positron data of Sueoka *et al* (1986) or our own. However, as discussed above, uncertainties on these data

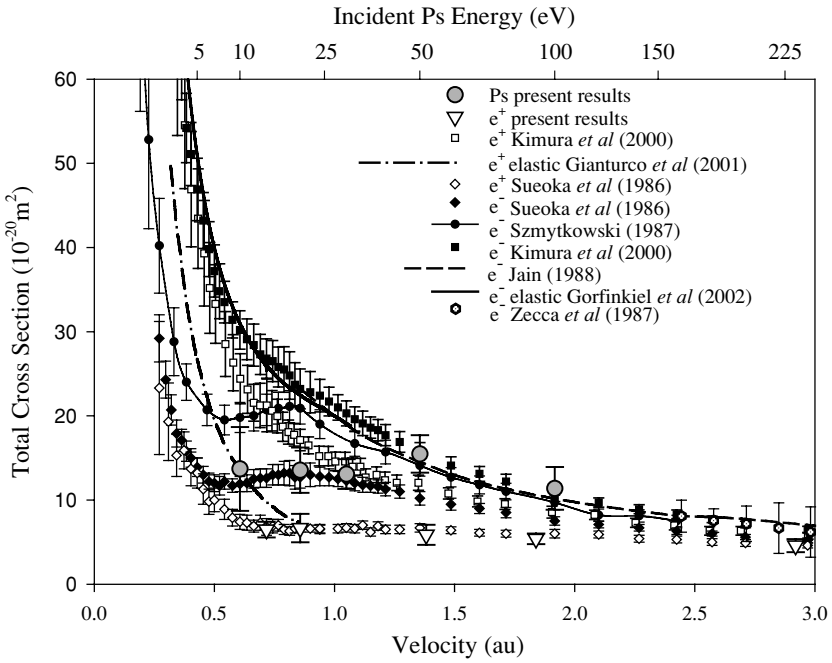

Figure 5. Total cross sections for Ps, positrons and electrons colliding with the H_2O molecule.

from forward-scattering errors remain outstanding and need to be evaluated before a firm conclusion may be drawn from the comparison.

4. Conclusion and outlook

Total cross sections have been measured for both Ps and positron scattering from H_2O. The positron total cross section values are approximately constant with velocity and agree with the original results of Sueoka *et al* (1986), i.e., prior to the correction for forward-scattering errors applied by Kimura *et al* (2000). The Ps total cross-section values are also approximately constant within errors across the velocity range investigated. It has been noted that, at high velocities, the Ps data are comparable with the electron data, e.g., of Szmytkowski (1987) and Kimura *et al* (2000). At lower velocities, the Ps data fall below the positron and electron data of Kimura *et al* (2000).

Experimental and theoretical determinations of differential cross sections for both positron and Ps projectiles from H_2O are needed in order to resolve current uncertainties on the influence of forward-scattering effects upon total cross-section measurements for this target. High-angular discrimination measurements of the total cross section, as currently being performed in another laboratory for positron projectiles on this target (Zecca 2005), are also expected to be valuable in this task.

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