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2013 J. Phys.: Conf. Ser. 443 012001

(http://iopscience.iop.org/1742-6596/443/1/012001)

Journal of Physics: Conference Series **443** (2013) 012001 doi:10.1088/1742-6596/443/1/012001

Positronium formation in molecular oxygen

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Abstract. New measurements of the positronium (Ps) formation cross section in molecular oxygen have been made using the 'positron loss' technique used in earlier measurements in the noble gases – the reduction in beam intensity being attributed to Ps formation. The results are generally in agreement, in terms of dependence on incident positron energy, with earlier measurements, but are lower in magnitude than those deduced from total ionization measurements below the first threshold for direct ionization, which employed ion detection. Q_{Ps} for molecular oxygen exhibits an interesting peak-like dependence on incident positron energy just above threshold, and possible reasons for this structure are discussed. These include the possibility of a dissociative attachment process; a unique peak at similar projectile energies has long been observed in electron-oxygen scattering. The most likely explanation, however, lies in coupling between Ps formation and excitation to the continuum, the latter having previously been shown also to have a peaked energy dependence above threshold.

1. Introduction

Total cross sections for positron scattering by molecular oxygen, Q_{tot}, have been measured by a number of laboratories since 1975 [eg 1-3]. Q_{tot} has an essentially smooth dependence on incident positron energy, and there is reasonable agreement between the different measurements.

In 1980 the positronium (Ps) formation cross section for O_2 was reported by Charlton et al [4]. The measurements were made by detecting the three annihilation gamma photons resulting from ortho-Ps decay in coincidence as the positrons passed through a gas cell. The resulting cross section, Q_{Ps}, exhibited a peak-like structure in the few eV above threshold, in common with most other gases studied. However, this structure was later found to be a result of premature pick-off annihilation of the o-Ps on the gas cell walls.

Interestingly, however, measurement of the total ionisation cross section for positrons on O_2 (ie, Ps formation plus direct ionization), involving ion detection, by the same group in 1993 showed that in O_2, unlike other gases, the near-threshold peak was real [5]. The authors linked this feature to coupling of the Ps formation channel with that for excitation to the continuum, to which we shall return.

In 2005 Marler and Surko [6] measured Q_{Ps} (not $Q_{Ps} + Q_{ion}$) using a trap-based system, by a method based on positron loss from the beam (as used by Fornari et al [7]). They confirmed the presence of a peak in Q_{Ps} above threshold and again invoking coupling with excitation to the continuum.

The present work was initially performed to attempt to settle the apparent differences in absolute magnitudes of the Q_{Ps} values reported in refs. [5] and [6], particularly at the near-threshold peak. This original aim was augmented by the need to try to understand more fully the reasons for the existence of the structure in the energy dependence of Q_{Ps}.

2. Experimental technique

The methodology used to measure Q_{Ps} is essentially that described by Thornton et al [8], and thus will only be briefly summarised here. Positrons from a ~20MBq ^{22}Na source are moderated by two annealed 50% transmission tungsten meshes and their energy distribution narrowed to ~ 800meV by a 92% tungsten cut-off mesh mounted immediately above the moderator. This latter mesh also improves the reflection of positrons scattered back towards the source. The moderated positrons, in a beam of 4mm diameter, are accelerated to the desired mean energy (in the range 2-40eV) by a potential applied to the moderator and are guided by a 7mT axial magnetic field through a 70mm-long gas cell to the 10mm-diameter cone of a channel electron multiplier (CEM) detector 360mm away. The front end of the CEM is held at -2kV, with a fine mesh cover to prevent the 'sucking out' of electrons from its cone and to reduce the background due to fast secondary electrons from the moderator meshes. Between the gas cell and detector is mounted a cylindrical retarding field analyser (RFA) to which a potential can be applied in order to discriminate against scattered positrons.

The positron signal count rate I_0 was determined by measuring the difference between the total CEM count rate and that recorded when the beam was *just* stopped by raising the potential on the cut-off mesh in front of the moderator. With gas continuously bled into the cell – at a density for which the maximum total attenuation was no more than 15% - the count rate is I. When a potential is applied to the RFA which only allows unscattered positrons to pass through (and thus effectively stops those positrons which have been scattered through elastic or inelastic channels), $(I_0\text{-}I)/I_0$ is the 'total' attenuation of the beam, A_{tot}. When no potential is applied to the RFA all positrons which survive the scattering process can pass through, and the only loss is via neutralisation by Ps formation (assuming that direct annihilation is negligible); $(I_0\text{-}I)/I_0$ is then designated A_{Ps}. Q_{Ps} is then deduced from $(A_{Ps}/A_{tot})Q_{tot}$, where Q_{tot} is the total scattering cross section measured previously [1-3].

3. Results

The total scattering cross section Q_{tot} is given by the Beer-Lambert expression $(-1/nL)\ln(1\text{-}A_{tot})$, where nL is the atomic number density-path length product for the gas cell. As nL was held constant for all positron energies, the measured A_{tot} values could be used to obtain the dependence on positron energy E of Q_{tot} for positron-oxygen scattering, which by normalisation could then be compared with earlier measurements – with which reasonable agreement was obtained, as shown in Fig.1.

Fig. 2 shows the results for Q_{Ps} from the current measurements and those of refs. [5] and [6]. The former used the thin-target approximation $Q_{Ps} = (1/nL)A_{Ps}$. If we use this expression then agreement between our results and those of ref. [6] is excellent for energies E up to 20eV. However, the disagreement in the magnitude of Q_{Ps} below the threshold for direct ionisation between the current results and those of ref. [5] is unexplained.

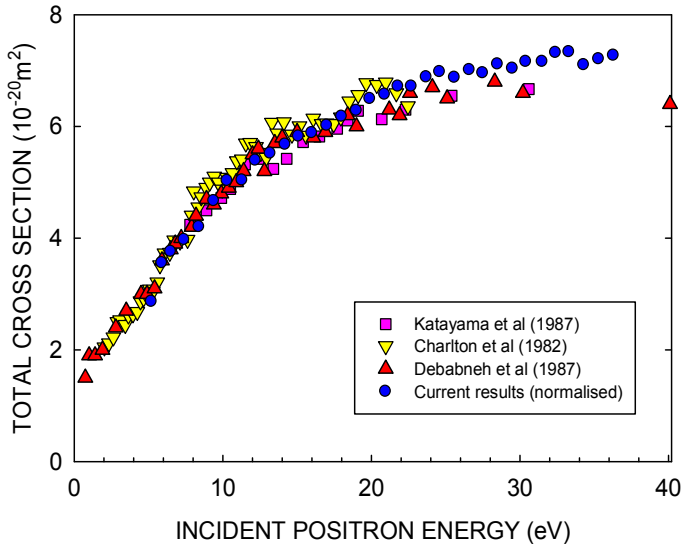

Figure 1. Total scattering cross sections for positron scattering by molecular oxygen.

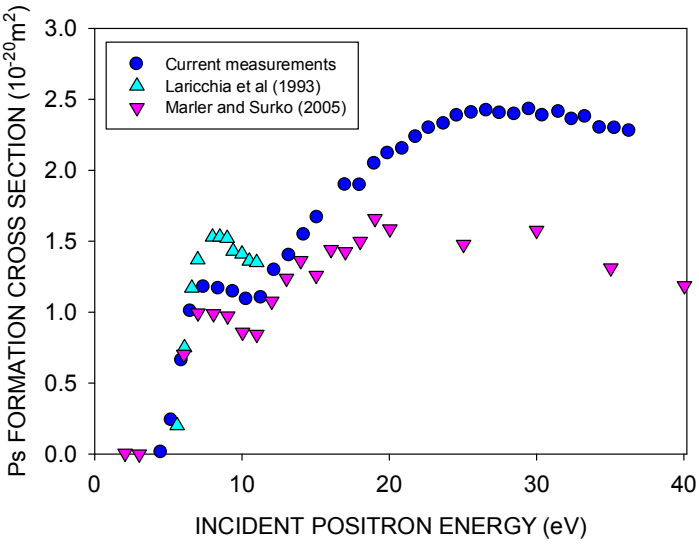

Figure 2. Cross sections for Ps formation in O_2. The results of Laricchia et al. are those for total positron-O_2 ionisation below the threshold for direct ionisation.

The divergence of the current results and those of ref. [6] at higher energies could be explained by an increasing underestimation of A_{tot} as E increases; but this implies a lack of discrimination against small-forward-angle elastic scattering, whereas inelastic processes (against which it is easy to discriminate) become increasingly important at higher E. Rather than discuss further this discrepancy in the magnitude of Q_{Ps} at higher energies between the current results and those of ref. [6], we shall turn instead to the mechanism for the peak in Q_{Ps} in the few eV above threshold.

4. Discussion

Coupling between scattering processes near the threshold for Ps formation has been seen in the noble gases (here between Q_{Ps} and the elastic cross section Q_{el}) [9,10]. It is thus appropriate to consider whether the peak in Q_{Ps} in O_2 above threshold is also evidence of channel coupling.

It is interesting to note that in electron-O_2 scattering there is a pronounced peak in the cross section for the formation of O^- ions (Fig. 3) which occurs at exactly the same energy as the peak in Q_{Ps} [11]. It is tempting to attempt to correlate this unusual peak with that in Q_{Ps}. One possible scenario would involve the dissociative attachment of a positron ($e^+ + O_2 \rightarrow Oe^+ + O$) or Ps ($e^+ + O_2 \rightarrow PsO + O^+$), whereby the positron would be lost directly (via annihilation) or after reorganisation into $Ps + O^+ + O$. Either route would leave an O^+ ion, which would be essential in the measurements of Laricchia et al [5]. However, Cheng et al [13] and others have shown that positrons do not bind to O atoms and, although PsO is possible there is an energy deficit against its formation of ~11.2eV in e^+-O_2 scattering.

Finally, note that the cross section at the peak in Fig. 3 is only about 1% of Q_{Ps} at the peak; we consequently should rule out any dissociative attachment mechanism to explain the structure in Q_{Ps}.

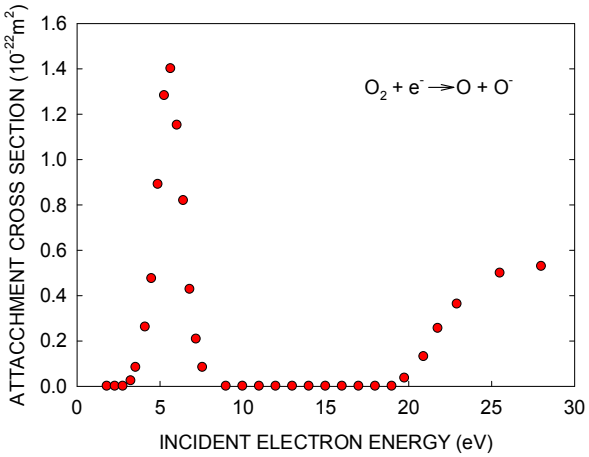

Figure 3. Cross section for the formation of O^- ions via electron-O_2 impact. Data taken from ref.[11] normalised to absolute values using ref [12].

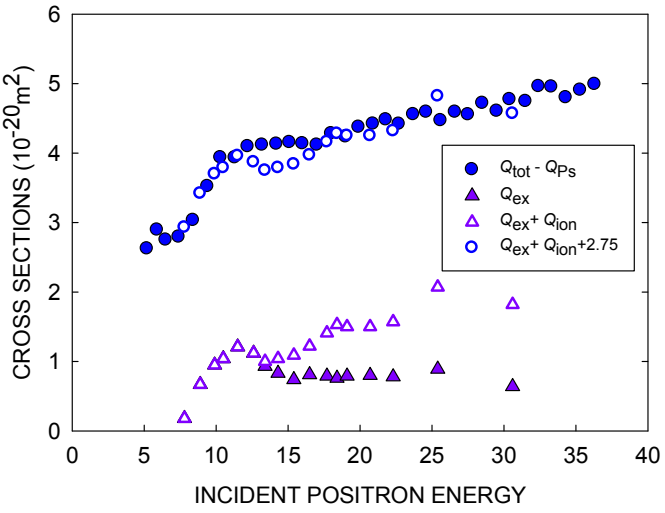

Figure 4. Solid circles: current (normalised) total minus Ps formation cross sections. Open circles: addition of the excitation and ionisation cross sections of Katayama et al [1] to a constant 2.75x10^{-20}m^2 to represent elastic scattering.

Laricchia et al and Marler and Surko [5,6] both linked the effect to coupling with excitation to the (Schumann-Runge) continuum – ie dissociative excitation. Katayama et al [1] measured the excitation cross section Q_{ex} by a positron time-of-flight method – ie the scattered positrons survive the interaction. They saw a significant peak in Q_{ex} above the Schumann-Runge threshold at 7.1eV, implying a broad resonance-like behaviour for positron (but not electron) excitation (see Fig. 4). However, they invoked a coupling between Ps formation and excitation which possibly involves the formation of a PsO_2^+ complex, which in the light of recent calculations does not now seem likely [14].

The question remains as to why Q_{tot} appears to vary smoothly with E, whereas the partial cross sections Q_{Ps} and Q_{ex} have structure. In Fig. 4 the current ($Q_{tot} - Q_{Ps}$) is compared with (Q_{ex}+Q_{ion}) of Katayama et al [1] added to a constant 2.75 x10^{-20}m^2 to represent Q_{el} in this energy range. Apart from the small discrepancy in the 10-15eV region, the two data sets are remarkably similar, suggesting that the structure in Q_{Ps} should not be regarded as a peak centred at ~ 8eV, but a dip at ~ 11eV, where there is strong coupling with dissociative excitation.

Thanks are due to David Schrader and Gleb Gribakin for stimulating discussions on positron and Ps binding with atoms and molecules.

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