Fragmentation in positronium collisions with atoms

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Abstract. Recent positronium collision experiments are reviewed in which the cross-sections for projectile- and/or target-fragmentation processes are determined. In contrast with theory, the likelihood for the latter process appears to be significant for positronium collisions with Xe at only $\sim 3$ eV above threshold. Measurements of the total direct ionization cross-section by positron impact, performed to test the reliability of the experimental apparatus and method, are also presented.

1. Introduction

Positronium (Ps) is the bound state of an electron, $e^-$, and its antimatter partner, the positron, $e^+$. It has half the reduced mass of hydrogen and thus its principal energy levels are correspondingly halved. Over the past twenty years, its controlled production has enabled investigations ranging from its reflection from LiF [1] to its fragmentation in collisions with He [2] and Xe [2, 3].

Fast positronium may be produced by neutralizing a positron beam, for example, by transmission through a thin foil [4], a gas [5, 6, 7, 8, 9], or by scattering it from a surface [10, 11]. The relative merits of these methods with respect to efficiency of production, energy and angular distributions, as well as quantum state population have been previously appraised [12]. At UCL, a beam of Ps atoms is produced using the gas technique according to the charge-exchange reaction [9, 13]:

$$e^+ + A \rightarrow \text{Ps} + A^+.$$  \hspace{1cm} (1)

Several gas targets (A) have been tried, the most efficient being H$_2$ at low energies [14] and N$_2$ up to 250 eV [15]. For these targets, the energy of the Ps beam, $E_{\text{Ps}}$, has been found to be consistent with the formation of ground-state Ps according to:

$$E_{\text{Ps}} = E_+ - E_i + \frac{6.8 \text{ eV}}{n^2},$$  \hspace{1cm} (2)

where $E_+$ is the energy of the positron beam, $E_i$ is the first ionisation energy of the production gas and $6.8 \text{ eV}/n^2$ is the binding energy of the Ps atoms of principal quantum number $n$. The formation of Ps in an excited-state has recently been found to make an increasingly significant contribution — of maximum probabilities 0.06, 0.12 and 0.23, respectively — to the total Ps formation cross-section of
Table 1. Lowest energy reactions in Ps - He scattering in which a free positron and/or electron is produced in the final state.

| Reaction                  | Threshold (eV) | Description                               |
|---------------------------|----------------|-------------------------------------------|
| (a) Ps + He → e⁺ + e⁻ + He | 6.8            | Ps fragmentation                          |
| (b) Ps + He → Ps⁻ + He⁺   | 24.2           | Ps negative ion formation                  |
| (c) Ps + He → Ps⁺ + e⁻ + He⁺ | 24.5         | Target-ionisation                          |
| (d) Ps + He → e⁺ + e⁻ + He* | 27.0           | Ps fragmentation and target-excitation     |
| (e) Ps + He → Ps*⁺ + e⁻ + He⁺ | 29.7     | Ps excitation and target-ionisation        |
| (f) Ps + He → e⁺ + 2e⁻ + He⁺ | 31.4           | Ps fragmentation and target-ionisation     |

Over the past two decades, the Ps beam at UCL has been used to measure total cross-sections, $Q_{Ps}^{T}$ [14, 15, 20, 21, 22, 23], and more recently fragmentation cross-sections [2, 3, 22]. In collisions with atoms and molecules, Ps may break-up if its kinetic energy exceeds 6.8 eV and experimentally this may be probed by detecting the residual positron or electron. Whilst detection of the positron is a unique signature of Ps fragmentation, detection of the electron may also indicate target ionisation.

2. Fragmentation in Ps–He collisions

Experimental investigations were carried out on He by Armitage et al. [2] in which the integrated fragmentation cross-sections, $Q_{f}^{T}$, were determined for the ejected positron at Ps energies $E_{Ps} = 13, 18, 25$ and $33$ eV as shown in Figure 1. Detection of the ejected positrons encompasses reactions (a), (d) and (f) in Table 1. In the study of Armitage et al. [2], the longitudinal energy distributions of the ejected positrons were also determined via a time-of-flight technique. As shown in Figure 2, a peak was found just below half of the residual energy, $E_{f} = E_{Ps} - 6.8$ eV, indicative of the electron loss to the continuum (ELC) process in which the ionised projectile and the ejected electron remain strongly correlated with a small relative velocity. The shift of the peak to lower energies from $E_{f}/2$ observed was ascribed to the ejected positrons being emitted in the laboratory frame within a ∼20° cone around the beam axis; a result supported by a classical trajectory Monte Carlo (CTMC) [24] and an impulse approximation calculation [25], the former agreeing in shape and the latter in shape and magnitude with the experimental data. In addition to the errors arising from statistical uncertainties in the absolute scale in Figures 1 and 2, there is a systematic uncertainty of (+8, -20-30)% arising from the determination of detection efficiencies. It is an energy-independent error which equally affects each value of $Q_{f}^{T}$ and $dQ_{f}^{T}/dE_{f}$. 
Figure 1. Integrated cross-sections for He. Experiment: $Q^+\!$: •, [2]; ▲, [22]. $Q^-\!$: ○, [22]. Theory: ———, CTMC [24]; — · · · · · , coupled-state calculation [26]; — — —, impulse approximation [25]; ·······, Coulomb–Born approximation [27]. An additional uncertainty of (+8, -(20-30))% exists on the absolute scale for the experimental $Q^+\!$ and $Q^-\!$.

Figure 2. Differential fragmentation cross-sections, $dQ^+\!/dE_\ell$, for Ps+He collisions at 13, 18, 25 and 33eV Ps incident energy. •, [2]; ———, CTMC [24] multiplied by 0.5; — — —, impulse approximation [25]. Average $dQ^+\!/dE_\ell < 1$ eV: ○, [25]. † indicates the position of $E_\ell/2 = (E_{Ps} - 6.8 \text{ eV})/2$. 
When detecting the ejected electrons from Ps–He collisions, the experiment is sensitive to all the reactions in Table 1. Thus the almost equality between $Q_f^-$ and $Q_f^+$, measured by Armitage et al. [22], implies a negligible cross-section for target-ionisation, $Q_{Ps}^i$, at $E_{Ps} = 30$ eV (see Figure 1). This result is supported by the First Born approximation calculation of Walters et al. [28] employed to determine the contribution from target-inelastic effects — i.e. target-excitation and -ionisation — to supplement the earlier, target-elastic impulse approximation of Starrett et al. [25]. It is noted that should the Ps negative ion, Ps$^-$, be produced, its lifetime of 0.48 ns against annihilation [29] means an electron would remain available for detection and contribute to the measured $Q_f^-$. To determine $Q_f^-$, the Ps beam, produced in cell 1 via the charge exchange reaction of Equation 1, was impinged against the target gas in cell 2 (see Figure 3). Any positrons not neutralised were reflected by a positive potential on CR1. Cell 2 was floated at a negative potential, $V_{cell}$, to accelerate the ejected electrons (from Ps fragmentation or target-ionisation) to energies much greater than secondary electrons released from surfaces. In this way, electrons from Ps–He collisions were separated from the background, and the longitudinal energies of the signal-electrons were analysed by a pair of retarding grids, R2 and R3. Electrons and positrons ejected from cell 2 were confined by an axial magnetic field of 22 mT and were detected by a channel electron multiplier array (CEMA). Whilst positrons ejected in the backwards direction were reflected by CR1 and thus detected, electrons ejected backward were lost due to attenuation. For this reason, all present experimental values of $Q_f^-$ should be considered to be lower limit estimates.

3. Fragmentation in Ps–Xe collisions

Preliminary investigations for Xe were performed by Armitage et al. [22], in which $Q_f^-$ — determined using the retarding field method above — was found to be almost a factor-of-two larger than $Q_f^+$ at $E_{Ps} = 30$ eV, suggesting the occurrence of target-ionisation (see Table 2). This work was supplemented by that of Brawley et al. [3], as shown in Figure 4, where the results of $dQ_f^+/dE_\ell$ for 18 and 30 eV Ps collisions are compared to an available theory [28]. Here $dQ_f^+/dE_\ell$ may be seen to display a peak just below $E_\ell/2$ — consistent with the ELC process, as observed in the time-of-flight work for He [2]. However, it should be noted that the extraction efficiency of positrons with energies below 1 eV from the main body of the cell is estimated to be $\lesssim 50\%$. Figure 4 also shows the results of $dQ_f^+/dE_\ell$ at $E_{Ps} = 30$ eV and those of the target-elastic impulse approximation theories of Walters et al. [28] at $E_{Ps} = 33$ eV. At this Ps impact energy, reactions (d) and (f) in Table 2 are energetically accessible, in
Table 2. Lowest energy reactions in Ps - Xe scattering in which a free positron and/or electron is produced in the final state.

| Reaction | Threshold (eV) | Description |
|----------|---------------|-------------|
| (a) Ps + Xe $\rightarrow$ e$^+$ + e$^-$ + Xe | 6.8 | Ps fragmentation |
| (b) Ps + Xe $\rightarrow$ Ps$^-$ + Xe$^+$ | 11.8 | Ps negative ion formation |
| (c) Ps + Xe $\rightarrow$ Ps$^-$ + e$^-$ + Xe$^+$ | 12.1 | Target-ionisation |
| (d) Ps + Xe $\rightarrow$ e$^+$ + e$^-$ + Xe$^*$ | 15.1 | Ps fragmentation and target-excitation |
| (e) Ps + Xe $\rightarrow$ Ps$^*$ + e$^-$ + Xe$^+$ | 17.2 | Ps excitation and target-ionisation |
| (f) Ps + Xe $\rightarrow$ e$^+$ + 2e$^-$ + Xe$^+$ | 18.9 | Ps fragmentation and target-ionisation |

Figure 4. $dQ^+/dE_\ell$ for Ps collisions with Xe at 18 eV and 30eV. •, [3] at $E_{Ps} = 30$ eV; ———, [28] for $E_{Ps} = 33$ eV; ↑, $E_t$/2; ↓, $E_t$.

addition to reaction (a), and may account for the differences between these results and the target-elastic data [28].

Measurements of $Q^+$ and $Q^-$ by Brawley et al. [3] are shown in Figure 5 together with the calculations of Walters et al. [28] and Starrett et al. [30]. An additional uncertainty in the absolute scale of (+30, -50)% of the same kind as described above applies to these measurements. As before, due to the fact that backward-ejected electrons remain undetected, $Q^-_t$ must be considered a lower limit. Also, the difference between $Q^+_t$ and $Q^-_t$ yields $Q^{Ps}_t$, for processes in which target-ionisation occurs without Ps fragmentation, namely reactions (b), (c) and (e) in Table 2. However, some overestimate from reaction (f) may occur if the two electrons in the final state are separated sufficiently at the CEMA to be detected independently. Considering the variety of energies and ejection angles that the two electrons may have, it seems likely that this will occur. Once again, the experimental $Q^-_t$ may include Ps$^-$ formation not yet included in the theories.
Figure 5. $Q^+_f$ and $Q^-_f$ for Ps collisions on Xe. Experiment: •, $Q^+_f$; ○, $Q^-_f$. Theory: ———, target-elastic integrated fragmentation work of [28] for the ejection of positrons; — — —, $Q^+_f$ work of [30]; ——·—·—, $Q^-_f$ work of [30]. There is an additional uncertainty of (+30, -50)% on the absolute scale for the experimental $Q^+_f$ and $Q^-_f$.

In view of the excess of $Q^-_f$ over $Q^+_f$ and the considerable difficulties in discriminating signal electrons from the significant backgrounds of secondary electrons, further tests were undertaken to gauge the reliability of the experimental system and method. These consisted of measurements of the direct ionisation cross-section for positron impact ($Q^+_i$). This has been determined at 42 and 62 eV for Ar, and at 40 and 55 eV for Xe. As in the case of Ps impact, ejected electrons are confined by the magnetic field — of the same strength as for the Ps experiments — and accelerated by $V_{cell}$ to be detected by the CEMA plates (see Figure 3). For this experiment, the positron collision energy in the cell is $E^+_c = E_+ + eV_{cell}$, and thus a value of $V_{cell} = -22$ V was chosen in order to accelerate the ejected electrons whilst not exceeding the maximum transverse energy of 64 eV which may be confined by the field. A positive potential of 250 V is applied to an electrode (RFA) to reflect any positrons that emerge from the scattering cell. The emerging electrons that reach the detection region are energy analyzed in the same way as in the case of the Ps fragmentation experiments above. The background originating from secondary electrons is taken into account and subtracted in order to determine the net number of electrons from ionisation events.

The ionisation cross-section by positron impact, $Q^+_i$, was then calculated as follows:

$$Q^+_i = \frac{N_-}{N^+_\text{scatt}} Q^+_T \left( \frac{\epsilon_+}{\epsilon_-} \right),$$

(3)

where $N_-$ is the net number of electrons detected from positron collisions, $N^+_\text{scatt}$ is the number of scattered positrons in cell 2, $Q^+_T$ is the positron total cross-section for the target gas, and the last term is the ratio of the detection efficiency of positrons to that of electrons by the CEMA plates (in this work equal to 1.60 ± 0.06 [31]). Whilst only electrons emitted within ±90° of the central beam axis may be detected, most of the contribution (~90%) to $N_-$ from the backward angles are included in the measurements by reflecting the transmitted positrons at the RFA. Although multiple-ionisation accounts for ≤ 10% of all direct ionisation events [32], it may contribute to the present cross-sections and, in principle, with enhanced sensitivity due to the availability of more than one electron in the final state. Thus in comparing with
other measurements, it is more meaningful to label the present measurements as $Q_{n+i}^+$, equal to the sum of the cross-sections for all degrees of direct ionisation. The present absolute results of $Q_{n+i}^+$ for Ar and Xe — as calculated using Equation 3 — are shown in Figure 6. In both cases, there is an additional error in the absolute scale of ± 5% in the present data due to uncertainties in calculating $(\epsilon_{+}/\epsilon_{-})$. For Ar, there is consistency between the present $Q_{n+i}^+$ and those of Marler et al. [33] and the $Q_i^+$ work of Moxom et al. [34] for 42 eV positron collisions. The agreement is less pronounced at the highest energy investigated, although it is within errors. For Xe, there is a fairly good consistency at both energies between the present work and that of Marler et al. [33]. At both energies, the present $Q_{n+i}^+$ are ∼10% higher than the $Q_i^+$ reported by Kara et al. [35].

The consistency of the present results for $Q_{n+i}^+$ with previous measurements (by two different methods), implies that contributions from secondary electrons to the background have been properly accounted for, thus affording confidence in $Q_i^+$.

4. Conclusion and outlook

In conclusion, Ps fragmentation and target-ionisation investigations carried out at 18 and 30 eV Ps collisions with He and Xe, suggest significant target-ionisation effects from the latter target at the higher energy, in disagreement with theory. The system reliability has been tested by performing positron impact ionization measurements which are found to be consistent with available data. Ps fragmentation was first extensively investigated theoretically by Ludlow and Walters [36] at high impact energy (500 eV). Whilst these conditions have not yet been amenable to experimental investigations, recently, Ps− (first observed by Mills [37]) has been produced with significant efficiencies [38] using a surface doping technique as suggested in [12, 32]. These developments open up the prospect of being able to produce monoenergetic Ps beams of useful intensities at high energies by accelerating Ps− and by photodetaching one electron — a method first proposed by Mills and Crane [4]. Much future activity is envisaged.
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