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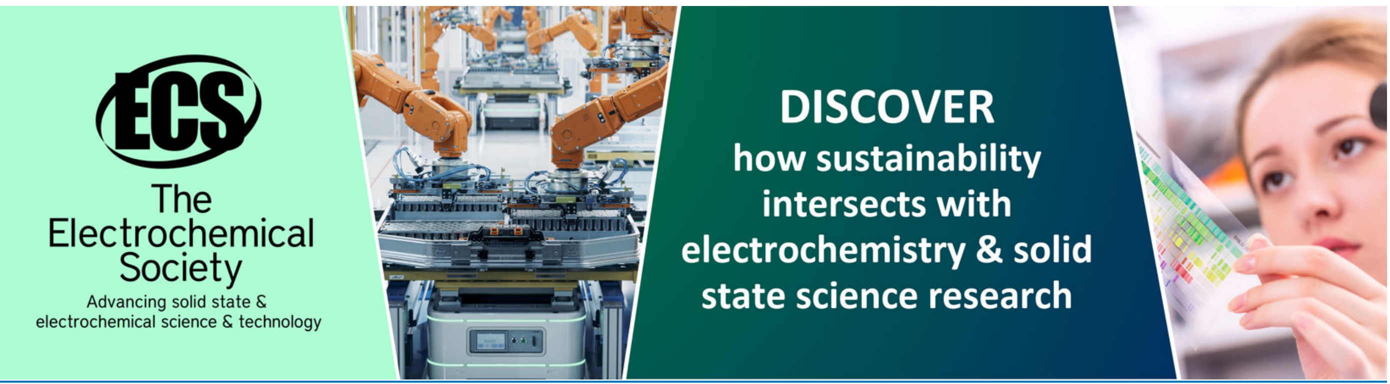

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Ionization in positron- and positronium- collisions with atoms and molecules

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Abstract. Recent progress in the experimental study of positron- and positronium-induced ionization of atoms and molecules is outlined. Investigations include integral and differential cross-sections, as well as formation of positronium in the first excited state. Future prospects are discussed.

1. Introduction

The study of positron and positronium collisions with atoms and molecules is motivated by the need to understand basic matter–antimatter interactions, to support the development of scattering theories, to assist the analysis of astrophysical events and tests of QED bound-state problems as well as calculations of radiation damage at the molecular level for positron-emission tomography e.g. [1–3].

In this article, we review recent progress made at UCL in the measurement of cross-sections for ionization processes arising from collisions of positrons and positronium atoms with atomic and molecular targets.

2. Positron induced ionization

In collisions between a positron and an atomic/molecular target (X), ionization may proceed via a number of channels: annihilation, transfer and direct ionization. These are summarized, respectively, by reactions 1–3 below:

$$X + e^{+} \longrightarrow X^{z+} + 2\gamma + (z-1)e^{-} \tag{1}$$

$$X + e^{+} \longrightarrow X^{z+} + Ps + (z-1)e^{-} \tag{2}$$

$$X + e^{+} \longrightarrow X^{z+} + e^{+} + ze^{-} \tag{3}$$

where Ps and/or X^{z+} in the final state may be excited, and z corresponds to the number of electrons removed from the target. If X is a molecule, the above reactions may be accompanied by dissociation. The total ionization cross-section (Q_i^t, defined as the sum of the cross-sections for all ion producing processes) is dominated by the cross-sections for Ps formation (Q_{Ps}) and single direct ionization (Q_i^+) (reactions 2 and 3 with $z = 1$) above their respective thresholds, E_{Ps}^{thr} and E_i^{thr}. Being an exothermic reaction, annihilation is the only possible ionization channel below E_{Ps}^{thr}. It is considered generally negligible except at very low energies [4], although enhancements in the annihilation probability have been observed below E_{Ps}^{thr} near vibrational excitation thresholds and associated with the formation of vibrational Feshbach resonances [3].

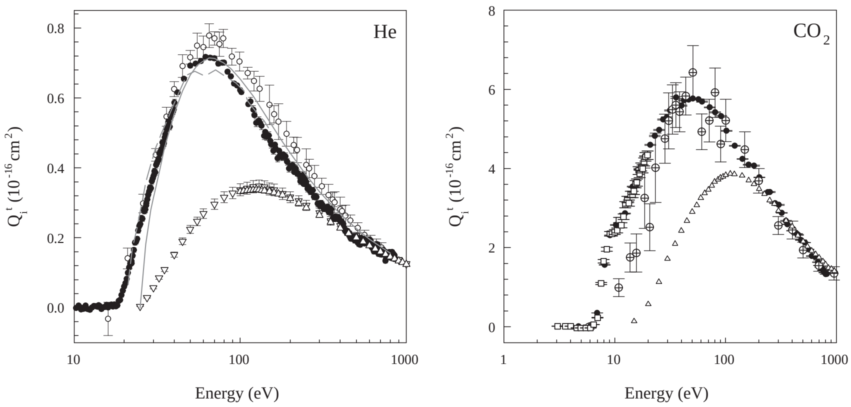

Figure 1. Theoretical and experimental determination of $Q_i^t(\mathrm{e}^+)$: **Left:** He ●—[5], ○—[6], grey dashed curve—[7], solid grey curve—[8]. Corresponding results for e$^-$ impact, $Q_i^t(\mathrm{e}^-)$, shown for comparison: △—[9], ▽—[10]; **Right:** CO_2, ●—[11], □—[12], ⊕—[13], △—$Q_i^t(\mathrm{e}^-)$ [14].

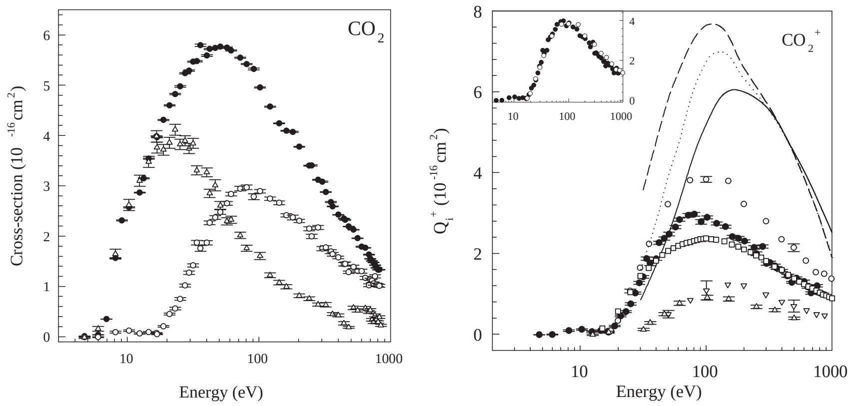

Figure 2. Left: Partitioning of Q_i^t for CO_2 [11, 15] into Q_{Ps}—△and Q_i^+—○. **Right:** Q_i^+ for CO_2 compared with theory and experiment. ●—[11], ○—[13], □—[14], ▽—Q_i^{diss} [13], △—Q_i^{diss} [15], all curves—[16]. **Inset:** normalized [15] and [13] to illustrate identical energy dependence.

Results of $Q_i^t(\mathrm{e}^+)$ for He and CO_2 are shown in figure 1 where some discrepancies may be noted among experiments. However in He, there is excellent agreement between the data of [5] and the coupled-pseudostate calculation of [7], the maximum being better described by the results of [8]. In CO_2, there is excellent agreement in shape between the high-resolution measurements of [12] and the absolute determination of [11], the latter also agreeing in magnitude at higher energies with the earlier data of [13]. $Q_i^t(\mathrm{e}^+)$ may be seen to exceed corresponding results for electron-impact $Q_i^t(\mathrm{e}^-)$ at low and intermediate energies primarily due to Ps formation, as illustrated for CO_2 in figure 2 (LHS). Whilst for He (and indeed all the noble gases) Q_{Ps} tends to zero around 100–150 eV, positronium formation in CO_2 remains a significant channel at much higher energies [11].

Concerning direct ionization, as discussed in [2], there is good accord for He among experimental determinations e.g. [5, 17, 18] and with theories [7, 19–22], however the energy region within 1 eV of the threshold remains a major experimental challenge. In the case of CO_2, as shown in figure 2 (RHS), there is excellent shape agreement over the whole energy range between experimental results [13, 15], the

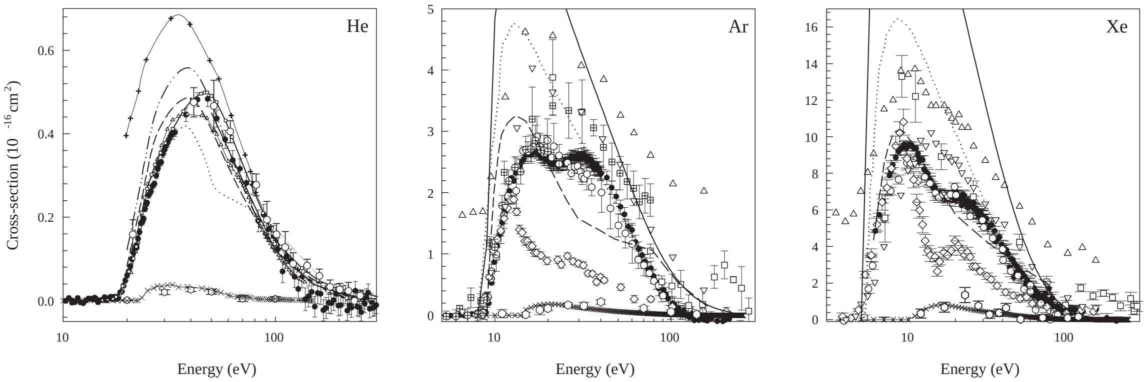

Figure 3. Left: Q_{Ps} for He, ●—[5], ○—[23], dotted black curve—[24], curve with △—[25], black chain curve—[7], solid black curve (from 50 eV)—[26], curve with + —[27], black double chain curve—[28], curve with □—[29], curve with ×—$Q_{Ps}(2P)$ [7], △—$Q_{Ps}(2P)$ [30]. **Centre:** Q_{Ps} for Ar, ●—[31], ○—[32], ◇—[33], ⊕—[34], □— [35], ⊞—[36], ▽—[37] LL, △—[37] UL, solid black curve—[38], dashed black curve—[39], dotted black curve—[40], curve with ×—$Q_{Ps}(2P)$ [38], ⬡—$Q_{Ps}(2P)$ [30]. **Right:** Q_{Ps} for Xe (symbols as for Ar).

discrepancy being entirely attributable to the electron data chosen for normalization (as illustrated in the inset) whilst the distorted-wave-Born-approximation (DWBA) results of [16] exceed experimental data by a factor of 2–3. At its maximum, the cross-section for dissociative direct ionization (Q_i^{diss}) accounts for approximately 20% of $Q_i^t(\mathrm{e}^+)$ for CO_2 [15].

Convergence has considerably improved in recent measurements of Q_{Ps} for the inert atoms [2], as illustrated in figure 3 for He, Ar and Xe. Whilst in helium, there is also good agreement between experimental and theoretical determinations, the situation for more complex targets is less satisfactory. Differences remain among experiments concerning structure around the peak and even greater discrepancies exist between experiment and theory, the latter overestimating measurements by a factor 2–3, although [40] and [41] found that inclusion of higher order processes leads to a significant reduction of the cross-section magnitude. Both the existence and the significance of the structure apparent in some of the experiments has been the subject of some speculation. Ps formation from higher thresholds has been considered either via capture of an inner-shell (*ns*) electron or Ps formation in an excited state (Ps*). An analysis based on an empirical scaling for ionization cross-sections [42] predicted increasing contributions of Ps* with decreasing ionization energy, I [31]. A DWBA method [38] found *ns* contributions to be very minor whilst Ps* gave rise to structure similar to that observed experimentally. Cross-sections for formation of Ps into the 2P state ($Q_{Ps}(2P)$) have now been measured [30]. The results are included in figure 3 where they are compared with corresponding theories. In He, the best agreement is with the coupled-pseudostate calculation [7]. In Ar and Xe, whilst the DWBA overestimates Q_{Ps}(all n) by factor 2–3, its predictions agree fairly well for 2P states. Interestingly, $Q_{Ps}(2P)$ is found to make a significant contribution to Q_{Ps}(all n) which increases from 6% in He to 23% in Xe.

Differential investigations of ionization by positron projectiles are scant. Triple differential studies have been carried out at UCL around 0° by measuring coincidence between scattered e^+ and ionized e^-. At 100 eV incident positron energy, a small peak was observed in the spectrum of the electrons ejected from the H_2 target at half-the-residual energy, E_r [47], a signature of the electron-capture to the continuum (ECC) phenomenon predicted ten years earlier [48]. Instead at 50 eV, an asymmetry between the energy spectra of electrons and positrons was found [49]. As shown in the LHS of figure 4, the electron spectrum was shifted by around 1.6 eV with respect to quantum theoretical expectations [44] whilst being in good agreement with classical-trajectory-Monte -Carlo calculations [45]. This latter

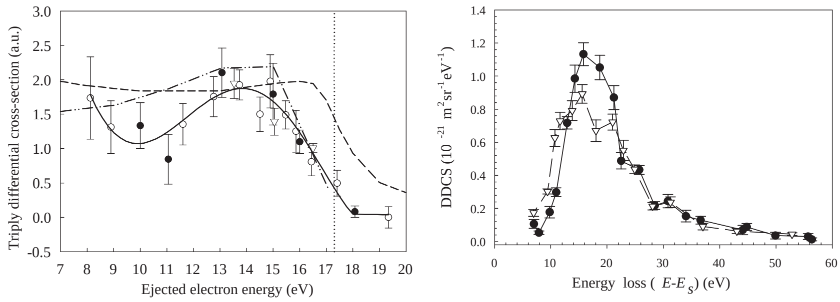

Figure 4. Left: Experimental and theoretical results for triply differential cross-sections for ejected electrons in 50 eV positron collision with H_2, D_2 and He: ●—D_2, ○—H_2, ▽—He at same E_r [43], solid curve—fit to experimental data as a guide to the eye; dashed curve—[44], double chain curve—[45], dotted line—$E_r/2$. **Right:** Double differential cross-section as a function of the energy loss of the scattered e^+ projectile in coincidence with H_2O^+ fragments: ●—100 eV [46], ▽—153 eV [46].

approach, however, failed to describe the 100 eV data [47]. All these findings are the subject of current theoretical scrutiny (e.g. [50]). More recently, investigations have been extended to H_2O because of its universal importance and in order to probe whether its strong dipole moment, responsible for strong forward-scattering of electron projectiles [51], might result in the ECC cusp becoming conspicuous at the doubly differential level as is the case by ion impact [52]. The energy distributions of e^+ scattered around 0° from H_2O were measured in coincidence with the remnant ions (H_2O^+, OH^+ and H^+) at 100 and 153 eV incident energy [46]. The maxima of the double differential cross-ections (DDCS) associated with the production of OH^+ and H^+ were found to be about 5–10 times smaller than that for H_2O^+ and the shape was observed to be similar to non-polar targets. As shown in the RHS of figure 4, at both incident energies, a small shoulder in the energy loss spectra associated with H_2O^+ production was seen around 28 eV. This feature appears consistent with e^- momentum spectroscopy results [53] which identifies it with the onset of a weak shake-up band at 27.1 eV connected with the $2a_1$ orbital. Further investigations would be justified.

3. Positronium induced ionization

A positronium atom makes an interesting projectile as it has no nucleus, its constituents having the same mass and opposite charge [e.g. 2]. Since both target and projectile have structure, ionization may be accompanied by excitations of either or both colliding partners, namely: projectile fragmentation, Ps^- formation, target ionization, projectile fragmentation with target excitation, target ionization with projectile excitation and, finally, projectile fragmentation with target ionization, as summarised in reactions 4–9 below:

$$A + Ps \to A + e^+ + e^- \tag{4}$$

$$A + Ps \to A^+ + Ps^- \tag{5}$$

$$A + Ps \to A^+ + e^- + Ps \tag{6}$$

$$A + Ps \to A^* + e^+ + e^- \tag{7}$$

$$A + Ps \to A^+ + e^- + Ps^* \tag{8}$$

$$A + Ps \to A^+ + 2e^- + e^+ \tag{9}$$

Reaction 4 is the only one not involving a change in the internal energy of the target and is referred to as target-elastic (TE); all the others are said to be target-inelastic (TI). Experimentally, these have been

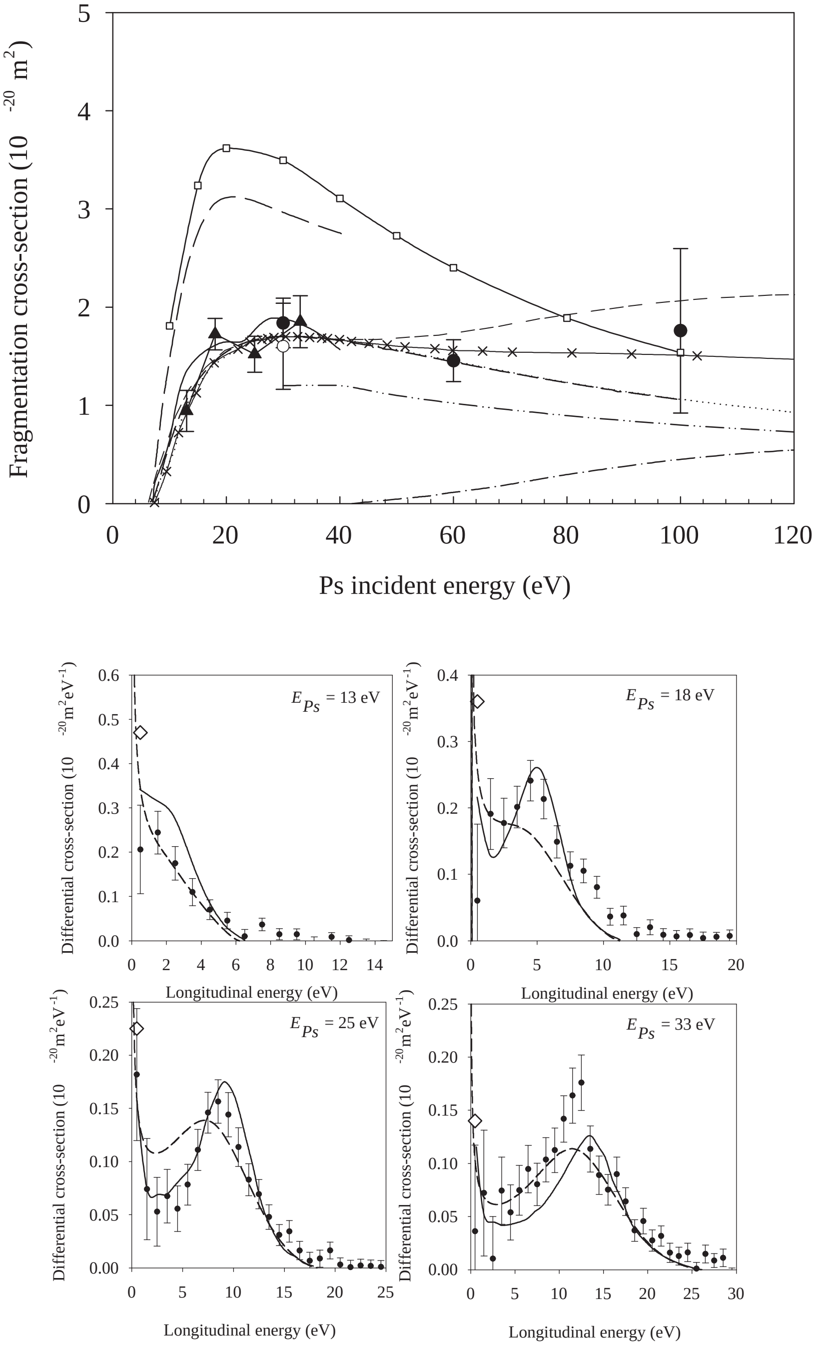

Figure 5. Top: The fragmentation cross-sections for Ps impact on He. ▲—Q_f^+ [54], ●—Q_f^+ [55], ○—Q_f^- [55], curve with □—TE [56], long dashed curve—TE [57], solid curve—TE [58], double chain curve—TE [59], medium dashed curve—TE [60], curve with ×—TE+TI Q_f^+ [61], short dashed curve—TE+TI Q_f^- [62]. **Bottom:** The absolute single differential cross-section dQ_f^+/dE_ℓ for the fragmentation of Ps in collision with He atoms. In this figure, longitudinal energy refers to that of the ejected positron and E_{Ps} to the incident energy of the Ps projectile. ●—[54], solid curve—[57] $\times 0.5$, dashed curve—[60], ◇—Average value < 1 eV [60].

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investigated by detecting the positron or the electron in the final state: the total Ps fragmentation cross-section Q_f^+ (corresponding to the sum of the cross-sections for all processes involving the break-up of Ps) is measured when detecting positrons; the total fragmentation cross-section Q_f^- (corresponding to the sum of the cross-sections for all target and projectile ionization channels) is determined when detecting electrons. The differential cross-section with respect to the (longitudinal) energy of the ejected positron (dQ_f^+/dE_ℓ) has also been determined by a time-of-flight method [54] and by retarding field analysis [55, 63].

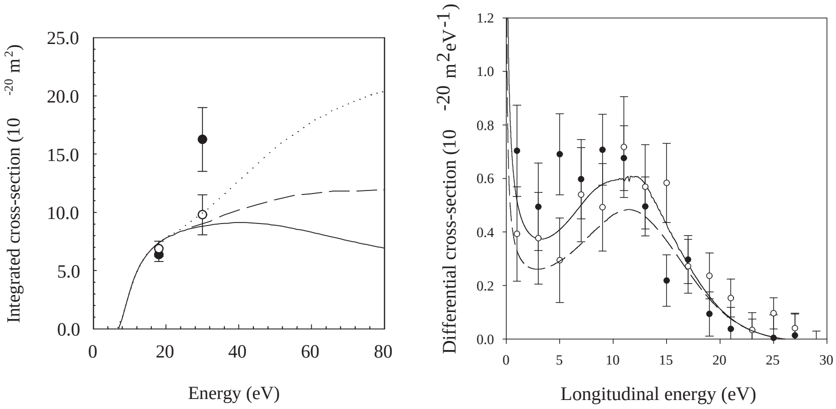

Figure 6. Left: The fragmentation cross-section for Ps impact on Xe. ●—Q_f^+ [63], ○—Q_f^- [63], solid curve—TE [61], dashed curve—TE+TI Q_f^+ [62], dotted curve—TE+TI Q_f^- [62]. **Right:** Longitudinal energy distributions of the ejected positrons form Ps collisions with Xe at 30 eV. ●—[63], solid line—[61]; ○—corresponding He data ×4 [54], dashed line— He×4 [60].

The results for He are shown in figure 5. In the top figure, both Q_f^+ and Q_f^- may be seen to agree with a coupled-pseudostate calculation [58] and an impulse approximation [60] supplemented by a first Born calculation for target inelastic processes [62]. The dQ_f^+/dE_ℓ shown in the bottom figure display a peak which grows in significance with positronium incident energy and arises from the occurrence of electron-loss to the continuum (a phenomenon related to ECC) where, following Ps break-up, the electron and the positron in the final state move with a small relative velocity. The agreement in shape with the results of the classical-trajectory Monte Carlo calculation [57] is very good and that with the impulse approximation [60] is good both in shape and absolute magnitude.

In figure 6, corresponding results for xenon are displayed. On the left, Q_f^- may be seen to exceed Q_f^+ at 30 eV, implying a degree of target ionization, contrary to theoretical expectations. On the right, the experimental dQ_f^+/dE_ℓ results for Xe [63] are compared with theory [61] with which they are in broad accord. Also included in the figure are the corresponding experimental results for He multiplied by a factor of 4 for shape comparison: the distributions for the two targets appears very similar, except perhaps at the lowest energy.

4. Conclusions and outlook

Recent progress in the study of ionization induced by positron and positronium impact on atoms and molecules has been presented. Results now comprise both integral and differential cross-sections, with and without Ps formation in the case of positron impact, and with and without target ionization for positronium projectiles. Whilst exploration of molecular targets is comparatively less advanced, investigations are now progressing to photon–ion coincidences to probe reactions where the target ion is left in an excited state [11]. The pace is expected to quicken further with the realization of positron reaction microscopes which are currently under development [e.g 64].

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