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

G. Laricchia, S. Brawley, D. A. Cooke, Á. Kövér*, D. J. Murtagh and A. I. Williams

UCL Department of Physics and Astronomy, University College London, Gower Street, London WC1E 6BT, United Kingdom

*permanent address: ATOMKI, Institute for Nuclear Research, Debrecen, Hungary

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:

\[ X + e^+ \rightarrow X^{z+} + 2\gamma + (z - 1)e^- \]  \hspace{1cm} (1)
\[ X + e^+ \rightarrow X^{z+} + Ps + (z - 1)e^- \]  \hspace{1cm} (2)
\[ X + e^+ \rightarrow X^{z+} + e^+ + ze^- \]  \hspace{1cm} (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 (\(Q_t\), 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].
Results of $Q^+_i(e^+)$ 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]. $Q^+_i(e^+)$ may be seen to exceed corresponding results for electron-impact $Q_i^-(e^-)$ 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
whilst being in good agreement with classical-trajectory-Monte Carlo calculations. This latter 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 ($Q_{P^i}$) accounts for approximately 20% of $Q_{P^i}(e^+)$ for CO$_2$ [15].

Convergence has considerably improved in recent measurements of $Q_{P^i}$ 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 2$P$ state ($Q_{P^i}(2P)$) 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 $Q_{P^i}(all n)$ by factor 2–3, its predictions agree fairly well for 2$P$ states. Interestingly, $Q_{P^i}(2P)$ is found to make a significant contribution to $Q_{P^i}(all n)$ 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 discrepancy...
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: $\bullet$—D$_2$, $\circ$—H$_2$, $\nabla$—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$_2$O$^+$ fragments: $\bullet$—100 eV [46], $\nabla$—153 eV [46].

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:

\[
\begin{align*}
A + \text{Ps} & \rightarrow A + e^+ + e^- \\
A + \text{Ps} & \rightarrow A^+ + \text{Ps}^-
\end{align*}
\]

\[
\begin{align*}
A + \text{Ps} & \rightarrow A^+ + e^- + \text{Ps} \\
A + \text{Ps} & \rightarrow A^+ + e^+ + e^- \\
A + \text{Ps} & \rightarrow A^+ + e^- + \text{Ps}^* \\
A + \text{Ps} & \rightarrow A^+ + 2e^- + e^+
\end{align*}
\]

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], solid curve—TE [58], long dashed curve—TE [57], medium dashed curve—TE [59], short dashed curve—TE+TI $Q^+_f$ [60], double chain curve—TE+TI $Q^-_f$ [62]. Bottom: The absolute single differential cross-section $dQ_f/dE_{\ell}$ 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] ×0.5, dashed curve—[60], ◊—Average value < 1 eV [60].
investigated by detecting the positron or the electron in the final state: the total Ps fragmentation cross-section \( Q^+ \) (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 \( Q^- \) (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 \( (dQ^+ / dE) \) 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. \( \bullet - Q^+_f \) [63], \( \bigcirc - Q^-_f \) [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 from Ps collisions with Xe at 30 eV. \( \bullet - [63], \) solid line—[61]; \( \bigcirc - \) corresponding He data \( \times 4 \) [54], dashed line—He \( \times 4 \) [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 \( dQ^+_f / dE \) 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 \( dQ^+_f / dE \) 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].

5. Acknowledgments
We wish to thank John Dumper and Rafid Jawad for expert technical support and express our gratitude to the Engineering and Physical Science Research Council UK (Grant no. GR/S16041/01, EP/E053521/1), the Hungarian Scientific Research Foundation (NKTH-OTKA, Grant No. 67719, OTKA K73703), the European Union (HPRN-CT-2002-00179 EPIC) and COST-STSM-P9-02555 for funding this research.

References
[1] Gribakin G and Walters H R J (eds) 2008 Proceedings of the XIV International Workshop on Low Energy Positron and Positronium Physics vol 266 (Nucl. Inst. Meth. B)
[2] Laricchia G, Armitage S, Kövér Á and Murtagh D J 2008 Adv. At. Mol. Opt. Phys. vol 56 (Elsevier) pp 1–47
[3] Surko C M, Gribakin G F and Buckman S J 2005 J. Phys. B 38 R57
[4] Van Reeth P, Laricchia G and Humberston J W 2005 Physica Scripta 71 C9
[5] Murtagh D J, Szluiniska M, Moxom J, P Van Reeth and Laricchia G 2005 J. Phys. B 38 3857
[6] Fromme D, Kruse G, Raith W and Sinapius G 1986 Phys. Rev. Lett. 57 3031
[7] Campbell C P, T McAlinden M, Kernoghan A A and Walters H R J 1998 Nucl. Instr. Meth. B 143 41
[8] Wu H, Bray I, Fursa D V and Stelbovics A T 2004 J. Phys. B 37 1165
[9] Sorokin A A, Beigman I L, Bobashev S V, Richter M and Vainshtein L A 2004 J. Phys. B 37 3215
[10] Rejoub R, Lindsay B G and Stebbings R F 2002 Phys. Rev. A 65 042713
[11] Cooke D A, Murtagh D J and Laricchia G 2009 In preparation
[12] Laricchia G and Moxom J 1993 Phys. Lett. A 174 255
[13] Bluhme H, Frandsen N P, Jacobsen F M, Knudsen H, Merrison J P, Mitchell R, Paludan K and Poulsen M R 1999 J. Phys. B 32 5825
[14] Straub H C, Lindsay B G, Smith K A and Stebbings R F 1996 J. Chem. Phys. 105 4015
[15] Cooke D A, Murtagh D J, Kövér Á and Laricchia G 2008 Nucl. Instr. Meth. B 266 466
[16] Tóth I, Campeanu R I, Chiş V and Nagy L 2006 Phys. Lett. A 360 131
[17] Moxom J, Ashley P and Laricchia G 1996 Can. J. Phys 74 367
[18] Ashley P, Moxom J and Laricchia G 1996 Phys. Rev. Lett. 77 1250
[19] Campeanu R I, McEachran R P and Stauffer A D 1996 Can. J. Phys 74 544
[20] Ihra W, Macek J H, Mota-Furtado F and O’Mahony P F 1997 Phys. Rev. Lett. 78 4027
[21] Deb N C and Crothers D S F 2002 J. Phys. B 35 L85
[22] Kuo T Y, Sun H L and Huang K N 2003 Phys. Rev. A 67 012705
[23] Overton N, Mills R J and Coleman P G 1993 J. Phys. B 26 3951
[24] Hewitt R N, Noble C J and Bransden B H 1992 J. Phys. B 25 557
[25] Sarkar N K, Basu M and Ghosh A S 1992 Phys. Rev. A 45 6887
[26] Igarashi A and Toshima N 1992 Phys. Lett. A 164 70
[27] Schulz D R and Olson R E 1988 Phys. Rev. A 38 1866
[28] Mandal P, Guha S and Sil N C 1980 Phys. Rev. A 22 2623
[29] Cheng Y and Zhou Y 2007 Phys. Rev. A 76 012704
[30] Murtagh D J, Cooke D A and Laricchia G 2009 Phys. Rev. Lett. 102 133202
[31] Laricchia G, Van Reeth P, Szłuinskiś M and Moxom J 2002 J. Phys. B 35 2525
[32] Marler J P and Surko C M 2005 Phys. Rev. A 72 062713
[33] Charlton M, Clark G, Griffith T C and Heyland G R 1983 J. Phys. B 16 L465
[34] Jin B, Miyamoto S, Sueoka O and Hamada A 1994 At. Coll. Res. Jap. 20 9
[35] Fornari L S, Diana L M and Coleman P G 1983 Phys. Rev. Lett. 51 2276
[36] Diana L M, Coleman P G, Brooks D L, Pendleton P K, Norman D M, Seay B E and Sharma S C 1986 Positron (Electron) - Gas Scattering ed Kauppila W E, Stein T S and Wadehra J M (World Scientific)
[37] Stein T S, Harte M, Jiang J, Kauppila W E, Kwan C K, Li H and Zhou S 1998 Nucl. Instr. Meth. B 143 68
[38] Gilmore S, Blackwood J E and Walters H J R 2004 Nucl. Instr. Meth. B 221 129
[39] McAlinden M and Walters H 1992 Hyperfine Interactions 73 65
[40] Dunlop L J M and Gribakin G F 2006 Nucl. Instr. Meth. B 247 61
[41] Maci P and Barrachina R 2009 Nucl. Instr. Meth. B 267 366
[42] Szłuinskiś M, P Van Reeth and Laricchia G 2002 J. Phys. B 35 4059
[43] Kövér Á, Paludan K and Laricchia G 2001 J. Phys. B 34 L219
[44] Fiol J, Rodriguez V D and Barrachina R O 2001 J. Phys. B 34 933
[45] Fiol J and Olson R E 2002 J. Phys. B 35 1173
[46] Arcidiacono C, Beale J, Pešić Z D, Kövér Á and Laricchia G 2009 J. Phys. B 42 065205
[47] Kövér Á and Laricchia G 1998 Phys. Rev. Lett. 80 5309
[48] Brauner M and Briggs J S 1986 J. Phys. B 19 L325
[49] Arcidiacono C, Kövér Á and Laricchia G 2005 Phys. Rev. Lett. 95 223202
[50] Fiol J, Maci P and Barrachina R O 2009 Nucl. Instr. Meth. B 267 211
[51] Kimura M, Sueoka O, Hamada A and Itikawa Y 2007 Adv. Chem. Phys. ed Prigogine S A R I pp 537–622
[52] Rudd M E, Goffe T V, DuBois R D and Toburen L H 1985 Phys. Rev. A. 31 492
[53] Ning C, Hajgat B, Huang Y, Zhang S, Liu K, Luo Z, Knippenberg S, Deng J and Deleuze M 2008 Chem. Phys. 343 19
[54] Armitage S, Leslie D E, Garner A J and Laricchia G 2002 Phys. Rev. Lett. 89 173402
[55] Armitage S, Leslie D, Beale J and Laricchia G 2006 Nucl. Instr. Meth. B 247 98
[56] Biswas P K and Adhikari S K 1999 Phys. Rev. A 59 363
[57] Sarkadi L 2003 Phys. Rev. A 68 032706
[58] Blackwood J E, Campbell C P, McAlinden M T and Walters H R J 1999 Phys. Rev. A 60 4454
[59] Ray H 2002 J. Phys. B 35 3365
[60] Starrett C, McAlinden M T and Walters H R J 2005 Phys. Rev. A 72 012508
[61] Walters H, Starrett C and McAlinden M T 2006 Nucl. Instr. Meth. B 247 111
[62] Starrett C, McAlinden M T and Walters H R J 2008 Phys. Rev. A 77 042505
[63] Brawley S, Beale J, Armitage S, Leslie D E, Kövér Á and Laricchia G 2008 Nucl. Instr. Meth. B 266 497
[64] Williams A I, Kövér Á, Murtagh D J and Laricchia G 2009 Journal of Physics: Conference Series in press