

Resonant Scattering of Positronium in Collision with CO₂

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The total cross sections of positronium (Ps) scattering from a carbon-dioxide molecule have been measured over the range (7–400) eV incident-Ps energy. For the first time in Ps collisions, a resonantlike structure is observed. For the present target, it occurs around 9.5 eV followed by a broader peak at ~60 eV. Following Brawley *et al.* [*Science* **330**, 789 (2010)] who have observed similarities between the total cross sections of positronium and of electrons incident upon a given target at the same velocity, a corresponding comparison is made for CO₂. The comparison suggests that the former peak corresponds to the well-known ${}^2\Pi_u$ shape resonance which occurs for electrons at an incident velocity of 0.5 a.u. Further features are discussed and theoretical input is sought.

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Positronium (Ps) is the hydrogenlike bound state of an electron and its antimatter partner, the positron. It has half the reduced mass of hydrogen, and thus twice the Bohr radius and half the binding energy, $6.8/n^2$ where n is the principal quantum number. The relative orientation of the spins of its constituents yields singlet (or para) and triplet (or ortho) states. The 1^1S_0 state has a lifetime of 125 ps (annihilating into predominantly 2 γ rays) while 1^3S_1 has a lifetime 142 ns (annihilating predominantly into 3 γ rays).

It is the relatively long lifetime of ortho-Ps which enabled its first observation in studies of positron annihilation in gases [1]. Since then, Ps has been found to be produced abundantly in positron collisions with matter. So significant is its probability of formation that 80% of all γ rays produced in positron-emission tomography [2], and around 95% of the e^+e^- annihilation emissions from the Galactic center [3], arise from Ps decay. Further annihilation studies, namely, angular correlation of the annihilation radiation and time-resolved Doppler broadening of the γ -ray energy, have facilitated the extraction of Ps momentum-transfer cross sections below 0.3 and 3 eV, respectively (e.g., [4,5]). At higher energies, Ps beams (e.g., [6–8]) have enabled the measurement of total [7,9–11] and fragmentation [12,13] cross sections in the range (7–250) eV with spreads around (1–2) eV [14]. Theoretically, a full description of Ps + atom (molecule) scattering is complicated by the internal degrees of freedom of both the target and the projectile and the two-center nature of exchange which is expected to be particularly important for Ps due to its neutrality and, indeed, the coincidence of its centers of mass and charge. Yet, a number of targets have been scrutinized using a variety of approaches, notably including elaborate close-coupling methods for H, He, and Li (e.g., [15–17] and references therein).

Recently, a striking similarity has been uncovered between the total cross sections for electrons (Q_T^-) and those for equivelocity Ps (Q_T^{Ps}) [18] incident upon a wide range of targets (He, Ne, Ar, Kr, Xe, H₂, H₂O, N₂, O₂, and SF₆)

over a broad range of velocities [0.5–2.2 a.u., corresponding to Ps incident energies (7–131) eV]. In that study, the total cross sections for the two projectiles were found to remain of comparable magnitude even in the proximity of prominent structures in those for electrons, such as the Ramsauer-Townsend minima and resonances. Neither feature was, however, established for Ps. Note that, henceforth in this Letter, velocities shall be given in atomic units (1 a.u. = 2.2×10^6 m s⁻¹) and energies in eV.

Even though the observation is not fully understood, Brawley *et al.* [18] drew attention to a classical trajectory Monte Carlo simulation of Ps fragmentation on He [19] which indicated that the electron in Ps may be on average closer to the target during a collision. Of course, exchange is completely absent in this type of calculation and any inference must be tempered accordingly. The same authors [18] also noted, with respect to an impulse approximation for the same process which treated the collision of Ps as a coherent combination of electron and positron scattering [20], that their findings might imply a negligible positron scattering amplitude in comparison to that of the electron.

While theoretical guidance remains very much needed, the current experimental investigation was undertaken to further probe the similarity between the two equivelocity projectiles. This was done by measuring Q_T^{Ps} for CO₂ for which Q_T^- displays a significant structure at a velocity, and of a width, accessible with the present Ps beam. CO₂ is a well-studied, linear, triatomic molecule of ${}^1\Sigma_g^+$ symmetry in its ground state [21]. It has three normal modes of vibration: a symmetric stretching mode ($n, 0, 0$) with Σ_g symmetry, a bending mode ($0, n, 0$) of Π_u symmetry, and an antisymmetric stretching mode ($0, 0, n$) of Σ_u symmetry, the latter two inducing a dipole moment in the molecule. The structure in Q_T^- , occurring at 3.8 eV ($\equiv 0.53$ a.u.), is a shape resonance in which the projectile electron attaches temporarily to the $2\pi_u$ orbital (e.g., [22,23]). Generally, the decay of a resonance by spontaneous electron emission leaves the target in a vibrationally

excited state [24]. For the specific case under consideration, the attachment is known to increase the probability of excitation to the accidentally degenerate (100) symmetric and (020) bending modes—the so-called Fermi dyad [22].

Figure 1 shows a schematic diagram of the Ps beam line at University College London (UCL). Ps atoms are produced in cell 1 where an atomic or molecular gas (A) is introduced to neutralize the beam of positrons (e^+) according to the charge-exchange reaction $e^+ + A \rightarrow \text{Ps} + A^+$ [25]. For this process, generally the differential cross section ($dQ_{\text{Ps}}/d\Omega$) becomes increasingly forward peaked starting from a few eV above threshold ($E_{\text{th}} = E_I - 6.8/n^2$, E_I being the first ionization energy of the target A). In this experiment, positrons are derived from the β decay of ^{22}Na , moderated by a thin layer of solid Kr frozen directly onto the source capsule, accelerated from it by an applied positive potential with respect to the chamber ground, and radially confined by an axial magnetic field of $\sim 10^{-2}$ T. The Wien filter serves to remove a line of sight between the source and the detectors, and to reduce the energy spread of the positron beam to (1.7 ± 0.1) eV FWHM. In this work, the neutralizer in cell 1 was H_2 gas which has been found to be an efficient $e^+ \rightarrow \text{Ps}$ converter particularly from $\sim(10\text{--}100)$ eV [9]. The Ps beam kinetic energy (E_{Ps}) is tunable via that of the positron (E_+) according to $E_{\text{Ps}} = E_+ - E_{\text{th}}$, as verified by time-of-flight measurements which also allow the dominant n state of Ps to be determined [26–28]. Positronium-beam production from H_2 is known to produce atoms predominantly in the ground state [28]. In the present work, great care was taken to ensure the stability of E_+ (and thus E_{Ps}) to within 0.1 eV.

The Ps production efficiency in cell 1 can be expressed as

$$\epsilon \propto (1 - e^{-\rho_1 \ell_1^+ Q_T^+}) \left(\frac{1}{Q_T^+} \int_0^{\theta'} \frac{dQ_{\text{Ps}}}{d\Omega} \sin\theta d\theta \right) e^{-\rho_1 \ell_1^{\text{Ps}} Q_T^{\text{Ps}}}, \quad (1)$$

where ρ_1 is the number density of the gas in cell 1, ℓ_1 the effective length of this cell, and Q_T the total cross section—the superscripts + and Ps on the latter two parameters refer to positrons and Ps, respectively; the beam half-angle θ' is typically around $(1^\circ\text{--}1.5^\circ)$. Positrons

transmitted through cell 1 are reflected electrostatically at an unannealed W grid (CR1). The neutral Ps beam—which has an energy spread very similar to that of the positron beam [14]—continues to cell 2, which contains CO_2 gas at a pressure in the range 0.2–0.3 Pa. Hits of Ps atoms on a pair of channel-electron-multiplier arrays (CEMA) are monitored in coincidence with an associated γ -ray photon detected by a CsI or NaI γ -ray counter. At the speeds appropriate for investigating atomic and molecular phenomena, para-Ps travels less than 1 mm in its lifetime and so it is ortho-Ps which has a non-negligible probability of traveling from the production cell through the interaction region to the detectors. The Ps beam intensity is highly dependent on its kinetic energy via the dependence of the relevant cross sections [as implied by Eq. (1)] and its survival probability. Thus Q_T^{Ps} was measured using two methods.

Direct.—Between 7 and 150 eV ($\equiv 0.5\text{--}2.4$ a.u.), Q_T^{Ps} was determined by measuring the incident (I_0) and transmitted (I) intensities of the Ps beam through cell 2 and applying the Beer-Lambert law:

$$Q_T^{\text{Ps}} = -\frac{1}{\rho_2 \ell_2} \ln\left(\frac{I}{I_0}\right), \quad (2)$$

where ρ_2 is the gas density in cell 2 and ℓ_2 its corresponding effective length.

Indirect.—At the highest energy investigated (400 eV), the Ps beam intensity produced through H_2 is inadequately low for method 1. Thus Q_T^{Ps} was extracted by least-square fitting Eq. (1) to measurements of $\epsilon \propto \frac{N_{\text{Ps}}}{N_+}$ (i.e., the measured yield of Ps atoms per positron) as a function of ρ_1 at a given E_{Ps} [29].

The present measurements for Q_T^{Ps} are plotted in Fig. 2 where they are compared with those for equivelocity electrons and positrons (Q_T^- [30,31] and Q_T^+ [30,32], respectively)—in the following discussion, energies refer to Ps projectiles only. With increasing velocity, the cross section increases rapidly from $(13.3 \pm 2.0) \times 10^{-20}$ m² at 0.51 a.u. (7.0 eV) to $(22.0 \pm 1.2) \times 10^{-20}$ m² at 0.60 a.u. (9.7 eV). A quick drop to $(17.7 \pm 0.5) \times 10^{-20}$ m² at 0.75 a.u. (15.3 eV) marks out a distinct peak of

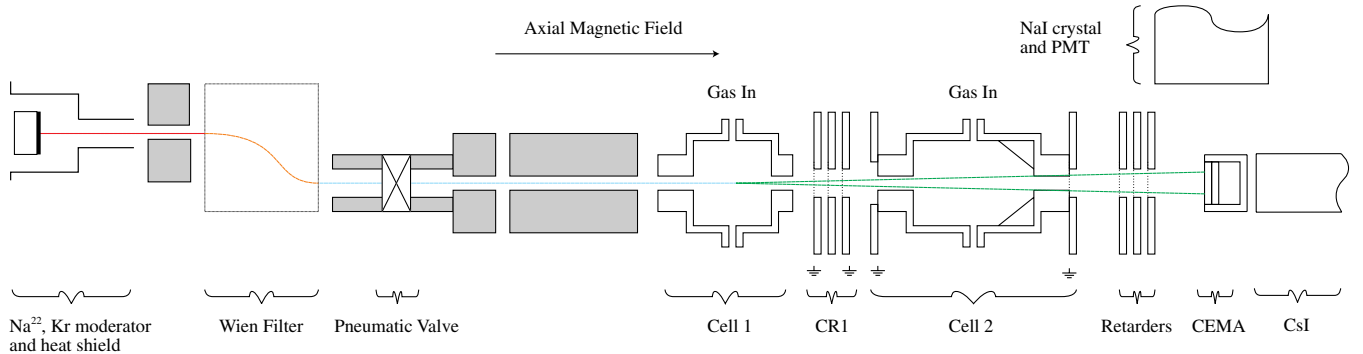


FIG. 1 (color online). Schematic of the Ps, positron, and electron beam line at UCL (in grey are various collimators).

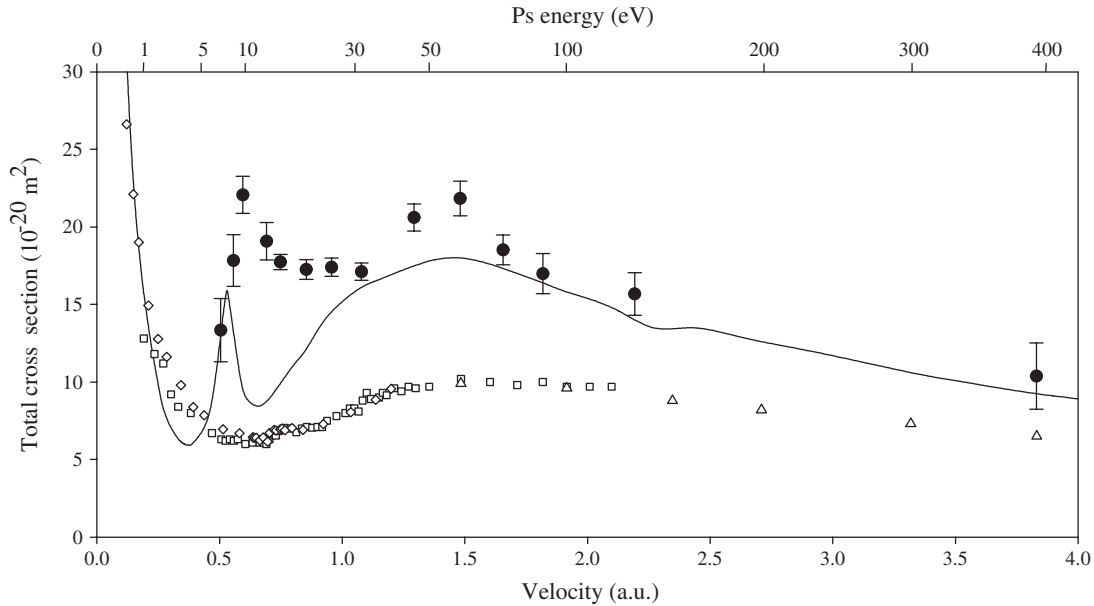


FIG. 2. Comparison of Ps, positron, and electron total cross-section measurements, Q_T^{Ps} , Q_T^+ , and Q_T^- , respectively, for collisions with CO_2 . ●, present Q_T^{Ps} ; solid line, average Q_T^- [21]; ◇, Q_T^+ [38]; □, Q_T^+ [30]; △, Q_T^+ [32]. Error bars represent the standard error of the mean.

FWHM ~ 5 eV, followed by a flat region up to around 1 a.u. (27 eV). A broad peak is observed from 1 to 2.2 a.u. (131 eV) which reaches a similar magnitude as the first. Q_T^{Ps} decreases approximately by a factor of 2 at the highest velocity investigated (3.8 a.u. \equiv 400 eV). While little correspondence is observed with Q_T^+ , Q_T^{Ps} shows a broad similarity with Q_T^- in shape over the entire velocity range, and also in magnitude above 1.5 a.u. The resemblance extends to the velocity region where the $^2\Pi_u$ resonance occurs in Q_T^- at 0.5 a.u. although the peak in Q_T^{Ps} occurs at a somewhat higher velocity (0.6 a.u.), possibly due to a weaker target polarization (e.g., [33]) by the neutral Ps in comparison with an electron projectile as well as perhaps contact-potential effects.

It is tempting to speculate on the possible physical origins of the excess of Q_T^{Ps} over Q_T^- around 0.9 and 1.5 a.u. Thus we note that, while not apparent in Q_T^- , two additional shape resonances of Σ_g and Σ_u symmetry emerge in the differential cross section for vibrational excitation of CO_2 by electrons at impact velocities of 0.86 and 1.48 a.u. [24,34,35]. Additionally, we note that, while uncertainties remain both on the threshold energies and the assignments of the electronically excited states of CO_2 [21], its differential cross sections by electron impact are rich in structure in the region from 0.8 to 1 a.u., including triplet-state excitations [36,37] to which Ps might be more sensitive, given the mediating role of exchange in such excitations and its presumed importance in Ps interactions with matter.

In summary, a peak has been observed in the total cross section for Ps + CO_2 scattering at a projectile velocity near

that of the well-known $^2\Pi_u$ shape resonance in the corresponding electron cross section. Observed enhancements of Q_T^{Ps} over Q_T^- might signal a comparatively greater sensitivity of Ps to additional shape resonances (i.e., Σ_g and Σ_u) and/or exchange-mediated electronic excitation. While the present work strengthens the finding of Brawley *et al.* [18] regarding the similarity in the overall scattering probabilities of Ps and equivelocity electron projectiles, the basic physical mechanism responsible for the phenomenon by which, at least over the velocity range 0.5–4 a.u., Ps scatters like a quasifree electron remains uncertain. Further investigations are eagerly anticipated.

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