Energy-Sharing Asymmetries in Ionization by Positron Impact

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(Received 13 May 2005; published 23 November 2005)

The triply differential cross section of molecular hydrogen for ionization by 50 eV positrons has been determined, for the first time, for both the ejected electron in coincidence with the remnant ion and for the scattered projectile. Asymmetries in the energy sharing between the two light particles in the final state are observed, with the electron spectrum being shifted to significantly lower (and the scattered positron to correspondingly higher) energies than expected. A similar shape is observed in the case of the ejected electron spectrum from a helium target at the same excess energy.

DOI: 10.1103/PhysRevLett.95.223202 PACS numbers: 34.85.+x

The correlated dynamics of few interacting particles is a fundamental physics problem that may be exemplified through the process of ionization. Despite the tremendous progress in its theoretical description during the past decade or so (e.g., [1,2]), concomitant experimental investigations remain essential in assessing the accuracy of the various approaches and in guiding further developments. In this respect, particularly sensitive are studies in which there are two or more *light* particles (e.g., an electron and a positron) in the final state and which yield cross sections which are differential in the energy and/or angular distribution of the ejected electron(s) and/or scattered projectile. The most stringent among these is the triply differential cross section (TDCS) in which all the kinematic parameters are determined. A significant body of data has been gained using the $(e, 2e)$ method (e.g., [3]) and, more recently, the COLTRIM technique (e.g., [4]), which has been applied to electron, photon, proton, and ion impact. Differential studies with positrons, mainly confined to doubly differential investigations (e.g., [5–7]), remain scarce but are desirable both intrinsically and for comparison with equivelocity electrons or protons to probe the role of the projectile charge or mass on the collision dynamics (e.g., [8,9]). In this Letter, we report experimental TDCS results for positron impact ionization of simple molecular and atomic targets that reveal major discrepancies with current quantum-mechanical treatments and should thus provide new insights into the understanding of three-body correlated dynamics.

Over the past decade, sophisticated distorted wave calculations have been developed based on the 3-Coulomb-wave final-state wave function (3C) of Brauner *et al.* [10] which approximates the strictly inseparable many-body system in terms of pairs of interacting particles. At lower energies, the use of the eikonal approximation for the initial state has been found to improve agreement with experiments (e.g., [11]). While such methods have been successful with a variety of projectiles and over a wide energy range, nonperturbative approaches remain superior at lower energies. Particularly noteworthy in this respect are the exterior complex scaling (ECS) method (e.g., [2]), which yielded the first accurate TDCS for e^--H ionization for the case of equal energy-sharing kinematics [12], and close-coupling techniques which have been used for electron and photon collisions with various atoms and ions (e.g., [13]). In the case of integrated cross sections, also for collisions with positrons (e.g., [14,15]) and positronium (e.g., [16]).

A special case of ionization is electron capture, where the ejected electron is captured by the projectile to a bound or low-lying continuum state [17,18]. This latter process, often referred to as electron capture to the continuum (ECC), arises from the dominance of the final-state Coulomb attraction between the scattered projectile and the ionized electron. It is well known in ion-atom ionizing collisions and easily observable in the energy spectrum of electrons ejected around the direction of the scattered ion which, owing to its mass, suffers little deflection through the collision [19]. Positrons, on the other hand, are light and easily deflected. For this reason, the observation of ECC with these projectiles had to await the first kinematically complete experiment where increased sensitivity was achieved by detecting the projectile scattered near 0° in coincidence with the electron ejected in the same direction [20]. In that study, the TDCS was determined for positrons at an incident energy of 100 eV in collision with a H_2 target, namely $e^+(100\text{ eV}) + H_2 \rightarrow e^+(\theta \sim 0°) + e^-(\theta \sim 0°, E_-) + {H_2}^+$. The ECC process was manifest by a small peak at half of the residual kinetic energy ($E_r/2$ where $E_r = E_i - I$, E_i being the positron incident energy and I the target ionization energy). The results were well described by the calculations [21,22] employing $3C$ wave functions for the final state. More recently, however, in an experiment at 50 eV incident energy, a significant shift of almost 2.5 eV from $E_r/2 = 17.3$ eV has been observed in the electron TDCS peak towards lower energies [23]. In comparison with the calculations of Fiol *et al.* [22], convoluted with the angular and energy resolutions, the experimental TDCS was shifted by around 1.6 eV. Although in [23], an error in the energy calibration was deemed unlikely, it could not be entirely excluded and thus the experimental results have remained preliminary until now. Kövér *et al.* [23] had conjectured that, if genuine, a possible cause of this energy shift could be a doubly inelastic process, e.g., ionization simultaneous to vibrational excitation or dissociation of the remnant ion.

Fiol and Olson [24] carried out calculations for impact ionization of H_2 using both the classical trajectory Monte Carlo (CTMC) method and the perturbative quantum-mechanical approach using $3C$ final-state wave function (CDW) at 50 and 100 eV positron incident energy. They found discrepancies between the results from the two theoretical methods and no consistent description of the experimental data, with CDW giving better agreement at 100 eV while the CTMC model better describes the experiment at 50 eV, where a strong correlation between the momenta of the positron and the recoil ion was noticed.

In this study, (i) we resolve the main uncertainties of the study of Kövér *et al.* [23], by calibrating the absolute energy through the identification of the threshold for positronium formation and by checking the remnant hydrogen ion for possible dissociation; (ii) we determine, for the first time, the energy spectrum of the positrons scattered from molecular hydrogen, and (iii) we measure, also for the first time, the energies of electrons ejected by positron impact ionization of helium at the same excess energy as in the molecular hydrogen study.

The experiment has been carried out at University College London using the apparatus previously described [5,25]. Briefly, positrons are transported through an electrostatic system from the moderator to a crossed gas jet. A ^{22}Na source (activity $\sim 3 \times 10^8$ Bq), in conjunction with an annealed tungsten mesh moderator, provides a positron beam intensity of $\sim 10^3\ s^{-1}$. The energy distribution of the particles ejected (or scattered) at around 0° has been measured using a single channel electron multiplier (CEM) at the end of a tandem parallel-plate energy analyzer (PPA) [26], as shown in Fig. 1. The particles scattered (or ejected) at the same angle have been detected in delayed coincidence with an assembly of microchannel plates (MCP) fixed within the first stage of the PPA. Time spectra have been recorded both with and without target gas. The overall measuring time at each energy was around 10^5 s. After normalizing the time spectra for the number of positrons incident upon the MCP, the gas pressure, and the possible variation in the detection efficiency, the difference between the normalized gas-on and gas-off spectra has been determined and the total coincidence signal calculated. The absolute energy of the beam has been obtained by determining the positronium formation threshold (E_{Ps}) in He. This has been done by measuring the ion yield versus moderator voltage, V_m, yielding $E_i = eV_m + (2.24 \pm 0.36)$ eV in agreement with the previous determination [23]. During this measurement, the positron beam was stopped while pulses ± 50 V high and 2 μs long, from

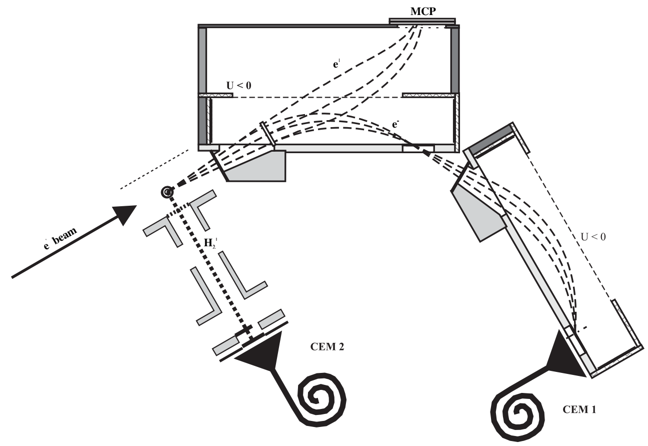

FIG. 1. Schematic diagram of the interaction region comprising the gas jet, the parallel-plate analyzer, and the ion extractor.

a generator operating at ~ 10 kHz, were applied to the ion-extractor plates in Fig. 1.

To investigate the possibility of dissociation, the charge-to-mass ratio (Q/M) of the ion in the final state was measured. For this purpose, a triple coincidence system has been set up between the ejected electron, the remnant ion and the scattered positron. Detection of an electron of a given energy triggered the application of the voltage pulses to the capacitor plates to extract possible ions present in the scattering region. In this measurement, D_2 was used as the target gas to increase the lifetime of the ion in the extraction region and to distinguish it from possible contributions from background gases. The correlated detection of an electron-ion pair has then been used to initiate a second measuring sequence stopped by a positron. From the time-of-flight spectra obtained from the delayed coincidence between CEM1 and CEM2, the ion Q/M has been determined.

In these measurements, no D^+ has been observed but this is not a conclusive proof that dissociative ionization is not responsible for the shift, as the extraction efficiency of the dissociation products may be significantly suppressed by their relatively large speed. However, as shown in Fig. 2(a), the energy dependence of the triply coincident $D_2{}^+$ signal has been found to be the same as that observed in [23]. It is this observation that excludes the involvement of dissociative ionization. In the figure, the triply differential electron spectrum for D_2 has been normalized to the theoretical calculation of Fiol *et al.* [22], as it was done in [23]. The most conspicuous feature of the comparison with the theory is the displacement of the experimental distribution towards lower energies: the theory of [22] peaks at 16.5 eV, while both sets of experimental data rise to a maximum at ~ 15 eV. Both theoretical and experimental data decrease with a similar slope above their respective peaks. Also shown in the figure are the CTMC calculations of Fiol and Olson [24]. As mentioned earlier, this approach failed to describe the 100 eV data of Kövér and Laricchia

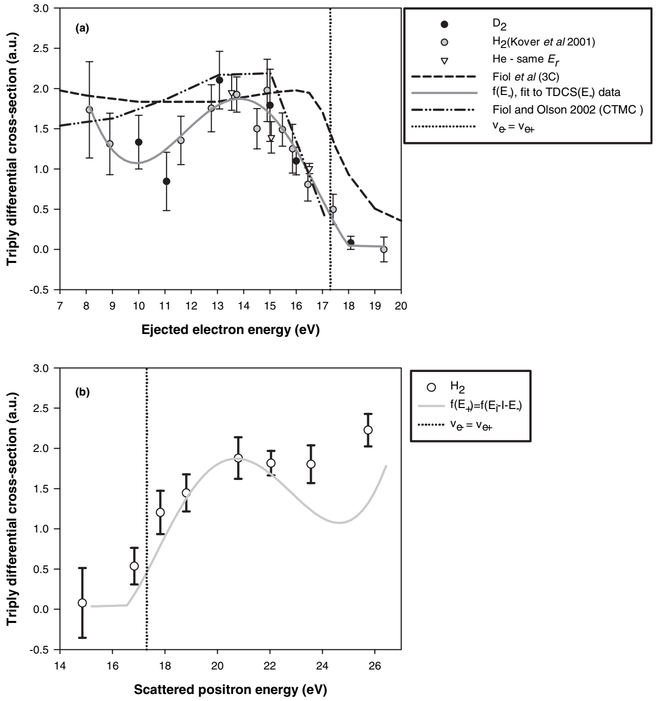

FIG. 2. Experimental and theoretical results for the triply differential ionization cross sections for ejected electrons (a) and scattered positrons (b) in 50 eV positron collision with molecular hydrogen. Data for ejected electrons from helium at the same residual energy are also presented. The gray lines are guides to the eye only.

[20], but it reproduces the main features of the 50 eV data of Kövér *et al.* [23] and of the present data.

The measured energy spectrum of the scattered positrons is shown in Fig. 2(b). The positron data have been normalized around the maximum to the peak value of the electron data. A close correspondence is observed between the energy distributions of the ejected electrons, TDCS (E_-), and that of scattered positrons, TDCS(E_+), with $E_+ = E_i - E_- - I$, as expected from energy conservation. This establishes that the shift does not arise from an energy loss to the target as, for example, through molecular excitations.

Finally, the TDCS (E_-) obtained by positron impact ionization of helium at the same residual energy, $E_r = 34.6$ eV, can also be seen in Fig. 2(a) to follow the same shape as for hydrogen, implying that the significant parameter for the shift is the final-state kinetic energy.

Recently, a study has been performed with H^+ incident on H_2 and He resulting in electrons being ejected with velocities comparable with those of the present study. At 10 and 20 keV impact energy, the ECC cusp formation around 0° has been found to be shifted below its standard position around the projectile velocity [27]. CTMC calculations by the same authors indicate that the long-range residual interaction of the electron with the remnant target ion is responsible for the shifts that manifest the pull of the target on the ejected electron.

Although we note that recent experimental and theoretical results [28] do not support the findings of [27], the interpretation of Shah *et al.* [27] might be compatible with the findings by Sarkadi [29], who investigated the fragmentation of positronium (Ps) in Ps-He collision also with the CTMC method and compared the results with the experimental data of Armitage *et al.* [30]. While good agreement has been found with the shape of the measured longitudinal energy distribution of the positron, he has predicted that the maximum of the electron peak should be shifted to lower energies due to the dynamical polarization of the target. While this prediction awaits experimental verification, the effect may reasonably be conjectured to be more pronounced in the case of a charged final state for the target.

Alternatively, it has been suggested that competition from the Ps formation channel might strongly influence the shape of the distributions at the lower energies [31]. Clearly, further work is needed in order to understand the observations.

In conclusion, the triply differential ionization cross section of molecular hydrogen has been determined for both ejected electrons and scattered positrons. Investigations have also been performed with a helium target. An unexpected asymmetry in the energy sharing between the two light particles in the final state has been observed around half of the residual energy: the electron spectrum is shifted to lower energies than predicted by perturbative calculations by around 1.5 eV while the positron distribution exhibits a shift of similar magnitude, but opposite sign from the equal energy-sharing value. From these studies, a significant factor in the shift appears to be the final-state kinetic energy and, in particular, perhaps, the low velocities of the light particles in the final state.

At present, the data have no consistent description by quantum-mechanical theoretical treatments and, in this respect, might be a suitable testing ground for recently developed *ab initio* approaches.

We wish to thank Laszlo Sarkadi and Jonathan Tennyson for helpful comments on this Letter. This work has been supported by the Engineering and Physical Science Research Council UK (Grant No. GR/S16041/01), the Royal Society, the Hungarian Scientific Research Found (OTKA No. T037203), and the European Union (HPRN-CT-2002-00179 EPIC). Akos Kover wishes to thank UCL for the excellent working conditions and hospitality.

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