Title

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Journal
Physical Review Letters, 111(1)

ISSN
0031-9007

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Publication Date
2013-07-05

DOI
10.1103/physrevlett.111.013003

Peer reviewed

Ejection of quasi-free electron pairs from the helium atom ground state by single photon absorption

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(Dated: August 1, 2012)

We investigate single photon double ionization (PDI) of helium at photon energies of 440 and 800 eV. We observe doubly charged ions with close to zero momentum corresponding to electrons emitted back-to-back with equal energy. These slow ions are the unique fingerprint of an elusive quasi-free PDI mechanism predicted by Amusia *et al.* nearly four decades years ago [J. Phys. B **8,** 1248, (1975)] . It results from the non-dipole part of the electromagnetic interaction. Our experimental data are in excellent agreement with calculations performed using the convergent close coupling and time dependent close coupling methods.

INTRODUCTION

The non-relativistic Hamiltonian of electromagnetic interaction is a one-body operator and thus it couples one photon to just one electron. Simultaneous emission of two electrons following absorption of a single photon is only possible due to electron-electron correlation. Single photon double ionization (PDI) of helium became the prototype process to investigate such correlations. The two specific correlation mechanisms, the shake-off (SO) and electron knock-off (KO) processes (the latter is also known as Two-Step-1 or TS1 [1]), are well established today to facilitate the ejection of two electrons by a single photon. In both cases, the photon couples primarily to the dipole formed by one electron and the nucleus. The two electrons interact with each other, either before (SO) or after (TS1) the photo-absorption takes place. At photon energies far from the threshold, the electron emission patterns characteristic for each of the mechanisms can be separated in fully differential cross sections (FDCS) [2]. The energy sharing between the two emitted electrons exhibits a U-shape [3, 4], which becomes deeper and deeper with increasing photon energies [5, 6]. The TS1 probability decreases with the increasing photon energy while the SO probability increases and finally saturates at the (non-relativistic) shake-off limit [7–10]. A common fingerprint of both mechanisms is a large momentum of the doubly charged ion and an almost dipolar angular distribution of the ion momentum with respect to the linear photon polarization axis. These two characteristic features are signatures of the initial coupling of the photon to the dipole formed by the primary electron and the nucleus [2, 8, 11–13].

Nearly four decades ago, Amusia et al. [14] predicted a third, so called quasi-free mechanism (QFM) of PDI. Its characteristic fingerprint is the ejection of two electrons back-to-back with similar energy while the nucleus is only a spectator [15] remaining almost at rest [14, 16, 17]. They argued that this mechanism ejects electrons mainly from the part of the initial state wave function at the electron-electron cusp, i.e. where both electrons are spatially close together. It is a contribution to the quadrupole part of PDI since emitting electrons back-to-back with equal energies is forbidden by the kinematic selection rule in a dipole transition [18]. This region of momentum space is, however, allowed in a quadrupole transition.

The QFM transition amplitude is extremely small, which is why it could not be verified experimentally so far. For instance, for a photon energy of 800 eV and the hydrogen Bohr radius, the non-dipole transition amounts to 1 % of the total PDI cross section only. From this quadrupole part, the QFM is only a small part; it can be estimated to 0.1 % of the total PDI cross section.

PDI of He in the low photon energy regime, where quadrupole transitions are negligible, has been investigated experimentally and theoretically in great detail in the past [19–21] Experimental FDCS are available up to 530 eV [2, 11, 22–24]. However, very little is known about PDI at higher photon energies. The coupling of higher orders of angular momentum of the incoming light, such as the electric quadrupole term, to the two electrons in the atom has only been addressed theoretically [25–30]. So far, no experiments have studied non-dipole effects in PDI due to extremely small cross sections. As the QFM has an even smaller cross section and requires, in addition, the coincident detection of two electrons emitted back-to-back with equal energy, it escaped experimental observation until now.

In the present Letter, we report on the observation of low momentum doubly charged ions for PDI of helium at 800 eV and thus present direct support for the QFM mechanism. This is also consistent with the accompanying *ab initio* non-perturbative calculations, performed using the convergent close coupling (CCC) and the time dependent close coupling (TDCC) methods, which reproduce our experimental findings.

The experiments were carried out with linear polarized light at 800 eV during two-bunch mode at beamline 11.0.2.1 of the Advanced Light Source (ALS) of LBNL. The technical realization of this project is very challenging. The cross section for quadrupole transitions amounts to $\simeq 2 \cdot 10^{-25}\text{cm}^2$ only. At the same time, a coincidence measurement of the momentum vectors of at least one electron and the recoiling doubly charged ion is necessary to cleanly single out the scarce PDI events (total cross section $\simeq 2 \cdot 10^{-23}\text{cm}^2$). The solution to this problem is the application of the COLd Target Recoil Ion Momentum Spectroscopy (COLTRIMS) method which is able to detect the 3D-momenta of the outgoing particles within the 4π solid angle in coincidence [31–33]. It is sufficient to measure one electron and the recoiling ion simultaneously. Momentum conservation is used to calculate the momentum vector of the second electron and the energy conservation is exploited to eliminate background in the offline analysis. In brief, the target is prepared in a supersonic gas jet (30 μm nozzle and 20 bar driving pressure) along the y-direction and intersected with the photon beam (propagating along the x-axis) in a weak electric field inside the momentum spectrometer. The field (16 V/cm) is just strong enough to separate the fragments by their charge and guide them towards two large position and time sensitive multi-channel plate (MCP) detectors with delay line readout [34, 35]; 80 mm diameter for the ions and 120 mm for the electrons. A magnetic field (23 Gauss) in parallel is used to prevent high energetic electrons from leaving the spectrometer by forcing these particles to gyrate towards the detector. With the knowledge of the dimensions of the spectrometer, the field strengths, the position of impact and time of flight of the particles, the 3D momentum vector of each particle can be deduced. To increase the ion momentum resolution and to compensate for the finite interaction volume (see [36] for general information about the time/space focusing), we employed an electrostatic lens and a 120 cm long drift region for focusing. This resulted in an ion momentum resolution of $\simeq$0.15 a.u. in the light polarization direction (z), which is also the time-of-flight-direction, and $\simeq$0.25 a.u. in the transverse direction. The axes layout is shown in Figure 1(a). The photon beam had a small contamination of lower harmonics. Due to an increase of the PDI cross section with decreasing photon energy, this small contamination leads to a non negligible amount of low momentum doubly charged ions. Our coincidence experiment however provides for each registered event the sum energy of both electrons. This has been used to identify the contamination of the ion signal by low energy photons and to unambiguously select events which resulted from the absorption of one 800 eV photon. This also discriminates against events from compton scattering, which also would produce low momentum ions [8]

In Figure 1(b) we present the momentum distribution of the doubly charged helium ions after PDI with linear polarized light of 800 eV. The solid circular line represents the maximum possible momentum an ion could receive in a double ionization ($\text{p}_{\text{max}} = 2\sqrt{(\text{E}_\gamma - 79\text{eV})/2}$, where 79 eV represents the PDI threshold). This is the case, when both electrons have half the excess energy and are emitted in the same direction. Nevertheless the vast majority of events are located close to the surface of a sphere in the momentum space with a radius of $\text{p}_{\text{single}} = \sqrt{\text{E}_\gamma - 79\text{ eV}}$ which corresponds to the recoil momentum of one electron that takes away all the available energy. These ions show a close to dipolar angular distribution. This is the characteristic pattern observed in all previous experiments [2, 12, 13]. It is created by both the SO and TS1 process and dominated by the dipole transition by far. In the center of the sphere, close to the momentum zero, we expect the events from the QFM. To make those events more visible, we plot the two momentum components in the xy-plane perpendicular to the polarization direction (Figure 1c-h). We define the momentum width out of this plane (z-direction) that we are going to select as $|\Delta p_z/p_{\max}|$ and show cuts corresponding to 7, 15 and 25 % of the maximum momentum in Figure 1c-d, e-f and g-h, respectively. This allows us to exploit the selection rules for the dipole transitions which forbid the emission of both electrons in the xy plane, irrespective of the energy sharing [18]. The outer ring of the pattern corresponds to transitions with maximum unequal electron energy sharing. In the center of this ring we find ions almost at rest, their relative contribution increases with photon energy; compare the left column (440 eV) with the right column (800 eV). As outlined above, we have performed a kinetically complete experiment, i.e. we obtained the momentum vectors of all particles. This allows for a full control of the experiment and a highly efficient suppression of all background. We therefore can be certain that the few events at small momentum are experimentally significant and definitely not background. A projection from (Figure 1g-h) with a width of 25 % (indicated by the yellow bars) is shown in (Figure 1i-j). Also here a peak around zero momentum is clearly visible for 800, but not for 440 eV.

The present experimental findings are supported by convergent close coupling (CCC) calculations [19] and time dependent close coupling (TDCC) theory [15] shown in Figure 2 for 440 eV (left) and 800 eV (right) photon energies and a cut in $p_z = \pm 15\%$. These calculations con-

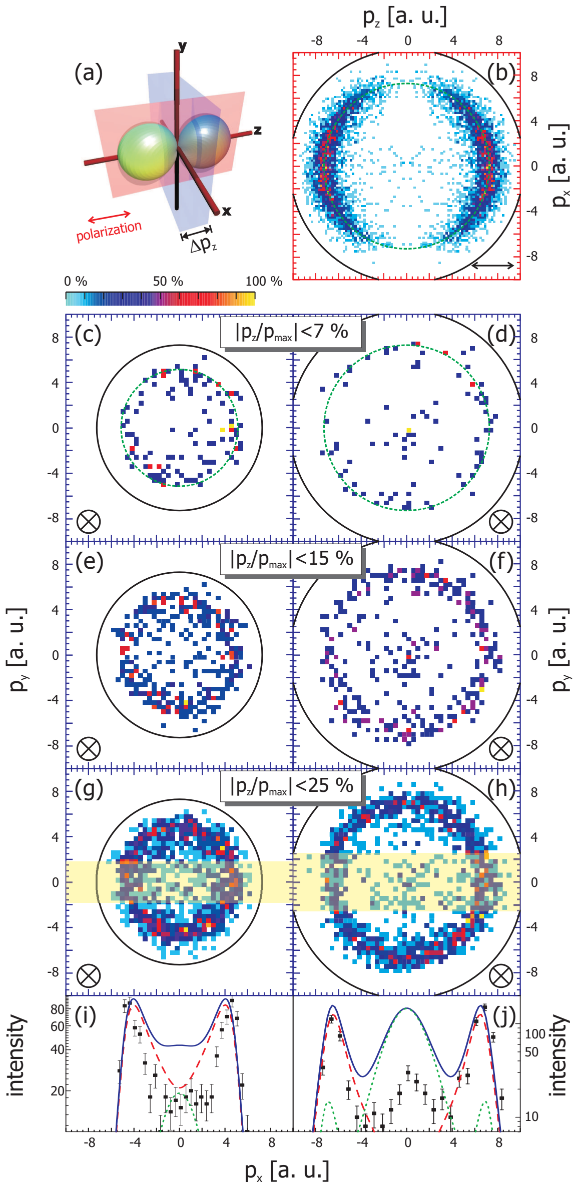

FIG. 1: (a) Sketch of the dipole structure, different planes and coordinates: light propagation x, light polarization z, gas jet direction y. (b) Slice ($|p_y| < 0.73 a.u.$) of the ion momentum distribution in the x,z-plane for $\hbar\nu$=800 eV. (c-h) Ion momentum distribution perpendicular to the light polarization vector for $\hbar\nu$=440 eV (left column) and $\hbar\nu$=800 eV (right column), with different cut widths along the momentum component parallel to the polarization vector of the doubly charged ion at a given photon energy (p_z) (c,d) $|p_z/p_{max}| \le 7\%$, (e,f) $|p_z/p_{max}| \le 15\%$, (g,h) $|p_z/p_{max}| \le 25\%$. The solid circular line represents the maximum possible momentum (p_{max}). The dashed green line in b-d represents (p_{single}), the case if one electron receives all momentum. (i,j) show a momentum cut of (g,h) $|p_y/p_{max}| \le 25\%$ indicated by the yellow bars. The various lines show TDCC calculations for dipole (dashed and red), quadrupole (dotted and green) as well as the coherent sum of dipole and quadrupole (blue and solid).

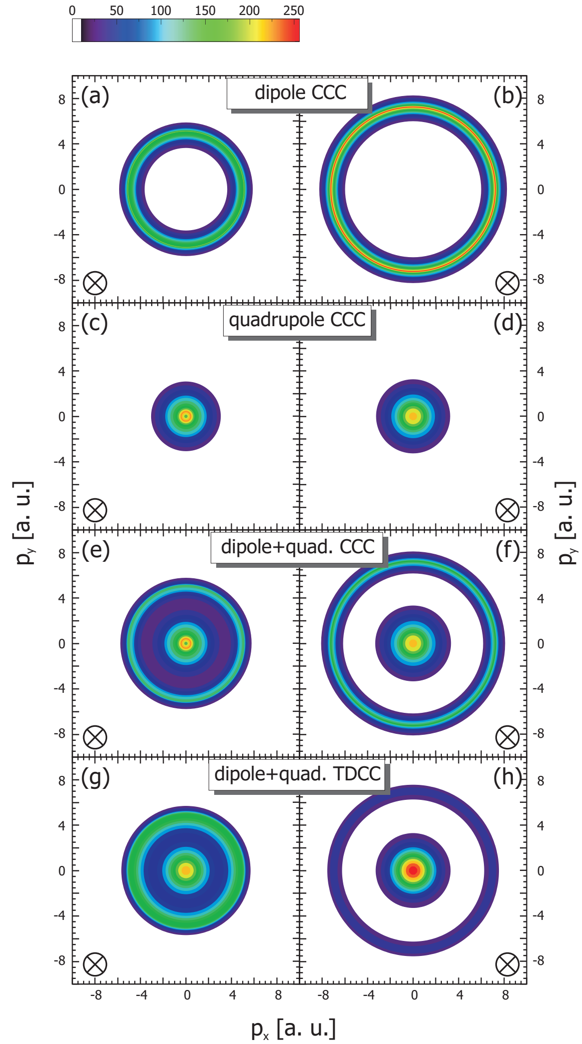

FIG. 2: Calculations for 440 eV (left column) and 800 eV (right column) photon energy with the same geometry as in the experiment (Figure 1c,d), for a width of $\Delta p_z/p_{\max} \le \pm 15\%$. In panels (a,b) the CCC calculations include only the dipole interaction, while (c,d) display solely the quadrupole terms and (e,f) show the full theory. The panels (g,h) show a full TDCC calculation including dipole and quadrupole terms and in contrast to CCC their interference terms.

tain the dipole part (Figure 2 a,b) and quadrupole contributions (Figure 2 c,d) as well as the coherent (TDCC) and incoherent (CCC) sum of the dipole and quadrupole matrix elements (Figure 2 e,f). Both calculations show ions with zero momentum for the quadrupole, but not for the dipole terms. The individual dipole and quadrupole contributions from CCC and TDCC agree well. The higher momenta contain contributions from both the dipole and the quadrupole term. The TDCC is a direct solution of the time dependent Schrödinger question. It is not possible to connect this to the intuitive picture of ionization mechanisms. As the calculation is exact up to the quadrupole term, it does, however, include all the possible PDI mechanisms. The CCC calculations, in turn, can be analyzed in terms of the Feynman diagrams [10]. The CCC theory does include the shake-off and the knock-off diagrams, but it does not contain the so-called butterfly diagram (Fig. 1c in Amusia et al. [14]), which has a vertex where the photon couples to both electrons simultaneously. As the CCC reproduces the observed low energy ions, it is clear that the QFM at these non-relativistic photon energies is not dominated by the butterfly diagram, as proposed when the QFM was first predicted [14]. This is in line with a more conventional interpretation, that refers to QFM as a specific kinematic region of equal energy sharing and low ion recoil, dominated by non-dipole PDI [30, 37]. The butterfly diagram is the extreme case of SO and TS1, when the time between the electron-electron vertex and the photo-absorption vertex is exactly zero, which is obviously not yet the case at 800 eV.

In conclusion, we performed kinematically complete experiments on single photon double ionization of helium at 440 and 800 eV with linearly polarized light. In contrast to all previous experimental work, we have observed, for the first time, the ions with close to zero momentum, originating from a back-to-back emission of two electrons with equal energy sharing. This observation confirms the quasi-free mechanism predicted nearly four decades ago by Amusia et al. [14]. Our CCC and TDCC calculations show that these slow ions are produced by the quadrupole interaction with the photon field and can be reproduced without including the butterfly diagram. The newly observed double ionization mechanism probes the two electron wave function at the electron-electron cusp, a region previously inaccessible. These cusp electrons can be thought of as a bosonic electron pair, which is virtually free as they compensate completely each other's momentum, while the nucleus is at rest. When this pair is photoionized, a total energy of 720 eV, stored in the form of a Coulomb repulsion, is released

We thank the staff of the Advanced Light Source, in particular H. Bluhm and T. Tyliszczak from beamline 11.0.2.1, for their outstanding support. M. S. Schöffler thanks the Alexander von Humboldt foundation for financial support. This work is supported by the Deutsche Forschungsgemeinschaft, DAAD, and the Office of Basic Energy Sciences, Division of Chemical Sciences, U.S. Department of Energy, and DOE-EPSCoR under Contracts No. DE-AC02-05CH11231 and No. DE-FG02-07ER46357. Resources of the Australian National Computational Infrastructure Facility were used in this work. We thank Miron Y. Amusia for encouraging us for 16 years to perform the present experiment.

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