Single photon double ionization of Helium at 800 eV – observation of the Quasi Free Mechanism

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Synopsis: In a kinematically complete experiment we have measured the photo double ionization of Helium at a photon energy of 800 eV and observed He 2+ ions with 0 momentum, corresponding to a back-to-back-emission of the two electrons. The results are in good agreement with theoretical calculations.

Photo double ionization (PDI) of Helium is widely believed to proceed through knock-off (TS1) or shake-off, with the last one dominating at high photon energies [1]. 40 years ago Amusia and coworkers [2] have predicted a third, high energy, mechanism, which they termed “Quasi Free Mechanism” (QFM). They predicted that this dipole forbidden mechanism becomes important at high photon energies. While the energy sharing for the well-established PDI exhibits a U-shape, for QFM an additional maximum in the central region (equal energy) appears. Here the photon is predicted to couple to the electron pair directly, leading to (dipole forbidden) back-to-back emission of the electrons with equal energy. The mechanism results from the electron-electron-cusp region of the ground state wave function, i.e. the region where both electrons are very close.

Here we provide the first experimental proof of this mechanism and additional theoretical calculations (see also [3]).

We used the COLTRIMS reaction microscope technique [4] with a specially optimized time and space focusing spectrometer that is capable to collect electrons with energies up to 420 eV (more than 50% of the excess energy) with nearly 4π solid angle and Helium ions with momenta up to 15 au. 800 eV linearly polarized photons from the Advanced Light Source (Berkeley) were intersected with a supersonic gas jet of Helium.

In Figure 1 the momentum distribution of the recoiling He 2+ ions is shown (perpendicular to the polarization direction) with a gate on the momentum along the polarization direction (pz<1.5 au). This condition highlights the quadrupole contribution appearing at 0 ion momentum, while the main contribution resulting from double ionization via shake-off is strongly suppressed. The remaining SO contribution can be observed in the outer ring in Fig. 1, as the shake-off-electron only carries negligible momentum (< 0.5 a. u.). As predicted for the QFM ions with 0 momentum appear in both, experiment and time dependent close coupling (TDCC) calculations.

Figure 1: Slice of the He 2+ momentum distribution perpendicular to the light propagation and polarization direction; experiment (left), TDCC (right).

References