Non-dipole and Quadrupole Photoionization of H_2

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Synopsis We report on a kinematically complete measurement of double ionization of H_2 by a single 800 eV circularly polarized photon. We observed the angular emission pattern of electrons originating from a pure quadrupole transition and discovered modifications of molecular frame photoelectron angular distributions by non-dipole contributions.

At high photon energies, the electric dipole approximation is no longer valid and the photon's linear momentum evidently modifies photoelectron angular distributions [1, 2]. We investigated such effects in the laboratory and molecular frames for the photo-double-ionization of H_2. We employed the COLTRIMS reaction microscope technique [3] and intersected a cold supersonic H_2 gas jet with a beam of 800 eV circularly polarized photons from beamline P04 at PETRA III (DESY, Hamburg).

The back-to-back emission at equal energy sharing between the two electrons is dipole-forbidden for photo-double-ionization of H_2 [1]. As Fig. 1(a) displays the angular emission pattern of such electrons in the laboratory frame, it shows a genuine quadrupole contribution to the photoionization process of molecular hydrogen. Once the photon's linear momentum cannot be neglected, it evidently modifies the dipole-dominated emission patterns in the molecular frame. The experimental data in the upper hemisphere of Fig. 1(b) displays the emission of high-energetic photoelectrons from double ionization of H_2. The photon hits the molecular axis at a 45° angle and makes the protons distinguishable, as one is closer to the incoming light than the other. The linear momentum changes the wavelength of the outgoing electron depending on its emission angle in the molecular frame, and the probability distribution is altered accordingly. The effect emerges in the observation as a higher chance for detecting an electron emitted towards the incoming light direction (than away from it) along the molecular axis. The lower hemisphere shows a corresponding calculation according to [2], that is consistent with this observation.

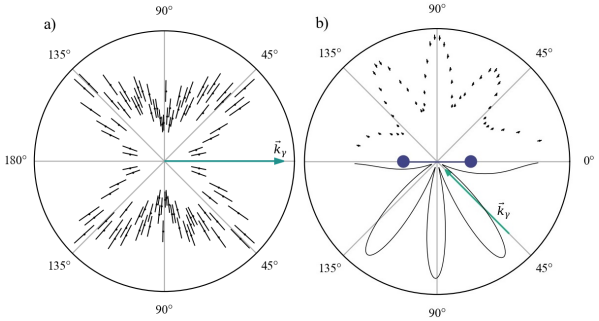

Figure 1. Photo-double-ionization of H_2 by a single 800 eV circularly polarized photon. **a**) Laboratory frame photoelectron angular distribution. Displayed electrons measured in coincidence with second electron emitted back-to-back (180°±20° enclosed by electron vectors) and equal energy sharing ($0.4 < E_{e1}/(E_{e1} + E_{e2}) < 0.6$). The light propagation axis is horizontal ($\vec{k}_\gamma$). Due to dipole selection rules, the emission pattern originates from pure quadrupole transition. **b**) Molecular frame photoelectron angular distribution. Data points: electrons measured with asymmetric energy sharing ($E_{e1} > 0.7(E_{e1} + E_{e2})$), molecular axis is horizontal and fixed in space with respect to the incident light at 45°±15°. Calculation according to [2]. The non-dipole modifications lead to the asymmetry between the left and right hemisphere.

References

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