

Spectral dependence of molecular frame photoemission in dissociative ionization of HD induced by circularly polarized light

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Dissociative photoionization (DPI) of H₂ and D₂ at resonance with the Q₁ and Q₂ doubly excited states gives rise to a rich ultrafast electron and nuclear dynamics in the femtosecond regime [1]. In this context, remarkable symmetry breaking in molecular frame (MF) photoemission induced by linearly [2,3] and circularly [4] polarized VUV synchrotron radiation have been demonstrated and assigned to interferences between indistinguishable DPI channels which involve ionic states of different u and g symmetry [3,4]. The outcome of the interferences depends on the time delays between the relevant channels, which are controlled by the kinematic of the dissociation process varying for DPI of the three isotopes H₂, D₂ and HD. Since HD is identical to H₂ and D₂ from the electronic perspective, noticeable differences observed in the recorded molecular frame photoelectron angular distributions (MFPADs) can a priori be interpreted as a signature of a coupling between electronic and nuclear dynamics.

We will report first results of a spectral study of resonant DPI of the HD molecule induced by circularly polarized synchrotron radiation in the 30-33 eV photon energy range. A comparison with previous measurements and calculations for DPI of H₂ and D₂ at comparable excitation energies [3,4,5] will be reported.

We have performed these investigations using the electron-ion vector correlation (VC) method [6], combining a double velocity spectrometer [7] with the well-defined polarized light [8] delivered at the beam line DESIRS at SOLEIL synchrotron.

The outcome of such experiments consists of recording MFPADs as a function of the kinetic energy release (KER) of the H⁺/D⁺ fragments. The MFPADs are expressed in terms of five one-dimensional F_{LN} functions [9] related to the dipole matrix elements of the PI reactions. As illustrated in [5], a convenient way to represent the KER dependence of the F_{LN} functions is to take advantage of their development in Legendre polynomials expansion [10]. We will focus in particular on the KER dependence of the F₂₁ and F₁₁ functions which are fingerprints of the relative phases of the dipole matrix elements for the parallel and perpendicular components of the DPI transitions.

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