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PAPER

Molecular frame photoemission by a comb of elliptical high-order harmonics: a sensitive probe of both photodynamics and harmonic complete polarization state

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Due to the intimate anisotropic interaction between an XUV light field and a molecule resulting in photoionization (PI), molecular frame photoelectron angular distributions (MFPADs) are most sensitive probes of both electronic/nuclear dynamics and the polarization state of the ionizing light field. Consequently, they encode the complex dipole matrix elements describing the dynamics of the PI transition, as well as the three normalized Stokes parameters s_1, s_2, s_3 characterizing the complete polarization state of the light, operating as molecular polarimetry. The remarkable development of advanced light sources delivering attosecond XUV pulses opens the perspective to visualize the primary steps of photochemical dynamics in time-resolved studies, at the natural attosecond to few femtosecond time-scales of electron dynamics and fast nuclear motion. It is thus timely to investigate the feasibility of measurement of MFPADs when PI is induced *e.g.*, by an attosecond pulse train (APT) corresponding to a comb of discrete high-order harmonics. In the work presented here, we report MFPAD studies based on coincident electron-ion 3D momentum imaging in the context of ultrafast molecular dynamics investigated at the PLFA facility (CEA-SLIC), with two perspectives: (i) using APTs generated in atoms/molecules as a source for MFPAD-resolved PI studies, and (ii) taking advantage of molecular polarimetry to perform a complete polarization analysis of the harmonic emission of molecules, a major challenge of high harmonic spectroscopy. Recent results illustrating both aspects are reported for APTs generated

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