High-Harmonic Source for Coincidence Spectroscopy nf Non-Linear XUV Photoionization

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Scrutinizing nonlinear photoionization dynamics in the extreme ultraviolet (XUV) regime typically requires the intense pulses provided by free-electron lasers. Here, we present the development of a powerful table-top XUV source and its application in coincidence measurements of two-photon double ionization (TPDI). Our high-flux source is based on high-harmonic generation (HHG) in argon, driven by a frequency-doubled, 100 kHz repetition-rate fiber laser (30 fs, 515 nm). By utilizing a suitably coated XUV mirror, we achieve a quasi-monochromatized, coherent XUV beam delivering an unprecedented flux of 10^{13} photons per second at 26.5 eV. The exceptional intensity of the focused XUV pulses enables the direct observation of nonlinear processes in a cold gas target using a coincidence reaction microscope.

Applying this setup, we investigated the TPDI of argon driven by femtosecond pulses tuned around the 26.5 eV photon energy. Through highly differential photoelectron spectra and coincidence measurements, we evaluated the contributing TPDI mechanisms. Supported by atomic structure calculations and Monte Carlo simulations, our results reveal that the prevailing ionization pathway is not direct, but rather proceeds via the excitation and prompt photoionization of an autoionizing resonance in neutral argon. Furthermore, the extracted energy spectra exhibit clear signatures of pronounced electron-electron interaction (Coulomb repulsion) during the ultrafast emission process in the continuum.

We demonstrate that the dominant TPDI mechanism can be effectively controlled and altered by finely tuning the photon energy of the driving XUV pulses. Our work establishes that state-of-the-art table-top HHG sources can now drive complex nonlinear coincidence spectroscopy, paving the way for advanced phase-resolved and pump-probe studies of electron correlations in the XUV regime.