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Transmitting the change: From watching to steering electron dynamics with intense FEL fields

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The advent of Free-Electron Lasers (FELs) has made a dream come true: Imaging and spectroscopy of atomic and molecular structural and electronic dynamics on their natural time scale by XUV/X-ray pump&probe pulses. Besides slowly realizing this dream, FELs keep fueling a “dream-on revolution” by inspiring new questions: Can we understand the vast variety of nonlinear (and complex multi-electron) interactions of XUV/x-ray light with matter? Can we even use the strong FEL fields at high frequencies to steer electron dynamics on an atomic level?

Here, we will discuss some of our recent experiments and insights into extreme FEL interactions with matter. A home-built transient-absorption spectroscopy beamline setup brought to FLASH at DESY/Hamburg allowed the observation of characteristic gas-phase atomic and molecular spectra in “Fraunhofer-type” transmission geometry. This approach allows to study the modification of fundamental resonances as a function of intensity and time delay of two coherently split XUV pulses from the same FEL shot.

Focusing intense FEL light near 60 eV on the 2s2p double excitation of a cloud of helium atoms, we observed a change of the characteristic Fano line shape. Modeling this interaction allows to identify the mechanism: A strong coupling between the ground and the (doubly) excited state at the onset of a two-electron Rabi cycle allows to exert phase control of the electronic excitation. Control over the phase is a prerequisite for shaping electronic wavefunctions. It can be shown that short FEL pulses (shorter than the lifetime/dynamics of the excited state) are key to this controllability, while longer pulses essentially only ionize the atom.

Towards exploring and understanding larger systems, we turn to neon atoms, where XUVpump/XUV-probe measurements of the absorbance change in neon gas allowed to directly watch the time-resolved buildup of doubly charged ions in the intense XUV field. Moreover, resonances of Ne2+ were observed as a function of intensity near 50 eV photon energy, exhibiting a spectral (Stark) shift on the order of 50 meV for one out of three lines of the 2p-3d multiplet transitions. A 2-fs coherence spike showed up in the nonlinear XUV-optical absorption spectrum near temporal overlap, pointing at the feasibility of gas-phase multidimensional spectroscopy even with self-amplified spontaneous-emission mode (SASE-)FELs.

We conclude with first time-resolved experimental results from iodine-containing molecules with site-specific pump&probe pulses resonant on the iodine 4d core resonance. Here, direct tracking of dissociation dynamics through different molecular geometries becomes possible, and is expected to gain massively by future two-color and broad-band XUV probing methods for extracting additional pathways and providing comprehensive control knobs even for steering chemistry on the fundamental level of atomic-site-selected electrons.

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