CFEL SCIENCE

SEMINAR

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Resonant absorption in molecules: Probing ultrafast dynamics and its laser control

Absorption spectra are the fingerprint of resonant bound–bound transitions between distinct quantum levels in atoms and molecules. Due to their complexity, including the nuclear motion and molecular bonds in the valence shell, spectroscopic structures in molecules are typically much broader as compared to their appearance in isolated atoms, and thus it is more challanging to disentangle them. In XUV and x-ray spectra, well defined core-to-valence transitions give atom-specific access to the molecule. Ultimately, with attosecond x-ray pump and probe pulses, a dream is to site-specifically initiate and probe a molecular wavepacket. Thus one would be sensitive to electronic coherences, which could then be quantified locally in the vicinity of individual atoms together with the motion of these nuclei. A deeper knowledge of these coherences, including their response to an external light stimulus, may also enable a new way to understand and optimize the laser control of molecular dynamics.

We build on experience of the laser control of multi-electron dynamics in isolated atoms, encoded in their absorption spectra. In this talk I will give an overview of our most recent activities to transfer these concepts to small molecular systems. More specifically, using intense XUV-FEL pulses from FLASH, we explore the site-specific resonant pumping and probing of an iodine 4d core-to-valence transition in gas-phase diiodomethane (CH_2I_2). While electronic dynamics and coherences have not yet been uncovered in this experiment, comparison with theory and previous static synchrotron measurements allowed us to extract the time scales of a non-trivial dissociation pathway of this molecule, which involves a transient isomeric geometry. In a second experiment we explore the possibilities of controlling coherent dynamics in a far more complex system, that is a metallo-organic phtaloyanine (AlClPc) complex in a liquid-phase environment. Hereby we stick to all-visible ultrashort laser pulses, hence avoiding the technical challenges of in-vacuum liquid jets. However, few-femtosecond and attosecond dynamics are in principle still encoded in the laser-driven real-time dipole response of the system, which can be extracted from the measurement. In a last but not least example, we demonstrate the laser control of a core-to-valence x-ray transition in sufur hexafluouride (SF_6) at the sulfur $L_{2,3}$ edge.

Hosts: Francesca Calegari / CFEL Molecular and Ultrafast Science Seminar