Ultrafast Dynamics: Pump-Probe Experiments at Free Electron Lasers

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One of the most exciting opportunities opened by Free Electron Lasers (FEL) is the feasibility of performing, for the first time, pump-probe experiments in the VUV, EUV and X-ray wavelength regimes with femtosecond time resolution or even below. Here, a first light pulse (IR, Vis, EUV, EUV or X-ray) initiates dynamics, like a chemical reaction, a phase transition, spin-, orbital, or charge-density waves in solids and a second pulse, impinging at a variable but well-defined time delay, probes the motion.

In the talk, a first series of such experiments, performed at the VUV-FEL in Hamburg, FLASH, the SCSS test facility in Japan as well as pioneering measurements at the LCLS X-ray FEL will be presented. At FLASH and SCSS the VUV-pulse has been split by a back-reflecting mirror that is cut onto two halves. One of the pulses can then be delayed by moving the two half-mirrors with respect to each other reaching sub-femtosecond accuracy. In a demonstration experiment the vibrational wave-packet motion in deuterium molecular ions with a round-trip time of about 22 fs could be traced, indicating a time-resolution of better than 10 fs. Moreover, the isomerization time in VUV-excited acetylene evolving into vinylidene cations proceeding within about 50 fs was measured for the first time, ending a 20 years controversial debate. More recently, the dynamics of a suite of other charge migration reactions was investigated via time-resolved many-particle fragment detection in a reaction microscope (REMI) watching atoms move in real-time. Using the CAMP instrument at LCLS first optical pump – X-ray probe experiments have been performed on aligned molecules, clusters and biological nano-crystals highlighting the rich future potential of these methods envisioning tracing Ångstrom spatial changes at femtosecond time resolution.
At the same time we witness tremendous technological progress at FELs: Non-linear autocorrelation traces showed sharp peaks with a FWHM in the order of the coherence length of the radiation (4 fs at FLASH and 10 fs at SCSS), explained by the statistical nature and coherence properties of the FEL pulses and pointing towards exciting possibilities to perform attosecond X-ray – X-ray pump-probe experiments at the LCLS. In addition, new machine developments, like “slotted spoilers”, “longitudinal space charge amplifiers” and high-harmonic generation or lasing in FEL-excited targets nurture the expectation to create attosecond pulses that might even be synchronized with an optical laser on a sub-femtosecond time-scale, which would open the door towards keV-attosecond science.