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Ultra-fast 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 wave-length regimes with femtosecond time resolution. Here, a first light pulse (IR, EUV, VUV 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 into 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 that 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. Autocorrelation measurements at FLASH and SCSS showed a sharp peak in the non-linear autocorrelation trace with a FWHM in the order of the coherence length of the radiation (4 fs at FLASH and 10 fs at at SCSS). This has been explained by the statistical nature and the coherence properties of the FEL pulses pointing towards exciting possibilities to perform attosecond X-ray - X-ray pump-probe experiments at the LCLS. Here, using the CAMP instrument, 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.