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Ultrafast pump-probe X-ray spectroscopy at SwissFEL

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With their unique combination of high per-pulse X-ray flux and femtosecond pulse durations, hard X-ray free electron lasers are an almost ideal source for time-resolved structural experiments. The SwissFEL, which is currently under construction at Paul Scherrer Institute, will be capable of generating femtosecond hard x-rays pulses in the photon energy range of 2-12 keV, with a planned emphasis on performing femtosecond pump-probe measurements. Experimental Station A (ESA) of the SwissFEL will focus on probing the ultrafast dynamics of systems in solution using a combination of X-ray spectroscopy and scattering. The primary goal of ESA will to enable users to perform X-ray absorption (XAS) and emission (XES/RIXS) spectroscopy pump-probe experiments with a focus on the 2-5 keV energy range with <50 fs time resolution (FWHM) using a range of excitation wavelengths (UV to IR). ESA will also be capable of using advanced methods for the injection of aerosol particles and sub-micron protein crystals. This presentation will provide an overview of the techniques we expect to have available at ESA and will present a prototypical 'ESA experiment' which uses a combination of femtosecond X-ray diffraction, scattering, and spectroscopy to investigate the electronic and structural dynamics. We will present recent results from an LCLS experiment devoted to study the interaction of intense femtosecond X-ray pulses with condensed matter. The electronic states of different forms of copper were probed on a shot-to-shot basis using a multi-crystal dispersive x-ray emission spectrometer. From these measurements we were able to establish the cross-sections of sequential-ionization and multi-photon absorption nonlinear processes.

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Large amplitude spin dynamics driven by a THz pulse in resonance with an electromagnon

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Multiferroics are materials with more than one type of ferroic ordering. Recently they have attracted strong interest for potential applications where electric fields control magnetic order. The ultimate speed of control via magnetoelectric coupling, however, remains largely unexplored.

Recent developments in ultrafast pump-probe experiments have introduced new methods to selectively stimulate and observe the structural dynamics in strongly correlated electron systems. Their key element is the use of the extremely intense sources of short-pulse electromagnetic radiation over a broad frequency range from the far-infrared to x-rays. Pump-probe experiments exploiting these parts of spectrum offer unique ways to gain important information about the energy-transfer processes in crystals and thus insight into the microscopic mechanisms governing their behavior.

Here we report on an experiment [1] in which we use a pump-probe technique to study ultrafast spin dynamics in $TbMnO_3$, a model spin-cycloid multiferroic where the ferroelectric polarization arises directly from the frustrated magnetic order [2]. We excite coherent spin motion with an intense few-cycle terahertz (THz) light pulse tuned to resonance with an electromagnon, an electric-dipole active spin excitation associated with the magnetoelectric coupling [3]. To directly see the coherent response of the magnetic structure, we perform

femtosecond resonant x-ray diffraction measurement at the LCLS x-ray free electron laser. We determine the motion of the Mn spins during the excitation process by measuring the first harmonic magnetic diffraction peak. By comparison with a static x-ray diffraction experiment, we are able to determine that the THz pulse induces coherent rotation of the spin-cycloid plane with amplitude of 4°.

We prove that the pump-probe experiments employing ultrashort x-ray pulses can serve as a powerful spectroscopic tool which gives direct access to the spin dynamics and allows to visualize the spin motion associated with a particular excitation. Furthermore, we demonstrate that the magnetic structure of multiferroics can respond within a fraction of a picosecond after the excitation, what is far above the previously established limit of several milliseconds and suggests a way to achieve ultrafast heatless multiferroic domain switching.

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