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Re.: Letter of Intent, Linac Coherent Light Source (LCLS) Experimental Program

Proposed experimental program: Ultrafast dynamics of condensed matter systems using novel pump-probe techniques.

The dynamics of the fundamental physical processes that govern the properties of many condensed matter systems occur on time scales that will become accessible with the ultra-short pulses produced by the LCLS. In particular, the LCLS will greatly expand the spectral range of radiation produced on such short timescales, which is a regime that is currently limited (primarily) to table-top laser sources operating in the visible, near-IR and soft-UV portion of the electro-magnetic spectrum. X-ray pulses from the LCLS can be used to interrogate the dynamics of many types of materials via a pump-probe kind of experimental architecture, where the system is perturbed in some fashion, and then, after a variable delay, LCLS pulses are used to investigate the relaxation mechanism of the system. The methods available for producing an ultrafast perturbation (*i.e.* the pump pulse) are currently limited, and most experiments rely on exposure to short pulses from a conventional laser, which heats the sample quickly, or changes to local carrier concentration on a short timescale. These excitations produce a mix of initial states, and the subsequent decay of this broad distribution can significantly complicate the analysis of data from the pump-probe experiment.

The accelerator at the LCLS, however, will be capable of producing extremely short and very intense pulses of radiation in the terahertz (THz) portion of the spectrum.

The THz pulses could be produced either via the use of a bend magnet, or through transition radiation when the electron bunches in the accelerator traverse a thin Al target in the accelerating cavity. Relatively simple optics can then be used to bring the THz radiation to an experimental station coincident with the x-ray radiation from the LCLS. The field strengths from the THz pulses from such a source are predicted to be extremely large, with peak electric fields over 10^9 V/m and magnetic fields over 10 Tesla. These fields would be concentrated in pulses of about 150 fs in length, and thus they would be ideal sources to coherently perturb materials that are investigated in pump-probe experiments at the LCLS. Experience gained from the the deep UV free-electron laser (DUV-FEL) at the National Synchrotron Light Source (NSLS) of Brookhaven National Laboratory (BNL) strongly suggests that these fields could be achieved with the LCLS with very few technical obstacles; peak electric fields of THz pulses produced at the DUV-FEL exceed the range of current detection methods, and extrapolations of low-power measurements indicate that the peak fields of the DUV-FEL are in excess of 10^8 V/m, with a pulse length of about 1 pico-second (ps).

Two classes of experiments are envisaged for the initial phases of the experimental program: THz excitation of ultra-fast magnetization dynamics, and coherent excitation of phonon modes and subsequent potential well mapping in ferroelectric materials. The BNL team has initiated experimental programs in both of these areas using the DUV-FEL and we are developing the techniques and expertise for ultrafast detection of intense THz pulses and synchronization of THz pump pulses with optical probes. In addition, the BNL team can draw upon the extensive experience of researchers at the NSLS to address issues such as x-ray probes of magnetic order and x-ray scattering techniques for examining lattice dynamics.

THz-driven Magnetization Dynamics:

Magnetization dynamics are often investigated in the frequency domain using swept field or frequency methods such as ferromagnetic resonance (FMR). However, it is often desirable to follow the magnetization of a sample in the time domain, particularly after a sharp perturbation to the initial magnetization. Time-domain techniques have been developed that provide such ultrafast (ps-scale) field profiles to magnetic materials. The most common method involves the placement of a planar RF waveguide (*e.g.* a microstrip) adjacent to a magnetic film, and then exciting precessional motion of the magnetic moments by the application of a fast RF pulse through the waveguide. Researchers at the NSLS, along with collaborators from Columbia University, have recently combined this technique with synchrotron-based x-ray magnetic circular dichroism (XMCD) to examine the precession of elemental moments in magnetically soft FM alloys [1]. The experiments made use of a pump-probe architecture similar to the proposed program at the LCLS.

The perturbing fields produced by planar RF waveguides are usually on the order of about 10 oersted (Oe). This restricts the application of this magnetic excitation technology to the study of soft ferromagnets (FM) such as permalloy ($Ni_{81}Fe_{19}$). Occasionally, ultrafast dynamics experiments have been performed on hard FM materials. For example, ultrashort magnetic field pulses (duration 2 to 8 ps) have been produced by a beam of relativistic 50 GeV electrons generated by the Stanford Linear Accelerator Center (SLAC) [2]. These electron beams have been used to examine the dynamic response of materials such as thin films of $Co_{28}Pt_{72}$ (H_c of 16 to 32 kOe). In these experiments, the magnetization of the samples was measured *ex-situ* and in the remanent state and the dynamics of the elemental moments had to be inferred from knowledge of the initial and final states.

The ultrashort and intense pulses of THz radiation readily available at the LCLS, combined with the unparalleled temporal resolution and coherence of the LCLS x-rays, can have a revolutionary impact on the study of magnetization dynamics in entire classes of magnetic materials. Many interesting dynamic phenomena in magnetism on ultrafast timescales occur in materials with considerably higher coercive fields. For example, spin-lattice (*i.e.* phonon coupling) relaxation times in manganites occur on a ps time scale. The coercive fields of even the "softest" ferromagnetic manganites, however, are on the order of several hundred Oe and are clearly out of the range of the fields produced by co-planar waveguides. THz fields should have both the magnitude to initiate a response in these and other hard magnetic materials and pulses short enough to provide a relatively clean kick to the magnetization of these samples.

One intriguing possibility that requires the combination of the THz pulses and x-ray radiation of the LCLS is the examination of spin-flop transition in anti-ferromagnets and ferrimagnets. Hematite (a-Fe2O3) provides a useful example of this class of materials. The 10 T peak field of the THz pulses should be sufficient to initiate the spin-flop mode in a-Fe2O3. The subsequent orientation of AF domains can then be examined with x-ray magnetic linear dichroism (XLMD) with sub-ps resolution. In addition, in systems where there is strong spin-orbit coupling, such as materials containing rare-earth dopants, x-ray scattering experiments can be designed to investigate the degree to which the re-orientation of the magnetic moments are accommodated or mediated by the lattice over fs to ps range timescales.

Dynamical Potential Well Study in Ferroelectrics

Ferroelectric materials of the ABO_3 perovskite structure (e.g. $PbTiO_3$) exhibit structural deformations near their ferroelectric transition temperature. The approach to this transition is often accompanied by changes to the atomic potential well and soft phonon behavior. Though models have been proposed for the potential well shape as the system passes through the transition, the actual shape is unknown.

With the advent of high-intensity coherent THz pulses from accelerators, a new method for studying potential wells may now be feasible. The method exploits the very high transient electric field associated with accelerator produced coherent THz pulses. Generated as transition radiation, pulses with ~ 100 μJ energy have been produced at the NSLS/SDL linac, consistent with theoretical predictions. Such a pulse, when focused into a diffraction-limited spot, creates transient fields on the order of 1 MV/cm. The bunch length, charge and relativistic energy of the SLAC/LCLS linac should be capable of 10 mJ pulses, and significantly higher peak fields, perhaps exceeding GV/m levels. Based on classical approximations for a low energy optic phonon, we can estimate the relative displacement for a particular set of atoms to be ~0.2 pm (0.02A) for a 1 MV/cm applied electric field, and 10 times larger for the anticipated performance of LCLS THz pulses. Even larger displacements can be anticipated in the vicinity of the ferroelectric transistion, and should be detectable in an x-ray diffraction experiment.

Therefore, we propose extracting THz radiation, produced as transition radiation from a thin metal foil, or as diffraction radiation from a small metal aperture, through a UHV quartz window and transporting it to a location where diffraction can be performed using x-rays produced by the same electron bunch. One of the optical paths would include a variable delay to enable pump-probe type time-resolved methods. In one experiment, the THz and X-ray pulses would be set to arrive simultaneously, and atomic positions would be determined by x-ray diffraction as a function of the applied THz field strength. From this, the detailed shape of the potential well could be extracted. In another experiment, the arrival time of the x-ray pulse (relative to a THz pulse) would be varied to track atomic position as a function of time following their initial, THz-induced displacement. Near to a soft mode, the relaxation to the equilibrium position is likely to be several picoseconds, and provide additional insight into the material's lattice dynamics.

[1] "Precessional dynamics of elemental moments in a ferromagnetic alloy," W.E. Bailey, L. Cheng, D. J. Keavney, C.-C. Kao, E. Vescovo, and D.A. Arena, *in preparation.*

[2] "The ultimate speed of magnetic switching in granular recording media, " I. Tudosa *et al., Nature* **428**, 821 (2004).