Letter of Intent
Study of Ultrafast Processes in Solids at SABER

By
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Here we describe experiments that we propose to perform at SABER in 2008. The experiments will probe dynamics in solids excited by passing through thin film samples one or several relativistic electron bunches. The excitation of dynamics is due to the unique electric and magnetic field pulses accompanying the intense and well focused relativistic electron bunches that will be available at SABER.

Our experiments are designed to address scientific issues associated with the interaction of an ultrafast electron beam and its associated large electric and magnetic fields with a material. Of particular interest are measurements as a function of pulse length, keeping the total charge per bunch and the lateral bunch dimensions unchanged. The main questions to be addressed are:

1. The magnetization dynamics and magnetic switching in *magnetic samples* triggered by the ultrafast and strong magnetic field.
2. The pulse-width dependent heat deposition of the electron beam in the sample.
3. The ultrafast switching of *ferroelectric samples* triggered by the variable-length electric field pulses associated with the beam.

Previous accomplishments and experimental technique

Previous work has been focused on magnetization dynamics and was carried out with electron bunches at the FFTB of SLAC. This work was highly successful and was published in premier scientific Journals and received world-wide publicity in the press. The work resulting in the following accomplishments:

(1) The invention of fast magnetic switching through precessional magnetization reversal [1],
(2) The establishment of the ultimate limits of magnetic recording in granular recording media [2],
(3) The first quantitative insight into the chaos created by magnetization precession at large angles [3].

Building on the expertise accumulated in these prior experiments, we now intend to continue the experiments with ultrafast field pulses, in particular by making use of the insight that can be gained by comparing the effects of uncompressed electron bunches possessing a duration of order of several ps [10^{-12}s] in the laboratory frame to the one of compressed bunches whose duration reaches far into the fs [10^{-15}s] range. Comparison of events occurring at ps- and fs-time scales is important in solid state dynamics as the electron-phonon relaxation takes place on the ps-time scale while on the fs-scale, electrons can have high temperatures while the crystal lattice is still cold [4]. Some of the results obtained in the last experiments at FFTB in August 2005 will serve as a

guideline and show that we have the unique possibility to access new physics of great general interest.

The experimental arrangement for our experiments at the FFTB is illustrated in Fig. 1 and explained in the caption. In the shown example and in our discussion below we shall consider ferromagnetic thin film samples with an in-plane easy magnetization axis. Each magnetic field pulse comes together with a perpendicular electric field pulse of amplitude E = cB where c is the velocity of light. The magnetic patterns are retained by the material according to the principle of magnetic recording and have been imaged with secondary electron microscopy (SEMPA) weeks after the experiment. The heat is deposited in the sample mainly by the action of the electric field pulse, but the temperature reached at the time of magnetization dynamics is revealed in the magnetic pattern. This arises because the magnetic properties determining the magnetic pattern show characteristic temperature dependences, vanishing completely when the Curie point T_c is reached. Exposing a thin magnetic film which has a preferred in-plane easy axis to a short linac magnetic field pulse generates a figure-8 pattern of the switched magnetization, as shown in Fig. 1, in those locations where the temperature of the crystal lattice of the sample remained below T_c. These results gave rise to the concept of precessional (or ballistic) switching based on the precession of the magnetization about a perpendicular magnetic field, where the dissipation of angular momentum, normally the bottleneck in magnetic switching, is transferred directly to the field itself [1].

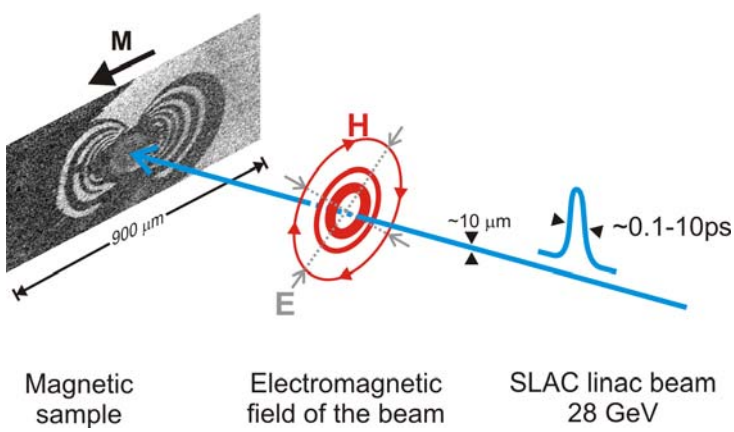

Fig. 1 Schematic of the magnetic switching experiment using an ultrashort electron bunch which is surrounded by an electric and a magnetic field, as shown. The high-energy electron bunch simply traverses the magnetic sample with an in-plane easy axis, imprinting a magnetic pattern or, in the case of a ferroelectric film, a ferroelectric pattern that depends on the number of electrons in the bunch and surprisingly also on the temporal length of the bunch. Before exposure to the beam the sample is uniformly magnetized in-plane. In the beam focus, a large gray area is visible where the sample was heated to T>Tc.

The focus of the new experiments is the effect of variable bunch *lengths* on magnetic patterns generated with electron bunches of constant electric *charge* $Q = N e \sim 1$ nC. The normal bunch length in the laboratory frame of reference of $\sigma_z = 700$ μm (corresponding to a temporal pulse length of 2.5 picoseconds) can be compressed to $\sigma_z = 21$ μm (70 femtoseconds) without loss of electrons, that is by keeping the total electrical charge Q of the bunch constant. The pulse duration in the lab frame is thus reduced from the ps-level far into the fs-regime.

Existing Observations in Experiments with Variable Bunch Lengths

Based on the above simple considerations, differences between the ps- and fs magnetic patterns recorded on the same sample are thus not expected. However, Fig.2 shows that differences in the pattern produced with the fs(~ 150 fs) and the ps(~5ps) bunch in the same sample do actually occur. The material is a 10 nm thick uniaxial $Co_{70}Fe_{30}$ film.

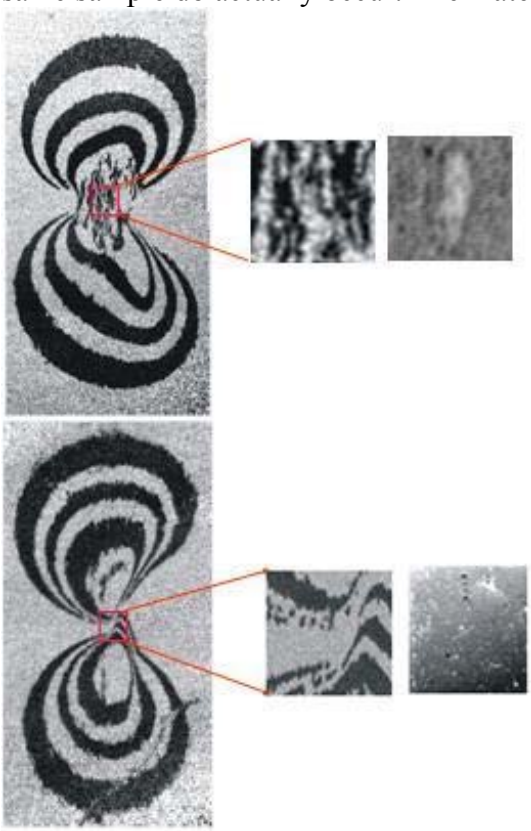

Fig. 2 Enlarged magnetic and topographical images of the region of electron bunch impact with one ps pulse (top) and one fs pulse (bottom). The topographic SEM pictures are the ones on the far right. The scales on all the pictures are the same; the entire magnetic patterns are ~450 x 900 microns and the zoomed in regions are all ~70 x 70 microns. Clearly visible in the ps topographic image is a spot of dimension 20 x 70 microns, while the fs central region remains remarkably damage free.

First we note that the patterns are quite similar in size and overall appearance as expected, showing the same number of black rings where the magnetization has switched from up to down. However, in the fs-pattern, the switched dark rings change from wide in the outer ring to narrow in the next ring and back to wide. This indicates that the damping of the magnetization relaxation after the field pulse depends strongly on time when the excitation occurred with the fs-pulse, becoming very large after several precessions leading to the wide innermost switched ring. The width of the switched rings is related to the magnetic chaos developing with time through spin wave instabilities [3], yet with ps-pulses one does not see such extreme chaotic behavior.

Furthermore, the fs pattern is clearly visible up to the location where the electron bunch passed. In fact, one notices a gap with no pattern in the center which is ~35 μm wide, corresponding to the width of the electron bunch focus for the compressed beam. Yet with the ps-pulse exposure, a large region with a stripe-like domain pattern appears in the center (Fig. 2). This indicates that the crystal lattice of the film has been heated in the environment of the electron beam focus to temperatures above the Curie temperature T_c with the ps-bunch, but remained below T_c with the fs-bunch. Closer inspection of the

central region of the patterns with high resolution scanning electron microscopy reveals ablation in the focus with the ps-pulse. Yet no beam damage whatsoever is visible with the fs-pulse. This indicates that the mechanism of heat transfer from the electron bunch to the crystal lattice is less efficient when the bunch length is reduced to the fs-time scale.

Proposed Future Work

Experiments with Magnetic Materials.

Our initial future goal is the understanding of the mechanisms involved in generating the unexpected features of the patterns shown in Fig. 2.

First, we would like to explore the important question of why the compressed electron bunch heats the sample to a lesser degree than the uncompressed bunch. It is this heating issue that is at the heart of all future experiments with ultra-short, ultra-intense beams of high energy particles including LCLS photon beams. The fundamental question is this: *During interaction of an ultra-short, ultra-intense beam with a thin film sample, the beam loses a large amount of energy. What are the ultrafast energy dissipation mechanisms and the magnitude of energy dissipation that precede the eventual transfer of energy to the lattice?*

Naturally, we want to investigate the heating of the crystal lattice as well as the occurrence of ablation by the ultrafast electron beam in more detail, changing the chemical and physical properties of the magnetic film by using Fe, Co, and other magnetic thin films with different magnetic properties and controlled crystallinity. We expect that any beam of high energy primary particles such as hard x-ray lasers will show reduced heating in a thin film as most of the secondary excitations escape the sample before producing low energy excitations in the form of phonons.

Additionally, we would like to investigate the magnetic pattern in more detail. Simple considerations lead one to conclude that with very short pulses, the switched regions should exhibit a figure 8 pattern composed of two exact circles if the sample properties are homogeneous [3,4]. Yet it is obvious that in the fs-pattern of Fig.2 the switched (black) regions are elongated and cannot be fitted with two circles. The deviation occurs on the right side of the pattern where the magnetic field of the pulse is at large angle, close to antiparallel to the magnetization. The deviation signals that the magnetic solid refuses to accept the Zeeman-energy when the applied field is at a large angle to the magnetization. This is a new phenomenon for which we have no explanation at this time. It obviously warrants more experiments for clarification.

Ferroelectric domain dynamics with SLAC electron beams.

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The shortest time scale ferroelectric domain dynamics were observed in experiments that utilized photoconductive Auston switches (very common in the magneto-dynamic experiments as well) to create short electric pulses applied to the ferroelectric capacitors $Pt/La_{0.5}Sr_{0.5}CoO_3$ /$Pb(Nb_{0.04}Zr_{0.28}Ti_{0.68})O_3$ /$La_{0.5}Sr_{0.5}CoO_3$ /Pt with thickness of 200 nm [5]. The shortest field rise time achieved in these experiments is ~70 ps with the field amplitude of 25 MV/m. The displacement current through the ferroelectric capacitor was measured with time resolution of 20 ps, from which polarization dynamics were inferred. The minimum switching time of this ferroelectric device was found to be ~220 ps, and it is considered the state-of-the-art switching speed at the moment.

The main disadvantage of this method is the need to have macroscopic contacts to apply the electric pulses and measure the response. It was found that the RC-characteristics of the contacts affect strongly the dynamic response of the ferroelectric at the shortest times. Accurate determination of the intrinsic switching time t_0 would require reducing the pulse rise time well below 70 ps (not possible with the current photoconductive switches) as well as reducing the capacitor planar dimensions (down from 5 x 5 um) to avoid RC effects. Another disadvantage of this method is that the measured response is averaged over the ferroelectric volume inside the capacitor and does not allow spatially resolved investigation of domains nucleation and propagation.

Using the SLAC electron bunches resolves the temporal limitations encountered in the experiments described above. The SLAC experiments would be somewhat similar to the magnetic switching experiment. The electric field is applied for a short time, and then the resulting domain pattern is imaged. Note that imaging will have to be done ex-situ, but since even the smallest domains (~20 nm) have been found stable for weeks after exposure, it should not pose a problem. The magnitude, direction and the pulse length of the applied field can be very accurately controlled in SLAC experiments. The field will be very uniform up to micrometer lengths; however it will be changing in magnitude and direction over larger areas, which would allow investigation of polarization dynamics as a function of field magnitude and direction in just one exposure. The dependence of polarization dynamics on pulse length from 100 ps down to 100 fs can be studied by changing the compression of the electron bunch. These experiments will determine the ultimate switching speed of ferroelectric devices.

References:

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