Ultrafast x-rays: radiographing magnetism

Project overview

The main purpose of the proposed postdoctoral fellowship is to gain unique training and expertise in time-resolved x-ray spectroscopy at the picoseconds (and eventually femtosecond) scales, focusing on magnetic materials. Such expertise is considered to be of strategic importance from a national point of view, with respect to the research planned at MaxIV.

Since their discovery in 1895 by Röntgen, x-rays have played a fundamental role in science and society. Probably, x-rays' most renown application is in radiography, which allows looking through the soft tissues of the human body. Furthermore, x-rays have always been and continue to be an invaluable tool in the field of physics. Most of the fundamental properties of matter, such as atomic composition, crystal structure, valence state, magnetic configuration, can all be retrieved using x-rays. That not being enough, recent developments at synchrotron light sources has allowed for a quantum leap in x-ray measurements. Indeed, it is now possible to achieve x-ray pulses that are a few femtoseconds long and that are focused within a few nanometers, thus allowing for an entirely new set of opportunities in condensed matter physics.

The driving idea behind the proposed fellowship is to apply time-resolved x-ray circular magnetic dichrosim (XMCD) to investigate the magnetic properties of two peculiar magnetic systems: a magnetic transition metal and a colossal magneto-resistive manganite. By choosing two different materials, the proposed fellowship can be divided in two phases, each of them roughly corresponding to one year of training.

The proposed host institution is Stanford PULSE at SLAC, and the principal investigator is Dr. Hermann Dürr, head of the ultrafast magnetism group. Stanford PULSE is a worldwide renowned centre for ultrafast science, and Dr. Hermann Dürr published, over the past few years, breakthrough results in the field of ultrafast magnetization dynamics, using x-rays as the investigation tool [1–4].

Phase 1: Spin wave dynamics in spin torque oscillators.

When a thin (few nm) metallic magnetic film is subjected to a high-density spin-polarized current (typically injected via a lithographically defined nano-contact), the excitation of highly non-linear spin waves in the magnetic film can take place. The mechanism behind this phenomenon is the spin transfer torque (STT), i.e. the transfer of magnetic moment from the spin-polarized current to the magnetic thin film. STT is currently a very active research topic in the magnetism community, since it is the basis of a potential new type of commercial non-volatile random access memory (STT-RAM). Despite a large number of achievements, a very fundamental question has not been confirmed experimentally: *what is the wavelength of the excited spin waves?* The answer to this question is not only of fundamental importance, but it has profound implications in applications related to spin torque. In this fellowship, we propose the use of short (10 – 50 ps FWHM) x-rays pulses focused down to 20 nm to create a time and space resolved map of the spin wave excited in a nano-contact spin torque oscillator. The typical excitation frequencies (in the GHz regime) and the theoretically predicted wavelengths (several hundreds of nm) are within the capabilities of the measurement technique.

Since the study of STT spin wave dynamics was the investigation topic of the applicant's Ph.D. thesis,

almost all of the efforts during the first year of the fellowship will be dedicated to the training at the synchrotron, including the time to perform the proposed experiment. This favourable situation, for which the only novelty of the first year of the fellowship will be the measurement technique, makes the realization of a second, distinct phase a realistically achievable goal in the second year of the fellowship.

Phase2: Photo-induced metal-insulator transitions in manganites

In 1986, the discovery of high temperature superconductivity [5] in ceramic oxides represented a breakthrough for the scientific community. As a consequence of this discovery, a great deal of research effort has been made towards understanding the properties of transition metal oxides (TMOs). However, despite this large effort, the fundamental physics behind the exotic properties of such oxides is still poorly understood [6, 7]. In the case of complex transition metal oxides, the interplay between the spin, charge and lattice degrees of freedom can have a fundamental impact on a wide range of the physical properties observed in this class of materials. A well known example are the manganites showing colossal magneto-resistance (CMR) behaviour [6, 7], where one can observe a phase transition from a ferromagnetic metallic state to a non-magnetic insulating state. Such a transition can be caused, for instance, by temperature, stress, or external magnetic fields. Most of these studies have been performed considering the "quasi-static" response of manganites system. However, in recent years, the possibility of ultra-fast spectroscopy has shown that a plethora of new effects can be observed when energy is deposed into the materials under very short time scales: photo-induced ultra-fast metal insulator transitions [8], melting of charge and orbital order [9], and emergence of coherent orbital waves [10] and of a transient "hidden" phase [11].

In this second phase, we propose the use of ultra-short (down to few ps achieved in the unique SSRL low alpha mode operation) x-ray pulses to study the time-domain evolution of a photo-induced metal-insulator transition in a manganite showing CMR effect, namely $La_{1-x}Sr_xMnO_3$ (LSMO). First, we want to look at time-evolution of the material when the system, pumped into the insulating non-magnetic state, relaxes back into the ferromagnetic conducting state. This can be achieved with x-rays in resonance with the oxygen *k*-edge where the evolution of the electronic band gap can be probed directly [4]. Second, we want to visualize the time evolution of the formation of ferromagnetic domains when the phase transition occurs. Since the appearance of ferromagnetic domains is related to the appearance of charge carriers, the visualization of such domain is expected to allow us to understand details of the metal-insulator transition that are still obscure. Here we use circular x-ray dichroism at the transition metal L-edges combined with high resolution imaging techniques.

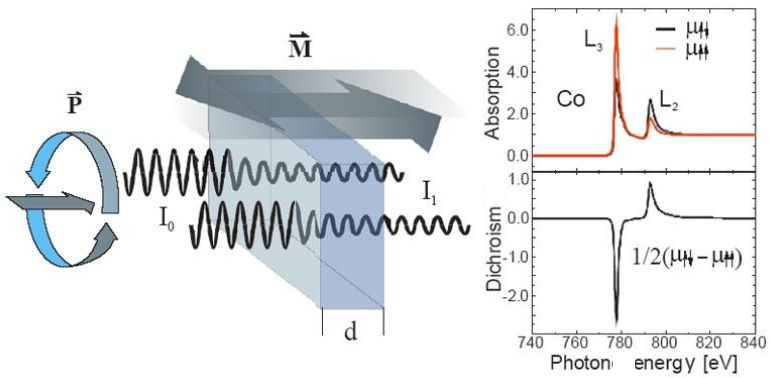

Figure 1. (Left) Attenuation of circularly polarized x-rays, dependent on sample magnetization. (Right) Absorption the Co $L_{2,3}$ edges for magnetization parallel and anti-parallel to the circular polarized x-ray beam. The lower part is the difference spectrum, i.e. the XMCD effect. [From Max Planck Institute for Intelligent Systems webpage]

Scientific case

Phase 1: Spin wave dynamics in spin torque oscillators.

In the first phase of the fellowship, the aim will be to determine the wavelength of the spin waves excited by spin transfer torque (STT) in nano-contact spin torque oscillators. Despite its fundamental nature, this question has never been answered experimentally after 15 years of research. The theoretical calculations that initiated the research in spin torque [12] assumed that the spin waves generated by spin transfer torque are exchange dominated and that they have a wavelength dependent solely on the nano-contact radius implying that they possess a cylindrical symmetry. However, recent works show that reality may be remarkably different. The Oersted field generated by the current flowing through the nano-contact breaks the cylindrical symmetry, and leads to an asymmetric propagation, according to micromagnetic simulations [13]. Furthermore, the contribution of the dipolar field to the spin wave energy spectrum may be even larger than the exchange term, for most nano-contact sizes of interest [14]. This may have important effects on the wavelength of the excited spin waves.

Time and space-resolved x-rays can be the tool that will allow for a definitive understanding of these questions. Indeed, with dynamics in the GHz range and with expected wavelengths in the hundreds of nm range, STT induced spin waves are within the capabilities of x-ray pulses generated at SLAC. The experiment we propose is based on the measurement of the x-ray magnetic circular dichroism (XMCD), i.e. the difference in the absorption spectra between left and right circularly polarized x-rays. The XMCD signal is directly proportional to the magnetization, and this can be used to determine the wavelength of the spin wave.

The idea of the measurement is as follows. We will phase lock the spin torque oscillator to an external microwave source. The dynamics will still be excited by the spin polarized direct current, but with the phase locking we will achieve a precise control over the phase of the oscillation. Now, by using time-resolved scanning transmission x-ray microscopy (STXM), we can retrieve both the temporal and spatial characteristics of the emitted spin wave, leading to a reliable extraction of the wavelength. The highly non-linear spin wave dynamics in spin torque oscillators leads to a large angle magnetization precession in the magnetic material. Hence, the peaks and the nodes of the spin wave amplitude will give rise to a measurable XMCD contrast. A STXM instrument exactly for this application is available at SSRL beam line 13. The SLAC group is using the instrument for current induced switching of nano-pillar structures. The proposed project will be able to utilize the instrumentation and provides an interesting extension of these activities.

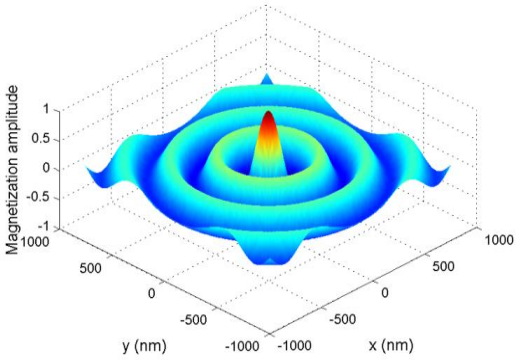

Fig. 2 Theoretical calculations of the magnetization amplitude in a spin torque oscillator, in the plane of the excited magnetic film. The nano-contact radius is 50 nm, and the expected wavelength about 300 nm.

The expected results are of fundamental importance for the research in magnetism. In fact, much of the current effort in the investigation of spin transfer torque dynamics focuses on the interaction between spin waves. Hence, the knowledge of the spin wave wavelength is a fundamental property that needs to be known, if one wants to predict and control more complex structures and devices. Also, this knowledge is particularly relevant for the emerging field of magnonics. Here, the modulation of the magnetic properties in a "magnetic lattice" affects the spin waves in analogous way as the refraction index affect the wavelength of light in photonic crystals.

Phase2: Photo-induced metal-insulator transitions in manganites

In the second phase of the fellowship, the aim will be to observe the metal-insulator transition in the manganite perovskite $La_{1-x}Sr_xMnO_3$ (LSMO), with $0.17 < x < 0.25$. At this stoichiometry, the system shows, at about room temperature, a phase boundary between a ferromagnetic metallic state and a paramagnetic insulating state. There is much interest around LSMO with this stochiometry because of its high Curie temperature and because it is supposedly a half metal, hence it could be used ideally as a 100% spin polarizer. Although in past years this phase transition has been subject of thorough investigation, its details are still poorly understood.

The difficulty in finding a deep understanding is due to the complexity induced by competing interactions between electrons, lattice and spins. This complexity can be disentangled in the time domain, since different interactions may have different characteristic time scales. For instance, laser heating drives the electronic system out of equilibrium nearly instantaneous. Then, within 1-2 ps, energy is transferred to the lattice via electron-phonon coupling. In magnets one also needs to consider the angular momentum. Typically ultrafast demagnetization in half metals happens slowly [15]. The question now is: *when does a metal become an insulator?* Is this happening on the same timescale as the ferromagnetic to paramagnetic phase transition or not? This information will shed light on the microscopic origin of magnetoresistance.

Ultrafast x-rays may allow these two processes to be studied separately, and hence allow for an improved understanding of this phase transition. The idea we propose in this fellowship is the following. At first, we will prepare a thin LSMO film with a composition that shows a ferromagnetic conducting state at room temperature. Then, we will deposit heat in the sample by means of a femtosecond laser pulse, which will lead the system into a non-magnetic insulating state. Finally, using x-ray pulses, we will probe the evolution of the oxygen *k*-edge as the system relaxes back to the magnetic metallic state. A recent work on photo-induced metal-insulator transitions in magnetite [4] suggests that such phase transitions may occur with dynamics at the ps time scales, and therefore observable with the time resolution available in x-ray pulses at the SLAC synchrotron. If measurements will show that faster dynamics are taking place, we will apply for beam time at the LINAC free electron laser, where x-ray pulses at the fs scale can be generated.

Finally, as an extra project in case time allows, we will try to look at the formation of ferromagnetic domains as the system relaxes back to the ferromagnetic state. In order to do this, we will use the differential holographic encoding technique recently developed at Stanford, and described in Ref. 16. With this technique, single shot coherent x-ray pulses can be used to recreate patterns in the sample down to a resolution of 16 nm.

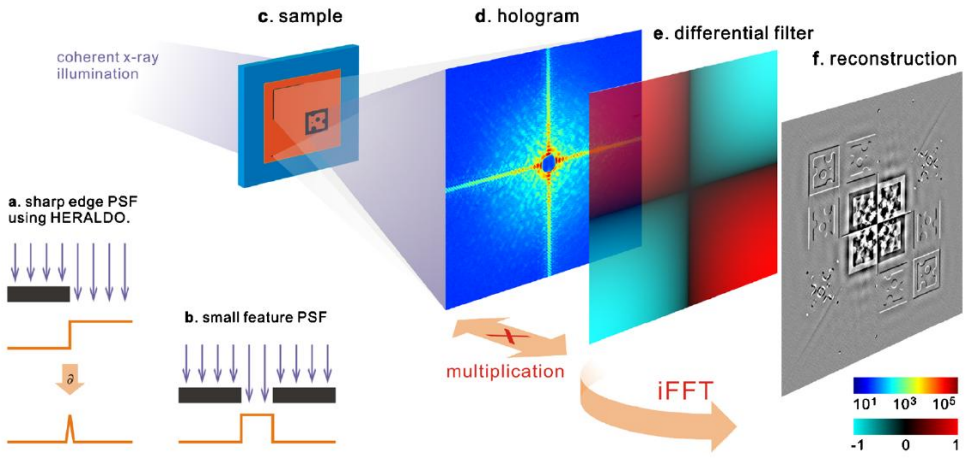

Figure 4 Schematic of the high resolution x-ray lensless imaging using the differential holographic encoding technique described in Ref. [16]

Experimental Methods

Synchrotron

The synchrotron that will be mainly utilized during the fellowship is the Stanford Synchrotron Radiation Lightsource (SSRL), which is one of the user facilities available at the SLAC National Accelerator Laboratory. The machine currently operating is the last upgrade of the synchrotron, and it goes under the name SPEAR3. It is a 234 m long ring, with beam energy of 3 GeV and an operational beam current which may be varied between 300 and 500 mA, and with a horizontal emittance of 10 nm rad.

The x-ray pulses currently generated at SSRL have a duration of 50 ps in normal operation and can be reduced to several ps (how many is part of active research, with a record value of 2.5 ps so far) in the so called low alpha mode [17]. In all the proposed experiments, we will measure XMCD (hence x-ray absorption) in transmission geometry. Samples will be grown on thin substrates, where the attenuation level of x-rays is still within an acceptable range. More details will be given below.

Phase 1: Spin wave dynamics in spin torque oscillators.

Sample fabrication

Nano-contact spin torque oscillators are typically made of a stack of metallic thin films, two of which are magnetic. One thicker magnetic layer (e.g. CoFe, 20 nm thick) is usually chosen as a reference layer, responsible for the spin polarization of the current, and a thinner magnetic layer with low Gilbert damping (e.g. NiFe, 5 nm thick) is the layer where spin waves can be excited. Because of their relative long wavelength compared to the atomic lattice, spin waves in soft magnetic materials are not sensitive to the crystal structure. Therefore, the thin film stack is typically polycrystalline and sputtered on non-lattice matched substrates (e.g. Si). This is a great advantage for the proposed experiment, since the substrate needs to be as transparent as possible to x-rays, and hence the choice of possible materials is restricted. The most straightforward option is to use commercially available Si_3N_4 membranes, which are the standard choice for scanning transmission x-ray microscopy, and which have thickness from a few hundred nm down to a few nm. (Alternatively, one may think to deposit a layer of a suitable material on top of a standard Si substrate, lithographically define openings on the back side of the substrate, and finally selectively etch away the silicon from the back side.)

After the materials deposition, the nano-contact and the electrodes are defined with a combination of electronic and optical lithography, dry and wet etching, and metal evaporation. The samples are available at Stanford, and there is interest in fabricating new samples in collaboration with the Materials Physics department at the Royal Institute of technology (KTH), where the applicant performed his Ph.D. studies.

Measurement details

In order to excite spin waves by spin transfer torque, one needs to provide a bias direct current to the sample. To do that, we will wire bond the sample to a carrier structure, a printed circuit board to which macroscopic wire can be connected and biased by a current source.

As previously mentioned, we want to create a spatial map of the spin waves. In order to do this, we will perform XMCD measurements in scanning transmission x-ray microscopy (STXM) configuration, recently implemented to image the spin torque induced vortex motion in a nano-magnet [18]. In order to perform such experiment, the periodicity of the spin wave pattern must be well defined over the entire experiment.

This will be achieved by locking the sample to a microwave source, so that the phase of the oscillation is well defined and the timing with the x-ray pulses can be determined accurately.

Phase2: Photo-induced metal-insulator transitions in manganites

Sample fabrication

In contrast to the experiment proposed in first phase, the investigation of metal-insulator transitions in LSMO requires samples of high crystalline quality. The growth of high quality crystals is a complex problem when the lattices of the substrate and of the oxide film are not matched. Since even in this case we will measure in transmission geometry, we will again need a substrate which is as transparent as possible to x-rays. Therefore, in order to fabricate samples that can be measured by XMCD, we will follow two approaches. At first, lattice-matched substrates will be back-etched using a combination of etching processes, including the focused ion beam (FIB) milling system available in Stanford's clean room. In the long run, samples will be grown directly on silicon nitride membranes, using suitable seed layers that are currently being investigated.

The fabrication activity has recently started at the hosting institution in collaboration with Harold Huang's group (also at Stanford), one of the world leading laboratories in growing manganites. High quality crystalline LSMO thin films will be grown by pulsed laser deposition (PLD) by Huang's group, and all the subsequent fabrication process will be carried out using Stanford clean room facilities in collaboration with that group.

Measurement details

Femtosecond laser pump – ps x-ray probe technique

The main measurement tool for the second phase of the fellowship will be a fs optical laser synchronized with the SSRL operating at 1.28MHz repetition rate.

X-ray free electron laser

The x-ray free electron laser (X-FEL) that is eventually planned to be used in the fellowship is the Linear Coherent Light Source (LCLS). The X-FEL is also installed at the SLAC National Accelerator Laboratory [19]. At present, the energy of the x-ray can be tuned from 540 eV to 9 keV. The x-ray pulse has typical energies of 2.5 mJ, corresponding to 10^{12} – 10^{13} photons per pulse (energy dependent). The pulse length can be tuned to a few tens of fs to hundreds of fs, and the tuning capability also depends on the chosen photon energy.

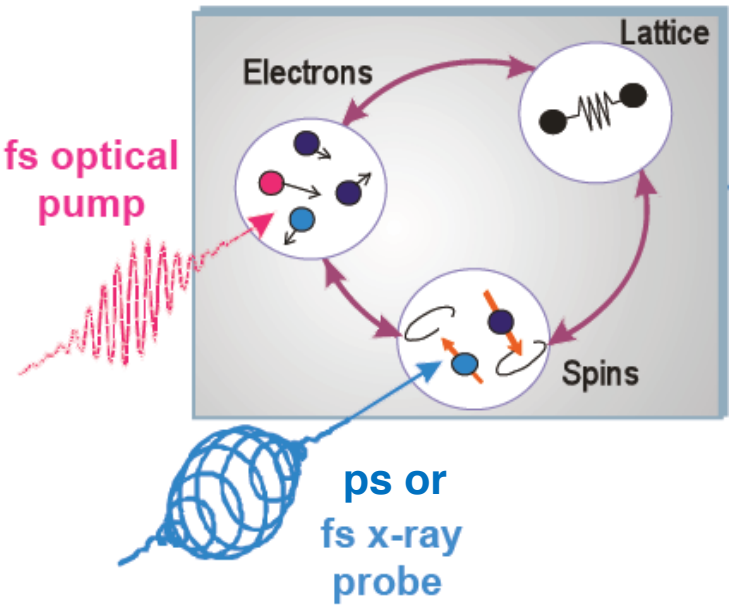

Figure 5 Pictorial view of the pump – probe experiment proposed in the second phase of the project. A fs optical pump pulse deposit excites electrons in the CMR manganite LSMO. The electronic excitation also affects the lattice and the spin degrees of freedom, and the spin excitation is probed by a circularly polarized x-ray pulse.

References

[1] C. Stamm *et al*, Nature Mater. **6**, 740 (2007)

[2] C. Boeglin *et al*, Nature **465**, 458 (2010)

[3] C. Stamm *et al*, Phys. Rev. B. **81**, 104425 (2010)

[4] N. Pontius *et al*, Appl. Phys. Lett. **98**, 182504 (2011)

[5] J.G. Bednorz and K.A. Mueller, Z. Phys. B **64**, 189 (1986)

[6] M. Imada *et al*, Rev. Mod. Phys. **70**, 1039 (1998)

[7] M. B. Salamon and M. Jaime, Rev. Mod. Phys. **73**, 583 (2001)

[8] K. Myiano *et al*, Phys. Rev. Lett. **78**, 4257 (1997)

[9] P. Beaud *et al*, Phys. Rev. Lett. **103**, 155702 (2009)

[10] D. Polli *et al*, Nature Mater. **6**, 643 (2007)

[11] H. Ichikawa *et al*, Nature Mater. **10**, 101 (2011)

[12] J. Slonczewski, J. Magn. Magn. Mat. **159**, L1(1996); L. Berger, Phys Rev. B **54**, 9353 (1996)

[13] M. A. Hoefer *et al*, Phys. Rev. B **77**, 144401 (2008). S. Bonetti *et al*, 55th conference on Magnetism and Magnetic Materials, November 14-18, 2010, Atlanta, GA.

[14] S. Bonetti and M. Madami *et al*, Nature Nanotechnology (2011). doi: 10.1038/nnano.2011.140

[15] Georg M. Müller *et al*, Nature Mater. **8**, 56 (2009).

[16] D. Zhu *et al*, Phys. Rev. Lett. **105**, 043901 (2010)

[17] J.Corbett, W.Cheng and X. Huang (SLAC), Short bunch measurements in SPEAR3,Proceedings of PAC09, Vancouver, BC, Canada (2009).

[18] X. Yu et al, Phys. Rev. Lett. **106**, 167202 (2011)

[19] http://home.slac.stanford.edu/pressreleases/2009/20090421.htm