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Magnetization Dynamic with Pulsed X Rays

Christine Boeglin
Institut de Physique et de Chimie de Strasbourg, Université de Strasbourg, France

1. Introduction

Lasers have become more and more useful and a large field of application is nowadays reached including medicine, biology but also fundamental research as physics for instance. It is also in the fundamental research area that recently a fast developing new field is growing: Ultra-short high-energy pulsed X rays. Compared with the lasers community where first technological developments were recently achieved [Spi1997, Dre2001 Schn1999, Kra2009] in order to reach higher energies (5-100 eV), the X-ray community is using high energy X rays from large facilities, for instance the synchrotron storage ring facilities were a large UV and X-ray energy range is produced but were time resolved spectroscopy is only starting since a few years [Sch2000, Scho2000, Hol2005]. It is my aim here to describe the actual state of the art in the field of X rays and especially concerning the different X-ray pulse length and intensities. In the second part I will develop the application in the field of magnetism of the time resolved X-ray spectroscopy and microscopy.
The description of the High-energy X-ray pulse section (2.) will include technical details about the energy range of the X rays, the different time resolution and density of photons produced in the facilities as Synchrotron and X-ray Free electron lasers (X-FEL). The f-slicing possibilities at BESSY (Germany) and also the X-FEL facilities in Europe and in USA will be developed. The recently launched free-electron laser at the FLASH facility in Hamburg and LCLS in Stanford are the two first free electron sources in the world.
Description and discussion of applications using the pulsed X-ray sources are given in section (3.) and will introduce some of the actual motivations in the field of ultrafast magnetization dynamics using ultrafast X-ray pulses. It is divide into two sub-sections; one concerning the spectroscopies performed using the time structures of X rays and the second the time resolved imaging techniques actually developed in the world.

2. Time resolved spectroscopy's using the temporal structure of X rays

In recent years, magnetism at ultrafast time scales has been a growing topic of interest. A thorough understanding of femtosecond magnetism will address the important questions of how fast the magnetization can be reoriented in a material and what physical processes are behind and limits to this speed. In the spatial domain, magnetism at nanometer length is a topic directly relevant to data storage, since future advances in this technology will require a further reduction in device dimensions to increase the storage density. These

considerations have motivated a variety of studies using magnetooptic effects in conjunction with ultrafast light pulses to explore these fundamental limits. These studies currently make use of visible-wavelength light from ultrafast lasers, or X-rays from large-scale synchrotron X-ray facilities. Ultrafast lasers produce short pulses (~30 fs), making possible femtosecond time resolution [Beau1996, Cin2006], but with a spatial resolution that is generally limited by the wavelength of the probe light. X rays, on the other hand, allow for high spatial resolution and high contrast imaging at the elemental absorption edges of ferromagnetic materials. However, the available time resolution to date is too slow to resolve the fastest dynamics. Because of this, significant efforts have been devoted to using short or isolated electron bunches of X rays pulses at synchrotron to perform time resolved microscopy with X rays. More recently femtosecond strong laser pulses are used to slice short burst (100 fs) of X rays from synchrotron radiation [Stam2007, Boe2010]. Magnetic imaging techniques as for instance X-ray PhotoEmission Electron Microscopy (X-PEEM), Scanning Transmission X-Ray Microscopy (STXM) or X-ray Resonant Elastic Scattering (XRES), are currently using the short X ray pulses in order to accede to time resolved imaging in the picosecond time range. Unfortunately, the f-slicing technique in synchrotrons produces a strongly reduced photon flux hindering the f-second magnetic imaging at facilities as synchrotrons.

2.1 Magnetic imaging using the ps time structure of the synchrotron

2.1.1 Magnetic domains and vortices under magnetic field pulse excitations

In order to move magnetic domains one of the simplest way one can think of is to apply a short magnetic field pulse perpendicular to the magnetization. In this way the field will exert a torque on the sample magnetization vector and induce a rotation of the spins. In a second step the out of equilibrium spins will start to relax in order to transfer the energy from the external field to the lattice, by characteristic precession and damping mechanism. Many experimental description of this process in soft and hard magnetic materials were performed aiming to model the dynamic of relaxation mechanisms in the pico and nanosecond time ranges. Even if the simple idea of a magnetic field pulse excitation is straight forward compared with electronic excitations, in practice this method suffers from the difficulties to produce strong and short magnetic pulses as well as sharp on and off sets (rise times) of the magnetic pulses. Several methods for the generation of magnetic field pulses have been used. Electrical pulse generators for instance (limited by the self-inductance of the electric circuit) with rise times of more than 100 ps and further lithography "stripe lines" were developed in order to reduce the rise times [Ele1996]. Further improvements of the rise-time was archived using optical switches, which can be optically controlled and which are based on lithography fabricated photoconductive " Austin" switches (based on metal-GaAs-metal junctions) [Ger2002] or alternatively "Schottky diodes" switches (based on metal-semiconductor junctions) [Acre2001)]. Beside the large ~50 ps rise times a second limitation is the low induced magnetic fields (~0.1 T) produced by the set-up at the sample location. This often limits the experiments to soft material as permalloy and soft CoFe alloy films (Fig1). Such systems where extensively studied in the past 10 years focusing on reduced dimensions in nanostructures and lithography designed vortices structures. [Cho2004, Schne2004, Raa2005, Weg2007, Kras2005, Kuc2004, Vog2005, Vogel 2005, Fuk2006, Vog2008, Hey2010, Uhl2011]

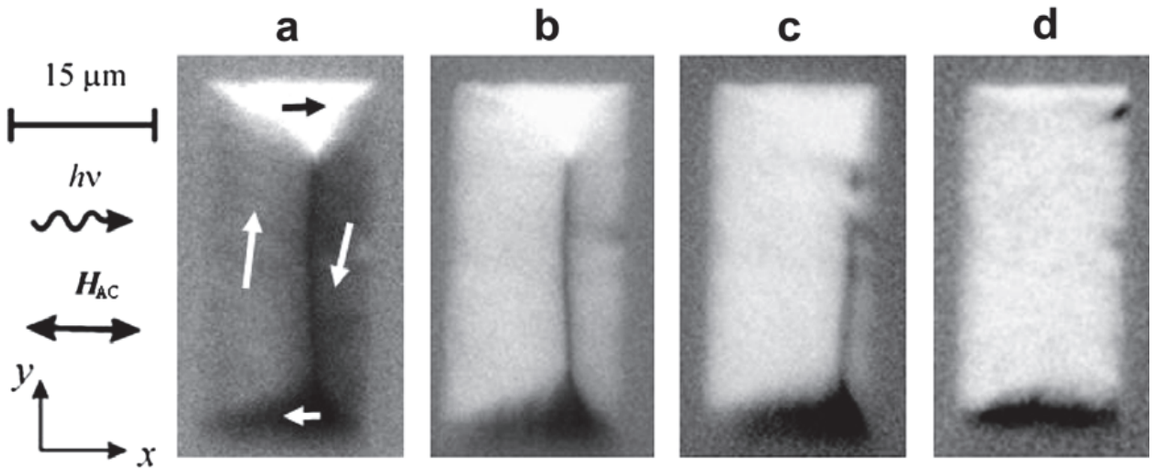


Fig. 1. Magnetic response of the x-component of the magnetization (bright areas are magnetized to the right, dark areas to the left) in a permalloy platelet of 16 ·32 μm2 size and 10 nm thickness for three different field amplitudes I (1.5 Oe), II (2.0 Oe) and III (2.5 Oe). (a) XMCD‐PEEM snapshot of the domain pattern in dynamic mode at excitation amplitude I; arrows denote the local magnetization direction. (b)‐(d) Snapshots of magnetic domain patterns at maximum magnetic response excited with increasing amplitudes. [Weg2007]

Furthermore, extremely large effective magnetic field pulses can be produced by femtosecond laser pulses combined with the heating of an exchange-biased system. Recently it was suggested that ultrafast switching could be induced via laser-induced reorientation of an exchange coupled antiferromagnet such as $TmFeO_3$ [Kim2005]. A strong magnetic field pulse has also be generated by a relativistic electron bunch combining short duration of 1ps and high field strength ~100 Tesla [Stam2005]. The counterpart of such experiments is that it is accompanied by a strong electric field. Up to now, no time resolved study using a pump-probe set-up has been archived using these high magnetic field pulses.

Time-resolved scanning transmission X-ray microscopy (STXM) in NiFe thin films was studied in order to define the role of domain wall pinning on the dynamic behavior of magnetic vortex structures [Van 2008]. The X-ray magnetic circular dichroism (XMCD) effect, was used as contrast mechanism for the imaging of the structures (Fig 2). In contrast with the X-PEEM, the STXM geometry is sensitive to the projection of the magnetization along the photon propagation direction; therefore, the in-plane magnetized sample was tilted over 60° with respect to the incoming photon beam in order to observe the magnetization. A full image can be constructed by scanning the sample along both in-plane directions. The lateral resolution is determined by the zone plate of the beam line and is about 30 nm. Time-resolved measurements were performed in order to investigate the dynamic behavior in magnetic vortex structures. The natural time structure in the storage ring of the synchrotron delivers photon flashes every 2 ns in the so-called multibunch mode. This allows the experiment to follow a typical pump-and-probe scheme, with the incoming photon flashes as probe and the externally applied in-plane magnetic field pulses as pump. The magnetic structures were repeatedly excited every 82 ns by sending an electric current in the stripline underneath the structures. The current pulses induce magnetic field pulses with amplitudes of about 10 mT and a full width at half maximum of about 1 ns (500 ps of

rise and falling time). The excitation was synchronized with the X-ray flashes of the synchrotron, which probe the magnetization at different times *t* after the pump. The analysis of the dynamic behavior of the vortex gyration frequency show that they are increased in square-shaped structures, where domain walls are present suggesting that the domain wall pinning is causing the increased frequency.

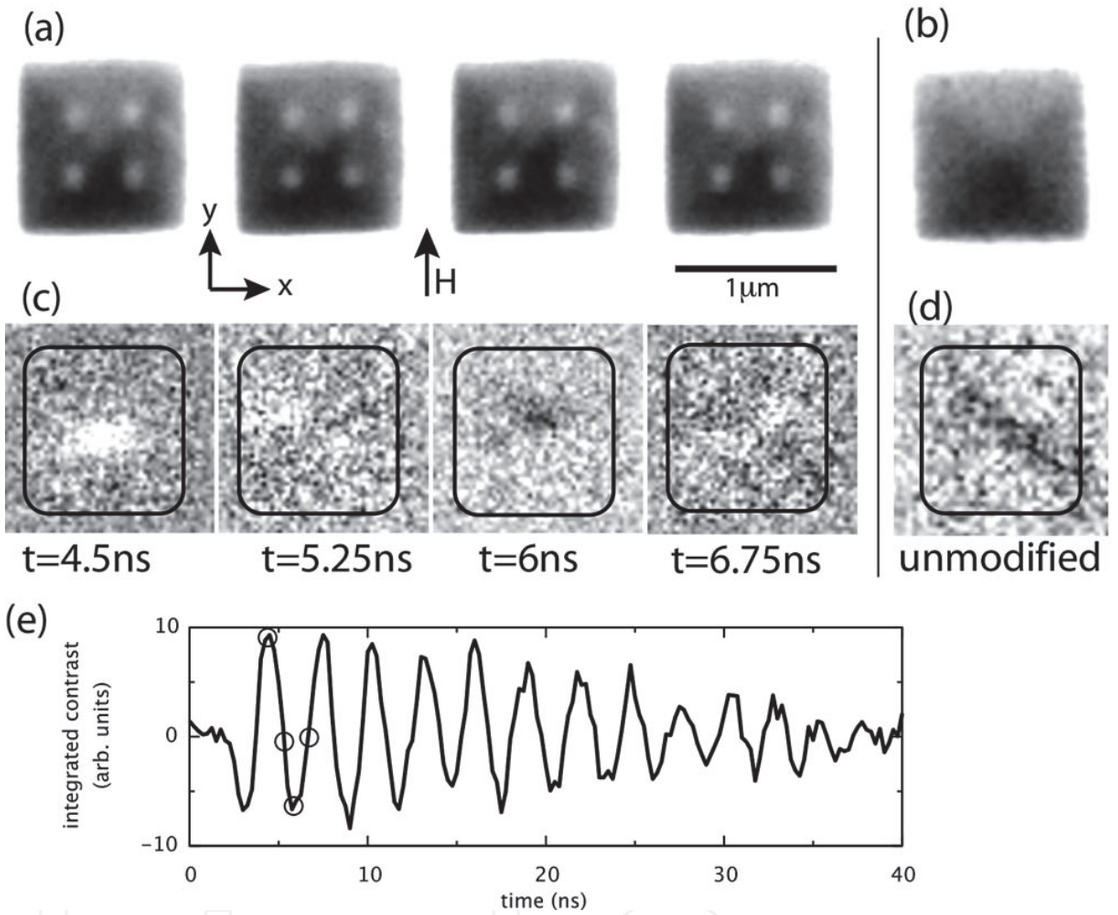


Fig. 2. (a) Sequence of STXM images for a 1 μm x 1 μm x 50 nm modified square during one period of the oscillation. (b) image of an unmodified structure and shows that the domain wall motion can span a larger area of the structure when no defects are created. The intensity is proportional to the *x* component of the magnetization, revealing the Landau configuration and the small gyrotropic motion of the vortex structure. The total contrast in this sequence oscillates with the resonance frequency, as shown in (e). The four STXM images in (a) and (c) correspond to the four markers in (e). The magnetic pulse *H* starts at *t*=2 ns. [Van 2008].

2.1.2 Magnetic domains under femtosecond laser excitation

In order to study the magnetization dynamics in oriented ferromagnetic domains, after a femtosecond pump laser excitation, a precise nanometer scale characterization of the magnetic domain contrast and domain configurations is of great importance. This more recent studied aspect of the space resolved dynamics aims to discribe the influence of a laser

excitation on the magnetic domains in different time ranges (nanosecond, picosecond and femtosecond scales). The characterization of the dynamic of the magnetic domain configurations helps to understand the demagnetization process because it provides a description of the magnetization in space. Using X rays these studies benefit from the chemical sensitivity of the circular polarized X rays and from the high spatial resolution (30 nm) of the magnetic imaging mode of X-PEEM instruments that are nowadays currently working at synchrotron storage rings. Appropriate femtosecond pump laser can easily be implemented one such instruments in order to address thermal effects of the laser pump in the ps range. The ultrafast modifications induced by an infra-red laser pump on the magnetic domain configurations is still unknown. Questions concerning the induced changes in the magnetic contrast in a magnetic domain, the size and shape of the domains are still pending. The typical time resolution of the actuel experiment is ~60 ps using the multibunch mode and 10 ps using the low alpha operation modes currently provided in synchrotron storage rings. The time resolution limitation is strongly related with the limited X-ray flux and with the imaging technique by them self where high flux is mandatory.

One of the interesting subjects today is the study of the dynamics in the picoseconde time range of domain sizes and of the magnetic contrast provided using the X-ray circular magnetic dichroism (XMCD) as a function of the pump probe delay. This can be studied either in in-plane oriented magnetic domains or in perpendicular oriented domains.

Following the excitation of ferromagnetic materials with ultra-short laser pulses, a sequence of relaxation mechanisms takes place. The first one is related to the ultra-fast demagnetization. The second mechanism is related to electron - spin and lattice energy transfer, most important within a few picoseconds after the excitation. This mechanism depends on several parameters: the electron-phonon coupling, the material's specific heat, the magneto-crystalline anisotropies and specific interactions like the ferromagnetic or anti-ferromagnetic coupling. One of the goal is to correlate the results with the laterally averaged spectroscopic information obtained using XMCD time resolved spectroscopy.

The experimental method consists in measuring the FM domain contrast in ferromagnetic materials set into a remanent state. Magnetic imaging in the pump probe configuration is setup using the triggered imaging detection mode to obtain a XMCD contrast image at the Fe, Co, or Ni L_3 edges. Moreover the laser fluence necessary to de-magnetize the films is typically in the order of a few mJ/cm^2 and, in the best cases, this allows achieving complete demagnetization of the films. Using focalization of the laser this can easily be achieved by focusing the laser spot onto a few 10 micronmeter on the sample surface. The XMCD signals are probed in a gated mode at different time delays between the laser pump pulse and the probe pulse of circularly polarized synchrotron radiation. The time resolved magnetic signal is extracted from a time-delay sequence of XMCD images and allows extracting the magnetic components in a semi-quantitative way as a function of time delay. Intensive research in this field is developing using the X-PEEM imaging technique and extension toward other techniques as time-resolved scanning transmission X-ray microscopy (STXM) is expected soon.

2.2 Spectroscopy using slicing techniques or X-FEL pulses

2.2.1 Pump probe with lasers using f -slicing

In order to perform experiments using ultrashort X-ray pulses of only ~100 fs in synchrotrons storage rings one had to modify the large electron bunch time structure of 60-80 ps. This can be performed by using a femtosecond laser pulse to slice the electron bunch. The first generation

of fs X-ray pulses in third generation synchrotron radiation sources was proposed [Zho1996] and experimentally demonstrated at the Advanced Light Source (ALS) in Berkeley [Sch2000, Scho2000] using X-ray radiation from a bend magnet. The first undulator-based facility was constructed and successfully commissioned at BESSY [Holl2005].
Such an installation has been set up at BESSY (Berlin) and also at SLS (Villigen) and consists on a slicing of the electron bunches using a femtosecond infra-red laser [Kah2005]. The source at BESSY is based on laser-induced energy modulation ("femtoslicing") and subsequent angular separation of the short-pulse X rays emitted by an elliptical undulator. The femtosecond X-ray source is thus delivering X-ray pulses of 100 fs (fwhm) duration with tuneable polarization.
The electronic synchronization between the laser pulse and the electron bunches is adjusted so that the electric field of the laser interacts with the bunches at the maximum of the intensity (Fig 3). A specific insertion device names Modulator hosts the laser-electron bunch interaction where the femtosecond laser pulse copropagates with an electron bunch, causing an oscillatory energy modulation of the electrons in the short overlap region. The off-energy electrons are transversely displaced by dispersive elements in order to extract the short component of radiation emitted in a subsequent device (the ”radiator”). The second device (Radiator) deviates the two electron bunches with a different angle, so that the angular separation allows extracting only the short radiation component.
The THz signal is the prime diagnostics tool for optimizing the femtoslicing source, when starting an experiment. In addition to being crucial for diagnostics of the laser-electron interaction, the THz radiation itself is useful for experiments where intense ultrashort THz pulses of well-defined temporal and spectral characteristics are required [Holl2006].

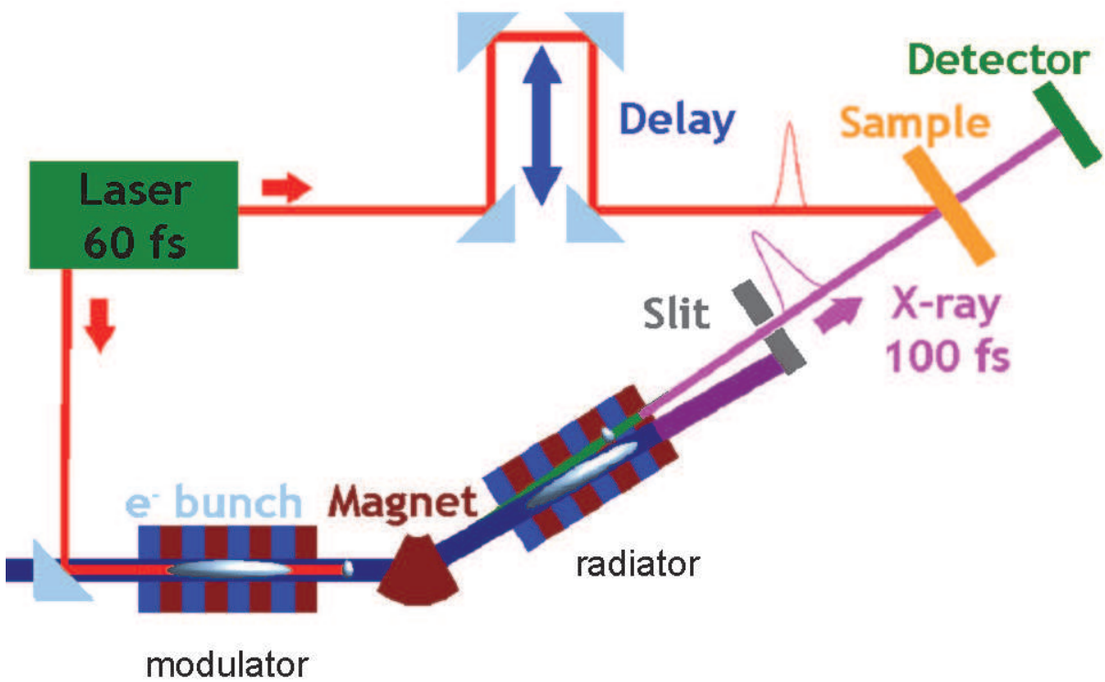


Fig. 3. Slicing experimental geometry : The femtosecond laser is divided into two branches. One is devoted to the slicing inside the ring and the second branche is used to pump the sample at the experimental end station.

The ultrashort X-ray pulses produced by slicing thus provides a strongly reduced flux of 10^4 photons s^{-1} mrad-2mm-2per 0.1% BW, compared to 10^6 photons s^{-1} mrad-2mm-2per 0.1% BW using the single electron bunch. The static measurements using all the bunches we can typically expect at 700 eV a flux of 10^{13} photons s^{-1} mrad-2mm-2per 0.1% BW. The reduction of the flux us thus extremely important when performing time resolved experiments and is in the limit of any experimental set up possibilities when using the sliced beam. This motivates to develop a Bragg-Fresnel zone plate beam line for which an improvement of the photon flux of more than a factor 10 is provided. The energy range of the X rays produced at the beam line at BESSY II ranges from 600 eV to 1400 eV.

The pump probe experiment using such slicing set up are done in a specific pump-probe geometry using the transmitted X rays at the element core level threshold (ex: Fe, Co, Ni L_2 and L_3), as a probe and a femtosecond laser as a pump. They were carried out using the circularly polarized X-ray femtosecond pulses [Sta2007, Boe2010]. The pump pulse with FWHM of 60 ± 5 fs is issued from an amplified Titanium Sapphire oscillator at a central wavelength 790 nm and amplified at 1.5 kHz repetition rate. The pump pulse is synchronized with the sliced electron bunch of the storage ring. The incident X-ray beam is perpendicular to the surface or at 30 degree from the normal and transmitted through the film deposited on a Si_3N_4 membrane. This geometry allows an optimization of the pumping through the film and of the amplitude of the X-ray magnetic circular dichroic (XMCD) amplitudes when performing the time resolved measurements. The transmitted X-ray intensity is measured using a fast Si avalanche photodiode, and a gated boxcar. The measurements are made by making the difference between the transmitted signals obtained for two opposite applied magnetic fields. The spins are then aligned either parallel or anti-parallel to the incoming circularly polarized X rays.

The XMCD contrast is obtained by subtracting the gated signals obtained with and without pump beam. The numerical XMCD values are obtained from the normalized difference of the signals recorded near the edges, for an energy position where the static XMCD signal is maximum. The results are normalized in order to account for the degree of circular polarization of the sliced X rays (70%) as well as the moderate energy resolution of the zone plate. The limited energy resolution of the zone plate (5 eV) used in such experiments ensures that a "integrated signal" over 5 eV is measured and allows us to apply the sum rules and to extract the spin and orbital magnetic moments [Car2009, Boe2010].

2.2.2 Pump-probe experiments using coherent X-FEL pulses

The recent development of ultrashort soft X-ray pulses, as provided by femto-slicing in conventional synchrotron storage rings, or by X-ray free electron lasers, opens today new perspectives in the femtomagnetism field. The free-electron lasers are now operating at Stanford (LCLS- USA) and at Hamburg (FLASH- Germany) producing very short and intense coherent X-ray pulses. The energy ranges at FLASH spreads from 20 eV to 200 eV and at LCLS from 400 eV to 2000 eV. One of the new opportunities at such sources are the pump-probe single shot imaging using the coherence of the source. For this purpose intensive work has been performed in order to define the imaging techniques that will permit to reach not only the ultimate time but also the ultimate space resolution in order to progress in the understanding of ultrafast magnetism.

The ultimate X-ray microscope provides a resolution that is only limited by the wavelength of the radiation. The resolution of STXM, however, is limited by the spot size on the sample.

Much simpler is the image formation process using Fourier transform holography (FTH), where the scattered radiation from the sample interferes with a reference wave and forms a hologram on the detector. Reverse Fourier transform of the measured diffraction pattern yields an unambiguous image of the object. As the phases are encoded in the hologram, several numerical contrast enhancing procedures, can be applied to the image. The spatial resolution in FTH-based methods is limited by the size of the reference aperture-today FTH masks can be routinely produced with reference holes of 30 nm size. However, the image obtained by reverse Fourier transform provides an excellent starting point for a further phase retrieval treatment. In such a way the resolution limitation of FTH can be overcome. FTH is especially attractive in the soft X-ray regime where the photon energy can be tuned to element-specific core level energies allowing for element-specific contrast in the images. This can be used for example to image magnetic domain structures, using X-ray magnetic circular dichroism or for anomalous diffraction imaging.

The recently launched free-electron lasers (FLASH and LCLS) are the first such sources covering the spectral range of relevance for magnetization studies in 3d metals. These novel X-ray sources are able to generate x-ray pulses as short as 10 fs with up to ~10^{12} linearly polarized photons. The short pulse duration, brightness, coherence, and well-defined polarization of the X-ray radiation are the main ingredients that may allow realizing femtosecond single-shot visualization of sub 100 nm magnetic domains [Eis2004].

In particular, it has been shown that if an ultrashort, bright, and coherent X-ray pulse illuminates a sample, the resulting far-field diffraction pattern will encode the image of the sample, from which it can be reconstructed [Eis2004, Chap2006, Gun2011]. The temporal resolution of such a single X-ray pulse snapshot image is then given by the duration of the X-ray pulse (10 fs – 100 fs).

One should remember that such an approach requires not only a very short, but also a very bright X-ray pulse and the large amount of energy deposited into the sample will ultimately turn it into a plasma. Chapman *et al.* demonstrated, however, that the destruction of the sample is not an obstacle for ultrafast "flash diffractive imaging" [Chap2006] as long as the coherent diffraction pattern is 'created' before the sample is destroyed. In order to reach higher energies than the one obtained at the fundamental wavelength (at FLASH -7.97 nm) one can also operated at the fifth harmonic originating from self-amplified stimulated emission at 1.59 nm. Using this operating mode resonant magnetic scattering at FLASH has been performed recently [Gut2009] by using a Co/Pd multilayer sample that was illuminated with 20-fs-long soft X-ray pulses tuned to the Co L_3 absorption edge at 778.1 eV.

More recently, Gutt *et al.* have applied the idea of ultrafast "flash diffractive imaging" to magnetic studies [Gut2010] performing a single-pulse resonant magnetic scattering experiments (Fig 4). By tuning the wavelength to one of the magnetically dichroic absorption resonances of cobalt (the M_3 edge around 60 eV in their case), one may achieve substantially different absorption of polarized X rays in the domains with different orientation of spins. Therefore, coherent and polarized X rays will diffract from such a sample and a far-field diffraction pattern will be formed. The authors performed a simple analysis of this pattern, being able to extract information about the size distribution of the magnetic domains. Due to the linear polarization of the FEL light, however, Gutt *et al.* did not obtain a real image of the magnetic domains. Nevertheless, the technique clearly demonstrates its ability to probe sub 100 nm magnetic domains with a single fs X-ray pulse.

In the near future, we anticipate that such a technique, in combination with further development of 4th generation synchrotron sources will dramatically improve our understanding of ultrafast magnetization dynamics and femtosecond laser control of magnetism. The advantage of such X-ray sources for our purpose is the high X-ray peak power, the very short pulse duration (down to ~10 fs), the high coherence and the tenability of the X-ray photon energy.

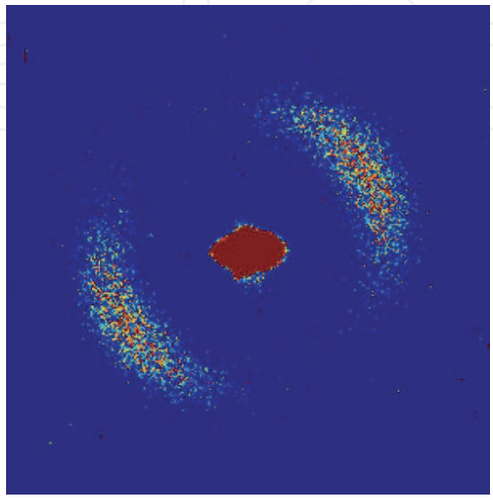


Fig. 4. CCD image of the magnetic diffraction pattern recorded with soft X-ray radiation at Co L_3 edge, using the fifth harmonic at FLASH (photon energy of 778.1 eV) [Gutt2009].

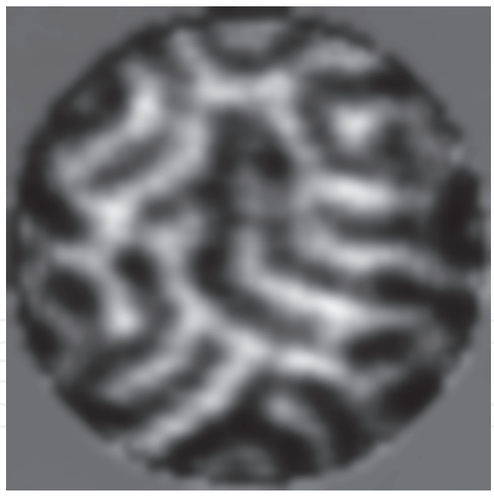


Fig. 5. Snapshot FTH image of a magnetic worm domain pattern recorded with a single LCLS X-ray pulse employing XMCD at the Co L_3 edge as contrast. The circular field of view is 1.5 μm in diameter [Lün2011].

Using the higher energies of the free-electron lasers at Stanford (LCLS) J. Lüning et al. [Lün2011] performed similar experiments on CoPd alloys using the spectral range of relevance (CoL_3). They achieved for the first time single shot magnetic images using HTF at the CoL_3 edge. The real images of the magnetic domains in CoPd alloys were obtained using monocromatized X rays of 100 fs duration, obtained by using a single circularly polarized X-

ray pulse of 10^{11} photons. The polarization was obtained by focusing the X rays thought a thin Cobalt ferromagnetic film. Different delays in the pump probe experiment give theme a serie of real magnetic domain images and reveal that 1 ps after the IR laser excitation the magnetic domains in CoPd only changes in contrast but not in shape (Fig 5).

3. Ultrafast magnetization dynamics on the nanoscale

3.1 Magnetization dynamics in magnetic solids

In solids the magnetization react upon external disturbances as for instance temperature, external magnetic field pulse or pulsed magnetic or electric fields. The induced changes in the magnetization shows different time scales, and different characteristic length scales and sizes for the magnetic structures, domains and domain walls, leading to intense work in this research field during the last decades.

Since the development of the magneto-optics using pulsed lasers has opened a new field of research named ultrafast magnetization dynamics many different experimental and theoretical work was performed. All this work concentrate on pump probe experiments were fs laser excite the ground state in ferromagnets. The development of this field was unambiguously correlated with the ability to perform time resolved spectroscopy below 1 ps which is the range of interest because they naturally corresponds to important magnetic energies, as given by the time-energy correlation $t = \hbar/E$ which links the cycle in time t to a characteristic energy E. For 3d elements this leads to characteristic times of a few ns for anisotropy energies in the 10^{-6} - 10^{-3} eV range, of a few ps for spin-orbit energies in the 10^{-2} - 10^{-1} eV range, and of a few fs for the inter-atomic exchange energy of $\sim 5.10^{-1}$ eV.

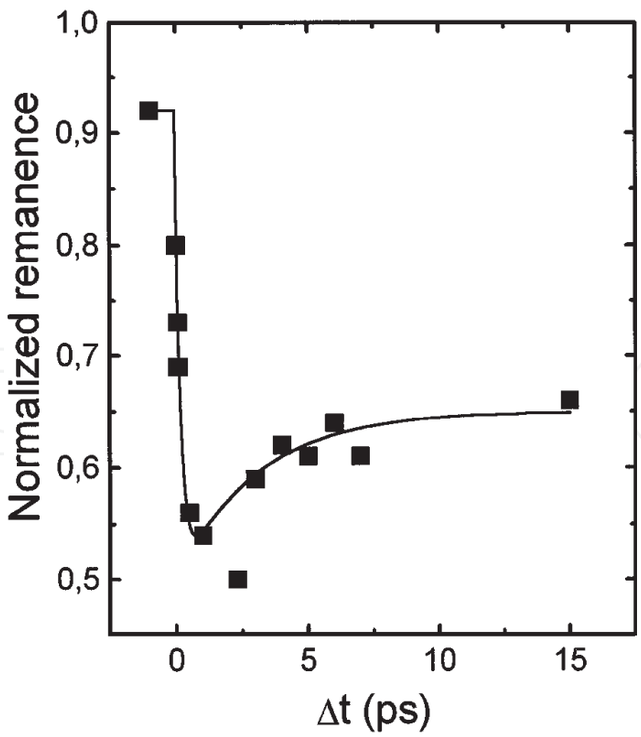


Fig. 6. Transient remanent longitudinal MOKE signal of a Ni(20 nm)/MgF_2(100 nm) film for 7 mJ /cm^2 pump fluence. [Bea1996]

From the discovery of subpicosecond demagnetization over a decade ago [Beau1996] (Fig.6) to coherent interactions between laser and spins [Zha2000, Zha 2008, Big2009] and to the recent demonstration of magnetization reversal by a single laser pulse [Stan2007], the manipulation of magnetic order by ultrashort laser pulses has become a fundamentally chanllenging topic with a potentially high impact for future spintronics, data storage and manipulation.

The recent development of ultrashort soft X-ray pulses, as provided by femto-slicing in conventional synchrotron storage rings, or by X-ray free electron lasers, opens today new perspectives in the femtomagnetism field. Indeed, thanks to the use of sum rules, time resolved XMCD might be viewed as a quantitative measurement of dynamical magnetism, allowing an unambiguous assessment of the magnetization relaxation time, thus confirming previous magneto-optical measurements.

In this context a new milestone has been set by Boeglin et al. [Boe2010] who observed, using time resolved X rays, how ultrashort laser light pulses modify the orbital angular momentum of electrons before it is transfered to the spins (Fig.7). By disentangling the changes in these two components the Strasbourg group showed that spin-orbit coupling can be manipulated on the femtosecond time scale before any lattice or structural transformations occur. The dynamics of spin and orbit angular moments were measured separately in out of plane oriented CoPd alloys, were different orbital and spin dynamics was evidenced [Boe2010]. This work shows that the projections along the easy magnetization axis z the orbital moment is faster by around 60fs than the spin showing that an ultra-fast quenching of the magneto-crystalline anisotropy occurs. This result could be a clue for understanding the laser induced demagnetization process, since microscopic theoretical interpretations take into account spin-orbit interaction (SOI) [Zha2008, Koo2009, Kaz2009] and demonstrates that at time scales shorter than 100fs, one may enter the regime of the SOI. Since the magneto-crystalline anisotropy, which dictates the direction where the magnetization is directed, also relies on the SOI, understanding magnetization dynamics at such time scales may help finding new routes for ultrafast magnetization manipulation.

Developments in the field of magnetization dynamics naturally lead us to ask if there is a physical limit to the speed at which magnetic moments can be switched. Moreover, exploring this limit is complicated, partly because spin reorientation and switching from one direction to the other can occur in multiple ways and along different paths. For example, magnetic and electric fields, electric currents, and laser pulses can all stimulate magnetic reorientation and the trajectory of the magnetization vector from its initial to its final state will vary with each of these mechanisms.

So far, groups have mainly looked at ways of turning and redirecting the magnetization continuously, typically by causing it to precess with magnetic field pulses [Schu2003]. Using purely optical methods, Vahaplar *et al.* show that a faster way to switch the magnetization is to temporarily quench it [Vah2009] and restore it immediately afterwards in the opposite direction, a scheme they call a *linear* reversal.

Their experiments are an ingenious combination of the different effects by which light interacts with magnetic moments. In their setup, Vahaplar *et al.* first stimulate the magnetization of amorphous 20 nm ferromagnetic films made of $Gd_xFe_{100-x-y}Co_y$ with a short and intense circularly polarized (pump) laser pulse and then image the magnetization with a second, equally short but linearly polarized (probe) laser pulse.

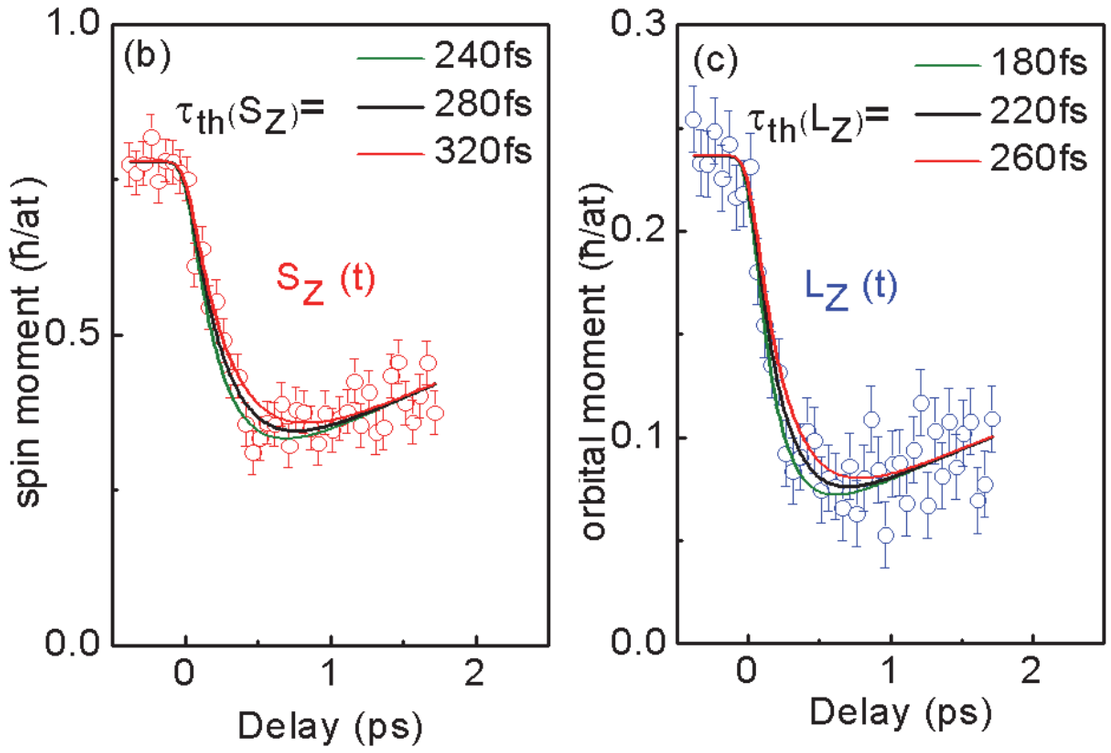


Fig. 7. Ultrafast dynamic of the spin and orbital magnetic moment measured using the pump probe set-up for a CoPd film at the $CoL_{2,3}$ edges. The X-ray probe beam are generated with the f-slicing set up at HZB-BESSY II leading to a time resolution of 130 fs [Boe2010]. Two different thermalization times were found for the spin and for the orbital magnetic moments. The best results of the fit procedure lead to τ_{th} (S_z) = 280 fs and τ_{th} (L_z) = 220 fs.

The first laser pulse has two effects on the magnetization. First, it rapidly pumps energy into the film, locally heating the material and demagnetizing it. Changes in the electronic temperature affect the magnetic properties on sub-ps time scales. Most importantly, the magnitude of the magnetization M decreases as the temperature of the electronic system approaches the Curie temperature. The first laser pulse also affects the magnetization via the inverse Faraday effect [Far1846, Ziel1965] : as the circularly polarized electromagnetic field pulse traverses the sample, it acts as an effective magnetic field along the pulse's propagation direction. This effective magnetic field is proportional to the intensity of the laser pulse and to its degree of circular polarization. The inverse Faraday effect provides outstanding possibilities to control the magnetization, since it can generate locally enormously strong effective magnetic fields of up to about 20 T. It can switch the magnetization as well, since the sign of the field only depends on the pulse's chirality. This optomagnetic, nonthermal control of the magnetization was first demonstrated by the Nijmegen group in 2007 [Stan2007]. Essentially, they showed that laser pulses as short as 40 fs could induce optomagnetic switching, but it was not clear how much time the magnetization required to complete the switching process after the exposure to such a short pulse.

By carefully varying the delay between the circularly polarized pump pulse and the linearly polarized probe pulse, the authors could obtain precise information on the spatiotemporal evolution of the magnetization in the film. They found that the switching process completes within a time well below 90 ps, which is very short but still much longer than the duration of the pulse. Recently the authors showed that 30-50ps is the ultimate limite for the swiching time using the invers Faraday mecanism.
The magnetization reversal is connected with a change of angular momentum, which must be provided from somewhere. Yet, it is generally agreed that the apparently simple assumption of a direct transfer of the photon spin to the magnetic system is not the solution [Koo2000], suggesting that the atomic lattice may play an important role in angular momentum conservation. This makes the question about spin to lattice (spin-phonon coupling) an important issu for a complet theoretical understanding of femtomagnetism. Recent experiments, using femto-slicing concluded that the angular momentum transfert is not using the orbital momentum to transfer the angular momentum from the spins to the lattice [Boe2010]. The measurements performed on CoPd alloys (fig 7.), show that ultrashort laser light pulses modify the orbital angular momentum of electrons before it is transfered to the spins, defining the correct sequence of transfert between orbit, spin and lattice. Different from the inverse Faraday effect the ultrafast manipulation of SOI is expected to transform and redirect the spins just by a single laser pulse by modifying the electronic anisotropy of any system at speeds down to the time speed of the laser pulse itself, ultimately atoseconds. The ultimate time speed limitation will be the electronic response to the laser field, typically faster than 1fs.

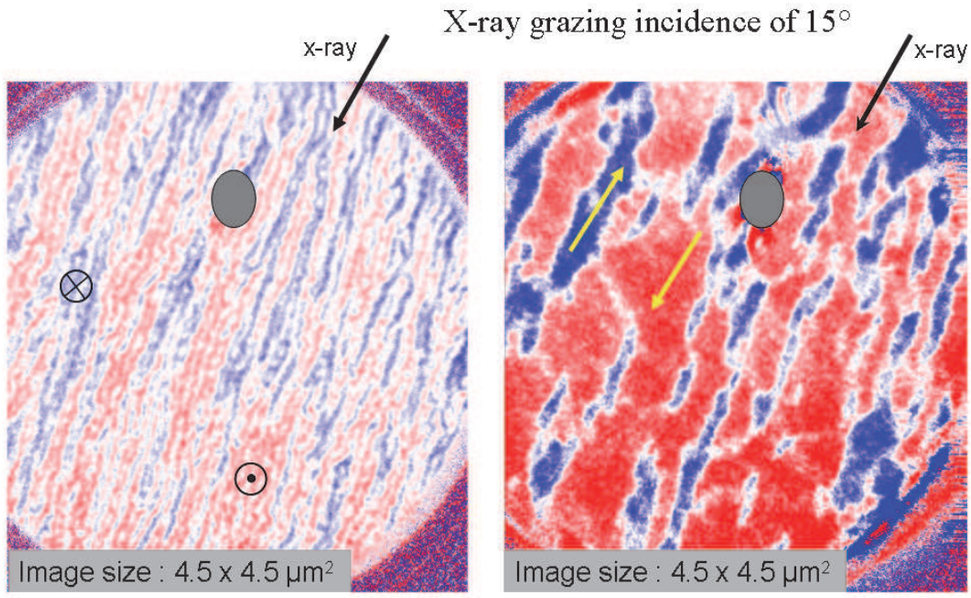
3.2 Magnetization dynamics at short length scales

Fundamental solid-state physics and electronics have progressed enormously in the last 20 years and this progress can be characterized by the words “smaller” and “faster.” In order to reach the ultimate ultrafast manipulations on the nanometer scale the challenge consists in improving our foundamental understanding of ultrafast magnetization dynamics and achieve ultrafast time resolved imaging at femtosecond time scales. In order to achieve this goal there are several aspects to consider related to ultrashort detection limits and spatial resolution capabilities (related to technical developments) which will ultimately enable a large step forward for the fundamental “ultrafast physics”.
Considering the spatial resolution in femto magnetism the experimental advances are much more recent and technical improvements still in progress. When excited by a very short subpicosecond stimulus with duration much shorter than the time of thermal equilibration in the spin system (~100 ps) the magnetic medium is set into a strongly nonequilibrium state, where a conventional description of magnetic phenomena in terms of thermodynamics is no longer valid, a macrospin approximation fails and the dynamics becomes often stochastic [Stö2006], totally different from scenarios that rely on classical magnetism [Tud2004, Vaha2009, Hert2009]. Experimental studies of the ultrafast dynamics of a stochastic process in a sub-100-nm magnet are very demanding as well. Indeed, the stochastic character of the studied process excludes the possibility of averaging in the experiment. This basically means that for such a study one would need to obtain a magnetic image of a sample within a picosecond period of time and with sub-100-nm resolution. So far, there has been no method that would satisfy these requirements. The recent development of ultrashort soft X-ray pulses, provided by X-ray free electron lasers, opens

new perspectives in this field. Several years after the pioneering work of S. Eisebitt et al. [Eis2004] demonstrating the possibility to image magnetic nanostructures by X-ray holography, C. Gutt and colleagues, report an experimental approach that may initiate a revolution in understanding ultrafast magnetic phenomena at the nanoscale. They show that by using one single 30 fs laser pulse it is possible to probe sub-100-nm magnetic domains in a Co/Pt multilayer sample [Gut2010]. The work was performed at FLASH free-electron laser facility at DESY in Hamburg. In fact, Gutt et al. have demonstrated an ultrafast probe of sub-100-nm magnets and thus have found a key to enter the uncharted world of femtosecond spin dynamics at the nanometer scale.

Similarly, the field of nanoscale magnetization dynamics is inseparably linked to the development of X-ray spin-sensitive methods as well as pulsed X-ray sources. Thanks to M. Faraday, who discovered the influence of a magnetic medium on the polarization of light [Far1846] magneto-optics in the visible spectral range has become one of the most popular tools for studies in magnetism. However, shorter wavelength is needed to observe down to the nanometer magnetic nanostructures. Since X rays sources have improved, since the 1990's advanced synchrotron radiation sources produce bright 50 ps pulses of polarized X rays. Using such sources, considerable progress has been achieved in understanding nanosecond magnetization dynamics at the sub-100-nm length scale [Waey2006].

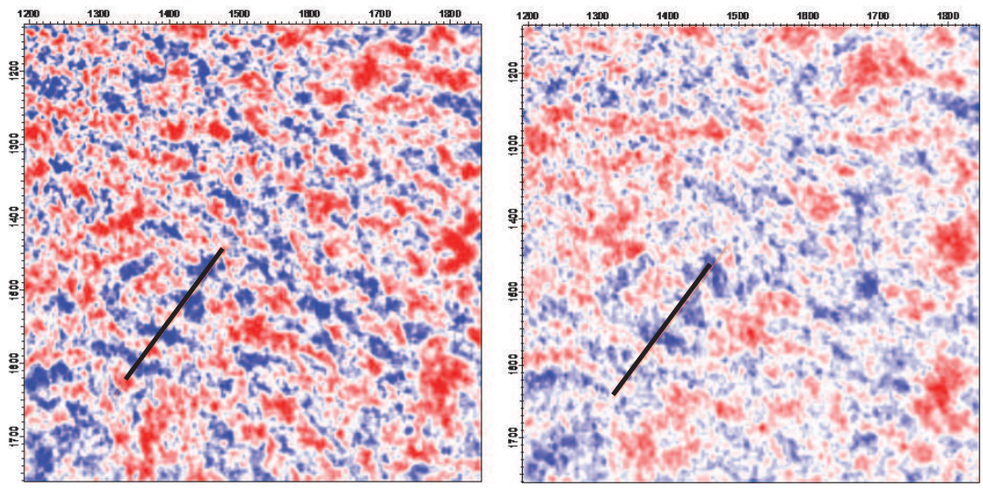
A recently started photoemission electron microscopy (PEEM) study at BESSY on thin CoPd alloy films allowed us to perform time resolved domain imaging in the 50 ps time range. Using the robust stripe domain pattern induced by the large out of plane anisotropy in this system, we aim to resolve the influence of a fs laser pump on the domain pattern and spin orientation in the domains. This work performed at BESSY-UE49 is still in progress, but has yielded already a first and important nanoscale description of the magnetization dynamics occurring in the thermalized regime of the first 100 ps. Figure 8 a,b show two static X-PEEM images (time integrated) taken during IR laser excitation (fs pulses at 5 MHz repetition rate) where thermal effects, reducing the out-of-plane anisotropy in the film generate a ~90° rotation of the spins from out-of-plane to in-plane. The spins thereby organizing in magnetic domains in the plane (fig 8 b), which are large in comparison to the initial, narrow stripe domains. The origin of this rotation is the reduction of the strong out-of-plane anisotropy by thermal effects. Moreover, the symmetry of the stripes also favors a 1D orientation of spins lying in the plane (figure 8b). This anisotropy of spins in the plane has been established by a complete azimuth analysis were complete extinction of the large magnetic domains was obtained after a +90 degree rotation of the sample (stripes) in respect of the X-ray incidence (not shown – see fig 8 c and d). These first results show that fs laser induced thermal effects can switch the anisotropy from out-of-plane to in-plane. When compared with the ultrafast reduction of the SOI observed for CoPd [Boe2010] we expect that the ultrafast change in SOI is able to switch the anisotropy in the fs time scale. This expectation is supported by the fact that the ultrafast SOI effect holds on for more than 2 ps (in the regime where spins and phonons are in thermal equilibrium [Boe2010]. In the pump probe experiment using lower fluency one can observe a limited 50 % reduction of the XMCD contrast in the stripe domains (fig. 8 c, d). Note that due to the azimuthal alignment (+ 90 deg) with respect to the incoming X rays (oriented along the stripe direction) the in-plane spins are here not observable at $t > t_0$.



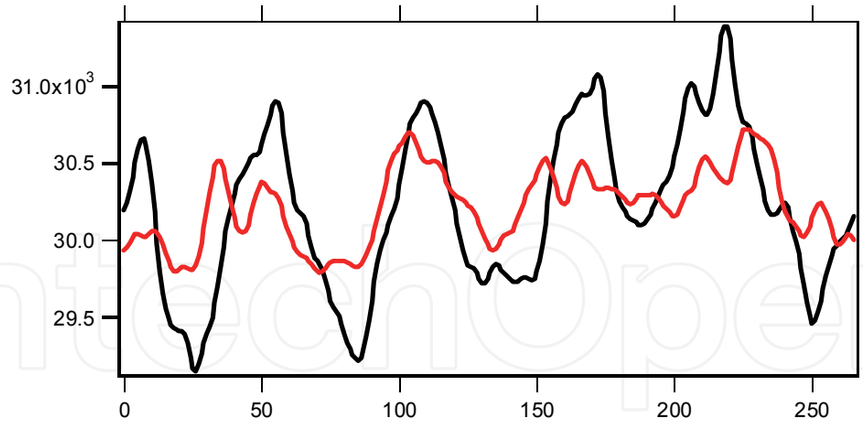
Stripe domains in CoPd film without laser. Out-of-plane spins (up = red; down = blue)

Domains in CoPd with pump laser on: In-plane spins

a) b)



c) t< to pump-probe d) t > to , pump-probe

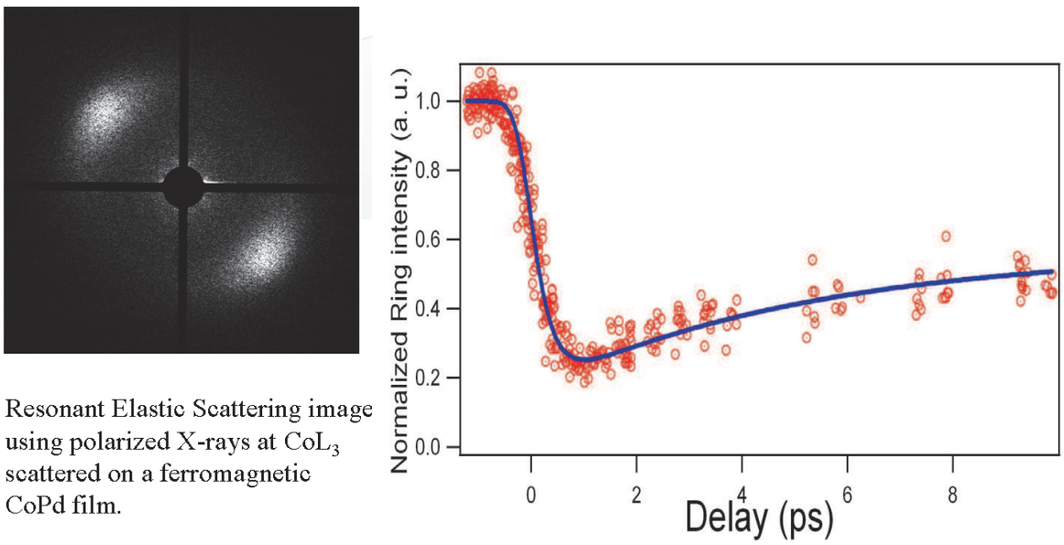


e) t < to for the black line (image c), while t > to in case of the red line (image d). Absice scale is in pixel. [Boe2011]

Fig. 8. (a and b) X-PEEM magnetic contrast images taken in the time-integrated mode. From a to b we observe the transformations induced by thermal effects of the fs laser excitation (5MHz). The magnetic domains show a stripe domain pattern (a) whereas large "in plane domains" are revealed in (b) induced by fs laser thermal heating. The color scale is common for the two images (a,b) and is proportional to the projection of the XMCD contrast along the x-ray incidence (15° grazing / surface). This leads to high sensitivity for in-plane contributions, while out-of-plane components are strongly reduced. For example, the projected value of +45% XMCD contrast at Co L_3 along the out-of-plane leads to only +2% XMCD contrast in the upper left PEEM image, which is obtained without laser pump pulse at room temperature. The grey pattern corresponds to the defect used to align the image position. Note that the left image shows modulation superimposed to the stripes. They are coming from small components of in-plane spins (less than 1%).
(c and d) Small field of view of 1,2 x 1,2 μm^2 of pump probe magnetic images taken in the time resolved mode (left: t = -100 ps before pump laser; right: t = 100 ps after pump laser). The color scale is common for the two images (c, d) and enhanced relative to the one of the two top images (a, b) in order to show the XMCD contrast reduction in this case. Note that due to the azimuth alignment in respect of the incoming X rays the in-plane spins are here not observable. The statistic is in the limit to clearly show the stripes but the line scans (e) performed along the straight line in the images c and d evidence the periodicity of stripes and show that indeed the XMCD contrast of the stripes is reduced by 40 %.

This specific orientation of the sample combined with reduced laser fluence allows to observe the very low XMCD contrast of 1% in the stripes at $t > t_0$. Finally, using the orientation dependent and fluence dependent information we can conclude that superposed to a contrast reduction of the stripes one can also expect a rotation of the spin into the plane and aligned along the stripe direction by fs laser pulse excitation. The results are revealing an important and new nanoscale "final state" of the fs laser induced SOI. And experimentally verifying this expectation is one of the major final goal in ultrafast magnetism.
Pump probe PEEM imaging is still in progress, but will yield a time resolution of 100 ps only. In order to bridge the time gap between 100 fs and 100 ps we will need to perform single shot imaging using Fourier transform Holography (FTH) with 100 fs time resolution using X-FEL sources. Compared with multi-shot imaging this single shot mode allows for imaging

with higher or less adjusted laser pulses (destructive for the stripe domains at ps time scales). Alternatively, the dynamic in worm domains can be studied with multi-shot imaging, suppressing the in-plane anisotropy of the spins observed with PEEM.
For instance, it has been shown by our previous work [Beau 1998] and [Boeg2010] that for magnetically saturated $CoPt_3$ and CoPd films a 100 fs laser pulse can partially or completely demagnetizes the film. Recently, we performed first time resolved resonant X-ray scattering experiments at LCLS on remanently aligned CoPd stripe domains [Lün2011]. In this case, due to the large sensitivity of the absorption coefficient to the local orientation of the magnetization vector, the X-ray beam is diffracted on the domain pattern, allowing measurement of the characteristic domain length scale. It can also be expected that the intensity of the diffraction pattern is proportional to the magnetic contrast between domains, providing information about magnetization dynamics. This experiment shows that stripe domains and bubble domains in CoPd films can be pumped by a 100 fs laser. We find that the time scale of the observed ultrafast demagnetization (see fig. 9) corresponds to the one of the spin dynamics reported in our above discussed femto-slicing paper [Boeg2010]. We note, however, that the temporal resolution of this first LCLS experiment was only ~500 fs due to jitter between the IR pump and the X-ray probe pulses.
We also successfully imaged magnetic domain structures at LCLS using multi as well as single shot illumination (fig. 10). To obtain circularly polarized X rays, a prerequisite for imaging of magnetic domain structures by Fourier Transform Holography, we employed a "resonant polarizer". This is based on a thin, semi-transparent magnetic Co film, which absorbs at the magnetically dichroic resonance one circular component more strongly than the other. This way, X-ray pulses with about 70% circular polarization can be obtained at the Co L3 absorption resonance at about ~10% total transmission.



Resonant Elastic Scattering image using polarized X-rays at CoL_3 scattered on a ferromagnetic CoPd film.

Fig. 9. Normalized intensity of the resonant magnetic scattering signal (Co L_3 edge) versus delay between the IR laser pump pulse and the X-ray scattering probe pulse. The observed time evolution reproduces well the ultrafast demagnetization dynamics observed by XMCD spectroscopy at the BESSY slicing source [Boe2010]. Each data point corresponds to 300 pump-probe cycles and the entire curve was measured on a single sample spot. The experiment was performed at LCLS by the collaboration detailed in Ref. [Lün2011].

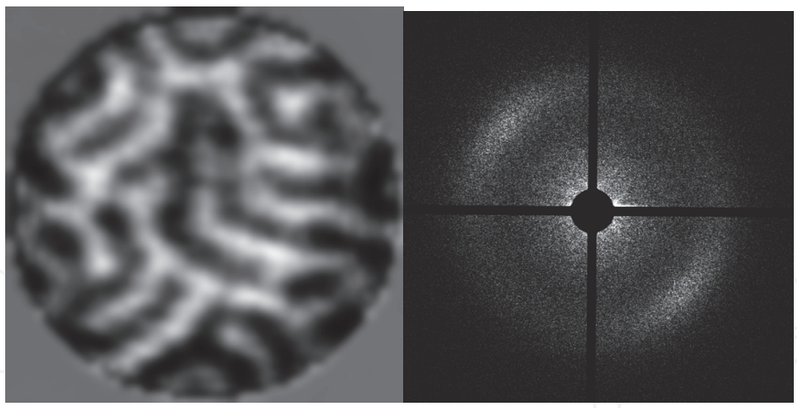


Fig. 10. (Left) Snapshot FTH image of a magnetic worm domain pattern recorded with a single LCLS X-ray pulse employing XMCD at the Co L_3 edge as contrast. The circular field of view is 1.5 □m in diameter (data treatment still in progress). (Right) Resonant elastic magnetic scattering pattern obtained using polarized X rays at the Co L_3 edge from a magnetic worm domain pattern [Lün2011].

Single shot snapshot imaging was in particular hampered by the photon energy jitter of the LCLS X-ray pulses, which leads to a high rate of "misfiring", since the XMCD effect is finite only close to the dichroic absorption edge. A monochromator was then used to select the Co L_3 resonance energy, which further reduced the available photon flux by nearly 2 orders of magnetitude. We note that IR pump – X-ray imaging probe experiments could not be realized during the last run at LCLS due to technical problems with the IR laser.
In order to describe the out-of-plane to in-plane transition we will carry out scattering experiments in two experimental geometries: First in a normal incidence geometry, in which the temporal evolution of the out-of-plane moment is probed. Then in grazing incidence geometry, in which both, the in-plane and the out-of-plane component, have finite projections. Hence, both contribute to the observed time dependence, but by comparison of the two time evolutions the in-plane component can be extracted from these data.
New aspects as for instance X-ray pump and X-ray probe will also been foreseen **in** the near future in order to study the interaction with X-ray at core level resonances. For instance X-ray single pulse intensities could be used to change the electronic configuration at the Fermi energy.
Note that FLASH and LCLS generate pulses with duration of down to 10 fs. This is already comparable with the characteristic time of exchange interaction in magnetic materials. It would be extremely intriguing to employ the elemental specificity of X-ray techniques and probe spin and orbital dynamics of TM and RE sub-lattices on a time scale pertinent to the time of the exchange interaction between them.

4. Conclusion

Pulsed X rays, are nowadays a promising route toward high temporal and spatial resolution allowing for quantitative and high contrast magnetic imaging at the elemental absorption edges of ferromagnetic materials. However, the available time resolution to date is too slow to resolve the fastest dynamics of 1 fs. Because of this, significant efforts have been devoted to using short or isolated electron bunches of X rays pulses at synchrotron to perform time resolved microscopy with X rays. Nowaday we are at time resolutions of 100 fs, and better

resolutions are forseen for near future using X-FEL's. Magnetic imaging techniques as for instance X-ray PhotoEmission Electron microscopy (X-PEEM), Scanning Transmission X-Ray Microscopy (STXM) or X-ray Resonant Elastic Scattering (XRES), are currently using the short X ray pulses in order to accede to time resolved imaging in the picosecond time range. Unfortunately, the f-slicing technique in synchrotrons produces a strongly reduced photon flux hindering the f-second magnetic imaging at those facilities. Recent projects using other synchrotron techniques are planed in order to increase the photons per pulse in the picosecond range allowing to image magnetic domains at 1 ps time resolution using synchrotron light. In parallel a large variety of new physics develops at X-FEL's where intense X-ray pulses modifies strongly the electronic and magnetic structures of mater. The development of such instruments will also allow new scientific approaches fare from the actual quasi-static physics.

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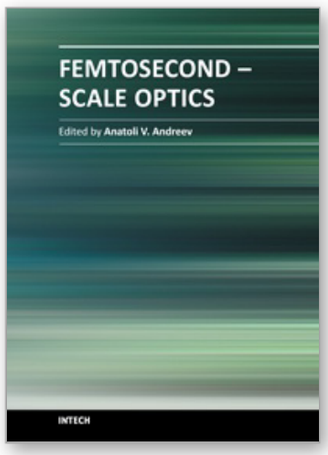

Femtosecond-Scale Optics
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With progress in ultrashort ultraintense laser technologies the peak power of a laser pulse increases year by year. These new instruments accessible to a large community of researchers revolutionized experiments in nonlinear optics because when laser pulse intensity exceeds or even approaches intra-atomic field strength the new physical picture of light-matter interaction appears. Laser radiation is efficiently transformed into fluxes of charged or neutral particles and the very wide band of electromagnetic emission (from THz up to x-rays) is observed. The traditional phenomena of nonlinear optics as harmonic generation, self-focusing, ionization, etc, demonstrate the drastically different dependency on the laser pulse intensity in contrast the well known rules. This field of researches is in rapid progress now. The presented papers provide a description of recent developments and original results obtained by authors in some specific areas of this very wide scientific field. We hope that the Volume will be of interest for those specialized in the subject of laser-matter interactions.

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