

# Exploring nanomagnetism with soft X-ray microscopy

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## Abstract

Magnetic soft X-ray microscopy images magnetism in nanoscale systems with a spatial resolution down to 15 nm provided by state-of-the-art Fresnel zone plate optics. X-ray magnetic circular dichroism (X-MCD) is used as the element-specific magnetic contrast mechanism similar to photoemission electron microscopy (PEEM), however, with volume sensitivity and the ability to record the images in varying applied magnetic fields which allows study of magnetization reversal processes at fundamental length scales. Utilizing a stroboscopic pump–probe scheme one can investigate fast spin dynamics with a time resolution down to 70 ps which gives access to precessional and relaxation phenomena as well as spin torque driven domain wall dynamics in nanoscale systems. Current developments in zone plate optics aim for a spatial resolution towards 10 nm and at next generation X-ray sources a time resolution in the fs regime can be envisioned.

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## 1. Introduction

Magnetism on the nanometer scale and its spin dynamics on a ns to fs time scale is currently a scientifically highly attractive topic [1,2] since it addresses both fundamental magnetic length scales such as magnetic exchange lengths in the sub-10 nm range [3] and interesting time scales of magnetism such as precessional and relaxation phenomena, domain wall and vortex dynamics [4–6]. The fundamental time scale in magnetism is given by the time required to transfer energy and momentum from the electronic into the spin system [7]. The aim is to understand the time scale on which magnetization can evolve upon excitation, e.g. with ultrafast optical pulses [8,9].

There is also a strong technological interest in fundamental magnetization processes on the nanometer length

and ns time scale due to current developments in ultrahigh density magnetic data storage media and miniaturized magnetic sensor technology. It is still an open question how to control, e.g. the switching field distribution in future magnetic storage devices where the bit size approaches the granular length scale.

New technological concepts such as spintronics, where in addition to the charge the spin of the electron is considered, require to control of the electron spin on a nanometer scale with ps timing.

Recently, it has been shown that apart from switching the magnetization with a magnetic (Oersted) field, a spin torque that acts on the local magnetization by injecting spin polarized currents into a magnetic device is a revolutionary concept to switch the magnetization on the nanoscale [10–18]. Logical elements for spintronics, non-volatile magnetic random access memories (MRAM) or 3dim magnetic data storage devices are just a few of the potential applications [19].

From the materials science point of view the quest is open to find novel and smart materials that will provide

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particular functionalities. Typical examples are, e.g. multiferroic systems which hold the promise to switch the magnetization, e.g. with electrical fields [20–23].

New analytical tools are required and a direct visualization of the magnetization is very appealing. The grand challenge to modern magnetic microscopies is therefore to provide both a spatial resolution in the nanometer regime, a time resolution on a ps to fs scale and elemental specificity which allows study of such advanced magnetic materials.

Magnetic transmission soft X-ray microscopy (MTXM) is a powerful novel technique that combines X-ray magnetic circular dichroism (X-MCD) as a huge and element-specific magnetic contrast mechanism with high spatial and temporal resolution down to 15 nm and 70 ps, respectively [24–28]. As a pure photon-in/photon-out based technique the images can be recorded in external magnetic fields giving access to study magnetization reversal phenomena on the nanoscale.

This paper discusses the current status of magnetic soft X-ray microscopy with regard to spatial and temporal resolution. It is complementary to X-PEEM regarding its bulk (volume) sensitivity compared to surface sensitivity in X-PEEM. This allows soft X-ray microscopy to access the nanomagnetism, e.g. in multilayered systems with a thickness much larger than the electron escape depth. Typical examples will demonstrate the achievements in spatial resolution and time resolved X-ray microscopy. Recent developments of novel X-ray optics allow for magnetic phase contrast imaging, which can be of importance at upcoming high brilliance X-ray sources that will give access to fs and single-shot spin dynamics with less than 10 nm spatial resolution.

## 2. Experimental details

There are three basic requirements to perform magnetic soft X-ray microscopy with high spatial and temporal resolution, namely high resolution X-ray optics, circular polarization of soft X-rays and a fast time structure of the X-ray source. Fresnel zone plates (FZPs) are the key components to soft X-ray microscopy. Due to the refractive index of soft X-rays, i.e. photons with energies between 200 and 2000 eV, being close to unity, conventional lenses cannot be used. State-of-the-art FZPs used as X-ray optical elements in soft X-ray microscopy currently provide a spatial resolution down to <15 nm [27,29]. FZPs are circular diffraction gratings with a radially increasing line density, i.e. they consist of alternating transparent and opaque rings. They can be fabricated using state-of-the-art nanolithography. The three parameters defining the performance of a FZP are: the outermost zone width ( $\Delta r$ ), the number of zones ( $N$ ) and the wavelength ( $\lambda$ ) [30]. From these three parameters all other ZP parameters can be derived such as focal length ( $f$ ), diameter ( $D$ ), and numerical aperture (NA). The spatial resolution of a zone plate based microscope is equal to  $k_1\lambda/NA_{MZP}$  where  $k_1$  is an illumination dependent constant, which ranges from 0.3 to 0.61. For a

partially coherent illumination  $k_1$  has a typical value of 0.4, and with the numerical aperture of the objective lens (the microzone plate in our case  $NA_{MZP} = \lambda/2\Delta r$  for high magnification the theoretical resolution for a full field microscope, as described here is  $0.8\Delta r$  [29].

Soft X-rays are abundantly available at synchrotron radiation sources. Moreover, they provide circularly polarized X-rays which are required when using XMCD as the magnetic contrast mechanism. For bending magnet sources the radiation viewed at an inclined angle achieves elliptical polarization with reversed helicity above and below the orbital plane which can be selected readily by appropriated apertures [31]. Dedicated helical undulator sources provide means to tune the circular polarization by mechanically moving the magnet array structures in the undulator source, and switching the helicity can be achieved within few seconds only.

Synchrotron radiation also exhibits an inherent time structure that is determined (and currently limited) by the lengths of the electron bunches circulating in the storage ring typically at 500 MHz. Thus, fast magnetization dynamics with typically <100 ps time resolution can be performed by applying stroboscopic pump–probe schemes.

Fig. 1 shows the schematics of XM-1, the high resolution soft X-ray microscope at beamline 6.1.2 at the Advanced Light Source in Berkeley CA (USA), which is currently the only system worldwide which is dedicated to studies of nanomagnetism [24]. Beamline 6.1.2 was built in 1994 by the Center for X-ray Optics (CXRO) and has been used by a large number of users since then for a wide variety of applications, ranging from nanomagnetism, materials and environmental science to biological applications. It exhibits a user-friendly design allowing recording of several thousands of X-ray images per day.

The principle of the soft X-ray microscope XM-1 for magnetic imaging is described in detail elsewhere [26]. Similar to an optical microscope XM-1 consists of two optical components, namely a condenser and an objective lens, however these comprise FZPs. First, a condenser zone plate together with a pinhole close to the sample forms a linear monochromator due to the wavelength dependence of the focal length of FZPs and second, the radiation passing through the sample is projected through the micro zone plate (MZP), which is the lens determining the lateral resolution, onto a CCD camera, where the images are recorded (see Fig. 1 [24]).

To image magnetic nanostructures the off-orbit emitted circularly polarized X-rays are selected by a movable aperture system upstream of the CZP. The spectral resolution that can be obtained at XM-1 with a typical value of  $\Delta E/E = 500$  is sufficient to discriminate the spin-orbit separated  $L_{3,2}$  edges ( $\sim 10$ – $20$  eV) in transition metals such as Fe, Co, Ni occurring around 700–900 eV, so that XMCD can be used as magnetic contrast mechanism.

As a pure photon-in/photon-out based microscopy technique external magnetic field can be applied during the recording so as to investigate magnetization reversal processes, local switching field distributions, etc.

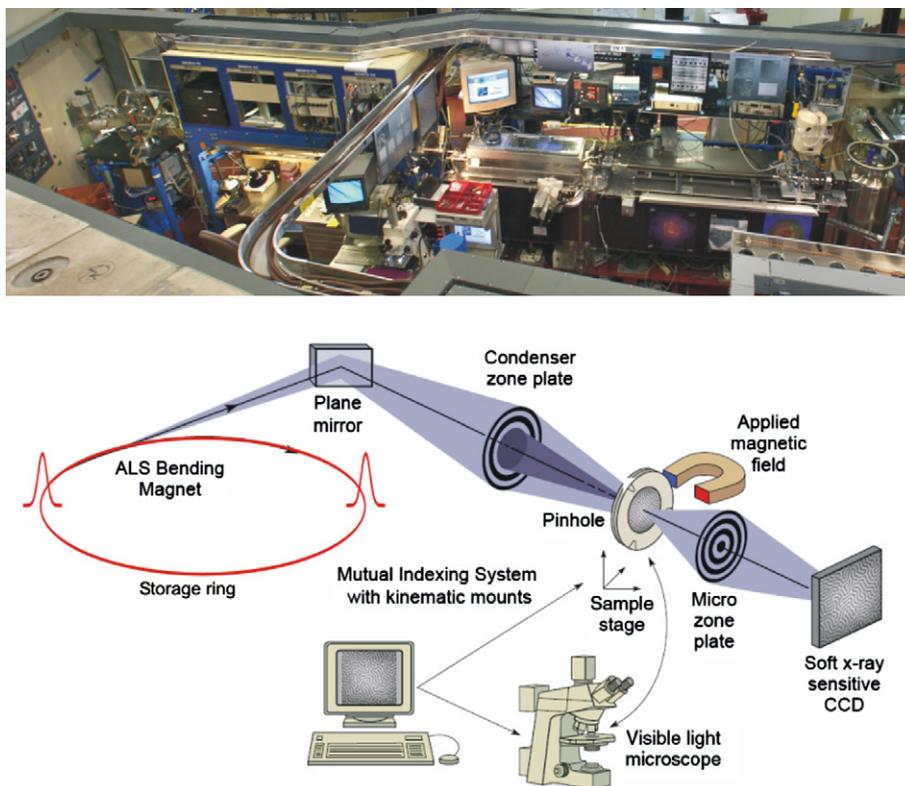


Fig. 1. Top: Bird's eye view of the soft X-ray microscope beamline 6.1.2 (XM-1) located at the Advanced Light Source in Berkeley, CA. The X-rays enter from the left through the concrete wall into the microscope. Bottom: Schematics of the X-ray optical layout of XM-1 [24].

Typical exposure times for magnetic imaging range from 0.5 to 3 s typically and each image covers a 10–15  $\mu\text{m}$  field-of-view. To eliminate non-magnetic background structures in the raw data, one can either normalize to a fully magnetically saturated image or reverse the magnetic structures by taking two images with opposite helicity.

Studies of spin dynamics on the nanoscale combine the high spatial resolution of magnetic soft X-ray microscopy with a stroboscopic pump–probe scheme utilizing the inherent time structure of the emitted X-ray pulses [32]. At the ALS there are two modes of operating the storage ring. In multibunch mode the electrons with a typical energy of 1.9 GeV circulate at a frequency of 499.642 MHz in 276 bunches each with a length of 70 ps and a distance between neighboring bunches of 2 ns thus providing a total electron current of 400 mA. With a 196.8 m circumference of the ring there is a gap in the storage pattern, which is filled with a single (camshaft) bunch of 10 mA. The second mode of operation is the two-bunch mode where there are two electron bunches stored, each of them with 20 mA current and 70 ps lengths and separated by 328 ns, i.e. emitting 3 MHz X-ray flashes that can be used for time resolved spin dynamics experiments at XM-1.

### 3. Results

X-ray magnetic circular dichroism detects the difference in the magnetic photoabsorption cross section which de-

pends on the relative orientation between the projection of the specimen's magnetization onto the photon propagation direction and the helicity of the transmitting X-rays. Strong X-MCD effects up to tens of percent occur in the vicinity of element-specific  $L_3$  and  $L_2$  absorption edges of transition metals, e.g. Fe, Co, Ni, which correspond to the binding energies of inner core atomic levels, such as the  $2p_{3/2}$  and  $2p_{1/2}$  levels, respectively. Since the magnetic ground state of a ferromagnetic system in general splits up into a magnetic domain configuration [33], i.e. in order to minimize competing magnetic energies, such as anisotropy, exchange, dipolar and Zeeman energy it is more favorable to create neighboring areas where the magnetization points in opposite directions, the transmission of circularly polarized soft X-rays will differ locally and can be visualized by imaging the transmitted X-rays with sufficient spatial resolution. Since magnetic thin films exhibit in general an in-plane magnetization to avoid magnetic charges, these samples have to be tilted with respect to the incoming photon beam to record the projection of the magnetization.

A typical example of high resolution magnetic imaging with soft X-ray microscopy is shown in Fig. 2 [27]. The magnetic domain structure of a 50 nm thin nanogranular  $(\text{Co}_{83}\text{Cr}_{17})_{87}\text{Pt}_{13}$  alloy film with a pronounced perpendicular magnetic anisotropy and its nucleation and magnetization reversal behavior in external applied magnetic fields was recorded at the Co  $L_3$  absorption edge. The dark and white areas in the image correspond to the direction

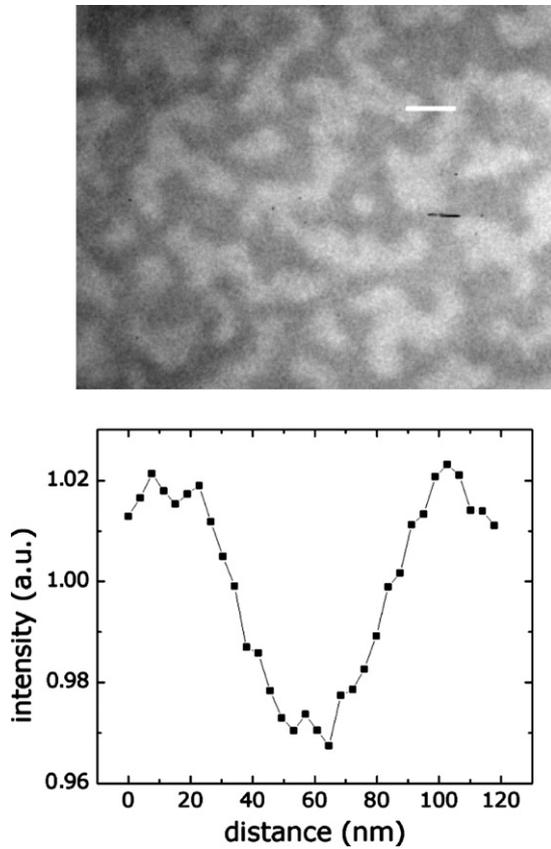


Fig. 2. Top: Magnetic X-ray image of the domain structure in a CoCrPt nanocrystalline thin film imaged at the Co L3 absorption edge (777 eV) with 15 nm MZP. Bottom: Intensity scan across the white bar in the X-ray image (above), indicating a spatial resolution of <15 nm [27].

of the Co magnetic moments. TEM analysis of this film revealed a grain size distribution with an average grain size of about 20 nm. From the intensity profile shown in the bottom panel of Fig. 2 taking into account the conventional 10–90% range of values, a 15 nm resolution, provided by the high resolution MZP was obtained, which indicates that these results provide insight into the switching of individual grains in that system which is discussed as a candidate for perpendicular recording.

Apart from imaging the static magnetism at fundamental magnetic length scales, there is an increasing interest to understand also fast spin dynamics occurring on the nanoscale [34–36]. However, because the number of photons per X-ray pulse at current facilities is not sufficient to create

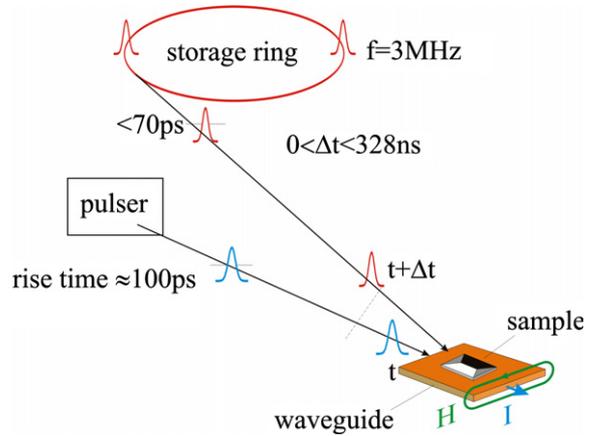


Fig. 3. Schematics of the stroboscopic pump-probe experiment at XM-1 to image time resolved spin dynamics in magneto nanoelements. The pump is an electronic pulse launched into a waveguide structure underneath the magnetic element to create a short magnetic field pulse (rise time 100 ps) onto the specimen. The probe pulse are X-ray flashes at 3 MHz with a length of 70 ps. To study the dynamics the pump pulse is delayed relative to the probe pulse up to several tens of ns.

single-shot images, a stroboscopic pump-and-probe measurement is used (see Fig. 3) to record the magnetization dynamics in nanoscale magnetic elements with time resolved magnetic X-ray microscopy. As a pump pulse a short electronic current pulse is used with a rise time of less than 100 ps and a typical pulse length of up to several 1 ns. When launched into, e.g. planar waveguide structures one can generate subsequent short and instantaneous magnetic field pulses, which excite various modes of the local spin configuration. There is increased interest to excite not only with classical Oersted fields, but moreover to inject spin polarized currents into nanoscale materials and thus create a spin torque, e.g. on the domain walls. Such dynamics can also be addressed by time resolved X-ray microscopy.

The probe is the X-ray flash of the synchrotron which can be delayed with respect to the pump pulse up to several ns in steps of several ps to study the temporal evolution of the magnetization after the exciting pulse. Typical accumulation times per single image are about 2–5 s and about 25–50 images are summed for each time step. As with the static imaging, non-magnetic background contributions can be suppressed by normalizing to a magnetically saturated image obtained by applying a strong external magnetic field up to few kOe field strength.

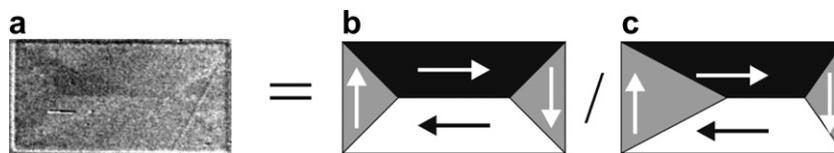


Fig. 4. Typical example of spin dynamics in a PY nanoelement exhibiting a four domain Lanau configuration. (a) Experimental difference image for the two delay times indicated in (b) and (c) showing the motion of the vortices and the associated domains. (b) and (c) Schematic diagram showing the configuration in the element at two distinct delay times between the pump and the probe pulse. The arrows indicate the direction of the magnetization in the film plane.

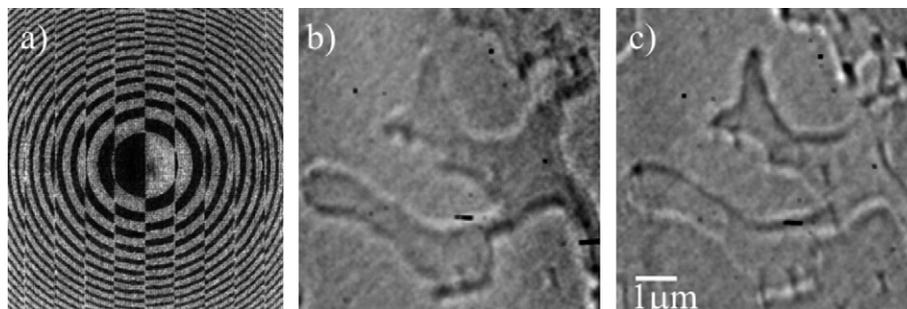


Fig. 5. (a) XOR Fourier optical element that replaces the conventional MZP to detect magnetic phase contrast. (b) Magnetic domain structure in an amorphous GdFe thin film recorded at 705 eV (below the absorption edge). (c) Magnetic domain structure recorded at 711 eV indication both a magnetic contrast at the boundary (differential contrast detection) and the reversal of the contrast [38].

A typical example where the vortex dynamics in a rectangular permalloy ( $\text{Fe}_{19}\text{Ni}_{81}$ ) microelement has been studied is shown in Fig. 4. Two ground state domain configurations can be observed, a seven domain and a four domain Landau pattern. The results of the seven domain Landau pattern and its spin dynamics can be found elsewhere [37]. Fig. 4 shows the schematics of the spin configuration at distinct delay times and an experimental difference image between these two delay times, which can be used for further analysis. One can clearly see that upon excitation the vortices in the Landau pattern have been displaced, i.e. they have moved along the central line towards each other. Following the full sequence of the dynamics, one can identify simultaneously domain wall motion, vortex dynamics, closure domain and spin wave dynamics emerging from both the vortices and the domain walls [37]. Thus, time resolved X-ray microscopy provides high quality experimental evidence to micromagnetic simulations even in more complex structures.

#### 4. Conclusion

Magnetic soft X-ray microscopy has achieved a spatial resolution of 15 nm which is close to fundamental magnetic length scales. Magnetic nanostructures, domain walls, and media at the grain size level can be imaged and its reversal behavior in external magnetic fields can be studied. The inherent elemental sensitivity due to XMCD as magnetic contrast mechanism is valuable in view to multilayered, multicomponent novel and advanced magnetic materials, such as magnetic multilayers, diluted magnetic semiconductors and multiferroic systems, which combine different order parameters and hold great promise as novel technologically relevant nanoscale (quantum) materials with outstanding novel functionalities.

So far, time resolved X-ray microscopy is limited in two senses. Firstly, only periodic (reproducible) processes can be studied due to the limited number of photons available at current X-ray sources. Secondly, the current time resolution of soft X-rays is far away from fundamental time scales that are associated with the time scale of exchange

interaction and magnetization fluctuations in the fs regime. However, upcoming X-ray free electron lasers and other high brightness coherent and incoherent sources for soft X-ray offer the opportunity for single-shot imaging of fs spin dynamics with soft X-ray microscopy. Even more, the continued development of suited X-ray optics is crucial. To minimize the photon load onto the sample, an interesting opportunity is to use magnetic phase contrast imaging. Recently, it has been shown that with Fourier optical elements, a magnetic differential interference contrast could be obtained [38]. An XOR zone plate, which was used instead of the conventional MZP demonstrated magnetic phase contrast showing up at a photon energy below the absorption edge, where the flux of photons can be reduced significantly. Fig. 5a shows the scheme of the zone plate optics and the results obtained around the  $L_{3,2}$  absorption edge of a  $\text{Gd}_{25}\text{Fe}_{75}$  amorphous film are displayed in Fig. 5b and c. The boundary of the magnetic domain patterns are enhanced at a photon energy of 705 eV and 711 eV, where no absorption contrast is expected. The observed reversal of contrast is in agreement with measurements of the magnetic real part of the refractive index [39].

It can be foreseen that magnetic soft X-ray microscopy will play an important role in the future to study spin structures on the nanoscale with fs time resolution.

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