Title

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Soft X-ray microscopy: Facing the mesoscale challenge in magnetism

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ABSTRACT

For more than a decade magnetism research focused on a fundamental understanding and controlling of spins on a nanoscale. The next step beyond the nanoscale will be governed by mesoscale phenomena. Those are expected to add complexity and functionality, which are essential design parameters e.g. for the realization of future spintronic devices. Advanced characterization techniques will play key roles in achieving mesoscience goals and multidimensional spectro-microscopies utilizing polarized soft x-rays offering a unique combination of spatiotemporal resolution, elemental and magnetic sensitivity, and tomographic capabilities are very promising.

As an example for complex behavior we show experimental results of the stochastic character of the nucleation of magnetic vortex structures in arrays of permalloy nanodisk. We have used magnetic soft x-ray microscopy to image at 25nm spatial resolution and a field of view of about 8 μm diameter both the circularity and the polarity of the disk, which allowed us to categorize the nucleated vortex state without ambiguity. We have found a symmetry breaking effect in the final vortex state, represented by a preferred handedness. We were able to identify as the origin of the asymmetry an internal Dzyaloshinskii-Moriya interaction arising from a broken inversion symmetry at the top and bottom surface/interface of the disk. Full 3-dimensional micromagnetic simulations confirmed our experimental observation. Here we present our observations with regard to disk diameter and disk thickness.

Various soft x-ray microscopy approaches are currently pursued to obtain full 3dimensional images of magnetic structures, including computed reconstruction of 2dim projection images.

Keywords: XMCD, magnetic soft x-ray transmission microscopy, Dzyaloshinskii-Moriya interaction, magnetic vortex, X-ray optics, mesoscience, soft X-ray tomography

1. INTRODUCTION

Over the last decade magnetism research focused on a fundamental understanding and controlling of spins on a nanoscale. A wealth of information has been achieved, which to a large extent was made possible by the development of advanced instrumentation providing nanoscale information. Recently, it has been recognized, that the next step beyond the nanoscale will be governed by mesoscale phenomena[1].

The mesoscale is not just defined as a length scale bridging the nanoscale - i.e. the length scale of single atoms- to the micro/macroscopic range. In that sense, mesoscale magnetism would correspond to the transition from single spin behavior and its fundamental exchange interaction with neighboring spins to a regime where magnetic properties act as a continuum, i.e. the world of micromagnetism[2]. Beyond that, mesoscience is expected to bring to bear complexity, stochasticity and functionality, which do not exist at the nanoscale. It can be anticipated that properties and behaviors emerging from those mesoscale phenomena will see an increased relevance in future applications. They will also require new and extended theoretical models reaching out into longer length and time scales.

Complexity in magnetic materials will become more important not only in combinatorial materials design approaches, such as the "materials genome initiative" (c.f. http://www.whitehouse.gov/mgi), which aims to discover and tailor specific properties from basic principle calculations of novel materials, but also in devices extending into the third dimension, where the "simple" cross-talk in conventional planar geometries will turn into corresponding volume effects. Closely related to this is the importance of interfaces and surfaces which are crucial in functionalizing of multi-component systems. The question as to whether magnetic processes on the nano- or the meso-scale exhibit an intrinsically deterministic character or follow stochastic behavior is not only scientifically fundamental, but will again be highly relevant in technological applications. Finally, considering static mesoscale structures alone is not sufficient, as functionality e.g. of magnetic devices has to take into account the spin dynamics across multiple time scales.

Prerequisite to achieve mesoscale scientific goals is the development and application of novel multidimensional characterization techniques. To understand magnetic properties of mesoscale systems, advanced characterization tools should meet the following requirements:

- a spatial resolution down to below 10 nm reflecting the fundamental exchange lengths in magnetic materials, and ultimately in all 3 dimensions,
- a temporal resolution from the sub-nsec down to the fsec regime, to investigate spin dynamics down to the time scales of exchange interactions,
- chemical and magnetic sensitivity with elemental specificity, which is highly relevant in multicomponent magnetic materials, and finally
- interface and surface sensitivity.

The unique properties of polarized soft X-rays[3], i.e. their wavelength between about 0.5 and 5nm, corresponding photon energies between 0.2 and 2keV, polarization characteristics, inherent time structure at synchrotrons (sub-nsec and psec) and x-ray free electron lasers (down to fsec), their abundant availability and high brightness, and the arsenal of powerful experimental techniques (spectroscopies, microscopies, and scattering) makes them a strong candidate as a probe for mesoscale challenges in magnetism.

2. MAGNETIC SOFT X-RAY MICROSCOPY

Here, we want to focus on magnetic soft x-ray microscopies, i.e. utilizing soft x-rays to image magnetic structures and their behavior. Magnetic contrast is obtained via X-ray dichroism effects, in particular the X-ray magnetic circular dichroism (XMCD) effect for the study of ferro- and ferrimagnetic materials and X-ray magnetic linear dichroism for antiferromagnetic materials. XMCD can be seen as the counterpart of the Kerr or Faraday effect in the X-ray regime. The absorption of circularly polarized in magnetic materials with photon energies corresponding to binding energies of inner core spin-orbit split electron levels, e.g the $2p_{3/2}$ and the $2p_{1/2}$ levels, depends strongly on the relative orientation of the helicity of the photons and the magnetization of the specimen. The characteristic resonant enhancement of x—ray absorption at those photon energies is referred to as the L_3 and L_2 x-ray absorption edges. Since these electronic levels are specific to the element and even its chemical state, the XMCD effect and thus the magnetic contrast in magnetic soft x-ray microscopies is inherently element specific, which translates into layer resolved capabilities, e.g. in a double layer consisting of different magnetic elements.

A variety of magnetic imaging techniques using soft X-rays is available. The first demonstration of magnetic x-ray imaging utilized photoemission electron microscope (X-PEEM), which records the local XMCD effect through imaging the secondary electrons generated in the X-ray absorption process and escaping the sample from a 5-10nm thin surface layer[4]. The latest generation of X-PEEM includes aberration correctors and is aiming for a <10nm spatial resolution. Next, there are two variants of soft x-ray microscopes, which utilize Fresnel zone plates (FZP) as x-ray optical elements and which are used for magnetic imaging[5]. FZP are circular grating, fabricated by state-of-the-art nanopatterning techniques, e.g. e-beam lithography. The FZP parameters, i.e. outermost zone width Δr, diameter D and number of zones N of the FZP as well as aspect ratio of the zones determine their performance in terms of spatial resolution, temporal bandwidth and diffraction efficiency. The latest generation of FZPs have demonstrated <10nm spatial resolution[6].

A full-field transmission soft x-ray microscope (TXM) uses the FZP as high resolution objective lens, which in a Scanning TXM (STXM) is used to focus the x-rays to a diffraction limited spot, which is then raster scanned across the sample. Whereas both TXM and STXM have similar spatial resolution, the parallel recording scheme, i.e. taking full images with a CCD detector in few secs of exposure time in TXM, offers clear advantages for multidimensional imaging approaches, such as spectromicroscopy and/or tomography studies. Compared to PEEM, with TXM and STXM as pure photon-in/photon-out techniques, the application of magnetic fields of in principle any strength and pointing into any direction is very easy to achieve. The limited absorption of soft x-rays around 1keV requires that the samples have to be prepared on x-ray transparent substrate, and Si_3N_4 membranes, which are also common in TEM studies, are the most widely used ones. The accessible range of sample thicknesses ranges from about 1nm to about 100-200nm. Whereas the lower end overlaps with accessible probing depth of X-PEEM, TXM/STXM provide information about bulk properties of the samples.

In addition to those "real-space" imaging tools, there is a strong effort with diffraction imaging approaches, such as coherent diffractive imaging (CDI)[7], x-ray holography[8] and ptychography. Most of them are still at the stage of instrument development and therefore have not yet been used extensively for studies of magnetic materials.

The major advantage of soft x-ray microscopy is the combination of spatial and temporal information. Time resolved magnetic soft x-ray microscopy studies depend on the inherent time structures of x-ray sources, i.e. the length of each x-ray pulse, its repetition rate, and the number of photons per pulse. At synchrotron sources, this is the sub-100 ps time scale, a MHz repetition rate and due to the low photon intensity per pulse, stroboscopic pump-probe scheme have to be used. Fully reproducible processes, such as resonant gyrational modes in magnetic vortex structures or domain wall motion have therefore been the focus of research. At X-ray free electron lasers (XFEL), where the intensity per shot is much higher, single shot imaging of ultrafast nanoscale magnetic behavior seems to become feasible.

So far, the full-field TXM at the Advanced Light Source in Berkeley (XM-1) is the only instrument worldwide used extensively for magnetic TXM (MTXM). There, magnetic fields of up to 2-3kOe in photon beam direction and up to 1-2kOe magnetic field along the sample's surface can be applied. Both in-plane and out-of-plane magnetization can be imaged and a rotary stage allows recording of tomographic datasets. In the following, we will show one example with MTXM to demonstrate its usefulness for mesoscale science.

3. EXAMPLE: MAGNETIC VORTICES

Magnetic vortex (MV) structures occur in soft ferromagnetic patterned elements, such as thin disks of permalloy (PY), a $Ni_{80}Fe_{20}$ alloy, as a result of the balance between exchange and dipolar energies[9]. They are characterized by a curling magnetization in the plane of the disk with a vortex core (VC) in the center, where the magnetization points perpendicular to the plane of the disk. Two binary properties are commonly used to describe this structure: the circularity (C), i.e. the counter-clockwise or clockwise curling of the in-plane magnetization, and the polarity (P), i.e. the up or down direction of the vortex core's magnetization, making a total of four independent MV configurations. The product CP defines the handedness, which can take the values of +1 and -1. Both the static and dynamic properties of MV structures have recently attracted an increased scientific interest both for fundamental and applied reasons. For example, magnetic vortex structures were suggested as potential future high-density and non-volatile recording systems, since the size of the vortex core is proportional to the magnetic exchange length Λ, which can extend into the sub-10 nm regime, and the magnetic core represents a very stable spin configuration, in fact protected by topology.

Figure 1 shows typical examples of MTXM images of MV structures in PY disk arrays with h=100nm disk thickness and various disk diameters D ranging from 400-1000nm. The left column shows C and was imaged with the sample's surface tilted at 30degree relative to the photon propagation direction since XMCD measures the projection of magnetization onto the photon beam direction. The right column shows images of P in the center of the disks pointing perpendicular to the disk surface in the identical elements as in the left column. Both clockwise and counterclockwise curling of C and both up and down orientation of P, which corresponds to black and white contrast, resp. can be observed. This raises the interesting question, as to whether the four independent MV configurations occur with equal probability, i.e. whether their energy is fully degenerate. To address this question, the nucleation of arrays of MV structures in permalloy disks was studied with MTXM[10].

The MVs in the permalloy disk arrays were generated by applying an external field to the sample, so that the MV core in each disk is completely expelled. Upon reducing the magnetic field to 0, the MV structure forms again This procedure was performed repeatedly, until a statistically significant number of events has been reached. Surprisingly, it was found, that there is a significant preference for a certain handedness. This constitutes an unexpected symmetry breaking effect in the nucleation process of MVs.

The Dzyaloshinskii-Moriya interaction (DMI) was considered as a potential origin for this asymmetry. The DMI originates from a broken inversion symmetry at the surface or interfaces in thin layers and has been identified to affect the shape and size of MV structures[11]. Full 3dim simulations were performed to study the effect of DMI on the nucleation process in the permalloy arrays. It was found, that the nucleation of the MV at the bottom and the top layer was asynchronous and while the overall nucleation process seems to be identical, details of the nucleation process are affected by the DMI. Excellent agreement with the simulation could be obtained by adjusting the DMI parameter.

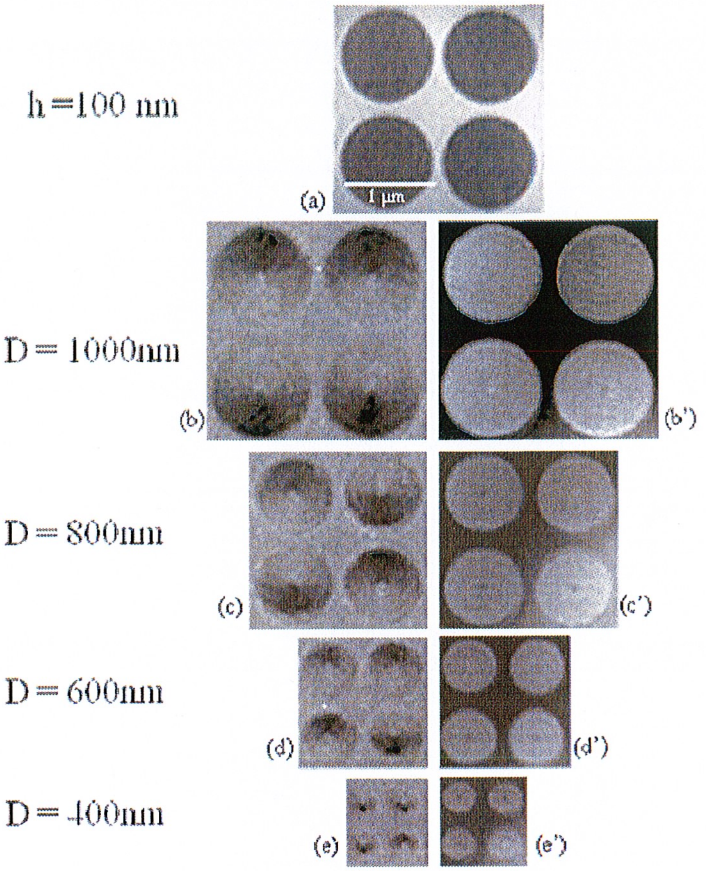

Figure 1 MTXM images of the circularity (left column) and polarity (right column) of an array of permalloy disk with disk height 100nm and varying disk diameters D. The top row shows the raw data, the C images were obtained by normalizing to saturated images, the P images were obtained by subtracting two images recorded with opposite circular polarization of the X-rays.

The capability of MTXM to image with sufficient high spatial resolution directly both C and P of identical elements and the short exposure times per image to acquire statistically significant data in a reasonable amount of time were the key to address and answer this question.

In the following we want to address the question, whether the geometry of the disks or the experimental procedure to generate the MV structure has an impact on the observed asymmetry in the nucleation process of MV structures. Figure 2 shows a sample of MTXM images of disks with a fixed diameter of 1000nm, but now with varying height ranging from h=40 to h=100nm. The circularity obtained in the MV nucleation process with the procedure described above was compared between subsequent cycles to determine, if there was a correlation to the thickness. The occurrence of the MV state can be clearly seen in each of the images. As can be seen from the overlapped images in Figure 2, there seems to be no direct correlation of the degree of stochasticity with the disk thickness. Since the asymmetry originates from the surfaces of the disk, one would assume that this effect should become more pronounced for reduced disk thicknesses, which cannot be confirmed by the obtained experimental data.

Figure 3 addresses the question, whether the experimental procedure has an influence on the MV nucleation process. The top row shows MTXM images of C obtained in opposite branches of the hysteresis cycle, i.e coming from positive or negative saturation to remanence. The bottom row compares the nucleation process obtained in the same branch of the hysteresis cycle. Again, the overlapped images for both of these cases allow to conclude the degree of stochasticity in the MV forming process. It is obvious, that the number of switched C states is significantly larger for opposite branches than for the same branch. This shows that data from both hysteresis branches have to be taken into account for investigating the stochastic character of the nucleation of MV structures in large arrays.

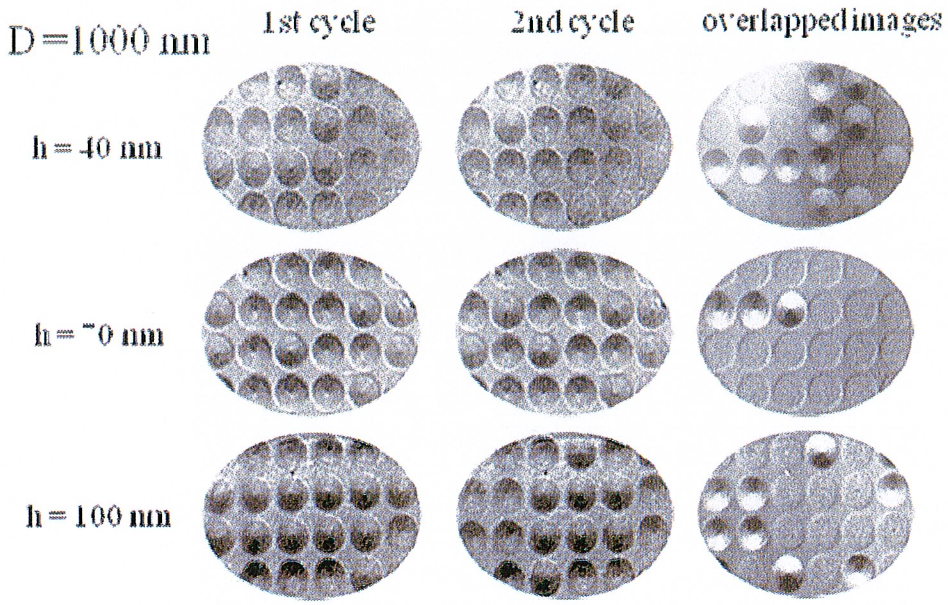

Figure 2. MTXM images of the circularity in arrays of permalloy nanodisk with fixed disk diameter (1000nm) and varying disk height. The degree of stochasticity in the nucleation process of the vortex structures seems to be uncorrelated with the disk thickness.

4. CONCLUSION

The nucleation behavior of ferromagnetic nanodisk arrays was studied with magnetic full-field transmission soft x-ray microscopy. An unexpected asymmetric behavior was observed, favoring a peculiar handedness. This could be traced back to an internal DMI interaction. The impact of disk thickness, diameter and measurement procedure was investigated. The asymmetric character of the nucleation process of MV arrays can be seen as a prototype example for mesoscale behavior.

The mesoscale era will bring new scientific challenges and technological opportunities. Novel characterization tools will be required to help understanding the underlying phenomena, which provide complexity, stochastic behavior and new functionalities. Soft X-ray spectro-microscopies offer a unique set of parameters, particularly the combination of spatio-temporal imaging with chemical and magnetic sensitivity and of particular interest will be the capability to obtain complete three-dimensional magnetic information.

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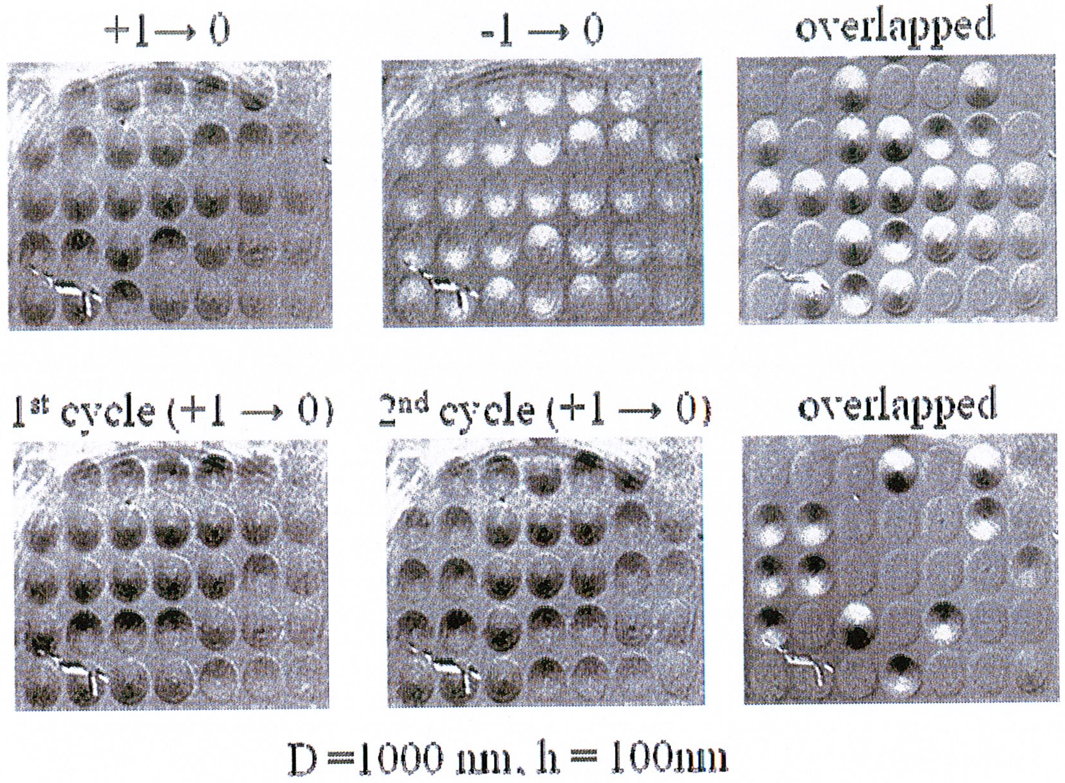

Figure 3. MTXM images of the circularity in arrays of permalloy disks with D=1000nm and h=100nm. The top row compares the first two images obtained in different branches of the hysteresis loop. The bottom row compares two images obtained in the identical branch of the hysteresis cycle. The nucleation process seems to be more stochastic, when coming from opposite saturation fields. The number of changes in the vortex structures, indicated by the overlapped images (right column) is significantly larger than for subsequent cycles coming from the same saturation field.

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