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Perpendicular magnetisation from in-plane fields in nano-scaled antidot lattices

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Abstract

Investigations of geometric frustrations in magnetic antidot lattices have led to the observation of interesting phenomena like spin-ice and magnetic monopoles. By using highly focused magneto-optical Kerr effect measurements and x-ray microscopy with magnetic contrast we deduce that geometrical frustration in these nanostructured thin film systems also leads to an out-of-plane magnetization from a purely in-plane applied magnetic field. For certain orientations of the antidot lattice, formation of perpendicular magnetic domains has been found with a size of several μm that may be used for an in-plane/out-of-plane transducer.

Keywords: antidot lattice, magnetic nanostructures, x-ray microscopy, geometric frustration, MOKE

(Some figures may appear in colour only in the online journal)

1. Introduction

Antidot lattices in magnetic materials, i.e. a periodic arrangement of holes in a thin metal film, have been investigated within a broad scientific and technical scope. Geometric frustration resulting in spin ice [1–3] and spin glass [4] behaviour have been discussed and observed in magnetic antidot lattices. These nanostructured systems also give rise to observable magnetic monopoles [5] and Dirac strings [5] and may be used in data storage applications [6]. Furthermore, they gained significant interest due to their ability to act as magnonic crystals which—similar to photonic crystals for photons—exhibit a periodic potential for magnons, allowing tuning of the spin wave dispersion [7, 8]. Thus, they are of interest for applications in the field of spintronics as spin wave filters or spin wave guides.

Hexagonal antidot lattices in in-plane magnetized materials, like the one shown in figure 1, have been broadly discussed regarding their effect on the in-plane magnetization of these materials. The coercivity is significantly increased [9–11] compared to the unstructured thin film and the anisotropy is clearly dominated by the antidot lattice symmetry [12–14]. In the hexagonal structure there are two distinguished directions: the nearest neighbour (nn) direction in which a string of close-packed holes is formed and which is the magnetic easy axis [12]; and the next-nearest neighbour (nnn) direction in which the longest distance between two adjacent holes is found and which is the magnetic hard axis (see inset of figure 1). The (nn)- and (nnn)-directions inclose an angle of 30° to each other.

Here we present a novel property of antidot lattices in magnetic materials, as we observe out-of-plane magnetization components from an applied in-plane magnetic field by means of magneto optical Kerr effect (MOKE) measurements with high spatial resolution as well as scanning x-ray microscopy (SXM) with x-ray magnetic circular dichroism (XMCD) as contrast mechanism. Transducing an in-plane excitation to an
out-of-plane magnetization may open up a range of new applications in sensing, spintronics and magnetic logic.

2. Results and discussion

The two distinguished directions of the hexagonal antidot lattice result in different magnetic behaviour for these two cases. Longitudinal MOKE measurements, that are sensitive to the in-plane magnetization of the samples, are shown in figures 2(a) and 3(a) for the (nn)- and (nnn)-direction respectively. For comparison the hysteresis loop of the in-plane magnetization of the samples, are shown in figure 2(a). In the case of an antidot lattice with a periodicity of 200 nm and a hole diameter of 100 nm the (nn)-direction clearly is the magnetic easy axis with increased coercive field and remanent magnetization compared to the (nnn)-direction which is the in-plane hard axis in this system. Although the longitudinal Kerr measurement is sensitive to any magnetization in the reflection plane of the laser beam, the signal is dominated by the in-plane signal [15].

In contrast to longitudinal MOKE, measurements in polar MOKE geometry probe the projection of the magnetization onto the surface normal [15]. The measurements of out-of-plane magnetization components for an applied in-plane magnetic field are shown in figures 2(b) and 3(b), with the magnetic field applied along the (nn)- and (nnn)-direction respectively. In order to exclude any in-plane magnetization leading to a significant signal in polar geometry, e.g. due to a possible misalignment of the setup, a permalloy reference sample was used and cross talk of less than 0.4 mdeg was achieved. Thus, the observed MOKE signal of 6 mdeg (figure 2(b)) is more than an order of magnitude larger than the experimental uncertainties. Furthermore, exactly the same measurement conditions applied for both the (nn)- and the (nnn)-direction as the sample was only laterally moved or rotated and thus the angle between the reflected laser spot and the sample surface was kept constant for measurements switching between the two directions. Additionally, this measurement was checked to be independent of the way this position was approached, i.e. the lateral movement or the rotation sense (clockwise or counter clockwise) to change from a (nnn)- to a (nn)-orientation with respect to the external field.

The results of the (nn)-direction measurements, shown in figure 2(b), are rather surprising as there is a prevailing magnetization component perpendicular to the applied magnetic field and perpendicular to the surface plane in pseudo-saturation. Moreover the out-of-plane (figure 2(b)) magnetization runs parallel to the in-plane (figure 2(a)) hysteresis loop. The behaviour of the (nnn)-direction, shown in figure 3(b), on the other hand is more straightforward as there is no net magnetization perpendicular to the surface plane when the in-plane field is large enough to saturate the sample.

To further confirm this behaviour and to localize the out-of-plane magnetization SXM investigations have been carried out at a photon energy of 707.0 eV, providing the maximum XMCD effect at the Fe L3 edge, in normal incidence geometry, being only sensitive to out-of-plane components of the magnetization [16]. An X-ray micrograph with an in-plane magnetic field of 240 mT applied along the (nn)-direction is shown in figure 4(a). The antidot lattice exhibits a locally homogeneous out-of-plane magnetization as indicated by the uniform blue contrast. This results in an averaged XMCD effect of 6% for the investigated 3 by 3 μm area, confirming a remaining out-of-plane magnetization at in-plane pseudo-saturation in agreement with the results obtained by MOKE (see figures 2 and 3). From this XMCD intensity compared to the XMCD effect along the magnetization direction [17], and also from the amplitude of the corresponding MOKE effect a homogeneous out-of-plane deflection of approximately $15° \pm 5°$ is estimated [15, 17].

For the case that the in-plane magnetic field is applied along the (nnn)-direction, as shown in figure 4(b), there is also a significant out-of-plane component that alternates on a very short length scale of less than two antidot lattice constants between an up and down orientation. As the out-of-plane magnetization is not homogeneous, averaging the XMCD signal over the sampled area results in no net out-of-plane magnetic moment. This corresponds to the MOKE measurements that average over a similarly sized beam spot and show no out-of-plane component in saturation for this orientation.

We therefore deduce that an out-of-plane deflection of the magnetization occurs for both the (nn)- and the (nnn)-direction. However, only the case of an applied magnetic field along the (nn)-direction results in domains that are large enough to be detected by MOKE measurements while a magnetic field along the (nnn)-direction results in an out-of-plane magnetization state that is disordered on a short length scale.

For the (nn)-direction domain boundaries between two homogeneously magnetized regions can be found in x-ray

5 Nanosphere lithography yields structurally homogenous domains with a size of approximately 10 by 10 μm [12]. Hence, it is possible to switch to a different orientation of the lattice either by rotating the sample or by laterally moving to a sample position with a different orientation. Both approaches were used to compare between the (nn)- and (nnn)-direction behaviour to exclude measurement artefacts.
micrographs. This is shown in figure 4(c), where a sample region with a domain-wall between up (blue) and down (red) magnetization components is depicted. Each homogeneously magnetized area exhibits a size of several μm, but is deflected in opposite out-of-plane directions. It can be assumed that the formation of homogeneous out-of-plane domains for the case of an applied field along the (nn)-direction is additionally stabilized by exchange coupling, favouring a homogenous alignment. This assumption of an involvement of the exchange coupling is further backed by the position of the out-of-plane domain wall, shown in figure 4(c), that runs along the shortest distance between two holes. Thus, the system minimizes the domain wall energy, as it is the case for in-plane domains in antidot lattices [10, 12].

Especially in the context of spin ice, monopoles and Dirac strings it has been discussed in literature that the introduction of an antidot lattice leads to a geometric frustration of the magnetic moments [1–3, 5]. Reduction of stray fields emerging from the hole edges leads to a misalignment of some magnetic moments with respect to the external magnetic field and thus is the driving force behind the frustration of the system. Previously, this misalignment has only been considered within the plane of the thin film in which the antidot lattice is hosted. However, in the light of our measurements this misalignment is not restricted to the surface plane of the sample, but also leads to an out-of-plane deflection of the magnetization. Because a deflection within the surface plane comes at the cost of exchange energy this small deflection out of the surface plane may instead become favourable. As deduced from the experiments presented here the geometric frustration induced by the antidot lattice structuring can, apart from the formation of a spin ice structure (at the cost of exchange energy), also generate an out-of-plane magnetization (at the cost of Zeeman energy). This behaviour is also reproduced in our micromagnetic simulations. Even in pseudo-saturation at 240 mT the in-plane magnetization is deflected off the external field direction in the proximity of the holes to avoid stray fields in the hole. As in this case the Zeeman energy is in competition with the stray field energy, we expect that the out-of-plane deflection can be overcome by much higher magnetic fields, resulting in all magnetic moments being aligned with the external magnetic field and within the surface plane.

Figure 2. (a) Longitudinal and (b) polar Kerr hysteresis loops with an in-plane magnetic field applied along the (nn)-direction of a hexagonal antidot lattice with 200 nm antidot periodicity and 100 nm antidot diameter in a 20 nm thick Fe film. For comparison the hysteresis loop of the unstructured material is additionally shown as dotted line in panel (a).

Figure 3. (a) Longitudinal and (b) polar Kerr hysteresis loops with an in-plane magnetic field applied along the (nnn)-direction of a hexagonal antidot lattice with 200 nm antidot periodicity and 100 nm antidot diameter in a 20 nm thick Fe film.
Generally, it is expected that the shape anisotropy contributes to the in-plane preference in an iron thin film. Cowburn et al have, however, shown that the local shape anisotropy due to the antidot lattice can favour an out-of-plane orientation of the magnetization [18]. Furthermore, the antidot anisotropy can indeed overcome any intrinsic anisotropy of the magnetic host material [18]. Considering the local geometry in an antidot lattice, it becomes evident that the aspect ratio is not comparable to a thin film anymore. In our case of a 20 nm thick film, an antidot periodicity of 200 nm and an antidot size of 100 nm the elements between two neighbouring holes have only an aspect ratio of 5:1 for the (nn)-direction. Merazzo et al have argued that for very low aspect ratios magnetic charges both at the antidot edge and the film surface can be of equal magnitude, effectively lifting any preference for an in-plane orientation of the magnetization [19]. Theoretical investigations into the anisotropy of antidot lattices by both Monte–Carlo simulation, e.g. by Ambrose and Stamps [20], and micromagnetic simulation, e.g. by Van de Wiele et al [21] also show that the in-plane orientation preference of the magnetization in a thin film antidot lattice can be, at least partially, lifted. Additionally, magnetic surface anisotropy contributes to the partial out-of-plane magnetization found here [22, 23].

Therefore the easy axes of the system do not necessarily need to be aligned completely in-plane and may be deflected partially out of the surface plane. In antidot arrays the formation of magnetic domains with a remarkably large out-of-plane component is coupled to the in-plane magnetization and its history. This coupling is plausible as there is no pure out-of-plane magnetization, but only an overall deflection of approximately 15° and may be related to a relaxation in pseudo-saturation into an easy axis that is aligned slightly out-of-plane. In a simple picture with a perfectly ordered system without any additional energy contributions a perpendicular deflection of the easy axis should have two-fold symmetry with respect to the surface normal, leading to an undetermined sign, i.e. up or down, of the perpendicular magnetization component. Yet, in our measurements we find that for any given sample position this degeneracy is lifted and one axis is more favourable.

Reproducible movement of magnetic domains through the antidot lattice has been observed in our SXM measurements. This is caused by structural defects in both the iron thin film and the antidot lattice, e.g. hole size variation; ordering defects; missing holes; and so on. The reproducible movement of magnetic domains through the antidot lattice leads in to a recurring intermediate stray field landscape during every magnetization reversal cycle. Typically thin magnetic films show Néel type domain walls to avoid stray field losses from out-of-plane magnetization components [24, 25]. However, from micromagnetic simulations, shown in figure 5, we find solely Bloch type domain walls in nano-scaled antidot lattices that exhibit an out-of-plane magnetization. As these Bloch type domain walls contribute significantly to the stray field landscape, their position and movement during magnetization reversal may sufficiently influence the out-of-plane deflected magnetization components to determine their orientation and lift the two-fold

Figure 4. Fe L₃ edge XMCD contrast of x-ray micrographs under normal incidence of a hexagonal antidot lattice with 200 nm antidot periodicity and 100 nm antidot diameter in a 20 nm thick Fe film. An in-plane magnetic field of 240 mT is applied in horizontal direction, i.e. along the (nn)-direction for images (a) and (c) and along the (nnn)-direction for image (b). The positions of the holes are indicated as grey overlays.

Figure 5. Cross section of a domain wall in an antidot lattice during magnetization reversal with the colour code indicating (a) in-plane components and (b) out-of-plane components of the magnetization. The geometrical restraints of the antidots favour a Bloch type domain wall, resulting in an out-of-plane magnetization in the domain wall.
The positions of the holes are indicated as grey overlays.

Figure 6. Fe $L_3$ edge XMCD contrast of x-ray micrographs under 30° grazing incidence of a hexagonal antidot lattice with 200 nm antidot periodicity and 100 nm antidot diameter in a 20 nm thick Fe film. An in-plane magnetic field corresponding to the respective coercive fields is applied in horizontal direction, i.e. along the (nn)-direction for image (a) and along the (nnn)-direction for image (b). The positions of the holes are indicated as grey overlays.

3. Methods

The antidot lattices were produced by nanosphere lithography using commercial polystyrene (PS) nanospheres (Invitrogen) with a mean diameter of 205±8 nm as starting material [26, 27]. Close-packed monolayers of these PS spheres are deposited on SiO$_2$(300 nm)/Si(001) substrates for MOKE measurements and Si$_3$N$_4$(500 nm, membranes)/Si(100) substrates for SXM measurements by dip coating at an extraction velocity of 10 $\mu$m s$^{-1}$ and an angle of 60° between the substrate surface and the air-water interface. Details of this preparation procedure can be found elsewhere [2, 12]. The deposited PS spheres were etched by an oxygen plasma (dc bias: −85 V) for 120 s to reduce their diameter to 100±8 nm. It has been shown that the etching process does not affect the spherical shape of the PS particles [26, 27]. On top of these templates a 20 nm thick polycrystalline Fe film was deposited with a 2 nm capping layer under UHV conditions. The thin films for MOKE and SXM measurements were deposited by pulsed laser deposition and ion beam sputtering respectively and capped with Pt or Al. Finally, the PS spheres including their metal caps were removed by chemo-mechanical polishing. Measurements with 1° angular resolution were performed to check that the unstructured iron thin films prepared on both substrate types had identical magnetic properties and were magnetically isotropic within the surface plane.

MOKE measurements were conducted with a Durham Magneto Optics NanoMOKE3 equipped with an air cooled vector electromagnet, capable of generating in-plane fields up to 120 mT. In polar geometry the laser beam was focussed with an aspheric lens ($f = 11$ mm) onto the sample, resulting in an imaging resolution of 1 $\mu$m and an area of smaller than $5 \times 5 \mu m^2$ that contributes to the magnetic signal. The orientation of the antidot lattice with respect to the applied in-plane field was indicated by markers that have been previously cut into the sample by FIB milling. MOKE hysteresis loops were averaged over 20 to 50 consecutive measurements at a cycling rate of 1.7 Hz. Three different samples with several positions where used for measurements and the experimental setup was repeated 16 times, resulting in approximately 500 acquired hysteresis loops. Additionally, two FIB milled antidot lattices in cobalt and permalloy, that are not shown here, were used to reproduce these results.

SXM measurements were conducted at the MPI IS operated MAXYMUS full spot end station at the UE46-PGM2 beam line at the BESSY II synchrotron radiation facility. The samples were illuminated under normal incidence by circularly polarized light in an applied in-plane field of up to 240 mT that was a generated by a set of four rotatable permanent magnets [28]. The photon energy was set to the absorption maximum of the Fe $L_3$ edge to get maximum XMCD contrast for imaging. The transmission through the holes, where there is no material and thus no x-ray absorption, was used as an internal intensity $I_0$ reference to normalize the measured intensities to the incident beam intensity. Intensities were locally averaged using a Gaussian filter in ImageJ [29]. Images taken at different photon helicities were registered using ImageJ with TurboReg [30]. Two individual samples were used and the measurement was repeated 19 times at different positions and for different orientations of the antidot lattice.
Micromagnetic simulations of magnetization reversal process of the hexagonally ordered Fe antidot arrays were performed using OOMMF software [31]. The antidot diameter was 100 nm and the centre to centre distance was 200 nm. The total size of the modelled system was 1000 nm × 1038 nm × 20 nm, the discretization was 2 nm × 2 nm × 10 nm and 2D periodic boundary conditions were used. The bulk material parameters of Fe were used: namely exchange constant $A = 2.1 \times 10^{-11}$ J m$^{-1}$, saturation magnetization $M_r = 1.7 \times 10^6$ A m$^{-1}$, and cubic anisotropy constant $K_u = 4.8 \times 10^5$ J m$^{-3}$. The polycrystallinity of the Fe antidot array was implemented by generation the Voronoi diagram of a random 2D map of 2000 grains and then the random orientations of the axes of the cubic anisotropy were assigned to each grain [32]. The magnetic field was applied in plane along (nn)-direction.

4. Conclusions

We have shown that geometric frustration in nano-scaled antidot lattices in thin iron films leads to an out-of-plane magnetization even for a magnetic field applied in the sample plane. This deflection could be estimated to an angle of 15° and is strongly linked to the in-plane magnetization reversal process of the sample.

The out-of-plane orientation, i.e. up or down, appears to depend on the stray field landscape history of the sample during magnetization reversal. These antidot lattices exhibit Bloch type domain walls which significantly contribute to the intermediate stray field landscape. Thus, the (nn)-direction that has undergone a more chaotic magnetization reversal with a multitude of fragmented domains and domain walls exhibits no long range out-of-plane order. On the other hand the (nn)-direction that reversed solely by domain wall movement exhibits out-of-plane domains with a size in the µm range as the moving domain wall pulls the deflected magnetization in one direction. This can be seen as a local easy axis that is slightly tilted with respect to the surface plane.

The transduction of in-plane magnetic fields to an out-of-plane magnetization signal may open up new ways to magnetic sensing or spintronics applications on the nanoscale by combining two devices that need different orientations of the magnetic signal. Furthermore, the transformation of in-plane magnetic information to an out-of-plane magnetic signal may advance 2D magnetic logic to the third dimension.

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