Polarized neutron studies on exchange-bias micro-stripe pattern

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Abstract. In the present study we examined the results of competing interactions in laterally exchange biased modulated thin films. For studying the magnetic interactions between neighboring magnetic elements we designed a model system. Considering that the model system should be of technological interest on the one hand, and should allow the application of several different and efficient experimental techniques for complementary analysis on the other hand, we have chosen films with in-plane stripe-like magnetic domains. The evolution of the magnetic domain structure along the hysteresis loop was analyzed with a combination of experimental techniques: magneto-optical Kerr effect, Kerr microscopy, polarized neutron reflectometry, and off-specular scattering of polarized neutrons with polarization analysis.

Due to the competition between interfacial exchange bias at the ferromagnetic-antiferromagnetic interface and intra-layer exchange interactions between neighboring ferromagnetic stripes the alignment is not perfectly antiparallel, but is periodically canted with respect to the stripe axis. Thus the net magnetization of the ferromagnetic film turns almost perpendicular to the stripes. At the same time the projection of the magnetization vector onto the stripe axis has a periodically alternating sign. The experimental observations are explained and quantitatively described within the frame of a phenomenological model. The model defines conditions which can be used for tailoring nano- and micro-patterned EB systems with different types of magnetic order.

1. Introduction
Patterned magnetic materials are extremely important for achieving maximum package densities in magnetic recording media. We used non-topographic magnetic patterning as an alternative approach to topographic patterning resulting in a local variation of only magnetic properties of thin films, which otherwise remain structurally homogeneous and maintain flat surfaces. With such systems one can avoid, e.g. the problem of very small feature sizes, where the long-term thermal stability of the magnetic elements is lost due to superparamagnetic fluctuations\cite{1}. For non-topographic magnetic patterning the exchange bias (EB) effect between ferromagnetic (F) and antiferromagnetic (AF) bilayers can be used \cite{2}. In those films the magnetization of the ferromagnet locally is pinned differently to the antiferromagnet at the interface implying that the complete film will adopt this lateral varying magnetization.

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We applied two routes for locally changing the EB. First, we prepared a CoFe/MnIr film by using Ion Beam Induced Magnetic Patterning (IBMP) \cite{3}\cite{4} for changing the EB between F and AF layers in one part of the film, while keeping the EB properties of the untreated regions unchanged. Second, we used a slightly changed film structure consisting of an extended soft ferromagnetic NiFe layer locally exchange biased to a periodic antiferromagnetic MnIr stripe array and studied several films with varied ferromagnetic film thickness and stripe width.

For a detailed study of the magnetic interactions between neighboring magnetic elements in a continuous film we designed model films which allowed the efficient use of different experimental techniques for complementary analysis. Therefore, we did not approach very small length scales but designed the magnetic elements to be in the micrometer range and used methods as the magneto-optical Kerr effect (MOKE), Kerr microscopy (KM) and polarized neutron reflectometry (PNR).

From the study of the first sample we found that while the EB in neighboring striped lateral areas was set antiparallel to each other the magnetization of each stripe appears to be tilted with respect to the EB frozen-in field so that the stripe magnetizations are aligned not completely antiparallel to each other. The reason is the additional ferromagnetic exchange interaction between the striped regions which favors a parallel orientation between the magnetization in the film volume\cite{5}. With the second set of samples we were able to investigate the overall remagnetization process in dependence of different EB and F exchange contributions.

2. Experimental Details

2.1. Sample preparation

The first sample studied is an EB Co\textsubscript{50}Fe\textsubscript{30}(28 nm)/Mn\textsubscript{83}Ir\textsubscript{17}(15 nm) F/AF bilayer with buffer layers Cu(28 nm)/SiO\textsubscript{2}(50 nm) on Si(111) and a TaO-Ta(9 nm) top layer prepared by magnetron sputtering. The initial EB direction was set by field cooling in a magnetic in-plane field of 1 kOe after an annealing step for 1 h at 275 °C which is above the blocking temperature of the antiferromagnetic material. Subsequently the film was covered by a photoresist, which was patterned into equally spaced stripes of width \(d = 2.5 \mu m\) and a periodicity of \(\Lambda = 5 \mu m\). The sample was bombarded with He\textsuperscript{+} ions with a fluency of \(1 \times 10^{15} \text{ ions/cm}^2\) at 10 keV. The resist thickness of 0.5 \(\mu m\) is sufficient to protect the covered regions from the He\textsuperscript{+} exposure. During bombardment a magnetic field of 1 kOe aligned opposite to the initial EB direction was applied. This resulted in a stripe-like pattern with alternate sign of the unidirectional anisotropy, and hence the sign of the EB in neighboring stripes\cite{6}.

The second set of samples are polycrystalline full Mn\textsubscript{77}Ir\textsubscript{23}(t\textsubscript{AF})/Ni\textsubscript{81}Fe\textsubscript{19}(t\textsubscript{F}) films deposited by magnetron sputtering on oxidized Si-wafers with a buffer layer of 4 nm Ta and covered by a TaO-Ta(3 nm) top layer. In order to obtain a 2-dimensional lateral modulation of the unidirectional exchange anisotropy, each layer stack was subjected to a subsequent lithography patterning and ion etching to remove the Ta cover layer in stripe-like parts of the sample. Therefore, only the uncovered parts of the sample were subsequently exposed to air for undergoing an oxidation process of MnIr resulting in a superimposed stripe array of MnIr and MnIrO (for more details see Ref. \cite{7}). MnIr is antiferromagnetic while MnIrO is non-magnetic. At a temperature of 250°C, which is above the blocking temperature of the system, an external field of 10 kOe was applied to set the unidirectional anisotropy direction of the F-AF stripes to be along the stripe axis. The resulting EB patterned film of alternating stripes with nominal TaO – Ta/Mn\textsubscript{77}Ir\textsubscript{23}(t\textsubscript{AF})/Ni\textsubscript{81}Fe\textsubscript{19}(t\textsubscript{F}) and MnIrO\textsubscript{3}/Ni\textsubscript{81}Fe\textsubscript{19}(t\textsubscript{F}) comprises exchange biased (AF-F) stripes and unbiased ferromagnetic stripes (F) through an underlying extended ferromagnetic layer. The stripe period \(\Lambda\) was chosen to be 4 \(\mu m\) and 12 \(\mu m\), respectively, to thus vary the amount inter-stripe interfaces. The magnitude of the unidirectional anisotropy was tuned by adjusting the ferromagnetic layer thickness \(t\textsubscript{F}\) to be either 20 nm or 30 nm while the antiferromagnetic layer thickness \(t\textsubscript{AF}\geq 7 \text{ nm}\) was always above the critical thickness for the
existence of EB.

2.2. Magneto-optical Kerr effect
We have analyzed the magnetization reversal of the first sample with a vector-MOKE set up as described in Ref. \cite{8}. For measuring the magnetization reversal process of the second set of samples we employed inductive magnetometry to map the macroscopic properties.

Further insight into the microscopic rearrangement of magnetization during cycling with an external field along the EB axis was achieved by a high resolution magneto-optical Kerr microscope (KM) \cite{9} that is sensitive to directions orthogonal to the field \cite{10}. The KM images provide good input data for constructing a remagnetization model which was then used to refine the physical parameters required for fitting polarized neutron reflectometry (PNR) measurements.

2.3. Polarized neutron scattering
Neutron scattering experiments were carried out with the ADAM reflectometer at the Institut Laue-Langevin, Grenoble, France \cite{11}, and with the HADAS reflectometer at the FRJ-2 reactor in Jülich, Germany. The measurements were performed with fixed neutron wavelengths of $\lambda = 0.441$ and 0.452 nm, respectively. To apply the external field $H$ along the stripe axis, an electromagnet with the field direction parallel to the sample surface and parallel to the neutron polarization and perpendicular to the scattering plane was used. The incoming neutron polarization was either set parallel (+) or antiparallel (-) to the external magnetic field. For specular reflectivity measurements, non-spin-flip (NSF) reflectivities $R^{++}$ and $R^{--}$ as well as neutrons with flipped polarization after the reflection, i.e. two spin-flip (SF) intensities $R^{+-}$ and $R^{-+}$, were analyzed.

The magnetic induction vector $\mathbf{B} = \mathbf{H} + 4\pi \mathbf{M}$ (which is mainly given by the magnetization vector) is assumed to lie in the sample plane (in-plane shape anisotropy). Furthermore, $\mathbf{B}$ may have an angle $\gamma$ with respect to the external magnetic field. We assume that a monochromatic neutron beam incident onto the sample surface under the angle $\alpha_i$ is scattered under the glancing angle $\alpha_f$, so that for specular reflection $\alpha_i = \alpha_f$.

3. Experimental results
The first sample (CoFe/MnIr) was studied by vector-MOKE measurements with the field applied along the EB axis which is parallel to the IBMP magnetically patterned stripe edges, see Fig. 1(a). A two step magnetization reversal is clearly visible in the longitudinal hysteresis, corresponding to a successive remagnetization of the two regions with alternative directions of the EB field. The transverse hysteresis loop in Fig. 1(a) shows a considerable transverse component of magnetization with plateaus in the field range where we expect an antiparallel alignment of the magnetization in irradiated and non-irradiated parts of the sample.

A detailed KM analysis of this sample was recently published by McCord et al.\cite{10}. With the microscope tuned to longitudinal magnetic contrast it was shown that starting from saturation magnetization the reversal proceeds through an antiparallel alignment of magnetization by head-on domain wall propagation along the stripes. In addition, the transverse magnetization contrast provides a rather detailed insight into the microscopic arrangement at different stages of the remagnetization process. Some representative examples of KM images taken with the Kerr sensitivity set close to the transverse direction, are reproduced in Fig. 1 (b)-(e). The images were taken in the ascending loop branch at fields just below the first reversal at $H_{c,1}$ (b), just above $H_{c,1}$ (c), in the antiparallel state between $H_{c,1}$ and $H_{c,2}$ (d), and at the second reversal at $H_{c,2}$ (e). Ripple domains can also be observed in Fig. 1 (b), (c) and (e) in stripes which are close to reversal. Note that rippling is typical for Co and Co rich CoFe alloys, \cite{12, 13} and in our sample always occurs around both coercive fields $H_{c,1}$ and $H_{c,2}$. 


Figure 1. (a) Vector-MOKE hysteresis loops of the CoFe/MnIr sample measured with the field applied along the EB axis and showing the longitudinal and transverse components of the magnetization vector. (b)-(e) Corresponding longitudinal Kerr images taken along the ascending magnetization loop.

Figure 2. (a) Magnetic hysteresis loop of the MnIr - MnIrO/NiFe (20 nm) film with $\Lambda = 4 \mu$m from the second set of samples. (b)-(e) Corresponding longitudinal Kerr images along the ascending magnetization loop.

In Fig. 2 the remagnetization process of one of the EB modulated films of the second set of samples with a thickness of $t_F = 20$ nm and a stripe period of $\Lambda = 4 \mu$m is presented. The inductively measured hysteresis loop and corresponding high-resolution Kerr images are depicted in Figures 2(a) and (b-e), respectively. The film structure also exhibits a two-step switching process, typical for such a hybrid magnetic two-phase system. Coming from negative magnetic fields, first, below 10 Oe the free F stripe magnetization starts to ripple (b), followed by the reversal of those stripes by head-on-domain wall motion (c). On the hysteresis step, a rather continuous change of the magnetization is observed, indicating rotation and relaxation.
Figure 3. (Color online) Polarized neutron reflectivity measurements performed at the CoFe/MnIr sample at different applied magnetic field. The symbols present measurements of non-spin flip reflectivities \( R^{++} \) and \( R^{--} \) and one spin flip reflectivity \( R^{+-} \). The lines represent fits to the data points. The sign of the field value corresponds to that in MOKE measurements in Fig. 1.

along the positive field direction. During the second step at 40 Oe, the inter-stripe domain walls move into the exchange biased stripes. Accompanied by head-on-domain wall motion (e) the pinned magnetization is finally reversed. During this reversal process, stripes of the same magnetic properties reverse in a correlated manner such that so called quasi-domains[14, 7] (or hyper-domains[15]) occur.

Specular polarized neutron reflectivity was measured at several field values on the ascending and descending branches of the hysteresis loops. In Fig. 3, several representative specular reflectivity curves of the CoFe/MnIr sample are displayed. The NSF reflectivity curves in the first row look quite similar to those in the last one, if the NSF reflectivity \( R^{++} \) in the latter row is substituted by its counterpart \( R^{--} \), and vice versa. This symmetry is just due to the opposite magnetization state of the sample in the upper and lower rows. The SF reflectivity \( R^{+-} \) in the first row, however, is stronger than that measured in saturation, where it is mostly due the limited efficiency of the polarization devices. An appreciable SF signal in the specular channel signifies that the magnetization averaged over the coherence length is tilted by the angle \( \gamma \neq 0 \) with respect to the polarization direction, and hence, to the stripe axis.

For measurements carried out at field value between \( H_{c,1} \) and \( H_{c,2} \) (middle row of Fig. 3), the NSF reflectivity curves almost merge together. The spin asymmetry \( R^{++} - R^{--} \) is close to zero, indicating that the mean magnetization within the neutron coherence range has almost no projection onto the direction of the applied field, in agreement with observations from the
Figure 4. (Color online) Experimental and simulated maps of the polarized neutron scattering intensity on a logarithmic scale measured on the EB Co$_{70}$Fe$_{30}$(28 nm)/Mn$_{83}$Ir$_{17}$(15 nm) F/AF bilayer at the descending loop branch at a magnetic field of -27 Oe. The intensities are plotted as a function of $\alpha_i$ and $\alpha_f$.

longitudinal hysteresis loop in Fig. 1 (a) for the plateau region. Such results can be interpreted in favor of an antiparallel alignment of the magnetization vector in neighboring stripes. However, at the same field values we observe a very strong SF signal comparable to the NSF intensities. This is an unambiguous proof for the fact that the mean magnetization over a number of stripes in the coherence length is tilted by a large angle $\gamma$ with respect to the polarization direction, and hence, to the stripe axis. Taking into account the degeneracy of NSF channels one concludes that $\gamma \approx 90^\circ$. The existence of a large magnetization component perpendicular to the applied field was already noted in the transverse MOKE and KM data.

A quantitative information about the magnetization distribution over the stripes is gained via fitting the specular PNR data to a theoretical model which takes into account that due to variations in the tilt angles $\gamma = \gamma + \Delta \gamma$ against the applied field one may observe local periodic variations $\Delta \gamma$ with respect to the mean value $\gamma$ within the coherence length. These deviations reduce the mean magnetic optical potential and modify the reflectivity profiles with respect to that in saturation. The ratio between SF and NSF reflectivities is regulated by the mean values of $\langle \cos \gamma \rangle$ and $\langle \sin^2 \gamma \rangle$, where angular brackets denote additional incoherent averaging. Such an averaging of reflected intensities is due to the fact that the coherence area of the neutron beam covers only a small portion of the sample surface and magnetization direction, and hence the tilt angle $\gamma$, may vary from one coherence area to the next.

The fitting procedure, basically determining $\langle \cos(\Delta \gamma) \rangle$, $\langle \cos \gamma \rangle$ and $\langle \sin^2 \gamma \rangle$ was carried out using an originally developed least-squares software package [16], which allows simultaneous evaluation of all four measured reflectivities in one cycle. It is based on the super-iterative algorithm, [17] which generalizes the Parratt algorithm [18].

It should be stressed that from MOKE and specular PNR results alone one cannot draw conclusions about the origin of the microscopic arrangement of magnetic states along the hysteresis loop. However, from fits to the Bragg diffraction and diffuse scattering of polarized neutron intensity maps (one representative example for experimental and simulated maps are shown in Fig. 4) we find a periodic deviation of the magnetization vector in neighboring stripes with an angle in the “antiparallel state” of only 120° (instead of expected 180°) to each other corresponding of $\Delta \gamma = 60^\circ$. In principle, deviation angles $\Delta \gamma$ consist of two parts. The first one is due to periodic alternation of $\Delta \gamma$. It contributes into superstructural off-specular Bragg reflection. The random fluctuations around this periodic alternation is a source of diffuse scattering.

In the maps in Fig. 4 the specular reflection ridge runs along the diagonal, where $\alpha_i = \alpha_f$. Besides little diffuse scattering Bragg diffraction concentrated along curved lines $\cos \alpha_i - \cos \alpha_f \approx$
Figure 5. (Color online) Results of the fits to the PNR data of the second set of samples. The graph shows the results with respect to $\Delta \gamma$ as function of ferromagnetic film thickness and EB stripe width $d_{st}$.

$n(\lambda/\Lambda)$ can be seen at $\alpha_i \neq \alpha_f$, where $n$ denotes the order of diffraction and $\Lambda = 5 \mu m$ is the period. Strong Bragg reflections for $n = \pm 1$ can be recognized, whereas for $n = \pm 2$ they are suppressed. This is caused by the structure factor, resulting from the equal width of adjacent magnetic stripes in a unit cell.

Similar results were also found from the second set of samples. For all three samples in the “antiparallel state” at the hysteresis step, the mean magnetization is canted about 90° away from the stripe axis. The sample with the lower unidirectional anisotropy of the biased stripes, i.e. a F thickness of 30 nm, and smallest pattern period of $\Lambda = 4 \mu m$ shows the smallest splitting of the neighboring stripe magnetizations of $\Delta \gamma = 18^\circ$. With increasing the period to $\Lambda = 12 \mu m$ and thus decreasing the number of stripe interfaces, the perpendicular magnetization component for both stripe types is reduced. For this case, the angle $\Delta \gamma$ is 38°. Keeping the stripe period constant but increasing the unidirectional anisotropy of the AF-F stripes, $\Delta \gamma$ also increases, now to 50°. The results from the three samples are summarized in Fig.5.

4. Discussion
We developed a simple phenomenological model explaining the reason for the intermediate state stable below the coercive field characterized by a canted domain structure in easy axis configuration. It is explained by the interplay between interfacial and bulk exchange coupling. The model also gives an estimate for the range of physical parameters (stripe width and film thickness) where one can expect a pure antiferromagnetic ordering of the stripe magnetization (the model is described in detail in Ref. [5]).

The basic result of the model is that the EB exchange field contribution is proportional to the number of interfacial ions, i.e., in first approximation being proportional to the width of the stripes $d$ times its length $l$. On the other hand, the bulk F exchange energy across the F domain wall is proportional to the number of sites belonging to the F domain wall running through the F film, i.e., in first approximation being proportional to the film thickness $t$ times stripe length $l$. Therefore the ratio between bulk and interfacial contributions is roughly proportional to $(t \cdot l)/(d \cdot l) = t/d$. This means that one can suppress the undesirable tilt instability by either increasing the width of the stripes or reducing the F film thickness, while keeping interfacial and bulk F exchange couplings unchanged.
5. Summary
In the first set of samples we have investigated a continuous ferromagnetic CoFe film alternately exchange-biased (with positive and negative EB, respectively) to an antiferromagnetic MnIr layer. In the second set of samples we studied three continuous NiFe layers with variable F film thickness alternately exchange-biased to MnIr layer or free to rotate. We have analyzed the magnetization reversal mechanism of these films in detail, which exhibit rich hysteresis and domain structures due to competing interfacial and intralayer exchange interactions. For a given set of parameters, including interfacial and bulk exchange coupling, stripe width, and film thickness, the intermediate state is stable below the coercive field and is characterized by a canted domain structure which results in a macroscopic magnetic moment directed perpendicular to the stripe direction and the EB axis. A simple phenomenological model suggests a reason for this phenomenon and gives an estimate for the range of physical parameters where one can expect a pure antiferromagnetic ordering of the stripe magnetization. This may provide directions for further advances in the design of magnetic micro- and nano-patterns with tailored properties.

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