Direct Imaging and Determination of the Uncompensated Spin Density in Exchange-Biased CoO/(CoPt) Multilayers

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Magnetic force microscopy (MFM) measurements were performed on an exchange-biased CoO/(CoPt) multilayer sample at 7.5 K. Applying an external magnetic field of up to 7 T saturates the ferromagnetic layer and the remaining uncompensated antiferromagnetic spins at the antiferromagnet-ferromagnet interfaces are imaged with high lateral resolution. The coupling between the uncompensated spins and the spins in the ferromagnet are found to be antiferromagnetic. Quantitative analysis of the MFM images revealed that 7% of the spins at the interface are uncompensated and contribute to the exchange biasing.

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The balance between exchange, anisotropy, and dipolar energies usually determines the domain structure in ferromagnetic (FM) films. However, coupling a ferromagnetic film to an antiferromagnetic (AF) layer can significantly alter the hysteresis processes and domain structure due to the additional FM-AF interfacial exchange [1,2]. Exchange-biasing results from field cooling such a sample through the Néel temperature (T_N) of the AF. The FM-AF interaction provides an effective bias field that gives rise to a horizontal shift in the hysteresis loop. Interaction with AF layers can also increase the anisotropy of the FM layer and often enhances the coercive fields. As such, exchange biasing has become an important tool for controlling domain formation in magnetic devices [3]. While routinely exploited, the microscopic origin of exchange bias is still open to debate. It is generally believed that the bias is related to defects in the antiferromagnetic order that leads to uncompensated spins inside the AF layers or at the AF interfaces [1,4]. There have been elegant studies that have measured the presence of uncompensated spins in thin AF layers [4–8] and the magnitude of these spins is consistent with the measured exchange bias. In particular, Ref. [8] shows that the biasing is correlated to the uncompensated spins that are pinned to the AF layer.

In this Letter we describe quantitative high-field magnetic force microscopy (MFM) imaging of the spatial distribution of uncompensated spins in CoO layers in a perpendicularly exchange-biased CoO/(CoPt) multilayer [9,10]. The sample is cooled through the T_N of CoO in a well defined striped domain pattern [11] which locally sets the CoO. External fields up to 7 T are then applied to saturate the FM layers and the remaining uncompensated spins are imaged. We find that the uncompensated spins replicate the original ferromagnetic domain structure but are antiferromagnetically aligned to the ferromagnetic moments. Upon field reversal the original domain structure returns showing a direct link between the uncompensated spins and the local bias.

A CoO(10 Å){[Co(4 Å)/Pt(7 Å)]$_4$Co(6 Å)CoO(10 Å)}$_{10}$ multilayer deposited onto a Si_3N_x-coated Si substrate by magnetron sputtering was used for all experiments presented here. Details of the sample production are found in [10]. The high interface anisotropy of the CoPt interfaces leads to uniaxial perpendicular magnetic anisotropy of the FM layers. The average magnetic properties are obtained by SQUID magnetometry. The domain structure is characterized by MFM. The MFM is a custom built low-temperature scanning force microscope [12] that images at 7.5 K in magnetic fields variable between -7 T and $+7$ T applied perpendicular to the sample surface. All images were acquired using a cantilever with an ultrahigh aspect ratio tip [13] coated with a thin FM layer. It had a resonance frequency of 61437 Hz and a stiffness of 0.5 N/m. In order to image the magnetic structure, the change of the cantilever resonance frequency is recorded when the tip is scanned over the sample at a constant tip-sample distance.

The AF nature of the CoO in the multilayer becomes apparent upon field cooling the sample below its T_N. Figure 1(a) shows the hysteresis loop measured after field cooling (filled squares) in 1.0 T that saturates the sample in the positive field direction. The resulting hysteresis fc loop shows an exchange-bias field of -105 mT with a coercive field of 376 mT. Figure 1(b) shows the temperature evolution of the exchange bias and the coercivity. The loss of exchange bias identifies a blocking temperature of 200 K while the coercive field enhancement suggests a

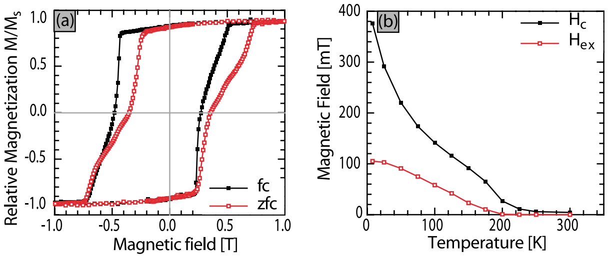

FIG. 1 (color online). (a) Hysteresis loops measured perpendicular to the sample after field cooling (filled squares) the sample in 1.0 T and zero-field cooling (open squares) in a stripe domain pattern. The hysteresis loops were measured from ± 5 T and were featureless above 1 T. The field-cooled loop shows an exchange-bias field of -105 mT and a coercive field of 376 mT. (b) Temperature evolution of the exchange bias and the coercivity after field cooling the sample.

somewhat higher T_N of about 250 K similar to that found in [9]. Cooling exchange-bias samples with the FM layer in different domain states has been studied by a variety of authors [14–16]. It is generally found that the spin structure of the AF layer during cooling is determined mainly by the local FM domain order. For the present samples, demagnetizing the films at room temperature with an in-plane field results in an aligned stripe domain pattern [11]. Cooling the sample through T_N imprints this pattern into the CoO layers. It is then expected that the sample will have locally biased regions depending on the local magnetic direction of the FM layer [14]. The measured hysteresis loop after cooling in the stripe domain pattern is shown in Fig. 1(a) (open squares). We observe the same coercivity enhancement as for the field-cooled sample but no macroscopic bias, as expected.

Shown in Fig. 2 is the field evolution of the domain pattern after cooling in the domain pattern. The initial pattern is depicted in Fig. 2(a). The domains have an average width of 83 nm and stretch over several micrometers in length. Because of the strong perpendicular anisotropy of the CoPt layers these domains have a magnetization either parallel or antiparallel to the sample normal. When a field of $+650$ mT is applied parallel to the initial magnetization direction of the MFM tip, the width of stripe domains with a magnetization direction opposite to the applied field (white domains) becomes smaller and their length is reduced [Fig. 2(b)]. This occurs either by an abrupt motion of an existing end of a domain [see circles 1 and 2 in Figs. 2(a) and 2(b)], or by the partition of an existing domain into two parts and predominantly occurs at domain bifurcations. According to the magnetization loop [Fig. 1(a)], in fields > 800 mT, the FM layers are saturated and no domain structure is expected to be visible. Surprisingly, a faint stripe domain structure with a 20 times smaller and granular contrast remains visible and persists to fields up to 7 T [Fig. 2(c)]. The total area of white domains equals that of the black domains as in the initial domain pattern [Fig. 2(a)], but the contrast is inverted. This is confirmed when comparing a line scan

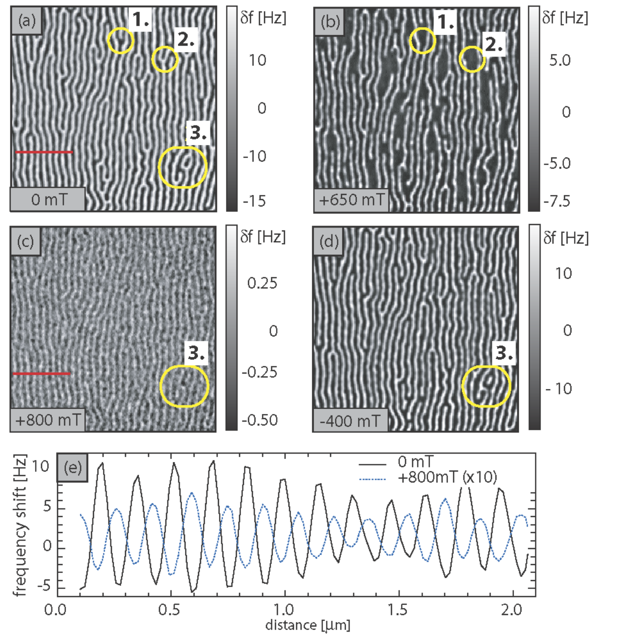

FIG. 2 (color online). Evolution of MFM images in an applied field after cooling in a stripe domain pattern. All images are $5 \times 5\ \mu$m. (a) MFM image of the original FM stripe domains after zero-field cooling. (b) In an applied field of $+650$ mT the white domains shrink in width and length. (c) At $+800$ mT the FM layers are saturated, nevertheless stripe domains with weak contrast remain visible and persist in fields up to 7 T. (d) Reversing the field to -400 mT the original domain pattern is recovered. However, due to the flipped tip magnetization the contrast is reversed when compared to (a). (e) Line scans from identical locations in (a) and (c) shown by the red lines. The high field image has inverted contrast compared to the original domains.

[Fig. 2(e)] taken at identical locations in the images of Figs. 2(a) and 2(b). They show an antiphase contrast. Reducing the field to zero leaves the faint domain structure unchanged but the contrast is slightly increased.

When the field is inverted and ramped to negative values, the contrast enhances again as domains are nucleated in the FM layers. At -400 mT [Fig. 2(d)], the original stripe domain pattern [Fig. 2(a)] is recovered, but the contrast is reversed because the magnetization of the MFM tip has flipped at a field of -65 mT. Circle 3 in Figs. 2(a) and 2(d) allows a comparison of the domain structure at one specific location. Upon raising the field to high negative values (-7 T) the faint domain pattern shown in Fig. 2(c) becomes again visible but is reversed due to the inverse tip magnetization. In subsequent field cycles, the original domain structure Fig. 2(a) can always be recovered.

From the data presented above the following conclusions can be drawn: (i) The hysteresis loop shows that the CoPt multilayer appears saturated in fields ≥ 800 mT. Therefore the magnetic contrast visible in Fig. 2(c) results from uncompensated spins that are pinned by the AF layers and persist to fields as high as 7 T. (ii) The contrast inversion found when comparing line scans from Figs. 2(a) and 2(c), shown in Fig. 2(e), reveals a collinear but antiferromagnetic alignment of the spins in the FM layer and the uncompensated spins at the interfaces. The inversion of the image rules out that the contrast at high field arises from incomplete saturation of the FM layers which should have the same phase as the original domains. (iii) The granular structure of the domain pattern shown in Fig. 2(c) is reproducible and its length scale is comparable to the structural grain sizes (15–25 nm). This suggests that the distribution of the uncompensated spins is not spatially uniform but is linked to the granular microstructure of the film.

To further elaborate on the above conclusions, the MFM data was quantitatively analyzed. First the cantilever was calibrated by a method developed by van Schendel *et al.* [17]. A sample with perpendicular magnetic anisotropy and artificially written domains was used to determine the tip transfer function $T_{\rm tip}(\mathbf{k})$. The MFM signal of a sample with an arbitrary but known magnetization direction and magnetization pattern can be calculated in Fourier space by

$$\mathrm{MFM}(\mathbf{k}) = T_{\rm tip}(\mathbf{k}) \cdot P(\mathbf{k}) \cdot H_z^{\rm tot}(\mathbf{k}), \tag{1}$$

where $\mathbf{k} = (k_x, k_y)$ is the in-plane wave vector , $H_z^{\rm tot}(\mathbf{k})$ is the Fourier transform of the z component of the stray field, and $P(\mathbf{k})$ is the transfer function containing all measurement parameters. To calculate the stray field of one single domain pattern existing in all the FM layers, the sample is considered as a collection of ten ferromagnetic thin films located at the tip-sample distances $d_{10} = d_{\rm AF} + d_{\rm ts}$ and $d_{i-1} = d_i + d_{\rm FM} + d_{\rm AF}$ for $10 > i \geq 1$. Here $d_{\rm AF} = 1$ nm denotes the thickness of an AF layer, $d_{\rm ts} = 30.4$ nm is the separation between the tip and the sample surface, and $d_{\rm FM} = 5$ nm is the thickness of a FM layer. From [17] we derive that the spatial Fourier components of the total field become

$$H_z^{\rm tot}(\mathbf{k}) = \frac{M_F}{2}(1 - e^{-kd_F}) \sum_{i=1}^{10} e^{-kd_i}, \tag{2}$$

where $M_F = 550$ kA/m is the saturation magnetization of the FM layers and d_i is the distance between the tip and the ith FM layer of the sample. Once $T_{\rm tip}(\mathbf{k})$ and $P(\mathbf{k})$ are known, the MFM image shown in Fig. 3(a) can be modeled. In a first step, the magnetization pattern of the sample is estimated [17] [Fig. 3(b)]. The up direction can be distinguished from the down direction by choosing an appropriate discrimination level for the frequency shift image [Fig. 3(a)]. When calculating the MFM contrast arising from a domain structure in the FM layers, the effect of the AF layers on the MFM signal is neglected. In a second step, the sample stray field (not shown here) is calculated from the magnetization pattern Fig. 3(b) using Eq. (2). In a final step, the MFM signal [Fig. 3(c)] is calculated from the stray field pattern using Eq. (1). The modeled and measured patterns show the same structures, and the gray scale distribution and the extrema of the frequency shift match extremely well.

When a sufficiently strong magnetic field is applied the FM layers become saturated. Since a homogeneously magnetized thin film does not generate a stray field, the magnetic stray field originates from uncompensated spins pinned at the AF-FM interfaces. To calculate the magnetic charge density at these interfaces a quantitative evaluation of the MFM contrast similar to that described above for the FM layers is applied to the AF layers. Again a magnetization pattern [Fig. 4(b)] is derived from the measured MFM data [Fig. 4(a)]. In contrast to the simulations performed for the FM layers, the magnetic charge density at the AF-FM interfaces depends on the density of uncompensated spins. Therefore the density of uncompensated spins in the model was iteratively adjusted to obtain the best agreement between the measured [Fig. 4(a)] and the modeled [Fig. 4(c)] images. The best agreement is obtained for a surface charge density $\sigma_{\rm CoO} = 118.4$ kAm.

CoO is an atomically layered antiferromagnet with alternating sheets of oxygen and cobalt ions along the [111] direction that is perpendicular to the sample surface. The Co^{2+} ions carry a magnetic moment of $+3.8\mu_B$ [18]. Assuming a monolayer of uncompensated spins at each interface that contributes to the magnetic surface charge density, one calculates an expected surface charge density of 1813.2 kAm. Compared to the charge density found in the simulations we estimate 0.07 monolayers of

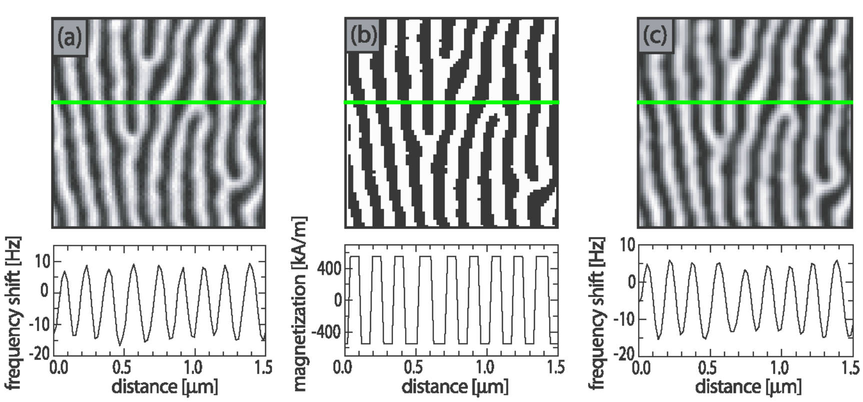

FIG. 3 (color online). (a) MFM image and typical cross section after zero-field cooling. (b) Magnetization pattern and cross section generated from the MFM image (a). The modeled (c) and measured (a) gray scale images agree extremely well.

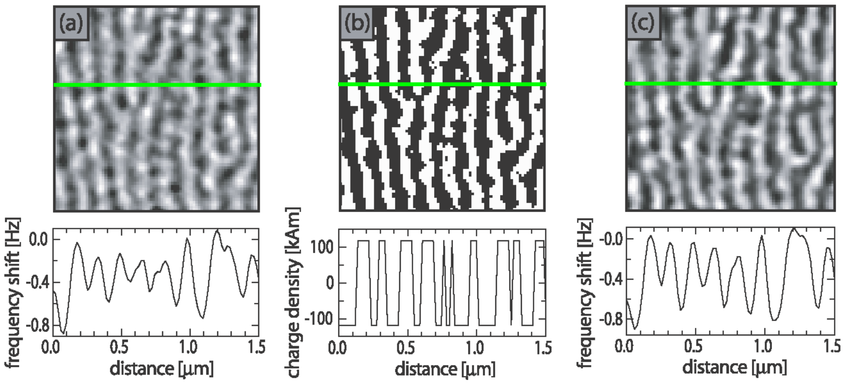

FIG. 4 (color online). (a) MFM image and a typical cross section at 800 mT. (b) Magnetization pattern and cross section generated from the MFM image (a). The modeled (c) and measured (a) gray scale images agree extremely well.

uncompensated spins at each interface [19]. The results compare well with the estimates of 0.04 monolayers for the Co/NiO system in [8] and the 0.01 monolayers for CoO/MgO multilayers [4].

In conclusion we have imaged the uncompensated spins at the AF-FM interfaces of a CoO/(CoPt) multilayer. Cooling the sample in a well defined domain state allows us to image the spatial variation of the uncompensated spins. Quantitative analysis of the MFM images reveals that about 7% of the interfacial spins are uncompensated. During reversal, the domain structure of the ferromagnetic layers exactly replicates the spatial variation of the uncompensated spins supporting the theory that they are responsible for the observed biasing. For the present sample the uncompensated spins are found to be coupled collinear but antiferromagnetically to the FM layers. The collinear alignment of the samples agrees with work by Nolting *et al.* [20] and is consistent with the model in [9] where the spins were assumed to align along the crystalline axis closest to the field cooling direction. In addition these results provide a direct measure of antiferromagnetic coupling between the FM and AF layers. Antiferromagnetic coupling of the AF and FM layers had been proposed to explain positive exchange bias observed in Fe/FeF_2 bilayers [21] but was not directly observed. The present finding suggests a superexchange mechanism for the AF-FM interface in which the layers are coupled via an indirect exchange across oxygen terminated AF surfaces. This is in contrast with the mean-field analysis of Parker *et al.* [22] that concluded direct exchange for the operating mechanism and Ohldag *et al.* [8] who found that for NiO the uncompensated spins were parallel to the ferromagnet. These differences could result from the different method of fabrication. In these studies the AF layer was deposited as an oxide, while for our samples the AF layer was formed by oxidation of the Co layer.

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