Spatially resolved magnetic reversal in a multilayered exchange bias system

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Abstract. We have observed the magnetic reversal of an exchange bias model system on both sides of the ferromagnet/antiferromagnet (FM/AFM) interface via nuclear resonant scattering of synchrotron radiation from \(^{57}\)Fe sensor layers. This method yields the spin direction and the fraction of uncompensated moments with nm depth resolution. The reversal of the ferromagnet along the easy axis proceeds via the formation of a domain structure that extends across the FM/AFM interface. This is responsible for archetypal exchange bias characteristics like the small magnitude of the bias and the asymmetric shape of the hysteresis loop.

Magnetic coupling effects constitute a unique possibility to tailor the properties of nanomagnetic systems. The most prominent example is the exchange bias effect where the magnetic reversal of a ferromagnetic layer is affected when brought into direct contact with a second layer with a pronounced magnetic anisotropy. The interface exchange coupling causes an anisotropy transfer into the ferromagnet (FM), which depends on the coupling strength and the net magnetization of the interface atoms and on the anisotropy of the antiferromagnet (AFM). A directed interface net magnetization of the AFM is created when the system is cooled through the Néel temperature of the AFM which results in a biasing of the FM magnetization and manifests itself in a shift of

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the hysteresis loop by the exchange bias field $H_{EB}$. This principle of a unidirectional exchange anisotropy is widely used in the magnetic recording industry to create a reference magnetization. The microscopic coupling mechanism, however, is still under controversial discussion.

Extensive research on exchange biased FM/AFM layer systems revealed a number of unusual magnetic properties of the FM (see [1, 2] and references therein) amongst which are the unexpectedly small shift and an asymmetric shape of the FM hysteresis loop. It was shown that both effects strongly depend on the magnetic field orientation relative to the anisotropy direction of the AFM and reach their maximum value for fields nearly parallel to the easy axis [3]. The simplest model where the FM is coupled to a rigid and fully uncompensated AFM surface [4] yields values of $H_{EB}$ about two orders of magnitude higher than the experimentally observed ones. A number of theories were developed to explain this discrepancy. Malozemoff [5] argues that interface roughness will cause magnetic domains with perpendicular domain walls in the AFM, which strongly lowers the interface net magnetization. Mauri predicts that the interface spin structure of the AFM will not be fixed during the reversal of the FM but a planar domain wall will wind up [6]. Both models deduce values of $H_{EB}$ close to the experimentally observed ones. The asymmetric shape of the FM hysteresis loop relates to different reversal modes on both branches like coherent rotation on one side and incoherent rotation or domain wall motion on the other side as it was measured with polarized neutron scattering techniques [7, 8] and Kerr microscopy [9]. It is expected that this effect is also caused by the interface domain structure of the AFM even though a general explanation is still missing [10].

Detailed information about the field-dependent spin structure of the AFM is necessary to understand the origin and magnitude of the exchange anisotropy. Unfortunately, depth-resolved investigations in this field are conceptually difficult. First, there are only a few experimental methods which are sensitive to the compensated magnetization of the AFM (polarized neutron scattering and x-ray magnetic linear dichroism) and model assumptions about the buried spin structure are necessary to derive their depth dependence from the experimental data [11, 12]. Additionally, it remains a challenge to determine the interface net magnetization in conventional systems, which is strongly affected by structural roughness.

In this paper, we investigate the magnetization reversal of an exchange bias model system which is characterized by a completely uncompensated FM/AFM interface in the magnetic ground state. Nuclear resonant scattering (NRS) of synchrotron radiation from ultrathin isotopic sensor layers is used to image the magnetic reversal depth resolved on both sides of the interface. This method is a very sensitive tool to probe the magnetization direction in thin films with a depth resolution in the nanometer range. Moreover, we show that the method can be applied to determine the fraction of compensated moments at the position of the sensor layer in the FM and in the AFM. Therefore, it is ideally suited for investigation of the exchange bias mechanism. Our results provide a qualitatively new picture of the reversal mechanism and the origin of the unidirectional exchange anisotropy.

The sample used here is sketched in figure 1(a). The AFM consists of an antiferromagnetically coupled superlattice of 14 bilayers natFe(3 nm)/Cr(1.4 nm) where every Fe layer represents an atomic layer of a crystalline AFM. The Cr thickness was chosen to coincide with the first minimum of the oscillatory bilinear coupling constant [13]. Due to the relatively large lattice constant (bilayer period) of the AFM, our system is much less sensitive to structural roughness than conventional (e.g. crystalline) systems are. To induce a unidirectional anisotropy into the AFM and to eliminate the lateral domain state in the (remanent) ground state which was detected via magnetic force microscopy and off-specular NRS on $^{57}$Fe/Cr.
multilayers, a hardmagnetic FePt buffer layer was used as a substrate. The FM is a Fe layer with a thickness of 6 nm. This is thin enough to obtain a relatively large bias field and thick enough for a sufficiently large Zeeman energy in an external magnetic field.

The polycrystalline layers were deposited on superpolished Si-wafers by rf sputtering. The FePt layer was prepared with a Fe_{55}Pt_{45} alloy sputtering target and flash annealed to 850 K for 14 min to form the hard magnetic L1_0 phase. A 3 nm thick Ta buffer layer suppressed grain growth during the thermal treatment to support smooth boundaries thus enforcing a high antiferromagnetic interlayer coupling in the adjacent Fe/Cr layer stack. X-ray reflectivity measurements showed that the rms interlayer roughness in the whole system is less than 0.3 nm.
MOKE hysteresis curves yielded a coercivity of the hardmagnetic FePt layer of 750 mT at room temperature and a saturation field $H_S$ of the multilayer AFM of 140 mT. A sputter-induced intrinsic uniaxial easy axis was determined from a set of hysteresis loops taken at different in-plane field orientations, which is independent of the magnetization of the hardmagnetic FePt buffer. X-ray diffraction measurements revealed a crystalline [110] texture of the Fe/Cr multilayer to cause this intrinsic anisotropy of the AFM which is one condition to realize the exchange bias effect. The remanent magnetization of the hardmagnetic FePt layer was induced parallel to this easy axis by saturation in a field of 1.5 T.

Figure 1(b) shows the major hysteresis loop of the artificial exchange bias system measured by MOKE. The central region reflects the switching of the coupled FM and the diagonal branches of the curve characterize the saturation behavior of the multilayer AFM. To induce a well-defined single-domain ground state in remanence an external magnetic field of 100 mT was temporarily applied perpendicular to the easy axis. In this way, the Fe layers within the AFM align in a spin flop phase and orient collinear with the easy axis in remanence. The magnetization of the hardmagnetic FePt layer determines the orientation of the first Fe layer in the multilayer stack and so the alignment of the whole system in the magnetic ground state (see figure 1(a)). The MOKE minor hysteresis loops of the coupled AFM/FM system show archetypal exchange bias characteristics (figure 1(c)). The hysteresis curve along the hard axis is symmetric and centered. However, the reversal parallel to the easy axis of the AFM shows clear asymmetry, a strong increase of the coercivity field and the loop is shifted by a field of $-1.3$ mT. This is about a factor of 20 less than the value expected from the Meiklejohn–Bean model for a rigid and uncompensated interface and coincides with the experimentally observed reduced bias fields in conventional systems. Along the easy axis the magnetic reversal is additionally characterized by a weak, progressive training effect. That means that the shift and asymmetry of the MOKE curves are slowly reduced with consecutive magnetization loops but no abrupt transition can be identified from the initial to the second MOKE curve. To avoid the influence of the training effect on this study, we always performed the magnetic field procedure explained above (which replaces the field cooling procedure) before a minor hysteresis loop was measured. This way all the minor hysteresis curves shown in this paper are initial ones.

In order to clarify the origin of the observed exchange bias effect, we spatially resolve the magnetic reversal via time-resolved NRS of synchrotron radiation from $^{57}$Fe sensor layers. Two chemically identical exchange bias systems were prepared with a 1.6 nm thick probe layer (enriched to 95% in $^{57}$Fe) either in the central position of the FM or in the second Fe layer of the AFM (see figure 1(a)).

The experiment was carried out at the nuclear resonance beam line ID18 [14] of the European Synchrotron Radiation Facility in the 16-bunch mode of operation. The energy of the linearly polarized x-rays was tuned to the magnetic dipole transition of the $^{57}$Fe nuclei at 14.41 keV with a natural lifetime of 141 ns. The samples were illuminated in grazing incidence geometry close to the critical angle to maximize the reflected nuclear signal. From the recorded beat pattern in time (time spectra) the orientation of the net magnetization $\mathbf{m}_{\text{net}}$ relative to the incoming photon wave vector $\mathbf{k}_0$ can be determined [15]–[17], because the measured signal contains a contribution that is proportional to $\gamma |\mathbf{k}_0 \cdot \hat{\mathbf{m}}|$, where $\mathbf{k}_0$ and $\hat{\mathbf{m}}$ are unit vectors along $\mathbf{k}_0$ and $\mathbf{m}_{\text{net}}$. $0 < \gamma < 1$ describes the relative degree of magnetization,

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5 We like to point out that the spin structure of the artificial AFM alone is stable against external fields up to 20 mT along the easy axis (measured with MOKE and nuclear resonant x-ray reflectometry).

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Figure 2. Every magnetization state of the buried sensor layers was characterized by three NRS time spectra at different azimuthal orientations of sample + magnet relative to the photon beam (see right column). This procedure allows one to determine the spin direction and the fraction $\gamma$ of uncompensated moments with high precision. The example shows time spectra of the sensor layer inside the AFM at an external magnetic field $B_{\text{ext}}$ of $+5$ mT. The in-plane magnetization axis was determined to be $+53(3)^\circ$ from the magnetic field direction with two antiparallel magnetic sublattices $m_1$ and $m_2$ (the green and white arrows present their orientation relative to $\vec{k}_0$) in a ratio of $9(0.2) : 1$, corresponding to $\gamma = 0.8$.

i.e. the fraction of uncompensated moments. Thus, a single time spectrum does not allow for a unique determination of the magnetization direction and the value $\gamma$. Only the simultaneous evaluation of more than two time spectra yields the relative angle $\varphi$ between the magnetization and the photon wave vector as well as the fraction $\gamma$ of uncompensated moments as illustrated in figure 2. The variation of $\gamma$ with the external field then allows one to distinguish reversal processes of coherent rotation and domain wall motion.

This is shown in figure 3 which displays the results for the magnetic reversal of the exchange bias system along the easy axis. The integral nuclear count rate was around 500 Hz for the sensor layer embedded in the FM and 200 Hz for the sample with the sensor layer inside the multilayer AFM. All time spectra with good statistics were acquired in less than 3 min each and could be simulated with a fitting procedure where only $\varphi$ and $\gamma$ were varied. The magnetic hyperfine field was fixed ($B_{\text{HF–FM}} = 33.1$ T; $B_{\text{HF–AFM}}$: two components, 90% at 33.05 T, 10% at 29.65 T). Figure 3(a) shows the layer-resolved longitudinal magnetization components inside the FM and the AFM obtained from the evaluation of the time spectra in the central scattering...
Figure 3. (a) Hysteresis loops and (b) polar graphs presenting the magnetic reversal of the sensor layers along the easy axis obtained from a series of NRS time spectra. The blue numbers display the magnetic field values for selected points of the hysteresis. (c) Magnetization model of the exchange bias system that explains the measured reversal of the sensor layers. Here, every circle displays the magnetization of a Fe layer.

position ($B_{\text{ext}} \parallel k_0$). The hysteresis curve of the sensor layer inside the FM (red) corresponds to that of the surface sensitive MOKE signal (figure 1(c)) with a horizontal shift of $-1$ mT. The hysteresis curve of the AFM near the interface reveals a switching behavior strongly correlated with the FM but the loop is additionally shifted in the vertical direction similar to the observation.
reported in [18]. A detailed picture of the reversal mechanism in both sensor layers is obtained by plotting the vectorial state of their magnetization (represented by $\varphi$ and $\gamma$) in a polar diagram, as shown in figure 3(b).

The shape of the polar graphs allows one to distinguish between different reversal modes of the system: for a coherent rotation of the magnetization, the graph would be a half-circle connecting $\varphi = 0^\circ$ and $180^\circ$. For a reversal via pure domain wall motion, the system would move along a straight line between these points. Here we deduce a reversal mode of the FM (red points) that is dominated by domain wall motion: at $+15$ mT the magnetization of the FM is oriented along the external field direction. With decreasing field the FM slightly rotates out of the easy axis ($\varphi = 10^\circ$) and the formation of antiparallel-oriented magnetic domains leads to a fraction of uncompensated moments of $\gamma = 0.90$ in remanence. The bending of the FM hysteresis curve introducing the first reversal is due to a further rotation of the magnetization axis by $5^\circ$ and an increase of antiparallel-oriented domains which yield $\gamma = 0.53$ at $-5$ mT. Then the magnetization axis switches to $165^\circ$ with $\gamma = 0.37$. At the end of the first reversal, the FM magnetization axis completely aligns with the external field direction and the amount of antiparallel-orientated domains is reduced to $8\%$. In the third quarter of the hysteresis loop, the magnetization axis remains at around $170^\circ$ up to $-2$ mT. But although stabilized by the external field a remarkable early demagnetization of the FM sensor layer due to an increase of antiparallel domains starts at $-10$ mT and results in $\gamma = 0.60$ in remanence and $\gamma = 0.41$ at $+3$ mT ($\varphi = 155^\circ$) before the backward switching takes place.

The polar graphs of the AFM sensor layer magnetization clarify the origin of the asymmetric reversal of the FM. Due to the interlayer coupling to the FM the individual moments of the AFM near the interface also take part in the first reversal. In the AFM sensor, however, the magnetization axis only reaches $155^\circ$ at the end of the first branch ($B_{\text{ext}} = -15$ mT) and antiparallel domains still lead to a relatively low value of $\gamma = 0.66$. This magnetization state persists in the third quarter of the hysteresis up to $-4$ mT and acts as a nucleation center for the increase of antiparallel domains in the FM. At the same time the magnetization axes get aligned and the magnetization states of both sensor layers become equal and the backward switching occurs ($+3$ mT). This observation shows that the incomplete reversal of the AFM near the interface at the end of the first magnetization branch causes a partial compensation of the FM magnetization in the third quarter of the hysteresis loop. The backward reversal takes place at a reduced external magnetic field which results in a horizontal shift of the hysteresis loop. Taking into account that the first Fe layer of the AFM in contact with the hardmagnetic FePt layer is fixed [15], a reversal mechanism of the whole layer system can be constructed which is consistent with the measured results (figure 3(c)). According to this model, after the first reversal of the FM two parts in antiphase within the AFM coexist but the antiferromagnetic interlayer coupling hinders parallel-oriented Fe layers at the boundary at this two parts. The coupling of both parts by a planar domain wall is energetically unfavorable due to the magnetocrystalline anisotropy energy, as we could see from the reversal of the sensor layers. Only a lateral domain formation of the Fe layers can reduce the increased coupling energy inside the AFM, which occurs upon reversal of the FM moment direction.

These results identify the domain split interfacial spin structure of the AFM at the end of the first reversal to be the origin of the main archetypal exchange bias characteristics. Firstly, the remaining existence of antiparallel-orientated domains inside the FM layer even at a strong external field of $-15$ mT can be explained. This effect is due to the strong interfacial coupling of the FM spins to the domain split interfacial spins structure of the AFM which was identified to
have a net magnetization of only 66% at this field value. A similar effect was already indicated by Radu et al [8] via off-specular neutron scattering measurements. Secondly, we reveal the AFM domain structure to be the nucleation center for the second magnetization reversal of the FM and in this way the origin of the shifted and biased FM hysteresis curve. This was already proposed before [8, 9] but could not directly be measured because of the lack of information about the AFM during the reversal of the FM. A comparable strongly correlated magnetic reversal of the FM and AFM interface magnetization with an additionally vertical shift of the AFM hysteresis loop was recently measured by Ohldag et al [18] via element-specific x-ray hysteresis curves. Here, a reversal mechanism via two types of uncompensated moments inside the AFM that are differently strong coupled to the FM was proposed to explain the observation. However, although this technique allows one to correlate the coupled magnetization behavior on both sides of the FM/AFM interface the physical results are, in general, based on the interpretation of a longitudinal magnetization signal which is additionally integrated over several atomic planes. From the layer-resolved study presented here, we can finally identify the reason for the weak, progressive training effect of this exchange bias sample. The polar graph of the magnetization cycle inside the AFM shows only a small change of the magnetization state near the interface after the first cycle. While the FM returns with its magnetization axis and net magnetization into its initial magnetization state at $B_{\text{ext}} = +15 \text{ mT}$ (the magnetization loop in figure 3(a) is closed), the AFM magnetization axis does not reach again the collinear orientation ($\Delta \phi \approx 10^\circ$). That is why the shift of the next hysteresis loop of the FM will be slightly reduced because the coupling energy parallel to the external field axis is decreased. Also the asymmetry will be weakened because the amount of antiparallel-orientated domains inside the AFM in the first quarter of the hysteresis loop increased. This phenomenon will decrease the net magnetization of the FM in remanence before the third reversal takes place.

For a magnetic field orientation perpendicular to the easy axis, there is no effective parallel component of the unidirectional anisotropy. The MOKE magnetization loop is symmetric and centered. The investigation of the sensor layer magnetizations shows that the FM as well as the AFM near the interface reverses by coherent rotation whereas the rotation angles inside the AFM are reduced. The results are summarized in figure 4. At $+15 \text{ mT} (-15 \text{ mT})$ the FM rotates $+69^\circ (-67^\circ)$ out of the easy axis and the AFM sensor layer reaches $+45^\circ (-43^\circ)$. The results indicate that the coherent reversal of the FM induces a planar domain wall into the AFM (figure 4(b)), which partially stores the Zeeman energy of the FM and reduces its rotation angle with decreasing field. This magnetization behavior is in accordance with the Mauri model.

In summary, we have revealed the microscopic origin of the asymmetric shape and horizontal shift of the FM hysteresis loop as well as an additional vertical shift of the layer-resolved AFM hysteresis loop in an exchange bias model system along the easy axis. We could identify the correlation of the magnetic reversal in selected depths on both sides of the FM/AFM interface. The reversal proceeds mainly via domain wall motion with an incomplete reversal of the AFM near the interface. The FM adopts the domain split interface magnetization of the AFM in the third quarter of the hysteresis loop due to the interlayer exchange coupling, which results in a partial compensation of its net magnetization and causes archetypal exchange bias characteristics in the FM hysteresis loop. Along the hard axis the creation of a Mauri-type antiferromagnetic spring structure was inferred, which was already identified in conventional and artificial exchange bias systems [11, 19]. The results identify the origin of the exchange bias effect in a clarity never seen before, possibly due to differentially probing the layer system with ultrathin isotopic sensor layers. We would like to point out that the experimental procedure...
Figure 4. (a) Rotation angles of the sensor layers inside the FM and AFM during the reversal along the hard axis. The reversal takes place by coherent rotation with reduced rotation angles inside the AFM which is in accordance with the Mauri model.

presented here can also be applied to study conventional exchange bias systems regarding the magnetic reversal inside the AFM or the effect of field cooling on its interface spin structure even though such experiments are then directed by the quality of the sample preparation.

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