Asymmetric magnetization reversal in the exchange bias system Fe/FeF₂ studied by MOKE

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The asymmetry of the magnetization reversal process in exchange biased Fe/FeF₂ has been studied by magneto-optical Kerr effect. Qualitatively different transverse magnetization loops are observed for different directions of the cooling and the measuring field. These loops can be simulated by a simple calculation of the total energy density which includes the relevant magnetic anisotropies and coherent magnetization rotation only. Asymmetric magnetization reversal is shown to originate from the unidirectional anisotropy and may be observed if the external measuring field is not collinear with either the exchange bias or the easy axis of the antiferromagnetic epitaxial FeF₂(110) layer.

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A bilayer system composed of an antiferromagnetic (AFM) and a ferromagnetic (FM) layer exhibits a shift of the hysteresis loop along the field axis, the so-called exchange bias (EB). This shift may be observed after cooling the system below the Néel temperature of the AFM either in an external magnetic field or with the FM layer magnetized to saturation. Exchange biased systems including Fe/FeF₂ can additionally exhibit a pronounced asymmetry of the hysteresis loops.

This reversal asymmetry was first investigated in Fe/FeF₂ by polarized neutron reflectometry (PNR) indicating different reversal mechanisms on either side of the hysteresis loop, which have been interpreted as coherent magnetization rotation near the left-side coercive field and as domain wall nucleation and propagation near the right-side coercive field. While the former mechanism was identified by a transverse magnetization component related to strong spin-flip scattering of the polarized neutrons, the latter was assigned by the absence of a transverse magnetization component. This behavior raises the question as to the origin of this unprecedented asymmetry in any switchable hysteretic physical system.

Asymmetric magnetization reversal has also been observed in other exchange bias systems. But the asymmetry in the loop might also be reversed. In, e.g., Co/CoO coherent rotation has been found on the right side of the magnetization loop only. A fast and powerful experimental probe of coherent magnetization rotation is the magneto-optic Kerr effect (MOKE). Our initial MOKE measurements on Fe/FeF₂ gave also evidence for coherent rotation on only the right side of the hysteresis loop in contrast to the PNR results. This calls for systematic studies of the reversal asymmetry as for different directions of the in-plane cooling field relative to the easy axis of the EB system. The angular dependence is also aimed at investigating the role of higher order anisotropies in asymmetric magnetization switching, especially of odd symmetry as considered in Ref.

Here we present a systematic MOKE study of magnetization reversal in exchange biased Fe/FeF₂. Particular emphasis is given to measure the net transverse magnetization component $M_T$, which is oriented perpendicular to the external magnetic field. It may build up during magnetization reversal mainly near the coercive fields and is a direct probe of coherent magnetization rotation. Furthermore, this technique allows to determine the rotational direction (chirality) of the magnetization vector via the sign of $M_T$. Asymmetric magnetization reversal can be identified by a non-zero $M_T$ at only one coercive field during reversal. A quite unexpected behavior of $M_T$ is found for different directions of the initial cooling field and the subsequent measuring fields in a small angular range around the easy axis of the antiferromagnet FeF₂. Within about $±3°$ the asymmetry in the transverse loops reverses, i.e. $M_T$ switches from the left to the right side of the hysteresis loop and changes its sign. To simulate these loops, we present a simple model describing the coherent rotation of a single magnetic moment using a total energy density comprising fourfold, twofold, and unidirectional anisotropies. The model reproduces all salient features of the magnetization reversal. We demonstrate that asymmetric magnetization reversal originates from the existence of the unidirectional anisotropy and can only be observed if the measuring field is non-collinear with either the easy axis direction of the antiferromagnetic layer or the exchange bias direction set by the cooling field. We argue that domain wall nucleation and propagation is not relevant for the existence of asymmetric magnetization reversal.

Polycrystalline Fe has been grown on epitaxial, twinned FeF₂(110) by molecular beam epitaxy in the multilayer structure MgO(100)/FeF₂(100 nm)/Fe(13 nm)/Al(10 nm), with Al as protective cap layer. Details of the sample preparation and structural characterization are given elsewhere.

MOKE measurements have been carried out inside a magneto-optical cryostat using a motorized sample rotator which enables sample rotation with a precision of...
± 0.1° in an external magnetic field aligned parallel to the film plane. For Kerr effect we chose a reflection plane parallel to the transverse magnetization $M_T$, i.e. perpendicular to the external magnetic field. To unambiguously detect a pure $M_T$ hysteresis loop, we chose s-polarized light for the incident beam. Details of the experimental setup are described in Ref. [4]. We first investigate how the magnetization reversal depends on the cooling field direction which we vary relative to the easy axis of the antiferromagnetic FeF$_2$ layer by rotating the sample. Note that for a sample angle $\varphi_H = 0°$ the cooling field is the easy axis of the AFM at 45° with respect to the AFM twins along the $<001>$ directions. Fig. 1(a) shows a series of transverse hysteresis loops taken at $T = 20$ K for different sample orientations (positive angles correspond to a clockwise rotation of the sample relative to the reflection plane). The sample is field cooled at each angle in a magnetic field of $H = 1$ kOe through its Néel temperature ($T_N = 78.2$ K) and is subsequently measured at the same angle. At $\varphi_H = 0°$ the transverse loop consists of two peaks of opposite sign close to the left and right coercive fields, respectively, $H_{c,l}$ and $H_{c,r}$, indicating a full 360° coherent rotation of the magnetization vector. However, the reversal becomes asymmetric after field cooling only slightly away from the easy axis direction. At $\varphi_H = -3.5°$, a transverse magnetization can only be detected near $H_{c,l}$, while at +3.5° it appears only near $H_{c,r}$ with opposite sign. This might explain the apparent difference between various experimental findings in previous reports [2, 3]. A slight misorientation of the cooling field direction with respect to the AFM easy axis may lead to qualitatively different reversal asymmetries in the transverse magnetization.

![Figure 1](image1.png)

**FIG. 1:** Transverse magnetization of a polycrystalline Fe film exchange coupled to a twinned antiferromagnetic FeF$_2$(110) layer taken by MOKE at 20 K after field cooling in $H = 1$ kOe (a) at several sample angles $\varphi_H$ at which also the measurements are performed and (b) at $-3.5°$ followed by rotations of the sample to the indicated measurement angles.

For exploring whether the observed asymmetries depend on the field cooling procedure, we have field cooled the sample at -3.5° in $H = 1$ kOe and recorded transverse $M_T$ loops at various sample angles (Fig. 1(b)). Along the easy axis direction ($\varphi_H = 0°$) we again find symmetric reversal with slightly reduced amplitude near $H_{c,l}$. The $M_T$ peak also switches sides and sign in going to a sample orientation of 2°. Although the angles of this switching do not exactly match with the previous measurements (Fig. 1(a)), all salient loop shapes are again observed. This suggests that the existence of the rather complex magnetization asymmetry is linked to the local anisotropies which are only weakly affected by the field cooling procedure. To further investigate how the asymmetric transverse magnetization loops evolve we show a series of loops in a broader range of sample angles $\varphi_H$ from -10° to +10° in Fig. 2(a). The data has been taken at $T = 20$ K after field cooling at 0° in $H = 1$ kOe. At -10° we clearly observe a transverse magnetization near both $H_{c,l}$ and $H_{c,r}$ indicating symmetric reversal. Note that the sign of $M_T$ is positive for both reversal directions in contrast to the loop at 0° with opposite signs of $M_T$. A similar reversal as at -10° is also seen at +10° with the opposite sign of $M_T$. Most interestingly, the sign of $M_T$ reverses at different angles for both reversal directions, i.e. at -3° near $H_{c,l}$ and +2° near $H_{c,r}$ with a smooth change of its respective amplitudes. Note that these angles of sign reversal mark the sample orientations of asymmetric magnetization reversal.

![Figure 2](image2.png)

**FIG. 2:** (a) Hysteresis loops of the transverse magnetization of Fe/FeF$_2$ (110) at $T = 20$ K after field cooling along the easy axis of the AFM layer in $H = 1$ kOe. (b) Simulated $M_T$ vs. $H$ hysteresis loops for different sample orientations using a simple model solely based on coherent rotation (solid line: increasing field, dashed line: decreasing field).
tion signal on only one side of the transverse loop is a rather exceptional case. This suggests that coherent rotation of the magnetization may be the dominant reversal process at all stages of reversal.

To support this scenario we use a simple model describing coherent rotation of a single magnetic moment to (i) explain the observed asymmetric reversal and to (ii) simulate the salient features of all transverse magnetization loops in Fig. 2(a). The reversal of the moment is induced by an external magnetic field varied in its angular orientation. For each field step the magnetic moment follows the minimum of a free energy density $E$ comprising fourfold ($K_e$), twofold ($K_u$) and unidirectional ($K_c$) anisotropies, with $E$ given by

$$E = -H \cos(\varphi_H)M \cos(\varphi_M) - H \sin(\varphi_H)M \sin(\varphi_M) + K_e (\cos^2 \varphi_m - \sin^2 \varphi_m) + K_u (\cos^2 \varphi_m - (\frac{\zeta}{180})\pi) + K_c (\varphi_m - (\frac{\alpha}{180})\pi),$$

where $H$ describes the external magnetic field and $M$ the saturation magnetization of the FM layer, $\varphi_H$ and $\varphi_M$ are the angles of $H$ and $M$, respectively, which are both measured relative to the easy axis of the AFM layer. The angle $\zeta$ represents the respective angle of the twofold easy axis of the FM layer and $\alpha$ the respective angle of the easy axis of the unidirectional anisotropy $K_u$ (exchange bias). The anisotropy constants are set to $K_e = -100000$ erg/cm$^3$, $K_u = -5000$ erg/cm$^3$ and $K_c = 26000$ erg/cm$^3$. These values provide the best results for the simulation of the experimental loops in Fig. 2(a). The uniaxial anisotropy constant $K_u$ of Fe is about an order of magnitude smaller than single crystal values in literature [3, 11]. This might be related to the large polycrystalline fraction of the Fe film. For $M$ we use the bulk value of 1670 Oe for iron [11]. The angle $\alpha$ is fixed to 75$^\circ$ which is approximately the easy axis direction of the ferromagnet, while $\alpha$ is set to be identical to the cooling field direction of 0$^\circ$ which is parallel to the easy axis direction of the AFM layer.

We first investigate how to obtain asymmetric reversal in our simulations. Fig. 3 depicts a series of simulated $M_T(H)$ loops as obtained for $\varphi_H = -3^\circ$ and $\alpha = 0^\circ$ for various values of $K_u$. A Stoner Wohlfarth-type magnetization reversal is observed for $K_c = 0$ (top). As expected, we observe an exchange bias shift with increasing $K_c$. While $M_T$ remains positive near $H_{c,L}$ and stabilizes at large values of $K_c$, it switches sign at around $K_c = -1.0 \times 10^5$ erg/cm$^3$ near $H_{c,R}$. This clearly demonstrates that the existence of $K_c$ is solely responsible for the asymmetry in $M_T$. We want to emphasize that we do not obtain asymmetric reversal (not shown) if the measuring field is collinear with both the easy axis of the AFM and the exchange bias direction, i.e. $\varphi_H = \alpha = 0$.

We now use the above anisotropy parameters with $K_c = -1.0 \times 10^5$ erg/cm$^3$ to simulate the measured $M_T(H)$ loops in Fig. 2(a). These simulations are displayed in Fig. 2(a). Note that the only adjustable parameter is the direction of the external magnetic field, which we change in the experiment by sample rotation. The sign change of $M_T(H)$ on either side of the loop proceeds at the same angles as for the measured loops thus reproducing the observed reversal asymmetry. Differences in the shape of the simulated and measured loops are attributed to the use of a simple macrospin model in the simulations, which neglects any changes of the local anisotropies that might result from the polycrystalline structure of the Fe layer. This is evident from a more realistic Monte Carlo simulation by Beckmann et al. [11], describing the magnetization reversal of an averaged ensemble of moments based on the domain state model. Nevertheless, our simple macrospin model of coherent rotation can explain all salient features of the magnetization reversal in the experiments. Hence we conclude that anisotropies of higher than fourth order and particularly of odd symmetry do not play a role in describing the reversal asymmetry in our EB system.

To further illustrate the origin of asymmetric reversal, we depict the total energy density at various magnetic fields from 0 Oe to -80 Oe in Fig. 4(a) using the parameter set of the asymmetric loop for $\varphi_H = -3^\circ$ (Fig. 2(b)). Upon field variation, the magnetization vector follows the local energy minimum. We initialize its longitudinal orientation close to 0$^\circ$ (A) at $H = 0$ Oe. In Figs. 4(b) and (c), the magnetization vector is decomposed into its $M_L$ and $M_T$ components, respectively. A polar plot of both is included in Fig. 4(d). Upon reversal from A to E the macrospin first reaches a local energy minimum near 90$^\circ$ (C) at -65 Oe which results in a stable transverse

![FIG. 3: Simulation of $M_T(H)$ as a function of the unidirectional anisotropy constant $K_c$. The exchange bias direction is assumed to be collinear with the easy axis of the AFM layer ($\alpha = 0^\circ$), while the external magnetic field is misaligned by $\varphi_H = -3^\circ$.](image)
magnetization between points C and D (Figs. 4c and 4d), which is also observed in the experiments (see Fig. 2a)). The magnetization is fully reversed in point E. In the opposite reversal direction the macrospin switches between F and G with no stable intermediate state along the transverse direction. Correspondingly, we do not observe a stable peak on the right side of the transverse loop, although the macrospin has to pass through the transverse direction. According to the asymmetry of the local energy density with respect to the direction of \(-M_L\) at 180°, the macrospin rotates backwards by changing its chirality. This observation confirms the asymmetric reversal mode simulated by Beckmann et al. for an angle of \(\varphi_H = 60°\) between the external field and the easy axis of an untwinned EB system.

According to our simulations, the interpretation of a vanishing component \(M_T\) in experiments as a sufficient indication of magnetization reversal by domain wall nucleation and propagation has to be revised. Our results clearly demonstrate that conventional domain wall nucleation and propagation is not needed to understand the observed asymmetric magnetization reversal. However, we want to emphasize that our simple macrospin model does not explain all details of reversal such as the exact values of the respective coercive fields as well as the continuous change of the net transverse magnetization through the critical field angles at which the sign reversal of \(M_T\) is observed in the experiments. Our model is capable of reproducing the magnetization reversal for different cooling field directions with the same set of anisotropy constants.

In summary, we have shown that the asymmetric magnetization reversal as probed by the transversal magnetization in Fe/FeF\(_2\)(110) depends strongly on the non-zero angle between the measurement field and the easy axis of the AFM or the EB direction. A simulation based on the field dependent total energy density considering all relevant anisotropies and the coherent rotation of a macrospin describes qualitatively the experiments. The unidirectional EB anisotropy is solely responsible for the asymmetry in the transverse magnetization loops. The agreement between simulation and experiment endorses the assumption that coherent rotation is sufficient to describe asymmetric magnetization reversal in Fe/FeF\(_2\).

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[1] W. H. Meiklejohn and C. P. Bean, Phys. Rev. 105, 904 (1957). For a review see J. Nogués and I. K. Schuller, J. Magn. Magn. Mater. 192, 203 (1999).
[2] For a first report see: M. R. Fitzsimmons et al., Phys. Rev. Lett. 84, 3986 (2000).
[3] F. Radu et al., Phys. Rev. B 67, 134409 (2003).
[4] A. Tillmanns et al., cond-mat/0508635.
[5] J. McCord et al., J. Appl. Phys. 93, 5491 (2003).
[6] T. Mewes et al., Phys. Rev. B 65, 224423 (2002).
[7] I. N. Krivorotov et al., Phys. Rev. B 65, 100402(R) (2002).
[8] J. Nogués et al., Phys. Rev. B 59, 6984 (1999).
[9] U. Gradmann, in: Ferromagnetic Materials Vol. 7, Ed. K. H. J. Buschow, North-Holland Publishing Company, Amsterdam-London-New York-Tokyo 1993 (p. 1).
[10] E. P. Wohlfahrt in: Ferromagnetic Materials Vol. 1, Ed. E. P. Wohlfahrt, North-Holland Publishing Company, Amsterdam-Oxford-New York-Tokyo 1980 (p. 1).
[11] B. Beckmann et al., Phys. Rev. Lett. 91, 187201 (2003).
[12] A. Tillmanns et al., unpublished
[13] J. Camarero et al., Phys. Rev. Lett. 95, 57204 (2005).