

Irreversibility of magnetization rotation in exchange biased Fe/epitaxial-FeF₂ thin films

Justin Olamit and Kai Liu^{a)}*Physics Department, University of California, Davis, California 95616*Zhi-Pan Li^{b)} and Ivan K. Schuller*Physics Department, University of California-San Diego, La Jolla, California 92093*

(Received 8 November 2006; accepted 11 December 2006; published online 19 January 2007)

Magnetization reversal via rotation is typical in ferromagnet/antiferromagnet exchange biased systems. The reversibility of the rotation is a manifestation of the microscopic reversal process. The authors have investigated the magnetization reversal in Fe/epitaxial-FeF₂ thin films using vector magnetometry and first-order reversal curves. The reversal is predominantly by rotation as the applied field makes an angle with the antiferromagnet spin axis, mostly irreversible at small angles and reversible at larger angles. A modified Stoner-Wohlfarth model reproduces the overall trend of the irreversibility evolution. The remaining discrepancies between the modeled and measured irreversibilities may be attributed to local incomplete domain walls. © 2007 American Institute of Physics. [DOI: 10.1063/1.2431784]

Exchange bias (EB) in ferromagnet/antiferromagnet (FM/AF) bilayers has attracted intense interests due to its elusive mechanism and important applications in “spin-valve” type devices.¹⁻³ The magnetization reversal mechanisms in EB systems have been the focus of many studies.⁴⁻¹⁴ In certain systems asymmetric reversal processes have been observed, where domain nucleation and growth dominate in one field sweep of the hysteresis loop while magnetization rotation prevails in the opposite field sweep.⁶⁻⁹ Other studies have shown the dependence of the reversal mechanism on the exchange and anisotropy fields in the sample.^{10,15} We have recently shown in FeF₂ and MnF₂ based systems that the switching mechanism depends on the AF crystallinity, especially on the alignment of the applied field with respect to the AF anisotropy axes.^{9,16,17} For example, FM/epitaxial-FeF₂ thin films have a single AF spin axis along which the exchange field would also point. With a small misalignment ($\sim 2^\circ$), the switching is clearly dominated by rotation.¹⁷ Indeed, magnetization rotation has been clearly identified in many experimental studies and found to be a common occurrence. However, despite being prominently featured in theoretical models,^{4,10} the nature of the rotations themselves has been rarely examined closely,¹³ in particular, the irreversibility and the implications on the microscopic magnetization switching process.

In this study, we have used a first-order reversal curve (FORC) method in conjunction with vector magnetometry to probe the reversal mechanism and quantitatively captured the irreversibility of the magnetization switching. The angular dependence of the reversal has been studied where the alignment of the applied field with respect to the AF anisotropy direction is changed. The results are compared with a modified Stoner-Wohlfarth model and the roles of local incomplete domain walls are highlighted.

Bilayer films of Fe/FeF₂ were fabricated by electron beam evaporation, similar to those reported earlier.^{15,18,19} An epitaxial layer of 500 Å FeF₂ was first evaporated onto a single crystal (110) MgF₂ substrate held at 300 °C. A layer of 300 Å Fe and a capping layer of 80 Å Al were subsequently grown at 150 °C. In-plane x-ray diffraction shows that the FeF₂ thus made is untwinned epitaxial (110). The Fe layer is polycrystalline. The bulk AF FeF₂ (110) surface is magnetically compensated with a single in-plane spin axis along [001] and an ordering temperature of $T_{\text{Néel}}=78$ K.

The sample was mounted in a Princeton Measurements Corp. vibrating sample magnetometer (VSM) so that the AF spin axis was approximately parallel to the applied field axis. The VSM was equipped with a helium flow cryostat and vector detection coils which were sensitive to moments parallel and perpendicular to the applied field. The sample was field cooled from 300 to 15 K in a 1 kOe field, a field sufficient to saturate the sample but still small enough not to induce a two-step reversal previously found in other exchange biased samples incorporating epitaxial FeF₂.^{18,19} With the cooling field applied in the sample plane along the AF spin axis direction, the sample became biased along this direction.¹⁷ Care was taken to align the AF spin axis with the applied field by monitoring the transverse loop. Alignment was achieved when the transverse loop features vanish (designated as the $\alpha=0^\circ$ orientation). Several orientations have been studied, where the misalignments α between the AF spin axis and applied field were 2° , 8° , 45° , and 90° . At each orientation, longitudinal and transverse hysteresis loops as well as FORC's were measured. Note that the sample orientation was such that the applied field stayed in the sample plane as the sample was rotated about its normal.

A FORC is measured by first saturating the sample at a large positive field, then decreasing the field to a reversal field H_R , and measuring the magnetization M as the applied field H is increased back to saturation.^{20,21} A subsequent FORC is measured after resaturating the sample and then ramping the field to a lower reversal field H_R . As the reversal fields are changed, the interior of the major loop is “filled”

^{a)} Author to whom correspondence should be addressed; electronic mail: kailiu@ucdavis.edu

^{b)} Present address: Cornell University, School of Engineering and Applied Physics, Ithaca, NY 14853.

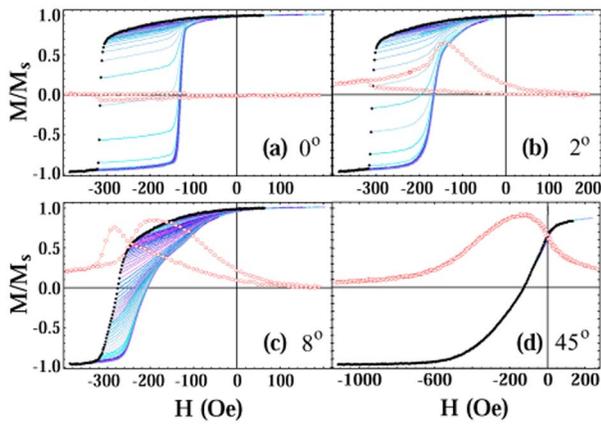


FIG. 1. (Color online) Family of FORC's (lines) and transverse hysteresis loops (open circles) measured at 15 K in the α =(a) 0° , (b) 2° , (c) 8° , and (d) 45° orientations after field cooling in 1 kOe. The black dots in the FORC data mark the beginning of each reversal curve. The outer boundary of FORC's traces out the longitudinal hysteresis loop.

with the FORC's. The FORC distribution $\rho(H_R, H)$ is calculated as

$$\rho(H_R, H) = -\frac{1}{2} \frac{\partial^2 M(H_R, H)/M_S}{\partial H_R \partial H}, \quad (1)$$

where M_S is the saturation magnetization. The partial derivative removes the purely reversible components of the magnetization. The FORC distribution measures the weight function of hysterons, or elements of hysteresis, with a particular local coercivity $H_C = (H - H_R)/2$ and bias field $H_B = (H + H_R)/2$, and in certain systems is equivalent to the Preisach distribution.²⁰ We have shown that the FORC distribution is very sensitive to distributions of magnetic properties and, in particular, irreversible switching processes.²¹ The integration $I_{\text{Irrev}} = \int \rho(H_R, H) dH_R dH = \int \rho(H_B, H_C) dH_B dH_C$ captures the amount of irreversible magnetic switching in a sample (where $I_{\text{Irrev}} = 100\%$ indicates that magnetic switching occurs entirely by irreversible processes).²²

The FORC data and transverse hysteresis loops for the different orientations are shown in Fig. 1. The longitudinal loops are contained in the families of FORC's as the outer boundaries. At $\alpha = 0^\circ$ [Fig. 1(a)], the longitudinal loop has an exchange field of $H_E = -220$ Oe and a coercivity of $H_C = 96$ Oe. The transverse loop is essentially flat and feature-

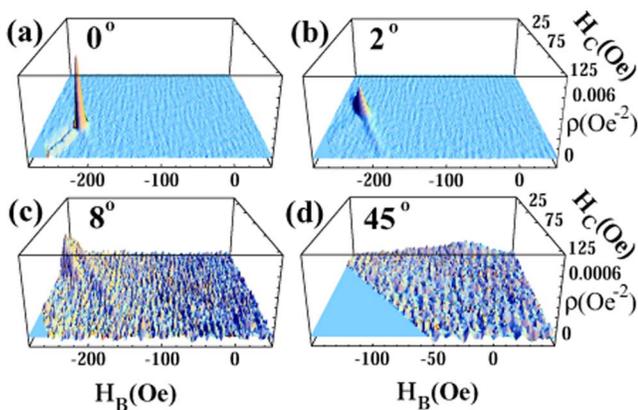


FIG. 2. (Color online) FORC distribution ρ plotted as a function of local coercivity H_C and bias field H_B in the α =(a) 0° , (b) 2° , (c) 8° , and (d) 45° geometries.

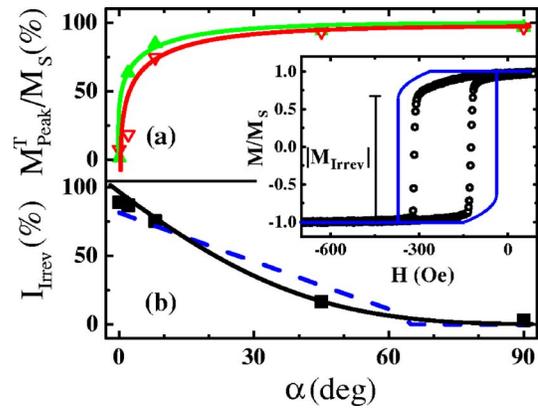


FIG. 3. (Color online) (a) Normalized transverse hysteresis loop peak magnitude M_{Peak}^T/M_S along decreasing and increasing field sweeps (open and filled triangles, respectively) and (b) the amount of irreversible switching obtained from FORC (squares) and the modified SW model (dashed line) in each geometry. Solid lines are guides to the eyes. The inset shows the measured (open circles) and simulated (lines) longitudinal hysteresis loop for $\alpha = 0^\circ$. The irreversible portion of the magnetization switching calculated in the modified Stoner-Wohlfarth model is shown by the vertical bar.

less. As α increases to 2° and 8° [Figs. 1(b) and 1(c)], the transverse loops develop distinct peaks, clearly showing that the reversal mechanism is dominated by rotation when the exchange field is not collinear with the AF spin axis. The corresponding longitudinal loops become highly asymmetric and the exchange field increases slightly. At very large misalignments, i.e., when $\alpha = 45^\circ$ [Fig. 1(d)] and 90° , the coercivity of the longitudinal loop collapses and the exchange field decreases and vanishes at $\alpha = 90^\circ$. Meanwhile, peaks in the transverse loops almost reach saturation magnetization.

The corresponding FORC distributions $\rho(H_B, H_C)$ are shown in Fig. 2. At $\alpha = 0^\circ$ [Fig. 2(a)], the FORC distribution has a distinct peak ($\rho_{\text{peak}} = 8 \times 10^{-3} \text{ Oe}^{-2}$). The peak is located at bias field $H_B = -220$ Oe and coercivity $H_C = 96$ Oe, which correspond to the exchange field and coercivity of the major loop, respectively. At $\alpha = 2^\circ$ [Fig. 2(b)], the peak feature broadens; however, its height is significantly reduced ($\rho_{\text{peak}} = 2 \times 10^{-3} \text{ Oe}^{-2}$). As α increases further, the peak feature is drastically suppressed at $\alpha = 8^\circ$ [$\rho_{\text{peak}} < 2 \times 10^{-4} \text{ Oe}^{-2}$ in Fig. 2(c)] and essentially vanishes at $\alpha = 45^\circ$ and beyond [Fig. 2(d)].

The percentage of the irreversible magnetization reversal, $I_{\text{Irrev}} = \int \rho(H_B, H_C) dH_B dH_C$, at each geometry is shown in Fig. 3, along with the corresponding peak heights of the transverse hysteresis loop M_{Peak}^T . At $\alpha = 0^\circ$, the transverse loop is virtually featureless and the irreversibility is at a maximum of 89%. With increasing α , the amount of irreversible switching decreases while the transverse peaks grow. At $\alpha = 2^\circ$ and 8° , the transverse loop has developed significant peaks (up to 64% and 85% of M_S , respectively), indicating that a majority of the film reverses via rotation. However, the irreversibility remains high (87% and 75%, respectively). These results demonstrate that most of the reversal is via *irreversible rotation processes*. At large α (45° and 90°), the transverse peaks are over 95% of M_S . The low I_{Irrev} ($< 16\%$) indicates that most of the rotation processes are reversible for large angles. Note that the angular dependence of irreversibility is monotonic, unlike that of the exchange field.

To further explore the effects of different geometries, we have modeled the reversal process using a modified Stoner-

Wohlfarth (SW) model. This type of model has been used to account for the angular dependence of exchange biased systems because the reversals of such systems occur largely through rotation.^{11,23} The energy density of the FM layer is modeled as

$$E = -H_E M_S \cos(\theta - \alpha) + K_1 \cos^2(\theta - \alpha) + K_2 \cos^4(\theta - \alpha) - H M_S \cos(\theta), \quad (2)$$

where K_1 and K_2 are the first and second order uniaxial isotropy constants,²⁴ respectively, and θ is the angle between the applied field and magnetization. Hysteresis loops are modeled by minimizing the energy for different H . K_1 and K_2 are determined by fitting the $\alpha=90^\circ$ loop and H_E is determined from the $\alpha=0^\circ$ loop. While the simulated and measured loops agree well for $\alpha=90^\circ$, they deviate for $\alpha=0^\circ$ (Fig. 3, inset). The discrepancy may be attributed to additional switching mechanisms involving domain nucleation and wall motion. For each α , the amount of irreversible switching was determined as $I_{\text{Irrev}} = |M_{\text{Irrev}}|/2M_S$, where $|M_{\text{Irrev}}|$ is the amount of magnetization that abruptly reverses at the steep drop in the loop where the energy becomes unstable.

The amount of irreversibility based on the SW model is plotted in Fig. 3 as a dashed line. The overall decreasing trend of SW irreversibility with increasing α is consistent with the FORC measurement, indicating that much of the irreversibility can be attributed to rotation. However, there are still discrepancies in the two I_{Irrev} curves. This is because the SW model is not always applicable. For example, with zero or a small misalignment ($\alpha=0^\circ$ or 2°), the transverse hysteresis loop shows little feature, indicating multidomain processes where SW model no longer applies.

A more realistic picture of the magnetization reversal is one that involves both rotations and domain nucleation, i.e., via local incomplete domain walls (LIDW's). These domain walls form in the depth of the FM layer, where the moments at the free FM surface rotate before those at the FM-AF interface, creating a partial spiral structure similar to that suggested by Kiwi *et al.*²⁵ We have observed the existence of such FM domain walls recently in similar samples.^{26,27} Furthermore, micromagnetic simulations have shown that these domains also have a lateral structure, where adjacent domains nucleate and rotate at different fields due to variations of local FM/AF coupling strengths.²⁶ Rotations of these local domains give rise to the large transverse component captured by vector magnetometry while the nucleation of the LIDW's—not included in the single domain SW model—may account for the discrepancy between the modeled and measured I_{Irrev} and coercivities.

The reversal mechanism at $\alpha=0^\circ$ is still unclear—the results do not rule out reversals by either rotation or domain wall nucleation and motion. However, extrapolating the irreversibility trend in Fig. 3 seems to suggest reversals via LIDW's. At $\alpha=0^\circ$, the bulk AF spin axis is aligned with the field but individual AF domains may be slightly misaligned, with equal numbers of domains having a net “up” or “down” transverse magnetization. These AF domains couple with the FM to create LIDW's in the FM that wind in opposite directions, resulting in the flat transverse loop. This has been observed in micromagnetic simulations.²⁸ As α increases, the population of up AF domains grows at the expense of down ones, causing more individual FM domains to twist “upward.” Indeed, the rapid growth of the transverse loop peaks

with misalignment may be an indication that the dispersion of local AF anisotropy directions is small. The dominance of rotation processes at zero or small misalignments is also consistent with previous polarized neutron reflectivity measurements.⁹

In summary, we have studied the magnetization reversal mechanisms of Fe/epitaxial-FeF₂ with vector magnetometry and FORC. The magnetization reversal is predominantly by rotations when the applied field makes an angle with the AF spin axis. The rotations are highly irreversible when the angle is small and becomes reversible at larger angles. A modified Stoner-Wohlfarth model reproduces the overall trend of the irreversibility evolution. The remaining discrepancies between the modeled and measured irreversibility may be due to the formation of local incomplete domain walls.

This work has been supported by the ACS-PRF (No. 43637-AC10), the U.S. Department of Energy, and the Alfred P. Sloan Foundation.

- ¹W. H. Meiklejohn and C. P. Bean, *Phys. Rev.* **102**, 1413 (1956).
- ²J. Nogués and I. K. Schuller, *J. Magn. Magn. Mater.* **192**, 203 (1999).
- ³A. E. Berkowitz and K. Takano, *J. Magn. Magn. Mater.* **200**, 552 (1999).
- ⁴M. Stiles and R. McMichael, *Phys. Rev. B* **59**, 3722 (1999).
- ⁵V. I. Nikitenko, V. S. Gornakov, A. J. Shapiro, R. D. Shull, K. Liu, S. M. Zhou, and C. L. Chien, *Phys. Rev. Lett.* **84**, 765 (2000).
- ⁶M. R. Fitzsimmons, P. C. Yashar, C. Leighton, I. K. Schuller, J. Nogués, C. F. Majkrzak, and J. A. Dura, *Phys. Rev. Lett.* **84**, 3986 (2000).
- ⁷K. Liu, S. M. Baker, M. Tuominen, T. P. Russell, and I. K. Schuller, *Phys. Rev. B* **63**, 060403 (2001).
- ⁸M. Gierlings, M. Prandolini, H. Fritzsche, M. Gruyters, and D. Riegel, *Phys. Rev. B* **65**, 092407 (2002).
- ⁹M. R. Fitzsimmons, C. Leighton, J. Nogués, A. Hoffmann, K. Liu, C. F. Majkrzak, J. A. Dura, J. R. Groves, R. W. Springer, P. N. Arendt, V. Leiner, H. Lauter, and I. K. Schuller, *Phys. Rev. B* **65**, 134436 (2002).
- ¹⁰B. Beckmann, U. Nowak, and K. D. Usadel, *Phys. Rev. Lett.* **91**, 187201 (2003).
- ¹¹J. Camarero, J. Sort, A. Hoffmann, J. M. Garcia-Martin, B. Dieny, R. Miranda, and J. Nogués, *Phys. Rev. Lett.* **95**, 057204 (2005).
- ¹²P. Blomqvist, K. M. Krishnan, and H. Ohldag, *Phys. Rev. Lett.* **94**, 107203 (2005).
- ¹³T. Gredig, I. N. Krivorotov, and E. D. Dahlberg, *Phys. Rev. B* **74**, 094431 (2006).
- ¹⁴A. Tillmanns, S. Oertker, B. Beschoten, G. Guntherodt, C. Leighton, I. K. Schuller, and J. Nogués, *Appl. Phys. Lett.* **89**, 202512 (2006).
- ¹⁵H. Shi and D. Lederman, *Phys. Rev. B* **66**, 094426 (2002).
- ¹⁶E. Arenholz and K. Liu, *Appl. Phys. Lett.* **87**, 132501 (2005).
- ¹⁷J. Olamit, Z. P. Li, I. K. Schuller, and K. Liu, *Phys. Rev. B* **73**, 024413 (2006).
- ¹⁸I. V. Roshchin, O. Petravic, R. Morales, Z. P. Li, X. Battle, and I. K. Schuller, *Europhys. Lett.* **71**, 297 (2005).
- ¹⁹J. Olamit, E. Arenholz, Z. P. Li, O. Petravic, I. V. Roshchin, R. Morales, X. Battle, I. K. Schuller, and K. Liu, *Phys. Rev. B* **72**, 012408 (2005).
- ²⁰C. R. Pike, A. P. Roberts, and K. L. Verosub, *J. Appl. Phys.* **85**, 6660 (1999).
- ²¹J. E. Davies, O. Hellwig, E. E. Fullerton, G. Denbeaux, J. B. Kortright, and K. Liu, *Phys. Rev. B* **70**, 224434 (2004).
- ²²J. E. Davies, J. Wu, C. Leighton, and K. Liu, *Phys. Rev. B* **72**, 134419 (2005).
- ²³M. Grimsditch, A. Hoffmann, P. Vavassori, H. T. Shi, and D. Lederman, *Phys. Rev. Lett.* **90**, 257201 (2003).
- ²⁴ K_2 reduces the squareness of simulated loops and is necessary to fit loops at both $\alpha=0^\circ$ and 90° .
- ²⁵M. Kiwi, J. Mejia-Lopez, R. Portugal, and R. Ramirez, *Appl. Phys. Lett.* **75**, 3995 (1999).
- ²⁶Z. P. Li, O. Petravic, R. Morales, J. Olamit, X. Battle, K. Liu, and I. K. Schuller, *Phys. Rev. Lett.* **96**, 217205 (2006).
- ²⁷R. Morales, Z.-P. Li, O. Petravic, X. Battle, I. K. Schuller, J. Olamit, and K. Liu, *Appl. Phys. Lett.* **89**, 072504 (2006).
- ²⁸Z. P. Li, Ph.D. thesis, University of California-San Diego, 2006.