Positive Exchange Bias in FeF_2-Fe Bilayers

J. Nogues,[1,*] D. Lederman,[2] T. J. Moran,[1,†] and Ivan K. Schuller[1]
[1]*Physics Department 0319, University of California at San Diego, La Jolla, California 92093-0319*
[2]*Physics Department, West Virginia University, Morgantown, West Virginia 26506-6315*
(Received 23 February 1996)

We have discovered a *positive* unidirectional exchange anisotropy in antiferromagnetic (FeF_2) and ferromagnetic (Fe) bilayers cooled through the antiferromagnetic critical temperature T_N in large magnetic fields. For low positive cooling fields, the ferromagnet's magnetization (M-H) loop center shifts to negative fields, as is normally observed in other systems. In contrast, large cooling fields can cause the shift to be positive. This can be explained if the FeF_2 surface spins couple to the external magnetic cooling field above T_N and the FeF_2-Fe interaction is antiferromagnetic. [S0031-9007(96)00418-8]

PACS numbers: 75.70.Cn, 75.30.Gw

Exchange anisotropy (EA) is caused by the magnetic interface interaction between a ferromagnet (FM) and an antiferromagnet (AFM). When a sample with a FM-AFM interface is cooled in a static magnetic field from above the ordering temperature of the AFM (T_N), with the FM Curie temperature greater than T_N, the FM magnetization (M-H) loop shifts away from $H = 0$. The magnitude of this shift is known as the exchange bias (H_E). Despite years of research since the discovery of EA in 1957 by Meiklejohn and Bean [1], the details of the mechanism responsible for this phenomenon remain unclear. Nevertheless, important technological applications of this effect include domain stabilizers [2] in magnetoresistive heads and "spin-valve"-based devices [3].

In a perfect, bulk AFM, two energetically equivalent spin configurations exist because the two spin sublattices are identical. However, in the traditional explanation of EA, the exchange interaction between an AFM and a FM breaks this symmetry, causing one of the AFM sublattices to couple to the FM as the sample is field cooled from $T > T_N$. Because of the AFM-FM interaction, the FM spins will tend to point in the direction of the cooling field at low temperatures even when the measuring field is reduced below $H = -H_C$, where H_C is the FM coercive field. Eventually, the field overcomes the interface interaction and the magnetization reverses at a field $H = -H_C - |H_E|$. When the field is increased, the FM magnetization reverses at a field corresponding to $H = H_C - |H_E|$. Therefore, *a positive cooling field results in a shift of the FM hysteresis loop toward negative fields* ($H_E < 0$), which is the usual experimental observation. This result is independent of whether the FM-AFM interaction is ferromagnetic or antiferromagnetic.

In the present work, we examine the dependence of EA on the cooling field H_{fc} in FeF_2/Fe AFM-FM bilayers. We find that H_E increases, i.e., becomes less negative, as H_{fc} is increased. For some samples, the M-H loops shift to *positive* fields when cooled in a large positive field, i.e., $H_E > 0$. The sign of H_E remains unchanged as a function of temperature until it disappears above T_N. The data can be explained if (1) the surface AFM spins couple to the magnetic field above T_N, and (2) the AFM-FM interface exchange is antiferromagnetic. Therefore, this experiment probes the magnetic interface interactions, and demonstrates that in this system it must be antiferromagnetic. This provides a way of determining the sign of the FM-AFM interface exchange interaction, which is difficult to determine in any other fashion.

The Fe^{2+} ions in FeF_2 form a body-centered tetragonal crystal structure ($a = b = 4.69$ Å, $c = 3.301$ Å) [4], with the ions at the unit cell center ordering antiferromagnetically with the ions at the corners [5]. FeF_2 has a large uniaxial magnetic anisotropy along the c axis [6]. The growth of FeF_2-Fe bilayers on MgO [100] has been described elsewhere [7]. Briefly, the films were grown by sequential e-beam evaporations of FeF_2 (~90 nm at a rate of 0.2 nm/s) and Fe (~13 nm at a rate of 0.1 nm/s). Substrates were heated to 450°C for 900 s prior to deposition, then cooled to the FeF_2 growth temperature $200 \leq T_S \leq 300$°C. At these temperatures the FeF_2 grows quasiepitaxially along the [110] direction with two in-plane domains (see below). The Fe layers were deposited at 150°C, resulting in polycrystalline films with mostly [110] and [100] orientations, and then capped with ~9 nm of Ag to prevent oxidation. The pressure during deposition was $< 1 \times 10^{-6}$ Torr. The film thicknesses were controlled by a calibrated quartz crystal oscillator.

Grazing-angle x-ray scattering data using Cu $K\alpha$ radiation ($\lambda = 1.5418$ Å is shown in Fig. 1. The inset shows the scattering geometry. As shown in Fig. 1(a), a scan of the angle ω, with $2\Theta = 55.54°$ fixed to the (002) FeF_2 in-plane reflection Bragg condition, revealed a fourfold symmetry. The in-plane $[1\bar{1}0]$ reflection was also fourfold symmetric. Because the FeF_2 [110] surface unit cell is rectangular, the film is twinned in the plane, presumably because the MgO [100] surface unit cell is a square. Figure 1(b) corresponds to the MgO substrate (200)

 0031-9007/96/76(24)/4624(4)$10.00

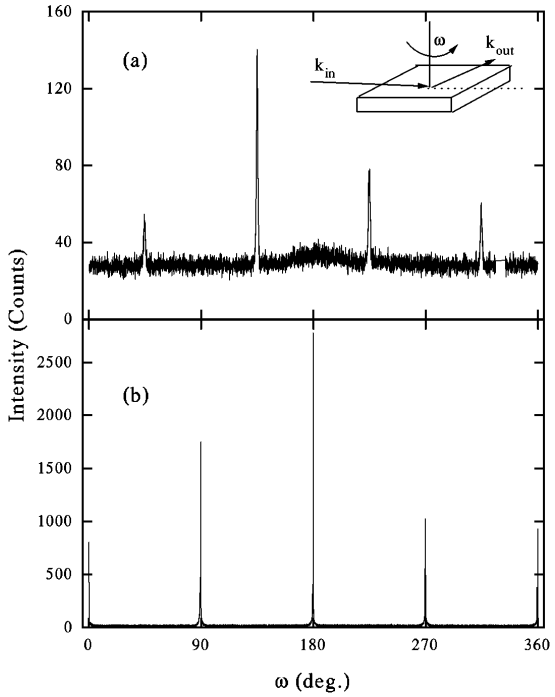

FIG. 1. (a) In-plane x-ray scattering of the FeF_2 (002) in-plane peak for an FeF_2-Fe bilayer film. (b) In-plane x-ray scattering of the MgO substrate in-plane (200) peak. The scans were performed without removing the sample from the goniometer. ω has been corrected for the difference in detector angles for the two reflections. Inset shows scattering geometry.

in-plane reflections ($2\Theta = 42.91°$). With respect to the MgO substrate, the two in-plane domains are determined by $FeF_2[001] \parallel MgO[110]$ and $FeF_2[1\bar{1}0] \parallel MgO[\bar{1}10]$ and its corresponding twin. Hence, two FeF_2 magnetic in-plane domains exist with their easy axes ([001] c axis) in the plane, but perpendicular to each other.

Samples were cooled from 120 K through the FeF_2 critical temperature ($T_N = 78.4$ K [8]), to 10 K in the presence of a magnetic field $H_{\rm fc}$. The Fe M-H loops were measured using a SQUID magnetometer in the -2 to $+2$ kOe range. In all cases, 2 kOe was enough to saturate the Fe magnetization.

Figure 2 shows the dependence of H_E at $T = 10$ K on $H_{\rm fc}$ for samples with the FeF_2 grown at different temperatures. $H_{\rm fc}$ was applied parallel to the MgO [100] in-plane direction. For samples grown at 300 and 250°C, H_E changed sign as $H_{\rm fc}$ was increased. When cooled in 70 kOe, the magnitude of H_E was in some instances as large or larger than the magnitude of the negative values obtained for low cooling fields. When samples were field cooled in low fields (2 kOe), and the magnetic field increased to 70 kOe at low temperatures (10 K), H_E remained unchanged to within 5% of the $H_{\rm fc} = 2$ kOe value. This demonstrates that the effect is a consequence of the AFM interaction with the magnetic field *during* the most recent cooldown procedure. It is important to note

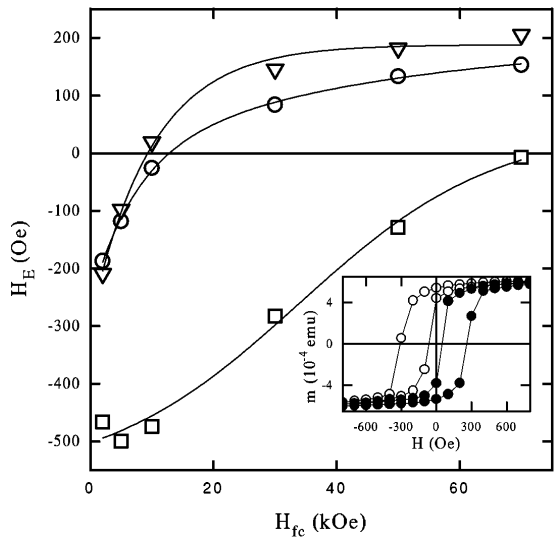

FIG. 2. Exchange bias field H_E as a function of the cooling field $H_{\rm fc}$ at $T = 10$ K for samples with the FeF_2 grown at $T_S = 200\,°C$ (□), $T_S = 250\,°C$ (▽), and $T_S = 300\,°C$ (○). Lines are guides to the eye. Inset: Magnetization loops of the $T_S = 300\,°C$ sample for $H_{\rm fc} = 2$ kOe (∘) and $H_{\rm fc} = 70$ kOe (•) at $T = 10$ K.

that all M-H loops remained unchanged when scanned repetitively [7].

Figure 3 shows the temperature dependence of the *absolute value* of H_E for the $T_S = 250\,°C$ sample field cooled in 2 kOe ($H_E < 0$) and 70 kOe ($H_E > 0$). The sign of H_E remains unchanged throughout the whole temperature range. In both cases, H_E disappears close to the T_N of FeF_2 (78.4 K), which indicates that the

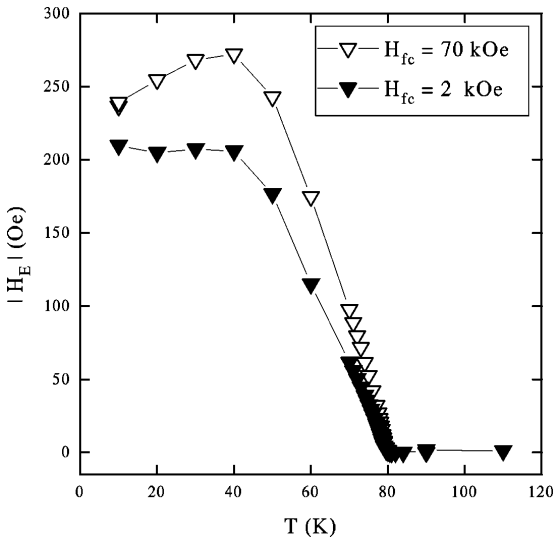

FIG. 3. Temperature dependence of the exchange bias field *magnitude* $|H_E|$ for the $T_S = 250\,°C$ sample shown in Fig. 2, field cooled in high and low fields. $H_E > 0$ for $H_{\rm fc} = 70$ kOe and $H_E < 0$ for $H_{\rm fc} = 2$ kOe in the temperature range $10\ {\rm K} \le T \le T_N = 78.4$ K. Lines are guides to the eye.

antiferromagnetic order is responsible for the presence of H_E. The coercivities for the two values of $H_{\rm fc}$ were similar, as shown in the inset of Fig. 2. This indicates that the FM domain structure is *not* responsible for the positive H_E.

A possible mechanism of the positive exchange bias is a competition between the FM-AFM exchange interaction and an external field–AFM surface magnetic coupling interaction. If $H_{\rm fc}$ couples to the AFM surface as the AFM is cooled through T_N, and the FM-AFM interaction is ferromagnetic, then the usual negative H_E is obtained because the system is in a low interface magnetic energy configuration (that is, there is no competition). However, H_E is positive if the FM-AFM interface magnetic interaction is *antiferromagnetic* and $H_{\rm fc}$ is large enough to align the AFM surface magnetization along $H_{\rm fc}$, thus overcoming the interface AFM-FM antiferromagnetic interaction. This is because, after field cooling, the system is in a state of high interface magnetic energy, assuming that the AFM magnetic surface remains fixed when the magnetic field is reversed. A similar argument was previously used to explain inverted hysteresis loops in CoO-Co granular samples, which were attributed to antiferromagnetic coupling between FM Co grains [9]. $H_{\rm fc}$ breaks the two-sublattice AFM symmetry during cooling, assuming the AFM surface is even slightly magnetically uncompensated. At low temperatures, the surface magnetic configuration remains locked by the bulk AFM magnetic structure. Note that if the interface interaction is antiferromagnetic, but there is no coupling between $H_{\rm fc}$ and the AFM surface, the resulting H_E is always negative. This is because, as in the case of ferromagnetic interface coupling, the system is field cooled into a low interface magnetic energy configuration where there is no competition.

In order to determine the plausibility of this mechanism, the magnetization of a 1000 layer Ising AFM film was calculated self-consistently using the mean-field equations for each layer [10]. In this calculation each antiferromagnetic sublattice is composed of a single atomic layer. Hence, the top and bottom layers are magnetically uncompensated. The calculation was carried out as a function of temperature, starting at low temperatures. The cooling field was $H_{\rm fc} = 0.01 H_{\rm AF}$, where $H_{\rm AF}$ is the exchange field of the antiferromagnet. For FeF_2 this corresponds to $H_{\rm fc} \sim 3.2$ kOe, much lower than the highest value used in our experiments (70 kOe). Figure 4 shows the magnetization of the first atomic layer (M_S) when $H_{\rm fc}$ is positive and negative. The sublattice magnetization near the center of the film, which represents the bulk material, is also shown for reference. Note that the magnetization of the top layer is positive or negative depending on whether the film is cooled in positive or negative fields. Similar calculations with 1001 layers yielded identical results for the surface magnetization, the only difference being that in the 1000 layer film a domain wall was formed. Hence, mean-field theory shows that for thicker AFM films with uncompensated magnetic surfaces cooling in a magnetic field through the Néel temperature breaks the symmetry of the sublattices.

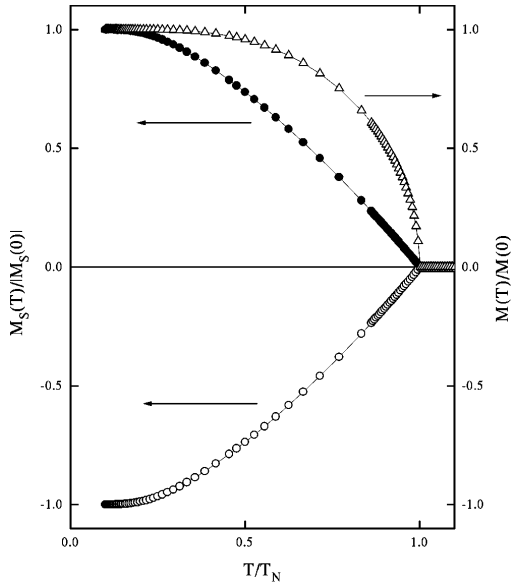

FIG. 4. Normalized magnetization as a function of temperature $[M_S(T)/\,|\,M_S(0)\,|]$ of the top layer of an uncompensated 1000 layer antiferromagnet calculated using mean-field theory. (∘) and (•) represent the calculation in negative and positive cooling fields, respectively ($H_{\rm fc} = 0.01 H_{\rm AF}$, where $H_{\rm AF}$ is the exchange field of the antiferromagnet). The sublattice magnetization of the bulk (△) is also shown for reference. Lines are guides to the eye.

In order to determine the value of $H_{\rm fc}$ necessary to induce the positive H_E, assume that the cooling field is applied along the AFM easy-axis direction. For *low cooling fields*, such that $|J_I| S_A S_F > H_{\rm fc} M_{SA}$ (where M_{SA} is the AFM surface magnetization and S_A and S_F are the values of the AFM and FM spins), but larger than the FM coercive field H_C, the AFM surface magnetization will lie antiparallel to $H_{\rm fc}$ at low temperatures. This will result in the usual *negative* H_E. For *high cooling fields* ($|J_I| S_A S_F < H_{\rm fc} M_{SA}$), the AFM surface orients along $H_{\rm fc}$, thus frustrating the AFM-FM exchange interaction. At low temperatures after the field is lowered, the AFM spin structure remains locked by the uniaxial anisotropy. Therefore, H_E should abruptly change sign when $H_{\rm fc} = |J_I| S_A S_F / M_{SA}$. However, in imperfect samples, with defects resulting in a distribution of J_I and/or M_{SA}, the change will be gradual, as shown in Fig. 2. Samples with larger J_I require larger $H_{\rm fc}$ to obtain $H_E > 0$. Notice that H_E in Fig. 2 never becomes positive for the sample grown at 200 °C. This sample has the smoothest interface, as determined from low-angle x-ray diffraction [7]. Hence, according to the model presented above, this sample has the largest average magnitude of J_I, therefore requiring larger cooling fields to increase H_E. A more quantitative analysis requires a value for M_{SA}, which is unknown.

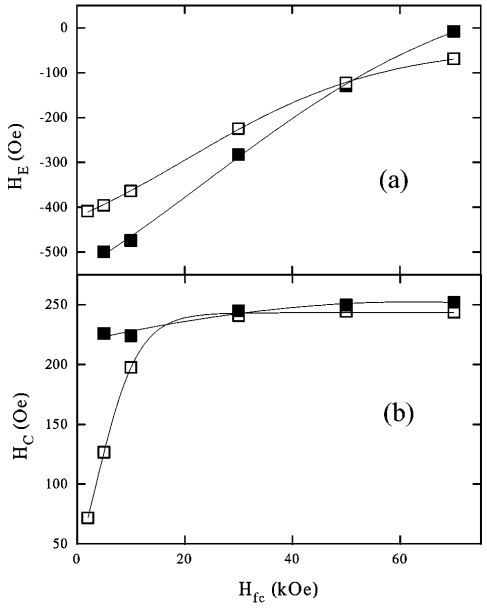

FIG. 5. Exchange bias field H_E (a) and coercive field H_C (b) as functions of the cooling field $H_{\rm fc}$, for the $T_S = 200\,^{\circ}$C sample in Fig. 2, with $H_{\rm fc}$ applied $\parallel$ or $\perp$ to (■) and at 45° (□) from the FeF_2 easy axes of the two FeF_2 in-plane domains. Lines are guides to the eye.

If the AFM uniaxial anisotropy direction is not parallel to $H_{\rm fc}$, the magnitude of the positive exchange bias will be reduced by a factor of $\cos\theta$, where θ is the angle between the anisotropy direction and $H_{\rm fc}$. In Fig. 5(a) H_E vs $H_{\rm fc}$ is plotted when $H_{\rm fc}$ is applied either parallel to or at 45° from the in-plane MgO [100] direction. When $H_{\rm fc} \parallel$ MgO[100], $\theta = \pi/4$ in both FeF_2 domains, while in the other configuration $\theta = 0$ or $\theta = \pi/2$, according to the x-ray data. Note that the FeF_2 thermodynamic spin-flop field is 419 kOe [11], well above the largest cooling fields, and thus it seems unlikely that the sublattice magnetization axis would change significantly when cooling at low or high fields. As shown in Fig. 5, the change of H_E with $H_{\rm fc}$ is larger when $\theta = \pi/4$. On average, the case of $\theta = 0$ or $\theta = \pi/2$ yields a factor of 0.5. When both domains are at $\theta = \pi/4$, the multiplicative factor is $1/\sqrt{2} > 0.5$. Therefore, qualitatively the H_E dependence on $H_{\rm fc}$ is expected to be stronger when $H_{\rm fc} \parallel$ MgO[100] ($\theta = \pi/4$), which is indeed the case in Fig. 5.

This simple model does not explain all of the data. As shown in Fig. 5(b), H_C, and hence the FM anisotropy, is strongly dependent on $H_{\rm fc}$ only for $\theta = 0$ or $\pi/2$. In these cases, H_C is small for small $H_{\rm fc}$, and saturates to a value similar to the $\theta = \pi/4$ case. This implies that the FM anisotropy changes with $H_{\rm fc}$, and could mean that at low temperatures the AFM surface spins do not align along the bulk easy axis. Another problem is that the growth of FeF_2 along the (110) direction implies a magnetically compensated surface. However, the magnetic surface could be effectively uncompensated if there is a surface reconstruction at the Fe-FeF_2 interface, and in this case, the cooling field could still break the sublattice symmetry. Nevertheless, the model does qualitatively explain how a positive H_E could arise. Direct quantitative measurements of the magnetic interface interactions are needed to describe this effect in more detail.

In conclusion, we have observed a positive exchange bias in FeF_2-Fe bilayers when they are field cooled in large fields through T_N. The effect is qualitatively attributed to a combination of an antiferromagnetic exchange at the AFM-FM interface and a FM coupling of the AFM surface spins to the cooling field above T_N. The observation of this effect provides a way of determining the sign of the AFM-FM interface magnetic exchange interaction.

This work was supported by the U.S. Department of Energy under Grant No. DE-FG03-87ER45332. J. N. thanks the Spanish Ministerio de Educación y Ciencia for its financial support.

*On leave from the Group d'Electromagnetisme, Universitat Autònoma de Barcelona, Spain.
†Present address: Department of Physics, University of Minnesota, Minneapolis, MN 55455.

[1] W. H. Meiklejohn and C. P. Bean, Phys. Rev. **102**, 1413 (1957).
[2] C. Tsang and R. Fontana, IEEE Trans. Magn. **18**, 1149 (1982).
[3] B. Dieny, V. S. Speriosu, S. S. P. Parkin, J. C. Scott, B. A. Gurney, D. R. Wilhoit, and D. Mauri, Phys. Rev. B **43**, 1297 (1991).
[4] J. W. Stout and S. A. Reed, J. Am. Chem. Soc. **76**, 5279 (1954).
[5] R. A. Erickson, Phys. Rev. **90**, 779 (1953).
[6] M. T. Hutchings, B. D. Rainford, and H. J. Guggenheim, J. Phys. C **3**, 307 (1970).
[7] J. Nogués, D. Lederman, T. J. Moran, I. K. Schuller, and K. V. Rao, Appl. Phys. Lett. **68**, 3186 (1996).
[8] D. P. Belanger, P. Nordblad, A. R. King, and V. Jaccarino, J. Magn. Magn. Mater. **31–34**, 1095 (1983).
[9] C. Gao and M. J. O'Shea, J. Magn. Magn. Mater. **127**, 181 (1993).
[10] D. L. Mills, Phys. Rev. B **3**, 3887 (1971).
[11] V. Jaccarino, A. R. King, M. Motokawa, T. Sakakibara, and M. Date, J. Magn. Magn. Mater. **31–34**, 1117 (1983).