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Influence of in-plane crystalline quality of an antiferromagnet on perpendicular exchange coupling and exchange bias

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We have undertaken a systematic study of the influence of in-plane crystalline quality of the antiferromagnet on exchange bias. Polarized neutron reflectometry and magnetometry were used to determine the anisotropies of polycrystalline ferromagnetic (F) Fe thin films exchange coupled to antiferromagnetic (AF) *untwinned single crystal* (110) FeF_2, *twinned single crystal* (110) FeF_2 thin films and (110) *textured polycrystalline* FeF_2 thin films. A correlation between the anisotropies of the AF and F thin films with exchange bias was identified. Specifically, when exchange coupling across the F-AF interface introduces an additional anisotropy axis in the F thin film—one perpendicular to the cooling field, the magnetization reversal mechanism is affected (as observed with neutron scattering) and exchange bias is significantly enhanced.

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I. INTRODUCTION

Investigations of exchange anisotropy (EA) at the interface between ferromagnetic (F) and antiferromagnetic (AF) materials have received renewed attention recently due to the importance of EA in technological applications.[1,2] Theoretical and experimental progress has been made understanding the phenomenology and mechanisms for exchange bias H_E (the shift of the F hysteresis loop along the field axis—a manifestation of unidirectional EA).[3] Experimentally, the effects of interface disorder[4] on H_E, the relation between H_E and coercivity, H_C,[5–7] the magnetization reversal mechanisms,[8–10] and the temperature dependence of H_E (Refs. 6, 11–13) have been studied in different systems. Theoretical studies have produced various models for H_E and H_C.[14,15] These models include: formation of AF domain walls parallel[16] and perpendicular[17] to the F-AF interface, spin-flop coupling across the F-AF interface,[18–21] collective excitations,[22,23] uncompensated free spin densities,[11] and AF domains with net magnetization.[24]

Coexistence of exchange bias and so-called perpendicular exchange coupling across the F-AF interface, which is manifested by a perpendicular orientation between the uniaxial anisotropy axis of the F relative to the uniaxial anisotropy axis of the AF, has been experimentally observed.[25,26] Coexistence of these phenomena may be coincidental, or may suggest an interdependence. Sophisticated numerical models of EA have predicted that exchange coupling across the F-AF interface (for compensated AF surfaces) will produce an arrangement where the spins of the F thin film are perpendicular to those of the AF.[18–21] So called spin-flop coupling is a low-energy configuration for a F layer on a compensated AF surface, and can give rise to experimentally observed perpendicular exchange coupling. Alternative mechanisms, including magnetoelasticity of the AF,[26] could also produce perpendicular exchange coupling between F and AF layers.

When the F and AF spins are constrained to lie parallel to the interface plane, Koon[18] found that spin-flop coupling led to exchange bias of the F-hysteresis loop. More recent studies removed this constraint and found that spin-flop coupling would enhance the coercivity of the F thin film via an increase in uniaxial anisotropy, but did not produce exchange bias. Rather, a canted magnetic structure of the AF at the F-AF interface in combination with an incomplete domain wall in the F,[19] or uncompensated moments in the AF (Ref. 20) were required to produce exchange bias.

Alternatively, Miltényi *et al.*[24] attribute exchange bias in Co-CoO bilayers to the exchange interaction between the net magnetization of finite-sized AF domains (bounded by domain walls perpendicular to the F-AF interface—as in the Malozemoff model[17]) with the F thin film. Their model predicts that a net magnetization of the AF layer establishes unidirectional anisotropy in the F layer parallel to the cooling field, so perpendicular exchange coupling (or spin-flop coupling) across the F-AF interface is not required to produce exchange bias.

Owing to the computational complexity of numerical models, modeling generally involves idealized F-AF structures, e.g., first-principle calculations are made for F-AF systems where the AF is an untwinned single crystal. An impediment towards proving/disproving some of the different models for exchange bias is attributable to a lack of experimental data for nearly idealized systems. Experimental studies have focused primarily on systems in which the AF is usually polycrystalline, sometimes textured, and least often, twinned (or multidomained) single crystals are studied.[25] Some exceptions exist,[26,27] for example, perpendicular exchange coupling was inferred from a magnetometry study of an F-untwinned bulk single crystal AF.[26]

Here, we report the results of a systematic experimental investigation that correlates the in-plane crystallinity of the AF layer with anisotropies created in the F thin film, magnetization reversal, and concomitant exchange bias upon field cooling (FC). Specifically, neutron scattering and magnetometry measurements were taken from samples with *untwinned single crystal* (110) FeF_2, and (110) *textured polycrystalline* FeF_2 AF thin films. (Measurements for the twinned thin-film single-crystal (110) FeF_2 system were previously reported in Ref. 8. For completeness, conclusions from that study are repeated as needed in the present manuscript.) The intent of this study is to understand how the in-plane crystalline structure of the AF influences EA across the F-AF interface by observing magnetization reversal processes in the F layer with neutron scattering, and by measuring exchange bias with magnetometry for samples in which the crystal structure of the AF layer is systematically changed. All AF films have (110) out-of-plane orientation (texture); however, the in-plane structure changes from single crystal to twinned to polycrystalline.

With a cooling field applied perpendicular to the spins of the untwinned single crystalline AF, a classic instance of perpendicular exchange coupling across the F-AF interface was observed, yet exchange bias was not observed. A cooling field applied to the sample with the textured polycrystalline AF, yielded exchange bias even though perpendicular exchange coupling was not observed. Therefore, perpendicular exchange coupling is neither a sufficient condition nor a required condition for exchange bias. Nevertheless, frustration of perpendicular exchange coupling between a F layer and a twinned AF is intimately linked to large exchange bias. In fact, for the Fe-FeF_2 thin film system, the tendency to form uncompensated moments in a polycrystalline AF thin film, would seem to be of less importance in determining the magnitude of exchange bias compared to frustration of perpendicular exchange coupling that can arise in a twinned AF system.

II. SAMPLE PREPARATION

This study involved investigations of three types of samples. In all cases the F thin film is polycrystalline Fe. The first sample, called the untwinned AF sample (u-AF), was prepared by sequential electron-beam deposition of ZnF_2 (25 nm), FeF_2 (65 nm), Fe (12 nm), and Al (10 nm) (a capping layer to prevent oxidation) onto a polished untwinned bulk single-crystal (110) FeF_2 substrate. The bulk FeF_2 substrate was heated to 773 ± 2 K to clean its surface prior to thin-film deposition. The nominal temperatures of the substrate during deposition of ZnF_2 and FeF_2 were 473 ± 2 K and 573 ± 2 K, respectively, and of Fe and Al were 423 ± 2 K. Using x-ray reflectometry, the roughness of the F-AF (Fe-FeF_2) interface (root-mean-square deviation about its mean) was determined to be 2.0 ± 0.5 nm. In-plane glancing incidence x-ray diffraction confirmed that the AF layer grew as an untwinned single-crystal film.

The purpose of the ZnF_2 buffer layer is to decouple AF order of the bulk (110) FeF_2 single-crystal substrate from that of the FeF_2 thin film, while maintaining heteroepitaxial growth conditions so that the FeF_2 thin film would grow as an untwinned (110) FeF_2 single crystal. The extra step taken to deposit a thin film of untwinned single-crystal (110) FeF_2, rather than simply depositing Fe onto the bulk (110) FeF_2 single crystal, facilitates a systematic and transparent comparison of in-plane microstructure ranging from untwinned AF films, to twinned AF films, to textured polycrystalline AF films.

The second sample, called the twinned AF sample (t-AF) was prepared by sequential electron-beam deposition onto a polished untwinned single-crystal (100) MgO substrate. Preparation of sample t-AF, and its characterization via x-ray diffraction, neutron reflectometry, and magnetometry were discussed previously in Ref. 8. Sample t-AF was composed of FeF_2 (90 nm), Fe (11 nm), and Ag (3 nm). The twinned structure of FeF_2 is a natural consequence of growing a rectangular lattice (i.e., the (110) plane of FeF_2) on a square lattice [i.e., the (100) plane of MgO] and produces twin crystal domains oriented 90° to one another. The roughness of the F-AF interface for sample t-AF was 1.2 ± 0.5 nm.

For the third sample, called the polycrystalline AF sample (p-AF), a $\sim$1-μm-thick MgO film was first grown onto glass using ion-beam-assisted deposition (IBAD).[28] IBAD involves bombarding the sample substrate with low-energy ions, as MgO is deposited via electron-beam deposition onto the substrate surface. The angle of incidence between the ion beam and the sample surface was chosen to preferentially sputter away MgO crystallites with crystallographic orientations that did not have the [100] direction parallel to the sample surface normal. This procedure produces a MgO film with a random orientation in the sample plane, and a (100) texture perpendicular to the film plane. After deposition of the MgO film, sequential electron-beam depositions of FeF_2 (90 nm thick), Fe (13 nm), and Al (20 nm) were made at temperatures of 473 ± 2 K, 423 ± 2 K, and 423 ± 2 K, respectively. X-ray diffraction confirmed the out-of-plane (110) texture of the FeF_2 AF thin film, while no evidence for in-plane texture was found using in-plane glancing incidence x-ray diffraction. X-ray reflectometry determined the roughness of the F-AF interface to be 4 ± 1 nm.

III. MAGNETOMETRY RESULTS—EXCHANGE BIAS AND EXCHANGE COUPLING

To confirm that the Fe thin films were exchange coupled to the AF thin films after cooling through the Néel point of the AF ($T_N=78$ K), the F-hysteresis loops of the samples were measured with a superconducting quantum interference device magnetometer. Exchange coupling between the F and AF thin films is evident if exchange bias is observed, or if the shapes of the F-hysteresis loops change upon cooling through T_N. The cooling field was $H_{FC}=2.00\pm0.01$ kOe ($=509$ kA/m). Two cooling field orientations were examined for sample u-AF—one with the cooling field applied parallel to the AF anisotropy axis, i.e., $\mathbf{H}_{FC}\|[001]$ FeF_2 [Fig. 1(a), inset], and one perpendicular to the AF anisotropy axis, i.e., $\mathbf{H}_{FC}\|[\bar{1}10]$ FeF_2 [Fig. 2(a), inset]. The values of H_E and H_C reported in Figs. 1(a) and 2(a) were obtained from hysteresis

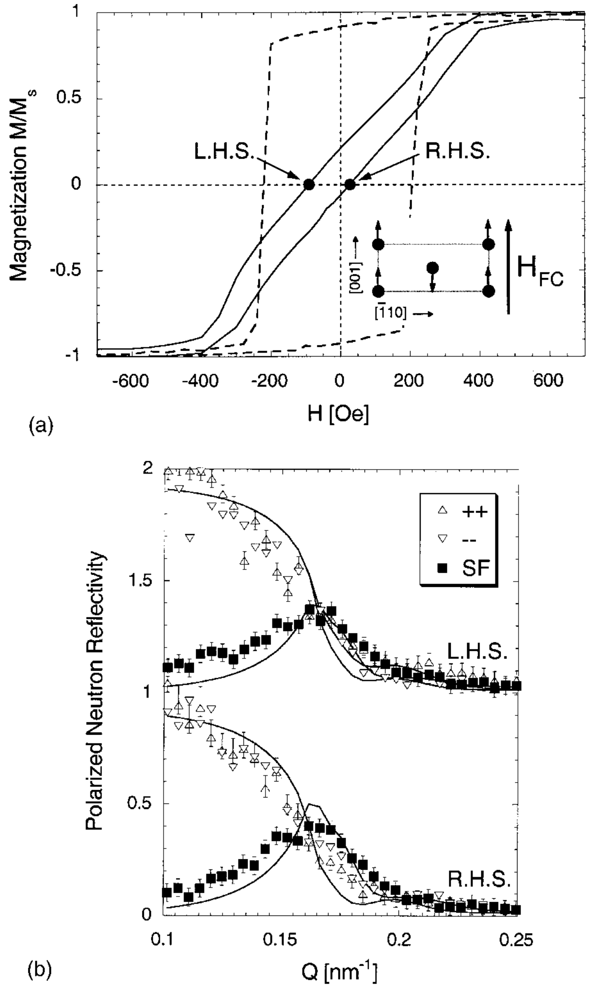


FIG. 1. (a) Hysteresis loop at 10 K (solid curve) and at 300 K (dashed curve) for sample *u*-AF, and the orientation of the cooling field $\mathbf{H}_{FC}\|[001]FeF_2$ (inset). This cooling field condition produces exchange bias ($H_E = -32 \pm 2$ Oe) of the F-hysteresis loop at 10 K, and coercivity of $H_C = 59 \pm 2$ Oe. (b) Polarized neutron reflectivity profiles taken at 20 K for applied fields shown by the closed symbols ● in (a) on the left-hand side (LHS) and right-hand side (RHS) for the same sample and cooling field orientation. Significant spin-flip (SF) scattering is observed for coercive fields on either side of the hysteresis loop, indicating magnetization reversal via rotation. Data corresponding to LHS are shifted for the sake of clarity. Solid curves were obtained from fitted models of the type discussed in Ref. 13.

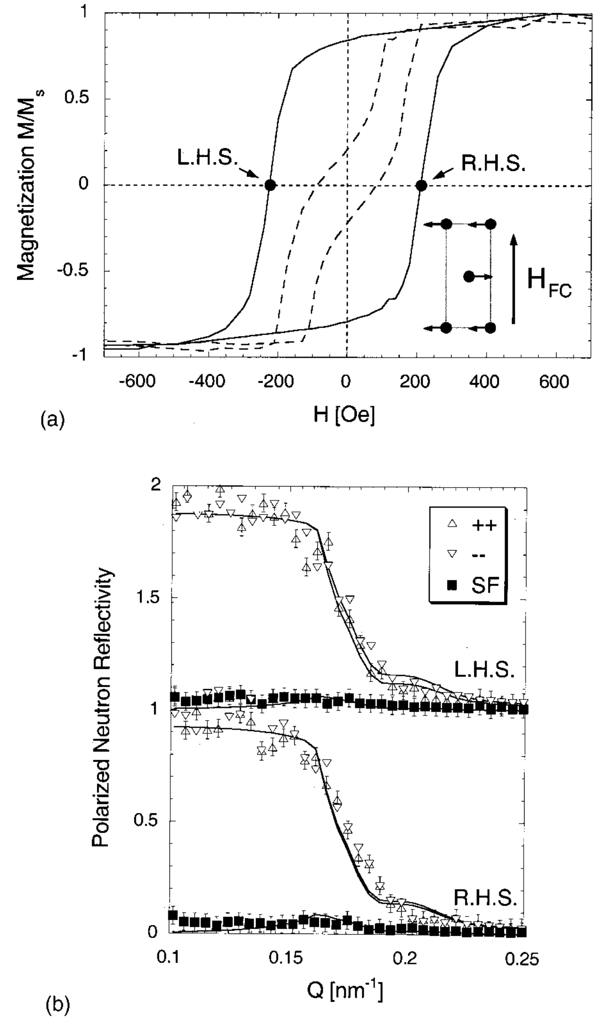


FIG. 2. (a) Hysteresis loop at 10 K (solid curve) and at 300 K (dashed curve) for sample *u*-AF, and the orientation of the cooling field $H_{FC} = 2$ kOe for $\mathbf{H}_{FC}\|[\bar{1}10]FeF_2$ (inset). H_E and H_C for this cooling field condition are -2 ± 2 Oe and 218 ± 2 Oe, respectively. (b) Polarized neutron reflectivity profiles taken at 20 K for applied fields shown by the closed symbols ●'s in (a) on the LHS and RHS for the same sample and cooling field orientation. No SF scattering is observed, indicating magnetization reversal via domain nucleation and wall motion. Data corresponding to LHS are shifted for the sake of clarity. Solid curves were obtained from fitted models of the type discussed in Ref. 13.

loops measured at 10 K (solid curves) with fields applied parallel (or antiparallel) to the cooling field direction. The hysteresis loops measured for the same crystallographic orientations at room temperature are shown by the dashed curves.

To see that the F and AF thin films were exchange coupled for sample *u*-AF, consider the first cooling field orientation (Fig. 1). At room temperature the hysteresis loop [dashed curve Fig. 1(a)] is square, indicating that the measurement field was applied parallel to an easy axis of the F thin film. Upon cooling through T_N, the loop [solid curve Fig. 1(a)] becomes sheared, indicating that the (measurement) field was applied parallel to a hard axis of the F layer. Yet, the direction of the measurement field always remained parallel to the cooling field direction and the anisotropy axis of the AF thin film. Therefore, the [001] FeF_2 direction, which corresponds to an easy axis in the F thin film at room temperature, is a hard axis in the F thin film at 10 K. This change (and the nonzero value of H_E), upon cooling is an evidence for the exchange coupling across the F-AF interface.

Now, consider the second cooling field orientation (Fig. 2)—one with the cooling field applied parallel to $[\bar{1}10]$ FeF_2. At room temperature, the hysteresis loop [dashed curve, Fig. 2(a)] is sheared, so the direction in the F thin film parallel to $[\bar{1}10]$ FeF_2 is a hard axis. Upon cooling in a field, the hysteresis loop becomes square, indicating that the axis in the F thin film is now an easy axis. This change from a hard to an easy axis upon cooling is evidence for perpendicular exchange coupling across the F-AF interface (since the easy axis of the F is perpendicular to the anisotropy axis of the AF); however, the exchange bias is nil.

The temperature dependence in the qualitative appearances of the hysteresis loops as square or sheared can be quantified by plotting the remanent magnetization (M_R in units of the saturation magnetization M_S) as a function of temperature for the two cooling field orientations [Fig. 3(a)]. For completeness, the temperature dependence of the coercivity H_C and exchange bias H_E are shown in Figs. 3(b) and 3(c), respectively. The temperature dependences of H_C and M_R/M_S indicate a gradual rotation of the F anisotropy axis, starting around T_N, but not finally completed until lower temperatures. The behavior is due to the competition between the intrinsic anisotropy of the F and the induced anisotropies due to the F-AF coupling. Similar behavior was previously observed for bulk FeF_2/Fe.[26]

The hysteresis loop shown in Fig. 4(a) (solid curve) was obtained from sample *p*-AF at 10 K. Hysteresis loops were also obtained by cooling and measuring in other orientations, and these did not differ significantly. The similarity of the hysteresis loops indicates that the ferromagnetic properties of the F layer were isotropic.

IV. NEUTRON-SCATTERING RESULTS—MAGNETIZATION REVERSAL MECHANISMS

The magnitude and orientation of **M** in the sample plane relative to the cooling field and details about the magnetization reversal process (i.e., whether magnetization reversal occurred via rotation or domain nucleation) were determined from the reflectivities of the samples measured with polarized neutrons. Polarized neutron reflectometry (PNR) involves specular reflection of a polarized neutron beam from a flat sample onto a polarization analyzer.[29] Four neutron cross sections were measured. Two cross sections correspond to the non-spin-flip (NSF) reflectivity profiles, where the intensities of the reflected radiation for spin-up (++) [and alternately spin-down (−−)] neutrons illuminating and reflecting from the sample were measured.[30] The difference between the ++ and −− NSF reflectivity profiles ΔNSF is related to the projection of **M** on the direction of the applied field $\mathbf{H_A}$, i.e., $\Delta\mathrm{NSF} \propto M_{\parallel}$. The remaining two cross sections are the spin-flip (SF) reflectivities. These are nonzero if the sample changes the neutron beam polarization from spin-up to spin-down (+−), and vice versa. For example, if **M** has a component perpendicular to the neutron spin (as for example would occur if the sample magnetization rotated away from the applied field), then the beam polarization will change, so $\mathrm{SF} \propto M_{\perp}$. Therefore, we can determine from the PNR data unambiguously the magnetization reversal mechanism, i.e., whether the reversal occurs via coherent rotation vs domain wall motion. Moreover, the PNR profiles were fitted using models of the type discussed in Ref. 13, from which the fraction of the sample with magnetization perpendicular to the applied field $M_{\perp}$ is obtained quantitatively.

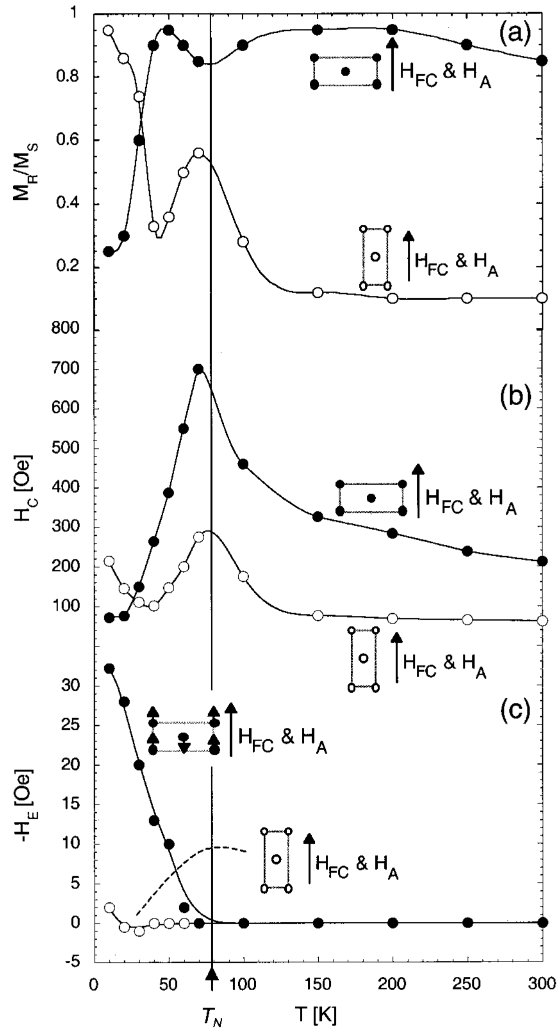


FIG. 3. (a) Remanent magnetization M_R, i.e., the magnetization of the sample for applied field $H_A=0$, normalized to the saturation magnetization M_S, is shown as a function of temperature. Solid symbols correspond to directions of H_{FC}, and the applied field H_A parallel to [001] FeF_2. Open symbols correspond to H_{FC}, and H_A parallel to $[\bar{1}10]$ FeF_2. The crossover of the remanent magnetization near 40 K suggests a change of the anisotropies in the Fe thin film. (b) The coercivity H_C is shown for the two cooling field orientations. H_C is peaked at the onset of AF order in FeF_2 at $T_N = 78$ K. (c) Exchange bias H_E is shown for the two cooling field orientations. All curves are guides to the eye.

A feature of PNR, which we exploit for this study, is the capability to determine in one measurement, the fraction of the sample magnetization perpendicular to the applied field even if the net magnetization of the sample perpendicular to the applied field is zero. Since PNR yields an average of a spatially varying signal (i.e., the microscopic sample magnetization) taken over dimensions of the order of a Fresnel zone width (typically having lateral dimensions of microns and often smaller than the lateral width of a F domain), and the measurement is one of intensity, i.e., phase information is lost, the fraction of a sample with magnetization perpendicular to the applied field (N.B., either +90° or −90°) can be obtained. In contrast, techniques whose "averaging dimen-

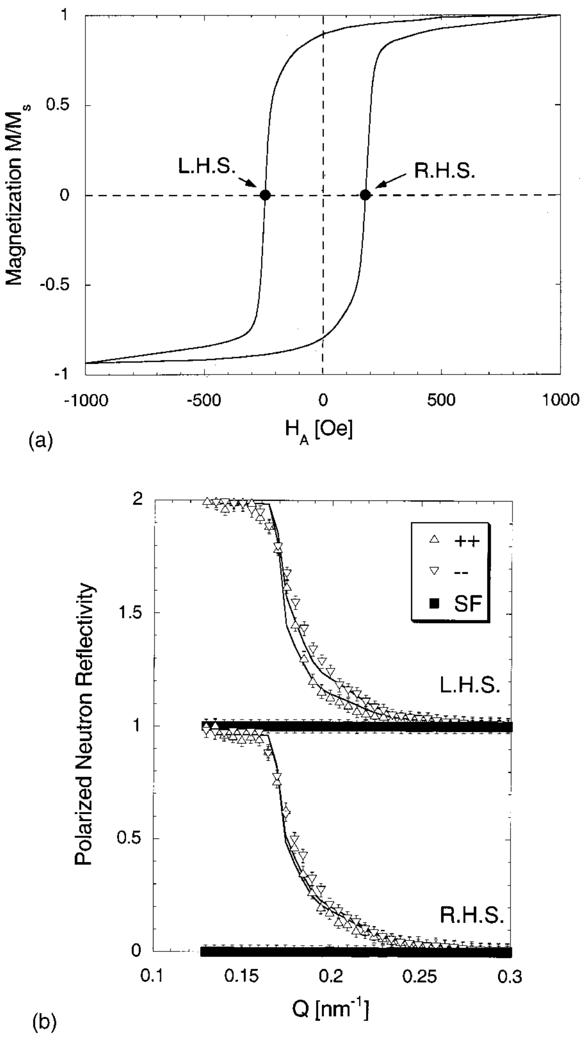


FIG. 4. (a) Hysteresis loop at 10 K for sample *p*-AF. The cooling field $H_{FC}=2$ kOe, was applied along the film plane (indicated by the "○" notation in the inset). The polycrystalline FeF_2 film is textured such that the (110) direction is perpendicular to the film plane. The exchange bias and coercivity for this sample are $H_E=-30\pm2$ Oe and $H_C=211\pm2$ Oe, respectively. H_E and H_C did not change significantly for different cooling field directions. (b) Polarized neutron reflectivity profiles taken at 11 K for applied fields shown by the closed symbols ● in (a) on the LHS and RHS for the same sample and cooling field orientation. No SF scattering is observed, indicating magnetization reversal via domain nucleation and wall motion. Data corresponding to LHS are shifted for the sake of clarity. Solid curves were obtained from fitted models of the type discussed in Ref. 13.

sion" encompasses the entire sample, for example, vector magnetometry, could yield zero signal for which a multitude of explanations are possible. We note that in the direction perpendicular to the sample surface, the "averaging dimension" for PNR is typically 1 nm thus, in principle, variations in the depth dependence of the sample magnetization can also be inferred.[31]

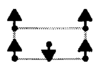
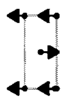
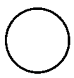
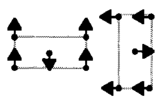
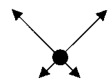
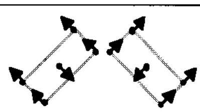
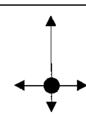
For the neutron-scattering experiment, the samples were cooled to $T \ll T_N$ (to 20 K for sample *u*-AF and 11 K for sample *p*-AF) in fields corresponding to the magnitudes and orientations used as cooling fields in the magnetometry study. Subsequent neutron measurements involved saturating the sample in a +2 kOe field, reducing the applied field to zero, reversing the direction of the applied field and then increasing the field strength until the NSF-reflectivity profiles (the ++ and −− profiles) were equal, i.e., ΔNSF=0. This field corresponds to $-H_C(T)+H_E(T)$ [●'s denoted as LHS (left-hand side) in Figs. 1, 2, and 4] where $M_\parallel=0$. The two non-spin-flip and two spin-flip cross sections were then measured for each sample and cooling field condition [upper panel of Figs. 1(b), 2(b), and 4(b)].[32] The right-hand sides (RHS) of the loops where $M_\parallel=0$ were measured by saturating the sample in a −2 kOe field, reducing the field to zero, reversing the field direction, and then increasing the field until the condition for $M_\parallel=0$ was achieved, i.e., ΔNSF=0, corresponding to $H_C(T)+H_E(T)$. The neutron reflectivity profiles for the RHS are shown in the lower panels of Figs. 1(b), 2(b), and 4(b). Results of the neutron and magnetometry measurements for samples *u*-AF and *p*-AF, along with those previously reported for the twinned AF sample (sample *t*-AF), for the different cooling field orientations are summarized in Table I.

Comparing the neutron-scattering results shown in Figs. 1(b) and 2(b), the cooling field orientation that produces exchange bias, i.e., $\mathbf{H}_{FC}\parallel[001]$ FeF_2 [Fig. 1(a), inset], is also one which leads to magnetization reversal via rotation. Magnetization reversal through magnetization rotation is evident by nonzero SF intensity in Fig. 1(b), since SF$\propto M_\perp$ [Fig. 1(b)].[33] The magnitude of the SF intensity suggests that 78% of the sample magnetization is perpendicular, i.e., $M_\perp=78\%$, to the applied field at coercivity. Magnetization rotation is promoted due to a *uniaxial* anisotropy in the F thin film that is *perpendicular* to the cooling field (and to the anisotropy axis of the AF, thus perpendicular exchange coupling is established).[34] Since exchange bias is observed, a unidirectional anisotropy in the F thin film *parallel* to the cooling field (and to the AF anisotropy axis) can be inferred. The directions of the anisotropy axes in the F layer are shown in Table I (Row 2, Column 5).

For the case of the second cooling field orientation, i.e., $\mathbf{H}_{FC}\parallel[\bar{1}10]$ FeF_2 [Fig. 2(a), inset], SF scattering was not observed [Fig. 2(b)], so magnetization reversal occurs via nucleation of a magnetic domain in the direction opposite to the saturating field, and motion of domain walls. In other words, cooling in a field with $\mathbf{H}_{FC}\parallel[\bar{1}10]$ FeF_2 produces only one uniaxial anisotropy in the F thin film. The uniaxial anisotropy lies along a direction parallel to the cooling field direction and perpendicular to the AF anisotropy axis. No exchange bias ($H_E=-2\pm2$ Oe) is observed in this prototypical example of a perpendicular exchange coupled system. This experimental observation reinforces a theoretical result of Schulthess and Butler[20,21] that spin-flop coupling does not by itself produce exchange bias.

From the study of sample *u*-AF, we conclude that perpendicular exchange coupling between F and AF layers is not a sufficient condition for exchange bias, since both cooling field conditions produce perpendicular exchange coupling, yet only one condition yielded exchange bias. This condition

TABLE I. Summary of results for untwinned AF (*u*-AF), twinned AF(*t*-AF), and textured polycrystalline AF (*p*-AF) samples.

Sample	$\uparrow H_{FC}$ Orientation (arrows show directions of Fe spins)	H_E [Oe]	$\sum_{domains} \lvert \vec{S_F} \bullet \vec{S_{AF}} \rvert$	F anisotropy axes (arrow length qualitatively indicates size of anisotropy)	Asymmetric reversal	Magnetization reversal mechanism
u-AF		-32±2	1		no	Rotation
u-AF		-2±2	0		no	Domain nucleation and wall motion
p-AF		-30±2	$\frac{2}{\pi}$[a]		no	Domain nucleation and wall motion
t-AF		-76±2	$\frac{1}{2}$		no	Rotation
t-AF		-325±2	$\sqrt{2}$		yes	Rotation (L.H.S.) Domain nucleation and wall motion (R.H.S)

[a]Reference 37.

is one where the F spins are oriented parallel (and antiparallel) to the spins of the AF during cooling. In other words, if exchange bias is desired, then the condition $|\mathbf{S_F} \cdot \mathbf{S}_{\mathrm{AF}}| \neq 0$ must be satisfied during field cooling. Generalizing to situations where the AF is twinned or polycrystalline, the cooling field condition leading to exchange bias is one where

$$\sum_{domains} |\mathbf{S_F} \cdot \mathbf{S}_{\mathrm{AF}}| \neq 0$$

with the sum taken over all AF domains. The sums are tabulated for the different samples in Table I.

Measurements of Sample *p*-AF (the sample with the textured polycrystalline AF film) observed only a unidirectional anisotropy (as indicated through exchange bias). By fabricating this sample in such a way that neither the AF film nor the F film could have macroscopic uniaxial anisotropies, perpendicular exchange coupling between the F and AF cannot exist. Yet, exchange bias was still observed; therefore, we conclude that perpendicular exchange coupling is neither a sufficient condition nor a requirement for exchange bias.

V. DISCUSSION

In comparing the neutron data [Figs. 1(b), 2(b), and 4(b)] for the different samples and cooling field orientations, one similarity is observed. Specifically, the neutron reflectivity profiles taken for coercive fields on either side of the same hysteresis loop are the same. In the first case (sample *u*-AF with $\mathbf{H}_{\mathrm{FC}} \| [001]$ FeF_2), SF scattering is observed on both sides of the loop indicating magnetization reversal via rotation, since $\mathrm{SF} \propto M_{\perp}$, so the magnetization reversal process is symmetric on either side of the loop. For the second case (sample *u*-AF with $\mathbf{H}_{\mathrm{FC}} \| [\bar{1}10]$ FeF_2), and in the case of sample *p*-AF (sample with the textured polycrystalline AF cooled in any field orientation (parallel to the sample plane), SF scattering is not observed on either side of the loop.[35] In the latter two cases, magnetization reversal occurs via domain nucleation (i.e., nucleation of domains with magnetization directed opposite to the saturating field) and domain wall motion. Even though the magnetization reversal process is different (rotation is not involved) from the first case (sample *u*-AF with $\mathbf{H}_{\mathrm{FC}} \| [001]$ FeF_2), the reversal processes are symmetric on either side of the same hysteresis loop. In other words, the samples with *untwinned single crystal or polycrystalline* AF thin films always exhibit symmetric magnetization reversal processes on either side of the F-hysteresis loop.

Previously, an asymmetry in the reversal process (rotation on the LHS and domain nucleation and wall motion on the RHS was reported for Fe layers exchange coupled to *twinned* (110) MnF_2 and FeF_2 single-crystal films.[8] (More recently a weak three fold anisotropy was identified in a similar Fe-twinned MnF_2 sample.[10] The three fold anisotropy may play an important role in asymmetrical magnetization reversal.) Asymmetrical magnetization reversal was observed when the twinned sample was cooled in a field applied along a direction that bisects the [001] axes of the FeF_2 twins (see figure inset in Table I, Row 6, Column 2). The exchange bias ($H_E = -325$ Oe) for sample *t*-AF is about one order of magnitude larger than those measured for samples *u*-AF or *p*-AF.

When the twinned sample (*t*-AF) was cooled in a field such that one half the sample had $\mathbf{H}_{\mathrm{FC}} \| [001]$ FeF_2 and the other half had $\mathbf{H}_{\mathrm{FC}} \| [\bar{1}10]$ FeF_2 (see figure inset in Table I, Row 5, Column 2), the magnetization reversal process occurred via rotation and was symmetric on both sides of the loop. For the symmetric reversal case, the exchange bias ($H_E = -76$ Oe) was reduced compared to the asymmetric reversal case ($H_E = -325$ Oe). This reduction is partly understandable in the context of the present results for sample *u*-AF, since unidirectional anisotropy may not be established in half of the twinned sample (i.e., the half with $\mathbf{H}_{\mathrm{FC}} \| [\bar{1}10]$ FeF_2).[36] In other words, during field cooling the quantity $\Sigma_{domains} |\mathbf{S}_{\mathbf{F}} \cdot \mathbf{S}_{\mathrm{AF}}|$ is smaller for the condition promoting symmetric magnetization reversal ($H_E = -76$) compared to the condition that promotes asymmetric magnetization reversal ($H_E = -325$ Oe).

However, the expression $\Sigma_{domains} |\mathbf{S}_{\mathbf{F}} \cdot \mathbf{S}_{\mathrm{AF}}|$ does not quantitatively account for the large exchange bias of the sample with the twinned AF. In the twinned system, the exchange bias is between two and ten times (depending upon cooling field orientation) larger than that measured for the untwinned or polycrystalline AF samples. A peculiar extrinsic feature of the twinned sample is the small (10 nm) lateral dimension of the twins, which, given that the anisotropy of FeF_2 is so large, likely limit AF domains (lateral) sizes to be equally small. Because the AF domains are so small and their orientations in the sample plane well defined (by the 90° twin relationship), interactions between the exchange coupling mechanism across different parts of the F-AF interface are likely important and may lead to frustration of the F layer (regardless of cooling field orientation). Frustration in the twinned system results, since one AF domain cannot adopt a perpendicular (low-energy) orientation with the F layer without another AF domain being forced into a parallel (high-energy) orientation. These interactions produce two uniaxial anisotropies in the F thin film that are rotated 45° from the anisotropy axes of the twinned AF thin film and lower the energy state for the entire system.

Frustration of perpendicular exchange coupling is intimately linked to large exchange bias. When the cooling field is applied parallel to a direction that will upon cooling become one of the two well-defined uniaxial anisotropies (produced through frustration of perpendicular exchange coupling), exchange bias is still further enhanced (Table I, Row 6, Column 2). This enhancement is correlated with an asymmetry in the magnetization reversal process on either side of the F-hysteresis loop, which tends to suppress reversal on one side of the loop while promoting reversal on the other side. Exchange bias is commonly assumed to be caused by an effective internal field; in fact, exchange bias may result from a modification of the magnetization reversal process, which is most dramatic in the twinned AF system.

VI. CONCLUSIONS

In summary, we systematically examined the influence of in-plane crystalline quality of the AF (e.g., untwinned single crystal, twinned single crystal, and textured polycrystal) on exchange coupling between an Fe thin film (the F) deposited onto (110) FeF_2 thin films (the AF). Regardless of cooling field orientation, perpendicular exchange coupling was always established in the sample with the untwinned single crystal AF. Yet, only the field orientation that aligned the F magnetization parallel to the anisotropy axis of the AF while cooling through T_N led to exchange bias. In the case of the sample with the textured (out-of-plane), polycrystalline (in-plane) AF, exchange bias was observed but perpendicular exchange coupling was not observed. These observations taken together lead us to conclude that perpendicular exchange coupling is neither sufficient nor required for exchange bias. However, frustration of the ferromagnet or perpendicular exchange coupling across the F-AF interface, can change the anisotropy of the ferromagnet, and alter magnetization reversal processes, which may play an important role in enhancing exchange bias.

In contrast, the orientations of individual grains within the plane of the sample with a textured (out-of-plane), polycrystalline AF thin film, are random, so well-defined uniaxial anisotropies in the F thin film were not formed. The inability to form a uniaxial anisotropy in the F thin film, crucial to enhancing exchange bias in samples with twinned (or untwinned) single crystal AF thin films, may preclude large exchange bias in the polycrystalline AF system.

The results from our systematic study of the influence of AF crystalline quality on EA, suggest that in order to enhance exchange bias three conditions should be fulfilled.

(1) The orientation between the spins in the AF and the F during field cooling must not be zero, i.e.,

$$\sum_{domains} |\mathbf{S}_{\mathbf{F}} \cdot \mathbf{S}_{\mathrm{AF}}| \neq 0$$

(see Column 4, Table I).

(2) By choice of cooling field orientation relative to the AF or by engineering the AF microstructure, a uniaxial anisotropy in the F layer should be formed in addition to and not collinear with the unidirectional anisotropy produced by field cooling (cf. Column 5, Table I).

(3) If multiple uniaxial anisotropies exist in the F layer, one anisotropy axis should be aligned with the cooling field, thus, promoting asymmetric magnetization reversal across the hysteresis loop (cf. Column 6, Table I).

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[30] "+" and "−" refer to whether the neutron spin is antiparallel or parallel, respectively, to the applied field.
[31] For example, the variation of $M_\perp$ with depth into the film can be obtained from PNR. To date measurements of SF scattering to momentum transfer Q as large as 2 nm^{-1} for Fe/FeF_2 and Fe/MnF_2 films have been obtained (by us) and the results indicate the magnetization in the F layer is uniform across the thickness of the film.
[32] The average of the two $+-$ and $-+$ cross-sections is shown as "SP" in Figs. 1(b), 2(b), and 4(b).
[33] In some cases, magnetization reversal through magnetization rotation can be inferred from magnetometry; however, hysteresis loops from Fe-Fe_2 or Fe-MnF_2 bilayers with the exception of exhibiting exchange bias can be otherwise symmetric even when the reversal process on either side of the loops are highly asymmetric (Ref. 8).
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[35] We note that the neutron reflectivity profiles from sample *u*-AF cooled in a field applied at 45° to the anisotropy axis were symmetric, and the SF scattering intermediate between those shown in Figs. 1(b) and 2(b).
[36] The relatively large exchange bias might still result from frustration in the F thin film of exchange coupling to differently oriented nanometer-sized AF twins. By extension from the study of the untwinned sample (sample *u*-AF), half of the twins promote unidirectional and uniaxial anisotropies (in different directions) in the F thin film, while the remaining half will promote only an uniaxial anisotropy in the F thin film. Competition between the formation of different anisotropies over lateral length scales of tens of nanometers may lead to frustration in the F thin film, and concomitant enhancement of exchange bias.
[37] The factor of $2/\pi$ is obtained by evaluating the integral representation, $\int_0^{2\pi}|\cos(\theta)|d\theta/2\pi$, of the sum $\Sigma_{\mathrm{domains}}|\mathbf{S}_\mathbf{F}\cdot\mathbf{S}_{\mathrm{AF}}|$.