Pinned magnetization in the antiferromagnet and ferromagnet of an exchange bias system

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Using polarized neutron reflectometry, we obtained separate depth profiles for pinned and unpinned magnetization across the interface of a ferromagnet/antiferromagnet bilayer as a function of the sign of exchange bias. The pinned and unpinned magnetization depth profiles are nonuniform and extend well beyond the chemical interface, suggesting an interfacial region magnetically distinct from its surroundings. A model that includes pinned and unpinned moments in the ferromagnet and antiferromagnet is developed for a complete description of the data.

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I. INTRODUCTION

The magnetization (M) of a ferromagnet (FM) can be pinned through exchange coupling with an antiferromagnet (AF). The magnetization (or moment) is pinned (M\text{\text{P}}) if it does not respond to modest fields (tens of kilo-oersted). A manifestation of exchange coupling between unpinned and pinned moments is the shift of the ferromagnetic hysteresis loop along the field axis—a phenomenon called exchange bias.6–9 The sign of exchange bias, positive or negative, refers to the shift of the loop with respect to the cooling field H\text{FC}. H\text{FC} is the field applied to the sample as it is cooled through the Néel temperature T\text{N} of the AF. If the loop is shifted in the direction opposite to H\text{FC}, then exchange bias is negative, –H\text{E}. Many FM/AF systems exhibit –H\text{E}. However, some systems containing FeF\text{2}, (Zn,Fe)F\text{2}, or MnF\text{2},6,7 or magnetic oxides, e.g., La_{0.67}Sr_{0.33}MnO\text{3}/SrRuO\text{3},8 and ferromagnetic systems, e.g., FeSn/FeGd,9 exhibit positive exchange bias, +H\text{E}. The sign of exchange bias can be influenced by environmental variables such as cooling field7 and temperature10,11 or by changes of the domain state12 in the AF bulk or at the FM/AF interface.10,13 +H\text{E} is commonly thought to arise from antiferromagnetic exchange coupling between unpinned moments in the FM and pinned uncompensated moments in the AF.6–9 Antiferromagnetic exchange coupling favors opposite (antiparallel) alignment of the unpinned and pinned moments, while ferromagnetic exchange coupling favors the same (parallel) alignment. An early model explaining the sign of exchange bias,9 considered two layers: the uncompensated pinned spins in the AF couple ferromagnetically to unpinned spins in the FM. The sign of H\text{E} was determined through the competition between the exchange coupling across the interface with the Zeeman interaction of the uncompensated AF spins with H\text{FC}. For weak H\text{FC}, antiferromagnetic coupling (across the FM/AF interface) prevailed, and the uncompensated moments in the AF were oriented and subsequently frozen in the direction opposite to H\text{FC}. This resulted in –H\text{E}. For large H\text{FC}, the Zeeman interaction between the field and the uncompensated moments in the AF overcame the exchange coupling across the AF/AF interface and aligned the uncompensated AF moments parallel to H\text{FC}, leading to +H\text{E} after cooling. The early model assumed that all uncompensated AF moments were located immediately at the FM/AF interface, and no depth dependence was considered. Further, the model assumed that all the FM moments were unpinned.

By identifying where the magnetization is pinned and the alignment of the unpinned magnetization with respect to the pinned magnetization as a function of field, the sign of exchange coupling, either ferromagnetic or antiferromagnetic, can be inferred. Key to understanding the origin of exchange bias is the measurement of pinned and unpinned magnetization depth profiles. Since the net magnetization of an ideal AF at zero field is zero, net magnetization in an AF refers to the uncompensated magnetization14 of a nonideal AF, which may also result from proximity to a FM.

Previously, Nogués et al.7 correlated exchange bias with pinned magnetization in Fe/FeF\text{2} and Fe/MnF\text{2} bilayers with twinned AF layers using a superconducting quantum interference device (SQUID) magnetometer.7 In their experiment, exchange bias was varied by changing the growth conditions of the samples or the strength of H\text{FC}. The pinned magnetization was obtained from vertical shifts of the hysteresis loops. For instance, an upward shift was related to a net pinned magnetization parallel to H\text{FC}. Ferromagnetic exchange coupling across the interface gave rise to an upward shift for all cooling fields with a minor change of exchange bias. In contrast, antiferromagnetic coupling led to a downward shift for small cooling fields. Thus, the sign of interfacial exchange coupling could be inferred from magnetometry, provided the pinned magnetization is confined in the AF close to the FM/AF interface. In particular, the pinned magnetization in the AF bulk must be negligibly small in comparison to the pinned interfacial magnetization.

Since the pinned magnetization of the AF cannot be distinguished from pinned interfacial magnetization with magnetometry, we previously measured the magnetization depth profile of an exchange bias system [Co/FeF\text{2} (untwinned)] using polarized x ray and neutron scattering.15 This study identified significant pinned magnetization in the AF bulk and showed that unpinned Fe moments beneath the Co/FeF\text{2} interface were antiparallel to unpinned Co moments above the interface. However, since neither x ray nor neutron-scattering measurements were taken in saturating fields applied in opposite directions and with both polarizations of the incident neutron (or x-ray) beam, the magnetization depth profiles could not be separated into pinned and unpinned...
components. Consequently, the previous study left unresolved the depth profile of the pinned magnetization across the Co/FeF₂ interface—a key issue in exchange bias.

Here, we report new neutron-scattering data that allow us to separate the magnetization depth profile into pinned and unpinned components with nanometer spatial resolution. We investigated a sample with exchange bias that could be switched from negative to positive just by cooling in weak or strong magnetic fields. This study yields insight into the sign of exchange coupling that gives rise to HE. Our observations motivate an exchange bias model with pinned magnetization not only in the AF as commonly assumed, but in the FM as well. Our model consistently explains the x-ray and neutron-scattering data.

II. SAMPLE PREPARATION

Exchange bias samples were prepared by sequential electron-beam evaporation of FeF₂, Co, and Al at a deposition rate of 0.05 nm/s onto MgF₂ polished substrates measuring 10×10 mm². The deposition temperatures were 300 °C for the FeF₂ layer and 150 °C for the Co and Al layers. The chemical depth profile of the sample was determined from x-ray reflectometry. The x-ray reflectivity is shown in Fig. 1 as a function of wave-vector transfer \( Q = k_f - k_i \), where \( k_f \) and \( k_i \) represent the final and incident wave vectors, respectively. Layer thickness and interface roughness are reported in Fig. 2. In-plane glancing angle x-ray diffraction (not shown) confirmed that the AF layer was an untwinned single-crystal film with [110]FeF₂∥[110]MgF₂. X-ray diffraction also indicates that the Co film is polycrystalline. A uniaxial magnetic anisotropy is present in the Co film with its easy axis along the FeF₂ easy axis, even for temperatures much higher than \( T_N \)—a property attributed to growth-induced anisotropy.

To promote +HE, a field of 7 kOe was applied along [001] FeF₂ at room temperature, and then the sample was cooled to 10 K. To promote −HE, the sample was cooled from 300 to 150 K in a field of 7 kOe (applied along [001] FeF₂), and then the sample was cooled to 10 K in a field of 100 Oe. The hysteresis loops measured with a SQUID magnetometer are shown in Fig. 3. The magnitude of HE was 2.2 kOe, regardless of the sign of exchange bias. For cooling field strengths intermediate between those we have chosen, the magnetization curves consists of two loops—one centered at −HE and the other centered at +HE. As \( H_{FC} \) is increased or decreased, one loop grows at the expense of the other. For \( H_{FC} = 7 \) kOe, a loop centered at −HE is mostly suppressed. Only about 4% of this loop remains as evidenced by the small (positive) increase in the magnetization (closed symbols, Fig. 3) near remanence. For \( H_{FC} = 100 \) Oe, a loop centered at +HE is completely suppressed.

III. MEASUREMENTS AND RESULTS

The magnetization depth profiles were obtained using polarized neutron reflectometry with polarization analysis.

![Fig. 1. (Color online) The x-ray reflectivity of the sample: observed (○) and calculated (solid curve) from the model structure (Fig. 2).](image1)

![Fig. 2. (Color online) Left: The chemical/nuclear model structure showing the chemical layers, their thickness (Δ) and interface roughness (σ). The real (solid curve) and imaginary (dashed) components of the x-ray scattering length density are shown. Right: The scattering model is shown.](image2)
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FIG. 3. Ferromagnetic hysteresis loops of the sample. The letters indicate the values of \( H_E \) and \( H_A \) corresponding to the neutron reflectivities in Fig. 4.

Neutron-scattering measurements were taken for \( \pm H_E \) and \( H_A = \pm 7 \text{ kOe} \).\(^{21}\) In each of these four configurations, two incident neutron beam polarizations (called up, i.e., neutron spin parallel to \( H_A \), and down) were used. From a comparative analysis of the resulting four pairs of reflectivities (Fig. 4), the magnetization depth profiles can be separated into pinned and unpinned components (Appendix) for each sign of exchange bias.

After field cooling the sample to 10 K, \( H_A \) was cycled \( \pm 7 \text{ kOe} \) three times—with the applied field of \( H_A = \pm 7 \text{ kOe} \) used for the neutron-scattering measurement. Positive applied field means that \( H_A \) and \( H_{FC} \) are parallel. The instrumental background corresponding to a reflectivity of order \( 10^{-7} \) has been removed. The data have been corrected for the nonperfect polarization of the neutron beam (polarization \( \sim 92\% \)), the slightly imperfect flipping efficiency of the neutron flipper (\( \sim 99.9\% \) efficient),\(^{22,23}\) and the wavelength dependence of the neutron spectrum. The error bars in Figs. 4 and 5 include contributions associated with these corrections and the number of neutrons detected for each spin state and \( Q \).

The two non-spin-flip reflectivities correspond to intensities measured for the same incident and reflected neutron beam polarizations, either both spin-up \( R^{++} \) or both spin-down \( R^{--} \). The difference between \( R^{++} \) and \( R^{--} \) is related to the component of the net magnetization vector \( \langle \vec{M}_1 \rangle \) parallel to \( H_A \) (Appendix). The averaging of \( \vec{M}_1 \) to obtain \( \langle \vec{M}_0 \rangle \) takes place over regions of the sample that scatter coherently—with a dimension we call the coherence dimension (typically microns in the sample plane).\(^{24}\) The specular reflectivity is sensitive to the average of \( \langle \vec{M}_0 \rangle \) within the coherence dimension. A nonzero value of \( \langle \vec{M}_0 \rangle \) modulates the specular reflectivity with periods in \( Q \) that are inversely related to the thicknesses of the magnetic layers (Appendix). The difference between \( R^{++} \) and \( R^{--} \) is related to the strength of the magnetization (Appendix). For the case where magnetic domains form with dimensions smaller than the coherence dimension, \( \langle \vec{M}_0 \rangle \) is reduced, and consequently, the difference between \( R^{++} \) and \( R^{--} \) is reduced. In addition, if the magnetizations of laterally separated domains are correlated, then off-specular diffuse (magnetic) scattering, where neutrons are reflected in directions other than specular, may be produced.\(^{25}\) Because off-specular diffuse neutron scattering is very weak, studies have been limited to systems of patterned films\(^{26–29}\) and multilayers of continuous films,\(^{30}\) which are constructed to amplify the diffuse scattering signal, or confined to the region of reciprocal space near the critical edge,\(^{31}\) where the specular reflectivity is close to unity. Spin dependence of off-specular neutron scattering is evidence for nonuniform laterally distributed magnetization, e.g., arising from domains, domain walls, and magnetic roughness. However, nonuniform laterally distributed magnetization is not a sufficient condition to produce off-specular scattering; for example, the lateral variations in the magnetization may not be correlated, or the correlation length may not be accessible for study with neutron reflectometry.\(^{32}\) We found no spin-dependent off-
spectral diffuse scattering either away from or near the critical edge (in contrast to observations in Ref. 31 of a different and much thicker magnetic system). For our system, diffuse scattering (of magnetic origin) is too weak to detect, or the dimensions of magnetic domains in our sample are not amenable for study with neutron reflectometry.

Qualitatively, the reflectivities (symbols in Fig. 4) (Ref. 33) for the same incident neutron beam polarization appear similar regardless of the orientation of \( H_A \) or the sign of \( H_E \). To obtain a quantitative measure of the effect of \( H_A \) and \( H_E \) on the reflectivities, we evaluate \( \zeta^2 = \sum_i (R_{0i} - R_{1i})^2 \) with standard deviation \( \Sigma_{0i} \) and \( R_{0i} = R(s, H_A, H_E, Q_i) \) with standard deviation \( \Sigma_{1i} \) represent two data sets (reflectivities) and neutron spin states up (+) or down (−). Table I shows \( \zeta^2 \), i.e., \( \zeta^2 \) normalized by the number of degrees of freedom \( v \) [equal to the number of observations (45)]. \( \zeta^2 \) is generally much greater than 1, indicating that the difference between a pair of reflectivities having the same incident neutron beam polarization is larger than what can be attributed to random fluctuations of the data.\(^{34}\) We show one such comparison in Fig. 5 for the pair of reflectivities corresponding to the conditions of \( H_A = ±7 \) kOe and \( H_E = ±2.2 \) kOe. The quasiperiodic variation of \( \zeta^2(Q) \) (Fig. 5) is a compelling evidence that differences between two reflectivities are not random. Since \( \zeta^2 \) is consistently greater than unity throughout Table I, we conclude that there are statistically significant differences between the reflectivities having different signs of \( H_E \) and \( H_A \) that warrant a quantitative analysis (model fitting).

The simplest exchange bias model treats the system as two magnetically distinct layers (e.g., FM and AF) coupled through an atomically sharp interface. However, more complex magnetic structures have been observed, including mixtures of ferromagnetic and antiferromagnetic exchange coupling,\(^7\) domain walls perpendicular \(^{13}\) and parallel \(^{15}\) to the FM/AF interface, and inhomogeneity of magnetic anisotropy in the FM and AF layers.\(^{35,36}\) Pertinent to our system are magneto-optical Kerr effect\(^7\) measurements of similarly prepared samples. These measurements identified an incomplete domain wall (i.e., a domain wall with a twist less than 180°) parallel to the FM/AF interface when a strong field was applied opposite to \( H_{FC} \) (or parallel to \( H_{FC} \) for \(-H_E\)).

In order to allow for complex magnetic structures, we generated magnetization depth profiles using a scattering model consisting of three different magnetic layers corresponding to the FM, interface (int) region, and AF. The spin-dependent neutron specular reflectivity was calculated using the dynamical formalism of Parratt.\(^{38}\) The process begins by choosing parameters for the scattering model, from which the spin-dependent neutron-scattering length density depth profiles \( \rho(z) = \rho_s(z) \pm C'M(z) \), where \( C' = 2.835 \times 10^{-7} \text{Å}^{-2} \text{cm}^3/\text{emu} \), are calculated. The “+” (“−”) sign is used for spin-up (down) incident neutron beam polarization. The parameters include the saturation magnetizations of each layer, \( M_{Co}, M_{int}, \) and \( M_{FeF2} \),\(^{39}\) magnetic roughness \( \sigma_1 \) and \( \sigma_2 \) on either side of the interface region, and the thicknesses of the three layers (Fig. 2).

We imposed several constraints on the scattering model. Since the nuclear scattering length density depth profile of the sample is not affected by \( H_A \) or \( H_{FC} \), it was constrained to be the same for all refinements. Second, the nuclear layer thickness and interface (chemical) roughness were optimized subject to the constraint that the optimal values lie within ranges shown in Fig. 2.\(^{40}\) Third, \( M_{Co} \) was constrained to be the same for all refinements. Fourth, the sum of the magnetic thickness of the Co layer and the magnetic thickness of the interfacial region on the Co side of the Co/FeF\(_2\) chemical interface was equal to the nuclear thickness of the Co layer. Fifth, the sum of the magnetic thickness of the interfacial region on the FeF\(_2\) side of the Co/FeF\(_2\) chemical interface and the magnetic thickness of the FeF\(_2\) layer was equal to the nuclear thickness of the FeF\(_2\) layer. Finally, the uncompensated magnetization in the bulk of the FeF\(_2\) layer \( M_{FeF2} \) was constrained to be pinned as determined by Roy et al.\(^{15}\) These

TABLE I. \( \zeta^2 \) for all possible combinations of reflectivity with the same incident neutron beam polarization but having different \( H_E \) and \( H_A \). \( \zeta^2 \leq 1 \) indicates statistically similar and \( \zeta^2 > 1 \) statistically different data sets.

<table>
<thead>
<tr>
<th>( H_E )</th>
<th>( H_A )</th>
<th>( \zeta^2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>−2.2 kOe</td>
<td>+7 kOe</td>
<td>6.5</td>
</tr>
<tr>
<td>+7 kOe</td>
<td>+7 kOe</td>
<td>31.5</td>
</tr>
<tr>
<td>−2.2 kOe</td>
<td>−7 kOe</td>
<td>22.1</td>
</tr>
<tr>
<td>+7 kOe</td>
<td>−7 kOe</td>
<td>30.3</td>
</tr>
<tr>
<td>+2.2 kOe</td>
<td>−7 kOe</td>
<td>21.5</td>
</tr>
<tr>
<td>−7 kOe</td>
<td>−7 kOe</td>
<td>1.8</td>
</tr>
<tr>
<td>+2.2 kOe</td>
<td>+7 kOe</td>
<td>4.0</td>
</tr>
<tr>
<td>−7 kOe</td>
<td>+7 kOe</td>
<td>6.4</td>
</tr>
<tr>
<td>−2.2 kOe</td>
<td>−7 kOe</td>
<td>4.4</td>
</tr>
<tr>
<td>+2.2 kOe</td>
<td>+7 kOe</td>
<td>7.8</td>
</tr>
<tr>
<td>−7 kOe</td>
<td>−7 kOe</td>
<td>5.6</td>
</tr>
<tr>
<td>+2.2 kOe</td>
<td>+7 kOe</td>
<td>1.8</td>
</tr>
<tr>
<td>−7 kOe</td>
<td>+7 kOe</td>
<td>2.0</td>
</tr>
</tbody>
</table>

FIG. 5. (Color online) Plot showing quasiperiodic variation of \( \zeta^2(Q) \) (open symbols) obtained from a comparison of the reflectivities taken for \( H_A = ±7 \) kOe and \( H_E = ±2.2 \) kOe and spin-down incident neutron beam polarization. These reflectivities times \( Q^2 \) are shown as the closed symbols.
constraints allow the interfacial region to have a magnetization depth profile containing pinned and unpinned components that may differ (though need not) from those of the adjoining layers, while preserving the chemical depth profile determined by x-ray reflectometry. The parameters were perturbed to minimize a measure of error $\chi^2$ between the calculated and observed reflectivities using the Powell optimization procedure. The calculated reflectivities that best fit the data are shown as solid curves in Fig. 4 and the magnetization in Fig. 6.

The projection of the pinned magnetization depth profiles along [001] FeF$_2$ for $\pm H_F$ can be obtained by determining the fractions of the magnetization depth profiles that do not change when $H_A$ is reversed. If the magnetization depth profile $M(z)$ is composed of pinned and unpinned components, $M^p(z)$ and $M^u(z)$, respectively, then $M(\pm H_A,z) = M^p(z) \pm M^u(z)$, where $M(z)$ is shown as the red ($-H_A$) and blue ($+H_A$) curves in Fig. 6, and $M^u(-H_A,z) = -M^u(+H_A,z)$ is assumed. Solving for $M^p(z)$ and $M^u(z)$ yields

$$M^p(z) = \frac{1}{2} [M(+H_A,z) + M(-H_A,z)],$$

$$M^u(z) = \frac{1}{2} [M(+H_A,z) - M(-H_A,z)].$$

$M^p(z)$ is plotted for the cases of $+H_E$ (orange) in Fig. 6(a) and $-H_E$ (dashed purple) in Fig. 6(b) and in much greater detail in Fig. 7(a). $M^u(z)$ is shown in Fig. 7(b) for $+H_E$ (orange) and $-H_E$ (dashed purple).

**IV. DISCUSSION OF THE MAGNETIZATION DEPTH PROFILE**

There is a strong correlation relating the direction of the pinned magnetization with the sign of exchange bias. For $-H_E$, the pinned interfacial magnetization is parallel to $H_{FC}$ (dashed purple curve, Fig. 7). For $+H_E$, the pinned interfacial magnetization is opposite to $H_{FC}$ (orange curve, Fig. 7). The alignments of unpinned and pinned magnetizations are shown by red and green arrows, respectively, for cases of $-H_E$ (upper panel) and $+H_E$ (lower panel), and $-H_A$ (left) and $+H_A$ (right) relative to $H_{FC}$ in Fig. 8. We find no evidence for a net pinned magnetization in the bulk of the AF for $-H_E$. In contrast, a net pinned magnetization parallel to $H_{FC}$ is observed in the AF bulk for $+H_E$, which is consistent with Roy et al.\textsuperscript{15}

The orientation of the pinned interfacial magnetization is contrary to the expectation based on the assumption of antiferromagnetic exchange coupling across an atomically sharp interface between unpinned moments in the FM and pinned moments in the AF. Specifically, the expected orientation is one with the green arrows corresponding to the interfacial magnetization in Fig. 8 reversed. In order to check whether the neutron data are truly sensitive to the orientation of the pinned interfacial magnetization, we repeated the refinement process constraining the pinned interfacial magnetization to be reversed from the orientation shown in Fig. 8. The best

![Image of magnetization depth profiles](image-url)
fitting reflectivities are shown as the dashed curves in the lower panel of Fig. 4. The disagreement between the dashed curves and the neutron data is so compelling that a model for the pinned interfacial magnetization being opposite to $H_{FC}$ for $-H_E$ and parallel to $H_{FC}$ for $+H_E$ can be rejected.

Our analysis shows that the magnetic interface has a thickness larger than that attributable to chemical roughness. This implies the existence of pinned moments in the FM layer and not just in the AF layer as commonly assumed—a realization that hints towards the presence of domain walls that extend from the interface into the FM and/or AF.

For $-H_E$, the full width at half maximum (FWHM) of the pinned interfacial magnetization depth profile is $52 \pm 2$ Å and extends $27 \pm 1$ Å into the Co layer and $25 \pm 1$ Å into the FeF$_2$ layer relative to the chemical position of the Co/FeF$_2$ interface (at $53 \pm 2$ Å from the Co/Al interface). The average of the pinned magnetization in the interfacial region is $\langle M'_{int} \rangle = 20 \pm 3$ emu/cm$^3$ (for $-H_E$). In the same region, the unpinned magnetization is $\langle M''_{int} \rangle = 575 \pm 16$ emu/cm$^3$. The ratio of $\langle M'_{int} \rangle$ to the sum of $\langle M'_{int} \rangle + \langle M''_{int} \rangle$ is the fraction $f'_{-H_E} = 3.4 \% \pm 0.6\%$ of pinned magnetization to the total magnetization in the interfacial region for $-H_E$.
TABLE II. Magnetization and areal moment density for ±H_E. The “+” (“−”) sign of the magnetization or areal moment density indicates that the quantity is parallel (antiparallel) to the cooling field.

<table>
<thead>
<tr>
<th>Measurement</th>
<th>For +H_E</th>
<th>For -H_E</th>
</tr>
</thead>
<tbody>
<tr>
<td>Co magnetization M_Co (emu/cm^2)</td>
<td>1299±30</td>
<td>1299±30</td>
</tr>
<tr>
<td>Magnetic interface width FWHM (nm)</td>
<td>5.2</td>
<td>2.7</td>
</tr>
<tr>
<td>Pinned interfacial magnetization (M_P^int) (emu/cm^3)</td>
<td>20±3</td>
<td>-47±3</td>
</tr>
<tr>
<td>Unpinned interfacial magnetization (M_U^int) (emu/cm^3)</td>
<td>575±16</td>
<td>601±14</td>
</tr>
<tr>
<td>FeF_2 bulk pinned magnetization (M_P^{FeF_2}) (emu/cm^3)</td>
<td>0±2.5</td>
<td>5.6±2.5</td>
</tr>
<tr>
<td>Percent pinned interfacial magnetization compared to the total interfacial magnetization, f_P^int = \frac{\langle M_P^int \rangle}{\langle M_P^int \rangle + \langle M_U^int \rangle} (%)</td>
<td>3.4±0.6</td>
<td>7.3±0.5</td>
</tr>
</tbody>
</table>

The areal moment density is the magnetization depth profile M(z) integrated over the entire sample, e.g., m^P = \int_{-125}^{125} M^P(z)dz (Table II). The ratio of the pinned interfacial areal moment density m^P to the areal moment density of the whole sample is related to a vertical shift of the ferromagnetic hysteresis loop. Since the pinned areal moment density is positive, i.e., its direction is parallel to H_FC, an upward shift of the hysteresis loop (measured with magnetometry) along the vertical axis corresponding to 1.9% ±0.3% of M_Co is expected for -H_E. For small H_FC, such as that used to establish -H_E for our sample, Nogués et al.7 found upward or downward shifts of the hysteresis loop, depending on the sample.

For +H_E, the pinned interfacial magnetization is more tightly confined near the chemical Co/FeF_2 interface than for -H_E, and a net pinned magnetization is present in the bulk of the FeF_2 layer. The FWHM of the pinned interfacial magnetization is 27±2 Å wide and extends 16±1 Å into the Co layer and 11±1 Å into the FeF_2 layer. As for the case of -H_E, the center of mass of the pinned interfacial magnetization for +H_E is shifted somewhat towards the Co side of the chemical Co/FeF_2 interface. The mean value of the pinned interfacial magnetization is \langle M_P^int \rangle = -47±3 emu/cm^3 (the negative sign indicates that the magnetization is opposite to H_FC) for +H_E. The unpinned magnetization in the same region is \langle M_U^int \rangle = 601±14 emu/cm^3. The fraction of pinned interfacial magnetization for +H_E is f_P^H_E = 7.3±0.5%—slightly more than twice that found for -H_E.

The point where the pinned magnetization reverses sign for +H_E is 21±2 Å below the chemical Co/FeF_2 interface (i.e., inside the FeF_2 layer and ~125 Å from the sample’s surface). The extension of the pinned interfacial magnetization into the FeF_2 layer is in the range of 20 to 35 Å previously identified from our x-ray and neutron-scattering studies. If we attribute the pinned magnetization above the depth of 125 Å to the interfacial region and that below this depth to the FeF_2 layer, we obtain areal moment densities of m^P = \int_{-125}^{125} M^P(z)dz = (1.6±0.1) × 10^{-5} emu/cm^2 for the interfacial region and m^{FeF_2} = \int_{125}^{200} M^P(z)dz = (2.1±0.1) × 10^{-5} emu/cm^2 for the bulk of the FeF_2 layer. A negative areal moment density is opposite to H_FC.

The interfacial areal moment density m^P is the number of pinned interfacial moments projected onto a plane per unit area. The fraction of pinned magnetization in the interfacial region compared to the total magnetization of the interfacial region is f^P. Our sample is one with an exchange bias of equal magnitude regardless of its sign, i.e., |H_E| = +H_E. If exchange bias results from pinned interfacial moments, we conclude that |H_E| is more closely related to the areal moment density than to the fraction of pinned interfacial magnetization since \langle m^{U^int}(H_E) \rangle = m^{P^int}(H_E) + \langle m^{U^int}(H_E) \rangle, whereas f^P_{-H_E} ≈ 2f^P_{+H_E}. Had the thickness of the interfacial region been the same for ±H_E, then the fraction of pinned interfacial magnetization would have been the same, too.

The pinned areal moment density for the whole +H_E sample is slightly positive m^P = m^{P^int} + m^{P^{FeF_2}} = (0.5±0.2)
\(10^{-5}\) emu/cm\(^2\). Using this value and that of the unpinned areal moment density \(n^i = (6.18 \pm 0.14) \times 10^{-4}\) emu/cm\(^2\), we expect an upward shift of the hysteresis loop along the vertical axis of 0.8\% \pm 0.3\% of \(M_C\) for \(+H_E\). The upward shift of the hysteresis loop is somewhat less than for the case of \(-H_E\) because the pinned interfacial areal moment density partially cancels that of the FeF\(_2\) layer. Note that upward shifts of the hysteresis loops are expected regardless of the sign of exchange bias for this sample; thus, the sign of exchange coupling across the Co/FeF\(_2\) interface cannot be inferred from a vertical shift of the hysteresis loop. It is noteworthy that the conclusions of Noguès et al.\(^7\) are based on the assumption that pinned moments are located only at the interface. Hence, that model does not capture the effects that are attributed to the difference between the pinned moments in the interfacial layer and those in the FeF\(_2\) bulk.\(^{45,46}\) More generally, pinned uncompensated magnetization in the bulk of an antiferromagnet (or a ferrimagnet) may dominate the vertical shift of the hysteresis loop; thus, the sign of exchange coupling across the FM/AF interface cannot be determined from the sign of the vertical shift of a hysteresis loop with magnetometry.

The absolute value of the gradient of \(M^i(z)\) is larger (i.e., steeper) on the Al side of the Co layer compared to that on the FeF\(_2\) side of the Co layer [Fig. 7(b)]. To understand the asymmetry, we compare \(M^i(z)\) to the profile that corresponds to the chemical variation of the Co number density profile (obtained from x-ray reflectometry). The fraction of the sample composed of Co as a function of depth into the sample \(F_{Co}(z)\) is calculated using

\[
F_{Co}(z) = \begin{cases} 
\frac{1}{2} 
& \text{if } z < \Delta_0 \\
1 + \text{erf}\left(\frac{z - \Delta_{Al/Co}}{\sqrt{2}\sigma_{Al/Co}}\right) & \text{if } z > \Delta_0
\end{cases}
\]  

(2)

where \(\Delta_{Al/Co}\) and \(\Delta_{Co/FeF_2}\) are the positions of the interfaces, \(\Delta_0 = \Delta_{Al/Co} + \Delta_{Co/FeF_2}\), and \(\sigma_{Al/Co}\) and \(\sigma_{Co/FeF_2}\) are the roughnesses of the interfaces (Fig. 2). The anticipated magnetization depth profile of the Co moments is the product \(F_{Co}(z)M_C\) [black-short-dashed curve in Fig. 7(b)]. Near the Co/FeF\(_2\) interface, \(F_{Co}(z)M_C\) is considerably larger than \(M^i(z)\) for \(\pm H_E\). The missing magnetization near the Co/FeF\(_2\) interface represents about 22\% of the anticipated areal moment density of the Co layer. Suppression of interfacial magnetization (along \(H_A\)) might result from a number of reasons including (1) domains that form such that the net magnetization of Co is diminished\(^47\) (2) the magnetization from unpinned uncompensated moments in the FeF\(_2\) layer negating some of the magnetization from unpinned Co moments (through antiferromagnetic exchange coupling)\(^15\) and (3) the magnetization that may be rotated away from the applied field, diminishing the projection of the magnetization along the applied field. Suppression of interface magnetization along the applied field is consistent with the previously identified loss of Co moment density near the Co/FeF\(_2\) interface.\(^15\)

V. MODEL FOR EXCHANGE BIAS

The magnetization depth profiles provide several important insights. First, the width of the pinned magnetization near the Co/FeF\(_2\) (FM/AF) interface is considerably larger than its chemical width \(52\pm2\) Å \((-H_E)\) and \(27\pm2\) Å \((+H_E)\) compared to \(9\pm1\) Å. Second, for \(\pm H_E\), the center of mass of interfacial pinned magnetization is on the Co side of the Co/FeF\(_2\) interface rather than the FeF\(_2\) side. Together, these observations suggest that the interfacial region of pinned magnetization is not only in the AF as generally assumed, but also in the FM. The existence of pinned moments in the FM implies the existence of incomplete domain walls that separate domains of pinned magnetization from unpinned magnetization. Additionally, the much larger width of the interfacial pinned magnetization compared to the chemical interface width (roughness) suggests a magnetic structure that is distinct from either the FM or AF and extends into both regions. Guided by these observations, we propose an exchange bias model that assumes

1. FM moments that greatly outnumber uncompensated AF moments,\(^15\)
2. ferromagnetic exchange coupling, \(J_{FM}\), between FM moments,
3. antiferromagnetic exchange coupling, \(J_{FM-AF}\), between FM and AF moments across the interface,\(^5,15\)
4. some uncompensated AF moments that couple to the rigid structure of compensated AF moments below \(T_N\), with a pinning field larger than \(H_A\) (these are pinned moments), and
5. other uncompensated AF moments, particularly at the interface, that remain unpinned below \(T_N\).\(^15,18\)

Shown to the right of the magnetization-depth-profile schematics in Fig. 8 are complementary schematics of the spin model for exchange bias. The magnetization in the FM and AF layers are dominated by FM and uncompensated AF moments, respectively. Since the magnetic interface straddles portions of the FM and AF, and because there are more FM moments than uncompensated AF moments, the orientation of the interfacial magnetization is mostly determined by the FM. Thus, for \(-H_E\), pinned FM (uncompensated AF) moments are parallel (antiparallel) to \(H_{FC}\) as expected when exchange coupling across the FM/AF interface is antiferromagnetic for \(+H_E\). The key element of the exchange bias model is the presence of pinned and unpinned moments in the FM and AF. This element is in contrast to the classical exchange bias model, where the moments in the FM are fully unpinned and the moments in the AF are pinned.

Our exchange bias model is not only consistent with the neutron- and soft-x-ray\(^15\) scattering studies but is also consistent with an important conclusion of Ohldag et al.\(^48\) Based on x-ray circular and linear dichroism studies, where the total electron yield was measured from a Co/FeF\(_2\) bilayer exhibiting large \(-H_E\), Ohldag et al.\(^48\) concluded that the coupling between pinned Fe (AF) moments and Co (FM) moments is antiferromagnetic below \(T_N\). Thus, at remanence Co moments will be antiparallel to pinned Fe moments. In our model, FM moments are antiparallel to the pinned AF moments at remanence for \(\pm H_E\).
The exchange bias model also explains why the same sample may exhibit \( \pm H_E \) depending on the strength of \( H_{FC} \). First, consider the case for \( +H_E \). To achieve \( +H_E \), strong \( H_{FC} \) is required. The field orient uncompensated moments in the AF layer and the moments in the FM layer parallel to \( H_{FC} \) (overcoming \( J_{FM\text{-}AF} \) at room temperature). After cooling to low temperatures, some of the uncompensated AF moments become pinned by coupling to the ordered AF moments. For fields that do not overcome (i.e., frustrate) \( J_{FM\text{-}AF} \) at low temperature, FM moments that are (antiferromagnetically) exchange coupled to pinned AF moments will be effectively pinned, but in a direction opposite to the pinned AF moments. FM moments that are exchange coupled to unpinned AF moments will be unpinned and will point along \( H_A \) (because the FM moments outnumber the AF moments). The unpinned AF moments will align in the opposite direction due to antiferromagnetic exchange coupling with the pinned FM moments. As the field is reduced below \( H_E \) from positive saturation, ferromagnetic exchange coupling between the unpinned and pinned FM moments will cause the magnetization of the FM to reverse its direction. Thus, the magnetization of the FM layer is negative for \( H_A \leq +H_E \), and the hysteresis loop is shifted in the direction of \( H_{FC} \) (yielding \( +H_E \)). Since the FM moments constitute the majority of the pinned and unpinned interfacial magnetization, this magnetization points opposite to the cooling field for \( H_A \leq +H_E \). As the field is increased from negative saturation above \( -H_E \), unpinned FM and pinned AF moments rotate together to maintain an antiparallel relationship with the FM magnetization parallel to \( H_A \) for \( H_A > +H_E \). Consequently, domain walls (dashed lines in Fig. 8) form between pinned and unpinned moments (for \( H_A > +H_E \)). Magnetic domains decrease the net magnetization along \( H_A \) of the FM moments in the vicinity of the FM/AF interface, as observed here and previously in suppression of the unpinned interfacial magnetization.\(^{15} \) (Experimental evidence for domains in similarly prepared samples has been published in Refs. 13 and 37.) The key requirement to produce \( +H_E \) is for the product \( H_{FC}M_{AF} \) to be large during cooling, so that \( M_{AF} \) will be parallel to \( H_{FC} \) at the expense of \( J_{FM\text{-}AF} \).

Now, consider the case for \( -H_E \). To achieve \( -H_E \), the sample is cooled in a field just large enough to keep the FM magnetization parallel to \( H_{FC} \), but not large enough to frustrate \( J_{FM\text{-}AF} \). Because \( H_{FC} \) is weak, the alignment of uncompensated AF moments in the bulk is not favored; thus, little net magnetization is induced in the AF bulk. However, some AF moments near the FM/AF interface are aligned opposite to \( H_{FC} \) due to antiferromagnetic exchange coupling between these moments and FM moments that are aligned parallel to \( H_{FC} \). At low temperatures, some of the AF moments become pinned in the same direction. These moments effectively pin the adjacent FM moments in the direction of the cooling field. Thus, when \( H_{FC} \) is weak, exchange coupling between FM and AF moments and the domain state of the FM layer determine the domain state of the AF. Subsequently, if \( H_A \) is increased towards positive saturation, FM moments, pinned or unpinned, will be parallel to \( H_A \), and the AF moments, pinned or unpinned, will be opposite \( H_A \). For a strong negative field, unpinned FM moments (and unpinned AF moments) move to be parallel (or opposite) to the \( H_A \). This behavior of the unpinned moments is similar to the case of \( +H_E \) and strong positive fields, while the pinned FM and pinned AF moments remain fixed (in directions opposite to the case of \( +H_E \)).

Regardless of the sign of \( H_E \), when a strong field is applied in the direction the hysteresis loop is shifted, domain walls form to accommodate the different directions of the pinned and unpinned moments (or magnetization) (Fig. 8). The work done in creating the domain walls is related to \( |H_E| \). For our sample, the work to create domain walls and thereby to reverse the sample magnetization is independent of the sign of \( H_E \).

In our exchange bias model, the direction of the pinned AF moments is correlated with the sign of \( H_E \) and anticorrelated with the direction of the pinned FM moments. The direction of pinned AF moments is determined by competition between \( H_{FC} \) and \( J_{FM\text{-}AF} \), which favor pinned moments in the AF that are parallel or antiparallel to strong or weak cooling fields, respectively.

\section{VI. Conclusions}

Good depth resolution and the ability to probe all depths in the sample with comparable sensitivity are intrinsic to neutron scattering. These attributes allowed us to characterize the magnetization depth profile across a Co/FeF\(_2\) bilayer as a function of the sign of exchange bias. The distribution of pinned magnetization across the interface is nonuniform and is correlated with the sign of exchange bias. The width of the interfacial magnetization exceeds the width of the chemical interface—extending between \( \sim 13 \) \text{Å} (for \( +H_E \)) and \( \sim 26 \) \text{Å} (for \( -H_E \)) away from the average chemical interface into the AF and FM layers. An exchange bias model that consistently explains all experimental data was proposed. In this model, the interfacial region of pinned magnetization is composed of both pinned Co and pinned Fe moments residing in the AF and FM layers, respectively. This arises from antiferromagnetic exchange coupling that pins the Co moments close to the interface to pinned Fe moments, thus making these Co moments unresponsive to modest applied magnetic fields.

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\section{Appendix}

To see that the unpinned and pinned magnetization depth profiles can be obtained from the polarized neutron (or x-ray)
reflectivities $R_{BA}^{\pm}(\pm H_A, Q)$, consider a uniformly magnetized film on a nonmagnetic substrate. Using the Born approximation, the reflectivity is given by

$$R_{BA}^{\pm}(Q) = \frac{16\pi^2}{Q^2} \left| \int_{-\infty}^{\infty} e^{iQz}\rho_{\pm}(z)dz \right|^2,$$

(A1)

where $\rho_{\pm}(z)$ is the neutron-spin-dependent scattering length density at a depth of $z$ into the sample averaged over its lateral dimensions. For a thin FM layer with a nuclear scattering length density of $\rho_0$ and magnetization $M$ on a nonmagnetic substrate with nuclear scattering length density $\rho_1$, $\rho_{\pm}(z)$ is

$$\rho_{\pm}(z) = \begin{cases} \rho_0 \pm C'M, & 0 < z < \Delta \\ \rho_1, & z > \Delta \end{cases}$$

(A2)

where $C' = 2.853 \times 10^{-9} \text{Å}^{-2} \text{cm}^3/\text{emu}$. Substituting Eq. (A2) into Eq. (A1), integrating to obtain $R_{BA}^{\pm}(Q)$, and neglecting terms in the delta function (i.e., considering only $Q \neq 0$), we obtain

$$R_{BA}^{\pm}(H, Q) = R_p[\rho_0 - \rho_1 + C'M(H)]^2(1 - \cos Q\Delta),$$

(A3)

where $R_p = 16\pi^2/Q^4$. If we consider $M$, the projection of $\mathbf{M}$ onto the direction of $H_A$, to be composed of pinned ($M^p$) and unpinned ($M^u$) magnetization, then $M = M^p + M^u$. For applied fields of $\pm H_A$ that saturate the unpinned magnetization, $M(\pm H_A) = M^p \pm |M^u|$, Eq. (A3) becomes

$$R_{BA}^{\pm}(\pm H_A, Q) = R_p[\rho_0 - \rho_1 + C'M^p \pm C'|M^u|](1 - \cos Q\Delta),$$

$$R_{BA}^{\pm}(\pm H_A, Q) = R_p[\rho_0 - \rho_1 - C'M^p \pm C'|M^u|](1 - \cos Q\Delta).$$

(A4)

From measurements using one incident beam polarization and opposite applied field directions, only two quantities can be obtained: $|M^u|$ and one of $\rho_0 - \rho_1 + C'M^p$ or $\rho_0 - \rho_1 - C'M^p$. However, if measurements are made using both incident beam polarizations and opposite field directions, then $|M^p|$ and $M^p$ can be obtained separately. Since complicated magnetization depth profiles can be represented by a sequence of arbitrarily thin uniform profiles, $M^p$ and $M^p$ can be separated in general. The primary motivation to use polarized neutron reflectometry is to obtain the magnetization depth profiles for systems that are not uniform.

Previously, we stated that the difference between the non-spin-flip reflectivities was related to the magnetization depth profile. To understand what is meant by "related," Eq. (A3) is rewritten as

$$R_{BA}^{+}(H, Q) - R_{BA}^{-}(H, Q) = 4R_p(\rho_0 - \rho_1)C'M(1 - \cos Q\Delta).$$

(A5)

The difference $R_{BA}^{+}(H, Q) - R_{BA}^{-}(H, Q)$ is proportional to the magnetization through terms that vary (oscillate) with $Q$. These terms are the Fourier components of the magnetization depth profile. Since the oscillatory term is always positive (for a uniform film), the sign of $R_{BA}^{+}(H, Q) - R_{BA}^{-}(H, Q)$ depends on the product of the signs of $\rho_0 - \rho_1$, i.e., the difference between the nuclear scattering length densities of the magnetic film and its substrate, and the magnetization $M$. Thus, the sign of $R_{BA}^{+}(H, Q) - R_{BA}^{-}(H, Q)$ for a single $Q$ is not necessarily indicative of the orientation of $M$ relative to $H_A$.

For example, the nuclear scattering length density of Co is about one-half that of FeF$_2$; therefore, for a uniformly magnetized Co film on FeF$_2$, $R_{BA}^{+}(H, Q) > R_{BA}^{-}(H, Q)$ for all $Q$. On the other hand, for the same Co film on Si, $R_{BA}^{+}(H, Q) > R_{BA}^{-}(H, Q)$ for all $Q$ because $\rho_{Co} > \rho_{Si}$. Provided the sample is simple, and the sign of $\rho_0 - \rho_1$ is known, the orientation of $M$ relative to $H_A$ can be obtained from inspection of $R_{BA}^{\pm}(H, Q)$ for a single $Q$. However, in practice, the sample structure is usually complex (e.g., the magnetic film may be capped to prevent oxidation, the magnetization may not be uniform, etc.). For complex structures, the orientation of $M$ relative to $H_A$ is obtained from an analysis (e.g., through model fitting) of many measurements of $Q$, as we have shown for our sample.

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14. The source of the uncompensated AF moments may be due to interfacial inhomogeneities, defects, or piezomagnetism.
16. The x-ray scattering length densities for Al and Co are in close agreement with literature values. The x-ray scattering length density for FeF$_2$ is about 17% larger than anticipated. This error could be explained by a misalignment of the sample by 0.025°.
17 By cooling the sample from room temperature to 150 K in a field of 7 kOe, we assure that the sample’s magnetization remains saturated above the Néel temperature of FeF$_2$.


21 7 kOe is the largest field that can be applied by our electromagnet when it accommodates the cryostat.


32 Indeed, in studies of patterned films, the dimensions of the patterning have been chosen to assure that off-specular scattering could be detected by neutron reflectometry.

33 Four weeks of neutron beam time were required to measure the eight reflectivity curves with a mean statistical precision of 5%.


39 $M_{\text{FeF}_2}$ represents the uncompensated magnetization of the antiferromagnetic FeF$_2$ layer.

40 For example, the optimal thickness of the Al layer determined by x-ray reflectometry was 15±2 Å, whereas the optimal value determined from the neutron reflectivity analysis was 13 Å.


42 Errors on the spatial extent of the layers were determined from a statistical analysis of the fit to the neutron data. Sensitivity of the fit to fine details of the magnetization profile is limited by the finite range of $Q$ over which the reflectivity was obtained. Averaging of the scattering amplitude occurs over lateral dimensions (tens of microns) related to the coherence of scattering and may further obscure fine details.

43 The error in the pinned areal moment density was determined from the variation of the pinned magnetization depth profile, which was obtained from a statistical analysis of the fit to the neutron data.

44 To define $f^p$, the peak value of $M^p$ could have been used instead of $\langle M^p \rangle$. This approach yields larger values for $f_{\text{fit}}$ and $f_{\text{exp}}$ of $\sim 9\%$ and $\sim 5\%$, respectively. The ratio of the two values is still about a factor of 2.


49 When fitting models to reflectivity data the dynamical formalism (Ref. 38) was used.

50 More precisely, the average is taken over the region of the sample which coherently scatters the neutron (or x-ray) beam.

51 A similar situation is encountered with resonant soft-x-ray reflectometry which makes interpretation of the sign of coupling between two magnetic species difficult, if not impossible, to determine from simple inspection of hysteresis loops. However, the sign of the coupling can be determined from model fitting (e.g., see Ref. 15).