

Parallel versus Antiparallel Interfacial Coupling in Exchange Biased Co/FeF₂

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By using the surface and element specificity of soft x-ray magnetic dichroism we provide direct experimental evidence for two different types of interfacial uncompensated Fe moments in exchange biased Co/FeF₂ bilayers. Some moments are pinned and coupled antiparallel to the ferromagnet (FM). They give rise to a positive exchange bias and vanish above $T_N = 78$ K together with the antiferromagnet (AF) order. Other interfacial Fe moments are unpinned and coupled parallel to the FM. They persist up to 300 K and give rise to magnetic order at the AF surface even above T_N .

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When a ferromagnet (FM) is grown on an antiferromagnet (AF), or *vice versa*, the magnetic anisotropy of the AF dictates the alignment of the magnetization in the FM through an effect called exchange anisotropy [1]. As a consequence, the FM exhibits an easy magnetization axis parallel to the anisotropy axis of the AF. This effect is referred to as *uniaxial* exchange anisotropy. When the magnetization of the FM prefers a specific direction we speak of a *unidirectional* exchange anisotropy or exchange bias. The preferred direction is typically introduced by cooling the sample from above the Néel temperature, T_N , in an applied field H_{CF} . As a result, the hysteresis loop exhibits a characteristic shift H_E along the field axis. In a wide variety of systems the loop shift is in the opposite direction of \vec{H}_{CF} , and this so-called “negative” exchange bias effect is used today in many magnetic devices [2,3].

In this work we address the origin of two phenomena related to AF-FM exchange anisotropy that were originally revealed by studies that used transition metal fluorides as the AF. Both effects are assumed to originate from a characteristic and prototypical magnetic arrangement at such an interface. For example, a distinct exchange induced uniaxial exchange anisotropy well above T_N has been reported [4–9]. This anisotropy manifests itself as an increase in coercivity of the FM layer parallel to the AF spin axis that persists well above the Néel temperature $T_N = 78$ K of FeF₂, up to 300 K in the case of Co/FeF₂(110). Grimsditch *et al.* have suggested that this effect may be the result of induced local magnetic order at the AF surface caused by the exchange interaction between the FM and uncompensated AF spins [9].

Another fascinating feature of these AF-FM exchange-coupled systems is the occurrence of so-called “positive” exchange bias in large cooling fields, first reported for Fe/FeF₂ [10]. The term positive refers to the fact that the FM hysteresis loop is shifted in the same direction as \vec{H}_{CF} . The origin of positive exchange bias is thought to be due to *antiparallel* coupling between the FM magnetization and interfacial moments in the AF [10].

Direct experimental verification of theoretical models developed to explain either positive exchange bias or the exchange induced anisotropy requires techniques that can detect the small magnetic moments arising exclusively from the buried interface. For this reason we have employed element-specific and depth-sensitive soft x-ray absorption measurements based on electron yield detection which provide magnetic sensitivity through linear (XMLD) and circular (XMCD) dichroism effects. We recently demonstrated that such an approach provides the necessary depth and submonolayer sensitivity to characterize the interface of an AF-FM bilayer [11,12].

Our samples, experimental method, and results differ from those recently reported by Roy *et al.* [13]. In the samples investigated by Roy *et al.* by means of soft x-ray and polarized neutron scattering, the exchange energy at the AF-FM interface was much larger than in the samples investigated by us [14]. This causes a significantly different distribution of moments in the AF during reversal of the FM because a domain wall in the AF may be wound up as suggested by Mauri *et al.* [15] and confirmed by Scholl *et al.* [16]. In the samples studied by Roy *et al.* [13], the formation of a domain wall consisting of twisted moments [16] is favored, while in our samples the weaker interfacial exchange energy resulted in a negligible domain wall contribution. Hence, we can assume that rotatable moments in our samples are not due to domain wall formation and the observed pinned AF moments are located right at the interface.

For our experiments a 2.5 nm Co/68 nm FeF₂(110) bilayer and a 68 nm FeF₂ single layer were grown at West Virginia University via molecular beam epitaxy on MgF₂(110), so that the FeF₂ layer was untwinned and epitaxial and the Co film was polycrystalline, as described elsewhere [7,17], and similar to the samples used in Ref. [9]. The FeF₂ layers for both samples were grown simultaneously. While the Co layer was grown on one sample the other one was removed from the holder without exposing it to air. Subsequently the bare FeF₂ sample was

placed back on the sample holder and both samples were capped with a 2 nm Pd layer to prevent the Co surface from oxidation during transport and the FeF_2 surface from x-ray damage during measurement [18]. $\text{Fe } L_2$ edge x-ray absorption spectra (XAS) obtained for each sample at room temperature (RT) were identical as shown in the top panel of Fig. 1, indicating that the $\text{FeF}_2(110)$ surface was not stoichiometrically changed by the proximity of the metallic Co or Pd layers. Furthermore, both samples exhibited an identical polarization dependence that is usually referred to as *charge dichroism* and is caused by the noncubic charge arrangement around the Fe atom, as shown in the inset of Fig. 1. We can therefore conclude that the atomic arrangement also was identical for the bare and exchange-coupled AF surfaces.

The x-ray dichroism experiments were performed using the elliptical undulator beam line 4.0.2. at the Advanced Light Source (ALS) in Berkeley which provides soft x rays in the energy range between 250–2000 eV with variable x-ray polarization. The x-ray absorption spectra were acquired by recording the sample current as a function of x-ray energy. For the XMCD measurements we used x rays that were incident at an angle of 30° from the surface, as in previous experiments [12]. The XMLD spectra were obtained at normal x-ray incidence with the linear polariza-

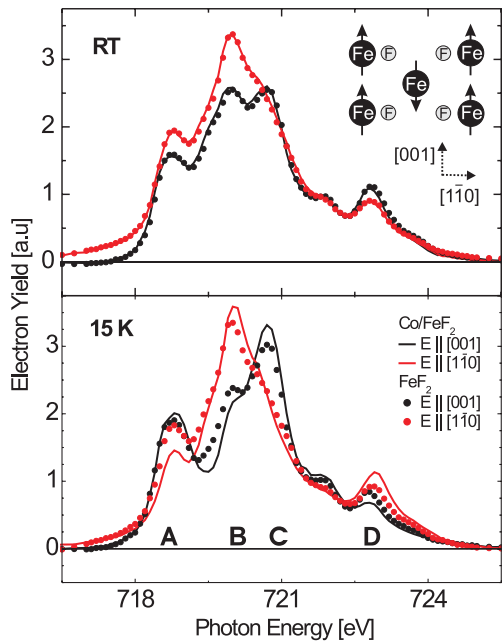


FIG. 1 (color). $\text{Fe } L_2$ edge XAS of $\text{FeF}_2(110)$ obtained at room temperature (top) and $T = 15$ K (bottom). The incoming linear polarization was aligned either parallel (black) or perpendicular (red) to the in-plane c axis ($[001]$). Spectra obtained from the bare FeF_2 (Co/ FeF_2) sample are represented by dots (lines). The inset in the upper panel shows a sketch of the tetragonal arrangement of Fe^{+2} and F^- ions at the (110) surface. F^- ions located below and above the center Fe^{+2} ion are not shown. The arrows indicate the direction of the spin moment of the Fe^{+2} ions.

tion (\vec{E} vector) aligned either parallel to the in-plane c axis ($[001]$) or $[1\bar{1}0]$ axis. The sample was mounted on a cryostat which could be cooled to 15 K. Additional field-dependent measurements were performed using the ALS vector magnetometer [20].

We first address the influence of AF-FM exchange coupling on the degree of AF order at the $\text{FeF}_2(110)$ surface. For this purpose we established the spectroscopic signature of Fe XMLD in FeF_2 and compared its magnitude for the two samples. In Fig. 1 we show $\text{Fe } L_2$ absorption spectra acquired from both samples above and below T_N . The most pronounced differences between spectra obtained at RT and 15 K are observed for the spectral features labeled A to D. For example, at RT the x-ray absorption intensity of peak A was smaller when \vec{E} was parallel to the c axis (black) than when \vec{E} was perpendicular to it (red). Below T_N , at 15 K, the situation was reversed and adjacent spectral features showed a similar behavior although with opposite sign. This dichroism could not have been caused by structural changes because the charge dichroism at the $\text{Fe } K$ edge (not shown) remained unchanged. We therefore conclude that the observed effect was caused by long-range antiferromagnetic order at the $\text{FeF}_2(110)$ surface. Although the XMLD was qualitatively the same for both samples, it was significantly more pronounced for the exchange-coupled sample (full lines) than for the bare surface (dotted lines). This constitutes clear experimental evidence that AF-FM exchange coupling in Co/ $\text{FeF}_2(110)$ leads to an increase in AF surface order relative to the bare FeF_2 surface, as suggested in Ref. [9].

To investigate the net magnetic moment that leads to the nonvanishing exchange coupling we acquired XMCD spectra of Fe and Co at 15 K. The x-ray absorption spectrum for Co/ $\text{FeF}_2(110)$ is shown in the top panel of Fig. 2, while the XMCD difference between spectra acquired with opposite helicities in a magnetic field of 2.8 kOe is shown in the bottom panel. The spectra are scaled to emphasize the Fe and F peaks which are much weaker than the signal from the Co metal layer on top. At the $\text{Fe } L$ edge, a clear

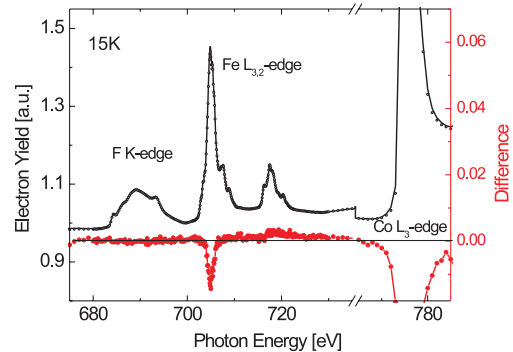


FIG. 2 (color). X-ray absorption spectrum of Co/ $\text{FeF}_2(110)$ across the Fe and Co L edges (black) and x-ray magnetic circular dichroism (XMCD) difference spectra shown in red underneath.

XMCD effect with opposite sign at the L_3 and L_2 resonances was observed. The sign of the XMCD effect at the L_3 resonance is the same for Fe and Co. This demonstrates that a net magnetic moment indeed exists in “antiferromagnetic” FeF_2 at the FeF_2/Co interface and that this moment is parallel to that of the Co. This moment does not originate from interdiffusion or loss of fluorination because the energy positions of the peaks in the Fe XMCD spectrum in Fig. 2 coincide with the maxima in the Fe absorption resonances of FeF_2 and are shifted ~ 1 eV higher with respect to the known peak positions for Fe metal. Hence, the XMCD signal is clearly due to FeF_2 and not to metallic Fe. Using a similar approach to the one used in Ref. [12], we estimate the thickness of the interface ferromagnetic layer to be approximately 1 monolayer. Since no XMCD signal was detected for the pure FeF_2 surface, the existence of the ferromagnetic Fe spins must be due to the interfacial interaction between FeF_2 and Co.

To address the role of the interfacial moment in exchange bias we acquired element-specific hysteresis loops of the uncompensated Fe moments and the Co moments as previously described [12]. The sample was cooled down from RT to 15 K in a relatively small cooling field $H_{\text{CF}} = +200$ Oe parallel to the FeF_2 [001] axis. The loops are shown in Fig. 3. The Co loop (black) exhibits a negative horizontal bias shift of $H_E = -265$ Oe because the Co layer favors a *positive* magnetization (parallel to \vec{H}_{CF}). It is slightly sheared, perhaps as a result of the 30° measurement geometry or the presence of Pd, which would tend to give the Co layer a slight perpendicular anisotropy. Overall the reversal of the ferromagnetic Co layer is asymmetric, indicating that the exchange coupling energy across the interface is small compared to the energy of an AF domain wall perpendicular to the interface [15]. We therefore assume that no domain wall is wound up in the AF and that the AF bulk spin lattice is not affected by the reversal

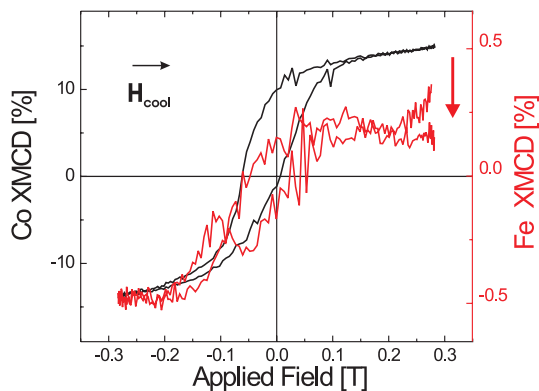


FIG. 3 (color). Element-specific Co (black) and Fe (red) hysteresis loops acquired at $T = 15$ K after field cooling in $+200$ Oe along the FeF_2 [001] axis, parallel to the AF spin axis. The direction of the cooling field and the vertical shift of the Fe loop at $T = 15$ K is indicated by arrows.

of the FM. The AF-FM exchange anisotropy is hence dominated by the uncompensated interface magnetization only. The Fe loop (red) partly follows the Co loop, but a vertical shift for positive field, indicated by an arrow, shows that some Fe moments remain pinned in the negative magnetization direction and do not rotate in an applied field. This means that a small fraction of interfacial Fe atoms are pinned in the direction antiparallel to \vec{H}_{CF} for a “small” cooling field [21]. This important result was corroborated by additional measurements using opposite alignment of the cooling field and the x-ray propagation direction, as discussed in Ref. [12]. We thus find direct proof of the preferred *antiparallel* coupling between the FM Co and the uncompensated *pinned* Fe moments at the AF-FM interface. We note that frozen ferromagnetic moments have been previously observed in the bulk of $\text{Fe}_x\text{Zn}_{1-x}\text{F}_2$ and FeF_2 single crystals when cooled in a large field and are believed to result from strain within the AF [22,23]. In our case, however, pinned moments are aligned during field cooling by the exchange interaction which acts as an effective field. They are likely confined to the interface itself, which explains why, to our knowledge, they have not been detected via direct magnetometry measurements neither in films nor in single crystals.

Our observation of antiparallel coupling between pinned interface moments and the FM can explain the occurrence of both negative and positive H_E as suggested by Nogués *et al.* [10]. For small cooling fields the FM is aligned along \vec{H}_{CF} yet the pinning direction in the AF is determined by the antiparallel exchange coupling with the ferromagnet. This leads to Fe moments pinned antiparallel to \vec{H}_{CF} as seen in Fig. 3. In “large” cooling fields the Zeeman energy dominates and the moment directions in both the FM and AF are aligned parallel to \vec{H}_{CF} . When the cooling field is turned off, the moments in the AF are frozen parallel to \vec{H}_{CF} but the favored antiparallel exchange leads to a rotation of the FM moments. The direction of the FM is now *antiparallel* to \vec{H}_{CF} , which corresponds to positive exchange bias. Indeed, when the sample was field cooled in $+2.8$ kOe, it did exhibit a small positive bias (~ 50 Oe). However, in this case the expected tiny vertical Fe loop shift of $+0.03\%$ was too small to be detected.

Our interpretation is supported by temperature dependent measurements shown in Fig. 4. The Fe XMCD intensities and the vertical loop shift were extracted from Fe hysteresis loops, and H_C and H_E were determined from Co hysteresis loops. The XMLD intensity was defined as the intensity ratio between peaks C and B in Fig. 1 with $\mathbf{E} \parallel [001]$. Clearly H_E (\square) is directly related to the vertical loop shift (∇) and hence to the existence of *antiparallel* coupled interfacial pinned moments. The two quantities disappear above T_N together with the XMLD effect (\star). Since the latter reflects the existence of AF order, this shows that the exchange bias effect is due to the anchoring of the pinned spins in the antiferromagnetic lattice.

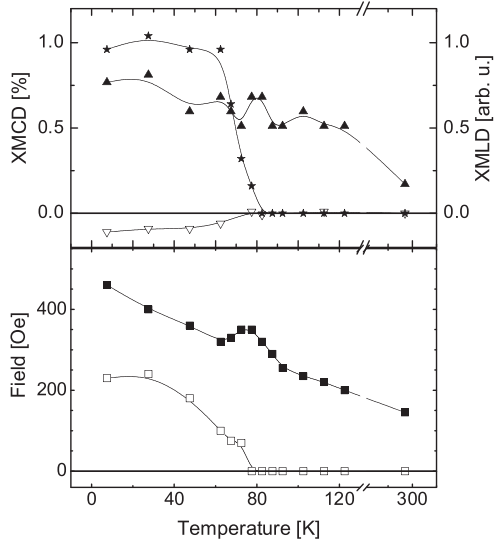


FIG. 4. Top: temperature dependence of the Fe XMCD (\blacktriangle , reflecting uncompensated Fe moments), Fe XMLD (\star , reflecting long-range AF order), and vertical Fe loop shift (∇ , reflecting pinned moments). Bottom: corresponding absolute values of the horizontal loop shift $|H_E|$ (\square) and coercivity H_C (\blacksquare), determined from element-specific Co hysteresis loops. Solid curves are guides to the eye.

In contrast, the unpinned Fe moments that are aligned parallel to the magnetization of the FM (\blacktriangledown) do not disappear above T_N . They follow the magnetization of the FM and we therefore conclude that they exist only in close proximity to the interface and are coupled more strongly to the FM than the pinned moments. Their magnitude decreases monotonically with increasing temperature and is not affected by the AF phase transition. Together with the temperature dependence of the *anisotropy* of the FM layer in Co/FeF₂ reported by Grimsditch *et al.* [9], our results show that magnetic order not only exists at the AF surface above T_N but that the preferred Fe spin direction is still governed by the FeF₂ lattice, not by Co. Hence we have the interesting situation that the exchange interaction (alignment) between the Fe moments at the FeF₂ surface is induced by Co yet the anisotropy direction is determined by the FeF₂ lattice, just as in the AF phase below T_N .

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