Induced anisotropy and positive exchange bias: A temperature, angular, and cooling field study by ferromagnetic resonance

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(Received 11 June 2001; published 15 January 2002)

Exchange-biased MnF_2/Fe bilayers, examined by variable angle and temperature ferromagnetic resonance (FMR), exhibit a sudden onset of a unidirectional and fourfold anisotropy below the MnF_2 Néel temperature. This unexpected fourfold symmetry arises from frustrated perpendicular coupling between the MnF_2 and the Fe overlayer in the presence of twinning in the antiferromagnet layer. These data are consistent with earlier polarized-neutron-reflectometry results. The FMR data show a clear reversal in the direction of the unidirectional anisotropy as a function of cooling field, switching sign at $H_{FC}=13$ kOe, which is consistent with the onset of positive exchange bias observed in conventional magnetometry experiments. The low-temperature FMR linewidth reflects the in-plane symmetry of the resonance itself, exhibiting surprising divergence in the hard directions. Temperature-dependent FMR measurements reveal a sharp reduction in the resonance field below the Néel point due to the ferromagnetic/antiferromagnetic coupling.

DOI: 10.1103/PhysRevB.65.064410 PACS number(s): 75.70.Cn, 75.30.Gw

The problem of exchange-induced anisotropy at the interface between an antiferromagnet (AF) and a ferromagnet (F) is an old, and essentially unsolved one.[1] Some of the central issues that remain to be resolved include the fundamental origin of exchange bias,[1–5] the spin dynamics[6,7] and structure in the AF[2–4] and F[8–10] constituents; the effects of microstructure and disorder;[11–13] measurement-technique-related discrepancies[14,15]; coercivity effects and the relation between coercivity and exchange bias[5,16,17]; and the magnetization-reversal mechanisms.[18] It is certainly clear at this point that if one hopes to understand exchange bias in a given system then it is necessary to understand fully the microstructure and behavior of the antiferromagnet. The explanation for the exchange bias in the polycrystalline system CoO/NiFe in terms of an uncompensated free spin density is a perfect example of this.[11] To this end, we have undertaken a comprehensive study of the exchange bias in transition-metal difluoride/Fe bilayers. This is a relatively simple AF system that has been very well studied. In particular, we have investigated the effects of microstructural disorder,[19] positive exchange bias,[20,21] magnetization-reversal asymmetry,[22] temperature dependence,[23] spin flop,[24] and coercivity[25] effects. Specifically we have seen that many of the effects are related to the twinned nature of the samples, highlighting once more the need to understand microstructure.

In this paper we probe, with ferromagnetic resonance (FMR), the F/AF exchange interaction in a Fe/MnF_2 bilayer, wherein the MnF_2 has a (110) epitaxy, but is orthogonally twinned. Neutron measurements[22] indicate that perpendicular coupling between the Fe and the AF-ordered MnF_2, combined with the frustration arising from the twinned AF domains produces an Fe easy axis at 45° from a MnF_2 easy direction. Superconducting quantum interference device (SQUID) magnetometry[21] reveals a crossover from negative to positive exchange bias as a function of cooling field. FMR measures directly, as a function of in-plane angle, the internal fields present in the system and as a result produces a complete characterization of the anisotropy topology. In addition, the FMR linewidth characterizes system magnetodynamics. While others have used FMR to examine various aspects of exchange bias,[7,26,27,29,30] this, to our knowledge, is the first investigation to map the anisotropy topology and magnetodynamics as a function of temperature on a structurally well defined, epitaxial, exchange bias system.

The sample is a thin film of structure (100)MgO/substrate/ZnF_2(110)/MnF_2(110)/Fe(polycrystalline)/Al, where the ZnF_2 is a buffer layer and the Al acts as a cap to prevent oxidation. Thicknesses are nominally 25, 60, 12, and 5 nm, respectively. The samples were deposited by high-vacuum electron-beam evaporation. Details of growth and characterization by high-angle x-ray diffraction, grazing-incidence x-ray reflectivity, reflection high-energy electron diffraction and SQUID magnetometry is provided elsewhere.[21] In summary, the fluorides are perpendicularly twinned quasiepitaxial layers with a (110) orientation (a compensated surface with the spins in the interfacial plane) while the Fe overlayers are essentially polycrystalline. The interface roughness (σ) can be tuned by varying the substrate temperature during deposition of the MnF_2 layers.[21] Previous studies have shown that smooth interfaces ($\sigma_{RMS}<1.2$ nm) exhibit AF coupling between the F/AF layers, positive exchange bias for large cooling fields[21] and enhanced coercivity effects,[25] whereas rougher interfaces ($\sigma_{RMS}>1.2$ nm) exhibit ferromagnetic coupling between the F/AF layers and show only negative exchange bias. The data presented in this paper are on a sample with smooth interfaces allowing us to access the positive exchange bias regime at moderate cooling fields of the order of 10 kOe.

FMR measurements were made with the sample lying film side down on the bottom of a homemade, TE 104 mode, rectangular cavity (loaded Q of approximately 1500) operating at 35 GHz, with the field applied in the plane of the

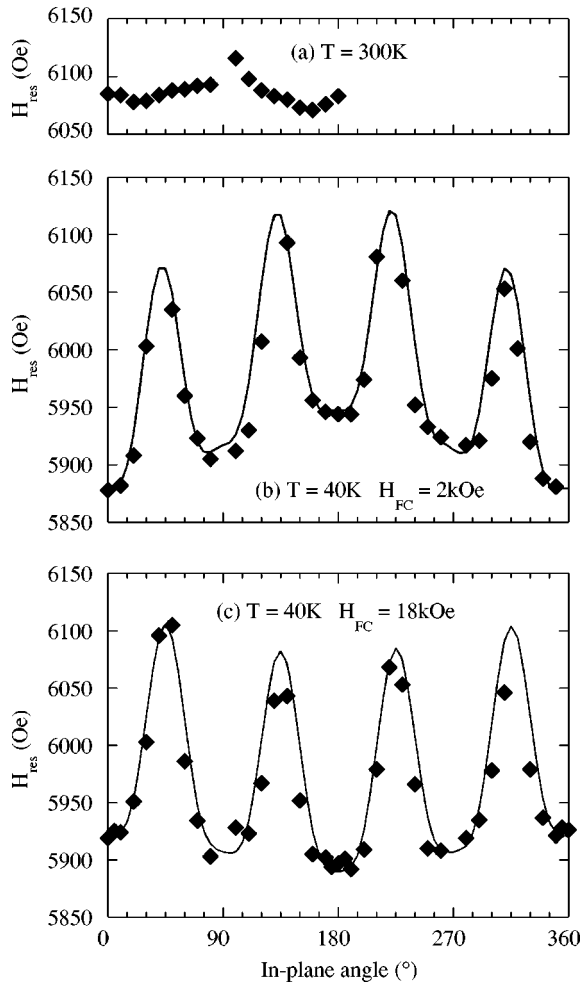

FIG. 1. Resonance field as a function of in-plane angle (a) $T=293$ K; (b) $T=40$ K and $H_{FC}=2$ kOe; (c) $T=40$ K and $H_{FC}=18$ kOe. Symbols are data points and lines are model calculations. The cooling field is applied at 0°, which is along the MgO(100), a direction bisecting the perpendicular (110) flouride twins.

bilayer film. At this frequency, the resonance fields are high enough to ensure magnetization saturation along any in-plane field direction. The cavity is situated inside an Oxford Instruments continuous-flow cryostat. The cavity/cryostat assembly is placed between the poles of a variable-gap, 10 inch electromagnet. Except for those at room temperature, angular measurements were taken at 40 K after cooling through the AF's T_N (approximately 67 K) in a field (H_{FC}) set at 150 K. The cooling field is applied along the MgO (100), a direction bisecting the perpendicular (110) fluoride twins mentioned above. A field of 6 kOe was set prior to reducing the temperature in each of the variable temperature measurements.

FMR spectra, at all temperatures and cooling fields, are characterized by a single, well-defined peak. In-plane angular measurements taken at room temperature [Fig. 1(a)] showed only a weak, probably growth induced, twofold anisotropy, confirming the polycrystalline nature of the Fe layer.

As mentioned above, 40 K FMR spectra were obtained as a function of in-plane angle at cooling fields ranging from 2 to 18 kOe. The results at these two extremes are depicted in Figs. 1(b) and 1(c). In each of the two extremes, one observes the superposition of fourfold and unidirectional symmetries, with the fourfold easy axis at approximately 45° from the weak twofold easy axis mentioned above. The anisotropy data are analyzed using the equation of motion

$$\frac{1}{\gamma}\frac{\partial \mathbf{M}}{\partial t}=\mathbf{M}\times\mathbf{H}_{\text{eff}}, \tag{1}$$

where M is the Fe magnetization, γ is the Fe gyromagnetic ratio, and the effective field arises from the magnetization gradient of the energy density E,

$$\mathbf{H}_{\text{eff}}=-\boldsymbol{\nabla}_M E(\theta,\phi), \tag{2}$$

and θ and ϕ are the angles the magnetization makes with the film normal and an in-plane easy direction, respectively. Energies pertinent to this system are Zeeman, out-of-plane anisotropy (usually dominated by thin-film-shape effects), and the fourfold and unidirectional anisotropies mentioned above. The angular dependence of these energies can be expressed as follows:

$$\begin{aligned} E_{\text{Zeeman}}&=-MH\sin\theta,\\ E_{\text{out}}&=-K_{\text{out}}\sin^2\theta,\\ E_{\text{4fold}}&=-[K_{\text{four}}\sin^2\theta\cos^2(2\phi)+K_{\text{four}}\sin^4\theta\cos^4(2\phi)],\\ E_{\text{ud}}&=-K_{\text{ud}}\sin\theta\cos(\phi), \end{aligned} \tag{3}$$

where the K's are coefficients to be extracted via model fitting to the data. Higher-order terms are required in the fourfold energy due to the obvious asymmetry in the peaks and valleys of the data. The above equations presume the magnetization is precessing about the applied field direction at resonance (approximately 6 kOe)—a condition verified in magnetization measurements.

MATHEMATICA™ is used to perform the energy derivatives and to provide an expression for the resonance field as a function of the in-plane angle. The energy coefficients are then adjusted to provide a best ''eyeball'' fit of the model to the data, the results of which are shown as solid lines in Fig. 1.[28] The resultant out-of-plane anisotropy ($K_{\text{out}}=1.57\times10^7$ erg/cm^3) is due, as expected, to demagnetization. Given the polycrystalline nature of the Fe layer, the appearance of a fourfold term is surprising. In addition, the values obtained ($K_{\text{four},1}=3\times10^4$ erg/cm^3) and $K_{\text{four},2}=-0.65\times10^4$ erg/cm^3 are quite small in comparison to bulk Fe values (47×10^4 erg/cm^3). A clue as to the source of the fourfold energy can be found in its easy axis, which is midway between the (110) easy axis of one of the MnF_2 twins and the (110) axis of its perpendicular counterpart. Each of the MnF_2 (110) twins has uniaxial in-plane anisotropy, so the uniform Fe overlayer sees an average of the inhomogeneous surface anisotropy, wherein the uniaxial terms cancel, leaving a residual second-order fourfold anisotropy. This fourfold term has its easy direction midway between the (110) directions of perpendicular MnF_2 twins. In addition, as seen in Fig. 2, the fourfold term appears suddenly at the MnF_2 Néel tempera-

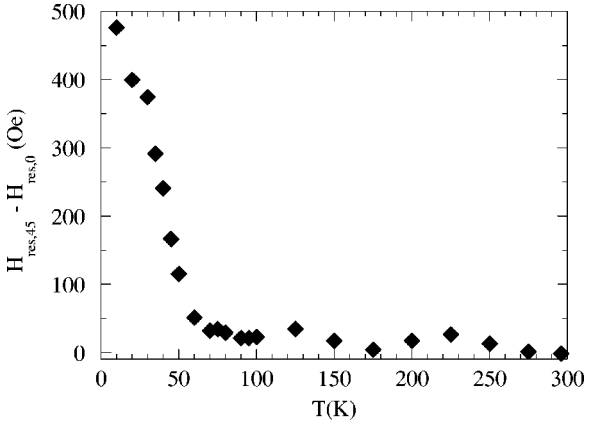

FIG. 2. Difference between resonance field at $\phi = 45°$ ($H_{res,45}$) and $\phi = 0°$ ($H_{res,0}$) as a function of temperature. This difference is proportional to the four-fold anisotropy energy density, K_1.

ture, increasing strongly with decreasing temperature. Therefore, one can conclude that the twinning in the AF is leading to fourfold anisotropy in the Fe layer due to the strong coupling between the layers at the interface. While others[29,30] observe the expected onset of unidirectional anisotropy, the dramatic appearance of the fourfold term below T_N is somewhat unexpected manifestation of the exchange interaction at the F/AF interface.

The low-temperature data in Fig. 1 also contains an obvious unidirectional component that arises from the orientation of the cooling field that freezes in the exchange-induced coupling. The change, with cooling field, in sign of the unidirectional exchange bias is readily observed in Figs. 1(b) and 1(c). In-plane measurements, as per Figs. 1(b) and 1(c), were made at a range of cooling fields and the exchange field ($H_{ex} = K_{ud}/M_{Fe}$) obtained is shown as a function of cooling field in Fig. 3 (the values of K_{out}, K_1 and K_2 are independent of cooling field). Here one sees, in detail, a crossover from negative to positive exchange bias in a cooling field of approximately 13 kOe, which is consistent with SQUID-based observations[21] on the same samples. This crossover has been explained[20] in terms of a competition between antiferromagnetic AF/FM interfacial exchange and Zeeman energies that

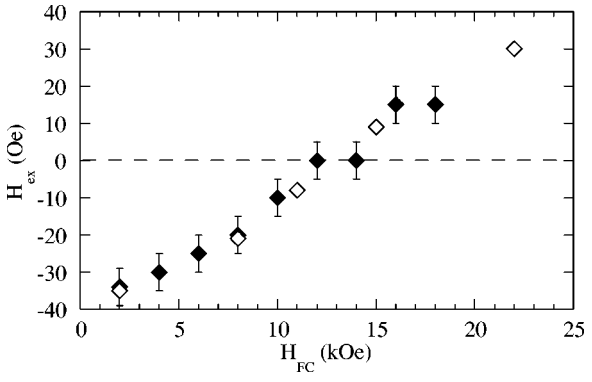

FIG. 3. Exchange bias as a function of cooling field. Solid diamonds are from the present FMR measurements and open squares are from SQUID measurements. The FMR measurement temperature is 40 K, whereas that of the SQUID data is 10 K.

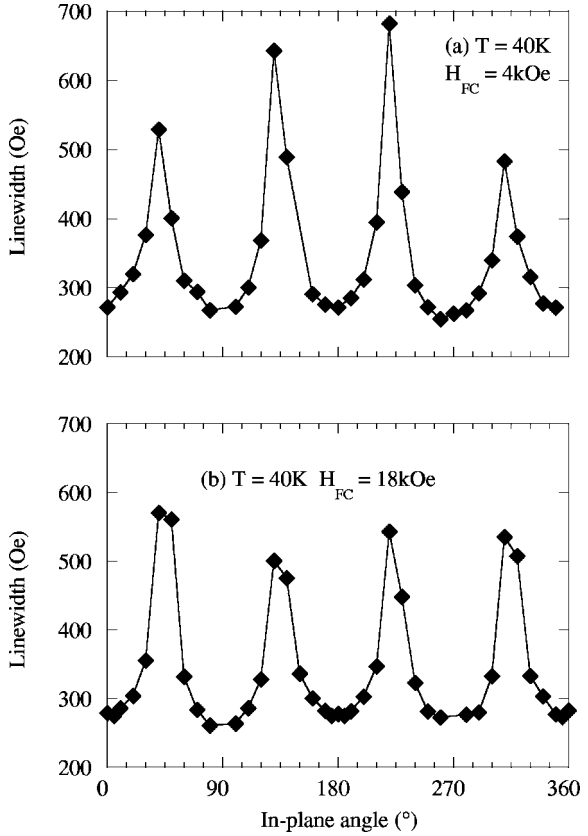

FIG. 4. FMR line-width as a function of in-plane angle at 40 K for cooling fields of (a) 4 kOe and (b) 18 kOe. The solid lines are a guide to the eye. The cooling field is applied at 0°, which is along the MgO (100), a direction bisecting the perpendicular (110) fluoride twins.

results, at high cooling fields, in parallel alignment of the FM and interfacial AF layers. In that case, the exchange energy of the Fe layer is higher when oriented in the field cooling direction than it is when oriented opposite that direction [see Fig. 1(c)]. Agreement between FMR (where the magnetization is saturated at all angles) and SQUID techniques indicates that the mechanism responsible for the size of the exchange bias is not influenced by irreversible phenomena, likely due to the epitaxial nature of the interface.[15] This is not to say that the reversal mechanism itself is not rich in detail,[31,32] but that there is likely an absence of training effects in this system. It should be noted that changing the cooling-field direction alters the position of the unidirectional peak, but does not affect the position of the fourfold peaks and that the fourfold amplitude does not depend upon size of the cooling field. These observations suggest that the fourfold term depends more upon the MnF_2 structure than it does upon the AF sublattice magnetization direction, similar to what was concluded in Fe on epitaxial FeF_2 from magnetization data.[33]

Interestingly, the in-plane variation of the FMR linewidth (Fig. 4) reflects the symmetry of the resonance itself, where the linewidth broadens in the hard directions and unlike other epitaxial systems[34] peaks sharply there. In addition, one notices the apparent influence of the exchange bias in the line-

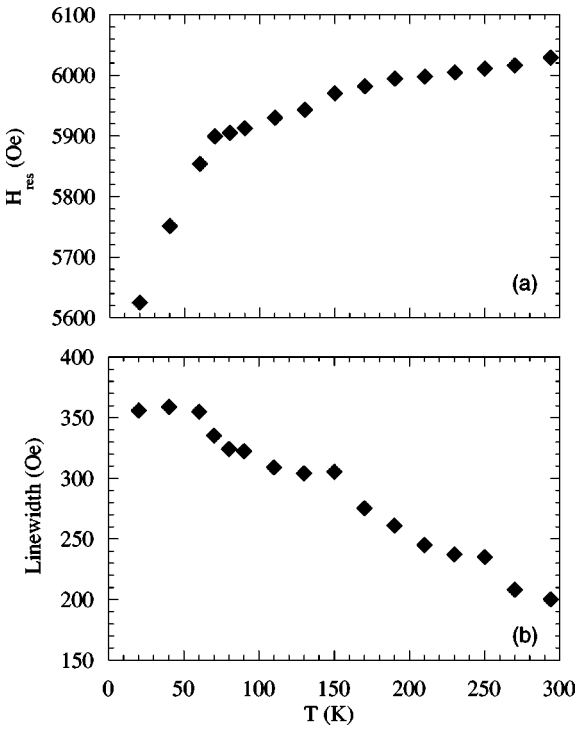

FIG. 5. FMR (a) resonance field and (b) linewidth, along the easy direction, as a function of temperature.

width peak heights. A qualitative interpretation of the fourfold linewidth anisotropy lies in the twinned nature of the MnF_2. When oriented along the easy direction, the Fe magnetization sees each MnF_2(110) twin in the same orientation, i.e., 45° from the (110). The homogeneity of this environment results in a narrow FMR linewidth. On the other hand, when oriented along the hard direction, the Fe is aligned parallel to the (110) of one MnF_2 twin and perpendicular to the (110) of the other, the resulting inhomogeneity giving rise to a broadened linewidth. While qualitatively satisfying, the above explanation accounts for neither the influence of the exchange bias, nor the sharp peaks in the hard directions. Another source of the linewidth broadening may lie in the *F*/AF coupling producing a spring-magnet-type spin inhomogeneity[8,9] in the Fe layer. The spring arises from the Fe at the AF interface being constrained along the easy direction, but Fe monolayers removed from the interface orient progressively toward the applied field. The resulting twist, and, therefore, the inhomogeneity, would be greatest along the hard direction. This interpretation would also account for the unidirectional feature in the size of the linewidth. At low (high) cooling fields, the negative (positive) exchange bias produces a higher (lower) energy configuration at 180°, thereby producing an increased (decreased) twist in the Fe layer and a concomitant increase (decrease) in the size of the linewidth. One might argue that the sharpness is a result of the interfacial Fe spontaneously switching from one easy direction to a perpendicular one as the field passes through the hard direction. The problem with this argument is that the interface coupling is very much less than the Fe-Fe exchange and, therefore, the amount of twist in the Fe layer would be quite small. Another possible source of the linewidth divergence is described by Arias and Mills.[35] Utilizing two magnon scattering processes in a system with a structurally asymmetric interface, they predict divergent behavior in the linewidth along certain symmetry directions. However, in their case their interface symmetry is broken by one-dimensional steps, a situation not present in our system. The source of this unusual linewidth behavior remains an open question.

Temperature variations of resonance field and linewidth along an easy direction are shown in Fig. 5. The resonance field exhibits a marked downturn at the MnF_2 Néel temperature, which results directly from the onset of the in-plane fourfold anisotropy at that same temperature. This can be seen in Fig. 1, where the, essentially isotropic, resonance field at 300 K becomes the hard direction field value at 40 K. The linewidth monotonically increases with decreasing temperature, reaching a constant value below the MnF_2 Néel temperature. This variation likely arises from short-lived AF fluctuations in the AF layer induced at the interface by the Fe layer. The temporal stability of the induced order increases with decreasing temperature, until at the Néel temperature the full AF structure is in place.

In summary, in-plane FMR measurements of Fe on MnF_2 reveal anisotropies dominated by the interfacial coupling between the Fe and the AF structure in the fluoride. Owing to the smooth interface, the unidirectional exchange anisotropy switches from negative to positive with increasing cooling field. A fourfold term in the anisotropy is shown to have its origin in the twinned structure of the MnF_2 surface. The origin of the sharply peaked, azimuthal variations in the linewidth remains an open question. Thermal variations in the FMR parameters are also dominated by the interfacial coupling of the Fe with the AF fluoride.

We gratefully acknowledge discussions with Eric Fullerton. This work was supported by the U.S. Department of Energy both at UCSD and at Miami and NSF at UCSD, J.N. acknowledges partial support from DGR (1999SDR00340).

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