

## Perpendicular coupling at Fe–FeF<sub>2</sub> interfaces

T. J. Moran,<sup>a)</sup> J. Nogués,<sup>b)</sup> D. Lederman,<sup>c)</sup> and Ivan K. Schuller  
*Physics Department 0319, University of California San Diego, La Jolla, California 92093-0319*

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We have studied the exchange anisotropy of ferromagnetic Fe films grown on antiferromagnetic FeF<sub>2</sub> single crystals. The behavior of the hysteresis loops of the Fe above and below the Néel temperature  $T_N$  of FeF<sub>2</sub> indicates a 90° rotation of the ferromagnetic easy axis due to the antiferromagnetic ordering. By examining the Fe hysteresis loops together with the FeF<sub>2</sub> susceptibility behavior we infer that below  $T_N$  the ferromagnetic and antiferromagnetic spins are coupled perpendicular to each other. This behavior can be explained by recent micromagnetic calculations on exchange bias systems, or by magnetoelastic effects. © 1998 American Institute of Physics. [S0003-6951(98)02405-X]

Exchange bias refers to the shift of a ferromagnetic hysteresis loop away from  $H=0$  due to the interaction between two magnetic materials.<sup>1</sup> Typically this happens when a ferromagnet (FM) and a neighboring antiferromagnet (AFM) are field cooled below the Néel temperature ( $T_N$ ) of the AFM. In recent years there has been a renewed interest in exchange bias especially in thin film form,<sup>2</sup> motivated by possible applications in magnetoresistive devices.<sup>3</sup> However, the microscopic mechanism responsible for this phenomenon remains unclear. To study the more fundamental aspects of exchange bias it is desirable to control the structure of the ferromagnet–antiferromagnet interface. For this reason some exchange bias studies have been carried out using single crystal antiferromagnets.<sup>4–7</sup>

In this study FeF<sub>2</sub> was chosen for the antiferromagnet because it has a simple crystal structure (body centered tetragonal),<sup>8</sup> simple spin structure,<sup>9</sup> and very strong uniaxial anisotropy for the AFM spins.<sup>10</sup> Assuming that the bulk magnetic structure is preserved, the surface magnetic structure can be changed by varying the crystalline orientation. For instance, the FeF<sub>2</sub>(110) surface has equal numbers of spins from the two antiparallel sublattices, and is therefore called compensated. The FeF<sub>2</sub>(100) surface is uncompensated since in that case a surface plane contains spins that point in a single direction. The FeF<sub>2</sub>(110) and FeF<sub>2</sub>(100) surfaces are similar in that the FeF<sub>2</sub>[001] direction, which is the ordered spin direction, lies in the plane of both. When Fe thin films are deposited on FeF<sub>2</sub> substrates with either of these orientations, we find a 90° rotation of the Fe easy axis below  $T_N$ , which is driven by a perpendicular coupling between the FM and AFM layers.

The FeF<sub>2</sub> single crystal was grown using the Bridgeman–Stockbarger method, aligned using a Laue x-ray camera, and cut with a diamond wire into wafers with two different orientations—(100) and (110). The crystals were polished and then loaded into a Riber ultrahigh vacuum mo-

lecular beam epitaxy (MBE) system ( $2 \times 10^{-10}$  Torr base pressure). To improve surface quality, the crystals were annealed in vacuum at 400 °C for 30 min. 20 nm of Fe was deposited onto the FeF<sub>2</sub> single crystals at  $T_{\text{deposition}} = 150$  °C using electron beam evaporation at a rate of 0.1 nm/s. To protect the Fe layer, a 20 nm capping layer of silver was deposited at a rate of 0.05 nm/s at  $T_{\text{deposition}} = 150$  °C using an effusion cell. During deposition the pressure was below  $5 \times 10^{-9}$  Torr. Deposition rates were controlled using electron impact emission spectroscopy.

The structure of the films was studied *in situ* by reflection high energy electron diffraction (RHEED) and *ex situ* by x-ray diffraction. The polished FeF<sub>2</sub> surfaces showed bright two-dimensional diffraction patterns that became sharper after annealing, implying highly ordered atomic arrangements. The Fe films deposited possessed a large fraction of the Fe(110) planes parallel to the surface, and were preferentially oriented in the plane of the film. This was deduced from the observation of spotty RHEED patterns, as well as from x-ray diffraction results. X-ray diffraction measurements from a Fe(110)/FeF<sub>2</sub>(100) sample were taken with the scattering vector at a 27° angle from the growth direction in order to detect the Fe(310) peaks and FeF<sub>2</sub>(510) peaks. By comparing the azimuthal variation in the intensity of these peaks, we determined that the Fe[001] direction was primarily parallel to the FeF<sub>2</sub>[001] direction. The x-ray diffraction rocking curve widths for the Fe(110) planes parallel to the surface were typically 4°.

The magnetic characterization was carried out using a semiconducting quantum interference device (SQUID) magnetometer. The samples were cooled from 300 to 10 K in a magnetic field of 2000 Oe applied parallel to the plane of the film. Afterward hysteresis loops were measured for several temperatures as the sample was warmed back to 300 K. Varying the cooling field up to 70 000 Oe had no effect on any of the results. The hysteresis loops had a large linear background due to the susceptibility of the FeF<sub>2</sub> crystals. This linear signal was subtracted from the data in order to observe clearly the Fe behavior. A temperature dependent vertical offset in the magnetization, which disappeared above 80 K, was also observed (about 0.002 emu per gram of FeF<sub>2</sub>). Because this behavior coincided with the  $T_N$  of FeF<sub>2</sub>,

<sup>a)</sup>Present address: IBM (E44/015), 5600 Cottle Rd., San Jose, CA 95193. Electronic mail: tmoran@us.ibm.com

<sup>b)</sup>On leave from the Grup d'Electromagnetisme, Universitat Autònoma de Barcelona, Spain.

<sup>c)</sup>Present address: Physics Dept., West Virginia University, Morgantown, WV 26506-6315.

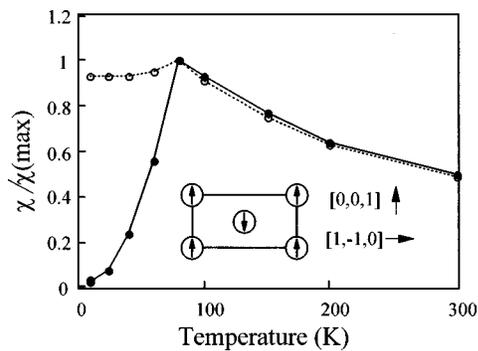


FIG. 1. Normalized susceptibility vs temperature for a  $\text{FeF}_2(110)$  crystal, when field is applied in two perpendicular directions: parallel to the  $\text{FeF}_2[001]$  direction (filled circles) and applied to the  $\text{FeF}_2[1,-1,0]$  direction (open circles). The inset shows the surface spin structure for the  $\text{FeF}_2(110)$  surfaces, assuming that the bulk structure is maintained. Ref. 13 contains a more complete diagram of the  $\text{FeF}_2$  structure.

this offset was attributed to piezomagnetism in the  $\text{FeF}_2$  and was also subtracted before analyzing the hysteresis loops.<sup>11</sup>

Figure 1 shows the susceptibility of a  $\text{FeF}_2$  single crystal when the applied field is either parallel or perpendicular to the AFM spin axis. The low temperature values approach zero for the parallel case and level off for the perpendicular case, as expected for single crystal antiferromagnets.<sup>12</sup>

The exchange bias of Fe films grown on  $\text{FeF}_2(100)$  crystals and  $\text{FeF}_2(110)$  is almost zero when the cooling field is applied in the plane and perpendicular to the AFM spin axis ( $H \parallel \text{FeF}_2[010]$  and  $H \parallel \text{FeF}_2[1,-1,0]$ , respectively). In other words, field cooling is ineffective when there is a large crystal anisotropy in the antiferromagnet which tends to align the spins perpendicular to the cooling field. The two interfaces behave differently when the cooling field is applied in the plane and parallel to the AFM spin axis;  $\text{FeF}_2(100)$  interfaces exhibit practically zero exchange bias while  $\text{FeF}_2(110)$  interfaces exhibit a "positive" exchange bias of about 80 Oe. Here positive exchange bias means that the hysteresis loop is shifted in the positive direction for positive cooling fields, which is opposite the direction observed in most exchange bias systems. Both positive and negative exchange bias conditions have been observed in Fe films coupled to  $\text{FeF}_2$  thin films.<sup>13,14</sup>

Now we will discuss the uniaxial anisotropy behavior, rather than the unidirectional anisotropy behavior (exchange bias). Figure 2(a) shows that the hysteresis loops taken at room temperature for a  $\text{Fe}/\text{FeF}_2(110)$  sample are different depending on the direction of the applied field. The curve taken parallel to the  $\text{FeF}_2[001]$  direction has large remanent magnetization and large coercivity compared to the curve taken with the field parallel to the  $\text{FeF}_2[1,-1,0]$  direction. We therefore conclude that the Fe has a uniaxial anisotropy with the easy axis parallel to the  $\text{FeF}_2[001]$  direction at 300 K. Based on x-ray diffraction and RHEED information, we attribute this easy axis to magnetocrystalline anisotropy, since the Fe crystallites have a preferred orientation in the plane of the film. Magnetoelastic anisotropy may also be present. Figure 2(b) shows the same curves taken at 10 K, where the situation has reversed. The large remanent magnetization and coercivity for the perpendicular case indicate that the Fe easy axis has rotated 90°. In other words, the Fe

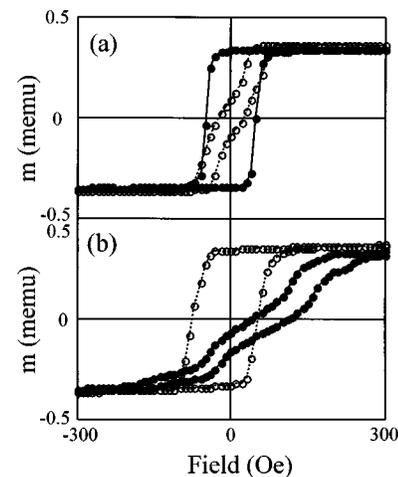


FIG. 2. Magnetization loops taken at (a) 300 K and (b) 10 K for a  $\text{Ag}(20 \text{ nm})/\text{Fe}(20 \text{ nm})/\text{FeF}_2(110)$  sample. Data were taken with the field applied in the plane parallel to the  $\text{FeF}_2[001]$  direction (filled circles) and  $\text{FeF}_2[1,-1,0]$  direction (open circles). In (b), the exchange bias for the filled circles is "positive" exchange bias, since the sample was cooled in a positive field of 2000 Oe.

spins have a low energy state when they are perpendicular to the ordered  $\text{FeF}_2$  spins. A similar rotation of the FM easy axis has also been observed in Fe films coupled to  $\text{FeF}_2(101)$  thin films.<sup>15</sup>

To investigate the origin of this rotation of the Fe easy axis we have measured hysteresis loops at increasing temperatures between 10 K and room temperature. Figure 3 shows the coercivity and the squareness, which is defined as the remanent magnetization divided by the saturation magnetization. The exchange bias shift was taken into account when determining the remanent magnetization values. One sees that the transition begins near the ordering temperature of the  $\text{FeF}_2$  (78.4 K). The same behavior is observed in  $\text{Fe}/\text{FeF}_2(100)$  samples.

The rotation of the Fe easy axis indicates that there is a perpendicular coupling between the Fe spins and the  $\text{FeF}_2$  spins, which is almost certainly influenced by the details of the atomic structure near the interface. These atomic struc-

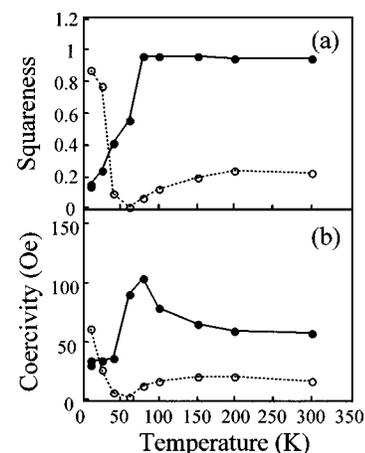


FIG. 3. Squareness (a) and coercivity (b) for an  $\text{Ag}(20 \text{ nm})/\text{Fe}(20 \text{ nm})/\text{FeF}_2(110)$  sample. Data taken with the field applied in the plane parallel to the  $\text{FeF}_2[001]$  direction (filled circles) and  $\text{FeF}_2[1,-1,0]$  direction (open circles). Here squareness = remanent magnetization/saturation magnetization.

ture details are difficult to obtain, although the temperature dependent behavior gives some information. A strong clue that the bulk atomic structure is maintained near the interface is the location of the transition points of the exchange bias and perpendicular coupling. Since both transitions occur near the bulk transition temperature of  $\text{FeF}_2$ , it is likely that the bulk atomic structure persists, even very close to the interface. Even if bulk atomic structure persists close to the interface, it is likely that the interface is not atomically flat, and that both  $\text{FeF}_2$  sublattices will contain spins which are coupled to the Fe spins.

This coupling of both sublattices can lead to frustration and uniaxial anisotropy in the following manner. Because the ordered  $\text{FeF}_2$  spins are along the  $\text{FeF}_2$   $c$  axis, interatomic exchange coupling would then cause half the surface to have Fe spins which tend to point parallel to the  $\text{FeF}_2$ [001] direction, while in the other half the Fe spins would tend to point in the opposite direction. In this model no uniaxial anisotropy would exist. Now suppose that the AFM spins near the interface are allowed to cant slightly in response to the interatomic exchange coupling. In the Fig. 1 (110) inset, if a ferromagnet were pointing to the right, this would mean that the two sublattices would rotate slightly to the right side of the page. Then each sublattice would have reduced frustration energy compared to the uncanted state. This process will be more effective at lowering the energy state of the system when Fe and  $\text{FeF}_2$  spin directions are perpendicular rather than parallel, therefore making the perpendicular arrangement a lower energy state.

This model is similar to the one proposed by Slonczewski to describe perpendicular coupling in Fe/Cr multilayers.<sup>16</sup> Micromagnetic calculations by Koon confirm that perpendicular coupling does result when canting is allowed and realistic parameters are used.<sup>17</sup> However, we should point out that Koon's calculations seem to imply larger exchange bias if the cooling field is applied perpendicular to the AFM spin axis, contrary to the results mentioned above.

Alternatively, it is possible that the Fe easy axis rotation is caused by magnetoelastic effects rather than by interatomic exchange coupling. The magnetoelastic model is supported by the observation of different thermal expansion behavior for the  $c$  lattice parameter and the  $a$  lattice parameter in  $\text{FeF}_2$ .<sup>18</sup> Compared to 293 K, the ( $c/a$ ) ratio  $R$  is larger at 100 K, [ $R(100\text{ K})/R(293\text{ K})=1.0006$ ], then gradually drops as the temperature drops below the  $\text{FeF}_2$  ordering temperature [ $R(60\text{ K})/R(293\text{ K})=0.9999$ ]. If this structural change causes a structural change in the neighboring Fe layer, the magnetoelastic properties of the  $\text{FeF}_2$  would produce a uniaxial anisotropy change similar to that observed in Figs. 2–4.

This model is also supported by the difference in exchange bias behavior of different  $\text{FeF}_2$  interfaces. Since the  $\text{FeF}_2$ (110) interface displays larger exchange bias than the  $\text{FeF}_2$ (100) interface, it is likely that the small-scale details of the exchange coupling are very different. The similar perpendicular coupling behavior of the two interfaces suggests that something other than exchange coupling, possibly magnetoelastic effects, are more important.

Besides the perpendicular coupling at  $\text{FeF}_2$  interfaces<sup>7</sup>

discussed here, similar effects have been reported at permalloy/CoO interfaces,<sup>6</sup> permalloy/FeMn interfaces,<sup>19</sup> and  $\text{Fe}_3\text{O}_4$ /CoO interfaces.<sup>20</sup> In fact, for the permalloy/CoO interfaces with CoO single crystals, perpendicular coupling due to antiferromagnetism was apparently the main coercivity mechanism. Therefore, controlling the perpendicular coupling could be useful in controlling the coercivity of exchange bias systems in general. This is true regardless of whether the mechanism is driven by exchange coupling or magnetoelastic effects.

In conclusion, we have studied the magnetization rotation behavior of Fe thin films coupled to  $\text{FeF}_2$  single crystals. We found that the Fe easy axis changes its direction as the  $\text{FeF}_2$  goes through its antiferromagnetic ordering temperature. The results for (110) and (100) surfaces clearly indicate that there is a perpendicular coupling between the Fe spins and the ordered  $\text{FeF}_2$  spins. This behavior is consistent with recent micromagnetic calculations on FM–AFM interfaces and with a magnetoelastic coupling mechanism.

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