

# Increased exchange anisotropy due to disorder at permalloy/CoO interfaces

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The exchange anisotropy of Ni<sub>80</sub>Fe<sub>20</sub> (permalloy) films deposited onto bulk single crystal CoO substrates using molecular-beam epitaxy is strongly affected by the interface structure. Contrary to expectations for uncompensated interfaces, the loop shift increases for samples with greater interface disorder. The results are discussed in the light of proposed exchange anisotropy models. The results suggest that antiferromagnet domain dynamics are an important part of the exchange anisotropy mechanism. © 1995 American Institute of Physics.

## I. INTRODUCTION

Magnetic anisotropy caused by the interaction of neighboring magnetic layers, known as exchange anisotropy, has been studied in a number of systems,<sup>1-9</sup> especially permalloy/FeMn,<sup>1-5</sup> and permalloy/Ni<sub>x</sub>Co<sub>1-x</sub>O.<sup>6-8</sup> An interesting feature of these ferromagnet/antiferromagnet systems is that if the sample is cooled through the Néel temperature ( $T_N$ ) of the antiferromagnet (AFM) in the presence of a magnetic field, the ferromagnet (FM) displays a unidirectional magnetic anisotropy. This unidirectional anisotropy is observed in magnetization loops as a shift  $H_E$  of the center of the loop along the field axis. The measured values of  $H_E$  cannot be explained using simple atomic models of exchange interactions; therefore models which allow for AFM domain dynamics have been proposed.<sup>10-13</sup> The application potential of exchange anisotropy has increased recently due to the role it plays in certain magnetic sensors based on the giant magnetoresistive effect.<sup>14,15</sup>

The mechanism responsible for exchange anisotropy is clearly dependent on the magnetic interaction across the interface between the FM and the AFM. However, the only studies that have been published to date which address the question of the atomic structure of exchange anisotropy interfaces have been complicated by their use of the antiferromagnetic FeMn, which can exist in different phases.<sup>5,15</sup> A systematic study of exchange anisotropy has been performed in samples where the interface structure was controlled with a variety of techniques. Contrary to expectations based on the simple exchange anisotropy models, interface disorder is shown to increase  $H_E$ .

CoO was chosen for the AFM because of its experimentally convenient Néel temperature ( $T_N=291$  K) and its simple crystal structure (NaCl with a slight distortion below  $T_N$ ).<sup>16</sup> CoO bulk single crystals with (111) orientation were used since the ordered state of CoO has (111) planes of ferromagnetically aligned Co spins<sup>17</sup> (Fig. 1). This was expected to raise the possibility of forming uncompensated spin planes parallel to the interface, leading to large values for the exchange bias. However, since four types of (111) planes are possible, the ordered (111) planes of CoO spins may or may not be parallel to the interface. There is some

controversy as to the spin orientation.<sup>18</sup> In addition, interface roughness, FM-AFM interactions, and the 17% lattice mismatch between CoO and permalloy may complicate the interface spin structure, making it different from Fig. 1. Permalloy was chosen for the FM because its small magnetic anisotropy causes relatively small coercivity, which simplifies the analysis of the loop shifts.

## II. EXPERIMENT

The samples were prepared using a Riber ultra-high vacuum molecular-beam epitaxy (MBE) system ( $2 \times 10^{-10}$  Torr base pressure) equipped with reflection high-energy electron diffraction (RHEED), low-energy electron diffraction (LEED), and Ar ion bombardment. CoO(111) bulk single crystals were used as substrates. To control the surface structure, some CoO crystals were sanded with 400 grit sandpaper while others were polished using 6 and 1 diamond powders, and 0.05  $\mu\text{m}$  alumina powders (from Buehler Ltd). All substrates were rinsed in methanol before introduction into the chamber, where they were bombarded with Ar ions for 30 min to clean the surface.

Crystals which were not well-polished displayed very faint RHEED patterns or none at all, indicating very rough or disordered surfaces. Polished samples showed transmission-type RHEED spots which became sharper after cycles of ion bombardment and 1100 °C annealing, indicating an improvement in the surface ordering. Ni and Fe were codeposited, at room temperature, using two electron beam guns with their rates controlled to produce the desired Ni<sub>80</sub>Fe<sub>20</sub> alloy at the substrate position. Deposition rates were controlled using Sentinel electron impact emission spectroscopy rate monitors from Inficon. During typical deposition conditions (total rate  $\sim 1$  Å/s) the pressure was better than  $5 \times 10^{-9}$  Torr. No aligning field was used during growth. The crystal structure was determined from x-ray diffraction using Cu  $K_\alpha$  radiation.

Magnetic measurements were performed using a superconducting quantum interference device magnetometer with the magnetic field in the plane of the film. Before each measurement run the samples were heated to 400 K, above the Néel temperature of CoO, and then cooled to 50 K in a field of 1500 Oe. Hysteresis loop measurements were then performed in steps of 50 K up to a maximum temperature of 400 K.

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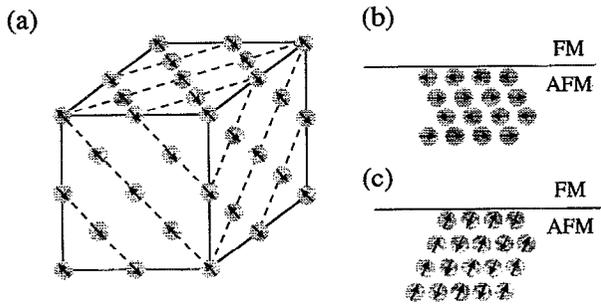


FIG. 1. (a) Bulk spin structure of CoO, with dotted lines connecting spins within identical (111) planes. Oxygen atoms and the interior spins of the unit cell are not shown. For simplicity the spins are shown as parallel to the (111) planes, although the actual structure is controversial. (b) Possible side view of CoO spins where the AFM periodicity is normal to the interface, producing an uncompensated interface. (c) Possible side view of CoO spins where the AFM periodicity is not normal to the interface, producing a compensated interface.

### III. RESULTS

Figure 2 shows the magnetization curves for a permalloy(200 Å)/CoO sample above and below  $T_N$ , from which a linear background due to the CoO has been subtracted. Care was taken to ensure that this subtraction did not introduce artifacts, such as vertical offsets, in the resulting permalloy magnetization data, which could distort  $H_E$  measurements. The offset field  $H_E$ , also known as the exchange bias, is defined as the midpoint of the two zero magnetization crossing points, while the coercivity  $H_C$  is defined as one half of the total separation between these points. Varying the in-plane orientation of the applied field with respect to the crystal had no significant effect (less than 10%) on  $H_E$  or  $H_C$ . This is somewhat surprising considering that the CoO crystalline anisotropy should cause the Co spins to point along preferred axes. When magnetization loops were measured repeatedly without changing conditions, small changes (less than 20%) in  $H_E$  or  $H_C$  were sometimes observed.

Figure 3 shows  $H_E$  and  $H_C$  as a function of temperature for two samples with identical preparation, except that one

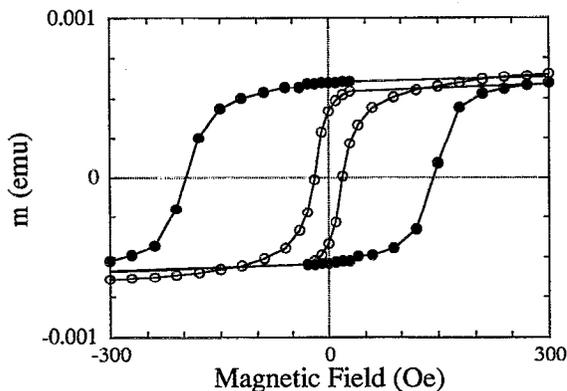


FIG. 2. Magnetization curves for permalloy (200 Å)/CoO measured at 50 K (●) and 350 K (○). The slight mismatch between the upper and lower 350 K curves is attributed to errors in the large CoO background subtraction.

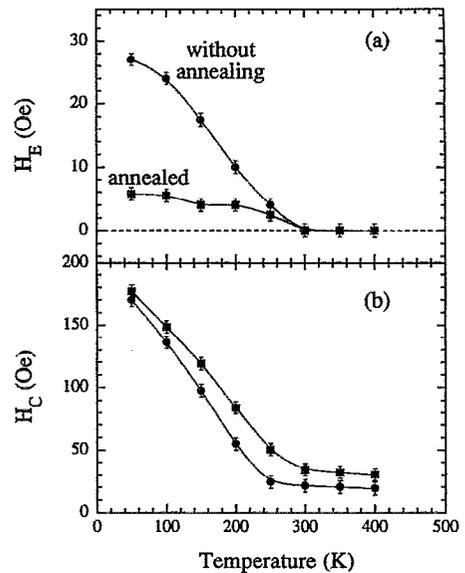


FIG. 3.  $H_E$  and  $H_C$  vs measurement temperature for two permalloy (200 Å)/CoO samples: (■) annealed at 1100 °C for 36 h prior to deposition, (●) without annealing.

was annealed at 1100 °C for 36 h prior to deposition. In these samples, as in all others measured,  $H_E$  disappears above the  $T_N$  of CoO, which demonstrates that the loop shift is due to the presence of the CoO. The approximately linear temperature dependence of  $H_E$  [Fig. 3(a)] has been observed previously<sup>2,3,7</sup> and may be related to the temperature dependence of the AFM order parameter.<sup>12</sup> The increase in  $H_C$  below  $T_N$ , observed previously in the permalloy/CoO system,<sup>6</sup> shows that the ordered CoO spins are causing a barrier to permalloy spin rotation. This implies that a large number of CoO spins must be rotating during each hysteresis cycle.<sup>10</sup> The higher  $H_C$  could be caused by AFM spins encountering coercive mechanisms analogous to those found within ferromagnets. This model is supported by torque magnetometer measurements on permalloy/FeMn samples,<sup>19</sup> and is in contrast to conventional models of coercivity which rely on mechanisms within the ferromagnet.<sup>20</sup> The sharp change in the slope of  $H_C$  versus  $T$  occurs at higher temperatures (260–290 K) for samples which had been annealed just prior to deposition, compared to (225–235 K) for the other samples.

A number of techniques were used to examine the structure of the interface, with some providing more useful information than others. Both a scanning electron microscope and a profilometer (Dektak) were used to examine the surfaces of the samples after deposition and showed that polished samples were smoother than sanded samples. The profilometer showed that the polished samples had ~500 Å of peak-to-valley roughness over a typical 50 μm range compared to ~4000 Å for the sanded samples. Unfortunately, neither of these techniques were able to differentiate between annealed and unannealed samples, which exhibit a large change in  $H_E$ . X-ray diffraction, being more sensitive to structural changes at small length scales (~100 Å), was more useful in detecting changes between annealed and unannealed

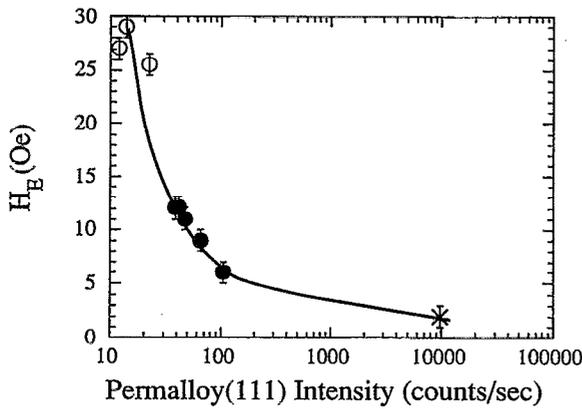


FIG. 4.  $H_E$  ( $T=50$  K) plotted vs intensity of permalloy (111) x-ray diffraction peak for permalloy (200 Å)/CoO samples. Different symbols show preparation method: (○) sanded, (●) sanded and annealed, (X) polished and annealed. The line is a guide to the eye.

samples. Figure 4 shows  $H_E$  at 50 K, as a function of the permalloy (111)  $\theta$ - $2\theta$  x-ray diffraction peak intensity. The fact that deposition conditions were identical for all samples, implies that the CoO surfaces of samples with lower x-ray intensities were more disordered before deposition. Therefore, the permalloy x-ray diffraction data is used to characterize the disorder of the resulting permalloy/CoO interfaces and implies that samples with disordered interfaces have larger  $H_E$  values.

To verify that interface disorder enhances  $H_E$  an additional ion bombardment experiment was performed (Fig. 5). For these samples the CoO crystals were polished, ion bombarded, and then annealed. After cooling to room temperature, some were ion bombarded again just prior to deposition. RHEED confirmed that ion bombardment disordered the surface of the CoO crystals, since RHEED images of the permalloy surface after deposition showed more streaked patterns for the samples without additional ion bombard-

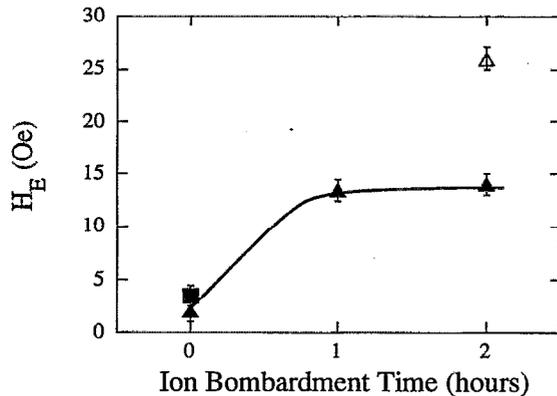


FIG. 5.  $H_E$  ( $T=50$  K) vs ion bombardment time: (▲) permalloy (200 Å)/CoO, (■) permalloy (200 Å)/CoO deposited at 300 °C, (△) permalloy (100 Å)/CoO. The line is a guide to the eye, showing that exposure to ion bombardment increased the measured exchange bias values. The sample deposited at a higher substrate temperature (■) agrees with this trend. The sample with a thinner FM layer (△) had a larger exchange bias value, indicating that coupling is an interface effect.

ment. Clearly  $H_E$  increases with ion bombardment time. Unlike the data shown in Fig. 4, a clear trend was not observed when  $H_E$  was plotted as a function of permalloy(111) peak height. This indicates that the structural effects of ion damage are different from those of mechanical damage, although both increase the exchange anisotropy. Note also that  $H_E$  scales inversely with the FM thickness, as expected for an interface effect such as exchange anisotropy.

#### IV. DISCUSSION

Exchange anisotropy models proposed to date are based on various forms of the following simplified Hamiltonian:

$$\begin{aligned} \mathcal{H} &= \mathcal{H}_1 + \mathcal{H}_2 + \mathcal{H}_3 + \mathcal{H}_4 + \mathcal{H}_5 + \mathcal{H}_6 \\ &= \sum_{i,j \text{ in FM}} J_{ij}^{\text{FM}} S_i \cdot S_j + \sum_{i \text{ in FM}} A_i^{\text{FM}}(S_i) + \sum_{i \text{ in FM}} H \cdot S_i \\ &\quad + \sum_{i,j \text{ in AFM}} J_{ij}^{\text{AFM}} S_i \cdot S_j + \sum_{i \text{ in AFM}} A_i^{\text{AFM}}(S_i) \\ &\quad + \sum_{\substack{i \text{ in FM} \\ j \text{ in AFM}}} J_{ij}^{\text{INTERFACE}} S_i \cdot S_j, \end{aligned} \quad (1)$$

where  $S$  is the magnetic moment,  $H$  the applied magnetic field,  $A(S_i)$  the anisotropy energy for the spin  $S_i$ , and the  $J$ 's are the various exchange coupling constants. Using Eq. 1,  $H_E$  can be calculated by comparing the energy difference between configurations with opposite FM spin directions. The challenge of explaining exchange anisotropy is to calculate the effect of  $\mathcal{H}_6$ , which couples the FM and AFM spins. The effects of ordered AFM planes, interface coupling disorder, AFM orientation, FM structural disorder, AFM domain formation, AFM pinning, and dipolar coupling will all be considered separately.

The simplest exchange anisotropy model describes the AFM as having planes of ferromagnetically ordered spins which are parallel to an atomically flat FM/AFM interface.<sup>2,20</sup> The planes of ordered spins result from cooling the sample through  $T_N$  in an applied magnetic field, and are assumed to remain fixed below  $T_N$ . All AFM spins at the interface point in the same direction and possess an identical magnetic exchange interaction with neighboring FM spins,  $J^{\text{INTERFACE}}$ . Within this model  $H_E$  can be calculated by considering only  $\mathcal{H}_3$  and  $\mathcal{H}_6$  in Eq. 1 since the other terms do not change during FM spin rotation. However, the  $H_E$  values that result are about two orders of magnitude larger than measured values. This is thought to be related to the failure to include changes in the AFM domain structure, which affect the other terms in Eq. 1.<sup>10,11</sup> If AFM spin rotation during FM spin rotation is included by calculating changes in  $\mathcal{H}_4$  and  $\mathcal{H}_5$  in Eq. 1, the calculated  $H_E$  values are in better agreement with experiments.

Atomically rough interfaces and nonuniform atomic coupling at different regions of the interface are thought to reduce net FM/AFM coupling. This is because structural disorder at the interface is expected to produce different types of exchange interactions between the FM and the AFM, expressed in Eq. 1 as causing a range of positive and negative  $J^{\text{INTERFACE}}$  values. Thus, different regions of the AFM will

tend to orient the FM in different directions, leading to a lower net exchange anisotropy. Therefore, within these models ordered interfaces should produce larger  $H_E$  values, contrary to the data presented in Figs. 4 and 5.

Another possible explanation is that increased disorder inside the FM layer, rather than inside the AFM or at the interface, could be causing the increased exchange anisotropy. However, FM structural disorder is expected to inhibit coupling between different FM regions, leading to smaller overall coupling between the FM and the AFM. This also contradicts the present data. The polycrystalline nature of the more disordered permalloy films, deduced from the presence of small ( $<40$  counts/s) permalloy(200) x-ray diffraction peaks, could also have some effect if planes other than the permalloy(111) couple differently with CoO, however there is no obvious mechanism which could explain such behavior.

Malozemoff proposed a model which assumes that real interfaces possess nonuniform interface couplings, and calculated the resulting exchange anisotropy.<sup>11-13</sup> Within this model the random  $J^{\text{INTERFACE}}$  values cause the AFM to break up into small domains along the interface, to minimize the total energy during field cooling. The magnitude of  $H_E$  is predicted to vary inversely with the lateral size of the AFM domains, their size determined by factors such as the interfacial couplings, structural disorder, crystal anisotropies, and thickness of the AFM layer. For FM/AFM samples with a defect-free thick AFM, this model predicts the formation of large AFM domains and  $H_E=0$ , and implies that structural defects may decrease the lateral size of the AFM domains and increase  $H_E$ , in agreement with data presented above. For the samples described above the model predicts AFM domain sizes on the order of 400 Å.<sup>21</sup> These AFM domains may be observable in certain systems using neutron diffraction or perhaps by their effects on FM domain structure.

Yet another explanation for the  $H_E$  increase with structural disorder could be related to rotation of the AFM spins during FM spin rotation, as suggested by the data in Fig. 3(b). Defects in the AFM could pin the AFM spins, preventing them from rotating together with the FM spins, making FM spin rotation more difficult. Therefore samples with more disordered interfaces would display a larger unidirectional anisotropy.

Dipolar coupling between FM and AFM spins is a possible exchange anisotropy mechanism due to the tendency for magnetic fields to be accentuated near protrusions of magnetic layers. Recent calculations<sup>22</sup> have shown that such effects may be comparable to magnetic exchange effects in FM/metal/FM structures in qualitative agreement with experimental data.<sup>23</sup> Perhaps a similar mechanism could be responsible for the increase in exchange anisotropy for disordered interfaces.

These results appear to contradict a recent study of exchange anisotropy in Fe layers deposited on top of FeF<sub>2</sub> layers on MgO substrates [Ag(90 Å)/Fe(120 Å)/FeF<sub>2</sub>(900 Å)/MgO].<sup>24</sup> In that case the interface roughness was controlled by varying the FeF<sub>2</sub> deposition temperature and measured using low angle x-ray diffraction. The exchange bias was lower in samples with greater interface roughness, which was attributed to roughness decreasing the amount of

interface adjacent to FeF<sub>2</sub>(110) planes. A similar mechanism might be at work here, assuming that the CoO(111) planes are relatively ineffective at producing exchange bias and interface disorder decreases the amount of interface adjacent to CoO(111) planes. It should be noted that since the interface preparation, disorder measurement, and AFM spin structures are different for the two experiments, the mechanisms determining the size of the exchange bias need not be the same. In fact, preliminary data for permalloy/CoO(100) samples suggest that the exchange bias has very little orientation dependence.

## V. CONCLUSION

In conclusion, it has been found that interfacial disorder induced by ion damage or mechanical treatments enhances  $H_E$  in the CoO/permalloy system, contrary to naive expectations and in qualitative agreement with models based on AFM domain dynamics.  $H_E$  and  $H_C$  exhibit a linear temperature dependence up to  $T_N(\text{CoO})$ . Further experiments are needed to provide information on the AFM domain structure to help identify the exchange anisotropy mechanism.

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