

Exchange biasing and interface structure in MnNi/Fe(Mo) bilayers

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The role of magnetic, structural, and chemical roughness on the origin of exchange biasing in polycrystalline Mn₅₂Ni₄₈/Fe₉₂(Mo₈) bilayers has been investigated by transmission x-ray magnetic circular dichroism (XMCD), vibrating sample magnetometry (VSM), and transmission electron microscopy (TEM). Three bilayer samples of MnNi(22 nm)/Fe(Mo) (6 nm) were grown by molecular beam epitaxy under ultrahigh vacuum conditions with the MnNi layer at temperatures of 200 °C, 250 °C, and 300 °C. The exchange bias, H_e , was observed to be the largest for the 250 °C growth sample. The angular dependence of H_e can be well modeled in terms of a cosine series with odd terms confirming the unidirectional nature of the anisotropy energy. However, the coefficients are different for the three samples indicating different microscopic magnetic interactions at the interface. XMCD measurements showed no magnetic moment for Mn and Ni but showed systematic variations of the Fe moment, i.e., a decrease in Fe moment with increase in H_e was observed. We have interpreted this decrease in Fe moment in terms of antiferromagnetic (AF) ordering of Fe at the interface with the extent of the AF ordering being related to the magnitude of the exchange. Thus, for samples grown at 250 °C, it is found that at least about 4 MLs of Fe appear to be AF. Preliminary energy-filtered imaging of cross-section samples shows that the Fe layer is chemically rough suggesting that the decrease in moment may arise from the intermixing of Fe with the MnNi layer.

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Antiferromagnetic (AF) exchange is important for both longitudinal and transverse pinning of soft adjacent-layer magnetoresistive and giant magnetoresistive spin valve heads.^{1,2} Investigations of exchange coupling between antiferromagnetic and ferromagnetic thin films have been carried out for several decades because of the interest in both physics and technology. The hysteresis loop of a FM layer exchange-coupled to an AF layer is shifted from the origin by an amount known as the exchange field (H_e) and is often accompanied by an enhanced coercivity (H_c).³ However, many questions concerning this exchange coupling remain unanswered. The earliest theory by Meiklejohn and Bean⁴ explained the effect in terms of an uncompensated monolayer of spin at the surface of the antiferromagnetic layer. However, this model predicted an exchange coupling strength 100 times greater than that observed experimentally. Several recent theories^{5,6-8} give improved predictions of the magnitude of H_e , but they differ on the physical explanation of the effect. For example, Malozemoff⁵ assumed that random structural roughness of the surface greatly reduces the number of uncompensated spins at the AF surface giving rise to a smaller H_e . Alternatively, Mauri's model⁶ assumes that AF films form domain walls at the interface which reduces H_e . Recently, Koon⁷ suggested 90° or spin-flop coupling

between the AF and FM layers and correctly predicted the magnitude of H_e . However, the sign of the bias was not definitely determined.

The experimental evidence for the origin of exchange is also far from conclusive. Ijiri *et al.*⁹ used neutron diffraction and found spin-flop coupling in Fe₃O₄/CoO superlattices in support of Koon's model. Qiu *et al.*¹⁰ used Fe films grown on stepped Cr surfaces and proved the existence of 90° coupling by measuring hysteresis loops that agreed with Koon's calculation. Recently, Takano *et al.*¹¹ performed direct measurements of uncompensated spins at the surface of the CoO layer in CoO/MgO and CoO/FeNi superlattices. They found approximately 1% of one monolayer (ML) of the Co surface spins were uncompensated and showed that these spins would lead to exchange biasing in such FM/AF bilayers in support of Malozemoff's model. However, most of those experiments were carried out on insulating systems. Here we used x-ray magnetic circular dichroism (XMCD), in conjunction with transmission electron microscopy (TEM), to study the spin structure in the metallic system MnNi/Fe(Mo) and explain the observed variation in exchange interaction between the FM and AF layers.

Polycrystalline bilayer samples of the structure Mn₅₂Ni₄₈(22 nm)/Fe₉₂(Mo₈)(6 nm) were grown on SiN (100 nm) membrane substrates by molecular beam epitaxy under ultrahigh vacuum conditions. The Fe(Mo) was grown at 50 °C followed by the growth of MnNi in a magnetic field of

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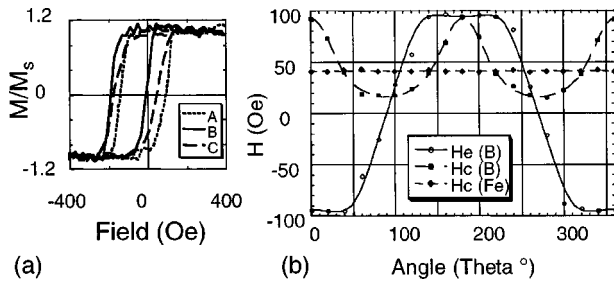


FIG. 1. (a) Hysteresis loops for the samples A, B, C. (b) Experimental results of angular dependence of H_e , H_c of sample B (\circ \blacksquare). Also shown is the angular dependence of H_c for a single Fe(Mo) layer (\blacklozenge). The curves are fitting results. ϑ is the inplane angle relative to the biasing field direction.

1 kG at three different growth temperatures (T_{sub}), 200 °C(A), 250 °C(B), and 300 °C(C). The Au (2 nm) capping layer was used to protect against oxidation. The 8% Mo was added in the Fe layer in order to reduce the coercivity. The external field serves to set the bias field direction of the ferromagnetic layers. The composition of the films was confirmed by particle induced x-ray emission (PIXE), Rutherford backscattering spectroscopy (RBS) measurements, and x-ray microanalysis. TEM bright field, selected area diffraction, high resolution imaging, and energy filtered imaging using inner-shell ionization edges of plan view and cross-section samples were carried out to determine the structural details of the bilayers at high resolution. Hysteresis loops were measured for all the samples using a vibrating sample magnetometer (VSM) to determine the exchange fields and coercivities. XMCD measurements using synchrotron radiation were carried out to get the interface spin structure.

Figure 1(a) shows the hysteresis loops for the MnNi/Fe(Mo) samples. The exchange field and coercivity (H_e , H_c) for the three samples A, B, C in the as-grown condition were (23, 120 Oe), (94, 92 Oe), and (59, 112 Oe), respectively. Detailed measurements of H_e and H_c as a function of the in-plane angle ϑ were carried out and a representative plot for sample B is shown in Fig. 1(b). The observed angular dependence is consistent with that measured previously for the NiFe/CoO bilayer system.³ In contrast to the exchange-coupled bilayer, a single Fe(Mo) layer shows much lower coercivity and no angular dependence. This indicates that for the exchange-coupled FM/AF bilayers, both H_e and H_c are closely related to the exchange coupling.

The angular dependence of H_e (H_c) can be expressed in terms of a cosine series with odd (even) terms, as they depend on unidirectional (uniaxial) parts of the anisotropy energy. The following expressions were obtained for samples A, B and C:

$$\begin{aligned}
 H_e(A) &= -26 \text{ Oe} [\cos(\vartheta) - 0.34 \cos(3\vartheta) \\
 &\quad + 0.13 \cos(5\vartheta) - 0.07 \cos(7\vartheta) + \dots], \\
 H_c(A) &= 80 \text{ Oe} [1 + 0.46 \cos(2\vartheta) - 0.04 \cos(4\vartheta) \\
 &\quad + 0.01 \cos(6\vartheta) + \dots], \\
 H_e(B) &= -111 \text{ Oe} [\cos(\vartheta) - 0.15 \cos(3\vartheta) \\
 &\quad - 0.01 \cos(5\vartheta) + 0.01 \cos(7\vartheta) + \dots],
 \end{aligned}$$

$$\begin{aligned}
 H_c(B) &= 44 \text{ Oe} [1 + 0.82 \cos(2\vartheta) + 0.24 \cos(4\vartheta) \\
 &\quad + 0.05 \cos(6\vartheta) + \dots],
 \end{aligned}$$

$$\begin{aligned}
 H_e(C) &= -62 \text{ Oe} [\cos(\vartheta) - 0.07 \cos(3\vartheta) \\
 &\quad + 0.01 \cos(5\vartheta) + 0.02 \cos(7\vartheta) + \dots],
 \end{aligned}$$

$$\begin{aligned}
 H_c(C) &= 59 \text{ Oe} [1 + 0.62 \cos(2\vartheta) + 0.13 \cos(4\vartheta) \\
 &\quad + 0.06 \cos(6\vartheta) + \dots].
 \end{aligned}$$

The significant difference in the coefficients for the three samples can be attributed to the different spin configuration or interaction at the interface.

The spin structure of each layer was determined by XMCD.¹² The transmission method was chosen for this experiment because it is more reliable compared to other methods such as the total electron yield, which is known to suffer from saturation and self-absorption effects that are difficult to correct. The principle of XMCD is to measure differences in x-ray-absorption spectra of circularly polarized photons for a fixed orientation between the magnetic moment of the sample and the photon helicity. The measurement is element specific and hence the magnetic structure of the different layers in the structure can be individually resolved. The XMCD measurements were carried out on the bending magnet beamline 9.3.2 at the Advanced Light Source. Typically, 90% circularly polarized light is obtained by using x rays emitted above or below the electron orbital plane. The x-ray beam is incident on the sample at an angle of 45° with respect to the surface normal. By switching the magnetic field by 180°, two transmission x-ray absorption spectra were recorded with a component of M parallel and antiparallel to the photon helicity. The difference between these two spectra is the XMCD and is proportional to the average magnetic moment per atom. This lets us characterize ferromagnetic properties of the Fe(Mo), NiMn, and their interface. Note that circular dichroism is insensitive to antiferromagnetic ordering and hence for MnNi only deviations from AFM ordering can be resolved.

XMCD spectra for all three elements (Ni, Mn, and Fe), using $L_{3,2}$ (excitation from filled 2p levels to unoccupied 3d states) absorption edges were measured. For all samples, no dichroism was observed for Mn suggesting that the Mn spins are perfectly compensated and remain an integral part of the antiferromagnetic layers. Similarly, our measurement in transmission did not find any discernible change in the moment for Ni. However, the XMCD signal (Fig. 2) for Fe was different for the three samples (normalized for thickness variations based on RBS measurement). The smallest value of the magnetic moment for Fe was for sample B which shows the largest value of H_e . Moreover, for these three samples, it is uniformly seen that the higher the exchange field, H_e , the lower the value of the moment for Fe as measured by XMCD.

The reduction in the magnetic moment of Fe could be attributed to either the compositional variation of Fe(Mo) or to the magnetic intermixing of Fe with NiMn leading it to order antiferromagnetically (AFM) at the interface. The former can be ruled out since the small variation in the com-

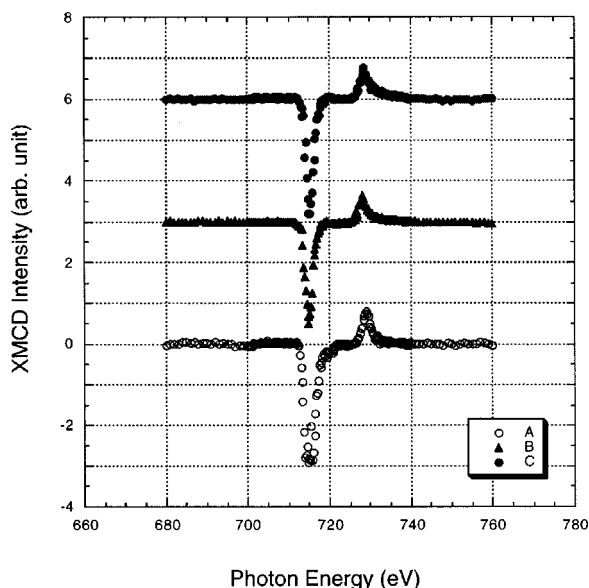


FIG. 2. XMCD intensity vs photon energy at $L_{3,2}$ edges of Fe for the samples A, B, C.

position of the film does not correlate systematically with the observed trend in the Fe moment vis-à-vis the H_e . Thus, the extent of the AFM ordering of Fe is closely related to the magnitude of the exchange field. This is quite plausible since the film shows (111) texture—as measured by x-ray diffraction—and the (111) surface of NiMn is magnetically compensated with an antiparallel spin arrangement.¹³ A possible mechanism to accomplish this ordering is the chemical interdiffusion of Fe into the MnNi layer, with the potential formation of an alloy or compound layer extending over a few monolayers at the interface. A first order estimate based on the magnitude of the XMCD signal suggests that this chemically mixed or AFM ordered layer is of the order of at least four monolayers.

TEM of cross-section samples have been initiated to resolve the physical and chemical roughness of the interface. A high resolution micrograph [Fig. 3(a)] of sample B shows that the interface between Fe and MnNi is truly indistinguishable. This is because the structure-factor and diffraction contrast mechanism do not easily resolve layers of materials of similar atomic numbers. On the other hand, energy-filtered imaging using characteristic inner-shell ionization edges in electron energy-loss spectroscopy can be carried out at high spatial resolution (~ 1 nm). Such energy-filtered images using the $L_{3,2}$ edges of Fe, Ni, and Mn (with 30 eV windows), after background subtraction and suitable correction to accommodate any thickness variations for sample B are shown in Figs. 3(b)–3(d). The individual layers corresponding to MnNi and Fe are clearly evident. However, it is clear that the Fe layer is not chemically sharp indicating a chemical intermixing at the interface. Systematic energy filtered imaging studies comparing these three samples as well as epitaxially grown films with well defined crystallographic interfaces are in progress.

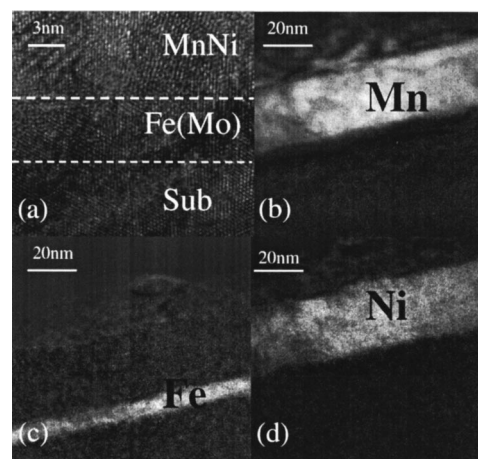


FIG. 3. (a) High resolution image of sample B and energy filtered image of sample B using the (b) Mn (c) Fe, and (d) Ni $L_{3,2}$ edges.

In conclusion, we have shown a systematic variation in the exchange field for samples grown at different temperatures. As expected, the angular dependence of H_e can be well modeled in terms of a cosine series with odd terms confirming the unidirectional nature of the anisotropy energy. However, the coefficients are different for the three samples indicating different microscopic magnetic interactions at the interface. Thus XMCD, used to measure the spin structure at the interface, showed a decrease in Fe moment with increase in H_e , i.e., a part of the Fe at the interface is AFM ordered and the extent of the AFM ordering is clearly related to the exchange. For sample B ($T_{\text{sub}} \sim 250^\circ\text{C}$), it is found that at least about 4 MLs of Fe appear to be AF. Preliminary, energy-filter imaging of cross-section samples shows that the Fe layer is chemically rough suggesting that the decrease in moment may arise from the interdiffusion of Fe into the MnNi layer. For example, interdiffusion could result in local Fe–Mn and FeMn is a well-known disordered AF.

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