SPIN-FLOP TENDENCIES IN EXCHANGE-BIASED Co/CoO THIN FILMS*

G. P. Felcher^a, J. A. Borchers^b, Y. Ijiri^b, S.-H. Lee^b, C. F. Majkrzak^b, K. Takano^c,
R. H. Kodama^c, and A. E. Berkowitz^c

^aMaterials Science Division
9700 South Cass Avenue
Argonne National Laboratory
Argonne, IL 60439

^bNational Institute of Standards and Technology
Gaithersburg, MD 20899

^cCenter for Magnetic Recording Research
University of California at San Diego
La Jolla, CA 94720

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J.A. Borchers, Y. Ijiri, S.-H. Lee, and C.F. Majkrzak

National Institute of Standards and Technology

Gaithersburg, Maryland 20899

G.P. Felcher

Argonne National Laboratory

Argonne, Illinois, 60439

Kentaro Takano, R.H. Kodama, and A.E. Berkowitz

Center for Magnetic Recording Research, University of California at San Diego

La Jolla, California, 94720

Abstract

In order to study the antiferromagnetic (AFM) spin structure near the interface of exchange-biased bilayers, polarized neutron diffraction measurements were performed on a series of (111) Co(7.5 nm)/CoO(X nm) and CoO(X nm) thin films where $X = 20, 40$ and 100 nm. In these samples, field cooling through the Néel temperature of the AFM increases the component of the CoO moment perpendicular to the applied field, relative to the parallel component. The subsequent application of a 500 Oe field perpendicular to the cooling direction rotates both the Co and CoO moments. Experiments on CoO films without Co showed a smaller difference between the parallel and perpendicular CoO moments in response to cooling and applied fields. Ex-

change coupling between the Co and CoO layers is apparently responsible for the increased projection of the AFM moments perpendicular to the cooling field.

The term "exchange-biasing" describes a phenomenon associated with interfacial coupling between ferromagnetic (FM) and antiferromagnetic (AFM) materials. In particular, a shift of the magnetic hysteresis loop along the field axis is observed after field cooling these composites through the Néel temperature of the AFM. Since its discovery over 40 years ago in oxidized Co particles¹, a number of experimental studies²⁻⁶ have focused on layered systems because of their potential applications in magnetic sensors.⁷ Still unresolved, however, is the link between exchange biasing and the microscopic orientation of the spins in the AFM layer. Early theories^{1,8,9} assumed that the AFM spins at the interface couple collinearly to the FM layer. Alternately, Koon¹⁰ has suggested that the AFM and FM spins can align perpendicular depending on the interfacial structure. Takano, *et al.*⁵ observed a correlation between the thermoremanent moment of polycrystalline CoO/MgO multilayers and the exchange biasing field of CoO/permalloy bilayers. This result suggested that uncompensated interfacial spins are the principal origin of exchange biasing in that system. Unfortunately, it is difficult to determine the AFM spin structures involved in exchange biasing because few experimental techniques can probe AFM order in buried and interfacial layers.

The scope of the present experiment is to determine the orientation of the AFM moments relative to the FM spins in (111) Co(7.5 nm)/CoO(X nm) thin films (X = 20, 40 and 100 nm) using polarized neutron diffraction methods. After field cooling our epitaxial samples, we find that the percentage of CoO spins perpendicular to the field exceeds the percentage of parallel spins. The difference between the parallel and perpendicular AFM moment projections in the Co/CoO bilayers appears to be induced by proximity to the FM layer. This behavior may arise from perpendicular coupling between the Co and CoO spins, as predicted by Koon.¹⁰

For this study, the CoO films were reactively sputtered at 100 °C in 2 mTorr of Ar and a small partial pressure of oxygen (0.09 mTorr) onto 1"-diameter (0001) Al₂O₃ substrates. We examined five single-layer and bilayer samples with differing compositions, SiO₂(10 nm)/CoO(X)/Al₂O₃ (X = 40 and 100 nm) and SiO₂(10 nm)/Co(7.5 nm)/CoO(X)/Al₂O₃ (X = 20, 40 and 100 nm). In order to induce

exchange-biasing and uniaxial anisotropy in the FM layer, the Co/CoO bilayers were deposited in a 300 Oe field applied along the $[11\bar{2}0]$ Al_2O_3 in-plane axis. High-angle x-ray diffraction confirms that the CoO layers are epitaxial with (111) preferred orientation and are twinned in the growth plane.¹¹ Fits to x-ray reflectivity data for the Co/CoO(40 nm) bilayer indicate that the width of the Co/CoO interface is approximately 1.0 nm due to roughness or interdiffusion.

Magnetization measurements performed in a SQUID magnetometer reveal that exchange-biasing is observed for a comparable epitaxial Co/CoO(100 nm) bilayer grown under similar conditions on smaller substrates. Figures 1 (a) and (b) show hysteresis loops at 10 and 200 K respectively after cooling in a 10 kOe field applied along the same direction as the growth field. Both the coercive field and the exchange-biasing field, H_{eb} , change as a function of temperature, as shown in Fig. 1. These field values are expected to differ for bilayers with thinner CoO and may depend on the film structure as determined by the substrate characteristics.

For these and related biased layers,¹² the temperature marking the onset of biasing is nearly equal to the Néel temperature, T_N , of the AFM component. Bulk CoO orders as a collinear antiferromagnet below $T_N = 291$ K. The spins are ferromagnetically aligned in (111) sheets and are nearly perpendicular to a $\langle 111 \rangle$ axis. The moment direction alternates in neighboring (111) planes.¹³ The CoO spins can thus align in any of four equivalent $\{111\}$ domains. For our thin films, the [111] "propagation-axis" of one of these domains is parallel to the growth directions. Due to twinning, however, there are six additional propagation-axis tilted 70.5° from the [111] growth direction. Complementary neutron diffraction studies¹⁴ revealed that a large portion of the CoO spins reside in these six off-axis domains. We restricted our study to the characterization of those spins in the (111) growth-axis domain, which give rise to a $(1/2 \ 1/2 \ 1/2)$ magnetic reflection in our neutron data.

To characterize the AFM structure associated with the biased state in the Co/CoO bilayers, we performed polarized neutron diffraction measurements on these samples using the SPINS triple-axis spectrometer at the NIST Research Reactor. An electromagnet provided

fields ranging from 50 to 2400 Oe. Since the maximum field is limited, we examined our samples at 200 K, rather than 10 K, in order to insure saturation of the Co layer upon field cooling (Fig. 1). On the SPINS spectrometer, polarizing elements were positioned before and after the sample to select the spin direction of the incident and scattered neutrons. The polarization efficiencies were approximately 92%. For all scans through the $(1/2\ 1/2\ 1/2)$ CoO reflection, we measured both the non-spin-flip (NSF) and spin-flip (SF) scattered intensities. The NSF intensity is proportional to the square of the average CoO moment projection parallel to the vertical applied field, while the SF intensity is proportional to the square of the average moment projection perpendicular to the field.¹⁵ These moment projections may belong to the same or to different (111) domains.

Figure 2 shows typical scans through the $(1/2\ 1/2\ 1/2)$ reflection for the Co/CoO(100 nm) bilayer at 200 K. After cooling the sample in zero field, the NSF and SF intensities are approximately equal [Fig. 2 (a)]. These results are consistent with random orientation of the CoO spins within the growth plane. Upon cooling in a -2200 Oe field and measuring in a 2200 Oe field, the SF intensity increases slightly relative to the NSF as shown in Fig. 2 (b). To determine if this difference is significant, we counted the NSF and SF intensities at the center of the $(1/2\ 1/2\ 1/2)$ reflection for extended times. Upon demagnetizing and zero field cooling, the ratio of the NSF to SF intensity is 1.001 ± 0.011 , compared to a ratio of 0.941 ± 0.017 for the same field preparation as in Fig. 2 (b). However, a single-layer CoO(100 nm) film also gives a NSF/SF ratio of 0.94 ± 0.012 upon field cooling in 2400 Oe (Fig. 3). The AFM spins in the Co/CoO(100 nm) bilayer do not clearly show any effects from coupling to the FM Co layer. Since the data in Fig. 2 represent the CoO spin configuration averaged both along the growth axis and across the growth plane, the direction of the CoO moments near the Co/CoO interface can not be directly distinguished from the direction of those moments in the remainder of the 100 nm layer. Any local deviations of the CoO spin direction at the Co/CoO interface may be masked by the bulk behavior of the thick AFM layer.

To determine if the perpendicular AFM spins are close to the interface, we measured the

($1/2\ 1/2\ 1/2$) NSF and SF peak intensities for Co/CoO samples with thinner CoO layers. Initially cooling in zero field, the Co/CoO(40 nm) bilayer shows a tendency for perpendicular spin alignment (NSF/SF = 0.933 ± 0.01). Upon field cooling and measuring in 2400 Oe, the NSF/SF ratio of 0.873 ± 0.01 is significantly smaller than that for the Co/CoO(100 nm) sample, as shown in Fig. 3. This ratio varies only slightly up to 0.895 ± 0.011 when the field is lowered to 150 Oe. Thus this spin configuration appears to “freeze-in” upon field cooling. The average perpendicular projection of the CoO moment is at least 3 % larger than the parallel projection. For the Co/CoO(20 nm) bilayer, a NSF/SF ratio of 0.928 ± 0.017 was measured in 2300 Oe after field cooling. This value is comparable to that for the Co/CoO(40 nm) bilayer. As shown in Fig. 3, the Co/CoO(20 nm) and Co/CoO(40 nm) films have a larger fraction of perpendicular spins than the Co/CoO(100 nm) film. This result may indicate that the canted spins are confined primarily to a finite region near the Co/CoO interface. For a more careful check of this hypothesis, the NSF/SF ratio should be accurately measured over the entire Q range of the Bragg reflection (e.g., Fig. 2).

Overall, these data suggest that the CoO spins in the biased bilayers prefer a perpendicular orientation relative to the cooling field. However, this alignment could either be a consequence of the Co/CoO interlayer coupling or a direct response to the cooling field. To distinguish between these two effects, we performed field-dependent diffraction measurements on a CoO(40 nm) film. Upon field cooling and measuring the sample in a 2300 Oe field, the ratio of the NSF to SF intensities for the ($1/2\ 1/2\ 1/2$) reflection is 1.02 ± 0.017 , which is significantly greater than the value measured for the corresponding Co/CoO(40 nm) bilayer (Fig. 3). The random orientation of the AFM spins is apparently unperturbed by cooling fields up to 2400 Oe. The preferred perpendicular orientation observed in Co/CoO bilayers thus appears to result from interlayer coupling. The following measurement for the Co/CoO(40 nm) bilayer further supports this conclusion. We first cooled the sample to 200 K in a 2400 Oe field applied in-plane parallel to the Co growth field direction. We lowered the field to zero and then applied an in-plane field of 500 Oe at an angle of 90° relative to the field cooling direction while maintaining a temperature of 200 K. The resultant NSF/SF

ratio is 1.02 ± 0.011 , which is larger than the 0.873 ± 0.01 ratio obtained in a 2400 Oe after initially field cooling along the easy axis (Fig. 3). The "frozen" CoO spins thus try to align perpendicular to the reoriented Co layer moment. Since the 500 Oe field is too small to rotate the CoO spins, the moment reorientation is a direct response to the change in the Co magnetization direction.

In conclusion, we observe a preferential orientation of the CoO moments away from the Co magnetization direction in (111) Co/CoO bilayers. Within the (111) growth-axis domain, the perpendicular projection of the CoO moment relative to the field direction is larger than the parallel projection for the three bilayers considered. This result is consistent with Koon's model,¹⁰ which predicts 90° coupling between the FM and AFM layers. The preferred perpendicular orientation of the AFM develops when the Co/CoO bilayer is field cooled through the AFM T_N into the biased state. Thus the CoO spin reorientation films appears to be a direct consequence of exchange coupling between the Co and CoO layers. Unfortunately, we can not as yet determine if the perpendicular spins reside in the interfacial region, or if the spins in the off-axis {111} domains play an important role. By controlling the AFM spin population in the growth-axis and off-axis domains, future diffraction studies of related CoO films may indeed provide a complete understanding of the physical origin of exchange biasing.

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FIGURES

FIG. 1. Magnetization as a function of field for a Co/CoO(10 nm) bilayer at (a) 10 K and (b) 200 K after field cooling from room temperature in a 10 kOe field applied along the in-plane easy axis.

FIG. 2. Polarized neutron diffraction scan through the $(1/2\ 1/2\ 1/2)$ reflection for a Co/CoO(10 nm) bilayer at 200 K. The wavevector Q is defined as $Q = \frac{4\pi}{\lambda} \sin \theta$ where the neutron wavelength $\lambda = 0.417$ nm and θ is the scattering angle. The shaded circles represent the non-spin-flip (NSF) intensity and the open circles mark the spin-flip (SF) intensity. The data in (a) were obtained after demagnetizing the sample and then cooling in zero field (ZFC). The data in (b) were obtained in a 2200 Oe field after field cooling (FC) in -2200 Oe from room temperature. These data have not been corrected for the efficiencies of the polarizing elements.

FIG. 3. Ratio of the non-spin-flip (NSF) to spin-flip (SF) peak intensities for the $(1/2\ 1/2\ 1/2)$ CoO magnetic reflection as a function of CoO layer thickness for several Co/CoO and CoO samples, as marked. The samples were cooled to 200 K in a 2200 - 2400 Oe field and then measured in the same field. These data have not been corrected for background contributions or for the efficiencies of the polarizing elements. Background corrections would increase the ratio for the thinnest samples with the weakest intensities.

Fig 1, Borchers, JAP, 60-06

Fig 2, Borchers, JAP, 60-06

Fig 3, Borchers, JAP, 60-06