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Pinned Co moments in a polycrystalline permalloy/CoO exchange-biased bilayer

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We have measured element-specific magnetization depth profiles across the interface between a polycrystalline ferromagnet and an antiferromagnet in an exchange-biased bilayer of Py/CoO. Using soft x-ray resonant reflectivity we have identified a thin (0.5 nm) layer containing uncompensated Co magnetization at the interface with the Py. The majority of this magnetization follows the external field; however, ~10% of the magnetization in this interfacial layer is pinned antiparallel to the cooling field used when biasing the sample, consistent with the negative exchange bias in this bilayer system, provided that the pinned Co spins are antiferromagnetically coupled to the ferromagnetic layer.

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When an exchange-coupled ferromagnet (FM) and an antiferromagnet (AF) are field cooled through the ordering temperature of the AF, exchange bias—a shift of the hysteresis loop—is observed.[1] The same effect is seen when depositing a ferromagnet on an (ordered) AF in a biasing field.[2] This has been exploited in several industrial applications using thin-film systems, but a detailed microscopic understanding has proved to be elusive.[3,4] The problem of exchange bias is further complicated by the myriad differences observed from system to system.

Interfacial effects are crucial in the development of exchange bias. In particular, the role of interfacial uncompensated spins in the AF is important.[5] However, because the interface is buried these uncompensated spins can be difficult to measure. Penetrating radiation, such as neutrons or x rays, is one of the few tools available to do this. Element-specific resonant magnetic reflectivity, measured as a function of applied field in the field-cooled state, can be used to obtain the depth dependence of the FM and AF components individually. Both pinned and unpinned contributions to the magnetization can be measured, thus providing details regarding the exchange bias mechanism unobtainable by other means. This information averages over a large sample area defined by the size of the beam spot. A series of recent studies has started to explore the ferromagnetic-antiferromagnetic interaction using these tools.[6–11]

Recent work by some of us[11] reported the unbiased room-temperature state of a polycrystalline permalloy (Py)/CoO bilayer, i.e., above the ordering temperature of the CoO (T_N=289 K). That investigation found a thin interfacial layer at the Py/CoO interface, in which some Co atoms possessed a net magnetization at room temperature.

In this Rapid Communication, we present soft x-ray reflectivity data that show that this interfacial layer is still present in the exchange-biased state (i.e., below T_N) and contains uncompensated Co magnetization. These uncompensated moments are divided into two groups. One group is pinned antiparallel to the cooling field, and the other aligns parallel to the external applied field (or the ferromagnet), independent of the applied cooling field.

The sample is the one used for the x-ray reflectivity experiments by Roy *et al.*[11] The bilayer was grown on a Si(100) wafer with a native oxide. The polycrystalline CoO film was deposited from a Co target by reactive sputtering in Ar and O_2. The polycrystalline Py film was deposited from a $Ni_{0.81}Fe_{0.19}$ target in an Ar atmosphere. The sample was capped with SiO_2. The cross-sectional transmission electron micrograph in Fig. 1 illustrates the boundary between the Py and the CoO. In Fig. 1 the micrograph shows both clear sharp interfaces and more diffuse regions; the latter are possibly the locations of redox reactions.

The resonant soft x-ray reflectivity measurements were carried out at beamline X13A of the National Synchrotron Light Source, Brookhaven National Laboratory. This beamline uses an elliptically polarized wiggler and flips between right- and left-hand circularly polarized x rays (frequency: 22 Hz). The x-ray energy was tuned 2 eV below the L_3 edges of Ni and Co to ensure that the (magnetic) scattering factors always have the same sign relative to the actual magnetiza-

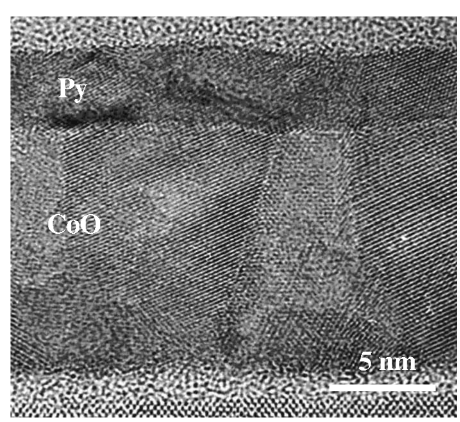

FIG. 1. Cross-sectional transmission electron micrograph image of the Py/CoO bilayer sample capped with SiO_2.

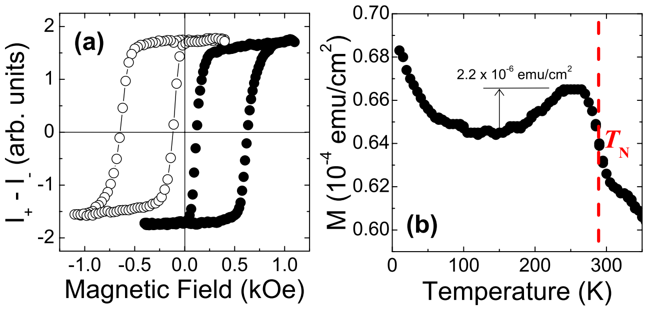

FIG. 2. (Color online) (a) The right-hand and left-hand circular difference signal as a function of magnetic field measured at the Ni L_3 edge on a $SiO_2/Py/CoO/Si$ sample. The measurements are taken at $T=235$ K, with open (closed) points representing positive (negative) field cooling measured at $2\theta=20°$ (10°). The open points have been scaled to ease comparison. (b) Thermoremanent magnetization measurements on the same sample measured using a superconducting quantum interference device (SQUID). The dashed line indicates T_N for CoO.

tion (see Ref. 11 for a discussion of this point), and the reflectivity was measured as a θ–2θ scan. The diffuse scattering background was measured by offsetting θ by 0.2° and then subtracted from the data.

The sample was cooled in a field of +7 kOe from room temperature to 235 K. The magnetic field was parallel to the sample surface along the projection of the x-ray beam on the sample. Figure 2(a) shows the hysteresis loop measured at the Ni L_3 edge by measuring the difference between the specularly reflected intensities at constant 2θ for the two different senses of circular polarization of the incident x rays. These bilayers always display *negative* exchange bias, in which the loop is shifted opposite to the applied cooling field. The reflectivity was measured at the Ni L_3 and Co L_3 edges at saturation after cycling round the hysteresis loop three times. This was repeated after cooling the sample in a field of −7 kOe. There was no indication of a difference in the overall energy line shape of the resonances after cooling in the two directions. Therefore, we conclude that there are no major changes in the electronic environment of the various constituent atoms in the biased state.

Figures 3 and 4 show the reflectivity data for the Ni and Co resonances, respectively. Element-specific magnetization density profiles have been extracted by fitting the data using magnetic scattering theory in the distorted-wave Born approximation (DWBA).[12] The basic model used assumed nominally uniform layers of charge and magnetization density with appropriate chemical and magnetic roughness. This is essentially identical to fitting a variable scattering density profile consisting of many fine slices, but it reduces the computational effort required. The experimental details are similar to those observed at room temperature,[11] and an interfacial layer model is necessary to explain the observed features. The model used for fitting splits the interfacial layer into two parts: one in the Py above the interface and one in the CoO below it. These interfacial layers are principally distinguished from the bulk layers by their differing magnetic properties. The fits are extremely sensitive to the thickness of the interfacial layers but rather insensitive to the associated magnetic roughness. Only the component of magnetization parallel to the projection of the beam onto the sample surface can be sampled; this is along the axis of the applied magnetic field in this experiment.

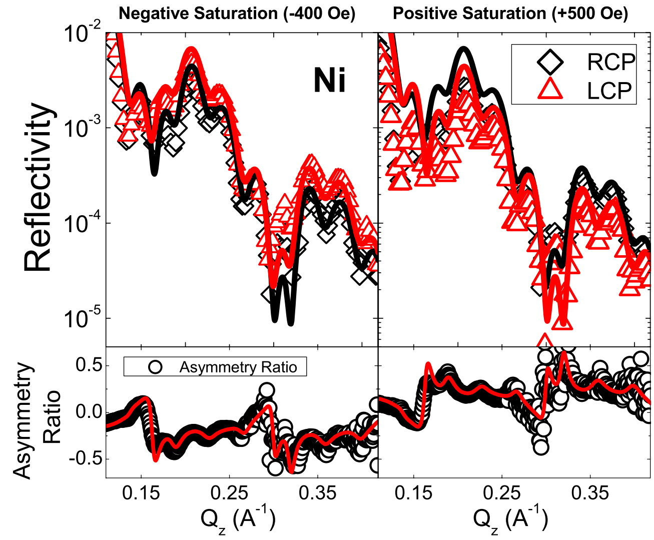

FIG. 3. (Color online) The normalized reflectivity and asymmetry ratio as measured with an incident photon energy 2 eV below the Ni L_3 edge at $T=235$ K in the biased state. The sample has been cooled in a field of −7 kOe. The reflectivity was measured at positive and negative saturations. For low Q_z there is a systematic error. The lines are fits to the model described in the text and Fig. 5. The reflectivity scans have been decimated for clarity.

Reflectivity data at the Ni L_3 resonant edge were taken for both positive and negative saturation fields after cooling the sample to 235 K from room temperature in a field of −7 kOe (Fig. 3). The lower panels show the asymmetry ratio, which is proportional to the in-plane magnetization of Ni atoms in

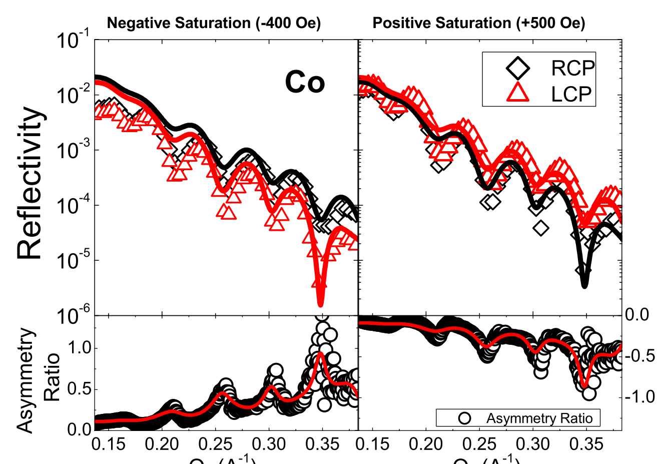

FIG. 4. (Color online) The normalized reflectivity and asymmetry ratio as measured with an incident photon energy 2 eV below the Co L_3 edge at $T=235$ K in the biased state. The sample had been cooled in a field of +7 kOe. The reflectivity was measured at positive and negative saturation. For low Q_z there is a systematic error. The lines are fits to the model described in the text and Fig. 5. The reflectivity scans have been decimated for clarity.

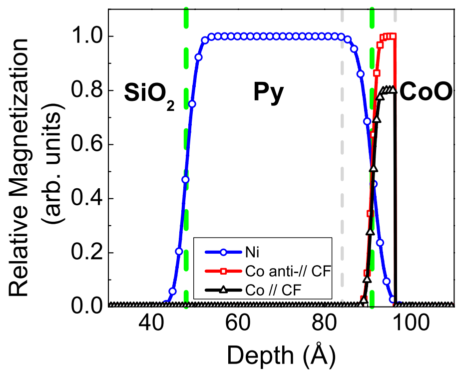

FIG. 5. (Color online) Magnetic density profile for the models described in the text. The magnetizations for both elements are normalized to 1. The dashed green lines are the chemical interfaces from Table I. The short-dash gray lines delineate the interface layers described in the text. Ni (blue circles) reverses completely on switching the field to the measured accuracy. Co magnetization parallel (antiparallel) to the cooling field direction is indicated by black triangles (red squares).

the sample.[13] Magnetization is confined to the permalloy layer, which is split into two magnetic layers: the bulk and an interfacial layer 6.9 Å thick. The magnitude of the Ni magnetization in this layer is slightly smaller than that in the bulk part. The best fit to this saturation magnetization density depth profile model is illustrated in Fig. 5, which shows the magnetization density parallel to the applied field for both positive and negative directions of saturation. The magnetization in the Py layer starts to decrease before the Co magnetization starts to appear; this could reflect different out-of-plane magnetic correlation length scales for the Py and CoO. The Py moments are affected by the presence of the interface over a larger distance.

On cycling around the hysteresis loop, the Ni magnetization reverses completely, within the measured accuracy, in both the bulk and the interfacial layer. Least-squares fitting indicates that the magnetization difference in the interfacial layer is $(1 \pm 3)\%$, indicating that any net pinned Ni magnetization in this interfacial layer must represent a fraction smaller than 1% of the total moments in the Ni interfacial layer. Assuming that in this layer the number density of atoms is the same as in the bulk layer and that FM moment is only pinned parallel to the applied field, this leads to the conclusion that less than 0.3% of the Ni moments in the entire sample is pinned. If the Fe moments follow Ni perfectly, this represents the total fractional amount of potential pinned (Ni,Fe) moments.

The Co resonance data indicate the net magnetization along the applied field axis of the Co moments (Fig. 4). The Co moments in the antiferromagnetic CoO layer have no net magnetization in the direction probed. However, a net magnetization is observed in an interfacial layer of thickness 5.4 Å (see Fig. 5). This magnetization does not reverse completely; there is a pinned component. Although most of the Co magnetization follows the magnetic field, the net pinned magnetization is aligned *antiparallel* to the cooling field. If the interfacial layer is assumed to have the same thickness in both saturation conditions, the magnitude of the magnetization is $(20 \pm 2)\%$ lower in the direction parallel to the cooling field (Fig. 5), indicating that 10% of the Co magnetization is pinned antiparallel to the cooling field.

TABLE I. Thicknesses and roughnesses for the sample, as obtained from Cu $K\alpha$ x-ray reflectivity data. The roughness quoted corresponds to the upper surface in each case. SLD refers to the scattering length density—the scattering length of a formula unit divided by the volume associated with that unit. ρ refers to the corresponding mass density. The standard bulk mass density is provided for reference. These values are used as fixed parameters in the soft x-ray reflectivity calculations.

Layer	Thickness (Å)	Roughness (Å)	SLD (10^{-6} Å^{-2})	ρ (g/cm^3)	ρ_{bulk} (g/cm^3)
SiO_2	48	5.4	6.8	2.09	2.53
Py	43	3.1	20.7	7.52	8.25
CoO	145	2.9	16.1	5.77	6.45
Si	∞	3.5	10.0	3.08	2.33

An estimate of absolute magnitudes can be made from the available data. Roy *et al.*[11] found a value of 0.53×10^{-4} emu/cm^2 for the Co interfacial layer magnetization for this sample at room temperature in the unbiased state (no pinned component was observed). If we assume the same magnetization at low temperature, the pinned Co moment in the biased state is 5.3×10^{-6} emu/cm^2. In support of this value, Abrudan *et al.*[14] estimated from x-ray magnetic circular dichroism studies on Fe/CoO bilayers that the uncompensated Co spins fill 1.1 monolayer (ML) if arranged together. The magnetic moment of Co^{2+} in bulk CoO is $3.8\mu_B$.[15] Assuming that this value applies to the Co in the thin film, Abrudan *et al.* find 0.4×10^{-4} emu/cm^2 of uncompensated moment,[16] or 45% of the Co moments in the 0.69 nm interfacial layer, of which 10% is pinned. If the Co is metallic, the magnetic moment is ~2 times smaller.

On warming through the T_N of CoO, these pinned moments unpin, with an accompanying increase in magnetization in the cooling field direction. Thermoremanent magnetization was measured in zero field after cooling to 10 K in 3 T [Fig. 2(b)]. A small peak of magnitude 2.2×10^{-6} emu/cm^2 is present close to T_N; this is the right order of magnitude to account for this increase.

Takano *et al.*[17] found that 1% of the moments at polycrystalline CoO interfaces is uncompensated. This is also found in calculations placing defects at FM/CoO interfaces with spin-flop coupling,[18] in which the pinned Co magnetization is antiparallel to the cooling field, and in Monte Carlo studies of FM-AFM interfaces.[19]

Since the net *pinned* Co magnetization lies antiparallel to the cooling field, it is possible that the pinned Co moments themselves lie antiparallel to the cooling field. In FM/FeF_2 bilayers there is a superexchange coupling mechanism across the interface with some FM magnetization pinned parallel to the cooling field. This mechanism allows for the establishment of positive exchange bias in large-enough cooling fields.[6,19,20] We see no clear evidence for this FM pinning in Py/CoO but possible mechanisms might include superexchange in a ferromagnetic or antiferromagnetic oxide. To date, positive exchange bias has not been observed in Py/CoO.

A second possibility is that the interfacial Co magnetization is in a canted perpendicular spin-flop state relative to the FM, yielding a net Co magnetization pinned *antiparallel* to the cooling field. Such a spin-flop state has been observed by polarized neutron diffraction in epitaxial Co/CoO with (111) planes parallel to the interface.[21] Schulthess and Butler[18] considered this coupling mechanism for a compensated, i.e., rough, interfacial (111) CoO plane, and found that for perfect single-crystal interfaces of FM/CoO, there is no net pinned magnetization and no exchange bias. However, if lattice defects are present, they found that an energy barrier is created, with net canted Co magnetization pinned antiparallel to the cooling field, producing negative exchange bias. In a large-enough cooling field, positive exchange bias could again be forced. The amount of uncompensated AF magnetization is estimated to be 1% of the moments in a CoO monolayer. From the estimates above, the pinned magnetization in the interfacial layer is 4.5% of the total potential magnetization; this yields ~2% for a single monolayer, which is in reasonable agreement with the Schulthess and Butler model.

The significant amount of unpinned Co magnetization that follows the applied field should be ferromagnetically coupled to the FM. We note that this is different from the behavior of the unpinned spins seen in FM/FeF2 bilayers,[6] in which the net unpinned magnetization is antiferromagnetically coupled to the FM. Van Lierop *et al.*[22] have previously noted that only a portion of the uncompensated magnetization in the AF appears to be implicated in the formation of exchange bias. This unpinned Co magnetization is produced by reduced interfacial Co cations, as described by Regan *et al.*[23]

In conclusion, there is an intermediate layer at the interface of the Py and the CoO, and the exchange bias in this sample is generated there. The Co magnetization consists of two independent components: the majority follows parallel to the applied field, and a small fraction is pinned antiparallel to the cooling field used to bias the sample. The Py magnetization shows no positive evidence for pinning, and therefore we conclude that the exchange coupling between the pinned CoO and the FM is sufficient to induce the observed exchange bias without any pinning of Py during the cooling process.

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