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Combined magnetic X-ray and polarized neutron reflectivity study of the origins of exchange bias in the Co/FeF_2 system

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Abstract

We discuss studies of the magnetic specular reflectivity of neutrons and X-rays from the exchange bias system consisting of a single crystal film of antiferromagnetic FeF_2 capped with a ferromagnetic Co film cooled in an applied magnetic field below the T_N of the FeF_2. This system exhibits a shift of the magnetic hysteresis loop along the direction of the cooling field H_c (positive exchange bias) or in the opposite direction (negative exchange bias) depending on the magnitude of the cooling field. The use of neutrons with polarization analysis enables the spatial distribution of different vector components of the magnetization to be determined, and the use of resonant magnetic X-ray scattering enables magnetization in a compound system to be determined element-selectively. Our results show that the coupling across the interface between the relatively few uncompensated Fe spins and the more numerous Co spins is antiferromagnetic. In a large cooling field which overrides this coupling, the Fe spins are oriented along H_c and some get pinned in this direction, in turn pinning the Co spins above the interface oppositely and thus creating positive exchange bias. For sall values of H_c the Fe spins get locked in the opposite direction producing negative exchange bias. In addition a significant fraction of the Fe spins at the interface are unpinned and always align opposite to the Co magnetization.

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1. Introduction

We present here a brief account of some recent work done on the problem of exchange bias, using both resonant magnetic X-ray reflectivity and polarized neutron reflectivity. The combined use of these powerful tools to study magnetism at buried interfaces can yield considerable insight into the fundamental mechanisms for exchange bias. It is appropriate to present these results in these proceedings, as the Zabel group has contributed importantly in this area using similar techniques [1–3].

Exchange bias (or exchange anisotropy) [4–8] is known to arise when a ferromagnetic (F) film is in contact with an antiferromagnetic (AF) film which has been cooled below its Neel temperature in an applied magnetic field, known as the "cooling field" (H_{fc}), which is then removed. The direction of the cooling field is henceforth defined as the positive x-direction in this paper. The exchange interaction between the F and AF spins across the interface results in a shift of the magnetic hysteresis loop of the F layer, usually in a direction along the magnetic field axis opposite to that of the cooling field H_{fc} (negative exchange bias) although in certain cases it can also cause a shift along the direction of the cooling field (positive exchange bias). This shift causes the magnetization of the F film to be "locked" in a certain direction for relatively weak applied fields, and the effect is exploited in many current and potential device applications, such as the read heads used with magnetic disc drives, random access memories, etc.

However, the details of how this comes about have been the subject of considerable work and controversy over the last decade or so, partly because detailed information about the magnetism at the interface has not been easily forthcoming. In this paper, we apply the techniques of resonant X-ray magnetic reflectivity and polarized neutron reflectivity to study the magnetic depth profiles around the interface of a polycrystalline Co film deposited on an epitaxially grown single crystal film of FeF_2. The T_N of FeF_2 is 78.4 K. This system exhibits negative exchange bias in relatively weak cooling fields, but shows positive exchange bias when the cooling field is large [9,10]. The crossover appears to depend on the roughness or other defect properties at the interface, and in some cases compound hysteresis loops, indicative of both positive and negative exchange bias are seen.

A key assumption of the theory of exchange bias is the existence of *pinned* net magnetization in the antiferromagnet or at the F/AF interface. The existence of this magnetization has been demonstrated in recent experiments [10–13]. Positive exchange bias is commonly thought to result from antiferromagnetic exchange coupling between unpinned and pinned spins. Nogues et al. [9,10] explained the occurrence of positive exchange bias in this system for large cooling fields in terms of a model that postulated pinned Fe spins at the interface antiferromagnetically coupled to the Co spins across the interface. Our present results confirm the antiferromagnetic interaction between the Co and Fe spins across the interface but yield a more complex picture involving transition layers on either side of the interface. Antiferromagnetic exchange coupling favours opposite (antiparallel) alignment of the coupled spins, while ferromagnetic exchange coupling favours the same (parallel) alignment. By identifying where the magnetization is pinned and the alignment of the unpinned magnetization with respect to the pinned magnetization as a function of field, the sign of the exchange coupling, either ferromagnetic or antiferromagnetic, can be inferred. Thus, key to understanding the origin of exchange bias is the measurement of pinned and unpinned magnetization depth profiles.

2. Experimental results

Here we describe a set of resonant magnetic X-ray reflectivity and polarized neutron reflectivity experiments on the model exchange bias system consisting of a polycrystalline ferromagnetic Co film on top of an antiferromagnetic FeF_2 film grown epitaxially on single crystal MgF_2. Exchange bias samples were prepared by sequential electron beam evaporation of FeF_2, Co and Al (the last to serve as a capping layer) at a deposition rate of 0.05 nm/s onto (110) oriented single crystal MgF_2 polished substrates measuring 10 mm by 10 mm. The deposition temperatures were 300 °C for the FeF_2 layer and 150 °C for the Co and Al layers. The chemical structure of the sample was determined from X-ray reflectometry. The thickness and chemical roughnesses for the layers determined by fitting to the Cu Kα reflectivity data were kept as constraints for fitting the magnetic X-ray and neutron reflectivity data. The resonant magnetic X-ray scattering experiments were carried out at Beamline 4 of the Advanced Light Source at Lawrence Berkeley National Laboratory, and the polarized neutron reflectivity experiments were done on the ASTERIX spectrometer at the Manuel Lujan Neutron Scattering Facility at Los Alamos National Laboratory. The first experiments were done on samples with positive exchange bias prepared by cooling in fields of approximately 1 T.

Since resonant X-ray magnetic scattering (RXMS) [14–16] is element-selective, we can use it to determine the magnetization in the FeF_2 and the Co separately. The results for the first set of resonant magnetic X-ray reflectivity and neutron reflectivity data (carried out on a sample which exhibited positive exchange bias) have been published earlier [9] and are summarized below.

The resonant X-ray magnetic reflectivity data were taken at low temperatures in a field cooled sample at applied fields corresponding to “saturation” along the positive and negative directions respectively (or more accurately beyond the point where the magnetic hysteresis loop closed for both directions of applied field). Reflectivity was measured as a function of q_z for each sense of the circular polarization of the incident beam. An analysis of the reflectivity data using the DWBA formalism for resonant magnetic X-ray scattering [17,18] showed that there were indeed two kinds of uncompensated magnetization in the FeF_2 sample, namely those arising from pinned Fe spins (which did not respond to the applied field) and those arising from unpinned Fe spins, and that the latter were coupled antiferromagnetically to the Co spins which aligned along the applied field, i.e. along the x axis, so that the Fe unpinned spins aligned along $-x$, i.e. antiparallel to the applied field. The unpinned Fe spins existed mainly in a layer of thickness $\sim$2 nm below the Co/FeF_2 interface. The analysis also showed that there was a decreased magnetization of the Co spins in a layer of thickness $\sim$3 nm above the Co/FeF_2 interface along the x-axis, indicating that some Co spins were pinned near the interface in the negative direction (see Fig. 1).

The polarized neutron reflectivity (PNR) data, which measured the depth profile of the total magnetization was taken after field cooling the sample, and then rotating the sample by 90° and then applying a field along the original field cooling direction and measuring the spin–flip and non spin–flip neutron reflectivities. The non spin–flip component is determined by the component of the total magnetization in the rotated sample which is now along the applied field direction, and thus measures the polarizable or unpinned component of the magnetization, while the spin–flip component is determined by the magnetization which is perpendicular to this direction and thus pinned in the rotated sample (although it should be noted that both pinned components parallel and antiparallel to the original cooling field direction contribute). The PNR data established that the magnetization in the Co layer away from the Co/FeF_2 interface was in the direction of the applied field and then the in-plane averaged magnetization formed a distorted spiral near the interface as the magnetization on the FeF_2 side tried to become antiparallel to the co magnetization

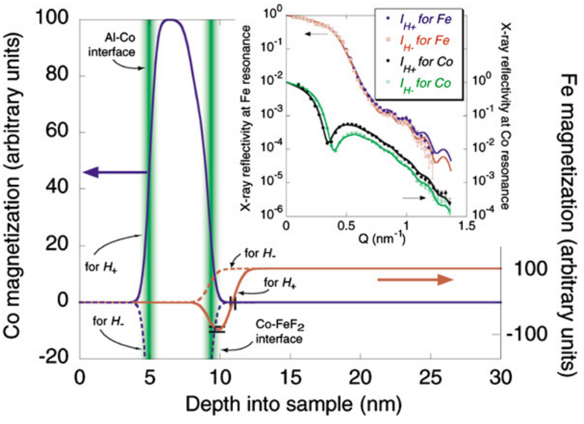

Fig. 1. Co magnetization profile (blue) and Fe magnetization profile (brown) for Co/FeF2 system at low temperature with positive exchange bias, as obtained with RXMS for both directions of applied field. Inset shows individual reflectivity curves for both senses of circular polarization and at the photon energy tuned to L-edge resonances in Fe (top curves) and for Co (bottom curves). The latter reflectivity curves have been shifted down for clarity. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

above, confirming the antiferromagnetic coupling across the interface obtained from the RXMS data. Since the spin–flip component of the PNR showed Kiessig fringes characteristic of the total thickness of the FeF_2 layer, it established that for this sample the pinned magnetization existed throughout the bulk of the FeF_2 film and not just at the interface. It also established absolute values for the magnetization, which indicated that approximately only ∼6% of the Fe spins were pinned. It further established that there were spins on the Co side of the interface that were pinned antiparallel to the Fe spins across the interface, and that the unpinned Fe spins existed only in the vicinity of the Co/FeF_2 interface, in agreement with the RXMS data.

Since the publication of the above results, we have carried out new studies of the PNR on new samples of Co/FeF_2, under conditions of both positive and negative exchange bias. It should be emphasized that PNR measures only the depth profile of the total magnetization, without distinguishing between the components coming from the Co or the Fe. Details of the measurements and the analysis will be published in a separate publication [19].

After cooling the sample appropriately so that it had exchange bias, an external field was applied at low temperatures sufficient to saturate the sample in both directions. By measuring the depth profile of the magnetization for each case, the PNR results yielded the depth profiles of both the pinned and unpinned spins in the sample. In Fig. 2, we show schematically the pinned and unpinned magnetization in the FeF_2, the interface region, and in the Co for positive and negative applied fields for both the positive and negative exchange bias samples at low temperatures. The separate magnetization contributions from the Co and Fe spins in the interface region have been inferred by combining the results from both the RXMS and neutron reflectivity data. It is the pinned Fe spins at the Co/FeF_2 interface which give rise to the exchange bias in this system.

The magnetization direction in the interface region is dominated by the direction of the majority of the Co spins which are ferromagnetically coupled to the spins in the rest of the Co layer, but one should remember that in the interface layer, there is a smaller amount of magnetization due to the Fe which is oppositely directed to the Co spins, as shown in Fig. 2. Thus, by examining the profiles in Fig. 2 and combining it with the results of the earlier RXMS

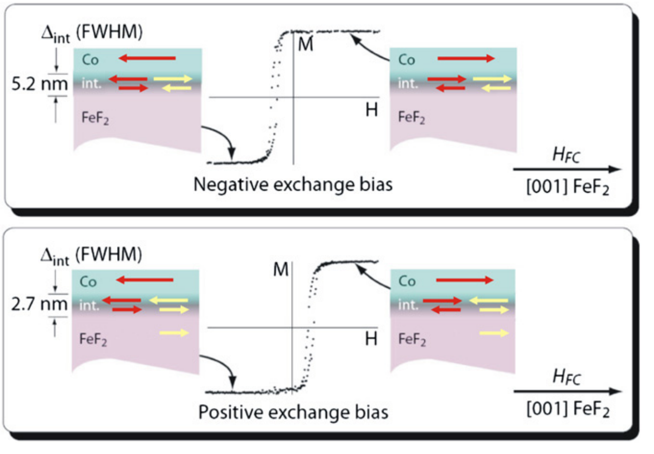

Fig. 2. Schematic diagram showing the alignment of the magnetization of the *unpinned* Co spins in the bulk of the Co film and of the unpinned Co and Fe spins on either side of the FM/AF interface (red arrows) and the *pinned* interfacial Co and FeF_2 layer magnetizations (yellow arrows) at extremes of applied field for the cases of $-H_E$ (upper) and $+H_E$ (lower) obtained from analysis of the neutron data [19]. For the positive exchange bias case, pinned Fe spins were also observed in the bulf of the FeF_2 layer, but not in the negative exchange bias case. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

and neutron results, we are led to propose a model for exchange bias in this system discussed in the next section.

3. Model and discussion

Basic assumptions

A: In FeF_2 there are both pinned and unpinned uncompensated spins even below T_N. Above T_N all spins are unpinned. (Although as T_N is approached there will be developing fluctuating short-range antiferromagnetic order amongst the Fe spins). The Co spins are all coupled to each other with a ferromagnetic exchange.

B: The Fe spins and Co spins are coupled across the interface by an *antiferromagnetic* exchange interaction J_{AF} (presumably due to superexchange through the F termination layer — this makes this specific to the Co/FeF_2 system and may not apply to other exchange biased systems, especially polycrystalline ones).

Model

Large H_{fc}

(1) At room temperature, H_{fc} is applied along x. If H_{fc} is large enough, it can override J_{AF} and induce a magnetization in both the Co layer and the FeF_2 layer parallel to the x axis. The magnetization in the FeF_2 layer can be significant because of the large field but smaller at the interface due to the inhibiting effect of J_{AF} since the Co is also magnetized parallel to x.

(2) The sample is cooled below T_N. The FeF_2 develops long range AF order but adjusts its domain structure to keep the majority of its uncompensated spins still pointing along x to minimize the free energy in the cooling field. Depending on where these spins are, a certain fraction (but not all) of the Fe spins get pinned in this direction, i.e. along $+x$, in the bulk of the FeF_2 and also at the interface.

(3) The cooling field is switched off. The Co spins are now basically unpinned but ferromagnetic with no preferred direction to point. The ones at the interface feel J_{AF} if they adjoin the pinned Fe spins at the interface, and thus want to switch in the $-x$ direction. In doing so, they carry the rest of the Co spins with them. (So zero applied field would give negative magnetization).
(4) If a field is now applied along $+x$, but is not large enough to override J_{AF}, at least some of the Co spins at the interface will be locked in the $-x$ direction, i.e. those adjoining the pinned Fe spins (probably forming a domain structure determined by the interface roughness) and because of their ferromagnetic interaction with the rest of the Co spins will *cause a positive exchange bias*. The ones adjoining the unpinned Fe spins will orient and force the unpinned Fe spins to orient oppositely. Thus a complementary domain structure of Co and Fe spins will be formed at the interface.

This would account for the fact that the magnetization of the Co layer appears to decrease from saturation as one approaches the interface (as obtained from both neutron and X-ray results). It explains positive exchange bias for large cooling fields.

Small H_{fc}

(1) At room temperature, there is very little polarization in the FeF_2 layer. The Co spins respond to the H_{fc} field and orient along $+x$. At the interface, J_{AF} can now override the field and causes the Fe spins at the interface to orient in the $-x$ direction.
(2) When the sample is cooled below T_N a fraction of these Fe spins get pinned in the $-x$ direction. When the cooling field is switched off, because of J_{AF} they lock the adjoining Co spins in the $+x$ direction. Again a complementary domain structure is formed at the interface but opposite to the one for large H_{fc}.
(3) These Co spins at the interface locked in the $+x$ direction give rise to *negative exchange bias* because of the ferromagnetic interaction between Co spins.

Thus, a combined RXMS and PNR specular reflectivity study has enabled us to flesh out a model which can account for many of the properties manifested by the Co/FeF_2 exchange bias system. Certain questions regarding the microscopic details remain to be answered, however. These include questions such as where the pinned and unpinned spins in the FeF_2 lie; what the exact lateral domain structure is on either side of the interface and how it correlates with the interface roughness; and how interface roughness determines the domain size and the distribution of J_{AF} at the interface. These can only be answered via off-specular scattering experiments, which are currently underway.

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