Exchange Bias in Spin-Engineered Double Superlattices

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Exchange bias has been observed in sputtered magnetic double superlattices which consist of a ferromagnetically coupled superlattice grown on an antiferromagnetically (AF) coupled superlattice. This system exhibits a parallel domain wall, a spin flop transition, and exchange bias when the anisotropy is large in the AF block. This work shows that neither the domain wall nor the spin flop are directly related to exchange bias but that the anisotropy is essential.

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The interaction between a ferromagnet (FM) and an antiferromagnet (AF) in atomic proximity has confounded researchers for over 40 years. The hysteresis loop $M(H)$ of a ferromagnet is generally centered at the origin with $M(H) = -M(-H)$ (where M is the magnetization and H is the externally applied field) thus satisfying time reversal symmetry. In an AF/FM system with a suitable AF the loop is offset from zero applied field, i.e., it exhibits unidirectional anisotropy. Such a phenomenon is known as exchange bias and was first discovered by Meiklejohn and Bean [1] in 1956. The importance of this discovery is manifested in spintronic devices known as spin valves [2]. Despite intense research the origin of exchange bias is still not fully understood.

Two theories have been quite successful in explaining the observed value of the exchange bias H_{ex}. Malozemoff proposed that the roughness at the interface sets up a random exchange field at the interface [3]. Depending on the anisotropy in the antiferromagnet, it becomes energetically favorable for the antiferromagnet to break up into domains with perpendicular domain walls. Alternatively, Mauri *et al.* [4] have argued that even at a very smooth interface, the observed exchange bias can be explained by parallel domain walls in the antiferromagnet, i.e., a spiraling spin structure formed in the AF layer which spreads the exchange energy into the domain wall. Recent theoretical work is in support of a spiral spin structure [5]. Experimentally, Yang and Chien [6] have used a model of a spiral spin structure in the AF layer to explain the behavior of the hysteresis loops from $Co/FeMn/Ni_{0.8}Fe_{0.2}$ trilayers.

However, due to the difficulties associated with measuring the spin configuration in the buried AF, there has been no experimental evidence to support the idea of parallel domain walls, nor the role of anisotropy in biasing. In particular, the anisotropy in the thin film AF layers is assumed to be large but reference is solely made to neutron diffraction measurements of bulk AF materials [7]. In this Letter we shall provide unequivocal evidence for the role of anisotropy in exchange bias. To this end we have grown artificially layered structures which consist of superlattices of Co and Ru. The spins in the magnetic layers can be tailored to be antiferromagnetically or ferromagnetically coupled, depending on the thickness of the Ru [8]. In order to mimic an exchange biased system, an artificial FM has been grown next to an artificial AF in the same manner as previous work on epitaxial Fe/Cr double superlattices [9]. The resulting samples had the nominal layer sequence: Si (001)/Ta (75 Å) [Co (35 Å)/Ru (15 Å)]$_{\times 9}$ Co (35 Å)/ Ru (15 Å)[Co (60 Å)/Ru (10 Å)]$_{\times 10}$/Ta (75 Å) where the first (second) superlattices were coupled antiferromagnetically (ferromagnetically). Such a system will be a suitable model for a very smooth AF/FM interface with near perfect coupling where layers of Co represent layers of atomic spins in a real system. Because of the ideal nature of the interface, it is a good system in which to test the Mauri model.

The films were prepared by dc magnetron sputtering in a chamber with a base pressure of 10^{-8} Torr. The working gas was Ar at a pressure of 2.5 mTorr. A 200 Oe field was applied in the sample plane during deposition. Typical deposition rates were 4 Å/s. The layer thicknesses and interfacial roughness (typically <3 Å) were determined by low angle x-ray reflectivity. The magnetization loop for the sample described above is shown in Fig. 1. The clean switching of the ferromagnet is clearly identifiable with a more complicated spin configuration for fields away from the coercive field. Since all of the magnetic layers are saturated at the extremities of the cycle, no exchange bias would be expected to be observable. An analysis of the individual AF component of the superlattice yielded a strong exchange coupling constant of -0.25 mJm^{-2}.

To understand the magnetic behavior, one can construct a simple Stoner-Wohlfarth-type model [10] by minimization of the areal energy density defined by

$$\varepsilon = -\sum_i [\mu_0 m_i H t_i \cos(\theta_i) + J_{i,i+1} \cos(\theta_i + \theta_{i+1}) + k_i t_i \cos^2(\phi_i)], \qquad (1)$$

where the terms are the Zeeman, exchange, and anisotropy energies, respectively. In more detail, t_i is the layer

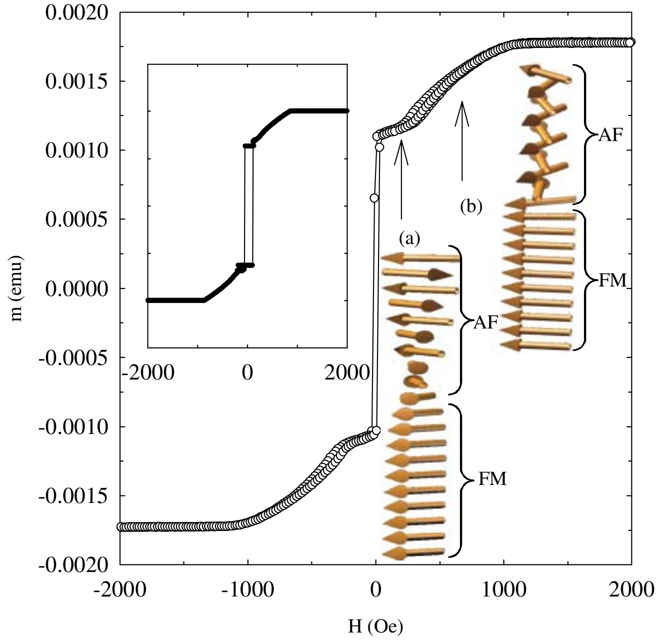

FIG. 1 (color online). The room temperature magnetization loop of the sample described in the text (open symbols). The inset diagrams show the calculated magnetization loop (closed symbols) and the calculated magnetic structure for the two fields indicated by the upwards pointing arrows. The states shown are the parallel domain wall (a) and the spin flop phase (b). The magnetic field is parallel to the moments of the ferromagnetic block.

thickness, m_i the magnetization, k_i the uniaxial anisotropy, θ_i is the angle of the average magnetization to the field H, and ϕ_i is the angle of the magnetization to the anisotropy axis of each layer i. In addition, $J_{i,i+1}$ is a measure of the exchange energy between the layers i and $i+1$. Because of the shape anisotropy the moments will be confined to the plane of the layers. All parameters were deduced from conventional magnetometry. The small anisotropy constant was $k = 1 \times 10^3 \mathrm{Jm}^{-3}$. The results of the calculation for the field along the easy axis are depicted in Fig. 1. Before saturation of the whole superlattice stack shown in Fig. 1, two distinct states are observed. Just after the saturation of the FM layer a Mauri type domain wall (see Fig. 1) is formed in the AF part of the stack. In the calculation, the domain wall does not significantly spread into the FM superlattice. Following this, it becomes energetically favorable for the spins in the AF to order themselves in a spin flop phase. In this phase, the exchange coupling between the antiferromagnetic spins is still strong enough to overcome the Zeeman term, such that most of the spin components are perpendicular to the externally applied field. After a further field is applied, the exchange energy succumbs to the Zeeman term and all the moments point in the direction of H.

To obtain depth dependent vector magnetometry information, we have performed polarized neutron reflectometry (PNR) measurements with polarization analysis. These artificial structures are ideally suited for such measurements [11,12]. In addition to the structure, neutrons are sensitive to magnetism through the interaction of the neutron magnetic moment with the magnetic induction inside the superlattice [13]. As the neutron is a spin-$\frac{1}{2}$ particle, there are two spin eigenstates. Components of the magnetic induction parallel to the neutron spin influence the potential the neutron experiences but do not change its spin eigenstate and are referred to as nonspin flip scattering (NSF). Components of the magnetization orthogonal to the neutron spin polarization result in a purely magnetic change in potential which can flip the spin state of the incident neutron, so-called spin flip scattering (SF).

PNR was performed on the CRISP reflectometer at the ISIS spallation source, Rutherford Appleton Laboratory [14]. Experiments were performed in time-of-flight mode. Specular reflectivity was thus measured for a Q range of 0.01–0.2 Å^{-1}. To obtain field dependent information, it was sufficient to obtain the overall scale factor and then to concentrate on the relevant Q range. The resolution of the perpendicular momentum transfer $\Delta Q/Q$ was 4%.

In Fig. 2, three reflectivities are shown for various values of the applied magnetic field. At saturation, shown in Fig. 2(a), all the Co spins are collinear with the applied magnetic field leading to NSF scattering and no AF ordering ($Q_{\mathrm{AF}} \approx 0.054$ Å^{-1}). Instead, two peaks can be seen in the nonspin flip channel. The first ($Q_{\mathrm{nucFM}} \approx 0.091$ Å^{-1}) is due to the chemical periodicity of the ferromagnetically coupled superlattice, and the second ($Q_{\mathrm{nucAF}} \approx 0.108$ Å^{-1}) arises from the antiferromagnetically coupled section of the superlattice. There is little SF scattering above the background level. Reducing the field to 600 Oe produces the spin flop phase, shown in Fig. 2(b), which has an AF component of spins orthogonal to the applied magnetic field therefore giving rise to SF scattering which dominates the specular reflectivity at the position of the half order peak (Q_{AF}). Reducing the field further to 150 Oe produces the scattering shown in Fig. 2(c). At the AF peak position there is still significant SF scattering but the NSF scattering is now dominating. We can therefore conclude that there is again an AF periodicity but with components parallel and perpendicular to the applied magnetic field. The exchange coupling with the FM layers means that the AF moments closest align antiferromagnetically with the FM layers but this relaxes to be orthogonal to the FM layers to minimize the Zeeman energy. The spin directions are fairly evenly distributed over 360° in the width of the AF leading to weaker SF scattering at the AF peak than in the spin flop case.

To analyze the PNR data the structure of the superlattices was fixed by an analysis of the x-ray data. A bulk moment was assumed for the Co of 1.7 μ_{B}/atom. The neutron reflectivities were then fitted using the fully dynamical computer code POLLY [15], employing a simulated annealing algorithm to vary solely the magnetization

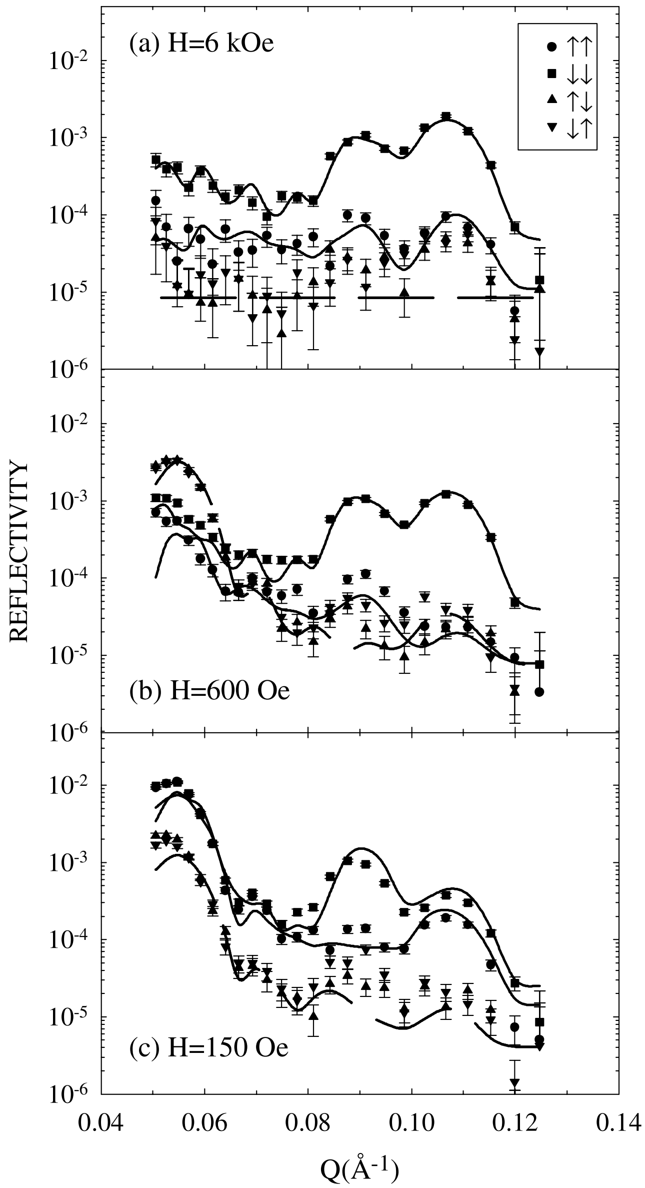

FIG. 2. PNR data at three different applied magnetic fields. The legend refers to the incident (outgoing) spin state of the neutron beam. The solid (and dashed lines) are the result of the calculations for the NSF (and SF) scattering described in the text.

direction in each Co layer. The calculated spin arrangement from the mean field calculation was used as a seed for the Co layer dependent spin configuration. The variation in the moment direction from the calculated arrangement after the simulated annealing was less than 10° in each layer. Bulk values for scattering length densities were assumed in agreement with the x-ray analysis. The fits shown as the solid and dashed lines in Figs. 2(a)–2(c), are the results of the calculation from a saturated film, a spin flop phase, and a parallel domain wall as shown in Fig. 1. The good agreement with the experimental data is self-evident confirming that the calculated spin configuration is a good description of the true one. In the analysis of both the PNR data and the calculations the majority of the spin reorientations between saturation of the FM and saturation of the whole stack happens in the AF layer in agreement with the theoretical work of Mauri *et al.* [4].

Performing minor loops (viz. a loop where only the FM layers are switched), both experimentally and in the calculations, yields no exchange bias. As a result of this it can be concluded that neither the spin flop phase nor the parallel domain wall give rise to exchange bias. In a real exchanged bias system the AF layer has a substantial anisotropy—such as Co/CoO ($k_{\mathrm{CoO}} \approx 5 \times 10^5\ \mathrm{Jm}^{-3}$) [1,16], or $\mathrm{Ni}_{0.8}\mathrm{Fe}_{0.2}$/FeMn ($k_{\mathrm{FeMn}} \approx 1.35 \times 10^4\ \mathrm{Jm}^{-3}$) [17]. Even a comparatively low k AF material, such as NiO, has a substantially higher anisotropy than the FM it is pinning. A large anisotropy in the AF part of the superlattice can be generated by the addition of a suitable dopant. Such a dopant is Pt, which forms a continuous series of solid solutions with Co [18] and has a large magnetocrystalline anisotropy [19–21]. This has been tested with magneto-optical Kerr Effect (MOKE) measurements and coercivities of the order of 200 Oe for plain CoPt films have been measured (cf. 20 Oe for pure Co). From a simple Stoner-Wohlfarth model [10] of a single domain state this corresponds to an anisotropy of $k \approx 1 \times 10^5\ \mathrm{Jm}^{-3}$. To test the role of anisotropy in exchange bias, a further set of samples were sputtered where the magnetic layers within only the AF stack were doped with Pt. Given that the domain wall does not penetrate significantly into the FM, a single layer of Co (which is too thin to contain a domain wall) was grown on top of the doped AF superlattice. The exact sequence of layers was as follows: Si(001)/[($\mathrm{Co}_{1-x}\mathrm{Pt}_x$)(61 Å)/Ru(10 Å)$]_{\times 9}$/($\mathrm{Co}_{1-x}\mathrm{Pt}_x$)(61 Å)/Ru(5 Å)/Co(56 Å)/Ru(19 Å) with $x \sim 0.25$. This CoPt composition is consistent with the magnetization of the sample as determined by vibrating-sample magnetometry (VSM) [22]. The major hysteresis loop is shown in Fig. 3(a). Minor loops were measured by MOKE to increase the sensitivity to the pure Co layer, as the laser does not penetrate significantly into the AF stack. These are shown as insets to Fig. 3(a) after saturation of the sample in a 1.5 T forward and reverse field. Here $\mathrm{H}_{\mathrm{ex}} \approx 200$ Oe, and it is set in either direction by the saturating field setting up the AF state in the CoPt/Ru.

Representative PNR data are shown in Fig. 3(b) for an applied field of 35 Oe on the positive going branch of the major loop. Again the curves are the result of the fitting methodology previously described. Clearly, a distribution of magnetic domains could produce the observed ratios of SF and NSF scattering. However, off-specular measurements showed an absence of diffuse scattering which would be indicative of a domain structure [23]. Moreover, we note that the splitting of the critical reflection wave vector for the two neutron spin states and the apparent shift in Q_{AF} (a dynamical effect due to a change in refractive index) are both consistent with a single domain

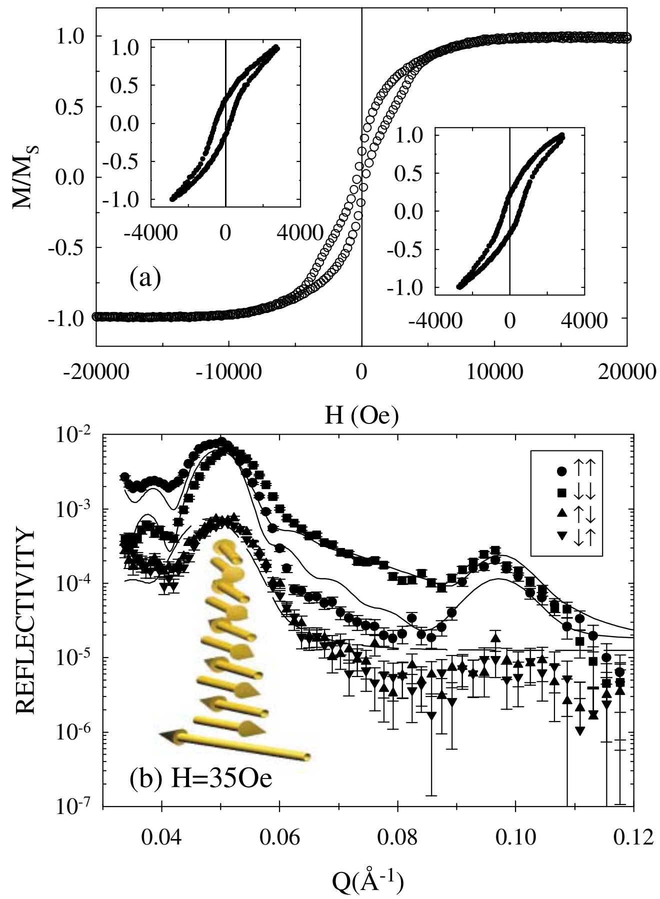

FIG. 3 (color online). (a) The magnetization loop as measured by VSM (open symbols). The insets present the exchange biased minor magnetization loops measured by MOKE for opposing directions of the bias set by application of a 1.5 T saturating field. The exchange bias field is ≈ 200 Oe. (b) The PNR data for an applied field of 35 Oe. The inset shows the derived spin structure.

state [24]. As the external field is decreased from saturation we obtain a parallel domain wall but that no spin flop phase is observed. In this case, we are in the domain wall state described by Mauri *et al.* after reversing a pinned layer. Since the moments would be pointing at nearly 90° to the easy axis it is not energetically favorable to form a spin flop phase due to the large anisotropy.

In conclusion, energy minimization calculations, MOKE, VSM, and PNR have been used to study the arrangement of spins in double superlattices and have allowed the direct observation of a magnetic parallel domain wall. It has been directly shown that it is necessary to introduce an anisotropy into the antiferromagnetically coupled superlattice to induce an exchange bias in the structure. In addition, the spin flop phase exists in the AF only if there is no or very little anisotropy. The parallel domain wall is primarily confined to the AF layers: the first direct experimental verification of the ideas of Mauri *et al.*, well over a decade after they were first published [4]. Since both the domain wall and the spin flop phase exist in the sample that does not have a large magnetocrystalline anisotropy in the antiferromagnetically coupled layer, neither phase is a direct consequence of exchange bias.

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