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Controlling the exchange interaction using the spin-flip transition of antiferromagnetic spins in $Ni_{81}Fe_{19}/\alpha$-Fe_2O_3

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We report anisotropy, coercivity, and exchange bias in ferromagnetic $Ni_{81}Fe_{19}$ layers coupled to antiferromagnetic (AF) (0001), $(11\bar{2}0)$, and $(1\bar{1}02)$ α-Fe_2O_3 layers. We show that AF spin configurations which permit spin-flop coupling give rise to a strong uniaxial anisotropy and hence a large coercivity, and that by annealing in magnetic fields parallel to specific directions in the AF we can control either coercivity or exchange bias. In particular, we show that a reversible temperature-induced spin reorientation in the AF can be used to control the exchange interaction.

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The interaction between a ferromagnet (FM) and an antiferromagnet (AF) across an interface gives rise to an exchange bias (H_{Ex}), i.e., a shift in the hysteresis loop as well as an enhanced coercivity (H_C) compared with the free FM (Ref. 1). This exchange bias is fundamental to the operation of spin valve devices such as magnetic read heads, nonvolatile memories, and various sensors.[2–4] Despite considerable work by many groups over the past two decades, the origin of the exchange bias and the enhanced coercivity are still unclear.[5–8] Mauri *et al.*[9] predict experimentally realistic values for H_{Ex} on the assumption that a domain wall parallel to the surface is formed in the AF layer. The random-field model of Malozemoff,[10] by considering interface roughness, qualitatively explained exchange bias in compensated AF surfaces. Koon[11] demonstrated that it is possible for the FM layer to minimize its energy when it aligns perpendicularly to the AF easy axis; this type of perpendicular exchange coupling has become known as spin-flop coupling because of its similarity to the spin-flop state of an AF material in a magnetic field. However, Schulthess and Butler[12] revealed that the spin-flop coupling alone cannot induce a unidirectional anisotropy, but instead gives rise to a uniaxial anisotropy which causes an enhanced coercivity. Experimentally, such spin-flop coupling has been observed in epitaxial FM/AF systems, such as $Ni_{80}Fe_{20}/Fe_{50}Mn_{50}$, Co/NiO, Fe_3O_4/CoO, and Fe/FeF_2 (Refs. 13–18). Nevertheless, a satisfactory understanding is not yet available because of complications at interfaces which include roughness, spin structure, and defects. Of particular relevance to the work presented here, Fitzsimmons *et al.*[18] have shown that exchange bias is dependent on the in-plane crystalline quality, and hence the net spin configuration at the interface, of an AF layer. The aim of the experiments reported here was to investigate the exchange bias and coercivity in a system in which the interfacial AF spin configuration could be controlled and changed without modifying the structural properties of the AF/FM interface.

Hematite α-Fe_2O_3 is attractive for exchange biased applications because of its high Néel temperature T_N ($\sim$680 °C) (Refs. 19 and 20). Bulk α-Fe_2O_3 undergoes an unusual temperature-controlled transition between two AF spin configurations—the so-called spin-flip (Morin) transition at $T_m \sim 260$ K—and has been identified as an ideal system in which to study exchange bias in general and spin flop in particular.[21]

Epitaxial α-Fe_2O_3 films on α-Al_2O_3 substrates have spin-flip transition temperatures that depend on the crystal orientation:[22,23] the T_m of $(11\bar{2}0)$ α-Fe_2O_3 is similar to that of the bulk material, while (0001) α-Fe_2O_3 films do not show a spin-flip transition above 2.5 K. In contrast, the spin-flip transition of $(1\bar{1}02)$ α-Fe_2O_3 films is increased to about 400 K; in this case the AF spins lying within the film plane above T_M flip to the out-of-plane direction below T_m. These changes are associated with lattice strain caused by the lattice mismatch between α-Fe_2O_3 and α-Al_2O_3 ($\sim$5.5%).

In this Letter, we report anisotropy, exchange bias, and coercivity FM layers coupled with (0001), $(11\bar{2}0)$, and $(1\bar{1}02)$ α-Fe_2O_3 layers. In contrast to previous experiments that have compared the exchange interaction associated with different *fixed* spin orientations associated with different AF crystal faces,[17] we show that a change of AF spin orientation across a *single* interface without structural disturbance is directly reflected in a modified exchange interaction with FM spins, i.e., in changes of anisotropy, coercivity, and/or exchange bias driven by the Morin transition.

Epitaxial α-Fe_2O_3 films were grown on α-Al_2O_3 substrates by pulsed laser deposition (PLD) with a substrate temperature of 700 °C and oxygen pressure of 20 mTorr. In order to fabricate the films under identical conditions, three substrates with different orientations were loaded side by side for simultaneous deposition. The 50-nm-thick α-Fe_2O_3 films were transferred into an ultrahigh vacuum dc sputtering chamber and a 5-nm $Ni_{81}Fe_{19}$ (NiFe) film was deposited in a magnetic field of 250 Oe at 295 K.

X-ray diffraction (XRD) measurements showed that all three α-Fe_2O_3 growth directions yielded films with excellent crystallinity: a full width at half maximum of $<0.08°$ and $\sim 0.9°$ in the rocking curve and in the ϕ scan, respectively. The rms surface roughness measured by atomic force microscopy was about 0.5 nm in each case. Therefore, effects caused by extrinsic factors such as roughness and defects should be virtually the same for all three samples, and thus differences in the H_{Ex} and the H_C should depend only on the

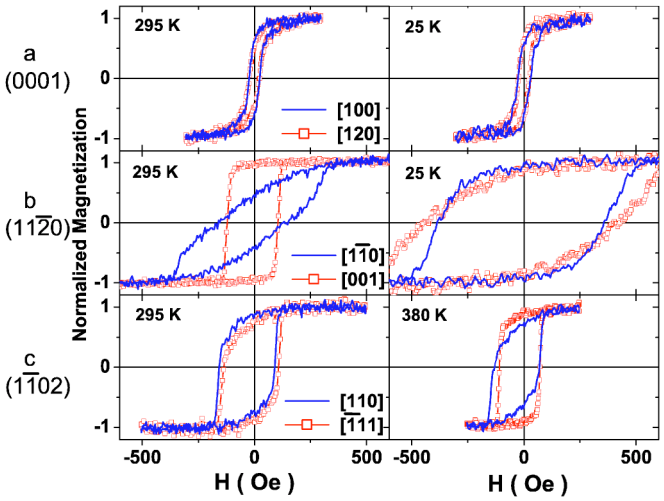

FIG. 1. The magnetic hysteresis loops of as-prepared $Ni_{81}Fe_{19}$ on (a) (0001) α-Fe_2O_3, (b) (11$\bar{2}$0) α-Fe_2O_3, and (c) (1$\bar{1}$02) α-Fe_2O_3 at several temperatures.

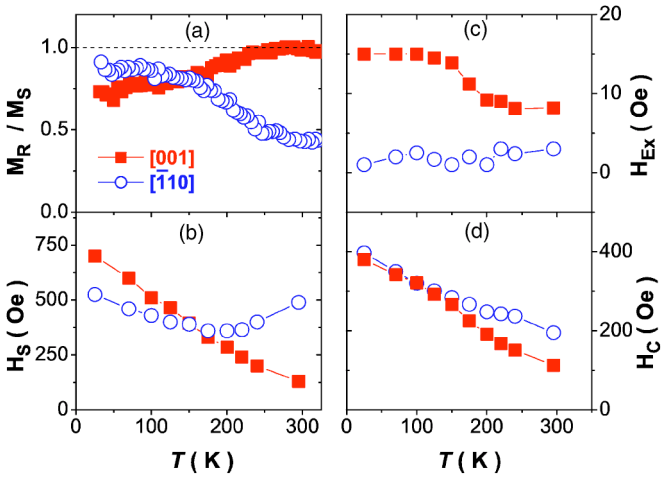

FIG. 3. $Ni_{81}Fe_{19}$ on (11$\bar{2}$0) α-Fe_2O_3: temperature dependence of (a) normalized remanent magnetization (M_r/M_s), (b) saturation field (H_s), (c) the H_{Ex}, and (d) the H_C with the in-plane direction.

spin structure of α-Fe_2O_3 at the surface. The strains observed in our films are similar to those reported by Fujii *et al.*[22] and so we expect a similar change in the Morin temperature.

The NiFe/α-Fe_2O_3 samples were measured in a variable temperature vibrating sample magnetometer (VSM). Figure 1 shows the temperature-dependent magnetic hysteresis loops of as-prepared samples for the different crystal orientations; each panel shows magnetization data collected for two orthogonal in-plane directions (see Fig. 2). For the (0001) orientation, the hysteresis loop is essentially independent of the temperature and the in-plane field direction: it shows minimal H_{Ex}, and a coercivity of about 22 Oe. In contrast, the hysteresis loops for the (11$\bar{2}$0) and (1$\bar{1}$02) orientations show large changes with the temperature and the in-plane field direction.

Figure 2 shows the crystal-direction dependence of the surface spin configuration of α-Fe_2O_3 (Refs. 22–24); the NiFe spin will always lie within the film plane because of large shape anisotropy. For spin-flop coupling to give rise to a strong uniaxial anisotropy, the FM spins must align perpendicular to the AF spins.[12] Accordingly, if the spin-flip transition of α-Fe_2O_3 at T_m results in a change of the in-plane spin direction, this should be reflected by a change in the easy

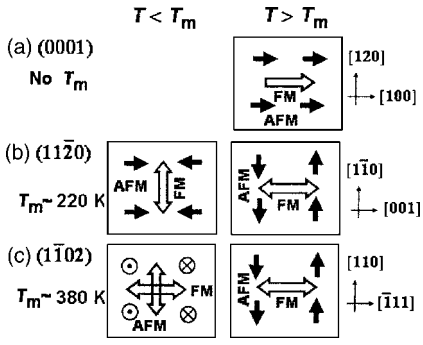

FIG. 2. Schematic surface spin structures of α-Fe_2O_3 on (a) (0001) α-Al_2O_3, (b) (11$\bar{2}$0) α-Al_2O_3, and (c) (1$\bar{1}$02) α-Al_2O_3.

axis of the NiFe. This expectation is consistent with our results for the (11$\bar{2}$0) and (1$\bar{1}$02) α-Fe_2O_3.

As seen in Fig. 1(b), the easy axis of the NiFe on (11$\bar{2}$0) α-Fe_2O_3 rotates by 90° with the temperature. Figures 3(a) and 3(b) show how the normalized remanent magnetization (M_R/M_S) and saturation field (H_S) of as-prepared NiFe/(11$\bar{2}$0) α-Fe_2O_3 for two in-plane directions depends on temperature. The 90° rotation of the easy axis of the NiFe on the (11$\bar{2}$0) α-Fe_2O_3 is associated with the spin-flip transition of α-Fe_2O_3 from the *ab* plane to the *c* axis [albeit associated with a reduced T_m, which is typical for 40–50 nm length scales in α-Fe_2O_3 (Ref. 25)]. Figures 3(c) and 3(d) show that the spin-flip transition modifies the H_{Ex} and the H_C for two in-plane directions with a clear decrease of H_{Ex} above T_m.

In contrast, hysteresis loops of the NiFe on (1$\bar{1}$02) α-Fe_2O_3 for two in-plane directions at 295 K are virtually identical except for a slight shift, but at 380 K there is a clear difference for the two in-plane directions, i.e., easy and hard magnetization axes [Fig. 1(c)]. The FM spins on (1$\bar{1}$02) α-Fe_2O_3 at room temperature have no preferential orientation because all in-plane directions equally satisfy a spin-flop coupling condition [Fig. 2(c)]. Upon warming, however, a preferential direction appears within the plane as the AF spins flip to one of the in-plane directions.

The substantially lower coercivity of NiFe/(0001) α-Fe_2O_3 is also consistent with the above picture since (0001) α-Fe_2O_3 has an uncompensated surface at all temperatures, and so cannot generate a large coercivity through spin-flop coupling. It is important to note that this uncompensated surface gave negligible exchange bias following any annealing procedure in contradiction to simple models for such systems; this may be a consequence of the small, but finite roughness in any practical sample. Although this may appear surprising, it is consistent with previous results in the exchange biased system Fe/FeF_2, which showed zero exchange bias for an uncompensated AF surface.[17] Nonzero exchange

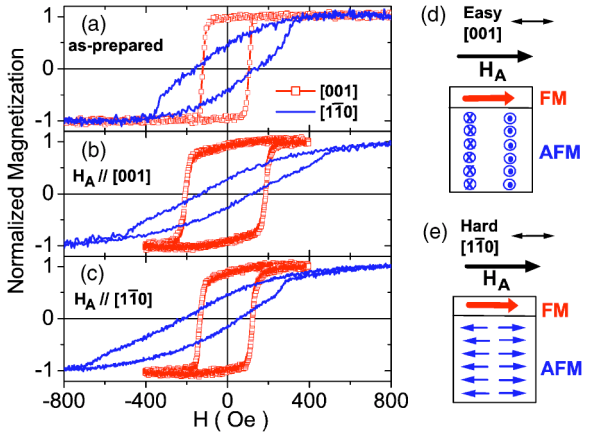

FIG. 4. The room-temperature magnetic hysteresis loops of $Ni_{81}Fe_{19}$ on $(11\bar{2}0)$ α-Fe_2O_3 (a) without MFA, (b) with MFA perpendicular to the AF spin direction, and (c) with MFA parallel to the AF spin direction. Two schematic MFA configurations were also displayed in (d) and (e). Here, H_A means the annealing magnetic field.

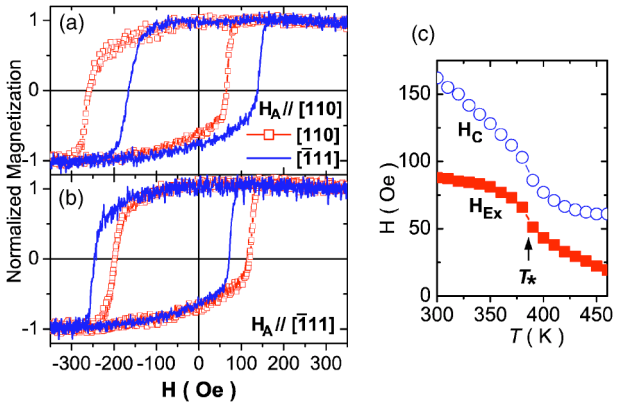

FIG. 5. The room-temperature magnetic hysteresis loops of $Ni_{81}Fe_{19}$ on $(1\bar{1}02)$ α-Fe_2O_3 (a) with MFA along the [110] direction and (b) with MFA along the $[\bar{1}11]$ direction. The temperature-dependent H_{Ex} and H_C was displayed in (c). The T_* roughly agrees with the Morin temperature of $(1\bar{1}02)$ α-Fe_2O_3.

bias by an uncompensated surface had been observed in Fe_3O_4/CoO with a very smooth surface by molecular-beam epitaxial growth.[15]

If the intrinsic anisotropy energy of a FM layer is negligible, the total energy per unit area in an exchange-coupled FM/AF system can be expressed as[26]

$$E = -J_1\cos\theta - J_2\sin^2\theta + K_{\mathrm{AFM}}\sin^2\phi, \qquad (1)$$

where J_1 and J_2 are, respectively, a direct (parallel) coupling constant and a spin-flop (perpendicular) coupling constant; θ and ϕ are the angles between the FM spin and the AF spin directions, and the AF spin and the AF anisotropy axis; K_{AFM} is the anisotropy constant of the AF layer. The lowest-energy state is thought to be a spin-flop-like state $\{\theta=90°,\ \phi=0°$ [see $T>T_m$ in Figs. 3(b) and 3(c)]}. The form of the spin-flop coupling is comparable with the classical uniaxial anisotropy energy, and thus the coercivity is mainly dependent on the second term of (1). If we associate exchange bias with a domain wall formed in the AF layer,[7,10,27] its stability is determined by a competition between a decrement of direct coupling energy and an increment of the AF anisotropy energy. From Eq. (1), we expect that magnetic-field annealing (MFA) perpendicular to the AF spin direction will stabilize the spin-flop coupling which will, in turn, enhance the coercivity. On the other hand, a MFA process parallel to the AF spin direction should induce an exchange bias because it enhances the direct coupling and suppresses the spin-flop coupling.

We performed a series of experiments in which MFA was performed under 10 kOe for 15 min at 200 °C, and the samples were cooled down to room temperature in the magnetic field. The $(11\bar{2}0)$ and $(1\bar{1}02)$ α-Fe_2O_3 films with compensated surfaces showed distinctive MFA effects; Fig. 4 shows the hysteresis loops of NiFe on $(11\bar{2}0)$ α-Fe_2O_3 before and after MFA. When the MFA was performed perpendicular to the AF spin direction [Fig. 4(d)], the exchange bias showed no change, but the coercivity approximately doubled [Fig. 4(b)]. In contrast, H_{Ex} of order 80 Oe was induced along the hard axis when the MFA was performed parallel to the AF spin direction [Fig. 4(c)]. A similar exchange bias along the hard axis has been observed in the epitaxial Fe/FeF_2 system.[17,18] Thus MFA with a configuration of Fig. 4(e) enhances the direct coupling in Eq. (1), and in turn it induces the H_{Ex} along the hard axis.

Finally, we applied MFA to the NiFe on $(1\bar{1}02)$ α-Fe_2O_3. A large exchange bias of 80–100 Oe was induced by the MFA along all directions within the plane, as seen in Fig. 5 (Ref. 28). If $(1\bar{1}02)$ α-Fe_2O_3 has the ideal spin structure of Fig. 3(c) below T_m, the NiFe should have shown no exchange bias because the spin configuration satisfies the spin-flop coupling condition. On the contrary, the large exchange bias suggests that the AF spin-flip transition to the out-of-plane direction during the cooling process of MFA is frustrated at the interface because of in-plane FM spins. In Fig. 5(c), the temperature-dependent exchange bias and coercivity clearly show an anomaly at T_*, which agrees with the T_m of $(1\bar{1}02)$ α-Fe_2O_3 in a previous report.[22] We conjecture that the cooling process of MFA assisted by FM spins enhances the direct coupling leading the formation of a domain wall in the AF layer, and thus the exchange bias is induced along all in-plane directions.

In α-Fe_2O_3 the Néel temperature is well above the experimental temperatures even for the MFA and so is largely irrelevant to the results reported here. Instead, in our experiments we see a distinctive decrease in H_{Ex} on heating through T_m, the transition between two different AF-ordered states [Figs. 3(c) and 5(c)]. This demonstrates that the spin reorientation at T_m effectively modifies the direct exchange coupling which has been established by cooling through T_m. Since the AF films in question have a single crystallographic orientation and compensated interfaces the net direct exchange energy can only arise from local exchange imbalances associated with surface roughness (the Malozemoff model[10]) and the micromagnetic AF structure. On warming

through T_m, the roughness is fixed and the micromagnetic structure need not be altered, but the local exchange imbalance will nevertheless change as a consequence of the global spin reorientation so as: firstly, to be orthogonal to the previous exchange bias direction and, secondly, to largely cancel since the direct exchange imbalance is equally likely to be positive or negative along to the new spin direction.

In summary, we have shown that there is a direct link between the spin direction in the AF and the anisotropy and coercivity induced by the exchange interaction in the FM. We have also demonstrated that magnetic-field annealing parallel to specific directions in the AF can alternatively modify either the coercivity or the exchange bias, in agreement with the Schulthess and Butler[12] and Malozemoff models,[10] respectively, demonstrating that these models are not mutually exclusive, but are part of a wider picture. We show that the exchange bias can only arise along specific spin directions in the AF and, most importantly, that spin reorientation in the AF directly affects the exchange bias. An ability to modify the exchange bias well below the T_N, albeit at a low level in this unoptimized system, offers the possibility of engineering different forms of control over the exchange interaction (for example by piezoelectric strain-driven variation of T_m) and so enhancing the role of the exchange interaction in magnetic data storage.

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