Coercivity Enhancement in Exchange Biased Systems Driven by Interfacial Magnetic Frustration

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We report the temperature and cooling field dependence of the coercivity of exchange biased MnF_2/Fe bilayers. When the antiferromagnetic surface is in a state of maximum magnetic frustration and the net exchange bias is zero, we observe a strong enhancement of the coercivity, which is proportional to the exchange coupling between the layers. Hence, the coercivity can be tuned in a reproducible and repeatable fashion in the same sample. We propose that a frustrated interface provides local energy minima which effectively pin the propagating domain walls in the ferromagnet, leading to an enhanced coercivity.

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The magnetic coercivity H_C (i.e., the half-width of the magnetic hysteresis loop) is an important parameter used to characterize magnetic materials. Control over the coercivity is desirable to tune the behavior of magnetic devices. In spite of this, the issue of controlling the magnetic coercivity has received little attention from physicists as it is supposed to be an extrinsic quantity often determined by such parameters as defect density. As such it is difficult to control in a reproducible fashion by changing an external parameter. Exchange bias H_E (the shift of the hysteresis loop along the field axis) has been extensively studied in antiferromagnetic (AF)/ferromagnetic (F) bilayers, although a quantitative understanding is still unavailable [1]. Despite this, some intriguing correlations exist between H_E and H_C. Moreover, recent theoretical work [2,3] claims that the behavior of H_C and the correlations between H_E and H_C provide important clues as to the microscopic origin of exchange anisotropy. However, experimental investigations of H_C, as well as H_E, in systems with well controlled and characterized microstructure are rare ([4–7] are examples).

Here we present measurements on an exchange biased system in which the coercivity can be tuned by the field applied (H_{FC}) when cooling through the AF Néel temperature T_N. In this fashion, the AF surface spin structure can be varied and its effect on the behavior of H_E and H_C observed. The crossover from negative to positive H_E, with increasing H_{FC}, is accompanied by an additional increase in the coercivity. This increase is in addition to that which occurs on cooling below T_N and increases with the exchange coupling between the AF and F layers. The dependence of the exchange bias on cooling field and temperature can be analyzed using a simple model in which the AF surface spin structure is modified by the applied H_{FC}, in the presence of an antiferromagnetic coupling between the F and AF layers [7–9]. We show that this enhancement is brought about by magnetic frustration at the AF/F interface, a result that has important implications for the physics of exchange biased systems.

ZnF_2(25 nm)/MnF_2(60 nm)/Fe(12 nm)/Al(3 nm) layers were deposited by electron beam evaporation. The nonmagnetic ZnF_2 layer serves as a buffer between the MgO(100) substrate and the AF MnF_2 layer ($T_N = 67$ K [8]), while the Al capping layer prevents oxidation. ZnF_2, Fe, and Al were deposited at 200 °C, 150 °C, and 150 °C, respectively, whereas MnF_2 was deposited at several temperatures in the 275–375 °C range. Varying the substrate temperature during deposition of the MnF_2 layer affords control over the roughness (σ) of the MnF_2/Fe interface [8] which in turn changes H_E. The pressure during fluoride deposition is around 6×10^{-7} Torr. X-ray diffraction, grazing incidence reflectivity, and reflection high energy electron diffraction were used for structural characterization, while magnetic measurements were made with a SQUID magnetometer between 4.2 and 100 K and in fields up to 70 kOe. Remnant fields were minimized by heating the superconducting magnet above its transition temperature after application of large fields. They were then measured and accounted for by measuring the hysteresis loops of single Fe films. It is noted that it has been suggested that hysteresis loops may provide a lower bound for H_E [10]. However, the absence of training effects, the reproducibility of the data, and earlier reversible measurements [11] all indicate that hysteresis loops provide a good measure of H_E.

ZnF_2 and MnF_2 layers have a body centered tetragonal structure with a (110) orientation perpendicular to the substrate surface, while the Fe overlayers are polycrystalline. The (110) fluoride reflection peak widths give a grain size equivalent to the thickness of the film. The roughness (σ) determined from refinement of the grazing incidence reflectivity [12] is identified as the rms thickness fluctuation at the fluoride-Fe interface over the relatively long length scale probed by grazing incidence reflectivity. This can be controlled from 0.6 nm up to $\gtrsim$4–5 nm without significantly varying the full width at half maximum of the MnF_2 (110) reflection or the rocking curve peak width, thus proving that the crystalline quality is not affected. As

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shown previously, MnF_2/Fe layers with "smooth" interfaces ($\sigma < 1.5$ nm) show positive H_E (i.e., in the same direction as H_{FC}) for cooling fields $\gtrsim 10$ kOe while layers with "rough" interfaces show only negative H_E (i.e., in the opposite direction to H_{FC}) [8].

Figure 1 shows the temperature dependence of H_E and H_C [defined as the half loop width at magnetic moment $(m) = 0$] for two H_{FC} values for a sample with a smooth MnF_2/Fe interface ($\sigma = 0.6$ nm). For low cooling field ($H_{FC} = 2$ kOe) typical behavior is observed: The *negative* H_E switches on close to T_N eventually saturating below 30 K [Fig. 1(a)], while H_C increases monotonically with decreasing temperature below T_N [Fig. 1(b)]. The temperature dependence of H_C for $ZnF_2/Fe/Al$, also in Fig. 1(b), shows a rather low H_C (<15 Oe) with a weak temperature dependence. These data clearly indicate that the large coercivities at $T < T_N$ in the MnF_2/Fe bilayers are due to exchange coupling across the AF/F interface. For $H_{FC} = 70$ kOe the temperature dependence of the *positive* H_E [Fig. 1(c)] and H_C [Fig. 1(d)] is similar to the $H_{FC} = 2$ kOe case. It is interesting that in both cases the H_C enhancement below T_N does not saturate at low temperatures, despite the fact that H_E reaches a temperature independent value (presumably this reflects the saturation of the AF sublattice magnetization and anisotropy [13]). Such behavior in H_E and H_C has been observed in several systems (e.g., [4–6,14–16]) and appears to be a common phenomenon. It seems clear that this behavior warrants further study. We note that the theoretical work mentioned earlier [2] suggests that H_E and H_C are of fundamentally different origins (H_C is due to the uniaxial anisotropy resulting from spin-flop coupling while H_E is created via a different mechanism, such as interfacial defects), suggesting that the temperature dependences should not necessarily be expected to be similar. However, we also note that identical mechanisms may give dissimilar temperature dependencies as coercivity is dependent upon thermally activated processes and is therefore expected to have a temperature dependence in the whole range of temperature measured here.

The cooling field dependence of H_E and H_C is shown in Fig. 2 for two representative samples: one with a smooth MnF_2/Fe interface ($\sigma = 0.6$ nm, shown in Fig. 1) and one with a rougher interface ($\sigma \approx 3$ nm). The smooth sample shows a crossover from negative to positive H_E at $H_{FC} = 10$ kOe, while the rough sample shows only negative exchange bias which is weakly dependent on H_{FC}. This behavior was found to be due to a crossover from AF exchange coupling to F exchange coupling with increasing roughness [8]. The coercivity behavior is intriguing; the rough sample (which shows only negative H_E) exhibits a very weak dependence of H_C on H_{FC} [Figs. 2(c) and 2(d)]. The smooth sample, on the other hand, shows a clear maximum in H_C close to the H_{FC} for which $H_E = 0$ [Figs. 2(a) and 2(b)]. The larger values of H_C measured for the sample showing only negative H_E are presumably due to the higher roughness of the Fe layer. Note that H_C of ZnF_2/Fe bilayers shows no measurable cooling field dependence.

Thus the cooling field provides an external agent by which the coercivity of the sample can be varied in a repeatable and reproducible way (i.e., the sample can be warmed above T_N, cooled to 10 K in the same field, and the same values of H_E and H_C recorded to within experimental uncertainty). In most other cases, changing H_C requires changing the density or nature of the defects in

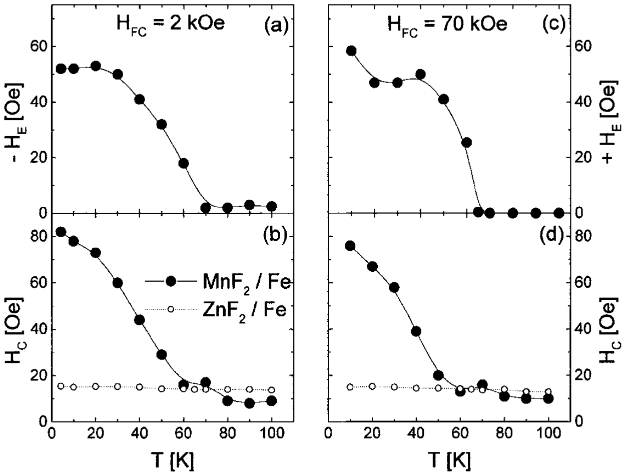

FIG. 1. H_E and H_C as a function of T for $H_{FC} = 2$ kOe and $H_{FC} = 70$ kOe [the sample has a smooth MnF_2/Fe interface ($\sigma = 0.6$ nm)]. (a) $H_E(T)$ for $H_{FC} = 2$ kOe, (b) $H_C(T)$ for $H_{FC} = 2$ kOe, (c) $H_E(T)$ for $H_{FC} = 70$ kOe, and (d) $H_C(T)$ for $H_{FC} = 70$ kOe. Note the sign reversal in H_E from (a) to (c). Open symbols are for ZnF_2/Fe. The lines are guides to the eye.

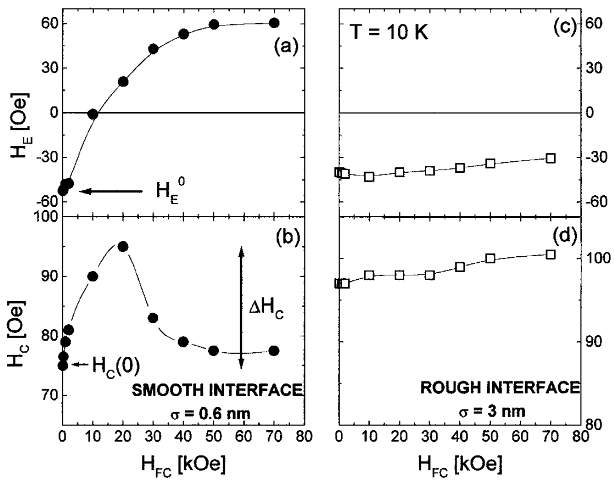

FIG. 2. H_E and H_C as a function of H_{FC} for a sample with a smooth MnF_2/Fe interface ($\sigma = 0.6$ nm; see Fig. 1) and a sample with a rougher interface ($\sigma \approx 3$ nm); (a) $H_E(H_{FC})$ for the smooth interface, (b) $H_C(H_{FC})$ for the smooth interface, (c) $H_E(H_{FC})$ for the rough interface, and (d) $H_C(H_{FC})$ for the rough interface. $T = 10.0$ K. The lines are guides to the eye.

the sample, or the crystallinity, with the consequent difficulties with interpretation.

We define the size of the "peak" in $H_C(H_{FC})$ [see Fig. 2(b)] as the increase of H_C from $H_{FC} = 0$ to the maximum value of H_C, labeled ΔH_C—the "coercivity enhancement." The percentage value can then be defined as $\Delta H_C/H_C(0)$, where $H_C(0)$ is the coercivity at zero cooling field. Figure 3 shows this percentage coercivity enhancement plotted against the exchange bias measured at low (2 kOe) cooling field (H_E^0), for a total of seven samples. The variation in H_E^0 is achieved by varying the interfacial roughness via the substrate temperature during growth [7,8]. These seven samples exhibit AF coupling between the AF and F layers and the low cooling field exchange bias is indicative of the coupling strength across the interface [8]. Hence the data of Fig. 3 show that the coercivity enhancement is more pronounced for strong coupling between layers and approaches zero as the AF coupling strength falls to zero.

Since in exchange biased systems the H_C enhancement is thought to originate from a finite H_E, the naive expectation is that H_C should reach a minimum close to $H_E(H_{FC}) = 0$. However, Fig. 2 clearly shows that the maximum in H_C occurs very close to the point at which $H_E(H_{FC}) = 0$. We first note that such coercivity maxima can generally be observed at magnetic phase transitions [17]. However, the AF surface or bulk spin flop can be ruled out as a possible mechanism as the fields involved are an order of magnitude larger than measured here [18]. The coercivity enhancement can be understood qualitatively and quantitatively as described below. Within the simple model for positive H_E [7–9], the AF exchange coupling across the interface ($J_{F/AF}$) is being frustrated by the coupling of the AF surface spins to H_{FC}. At low cooling fields the AF coupling dominates and a low energy state is "frozen in" at T_N, but for high cooling fields the coupling of the AF surface spins to H_{FC} dominates and leads to an unstable state being frozen at T_N. This leads to the positive exchange bias. It is in the intermediate region where H_E is close to zero that we observe the enhancement of the coercivity. Here some fraction of the spins are aligned with the cooling field and are therefore frustrating the AF exchange coupling, while others remain in the low potential energy, AF-coupled, state. *It is at this point of maximum frustration that we observe the largest* H_C. In other words, the AF surface splits into regions or "domains" [19], which are aligned either with H_{FC} or in the original AF-coupled configuration. Given that H_C in these Fe films is dominated by domain wall pinning, an enhancement of H_C can be understood in terms of increased pinning of the Fe domain walls at the edges of the AF domains. This explanation is also consistent with the data of Fig. 3 where it is seen that the coercivity enhancement becomes more pronounced with increasing exchange coupling. Essentially, the effectiveness of the AF domain walls as pinning sites for the propagating F domain wall increases as the exchange coupling between the layers is increased.

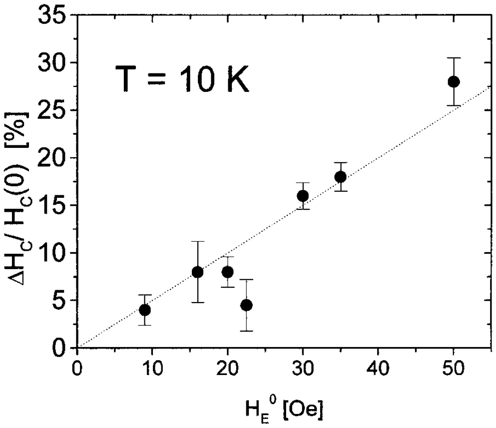

FIG. 3. ΔH_C (the percentage increase in coercivity over the $H_{FC} \to 0$ value) as a function of H_E^0, the low cooling field exchange bias (measured at $H_{FC} = 2$ kOe) for seven samples. The rms roughnesses as determined by low angle reflectivity are 0.60, 0.72, 0.80, 0.92, 1.00, 1.13, and 1.2 nm. The dotted line is a least squares linear fit through the origin. ΔH_C and H_E^0 are labeled in Fig. 2.

A simple modeling of this scenario can be realized by considering a domain wall in an F layer (in the x-y plane) of thickness t, domain wall width W, and length L, propagating over a semi-infinite AF block split into N square shaped domains aligned parallel or antiparallel to H_{FC}. Balancing the energy due to an applied field H with the energy change of the domain wall on propagating a distance δy gives the following integral [20]:

$$2M_S HLt = \frac{d\gamma}{dy_0} = \int_{x_0-L/2}^{x_0+L/2} \gamma(x, y_0 + W/2) - \gamma(x, y_0 - W/2)\, dx\,, \qquad (1)$$

where M_S is the saturation magnetization and γ is the domain wall energy. γ can be written $\gamma(x,y) = -M_S a_0 \underline{\hat{m}}(x,y)\underline{h}(x,y)$, where $\underline{\hat{m}}(x,y)$ is a unit vector in the direction of the local magnetization, $\underline{h}(x,y)$ is the local field, and a_0 is the interfacial atomic separation. We then define $\underline{h}(x,y)$ as a series of delta functions at each domain edge which crudely models the real situation where the domain walls in the AF provide a local energy perturbation for the propagating F domain wall. Evaluating the integral we obtain $\Delta H_C = a_0 h_0 L/t d_{AF}$, where ΔH_C is the enhancement of H_C, d_{AF} is the AF domain size, and the field h_0 is the delta function weight. To evaluate ΔH_C a suitable value for h_0 is required. As a first approximation, for an order of magnitude estimate we simply use the amplitude of the Malozemoff random field [21], $h_0 = J_{AF/F}/M_S a_0^3$, giving

$$\Delta H_C = J_{AF/F} L/d_{AF} t M_S a_0^2\,. \qquad (2)$$

At low H_{FC} and high H_{FC} the AF is aligned antiparallel or parallel to H_{FC}, there are no AF domains, d_{AF} approaches the sample size, and ΔH_C is minimized. At the point $H_E = 0$ the number of AF domains reaches a maximum, hence d_{AF} reaches a minimum value and there is a consequent maximum enhancement in H_C. Taking literature values [22] for the parameters involved we obtain the reasonable value $d_{\mathrm{AF}} \sim 1000$ nm, required to reproduce our observed ΔH_C of 30%. In addition to this, Eq. (2) implies that if the AF domain size (d_{AF}) is constant at the H_C peak, the H_C enhancement is directly proportional to $J_{\mathrm{AF/F}}$. Given that H_E is proportional to $J_{\mathrm{AF/F}}$ [23] this dependence is exactly that observed in Fig. 3, where the H_C enhancement increases linearly with the low field H_E^0.

In summary, we have measured the temperature and cooling field dependence of the coercivity of $\mathrm{MnF_2/Fe}$ exchange biased bilayers. In this system the surface spin structure of the AF layer can be varied by the cooling field which systematically varies the exchange bias and coercivity. A strong coercivity enhancement is observed in the region where the AF surface spin structure exhibits maximal frustration. The coercivity enhancement is shown to be proportional to the exchange coupling between the layers. Simple modeling shows that the increased coercivity is due to enhanced pinning of the propagating domain wall in the F layer resulting from the interfacial magnetic frustration.

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