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Competing interfacial exchange and Zeeman energies in exchange biased bilayers

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The exchange bias in antiferromagnet (MnF_2)/ferromagnet (Fe) bilayers has been studied as a function of temperature, cooling field, and interfacial roughness. Positive exchange bias is observed under certain conditions, along with a nonmonotonic roughness dependence. The unusual cooling field dependence can be understood within a simple model in which the exchange bias is determined by a competition between a roughness-dependent average exchange energy and a Zeeman energy for the antiferromagnet surface spins. Our results are consistent with the existence of a domain wall parallel to the interface in the Fe layer, contrary to earlier theoretical models. [S0163-1829(99)08541-0]

Exchange anisotropy at the interface between ferromagnetic (FM) and antiferromagnetic (AF) materials has received renewed attention in recent years.[1] In addition to the fundamental interest in exchange bias, interest has been stimulated by the use of exchange pinned ferromagnetic layers in hybrid spin valve magnetic field sensors.[2] Despite the recent revival and its discovery over four decades ago,[3] there is little quantitative understanding of exchange bias. Several theories[4–9] have been advanced to explain exchange bias (H_E) quantitatively, while recent experimental work has investigated the situation where the Curie temperature (T_c) of the FM is lower than the Néel temperature (T_N) of the AF,[10] memory effects,[11] uncompensated interfacial spins,[12] and the related phenomenon of exchange spring magnetism.[13] Further experimental input is required to ascertain the effect of important material parameters on H_E. In this paper, we examine the behavior of H_E in the simple epitaxial bilayer system MnF_2/Fe, as a function of temperature, cooling field, and interfacial roughness. We observe an unusual cooling field dependence as well as a nonmonotonic roughness dependence of H_E. The cooling field dependence can be explained within a simple model of competing exchange coupling and Zeeman energies *if* the interfacial exchange coupling is assumed to evolve from AF to FM with increasing roughness. This behavior is confirmed by a nonmonotonic roughness dependence, where the magnitude of H_E reflects the magnitude of the coupling. Ordinarily, AF/FM bilayer hysteresis loops are shifted along the field axis in the opposite direction to the applied cooling field[1] (''negative'' exchange bias). Positive exchange bias can also be observed under certain conditions,[14] and is thought to be a consequence of AF exchange coupling between the FM and AF layers. The roughness-dependent exchange coupling discussed in this paper leads to a remarkable situation in MnF_2/Fe where different behavior is observed in different roughness regimes. Note that in this paper we adopt the same model to interpret data on positive exchange bias as Ref. 14.

ZnF_2/MnF_2/Fe/Al thin films were grown on (100) MgO substrates by sequential electron beam evaporation. Thicknesses were nominally 25, 55, 12, and 3 nm, respectively. The ZnF_2 layer serves as a buffer layer, while the Al is used as a capping material to prevent oxidation. The deposition temperatures are 200, 275–375, 150, and 150 °C, respectively. The deposition temperature of the MnF_2 layer is varied to control the interfacial roughness (cf. Ref. 14). The base pressure in the system is 3×10^{-8} Torr, while the pressure during fluoride deposition is around 6×17^{-7} Torr. After growth, the films were characterized by low- and high-angle x-ray diffraction and reflection high-energy electron diffraction (RHEED). X-ray refinement based on the SUPREX model[15] were used to extract the interfacial roughnesses from low-angle x-ray data. Hysteresis loops were measured in a superconducting quantum interference device (SQUID) magnetometer from 4.2 up to 120 K and in magnetic fields up to 70 kOe. Note that it has been suggested that measurement of H_E via a hysteresis loop is likely to give a lower bound for the value of H_E.[16] On a similar note, we would like to point out that no magnetic training effect was observed under any of the experimental conditions reported in this paper. Checks for training effects were made by measuring three consecutive hysteresis loops in the limit of low (234 Oe) and high (70 kOe) H_{FC}, and at the extremes of roughness attained in this study. No training effect was observed within the experimental uncertainty.

High angle θ-2θ x-ray diffraction profiles show only the (110) orientation of ZnF_2 and MnF_2 with a full width at half maximum that corresponds to a grain size equivalent to the film thickness. Rocking curves through the (110) MnF_2 reflection have full widths at half maximum from 2.0° to 2.6°, indicating that we can control the roughness over a wide range without significantly changing the crystallinity. RHEED images of the surface of the ZnF_2/MnF_2 layers suggest ''quasiepitaxial'' (twinned[14]) growth, whereas the Fe overlayers are polycrystalline. Figure 1 shows the low-angle x-ray diffraction data for three representative samples (over 30 were grown, characterized, and measured) with varying interfacial roughnesses. The short-period oscillations are due

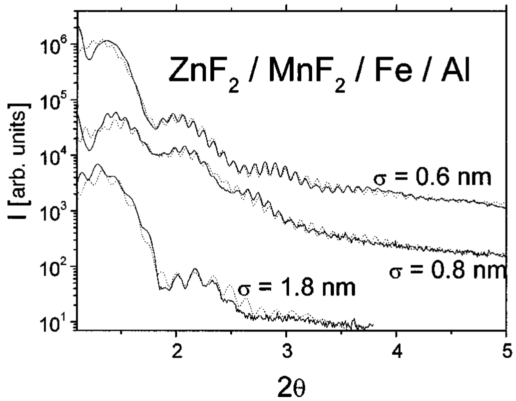

FIG. 1. Low-angle x-ray diffraction data on three representative samples of ZnF_2/MnF_2/Fe/Al (solid lines) along with fits (dashed line) based on the model described in Ref. 15. Each fit is labeled with the corresponding value of the rms roughness.

to the thickness of the Fe layer while the long-period oscillations are due to the fluoride thickness. Quantitative analysis of all of the samples in this study leads to MnF_2 thicknesses in the range 55.5–61 nm and Fe thicknesses between 10.6 and 12.6 nm. Here, the roughness is controlled by varying the deposition temperature from 275 to 375 °C, although similar results can be obtained by changing the ZnF_2 buffer layer thickness. The roughness can be varied from 0.6 nm up to approximately 4 nm. The roughness determined by the x-ray refinement is an rms value of the vertical thickness fluctuations on the relatively long lateral length scale probed by grazing incidence reflectivity.

H_E exhibits a strong dependence on H_{FC} as shown in Fig. 2. Typical H_E values are between 20 and 60 Oe. The smoothest sample ($\sigma = 0.6$ nm) shows a negative H_E at low cooling field crossing over to positive H_E in a field of less than 10 kOe, and saturating at around 60–70 kOe. The roughest sample ($\sigma = 1.8$ nm) shows a very different behavior, with H_E saturating at a negative value. The temperature dependence of $H_E(H_{FC} = 2$ kOe) for the $\sigma = 1.8$ nm sample is shown in the inset of Fig. 3. H_E approaches a constant value at low temperatures ($T < 30$ K), and falls to zero very

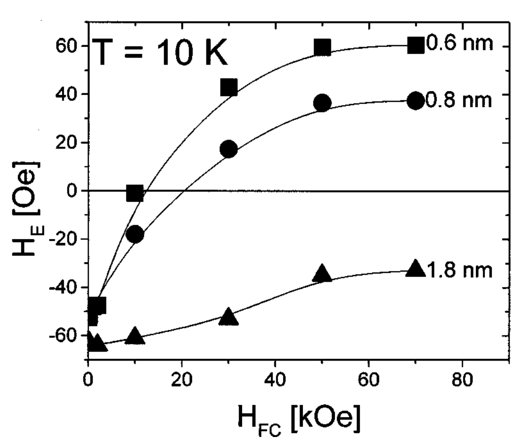

FIG. 2. Cooling field dependence of the exchange bias for the three samples shown in Fig. 1 from 234 Oe up to 70 kOe at $T = 10$ K. The curves are labeled with a value for the interfacial roughness. The solid lines are guides to the eye.

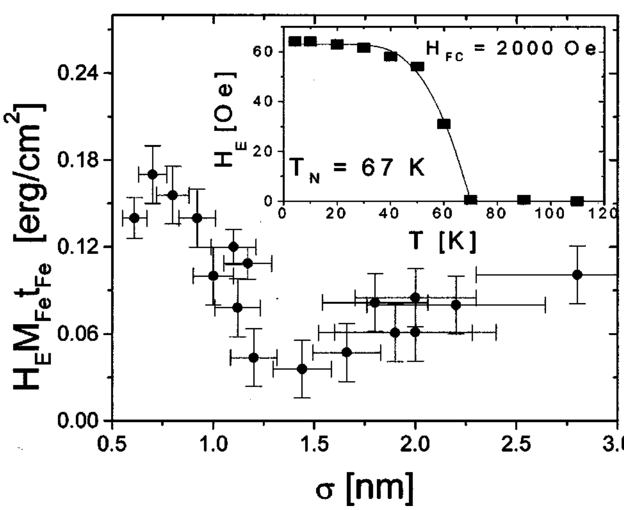

FIG. 3. Roughness dependence of the exchange bias energy per unit area, evaluated at $T = 10$ K and $H_{FC} = 2$ kOe. Inset, temperature dependence of the exchange bias from 4.2 K up to 120 K for the $\sigma = 1.8$ nm sample shown in Figs. 1 and 2 at $H_{FC} = 2$ kOe. The solid line is a $S = \frac{5}{2}$ Brillouin function fit. MnF_2 thicknesses are in the range 55.5–61 nm while the Fe thicknesses are between 10.6 and 12.6 nm.

close to the expected $T_N = 67$ K. This behavior is observed in all samples, regardless of the value of the interfacial roughness. The solid line in this figure represents a fit to the data with a Brillouin function for $H_E(T)$. Such behavior is expected if the magnitude of the exchange bias is proportional to the AF sublattice magnetization.[17] Note that the temperature dependence of the Fe magnetization is neglected here, as $T_c \gg T_N$. Models based on the formation of a domain wall parallel to the interface in the AF layer predict that H_E is controlled by a factor $(AK)^{1/2}$ where A is the AF spin stiffness and K is the AF anisotropy. In this case one expects that $H_E(T)$ would be given by $[A(T)K(T)]^{1/2}$, whereas if the AF anisotropy plays no role one would simply expect to see the temperature dependence of the AF sublattice magnetization as observed here. We return to this point later.

Further interesting behavior is observed in the roughness dependence of the low cooling field H_E as shown in Fig. 3. Initially, increasing the roughness from 0.6 nm results in a decreased value of the exchange bias energy per unit area, which reaches a minimum around 1.2 nm, eventually increasing for larger roughnesses. To our knowledge, this remarkable behavior is the first observation of a nonmonotonic dependence of the exchange bias energy on interfacial roughness. *It should be noted that the roughness at which the energy reaches a minimum corresponds to the maximum roughness value for which positive exchange bias effects are observed,* i.e., the point $\sigma \approx 1.2$ nm delineates two distinct regimes of behavior. Below this value of roughness all samples show positive exchange bias at high H_{FC}. Above this value of roughness the samples exhibit only negative exchange bias with no tendency towards positive bias up to $H_{FC} = 70$ kOe.

An exhaustive study of exchange bias in FeF_2/Fe bilayers[14] showed that H_E decreases rapidly with increasing roughness. Moreover, positive H_E was observed for large H_{FC} and explained in terms of AF exchange coupling between the AF and the FM. For low H_{FC}, the interfacial FM and AF spins align antiferromagnetically in an energetically stable state that leads to the usual negative bias. For large H_{FC} it was postulated that the AF surface spins couple to the applied magnetic field and overcome the AF exchange between AF and FM. An unstable state is frozen in at T_N, leading to positive H_E. In essence there is a competition between the exchange coupling energy and the Zeeman energy for the AF surface spins. It was further observed that positive H_E existed only for rough interfaces. In samples with smooth interfaces the exchange coupling is so large that it always dominates over the Zeeman energy for the AF surface spins. Random roughening of the interfaces leads to the formation of FM coupled regions, which lowers the *magnitude* of the average exchange coupling, J_{AV}. As a result, J_{AV}, which is still negative, has been reduced to a level where the Zeeman energy dominates at high H_{FC}, leading to positive H_E. The AF coupling as well as an increasing FM coupling with roughness can be qualitatively understood assuming the AF coupling is due to superexchange mediated by F^- ions at the interface.[18]

There are several clear experimental differences between the MnF_2/Fe and the FeF_2/Fe systems. In FeF_2/Fe the smooth samples show no positive H_E up to 70 kOe, while the rough samples crossover to positive H_E at around 10 kOe.[14] This is apparently opposite to that seen in MnF_2/Fe (Fig. 2). Also, in FeF_2/Fe even the curves that show only negative H_E might cross the H_{FC} axis at over 70 kOe (the largest measured field), while in MnF_2/Fe the negative H_E saturates at 50–60 kOe. Note that MnF_2 has a very similar crystal structure, spin structure, and magnetic properties to FeF_2 but a very different anisotropy field [$K_{\mathrm{AF}}=7$ kOe in MnF_2 ($S=\frac{5}{2}$) compared to 149 kOe in FeF_2 ($S=2$) (Ref. 19)].

First we address the seemingly opposite dependence of $H_E(H_{FC})$ on the interfacial roughness, when compared to the case of FeF_2. This can be explained using a simple model based on competition between the AF coupling at the FM/AF interface and the Zeeman energy of the AF surface spins. If the intrinsic coupling between the FM and the AF layers is significantly lower in MnF_2/Fe than FeF_2/Fe, the AF surface spin Zeeman energy ($g_{\mathrm{AF}}\mu_B H_{FC}$) dominates over the exchange coupling across the interface ($J_{\mathrm{FM/AF}}$). Hence positive H_E will occur at a value $H_{FC}^{*}=J_{\mathrm{FM/AF}}/g_{\mathrm{AF}}\mu_B$. However, when the interface is rough and regions of ferromagnetic coupling occur (just as in FeF_2/Fe), the magnitude of the average exchange coupling is reduced, eventually passing through zero and becoming positive (i.e., a net FM exchange coupling). At this point, positive exchange bias is prohibited since, within this model, it requires the existence of AF exchange coupling. This is exactly the behavior shown in Fig. 2, where the rough sample shows only negative H_E. Moreover, this simple model naturally explains the nonmonotonic dependence of the exchange bias energy on roughness, as shown in Fig. 3. In a simple model, the value of H_E is proportional to the magnitude of the exchange coupling energy $|J_{\mathrm{AV}}|$, which, according to the above argument, is finite at low σ (where $J_{\mathrm{AV}}<0$), falls to zero at a critical σ, and increases again at high values of σ (where $J_{\mathrm{AV}}>0$). This is exactly the observed behavior in $H_E M_{\mathrm{Fe}} t_{\mathrm{Fe}}(\sigma)$, where Fig. 3 shows an initial decrease to a point where H_E almost falls to zero, followed by an increase in H_E at higher values of σ.

We point out at this stage that we are taking the previously advanced model for positive exchange bias[14] and extending it to incorporate the data on MnF_2/Fe. To do this we are forced to *assume* that the intrinsic exchange coupling is weaker for MnF_2/Fe than for FeF_2/Fe leading to the crossover from AF to FM coupling with increasing roughness. The roughness dependence of H_E is completely consistent with this inferred crossover from AF to FM coupling and must be taken as strong evidence for it. Further evidence that the assumption on the size of $J_{\mathrm{FM/AF}}$ is reasonable is provided by the relative magnitudes of H_E and H_{FC}^{*} in FeF_2/Fe and MnF_2/Fe. Note that the smoothest MnF_2/Fe sample in this study ($\sigma=0.6$ nm) has the same Fe layer thickness as the smoothest FeF_2/Fe in Ref. 14 ($\sigma=0.6$ nm). $H_E\approx 450$ Oe ($H_{FC}=2$ kOe) for FeF_2 compared to $H_E\approx 50$ Oe for MnF_2. Since H_E is proportional to $J_{\mathrm{FM/AF}}$, we expect $J_{\mathrm{FeF_2/Fe}}/J_{\mathrm{MnF_2/Fe}}\sim 9$. Then, $H_{FC}^{*}=J_{\mathrm{FM/AF}}/g_{\mathrm{AF}}\mu_B$ implies that H_{FC}^{*} for FeF_2 and MnF_2 are in the ratio 9:1. For the smoothest samples this ratio is ~7:1 (see Fig. 2 and Ref. 14), in good agreement with our prediction. The origin of the reduced $J_{\mathrm{FM/AF}}$ in MnF_2/Fe is unclear although it is possible that weakened superexchange between Mn and Fe ions across the interface is partly responsible. A reduction in superexchange interaction energy for Fe^{2+}-F^--Mn^{2+} compared to Fe^{2+}-F^--Fe^{2+} can be estimated from the Anderson superexchange model,[20] by calculating the intersite hopping integrals and the Coulomb repulsion energy. It should also be noted that the *a* axis lattice parameter of MnF_2 is significantly larger than FeF_2, so possible effects of interfacial strain cannot be dismissed.

The intriguing issue that remains is the effect of the reduced anisotropy on the exchange bias. As discussed above, positive exchange bias is observed despite the low anisotropy field. Moreover, the smaller exchange bias in MnF_2/Fe compared to FeF_2/Fe can be accounted for by a decrease in the value of $J_{\mathrm{FM/AF}}$. This is further supported by comparing H_{FC}^{*} in the two systems. This seems to imply that the reduced value of K_{AF} has little effect on the magnitude of H_E. Also, the temperature dependence of H_E can be adequately accounted for by the AF sublattice magnetization alone, without the AF anisotropy field. A possible explanation for the lack of sensitivity to the AF anisotropy is that the exchange bias energy is stored in a domain wall parallel to the interface *in the Fe layer*, rather than in the MnF_2 layer. This was found by Kiwi *et al.*,[9] based on micromagnetic calculations and theoretical modeling of the FeF_2/Fe system. Further evidence for the existence of a domain wall in the Fe comes from the observation of a vertical shift in the exchange biased hysteresis loops and neutron scattering measurement in FeF_2/Fe.[21]

In summary, we have measured the dependence of the exchange bias in MnF_2/Fe bilayers on temperature, cooling field, and interfacial roughness. The effect of roughness on $H_E(H_{FC})$ can be explained in terms of a competition between a roughness-dependent average exchange coupling and a Zeeman energy of the AF surface spins. This simple model naturally explains the unusual nonmonotonic dependence of the exchange bias energy on roughness. The reduced exchange bias compared to FeF_2/Fe is due mainly to a reduction in the coupling between the layers rather than the effects of reduced anisotropy. This is consistent with recent theoretical arguments for domain formation in the ferromagnetic overlayer.

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