Exchange anisotropy and the antiferromagnetic surface order parameter

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The temperature dependence of the exchange bias (H_E) near the FeF_2 Néel temperature ($\sim$ 78.4 K) was correlated with structural measurements in FeF_2-Fe bilayers. Low-angle x-ray diffraction and atomic force microscopy show that samples with larger height fluctuations have larger lateral grain sizes. Samples with larger lateral grain sizes exhibit a surface critical exponent ($\beta_S \sim 0.8$) while samples with smaller grains and smaller height fluctuations have a decreased β_S, indicating a more three-dimensional-like phase transition or an increase in the FeF_2 surface exchange interaction. [S0163-1829(97)05230-2]

The critical exponents corresponding to a bulk magnetic material and those corresponding to its surface and the atomic planes close to the surface are in general different from each other. This occurs because spins at the surface have fewer nearest neighbors than those in the bulk.[1,2] Early mean-field calculations[3] of the surface magnetization in Heisenberg ferromagnets (FM) and antiferromagnets (AF) showed that the surface order parameter has the temperature dependence $\langle M_S \rangle \sim t^{\beta_S}$, where $\langle M_S \rangle$ is the surface magnetization for a FM or sublattice magnetization for an AF, β_S is the surface magnetization critical exponent, and $t = 1 - T/T_C$ is the reduced temperature, with T_C representing the Curie temperature for a FM or the Néel temperature for an AF. Within the mean-field theory, $\beta_S = 1$ and $\beta = 0.5$. The mean-field calculations were inspired by low-energy electron diffraction data,[4] which showed half-order spots induced by the appearance of antiferromagnetic order at the surface of NiO (100). Subsequent experiments[5] showed that $\beta_S = 0.89 \pm 0.01$ in NiO (100). Similar behavior was later observed in ferromagnetic Ni surfaces using spin-polarized low-energy electron scattering (SPLEED),[6] with $\beta_S = 0.83 \pm 0.03$. Although the latter two values of β_S do not agree with the mean-field result, they do agree with more sophisticated calculations using renormalization group theory,[7] where β_S ranges from 0.78 for the 3D Ising model to 0.88 for the 3D Heisenberg model. Monte Carlo simulations not only confirmed these results,[8] but also demonstrated that within the Ising model $\beta_S = 0.78$ only if the surface exchange interaction J_S is equal or smaller than the bulk J. If $J_S > J$, β_S decreases steadily, and for $J_S/J > 1.6$, the surface orders at a temperature higher than the bulk T_C. This occurs because, as J_S increases, the surface tends to behave as a two-dimensional magnetic material near the phase transition, rather than the surface of a three-dimensional material. Previous mean-field calculations qualitatively agree with this result.[3,9] Therefore, there are three phase transition effects associated with the surface of a magnetic material: (1) the atomic layers at or near the surface order with different critical exponents than the bulk, (2) the surface critical exponent β_S corresponding to the order parameter of the surface is greater than the bulk exponent β if $J_S/J < 1.0$, and (3) β_S decreases and the surface transition temperature increases for $J_S/J > 1.6$.

An interesting interfacial effect occurs when an antiferro-magnet, such as FeF_2, is placed in close proximity to a ferromagnetic material. When a ferromagnetic Fe thin film is grown on FeF_2, and the sample is field cooled through the FeF_2 Néel temperature, an exchange anisotropy arises.[10] This exchange anisotropy is characterized by a shift of the $M-H$ loop away from $H=0$, commonly denoted as the exchange bias H_E. Although this effect was discovered many years ago,[11] the detailed mechanism remains unclear. In the simplest model, the exchange bias arises from interface magnetic exchange, and is given by $H_E = 2J_I S_A S_F / M_F t_F a^2$, where J_I is the interface exchange energy, S_A and S_F are the spins of the antiferromagnet and ferromagnet, M_F is the magnetization of the ferromagnet, t_F the thickness of the ferromagnet, and a is the interface lattice parameter.[11] This model is inadequate for two reasons: (1) the values it yields for H_E are usually one to two orders of magnitude larger than determined from experiment, assuming that J_I has the same magnitude as the exchange interaction in the bulk FM, and (2) interface disorder or magnetically compensated surfaces would destroy H_E, while in practice the effect is observed in both cases.[10,12] More sophisticated models rely on domain wall creation, either parallel to the interface, created during the cool-down procedure,[13] or the rotation of the FM magnetization,[14] respectively. An important common feature of these models is that H_E is proportional to the strength of the interface magnetic interaction if the AF has uniaxial magnetic anisotropy. Moreover, because the AF domain wall size is independent of temperature for AF with uniaxial anisotropies, the model based on domain creation during cooling is only proportional to the interface interaction.[15] Hence, if the FM Curie temperature is much larger than the AF Néel temperature T_N, H_E reflects the order parameter of the AF surface near T_N because in the temperature range of interest the FM magnetization is almost constant.

In the present study we investigate the surface order parameter of antiferromagnetic FeF_2 by measuring the exchange bias of FeF_2-Fe bilayers near the FeF_2 T_N. This approach is experimentally easier than the SPLEED technique, but is limited to films that exhibit exchange bias and have an AF with strong uniaxial anisotropy. We find that $H_E \sim t^{0.80 \pm 0.04}$ for samples with large grain sizes, with the exponent decreasing as the grain size becomes smaller. For the large grain size samples, the exponent agrees with predictions of surface ordering of the three-dimensional (3D)

Ising model. The decrease in exponent as the grain size decreases may be due either to an effective decrease in lateral terrace size or an increase in the interface exchange interaction with decreasing roughness.

The Fe^{2+} ions in FeF_2 form a body-centered tetragonal crystal structure ($a=b=4.69$ Å, $c=3.301$ Å),[16] with the ions at the center of the unit cell ordering antiferromagnetically with the ions at the corners.[17] FeF_2 has a large uniaxial magnetic anisotropy ($K\sim1.39\times10^8$ erg/cm^3) along the c axis.[18] Because of its large anisotropy, FeF_2 behaves as an Ising model system over a wide temperature range.[19]

The growth of FeF_2-Fe bilayers on MgO (100) has been described elsewhere.[10] Briefly, the films were grown by sequential e-beam evaporation of FeF_2 ($t_A\sim90$ nm at a rate of 0.2 nm/s) and Fe ($t_F\sim14$ at a rate of 0.1 nm/s) with the rates controlled by a calibrated quartz crystal oscillator. Substrates were heated to 450 °C for 900 s prior to deposition in order to reduce the brucite layer on the MgO substrates, then cooled to the FeF_2 growth temperature $200\leqslant T_S\leqslant300$ °C. At these temperatures the FeF_2 grows quasiepitaxially along the (110) direction with two in-plane domains. The Fe layers were deposited at 150 °C, resulting in polycrystalline films with mostly (110) and (100) orientations, and then capped with ~9 nm of Ag to prevent oxidation. The pressure during deposition was $<1\times10^{-6}$ Torr.

Samples were cooled from 120 K through the FeF_2 critical temperature [$T_N=78.4$ K (Ref. 19)], to 10 K in the presence of a magnetic field $H_{FC}=2$ kOe. The Fe M-H loops were measured using a SQUID magnetometer in the -2 kOe to $+2$ kOe range (Fig. 1, top inset). In all cases, 2 kOe was enough to saturate the magnetization of the films. The Fe magnetization was measured every 5 Oe near $M=0$ in order to determine H_E more accurately.

Figure 1 shows the typical behavior of H_E near the FeF_2 Néel temperature for a sample with $t_A=100$ nm FeF_2 layer grown at 300 °C and $t_F=14$ nm. H_E was determined from the M-H loop (top inset). The H_E data were fit to a ''rounded'' power law:

$$H_E=\frac{H_{E0}}{\delta_T\sqrt{2\pi}}\int_0^\infty t_0^{\beta_S}\exp[-(T_C-T_{C0})^2/2\delta_T^2]dT_{C0}, \quad (1)$$

where $t_0=1-T/T_{C0}$ for $T<T_{C0}$ and $t_0=0$ for $T>T_{C0}$. H_{E0} is the exchange bias at $T=0$, T_C is the center of the critical temperature distribution, and δ_T is its width. This equation is valid for a Gaussian distribution of transition temperatures, of width δ_T, due to disorder, strain, or other defects. The parameters β_S, T_C, δ_T, and H_{E0} were simultaneously fit. The errors from these parameters were estimated by fitting all parameters but one and plotting χ^2 vs each fixed parameter. The uncertainty for β_S was ±0.04. The solid line in Fig. 1 is a result of this fit. The bottom inset shows the data in a log-log scale, where the straight line is a power law with $\beta_S=0.8$. δ_T was approximately 0.7 ± 0.2 K for all samples, regardless of the transition temperature. Given that the samples had different amounts of interface disorder (see below) yet similar values of δ_T, the rounding could also be due to the Fe magnetically ordering the FeF_2 at a temperature above the T_N of FeF_2.

In order to determine the effects of the interface structure, the FeF_2 was grown at different temperatures in the
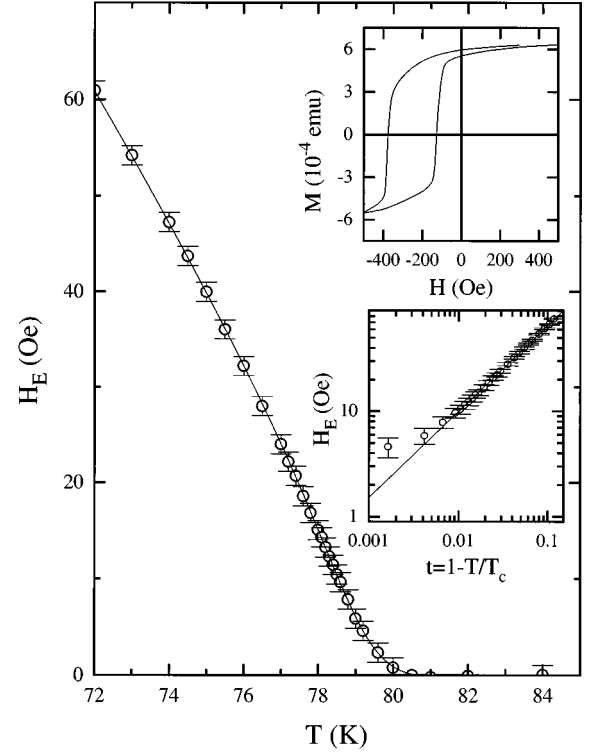

FIG. 1. H_E as a function of temperature near T_C for a sample with the 100 nm FeF_2 layer grown at 300 °C and an Fe thickness $t_F=14$ nm. The solid line is a result of a fit to Eq. (1), with $T_C=79.33\pm0.2$ K, $\delta_T=0.7\pm0.2$ K, $\beta_S=0.80\pm0.04$, and $H_{E0}=718\pm34$ Oe. Top inset: M-H loop at 10 K for the same sample after field cooling. H_E is the displacement of the center of the loop away from $H=0$. Bottom inset: log-log graph of H_E vs the reduced temperature t. The straight line is the power law $H_E \propto t^{0.80}$.

$200\leqslant T_S\leqslant300$ °C range. Small-angle x-ray diffraction[20,21] and atomic force microscopy techniques were used to quantitatively analyze the interface structure. In general, samples grown at higher temperatures had larger thickness fluctuations (roughness) as well as larger lateral correlation lengths (grain size).[10] By fitting the x-ray data to an optical model which takes into account the diminished reflectivity at each interface due to disorder,[22] roughness parameters were obtained for some of the samples. It was difficult to fit, using this model, data from samples that were too rough (i.e., which did not show many low-angle peaks).

It is known that the magnitude and sign of H_E in the FeF_2-Fe system depends strongly on the cooling field, H_{FC}.[23] However, it is important to note that despite these changes in H_E, we found that β_S *remains unchanged as a function of* H_{FC}.

The interface magnetic energy is defined as $\Delta E=2M_F t_F H_E$, where $M_F=1740$ G is the magnetization of Fe at low temperatures and t_F is the Fe thickness. In Fig. 2 we show ΔE as a function of the parameter σ obtained for the Fe-Ag interface, which represents the film's average thickness fluctuations. As has been previously discussed,[10] σ for this interface must be related to the σ at the FeF_2-Fe interface because, for these samples with similar FeF_2 and Fe thicknesses (to within 10%), the growth temperature of the FeF_2 was the only growth parameter which varied from
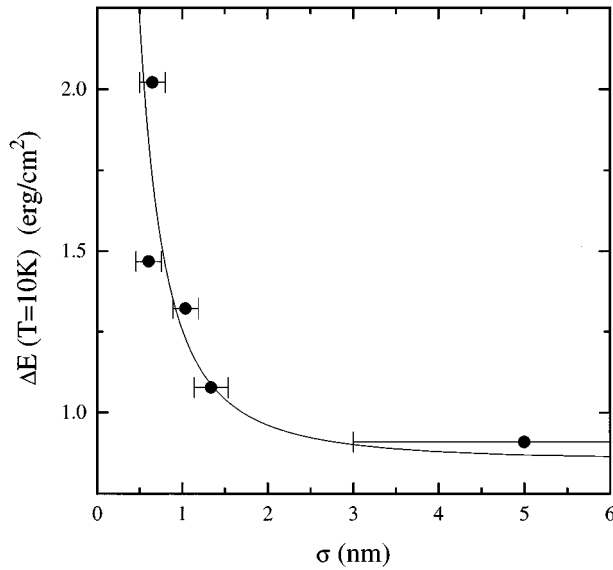
FIG. 2. ΔE vs Fe-Ag interface roughness σ obtained from grazing x-ray diffraction analysis for different samples. The solid curve is a guide to the eye. Note that samples with larger σ also have larger terraces (island size).

FIG. 4. T_C vs β_S for different samples. The straight line is a guide to the eye.

sample to sample. The parameters obtained for the FeF_2-Fe interface had large uncertainties, presumably due to the lower contrast between FeF_2 and Fe, and are thus unreliable. It is clear from these data that the exchange bias tends to decrease as σ increases. However, it is important to keep in mind that samples with larger values of σ have larger lateral grain sizes ξ_g.[10] As will be shown below, it is the changes in ξ_g that cause β_S to vary.

The critical exponent β_S for all samples, including those whose x-ray spectra were not fit, are shown in Fig. 3 as a function of ΔE. The general trend is for β_S to decrease as ΔE increases. Because ΔE decreases as ξ_g (and σ) increases (compare with Fig. 2), β_S decreases as ξ_g decreases. Note that for the samples with larger ξ_g (samples with lower
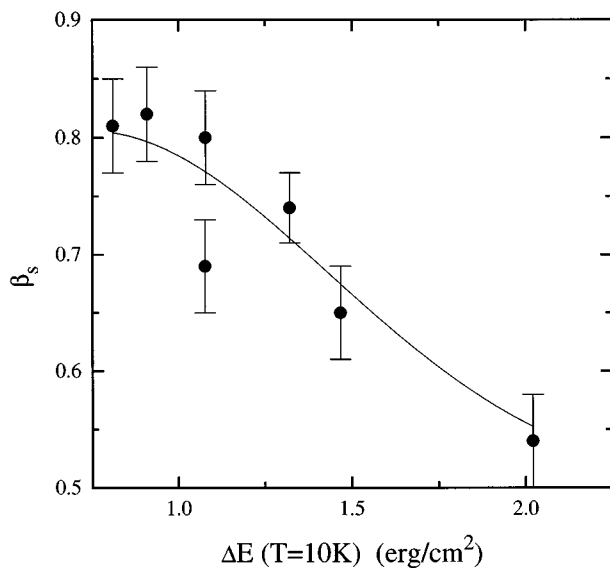

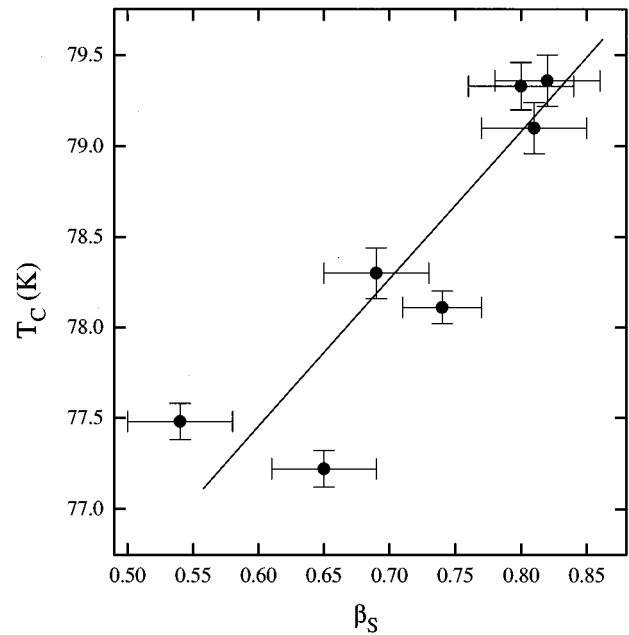FIG. 3. β_S vs ΔE for different samples. The solid curve is a guide to the eye.

ΔE), $\beta_S \sim 0.80 \pm 0.04$, which agrees well with the critical exponent of the surface of a 3D Ising system ($\beta_S = 0.78$). The samples with lower values of ξ_g have a lower critical exponent, but are still significantly larger than bulk FeF_2 ($\beta = 0.325$). This means that as ξ_g increases, the transition becomes more sensitive to the interface, i.e., β_S approaches the value of the surface of a 3D Ising system. One possible reason for this behavior is related to the exchange at the FeF_2 surface. If the exchange at the interface is greater than in the bulk FeF_2, β_S should be lower than 0.80 because the surface would order independently from the bulk (i.e., 2D like), tending to order with an exponent closer to the well-known 2D Ising value[24] of 0.125. In the range $1.0 < J_S/J < 1.6$, Monte Carlo simulations predict that β_S should decrease *without* increasing T_C, while T_C should increase for $J_S/J > 1.6$. Figure 4 shows T_C vs β_S. The changes in T_C are small, and in any case the trend is contrary to the theoretical prediction because β_S decreases as T_C decreases. Nevertheless, this slight decrease could be due to finite size effects, resulting from an effective reduction of the lateral correlation length of the order parameter as the terrace size decreases, which in FeF_2 is known to be important for sizes less than ~ 70 Å.[25] Hence, this may indicate that $1.0 < J_S/J < 1.6$ for all the samples. Note that T_C for some samples in Fig. 4 is larger than $T_N = 78.4$ K, the bulk FeF_2 Néel temperature. This could be due to the interaction with the Fe, which would tend to order the FeF_2 surface at $T > T_N$.

Another explanation is directly related to the terrace sizes. The interface structural analysis mentioned above showed that as σ increases, the lateral grain size ξ_g also increases. Samples with smaller grain sizes tend to have a larger number of atoms at island or step edges which will have more nearest neighbors than atoms on the flat regions of the surface. This effectively results in a smaller value of β_S. However, samples with larger grains may have larger flat regions,

resulting in a value of β_S closer to the value corresponding to a surface $\beta_S \sim 0.8$. The effective value for β_S is a result of the average value over the whole sample, which includes averaging over regions where the Fe is in contact with FeF_2 atoms belonging to the second and perhaps third monolayer, as well as with atoms near corners. Hence, the exchange bias probes a distribution of values of β_S because of the interface disorder.

Finally, in antiferromagnets with cubic anisotropy, such as CoO or NiO, the temperature dependence of H_E is more complex. In the model proposed by Malozemoff,[15] for example, the temperature dependence is proportional to both the square of the order parameter in the AF bulk and to the interface order parameter for AF's with cubic anisotropy. In the present context FeF_2 has an intrinsic advantage, since it has a uniaxial anisotropy, and theoretically the temperature dependence of H_E should be directly proportional only to its surface order parameter.

In conclusion, we have systematically studied the temperature dependence of the exchange bias in FeF_2-Fe AF-FM bilayers as a function of interface disorder. This provides an indirect measurement of the FeF_2 surface order parameter. The critical exponent of H_E near T_C, β_S, is found to increase as the AF film thickness fluctuations and the lateral grain size increase. This could be the result of an increase in the surfacelike exchange interaction at the interface or the presence of larger terraces as the lateral island size increases.

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