Calculations of Exchange Bias in Thin Films with Ferromagnetic/Antiferromagnetic Interfaces

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A microscopic explanation of exchange bias in thin films with compensated ferro/antiferromagnetic interfaces is presented. Full micromagnetic calculations show the interfacial exchange coupling to be relatively strong with a *perpendicular* orientation between the ferro/antiferromagnetic axis directions, similar to the classic "spin-flop" state in bulk antiferromagnets. With reasonable parameters the calculations predict bias fields comparable to those observed and provide a possible explanation for both anomalous high field rotational hysteresis and recently discovered "positive" exchange bias. [S0031-9007(97)03407-8]

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The phenomena of exchange bias was discovered more than 40 years ago by Meiklejohn and Bean [1], who found that fine particles of partially oxidized Co exhibit magnetization curves with an unusual displacement along the field axis, as though there were a "bias" field ($\mathbf{H_{EB}}$) in addition to the applied field. Bias fields are observed *only* if an external magnetic field is applied while cooling the ferromagnetic (*F*) Co particles below the Néel temperature of their antiferromagnetic (*A*) CoO coating. Experiments by Bean [2] on Co/CoO thin films demonstrated that exchange bias is primarily an interface phenomena, although it has also been observed in inhomogeneous F/A mixtures [3]. Meiklejohn and Bean [1] also showed there was a close connection between anomalous high field rotational hysteresis and exchange bias, although the two do not generally coexist.

In recent years exchange bias in thin films has found important technological application in such devices as magnetoresistive sensors. However, its fundamental origin is still unclear [4,5]. The simplest model [1], which assumes that the F/A interface occurs at an ideal uncompensated (all spins aligned) plane of the antiferromagnet, predicts bias fields of orders of magnitude larger than those observed and fails to explain biasing when the interface plane of the antiferromagnet is fully compensated (zero net moment). Mauri *et al.* [4] provided an explanation for the reduced bias fields by showing that the formation of a domain wall parallel to the interface dramatically lowers the energy required to reverse the magnetization. However, exchange coupling across the F/A interface was simply assumed, and questions regarding the origin of the F/A coupling or what happens at fully compensated interfaces were not addressed. Malozemoff [5] interpreted exchange bias in terms of random exchange fields due to interface roughness. However, his model has extrinsic features which depend on details of the microstructure [6].

Recently, several groups have reported experimental results which cannot be readily explained by the theories discussed above. Jungblut, *et al.* [6] studied molecular-beam epitaxy grown epitaxial wedge structures of NiFe/FeMn, as a function of NiFe (*F*) and FeMn (*A*) thicknesses as well as orientation of the FeMn, and found critical thicknesses of the antiferromagnet for the onset of exchange bias as well as the fact that the *F* magnetization ($\mathbf{M}_F$) tended to orient perpendicular to the *A* easy axis. Nogues *et al.* [7] found for Fe on a single crystal FeF_2 that the largest exchange bias occurred for the fully compensated (110) interface orientation and that increasing interface roughness *decreased* the bias fields. The same group [8] also discovered that field cooling Fe/FeF_2 in large applied fields ($\sim$70 kOe) can result in *positive* exchange bias, where the magnetization reversal occurs as the field is lowered while it still has the *same* sign as the cooling field rather than the opposite sign, as normally observed.

This Letter presents results of full micromagnetic (generalized mean field) numerical calculations on a simple model of a thin film with compensated F/A interfaces. The main difference from normal mean field calculations is that each spin in the primitive magnetic cell is allowed to have its own direction. In spite of the simplicity of the model, many of the established properties of thin film exchange bias systems appear naturally, as do some which are not so well established. Fully compensated perfect interfaces are found to be favorable for exchange bias, and roughness does *not* generally play an essential role. The mechanism for storage of magnetic exchange bias energy is found to be parallel domain walls, as proposed by Mauri *et al.* [4]. The 90° angle between magnetization directions in the ferromagnet and the antiferromagnet [6] is predicted to be a normal occurrence and is shown to be related to the well-known "spin-flop" state in antiferromagnets. Irreversible magnetic transitions in the antiferromagnet are found which provide a mechanism to account for the high field rotational hysteresis normally associated with exchange bias [1]. Reasonable values of exchange and anisotropy energies lead to bias fields the same order of magnitude as those observed experimentally at low temperatures. Finally, an explanation for positive exchange bias [8] is presented which may permit determination of the sign of the microscopic exchange interaction J_{FA} between *F* and *A* spins.

The model assumes a simple body centered tetragonal magnetic structure with exchange interactions along the body diagonals. For the antiferromagnet this leads to the simplest type of antiferromagnetic order in which spins on the two sublattices are oppositely directed and only interact with spins on the other sublattice, as indicated in Fig. 1. The magnetic unit cell of a (110) film with this structure contains two spins per plane because of the lack of translational invariance in the [110] direction. Classical spins with $S = 1$ were assumed, and uniaxial anisotropy K_U favoring the [001] was allowed only on the A sites. Only zero temperature calculations are presented.

Although relaxation methods can be used directly to calculate exchange biased magnetization curves, it is instructive to first consider some simple cases. The key issue of exchange coupling or "pinning" across a F/A interface is examined by considering a (110) oriented F/A film with thicknesses t_F and t_A of 15 monolayers (ML) each. It was assumed that $J_{FF} = -J_{AA} = -J_{FA} = 1$ meV. In each of the two outer layers (one F and one A) all the spins are constrained to lie along the same axis, with an angle φ between the axes of the layers. The initial configuration was with the axis directions parallel ($\varphi = 0$) and with the inner spins random in orientation. Relaxation methods were used to solve for the energy per unit area and spin configurations as φ increased.

In Fig. 2 the energy per unit area is shown as a function of the fixed angle φ for two cases: one as described above with a frustrated F/A interface, and a second with either all F or all A exchange interactions of the same magnitude ($|\mathbf{J}| = 1$ meV). For pure F or A films the minimum energy occurs at $\varphi_0 = 0°$, as expected, while the film with a frustrated F/A interface has a minimum at $\varphi_0 = 90°$. In both cases the angular dependence of the energy is well described by a polynomial of the form $c(\varphi - \varphi_0)^2$. For the mixed F/A film $c = 1.98 \times 10^{-5}$ meV deg^{-2}, and for pure F or A films $c = 2.10 \times 10^{-5}$ meV deg^{-2}. Since c is proportional to the exchange constant J, one arrives at the striking conclusion that the effective exchange coupling between the top and bottom layers of the 30 layer film with a completely frustrated F/A interface is not only shifted by 90°, it is reduced in magnitude by a relatively small amount (~6% in this case) compared to a homogeneous (A or F) film with the same magnitude of exchange and same thickness.

Such a strong "macroscopic" exchange coupling can only occur if the magnetic symmetry is broken by interactions at the F/A interface. The nature of this broken symmetry is illustrated in Fig. 3, which gives the configuration of spins near the F/A interface at the minimum energy angle $\varphi = 90°$. If J_{FA} is antiferromagnetic the A spins cant away from the F direction as shown. If J_{FA} is ferromagnetic they cant toward the F direction. In both cases the canting angle decays rapidly as a function of distance from the interface, becoming essentially zero at 5–6 ML.

Thus it can be concluded that frustration at a fully compensated F/A interface does *not* lead to zero exchange coupling as the simplest model would suggest [1] but to a macroscopic 90° coupling only somewhat weakened compared to homogeneous full exchange. This 90° coupling state is precisely analogous to the well-known spin-flop state of antiferromagnets in an applied field if one identifies the applied field with frustrated microscopic exchange across the interface.

This result also seems to suggest that interface roughness plays a somewhat different role than usually assumed [5]. Because maximum frustration is already present in a perfectly compensated F/A interface plane, it appears plausible that surface roughness could only reduce frustration at such an interface and thus reduce the bias field compared to that of a perfect interface. This observation is consistent with experimental work on Fe/FeF_2, which shows that the bias fields are largest for the compensated (110) orientation and decrease with increasing roughness of the interface [7].

A second key issue is the nature of coupling to the lattice, without which exchange bias could not exist. Since anisotropy in a ferromagnet does not, by itself, lead to exchange bias, it is reasonable to guess that the essential pinning comes from uniaxial anisotropy K_U favoring [001]

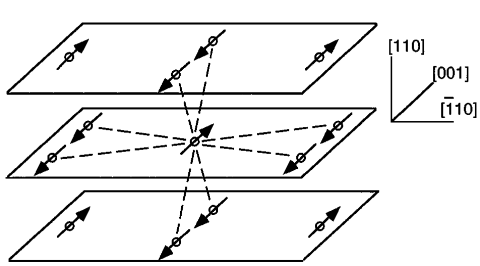

FIG. 1. Magnetic structure of the model for only antiferromagnetic interactions and emphasizing the (110) planes. Exchange bonds are shown by the dashed lines.

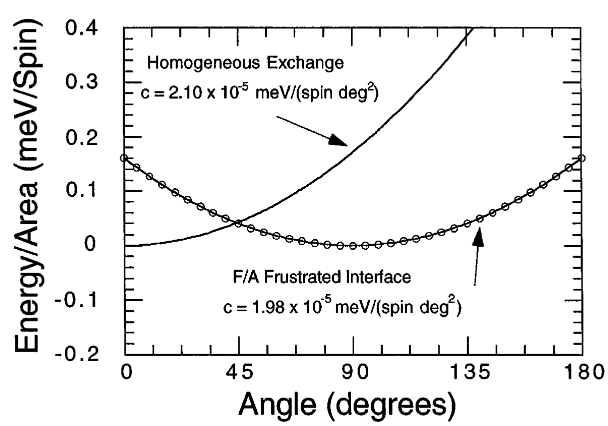

FIG. 2. Energy per unit area of a 15/15 ML (110) F/A film as a function of angle between the top (F) and bottom (A) magnetization axes (curve with data points). The smooth curve corresponds to a structurally identical film with only A or only F spins. For both films $|\mathbf{J}| = 1.0$ meV.

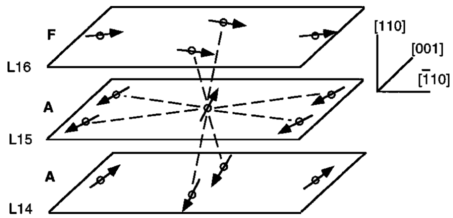

FIG. 3. Spin configuration near the interface plane for a 15/15 ML F/A film with lowest energy orientation ($\varphi = 90°$). Parameters are the same as for Fig. 2. The two interface planes are $L15$ and $L16$. Angles are approximately to scale.

on the A sites. To examine this issue a simplified model is considered where the F spins are locked together ($J_{FF} = \infty$) and fixed at an angle θ in the (110) plane relative to [001]. In accordance with the preceding calculation, one would expect two degenerate minimum energy orientations of $\mathbf{M}_F$ at [$\bar{1}$10] and [1$\bar{1}$0] ($\theta_0 = 90°$ and 270°, respectively). To calculate the energy curves as a function of t_A in Fig. 4 it was assumed that the initial direction of $\mathbf{M}_F$ was [$\bar{1}$10] ($\theta_0 = 90°$), while the A spins were randomized. The angle θ was incremented away from 90°, with each final spin configuration serving as the starting configuration for the next angle. For a choice of $\theta_0 = 270°$, one obtains an essentially identical set of curves (not shown) shifted by 180°. For t_A much less than the A domain wall width w ($\sim(4J/K_u)^{1/2} = 9$ ML) the angular dependence of the energy is reversible and similar to that of K_U, except for the 90° interfacial shift. The curves also exhibit mirror symmetry about $\theta = 0°$ and 180°, reflecting the mirror symmetry of the ($\bar{1}$10) plane. When $t_A \gtrsim w$, however, the energies increase smoothly as $\mathbf{M}_F$ rotates through $\theta = 0°$ and 180°, and the energy curves no longer have mirror symmetry. After $\mathbf{M}_F$ passes through the mirror plane the spin configuration follows a metastable high energy branch, blocked by an energy barrier from transitioning to a lower energy branch until $\theta = \theta_{CR}(t_A)$. The two different states of the A spins at each angle correspond to one or the other of the two degenerate ground state directions of $\mathbf{M}_F$ in zero field. Transitions between the states depend mostly on the orientation of $\mathbf{M}_F$ (driven by $\mathbf{H}$) and are weakly dependent on the interaction of $\mathbf{H}$ with A because the net moment of A is small for normal laboratory fields. Such discontinuous rotations, primarily in the antiferromagnet, therefore appear to be a plausible mechanism for the anomalous high field rotational hysteresis observed by Meiklejohn and Bean [1].

More importantly, the energy curves in Fig. 4 for large t_A ($\theta_{CR} > 90°$) can lead to exchange bias. For 50 ML, for example, there is a minimum energy centered at 90° [$\bar{1}$10] with an angular range of 344° over which $\mathbf{M}_F$ can rotate reversibly and only a $\delta = 16°$ range where irreversible behavior occurs, as is illustrated in the inset of Fig. 4. If the direction of $-\mathbf{H}$ ($\mathbf{H}$ is along the field cooling direction) falls outside the range of δ, then the energy curve will be entirely reversible, regardless of the magnitude of $\mathbf{H}$. The range δ decreases rapidly with increasing t_A/w greater than one, which clearly reflects the need to have a minimum thickness of the antiferromagnet in an exchange bias system.

The preceding discussion is subject to the approximations that the F spins have infinite stiffness and that the effect of the applied field on A can be neglected, neither of which is correct in general. It is therefore essential to calculate an exchange bias curve with all restrictions on the spin orientations removed and an external field applied to both the ferromagnet and antiferromagnet using realistic anisotropy and exchange parameters. The results are shown in Fig. 5 for a case where $\mathbf{H}$ is applied 10° from [$\bar{1}$10], $t_F = t_A = 50$ ML, and $J_{FF} = 16$ meV, which corresponds approximately to Fe or Co. The angular offset from [$\bar{1}$10] was used for convenience to reduce t_A and computation time. Other parameters are the same as for Fig. 4. The orientations of all spins were first randomized and

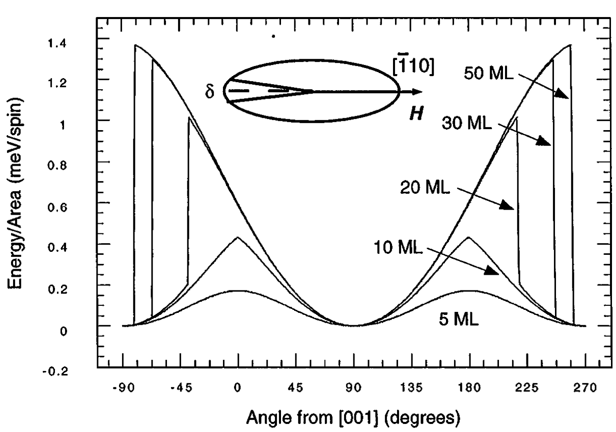

FIG. 4. Energy per unit area as a function of angle θ between $\mathbf{M}_F$ and [001] in the (110) plane for F/A films with different t_A. Parameters are defined in the text. Calculated antiferromagnetic domain wall width is 9 ML. Only curves with $\mathbf{M}_F$ initially along [$\bar{1}$10] ($\theta = 90°$) are shown. The inset shows the range δ where irreversible transitions occur.

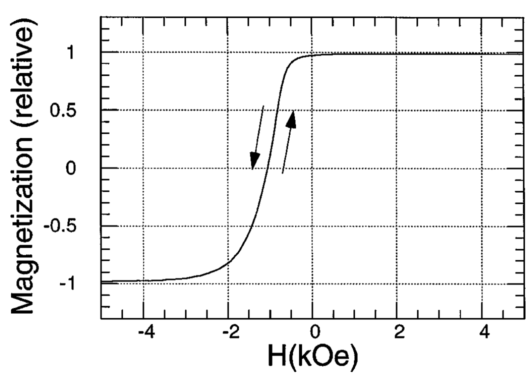

FIG. 5. Calculated magnetization curve for a 50/50 ML F/A film. Parameters are the same as for Fig. 4, except $J_F = 16$ meV. For convenience, the field was applied 10° from the [$\bar{1}$10].

then allowed to relax with a 5 kOe field applied. The field was then decreased in small increments to -5 kOe, with each spin configuration serving as the starting point for the subsequent one. A completely reversible (no hysteresis) exchange bias curve is obtained with $\mathbf{H_{EB}} \sim 1.0$ kOe, as shown in Fig. 5. This is a larger bias field than typically seen at room temperature, but is comparable to the low temperature bias fields observed in Co/CoO [1] and Fe/FeF_2 [8].

The physical picture emerging from this model is that exchange bias results from the formation of a mainly antiferromagnetic parallel domain wall [4] as a reverse field rotates $\mathbf{M}_F$ away from the field cooled direction. The domain wall is made possible by strong macroscopic F/A exchange coupling and pinning of the A by anisotropy. If this picture is correct, then the recent observation of positive exchange bias [8] implies that in sufficiently large fields the ground state of the system *must* contain such a domain wall, since it is the "unwinding" of the wall which causes the magnetization to reverse *before* the magnitude of the field decreases to zero. Preliminary calculations for the present model confirm that the ground state can generally contain such a domain wall if J_{FA} is negative (antiferromagnetic), but not if it is ferromagnetic. Observation of positive exchange bias would then imply antiferromagnetic J_{FA}. Normal negative bias, on the other hand, is only weakly dependent on the sign of J_{FA}.

In summary, the broad qualitative agreement between calculations utilizing this simple model and a variety of observed exchange bias phenomena suggests that the model does contain much of the essential physics of interface dominated exchange bias systems. A more detailed discussion of these calculations as well as applications to real thin film exchange bias systems will be presented elsewhere.

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