Consequences of Spin-Flop Coupling in Exchange Biased Films

T. C. Schulthess and W. H. Butler

Metals and Ceramics division, Oak Ridge National Laboratory,Oak Ridge, Tennessee 37831-6114

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Using a microscopic Heisenberg model which includes magnetostatic interactions, the Landau-Lifshitz-Gilbert equation of motion is solved in order to study several magnetic properties of ferromagnetic/antiferromagnetic bilayers. For perfectly flat interfaces, it is shown that spin-flop coupling does not lead to exchange bias as has been proposed, but rather gives rise to a uniaxial anisotropy which in turn causes the large coercivities observed in exchange biased films. The introduction of interfacial defects leads to exchange bias of the correct order of magnitude. [S0031-9007(98)07656-X]

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"Exchange bias" is a shift (H_{eb}) in the magnetization curve away from the zero field axis that can occur when a ferromagnetic (FM) film is in contact with a variety of antiferromagnetic (AFM) materials. The shift occurs when the AFM is ordered in the presence of a field or an already ordered FM film. Even though this effect was discovered four decades ago [1], its microscopic origin has not yet been established. Recent applications of exchange biased films to magnetoresistive sensors, however, have renewed interest in the effect and in its underlying physics.

One major difficulty in understanding exchange bias lies in understanding the coupling between FM and AFM materials for the case in which the AFM interface is compensated [2], i.e., when there is an equal number of positive and negative exchange interactions across the interface so that the net exchange interaction vanishes. The first theory to successfully describe this case is that of Malozemoff [3], who explained the coupling as due to a random field which he attributed to interface roughness and to the formation of domains in the AFM when the system is cooled through the ordering temperature. While this theory successfully describes most of the common phenomena related to H_{eb}, it fails to explain some of the more recent experimental findings, in particular, the tendency of the FM to align perpendicularly to the AFM easy axis [4,5]. This perpendicular coupling can be understood [6,7] within a microscopic Heisenberg model where, due to frustration of the moments at the interface, the FM minimizes the energy when it aligns perpendicular to the AFM easy axis. The name *spin-flop coupling* has been used to describe this type of effective exchange coupling between AFM and FM films. By restricting the motion of the spins during field reversal to the plane parallel to the interface, Koon [7] was able to make his model show the exchange bias effect through a mechanism proposed by Mauri *et al.* [8], in which a domain wall forms in the AFM when the FM magnetization rotates. The resulting magnetization curve is reversible and is shifted by realistic values of H_{eb}. This mechanism differs entirely from Malozemoff's theory and it is, at present, unclear as to which of the two theories should prevail. While the observation of perpendicular alignment clearly supports the view of spin-flop coupling, other evidence, such as a recent experiment by Takano *et al.* [9] which shows the relation between net uncompensated AFM magnetization at the interface and H_{eb}, tends to support Malozemoff's theory. Two additional observations which are not addressed by either of the theories but which seem to be related to the AFM-FM coupling are as follows: (1) A large coercive field (H_c) is almost always observed in exchange biased films [10]. (2) The AFM-FM coupling measured in reversible experiments can be several times larger than the coupling derived from H_{eb} [11].

In the present contribution it will be shown that, contrary to Koon's expectation, spin-flop coupling does not lead to the formation of a domain wall during magnetization reversal but rather introduces a large uniaxial anisotropy. It will further be shown that the two theories, Malozemoff's random field and Koon's spin-flop coupling, do not conflict but that rather a combination of both points of view provides an explanation for all of the above-mentioned phenomena.

Koon calculated magnetization curves under the constraint that the spin motion is parallel to the interface. While this constraint has some justification in the FM, where the spins are generally held in the film plane by the magnetostatic field, it is not obvious for AFM spins. In order to remove the constraint from the calculations in a proper way, the magnetostatic interactions have to be included explicitly. Thus, in the present approach, the microscopic Heisenberg model is extended to include, in addition to the usual exchange, Zeeman and anisotropy energies, the magnetostatic energy, $E_D = \sum_{i \neq j} \{\vec{\mu}_i \vec{\mu}_j - 3(\vec{\mu}_i \hat{n}_{ij})(\vec{\mu}_j \hat{n}_{ij})\}/|\vec{R}_i - \vec{R}_j|^3$, of a configuration of atomic moments $\{\vec{\mu}_i\}$, where $\hat{n}_{ij}$ is the unit vector that points in the direction that connects the sites at $\vec{R}_i$ and $\vec{R}_j$. Since all systems considered here are periodic in two dimensions (2D), the lattice sums for E_D are performed using an Ewald method outlined by Adams and McDonald [12] but adapted to 2D periodic lattices. Magnetic properties are calculated using the usual

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approach of classical micromagnetics, i.e., the local magnetic field is determined from the gradient of the energy, $H_i[\{\vec{\mu}_i\}] = -\frac{\partial}{\partial \vec{\mu}_i} E[\{\vec{\mu}_i\}]$, and $\{\vec{\mu}_i\}$ is required to satisfy the Landau-Lifshitz equation of motion (EOM) with the Gilbert-Kelley form for the damping term: $\frac{\partial}{\partial t}\vec{\mu}_i = -\gamma(\vec{\mu}_i \wedge H_i) + \sigma(\vec{\mu}_i \wedge \frac{\partial}{\partial t}\vec{\mu}_i)\frac{1}{|\vec{\mu}_i|}$, where γ and σ denote, respectively, the gyromagnetic ratio and the damping parameter. While the material parameters that enter the expression for the energy have direct physical justifications, the damping term is phenomenological and is included to remove the energy from the system and to ensure that the magnetic configuration is in a stable or metastable equilibrium. Care has been taken to ensure that all quantitative results presented here are independent of the magnitude of the damping parameter.

To illustrate the first important result of the present paper, the approach just outlined is applied to the generic model for AFM/FM bilayers used by Koon [7]. In order to simplify the interpretation of the results, a square lattice was used to ensure that the magnetostatic contribution (which was omitted from Koon's model) is isotropic in the plane parallel to the interface. Koon did not specify the anisotropy used in his calculations, but the magnitude does not qualitatively affect the results.

A reversible magnetization curve (Fig. 1a, solid line), similar to the one obtained by Koon, with a domain wall forming in the AFM, could only be generated when the precession term was removed from the EOM and the initial configuration had all moments perfectly parallel to the interface plane. In this case, the torque acting on the spins has no out-of-plane component which is equivalent to the constraints used by Koon. When the full EOM is used, the torque has an out-of-plane component which allows the spins in the interface region to relax into the second spin-flop state that corresponds to the energy minimum of the bilayer with reversed magnetization (Fig. 2). The resulting magnetization curve is irreversible (Fig. 1a, dotted line). In other words, when the physical EOM is solved, the energy barrier that the configuration has to overcome to switch between the two spin-flop states shown in Fig. 2 is smaller than the energy of the domain wall in the AFM (Fig. 1b).

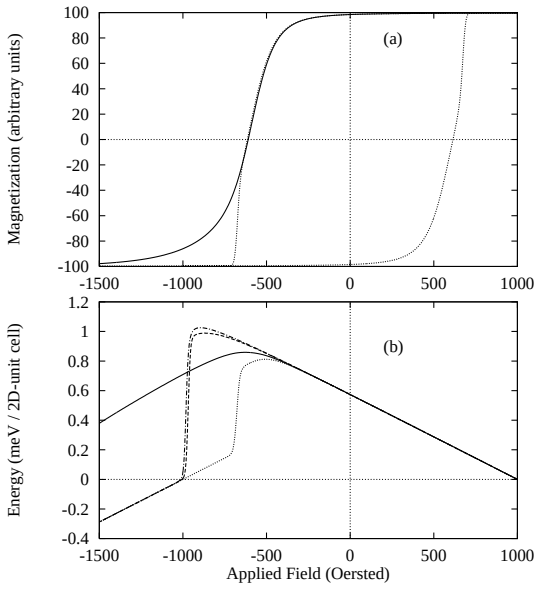

FIG. 1. Magnetization curves (a) and energy (b) for applied field changed at a slow but constant rate. Solid lines represent the reversible solution of EOM without the precession term; dotted lines represent the irreversible solution of the full EOM. Note that only one leg of the energy curve is shown. The AFM anisotropies used: 0.01 meV/site (solid and dotted lines); 0.1 meV/site (dashed line); 1.0 meV/site (dash-dotted line).

The physical conclusion of the previous discussion is straightforward. For flat interfaces, spin-flop coupling does not lead to a shifted magnetization curve (unidirectional anisotropy) but rather introduces a uniaxial anisotropy which leads to irreversible magnetization curves with finite coercivity. Additional mechanisms are required to obtain a unidirectional anisotropy necessary for exchange bias. To further substantiate these conclusions, the microscopic Heisenberg model will now be applied to CoO/FM bilayers, where either Permalloy (Py) or Co will be used as a FM. The (111) plane will be chosen for the AFM-FM interface, since, as will become apparent, it leaves the least ambiguity for the AFM-FM exchange parameters.

The present model for CoO/FM (111) bilayers (Fig. 3) assumes the usual NaCl structure for the AFM, where the Co^{2+} ions occupy an fcc lattice with a 4.27 Å lattice constant and have a magnetic moment of $3.8\mu_B$ [13]. Since the anisotropy direction in films is not known, two cases will be considered for the AFM easy axis: $[\bar{1}\,\bar{1}7]$ as in bulk CoO [13] and $[\bar{1}01]$ which is in the interface plane and was found to be the easy axis in some (001) films [5]. In both cases the anisotropy constant is chosen to be 3 meV/Co^{2+} [13]. Because of the strong anisotropy, the only exchange interaction that is significant in the AFM, after the magnetic configuration has ordered, is the next-nearest-neighbor interaction which is $J_{A\text{-}A} = -2$ meV [13]. In order to keep the problem solvable from a microscopic point of view, the AFM is assumed to be in a single domain state with compensated (111) planes at the interface [14] and only the case where the FM magnetization rotates coherently will be considered explicitly in the calculations. In this context the FM

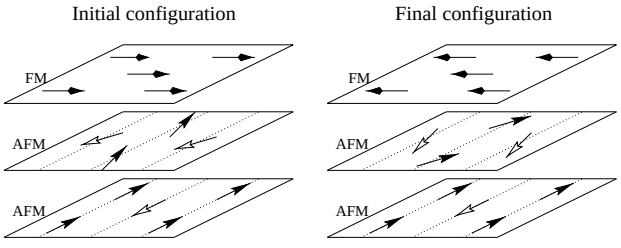

FIG. 2. Schematic of initial and final spin-flop states corresponding to the solution of the full EOM. Note that AFM sublattice magnetization (lower two planes) is the same in both cases, except very near the interface.

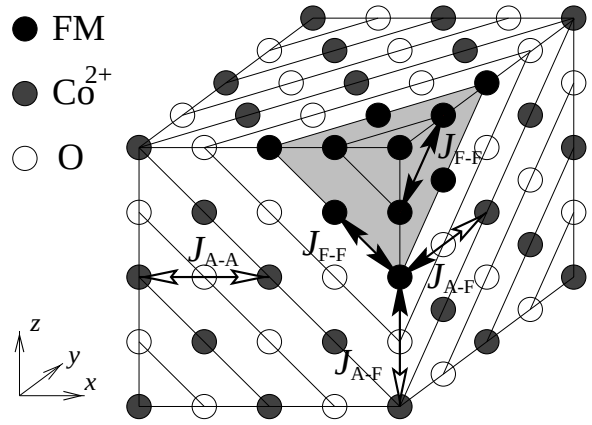

FIG. 3. Model of CoO-Co (111) interface. Values chosen for exchange interactions (indicated by arrows) are next-nearest-neighbor AFM, $J_{A\text{-}A} = -2$ meV; nearest-neighbor FM, $J_{F\text{-}F} = 16$ meV; next-nearest-neighbor AFM-FM, $J_{A\text{-}F} = -2$ meV.

lattice can be matched to that of the AFM with an atomic volume that corresponds to a lattice constant of 3.55 Å. For flat interfaces the magnetic 2D unit cell thus contains two sites per monolayer. Furthermore, since the exchange interactions in the FM are much larger than in the AFM and the current work is only concerned with anisotropies that are induced by the AFM-FM coupling, the only material-dependent property that is relevant in the FM is its magnetization. The magnetic moments used for Co and Py are, respectively, $1.7\mu_B$ and $0.9\mu_B$. The magnetocrystalline anisotropy in the FM is neglected, and the exchange interactions are assumed to be $J_{F\text{-}F} = 16$ meV [15] between nearest neighbors. The only parameter that is not known *a priori*, but to which the results are expected to be sensitive, is the exchange between the FM and CoO. It will be assumed that $J_{A\text{-}F} = J_{A\text{-}A}$, which for CoO/Co may be reasonable but is not obvious for CoO/Py. Thus the sensitivity of the spin-flop coupling strength to $J_{A\text{-}F}$ will be investigated. Finally, all results presented here are for FM layers which are 200 Å thick.

As in the case of Koon's model, the present calculations for a CoO/FM bilayer yield spin-flop coupling, as well as symmetric and irreversible magnetization curves for flat interfaces. The coupling axis, defined by the direction of FM magnetization at zero applied field, is perpendicular to the AFM easy axis and in the film plane. To determine the spin-flop coupling strength, a field is applied in the film plane perpendicular to the coupling axis ($\phi_H = 90°$). The EOM is solved and for small enough field [16] the total energy per unit area is found to fit form: $E(H_\perp) = E(0) + K_{\text{eff}} \sin^2 \phi$, where ϕ is the angle between the total magnetization in the unit cell and the coupling axis. Results for the effective coupling constant, K_{eff}, are given in Table I. They are relatively insensitive to the anisotropy direction as well as to the choice of the magnetic moments (μ_{Py} versus μ_{Co}). K_{eff} is also relatively robust to changes in $J_{A\text{-}F}$, as long as the latter is not much smaller than $J_{A\text{-}A}$.

TABLE I. Spin-flop coupling constant, $K_{\text{eff}}(\text{erg/cm}^2)$, for different AFM-FM exchange parameters and AFM easy axis.

FM	AFM easy axis	$J_{A\text{-}F}$	K_{eff}
Py	$[\bar{1}01]$	-1	0.54
Py	$[\bar{1}01]$	-2	0.87
Py	$[\bar{1}01]$	-3	0.83
Py	$[\bar{1}\,\bar{1}7]$	-2	0.64
Co	$[\bar{1}01]$	-2	0.88

For coherent rotation of the magnetization, the coercivity can be determined by reversing the field and finding the equilibrium solution of the EOM for constant applied field. When the applied field is larger than a certain threshold, the metastable solution becomes unstable and the FM magnetization switches. It is this critical field that corresponds to the coercivity. Results for the present model of CoO/Py bilayers, with the external field applied parallel to the interface plane at an angle $\phi_H = 10°$ from the coupling axis, are shown in Table II. The values of H_c clearly depend on ϕ_H and are about 10%–20% larger for $\phi_H = 0°$. The reason for the particular choice of $\phi_H = 10°$ is discussed in the next paragraph.

Defects at the interface, such as steps, islands, or point defects, can be included by increasing the size of the 2D unit cell parallel to the interface and including a corresponding arrangement of AFM sites on the FM side of the interface. Table II includes results for the case in which the 2D unit cell was increased form 2×1 to 4×4, with one interfacial FM site replaced by an AFM site by accordingly changing the moments as well as the exchange and anisotropy parameters. The defect site is decoupled from one of the AFM sublattices and antiferromagnetically coupled to the other. Consequently, the two spin-flop states (Fig. 2) for a given AFM configuration no longer have the same energy. This shifts the magnetization curve and gives exchange bias. Note that it is the coupling of the FM to the AFM through the uncompensated defect that gives rise to this unidirectional shift, the spin-flop coupling is not a necessary requirement for exchange bias. In the present calculation, the magnetic moment of the uncompensated AFM defect points roughly along the AFM easy axis. For $\phi_H = 10°$ the amount of uncompensated AFM magnetization projected onto the applied field axis is about 1% of the moments in a CoO monolayer and thus comparable to the amount Takano *et al.* [9] have measured. With this choice of arrangement, the calculated and experimentally measured loop shift should be of comparable magnitude, which is

TABLE II. H_c for flat CoO/Py interface as well as H_c and H_{eb} for interface with uncompensated AFM defects (in Oe).

AFM easy axis	Flat interface H_c	Interface with defects H_c	H_{eb}
$[\bar{1}\,\bar{1}7]$	885	575	75
$[\bar{1}01]$	1625	1250	74

indeed the case for the results given in Table II and the values for H_{eb} given by Takano *et al.* [9]. Introducing uncompensated defects also reduces the coercivity. This is because the exchange field is disturbed locally, which reduces the energy barrier that must be overcome in order to switch the FM magnetization. However, H_c in Table II is still much larger than the values actually measured for H_c in CoO/Py films [4] ($\approx$200 Oe for 200 Å of Py). The likely explanation for this is that coherent rotation is not the actual reversal mechanism in these films.

More realistically, the reversal is nucleated at film edges or at defects and then proceeds via propagation of FM domain walls. The coercivity arises from the pinning of these domain walls at defects in the film. A direct microscopic description of such a mechanism is not presently feasible and the effect of spin-flop coupling on the coercivity has to be discussed indirectly. The effective coupling strengths determined for 200 Å (Table I) correspond to an averaged uniaxial anisotropy constant $K_{\text{spin-flop}} \approx 5 \times 10^5$ erg/cm^3 which is much larger than the anisotropy of Py ($K_{\text{Py}} \approx 20 \times 10^3$ erg/cm^3 for the bulk). Since the AFM-FM coupling energy is concentrated in the interface region, the averaged value, $K_{\text{spin-flop}}$, is only a lower estimate of the real anisotropy that is induced in the FM near the interface. The spin-flop coupling thus reduces the size of the domain walls which in turn increases the density of defects that can pin the domain wall and thus increases the coercivity. As in the case of coherent rotation, uncompensated defects at the interface give rise to an effective field which will shift the magnetization curve. The origin of such defects may be simply interface roughness which, in the spirit of Malozemoff's theory, would lead to such an effective field. Other defects, such as dislocations at the interface, could also induce locally uncompensated regions which, in the presence of the cooling field, are oriented preferentially and thus break the symmetry. Thus, while the strong increase in the coercive field is an immediate consequence of the direct AFM-FM coupling, exchange bias requires indirect coupling through a defect. This would explain, why H_{eb} can be strongly reduced when CoO/Py films are annealed while H_c remains essentially unchanged [14]. In general, however, the AFM-FM coupling may also depend on the interface morphology. The experimental observation that coupling constants determined from reversible techniques are much larger than those derived from H_{eb} is an immediate consequence of the different origins of H_{eb} and the coupling. While reversible techniques measure the contribution of both the uniaxial (spin-flop coupling) and the unidirectional (H_{eb}) anisotropies, the irreversible techniques measure only the latter. In CoO, as is indicated by the large difference between reversible and irreversible coupling measurements, the unidirectional contribution is much smaller than the uniaxial coupling. Therefore, the reversible measurements sense mainly the uniaxial coupling, which for 200 Å of Co is [17] $2K_{\text{eff}} \approx 1$ erg/cm^2 [18]. The present theory thus not only explains qualitatively the difference between reversible and irreversible experiments but also predicts coupling constants (Table I) with the correct order of magnitude.

In conclusion, it has been shown that spin-flop coupling between FM and AFM films gives rise to a uniaxial rather than a unidirectional anisotropy. A necessary consequence is that additional mechanisms, such as those induced by interfacial defects, are required to shift the magnetization curve for exchange bias. For CoO the present calculations yield realistic values for the coupling constants as well as the loop shift, and explain the increase in coercivity due to the AFM-FM coupling.

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[17] Figure 2 in Ref. [11] extrapolated to a 200 Å thick Co film. The dependence of K_{eff} on the Co film thickness will be discussed elsewhere.
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