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Exchange coupling and coercivity in ferro-antiferromagnetic bilayers

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Abstract

A microscopic Heisenberg model which includes the magnetostatic interactions is constructed for CoO/Co bilayers. The Landau–Lifshitz equation of motion is used to study the equilibrium properties of spin configurations and it is shown how magnetization curves can be calculated. Flat interfaces with compensated antiferromagnetic interface planes show spin-flop coupling but no exchange bias. The magnetization reversal is however irreversible with very large coercivity. © 1999 Elsevier Science B.V. All rights reserved.

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Bilayers in which a ferromagnetic (FM) film is in contact with certain antiferromagnetic materials (AFM) that have been ordered in presence of the FM or an external field, show a shift in the magnetization curves away from zero field axis which is commonly referred to as exchange bias [1]. The effect is naturally thought to be a consequence of the AFM–FM exchange coupling. However, when the AFM interface plane is compensated, which is in fact the case for most realistic interfaces, it is not obvious how microscopic exchange can lead to a macroscopic coupling between the AFM and the FM. A solution for this long-standing problem has been given by Hinchey and Mills [14] and independently by Koon [2] who showed with a generic Heisenberg model that the FM minimizes the energy when it aligns perpendicular to the AFM easy axis. This coupling is due to frustration of interfacial spins and its similarity to the classical spin-flop state has led to the name 'spin-flop coupling'. With this effective AFM–FM coupling established, Koon also showed that his model yields a shifted magnetization loop through a mechanism proposed by Mauri et al. [3], in which the motion of the spins is assumed to be constrained parallel to the interface such that a domain wall forms in the AFM during the rotation of the FM magnetization. The resulting magnetization curve of the AFM–FM bilayer is shifted from the zero field axis and reversible. Although the perpendicular coupling between the FM and the AFM easy axis has been observed for several AFMs that show exchange bias [4–7], it is unclear whether spin-flop coupling really leads to exchange bias. From a theoretical perspective it is not obvious that the domain wall which is wound up in the AFM during field reversal is stable. A proper manner to investigate this is to replace the constraint on the spin motion during field reversal by the actual demagnetizing field and use an equation of motion for the time evolution of the spin configuration. In the present article we include the magnetostatic interactions into the Heisenberg model and show how the Landau–Lifshitz equation can be used to study magnetic properties of the AFM–FM bilayer from a microscopic point of view. The results presented here are for CoO/Co bilayers, a system which shows both, exchange bias and perpendicular coupling [7].

For CoO we use model parameters that have been extracted from neutron scattering measurements [9]. The Co^{2+} ions have a moment of 3.8 μ_B, are located on an FCC lattice ($a = 4.27$ Å) with next nearest neighbor exchange interactions $J_{A\text{-}A} = -2$ meV, and a uniaxial anisotropy that has the easy axis along the $[\bar{1}\,\bar{1}\,7]$ and an anisotropy constant of 3 meV. The (1 1 1) plane is

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used for the interface and the Co film is assumed to be coherent with the Co^{2+} sublattice of CoO and the Co layer spacing is chosen such that the bulk atomic volume is conserved. In the model, Co atoms have a moment of 1.7 μ_B, nearest neighbor exchange interactions, J_{F-F}, of 16 meV, and no anisotropy is assumed for these FM sites. So far all parameters are specified by the bulk properties of either CoO or Co. The only parameter that remains to be chosen is the AFM–FM exchange interaction, J_{A-F}. For the present interface it is reasonable to assume that $J_{A-F} = J_{A-A}$ since there is always an O layer between FM Co and AFM Co^{2+} sites. All calculations presented here are for a 200 Å thick Co film. The thickness of the CoO film is 22 Å (9 layers of Co^{2+}) which is a realistic value [8] for CoO/Co and is large enough to prevent the AFM configuration from simply following the FM during magnetization reversal. In what follows we will refer to Co as FM sites and Co^{2+} in CoO as AFM sites.

The energy of a spin configuration, $\boldsymbol{M} \equiv \{\boldsymbol{\mu}_i\}$ where i labels the atomic site, consists of four terms,

$$E[\boldsymbol{M}] = E_Z + E_J + E_A + E_D,$$

of which the first three are, respectively, the Zeeman energy, $E_Z = \sum_i \boldsymbol{\mu}_i \boldsymbol{H}_{ext}$, the exchange energy, $E_J = -\sum_{i \neq j} J_{ij} \boldsymbol{s}_i \boldsymbol{s}_j$, with $\boldsymbol{s} = \boldsymbol{\mu}/|\boldsymbol{\mu}|$, and the anisotropy energy, $E_A = \sum_i K_i \sin^2 \theta_i$. The magnetic moments, μ_i, the exchange parameters, J_{ij}, and the anisotropy constants, K_i, are specified by the model discussed in the previous paragraph. The magnetostatic contribution to the energy is

$$E_D = \sum_{i \neq j} \frac{\boldsymbol{\mu}_i \boldsymbol{\mu}_j - 3(\boldsymbol{\mu}_i \hat{n}_{ij})(\boldsymbol{\mu}_j \hat{n}_{ij})}{|\boldsymbol{R}_i - \boldsymbol{R}_j|^3},$$

where $\hat{n}_{ij} = (\boldsymbol{R}_i - \boldsymbol{R}_j)/|\boldsymbol{R}_i - \boldsymbol{R}_j|$. Since we will consider only systems that are periodic in two dimensions (2D), the lattice sums for E_D will be performed using the Ewald method outlined by Adams and McDonald [10] but adapted to 2D periodic lattices [11]. The magnetic moments are subject to the Landau–Lifshitz equation of motion (EOM) with the Gilbert–Kelley form for the damping term,

$$\frac{\partial}{\partial t} \boldsymbol{\mu}_i = -\gamma(\boldsymbol{\mu}_i \wedge H_i) + \sigma \left(\boldsymbol{\mu}_i \wedge \frac{\partial}{\partial t} \boldsymbol{\mu}_i \right) \frac{1}{|\boldsymbol{\mu}_i|},$$

where $H_i[\boldsymbol{M}] = -\partial/\partial \boldsymbol{\mu}_i E[\boldsymbol{M}]$ is the local magnetic field, γ is the gyromagnetic ratio, and σ is an arbitrary damping parameter. This EOM is usually used in phenomenological micromagnetics calculations at a much larger length scale [12]. At the atomic scale it was used by Kubo and Hashitsume to study the Brownian motion of classical spins [13], where a stochastic term is added to the local field. In this case the friction term is still phenomenological but is justified on the basis of the fluctuation–dissipation theorem and has the purpose of driving the system to equilibrium. In the present case we will primarily be concerned with stability of equilibrium solutions in the limit of zero temperature. The stochastic term is therefore dropped and the damping constant is treated as an arbitrary number. Great care has therefore to be taken that the results are independent of its magnitude.

We consider spin configurations where CoO is in a single domain state with a compensated interface plane. The magnetic unit cell therefore contains only two sites per (1 1 1) layer which greatly simplifies the calculations. This only excludes AFM domain walls that are perpendicular to the interface. The possibility for a domain wall to form parallel to the interface during magnetization reversal is still allowed. In the equilibrium solution for the spin configuration with zero applied field, the AFM moments point along the AFM easy axis, the [$\bar{1}$ $\bar{1}$ 7], and the FM moments are in the film plane and perpendicular to the projection of the AFM easy axis onto the (1 1 1) plane, i.e., they point along the [1 1 0]. There are two degenerate solution for the same AFM configuration as is illustrated in Fig. 1. When an external field is applied the FM magnetization changes to align somewhere between the axis of the field and the spin-flop coupling axis ([1 1 0]). Provided the field is not applied perpendicular to the [1 1 0] the degeneracy of the two solutions is removed.

In Fig. 2 we show the magnetization and energy curves for the case where the field is slowly reversed at a constant rate. The initial condition for this calculation is the solution of the EOM with a constant field of 1 kOe applied in the (1 1 1) plane at an angle $\phi_H = 10°$ from the spin-flop coupling axis. When the field approaches zero magnitude the FM moments align with the spin-flop coupling axis and then slowly start to rotate in the opposite direction. As can be seen from the energy curve, the magnetic configuration is clearly in a metastable state which, when the magnitude of the external field further

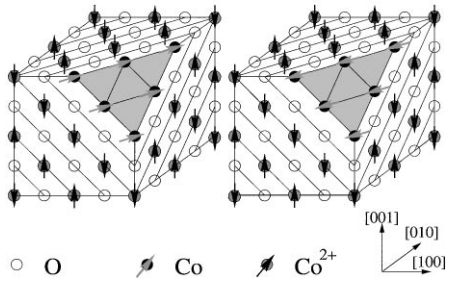

Fig. 1. Schematic of equilibrium spin configuration in CoO–Co (1 1 1) bilayers. For clarity of presentation the AFM moments are drawn to point along the [0 0 1] instead the [$\bar{1}$ $\bar{1}$ 7] and the relaxation of moments near the interface is neglected. The shaded layer is the first FM Co layer.

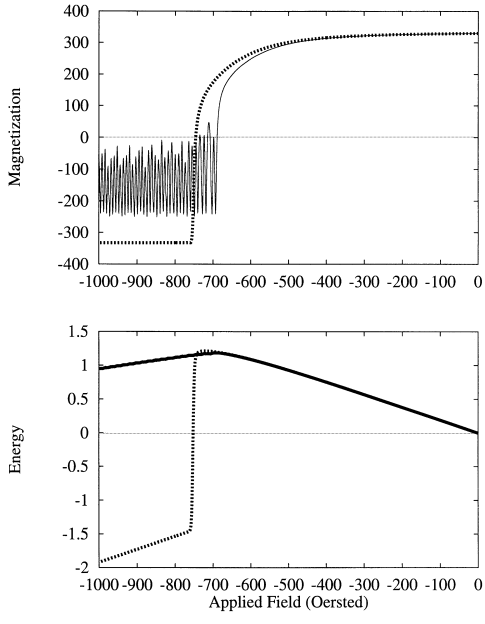

Fig. 2. Magnetization (upper panel) and energy (lower panel) for applied field decreased at slow but constant rate (energy zero is arbitrarily set to value at zero external field). Solutions of the undamped and damped EOM are represented by solid and dashed lines, respectively.

increases, eventually becomes unstable. At this point the solution for the damped EOM evolves into the second spin-flop state, which now has minimum energy, while in the undamped EOM the configuration remains unstable, since the total energy of the system is conserved. Note that in Fig. 2 the constant change in energy of the undamped solution is due to the changes in the external field. The exact form of the magnetization curve depends on the rate at which the applied field is changed and on the magnitude of the damping term. Since no physical studies of the dynamics can be performed with the present form of the dissipative term in the EOM, a different approach has to be used to calculate the magnetization curve. We note however, that despite this restriction the actual result that the remanent state becomes unstable for large applied field has a proper physical meaning.

A well-defined way to determine the magnetization curve is to calculate the solutions of the EOM at constant applied field. One starts with a solution for a given field, changes the magnitude of the applied field and solves the EOM with the old solution as an initial condition. This procedure is independent of the magnitude of the damping parameter in the EOM. The point at which the metastable solution becomes unstable can be determined to arbitrary accuracy by decreasing the steps in the applied field. Such a magnetization curve, for the case where the field is applied in the (1 1 1) plane with $\phi_H = 10°$, is shown in Fig. 3. Since the configuration at

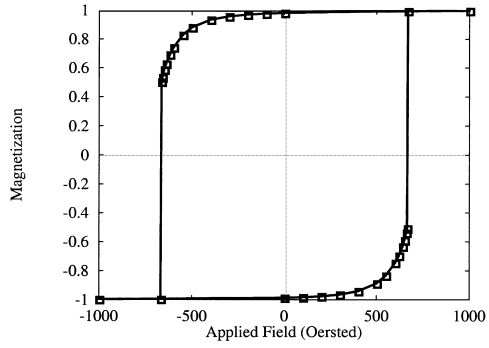

Fig. 3. Full MH-loop for an applied field changing at arbitrarily slow rate.

the endpoints of the magnetization curves are the two spin-flop states of Fig. 1, which are the respective lowest energy states, no energy is stored in the system and the magnetization curves are symmetric with respect to the zero field axis. The flat CoO/Co interface does not show exchange bias despite the spin-flop coupling. However, since the remanent state is stable up to a rather large value of the applied field, the coupled AFM–FM bilayer shows a coercive field (H_c) which is large. For $\phi_H = 0°$ the shape of the loop is similar where H_c is about 10–20% larger. On the other side, when $\phi_H = 90°$, the magnetization curve is closed and has the expected shape of the hard axis loop.

The easy axis direction in CoO films may not be the same as in bulk. In fact, for CoO/Fe_3O_4 multilayer with [0 0 1] growth direction, the easy axis is along the [1 1 0] in the interface plane [5]. While the choice of AFM easy axis in the model does affect the results quantitatively, the calculations show that it has no effect on the qualitative aspects. Even when the easy axis is chosen to be parallel to the interface plane the domain wall does not form in the AFM and the magnetization reversal remains an irreversible process.

Our results for the equilibrium solution agree with Koon's [2], i.e. the FM aligns perpendicular to the AFM easy axis, but they are qualitatively different when it comes to the properties during magnetization reversal, since in our case no domain wall forms in the AFM when the FM magnetization switches. To show, that this is not a consequence of the different AFM used in the study, we have repeated Koon's calculations[1] using the EOM approach. The resulting magnetization curves are shown in Fig. 4. Koon's reversible magnetization curve, with

[1] See Ref. [2] for details. Note that in our case the geometry of the lattice explicitly enters the expression for the energy. In order to simplify the interpretation of the results, a square lattice was used to ensure that the magnetostatic contribution is isotropic in the plane parallel to the interface.

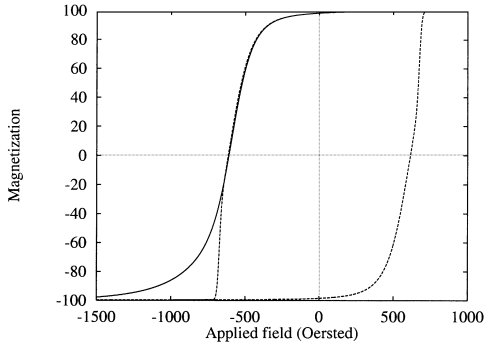

Fig. 4. Magnetization curves for applied field changed at slow but constant rate. Solid lines represent reversible solution of EOM without precession; Dotted lines represent the irreversible solution of the full EOM.

a domain wall forming in the AFM during field reversal, could only be generated when the precession term in the EOM was suppressed[2] and the initial spin configuration had all moments perfectly parallel to the interface plane. In this case the torque on the spins has not component out of the interface plane and therefore the spin motion is restricted to be parallel to the interface plane. The magnetization curves are irreversible when the full EOM (including precession) is used. This is because spins in the interface region can move out of the plane and rearrange in such a way that the systems evolves into the second spin-flop state which corresponds to the initial state but with reversed FM magnetization.

In conclusion, the present example of CoO/Co shows that the introduction of a domain wall into the antiferromagnet during magnetization reversal is not obvious from an atomic-level perspective. Spin-flop coupling by itself does not lead to exchange bias, but rather extrinsic effects related to interfacial roughness or defects would have to be introduced to break the symmetry. However, the results reveal that a large coercivity is intrinsically related to the spin-flop coupling and that the magnetization curves of the coupled AFM–FM system are naturally irreversible. Quantitative results and a more detailed study of the sensitivity to ambiguous model parameters, such as the AFM anisotropy direction and the AMF–FM exchange coupling, will be presented elsewhere.

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References

[1] W.H. Meiklejohn, C.P. Bean, Phys. Rev. 105 (1957) 904.
[2] N.C. Koon, Phys. Rev. Lett. 78 (1997) 4865.
[3] D. Mauri, H.C. Seigmann, P.S. Bagus, E. Kay, J. Appl. Phys. 62 (1987) 3047.
[4] T.J. Moran, I.K. Schuller, J. Appl. Phys. 79 (1996) 5109.
[5] Y. Ijiri, J.A. Borchers, R.W. Erwin, S.-H. Lee, P.J. van der Zaag, R.M. Wolf, Phys. Rev. Lett. 80 (1998) 608.
[6] T.J. Moran, J. Nogués, D. Lederman, I.K. Schuller, Appl. Phys. Lett. 72 (1998) 617.
[7] J.A. Borchers, Y. Ijiri, S.-H. Lee, C.J. Majkrzak, K. Takano, R. Kodama, A.E. Berkowitz, J. Appl. Phys., submitted for publication.
[8] B.H. Miller, E. Dan Dahlberg, Appl. Phys. Lett. 5003 (1996).
[9] V. Wagner, D. Hermann-Ronzaud, Neutron Inelastic Scattering 1977 Part II, 1971, p. 135–143.
[10] D.J. Adams, I.R. McDonald, Mol. Phys. 32 (1976) 931.
[11] T.C. Schulthess, unpublished.
[12] L.D. Landau, E.M. Lifshitz, Statist. Phys., Part 2.
[13] Ryogo Kubo, Natsuki Hashitsume, Prog. Theor. Phys. Suppl. 46 (1970) 210.
[14] L.L. Hinchey, D.L. Mills, Phys. Rev. B. 34 (1986) 1689.

[2] In the present case we have actually removed the precession term for the EOM. This corresponds to the limit of a very large damping constant which we consider to be unphysical.