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A quantum exchange bias model

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Abstract

The origin of the exchange bias phenomenon is investigated on the basis of a quantum mechanical model. In particular, the mechanisms that determine the magnetic structure in the vicinity of an antiferromagnetic–ferromagnetic interface are reexamined. This way we establish how the breaking of translational invariance modifies quantum spin fluctuations. It is found that non-uniform fluctuations induce uncompensated spins in the antiferromagnet, which in turn give rise to a dipole field that couples to the magnetization of the ferromagnet. This coupling yields an exchange bias field that is of the order of magnitude of the one observed experimentally. A net surface magnetization should also be experimentally observable in a clean antiferromagnetic surface. © 2007 Elsevier B.V. All rights reserved.

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1. Introduction

The origin of the exchange anisotropy phenomenon is the interaction of two differently ordered magnetic materials which are in contact, usually one ferro- and the other antiferromagnetic, which have been cooled below their ordering temperatures $T_{\rm C}$ and $T_{\rm N}$ (Curie and Néel, respectively) in an external magnetic field. It has been observed in clusters or small particles, ferromagnetic (FM) films deposited on single-crystal or polycrystalline antiferromagnetic (AF) substrates, FM/AF thin film bilayers, and spin glasses. The most characteristic signature of exchange bias (EB) is that the center of the hysteresis loop is shifted by an amount called the EB field $H_{\rm EB}$, as illustrated in Fig. 1. With the convention that the positive field direction is that of the cooling field, $H_{\rm EB}$ is usually negative.

Although exchange anisotropy has attracted the attention of physicists and material scientists for half a century [1-4] and has resulted in extensive technological applications in the storage and sensor industries [5,6], a full

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understanding of its physics has not yet been achieved. Experimentally, it has been ascertained that EB is related to the spin ordering on the AF side of the interface [7,8]. However, the nature of this arrangement, and the precise microscopic mechanism that determines the magnitude of the EB field, are still open questions. Several EB theories have been advanced with varying degree of success [9–17]. Early models [11,12], which yield much too large values of $H_{\rm EB}$, assume a domain wall (DW) in the antiferromagnet. Koon [10] gave strong arguments in favor of the FM/AF interface structure sketched in Fig. 2, with the FM bulk magnetization orthogonal to the bulk AF easy axis. This was confirmed theoretically [15–17] and experimentally [18] for Fe/FeF_2 and also for the Fe_3O_4/CoO systems [19]. Recent experimental neutron reflectometry [20] and reversible anisotropic magnetoresistance [21] results confirmed the presence of a DW in the FM slab. Another important experimental information [21,22] is that $H_{\rm EB} \propto t_{\rm F}^{-1}$. Moreover, quite a number of semi-classical models have been put forward [4], an exception being the contribution of Suhl and Schuller [23] who interpreted the exchange field as a self-energy shift due to the emission and reabsorption of AF spin waves.



Fig. 1. (Color online) Idealized representation of a hysteresis loop EB shifted by a field of magnitude $H_{\rm EB}$.



Fig. 2. (Color online) Cartoon of the magnetic configuration in the vicinity of a FM/AF interface. Notice the canting of the spins in the AF monolayer closest to the interface.

In this paper, we will succinctly review semi-classical approaches that have been put forward and present, in detail, a theory based on the idea that quantum fluctuations lead to a two-dimensional dipole moment density in the AF. This, in turn, generates a dipolar field which can account for the observed values of the EB anisotropy.

Semi-classical EB theories basically rely on some sort of rearrangement of the magnetic ordering in the vicinity of the interface, either in the FM [10], in the AF [11,12,24] or in both [4,15–17]. Essentially the interfacial magnetic structure rearrangement gives rise to an exchange field which couples the FM and the AF, generating a transition from uniaxial to unidirectional anisotropy. The basic idea behind this type of models is that as the sample is field cooled, from a temperature $T_{\rm C} > T > T_{\rm N}$ to temperatures below $T_{\rm N}$, a spin glass structure develops in the immediate vicinity of the interface. The stability of this magnetic spin glass configuration is due to the FM/AF interface lattice mismatch and the presence of defects in the interfacial region. They, in turn give rise to a metastable magnetic interface configuration and consequently to quite stable pinning centers. This picture is consistent with the training and thermal memory effects, and allows to understand them.

In what follows we venture into the formulation of an alternative model for EB, exclusively quantum mechanical in origin.

2. Quantum theory

Here we put forward the idea that quantum fluctuations lead to a two-dimensional dipole moment density in the AF, which in turn generates a dipolar field that can account for the EB anisotropy. The relevant order parameter is the staggered magnetization

$$\langle \hat{\mathbf{M}}_{\mathrm{AF}} \rangle = g \mu_{\mathrm{B}} \left\langle \sum_{\alpha} \hat{\mathbf{S}}_{\alpha} - \sum_{\beta} \hat{\mathbf{S}}_{\beta} \right\rangle, \tag{1}$$

where $\hat{\mathbf{S}}_{\alpha}$ and $\hat{\mathbf{S}}_{\beta}$ denote the spin operators at the spin-up and spin-down sublattices. If we assume a Heisenberg Hamiltonian $\mathscr{H} = \sum_{ij} J_{ij} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j$, we can see that $[\hat{\mathbf{M}}_{AF}, \mathscr{H}] \neq 0$, in other words, that $\hat{\mathbf{M}}_{AF}$ is not constant in time. Therefore its time-averaged value must be smaller than its maximum value, even at T = 0. (If N is the number of spins and $\sqrt{S(S+1)}$ is the spin magnitude, the maximum value of $|\hat{\mathbf{M}}_{AF}^{max}| = NS$.) In a translationally invariant system the reduction of the magnetization is equally shared by all spins. Moreover, since the spin up and spin down sublattices are equivalent then $\sum_{\alpha} \hat{\mathbf{S}}_{\alpha} = -\sum_{\beta} \hat{\mathbf{S}}_{\beta}$, that is, the system magnetic moment is zero. But when translational symmetry is broken by surfaces, interfaces, or other defects, a net magnetic moment can appear because of the decrease of the spin-up average need not equal the corresponding spin-down reduction [25–27].

We will show how, for an ideal FM–AF interface, such a net magnetic moment does appear. The resultant AF magnetization is confined to the immediate vicinity of the interface and may be regarded as a two-dimensional dipole density. This dipole sheet produces a magnetic field which interacts with the moments in the FM and contributes an additional term to the magnetic energy. This additional energy can explain EB. Numerical results indicate that the magnitude of this magnetic moment is only a small fraction of that of a single AF spin, which leads to a value of $H_{\rm EB}$ that is consistent with experimental results.

3. Ideal ferromagnetic-antiferromagnetic interface

We now describe our model system, in which atomic spins of magnitude $\sqrt{S(S+1)}$ are located at BCC lattice sites. Our system is divided in two halves by a (001) interface. On one side of the interface nearest neighbor spins are coupled ferromagnetically by the exchange integral $-J_F$. On the other side, nearest neighbor spins are coupled antiferromagnetically by the exchange integral J_A . Across the interface, they are coupled by the exchange integral $-J_0$.

We decompose this BCC lattice into planes parallel to the interface. On the AF side, each of these planes is ferromagnetically ordered and its spin direction alternates from one plane to the next; these planes are combined into pairs and each *pair* is labelled with the index $l \ge 0$, with l = 0 labelling the pair closest to the interface; within each pair we label the corresponding planes with the subscript α for spins up, and β for spins down. In contrast, on the FM side each index (l < 0) denotes a single, spin up, layer. This choice of notation reflects the doubling of the unit cell on the AF side. The Hamiltonian of our system is written as

$$\mathcal{H} = \sum_{l=0}^{+\infty} \sum_{\mathbf{R},\delta} [J_{l,l} \mathbf{S}_{\alpha}(l, \mathbf{R}) \cdot \mathbf{S}_{\beta}(l, \mathbf{R} + \delta) + J_{l,l+1} \mathbf{S}_{\beta}(l, \mathbf{R} + \delta) \cdot \mathbf{S}_{\alpha}(l+1, \mathbf{R})] + \sum_{l=-\infty}^{-1} \sum_{\mathbf{R},\delta} J_{l,l+1} \mathbf{S}_{\alpha}(l, \mathbf{R} + \delta) \cdot \mathbf{S}_{\alpha}(l+1, \mathbf{R}),$$
(2)

where $\mathbf{R} = a(n_1\hat{\mathbf{x}} + n_2\hat{\mathbf{y}})$ specifies a two-dimensional lattice point, *a* is the lattice constant, n_1 and n_2 are integers, $\delta = a(\pm \hat{\mathbf{x}} + \pm \hat{\mathbf{y}})$, and $\mathbf{S}_A(l, \mathbf{R}) [\mathbf{S}_B(l, \mathbf{R})]$ is a spin in the α (β) plane of the *l*th pair (and at site **R** in that plane).

We use the Holstein–Primakoff transformation to rewrite the Hamiltonian in terms of a, a^{\dagger} , b and b^{\dagger} boson destruction and creation operators. Spin wave interactions are neglected, thus discarding quartic and higher order terms. To take advantage of the in-plane translational symmetry we write the boson operators as functions of the layer index l and the two-dimensional wave vector \mathbf{k} . The Hamiltonian then decouples into a set of independent semiinfinite chains Hamiltonians, each one corresponding to a specific wave vector \mathbf{k} . These chains are conveniently analyzed using Green functions, which we define as

$$G_{ll'}^{aa}(\omega, \mathbf{k}) = -\frac{\mathrm{i}}{\hbar} \int_{-\infty}^{\infty} \mathrm{d}t \, \mathrm{e}^{\mathrm{i}\omega t} \theta(t) \langle [a(l, \mathbf{k}, t), a^{\dagger}(l', \mathbf{k}, 0)] \rangle, \qquad (3)$$

where $\langle A \rangle$ denotes the thermal average of A and the operators are in the Heisenberg picture. The functions $G_{ll'}^{bb}$, $G_{ll'}^{ab}$, and $G_{ll'}^{ba}$ are defined analogously. We use the transfermatrix method [28,29] to obtain analytic expressions for these Greens functions. When the interface coupling is FM we find the following expressions for the diagonal elements of the Greens function

$$G_{ll}^{aa}(z,\mathbf{k}) = G_{\rm AF}(z,\mathbf{k})[1 - T_{\rm AF}^{2l}(z,\mathbf{k})f_{\rm R}(z,\mathbf{k})], \quad l \ge 0, \tag{4}$$

$$G_{ll}^{bb}(z, \mathbf{k}) = G_{\rm AF}(z, \mathbf{k})[1 - T_{\rm AF}^{2l+1}(z, \mathbf{k})f_{\rm R}(-z, \mathbf{k})], \quad l \ge 0, \quad (5)$$

$$G_{ll}^{aa}(z, \mathbf{k}) = G_{\rm F}(z, \mathbf{k})[1 - T_{\rm F}^{-2(l+1)}(z, \mathbf{k})f_{\rm I}(z, \mathbf{k})], \quad l \le -1.$$

$$[z, \mathbf{k})[1 - I_{\mathrm{F}} \land (z, \mathbf{k})]_{\mathrm{L}}(z, \mathbf{k})], \quad l \leq -1.$$
(6)

In these $G_{AF}(z, \mathbf{k})$ is the diagonal (l = l') element of the bulk Greens function for the AF and $G_F(z, \mathbf{k})$ the diagonal element of the bulk FM Greens function. From these diagonal elements and the transfer matrices $T_{AF}(z, \mathbf{k})$ and $T_F(z, \mathbf{k})$ one readily obtains the full bulk Greens functions. Physically the transfer matrices describe plane-wave propagation in one-dimensional chains. Indeed, it is possible to define a one-dimensional wave vector k_z by the relation $T = \exp(ik_z)$. The interference effects at the right and left sides of the interface are contained in the functions $f_R(z, \mathbf{k})$ and $f_L(z, \mathbf{k})$. More explicitly

$$G_{\rm AF}(z,\mathbf{k}) = \frac{z+2J_{\rm A}}{Q_{\rm A}(z,\mathbf{k})},\tag{7}$$

$$Q_{\rm A}(z,\mathbf{k}) = [(z^2 - 4J_{\rm A}^2)(z^2 - 4J_{\rm A}^2 + 4J_{\rm A}^2|\gamma_{\mathbf{k}}|^2)]^{1/2}, \tag{8}$$

$$T_{\rm AF}(z,\mathbf{k}) = \frac{1}{2J_{\rm A}^2 |\gamma_{\mathbf{k}}|^2} [4J_{\rm A}^2 - z^2 - 2J_{\rm A}^2 |\gamma_{\mathbf{k}}|^2 + Q_{\rm A}(z,\mathbf{k})], \quad (9)$$

$$G_{\rm F}(z,\mathbf{k}) = \frac{1}{Q_{\rm F}(z,\mathbf{k})},\tag{10}$$

$$Q_{\rm F}(z, \mathbf{k}) = \left[(z - 2J_{\rm F} - 2J_{\rm F} |\gamma_{\mathbf{k}}|)(z - 2J_{\rm F} + 2J_{\rm F} |\gamma_{\mathbf{k}}|) \right]^{1/2},$$
(11)

$$T_{\rm F}(z,\mathbf{k}) = \frac{1}{2J_{\rm F}} |\gamma_{\bf k}|^2 [2J_{\rm F} - z + Q_{\rm F}(z,\mathbf{k})], \qquad (12)$$

$$f_{\rm R}(z,\mathbf{k}) = \frac{(z-2J_0+Q_{\rm F})[(z+2J_{\rm A})(z-2J_0)-Q_{\rm A}]-4J_0^2|\gamma_{\rm k}|^2(z+2J_{\rm A})}{(z-2J_0+Q_{\rm F})[(z+2J_{\rm A})(z-2J_0)+Q_{\rm A}]-4J_0^2|\gamma_{\rm k}|^2(z+2J_{\rm A})}$$
(13)

and

$$f_{\rm L}(z,\mathbf{k}) = \frac{(z-2J_0-Q_{\rm F})[(z+2J_{\rm A})(z-2J_0)+Q_{\rm A}]-4J_0^2|\gamma_{\rm k}|^2(z+2J_{\rm A})}{(z-2J_0+Q_{\rm F})[(z+2J_{\rm A})(z-2J_0)+Q_{\rm A}]-4J_0^2|\gamma_{\rm k}|^2(z+2J_{\rm A})}.$$
(14)

When the interface coupling is AF we obtain very similar expressions, the only difference being that the exponents of T_{AF} in Eq. (9), and T_{F} in Eq. (12), are shifted by -1 and +1, respectively.

The average spin on the *l*th plane is finally given by

$$\langle S_{l,a}^{z} \rangle = S + \sum_{\mathbf{k}} \frac{1}{\pi} \operatorname{Im} \int_{-\infty}^{\infty} \mathrm{d}\omega \frac{G_{ll}^{aa}(\omega, \gamma_{\mathbf{k}})}{e^{\hbar\omega/k_{\mathrm{B}}T} - 1},$$
(15)

$$\langle S_{l,b}^{z} \rangle = -S - \sum_{\mathbf{k}} \frac{1}{\pi} \operatorname{Im} \int_{-\infty}^{\infty} \mathrm{d}\omega \frac{G_{ll}^{bb}(\omega, \gamma_{\mathbf{k}})}{e^{\hbar\omega/k_{\mathrm{B}}T} - 1}.$$
 (16)

For our numerical calculations we choose units such that $J_F + J_A = 1$. We use as our parameters J_0 and $x \equiv J_F - J_A$. In Fig. 3 we show the *net* magnetization in the AF side of the interface, as a function of x and $J_0 > 0$. In Fig. 4 we show the corresponding results for $J_0 < 0$.

For this specific model, we observe that the net magnetization per atom is of the order of a few percent. Coincidentally, Bulut et al. [26] investigated the magnetization around a vacancy in a 2D AF obtaining results consistent with ours, but for a very different spin arrangement. In addition, we notice that in these systems the correlation length, away from the critical points, is of the order of the interatomic distance. Therefore the results for the idealized interface studied here should hold for nonideal interfaces as well.

3.1. Surface dipole density and exchange bias

The uncompensated AF spins give rise to a dipolar magnetic field, \mathbf{B}_{AF} . This results in a Zeeman contribution to the energy density inside the FM, given by $-\mathbf{M}_{F} \cdot \mathbf{B}_{AF}$, where \mathbf{M}_{F} is the FM magnetization. This allows us to estimate the interface anisotropy energy per unit area, ΔE [30,31]. To obtain this estimate we assume a circular AF



Fig. 3. (Color online) Net magnetization in the antiferromagnetic side of the interface as a function of $x = (J_F - J_A)/(J_F + J_A)$ and $J_0/(J_F + J_A)$. The units we choose correspond to $J_F + J_A = 1$. The coupling across the interface is ferromagnetic.



Fig. 4. (Color online) Net magnetization in the antiferromagnetic side of the interface as a function of $x = (J_F - J_A)/(J_F + J_A)$ and $J_0/(J_F + J_A)$. The units we choose correspond to $J_F + J_A = 1$. The coupling across the interface is antiferromagnetic.

domain of radius *R*. On this domain there is a uniform dipole distribution $\sigma \sim 4g\mu_{\rm B}\delta S/a^2$, due to the uncompensated fluctuations (here *g* is the gyromagnetic ratio, $\mu_{\rm B}$ is the Bohr magneton, and *a* is the distance between neighboring spins). Adjacent to this domain there is a semi-infinite cylindrical ferromagnet, also of radius *R*. This FM is assumed to be in a single domain state, of magnetization $M_{\rm F}$ parallel to the interface and at an angle ϕ with respect to the AF positive axis. Integration of the Zeeman energy density over the cylinder yields

$$\Delta E = \alpha (M_{\rm F} \mu_0 g \mu_{\rm B} \delta S / a^2) \cos \phi. \tag{17}$$

The constant α is the adimensional integral that results from scaling distances by *R* and angles are by 2π . Numerical computation yields $\alpha = 0.47$ [32].

With $\delta S \sim 10^{-2}$, $a \sim 0.1$ nm, and M_F equal to the saturation magnetization of Co, we find that $\Delta E \sim 0.08 \text{ ergs/cm}^2$. This falls within the range of experimentally observed [30,33] interface energies.

The AF free surface is a special case which corresponds to the $J_0 = 0$ limit of the model presented here. As can be seen in Figs. 3 and 4, this free surface also develops a net magnetization. Takano et al. [34] have observed a thermoremanent magnetization at the free surface of CoO films that have been field cooled through the Néel temperature. They also studied bilayers of $Ni_{81}Fe_{19}/CoO$ and found that the EB field, after field cooling, has the same temperature dependence as the free film thermoremanent magnetization. This suggests that uni-directional anisotropy arises, in the context of our model, due to the coupling between the FM and the AF *net* adjacent surface magnetizations.

In nanoparticles, in which the surface to bulk ratio is significant, a surface magnetization can be observed. A net magnetization has indeed been reported in AF ferritin [35] and ferrihydrite [36] nanoparticles. With $S = \frac{5}{2}$ and $\delta S = 0.03$ we estimate magnetizations per particle of $258\mu_{\rm B}$ for ferrihydrite and $368\mu_{\rm B}$ for ferritin. These are consistent with the experimental values of $250\mu_{\rm B}$ [36] for ferrihydrite and $345\mu_{\rm B}$ [35] for ferritin.

4. Summary and conclusion

In summary, after a brief review of experimental results and theoretical models of exchange bias, we have put forward an alternative mechanism to generate exchange anisotropy in a system where an antiferromagnet (AF) is in contact with a metallic ferromagnet (FM). It is based on the fact that ground-state fluctuations reduce the zerotemperature magnetic moments of the spins in a quantum AF, giving rise to an exchange field, an alternative that to the best of our knowledge had not been explored before. The model put forward here differs from the available literature in that it does not depend on interface lattice mismatch nor defects in the vicinity of the interface, but only on the presence of the latter. In fact, in the vicinity of interfaces, and other defects which break translational symmetry, the above quantum fluctuations are not uniform. Consequently, the magnetic moments of oppositely oriented spins do not compensate exactly, as they do in a bulk AF. Thus, close to a surface or interface, a relatively small magnetic dipole density is generated.

We have shown that this dipole field yields an interface energy which is of the same order of magnitude as the one obtained in experimental measurements, which suggests that a better understanding of exchange bias, and the underlying physical phenomena, could be achieved by means of magnetization measurements, Brillouin lightscattering, and spin polarized electron energy loss spectroscopy of clean AF surfaces.

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