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Positive exchange bias model: Fe/FeF_2 and Fe/MnF_2 bilayers

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Abstract

Positive exchange bias (PEB) is a remarkable phenomenon, which was recently observed experimentally. Normal (negative) exchange bias (NEB) was discovered more than 40 years ago. Its signature is the shift of the hysteresis loop along the applied field axis by $H_E < 0$, in systems where a ferromagnet (FM) is in close contact with an antiferromagnet (AFM). This occurs after the system is cooled below the Néel temperature in an external field H_{cf} of a few kOe. As H_{cf} is substantially increased H_E adopts positive values. Here we explain this rather unexpected behavior on the basis of an incomplete domain wall model that develops in the FM, for Fe/FeF_2 and Fe/MnF_2 systems. A consistent and unified picture of both NEB and PEB, and satisfactory quantitative agreement with experimental results are obtained on the basis of our theory. © 2000 Published by Elsevier Science Ltd.

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1. Introduction

Exchange bias (EB) is a phenomenon that occurs in systems where a ferromagnet (FM) is in atomic contact with an antiferromagnet (AFM). When a sample with a FM/AFM interface is cooled below the Néel temperature T_N (assuming that the FM Curie temperature is larger than T_N) in a static external magnetic field H_{cf}, the center of the FM magnetization loop (M versus H) shifts away from $H = 0$. The magnitude of this shift is known as the exchange bias field H_E. Since the discovery of EB, by Meiklejohn and Bean [1,2] more than 40 years ago, extensive efforts have been devoted [3] to develop a full understanding of the phenomenon [1,2,4–9], because of its fundamental interest and the important technological applications that it has. Among the latter the most notable are the domain stabilization of magnetoresistive heads [10] and "spin-valve"-based devices [11].

The main feature of normal EB is the displacement of the hysteresis loop to fields that point opposite to the applied cooling field $\vec{H}_{cf}$. This shift of the hysteresis loop in the $-\vec{H}_{cf}$ direction was invariably observed in weak field-cooled samples, until in 1996 Nogués et al. [12] applied strong cooling fields, to the same samples where normal (or negative) exchange bias (NEB) had been measured in weak H_{cf}, and discovered positive exchange bias (PEB) in Fe/FeF_2 and Fe/MnF_2 systems; i.e. a shift to the right of the hysteresis loop ($H_E > 0$).

The discovery of PEB is an interesting and rather unexpected phenomenon in itself, reflected in the fact that it took no less than 40 years between the original discovery [1,2] and the detection of PEB [12]. In addition, it also constitutes an important clue to understand the mechanism that governs EB in general. Theoretically PEB was investigated for uncompensated interfaces, on the basis of a spin wave model, by Hong [13] who generalized work on NEB by Suhl and Schuller [14].

Most of the available EB models (more properly NEB models) make different assumptions on the interface structure [15–17], and postulate a domain wall in the AFM [1,2,4–9,18,19]. However, quite recently we put forward a model based on an incomplete domain wall (IDW) in the FM [20], in order to properly take into account new

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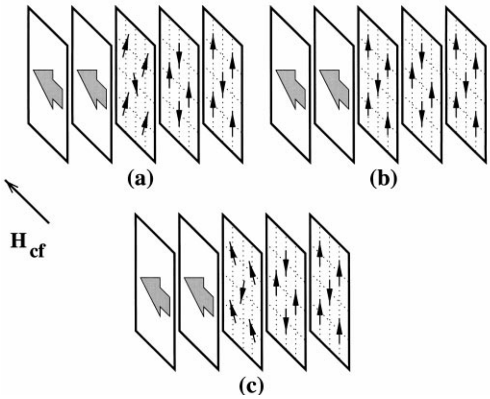

Fig. 1. Spin configuration of the AFM interface monolayer and both the two FM and the two AFM monolayers closest to the interface, after it is field-cooled through T_N. The canting angle θ_c is measured relative to the cooling field $\vec{H}_{cf}$, applied parallel to the $(\bar{1}10)$ AFM crystal direction: (a) corresponds to weak; (b) to the critical; and (c) to strong $|\vec{H}_{cf}|$ values.

experimental information [21–26]. This model, which received strong support by the results recently published by Fitzsimmons et al. [27], assumes that the AFM interface monolayer reconstructs, close to Néel temperature T_N, into an almost rigid canted magnetic structure which freezes, into a spin glass like configuration, as the AFM bulk orders. Moreover, it remains frozen, in a metastable state, during the cycling of the external magnetic field, when performed for $H < H_{cf}$. The analytic formulation of our model was outlined in Ref. [20] and some of its consequences explored in Ref. [28]. In particular, the magnetization cycle was obtained both by numerical simulation (simulated annealing) and by an analytic micromagnetic calculation which is carried out solving a system of nonlinear equations. The theory incorporates only one adjustable parameter: the FM/AFM interface exchange coupling and yields values of H_E which are in excellent agreement with experiment [20].

This paper is organized as follows. After the introduction in Section 1 our model is presented and results are obtained in Section 2, and the contribution is closed, with a brief discussion of the main results, in Section 3.

2. Model and results

In this paper we present an IDW model for PEB that relies on the same hypothesis outlined above [20], and which fits quite well with most of the available experimental information on PEB [12,24,25], with the same parameter values of our previous work on NEB [20,28]. As our prototype systems we have chosen Fe deposited on the (110) crystal face of FeF_2 and MnF_2 [12]. They share a very small AFM/DW width, (of the order of monolayers) and a well characterized, controlled and simple FM/AFM interface structure [3]. As in the experiment we assume that the samples are cooled from 120 to 10 K, through the FeF_2 or MnF_2 Néel temperature ($T_N \approx 79$ K for FeF_2 is very close to the MnF_2 value, which is $T_N \approx 72$ K) in a cooling field H_{cf} parallel to the Fe slab surface and large enough to saturate it [12]. In the absence of the Fe slab, or if the interfacial interaction is negligible, the AFM layers order at, or very close to T_N the bulk transition temperature. But, the presence of the Fe slab (which adopts a magnetization perpendicular to AFM bulk easy axis [9,12,15–17]) acts on the AFM interface monolayer, via the interface exchange constant $J_{F/AF}$, generating a canted spin interface configuration. In fact, even for $T > T_N$, the (110) interface AFM layer starts to order in a canted configuration, as illustrated in Fig. 1a and c, since the spins in this monolayer have a different magnetic environment than the bulk spins. This precursor ordering was observed experimentally by Gökemeijer et al. [29] in EB systems where the Curie temperature of the FM is lower than the ordering temperature T_N of the bulk AFM.

Our model is specified analytically by the following Hamiltonian:

$$\hat{\mathbf{H}} = \hat{\mathbf{H}}_{AF} + \hat{\mathbf{H}}_{F/AF} + \hat{\mathbf{H}}_F, \tag{1}$$

where $\hat{\mathbf{H}}_{AF}$, $\hat{\mathbf{H}}_{F/AF}$ and $\hat{\mathbf{H}}_F$ describe the AFM substrate, interface coupling and the FM slab, respectively. For the single interface magnetic cell, illustrated in Fig. 1, they can be written as

$$\begin{aligned}\hat{\mathbf{H}}_{AF} = {} & -J_{AF}[S\hat{e}_{AF}\cdot(\vec{S}^{(\alpha)} - \vec{S}^{(\beta)}) + 2\vec{S}^{(\alpha)}\cdot\vec{S}^{(\beta)}] \\ & - \tfrac{1}{2}K_{AF}[(\vec{S}^{(\alpha)}\cdot\hat{e}_{AF})^2 + (\vec{S}^{(\beta)}\cdot\hat{e}_{AF})^2] \\ & - \tfrac{1}{2}\mu_B g_{AFM}(\vec{S}^{(\alpha)} + \vec{S}^{(\beta)})\cdot\vec{H},\end{aligned} \tag{2}$$

$$\hat{\mathbf{H}}_{F/AF} = -J_{F/AF}(\vec{S}^{(\alpha)} + \vec{S}^{(\beta)})\cdot\vec{S}_1, \tag{3}$$

$$\hat{\mathbf{H}}_F = -2J_F\sum_{k=1}^{N-1}\vec{S}_k\cdot\vec{S}_{k+1} - \sum_{k=1}^{N}\left[\frac{K_F}{H^2}(\vec{S}_k\cdot\vec{H})^2 + \mu_B g_{FM}\,\vec{S}_k\cdot\vec{H}\right]. \tag{4}$$

Above $S = |\vec{S}|$, while μ_B and g denote the Bohr magneton and the gyromagnetic ratios, respectively, and $\vec{H}$ is the external applied magnetic field. In Eq. (2) the unit vector $\hat{e}_{AF}$ defines the AFM uniaxial anisotropy direction, $\vec{S}^{(\alpha)}$ and $\vec{S}^{(\beta)}$ are canted spin vectors in the AFM interface, belonging to the α and β AFM sub-lattices. The vectors $\vec{S}_k$ are the classical spin vectors of the k-th FM layer, with $k = 1$ denoting the FM interface ($1 \le k \le N$). We have adopted the value $N = 65$ for the number of FM layers (which corresponds to an Fe slab width of ≈ 13 nm).

As already discussed above, in the expression for $\hat{H}_{AF}$ we assume that spin canting in the AFM is significant only in the interface monolayer [20]. However, in Eq. (4) we allow the FM spins $\vec{S}_k$ to rotate in each of the $1 \le k \le N$ layers of the Fe slab, parallel to the each other and to the interface.

In Fig. 1 we display the interface spin configuration of the system, after it is field-cooled through T_N, for three different values of H_{cf}. Fig. 1a corresponds to low fields, such that the interfacial exchange energy is larger than the Zeeman energy; on the contrary, in Fig. 1c the Zeeman energy is dominant. Fig. 1b illustrates the configuration that corresponds to the critical H_{cf} value, when a crossover from one regime to the other does occur. In each of these three cases the FM slab magnetization is fully saturated. On the other hand, the AFM spins are fixed, except those at the interface monolayer [20]. Therefore, the only energy difference is due to changes of interface configuration. From Eq. (1), and using the definitions given in Eqs. (2)–(4), we obtain

$$E = 2|J_{AF}|\left[\cos(2\theta) + \cos\left(\theta + \frac{\pi}{2}\right)\right] - K_{AF}\cos^2\left(\theta - \frac{\pi}{2}\right) + (2|J_{F/AF}| - \mu_B g_{AFM} H_{cf})\cos\theta, \quad (5)$$

where E is the energy per spin (constant terms have been omitted). Due to symmetry considerations, we assume that $\theta = \theta^{(\alpha)} = -\theta^{(\beta)}$, where $\theta^{(\alpha)}$ ($\theta^{(\beta)}$) is the angle between $\vec{S}^{(\alpha)}(\vec{S}^{(\beta)})$ and the cooling field $\vec{H}_{cf}$, which is taken as the reference direction. The values we adopt for the Fe/FeF_2 parameters in Eq. (5) are $J_{F/AF} = J_{AF} = -1.2$ meV, $K_{AF} = 2.5$ meV/spin, while for Fe/MnF_2 they are $J_{F/AF} = -0.35$, $J_{AF} = -1.3$ meV and $K_{AF} = 0.35$ meV/spin, exactly as in Ref. [28].

To obtain the energy minimum we set equal to zero the derivative of Eq. (5), which reads

$$\frac{\partial E}{\partial\theta} = (K_{AF} - 4|J_{AF}|)\sin 2\theta - 2|J_{AF}|\cos\theta - (2|J_{F/AF}| - \mu_B g_{AFM} H_{cf})\sin\theta = 0. \quad (6)$$

Eq. (6) embodies the qualitative explanation of the crossover from NEB to PEB. For low cooling fields ($H_{cf} < 2|J_{F/AF}|/\mu_B g_{AFM}$), the energy E is minimum for $\theta = \theta_c > \pi/2$, where θ_c is the canting angle of the AFM interface monolayer measured relative to $\vec{H}_{cf}$ (Fig. 1a). As outlined in Ref. [20] this configuration accounts for NEB. But, when the sample is cooled in high fields ($H_{cf} > 2|J_{F/AF}|/\mu_B g_{AFM}$), the minimum of E shifts to $\theta = \theta_c < \pi/2$, as shown in Fig. 1c. This implies a frustrated configuration of the FM spins relative to the AFM interface, which is due to the large Zeeman energy that tends to align the AFM spins in the external field ($\vec{H}$) direction. Thus, as $|\vec{H}|$ is lowered during the measurement of the hysteresis loop, frustration is relieved through the rotation of the FM spins before $H = 0$ is reached. This explains the onset of PEB. Finally, $H_{cf} = 2|J_{F/AF}|/\mu_B g_{AFM}$ implies $\theta_c = \pi/2$, such that the spin configurations on the $H > 0$ and $H < 0$ sides of the hysteresis loop are mirror images of each other (Fig. 1b). The absence of a symmetry breaking mechanism, which occurs at this critical H_{cf} value, implies the quenching of EB.

Experiments exhibit a quite complex dependence of the magnitude of H_E on interface roughness [12,24]. Nogués et al. [12] relate high growth temperatures with increased interface roughness and observe that low temperature grown samples have smoother interfaces. While in Fe/FeF_2 the maximum value of $|H_E|$ increases with roughness the opposite occurs for Fe/MnF_2. This fact also implies that several different values for the critical H_{cf} are obtained depending, mainly, on the growth temperature of the sample. For example, the critical H_{cf} value of Fe/FeF_2 grown at 300°C is close to 10 kOe, but when the growth temperature is lowered to 200°C PEB is not achieved for fields as high as 70 kOe.

Our model applies to *ideal* flat interfaces. Consequently, we compare our estimates with the results obtained for the most perfect (smoothest) interfaces experimentally reported. We estimate H_{cf} using Eq. (4), which allows to compute the full magnetization M versus applied field H relation. The relevant terms of the Hamiltonian, with the frozen interface, are

$$\epsilon = -h\sum_{k=1}^{N}\cos\theta_k - \sum_{k=1}^{N-1}\cos(\theta_{k+1} - \theta_k) - \kappa\cos\theta_1 - D\sum_{k=1}^{N}\cos^2\theta_k. \quad (7)$$

Above we define the following dimensionless quantities: the energy ϵ in units of J_F, the applied field $h = \mu_B g_{FM} H/2J_F < 10^{-3}$, the effective interface coupling $\kappa = (J_{F/AF}/J_F)\cos\theta_c$, and the anisotropy $D = K_F/2J_F < 10^{-5}$. We differentiate respect to θ_j, to obtain

$$\frac{\partial\epsilon}{\partial\theta_j} = h\sin\theta_j - (1 - \delta_{j,N})\sin(\theta_{j+1} - \theta_j) + (1 - \delta_{j,1}) \times\sin(\theta_j - \theta_{j-1}) + \delta_{j,1}\kappa\sin\theta_1 + 2D\sin\theta_j\cos\theta_j, \quad (8)$$

where $\delta_{i,j}$ is the Kronecker symbol. Eqs. (7) and (8) are valid for all values of H_{cf}, which is present through κ via the canting angle θ_c. In fact, the sign of κ is the *signature* of EB. If $\kappa > 0$, NEB is obtained, but if $\kappa < 0$ PEB sets in; $\kappa = 0$ determines the crossover from one to the other. To find the minimum energy configuration we set all the Eq. (8) ($1 \le j \le N$) equal to zero.

In the absence of interface coupling ($J_{F/AF} = 0$) or if the sample is field-cooled at the critical H_{cf} [i.e. $\kappa = (J_{F/AF}/J_F)\cos(\pi/2)$] the value $\kappa = 0$ is obtained and the trivial solutions $\theta_k = 0$ or $\theta_k = \pi$, for all k's, are immediately recovered. But, if $\kappa > 0$ at least another solution exists for $h < 0$ yielding NEB [20]. In the $\kappa < 0$ case solutions are obtained for $h > 0$. In both cases the FM spins adopt an incomplete domain wall structure (IDW). For NEB this IDW develops from the free surface, propagating into the FM slab, while for PEB the IDW generates at the interface, due to frustration between the first FM monolayer and the AFM interface layer.

The results for H_E versus cooling field, both for Fe/FeF_2

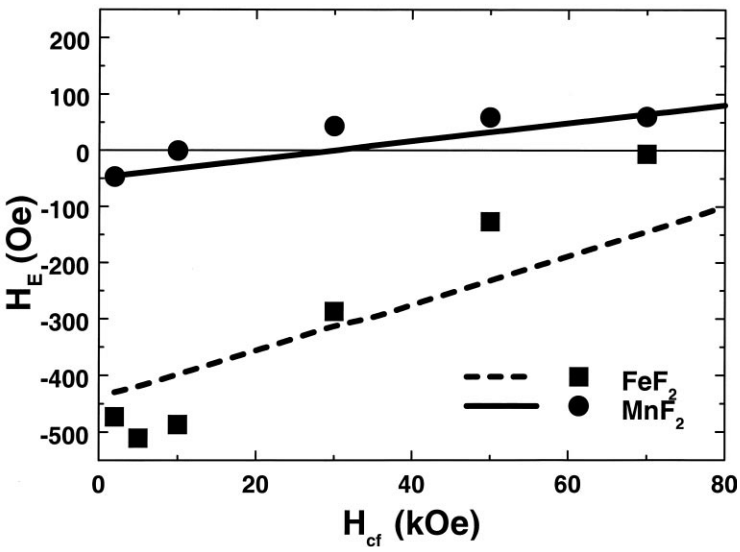

Fig. 2. H_E versus cooling field H_{cf} for Fe/FeF_2 and Fe/MnF_2. The continuous lines and the full circles correspond to Fe/MnF_2 and the dashed line and full squares to Fe/FeF_2. The lines are our theoretical results and the circles and squares correspond to the experiments of Ref. [25].

and Fe/MnF_2, are illustrated in Fig. 2. The fit of our theory of the experimental results, assuming only one adjustable parameter $J_{FM/AFM}$ (for which we also used the same values as in Ref. [28]) is in good agreement with experimental observation for Fe/MnF_2 and Fe/FeF_2.

3. Discussion

Above we have developed a model that provides an explanation of positive exchange bias. It is based on a single assumption: that the AFM interface monolayer reconstructs, close to Néel temperature T_N, into a fairly rigid canted magnetic structure which freezes, into a spin glass like configuration, as the AFM bulk orders [20,28]. Moreover, it remains frozen (in a metastable state) due to the presence of interface pinning centers, during the cycling of the external magnetic field, when performed for $H < H_{cf}$. Our model accounts qualitatively and quantitatively for the crossover of a specific sample from normal ($H_E < 0$) to positive EB, as a function of the cooling field intensity H_{cf}. All the above is achieved without the introduction of a single additional parameter. In fact, the same values of Refs. [20,28] were used, and they in turn only imply a single free parameter: the interface exchange coupling $J_{F/AF}$. Thus, a unified picture of negative and positive EB is provided, that is consistent with the experiment, for systems in which the AFM is strongly anisotropic, like is the case of Fe/MnF_2 and Fe/FeF_2.

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