

## Relaxation times in exchange-biased nanostructures

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We calculated the energy barrier,  $\Delta E$ , for exchange-biased (EB) systems, using the ferromagnetic domain wall model. The temperature dependence of the EB is in good agreement with experimental results. For Fe-FeF<sub>2</sub>, Fe-MnF<sub>2</sub>, and Ni-NiO,  $\Delta E$  is proportional to a power of the interfacial coupling constant and inversely to the ferromagnetic film thickness. The temperature and volume dependence of the relaxation time show that exchange coupling increases the superparamagnetic blocking temperature of nanostructured ferromagnets. © 2003 American Institute of Physics. [DOI: 10.1063/1.1592637]

During the last decade, the areal density of hard disk drives increased over 100% per year and 49.8 Gbit in<sup>-2</sup> has been reported in 2002 by Toshiba. Single bit information is encoded into two magnetic states, separated by an energy barrier. The thermal decay to equilibrium of the magnetic moment of a single-domain ferromagnetic nanoparticle<sup>1</sup> follows a simple Arrhenius switching probability<sup>2</sup> with a relaxation time  $\tau = \tau_0 \exp(\Delta E'/k_B T)$ , where  $k_B$  is the Boltzmann constant,  $T$  is the temperature, and  $\tau_0$  is a constant of the order of 10<sup>-9</sup> s.  $\Delta E' = KV$  is the energy barrier each grain has to overcome to minimize its energy,<sup>3</sup> with  $V$  as an effective switching volume and  $K$  as the magnetic anisotropy per unit volume. A 5% maximal loss over 10 years of stored information at ambient temperatures<sup>4,5</sup> implies  $\Delta E' = 40k_B T$ , then the use of smaller volumes necessarily requires an increase of  $K$ .

In ferromagnetic (FM)-antiferromagnetic (AF) bilayers,  $\Delta E'$  may increase respect to the free FM because of the exchange coupling present.<sup>6</sup> Recently exchange bias (EB) has been intensively studied,<sup>7,8</sup> and models which include different assumptions on the interface structure,<sup>9,10</sup> the formation of AF<sup>11-17</sup> and FM<sup>18-20</sup> domains were proposed. Jensen<sup>21</sup> found an increase of  $\Delta E'$  of a FM particle on top of an AF substrate due to the exchange coupling. However, this model does not exhibit EB.<sup>15</sup>

Here we calculate the temperature dependence of  $\Delta E'$  and  $\tau$  in FM-AF bilayers using the ferromagnetic domain wall model (FM-DW).<sup>20</sup> In this model the first AF interface layer is frozen during the cooling process across the AF Néel temperature  $T_N$  into a canted spin configuration at an angle  $\theta_c$  with respect to the cooling field  $H_{cf}$ . Thus, at  $T < T_N$ , only terms related to the FM will contribute to the energy. In an external field  $H$ , the energy per ferromagnetic spin is given by

$$E = -\frac{2J_F S_F^2}{N_F} \left[ \sum_{k=1}^{N_F-1} \cos(\theta_{k+1} - \theta_k) + \frac{K_F}{2J_F} \sum_{k=1}^{N_F} \cos^2 \theta_k \right] - \frac{2J_F S_F}{N_F} \left[ h \sum_{k=1}^{N_F} \cos(\theta_k - \Phi) + S\kappa \cos \theta_1 \right], \quad (1)$$

where  $N_F$  is the number of FM layers,  $J_F$  is the FM exchange constant, and  $S_F$  and  $S$  are the magnitude of the FM and interface spin at  $T=0$ , respectively. The terms in Eq. (1) describe (1): the exchange interaction between the FM layers, with  $\theta_k$  the angle between the spins in the  $k$ th FM layer and the  $\mathbf{H}_{cf}$  direction,  $k=1$  denoting the FM interface layer; (2) anisotropy energy defined by the ferromagnet's anisotropy constant  $K_F$ ; (3) Zeeman energy proportional to  $h = \frac{1}{2} g_F \mu_B H / J_F$ , with  $g_F$  as the ferromagnet's gyromagnetic ratio,  $\mu_B$  as the Bohr magneton, and  $\Phi$  as the angle between the external magnetic field and  $\mathbf{H}_{cf}$ ; and (4) interface coupling energy with an effective coupling constant (conserving earlier notation)<sup>22</sup>

$$\kappa = \frac{|J_{F/AF}|}{J_F} \left[ \frac{2|J_{F/AF}| - g_{AF} \mu_B H_{cf}}{10|J_{AF}| + 2K_{AF}} \right], \quad (2)$$

with  $J_{F/AF}$  and  $J_{AF}$  as the exchange constants of the interface and AF bulk respectively,  $K_{AF}$  as the anisotropy constant, and  $g_{AF}$  as the gyromagnetic ratio of the antiferromagnet.  $\kappa > 0$  yields negative and  $\kappa < 0$  positive EB. When  $\kappa < \kappa_0 = \sqrt{24/(N_F^2 - 1)}$ , the EB field<sup>22</sup> is

$$H_{EB} = -\frac{2J_F S \kappa}{g_F \mu_B N_F}. \quad (3)$$

Generally<sup>23</sup> the ferromagnet's Curie temperature  $T_C$  is much larger than  $T_N$  (for Fe-FeF<sub>2</sub>,  $T_C = 1063$  K, and  $T_N = 79$  K). Therefore, at  $T < T_N \ll T_C$ , the magnetic properties (magnetization) of the FM are temperature independent. Within the FM-DW model, the main effect of temperature is to change the magnitude of the AF interface spins  $S$ , i.e., the effective coupling  $S\kappa$  becomes  $\langle S \rangle_T \kappa$ . This is equivalent to assume that the FM-AF coupling is temperature independent. Assuming that the AF interface magnetization has the bulk mean field temperature dependence,  $\langle S \rangle_T = S B_S(x)$ , where  $B_S(x)$  is the Brillouin function,  $x = -z J_{AF} S \langle S \rangle_T / k_B T$ , and  $z$  is the number of nearest AF neighbors in the AF bulk. This is equivalent to consider a very high AF anisotropy. Replacing  $S$  by  $\langle S \rangle_T$  in Eq. (3), the temperature dependence of  $H_{EB}$  becomes

$$H_{EB}(T) = -\frac{2J_F S B_S(x) \kappa a_F}{g_F \mu_B T}, \quad (4)$$

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TABLE I. Magnetic parameters for Fe–FeF<sub>2</sub>, Fe–MnF<sub>2</sub>, and Ni–NiO.  $J$  and  $K$  are in milli-electron-volt.

System	$J_F$	$K_F$	$J_{AF}$	$K_{AF}$	$g_F$	$g_{AF}$
Fe–FeF <sub>2</sub>	16 <sup>a</sup>	0.001 <sup>a</sup>	–1.26 <sup>c</sup>	2.500 <sup>e</sup>	2.2 <sup>a</sup>	3.75 <sup>f</sup>
Fe–MnF <sub>2</sub>	16 <sup>a</sup>	0.001 <sup>a</sup>	–1.08 <sup>d</sup>	0.120 <sup>e</sup>	2.2 <sup>a</sup>	2.50 <sup>f</sup>
Ni–NiO	14 <sup>b</sup>	0.043 <sup>b</sup>	–11.0 <sup>b</sup>	0.043 <sup>b</sup>	2.5 <sup>b</sup>	2.50 <sup>b</sup>

<sup>a</sup>See Ref. 24.

<sup>b</sup>See Ref. 21.

<sup>c</sup>See Ref. 10.

<sup>d</sup>See Ref. 27.

<sup>e</sup>See Ref. 25.

<sup>f</sup>See Ref. 26.

with  $a_F$  as the distance between adjacent FM planes and  $t_F = N_F a_F$  as the FM thickness. To validate this model, we calculate  $H_{EB}$  for Fe–FeF<sub>2</sub> at different temperatures using the parameters in Table I, and  $a_F = 0.2$  nm,  $t_F = 13$  nm, and the only adjustable parameter  $J_{F/AF} = -0.80$  meV. We always used  $S = 1$  and  $H_{cf} = 2000$  Oe. Our results in Fig. 1 are in good agreement with experiment.<sup>10</sup>

To calculate at  $T = 0$  the energy barrier per spin  $\Delta E$ , we apply a strong external magnetic field,  $\mathbf{H}$ , which forces the FM to align along this direction. Then,  $\theta_k = \Phi$  for every  $k$  and Eq. (1) reduces to

$$E(\Phi) = -2K_F \left[ \frac{J_F(N_F - 1)}{N_F K_F} + \cos^2 \Phi + \frac{hJ_F}{K_F} + \gamma \cos \Phi \right]. \quad (5)$$

$\gamma \equiv J_F S \kappa / K_F N_F$  compares the strength of the FM–AF exchange coupling energy ( $J_F S \kappa$ ) with the anisotropy energy of the FM ( $K_F N_F$ ). When  $\gamma < 1$ , Eq. (5) exhibits minima at  $\Phi_1 = 0$  and at  $\Phi_2 = \pi$  and a maximum at  $\Phi_3 = \arccos(-J_F S \kappa / 2K_F N_F)$  [solid line in the inset of Fig. 2(a)]. For  $\gamma > 1$  there is a minimum at  $\Phi_1 = 0$  and a maximum at  $\Phi_2 = \pi$  [dashed line in the inset of Fig. 2(a)].  $\Delta E$  is given by the difference between a maximum and a minimum of the energy. At  $\Phi_1$ :

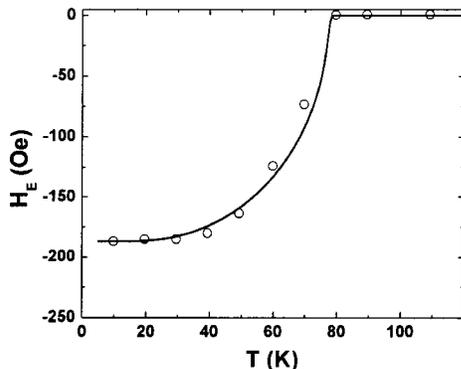
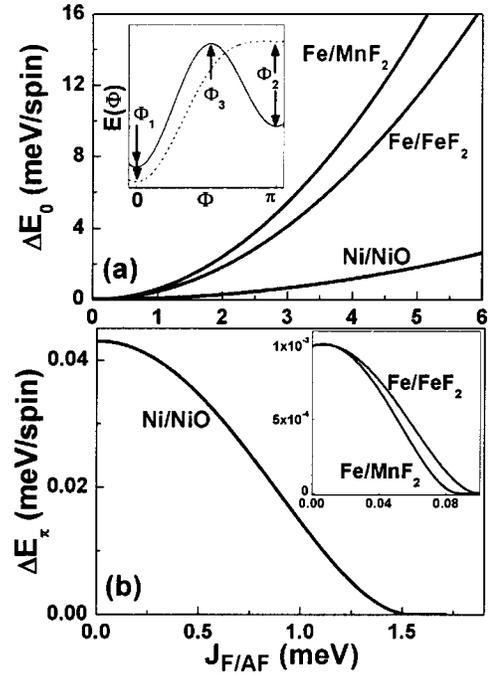
$$\Delta E_0 = K_F(1 + \gamma)^2 \quad \text{for } \gamma < 1, \quad (6)$$

$$\Delta E_0 = 4K_F \gamma \quad \text{for } \gamma > 1, \quad (7)$$

and at  $\Phi_2$ :

$$\Delta E_\pi = K_F(1 - \gamma)^2 \quad \text{for } \gamma < 1, \quad (8)$$

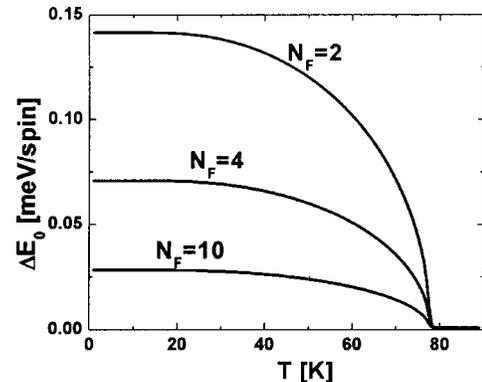
$$\Delta E_\pi = 0 \quad \text{for } \gamma > 1. \quad (9)$$


 FIG. 1.  $H_{EB}$  as a function of  $T$  for Fe–FeF<sub>2</sub>; solid line depicts our results, circles illustrate experiment (see Ref. 10).

 FIG. 2. Energy barrier,  $\Delta E_0$  and  $\Delta E_\pi$ , as a function of  $J_{F/AF}$ . Inset (a): Total energy  $E$  as a function of  $\Phi$  for  $\gamma < 1$  (solid line) and  $\gamma > 1$  (dashed line).

From these expressions and Eq. (2) when  $\gamma > 1$  or  $\gamma < 1$ ,  $\Delta E_0$  is proportional to  $J_{F/AF}^2$ <sup>21</sup> or  $J_{F/AF}^4$ , respectively. Figures 2(a) and 2(b) illustrate  $\Delta E_0$  and  $\Delta E_\pi$  for Fe–FeF<sub>2</sub>, Fe–MnF<sub>2</sub>, and Ni–NiO, with  $N_F = 1$  and using parameters in Table I. The Ni–NiO curve exhibits the same qualitative behavior as presented by Jensen<sup>21</sup> in Fig. 3(c).

Replacing  $S$  by  $\langle S \rangle_T$  in Eqs. (6) and (7) we obtain the temperature dependence of the energy barrier.  $\Delta E_0$  decreases with increases FM thickness (Fig. 3). As expected, the three curves collapse in one for  $\Delta E_0$  per unit interface area,  $A$ . For every thickness,  $\Delta E_0$  remains almost constant over a large interval of temperatures, until it drops to the FM value at  $T_N$ .

The relaxation time  $\tau$  for different sized FM and temperatures (Fig. 4) was evaluated for Fe–FeF<sub>2</sub> using  $\Delta E'(T) = N \Delta E_0(T)$ , with  $N$  the total number of FM spins. A volume built from a unit cell (formed by an Fe monolayer along the [110] direction) of  $3.96 \times 2.8 \times 1.98 \text{ \AA}^3$  repeats  $N_F$  times in the three dimensions. Below  $T_N$ , for a FM coupled


 FIG. 3. Energy barrier,  $\Delta E_0$ , per spin, as a function of  $T$  for different  $N_F$ , using the same parameters as in Fig. 1.

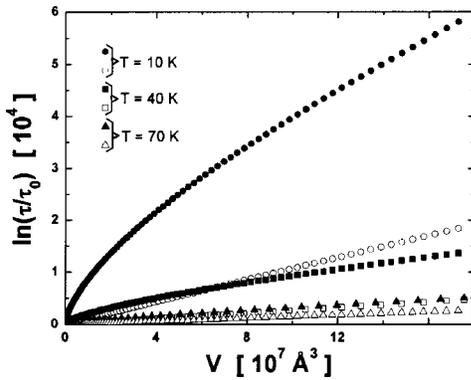


FIG. 4.  $\ln(\tau/\tau_0)$  as a function of  $V$ . Filled symbols represent Fe nanostructures coupled to  $\text{FeF}_2$ , while open symbols represent the same Fe cluster without coupling.

to an AF,  $\tau$  is greater than for an isolated FM case (Fig. 4), i.e., exchange coupling increases the transition temperature from ferromagnetism to superparamagnetism for small particle volumes. Figure 5 shows the temperature dependence of  $\ln(\tau/\tau_0)$  for two FM clusters of volumes  $1.12 \times 10^7 \text{ \AA}^3$  (lower curves) and  $3.8 \times 10^5 \text{ \AA}^3$  (upper curves) corresponding to the superparamagnetic limit ( $KV = 40k_B T$ ) for isolated FM clusters at  $T = 300 \text{ K}$  and  $T = 10 \text{ K}$ , respectively.

This model provides a clear description for experimentally measurable relaxation time, however it mainly refers to the dependence of  $\tau$  on the anisotropy. It does not include the algebraic dependence found in a quantum mechanical model<sup>28</sup> or the enhancements of the coercivity concomitant with exchange bias.<sup>7,8</sup> Preliminary calculations using the Mauri model<sup>12</sup> indicate that the conclusions obtained here are valid as long as changes in the effective switching volume while crossing  $T_N$  are forbidden. On the other hand, models which rely on the formation of domain walls<sup>8,17</sup> in

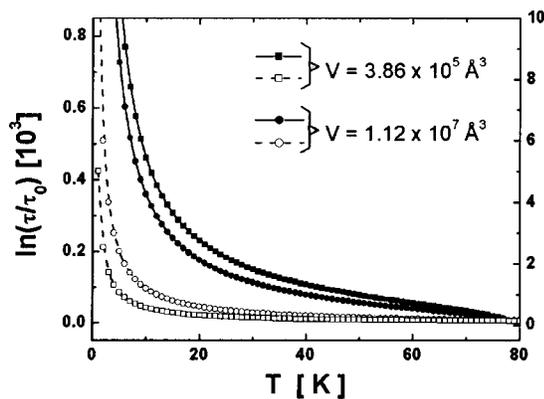


FIG. 5.  $\ln(\tau/\tau_0)$  as a function of  $T$  for two different volumes. Filled symbols represent an Fe cluster coupled to  $\text{FeF}_2$  while open symbols relates to the same Fe cluster without coupling.

the AF, for instance, may show an enhanced  $\tau$  for both spin directions.

In conclusion, we derived the temperature dependence of the exchange bias in FM–AF bilayers within the FM–DW model, assuming it to be given solely by the temperature dependence of the interfacial AF spins. These results are in agreement with experimental measurements in Fe– $\text{FeF}_2$ . From these we calculated the  $\Delta E$  as a function of temperature and  $\tau$  for various sized systems. A strong increase of  $\Delta E$  of spins initially oriented along the direction of the cooling field is found. As a consequence, an increase of the magnetic stability appears. However, spins initially pointing in the opposite direction exhibit a decreased energy barrier at zero field. But, as mentioned earlier, this would not be true if one considers for example domain walls in the antiferromagnet.

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