PHYSICAL REVIEW B **75**, 014434 (2007)

Exchange bias relaxation in reverse magnetic fields

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(Received 14 August 2006; revised manuscript received 24 September 2006; published 25 January 2007)

The time dependence associated with the so-called exchange bias in coupled polycrystalline ferromagnetic/antiferromagnetic films has been suspected as arising from antiferromagnetic domain dynamics. In this paper we present a quantitative description of the nucleation and growth of antiferromagnetic domains based on the Kolmogorov-Avrami model, which describes the time dependence of the exchange bias in all the systems for which data could be found.

DOI: 10.1103/PhysRevB.75.014434 PACS number(s): 75.70.Cn, 75.30.Gw, 75.50.Ss, 75.60.Ej

I. INTRODUCTION

The exchange bias effect, which arises from the exchange coupling between a ferromagnet (FM) and an antiferromagnet (AF) at the interface, was discovered about 50 years ago.[1] It is so named because the phenomenon manifests itself in a shifted hysteresis loop for the FM/AF bilayer film. The importance of the effect lies in its critical application in magnetoelectronic devices and there has been intensive study in recent decade.[2,3] Through numerous theoretical studies,[4–8] knowledge has been gained in understanding the origin and the magnitude of the unidirectional exchange anisotropy. All of these lead to the conclusion that both the FM/AF interface and the domain structure of the AF play crucial roles in the exchange bias. Neither can be directly observed in experiments without sophisticated apparatus[8,9] in the FM/AF bilayer heterostructures.

Recent years have also seen increased attention to the dynamical aspects of exchange bias, which is of practical importance as well as having scientific merit. Experiments including those in Refs. 10–14 have been conducted to investigate the relaxation of the FM magnetization of a bilayer in a reverse field and the training effect, which describes the decrease of exchange bias with cycling of the field. These studies suggest that changes are occurring in the state and structure of the polycrystalline AF layers in response to the dynamical environment. In fact, the underlying mechanism for these results is the same as that leading to the exchange bias reduction and reversal observed in the earlier experiments performed by Heijden *et al.*[15,16] In their study, exchange-biased polycrystalline FM/AF bilayer films were placed in a magnetic field, which was opposite to the exchange bias direction. The field was strong enough to reverse the FM magnetization. At subsequent times, the exchange bias field H_e was measured by the shift of the hysteresis loop providing a time dependence of the exchange bias. In Binek *et al.*'s study[14] to understand the training effect of exchange bias, a Landau-Khalatnikov theory was employed to characterize the time evolution of the interface magnetization in the AF layer when the bilayer system approaches equilibrium. In this paper, we provide a theory based on the Kolmogorov-Avrami (KA) model[17,18] that describes the dynamics of the AF layers and quantitatively explains the observed time dependence of exchange bias in a variety of exchange-biased systems. The KA model has been successfully applied to ferroelectric domain reversal.[19] We believe it can be used in magnetic systems. In fact, domain sizes of micrometers have been observed in epitaxial AF films coupled with FM films.[9] AF domains can be formed in polycrystalline AF/FM films through intergranular coupling.[20,21] In this paper, we establish a relationship between the exchange bias and AF domain growth and therefore provide an understanding of the relaxation behavior of the exchange bias.

II. MODEL

The subject in this study is a polycrystalline FM/AF bilayer that is described in Ref. 15. The FM layer is a typical magnetically soft material. Exchange bias is obtained by the coupling between the FM magnetization and the net moments of the AF grains at the interface.[6] The AF grains are assumed to be single domains. At equilibrium, the exchange field H_E is $J_\mathrm{int}/M_s t_\mathrm{FM}$, where M_s and t_FM are the saturation magnetization and thickness of the FM layer, respectively, and J_int is the average interface coupling energy per unit area between the FM magnetization and the AF grains. When a strong magnetic field of 5 kOe or greater is applied on the bilayer in the direction against the exchange bias, the FM magnetization reverses instantly to align with the external field, leaving the AF grains in a position that is not energetically favored. From the magnetizing measurements, we know that the FM magnetization is saturated in the field direction and that most the moments are aligned with the applied field.[15] The AF grains must overcome the uniaxial anisotropy energy barriers via thermal activation in order to switch over to reach the lower-energy state. For simplicity, we consider that the switch is accomplished by coherent rotation of the AF state of a grain, meaning there is no twist on the magnetic moments in the thickness direction. This means the AF film thickness is less than the AF domain wall width.

Two kinds of behavior could occur simultaneously for the AF grains during the relaxation process. First, in the presence of a reverse magnetic field, AF grains keep reversing due to thermal excitation until the whole AF layer is reversed. Second, AF grains next to each other are coupled via the exchange coupling between the moments at the grain boundary since both sublattices of the AF magnetic moments are exposed at the grain boundaries. Similar to Malozemoff's

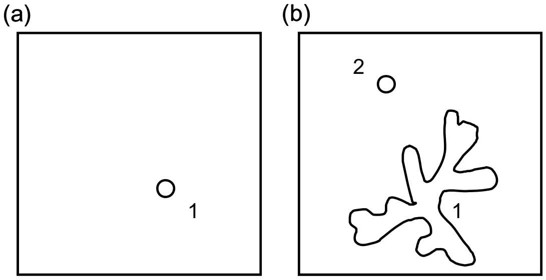

FIG. 1. (a) Schematic of the AF layer with a reversed grain appearing on site 1 at time τ_1. (b) At time $\tau_2 > \tau_1$, a domain grows from the grain while a second reversed grain appears on site 2.

argument on the random field theory,[4] the coupling strength is proportional to $\sqrt{N}$, where N is the number of moments at the boundary. Therefore, there exists an intergranular coupling in the AF layer and the "averaged-out" coupling should be weaker than the coupling in soft FM films.[20] In addition, the distance across the grain boundary reduces the coupling strength as well.[5] Therefore, the AF grains adjacent to the reversed grain are more likely to switch than others. In this case, a domain is formed and grows over time from the initial reversed grain, which is considered as a nucleus. Note that when an AF grain flips, its net moment at the interface also flips, which contributes to the exchange bias relaxation. In the following discussion, the reversed domains are characterized by the areas covering the FM/AF interface as shown in Fig. 1.

Suppose a reversed domain is formed at time τ in an AF layer of a unit surface area. Let us denote the size of the domain after time t as $s(t-\tau)$. We assume that the domain growth is independent of the time τ at which it is formed. The probability of nucleation in a time interval $\Delta\tau$ is denoted as $n(\tau)\Delta\tau$. Therefore, the domains nucleated in $\Delta\tau$ result in a reversed area of $n(\tau)s(t-\tau)\Delta\tau$ at time t. In other words, the AF grains that do not switch occupy an interface area of

$$1 - n(\tau)s(t-\tau)\Delta\tau.$$

The magnetic structure of the AF layer at time t is in fact the consequence of all the occurrences of nucleation and domain growth since the reverse field is applied. Dividing time t by $\tau_0=0$, τ_1, $\tau_2,\ldots,$ $\tau_i,\ldots,$ $\tau_N=t$, and writing $\Delta\tau_i=\tau_i-\tau_{i-1}$, the area of nonreversed AF grains is

$$A(t) = \prod_{i=1}^{N} [1 - n(\tau_i)s(t-\tau_i)\Delta\tau_i]. \tag{1}$$

Taking the logarithm of $A(t)$, we obtain that

$$\ln A(t) = \sum_{i=1}^{N} \ln[1 - n(\tau_i)s(t-\tau_i)\Delta\tau_i]. \tag{2}$$

Letting $\Delta\tau_i \to 0$ and $N\to\infty$ and noting that $\Sigma_i \Delta\tau_i = t$, Eq. (2) is transformed to

$$\ln A(t) = -\sum_{i=1}^{\infty} n(\tau_i)s(t-\tau_i)\Delta\tau_i = -\int_0^t n(\tau)s(t-\tau)d\tau. \tag{3}$$

It simply says

$$A(t) = \exp\left(-\int_0^t n(\tau)s(t-\tau)d\tau\right). \tag{4}$$

The net interface coupling strength to the FM magnetization at the given time t is

$$\begin{aligned} J_{\mathrm{int}}(t) &= J_{\mathrm{int}}A(t) - J_{\mathrm{int}}(1-A(t)) \\ &= J_{\mathrm{int}}\left[2\exp\left(-\int_0^t n(\tau)s(t-\tau)d\tau\right) - 1\right]. \end{aligned} \tag{5}$$

The exchange bias field then becomes

$$H_e(t) = H_{\mathrm{E}}\left[2\exp\left(-\int_0^t n(\tau)s(t-\tau)d\tau\right) - 1\right]. \tag{6}$$

The relaxation of exchange bias is therefore determined by the nucleation probability and the characteristics of the domain growth. Neither can be easily measured, especially for AF layers. Let us consider several limiting cases. The first is the case I when there is no exchange coupling between AF grains, $s(t-\tau)=s_0$, and the nucleation, i.e., the switching of AF grains, occurs with a constant rate $n(\tau)=n_0$. Thus, the exchange bias field decreases exponentially over time, $H_e(t)=H_{\mathrm{E}}[2\exp(-n_0 s_0 t)-1]$.

Since real magnetic films do not have a uniformity in size and orientation of the grains and anisotropies and in the intergranular coupling, the domains expand selectively, leading to a fractal structure as shown in Fig. 1(c). This kind of structure has been demonstrated in AF layers by Monte Carlo simulations.[21] To characterize the domain growth, we express it as a power law,

$$s(t-\tau) = c(t-\tau)^{\gamma}, \tag{7}$$

where c is the domain growth speed and $\gamma > 0$. Strictly speaking, the domain growth speed c depends upon the stage and nature of the domain development and varies with time. When the domain grows to a size much larger than the AF grains, it can be approximated to be a constant since the switching grains at the domain boundary just interact with their neighboring grains. In a homogeneous system, γ for domain growth is its dimensionality d. However, the defects and pinning sites formed in the AF films during deposition play an important role in determining the domain structure and slow down the domain growth. The defects can be the FM moments that are not reversed by the applied field. Furthermore, there may be minority AF grains with net surface moments opposite to the bias direction initially. When the applied field is reversed, these grains will inhibit domain growth. Therefore, γ will be smaller than d.

Under the domain growth scenario, let us consider two limiting cases: (II) nucleation at $\tau=0$ immediately after the reverse field is applied, i.e., $n(\tau)=\delta(\tau)$; and (III) a constant

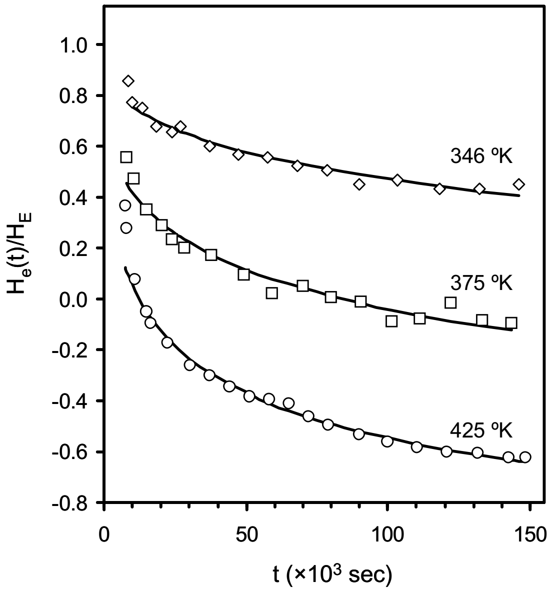

FIG. 2. Time dependence of normalized exchange field in the presence of an external magnetic field that is opposite to the initial exchange bias direction. Data are taken from Ref. 15 and fitted to Eq. (10).

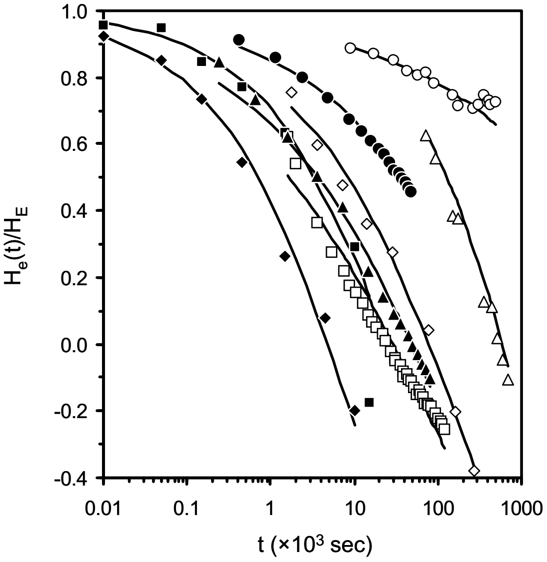

FIG. 3. Normalized exchange field as a function of time in the presence of reverse magnetic fields. Data are taken from Ref. 22 (□) for NiFe/IrMn at 400 K; Ref. 23 (◆) for NiFe/PtMn and (■) for NiFe/NiMn at 413 K; Ref. 24 (▲) for CoFe/NiMn at 473 K and (●) for CoFe/IrMn at 493 K; Ref. 25 (△) for NiFe/NiO at room temperature; Ref. 26 (◇) for CoFe/IrMn at room temperature; and Ref. 27 (○) for NiFe/NiO at room temperature. Data for each system are fitted to Eq. (10).

nucleation rate, i.e., $n(\tau)=n_0$. Inserting the nucleation rate $n(\tau)$ and Eq. (7) into Eq. (6), the exchange bias field for case II is obtained as

$$H_e(t) = H_{\mathrm{E}}\left[2\exp\left(-\int_0^t \delta(\tau)c(t-\tau)^{\gamma}d\tau\right)-1\right]$$
$$= H_{\mathrm{E}}[2\exp(-ct^{\gamma})-1] \qquad (8)$$

and for case III it is

$$H_e(t) = H_{\mathrm{E}}\left[2\exp\left(-\int_0^t n_0 c(t-\tau)^{\gamma}d\tau\right)-1\right]$$
$$= H_{\mathrm{E}}\left[2\exp\left(-\frac{cn_0}{\gamma+1}t^{\gamma+1}\right)-1\right]. \qquad (9)$$

For these two cases, the exchange bias relaxation can be generically written as

$$H_e(t) = H_{\mathrm{E}}\{2\exp[-(t/\tau_D)^{\sigma}]-1\}, \qquad (10)$$

where τ_D is defined as the characteristic relaxation time for exchange bias and the power index $\sigma=\gamma$ for case II and $\sigma=\gamma+1$ for case III. Note that $\sigma=1$ for case I where the AF grains switch independently with a constant rate, i.e., $n(\tau)=n_0$ and $s(t-\tau)=s_0$.

III. ANALYSIS

Figure 2 shows that Heijden *et al.*'s time dependence data[15] of NiO biased bilayers may be fitted to Eq. (10). Furthermore, this analytical expression seems to give a better fit than numerical calculations based on the thermal excitation model of independent AF grains in Ref. 15. The power index σ obtained from the fitting is 0.373, 0.329, 0.317, and 0.365 for 346, 375, 400, and 425 K, respectively, showing little dependence on temperature. These values are less than unity and rule out the case I ($\sigma=1$) for independent AF grain switching and case III ($\sigma>1$) for AF domain growth with a constant nucleation rate. In AF layers with dispersion in grain size, anisotropy, and interface coupling, smaller grains with lower energy barriers will switch faster than larger ones with higher energy barriers due to thermal excitation, as described by an Arrhenius law for the time to nucleate τ, $\tau=\nu_0^{-1}\exp(E_V/k_BT)$, where ν_0 is the attempt frequency, E_V is the barrier energy for an AF grain with an effective activation volume V, T is the ambient temperature, and k_B is the Boltzmann constant. In addition, nuclei may appear near the sites of film defects such as voids when a reverse field is applied. Therefore, the nucleation probability $n(\tau)$ decreases rapidly over time and resembles a delta function $\delta(\tau)$.

We also observe that the measured exchange field is higher than the fit value when the time t is less than 10^3 s. A possible reason for this is that in the hysteresis loop measurements (25 s each in Ref. 15) the FM magnetization is switched back and forth, which interrupts the relaxation of the exchange bias. Another possibility is that the domain growth is slower when it is just formed. In this phase, the AF grains at the domain boundary experience more nonreversed grains than reversed ones, making them more difficult to switch.

The exchange bias relaxation and reversal in the presence of reverse fields have been widely observed in many other FM/AF thin films.[12,22–27] Figure 3 shows that all the results that we can find may be fitted to Eq. (10), regardless of material, temperature, or time scale. The fitting may appear

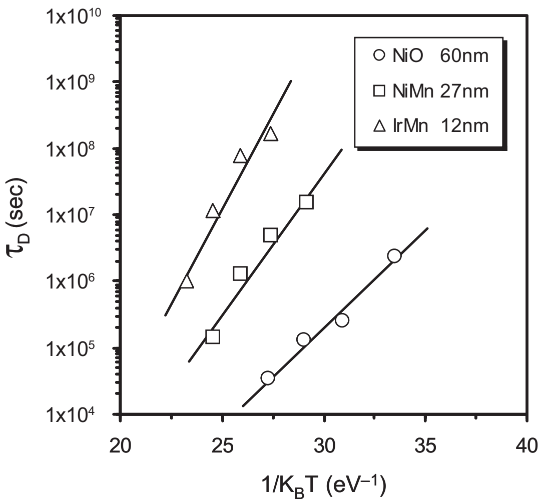

FIG. 4. Temperature dependence of the relaxation time constant τ_D for 60-nm-thick NiO in Ref. 15 and 27-nm-thick NiMn and 12-nm-thick IrMn in Ref. 24.

trivial since two parameters are used to describe a monotonic behavior. However, a number of functions have been tried on the sufficient data points and the best fit is obtained from Eq. (10). Furthermore, Eq. (10) makes the most sense because it describes the relaxation behavior under the KA mechanism illustrated above. The power index σ varies from 0.253 for NiFe/IrMn in Ref. 22 to 0.583 for CoFe/IrMn in Ref. 25 but is close to 1/3 in many cases. We are not aware of any plausible reason for this, but 1/3 is generally found in the relaxation of spin-glass systems.[28] Despite varying from sample to sample, σ changes little for measurements on the same samples.[15,23,24] This implies that σ depends strongly upon the sample preparation and the film microstructure.

Figure 4 shows the characteristic relaxation time length τ_D extracted from the fitting to Eq. (10) for the NiFeCo/NiO sample in Ref. 15 and the CoFe/NiMn and CoFe/IrMn samples in Ref. 24, on which temperature measurements were performed. The temperature dependence of τ_D is well described by an Arrhenius law,

$$\tau_D = \nu_\sigma^{-1} \exp(E_\sigma / k_B T). \tag{11}$$

It is well known that the attempt frequency ν_0 for magnetic grains related to spin-phonon interaction is generally believed to be 10^9 Hz. The ν_σ found from our data fitting, 1.7×10^3, 1.4×10^5, and 2.0×10^6 Hz for NiO, NiMn, and IrMn, respectively, is orders of magnitude lower. The significantly low attempt frequency ν_σ is evidence of the magnetic viscosity of the systems. Thermal excitation of AF grains has been effectively damped by the interaction between the AF grains and the FM magnetization. It is strongly dependent on the film microstructure. Moreover, the energy barrier E_σ is 0.66, 0.99, and 1.24 eV for the three samples. Taking the lateral grain sizes of 5 and 15 nm in Refs. 15 and 24, respectively, and assuming a typical uniaxial anisotropy constant of 10×10^5 erg/cm^3 for AF thin films at room temperature, the anisotropy energy is just 0.094 eV for NiO and 0.169 eV for NiMn and IrMn. Note that in order to obtain the exchange bias, the FM/AF interface coupling strength must be less than the anisotropy energy of the AF grains. The energy barrier for independent AF grains to switch in a reverse field must be less than the anisotropy energy of the grains.[24] The fact that the barrier energy E_σ in Eq. (11) is higher than the anisotropy energy by a factor of 6 indicates the existence of the coupling between the AF grains. The intergranular coupling results in a lower frequency for AF grain reversals and the formation of reversed domains. References 29 and 30 also show that the exchange bias relaxation time is longer for thicker AF bilayers. While this is intuitively understandable since the energy barrier should be a function of the volume of the AF grains, there is not sufficient data to establish a quantitative relationship between the relaxation time τ_D and the AF layer thickness.

IV. SUMMARY

In summary, we have described the nucleation and domain growth behavior of AF grains in polycrystalline exchange-biased bilayer systems based on the Kolmogorov-Avrami model. The change in the magnetic structure of the AF layer manifests itself macroscopically in the exchange field of the biased system. An analytical expression for the time dependence of the exchange bias has been derived. The theory not only explains the exchange bias relaxation and reversal in reverse magnetic fields that are found in a number of biased systems but also describes the training effect of exchange bias very well.[31] Further study will consider the random distribution and dispersion of the AF grains in size, anisotropy strength and orientation, and intergranular coupling. The study should also include the film microstructure and defects in the films that, as mentioned above in the text, play a critical role in the exchange bias relaxation.

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