Universal time relaxation behavior of the exchange bias in ferromagnetic/antiferromagnetic bilayers

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The resilience of the exchange bias ($H_{\text{Ex}}$) in ferromagnet/antiferromagnet bilayers is generally studied in terms of repeated hysteresis loop cycling or by protracted annealing under reversed field (training and long-term relaxation, respectively). In this paper we report measurements of training and relaxation in NiFe films coupled with polycrystalline FeMn and epitaxial $\alpha$-Fe$_2$O$_3$. We show that $H_{\text{Ex}}$ suppressed both by training and relaxation was partially recovered as soon as a field cycling for consecutive hysteresis loop measurement was stopped or the magnetization of the ferromagnet was switched back to the biased direction. In both cases we can model the observed logarithmic time relaxation behavior, and its film thickness and temperature dependence, in terms of a thermally activated reversal of the antiferromagnetic domain configuration to reduce the total magnetic energy. © 2006 American Institute of Physics. [DOI: 10.1063/1.2169876]

I. INTRODUCTION

The stability of exchange bias ($H_{\text{Ex}}$) in exchange-coupled ferromagnet (FM)/antiferromagnet (AF) systems is an important issue because it is related to the reliability of spin-valve devices such as magnetic read heads, nonvolatile memories, and various sensors.\(^{1,2}\) Up to now, most studies have focused on the thermal stability with the blocking temperature ($T_B$), i.e., the temperature at which $H_{\text{Ex}}$ vanishes.\(^3\)–\(^11\) Many exchange bias systems show a decrease of $H_{\text{Ex}}$ with the consecutive measurement of hysteresis loops. The so-called “training effect” has been used to describe such irreversible change of the $H_{\text{Ex}}$ with the number of measurements.\(^12\)–\(^19\) The training effect is generally distinctive in polycrystalline AF but very small or nonexistent in exchange bias systems based on single crystal (bulk or thin films).\(^213\) However, the origin of training effect is still unclear: Zhang et al. reported that the training effect in exchange bias systems based on polycrystalline AF materials can be interpreted by a model with positive and negative exchange couplings between AF grains.\(^16\) Recently, Hoffmann’s numerical simulation showed that the training effect may be caused by the existence of multiple anisotropy axes in the AF.\(^17\)

Several reports have suggested that the training effect is related to the reorientation of AF domains which takes place during magnetization reversal of the neighboring FM layer.\(^11\)–\(^14\) If this is true then $H_{\text{Ex}}$ should depend on the measurement time for the hysteresis loop as well as the number of measurements. Recently, van der Heijden et al. reported that the decrease of exchange bias with time, which could be explained by an Arrhenius model, was significant near the blocking temperature, so it is interpreted as a thermally assisted movement of AF domains.\(^20\) Hughes et al. reported that the recoil hysteresis loop was modified by the duration of time which spent with the FM layer saturated in the negative direction.\(^19\) In this report, we show that the training effect in many exchange bias systems coexists with a time-dependent relaxation of the $H_{\text{Ex}}$ because of the finite measurement time of hysteresis loop. We show that both training and long-term relaxation of the exchange bias show a logarithmic time dependence. We introduce a universal model which gives good fits to both the long-term relaxation and training data.

II. EXPERIMENT

We prepared two types of exchange-coupled FM/AF bilayer systems based on polycrystalline and single-crystalline AFs. Polycrystalline NiFe/FeMn bilayers on Si substrates were prepared by ultrahigh-vacuum dc sputtering with Ar pressure of 0.5 Pa and base pressure of $2 \times 10^{-6}$ Pa. During deposition, the samples were held in a magnetic field of about 250 Oe in order to induce a unidirectional anisotropy.\(^21\) FeMn/NiFe bilayers were also prepared on a long rectangular Si substrate within a magnetic shield, resulting in a uniaxial domain structure in the NiFe that was locked into the biased layer.\(^22\)

For comparison with the polycrystalline FeMn, an epitaxial AF $\alpha$-Fe$_2$O$_3$ layer was grown on $R$-plane (1102)$\alpha$-Al$_2$O$_3$ substrate by pulsed laser deposition (PLD), with a substrate temperature of 700 °C and oxygen pressure of 20 mTorr. The 50 nm $\alpha$-Fe$_2$O$_3$ films were transferred in air into...
an ultrahigh-vacuum dc sputtering chamber and 5 nm NiFe was deposited. For the NiFe/α-Fe₂O₃, a postannealing was performed in a magnetic field of 10 kOe for 15 min at 200 °C; previous measurements have shown a strong correlation between the exchange interaction and the α-Fe₂O₃ crystal orientation implying that the exposure to air has a limited effect on the bias.²³ X-ray-diffraction (XRD) measurements showed that the α-Fe₂O₃ film has excellent epitaxy.

The magnetic hysteresis loop \( M(H) \) of NiFe layer was measured by a vibrating-sample magnetometer (VSM). It took about 4 min to collect each \( M(H) \) loop data.

### III. RESULTS

Figure 1 shows the FeMn thickness dependence of the blocking temperature \( (T_B) \), coercivity \( (H_C) \), and exchange bias \( (H_{Ex}) \) for the FeMn/NiFe(3 nm) system. \( T_B \), the temperature at which \( H_{Ex} \) becomes zero, was estimated from the temperature dependence of the hysteresis loop. Our data were basically consistent with those in the previous reports.⁶–⁸ It is worth noting that \( T_B \) drastically decreases as the FeMn thickness is reduced below 5 nm.

We performed consecutive measurements of several hysteresis loops in order to investigate the training effect. Figure 2 shows the representative training hysteresis loops for the 4 nm FeMn sample at 295 and 77 K. Two samples deposited together were used for the measurements at the different temperatures. The magnetic field \( (H) \) was cycled in the form of \( +H \rightarrow -H \rightarrow +H \). The hysteresis loops \( M(H) \) were continuously collected up to the sixth loop, while the seventh one was obtained after the field was held at \( +H \) for 30 min. The measurement time for each \( M(H) \) loop was about 4 min. The \( H_{Ex} \) in the second hysteresis loop was distinctly decreased in comparison to that in the first loop, but after then the decrease of the \( H_{Ex} \) was relatively small. On the other hand, the \( H_{Ex} \) in the seventh loop was clearly increased. This means that \( H_{Ex} \) is partially but quickly recovered once the field cycling is stopped. The dependence of the training effect on the FeMn thickness is summarized in Fig. 3; the...
training effect decreases significantly at both temperatures for thicker FeMn layers and is much weaker at low temperature. Importantly for the discussion later, $H_{\text{Ex}}$ for all samples was partially recovered by a time relaxation for 30 min before the seventh hysteresis loop was recorded.

Figure 4(a) shows the long-term relaxation behavior of the $H_{\text{Ex}}$ for several FeMn thicknesses. The magnetic field was cycled in the form of $-H \rightarrow +H \rightarrow -H$ and the time-dependent hysteresis loops were collected after the cycling field was held at $-H$ for a time $t$. The $H_{\text{Ex}}$ gradually decreases with increasing $t$ and appears to depend linearly on $\log(t)$. The slope rapidly decreases as the FeMn thickness increases. Figure 4(b) demonstrates that a log(time) decay or recovery behavior could be induced by successive annealings at $-H$ or $+H$, respectively.

Exchange bias can only be induced or modified through changes in the AF induced by the interfacial exchange interaction with the FM. This is due to the fact that the AF does not respond to moderate fields as it has no net Zeeman energy. Thus the increase or decrease of the time-dependent $H_{\text{Ex}}(t)$ should be determined only by the relative directions of the FM magnetization and $H_{\text{Ex}}$. This was confirmed by the experiment in which the NiFe layer was deposited first on a long rectangular Si substrate in a magnetic shield. Under these conditions a uniaxial domain structure is formed in the NiFe which sets the domain structure of the subsequently deposited FeMn so as to lock the opposite exchange bias into the different FM domains. The resulting hysteresis loop (Fig. 5) is a combination of two loops with $H_{\text{Ex}} = +400$ and $-400$ Oe, respectively. In the third hysteresis loop in Fig. 5, which was measured after the magnetic field was held at $-600$ Oe for 2 h, the left partial loop (A) clearly shifts towards zero field while right side loop (B) was unchanged. On the other hand in the fourth hysteresis loop, measured after 2 h at $+600$ Oe, both sides A and B shifted to the negative field direction, indicating that $H_{\text{Ex}}$ in the A side recovered while that in the B side was suppressed.

It has been reported that a training effect is very small or nonexistent in the exchange bias system based on single-crystalline AF (bulk or thin films). In order to investigate a time relaxation behavior in such a single-crystal AF system, we prepared a NiFe/(1102) $\alpha$-Fe$_2$O$_3$ sample. As seen in Fig. 6, the time relaxation behavior was very small, but it is still observable.

IV. DISCUSSION

These results show that a long-term decay and recovery behavior of $H_{\text{Ex}}$ is observed in these samples in addition to a “conventional” training effect. Since both represent changes
in the magnitude of $H_{\text{Ex}}$, it is reasonable to ask whether they are in fact separate effects or different manifestations of the same mechanism.

In these experiments, the only external driving force on relaxation processes occurring within the AF which reduce or enhance the total bias is the interfacial exchange interaction; the magnitude of the magnetic field is not a critical factor as long as the applied magnetic field is larger than the saturation field of FM layer and less than the spin-flop field of the AF. The results from Figs. 4 and 5 demonstrate that $H_{\text{Ex}}$ is progressively reduced when the relative direction between the $H_{\text{Ex}}$ and the FM magnetization is antiparallel, while it recovers when the relative direction is parallel.

The strong log(time) kinetics evident from Fig. 4 immediately suggest a thermally activated reversal process involving a range of activation energies in the AF layer.\textsuperscript{3,20,24} To model our data we have adopted the activation energy spectrum model originally introduced for relaxation in amorphous metals.\textsuperscript{25} In its most general form, this model considers relaxation within a two-level system which, for the particular case of exchange bias, we take to be an individual AF grain or domain switching from a positive to a negative exchange energy with respect to the FM layer. The simplification used here is that following an isothermal annealing for a particular time $t$, it is assumed that all processes with energy below $E_0$ have relaxed while higher energy processes only contribute a later time. This leads to the general equation,

$$\Delta P = p_0(t) k_B T \ln(v_0 t), \quad (1)$$

relating the change in the observed quantity $\Delta P$ (exchange bias in our case), to the attempt frequency $v_0$ and the activation energy spectrum $p_0(E)$. If the latter is approximately constant over the range of the experiment then log(time) kinetics are recovered.\textsuperscript{26}

To apply this model to the data obtained from our experiments we define an activation energy spectrum $p_{\text{R}}(E)$ for the reversal energy of AF grains or domains. This spectrum is a characteristic of the particular AF and should depend on the details of the microstructure. If we retain the assumption that $p_{\text{R}}(E)$ is approximately independent of energy for the spectrum of relevance to the model, the distribution of surface relaxation energies $p_{\text{S}}(E)$ must be related to $p_{\text{R}}(E)$ by an expression of the form

$$P_s(E) = \frac{p_{\text{R}}(E)}{(d - d_0) K(T)}, \quad (2)$$

where $d$ is the AF layer thickness, $d_0$ is the AF thickness for which the blocking temperature is equal to the measurement temperature (the “blocking thickness”), and $K(T)$ is the temperature-dependent anisotropy energy per unit volume of the antiferromagnet. What this expression does in essence is to rescale the energy axis so that the density of available processes at a particular energy increases as the thickness and anisotropy decrease and diverges as it approaches the blocking thickness so as to give the instantaneous relaxation expected at $T_B$. Substituting (1) into (2) gives the change in $H_{\text{Ex}}$ with time,
V. CONCLUSIONS

We have investigated the training effect and the time relaxation behavior in exchange-coupled FeMn/NiFe and NiFe/\(\alpha\)-Fe\(_2\)O\(_3\) bilayers. Our systematic study revealed that in exchange bias systems the \(H_{ex}\) is dependent on measurement time as well as measurement number, and thus a change in the \(H_{ex}\) is caused by a time relaxation behavior of AF domain configuration to minimize the total magnetic energy.

Despite its simplicity, we have demonstrated that our universal model gives good fits to both the long-term relaxation and training data. The significance of this result is that, at least up to \(d=20\) nm, the whole thickness of the AF works to stabilize the relaxation during annealing. Related experiments have recently been performed by Pina et al.,\(^{25}\) however, they observed the relaxation behavior as a function of the degree of FM reversal which makes the analysis much more complicated.

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FIG. 8. Plot of \(\Delta H_{ex}/H_{ex}\) vs \((d-d_0)^{-1}\) between measurements 1 and 2 from Fig. 3(a). The solid line displays a best fit using \(d_0=3.3\) and \(d_0=1.8\) nm for 295 and 77 K, respectively.

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applied to reverse the FM. Starting from the as-grown or as-annealed state, a full hysteresis loop therefore contains time periods in which AF relaxation acts so as to reduce the total bias, followed by partial recovery once the alignment of the FM returns to that of the exchange bias. Microscopically, the local AF rearrangements depend on the local state of the FM, but it is reasonable to assume a square loop and take each hysteresis loop as consisting of discrete relaxation and recovery periods. In Fig. 8, we plot the reduction in normalized \(H_{ex}\) vs \((d-d_0)^{-1}\) between measurements 1 and 2 from Fig. 3(a). Since all loops reported in this paper take the same time to collect, this is effectively a measurement at constant \(t\) provided that the recovery between measurements 1 and 2 is assumed to be small and so we again expect a linear dependence which is caused by the relaxation for half the period of the loop (~2 min). Using the same value of \(d_0=3.3\) nm used in the analysis reported above we again get a good linear fit to the data; the gradient of the least-squares fit to the data is 0.15 nm in Fig. 8. If we take the best fit to the relaxation rate in Fig. 7 to determine the reduction in normalized \(H_{ex}\) expected for 2 min we obtain a value of ~0.13 nm. Thus there is a good internal consistency for the same model applied to both long-term relaxation and training. Since \(d_0\) is temperature dependent by definition we cannot quantitatively investigate the temperature variation of the relaxation, but if \(d_0\) is assumed to be 1.8 nm at 77 K then reasonable agreement is obtained between the data in Figs. 3(a) and 3(b) as shown in Fig. 8.

Our results demonstrate that an experimentally observed \(H_{ex}\) is dependent on the time required to measure a hysteresis loop.\(^{3,27}\) Recently, Hughes et al. performed a similar study of training with the measurement time.\(^{19}\) A measurement time of a hysteresis loop is generally about a few minutes in a commercial VSM, and it takes a much longer time in a superconducting quantum interference device (SQUID) magnetometer. As seen in Figs. 2 and 3, such a finite measurement time should be large enough to display a time relaxation behavior of exchange bias. Therefore, in practice, a decrease of the \(H_{ex}\) by the training effect is a manifestation of the change of the \(H_{ex}\) by a time relaxation.