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

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The resilience of the exchange bias (H_{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 α-Fe_2O_3. We show that H_{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_{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_{Ex} vanishes.[3–11] Many exchange bias systems show a decrease of H_{Ex} with the consecutive measurement of hysteresis loops. The so-called "training effect" has been used to describe such irreversible change of the H_{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).[2,13] 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_{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 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_{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 2×10^{-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 α-Fe_2O_3 layer was grown on *R*-plane $(1\bar{1}02)\alpha$-Al_2O_3 substrate by pulsed laser deposition (PLD), with a substrate temperature of 700 °C and oxygen pressure of 20 mTorr. The 50 nm α-Fe_2O_3 films were transferred in air into

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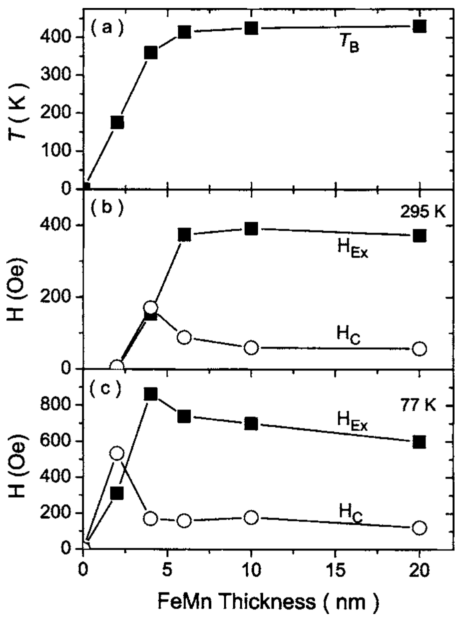

FIG. 1. Thickness of FeMn vs (a) blocking temperature (T_B), (b) coercivity (H_C) and exchange bias (H_{ex}) at 295 K, and (c) coercivity (H_C) and exchange bias (H_{ex}) at 77 K.

an ultrahigh-vacuum dc sputtering chamber and 5 nm NiFe was deposited. For the NiFe/α-Fe_2O_3, 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-Fe_2O_3 crystal orientation implying that the exposure to air has a limited effect on the bias.[23] x-ray-diffraction (XRD) measurements showed that the $(1\bar{1}02)\alpha$-Fe_2O_3 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

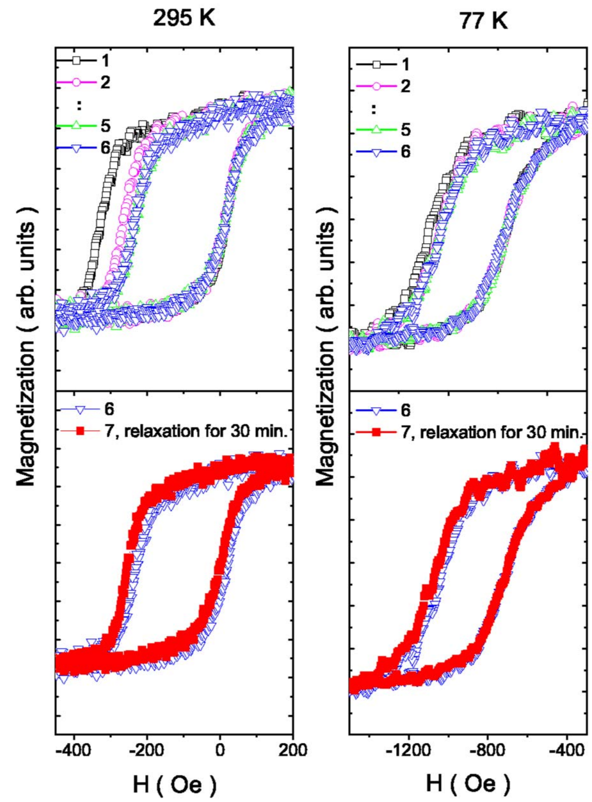

FIG. 2. (Color online) The representative hysteresis loops of the training effect for the 4 nm FeMn sample (a) at 295 K and (b) at 77 K. The inset label refers to the number of cycle.

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

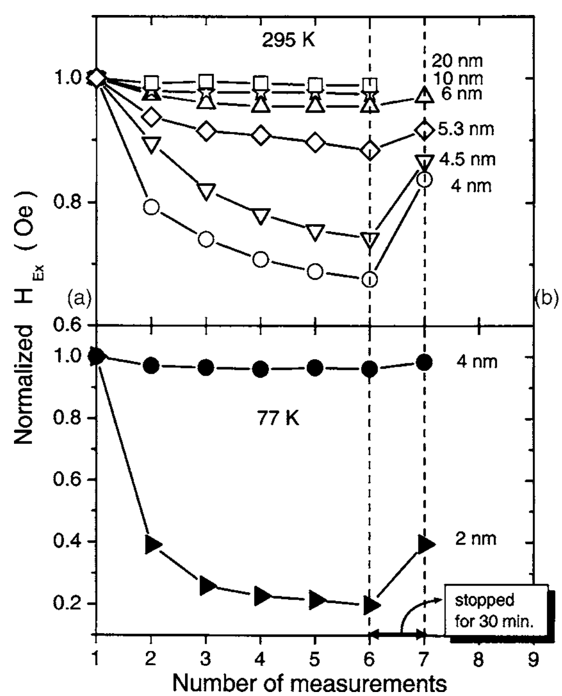

FIG. 3. Normalized H_{ex} as a function of the number of loop measurement for 4 nm FeMn (a) at 295 K and (b) at 77 K.

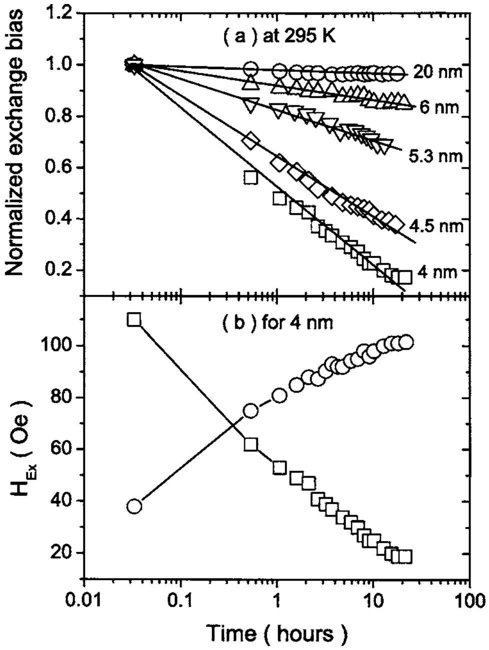

FIG. 4. (a) Normalized H_{ex} as a function of the time for several FeMn thicknesses at 295 K. The solid lines display the linear fits of a log(t) function. (b) The suppression and recovery behaviors as a function of the time for 4 nm FeMn at 295 K.

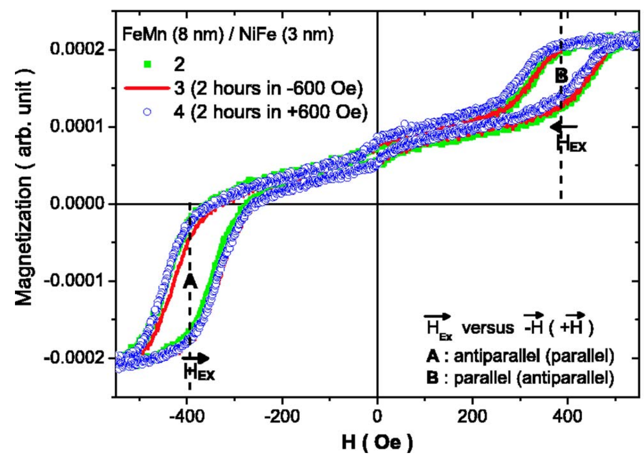

FIG. 5. (Color online) Sequential magnetic hysteresis loops for a FeMn(8 nm)/NiFe(3 nm) film deposited under a magnetic shield. The inset label refers to the number of cycle. The second loop was measured after the cycling magnetic field stayed at −600 Oe for 2 h, while the third loop was measured after the cycling magnetic field stayed at +600 Oe for 2 h.

training effect decreases significantly at both temperatures for thicker FeMn layers and is much weaker at low temperature. Importantly for the discussion later, H_{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_{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_{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 $H_{Ex}(t)$ should be determined only by the relative directions of the FM magnetization and H_{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.[22] The resulting hysteresis loop (Fig. 5) is a combination of two loops with $H_{Ex}=+400$ 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_{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).[2,13] In order to investigate a time relaxation behavior in such a single-crystal AF system, we prepared a NiFe/(1$\bar{1}$02) α-Fe_2O_3/(1$\bar{1}$02) α-Al_2O_3.[23] 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_{Ex} is observed in these samples in addition to a "conventional" training effect. Since both represent changes

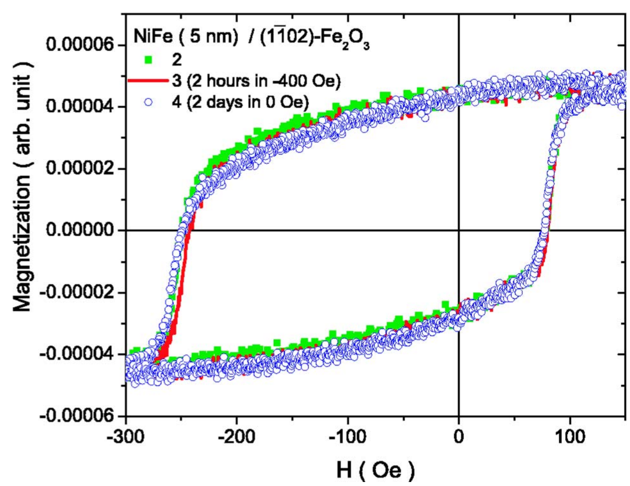

FIG. 6. (Color online) Sequential magnetic hysteresis loops for NiFe/50 nm(1$\bar{1}$02) α-Fe_2O_3 sample. The inset label refers to the number of cycle. The second loop was measured after the cycling magnetic field stayed at −400 Oe for 2 h, while the third loop was measured after 2 days in zero field.

in the magnitude of H_{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_{Ex} is progressively reduced when the relative direction between the H_{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.[3,20,24] To model our data we have adopted the activation energy spectrum model originally introduced for relaxation in amorphous metals.[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,

$$\Delta P = p_0(E) k_B T \ln(\nu_0 t), \tag{1}$$

relating the change in the observed quantity ΔP (exchange bias in our case), to the attempt frequency ν_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.[26]

To apply this model to the data obtained from our experiments we define an activation energy spectrum $p_R(E)$ 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 $p_R(E)$ is approximately independent of energy for the spectrum of relevance to the model, the distribution of surface relaxation energies $p_S(E)$ must be related to $p_R(E)$ by an expression of the form

$$P_S(E) = \frac{p_R(E)}{(d-d_0)K(T)}, \tag{2}$$

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_{Ex} with time,

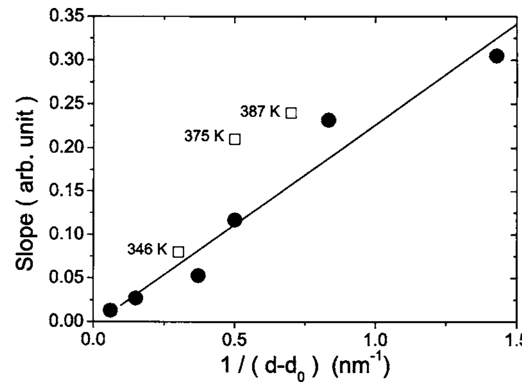

FIG. 7. Plot of the gradient of the logarithmic decay in Fig. 4(a) vs $(d-d_0)^{-1}$ (solid circles); the dashed line shows the best fit using d_0=3.3 nm. The open squares show the data from van der Heijden *et al.*[20] plotted with $(d-d_0)^{-1}$ scaled by $(T_N-295)/(T_N-T)$ where T_N=425 K and T corresponds to the different measurement temperatures marked on the plot.

$$\Delta H_{Ex} = \frac{p_R(E)}{K_0(d-d_0)(T_N-T)} k_B T \ln(\nu_0 t), \tag{3}$$

where we have used a linear approximation for the anisotropy $K(T) \sim K_0(T_N-T)$ which should be valid close to the Néel temperature T_N. Therefore a plot of the gradient of the isothermal logarithmic decay in Fig. 4(a) versus $(d-d_0)^{-1}$ should be linear provided that the correct value of d_0 is chosen. This is plotted in Fig. 7 using $d_0-3.3$ nm derived from the data of Fig. 1(a). This value gives optimum linearity for the data used and suggests that this type of model is appropriate for understanding the long-term annealing effects. It is important to note that the initial gradient for the recovery annealings in Fig. 4(b) is approximately identical to the longer-time gradient for the relaxation in the same plot: this shows that relaxation and recovery are identical two-level processes driven by the relative orientation of the magnetization and H_{Ex}. The gradient of the recovery at longer time scales decreases as the available reversal processes are exhausted close to full recovery.

In Fig. 7 we also plot the data extracted for a 10-nm-thick FeMn film from Fig. 3 of van der Heijden *et al.*[20] for which a log(t) dependence gives a good fit for $t > 10$ min. The blocking temperature for their film agrees with our value for 10 nm FeMn and so we plot the data, scaled for the appropriate blocking thickness and temperature-dependent anisotropy (assuming that T_B for an infinite-thickness film is equal to T_N). Given that the model makes a very simple assumption about the temperature dependence of the anisotropy and we are comparing materials grown in different vacuum systems under different conditions, the level of agreement leads us to conclude that our activation energy spectrum model gives a good description of the relaxation and recovery process. At some limit we expect the spectrum of available processes to become exhausted, but the experiments discussed here do not reach this time limit.

We now consider the training effects summarized in Fig. 3. For the samples in our experiments (and for practical exchange bias systems in general), the hysteresis loop is shifted so that the FM is more or less saturated at zero field. Relaxation of the AF is therefore minimal unless sufficient field is

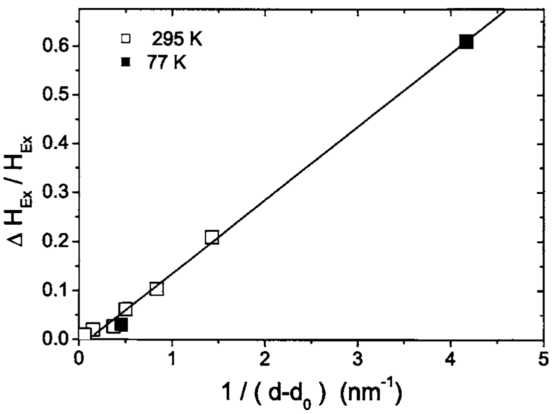

FIG. 8. Plot of the $\Delta H_{\mathrm{Ex}}/H_{\mathrm{Ex}}$ vs $(d-d_0)^{-1}$ between measurements 1 and 2 from Fig. 3(a). The solid line displays a best fit using d_0=3.3 and d_0 =1.8 nm for 295 and 77 K, respectively.

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 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.

V. CONCLUSIONS

We have investigated the training effect and the time relaxation behavior in exchange-coupled FeMn/NiFe and NiFe/α-Fe_2O_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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