Dynamics of Spontaneous Magnetization Reversal in Exchange Biased Heterostructures

Zhi-Pan Li, Casey W. Miller, Igor V. Roshchin, and Ivan K. Schuller

Physics Department, University of California, San Diego, La Jolla, CA, 92093-0319, USA

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The dependence of thermally induced spontaneous magnetization reversal on time-dependent cooling protocols was studied. Slower cooling and longer waiting close to the Néel temperature of the antiferromagnet ($T_N$) enhances the magnetization reversal. Cycling the temperature around $T_N$ leads to a thermal training effect under which the reversal magnitude increases with each cycle. These results suggest that spontaneous magnetization reversal is energetically favored, contrary to our present understanding of positive exchange bias.

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Exchange bias (EB) arises when a ferromagnet/antiferromagnet (FM/AF) heterostructure is cooled in a magnetic field ($H_{FC}$) below the Néel temperature $T_N$ of the AF. EB is manifested as a shift of the hysteresis loop along the field axis by an amount $H_{EB}$, dubbed the exchange bias field. This phenomenon has been intensely studied in the past ten years due to its significance in providing a magnetic reference in spin valve devices. More fundamentally, EB is also of physical importance for understanding competing interactions in coupled magnetic materials. A rich variety of physical phenomena are associated with EB, including thermal stability, positive EB, training effect, and exchange bias. A lower energy state would have positive EB, rather than observed positive EB. This contradiction implies that either spontaneous reversal is a novel metastable state, or our present understanding of positive EB is incomplete.

This work reports slow dynamics and thermal training of the spontaneous reversal effect. We show that a slow cooling rate enhances the magnetization reversal magnitude, and that reversal is strongly related to dynamic processes around $T_N$. Relaxation of the system at $T_N$ over a long period of time causes increased reversal at low temperatures. Successive thermal cycling about $T_N$ allows the system reach a global equilibrium state. These results show that spontaneous reversal is energetically favored rather than a metastable state as predicted by the existing positive exchange bias model. Possible directions for a modified theory of positive exchange bias are suggested.

The same Ni(3nm)/FeF$_2$(30nm) sample on a MgF$_2$ substrate was studied as in our previous work. FeF$_2$ is an AF with $T_N$ = 78 K, and grows epitaxially untwinned in the (110) direction on MgF$_2$ (110) substrates. The FM exhibits uniaxial anisotropy with the easy axis parallel to FeF$_2$ [001] (the spin axis of the AF). The magnetic
field was always applied along the easy axis of the FM. Prior to cooling, the FM was saturated with a 5 kOe field, well above the 150 K coercive field of $H_C = 0.35$ kOe, then reduced to $H_{FC}$. Fig. 1(a) shows the thermally induced magnetization reversal in $H_{FC} = 0.1$ kOe. Hysteresis measurements at $T = 10$ K find negative EB for $-0.25$ kOe $\leq H_{FC} \leq -0.1$ kOe, positive EB at $H_{FC} > 0.5$ kOe, and coexistence for $-0.1$ kOe $< H_{FC} < 0.5$ kOe (Figure 1(b)). $H_{EB} = (3.9 \pm 0.1)$ kOe for all cooling fields. Coexistence of positive and negative EB at an intermediate $H_{FC}$ has been interpreted as the AF breaking into “domains” with uncompensated moments of either sign. When the lateral size of these “domains” is much larger than the FM domain wall width, they independently induce either positive or negative EB in the FM, causing the experimentally observed double hysteresis loop. Since only positive EB is essential for spontaneous reversal, partial reversal was observed for $H_{FC}$ associated coexistence (Fig. 1(a)).

Two different cooling protocols were used to investigate the time dependence of the reversal magnitude $\Delta M = M(T = 10$ K$) - M(T = 150$ K$)$. We only consider $H_{FC} = 0.1$ kOe, for which the magnetization reverses by about 50% upon cooling. The first protocol cooled the sample from $T = 150$ K to 10 K with two decades (0.1-10 K/min) of uniform cooling speeds (Fig. 2(a) inset). The second protocol cooled the sample at 10 K/min from 150 K to an intermediate temperature $T_w$, where the temperature was held constant for a time $\tau$, then cooled to 10 K at 10 K/min (Fig. 2(b) inset). For both protocols, $M(T = 10$ K$)$ was measured after the sample temperature stabilized at 10 K.

Fig. 2(a) shows that slower cooling leads to a larger $|\Delta M|$. With the present definition, $\Delta M = -2$ implies complete magnetization reversal. With the largest cooling speed of 10 K/min, the FM reverses by $\Delta M = -0.9M_S$. When cooled at 0.1 K/min, $|\Delta M|$ increases by 0.2$M_S$. Moreover, the dependence of $\Delta M$ on the cooling speed is well fit by an exponential function $\Delta M = \Delta M_0 + A \exp(\alpha dT/dt)$. This fit implies $\Delta M$ of -0.88 and $-1.15M_S$ in the limits of infinite and zero cooling speed, respectively.

The second cooling protocol demonstrates that $|\Delta M|$ is sensitive to the time spent with $T \sim T_N$. The dependence of $\Delta M$ on the wait temperature $T_w$ shows the largest reversal for $T_w = 80$ K, closest to $T_N$ (Fig. 2). As $\tau$ increases beyond 35 min, $|\Delta M|$ increases from 0.86$M_S$ to 1.18$M_S$. For $T_w = 85$ K, $|\Delta M|$ only changes by 0.07$M_S$ after waiting for 50 minutes. For $T_w = 75$ K, $|\Delta M|$ saturates after $\sim 15$ minutes at 1.05$M_S$. This $T_w$-dependent behavior was not observed when waiting at the reversal temperature ($T = 63$ K). The results from these two cooling protocols show that spontaneous magnetization reversal exhibits slow dynamics with a relatively long time scale. The fact that the dynamics are most pronounced around $T_N$ hints that this effect depends on the establishment of AF domain states.

Several tests were performed to ensure that the dynamics were not experimental artifacts. First, measuring the magnetic moment via SQUID involves moving the sample through the SQUID coils by 4 cm, thus subjecting the sample to magnetic field inhomogeneity. To exclude this as an artifact, the cooling procedure used to obtain the data of Fig. 1 was repeated, but measuring only the initial and final the magnetization values, rather than several intermediate temperatures. The sample was thus only exposed to field inhomogeneity at these extreme temperatures. The reversal magnitude only differs by $3 \times 10^{-4}M_S$ between these two methods, which is negligibly small. Temperature fluctuations during cooling are another source of potential signal artifacts. To investigate this, the sample was heated from 10 K to a temperature $T_x$, then cooled back down to 10 K. When $T_w \leq 80$ K, $\Delta M$ varies by no more than 0.01$M_S$, too small to account for any $\Delta M$ variation found earlier (Fig. 3). When $T_x > 80$ K, a significant additional magnetization reversal was observed (more below). These checks demonstrate that the time-sensitivity of spontaneous re-

\begin{figure}[h]
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\includegraphics[width=\textwidth]{fig2.png}
\caption{(Color online) (a) $\Delta M/M_S$ as function of uniform cooling speed $dT/dt$ for $H_{FC} = 0.1$ kOe. The line is a fit to an exponential function. (Inset) Temperature vs time for uniform cooling speeds of 2.5 (blue) and 0.6 K/min (red). (b) $\Delta M/M_S$ as a function of wait time $\tau$ at temperatures $T_w = 75\text{ (green)}, 80\text{ (black)},$ and 85 (red) K for $H_{FC} = 0.1$ kOe. (Inset) Schematic of this cooling protocol. Lines are a guide to the eye.}
\end{figure}
versal is not an experimental artifact but rather is intrinsic to the system, and clearly related to the AF phase transition. Larger reversal for slower cooling rates and longer wait times around $T_N$ suggests that spontaneous reversal is thermodynamically favorable.

The slow evolution of the system toward a larger reversal implies the presence of large energy barriers. The additional large reversal during thermal cycling above 80 K (Fig. 3) suggests that the system can overcome this energy barrier by thermal activation. *Thermal training* refers to successive magnetization reversal when the system is cycled above and below $T_N$. With the FM saturated, the sample was first cooled in $H_{FC} = 0.1 \text{kOe}$ from 150 K to 10 K at 0.1 K/min, followed by heating to 150 K, just below the temperature for the FM to reverse back along the field direction (Fig. 1). After that, the sample was cycled between 150 K and 10 K. The magnetic field was held constant at $H_{FC} = 0.1 \text{kOe}$ throughout the thermal cycles. The FM reverses with each additional cooling (with decreasing incremental reversal magnitude) until the total magnetization reversal reaches $1.8 M_S$, significantly larger that the initial reversal for any cooling speed or wait time (Fig. 1(a)).

Fig. 1(b) shows the dependence of $M(10 \text{K})$ on the number of cycles $N$ for different $H_{FC}$ and cooling speeds. For all cases, they follow an exponential dependence, $M_N(10 \text{K}) = M_{\infty} + (M_S - M_{\infty}) \exp(-N/\eta)$, where $M_{\infty}$ is the convergent $M(10 \text{K})$ when $N \to \infty$, and $\eta$ is a characteristic cycle number for each $H_{FC}$ and cooling speed. $M_N(10 \text{K})$ for $N = 0$ is defined as $M_S$. $M_{\infty}$ appears to be linearly dependent on $H_{FC}$ for constant cooling speed (Fig. 1 inset). Larger $H_{FC}$ results in smaller $\eta$, which means a faster approach to $M_{\infty}$. This makes qualitative sense because a larger magnetic field should facilitate reversal by lowering the energy barrier, so that more AF moments are aligned in the field direction.

These experiments suggest that it is energetically favorable for the FM to reverse against $H_{FC}$, albeit counterintuitive since $|m_{FM}| \gg |m_{AF}|$. This behavior cannot be explained simply by the competition between the Zeeman energy and interfacial coupling. A new mechanism for determining the sign of AF uncompensated moments is necessary to explain the features we observe experimentally. Consider that $(M_S - M(10 \text{K}))/2M_S$ gives the percentage of sample that exhibits positive EB at 10 K for an intermediate $H_{FC}$. For $H_{FC} = 0.1 \text{kOe}$, the sample is nearly 90% positively exchange biased at 10 K after 6 thermal cycles at 0.1 K/min. The interfacial coupling energy in this sample is $E_{int} = J_{FM/AF}S_{FM}\cdot S_{AF} = \mu_0 H_{EB} M_{FM} M_{AF} = 0.79 \text{erg/cm}^2$, close to that previously found in similar systems. However, the onset $H_{FC}$ for positive EB in this case is about two orders of magnitude smaller than previously found. This very small $H_{FC}$ necessary for positive EB challenges the present interpretation of positive EB.

One possible explanation for these observations considers pinned uncompensated moments in the bulk of the AF. Neutron scattering results show that parallel AF domain walls can form between the bulk and interfacial AF moments when the interfacial moments are more strongly coupled with the FM. For small (large) cooling fields, interfacial AF moments need to orient in the negative (positive) direction to establish negative (positive) EB. Here, antiferromagnetic interfacial coupling is assumed. However, bulk AF moments far away from the interface are dominated by the applied field and align parallel to it. This is independent from the orientation of the interfacial AF moments. Therefore, a parallel AF domain wall forms in case of negative EB, but not for positive EB.

A second scenario considers a parallel domain wall in
the FM. Spontaneous rotation found previously supports this possibility. When cooling down, such a domain wall occurs in positively exchange biased thick FMs because the antiferromagnetic interfacial coupling locks interfacial FM moments in the negative direction while FM moments far from the interface only sense the external field. With the inclusion of the parallel AF and/or FM domain wall energies, the sign of exchange bias is no longer determined simply by the competition between $|E_{\text{int}}|$ and $|E_{AF-\text{Zeeman}}|$, and the paradox $|m_{FM}| < |m_{AF}|$ can be avoided. However, detailed calculations of the different energies involved in negative and positive EB are necessary to explicitly develop the pertinent relationships.

The observed slow dynamics may arise from the competition of these energies around $T_N$, which also determine the sign of EB while the AF order is established. The competition of these energies may result in multiple local energy minima that are separated by significant anisotropy barriers. These barriers grow larger compared with $k_B T$ with decreasing temperature, and the time it takes for the system to evolve into a lower energy state exponentially increases. Thermal training allows the system to seek out the global energy minimum because the FM domains are approximately unchanged with temperature, but the AF order is cyclicly perturbed. After cooling to $T_x < T_N$ for the first time, the AF orders, and a portion of the originally saturated FM spontaneously reversing because $|E_{\text{int}}| > |E_{FM-\text{Zeeman}}|$. Next, the AF order is cyclicly perturbed. After cooling to $T_x > T_N$ with the FM domains relatively unchanged. Subsequent cooling causes the population of positive EB regions to increase because the FM moments associated with the domain walls deviate from the field direction. This results in a smaller coupling energy with the AF, making it easier for the AF moments to align with the field. This gives rise to a larger fraction of the sample that shows positive EB, and thereby increases the magnetization reversal magnitude. This process is successful because the FM domain wall width is an order of magnitude larger than the AF domain wall width. The details of the time-dependent reversal, what determines the system ground state, and the various paths to reach this state are not presently understood.

In summary, two different cooling protocols revealed that spontaneous magnetization reversal in exchange biased heterostructures is strongly time-dependent. Slower cooling speeds and longer waiting times around $T_N$ lead to larger magnetization reversal. Thermal training was discovered by cycling the sample temperature about $T_N$, causing the FM to reverse successively with each cycle. This effect reflects the incremental conversion of negative to positive EB regions. These results suggest that spontaneous reversal is thermodynamically stable rather than metastable, contradicting our present understanding of positive EB. Additional energy terms that describe parallel domain walls in the antiferromagnet and/or ferromagnet are necessary to explain these results, and to refine positive exchange bias models.

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* E-mail: zl65@cornell.edu. Present address: Center of Nanoscale Systems, Cornell University, Ithaca, NY 14853.
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