Dynamical properties of magnetization reversal in exchange-coupled NiO/Co bilayers

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Real time dynamic magnetization reversal measurements at room temperature have been performed on polycrystalline exchange-coupled NiO/Co bilayers over 10 decades of applied field sweep rates. Domain wall displacement and domain nucleation regimes govern the magnetization reversal at low and high sweep rates, respectively. The crossover between the two regimes depends on the relative thickness of the two layers. Thicker Co favors propagation, whereas thicker NiO favors nucleation. The coupling energy at the interface was found to be inversely proportional to the square root of the area corresponding to the activation (Barkhausen) volume. These results are consistent with a model of exchange anisotropy in which a *randomness* of the coupling along the ferromagnetic (F)/antiferromagnetic (AF) interface is combined with a thermally activated switching process of the AF magnetization.

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The effects of the interfacial exchange interaction between a ferromagnet (F) and an antiferromagnet (AF) were discovered more than 40 years ago.[1] The most notable among these effects are an enhanced coercivity (H_C) and a shift in the hysteresis loop of the ferromagnetic layer, called exchange bias (H_E). Apart from their fundamental interest, exchange-biased AF/F systems are largely studied because of their applications in the data storage industry. Antiferromagnetic layers are used to induce either longitudinal or transversal pinning of adjacent ferromagnetic layers in magnetoresistive and giant magnetoresistive spin-valve heads.[2] However, the physics of exchange coupling is not fully understood yet and its microscopic origin is still controversial.[3]

In the first model to explain the exchange bias, H_E was assumed to arise from the exchange coupling at an uncompensated interface between the AF and F layers.[1] The AF spin configuration was assumed to remain frozen during the F reversal.[1] This model gives a good intuitive idea of the origin of the exchange bias, but the resulting coupling is several orders of magnitude too strong. Introducing features such as roughness and structural defects, which prevent the interface from being perfectly compensated, Takano *et al.* obtained the correct order of magnitude for H_E.[4] The enhancement of the coercivity was, however, not addressed. In other models, the reversal of the F layer is assumed to provoke a rearrangement of the AF moments at the interface.[5–9] Also, these models predict the correct order of magnitude for H_E. The switching of the F moments induces a torque on the magnetic moments of the AF at the interface, which leads to the formation of partial domain walls parallel to the interface. In addition, Malozemoff[6] assumed that the AF moment unbalance originating from features such as roughness and structural defects causes a ''random field'' leading to the formation of AF domain walls perpendicular to the AF/F interface. Recently, it has been shown that the ''random field model'' can also account for the observed enhancement of the coercivity as a consequence of the formation of lateral domains in the F layer.[10,11]

In a uniaxial ferromagnetic single film, the magnetization reversal for a field applied along the easy axis occurs by nucleation and growth of reversed domains. In systems with a stable multidomain magnetic structure at zero field it has been shown that domain nucleation dominates at high field sweep rates while domain wall propagation dominates at lower ones.[12–14] In the case of pinned ferromagnetic layers the interface exchange interactions can alter the geometry of the domain structure. The domain size during magnetization reversal has been observed to be much smaller in exchange-biased F layers than in single layers.[15,16] Additionally, due to surface imperfections, the strength of the exchange coupling varies at a microscopic scale. Microscopic regions in the F layer weakly and strongly coupled to the AF layer are found to coexist.[15,17] This should have an influence on the dynamic reversal processes. Several works have been reported on relaxation and thermally assisted magnetization reversal in exchange-biased systems, focusing on the temperature and time dependence of the biasing field.[18–21] A thermally activated reorganization of the magnetization in the AF layer takes place when the magnetization of the adjacent F layer switches. This implies that the reversal in the FM pinned layer is dependent upon the magnetic history of the AF layer.[21]

In order to obtain a better insight into the fundamental aspects of the reversal in AF/F bilayers, we performed room temperature dynamic magnetization measurements over 10 decades of applied field sweep rates on exchange-coupled polycrystalline NiO/Co bilayers. First, we will focus on the dynamical coercive field, the magnetization reversal process, and their dependence on both AF and F layer thickness. Second, using a simple phenomenological model, we will estimate the interface coupling energy of the AF/F bilayers. We will show that this is directly proportional to the inverse of a characteristic length related to the Barkhausen volume. This result is an experimental confirmation of the random field model for the AF/F coupling at the interface, first proposed in 1987 by Malozemoff.[6]

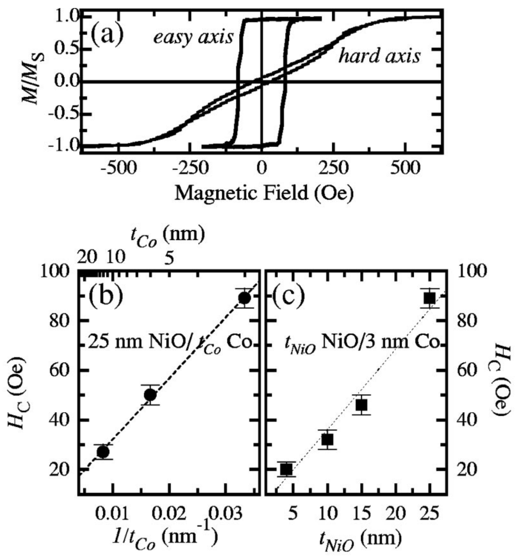

FIG. 1. Quasistatic magnetization reversal of NiO-Co bilayers. (a) Typical VSM RT magnetization curves of a 25 nm NiO/3 nm Co bilayer film along the easy (black) and hard (gray) axes. Coercive field as a function of the thickness of the (b) Co and (c) NiO layer. The symbols are experimental data and the lines are linear fits to $1/t_{\mathrm{Co}}$ and t_{NiO}, respectively.

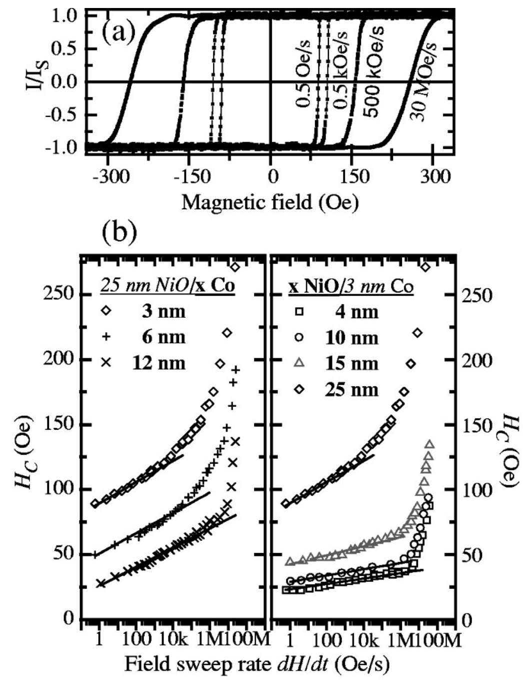

FIG. 2. Magnetization reversal dynamics of NiO-Co bilayers. (a) Longitudinal Kerr curves along the easy axis for different applied field sweep rate values of a 25 nm NiO/3 nm Co bilayer film. (b) Applied field sweep rate dependence of the coercive field of different exchange-coupled NiO-Co bilayers. Right and left panels show the NiO and Co thickness dependence, respectively. The symbols are experimental data and the lines are fits ($\chi^2<1$) using Eq. (1) (see text).

The NiO/Co bilayers investigated in this work were grown at room temperature in zero field on thermally oxidized Si wafers in a multisource sputtering unit described elsewhere.[22] NiO layers were deposited at oblique incidence by rf sputtering from a NiO target, leading to small grain sizes (4–7 nm) and a well-defined in-plane uniaxial anisotropy[22,23] of the bilayers [see Fig. 1(a)]. Co layers were deposited by dc sputtering. To determine the relevant mechanisms in the Co reversal process, we measured the hysteresis loops of the films using the longitudinal Kerr effect with applied field sweep rates (dH/dt) up to 10^9 Oe/s.[24] Hysteresis curves and coercivity were obtained by averaging over several (10–1000) magnetization cycles to improve statistics. The influence of both Co and NiO thicknesses on the reversal process was studied.

In quasistatic conditions, an enhancement of the coercive field with respect to a single Co thin film has been observed at room temperature for all films, without a significant shift of the hysteresis loops. The absence of exchange bias is expected if the NiO magnetization is dragged irreversibly by the Co magnetization when the latter reverses at the coercive field. This corresponds to the formation of a parallel domain wall in the NiO as in the Néel,[5] Malozemoff,[6] and Mauri *et al.* models,[7] and the subsequent thermally activated propagation of this domain wall throughout the AF layer. The easy thermal switching of the NiO in our samples is probably due to the small grain size.[9] The observed blocking temperature of our samples is around 200 K (depending on the NiO thickness).

Figure 1 shows that the coercivity depends on the thickness of the Co and NiO layers. The coercive field decreases (increases) for thicker Co (NiO) films. The dependence on Co thickness can be explained in terms of balance between the interfacial NiO/Co energy and the Zeeman volume energy of the Co layer. This idea was already given in the first papers on exchange bias.[1,6] As a result, the effective uniaxial anisotropy energy per unit volume associated with the interfacial coupling is expected to decrease as $1/t_{\mathrm{Co}}$ with increasing Co thickness (t_{Co}), which is indeed observed experimentally [Fig. 1(b)]. The coercivity dependence on the NiO thickness can be understood with the model described above. The torque imposed by the Co switching overcomes the torque due to the NiO anisotropy, and the NiO magnetization switches. The reorganization of the AF magnetization generates a large dissipation roughly proportional to the volume of the NiO layer. Consequently the coercive field of the Co layer increases with the NiO thickness, as observed in Fig. 1(c).

Some results of our dynamic magnetization measurements are shown in Fig. 2(a). Dynamical effects during the Co reversal are observed such as increasing coercivities and widening transitions when the applied field sweep rate dH/dt is increased. These observations indicate that the magnetization reversal process is thermally activated. As found by Raquet *et al.*[13,14] for Au/Co layers, these results suggest that the reversal is mainly governed by domain wall propagation at lower dH/dt, while for higher sweep rates domain nucleation processes dominate. Direct observation of the magnetization reversal process would be necessary to validate the above statement.

The dependence of the coercivity on dH/dt for a series of NiO/Co bilayers is plotted in the bottom graphs of Fig. 2. For all Co and NiO thicknesses the coercivity increases with increasing sweep rate dH/dt. On the other hand, our experimental results do not show any shift in the hysteresis loops whatever the chosen sweep rate and thickness. Stamps[18] suggests that an exchange bias should appear upon increasing dH/dt. We attribute the absence of exchange bias in our dynamic measurements to the so-called training effect, causing the interface spin arrangement of the NiO grains after several cycles to be random at zero Co magnetization (at H_C).

Dynamic measurements carried out with constant dH/dt point out unambiguously that also the dynamic processes in the AF layer *after* F reversal play a role in both exchange bias[19,21] and F coercivity.[21] Hughes *et al.* have observed that an increase of the waiting time with the F layer saturated in one direction causes an increase of the F reversal field in the opposite direction.[21] In our measurements, we have not observed any waiting-time dependence of the reversal. This indicates that the time needed to completely switch the NiO by thermal activation after F reversal is longer than the time scale used in our experiments. In the frequency range used here, the accessible waiting times after Co reversal are between some seconds and some microseconds. These times are small compared to the times (several minutes to hours) in which these effects have been observed at room temperature.[19,21]

Phenomenological models based on domain wall dynamics predict a logarithmic dependence of the coercive field on the applied sweep rate.[12,13] These models are based on thermally activated relaxation ("single relaxation time approximation") and assume that the energy barrier for magnetization reversal varies linearly with the applied magnetic field (i.e., domain wall propagation with weak pinning centers). Assuming a driving field varying at a rate dH/dt, the equations describing dM/dt can be integrated to yield $M(dH/dt)$, from which an expression of the dynamic coercive field $H_C(dH/dt)$ is obtained by solving $M(H_C)=0$:[13]

$$H_C=\frac{kT}{V^*M_S}\ln\left[\tau_{w_0}\left(\frac{dH}{dt}\right)\frac{V^*M_S}{kT}\ln 2+1\right], \tag{1}$$

where M_S is the saturation magnetization. V^* is the Barkhausen volume, i.e., the characteristic volume which reverses magnetization during a wall jump. τ_{w_0} is the relaxation time, i.e., the time to overcome the activation energy barrier ΔE in the absence of an applied field, with an attempt frequency τ_0 [$\tau_{w_0}=\tau_0\exp(\Delta E/kT)$]. V^* and τ_{w_0}, have been adjusted to fit the experimental results in the lower sweep rate range.

In a macroscopic sample many pinning centers and domain walls coexist. To give an accurate description of the observed relaxation phenomena [i.e., to model the $M(H,dH/dt)$ curves during the reversal] one should therefore take explicitly into account this distribution of activation energies. However, the hypothesis of a single activation barrier already allows us to interpret the gross features of the observations, $H_C(dH/dt)$, and yields an order of magnitude of the characteristic volume V^* and stability time τ_0 in-

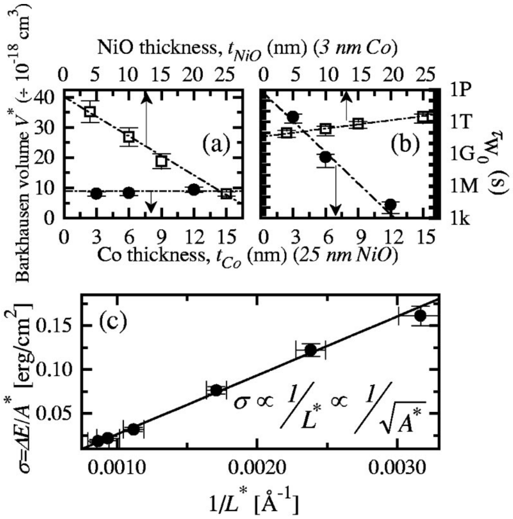

FIG. 3. (a) Barkhausen volume V^* and (b) characteristic time τ_w (b) obtained from the fits in Fig. 2, as a function of NiO (empty circles, top axis) and Co (filled squares, bottom axis) thickness. (c) Plot of σ vs $1/L^*$. The corresponding values for the energy barrier $\Delta E/A^*$ and the diameter of the activated area L^* are taken from Table I. Symbols are the experimental data and the discontinuous line is a linear fit.

volved in the activation process. The fits are displayed with solid lines on the bottom graphs of Fig. 2. The dH/dt validity range of the fits indicates that the transition from the domain wall propagation regime at lower dH/dt to the domain nucleation regime at higher dH/dt shifts to lower sweep rates [right panel of Fig. 2(b)] as the NiO thickness increases. The absence of a clear transition for the 25 nm NiO/Co bilayers reflects the coexistence of the two mechanisms during the reversal process. A similar crossover from domain wall propagation to nucleation can be observed with varying t_{Co} (left panel). In this case, as the Co thickness increases the transition takes place at higher dH/dt and the switching behavior approaches that of a Co single film. These results are coherent with the ones of Chopra *et al.*[15] who observe that in a NiO/Co bilayer with thicker NiO (50 nm) and thinner Co (1 nm) layers the propagation mechanism does not dominate the reversal even in a quasistatic regime.[15]

The parameters obtained from the fits are displayed in Fig. 3. The Barkhausen volume V^* decreases linearly with increasing NiO thickness while it stays more or less constant with Co thickness. The relaxation time at zero field τ_{w_0} increases slowly with NiO thickness and decreases quickly with Co thickness. For small t_{Co} the Barkhausen volume includes the whole Co thickness ($V^*=A^*t_{\mathrm{Co}}$). The ratio between the energy barrier ΔE and the area of the activation volume A^* is a measure of the interfacial coupling energy ($\sigma=\Delta E/A^*$). The latter and the length L^* corresponding to the activation volume ($L^*=2\sqrt{V^*/\pi t_{\mathrm{Co}}}=2\sqrt{A^*/\pi}$), calculated from the parameters V^* and τ_w in Fig. 2(c), are given in Table I. We find that the interface energy is inversely proportional to the effective activation length ($\sigma\propto 1/L^* \propto 1/\sqrt{A^*}$) as shown in Fig. 3(c). This is in agreement with

TABLE I. Thickness dependence of the effective length of the activation volume (L^*) and of the interface coupling ($\sigma \approx \Delta E/A^*$). Both have been extracted using the parameters V^* and τ_{w_0} displayed in Figs. 3(a) and 3(b), respectively: $L^* = 2\sqrt{V^*/\pi t_{Co}}$ and $\Delta E = \ln(\tau_{w_0}/t_0)/kT$. The attempt frequency has been taken equal to 10^9 Hz (Ref. 25).

t_{NiO}/t_{Co} (nm)/(nm)	4/3	10/3	15/3	25/3	25/6	25/12
L^* (nm)	115 ± 10	107 ± 9	89 ± 6	59 ± 3	42 ± 2	32 ± 2
$\Delta E/A^*$ (merg/cm^2)	18 ± 2	22 ± 2	32 ± 3	76 ± 4	122 ± 7	161 ± 10

models such as the one proposed by Malozemoff, where a random walk argument is used to show that the exchange anisotropy energy is inversely proportional to some typical length scale (square root of area) in the AF material. This length scale can be imposed by random interface roughness, but can also be related to the local average of the random anisotropy of the NiO grains in the activation area.

In summary, we have presented a study at room temperature of the magnetization reversal in polycrystalline exchange-coupled NiO/Co bilayers as a function of applied sweep rate dH/dt, for different NiO and Co thicknesses. For all sweep rates and thicknesses, the symmetry of the hysteresis loops reflects that an identical pinning strength has to be overcome in both directions of the reversal. The experimental results suggest that domain wall displacement and domain nucleation regimes govern the magnetization reversal at low and high sweep rates, respectively. The sweep rate of the crossover depends on the relative weight of the Co and NiO layers: thicker Co favors propagation and thicker NiO favors nucleation. For a constant sweep rate the coercivity increases (decreases) for thicker NiO (Co). It was found that the interface energy is inversely proportional to the area of the activation (Barkhausen) volume. These results are consistent with a model of exchange anisotropy in which a random walk argument of the coupling between AF and F layer due to the high frustration of exchange interactions along the AF/F interface caused by interfacial roughness and/or random anisotropy of the NiO grains is combined with a thermally activated switching process of the NiO magnetization.

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[25] One (two) order of magnitude difference in τ_0 gives ΔE values a 5% (10%) difference.