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Magnetic relaxation in thin Co films with in-plane magnetization

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Magnetic relaxation along two nonequivalent axes has been investigated in Co films on stepped Cu(001) by magneto-optical Kerr effect and correlated to magnetic domain patterns and structural defects by spin-polarized scanning electron microscopy. The results show that nucleation is dominated by anisotropy rather than magnetostatics. Domain wall velocity is not constant for a given field, but depends on the initial state of the system.

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Magnetic switching in thin films is of great technological importance because numerous devices rely on the fact that a magnetic system can be bistable. Switching from one state to the other is achieved by a magnetic field. As soon as the field is applied, one state becomes metastable, and the system proceeds through a large number of metastable states of decreasing energy until the ground state is reached. Temperature and applied field determine the probability to overcome the barriers separating metastable states, and thus the system evolves with time even though field and temperature are kept constant. This relaxation phenomenon is also referred to as an after-effect or a creep. The long-term stability of magnetic storage media is largely determined by this effect.[1] Despite its importance, only a few studies have investigated relaxation in thin films magnetized in plane.[2,3] Most studies were performed in perpendicularly magnetized films.[4–10] Although these are analogous to in-plane magnetized systems with uniaxial symmetry from an anisotropy perspective, the magnetostatic energy might make reversal behave differently.

A conceptually more complex case are in-plane systems with higher symmetry, for which higher orders become relevant for in-plane anisotropy. Apart from easy and hard magnetization axes, further nonequivalent ''intermediate'' axes can exist. Consequently, magnetization relaxation along different axes can be compared.

We show that relaxation in such systems displays pronounced differences along nonequivalent directions, both with respect to nucleation and domain-wall propagation. This allows us to disentangle qualitatively the relative influence of crystalline anisotropy and the magnetostatic energy contribution on relaxation. No study has reported magnetization relaxation experiments on the identical sample along two nonequivalent directions.

A 30-monolayer (ML)-thick epitaxial cobalt film has been prepared by molecular beam epitaxy on a Cu(001) single crystal with 1° miscut. The step edges were preferentially oriented along the [110] direction. The in-plane anisotropy is described by a superposition of twofold and fourfold anisotropy, $K_u\sin^2(\phi)+(K_1/4)\sin^2(2\phi)$, where K_u and K_1 are the uniaxial and the cubic anisotropy constants, and ϕ the angle between the magnetization and the $[1\bar{1}0]$ direction.[11] For this system, $K_u=7.8\times10^3$ J/m^3 and $K_1=89.2\times10^3$ J/m^3. For this film thickness, the easy axis is perpendicular to the step edges, i.e., along the $[1\bar{1}0]$ direction, whereas the intermediate axis is parallel to the step edges, i.e., along the [110] direction.[12]

The relaxation experiments were performed by *in situ* magneto-optical Kerr effect in the transverse and longitudinal geometry for measurements with field along the easy and intermediate axis, respectively. The laser spot diameter is about 1 mm. Two pairs of coils are positioned at 90° to each other in the plane of the sample. A field 30 times larger than the coercive field was first applied to fully saturate the magnetization. Then the field was reversed to a value smaller than that of the coercive field, and the Kerr signal was recorded as a function of time. The measured rise time of the field step is <1 ms. After removal of the field, magnetic domain images were taken by spin-polarized scanning electron microscopy (spin-SEM).[13] All experiments were performed at room temperature.

Relaxation along the easy axis proceeds by a succession of jumps in the magnetization, see Fig. 1. For a given field, the time at which jumps occur as well as their height are not constant because the process is thermally activated. Usually, such relaxation curves are analyzed within a model extending Fatuzzo's work.[5] The model assumes that reversal takes place by nucleation of domains and their subsequent expansion by wall propagation, described by the nucleation rate, the initial size of the nuclei, and the wall velocity. In the simplest approximation, the observed crossover in the shape of the relaxation curve from an almost exponential decay at low fields to an *S*-shaped curve at high fields is associated

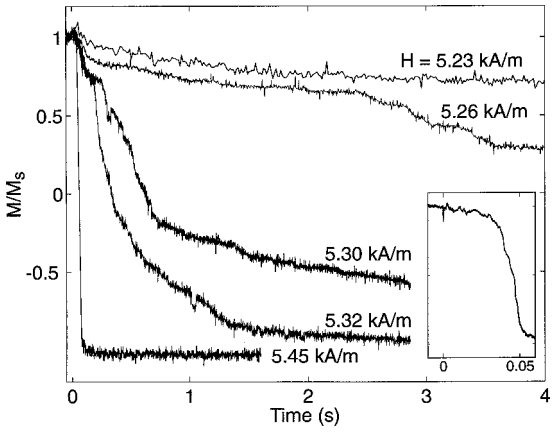

FIG. 1. Magnetic relaxation with field applied along the easy axis. At the time $t=0$ s, the magnetic field was switched from saturation to a reverse field H given at each curve. The Kerr signal M is normalized to saturation magnetization M_s. The inset shows the *S*-shaped initial relaxation curve at large fields, $H=5.45$ kA/m.

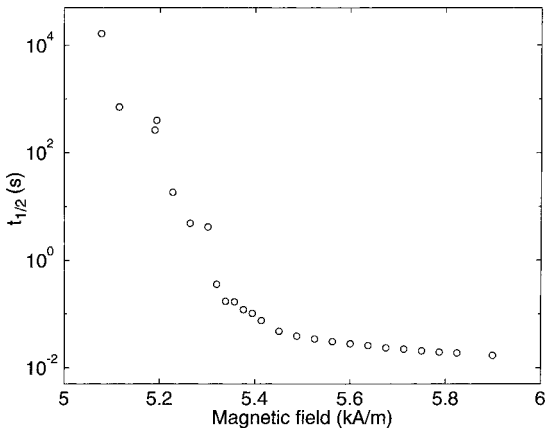

FIG. 2. Logarithmic plot of the relaxation time $t_{1/2}$ vs reverse field H; $t_{1/2}$ is the time needed to reverse half of the magnetization, i.e., $M(t=t_{1/2})=0$. Note the steep slope at small fields and the flat curve at large fields.

with a decay governed by domain nucleation and wall propagation, respectively.[6] As three parameters are involved, however, such an identification is generally not unambiguous and requires additional analysis.

Relaxation can be characterized by the time $t_{1/2}$ needed to reverse half the magnetization over the probed area. At low fields, an Arrhenius-type model yields $\ln(t_{1/2}/\tau)=-2M_sV_B(H-H_p)/kT$. The characteristic time τ comprises all atomistic quantities related to a typical reversal time, M_s is the saturation magnetization, V_B the Barkhausen volume, H_p the wall propagation field, and kT the Boltzmann constant times temperature. The Barkhausen volume, i.e., the volume swept by the wall in moving from one minimum to the other, can be determined from the linear slope of the $\ln t_{1/2}(H)$ curve in Fig. 2 at low fields,[4] $V_B=86\times10^{-24}$ m^3. At higher fields, a break in slope is observed. It has been shown earlier that the high-field regime corresponds to a viscous motion of domain walls rather than thermal activation.[14] Correspondingly, the logarithmic dependence of $\ln t_{1/2}$ with H is no longer valid.

Additional information on the relaxation process is obtained by direct visualization of the domain structure by spin-SEM. We find that reversal takes place by nucleation at only a few structural defects in the film, enlarging to big inverted domains, see Fig. 3(a). Once a domain has been nucleated at such a macropin,[3] it expands to a certain size determined by field strength and pulse duration, and eventually pins at local barriers for wall propagation. Qualitatively the same behavior is found for the entire field range probed in Fig. 2.

To study unpinning of a domain at local barriers we performed reversal experiments with a sequence of two field pulses separated by a variable time delay and measured the Kerr signal during this sequence. Figure 4 shows that the magnetization decay after two separated pulses is strongly suppressed compared to a single pulse of double length. The corresponding domain image after the second pulse confirms this picture, see Fig. 3(b). The existing domain of Fig. 3(a) has expanded only marginally even though the second pulse has been applied during a much longer time. Hence, the

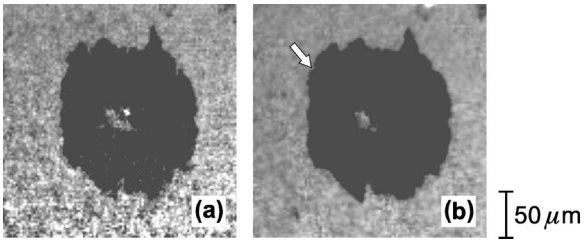

FIG. 3. Magnetization images taken by spin SEM. The magnetization component along the easy axis is measured. A reversed (black) domain has nucleated at a defect in the image center and expanded (a) after the application of a reversed field of 5.08 kA/m for 11 s and (b) after a second pulse of identical strength for 1000 s. The small region expanded with the second field pulse is indicated by an arrow.

magnetization decay during the second pulse in Fig. 4 is caused by nucleation and subsequent expansion of an additional, previously nonexistent domain. The inefficiency of the second pulse is present even for the shortest time delays applied between the two pulses, i.e., for 0.1 ms. During this short time in zero field the system continues to relax to lower its energy, albeit on a path different from the one with field applied. Such a path is governed either by diffusion or thermal activation.[15] In a diffusional process, atoms or impurities move at the Co/Cu interface or in the Co film, thereby inducing a change in magnetic anisotropy.[16] However, the time scale for such a process is generally on the order of seconds rather than milliseconds. Moreover, diffusion processes follow the ''superposition principle'' of magnetic after-effects[15] which is not compatible with the inefficiency of the second pulse. The process to lower the energy is therefore a thermal activation in which the wall can overcome small barriers to reduce its energy further, with a competition between two terms typical for creep:[10] the first one drives the wall to a local position where the energy density is minimum; the second tends to reduce the wall length. When the next pulse starts, the system will thus be in a different, lower energy

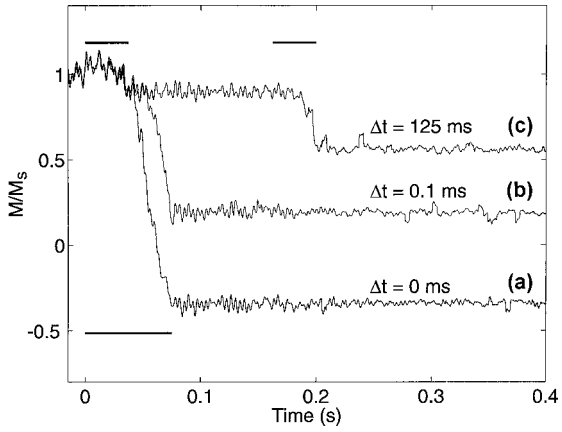

FIG. 4. Magnetic relaxation with field along the easy axis with two reverse field pulses $H=5.43$ kA/m of 40 ms duration, and a delay time Δt between the two pulses. Between the pulses, no field is applied. The temporal position of the pulses is indicated at the bottom for (a) and (b), at the top for (c).

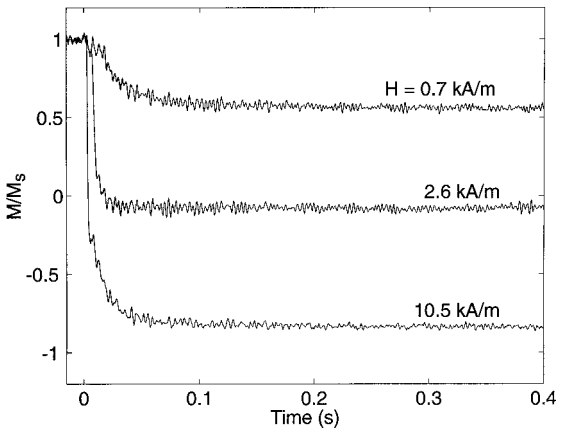

FIG. 5. Magnetic relaxation with field applied along the intermediate axis. The magnetic field was switched from saturation to a reverse field H given at each curve. The magnetization stays constant after a fast initial decay.

state than at the end of the previous pulse. Therefore, the wall velocity is not constant in a given field, but depends on the initial state of the system.

The stepped Co/Cu(001) system offers the unique possibility to directly compare relaxation along two nonequivalent crystallographic directions. In addition to the reversal experiments along the easy axis, we performed the same set of experiments along the intermediate axis. In contrast to reversal along the easy axis, the relaxation curve has the same shape for all fields applied, see Fig. 5. Within a few tens of milliseconds, the Kerr signal reaches a value with no additional decrease for several minutes. This value is the same as the one attained at this field while cycling a hysteresis loop.

The domain image in Fig. 6(a) shows that a large number of nucleation sites was present before wall propagation, distinctly different from the images taken along the easy axis. Moreover, reversal proceeds through an intermediate step with domains aligned along the easy axis at 90° to the field direction.[2] For an applied field smaller than the uniaxial anisotropy field of 4.3 kA/m, newly created domains are magnetized along the easy axis; only for applied fields larger than the uniaxial anisotropy field does the magnetization in

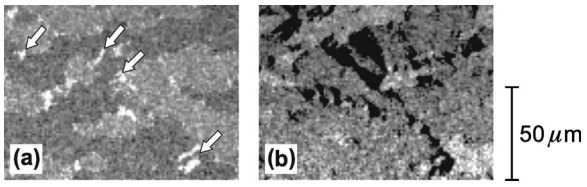

FIG. 6. Magnetization images taken by spin-SEM at the identical area of the sample. After saturation of the sample, a reverse field pulse H of 0.2 s has been applied along the intermediate axis. The magnetization component along the intermediate axis appears black and white, the component along the easy axis light and dark gray. (a) $H=1.9$ kA/m. The two gray levels correspond to domains oriented in opposite directions along the easy axis, except at defects in the film as seen in topography (arrows) which keep their original white magnetization direction. (b) $H=9.3$ kA/m. Black regions appear in which the magnetization has reversed.

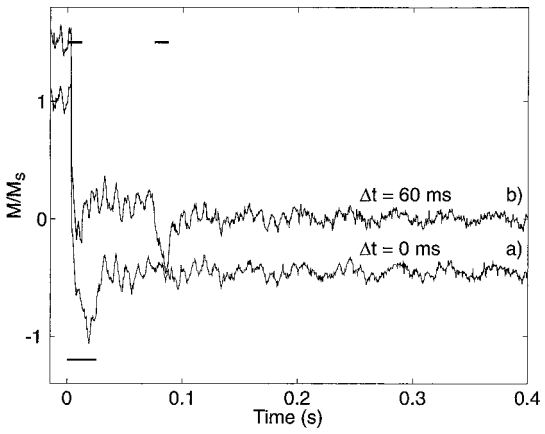

FIG. 7. Magnetic relaxation with field along the intermediate axis with two reverse field pulses $H=11.2$ kA/m of 60 ms duration and a delay time Δt between the two pulses. Between the pulses, no field is applied. The temporal position of the pulses is indicated at the bottom for (a), at the top for (b). The two curves are vertically displaced for clarity, the magnetization reaches the same final value in (a) and (b).

some domains point along the applied field direction, see Fig. 6(b).

Strikingly, the magnetization at defects visible in topography still points along the original direction even though most of the film has already switched magnetization by 90°, see Fig. 6(a). Thus, nucleation at defects is more difficult than in the rest of the film. This means that these defects break the symmetry: although they favor nucleation for the magnetization along the easy axis, they hinder it if the magnetization points along the intermediate axis. As magnetostatic energy at defects is on average independent of direction, we conclude that nucleation in the stepped Co/Cu(001) system is controlled by the uniaxial anisotropy field rather than by the demagnetizing field. Anisotropy is hence the key factor to control nucleation.

The uniaxial anisotropy is reduced at defects because step edges are no longer preferentially oriented along [110]. On an atomic scale, any step orientation is composed of segments along [110] and $[1\bar{1}0]$.[17] A deviation of the step edges from the [110] direction thus leads to a reduction of K_u. At defects, this leads to a decrease of the nucleation barrier for a sample initially saturated along the easy axis, and an increase of the barrier for a sample initially saturated along the intermediate axis, so that the spatial distribution of nucleation centers depends on the saturation direction. This explains why defects play a completely different role in magnetization reversal along the two inequivalent directions. The height of the barrier also varies with the step density, but this effect seems less drastic than the influence of step orientation. We note that anisotropy changes at defects are dominant in the nucleation process over demagnetization effects at the sample edges. Indeed, if nucleation were easier at the edge than at macropins, reversal should proceed by nucleation at an edge or a corner, and sweep a wall through the sample, eventually pinning at a defect. This is not what is observed (see Fig. 3).

To check whether the different reversal behaviors are re-

flected in the wall pinning, we performed relaxation experiments with two pulses separated by a time delay (see Fig. 7). The behavior is completely different from the one along the easy axis. When the field pulse is switched off, an appreciable relaxation of the magnetization towards the easy axis is observed. We conclude that the 90° walls are able to adjust over large distances, contrary to the 180° walls governing reversal along the easy axis. This difference can be traced back to the fact that the energy for a 90° wall is about a factor of 2 smaller than that for a 180° wall. Hence the wall can locally adjust its position at energetically favorable pinning sites, preferably by an increase in length of the 90° walls.

In conclusion, the study of the time dependence of magnetization relaxation and the corresponding domain configurations in a stepped Co/Cu(001) film has led to insight into domain nucleation and wall propagation. Whereas nucleation and subsequent wall propagation evolves through 180° walls along the easy axis, 90° walls are the route for reversal along the intermediate axis. We have shown that magnetic anisotropy rather than local demagnetization energy is the key determining factor of domain nucleation at defects. In particular, defects act as nucleation centers for 180° walls, but not for 90° walls. This observation is related to the perturbed step orientation at defects, which leads to a reduced local uniaxial anisotropy. Correspondingly, the nucleation barrier decreases for 180° walls, but increases for 90° walls. We have shown that wall propagation and pinning are largely influenced by anisotropy as well. More studies are needed to understand wall velocity in a system of superimposed uniaxial and fourfold anisotropy, and its dependence on magnetic history. The topic is complicated by the fact that both step density and step orientation are important, so that a connection must be made between wall position and nanoscale defects acting as propagation barrier.

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