Magnetization Direction Switching in Fe/Cu(100) Epitaxial Films: Temperature and Thickness Dependence

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The magnetization direction switching is investigated in epitaxial Fe/Cu(100) films by spin-polarized scanning electron microscopy. We follow the transition from perpendicular to in-plane magnetization with both increasing film thickness and varying temperature. No variation of magnetic moment with magnetization direction change is found. A perpendicular stripe domain configuration is identified which evolves from the low-temperature single-domain state during the reorientation phase transition.

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Ferromagnetic ultrathin films display a wealth of spectacular properties characteristic of two dimensions. One of their most remarkable features is that 2D ferromagnetism is stabilized exclusively by magnetic anisotropies [1]. There are several contributions to these anisotropies, the most prominent of which is the generally large uniaxial surface anisotropy predicted for [2] and observed in [3-6] numerous epitaxial systems. It is essentially the competition between this surface anisotropy and the shape anisotropy that determines the easy magnetization direction. Quite generally, if the balance between these quantities is changed, the magnetization direction can undergo a transition from being oriented perpendicular to the film surface towards being oriented in the plane. Remarkably this subtle balance is experimentally approachable since it hinges on such parameters as temperature, film thickness, and magnetic field.

This magnetization direction switching is particularly appealing in 2D. Recent theoretical work [7,8] points out the importance of thermodynamic fluctuations and entropy for the magnetization reorientation in 2D. No predictions, however, exist for the detailed nature of this phase transition. Experimentally such a magnetization direction switching has been observed for a variety of systems [3,5,9-11]. An easily accessible quantity for monitoring the transition from perpendicular to in-plane magnetization is the remanent magnetization $\mathbf{M}_R \equiv \mathbf{M}(H=0)$ measured along the various directions after applying field pulses. For fcc Fe/Cu(100), Pappas *et al.* [3] find a film thickness range of 5 ML $\leq d_s \leq$ 6.5 ML (ML denotes monolayer) where a reversible magnetization direction switching occurs at temperatures in the range 300 K $\geq T_s \geq$ 140 K. The most intriguing observation is that $\mathbf{M}_R$ vanishes along all three spatial directions at the film thickness d_s where switching occurs. Even more spectacular is that $\mathbf{M}_R$ vanishes not only at the switching temperature T_s but also in an extended finite temperature interval of $\Delta T_s \simeq 30$ K. This has not yet been satisfactorily explained, but we expect that it is intrinsic to the 2D nature of these films. It has been speculated that a paramagnetic intermediate state might exist, since the surface anisotropy and the short-range part of the shape anisotropy cancel at T_s. The symmetry arguments of Ref. [1]—although not strictly fulfilled because of the long-range part of shape anisotropy—might then become more relevant. Another likely possibility is that domains form during reorientation. Note that even an antiferromagnetic state cannot be ruled out by the present experimental results [9].

The second important observation concerns films of thickness $d > d_s$, in which $\mathbf{M}_R$ is considerably reduced. This reduction has been attributed to a loss of magnetic moment from $2.2\mu_B$ to $1.4\mu_B$ in fcc Fe/Cu(100) [9]. We note that such a reduction has not been observed in bcc Fe/Ag(100). Because of this dissimilar behavior one could argue that the reduced $\mathbf{M}_R$ in Fe/Cu(100) is a consequence of the structural transition from a tetragonally distorted to a complex phase at $\simeq 6$ ML [12].

Our aim is to resolve these fundamental aspects of the magnetization direction reorientation in fcc Fe/Cu(100). Domain imaging experiments were used to investigate locally the switching behavior with increasing film thickness and temperature. The technique employed is known as spin-polarized scanning electron microscopy (spin-SEM [13] or SEMPA [14]) and, with its ML sensitivity, is exceptionally well suited to addressing these questions. We will show how the spontaneous magnetization direction switching takes place with increasing film thickness, and compare it to the remanent magnetization. The main experimental effort is devoted to investigating the formation of domains with increasing temperature, starting with a uniformly magnetized film at low temperature. A characteristic series of domain patterns develops with temperature during magnetization reorientation that has not been observed before. It can be understood in the framework of renormalization-group theory.

All films were epitaxially grown onto a Cu(100) single crystal held at low temperature, $T=90$ K, which is known to produce sharp interfaces between substrate and film [15]. The pressure during evaporation was $<2 \times 10^{-10}$ mbar, and the films were subsequently annealed to 300 K. This procedure has been adapted from Ref. [3], and characterization by LEED indeed shows that their reconstruction patterns evolve with thickness. For details of domain imaging with our spin-SEM, see Ref. [16]. Compared to our earlier experiments, we decreased the energy of the probing electron beam from 10 to 2 keV, and kept the beam current below 1 nA. This was done to avoid structural and magnetic changes of the Fe films, which we have found to occur when our standard electron gun parameters are used. In particular, d_s has been observed to shift to smaller values if the films are contaminated with minute traces of oxygen [3].

The thickness dependence of the spontaneous magnetization $\mathbf{M}_S$ (and hence of the magnetization reorientation) can be investigated on a single sample by growing a shallow Fe wedge [17,18]. Figure 1 displays domain images for both perpendicular and parallel magnetization along a wedge with an Fe thickness of $0 \le d \le 10$ ML measured at $T=175$ K. A magnetic field pulse has been applied to eliminate perpendicular domains present in the as-grown film. For $d \le 2.3$ ML, no magnetization is found perpendicular or parallel to the sample surface since $T_C < 175$ K [9]. In the range 2.3 ML $\le d \le 5.3$ ML, perpendicular magnetization prevails. Above $d_s = 5.3$ ML, in-plane magnetization is observed. We extract twenty line scans along the wedge to plot $\mathbf{M}_S(d)$ directly. Figure 2 displays both perpendicular and parallel spin polarization components along the [001] direction, $M_{S\perp}$ and $M_{S\parallel}$, and the calculated magnitude of the spontaneous magnetization, $M_S = [(M_{S\perp})^2 + (M_{S\parallel})^2]^{1/2}$.

Two remarkable features are observed:

(i) The spontaneous magnetization is not reduced for the in-plane films ($d > d_s$), but follows an increase as expected for a thickness-independent magnetic moment and an inelastic mean free path of secondary electrons of 3.8 ML. The earlier discovery of a reduced remanent magnetization must therefore be attributed to the occurrence of domains within the film, i.e., to the inability to establish a single-domain remanent state over the sampling area of 1 mm. Note that in our wedge sample we also find reversed domains for $d > d_s$; see Fig. 1. A recent theoretical study calculates that the magnetic moment depends on the magnetization direction, since spin-orbit coupling might no longer be quenched in ultrathin films [19]. This effect is predicted to be small, of the order of a few percent. The statistics of our data currently do not allow us to check this prediction with sufficient accuracy. We emphasize, however, that no change of the order of 10% occurs. Therefore Fe/Cu(100) behaves analogously to previously investigated systems, such as Fe/Au(111) [6], Fe/Ag(100) [9], and Co/Au(111) [16], contrary to earlier belief.

(ii) We clearly find no thickness range where both $M_{S\perp}$ and $M_{S\parallel}$ vanish. As $M_{S\perp}$ decays, $M_{S\parallel}$ already sets in. On the other hand, we see a pronounced drop of M_S at switching thickness. However, small irregular in-plane domains with reversed magnetization exist just at d_s; see Fig. 1(b). The apparent reduction in M_S therefore might originate from the average over these small domains by large-scale scans rather than from a truly reduced magnetization [20]. One might argue that this reasoning is partly invalidated in wedge-type samples by mutual magnetostatic interaction between adjacent regions of different thickness. A typical stray-field range is of the order of 10 μm, which translates into a thickness change of $\simeq 0.4$ ML in our wedge sample. To avoid this possible pitfall, the characteristic drop at d_s shown in Fig. 2 also

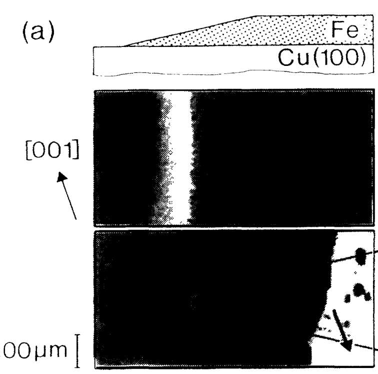

FIG. 1. (a) Magnetic domain images of a $0 \le d \le 10$ ML Fe wedge atop the Cu(100) substrate, grown at $T=90$ K and measured at $T=175$ K. A side view of the position of the wedge relative to the images is shown on top. Upper panel: magnetization component perpendicular to surface; lower panel: magnetization component parallel to surface along the [001] direction. Gray scale varies from spin polarization $P=-30\%$ (black) to $P=+30\%$ (white). (b) Enlargement of region at switching thickness d_s for the in-plane magnetization component. Small reversed domains of several micrometers in size appear.

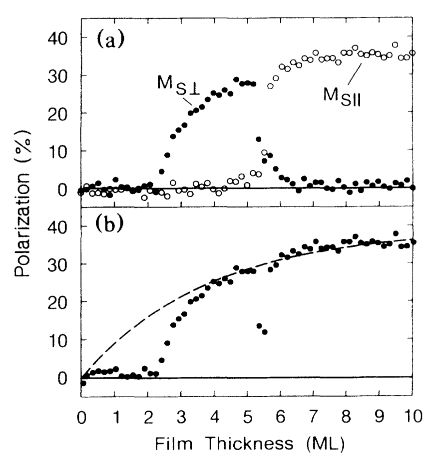

FIG. 2. (a) Perpendicular ($M_{S\perp}$,●) and parallel ($M_{S\parallel}$,○) spin polarization vs film thickness, averaged over twenty line scans for the wedge in Fig. 1. (b) Total spin polarization (or spontaneous magnetization) $M_S = [(M_{S\perp})^2 + (M_{S\parallel})^2]^{1/2}$ vs film thickness, as calculated from (a). The expected increase with film thickness is given by the dashed line, assuming an inelastic mean free path of 3.8 ML. No reduction of magnetic moment is observed at $d > d_s$.

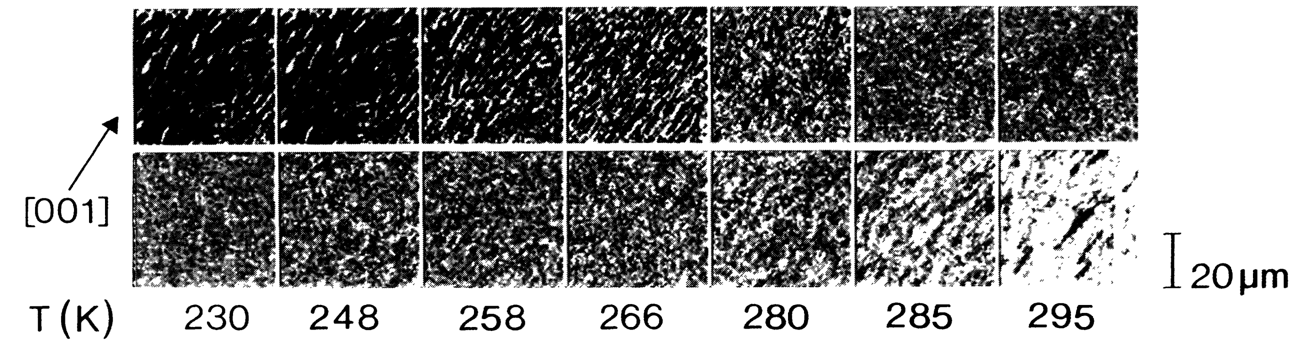

FIG. 3. Evolution of domain pattern with increasing temperature T for an Fe/Cu(100) film of constant thickness d_s grown at $T=90$ K. Upper panel: magnetization component perpendicular to surface; lower panel: one magnetization component parallel to the surface. Below $T=230$ K, the film is a perpendicular single domain. Note the change to stripe domains during magnetization direction reorientation.

needs to be investigated in films of *constant* thickness. These results will be discussed in the following.

The evolution of the domain pattern with increasing temperature is shown in Fig. 3 for a film of fixed thickness d_s. Below $T=230$ K, the film is perpendicularly magnetized and has been made single domain by applying a short field pulse. At $T=230$ K, some irregularly shaped reversed domains nucleate at a typical distance of $\simeq 20$ μm, preferably at defects visible in topography. At $T=248$ K, more nucleation centers appear, and they tend to be elongated in shape. At $T=258$ K, stripe domains have formed along the [001] direction with distances narrowing with increasing temperature. Between 280 K $\leq T \leq 285$ K the transition to in-plane magnetization takes place, which is completed at $T=295$ K where in-plane domains of varying size are visible.

Note that the magnetization reorientation from perpendicular to parallel occurs over a finite temperature interval of $\Delta T_s \simeq 50$ K. Our data definitely rule out that a loss of long-range ferromagnetic order is responsible for the vanishing remanence. To elucidate this finding further, Fig. 4 shows a direct comparison of spontaneous and remanent magnetization as deduced from the domain images. The value of $\mathbf{M}_R$ is approximated by the polarization average over the scan area. Figure 4 demonstrates that $\mathbf{M}_R$ vanishes in a finite temperature interval around T_s, and decays appreciably in a broad temperature range. $\mathbf{M}_S$, on the other hand, does not vanish in such an extended transition, and switches within a narrower interval. Note that Fig. 4 implies that $\mathbf{M}_S$ might also be reduced in a much narrower temperature interval of < 10 K at T_s. The investigation of the critical behavior of this remarkable phase transition is beyond the scope of this paper. At present we concentrate on the series of domain patterns of Fig. 3 in the extended temperature range of 60 K.

The most striking observation is certainly the formation of a stripe pattern initiated by the nucleation of small reversed domains. Yafet and Gyorgy [21] have predicted that stripe domains are energetically preferred over a uniformly magnetized state owing to the presence of long-range magnetic dipolar interaction. The stripe period, however, is only accessible to the experiment if the surface and shape anisotropy nearly cancel. Otherwise, a single domain should prevail on the length scale of any experiment. The observed transformation of the pattern from a single domain at low temperature to a striped configuration near T_s qualitatively confirms these predictions. This is quite astonishing in view of the fact that their calculation was limited to zero temperature and fails near the phase transition where magnetization is no longer saturated.

A more detailed theory lifts these restrictions by taking the finite-temperature spin fluctuations into account in a renormalization-group analysis [22]. These authors are able to confirm the stripe pattern of Ref. [21]. In addition, they find a preferential orientation of the stripes that is stable against thermal fluctuations. They predict the [001] directions to be favored, in agreement with our observation. In their theory the narrowing of the stripes on

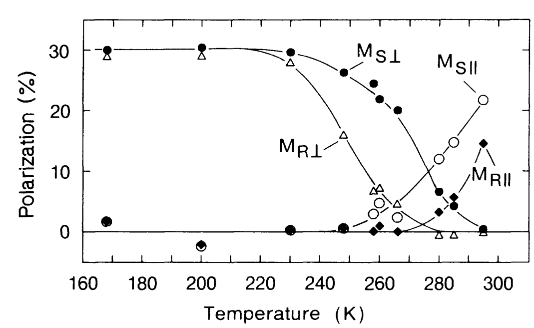

FIG. 4. Perpendicular and parallel components of spontaneous magnetization $\mathbf{M}_S$ and remanent magnetization $\mathbf{M}_R$ vs temperature as deduced from the analysis of the images in Fig. 3. The value of $\mathbf{M}_R$ is taken as the average over the scan area. Note the drop in $\mathbf{M}_R$ in a much wider temperature range than that of $\mathbf{M}_S$.

approaching T_s occurs because of the strong decrease of the surface anisotropy. The limiting domain width L_d is determined at switching by exchange integral and dipolar energy. The agreement between the observed minimum width of $\simeq 2$ μm and $L_d \simeq 0.6$ μm is reasonably good, given the fact that a very small external magnetic field is predicted to partly suppress the formation of equally spaced stripes [23].

In conclusion, we have investigated the magnetization direction reorientation in fcc Fe/Cu(100) films with changing film thickness and temperature. We show that, in contrast to earlier indications, the magnetic moment is not reduced for the in-plane films. With increasing temperature, the switching evolves in a distinct pattern: perpendicular single domain, followed by nucleation sites, stripe domains, and finally irregularly shaped in-plane domains. We compare our results with calculations based on a renormalization-group approach. We expect that such a characteristic evolution of the magnetization direction might occur not only with film thickness and temperature, but also with applied field [24]—a fascinating topic for future experimental work.

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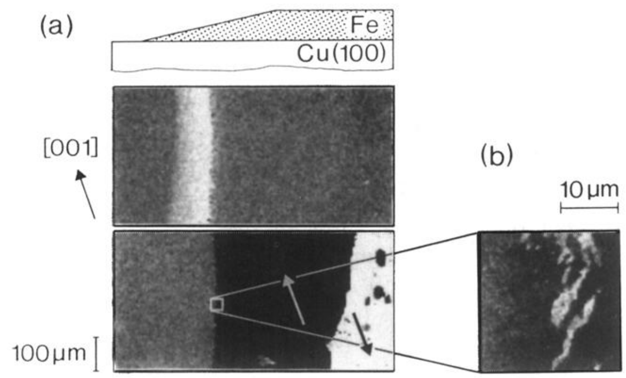

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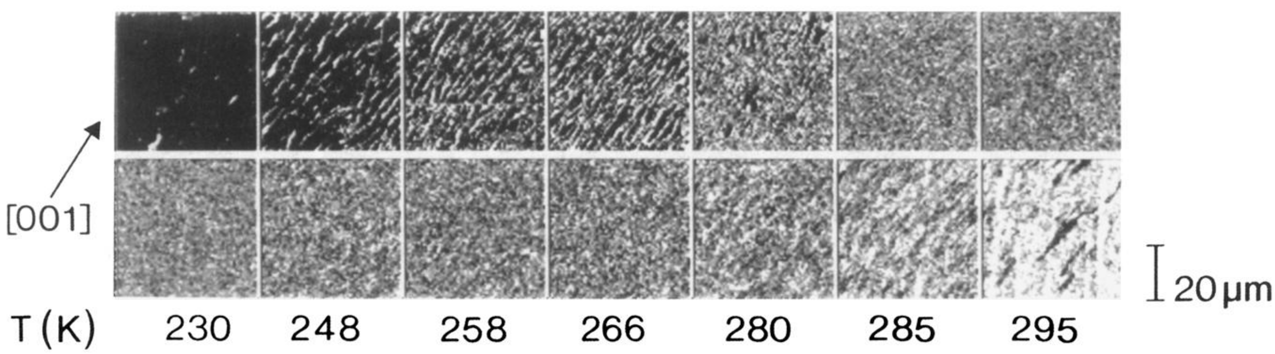

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