Magnetic Domain Formation in Fe Films on Cr(100)

H. Hopster

Department of Physics and Astronomy and Institute for Surface and Interface Science,
University of California, Irvine, California 92697

Magnetic domain images of thin (2 nm) Fe films on Cr(100) are presented. Upon cooling a single-domain state transforms into a state with a locally varying in-plane magnetization direction. This transformation is partially reversible and highly reproducible. It is suggested that the in-plane rotation is driven by frustration that favors 90° coupling, and that the local magnetization direction reflects μm-scale variations of the atomic scale roughness.

There is currently a great interest in ferromagnetic/antiferromagnetic (FM/AF) interfaces. "Exchange biasing" is becoming increasingly important in novel magnetic devices. In exchange biased films the hysteresis loop due to the coupling to the AF film is shifted from zero field as if an external field was present. Although discovered more than forty years ago the effect is not well understood on an atomic scale [1]. In addition, short-period oscillations in the exchange coupling between Fe films through Cr in very flat samples [2] have kindled interest in the magnetic structure of Cr layers since the coupling is believed to be closely tied to the AF structure of the Cr films. The magnetic order in thin Cr layers in relation to the well-known incommensurate spin density wave ordering of bulk Cr [3] is actively being investigated [4].

The present study was motivated by a magneto-optical Kerr effect investigation of Fe films on Cr(100) [5], which showed unusual temperature dependent magnetic properties. Fe films thinner than 5 nm show anomalous temperature dependencies of the coercive field and the remanent magnetization decreases below the Cr Néel temperature (311 K). The effects were attributed to frustrated magnetization structures. Frustration is a common phenomenon when AF interactions and steps are present simultaneously, and it plays a crucial role in most theoretical models of AF/FM interface structures [6]. A recent example is "biquadratic" (or 90°) coupling between FM layers [7]. Steps were recently shown to induce the Fe magnetization on stepped Cr surfaces to switch between parallel and perpendicular to the steps as a function of step density [8].

This paper gives a microscopic picture of the temperature dependent magnetization of an ultrathin FM film (2 nm Fe) on a nominally flat AF substrate (Cr). A single-domain state evolves into a state in which the magnetization direction varies on the micron scale when cooled below T_N.

The experiments were performed in a new UHV system that combines molecular-beam epitaxy growth and characterization capabilities with magnetic microscopy using secondary electron microscopy with polarization analysis (SEMPA) [9]. The rectangular Cr(100) crystal and the cleaning and growth conditions are the same as the ones used in Ref. [5]. However, the Cr crystal has been reoriented and repolished [10]. The electron gun for SEMPA measurements consists of a Shottky field emission tip, single-lens focusing, and an octupole deflection unit [11]. For sample alignment the SEM can be operated in the usual TV mode. The low-energy secondary electrons are extracted into a medium-energy Mott detector [12]. Two components of the spin polarization can be measured simultaneously. Figure 1 shows the sample and measurement geometry. The incoming electron beam is about 35° off normal and the extraction lens into the Mott detector about 10° off normal so that one measures basically the two in-plane magnetization components (P_x and P_y), as indicated. In addition, an intensity image can be obtained by adding the four channels from the Mott detector. In the intensity images the samples show no contrast except for a few "spots," as the one shown in the image in Fig. 1. These spots were used to position the sample reproducibly. All images shown are 256 × 256 pixels and about 85 μm × 70 μm (due to the off-normal alignment of gun and sample). The primary electron beam energy is 5 keV and the current is a few nA. The lateral resolution is about 500 nm. Count rates in the Mott detector are typically 5×10^4 counts/sec per channel. The Fe films can be magnetized by a field pulse of 50 Oe in the x direction, which coincides with an easy axis. The Fe films show a high secondary electron polarization of about 40%. The sample stage can be cooled to about $-100\,°C$ and heated to about $80\,°C$, which is well above the Cr Néel temperature.

Figure 2 contains the principal observations of this paper. It shows the two in-plane polarization components starting in the top row with a one-domain state at RT after the film was magnetized above T_N. There is a uniform high x-polarization (left column) except for a spot with zero polarization in the center of the image. Upon cooling, a domain pattern as shown in the second row
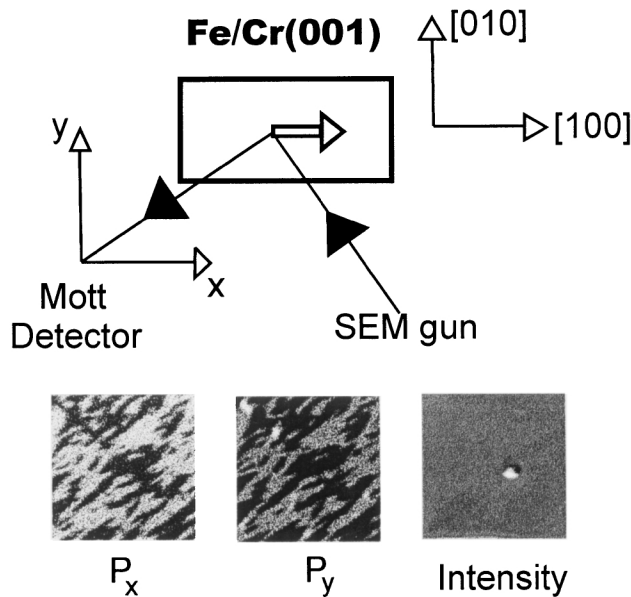
FIG. 1.   Geometry of the experiment and sample orientation with respect to the polarization measurement. The two in-plane components (x and y) of the magnetization are measured. An external magnetic field can be applied in the x direction.

(LT) evolves which was taken at the final temperature of $-100\,°C$. This domain pattern with the emergence of a perpendicular component (P_y) and a corresponding reduction in P_x is, of course, a microscopic picture of the observed magnetization reduction reported in Ref. [5]. A pixel-by-pixel analysis shows that the absolute polarization values ($\sqrt{P_x^2 + P_y^2}$) stay constant at about 40%. This proves that the magnetization stays in plane (no "missing" out-of-plane component) and only *rotates* in plane as a function of temperature while the *local* magnetization value is temperature independent. It also proves that there are no significant magnetic structures smaller than the beam diameter (500 nm) since averaging over those structures would lead to a reduction of the polarization values. The magnetization structures are quite irregular and range from a few μm to some tens of μm. When the film warms to above the Néel temperature the domain pattern changes to the one shown in the third row (RT). The images become sharper. The domains boundaries become smoother and most of the small-scale structures have coalesced into bigger domains. The change between the LT and RT state is reversible. When cooled again the domain images in the lower row of Fig. 2 result in almost all detail identical to the second row.

A survey of several spots on the sample showed that the magnetization behavior described above is indeed typical. A quantitative evaluation of the images reveals more details about the magnetization state. From the measured P_x and P_y values one can determine the local magnetization angle for every pixel. Figure 3 shows the distribution of angles with respect to the macroscopic magnetization di-
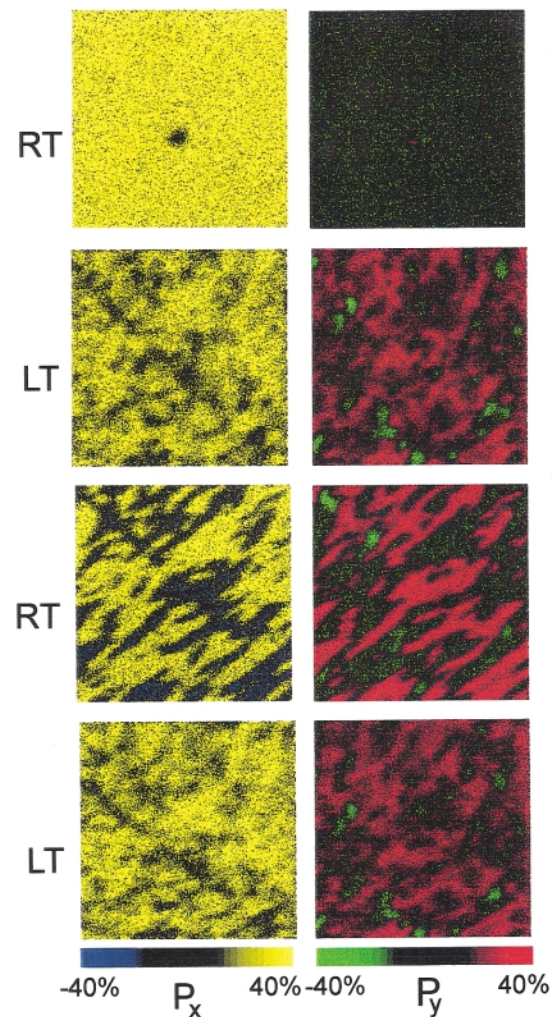

FIG. 2 (color).   The evolution of the magnetic domain state as the temperature is changed. The images show on the left (yellow-blue contrast) the P_x and on the right the P_y component (red-green). All images are taken on the same spot, except for a small thermal drift of the sample stage. The images are 85 μm $\times$ 70 μm. Top row: One-domain state at RT; second row: cooled down to low temperature ($-100\,°C$). Third row: After warming above T_N; fourth row: cooled down again to LT.

rection. A four-pixel average was applied in order to improve statistics. The single-domain state shows a fully aligned magnetization. The width of the distribution is solely due to counting statistics. The LT state, on the other hand, shows a broad asymmetric distribution of angles with a maximum around 25° and the average at about 45°. After warming to above T_N the maxima at 0° and at 90° correspond to easy axes of the bcc Fe films. Thus, above T_N the magnetization direction is governed by crystalline anisotropies. By comparing the RT and LT images in Fig. 2 it is clear that the areas that have a strong perpendicular component at LT are most likely to "flip over" into the 90° direction upon warming to RT.

Not only is the temperature cycling between LT and RT reversible but even the domain patterns that result after
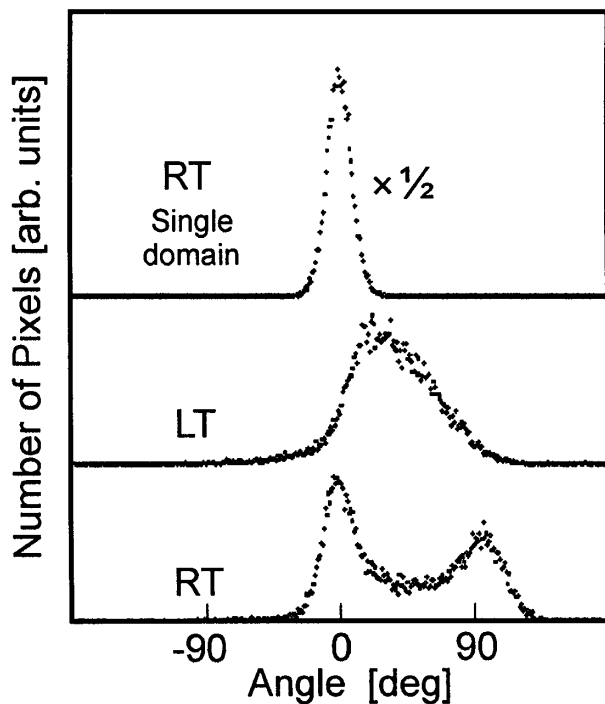
FIG. 3. Histogram of the angular distribution of the local magnetization with respect to the axis of the macroscopic magnetization. The images from Fig. 2 were used after a four-pixel average to improve statistics.

remagnetization (above T_N) and cooling again are highly reproducible. Reversing the magnetization direction, as well as rotating the sample 90° and magnetizing in that direction, shows that it is always the same areas that *turn* away from the macroscopic magnetization direction, independent of the initial magnetization direction.

What drives the magnetization reorientation? It seems unlikely that the images reflect some aspect of the magnetic structure of the Cr, e.g., an antiferromagnetic domain structure, because of the reproducibility of the structures even after heating above the Cr ordering temperature. It can also be safely excluded that the domains reflect the atomic terrace structure of the Cr substrate. Atomic terrace widths on well-prepared single crystal metal surfaces are typically not more than a few hundred Å wide [13]. Thus, the magnetic structures are a hundred times larger than the terraces. The mechanism that then springs to mind is a roughness driven transition due to frustration, very similar to the mechanism that leads to biquadratic coupling between FM layers due to interface roughness. Since the Fe magnetization cannot follow the Cr terrace structure, because it would cost too much energy in domain walls, it tends to turn perpendicular to the Cr surface moments. The Cr surface magnetization was induced by the single-domain Fe film and was frozen in when the sample was cooled through T_N. Thus the images show the biquadratic coupling already at the single AF/FM interface. Evidence for perpendicular spin orientations has been reported re-

cently in very different systems: Fe_3O_4/CoO superlattices [14] and Fe films on FeF_2 [15].

In order to demonstrate the roughness/magnetization connection a little better, Fig. 4 shows the polarization and the corresponding intensity image (digitally enhanced) taken very close to the sample edge, where one can actually see scratches in the SEM image as well as under an optical microscope. A correlation between magnetization and topography is obvious in a very general, qualitative way. On this "rough terrain" hardly any horizontal (P_x) magnetization is left over, i.e., everything has turned almost by a full ±90° with respect to the original magnetization direction. Note that it is not the direction of the scratches that determines the magnetization direction but that the magnetization is perpendicular to the original magnetization direction, i.e., dominated by the Cr interface moments. Also, during cooling the rough regions start switching sooner than the flat areas consistent with a roughness driven mechanism. Some slight preferential orientation of the roughness, e.g., step edges, probably is also the cause of the preferential angle in which the magnetization turns at low temperature. In Fig. 2 almost everything turns towards +90° and very little towards −90°. This preference seems to vary locally over the sample. Finally, because of the similarity of the images I want to mention a study of the exchange coupling in Co/Cr/Co trilayers by TEM during magnetization reversal [16]. It is shown that some areas start switching consistently sooner than others. This is attributed to a spatially varying exchange coupling due to thickness fluctuations.
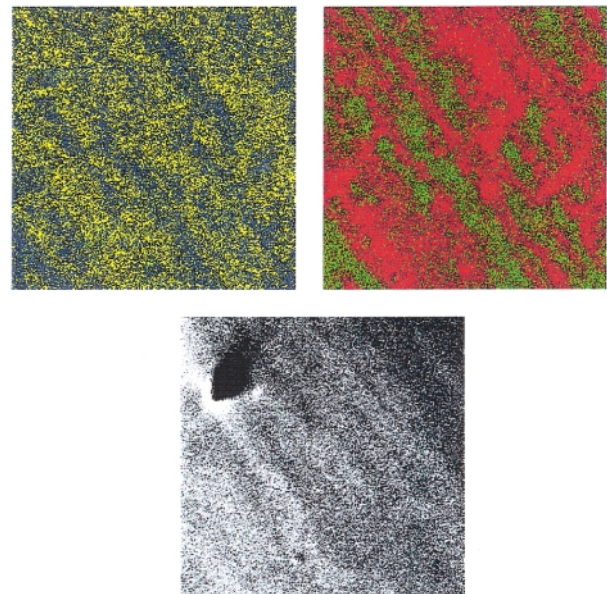

FIG. 4 (color). Domain (P_x and P_y) image in the top row taken at RT (same parameters as in Fig. 2); below: SEM intensity image derived from the Mott detector counts, with the contrast greatly enhanced. This image was obtained on a rough area very close to the sample edge, showing the general correlation between roughness and magnetic structure.

In summary, it was shown that upon cooling one-domain 2-nm Fe films on nominally flat Cr(100) surfaces form a nonuniform magnetization state with a spatially varying 90° (in-plane) magnetization component. The domain evolution is partially reversible and highly reproducible. It is suggested that atomic scale roughness is the driving force for the magnetization reorientation, and that the magnetization direction reflects the average local step density. When warmed to above the Cr Néel temperature the Fe magnetization relaxes into the easy magnetization directions. Evidence for the correlation between roughness and magnetization direction was presented. The origin of the 90° alignment is suggested to be frustration due to interface roughness similar to the mechanism in biquadratric coupling in FM/AF/FM structures.

[1] W. H. Meiklejohn and C. P. Bean, Phys. Rev. **102**, 1413 (1956).

[2] J. Unguris, R. J. Celotta, and D. T. Pierce, Phys. Rev. Lett. **67**, 140 (1991).

[3] E. Fawcett, Rev. Mod. Phys. **60**, 209 (1988).

[4] E. E. Fullerton, S. D. Bader, and J. L. Robertson, Phys. Rev. Lett. **77**, 1382 (1996); P. Bodeker, A. Hucht, A. Schreyer, J. Borchers, F. Guthoff, and H. Zabel, Phys. Rev. Lett. **81**, 914 (1998); S. Demuynck, J. Meersschaut, J. Dekoster, B. Swinnen, R. Moons, A. Vantomme, S. Cottenier, and M. Rots, Phys. Rev. Lett. **81**, 2562 (1998).

[5] A. Berger and H. Hopster, Phys. Rev. Lett. **73**, 193 (1994).

[6] J. C. Slonczewski, Phys. Rev. Lett. **67**, 3172 (1991); N. C. Koon, Phys. Rev. Lett. **78**, 4865 (1997).

[7] M. Rührig, R. Schäfer, A. Hubert, R. Mosler, J. A. Wolf, S. Demokritov, and P. Grünberg, Phys. Status Solidi A **125**, 635 (1991); M. E. Filipkowski, J. J. Krebs, G. A. Prinz, and C. J. Gutierrez, Phys. Rev. Lett. **75**, 1847 (1995); A. Azevedo, C. Chessman, S. M. Rezende, F. M. de Aguiar, X. Bian, and S. S. P. Parkin, Phys. Rev. Lett. **76**, 4837 (1996); J. Schwabenhausen, T. Durkop, and H.-J. Elmers, Phys. Rev. B **15**, 15 119 (1997).

[8] E. J. Escorcia-Aparicio, H. J. Choi, W. L. Ling, R. K. Kawakami, and Z. Q. Qiu, Phys. Rev. Lett. **81**, 2144 (1998).

[9] H. Hopster, Y. Iwasaki, J. Barthel, E. B. Maiken, B. P. Miller, J. Kondis, and Y. Yu, Mater. Res. Soc. Symp. Proc. **475**, 569 (1997).

[10] The Cr(100) single crystal sample was commercially oriented and polished to within 0.1° (0.25° guaranteed) by Mateck, Jülich.

[11] FEI Company, Hillsboro, Oregon.

[12] D. P. Pappas, Ph.D. thesis, University of California, Irvine, 1990.

[13] At present there is no direct information available on the atomic terrace widths on this sample. The smoothest metal substrates available seem to be whiskers (see, e.g., Ref. [2]). Here a terrace width can reach the micron scale. STM experiments will be combined with SEMPA in the future.

[14] Y. Ijiri, J. A. Borchers, R. W. Erwin, S.-H. Lee, P. J. van der Zaag, and R. M. Wolf, Phys. Rev. Lett. **80**, 608 (1998).

[15] T. J. Moran, J. Nogues, D. Lederman, and I. K. Schuller, Appl. Phys. Lett. **72**, 617 (1998).

[16] A. C. Daykin, J. P. Jakubovics, and A. K. Petford-Long, J. Appl. Phys. **82**, 2447 (1997).