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Domain images of ultrathin Fe films on Ag(100)

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Scanning electron microscopy with electron polarization analysis has been used to image domains of ultrathin Fe films grown epitaxially on a Ag(100) substrate. Room-temperature measurements show clearly the existence of large domains of in-plane magnetization for film thicknesses of 3.4 monolayers or more. No in-plane domains were observed for thinner films.

Owing to recent advances in theoretical and computational techniques,[1] epitaxial growth methods,[2] and methods of surface magnetization measurement,[3] the study of ultrathin magnetic films is enjoying very rapid growth. An immediate objective of these endeavors is to understand the physical principles at work in the first few monolayers of a magnetic material epitaxially grown on a noble metal single-crystal surface. This letter demonstrates that scanning electron microscopy with polarization analysis can be used to observe the magnetic characteristics of such ultrathin films. Specifically, this technique allows the direct observation of the change in domain structure of epitaxial α-Fe(100) films on Ag(100) that takes place at an Fe thickness of approximately 3 monolayers.

In 1986 Jonker *et al.*[4] studied the Fe/Ag(100) system using spin polarized photoemission. They saw the in-plane component of polarization vanish for Fe thicknesses below 3 monolayers (ML) in their room-temperature measurements. Possible explanations for this loss of observable magnetization included a perpendicular anisotropy which caused a perpendicular magnetization not measurable in their experiment, the formation of small domains, or a reduced Curie temperature for the thinner Fe films. Gay and Richter[5] applied band structure theory to the problem of a free-standing monolayer of Fe and concluded that the anisotropy normal to the surface was of sufficient strength to overcome the demagnetization field, which normally forces the magnetization to lie in the surface plane. Subsequent calculations[6] for a monolayer (ML) of Fe on Ag(100) were unable to specify a favored direction of magnetization.

Heinrich *et al.*[7] used ferromagnetic resonance to study Fe(100) films grown on Ag(100) in thicknesses of 28, 17, 5, and 3 ML. Each Fe film was covered by Au(100). They extrapolate their results to 2 ML and concluded that at 2 ML or less the magnetization should be perpendicular to the surface.

Koon *et al.*[8] utilized the conversion-electron Mössbauer technique to study the Fe/Ag(100) system at temperatures down to 15 K. To achieve the necessary sensitivity, a superlattice was grown with from 7 to 45 periods of Fe layers with thicknesses of 1 to 5.5 ML separated by Ag layers of 4 to 7 ML. They concluded that at 15 K the zero field magnetization was orientated out of the plane for both the 1 and 2.4 ML films. At 5.5 ML thickness, they found the magnetization to lie in-plane at room temperature and to have an out-of-plane component at low temperatures.

Finally, Stampanoni *et al.*[9] used threshold spin polarized photoemission to measure the perpendicular component of magnetization of the Fe/Ag(100) system over a range of temperatures and applied magnetic fields. At $T = 30$ K, they found remnant perpendicular magnetization only for overlayers in the thickness range of 3.5 to 5 ML. Above 100 K, they conclude from their data that the magnetization lies in-plane. They also show that the Curie temperature for bcc Fe films is approximately 400 K for a 1 ML and rises linearly to the bulk value at 5 ML thickness.

Scanning electron microcopy with polarization analysis (SEMPA) offers an alternative way of observing the surface magnetization of ultrathin films. In this technique,[10] the polarization of the secondary electrons produced at the sample surface in a scanning electron microscope is measured by transporting them to a detector that measures both their spin polarization and flux. The spin polarization determination

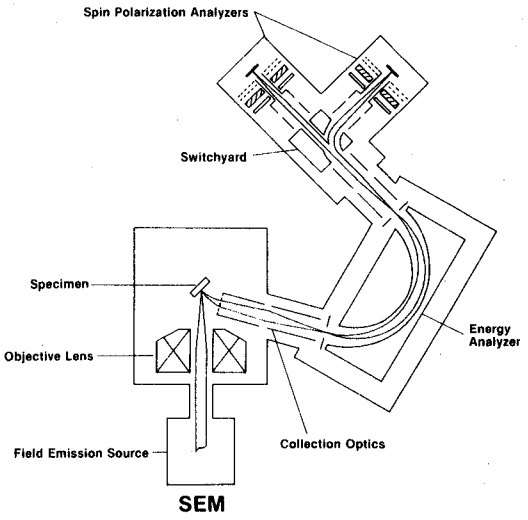

FIG. 1. Schematic representation of the apparatus showing the field emission microscope, the extraction optics, optional energy analyzer, and polarization detectors.

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provides a direct measurement of the magnetization vector of the sample area under the focused incident electron beam. The apparatus is illustrated schematically in Fig. 1. The polarization detector positioned at the end of the electron optical path simultaneously determines the two orthogonal components of the magnetization lying in the plane of the sample. For in-plane magnetization, these provide complementary gray map images of the domain structure of the sample.

The thin-film surfaces were prepared *in situ* in an ultrahigh-vacuum scanning electron microscope with a base pressure of 5×10^{-10} Torr. The preparation procedure consisted of mild sputter cleaning of a Ag(100) single-crystal substrate, followed by annealing to 300 °C for several minutes. Reflection high-energy electron diffraction was used to continuously monitor the long range order of the sample surface during cleaning and deposition. The Fe was deposited using electron bombardment heating of a pure Fe wire and was monitored with a quartz crystal thickness monitor. The deposition rate was determined before and after the experiment by x-ray fluorescence measurements of Fe deposited under the same conditions on GaAs substrates. The fluorescence measurements were made using a calibrated reference standard. α-Fe(100) can be grown on Ag(100) because the bcc Fe lattice, when rotated 45°, is commensurate[11] with the Ag lattice within 0.8%. The growth mechanism is thought to be layer by layer for the first three layers, with island growth thereafter.[4,12]

Figure 2(a) is a polarization image for an Fe film 3.4 ML thickness. The gray scale represents the amplitude of the component of magnetization in one direction along the sample surface. The areas of uniform intensity represent domains. Figure 2(b) shows the intensity topography from the same region. The total difference in the polarization between the two major domains shown in Fig. 2(a) is approximately 40%. The dimensions of this image are approximately 150×200 μm. Most of the film consists of a single domain except for a 1-mm-wide strip along the sample edge where multidomain structures, such as this one, are present. Measurements made in the same region at a layer thickness of 2.6 ML or less showed no variation in either in-plane or perpendicular polarizations characteristic of domain formation.

For some of the Fe depositions we placed a mesh within 2 mm of the sample surface. In this manner we produced a regular array of 310×310 μm patches of thin Fe films on a background of Ag. It was possible to image these patches of Fe using SEM techniques alone,[13] even at thicknesses below 1 ML. Figure 3 is a polarization image which shows some of the Fe islands on the Ag crystal background. The Fe film thickness was 7.7 ML for this image and the polarization was in-plane. This image is the result of magnetizing the Fe by applying a pulsed magnetic field in two opposite directions and subtracting the resulting polarization images. This verifies that the polarization measurements are determining the surface magnetization and rules out any spurious effects. Figure 3 therefore shows the change in the magnetization on field reversal. The total change in polarization, as depicted in the superimposed line scan, is approximately 55%. This is in rough agreement with, though somewhat higher than, the

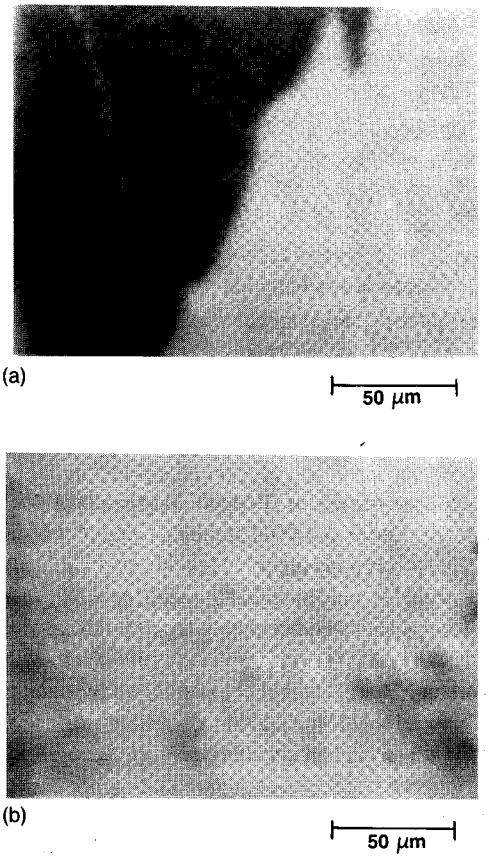

FIG. 2. (a) Polarization image with intensity representing the magnitude of the in-plane polarization of a 3.4 ML Fe film. Two large domains are seen. (b) Simultaneously obtained normal SEM image showing a featureless area approximately 150×200 μm in size.

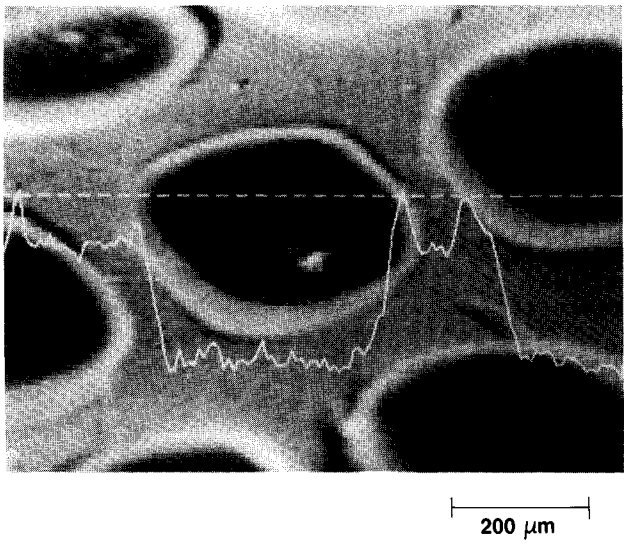

FIG. 3. Image represents the change in polarization on reversal of an external, pulsed magnetic field. The light background is the nonmagnetic Ag substrate. The dark regions are ferromagnetic Fe spots whose magnetization was reversed by the magnetic field pulse. The line scan shows the change in magnetization sampled along the dashed line.

maximum changes in polarization we have observed in bulk Fe-3% Si crystals. The light background comes from the nonmagnetic Ag, and the light rings surrounding the Fe patches are not yet fully understood.

These measurements demonstrate that, in the room-temperature Fe/Ag(100) system, in-plane magnetic domains can be seen to exist at thicknesses down to about 3.4 ML, but are not visible under identical conditions at 2 ML thickness even though the SEMPA technique has sufficient sensitivity to observe them. Our measurements agree with the results of Jonker *et al.*; the in-plane magnetization of uncoated, room-temperature Fe films vanishes below 3.4 ML. Also, the domains are seen to be large at 3.4 ML and it is unlikely that unresolved domain structure at lower Fe thickness can account for either result. The SEMPA technique is shown to be capable of studying the magnetization of films at the few monolayer level, which should be an important asset to the study of the new magnetic systems mentioned earlier.

Our future work will include studies of this and similar systems at low temperatures, with special emphasis on the perpendicular component.

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