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Thickness- and temperature-dependent spin anisotropy of ultrathin epitaxial Fe films on Ag(100)

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Magneto-optical techniques have been used to study the thickness- and temperature-dependent spin anisotropy of ultrathin epitaxial Fe films grown on Ag(100) substrates. Films having a thickness of one and two monolayers exhibit preferred spin orientation perpendicular to the surface and a temperature-dependent coercive force. Thicker films exhibit in-plane spin orientation, layer-dependent coercive forces, and evidence of layer-dependent in-plane spin anisotropy.

Recent experimental[1-4] and theoretical[5-7] studies of ultrathin epitaxial magnetic films have begun to explore the striking departure of their magnetic properties from those of bulk materials. Novel properties of films having thickness of a few monolayers (ML) include layer-dependent enhanced magnetic moments,[6] thickness-dependent spin-anisotropy,[1-5] linear temperature dependence of the magnetization,[8] and Curie temperatures that are significantly different from those for bulk materials.[8,9] These properties, which are intrinsic to ultrathin films, result from the loss of translational symmetry perpendicular to the surface (reduced dimensionality), reduced coordination associated with surface atoms, and subtle changes in the spin-dependent terms[5] that contribute to the total energy such as spin-orbit effects and the surface-to-volume ratio of the thin-film structure.

In the present work, we have explored the thickness and temperature-dependent magnetic properties of ultrathin epitaxial Fe films on Ag(100) surfaces using magneto-optical techniques. Our results yield unambiguous verification of the predicted perpendicular spin anisotropy[5] of single monolayer films, in agreement with conversion-electron Mössbauer experiments[3] conducted on epitaxial multilayer films, and spin-polarized photoemission experiments[2] and ferromagnetic resonance studies[4] conducted on epitaxial Fe films on Ag(100). Our experiments, in addition, reveal the layer dependence of the in-plane magnetic anisotropy, and characterize the temperature dependence of the easy axis rotation.

In ferromagnetic materials, magneto-optical effects are dominated by spin-orbit effects which couple the electron-spin moment to the spatial components of the wave function.[10] Optical dipole transitions are affected by the spin direction which produces a first-order (linear) magnetic contribution to the off-diagonal component of the optical conductivity tensor. This property, known as the magneto-optic Kerr effect, is manifested in the reflection of polarized light from a magnetic material as a small (10^{-5} rad) magnetization-dependent rotation of the polarization axis and an accompanying optical phase shift (ellipticity). Previous experiments have established the utility of the Kerr effect as a probe of magnetic properties of bulk[11] and thin-film[12] magnetic materials.

Polar and longitudinal magneto-optic Kerr effect signals are proportional to the spin polarization projected onto the propagation vector of light in the magnetic material and averaged over the optical penetration depth (typically 200 Å for metals). Optical interference effects[13] are negligible in cases where the film thickness ΔX is small compared to the wavelength of light in the film (also typically 200 Å for visible range light). Under these conditions (clearly valid for our experiments) Kerr effect signals will scale linearly with thickness of ultrathin films provided there are no layer dependencies in the spin polarization. In this regime ($\Delta X \ll$ optical penetration depth), the Kerr effect is quantitatively different from the classical magneto-optic Kerr effect (which assumes $\Delta X \gg$ optical penetration depth), and a new name for the effect appears to be justified: the surface magneto-optic Kerr effect[12] (SMOKE). Dramatic layer-dependent departures from linear thickness-dependent behavior of SMOKE signals from ultrathin films can provide a probe of layer-dependent changes in magnetic properties.

Magnetic anisotropy of a specific thin-film structure can also be probed by the Kerr effect. Kerr rotations and ellipticities are proportional to the average direction of magnetization induced by an applied field, and can, therefore, be used to determine a hysteresis curve of a thin magnetic film. The properties of hysteresis curves as a function of applied field strength and direction in relation to the film surface and crystallographic directions establish easy and hard directions of magnetization.

Our epitaxial films were prepared and studied *in situ* under ultrahigh vacuum conditions (1×10^{-10} Torr). Conventional techniques were used to cut and align the 1-cm-diam. Ag(100) crystals, and to clean the surface (Ne sputtering at 500 V followed by annealing). Epitaxial layers were grown by electron beam evaporation from the tip of a high-purity Fe wire. The Ag(100) crystal was held at temperatures ranging from 300 to 350 K during film growth. This range of temperatures yields good epitaxial layers without causing diffusion of deposited atoms into the substrate. Crystal order and chemical purity of the Ag(100) surface and the epitaxial films were monitored by low-energy electron diffraction (LEED) and Auger analysis. Film growth (typically at 0.25 ML/min) was monitored in real time by Auger spectroscopy and by a calibrated quartz thickness monitor near the source. A sharp break in the slope of Auger intensities versus thickness observed during growth of our films supports previous

work[1,14] that has established that Fe initially grows layer-by-layer on Ag(100). Island formation[15] apparently begins to occur for films greater than 4–5 ML, although our LEED patterns did not suggest departure from good epitaxy. We also observed the reported broadening of LEED spots between 2 and 4 ML reported by Stampanoni, Vaterlans, Aeschlimann, and Meier.[2]

Figure 1 displays hysteresis curves for epitaxial films of Fe on Ag(100) using the longitudinal configuration in which the applied magnetic field is in the plane of the film and parallel to the plane of incidence. Our SMOKE data were taken using a He-Ne laser ($\lambda = 632.8$ nm). The *s*-polarized light was incident at 40° measured from the film normal direction. Two orientations of the crystal were investigated, one in which the [11] direction was parallel to the applied field H and one in which the [10] direction was parallel to H.

Figure 2 displays corresponding hysteresis curves for the polar configuration in which H is applied along the crystal's normal direction. Polar configuration measurements were made using the same angle of incidence, 40° from the film's normal direction, and with the plane of incidence along a [01] crystal axis. A rotatable magnet permitted the same film to be studied in both polar and longitudinal configurations.

The hysteresis curve obtained in polar geometry for a 1-ML film proves that a single layer film is ferromagnetic. The coercive force of the film at room temperature is small (suggesting that multiple domains are present for zero applied field) and temperature dependent. Applied fields of several hundred oersted are required to saturate the film (form a single domain).

No Kerr-effect signals were detected for H parallel to the surface for films less than 2-ML thick with applied fields ~ 1.2 kOe and temperature above 100 K. Polar Kerr-effect signals were observed for single- and double-layer films at 300 K. These features are consistent with preferred spin alignment perpendicular to the surface for $\Delta X < 2$ ML and parallel to the surface for $\Delta X > 2$ ML.

Figure 3 displays reduced data consisting of SMOKE amplitude (at saturation) as a function of film thickness (in ML) for H along the [01] and [11] crystal directions. Both plots exhibit the loss in signal for films below 2 ML

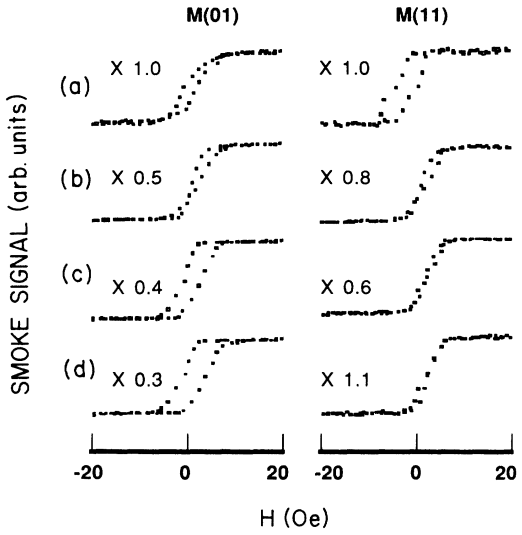

FIG. 1. Hysteresis curves obtained using the SMOKE longitudinal configuration for epitaxial bcc Fe films on Ag(100). Vertical axes of the curves have been scaled by the amount indicated. The temperature is 300 K. The number of layers is (a) 2.5, (b) 4.5, (c) 6.5, and (d) 8.5.

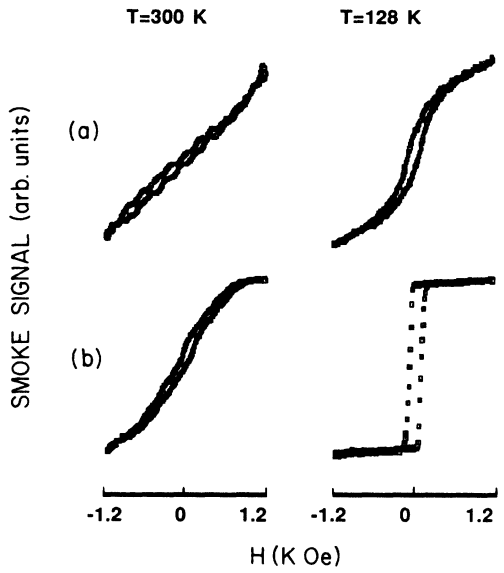

FIG. 2. Temperature-dependent hysteresis curves for bcc Fe/Ag(100) obtained using the SMOKE polar configuration $M(100)$. The number of layers is 1.0 for (a) and 2.0 for (b).

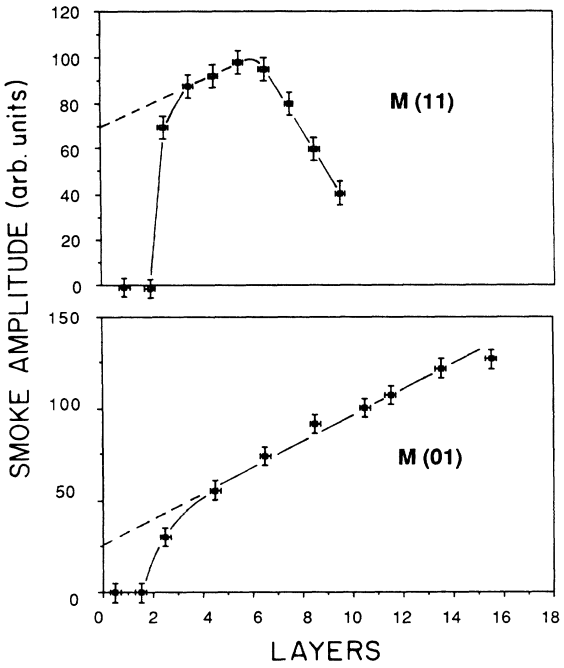

FIG. 3. Layer dependence of SMOKE amplitude for longitudinal configuration (H parallel to the surface) for applied fields along [01] and [11] crystal axes. Sample temperature was 300 K.

attributed to perpendicular anisotropy. Both curves exhibit a linear layer dependence of film thickness greater than approximately 3 ML which extrapolates to a nonzero value for zero film thickness. The linear dependence of Kerr signal as a function of film thickness for H along the [01] direction supports our previous statement that for uniform magnetization and $\Delta X \ll \lambda_{\text{metal}}$ ($\lambda_{\text{metal}} \sim 200$ Å) the Kerr effect will be proportional to thickness.

The nonzero extrapolated Kerr signal for $\Delta X = 0$ is an interesting feature of our data. Various consistency tests were conducted to assure the accuracy of the results. These checks involved split-beam experiments in which half of the crystal was first masked to maintain a clean surface reference for film studies of various values of ΔX. We also conducted additional experiments in which the clean half of the crystal was subsequently covered so that two film thickness could be studied simultaneously. All of the results were consistent. The extrapolated nonzero Kerr signal could be attributed to enhanced surface moments associated with atoms in contact with the substrate and/or atoms at the surface. This interpretation, however, requires enhanced surface moments considerably larger (factor of 10) than predicted (40%) enhancements which is unreasonable. Precise understanding of this effect will probably require a more detailed macroscopic model of the Kerr-effect reflection process than is presently available.

Independent of the other properties just discussed (departure from linear thickness dependencies for $\Delta X < 3$ ML, nonzero extrapolated signal at $\Delta X = 0$) the simple fact that the two panels of Fig. 3 are not identical must be regarded as an indication of a layer-dependent in-plane anisotropy. Previous experimental evidence for layer-dependent in-plane magnetic surface anisotropy appears to be limited to a few examples that involve only twofold in-plane symmetry: Fe(110) grown on W(110),[16,17] or on GaAs (Ref. 18). In these cases, a spontaneous switching of M from a [001] to a [110] direction with decreasing film thickness was observed at $\Delta X \sim 30$ (80) layers of Fe on GaAs [W(110)]. Referring to Fig. 1, it is clear that the coercive force of H along the [11] direction becomes very small, and the SMOKE amplitude (at saturation) exhibits a dramatic decrease starting at $\Delta X = 6$ ML. Similar effects are not observed for H along the [01] direction, and since at $\Delta X = 6$ ML, the shape anisotropy prohibits spin alignment perpendicular to the surface, it it reasonable to assume that the behavior illustrated in Fig. 3 is an indication that spins prefer alignment along the [01] direction. This behavior can be attributed to a layer-dependent in-plane anisotropy characterized by a very small anisotropy energy up to about six layers. The absence of large in-plane anisotropy at room temperature for 3-ML films has been observed by ferromagnetic-resonance studies.[4] However, these studies require the Fe films to be protected with Ag or Au coatings that can influence the anisotropic character of the ultrathin films[17] and also assume a constant (bulk) value for the saturation magnetization in order to calculate the anisotropy constant.

Our temperature-dependent studies of the SMOKE amplitude are summarized in Table I. For very thin films ($\Delta X < 4$ ML), we observe a linear dependence (within experimental error) of the SMOKE amplitude with temperature, and thickness-dependent extrapolated Curie temperatures. Both of these features establish the essential two dimensionality of our epitaxial films. These results are in reasonably good agreement with corresponding spin-polarized photoemission measurements reported by Stampanoni *et al.*[2] for the same system. However, our monolayer films at low temperature (128 K) and double-layer films at room and low temperature, exhibit sizable coercive forces that contradict their results above 110 K. The lack of coercive force was attributed to a possible in-plane anisotropy which is again inconsistent with our measurements. The agreement of SMOKE results and spin-polarized photoemission results for films having ΔX greater than 2–3 layers is probably accidental because of the difference of probe depth associated with the two

TABLE I. Linearly extrapolated Curie temperature as a function of thickness for epitaxial Fe on Ag(100). Extrapolation is based on saturated magnetization ($H \gg 20$ Oe refer to Fig. 1).

Layers	T_c (K)
2.4	475 ± 50
2.5	775 ± 50
9.5	1160 ± 200

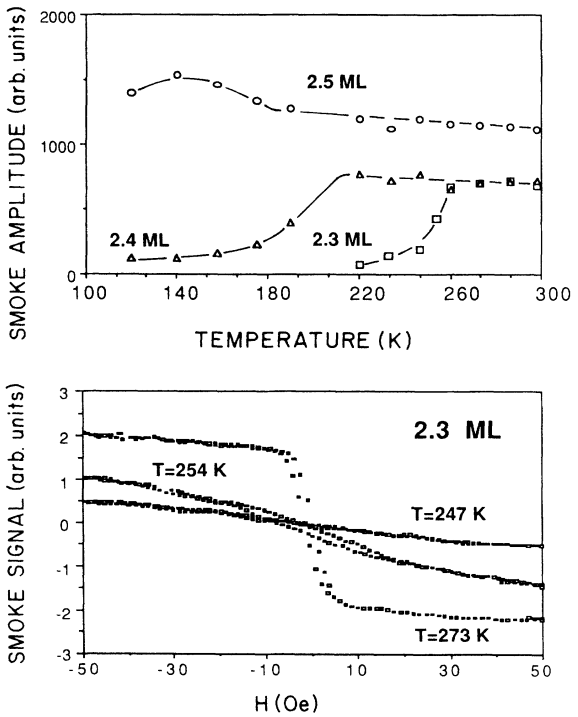

FIG. 4. Temperature dependence of SMOKE signals for 2.4 ± 0.1 ML films. Note the dramatic changes that occur at approximately 220 K. Lower panel, hysteresis curves for a 2.3-ML film for three temperatures above 220 K. (Longitudinal configuration, H along [01].)

methods (photoemission being much more sensitive to the top few layers).

An interesting temperature-dependent and strongly layer-dependent behavior is observed for films of 2.4 ± 0.3 ML. The SMOKE amplitude (as shown in the upper panel of Fig. 4) exhibits a linear dependence with temperature (again consistent with the low dimensionality of the films) but at a layer-dependent transition temperature the amplitude decreases for the 2.3- and 2.4-ML films. The departure of linear behavior is in this case consistent with temperature and layer dependencies of the perpendicular anistropy constant that accompany the rotation of the easy magnetization direction from a direction perpendicular to the surface ($\Delta X < 2.4$ ML) to an in-plane direction ($\Delta X > 2.4$ ML). As it is illustrated in the lower panel of Fig. 4, there is a dramatic change in the shape of the hysteresis curve as a function of temperature that suggests an increase in the perpendicular anisotropy energy as the temperature decreases.

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