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Reduction of macroscopic moment in ultrathin Fe films as the magnetic orientation changes

D. P. Pappas and C. R. Brundle
IBM Almaden Research Center, San Jose, California 95120

H. Hopster
Department of Physics and Institute for Surface and Interface Science, University of California, Irvine, California 92717

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A study of the remanent magnetization $\mathbf{M}_r$ in ultrathin Fe films grown at 125 K on Cu(100) and Ag(100) substrates was conducted using spin-polarized secondary-electron spectroscopy. For bcc-Fe/Ag(100) and fcc-Fe/Cu(100) films, perpendicular $\mathbf{M}_r$ was observed for film thickness less than 7 and 11 Å, respectively. In-plane $\mathbf{M}_r$ was observed for thicker films. For thickness below the critical thickness, both systems showed a reversible switching transition of the magnetization orientation from perpendicular to in plane as the temperature was increased. For the Fe/Cu(100) films the switching could be observed for many temperature cycles providing that the film was not annealed above 350 K, while the Fe/Ag(100) films switching was stable for only 1–2 annealing cycles to 300 K. A loss of $\mathbf{M}_r$ during the switching transition is observed, and is attributed to a change in the magnetic nature of the film. The details of the loss of magnetization observed in Fe/Ag(100) and Fe/Cu(100) are significantly different.

The magnetic properties of ultrathin transition metal films have revealed a variety of useful and interesting effects. In particular, the magnetic easy axis and domain structure is of vital interest when considering thin films as storage media. Remanent magnetization normal to the plane of the film is desirable, e.g., for magneto-optic recording media, while the intrinsic tendency of a film to form magnetic domains limits the data bit size and signal-to-noise ratio when reading the information. Néel[1] first postulated the existence of a perpendicular anisotropy at an interface, and it has been shown that in many thin-film systems the perpendicular anisotropy can overcome the magnetic dipole energy of the film.[2] The switching transition between perpendicular and in-plane $\mathbf{M}_r$ as the film thickness and temperature is increased has been studied theoretically by Jensen and Bennemann[3] using entropy considerations, and also by Pescia and Pokrovsky[4] in a renormalization-group theory approach. Both theories predict that there is a thickness and temperature range where the remanent magnetization, $\mathbf{M}_r$, switches its orientation from perpendicular to in plane. However, the nature of $\mathbf{M}_r$ near the switching transition cannot be predicted by these theories since the free energy in this region is essentially constant as a function of angle. On the other hand, it has been suggested that a multidomain configuration is energetically favorable over a single-domain state when the perpendicular anisotropy and dipole energies are nearly equal,[5] which can be expected near the switching region. Experimentally, a multidomain ground state has been directly observed in thin films of Co films using scanning electron microscopy with polarization analysis[6,7] (SEMPA) and spin-polarized low-energy electron microscopy (SPLEEM).[8] The breakup of $\mathbf{M}_r$ into small domains was also correlated with the switching from perpendicular to in-plane magnetization as the thickness was increased.[6]

Magnetism in a wide variety of well-characterized thin-film systems has by now been studied.[2] For Fe, in particular, the direction of $\mathbf{M}_r$ has been shown to depend sensitively on film thickness, temperature, contamination level, and crystallographic orientation.[9–13] Fe is also interesting because it can be stabilized in either the low temperature, ferromagnetic bcc phase on a Ag(100) substrate,[14,15] or in a metastable, fcc structure on Cu(100) in which the magnetic state is extremely sensitive to small changes of the lattice constant.[15–18] The structural and magnetic properties of thin Fe overlayers (1–20 Å) on these substrates have been a subject of controversy.[10,19–23] For the most part, both the Fe/Ag(100) and Fe/Cu(100) systems are reported to be ferromagnetic when grown at low temperature,[9–11] and undergo a switching transition (at that temperature) of $\mathbf{M}_r$ from perpendicular to in plane as the film thickness is increased. A similar transition was reported in the temperature dependence of $\mathbf{M}_r$ (at a given thickness) for Fe/Ag(100),[10] with evidence of $\mathbf{M}_r$ perpendicular to the film

at 30 K and n plane at room temperature. In Fe/Cu(100), the transition was found to be reversible with temperature cycling.[11] The transition temperature was observed to decrease with increasing film thickness, which is in line with the assumption that the in-plane dipole energy of the film is increasing with increasing thickness while the perpendicular surface anisotropy is constant. While there are similarities in the magnetic behavior of Fe grown on Ag(100) and Cu(100) substrates, there is considerable disagreement in the published literature of the exact thickness and temperature dependence of $\mathbf{M}_r$ in these systems.

In general, it is difficult to compare results taken under different growth conditions and with different magnetization characterization techniques. The relative film quality is also a primary concern, since it has been shown, e.g., that surface contamination ($\simeq$0.5 langmuir O_2) induces a rotation of $\mathbf{M}_r$ from perpendicular to in plane.[12] In this paper, a study of magnetism in Fe films grown on Ag(100) and Cu(100) substrates under identical conditions is presented. Spin-resolved secondary-electron emission spectroscopy (SPSEES) was used due to its sensitivity to surface magnetization, and all three components of polarization were measured to determine the orientation of $\mathbf{M}_r$. We observe differences in the reversibility of the magnetic properties of the films with temperature cycling, and also in the thickness and temperature dependence of the orientation of $\mathbf{M}_r$ with respect to the plane of the film. In both systems a decrease of the macroscopic magnetic moment is observed near the switching transition in both the thickness and temperature-dependent measurements.

The samples were grown in UHV (10^{-10} Torr) conditions at $T\simeq125$ K and then annealed to room temperature.[24] The film quality was determined using low-energy electron diffraction (LEED) and Auger electron spectroscopy (AES). Sharp LEED patterns were obtained from both substrates prior to growth of the films. LEED patterns from the Fe/Cu(100) films showed a fourfold cubic pattern with 5×1 superstructures.[12] The LEED patterns from the Fe/Ag(100) showed a broadened, diffuse 1×1 pattern similar to that described by Li.[20] The AES analysis revealed only trace contamination levels of oxygen ($\simeq$1% eq. monolayer). Here we note that in the magnetic characterization of the films, the Fe/Cu(100) switching transition was reversible only for $T<350$ K.[11] However, as indicated by the stability of the Fe/Cu Auger intensity ratios,[25] films thicker than 3 monolayers (ML) do not begin to interdiffuse until $\simeq$500 K. An irreversible transition near $T=350$ K from a tetragonally distorted fcc to an unstrained fcc structure has recently been reported in thin Fe/Cu(100) films[26] which may be related to the irreversible loss of polarization we observed. In the Fe/Ag(100) system, however, the magnetization tends to switch irreversibly into the plane after annealing to above 300–325 K, depending on film thickness and the annealing rate. This change in the magnetic properties is correlated with a decrease in the Fe/Ag AES peak ratio, indicating that interdiffusion or surface segregation begins in at relatively low temperatures.

The SPSEES analysis was performed *in situ* with the apparatus described elsewhere.[11] An important aspect of the SPSEES measurement is that it averages over the area emitting secondary electrons, which are generated using a 1 mm^2 primary beam of 1-keV electrons. All polarizations were measured in zero applied field, and at every temperature the perpendicular and in-plane polarization measurements were conducted separately by reversing the magnetization with 400 Oe **B** pulses. In this manner it was possible to measure the direction of $\mathbf{M}_r$.

In Fig. 1 the thickness dependence of the low-energy (0–2 eV) secondary electron spin polarization, P_{sec}, is shown for both Fe/Ag(100) and Fe/Cu(100) at 125 K. The solid lines show a fit of the polarization which assumes that the film forms a single magnetic domain and the polarized electrons from the film are diluted by exponentially attenuated, unpolarized electrons from the substrate. The dotted line traces out the observed polarization behavior of the films. The thickness at which the polarization first appears is $\simeq$1.5 and 2 ML for the Fe/Ag(100) and Fe/Cu(100), respectively [using layer spacings of 1.44 and 1.8 Å for the (100) bcc- and fcc-Fe faces]. This sudden onset of long-range order is consistent with, e.g., a reduced critical temperature ($T_c<125$ K) for film thickness below these values.

For thicknesses up to $\simeq$4.9 ML in Fe/Ag(100) and $\simeq$6.1 ML in Fe/Cu(100), perpendicular $\mathbf{M}_r$ is observed, and the polarization fits well to the single-domain model. This agrees with magneto-optic Kerr effect measurements,[9] where a square hysteresis loop is reported. The subsequent observation of $\mathbf{M}_r$ switching into the plane of the film as the film thickness increases is accompanied by an initial loss of spin polarization, then the in-plane polarization component increases rapidly again as the thickness

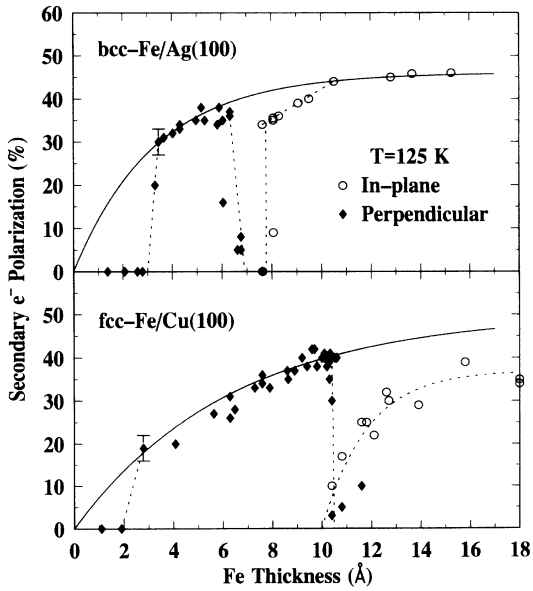

FIG. 1. Low-energy secondary electron spin polarization from Fe/Ag(100) (top panel), and Fe/Cu(100) (bottom panel) at 125 K. The solid lines represent a single-domain model for the increase of the polarization, with saturation polarizations of 45% and 50%, respectively. The dotted lines trace the actual polarization behavior of the films.

is increased further. The top panel of Fig. 1 shows that the in-plane polarization for the Fe/Ag(100) films returns to the polarization extrapolated from the thinner, perpendicularly magnetized films, with a saturation polarization of 45%. In the lower panel, however, it can be seen that the in-plane polarization from the Fe/Cu(100) saturates at $\simeq$35%. This is considerably lower than the extrapolated perpendicular polarization of $\simeq$50%.

The two main features in Fig. 1 which we would like to stress are the generally reduced polarization near the switching thickness and the saturation polarization for the thick films.

First, a reduction of the polarization is indicative of a loss of long-range order at that particular thickness and temperature range. This could be caused, e.g., by the collapse of the magnetization into microscopic closure domains, as seen for Co/Au(111).[6] Alternative explanations for the loss of polarization, such as a paramagnetic or antiferromagnetic state of the Fe films, however, cannot be ruled out at this point.

Second, the values of the secondary electron saturation polarization are significant since for bulk Ni, Co, and Fe surfaces the polarization increases nearly linearly with the local magnetic moment.[27-29] Specifically, bulk bcc Fe, with a moment of $2.2\mu_B$, shows a $P_{sec}=47\%$. Clearly, then, the top panel of Fig. 1 shows that the bcc Fe/Ag(100) remains magnetized in a single domain (for thicknesses $3.5 < d < 6.5$ Å and $d > 10$ Å), with a moment comparable to that of bulk bcc Fe. The case of fcc Fe/Cu(100) is more complicated. P_{sec} from the perpendicular films ($3 < d < 10$ Å) follows the single-domain curve, with an extrapolated saturation polarization of 50%. Again, this is indicative of a moment comparable to that of bcc Fe, in agreement with neutron scattering experiments.[30] However, the in-plane films ($d > 12$ Å) appear to be saturating at a substantially lower polarization (35%), which would be consistent with an $\simeq$25% reduced moment in this thickness range. These data illustrate the possible pitfalls of equating the magnetic properties of thick Fe/Cu(100) films with those of thin films, as in Ref. 31, where the moment and exchange splitting of Fe/Cu(100) are suggested to be linearly related with a slope of 1 eV/μ_B. Although we consider it unlikely that this relation holds for all transition metals irrespective of the environment[31] (i.e., atom, solid, alloy), a more correct comparison of moment and exchange splitting for the particular case of the thick Fe films gives a better fit to this trend than is shown in Ref. 31. In order to support their point, the authors of Ref. 31 correlate a 1.2 ± 0.1-eV exchange splitting which was actually measured on a 15-Å film to a $(1.9\pm0.6)\mu_B$ moment for a 5.4-Å-thick film.[31] If the 15-Å film does have a reduced moment of $1.4\mu_B$, as the reduction of P_{sec} suggests, then the ratio of exchange splitting to moment is $\simeq$0.8 eV/μ_B.

Finally, further indications of domain formation during the switching transition of $\mathbf{M}_r$ are apparent in the temperature dependence of P_{sec}. The switching of $\mathbf{M}_r$ from perpendicular to in plane with temperature in Fe/Cu(100) and Fe/Ag(100) is shown in Figs. 2 and 3. For the Fe/Cu(100), the transition is totally reversible unless the annealing temperature is above $\simeq$350 K. The data shown in Fig. 2 were taken after the film had been annealed to 300 K once. The polarization of the Fe/Cu(100) films during the switching transition goes through several stages as the temperature is increased. $P_{sec}=41\%$ at 125 K. It decreases slowly with temperature, then as $\mathbf{M}_r$ starts to switch the perpendicular component first decreases sharply, and then exhibits a slight increase just above the switching temperature. This could be caused by either a canting of the magnetization in the domains, as reported in Ref. 6, or a co-existence of microscopic perpendicular and in-plane domains. Additionally, the $\mathbf{M}_r$ appears to favor the in-plane (110) axis (as defined in Fig. 2) during the transition, switches to the (001) axis up to 300 K, then

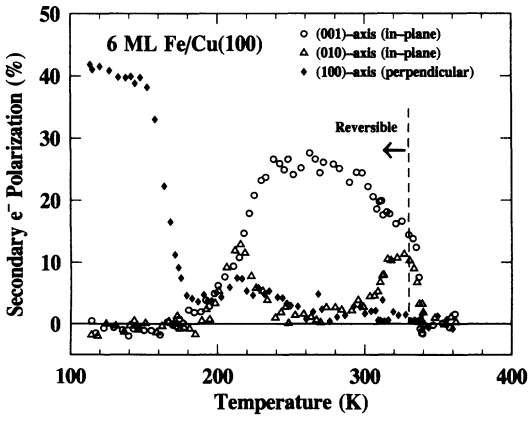

FIG. 2. Temperature dependence of the low-energy secondary electron spin polarization from 6 ML Fe/Cu(100). The film had been annealed once to 300 K.

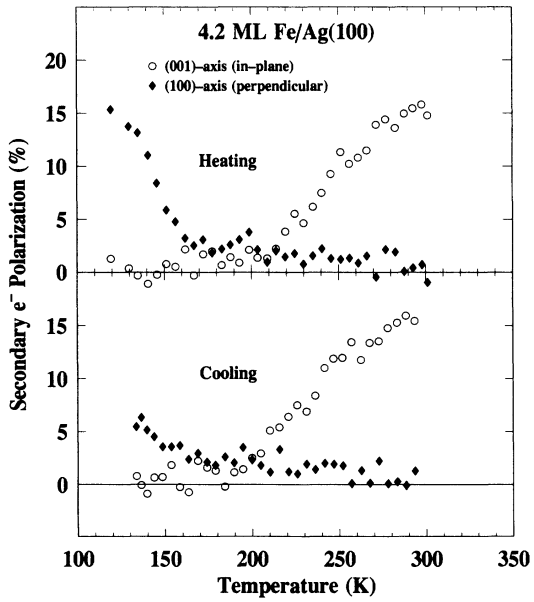

FIG. 3. Temperature dependence of the low-energy secondary electron spin polarization from 4.2 ML Fe/Ag(100). The first anneal cycle to 300 K is shown in the top and bottom panels after the film was grown at 125 K.

goes back to the in-plane (110) axis just before the irreversible loss of P_{sec} at $\simeq 350$ K.

The switching transition with temperature for Fe/Ag(100) is qualitatively broader than the transition in Fe/Cu(100), as seen by comparing Fig. 3 to Fig. 2. The first anneal cycle of an Fe/Ag(100) film to 300 K is plotted in the top and bottom panels of Fig. 3, and illustrates that the $\mathbf{M}_r$ switching transition in Fe/Ag(100) is reversible. A slight shift of the transition down by $\simeq 10$ K is observed after the 300-K anneal, showing that the perpendicular anisotropy energy term is slightly reduced. This is probably due to Ag interdiffusion, as discussed above. In contrast to Fe/Cu(100) films, which could be annealed reversibly many times to temperatures less than 350 K, the Fe/Ag(100) films typically could only be annealed 1–2 times to 300 K before the perpendicular remanence at low T was gone.

In conclusion, we have measured the secondary electron spin polarization from Fe films grown epitaxially on Cu(100) and Ag(100) substrates at 125 K. The effects of varying the temperature and thickness on the remanent magnetization were investigated. A switching transition of $\mathbf{M}_r$ could be induced in both systems by varying the thickness and temperature. At 125 K, the switching transition occurs at $\simeq 7$ and 11 Å ($\simeq 4.9$ and 6.1 ML) for Fe/Cu(100) and Fe/Ag(100), respectively, and the transition is reversible with temperature cycling in both systems. A reduction of P_{sec} was observed during the switching transition. By comparison to spatially resolved studies on other systems we would conclude that the remanent magnetization collapses into microscopic closure domains as the orientation of $\mathbf{M}_r$ switches. However, the possibility remains that the Fe passes through either a paramagnetic or antiferromagnetic state near the switching thickness and temperature. The application of SEMPA or SPLEEM to these systems should provide an answer to this question.

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