

# Magnetism of ultrathin films of Fe on Cu(100)

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Magnetism in ultrathin (1–10 ML) Fe films grown on Cu(100) has been studied by spin-polarized secondary electron emission spectroscopy. The variation of the magnetization with temperature and oxygen adsorption was investigated for various film thicknesses. The orientation of the magnetization for films between 5 and 6 ML thick switches reversibly between perpendicular (at low temperature) to in-plane (at high temperature). The switching transition temperature decreases with increasing film thickness, and is accompanied by a loss of long-range order over a range of 20–30 K. The transition is attributed to the temperature dependence of the perpendicular anisotropy. The effect of oxygen adsorption onto films with perpendicular remanence is to first suddenly turn the magnetization into the plane at a critical coverage, and then to kill the magnetization gradually with continued exposure. This indicates that the uniaxial surface anisotropy at the Fe-vacuum interface plays a major role in the magnetization of the film.

Magnetic thin films are an exciting field for the study of the magnetic properties of condensed matter in novel systems and at interfaces.<sup>1</sup> The two-dimensional nature of the films indicates that the magnetism is driven by anisotropies,<sup>2,3</sup> which are dominated by the surface anisotropy  $K_u$  and dipole demagnetizing energy.<sup>4</sup> For thin fcc Fe overlayers on Cu(100),  $K_u$  is perpendicular to the film,<sup>5,6</sup> while the dipole demagnetizing energy always favors in-plane magnetization.

In this study we have observed the competition between these two anisotropies in Fe/Cu(100) by measuring the direction of the remanent magnetization for different film thicknesses, temperatures, and oxygen coverages. The Fe films were grown at low temperature ( $\approx 125$  K) in order to minimize Fe/Cu intermixing,<sup>7</sup> and then annealed to 300 K before data were taken. The base pressure of the UHV system was  $5 \times 10^{-11}$  Torr, and rose to  $3 \times 10^{-10}$  Torr during the Fe deposition. The film thickness was monitored by a quartz-crystal microbalance, which was calibrated independently against an optical interferometer. This resulted in identifying the first break of slope in the normalized Auger Fe uptake curve with the completion of a bilayer (within 10% accuracy), in agreement with thickness calibrations from Refs. (8) and (9). Therefore, the thicknesses presented here are a factor of 2 larger than our first measurements,<sup>10</sup> which were calibrated using the first kink in the Auger uptake curve as 1 ML. The only detectable impurity in the Auger spectra of the films was oxygen, at levels of  $O_{503 \text{ eV}}/Fe_{703 \text{ eV}} \sim 1/40$ .

The structure of the films was monitored by LEED. Figure 1 shows the LEED evolution of a 3.4 ML film (deposited at 125 K) as it is annealed. The as-evaporated LEED is a diffuse  $1 \times 1$  pattern, and is not shown. At 300 K, streaky fourth (or fifth)-order superstructure spots are apparent between the (10) and (11) spots, as in a  $4 \times 1$  (or

$5 \times 1$ ) superstructure<sup>11</sup> with the central spot(s) missing. Annealing to 400 K sharpens the pattern, and a faint  $p(2 \times 2)$  reconstruction appears to be superimposed onto the superstructure. After a 500 K anneal, the superstructure is almost gone, and a  $p(2 \times 2)$  reconstruction is evident. At  $\approx 650$  K, the Fe/Cu Auger ratio begins to decrease, and there is a transition to a  $c(2 \times 2)$  reconstruction, shown in Fig. 1 at 700 K.

The magnetization in the films was monitored *in situ* by spin-polarized secondary electron emission spectroscopy. In our apparatus the full vector polarization of the secondary electrons is analyzed, giving a signal proportional to the film magnetization averaged over the primary spot size ( $\approx 1$  mm diameter). It is possible to acquire all three components of polarization in a few seconds, thus allowing essentially continuous monitoring of the magnetization during temperature cycling and oxygen exposure.

Magnetization curves for 4.6- and 10-ML films are shown in Fig. 2. The coordinate system is defined with the  $z$  axis perpendicular to the plane of the film, along the [100] direction, and the  $x$  axis in the plane of the film, along the [010] direction. The magnetization at each temperature is measured in remanence, i.e., after a magnetizing pulse ( $\approx 200$  Oe) is applied in the  $z$  and then  $x$  direction for the perpendicular and then in-plane magnetization measurement.

The 4.6- and 10.0-ML films, in Fig. 2, show only magnetization perpendicular and in the plane of the film, respectively. These results match with previous studies on this system, where the border between perpendicular and in-plane magnetization was at  $\approx 6$  ML.<sup>6</sup>

The temperature dependence of the anisotropy is evident for films that are right on the borderline. Figure 3 shows the magnetization for 5.0- and 6.0-ML films switching from perpendicular to in-plane as the temperature is

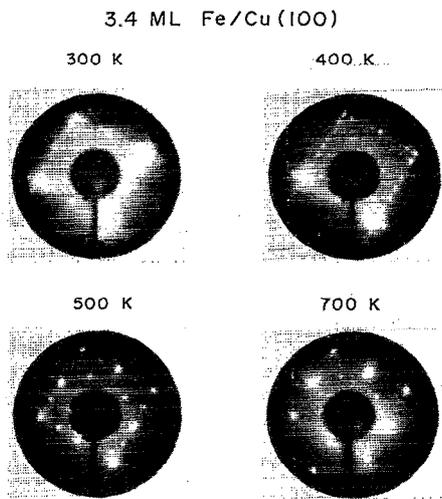


FIG. 1. LEED evolution of a 3.4-ML film of Fe/Cu(100) as it is annealed. The growth temperature was 125 K. The LEED energies are 174.6, 170.3, 162.4, and 174.0 eV for 300, 400, 500, and 700 K, respectively. The top spot in each pattern is an (11) spot.

increased. This switching behavior is completely reversible for  $T < 350$  K. Above 350 K there is an irreversible loss of polarization accompanied by a sharpening of the LEED pattern with similar, but much weaker, superstructure spots as the 3.4-ML film in Fig. 1. At the switching temperature  $T_s$ , there is a characteristic loss of polarization. The loss of polarization is attributed to either: (i) the loss of long range order when the perpendicular anisotropy energy is approximately equal to the dipole demagnetizing energy (this would correspond to a paramagnetic state of the film, because at finite temperature a minimum anisotropy is required to sustain long-range order in a 2D system<sup>3</sup>); (ii) the existence of microscopic ( $< 1$  nm) domains, where the domain size varies with the anisotropy.<sup>12</sup>

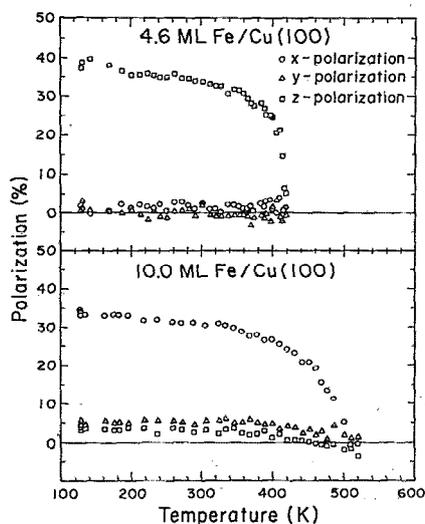


FIG. 2. Temperature dependence of the spin polarization of secondary electrons from a 4.6-ML film (top panel) and a 10-ML film (bottom panel) of Fe/Cu(100).

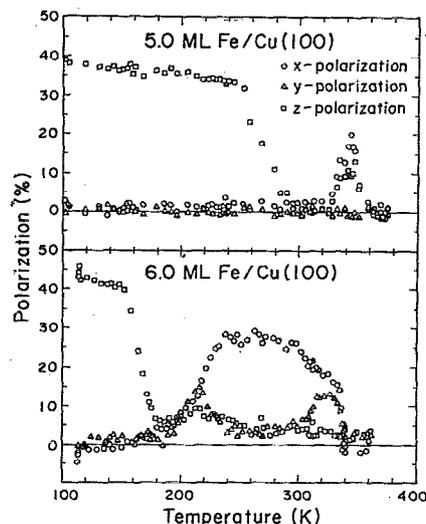


FIG. 3. Temperature dependence of the spin polarization of secondary electrons from a 5.0-ML film (top panel) and a 6.0-ML film (bottom panel) of Fe/Cu(100).

The temperature dependence of the perpendicular uniaxial anisotropy can be inferred by the dependence of  $T_s$  on film thickness  $d$ . At  $T_s$  the perpendicular surface field compensates the demagnetizing field,<sup>13</sup> i.e.,

$$2K_u(T_s)/M_s d = 4\pi D_{\text{eff}} M_s \quad (1)$$

where  $D_{\text{eff}}$  is the correction factor for the discrete nature of the film layers.<sup>13</sup> The switching temperature is observed to vary dramatically with film thickness, as illustrated in Fig. 4. Since the demagnetizing field is relatively constant for  $5 \text{ ML} < d < 6.5 \text{ ML}$ , from Fig. 4 one finds that

$$K_u(300 \text{ K})/K_u(150 \text{ K}) = 5 \text{ ML}/6.5 \text{ ML}, \quad (2)$$

and  $K_u$  must decrease by  $\approx 25\%$  between 150 and 300 K.

The quenching of the perpendicular anisotropy was then studied by oxygen adsorption. In Fig. 5 it can be seen that the magnetization begins to turn into the plane of the film at 0.4 L oxygen exposure, and is completely in the

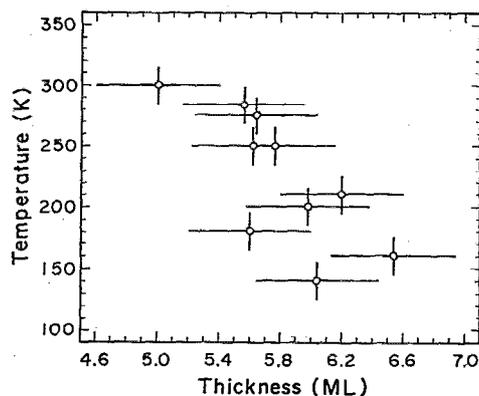


FIG. 4. Thickness dependence of the transition temperature  $T_s$  (switching temperature) between perpendicular and in-plane magnetization.

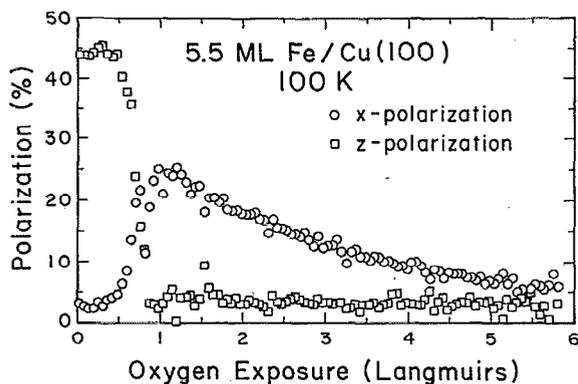


FIG. 5. Spin polarization of secondary electrons from a 5.5-ML film during oxygen exposure at  $1 \times 10^{-9}$  Torr (1 L = 1 s at  $1 \times 10^{-6}$  Torr) at  $T = 100$  K.

plane after 0.8 L. The sample was held at low temperature ( $\approx 125$  K), and therefore the sticking coefficient is expected to be close to 1.

Films from 3.4 to 6 ML have all shown switching of the easy axis after  $\approx 0.5$  ML oxygen adsorption. The magnetization of the films under oxygen exposure shows three distinct stages (Fig. 5) as follows.

(i) In the initial stage of oxygen adsorption the total polarization is relatively constant up to a critical coverage, when the magnetization suddenly begins to switch into the plane of the film.

(ii) during the intermediate stage, i.e., while the film is switching, the polarization begins to decrease. If the oxygen treatment is stopped while the easy axis is switching, the magnetization can be frozen at an angle and then pulled back perpendicular by cooling the crystal to a lower temperature.

(iii) The final stage is characterized by the magnetization completely in the plane of the film and decreasing gradually to zero.

A total loss of the polarization during switching has not been observed under oxygen exposure. The inherent difference between oxygen and temperature cycling (i.e., a change in the Fe-vacuum interface morphology) must inhibit the mechanism responsible for the loss of long-range

order near  $T_s$ . We conclude, therefore, from the sensitivity of the anisotropy to oxygen, that the Fe-vacuum interface makes a significant contribution to the total anisotropy of Fe/Cu(100) overlayers. This supports the conclusions made by Heinrich *et al.*,<sup>13</sup> from FMR data of Fe on Ag, where the vacuum interface showed the strongest surface anisotropy.

In this study of the magnetization of thin Fe overlayers on Cu(100) we have identified a thickness range (5–6 ML) in which the magnetic easy axis switches reversibly from perpendicular to in-plane between 100 and 300 K. The switching is attributed to the temperature dependence of the surface uniaxial anisotropy  $K_u$ . The dependence of the switching temperature on the film thickness was used to show that the uniaxial surface anisotropy is reduced by  $\approx 25\%$  between 150 and 300 K. There is a characteristic loss of long-range order in the region of the switching temperature.

Small amounts of oxygen ( $\approx 0.5$  ML) also caused a switching of the easy axis into the plane of the film, however, an analogous loss of long-range order was not observed. We conclude that the Fe-vacuum interface plays a major role in the anisotropy of thin Fe overlayers on Cu(100).

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