Magnetism of Epitaxial fcc Iron Films on Cu(001) Investigated by Spin-Polarized Photoelectron Emission

D. Pescia,[a] M. Stampanoni, G. L. Bona, A. Vaterlaus, R. F. Willis,[b] and F. Meier

Laboratorium für Festkörperphysik, Eidgenössische Technische Hochschule–Hönggerberg, CH-8093 Zürich, Switzerland

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One-, three-, and five-monolayer (ML) films of fcc iron grown epitaxially on Cu(001) are found to be ferromagnetic by spin-polarized photoelectron emission measurements of the magnetization as function of perpendicularly applied field and temperature. The hysteresis loop of the 5-ML film at 30 K is quadrangular, and shows a remanence magnetization directed along the surface normal and a coercive field of 650 Oe. The extrapolated temperature dependence of the saturation magnetization leads to a Curie temperature of 230 K for the 1-ML film and 390 K for the 3- and 5-ML films.

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Iron crystallizes in two phases: up to 1184 K, it is bcc, between 1184 and 1665 K, fcc, and from there up to the melting point at 1809 K it is again bcc.[1] From many sides, much activity is presently focused on the exploration of the ground-state magnetic properties of fcc iron. Alloy-stabilized microcrystallites of fcc iron are the key ingredient of nonmagnetic (austenitic) stainless steels and therefore of enormous technological importance.[2] Thin films of fcc iron can be grown epitaxially on metallic substrates, the lattice structure being preserved over the whole temperature range. However, the study of the magnetic properties has produced conflicting results.[3,4] The discrepancies are yet unexplained. One possible source of disagreement—the crystallographic orientation of the surface—can be excluded according to Ref. 4. In Fernando *et al.*[5] the magnetization of epitaxially grown fcc iron on Cu(001) is studied by the surface magneto-optic Kerr effect. The iron films showed a complicated magnetic behavior: Ferromagnetism appeared if the film was deposited at an elevated substrate temperature of 190 °C and disappeared after cooling to room temperature or keeping the sample for 1 h at 190 °C. Films evaporated onto a substrate held at room temperature were not ferromagnetic. Finally, fcc iron is a test ground for highly developed band-theory investigations. These methods apparently have become sophisticated to the point where they are able to discriminate between the small energy differences of various magnetic configurations.[6]

In the present experiment, clean 1-, 3-, and 5-monolayer (ML) films of fcc Fe grown on Cu(001) were studied by spin-polarized photoelectron emission. All films were ferromagnetic, the 3- and 5-ML films having a Curie temperature of 390 ± 30 K. At room temperature the 3- and 5-ML films showed no measurable remanence, but at 30 K hysteresis loops with coercive fields of 350 Oe and 650 Oe, respectively, were observed. The easy direction of the magnetization is perpendicular to the plane of the film. For the 1-ML film no hysteresis loop was resolved even at low temperatures. Contamination of the 5-ML film by about 10-at.% carbon destroys the ferromagnetic order.

The thin iron films were prepared by evaporation from a resistively heated Fe filament. Extensive outgassing is necessary to minimize carbon contamination. On Cu(001) the Fe film grows in a layer-by-layer mode as verified by the typical dependence of the Auger amplitudes on film thickness: During growth of a monolayer, they increase linearly with the thickness but there is a change of slope when one layer is completed and the next starts developing.[7] The $p(1\times1)$ LEED pattern of the Fe film is identical to that of the copper substrate. Carbon contamination on top of the surface of the film is found to cause extra spots at the $\pm\frac{1}{2}$ positions, as for Co on Cu(001).[8] However, in contrast to the Co/Cu(001) system, carbon seems to be easily incorporated into the Fe epitaxial films. Although no extra spots are then visible, this is recognized by an enhanced diffuse luminosity of the LEED screen together with an Auger C signal well above the noise limit (about 2 at.%). During evaporation the copper substrate was at room temperature. Perfect epitaxial growth of fcc Fe occurs for films up to 5 ML. The formation of bcc inclusions prevents the preparation of thicker films.

The technique of spin-polarized photoelectron emission is described by Campagna *et al.*[9] The polarization of the emitted electrons is $P=(N_\uparrow-N_\downarrow)/(N_\uparrow+N_\downarrow)$ where $N_\uparrow$ ($N_\downarrow$) is the number of electrons with spin magnetic moment parallel (antiparallel) to the magnetization of the sample. The magnetic field to saturate the film is applied perpendicularly to the surface. As light source, the full spectrum of a Hg-Xe lamp was used, $h\nu \leq 5.5$ eV. The photothreshold of all samples was 4.7 ± 0.1 eV. An extensive discussion about the proportionality between the spin polarization of photoelectrons and the magnetization generally, and in particular for iron, has been given by Kisker and co-workers.[10] Since no energy and angular resolution is made in the present experiment, the measured P is determined by the spin density of states of those electrons which are emitted by

the spectral intensity distribution of the light source. Analysis of the polarization values versus film thickness gives an average escape depth of the photoelectrons of 6 Å in agreement with Ref. (5).

Magnetization curves of the 5-ML film taken at 215, 267, and 375 K are shown in Fig. 1(a). At these temperatures the magnetic field dependence $P(H)$ is reversible with the magnetization lying in the plane at $H=0$. However, the effect of increasing perpendicular anisotropy upon lowering of the temperature is noticed by the increasingly smaller saturation field. Finally, at 30 K, a quadrangular hysteresis loop is observed [see Fig. 1(b)]. The remanence is equal to the full saturation magnetization, i.e., the perpendicular anisotropy is strong enough to keep the film in a perpendicularly magnetized single-domain state.

It is tempting to infer from the shape of the hysteresis curve the mechanism of the magnetization reversal. Two types of anisotropies must be present: the shape anisotropy—corresponding to the energy $2\pi M_s^2$, where M_s is the saturation magnetization—tending to keep the magnetization in the plane, and the magnetocrystalline, uniaxial anisotropy K_u tending to keep the magnetization perpendicular to the plane. Evidently, at 30 K, K_u wins, which means that $K_u > 2\pi M_s^2$ at this temperature.

With the assumption of a uniformly magnetized film where M is the same for all five layers, the simplest mechanism of magnetization reversal is by coherent rotation of all spins, i.e., the film always remains in a single-domain state.[11] Then, as one moves on the upper branch of the hysteresis loop, a discontinuous jump from M_s to $-M_s$ occurs at the external field $H=-2K_u/M_s+4\pi M_s$, where $2K_u/M_s$ is the anisotropy field and $4\pi M_s$ the demagnetizing field of the perpendicularly magnetized film. Obviously such behavior, which would result in a rectangular hysteresis loop, is not observed.[12]

A prerequisite for obtaining insight into the mechanism of the magnetization reversal in these films is the experimental observation of the domain structure. Suitable, highly surface-sensitive magnetic probes with good spatial resolution are now being developed: spin-polarized secondary electron emission,[13] and, based on it, spin-polarized scanning electron microscopy,[14] or even spin-polarized scanning tunneling microscopy.

Magnetization curves at 30 K for a 3- and a 1-ML film are shown in Figs. 2(a) and 2(b), respectively. The 3-ML film still produces a clearly resolved hysteresis, but (compared to the 5-ML film) with reduced coercivity, reduced remanence, and a shape which is no longer quadrangular, but rounded. For the 1-ML film no hysteresis is observed. Evidently, the magnetocrystalline anisotropy depends on the thickness of the film—in a way, however, which was hardly anticipated. For a 1-ML fcc iron film at the lattice spacing of Ag, the direction of spontaneous magnetization is predicted to be perpendicular.[15] Since the magnetic properties of fcc iron vary sensitively with the interatomic spacing[16] the observed thickness dependence of the anisotropy may well be an effect of the smaller lattice constant of Cu as compared to Ag. Apart from this geometrical factor, the specific electronic structures of Ag and Cu are believed to have

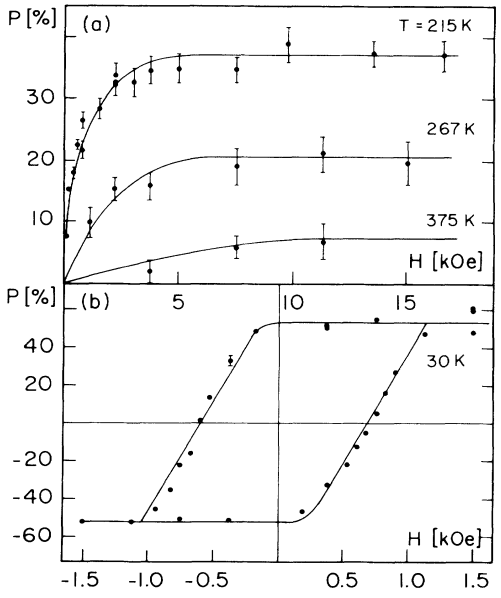

FIG. 1. Polarization $P(H)$ of a 5-ML fcc iron film on Cu(001) (a) for sample temperatures $T=215$, 267, and 375 K, (b) at $T=30$ K. H is the applied magnetic field perpendicular to the plane of the film. The error bars indicate the statistical error.

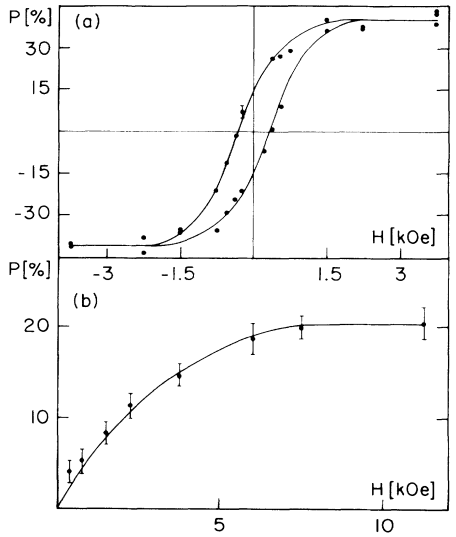

FIG. 2. Polarization $P(H)$ measured at $T=30$ K (a) for 3-ML fcc Fe on Cu(001), (b) for 1 ML. H is the applied magnetic field perpendicular to the plane of the film. The error bars indicate the substantial error.

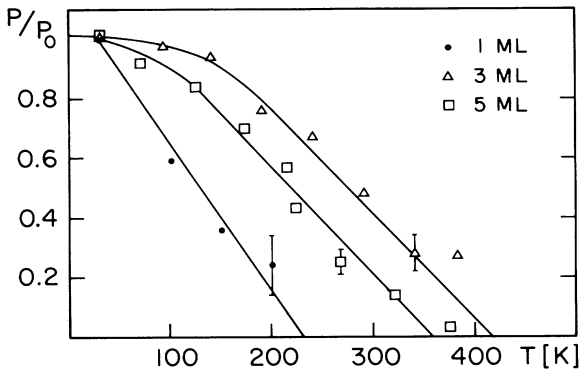

FIG. 3. Temperature dependence of the reduced polarization P/P_0 for 1-, 3-, and 5-ML films of fcc-Fe on Cu(001). P_0 is the saturation polarization at low temperatures: 54%, 40%, and 20% for 5, 3, and 1 ML, respectively. The Curie temperatures are 230 ± 30 K for the 1-ML film and 390 ± 30 K for the 3- and 5-ML films. The error bars indicate the statistical error.

no profoundly different effects on the magnetic Fe overlayer since the d bands of Cu and Ag are both energetically well separated from those of Fe.[6]

The temperature dependence of the saturation magnetization is shown in Fig. 3 for all three films. The external magnetic field was 7.5 kOe for the 1-ML film and 3.8 kOe for the 3- and 5-ML films. As a result of the H field the transition temperatures are not well defined. For bulk materials measured in an applied magnetic field it is customary to determine the Curie temperature by extrapolation to $M_s = 0$ of the linear portion of the $M_s^2(T)$ relation.[17] The magnetization of thin films—or surfaces—is known to vary linearly with temperatures near T_C.[18] Therefore, with adoption of a correspondingly modified procedure the Curie points are obtained by extrapolation of the linear parts of $M_s(T)$ instead of M_s^2. The following values are obtained: T_C of the 1-ML film is 230 K, for the 3- and 5-ML films 390 K. The uncertainty in all three cases is estimated to be ± 30 K. No diffusion of Fe into the Cu substrate occurred, since the $P(T)$ curves were perfectly reversible. This is in agreement with the metallurgical fact that the solubility of Cu in Fe is negligible below 100 °C [19]; in fact, iron is used as a diffusion barrier for Cu atoms.[20] Although there is a systematic difference between the measured $P(T)/P_0$ data of the 3- and 5-ML films, we hesitate to attach intrinsic significance to it but prefer to attribute it to unknown, spurious effects of the film quality.

Thus, fcc iron stabilized at the lattice constant of Cu has been found to have a ferromagnetic ground state. This is the main result of the present study. It is difficult at present to account for the discrepancies with Ref. 5. One of the problems is certainly the extreme sensitivity of the magnetic properties of fcc iron on Cu(001) to changes of the lattice constant; see Ref. 16. An unexpected phenomenon is the observed dependence of the anisotropy on temperature and film thickness, which certainly deserves more attention. Since the electronic structure of a 4-ML film has been found to be already fully bulklike,[21] the T_C of the 3- and 5-ML films is considered to be the transition temperature of the bulk fcc Fe on Cu(001).

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[a]Permanent address: Institut für Festkörperforschung der Kernforschungsanlage Jülich GmbH, D-5170 Jülich 1, Federal Repubic of Germany.

[b]Permanent address: Cavendish Laboratory, University of Cambridge, CB3 0HE Cambridge, United Kingdom.

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