Magnetism of atomically thin fcc Fe overlayers on an expanded fcc lattice: $Cu_{84}Al_{16}(100)$

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We present experimental data on the magnetic properties of atomically thin fcc (γ-phase) Fe films (1–6 atomic layer nominal thickness) epitaxially grown on $Cu_{84}Al_{16}(100)$ obtained by linear magnetic dichroism in the angular distribution of Fe $3p$ core photoelectrons excited by linearly polarized synchrotron radiation. The sign and magnitude of the Fe $3p$ photoemission magnetic asymmetry indicates the onset of in-plane ferromagnetism at 2.5(2) monolayer (ML) thickness of γ-Fe. The Curie temperature is 288(2) K for 4 ML thickness. The magnetic splitting of the Fe $3p$ mj core hole sublevels is 1.10(2) eV, i.e., the same value as measured for a bcc-Fe(100) surface where large surface and near-surface enhanced moments contribute. These results characterize the epitaxial γ-Fe on $Cu_{84}Al_{16}(100)$ as a high-spin ferromagnet for thickness up to 4 ML, with an average magnetic moment per iron atom of $2.5(1)\mu_B$. A phase transition occurs between 4 and 5 ML thickness: the magnetic order of the pseudomorphic γ-Fe film decreases consistently with the breaking into two phases with the deeper layers in a low-spin and/or antiferromagnetic phase and surface restricted ferromagnetism, similar to the case of γ-Fe/Cu(100). [S0163-1829(98)09833-6]

INTRODUCTION

Achieving an understanding of the properties and behavior of artificial nanostructures obtained by epitaxial growth and other new techniques in the search for unusual properties of materials is one of the dominant trends in physics and materials science nowadays. For magnetic materials, the search for correlation between structure and magnetic properties of metastable phases of transition metals and other nonconventional materials is particularly interesting. Among these metastable phases, fcc Fe (γ-Fe) has attracted much attention from theoretical and experimental groups. The relevance of this fact lies in the possibility of studying the magnetic properties of iron in metastable phases where it is expected that magnetovolume effects determine the ground-state configuration.[1–3] Theoretical calculations of the ground-state of bulk γ-Fe predict a nonmagnetic phase, an antiferromagnetic phase, and two ferromagnetic phases, one with a small magnetic moment of $1.1\mu_B$, and one with a high magnetic moment of $2.5\mu_B$ per atom.[1–3] The lattice parameter, i.e., the volume available for the iron atoms, determines the ground state of γ-Fe and a magnetovolume instability is predicted at 3.66 Å lattice spacing (or 12.25 $Å^3$ volume) inducing a first-order phase transition from the antiferromagnetic and low-spin phases to the ferromagnetic high-spin state.[1–3] Moreover, within the low-spin and in the high-spin states, the fcc Fe magnetic moment is predicted to increase monotonically with increasing lattice spacing.[1–4] Experimentally, bulk fcc Fe is stable only at elevated temperatures (>910 °C) as a paramagnetic metal and can be stabilized at lower temperatures as coherent precipitates in Cu and CuAl matrixes[5,6] or by epitaxy onto suitable fcc substrates.[7–17] Cu has a lattice parameter of 3.61 Å and therefore a volume per atom of 11.76 $Å^3$ that would correspond to a γ-Fe antiferromagnetic or low-spin phase.[1] Ferromagnetic order of γ-Fe was observed first for films grown by electrolytic methods on Cu(110) (Ref. 8) and for ultrathin films grown by molecular-beam epitaxy (MBE) methods on Cu(111). The experimental evidence of the stabilization of the high-spin phase, with a measured moment of $2.6\mu_B$, was obtained for γ-Fe as grown on CuAu substrates.[11] The growth of γ-Fe on Cu(100) is in fact pseudomorphic: an expansion of the lattice perpendicular to the surface can lead the system to the magnetovolume instability. The complexity of the experimental results obtained on Fe/Cu(100) is understood as a consequence of the exact structure taken by the γ-Fe as a function of thickness, deposition temperature, and interdiffusion with the substrate. A complex correspondence of magnetism and structure has been recently established.[18–28] Nondistorted fcc Fe/Cu(100) shows antifer-

romagnetic order, while an expansion of the Fe lattice perpendicular to the surface can lead to ''fcc-like'' Fe in the ferromagnetic state.[19–26]

In this paper we present surface sensitive magnetometric results obtained on epitaxial ultrathin Fe films grown on $Cu_{84}Al_{16}(100)$, a fcc substrate chosen in order to favor the stabilization of the ''high spin'' phase of γ-Fe since it has a lattice parameter 1% larger than pure fcc-Cu and therefore a 12.15 $Å^3$ volume, tuned to that predicted for the high-spin fcc Fe. The magnetism of the fcc iron epitaxial layers has been probed by linear magnetic dichroism in the angular distribution of photoelectron intensity (LMDAD) of the Fe $3p$ core levels. This novel method[29–32] allows us to analyze, in a fairly independent way,[33,34] the *magnetic order* of the fcc iron surfaces via the *magnitude* of the LMDAD asymmetry, and the relative changes of the *local magnetic moment* of the Fe atoms via the changes of the *energy splitting* of the magnetic sublevels of the Fe $3p$core hole. This is a photoelectron spectroscopy experiment: the magnetic information that is derived is an average of the contributions of the top layers, weighted by the photoelectron escape depth.

EXPERIMENT

Epitaxial Fe overlayers (1 to 6 ML thick) were grown under MBE conditions by *e*-beam evaporation of a high-purity Fe wire $(5N)$ onto a clean surface of a $Cu_{84}Al_{16}(100)$ single crystal that presents a lattice parameter of 3.65 Å, as determined by x-ray diffraction. Both growth and measurements were performed in an ultrahigh vacuum system with a base pressure of 1×10^{-10} mbar connected to the SU7 undulator beam line at the SuperAco storage ring at Orsay. The energy of the linearly polarized undulator radiation was selected at 170 eV in order to obtain Fe $3p$ photoelectrons with kinetic energies such to give a minimum probing depth $\lambda=5(1)$ Å. This means that about 30% of the photoemission intensity originates from the surface and that the subsurface contributions are weighted by $e^{-d/\lambda}$, where d is the distance of the deeper layer from the vacuum interface. The $Cu_{84}Al_{16}(100)$ surface was cleaned by cycles of Ar^+ sputtering and annealing. Reproducible results were obtained for 30 min Ar^+ sputtering at 1.5 kV at room temperature (RT) followed by 20 min annealing at 650 K: sharp $p(1\times1)$ low-energy electron diffraction (LEED) patterns for 75 eV primary electrons were obtained. No traces of C were measured, but a small oxide signal on the Al $2p$ core level photoemission peak indicated the presence of some oxidized aluminum at the surface. The Fe evaporation rate was 0.3 Å/min, as determined by a calibrated quartz oscillator, and the residual gas pressure during the Fe evaporation was always better than 8×10^{-10} mBar. Two growth conditions were explored: the $Cu_{84}Al_{16}$ substrate was held at RT and at 150 K during iron deposition. The magnetic measurements were done by LMDAD using the same experimental setup and chiral geometry as described elsewhere.[35]

We have measured the Fe $3p$ core level from the Fe monolayers and mirror experiments were achieved by aligning the in-plane magnetization of the Fe surface via an external field up or down along the vertical direction, perpendicular to the scattering plane. The magnetic (LMDAD) asymmetry is defined as A_{LMDAD} $=[(I_{up}-I_{down}/I_{up}+I_{down})]$, where $I_{up}(I_{down})$ were the photoelectron spectral intensities obtained with the sample magnetization in the upward (up) or downward $(down)$ directions. In this geometry A_{LMDAD} is nonzero only if the surface magnetization has a sizable in-plane component. The sample position and the photon energy were kept fixed in order to avoid photoelectron diffraction effects.[36]

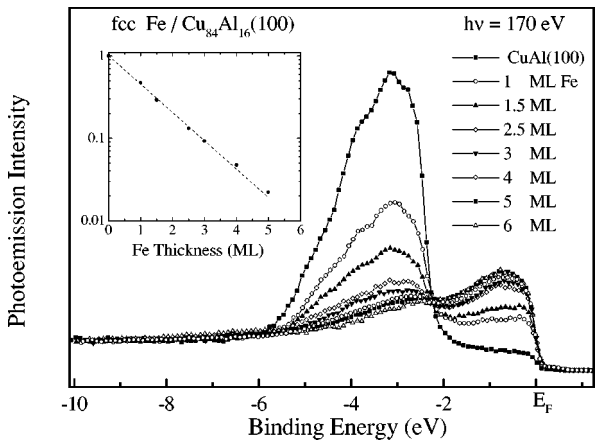

FIG. 1. SR-PES valence band spectra as a function of the Fe coverage, for the Fe/$Cu_{84}Al_{16}(100)$ system. Inset: valence band intensity decay. The result of the fitting in the inset indicates a layer-by-layer growth of the Fe films.

RESULTS AND DISCUSSION

The epitaxial growth of ultrathin Fe layers on the $Cu_{84}Al_{16}(100)$ surface at RT and 150 K was confirmed by LEED. The LEED patterns from the Fe films showed a sharp $p(1\times1)$ structure for RT growth and the same pattern with somewhat broadened spots for growth at 150 K with the same spacing as the fcc substrate. Samples grown at RT exhibited Al diffusion from the substrate through the Fe overlayers. These samples did not show magnetic dichroism either at RT or after cooling down to 150 K. The low-temperature grown films were magnetically ordered and no interdiffusion was observed within the time of the experiments. We restrict the analysis and discussion to the magnetic γ-Fe layers grown at 150 K.

Figure 1 presents the valence-band spectra taken at 170 eV photon energy for the clean $Cu_{84}Al_{16}(100)$ substrate and for different Fe coverages, deposited and measured at 150K. The spectrum of the clean substrate is dominated by the Cu $3d$ band between 5 and 2.5 eV below the Fermi level. In the spectra of the overlayers, the intensity within 1.5 eV from the Fermi level is mostly due to the $3d$ band of iron. The inset shows the intensity decay of the Cu $3d$ band of the substrate as a function of the Fe coverage. The exponential decay indicates that the γ-Fe films cover the substrate uniformly.

Figure 2 shows the Fe $3p$ LMDAD for 4 and 5 ML γ-Fe on $Cu_{84}Al_{16}(100)$, prepared and measured at 150 K with linearly polarized, monochromatic synchrotron radiation of 170 eV. The LMDAD spectra indicate in-plane ferromagnetism at these coverages. Figure 3 shows the evolution of A_{LMDAD} with Fe thickness on $Cu_{84}Al_{16}(100)$. Up to 2 ML, no magnetic order is observed in the direction specified by

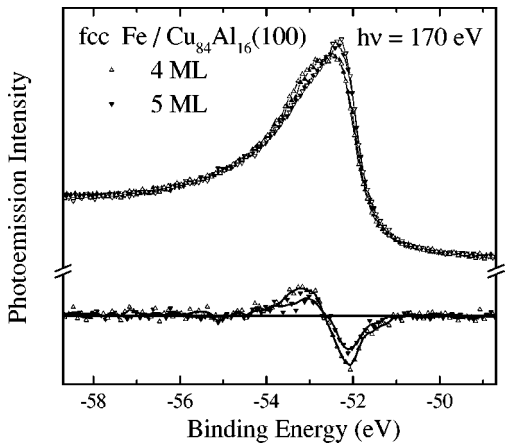

FIG. 2. Fe $3p$ LMDAD spectra for 4 and 5 ML fcc $Fe/Cu_{84}Al_{16}(100)$ prepared and measured at 150 K. The existence of magnetic asymmetries indicates in-plane ferromagnetism for the 4 and 5 ML-thick γ-Fe films. Up (down) triangles refer to upward (downward) direction of the sample magnetization with respect to the photoemission plane. Empty (full) symbols refer to the 4 (5 ML) thick iron layer.

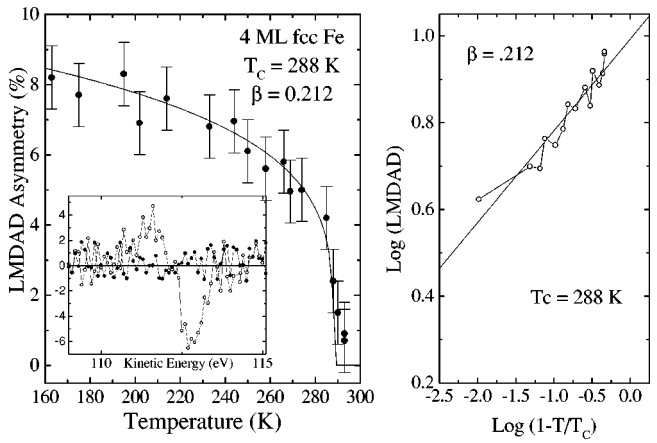

FIG. 4. Temperature dependence of the Fe $3p$ LMDAD for 4 ML fcc Fe on $Cu_{84}Al_{16}(100)$. The data are fitted by the power law $[1-(T/T_C)]^{\beta}$. The inset shows the LMDAD asymmetries at 150 K (empty circles) and 300 K (dots). The second panel shows the fitting procedure based on the power-law hypothesis, according to Dürr *et al.* (Ref. 40).

the external applied field (in-plane and perpendicular to the photoemission plane). In-plane ferromagnetism appears at 2.5 ML and A_{LMDAD} reaches a maximum value for 3–4 ML. Assuming that below 2.5 ML our Fe films present perpendicular anisotropy, this result would indicate a lower thickness threshold for the reorientation transition of the ferromagnetic easy axis than reported in the case of a Cu(100) substrate.[20,22,25,37]

The Curie temperature of the in-plane magnetized ultrathin γ-Fe phase has been determined for 4 ML thickness by measuring A_{LMDAD} as a function of temperature, as shown in Fig. 4. The experiment was performed by varying the temperature of the 150 K deposited sample between 150 and 300 K. The data are fitted by a function describing the power-law dependence of the order parameter of ferromagnetism in the neighborhood of the Curie temperature T_C.[38] The fit is obtained by maximizing the function $\log[1-(T/T_C)]$ in the region where log(LMDAD) vs $\log[1-(T/T_C)]$ is linear. This procedure yields an exponent $\beta=0.212(5)$ for the power law $[1-(T/T_C)]^{\beta}$ and a Curie temperature $T_C=288(5)$ K.

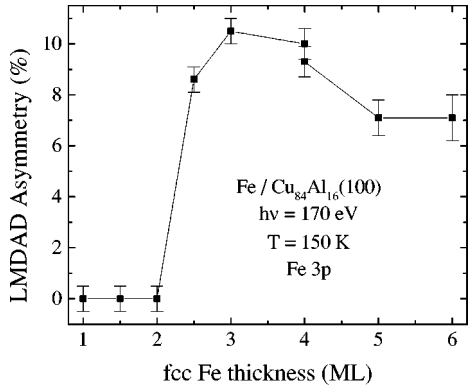

FIG. 3. Fe $3p$ LMDAD asymmetry for increasing Fe coverage on $Cu_{84}Al_{16}(100)$, indicating the onset of in-plane magnetization at 2.5 ML Fe and the phase transition between 4 and 5 ML, with 35% reduction of magnetization.

Similar T_C values have been reported for Fe/Cu(100) (Refs. 26 and 15) and Fe/CuAu(111).[11] Our β value agrees well with that predicted by the two-dimensional XY model[39] and with values measured on Fe/Au(100) (Ref. 40) and on Fe/W(100).[41] This extrapolation method for T_C allows for the existence of a deviation from the power law above a certain temperature.[40,42] The deviation is connected to the low dimensionality of the system: in quasi-two-dimensional systems the spin fluctuations related to the phase transition are very important and affect a larger temperature range near T_C than in bulk ferromagnetism. By reaching T_C from below, the magnetization may not vanish because of the formation of short-range ordered spin clusters that can show different T_C values, depending on their size.[43] The spread of the measured A_{LMDAD} values near T_C provides evidence for a limited coherence length that is connected to the high density of defects in the low-temperature grown layer. The possible onset of interdiffusion at the interface, when annealing at room temperature, may have a direct consequence on the deviation from the power-law behavior.

The analysis of the energy splitting of the Fe $3p$ core level can give insight on the *local* magnetic moment of the iron atoms in the epitaxial films. In Fig. 5 we compare the Fe $3p$ LMDAD splitting of 3 ML fcc-Fe on $Cu_{84}Al_{16}(100)$ $[\Delta E=1.10(2)$ eV] of a standard bcc-Fe(100) surface. The energy width of the dichroism spectrum represents the energy splitting of the $J=\frac{3}{2}$ multiplet due to the exchange interaction for the $3p$ core hole and the spin-polarized valence band. The splitting reflects therefore the value of the magnetic moment of the excited atom, i.e., a local property.[30,32] Recent experiments on Fe-Co and Fe-Ni surface alloys[34] have demonstrated that the width of the $3p$ core level LMDAD spectra is proportional to the *local* magnetic moment. This means that sizable changes of the iron *local* magnetic moment are reflected in changes of the splitting energy of the core hole magnetic sublevel, and consequently on the width of the LMDAD spectrum. Roughly speaking, the width of the LMDAD spectrum for the predicted low-spin ferromagnetic phase with $1.1\mu_B$ should be reduced to one-

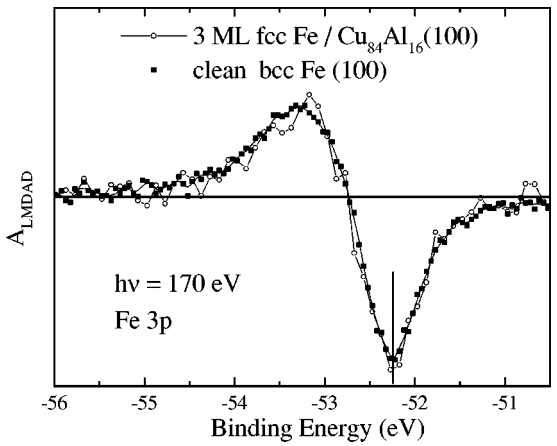

FIG. 5. Fe 3*p* LMDAD for 4 ML fcc Fe on $Cu_{84}Al_{16}(100)$ (open circles) and for bcc-Fe(100) (filled circles). The bcc-Fe(100) spectrum represents the average of a surface enhanced moment of $\sim 3\mu_B$ and subsurface and bulk contributions of $\sim 2.2\mu_B$. According to an escape depth $\lambda = 5(1)$ Å, ~30% of the photoemission intensity originates from the surface and the LMDAD lineshape of bcc-Fe(100) is equivalent to that of a homogeneous Fe film with $\sim 2.5\mu_B$.

half of the width of the LMDAD spectrum for bulk bcc Fe $(2.2\mu_B)$. From Fig. 5 we observe that the width of the LMDAD spectrum of 3 ML γ-Fe/$Cu_{84}Al_{16}(100)$ and that for the α-Fe(100) are identical within experimental accuracy. A discussion of the LMDAD signal of bcc-Fe is needed at this stage: the reported LMDAD spectra were obtained in highly surface sensitive conditions, which implies that the measured LMDAD line shapes represent the average of surface, subsurface, and substrate contributions.[34] The width of the reference bcc Fe(100) surface spectrum is determined by the surface enhanced moment, the subsurface, and the bulk moments averaged with relative weights set by the surface sensitivity of the measurement. A numerical simulation of the Fe(100) LMDAD spectrum, based on the hypothesis of linear dependence of the LMDAD width upon the magnetic moment and on an escape depth of $\lambda = 5$ Å, gives an average value of the magnetic moment of the sampled bcc iron layers of $2.5(1)\mu_B$. The enhanced values of the magnetic moment at the bcc Fe(100) surface and subsurface layers have been taken from theory ($2.97\mu_B$ for the surface).[44] It appears from Fig. 5 that the magnetic moment of the iron atoms in 3 ML γ-Fe/$Cu_{84}Al_{16}(100)$ is of the same value. If we make the hypothesis that the magnetic moment of the 3 ML-thick γ-Fe film is uniform through the layer, then our data are consistent with the formation of the theoretical high-spin ferromagnetic phase characterized by a magnetic moment of $2.5(1)\mu_B$, and with the experimental value from Gradmann and Isbert[11] larger than in bulk α-Fe. In the high-spin state of the γ-phase, iron is a strong ferromagnet, with an almost full majority 3*d* band. In these conditions, the surface enhancement of the magnetic moment should be limited to a 10% effect at most, likewise in the case of hcp or bcc cobalt.

Above 4 ML, the LMDAD asymmetry is reduced by 35%, as can be seen in Fig. 2. We could not check structural changes on our samples grown on $Cu_{84}Al_{16}(100)$, but the reduction of the LMDAD asymmetry signal is consistent with a picture known from (RT grown) Fe/Cu(100): by increasing the Fe thickness, the deeper Fe layers undergo a transformation from a distorted fcc structure towards a more compact undistorted one.[19,22,26,27] If the top layers remain in the perpendicularly expanded, large volume structure, then a magnetovolume instability in the vertical direction may explain the reduction of LMDAD asymmetry (fewer layers in the high spin state) as well as the permanence of a high magnetic moment near the surface. The dense deep layers could be in the antiferromagnetic state at sufficiently low temperature (paramagnetic at 150 K) while the surface/near-surface layers would remain in the high-spin ferromagnetic state with magnetic moments of $2.5(1)\mu_B$. For Fe/$Cu_{84}Al_{16}(100)$, the formation of an antiferromagnetically ordered phase in the deep layers for samples thicker than 4 ML is compatible with the LMDAD results, since it would add a nondichroic spectral contribution with a reduced total photoemission peak width (due to the reduced magnetic moment of Fe atoms in the antiferromagnetic phase) to the surface (ferromagnetic) dominated LMDAD spectrum. This contribution cannot severely modify the line shape of the surface sensitive Fe 3*p* spectra, but can certainly reduce the A_{LMDAD}. On the other hand, if the observed reduction of magnetization for thicknesses larger than 4 ML was due to a transition of the whole γ-Fe film to a low-spin ferromagnetic phase, as suggested by Durrand *et al.* for Fe/Cu(111),[45] then a large reduction of the LMDAD splitting should be observed, which is not the case.

CONCLUSIONS

In conclusion, we have observed in-plane ferromagnetism in ultrathin fcc-Fe(100) layers on $Cu_{84}Al_{16}(100)$ for thickness between 2.5 and 6 ML, which were prepared and measured at 150 K. The magnetization is oriented in plane at lower coverage than reported in the studies of Fe/Cu(100), perhaps as a consequence of the laterally expanded $Cu_{84}Al_{16}(100)$ lattice, which reduces the need of expanding the interlayer distance perpendicularly in order to reach the favorable atomic volume for the high-spin ferromagnetic phase, and consequently reduces the perpendicular anisotropy. The ferromagnetic fcc-Fe(100)/$Cu_{84}Al_{16}(100)$ phase has an average magnetic moment of the order of $2.5\mu_B$, as deduced by comparison with the data from a clean α-Fe(100) surface, and has a Curie temperature close to room temperature for 4 ML thickness. A phase transition is observed between 4 and 5 ML: the order parameter is suddenly reduced but the magnetic splitting of the top layers remains basically unchanged. This can be understood by analogy with the behavior established for γ-Fe/Cu(100) films of similar thickness that exhibit antiferromagnetically ordered (or paramagnetic) deep layers and surface restricted ferromagnetism.

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