PHYSICAL REVIEW B, VOLUME 65, 235435

Magnetic force microscopy study of perpendicular magnetization reorientation for Fe grown on Cu/Si(111)

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(Received 10 January 2002; published 21 June 2002)

Iron films with thicknesses ranging from 1 ML to 40 ML have been grown on a Si(111)7×7 reconstructed substrate on which a 15-ML copper layer has been previously deposited. Samples have been analyzed *in situ* by scanning tunneling microscopy and magnetic force microscopy (MFM) techniques for each step of the iron deposition. Imaging has been performed at room temperature and zero magnetic field. MFM images collected for Fe thickness lower than or equal to 4 ML showed randomly assembled magnetic domains characterized by a perpendicular magnetization. The average magnetic domain size increased with increasing Fe amount up to a thickness of 4 ML. For an Fe film of 6 ML we observed a coexistence of out-of-plane and in-plane magnetic domain structures. The reported magnetic out-of-plane–in-plane transition has been found to occur for iron thicknesses slightly greater than those reported by using electron-diffraction measurements for the structural fcc-bcc transition. For higher iron coverage the presence of domain walls indicated that the thin films are completely magnetized in the plane of the surface. An evaluation of the width of the domains wall is also given.

DOI: 10.1103/PhysRevB.65.235435 PACS number(s): 68.37.Rt, 87.64.Dz, 75.70.−i, 75.75.+a

I. INTRODUCTION

Ferromagnetic ultrathin films have attracted a lot of interest both fundamentally and in view of their technological application for magnetic recording media.[1–9] An interesting recent subfield concerns the investigations of these magnetic ultrathin films grown on silicon substrates in view of their potential applications in silicon technologies. One of the most intriguing questions is the correlation between the structural and magnetic properties of these systems. For the thermal growth of iron on a Cu(111) substrate the development of iron structural environment has been generally followed by analyzing low-energy electron diffraction (LEED) patterns. At first stages, iron has been found to grow pseudomorphically keeping the fcc crystalline phase of the substrate (γ-Fe) then, at a critical thickness, to form six bcc Fe(110) domains rotationally related in the Kurdjumov-Sachs orientation.[4,5] The critical thickness has been reported to vary from one study to another, mainly because of the different experimental growth conditions.[4,6–9] Anyway, to our knowledge, only a series of experimental studies investigated, at the same time and under the same growth conditions, the structure (by LEED), the morphology (by scanning tunneling microscopy, STM) and the magnetic behavior (by surface magneto-optical Kerr effect, SMOKE) of these films.[7,9] These studies indicated that at 3 ML (1 ML=2.08 Å) of iron deposition the typical LEED pattern due to the Kurdjumov-Sachs orientation has been recorded, thus suggesting the occurrence of the fcc-bcc structural transition. At the same time, STM images suggested that small bcc iron structures are present even at very low coverage, become significant at around 3 ML and are completely dominant at 5 ML.[8] In correspondence of the fcc-bcc structural transition the authors observed a reorientation of the magnetization axis from out-of-plane to mainly in-plane.[7,9] A similar evolution appears when iron is deposited on a Si(111) substrate, after the pre-evaporation of a 15-ML thick Cu buffer layer on the clean 7×7 reconstructed surface.[10,11] In fact, Cu thin film deposited on Si(111) relaxes towards its (111) oriented bulk structure just after a few monolayers,[12,13] and therefore it supplies a good seeding for iron deposition. Previous SMOKE and BLS (Brillouin light scattering) investigations have shown that the film magnetization tilts from the out-of-plane to the in-plane direction, between 3 and 4 ML not far from the structural fcc-bcc transition detected by LEED and Kikuchi electron diffraction.[10,11] To our knowledge, the magnetization reorientation of iron ultrathin films has never been tested by magnetic microscopy and here we report on a topographic and magnetic study on the growth of Fe thermally deposited on a Cu/Si(111) heterostructure by using LEED diffraction, STM and magnetic force microscopy (MFM) imaging. The interest in the last technique relies on the magnetic spatial resolution, which is completely lacking for the other optical methods (SMOKE and BLS). Furthermore, the existence of the perpendicular magnetization has been tested by magneto-optics at a temperature below the Curie temperature (about 60 K) of the film[10,11] when the domains size is of the order of the sample surface. MFM allowed to probe the orientation of the magnetization and get information on the surface domains and their evolution with the increasing coverage of the metal film[14] at zero field and above the Curie temperature. In this paper we have evidenced the presence of an out-of-plane magnetization up to 6 ML of Fe amount. For this Fe coverage a coexistence of both perpendicular and in-plane magnetization have been measured. Interestingly the perpendicular magnetic domain's average size increases on passing from 2 ML to 4 ML and then decreases considerably for 6 ML. For higher iron coverages a complete in-plane magnetization is clearly evidenced by MFM images.

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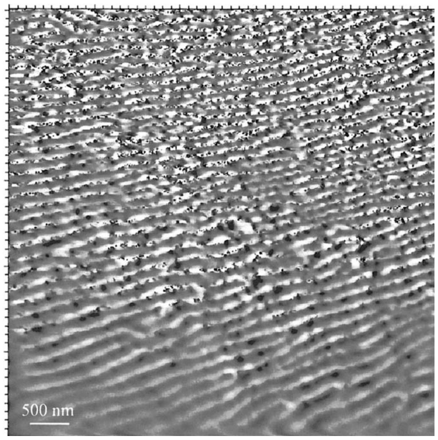

FIG. 1. MFM image (5 μm×5 μm) of 1100 ML Fe/Cu/Si(111) sample obtained by varying continuously the distance between the tip and the sample in order to show the transition between a mainly van der Waals regime (upper part of the figure, topographic and magnetic effects) to a pure magnetic regime (lower part of the image).

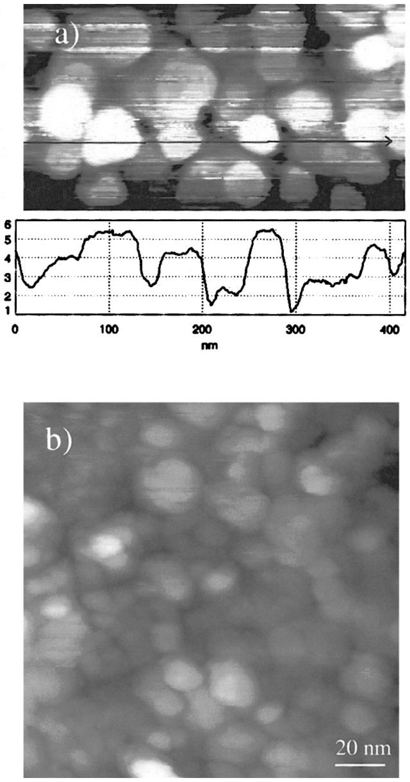

FIG. 2. (a) STM image (470 nm×250 nm) of 15 ML of Cu/Si(111), a typical line profile is shown in bottom part of the figure. (b) STM image (150 nm×150 nm) of 6 ML Fe/Cu/Si(111).

II. EXPERIMENT

All the samples have been investigated *in situ* in an ultrahigh vacuum chamber, with a base pressure of 5×10^{-10} torr. The substrate has been cut from a *p*-doped Si wafer with the (111) orientation, 1–10 Ω cm, and cleaned *in situ* by flowing a current of around 8 A through the sample. The Auger analysis showed no significant presence of contaminants such as oxygen or carbon and the LEED pattern ensured obtaining a clean surface showing a sharp 7×7 silicon reconstruction. A 15-ML Cu layer is then deposited on the substrate, in order to better match the Fe lattice parameter and thus favoring the epitaxial Fe growth without the formation of iron silicides. The thickness of the Cu layer has been chosen as the one enough to recover the Si substrate, in order that, after such a deposition, the Auger technique did not detect any appreciable signal in correspondence of the Si $L_{2,3}VV$ Auger transition. The LEED pattern showed six spots organized as an enlarged hexagon, rotated by 30° with respect to the one showed by Si. Iron is then thermally deposited from high-purity ingots contained in a tungsten basket, by controlling the thickness with an Inficon quartz microbalance. At each step of deposition, LEED diffraction, STM, and MFM imaging have been performed. We used a STM/AFM apparatus by Omicron. For the STM, a home-made tungsten tip has been used, with a gap voltage of 0.5 V applied between sample and tip and a tunneling current of 1 nA. The MFM images have been performed through the noncontact detection mode, by oscillating a magnetic tip close to the sample surface. The maximum lateral scan range of the microscope is 5 μm. The tip is a Si_3N_4 cantilever covered by 40 nm of a Co alloy. The tip magnetization should be ensured by its pyramid shape, however we preliminarily magnetized the tip in the *z* direction (i.e., perpendicular to the scanned surface) in an external magnetic field. The cantilever resonant frequency $\omega=\sqrt{k/m}$ is of 60 kHz, with *k* indicating the cantilever force constant (1.3 N/m in our case) and *m* its mass. As the tip is scanned some tens of nanometers above the sample surface, a change of the force gradient $\partial F/\partial z$, whether its origin is atomic or magnetic, produces the shift of the resonance frequency, according to the formula $\omega=\sqrt{(k-\partial F/\partial z)/m}$. The frequency shift, measured via a frequency modulation detector, is the used feedback signal. The minimum signal obtainable is of the order of one-tenth of hertz, corresponding for the cantilever used to a force gradient sensitivity of 1×10^{-6} N/m well below the typical value of frequency shift observable at a Neel or Bloch wall.[15] The MFM images have been obtained both by allowing tip vertical displacements, in order to maintain a constant force gradient and thus following the surface profile, and by keeping a constant tip-sample distance, so measuring the resonance frequency shift. The latter measurement mode is particularly useful in order to increase the tip-sample distance and to quench the short-range van der Waals interactions, thus letting the magnetic signal to prevail and producing a greater contribution to the overall signal. Finally, the tips and detection mode were checked by performing an MFM imaging on a 1100 ML of bcc (110) iron on Cu/Si(111) substrate. By varying the tip distance with respect to the sample (from 40 nm to 100 nm) we succeeded in recording the transition from the topographic to the magnetic regime (see Fig. 1, from top to bottom). In the topographic part the nanostructures constituting the iron film are clearly visible while in the magnetic part a fine stripes pattern is shown. The pattern consisted of alternating black and white stripes indicative of a normal magnetization component that alternates in sign. This is due to the fact that for iron bulk crystal the easy magnetization axis is the (100) and therefore we are

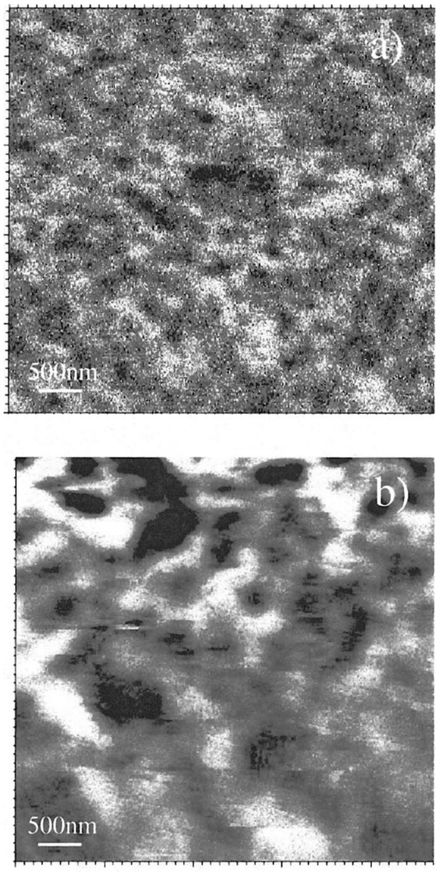

FIG. 3. MFM images (5 μm×5 μm) (a) 2 ML Fe/Cu/Si(111), (b) 4 ML Fe/Cu/Si(111).

observing a component parallel to (110) direction of this magnetization direction. The stripes are wavy and sometimes contain forks. These forks are commonly observed defects and are generated when a stripe of one sign met two other stripes of opposite sign. Moreover, we checked that the stripe's average width roughly corresponds to the iron film thickness in agreement with previous studies.[16]

III. RESULTS AND DISCUSSION

After the deposition of 15 ML of copper on the Si(111)7×7 reconstructed substrate we observed the typical LEED pattern of (1×1) hexagonal net of a twinned fcc lattice.[10,13] In Fig. 2(a) we report the STM image (410 ×250 nm) of this sample, which is formed by rather flat islands whose average lateral dimension is about 50 nm. This picture can explain the presence of the LEED pattern because the islands have an area and a flatness much greater than that of the spatial coherence of the diffracted low-energy electrons, which is normally estimated to be of the order of 5–10 nm. Furthermore, the islands appeared to be hexagonal and this can be related to the twinned growth of copper on the Si(111) substrate.[10,13] In Fig. 2(b) we report the STM image (150×150 nm) of 6 ML of iron deposited on Cu/Si(111). Islands appeared to be smaller than those reported for copper, ranging between 10 and 25 nm. At the same time the LEED pattern showed spots arranged according to the Kurdjumov-Sachs[10] orientation and a diffuse background. Interestingly, the topography of the samples with a lower iron coverage has been found to be nanostructured: the nanostructures average lateral dimension and their density increases with iron coverage. The average size of these nanostructures is comparable with that reported in the case of iron

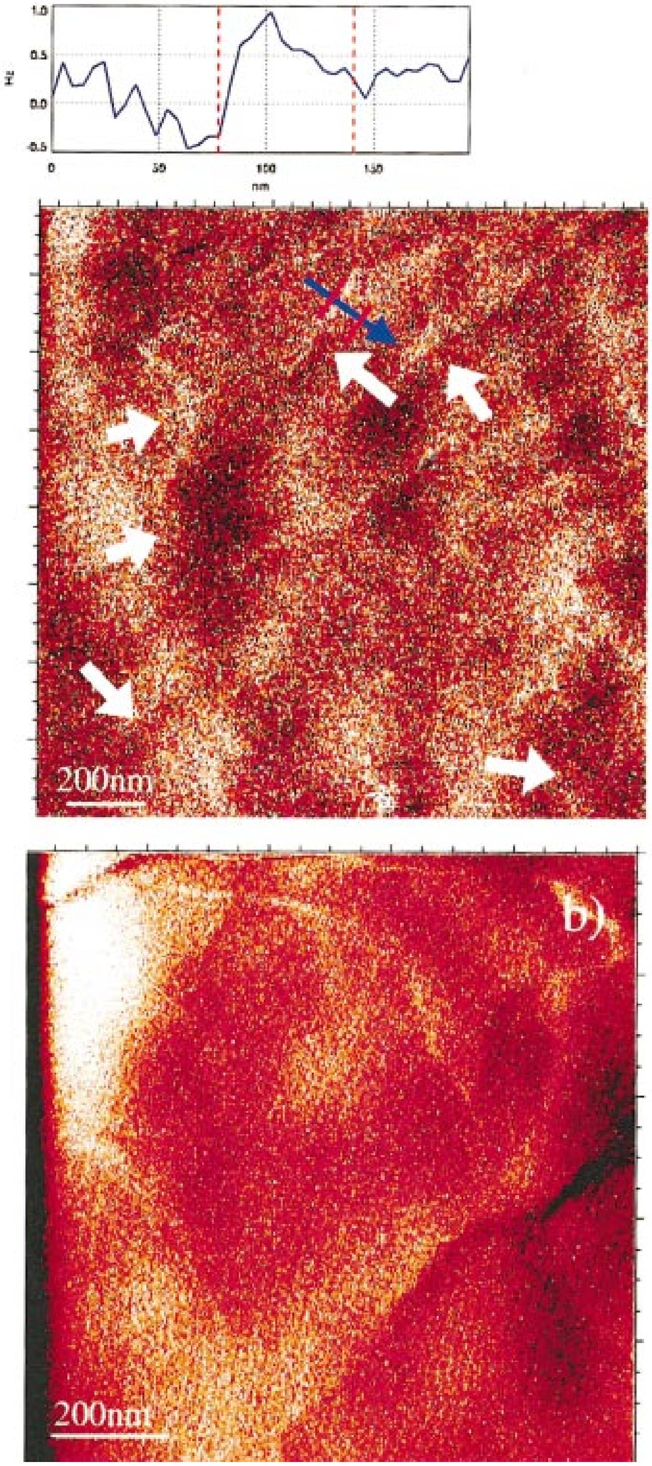

FIG. 4. (Color) (a) MFM image (1.5 μm×1.5 μm) of 6 ML Fe/Cu/Si(111) and (b) MFM image (1 μm×1 μm) of 15 ML Fe/Cu/Si(111). A typical line profile of the wavy lines indicated by the white arrows for the 6 ML Fe/Cu/Si(111) sample is shown in the upper part of the figure.

deposition on Cu(111) single crystal.[8,9] The fcc-bcc structural transition has been even observed by LEED to occur between 2 and 3 ML.[10] Figure 3(a) is a 5 μm×5 μm MFM image representing the magnetic force gradient detected above the surface of the 2-ML-thick Fe film. Since the tip is perpendicularly magnetized, dark and bright regions correspond to out-of-plane magnetized domains, whose interaction with the tip is attractive or repulsive according to the up or down direction of their magnetic moments. Domains are meanderlike and give rise to a zero net global magnetization. Their average linear size is about 250 nm. The domain contrast is of the order of 1 Hz and corresponds to a force gradient of around 10^{-5} N/m. The corresponding LEED pattern showed that the Fe growth is pseudomorphic to the Cu buffer layer, with Fe keeping its fcc structure. The lattice parameter of such a (γ-Fe) can be evaluated to be the same of copper. Figure 3(b) showed the magnetic domain structure of the 4-ML-thick sample. The image (5 μm×5 μm) indicates that the out-of-plane anisotropy still prevails in shape 1 and gives rise to perpendicularly magnetized domains. The approximate measure of a domain width is around 350 nm. An interesting feature emerges when comparing Fig. 3(a) to Fig. 3(b), namely, the domains widening. The increase of the domains size suggests the magnetization is far from the in-plane reorientation, because just before such a reorientation a domains shrinking is expected.[14] A completely different magnetic pattern appeared for the 6-ML-thick sample. The MFM image [Fig. 4(a), 1.5 μm×1.5 μm] still reveals the out-of-plane magnetized domains whose average size has now decreased to about 250 nm. Besides, also wavy lines [Fig. 4(a), white arrows] crossing the whole image are visible possibly due to the formation of a magnetic ripple structure.[14] Since the perpendicularly magnetized tip does not interact with an in-plane magnetized domain but it is anyway sensitive to regions where magnetization rotates, thus exerting a force gradient on the tip, the image contrast must be ascribed to walls of in-plane magnetized domains. The lateral dimension of such wavy lines, as measured in Figs. 4(a) and 4(b), has been found to be about 30–80 nm. A typical line profile of one of these wavy lines has been reported in the upper part of Fig. 4. Moreover, it is worth underlining that this value is not necessarily equal to the actual wall width since (a) we are at the edge of the MFM resolution, (b) an MFM image shows the distribution of the second derivative of the normal film surface component of stray field measured at some distance from the film surface. At very low film thickness (20 nm) 180-deg spin transitions can occur by the formation of Neel walls composed by spins that do change direction without leaving the plane of the surface. Nevertheless it has been reported that MFM technique can detect this Neel wall.[15] It is worthwhile to note, in addition, that the resonance frequency shift corresponding to the walls is one order of magnitude less when compared to that previously obtained for perpendicularly magnetized domains. The corresponding force gradient is only 10^{-6} N/m. In Fig. 4(b) we report the MFM image (1.0 μm×1.0 μm) of 15 ML of iron deposited on Cu/Si(111) substrate. It is easy to note that no structure from perpendicular magnetic domains is evidenced but only wavy lines ascribed to domains walls are visible. This means that for this Fe thickness a complete in-plane magnetization occurred. This picture is confirmed for all the higher iron coverage (up to 40 ML) that we investigated. The average width of the domains has been found to increase with iron coverage. Interestingly the MFM probe is very sensitive to the out-of-plane magnetization due to the preferential direction of the magnetic field of the tip while it is only indirectly sensitive to the change of the spin orientations in the in-plane magnetized domain walls. To this reason can be ascribed the capability of this technique to detect a perpendicular magnetization up to 6 ML of iron deposition, even though the other magneto-optical techniques have been reported to show a quasi-complete magnetization reorientation.[10,11] At the same time, we cannot observe the presence of the domain walls, characteristic effects of the presence of an in-plane magnetization in a MFM image, at Fe coverage lower than 6 ML because of the great contrast (frequency shift) induced by the domains magnetized perpendicular to the surface plane with respect to the others. The observed behavior of the average size of the out-of-plane magnetized domains deserves to be deepened. In fact, we measured first an increase of these spatial dimensions (up to 4 ML) and then a sizeable decrease for 6 ML. This technique, therefore, allowed us to follow the magnetization flip and indicated that the beginning of the reorientation of the magnetization in the out-of-plane domains occurred for iron thickness higher than 5 ML. The coexistence of both the magnetization for 6 ML of iron, the presence, measured by resonant magnetic scattering, of a small local in-plane magnetic moment even for Fe 1 ML (Ref. 17) together with the results of our Auger electron-diffraction analysis[18] pointing out to the occurrence of small bcc clusters even at very low iron coverage, suggested us a picture of the magnetic behavior of these systems where both the fcc and bcc structural phases are present already at very first stages of iron growth, each characterized by its own magnetic behavior. Finally, for iron coverages higher than 8 ML the complete transition to the in-plane magnetization has occurred and the magnetic in-plane domains have been found to increase their area with iron thickness. In conclusion, we collected MFM images for several iron thicknesses deposited on Cu/Si(111) substrate. The MFM technique has been shown to be very sensitive to the magnetic behavior of even 1 ML of iron and enabled us to follow the evolution of the area of these out-of-plane magnetized domains as a function of Fe coverage. Moreover, for Fe 6 ML we observed a coexistence of both in-plane and out-of-plane magnetized domains while for higher Fe deposited amount only the in-plane magnetized domains have been detected.

ACKNOWLEDGMENTS

This research has been carried out thanks to the financial support of COFIN 2000 and INFM (National Institute of Physics of Matter) SIMBRIS Advanced Research Project and MAGDOT PAIS.

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