Magnetic Moment of fcc Fe(111) Ultrathin Films by Ultrafast Deposition on Cu(111)

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Thermally deposited ultrathin Fe films on Cu(111) at room temperature or below have been known to grow in a multilayer mode with a low net magnetic moment ($\sim 0.5\mu_B$). In the present work we have used pulsed laser deposition to produce isotropic fcc Fe/Cu(111) films which grow layer by layer with a magnetic moment of more than $2\mu_B$. We attribute the larger magnetic moment of the pulsed laser deposited films to their structural perfection as seen by low energy electron diffraction and scanning tunneling microscopy. [S0031-9007(98)05386-1]

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fcc γ-Fe is known to have various magnetic structures. These include paramagnetic, antiferromagnetic, ferrimagnetic, low-moment ferromagnetic, or high-moment ferromagnetic states depending on the lattice constant [1,2]. Generally for γ-Fe a larger lattice constant favors a larger moment as well as the ferromagnetic alignment. However, the most interesting high-moment ($>2\mu_B$) ferromagnetic phase has never been experimentally observed in any isotropic fcc Fe system despite theoretical predictions [3,4]. In bulk, fcc γ-Fe has zero net moment: It can only be stabilized either above 1200 K in a paramagnetic state, or quenched as antiferromagnetic inclusions in a Cu matrix [5]. In the case of ultrathin films, a paradoxical situation arises: On the one hand, one has to epitaxially grow Fe on an fcc substrate with relatively large lattice constant to achieve large atomic volume and thus the high-moment phase of Fe; on the other hand, growing Fe on such substrates will unavoidably lead to a vertical expansion of the Fe lattice constant, which makes the films no longer fcc-like. A possible substrate for growing isotropic fcc Fe is copper due to the small lattice misfit. On the (100) surface, the high-moment phase was observed to exist up to 4 ML [6] but in this thickness region the films are tetragonally distorted [7]. The net moment of Fe films falls immediately [8] after they become fcc-like (5 to 10 ML) [7]. On the (111) surface, the Fe films appear to have near isotropic fcc structure [9] but magnetic measurements on films capped by copper [10] as well as on films supported by a stepped substrate [11] indicate that Fe possesses a significantly smaller net moment of about $0.5\mu_B$. Therefore, the very fundamental question, i.e., whether an isotropic fcc Fe system can be high-moment ferromagnetic or not, has yet to be answered experimentally.

In this Letter we demonstrate the first experimental finding of an isotropic fcc Fe system which has a high-moment ferromagnetic phase. The system is ultrathin Fe film on Cu(111) prepared by pulsed laser deposition (PLD). The PLD technique greatly increases the instantaneous deposition rate by 5 or 6 orders relative to that of thermal deposition (TD). The PLD films have an isotropic fcc structure and a layer-by-layer morphology up to 6 ML as compared to the multilayer-island morphology of the TD Fe films. Most importantly, we have observed the high-moment ferromagnetic phase in the PLD Fe films up to 3 ML. We will discuss the mechanisms supporting the high-moment phase later in the paper.

The experiments were performed in an ultrahigh vacuum (UHV) multichamber system including facilities for scanning tunneling microscopy (STM), Auger electron spectroscopy (AES), low energy electron diffraction (LEED), and magneto-optical Kerr effect (MOKE). The base pressure is better than 5×10^{-11} mbar and never exceeds 2×10^{-10} mbar during deposition. Prior to film deposition the copper(111) substrate (miscut $<0.1°$) was cleaned by cycles of Ar^+ sputtering followed by 700 K annealing until a contamination free Auger spectrum and a sharp LEED (1×1) pattern have been achieved. The substrate was then kept at 220 K and placed 100 mm away from an Fe target (99.99% purity). The output of an excimer laser with KrF (248 nm wavelength, 34 ns pulse length, typical pulse energy 270–300 mJ, and repetition rate 5 Hz) was focused onto the Fe target, resulting in an instantaneous deposition rate, i.e., the deposition rate during the duration of the plasma plume (on the order of microsecond) created by each laser pulse, of Fe on Cu(111) of about 10^6 ML/min. The film thickness was controlled by reflection high energy electron diffraction and cross-examined by STM and AES afterwards. For comparison, films on the same substrate at the same temperature were also prepared by thermal evaporation from an iron wire (5N purity) heated by *e*-beam bombardment. The magnetic properties of both types of the films were recorded by MOKE.

By pulsed laser deposition, the morphology of the Fe/Cu(111) films has been improved remarkably towards layer by layer. Figure 1 shows a side-by-side comparison of the morphology of 1.0, 2.0, and 3.5 ML Fe/Cu(111) films prepared by thermal (left column) and pulsed laser (right column) deposition. The thermally deposited Fe films have a typical multilayer morphology: The substrate has not been wetted after 2 ML of Fe deposition. All the islands are bilayer (marked as 2) or trilayer (marked as 3) high at nominal thickness of 1 ML, and contain five

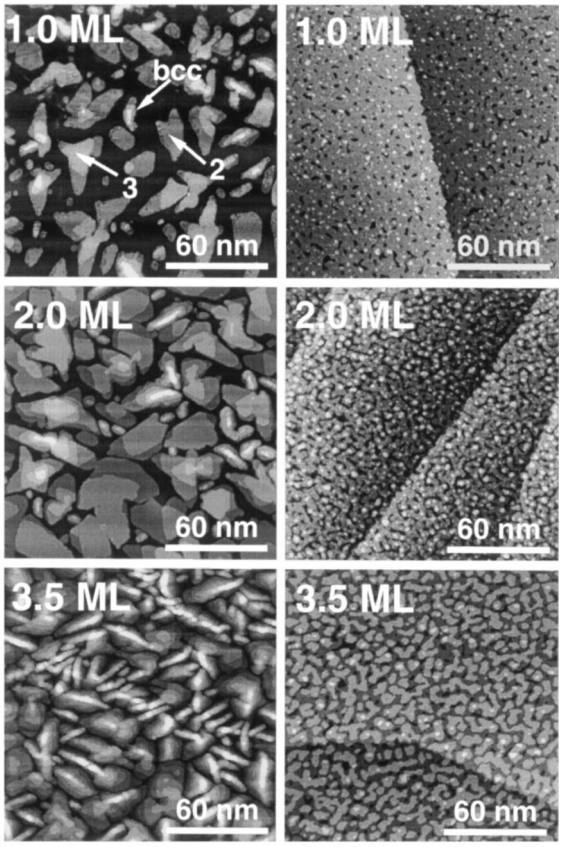

FIG. 1. STM topography images of Fe/Cu(111) films prepared by thermal deposition (left column) and pulsed laser deposition (right column). The pulsed laser deposited films show typical morphology of a good layer-by-layer growth. The thermally deposited films grow in a multilayer mode. At thickness as low as 1 ML, the thermal film is formed by bilayer islands (marked as 2), trilayer islands (marked as 3), and bcc precipitates. At 2 ML, the islands typically contain five layers in height. Note at 3.5 ML the pulsed laser deposited film stays fcc-like, while the thermal film has become mainly bcc-like.

layers at nominal thickness of 2 ML. In addition, some elongated ridgelike structures have been observed in the films. These structures are recognized as bcc(110) precipitates because they have the same elongated morphology (along ⟨011⟩ directions) as that of the thick films (>4 ML) whose structure has been determined to be bcc(110) by LEED. When the thickness increases to 3.5 ML, the bcc precipitates become dominant and the LEED pattern (not shown here) accordingly shows structure of bcc(110) with Kurdjumov-Sachs orientation. The fcc → bcc phase transformation quickly proceeds between 2 and 4 ML, above which the films have been fully transformed into bcc morphology according to our STM studies.

For the pulsed laser deposited films, the morphology is close to a layer-by-layer one. At 1 ML, more than 90% of the substrate surface has been covered by the Fe atoms, while in the 2 ML film the second layer contributes more than 85% of the total surface area. Moreover, we have obtained clear evidence that the pulsed laser deposited films have a significantly more stable fcc structure with respect to the fcc → bcc phase transformation. Figure 2 shows the interlayer spacing of the PLD films as a function of the thickness as calculated from LEED intensity vs energy curves based on a kinematic model. Below 6 ML the interlayer spacing is close to that of the substrate and above 6 ML it decreases to a distinctly smaller value. The inset LEED patterns indicate that the films are in fcc(111) structure below 6 ML and transform into the bcc(110) structure with Kurdjumov-Sachs orientation at higher thickness. Therefore, both our LEED and STM studies suggest that the pulsed laser deposited films remain *isotropic fcc* below 6 ML. Figures 1 and 2 unambiguously prove that by means of ultrafast deposition, i.e., pulsed laser deposition, the morphology of the Fe/Cu(111) films is indeed largely improved towards layer by layer, and the fcc → bcc phase transformation has been considerably delayed.

Knowing the morphology and structure of the films, we now compare the magnetic properties of thermally and pulsed laser deposited films. As the present Letter is focused on the magnetic moment, we will concentrate here only on the saturation magnetization while we leave most of the magnetic properties such as Curie temperature, magnetic anisotropy, and the spin reorientation to a forthcoming paper [12]. The saturation magnetization was obtained from saturated MOKE hysteresis loops, which were measured *always* in the same polar geometry

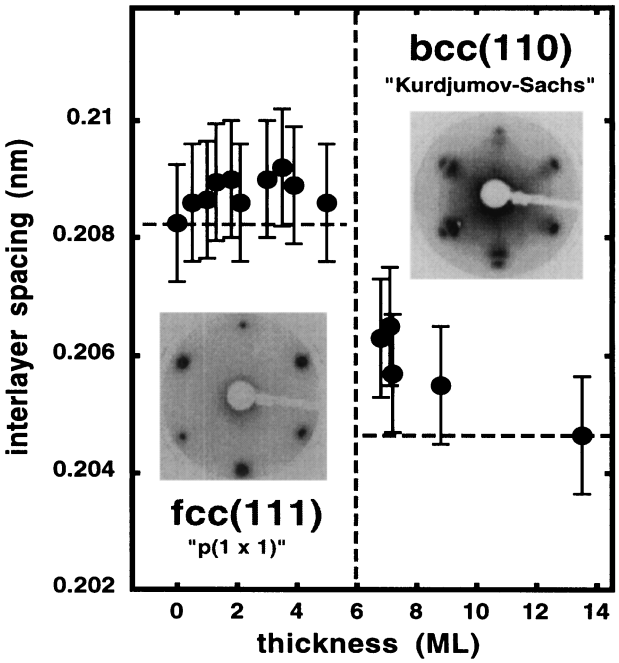

FIG. 2. Interlayer spacing of the pulsed laser deposited Fe/Cu(111) films as a function of thickness. The inset pictures are typical LEED patterns corresponding to films below and above 6 ML, respectively. The decrease of the interlayer spacing at 6 ML is consistent with the LEED patterns indicating a structural transformation from fcc(111) to bcc(110).

irrespective whether the easy magnetization axis is parallel or perpendicular to the film surface. Typical hysteresis loops of both thermally and pulsed laser deposited films at 1.6 ML are shown in Fig. 3. The two loops, measured at 40 K, differ strongly. The PLD loop has a well-defined rectangular shape and a very small coercivity of 135 Oe. The TD loop is significantly tilted with a large coercivity of 1150 Oe, reflecting the imperfection of the TD films as shown in Fig. 1. In this respect, the very small coercivity and rectangular shape of the hysteresis of the PLD film is yet another proof of its structural perfection.

The most remarkable difference, however, is the saturation magnetization. At the same nominal thickness of 1.6 ML, the saturation magnetization of the PLD film is almost 4 times larger than that of the TD film. In Fig. 4 the saturation magnetization is displayed as a function of thickness. Here all the data points, except two points for the PLD films below 1.5 ML, were acquired at 150 K by liquid N_2 cooling instead of 40 K by liquid He cooling. Since the Curie temperature of both PLD and TD films is above 200 K [12], 150 K turns out to be sufficiently below the Curie temperature where the magnetization falls rapidly. Below 1.5 ML, the Curie temperature of the PLD films is lower than 150 K; therefore, the two data points were measured at 40 K. It is also important to note here that we consider the measured Kerr intensity, no matter from the fcc or transformed bcc films, to be proportional to the magnetization [11].

For the thermally deposited films in Fig. 4, their magnetization is strongly correlated with the fcc → bcc phase transformation. In the thickness range where the fcc phase is dominant (<2 ML), the magnetization of the films has low values increasing near linearly with film thickness. Between 2 and 4 ML, the magnetization increases steeply, in accordance with the rapid progress of the fcc → bcc phase transformation in this region. In the bcc region (>4 ML), the magnetization of the films again rises proportionally with thickness but is nearly 4 to 5 times larger than the expected values based on the linear extrapolation from the fcc films (<2 ML). This means that by thermal growth, the net magnetic moment of the fcc films is about 4 to 5 times smaller than that of the bcc films. Assuming the typical moment of $2.2\mu_B$ for the transformed bcc films, we estimate the net moment of the TD films in the fcc regime to be about $0.5\mu_B$, which is consistent with previous results from the copper capped films [10] and the films supported by a stepped substrate [11].

For the pulsed laser deposited films, two different regions can be distinguished in Fig. 4. Below 3 ML (region I), the magnetization increases linearly with increasing thickness, reaching its maximum at about 3 ML. The magnetic moment in this region appears to be close to that of the transformed TD bcc films, i.e., $2.2\mu_B$ per atom. This moment value corresponds to the high-moment ferromagnetic phase since it is clearly larger than the net moment of any other magnetic structure predicted by the theories [1,2]. Above 3 ML (region II), the magnetization falls abruptly to 30% to 40% of that of the 3 ML film. Upon further thickness increasing, the

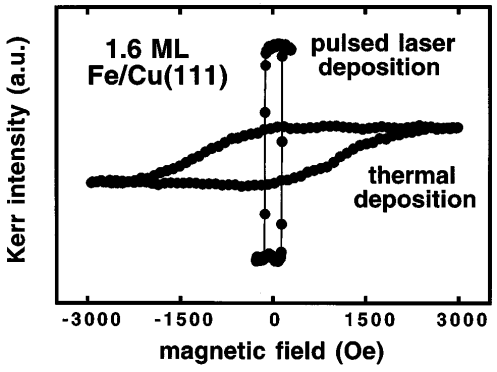

FIG. 3. Comparison of Kerr hysteresis loops of a 1.6 ML Fe/Cu(111) film prepared by thermal deposition and pulsed laser deposition. The loops were measured at 40 K. The saturation signal of the pulsed laser deposited film is about 4 times larger than that of the thermal film. The former has also a square shape with a much smaller coercivity reflecting the improved morphology shown in Fig. 1.

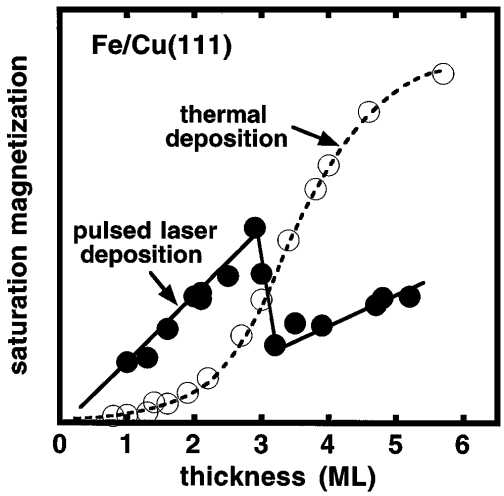

FIG. 4. Saturation magnetization as a function of the thickness of the thermally (open circles) and pulsed laser deposited (full circles) Fe/Cu(111) films. The thermal films have low magnetization below 2 ML ($0.5\mu_B$ per atom per monolayer). Between 2 and 4 ML, their magnetization increases quickly, in accordance with the rapid progress of the fcc → bcc phase transformation in this region. In the bcc region (>4 ML), the magnetization of the films is nearly 4 times larger than the expected values based on the linear extrapolation from the fcc films (<2 ML). For the pulsed laser deposited films, the magnetization initially increases linearly at a rate of about $2\mu_B$ per atom per monolayer reaching the maximum at about 3 ML. Above 3 ML, the magnetization first falls down to 30% to 40% of that of the 3 ML film, and then linearly increases with a slope of about $0.7\mu_B$ per atom per monolayer. Note that at a given nominal thickness below 3 ML, the magnetization of the pulsed laser deposited films is strongly enhanced, up to a factor of 4 to 5, as compared to that of the thermal films.

magnetization again rises linearly with a 3 times smaller slope than the initial slope below 3 ML. We estimate the net moment of the films in region II to be about $0.7\mu_B$ per atom averaged over the film.

As mentioned, the transition from low-moment to high-moment phase in the TD Fe films is the direct result of the fcc → bcc phase transformation in this system. For the PLD films, the structural origin of the abrupt decrease of the net magnetic moment around 3 ML, however, appears to be less clear. Figure 2 indicates that neither a structural transformation nor a distinct change of the lattice constant occurs around 3 ML thickness. In this respect, the PLD Fe/Cu(111) system has to be distinguished from the well-studied TD Fe/Cu(100) system, where an abrupt drop of the magnetization has also been observed around 4 ML thickness [8]. The latter is generally considered as a result of the structural relaxation from fct to fcc which apparently does not occur in the PLD Fe/Cu(111) case.

An obvious question is as follows: Why do the thermally grown Fe/Cu(111) films have a low net moment in the first few monolayers? Two possible mechanisms may be responsible for the small moment. First, there exists two additional paths for the strain relaxation in the TD Fe films: (1) via the edge atoms [13] of the multilayer island and (2) via the bcc precipitates [14]. Therefore strain relaxation is more likely to occur in the TD Fe films than in the PLD films, which could result in a smaller atomic volume, thus a smaller moment, for the TD films. Second, the thermally grown Fe films may have mixed significantly with copper due to the high diffusivity of the (111) surface. The net moment of Fe will be reduced by copper diffusion: In an extreme case, the films become nonmagnetic when deposited at 370 K where a significant interdiffusion occurs [15]. The pulsed laser deposited films, however, are less likely affected by copper diffusion because of the fast deposition rate.

The finding of the high-moment phase for the pulsed laser deposited Fe/Cu(111) films (<3 ML) has also clearly demonstrated that it is not essential for the fcc Fe films to expand tetragonally to stabilize the high-moment phase. Both the tetragonally expanded Fe/Cu(100) films and the isotropic Fe/Cu(111) films have high-moment ferromagnetic phase at low thicknesses, but the transition from the high-moment to a low-moment phase has different origins in the two systems. The instability of the moment of fcc Fe may be understood in the following way: If changes of the lattice constant do occur, as in the Fe/Cu(100) system, the magnetic moment of the fcc Fe films will change accordingly; but even if the lattice constant remains unchanged, the magnetic moment of the film as a whole will still be changed if the thickness is high enough and the influence of the Fe/Cu interface is low enough. Our experimental data (Fig. 4) suggest that in the fcc Fe(111) films the transformed phase is a low-moment ferromagnetic or ferrimagnetic phase in contrast to the antiferromagnetic phase in the fcc Fe(100) films. The determination of the detailed magnetic structure of the Fe/Cu(111) films, however, needs theoretical studies using *ab initio* calculations.

In summary, we have successfully modified the morphology of the Fe/Cu(111) films by increasing the deposition rate by about 5 to 6 orders of magnitude using pulsed laser deposition. The pulsed laser deposited films (<3 ML) have a significantly enlarged magnetic moment indicating that the isotropic fcc Fe/Cu(111) ultrathin films are high-moment ferromagnetic.

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