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In-plane anisotropy and reversed spin reorientation of fcc Fe ultrathin films on Cu(100) by pulsed-laser deposition

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Abstract. – Fe ultrathin films on Cu(100) have been prepared by pulsed laser deposition (PLD). These films show good layer-by-layer morphology and an almost ideal fcc structure at all thicknesses below 10 ML. Above 10 ML the films transform into bcc(110) structure. The magnetization of the PLD Fe films lies in-plane at low thickness (2 to 5 ML), and undergo a reversed spin reorientation switching to perpendicular at higher thickness.

Among magnetic ultrathin-film systems, Fe/Cu(100) is the most extensively studied system in the recent years [1-8]. This interest arouse since it appeared possible to study the magnetism of the thermodynamically unstable γ-Fe phase at room temperature. Without the stabilization by the fcc Cu substrate, bulk γ-Fe is stable only above 1200 K, *i.e.* in the paramagnetic state, or as inclusions in a Cu matrix which were found to be antiferromagnetic [9]. However, the Fe/Cu(100) films, if prepared by the widely used thermal deposition (TD) method, have been observed to show rather complex structural and magnetic properties. At low thickness ($<$ 5 ML), the Fe films are ferromagnetic with high moment ($\sim 2.5\mu_{\mathrm{B}}$) [6], though their structure is distinctly distorted with respect to the fcc structure of the copper substrate [10,11]. The distortion manifests itself in a tetragonal expansion of the interlayer spacing and in the buckling of the atoms in both lateral and vertical directions. At higher thickness (between 5 and 11 ML), the structure of the whole films except the top one or two layers becomes fcc-like showing no obvious distortion [3]. Meanwhile the films undergo a structural phase transformation from fcc to bcc which are characterized by the formation of $\langle 011 \rangle$-oriented ridge-like structures [2].

According to the previous knowledge from the studies of the TD Fe/Cu(100) films, Fe is either high-moment ferromagnetic with fct structure [1, 6], or antiferromagnetic when the structure is fcc-like [4]. However, a recent study of Fe films with fcc-like structure grown on Cu(111) by pulsed-laser deposition (PLD) [12] indicates that the Fe films have a high-moment ferromagnetic phase at low thickness ($<$ 3 ML). It is of obvious interest to study whether

a fcc-like structure can be achieved in Fe/Cu(100) films when prepared by PLD, and more importantly, whether the fcc-like Fe can be ferromagnetic with high moment.

In this letter we present the first experimental data on the structure and magnetism of the PLD Fe/Cu(100) films. Neither tetragonal distortion nor atomic buckling has been observed in such films. These fcc-like PLD Fe films do possess high-moment ferromagnetic phase at low thickness (< 4 ML). Their characteristic magnetic features include in-plane magnetization at low thickness (2–5 ML), and a reversed spin reorientation from in-plane to perpendicular around 5–7 ML.

The PLD Fe/Cu(100) films were prepared in a multi-chamber ultrahigh vacuum (UHV) system with a base pressure $< 5 \times 10^{-11}$ mbar and less than 2×10^{-10} mbar during deposition. Prior to film deposition the copper substrate (miscut $< 0.1^\circ$) was cleaned by cycles of Ar^+ sputtering followed by 600 °C annealing until a contamination free Auger spectrum and a sharp LEED (1×1) pattern have been achieved. The substrate was then kept at room temperature $(300 \pm 5$ K) and placed 100 mm away from an Fe target (4N 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 rate of Fe deposition on Cu(100) of about 10^5 ML/min, *i.e.* a value which is nearly 5 to 6 orders of magnitude larger than during thermal deposition of Fe. The film thickness was controlled by reflection high-energy electron diffraction (RHEED) and cross-examined by scanning tunneling microscopy (STM) and Auger electron spectroscopy (AES) afterwards.

Compared to the thermally deposited Fe/Cu(100) films, the PLD Fe films have a more ideal 2-dimensional morphology, particularly in the initial stages of growth. For example, at the nominal thickness of 1 ML, 2nd layer islands contribute by 5% for PLD films, while they contribute by 20% of the total area for TD Fe films. The average island size of the PLD films is smaller than that of the TD films due to the high flux. In terms of the fcc-to-bcc structural transformation, the TD and PLD films are rather similar: bcc-linked features start to appear only at thickness above 4 ML in both films. The detailed morphology study, due to the space limit, will be presented in a forthcoming paper [13].

The structure of the PLD Fe/Cu(100) films is fcc-like without distinct tetragonal distortion at all thicknesses below 11 ML. As an example, in fig. 1 we show the IV-LEED spectra of the (00) beam of a 3 ML PLD Fe/Cu(100) film (upper panel) and a 3 ML TD Fe/Cu(100) film (lower panel) recorded at room temperature [14]. For the PLD films, no distinct changes of the IV-LEED curves have been observed at lower temperatures (down to 145 K). The primary beam is about 6° off the surface normal lying in the (001) plane. The inset pictures are the corresponding LEED patterns taken at 105 eV with normal incidence. The *I-V* spectrum of the TD film exhibits two families of characteristic peaks, which correspond to those of fct (dashed lines) and fcc (solid lines), respectively. The fcc peaks are located at the same energy positions as those of the copper substrate, and their intensity decreases with increasing thickness. These findings, together with previous LEED studies [10,11,15], consistently suggest that the fcc peaks are largely contributed from the copper substrate, while the fct peaks stand for the tetragonal structure of the Fe films. A full dynamic IV-LEED calculation [3] further indicates that the Fe film, prepared by TD, is not only vertically expanded, but also atomically buckled in both lateral and vertical directions. As a result of the surface buckling, the LEED pattern exhibits a complicated (5×1) superstructure as viewed in the LEED picture in fig. 1. The PLD films, remarkably, do not show signs of similar atomic buckling, as the LEED pattern of the PLD film is that of a sharp (1×1) diffraction pattern, which has been observed for all films (2 to 10 ML) at room temperature and below (down to 145 K). In fig. 1 the background of the LEED pattern of the PLD films is somewhat higher than the TD films, which may be due to the significantly smaller island size of the PLD films. Furthermore, except for peaks

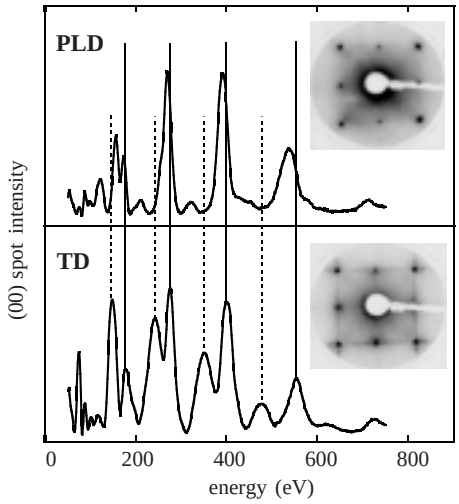

Fig. 1

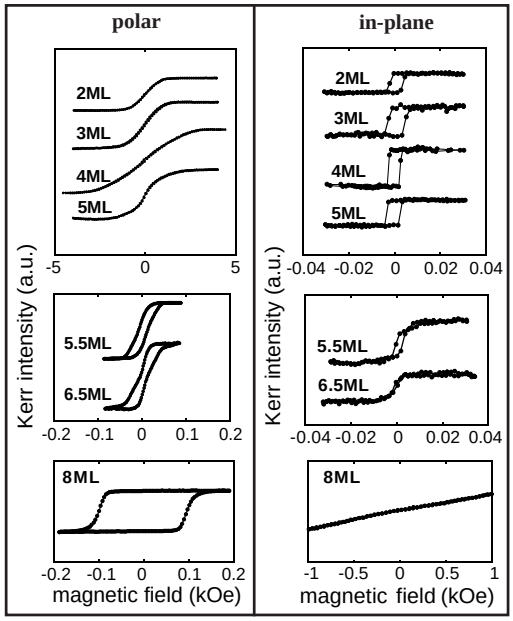

Fig. 2

Fig. 1. – Low-energy electron diffraction data from two 3 ML thick Fe films on Cu(100), prepared by pulsed-laser deposition (upper panel) and thermal deposition (lower panel). Inset pictures are the LEED patterns taken at 105 eV. The curves give the intensity of the specular beam as a function of the energy at near-normal incidence. Both fct (dashed lines) and fcc (solid lines) families of peaks are visible in the thermal deposition film, while in the pulsed-laser deposition film only fcc peaks are present. The LEED pattern of the pulsed-laser deposition film is (1×1), whereas the thermal deposition film has a (5×1) superstructure.

Fig. 2. – Kerr loops of pulsed-laser–deposited Fe/Cu(100) films at various thicknesses. With increasing thickness, the polar loops (left column) become easy-axis like and the in-plane loops (right column) become hard-axis like. Note that the scales of magnetic field are different and the units of Kerr intensities are different for polar and in-plane magnetization.

at low energies, only one family of peaks corresponding to the fcc structure exists in the *I-V* spectrum for PLD films. In fact, in the thickness range from 2 to 10 ML, the IV-LEED curves and the LEED patterns of the PLD films closely resemble the one shown in the upper panel of fig. 1. The existence of some fct-like peaks at low energies may imply that the topmost layers of the PLD Fe film are still expanded to some extent, though a full dynamic analysis is necessary for clarifying this. Nevertheless, the vanishing of both the fct peaks and the superstructure LEED patterns strongly suggests that the PLD Fe/Cu(100) films have a nearly isotropic fcc structure between 2 and 10 ML. Above 10 ML, the structure of the PLD films transforms into bcc(110), in close agreement with TD Fe films [10]. The obvious question, *why* PLD films differ structurally that strongly from TD films, goes beyond the scope of the present letter and is discussed in a future publication [13].

These PLD Fe/Cu(100) films show remarkably different magnetic properties as compared to the TD films. Figure 2 shows typical MOKE hysteresis curves of the PLD films with various thicknesses at 145 K. In contrast to the perpendicularly magnetized TD Fe films, between 2 and 5 ML the PLD Fe films have a clear in-plane easy magnetization axis as characterized

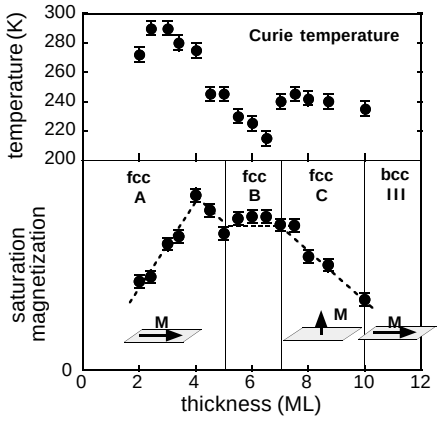

Fig. 3. – Plots of Curie temperature (upper panel) and saturation magnetization M_s (lower panel) *vs.* thickness of the pulsed-laser–deposited Fe/Cu(100) films. In the M_s plot, four regions can be distinguished: the pulsed-laser–deposited films in A, B, and C show remarkably different magnetic behavior from the thermally deposited films [1]. Both types of films in region III are bcc films.

by the rectangular in-plane Kerr loops and the hard-axis perpendicular loops. Above 5 ML, the films start to experience a reversed spin reorientation. Polar loops with finite remanent magnetization (M_r) start to appear in the 5.5 and 6.5 ML films, and the M_r/M_s ratio of their in-plane loops becomes distinctly smaller than 1. In the transition thickness region (between 5 and 7 ML), the M_r values of both polar and in-plane loops are far less than 100% saturation magnetization (M_s). Whether the films have a canted easy magnetization axis or coexisting in-plane and perpendicular domains remains to be examined by further studies. When increasing the thickness over 7 ML, a well-defined perpendicular easy magnetization axis is established in the films, as indicated by the rectangular polar loop of a 8 ML film in fig. 2.

The anisotropy behavior has been summarized in the phase diagrams (fig. 3 bottom) of M_s *vs.* thickness. The M_s values were always taken from the well-saturated polar Kerr loops, irrespective of the direction of the easy magnetization axis. Four regions, denoted as A, B, C, and III, can be distinguished in the M_s *vs.* thickness plot. The division of regions A, B, and C is remarkably different from the previous division [1], *i.e.* I (2–5 ML) and II (5–11 ML), in the TD Fe/Cu(100) films. In region A (2 to 5 ML), M_s increases near linearly to the maximum at about 4 ML, and then drops to about 80% of the maximum when reaching 5 ML. The easy magnetization axis is in-plane in this region. In the transition region B (5 to 7 ML), M_s is nearly constant and close to that of the 5 ML film. In the perpendicular magnetized region C (7 to 10 ML), M_s starts to decline, resulting in a nearly equal saturation magnetization value of the 10 ML film to that of the 2 ML film. Above 10 ML, as mentioned earlier, a fcc to bcc structural transformation proceeds rapidly, leading to an in-plane easy magnetization axis which is similar to the magnetic behavior of TD Fe films in region III (> 11 ML). We therefore also denote the bcc region in the PLD films to be III for comparison to the TD films.

The thickness dependence of the Curie temperature (T_c) of the PLD Fe films is shown in the upper panel of fig. 3. At 3 ML T_c hits the maximum of about 290 K, which is significantly lower than the Curie temperature of the 3 ML TD film (370 K) [1]. Above 3 ML T_c starts to decrease with increasing thickness. The minimum T_c value, *i.e.* 210 K, is reached for films in the transition region B. In the perpendicular region C, the T_c is almost constant around

240 K. T_c is either too low (< 150 K) below 2 ML or too high (> 500 K) above 10 ML to be measured in the present MOKE setup, and thus is not included in the phase diagram.

The different magnetic behavior between the TD and PLD films again indicate the complex and rich magnetic structures of fcc Fe. The initial linear increase of M_s suggests that the films have a ferromagnetic phase below 4 ML. The measured Kerr intensities of the PLD and TD Fe films are practically the same (not shown here) indicating that the PLD fcc-like films (< 4 ML) have also a high-moment ferromagnetic phase. The decrease of M_s above 4 ML is an indication that the Fe films are no longer uniformly magnetized. Recent theoretical calculations [16, 17] have demonstrated a large amount of possible AFM-type spin configurations of the Fe/Cu(100) films at higher thickness (> 4 ML). However, the suggested AFM structure would result in a considerably smaller net magnetization than the experimental values between 5 and 7 ML, and is particularly inconsistent with the fact that the M_s continuously decreases with increasing thickness from 7 to 10 ML.

In the following we provide a plausible explanation for the inconsistency between the theory and the experiment. While the theory assumes an unchanged fcc structure for all film thicknesses, the lattice constant of the experimental films may be changed by increasing thickness, because above 4 ML the films start to experience a fcc-to-bcc structural transformation. The transformed bcc precipitates have a larger lattice constant and tend to partially accommodate the strain in the films. As a result, a portion of the films may relax to bulk-like fcc, *i.e.* nonpseudormorphic, which is known to be nonmagnetic above Néel temperature (67 K in case of bulk). Because the fcc-to-bcc transformation initially proceeds very slowly, the portion of the nonpseudormorphic part of the Fe films varies little explaining the rather constant M_s between 5 and 7 ML. At higher thickness the transformation proceeds rapidly leading to a larger and larger portion of Fe films to become nonpseudormorphic. This explains why the M_s of the films decreases with increasing thickness from 7 to 10 ML. It is not clear whether the transformed bcc precipitates are magnetic or not, but they do not contribute to the measured M_s because our applied magnetic field is not large enough to saturate bcc films (> 5 ML) in the perpendicular direction.

A similar mechanism can be used to solve the puzzle why the fcc-like PLD films ($<$ 4 ML) have high-moment ferromagnetic phase while the fcc-like TD films (5–11 ML) are antiferromagnetic. According to our STM studies the PLD films (< 4 ML) is free of bcc structures while the TD films (5–11 ML) contains bcc precipitates which tend to accommodate the strain in the film. Thus part of the TD films (5–11 ML) should relax to have bulk lateral lattice, *i.e.* nonpseudormorphic. Because full dynamic LEED calculation indicated that the topmost layers are still fct-like and thus strained, it is likely that the fcc-like inner layers are actually nonpseudormorphic. The PLD films (< 4 ML), on the other hand, should adopt the lateral lattice constant of copper, which is slightly larger than that of the bulk fcc Fe. Under these assumptions, it is straightforward to understand that the TD films (5–11 ML) have an antiferromagnetic phase which is similar to the bulk fcc Fe, while our studies indicate that the pseudormorphic fcc Fe/Cu(100) has a high-moment ferromagnetic phase.

The most astonishing finding is the in-plane magnetization in region A, as all the TD Fe films (< 11 ML) exhibit clear perpendicular magnetization. Concerning the anisotropy, it remains a matter of dispute in the calculations whether the positive surface anisotropy is large enough to overcome the shape anisotropy. Ujfalussy *et al.* [18] have found for the FM Fe films that the surface anisotropy is too small to overcome the shape anisotropy at any thickness. This is consistent with the measured in-plane magnetization below 5 ML (fig. 2), where the films are in FM state. Results by Lorentz and Hafner [16], however, predict perpendicular magnetization for the FM Fe films. For the AFM films things seem to be more consistent, both groups predicting the direction of the moment to be perpendicular to the surface. This

could be the reason for the spin reorientation in region B, in which the films start to transit from FM phase to some type of AFM phase.

Apparently the present study suggests that the magnetism of the already complex Fe/Cu(100) system is far away from being thoroughly understood. More detailed structural characterization is required to achieve a comprehensive understanding of the correlation between the structure and magnetism of the PLD Fe/Cu(100) films.

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