Comparison of pulsed laser deposition and thermal deposition: Improved layer-by-layer growth of Fe/Cu(111)

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In the search for a correlation between the magnetism and the microstructure of ultrathin films, straightforward layer-by-layer growth is desirable. The thermal deposition of Fe onto Cu(111), however, does not result in this growth mode. In this letter, we compare the initial growth of Fe on Cu(111) prepared by pulsed laser deposition (PLD) with thermally deposited Fe/Cu(111) using scanning tunneling microscopy (STM). In PLD, from the beginning there is two-dimensional nucleation and growth, in contrast to the initial bilayer nucleation and growth found for thermal deposition. Therefore, it is shown by STM that PLD grown films exhibit greatly improved layer-by-layer growth. The different experimental results are interpreted in terms of the very high deposition rate during PLD. © *1996 American Institute of Physics.* [S0003-6951(96)01448-9]

The deposition of Fe on Cu single crystals allows the room temperature stabilization of γ-Fe, an fcc-Fe phase that promises interesting magnetic properties but usually only exists in bulk above 1186 K. While there has been a lot of attention regarding the preparation and characterization of the Fe/Cu(100) system,[1] comparably few publications have shown an effort to grow Fe on the Cu(111) surface.[2–10] Although similar thermal deposition techniques were applied, these reports give contradictory information regarding the growth mode of Fe/Cu(111): while layer-by-layer growth of Fe on Cu(111) was concluded from low-energy electron diffraction (LEED) oscillations and Auger electron spectroscopy (AES) studies,[2–7] scanning tunneling microscopy (STM) observations[8,9] and x-ray photoelectron forward scattering experiments[10] demonstrated that, instead, a 3D growth behavior exists. Pulsed laser deposition (PLD) was investigated as an alternative deposition technique for Fe on Cu(111). PLD has not played a major role in the study of magnetic ultrathin films. Its main advantages are the experimental simplicity, as the laser light is coupled into the vacuum chamber through a viewport, and the possibility to achieve a congruent deposition of virtually any target material.[11] In this letter we present STM images of the initial stages of the growth of Fe/Cu(111) prepared by both thermal deposition and PLD and discuss the observed differences in the growth modes.

Both deposition techniques were performed in a multichamber ultrahigh vacuum (UHV) system with a base pressure $\leq 7 \times 10^{-9}$ Pa and a pressure $\leq 4 \times 10^{-8}$ Pa during deposition, holding the sample at room temperature, i.e., at (300 ± 5) K. Prior to the evaporation, the Cu(111) substrate was subjected to cycles of 1 keV Ar$^+$ sputtering and annealing at 700 K until clean AES spectra and sharp LEED spots were obtained. For the thermal deposition, an Fe wire of 99.999% purity was heated by electron bombardment to deposit Fe at a rate of about 0.4 ML/min onto the Cu(111) crystal. For PLD the output of an excimer laser[12] with KrF (248 nm wavelength, 34 ns pulse length, max pulse energy 600 mJ, max repetition rate 30 Hz) was focused onto an Fe target of 99.99% purity resulting in a fluence of about 5 J/cm^2 to deposit Fe onto the Cu(111) substrate, 100 mm away

from the target. Samples were then transferred under UHV to a STM chamber for topographic characterization and afterwards to an analysis chamber for LEED and AES.

The initial stages of growth of Fe on Cu(111) by thermal evaporation, as seen in Fig. 1, show a 1-ML high decoration of the Cu terrace edges, islands of 2 and sometimes 3 ML height on the terraces, and 1-ML-deep extended holes in the Cu substrate. This is in agreement with Refs. 8 and 9. The mechanism of the hole formation is described in Ref. 13. A height profile along the white line indicated in Fig. 1(a) is given in Fig. 1(b). Higher coverages result in a continuous decoration of the step edge and an increasing size of the 2- and 3-ML-high islands. There are no 1-ML-high islands observed in the initial stages of the growth of Fe/Cu(111) by thermal evaporation. Figures 1(c) and 1(d) show STM images of Fe thermally deposited onto a Cu(111) crystal with an average step width of approximately 10 nm. The 1-ML-deep holes in the Cu substrate coalesce and form larger grooves resulting in an increase of the overall surface roughness.

The features of the initial growth of Fe on Cu(111) as prepared by PLD are shown in Fig. 2. It is remarkable at first glance that there is a complete absence of the decoration effect observed in thermal evaporation. There is also no indication for a hole formation in the Cu(111) substrate. At a low coverage [Fig. 2(a)] there are both small islands approximately 1 nm in diameter and large islands about 10 nm in diameter, but all of 1 ML height and randomly distributed over the terraces. The extended shape of some islands and the irregularity of the step edges in Fig. 2(b) suggest that islands are connecting with each other and with Cu steps. Thus, the initial growth of Fe on Cu(111) by PLD is clearly in the layer-by-layer mode. As seen from Fig. 2(c), at a coverage of around 0.4 ML the second Fe layer starts appearing on top of the 1-ML-high islands. Figure 2(d) shows a height profile along the white line indicated in Fig. 2(c). At higher coverages there is still a large number of smaller islands ($\sim$1 nm in diameter) which indicate that there is no nucleation saturation. This is unlike thermal evaporation where incoming atoms would nearly exclusively grow on already present islands resulting in a comparably small number of larger
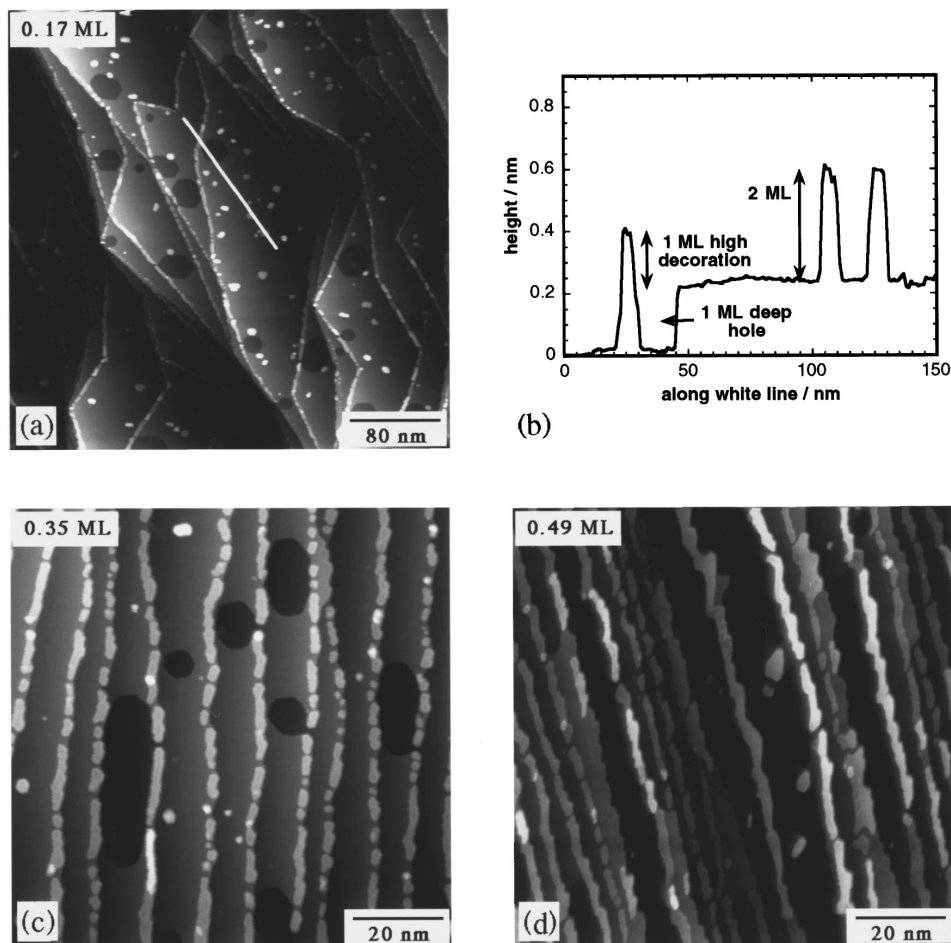
FIG. 1. Fe on Cu(111) deposited by thermal evaporation at 300 K demonstrating the characteristic features of the growth: 1-ML-high decoration of the Cu terrace edges, 2- and 3-ML-high islands in the terraces, and 1-ML-deep extended holes in the Cu substrate. (a) 0.17 ML Fe/Cu(111), (b) height profile along the white line indicated in (a), (c) 0.35 ML, and (d) 0.49 ML Fe on Cu(111) with an average step width of approximately 10 nm.

islands, as seen in Fig. 1. There might be a transition to a multilayer growth mode at higher coverages, as the third Fe layer is observed to start at a coverage of around 0.8 ML.

LEED studies on the above sub-monolayer coverages of Fe on Cu(111) grown by PLD have shown that there is no apparent difference between the LEED pattern of the clean Cu(111) substrate and the partially Fe covered sample. From AES measurements there is no evidence of contamination in the sub-monolayer films, i.e., carbon or oxygen related peaks are below the noise level, which compares well to the contamination level of thermal deposition. There is no indication of any implantation of Fe into the Cu(111) surface during PLD.

It is obvious from the STM images that there are distinct differences between the thermal evaporation and PLD of Fe onto the Cu(111) surface. In thermal evaporation, nucleation and island growth occur by collision of Fe atoms diffusing across the surface either randomly within a terrace or preferentially at energetically favorable positions on the substrate, i.e., on top of the step edge. This is governed by the need for a total surface energy minimization and results in the observed initial bilayer nucleation in the terrace, as well as the decoration of the step edges.

In PLD, no such growth behavior is observed. The Fe appears to be growing on the Cu(111) surface in a layer-by-layer fashion following random nucleation on the terrace. There is no decoration effect and no hole formation. It is known that the material deposition during PLD is occurring during a time of the order of microseconds after the short (ns) laser pulse,[11] such that by depositing 0.01 ML per laser pulse, as in our experimental setup, the deposition rate is in excess of 10^4 ML/s per laser pulse. Thus, the deposition rate is by a factor of 10^7 larger than in thermal deposition.[14] With this very high instantaneous deposition rate[11] the density of nucleation sites is significantly increased. There is a much higher probability for nucleation due to the fact that at an arrival rate of 0.01 ML within one pulse the mean distance between individual impinging atoms is around 10 atoms. Therefore, a large number of small two-dimensional islands is formed. Nevertheless, in our setup, at a laser pulse repetition rate of 1 Hz the integral deposition rate is 0.6 ML/min, which is within the range of the thermal deposition.

The Fe atoms deposited in the PLD process typically have a higher kinetic energy (about 1–10 eV on average) than in thermal evaporation. But there is no evidence that this high kinetic energy is leading to implantation or surface sputtering effects. The uncovered Cu surface does not look damaged and step edges appear quite smooth in the STM images. There are suggestions[11,14–17] that the high kinetic energy of the impinging atoms can be turned into a high
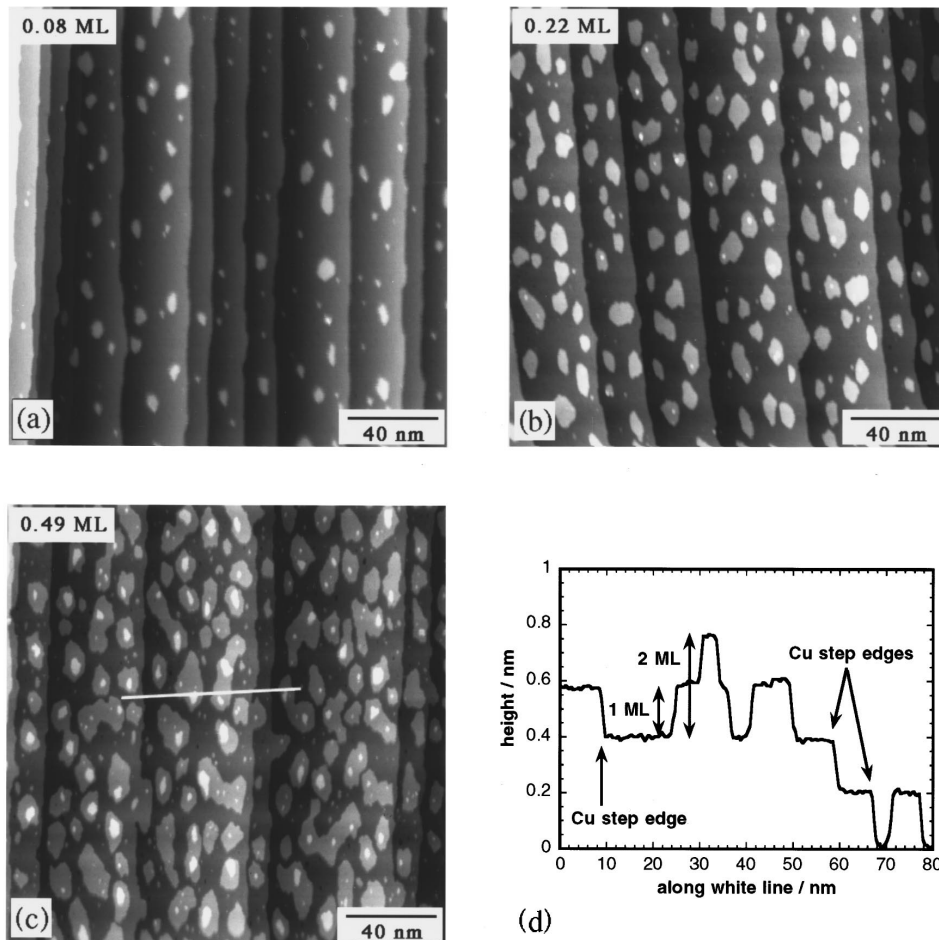
FIG. 2. Fe on Cu(111) deposited by PLD at 300 K showing that the initial growth proceeds via the layer-by-layer mode. (a) 0.08 ML, (b) 0.22 ML, (c) 0.49 ML Fe/Cu(111), and (d) height profile along the white line indicated in (c).

mobility of these atoms on the surface and, thus, further increase the nucleation probability and improve the growth. It is also possible that the impact of Fe atoms of higher kinetic energy results in additional nucleation sites and, therefore, also furthers layer-by-layer growth. A common problem in PLD is the presence of droplets in the deposited film, but during this STM study there was no indication for the presence of particulates on the samples. Both droplets and atoms with high kinetic energies, however, might be effectively suppressed by our experimental conditions, i.e., by choosing a low laser fluence just above the ablation threshold and working with a large target-to-substrate distance. Recently, we obtained nearly ideal layer-by-layer growth using PLD at substrate temperatures around 180 K. We found similarly successful results for Co/Cu(111) and also achieved true layer-by-layer growth in the initial stages (<2 ML) of Fe/Cu(100) and Co/Cu(100). These results will be presented in a forthcoming paper.

Therefore, we have proved by STM studies of the initial stages of growth that the characteristic features of PLD, i.e., the instantaneous deposition, are leading to a layer-by-layer growth mode. Thus, we present PLD under UHV conditions as a useful technique to improve the growth of Fe on Cu(111).
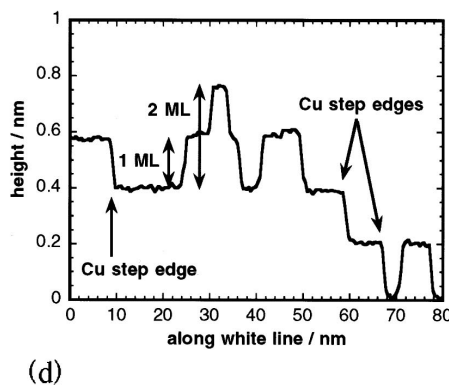
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