Abstract

The structure and quality of ultra-thin iron films on Cu(100) prepared by pulsed laser deposition (PLD) were studied by means of low energy electron diffraction (LEED). As in the case of thermal deposition (TD), the film undergoes a structural transition from a tetragonally expanded phase to the undistorted fcc structure at an iron coverage of 5 monolayers (ML). The changed structure of the Fe/Cu interface and the improved film quality are discussed in terms of the laser-fluence dependency of the deposited particle energies for PLD. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Laser methods; Low energy electron diffraction (LEED); Metal–metal magnetic thin film structures

1. Introduction

Epitaxial growth of thin metal films on single-crystal substrates offers the possibility to stabilize materials with novel physical properties. Magnetic thin films have attracted particular interest, since surface magnetism and structure are closely connected. One of the prototype systems for which this intimate relation was demonstrated is Fe/Cu(100), where only slight structural changes upon an increased film thickness result in a complete change of the magnetic behavior [1–4]. Up to 4 monolayers (ML), the iron films are tetragonally distorted (fct), and the whole film is ferromagnetic [2,3]. Above 4 ML, the films undergo a structural change, and ferromagnetism is restricted to the reconstructed surface layer, while the remaining undistorted fcc film shows paramagnetic [2] or antiferromagnetic [5] behavior. In accordance with theory [6], these differences come from a change in average vertical layer distance by about 5%. Up to now, ultra-thin magnetic films have usually been prepared by thermal deposition (TD) techniques. Though successfully used in material science (see, for example, Ref. [7]), it is only recently that pulsed laser deposition (PLD) was introduced for the preparation of Fe/Cu(100) upon an increased film thickness result in a complete change of the magnetic behavior [1–4]. Up to 4 monolayers (ML), the iron films are tetragonally distorted (fct), and the whole film is ferromagnetic [2,3]. Above 4 ML, the films undergo a structural change, and ferromagnetism is restricted to the reconstructed surface layer, while the remaining undistorted fcc film shows paramagnetic [2] or antiferromagnetic [5] behavior. In accordance with theory [6], these differences come from a change in average vertical layer distance by only 5%.

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While the kinetic energies of the atoms range between 5 and 10 eV [7], the kinetic energy of Fe$^+$ ions exhibits an almost linear dependence on laser intensity [12,13]. Already at a laser fluence of 4 J/cm$^2$, i.e. just above the ablation threshold (2 J/cm$^2$), mean kinetic energies and Fe$^+$ ionization yields above 50 eV and 35% are reported [12,13]. These ions can cause undesirable damage and intermixing. However, layer-by-layer growth may also be improved by particles with an elevated kinetic energy, since temporarily induced defects can act as nucleation centers, and higher kinetic energies may help to overcome surface potential barriers, e.g. at step edges of islands [14,15].

In a recent study, we have investigated the structure of PLD-grown iron films for a fixed laser fluence of 3–4 J/cm$^2$ by means of LEED [16]. In the present paper, we have extended this work to study the dependence of film quality and structure for laser fluences between 2 and 8 J/cm$^2$. The films were prepared in UHV (base pressure $8 \times 10^{-11}$ mbar) at room temperature by focusing XeCl excimer laser pulses on an Fe target. LEED intensity spectra $I(E)$ for the (10) beam are depicted in Fig. 1 for various coverages. The increased $R$-factors between 0/1, 4/5 and 10/12 ML roughly divide the set into three coverage regimes.

1. Between 1 and 4 ML, we observe a shift in individual peak structures to lower energies. This suggests an increase of the vertical layer distance, i.e. films with a fct structure. A tensor-LEED analysis of the 4 ML PLD film [16] reveals an average vertical expansion of the top two layers by 5%. Similar to the TD films [3], a complex $(5 \times 1)$ reconstruction with in-plane shifts and vertical displacements of the iron atoms compared to the ideal fcc position is deduced. However, the displacements diminish, and the iron interface layer exhibits the undistorted fcc configuration.

2. Between 5 and 10 ML, the shift direction of the individual peak groups reverses, indicating a reconstruction of the interlayer distances. A comparison with TD grown films demonstrates that the structures are seemingly identical for this coverage regime. Thus, a reconstructed surface layer resides on an undistorted fcc iron bulk. Above 10 ML, the iron film undergoes the well-established phase transition to bcc iron [4,9,16]. In a previous study, the magnetic properties of PLD-grown Fe films have been investigated [9]. A linear increase in the Kerr signal up to a coverage of 4 ML has been found, albeit with an in-plane easy axis of magnetization. This in-plane magnetization is not observed for TD films and is thus, from a magnetic point of view, the main difference. Around 4 ML, the Kerr signal drops to remain constant from 5 to 7 ML and then decreases to the value of TD films at 10 ML with the easy axis reoriented perpendicularly [8,9]. The combination of both results reveals that the structural change from fct to fcc phase entails a change in magnetism (see Refs. [9,16]).
3. Dependence on laser fluence

We turn now to the dependence of the film quality and structure on laser fluence. As described above, significant structural differences between TD and PLD grown films occur in the low coverage regime and can be mostly ascribed to the changed structure of the Fe/Cu interface. Note that the sensitivity of LEED for the interface drops with increasing iron coverage. Thus, possible changes remain unresolved at higher Fe coverages. Following the arguments in Section 1, it seems plausible that the observed changes result from a mixing of Cu and Fe atoms. As shown in Ref. [9], PLD of Fe on Cu(100) leads to layer-by-layer growth from the very beginning. Thus, Fe/Cu mixing will predominantly occur in the initial stage of growth. The incorporation of Cu atoms in the Fe interface layer can reduce the tensile stress induced by pseudomorphic growth and promote surface wetting. The penetration threshold for Cu atoms impinging on a Cu(100) sample was calculated in a molecular dynamic simulation to 150 eV [17], while a recent theoretical study yields a much lower value of 20 eV [14,15]. An alternative explanation is derived from the high kinetic energy of the ablated particles itself. It is well known that a repulsive barrier at island edges can cause 3D growth. A higher momentum parallel to the surface to the incident electron flux and the obtained averaged intensities for different spots aligned on a common scale will help to overcome this barrier and therefore support 2D growth. As the kinetic energy of the ablated ions depends on the laser fluence, some insight into the growth mechanisms can be extracted by changing the laser pulse energies. The LEED spectra of the (10) beam for 4 ML Fe shown in Fig. 2 for various laser fluences are practically identical. Consequently, the evaluation by pairs of the Pendry R-factors results in values below 0.085 for 4 ML and 0.072 for 2 ML (not shown). Thus, the structure of the film is not changed, while the intensity depends on the laser fluence. To illustrate this more clearly, integrated intensities for the (10), (11) and (20) beams are depicted in Fig. 2 (bottom). The corresponding value for the TD films is 0.4 at 4 ML [16]. The individual preparations differ only by ±0.1 ML. Thus, under the given experimental conditions, the quality of the films is significantly improved by PLD (a comparable behavior is observed for 2 ML Fe). Obviously, the best film order is obtained just above the ablation threshold while the intensity drops at higher fluences. As can be seen from Fig. 2, the instantaneous growth rate of the film changes only from 1.7 × 10^4 to 6 × 10^4 ML/min, assuming a constant deposition time of 1 μs. Thus, the observed continuous intensity decrease for increasing laser fluence is ascribed to the increasing kinetic energy of the impinging ions which, consequently, lead to a greater degree of surface damage. The observed improved growth and film quality at low coverages may be attributed to a change in growth kinetics. As discussed above, a possible scenario is a repulsive barrier at island edges that has to be overcome already for particle kinetic energies of 5–10 eV at the ablation thresh-
old. However, the incorporation of Cu atoms in the Fe film at the interface can lead to a reduction in film stress. This should result in an improved film quality if the early stage of growth is important for the growth mode and structure. In this case, mixing at the interface has to be sufficient already at low laser fluences since the structure is independent on laser fluence.

4. Conclusion

Pulsed laser deposition is a valuable tool for the preparation of thin epitaxial films on single crystal substrates. As demonstrated, for Fe on Cu(100), improved growth and changed film structures are obtained at low coverages. Nevertheless, PLD introduces many more experimental parameters, as compared with thermal deposition, that may offer further possibilities to improve thin-film growth but have to be controlled.

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