

## Annealing effect on morphology and magnetism of ultrathin films of Fe and Ni on Cu(100)

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### Abstract

Scanning tunneling microscopy (STM) and the magneto-optical Kerr effect (MOKE) have been used to investigate the effects of annealing on the morphology and magnetism of Fe and Ni films on Cu(100). Relatively mild annealing (490 K) created pinholes in the Fe/Cu(100) films, opening channels for the surface diffusion of copper to the top of the film surface along the walls of the pinholes. The magnetization of the Fe films is strongly affected by annealing. Annealing smooths the surface of Ni/Cu(100) films significantly, but no interdiffusion was observed upon annealing below 500 K. For Ni films thicker than 7 ML, the magnetization remains perpendicular after annealing, with only a slight reduction in its amplitude.

The magnetism of magnetic ultrathin films is influenced by a number of parameters such as the crystallographic structure, the strain, and the morphology, which usually undergo certain changes above a critical film thickness due to the lattice mismatch. Thermal annealing is one method of overcoming this difficulty, since it may convert a 3D morphology to a layer-by-layer one. In addition, once the Curie temperature ( $T_C$ ) is above room temperature, the annealing process when measuring  $T_C$  may cause irreversible changes in the magnetic behavior, which requires an analysis of the respective changes in the structural properties. In this paper, we report the annealing effect on ultrathin film systems of Fe and Ni on Cu(100), which show distinctly different behavior in morphology and magnetism upon annealing. STM, Auger electron spectroscopy (AES) and MOKE were used in this work; details of the experimental procedure are described elsewhere [1,2].

Fig. 1 demonstrates the annealing effect on a 3 monolayer (ML) Fe film. The morphology of the film before annealing is shown in Fig. 1(a). Three gray levels are visible on the surface, corresponding to the second (dark), the third, and the fourth (bright) layers, respectively. After annealing at 490 K for 10 min microscopic pinholes (the darkest patches in Fig. 1b) formed in the film. On the surface area between these pinholes, small dark patches are uniformly distributed which have an apparent fractional step height (0.7 Å at a sample positive bias of 900 mV).

The depth of the 'pinholes' is about 21 Å, independent of the bias. Since the vertical dimension of the 3 ML Fe film is only about 5.5 Å, the pinholes must extend well into the Cu substrate. The material removed from the pinholes has to diffuse to the top of the surface, because the annealing temperature (490 K) is far below the evaporating temperatures of both Fe and Cu. The AES Cu(60)/Fe(47) ratio of the 3 ML film after annealing is almost three times larger than that of the film before annealing, indicating that the surface region of the film became copper-rich after annealing. The fractional step height features are tentatively attributed to the formation of a Fe–Cu surface alloy. A full discussion of the Fe–Cu surface alloy has been presented in Ref. [1].

Fig. 2 shows the annealing effect on the perpendicular magnetization of the 3 ML Fe film. Before annealing (Fig. 2a) the Kerr intensity at saturation ( $M_s$ , scaled in arbitrary units) at room temperature is about 480, and the coercive field ( $H_c$ ) is about 7 Oe. After annealing (Fig. 2b), the perpendicular magnetization existed only below 205 K. At 190 K,  $M_s$  is about 120 and  $H_c$  is about 130 Oe. From our temperature-dependent MOKE measurements, the Kerr intensities at 0 K,  $M(0)$ , of the film before and after annealing were extrapolated to be about 750 and 220, respectively.

In contrast with the Fe/Cu(100) films, Ni/Cu(100) films did not show any detectable interdiffusion by AES as long as they were annealed at temperatures below 500 K. The surface of the Ni films after annealing was found to be much smoother than before annealing. This is demonstrated in Fig. 3, which shows the annealing effect (450 K,

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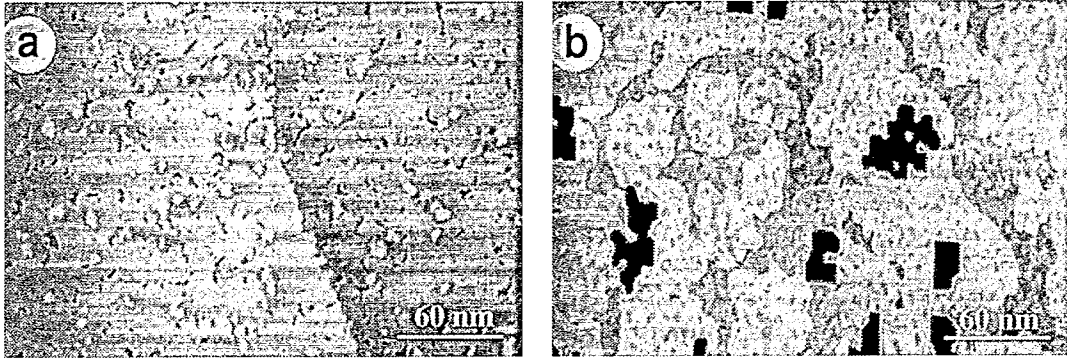


Fig. 1. STM images of 3 ML Fe/Cu(100) film. (a) Before annealing, the layer fillings of the third and fourth layers (brightest islands) are 95% and 5%, respectively. (b) After annealing at 490 K for 10 min, 'pinholes' (darkest patches) formed in the film and small depressions are clearly visible between the pinholes.

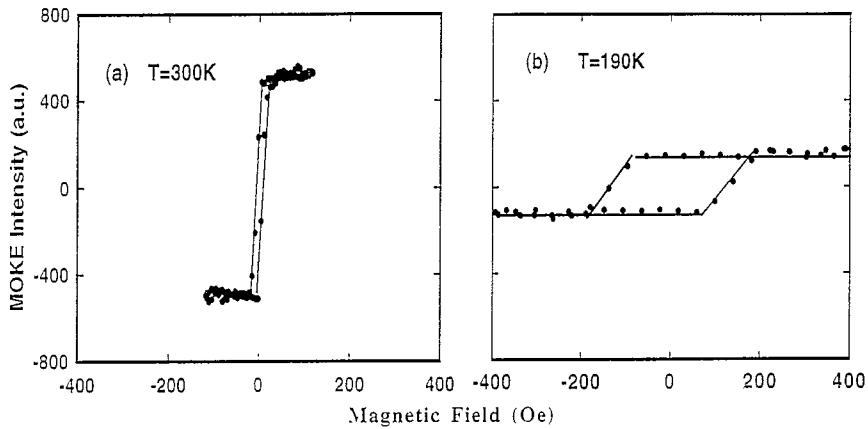


Fig. 2. Polar Kerr loops of the 3 ML Fe/Cu(100) film. (a) 3 ML before annealing; (b) 3 ML after annealing. Note that the two curves are plotted on the same scale.

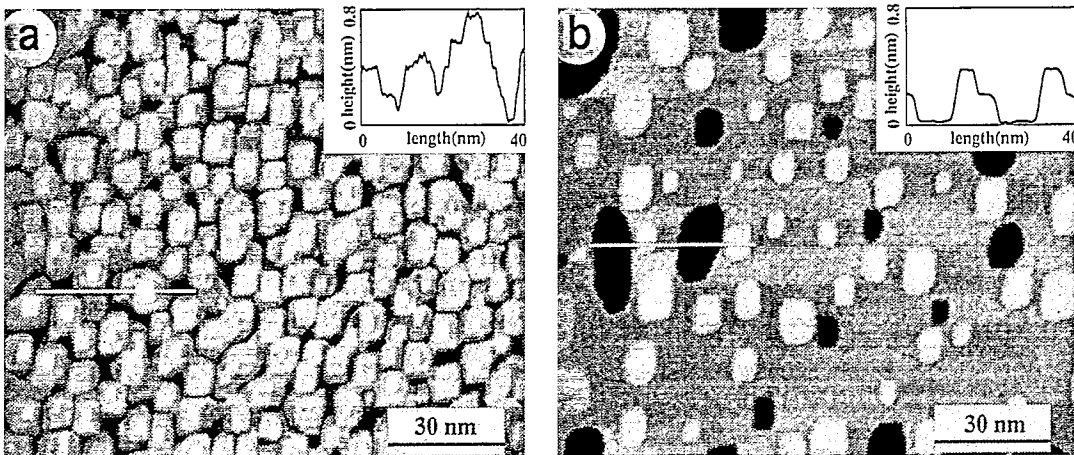


Fig. 3. STM images of 10.2 ML Ni/Cu(100) film. (a) Before annealing; (b) after annealing at 450 K for 10 min. The line profiles in the insets demonstrate that the peak-to-peak roughness of the film after annealing is about twice as small as that before annealing.

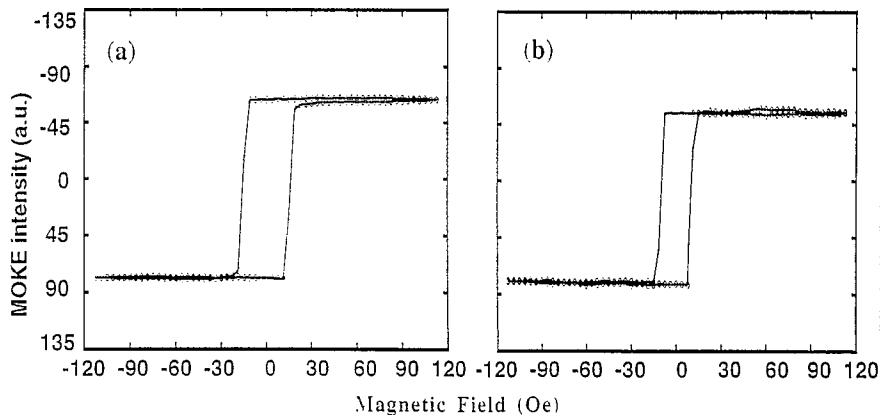


Fig. 4. Polar Kerr loops of the Ni/Cu(100) films. (a) 10.2 ML before annealing; (b) 10.2 ML after annealing. Note that the two curves are plotted on the same scale.

10 min) on the morphology of a 10.2 ML Ni film. The line profiles are plotted on the same scale in the insets of each image, showing that the peak-to-peak roughness after annealing is almost twice as small as that before annealing. Before annealing, well-separated 3D rectangular islands are observed, with an average size of  $10 \times 10 \text{ nm}^2$  and an average height of 0.8 nm. After annealing, the islands coalesced, resulting in a morphology similar to that of layer-by-layer growth. No pinholes were observed in the annealed film, which is consistent with the fact that the AES ratio of Ni(848)/Cu(920) hardly changed during annealing.

The difference in the morphological behavior of Fe/Cu(100) and Ni/Cu(100) films upon annealing could be due to the different surface free energies of Fe and Ni. According to Bauer [3], the expression  $\Delta_{B/A} = \sigma_B - (\sigma_A - \sigma_{B/A})$  indicates the tendency to lower the total energy of a thin film system B on A by 3D islanding. Interdiffusion is another possible mechanism to lower the energy in such a system. Here  $\sigma_B$  and  $\sigma_A$  denote the surface free energies of the film and the substrate, respectively, while  $\sigma_{B/A}$  denotes the interfacial energy. Since  $\Delta_{\text{Fe/Cu}}$  ( $1.64 \text{ J m}^{-2}$ ) is larger than  $\Delta_{\text{Ni/Cu}}$  ( $0.59 \text{ J m}^{-2}$ ) [4–6], the Fe/Cu system is expected to show a stronger tendency towards interdiffusion than the Ni/Cu system.

Fig. 4 displays the polar Kerr loops of a 10.2 ML Ni film before and after annealing at 450 K for 10 min. The Kerr intensity of the 10.2 ML film is measured to be 140 before annealing (Fig. 4a), and 135 after annealing (Fig. 4b) at room temperature. The coercive field of the film measured at room temperature decreases upon annealing, from 15 Oe before annealing down to 9 Oe after annealing.

The strong reduction (by more than a factor of 3) of  $M(0)$  of the 3 ML Fe film upon annealing can not be simply attributed to the formation of the Fe–Cu surface alloy, since the reduction of the Fe magnetic moment in Fe–Cu alloy is expected to be small [7]. A possible mechanism is that upon annealing a fct to fcc structural transition takes place, which reduces the atomic volume of iron atoms in all the iron layers except the top one to a value corresponding to that of the non-ferromagnetic state [8].

The perpendicular magnetization of the 10.2 ML Ni film hardly changed after annealing, despite the coalescence of the 3D islands upon annealing. This strongly suggests that the perpendicular magnetization of Ni/Cu(100) films does not originate from the morphology. Our result is consistent with an FMR study [9], which suggests that the perpendicular magnetization is driven by the magnetoelastic anisotropy.

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