Changes of Morphology, Structure, and Magnetism of Fe on Stepped Cu(111)

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Abstract—We have studied the morphology, structure and magnetism of Fe films thermally deposited on stepped Cu(111) by scanning tunnelling microscopy (STM), low energy electron diffraction (LEED) and magneto-optical Kerr effect (MOKE). At room temperature, in the submonolayer range Fe films grow as quasi one-dimensional stripes along the upper step edges due to a strong decoration effect. Between 1.4 and 1.8 monolayer coverage the stripes percolate and become two-dimensional films. Between 2.3 and 2.7 monolayer coverage the films undergo a structural transition from fcc(111) to bcc(110) with Kurdjumov-Sachs orientation. In the fcc range the films have a low net magnetic moment, and a perpendicular easy magnetization axis. The magnetization switches to an in-plane high-moment phase correlated with the fcc to bcc structural transformation.

Index Terms—Stepped metal surfaces, ultra thin film growth and magnetism

Magnetism in reduced dimensions is a research field of topical interest in recent years. One way to go beyond two-dimensional (2D) magnetism is to benefit from a special decoration effect at atomic steps to produce quasi one-dimensional Fe stripes on a stepped Cu(111) substrate [1,2]. It has been shown that the magnetism of parallel Fe stripes at submonolayer coverages has a superparamagnetic nature which differs from that of 2D ferromagnetic films by its time-dependent remanent magnetization [3]. By varying the thickness from submonolayer to a coverage of some monolayers the system Fe on stepped Cu(111) allows to follow the transition from 1D to 2D magnetism.

Another very interesting aspect of this system is the relationship of morphology, crystallographic structure and magnetic properties. In the low thickness limit the Fe/Cu(111) films have a metastable fcc structure [4,5], which does not exist in bulk below 1186 K. It has been found theoretically that fcc Fe can exist in nonmagnetic, antiferromagnetic, and low-moment or high-moment ferromagnetic phases depending on the lattice parameters [6]. Experimental checks of these theoretical predictions have been performed mainly on the Fe/Cu(100) system, while the structure and magnetism of the Fe/Cu(111) system is much less understood due to the very limited published work. At room temperature Fe grows pseudomorphically in a metastable fcc structure up to 4 monolayers [4] changing then to the bcc structure with (110) orientation [4,7,8]. STM studies revealed an initial bilayer growth changing to multilayer growth at room temperature [1,9]. Magnetic measurements from a copper capped Fe/Cu(111) film yielded a rather small magnetic moment (~0.6 μ_B) of Fe and the easy magnetization direction depending strongly on the growth temperature [10].

In this contribution we report on morphological, structural and magnetic studies of Fe films on stepped Cu(111) in the submonolayer to 5 monolayer coverage range by using scanning tunneling microscopy (STM), low energy electron diffraction (LEED) and magneto-optical Kerr effect (MOKE) measurements.

The experiments were performed in a multi-chamber system including an STM chamber, a MOKE chamber, and an analysis chamber equipped with Auger electron spectroscopy (AES), LEED and thin film growth facilities. The base pressure in each chamber is better than $5x10^{-11}$ bar. The vicinal Cu(111) substrate has a miscut of 1.2 ° normal to the [1-10] direction which gives a mean terrace width of about 10 nm of atomic steps in the [1-10] direction with a (001) microfacet. The Cu substrate was cleaned by cycles of 1 keV Ar^+ sputtering and annealing at 700 K . Its crystallographic quality and cleanness were checked by sharp LEED spots and contamination free Auger spectra. Fe films were grown by electron beam evaporation from a high purity wire at 273 K substrate temperature to suppress strain induced Cu surface diffusion during the deposition [2]. Polar and in-plane Kerr measurements were performed in the temperature range between 50 K and 270 K. STM images were recorded at room temperature after the MOKE measurements.

Fig. 1 shows 3 examples of STM topographical images from a series of Fe films on stepped Cu(111) at different thicknesses. At 0.8 monolayer (ML) thickness in Fig. 1a Fe forms quasi 1 D parallel stripes on the upper edges of the steps [2]. The height of the stripes is either one monolayer (grey) or two monolayers (brighter). The two darker contrasts between the Fe stripes correspond to the Cu substrate and 2D holes in the Cu terrace [2]. The width of the stripes of about 5 to 6 nm is nearly half the terrace width of 10 nm. Along the <110> direction of the steps the stripes have coalesced thus forming continuous wires with only rare disconnections. This situation describes the stage of 1 D percolation. With increasing thickness beyond 0.8 ML the stripes become wider and finally percolate between 1.4 and 2 ML (Fig. 1b) with direct connections between most of the stripes across the terraces. This represents the stage of 2D percolation. The film morphology changes from stripes to triangular-shaped islands with increasing thickness to 2.3 ML. The triangular island

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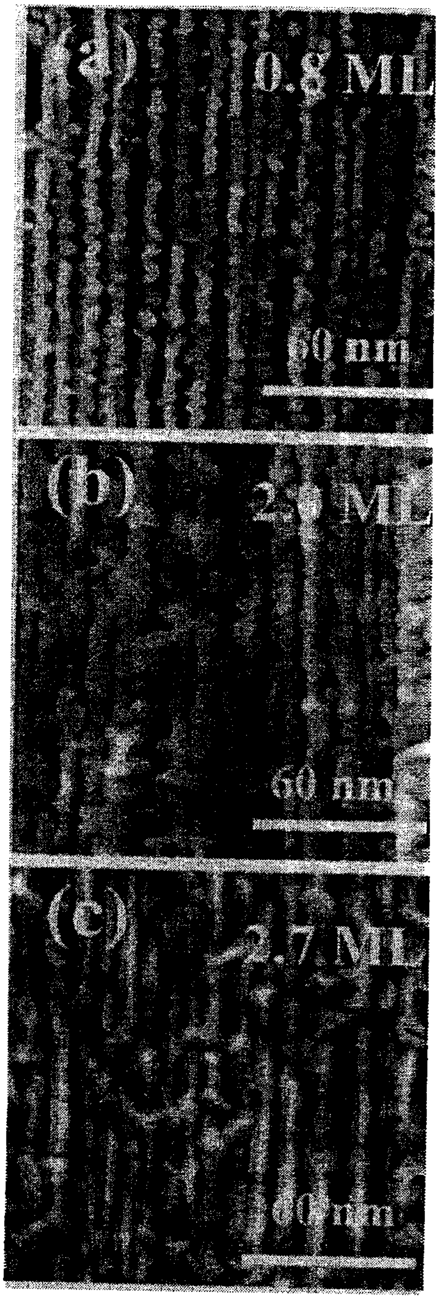

Fig. 1. STM images of Fe films on stepped Cu(111) in dependence on thickness. At 0.8 ML (a) quasi 1 D stripes are formed. The height of Fe stripes is either 1 ML (grey) or 2 ML (brighter). The two darker contrasts between the stripes correspond to the Cu substrate and 2 D holes in Cu. At 2 ML (b) a 2 D percolation across the terraces occurs. At 2.7 ML (c) a ridge-like morphology appears mainly along the <110> step direction and very rarely along the other two <110> directions.

shape in this stage of multilayer growth reflects the 3 fold symmetry of the fcc(111) structure. At a thickness of 2.7 ML in Fig. 1c the film morphology changes once again to elongated ridges mainly oriented along the <110> step direction but developing also along the two other <110> directions. The last stage of 3 fold ridge-like morphology is caused by a structural transition from fcc to bcc.

It has already been discussed in previous LEED work that this phase transition occurs between 4 and 5 ML thickness for Fe films grown on flat Cu(111) surfaces [5]. Additional satellite spots indicate that fcc(111) planes change to bcc (110) planes normal to the surface in the special Kurdjumov-Sachs orientation [5]. In our system Fe on stepped Cu(111) LEED pattern analysis shows that the fcc - bcc transformation occurs already at a lower critical thickness between 2.3 and 2.7 ML [11]. This conclusion is supported by an I/V LEED study of this system. In a kinematical analysis of the thickness dependent intensity curves a decrease of the interlayer distance of the fcc(111) Fe to the bcc(110) Fe distance is found near 2.5 ML thickness [11]. The smaller critical thickness in our system might be due to the high density of <110> oriented steps. At steps the Fe stripes are locally thicker than islands on the terraces and they are elongated preferentially along the step direction. Both these facts may result in an easier path for the fcc - bcc transformation as compared to the normal triangular-shaped islands on a flat Cu(111) surface.

As already shown in Fig. 1a the Fe films on stepped Cu(111) form quasi 1 D stripes at submonolayer coverage. In our previous polar MOKE measurements [3] we found that the easy magnetization axis of the stripes was perpendicular to the surface. No in-plane magnetic signal could be detected in longitudinal geometry.

Here we discuss the MOKE measurements over the thickness range from 1 ML to 5 ML. Fig 2 shows magnetization curves in polar geometry with increasing thickness. From longitudinal magnetization curves (not shown here) it follows that below 2.3 ML thickness the films exhibit magnetic signals only in the polar geometry. This indicates that up to 2.3 ML the easy magnetization axis is perpendicular. At 160 K, the polar hysteresis loops have very small remanence and coercivity. With increasing thickness the saturation field increases continuously from 400 Oe for 2 ML to 850 Oe for 2.5 ML. For a 3 ML film the saturation field jumps to about 4500 Oe, and in-plane hysteresis loops with near rectangular shape start to appear. This indicates that the easy axis switches from out-of-plane to in-plane in the thickness range between 2.3 and 3 ML. The origin of this spin reorientation can be explained by two different arguments. For very thin films it is generally expected that the interplay between the constant positive surface anisotropy and the thickness-dependent negative shape anisotropy results in a critical thickness at which the easy magnetization axis switches from perpendicular to in-plane. On the other hand the spin reorientation occurs in the same thickness range as the structural transformation from fcc(111) to bcc(110). The two-fold in-plane symmetry of the bcc(110) plane contributes an additional in-plane anisotropy term. Thus the easy magnetization axis switches to in-plane as soon as the bcc(110) structure is attained.

A very remarkable change of the magnetism can be revealed if we determine the polar saturation magnetization as a function of film thickness, as shown in Fig. 3. We have to

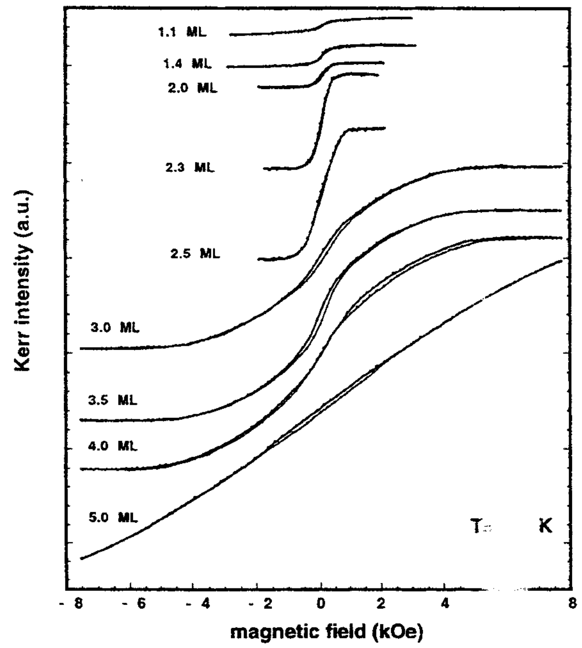

Fig. 2. Polar magnetization curves of Fe films with increasing thickness. In the thickness range between 2.5 and 3 ML the saturation field in the perpendicular direction substantially enhances by a factor of 5.

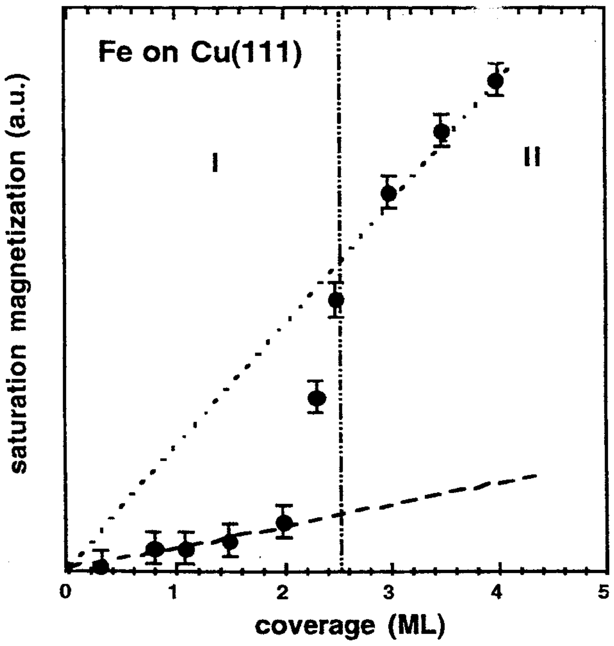

Fig. 3. Polar saturation magnetization as a function of film thickness measured at about 160 K. The magnetization abruptly increases near the vertical line indicating the phase transition from fcc to bcc.

emphasize that all films were saturated along the surface normal no matter whether the easy axis is perpendicular or in-plane. In the low thickness range < 2.3 ML the saturation signals have small values whose linear extrapolation (dashed line) goes approximately through zero. If the magnetic moment keeps constant, then with increasing thickness beyond 2.3 ML the saturation Kerr signals should fall on the dashed line. However, in the thickness range between 2.3 and 3 ML corresponding to the fcc to bcc transformation the saturation Kerr signals increase sharply. Beyond the vertical line indicating the completion of the phase transformation the saturation signal increases again linearly, however, with a 5 times larger slope. Since the MOKE does not measure the magnetization directly any conclusion concerning the magnetic moments needs a careful discussion of all relevant aspects. The result of such an analysis in [11] can be summarized as follows. The fcc Fe films on stepped Cu(111) below 2.7 ML are in a low-spin ferromagnetic or a ferrimagnetic phase. The estimated magnetic moment is about 0.4 to 0.5 µ which is close to an early experimental value of 0.5 to $0.6\mu_B$ reported for a copper capped Fe/Cu(111) film [10]. Theoretical calculations predicted two low-spin phases [12] the lower one with a moment of $0.4\mu_B$ in agreement with our estimation. The bcc Fe films above 3 ML thickness exist in the high-spin ferromagnetic phase with the bulk moment of $2.2\mu_B$.

In conclusion, we have studied the morphology, structure and magnetism of Fe films grown on stepped Cu(111). In the submonolayer range the Fe films grow as quasi 1D stripes along step edges. Between 1.4 and 1.8 ML the stripes percolate to 2 D films. Between 2.3 and 2.7 ML the films undergo a fcc to bcc transition as well as a spin reorientation from perpendicular to in-plane. Experimental evidence suggests that the fcc films have a low-spin phase, whereas the bcc films are in a high-spin phase.

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