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ABSTRACT

Epitaxial Fe wedges with a thickness gradation from 0 - 20 Å were grown on diamond(100) at room temperature, subsequently annealed, and investigated with reflection high-energy electron diffraction and the surface magneto-optical Kerr effect. The results indicate that for < 5 monolayer thicknesses the Fe grows on C(100) as smooth, epitaxial *fcc* films, which are not ferromagnetic, but that thicker films undergo a transition to become rough and the ordinary bcc ferromagnetic phase.

I. INTRODUCTION

Face-centered-cubic (fcc) Fe (also referred to as γ -Fe) thin films have provided a rich variety of magnetic and structural phase transitions. Unlike the more stable ferromagnets Co and Ni, fcc Fe is predicted to have both ferromagnetic (FM) and antiferromagnetic (AFM) phases depending on atomic volume.¹ In addition, fcc Fe, as a high-temperature phase in nature, can only exist as a metastable phase at room temperature (RT) via epitaxial stabilization on suitable substrates² or in a Cu host as coherent precipitates. As a result, epitaxial Fe films can form several structurally similar, yet subtly different face-centered phases³ with strikingly different magnetic properties. For example, one of these phases has a face-centered-tetragonal (fct) structure and FM ordering. Another less distorted fcc phase has an FM “live” surface and AFM interior layers.^{2,4} This latter phase is one of few beside Gd(0001),^{5,6} for which the surface magnetic ordering temperature is higher than that of the interior. It also shares a relatively unique position with Cr(100)⁷ in having a FM surface coupled to an AFM bulk. The complex multiplicity of face-centered Fe phases were recently described theoretically as arising from frustrated exchange interactions.⁸ Epitaxy of Fe on different substrates promotes the stabilization of different metastable face-centered phases. So far most of the substrates used to grow fcc Fe, such as Cu(100) (lattice constant $a = 3.61 \text{ \AA}$),^{2,4,9,10} Co(100),^{11,12} Cu-Au alloy¹⁰ and Ni-Fe alloy,¹³ have slightly larger lattice constants than the extrapolated RT fcc Fe value of $3.59 \text{ \AA}$ and therefore represent an in-plane *expansion* for fcc Fe. Diamond C(100), on the contrary, has a lattice constant of only $3.57 \text{ \AA}$, and therefore represents an in-plane *contraction*. Recently, fcc Fe was grown epitaxially onto C(100),^{14,15} though the magnetic characterization was inconclusive. In the present work, we investigate wedge-shaped Fe films of $0 - 20 \text{ \AA}$ on C(100) by means of reflection high-energy electron diffraction (RHEED) and the surface magneto-optic Kerr effect (SMOKE) to systematically identify the structural and magnetic phases. We demonstrate that the films < 5 monolayers (ML)

thick are flat, well-ordered fcc films, and are consistent with a prior Mössbauer identification¹⁶ as being a low-moment AFM. Films > 5.5 ML thick exhibit a three-dimensional (3D) bcc structure and are the normal ferromagnetic phase with in-plane easy axis of magnetization. The results provide insights into the competing driving forces that govern the formation of structural and magnetic phases in metastable, ultrathin magnetic films.

II. EXPERIMENTAL

The experiments were carried out in a ultrahigh vacuum (UHV) chamber equipped with RHEED, low-energy electron diffraction (LEED), Auger electron spectroscopy and SMOKE. The RHEED images were obtained using a charge-coupled diode (CCD) camera. The streak and/or spot spacings and widths of the RHEED patterns were analyzed digitally to quantify the lattice spacings and terrace sizes. Synthetic diamond C(100) substrates used in this work were first etched with acid solution as previously reported,^{14,16} and then transferred into the UHV chamber through a load-lock system, and heated to 650°C for ~ 20 min. This procedure results in well-ordered, clean C(100) surfaces that have sharp RHEED and LEED patterns and are free of oxygen or any other impurity as detected with Auger using a hemispherical analyzer. Fe films were deposited at RT from a resistively heated crucible. The background pressure during deposition was typically $\sim 1 \times 10^{-9}$ Torr, while the base pressure of the chamber is $< 1 \times 10^{-10}$ Torr. The films were subsequently annealed at 400°C after each ~ 4 Å dosage to ensure fcc Fe growth, while minimizing interdiffusion. The nominal Fe thickness was determined with a quartz crystal monitor that was calibrated against the RHEED oscillations of Fe/Cu(110). Fe wedges of 0 - 20 Å were grown while translating the substrate behind a knife-edge mask using a stepping motor. The films are free of oxygen contamination as confirmed by Auger, though a C signal is always visible, suggesting exposure of the C(100) substrate and/or C-interdiffusion. After each experiment, the substrate was removed from UHV and acid etched as before for re-

use. *In-situ* magnetic characterization was performed with longitudinal and polar SMOKE measurements utilizing a focused He-Ne laser scanned along the wedge at temperatures between 120 - 300 K. The Kerr signal proportional to the saturation magnetization M_s (in arbitrary units) was obtained from Kerr-ellipticity hysteresis loops.

III. RESULTS AND DISCUSSIONS

Figure 1 shows typical RHEED patterns of a C(100) substrate and the Fe films of 4.0 and 7.8 ML (where $1\text{ML} \equiv 1.8 \text{ \AA}$). The former two [Fig. 1(a) and (b)] exhibit sharp 1×1 streaks with similar spacing, indicating flat, well-ordered fcc surfaces. The latter [Fig. 1(c)] shows a spotty pattern typical of a 3D rough surface, while some ring-shaped features indicate the coexistence of a polycrystalline phase. A detailed analysis of spacings¹⁶ indicates that the 7.8-ML Fe is bcc with its $[111]$ oriented along the fcc $[110]$, and its row spacing $2.01 \pm 0.02 \text{ \AA}$ perpendicular to $[110]$. This is in excellent agreement with the bulk bcc Fe value of $2.03 \text{ \AA}$. Our results, therefore, indicate a structural phase transition from the metastable fcc phase to the bcc ground state.

While such a structural phase transition occurs in all the samples, some transitions are more gradual than the one shown in Fig. 1. Figure 2 shows another set of RHEED patterns of Fe/C(100) along a wedge [(a) 1.6 ML; (b) 3.1 ML; (c) 4.5 ML; (d) 6.5 ML] on another C(100) substrate for comparison. While the films remain fcc in Figs. 2(a)-(c), the patterns become increasingly 3D-like and the widths of the streaks/spots increase before the film completely transforms into a 3D bcc phase [Fig. 2(d)].

This fcc - bcc structural phase transition is better illustrated in Fig. 3. Here the RHEED streaks / spots spacing and width are plotted vs. the nominal Fe thickness along the wedges, where the top panel (a) is from the wedge for Fig. 1 and the bottom panel from the wedge for Fig. 2. It is clear that in both cases, there is a structural phase transition at $\sim 5 \text{ ML}$ as evident from the drastic changes in the RHEED spacings and widths. These variations indicate the changes in row spacing and grain size in real spacing that accompany

the structural phase transition, which occurs at ~ 5 ML for all the clean samples. The *differences* among the different wedges [see, for example, Fig. 3(a) and (b)] is mainly in the sharpness of the transitions. In Fig. 3(b), the RHEED streak spacing starts to relax gradually even before 5 ML, and the streak widths broaden, indicating increasingly finer grains. These results point to the appearance of a small amount of bcc phase even before the main transition at ~ 5 ML. The apparent small spacing changes for Fe thickness < 5 ML should be a result of averaging over the fcc and the bcc grains. This type of quantitative differences is common among the wedges grown on different substrates or on the same substrate after repeated acid etching. We believe that these differences are the result of different defect densities on the surfaces. Nevertheless, these minor details do not affect our main result that a structural phase transition occurs at ~ 5 ML.

Figure 4 shows M_s vs. Fe thickness at 130 K obtained from longitudinal SMOKE measurements, as compared with the changes of RHEED streak spacing from Fig. 3(b). It is evident for all the wedges (shown by different symbols in Fig. 4) that M_s increases with Fe thickness linearly with the same onset of $\sim 5.5 - 6$ ML. The Curie temperature (not shown) also has a similar onset at ~ 5.5 ML and quickly increases to 300 K within 1 ML.¹⁶ Such an onset coincides with the fcc - bcc structural phase transition as evident from the RHEED streak spacing. There is no ferromagnetic signal detected observed between 120 - 300 K in either polar or longitudinal measurements. Therefore, we only observe ferromagnetic ordering in the 3D bcc films > 5.5 ML, but not in the 2D fcc films < 5 ML. An *in-situ* Mössbauer measurement¹⁶ in a companion study indicates that this fcc phase is paramagnetic at temperatures ≥ 70 K and is a low-moment AFM below 40 K.

While all the clean films exhibit the fcc - bcc phase transition at ~ 5 ML, we note that thicker fcc films are sometimes retained on dirty substrates subjected to a higher background pressure ($> 5 \times 10^{-9}$ Torr). In these cases, Auger detects oxygen contamination of both the substrate and the films. The corresponding RHEED patterns of the Fe films, although fcc, show additional half-order streaks, as seen in Fig. 5. These

films also exhibit $c(2 \times 2)$ reconstruction in LEED (not shown), which is common for the contaminated fcc (100) transition metal surfaces. The fcc phase probably persists due to surfactant-mediated growth as occurs in Fe/Cu(100).^{17,18}

IV. SUMMARY

We have systematically studied the epitaxial growth of fcc Fe on C(100) utilizing wedged structures. RT-grown, annealed Fe films are 2D fcc with the same lattice spacing as C(100) for Fe thicknesses < 5 ML, and are 3D bcc for thicknesses > 5 ML. The fcc Fe/C(100) is paramagnetic at RT (and AFM at low temperature based on a separate Mössbauer study¹⁶), while the bcc Fe is the ordinary ferromagnetic phase.

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Figure Captions

- Figure 1. Typical RHEED images taken at an electron energy of 9 kV with a CCD camera. (a) clean C(100) substrate; (b) 4.0 ML Fe/C(100); (c) 7.8 ML Fe/C(100).
- Figure 2. RHEED images of another Fe wedge at (a) 1.6 ML; (b) 3.1 ML; (c) 4.5 ML; (d) 6.5 ML taken at an electron energy of 9 kV with a CCD camera.
- Figure 3. The RHEED streak/spot spacings and widths along two typical Fe wedges [(a) and (b)] on diamond (100). A structural phase transition is apparent at ~ 5 ML, while quantitative differences of the transition exist among samples.
- Figure 4. The saturation magnetization M_s at $T = 130$ K as measured from longitudinal Kerr hysteresis loops along the Fe wedges on C(100). Different symbols represent measurements from different wedges. The RHEED streak spacing in Fig. 3(b) is re-plotted for comparison.
- Figure 5. Row scan across the RHEED pattern of a 7.5-ML Fe film on dirty C(100). Note the weak half-order peaks associated with surface reconstruction.

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