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Scanning Tunneling Microscopy Observation of Epitaxial bcc-Fe(001) Surface

Agus SUBAGYO[1,*], Hirofumi OKA[1], Guido EILERS[2], Kazuhisa SUEOKA[1,3] and Koichi MUKASA[1,2]
[1]*Graduate School of Engineering, Hokkaido University, Kita-13, Nishi-8, Kita-ku, Sapporo 060-8628, Japan*
[2]*CREST, Japan Science and Technology Corporation (JST), Kita-13, Nishi-8, Kita-ku, Sapporo 060-8628, Japan*
[3]*PRESTO, Japan Science and Technology Corporation (JST), Kita-13, Nishi-8, Kita-ku, Sapporo 060-8628, Japan*

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We report the first atomic-resolution scanning tunneling microscopy (STM) image of epitaxial bcc-Fe(001) films grown on MgO(001) substrates. A 50-Å-thick Fe film grown at a growth temperature of 550 K formed square pyramidal islands with atomically flat terraces. The terraces were found to range between 5 nm and 20 nm in width separated by monoatomic high steps. The film exhibited a (1 × 1) unreconstructed structure at a film thickness below 19 Å; however, a reconstructed surface was found on thicker films. The atomic-resolution STM image and low energy electron diffraction (LEED) observation indicated that the reconstructed structure is a c(2 × 2) structure.

KEYWORDS: scanning tunneling microscopy, bcc-Fe(001), MgO(001), epitaxial growth, surface reconstruction, c(2 × 2)

1. Introduction

In recent years, since a bcc-Fe(001) surface is expected to have highly spin-polarized surface states,[1)] there has been growing interest in the investigation of atomically flat Fe films on MgO(001) with respect to the preparation of a suitable surface for spin-polarized tunneling experiments[2,3)] and electron spin polarimetry.[4)] By using this material as a sample for spin-polarized scanning tunneling microscopy (SP-STM) experiments, we found that the tunneling current between an optically pumped GaAs tip and the magnetic sample was dependent on the magnetic condition of the sample.[5)] This material is also interesting for technological applications, for example, as a seed layer for the epitaxial growth of other layers and multilayers,[6)] and for the fabrication of submicron particles[7)] and wires.[8)]

By using STM, Thurmer *et al*. have reported that Fe/MgO is a thin-film system with effective Schwoebel barriers at the step edges[9)] so that a specific structured film can be obtained by controlling the growth temperature. They found that atomically flat films can be obtained at growth temperatures above 500 K. Jordan *et al*. also reported a similar result for thinner films.[2)] A flat surface with terraces separated by atomic steps on a magnetic sample can be expected to provide many advantages for the spin-polarized tunneling experiments, since the spin-polarization of surface electron states strongly depends on the local arrangement of atoms. An atomically flat film can be obtained easily; however, an atomically resolved STM image on the surface has not been reported so far. Understanding of the atomic structure of this surface is very useful for obtaining high-resolution spin-polarized information; therefore it is also essential to study the correlation between surface structures and magnetism.

In this work, we report the first atomically resolved STM image of Fe film surfaces grown on MgO. At a growth temperature of 550 K, we can produce an atomically flat film with wide terraces and obtain atomic images of the flat terraces. A previous low energy electron diffraction (LEED) study indicated that the high-quality Fe single crystal films had a (1 × 1) unreconstructed surface, and segregated or adsorbed oxygen or other atoms induced other reconstructed surfaces. By observing the film thickness dependency of the surface structure, we showed that the possibility remains that the c(2 × 2) reconstructed surface is composed of Fe atoms.

2. Experiment

The experiments were performed in an ultrahigh-vacuum system equipped with electron beam evaporators (Omicron EFM3), reflection high energy electron diffraction (RHEED), and Ar^+ ion bombardment for the preparation chamber ($p = 1 \times 10^{-8}$ Pa); and LEED, Auger electron spectroscopy (AES), X-ray photoelectron spectroscopy (XPS), atomic force microscopy (AFM) and STM system for the analysis chamber ($p = 3 \times 10^{-9}$ Pa).

As substrates, commercially available, mechanically polished MgO(001) substrates were used. The substrates were ultrasonically cleaned in acetone, 2-propanol and methanol, and then were exposed in boil acetone at 323 K. An Au strip was deposited on both edges of substrates to provide an electrical contact between Fe films and the sample plate. The substrates were cleaned by 3 keV Ar^+ ion bombardment for 5 minutes at a grazing angle of 30° followed by flashing at 1173 K. The cleanliness of the substrates was monitored by AES.

An Fe film was deposited at a growth temperature of 550 K at a deposition rate of 0.5 ML/min (1 ML = 1.433 Å) from a high-purity (99.99+%) Fe wire. In order to avoid any effect from the impurities, the Fe wire was preheated for more than 6 h. The flux of Fe atoms was directed along the Fe[110] direction at an incident angle of 20° from the surface normal. The pressure during the deposition never exceeded 3.0×10^{-8} Pa. An electrochemically etched W wire was used as a tip. The tip was cleaned by Ar^+ ion bombardment and *in situ* annealing. STM observation was performed at room temperature. Topographic STM images were obtained in a constant current mode.

3. Results

RHEED and LEED observations show that MgO substrates exhibit a (1 × 1) bulk terminated structure even before any cleaning treatment in vacuum. As confirmed by AES, the cleaning procedures described in §2 can remove the contaminant. The very sharp RHEED and LEED patterns indicated that a clean surface was obtained. According to the AFM images, the substrate exhibits a flat surface with average rms surface roughness of about 1.5 ± 0.05 Å.

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[*]E-mail address: agus@nano.eng.hokudai.ac.jp

As we have reported previously,[3,10)] and in agreement with the findings of other groups,[9,11)] Fe films ($a_{\text{Fe-bulk}} = 2.867$ Å) grow epitaxially in the bcc phase on the rock-salt structure of MgO substrates ($a_{\text{MgO}} = 4.213$ Å) with a lattice mismatch of about 3.8%. The epitaxial relations are Fe(001)//MgO(001) and Fe[110]//MgO[100].

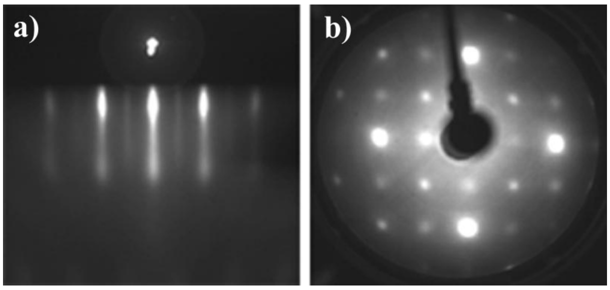

Fig. 1. RHEED and LEED patterns of a 50-Å-thick Fe film grown on MgO(001) at a growth temperature of 550 K. (a) RHEED patterns obtained in the Fe[110] direction. Beam energy: 20 kV. (b) LEED patterns obtained at a primary energy of 171 eV.

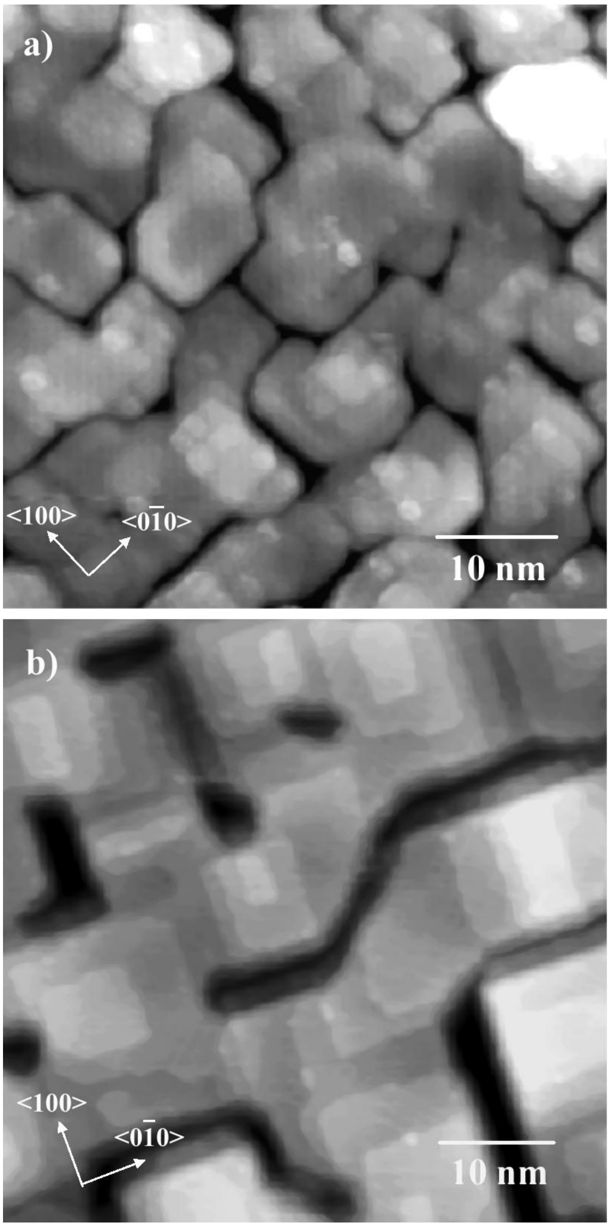

Fig. 2. STM image of Fe films grown on MgO(001) substrates at a growth temperature of 550 K. (a) 35-Å-thick film, (b) 50-Å-thick film. Scan size of both images: $50 \times 50\,\text{nm}^2$. Terrace width in image (b) ranged between 5 and 20 nm wide. The steps run along the Fe[100] direction. The steps are monoatomically high which is consistent with $a/2$ of bulk Fe.

RHEED observation during growth shows that the films exhibit a (1×1) unreconstructed structure at low coverage films. When the film thickness achieved about 19 Å, RHEED patterns became two-fold patterns, indicating the occurrence of surface reconstruction. This reconstructed structure never changed and becomes more clearly observed as the film thickness increases to 50 Å, as shown in Fig. 1(a). As revealed by LEED patterns shown in Fig. 1(b), this reconstructed structure can be referred to as a $c(2 \times 2)$ structure.

Figure 2 shows STM images of the surface structures of 35-Å (Fig. 2(a)) and 50-Å (Fig. 2(b)) thick Fe films. At a film thickness of 35 Å, Fe film forms flat square islands with square terraces (or 2D-nuclei) on the surface. The island edges run along the Fe[100] direction. The terraces were very narrow. Although the film exhibits an atomically flat surface, it was difficult to obtain an atomically resolved STM image on this surface.

By increasing the film thickness to 50 Å, the edges of the islands become clearly defined and straight, as shown in Fig. 2(b). The film forms square pyramidal islands with 4 to 6 layers of atomically flat terraces. The terrace width ranged between 5 and 20 nm. The steps run along the Fe[100] direction, parallel to the edges of the islands and are monoatomically high which is consistent with $a/2$ of bulk Fe.

The formation of an atomically flat surface at a growth temperature of 550 K is consistent with previous works, in which the atomically flat surface was obtained at growth temperatures typically above 500 K.[2,9)] In this temperature range, the so-called Schwoebel barrier,[12)] which prevents adsorbed atoms from transferring to the lower terraces through steps, is overcome, and then Fe atoms can diffuse not only in an upward direction but also in a downward direction on the step edges. Thus, at this temperature, 2D growth, which is favored

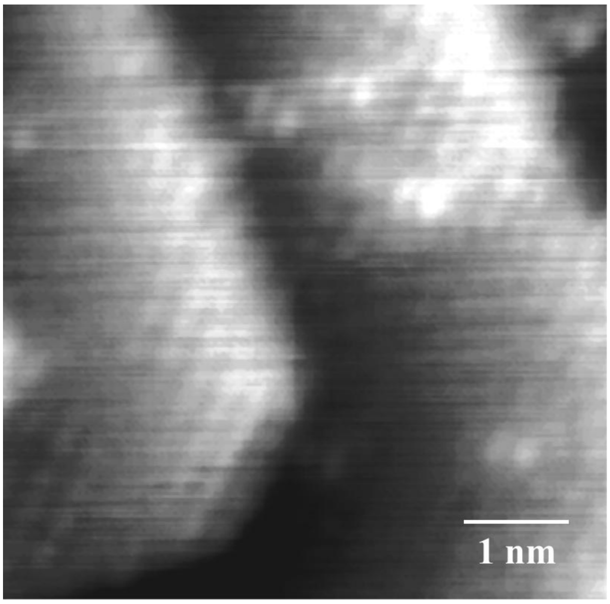

Fig. 3. Atomically resolved STM image of a 50-Å-thick Fe film. Scan size: $6 \times 6\,\text{nm}^2$. The atomic structure is a $c(2 \times 2)$ reconstructed structure. Atomic corrugation is 0.15 Å.

by thermodynamics, becomes dominant, and leads to formation of an atomically flat film.

An atomically resolved STM image on the flat terraces of a 50-Å-thick Fe film is shown in Fig. 3(a). The scan size is 60×60 Å. The distance of the atomic periodicity of Fe atoms in the [100] direction was found to be typically twice the distance of Fe atoms of the bulk. Thus, the atomic structure shown in the image is revealed to be a c(2×2) reconstructed structure. This structure is consistent with the LEED patterns shown in Fig. 1(b). The atomic corrugation is about 0.15 Å.

4. Discussion

There are several possible reasons for surface reconstruction in material surfaces, for example, surface segregation between substrates and overlayers, and surface adsorption. Oxygen segregation into the Fe film surface induced by the annealing at about 873 K of postdeposited Fe films on MgO(001) has been reported to give rise to surface reconstruction.13) Interdiffusion and surface segregation of magnesium in Fe_3O_4 films grown on MgO(001) induced by annealing at 700 K–800 K has also been reported. However, magnesium did not segregate to the film during annealing at 630 K, or even during growth at a temperature of 570 K.14) Thus, we conclude that the reconstruction in our film is not caused by magnesium or oxygen segregation, since the growth temperature of our films was 550 K, which is sufficiently lower than 700 K.

We want to emphasize that the reconstruction of our film is not caused by impurities coming from the evaporated Fe itself during growth, since our film did not contain any contaminants such as sulfur or other metal impurities, as confirmed by XPS measurements. In our study, we did not observe any reconstruction or atomically flat surfaces on films grown at growth temperatures below 493 K. Furthermore, as confirmed by RHEED observations and STM images of the growth morphology of films grown at growth temperatures above 493 K, we found that the reconstruction was formed at the same time as the formation of an atomically flat surface. The critical thickness at which the reconstructed surface was formed strongly depended on the growth temperature. These indicate the strong relationship of reconstruction with the growth process of Fe film itself.15) Therefore, although there still remains a possible effect from the migration of remaining contaminants from substrates to the film surface during growth, we believe that the reconstruction formation is related to the growth process of Fe film itself.

5. Summary

The surface structure of bcc-Fe(001) films epitaxially grown on MgO(001) substrates was studied by STM. Films grown at a growth temperature of 550 K formed pyramidal square islands with atomically flat terraces separated by monoatomic steps. The atomically resolved STM images revealed that the film had a c(2×2) reconstructed surface and its atomic corrugation was 0.15 Å.

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