〈연구논문〉

Determination of Mixing by a Scaling Behavior in Fe on Cu(001) System

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Scaling 형태분석을 통한 Fe/Cu(001)계의 혼합 여부 결정

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Abstract – The growth structure of Fe on Cu(001) was studied by scanning tunneling microscope. An analysis of size distribution of Fe islands on Cu(001) surface was made to determine whether Fe atoms mix with substrate Cu. The size distribution deviates from the standard scaling behavior, indicating that atomic density of Fe decreases with coverage up to 1 ML. The growth can be characterized by layer-by-layer scheme from 1 ML to 5 ML. This result agrees well with previously studied, Auger spectroscopy and RHEED result.

1. Introduction

The growth of Fe on Cu(001) has drawn much attention since this system can be used as a film magnetic recording or a reading device. Even though it is generally believed that Fe atoms grow epitaxially as an fcc structure with layer by layer scheme up to 11 ML, the growth structure at early stage is still controversial [1-8]. It was theoretically proven that the bcc Fe is more stable at room temperature (RT), but the fcc Fe which is stable only above 913℃ in bulk can be epitaxially grown up to 11 ML in the presence of Cu(100) substrate [1, 2]. That is another reason why this system has been widely studied for the last decade. This epitaxial Fe film is rather defect free but the magnetic property varies with substrate temperature and film thickness[5,6]. At the coverage of 2~4 ML, ferromagnetic Fe is present due to the lattice distortion of the fcc film to a face centered tetragonal structure or a reconstructed fcc structure [5, 6]. Above ~5 ML, Fe film reveals antiferromagnetism [5, 6], consistent with the magnetism of bulk fcc Fe. The geometrical structure of the film or the magnetic property was measured independently, but there is not much interpretation which can link these two observations. The surface phase diagram of this film layers has never been proposed, but the mixing due to the instability at elevated temperature or at low coverage were reported by ion scattering [7], Auger [8] and STM [9] experiments, suggesting Cu segregation. Some pointed out heavy mixing at < 1 ML even at room temperature and some at elevated temperature, above 370℃, through a pin holes developed during the mixing process.

The oscillation curves observed by RHEED [3] or MEED [4] due to the growth morphology indicate a layer-by-layer growth of fcc Fe film at room temperature with the coverage of >2 ML. But, they differ at the early stage growth. We have reported that, above 3 ML, Fe grows layer by layer up to 10 ML. When the coverage is fractional, i.e. in between two consecutive integers, the island size distribution of the Fe islands can be represented by a unique curve suggested by a scaling theory [11]. In this report, we report that the island size distribution cannot be scaled to single curve at low coverage of <1 ML.

With this we can conclude Fe mixes with the substrate Cu even without doing macroscopic Auger experiment, in which it is not sure whether the Auger electron beam is probing the same area where the STM image is taken.

2. Experimental

A Cu(001) single crystal was cut with a miscut $<0.5°$. It was mechanically and electro-chemically polished before loaded into a UHV chamber. After outgassed at 650℃, (measured by an optical pyrometer) the Cu sample was cleaned by several cycles of Ar^+ sputtering followed by 600℃ annealing, till a sharp LEED pattern was obtained without trace of C or S contamination. The prepared sample showed atomically flat terraces with the width of $400 \sim 1000 Å$ in STM images. A 99.999 % Fe wire was wrapped on an outgassed W filament and was outgassed again to remove the iron oxide. Before Fe was deposited, the sample was cooled to RT and held at RT during deposition. The Fe coverage was determined by the exposure time and areal coverage of deposited Fe in the STM images and Auger spectra. In most experiments, the deposition rate was set to be ~ 0.01 ML/s. During the Fe deposition, the pressure of the chamber was increased to 4×10^{10} Torr from the base pressure of $<1.5 \times 10^{10}$ Torr. The

results reported in this paper are with Fe/Cu(100) sample kept at RT during deposition and the image was taken at RT.

3. Results and Discussion

From recent experiments it has been proven that the island can be distributed with non-equilibrium distribution in several adsorption systems. Traditional adsorption model, therefore, has to be modified to explain the growth morphology [12, 13]. Scaling of the island size distribution for large diffusion rate depends on the detailed structure of islands shapes. The structure is strongly system dependent on every system of interest. In the island size scaling, the structural effect can usually be neglected. When atoms are deposited at random square lattice with arrival rate r, the atoms diffuse around till they meet others or an immobile island. With the consideration of hop rate h, an atom will hop around till it meets an island with size s equal or larger than 2 ($s \geq 2$) , producing an island with the size of $s+1$. (We call an isolated atom when $s=1$.) The island size distribution is in general determined by a ratio h/r. If it is large, atoms travel further and result in lower island density and larger average size. If we define N_s as the density of the island of size s and total island is defined by $N = \sum_{s \geq 2} N_s$ and s is the number of atoms in the island, the average island size can be defined by

$$s_{av} = \sum_{s \geq 1} sN_s / \sum_{s \geq 1} N_s = \theta/(n+N) \sim \theta/N \quad (1)$$

when $N \gg n$ where θ is the coverage and n is the particle density. The island size distribution varies greatly with temperature due to the change of hop rate. However for the large hop rate, the island size distribution N_s can be scaled by

$$N_s \sim \theta \, s_{av}^{-2} \, g \, (s/s_{av}) \quad (2)$$

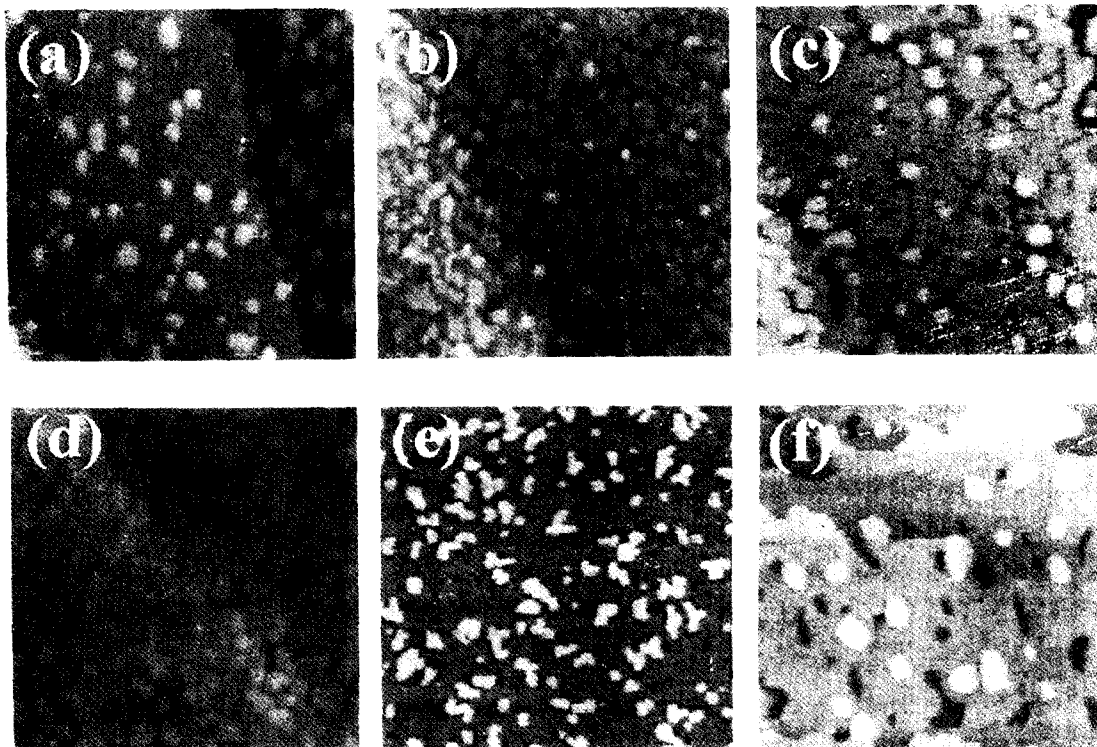where g is a scaling function. It means that the island size distribution can be represented by a sin-

Fig. 1. Morphology change of room-temperature grown Fe depending on coverage. Deposition rate is $0.5 \sim 1.5$ ML/min, $I_t = 500$ pA.   (a) 0.26 ML, 540×540 (Å^2), (b) 0.6 ML, 900×900 (Å^2), (c) 1.2 ML, 800×800 (Å^2), (d) 1.7 ML, 900×900 (Å^2), (e) 3.2 ML, 1500×1500 (Å^2), (f) 4.1 ML, 1200×1200 (Å^2).

gle function despite of coverage as far as they do not coalesce. Therefore if we plot the island density as a function of scaled island size, the curves at various coverage (but much less than 1 ML) should fall down to a scaled curve.

If Fe deposition on Cu(100) follows the scaling behavior, one can safely conclude the growth behavior is simply governed by deposition and ripening with a high hop rate, resulting a layer by layer growth. If not, the growth behavior is interrupted by other phenomena, such as interdiffusion. Fig. 1 shows the coverage dependent 6 STM images. At lower coverage (Fig. 1a, b), the images look more like layer by layer growth when we judge only by appearance without an quantitative analysis. From these STM image only, it is difficult to determine whether this images indicate the mixing of Fe and

Cu substrate. Earlier RHEED studies [3, 4] suggested that the growth behavior is perfect layer by layer growth at > 3 ML, but the oscillation is not observed or damped substantially at lower coverage. It was also from combined STM and Auger spectroscopy study [8, 9] that Cu substrate atoms segregate toward surface through pinholes which are observe in some STM images at low coverage. In the present experiment, the pinhole was occasionally observed but it is not clear Cu atoms diffuse through them as suggested by other results. Although this combined result [8, 9] are convincing, in one sense there is no guarantee that the same area was probed in Auger and STM studies since one probes 1 mm area and the other probes 10 nm area. Unfortunately, STM and STS (scanning tunneling spectroscopy) are not chem-
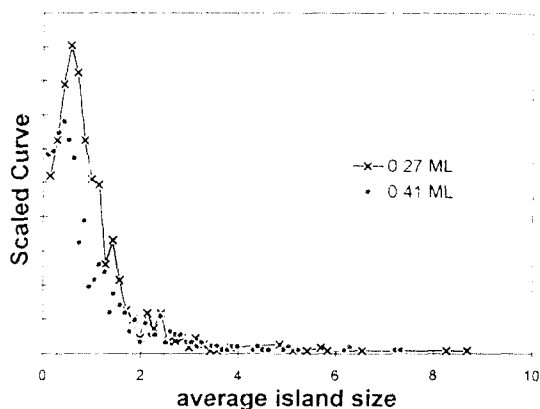
Fig. 2. Scaled curve obtained at the Fe coverage of 0.27 and 0.41 ML. X axis denotes $s/\langle s \rangle$ and y axis does N_s. Two curve should fall down to the same one but deviates because of mixing.

ical identity specific, therefore the proposed mixed first layer is hard to be determined by STM only. However, since simple layer by layer growth would follow a single scaling curve, it can be determined whether the grown structure is represented by a deposition and ripening process, which follows scaling behavior, or intermixing, which results in deviation from a scaling behavior. In this paper, the proposed scaling theory is used to determine the growth behavior for the first time in this system.

Above 3 ML, growth morphology shows a near perfect layer-by-layer growth, which would result in very slow decrease in the RHEED intensity envelope [3,4]. Strain-induced structures are observed at intermediate coverages (Fig. 1d, e, f). It was found by SMOKE study [5,6], it reveals a ferromagnetic behavior, unlike antiferromagnetic fcc Fe calculated by theory. Fe film is gradually relaxed from the strained epitaxial structure with increasing temperature and film thickness, restoring Cu lattice spacing in the Fe thick film between 4-11 ML. The tendency of layer by layer growth is enhanced before the structural transformation, suggesting the substrate effect is diminished above 3 ML. If one assumes any integral coverage (4 ML

for example) a perfect scaling behavior can be observed. When we annealed the surface, substantial mixing was observed [11].

In order to determine the mixing behavior, coverage dependent island size distribution was calculated. The island size was normalized by the average island size. The abundance of islands at each size was also calibrated by multiplying the square of average island size and dividing by the coverage. The scaling curve then should fall down to a single curve as described by Eq. 2 if the growth structure can be explained by layer by layer growth. The scaling curves at two different coverage is shown in Fig. 2. At 0.27 ML, the curve reveals a nearly perfect universal scaling. If the hop rate of Fe atoms is high (indeed the case), it should show the universal scaling. However at 0.41 ML, the scaling curve is rather distorted, the peak moves toward 0. The tendency is consistent up to 0.5 ML (the data not shown here). It can be explained by two ways; this system deviates from the scaling behavior due to strong structural dependence of Fe deposition on Cu(100) or Fe atoms displace the substrate Cu atoms and Cu diffuse out of the substrate plane. For the first possibility, there is not much reason why this system should be different from Fe/Fe system in terms of pair interaction [12]. Therefore, it is safe to conclude Fe atoms diffuse into the substrate while the substrate atoms diffuse out. This behavior was also observed in previously studied Mn/Cu(100) and Pd/Cu(100) system [9], resulting surface alloys. The scaling curves move toward to 0 with increasing coverage up to 0.5 ML but it is difficult to use the scaling theory above the coverage because of percolation behavior and the appearance of the second layer. In order to cross check the present conclusion, one may consider a Auger in nanometer scale or applying the scaling theory to a system where two elements can be identified by STM. As many understood the mixing at low coverage by macroscopic Auger [8,9], the present

study open a possibility of using a scaling theory to determine the mixing behavior.

4. Summary

The structure of fcc Fe film grown on the Cu (001) surface was investigated using scanning tunneling microscopy. Room temperature growth is predominantly governed by the layer-by-layer growth. At low coverage of <1 ML, the epitaxy is rather poor and possibility of Fe-Cu intermixing at the first layer is noticed from the scaling-theory results, which is contrasting with the recent RHEED study. Above 4 ML, layer-by-layer growth is markedly improved as observed by MEED oscillation. Contradictory results could be due to atomic defects or dislocations on prepared sample. Scaling theory can be used to determine the mixing of the overlayer and substrate at low coverage.

Acknowledgment

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