Dynamic Observation of Sn/Si(111) Surface Structures by Low Energy Electron Microscope*

Fangzhun Guo,† Keisuke Kobayashi, and Toyohiko Kinoshita
Japan Synchrotron Radiation Research Institute, SPing-8,
Kouto 1-1-1, Mikazuki, Sayo, Hyogo 679-5198, Japan
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Dynamic surface structures of Tin (Sn) on Si(111) were studied by using low energy electron microscope (LEEM). Different from the reported phase diagram, $(\sqrt{3} \times \sqrt{3})$ -$R30^\circ$ Sn structure is found existing only in a narrow Sn film thickness range and disappearing quickly over 1/3 monolayer (ML) Sn deposition at substrate temperature of 400°C. Sn islands, which are different from the S-K mode growth, begin to grow from around 1/5 ML along steps or domain boundaries. The models of Sn growth on Si(111) are proposed. [DOI: 10.1380/ejssnt.2005.350]

Keywords: Low-energy electron microscopy (LEEM); Low energy electron diffraction (LEED); Surface structure, morphology, roughness, and topography

Growth of Sn on Si attracts much interest for the colorful phase structures and the application as a surfactant to control the self-assembly technique with Stranski-Krastaner (SK) growth [1–3]. Although previous papers had reported the surface structures of Sn on Si(111) and provided models [4–6], few dynamic observations have been reported. Moreover, the traditional surface structure analysis, such as Scanning Tunneling Microscope (STM) [7], LEED-Auger, etc. may miss some processes during Sn growth. In recent years, LEEM has been widely used for surface studies. The low energy electron originated surface sensitivity, the selected area structure analysis and the dynamic surface growth observation make LEEM possible to study the surface comprehensively. In this paper, we report the dynamically observed surface structures of Sn on Si(111) by LEEM (ELMITEC manufactured LEEM III) [8, 9] and propose growth models.

N-type Si(111) wafers (5-10 Ωcm) was used as substrate. The substrate was cleaned by flash direct-current heating at about 1200°C and cooled down immediately. This treatment resulted in a clean Si(111) 7×7 and 1×1 complex structures on which atomic steps and atomically flat terraces could be produced. During Sn deposition, the substrate was kept at temperature 400°C and Sn was deposited onto the substrate using electron bombard evaporator in the analysis chamber. The deposition rate was estimated as 2.09 ML/h supposing the sticking probability is unity through the low energy electron diffraction (LEED) spot intensity analysis.

Sn reconstruction $(\sqrt{3} \times \sqrt{3})$-R30° ($\sqrt{3}$ in short) appears under sub-monolayer Sn deposition on Si(111) 7×7 surface and the intensities of $\sqrt{3}$ spot changing as Sn deposition are shown in Figure 1. Here, the intensities are defined as the background subtracted (2/3, 1/3) spots from the LEED patterns obtained synchronously during Sn deposition. The incident electron energy is 25 eV. $\sqrt{3}$ reflections grow in intensity starting from about 0.1 ML and reaches its peak at Sn deposition 1/3 ML (1ML =7.81 × 10¹⁴ atoms/cm²). Upon further Sn deposition, $\sqrt{3}$ structure disappears quickly. Our experimental results show that the $\sqrt{3}$ structure exists only in a narrow Sn film thickness range. It is different from the previous report that $\sqrt{3}$ structure coexists within wide Sn coverage until about 1 ML in wide substrate temperature [4]. Different from the automatically LEED measurement by using LEEM, Ichikawa, et al. studied the structures of different Sn thicknesses during annealing process by RHEED (reflection high energy electron diffraction) measurement. Some effects, for example Sn desorption, during the annealing process are thought to be the reasons for the discrepancy to our experiment results.

After measuring the series LEED patterns, the substrate was flashed again to obtain the same clean surface. Next, LEEM images were taken automatically using the same evaporation ratio and substrate temperature as those used for LEED experiments. The incident electron energy is 11.4 eV and the main contrast mechanisms are the different diffraction intensities from different surface structures.

Based on the LEED spot intensity behavior shown in Figure 1, LEEM images corresponding to five typical Sn depositions 0, 1/5, 1/3, 1/2, 3/4 ML are shown in Figure 2. The corresponding growth models are supposed and illustrated on the right side. All the LEEM images were

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†Corresponding author: fz-guo@spring8.or.jp

FIG. 1: Diffraction spot intensities of $(\sqrt{3} \times \sqrt{3})$ -$R30^\circ$ structure change as Sn deposition. The incident electron energy is 25 eV.
FIG. 2: LEEM images with field of view of 5 µm and the proposed modes corresponding to five typical Sn depositions 0, 1/5, 1/3, 1/2, 3/4 ML are shown.

For clean Si(111) surface (0 ML), three-fold symmetry 7×7 (bright) and 1×1 (dark) structures are observed in the LEEM image. The 7×7 surface structures are separated by domain boundaries on atomically flat terraces, while 1×1 structures mainly lie along the atomic steps. At the Sn deposition of about 1/5 ML, the contrasts in the LEEM image between 7×7 and 1×1 structures become diffuse due to the diffraction intensity decrease. Thinking about the √3 diffraction spot intensity shown in Figure 1, Sn atoms with √3 structure are considered to appear on 7×7 surface at random distributed small areas, while Sn atoms deposited on Si 1×1 surface have the same 1×1 arrangements. When the Sn deposition reaches 1/3 ML, the LEEM image shows almost no contrast because the Sn √3 structure covers almost all the Si 7×7 surface, and the Sn 1×1 structure covers the Si 1×1 surface completely. For √3 surface structure, Sn atoms occupy the position of T4 model between the two types of three-fold triangular sites lying in the last plane of a (111) Si double layer, which contains a Si atom directly in the bottom of the double layer. In the LEEM image with 1/3 ML Sn deposition, four bright Sn islands are observed lying along steps or domain boundaries. The islands are formed because the coming Sn atoms are trapped near steps or domain bound-
aries, and the surrounding Sn atoms gather together on the 400°C substrate. These islands are not the S-K mode growing islands whose on-set coverage is reported as 1.6 ML [10, 11]. In the LEEM image of 1/2 ML Sn deposition, the former 7×7 structure areas become dark with bright domain boundaries. Reminding the decrease of $\sqrt{3}$ diffraction spot intensity at 1/2 ML shown in figure 1, the structure seems destroyed and turns to disordered surface structure. Some of the Sn atoms gather along steps or boundaries, thus makes the steps and boundaries bright. The former Si 1×1 areas have higher intensity indicating that the Sn atoms on them keep the same 1×1 structure as Sn deposition. The structure of Sn atoms around steps or domain boundaries are thought having the same 1×1 structure because of the same contrast as that on Si 1×1 surface. Moreover, the marked big Sn island size looks smaller at 1/2 ML deposition than that at 1/3 ML. It is clearly observed that the island decomposes partially and flows along the former 7×7 domain boundaries. The flowing Sn atoms make the boundaries near the island wider than those far from it. At the Sn deposition 3/4 ML, the former 7×7 areas are covered by more Sn 1×1 areas and the islands begin to grow again. All the surface are thought to be covered by Sn 1×1 structure for 1 ML Sn deposition.

Dynamic surface structures of Sn on Si(111) surface were studied by using LEEM. The in situ observation during Sn deposition makes it possible to follow both the structures and surface changes accurately. The $\sqrt{3}$ structure is found existing only in a narrow Sn film thickness range, and a disordered phase seems existing between $\sqrt{3}$ and 1×1 structures on the Si 7×7 atomically flat terraces. Sn islands are found forming below the on-set coverage of S-K mode island growth. Micro-size LEED observations are necessary for further surface structure decision of Sn on Si(111).

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