Self-organized Growth of Nanopucks on Pb Quantum Islands

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Abstract

Electronic Moire patterns found on lead (Pb) quantum islands can serve as a template to grow self-organized cluster (nanopucks) arrays of various materials. These patterns can be divided into fcc- and hcp-stacked areas, which exhibit different binding strengths to the deposited adatoms. For Ag adatoms, the binding energy can differ substantially and confined nucleation thus occurs in the fcc sites. Both the size distribution and spatial arrangement of the Ag nanopucks are analyzed and found to be commensurate with the characteristics of the template island, which exhibits a bi-layer oscillatory behavior.

Several routes were taken to search for a suitable template on which the self-organized growth could be realized in practice [1-3]. In this work, we demonstrate for the first time that a regular pattern of electronic origin can serve as a template for the growth of self-organized nanostructures. We also provide evidence that the growth of self-organized Ag nanostructures reflects the characteristics of a Pb quantum island substrate [4].

The experiments were carried out in a UHV chamber where the base pressure was less than $5 \times 10^{-11}$ torr. The chamber was also equipped with a variable...
temperature scanning tunneling microscope and two well-collimated e-beam evaporators for depositing high purity Pb and Ag atoms. A clean Si(111)-7×7 surface was first prepared and over one monolayer of Pb was evaporated onto the 7×7 at room temperature, followed by annealing at 480°C for a few seconds to generate the stripe incommensurate phase (SIC) [5]. The sample was then cooled to 200 K and an extra amount of lead was further added to generate Pb quantum islands of various thicknesses. To form nanoclusters on the Pb quantum islands, we deposited a suitable amount of Ag while the sample was held at different temperatures. STM observations and measurements were carried out after the deposition.

The characteristics of the template employed in this study for the further growth of two-dimensional nanoclusters are detailed in ref. 6 and summarized with the STM images shown in Figure. 1. The prominent apparent corrugation of the superstructures observed on the islands of three atomic layers (Figure. 1(a)) is beyond simple geometrical consideration. In addition, the larger islands in this image are all three layers thick above the Si substrate, yet one can immediately distinguish two types of islands (as marked) with the image contrast of the superstructure. Since the only dissimilarity between the two types of islands results from the different stacking sequences of the grown films with respect to those of the Si substrate, geometry effect alone cannot explain such a huge disparity in their apparent heights as measured by STM. This is the first indication of which the electronic origin should take hold.

The second indication of the electronic origin is that the contrast of the pattern also strongly depends on the bias voltage as shown in Figure. 1(b). It is more obvious for the type II islands where the contrast goes from weak to strong as the bias changes from 2 V (upper image) to 0.4 V (lower image). Furthermore, the image contrast also varies with the island thickness (Figure. 1(c)), which provides the third piece of evidence of the electronic origin of the superstructures. At the same bias voltage, the image contrast will alternate between the high and the low as the island grows one more layer. This bi-layer oscillatory behavior can be correlated to the phase shift of the confined electrons [6].

These patterns found on the Pb quantum islands of electronic origin can serve as a template for self-organized growth of nanostructures. The STM image of deposited Ag atoms at 120K on the Pb island of three atomic layers demonstrates this as
shown in Figure 2. It can be seen that Ag atoms form a very ordered periodic cluster array with a uniform size, indicated by the size distribution curve (Figure 2(b)) and the Fourier-transfer pattern of sharp spots (inset in Figure 2(a)). A closer examination of the structure of these clusters shows that they are of one layer in height and have either an imperfect hexagonal shape or a roughly circular shape. We thus call them nanopucks. Taking the nanopucks in Figure 2(a) as an example, the standard deviation (STD) of the size distribution after normalizing with the average size is only 0.11 at a coverage of 0.2 monolayers (ML). If there were no template, the formation of nanoclusters on a crystalline surface would be governed only by surface adatom diffusion, and the size distribution (STD=0.59) according to the scaling law [7] would be much wider as displayed in Figure 2(b). Thus, the superstructure must have provided extra diffusion barriers to confine the nucleation of the nanopucks. This fact is also reflected in the positions of the nanopucks. They are not random and tend to occupy only one half of a unit cell (see Figure 2(c)). The rhombic unit cell can be divided into two triangular halves by the black dashed lines. One has the face-centered cubic (fcc) stacking and the other hexagonal close-packed (hcp) stacking (Figure 2(d)). For many heteroepitaxial systems, it is generally accepted that adsorption favors the fcc side. It also indicates a strong binding difference between two half triangular cells.

It is also desirable to obtain quantitative values specifying the trapping strength of the template. For the current system the number of atoms in a critical nucleus is assumed to be 1, which is pertinent to many cases of Ag deposited on metal systems. We then follow the analytical procedure of Ref. 1 and apply the nucleation theory for complete condensation [8]. The saturated island density (N) is proportional to \( \exp\{E_d/kT\} \), where \( E_d \) is the activation energy for surface diffusion, and \( k \) a fitting parameter accounted for diffusion on an inhomogeneous substrate. For the Ag/Pt(111) system, however, we find the above equation can become immediately applicable if \( k = 1.8 \) is taken for that system. Experiments were performed by depositing a suitable amount of Ag onto quantum islands at various temperatures as shown in Figure 3(a)-(c), corresponding to three regions of different slopes in Figure 3(c). The amounts of coverage have been determined to result in a saturated island density. From the Arrhenius plots of N versus 1/T for Ag nanopucks formed on Type I Pb islands of three atomic layers in Figure 3(e), we can derive the conventional site diffusion within the unit cell \( E_a = 55 \) meV and the adatoms can overcome the barrier between two half cells \( E_d = 340 \) meV for Ag nanopucks nucleated on the Type I Pb islands of three layers.

Furthermore, a prominent difference has been observed for Ag nanopucks grown on the islands of dissimilar types.
Figure 4(a) one can quickly set apart the Ag nanopucks grown on the Type II island (marked on the top left of the figure) by their randomness of the size and arrangement. For the Type II island, with a similar analysis, we find that the cross-cell barrier energy $E_d$ reduces to around 150 meV and the site diffusion barrier $E_\delta$ to about 50 meV.

Comparing the Ag nanopucks grown on the four-layer islands of Type I and II in Figure 4(c), we discover that those on Type II have smaller and more uniform sizes and their arrangement is also more orderly, which is just opposite to what was found for Ag nanopucks on the islands of three atomic layers. According to the tabulated values in Figure 4(b), it is evident that the growth orderliness of Ag nanopucks reflects the image contrast of the substrate’s superstructure.

We have also made attempts in growing nanopucks of other materials, such as Pb and Al. The trapping strengths of the superstructure templates for Pb and Al adatoms are nearly equivalent and comparable to those of the surface dislocation templates. This suggests that the Ag case may contain more electronic interactions between the clusters and the substrate, which render a stronger chemisorption likely involving charge transfer. This electronic character has also made the Ag nanopucks grown on two types of quantum islands quite distinguishable while, for Pb and Al nanopucks, no similar difference can be observed.

This work was supported by the NSC, Academia Sinica, and the academic excellence program of the Ministry of Education of Taiwan, ROC.


References: