Correlation between morphological transition and preferred thickness of Pb and Ag islands on Si(111)7×7

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It is known that a quantum size effect can induce a morphological transition of Pb nanostructure, from three-dimensional clusters to multilayer two-dimensional islands, grown on the Si(111)7×7 at low temperature. We use scanning tunneling microscopy to *in situ* observe the formation of an individual island to figure out the transition process. Our results reveal that every island differing in thickness can be correlated with a unique transition pathway. A similar behavior is also observed in the growth of flat Ag islands on the Si(111) 7×7 substrate at room temperature.

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For technological applications, the growth of a metal film on the semiconductor is often required to be atomically flat and of uniform thickness. This expectation has recently been fulfilled in some metal/semiconductor systems. For example, using the procedure of low temperature deposition followed by annealing to room temperature, flat Ag films with preferred thickness of seven and two atomic layers can be created on GaAs(110) and Si(111)7×7 surfaces, respectively.[1,2] Flat top Pb islands with thickness of two layers can be directly grown on the incommensurate Pb/Si(111) surface at low temperature.[3] A common characteristic of the grown flat films in those systems is the appearance of a single preferred thickness, which is now attributed to the quantum size effect (QSE) of electrons in general, and such growth phenomena are termed the electronic growth.[4] Although growth of Pb on the Si(111)7×7 at low temperature can also form flat top islands, in comparison with the above systems, it reveals an abnormal thickness distribution in the range of 4–9 layers instead of one single thickness.[5] This unusual growth system at room temperature follows the Stranski-Krastanov mode, i.e., 3D growth after the completion of a wetting layer.[6] Even though at low temperature the QSE drives the growth of Pb islands into the quasi-two-dimensional,[7] STM observations in Ref. 8 have demonstrated that the 3D-cluster growth still governs the initial growth stage. Thus the formation of 2D islands involves a 3D-to-2D morphological transition. In that report, however, the pathway of 3D clusters transforming into 2D islands at various heights remains unanswered. We believe that the cause of the dispersion in island thickness can be understood if the transition pathway for the island of each thickness is traced out. This motivates us to *in situ* observe the growth process of an individual island with STM. Our observations show that growing clusters can transform into islands via several transition pathways that result in a multiple-thickness distribution. The different pathways originate from a sporadic timing of the 3D-to-2D transition. The timing of transition for a thinner island is earlier than that for a thicker island. We will discuss later that this timing fluctuation may be associated with the interface properties of Pb/Si(111)7×7. Flat silver islands with the single preferred thickness of two atomic layers can also be directly grown on the Si(111)7×7 surface at room temperature, and the growth is two dimensional.[9] The quantum-well states in the epitaxial Ag film on the Si(111)7×7 have been observed,[10,11] thus the formation of 2-layer Ag islands would also be driven by the QSE. These observations imply that there exists a morphological transition with a single pathway in the formation of Ag islands. We will show that this argument can be verified by the scaling theory.[12]

In our experiment, a Si(111)7×7 surface was obtained by annealing the sample to 1200 °C followed by slow cooling to room temperature. Lead was deposited *in situ* onto the sample on STM stage at 170 K, which allowed us to observe

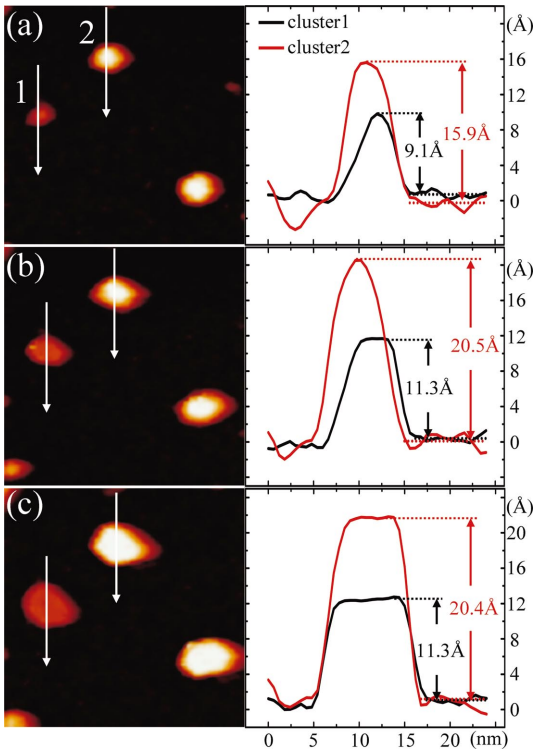

FIG. 1. (Color) *In situ* observations for the growth of two 3D clusters as marked by the numbers in (a). Line profiles at the right side of (a)–(c) represent the morphology evolution of cluster 1 and 2 along arrows in (a). The image size is 52×52 nm^2.

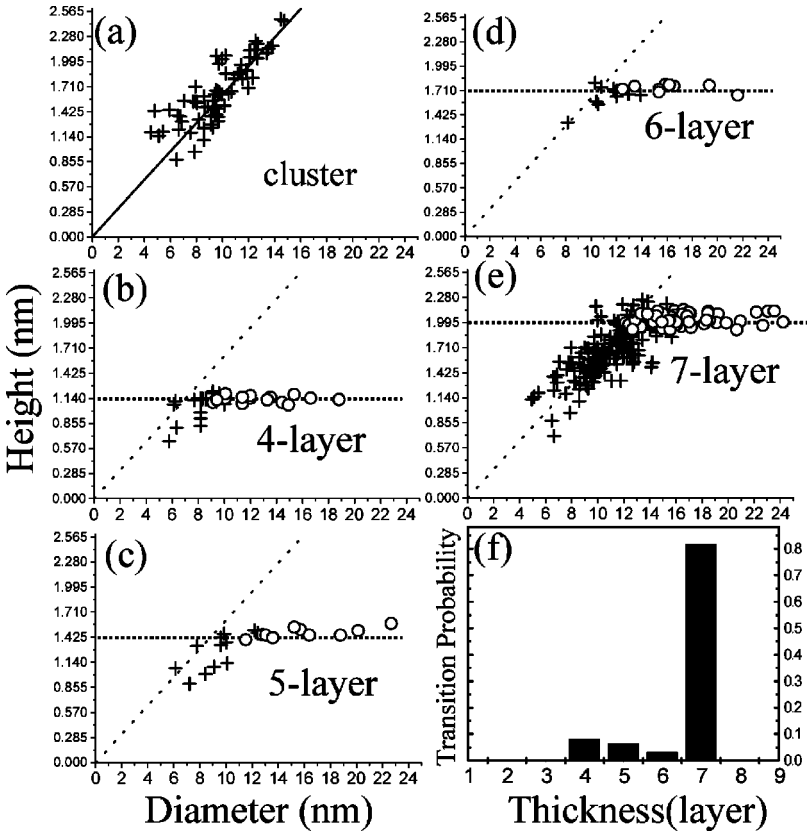

FIG. 2. (a) A statistic result of the height as a function of the base diameter for clusters without undergoing the growth transition. It can be fitted by a linear line to show the 3D growth of clusters. (b)–(e) Clusters experiencing the transition are separated in terms of the subsequent island thickness, which is also represented with the height as a function of the diameter for both clusters (cross) and islands (open circle). (f) The transition probability of clusters transforming into islands of different thickness.

the same area after the deposition. The flux of evaporated lead was 0.16 ML per minute and the pressure during the deposition was kept below 2×10^{-10} torr. Silver was deposited onto the Si(111)7×7 surface at room temperature with a flux of 0.06 ML per minute.

Figures 1(a)–1(c) show *in situ* observations of the growth of two 3D clusters as marked by numbers in Fig. 1(a) with increasing coverage. Line profiles at the right side of corresponding STM images represent the morphology evolution of cluster 1 and 2 along the arrows. After twice deposition of 0.02 ML, cluster 1 with a height of 9.1 Å in Fig. 1(a) transformed into a flat-top island with a 11.3 Å thickness, then grew to a larger island of the same thickness, as shown in Figs. 1(b) and 1(c), respectively. On the other hand, cluster 2 of 15.9 Å height grew into a higher cluster of 20.5 Å height, then transformed into an island with a 20.4 Å thickness. Those *in situ* observations are consistent with previous studies, where the existence of a 3D-to-2D growth transition in the formation of Pb islands was demonstrated.[8] However, Fig. 1 reveals that an island thickness can be directly correlated to a cluster height before transition, e.g., a thinner island being transformed from a lower cluster. It indicates that each preferred thickness of an island can correspond to an independent transition pathway. We thus accumulated hundreds of transformation events to find out the correlation between a transition pathway and a preferred thickness. Figure 2(a) is a statistical result of the heights as a function of the base diameters for the clusters that are not undergoing the growth transition. On the average, the height is linearly proportional to the diameter, exhibiting a 3D growth nature. Clusters experiencing the transition are separated in terms of the subsequent island thickness. Figure 2(b) shows the events of clusters finally transforming into 4-layer islands, which is also represented with the height as a function of the diameter for both clusters (cross) and islands (open circle). The diameter of an island can vary from 10 nm to 20 nm, indicating that once a 4-layer island is formed, it starts to grow in size laterally. Right before the transition, the height of the cluster reaches 1.14 nm, which is four atomic layers in terms of 0.285 nm interlayer spacing. This implies the cluster is necessary to gain the equivalent height before becoming 4-layer islands. The lateral growth behavior and equivalent height are also observed in the events of clusters transforming into the 5-, 6-, 7-layer island as shown in Figs. 2(c)–2(e). We therefore conclude that the transition pathway should be that an *N*-layer island is transformed from an *N*-layer height cluster. Islands of each thickness thus have a unique growth path. The distribution of cluster heights versus sizes in Figs. 2(b)–2(e) follows a linear dash line, which is the same as the solid line in Fig. 2(a), indicating the clusters experiencing the transition are identical to the clusters maintaining 3D growth. The height distribution of clusters in Fig. 2(a) covers all island thickness, reflecting that the fate of a growing cluster has two choices: one is to follow the transition pathway to become an island; the other is to continue to grow into a higher cluster. Therefore, the transition timing for the formation of a thinner island is earlier than that for forming a thicker island. The multiple-thickness distribution thus basically manifests a sporadic nature in the timing of the 3D-to-2D transition. Figure 2(f) shows transition probabilities of the clusters transforming into the islands of different thickness. The transition probability for 7-layer islands is much

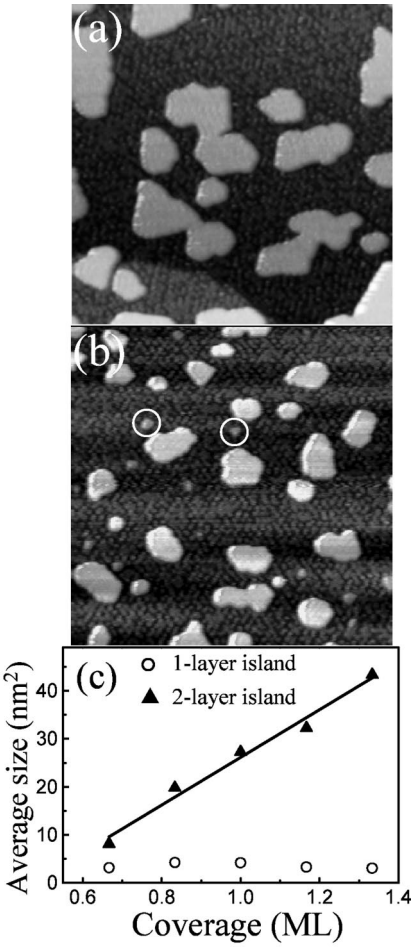

FIG. 3. (a) The growth of flat Ag islands with a preferred thickness of 2 atomic layers on Si(111)7×7 surface at room temperature at the coverage of 1 ML. (b) At the coverage of 0.6 ML, not only 2-layer islands but also 1-layer islands, as marked by circles, are formed on the surface. (c) Average sizes of 1-layer and 2-layer islands as a function of coverage. Both image size in (a) and (b) are 75×75 nm^2.

larger than those for the islands of other thicknesses, this is the reason why the 7-layer island is observed to be the most abundant in previous studies.[3,7] Although the probabilities for forming 4-, 5- ,6-layer islands are small, their existence reveals an interesting point: identical clusters might possess dissimilar electronic structures.

In contrast to the islands of multiple thicknesses in Pb/Si(111)7×7 growth system, Pb islands formed on the incommensurate Pb/Si(111) surface at a similar coverage and temperature clearly demonstrates a single thickness of two atomic layers.[3] This difference can be attributed to the different Pb/Si interface properties.[13] Recently, two studies by Altfeder *et al.* have shown that periodic patterns, observed by STM on the top surface of Pb islands grown on both surfaces, can be directly related to the structures of these two interfaces.[14,15] The manifestation of periodic patterns on Pb islands is because the quantization of electrons along the surface normal is not coherent but changes with the location at the interface. The *I-V* spectra measured on the islands indeed reveal that the quantum-well states vary with the site

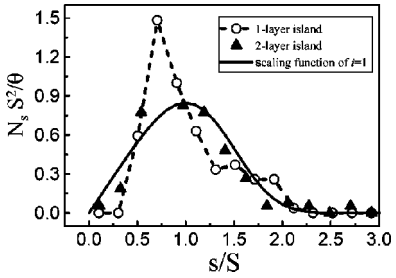

FIG. 4. Scaled size distributions of 1-layer and 2-layer Ag islands. The dash lines connecting data points of 1-layer islands are drawn for the purpose of guiding the eye. The solid curve is the theoretical scaling function of one-atom critical size.

for both systems. For the island (10 nm height) grown on Pb/Si(111)7×7 wetting layer at 0 °C,[14] their STM images show a pattern of the 7×7 reconstruction on the island, and the quantum-well state appears at the adatom site, but disappears at the corner hole and dimer row sites on the pattern. This experimental result reflects that the Pb/Si(111)7×7 interface is not uniformly perfect for confining electrons. Electrons at the corner holes and dimer rows of the 7×7 reconstruction are only weakly reflective. Quantized and non-quantized electrons thus coexist in the Pb film. This observation guides us to explore the origin of the timing fluctuation in the morphological transition. When a 3D cluster is created on the Pb wetting layer, its electronic structure would naturally consist of quantized and non-quantized electrons because of the interface imperfection. The strength of the QSE in clusters of even the same thickness can be different, depending on the base area covered by each cluster. The driving force for forming a 3D cluster originates from different surface energy between silicon and lead, which should be the same for every cluster. The occurrence of the transition marks the onset of surpassing of the QSE over the 3D growth driving force in the cluster. Since the strength of the QSE in each cluster can be varied, the transition timing may thus fluctuate. Therefore several transition pathways correlating to the preferred thickness of islands basically originates from the interface imperfection. Following to this argument, we propose a way to eliminate the timing fluctuation, i.e., to prepare a better interface for quantum confinement. This argument can be examined by another STM result from Altfeder *et al.*[15] for the Pb island (1.5–10 nm height) grown on incommensurate Pb/Si(111) surface. Their STM images show several superlattice patterns, reflecting interface structures, but there is no observation of disappearance of quantum-well state feature in their *I-V* measurements on the patterns. It indicates that this interface, the incommensurate Pb/Si(111), is better in establishing the quantum confinement of electrons than the Pb/Si(111)7×7 interface. The island growth on incommensurate Pb/Si(111) surfaces indeed reveals the characteristic of a single preferred thickness, thus is consistent with our argument.

Flat top Ag islands of 2-layer thickness can actually grow on the Si(111)7×7 surface at room temperature directly,[9] which is unnecessary to adopt the procedure of low temperature deposition followed by annealing to room temperature. Figure 3(a) is an STM image of formation of Ag islands at

the coverage of 1 ML silver, showing a uniform thickness of 2 atomic layers. In the observations of Sobotík *et al.*, at low coverage, there also exist 1-layer islands as marked by the circles in Fig. 3(b), but only 2-layer islands exhibit the 2D growth behavior with coverage. Figure 3(c) shows that the average size of 1-layer islands almost remains at a constant whereas that of 2-layer islands increases linearly with the coverage, which is consistent with the results of Sobotík *et al.* Since the quantum-well states in the epitaxial Ag film on Si(111)7×7 have been observed by photoemission studies[10,11] the 2D growth behavior of 2-layer islands is also likely driven by the quantum size effect. The scaling theory[12] is useful to explain the size distribution of single-layer islands in homoepitaxial system,[16,17] and a recent study has demonstrated that it can also be applied to describe the size distribution of multilayer 2D islands.[8] We thus apply the scaling theory to analyze the size distribution of 2-layer Ag islands. Figure 4 shows that the scaled data of size distribution (solid triangle) and they can be well fitted by the scaling function of critical size 1. We also measure the size distribution of 1-layer islands but the scaled data (open circle) deviate from the scaling function as shown in Fig. 4. Because both 1-layer and 2-layer islands are grown at room temperature, they are supposed to nucleate from the same critical size. The disagreement between the scaled size distribution of 1-layer islands and the scaling function implies that the size of 1-layer islands is constrained by a 3D growth driving force. When a 1-layer island reaches a certain size, it can grow upward into a 2-layer island. The 3D growth is then suppressed by the 2D growth right after the formation of 2-layer islands. This process can explain why the average size of 1-layer islands is independent of the coverage, and only a single transition pathway exhibits in the formation of 2-layer Ag islands.

In conclusion, we have observed that each preferred thickness for 2D island growth induced by the QSE corresponds to a unique transition pathway. For the formation of Pb islands on the Si(111)7×7 substrate, several independent pathways guide the 3D clusters to transform into the 2D islands at a scattered thickness. From our current understanding, the origin of various transition pathways most likely lies in the imperfection of the Pb/Si(111)7×7 interface for quantum confinement. Using the scaling theory, a single transition pathway for forming 2-layer Ag island on Si(111)7×7 is explored. In comparison with various thicknesses of Pb islands, the single preferred thickness of Ag islands suggests that the Ag/Si(111)7×7 has a better interface than the Pb/Si(111)7×7 for confining electrons.

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