Formation of multilayer two-dimensional Pb Islands on Si(111)7×7 at low temperature: From nucleation to growth

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The morphology of Pb islands formed on Si(111)7×7 at low temperature has the characteristics of both three-dimensional (3D) and 2D structures as they are of multilayer thickness but have a flat top surface. We use scanning tunneling microscopy to observe the growth behavior from the incipient stage to the formation of islands. Our observations show that the growth is 3D (Stranski-Krastanov mode) right after the nucleation, but that there is a transition from 3D to 2D when the nucleus reaches a certain size. This transition is driven by the quantum size effect, and it changes the growth behavior of Pb islands to be similar to that of single-layer islands in homoepitaxy systems. This similarity allows us to apply the scaling theory to analyze the size distribution of Pb islands, and also to conclude the critical size to be three atoms at the nucleation stage.

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Low temperature deposition has been an effective way of producing a flat metal film on a semiconductor surface. For example, combining the low temperature deposition with the subsequent process of annealing to room temperature, a flat Ag film with a critical thickness can be created on the GaAs(110) surface.[1] Later, the electronic growth model explains that this growth behavior is driven by the quantum size effect of electrons in the surface normal.[2] That is, the nanocluster film formed by low temperature deposition becomes a crystalline film with annealing, and as well as the quantum size effect appears in the Ag film, causing the film to be flat and be of a critical thickness. Recent studies by Spot-Leed have demonstrated that Pb islands with the features of the steep edge, flat top surface, and preferred thickness of seven atomic layers can be directly created on Si(111)7×7 surface without further annealing.[3] This growth behavior has been directly observed by scanning tunneling microscopy.[4,5] It is believed that this unusual growth behavior also originates from the quantum size effect; however, Pb islands directly formed without annealing implies that the manifestation of the quantum size effect is not necessary in the thin film structure. Therefore, this growth process must be different from that of an Ag/GaAs(110) system, and it was not clearly described in previous studies. This motivates us to explore the process of how Pb islands are created from initial nucleation to growth.

Our observations show that after the nucleation, the growth follows a Stranski-Krastanov (SK) mode to form three-dimensional (3D) clusters on a wetting layer. However, we also observe a transition from 3D to 2D growth. The transition indicates that the quantum size effect begins to influence the growth behavior of 3D clusters. This effect suppresses the vertical growth and enhances the lateral growth of the clusters. The combination of the SK growth and the quantum size effect results in the subsequent morphology of Pb islands to be of a multilayer thickness and a flat top surface. In the low coverage limit, we have observed that the average island size is insensitive to the island thickness. This property reflects that after the quantum size effect governs the growth, the island size of the different thickness grows nearly at the same rate. Thus we can view these multilayer islands effectively as single-layer 2D islands. The scaling theory[6] is powerful to investigate the critical size of single-layer 2D islands. We present an argument that it is also possible to analyze multilayer islands with the same scaling theory. The size distributions of multilayer flat top Pb islands at different coverages can be scaled into an universal distribution agreeing with the scaling function of the critical size of three atoms. This implies that the Pb tetramer is the smallest stable nucleus. In terms of these observations and analysis, the entire process of Pb island formation from nucleation to growth can be understood.

In our experiment, Si(111) samples were cleaned in ultrahigh vacuum at a base pressure of 5×10^{-11} Torr. The reconstructed 7×7 surface was obtained by annealing the sample to 1200 °C followed by slow cooling to room temperature. Lead was evaporated *in situ* onto the sample at 170–210 K with a flux of 0.16 ML per minute. The pressure was kept below 2×10^{-10} Torr during the deposition.

Figure 1(a) is a typical scanning tunneling microscope (STM) topography image of a Pb film grown at 208 K with a coverage of 3.52 ML. Before the formation of islands, about 2 ML of Pb are consumed in wetting the Si(111)7×7 substrate. The rest of Pb grows into islands above the wetting layer. The surface of the islands is very flat and its orientation is along the [111] direction.[7] The relative abundance ratio distribution as a function of island thickness shown in Fig. 1(b) reveals that the thickness is confined to 4–9 atomic layers. Islands with a seven-layer thickness are the most abundant, indicating this is the magic thickness. There is no island with a thickness below four layers, indicating the critical thickness is four layers. That critical thickness is far more than one layer already implies the initial growth is 3D. On the other hand, the existence of the magic thickness and a flat top surface indicates that, after some time, the 3D growth will stop, and the subsequent growth is governed by the lateral growth. Therefore, there must exist a transition of the growth from 3D to 2D.

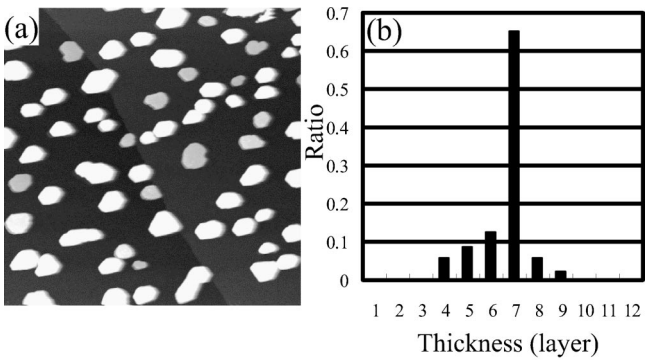

FIG. 1. (a) STM image of Pb islands formed on Si(111)7×7 surface at 208 K by depositing 3.52-ML Pb (image size: 300 ×300 nm^2). (b) Ratio distribution as a function of island thickness, showing the thickness is confined to 4–9 atomic layers and that islands with a seven-layer thickness are the most abundant.

In order to confirm the existence of the 3D-to-2D growth transition, we observe the growth stage right after the completion of the wetting layer at a lower temperature of 170 K. Figure 2(a) shows that clusters and 2D islands are formed concurrently on the wetting layer at a coverage of 2.32 ML. These clusters are the seeds for growing 2D islands at a higher coverage. The line profile across a cluster and an island in Fig. 2(a), indicated by an arrow, shows the 3D morphology of the cluster and flat top surface of the island.

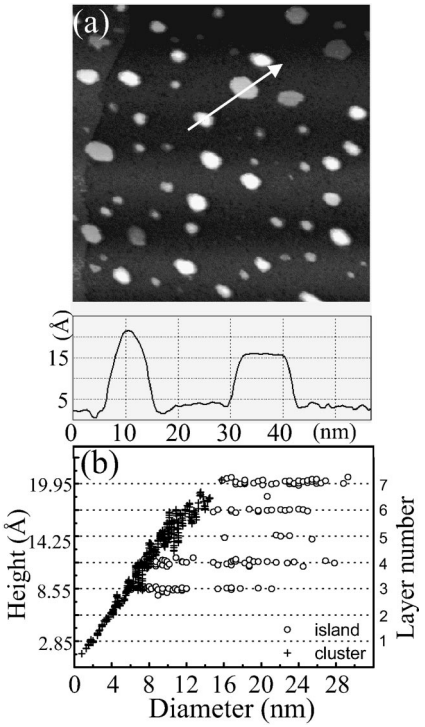

FIG. 2. (a) STM image with 2.32-ML Pb on the Si(111)7×7 surface at 170 K, showing 3D clusters and 2D islands are formed concurrently on the wetting layer. The line profile across a cluster and an island, indicated by an arrow in the STM image, shows a 3D morphology and a flat top surface, respectively (image size: 300 ×300 nm^2). (b) A statistic result of the height as a function of the diameter for both 3D clusters (cross) and 2D islands (open circle).

We sample hundreds of clusters and islands to measure their diameter and height. Figure 2(b) shows the height as a function of the diameter for clusters and islands. The distribution can be separated into two regimes. In the first regime (cross), the height is linearly proportional to the diameter, demonstrating the 3D growth feature of the clusters. In the second regime (open circle), several discrete heights, corresponding to the layer number of the islands, are independent of the island size, showing the 2D growth feature of these islands irrespective of the thickness. Therefore, there indeed exists a transition from 3D to 2D in the growth process. Lead is known to grow into 3D islands on the Si(111)7×7 surface at room temperature with the SK growth mode.[8] Though in our case, the growth is performed at low temperature, after nucleation, the amount of atoms in the nucleus is still too small to establish the quantum size effect strong enough to affect the growth. Therefore, the growth is still dominated by the SK mode, which causes the nucleus to grow into a 3D cluster. Once the aggregated atoms of the cluster are large enough to incite the quantum size effect to overcome the driving force of forming a 3D cluster, a 3D-to-2D growth transition occurs. The 3D growth is suppressed and the subsequent growth is in the lateral direction. This causes the Pb islands to have a flat top surface and to be of a multilayer thickness. The growth transition reflects interesting information that even a nanosize 3D cluster, its subsequent growth can be affected by the quantum size effect. This does not follow the general concept that the influence of the quantum size effect on the growth can only manifest itself the thin film structure as in the growth of Ag/GaAs(110). Since Fig. 2(b) is a statistic result, it cannot demonstrate directly the atomic path of how a 3D cluster is transformed into a 2D island, nor how the island thickness is correlated to the height of the 3D cluster. There is an interesting feature worthy of our attention, however. As seen in Fig. 2(b), the height of the 3D clusters can reach seven layers. This is exactly the magic thickness of the 2D islands. Unfortunately, this does not tell us how 3D clusters of different heights are transformed into 2D islands of different thicknesses. It is possible that such a transformation is also affected by the kinetics of atomic motion and by thermal fluctuations. We would like to point out here that, while at 208 K the critical thickness, or the least thickness of the 2D islands formed, is four layers, at 170 K this thickness reduces to three layers. This again points to the fact that a 3D to 2D transformation, which is driven by the electronic effect of quantum sizes, is also affected by the atom mobility and thermal effect.

Another question we ask is whether the growth of multilayer 2D islands can be described by a 2D theory of growth. To answer this question, we analyze the average island size (area) at different coverages. Figure 3(a) shows the average size as a function of thickness at coverages of 0.8 and 1.6 ML above the wetting layer at 208 K. The average size is insensitive to the thickness, indicating that the growth rate of the island size is nearly independent of the island thickness. Figure 3(b) demonstrates that the average size of each thickness becomes double when the coverage changes from 0.8 to 1.6 ML, implying islands of any thickness grow in size instead of in thickness, again showing the 2D growth

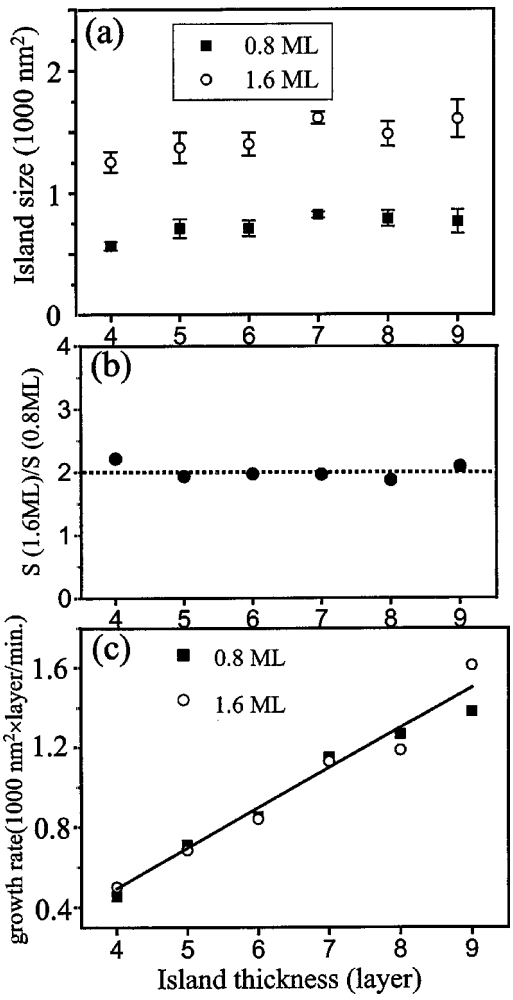

FIG. 3. (a) The average island size (area) as a function of island thickness at the coverage of 0.8 ML and 1.6 ML above wetting layer at 208 K. (b) The average size of each thickness doubles when the coverage changes from 0.8 ML to 1.6 ML, showing a 2D growth behavior. (c) The growth rate of island volume as a function of island thickness, showing a linear dependence.

behavior. Thus the growth rate of the island volume or its ability to absorb free atoms of the islands linearly increase with thickness, as shown in Fig. 3(c). This linear relationship is due to the fact that the vertical growth is suppressed after the 3D-to-2D growth transition. The probability that a mobile atom is adsorbed into the multistep edge of an island is linearly proportional to the island thickness. Since the maximum probability is 1, the linear dependence indicates that the absorbed probability should be smaller than 1, which means that the mobile atoms at the island edge have a chance to escape from islands for all kinds of thicknesses. Since the number of absorption site of the island edge is linearly proportional to the thickness, the absorbed probability, and thus the growth rate of the island volume, linearly increase with the island thickness. This results in the island size being insensitive to the thickness. Therefore the growth of these multilayer 2D islands should be similar to that of single-layer 2D islands.

The scaling theory has been used to derive the critical size

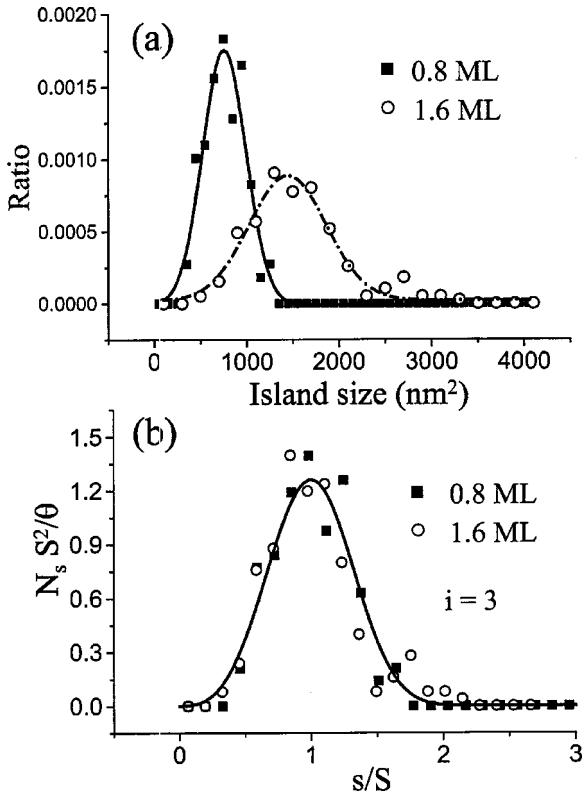

FIG. 4. (a) Pb island size distributions at the coverage of 0.8 and 1.6 ML above the wetting layer. The solid and dash-dotted lines are smooth fitting lines to the data points. (b) The two size distributions in (a) can be scaled into nearly identical distributions. The solid curve is the theoretical scaling function of three-atom critical size.

of single-layer islands in homoepitaxy systems.[9,10] According to this theory, there exits a universal scaling function $f_i(u)$ which depends only on the critical size i, and it can be related to the size distribution of the single-layer island according to

$$f_i(s/S) = N_s(\theta) S^2/\theta, \tag{1}$$

where $N_s(\theta)$ is the island-size distribution, corresponding to the number density of islands at size s at coverage θ, and S is the average island size. Theoretical scaling functions of different critical sizes are derived from the scaling theory. Using Eq. (1), one can obtain the experimental scaling data from the island-size distribution. By comparing the theoretical scaling function with the experimental scaling data, the critical size can then be derived.

We have described the growth process after nucleation; now we further study the critical size of the nucleus. Since 2D Pb islands behave similarly to single-layer islands, an interesting question is whether the scaling theory can be used to analyze these multilayer islands to find their critical size. We pose an argument. Let us assume that there is a size distribution of single-layer islands, and that it obeys the scaling law of Eq. (1). If each one-layer island is folded into a two-layer island, the size of each island and the average size as well as the coverage, which is the total area of all single-

layer islands, would be a half the original ones. The number density of islands would be doubled. Therefore, Eq. (1) for two-layer islands becomes

$$f_i(s/2)/(S/2) = 2N_s(\theta)(S/2)^2/(\theta/2); \quad (2)$$

after rearranging Eq. (2), it becomes

$$f_i(s/S) = N_s(\theta)S^2/\theta. \quad (3)$$

Equation (2) returns to the same scaling form, indicating that the scaling theory is still valid for understanding the critical size of multilayer 2D islands. However, the nucleation has to be independent of the vertical growth mechanism in the formation of a multilayer island. In some homoepitaxy experiments, the Arrehinus plot of the single-layer island density as a function of temperature always reveals a linear relationship,[9,10] and it also appears in the growth of multilayer Pb islands.[11] This implies that the nucleation of Pb islands should be similar to that of single-layer islands since the Pb islands are created on the Pb wetting layer. We thus apply the scaling theory to investigate the critical size of the Pb islands. As the island size is nearly independent of the island thickness, we analyze the size distribution of all islands without distinguishing their thickness. Figure 4(a) shows the size distribution at coverages of 0.8 and 1.6 ML. These two distributions can be scaled into two nearly identical distributions by the scaling equation as shown in Fig. 4(b), showing a behavior similar to those of single-layer islands. The solid curve is the theoretical scaling function of critical size of three atoms,[6] and it matches these two groups of the experimental scaling data well. Thus we conclude that the Pb tetramer is the smallest stable nucleus in the incipient growth stage.

In conclusion, based on the above analysis and observations, we propose a mechanism for the formation of multilayer 2D Pb islands. First, after the completion of the wetting layer, every four Pb atoms form a tetramer as a stable nucleus for the further growth. Following the nucleation, the SK growth mode provides the initial growth mechanism for forming 3D clusters. When the amount of aggregated atoms of 3D clusters is large enough to establish the quantum size effect to suppress the SK growth, the lateral growth governs the final stage of the formation of a 2D Pb island. The energetics is optimum for the quantum size effect when the height of 3D clusters is seven layers; thus the magic thickness is seven layers. The existence of other thicknesses is the result of atom kinetics and thermal fluctuations. In addition, our observations show that the island size is insensitive to the thickness; thus we can treat multilayer flat top Pb islands as single-layer islands, and derive their critical size from the scaling theory. We believe our analysis provides atomic scale insights for understanding the mechanism in low-temperature epitaxy of metals on semiconductor surfaces.

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