Electronic growth of Pb islands on Si(111) at low temperature

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(Received 19 December 2001; published 28 May 2002; publisher error corrected 3 June 2002)

The growth of Pb films on the Si(111)7×7 surface has been investigated at low temperatures using scanning tunneling microscopy. Flat-top Pb islands are formed and at low coverage the thickness of islands is confined in the range of four to nine atomic layers. Among these islands, those of seven-layer height are the most abundant. In low coverage limit, these multilayer islands prefer to grow in size instead of in thickness, showing a quasi-two-dimensional growth property. This growth behavior, different from the conventional growth modes, arises from the quantum size effect. At higher coverage, the growth also reveals layer-by-layer behavior. The Arrhenius plot of the island density versus temperature shows a linear relationship, indicating the formation of islands can be explained by the nucleation and growth theory. We also study the growth of Pb films on the incommensurate Pb/Si(111) surface at low temperatures. Flat Pb islands can be grown as well, but the threshold thickness is reduced to two atomic layers instead of four.

DOI: 10.1103/PhysRevB.65.245401 PACS number(s): 68.55.Ac, 68.55.Jk, 68.37.Ef

I. INTRODUCTION

Growth of thin metal films with atomic flatness on a semiconductor has been an important subject in recent years due to both scientific interest and possible applications in the integrated circuit industry. However, because of lattice mismatch, the stress existing at the metal/semiconductor interface may drive the morphology of the metal film to three-dimensional(3D) island formation[1–3] instead of a flat film. For this reason, the growth of a flat metal film on a semiconductor surface remains a formidable task. Recent discoveries have demonstrated that this barrier can be overcome by a new growth procedure: low-temperature deposition followed by annealing to room temperature. By using this two-step process, flat silver overlayers with preferred thickness of seven and two atomic layers have been created on GaAs(110) and Si(111) surfaces, respectively.[4,5] Recently Budde *et al.* have also observed that flat Pb islands with seven-step thickness can be grown on the Si(111)7×7 surface at 185 K by analyzing the spot profiles acquired from low-energy electron diffraction.[6] A specific characteristic of these growth phenomena, not observed in conventional growth, is that the preferred thickness is not changed with increasing coverage until the film has been completely grown. In order to elucidate such peculiar growth behavior, a model named ‘‘electronic growth’’ has been proposed to explain why some flat metal films can be grown on semiconductor surfaces under the low-temperature growth condition.[7] The essence of this model is based on the quantum size effect in the metal film and the inclusion of charge transfer occurring at the metal/semiconductor interface. The preferred thickness of the metal film is determined by these two factors.

Pb is known to be chemically inactive and it does not intermix with Si.[8] Therefore, the Pb/Si interface is sharp, which facilitates the study of the growth mechanism and electronic properties. At room temperature, different phases of Pb atoms on the Si(111) surfaces are formed with varying degrees of coverage up to 2 ML.[9–14] Some properties such as electronic structure and Schottky-barrier height may change with these complex surface phases as well.[9,15] At higher coverage, Pb is known to grow on Si(111) with the Stranski-Krastanov mode, i.e., to grow in 3D islands following the completion of a wetting layer. By increasing the coverage up to hundreds of ML, the islands start to connect to each other and finally a continuous film is formed.[9] In our previous studies,[16,17] we have investigated the growth of a Pb film on the Si(111)7×7 surface at 200 K with scanning tunneling microscopy(STM). Our results show that flat-top Pb islands of thickness confined in the range of four to nine atomic layers can be formed and the seven-layer island is the most abundant one. In addition, I-V measurements on individual islands reveal the quantum-well states, manifesting that the quantum size effect indeed exists in Pb islands. In this paper, we focus on the essential factors influencing the growth of islands such as the coverage, temperature, and interface. Our findings show that in low coverage limit the islands tend to grow in lateral direction. Thus the growth of the multilayer islands is of quasi-2D growth behavior, similar to the homoepitaxy of single-layer 2D islands.[18] At higher coverage, the islands start to coalesce, and the lateral growth as well as the on-top growth proceeds simultaneously. The measured island density shows an Arrhenius dependence on temperature, indicating that though the quantum size effect indeed takes part in the island formation, the growth of the islands is still governed by factors in the conventional nucleation process, such as the activation energy of surface diffusion, the binding energy of the critical size, the growth temperature, etc.[20] Flat islands can also be formed when Pb is deposited on the incommensurate Pb/Si(111) surface at 208 K, but the threshold thickness is reduced to two atomic layers. This observation demonstrates that the interface structure can be important for the electronic growth.

II. EXPERIMENT

The experiment was carried out in a UHV chamber (base pressure less than 1×10^{-10} mbar) equipped with a coldfinger type variable temperature STM and a well-collimated *e*-beam heating evaporator of high purity Pb. Samples were cut from boron dopped Si(111) wafers with the resistivity 0.1∼1 Ωcm and a miscut angle of less than 0.1°. The reconstructed 7×7 surface was obtained by annealing samples to 1200 °C and followed slow cooling to room temperature. Lead atoms were evaporated onto the predetermined sample at low temperatures with a rate of 0.16 ML per minute. The pressure was kept below 2×10^{-10} mbar during the deposition. To prepare the incommensurate $\sqrt{3}$ surface, 2 ML of Pb was first deposited onto the 7×7 surface at room temperature and followed by annealing at 480 °C for a few seconds.

III. GROWTH CHARACTERISTICS

Figure 1 shows some STM topographic images of Pb overlayers grown at ∼208 K with three different coverages. Before islands are formed, about 2 ML of Pb are consumed in wetting the Si(111)7×7 surface. The remaining Pb begins to grow into Pb islands with a flat-top surface above the wetting layer. Previous studies have shown that the orientation of the top surfaces of these islands is along the [111] direction.[19] Comparing these three images, the island density does not change significantly with the coverage. Figure 2(a) shows the statistical data of island density as a function of Pb coverage. It demonstrates that the island density is at a steady-state regime when the coverage is between 2.72 ML and 5.12 ML. The island density drops rapidly at higher coverages, indicating islands start to contact each other, i.e., island coalescence occurs. Since the island density is almost constant at lower coverages, the deposition of Pb is to increase the thickness of island and the island size. In order to understand the changing of island thickness at different coverages, we sampled more than two hundred islands to analyze their thickness for each coverage. The thickness of an island is measured from the wetting layer to the top surface of the island. Fig. 2(b) shows the ratio distribution as a function of island thickness at three coverages. It is interesting to note that the thickness of islands is confined within the range of four to nine atomic layers. Since four layers is the minimum thickness we have observed, it represents the critical or threshold thickness for the growth of Pb islands. Islands with seven-layer thickness have the highest abundance ratio, indicating that the seven-layer is the magic thickness for the growth of Pb islands. We analyze the average thickness of islands and it only increases ∼7% even though the coverage (above wetting layer) increases by a factor of three. Thus the growth of islands is mostly along the lateral direction. Figure 2(c) demonstrates that the average size of the islands increases with the coverage linearly, further illustrating the two-dimensional growth behavior of these islands. The Pb islands are of flat top, preferred thickness, and grown laterally at low coverage. These characteristics are very similar to those by the electronic growth, therefore the quantum size effect should play an important role in the formation of Pb islands.

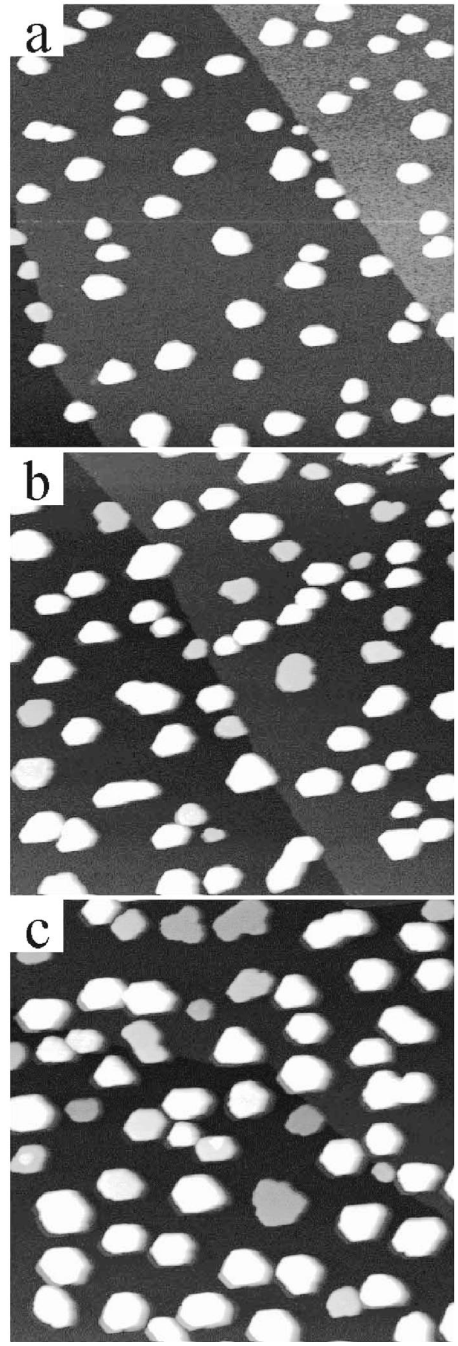

FIG. 1. STM images (500×500 nm^2) of Pb islands grown at ∼208 K with a coverage of (a) 2.72 ML, (b) 3.52 ML, and (c) 4.32 ML.

IV. GROWTH AT HIGHER COVERAGE

Figure 3(a) shows the morphology of Pb islands at a coverage of 6.72 ML. An obvious change is that the island density is reduced and the island size increases rapidly because of the coalescence of islands. Thickness measurement shows that islands with thickness above nine layers appear and seven-layer thickness is no longer the most abundant one, indicating the vertical growth has occurred. In Fig. 3(b), the island size at coverage of 8.32 ML is on average larger than the one in Fig. 3(a). Thus after the coalescence, islands can still grow in the lateral direction. Arrows in Fig. 3(b) point to the islands with an incompletely grown top layer. The appearance of incomplete islands indicates that the vertical growth is restricted by the supply of incoming atoms as the

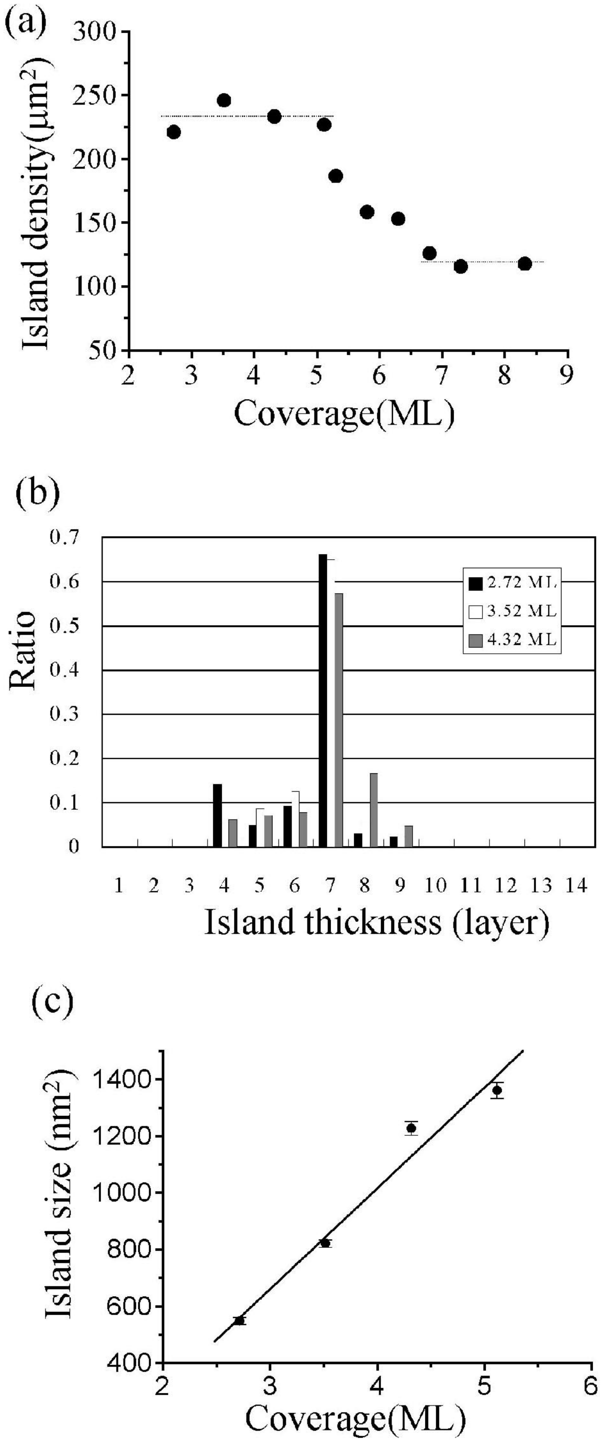

FIG. 2. (a) Island density as a function of coverage at $\sim$208 K; at low coverage (2.72–5.12 ML), the island density is saturated and it is reduced at higher coverage because of the coalescence. (b) The appearance ratios as a function of island thickness at three different coverages. (c) The average size of islands linearly increases with coverage at saturated regime, showing a quasi-two-dimensional growth behavior.

island size becomes large. In our observation, the thickness of the incomplete top layer is always one atomic layer, indicating that the vertical growth of islands may be the layer-by-layer growth. To check this point of view, we observe the growth of an individual island by *in situ* deposition. During deposition we withdraw the tip away from the surface only a few hundred angstroms in order to observe the same scanned area before and after deposition. Figure 4(a) shows two Pb islands with five-layer (bottom left) and eight-layer (up right) thickness grown at $\sim$205 K for the coverage of 2.77 ML, but observed at 160 K. At 160 K, the growth speed is suppressed so we can observe the growth process in steps. Figure 4(b) demonstrates that the five-layer island becomes larger and an incomplete single-step layer is created on the base island after additional deposition of 0.19 ML Pb. With 0.04 ML more deposition, basically the shape of the incomplete layer is not changed. However, the incomplete layer gradually changes to a complete layer with time even without further deposition, as shown in Figs. 4(c)–4(f). The evolution of the incomplete layer reveals that there are still free Pb atoms moving on the surface and they take time to meet the incomplete layer due to the lower mobility at 160 K. Figures 4(d)–4(f) thus illustrate the vertical growth of islands following the mechanism of layer-by-layer growth.

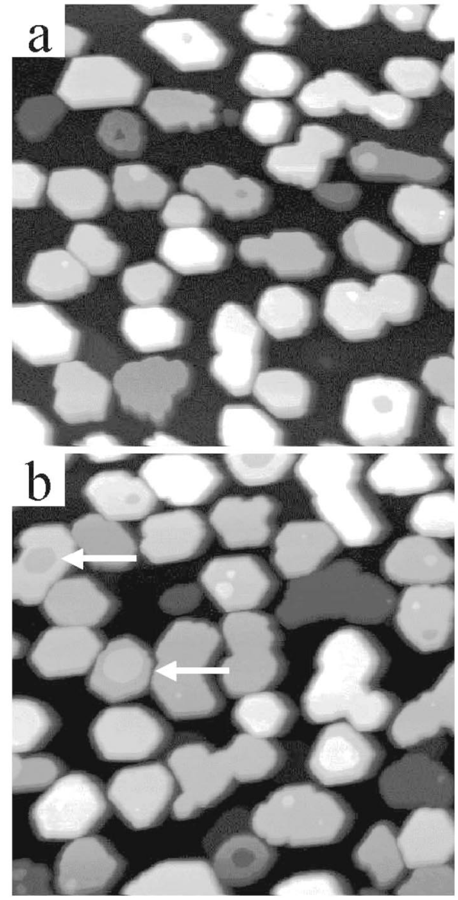

FIG. 3. (a) The growth of Pb islands at $\sim$208 K with a coverage of 6.72 ML; island density becomes less, compared to the one in Fig. 1(b) At the coverage of 8.32 ML, the island size increases and single-step incomplete layers (indicated by arrows) appear on some islands. The size of both images is 500×500 nm^2.

V. TEMPERATURE EFFECT

Figure 5 displays the growth of islands at temperatures from 190 K to 250 K at a coverage of 3.2 ML. In this tem-

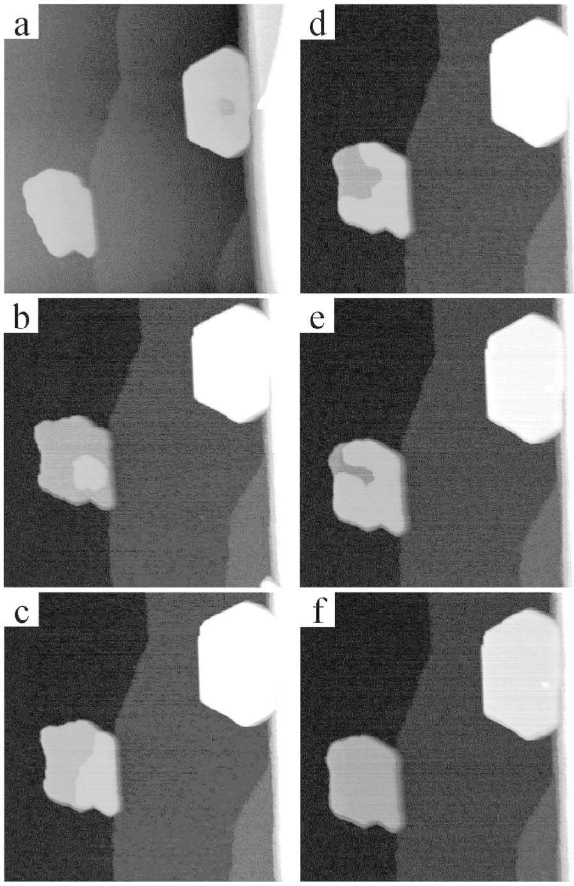

FIG. 4. (a) Islands grown at 208 K but observed at 161 K. (b) After more 0.19 ML *in situ* deposition at 161 K, the left-down island becomes larger and an incomplete single-step layer is created on its surface. After more 0.04 ML deposition, this layer changes into a complete layer with time as shown in (c)–(f). Each image size is 300×300 nm^2.

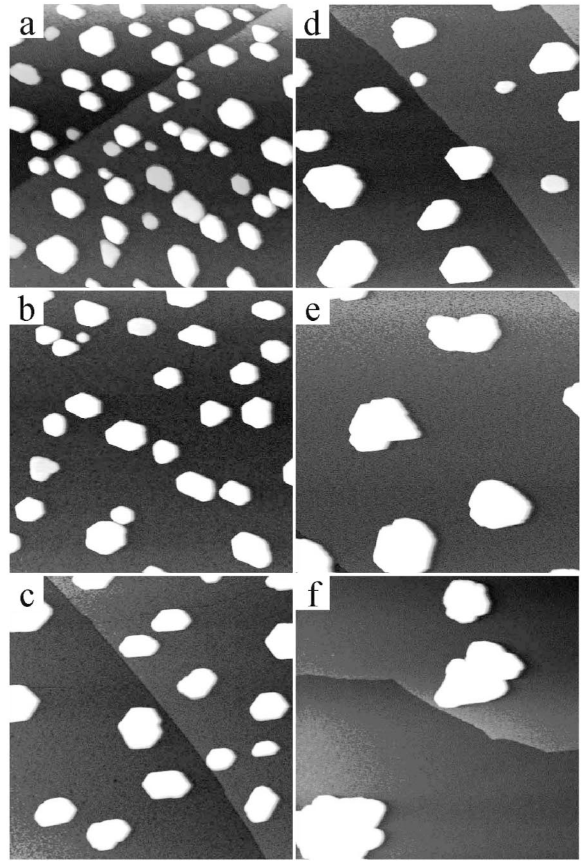

FIG. 5. The growth of Pb islands at (a) 189 K, (b) 198 K, (c) 208 K, (d) 217 K, (e) 226 K, and (f) 254 K at a coverage of 3.2 ML. The image size of (a)–(c) is 300×300 nm^2, (d) and (e) is 500×500 nm^2, (f) is 1000×1000 nm^2.

perature range, the top surfaces of the islands maintain their atomic flatness. The thickness measurement reveals that the height of Pb islands is still confined in the four to nine layers range. It indicates that the effect of quantum confinement sustains even when temperature reaches 250 K. As the temperature is raised further, the island density decreases, and the island size increases, manifesting a competing process between the island nucleation and growth. The competition arises because Pb adatoms diffuse faster at higher temperature, the probability for an adatom to attach to an existing island instead of nucleating a new island thus increases. According to the nucleation theory, in the low coverage limit with isotropic diffusion, the island density N is given by[20]

$$N \sim \exp[(iE_d+E_i)/(i+2)k_BT], \tag{1}$$

where E_d is the activation energy for diffusion, and E_i is the binding energy for the critical size i. The Arrhenius plot of the island density versus temperature shows a linear relationship as demonstrated in Fig. 6. It implies that Eq. (1) can be applied to describe the growth of Pb islands here. For some homoepitaxy systems in which single-step 2D islands are formed, the critical size of the nucleation of islands is one or two atoms, depending on the epitaxy temperatures.[21,22] Since the Pb islands, in our case, are formed on the wetting layer, the nucleation process is supposed to be similar to the homoepitaxy and the critical size should be close to that in the homoepitaxy system. The number of aggregated atoms for manifesting the quantum size effect should be much larger than the critical size. We believe that the nucleation process must occur before the quantum size effect takes place. Thus the nucleation and the quantum size effect are two independent factors in the formation of an island, the former results

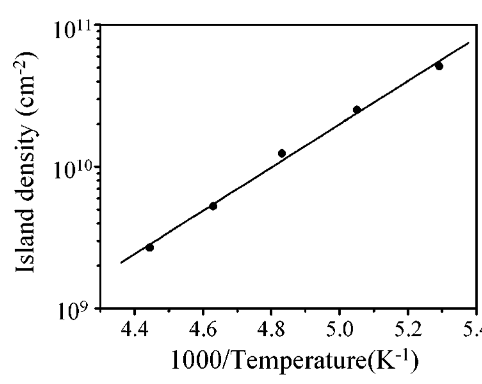

FIG. 6. The Arrhenius plot of the island density versus temperature, showing a linear relationship.

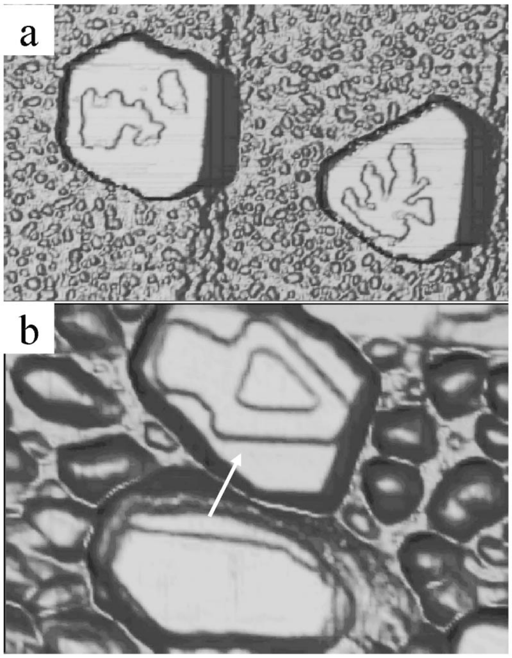

FIG. 7. Post deposition on the island-creation surface at (a) 56 K; the single-step and fractal-like layer is formed on islands. (b) 121 K; the compactlike layer is created on an island (indicated by an arrow). The image size of (a) and (b) is $138\times80\ \mathrm{nm}^2$ and $106\times72\ \mathrm{nm}^2$, and both are the 3D images.

in the creation of an island and the latter determines the thickness of the created island.

Figure 7(a) shows the 3D topographic image acquired at 56 K after first depositing 2.72 ML Pb at 208 K and subsequently depositing additional 0.8 ML at 56 K. This two-step deposition allows us to observe the growth on wetting layer as well as on islands at lower temperatures. On the wetting layer, clusters with a size much smaller than the island are formed due to the low mobility of atoms. In addition, the height of clusters is smaller than the critical thickness of islands, indicating the quantum size effect does not contribute to the growth at this temperature. Therefore, if the epitaxy temperature is too low, the deposited atoms are not mobile enough to exhibit the effect. On islands, single-step and fractal-like Pb layer is grown, showing the top face of the island is the (111) surface[23–25] and the diffusion of Pb atoms on the island is very different from that on the wetting layer. Because of the amorphous structure of the wetting layer, the arriving Pb atoms are supposed to face a larger barrier in diffusion, therefore the Pb atoms are formed into small clusters and the distance between each cluster, which reflects the diffusion length, is also short. On the other hand, atoms moving on the island possess a much larger mobility, the number of created single-step layers on the island is thus only one or two. However, at 52 K, the diffusion of atoms at the edge of the layer is sufficiently low, therefore the shape of the layer is fractal-like instead of compactlike.[25] Figure 7(b) shows that the compactlike layer is grown (indicated by an arrow) when the additional deposition is performed at 121 K. This indicates that Pb atoms can migrate along the layer edge, thus leading to a smoother shape at this temperature.

VI. INTERFACE EFFECT

It is well known that Si(111)7×7 surface can be converted into a 1×1 bulk-terminated structure if further annealing is performed after the deposition of Pb on Si(111)7×7 surface. Pb atoms on a bulk-terminated surface can give rise to several different phases depending on the coverage. For example, incommensurate (IC) phase, 1×1 phase mixed with $\sqrt{3}\times\sqrt{3}$ phase exist at a coverage just above and below 1 ML, respectively.[14] Previous studies have shown that the Schottky-barrier at Pb/Si(111)7×7 interface is different from that at Pb/Si(111)1×1 interface,[15] indicating that the charge transfer between Pb and Si for these two interfaces is different. Since the charge transfer at the metal/semiconductor interface is an important factor in the electronic growth of a film, we expect that the Pb growth on the IC phase would be different from that on the Si(111)7×7.

Figure 8(a) shows a typical STM image of the incommensurate Pb surface,[14] which consists of domains (bright region) and domain walls (dark region). In a domain, every three Pb atoms gather to form a trimer and the lattice of the trimers displays a periodicity of $\sqrt{3}\times\sqrt{3}$. Every two Pb atoms in the domain walls form a dimer. In contrast with the Pb growth on the Si(111)7×7, the islands can be grown on the IC phase directly without an additional wetting layer. Figure 8(b) demonstrates the growth of Pb islands on the

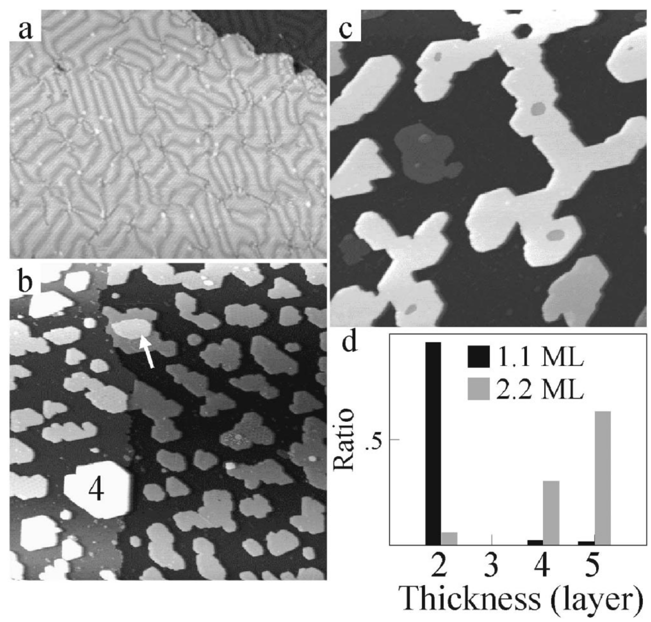

FIG. 8. (a) The typical topography image of the incommensurate Pb/Si(111) surface. (image size: $100\times78.5\ \mathrm{nm}^2$) (b) The growth of flat top Pb islands on the incommensurate Pb surface after depositing 1.1 ML. At this coverage, there are few four-layer islands (mark by number) and the three-layer island is never found but the third layer on some two-layer islands can be observed (indicated by an arrow). (c) The topography image at coverage of 2.2 ML, showing the island density is reduced and island size becomes large. (d) The ratio distribution as a function of island thickness, showing the preferred thickness is two layers at 1.1 ML and the four and five layers become the next preferred thickness at 2.2 ML. The image size of (b) and (c) is $300\times300\ \mathrm{nm}^2$.

incommensurate surface after depositing ~1.1 ML of Pb at 208 K. The ratio distribution of islands as a function of thickness is plotted in Fig. 8(d), which reveals that most of islands are of two atomic layers in thickness, which marks the critical thickness for the growth on this substrate. At this coverage, four-layer [indicated by number in Fig. 8(b)] and five-layer islands are seldom found, and they do not appear at lower coverages. In comparison with the growth on the substrate of Si(111)7×7 surface, which can have six different kinds of preferred thickness, the primary thickness of the grown islands on IC surface is just of one kind. It demonstrates that the interface indeed plays an important role in the electronic growth. Figure 8(c) shows that the island density is reduced and the island size becomes larger as the coverage becomes 2.2 ML. The ratio distribution of this coverage shown in Fig. 8(d) demonstrates that the dominant thickness is four and five layers, and two-layer thickness becomes a minor one. Thus four-layer and five-layer are the next preferred thickness for growing islands. Recent studies by spot profile analysis low-energy electron diffraction have shown that the five-step islands are the most stable islands.[26] This is consistent with our result at higher coverages. In our observation, the complete three-layer island is never found and the third layer [indicated by an arrow in Fig. 8(b)] can only be grown on two-layer islands of sufficient sizes. Therefore the three-layer island is unfavorable for the Pb growth on the IC surface. This is a further distinct behavior compared to the growth on the Si(111)7×7 surface, in which there is no "gap" in the window of preferred thickness.

VII. CONCLUSIONS

Using the scanning tunneling microscopy, we have investigated the growth of flat-top Pb islands on the Si(111)7×7 surface at low temperatures. When the coverage is less than 5 ML and the temperature is lower than 250 K, six kinds of preferred thickness, i.e., four to nine atomic layers above the wetting layer, appear among the grown islands, and islands of seven-layer height are the most abundant. These growth characteristics indicate the important role of the quantum size effect in the growth mechanism and the growth behavior of Pb islands exhibits a quasi-two-dimensional property. At higher coverage and substrate temperature, both factors will promote the growth out of the above range. We also studied the growth of Pb islands on the incommensurate Pb/Si(111) surface at 208 K to explore the interface effect. Quasi-2D flat-top islands with sharp edges were also observed, implying the electronic growth model can still apply. Below the coverage of 2.5 ML, the distribution of the island thickness has been changed to two to five layers.

ACKNOWLEDGMENTS

The authors would like to thank Dr. Ing-Shouh Hwang for valuable discussions. This work was supported by the National Science Council of Taiwan, Republic of China.

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