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Ultrafast kinetics in Pb/Si(111) from the collective spreading of the wetting layer

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Although quantum size effects in Pb/Si(111) control the selection of uniform island heights, the fast formation time (after deposition of few minutes at T ~ 180 K) has been puzzling so far. By studying the island growth in stepwise deposition experiments, we demonstrate the important role of the wetting layer in the kinetics. Pb is transferred to unstable islands, not by independent atom hops, but by the continuous spreading of the wetting layer from the surrounding area to the island top. As Pb is moved to the top in ring-like morphology the underlying 7×7 reconstruction in the surrounding area emerges gradually.

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I. INTRODUCTION

One of the unusual systems that has generated a lot of interest recently is Pb/Si(111). Numerous experiments by different techniques (SPA-LEED,[1] STM/STS,[2,3] x-ray,[4,5] ARPES,[6,7] *in situ* conductivity,[8] etc.) and corresponding theoretical calculations[9,10] have shown that the reason the system displays such extraordinary selectivity in island height is quantum size effects (QSE), i.e., the dependence of the quantized electronic energy on the island height. Theoretical calculations have confirmed the bilayer height stability with first principles and other approximate techniques which was attributed to the special relation between the Fermi wavelength λ_F and interlayer spacing d for Pb(111) $d\approx(3/4)\,\lambda_F$. The island stability was observed directly from oscillations of the position of the highest occupied band (HOB) with respect to the Fermi level, i.e., heights with HOB further away from the Fermi level correspond to stable heights, since the confined electron energy is lower. However, this oscillatory dependence of the energy level positions with respect to the Fermi level has been also seen in other systems[11–13] where QSE also are present, but it is still not clear why Pb/Si(111) is so special. In these systems the position of the HOB with respect to the Fermi level also oscillates with height, so it is puzzling why preferred heights have not been observed in these systems as well. This suggests that possibly there are additional reasons for the extraordinary height selectivity in Pb/Si(111). One possible candidate is kinetics, i.e., unusually fast mass transport mechanism that enables the system to find the QSE-driven energy minima within the duration of the experiment, a condition which possibly is unique to Pb/Si(111).

It is the goal of this work to report stepwise deposition experiments to understand how the Pb/Si(111) islands are built and to check whether kinetic processes are unique to Pb/Si(111). [Although Pb/Cu(111) has also been shown to have preferred heights,[14] the system has not been examined so far with as many techniques as Pb/Si(111) and the optimal temperature in Pb/Cu(111) is higher ~300 K.] It is clear that if these kinetic processes are better understood, they can lead to more educated search to discover other epitaxial systems where height selectivity will be as successful as for Pb/Si(111).

II. STEPWISE SECOND LAYER DEPOSITION EXPERIMENTS

The growth at ~180 K on different initial Si(111) interfaces [i.e., 7×7, Pb-β-$\sqrt{3}\times\sqrt{3}$, Pb α-$\sqrt{3}\times\sqrt{3}$, In-(4×1)] has led to mono-dispersive height distributions (with typical distribution widths less than 1% from the average). Stepwise deposition experiments are described as a function of temperature to observe how, starting from a nonoptimal initial height distribution (that includes stable and unstable heights), a sharper distribution with mostly stable heights is built after deposition of an additional amount $\Delta\theta$. From these experiments we have observed the crucial role of the wetting layer which not only is an additional source of Pb, but it diffuses in a novel collective mode from the region between the islands to the island top in a continuous, uninterrupted way. After reaching the top, it forms very quickly a ring around the unstable island perimeter with a width ~20 nm. The ring after its completion moves more slowly towards the center until the whole layer is fully occupied. It seems that, once this process is initiated, there is no barrier to be overcome and Pb proceeds as a large "spreading" drop from the wetting layer to the island top. This Pb transfer is seen from the comparison of the final versus initial Pb amount in the ring which shows the need of additional Pb to diffuse from the wetting layer to the islands (than the amount available from the coarsening of smaller Pb islands). As Pb is removed from the wetting layer, features of the underlying 7×7 reconstruction partially emerge. This is a local process where the transfer occurs close to unstable islands as revealed both by the growing ring on their top and the depletion of Pb in the wetting layer. This highly correlated uninterrupted motion of the wetting layer can account for the ultrafast kinetics and the extremely sharp height distributions observed so far only on Pb/Si(111). This might be the crucial process to be better understood if other systems will be found with the same behavior as Pb/Si(111).

An initial distribution of laterally big islands is first prepared at 240 K. The islands form after stepwise deposition with the first dose θ=1 ML followed by a smaller dose $\Delta\theta$ =0.5 ML to allow for more diffusion. Mostly 4-layer and 5-layer islands (and few 6-layer 7-layer islands form). The island height is measured from the wetting layer and 4-layer is an unstable and 5-layer is a stable height. The 4-layer

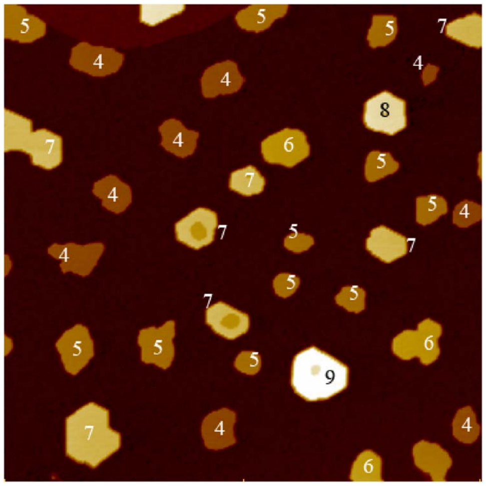

FIG. 1. (Color online) 1000×1000 nm^2 θ=1.5+0.25 ML grown stepwise at 240 K. The island heights are 4-, 5-, 6-, 7-, and 9-layers as followed by the color. Only few 6-layer islands have rings but no second layer islands on top at this high temperature. The 3-pronged island right-bottom is used as a marker that the same area is followed during coarsening.

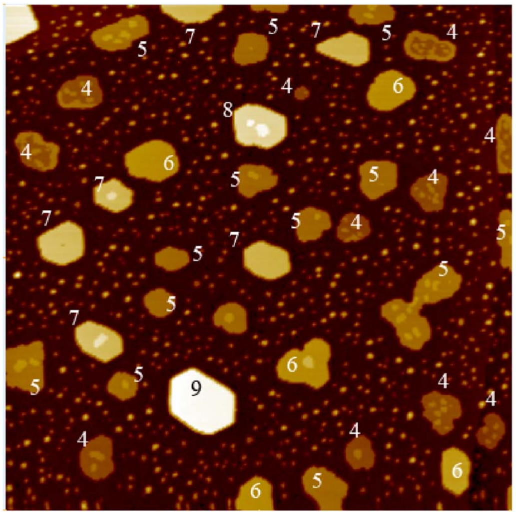

FIG. 2. (Color online) 1000×1000 nm^2 of the surface in Fig. 1 cooled at 180 K and, after an additional deposition, $\Delta\theta$=0.25 ML. Most 4-layer islands have both second layer islands and rings on top. Some stable islands (5- and 7-layer) have second layer islands on top but never rings. On top of the wetting layer many smaller (by two orders of magnitude in lateral size) islands have formed.

islands convert eventually to the 5-layer islands. For growth on the 7×7, 7-layer is the superstable height and the 5-layer islands will transform over longer times to 7-layer islands but because their lateral size is big (~100 nm) the time necessary to transform can be exceedingly long.

This initial wider height distribution is due to the growth at higher temperature (240 K) and at low flux rates (0.025 ML/min) that result in laterally big islands, which are unusually stable (when compared to islands grown at high flux rates). Also, most of the islands have irregular shapes and a three-prong island (see Fig. 1) at the right and lower half serves as a marker to follow the same area. The irregular island shape most likely indicates coalescence of neighboring smaller islands mediated by the moving wetting layer (as will be seen next), but such coalescence must be happening during deposition, so it has not been directly observed. Laterally big islands are necessary for the transfer to be initiated because the ring morphology requires a minimum initial island size larger than ~50 nm.

Figure 1 shows the result of an additional dose $\Delta\theta$ ~0.25 ML at 240 K on top of the starting distribution with island density 5×10^{-5} islands/nm^2. No new islands have formed, but more islands have hexagonal shapes. One island grows higher (9-layers). There are only two 6-layer islands with rings and the thickness of the ring is 1-layer (because 6-layer is an unstable height and the next stable height is 7-layer). The 7-layer islands do not show rings and this is true in all cases irrespectively of preparation. This is due to their superstability (among all odd heights) for growth on the Si(111)-7×7. No atoms "climb" from the wetting layer to the top of the 7-layer islands; and as deduced recently,[15] because of the zero step edge barrier at their perimeter atoms deposited directly on top descend quickly to the wetting layer.

After the surface was cooled to 180 K (over ~40 min), an additional deposition $\Delta\theta$~0.25 ML was made with the result shown in Fig. 2. The image size is 1000×1000 nm^2. Small islands are seen between the initial big islands and these smaller islands are mostly of 4- or 7-layer height, but differ in lateral size by ~two orders of magnitude from the islands grown at 240 K. The reason for cooling the surface down to 180 K is to slow down the transfer of Pb from the wetting layer so to study the ring front evolution with the STM acquisition speed.

The Pb amount transferred from the wetting layer to the islands depends on temperature. In other controlled experiments at 240 K for a similar initial island distribution of 4- and 5-layer islands, the coarsening outcome for one hour is mainly for the islands to grow to 7- and 9-layer height. This is seen in Fig. 3. The global amount transferred from the wetting layer to the islands ~0.32 ML.

However, at the lower temperature we can monitor other related processes, i.e., how the growth front of the ring moves on top of the islands (Figs. 4 and 5), how the wetting layer is depleted (Figs. 7 and 8) and the large growth front fluctuations (Fig. 9). At 240 K these processes are too fast and are completed within minutes (1–2 frames).

Visual inspection of the image shows differences in the second layer nucleation on top of the bigger islands for unstable (4-layer) versus stable (5-layer) islands. Most of the 4-layer islands have small islands at their center and a few have rings at their perimeter, but only a fraction of the 5-layer islands have small islands at their centers and none has rings. The small islands on top are the result of direct deposition. Since the majority of the 5-layer islands have no small islands on top, this indicates that at their perimeter the step edge barrier is low (and for the 7-layer islands is even lower as suggested in Ref. 16).

Comparing the images in Figs. 1 and 2, it is clear that more islands have rings (and small islands on top) at 180 K than at 240 K. This is due to the higher probability for the

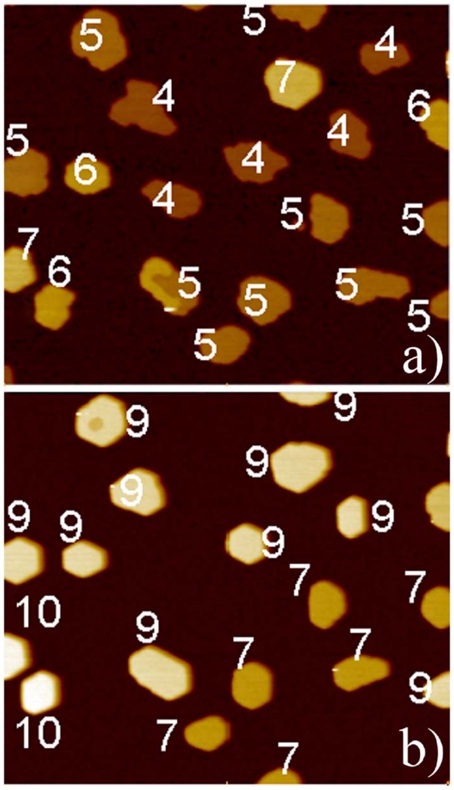

FIG. 3. (Color online) Two images showing coarsening at higher temperature 240 K. The scale is $414 \times 473\ \mathrm{nm}^2$ and the deposited amount is 1.5 ML. The time between the two images is 1 hour. Comparison of the global coverage in the two images shows a coverage increase of 0.32 ML, which confirms the larger transfer of Pb at 240 K than at 190 K.

atoms deposited directly on top to fall off at 240 K than at 180 K; while the atoms transferred from the wetting layer to the rings of unstable islands complete the next stable height very quickly. From the 180 K data it is possible to estimate the difference in step edge barriers for 4- versus 5-layer islands.[16]

After the second deposition at 180 K and the formation of the morphology shown in Fig. 2, there is no visible change in the islands for 30 min so the surface is heated to slightly higher temperature 190 K which initiates slow coarsening of the islands. Figure 4 is a $370 \times 400\ \mathrm{nm}^2$ zoomed area at the beginning of the heating and Fig. 5 shows the same area 1.5 h later (with the temperature kept at 190 K). The changes are visible with both the density of the smaller islands on the wetting layer decreasing and the rings growing on top of the 4-layer islands and in some cases engulfing some of the small islands on top.

The speed of the advancing front of the rings can be estimated from the average distance the front moves over the 1.5 h between the time to obtain Figs. 4 and 5. It is found to be $15\ \mathrm{nm}/5400\ \mathrm{sec} \sim 3 \times 10^{-3}\ \mathrm{nm/sec}$. However, this distance is not a measure of the diffusion coefficient since, as will be seen, the rate limiting step is the supply of Pb from the wetting layer.

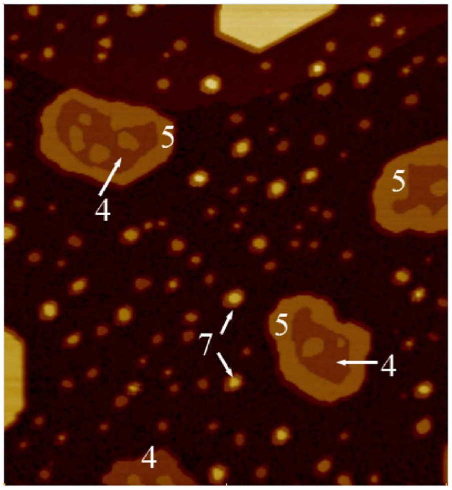

FIG. 4. (Color online) $370 \times 400\ \mathrm{nm}^2$ image $T=190$ K showing three 4-layer islands with smaller islands on top and with rings. The amount released from the deposition within the denuded zone is not sufficient to account for the growth of the rings on the big 4-layer islands.

III. GROWTH OF RING

The density of the smaller islands between the big islands decreases from 1.4×10^{-3} islands/nm^2 to 0.7×10^{-3} nm^2) within the time separating Figs. 4 and 5. A large fraction of the small 4-layer islands have decayed while on average the small 7-layer islands have doubled in lateral size. Most of the Pb released by the decay of the smaller 4-layer islands have moved to these growing 7-layer islands. It is not clear[17] if these superstable 7-layer islands eventually will decay and the Pb will move to the few bigger original islands (of Fig. 1); but if this happens, it will take much longer than the time in the current experiments.

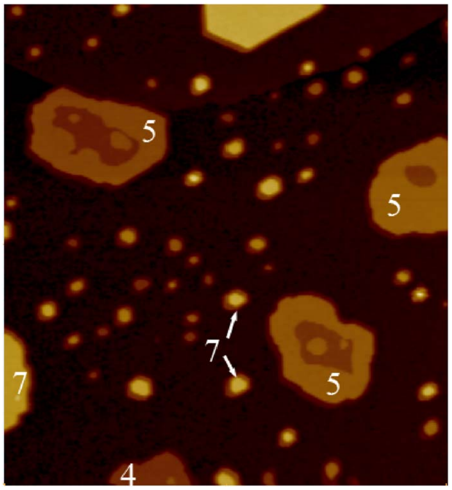

FIG. 5. (Color online) The same area as in Fig. 4, 92 minutes later with the surface still at $T=190$ K showing the growth of the rings which have engulfed some of the smaller islands. Most of the small 4-layer islands on the wetting layer have decayed, but the amount released is not sufficient to account for the growth of the rings.

The number of the smaller 4-layer islands has decreased by 23 islands within the (almost) parallelogram in Fig. 4 defined by the 4 bigger islands in the middle of the image. We can estimate the amount of Pb which has fed the growth of the rings on the bigger 4-layer islands to compare the two amounts, i.e., the added amount in the rings versus the Pb amount released by the decay of the smaller islands. The small decaying islands have an average radius of 5 nm and are 4-layer high, so each one has approximately 3000 atoms. All 23 islands have less than 7×10^4 atoms. As seen from the doubling of the lateral size of the 12 neighboring 7-layer islands with their average radius changing from 7.4 to 9.2 nm, the amount needed is 6.2×10^4 atoms. The amount remaining to move to the rings is 0.8×10^4 atoms, which is only $\sim\frac{1}{4}$ of the amount needed for the rings. This amount is estimated with high precision from the change of the area 3371 nm^2 covered by the rings in the three 4-layer islands (not the left bottom 7-layer islands which has only a small island at its center). The area is converted to the number of atoms 3×10^4 using the Pb(111) density 9.41 atoms/nm^2 (i.e., a simple visual estimate of the growth of the ring width is ~10 nm and the average size of the almost elliptical size hole is 80 nm × 50 nm). Since this amount is four times the amount released by the decaying small 4-layer islands, there must be another source of Pb atoms to account for the additional material. Despite these remarkable changes on the 4-layer islands, the stable 7-layer islands have no rings and practically no islands on top (with a very small number of exceptions).

This estimated number of atoms for the growth of the rings 3×10^4 atoms can be converted into 0.033 ML simply by dividing the total area of the wetting layer exposed in the image 103 000 nm^2 (in Figs. 4 and 5 assuming that it has only originated from the area on the same terrace with the three big 4-layer islands). This amount was checked also in a different way to be ~0.02 ML. Histograms were built for each height for the two images Figs. 4 and 5 (which include all the islands in the image) and the initial and final volumes are compared. It is found that the amount of Pb in the islands in Fig. 5 is 0.82 ML while in Fig. 4 it is 0.8 ML which is consistent with the net increase in the islands from Pb originating from the wetting layer. This apparent low value for the global coverage change is simply because the coverage is averaged over the whole image. However, the low global coverage 0.02 ML is not the best parameter to describe the Pb transfer from the wetting layer to the islands, but it is quoted to be compared to the much higher transfer amount 0.32 ML at 240 K. The transfer is highly localized and selective, i.e., it happens close to unstable 4-layer islands which are the only ones with rings. The quoted 0.02 ML is an average over "inactive" regions (close to stable 5-layer islands where no rings are seen and no transfer occurs). The coverage balance should be compared in the "active" regions, i.e., the growth of the three 4-layer unstable islands versus the decay of the small 4-layer islands minus the growth of the 7-layer islands between them. These three coverages are known accurately, i.e., the area on top of 3 large 4-layer islands is easily measured and the number of the small 4-layer islands and their lateral size distribution is known; as shown above the Pb amount needed for the growth of the rings is approximately four times the available amount from the decay of the small 4-layer islands.

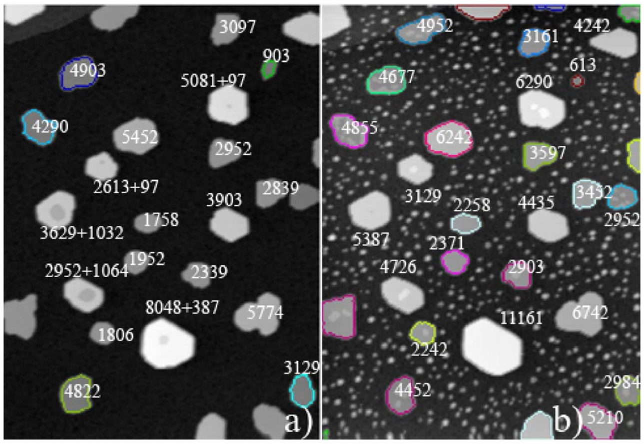

FIG. 6. (Color online) (a) The large islands grown at 240 K and cooled to 180 K with their lateral size denoted on top. The scale is 725×1000 nm^2. (b) The same area after deposition of 0.25 ML at 180 K. All islands increase in lateral size by 15–20% which shows that the amount transferred from the denuded zones is not sufficient to account for this lateral growth and the transfer from the wetting layer described in Figs. 4 and 5. (These are parts of Figs. 1 and 2.)

The transfer of Pb from the wetting layer to the rings can also be inferred directly from Fig. 2. Both stable and unstable islands show mostly lateral size increase which is larger than the deposited amount on top plus the amount collected within the denuded zones surrounding the islands. The lateral size increase is on the average 15–20%.

Stable height islands have no nucleation on top. Since the height of these islands is either 5- or 7-layer this means that the added amount for lateral growth is between 0.8 to 1 ML (measured with respect to their saturation coverage of the top layer). This is 1.5 to 2 times more than the amount $\Delta\theta$ ~0.25 ML fallen directly on top and 0.25 ML the amount in the denuded zones (since the area covered by the denuded zones is at most equal to the area covered by the island). Examining the unstable 4-layer islands with rings, the amount in the rings is more than 0.5 ML measured with respect to the saturation coverage on top (estimated by $2\,w/R$ where $R=80$ nm is the average radius of the big 4-layer islands and $w=20$ nm the initial average ring width).

The role of the denuded zones is better seen in Fig. 6 showing the surface at 180 K prior to the 0.25 ML deposition (that has generated Fig. 2). Each number denotes the top view of the outlined area; when a ring is present, then two numbers are shown, one the area covered by the ring and the other by the empty area in the island middle. The islands maintain their height and most of them grow laterally by ~15–20% as seen from the comparison of the numbers shown in the two images of Fig. 6. The entire 0.25 ML amount fallen within the denuded zone was used for the lateral growth of the island. From the lateral island growth the islands require more than 4 times 0.15 ML (or 0.6 ML) of Pb (this is the amount for the lower height 4-layer islands and the amount for the larger heights will be even larger). Furthermore, in Fig. 6 all heights (stable and unstable) grow laterally while in the images shown in Figs. 4 and 5 the growing rings are present only on unstable heights. This shows that the transfer in Figs. 4 and 5 is not only simple mass transfer as from the denuded zones but is driven by differences in island stability.

The needed amount 0.6 ML for the 4-layer islands between the two images of Fig. 6 (i.e., the difference between 1.1 ML the amount in the rings (0.5 ML) and the lateral growth (0.6 ML) minus the amount fallen on top (0.25 ML) and in the denuded zones (0.25 ML) can be converted into absolute coverage 0.085 ML by simply dividing it by the ratio (1/7) of the area covered by the three islands ($\sim$15 600 nm^2) over exposed area in the wetting layer (10 300 nm^2), since this is the area it has originated from. (This is higher than the previous global estimate between Figs. 4 and 5 because it includes the amount from the deposition. We use the different ways to evaluate the amount transfer from the wetting layer to show that the conclusions are very robust.) Figures 2 and 6 show the increase of the lateral size of the islands during deposition, the faster transfer from the wetting layer to the islands and that the total amount moved to the islands is 4–5 times larger than the amount transferred during annealing with the flux off as in Figs. 4 versus 5.

A similar conclusion is reached either from the growth in the rings at 190 K (difference between Figs. 4 and 5) or from the lateral growth and/or ring formation after the second deposition at 180 K (Figs. 2 and 6), i.e., the rings must have originate from the wetting layer. The amount is a strong function of temperature and also of the local environment of the islands and at 240 K is as high as 0.32 ML. The transfer depends on the local environment because it occurs mostly close to unstable 4-layer islands. In regions with predominantly stable heights there is no transfer.

IV. WETTING LAYER TRANSFER

It is interesting that, in most cases, the initial ring closes along the perimeter much faster ($\sim$over minutes) at 190 K while the completion of the open hole at the island center happens much slower ($\sim$over tens of minutes). This has been also observed for rings generated under different conditions, i.e., room temperature and with the application of a high voltage pulse.[18,19] The fast completion of the initial ring is seen from the continuous flow along the perimeter so there must be uninterrupted supply of Pb. It is difficult to imagine that if this Pb was coming from the uncorrelated decay of separate small 4-layer islands, then either these islands have decayed at the same time or that the released Pb very quickly has aggregated into a "drop," which provided the Pb to diffuse around the island perimeter so quickly. It is also not clear what defines the lateral size of the ring, why it remains at $\sim$20 nm for so long, and then why it propagates radially towards the center until much longer time?

Changes are also seen in the wetting layer after Pb is transferred to the islands. The wetting layer does not look featureless but clear structure develops and some order becomes visible. This order seen in Fig. 7 evolves from the partial recovery of the 7×7 reconstruction which is still intact on the surface with the 7×7 cell containing a mixture of Pb and Si adatoms.[20] As the Pb atoms escape the 7×7 unit cell and move towards the big islands[21] the reconstruction becomes uncovered, and lines parallel to the Si 7×7 unit cell start to be seen again.

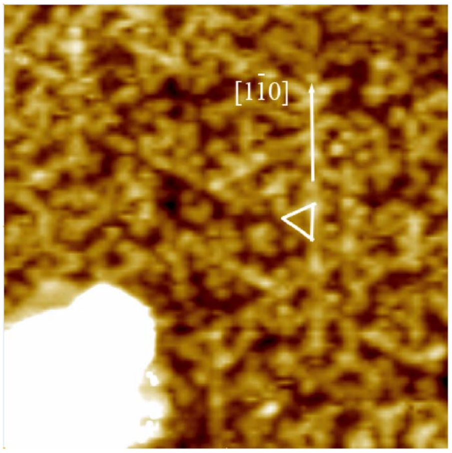

FIG. 7. (Color online) 30×30 nm^2 area on the wetting layer next to a small 7-layer island showing that partially order develops as Pb in the wetting layer is transferred to the rings. The arrow indicates the [1$\bar{1}$0] direction and the marked triangle is half the size of the 7×7 unit cell.

The emerging features of the 7×7 in the wetting layer are only observed in cases when the islands are laterally sufficiently big (more than $\sim$50 nm) so they can support the formation of the rings. This is possible for growth at low flux rates and high temperatures as in Fig. 1. For islands grown at high flux rates when the island are smaller, the wetting layer is featureless. Similar featureless wetting layer is also seen in the original data of Ref. 2 when the islands are small without rings. These results are summarized in the sequence of Figs. 8(a)–8(f). The wetting layer is shown close to a large $\sim$100 nm island where rings develop (top left). This island was grown at low flux rate 0.05 ML/min. The middle left image of Fig. 8(b) shows the wetting layer with the 7×7 features developing as in Fig. 7. This is further confirmed from the bottom left of Fig. 8(c) that shows the autocorrelation function of the previous image. The autocorrelation shows better the emerging 7×7 periodicity. The three images to the right show growth at high flux rates 0.5 ML/min when the islands are smaller with average size less than 20 nm [Fig. 8(d)], the wetting layer is featureless [Fig. 8(e)] and the autocorrelation function does not show the 7×7 periodicity [Fig. 8(f)]. Clearly this confirms the correlation between Pb transfer from the wetting layer, the ring morphology on top of unstable heights, and the revealed 7×7 features on the wetting layer.

Figure 9 shows a close-up view of the growing ring at the top left 4-layer island. The scale is 40×40 nm^2 and the ring front shows curvature and fluctuations at its perimeter. Also, fluctuations are observed at the perimeter of the small island on top of the 4-layer island, which was present prior to the spreading of the ring. The ring advances continuously as a single layer, which extends from the top of the ring, over the facet planes at the sides of the islands to the surrounding

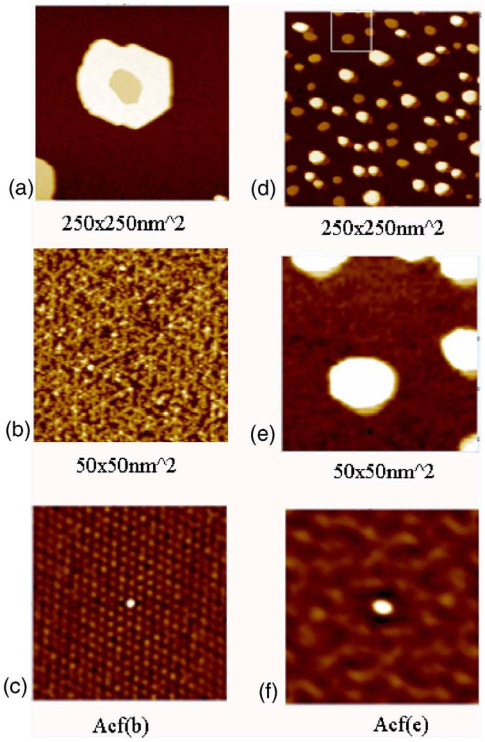

FIG. 8. (Color online) (a)–(c) Comparison of the emerging 7×7 features in the wetting layer for large islands. (a) A $250\times250\ \mathrm{nm}^2$ area showing a 4-layer island with a ring on top. The island was grown at a low flux rate 0.05 ML/min. (b) $50\times50\ \mathrm{nm}^2$ of the wetting layer showing the emerging features of the 7×7. (c) Autocorrelation function of image (b) showing the 7×7 periodicity; on the other hand (d)–(f) show no wetting layer transfer when the islands are smaller. (d) A $250\times250\ \mathrm{nm}^2$ area with smaller islands ~20 nm grown at higher (0.5 ML/min) flux rate. (e) No features of the 7×7 reconstruction are seen in the wetting layer. (f) This is confirmed from the autocorrelation function Acf of (e) which shows no periodicity.

wetting layer close to the growing island. If the atoms were climbing on top of the ring and were falling down randomly to advance the ring, then the ring front should be rougher.

These observations provide more information about the kinetic barriers in the system. The absence of second layer islands at 240 K versus the finite island density at 180 K can be used to deduce the surface diffusion of the Pb(111) islands and the wetting layer. These estimates are very consistent with previous independent estimates of the diffusion barriers.[15,22,23] In addition the difference in the occupation of 4-layer versus 5-layer islands after deposition at 180 K indicates a difference in the step edge barriers for stable versus unstable islands as conjectured elsewhere.[15] The consistency of these conclusions with the results of other work[15] supports the rest of the observations reported here about the wetting layer mobility.

An example of the diffusion barrier estimate is given below. The average size of the big island is 100 nm grown at 240 K. The islands have no second layer islands on top (Fig. 2), while they do after depositing $\Delta\theta\sim0.25$ ML at 180 K (Fig. 4) with average distance between the small second layer islands on top ~20 nm. If we take these distances to be the diffusion lengths at the two temperatures 240 K (i.e., 100 nm) versus 180 K (i.e., 20 nm), their ratio is 5 and if we use the relation that the diffusion length depends only on the ratio $1=(D/F)^n$ (Ref. 24) then we obtain the terrace diffusion barrier

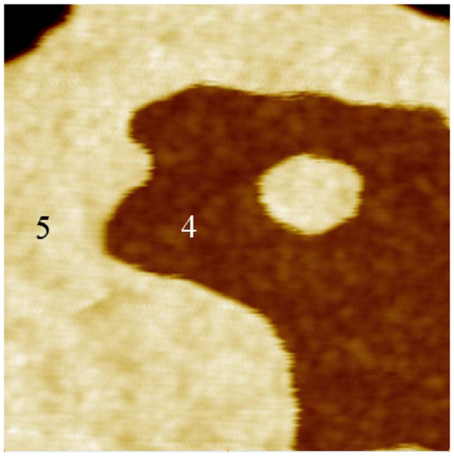

FIG. 9. (Color online) $40\times40\ \mathrm{nm}^2$ showing the spreading growth front of the ring on top of the right 4-layer island of Fig. 5. The curved front of the ring slowly approaches the small island and clear fluctuations are visible at the front and the small island perimeter.

$$\Delta E = \ln 5^{*}\frac{1}{\frac{1}{240}-\frac{1}{180}} = 0.06\ \mathrm{eV}$$

assuming $n=1$ since the critical size cluster is $i_c \gg 1$ ($n=i_c/(i_c+2)$ (Ref. 25). Otherwise, if $i_c=1$, the exponent should be $\frac{1}{6}$ and make the diffusion barrier higher. The assumption $i_c \gg 1$ seems reasonable, since it should take a large number of Pb atoms to aggregate and form the very first Pb islands as expected from the weak QSE energies controlling island stability (several tens of meVs). (There is no need to know the absolute value of the prefactor since only the ratio of diffusion lengths is used.)

This lower diffusion barrier on top of the islands versus on top of the wetting layer is also seen directly from Fig. 2, since the island density is higher by a factor of two which implies that the diffusion barrier on top of the wetting layer is 0.1 eV. (The island density on top of the wetting layer is 1.4×10^{-3} islands/nm^2.)

In summary, we have presented stepwise deposition experiments for Pb growth on Si(111)-7×7 showing the transition from an initial unstable to the next stable height islands after the deposition of a small amount, $\Delta\theta\sim0.25$ ML. It is found that the growth is based on the formation of rings which nucleate at the island perimeter of unstable 4-layer islands. These islands have rings of single layer growing to the next stable 5-layer height. A large fraction of the Pb amount needed for building the rings originates from the wetting layer, as seen from the imbalance between the amount in the rings versus the amount released by the decay of the smaller islands. This provides an easy source of Pb needed to build up the islands to the next stable height and explains why the kinetics is faster than what expected from classical coarsening processes. It is even more extraordinary the way the wetting layer moves: as a continuous, uninterrupted film (from far away the islands climbing the island facets quickly). It spreads within minutes along the island perimeter but the refilling of the remaining vacant hole in the island center is much slower. This novel type of diffusion where the layer moves in a continuous way without uncorrelated atom hops also explains why these islands are completed in such a short time, i.e., after deposition of several minutes. This reveals unexpected collective pathways during epitaxy that not only operate at low temperatures, but they decrease the diffusion times for mass transport and, therefore, the completion time of nanostructures by orders of magnitude, than what expected from classical kinetic processes.

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