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SCANNING TUNNELING MICROSCOPY STUDY OF EPITAXIAL GROWTH OF Si AND Ge ON SILICON

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Abstract. We use a scanning tunneling microscope (STM) capable of imaging the growing layer during MBE-growth at high temperatures. This method (MBSTM) opens the possibility to follow MBE growth processes dynamically on the atomic scale and gives access to the evolution of specific features during growth. The influence of surface reconstructions on growth kinetics can be studied directly. For the case of growth of Si islands on Si(111) we find lateral growth of rows of the width of the 7X7 reconstruction unit cell at the edges of twodimensional islands. This leads to a kinetic stabilization of magic island sizes. The evolution of size and shape of individual {105} faceted Ge islands (hut cluster) on Si(001) is measured during growth. A slower growth rate is observed when an island grows to larger sizes. This behavior can be explained by kinetically self-limiting growth.

1. INTRODUCTION

Recently scanning tunneling microscopy (STM) has become a powerful method to study epitaxial growth on the atomic level. However, STM has been limited in the past to “snapshots” of certain growth stages. The growth was interrupted at a specific coverage and the sample was quenched to room temperature and transferred to the STM for imaging. The main disadvantage of this mode of STM operation is that local growth structures which are accessible by a real space microscopy cannot be studied in their dynamic evolution during growth. Recently some effort was made to overcome these disadvantages [1-3]

Here we describe a mode of STM operation which opens the possibility to observe the dynamics during epitaxial growth “in vivo” (MBSTM). The experiments were performed in a standard ultra-high vacuum system. Due to the open design of the STM the molecular beam from an evaporator can be directed towards the sample while the STM is scanning the surface [4,5]. During imaging the sample is held at MBE growth temperatures of 600-900K.

Using this method of simultaneous scanning and molecular beam epitaxy the area behind the tip is shaded. This area can be measured if we evaporate Germanium at room temperature (small diffusion length) while the tip is fixed (i.e. not scanning). Scanning this area afterwards shows the silhouette of the tip on the surface (Fig. 1). The tip diameter can be measured to about 400 Å so that for images of several thousand Å scan range the shaded area is sufficiently small.

2. INFLUENCE OF THE (7 X 7) RECONSTRUCTION IN Si/Si(111) EPITAXY

During the epitaxial growth of 2D Si islands on Si(111) the growth of a selected island can is observed as a function of time. Fig. 2 (a-f) shows STM images from a growth sequence of such an island. In (a), the shape of the island is triangular. Images (b-d) show the same island at a later stage during growth. As shown by images (b-d), growth proceeds by advancement of a row of a certain width along the right island edge. The position of the kink at which the row is ending is shown by an arrow in (b-d). An analysis of the width of this row and further atomically resolved images show that the width of such a row is

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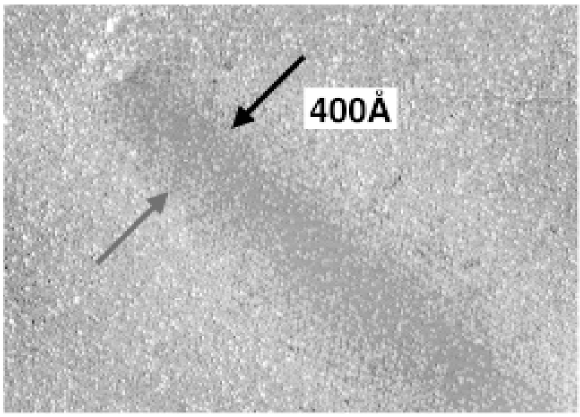

Fig. 1. **G**ray-scale image of the silhouette of the STM tip on the surface produced by the molecular beam. The Ge covered region is shown brighter while the darker region was shaded by the STM tip. The tip diameter can be measured to be 400 Å.

27 Å, which is just the width of one 7X 7 reconstruction unit cell [6].

In the following we discuss the influence of the 7X7 reconstruction on the growth behavior. The rhombic unit cell of this reconstruction consists of two triangles. One of this triangles has a stacking fault in the surface layers, relative to the substrate stacking (F-HUC). The other triangle of the reconstruction unit cell is unfaulted relative to the substrate (U-HUC), Fig. 3a.

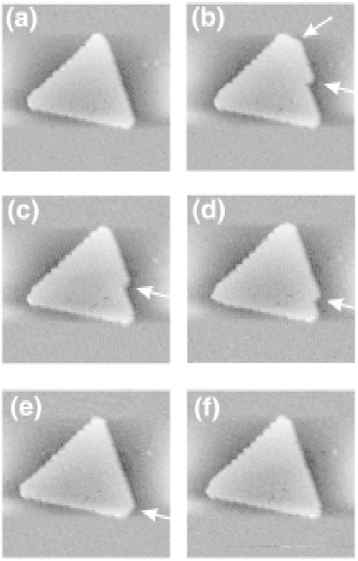

Fig. 2. Sequence of images showing the lateral growth of a triangular Si(111) island. A row of the width of the 7x7 unit cell is growing along the right edge of the island (b-f). The image size is 500x500 Å^2, T=575K. The complete growth sequence is available as a movie on the World Wide Web: http://www.fz-juelich.de/video/voigtlaender.

During lateral growth of an island, the surface reconstruction of the substrate has to be lifted and the substrate atoms have to rearrange to the bulk structure. This transformation of the reconstructed surface layer towards the bulk structure is a general phenomenon which has to occur in any epitaxial growth at reconstructed surfaces. It requires a higher energy to lift the reconstruction of the F-triangle as compared to the U-triangle [7]. To lift the reconstruction in the U-triangle, only atoms in the

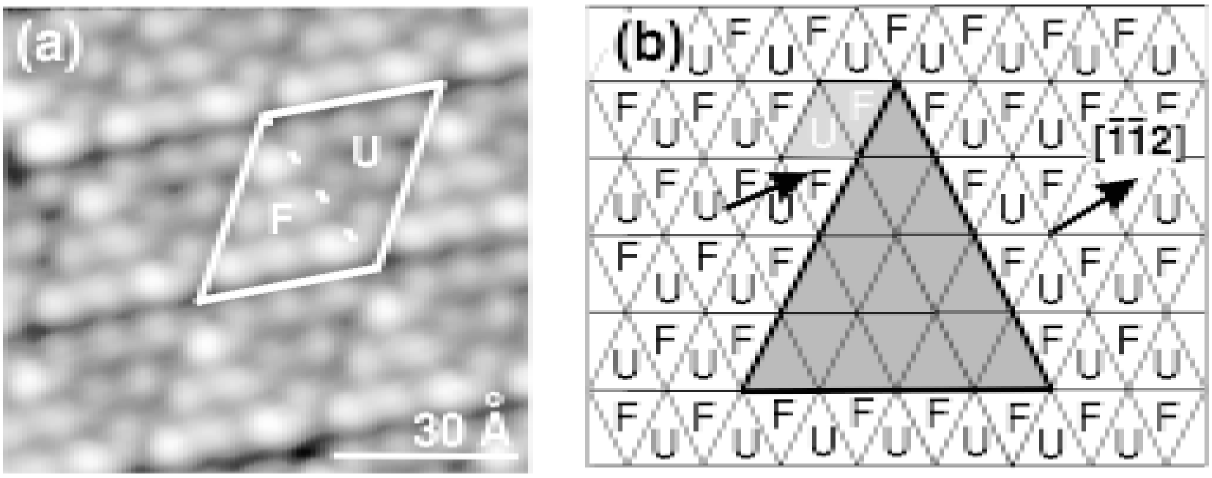

Fig. 3. (a) STM image of the Si(111) – 7x7 reconstruction. The white protrusions are the Si adatoms. The unit cell is indicated by a white rhombus. A dashed line divides two triangular subunits. Since the image is taken at negative sample bias (-2V), the adatoms in the faulted half of the unit cell (F) appear brighter than those in the unfaulted half (U). (b) Arrangement of the U and F parts of the 7x7 unit cell on the substrate around a triangular island (shown in gray).

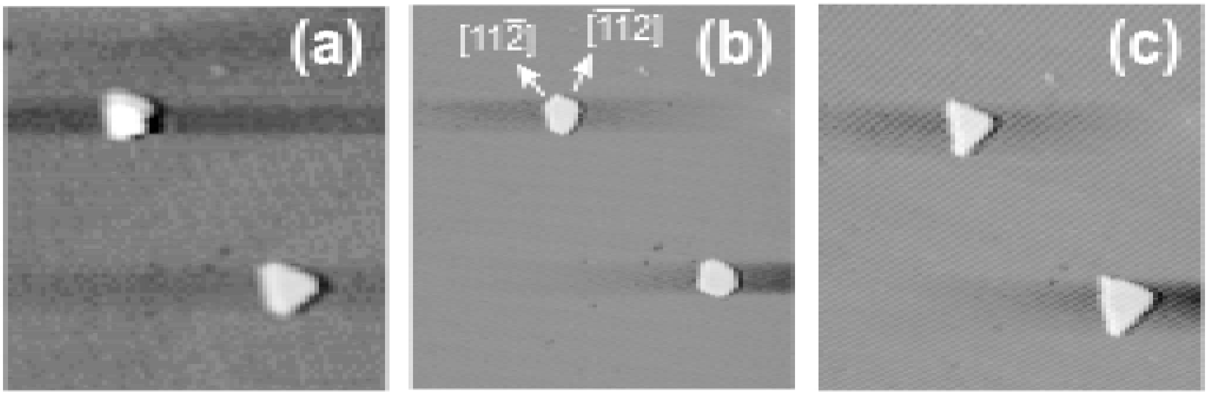

Fig. 4. Evolution of island shapes during growth and growth interruption (at a substrate temperature of 725K). Imaged during growth, islands have a triangular form. (a) After 18 minutes of annealing at 775K, the islands have changed their shape to hexagonal (rounded); (b) After growth commenced again, the island shapes changed back to triangular; (c) In all three images, snapshots of the same two islands are shown. The image size is 550 Å x 550 Å.

uppermost adatom layer have to rearrange. This is associated with a relatively low energy barrier. Lifting the reconstruction of the F-triangle requires the removal of the stacking fault in the layer below the adatoms. This rearrangement of atoms in deeper layers is associated with a larger energy barrier. This should lead to a high activation barrier for overgrowth of the F-triangle compared to overgrowth of the U-triangle.

In Fig. 3b, a Si island (gray) and the U and F triangles of HUCs of the surrounding reconstructed substrate surface are shown. Due to the crystallographic orientation of the island, it is surrounded only by substrate F-HUCs. This means that further lateral growth (initiated by the overgrowth of an F-triangle) is hindered by a high energy barrier. Once an F-triangle has nucleated, the neighboring U-triangles can be overgrown more easily (no stacking fault has to be removed). The overgrowth of the next F triangle is facilitated by the existence of a “macro kink” (arrow in Fig. 3b). Here the cost of the stacking fault energy is reduced by a gain in the island edge energy: The edge length is reduced after growth of an F triangle. Therefore, neighboring U and F units can be overgrown in quick succession, leading to the fast growth of a stripe of the width of the 7X7 unit cell.

Due to the quick growth of a row once it is nucleated and a longer time until the nucleation of the next row the island shape of a completed triangle is found often in snapshot images of the island morphology. Islands of complete triangular shape are kinetically stabilized. This higher stability for the closed shell triangular islands leads to pronounced peaks of islands of magic sizes in the measured island size distributions [7].

The two-dimensional triangular islands form during submonolayer growth of Si on Si(111) are 1 BL high. In the following it will be shown that this is a *kinetically* limited growth shape evolving during growth. Equilibration of these islands without external flux results in a transition to the hexagonal equilibrium form.

In Fig. 4 two islands are imaged first during growth (Fig. 4a), then after a growth interruption of 18 min (Fig. 4b), and finally after the growth was resumed (Fig. 4c). These experiments show that when growth is interrupted, the islands quickly lose their triangular shapes. First, the atoms from the apex regions detach creating islands with rounded corners. Subsequently, the island shapes more slowly turn into a hexagon-like (compact) shape as shown in Fig. 4b. When growth is continued, the islands quickly resume triangular shapes again. This evolution of growth morphology from triangular islands during evaporation to hexagonal islands upon equilibration of the surface at 725K and the final transition back to triangular islands when the external flux is resumed and the equilibrium conditions are no longer maintained is clearly shown in Fig. 4. This process shows that the triangular island shapes are non-equilibrium ones while equilibrium shapes are close to hexagons.

The equilibrium shape of an island is the form for which the energy of the island is minimized. If the steps in the $[\bar{1}\bar{1}2]$ and the $[11\bar{2}]$ direction were equivalent, the equilibrium shape would be an equilateral hexagon. During annealing the form of the islands approaches such a hexagon. However several observations after long times of annealing show that the edges perpendicular to $[11\bar{2}]$ are a bit shorter than the edges perpendicular to the [$\bar{1}\bar{1}2$] direction. This indicates that the edges perpendicular to [$11\bar{2}$] have a higher energy.

3.KINETICALLY SELF-LIMITING GROWTH OF Ge ISLANDS ON Si(001)

In Ge epitaxy on Si(001) three-dimensional Ge islands with {105} facets nucleate after the completion of the 2D wetting layer [8]. The island edges are oriented along <100> directions, i.e. at 45° to the direction of the reconstruction dimer rows. The strain of these islands is partially relaxed elastically, but they are coherent with the substrate lattice (i.e., free of dislocations between the substrate and the hut clusters) [9]. The evolution of the growth of these hut clusters is observed by STM during growth. A sequence of STM images for increasing coverage is shown in Fig. 5. From the images for instance the volume of the hut clusters can be calculated.

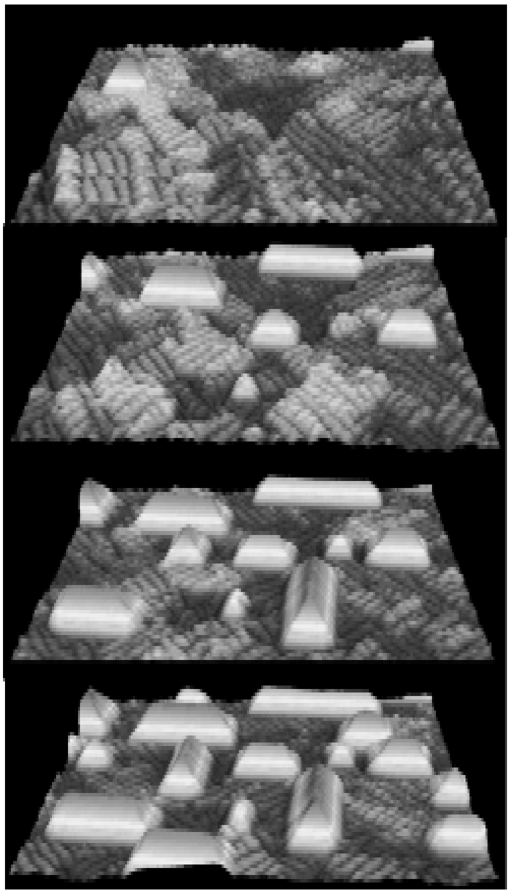

Fig.5. Perspective view of STM sample images of the hut cluster growth as function of coverage beyond the wetting layer. (0.07 ML, 0.46 ML, 1.33 ML and 1.81 ML from the top to the bottom; Image area: 1300 x 1000 Å^2, T=575K. The complete growth sequence is available as a movie on the World Wide Web: http://www.fz-juelich.de/video/voigtlaender/.

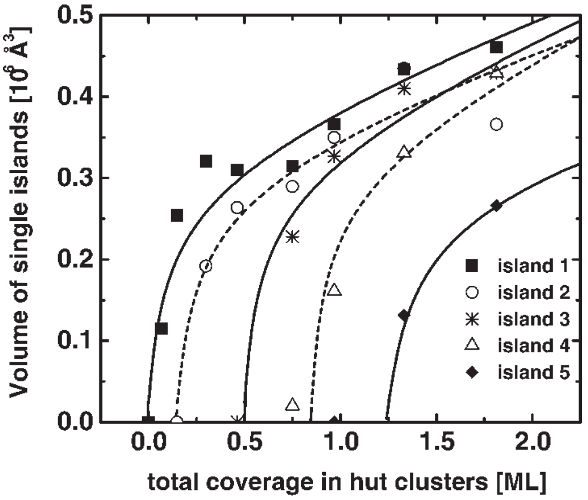

Fig. 6. Evolution of the volume of individual hut clusters. The different symbols correspond to different individual islands. The size evolution shows self-limiting behavior: The initially larger growth rate (large slope) just after the nucleation decreases when the islands grow larger. Results of a model calculation of kinetically self-limiting growth including a kinetic energy barrier for the nucleation of new material on the facets are shown as solid and dashed lines.

Fig. 6 shows the evolution of the volume of several individual islands as a function of total deposited coverage. The initially higher growth rate of individual islands just after the nucleation, indicated by initially large slopes in Fig. 6, decreases when the islands grow to a larger size. In the following, we use a model which shows that kinetically self-limiting growth explains the observed slower growth for larger island sizes. This kinetic model for the growth of hut clusters relies on a barrier for the nucleation of each successive atomic layer on the {105} facets [10]. The barrier for the nucleation of a new facet increases with the facet size. This size dependent nucleation barrier for the repeated overgrowth of the {105} facets is the reason for the self-limiting growth behavior [11]. Results of the model calculations for the evolution of the hut cluster volume are shown as solid and dashed lines in Fig. 6 and are in good agreement with the experimental data. The experimentally observed slower growth rate for larger islands is clearly reproduced. This indicates, that a kinetic self-limitation is effective during the growth of larger hut clusters. This kinetic self-limitation arises due to an energy barrier for the nucleation of new material on completely filled {105} facets.

4. SUMMARY

In summary we have shown that STM can be operated in real time during molecular beam epitaxy at high temperatures. MBSTM opens the possibility to observe the growth history of single growth features in real time. In the case of the homoepitaxial growth of Si on Si(111) the influence of the surface reconstruction on the lateral growth of Si islands was demonstrated. For the case of heteroepitaxial growth of Ge on Si(001) a slower growth rate is observed when an island grows to larger sizes. This behavior can be explained by self-limiting growth. A kinetic growth model involving a nucleation barrier for each repeated growth of a new atomic layer on the {105} facets agrees with the experimental results for the evolution of the island volume.

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