Magic Islands in Si/Si(111) Homoepitaxy

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The island size distribution after submonolayer deposition of Si on Si(111) exhibits pronounced peaks of *magic* sizes. Scanning tunneling microscopy studies during growth enable us to study directly the influence of surface reconstructions on growth kinetics. Lateral growth of rows of the width of the 7×7 reconstruction unit cell leads to kinetic stabilization of magic islands. Monte Carlo calculations are performed that reproduce the main experimental results and make it possible to estimate important energy barriers. [S0031-9007(98)06691-5]

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The phenomenon of surface reconstruction—the rearrangement of atoms at the surface due to the termination of the bulk structure at the solid-vacuum interface—has been studied in detail by scanning tunneling microscopy (STM). However, the effect of surface reconstructions on kinetics of epitaxial growth has received more attention only recently [1,2].

The simplest case to be studied is homoepitaxial growth. Most theoretical studies focus on a model in which atoms are deposited on sites of a regular lattice, migrate on the surface, and nucleate islands that continue to grow and eventually complete a new atomic layer. However, surface reconstructions can drastically change established patterns of the surface morphology evolution. In this study, we show that the interplay of surface reconstruction and growth kinetics lead to a distribution of island sizes exhibiting a series of peaks at *magic* sizes [3].

We use high temperature STM measurements in which the growing surface is imaged continuously during growth. This enables us to study directly the influence of the surface reconstruction on growth kinetics and to show how the lateral growth of rows with the width of the reconstruction unit cell leads to a creation of islands of magic sizes. We also perform kinetic Monte Carlo (KMC) simulations of a simple model that captures the main elements of the growth process on the 7×7 reconstructed surface, i.e., the existence of faulted and unfaulted half unit cells and of activation barriers to island growth. With the help of this model we uncover the underlying kinetic mechanisms for the formation of magic islands and estimate the effective barriers involved.

We used a beetle-type STM, which is described in detail in Ref. [4]. Thanks to the open design of the STM, the molecular beam from an Si evaporator can be directed towards the sample which is located in the STM position. Evaporation is done continuously while the STM is scanning the growing film. Si is evaporated from a homemade *e*-beam evaporator. Because of the crystallography of the Si(111) surface, the growth in the vertical direction occurs in units which are 3.1 Å high. We call this unit of 1.56×10^{15} atoms/cm^2 one monolayer or 1 ML. The STM measurements were performed in an ultrahigh vacuum chamber (base pressure 3×10^{-11} mbar). We used a sample bias voltage of $+2$ V and a tunneling current of 300 pA.

During submonolayer growth of Si on Si(111), two-dimensional (2D) islands (1 ML high) are observed. The form of the islands is triangular. This is the *kinetically* limited growth shape evolving during growth. Equilibration of these islands without external flux results in a transition to a hexagonal equilibrium form [4,5]. In Fig. 1(a), the Si island size distribution after deposition of 7% of an atomic layer is shown (sample temperature 725 K, deposition rate 0.5 ML/min). The size of the islands is plotted in units of half a 7×7 unit cell (HUC). Several peaks are observed in the distribution of Fig. 1(a), in particular, a narrow peak at a size of 4 HUC. This observed multiple-peak shape of the island size distribution is quite different from the island size distributions with only one broad maximum observed experimentally for other systems [6].

The typical single-peaked distribution can be explained by assuming that island growth is limited only by the number of adatoms deposited in a "capture zone" closer to this island than to other islands. The island size distribution is thus similar to the distribution of Voronoi polygons around the islands [7,8]. Kinetic constraints due to the barriers to attachment of adatoms to the islands on Si(111) substrate completely change this simple picture.

In the following, it will be shown qualitatively how the well-known 7×7 reconstruction of the Si(111) substrate influences the growth behavior and leads to the experimentally observed multiply peaked island size distribution. The rhombic unit cell of this reconstruction consists of two triangles. One of these triangles has a stacking fault in the surface layers, relative to the substrate stacking

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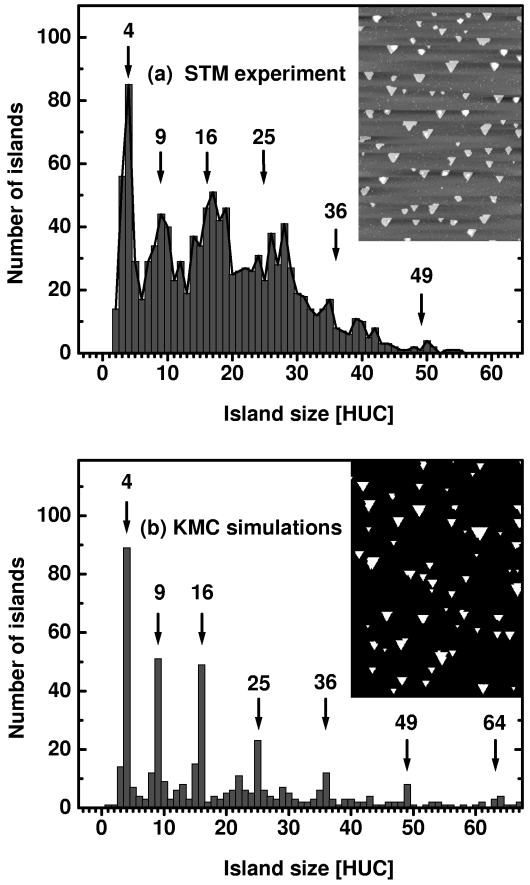

FIG. 1. (a) Experimentally observed island size distribution of two-dimensional Si islands epitaxially grown on Si(111) at 7% surface coverage. The distribution consists of several peaks at magic sizes. The size is expressed in half unit cells of the 7×7 reconstruction unit cell. An STM image of the triangular islands is shown in the inset (2000×3000 Å^2). (b) Island size distribution and the surface morphology (100×120 part of a 400×400 lattice is shown in the inset) observed in KMC simulations.

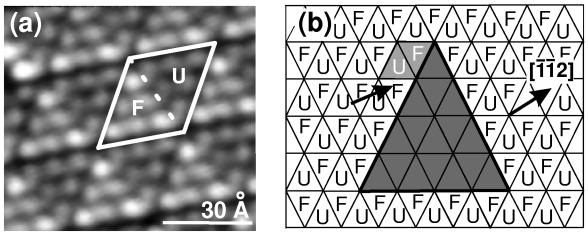

FIG. 2. (a) STM image of the Si(111)-7×7 reconstruction. The white protrusions are the Si adatoms. The unit cell is indicated by a white rhombus. A dashed line divides two triangular subunits. Since the image is taken at a negative sample bias (-2 V), the adatoms in the faulted half of the unit cell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

(F-HUC). The other triangle of the reconstruction unit cell is unfaulted relative to the substrate (U-HUC), Fig. 2(a).

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. The main ingredient of the model for island growth which we introduce in the following is the assumption that it requires different energy barriers to lift the reconstruction of the U triangle as compared to the F triangle [9]. To lift the reconstruction in the U triangle, only atoms in the 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. 2(b), a Si island (gray) and the U and F triangles of HUCs of the surrounding reconstructed substrate surface are shown. Because of the crystallographic orientation of the island, it is surrounded only by substrate F-HUCs. This means that further lateral growth (requiring 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. 2(b)]. 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 7×7 unit cell.

Because of our capability to perform growth and STM imaging simultaneously, growth of a selected island can be observed as a function of time. Figures 3(a)–3(f) show STM images from a growth sequence of such an island. In Fig. 3(a), the shape of the island is triangular. Images 3(b)–3(d) show the same island at a later stage during growth. As shown by images 3(b)–3(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 Figs. 3(b)–3(d). An analysis of the width of this row and further atomically resolved images show that the width of such a row is 27 Å, which is just the width of one 7×7 reconstruction unit cell [10].

A deeper insight into kinetics of island growth on Si(111) surface has been obtained with the help of KMC simulations. We used a simple coarse-grained model with the HUC as a basic unit. This spatial coarse graining

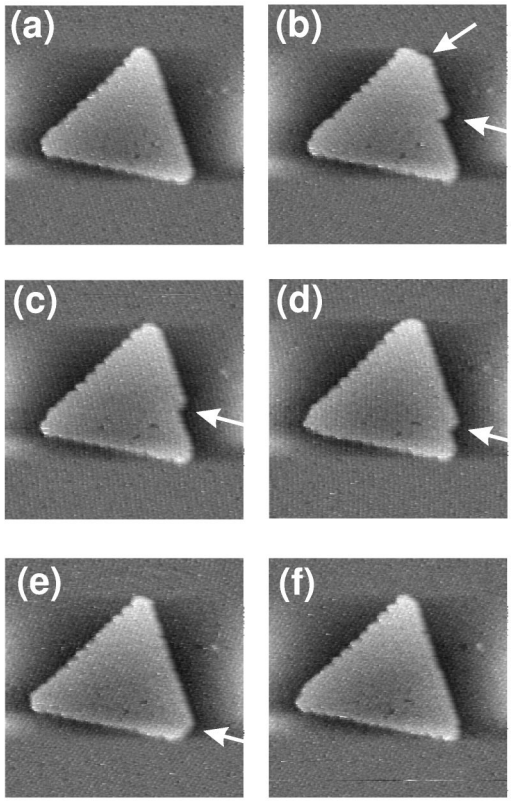

FIG. 3. Sequence of images showing the lateral growth of a triangular Si(111) island. A row of the width of the 7×7 unit cell is growing along the right edge of the island (b)–(f). The image size is 500×500 Å^2, $T = 575$ K. The complete growth sequence is available as a movie on the World Wide Web: (http://www.kfa-juelich.de/video/voigtlaender).

enables us to study the surface morphology evolution in an effective way and focus on the observed behavior of the growing islands. On the other hand, spatial coarse graining leads also to coarse graining in the time domain [11] and means that the energy barriers obtained are effective model parameters.

In the model, the surface is represented by a honeycomb lattice consisting of F and U sites representing HUCs. Deposition and transport of material toward islands take place in the units of HUCs [12]. The hopping rate is given by an Arrhenius expression, $k_0 \exp(-E_D/k_BT)$, where $k_0 = 10^{13}$ s^{-1}, $E_D = E_S + nE_N$ is the barrier to hopping consisting of a substrate contribution E_S and a contribution nE_N from n lateral nearest neighbors ($n = 0, 1, 2, 3$), T is the substrate temperature, and k_B is the Boltzmann's constant. A new island nucleates with the rate $k_0 \exp(-E_{\text{nuc}}/k_BT)$ if at least three neighboring HUCs (U-F-U) become occupied [9].

New HUCs attach to existing islands with the rate of $k_0 \exp(E_{\text{att}}/k_BT)$ where the attachment barrier E_{att} depends both on the type of the underlying HUC (U or F) and the number n_{edge} of nearest-neighbor HUCs that are already a part of the growing island, $E_{\text{att}} = E_{U/F} - n_{\text{edge}}E_{\text{edge}}$. In order to simplify the model, we set the barrier for overgrowth of U-HUCs to zero ($E_U \equiv 0$), set the barrier to detachment of HUCs from islands to a very large value, and forbid nucleation on top of islands. None of these restrictions is inherent to the model and any of them can be easily lifted.

Simulations can be used to infer effective values of the activation barriers playing a major role in kinetics of island growth. Experiments provide the information that can be used to determine these barriers, such as the island density and shapes, the average time for growing a complete row, and the average delay between growth of successive rows for an island of a certain size. The results shown in this paper were obtained using the values that provided the best agreement with experimental data: $E_S = 1.3$ eV, $E_N = 0.2$ eV, $E_F = 2.6$ eV, and $E_{\text{edge}} = 0.35$ eV at $T = 700$ K and $F = 0.01$ ML/s (island size distributions) or 10^{-5} ML/s (a single island evolution). In order to keep the number of free model parameters as low as possible, we deliberately set $E_S = 1.3$ eV and $E_{\text{nuc}} \equiv E_F - E_{\text{edge}} = 2.25$ eV and vary only E_N, E_F, and E_{edge}. It is interesting to note that the best correspondence with the experiment has been achieved with $E_F = 2.6$ eV, which corresponds just to the measured stacking fault energy density for Si (4.28×10^{-3} eV/ Å^2) [13] multiplied by the area of a HUC.

From the model calculations, the evolution of individual islands as a function of time can be extracted. Figure 4 shows the size of one growing island as a function of time. Several plateaus are visible, most of them at a horizontal line indicating completion of one row (a magic size). The existence of the plateaus shows that the island does not grow for awhile after completion of a row of the 7×7 unit cell width. This is related to the large activation barrier for the nucleation of a new row ($E_{\text{att}} = E_F - E_{\text{edge}}$).

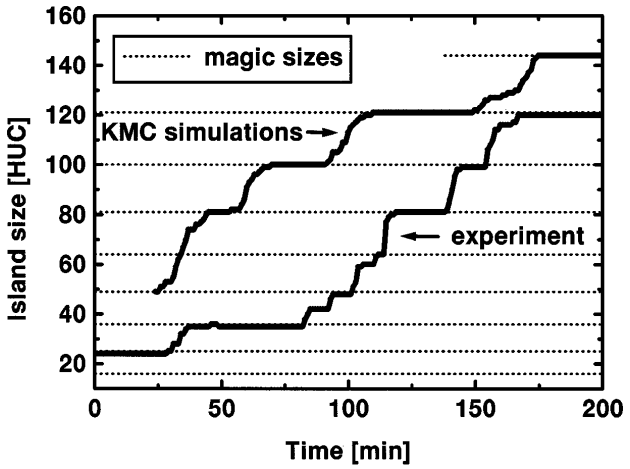

FIG. 4. KMC and STM results for the evolution of a single island size as a function of time. After rapid growth of rows, longer times without further growth result in plateaus in the time evolution. These plateaus occur just at the magic sizes of perfectly triangular islands.

The rate-determining step during lateral growth is the nucleation of a new row. A steep increase of the island size in between the plateaus corresponds to a fast growth of a new row. Dynamical STM measurements of the evolution of single islands show behavior very similar to simulations, Fig. 4, with plateaus near the magic sizes.

These results, obtained by dynamical measurements during growth of a single island and KMC calculations, explain naturally the experimentally observed island size distribution in Fig. 1(a). The interruption of island growth after the completion of a row and the formation of a "closed shell" structure leads to a larger quantity of islands of this magic size. Since the sizes between the magic ones are run through faster due to the fast growth of the rows, the number of such islands in a snapshot image is lower. This leads to the minima in the island size distribution between the magic sizes.

An island size distribution obtained in simulations is shown in Fig. 1(b). The observed qualitative agreement suggests that the model correctly captures the basic features of homoepitaxy on Si(111) 7×7 reconstructed substrate. As can be seen from the schematic image of an island in Fig. 2(b), the magic sizes of the closed shell triangular islands have a size of n^2 HUCs ($n > 1$). The smallest triangle, consisting of 4 HUCs, shows up as the highest and narrowest peak in the experimental island size distribution. Also at the subsequent magic sizes of 9, 16, and 25 HUCs maxima in the experimental island size distributions are observed. For larger magic sizes, the number of observed islands is too low to resolve peaks clearly. In contrast, it is possible to distinguish even the peak at $n^2 = 64$ in simulations, Fig. 1(b).

Note, however, that the "magic" peaks are more broadened in experiment because growth of rows in reality is more complicated. The rows can get pinned by defects and, in addition, nucleation on top of existing islands may shift the island size from the magic one [14].

In conclusion, our results demonstrate that islands of magic sizes can occur not only due to their energetically stable configurations [3] but also due to growth kinetics. The surface reconstruction of Si(111) is responsible for the existence of a nucleation barrier to further growth of magic islands with a closed shell structure. We believe this to be only one example of the strong influence of surface reconstructions on the surface morphology evolution during epitaxial growth.

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