Direct Tests of Microscopic Growth Models using Hot Scanning Tunneling Microscopy Movies

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We use a hot scanning tunneling microscope to make time lapse movies of the growth of Si on a Si(001) substrate. In the initial stages of molecular beam epitaxial growth at 530 K, many small one dimensional (1D) islands are formed. The explanation of this curious shape anisotropy has been controversial. We analyze movies acquired during deposition and follow changes in *individual islands* to find that the growth rate of 1D islands is independent of length, supporting a model of anisotropic sticking to explain island shape anisotropy. We find the ratio of sticking at side sites versus end sites is 0.019 ± 0.003.

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With the development of the scanning tunneling microscopy (STM), it is now possible to observe growth on an atomic scale [1,2]. The hot STM, which images surfaces at elevated temperatures, provides the opportunity to observe the growth process as it occurs [3]. Sequential imaging and growth has been demonstrated and was used to observe the large scale evolution of three dimensional Ge islands on Si [4]. We introduce the use of hot STM movies to follow the growth of individual islands on the atomic scale. This will allow us to visualize the growth process and to test atomistic models of growth. In this paper we study the initial stages of Si molecular beam epitaxial (MBE) growth on Si(001) surfaces using a hot STM in ultrahigh vacuum. We track the growth and evolution of *individual islands* through time lapse STM movies.

The Si(001) surface is the basis for most lithographic and nanofabrication methods and consequently has been the subject of many experimental and theoretical growth studies. The growth of Si on Si(001) has been used as a model system of the study of homoepitaxial growth in general. It is known that the Si(001) surface reconstructs to form parallel rows of surface dimers. Because of the crystal structure, the rows are perpendicular in adjacent atomic terraces, as one can see in Fig. 1. Therefore, terrace edges are alternately aligned parallel (type *A* step edge) or perpendicular (type *B* step edge) to the dimer rows in the upper terrace [5]. Previous STM experiments [6,7] and theoretical calculations [8–11] indicate the Si(001) surface exhibits highly anisotropic diffusion. The mobility of Si monomers and dimers is high along the dimer rows and low across them. It has been observed that for certain growth conditions deposited Si atoms form islands a single dimer row wide (1D islands). The long axes of these 1D islands are aligned perpendicular to the substrate dimer rows [1,2]. Perpendicular 1D islands are also observed in the initial stages of chemical vapor deposition of Si on Si(001) [12]. Low coverages of Al, Ga, In, and Sn form perpendicular 1D islands on Si(001) as well [13]. It is puzzling that the islands grow perpendicular to the direction of fast diffusion. Because the mobility is high along the rows, one might expect that islands would capture atoms from a greater distance in this direction. This would lead to growth predominantly along the rows, contrary to what is observed. Several models have been proposed to account for this surprising growth feature.

Mo *et al.* annealed 1D Si islands to determine whether the 1D growth mode is energetically favored [14]. They found that long 1D islands with aspect ratios up to 20:1 were converted into large 2D islands with aspect ratios near 3:1. This equilibrium aspect ratio of 3:1 corresponds to the factor of 3 energy difference between the two different step edge types [15]. The fact that low aspect

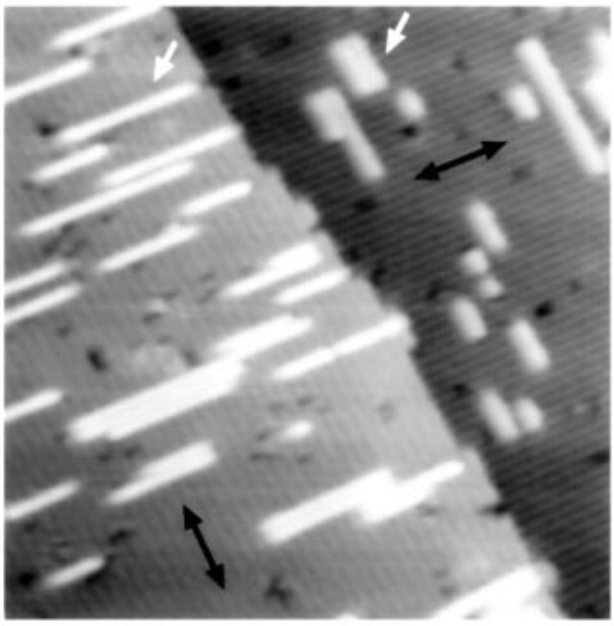

FIG. 1. 40 nm × 40 nm image of the Si(001) surface at 536 K showing many small Si islands. The direction of fast monomer diffusion is parallel to the dimer rows, indicated by the black arrows. Islands growing on both the upper and lower terraces have their long axes aligned perpendicular to the substrate dimer rows. Examples of 1D and 2D islands are indicated with white arrows.

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ratios are energetically favored suggests that a kinetic mechanism is responsible for the increased island shape anisotropy observed during growth.

Mo *et al.* proposed a kinetic model based on sticking anisotropy to explain the island shape anisotropy [14]. If island ends are stickier than island sides, arriving adatoms stick preferentially to the ends, resulting in the growth of many 1D islands. Metiu, Lu, and Zhang proposed an alternative model based on an exchange mechanism [16]. Such mechanisms have recently been discovered in metal growth and diffusion experiments [17]. Metiu suggested that an adatom arriving on the side of a Si island may displace an existing island atom to the top of the island. The displaced atom diffuses rapidly along the top of the island (the direction of fast diffusion) until reaching an end, where it can fall over the edge and stick. Adatoms arriving on island sides are transported to the ends via this exchange mechanism, resulting in enhanced 1D growth.

In Fig. 1 we show an image of the Si(001) surface decorated with many Si islands. The surface steps down from the lower left to the upper right. The upper terrace terminates with a type *A* step edge. As one can see, islands have grown on the upper and lower terraces with their long axes aligned perpendicular to the substrate dimer rows. Closer examination of the image reveals the complexity of the growth process. Both 1D (aspect ratios as high as 15:1) and 2D islands are visible. Missing dimer defects are visible as black areas on the substrate. A complete model for growth must include annealing, coalescence, and nucleation effects, as well as substrate defects, substrate temperature, and sticking anisotropy. Because of the large number of free parameters, developing a complete growth model based only on comparison to grown films can be difficult.

Our experimental apparatus, sample preparation, and temperature determination have been discussed elsewhere [18]. Si is evaporated from a resistively heated wafer inclined at a 30° angle to the sample plane. During the deposition, the STM tip is retracted 500 nm from the surface to avoid obstructing the sample. A deposition lasts 10 sec at a rate of 0.001 monolayer (ML) per second and a substrate temperature of 530 K. This produces optimal growth of 1D islands, as shown in Fig. 2. We find that for low deposition rates, 1D islands grow well within a 50 K window around 530 K. At lower temperatures, mobility is decreased, so atoms deposited on the surface nucleate new islands before attaching to existing islands. This results in increased island number density and smaller 1D islands. At higher temperatures islands quickly anneal and become 2D (with aspect ratio of roughly 3:1). We can image the surface immediately after the deposition, but we wait in order to minimize image distortion. The distortion arises from a thermal pulse caused by evaporator radiation. Waiting roughly 10 min after a deposition allows the thermal pulse to dissipate, thereby reducing image distortion. We use surface features to identify the same spot on the sample before and after a deposition. A series of images of the surface after sequential evaporations composes a time lapse movie of the growth process.

A portion of one movie is shown in Fig. 2. The first image shows the clean substrate before any deposition and the following frames (excerpts from the movie) show the surface as the coverage increases from 0.03 to 0.07 ML.

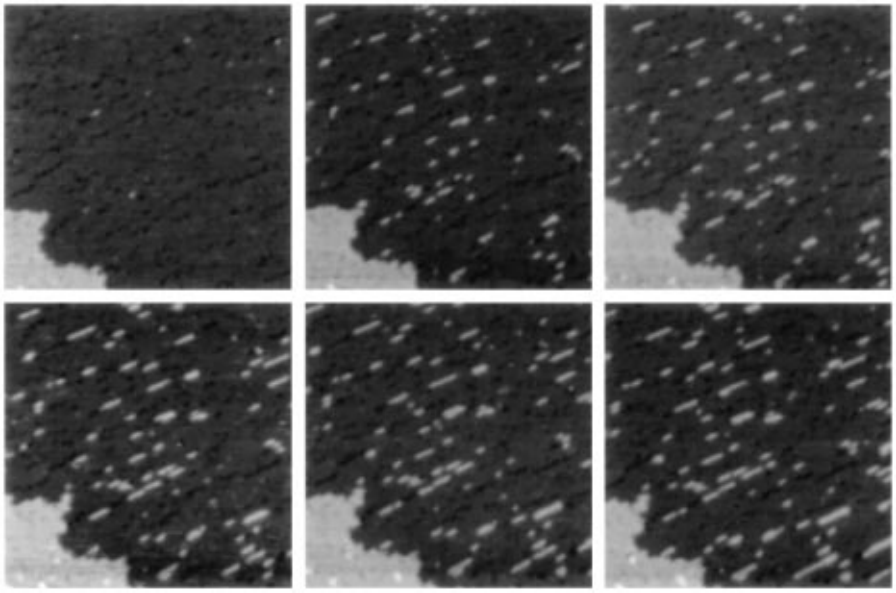

FIG. 2. Six 80 nm × 80 nm images from a longer movie showing the growth of 1D islands at 523 K. Time advances from left to right, top to bottom. Coverage increases from 0 to 0.07 ML. The movie begins with the clean substrate before deposition. The following frames show the condition of the surface after sequential depositions starting at 0.03 ML. One can follow individual islands from frame to frame as they grow.

We find that the movies aid considerably in visualizing the growth [19]. The ability to observe microscopic changes on any given island from frame to frame allows us to study changes in island morphology directly. We observe that changes to islands, either from annealing or from growth, occur in sets of four atoms. This is consistent with changes observed in step edges annealing [20–23]. One set of four atoms, which we term a block, occupies a 0.77×0.77 nm square on the substrate.

As discussed above, it has been shown that on this surface islands transform under annealing at 575 K, becoming 2D [14]. To test for the amount of annealing occurring during the sequential depositions, we repeatedly image one area at 543 K for 35 min with no deposition. By counting the number of blocks attaching or detaching from the islands, we set an upper bound of 1.1×10^{-5} changes per end site per second on the islands. At this slow rate, annealing will not have a significant effect on our growth results. To image faster or to follow growth at higher temperatures in real time will require the development of new and faster STM instruments.

According to the simple sticking anisotropy model of 1D island growth, a long 1D island should be no more likely to capture material at its ends than a short one, for both islands have just two ends. The Metiu model, however, implies that the effective end capture probability should increase linearly with length. Because the exchange mechanism transports adatoms arriving on island sites to island ends, doubling an island's length should double the rate at which material is added to its ends.

To test this prediction we analyze three growth movies with temperatures ranging from 523 to 533 K. We use custom analysis software to locate the islands on each image and compute their positions, lengths, widths, and areas [24]. By tracking island positions in subsequent frames of the movie, the computer follows the growth of individual islands. The three movies consist of 46 separate frames with an average of 50 1D islands per frame. In two of the movies we track 80% of the 1D islands, while in the third we track 65%.

We observe 393 cases of 1D islands growing longer by gaining blocks at end sites. We compute the average gain for islands of each particular length l and divide it by the average gain for all islands. We find the relative gain R = (average gain for 1D islands of length l)/(average gain for all 1D islands). We choose this ratio because we expect it to be independent of factors that could change as the growth progresses, such as the nucleation rate, coverage, and island size distribution. One advantage of our method is that we can select and follow just the 1D islands. We stress that because the relative gain incorporates only averages of independently tracked microscopic events, its calculation and interpretation require no model specific arguments or parameters.

Figure 3 shows a plot of the relative gain R versus length l. According to the Metiu model, larger islands should capture a higher than average amount of material, giving the plotted points a pronounced positive slope. In fact, we see that the ratios are very close to unity for islands 1 to 9 blocks long. A relative gain ratio of unity means that growth is independent of length, supporting the anisotropic sticking model. In response to this result, Metiu has suggested that the exchange mechanism may still be active, but that after the atom is pushed on top of the island it may fall off the side before reaching the end [25]. In any case, it is clear that the exchange mechanism does not enhance the shape anisotropy. The simple anisotropic sticking model is therefore confirmed as the cause the island shape anisotropy observed during growth. We note that for lengths above 5 blocks the ratios drop slightly below unity. This is likely due to the presence of defects on the surface. As an island grows longer, it is more likely that an end will become pinned by a defect, thereby suppressing the relative gain ratio.

We now use our data to measure the anisotropic sticking ratio. We observe 41 cases of islands becoming 2D by gaining blocks at side sites (we omit the few cases where the block disappears in the next frame). We find that the probability of side growth events increases linearly with island length, whereas the probability of end growth events is independent of length. This suggests that the probability of growth at a side or end site is a constant, and that the ratio of these constants is a measure of the sticking anisotropy. The non-normalized probabilities of growth at a side site, P_S, and at an end site, P_E, are

$$P_S = \frac{\sum_l [n_S(l)/2l]}{N_S}, \quad P_E = \frac{\sum_l [n_E(l)/2]}{N_E}. \qquad (1)$$

In each case $n(l)$ is the number of growth events observed for islands of length l and N is the total number of

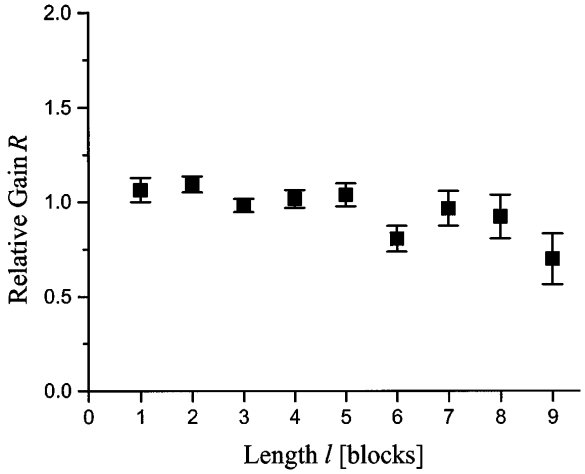

FIG. 3. Plot of relative gain R as a function of 1D island length. The relative gain is R = (average gain for 1D islands of length l)/(average gain for all 1D islands). The growth of 1D islands is found to be independent of length. This result supports a model based on anisotropic sticking to explain island shape anisotropy observed during growth.

observations. Note that we divide $n_S(l)$ by $2l$, the number of sided sites, and $n_E(l)$ by 2, the number of end sites. We find P_S/P_E for each movie and compute the weighted average, yielding a sticking anisotropy ratio of 0.019 ± 0.003. Thus, an end site is roughly 50 times more likely to gain a block than a side site.

This result gives us insight into MBE growth of Si on Si(001). Although the temperature dependence of the sticking anisotropy has yet to be explored, we expect the large anisotropy to survive at the slightly higher temperatures used in real MBE growth. It is known that in step flow growth, type *B* step edges grow faster than type *A* edges, eventually causing double height steps to form [26]. Although most material arriving at a step edge is incorporated at existing kink sites [23], the creation of new kinks (by addition of material to previously flat sections) is the rate limiting step for the advance of the edge. Because the side of a 1D is a type *A* edge, adding a block there is like adding a block to a flat section of a type *A* step edge. The end of a 1D island is a type *B* edge; adding a block there is like adding a block to a flat section of type *B* step edge [27]. One therefore expects the creation of kinks (addition of blocks) at the *B* edge to be much more likely than at the *A* edge, with a similar disparity in step growth rates. Thus the rapid growth of the type *B* step edge and the highly anisotropic island shapes are both results of the sticking anisotropy. This is in agreement with recent *ab initio* calculations which show that the type *B* step edge acts as a strong sink for adatoms [28]. The stronger binding at the end of a row localizes the atoms until another adatom arrives, thereby facilitating growth at that site. Our measured sticking anisotropy, together with our previous measurements of edge fluctuations, can also provide a detailed quantitative picture of the coarsening of small islands or features on the Si(001) surface.

We present a novel technique to study island nucleation and growth on an atomic scale. Using a combination of hot STM movies and analysis of individual islands, we test two competing kinetic growth models. We find that the exchange mechanism that operates in some metal on metal systems does not significantly affect the growth of Si islands. For growth on the Si(001) surface, the large sticking anisotropy of 50:1 overwhelms the large diffusional anisotropy. The method presented here can also be used to study the growth of larger 2D islands and step edges as a function of their initial size or shape. It can be applied to a wide range of growth systems, providing accurate parameters needed for the refinement of growth models. We are at an exciting moment when the combined experimental and theoretical work on the Si surface will soon permit an accurate and complete understanding of diffusion, coarsening, and growth processes over a wide range of temperatures.

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