Dynamic observation of Si crystal growth on a Si(111)7×7 surface by high-temperature scanning tunneling microscopy

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The dynamic process of Si crystal growth on a Si(111)7×7 surface was studied *in situ* using high-temperature scanning tunneling microscopy. Si was evaporated onto a Si(111)7×7 surface, kept at 350 °C, and the crystal growth was observed. The step-flow growth was observed as the appearance of new adatoms at the step edge. The $[11\bar{2}]$ steps became jagged with $[\bar{1}\bar{1}2]$ steps. At the $[\bar{1}\bar{1}2]$ steps, new adatoms appeared in rows along the step edges.

The homoepitaxial growth of Si on Si(111) surfaces has been extensively studied because of both its technological importance and scientific interest. A clean Si(111) surface has a dimer adatom stacking-fault (DAS) (Ref. 1) structure, which is very different from a 1×1 structure. As a result, its growth mechanism has attracted much attention, and has been studied using a variety of analytical techniques, such as reflection high-energy electron diffraction (RHEED),[2] reflection electron microscopy (REM),[3,4] and scanning reflection electron microscopy (SREM).[5] These studies revealed the dynamic growth processes, such as the movement of steps and the nucleation of two-dimensional (2D) islands. Scanning tunneling microscopy (STM) has also been employed to reveal growth at an atomic resolution. An *in situ* study of the morphology of the grown surface[6] revealed epitaxial triangular islands with $[\bar{1}\bar{1}2]$ steps, which have lower energy than $[11\bar{2}]$ steps.

The termination of both higher and lower terraces on a Si(111)7×7 surface is of great interest in view of how the 7×7 structure grows. The step structure of the annealed surface has been investigated with STM.[7,8] Tochihara *et al.*[8] discussed stable terminations of the 7×7 structure at the steps which depended on the phase difference between the 7×7 structures of the higher and lower terraces. They showed that a step was always terminated at a stable position for an *annealed* surface. However, it is generally believed that a step does not always coincide with the stable position *during growth.* Ragged arrangements of adatoms at step edges of the grown surface[6] suggest that the step-flow growth is not quantized in units of the 7×7 unit mesh to maintain the stable termination of the step.

STM has been widely used in surface science because of its atomic resolution. In addition, recent advancements in the STM technique have enabled the observation of dynamic surface processes at high substrate temperatures, such as observing the phase transition of a Si(111) surface from the 1×1 to the 7×7 structures,[9] and the metal adsorption process onto a Si surface.[10]

This paper reports on a STM observation of dynamic Si growth on a Si(111)7×7 surface to show how new step edges are formed. The sample was a Si wafer (0.01 Ω cm, 7×2×0.1 mm^3), whose surface was cleaned by flashing at 1200 °C and then kept at 350 °C by direct joule heating during the observation of the growth. The orientation of the sample was determined from filled state images. Si atoms were supplied to the wafer surface from a Si evaporator, which was a tiny piece of the same wafer positioned 15° from the sample surface and heated to more than 1200 °C. Although the tip was very close to the sample surface in the STM observation, Si was able to grow in the areas under observation due to the small angle of the Si evaporator. The pressure in the vacuum chamber was maintained below 1×10^{-9} Torr (the base pressure was 3×10^{-10} Torr) during the evaporation. STM images were taken every 15 sec and recorded on a videotape. The growth rate was controlled less than 0.5 bilayer per hour so that the dynamic processes at the atomic level could be observed in the observations that occurred every 15 sec. The images were recorded in a differential mode to eliminate the effect of the severe thermal drift in the *z* direction caused by the evaporation.

A series of STM images taken during the Si growth, at 350 °C, on the Si(111)7×7 surface is shown in Fig. 1. These are large scanned images, so they do not have atomic resolution. The jagged dark line running vertically is a monatomic-height (3.2 Å) step, and the terrace on the left side is higher than that on the right side.

Just after the flashing, most $[11\bar{2}]$ steps were observed to have straight edges. However, they then developed jagged shapes (as shown in Fig. 1) as the growth proceeded. The arrows in each image indicate the places where significant growth was observed. For instance, the step edges indicated by the arrows in Fig. 1(b) move to the right compared with Fig. 1(a). This shows that the higher terrace has grown by step-flow growth in the intervening 15 sec. The island on the higher terrace has also grown. In the first 15 sec, substantial growth was observed at the two places indicated by the arrows in Fig. 1(b).

The step between the higher and lower terraces, which was observed to be a straight $[11\bar{2}]$ step just after flashing, has grown in a parallel shape to that of the island. This growth into a jagged shape can be explained by the transition from an unstable $[11\bar{2}]$ step to a stable $[\bar{1}\bar{1}2]$

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step.

A series of images showing the step-flow growth at atomic resolution is shown in Fig. 2. The right side terrace of the step is higher than the left side terrace. The dark depression at the lower edge of the step is an artifact due to the differential imaging. The arrows in Figs. 2(b) and 2(c) indicate adatoms which have newly appeared in each image. For instance, three new adatoms appeared in the first 15 sec, as indicated by the arrows in Fig. 2(b). They correspond to adatoms at the 7×7 unit mesh edge. In the next 15 sec, two new adatoms appeared. These images suggest that single adatoms appear one by one during growth. The arrowheads in Fig. 2(a) indicate some adatoms whose locations are different from the 7×7 unit mesh edge. These observations indicate that the step edge where growth occurs does not always coincide with the 7×7 unit mesh edge, and that the structure of the 7×7 unit mesh or half mesh is not formed at one time during growth.

It should be noted that adatoms always exist at the step edge where the growth occurs. Neither a Si atom in a stacking-fault layer nor a dimer of a DAS structure was observed at the step edge where the growth took place. An adatom of a DAS structure, and not a 7×7 unit or a half unit mesh, appears to be the smallest unit of the growth.

The arrangement of adatoms at the higher edge of the step is slightly disordered. In particular, that of the sharp edge of the step in Fig. 2(a) is disordered. This displacement of adatoms suggests that the DAS structure is not always the most stable structure at the step edge during growth, although adatoms are essential to reduce the surface energy. As growth proceeds, the arrangement of such adatoms becomes similar to that of a DAS structure. The half unit cell drawn in Fig. 2(c) indicates an area where such rearrangement occurred. Notice a corner-hole-like structure cannot be seen at one of the apexes of this half unit cell, where there is a new step edge.

A part of the jagged step edge is shown in Fig. 3. The right side terrace of the step is higher than the left side terrace. The triangular shaped area in the higher terrace was formed with stable $[\bar{1}\bar{1}2]$ steps, as discussed above. The $[\bar{1}\bar{1}2]$ step edges in Fig. 3 are almost straight, although they do not correspond to the 7×7 unit mesh edge. At the edges, new adatoms appear in the form of a row along the edges. For instance, the arrow indicates a short row consisting of four adatoms.

A series of STM images of the area indicated by white lines in Fig. 3 is shown in Fig. 4. The images are shown at 30-sec intervals. There are three rows of adatoms along the step edge in Fig. 4(a). Two of them indicated

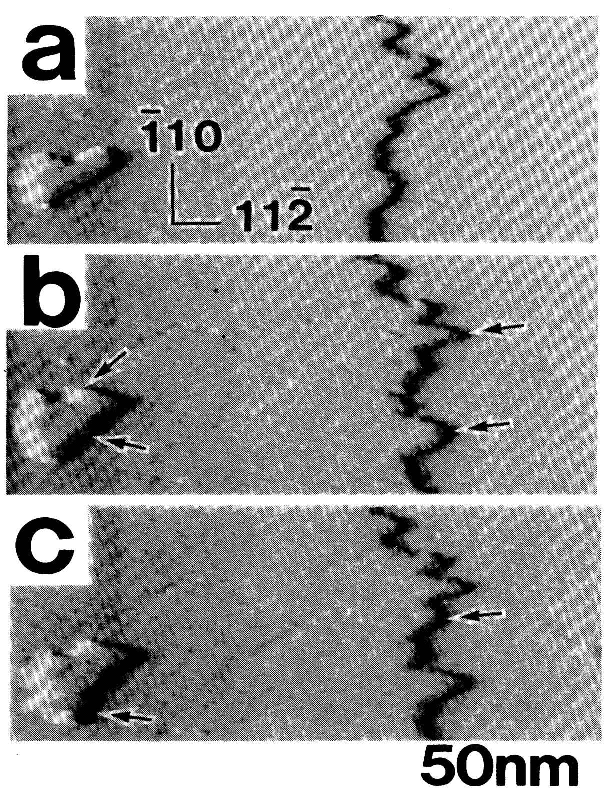

FIG. 1. A series of STM images taken every 15 sec during Si growth on a Si(111)7×7 surface at 350°C. Both step-flow growth and island growth can be seen. Arrows indicate the places where significant growth was observed compared to the previous image.

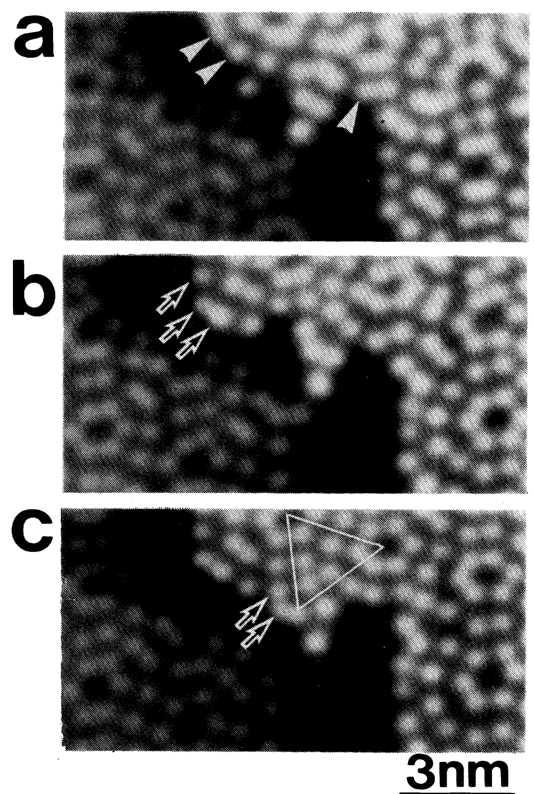

FIG. 2. A series of STM images showing step-flow growth. The step edge does not always coincide with the 7×7 unit mesh edge during the growth. A few new adatoms of a DAS structure appear in each image, and these are indicated by the arrows.

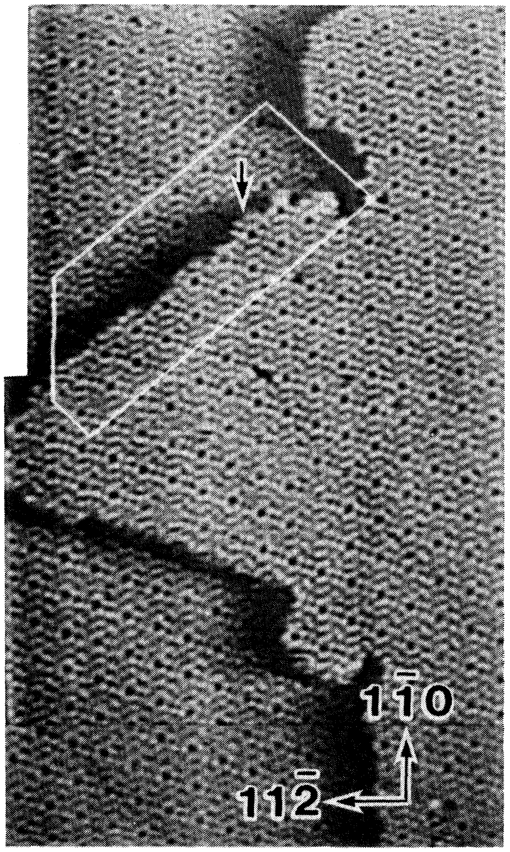

FIG. 3. An image of the terrace grown into a jagged shape with $[\bar{1}\bar{1}2]$ step edges. The formed $[\bar{1}\bar{1}2]$ step edges are almost straight although they do not coincide with the 7×7 unit mesh edge.

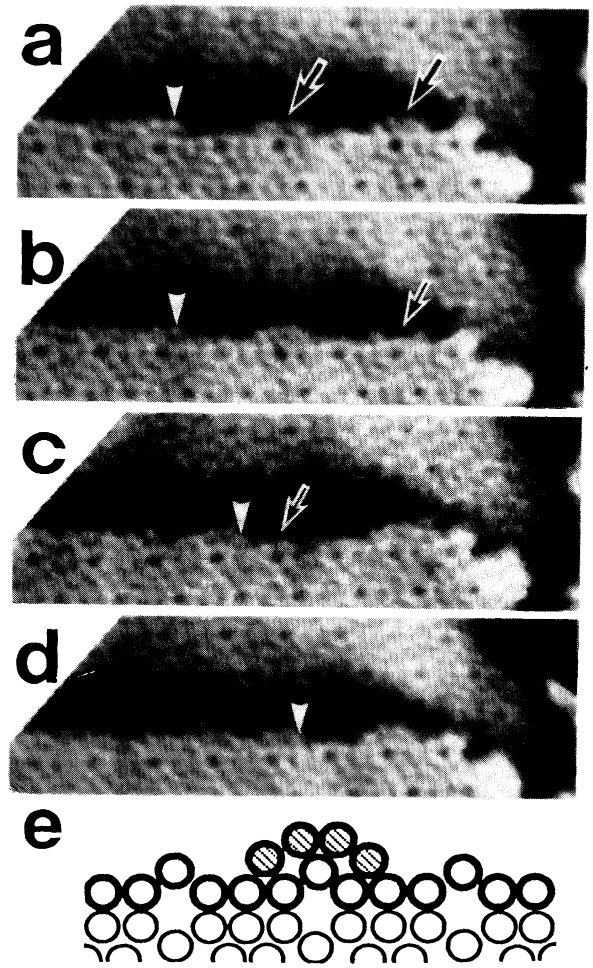

FIG. 4. (a)–(d) A series of STM images showing the step-flow growth at the $[\bar{1}\bar{1}2]$ step edge. Short rows of adatoms indicated by arrows are observed to be unstable. (e) A schematic of the short row. Four adatoms drawn by hatched circles form a row along the step edge, which is formed by a complete row of adatoms drawn by heavily outlined open circles.

by arrows consist of four adatoms each, as illustrated in Fig. 4(e). They were observed to be unstable; two adatoms of the row on the right side disappear in Fig. 4(b) and the other row disappears in Fig. 4(c), as indicated by arrows in each image. On the other hand, the longer row which is prolonged from the left side of the images did not disappear during the growth. The row grew two unit mesh lengths of the 7×7 structure from Figs. 4(a) to 4(d). Arrowheads indicate the end of the row. The formation of the rows along the step edge was observed only during the growth.

At the end of the row, the lower terrace cannot be smoothly bonded to the upper terrace and some dangling bonds remain in the lower terrace. Therefore, short rows, which have both ends close to each other, are energetically unstable, and they are thermally diffused to form a longer row which is energetically stable. We believed that the $[\bar{1}\bar{1}2]$ step maintains a straight edge by forming complete rows along the edge one by one.

In conclusion, the dynamic process of Si growth on a Si(111)7×7 surface was studied using STM. The new adatoms in the step-flow growth were observed at the step edge. An adatom was observed to be the smallest unit of growth, and a step edge does not always coincide with a 7×7 unit mesh edge during growth. At the $[\bar{1}\bar{1}2]$ step edge, new adatoms formed rows along the edge. The $[\bar{1}\bar{1}2]$ step edge is believed to expand row by row to reduce the number of ends of rows, that is, to reduce the number of dangling bonds in the lower side terrace of the step.

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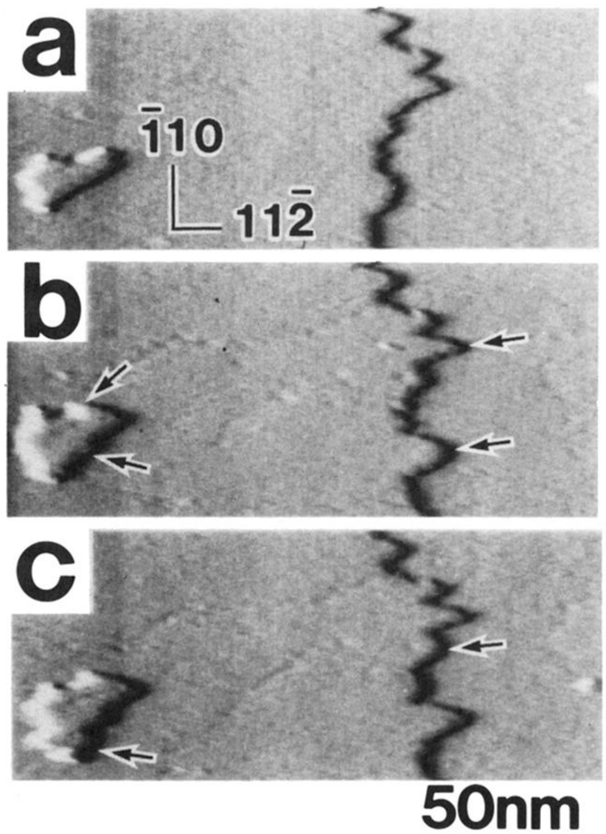

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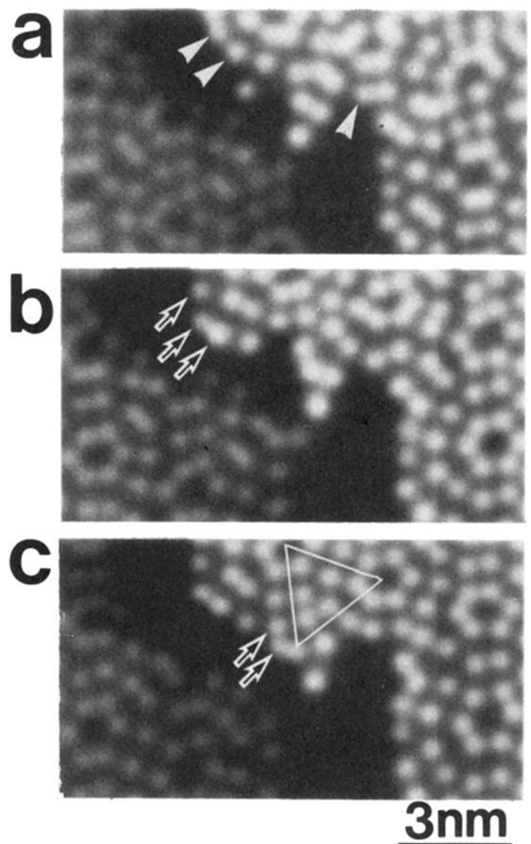

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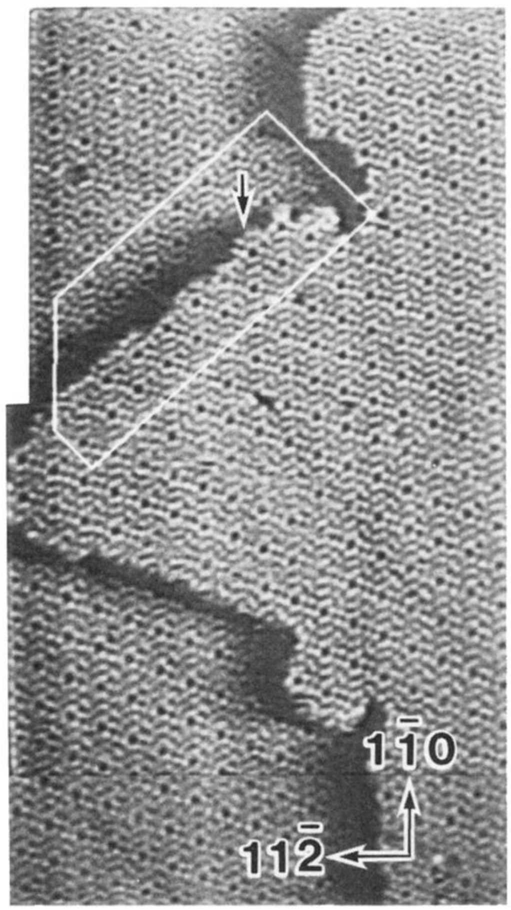

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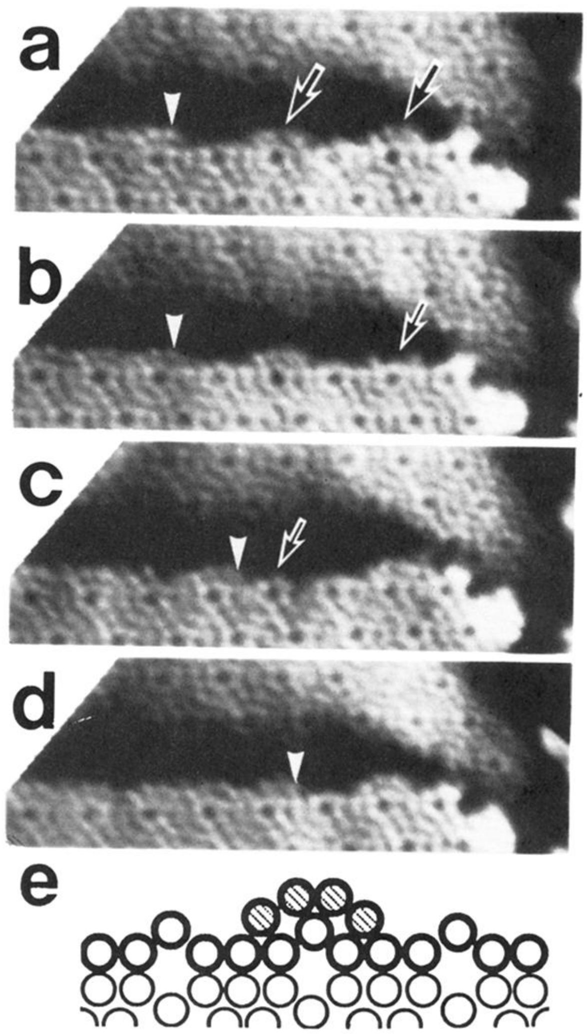

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