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Observation of Si(001) surface topography at temperatures below 1140 °C using a reflection electron microscope

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The changes in the surface topography of a Si(001) vicinal sample caused by heating it to temperatures below 1140 °C by direct current are investigated by reflection electron microscopy: the surface normal of the sample is misoriented by less than 2′. At temperatures below 1000 °C, the surface is mostly covered by preferential terraces (for example, 2×1 terraces) determined by the direction of the heating current. At temperatures below about 850 °C, pairs of steps are uniformly distributed on the surface, while step bands are formed by the aggregation of steps at temperatures near 950 °C. A few wide terraces remain between the step bands: in our sample, they are about 5 μm wide in the direction parallel to the current. At temperatures between 1000 °C and 1100 °C, the step bands relax and the narrow terraces (1×2 terraces) widen due to the evaporation of Si atoms. At temperatures between 1070 °C and 1100 °C, atomic steps appear due to the relaxation in pair steps; both the 2×1 and 1×2 terraces coexist with approximately equal widths (about 1 μm). At temperatures above 1100 °C, the surface topography changes to one composed of mosaic domains due to the evaporation of the atoms. For the Si(001) vicinal surface, the evaporation temperature of Si atoms is about 1000 °C for Si atoms released from the S_b step and about 1100 °C for the atoms released from the S_a step, where the evaporation temperature is determined by the activation energy, which consists of the release energy and the evaporation energy. [S0163-1829(97)05004-2]

I. INTRODUCTION

Surface topography affects many physical and chemical properties, including crystal growth, catalysis, and adsorption. Surface topographies have been investigated using optical microscopy,[1–3] scanning electron microscopy,[4–7] reflection electron microscopy (REM),[8–15] low-energy electron microscopy,[16,17] and scanning tunneling microscopy (STM).[18–20]

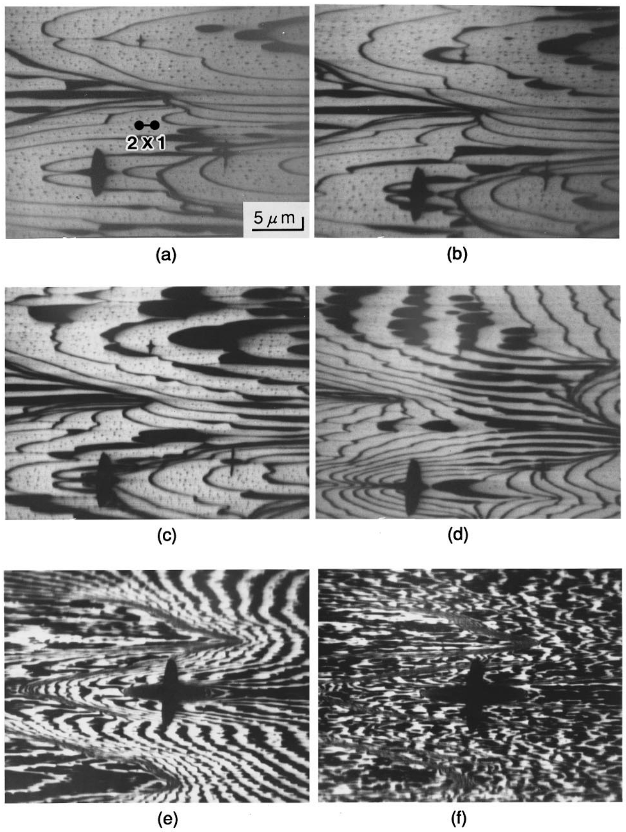
The surface topography of a clean Si(111) surface has been investigated at temperatures below about 1400 °C using REM.[10] It has also been investigated using STM at temperatures below about 1000 °C.[20] In both investigations, atomic steps aggregated or separated on the surface depending on the heating conditions (the direction of the heating current and the temperature). On the other hand, the surface topography of a clean Si(001) surface has been investigated using REM (Ref. 11) and STM (Ref. 20) at temperatures below about 1000 °C. On the Si(001) surface, dimers are formed by reconstruction of the surface atoms. Depending on the direction of the heating current, the surface is mostly covered by a 2×1 terrace (with a step-up current) or a 1×2 terrace (with a step-down current). On the 2×1 terrace, the 2×1 dimer is parallel to the direction of the heating current, while on the 1×2 terrace, the 1×2 dimer is perpendicular to the 2×1 dimer on the surface.

We investigated the surface topography of a Si(001) vicinal sample at temperatures below about 1140 °C using REM. Based on the results, we estimated the evaporation temperatures of the Si atoms using a simple model.

II. EXPERIMENT

We used a Si(001) vicinal sample (CZ-N, 8–12 Ω cm, <2′) with four sides corresponding to the ⟨110⟩ directions. A clean Si(001) surface was obtained after the sample was flash heated to about 1200 °C for 2–3 s by passing a direct current through it. When the sample was heated to 950 °C for 20 min by a step-up current, the Si(001) surface was mostly covered by 2×1 terraces.[13–15] When it was heated to 950 °C for 20 min by a step-down current, the surface was mostly covered by 1×2 terraces. The step-up direction is the direction from the down side to the up side at surface steps (the step-down direction is the reverse). This direction corresponds to the ⟨110⟩ direction on the Si(001) surface.

We investigated the surface topography of the Si(001) sample at temperatures below about 1140 °C using REM when the sample was heated by direct current. To fix the surface structures against electromigration, the sample was heated by the step-up current for 2×1 terraces and by the step-down current for 1×2 terraces.[13–15] The REM images were produced using the 2×1 spot in the diffraction pattern; the bright region in the REM image always corresponds to the 2×1 terrace. The images were mostly observed after the sample was cooled to room temperature. In some cases, REM images were observed *in situ* at the heating tempera-



(a) (b)

(c) (d)

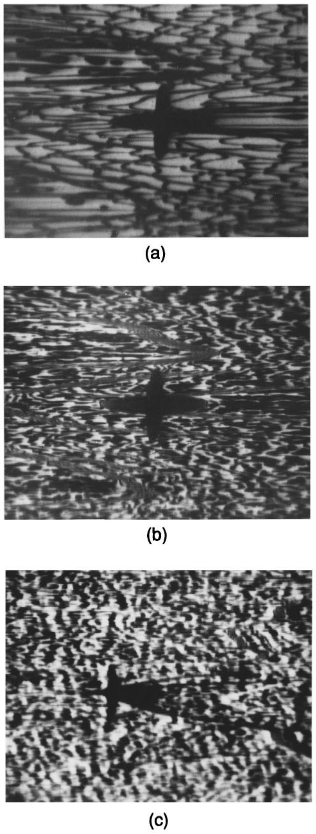
(e) (f)

FIG. 1. REM images of a Si(001) 2×1 surface heated to (a) 980 °C, (b) 1000 °C, (c) 1025 °C, (d) 1070 °C, (e) 1100 °C, and (f) 1140 °C. The images were observed after the sample was cooled to room temperature.

ture. The base pressure of the vacuum chamber was kept at about 1×10^{-8} Pa, the acceleration energy of the electron beam was 20 keV, the glancing angle of the beam was about 3°, and the beam diameter was about 20 nm.[21]

III. RESULTS

For the Si(001) 2×1 sample heated by the step-up current, REM images after the sample was cooled to room temperature are shown in Fig. 1. We assume that the atom inflow balanced the atom outflow at each step on the surfaces. The direction of the 2×1 dimer is shown in Fig. 1(a). At temperatures below about 980 °C, the surface was mostly covered by the 2×1 terraces (bright regions). Although the atomic steps on the surface may have changed their positions, the surface topography after heating was similar to the topography before heating. However, at temperatures between 1000 °C and 1070 °C [Figs. 1(b)–1(d)], the 1×2 terraces (dark regions) widened in the direction parallel to the current due to heating. When the sample was heated to about 1100 °C [Fig. 1(e)], both the 2×1 and 1×2 terraces of approximately equal width coexisted on the surface. Furthermore, the atomic steps were approximately parallel to a certain direction on the surface. At a temperature of about 1140 °C [Fig. 1(f)], many mosaic domains were created on the terraces. These domains had areas less than 500×20 nm^2; the limited microscopic resolution prevented a more precise measurement.



(a)

(b)

(c)

FIG. 2. REM images of a surface (a) before heating, (b) after 60 s of heating at 1140 °C, and (c) after flash heating. The images were observed after the sample was cooled to room temperature.

REM images of the sample heated to 1140 °C are shown in Fig. 2. After heating for 60 s [Fig. 2(b)], the topography had similar properties to that after flash heating [Fig. 2(c)]. These results show that at 1140 °C the Si atoms at the surface had already started to evaporate.

For the Si(001) 1×2 sample heated by the step-down current, REM images after the sample was cooled to room temperature are shown in Fig. 3. The direction of the 1×2 dimer is shown in Fig. 3(a). At temperatures below about 980 °C, the surface was mostly covered by 1×2 terraces (dark regions). Again, the surface topography after heating was similar to the topography before heating. At temperatures between 1000 °C and 1070 °C. [Figs. 3(b)–3(d)], the 2×1 terraces (bright regions) widened in the direction parallel to the current. At about 1100 °C [Fig. 3(e)], the 2×1 and 1×2 terraces also coexisted on the surface. Furthermore, mosaic domains had already formed at about 1140 °C [Fig. 3(f)].

To estimate how the cooling process of the sample affects the surface topography, we compared the REM images obtained at the heating temperatures with those obtained at room temperature. The Si(001) 2×1 sample was heated to temperatures below about 1090 °C by the step-up current, as shown in Fig. 4. The REM image obtained at 1050 °C had properties similar to the image taken after cooling the sample to room temperature [Figs. 4(a) and 4(b)]. The image obtained at about 1090 °C also had properties similar to those of the image obtained at room temperature [Figs. 4(c) and 4(d)]. In this experiment, the sample was cooled to a temperature below 500 °C in less than 3 s. We neglected the

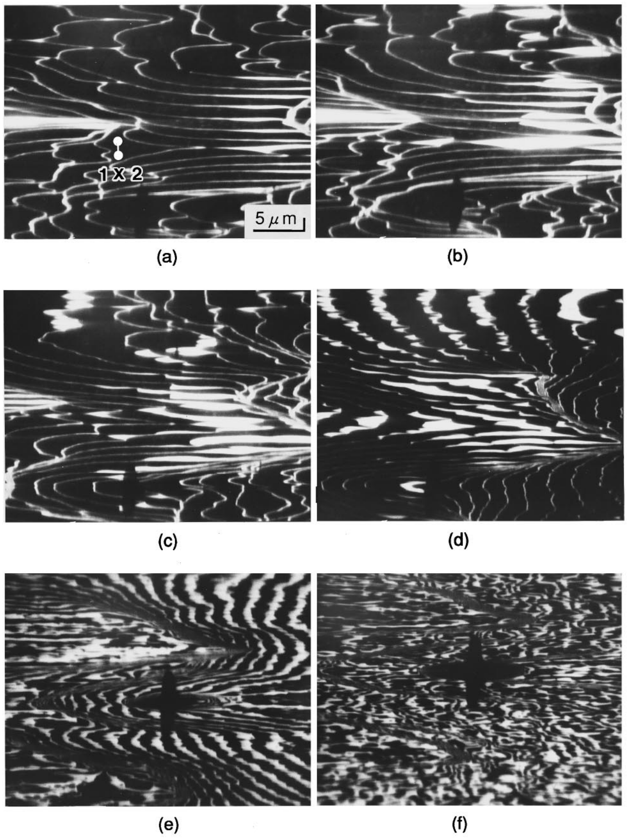


FIG. 3. REM images of a Si(001) 1×2 surface heated to (a) 980 °C, (b) 1000 °C, (c) 1025 °C, (d) 1070 °C, (e) 1100 °C, and (f) 1140 °C. The images were observed after the sample was cooled to room temperature.

effect of thermal diffusion because of the small diffusion constant at temperatures below 500 °C. On the Si(001) surface, therefore the REM images obtained *in situ* at heating temperatures below 1090 °C matched the images obtained at room temperature well. Based on these results, we conclude that the cooling process of the sample does not affect the surface topography.

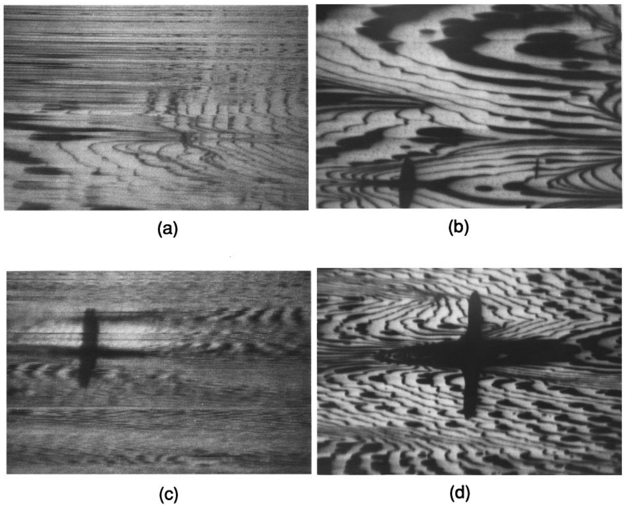


FIG. 4. *In situ* observations of a Si(001) surface: (a) the REM image obtained at 1050 °C and (b) the image after cooling the sample to room temperature; (c) the image obtained at 1090 °C and (d) the image after cooling it to room temperature.

IV. DISCUSSION

When the sample was heated by the step-up current for 2×1 terraces and by the step-down current for 1×2 terraces, we observed that the width of the preferential terraces (for example, 2×1 terraces) became narrow at temperatures between 1000 °C and 1100 °C. If thermal diffusion of Si atoms dominated the changes in the surface topography, then every narrow terrace (for example, the 1×2 terrace) would widen on the surface: thermal diffusion has a strong effect on a narrow terrace. Furthermore, every narrow terrace on the surface would have the same width. However, we actually observed that the wider preferential terraces started to narrow at lower temperature, as shown in Figs. 1 and 3. In addition, as shown in Fig. 2, the atoms at the surface already started to evaporate at 1140 °C. From these results, we conclude that the surface topography changes due to the evaporation of Si atoms at about 1000 °C and again at about 1100 °C.

The evaporation of Si atoms involves electromigration as well as the evaporation process (elementary process) itself. The electromigration of the Si substrate atoms consists of three elementary processes: release, diffusion, and capture.[22] The Si atoms are released from a substrate step and migrate across the terrace in the same direction as the electric field. Then Si atoms may be captured by the neighboring substrate step. At temperatures below the evaporation temperature T_e of the Si substrate atom, Si atoms are always captured when they arrive at the substrate step, so we regarded the probability of capture as being 1. The surface topography of the sample is determined by the electromigration of Si substrate atoms. According to the model, the activation energy of this process (the surface diffusion energy) consists of the release energy and the diffusion energy. On the other hand, at temperatures above T_e, the Si atoms evaporate from the terrace instead of being captured. Here the surface topography of the sample is determined by the evaporation of Si substrate atoms. Likewise, according to the model, the activation energy of this surface evaporation consists of the release energy and the evaporation energy. Based on this result, we conclude that the surface topography changes at the evaporation temperature T_e due to the evaporation of Si atoms.

A. Surface topography of a Si(001) sample at high temperatures

We investigated how the surface topography of the Si(001) 2×1 sample varied with the heating temperature (Fig. 5). There are two types of steps native to a Si(001) surface: the S_a step, which is the front edge of the 2×1 terrace, is more stable than the S_b step, which is the front edge of the 1×2 terrace.[23] When the sample was heated by the step-up current, at temperatures below 850 °C [Fig. 5(a)], the surface separated into the 2×1 terraces (bright regions) and the steps had small radii. At temperatures between 900 °C and 1000 °C [Fig. 5(b)], the steps aggregated and step bands more than ten atomic layers high (above 2 nm) were formed on the surface. The 2×1 terraces remaining between the step bands became more than 5 times as wide as those obtained at temperatures below 850 °C and the steps

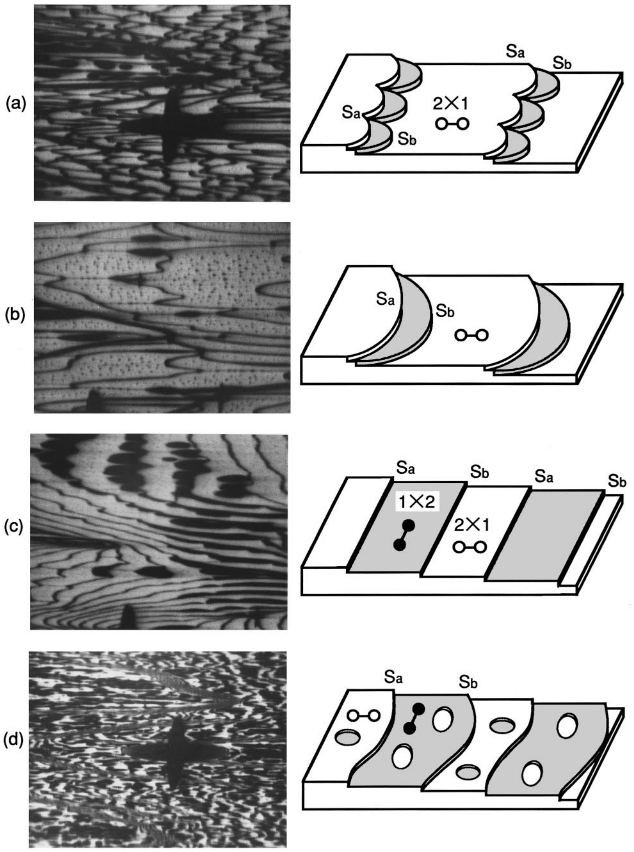


FIG. 5. Surface topographies of a Si(001) 2×1 sample heated to (a) below 850 °C, (b) between 900 °C, and 1000 °C, (c) between 1000 °C and 1100 °C, and (d) above 1100 °C.

remaining between the step bands had a large radius. We observed that the 2×1 terraces had an average width of about 5 μm in the direction parallel to the current.

At temperatures between 1000 °C and 1100 °C [Fig. 5(c)], the number of step bands on the surface decreased and the width of the wide 2×1 terraces became narrower due to the evaporation of Si atoms. We conclude that the atoms released from the S_b steps evaporated from the surface before they could be captured by the neighboring S_a step, while atoms released from the S_a steps were captured by the neighboring S_b step before they could evaporate. This imbalance caused the S_a steps to retreat, which widened the 1×2 terraces (dark regions). In the equilibrium condition, the surface consisted of both the 2×1 and 1×2 terraces with a constant ratio between their widths. At about 1100 °C, the step bands disappeared from the image, except for those higher than at least 2 nm. Therefore, the terrace width was approximately equal to the width determined by the tilt angle from the [001] direction: all steps were approximately parallel to each other on the surface. Here we observed that the terraces had an average width of about 1 μm in the direction parallel to the current (the ratio ≈1).

At temperatures above 1100 °C, the atoms released from the S_a and S_b steps mostly evaporated from the surface before they could be captured at neighboring steps. Here, at about 1140 °C, mosaic domains are created on the terraces, as shown in Figs. 1 and 3. If the Si atoms evaporate from every dimer row on the surface, then the terraces would change to a rugged surface with heights varying by a few atomic layers [Fig. 5(d)]. We conclude that every dimer row on the surface corresponds to an atomic step, as shown in Fig. 6.

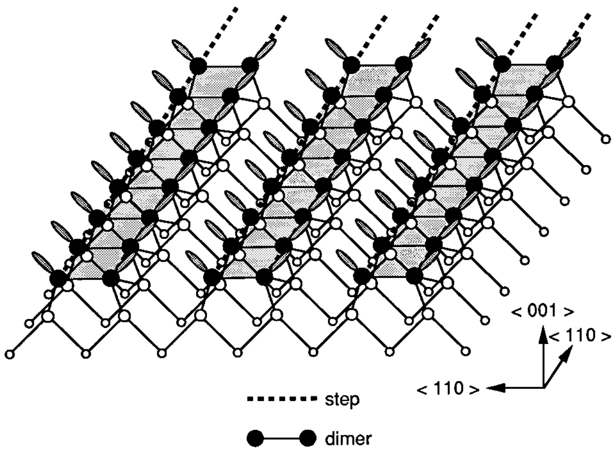


FIG. 6. Each dimer row corresponds to an atomic step. At high temperatures, Si atoms evaporate from every dimer row.

Similar results were obtained for the 1×2 surface when the sample was heated by the step-down current. However, our results do not agree with those previously obtained using REM.[11] In that study, a double-domain surface, which was covered with periodic arrays of the 2×1 and 1×2 domains, was obtained at temperatures between 800 °C and 900 °C. Conversely, a single-domain surface was obtained at temperatures above 900 °C. The width of each domain was about 80 nm at temperatures between 800 °C and 900 °C, while the domain width in our study was about 1 μm at 1100 °C. The difference was because the tilt angle in the previous study was larger than that in our study. We suspect that the double-domain surface was obtained at lower temperatures in the previous study because of the narrower terrace width.

B. Evaporation of Si atoms from a Si(001) surface

Since all Si atoms on the surface (but not those at steps) have the same properties in the direction of the surface normal, the evaporation energy (elementary process) of Si atoms on the 2×1 terrace is the same as that on the 1×2 terrace. According to the model, at temperatures above T_e, the difference in the surface evaporation energies is due to the difference in the release energies. We considered the release processes from steps for four different conditions: (i) the S_b step in the step-down direction, (ii) the S_a step in the step-down direction, (iii) the S_b step in the step-up direction, and (iv) the S_a step in the step-up direction. We assumed that the release energy increases in this order, with the energy in case (i) being the smallest and that in case (iv) being the largest.

The growth of narrow 1×2 terraces (dark regions) on a surface with wide 2×1 terraces (bright regions) is illustrated in Fig. 7. Here the sample was heated by the step-up current. At temperatures below the evaporation temperature (1000 °C) [Fig. 7(a)], since Si atoms remain on the terrace and migrate across it to the neighboring step by electromigration, the surface after heating retains a structure similar to

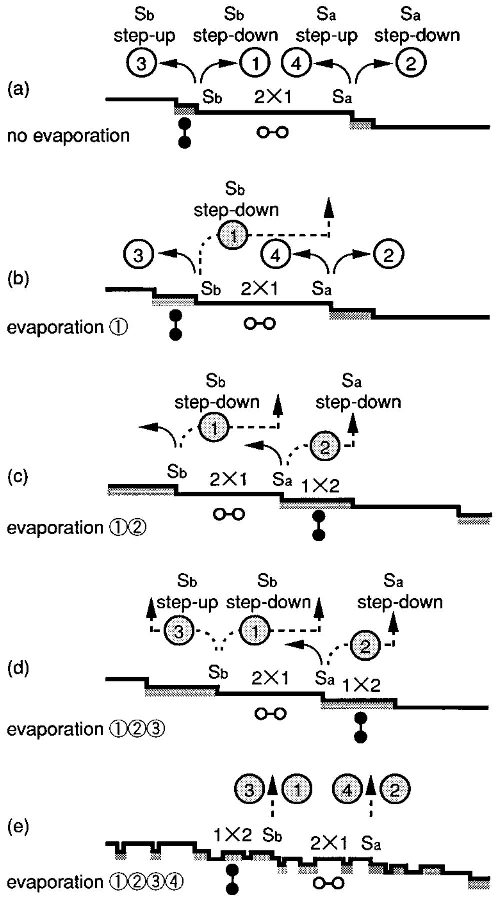


FIG. 7. Growth of a narrow 1×2 terrace on a surface with wide 2×1 terraces. (a) At temperatures below 1000 °C, the surface retains its structure after heating. (b) At 1000 °C, Si atoms released from S_b steps in the step-down direction evaporate from the terrace. The activation energy of this surface evaporation consists of the release energy from S_b steps and the evaporation energy. (c) At temperatures above 1000 °C, Si atoms released from S_a steps in the step-down direction also evaporate; however, Si atoms released from the S_b steps in the step-down direction dominate the evaporation of Si atoms. The surface topography does not change even though there is the step movement. (d) Then, Si atoms released from the S_b steps in the step-up direction evaporate; however, Si atoms released from the S_b steps in the step-down direction still dominate the evaporation of Si atoms. (e) At temperatures above 1100 °C, Si atoms released from the S_a steps in the step-up direction evaporate. The activation energy of this surface evaporation consists of the release energy from the S_a steps and the evaporation energy. Here all atoms evaporate from the surface.

that before heating. However, at 1000 °C [Fig. 7(b)], Si atoms start to evaporate from the surface. At first, Si atoms released from S_b steps in the step-down direction evaporate from the terrace. Since they do not reach the neighboring S_a steps, the atom inflow at the step does not balance the atom outflow there. This causes the 1×2 terraces (dark regions) to widen. At temperatures above 1000 °C (below 1100 °C), Si atoms released from the S_a steps in the step-down direction also evaporate from the surface. However, based on the time it takes the atoms to evaporate from the terrace, the atoms released from the S_b steps in the step-down direction dominate the evaporation of Si atoms. Therefore, the surface topography does not change even though there is the step movement, as shown in Fig. 7(c). next [Fig. 7(d)], Si atoms released from the S_b steps in the step-up direction also evaporate from the surface. Likewise, based on the time it takes the atoms to evaporate from the terrace, the atoms released from the S_b steps in the step-down direction still dominate the evaporation of Si atoms. Here the surface consists of both the 2×1 and 1×2 terraces with approximately equal widths. At temperatures above 1100 °C [Fig. 7(e)], the atoms released from the S_a steps in the step-up direction evaporate from the surface; thus all atoms evaporate from the surface. Since the atoms evaporate from every dimer row, a rugged surface is created on each terrace.

The process on a surface with wide 1×2 terraces is similar to that on one with wide 2×1 terraces. Likewise, we conclude that the atoms released from the S_b steps in the step-down direction evaporate from the surface at about 1000 °C and the atoms released from the S_a steps in the step-up direction evaporate from the surface at about 1100 °C.

From the electromigration of Si atoms on a Si(001) surface,[22] we estimated the time it takes the atoms to evaporate from the surface. We have previously determined the migration speed of atoms on both the 2×1 and 1×2 terraces.[22] Here, for both the S_a and S_b steps, we assume that the release energy in the step-up direction is the same as that in the step-down direction, though there is a little difference between those energies. At temperatures between 1000 °C and 1100 °C, the speed on the 2×1 terrace is approximately equal to that on the 1×2 terrace: about 4.4×10^{-6} cm/s at 1000 °C and about 1.1×10^{-5} cm/s at 1100 °C. Thus, at 1000 °C, the width of the 2×1 terrace is about 5 μm, as shown in Fig. 1(b). We estimated that Si atoms released from S_b steps in the step-down direction evaporate from the surface within about 2 min. At 1100 °C, the width of the 2×1 terrace is about 1 μm, as shown in Fig. 1(e). We estimated that Si atoms released from S_a steps in the step-up direction evaporate from the surface within about 10 s. These estimations support the changes in the surface topography being due to the evaporation of Si atoms.

C. Step geometry on a Si(001) surface

The distribution of steps varied with the heating temperature for both the 2×1 and the 1×2 terraces (the former is shown in Fig. 8). We call the two steps at the edges of a narrow terrace (in Fig. 8, the 1×2 terrace) a pair of steps. At temperatures below about 850 °C [Fig. 8(a)], we assumed that these pairs of steps are uniformly distributed on the surface: a dispersion of pairs of steps. At temperatures near 950 °C [Fig. 8(b)], step bands higher than ten atomic layers (above 2 nm) are formed on the surface: an aggregation of (pairs of) steps. However, a few pairs of steps remain between the step bands, as shown in Figs. 1 and 3. At temperatures between 1000 °C and 1070 °C [Fig. 8(c)], the step bands relax and pairs of steps are formed on the surface: a dispersion of pairs of steps. At temperatures between 1070 °C and 1100 °C [Fig. 8(d)], the pairs of steps become atomic steps: a dispersion of steps. Then, at temperatures above 1100 °C, the atoms released from all steps evaporate from the surface, producing a rugged surface. This aggrega-

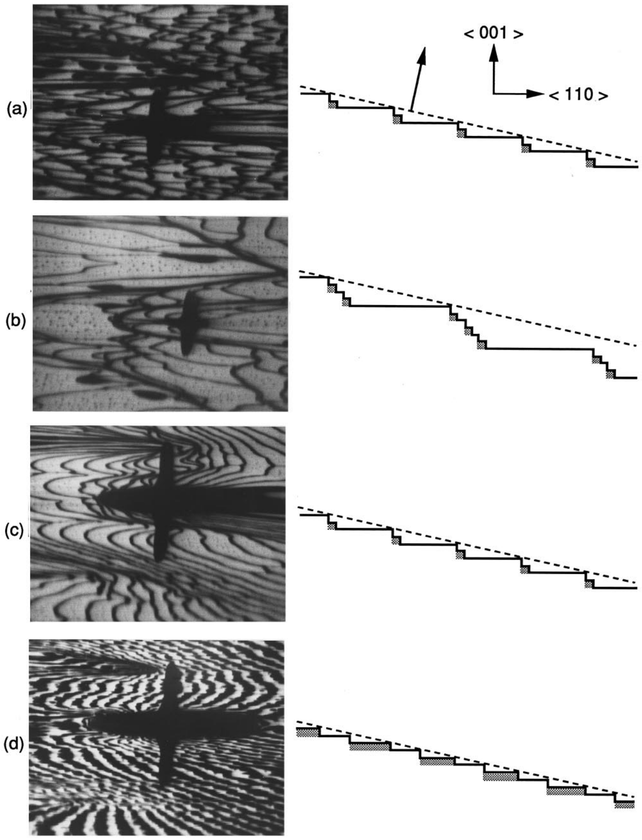


FIG. 8. Distribution of steps on a 2×1 terrace: (a) dispersion of pairs of steps at temperatures below 850 °C, (b) aggregation of steps at temperatures near 950 °C, (c) dispersion of pairs of steps at temperatures between 1000 °C and 1070 °C, and (d) dispersion of steps at temperatures between 1070 °C and 1100 °C.

tion of steps and formation of step bands on a Si(001) surface corresponds to previous theoretical results.[24]

These results suggest that if we control the terrace width on a Si(001) surface, it will be possible to form high-quality films at low temperature by accelerating the step flow growth on the surface. It will also be possible to form low-dimensional structures on the surface.

V. CONCLUSION

For the Si(001) vicinal surface misoriented by less than $2'$, we used reflection electron microscopy to investigate the change in surface topography of the sample when it was heated to temperatures below 1140 °C by direct current. At temperatures below 1000 °C, the surface was mostly covered by a preferential terrace: the 2×1 terrace with the step-up current and the 1×2 terrace with the step-down current. In our sample, the width of the preferential terrace was about 5 μm in the direction parallel to the current. At temperatures between 1000 °C and 1100 °C, the narrow terraces widened in the direction parallel to the current. At temperatures between 1070 °C and 1100 °C, the 2×1 and 1×2 terraces coexisted with approximately equal widths (about 1 μm for our sample). At temperatures above 1100 °C, the surface topography changed to one composed of mosaic domains due to the evaporation of Si atoms.

We estimated the evaporation temperatures of the Si atoms, where the activation energy consists of the release energy and the evaporation energy. The Si atoms released from the S_b step in the step-down direction evaporated from the terrace at about 1000 °C. On the other hand, the Si atoms released from the S_a step in the step-up direction evaporated from the terrace at about 1100 °C. We conclude that the difference in the evaporation temperatures is due to the difference in the release energies of Si atoms.

We also observed how the distribution of the steps changed with the temperature. At temperatures below 850 °C, pairs of steps were uniformly distributed on the surface: a dispersion of pairs of steps. Step bands were formed at temperatures near 950 °C and a few wide preferential terraces remained between the step bands: an aggregation of steps. Then the step bands relaxed at temperatures above 1000 °C: a dispersion of pairs of steps. At temperatures between 1070 °C and 1100 °C, the pairs of steps became atomic steps: a dispersion of steps.

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