Step morphologies on small-miscut Si(001) surfaces

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The morphologies of single height atomic steps were studied on Si(001) samples with a miscut of $\sim 0°–0.05°$ using dark-field low-energy electron microscopy. In addition to the previously observed wavy step phase we identify a "hilly" phase at miscut angles $<0.03°$. This phase exhibits steps in excess of those required by the local miscut, but their morphology is qualitatively different from the straight steps predicted by theory.

The structure of steps on the Si(001) surface has been a subject of intense experimental and theoretical study in recent years.[1–9] It was found early on that large miscut angles result in a dominance of double height (DH) steps, with single height (SH) steps prevailing at small-miscut angle. This DH-to-SH transition has been studied in detail, and is relatively well understood.[7] Recently, an additional phase transition was found at very-small-miscut angles, from *straight* to *wavy* SH steps.[8,9]

To understand the physical driving forces behind these transitions we must consider the atomic structure of the clean Si(001) surface. In the unreconstructed surface, surface atoms have two backbonds, and two dangling bonds. Pairs of neighboring atoms undergo lateral displacements to form a dimer bond, eliminating two dangling bonds per (2×1) unit cell. These dimers, arranged in dimer rows, were directly observed in scanning-tunneling-microscopy (STM) experiments several years ago.[10] Crossing a SH step, the dimer orientation rotates 90°, changing the reconstruction from (2×1) to (1×2) or vice versa. It was shown both theoretically[11] and experimentally[12] that the surface stress tensor of the dimer-reconstructed surface is anisotropic; tensile parallel to the dimer bond, compressive normal to it. The stress tensor rotates 90° at each SH step, but not at a DH step.

At large-miscut angles the surface energy is dominated by the cost of the steps. Due to strong repulsion between SH steps, DH steps have lower energy at large-miscut angles, explaining their predominance. However, since the surface stress tensor is now uniquely aligned over the entire surface, the energy gain of the DH steps is lowered by the surface stress. At smaller-miscut angles, the balance shifts from step energies to surface-stress-related energies. Breaking up the surface in both (2×1) and (1×2) domains results in a lowering of the surface energy. Thus, at smaller-miscut angles DH steps dissociate into SH steps. A theoretical phase diagram on the DH-SH phase transition was recently published, and appears to be in general agreement with experimental results.[7] As the miscut angle gets even smaller, the terrace widths get larger, increasing the domain sizes, which leads to an increase in surface energy at very-small-miscut angles. Theoretical studies suggested that the energy may be lowered by introducing excess pairs of up and down SH steps, thus reducing the size of the reconstructed domains.[5] Alternatively, it was recognized that the surface energy may be lowered by making the steps wavy instead of straight, giving rise to a reduction of domain size parallel instead of normal to the global step direction.[8] Theoretical studies of step waviness and experimental observations with low-energy electron microscopy (LEEM) appear to be in good agreement. No excess steps as suggested by previous theories were observed. Wavy, large terrace width SH steps were found to coexist with narrow straight steps, indicating a first-order phase transition from straight to wavy steps, in agreement with theory.[9]

As the miscut angle is reduced to the limit of 0°, it is obvious that at some point excess steps *must* occur. For this reason we decided to complement our previous studies, at miscut angles of $\sim 0.1°$, with LEEM observations on samples with a miscut of $\sim 0°–0.05°$. On these samples we find wavy steps, as on the 0.1° miscut samples. In addition, we find clear examples of excess step generation, in the form of step loops (hills and valleys) as the miscut angle approaches 0°. This new "hilly" phase is thermally stable and appears to complete the Si(001) step phase diagram.

Our experiments were performed in a recently designed and constructed low-energy electron microscope.[13,14] Briefly, a 15-keV electron beam is focused in the backfocal plane of a magnetic cathode objective lens. Between this lens and the sample (a gap of 2 mm), a potential difference close to 15 kV is maintained, with the sample potential only a few V different from the electron source potential. Thus, the coherent electron beam strikes the sample with an energy of a few eV, and undergoes low-energy electron diffraction (LEED). Then the electrons are accelerated back into the objective lens. A focused LEED pattern is formed in the backfocal plane, and an image of the sample (at magnification 20) at a distance of 30 cm. An aperture in a conjugate diffraction plane is used to select one of the diffracted beams. Selection of the (0,0) beam results in a bright field image, selection of a fractional order beam gives a dark field image. In this study all images were formed with a $(\frac{1}{2},0)$ beam. With SH steps, every other terrace diffracts into this beam and is imaged bright. The terraces in between are dark. Thus, this imaging condition gives strong contrast and is eminently suited to study the morphologies of SH steps. To optimize image resolution, the incident beam does not travel along the optical axis, but is incident on the sample at an angle such that the $(\frac{1}{2},0)$ beam is on the optical axis. This condition is achieved with the aid of electromagnetic steering coils.

Si(001) samples (kindly donated by Wacker Chemitronic) were cut into disks (8-mm diam) and mounted in a Mo cap at the end of an alumina tube. Clean surfaces were obtained after careful degassing at 600 °C by repeated flashing to 1250 °C. The samples were heated by electron bombardment from behind. Temperatures were measured with an optical pyrometer. The LEED pattern was observed immediately after cleaning, and images were obtained within 1 or 2 min after cleaning. The base pressure in the microscope during these experiments was 4×10^{-10} Torr. In these experiments it is essential to obtain a surface without SiC particulates (which cause step pinning). Fortunately, such pinning centers are easily visible in the microscopy, leading to very characteristic step morphologies, quite different from the step morphologies presented in this paper. Good vacuum during sample flashing (in the 10^{-10} Torr regime) and careful degassing of the sample holder are essential.

Figure 1 shows a few typical images of regions of the surface exhibiting wavy SH steps. The waviness is very regular in Figs. 1(a) and 1(b), and very closely analogous to the theoretical results of Tersoff and Pehlke. Images 1(c) and 1(d) are somewhat less regular, but are still rather similar to 1(a) and 1(b). A few two-dimensional (2D) islands are seen in 1(c). These images also bear a close resemblance to our previous results at 0.1° miscut angle.

Very different are the images shown in Fig. 2. Figures 2(a) and 2(b) show a large number of 2D islands, as well as protrusions and depressions which are several SH steps high or deep. Long steps can be seen to wind their way around and between these features. Bandlike areas with relatively large domain sizes are seen in Figs. 2(c) and 2(d). Both the hill- and valley-type features seen in Figs. 2(a) and 2(b) and the bandlike features in Figs. 2(c) and 2(d) correspond to the local occurrence of excess steps on the surface. These excess steps are rather limited in spatial extent, and typically close upon themselves.

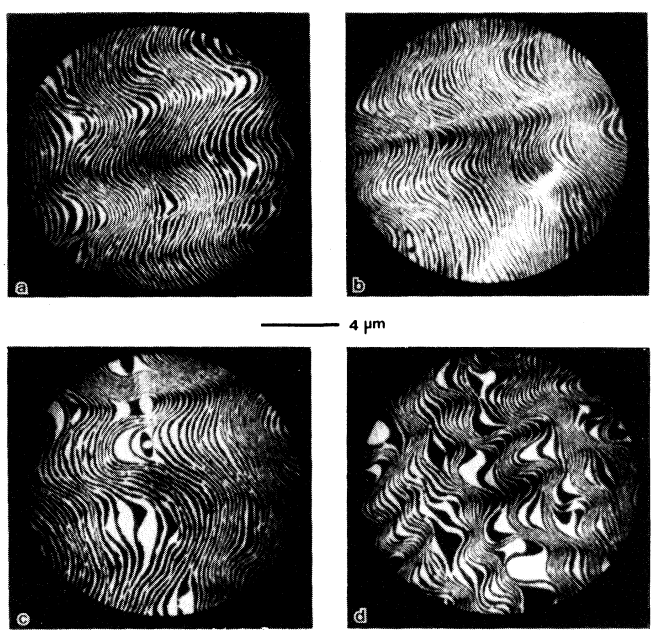

FIG. 1. Dark field LEEM images of the Si(001) surfaces. Miscut angle in (a)–(c) is ~0.05°, in (d) ~0.04°.

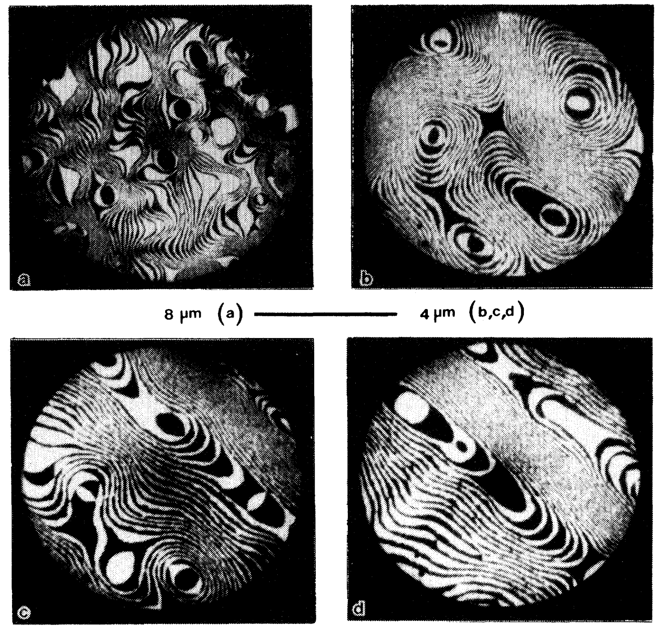

FIG. 2. As Fig. 1. Miscut angle in (a) is ~0.03°, in (b) ~0°. The patterns observed are intrinsic to small miscut. The elongated features seen in (c) and (d) are thought to be extrinsic in nature.

The wave-type regions shown in Fig. 1 and the somewhat rougher regions in Fig. 2 were often found in close proximity of each other. We have investigated the stability of local step configurations during high-temperature sublimation of Si. Under such conditions surface mobilities are very high, and the surface steps (which flow due to evaporation) would have ample opportunity to rearrange themselves, if a driving force for rearrangement were present. Figure 3(a) shows a surface at the onset of evaporation. Figure 3(b) shows the same area at a temperature of ~1130 °C, with rapid evaporation in progress. It is easily recognized that the overall step morphology is not much changed, and is rather stable under these conditions. This can also be seen in Figs. 3(c) and 3(d). In 3(c) a small hill is seen (arrow). Figure 3(d) shows the same area after 40 monolayers of Si have evaporated, much more than the height of the hill, which has nonetheless survived (arrow). These evaporation experiments show that local step morphologies are indeed quite stable, and appear to correspond to (local) energy minima. Only after rapid quenching from 1250 °C to room temperature (by instantaneous disconnection of the heating filament) do we observe structures that are quite unstable. Figure 4(a) shows a clear example, with quaint, distorted domains and ill-defined step morphologies. On somewhat steeper areas, the steps are well defined, but an unusually high number of kissing sites[15] is observed [Fig. 4(b)]. These kissing sites (arrows) mask antiphase domain

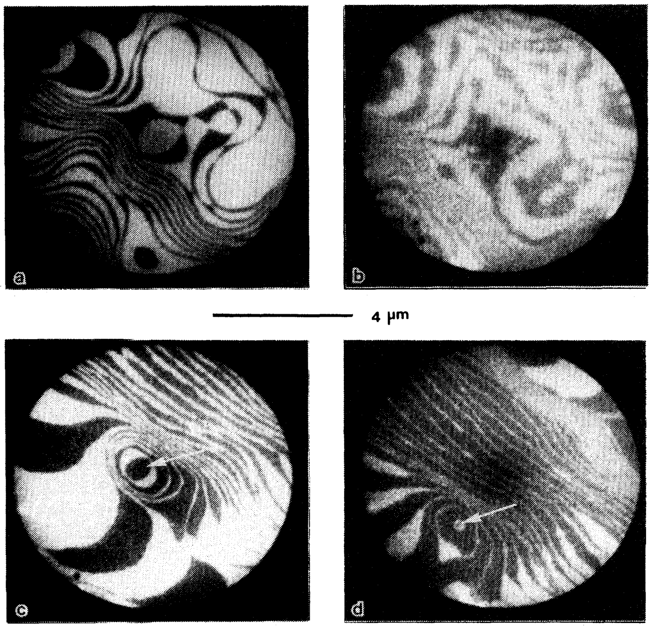

FIG. 3. Local step structures before [(a) and (c)] and during [(b) and (d)] evaporation. The local step morphology does not change during step flow. The arrows in (c) and (d) indicate a stable hill, before and after 40 monolayers of evaporation.

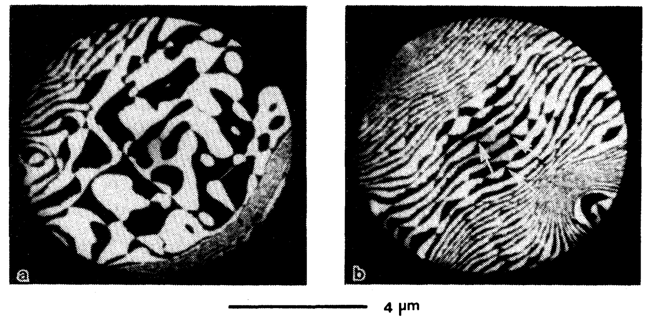

FIG. 4. Metastable structures after quenching from 1250 °C. In (b) arrows indicate a few of the many kissing sites.

boundaries on the underlying terrace. During the rapid quench, reconstructed domains nucleate in many places at the same time, resulting is small domain size and large number of antiphase boundaries.

Figure 1 confirms our previous observations of a distinct wavy phase at very-small-miscut angles. In addition, Fig. 2 provides the first experimental evidence for the occurrence of excess steps on such samples. Both these possibilities have been suggested in previous theoretical studies.[5,8] Once a given morphology has developed, it is remarkably stable, indicating that the surface arrives at least at a local energy minimum, with only weak driving forces to thermodynamic equilibrium. It is important to note that at these very-small-miscut angles, tiny deviations in local sample slope from the average miscut may have large effects. A slope of 7 Å ($\sim$5 SH steps) per μm corresponds to an angle of 0.04°. Thus, if the polishing and sample cleaning procedures leave a waviness in the surface plane of $\sim$7 Å/μm, then the local variations in miscut may be as large as the miscut itself.

With this in mind, the results may be somewhat easier to understand. Figures 1(a) and 1(b) correspond to a local miscut of $\sim$0.05°, Fig. 1(d) to $\sim$0.04°, and Fig. 2(a) to $\sim$0.03°. (Miscuts angles are determined by counting the number of steps in the field of view.) As the miscut gets smaller, larger terraces develop with a pronounced tendency to 2D island and hole formation. It is much harder to determine the local miscut in image 2(b), but from this tendency to form 2D holes and islands as the local miscut approaches 0° we conclude that it is very close to 0°. The bandlike features of Figs. 2(c) and 2(d) are more rare and may be the result of more strongly localized remnants of surface preparation (polishing and cleaning) induced surface undulations.

Thus, as the miscut approaches 0°, the following sequence of phases is observed. At a few degrees miscut the surface undergoes a transition from DH to SH steps. Below $\sim$0.2°–0.1° straight SH steps become unstable versus wavy SH steps, and a first-order phase transition (characterized by the phase separation of uniquely spaced straight SH steps and wavy steps[9]) between these two phases is seen. As the miscut becomes smaller than $\sim$0.03°, the steps are no longer exclusively aligned with the global direction of miscut, and closed step loops are found (2D holes and hills). Very close to 0°, step loops dominate the surface step morphology. These last observations agree with the theoretical expectation that excess steps must form as the miscut angle tends to 0°. However these excess steps are not one dimensional as envisioned by theory, but are, rather, two dimensional and predominantly looped. Of course, these excess steps reduce the reconstruction domain sizes, allowing more efficient relaxation of surface stress. Like the wavy steps, this reduction is more effective in a 2D pattern than in a 1D striped pattern. In any case, if the miscut angle is exactly 0° no preferential excess step direction should exist on a global scale. The images shown here indicate that there is no local preference either, i.e., the surface does not break up in areas with *locally* 1D step patterns, but exhibits 2D step patterns everywhere. None of the step morphologies observed are extremely regular, but they do appear to correspond to a local minimum in surface free energy. This is demonstrated by the stability of local morphologies during evaporation, as shown in Fig. 3. The lack of long-range regularity is the result of this local stability, which impedes the progress towards thermodynamic equilibrium. It was shown by Tersoff and Pehlke that the driving force toward equilibrium (given the presence of wavy steps) is quite weak anyway, further explaining the lack of regularity.

In conclusion, four different phases occur on the Si(001) surface with decreasing sample miscut: DH steps, SH straight steps, SH wavy steps, and a new SH "hilly" phase, exhibiting excess step loops. This last phase has not been considered previously and appears to complete the Si(001) step phase diagram.

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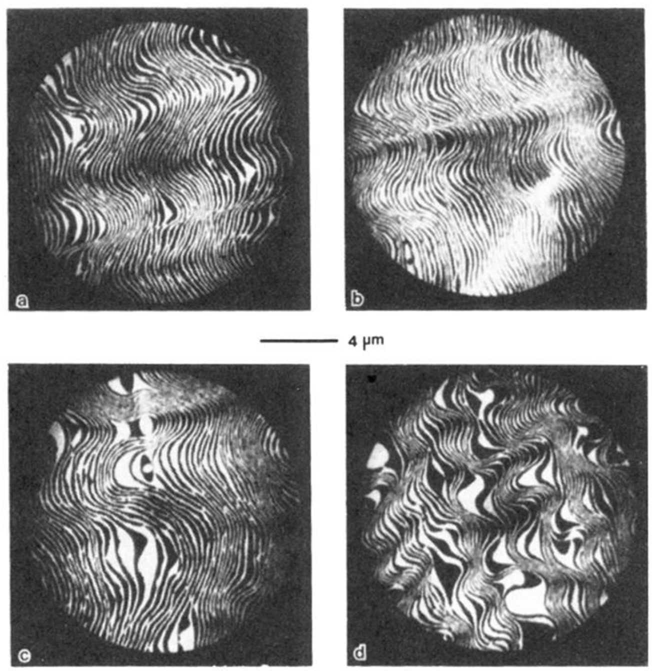

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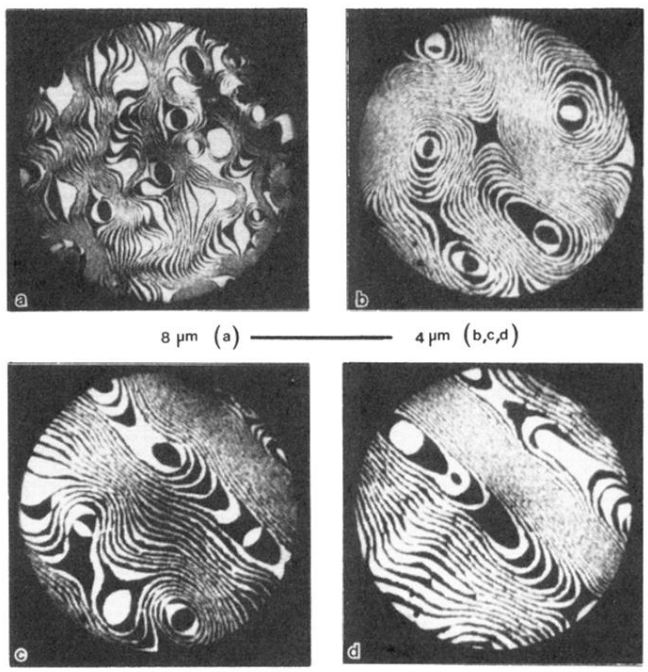

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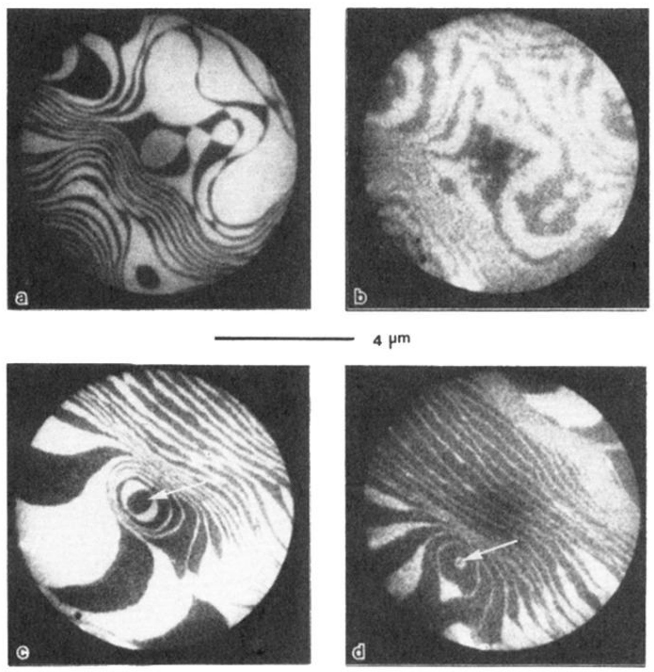

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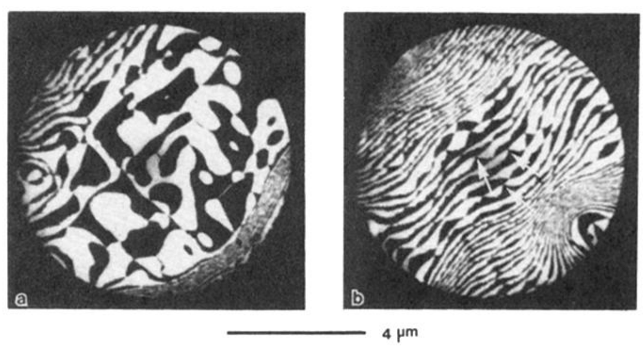

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