Wavy Steps on Si(001)

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Using low-energy electron microscopy we have found a new phase transition on the Si(001) surface at miscut angles smaller than $-0.1°$. The surface phase separates into facets with ~ 300 Å terrace width, and regions with much larger, wavy terraces. This wavy phase is stabilized by a reduction of surface-stress-induced strain energy. A theoretical study by Tersoff and Pehlke compares favorably with our observations.

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The step structure of Si(001) surfaces intentionally miscut towards (111) has been the subject of many recent studies [1–7]. This lively interest was sparked by experimental observations of a double-step predominance at larger miscut angles, with a phase transition to single-height steps at smaller miscut angles. Much less work has been done on samples cut very close to the (001) direction. Diffraction techniques and scanning tunneling microscopy (STM), used with much success on the vicinal surfaces, are not very suitable for small miscuts, due to limited coherence length in the case of diffraction, and due to the limited field of view in STM. In this study we have used low-energy electron microscopy (LEEM) to show that in addition to the double–single-height phase transition at large miscut angles, the surface undergoes another phase transition at very small miscut angles, from straight single-height steps to a coexistence of uniquely spaced straight steps and large-terrace-width wavy steps. Step waviness allows for alternation of (1×2) and (2×1) domains not only normal to, but also parallel with, the global step edges, resulting in a reduction of surface-stress-induced strain energy. These observations are in good agreement with a theoretical study by Tersoff and Pehlke [8].

The Si(001) surface exhibits a (2×1) dimer reconstruction. At a single-height atomic step the dimer orientation rotates 90° from (2×1) to (1×2) or vice versa. Experiments [9] and calculations [10] have shown that the surface stress tensor is anisotropic; tensile along the dimer direction, and compressive normal to the dimer. At small miscut angles an alternation of (2×1) and (1×2) domains separated by single-height atomic steps is favorable as it reduces the net stress in the surface. Because of the strong repulsion of S_A steps (dimers on the upper terrace normal to the step edge) and S_B steps (dimers parallel with the step edge) at close step separation [11], double-height steps become predominant at large miscut angles, although the energy gain is reduced by buildup of a net surface stress. The temperature-miscut phase diagram has been calculated theoretically [6,7], and appears to be in general agreement with experimental results. One of the unresolved issues, however, is the prediction that surface stress will be reduced at very small miscut angles by spontaneous generation of steps, in excess of those imposed by the miscut [5]. Such an effect has not been observed experimentally, and based on our observations we argue that this prediction results from the assumption of straight steps, and that step waviness is an effective way to reduce stress at small miscut angles, without excess step generation.

We have used low-energy electron microscopy to image the clean Si(001) surface with relatively large fields of view (4–10 μm). LEEM was developed in recent years by Bauer and Telieps [12]. In our microscope [13] a 15-keV electron beam is focused in the back focal plane of a magnetic cathode-type objective lens. The sample potential is close to the source potential. Thus, the electrons are decelerated between objective lens and sample (a gap of 2 mm) to an energy of a few eV. At this energy the electrons undergo conventional low-energy electron diffraction (LEED), and are subsequently accelerated back into the objective lens. A focused LEED pattern is formed in the back focal plane. A Gaussian image at magnification 20× is formed at a distance of 30 cm. An aperture is used in a conjugate diffraction plane to select an appropriate diffracted beam to form the final image at magnification 4000× to 10000× onto a 40-mm channel plate intensified phosphor screen. The images shown in this paper were formed using the $(\frac{1}{2},0)$ LEED beam at -3.5 eV energy. Terraces separated by single-height atomic steps alternate from black to white due to a 90° rotation of the dimer reconstruction at the step edges.

The Si(001) surface was prepared by repeated flashing to 1250 °C to desorb the native oxide and carbon impurities. Samples (n type, 2 mΩ cm) are heated by electron bombardment from behind, and show sharp and intense two-domain LEED patterns. Figure 1 shows four representative images obtained on one sample. Surface height increases from left to right. The field of view in (a)–(c) is 8 μm, and 4 μm in (d). Clearly, the steps are not straight, or even approximately straight. This is in sharp contrast with STM images obtained on samples with much larger miscuts, in which S_A steps are very straight, and the S_B steps undergo *random* position fluctuations. Image (a) shows groups of steps oscillating together. The oscillations are not coherent over very large distances, and are separated by larger terraces. The average terrace width is in the 1000–2000-Å range. Image (b) shows another area, qualitatively similar to (a). In image (c) we point out two so-called "kissing" sites [14],

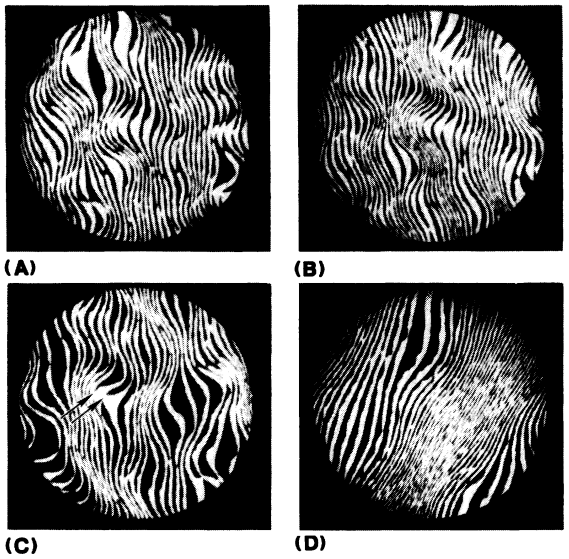

FIG. 1. Representative images of Si(001) with ~0.1° miscut. (a),(b) Two different, but similar regions on the sample. (c) A region with a few "kissing" sites (arrow). (d) A faceted area with ~300 Å terrace widths. Field of view in (a)–(c) is 8 μm; in (d), 4 μm.

related to reconstruction domain boundaries on the same terrace (arrows). In these images the terrace widths are not very uniform. This is unlike the area with tightly spaced steps seen in image (d). Here, the step spacing is quite uniform (about 300 Å) and the steps are quite straight. This bunched-step area may be viewed as a narrow facet, coexisting with the flatter areas with larger step spacing. Remarkably, we have observed similar facets, with the same average step spacing of about 300 Å, at various places on the sample. The facets always extend over large distances parallel to the step edges, in contrast with the wavy regions shown in (a)–(c) which usually do not extend coherently over a large distance. We note that the step waviness seen in (a)–(c) is very different from the step morphology observed near SiC particles and cannot be explained by such impurities. Indeed, no such particles are seen in Fig. 1.

While the images in Fig. 1 were obtained at room temperature, the surface steps reach their final configuration at much higher temperature, during cooldown after cleaning. To better understand the high-temperature starting phase of the surface we have obtained images in the temperature range of 1100–1200 °C. Figure 2 shows the most important results. The area shown was selected for its large step separation to make the effects more clearly visible. Image (a) was taken at 1110 °C. During real-time observations we see evaporation by step flow; the steps recede in the direction of the arrows, and are well defined at all times. The situation is different at 1130 °C, image (b). Now evaporation no longer proceeds by simple step flow. Instead, single-height holes form on

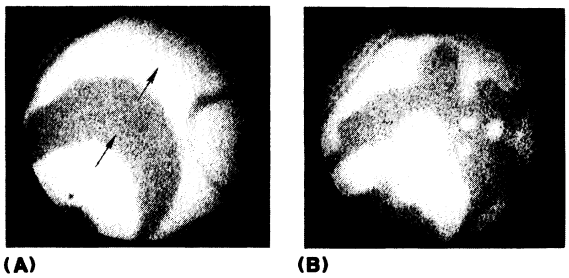

FIG. 2. Images of Si(001) at (a) 1110 °C and (b) 1130 °C. Because of evaporation the steps move in the direction of the arrow. The steps are well defined in (a) but start to dissolve in (b). At 1150 °C steps are no longer visible, and contrast disappears completely due to surface roughness. Field of view is 4 μm.

the terraces, and then grow and run into other holes or remnants of steps. This process is very dynamic and chaotic. At 1150 °C all contrast disappears. Evaporation occurs now so rapidly all over the surface that holes no longer have time to ripen and grow, and the surface becomes rough. At this stage steps are no longer well defined and certainly not well localized [15]. During surface cleaning we reverse this process, starting with a rough phase in which steps are not well defined, cooling through step flow, and finally arriving at the patterns shown Fig. 1.

Here we compare our results with a recent theoretical study by Tersoff and Pehlke (TP) [8], in which the effect of sinusoidal step oscillations on surface energy is analyzed. For terrace lengths (L) exceeding ~600 Å, TP find that such oscillations are stable with respect to straight steps over a very wide range of amplitude (A) and wavelength (λ). In our images we typically see $A/L = 2$–4, $L = 1000$–2000 Å, $\lambda = 1$–2 μm, and $A/\lambda = 0.5$–1. This is well within the stability region, but not close to the *minimum*-energy structures found in the calculations, which would correspond to $A/\lambda = 4$–6 for $L = 1000$–2000 Å. There are three possible explanations for this apparent discrepancy. First, the numerical values for step energies and stress anisotropies that enter the theory may be insufficiently accurate. This would affect the quantitative predictions, but not the qualitative conclusions. Second, the theory calculates energies of perfectly coherent, defect-free oscillations. The images show that the real surface is not perfectly coherent, and not defect free (kissing sites [14] being the predominant defect). Third, there is no guarantee that the surface reaches thermodynamic equilibrium (even in the absence of surface defects), considering the high-temperature surface morphology from which the sample is cooled. In addition, the theoretical results indicate that the driving force to approach equilibrium from a small A/λ ratio is relatively small.

As mentioned, the frequent occurrence of kissing sites

is likely to hinder equilibrium of the step morphology. Two such sites are indicated by arrows in Fig. 1(c). A given terrace may have two regions of (2×1) [or (1×2)] reconstruction, separated by an in-plane domain boundary. Such domain boundaries may form during cooldown as the reconstruction nucleates simultaneously on different regions of a terrace. Alternatively, we have observed the formation of domain boundaries as screw dislocations move through the bulk, extending to the surface, at high temperature. In either case, the domain boundaries have high energy, which can be lowered by covering them with a protrusion from the next terrace level, forming a local double step at the kissing site. The step undulation at the kissing site is similar to the sinusoidal undulations occurring without kissing sites, but extrinsic in nature. As a result of its effect on the local surface stress, a kissing site will place a restriction on the step undulations in its vicinity, which in effect get "locked" to the kissing site undulation.

We find a pronounced tendency to form facets consisting of straight steps, with a terrace width of $\sim$300 Å, coexisting with large regions of wide, wavy terraces. This phase separation is indicative of two distinct regions of thermodynamic stability, one at terrace widths of $\sim$300 Å, and another for very large terrace widths, and is a clear signature for a first-order phase transition from a straight step phase to a wavy step phase. TP's calculations show that the surface energy has two distinct minima with respect to L, one for straight steps at $L=200$ -300 Å (somewhat depending on the choice of input parameters), and another region with only slightly higher energy for L in the range of a few 1000 Å (wavy steps), in excellent agreement with our experimental observations.

While the kinetic limitations during cooldown and the occurrence of kissing sites limit the degree of quantitative agreement to be expected between experiment and theory, the qualitative agreement is very satisfactory. Theory predicts strong step oscillations for terrace widths exceeding 600 Å, as seen in Fig. 1. In addition, a phase separation is expected, with facets of average terrace width $L=200$–300 Å coexisting with terraces larger than $\sim$1000 Å. This again is in excellent agreement with our observations. The amplitude of the step oscillations appears to be larger than the lowest-energy oscillations predicted in theory, but the structures observed are well within the stability regions calculated by TP. Inside these regions (and particularly for large wavelengths) the driving force toward thermodynamic equilibrium is rather small. Our studies at high temperature show that the surface starts in a very rough phase from which the sample tries to approach thermodynamic equilibrium. Spontaneous step formation as predicted in earlier theoretical studies (assuming straight steps) has not been observed in these experiments. Step-flow evaporation prevents the formation of such extra steps, and the surface can more effectively reduce stress by step oscillations. Interestingly, at both large and small miscut angles the surface morphology is determined by a competition of step energies and surface stress. At large miscut angles there is the additional competition between single- and double-height steps. Over the very large range of terrace widths now studied (10 to $\sim$5000 Å), two distinct phase transitions have now been observed: from double-height steps to straight single-height steps at large miscuts, and then from straight single-height steps to a phase coexistence of uniquely spaced straight single-height steps and wavy steps as observed for the first time in this study.

The authors have enjoyed many enlightening discussions with Jerry Tersoff and Eckhard Pehlke.

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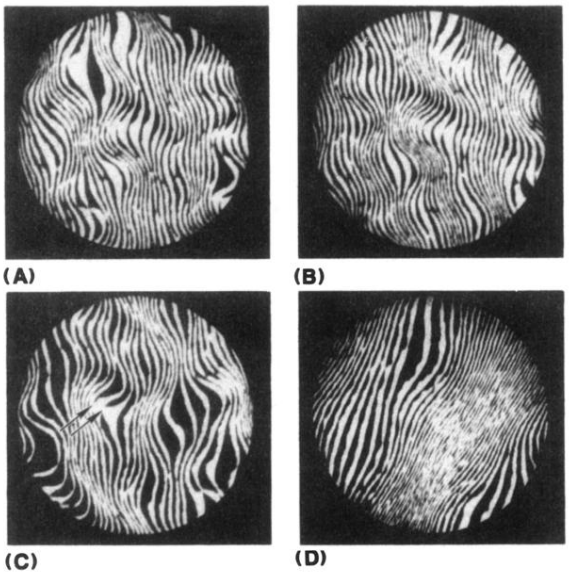
[15] The picture that emerges is very similar to that of growth, with increasing flux rates. At low flux rate the surface grows by step flow. At higher flux rate step flow combines with 2D island nucleation on the terraces. At still higher flux rate the mobility is too low even for island nucleation, and the surface becomes rough. In analogy, increasing the temperature during evaporation results in an increasing *negative* flux rate, or rate of surface vacancy formation. The phenomena in Fig. 2 also appear similar to the kinetic roughening transition recently discussed by D. E. Wolf, Phys. Rev. Lett. **67**, 1783 (1991).



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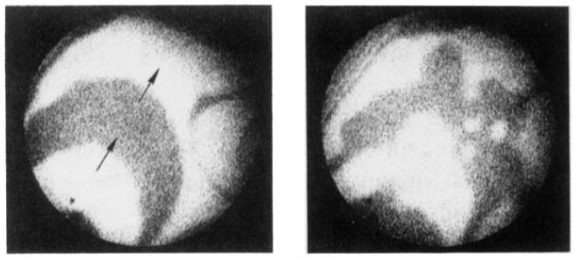

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