

Step Fluctuations on Vicinal Si(113)

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(Received 4 March 1998)

The fluctuation properties of steps relevant to step coalescence have been measured using scanning tunneling microscopy on a Si(113) surface miscut along a low symmetry azimuth. In local thermal equilibrium at 710 °C, coexistence of single, double, triple, and quadruple steps has been observed. From direct measurement of the step-correlation function, the step stiffness is shown to be proportional to the step height, with an additional stabilization of double-height steps. This result is shown to require a step-step attraction energy close to kT . [S0031-9007(98)06237-1]

PACS numbers: 68.35.Md, 61.16.Ch, 68.35.Bs, 68.60.Dv

Understanding of the role of steps and surface curvature in surface mass transport [1,2] is the key to descriptions of growth and structural stability in the fabrication of nanomaterials. Relatively recently, quantitative connections between the early kinetic and thermodynamic descriptions of step behavior have been developed [3]. This advance offers the promise of an effective parameterization scheme for material evolution at surfaces and interfaces [4,5]. To date, the step-based approach has proven extremely powerful in describing mass transport in the decay of metastable structures and in the description of reversible faceting transitions in which behavior is governed by the repulsive interactions between steps. In these systems, the surface can be described as composed of discrete steps. A major extension of the theoretical approach is required for the important problem of the formation of faceted structures via step motion. In this paper, we investigate quantitatively the fluctuation characteristics of interacting steps which are required to bind into well-defined facet structures.

The vicinal surfaces of Si(113) are a model system for studying the evolution of steps into stable facets. For this system, Song and co-workers discovered faceting transformations involving step-step attractions using surface x-ray scattering [6]. Because there is no physical basis for expecting a long-range attractive interaction between steps, this work has inspired theoretical interest [7,8]. Recently, Lässig [8] has predicted that a long-range repulsion and a short-range attraction conspire to produce the complicate orientational phase diagram as shown by Song and co-workers. Song and co-workers [9,10] have adapted Lässig's concept to understand their observations on the kinetics of faceting, where they have found that the characteristic length scale, for instance, the average terrace width, increases as a power law versus time with a coarsening exponent $\frac{1}{6}$ [9,10]. To explain the $\frac{1}{6}$ power law, they had to require that as steps bind together the step stiffness increases linearly with the number of steps and

the step mobility decreases linearly with the number of steps. In this paper we have determined experimentally the behavior of step groups of variable size to test these assumptions.

To quantify the step stiffness, we measure correlation functions directly using scanning tunneling microscopy (STM). The experimental design uses formalisms demonstrated previously for STM [11], reflection electron microscopy [12], and low-energy electron microscopy [13]. We show that for steps with heights from single layer up to quadruple layers the step stiffness is proportional to the number of steps in the step bunch, with an additional increase in stability for double-layer steps. This result is compared with microscopic models to understand the nature of the step-binding energy.

The experiment was conducted in a ultrahigh vacuum (UHV) chamber with base pressure of 3.0×10^{-8} Pa, equipped with a variable-temperature STM. Samples were cut from a *n*-type Si(113) wafer miscut by 1.7° towards a low symmetry azimuth which is 33° away from the $[\bar{1}10]$ to the $[\bar{3}\bar{3}2]$. The vicinality was measured by Laue x-ray diffraction in agreement with the STM images, which showed that the average step edge direction is 35° away from the $[\bar{1}10]$ direction. To obtain a clean silicon surface, the samples were flashed at 1200 °C for one minute by a direct current through a sample. The temperature was monitored by an optical pyrometer. Structure was measured at room temperature after quenching at a cooling rate of 200 °C/s, which is achieved by cutting off the heating current.

To find conditions for the preparation of equilibrated steps of variable height, we compared surfaces quenched after annealing at various temperatures in the range 600–1000 °C. When the sample is annealed below 690 °C, the surface breaks up into a hill and valley structure composed of (113) facets and densely stepped regions. The period of the structure increases with increasing annealing time following the $\frac{1}{6}$ power law [14].

Annealing above the transition temperature of 720 °C always results in a mixture of single steps (1.63 Å), double steps, and triple steps, indicating that the quenching rate is not fast enough to retain the high-temperature equilibrium structure which contains only single-layer high steps. Following annealing at 690–720 °C, the average terrace width becomes slightly larger than that of the surface annealed above 720 °C, because of the existence of steps with more than triple-layer height, as shown in Fig. 1. The size of the (113) terraces saturates during annealing within 5 min, implying that the surface reaches a local thermal equilibrium. Thus, at this temperature regime, equilibrium step configurations can be frozen by the quenching.

In Fig. 2, high resolution STM images of the step edge with various step heights are displayed. The crystallographic axes shown in Fig. 2 were identified by the (3×2) surface reconstruction [15]. The single steps [Fig. 2(a)] and the double steps [Fig. 2(b)] have sawtooth structures composed of step edges along the $[\bar{1}10]$ and $[0\bar{3}1]$ directions, which accommodate the low-symmetry orientation of the steps. The double steps frequently have sharp edges, suggesting that the double step is stable and most likely forms with a well-defined bonding configuration at the step edge. On the other hand, the triple [Fig. 2(c)] and quadruple steps [Fig. 2(d)] have more complicated and rough structures due to splitting into single and double steps at the step edge.

To quantify the fluctuation properties of the steps, we set up a $x - y$ coordinate system with the y direction taken to be the mean running direction of the step. After

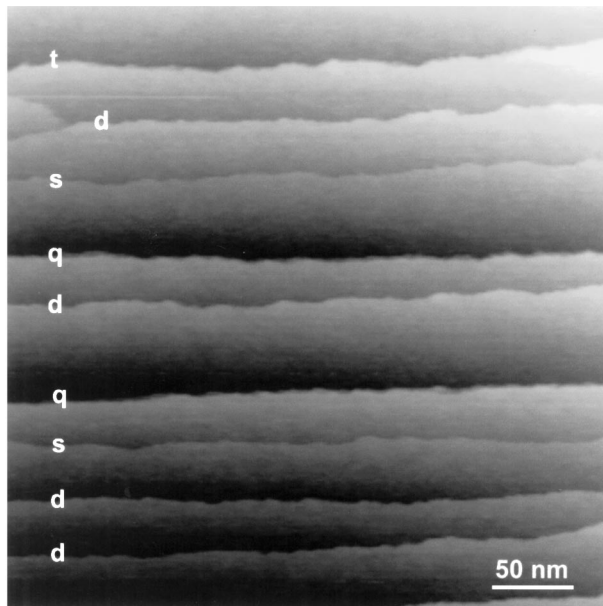


FIG. 1. A 360 nm \times 360 nm STM image of the Si(113) surface quenched after annealing at 710 °C for 10 min. Single-, double-, triple-, and quadruple-layer steps are marked as s , d , t , and q , respectively.

coarse graining, each of the steps is treated as a string with energy, $E = \tilde{\beta}/2 \int [\partial x(y)/\partial y]^2 dy$, where $x(y)$ is the coarse-grained step configuration and $\tilde{\beta}$ is the step stiffness. In general, the fluctuations of the steps can be characterized by the mean-square displacement as a function of distance y : $g(y) = \langle [x(y) - x(0)]^2 \rangle$. If the step is isolated, the step edge can be considered as a random walker, and therefore $g(y)$ increases linearly as $g(y) = b^2 \cos^3 \theta y$, where b^2 is the diffusivity and θ is the angle between the step edge and the high-symmetry direction, in this case approximately 35°. Using the equipartition law, we easily find that the diffusivity is related to the stiffness as $\tilde{\beta} = kT/b^2 \cos^3 \theta$ [16]. Thus the stiffness can be determined directly from the measured slope of the correlation function versus distance, $b^2 \cos^3 \theta$. If the area of the step bunch is small enough to be negligible compared to that of the terrace, the above formalism can be simply extended to step bunches of N layers, which will be characterized by their own values of the diffusivity and the stiffness b_N^2 and $\tilde{\beta}_N$.

In Fig. 3, the measured mean-square displacements $g(y)$ for the steps with different heights are plotted as a function of y . At small enough y compared to the length of the analyzed step of 650 nm, the linear behavior of $g(y)$ is confirmed. We find that the slopes of the correlation functions are 0.15 ± 0.01 for single steps, 0.100 ± 0.005 for double steps, 0.052 ± 0.001 for triple steps, and 0.040 ± 0.005 nm for quadruple steps. The length scale on which the effect of the step collisions is not negligible is roughly estimated by the collision distance y_{coll} : $y_{\text{coll}} = \bar{l}^2/4b^2 \cos^3 \theta$, where \bar{l} is the average distance between neighboring steps [16]. Using $\bar{l} = 35$ nm and $b^2 \cos^3 \theta = 0.15$ nm for single steps, we find that the collision distance is $\sim 2 \mu\text{m}$. The range of the linear dependence seen in Fig. 3 is well within this limit. We also found that the linear dependence of $g(y)$ breaks down for larger y , in qualitative agreement with theories [16] which predict that the step-step collisions change the linear behavior into a logarithmic form, though we could not determine the exact functional form at large

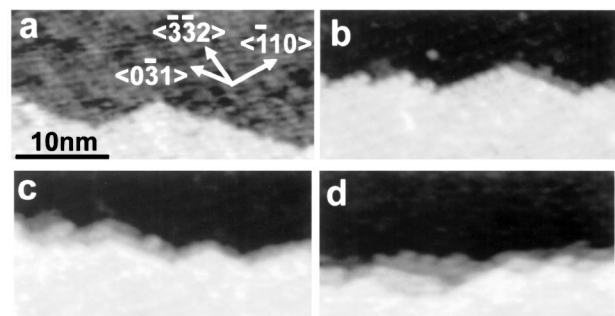


FIG. 2. High resolution 32 nm \times 16 nm STM images of the step structure of (a) a single step (1.63 Å), (b) a double step, (c) a triple step, and (d) a quadruple step.

y . Note that the offsets of $g(y)$ for multilayer height steps in Fig. 3 originate from uncertainties in the measurement of the step position due to the finite width of the step edge (as seen in Fig. 2). The short wavelength fluctuations responsible for the width of step edge do not affect the scaling features of $g(y)$.

From the diffusivity deduced from Fig. 3, we estimate the step stiffnesses of the single steps, the double steps, the triple steps, and the quadruple steps at 710 °C to be $\tilde{\beta}_1 = 57$, $\tilde{\beta}_2 = 85$, $\tilde{\beta}_3 = 163$, and $\tilde{\beta}_4 = 220$ meV/Å, respectively. The step stiffness increases linearly with the step height as shown in Fig. 4, implying that bending the higher step bunches costs more energy than bending the lower ones. The slightly lower stiffness of the double-layer step than $2\tilde{\beta}_1$ suggests a special bonding configuration, consistent with the sharp structure of the double-layer step revealed in Fig. 2.

We can obtain a qualitative understanding of the dependence of step stiffness on step height in terms of a terrace-step-kink (TSK) model, in which steps wander due to thermal excitation of kinks. On a rectangular lattice with lattice constants a_y and a_x , consider a group of N single steps with a short-range step-step attraction whose Hamiltonian is

$$H = \sum_{i=1}^N \sum_{y=1}^L \epsilon |n_i(y)| - \sum_{i=1}^{N-1} \sum_{y=1}^L \epsilon_a \delta_{x_i(y), x_{i+1}(y)}, \quad (1)$$

where ϵ is the kink energy, which governs the thermal excitations and thus the stiffness of the steps, ϵ_a is the short-range attractive interaction, which acts only between elements of adjacent steps that physically intersect [$x_i(y) - x_{i+1}(y) = 0$], and the depth of a kink at each position of a step edge y is $n_i = [x_i(y) - x_i(y +$

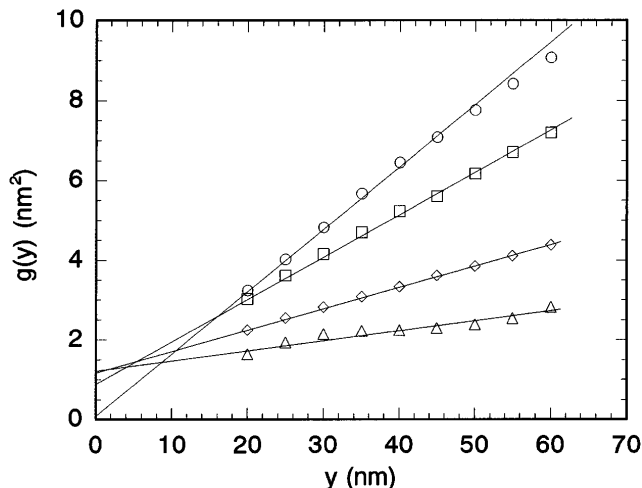


FIG. 3. The mean-square displacements are plotted versus the separation distance parallel to the mean step edge y for circles—single-height steps; squares—double-height steps, diamonds—triple-height steps; and triangles—quadruple-height steps. Each is an average of individual data measured for 5–12 steps.

)]/ a_x . This model has previously been shown to reproduce the characteristic kinetics of step coalescence on Si(113) [14]. As well as the stiffness of a single step, that of a group of N single steps can be defined as $\tilde{\beta}_N(T) = a_y kT / b_N^2(T)$ via the diffusivity of the average position of the component steps, $b_N^2(T) = a_x^2 \langle \bar{n}^2 \rangle$, where $\bar{n} = \sum_{i=1}^N n_i(y) / N$. At low temperatures, a bunched step which consists of N single steps acts as a single step with a kink energy $N\epsilon$. Since the step stiffness of a single step is given by $\tilde{\beta}(T) = 2(a_y/a_x^2)kT \sinh^2(\epsilon/2kT)$ [16], the dependence of the step stiffness on the step height results in

$$\tilde{\beta}_N = 2 \frac{a_y}{a_x^2} kT \sinh^2 \frac{N\epsilon}{2kT}. \quad (2)$$

At high temperatures, we are no longer able to treat a step bunch as a single step with kink energy of $N\epsilon$ because not only kinks but also unbinding of steps is partially excited. In the high-temperature limit, the interactions between steps disappear and the length of kinks, na_x , becomes independent, therefore, $b_N^2(T) = a_x^2 \langle n^2 \rangle / N$. Thus, as the form in the high-temperature limit, we find that

$$\tilde{\beta}_N = N\tilde{\beta}_1. \quad (3)$$

To quantify the temperature range over which step unbinding becomes important, we performed Monte Carlo calculations of $\tilde{\beta}_N$ for various T . In all of the calculations, we set $\epsilon = \epsilon_a$. In this case, the faceting temperature, at which a step bunch breaks up, is $kT/\epsilon \sim 1.15$. Figure 5 shows the computed step stiffnesses. Deviations from the form of Eq. (2) become pronounced when the simulated step structures begin to show significant step unbinding at about $kT = \epsilon_a$. The dependence changes to the linear behavior of Eq. (3) near the faceting temperature. Thus, the observed approximately linear dependence

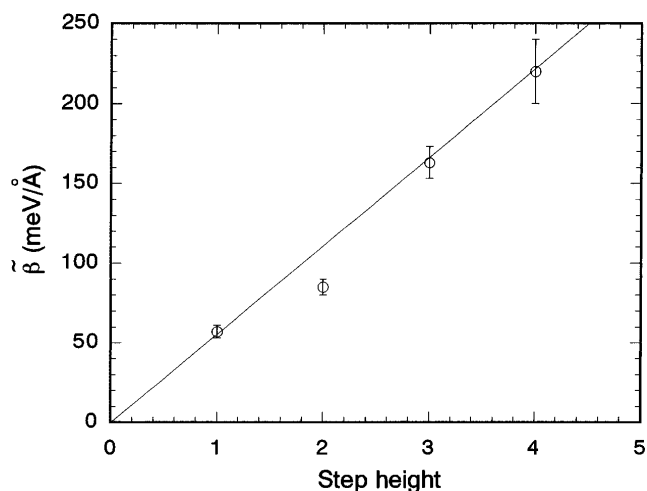


FIG. 4. The step height dependence of the step stiffness, calculated from the slopes shown in Fig. 3 assuming an equilibration temperature of 710 °C. Step heights are measured in units of single layer height (1.63 Å).

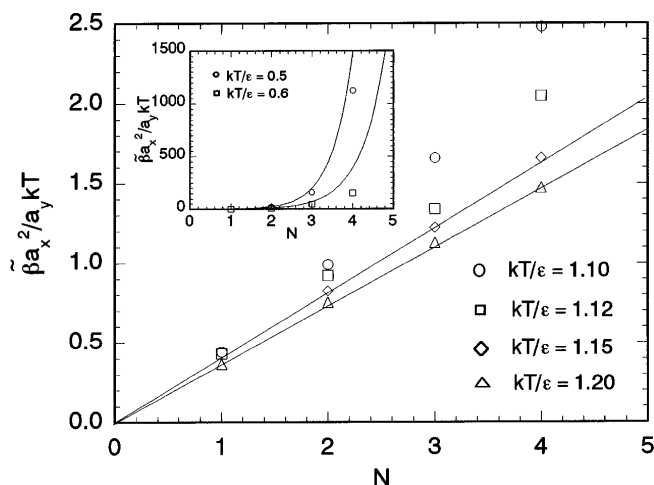


FIG. 5. The step height dependence of $\tilde{\beta}$ for the modified TSK model, whose Hamiltonian is given by Eq. (1), for temperatures near T_f . The solid lines show $\tilde{\beta}$ calculated from Eq. (3). The inset shows the step height dependence of $\tilde{\beta}$ at low temperatures. The solid lines show $\tilde{\beta}$ calculated from Eq. (2). In all of the calculations, the ratio of ϵ_a to ϵ is 1:1.

of the step stiffness on the step height at 710 °C, which is near T_f , qualitatively agrees with the prediction for high temperatures in the modified TSK model of Eq. (1). Figure 2 indeed shows that the steps are partially unbound except for the double steps.

These results provide a physical basis for understanding Song *et al.*'s observation of a $\frac{1}{6}$ power law dependence of average terrace width versus time during faceting. To explain this result, their model requires that the step stiffness scales with height as $\tilde{\beta}_N = N\tilde{\beta}_1$. Our results show that this is an appropriate physical constraint only near the faceting temperature where step unbinding becomes facile. The special stability of the double-height steps will not comprise this conclusion because formation of the double steps occurs in the early stage of the faceting. The absolute magnitudes of the step stiffness that we have measured can, in principle, be used to develop a quantitative description of the energetics of excitations at the step edges [17]. Previously, a much lower step stiffness of a single step on Si(113), 0.72 meV/Å was estimated at 920 °C [18] for a step in the high-symmetry direction. Our results are not inconsistent with this, since the step stiffness decreases rapidly just below the roughening temperature. For comparison, we note that the roughening temperature of the Si(001) is ~ 1200 °C [13,17]. In addition, a higher value of the step diffusivity, 0.82 nm, was measured for single steps oriented orthogonal to the high-symmetry direction [19] following a quench from high temperature. The higher diffusivity implies that the kink excitation energy at the step edge along the $[3\bar{3}2]$ direction is lower than at the step edge along the $[\bar{1}10]$ direction.

In conclusion, we have studied the fluctuation properties of steps on the Si(113) surface miscut along a low symmetry azimuth as they relate to the binding of steps to form stable facets. We demonstrated that, in thermal equilibrium at 710 °C, single, double, triple, and quadruple steps are coexistent. We have shown that, except for the double steps, the step stiffness is proportional to the number of steps in the step bunch. By comparison with a TSK simulation, we conclude that this result can occur only for step bunches very close to their unbinding transition temperature.

This work was supported by a Grant-in-Aid for Scientific Research Project on Priority Area "Single Electron Devices and Their High Density Integration" from the Ministry of Education, Science, Sports and Culture, Japan (No. 08247214). One of us (E.D.W.) also acknowledges support from the University of Maryland NSF-MRSEC (DMR-96-324521). We acknowledge useful discussions with Professor T. L. Einstein, Professor J. D. Weeks, and Professor N. Akutsu.

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