

Terrace-Width Distributions on Vicinal Si(111)

X.-S. Wang, J. L. Goldberg, N. C. Bartelt, T. L. Einstein, and Ellen D. Williams

Department of Physics, University of Maryland, College Park, Maryland 20742-4111

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Using scanning tunneling microscopy, we have quantitatively characterized the configurations of steps on vicinal Si(111) surfaces misoriented by 1.2° and 2.3° towards the $[\bar{1}\bar{1}2]$ direction. The measured terrace-width distributions are strongly peaked, consistent with predictions for thermally wandering steps. However, the distributions are much narrower than predicted for the simple terrace-step-kink model, indicating that the steps interact with energetic short-range repulsions. The magnitude of this energetic repulsion is gauged from a Gaussian fit to the data. The width of the distribution scales with step density as expected for repulsions which decay as the inverse square of step separation.

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A fundamental question in surface science is how surface structure is related to surface energetics. An important aspect of this question is what determines the overall morphology of a surface, e.g., what determines whether a surface is faceted or rough. Under equilibrium conditions, this is a question of surface thermodynamics, which is determined by the orientational dependence of the surface free energy.^{1,2} As has long been recognized,¹ this orientational variation depends crucially on the energies of single steps and their interactions. Recently, much progress has been made in understanding the statistical mechanics of stepped surfaces through the consideration of simple models.^{3,4} To assess the applicability of these models, there have been beautiful measurements of equilibrium crystal shapes,^{2,5} which are again directly determined by the orientational dependence of the surface free energy. There have also been a large number of diffraction measurements of the temperature (T) dependence of surface morphology.⁶ In this Letter we present a different approach. Using a scanning tunneling microscope (STM),⁷ we have directly imaged, with atomic resolution, over a thousand steps on vicinal Si(111) surfaces. By analysis of the statistics of these step configurations, in particular by determining $P(L)$, the distribution of terraces of width L , we have drawn important conclusions about the interactions between steps.

In deducing the effects of interactions between steps from $P(L)$, it is important to include correctly the role of step wandering. In configurations with closely spaced steps, the steps can wander less than in configurations with wide separations, so the former contribute less to the entropy.⁸ With this entropic consideration, small L 's become unfavorable, giving rise to an effective "entropic repulsion."³ We find that $P(L)$'s measured on Si(111) are consistent with freely wandering steps suffering such entropic repulsion, with additional short-range energetic repulsions. By studying the dependence of $P(L)$ on mean terrace size, i.e., net surface misorientation, we are able to demonstrate that $P(L)$ varies with misorientation angle in a manner consistent with $1/L^2$ step-step interac-

tions, the form expected for elastic or electric-dipole-induced interactions.⁴ In previous studies on stepped Si(100) surfaces, which are expected⁹ to have a different form of step interactions due to the anisotropy of the (2×1) reconstruction, evidence for direct energetic repulsions has also been observed.^{10,11} Drawing from recent statistical-mechanical theories,^{12,13} we are the first to focus on the details of $P(L)$ as a key to thorough understanding of interactions between steps, and on the orientational dependence of $P(L)$ as a way to demonstrate the form of the energetic repulsions between steps. By varying sample preparation conditions, we can monitor the importance of nonequilibrium effects on surface morphology: We find that the distributions do depend on sample cooling rates.

We have studied Si(111) misoriented by 1.2° and 2.3° towards the high symmetry $[\bar{1}\bar{1}2]$ direction. On these surfaces the low- T equilibrium structure on the terraces is (7×7) reconstructed. The (7×7) unit cell is diamond shaped and can be described as two equilateral triangles of side length 27 \AA placed base to base. The $[\bar{1}\bar{1}2]$ direction lies along the perpendicular bisector of these triangles (bisector length = 23 \AA). It has been shown in STM studies that the reconstruction continues up to the step edges.¹⁴ Previous low-energy electron-diffraction (LEED) results on 6° and 12° misorientations, where the average single-layer height step-step separations are 30 and 15 \AA , respectively, have shown that the step structure at low T is not simple: Phaneuf and Williams¹⁵ conclude that these surfaces contain triple-layer height steps (possibly mixed with single-layer height steps). However, as discussed below, on the 1.2° - and 2.3° -misoriented surfaces, where the average single-layer height step-step separations were measured to be 146 \AA (~ 6.3 bisector lengths) and 79 \AA (~ 3.4 bisector lengths), respectively, the ratio of triple-layer height to single-layer height steps is small and decreases with annealing, suggesting that triple-layer steps are nonequilibrium features¹⁶ for small misorientation angles.

The samples used were n -type Si(111) wafers ($7 \text{ m}\Omega \text{ cm}$). Each sample was cleaned via the standard pro-

cedure^{15,17} of a 1250°C anneal for 1 min with the background pressure below 1×10^{-9} Torr, followed by quick cooling to 900°C, then slow cooling ($\leq 0.05^\circ\text{C}/\text{sec}$) to room temperature. Parallel high-resolution LEED measurements in a separate chamber showed that at any faster cooling one obtains a surface which is obviously out of equilibrium as marked by a diffuse diffraction pattern. The rate of cooling through the $(1 \times 1) \rightarrow (7 \times 7)$ phase transition at $\sim 850^\circ\text{C}$, where changes in step structure occur,^{15,16} is most crucial. Also examined with STM were samples prepared by quick cooling (faster than $100^\circ\text{C}/\text{sec}$) and others annealed for up to 20 h at $\sim 700^\circ\text{C}$ following slow cooling through the phase transition.

After allowing a few hours for the system to cool following cleaning, samples were transferred to the STM. In order to compile a statistically significant number of steps, we scanned macroscopically distant areas on each sample in addition to imaging different surfaces of the same orientation and treatment. This procedure requires several important features of the STM and its environment.¹⁸ To measure terrace sizes over large regions of the surface, the STM must have the stability to take repeated large-area ($\sim 1000 \text{ \AA} \times 500 \text{ \AA}$) atomically resolved images. The configuration of our STM allows us to scan macroscopically distant regions by moving the sample or by changing tips. A final requirement is the ability to maintain a clean sample. For the (111) surface, we find that a base pressure below 1×10^{-10} Torr maintains the integrity of the STM images for up to 4 days after the initial cleaning. All images were taken in constant-current mode at a -2.0-V tip bias with 1.2-nA tunneling current.

A typical image from a 1.2° sample annealed for approximately 20 h at $\sim 700^\circ\text{C}$ following the normal cleaning procedure is shown in Fig. 1. Although slowly

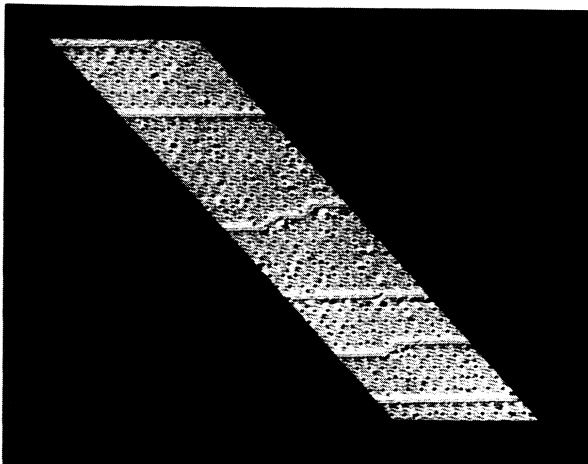


FIG. 1. Scanning-tunneling-microscope image of a $\sim 800 \text{ \AA} \times 200 \text{ \AA}$ region of a stepped Si surface misoriented by 1.2° towards the $[\bar{1}\bar{1}2]$ direction. Notice that the kink spans one (7×7) unit cell.

cooled 1.2° samples which did not receive any post-cleaning treatment had a triple- to single-height step ratio of about 1:5, the long annealing reduced this ratio to less than 1:12. On the 2.3° surface it is quite difficult to reduce this ratio below 1:6. Both surfaces have a relatively low density of kinks at step edges, such as the one seen in Fig. 1. Such kinks appear irregularly every few hundred angstroms. Notice that the kinks follow the corner holes of the (7×7) reconstruction.¹⁶

The question of equilibrium is important in interpreting the observed step structure. We noted above the significant changes in step structure (i.e., the density of triple-height steps) following annealing at 700°C for many hours. After 20 h, LEED and STM show no further changes on the time scale of hours: The surfaces are as close to equilibrium as we can achieve. The amount of surface diffusion is the limiting factor in attaining equilibrium. From studies on $\text{Si}(111)$ misoriented towards the $[\bar{2}11]$ direction, we know that cooling rates of $0.2^\circ\text{C}/\text{sec}$ allow the surface to facet reversibly¹⁹ at temperatures between 700 and 850°C , depending on the angle of misorientation. Since the amount of mass transport required to move steps across our present surfaces dwarfs that needed for faceting, equilibration via mass transport must be possible at 700°C . It would be surprising, however, if the sample were in thermal equilibrium down to room temperature. Since noticeable step motion has been reported on laser-quenched Si surfaces at $T \approx 400^\circ\text{C}$,²⁰ our observed step configurations are probably representative of equilibrium at a T between 400 and 700°C .

Our data show that the (7×7) reconstruction is strongly correlated across steps on slowly cooled sur-

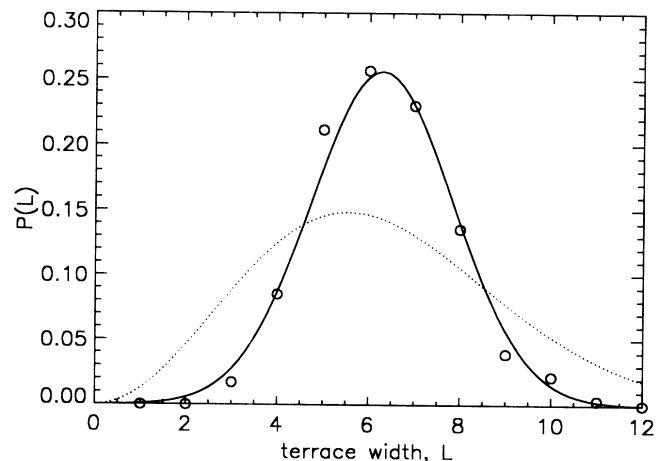


FIG. 2. Terrace-width distribution for an annealed 1.2° -misoriented sample. $P(L)$ is the probability of occurrence of terrace width L , in units of the (7×7) unit-cell dimension in the $[\bar{1}\bar{1}2]$ direction (23 \AA). Circles are data measured on a sample as close to equilibrium as we can obtain; the dotted curve is the distribution derived from the TSK model; the solid curve is the result of a least-squares fitted by the Gaussian form expected for strongly interacting steps.

faces; approximately 90% of single-height steps exhibit the same displacement of the (7×7) reconstruction across step edges.¹⁶ The widths of terraces bounded by two single-height steps, therefore, come in multiples of the distance between adjacent rows of corner holes, i.e., the 23-Å bisector distance which henceforth is our unit length. Measuring terrace widths on the slowly cooled, long-annealed 1.2° and 2.3° misorientations yields the terrace-width distributions $P(L)$ plotted as circles in Figs. 2 and 3. Terraces neighboring triple-height steps were not counted in these compilations. Observations from 151 to 482 terraces were used in obtaining Figs. 2 and 3, respectively. To obtain the 1.2° data, five distinct regions on two different samples were examined; for the 2.3° data, six regions on two samples were used. Data from each region were statistically consistent with the final $P(L)$; we estimate the statistical uncertainty in the distributions to be $\pm 10\%$ near the peaks.

To interpret these distributions, we first consider the simplest model of a stepped surface in equilibrium, the terrace-step-kink (TSK) model. It makes three basic assumptions: (1) The surface is in equilibrium; (2) the terraces are sufficiently large (i.e., the misorientation angle sufficiently small) so that there are no energetic interactions between steps; and (3) the only thermal excitations of the surface are kinks in the step edges, which cause the steps to wander. In deducing interactions between steps from $P(L)$, it is important to include the effect of this wandering as mentioned earlier.

In Fig. 2 the $P(L)$ recently computed^{12,13} for the TSK model is coplotted (dotted curve) with the experimental $P(L)$ (circles) from the 1.2° samples. The calculation relies on the analogy of steps with free fermions^{4,12,13} and thus assumes that the kink density is low, as observed: In this limit the distribution is independent of T

(and remains so to $\sim 80\%$ of the roughening T of the terraces¹²). Thus our previously mentioned lack of knowledge of the T characterizing the equilibration of the step structure does not hinder comparison with experiment. Notice the signature of entropic repulsions: Despite the assumed lack of energetic interactions between steps, there is a bias against small step separations. The model does not describe the data adequately: The experimental $P(L)$ is more sharply peaked and has fewer occurrences of small L than the theoretical model. For sufficiently large repulsive energetic interactions, $P(L)$ can be well approximated by a Gaussian,¹² as is the case apparently for our data, plotted in Fig. 2.

The width w of the Gaussian is determined by a balance between the large energy cost of small terraces and the entropy cost of restricting step widths. Elastic or dipole-induced step interactions are expected to decay as A/L^2 ,^{2,3} similar to the $g/\langle L \rangle^2$ decay of the free-energy cost due to step collisions.^{3,4,8} (Here g , which gauges the amount of step wandering, increases with T and is inversely to proportional to step "stiffness."^{8,12}) Consideration of simple models¹² shows that a signature of this form of energetic repulsion is

$$w \approx (g/4\pi^2 A)^{1/2} \langle L \rangle. \quad (1)$$

This *linear* relationship can be tested by our results on the 2.3° -misoriented surface, shown in Fig. 3: Again the data are poorly fitted by the noninteracting TSK model; however, they are remarkably well fitted by a Gaussian with a width obtained by using Eq. (1) to scale from the value of the width for the 1.2° data in Fig. 2 ($w_{1.2^\circ} \approx 1.56$ bisector units, or 35.9 Å). From the value of the Gaussian width we also estimate that $A/g \approx 6.7$.²¹

To emphasize the importance of equilibrium, we show in Fig. 4 for the same surface as Fig. 2, the much

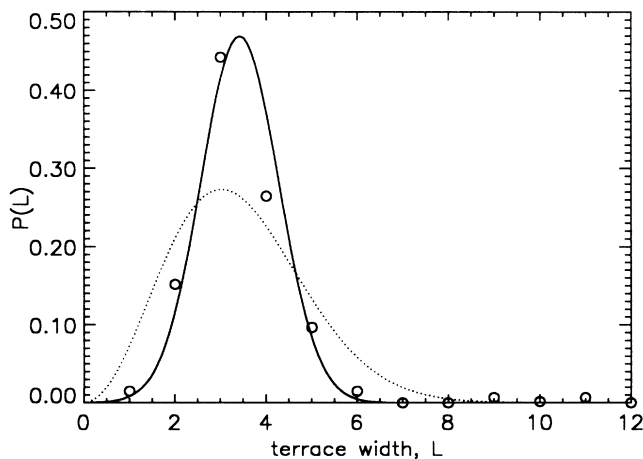


FIG. 3. Terrace-width distribution for an annealed 2.3° -misoriented sample (circles). The dotted curve is the prediction of the simple TSK model; the solid line is the Gaussian with a width obtained by using Eq. (1) to extrapolate from the value of the width found for the 1.2° data in Fig. 2.

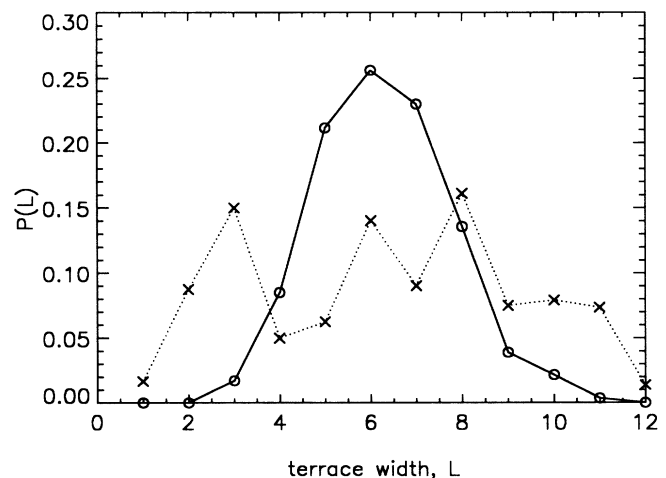


FIG. 4. Terrace-width distribution for a 1.2° -misoriented Si(111) surface quenched from 1250°C (crosses). The distribution for the slowly cooled surfaces (circles) of Fig. 2 is coplotted for comparison.

broader distribution, with a finite population of one- and two-unit terrace widths, that occurs for a *quenched* sample. Moreover, the step edges wander erratically and do not lie along corner holes.

Finally, we note that the existence of an energetic step-step repulsion also provides a natural mechanism for the formation of triple-layer height steps observed on highly misoriented surfaces.^{15,16} An isolated triple-layer height step costs more energy than three isolated single-height steps. However, with increasing step density this energy is outweighed by the decrease in step repulsion energy since triple-layer steps increase the average step separation.²²

In conclusion, we find that comparison of experimentally observed terrace-width distributions to those generated from the TSK model allows us to deduce the presence of short-range repulsions between steps in addition to the expected entropic repulsions. These interactions could be due to elastic strain: Empirical potential calculations for unreconstructed Si(111) predict substantial elastic step-step interactions.²³ Finally, we mention that STM can be used to probe other interesting statistical properties of stepped surfaces besides $P(L)$. In related work on 3.8°-misoriented samples,¹⁸ we have measured height-height correlation functions, and find agreement with the logarithmic divergences predicted by the statistical mechanics of "rough" surfaces.

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²¹In principle, one could estimate A from knowledge of g . However, to gauge g requires an estimate of the entropy of step wandering, which in turn requires an accurate estimate of kink energies and equilibrium temperature: The entropy cost of restricting a step depends *exponentially* on the kink energy (Ref. 4). On this surface obtaining this information is difficult because of the low density of kinks. On other surfaces, this program might be feasible: e.g., we extracted $P(L)$ from published pictures of Si(100) (Refs. 10 and 11), where the kink density is much higher; small L 's appeared less frequently than could be explained exclusively by entropic repulsions.

²²Our results thus contrast with the plausible theory of Alerhand *et al.* (Ref. 9) for the step-height doubling observed on Si(100): They propose that interactions induced by the anisotropy of the (2×1) reconstruction are responsible. We find a similar transition to triple-layer height steps in a case when such interactions do not exist.

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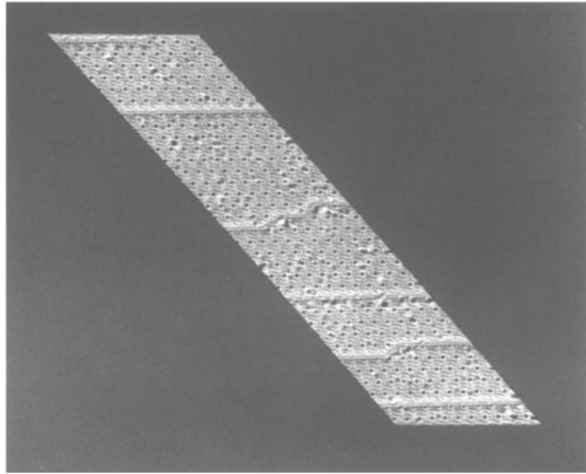


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