Sinuous Step Instability on the Si(001) Surface

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On slightly miscut Si(001) surfaces, straight steps are predicted to be unstable against the formation of long-wavelength undulations. These undulations lower the energy, by, in effect, reducing the size of the stress domains; they are thus analogous to the spontaneous step formation proposed by Alerhand *et al.* However, step undulations are expected to be kinetically favored, and therefore to preempt spontaneous step formation. Moreover, they lead to an unexpected distinct thermodynamic phase in the surface phase diagram at small angles.

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Steps on vicinal Si(001) surfaces have been intensely studied, especially since Alerhand *et al.* predicted such remarkable effects as spontaneous formation of steps [1], and a transition in step height with angle of miscut [2–4]. However, theoretical analyses to date have universally assumed that these steps are straight, except for random thermal meandering [1–7]. Yet recently, Tromp and Reuter [8], using low-energy electron microscopy (LEEM), observed steps on rather flat Si(001) surfaces to be sinuous rather than straight on a submicron length scale.

Here we show that, for sufficiently low step densities, straight steps are unstable against long-wavelength distortions, leading to a new phase transition on this surface. The cause is the interaction between surface stress domains. These results lead to a new picture of the structure and phase diagram of vicinal surfaces, and offer a natural explanation for the remarkable observation of Tromp and Reuter.

Alerhand *et al.* first recognized the importance of steps in creating stress domains on Si(001)2×1, and showed that a surface with sufficiently low step density could reduce its energy by introducing extra steps [1]. Given the strength of their argument, the failure to observe such extra steps has been a puzzle. The results here finally resolve this puzzle—step undulations can relieve stress and hence preempt the formation of extra steps.

Moreover, such undulations are kinetically preferred. There is a large barrier to nucleating extra steps, but little barrier to step undulations. Also, during either growth or sublimation (e.g., while heat cleaning), step flow places severe kinetic constraints on the step geometry. Unlike spontaneous formation of up-and-down steps, step undulations are compatible with step flow.

We begin by recalling the relevant features of the Si(001)2×1 surface, and of the continuum elastic model which has been successfully used to describe step interactions on this surface [1–5]. For unreconstructed Si(001), the surface lattice constant in the [110] direction is $a=3.84$ Å. For a surface miscut by an angle θ in the [110] direction, the separation between equally spaced single-layer steps is

$$L=a/2\sqrt{2}\tan\theta. \quad (1)$$

The Si(001) surface exhibits a 2×1 reconstruction in which pairs of atoms form dimers. Because of the atomic geometry, at single-layer steps the dimerization necessarily rotates by 90°, from 2×1 to 1×2 or vice versa. If the dimers on the upper terrace are perpendicular to the step edge, the step is called [5] S_A, or if parallel, S_B.

Because the stress is anisotropic and the domain rotates 90° at a step, the stress is discontinuous at the step. Using the known [9] stress tensor of the surface, we take the divergence of the stress to obtain the force on a step, referred to as a "force monopole" [1,6]. The elastic energy of the steps is then $-\frac{1}{2}\int d^2x\, d^2x' \chi_{ij}(\mathbf{x}-\mathbf{x}')f_i(\mathbf{x})f_j(\mathbf{x}')$, where f_i is the force density at the surface, and χ is the elastic Green's function of the surface. We calculate the Green's function numerically for a semi-infinite geometry, using the full cubic anisotropy with the experimental elastic constants. For sinusoidal steps, the Fourier transform of the force density can be calculated analytically. The integral for the elastic energy then transforms into a reciprocal-lattice sum, which is performed numerically.

The only other property needed to describe the steps is an energy per length for each type of step (S_A or S_B), reflecting a "local" energy in addition to the energy of the strain field. We do not include any "corner energy" [3], so that we can treat the continuum limit without considering the microscopic distribution of kinks in the meandering steps.

We omit thermal and entropic effects here. These have been extensively discussed already [2–4]. At large step separations, the steps meander about their minimum-energy positions. This meandering has a short correlation length; thus while it results in a renormalization of the local energies [2], it should not qualitatively affect the long-wavelength properties studied here.

We restrict consideration here to equally spaced identical sinusoidal steps, as shown in Fig. 1. Besides simplifying the elastic calculation, this allows the steps to be fully characterized by two numbers: the period λ and amplitude A of the sine wave. No distinction need be made here between S_A and S_B steps; because of the symmetrical step pattern assumed, only the sum of their energies enters. These restrictions are discussed further below.

For straight steps the energy E_s can be calculated analytically, giving the well-known [1,6] logarithmic

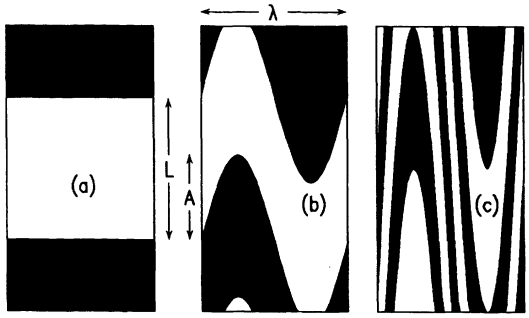

FIG. 1. Pattern of equally spaced sinusoidal steps used here. Black and white regions correspond to 2×1 and 1×2 domains, which are separated by single-layer steps. Step spacing L, wavelength λ, and amplitude A are indicated. (a) $A=0$, (b) $A=0.6L$, (c) $A=3L$. L and λ are the same in all three figures.

dependence on step separation L, $E_s=C_1-C_2\ln(L/S)$, where C_2 reflects the strength of the interaction, and C_1 characterizes the local energy of the step. Our value of C_2 is 29 meV/a, considerably larger than that suggested previously [1], mainly because of more accurate recent calculations of the stress anisotropy [9]. C_1 here represents an average of S_A and S_B local energies; its numerical value depends upon the (arbitrary) choice of S, and we adopt the convention $S=\pi a$ chosen by Alerhand *et al.* [1]. Since the actual step energy for Si is not well known, we somewhat arbitrarily choose a local energy such that $C_1=58$ meV/a. The effect of this choice and of other approximations is discussed below.

The energy per area, E_s/L, has a minimum at the step separation

$$L_0=S\exp(1+C_1/C_2)\,, \tag{2}$$

giving $L_0\approx 63a$ for the parameter values used here. [For the small angles of interest here, in discussing energy per area it is not necessary to distinguish between surface area and the projection of that area onto the (100) plane, or between θ and $\tan\theta$.] As was first pointed out by Alerhand *et al.* [1], a sufficiently flat surface could lower its energy by spontaneously forming additional steps to decrease the step separation to L_0. L_0 thus provides a second natural length scale in addition to the step separation L imposed by the miscut.

We now turn to the properties of sinusoidal steps like those in Fig. 1. Figure 2 shows the step contribution to the surface energy, on a surface with step separation $L=1000a$, corresponding to a miscut of 0.02°. Contours of constant energy are shown as a function of the amplitude A and wavelength λ of the undulations. Only contours with energy lower than that for straight steps are shown, so straight steps are unstable over the entire region within the outermost contour.

The behavior is surprisingly complex. There is a shallow local minimum in energy for straight steps, i.e., $A=0$. Increasing the amplitude raises the energy up to a

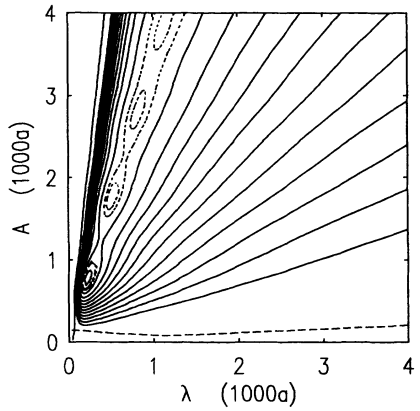

FIG. 2. Contours of constant step energy per surface area, as a function of amplitude A and wavelength λ, for $L=1000a$. Only contours with energy lower than -0.11 meV/a^2, the energy for straight steps at this separation, are shown. There are a series of local minima along the line $\lambda\approx 0.3A$, and a weak local minimum along the line $A=0$. The energy has a local maximum with respect to A along a ridge indicated by the dashed line. Successive contours differ in energy by 0.02 meV/a^2, with some supplemental dotted contours to better show the minima.

ridge indicated by the dashed line in Fig. 2. For still larger A, the energy drops, and a series of local minima are clearly seen, falling nearly along a line defined by $\lambda\approx 0.3A$.

To show the behavior along the minima more clearly, for each value of A we minimize the energy with respect to λ, and plot the resulting energy and wavelength in Fig. 3. In all cases studied, the first minimum with respect to

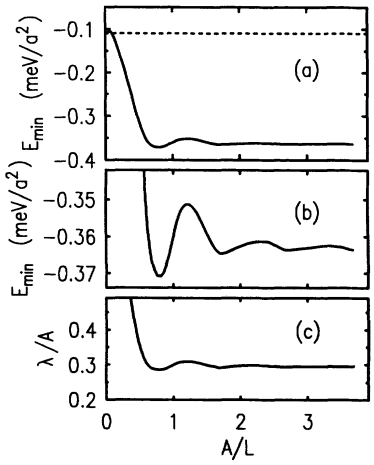

FIG. 3. Properties of steps on a surface with step separation $L=1000a$. For each amplitude A, the wavelength λ is that which minimizes the energy. (a) Energy per surface area vs reduced amplitude A/L. Amplitude is scaled by L to emphasize nearly perfect periodicity. Dotted line is energy for straight steps, for comparison. (b) Same as (a), on different scale to show oscillations. (c) Reduced wavelength λ/A vs A/L.

A was the deepest, but there were subsequent small oscillations about an asymptotic value. The period of these oscillations is simply the step spacing L, suggesting that the oscillations are due to a preference for having a specific alignment of the extrema of different steps. The ratio λ/A remains virtually constant beyond the first minimum in Fig. 3, consistent with the nearly straight trough in the energy surface seen in Fig. 2. The approach to an asymptotic value represents an approximate scaling relationship: The energy depends primarily on A/λ, with corrections due to the discrete step structure with period L.

Finally, to obtain an overview of the behavior of the surface, we calculate the minimum-energy step shape for a range of L. In Fig. 4, we see that, for step separations of about $200a$ or less, the straight steps have lower energy. For separations less than $150a$, we could not even find a local minimum with respect to λ and A. However, for step separations larger than $200a$, straight steps can lower their energy by developing undulations. At large step separations the energy appears to be approaching that of the minimum-energy surface, i.e., of the surface with step separation L_0.

The amplitude A of the minimum-energy steps, shown in Fig. 5, scales nearly perfectly with L as $A \approx 0.8L$, over the entire range $L > 200a$ where wavy steps are favored. The wavelength λ actually decreases with increasing L, so A/λ increases with L.

From Fig. 4, we see that the surface should undergo a phase transition with respect to angle of miscut, from a phase of straight steps to one of wavy steps. Surfaces with intermediate miscut should (if kinetics allow) facet into regions with $L \approx L_0$ and very flat regions of large L. Such faceting would still be compatible with step flow, and has apparently been observed by Tromp and Reuter [8]. Alternating up-and-down facets of miscut $L = L_0$ might have slightly lower energy; but like extra up-and-down steps, such up-and-down faceting would be incompatible with step flow.

We can get a semiquantitative understanding of the formation of step undulations in a rather simple way. In Fig. 1, we see that for large A substantial portions of the surface are covered with nearly straight steps at a spacing much smaller than L. Intuitively, we expect that the step undulations form in order to decrease the step spacing to a value closer to the minimum-energy spacing L_0.

The length of wavy steps (composed on an atomic scale of rectilinear segments) is increased by a factor of $1+4A/\lambda$; so we can think of the characteristic step spacing as being reduced roughly by that factor to $L/(1+4A/\lambda)$. If we assume that the energy is minimized when this characteristic spacing approaches L_0, we would expect that $L/(1+4A/\lambda) \approx L_0$, i.e.,

$$A/\lambda \approx (L - L_0)/4L_0 . \tag{3}$$

In Fig. 5(b), this linear relationship is included as a dotted line. (It appears as a curve due to the logarithmic scale.) The actual calculated results are seen to correspond rather well to the crude prediction (3), confirming our picture of the driving mechanism here.

Finally, it is important to address the limitations of the present study. Any inaccuracy in the stress anisotropy and in the local step energy C_1 simply changes the overall energy scale and the length L_0. A moderate change in the energy scale has no affect on our conclusions. While we have only studied one value of L_0, it is clear that step waviness should in general occur whenever L becomes much larger than L_0. We also note that the value of L_0 here is fortuitously close to that inferred by Tromp and Reuter.

We have treated the case of no applied external strain. However, even a modest external strain can significantly affect the stress-domain patterns at small miscut [1,10]. Small strains can easily occur accidentally in experi-

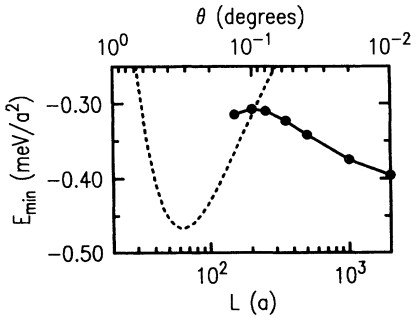

FIG. 4. Step energy per surface area vs step separation L (bottom scale), or angle of miscut (top scale). Dots correspond to sinusoidal steps, whose amplitudes A and wavelengths λ are those which minimize energy; solid curve is a spline fit to guide the eye. Dotted curve is the corresponding energy for straight steps.

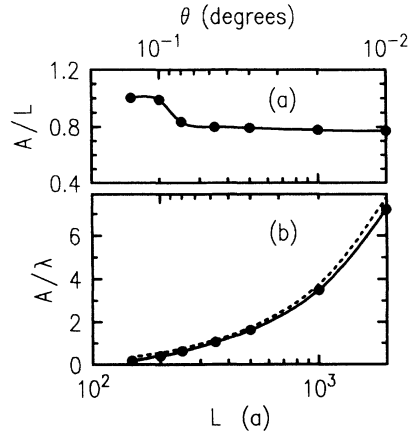

FIG. 5. Properties of minimum-energy steps vs step separation L (bottom scale), or angle of miscut (top scale). Each point corresponds to a point in Fig. 4. (a) Amplitude as fraction of L. (b) Dimensionless amplitude A/λ, along with linear relationship of Eq. (3) (dotted line).

ments, and so should be considered before attempting a detailed comparison with measurements.

We have also assumed a specific shape for the steps, based on analytic convenience. Thus our variational calculation in the parameters λ and A actually provides an upper bound on the energy of the wavy phase. This is enough to guarantee our central result, the instability of straight steps. Moreover, the assumed sinusoidal shape is physically reasonable, and is qualitatively consistent with experimental observations [8]; so it seems highly unlikely that a more accurate shape would greatly affect the overall behavior, see Fig. 4.

It would certainly be of interest to determine the actual step shape which minimizes the energy. In particular, meandering of the S_B steps is favored, since it creates segments of S_A step, which are believed to have rather small local energy. Meandering of S_A steps creates higher-energy S_B segments. Thus we expect S_B steps to have undulations of larger amplitude. Aside from the shapes of the individual steps, more complicated patterns of steps are possible, which would not repeat every two steps. Also, the presence of "kissing site" defects [11], associated with antiphase boundaries in the dimerization, appears to cause significant deviations from ideal behavior [8].

The only apparent discrepancy between theory and experiment [8] is the failure to observe the predicted large values of A/λ for large L. Step flow kinetics would tend to suppress large A/λ, especially given the rather weak dependence of energy on λ in Fig. 2. Large A/λ could also be disfavored if the step undulations are coherent only over small patches, as in the experiments, due to defects. And at very large L, even small external strains could affect the results.

Spontaneous step formation [1] has the advantage that only the low-energy S_A steps are created. However, even if this should prove to be the structure of lowest energy, it might not be kinetically accessible. A sequence of S_A steps necessarily has an up-and-down pattern. Step flow would quickly eliminate such steps, leaving only the monotonic sequence of steps associated with the miscut. Such step flow occurs not only while growing by vapor deposition, but also during sublimation while heat cleaning the surface [8]. At temperatures low enough to suppress sublimation, the energetic barrier to spontaneous step formation might be prohibitive.

In contrast, step waviness can reduce the elastic energy without interfering with step flow. And the phenomena observed experimentally [8] are all in accord with this picture, including step flow during high-temperature cleaning, coherent step undulations over large areas, and apparently even faceting into regions of more closely spaced straight steps and widely spaced wavy steps. Thus very flat Si(001) surfaces provide a window onto an unexplored regime with a wealth of fascinating new phenomena.

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[1] O. L. Alerhand, D. Vanderbilt, R. D. Meade, and J. D. Joannopoulos, Phys. Rev. Lett. **61**, 1973 (1988).

[2] O. L. Alerhand, A. N. Berker, J. D. Joannopoulos, D. Vanderbilt, R. J. Hamers, and J. E. Demuth, Phys. Rev. Lett. **64**, 2406 (1990).

[3] T. W. Poon, S. Yip, P. S. Ho, and F. F. Abraham, Phys. Rev. Lett. **65**, 2161 (1990).

[4] E. Pehlke and J. Tersoff, Phys. Rev. Lett. **67**, 465 (1991); **67**, 1290 (1991).

[5] D. J. Chadi, Phys. Rev. Lett. **59**, 1691 (1987).

[6] V. I. Marchenko and A. Y. Parshin, Zh. Eksp. Teor. Fiz. **79**, 257 (1980) [Sov. Phys. JETP **52**, 129 (1980)].

[7] The possibility that straight steps might be energetically unstable was previously suggested by X. Tong and P. A. Bennett, Phys. Rev. Lett. **67**, 101 (1991). However, they had in mind much smaller step separations, where the effect discussed here does not occur.

[8] R. M. Tromp and M. C. Reuter, following Letter, Phys. Rev. Lett. **68**, 820 (1992); R. M. Tromp (private communication).

[9] R. D. Meade and D. Vanderbilt, in *Proceedings of the Twentieth International Conference on the Physics of Semiconductors,* edited by E. M. Anastassakis and J. D. Joannopolous (World Scientific, Singapore, 1990), p. 123; M. C. Payne, N. Roberts, R. J. Needs, M. Needels, and J. D. Joannopoulos, Surf. Sci. **211/212**, 1 (1989).

[10] F. K. Men, W. E. Packard, and M. B. Webb, Phys. Rev. Lett. **61**, 2469 (1988).

[11] B. S. Swartzentruber, Y. W. Mo, and M. G. Lagally, Appl. Phys. Lett. **58**, 822 (1991).