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Step-step interactions due to anisotropic surface stress

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I have derived the effective potential that controls the motion of a single step on a crystal like Si(001) that has two inequivalent surface domains. The behavior of the steps is controlled by such a potential times a correlation length that measures the stiffness of the step edge against kinking. This correlation length may be of order 100 lattice constants. The potential derived here is compared with published images, by measuring the amplitude of thermal fluctuations in the step-edge positions.

One of the surprising things in nature is the near periodicity of the spacing of steps on crystalline surfaces. For a surface in equilibrium, this regularity must be the result of a balance between thermal fluctuations and an interaction between steps. Here, I generalize the calculations of Alerhand, Vanderbilt, Meade, and Joannopoulos[1] to derive an interaction potential per unit length of step between one step in an array and the rest of the step array. Further, the kink energies are shown to have a dramatic effect on the effective interaction by establishing a correlation length along the step edge that can become large.

Alerhand *et al.* showed for Si(001), or any surface with two inequivalent domains with anisotropic surface stress tensors, that the ground state of the crystal contains steps. The strain energy of such a crystal diverges logarithmically with the distance between steps. With large spacing between steps, it becomes energetically favorable to create more steps. Their theory nicely explains the low-energy electron diffraction experiments of Men, Packard, and Webb,[2] which showed that the predominant domain on the Si(001) surface can be changed from 1×2 to 2×1 by application of external strain. An interaction that is large enough to make it favorable to create steps can reasonably be expected to control the motion of steps.

The calculation of interstep interactions centers on an evaluation of the elastic strain energies. For a Si(001) surface covered by single-layer steps, one must cross two steps to reach an equivalent surface. Si cut within 2° of the (001) direction behaves in this manner: The basic reconstruction is a dimerization of the surface atoms, and the orientation of the dimers differs by 90° on opposite sides of each step. The two varieties of step are denoted "*A*" and "*B*;" type-*A* steps have the dimers on the upper terrace perpendicular to the step edge.

The strain energy calculated here is a result of bond bending as the surface reconstructs to reduce the number of dangling bonds. There are two equivalent ways of looking at the problem: as terraces with a nonzero stress tensor that are separated by domain walls, or by considering the forces that would produce the same strain in an unreconstructed crystal. One can think of the strain fields as being generated by attaching springs between the steps. The forces necessary to produce the strain field for Si(001) are[1]

$$f(x)=\sum_i(-1)^i F_0\delta(x-x_i) \tag{1}$$

in the plane of the surface, perpendicular to each step. For Si(001), $F_0=0.07$ eV/Å (Ref. 3) is the anisotropy of the surface stress, and x_i is the position of the ith step. The calculation is done in terms of the Fourier transforms of position coordinates for simplicity. I use a continuous, rather than the discrete, Fourier transform used in Ref. 1 to calculate the elastic energy of an infinite periodic array of alternating type-*A* and type-*B* steps, with a particular step displaced by a distance d from its ideal position:

$$x_n=\begin{cases} nL/2 \text{ if } n\neq 0\,, \\ d \text{ if } n=0\,. \end{cases} \tag{2}$$

For this situation, the force is given by

$$f(g)/F_0=\left[-1+e^{idg}+\frac{(1-e^{iLg/2})}{L}\times\sum_{n=-\infty}^{\infty}\delta(g-2\pi n/L)\right]e^{-a|g|}\,. \tag{3}$$

A microscopic cutoff length on the scale of the lattice constant is introduced by smoothing the force distribution of Eq. (1) by a Lorentzian of width a. This length scale corresponds to the size of a step.

The elastic Green's function for interacting domain walls can be obtained from the Green's function of a point force at the origin applied to a half-space of material:[4,5]

$$X_{\text{point}}(\mathbf{r})=\frac{1+\sigma}{2\pi E_y}\frac{1}{r}\left[2(1-\sigma)+2\sigma\frac{x^2}{r^2}\right]\,, \tag{4}$$

where E_y is the Young's modulus, σ is Poisson's ratio, $r^2=x^2+y^2$; $\mathbf{r}=(x,y)$. Calculations that attempt to model steps slightly more closely by changing the distribution of forces yield a Green's function with the same dependence on $\mathbf{r}$ or g. We assume the step extends along the y axis, so the domain wall applies a line of force along the y axis in the x direction. We can then write the Green's function for a wall as

$$X(\mathbf{r})=\int_{-\infty}^{\infty}X_{\text{point}}(\mathbf{r})dy \tag{5}$$

or

$$X(g)=2\frac{1-\sigma^2}{E_y}\frac{1}{|g|}\,. \tag{6}$$

The elastic energy is

$$E_{\mathrm{el}}=\frac{-1}{2\pi}\int_{-\infty}^{\infty}f(g)X(g)f^{\dagger}(g)dg\,. \tag{7}$$

If this is evaluated, the d-dependent part of E_{el} is

$$E_{\mathrm{el}}(d,L)=\frac{1-\sigma^2}{\pi E_y}\frac{2F_0^2}{L}\left[-\ln\left(\frac{4a^2+d^2}{4a^2}\right)+\ln\left(\frac{1+2e^{-4\pi a/L}\cos(2\pi d/L)+e^{-8\pi a/L}}{1-2e^{-4\pi a/L}\cos(2\pi d/L)+e^{-8\pi a/L}}\right)\right], \tag{8}$$

where the first term comes from the product of the continuous part of f with itself, and the other term comes from the continuous part times the discrete parts.

Figure 1 displays the step-step interaction [Eq. (8)] for several values of mean step spacing. The interaction is approximately $E_{\mathrm{el}}(d,L)\approx\alpha(L)\beta(d/L)$, where α is a slowly (logarithmically) increasing function of L, and β describes the shape of the potential. This functional form implies that thermal fluctuations in the step position should be a nearly constant fraction of the distance between steps, independent of L.

We can compare this calculation to available scanning-tunneling-microscopy (STM) images by measuring the variance in interstep spacings and ascribing it to thermal fluctuations. The calculation yields a step energy per unit length, which must be multiplied by a correlation length ζ that measures how much of the step edge moves as a unit. Under the assumption that the surface has equilibrated, any interaction will have an expectation value per unit length of approximately kT/ζ. This length is certainly larger than two unit cells, as STM images show that steps never kink by splitting a dimer (see, for instance, Fig. 3 in Ref. 6 or Fig. 2 in Ref. 7). Casual inspection of images (Fig. 1 in Ref. 8, Fig. 5 in Ref. 9, or Fig. 5 in Ref. 10) shows that the type-A steps are straight for more than ten unit cells, while type-B steps typically kink after four.

However, ζ is not necessarily the distance between kinks. If a step is forced to lie in some direction other than [110], a minimum density of kinks is enforced by the geometry, but the step can still be correlated across several kinks. To calculate ζ, and to investigate this, I ran a series of Monte Carlo simulations of a step edge in a gentle parabolic potential of strength c. The step edge was just described by a single kink energy E_{kink}, and the following Hamiltonian:

$$H=c\sum_i x_i^2+E_{\mathrm{kink}}\sum_i|x_i-x_{i+1}|\,, \tag{9}$$

where x_i is the position of a step at location i, and i is measured in units of dimer spacings. Equation (9) does not contain direct kink-kink interactions; they are estimated to be small. Simulations were run in periodic boundary conditions on arrays of 128–1024 dimers, and step directions from [110] to [100], and $\langle x^2\rangle$ was measured. If $E_{\mathrm{kink}}=0$, the dimers are uncorrelated so $\zeta=2a_0$ (one dimer spacing), and we recover the result of the equipartition theorem: $c\langle x^2\rangle=kT/2$. If we consider a region of length ζ to move as a unit, we can approximate the system as a collection of independent objects. To satisfy the equipartition theorem, the correlation length must be $\zeta\equiv kT/c\langle x^2\rangle$. We have checked that ζ is independent of c for a broad range of c about these simulations.

Figures 2 and 3 show ζ as a function of E_{kink}/kT and as a function of the density of geometrically required kinks.

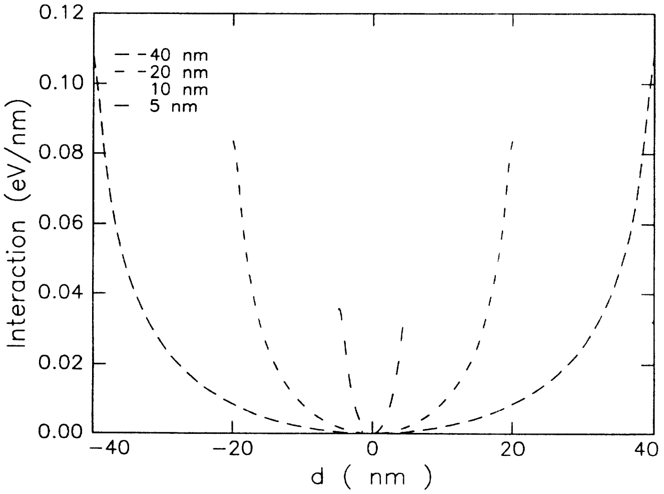

FIG. 1. A plot of the calculated interaction energy per unit length of step vs the displacement of a step from uniform spacing. The curves are calculations for mean step spacings of 50, 100, 200, and 400 Å.

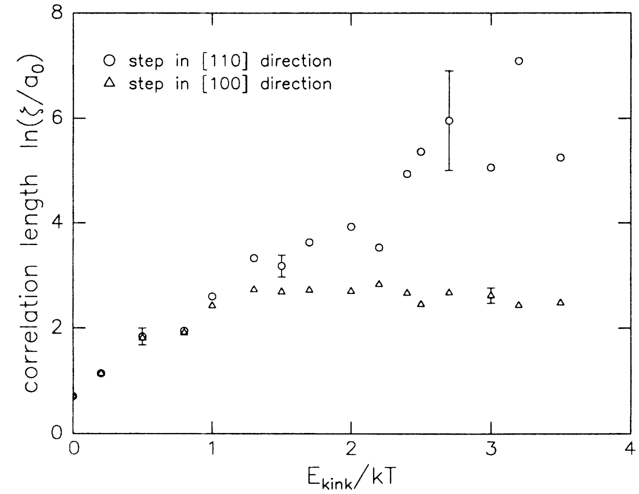

FIG. 2. The correlation length along the step edge ζ vs E_{kink}/kT. ζ measures how much of the step edge moves as a unit in response to interactions with other steps. The circles are calculations for steps with no geometrically required kinks (running along [110]) and the triangles correspond to a step along [100], with a high density of enforced kinks. The error bars are based on differences between three independent runs.

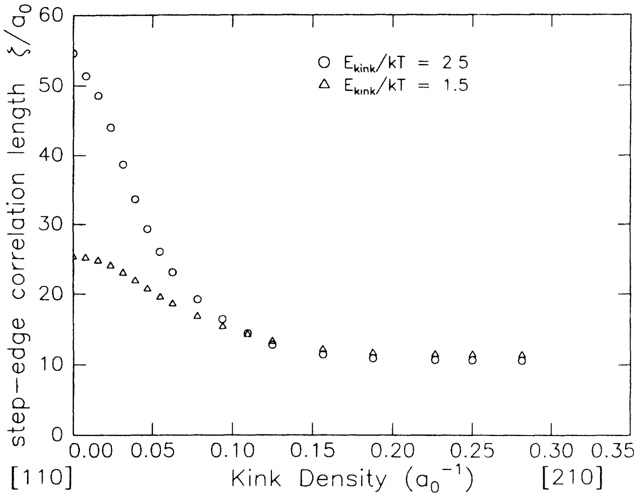

FIG. 3. The correlation length, ζ vs the direction of the step. The horizontal axis shows the density of geometrically enforced kinks, corresponding to steps from [110] to [210].

It can be seen that forcing kinks by tilting the step away from [110] rapidly reduces ζ to a value that is independent of E_{kink}.

Consideration of the atomic structure of a kink leads to the conclusion that a type-*B* step is a reasonable approximation for a kink in a type-*A* step, and vice versa. A calculation of step energies by Chadi[11] gives an energy of 0.30 eV for a type-*B* step $2a_0$ long, and 0.02 eV for a similar type-*A* step. If we assume Chadi's energies and that the surface freezes at $T=900$ K, ζ is $3a_0$ for a type-*B* step, and can be as large as $1000a_0$ for a type-*A* step oriented exactly along $[1\bar{1}0]$. Defects and misorientation of the surface will reduce these values for ζ, but large values are clearly plausible.

Before we compare the data to the calculations, we need to consider possible problems with interpreting the data. Virtual interactions between steps are possible, mediated by collisions between adjacent steps.[12] However, in general, and in the data used here, it is rare for two steps to collide. Alternatively, Schwoebel and Shipsey[13] proposed a kinetic effect that can force steps to be evenly spaced if the crystal is growing or subliming. This effect could be avoided by low-temperature annealing, so that substantially less than a monolayer is evaporated from the surface. Even normal cooling rates after high-temperature processing (on the order of $1\ \mathrm{K\,s^{-1}}$) may be slow enough to allow the step configuration to equilibrate below 1200 K, where evaporation becomes negligible; Sakamoto *et al.*[14] observed significant step motion at temperatures as low as 750 K within just a few minutes.

If all the different excitations of the surface (e.g., different sizes of kinks) freeze out at nearly the same temperature, the STM images should be a faithful representation of the silicon surface at this freeze-out temperature. If this were not the case, the STM would display a surface that is not in thermal equilibrium.

Bearing these difficulties in mind, we can compare the data with these calculations. The data selected, Fig. 1 of the paper by Hoeven *et al*,[8] and Fig. 5 of Swartzentruber *et al.*,[9] show a large area, yet individual dimer rows can be counted. The step edges were digitized by visually counting dimer rows in enlarged images. The expectation value of $E_{\mathrm{el}}(d)$ was computed from Eq. (7) by averaging at intervals of one dimer spacing. L was taken to be the mean spacing of the triplet of steps surrounding the step of interest, and d was measured from the mean position of the middle step. This procedure will slightly underestimate $\langle E_{\mathrm{el}}(d)\rangle$, as it neglects the long-wavelength fluctuations in the step position, but this is a small effect. Errors in the counting process are ± 1 dimer, yielding a $\approx 10\%$ error in $\langle E_{\mathrm{el}}(d)\rangle$.

For the type-*B* step in Hoeven *et al.*, $\langle E_{\mathrm{el}}\rangle = 3.6\times10^{-4}$ eV/a_0, which would require $\zeta \approx 260a_0$ to be in thermal equilibrium at 900 K. The equivalent value for the steps in Swartzentruber *et al.* is $\langle E_{\mathrm{el}}\rangle = 1.8\times10^{-3}$ eV/a_0 with the corresponding value of ζ being $50a_0$. Calculations that use the measured directions of the steps yield $\zeta = 3a_0$, assuming Chadi's energies. At 900 K, no value of E_{kink} can make the calculated and experimental values agree. Essentially, the experimental step edges are significantly smoother than these calculations can reasonably produce. Agreement between the data and calculations can only be forced if $E_{\mathrm{kink}} \geq 0.1$ eV for a $2a_0$ kink in a type-*B* step, and if we make the unreasonable assumption that the step edge equilibrates at 300 K.

The type-*A* step is even straighter, with $\langle E_{\mathrm{el}}\rangle = 1.3\times10^{-4}$ eV/a_0, or $\zeta \approx 700a_0$ at 900 K (Hoeven *et al.*). The other image yields 4×10^{-4} eV/a_0 and $250a_0$. For the measured step directions, I calculate $\zeta = 50a_0$ and $15a_0$, respectively. The calculated steps cannot be straightened enough by increasing E_{kink} or lowering the temperature even to 300 K. It is worth noting, however, that the calculations agree with the data that the steps in the data of Hoeven *et al.* should be the straighter of the two sets because the steps have a lower density of geometrically required kinks.

These are several possibilities that could account for the general behavior of the steps. First, the STM may be imaging a nonequilibrium surface. Different aspects of the surface may freeze out at different temperatures, and STM images of silicon may not represent the high-temperature structure. Second, Eq. (9) may be incorrect in its assumption that the total energy of a kink is proportional to the size of the kink. If the middle segments of a large kink are assumed to cost less energy than the corner sections, the data can be reproduced with Chadi's energies for the middle segments. This is physically reasonable, but the data are not sufficient to justify extracting another parameter. Third, kinks may have long-range interactions.

This paper has presented a calculation showing a strong repulsive interaction between single steps on Si(001), Ge(001), and similar surfaces that have multiple domains and anisotropic surface stress tensors. Step edges may be stiff on length scales of tens of hundreds of lattice constants because kinks can be too energetically costly to be thermally excited.

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

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

[3] M. C. Payne, N. Roberts, R. J. Needs, M. Needels, and J. D. Joannopoulos, Surf. Sci. **211**, 1 (1989).

[4] L. D. Landau and E. M. Lifshitz, *Theory of Elasticity* (Addison-Wesley, Reading, MA, 1959), pp. 26–29.

[5] P. Wynblatt, in *Interatomic Potentials and Simulation of Lattice Defects,* edited by P. C. Gehlen, J. R. Beeler, and R. I. Jaffee (Plenum, New York, 1972), p. 633.

[6] J. E. Griffith, J. A. Kubby, P. E. Wierenga, and G. P. Kochanski, in *Heteroepitaxy on Silicon: Fundamentals, Structures, and Devices,* edited by H. K. Choi, R. Hull, H. Ishiwara, and R. J. Nemanich, MRS Symposia Proceedings No. 116 (Materials Research Society, Pittsburgh, PA, 1988).

[7] J. A. Kubby, J. E. Griffith, R. S. Becker, and J. S. Vickers, Phys. Rev. B **36**, 6079 (1987).

[8] A. J. Hoeven, J. M. Lenssinck, D. Dijkkamp, E. J. van Loenen, and J. Dieleman, Phys. Rev. Lett. **63**, 1830 (1989).

[9] B. S. Swartzentruber, Y.-W. Mo, M. B. Webb, and M. G. Lagally, J. Vac. Sci. Technol. A **7**, 2901 (1989).

[10] J. E. Griffith, G. P. Kochanski, J. A. Kubby, and P. E. Wierenga, J. Vac. Sci. Technol. A **7**, 1914 (1989).

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

[12] G. H. Gilmer and J. D. Weeks, J. Chem. Phys. **68**, 950 (1978).

[13] R. L. Schwoebel and E. J. Shipsey, J. Appl. Phys. **37**, 3682 (1966).

[14] K. Sakamoto, T. Sakamoto, S. Nagao, G. Hashiguchi, K. Kuniyoshi, and N. Taskahashi, in *Proceedings of the Second International Symposium on Silicon Molecular Beam Epitaxy, Honolulu, 1987,* edited by J. C. Bean and L. J. Schowalter (Electrochemical Society, Pennington, NJ, 1988).