Step-Bunching Instability of Vicinal Surfaces under Stress

J. Tersoff
IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598

Y. H. Phang, Zhenyu Zhang,* and M. G. Lagally
University of Wisconsin, Madison, Wisconsin 53706
(Received 9 June 1995)

On a vicinal surface under stress, elastic relaxation at steps produces a long-range attractive interaction between the steps. As a result, the surface is unstable against step bunching, driven by the energetics of the system rather than by the kinetics of step flow. This bunching instability differs from the predictions of previous continuum models by its lack of a characteristic wavelength. Instead, it evolves by progressive coalescence of step bunches. Flux can dramatically modify this evolution, limiting the growth of step bunches.

PACS numbers: 68.35.Fx, 68.35.Bs

For several years it has been recognized that the surface of a solid under stress exhibits a morphological instability, characterized by spontaneous roughening [1–5]. Such roughening is important for semiconductor technology, given the growing role of strained layers in electronic devices. Yet the understanding of stress-induced roughening is still incomplete. Early work [1–3] focused on continuum models and predicted a true instability. More recently it has been noted that stress-induced roughening of an initially flat surface requires nucleation of steps or facets [5,6], so that a flat surface may be metastable rather than unstable.

In practice most semiconductor devices are fabricated on vicinal substrates, i.e., substrates with an orientation that is a few tenths of a degree off the crystallographic plane. This misorientation creates a substantial density of steps, which are intended to facilitate smooth growth by step flow. Nevertheless, steps are sometimes observed to form bunches, giving a rougher surface. Such bunching is usually attributed to the kinetics of step flow [7–12]. There have been suggestions that attractive interactions between steps could also lead to step bunching [13,14]. However, the proposed interactions are rather short ranged, and even their existence remains speculative.

Here we show that on vicinal strained layers, there is quite generally a long-ranged attraction between steps, which leads to a robust step-bunching instability with several novel features, even under ideal conditions of step-flow growth. In contrast to continuum models [1–4], this instability has no characteristic wavelength, but proceeds by progressive coalescence of step bunches. However, if the surface is exposed to a flux of atoms, coalescence terminates at a maximum bunch size that depends on the flux. Step bunching on strained layers has already been observed by several groups [15–18], and we believe these observations represent precisely the effect discussed here.

Recently Duport, Nozières, and Villain [10] also proposed a step-bunching instability. However, that instability is purely kinetic, caused by the presence of a kinetic barrier to diffusion at steps [12]. Stress enters only in determining the magnitude of the diffusion barrier. Thus there would be no instability in the limit of high temperature, where such a barrier becomes unimportant. Moreover, that instability occurs only for certain values of the material parameters. In contrast, the instability discussed here is present for any strained layer, even for zero flux or high temperature.

We begin by analyzing the interaction between steps and deriving their equation of motion. Steps move discretely by the incorporation or detachment of atoms at kinks. However, if steps are not pinned, and their meandering is not too large, we need only consider the average position of each step. The problem then becomes one dimensional, and the step position is a continuous variable.

The principal interactions between steps (aside from contact interactions) are elastic in nature. For simplicity, we assume that the system has only one type of step and that all terraces are equivalent. Without strain, the only elastic interaction is the repulsion arising from the intrinsic stress (the "force dipole") of the steps [19], which has the form $\frac{1}{2}\alpha_2 L^{-2}$ for steps at distance L. There is also a logarithmic repulsion when successive terraces are inequivalent [20].

However, there is an additional effect at the surface of a solid under stress, e.g., a strained layer. At each step, there is a discontinuity in the surface height. Thus the lateral force from the strained material on one side is not balanced by an equal force from the other side. The result is a net force at the step, i.e., a "force monopole." The magnitude of this force is simply the discontinuity in the stress, i.e., the bulk stress σ times the step height h. There is an equal and opposite force distributed over the terraces, but this has no effect on the step motion.

It is well known [20] that such force monopoles lead to logarithmic interactions between steps. This

0031-9007/95/75(14)/2730(4)$06.00

interaction has very recently been directly observed in atomistic simulations [21]. The novel feature here is that successive steps have forces in the same direction, so the interaction is attractive, of the form $\alpha_1 \ln(L)$. Here $\alpha_1 = \sigma^2 h^2/M$, M being an elastic constant. Taking the derivative of step energy with respect to position, including both terms discussed above, the total force per unit length on the mth step, f_m, is

$$f_m = \sum_{n \neq m} \left(\frac{\alpha_1}{(x_n - x_m)} - \frac{\alpha_2}{(x_n - x_m)^3} \right). \tag{1}$$

where x_m is the position of the mth step in the direction perpendicular to the steps. Equation (1) represents the force conjugate to the step position, i.e., the force driving step motion, and should not be confused with the "force monopole" [20] discussed above, which is conjugate to elastic distortion of the surface.

To derive the resulting step motion, we assume that the adatom densities on both sides of the step are in equilibrium with the step. (This neglects for simplicity the possibility [12] of a rate limiting diffusion barrier at the step, which can either drive step bunching or oppose it.) The adatom density η per unit site at the mth step is then

$$\eta(x_m) = e^{-(E_1 + f_m A)/kT}, \tag{2}$$

where E_1 is the formation energy of an adatom (i.e., the energy to dissociate an adatom from a kink in a step) in the absence of any force, and A is the area per surface site.

We solve the diffusion equation with an adatom flux F per site,

$$\frac{\partial \eta}{\partial t} = D \nabla^2 \eta + F, \tag{3}$$

where t is time and D is the adatom diffusion coefficient, subject to the boundary conditions of Eq. (2). Evaluating the divergence of the adatom current at the step gives the velocity v_m of the mth step:

$$v_m = F\left(\frac{x_{m+1} - x_{m-1}}{2} \right) - B\left(\frac{f_{m+1} - f_m}{x_{m+1} - x_m} - \frac{f_{m-1} - f_m}{x_{m-1} - x_m} \right), \tag{4}$$

where

$$B = \frac{DA}{2kT} e^{-E_1/kT}. \tag{5}$$

B is extremely sensitive to temperature, so we expect that stress-driven step bunching should be a strong effect at high temperature, but negligible at low temperature. The data needed to quantify B do not exist, to our knowledge.

We address two distinct issues here: an initial bunching instability and the subsequent evolution of the surface morphology. The existence of a bunching instability can be determined by perturbing the steps from their initially equispaced positions and calculating the linear response. Let $u_m(t)$ denote the deviation of the step from the position it would have under ideal step flow, i.e.,

$$u_m(t) = x_m(t) - L_{\text{av}}(m + Ft), \tag{6}$$

where L_{av} is the average step separation. For a perturbation of amplitude Δ and period N steps, we substitute $u_m(t = 0) = \Delta \cos(2\pi m/N)$ into Eq. (4) and integrate the velocity. The result, for small times and to first order in Δ is

$$u_m(t) \approx e^{rt} \Delta \cos\left(\frac{2\pi}{N} m + \frac{2\pi}{N} Ft \right), \tag{7}$$

where

$$r = \left(\frac{B\alpha_1^2}{\alpha_2} \right) \frac{L_0^2}{L_{\text{av}}^3} \frac{8\pi^4}{N^3} \left\{ 1 - \frac{1}{N} + \frac{L_0^2 \pi^2}{L_{\text{av}}^2 N} \left(-1 + \frac{2}{N} - \frac{1}{N^2} \right) \right\}, \tag{8}$$

and $L_0 = \sqrt{(\alpha_2/\alpha_1)}$ is the minimum-energy separation for an isolated step pair. Separately, we have verified by direct numerical integration that Eq. (7) gives a good description of the short-time evolution.

For a given period N of the sinusoidal perturbation, Eq. (8) describes a step-bunching instability if $r > 0$. Note that the existence of an instability, and the rate r of initial bunching, is entirely independent of the flux. However, from Eq. (8), it is clear that the instability depends sensitively on L_0/L_{av}. This dependence is shown in detail in Fig. 1. For widely spaced steps, where the short-ranged repulsion is unimportant, the bunching instability exists for all wavelengths, i.e., $r > 0$ for all N. Moreover, the rate of bunching increases rapidly with decreasing wavelength.

For more closely spaced steps, the most unstable wavelength moves to a larger value, and the corresponding bunching rate r decreases dramatically. (Note the different vertical scales in Fig. 1.) No matter how densely spaced the steps, there is, in principle, a bunching instability. However, because of the long wavelength and very slow rate, the instability may become kinetically irrelevant for $L_{\text{av}} \ll L_0$.

We now address the subsequent evolution of the step bunching in the physically interesting regime $L_{\text{av}} \gg L_0$. The evolution of the surface morphology in the most general case can only be obtained numerically by direct integration of Eq. (4). In such calculations, we typically use a system size containing 500 steps, with periodic boundary conditions. A specific example is shown in Fig. 2 for $F = 0$ (no deposition flux). The initial configuration is chosen to have small random deviations from uniform step spacing. Figure 2(a) shows that, initially, small step bunches are formed, and these coalesce into progressively larger bunches over time,

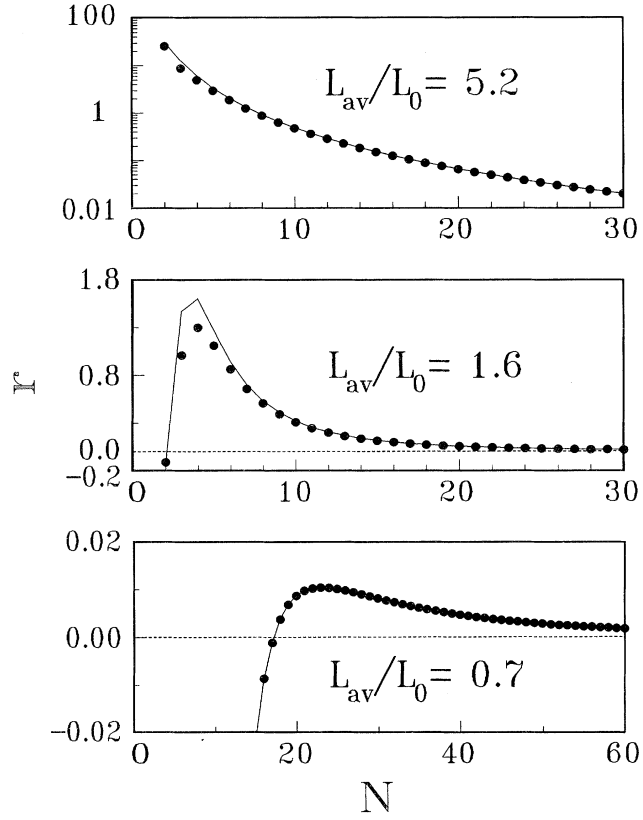

FIG. 1. Rate r of initial step bunching after a sinusoidal perturbation of period N steps is created, vs N. The solid lines and dots are results of Eq. (8) and of numerical integration, respectively. Values of the dimensionless step spacing L_{av}/L_0 are indicated. Note the very different vertical scales in the three panels.

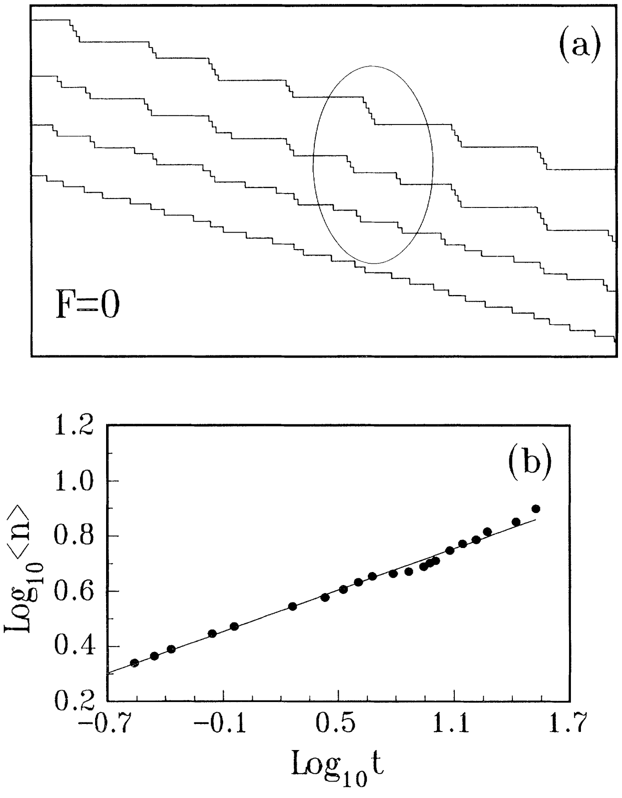

FIG. 2. Evolution of surface morphology without flux. (a) Surface morphology at simulation times (from bottom to top) of 0, 0.1, 0.7, and 4.7 for $B = 1 \times 10^5, \alpha_1 = 1, \alpha_2 = 100$, and $L_{\mathrm{av}} = 52$. Five steps are indicated, which over time coalesce into one bunch. The length of the system shown is $150L_0$. (b) Characteristic bunch size $\langle n\rangle$ vs time. The slope of the straight line, which is the linear least-square fit to the data, is 0.25 ± 0.01.

consistent with the faster bunching at shorter length scales predicted in Fig. 1.

Figure 2(b) shows the evolution of the characteristic bunch size $\langle n\rangle = (\sum_k n_k^2)/\sum_k n_k$, where n_k is the number of steps in the kth bunch. In calculating $\langle n\rangle$, two bunches are considered separate if the terrace separating them is larger than L_{av}. The bunches grow monotonically with time. The result is consistent with a dependence $\langle n\rangle \propto t^{1/4}$, although we have not analyzed this behavior in detail.

The long-time evolution in the presence of a flux raises complex issues of kinetics, quite aside from the underlying instability, and so a complete discussion lies outside the scope of this paper. Here we simply note the most salient features, summarized in Fig. 3. As for $F = 0$, the bunch size initially grows with time. However, the slope in Fig. 3 implies a $\langle n\rangle \propto t^{1/6}$ behavior, compared with $\langle n\rangle \propto t^{1/4}$ in Fig. 2. While the data in both cases are over too narrow a range to take these exponents very seriously, both power laws do appear in discussions of surface morphology [22].

The important point to note in Fig. 3, though, is that the bunching does not proceed indefinitely. The bunches appear to reach a maximum size, at which they stop growing. We have confirmed that this maximum bunch size decreases with increasing flux. A detailed discussion will be presented elsewhere, but the qualitative origin of this behavior can be simply described. Consider large bunches of many steps, separated by very large step-free terraces. Most of the flux then lands on the terrace, where it diffuses to the first (lowest) and the last (uppermost) steps of the bunches. If the flux is sufficiently large, relative to the intrabunch diffusion and step binding, the effect will be to eject the leading (lowest) step of the bunch. This "debunching" mechanism was recently addressed theoretically by Kandel and Weeks [23], who showed that the essential properties are correctly captured in a one-dimensional model such as ours.

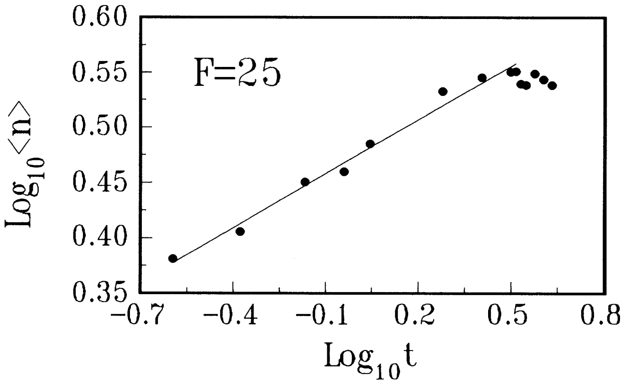

FIG. 3. Evolution of the bunch size $\langle n \rangle$ with time, for a flux $F = 25$. Other parameters are the same as for Fig. 2. The slope of the straight line, which is the linear least-square fit to the data before saturation, is 0.16 ± 0.01.

Thus, while the energetics drive roughening of the strained layer via step bunching, this bunching can be substantially suppressed by a high growth rate (large flux), or equivalently by low growth temperature. Not only is the maximum bunch size reduced for large flux and wide terraces, but each bunch will also eject many steps before capturing any steps from the upstream bunch. We therefore anticipate a steady state in which there are a finite number of "free" steps between bunches at any given moment [23], further reducing the total roughness.

This effect is potentially crucial for the growth of electronic devices that contain strained layers as one component. While we have shown that the tendency to step bunching can never be entirely eliminated, the presence of weakly bound bunches of only a few steps is probably acceptable from a technological viewpoint. Of course, whether the flux and temperature required to suppress roughening are consistent with other processing requirements depends upon the specific system.

In conclusion, for a vicinal surface under stress, the elastic relaxation around each step causes a logarithmic attraction between steps, leading to a step-bunching instability, regardless of the flux or step density. However, the ultimate surface morphology depends in a crucial way upon flux. It is likely that the step bunching observed in strained layers [15–18] has as its foundation the mechanism we describe here. Careful experiments to explore the relationship between bunch morphology and kinetic parameters will allow quantification of the final essential point made above: that surface morphology of a film under stress can actually be kept smoother when the film is grown far from equilibrium than it would be for a film grown near equilibrium.

The work of Y. H. P. and M. G. L. was supported by NSF Grant No. DMR 92-01856.

Note added.—After this paper was submitted, we received a preprint that also addresses this problem, though with a different emphasis [24].

---

*Present address: Oak Ridge National Laboratory, Oak Ridge, TN 37831.

[1] R. J. Asaro and W. A. Tiller, Metall. Trans. **3**, 1789 (1972).
[2] M. A. Grinfeld, J. Nonlinear Sci. **3**, 35 (1983).
[3] D. J. Srolovitz, Acta Metall. **37**, 621 (1989).
[4] B. J. Spencer, P. W. Voorhees, and S. H. Davis, Phys. Rev. Lett. **67**, 3696 (1991).
[5] J. Tersoff and F. K. LeGoues, Phys. Rev. Lett. **72**, 3570 (1994).
[6] Y. H. Xie, G. H. Gilmer, C. Roland, P. J. Silverman, S. K. Buratto, J. Y. Cheng, E. A. Fitzgerald, A. R. Kortan, S. Schuppier, M. A. Marcus, and P. H. Citrin, Phys. Rev. Lett. **73**, 3006 (1994).
[7] S. Stoyanov, Jpn. J. Appl. Phys. **30**, 1 (1991).
[8] D. Kandel and J. D. Weeks, Phys. Rev. Lett. **69**, 3758 (1992); Phys. Rev. B **49**, 5554 (1994).
[9] J. Ishizaki, S. Goto, M. Kishida, T. Fukui, and H. Hasegawa, Jpn. J. Appl. Phys. **33**, 721 (1994).
[10] C. Duport, P. Nozières, and J. Villain, Phys. Rev. Lett. **74**, 134 (1995).
[11] W. W. Mullins and J. P. Hirth, J. Phys. Chem. Solids **24**, 1391 (1963).
[12] R. L. Schwoebel, J. Appl. Phys. **40**, 614 (1969).
[13] J. Frohn, M. Giesen, M. Poensgen, J. F. Wolf, and H. Ibach, Phys. Rev. Lett. **67**, 3543 (1991).
[14] A. C. Redfield and A. Zangwill, Phys. Rev. B **46**, 4289 (1992).
[15] T. Marschner, S. Lutgen, M. Volk, W. Stolz, E. O. Göbel, N. Y. Jin-Philipp, and F. Philipp, Superlattices Microstruct. **15**, 183 (1994).
[16] Y. H. Phang, C. Teichert, M. G. Lagally, L. J. Peticolas, J. C. Bean, and E. Kasper, Phys. Rev. B **50**, 14 435 (1994).
[17] T. Kikkawa, K. Makiyama, H. Ochimizu, K. Kasai, and J. Komeno, J. Cryst. Growth **145**, 799 (1994).
[18] K. Hiramoto, T. Tsuchiya, M. Sagawa, and K. Uomi, J. Cryst. Growth **145**, 133 (1994).
[19] V. I. Marchenko and A. Ya. Parshin, Sov. Phys. JETP **52**, 129 (1980).
[20] O. L. Alerhand, D. Vanderbilt, R. D. Meade, and J. D. Joannopoulos, Phys. Rev. Lett. **61**, 1973 (1988). V. I. Marchenko, JETP Lett. **33**, 381 (1981)
[21] J. Tersoff, Phys. Rev. Lett. **74**, 4962 (1995); Y. H. Xie, G. H. Gilmer, C. Roland, P. J. Silverman, S. K. Buratto, J. Y. Cheng, E. A. Fitzgerald, A. R. Kortan, S. Schuppier, M. A. Marcus, and P. H. Citrin, Phys. Rev. Lett. **74**, 4962 (1995).
[22] F. Liu and H. Metiu, Phys. Rev. B **48**, 5808 (1995).
[23] K. Kandel and J. D. Weeks, Phys. Rev. Lett. **74**, 3632 (1995).
[24] C. Duport, P. Politi, and J. Villain, J. Phys. (to be published).