

Convective instability on a crystal surface

Navot Israeli and Daniel Kandel

Department of Physics of Complex Systems, Weizmann Institute of Science, Rehovot 76100, Israel

Michael F. Schatz and Andrew Zangwill

School of Physics, Georgia Institute of Technology, Atlanta, GA 30332

The distinction between absolute and convective instabilities is well known in the context of hydrodynamics and plasma physics. In this Letter, we examine an epitaxial crystal growth model from this point of view and show that a strain-induced step bunching instability can be convective. Using analytic stability theory and numerical simulations, we study the response of the crystal surface to an inhomogeneous deposition flux that launches impulsive and time-periodic perturbations to a uniform array of steps. The results suggest a new approach to morphological patterning.

PACS numbers:81.15.Aa, 05.45.-a, 47.20.-k

Instabilities dominate the behavior of many nonlinear systems. In hydrodynamics [1] and plasma physics [2,3], it is commonplace to distinguish between an *absolute* and a *convective* instability. In the first case, an initial perturbation spreads more rapidly than it advects and the system evolution is insensitive to subsequent perturbations. In the second case, an initial perturbation advects more rapidly than it spreads and the system evolution remains sensitive to subsequent perturbations. The latter characteristic presents an opportunity for control. In this Letter, we show that a strain-induced instability that can occur during epitaxial growth is convective and suggest a novel approach to morphological patterning based on the implied control.

We consider a regular, staircase-like surface composed of flat terraces of average width ℓ . The terraces are separated by straight, parallel, atomic height steps with horizontal positions x_n , where the index n grows in the direction of negative surface slope. We assume that atoms impinge on each lattice site at a rate F . This leads to a build up of a finite concentration of adatoms on the terraces. Adatoms diffuse on terraces and attach to the bottom of steps at a rate K . Atoms can also detach from steps towards neighboring terraces. These processes lead to step motion which can be described by simple equations [4]. If the flux is large enough, the steps acquire a net positive velocity, inducing vertical growth of the crystal by one atomic unit after every step has moved a distance of one terrace width.

The equations of motion for the steps are much more complicated if there are long-range interactions between steps. An example is the growth of a strained film where the lattice constant of the deposited material differs from the lattice constant of the substrate. The corresponding equations of motion [5] can be simplified if we assume that diffusion is fast. In this limit, the step velocities are given by

$$v_n = \frac{F}{2}(x_{n+1} - x_{n-1}) + \frac{K}{2}(\mu_{n+1} + \mu_{n-1} - 2\mu_n), \quad (1)$$

where μ_n is the chemical potential associated with adding an atom to the solid at the n th step. The μ_n are

$$\mu_n = \sum_{m \neq n} \left(\frac{\beta}{(x_m - x_n)^3} - \frac{\alpha}{x_m - x_n} \right), \quad (2)$$

where α reflects the attractive interaction arising from the elastic relaxation around each step on a strained layer, and β reflects the repulsion arising from the inherent stress of each step.

One solution to this model is uniform step-flow, where every step moves with velocity $F\ell$. Under certain circumstances, this steady state becomes unstable and groups of steps bunch together [5]. In this paper, we analyze the bunching instability from a new point of view.

The distinction between an absolute and a convective instability is most significant for a problem with at least one preferred frame of reference. For our problem, the *lab frame* is one such frame. We will also be interested in perturbing the step train by supplementing the uniform deposition flux with a very narrow beam of atoms that can be moved across the surface. This beam is at rest in the *source frame* of reference.

In the lab frame, the linear stability of uniform step-flow motion against a perturbation of the step positions $\delta x_n(t) = \epsilon \exp[i(n\ell q - \omega t)]$ leads to the dispersion relation

$$D_{lab}(q, \omega) = -i(\omega - F\ell q + F \sin \ell q) - 2K(\cos \ell q - 1) \sum_{m=1}^M (\cos m\ell q - 1) \left(\frac{\alpha}{m^2 \ell^2} - \frac{3\beta}{m^4 \ell^4} \right). \quad (3)$$

Here M is the number of neighbors a step interacts with on each side. In a general frame of reference, $D(q, \omega) = D_{lab}(q, \omega + qv_f)$, where v_f is the velocity of the frame of reference with respect to the lab frame. Conventional stability theory seeks the complex zeroes $\omega(q) = \omega_R(q) + i\omega_I(q)$ of $D(q, \omega)$ for given real q . $\omega_I(q) > 0$ is a sufficient condition for instability of uniform step-flow. Figure 1

shows $\omega_I(q)$ of our model for different values of α . The $q = 0$ mode is marginal for all values of α . When $\alpha > 0$ is small there are two additional marginal modes with $q = \pm q_m$. All the modes with $-q_m < q < q_m$ (except for $q = 0$) are unstable. When α is increased, q_m increases towards π/ℓ and the interval of unstable modes becomes wider.

To distinguish between different types of instability, we consider the long-time behavior of the mode with wave-number q^0 that has a zero group velocity: $\partial\omega_R(q)/\partial q|_{q=q^0} = 0$. By definition, the system is absolutely unstable if $\omega_I(q^0) > 0$ and convectively unstable if $\omega_I(q^0) < 0$. In physical terms, this is equivalent to examining the time-asymptotic behavior of a disturbance launched by an impulse-type (localized in space and time) perturbation. When the instability is absolute, the disturbance spreads in space more rapidly than it advects; an observer at any fixed position sees asymptotic growth since $\omega_I(q^0) > 0$. When the instability is convective, the disturbance advects more rapidly than it spreads; an observer at any fixed position may see transient growth as the disturbance advects by, but finds asymptotic decay since $\omega_I(q^0) < 0$.

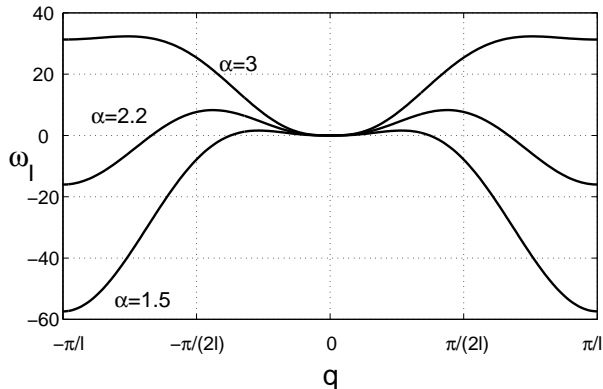


FIG. 1. $\omega_I(q)$ for real q and different values of α . The values of the other parameters in units where $\ell = 1$ are $F = 10$, $K = 6$, $\beta = 1$ and $M = 299$.

Figure 2 is a space-time plot of step trajectories (in the lab frame) determined by a numerical solution of the equations of motion with an impulsive perturbation applied at $t = 0$ to a single step. In the laboratory, a perturbation of this kind can be generated using the narrow beam source mentioned above [6]. The perturbation has no effect on uniform step flow in the portion of the surface labeled Region B in Fig. 2. However, in Region A, the perturbation creates a disturbance that spreads and amplifies in the direction of step flow. v_{min} and v_{max} are the minimal and maximal group velocities (in the lab frame) of unstable Fourier modes (for which $\omega_I(q) \geq 0$). As it happens, $v_{min} = 0$ for this model so the disturbance neither spreads out over the entire crystal nor advects away from the point where the impulse

was applied. In other words, step bunching as observed in the lab frame is “on the border” between absolute and convective instability. This fact can be used to test the step-bunching model experimentally because it does not depend on any of the model parameters. By contrast, the transition between absolute and convective instability in hydrodynamic systems typically occurs at a single value of the control parameter.

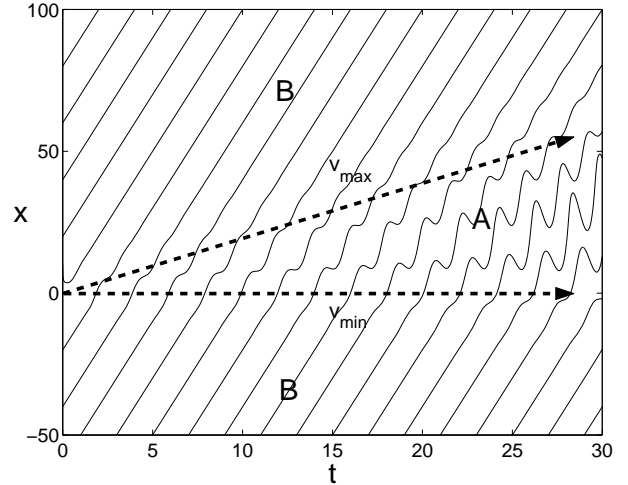


FIG. 2. Space time plot of a system of 300 steps with periodic boundary conditions after the application of an impulsive perturbation to a single step at $x = 0$ and $t = 0$. Each line shows the trajectory of a single step in the lab frame. Only a small portion of the system is plotted and the step motion is amplified. The choice of parameters for this specific system in units where $\ell = 1$ is $F = 10$, $K = 6$, $\alpha = 0.9$, $\beta = 1$ and $M = 10$.

We turn next to the response of the growing crystal to spatially localized but *time-periodic* perturbations produced by the narrow beam source. Such perturbations generate two types of asymptotic behavior which we will call *switch-on bunching* and *time-periodic bunching*. If the source moves with velocity v_s (in the lab frame) in the interval $v_{min} < v_s < v_{max}$, the system is absolutely unstable in the source frame and switch-on bunching occurs exclusively. For this situation, the step pattern develops analogously to the impulsive case (Fig. 2). However, if $v_s > v_{max}$ or $v_s < v_{min}$, the system is *convectively* unstable in the source frame. Switch-on bunching still occurs on one portion of the crystal surface, but, in addition, time-periodic bunching (which is sensitive to the nature of the forcing) *can* occur on an adjacent portion of the surface (Region C in Fig. 3).

To determine whether time-periodic bunching *does* occur in the regime of convective instability, it is sufficient to examine the asymptotic linear response of the step system to a spatially localized, time-harmonic source,

$$S_n(t) = \exp\left(-\frac{4[n\ell - (v_s - F\ell)t]^2}{a^2} - i\omega_s t\right), \quad (4)$$

where ω_s , a and v_s are the source frequency, width and velocity in the lab frame. This is called the *signaling problem* [1,3]. In the source frame ($n_s = n - (v_s - F\ell)t/\ell$) we find that

$$\delta x_{n_s}(t) \propto \exp(-i\omega_s t) \int_C \frac{\exp(in_s \ell q - a^2 q^2)}{D_{lab}(q, \omega_s + v_s q)} dq, \quad (5)$$

where D_{lab} is the dispersion relation (3) continued to the complex q plane and C is a suitable contour in this plane. The zeroes of $D_{lab}(q, \omega_s + v_s q)$ in the q plane dominate the integral. Among these, the most important zero corresponds to the single mode whose wave-vector $q^*(\omega_s)$ has a real part $q_R^*(\omega_s)$ in the interval $[-\pi/\ell, \pi/\ell]$ and an imaginary part $q_I^*(\omega_s)$ that can change sign as ω_s changes.

The main result is that there exists a critical frequency

$$\omega_c = |F \sin \ell q_m + q_m (v_s - F\ell)|. \quad (6)$$

If $|\omega_s| > \omega_c$, the amplitude of time periodic step bunching decays as the distance from the source increases. The source has little effect on the step-flow pattern in this case. However, if $|\omega_s| < \omega_c$, the source induces time-periodic step bunching that grows exponentially in space:

$$\delta x_{n_s}(t) \propto \frac{\exp[in_s \ell q - i\omega_s t - a^2 q^2]}{\left. \frac{dD_{lab}(q, \omega_s + v_s q)}{dq} \right|_{q=q^*(\omega_s)}}. \quad (7)$$

There are two cases to consider. If $v_s > v_{max}$, the disturbance grows in the direction opposite to step flow because $q_I^*(\omega_s) > 0$ (Region C of Fig. 3(a)). Conversely, if $v_s < v_{min}$, we find $q_I^*(\omega_s) < 0$ and the disturbance grows along the direction of step flow (Region C in Fig. 3(b)). Regions A and B correspond to switch-on bunching and uniform step flow similar to the corresponding regions in Fig. 2.

For a step-bunch that grows from a time-harmonic perturbation, there is very little nonlinear distortion of the bunch shape close to the source, as expected. Frequency spectra collected at different spatial locations in Region C show that higher harmonics contribute more as the distance from the source increases. Nevertheless, the amplitudes of the fundamental and all higher harmonics *saturate* for distances sufficiently far from the source.

The stabilizing influence of nonlinearity in the step-flow case prevents the system from wandering too far away from the linear character of the imposed perturbation. This provides an opportunity to exploit the bunching instability to intentionally pattern the crystal surface in Region C. The idea is to apply a perturbation prepared as a superposition of terms of the form (4) with frequencies in the range $|\omega_s| < \omega_c$. As long as nonlinear effects can be ignored, each of these terms evolves according to Eq. (7) in Region C. We can therefore tune the values of the amplitudes and phases of the various terms of the

perturbation, so that at a specific time the step configuration in Region C would be close to a pre-designed morphology.

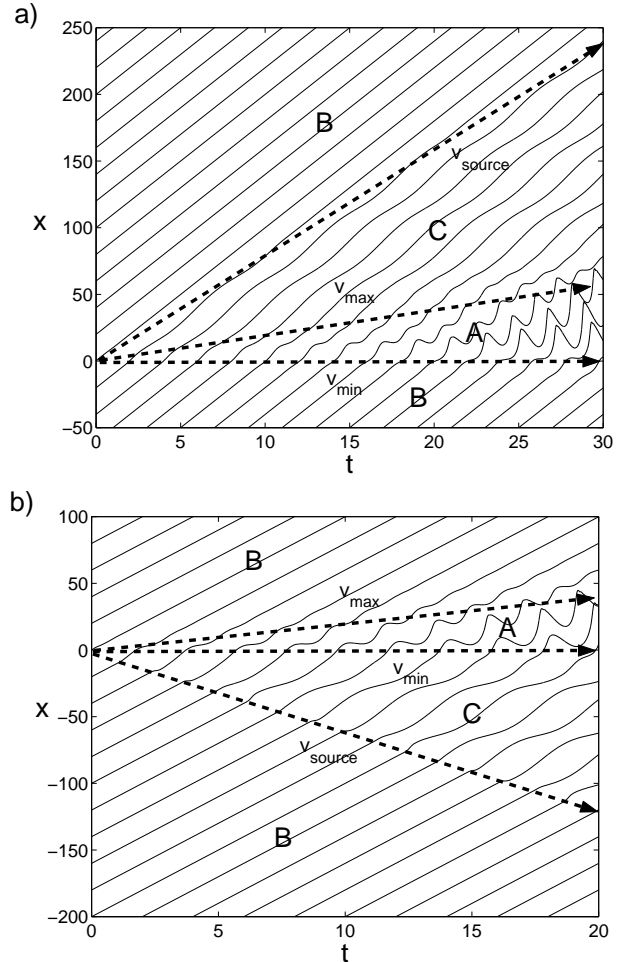


FIG. 3. Space time plots of systems of 300 steps with periodic boundaries perturbed by a narrow harmonic source. Each line shows the trajectory of a single step in the lab frame. Only a small portion of the system is plotted and the step motion is amplified. We have indicated the rays which correspond to the source velocity and the velocities v_{min} and v_{max} . When the source velocity $v_{source} > v_{max}$ and $|\omega_s| < \omega_c$, periodic step bunching is amplified in Region C in the direction opposite to step flow (a). When $v_{source} < v_{min}$ and $|\omega_s| < \omega_c$, periodic step bunching is amplified in Region C along the direction of step flow (b). The choice of parameters for these specific systems in units where $\ell = 1$ is $F = 10$, $K = 6$, $\alpha = 0.9$ and $\beta = 1$. The source width is $a = \ell$.

In order to demonstrate the applicability of this idea, we attempted to induce a groove-like pattern in a region which contained 125 steps. To construct this pattern, we used the linear analysis to optimize a small set of amplitudes and phases for waves with frequencies in the range $|\omega_s| < \omega_c$. We then numerically solved the step equations of motion with the designed source. The resulting surface height as a function of position is shown as circles

in Fig. 4. In Fig. 4(a) we have indicated regions analogous to Regions A, B and C of Fig. 3(a). Figure 4(b) is a magnification of the section marked by a two-sided arrow in Fig. 4(a). The region of 125 steps we attempted to pattern is marked. The solid line in Fig. 4(b) shows the predicted linear response of the surface to the designed source. Inside the patterned region it is very similar to the desired pattern. Near the source ($x/\tilde{\ell} = 1200$), the shape of the surface obtained from the numerical solution of the step equations of motion closely follows the linear dynamics. Further from the source, nonlinearity acts and we observe a regular sequence of grooves which are recognizably “echoes” of the original pattern for many periods. We have checked that this behavior is robust in the presence of deposition shot noise.

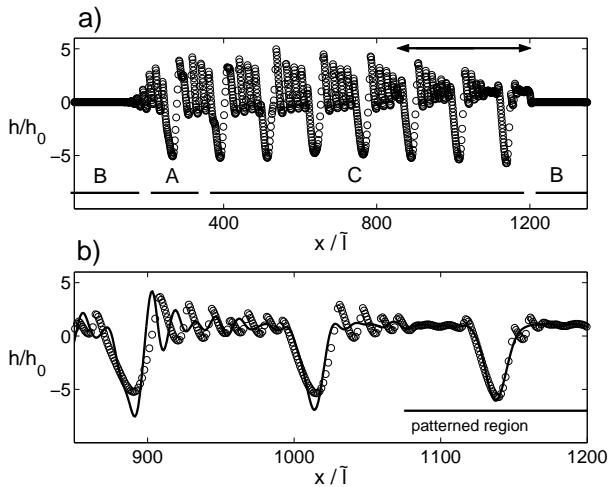


FIG. 4. Surface height (in units of the height of a single step, h_0) as a function of position evolving from a superposition of sinusoidal disturbances. $h = 0$ corresponds to the plane $x_n = n\ell$ and the unit of length is the average step separation on this plane, $\tilde{\ell} = \sqrt{\ell^2 + h_0^2}$. The source velocity is $v_s = F\ell$ and it is located at $x/\tilde{\ell} = 1200$. Regions A, B and C in (a) are analogous to the corresponding regions in Fig. 3(a). (b) is a magnification of the region marked by the two-sided arrow in (a). The solid line in (b) shows the predicted linear response, while the actual surface morphology, resulting from the solutions of the step equations of motion, is shown as circles.

In a typical experimental system the dispersion relation is not known. Nevertheless, one can in principle investigate the response of the step system to sources of different frequencies experimentally. For each frequency, one can measure the induced wavelength (q_R^*) and amplification rate (q_I^*), as well as the prefactor multiplying the exponential in Eq. (7). This information is sufficient for the implementation of the design procedure outlined above.

In summary, we have demonstrated the convective nature of a step-bunching instability that occurs in a recently proposed model of epitaxial, strain-induced, step-

flow growth. A variety of step bunching scenarios arise when conventional step-flow is perturbed by a beam of atoms whose flux can be controlled as a function of space and time. In particular, there is a regime of time-periodic bunching that can be used to launch a sequence of pre-designed step-bunch patterns. The nonlinearity of the model is such that the bunches do not distort appreciably as growth proceeds.

D.K. acknowledges the support of the Israel Science Foundation. M.F.S. is a Cottrell Scholar of the Research Corporation and gratefully acknowledges the support of the National Science Foundation under Grant CTS-9876590. A.Z. gratefully acknowledges the support of the Department of Energy under Contract DE-FG02-97ER45658.

-
- [1] C. Godreche and P. Manneville, *Hydrodynamics and Non-linear Instabilities* (University Press, Cambridge, 1998).
 - [2] M.N. Rosenbluth and R.Z. Sagdeev, *Handbook of Plasma Physics* (North-Holland, Amsterdam, 1983).
 - [3] E. M. Lifshitz and L. P. Pitaevskii, *Physical Kinetics*, (Pergamon Press, 1981).
 - [4] W.K. Burton, N. Cabrera, and F.C. Frank, *Philos. Trans. R. Soc. London, Ser. A* **243**, 299 (1951); R. Ghez and S.S. Iyer, *IBM J. Res. Develop.* **32**, 804 (1988).
 - [5] J. Tersoff, Y. H. Phang, Z. Zhang and M. G. Lagally, *Phys. Rev. Lett.* **75**, 2730 (1995); F. Liu, J. Tersoff and M. G. Lagally, *Phys. Rev. Lett.* **80**, 1268 (1998).
 - [6] Our main conclusions should remain valid even if many steps are perturbed in an actual experiment.