Convective instability of strained layer step-flow

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Abstract

We examine an epitaxial crystal growth model in the context of absolute and convective instabilities 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.

Keywords: models of non-equilibrium phenomena, models of non-linear phenomena, models of surface kinetics, growth, step formation and bunching, single crystal epitaxy

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Fifty years ago, Burton, Cabrera and Frank proposed their step-flow model for crystal growth onto a vicinal surface [1]. By now, it is well-verified that macroscopically flat steps can move parallel to themselves when deposited atoms diffuse across flat crystal terraces and attach to step edges. However, it is also understood that under suitable conditions, kinetic instabilities occur that lead to wandering and bunching of the steps as growth proceeds [2]. To uncover such instabilities, it is usual to ask whether the amplitude of an imposed periodic displacement of the steps grows or decays as time goes on. This was done, for example, in a recent interesting study of step bunching during strained layer growth onto a vicinal surface [3].

In this Letter, we generalize the analysis of Reference [3] and distinguish between an absolute step instability and a convective step instability. This type of distinction is widely appreciated in hydrodynamics [4], plasma physics [5], and the theory of dendritic crystal growth [6]. When an instability is absolute, an initial, localized perturbation spreads more rapidly than it propagates and the system is not sensitive to subsequent perturbations. When an instability is convective, an initial localized perturbation propagates more rapidly than it spreads and the system is sensitive to subsequent perturbations. In the present context, we will see below that this sensitivity presents an opportunity for morphological control at the nanoscale.

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 (the index n increases in the direction of negative surface slope). We assume growth conditions where a flux of atoms impinges 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 [1]. 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 [3] can be simplified if we assume that diffusion is fast. In this limit, the step velocities are given by

$$v_n = \frac{F}{2}\left(x_{n+1} - x_{n-1}\right) + \frac{K}{2}\left(\mu_{n+1} + \mu_{n-1} - 2\mu_n\right) \ . \tag{1}$$

The first term accounts for the deposited material each step collects from its neighboring terraces. The second term accounts for mass exchange between steps by detachment-attachment events. μ_n is the chemical potential associated with the addition of atoms to the solid at the nth step. It is given by

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

where α characterizes a step-step attraction due to local elastic relaxations and β characterizes a step-step repulsion due to intrinsic elastic stress.

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 [3].

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 $\delta x_n(t) = \epsilon \exp\left[i\left(n\ell q - \omega t\right)\right]$ leads to the dispersion relation

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

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 $\omega_I\left(q^0\right) > 0$ and convectively unstable if $\omega_I\left(q^0\right) < 0$. In physical terms, this is equivalent to examining the long time 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 propagates; an observer at any fixed position sees asymptotic growth since $\omega_I\left(q^0\right) > 0$. When the instability is convective, the disturbance propagates more rapidly than it spreads; an observer at any fixed position may see transient growth as the disturbance moves by, but finds asymptotic decay since $\omega_I\left(q^0\right) < 0$.

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. (Our main conclusions should remain valid even if many steps are perturbed in an actual experiment.) 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 propagates 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.

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 long time linear response of the step system to a spatially localized, time-harmonic source,

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

where ω_s, a and v_s are the source frequency, width and velocity in the lab frame. This is called the *signaling problem* [4]. In the source frame ($n_s = n - (v_s - F\ell)t/\ell$) we find that

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

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

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

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:

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

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 response 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.

In order to demonstrate 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.

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.

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Figure Captions

Figure 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: 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 $M = 10$.

Figure 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 $v_s > v_{max}$ and $|\omega_s| < \omega_c$, periodic step bunching is amplified in Region C in the direction opposite to step flow (a). When $v_s < v_{min}$ 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 $a = \ell$.

Figure 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.

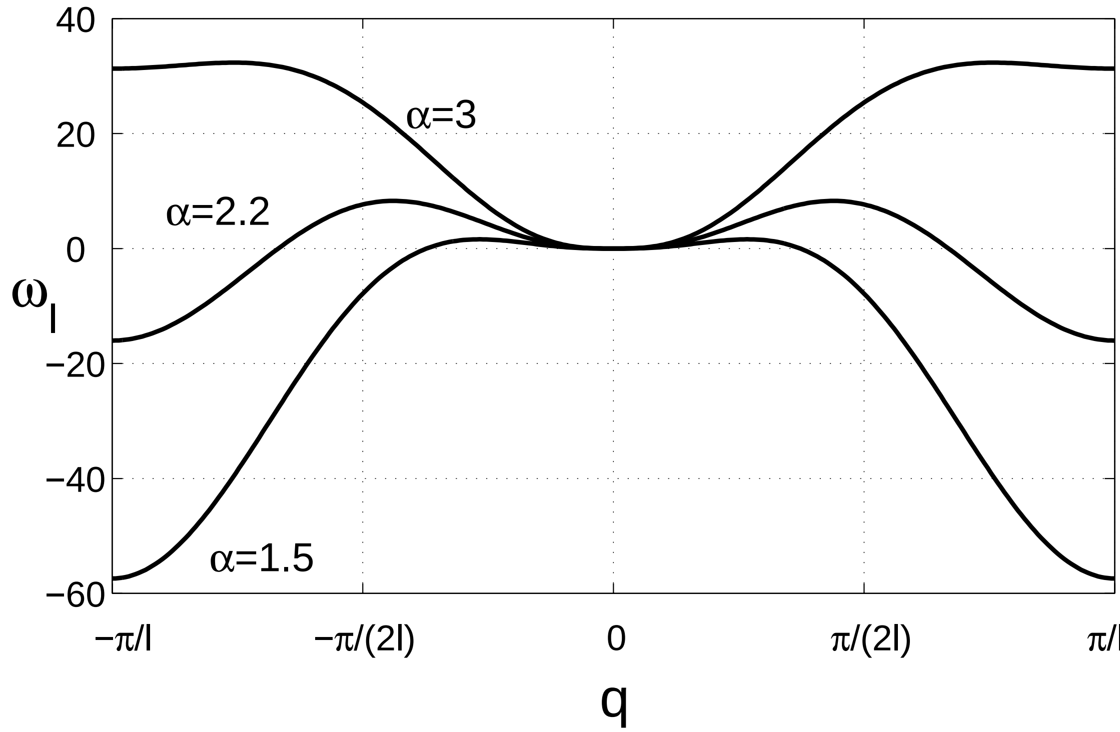


Figure 1: Israeli, *et al.*, Convective instability of strained layer step-flow

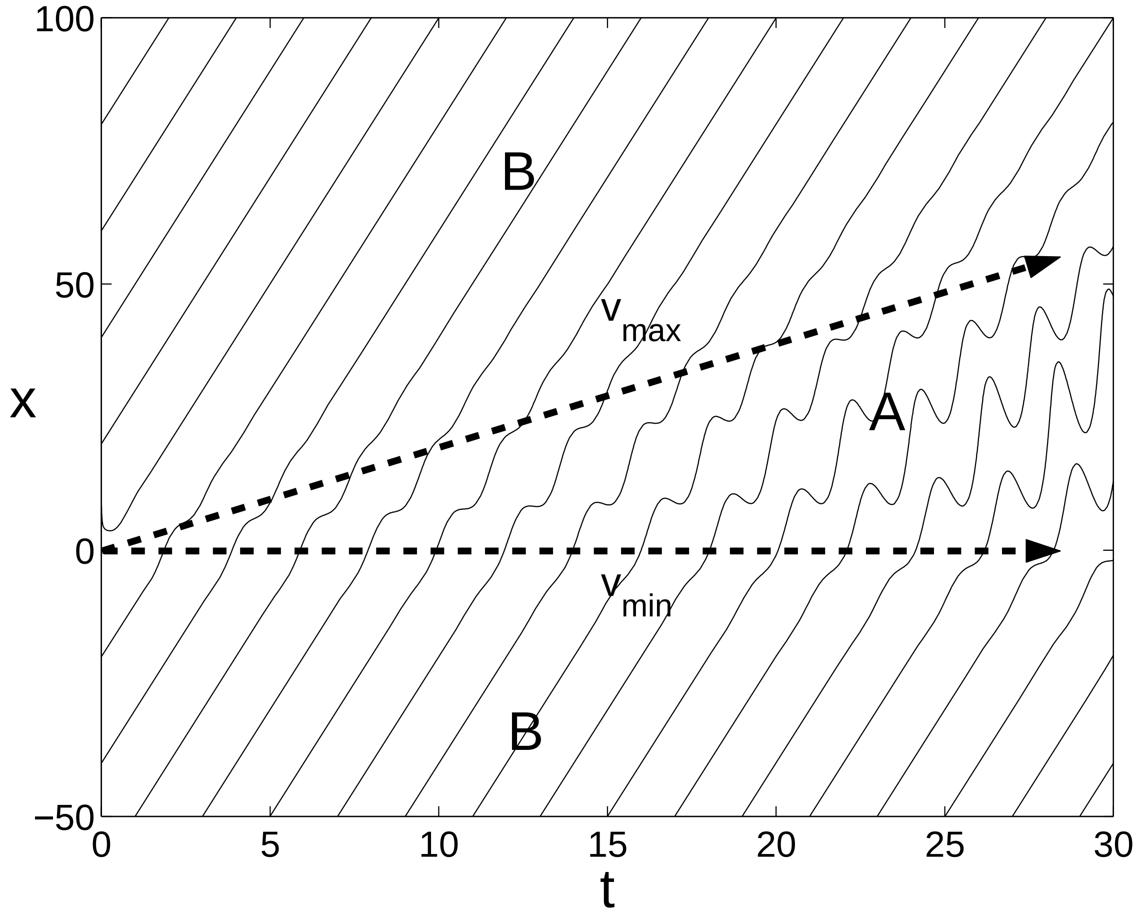
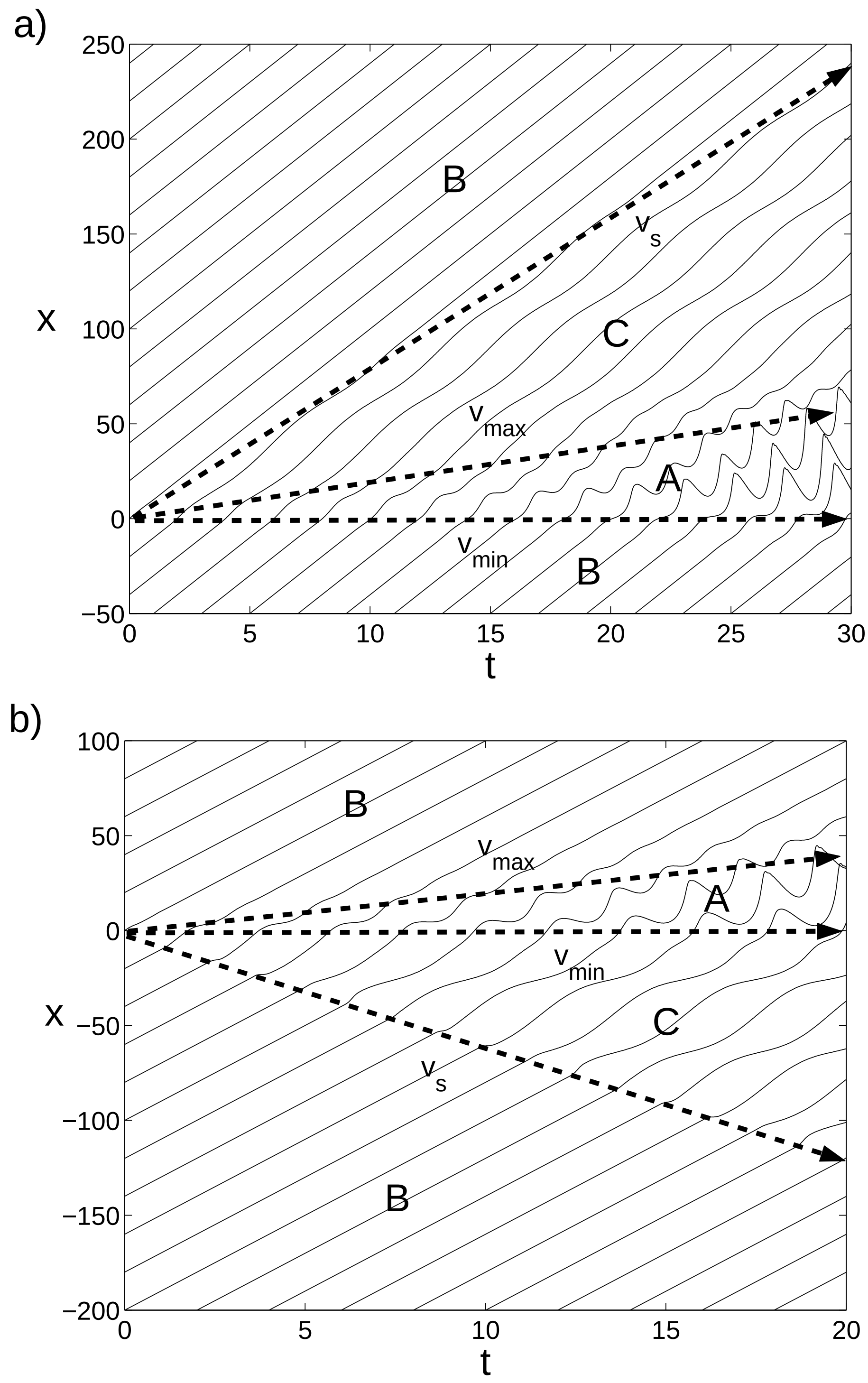


Figure 2: Israeli, *et al.*, Convective instability of strained layer step-flow



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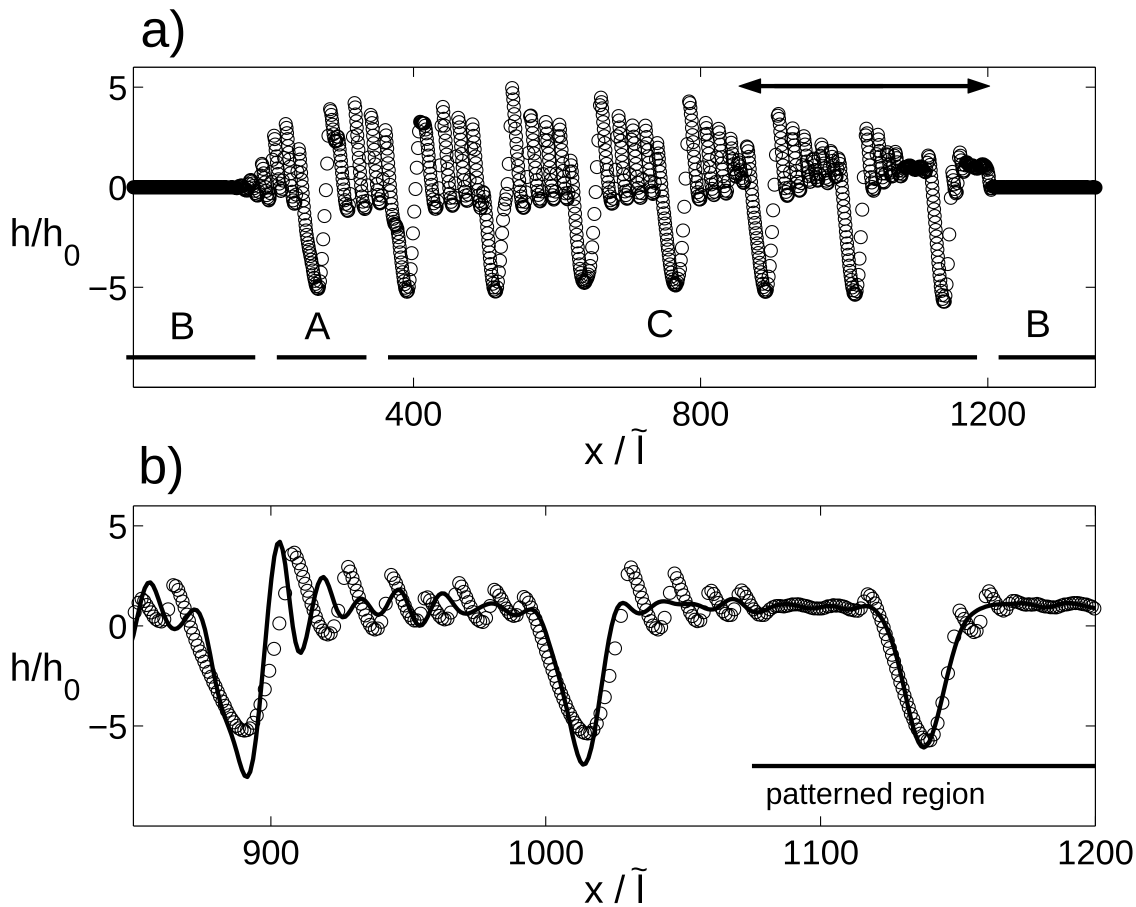

Figure 4: Israeli, *et al.*, Convective instability of strained layer step-flow