

## Crystallization kinetics and self-induced pinning in cellular patterns

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Within the framework of the Swift-Hohenberg model it is shown numerically and analytically that the front propagation between cellular and uniform states is determined by periodic nucleation events triggered by the explosive growth of the localized zero-eigenvalue mode of the corresponding linear problem. We derive an evolution equation for this mode using asymptotic analysis, and evaluate the time interval between nucleation events, and hence the front speed. In the presence of noise, we find the velocity exponent of “thermally activated” front propagation (creep) beyond the pinning threshold.

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Understanding the dynamics of localized structures in cellular patterns, such as dislocations, grain boundaries, and other defects, is a long-standing problem of pattern formation [1]. An important aspect of this problem is the propagation of a patterned state into a uniform state. This is a classical front propagation problem complicated by the fact that the patterned state provides potential barriers for the front, and close to the threshold leads to *self-induced* front pinning or stick-slip front motion. Near the threshold of a pattern-forming instability these effects can be studied within the framework of the generic Swift-Hohenberg (SH) model [1]. Despite relative simplicity of this model, and a serious limitation related to the fact that it is of the gradient type, it gives rise to a remarkably large variety of solutions. The SH equation has been intensively studied in the past as a paradigm for pattern formation in large aspect ratio systems [1,2]. More recent computations brought attention to the propagation of fronts between uniform stationary states of this equation and coarsening [3]. The computations have also demonstrated formation of stationary solitons, i.e., stable localized objects in the form of a domain of one phase sandwiched inside another phase [3]. The interest was supported by applications of the SH model to marginally unstable optical parametric oscillators [4,5]. The SH model has also served as a convenient testing tool for the problem of pattern propagation into an unstable trivial state [6,7]. In this Rapid Communication we elucidate another aspect of pattern formation in nonequilibrium media that can be modeled by the SH equation: self-induced pinning and stick-and-slip motion of the interphase boundary, which can be thought of as a particular kind of crystallization or melting.

We write the basic equation in the form

$$u_t = -(1 + \nabla^2)^2 u + \epsilon u - u^3. \quad (1)$$

At  $\epsilon > 0$  the trivial state  $u = 0$  undergoes a supercritical stationary bifurcation leading to a small-amplitude pattern with unit wave number  $k$ . The band of unstable wave numbers widens with growing  $\epsilon$ , until it reaches the limiting value  $k = 0$ , which signals the appearance of a pair of nontrivial

uniform states,  $u = \pm \sqrt{\epsilon - 1}$ . The two symmetric states are stable to infinitesimal perturbations at  $\epsilon > \frac{3}{2}$ . At still higher values of  $\epsilon$ , a variety of metastable states become possible: (1) a kink separating the two symmetric nontrivial uniform states [Fig. 1(a)]; (2) a semi-infinite pattern, coexisting with either of the two nontrivial uniform states; (3) a finite patterned inclusion, sandwiched, either symmetrically or anti-symmetrically, between semi-infinite domains occupied by nontrivial uniform states [8]; (4) an isolated solution [Fig. 1(b)].

The energy of the uniform state is higher than that of the regular pattern with the optimal wave number at  $\epsilon < 6.287$  [11]. In spite of the difference between the energies of the uniform and patterned state, the interface between them remains immobile at moderate and large values  $\epsilon$ , and a multiplicity of localized states is linearly stable. The pattern propagation into a metastable uniform state or the reverse “melting” process at  $\epsilon \gg 1$  is impeded by the *self-induced pinning* attributed to the oscillatory character of the

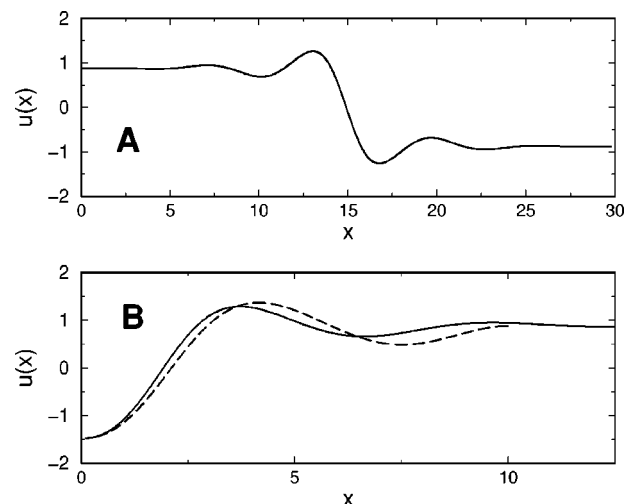


FIG. 1. Stationary solutions to Eq. (1) for  $\epsilon = 1.78$ . (a) kink solution connecting  $u = \pm \sqrt{\epsilon - 1}$ ; (b) 1D localized solution (solid line) and radially symmetric solution to Eq. (1) (dashed line).

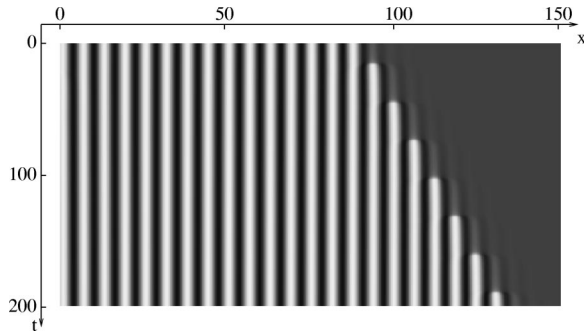


FIG. 2. Space-time plot of the large ( $N=29$ ) cellular cluster expansion into the stable uniform phase at  $\epsilon=1.757$  (only the right half of the cluster is shown).

asymptotic perturbations of the uniform state, characterized by the complex wave number  $k^2 = -1 \pm i\sqrt{2\epsilon-3}$ . As  $\epsilon$  decreases, the stationary localized solutions lose stability and give rise to propagating solutions.

We shall show below that the front propagation process can be described in terms of periodic nucleation events triggered by an explosive growth of the localized zero-eigenvalue mode of the corresponding linear problem. Using asymptotic analysis, we shall derive the evolution equation for this mode which allows us to estimate the time interval between the nucleation events and hence the front speed. We shall also study ‘‘thermally activated’’ front propagation (creep) at  $\epsilon > \epsilon_c$  and derive the creep velocity exponent.

We studied numerically Eq. (1) in one spatial dimension for various initial cluster sizes from a single ‘‘roll’’ to a semi-infinite roll pattern. As an initial condition we chose a section of a sine wave  $u = (-1)^N u_0 \cos x$  having  $N$  maxima within  $-N\pi < x < N\pi$ , surrounded by a uniform state  $u = u_0$ , where  $u_0 = -(\epsilon-1)^{1/2}$ . At large  $\epsilon \approx 1.8$ , this initial state quickly transforms into a stationary cluster of  $N$  ‘‘rolls.’’ Then we slowly decreased  $\epsilon$  to detect a transition from static to expanding cluster. The depinning transition for the single soliton occurs at  $\epsilon \approx 1.74$ . As  $N$  increases, the depinning threshold increases, rapidly converging to the limiting value  $\epsilon = \epsilon_c \approx 1.7574 \dots$  for the semi-infinite pattern. Figure 2 illustrates expansion of a large cellular cluster into a uniform stable state at  $\epsilon = 1.757$ . The front propagation takes the form of well separated in time periodic nucleation events of new ‘‘atoms’’ of the ‘‘crystalline’’ state at the front. Between successive nucleation events, the solution remains close to the stationary semi-infinite pattern found at  $\epsilon_c$ . This process resembles crystallization in equilibrium solids, with the important distinction that the new ‘‘atoms’’ are created directly from the metastable ‘‘vacuum’’ state. The time between consecutive nucleation events diverges as  $\epsilon$  approaches the pinning threshold [9]. Figure 3 presents the average front speed as a function of  $\epsilon - \epsilon_c$ . This function can be fitted by  $V = V_0 \sqrt{\epsilon_c - \epsilon}$ , with  $V_0 = 2.292$ .

Near the critical value  $\epsilon = \epsilon_c$  front dynamics can be analyzed within the framework of the perturbation theory. At  $|\delta| \ll 1$ , we can present the front solution as

$$u(x,t) = U_0(x) + |\delta|^{1/2} u_1(x,t),$$

where  $U_0(x)$  is the stationary front solution at  $\delta=0$  and  $|\delta|^{1/2} u_1(x,t)$  is a small correction. This solution is uniformly

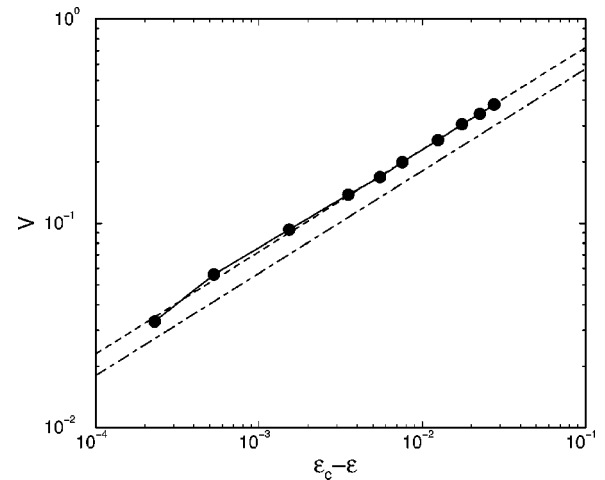


FIG. 3. Average front speed as a function of the control parameter  $\epsilon$  near the critical point  $\epsilon_c = 1.7574 \dots$ . Points represent numerics, the dashed line is the best fit  $V = 2.29(\epsilon_c - \epsilon)^{1/2}$ , and the dotted-dashed line is the theoretical prediction  $V = 1.80(\epsilon_c - \epsilon)^{1/2}$ .

valid at small positive  $\delta$ ; it is also valid during quasistationary phases (away from nucleation events) for small negative  $\delta$ . Plugging this ansatz in Eq. (1), we obtain

$$\partial_t u_1 = L[U_0]u_1 - |\delta|^{1/2} [3U_0 u_1^2 - \text{sgn}(\delta)U_0] + \dots, \quad (2)$$

where  $L[U_0] \equiv \epsilon_c - 3U_0^2 - (1 + \nabla^2)^2$  is the linearized SH operator at  $\epsilon_c$ . In the lowest order in  $\delta$ , Eq. (2) yields the linear equation  $\partial_t u_1 = L[U_0]u_1$ . This equation is always satisfied by the stationary translational mode  $U_0'(x)$ . In addition, the linearized operator  $L[U_0]$  has a localized neutral eigenmode  $U_1$  which we found numerically; see Fig. 4. Since all other eigenmodes have negative eigenvalues, the evolution of the system close to the bifurcation point can be reduced to single-mode dynamics [12],  $u_1(x,t) = a(t)U_1(x)$ , where  $a(t)$  is the amplitude of the zero mode.

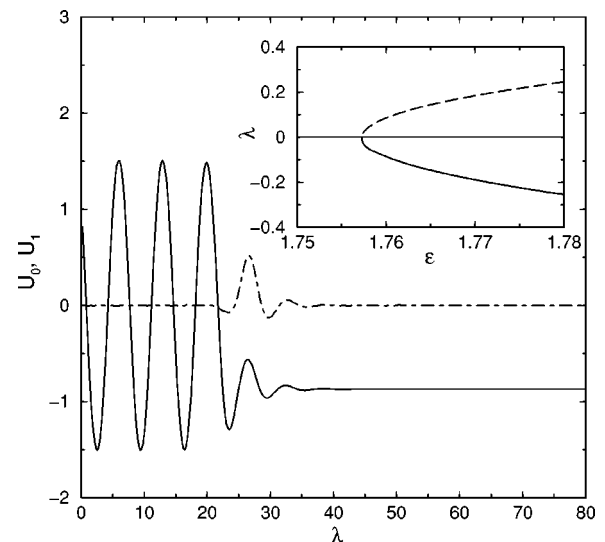


FIG. 4. Stationary structure  $U_0(x)$  (solid line) and the corresponding localized zero mode  $U_1(x)$  (dashed line) at  $\epsilon = \epsilon_c$ . Inset: eigenvalues of the stable and unstable localized modes of the stationary front solutions at  $\epsilon > \epsilon_c$ .

At  $\delta = \epsilon - \epsilon_c > 0$ , the immobile front solution  $u_0(x, \epsilon)$  is linearly stable. Numerical stability analysis [10] shows that the negative eigenvalue  $\lambda_s$  of the localized mode  $u_1(x, \epsilon)$  approaches zero as  $\delta^{1/2}$ . In addition, there is also an unstable front solution and a corresponding mode with positive eigenvalue  $\lambda_u$  which also approaches zero as  $\delta^{1/2}$ . At the pinning threshold, these two solutions collide and disappear via a saddle-node bifurcation. In the inset of Fig. 4, we show the stable and unstable eigenvalues  $\lambda_{s,u}$  as functions of  $\epsilon$ . At  $\epsilon < \epsilon_c$ , the front solution becomes nonstationary. Nonetheless, at  $|\delta| \ll 1$ , the solution remains close to the stationary front solution  $U_0(x)$  all the time except short intervals when a new roll nucleates.

Close to the bifurcation threshold  $\partial_t u_1 = \dot{a} U_1$  can be treated as perturbation. Therefore, in the second order we derive

$$L[U_0]u_2 = \dot{a} U_1 + |\delta|^{1/2} [3a^2 U_0 U_1^2 - \text{sgn}(\delta) U_0]. \quad (3)$$

Equation (3) has a bounded solution if its rhs is orthogonal to the zero mode  $U_1$  of the operator  $L$ . This results in the solvability condition for the amplitude  $a$ :

$$\alpha \dot{a} = |\delta|^{1/2} [\text{sgn}(\delta) \beta - \gamma a^2], \quad (4)$$

where

$$\alpha = \int_{-\infty}^{\infty} U_1^2 dx, \quad \beta = \int_{-\infty}^{\infty} U_1 U_0 dx, \quad \gamma = 3 \int_{-\infty}^{\infty} U_1^3 U_0 dx.$$

At  $\delta > 0$ ,  $a(t)$  reaches the stationary amplitude  $a_0 = (\beta/\gamma)^{1/2}$ . This value corresponds to the difference between the stable front solutions at  $\epsilon$  and  $\epsilon_c$ . At small *negative*  $\delta$ , Eq. (4) describes *explosive* growth of  $a$ , which passes from  $-\infty$  to  $\infty$  in a finite time  $\tau_e = \pi\alpha/(|\delta|\beta\gamma)^{1/2}$ . This explosion time gives an upper bound for the period between the nucleation events, after which the whole process repeats. The front speed is found as  $V = \Lambda/\tau_e$ , where  $\Lambda$  is the asymptotic spatial period of the pattern selected by the process of roll nucleation, and  $\tau_e$  is the time interval between nucleation events. Our calculations give the value  $V = 1.8|\delta|^{1/2}$ . This scaling is in a good qualitative agreement with the results of the numerical simulations; see Fig. 3. However, the prefactor 1.8 is noticeably lower (about 25%) than the corresponding value  $V_0 = 2.29$  obtained by numerical simulation of Eq. (1).

Let us discuss a possible reason for the discrepancy. Our numerical simulations show that the nucleation events produce slowly decaying distortions behind the moving front. These distortions may effectively “provoke” a consequent nucleation event by creating an initial perturbation of the zero mode  $U_1(x)$ . It will lead to an increase of the front velocity. Although we have evidence for the importance of this effect, a systematic treatment of this process is very complicated and goes beyond perturbation theory.

*Effect of noise.* Let us consider the effect of weak additive noise  $\eta(x, t)$  in the rhs of Eq. (1). For simplicity we assume that  $\eta$  is delta-correlated with the intensity (temperature)  $T$ :

$$\langle \eta(x, t) \eta(x', t') \rangle = T \delta(x - x') \delta(t - t').$$

In the presence of noise, there is no sharp threshold for the onset of motion at  $\epsilon < \epsilon_c$ . Instead, thermally activated mo-

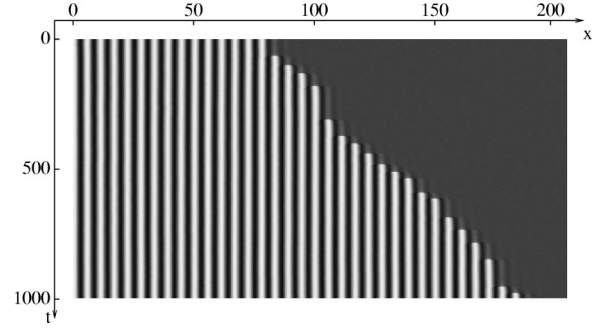


FIG. 5. Space-time plot of the switching wave propagation triggered by white noise with the temperature  $T=0.00042$  at  $\epsilon=1.758 > \epsilon_c$ .

tion (creep) occurs at  $\epsilon > \epsilon_c$  (see Fig. 5). In this case the average creep velocity is determined by the noise intensity. For  $\epsilon < \epsilon_c$  the noise will slightly increase the speed of the front. In contrast to the deterministic motion, the intervals between consecutive nucleation events are random.

In order to estimate the effect of the noise at  $\delta > 0$ , we will treat  $\eta$  as a small perturbation. In this analysis, we shall not introduce scaling explicitly, since the scaling of noise that is balanced with deterministic perturbations cannot be determined *a priori*. Following the lines of the above analysis, we project noise onto the zero mode to obtain the solvability condition

$$\alpha \dot{a} = \delta^{1/2} (\beta - \gamma a^2) + \delta^{-1/2} \tilde{\eta}(t), \quad (5)$$

where  $\tilde{\eta}(t) = \int_{-\infty}^{\infty} \eta(x, t) U_1(x) dx$ . For  $T=0$ , Eq. (5) has a stable fixed point  $a_s = \sqrt{\beta/\gamma}$  and an unstable one  $a_u = -\sqrt{\beta/\gamma}$ . At  $a < a_u$ , one has explosive growth of the solution to Eq. (5), while  $a \rightarrow a_s$  at  $a > a_u$ . Thus, we have to estimate the probability  $P$  for the amplitude of the zero mode  $a$  to be smaller than  $a_u$ . This quantity can be derived from the corresponding Fokker-Planck equation for the probability density  $p(a, t)$  [13]:

$$p_t = -\frac{\delta^{1/2}}{\alpha} \frac{\partial}{\partial a} [(\beta - \gamma a^2)p] + \frac{T}{2\alpha\delta} \partial_a^2 p, \quad (6)$$

where we used  $\langle \tilde{\eta}^2 \rangle = \alpha T$ . This is the standard Kramers problem. The stationary probability  $p(a)$  is given by

$$p \sim \exp \left[ \frac{2\delta^{3/2}(\beta a - \gamma a^3/3)}{T} \right]. \quad (7)$$

The probability  $P$  is given by the integral  $P = \int_{-\infty}^{a_u} p(x) dx$ . For  $T \ll \delta^{3/2}$ , we can use the saddle-point method, which gives the following result:

$$\begin{aligned} P(a < a_u) &\sim \exp \left[ -\frac{4\delta^{3/2}\beta^{3/2}}{3\gamma^{1/2}T} \right] \\ &= \exp \left[ -\frac{0.57\delta^{3/2}}{T} \right]. \end{aligned} \quad (8)$$

Since the time between the nucleation events  $\tau_n \sim 1/P$ , we find that the velocity of the front in the stable region is given by  $v \sim 1/\tau_n \sim \exp[-0.57\delta^{3/2}/T]$ .

At very large  $\epsilon > \epsilon_0 = 6.287$ , the flat state  $u = \pm \sqrt{\epsilon - 1}$  has lower energy than the periodic state. Nevertheless, the uniform state does invade the periodic state at any  $\epsilon$  because of the strong self-induced pinning. With large-amplitude noise there will be some probability for the flat state to propagate towards the periodic state by thermally activated annihilation events at the edge of the periodic pattern; however, for large  $\epsilon$  the probability of annihilation at the edge is of the same order as that in the bulk of the patterned state. Thus, for very large  $\epsilon$  and large  $T$  we may expect melting of the periodic structure both on the edge and in the bulk.

The above results can be trivially extended to regular two-dimensional periodic structures (rolls) selected by the SH equation [1]. More interesting is the behavior of a 2D hexagonal lattice which exists near the bifurcation of non-trivial uniform solutions at  $\epsilon = \frac{3}{2}$  [14]. We anticipate that propagation of the hexagonal structure into the uniform state will exhibit the same features of stick-and-slip motion as described above, and can be studied by similar methods. For the SH equation (1) at  $\epsilon > \frac{3}{2}$ , hexagonal lattices coexist with roll patterns which have lower energy, so the front propaga-

tion may in fact give rise to rolls. However, hexagons may become dominant in the modified SH equation with an added quadratic nonlinearity near  $\epsilon = 0$ , where the robust ‘‘crystallization’’ of the hexagonal lattice is expected. The work on this subject is now in progress.

Finally, we want to emphasize that although our results are obtained in the framework of the Swift-Hohenberg model, they can be applicable in a more broad context of pattern propagation into a uniform state. In particular, we recently observed and are currently studying the stick-slip motion of the interface between cellular and uniform states in experiments with thin vibrated layer of granular material [15].

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