Transition to Phase Chaos in Directional Solidification: A Two-Mode Interaction Picture

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Interface dynamics are analyzed in directional growth of liquid crystals in a two-mode interaction picture. We find that this simple model retains many interesting features observed experimentally including order and chaotic motion. We discover that the parity-broken mode undergoes an instability characterized by a permanent hopping between q and $2q$ states (where q is the wave number). On increase of the two-mode coupling the interface motion enters a chaotic regime via a quasiperiodicity route. Chaos manifests itself by an erratic change of the drift direction of the pattern between left and right. We tentatively call it "phase chaos."

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Since the discovery by Simon, Bechhoefer, and Libchaber [1] of the so-called "solitary mode" during directional growth of nematic crystals, the problem of interface dynamics has gained renewed interest. This type of growth is characterized by the existence of a few parity-broken (PB) cells that are approximately twice as wide as the symmetric ones and travel at constant speed transversely to the growth front. Soon after this discovery it became clear that this dynamical manifestation is common to a large variety of one-dimensional pattern-forming systems [2-6]. Another seemingly generic feature pertains to the so-called "vacillating-breathing" (VB) mode where the cell width oscillates in phase opposition with its neighbors [3,7-10]. On variation of external constraints the system often seems to develop an irregular pattern, an irregularity which was tentatively called chaos [3,5,7-10].

An important first step toward the understanding of the parity-broken state was made by Coullet, Goldstein, and Gunaratne [11] who suggested that parity breaking results from the loss of stability of the initially symmetric state. It was shown later that the "microscopic" models of growth do indeed support parity-broken solutions extending along, and moving transversely to, the whole front [12,13].

More recently [14] a more extensive analysis of interface dynamics has been made possible by focusing on the "large growth velocity" regime explored by experiments on liquid crystals [7]. Indeed, in this regime the boundary integral equation reduces to a local equation which naturally is much simpler for the analysis [15]. It has been shown [14] that this equation possesses symmetric steady solutions, PB and VB modes. An interplay between PB and VB modes causes the interface to enter a chaotic regime via quasiperiodicity. As mentioned in a previous article [14], the front profile is mainly composed of the fundamental and the first harmonic. This observation strongly suggests that a description in terms of two modes should be sufficient to capture the essential features of interface dynamics including chaotic behavior. Such a model would provide a much simpler tool for the understanding of the complex system dynamics in a certain region of phase space. The main purpose of this Letter is to deal with this question.

We find that besides parity-broken states, the two-mode interaction model accounts for the existence of an oscillatory mode that has not been discussed before. This new growth mode results from an instability of the PB state and is characterized by a permanent hopping between q and $2q$ states, while the cells globally drift sideways. For certain parameter values the two modes (the traveling mode and the oscillatory one) maintain their identity: The system behaves in a quasiperiodic manner. A change in the parameters leads to a mode-locking state before the dynamics enter a chaotic regime. Chaos manifests itself essentially as erratic behavior of the phase of the fundamental harmonic: The front erratically drifts to the left or to the right. This chaotic motion differs from the one discovered in [14]: There the second "oscillator" that mixes to the PB one is of VB type, while in the present case it is the one associated to the $q \to 2q$ hopping that couples to the PB mode. This supports the idea that the transition to chaos involving quasiperiodicity should be generic for systems with PB states, regardless of the specific nature of the second oscillator.

Our starting point is the evolution equation of the front profile $\zeta(x,t)$ derived in the quasilocal regime of directional growth of liquid crystals [15]:

$$3\zeta_{xxxx} - 4\zeta_{xxt} + \zeta_{tt} + 8\zeta_{xx} + 8 l_T^{-1}\zeta = 4\zeta_x\zeta_{xt} + 2\zeta_{xx}\zeta_t - 6\zeta_{xx}^2 - 6\zeta_{xx}\zeta_x^2 - 8\zeta_x\zeta_{xxx}\,. \quad (1)$$

The only parameter that remains in this equation, l_T^{-1}, is proportional to G/V, where G is the applied thermal gradient and V is the growth speed. The proportionality coefficient depends on material properties only [15].

In order to derive the coupled-amplitude equations from Eq. (1) we expand the interface position in Fourier series $\zeta(x,t) = A_0(t) + \sum_{n=1}^{\infty}[A_n(t)e^{iqnx} + \text{c.c.}]$. Inserting this expression into Eq. (1), we obtain a general equation for A_n:

$$L(nq)A_n(t)=\sum_m \{b(n-m,m)q^2A_{n-m}\dot{A}_m+c(n-m,m)q^4A_{n-m}A_m\}+\sum_{m,p}\{d(n-m,m)q^4A_{n-m-p}A_mA_p\}\,, \tag{2}$$

with

$$L(q)=\frac{d^2}{dt^2}+4q^2\frac{d}{dt}+(3q^4-8q^2+8l_T^{-1})\,. \tag{3}$$

b, c, and d are polynomials. Setting $n=1$ and $n=2$ into Eq. (2) we can write the following set of amplitude equations for A_1 and A_2:

$$L(q)A_1=\alpha_1A_1^*A_2+\beta_1A_1|A_1|^2+\gamma_1A_1|A_2|^2+\delta_1A_1\dot{A}_2 +\mu_1A_1\dot{A}_2A_2^*+\nu_1\dot{A}_1|A_2|^2+\rho_1A_1|\dot{A}_2|^2+\sigma_1\dot{A}_1A_2\dot{A}_2^*\,, \tag{4}$$

$$L(2q)A_2=\alpha_2A_1^2+\beta_2A_2|A_2|^2+\gamma_2A_2|A_1|^2+\delta_2A_1^*\dot{A}_1 +\lambda_2A_2^2\dot{A}_2^*+\mu_2\dot{A}_2|A_2|^2+\nu_2\dot{A}_2|A_1|^2+\xi_2A_1\dot{A}_1^*A_2+\sigma_2\dot{A}_1A_1^*A_2+\rho_2A_2|\dot{A}_1|^2+\sigma_2A_1\dot{A}_1^*\dot{A}_2\,. \tag{5}$$

The coefficients appearing in Eqs. (4) and (5) are related to the control parameter l_T^{-1} and to the wave number q, and are easily derived. It is important to mention that these coefficients have been renormalized by the higher harmonic amplitudes A_3 and A_4. Indeed, in order to perform a consistent expansion we must keep in the development of Eq. (2) all terms up to the third order in A (where $A\propto A_1,A_2$). Thus, as $A_3\propto A_1A_2$ and $A_4\propto A_2^2$, as a result of translational invariance, the terms like $A_2^*A_3$ and $A_2^*A_4$ appearing in Eq. (2) for $n=1$ and $n=2$, respectively, must be taken into account. A_0 obeys a detached equation, $8l_T^{-1}A_0=4q^4|A_1|^2+64q^4|A_2|^2$.

In order to study this two-mode interaction system it is useful to write the complex amplitudes A_1 and A_2 as $a_1e^{i\varphi_1}$ and $a_2e^{i\varphi_2}$, respectively. Substituting in Eqs. (4) and (5) and equating real and imaginary parts, we obtain a system of four coupled equations for the amplitudes a_1,a_2 and the phases φ_1,φ_2. The result emerging from the analysis of this system is summarized by the bifurcation diagram shown in Fig. 1. This diagram is a plot of the real amplitude a_2 against the wave number q for a given value of l_T^{-1}. The basic steady branches are the "mixed" modes (M^+ and M^-) with both a_1 and a_2 nonzero and the pure "period-doubled" solution P with $a_1=0$ and $a_2\neq0$. In addition, a parity-broken state solution merges from the branch M^+ at the locus where this one becomes linearly unstable. This type of diagram has been given already in [13], and its topology is quite generic.

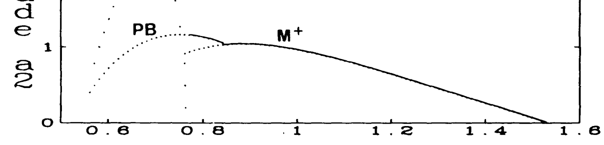

FIG. 1. Bifurcation diagram for $l_T^{-1}=0.28$ showing M^+, M^-, and P branches, and the parity-broken state solution (PB). Stable branches are represented by solid lines and unstable ones by dotted lines.

To check the relevance of the system described above, we have solved numerically the full equation (1) for symmetric and asymmetric states. We found that close to the codimension-two point (where both q and $2q$ state modes destabilize the flat interface simultaneously), which has the coordinates $l_T^{-1}=0.427$ and $q=0.730$, the two-mode interaction system is almost quantitatively and qualitatively accurate. As we move away from this point, the results differ quantitatively but remain qualitatively in agreement. Therefore, the overall picture found close to the codimension-two point should persist in a wide range of parameters.

An important question is whether the PB mode may itself undergo an instability and if so, what type of dynamics would then emerge. We have first analyzed the linear stability of the PB mode. We find that for $l_T^{-1}=0.28$ (not too far from the codimension-two point) the PB mode remains stable near the location of its birth ($q=0.85$). By decreasing the wave number q down to a value slightly below 0.76 the PB mode undergoes an oscillatory instability. The frequency of oscillations changes slightly with l_T^{-1} and is approximately of the order of $\mathrm{Im}(w)\propto0.8$.

In order to understand the long-time behavior of the growing instabilities we must perform a full nonlinear analysis of the coupled equations (4) and (5). To do so, we have solved these equations numerically. We use Gear's [16] backward difference method. This method has proven to be more appropriate than the usual Runge-Kutta method, and it is robust against the stiffness of the chaotic dynamics. We first concentrate on the situ-

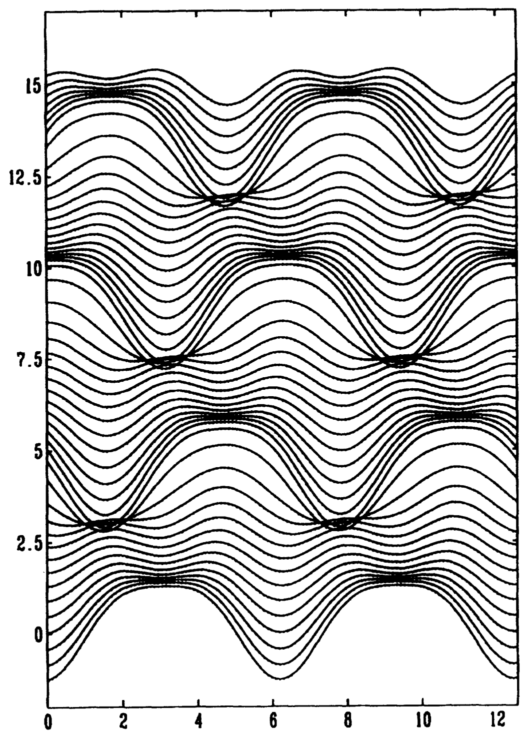

FIG. 2. Interface dynamics exhibiting both a temporal oscillation (the $q \rightarrow 2q$ hopping) and a global drift of the patterns due to parity breaking.

ation close to the onset of the oscillatory instability found from the linear analysis described above. Figure 2 shows the evolution of the interface for $l_T^{-1}=0.28$ and $q=0.75$ after transients have decayed. As we could expect from the linear analysis, the interface shows a temporal oscillatory instability. The pattern oscillates in time while the PB mode persists. The oscillatory mode (Fig. 2) is characterized by a permanent hopping of the front between a state with wave number q and a state with wave number $2q$. In other words the interface undergoes periodic tip splitting. We believe this mode of growth is the origin of the periodic droplet emission in liquid-crystal experiments [5,7]. In that situation the interface tip splits periodically. When the amplitude of oscillation is large enough the grooves pinch off, resulting in the detachment of droplets. Our suggestion is strongly supported by Oswald's experiments where he observes that this oscillatory regime first occurs in the tilted domain. Of course the eventual droplet formation occurs for a 2D interface; it is the well-known Rayleigh instability. It cannot therefore be accounted for in our 1D interface model.

Since the drift of the pattern subsists, each point on the front is now subject to the oscillation resulting from the drift with a frequency f_1, and to the $q \rightarrow 2q$ hopping (with a frequency denoted by f_2). Indeed the Fourier spectrum (Fig. 3) reveals sharp peaks at f_1 and f_2. All the other peaks are either higher harmonics or combinations of the form $|mf_1+nf_2|$ with m,n integers. The

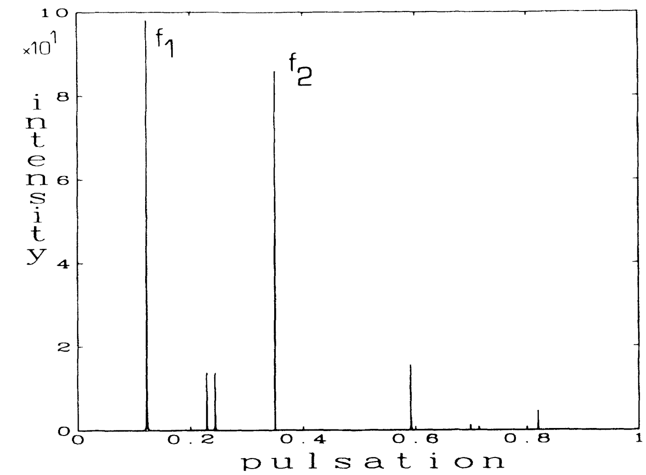

FIG. 3. Fourier spectrum in the quasiperiodic regime at $l_T^{-1}=0.28$ and $q=0.75$.

motion is quasiperiodic. The Poincaré map (not shown here) is dense, thus indicating that the trajectories in phase space cover the torus densely. Then by decreasing the value of q—which amounts to increasing the importance of the $2q$ mode—the interface develops the typical dynamics of a quasiperiodic system. More precisely, at $q=0.745$ a mode-locking state of order 3 appears. The Poincaré map reduces to three distinct points. The interface in the mode-locking situation does not, however, look fundamentally different from the incommensurate case. Upon variation of the wave number the motion again becomes quasiperiodic but with a much higher density of trajectories in phase space about the previously mode-locked state of order 3. At $q=0.743$ the interface motion enters a chaotic regime, via a destruction of the torus. The chaotic dynamics are characterized by an erratic drift of the pattern between left and right. The spectrum forms a continuum for low frequencies (Fig. 4).

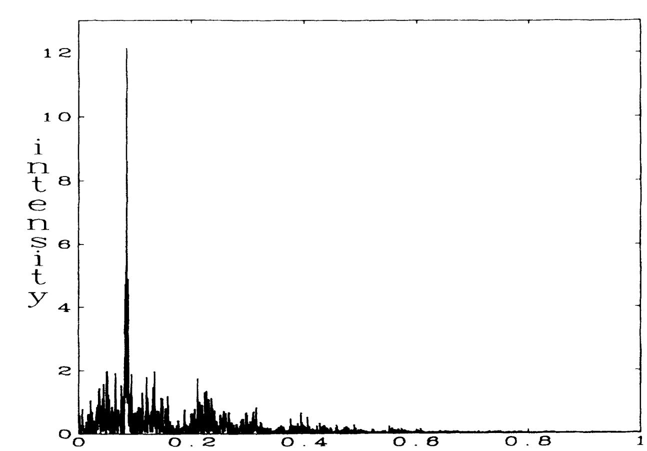

FIG. 4. Fourier spectrum in the chaotic regime at $l_T^{-1}=0.28$ and $q=0.743$.

Note that since Eqs. (4) and (5) are of second order in time (and that they are both complex), the total number of degrees of freedom is equal to 8. However, because of the translational invariance, φ_1 and φ_2 are not independent; the appropriate variable is $\theta = 2\varphi_1 - \varphi_2$. Therefore the number of degrees of freedom is in fact 7. By inspecting the dynamics of the different degrees of freedom, we find that $a_1(t)$ and $\dot{a}_1(t)$ are "regular" functions of time in the chaotic regime, while the rest are erratic. That is to say, the number of degrees of freedom that are involved in the chaotic regime is equal to 5. In other words $a_1(t)$ and $\dot{a}_1(t)$ simply act as parametriclike excitations. We hope to report on details of this question in the future.

A surprising feature is that, although the other degrees of freedom $(a_2,\dot{a}_2,\varphi_1,\varphi_2,\dot{\varphi}_1,\dot{\varphi}_2)$ are chaotic in time, the combination $a_2\cos(\varphi_2)$ [and consequently $a_2\sin(\varphi_2)$] is completely periodic in time. Since the front profile in a two-mode model can be written as

$$\zeta = 2a_1\cos(qx+\varphi_1) + 2a_2\cos(\varphi_2)\cos(2qx) - 2a_2\sin(\varphi_2)\sin(2qx),$$

only the first term is chaotic via the phase φ_1. Figure 5 shows the behavior of $\dot{\varphi}_1$ (the derivative suppresses the constant drift). We do not have at present an explanation of this "particular" behavior where all information on chaos is solely supported by the phase φ_1. It is natural to refer to this type of dynamics as "phase chaos." It is an important task for future investigations to see whether the understanding of the present chaos can be achieved with fewer degrees of freedom.

In summary, we have shown that a simple two-mode interaction model accounts for a large variety of dynamical behaviors. The parity-breaking instability has been discussed before [13]. However, the existence of an oscillatory tip-splitting instability is one of the new aspects of this model. Another interesting property of the model is that it supports chaotic solutions. A striking feature is that generically the systems possessing PB modes seem to transit, when coupled to another oscillator, into chaos via quasiperiodicity. We believe that this feature is analogous to the one encountered in the Taylor-Couette system [17]. In that system, the incommensurate character of the two oscillators can be traced back to the underlying symmetries. We hope that our work will incite new experiments with the aim to carefully study the possible transition into chaos.

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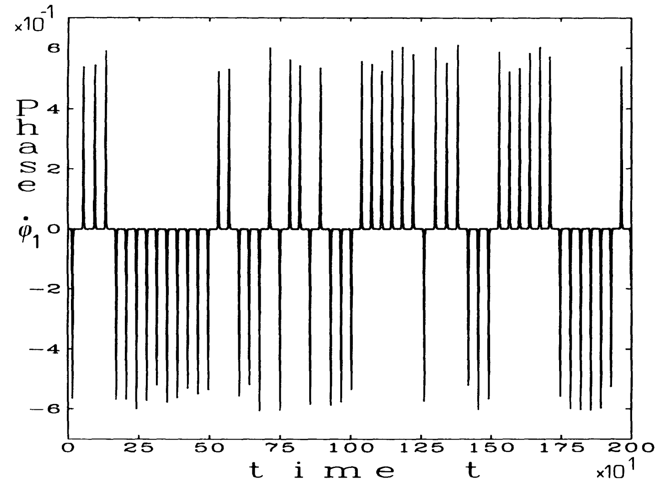

FIG. 5. Temporal evolution of the phase $\dot{\varphi}_1$ in the chaotic regime.

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