

Resonant interactions and traveling-solidification cells

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Recently a set of new phenomena has been observed in the directional solidification of liquid crystals, which include solitary inclusions of traveling waves and spatial period doubling. We propose that these patterns can be naturally explained as arising from the resonant interaction of modes with wave vectors q and $2q$. This picture is substantiated by numerical calculations employing the material parameters of the liquid-crystal system of Simon, Bechhoefer, and Libchaber [Phys. Rev. Lett. **61**, 2574 (1988)]. Our approach correctly predicts period-doubled (mixed) modes and the parity-breaking bifurcation to the traveling-wave state. Possible causes for the disagreement of some stability assignments as compared with the experiments are sketched.

The study of spatial patterns in nonequilibrium systems is becoming increasingly popular. One reason for this activity is the recognition that certain features of pattern-forming systems can be explained within a general framework by identifying universal mechanisms which depend essentially on the symmetries of the underlying physical systems. This approach allows the correlation of experiments in systems as diverse as alloy solidification, convection, and parametrically excited waves. Amplitude equations for the relevant degrees of freedom of the macroscopic system play an important role in exploiting these general concepts.

Within the context of solidification, recent experiments by Simon, Bechhoefer, and Libchaber¹ have exhibited an entirely new phenomenology. By using the nematic-to-isotropic transition in liquid crystals, they are able to explore a parameter range not accessible in experiments using other materials. They discovered an interesting transition from a spatially periodic, stationary pattern to one with solitary inclusions which propagate through this stationary pattern. For larger wave numbers they find that the pattern undergoes a period-doubling transition in space. For still larger wave numbers, the ensuing steady pattern becomes unstable with respect to an oscillatory instability in time. Subsequently, some of these patterns have also been seen in the "printer instability" of the meniscus of a fluid between two rotating cylinders.²

In a recent Letter, Coulet, Goldstein, and Gunaratne³ provide a natural explanation for the propagating inclusions. They postulate the existence of a secondary bifurcation which breaks the reflection symmetry of the original periodic steady-state pattern. If such a parity-breaking bifurcation is subcritical, kinks connecting the reflection-symmetric with the -asymmetric state can arise. A kink-antikink pair can propagate through the pattern exhibiting all qualitative features observed in the experiment.

In this Rapid Communication, we propose that all three phenomena, the parity-breaking bifurcation, the transition to a period-doubled state as well as its oscillatory instability, arise quite generally in pattern-forming systems through the resonant interaction of modes with wave numbers q and $2q$. We show by explicit numerical calcu-

lation the existence of the period-doubled state as well as of the aforementioned parity-breaking bifurcation in a microscopic model for directional solidification. We interpret these findings using previous results from the bifurcation theory of such resonating modes.⁴⁻⁷ This allows us to give all essential features of the experimental phase diagram along with a prediction for an additional transition. At the same time, a unified picture for the origin of these transitions is obtained which applies to a whole class of pattern-forming systems.

If the pulling speed v and the wave number q are close to the codimension-two point (q_{CT}, v_{CT}) , where both modes destabilize the flat interface simultaneously, the resonant interaction of two modes with wave numbers q and $2q$ can be described by two coupled amplitude equations. In the presence of translation and reflection symmetry these equations read

$$\dot{z}_1 = az_1 + c_1 z_1^* z_2 + a_1 z_1 |z_1|^2 + b_1 z_1 |z_2|^2 + \text{higher-order terms}, \quad (1)$$

$$\dot{z}_2 = \beta z_2 + c_2 z_1^2 + a_2 z_2 |z_1|^2 + b_2 z_2 |z_2|^2 + \text{higher-order terms}. \quad (2)$$

The pattern (interface position) ζ is given in terms of the complex amplitudes z_1 and z_2 by $\zeta(x, t) = z_1(t)e^{iqx} + z_2(t)e^{2iqx} + \text{c.c.} + \text{higher-order terms}$, and a and β are related to the control parameter v and the wave number q through $a = \mu(v - v_{CT}) + \kappa(q - q_{CT})$, $\beta = \mu'(v - v_{CT}) - \kappa' \times (q - q_{CT})$. The coefficients in Eqs. (1) and (2) can be calculated by performing a weakly nonlinear analysis of the microscopic equations. The results of this calculation will be published elsewhere.⁸ An independent calculation has been performed by Haug.⁹

These amplitude equations have been studied previously by various authors.⁴⁻⁷ A typical (schematic) bifurcation diagram arising from these analyses^{6,7} is shown in Fig. 1(a) for the case $c_1 c_2 < 0$. The basic steady-state solution branches are the mixed mode S_+ with $(z_1, z_2) = (z_1^+, z_2^+)$ which bifurcates from the neutral curve for large values of the wave number q and which corresponds to the solution

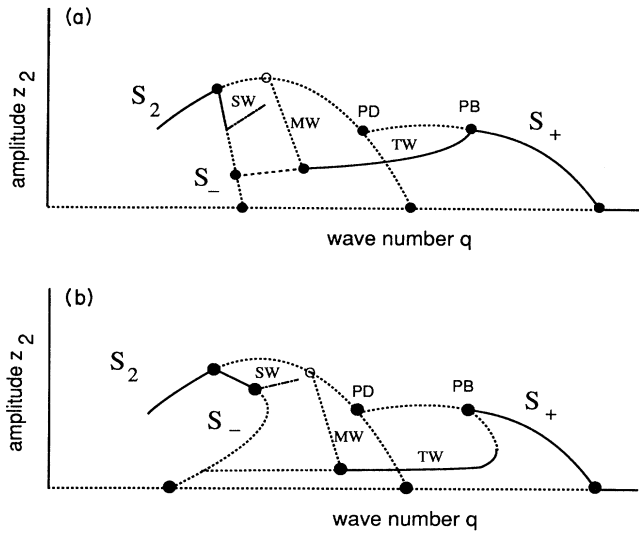


FIG. 1. (a) Typical bifurcation diagram for a $(q, 2q)$ -mode interaction for $c_1 c_2 < 0$. Solid lines represent stable solutions and dashed lines represent unstable solutions. For an explanation for the various symbols we refer to the text. (b) Proposed bifurcation diagram yielding traveling inclusions and stable period-doubled states.

commonly observed in the experiments, and the mixed mode $S_-(z_1^-, z_2^-)$ which bifurcates from the neutral curve for small q , and the pure mode $S_2(0, z_2^{(2)})$. It is crucial for our discussion to note that a pure mode S_2 with wave number q can be identified with a mixed mode S_+ with wave number $2q$. The transition from S_2 to S_- at period-doubled state (PD) constitutes a period-doubling bifurcation. In addition a traveling-wave (TW) solution $(z_1^{TW} e^{i\Omega t}, z_2^{TW} e^{2i\Omega t})$ bifurcates off S_+ in a parity-breaking (PB) bifurcation and a standing wave (SW) $(z_1^{SW}(t), z_2^{SW}(t))$ bifurcates from the S_- in a Hopf bifurcation. Finally a modulated wave (MW) can connect TW and S_2 . Note that the bifurcation to the traveling waves is supercritical close to the codimension-two point.

To show the relevance of the above analytical results to the phenomena of interest here, we have solved numerically the usual microscopic equations describing the directional solidification system investigated by Simon *et al.*¹ It is crucial to realize that these calculations described below are *not* performed close to the codimension-two point; they provide evidence that the states found close to this point, in fact, persist at a wide range of parameters and govern many of the interesting physical states seen in the experiment.

Using the integro-differential method¹⁰ we have calculated the nonlinear solutions for $v = 2 \mu\text{m/s}$ and examined their stability by perturbing the interface and solving the resulting eigenvalue problem.¹¹ The material parameters used in these calculations are detailed in Table I. Note that since $(v - v_{CT})/v_{CT} = 0.2$ we are not close to the codimension-two point. The main results of this calculation are given in Fig. 2(a), which shows the amplitude of the interface deformation as a function of the wave number for the steady-state solutions. Stable solutions are

TABLE I. Parameter values of the liquid-crystal system used in the numerical calculation.

$d_0 = 2 \times 10^{-8} \text{ cm}$
$T_M = 313.65 \text{ K}$
$m_l = 1^\circ/\text{mol}\%$
$c_\infty = 1.2 \text{ mol}\%$
$D_l = 3.8 \times 10^{-7} \text{ cm}^2/\text{s}$
$D_s = D_l$
$k = 0.88$
$G = 23 \text{ K/cm}$
$q_{CT} = 5.75 \times 10^{-2} \mu\text{m}^{-1}$
$v_{CT} = 1.66 \mu\text{m/s}$

denoted by solid, unstable solutions by dashed lines. Similar to Fig. 1(a) it shows the existence of the mixed mode S_- branching off S_2 . Figure 2(b) gives S_- in detail and clearly demonstrates its period-doubled structure in space. Because of the identification of S_2 with S_+ this establishes the existence of a period-doubling transition off the regular pattern for large wave numbers. This transition point is also indicated in Fig. 1(a). Using a stability analysis which also allows antisymmetric perturbations we find that there is indeed a parity-breaking bifurcation from the S_+ branch to traveling waves. [Our method places the solution (and any perturbations thereof) into a computational box of one-half the physical wavelength, $0 \leq x \leq \lambda/2$. Possible parity-breaking bifurcations are *antisymmetric* around $z=0$ and also around $z=\lambda/2$. These modes can be found in a straightforward generalization of the stability equations derived in Ref. 11; there is even a simple check of the computation since translation invariance (parallel to the front) ensures that there is an antisymmetric mode at zero growth rate.] Its position is denoted by PB in Fig. 2. (Meanwhile, we have obtained numerical solutions for these traveling waves.¹²)

The experimental results can now be explained quite naturally if two additional assumptions are made. To ob-

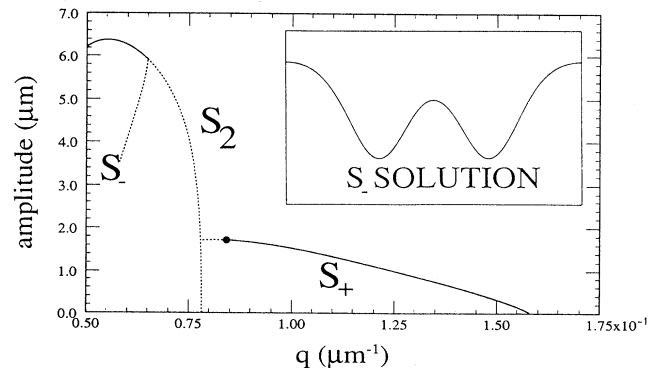


FIG. 2. (a) Numerical bifurcation diagram at $v=2$ showing the S_- , S_+ , and S_2 branches and the location of the parity-breaking bifurcation (dot). As in Fig. 1, solid lines represent stable solutions and dashed lines represent unstable solutions. Bifurcation is represented by a solid circle. The inset shows a typical period-doubled (S_-) solution.

tain traveling inclusions within the framework proposed by Coulet *et al.*³ the parity-breaking bifurcation of the TW off S_+ has to be backward in the experimental regime. Thus it would have to turn when increasing the pulling speed. Such a behavior is not uncommon and has been found in the Taylor vortex flow⁷ where it is found that a similar bifurcation analysis, which is strictly valid only close to the codimension-two point, organizes the numerically obtained stability diagram over a wide range of Reynolds numbers and wave numbers. Stable period-doubled states are obtained if the bifurcation of S_- off S_2 is forward, whereas our numerical calculations give a backward bifurcation. It is, however, reasonable to expect that various assumptions entering the microscopic equations have a strong influence on this feature. Foremost, the experiments were performed in very thin samples whereas our calculations assume a two-dimensional geometry. This is supported by the strong dependence of the observed states on the sample thickness.^{13,14} In addition, the capillary length of this system is not very well known. The effect of varying the capillary length on the stability is presently under investigation. Finally, there might be complicating features due to the use of liquid crystals. In particular, the interface elasticity appears to give a much larger microscopic cutoff length on interface deformations than the chemical capillary length.¹⁴ These effects might also explain the quantitative discrepancies between the numerically obtained transition points for PD and PB and those observed in the experiments.

With these additional assumptions one obtains the bifurcation diagram sketched in Fig. 1(b). In addition to the traveling inclusions and the period-doubled state it also shows the oscillatory instability of the period-doubled state to standing waves. Note that this instability does not saturate in the vicinity of the codimension-two point.⁶ We conjecture that this instability is connected to the oscillatory phenomena observed in the experiments. However, this last part must await a more detailed understanding of the time dependence observed in this parameter regime. Figures 1(a) and 1(b) taken together yield the phase diagram shown in Fig. 3, which sketches the regions in which traveling inclusions, period-doubled states, and the oscillatory instability are to be found. The bifurcation diagrams of Figs. 1(a) and 1(b) correspond to traversing this phase diagram at the pulling speeds indicated by the open and solid arrows, respectively. Note that for small pulling speeds this analysis predicts a transition from the steady pattern directly to traveling waves *without* the appearance of localized traveling inclusions. This can be checked experimentally. We believe that such a diagram can be matched to the experiments at each value of the channel thickness, providing a coherent picture of the possible nonlinear patterns.

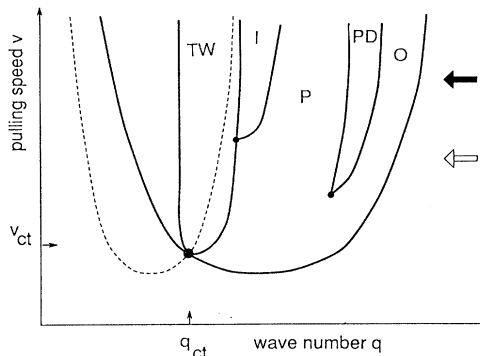


FIG. 3. Phase diagram: P, periodic state; PD, period-doubled state; I, inclusions; TW, traveling waves; O, oscillatory instability. The outer solid line is the neutral stability curve for the q mode and the dotted line is the neutral stability curve for the $2q$ mode.

In conclusion, we have argued that the general structure of the nonlinear states seen in directional solidification is due to a resonant mode interaction. Close to the codimension-two point this interaction can be treated analytically using amplitude equations. Our numerical calculation of the microscopic equations shows that two key features of the bifurcation diagrams—the period-doubled state and the parity-breaking bifurcation—can be followed beyond the validity of the amplitude equations into the experimentally relevant regime. Because of its general nature, this bifurcation analysis also elucidates the origin of similar phenomena in other systems.^{2,15} We have not discussed the influence of long-wavelength perturbations on the stability of the patterns. In other systems⁷ such perturbations often obscure the transitions which are of interest here. For the present purpose, however, this may be of minor importance since such diffusive instabilities involve much longer time scales which may not be observable in the experiments in question. Nevertheless, this question requires further work, both theoretical and experimental. A quantitative comparison of the numerical calculation with experimental results will probably require a more sophisticated model, taking into account three-dimensional phenomena and liquid-crystal physics.

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¹A. Simon, J. Bechhoefer, and A. Libchaber, *Phys. Rev. Lett.* **61**, 2574 (1988).

²M. Rabaud, S. Michalland, and Y. Couder, *Phys. Rev. Lett.* **64**, 184 (1990).

³P. Coulet, R. E. Goldstein, and G. H. Gunaratne, *Phys. Rev. Lett.* **63**, 1954 (1989).

⁴L. H. Ungar and R. A. Brown, *Phys. Rev. B* **29**, 1367 (1984).

⁵G. Dangelmayer, *Dyn. Stab. Syst.* **1**, 159 (1986).

⁶C. A. Jones and M. R. E. Proctor, *Phys. Lett. A* **121**, 224 (1987).

⁷H.-G. Paap and H. Riecke, *Phys. Rev. A* **41**, 1943 (1990).

⁸H. Levine, W. J. Rappel, and H. Riecke (unpublished).

⁹P. Haug, *Phys. Rev. A* **40**, 7253 (1989).

¹⁰H. Levine and D. Kessler, *Phys. Rev. A* **39**, 3041 (1989).

¹¹D. Kessler and H. Levine, *Phys. Rev. A* **41**, 3197 (1990).

¹²H. Levine and W.-J. Rappel, *Phys. Rev. A* **42**, 7475 (1990).

¹³J. Bechhoefer, Ph.D. thesis, University of Chicago (unpublished).

¹⁴A. Simon (private communication).

¹⁵G. Faivre, S. deCheveigne, C. Guthmann, and P. Kurowski, *Europhys. Lett.* **9**, 779 (1989).