Cellular self-propulsion of two-dimensional dissipative structures and spatial-period tripling Hopf bifurcation

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Instabilities arising in two-dimensional patterns are analyzed; in particular, we report on two generic instabilities of an ordered dissipative stationary structure. The first of these corresponds to several-point symmetry breakings, such as mirror and rotation symmetry, which cause the patterns to travel and may give rise to spirals or labyrinths. The second type manifests itself as a collective out-of-phase temporal oscillation, resulting in spatial-period tripling. We present both a numerical and an analytical analysis of the new emerging patterns. [S1063-651X(97)03006-7]

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There is an overabundance of dissipative systems that may spontaneously build up a highly organized pattern from an initially structureless state when they are moved away from thermodynamic equilibrium. Typical examples of two-dimensional (2D) patterns include reaction-diffusion [1] systems, Marangoni convection [2], Faraday waves [3], moving fronts [4], granular media [5], and perhaps biological objects. The most frequent ordered structures are hexagons, although there is evidence of various other patterns (squares, etc.).

In studies on secondary instabilities (i.e., instabilities of the ordered structure) of *one-dimensional systems*, considerable progress has been made both experimentally [6–9] and theoretically [10–14] since the discovery by Simon, Bechhoefer, and Libchaber [15] of the so-called ''solitary'' mode (consisting of one or two asymmetric cells traveling sideways).

To date, however, work on two-dimensional structures has been primarily directed toward relative stability between ordered patterns and their stability against phase modulations (the Eckhaus instability). From symmetry considerations, the pattern is expected to undergo myriad secondary instabilities. The aim of this paper is to report explicitly on two generic instabilities. The basic state is taken to be hexagonal, both for definiteness and because hexagons are a generic pattern. The first instability is accompanied by the loss of several mirror and rotation symmetries, which may cause the pattern to drift in one of six possible directions. Conflicts in the choice of drift direction may cause spirals or labyrinths to develop. The second one appears as a Hopf bifurcation where the six cells forming the corners of the hexagon oscillate in an alternating manner with a temporal phase shift between two successive cells of $2\pi/3$, whereas the cell in the middle of the hexagon has a temporal phase shift of $-2\pi/3$. Other types of collective out-of-phase oscillations are also possible.

Our analysis will be exemplified both analytically and numerically on a generic equation, the 2D version of the damped Kuramoto-Sivashinsky (DKS) equation

$$\frac{\partial h}{\partial t} = -\alpha h - \nabla^2 h - \nabla^4 h + (\nabla h)^2, \tag{1}$$

which is a paradigm in dissipative systems. This equation arises in a large variety of physical and chemical situations [16], and we therefore expect the results to have a general consequence. Moreover, we have shown that in 1D [14] this equation produces generic instabilities (parity breaking, vacillating breathing, etc.) observed in various systems. For all these reasons we have chosen to work on this equation. It will be recognized that all our reasoning can be readily used with any other starting model equations.

The scalar h may mimic, for example, the instantaneous position of a surface separating two phases. For example, Eq. (1) describes front dynamics in free growth at large speed [17]. The damping term α acts as a stabilizer and plays the role of a tuning parameter for complexity (i.e., a control parameter, such as the thermal gradient in directional solidification).

The linear dispersion relation (for perturbations in the form $e^{i\mathbf{q}\cdot\mathbf{r}+\omega t}$, where $\mathbf{r}$ designates the two-dimensional position vector) reads $\omega = -\alpha + q^2 - q^4$. The critical condition for the onset of instability is given by $q_c = 1/\sqrt{2}$ and $\alpha_c = 1/4$. Below α_c, the homogeneous state becomes unstable against the formation of cells. All ordered structures that are compatible with translational symmetries are allowed (squares, rolls, hexagons, etc.). We find that close to α_c the hexagons prevail (note that hexagons appear for $\alpha < \alpha_c$ as a transcritical bifurcation due to the loss of the up-down symmetry).

The richness of nonequilibrium systems stems from the fact that their dynamics do not generally possess a Lyapunov functional, except for specific situations very close to the threshold. The variational character may be broken either because other modes become active, a situation that may lead to several symmetry breakings and drifts (drift is intimately related to nonvariational effects), or because inhomogeneous fluctuations set in that detect the underlying periodic state through nonlinearities. This results in wave interferences that lead to collective—generally out-of-phase—oscillations and in reduction of the translational symmetries (here, tripling of the spatial wavelength). Here again, nonvariational effects lead to permanent motions.

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Let us begin with the first category in which the original translational invariance is preserved. When lowering the value of α, there are new modes that become unstable. The initial hexagonal structure is represented on the basis of the unit vectors $\mathbf{u}_1$, $\mathbf{u}_2$, $\mathbf{u}_3$ (where the three vectors make an angle of 120° with each other) with amplitudes A_1, A_2, and A_3 respectively. The basic wave number is denoted by q. For a hexagonal pattern, the next mode to become unstable is the one whose wave vector lies on the circle with radius $\sqrt{3}q$. It is the interaction between this mode and the basic one that induces several symmetry breakings and drifts. This mode (which is naturally generated from nonlinearities) is built on the new basis, $\mathbf{u}_2-\mathbf{u}_3$, plus cyclic permutations. Let the three new amplitudes be denoted by B_i ($i=1,2,3$; B_1 is the amplitude associated with $\mathbf{u}_2-\mathbf{u}_3$). The amplitude equations involving interactions between both harmonics take the form [18]

$$\dot{A}_1=\mu_1A_1+\mu_2A_2^*A_3^*+\mu_3(A_2B_3+A_3B_2^*)+\mu_4(A_2^{*2}B_1+A_3^{*2}B_1^*)+\mu_5|A_1|^2A_1+\mu_6(|B_2|^2+|B_3|^2)A_1, \quad (2)$$

$$\dot{B}_1=\mu_7B_1+\mu_8A_2A_3^*+\mu_9B_2^*B_3^*+\mu_{10}(A_1A_2^2+A_1^*A_3^{*2})+\mu_{11}(|A_2|^2+|A_3|^2)B_1+\mu_{12}|B_1|^2B_1+\mu_{13}(|B_2|^2+|B_3|^2)B_1, \quad (3)$$

plus cyclic permutations. The forms of these equations could also be inferred from translational and rotational symmetries. The forms of the equations are general and should arise regardless of the strarting model equations.

The explicit form of the coefficients entering the amplitude equations will not be listed in this brief account. It is a simple matter to recognize that, in the absence of the B terms, the equation of motion possesses a Lyapunov functional. The interaction between A and B breaks the variational character, and this is precisely the source of permanent drift outlined below. Introducing the amplitude and phase for the complex quantities, $A_j=R_a^je^{i\alpha_j}$ and $B_j=R_b^je^{i\beta_j}$, we can deduce real equations. There is a plethora of possibilities, but we shall limit ourselves to a typical situation in this brief exposition, while leaving the details to a forthcoming publication. We consider here the case where the amplitudes in the three hexagonal directions are identical: $R_a^j=R_a$ and $R_b^j=R_b$. Then, straightforward algebra on the real and imaginary parts of Eqs. (2) and (3) yields the following condition [18]:

$$|\theta_1|=|\theta_2|=|\theta_3|=\theta, \quad \theta_1\equiv\beta_1-(\alpha_2-\alpha_3). \quad (4)$$

The quantity θ represents the phase shift between the harmonic built on the direction $\mathbf{u}_2-\mathbf{u}_3$, with phase β_1, and the phases of the principal harmonics in the $\mathbf{u}_2$ (phase α_2) and $\mathbf{u}_3$ (phase α_3) directions that serve to construct the next resonant harmonic. This is again a consequence of rotational symmetry. There are two possibilities that may arise: either all the θ's have the same sign, or one θ has a sign opposite to that of the two others. We consider here only the second case, although the first one is also possible [18] from general arguments, although we have not yet seen any dynamical signature for it. It is convenient to write the field h in a real form:

$$\begin{aligned} h=&2R_a[\cos(q\mathbf{u}_1\cdot\mathbf{r})+\cos(q\mathbf{u}_2\cdot\mathbf{r})+\cos(q\mathbf{u}_3\cdot\mathbf{r})]\\ &+2R_b\{\cos\theta\cos[q(\mathbf{u}_2-\mathbf{u}_3)\cdot\mathbf{r}]-\sin\theta\sin[q(\mathbf{u}_2-\mathbf{u}_3)\cdot\mathbf{r}]\}\\ &+2R_b\{\cos\theta\cos[q(\mathbf{u}_3-\mathbf{u}_1)\cdot\mathbf{r}]+\sin\theta\sin[(q(\mathbf{u}_3-\mathbf{u}_1)\cdot\mathbf{r}]\}\\ &+2R_b\{\cos\theta\cos[q(\mathbf{u}_1-\mathbf{u}_2)\cdot\mathbf{r}]+\sin\theta\sin[q(\mathbf{u}_1-\mathbf{u}_2)\cdot\mathbf{r}]\}, \end{aligned} \quad (5)$$

where the phases α_i and β_i have been absorbed through an appropriate translation, $\mathbf{r}\to\mathbf{r}-\mathbf{r}_0$, with $q\mathbf{r}_0\cdot\mathbf{u}_i=\alpha_i$. It is a simple matter to see that as soon as $\theta\neq 0,\pi$, several symmetries are broken. It is clear from the above expression that both symmetries with respect to the mirrors that are orthogonal to the axes $\mathbf{u}_2$ and $\mathbf{u}_3$ are broken, while the one on the mirror orthogonal to $\mathbf{u}_1$ is preserved. Similarly, several rotational symmetries of the hexagonal group together with central symmetries are broken as well (note that the latter corresponds in 2D to a rotation by an angle of π). Some of these symmetries are easy to recognize from Eq. (5) (see below also). A complete enumeration of the spatial group symmetries that are broken will be published elsewhere [18]. As a consequence of these symmetry breakings (combined with nonvariational effects), the pattern drifts. Indeed, an analysis of the imaginary part of Eqs. (2) and (3) shows that $\alpha_j=-q\mathbf{C}\cdot\mathbf{u}_jt$ [recall that α_j had been absorbed into Eq. (5) via a translation by $\mathbf{r}_0$], where $\mathbf{C}=4qR_b^*\sin\theta^*(\mathbf{u}_2-\mathbf{u}_3)$ is the drift velocity (the stars refer to fixed points of the dynamical system). This obviously shows a connection between the drift and the parameter θ, which is directly connected to several-point symmetry breaking. The pattern drifts along the direction given by $\mathbf{u}_2-\mathbf{u}_3$ (which is orthogonal to $\mathbf{u}_1$). That is to say, the pattern drifts precisely along the mirror that preserves the symmetry, a fact which can be understood quite intuitively. Had we chosen the phase along $\mathbf{u}_2$ to have a sign opposite to the two others, the drift would then have been in the direction given by $\mathbf{u}_3-\mathbf{u}_1$. In total (and due to the original symmetry), there are six drift directions which are possible and are given by the $\pm(\mathbf{u}_1-\mathbf{u}_2)$ (plus cyclic permutations) directions. The choice depends upon the actual fluctuations in a given system. Note that the drift direction is fixed by the second excited harmonic (here, along $\mathbf{u}_2-\mathbf{u}_3$), a result which is not (at least to the authors) obvious *a priori*! The six drift directions arising from the interaction of modes q and $\sqrt{3}q$ are perpendicular to the sides of the hexagonal cells [19].

An important question arises. Under which circumstance does the solution discussed above occur? Our analysis shows that this basically happens as soon as the harmonic with wave number $q\sqrt{3}$ becomes active. The region in parameter space where this situation is encountered is approximately determined by the condition $\omega(q\sqrt{3})>0$. This defines a boundary beyond which the new solution takes over. We believe this condition to be quite general and it should apply to diverse situations; in particular, the same conclusion is reached with a more complex equation arising in the problem of a moving nematic-isotropic boundary [18]. This means that the knowledge of the linear dispersion relation may be

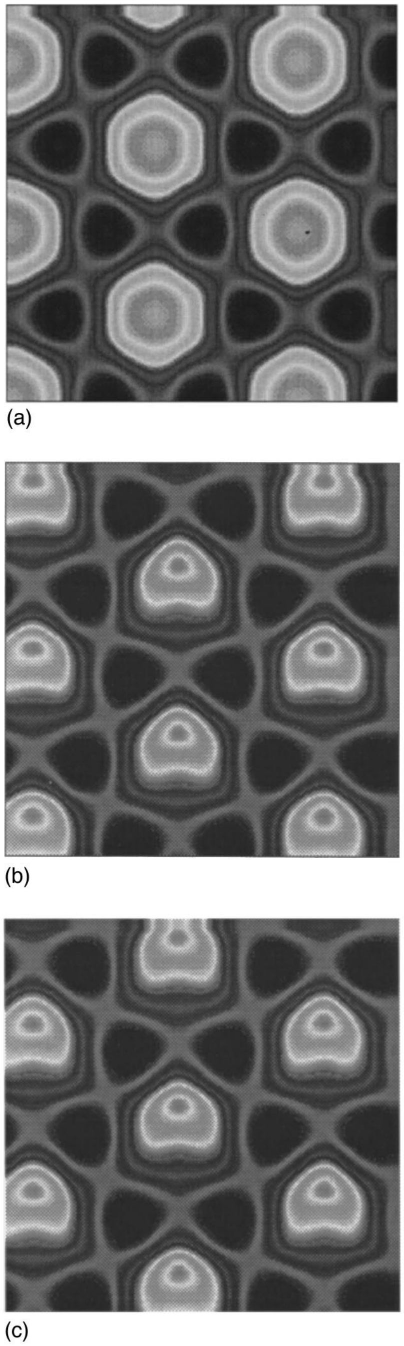


FIG. 1. The original hexagonal pattern and two other figures (taken at different times) showing the symmetry breaking and the upward drift.

sufficient in a given system to hint at the boundary in parameter space that separates the stable ordered structure from the one leading to drift.

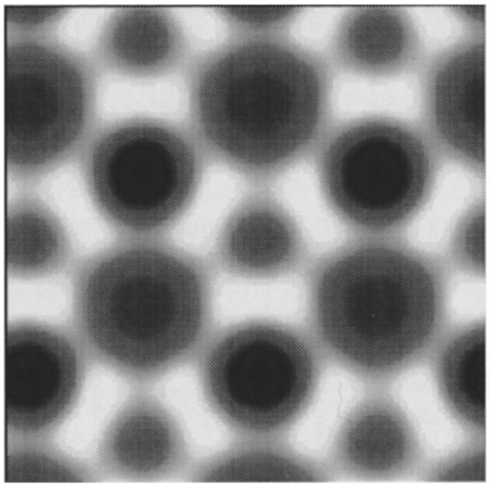
Our analytical results have been confirmed by numerical calculations in two ways. First, we have integrated the dynamical system of amplitude equations (2) and (3). Starting from arbitrary solutions in the region just below the line fixed by $\omega(q\sqrt{3})=0$, dynamics tend toward the above-mentioned symmetry breaking bifurcation. Figure 1 displays the initial pattern [see Fig. 1(a)] and its evolution after the instability develops [see Figs. 1(b) and 1(c)]. Several broken symmetries are easily identified in the figure. Note also that Figs. 1(b) and 1(c) demonstrate that the pattern drifts upward. Because the above amplitude equations are expected to be quantitatively and qualitatively accurate only close enough to the codimensional two-bifurcation point, we have solved numerically the full original DKS equation in order to ascertain the validity of amplitude equation truncations. We have used a hexagonal box with periodic boundary conditions. The spatial derivatives are evaluated using second-order finite differences, and the resulting dynamical system is solved by means of a four-point Runge-Kutta method. For spatial derivatives we have made use of a particular property of a hexagonal grid, $(\Sigma_{l=1}^{6}h_l-6h_0)/a^2=\nabla^2 h_0+O(a^2)$, where a is the mesh spacing and the sum is over the field values at the six corners of a hexagon, whereas h_0 refers to its value in the center of the hexagon. This trick has the advantage of accurately preserving the rotational symmetry of the original equation. The numerical analysis has here again confirmed the overall picture of the results described above. A similar analysis has been performed for the NIB equation.

Our next investigation focuses on oscillatory modes. Before presenting the underlying ideas that guided our work, we shall first discuss the results that emanate from the full numerical analysis. By increasing the ''aspect ratio'' (the ratio between the box size and the critical wavelength), dynamics have proven to be richer and richer. If this ratio is large enough (at least equal to three), and we use a hexagonal box, we have identified an oscillatory mode close enough to the threshold, the snapshot of which is shown in Fig. 2. Each cell in the center is oscillating with a phase shift with the six cells surrounding it (forming the corners of the hexagon). Each corner is oscillating with a phase shift with the adjacent corner, but in phase with the following corner. In all, we have three oscillators (one in the middle of the hexagon and two in the corners). The temporal phase shift between each oscillator is found to be equal to $2\pi/3$. It is clear from the figure that this oscillation is accompanied by an increase of the spatial wavelength. Close inspection shows that the wavelength has tripled.

In order to illustrate the lines of reason that guided this investigation, we briefly develop a two-dimensional analogy with band theory in solid-state physics, as we have previously suggested for a one-dimensional structure [14]. For that purpose, let $h_0(\mathbf{r})$ be a steady and spatially periodic solution and $h_1(\mathbf{r},t)$ a small perturbation. Linearization of Eq. (1) yields

$$\frac{\partial h_1}{\partial t}=-\alpha h_1-\nabla^2 h_1-\nabla^4 h_1+2\nabla h_0\cdot\nabla h_1. \tag{6}$$

This equation is reminiscent of the Schrödinger equation in imaginary time for an electron in a crystal, but with a fundamental difference (see below). ∇h_0 plays the role of the periodic potential. The Floquet-Bloch theorem states that the general solution has the form $h_1=e^{\sigma t}e^{i\mathbf{Q}\cdot\mathbf{r}}\hat{h}(\mathbf{r})$, where σ is the growth rate (or the energy in solid-state physics), $\mathbf{Q}$ is a wave vector which belongs to the first Brillouin zone,


(a)

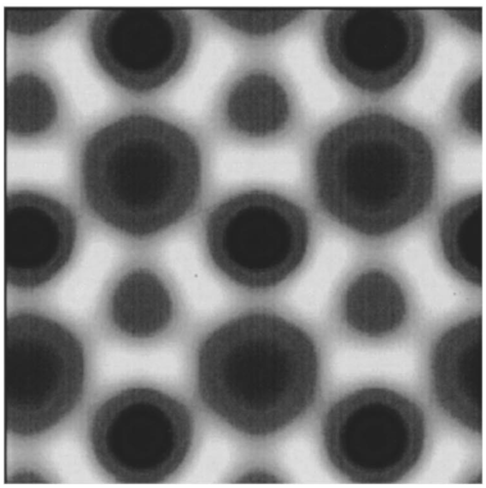

(b)

FIG. 2. Out-of-phase oscillations. Cells have exchanged their role from one figure to the next (not a full temporal period is shown, but approximately 2/3).

$|\mathbf{Q}| \leqslant |\mathbf{q}|$, and $\hat{h}(\mathbf{r})$ is a periodic function having the periodicity of the basic solution h_0. If $\sigma(\mathbf{Q})$ is an eigenvalue, then this is equally true for $\sigma(\mathbf{q}+\mathbf{Q})$, due to the Goldstone mode associated with translational invariance. Strong wave coupling occurs only when $\omega(\mathbf{Q}) \simeq \omega(\mathbf{q} \pm \mathbf{Q})$ (recall that ω is the bare eigenvalue, and in a perturbative scheme it suffices to consider this quantity), and this leads to gap opening. Let us be more specific here and focus on the direction perpendicular to $\mathbf{u}_1$. Inspection of the relation $\omega(\mathbf{Q}) \simeq \omega(\mathbf{q} \pm \mathbf{Q})$ reveals some interesting intersections. The first one is obvious and is given by $Q = q/\sqrt{3}$. This gives rise to a resonance between the incident wave [22] and the reflected one [22]. This is a direct analogue of the so-called Bragg reflection. Treating the term ∇h_0 as a perturbation (in a similar way as for the quasifree electron problem in a crystal) and expanding the wave on the space spanned by the two degenerate states, we find an ''energy'' gap at $Q = q/\sqrt{3}$ [18], which is unimportant here. Most important in our case is the second degeneracy (which is easy to determine by writing down explicitly the bare dispersion relation ω), which occurs at $Q \sim q/\sqrt{3}$ (for q close to q_c). This resonance follows from interaction of the incident wave and the transmitted one [22]. Perturbation theory reveals here a nonclassical result (which has no analogue in quantum mechanics): the resonance opens a gap on the $\mathbf{Q}$ axis. This gap corresponds to complex eigenvalues σ. This is possible here since the linear operator in Eq. (6) is not self-adjoint (contrary to Hamiltonians in quantum mechanics). A simple calculation leads (at the crossing point) to $\mathrm{Im}(\sigma) = \sqrt{15} R_a q^2/2$. On one hand, the fact that the resonance occurs at $Q \sim q/3$ implies a period tripling of the structure in the direction of **ui**. On the other hand, the opening of a gap on the wave number axis means that the instability must be oscillatory in time. This completes our analysis, on which we shall give an extended account in the near future.

Finally, an important question remains to be addressed. This concerns the experimental access to these dynamics. To the authors' best knowledge, no experimental evidence has been reported on the instabilities we have described. While we think that several systems may be good candidates, eutectics have proven in one dimension to be appropriate for both drifting patterns and oscillations. According to one of our suggestions [20], a sudden change in the growth velocity by a factor of about 4 had led to pattern drifting as a whole. Most experiments on two-dimensional fronts are performed on metals where *in situ* analysis is a formidable challenge. We believe that transparent materials (such as those used for thin samples) are good candidates on which to perform experiments. Here, of course, visualization based on optical transmission (as with thin samples) is probably not suitable. Imaging using reflection on the solid-liquid front would most likely be promising. A sudden change of the velocity (presumably by a factor of 3 for a hexagonal structure, due to the law $\lambda^2 V \sim$ const and because wavelength adjustment is very slow, so that a sudden change in velocity is practically equivalent to a wavelength increase; V is the growth velocity and λ the wavelength) should cause symmetry breaking along the whole front. Oscillations may be generated by acting on the eutectic concentration. Chemical or hydrodynamic systems may also offer the possibility of accessing a drift, provided one can monitor a wavelength modification, for example, via a thermal memory, as has been devised for the study of the Eckhaus instability [21]. Once the condition for the drift is reached, it is likely that a conflict in the choice of the traveling direction can cause several interactions between waves in different directions. This may lead to spiral-like defects or labyrinth patterns. Preliminary simulations on large scales seem to reveal evidence of such structures.

It must be emphasized that the same analysis can be performed for any symmetry other than hexagonal. The present work has given a short view of the richness of what we may call the theory of stability of ''two-dimensional nonvariational crystals.'' There are 17 two-dimensional space groups. Combinations of simple analytical concepts such as those presented here—group theory, and numerical works—promise to provide a wide program of nonlinear physics of 2D dissipative structures.

What we have learned so far is that a simple nonlinear equation based on symmetries and nonvariational ingredients (which are inherent in nonequilibrium systems) reveals a variety of fascinating static and dynamical phenomena. These continue to prove to be a disguised form for only a few prototypes and may embrace diverse physical and chemical systems.

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[1] V. Castets, E. Dulos, J. Boissonade, and P. de Kepper, Phys. Rev. Lett. **64**, 2953 (1990).
[2] P. Kolodner, Physica D **37**, 319 (1989).
[3] B. Christiansen, P. Alstrom, and M.T. Levinsen, Phys. Rev. Lett. **68**, 2157 (1992).
[4] B. Billia, H. Jamgotchian, and H. Nguyen Thi, Metall. Trans A **22A**, 3041 (1991).
[5] F. Melo, P. B. Umbanhowar, and H. L. Swinney, Phys. Rev. Lett. **75**, 21 (1995).
[6] C. Faivre, S. de Cheveigné, C. Guthmann, and P. Kurowski, Europhys. Lett. **9**, 779 (1989).
[7] M. Rabaud, S. Michalland, and Y. Couder, Phys. Rev. Lett. **64**, 184 (1990).
[8] L. Limat, P. Jenffer, B. Dagens, E. Touron, M. Fermigier, and J.E. Wesfreid, Physica D **61**, 166 (1992).
[9] I. Mutabazi, H. Hegset, C.D. Andereck, and J.E. Wesfreid, Phys. Rev. Lett. **64**, 1729 (1990).
[10] K. Kassner and C. Misbah, Phys. Rev. Lett. **65**, 1458 (1990).
[11] H. Levine and W.J. Rappel, Phys. Rev. A **42**, 7475 (1990).
[12] P. Coullet and G. Iooss, Phys. Rev. Lett. **64**, 866 (1990).
[13] K. Kassner, C. Misbah, H. Müller-Krumbhaar, and A. Valance, Phys. Rev. E **49**, 6 (1994).
[14] C. Misbah and A. Valance, Phys. Rev. E **49**, 1 (1994).
[15] A. J. Simon, J. Bechhoefer, and A. Libchaber, Phys. Rev. Lett. **63**, 2574 (1989).
[16] Y. Kuramoto and T. Tsuzuki, Prog. Theor. Phys. **55**, 356 (1976); I. Sivashinsky, Acta Astronautica **4**, 1177 (1977) D.J. Benney, J. Math. Phys. **45**, 150 (1966); R. Cuerno *et al.*, Phys. Rev. Lett. **74**, 4746 (1995); J. Krug *et al.*, Phys. Rev. Lett. **75**, 3894 (1995).
[17] C. Misbah, H. Müller-Krumbhaar, and D.E. Temkin, J. Phys. (France) I **1**, 585 (1991).
[18] I. Daumont, K. Kassner, C. Misbah, and A. Valance (unpublished).
[19] It is equally possible to obtain drifting solutions by an interaction between modes q and $2q$, which is the direct analogue to the one-dimensional case. In that situation there are also six drift directions, but now they point towards the *corners* of the basic hexagons. So far, we have investigated these additional solutions only numerically, but an analytical description along the lines exhibited here should be feasible.
[20] K. Kassner and C. Misbah, Phys. Rev. A **44**, 6533 (1991).
[21] M. Lowe and J.P. Gollub, Phys. Rev. Lett. **55**, 2575 (1985).
[22] Since the incident wave $e^{i\mathbf{Q}\cdot\mathbf{r}}$ is perpendicular to $\mathbf{u}_1$, the reflected waves are $e^{i(\mathbf{Q}-q\mathbf{u}_2)\cdot\mathbf{r}}$, $e^{i(\mathbf{Q}+\mathbf{qu}_3)\cdot\mathbf{r}}$ and the transmitted ones $e^{i(\mathbf{Q}+\mathbf{qu}_2)\cdot\mathbf{r}}$ and $e^{i(\mathbf{Q}-\mathbf{qu}_3)\cdot\mathbf{r}}$.