Hydrodynamic coarsening in striped pattern formation with a conservation law

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We observed in numerical simulations that the interaction of striped-pattern-forming instability and a neutrally stable zero mode induces patterns of domains of upflow hexagons coexisting with domains of downflow hexagons. They appear only when hydrodynamic flow is present.

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Our aim is to describe the behavior of systems in which the onset of pattern formation occurs with a finite wave number k_0 (hence resulting in spatially periodic patterns) when hydrodynamic interactions are present at the same time. We restrict our attention in this paper to two-dimensional systems. We model the system by an order-parameter field, ψ, which couples to the velocity field, $\boldsymbol{v}$. Beside the usual space and time translational symmetry, it is assumed to be invariant under the transformation

$$(\psi,\boldsymbol{v}) \to (-\psi,\boldsymbol{v}), \tag{1}$$

so that there is no preference for hexagons over stripes (rolls). We further assume that the system satisfies the symmetry under reflection in horizontal coordinates x and y;

$$(x,y) \to (-x,-y), \quad \boldsymbol{v} \to -\boldsymbol{v}, \quad \psi \to \psi. \tag{2}$$

As a specific example, we consider the case where the velocity field is driven by distortions of the field ψ, and the length scales of the generated flow are large compared to k_0^{-1}. The minimal dynamical model which encompasses the symmetry constraints listed above takes the form

$$\partial_t\psi = (i\,\boldsymbol{\nabla})^p[\varepsilon\psi - \tilde{\xi}_0^4(\nabla^2 + k_0^2)^2\psi - u\psi^3] - \boldsymbol{v}\cdot\boldsymbol{\nabla}\psi, \tag{3}$$

$$\tau\partial_t\boldsymbol{v} = \nu(\nabla^2 - c^2)\boldsymbol{v} - \frac{1}{2}g_0\,\boldsymbol{\nabla}(\boldsymbol{\nabla}\psi)^2. \tag{4}$$

Here $p=0$ for a nonconserved order parameter (NCOP), whereas for a conserved order parameter (COP) $p=2$. The ε is the control parameter with the transition to stripes occurring for $\varepsilon > 0$. The coefficient g_0 of the second term on the right-hand side of Eq. (4) represents the strength of the hydrodynamic coupling between $\boldsymbol{v}$ and ψ, while the first term represents the viscous dissipation; τ, c^2 are other phenomenological parameters of the velocity field. The main task in what follows is to discuss the COP case regarding spatial patterns that may be induced by this dynamical model.

The terms inside the square brackets in Eq. (3) are just those of the much-studied Swift-Hohenberg (SH) equation [1]. In particular, in the NCOP case the above model equations (3) and (4) are equivalent to the model proposed [2] to describe the mean-flow effect in Rayleigh-Bénard convection. In fact, by taking the curl of Eq. (4) and introducing the vertical vorticity potential, ζ, with

$$-\nabla^2\zeta = \hat{z}\cdot(\boldsymbol{\nabla}\times\boldsymbol{v}), \tag{5}$$

where $\hat{z}$ is the unit vector in the z direction, we find

$$[\tau\partial_t - \nu(\nabla^2 - c^2)]\nabla^2\zeta = g_0\hat{z}\cdot\left(\boldsymbol{\nabla}\nabla^2\psi\times\boldsymbol{\nabla}\psi\right). \tag{6}$$

This is the vorticity equation given in Ref. [2]. The set of equations (3) and (6) has been shown by analytical and numerical studies [3] to reproduce complex pattern formation of convective fluids near onset. Among others, spiral-defect chaos [4] was found to be reproduced by numerically simulating these generalized SH equations; see Fig. 1(a). In the following, assuming a slowly varying vorticity, we neglect the inertial effect and take $\tau=0$ (the so-called passive vorticity case [3]).

The model for the COP ($p=2$) is particularly interesting in that the system's dynamics is expected to drastically differ from that in the NCOP case. This is due to the coupling of long-wavelength modes belonging to a zero-mode (Goldstone) branch of the spectrum (see Fig. 2) originated in the conservation law to random long-wavelength modulations of the short-wavelength ($\propto k_0^{-1}$) pattern [5,6]. In many physical situations, the presence of a conservation law is related to the long-wavelength instability. However, here in Eq. (3) the instability is a short-wavelength one. It is conjectured [5] that such coupling would lead to instability of all weakly nonlinear spatially periodic patterns that may occur in such sys-

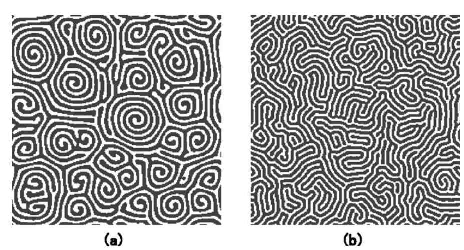

FIG. 1. (a) Spiral-defect pattern in the nonconserved case ($p=0$) as it occurs in the cell-dynamical-system simulations of Eqs. (3) and (6). (b) Bicontinuous roll (striped) pattern in the conserved case ($p=2$) without hydrodynamic interactions. The same parameters and random initial conditions are used in both cases. The white regions denote positive values of the order parameter ψ and the gray ones negative ψ.

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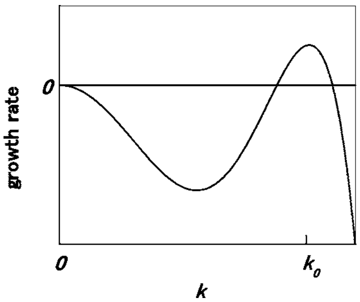

FIG. 2. The growth rate of infinitesimal perturbations of the trivial solution $\psi=0$ of Eq. (3) with COP, where k is the perturbation wave number. For $0<\varepsilon\ll 1$, there is a narrow band of unstable wave numbers near $k=k_0$. Furthermore, for small k the conservation law generates a neutrally stable zero mode.

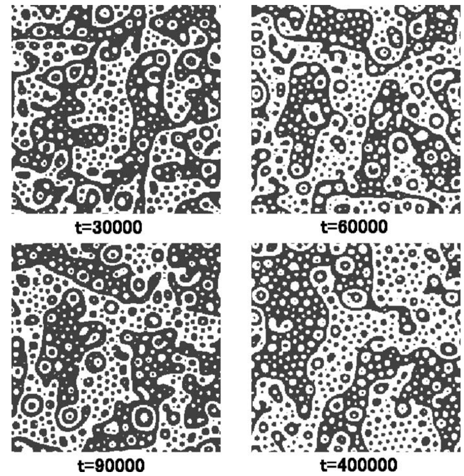

FIG. 3. Evolution of coexisting H0 and Hπ hexagons occurring in simulations of COP Eqs. (3) and (4) for hydrodynamic interactions $g_0=20$; all other parameters are the same as in Fig. 1 with the same initial conditions.

tems. It is then expected that the pattern retains short-range order originating in the short-wavelength instability while the long-wavelength modulation of such spatially periodic patterns of meso-scales persists indefinitely.

The simplest realization of this type of coupling has been found in the Nikolaevskii equation [5]:

$$\partial_t\psi = -\partial_x^2[\varepsilon - (\partial_x^2 + k_0^2)^2]\psi - \psi\partial_x\psi, \tag{7}$$

which was originally proposed to describe propagation of longitudinal seismic waves in viscoelastic media. It is shown by means of direct simulations of Eq. (7) that spatially periodic steady states do not occur, and instead spatiotemporal chaos arises supercritically.

Hydrodynamic effects on pattern formation for which $k_0=0$ have been intensively studied both experimentally and theoretically. The phenomena are commonly observed in spinodal decomposition of binary fluid mixtures [7]. However, even for these simple fluids, the hydrodynamic effects have not been fully understood due to the nonlocal nature of hydrodynamic interactions, awaiting further clarification to gain complete understanding of the effects.

One of the most intriguing recent discoveries in hydrodynamic coarsening of binary fluid mixtures is the observation of phenomena called spontaneous double phase separation (DPS) [8,9]. When the hydrodynamic flow is strong enough, the hydrodynamic coarsening can be too quick for the order parameter in each domain to attain its equilibrium value. With domains rendered metastable or unstable in this way, this effectively causes a kind of further quench effect, and the secondary spinodal decomposition can take place within each domain. The resulting pattern is a nested circular droplet formation within the large bicontinuous domains. This scenario for DPS, first proposed by Tanaka [8], seems to be quite universal for any hydrodynamic coarsening systems, including not only the fluid mixtures (for which $k_0\equiv 0$) but also systems in which the forming pattern is a spatially periodic state.

Given all the facts mentioned above, we now pose the following questions. (1) Can spirals, which appear for parameter values where the stripe state is known to be stable in the system described by the NCOP model (3) and (6), survive the presence of the long-wavelength zero mode? (2) What is the outcome (pattern morphology) of the two competing growth mechanisms, i.e., the spontaneous DPS and the spiral-defect generation, which may be operative in hydrodynamic systems developing spatially periodic patterns?

In pursuing these goals, we performed numerical study of Eqs. (3) and (4). We employed the cell-dynamical-system (CDS) method [10,11] on a square lattice of size 256×256 with periodic boundary conditions, and applied the predictor-corrector algorithm to upgrade the order-parameter field. In order to solve the implicit equation for ζ, we used the pseudo-spectral technique with a fast Fourier transform method. The initial conditions were a random distribution of ψ of amplitude 0.1. We have presented results for $g_0=0$ [Fig. 1(b)] and $g_0=20$ (Fig. 3) to assess the importance of the hydrodynamic interactions.

We surprisingly found that the spiral pattern gives way to the hexagonal planform in which hexagons of the type H0 and Hπ [12] coexist. Furthermore, we observed that patches of coexisting hexagonal patterns underwent slow evolution in time and showed no tendency to reach any stationary state (Fig. 3).

We now discuss the physical origin of the unusual patterns shown in Fig. 3. To that end, we apply the standard method of multiple scales approach. Namely we seek a solution of Eqs. (3) and (4) when ε is small in the form

$$\psi = \varepsilon^{1/2}\left[B + \left(\sum_{j=1}^{3} A_j e^{i\boldsymbol{k}_j\cdot\boldsymbol{r}} + \text{c.c.}\right)\right] + \varepsilon\psi_1 + \cdots, \tag{8}$$

$$\boldsymbol{v} = \varepsilon\boldsymbol{v}_0 + \varepsilon^{3/2}\boldsymbol{v}_1 + \cdots, \tag{9}$$

where $|\boldsymbol{k}_j|=k_0$ and the wave vectors satisfy the resonance condition $\Sigma_j\boldsymbol{k}_j=0$. The amplitudes A_j and B depend on slow space and time variables $X=\varepsilon^{1/2}x$, $Y=\varepsilon^{1/2}y$, and $T=\varepsilon t$. In the following analysis we neglect the ∇^2 term on the RHS of Eq. (4) because ∇^2 is small when compared to a constant $-c^2$ in the case of thin films; recall that physically this constant arises from averaging of ∂_z^2 over the thickness of the horizon-

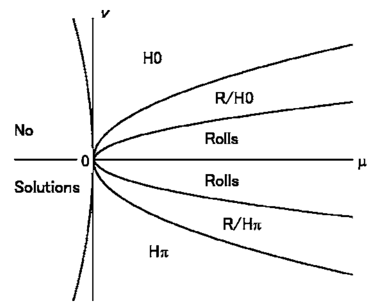

FIG. 4. Linear stability regions associated with Eq. (10) with constant parameters μ and v when $\gamma>1$. The region denoted by $R/\mathrm{H}0(\mathrm{H}\pi)$ corresponds to the mixed state of rolls and $\mathrm{H}0(\mathrm{H}\pi)$ hexagons; for $\gamma<1$ the rolls become unstable.

tal films [2]. Also we use the units in which $\tilde{\xi}_0=u=1$.

Considering terms for A_j at $O(\varepsilon^{3/2})$ and $\boldsymbol{v}_0$ at $O(\varepsilon)$, we find the amplitude equations to be

$$\partial_T A_j = [\mu(B) + 4(\boldsymbol{n}_j \cdot \boldsymbol{\nabla}_X)^2]A_j + v(B)A_{j-1}^* A_{j+1}^* - g(h)[|A_j|^2 + \gamma(h)(|A_{j-1}|^2 + |A_{j+1}|^2)]A_j \quad (j=1,2,3 \bmod 3), \tag{10}$$

$$\partial_T B = \nabla_X^2 B + 3h(A_1A_2A_3 + A_1^*A_2^*A_3^*), \tag{11}$$

with

$$\mu(B) = 1 - 3B^2, \quad v(B) = -6B,$$

$$g(h) = 3 + 2h, \quad \gamma(h) = (6+h)/(3+2h), \tag{12}$$

where $\nabla_X^2 = \partial_X^2 + \partial_Y^2$ and $\boldsymbol{n}_j \equiv \boldsymbol{k}_j/k_0$. In Eqs. (10) and (11), we have rescaled the slow variables (X,Y) and T by the cosmetic factors k_0 and k_0^{-2}, respectively, and the parameter $h \equiv g_0 k_0^2/(2\nu c^2)$ representing the hydrodynamic coupling.

For the present purpose we may consider Eqs. (10) and (11) for the case of perfect structures, i.e., those with uniform amplitudes. Then the amplitude equations (10) for A_j's are of the well-known form [12,13], except that, in the present case, the parameters μ and v are B-field dependent. In this connection we remark that the nonzero value of v is the prerequisite for formation of hexagonal patterns. Although analytic study of these coupled equations is hardly possible, we can make a qualitative prediction for the selected pattern based upon the linear stability diagram (Fig. 4) that has been obtained for the equation (10) with constant parameters μ and v, which we will give below.

Assume, for the moment, that the B-field attains the stationary value. Then (i) *in the absence of hydrodynamic interactions* ($h=0$): If one starts from small random perturbations as initial conditions, the neutrally stable zero modes (B) simply damp out (and eventually yield $\mu=1, v=0$). Hence the strength of quadratic coupling (v) in (10) is too weak to give rise to hexagonal patterns. We will thus find that a striped state is the selected planform. See Fig. 1(b) for confirmation.

(ii) *The case* $h\neq 0$: Now the zero mode couples to the short-wave instability through the nonlinear coupling to A_j's, so that the strength of v may become strong enough to induce hexagons. Since the sign of v can be locally both positive and negative depending on the value of the B field, we expect to observe the coexisting hexagonal patterns of two types, H0 (for $B<0$) and Hπ (for $B>0$); hexagons must be certainly degenerate in view of the symmetry $\psi\to-\psi$ in Eqs. (3) and (4). As already shown in Fig. 3, this effect has in fact been observed in our simulations. Noteworthy also in this figure is the existence of small droplets of the complementary phase in both H0 and Hπ phases.

In actuality, the B field, which is now blowing up due to the nonlinear coupling to A_j fields, is evolving slowly, both in time and in space as well, according to Eq. (11). Note at the same time that when μ is rendered too small by the increase of B^2 value, hexagons cease to exist; this in turn changes the B field into a stable mode. This being repeated, a slow coarsening dynamics of coexisting hexagons will persist with concomitant domain-wall motions, and the steady state would never be reached. Thus we expect our model should share the same features as the so-called [5] soft turbulence discovered in the Nikolaevskii model (7). The distinctive feature pertinent to our model, however, is the coexistence of H0- and Hπ-hexagonal states, and this is made possible only when the hydrodynamic interactions are present in the system.

Although the present work has not been motivated directly by experiments, the possible application of COP model (3) and (4) is to a microphase separation near its transition in an admixture of homopolymers and diblock copolymers. In this system, when the long-range repulsive interaction of monomer concentration deviation is screened, the situation can occur in which the short-wave instability couples to neutrally stable zero modes. (Note in passing that for pure copolymer systems, the long-range interaction will not be screened [14]. As a consequence, the zero mode is always absolutely stable.) Another possible experimental application is to a Langmuir monolayer. However, it is not clear at present how much of the present finding based on the modeling (4) of the hydrodynamic effects would be relevant in both systems. For example, the kinetics of the microphase separation in the presence of hydrodynamic interactions have been studied [15] by the time-dependent Ginzburg-Landau equations, which are generalization of the so-called model H for critical binary mixtures. In that case the basic equations possess the effective free-energy (Lyapunov) functional, in contrast to our model equations. In the case of Langmuir monolayers, the specific features of an adsorbed system might exert much larger effects than the neutral zero mode considered in the present paper.

Finally, let us remark that the coexistence of upflow and downflow hexagons [12] was experimentally observed in Boussinesq Rayleigh-Bénard convection (RBC) at higher values of Rayleigh number [16]. However, the mean-flow effect is not crucial here for the appearance of hexagons since no stable hexagons were found for low Prandtl numbers [17]. Therefore the hexagons in the present paper are not related to the RBC hexagons.

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