Resonant Patterns through Coupling with a Zero Mode

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The interaction between a diffusive instability and a quasineutral zero mode favors the onset of resonant structures in systems exhibiting inversion symmetry and stabilizes reentrant hexagonal patterns when this symmetry is absent.

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In driven systems, structures of hexagonal symmetry are ubiquitous. They have been observed in fields as diverse as hydrodynamics (Bénard-Marangoni [1] and non-Boussinesq Rayleigh-Bénard [2] convection), chemistry (Turing patterns) [3], and nonlinear optics (tranverse patterns) [4]. All these systems are described by a set of nonlinear partial differential equations for the state vector $\mathbf{U}$

$$\frac{\partial \mathbf{U}}{\partial t} = \mathbf{N}(\mathbf{U}, r; \nabla), \tag{1}$$

where $\mathbf{N}$ is a vector function of $\mathbf{U}$ and its spatial derivatives that describes the various kinetic processes taking place in the system. It also depends on some control parameter r. Generically, in 2D, resonant hexagonal patterns stabilized by triplet interactions appear as the result of a symmetry breaking instability in systems lacking inversion symmetry, i.e., for which the equality

$$\mathbf{N}(\mathbf{U}, r; \nabla) = -\mathbf{N}(-\mathbf{U}, r; \nabla) \tag{2}$$

is violated. In 3D, this property is also responsible for the onset of resonant patterns such as body centered cubic structures (bcc) or hexagonally packed cylinders (hpc) [5]. Some of these 3D structures have recently been obtained in open spatial chemical reactors [6]. In this Letter we study the influence of a quasineutral zero mode generated by a secondary steady-state bifurcation (that may mimic a phase transition) on such symmetry breaking instabilities. We show that it induces resonant structures when inversion symmetry is present. In the converse case it leads to reentrant resonant patterns.

We first consider a system described by Eq. (1) and for which the condition Eq. (2) is satisfied. We suppose, to fix the ideas, that for homogeneous conditions, the thermodynamic branch $\mathbf{U}_0$, such that $\mathbf{N}(\mathbf{U}_0, r) = 0$, undergoes a "pitchfork bifurcation" at $r = r_p$ giving rise to two new homogeneous steady states (HSS): $\mathbf{U}_+ = -\mathbf{U}_-$. We furthermore impose that the trivial state $\mathbf{U}_0$ can be destabilized by inhomogeneous perturbations of wave number q_c leading to a diffusive-type instability [3] occurring at $r = r_T < r_p$.

Standard bifurcation theory may be applied to describe the patterned solutions that branch off the state $\mathbf{U}_0$ at $r = r_T$ [7]. In large aspect ratio systems, the field $\mathbf{U}$ may be approximated by any linear superposition of m critical modes

$$\mathbf{U} = \mathbf{U}_0 + \mathbf{e}_T \sum_{i=1}^{m} A_i \exp(i\mathbf{q}_i \cdot \mathbf{r}) + \text{c.c.}, \tag{3}$$

where ($|\mathbf{q}_i| = q_c$) and $\mathbf{e}_T$ is the critical eigenvector of the corresponding linearization of $\mathbf{N}$. In 2D, and if the instability is saturated at third order, the complex amplitudes A_i obey a set of Landau equations

$$\frac{\partial A_i}{\partial t} = \mu A_i - g_1|A_i|^2 A_i - g_2 \sum_{j=1}^{m-1} |A_j|^2 A_i \quad (j \neq i), \tag{4}$$

where $\mu = (r - r_T)/r_T$ and $g_2 > g_1 > 0$. Owing to the absence of quadratic terms [Eq. (2)] and because the cubic interaction is competitive (i.e., the cross-mode coupling g_2 is larger than the self-coupling feedback g_1), the only stable structure corresponds to stripes with $m = 1$ [8]. They appear supercritically at $r = r_T$ with an amplitude A_1 such that $|A_1| = R_1 = \sqrt{\mu/g_1}$. However, when $r \rightarrow r_p$, the growth rate ω_0 of a homogeneous perturbation of amplitude A_0 about the state $\mathbf{U}_0$ tends to zero. As a result the uniform mode A_0 progressively rejoins the set of active modes. Its resonant interaction with the critical modes must then be taken into account since it can modify their stability. In particular, when $r_p - r_T$ is small this interaction may be described by coupled amplitude equations [9]

$$\begin{aligned} \partial_t A_0 &= \omega_0 A_0 - A_0^3 - \beta_1 \left[\sum_{i=1}^{3} |A_i|^2 \right] A_0 \\ &\quad - \beta_2 (A_1^* A_2^* A_3^* + A_1 A_2 A_3), \\ \partial_t A_1 &= \mu A_1 - g_1 |A_1|^2 A_1 - g_2 [|A_2|^2 + |A_3|^2] A_1 \\ &\quad - g_3 A_1 A_0^2 - g_4 A_0 A_2^* A_3^*, \end{aligned} \tag{5}$$

where β_1, β_2, g_3, and g_4 are positive and $\mathbf{q}_1 + \mathbf{q}_2 + \mathbf{q}_3 = 0$. The equations for A_2 and A_3 are obtained by cyclic permutation of the subscripts. As discussed recently by one of us [10], the interference between the pitchfork bifurcation, at r_p, and the symmetry breaking instability, at

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$r = r_T$, generates a quadratic coupling term proportional to A_0 in the amplitude equation for the patterned modes. These equations admit four types of global solutions: (i) the reference state and the homogeneous solutions $A_0 = 0$ or $A_{0\pm} = \pm\sqrt{\omega_0}$, $A_i = 0$; (ii) pure striped modes $|A_1| = R_1 = \sqrt{\mu/g_1}$, $A_0 = A_2 = A_3 = 0$; (iii) mixed modes of smectic symmetry $A_0 = \mathcal{R}_{M1} \neq 0$, $|A_1| = R_{M1}$ and $A_2 = A_3 = 0$, (iv) mixed modes of hexagonal symmetry $A_0 = \mathcal{R}_{M3} \neq 0$ and $A_i = R_{M3}\exp(i\phi_i)$. The first two have already been considered above; the third type, mixed modes, are unstable. In the last case the sum of phases of the modes forming the resonant triad, $\Phi = \phi_1 + \phi_2 + \phi_3$, obeys the equation

$$\frac{\partial \Phi}{\partial t} = g_4 \mathcal{R}_{M3} R_{M3} \sin\Phi \,. \qquad (6)$$

Therefore Φ relaxes to 0 (π) when $\mathcal{R}_{M3} = \mathcal{R}^0_{M3} < 0$ ($\mathcal{R}_{M3} = \mathcal{R}^\pi_{M3} > 0$). In the former case, the maxima of a component of the field $\mathbf{U}$ form a triangular lattice (h_0). In the latter, a honeycomb lattice (h_π) is obtained. The other components of the field are in phase or in phase opposition. The corresponding amplitudes are given by the solutions of

$$\omega_0 \mathcal{R}_{M3} - \mathcal{R}^3_{M3} - 3\beta_1 R^2_{M3}\mathcal{R}_{M3} \pm 2\beta_2 R^3_{M3} = 0\,,$$
$$\mu - (g_1 + 2g_2)R^2_{M3} - g_3\mathcal{R}^2_{M3} \pm g_4\mathcal{R}_{M3}R_{M3} = 0\,, \qquad (7)$$

with the signs, respectively, corresponding to an h_π or an h_0 pattern.

For the sake of concreteness we have analyzed the Swift-Hohenberg model [11] that has been widely used [7] to give a qualitative description of stable structures appearing in systems exhibiting inversion symmetry. Its defining kinetic equation is

$$\frac{\partial u}{\partial t} = ru - [\nabla^2 + q_c^2]^2 u - u^3. \qquad (8)$$

For this model, $\mathbf{U}_0 = u = 0$, $r_T = 0$, and $r_p = q_c^4$ [12]. The amplitudes of the predicted hexagonal mixed-mode solutions are shown on the bifurcation diagram (Fig. 1). They are stable for $r > \frac{51}{74}q_c^4$ and thereafter coexist with stripes, and with the homogeneous states $\mathbf{U}_\pm = u_\pm = \pm\sqrt{r - q_c^4}$ as soon as $r > \frac{3}{2}q_c^4$. Hence there exists a large multiplicity of states. This diagram is different from that of the standard "hexagons-stripe" competition that occurs near the instability point of systems lacking inversion symmetry [13]. These stable hexagonal mixed-mode states can only be reached by finite amplitude perturbations from the HSS or by quenches ($\Delta\mu$) through the diffusive instability. For the same operating conditions, one may therefore obtain either a triangular ($\mathcal{R}^0_{M3}, R_{M3}$) or a honeycomb ($\mathcal{R}^\pi_{M3}, R_{M3}$) lattice, or stripes ($R_1$), depending on the initial conditions. Stable fronts between h_0 and h_π patterns can then be constructed, and an example is shown in Fig. 2. Convection experiments where the non-Boussinesq character is easily tunable have recently been carried out with SF_6 near its critical point [14]. When non-Boussinesq effects are negligible, a scenario where hexagonal patterns appear further from threshold than the rolls has been observed [15]. Coexistence of h_0 and h_π may then also be present. At larger values of the control parameter, when the first overtone ($|\mathbf{q}| = 2q_c$) becomes quasineutral, its coupling with the zero mode A_0 also induces a secondary subharmonic bifurcation of the stripes that have appeared at $r = r_T$.

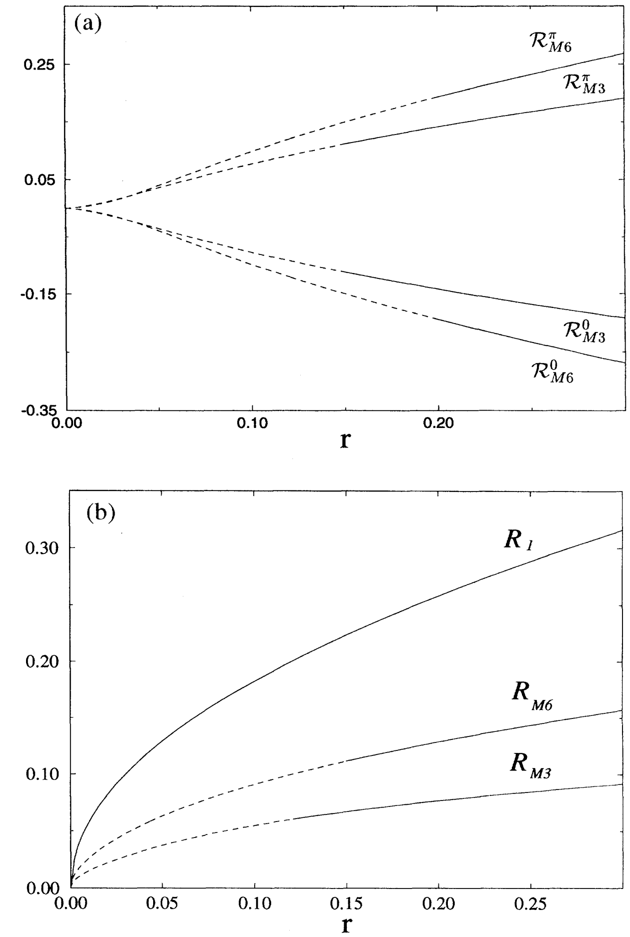

FIG. 1. (a) Moduli of the amplitudes for the 3D Swift-Hohenberg model with $q_c = 0.5$. R_1 is the standard "stripes" solution branch. R_{M3} and R_{M6} are the spatially modulated components of the hpc and bcc mixed modes. (b) Amplitudes of the zero mode components of the hpc and bcc mixed modes for the 3D Swift-Hohenberg model with $q_c = 0.5$.

In 3D the same effect biases the onset of all patterns characterized by modes forming equilateral triangles. Here also the phases of the modes belonging to a resonant triad adjust in such a way that the quadratic coupling terms are destabilizing. The following types of stable patterned solutions may then be obtained: (i) pure structures consisting of a smecticlike ordering of lamellae ($m = 1$) and appearing supercritically at $r = r_T$; (ii) hpc_0 and hpc_π ($m = 3$) hexagonally packed cylindrical mixed

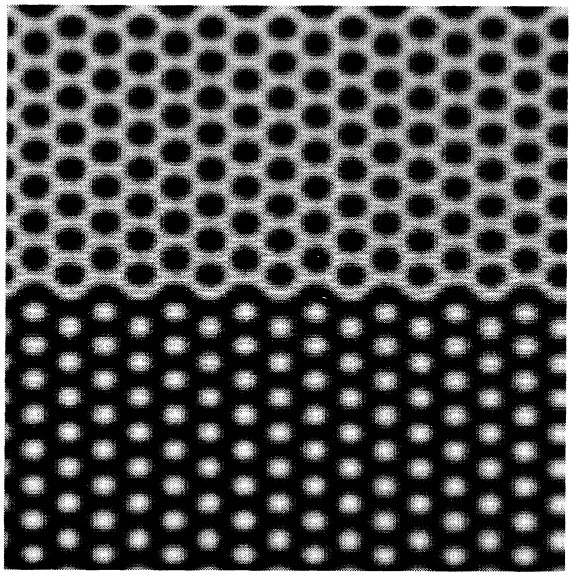

FIG. 2. Stable stationary front separating the two different hexagonal phases obtained for the same condition as in Fig. 1, $r = 0.2$.

modes which are the natural 3D extension of the h_0 and h_π mixed modes described above, and which also obey [Eq. (7)]; and (iii) mixed modes resulting from the superposition of modes of bcc symmetry ($m = 6$) and a uniform mode $A_0 = \mathcal{R}_{M6}$. The amplitudes $\mathcal{R}_{M6}$ and R_{M6} are given by the solutions of

$$\omega_0 \mathcal{R}_{M6} - \mathcal{R}_{M6}^3 - 6\beta_1 R_{M6}^2 \mathcal{R}_{M6} \pm 8\beta_2 R_{M6}^3 = 0,$$

$$\mu - (g_1 + 5g_2) R_{M6}^2 - g_3 \mathcal{R}_{M6}^2 \pm 2g_4 \mathcal{R}_{M6} R_{M6} = 0, \quad (9)$$

where the signs, respectively, correspond to bcc_π and bcc_0 type patterns. For the zero phase structures (bcc_0) the maxima of one component of the field are located at the corners and center of a cube. They are sited in the interstices for the π phase pattern. The same remark as above applies for the other component. For the same given value of the parameters one may thus obtain the following set (Fig. 1) of structures

$$\mathrm{bcc}_\pi(\mathcal{R}_{M6}^\pi, R_{M6}),\ \mathrm{hpc}_\pi(\mathcal{R}_{M3}^\pi, R_{M3}),\ \text{lamellae } (R_1),$$
$$\mathrm{hpc}_0(\mathcal{R}_{M3}^0, R_{M3}),\ \mathrm{bcc}_0(\mathcal{R}_{M6}^0, R_{M6}).$$

This succession is similar to that reported in the phase separation of block copolymers [16]. We may thus conclude that the interaction between a diffusive-type instability and a steady-state bifurcation favors the onset of resonant structures, *even in systems exhibiting inversion symmetry*.

We now consider the effect of the resonant zero mode on the physical systems cited in the introduction and that lack such inversion symmetry. As is well known the amplitude equations, Eq. (4), then contain the intrinsic quadratic coupling terms $v A_k^* A_j^* \delta(\mathbf{k}_i + \mathbf{k}_j + \mathbf{k}_k)$ that already induce a subcritical branch of hexagonal patterns near $r = r_T$. When $v \neq 0$ the distinction between pure patterned states and hexagonal mixed modes is irrelevant since the quadratic term also induces the onset of harmonics already near the instability point $r = r_T$. In this case the interaction with the quasineutral zero mode near $r = r_p$ leads to a renormalization of the quadratic coupling coefficient $v_{\mathrm{eff}} = v - g_4 A_0$. It now depends on the control parameter through A_0. This dependence may lead to the onset of new bifurcation scenarios. For example, when the system is prepared in the trivial state $\mathbf{U} = \mathbf{U}_0$ and v (>0) is sufficiently small, the following sequences of patterns may be observed in 2D by increasing the bifurcation parameter. A subcritical h_0 triangular structure (R_h^0) with $A_0 > 0$ prevails near $r = r_T$ and then becomes unstable to the formation of stripes (R_s) in the range of values of the parameter where $v_{\mathrm{eff}} \approx 0$. Patterns of hexagonal symmetry may reappear at still higher values of v: h_0 (with $v_{\mathrm{eff}} < 0$) and $h_\pi(R_h^\pi)$ with $A_0 > 0$ ($v_{\mathrm{eff}} > 0$). They correspond to the remnants of the mixed-mode solutions discussed above in the limit $v = 0$. For a larger value of v, the h_0 structures remain stable in all the range of values of the bifurcation parameter r. Such scenarios have been obtained for the generalized Swift-Hohenberg model with a quadratic term vu^2 [17]. The corresponding bifurcation diagram is displayed in Fig. 3. A complete analysis of all the possible bifurcation diagrams will be given elsewhere. This reappearance of h-type solutions should not be confused with the reentrant hexagons that have been observed in some chemical models where the nonlinear terms depend on the bifurcation parameter [18]. This latter effect strongly depends on the specification of the models, whereas the mechanism of reentrance induced by the coupling with a zero mode is more generic. Interestingly a sequence of structures analogous to those presented in Fig. 3 has recently been obtained

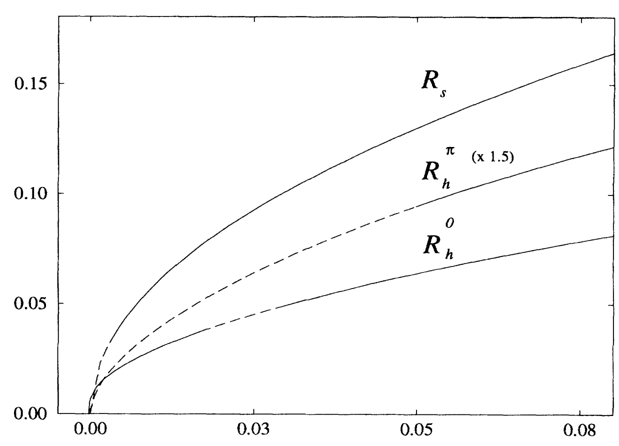

FIG. 3. Moduli of the amplitude for the 2D generalized Swift-Hohenberg model with $q_c = 0.5$ and $v = 0.05$. R_s and R_h, respectively, correspond to striped or hexagonal structures.

in the CIMA reaction [19]. Similar reentrant inverted hexagonal patterns have also been reported in the case of thermal convection in a liquid crystal sample near the nematic-isotropic phase transition [20].

The coupling presented in this note will be operative in all degenerated bifurcations involving a diffusive instability and a zero mode, such as in the Turing–saddle-node interaction or the formation of supersolids [21]. The longitudinal and transverse modes in anisotropic media or systems submitted to ramps of the control parameter also fall in this class of problem [22].

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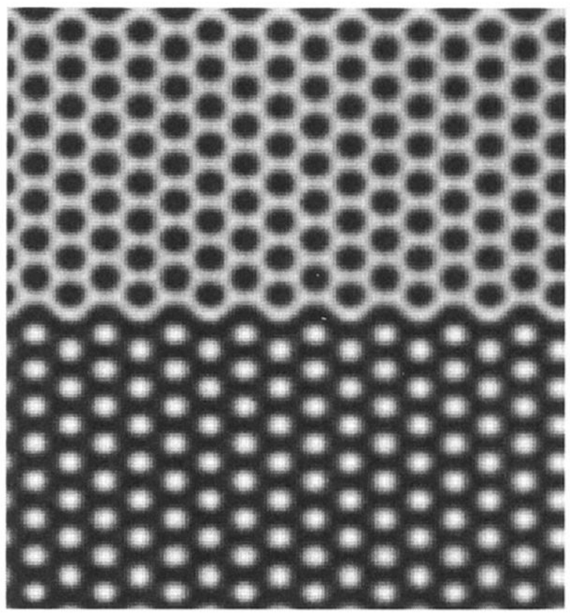

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