Two-layer model showing a variety of pattern types near nonequilibrium phase transitions

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It is shown analytically as well as by direct numerical integration that a simple model of two coupled two-dimensional order parameter equations may lead to the formation of a large variety of basic pattern types. The same pattern symmetries have been obtained recently in reaction-diffusion systems and are known as mixed states or beans and triangles. Finally we discuss a nonvariational extension of our model that may also show time-stable rhombic structures. [S1063-651X(96)03305-3]

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I. INTRODUCTION

A large number of spatially extended pattern forming systems far from equilibrium were extensively studied both experimentally and theoretically in the last years (for a review see [1] and references therein). Until now interest was mainly focused on two spatial dimensions. Two-dimensional (2D) order parameter equations could be derived directly for the case of the three-dimensional (3D) convection instability in various systems and show good agreement between 3D experiments or direct 3D-numerical solutions of the hydrodynamic equations [2]. Beyond the reduction on two spatial dimensions the order parameter equations have a unifying character and allow for the classification of quite different systems into a few classes of basic instabilities [3,4]. In this paper we shall be concerned with an instability of a temporally and spatially homogeneous system that selects a finite critical wave number in an isotropic two-dimensional plane, leading to a typical length scale in real space. Moreover, the unstable modes may grow with a real rate, leading very often to transient states that end in stationary, spatially more or less ordered structures. We adopt the nomenclature of [1] and call this a type III_s instability. Type III_s instabilities are encountered in many systems. We mention convection in a pure fluid [4], transverse optical patterns in a laser with a nonlinear medium [5], and the chemical structures caused by the Turing instability [6]. The normal form of type III_s reads

$$\partial_t \Psi(\mathbf{x},t) = L(\varepsilon,\Delta)\Psi(\mathbf{x},t) + N(\Psi(\mathbf{x},t),\delta), \tag{1}$$

where $\Psi(\mathbf{x},t)$ is the real order parameter field depending on the two spatial coordinates $\mathbf{x}=(x,y)$ and L is a linear differential operator that accounts for the linear growth of modes with a wavelength in the vicinity of $\lambda_c = 2\pi$ [7]

$$L(\varepsilon,\Delta) = \varepsilon - (1+\Delta)^2. \tag{2}$$

Here Δ is the 2D Laplacian and ε is the control parameter where $\varepsilon \geqslant 0$ denotes instability of the homogeneous state and the emergence of spatial structures. $N(\Psi,\delta)$ is a nonlinear (in general, also nonlocal) function of Ψ as well as of its spatial derivatives, δ stands for one or more coefficients that may be controlled also from the outside. Without further requirement to the symmetry of N, the generic patterns produced by (1,2) at onset are regular hexagons [8], in agreement with the first experimental results in convection by Bénard [9], as well as in reaction diffusion systems, there found for the first time by Castets *et al.* [10] and later by Ouyang and Swinney [11] examining the chlorite-iodide-malonic-acid (CIMA) reaction. If the condition $N(\Psi,\delta) = -N(-\Psi,\delta)$ is fulfilled, stripes (or rolls) are stable above threshold. A general solution of (1) in lowest order of ε reads

$$\Psi(\mathbf{x},t) = \sum_j^3 \xi_j(t)\exp(i\mathbf{k}_j\mathbf{x}) + \text{c.c.}, \tag{3}$$

with $\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3=\mathbf{0}$ and $|\mathbf{k}_j|=\mathbf{1}$. In (3) $\xi_1=\xi_2=\xi_3=Z$ corresponds to (+)hexagons (Z real, $Z>0$), (−)hexagons ($Z<0$), and triangles ($\mathrm{Re}Z=0$, $\mathrm{Im}Z\neq 0$). Here (+,−)hexagon denotes the case where Ψ is (positive, negative) valued in the center of each hexagon. The case of only one nonvanishing ξ_j corresponds to stripes. Rhombic cells are obtained if two amplitudes in (3) are equal and the third vanishes. Finally a mixture between hexagons and stripes, named ''mixed state '' [12] or, synonymously ''beans'' [13] is presented by $\xi_1>\xi_2=\xi_3$ and real positive ξ_j. A linear analysis in the vicinity of (3) shows that (+,−)hexagons and/or stripes may be stable, depending on the form of $N(\Psi)$ and of the value of ε [14]. All other structures are unstable.

II. THE MODEL

Nevertheless triangles, beans, and rhombs were found by different groups in the CIMA reaction [12,15,16]. Recently, De Kepper [15] argued that the reason for the occurrence of these types could be the existence of the spatial extension of the chemical reactor vertical to the plane where the patterns are usually visualized (Fig. 1). The vertical size could be smaller or of the same order than the critical wavelength. To sustain the reaction far from equilibrium, the reactor must be fed continuously from the outside. Therefore a gradient in the control parameters, a so-called parameter ramp may occur, leading to different selected patterns in the different vertical planes. To demonstrate the stability of triangles and beans we studied the simplest possible two-layer model obtained by two linearly coupled Haken equations [3]

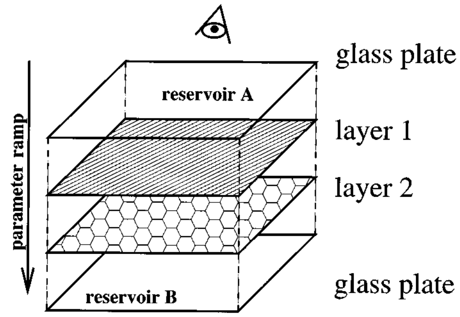

FIG. 1. Schematic drawing of the chemical reactor. At the glass plates, the concentration of the control variables is kept constant by coupling to the reservoirs A and B from the outside and a parameter ramp can occur in the vertical direction. This may lead to the stabilization of different pattern types in the two layers. The visually obtained pattern is simply regarded as a superposition of the patterns formed in each layer.

$$\partial_t\Psi_1(\mathbf{x},t)=L(\varepsilon_1,\Delta)\Psi_1(\mathbf{x},t)+\alpha\Psi_2(\mathbf{x},t)+\delta_1\Psi_1^2(\mathbf{x},t)-\Psi_1^3(\mathbf{x},t),$$

$$\partial_t\Psi_2(\mathbf{x},t)=L(\varepsilon_2,\Delta)\Psi_2(\mathbf{x},t)+\alpha\Psi_1(\mathbf{x},t)+\delta_2\Psi_2^2(\mathbf{x},t)-\Psi_2^3(\mathbf{x},t), \tag{4}$$

where $i=1,2$ denotes the two vertically stratified layers. The linear coupling reflects diffusion ($\alpha>0$) of the concentration fields in vertical direction. To model parameter ramps of the control parameters we allow for different values of δ and ε in the two layers. Looking through both layers, the visualized structure is given by a superposition of the patterns in each layer

$$\Psi(\mathbf{x},t)=\Psi_1(\mathbf{x},t)+\Psi_2(\mathbf{x},t). \tag{5}$$

Our model (4) has a potential of the form

$$V[\Psi_1,\Psi_2]=-\frac{1}{2}\int dxdy\{\Psi_1L(\varepsilon_1,\Delta)\Psi_1+\Psi_2L(\varepsilon_2,\Delta)\Psi_2+2\alpha\Psi_1\Psi_2+\tfrac{2}{3}(\delta_1\Psi_1^3+\delta_2\Psi_2^3)-\tfrac{1}{2}(\Psi_1^4+\Psi_2^4)\}. \tag{6}$$

The most interesting results discussed in the next section are obtained for substantially different values for δ_i, namely, where the two coefficients have opposite signs. This can be justified by the assumption that the control parameters of the system are in a region where δ of a one layer model goes through zero, i.e., where the transition from (+) to (−) hexagons occurs via stripes (this situation is similar to convection in a pure fluid, where the transition from gas hexagons to liquid hexagons can be obtained for certain fluid parameters [17]). The ''unfolding'' of δ in the third dimension by means of a parameter ramp may give rise for a variety of stable basic structures as will be shown now.

III. RESULTS

To demonstrate the stability of the above explained basic pattern types we introduced the expressions

$$\Psi_1(\mathbf{x},t)=\sum_j^3 \xi_j(t)\exp[i\mathbf{k}_j(\mathbf{x}+\mathbf{b}_1)]+\text{c.c.},$$

$$\Psi_2(\mathbf{x},t)=\sum_j^3 \eta_j(t)\exp[i\mathbf{k}_j(\mathbf{x}+\mathbf{b}_2)]+\text{c.c.} \tag{7}$$

into (6) and search for the absolute minimum of V with respect to the amplitudes ξ_i, η_i and the shift $\mathbf{b}_1$ (due to translation symmetry of the whole system only the relative shift $\mathbf{b}_1-\mathbf{b}_2$ is important and we may put arbitrarily $\mathbf{b}_2=0$). The resulting perfect patterns may be classified as above. Figure 2 shows a phase diagram in the δ_i plane for fixed

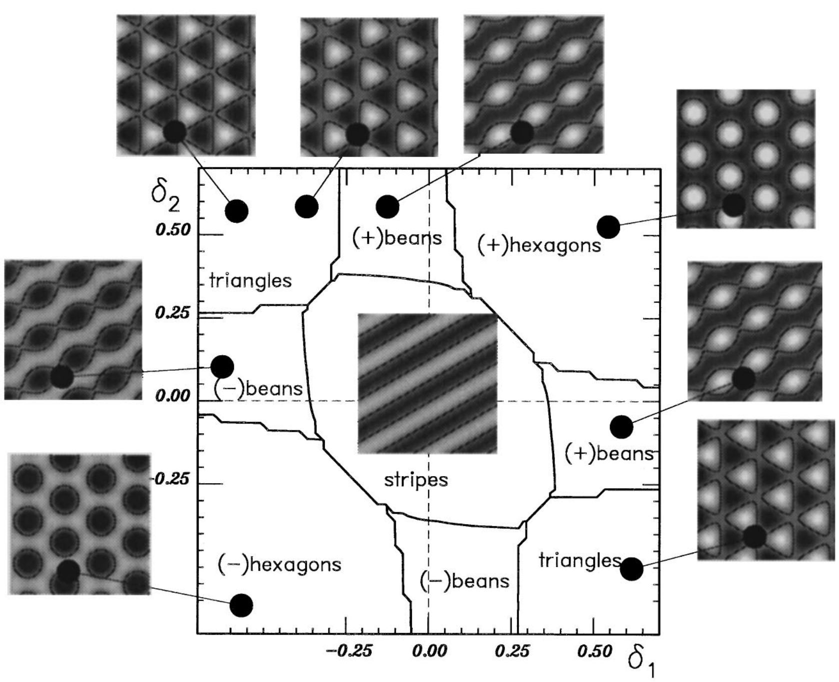

FIG. 2. Parameter plane for fixed $\varepsilon_i=0.07$ and $\alpha=0.03$. The patterns plotted are those which minimize the potential (6) and that are therefore globally stable.

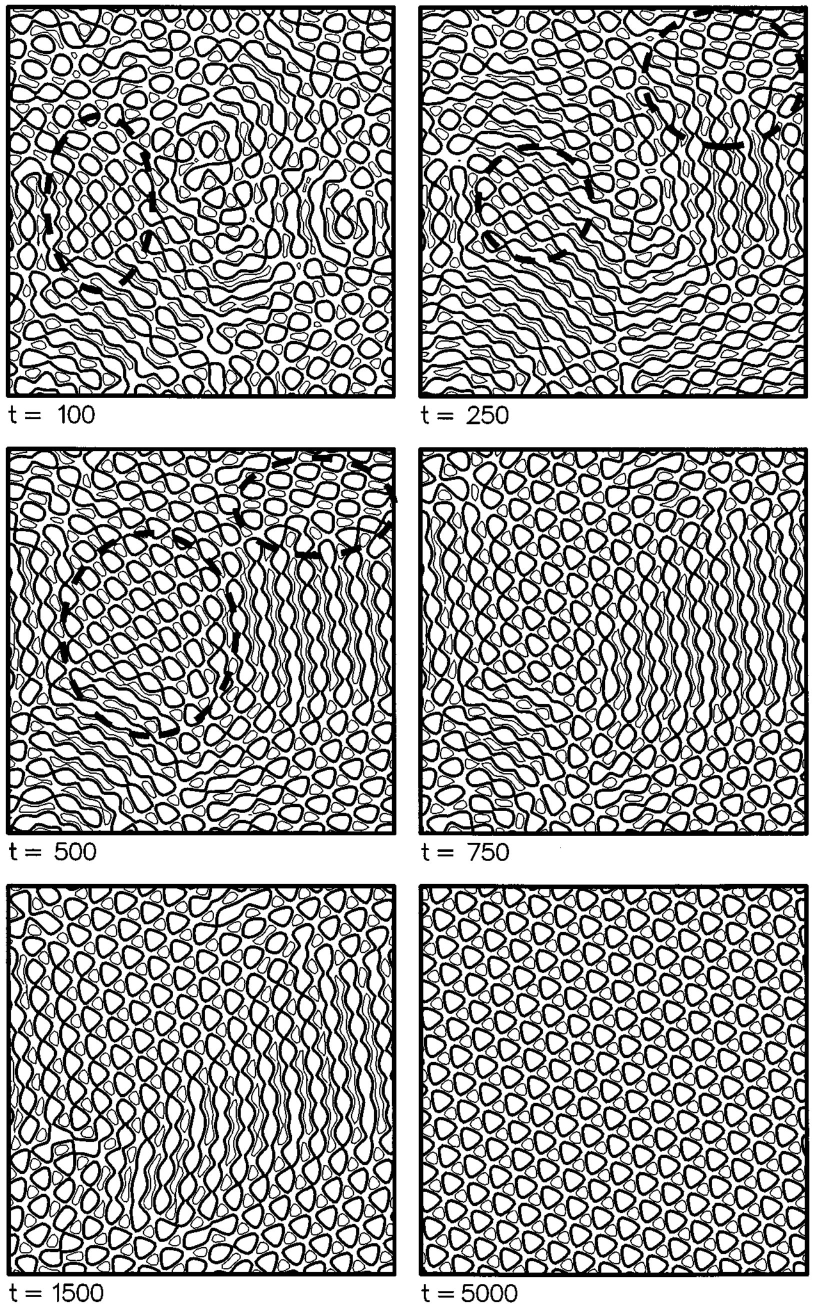

FIG. 3. Numerical solution of the model (4) in a domain between beans and triangles where rhombs are metastable ($\delta_1=-0.7, \delta_2=0.35$). They are formed as transients in the circled areas and vanish eventually. The state at $t=5000$ is stable. Plotted are contour lines of the superposition [Eq. (5)], bold: $\Psi=0$, thin: $\Psi=\Psi_{min}/2$. Time is in dimensionless units of Eqs. (4).

$\varepsilon_1=\varepsilon_2=0.07$ and $\alpha=0.03$. If the coefficients δ_i have opposite signs and about the same values pattern selection goes to (+)hexagons in one layer, (−)hexagons in the other. In the coupled system, this ''frustration'' results either in a stripe pattern (small δ_i) in both layers or in the formation of the two oppositely orientated hexagons (large δ_i). The latter have a relative shift $|\mathbf{b}_1|=\lambda_c/3$ and $\mathbf{b}_1$ parallel to one of the $\mathbf{k}_j$. A superposition according to (5) yields a symmetric (with respect to $\Psi\rightarrow-\Psi$) triangular structure. Asymmetric triangles are obtained near the borders to the bean states. Here, a superposition of hexagons and triangles in one layer, pure hexagons in the other, takes place.

Mixed states or beans are obtained if one layer shows hexagons, the other stripes. This is the case for one δ_i large, the other near zero. Experimental evidence for such a configuration is given in [13]. There the two planes with the different structures can be distinguished clearly and the pattern seen by superposition can be identified as a mixed state.

Rhombs are only metastable in our model and can never totally minimize the potential. Depending on parameters, beans or asymmetric triangles are globally stable and therefore eventually the preferred structure. Thus rhombs are observed mainly during long transient phases. To demonstrate this we performed a direct numerical solution of (4). The method, a pseudospectral semi-implicit scheme was developed and used before for the one layer problem and is described in detail in [18]. Figure 3 shows the spatiotemporal evolution of randomly distributed initial patterns in both layers in a parameter region where rhombs are metastable. They are present in large regions during the transient phase but finally vanish and give way to a regular time-stable structure of triangles. At first sight this seems to be in contradiction

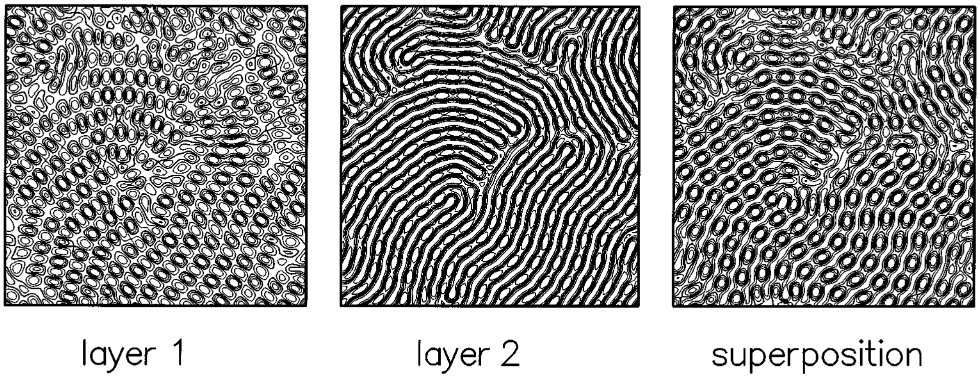

FIG. 4. Time stable but spatially disordered state of (8). Note that rhombs are now stable in layer 1. ($\delta=0.4,\varepsilon_1=0,\varepsilon_2=0.2.$) Contour lines of Ψ_1 (left), Ψ_2 (middle) and the superposition [Eq. (5)], right.

with [16], where stable rhombs are selected under certain conditions. Gunaratne, Ouyang, and Swinney argued quite right that metastable rhombs may follow from a 2D-variational model. Then they state that the linear stability should be sufficient for the selection of rhombs. In our opinion, and this is also supported by a numerical integration of any variational model including spatial diffusion, the eventually selected pattern is always the one leading to the lowest potential. If there exist some regions where the globally stable pattern has been formed, the domain walls will move in such a way that these regions blow up until the metastable domains have completely vanished. Nevertheless stable rhombs found experimentally [16] can be explained in several ways: (1) The initially prepared rhombic pattern is perfect and no seed for the globally stable pattern exists. This seems to be the case in Fig. 6(b) of [16]. (2) domain boundaries between rhombs and the globally stable pattern are pinned by the small scale structure or defects (on pinning of a wall between stripes and hexagons see, e.g., [19]). (3) The dynamics of the pattern cannot be described by a variational model, as discussed in the following section. This view is supported by the fact that rhombs are, until now, never obtained in the Bénard convection. Convection in a pure fluid just above onset behaves very much like a variational system [20] and should give no reason for the selection of a metastable pattern (see, e.g., [22]).

An extension of (4) could be the inclusion of nonlinear coupling terms between the two layers. This could also give rise to nonvariational models and the stabilization of other structures, e.g., rhombs. Nonvariational systems may also show stable states that are far from being perfect but exhibit a lot of defects and grain boundaries between stripes with different orientation in both layers. Figure 4 shows a time-stable state of a model with a nonvariational and nonlinear extension

$$\partial_t\Psi_1(\mathbf{x},t)=L(\varepsilon_1,\Delta)\Psi_1(\mathbf{x},t)+\delta(\Psi_1-\Psi_2)^2-\Psi_1^3(\mathbf{x},t),$$

$$\partial_t\Psi_2(\mathbf{x},t)=L(\varepsilon_2,\Delta)\Psi_2(\mathbf{x},t)+\delta(\Psi_1-\Psi_2)^2-\Psi_2^3(\mathbf{x},t). \tag{8}$$

Let $C(\mathbf{x},z_i)\equiv\Psi_i(\mathbf{x})$ be a three-dimensional concentration field with $z_i\propto i$, then the quadratic term in (8) may be justified by introducing a vertical gradient of the form $[\partial_z C(\mathbf{x},z)]^2$. Now layer one shows clearly the stable rhombs, layer two a mixed state that is also dominating the superposition. In contrast to the transient evolution of the potential model where in the long time limit all defects and domain boundaries eventually vanish and a perfect pattern remains stable, the steady state obtained with (8) is spatially quite irregular.

IV. CONCLUSIONS

We showed that even the simplest model of two linearly coupled 2D-order parameter equations may account for a large variety of patterns. The existence of pattern types like triangles or mixed states follows naturally, and both of them were obtained experimentally in Turing systems. More complicated models can be discussed: A fully three-dimensional extension of (1,2) in the case of a relatively large vertical extension of the reactor was studied in [23] with respect to hexagons orientated in the basic planes of the three spatial dimensions. Since this problem is much more complex then our model, an analysis in the way of (7) is not possible and a full classification of stable basic patterns is still lacking.

The results of the present paper support strongly the hypothesis of De Kepper: triangles, mixed states, and even rhombs, although unstable or at most metastable solutions of the generic 2D-order parameter equation of a type III_s instability, can be shown to be globally stable in a simple model that takes the spatial dimension vertically to the reactor into account and that allows for the modeling of parameter ramps in that direction.

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