General theory of instabilities for patterns with sharp interfaces in reaction-diffusion systems

C. B. Muratov
Department of Physics, Boston University, Boston, Massachusetts 02215

V. V. Osipov
Department of Theoretical Physics, Russian Science Center "Orion," 2/46 Plekhanov St., Moscow 111123, Russia
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An asymptotic method for finding instabilities of arbitrary d-dimensional large-amplitude patterns in a wide class of reaction-diffusion systems is presented. The complete stability analysis of two- and three-dimensional localized patterns is carried out. It is shown that in the considered class of systems the criteria for different types of instabilities are universal. The specific nonlinearities enter the criteria only via three numerical constants of order 1. The analysis performed explains the self-organization scenarios observed in the recent experiments and numerical simulations of some concrete reaction-diffusion systems.

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I. INTRODUCTION

In the last two decades the problem of pattern formation and self-organization has become a paradigm of modern science [1–8]. Patterns are observed in a wide variety of physical systems, such as gas and electron-hole plasmas; various semiconductor, superconductor, and gas-discharge structures; some ferroelectric, magnetic, and optical media; systems with uniformly generated combustion material (see [5–7,9] and references therein). Pattern formation and self-organization are most conspicuous in chemical and biological systems (see [1–5] and references therein). As a rule, all these systems are extremely complicated. In order to describe pattern formation phenomena in them a number of simplifications are made. The majority of the simplified models reduce to a pair of reaction-diffusion equations of the activator-inhibitor type [5–7]:

$$\tau_\theta \frac{\partial \theta}{\partial t} = l^2 \Delta \theta - q(\theta, \eta, A), \tag{1}$$

$$\tau_\eta \frac{\partial \eta}{\partial t} = L^2 \Delta \eta - Q(\theta, \eta, A), \tag{2}$$

where θ and η are the distributions of the activator and the inhibitor, respectively; $q(\theta,\eta,A)$ and $Q(\theta,\eta,A)$ are certain nonlinear functions; l and L are the characteristic length scales, and τ_θ and τ_η are the characteristic time scales of θ and η, respectively; and A is the control parameter. The well-known models of certain autocatalytic reactions, such as the Brusselator [1], the two-component version of the Oregonator [2] and the Gray-Scott [10] models, the classical model of morphogenesis proposed by Gierer and Meinhardt [11], and the piecewise-linear [12,13] and FitzHugh-Nagumo [14,15] models describing the propagation of impulses in the nerve tissue are special cases of Eqs. (1) and (2). These models are most widely used in the analytical investigations of different types of patterns [11–26].

The main self-organization phenomenon in the considered systems is spontaneous transformation of one type of pattern to another as certain parameters of the system are varied. Self-organization scenarios become extremely diverse in two- and three-dimensional systems. In this situation the most interesting are the spontaneous transformations of simple patterns, especially localized steady patterns [autosolitons (AS's)], into much more complicated ones. Many different types of these transformations were recently observed in experiments with semiconductor and gas-discharge structures [27–29], chemical reaction-diffusion systems [30–32], and in numerical simulations [33–35]. At the same time, a general theory of such transformations is absent.

The general asymptotic method of constructing solutions in the form of large-amplitude localized, periodic, and more complex one-dimensional patterns, and the qualitative analysis of their stability in the systems described by Eqs. (1) and (2), were developed by Kerner and Osipov [5,6,36–42]. A more formal and mathematically rigorous analysis of one-dimensional patterns was carried out by Nishiura, Mimura, and their co-authors [43–45]. Static, pulsating, and traveling large-amplitude patterns in simple reaction-diffusion systems have been studied in detail [13,16–21,25,12,46].

In two and three dimensions Kerner and Osipov constructed asymptotic solutions for radially symmetric patterns, and also analyzed the stability of one-dimensional patterns in higher dimensions [5,6,39–42]. Ohta, Mimura, and Kobayashi developed an approach which allowed them to study the stability of one-dimensional and radially symmetric patterns in two- and three-dimensional versions of a simple piecewise-linear model of a reaction-diffusion system [47]. This approach was further developed by Petrich and Goldstein, who applied it to a version of the FitzHugh-Nagumo model [23].

In this paper we develop a systematic procedure to find the bifurcation points of an arbitrary d-dimensional pattern. Using this procedure, we analyze the stability of the major types of patterns in arbitrary dimensions. On the basis of this analysis, we draw conclusions about possible scenarios of the transformations of patterns.

Our paper is organized as follows. In Sec. II we generalize the method of constructing the asymptotic solution for one-dimensional and radially symmetric patterns developed in Refs. [5,6,36–42] to the case of arbitrary d-dimensional

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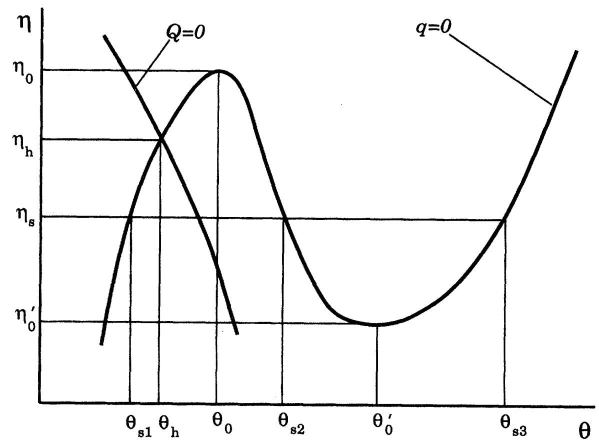

FIG. 1. Qualitative form of the nullclines of Eqs. (3) and (4).

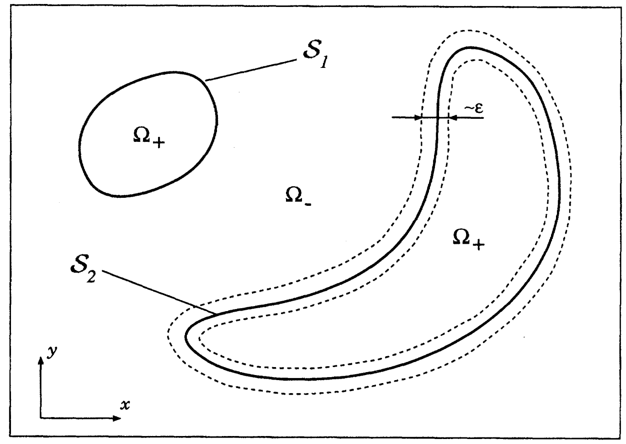

FIG. 2. "Hot" and "cold" regions forming a pattern. The walls of the pattern are localized in the region of order ϵ around $\mathcal{S}_i$.

patterns. In Sec. III we present the derivation of the general dispersion relation governing the stability of an arbitrary pattern. In Sec. IV we apply the obtained results to one-dimensional AS's in higher dimensions. In Sec. V we analyze the instabilities of the spherically symmetric AS in three dimensions, and the cylindrically symmetric AS in two and three dimensions. In Sec. VI we summarize the results obtained and discuss their implications for the evolution of patterns and also give comparisons with the experimental and numerical data.

II. ASYMPTOTIC SOLUTIONS FOR ARBITRARY *d*-DIMENSIONAL PATTERNS

If we choose L and τ_η as the units of length and time, we can write Eqs. (1) and (2) as

$$\alpha \frac{\partial \theta}{\partial t} = \epsilon^2 \Delta \theta - q(\theta, \eta, A), \tag{3}$$

$$\frac{\partial \eta}{\partial t} = \Delta \eta - Q(\theta, \eta, A), \tag{4}$$

where $\epsilon \equiv l/L$ and $\alpha \equiv \tau_\theta / \tau_\eta$ are the ratios of the characteristic lengths and times of the activator and the inhibitor, respectively. The boundary conditions for Eqs. (3) and (4) may be neutral or periodic.

Kerner and Osipov developed a qualitative theory of large-amplitude pattern in reaction-diffusion systems [36–40] (for a comprehensive review on the subject see Refs. [5–7]). They showed that the overall type of patterns is determined by the values of ϵ and α, and the form of the nullcline of Eq. (3), that is, the dependence $\eta(\theta)$ implicitly determined by the equation $q(\theta, \eta, A) = 0$ for a fixed value of A. For many systems where patterns may form this nullcline is N or inverted N (Fig. 1).

According to the general qualitative theory [5,6], when $\epsilon \ll 1$ and $\alpha \gg 1$, only static patterns may form in the system; when $\alpha \ll 1$ and $\epsilon \gg 1$ only traveling patterns may form; and when both $\epsilon \ll 1$ and $\alpha \ll 1$, all types of patterns — static, traveling, and pulsating — may form.

From the mathematical point of view the fact that θ is the activator means that in some range of the system's parameters $q'_\theta < 0$. In N systems this condition is satisfied for $\theta_0 \lesssim \theta \lesssim \theta'_0$ (see Fig. 1). The fact that η is the inhibitor means that the following conditions hold [5,6]:

$$Q'_\eta > 0, \quad q'_\eta Q'_\theta < 0, \tag{5}$$

and in the whole range of the system's parameters the derivatives Q'_θ, Q'_η, and q'_η do not change signs.

The systems we are considering have a unique homogeneous state $\theta = \theta_h$ and $\eta = \eta_h$, where θ_h and η_h satisfy

$$q(\theta_h, \eta_h, A) = 0, \quad Q(\theta_h, \eta_h, A) = 0. \tag{6}$$

The homogeneous state is stable for $A < A_0$, where A_0 is the point where $\theta_h = \theta_0$ (see Fig. 1) [5,6].

As follows from the qualitative theory [5,6], the condition $\epsilon \ll 1$ is necessary for the existence of AS's and other large-amplitude patterns in the considered reaction-diffusion systems. This fact allows one to use ϵ as a natural small parameter and construct asymptotic solutions by means of the singular perturbation theory [5,6,38,41]. Kerner and Osipov have shown that as $\epsilon \to 0$, a pattern looks like a collection of "hot" (high values of the activator) and "cold" (low values of the activator) regions, separated by walls whose width is of the order of ϵ [5,6,37–40]. Thus, in the limit $\epsilon \to 0$ any pattern in a d-dimensional N system can be described as a $(d-1)$-dimensional manifold $\mathcal{S}$, corresponding to the walls of the pattern, which separates hot and cold regions Ω_+ and Ω_-, respectively (Fig. 2). In general, $\mathcal{S}$ is a collection of an (infinite) number of disconnected orientable submanifolds $\mathcal{S}_i$.

Let us introduce the orthogonal curvilinear coordinates around each submanifold $\mathcal{S}_i$. For a point x let ρ_i be the distance from x to $\mathcal{S}_i$, and the $(d-1)$-dimensional coordinate $\vec{\xi}_i$ the projection to the submanifold $\rho_i = \text{const}$. The value of ρ_i is assumed to be positive if $x \in \Omega_+$ and negative otherwise. In the region of size $\sim \epsilon$ around each $\mathcal{S}_i$ the variation of the inhibitor in the direction perpendicular to $\mathscr{S}_i$ and the variation of the activator along $\mathscr{S}_i$ are negligible compared to the variation of the activator in the direction perpendicular to $\mathscr{S}_i$ [5,6,47]. Therefore, in the vicinity of $\mathscr{S}_i$ the stationary Eq. (3) can be approximately written as

$$\epsilon^2 \frac{d^2\theta_i}{d\rho_i^2} + \epsilon^2 K_i(\rho_i,\xi_i)\frac{d\theta_i}{d\rho_i} = q(\theta_i, \eta_s^i, A), \tag{7}$$

where $K_i(\rho_i,\xi_i)$ is the curvature of $\mathscr{S}_i$ at a point with the curvilinear coordinates ρ_i and ξ_i. The boundary conditions for θ_i in Eq. (7) are

$$\theta_i(-\infty) = \theta_{s1}^i, \quad \theta_i(+\infty) = \theta_{s3}^i, \quad \theta_i(0) = \theta_{s2}^i, \tag{8}$$

where θ_{sk}^i satisfy $q(\theta_{sk}^i, \eta_s^i, A) = 0$, with $\theta_{s1}^i < \theta_{s2}^i < \theta_{s3}^i$, and η_s^i satisfies the consistency condition

$$\epsilon^2 \int_{-\infty}^{+\infty} K_i(\rho_i,\xi_i)\left(\frac{d\theta_i}{d\rho_i}\right)^2 d\rho_i = \int_{\theta_{s1}^i}^{\theta_{s3}^i} q(\theta, \eta_s^i, A)\, d\theta, \tag{9}$$

which follows from Eq. (7) if we multiply it by $d\theta_i/d\rho_i$ and integrate over ρ_i. Of course, the infinities in Eq. (8) actually mean that the boundary conditions for Eq. (7) should be satisfied sufficiently far from $\mathscr{S}$, namely, for $|\rho| \gg \epsilon$. Note that in the equations above ξ_i appears only as a parameter.

When $K_i \to 0$ the solution of Eqs. (7)–(9) naturally transforms to the one-dimensional sharp distribution (inner solution) [5,6,37,38]:

$$\epsilon^2 \frac{d^2\theta_{sh}}{d\rho_i^2} = q(\theta_{sh}(\rho_i), \eta_s, A), \tag{10}$$

where η_s is a constant determined by the equation

$$\int_{\theta_{s1}}^{\theta_{s3}} q(\theta, \eta_s, A)\, d\theta = 0, \tag{11}$$

and θ_{sk} are constants satisfying $q(\theta_{sk}, \eta_s, A) = 0$. The boundary conditions for Eq. (10) are given by Eq. (8) with $\eta_s^i = \eta_s$ and $\theta_{sk}^i = \theta_{sk}$. Thus Eqs. (7)–(9) describe the sharp distributions of the activator around $\mathscr{S}_i$ and take into account the curvature of $\mathscr{S}_i$.

Far from $\mathscr{S}$ the characteristic length of the activator variation is of order 1, so for $|\rho_i| \gg \epsilon$ the solution of Eqs. (3) and (4) is approximately given by the smooth distributions (outer solutions) $\theta_{sm}^{\pm}(x)$ and $\eta_{sm}^{\pm}(x)$ defined for $x \in \Omega_{\pm}$, respectively, which satisfy [5,6,37–40]

$$\Delta \eta_{sm}^{\pm} = Q(\theta_{sm}^{\pm}, \eta_{sm}^{\pm}, A), \quad q(\theta_{sm}^{\pm}, \eta_{sm}^{\pm}, A) = 0 \tag{12}$$

for $x \in \Omega_{\pm}$, respectively, with the boundary conditions

$$\eta_{sm}^{\pm}(x_i) = \eta_s^i, \quad \frac{\partial \eta_{sm}^{+}}{\partial \rho_i} = \frac{\partial \eta_{sm}^{-}}{\partial \rho_i}, \quad \theta_{sm}^{+}(x_i) = \theta_{s3}^i,$$

$$\theta_{sm}^{-}(x_i) = \theta_{s1}^i \tag{13}$$

for any $x_i \in \mathscr{S}_i$. Note that the shape of $\mathscr{S}$ itself is determined self-consistently via Eqs. (12) and (13).

According to the singular perturbations theory [5,6], for $x \in \Omega_{\pm}$ the asymptotic solution of Eqs. (3) and (4) is given by the following composition of the sharp and smooth distributions:

$$\theta^{\pm}(x) = \theta_{sm}^{\pm}(x) + \sum_i \left[\theta_i(x) - \theta_{s1,3}^i\right], \quad \eta^{\pm}(x) = \eta_{sm}^{\pm}(x), \tag{14}$$

where the plus sign goes with θ_{s3}^i and the minus sign goes with θ_{s1}^i.

III. GENERAL METHOD FOR CALCULATING INSTABILITIES

Let us consider the problem linearized about the static solutions of Eqs. (3) and (4) with respect to the fluctuations

$$\delta\theta(x,t) = \delta\theta(x)e^{i\omega t}, \quad \delta\eta(x,t) = \delta\eta(x)e^{i\omega t}. \tag{15}$$

The equations describing the fluctuations with the frequency ω will become

$$i\alpha\omega\,\delta\theta = \epsilon^2 \Delta\,\delta\theta - q'_\theta(\theta(x),\eta(x))\,\delta\theta - q'_\eta(\theta(x),\eta(x))\,\delta\eta, \tag{16}$$

$$i\omega\,\delta\eta = \Delta\,\delta\eta - Q'_\theta(\theta(x),\eta(x))\,\delta\theta - Q'_\eta(\theta(x),\eta(x))\,\delta\eta. \tag{17}$$

As was shown in the previous section, the solutions $\theta(x)$ and $\eta(x)$ in the form of a static pattern, around which Eqs. (3) and (4) are linearized, are approximately given by Eqs. (14) for sufficiently small ϵ.

According to the general qualitative theory [5,6], the stabilization of a pattern occurs due to the damping effect of the inhibitor on the fluctuations of the activator. It was shown that only those fluctuations of the activator that are localized in the walls of the pattern and lead to their small displacements are dangerous for the pattern's stability. To incorporate this fact into our analysis, let us first consider the fluctuations in the vicinity of $\mathscr{S}_i$ with a fixed distribution of the inhibitor. Putting $\delta\eta = 0$ in Eq. (16), we may write

$$i\alpha\omega\,\delta\theta = -(\hat{H}_\theta^0 + \hat{H}_\theta^1 + \epsilon^2 \hat{S}_i)\,\delta\theta, \tag{18}$$

where

$$\hat{H}_\theta^0 = -\epsilon^2 \frac{\partial^2}{\partial \rho_i^2} + q'_\theta(\theta_{sh}(\rho_i), \eta_s), \tag{19}$$

$$\hat{H}_\theta^1 = -\epsilon^2 \hat{K} + q'_\theta(\theta(x),\eta(x)) - q'_\theta(\theta_{sh}(\rho_i),\eta_s), \tag{20}$$

and the operator $\hat{S}_i$ is the part of the Laplacian acting on the $(d-1)$-dimensional coordinates ξ_i, evaluated at $\mathscr{S}_i$ and taken with the minus sign; $\hat{K}$ is the rest of the Laplacian associated with the curvature of $\mathscr{S}_i$.

The lowest bound eigenstate of the operator $\hat{H}_\theta^0$ is $\delta\theta_0 = d\theta_{sh}/d\rho_i$, which corresponds to the eigenvalue $\lambda = 0$ [5,6,37–40]. Indeed, if we differentiate Eq. (10) with respect to ρ_i, we will get

$$\hat{H}_\theta^0 \frac{d\theta_{sh}}{d\rho_i} = 0. \tag{21}$$

The function $\delta\theta_0 = d\theta_{sh}/d\rho_i$ has no nodes; therefore it corresponds to the lowest eigenstate. The eigenvalues corresponding to the excited states are all of order 1. This means that the excited states, whose eigenvalues are all positive and of order 1, are highly damped and are therefore not important for our analysis of the instabilities [5,6,37–40].

The operators $\hat{H}_\theta^1$ and $\epsilon^2 \hat{S}_i$ in Eq. (18) can be treated as perturbations to the operator $\hat{H}_\theta^0$. Since the unperturbed operator $\hat{H}_\theta^0$ has no dependence on ξ_i, we need to introduce the orthogonal basis of the states corresponding to the surface modes on $\mathscr{S}_i$. As such a basis, we may choose the eigenfunctions of the operator $\hat{S}_i$. Then the dangerous fluctuations $\delta\theta_{sh}$ are linear combinations of the functions $\delta\theta_{il}$, where

$$\delta\theta_{il} = \phi_l(\xi_i) \frac{d\theta_{sh}}{d\rho_i}, \quad \hat{S}_i \phi_l = \nu_{li} \phi_l, \tag{22}$$

and ϕ_l satisfy

$$\int_{\mathscr{S}_i} \phi_l^*(\xi_i) \phi_{l'}(\xi_i) d^{d-1}\xi_i = \delta_{ll'}. \tag{23}$$

Of course, for each $\mathscr{S}_i$ there is its own set of ϕ_l. We will frequently omit the indices such as i and ω, wherever it does not lead to ambiguities, in order to simplify the notation.

Up to now we have ignored the reaction of the inhibitor on the fluctuations of the activator. According to the general qualitative theory, this reaction can be included into our analysis by means of the perturbation theory [5,6]. The main problem here is to correctly find the response of the inhibitor to the dangerous fluctuations of the activator. The formal solution of the problem is of no practical use since one has to diagonalize a complicated operator, nor is the expansion in the eigenfunctions since one has to consider the whole spectrum of the problem. A way to do this is to use the idea of singular perturbation theory and separate $\delta\theta$ into the localized (sharp) and the delocalized (smooth) parts (for a more rigorous derivation see Appendix A):

$$\delta\theta = \delta\theta_{sh} + \delta\theta_{sm}. \tag{24}$$

Far from $\mathscr{S}$ Eq. (16) will become

$$i\alpha\omega\delta\theta_{sm} = -(q'_\theta)_{sm}\delta\theta_{sm} - (q'_\eta)_{sm}\delta\eta, \tag{25}$$

where the subscript sm means that the derivatives are evaluated at the smooth distributions, that is,

$$(q'_\theta)_{sm} = q'_\theta(\theta_{sm}(x), \eta_{sm}(x)),$$
$$(q'_\eta)_{sm} = q'_\eta(\theta_{sm}(x), \eta_{sm}(x)). \tag{26}$$

As follows from the general qualitative theory [5,6], for all types of instabilities the condition $\alpha\omega \ll 1$ is satisfied. For this reason we may neglect the left-hand side in Eq. (25), and obtain that

$$\delta\theta_{sm} = -\frac{(q'_\eta)_{sm}}{(q'_\theta)_{sm}} \delta\eta. \tag{27}$$

Let us substitute Eq. (24) into Eqs. (16) and (17). Using Eqs. (21) and (27), we can rewrite Eq. (16) around $\mathscr{S}$ and Eq. (17) in the whole space as

$$(i\alpha\omega + \epsilon^2 \hat{S}_i + \hat{H}_\theta^1)\delta\theta_{sh} = -q'_\eta \delta\eta, \tag{28}$$

$$\left[i\omega - \Delta + (Q'_\eta)_{sm} - \frac{(q'_\eta)_{sm}(Q'_\theta)_{sm}}{(q'_\theta)_{sm}} \right] \delta\eta = -Q'_\theta \delta\theta_{sh}. \tag{29}$$

Note that we neglected the term $\hat{H}_\theta^0 \delta\theta_{sm}$ in the left-hand side of Eq. (28) since it does not contribute in the first order of the perturbation theory. Also note that in writing Eq. (29) we replaced the true distributions $\theta(x)$ by the smooth distributions $\theta_{sm}(x)$. It is easy to see that this replacement gives negligible difference in $\delta\eta$.

Let us solve Eq. (29) for $\delta\eta$ by means of the Green's function

$$\delta\eta(x) = -\int Q'_\theta(x') G(x,x') \delta\theta_{sh}(x') dx', \tag{30}$$

where $G(x,x')$ satisfies

$$[i\omega - \Delta + C + V(x)] G(x,x') = \delta(x - x'), \tag{31}$$

where

$$C = Q'_\eta(\theta_h, \eta_h) - \frac{q'_\eta(\theta_h, \eta_h) Q'_\theta(\theta_h, \eta_h)}{q'_\theta(\theta_h, \eta_h)}, \tag{32}$$

and

$$V(x) = \left[(Q'_\eta)_{sm} - \frac{(q'_\eta)_{sm}(Q'_\theta)_{sm}}{(q'_\theta)_{sm}} \right] - C. \tag{33}$$

In view of Eq. (30), the right-hand side of Eq. (28) can be regarded as an operator $\hat{R}$ acting on $\delta\theta_{sh}$:

$$\hat{R}[\delta\theta_{sh}] = q'_\eta(x) \int Q'_\theta(x') G(x,x') \delta\theta_{sh}(x') d^d x'. \tag{34}$$

Since the sharp fluctuation $\delta\theta_{sh}$ is the linear combination of the functions $\delta\theta_{il}$ defined in Eq. (22), the integral in Eq. (34) can be easily calculated. Taking into account that $d\theta_{sh}/d\rho_i$ are close to δ functions [5,6], we may write

$$\hat{R}[\delta\theta_{il}] = [Q(\theta_{s3}, \eta_s) - Q(\theta_{s1}, \eta_s)] q'_\eta(x) \int_{\mathscr{S}_i} G(x, \xi_i) \phi_l^i(\xi_i) d^{d-1}\xi_i, \tag{35}$$

where ξ_i denotes the point on $\mathscr{S}_i$ and the integration is over $\mathscr{S}_i$. The matrix elements of $\hat{R}$ can be calculated analogously. The result of the calculation is

$$\langle i'l'|\hat{R}|il\rangle = -B\int_{\mathscr{S}_{i'}}\int_{\mathscr{S}_i} G(\xi_{i'},\xi_i)\phi^*_{l'}(\xi_{i'}) \times \phi_l(\xi_i)d^{d-1}\xi_{i'}d^{d-1}\xi_i, \quad (36)$$

where

$$B = -[Q(\theta_{s3},\eta_s) - Q(\theta_{s1},\eta_s)]\int_{\theta_{s1}}^{\theta_{s3}} q'_\eta(\theta,\eta_s)d\theta \quad (37)$$

is a constant depending on A only. Note that in accordance with Eq. (5) the value of B is positive.

When the distance between the different $\mathscr{S}_i$ is much greater than ϵ, the overlap between the different $\delta\theta_{il}$ is negligible, so the operator $\hat{H}^1_\theta$ is diagonal in the i indices. Then, in the first order of the perturbation theory Eqs. (28), (34), and (36) reduce to

$$(i\alpha\omega + \epsilon^2\nu_{li})\delta_{ii'}\delta_{ll'} = \epsilon Z^{-1}[\langle i'l'|\hat{R}|il\rangle - \delta_{ii'}\langle il'|\hat{H}^1_\theta|il\rangle], \quad (38)$$

where

$$Z = \epsilon\int_{-\infty}^{+\infty}\left(\frac{d\theta_{sh}}{d\rho}\right)^2 d\rho. \quad (39)$$

Note that the value of Z is of order 1 since the characteristic length of the activator variation is ϵ.

Equation (38) is the principal equation which determines the stability of an arbitrary d-dimensional pattern in N systems. This equation was derived with an accuracy to $\epsilon \ll 1$ and $\epsilon K_{max} \ll 1$, where K_{max} is the maximum curvature of a given pattern.

If a pattern possesses certain symmetries, the operators $\hat{R}$ and $\hat{H}^1_\theta$ are diagonal in the l indices. In this case the operators in Eq. (38) can be easily diagonalized (see the following sections). The main problem is to find the Green's function $G(x,x')$. Once this is done, we can obtain the "dispersion relation," which relates ω to the values of A, ϵ, and α for different types of fluctuations.

IV. STATIC ONE-DIMENSIONAL AUTOSOLITON IN TWO AND THREE DIMENSIONS

Let us apply the procedure developed in the previous section to the simplest pattern — a static one-dimensional autosoliton in two or three dimensions (Fig. 3). Since the AS is localized, the distributions of the activator and the inhibitor on its periphery go to the stable homogeneous state $\theta = \theta_h$ and $\eta = \eta_h$, where θ_h and η_h are determined by Eq. (6). In this case, according to Eqs. (5) and (32), the value of $C > 0$ since for $\theta_h < \theta_0$ the value of $q'_\theta(\theta_h,\eta_h) > 0$ [5,6].

For a one-dimensional (1D) AS the manifold $\mathscr{S}$ consists of two parallel planes where the AS walls are localized. We can choose the coordinate directions in such a way that these planes are perpendicular to the z axis. Then the solution for the AS will depend only on z.

Since the considered static 1D AS is symmetric with respect to its center [5,6], we can assume that the positions of its left and its right walls are $z_1 = -\mathscr{L}_s/2$ and $z_2 = \mathscr{L}_s/2$, respectively, where $\mathscr{L}_s$ is the distance between the walls.

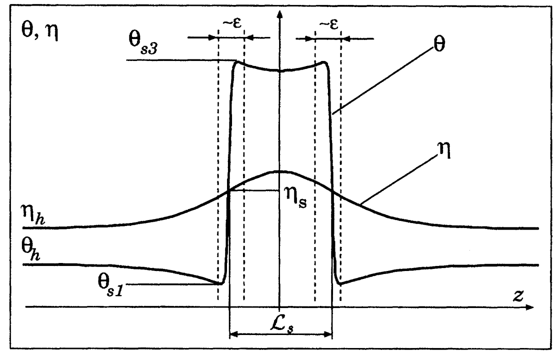

FIG. 3. Distributions of θ and η in the form of a one-dimensional AS. On the AS periphery θ goes to θ_h and η goes to η_h. Dashed lines indicate the regions where the AS walls are localized.

Note that the value of $\mathscr{L}_s$ itself can be used as a bifurcation parameter instead of A, since there is a one-to-one correspondence between them in the whole region of AS existence [5,6]. For this reason, here and further we will use the AS width $\mathscr{L}_s$ as the bifurcation parameter instead of A.

Since the $\mathscr{S}_{1,2}$ are flat, the curvilinear coordinates $\rho_{1,2}$ coincide with $z - z_1$ and $-(z - z_2)$, respectively (the signs are consistent with our definition of ρ_i given in Sec. II), and the coordinates ξ_i coincide with the rest of the coordinates of space. Of course, $\hat{K} \equiv 0$.

According to our procedure, let us first look at the operator $\hat{S}_i$. Here $\hat{S}_{1,2} = -\Delta_{\xi_{1,2}}$ is the Laplacian, acting on the local coordinates on $\mathscr{S}_{1,2}$. The eigenfunctions of this operator are just plane waves with the wave vector k along $\mathscr{S}_{1,2}$, whose eigenvalues are $\nu_k = k^2$.

If we substitute the eigenfunctions of $\hat{S}_{1,2}$ into Eq. (36) and use the fact that the system has translational invariance in the ξ directions, by integrating over the ξ_i's we will get

$$\langle 1,k|\hat{R}|1,k'\rangle = -BG_k\left(\frac{\mathscr{L}_s}{2},\frac{\mathscr{L}_s}{2}\right)\delta(k-k'), \quad (40)$$

$$\langle 1,k|\hat{R}|2,k'\rangle = -BG_k\left(-\frac{\mathscr{L}_s}{2},\frac{\mathscr{L}_s}{2}\right)\delta(k-k'), \quad (41)$$

where $G_k(z,z')$ is the Fourier transform of $G(x,x')$ in ξ. Because the AS is symmetric with respect to its center, the values of the matrix elements satisfy $\langle 1,k|\hat{R}|1,k\rangle = \langle 2,k|\hat{R}|2,k\rangle$ and $\langle 1,k|\hat{R}|2,k\rangle = \langle 2,k|\hat{R}|1,k\rangle$. Thus the matrix elements of the operator $\hat{R}$ in this case can be expressed in terms of the values of the Fourier-transformed Green's function at the particular points of the z axis.

Let us now turn to the operator $\hat{H}^1_\theta$. As was shown in the general qualitative theory [5,6], the functions $d\theta_{sh}/d\rho_{1,2}$ decay exponentially at distances much larger than ϵ. Because of this, for $\mathscr{L}_s \gg \epsilon\ln(\epsilon^{-1})$ the overlap of these functions can be neglected. Following the notation of Refs. [5,6], we write the diagonal elements of $\hat{H}^1_\theta$ as

$$\langle 1,k|\hat{H}_\theta^1|1,k'\rangle=\langle 2,k|\hat{H}_\theta^1|2,k'\rangle=\epsilon^{-1}Z\lambda_0\delta(k-k'), \tag{42}$$

where λ_0 is of order ϵ. The operator in the right-hand side of Eq. (38) can be trivially diagonalized. Since the AS possesses central symmetry, the eigenstates of this operator are the symmetric and the antisymmetric combinations of $\delta\theta_{1k}$ and $\delta\theta_{2k}$. These states correspond to the symmetric and the antisymmetric deformations of the AS walls. As a result, introducing the functions

$$R_0(k,\omega)=G_k\left(\frac{\mathscr{L}_s}{2},\frac{\mathscr{L}_s}{2}\right)+G_k\left(\frac{\mathscr{L}_s}{2},-\frac{\mathscr{L}_s}{2}\right), \tag{43}$$

$$R_1(k,\omega)=G_k\left(\frac{\mathscr{L}_s}{2},\frac{\mathscr{L}_s}{2}\right)-G_k\left(\frac{\mathscr{L}_s}{2},-\frac{\mathscr{L}_s}{2}\right), \tag{44}$$

from Eq. (38) we obtain the dispersion relation

$$i\alpha\omega+\epsilon^2k^2+\lambda_0=-\epsilon BZ^{-1}R_{0,1}(k,\omega). \tag{45}$$

Here the subscript "0" corresponds to the symmetric and the subscript "1" corresponds to the antisymmetric mode of fluctuations. The value of λ_0 can be calculated indirectly. Since the AS possesses translational invariance in the z direction, Eq. (45) should be identically satisfied for $k=0$ and $\omega=0$ for the antisymmetric fluctuation. This immediately means that

$$\lambda_0=-\epsilon BZ^{-1}R_1(0,0). \tag{46}$$

Equations (43) and (44) define the functions $R_0(k,\omega)$ and $R_1(k,\omega)$, which describe the inhibitor reaction on the dangerous fluctuations of the activator. In general the potential $V(z)$ in Eq. (31) which determines the Green's function is some nontrivial function of z, so $R_{0,1}(k,\omega)$ are some complicated functions of $\mathscr{L}_s$. In the case of the piecewise-linear model one can calculate the values of $R_0(k,\omega)$ and $R_1(k,\omega)$ explicitly (see Appendix B). One can see that the dispersion relations for the fluctuations obtained in this case are identical to those found earlier by Ohta, Mimura, and Kobayashi in Ref. [47], who used a different approach.

To calculate the critical values of A and the parameters of the critical fluctuations in general, we need to know the detailed form of the Green's function. According to Eq. (31), the Fourier-transformed Green's function $G_k(z,z')$ is governed by

$$\left[-\frac{\partial^2}{\partial z^2}+i\omega+k^2+C+V(z)\right]G_k(z,z')=\delta(z-z'). \tag{47}$$

As was said earlier, the potential in the operator in the left-hand side of Eq. (47) is some complicated function of z. However, the problem is greatly simplified for finding instabilities, since, as was shown in the general qualitative theory [5,6], most of the instabilities occur when $\mathscr{L}_s \ll 1$ (see also the results below). This allows one to use the value of $\mathscr{L}_s$ as a small parameter and expand the functions $R_{0,1}(k,\omega)$ in terms of it.

Considering all this, we are now able to construct the perturbation expansion for the Green's function, considering the potential $V(z)$ in Eq. (47) as a perturbation. The unperturbed Green's function satisfies

$$\left[-\frac{\partial^2}{\partial z^2}+k^2+i\omega+C\right]G_k^{(0)}(z,z')=\delta(z-z'). \tag{48}$$

The solution of Eq. (48) is well known:

$$G_k^{(0)}(z,z')=\frac{\exp\{-\sqrt{C+k^2+i\omega}|z-z'|\}}{2\sqrt{C+k^2+i\omega}}. \tag{49}$$

To calculate the corrections to the Green's function, we use the formula

$$G_k^{(n)}(z,z')=-\int_{-\infty}^{+\infty}V(z'')G_k^{(0)}(z,z'')G_k^{(n-1)}(z'',z')dz'', \tag{50}$$

where $G_k^{(n)}(z,z')$ is the nth correction.

Let us denote the contributions from $G_k^{(n)}$ to $R_{0,1}(k,\omega)$ as $R_{0,1}^{(n)}(k,\omega)$, respectively. Since for small values of $\mathscr{L}_s$ the coefficients B, C, and Z only weakly depend on A, we may replace them by their values at $A=A_b$, where the AS size becomes formally zero when $\epsilon\to 0$ [5,6]. Then the functions $R_0^{(0)}(k,\omega)$ and $R_1^{(0)}(k,\omega)$ can be written as

$$R_0^{(0)}(k,\omega)=\frac{1}{2\sqrt{C+k^2+i\omega}}\{1+\exp(-\mathscr{L}_s\sqrt{C+k^2+i\omega})\}, \tag{51}$$

$$R_1^{(0)}(k,\omega)=\frac{\mathscr{L}_s}{2}-\frac{\mathscr{L}_s^2\sqrt{C+k^2+i\omega}}{4}+O(\mathscr{L}_s^3). \tag{52}$$

Note that $R_0^{(n)}=O(\mathscr{L}_s^n)$, and because of the central symmetry of the potential $R_1^{(n)}=O(\mathscr{L}_s^{2n+1})$.

Substituting these values of $R_{0,1}(k,\omega)$ into Eq. (46), we will obtain that the leading term of λ_0 is

$$\lambda_0=-\frac{\epsilon Z^{-1}B\mathscr{L}_s}{2}. \tag{53}$$

Having calculated the value of λ_0, we may write the dispersion relation for the symmetric fluctuations

$$\begin{aligned} i\alpha\omega+\epsilon^2k^2=\epsilon BZ^{-1}\Bigg\{&\frac{\mathscr{L}_s}{2}-\frac{1}{2\sqrt{C+k^2+i\omega}} \\ &\times\{1+\exp(-\mathscr{L}_s\sqrt{C+k^2+i\omega})\} \\ &-R_0^{(1)}(k,\omega)\Bigg\}+O(\mathscr{L}_s^2). \end{aligned} \tag{54}$$

As was shown in the general qualitative theory [5,6,39,40], when α is big enough, for $\mathscr{L}_s>\mathscr{L}_{c1}$ the one-dimensional AS becomes unstable with respect to the fluctuation with $Re\,\omega=0$ and $k=k_c\gg 1$, corresponding to the corrugation of the AS walls. Note that, according to Eq. (50), for $k_c\gg 1$ the value of $R_0^{(1)}(k,\omega)$ which is of order $\mathscr{L}_s$, contains a small factor $\propto k^{-2}$, so it can be neglected. Let us calculate the values of $\mathscr{L}_{c1}$ and k_c. Putting $\omega=0$ and neglecting C in comparison with k^2 in Eq. (54), we will get a transcendental equation, which can be solved for small values of $\mathcal{L}_s$. The result is

$$k_c=0.71\left(\frac{\epsilon Z}{B}\right)^{-1/3}, \quad \mathcal{L}_{c1}=2.64\left(\frac{\epsilon Z}{B}\right)^{1/3}. \tag{55}$$

Note that the dependences of $\mathcal{L}_{c1}$ and k_c on ϵ coincide with those obtained in the qualitative theory for $\mathcal{L}_s \ll 1$ [5,6].

In the case $\alpha \ll \epsilon$ there is an instability for $\mathcal{L}_s > \mathcal{L}_\omega$ with respect to the fluctuation describing the pulsations of the AS with the frequency $\omega=\omega_c \gg 1$ and $k=0$ [5,6,41,42]. As before, the term $R_0^{(1)}(k,\omega)$ contains the small factor ω^{-1} and can be neglected. Solving the transcendental equation obtained in this case, we have

$$\omega_c=0.73\left(\frac{\alpha Z}{\epsilon B}\right)^{-2/3}, \quad \mathcal{L}_\omega=0.96\left(\frac{\alpha Z}{\epsilon B}\right)^{1/3}. \tag{56}$$

Again, the calculated dependences of $\mathcal{L}_\omega$ and ω_c on the ratio α/ϵ coincide with those obtained in the qualitative theory [5,6], and in the analytic studies of the piecewise-linear model [25]. Also, comparing $\mathcal{L}_{c1}$ and $\mathcal{L}_\omega$, one can see that the pulsating instability occurs before the corrugation instability if $\alpha < 21\epsilon^2$.

When the size of the AS is greater than the critical size determined by Eq. (55), the increment of the growing fluctuations may be very small. Indeed, according to Eq. (54), for $\mathcal{L}_s \gg \mathcal{L}_{c1}$ we obtain that the increment of the most dangerous fluctuations is $\gamma \simeq \epsilon B \mathcal{L}_s/(2\alpha Z)$.

Now let us turn to the antisymmetric fluctuations. According to Eq. (52), the first term in $R_1(k,\omega)$ which depends on ω and k is of order $\mathcal{L}_s^2$. As was mentioned earlier, the first correction $R_1^{(1)}(k,\omega)$ is of the order $\mathcal{L}_s^3$, so it can be neglected. Then Eq. (45) for the antisymmetric fluctuations can be written as

$$i\alpha\omega+\epsilon^2k^2=\frac{\epsilon B Z^{-1}\mathcal{L}_s^2}{4}\{\sqrt{C+k^2+i\omega}-\sqrt{C}\}+O(\mathcal{L}_s^3). \tag{57}$$

According to the general qualitative theory [5,6], there are two types of antisymmetric instabilities: wriggling of the AS walls, and formation of a traveling AS. The first instability is realized when $\mathrm{Re}\omega=0$, $k\to 0$, and $\mathcal{L}_s > \mathcal{L}_{c2}$. The second is realized when $\alpha \ll 1$, $k=0$, and $\mathcal{L}_s > \mathcal{L}_T$. We may expand the the right-hand side of Eq. (57) in powers of k and ω and obtain the expression

$$i\alpha\omega+\epsilon^2k^2=\epsilon b \mathcal{L}_s^2(k^2+i\omega), \tag{58}$$

where we retained only the first nonvanishing terms. The constant b is given by

$$b=\frac{B}{8ZC^{1/2}}. \tag{59}$$

One can see from Eq. (58) that the instability $\mathrm{Im}\omega<0$ occurs at $k=0$ for $\mathcal{L}_s > \mathcal{L}_T$, where

$$\mathcal{L}_T=\left(\frac{\alpha}{b\epsilon}\right)^{1/2}, \tag{60}$$

or for $\mathrm{Re}\omega=0$ and $\mathcal{L}_s > \mathcal{L}_{c2}$, where

$$\mathcal{L}_{c2}=\left(\frac{\epsilon}{b}\right)^{1/2}. \tag{61}$$

Comparing these two formulas, one can see that a traveling AS forms before the walls of the static AS become unstable with respect to wriggling, when $\alpha < \epsilon^2$. This fact is also an obvious consequence of the additional symmetry present in Eqs. (3) and (4) at $\alpha=\epsilon^2$.

Although for $\mathcal{L}_s > \mathcal{L}_{c2}$ the AS is unstable, the increment of the most dangerous fluctuations may be extremely small. Indeed, according to Eq. (57), for $\mathcal{L}_{c2} \ll \mathcal{L}_T$ and $\mathcal{L}_{c2} \ll \mathcal{L}_s \ll \mathcal{L}_{c1}$ we have

$$\gamma_{max}=\alpha^{-1}\left(\frac{\mathcal{L}_s^2 B}{8Z}\right)^2 \ll 1 \quad \text{for} \quad k=k_{max}=\frac{\mathcal{L}_s^2 B}{8\epsilon Z} \gg 1. \tag{62}$$

Comparison of the expressions in Eqs. (55) and (61) for the critical values of $\mathcal{L}_s$ for symmetric and antisymmetric fluctuations shows that for $\epsilon \ll 1$ the wriggling instability always emerges before the corrugation instability. Likewise, according to Eqs. (56) and (60), for sufficiently small ratios α/ϵ the traveling AS forms before the pulsating one. Notice that these general conclusions are in agreement with the analytic investigation of AS in the piecewise-linear model [47,25].

When the size of the AS becomes comparable with ϵ, the overlap between the eigenfunctions of the operator $\hat{H}_\theta^0$ becomes significant, which leads to instability of the AS. As was shown in the general qualitative theory [5,6], the instability of an AS with $\mathcal{L}_s \sim \epsilon$ occurs with respect to symmetric fluctuations. Since the asymptotic behavior of the sharp solutions is exponential, an extra piece in Eq. (54) from the operator $\hat{H}_\theta^0$ will have the form $a\exp(-\mathcal{L}_s/\tilde{l})$, where $\tilde{l}=\epsilon q_\theta'(\theta_{s3},\eta_s)^{-1/2}$, and a is some constant of order 1 [5,6,37–40]. Then we obtain that for $\alpha \gg \epsilon$ the instability occurs with respect to the fluctuations with $\mathrm{Re}\omega=0$ and

$$k=k_c=2^{-1/3}\left(\frac{\epsilon Z}{B}\right)^{-1/3}, \quad \mathcal{L}_s < \mathcal{L}_{cb}=\frac{4\tilde{l}}{3}\ln\epsilon^{-1}, \tag{63}$$

whereas for $\alpha \ll \epsilon$ the instability is realized with respect to the fluctuations with $k=0$ and

$$\omega=\omega_c=2^{-1/3}\left(\frac{\alpha Z}{\epsilon B}\right)^{-2/3}, \quad \mathcal{L}_s < \mathcal{L}_{b\omega}=-\frac{\tilde{l}}{3}\ln\alpha\epsilon^2. \tag{64}$$

Equations (63) and (64) were calculated with logarithmic accuracy and coincide with those obtained in the qualitative theory [5,6].

In one-dimensional systems with $\alpha \gg \epsilon$ the value of $\mathcal{L}_b$ at which the AS disappears will be slightly different from $\mathcal{L}_{cb}$, since the only possible value for k there is zero. Putting $k=0$ in the dispersion relation we obtain that $\mathcal{L}_b=\tilde{l}\ln\epsilon^{-1}$ [5,6].

Up to now we have considered an AS whose width $\mathcal{L}_s \ll 1$. This is justified in two- or three-dimensional systems since, according to Eqs. (55) and (61), the AS becomes unstable when $\mathcal{L}_s \ll 1$. In one dimension, however, an AS remains stable up to the values of $\mathscr{L}_s \sim 1$, if $\alpha \gg \epsilon$. In this situation, according to the general qualitative theory [5,6], there is another effect causing the instability; namely, when $\mathscr{L}_s$ is close to some $\mathscr{L}_d \sim 1$ at which the activator in the AS center reaches the value of θ_0', such that $q'_\theta(\theta_0', \eta_0') = 0$, the solution in the form of an AS disappears. As a result, a local breakdown occurs at the AS center, causing the AS to split, so that eventually the whole system becomes filled with a complex pattern [5,6]. This can also be seen from Eq. (45), if we take into account that at the point $z=0$ where $\theta = \theta_0'$ the potential $V(z)$ in Eq. (47) becomes singular. In the case $\alpha \lesssim \epsilon$ the values of $\mathscr{L}_\omega$ and $\mathscr{L}_T$ corresponding to the instabilities leading to formation of pulsating and traveling AS's, respectively, may be less than $\mathscr{L}_d$, so the AS cannot reach the point where local breakdown occurs. There is also a possibility that the point $A = A_d$ where $\mathscr{L}_s = \mathscr{L}_d$ is preceded by the point $A = A_c$ where the homogeneous state of the system becomes unstable [5,6]. In this case the periphery of the AS becomes unstable. This can also be seen from Eq. (45) if we take into account that, according to Eq. (47), for $A > A_0$ the value of C becomes negative and therefore the tails of the Green's function become oscillatory. When α gets larger, the right-hand side of Eq. (45) always remains of order 1, so at some critical values of $\alpha \gtrsim \epsilon$ the instability leading to the formation of traveling and pulsating AS's disappears.

V. HIGHER-DIMENSIONAL RADIALLY SYMMETRIC AUTOSOLITONS

Let us now turn to higher-dimensional AS's. First we consider spherically symmetric AS's in three dimensions. In this case there is only one manifold $\mathscr{S}$ which is simply a sphere of radius $\mathscr{R}_s$. As a set of orthogonal curvilinear coordinates we choose the usual spherical coordinates ρ, ϑ, and φ, except ρ will be measured from the surface of the sphere rather than from the origin, in order to be consistent with our initial definition.

The operator $\hat{S}$ in the considered case is

$$\hat{S} = -\mathscr{R}_s^{-2}\left\{ \frac{1}{\sin\vartheta}\frac{\partial}{\partial\vartheta}\sin\vartheta\frac{\partial}{\partial\vartheta} + \frac{1}{\sin^2\vartheta}\frac{\partial^2}{\partial\varphi^2} \right\}. \quad (65)$$

The eigenfunctions of the operator $\hat{S}$ are just spherical harmonics $\phi_{lm} = Y_{lm}(\vartheta,\varphi)/\mathscr{R}_s$ with the eigenvalues $\nu_l = l(l+1)/\mathscr{R}_s^2$. The factor $1/\mathscr{R}_s$ in the eigenfunction ensures the proper normalization.

Now let us calculate the matrix elements. First of all, since the system possesses spherical symmetry, the only nonvanishing matrix elements of the operator $\hat{H}_\theta^1$ are the diagonal elements, which are all equal to each other. Due to the same symmetry, the operator $\hat{R}$ is also diagonal, and its diagonal elements are independent of m. According to Eq. (36), the diagonal matrix elements of $\hat{R}$ are

$$\langle lm|\hat{R}|lm\rangle \equiv -BR_l(\omega), \quad (66)$$

where

$$R_l(\omega) = \int\int G(\mathscr{R}_s,\vartheta,\varphi;\mathscr{R}_s,\vartheta',\varphi') \times Y_{lm}^*(\vartheta,\varphi)Y_{lm}(\vartheta',\varphi')\mathscr{R}_s^2 do' do, \quad (67)$$

do is the element of solid angle, and the Green's function is written in terms of the spherical coordinates.

To calculate $R_l(\omega)$ let us note that $R_l(\omega) = \mathscr{R}_s^2 G_l(\mathscr{R}_s, \mathscr{R}_s)$, where

$$G_l(r,r') = \int\int G(r,\vartheta,\varphi;r',\vartheta',\varphi') \times Y_{lm}^*(\vartheta,\varphi)Y_{lm}(\vartheta',\varphi') do do' \quad (68)$$

is the Green's function satisfying

$$r^2\left[-\frac{d^2}{dr^2} - \frac{2}{r}\frac{d}{dr} + \frac{l(l+1)}{r^2} + C + i\omega + V(r) \right] G_l(r,r') = \delta(r-r'). \quad (69)$$

Equation (69) follows from Eq. (31) if we first rewrite it in the spherical coordinates r, ϑ, and φ, multiply both sides by $Y_{lm}^*(\vartheta,\varphi)Y_{lm}(\vartheta',\varphi')$, and then integrate over do and do', taking into account the Hermiticity of the angular part of the Laplacian and the orthonormality of the spherical harmonics.

Using the above mentioned properties, we can now write Eq. (38) for the spherically symmetric AS of radius $\mathscr{R}_s$ in the form

$$i\alpha\omega + \frac{\epsilon^2 l(l+1)}{\mathscr{R}_s^2} + \lambda_0 = -\epsilon B Z^{-1} R_l(\omega), \quad (70)$$

where λ_0 is the contribution from $\hat{H}_\theta^1$. Equation (70) is the dispersion relation for a fluctuation characterized by the number l.

It can be shown by direct calculation that to first order in $\epsilon/\mathscr{R}_s$ the value of λ_0 is zero. This means that in order to calculate λ_0 from its definition one should know the distributions $\theta(x)$ and $\eta(x)$ for the AS with greater accuracy than that of Eq. (14). Also, the second order of the perturbation theory has to be taken into account. However, these difficulties can be avoided, if we use the fact that the system possesses translational invariance. Indeed, Eq. (67) should be identically satisfied for $l=1$ and $\omega=0$, so we immediately obtain that

$$\lambda_0 = -\frac{2\epsilon^2}{\mathscr{R}_s^2} - \epsilon B Z^{-1} R_1(0). \quad (71)$$

As we will show below, the instability of a spherically symmetric AS occurs at $\mathscr{R}_s \ll 1$. To find the function $R_l(\omega)$ let us use the idea of the previous section and seek for the Green's function of Eq. (69), treating the potential $V(r)$ as a perturbation. The zeroth-order Green's function is well known:

$$G_l^{(0)}(r',r'')=\begin{cases}\dfrac{I_{l+1/2}(\kappa r')K_{l+1/2}(\kappa r'')}{\sqrt{r'r''}}, & r'<r''\\[2ex] \dfrac{I_{l+1/2}(\kappa r'')K_{l+1/2}(\kappa r')}{\sqrt{r'r''}}, & r'>r'',\end{cases} \tag{72}$$

where $\kappa=\sqrt{C+i\omega}$; $K_{l+1/2}(z)$ and $I_{l+1/2}(z)$ are the modified Bessel functions. The corrections to $G_l^{(0)}(r',r'')$ are then given by

$$G_l^{(n)}(r',r'')=-\int_0^\infty V(r)G_l^{(0)}(r',r)G_l^{(n-1)}(r,r'')r^2dr. \tag{73}$$

As a result, from Eq. (72) we obtain that

$$R_l^{(0)}(\omega)=\mathscr{R}_s I_{l+1/2}(\kappa\mathscr{R}_s)K_{l+1/2}(\kappa\mathscr{R}_s), \tag{74}$$

and, as can be seen from Eq. (73), $R_l^{(n)}(\omega)\equiv \mathscr{R}_s^2 G_l^{(n)}(\mathscr{R}_s,\mathscr{R}_s)=O(\mathscr{R}_s^{2n+1})$.

As before, we will use the values of B, C, and Z evaluated at the point $A=A_b$ where $\mathscr{R}_s\to 0$ as $\epsilon\to 0$. Expanding the Bessel functions at $\mathscr{R}_s\ll 1$, we may write Eq. (70) as

$$i\alpha\omega+\epsilon^2\mathscr{R}_s^{-2}(l+2)(l-1)=\epsilon BZ^{-1}\mathscr{R}_s\left(\frac{1}{3}-\frac{1}{2l+1}\right). \tag{75}$$

Equation (75) describes the fluctuations in the case $|\omega|\mathscr{R}_s^2\ll 1$. This is satisfied for the thresholds of the aperiodic instabilities. Simple calculation shows that an AS becomes unstable with respect to the aperiodic $l=0$ mode when

$$\mathscr{R}_s<\mathscr{R}_b\equiv\left(\frac{3\epsilon Z}{B}\right)^{1/3}. \tag{76}$$

For $\mathscr{R}_s>\mathscr{R}_b$ it becomes unstable with respect to the aperiodic fluctuations with $l>1$ at

$$\mathscr{R}_s>\mathscr{R}_{cl}\equiv\left(\frac{3(l+2)(2l+1)\epsilon Z}{2B}\right)^{1/3}. \tag{77}$$

One can see from this equation that the first instability point corresponds to $l=2$:

$$\mathscr{R}_{c2}=\left(\frac{30\epsilon Z}{B}\right)^{1/3}. \tag{78}$$

Thus a spherically symmetric AS can be stable only if its radius satisfies $\mathscr{R}_b<\mathscr{R}_s<\mathscr{R}_{c2}$, where $\mathscr{R}_b$ and $\mathscr{R}_{c2}$ are given by Eqs. (76) and (78), respectively.

The $l=1$ mode corresponds to the translation of the AS as a whole. As in the case of the one-dimensional AS, for some value of $\alpha\ll 1$ the spherically symmetric AS becomes unstable with respect to the fluctuation, leading to the formation of a traveling AS. The instability with respect to the $l=1$ mode occurs for some $\mathscr{R}_s>\mathscr{R}_T$ when $\omega\to 0$. Expanding the Bessel functions in the right-hand side of Eq. (70) for small $\mathscr{R}_s$ and ω, we get

$$i\alpha\omega=\epsilon BZ^{-1}\mathscr{R}_s^3\left\{i\omega\frac{2}{15}-(i\omega)^2\frac{\mathscr{R}_s}{24\sqrt{C}}\right\} + \text{higher-order terms}. \tag{79}$$

According to this equation, the static AS transforms into a traveling one when $\mathscr{R}_s>\mathscr{R}_T$, where

$$\mathscr{R}_T=\left(\frac{15\alpha Z}{2\epsilon B}\right)^{1/3}. \tag{80}$$

Now let us study the instabilities with $\mathrm{Re}\,\omega\neq 0$. Consider an AS stable when $\alpha\gg 1$; its radius is therefore of order $\epsilon^{1/3}$. As follows from Eq. (70), in order for an instability to occur, the frequency $\mathrm{Re}\,\omega$ at the threshold of the instability should be big enough so that the argument of the Bessel functions in Eq. (74) is of order 1. This means that $\omega\sim\mathscr{R}_s^{-2}\sim\epsilon^{-2/3}$ and therefore the critical values of α are of order ϵ^2. Indeed, let us introduce the new variables

$$\bar{\alpha}=\alpha/\epsilon^2,\quad \bar{\omega}=\omega\left(\frac{\epsilon Z}{B}\right)^{2/3},\quad p=\frac{B\mathscr{R}_s^3}{\epsilon Z}. \tag{81}$$

Substituting these variables into Eqs. (70), (71), and (74), after some algebra we obtain the following transcendental equation which has explicit dependence on $\bar{\alpha}$, p, and $\bar{\omega}$ only:

$$i\bar{\alpha}\bar{\omega}+(l+2)(l-1)p^{-2/3}=p^{1/3}\left(\frac{1}{3}-I_{l+1/2}\{p^{1/3}\bar{\omega}^{1/2}\sqrt{i}\}\times K_{l+1/2}\{p^{1/3}\bar{\omega}^{1/2}\sqrt{i}\}\right). \tag{82}$$

We solved Eq. (82) numerically for $l=0$ and 2 in the region of p where the AS is stable with respect to the aperiodic fluctuations. The resulting stability diagram is shown in Fig. 4. The rescaled critical frequency as a function of $\bar{\alpha}$ for $l=0$ is also presented in Fig. 5. From Fig. 4 it is clear that when α gets smaller, the AS always loses stability with respect to the $l=0$ pulsations first. The AS is always unstable if $\alpha<\alpha_c=4.4\epsilon^2$. For $\alpha>6.7\epsilon^2$ the AS destabilizes with respect to the aperiodic $l=2$ mode first, if its radius is increased. All other instabilities, including the one leading to the formation of a traveling AS, occur at smaller values of α and are, therefore, secondary.

Let us now consider the case of the radially symmetric AS in two and three dimensions. Because of the close analogy with the case of the spherically symmetric AS, we will only outline the derivations, focusing mainly on the obtained instability criteria.

If r, φ, and z are the cylindrical coordinates, then the coordinate $\rho=-(r-\mathscr{R}_s)$, and the operator $\hat{S}$ is

$$\hat{S}=-\frac{1}{\mathscr{R}_s^2}\frac{\partial^2}{\partial\varphi^2}-\frac{\partial^2}{\partial z^2}. \tag{83}$$

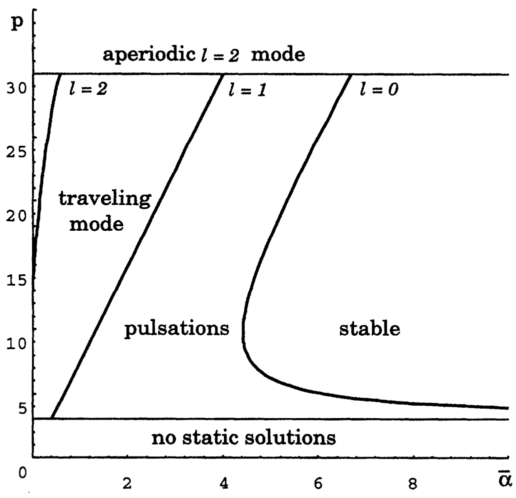

FIG. 4. Stability diagram for the spherically symmetric AS in the rescaled variables $\bar{\alpha}$ and p. The solid curves correspond to the thresholds of the instabilities for $l=0$, $l=1$, and $l=2$. The bottom and top horizontal lines correspond to the thresholds of the aperiodic instabilities for $l=0$ and $l=2$, respectively.

The eigenfunctions of this operator are $\phi_{km}=(4\pi^2\mathscr{R}_s)^{-1/2}e^{ikz+im\varphi}$ with the eigenvalues $\nu_{km}=k^2+m^2/\mathscr{R}_s^2$.

The nonvanishing matrix elements of the response operator in this case are

$$\langle mk|\hat{R}|mk'\rangle \equiv -BR_m(k,\omega)\delta(k-k') \tag{84}$$

where the function $R_m(k,\omega)$ can be expressed in terms of the Green's function given by

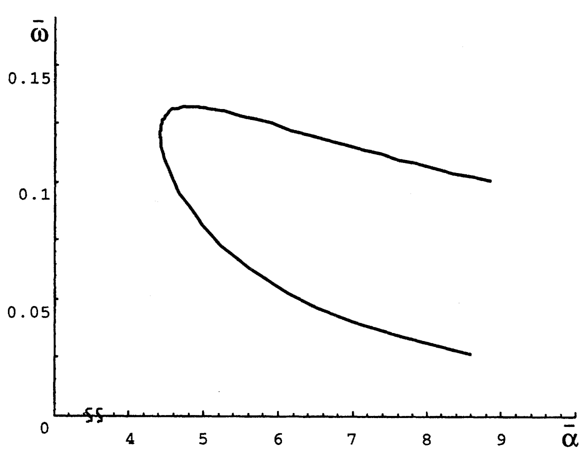

FIG. 5. The rescaled frequency $\bar{\omega}$ vs $\bar{\alpha}$ at the threshold of the $l=0$ instability of the spherically symmetric AS.

$$r\left[-\frac{d^2}{dr^2}-\frac{1}{r}\frac{d}{dr}+\frac{m^2}{r^2}+C+k^2+i\omega+V(r)\right]G_{km}(r,r') = \delta(r-r'), \tag{85}$$

as $R_m(k,\omega)=\mathscr{R}_s G_{km}(\mathscr{R}_s,\mathscr{R}_s)$.

The dispersion relation for the fluctuations with particular values of k and m, obtained from Eq. (38), is

$$i\alpha\omega+\epsilon^2k^2+\frac{\epsilon^2m^2}{\mathscr{R}_s^2}+\lambda_0=-\epsilon BZ^{-1}R_m(k,\omega). \tag{86}$$

Because of the translational invariance, this equation should be satisfied identically for $k=0$, $\omega=0$, and $m=1$. This gives us

$$\lambda_0=-\frac{\epsilon^2}{\mathscr{R}_s^2}-\epsilon BZ^{-1}R_1(0,0). \tag{87}$$

As before, for small values of $\mathscr{R}_s$ we will seek for the function $R_m(k,\omega)$ perturbatively. The zeroth-order Green's function here is

$$G_{km}^{(0)}(r',r'')=\begin{cases} I_m(\kappa r')K_m(\kappa r''), & r'<r'' \\ I_m(\kappa r'')I_m(\kappa r'), & r'>r'', \end{cases} \tag{88}$$

where $\kappa=\sqrt{C+k^2+i\omega}$, and $I_m(z)$ and $K_m(z)$ are the modified Bessel functions. The corrections to the Green's function are given by

$$G_{km}^{(n)}(r',r'')=-\int_0^\infty V(r)G_{km}^{(0)}(r',r)G_{km}^{(n-1)}(r,r'')rdr. \tag{89}$$

From this we find that

$$R_m^{(0)}(k,\omega)=\mathscr{R}_sI_m(\kappa\mathscr{R}_s)K_m(\kappa\mathscr{R}_s), \tag{90}$$

and that the corrections to $R_m(k,\omega)$ from $G_{km}^{(n)}$ are $R_m^{(n)}(k,\omega)\equiv\mathscr{R}_sG_{km}^{(n)}(\mathscr{R}_s,\mathscr{R}_s)=O(\mathscr{R}_s^{2n+1})$.

Let us study the stability of the radially symmetric AS in two dimensions first. Putting $k=0$ in Eq. (86) with $R_m(k,\omega)$ given by Eq. (90), we obtain that for $m>1$ and $\mathscr{R}_s\ll 1$ the aperiodic instability occurs at $\mathscr{R}_s>\mathscr{R}_{cm}$, where

$$\mathscr{R}_{cm}=\left(\frac{2\epsilon m(m+1)Z}{B}\right)^{1/3}. \tag{91}$$

According to this equation, when the value of $\mathscr{R}_s$ is increased the AS becomes unstable with respect to the fluctuation with $m=2$ when $\mathscr{R}_s>\mathscr{R}_{c2}$, where

$$\mathscr{R}_{c2}=\left(\frac{12\epsilon Z}{B}\right)^{1/3}. \tag{92}$$

When the value of $\mathscr{R}_s$ is decreased, at $\mathscr{R}_s<\mathscr{R}_b$ the radially symmetric AS in two dimensions becomes unstable with respect to aperiodic fluctuations with $m=0$. According to Eq. (86), for small values of $\mathscr{R}_s$ we have

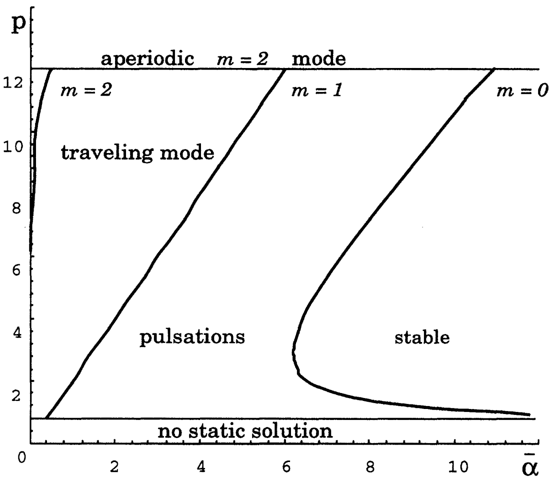

FIG. 6. Stability diagram for the radially symmetric AS in two dimensions in the rescaled variables $\bar{\alpha}$ and p. The solid curves correspond to the thresholds of the instabilities for $m=0$, $m=1$, and $m=2$. The top horizontal line corresponds to the thresholds of the aperiodic instability for $m=2$. The bottom horizontal line shows schematically the threshold of the aperiodic instability for $m=0$.

$$\mathcal{R}_b=\left(\frac{\epsilon Z}{Bb_1}\right)^{1/3}, \tag{93}$$

where $b_1=-\ln(1.47\mathcal{R}_s\sqrt{C})$. Since b_1 only weakly depends on $\mathcal{R}_s$, for $\mathcal{R}_s\sim 0.1$ we may put $b_1\simeq 2$, and for $\mathcal{R}_s\sim 0.01$ we may put $b_1\simeq 4$. Thus, a static radially symmetric AS in two dimensions can be stable only if $\mathcal{R}_b<\mathcal{R}_s<\mathcal{R}_{c2}$ with $\mathcal{R}_b$ and $\mathcal{R}_{c2}$ given in Eqs. (93) and (92), respectively.

When α gets sufficiently small, the radially symmetric AS in two dimensions typically destabilizes with respect to the $m=0$ pulsations first. If we introduce the variables of Eq. (81) into Eq. (86), for small $\mathcal{R}_s$ we will obtain an equation similar to Eq. (82), which depends on p, $\bar{\alpha}$, and $\bar{\omega}$ only. Solving this equation numerically for $m=0$ and 2 (the case $m=1$ will be discussed below) we obtain the stability diagram for the radially symmetric AS in two dimensions (Fig. 6). Figure 6 shows that when $\alpha<\alpha_c=6.1\epsilon^2$, the AS is unstable for all values of $\mathcal{R}_s$. When $\alpha>11\epsilon^2$ the AS first destabilizes with respect to aperiodic fluctuations with $m=2$. The dependence of the rescaled critical frequency $\bar{\omega}$ on $\bar{\alpha}$ for the $m=0$ pulsations is presented in Fig. 7. All instabilities with $m>2$ occur at smaller values of α, so they are secondary.

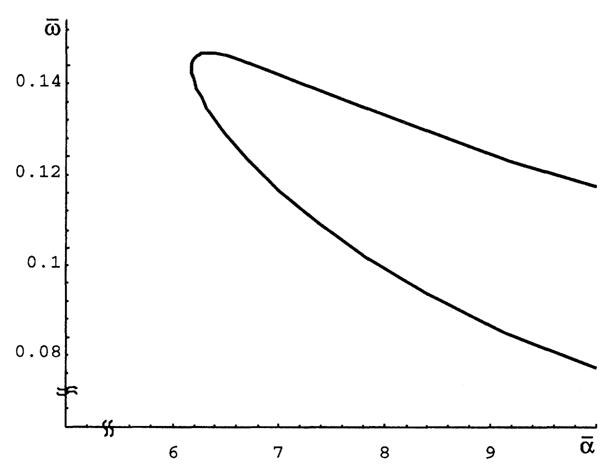

FIG. 7. The rescaled frequency $\bar{\omega}$ vs $\bar{\alpha}$ at the threshold of the $m=0$ instability of the radially symmetric AS in two dimensions.

Now let us turn to the radially symmetric AS in three dimensions. Solving the dispersion equation for small $\mathcal{R}_s$ for $m=0$, we obtain that the AS becomes unstable when $\mathcal{R}_s<\mathcal{R}_b$, where

$$\mathcal{R}_b=\left(\frac{2.25\epsilon Z}{B}\right)^{1/3}, \tag{94}$$

or when $\mathcal{R}_s>\mathcal{R}_c$, where

$$\mathcal{R}_c=\left(\frac{7.5\epsilon Z}{B}\right)^{1/3}, \tag{95}$$

when

$$k=k_c=\left(\frac{4.1\epsilon Z}{B}\right)^{-1/3}. \tag{96}$$

Comparing Eq. (95) with (92), one can see that in contrast to the radially symmetric AS in two dimensions, when $\mathcal{R}_s$ is increased, the radially symmetric AS in three dimensions destabilizes with respect to the $m=0$ mode first.

In the case $m=1$ an AS destabilizes with respect to fluctuations with small k at $\mathrm{Re}\,\omega=0$. Expanding the Bessel functions in the right-hand side of Eq. (86), we obtain the equation

$$i\alpha\omega+\epsilon^2k^2=\epsilon BZ^{-1}b_2\mathcal{R}_s^3(k^2+i\omega), \tag{97}$$

where $b_2=-\ln(1.14\mathcal{R}_s\sqrt{C})/4$. The value of b_2 weakly depends on $\mathcal{R}_s$, so for $\mathcal{R}_s\sim 0.1$ we may put $b_2\simeq 0.5$, and for $\mathcal{R}_s\sim 0.01$ we may put $b_2\simeq 1$. It can be seen from Eq. (97) that when $\alpha\gg\epsilon^2$ an AS becomes unstable when $\mathcal{R}_s>\mathcal{R}_{c1}$ at $k\to 0$, where

$$\mathcal{R}_{c1}=\left(\frac{\epsilon Z}{Bb_2}\right)^{1/3}. \tag{98}$$

Comparing Eq. (98) with Eq. (94) one can see that for $\epsilon\lesssim 10^{-3}$ we have $\mathcal{R}_{c1}<\mathcal{R}_b$, and, therefore, the cylindrically symmetric AS is always unstable. However, note that the increment of the fluctuations with $m=1$ and small k may be extremely small.

Similarly, as follows from Eq. (97), when $\alpha\sim\epsilon^2$ the AS becomes unstable for $\mathcal{R}_s>\mathcal{R}_T$ at $k=0$, where

$$\mathcal{R}_T=\left(\frac{\alpha Z}{\epsilon Bb_2}\right)^{1/3}, \tag{99}$$

and transforms into the traveling AS. As we already noticed, when $\mathcal{R}_s$ is not very small, the instability leading to the formation of a traveling AS occurs at smaller values of α than the instability with respect to the $m=0$ pulsations. However, when $\mathcal{R}_s\lesssim 0.01$ the coefficient b_2 in Eq. (99) becomes such that the line $p(\bar{\alpha})$ in Fig. 6 corresponding to the instability with $m=1$ crosses the curve corresponding to the threshold of the $m=0$ pulsations when $p<12$. In this situation a radially symmetric AS in two or three dimensions may become unstable and transform into the traveling AS before it destabilizes with respect to the $m=0$ pulsations. However, this is a rather unrealistic situation since in order for an AS to have a radius $\mathcal{R}_s \lesssim 0.01$ and be unstable with respect to the $m=1$ mode, one should have $\epsilon \lesssim 10^{-6}$ and $\alpha \lesssim 10^{-12}$.

VI. CONCLUSION

Thus, in the present paper we developed an asymptotic theory of the instabilities of arbitrary d-dimensional static patterns which may form in a wide class of reaction-diffusion systems of the activator-inhibitor type. This theory is based on the natural smallness of the parameter $\epsilon = l/L$, where l and L are the characteristic length scales of the activator θ and the inhibitor η, respectively. In fact, as was already mentioned, if the length scales l and L, as well as the normalization of θ and η, are chosen properly, the condition $\epsilon \ll 1$ is not only sufficient, but also necessary for the existence of AS and other large-amplitude patterns [5,6].

Within the presented theory we analyzed different types of spontaneous transformations of the simplest static patterns in two- and three-dimensional systems into more complex static, pulsating, and traveling patterns. We showed that the criteria corresponding to these transformations are universal in the sense that they are practically independent of the specific nonlinearities of the system and are determined only by the two parameters ϵ and α and three numerical constants B, C, and Z, which have all necessary information about the nonlinearities. If the length scales and the normalization of θ and η are chosen properly, the constants B, C, and Z are necessarily of order 1, and the constant B can in principle be small.

Let us summarize the results of our analysis.

According to the formulas of Sec. IV, when $\alpha \gg \epsilon$ a one-dimensional AS in two and three dimensions (stripe) is less stable than the AS in one dimension. As follows from Eqs. (55) and (61), for $\epsilon \ll 1$ we have $\mathcal{L}_{c2} < \mathcal{L}_{c1}$. This means that when the control parameter A is increased and the AS widens, the AS always destabilizes with respect to the wriggling of its walls first. It is natural to expect that for $\mathcal{L}_{c2} < \mathcal{L}_s < \mathcal{L}_{c1}$ a one-dimensional AS will deform into a wriggled stripe that fills the whole volume of the system. When A is further increased so that $\mathcal{L}_s > \mathcal{L}_{c1}$, each wall of the pattern becomes unstable with respect to the fluctuations with the characteristic wave vectors $k_c \sim \epsilon^{-1/3}$. As a result of the development of this instability, fingers will start to grow from the walls of the pattern. Eventually, the volume of the system will become filled with a labyrinthine pattern. The instability will persist until the distance between the walls and their curvature radii become of order $\epsilon^{1/3}$. This phenomenon was observed recently in the experiments by Lee and Swinney [31] and in the numerical simulations of a two-dimensional reaction-diffusion system [33,23].

When A is decreased and the AS narrows, at $\mathcal{L}_s < \mathcal{L}_{cb}$ it destabilizes with respect to the corrugation of its walls with the wave vector $k_c \sim \epsilon^{-1/3}$. As a result, granulation of the static AS will occur and eventually the resulting granules with the radius of order ϵ will disappear.

According to Eqs. (61) and (60), for $\alpha < \epsilon^2$ we have $\mathcal{L}_T < \mathcal{L}_{c2}$. This means that as $\mathcal{L}_s$ is increased, a one-dimensional AS will always transform to a traveling stripe first. Note that the condition $\alpha < \epsilon^2$ here is exact and does not depend in any way on the nonlinearities of the system.

As follows from Eqs. (56) and (60), there may be a rather wide range of the parameters α and ϵ for which the pulsation instability emerges before the instability leading to the formation of traveling AS's as the width of the AS is increased or the value of α is decreased. Indeed, the condition $\mathcal{L}_\omega < \mathcal{L}_T$ is satisfied when

$$\alpha > \epsilon \beta_\omega, \tag{100}$$

where

$$\beta_\omega = \frac{B}{650 Z C^{3/2}}. \tag{101}$$

Note the huge numerical factor in the denominator of Eq. (101). Because of it the instability with respect to pulsations will emerge before the instability to traveling AS's in the majority of real systems. At the same time, as follows from Eqs. (56) and (61), the instability with respect to pulsations is the first, i.e., $\mathcal{L}_\omega < \mathcal{L}_{c2}$, if

$$\alpha < \epsilon^{5/2} \beta_\omega^{-1/2}. \tag{102}$$

These two conditions can be satisfied at the same time only if

$$\epsilon > \beta_\omega. \tag{103}$$

As a result of the instability with respect to pulsations a static AS may collapse or transform into a stationary breathing pattern (pulsating AS), if the parameters of the system are finely adjusted [5,6]. However, as we see from our numerical simulations, in most cases the walls of the AS go so far apart that a local breakdown occurs at the AS center, and eventually the AS produces two one-dimensional AS's traveling in the opposite directions.

When the width of the AS is decreased, at $\alpha < \epsilon^2$ the AS destabilizes with respect to pulsations and disappears, if $\mathcal{L}_s < \mathcal{L}_{b\omega}$, where $\mathcal{L}_{b\omega}$ is given in Eq. (64). When the value of α decreases, at $\alpha = \alpha_c$ we have $\mathcal{L}_{b\omega} = \mathcal{L}_T$. For smaller values of α the AS is always unstable. According to Eqs. (60) and (64), the value of α_c is given by

$$\alpha_c \simeq \epsilon^3 (\ln \epsilon^{-1})^2. \tag{104}$$

The transformations and the evolution of the static spherically symmetric AS in three dimensions and static radially symmetric AS in two dimensions are very similar. These AS's are stable only in relatively narrow range of their radii. When α is big enough, the AS is stable if $\mathcal{R}_b < \mathcal{R}_s < \mathcal{R}_{c2}$, where $\mathcal{R}_b$ and $\mathcal{R}_{c2}$ are of order $\epsilon^{1/3}$ and are given by Eqs. (76) and (78) for the spherically symmetric and by Eqs. (94) and (92) for the radially symmetric AS in two dimensions. As the control parameter A is decreased, at $\mathcal{R}_s < \mathcal{R}_b$ the AS will abruptly disappear. As A is increased and the AS radius becomes greater than $\mathcal{R}_{c2}$, the AS loses stability with respect to the radially nonsymmetric fluctuations with $l=2$ first. The growth of these fluctuations may lead to the split-

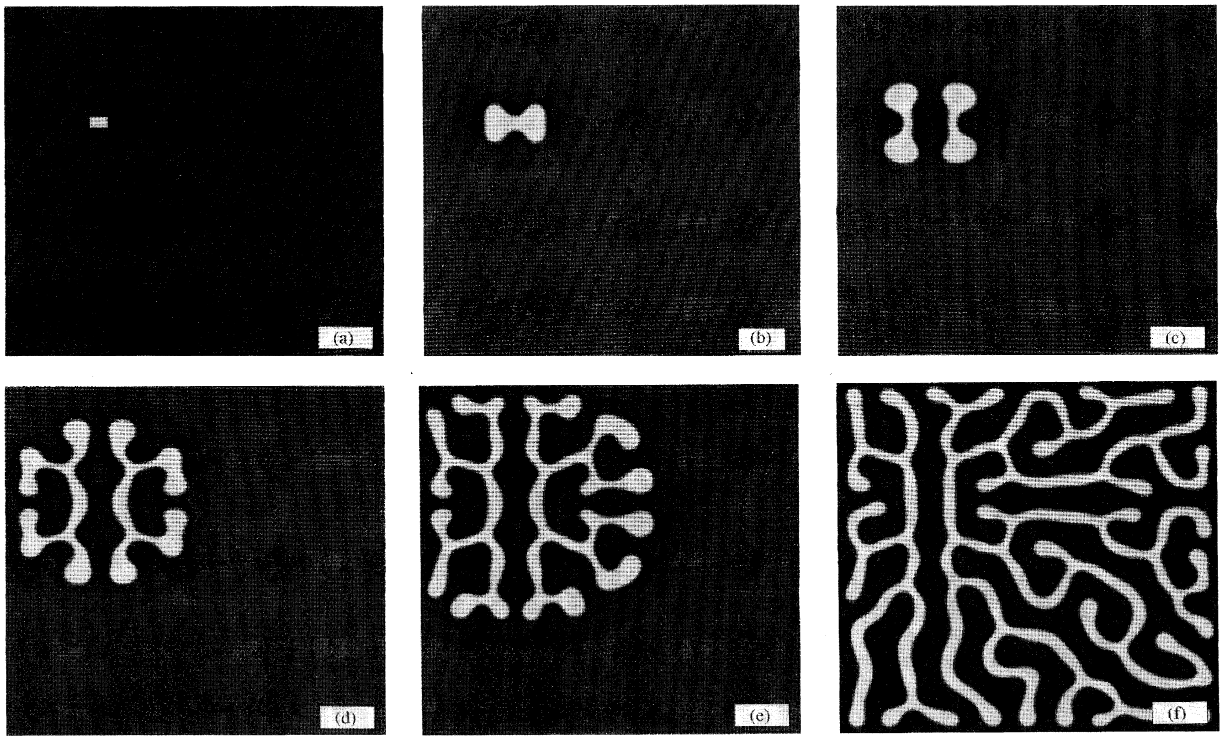

FIG. 8. Formation of a labyrinthine pattern. Numerical solution of Eqs. (3) and (4) with $q=\theta^3-\theta-\eta$ and $Q=\theta+\eta-A$ with $\epsilon=0.05$, $\alpha=0.1$ and $A=-0.3$ at different times. The system size is 20×20. At $t=0$ the homogeneous state of the system is excited in the region 0.5×0.7. Distributions of the activator at times $t=0$, 16.5, 30, 65, 100, 300.

ting of the AS into two, or to the growth of a pattern with sophisticated geometry. This complex pattern may further get more and more complicated as a result of the fingering instability, if the distance between the pattern's walls exceed a value of order $\epsilon^{1/3}$. The process of splitting and complicating will go on until the whole system becomes filled with a very sophisticated labyrinthine pattern (Fig. 8). The pattern should not necessarily be connected because of the possibility of splitting. Thus, there is a remarkable phenomenon characteristic of the considered class of nonlinear systems: *as a result of the instability of an AS localized in a small portion of an extended system the whole system becomes filled with a complicated pattern.* These conclusions explain the effects of splitting, self-replication, and formation of labyrinthine patterns found recently in the experimental and numerical investigations of some two-dimensional reaction-diffusion systems [31,33,23].

Static cylindrically symmetric AS's in three dimensions can only be stable if $\epsilon\gtrsim10^{-3}$. If this condition is satisfied, the cylindrically symmetric AS will destabilize with respect to wriggling ($m=1$ mode) when $\mathcal{R}_s>\mathcal{R}_{c1}$, where $\mathcal{R}_{c1}$ is given by Eq. (98), as its radius is increased. If the radius of the AS is decreased, at $\mathcal{R}_s<\mathcal{R}_b$, where $\mathcal{R}_b$ is given by Eq. (94), the AS will destabilize with respect to the corrugation of its walls [$m=0$ and $k=k_c$, where k_c is given by Eq. (96)]. As a result, the AS will granulate and transform into a number of spherically symmetric AS's.

When the value of α is decreased, a stable radially symmetric AS in two dimensions and a spherically symmetric AS in three dimensions lose their stability with respect to the radially symmetric fluctuations oscillating with some characteristic frequency (see Figs. 4–7). Only a radially symmetric AS in two dimensions whose radius $\mathcal{R}_s\lesssim0.01$ can spontaneously transform to a traveling AS before it destabilizes with respect to the $m=0$ pulsations. However, as we already mentioned, this situation is possible only for unrealistically small values of α and ϵ; therefore this bifurcation, which was recently discussed in Ref. [24], is secondary in most real reaction-diffusion systems. As a result of the instability with respect to radially symmetric pulsations the AS may collapse, or, if the parameters of the system are finely adjusted, a stationary pulsating radially symmetric AS may form [5,6]. However, as we see from our numerical simulations, in most cases the growth of the amplitude of the AS pulsations leads to local breakdown in the AS center and the formation of a traveling wave in the form of a ring with the radius monotonically growing with time.

Static radially symmetric AS's of any radius are always unstable if $\alpha<4.4\epsilon^2$ in three dimensions, or if $\alpha<6.1\epsilon^2$ in two dimensions (in the case of extremely small ϵ and α this value may be greater; see the discussion above). In this situation only traveling waves and pulsating patterns will form in the system. Note that these conclusions are totally independent of the specific nonlinearities of the system.

In our analysis we considered only monostable systems. However, the results obtained by us remain true in bistable systems as well. In particular, it can be easily seen that a static one-dimensional front connecting two stable homogeneous states is always unstable with respect to fluctuations with the wave vector $k\sim\epsilon^{-1/3}$.

So we see that our results are universal and applicable to a wide class of physical, chemical, and biological systems which can be described by Eqs. (1) and (2), if the nullcline of Eq. (1) is N or inverted N (Fig. 1). In such N systems the universality of the obtained results is related to both the form of the nullcline and the smallness of the parameter ϵ. At the same time there are systems which are described by Eqs. (1) and (2), in which the nullcline of Eq. (1) is V or Λ. For example, the biological morphogenesis system of Gierer and Meinhardt [11], the models of axiomatic chemical reactions (the Brusselator [1], and the Gray-Scott model [22]) all have V or Λ nullclines. In such V systems at $\epsilon \ll 1$ spike patterns of giant amplitude may form [5,6]. The properties of such patterns essentially differ from those forming in the N systems we considered here. For this reason it would be incorrect to use the results of present paper to interpret the results of the numerical simulations in V systems. However, the simulations performed by Pearson in the two-dimensional Gray-Scott model showed [22] that the effects of the granulation of static one-dimensional AS's, splitting, and self-replication leading to the formation of complex patterns which fill the whole space are seen in V systems as well.

APPENDIX A: THE EIGENVALUE PROBLEM

Let us consider the exact stability problem for a static pattern. Equations (16) and (17) can be written in operator form as

$$\hat{H}_1 \delta\theta = \hat{U}_1 \delta\eta, \tag{A1}$$

$$\hat{H}_2 \delta\eta = \hat{U}_2 \delta\theta, \tag{A2}$$

where

$$\hat{H}_1 = i\alpha\omega - \epsilon^2 \Delta + q'_\theta, \tag{A3}$$

$$\hat{H}_2 = i\omega - \Delta + Q'_\eta, \tag{A4}$$

and

$$\hat{U}_1 = -q'_\eta, \quad \hat{U}_2 = -Q'_\theta. \tag{A5}$$

Substituting Eq. (A2) into Eq. (A1), we obtain

$$(\hat{H}_1 - \hat{U}_1 \hat{H}_2^{-1} \hat{U}_2)\delta\theta = \lambda\,\delta\theta, \tag{A6}$$

where λ should be put to zero. Solving this eigenvalue problem and then requiring that $\lambda = 0$, we may obtain the value of ω. In fact, this allows us to think of λ as an infinitesimally small quantity.

In the problem considered the operator $\hat{U}_1 \hat{H}_2^{-1} \hat{U}_2$ may be treated as a perturbation to the operator $\hat{H}_1$ [5,6]. We would like to find the solution of Eq. (A6) corresponding to the lowest eigenvalue. In view of the discussion in Sec. III, to the zeroth order in ϵ the eigenfunctions of the operator $\hat{H}_1$ are linear combinations of the functions $\delta\theta_{il}^{(0)} = \sqrt{\epsilon/Z}\,\delta\theta_{il}$ where $\delta\theta_{il}$ are defined in Eq. (22) and Z is defined in Eq. (39) (the coefficient in front of $\delta\theta_{il}$ ensures the proper normalization), and their corresponding eigenvalues are of order ϵ. It can be easily seen that

$$\langle i'l' | \hat{U}_1 \hat{H}_2^{-1} \hat{U}_2 | il \rangle \sim \epsilon, \tag{A7}$$

where the matrix element is calculated with the functions $\delta\theta_{il}^{(0)}$. However, one should be careful in calculating λ since the matrix element from the bound state $\delta\theta_{il}^{(0)}$ to the state of the continuous spectrum of the operator $\hat{H}_1$ with the wave vector $k \sim 1$ has the following estimate:

$$\langle k | \hat{U}_1 \hat{H}_2^{-1} \hat{U}_2 | il \rangle \sim \sqrt{\epsilon}. \tag{A8}$$

So the second- and higher-order corrections of perturbation theory given by the transitions from the bound states to the long-wave continuous spectrum will be of the same order as the first-order contribution from the diagonal element.

According to Eq. (A6) and the fact that the unperturbed eigenvalues of the operator $\hat{H}_1$ are of order ϵ, the improved function $\delta\theta_{il}^{(1)}$ which contains the corrections of order $\sqrt{\epsilon}$ can be written as

$$\delta\theta_{il}^{(1)} = \{1 + \hat{H}_1^{-1} \hat{U}_1 \hat{H}_2^{-1} \hat{U}_2 + (\hat{H}_1^{-1} \hat{U}_1 \hat{H}_2^{-1} \hat{U}_2)^2 + \cdots\}\delta\theta_{il}^{(0)}. \tag{A9}$$

Of course, as it should in the perturbation theory, the operator $\hat{H}_1^{-1}$ actually projects out the $\delta\theta_{il}^{(0)}$ components. If we now substitute this function for $\delta\theta$ into Eq. (A6), multiply it by $\delta\theta_{il}^{(0)*}$, and then integrate over the volume of the system, to the first order in ϵ we will arrive at the equation

$$\langle i'l' | \hat{H}_1 | il \rangle - \langle i'l' | \hat{R} | il \rangle = \lambda\,\delta_{ii'}\delta_{ll'}, \tag{A10}$$

where

$$\hat{R} = \hat{U}_1 \hat{H}_2^{-1} \hat{U}_2 \{1 + \hat{H}_1^{-1} \hat{U}_1 \hat{H}_2^{-1} \hat{U}_2 + (\hat{H}_1^{-1} \hat{U}_1 \hat{H}_2^{-1} \hat{U}_2)^2 + \cdots\}. \tag{A11}$$

One should not confuse the matrix elements in Eq. (A10) with those of Eqs. (36) and (38), since they are calculated with eigenfunctions which have a different normalization.

To the first order in ϵ, we may replace the true distributions of the activator and the inhibitor in the operator $\hat{R}$ by smooth distributions. According to Eq. (A9), the function $\delta\theta_{sm} = \delta\theta_{il}^{(1)} - \delta\theta_{il}^{(0)}$ has the characteristic length scale 1. If we neglect the term $\alpha\omega$ in Eq. (A3), the operator $\hat{H}_1^{-1}$ reduces to $q'_\theta(\theta_{sm}(x), \eta_{sm}(x))^{-1}$. Then the definition of the operator $\hat{R}$ in Eq. (A11), together with the above mentioned property of the operator $\hat{H}_1$, means that in the calculation of the inhibitor response one should consider the fluctuations $\delta\eta$ and $\delta\theta_{sm}$ to be related by Eq. (25). With all these approximations, Eq. (A10) is equivalent to Eq. (38).

APPENDIX B: PIECEWISE-LINEAR MODEL

It seems that the only model in which it is possible to find the exact Green's function of Eq. (31) is the well-known piecewise-linear model of a reaction-diffusion system, which is described by the equations [13]

$$\alpha \frac{\partial\theta}{\partial t} = \epsilon^2 \Delta\theta - \theta - \eta + H(\theta - A), \tag{B1}$$

$$\frac{\partial \eta}{\partial t}=\Delta \eta+\theta-\gamma \eta, \tag{B2}$$

where $H(x)$ is the Heaviside function.

The homogeneous state of this system is $\theta_h=0$, $\eta_h=0$. It can be easily verified that the values of the parameters describing the sharp distribution of the activator are $\theta_{s1}=A-\frac{1}{2}$, $\theta_{s2}=A$, $\theta_{s3}=A+\frac{1}{2}$, and $\eta_s=\frac{1}{2}-A$ [13,16–18]. In view of Eqs. (B1), (B2), (37), and (32), we obtain that in this model $B=1$ and $C=1+\gamma$.

In order to find Z we need to know the sharp distribution. According to Eq. (10), for this model

$$\theta_{sh}(\rho)=\begin{cases} A-\dfrac{1}{2}+\dfrac{1}{2}\exp(\rho/\epsilon), & \rho<0 \\ A+\dfrac{1}{2}-\dfrac{1}{2}\exp(-\rho/\epsilon), & \rho>0. \end{cases} \tag{B3}$$

From Eqs. (B3) and (39) we obtain that $Z=1/4$. Note that B, C, and Z are just constants independent of A.

Having calculated the constants B, C, and Z, we can easily find all instability points. Let us consider the one-dimensional AS, for example. According to Eq. (55), for $\alpha \gg \epsilon$ the AS becomes unstable with respect to symmetric fluctuations with

$$k_c=1.13\epsilon^{-1/3} \quad \text{at} \quad \mathscr{L}_c=1.66\epsilon^{1/3}, \tag{B4}$$

whereas, according to Eq. (56), in the case $\alpha \ll \epsilon$ it destabilizes with respect to fluctuations with

$$\omega_c=1.84\left(\frac{\alpha}{\epsilon}\right)^{-2/3} \quad \text{at } \mathscr{L}_\omega=0.60\left(\frac{\alpha}{\epsilon}\right)^{1/3}. \tag{B5}$$

Equation (B5) improves the accuracy of the results obtained in Ref. [25].

Similarly, from Eq. (59) one concludes that in this system $b=1/(2\sqrt{1+\gamma})$. According to Eq. (61), in the case $\alpha \gg \epsilon$ a one-dimensional AS becomes unstable with respect to antisymmetric fluctuations with $k \to 0$ at

$$\mathscr{L}_s>\mathscr{L}_{c2}=(1+\gamma)^{1/4}(2\epsilon)^{1/2}, \tag{B6}$$

what agrees with the result of Ref. [47]. In the case $\alpha \ll \epsilon$, according to Eq. (60), a static AS spontaneously transforms into a traveling AS when

$$\mathscr{L}_s>\mathscr{L}_T=(1+\gamma)^{1/4}\left(\frac{2\alpha}{\epsilon}\right)^{1/2}. \tag{B7}$$

This result coincides with the one obtained in Ref. [25].

When $\mathscr{L}_s$ becomes comparable with ϵ, an AS becomes unstable with respect to symmetric fluctuations. According to Eq. (63), for $\alpha \gg \epsilon$ the instability occurs at $\mathscr{L}_s<\mathscr{L}_b$ with respect to fluctuations with $k=k_c$, where

$$k_c=1.26\epsilon^{-1/3}, \quad \mathscr{L}_b=\frac{4\epsilon}{3}\ln \epsilon^{-1}, \tag{B8}$$

and, for $\alpha \ll \epsilon$, according to Eq. (64), at $\mathscr{L}_s<\mathscr{L}_{b\omega}$ with respect to the fluctuations with $\omega=\omega_c$, where

$$\omega_c=2\left(\frac{\alpha}{\epsilon}\right)^{-2/3}, \quad \mathscr{L}_{b\omega}=-\frac{\epsilon}{3}\ln \alpha\epsilon^2, \tag{B9}$$

since in this case $\tilde{l}=\epsilon$. Similarly, in one dimension $\mathscr{L}_b=\epsilon\ln\epsilon^{-1}$.

As we noted in Sec. IV, there is a one-to-one correspondence between the AS width $\mathscr{L}_s$ and the control parameter A. In this system this correspondence is given by

$$A=\frac{\gamma+e^{-\mathscr{L}_s\sqrt{1+\gamma}}}{2(1+\gamma)}, \tag{B10}$$

where $\gamma/2(1+\gamma)<A<\frac{1}{2}$. Thus, knowing the critical values of $\mathscr{L}_s$, we can easily calculate the values of A at which the instabilities occur.

Finally, the value of β_ω, which appears in Eq. (101) and determines the region where the AS becomes unstable with respect to pulsations before it transforms into a traveling AS, for this model is

$$\beta_\omega=6.2\times 10^{-3}(1+\gamma)^{-3/2}. \tag{B11}$$

This formula also improves the accuracy of the results obtained earlier in Ref. [25].

Because of the singular character of the nonlinearity, the potential defined in Eq. (33) is identically zero, so the dispersion relation for the one-dimensional AS of an arbitrary size in this model becomes

$$i\alpha\omega+\epsilon^2k^2+\lambda_0=-\frac{2\epsilon}{\sqrt{1+\gamma+k^2+i\omega}} \times\{1\pm e^{-\mathscr{L}_s\sqrt{1+\gamma+k^2+i\omega}}\}, \tag{B12}$$

where the plus sign goes with symmetric fluctuations, whereas the minus sign goes with the antisymmetric ones. According to Eq. (46),

$$\lambda_0=-\frac{2\epsilon}{\sqrt{1+\gamma}}\{1-e^{-\mathscr{L}_s\sqrt{1+\gamma}}\}. \tag{B13}$$

One can see that Eq. (B12) coincides with Eq. (5.4b) of Ref. [47] obtained by a different method.

As in the case of one-dimensional AS's, the unperturbed Green's functions from Eqs. (72) and (88) for spherically and cylindrically symmetric AS's, respectively, are the exact Green's functions. For this reason the exact dispersion relations for radially symmetric AS's in this model are given by Eqs. (70) and (86) with the functions $R_l(\omega)$ and $R_m(k,\omega)$ given by Eqs. (74) and (90), respectively. It is easy to see that the dispersion relations obtained in this fashion coincide with those obtained in Ref. [47] as well.

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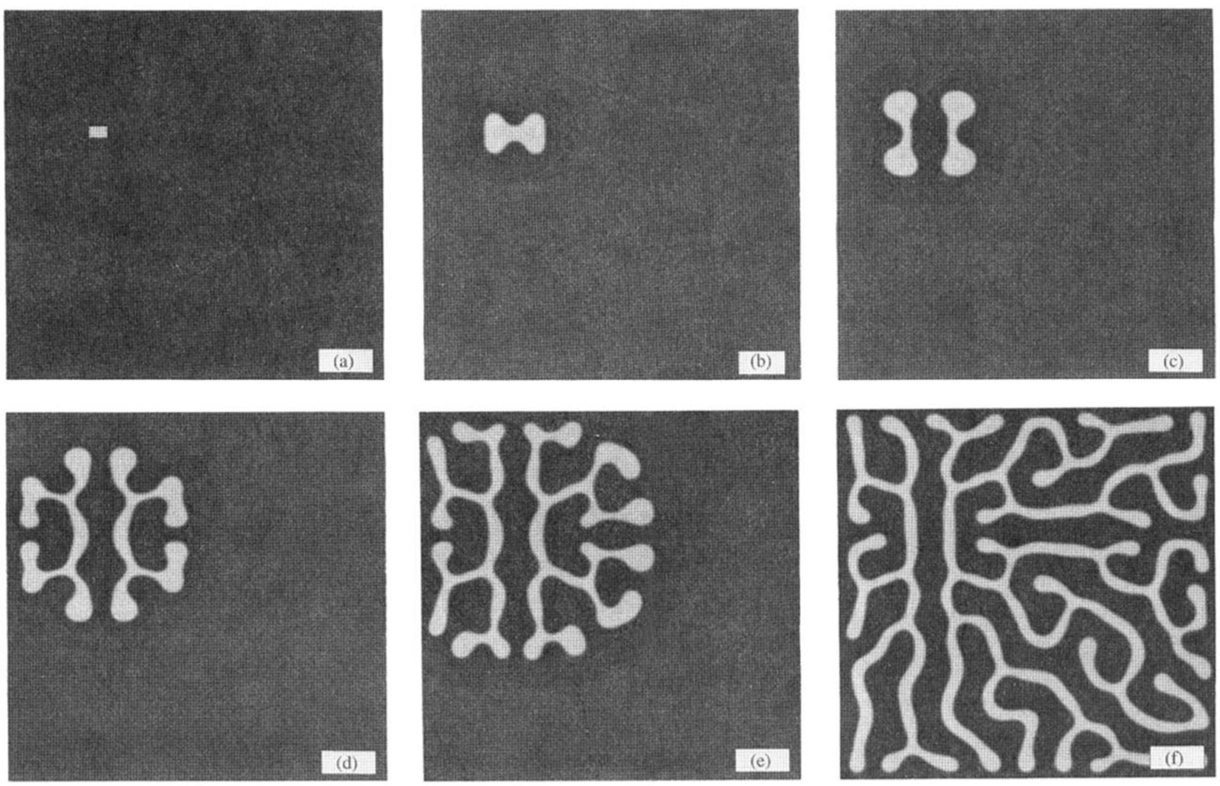

FIG. 8. Formation of a labyrinthine pattern. Numerical solution of Eqs. (3) and (4) with $q=\theta^3-\theta-\eta$ and $Q=\theta+\eta-A$ with $\epsilon=0.05$, $\alpha=0.1$ and $A=-0.3$ at different times. The system size is 20×20. At $t=0$ the homogeneous state of the system is excited in the region 0.5×0.7. Distributions of the activator at times $t=0$, 16.5, 30, 65, 100, 300.