

Interaction of a Hopf Bifurcation and a Symmetry-Breaking Bifurcation: Stochastic Potential and Spatial Correlations

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The multivariate master equation for a general reaction-diffusion system is solved perturbatively in the stationary state, in a range of parameters in which a symmetry-breaking bifurcation and a Hopf bifurcation occur simultaneously. The *stochastic potential* U is, in general, not analytic. However, in the vicinity of the bifurcation point and under precise conditions on the kinetic constants, it is possible to define a fourth-order expansion of U around the bifurcating fixed point. Under these conditions, the domains of existence of different attractors, including spatiotemporal structures as well as the spatial correlations of the fluctuations around these attractors, are determined analytically. The role of fluctuations in the existence and stability of the various patterns is pointed out.

KEY WORDS: Multivariate master equation; reaction-diffusion system; codimension-two bifurcation; nonanalytic potential; spatiotemporal structure; fluctuations; spatial correlations.

1. INTRODUCTION

The description of non-equilibrium systems near bifurcation points has been considerably developed in the last years. From a deterministic point of view, local bifurcations may be studied analytically through the use of the normal form⁽¹⁾ of the differential equations expanded around a fixed point. Bifurcation cascades, which usually give rise to global phenomena, are also frequently amenable to local analysis by bringing the system to the vicinity of a degenerate situation thanks to the control of a sufficient number of parameters. Among such *high codimension bifurcations*, the case in

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which the linearization of the equations around a fixed point gives rise to a simple zero eigenvalue and a simple pair of pure imaginary eigenvalues has been investigated extensively.⁽²⁻⁴⁾ Depending on the parameter values, a wide range of behaviors is observed. So far, the analysis of this codimension two bifurcation has been limited to abstract dynamical systems or to physical systems of *small size*.^(2,4) In both cases, by virtue of the center manifold theorem, it is guaranteed that the evolution can be restricted to a three-dimensional manifold in which the *critical variables* obey a set of coupled ordinary nonlinear differential equations. Notice that the existence of universal unfoldings around criticality remains an open question.

In the present paper, attention is focussed on a general class of reaction-diffusion systems of *large spatial extension* operating in the vicinity of a local codimension two bifurcation of the type mentioned above. Specifically, it is assumed that for a critical set of parameter values λ_0, μ_0 the spectrum of the linear stability operator is quasi-continuous and behaves in the way depicted in Fig. 1. It is expected that for slight deviations of λ, μ from these critical values, a symmetry-breaking bifurcation will interact with a Hopf bifurcation, giving rise to interesting dynamical phenomena. We show how the center-manifold theory extends in this case and obtain the *normal form* of the equations for the critical variables. The latter are expected to define a three-component vector whose components are *fields* obeying coupled nonlinear partial differential equations. Actually, we shall deal with the discretized form of these equations obtained by dividing the physical space into cells of sub-macroscopic size.

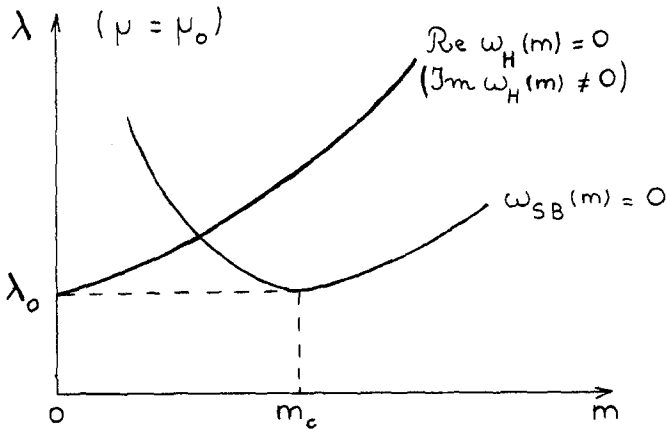


Fig. 1. Linear stability diagram associated with the *degenerescence* of a symmetry-breaking bifurcation [$\omega_{SB}(m) = 0$] and a Hopf bifurcation [$\text{Re } \omega_H(m) = 0$]. For $\mu = \mu_0$, the bifurcation parameter λ is plotted against wave number m .

The main objective of the present work is however to carry out a *stochastic analysis* of this kind of bifurcation and find the probabilistic analogue of the center manifold and normal forms theory. Rather than start with the deterministic laws of evolution and add random (Langevin) forces, we prefer to adopt the master equation description,⁽¹¹⁾ in which fluctuations arise as natural consequences of the very dynamics of the system. We shall model chemical reactions as birth-and-death processes and diffusion as a random walk. Our purpose will be to find asymptotic (long-time) solutions for the multivariate probability distribution. We recall that for simple (nondegenerate) symmetry-breaking bifurcations⁽⁵⁻⁷⁾ and Hopf bifurcations⁽⁸⁻¹⁰⁾ this program has been carried out using both Langevin^(5,8,9) and master equation^(6,7,10) descriptions.

The main thrust of our method is to seek for asymptotic expressions of the cologarithm of the probability distribution U , hereafter referred to as *stochastic potential*.⁽¹²⁾ In previous analyses dealing with simple bifurcations, adequate expressions were obtained by expanding U in Taylor series around the (unstable) fixed point and truncating to fourth-order terms. However, this procedure breaks down in the case of degenerate bifurcations, unless specific conditions are imposed on the coefficients of the master equation. The existence of a smooth potential in these more complex situations is a question of interest for several authors^(13,14) who analyze this problem in a different context and for systems with finite degrees of freedom. More explicitly, the general problem of the existence of polynomial expansions of the stochastic potential has been recently analysed in detail by Descalzi and Tirapegui⁽²⁰⁾ in a context similar to our own previous short work.⁽¹⁵⁾

We explore here the dynamical behavior of the system under the conditions of existence of a quartic potential, and compute the spatial correlations of the fluctuations around the possible attractors. We show that in low-dimensional systems, long-range order cannot be sustained. It is thus expected that for such systems fluctuations will destroy the attractors predicted by the deterministic analysis.

The paper is organized as follows: in Section 2, we recall briefly the main principles of the perturbation method used to solve the master equation. The conditions of existence of an analytic stochastic potential are determined in the case of the codimension two bifurcation of interest. Section 3 is devoted to the search of the bifurcating attractors as extrema of the potential. Finally, we estimate in Section 4 the spatial correlations of the fluctuations around these attractors.

2. SEARCH OF THE STOCHASTIC POTENTIAL ASSOCIATED WITH A CODIMENSION-TWO BIFURCATION

The perturbation method used to solve the multivariate master equation associated with a general reaction-diffusion system has been introduced by Kubo *et al.*⁽¹⁷⁾ and developed further by Lemarchand and Nicolis.^(6,7) We summarize here the main steps of the procedure leading to a Taylor expansion of the stationary stochastic potential around a fixed point.

2.1. Main Principles of the Perturbation Method

The reaction-diffusion model consists of a set of chemically active constituents in a volume \mathcal{V} in a d -dimensional space. This space is divided into n submacroscopic cells. The numbers of cells along each axis are denoted by n_1, n_2, \dots, n_d such that $n_1 \times n_2 \times \dots \times n_d = n$. A vector $\mathbf{r} = (r_1, r_2, \dots, r_d)$, with integer components locates a given cell. The number of particles of species α in a cell \mathbf{r} will be denoted $X_{\mathbf{r}\alpha}$. The kinetic characteristics of the chemical reactions are the following.

$\bar{v}_{\rho\alpha}$ is the order of the ρ th reaction with respect to X_α .

$v_{\rho\alpha}$ is the stoichiometric coefficient of X_α in the ρ th reaction ($v_{\rho\alpha} > 0$ for particles formed as a result of the reaction and $v_{\rho\alpha} < 0$ for particles disappearing as a result of the reaction).

k_ρ is the normalized kinetic constant of the ρ th reaction, including externally controlled concentrations.

Each constituent X_α may diffuse between two adjacent cells with a jump frequency D_α depending on the length Δl of the cell and related to Fick's coefficient \mathcal{D}_α through $\mathcal{D}_\alpha = D_\alpha(\Delta l)^2$.

The usual stochastic description⁽¹¹⁾ of chemical reactions as birth-and-death processes and of diffusion as a random walk between adjacent cells leads to the multivariate master equation for the probability distribution $P(\{X_{\mathbf{r}\alpha}\})$:

$$\begin{aligned} \frac{dP}{dt} = & \sum_{\rho} k_{\rho} \sum_{\mathbf{r}} \left[\left(\prod_{\alpha} \frac{(X_{\mathbf{r}\alpha} - v_{\rho\alpha})!}{(X_{\mathbf{r}\alpha} - \bar{v}_{\rho\alpha} - v_{\rho\alpha})!} \right) P(\{X_{\mathbf{r}\alpha} - v_{\rho\alpha}\}) \right. \\ & \left. - \left(\prod_{\alpha} \frac{X_{\mathbf{r}\alpha}!}{(X_{\mathbf{r}\alpha} - \bar{v}_{\rho\alpha})!} \right) P \right] \\ & + \sum_{\alpha} D_{\alpha} \sum_{\mathbf{r}\mathbf{z}} ((X_{\mathbf{r}\alpha} + 1) P(X_{\mathbf{r}\alpha} + 1, X_{(\mathbf{r}+\mathbf{a})\alpha} - 1) - X_{\mathbf{r}\alpha} P) \quad (2.1) \end{aligned}$$

where \mathbf{a} denotes the first neighbors of cell \mathbf{r} . Only the arguments of P which differ from $\{X_{\mathbf{r}\alpha}\}$ are explicitly indicated.

Introducing N , the mean number of particles in a cell, as an expansion parameter, we assume that the probability distribution has the asymptotic form

$$P(\{X_{\mathbf{r}\alpha}\}, t) = C(N) \exp\{-nN[U(\{x_{\mathbf{r}\alpha}\}, t) + O(1/N)]\} \quad (2.2)$$

where $C(N)$ is a normalization constant and where the *stochastic potential* U is defined as a continuous function of the reduced variables $x_{\mathbf{r}\alpha} = X_{\mathbf{r}\alpha}/N$.

Substituting Eq. (2.2) into Eq. (2.1) and expanding in terms of $1/N$, we obtain to zeroth order a Hamilton–Jacobi type of equation,

$$-\frac{\partial U}{\partial t} = H(\{x_j\}, \{U^j\}) = \sum_{\mathbf{r}} M(\{x_{\mathbf{r}\alpha}\}, \{U^{\mathbf{r}\alpha}\}) + \mathbf{H}^{\text{diff}}(\{x_j\}, \{U^j\}) \quad (2.3)$$

The “Hamiltonian” H is a function of the variables x_j (we use here the contracted notation $j = \mathbf{r}\alpha$) and of their “conjugate momenta” $U^j = \partial U / \partial x_j$. It can be expressed as a sum of a term associated with diffusion processes

$$H^{\text{diff}}(\{x_j\}, \{U^j\}) = \sum_{\alpha} D_{\alpha} \sum_{\mathbf{r}\mathbf{a}} x_{\mathbf{r}\alpha} \left\{ \exp \left[-\frac{\partial U}{\partial x_{\mathbf{r}\alpha}} + \frac{\partial U}{\partial x_{(\mathbf{r}+\mathbf{a})\alpha}} \right] - 1 \right\}$$

and terms such as $M(\{x_{\mathbf{r}\alpha}\}, \{U^{\mathbf{r}\alpha}\})$ associated with the chemical reactions in a cell \mathbf{r} . The *function* $M(\{x_{\alpha}\}, \{U^{\alpha}\})$ itself is space independent and it coincides with the Hamiltonian of a purely chemical system without diffusion. It is given by

$$M(\{x_{\alpha}\}, \{U^{\alpha}\}) = \sum_{\rho} k_{\rho} \left(\prod_{\alpha} x_{\alpha}^{v_{\rho\alpha}} \right) \left[\exp \left(\sum_{\alpha} v_{\rho\alpha} U^{\alpha} \right) - 1 \right]$$

The derivatives of H (respectively M) with respect to x_j, x_j, \dots (respectively $x_{\alpha}, x_{\alpha}, \dots$) and U^j, U^j, \dots (respectively $U^{\alpha}, U^{\alpha}, \dots$), evaluated for $U^j = 0$ (respectively $U^{\alpha} = 0$) will be denoted by $H_{j_1 j_2 \dots}^j(\{x_j\})$ [respectively $M_{\alpha_1 \alpha_2 \dots}^{\alpha}(\{x_{\alpha}\})$]. It can be shown^(6,7) that H_j, H_{jj}, \dots are the first, second, ... moments of the transition probability. Specifically, H_j is the vector field of the deterministic equations. Drawing a parallel between a local deterministic analysis beginning with an expansion of the differential equations around a fixed point (i.e., $H_j = 0$) and our stochastic approach, we seek a stationary solution of Eq. (2.3) in the form of a Taylor expansion around an extremum $\bar{x}_j(t)$ of U (i.e., $\partial U / \partial x_j = 0$). It is possible to show^(6,7) that the extrema of U obey the deterministic equations

$$d\bar{x}_j/dt = H_j(\{\bar{x}_j\}) \quad (2.4)$$

Choosing then to expand U around an extremum $\bar{x}_{r\alpha}(t) = \bar{x}_\alpha$ which is a stationary homogeneous state of the deterministic equations yields

$$U = \frac{1}{2} U^{l^1 l^2} \sigma_{l^1} \sigma_{l^2} + \frac{1}{3!} U^{l^1 l^2 l^3} \sigma_{l^1} \sigma_{l^2} \sigma_{l^3} + \dots \tag{2.5}$$

where the indices with numerical exponents such as l^1 are implicitly summed over and where σ_i are the new coordinates in the representation of the eigenvectors C^l of the linear stability operator $H_j^{l'}$ at the reference state \bar{x}_α :

$$x_j - \bar{x}_\alpha = C_j^{l^1} J_{l^1} \tag{2.6}$$

For periodic boundary conditions, suitable for the description of large systems, the components $C_j^l = C_{r\alpha}^{m\beta}$ of the eigenvector C^l associated with the eigenvalue $\omega_{m\beta}$ satisfy

$$C_j^l = (c_m)_\alpha^\beta e^{i r m} \tag{2.7}$$

where $\mathbf{m} = 2\pi(m_1/n_1, m_2/n_2, \dots, m_d/n_d)$ with $m_i \in \mathbb{N}$ plays the role of a Fourier variable and where $(c_m)_\alpha^\beta$ obeys^(6,7)

$$[M_\alpha^{\alpha^1} - K_m D_\alpha \delta(\alpha - \alpha^1)] (c_m)_{\alpha^1}^\beta = \omega_{m\beta} (c_m)_{\alpha^1}^\beta \tag{2.8}$$

with

$$K_m = \sum_{i=1}^d 4 \sin^2 \pi m_i / n_i \tag{2.9}$$

As the number n of cells (size of the system) is large, the eigenvalue spectrum is dense and so is the set of K_m . It should be noted here that in the limit of a continuous space description ($\Delta l \rightarrow 0$, where Δl is the cell's side length), the wave vectors \mathbf{p} are defined as $\mathbf{m}/\Delta l \rightarrow \mathbf{p}$ and it can be shown that $K_m/(\Delta l)^2 \rightarrow \mathbf{p}^2$.

In the representation σ_i , the successive derivatives of U have the following expressions^(6,7):

$$U_{ll'}^{-1} = -\frac{H_{ll'}}{\omega_l + \omega_{l'}} \tag{2.10}$$

$$U^{ll'lr} = \frac{U^{ll'} U^{l'l^2} U^{l''l^3}}{\omega_{l^1} + \omega_{l^2} + \omega_{l^3}} [\mathcal{S}_{l^1 l^2 l^3} (H_{l^1 l^5}^{-1} U_{l^4 l^2}^{-1} U_{l^5 l^3}^{-1} + H_{l^1 l^2}^4 U_{l^4 l^3}^{-1}) + H_{l^1 l^2 l^3}] \tag{2.11}$$

The symmetrization symbol $\mathcal{S}_{ll' \dots}$ applied to a function of the indices $ll' \dots$ represents the sum of all distinct terms obtained by permutation of these

indices. These exact expressions have already been evaluated in the vicinity of a codimension-one bifurcation leading either to spatial structures^(6,7) or to temporal oscillations.⁽¹⁰⁾ The question is: does the same method apply to a codimension-two bifurcation, leading in particular to spatiotemporal structures?

2.2. Critical Eigenvalues. Quasiresonances

The system of interest is a general reaction-diffusion system of two chemical components and of large size. It depends at least on two parameters η_c and η_0 such that there exist conditions for which a symmetry-breaking bifurcation ($\eta_c = 0$) and a Hopf bifurcation ($\eta_0 = 0$) occur simultaneously. (η_c and η_0 are combinations of the parameters λ and μ referred to in the introduction). Guckenheimer⁽²⁾ shows that these degeneracy conditions may be obtained, e.g., for the trimolecular model, the so-called Brusselator.⁽¹¹⁾ In the vicinity of such a codimension-two bifurcation, a local analysis is possible. At the bifurcation point ($\eta_c = \eta_0 = 0$) all the eigenvalues $\omega_{\mathbf{m}\beta}$ of the linear stability operator H_j^l have negative real parts except one of them, denoted by $\omega_{\mathbf{m}_c 0}$ (or simply $\omega_{\mathbf{m}_c}$), which vanishes, and a pair of complex conjugate eigenvalues denoted by ω_{01} and $\omega_{0\bar{1}}$, which are purely imaginary.

The critical value of the parameter η_c and the critical wave vector \mathbf{m}_c associated with the spatial structure emerging from the instability are defined as solutions of the system

$$\omega_{\mathbf{m}\beta} = 0, \quad \frac{\partial \omega_{\mathbf{m}\beta}}{\partial K_{\mathbf{m}}} = 0 \quad (2.12)$$

with $\omega_{\mathbf{m}\beta}(K_{\mathbf{m}})$ satisfying Eq. (2.8). Because of the presence of a space-dependent part in Eq. (2.8), in a system of large spatial extension (large n), the transition to instability will be marked not only by one eigenvalue going to zero, but by the accumulation to zero of a large number of closely packed eigenvalues $\omega_{\mathbf{m}_0 0}$ (denoted simply by $\omega_{\mathbf{m}_0}$ when there is no ambiguity) characterized by vectors \mathbf{m}_0 of any direction and of modulus close to $|\mathbf{m}_c|$. Taking Eq. (2.12) into account, an expansion of $\omega_{\mathbf{m}_0}$ in powers of η_c and $(K_{\mathbf{m}_0} - K_{\mathbf{m}_c}) = (K_{\mathbf{m}_0} - K_c)$ reduces at dominant order to

$$\omega_{\mathbf{m}_0} = \eta_c + \theta_c (K_{\mathbf{m}_0} - K_c)^2 \quad (2.13)$$

where θ_c is negative and where the bifurcation parameter η_c is taken to be simply $\eta_c = \omega_{\mathbf{m}_0}(K_c)$.

As the bifurcation parameter η_0 vanishes, the first instability leading to temporal oscillations in a system of two chemical components appears for a

vanishing wave vector. Again, because of the large spatial extension of the system, the transition is marked by a set of eigenvalues $\omega_{\mathbf{q}\beta}$ with small real parts characterized by small wave vectors \mathbf{q} or equivalently small values of $K_{\mathbf{q}}$. Their expansion in powers of η_0 and $K_{\mathbf{q}}$ reduces at dominant order to

$$\omega_{\mathbf{q}\beta} = \eta_0 + \eta_1 K_{\mathbf{q}} + i\beta(\theta_0 + \theta_1 K_{\mathbf{q}}) \quad (2.14)$$

where η_1 is negative, and θ_0 and θ_1 are constants. β takes the two values 1 and $\bar{1} = -1$. (We have chosen the second bifurcation parameter to be $\eta_0 = \text{Re } \omega_{\mathbf{q}=0, \beta}$.)

The set of eigenvalues with small real parts $\omega_{\mathbf{m}_0}$ and $\omega_{\mathbf{q}\beta}$ are called “critical eigenvalues” and are denoted globally by ω_{i_0} . The corresponding variables σ_{i_0} are called critical variables. Conversely, the eigenvalues with nonsmall real parts or noncritical eigenvalues are denoted by ω_{i_p} . In the vicinity of the bifurcation point and for a large system, it is now possible to evaluate the successive derivatives (2.10), (2.11),... of U through a systematic expansion in powers of the small parameters $\text{Re } \omega_{i_0}$ which reflect the distance to the bifurcation point through η_c or η_0 as well as the size of the system through $(K_{\mathbf{m}_0} - K_c)$ or $K_{\mathbf{q}}$ [see Eqs. (2.13), (2.14)]. It should be remarked, considering expressions (2.10), (2.11),..., that the derivatives $U^{(1)'} \dots$ of the stochastic potential can be written as a sum of terms, each of which is in inverse ratio to a sum $\omega_{i_1} + \omega_{i_2} + \omega_{i_3} + \dots$ of eigenvalues. It is clear that the terms with a small denominator will be dominant and it can now be understood why the critical eigenvalues play an essential role in our perturbation theory.

We define a relation of *quasiresonance* by

$$\sum_{i=1}^p \omega_{i_0} = O(\text{Re } \omega_{i_0}) \quad \text{with } p \in \mathbb{N}^* \quad (2.15)$$

extending thus the concept of resonance introduced in the theory of normal forms.^(1,4) In the case of the bifurcation of interest, three different types of quasiresonances occur. The relation

$$\sum_{i=1}^p \omega_{\mathbf{m}_0^i} = O(\omega_{\mathbf{m}_0}) \quad \text{with } p \in \mathbb{N}^* \quad (2.16)$$

already defined a quasiresonance of order p for a symmetry-breaking bifurcation^(6,7) and remains true here. The quasiresonance

$$\sum_{i,j=1}^s \omega_{\mathbf{q}^i\beta} + \omega_{\mathbf{q}^j\beta} = O(\text{Re } \omega_{\mathbf{q}\beta}) \quad \text{with } s \in \mathbb{N}^* \quad (2.17)$$

associated with a simple Hopf bifurcation⁽¹⁰⁾ still holds. Note that quasiresonances of this type are of even order $2s$. In the case of the inter-

action of the above bifurcations, a third kind of quasiresonance of order $2s + p$ appears in the form

$$\sum_{i=1}^p \omega_{m_0^i} + \sum_{j,k=1}^s \omega_{q^j\beta} + \omega_{q^k\beta} = O(\text{Re } \omega_{l_0}) \quad \text{with } (p, s) \in (\mathbb{N}^*)^2 \quad (2.18)$$

The first quasiresonance of this third kind is of order three.

The existence of three types of quasiresonances implies that an expansion of the stochastic potential reduces, at dominant order with respect to $\text{Re } \omega_{l_0}$, to a sum of three terms⁽¹²⁾:

$$U = U^{\text{SB}} + U^{\text{H}} + U^{\text{I}} \quad (2.19)$$

where U^{SB} is associated with a symmetry-breaking bifurcation^(6,7) or with the quasiresonances (2.16); U^{H} is associated with a Hopf bifurcation⁽¹⁰⁾ or with the quasiresonances (2.17); U^{I} is an interaction term associated with the quasiresonances (2.18).

2.3. Symmetry-Breaking Bifurcation. Determination of U^{SB}

A symmetry-breaking bifurcation is characterized by a set of critical eigenvalues ω_{m_0} given by Eq. (2.13): ω_{m_0} is real and small in the vicinity of the bifurcation. An expansion of the stochastic potential U^{SB} limited to its quadratic part can only describe the close neighborhood of the reference fixed point ($\sigma_1 = 0$) whenever this point is stable. We know from the deterministic analysis that when the parameter η_c increases above its bifurcation value $\eta_c = 0$, the fixed point is destabilized and gives rise to new attractors. Expecting the stochastic potential to give specific information on this new behavior, Lemarchand and Nicolis^(6,7) carried out the expansion of U^{SB} to higher orders so as to include the description of the bifurcation attractors. They showed that this expansion can be written in a form exhibiting separately the contributions from critical modes and those containing at least one noncritical mode:

$$\begin{aligned} U^{\text{SB}} = & \frac{-\omega_{m_0^1}}{H_{m_c, m_c}} \sigma_{m_0^1} \sigma_{m_0^1} + \frac{1}{3!} U^{m_0^1 m_0^2 m_0^3} \sigma_{m_0^1} \sigma_{m_0^2} \sigma_{m_0^3} \\ & + \frac{1}{4!} (U^{m_0^1 m_0^2 m_0^3 m_0^4} - 3U^{m_0^1 m_0^2 l_\phi^1} U_{l_\phi^1 l_\phi^2}^{-1} U_{l_\phi^2 l_\phi^3}^2 m_0^3 m_0^4) \sigma_{m_0^1} \sigma_{m_0^2} \sigma_{m_0^3} \sigma_{m_0^4} \\ & + \frac{1}{2} U_{l_\phi^1 l_\phi^2}^1 \left(\sigma_{l_\phi^1} + \frac{1}{2} U_{l_\phi^1 l_\phi^3}^{-1} U_{l_\phi^3 l_\phi^4}^3 m_0^1 m_0^2 \sigma_{m_0^1} \sigma_{m_0^2} \right) \\ & \times \left(\sigma_{l_\phi^2} + \frac{1}{2} U_{l_\phi^2 l_\phi^4}^{-1} U_{l_\phi^4 l_\phi^3}^4 m_0^3 m_0^4 \sigma_{m_0^3} \sigma_{m_0^4} \right) \end{aligned} \quad (2.20)$$

where the expression of the second transition moment $H_{\mathbf{m}_c, \bar{\mathbf{m}}_c}$ in terms of the characteristics of the reaction-diffusion model is given in refs. 6 and 7. In this expansion, the coefficients $U^{\mathbf{m}_0 \mathbf{m}'_0 \mathbf{m}''_0}$ are supposed to verify the condition

$$U^{\mathbf{m}_0 \mathbf{m}'_0 \mathbf{m}''_0} \lesssim (\omega_{\mathbf{m}_0})^{1/2} \quad (2.21)$$

which amounts to gathering the bifurcating attractors in a small neighborhood of the fixed point [$|\sigma_{\mathbf{m}_0}| \lesssim (\omega_{\mathbf{m}_0})^{1/2}$].

Equation (2.20) states that, thanks to a nonlinear change of variables defining the new variables

$$s_{l_\phi} = \sigma_{l_\phi} + \frac{1}{2} U_{l_\phi l_\phi}^{-1} U_{l_\phi}^l \mathbf{m}'_0 \mathbf{m}''_0 \sigma_{\mathbf{m}'_0} \sigma_{\mathbf{m}''_0} \quad (2.22)$$

the noncritical modes may be entirely cast in a polynomial of order two [third and fourth lines of Eq. (2.20)]. The probability associated with the variables s_{l_ϕ} is thus simply a Gaussian distribution. In other words, Eq. (2.22) states that the noncritical modes may be adiabatically eliminated as anticipated in ref. 5. It is then possible to define separately a “critical potential” $U_{\text{cr}}^{\text{SB}}$ as a polynomial of order four, depending only on the critical variables [first and second lines of Eq. (2.20)]. The determination of $U_{\text{cr}}^{\text{SB}}$ is sufficient to describe a symmetry-breaking bifurcation:

$$U_{\text{cr}}^{\text{SB}} = \frac{1}{H_{\mathbf{m}_c, \bar{\mathbf{m}}_c}} [-\omega_{\mathbf{m}'_0} \sigma_{\mathbf{m}'_0} \sigma_{\bar{\mathbf{m}}_0} + \gamma \delta(\mathbf{m}'_0 + \mathbf{m}''_0 + \mathbf{m}'''_0) \sigma_{\mathbf{m}'_0} \sigma_{\mathbf{m}''_0} \sigma_{\mathbf{m}'''_0} + v_1 \delta(\mathbf{m}'_0 + \mathbf{m}''_0 + \mathbf{m}'''_0 + \mathbf{m}^{\text{IV}}_0) \sigma_{\mathbf{m}'_0} \sigma_{\mathbf{m}''_0} \sigma_{\mathbf{m}'''_0} \sigma_{\mathbf{m}^{\text{IV}}_0}] \quad (2.23)$$

where γ and v_1 are the dominant orders of the cubic and quartic coefficients with respect to η_c and $(K_{\mathbf{m}_0} - K_c)$. They are identical to the real parts of coefficients of the normal form of the deterministic equations (see the Appendix). Their expressions as a function of the characteristics of the reaction-diffusion model are given in refs. 6 and 7. Let us mention that, according to Eq. (2.21), we impose

$$\gamma \sim (\omega_{\mathbf{m}_0})^{1/2} \quad (2.24)$$

The existence of a critical potential gives the stochastic equivalent of a theorem of bifurcation theory, the *center manifold theorem*.^(3,4) This theorem proves that, thanks to a nonlinear change of variables, it is possible to write locally a system of differential equations in an uncoupled form separating the evolution of the critical variables s_{l_c} from the evolution of the noncritical ones s_{l_ϕ} . The equations of s_{l_ϕ} are linear, whereas the nonlinearities are “concentrated” in the center manifold which is tangent to the eigenspace associated with the critical eigenvalues. The *theory of normal*

forms^(1,4) used in analysis states analogous properties and gives also the way to select the nonlinear terms appearing in the equations of evolution of s_i according as they are associated with *resonances*. Here, the nonquadratic terms which have to be retained in the expansion of the critical stochastic potential are associated with *quasiresonances*. Note that there exists no rigorous deterministic theory about quasiresonances.

Finally, it is interesting to note that changing $\sigma_{\mathbf{m}_0}$ into $\sigma_{\mathbf{m}_0} \exp(i\mathbf{m}_0 \cdot \mathbf{l})$ (where the vector \mathbf{l} corresponds to any translation) leaves the critical potential (2.23) invariant. This translational invariance is reflected in the proportionality of the coefficients of U^{SB} to the Kronecker delta; it has been preserved by the periodic boundary conditions imposed on the system.

2.4. Hopf Bifurcation. Determination of U^H

Considering now a set of complex conjugate critical eigenvalues $\omega_{\mathbf{q}_1}$ and $\omega_{\mathbf{q}_1}$ given by Eq. (2.14), we follow at first the same method as in Section 2.3. Applying Eqs. (2.10), (2.11),... only to quasiresonances (2.17) allows us to determine the dominant contributions of the critical potential U_{cr}^H associated with a Hopf bifurcation. Because of the even order of the quasiresonances, only contributions $U^{(2p)}$ of order $2p$ appear in U_{cr}^H :

$$\begin{aligned}
 U_{cr}^H = & \frac{1}{M_{11}} \left[-2 \operatorname{Re} \omega_{\mathbf{q}_1} \sigma_{\mathbf{q}_1} \sigma_{\mathbf{q}_1} \right. \\
 & + \left(u_1 + \frac{i \underline{K} (\eta_1 u_2 - \theta_1 u_1)}{4\eta_0 + \eta_1 K + i\theta_1 \underline{K}} \right) \delta(\mathbf{q}^1 + \mathbf{q}^2 + \mathbf{q}^3 + \mathbf{q}^4) \sigma_{\mathbf{q}_1} \sigma_{\mathbf{q}_2} \sigma_{\mathbf{q}_3} \sigma_{\mathbf{q}_4} \\
 & \left. + \sum_{p=3}^{\infty} U^{(2p)} \prod_{i=1}^p \prod_{j=p+1}^{2p} \sigma_{\mathbf{q}_i} \sigma_{\mathbf{q}_j} \right] \tag{2.25}
 \end{aligned}$$

where

$$K = K_{\mathbf{q}_1} + K_{\mathbf{q}_2} + K_{\mathbf{q}_3} + K_{\mathbf{q}_4}, \quad \underline{K} = -K_{\mathbf{q}_1} - K_{\mathbf{q}_2} + K_{\mathbf{q}_3} + K_{\mathbf{q}_4}$$

The coefficients u_1 and u_2 , as well as the second transition moment M_{11} , have explicit expressions in terms of the characteristics of the model.⁽¹⁰⁾ Let us emphasize that the noise component M_{11} does not need a “non-equilibrium fluctuation-dissipation theorem” to be expressed in terms of the dissipative characteristics of the system, as it does in the Langevin approach.⁽⁵⁾ Apart from this, our method yields the same form of fourth-order potential as obtained by Walgraef *et al.*

An essential point which has not been discussed in ref. 5 or in ref. 10 is that the fourth-order terms have no limit as η_0 , \underline{K} , and K tend to zero unless the condition

$$\eta_1 u_2 - \theta_1 u_1 = 0 \quad (2.26)$$

is satisfied.

Moreover, we prove through a recurrence that higher order derivatives $U^{(2p)}$ with $p > 2$ satisfy

$$U^{(2p)} = (\eta_1 u_2 - \theta_1 u_1) O(\text{Re } \omega_{q_1})^{2-p} \quad (2.27)$$

so that they diverge when $\text{Re } \omega_{q_1} \rightarrow 0$ if condition (2.26) is not satisfied. In this latter case, an expansion of U_{cr}^H is only valid in a neighborhood of the fixed point defined by

$$\sigma_{q_1} \ll (\text{Re } \omega_{q_1})^{1/2} \quad (2.28)$$

U_{cr}^H reduces then to its quadratic part and does not bring any new information about the probability surface after the bifurcation.

Hence, condition (2.26) appears as a necessary and sufficient condition to define a fourth-order expansion of U_{cr}^H , valid in a neighborhood of the fixed point, defined by

$$\sigma_{q_1} \lesssim (\text{Re } \omega_{q_1})^{1/2} \quad (2.29)$$

In the sequel, the condition (2.26) is supposed satisfied, so that U_{cr}^H reduces to

$$U_{\text{cr}}^H = \frac{1}{M_{11}} [-2 \text{Re } \omega_{q_1} \sigma_{q_1} \sigma_{q_1^\dagger} + u_1 \delta(\mathbf{q}^1 + \mathbf{q}^2 + \mathbf{q}^3 + \mathbf{q}^4) \sigma_{q_1} \sigma_{q_2} \sigma_{q_3} \sigma_{q_4}] \quad (2.30)$$

This simple form was previously obtained by Szeffalussy and Tel,⁽⁸⁾ who mentioned condition (2.26) as a special case which allowed them to reduce the model to a solvable "time-dependent Ginzburg–Landau model," well known in the critical dynamics of the equilibrium phase transition. Our systematic method proves that this condition is the only one under which such a polynomial form of U_{cr}^H is valid.

Note, finally, as for $U_{\text{cr}}^{\text{SB}}$, that U_{cr}^H possesses an invariance property: it remains unchanged under any phase deviation in the form $\sigma_{q_1} \rightarrow \sigma_{q_1} e^{i\phi}$.

2.5. Interaction Term. Determination of U^I

One of the major advantages of our method is its generality. Using the concept of quaresonance, we are able to select all the relevant terms in the

Taylor series (2.5), whatever their origin. In particular, the quairesonances of the third kind [Eq. (2.18)], originating from the complex situation of degenerate bifurcations, can be handled in the same way as the simple cases previously discussed.

We first state that the third derivatives (2.11) associated with the quairesonance

$$\omega_{\mathbf{m}_0} + \omega_{\mathbf{q}\beta} + \omega_{\mathbf{q}'\beta} = O(\text{Re } \omega_{l_0})$$

always vanish due to their proportionality^(6,7) to $\delta(\mathbf{m}_0 + \mathbf{q} + \mathbf{q}')$. Knowing the expressions (2.13), (2.14) of the critical eigenvalues, we evaluate the fourth-order derivatives associated with the quairesonance

$$\begin{aligned} Q &= \omega_{\mathbf{m}_0} + \omega_{\mathbf{m}'_0} + \omega_{\mathbf{q}\beta} + \omega_{\mathbf{q}'\beta} \\ &= 2\eta_c + \theta_c [(K_{\mathbf{m}_0} - K_c)^2 + (K_{\mathbf{m}'_0} - K_c)^2] \\ &\quad + 2\eta_0 + \eta_1(K_{\mathbf{q}} + K_{\mathbf{q}'}) + i\theta_1(-K_{\mathbf{q}} + K_{\mathbf{q}'}) \end{aligned} \tag{2.31}$$

and obtain the rather involved expression

$$\begin{aligned} &U^{\mathbf{m}_0\mathbf{m}'_0\mathbf{q}\mathbf{q}'1} \\ &= \delta(\mathbf{m}_0 + \mathbf{m}'_0 + \mathbf{q} + \mathbf{q}') \left(\frac{\eta_c A/H_{\mathbf{m}_c\bar{\mathbf{m}}_c} + \eta_0 B_1/M_{1\bar{1}}}{\eta_0 + \eta_c} \right. \\ &\quad + \theta_c [(K_{\mathbf{m}_0} - K_c)^2 + (K_{\mathbf{m}'_0} - K_c)^2] \left(\frac{A}{H_{\mathbf{m}_c\bar{\mathbf{m}}_c}} - \frac{B_1}{M_{1\bar{1}}} \right) \frac{\eta_0}{Q(\eta_0 + \eta_c)} \\ &\quad - \eta_1(K_{\mathbf{q}} + K_{\mathbf{q}'}) \left(\frac{A}{H_{\mathbf{m}_c\bar{\mathbf{m}}_c}} - \frac{B_1}{M_{1\bar{1}}} \right) \frac{\eta_c}{Q(\eta_0 + \eta_c)} \\ &\quad \left. - i(-K_{\mathbf{q}} + K_{\mathbf{q}'}) \frac{(A/H_{\mathbf{m}_c\bar{\mathbf{m}}_c})\theta_1\eta_c + (B_1/M_{1\bar{1}})\theta_1\eta_0 + (B_2/M_{1\bar{1}})\eta_1(\eta_0 + \eta_c)}{Q(\eta_0 + \eta_c)} \right) \end{aligned} \tag{2.32}$$

where the coefficients A and $B = B_1 + iB_2$ appear in the normal form of the deterministic equations. Their expressions in terms of the characteristics of the chemical model are given in the Appendix. Apart from the Kronecker delta $\delta(\mathbf{m}_0 + \mathbf{m}'_0 + \mathbf{q} + \mathbf{q}')$, the first term in expression (2.32) is independent of the wave vectors \mathbf{m}_0 , \mathbf{m}'_0 , \mathbf{q} , and \mathbf{q}' . Indeed, it is equal to the exactly resonant fourth derivative

$$U^{\mathbf{m}_c\bar{\mathbf{m}}_c010\bar{1}} = \frac{\eta_c A/H_{\mathbf{m}_c\bar{\mathbf{m}}_c} + \eta_0 B_1/M_{1\bar{1}}}{\eta_c + \eta_0} \tag{2.33}$$

The point is that this *interaction* term, which contains explicitly the two different types of critical modes, becomes infinite on the line $\eta_c + \eta_0 = 0$ and has no limit when η_c and η_0 tend to zero. The analogy with our results for the interaction of two Hopf bifurcations⁽¹⁵⁾ is striking. Due to the *codimension-two* character of the bifurcation, the fourth derivative $U^{m_c \bar{m}_c 010\bar{1}}$ is singular unless the following relation between the coefficients of the reaction-diffusion model is satisfied:

$$A/H_{m_c \bar{m}_c} = B_1/M_{1\bar{1}} \quad (2.34)$$

As the second transition moments $H_{m_c \bar{m}_c}$ and $M_{1\bar{1}}$ are positive quantities, condition (2.34) imposes

$$AB_1 > 0 \quad (2.35)$$

In the sequel, condition (2.34) is supposed satisfied, so that the exactly resonant fourth derivative reduces to

$$U^{m_c \bar{m}_c 010\bar{1}} = A/H_{m_c \bar{m}_c} \quad (2.36)$$

and so that the quasis resonant fourth derivatives (2.32) take the simpler form

$$U^{m_0 m'_0 q_1 q'_1} = \delta(m_0 + m'_0 + q_1 + q'_1) \left[\frac{A}{H_{m_c \bar{m}_c}} - \frac{i(-K_q + K_{q'}) (\theta_1 B_1 + \eta_1 B_2)}{QM_{1\bar{1}}} \right] \quad (2.37)$$

where Q is the quasisresonance (2.31).

Nevertheless, it is clear from expression (2.37) that the condition (2.34) does not suffice to define the full set of the fourth derivatives. Indeed, due to the *spatial dependence*, the derivatives (2.37) have no limit as $(-K_q + K_{q'})$ and Q tend to zero unless the condition

$$\theta_1 B_1 + \eta_1 B_2 = 0 \quad (2.38)$$

is satisfied. Moreover, higher order derivatives $U^{(2p)}$ associated with quasisresonances satisfy

$$U^{(2p)} = (\theta_1 B_1 + \eta_1 B_2) O(\text{Re } \omega_{l_0})^{2-p}, \quad p > 2 \quad (2.39)$$

so that they diverge for $\text{Re } \omega_{l_0} \rightarrow 0$ if condition (2.38) is not satisfied. Analogous singularities have been pointed out in Section 2.4 for a Hopf bifurcation with a spatial dependence. If condition (2.38) does not hold, the expansion of U_{cr}^I is only valid in a small neighborhood of the fixed point which does not include the bifurcating attractors. It is thus necessary to impose condition (2.38) to define an expansion of U_{cr}^I , valid in a neighborhood of the fixed point defined by

$$\sigma_{q_1} \lesssim (\text{Re } \omega_{q_1})^{1/2}, \quad \sigma_{m_0} \lesssim (\omega_{m_0})^{1/2}$$

Under condition (2.38), the quasis resonant fourth derivatives (2.37) are simply

$$U^{\mathbf{m}_0 \mathbf{m}'_0 \mathbf{q}^1 \mathbf{q}'^1} = \delta(\mathbf{m}_0 + \mathbf{m}'_0 + \mathbf{q} + \mathbf{q}') \frac{A}{H_{\mathbf{m}_c \bar{\mathbf{m}}_c}} \tag{2.41}$$

so that the expansion of the interaction term U_{cr}^1 reduces finally to

$$U_{cr}^1 = \frac{A}{2H_{\mathbf{m}_c \bar{\mathbf{m}}_c}} \delta(\mathbf{m}_0^1 + \mathbf{m}_0^2 + \mathbf{q}^1 + \mathbf{q}^2) \sigma_{\mathbf{m}_0^1} \sigma_{\mathbf{m}_0^2} \sigma_{\mathbf{q}^1} \sigma_{\mathbf{q}^2} \tag{2.42}$$

2.6. The Critical Potential Associated with the Interaction of a Symmetry-Breaking Bifurcation and a Hopf Bifurcation

We summarize here the results of the preceding subsections.

The first point is that, near the bifurcation, the search for the stochastic potential U may be reduced to the determination of its critical part U_{cr} which depends only on the critical variables.

The second point is that, due to the spatial dependence and to the codimension-two character of the bifurcation, it is in general not possible to define a Taylor expansion of U_{cr} valid in a neighborhood of the fixed point which describes properly the bifurcating attractors.

Nevertheless, we proved that a fourth-order expansion of U_{cr} , valid in a sufficiently large neighborhood of the fixed point defined by

$$\sigma_{\mathbf{q}^1} \lesssim (\text{Re } \omega_{\mathbf{q}^1})^{1/2}, \quad \sigma_{\mathbf{m}_0} \lesssim (\omega_{\mathbf{m}_0})^{1/2} \tag{2.43}$$

exists if and only if the following conditions between the coefficients of the reaction-diffusion model are satisfied:

$$\frac{\eta_1}{\theta_1} = \frac{u_1}{u_2} = -\frac{B_1}{B_2}, \quad \frac{A}{H_{\mathbf{m}_c \bar{\mathbf{m}}_c}} = \frac{B_1}{M_{1\bar{1}}} \tag{2.44}$$

This expansion reduces to

$$\begin{aligned} U_{cr} = & \frac{1}{H_{\mathbf{m}_c \bar{\mathbf{m}}_c}} [-\omega_{\mathbf{m}_0^1} \sigma_{\mathbf{m}_0^1} \sigma_{\mathbf{m}_0^1} + \gamma \delta(\mathbf{m}_0^1 + \mathbf{m}_0^2 + \mathbf{m}_0^3) \sigma_{\mathbf{m}_0^1} \sigma_{\mathbf{m}_0^2} \sigma_{\mathbf{m}_0^3} \\ & + v_1 \delta(\mathbf{m}_0^1 + \mathbf{m}_0^2 + \mathbf{m}_0^3 + \mathbf{m}_0^4) \sigma_{\mathbf{m}_0^1} \sigma_{\mathbf{m}_0^2} \sigma_{\mathbf{m}_0^3} \sigma_{\mathbf{m}_0^4}] \\ & + \frac{1}{M_{1\bar{1}}} [-2 \text{Re } \omega_{\mathbf{q}^1} \sigma_{\mathbf{q}^1} \sigma_{\mathbf{q}^1} \\ & + u_1 \delta(\mathbf{q}^1 + \mathbf{q}^2 + \mathbf{q}^3 + \mathbf{q}^4) \sigma_{\mathbf{q}^1} \sigma_{\mathbf{q}^2} \sigma_{\mathbf{q}^3} \sigma_{\mathbf{q}^4}] \\ & + \frac{A}{2H_{\mathbf{m}_c \bar{\mathbf{m}}_c}} \delta(\mathbf{m}_0^1 + \mathbf{m}_0^2 + \mathbf{q}^1 + \mathbf{q}^2) \sigma_{\mathbf{m}_0^1} \sigma_{\mathbf{m}_0^2} \sigma_{\mathbf{q}^1} \sigma_{\mathbf{q}^2} \end{aligned} \tag{2.45}$$

This explicit potential contains all the information about the system. Using only Eq. (2.45), we shall now deduce both the deterministic and the stochastic properties of a general reaction-diffusion system in the vicinity of the bifurcation of interest.

3. DETERMINATION OF ATTRACTORS

As already pointed out, one of our aims is to deduce the limit sets emerging from the complex bifurcation of interest as extrema of the stochastic potential [cf. Eq. (2.4)]. Actually, we show that the stochastic potential U defined as a solution of the Hamilton–Jacobi equation (2.3) is a generalized Lyapunov function⁽¹¹⁾ of the deterministic flow.

3.1. Extrema of the Stochastic Potential U_{cr}

Since the behavior of the system is entirely described by the critical potential U_{cr} given by Eq. (2.45), we look for the states for which all the first derivatives of U_{cr} vanish,

$$\frac{\partial U_{cr}}{\partial \sigma_{m_0}} = 0, \quad \frac{\partial U_{cr}}{\partial \sigma_{q_1}} = 0, \quad \frac{\partial U_{cr}}{\partial \sigma_{q_1}} = 0, \quad \forall \mathbf{q}, \mathbf{m}_0 \quad (3.1)$$

The system (3.1) has a great number of solutions, but we restrict our analysis to solutions in the following form:

$$\begin{aligned} \sigma_{m_0} &= \sigma_c \delta_{m_0, \mathbf{k}} + \sigma_c^* \delta_{m_0, -\mathbf{k}} \\ \sigma_{q_1} &= \sigma_0 \delta_{q, 0} \\ \sigma_{q_1} &= \sigma_0^* \delta_{q, 0} \end{aligned} \quad (3.2)$$

where the asterisk indicates complex conjugate and where the direction of the vector \mathbf{k} is fixed [its modulus is imposed by the wave vector \mathbf{m}_c solution of system (2.12)].

The type of solution σ_{m_0} we look for limits the study to spatial structures built on a single wave vector \mathbf{k} .

Substituting σ_{m_0} , σ_{q_1} , and σ_{q_1} from Eqs. (3.2) into Eqs. (3.1) yields

$$(-2\eta_c + A|\sigma_0|^2 + 12v_1|\sigma_c|^2)\sigma_c = 0 \quad (3.3)$$

$$(-2\eta_0 + 2u_1|\sigma_0|^2 + B_1|\sigma_c|^2)\sigma_0 = 0 \quad (3.4)$$

where A and B_1 satisfy condition (2.34).

As Eqs. (3.3), (3.4) impose only the moduli of σ_c and σ_0 , we introduce polar coordinates

$$\sigma_c = R_c e^{i\phi_c}, \quad \sigma_0 = R_0 e^{i\phi_0} \quad (3.5)$$

defining the moduli R_c and R_0 and the phases ϕ_c and ϕ_0 . Equations (3.3), (3.4) admit the four solutions gathered in Table I.

The first solution

$$R_c = 0, \quad R_0 = 0 \tag{3.6}$$

corresponds to the initial fixed point.

The second solution

$$R_c^2 = \eta_c / 6v_1, \quad R_0 = 0, \quad \forall \phi_c, \phi_0 \tag{3.7}$$

corresponds to a spatial structure which exists after the bifurcation point if $v_1 > 0$. In the sequel, we suppose that this condition is satisfied. To obtain a better representation of this structure, it is instructive to return to the initial variables $x_{r\alpha} - \bar{x}_\alpha$ through Eq. (2.6):

$$x_{r\alpha} - \bar{x}_\alpha = 2R_c (c_{\mathbf{k}})_\alpha^0 \cos(\mathbf{k}\mathbf{r} + \phi_c)$$

where $(c_{\mathbf{k}})_\alpha^0$ is one of the eigenvector solutions of Eq. (2.8) associated with the eigenvalue $\omega_{\mathbf{k}0} = \eta_c$.

The structure corresponds to a spatial oscillation of the concentrations with an amplitude $2R_c (c_{\mathbf{k}})_\alpha^0$, a pulsation \mathbf{k} , and a phase ϕ_c . As Eqs. (3.7) impose only the modulus of σ_c , the structures obtained when ϕ_c varies are equally probable. A variation $\Delta\phi_c$ of the phase corresponds to a translation of the patterns by $\Delta\mathbf{r}$ such that $\Delta\mathbf{r} \cdot \mathbf{k} = \Delta\phi_c$. This property is a consequence of the translational invariance of the stochastic potential U_{cr}^{SB} given by Eq. (2.23). Hence, Eqs. (3.7) define a set of spatial structures obtained from one another by *translation*.

The third solution

$$R_c = 0, \quad R_0^2 = \eta_0 / u_1, \quad \forall \phi_c, \phi_0 \tag{3.8}$$

Table I. Extrema of the Stochastic Potential U_{cr} ^a

	Fixed point (3.6)	Spatial structure (3.7)	Limit cycle (3.8)	Spatiotemporal structure (3.9)
R_c^2	0	$\frac{\eta_c}{6v_1}$	0	$\frac{2(2u_1\eta_c - A\eta_0)}{24u_1v_1 - AB_1}$
R_0^2	0	0	$\frac{\eta_0}{u_1}$	$\frac{2(12v_1\eta_0 - B_1\eta_c)}{24u_1v_1 - AB_1}$

^aThe analysis is limited to solutions in the form (3.2). Only the moduli R_c and R_0 are imposed; any values of the phases ϕ_c and ϕ_0 agree.

corresponds to a limit cycle, identical to the limit cycle issuing from a simple Hopf bifurcation.⁽¹⁰⁾ It exists after the bifurcation point if $u_1 > 0$, which we shall assume in the following.

The fourth solution

$$R_c^2 = \frac{2(2u_1\eta_c - A\eta_0)}{24u_1v_1 - AB_1}, \quad R_0^2 = \frac{2(-B_1\eta_c + 12v_1\eta_0)}{24u_1v_1 - AB_1} \quad (3.9)$$

corresponds to a spatiotemporal structure characterized by oscillations of the concentrations in both space and time. Before discussing the conditions of existence of this structure, let us give a more evocative picture of it by returning to the initial variables

$$x_{r\alpha} - \bar{x}_\alpha = 2R_c(c_k)_\alpha^0 \cos(\mathbf{k}\mathbf{r} + \phi_c) + 2R_0|(c_0)_\alpha^1| \cos[\arg(c_0)_\alpha^1 + \phi_0]$$

where $(c_0)_\alpha^1$ is a complex eigenvector of M_α' , a solution of Eq. (2.8) with $\omega_{01} = \eta_0 + i\theta_0$.

The states associated with different values of the phase ϕ_0 are reached successively when time is varied, as is shown in the Appendix [see Eq. (A.9) for the justification of $\phi_0 \simeq \theta_0 t$]. Figure 2 represents the temporal oscillations, or better the “breathing” of the spatial structure, which is not distorted in the course of time [see Eq. (A.9) for the justification of $\dot{\phi}_c \simeq 0$].

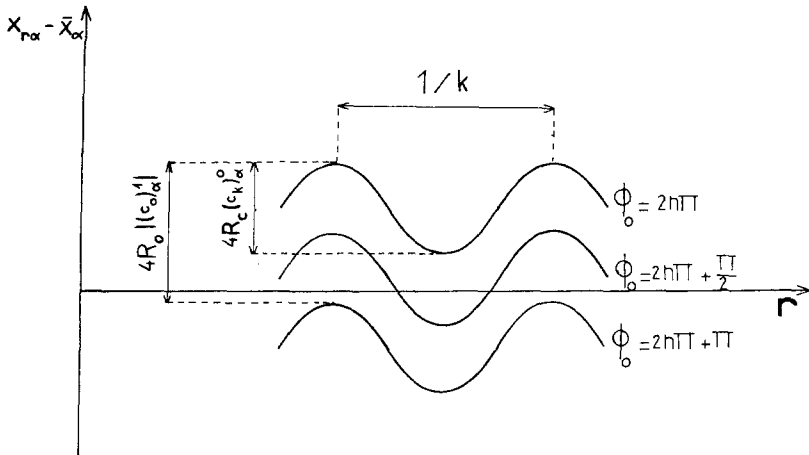


Fig. 2. Amplitude of the spatiotemporal structure (3.9) $\{x_{r\alpha} - \bar{x}_\alpha = 2R_c(c_k)_\alpha^0 \cos(\mathbf{k}\mathbf{r} + \phi_c) + 2R_0|(c_0)_\alpha^1| \cos[\arg(c_0)_\alpha^1 + \phi_0]\}$ for different values of ϕ_0 [$= 2n\pi, \pi/2 + 2n\pi,$ and $(2n + 1)\pi,$ with $n \in \mathbb{N}$]. As is justified in the Appendix, ϕ_0 plays the role of time. When time is varied, the spatial oscillations are not distorted, but the amplitude $x_{r\alpha} - \bar{x}_\alpha$ oscillates between two extreme sinusoids associated with $\phi_0 = 2n\pi$ and $\phi_0 = (2n + 1)\pi$.

Table II. Conditions of Existence of the Spatiotemporal Structure Defined by Eq. (3.9)

	$24u_1v_1 - AB_1 > 0$ $A < 0$ Case a	$24u_1v_1 - AB_1 > 0$ $A > 0$ Case b	$24u_1v_1 - AB_1 < 0$ $A > 0$ Case c
R_c	$\eta_c > \frac{A}{2u_1} \eta_0$	$\eta_c > \frac{A}{2u_1} \eta_0$	$\eta_c < \frac{A}{2u_1} \eta_0$
R_0	$\eta_c > \frac{12v_1}{B_1} \eta_0$	$\eta_c < \frac{12v_1}{B_1} \eta_0$	$\eta_c > \frac{12v_1}{B_1} \eta_0$

We gather in Table II the conditions of existence of R_c and R_0 defined by Eqs. (3.9). Let us recall that the parameters u_1 and v_1 are suppose to be positive and that, according to Eq. (2.38), $AB_1 > 0$. We exclude the case $A < 0$ and $24u_1v_1 - AB_1 < 0$, for which R_c and R_0 never exist after the bifurcation point, as we excluded the cases $u_1 < 0$ and $v_1 < 0$.

3.2. Nature of the Extrema of U_{cr}

From now on, we shall restrict our description of the probability surface to a small neighborhood of any extremum previously obtained [see Eqs. (3.7)–(3.9)]. The natural variables for this description are those for which the local second derivatives of U_{cr} at one arbitrary point of the family of extrema reduce to a diagonal matrix. In particular, the sign of the eigenvalues of this matrix gives the nature of the extrema. The first step is thus to evaluate the quadratic terms $V^{(2)}$ of the potential expanded around an extremum in the form (3.2).

Defining the deviations from an extremum through

$$\begin{aligned}
 \zeta_{\mathbf{m}_0} &= \sigma_{\mathbf{m}_0} - (\sigma_c \delta_{\mathbf{m}_0, \mathbf{k}} + \sigma_c^* \delta_{\mathbf{m}_0, -\mathbf{k}}) \\
 \zeta_{\mathbf{q}1} &= \sigma_{\mathbf{q}1} - \sigma_0 \delta_{\mathbf{q}0} \\
 \zeta_{\mathbf{q}\bar{1}} &= \sigma_{\mathbf{q}\bar{1}} - \sigma_0^* \delta_{\mathbf{q}0}
 \end{aligned}
 \tag{3.10}$$

it is possible to show that $V^{(2)}$ may be split into two parts: the first part, $V_{\mathbf{q}}^{(2)}$, depends only on the variables $\zeta_{\mathbf{q}+\mathbf{k}}$, $\zeta_{\mathbf{q}-\mathbf{k}}$, $\zeta_{\mathbf{q}1}$, $\zeta_{\mathbf{q}\bar{1}}$, and their complex conjugates, whereas the second part, $V_{\mathbf{m}_0}^{(2)}$, contains all other critical variables, denoted by $\zeta_{\mathbf{m}_0}$ with $\mathbf{m}_0 \neq \pm(\mathbf{k} \pm \mathbf{q})$ (let us recall that the direction of the wave vector \mathbf{k} is imposed). From a mathematical point of view, this property of $V^{(2)}$ is a direct consequence of the proportionality of the

coefficients of the critical potential U_{cr} [Eq. (2.45)] to the Kronecker delta. Hence, $V^{(2)}$ may be written as

$$V^{(2)} = \sum_{\mathbf{q}} V_{\mathbf{q}}^{(2)} + \sum_{\mathbf{m}_0 \neq \pm(\mathbf{k} \pm \mathbf{q})} V_{\mathbf{m}_0}^{(2)} \tag{3.11}$$

From a physical point of view, these two different parts of $V^{(2)}$ express the existence of two kinds of eigendirections in the phase space, pointing toward states of clearly different nature. Changing the sign of an eigenvalue of $V_{\mathbf{q}}^{(2)}$ or $V_{\mathbf{m}_0}^{(2)}$ would imply a destabilization of the initial structure in favor of two different types of new structures. In the first case, the new structure would approximatively keep the essential characteristics of the initial one; in particular, the description with the wave vectors $\mathbf{q} = 0$ and \mathbf{k} is still valid. In the second case, the new structure would be built on different wave vectors.

Let us first consider the second term of Eq. (3.11), $V_{\mathbf{m}_0}^{(2)}$, which may be written in the form

$$\begin{aligned} & \sum_{\mathbf{m}_0 \neq \pm(\mathbf{k} \pm \mathbf{q})} V_{\mathbf{m}_0}^{(2)} \\ &= \sum_{\substack{\mathbf{m}_0 \neq \pm(\mathbf{k} \pm \mathbf{q}) \\ \mathbf{k} + \mathbf{m}_0 \neq \mathbf{m}_0}} \frac{1}{H_{\mathbf{m}_c \bar{\mathbf{m}}_c}} \left(-\omega_{\mathbf{m}_0} + 12v_1 R_c^2 + \frac{A}{2} R_0^2 \right) \zeta_{\mathbf{m}_0} \zeta_{\bar{\mathbf{m}}_0} \\ &+ \sum_{\substack{\mathbf{m}_0 \neq \pm(\mathbf{k} \pm \mathbf{q}) \\ \mathbf{k} + \mathbf{m}_0 = \mathbf{m}_0}} \frac{1}{H_{\mathbf{m}_c \bar{\mathbf{m}}_c}} \left[\left(-\omega_{\mathbf{m}_0} + 12v_1 R_c^2 + \frac{A}{2} R_0^2 \right) \zeta_{\mathbf{m}_0} \zeta_{\bar{\mathbf{m}}_0} \right. \\ &\left. + 3\gamma(\sigma_c \zeta_{\mathbf{m}_0} \zeta_{-\mathbf{k} - \mathbf{m}_0} + \sigma_c^* \zeta_{\bar{\mathbf{m}}_0} \zeta_{\mathbf{m}_0 + \mathbf{k}}) \right] \tag{3.12} \end{aligned}$$

The last term in Eq. (3.12), proportional to γ , arises from the cubic terms of the critical potential (2.45). Because of their proportionality to a Kronecker delta, the last term in Eq. (3.12) is only summed on vectors \mathbf{m}_0 such that $-\mathbf{m}_0 - \mathbf{k}$ is critical and such that \mathbf{k} , \mathbf{m}_0 , and $-\mathbf{k} - \mathbf{m}_0$ form a quasi-equilateral triangle (see Fig. 3). The first sum in Eq. (3.12) is already a diagonal Hermitian form, the eigenvalues of which are easily evaluated for R_c and R_0 associated with the various extrema. For cases (3.7), (3.9), the above eigenvalues denoted by $\lambda_{\mathbf{m}_0}$ are positive as soon as the corresponding extrema exist. A condition of stability, different from the conditions of existence, is obtained for case (3.8): taking into account the expression (2.13) of $\omega_{\mathbf{m}_0}$ with $\theta_c < 0$, we find that the eigenvalues $\lambda_{\mathbf{m}_0}$ are positive if $A\eta_0 - 2u_1\eta_c > 0$. Comparing this inequality with the results summarized in Table II, we see that the limit cycle (3.8) is unstable when the spatio-temporal structure (3.9) exists for cases a and b, but is stable for case c.

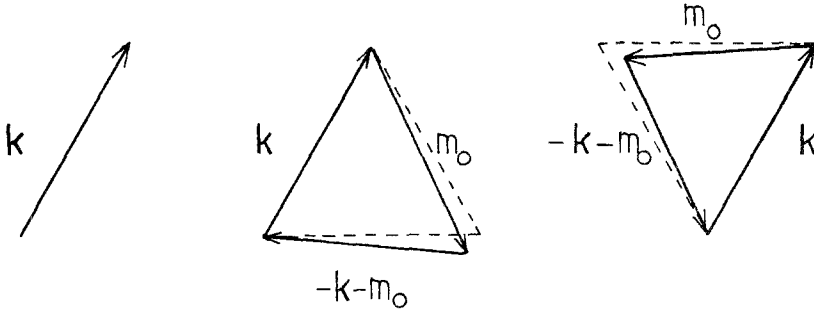


Fig. 3. Illustration of the different possibilities for the vector \mathbf{m}_0 appearing in the last term in Eq. (3.12). The modulus and the direction of the wave vector \mathbf{k} are imposed by the spatial structure of interest, and so \mathbf{m}_0 is such that $-\mathbf{k} - \mathbf{m}_0$ is critical and such that \mathbf{k} , \mathbf{m}_0 , and $-\mathbf{k} - \mathbf{m}_0$ form a quasiequilateral triangle.

The second sum in Eq. (3.12) may be written in a matrix form and diagonalized. Writing the condition of stability for case (3.8) does not bring any new information, but for cases (3.7), (3.9) the eigenvalues are positive if, respectively,

$$\left(\frac{\eta_c v_1}{6}\right)^{1/2} \pm \gamma > 0, \quad v_1 \left(\frac{4u_1 \eta_c - 2A\eta_0}{24u_1 v_1 - AB_1}\right)^{1/2} \pm \gamma > 0 \quad (3.13)$$

According to Eq. (2.24), if $\gamma \sim (\omega_0)^{1/2}$, we can find values of η_c and η_0 for which one of these inequalities is not satisfied. The spatial and spatio-temporal structures built on a single wave vector \mathbf{k} then become unstable and structures built on three or more vectors appear, as mentioned by Walgraef *et al.*⁽⁵⁾ In the sequel, we suppose that γ may be neglected with respect to $(\omega_0)^{1/2}$, so that the inequations (3.13) are always satisfied.

We now consider the first part $\sum_{\mathbf{q}} V_{\mathbf{q}}^{(2)}$ of Eq. (3.11). It may be written in the following matrix form:

$$\sum_{\mathbf{q}} \begin{pmatrix} \zeta_{\mathbf{q}+\mathbf{k}}^* & \zeta_{\mathbf{q}-\mathbf{k}}^* & \zeta_{\mathbf{q}\uparrow}^* & \zeta_{\mathbf{q}\downarrow}^* \end{pmatrix} \begin{pmatrix} E_{\mathbf{q}} & \frac{6v_1 \sigma_c^2}{H_{\mathbf{m}_c \bar{\mathbf{m}}_c}} & \frac{B_1 \sigma_0^* \sigma_c}{2M_{11}} & \frac{B_1 \sigma_0 \sigma_c}{2M_{11}} \\ \frac{6v_1 \sigma_c^{*2}}{H_{\mathbf{m}_c \bar{\mathbf{m}}_c}} & E_{-\mathbf{q}} & \frac{B_1 \sigma_0^* \sigma_c^*}{2M_{11}} & \frac{B_1 \sigma_0 \sigma_c^*}{2M_{11}} \\ \frac{A\sigma_0 \sigma_c^*}{2H_{\mathbf{m}_c \bar{\mathbf{m}}_c}} & \frac{A\sigma_0 \sigma_c}{2H_{\mathbf{m}_c \bar{\mathbf{m}}_c}} & F_{\mathbf{q}} & \frac{u_1 \sigma_0^2}{M_{11}} \\ \frac{A\sigma_0^* \sigma_c^*}{2H_{\mathbf{m}_c \bar{\mathbf{m}}_c}} & \frac{A\sigma_0^* \sigma_c}{2H_{\mathbf{m}_c \bar{\mathbf{m}}_c}} & \frac{u_1 \sigma_0^{*2}}{M_{11}} & F_{-\mathbf{q}} \end{pmatrix} \begin{pmatrix} \zeta_{\mathbf{q}+\mathbf{k}} \\ \zeta_{\mathbf{q}-\mathbf{k}} \\ \zeta_{\mathbf{q}\uparrow} \\ \zeta_{\mathbf{q}\downarrow} \end{pmatrix} \quad (3.14)$$

Table III. Eigenvalues of $V_a^{(2)}$ Associated with the Matrix (3.14) Evaluated for Each Extremum of the Stochastic Potential^a

Fixed point (3.6)	Spatial structure (3.7)	Limit cycle (3.8)	Spatiotemporal structure (3.9)
$\frac{-\eta_c - \theta_c K_+}{H_{m_c, m_c}}$	$\lambda_{q0} = \frac{-\theta_c}{2H_{m_c, m_c}} (K_+ + K_-)$	$\lambda'_{q0} = \frac{-\eta_1 K_q}{M_{11}}$	$\lambda_{q0} = \frac{-\theta_c}{2H_{m_c, m_c}} (K_+ + K_-)$
$\frac{-\eta_c - \theta_c K_-}{H_{m_c, m_c}}$	$\lambda_{q1} = \frac{4\eta_c - \theta_c (K_+ + K_-)}{2H_{m_c, m_c}}$	$\lambda'_{q1} = \frac{2\eta_0 - \eta_1 K_q}{M_{11}}$	$\lambda'_{q0} = \frac{-\eta_1 K_q}{M_{11}}$
$\frac{-\eta_0 - \eta_1 K_q}{M_{11}}$	$\lambda_{q2} = \frac{-12v_1(\eta_0 + \eta_1 K_q) + B_1 \eta_c}{12v_1 M_{11}}$	$\lambda'_{q2} = \frac{-2u_1(\eta_c + \theta_c K_+) + A\eta_0}{2u_1 H_{m_c, m_c}}$	$\mu_{q1} = \frac{6v_1 R_c^2}{H_{m_c, m_c}} + \frac{u_1 R_0^2}{M_{11}} + \left(\frac{u_1 R_0^2}{M_{11}} - \frac{6v_1 R_c^2}{H_{m_c, m_c}} \right)^{1/2}$ $\times \left(1 + \frac{AB_1 R_0^2 R_c^2}{H_{m_c, m_c} u_1 R_0^2 - M_{11} 6v_1 R_c^2} \right)^{1/2}$
$\frac{-\eta_0 - \eta_1 K_q}{M_{11}}$	$\lambda_{q3} = \frac{-12v_1(\eta_0 + \eta_1 K_q) + B_1 \eta_c}{12v_1 M_{11}}$	$\lambda'_{q3} = \frac{-2u_1(\eta_c + \theta_c K_-) + A\eta_0}{2u_1 H_{m_c, m_c}}$	$\mu'_{q1} = \frac{6v_1 R_c^2}{H_{m_c, m_c}} + \frac{u_1 R_0^2}{M_{11}} - \left(\frac{u_1 R_0^2}{M_{11}} - \frac{6v_1 R_c^2}{H_{m_c, m_c}} \right)^{1/2}$ $\times \left(1 + \frac{AB_1 R_0^2 R_c^2}{H_{m_c, m_c} u_1 R_0^2 - M_{11} 6v_1 R_c^2} \right)^{1/2}$
			$-\frac{\theta_c}{H_{m_c, m_c}} \frac{G^2}{2} (K_+ + K_-) - \frac{\eta_1}{M_{11}} (1 - G^2) K_q$
			$-\frac{\theta_c}{H_{m_c, m_c}} \frac{1 - G^2}{2} (K_+ + K_-) - \frac{\eta_1}{M_{11}} G^2 K_q$

^a R_0 and R_c are given by (3.9). We have adopted the following notation: $K_+ = (K_{q+k} - K_c)^2$, $K_- = K_{q-k} - K_c^2$, and

$$G^2 = \left\{ \frac{u_1 R_0^2}{M_{11}} - \frac{6v_1 R_c^2}{H_{m_c, m_c}} + \left[\left(\frac{u_1 R_0^2}{M_{11}} - \frac{6v_1 R_c^2}{H_{m_c, m_c}} \right)^2 + \frac{AB_1 R_0^2 R_c^2}{H_{m_c, m_c} M_{11}} \right]^{1/2} \right\}^2 + \frac{AB_1 R_0^2 R_c^2}{H_{m_c, m_c} M_{11}}$$

where

$$\begin{aligned}
 E_{\mathbf{q}} &= \frac{1}{H_{m_c \bar{m}_c}} \left(-\omega_{\mathbf{q}+\mathbf{k}} + \frac{A}{2} |\sigma_0|^2 + 12v_1 |\sigma_c|^2 \right) \\
 F_{\mathbf{q}} &= \frac{1}{M_{11}} \left(-\text{Re } \omega_{\mathbf{q}1} + 2u_1 |\sigma_0|^2 + \frac{B_1}{2} |\sigma_c|^2 \right)
 \end{aligned}
 \tag{3.15}$$

and where σ_0 and σ_c satisfy system (3.4). Table III gives the eigenvalues of matrix (3.14) evaluated for each extremum of the stochastic potential U_{cr} given by Table I.

Note that the spatial structure (3.7) is characterized by a vanishing eigenvalue $\lambda_{\mathbf{q}=00} = 0$ associated with an eigenvector tangent to the set of spatial structures obtained from one another by translation. Similarly, the limit cycle (3.8) is characterized by a vanishing eigenvalue $\lambda'_{\mathbf{q}=00} = 0$ associated with an eigenvector tangent to the limit cycle. The spatiotemporal structure admits two vanishing eigenvalues $\lambda_{\mathbf{q}=00} = 0$ and $\lambda'_{\mathbf{q}=00} = 0$ in the two directions defined previously.

Determining the sign of the eigenvalues of $V_{\mathbf{q}}^{(2)}$ whatever \mathbf{q} , we deduce immediately from Table III the conditions under which the extrema (3.6)–(3.9) of U_{cr} are minima. The conditions of stability of structures (3.6)–(3.9), or, equivalently, the domains of existence of the corresponding attractors, are summarized in Table IV.

As expected, the conditions of stability of the fixed point (3.6) deduced from $V_{\mathbf{q}}^{(2)}$ are totally in agreement with the deterministic conditions deduced from the linear stability operator H'_j associated with the eigenvalues (2.13) and (2.14).

Note that the conditions of stability of the limit cycle (3.8) imposed by $V_{\mathbf{m}_0}^{(2)}$ defined by Eq. (3.12) are identical with the conditions deduced from the analysis of $V_{\mathbf{q}}^{(2)}$ defined by Eq. (3.14).

Comparing Table II and IV, we conclude, for cases a and b, that the spatial structure (3.7) and the limit cycle (3.8) are unstable as soon as the

Table IV. Conditions of Stability of Structures (3.6)–(3.9)

	Fixed point (3.6)	Spatial structure (3.7)	Limit cycle (3.8)	Spatiotemporal structure (3.9)
Domains of parameter space	$\eta_c < 0$ $\eta_0 < 0$	$\eta_c > 0$ $B_1 \eta_c - 12v_1 \eta_0 > 0$	$\eta_0 > 0$ $-2u_1 \eta_c + A \eta_0 > 0$	$24u_1 v_1 - AB_1 > 0^a$

^a If R_0 and R_c given by (3.9) exist.

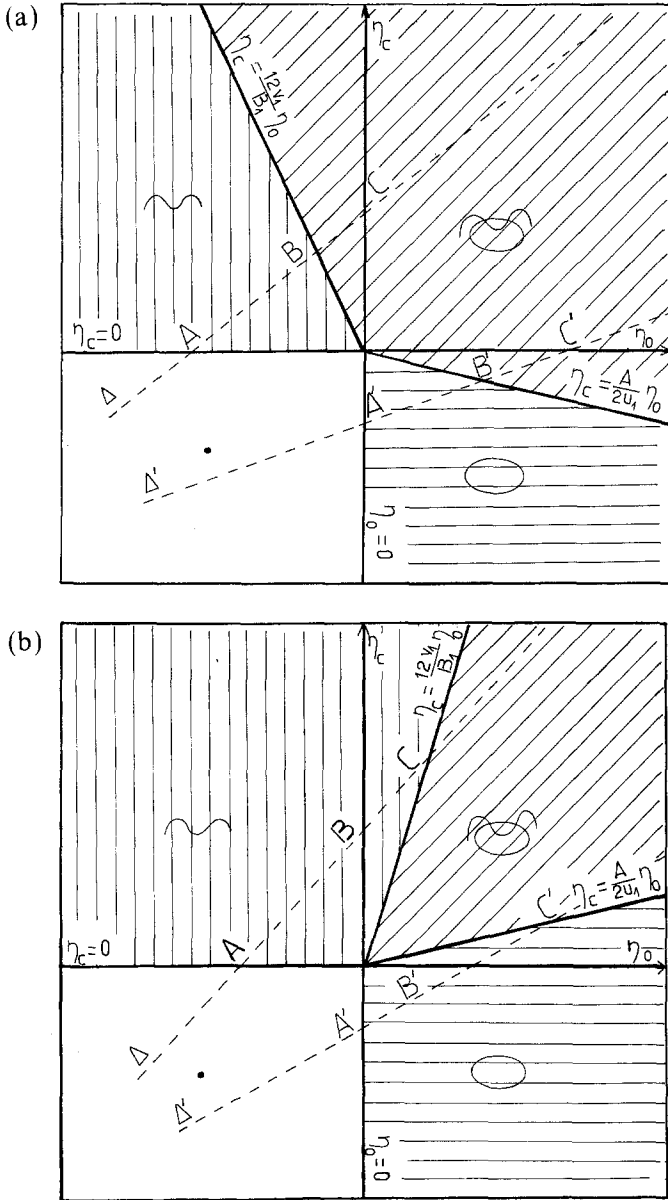


Fig. 4. (a) Domain of stability of the different attractors in the parameter space (η_0, η_c) in case a ($A < 0, 24u_1v_1 - AB_1 > 0$). Open sector, lower left: domain of stability of the initial fixed point (dot). Horizontal lines mark the domain of stability of the limit cycle (oval). Vertical lines mark the domain of stability of the spatial structure (wiggly line). Diagonal lines mark the domain of stability of the spatiotemporal structure (wiggly line plus oval). (b) Domain of stability of the different structures in case b ($A > 0, 24u_1v_1 - AB_1 > 0$). Same key as for part (a). (c) Domain of stability of the different structures in case c ($A > 0, 24u_1v_1 - AB_1 < 0$). Same key as for part (a), apart from the domain of coexistence of the limit cycle and the spatial structure.

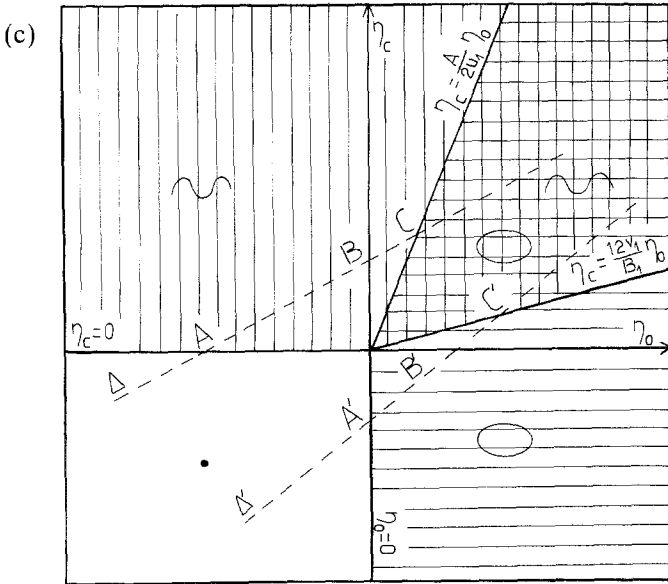


Fig. 4 (continued)

spatiotemporal structure (3.9) is stable. Conversely, for case c, the spatiotemporal structure is never stable, but there exists a domain of coexistence of the spatial structure (3.7) and of the limit cycle (3.8). These results are illustrated by Fig. 4.

The domain of existence of a spatiotemporal structure (cases a and b) or the domain of coexistence of a spatial structure and a limit cycle (case c) may be reached by two qualitatively different ways. These two possibilities are illustrated by sections of the bifurcation diagrams along either a line Δ crossing the domain of stability of the spatial structure (3.7) (see Figs. 4 and 5) or a line Δ' crossing the domain of stability of the limit cycle (3.8) (see Figs. 4 and 6). It is clear from Figs. 5 and 6 that, on the boundary between the domains of stability of the spatial structure (3.7) and the spatiotemporal structure (3.9), the amplitude of the spatial component R_c does not undergo any discontinuity. The same property is observed on the boundary between the domains of stability of the limit cycle (3.8) and the spatiotemporal structure (3.9) for the amplitude of the temporal component R_0 . Note the continuity between the eigenvalues of $V_q^{(2)}$ given by Table III on these boundaries, too.

It is interesting to compare cases a and b: this brings out the effect on the bifurcation diagrams of the sign of A which appears through Eq. (2.42) as the "interaction coefficient" of the stochastic potential. If $A < 0$ (case a),

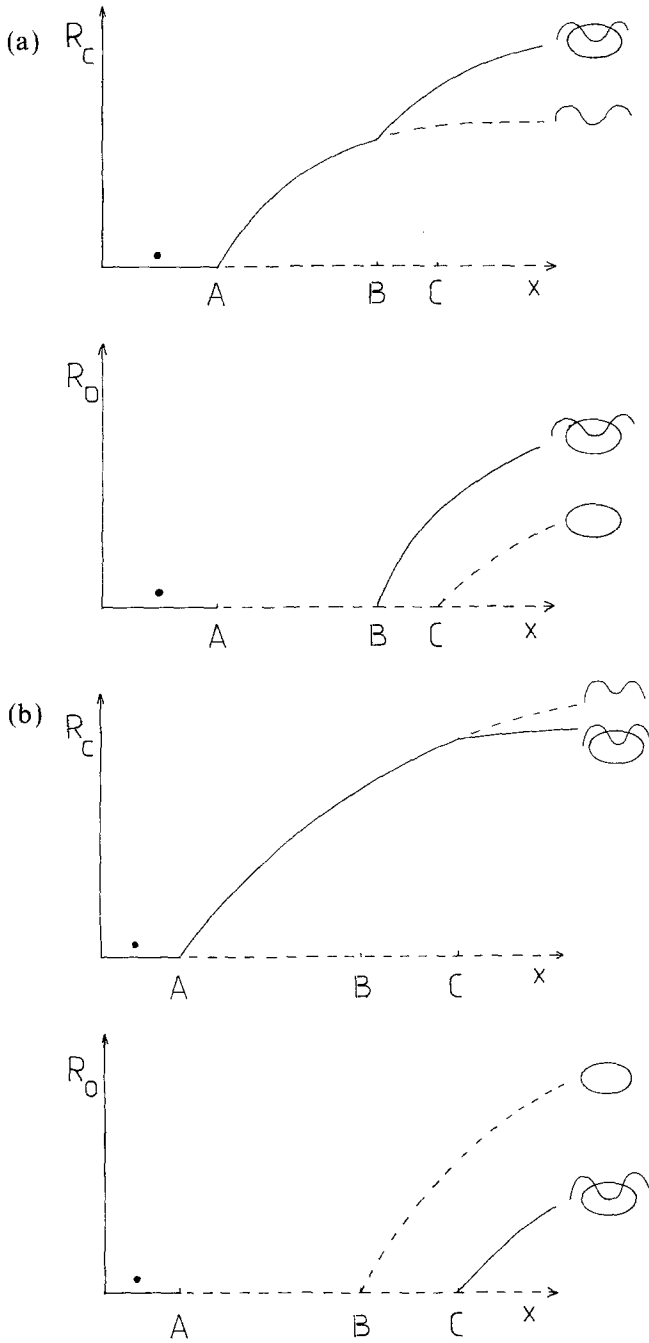


Fig. 5. (a) Section of the bifurcation diagram along the line \mathcal{A} in cases a-c of Fig. 4. initial fixed point; oval, limit cycle; wiggly line, spatial structure; wiggly line plus oval, spatiotemporal structure; (—) stable and (---) unstable structures.

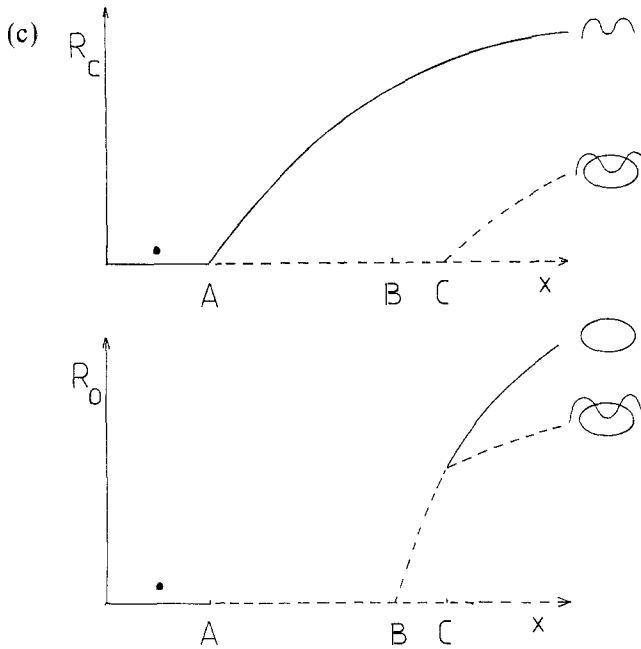


Fig. 5 (continued)

the domain of stability of the spatiotemporal structure (see Fig. 4a) is always greater than the quadrant defined by $\eta_c > 0$, $\eta_0 > 0$ and it increases with $|A|$ up to a half-plane. Moreover, the amplitudes R_c and R_0 of the spatiotemporal structure (3.9) are larger than the amplitude R_c of the spatial structure (3.7) and the amplitude R_0 of the limit cycle (3.8) (see Figs. 5a and 6a). Conversely, if $A > 0$ (case b), the domain of stability of the spatiotemporal structure (see Fig. 4b) is smaller than the quadrant defined by $\eta_c > 0$, $\eta_0 > 0$ and it decreases with $|A|$. The amplitudes R_c and R_0 of the spatiotemporal structure (3.9) are smaller than the amplitudes R_c and R_0 given, respectively, by (3.7) and (3.8) (see Figs. 5b and 6b).

For case c, the domain of existence of structures (3.7) and (3.8) increases with $|A|$ but is never larger than the quadrant defined by $\eta_c > 0$, $\eta_0 > 0$.

3.3. Comparison with Deterministic Results

We give in the Appendix the normal forms of the deterministic equations describing the evolution of the concentrations. We immediately verify that the extrema of the stochastic potential U_{cr} given by Table I are

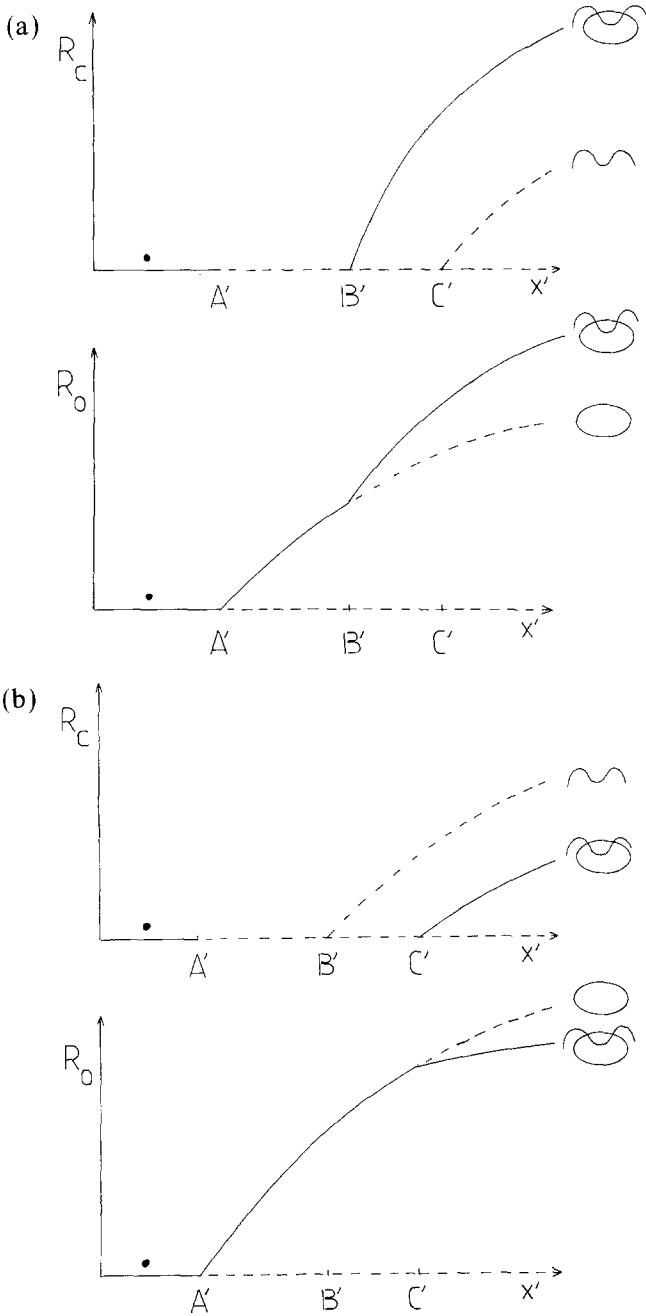


Fig. 6. Section of the bifurcation diagram along the line Δ' in cases a-c of Fig. 4.

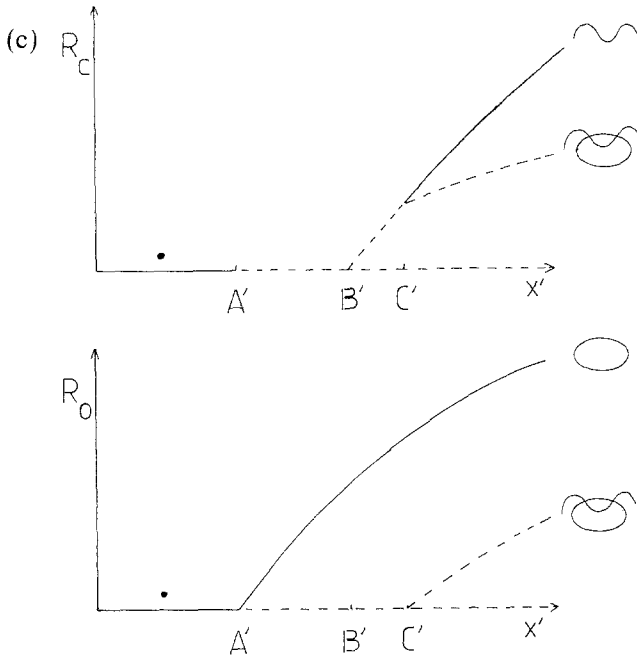


Fig. 6 (continued)

identical with the deterministic limit sets. Note that the expansion (2.45) of U_{cr} is only valid if conditions (2.44) between its coefficients are satisfied. Although these constraints may seem drastic, they actually correspond to a generic situation. Indeed, any small deviation from the imposed conditions does not modify the topological structure of the bifurcation diagram. Thus, our stochastic analysis gives a satisfying description of the systems associated with the same topological types. Nevertheless, conditions (2.44) preclude some interesting behaviors predicted by the deterministic analysis.

In particular, it is known that the limit cycle (3.8) may be destabilized through inhomogeneous perturbations that may lead to phase turbulence.⁽¹⁶⁾ According to deterministic results,⁽¹⁶⁾ this may happen if the coefficient $\eta_1 u_1 + \theta_1 u_2$ is positive. Because of condition (2.26), namely $\eta_1 u_2 - \theta_1 u_1 = 0$, this coefficient reduces here to $(\eta_1/u_1)(u_1^2 + u_2^2)$, which is always negative. Hence, an expansion around the fixed point of the stochastic potential does not describe the inhomogeneous destabilization of a limit cycle. In a less ambitious approach, it is possible to describe this bifurcation provided that the probability is limited to a small neighborhood of the limit cycle which does not include the fixed point.

Another type of bifurcation is ruled out because of condition (2.34),

which imposes $AB_1 > 0$. Indeed, if the signs of A and B_1 were different, it would be possible to observe a Hopf bifurcation in the space of moduli (R_c, R_0). Following then the deterministic analysis of Guckenheimer and Holmes,⁽⁴⁾ we know that the next step in the bifurcation cascade consists in a bifurcation from the quasiperiodic attractor to a homoclinic orbit. However, such a complex behavior is not described by a normal form truncated to its cubic terms. It is thus hardly surprising that such a complex situation is not described by a smooth probability surface.

Beyond the comparison with deterministic results, our aim is to take advantage of the specific information given by the stochastic potential. In particular, this quantity describes the fluctuations of the concentrations around their deterministic values. In order to establish whether a structure associated with a given attractor is observable at a macroscopic level, it is crucial to determine how these fluctuations of concentrations at different points of the system are correlated. Indeed, the spatial correlations between two distant points measures the ability of the system to maintain a coherent behavior at long range.

4. SPATIAL CORRELATIONS OF THE FLUCTUATIONS AROUND THE DIFFERENT ATTRACTORS

One of the results of the previous sections is the existence, in the spectrum of the quadratic form $V^{(2)}$ around a point of an attractor (3.7)–(3.9), of one or two vanishing eigenvalues $\lambda_{\mathbf{q}=00}$ or $\lambda'_{\mathbf{q}=00}$ (see Table III). These vanishing eigenvalues reflect the fact that the attractor is not a point of the phase space but a one- or two-dimensional manifold. The corresponding eigenvectors are tangent to this manifold. In addition, for a large system, the eigenvectors associated with $\lambda_{\mathbf{q}0}$ or $\lambda'_{\mathbf{q}0}$, with nonzero but small \mathbf{q} , define directions of the phase space along which the probability does not remain rigorously constant but is very slowly varying compared to the other eigendirections. For given η_0 and η_c all the eigenvalues $\lambda_{\mathbf{q}0}$ and $\lambda'_{\mathbf{q}0}$ have orders of magnitude much smaller than the others, provided that \mathbf{q} obeys the following inequalities (see Table III):

$$\begin{aligned} \theta_c(K_{\mathbf{q}+\mathbf{k}} - K_c)^2 &\ll \eta_c \\ \theta_c(K_{\mathbf{q}-\mathbf{k}} - K_c)^2 &\ll \eta_c \\ \eta_1 K_{\mathbf{q}} &\ll \eta_0 \end{aligned} \quad (4.1)$$

Among all the states in the neighborhood of our arbitrary reference point on the attractor, the states which have nonnegligible components only in the eigendirections associated with $\lambda_{\mathbf{q}0}$ or $\lambda'_{\mathbf{q}0}$ [with \mathbf{q} obeying (4.1)] have a probability very close to its maximum value. For these particular states, the

Taylor expansion of the stochastic potential has to include terms of order higher than two, or, in other words, fluctuations may reach very large values in these eigendirections. This is why the correlation of these fluctuations plays a crucial role in sustaining or destroying the attracting structures predicted by the deterministic theory.

4.1. Local Potential around an Attractor

By virtue of the previous analysis, the local description of the stochastic potential around an arbitrary point of an attractor begins with the diagonalization of the quadratic form $V_{\mathbf{q}}^{(2)}$. The new variables associated with the various eigenvalues (given for each attractor by Table III) will be globally denoted by $\{y\}$. For example, $y_{\mathbf{q}0}$ is associated with the small eigenvalue $\lambda_{\mathbf{q}0}$, whereas $y_{\mathbf{q}\phi}$ denotes globally the eigendirections associated with nonsmall eigenvalues. The cubic and quartic terms of the expansion are then computed only for the states having nonnegligible $y_{\mathbf{q}0}$ (and/or $y'_{\mathbf{q}0}$) and small components in other eigendirections. To separate the various terms in terms of critical and noncritical contributions, we use arguments of the same type as used by Lemarchand and Nicolis^(6,7) in the case of the fourth-order expansion U^{SB} around a fixed point [see Eq. (2.20)]. The results are given for each attractor in Table V.

The point is that the expansion of the potential reduces here to a sum of quadratic terms in the variables y themselves and in some particular nonlinear combinations of them, $y_{\mathbf{q}\phi} - g(\{y_{\mathbf{q}0}\})$. This was not the case in the fourth-order form (2.20), in which cubic and quartic terms remained even after the nonlinear change of variables (2.22). This different behavior may be interpreted as follows: though the separation of terms in the two types of fourth-order expansions of U is based on the same idea, namely the existence of small eigenvalues in the matrix of the quadratic part, the difference comes from the distinct nature of the vanishing eigenvalues. In the first case [described by Eq. (2.20)], the vanishing eigenvalue originated from the bifurcation of the fixed point. In the present case, a zero eigenvalue is a consequence of the nonzero dimension of the attractor. In other words, it originates from the invariance of U when one moves along this attractor, i.e., on the manifold $\{y_{0\phi} = g(\{y_{00}\}); y_{\mathbf{q}\neq 0\phi} = 0\}$. Clearly, this invariance property would not be preserved if other nonquadratic terms in $y_{\mathbf{q}0}$ would have remained. As a result, the probability of the states of components $\{y_{\mathbf{q}0}\}$, whatever $\langle y_{\mathbf{q}\phi} \rangle$, is properly given by the Gaussian approximation.²

² The Gaussian approximation fails only if another eigenvalue $\lambda_{\mathbf{q}0}$ with $\mathbf{q} \neq 0$ vanishes. Analyses of this new type of bifurcation would need to compute higher order terms in $K_{\mathbf{q}}$ or K_{+} and K_{-} in the expansion of $\lambda_{\mathbf{q}0}$ or $\lambda'_{\mathbf{q}0}$ (see Table III). This is the aim of the phase diffusion analysis.⁽¹⁶⁾

Table V. The Stochastic Potential V_q around an Attractor^a

	V_q
Spatial structure (3.7)	$\sum_q \lambda_{q0} y_{q0} ^2 + \lambda_{q1} \left y_{q1} - \sum_{q'} \frac{1}{2} \left(\frac{3v_1}{\eta_c} \right)^{1/2} y_{q0} y_{q-q0} \right ^2$ $+ \lambda_{q2} y_{q2} ^2 + \lambda_{q3} y_{q3} ^2$
Limit cycle (3.8)	$\sum_q \lambda'_{q0} y'_{q0} ^2 + \lambda'_{q1} \left y'_{q1} - \sum_{q'} \frac{1}{2} \left(\frac{u_1}{2\eta_0} \right)^{1/2} y'_{q0} y'_{q-q0} \right ^2$ $+ \lambda'_{q2} y'_{q2} ^2 + \lambda'_{q3} y'_{q3} ^2$
Spatiotemporal structure (3.9)	$\sum_q \lambda_{q0} y_{q0} ^2 + \lambda'_{q0} y'_{q0} ^2$ $+ \mu_{q1} \left y_{q1} - \sum_{q'} \frac{2u_1 GR_0 + B_1(1-G^2)^{1/2} R_c}{2\sqrt{2} M_{1\bar{1}} \mu_{q1}} y_{q0} y_{q-q0} \right.$ $\left. - \sum_{q'} \frac{AGR_0 + 12v_1(1-G^2)^{1/2} R_c}{2\sqrt{2} H_{m,\bar{m}} \mu_{q1}} y'_{q0} y'_{q-q0} \right ^2$ $+ \mu'_{q1} \left y'_{q1} - \sum_{q'} \frac{-2u_1(1-G^2)^{1/2} R_0 + B_1 GR_c}{2\sqrt{2} M_{1\bar{1}} \mu'_{q1}} y_{q0} y_{q-q0} \right.$ $\left. - \sum_{q'} \frac{-A(1-G^2)^{1/2} R_0 + 12v_1 GR_c}{2\sqrt{2} H_{m,\bar{m}} \mu'_{q1}} y'_{q0} y'_{q-q0} \right ^2$

^a The eigenvalues of $V_q^{(2)}$ are given by Table III. R_c and R_0 are given by Eqs. (3.9) and G is given in Table III.

The large fluctuations of these components are simply expressed by the covariances:

$$\langle y_{q0} y_{q'0} \rangle = -\frac{\delta(\mathbf{q} + \mathbf{q}')}{2nN\lambda_{q0}}, \quad \langle y_{q0} y'_{q'0} \rangle = 0 \tag{4.2}$$

$$\langle y'_{q0} y'_{q'0} \rangle = -\frac{\delta(\mathbf{q} + \mathbf{q}')}{2nN\lambda'_{q0}}$$

4.2. Evaluation of the Spatial Correlation Function

All the essential deterministic and stochastic characteristics of the new temporal and/or spatial structures can be described in the space of the complex variables $\{\sigma_{q1}\}$, $\{\sigma_{q+k}\}$, where \mathbf{q} runs in a large set of closely packed small values. Switching to space-dependent variables, we define the following complex order parameters

$$\sigma_r^H = \sum_q \sigma_{q1} e^{-i\mathbf{q}r}, \quad \sigma_r^{SB} = \sum_q \sigma_{q+k} e^{-i\mathbf{q}r} \tag{4.3}$$

With these definitions, any point on the new attractor (3.2) is given by homogeneous values of the order parameters:

$$\sigma_r^H = R_0 e^{i\phi_0}, \quad \sigma_r^{SB} = R_c e^{i\phi_c} \tag{4.4}$$

We know from the previous analyses⁽¹⁰⁾ that fluctuations will play an important role for some particular variables upon which the stochastic potential (2.45) does not depend. In order to bring out the role of this kind of variable, we use a polar representation of the order parameters and set

$$\sigma_r^H = R_r \exp(i\phi_r), \quad \sigma_r^{SB} = \rho_r \exp(ik \cdot I_r), \tag{4.5}$$

It is easily verified that any homogeneous translation of the “phase” variables $\{\phi_r\}$ and $\{\mathbf{k}I_r\}$ has no effect on the stochastic potential U . This is why fluctuations of these variables play a dominant role. Indeed, the explicit calculation of the variables $\{y\}$ in which the local quadratic form $V_q^{(2)}$ is diagonal proves that the Fourier transforms of the phase variables are approximately proportional to the “most fluctuating” variables y_{q0} and y'_{q0} . We obtain explicitly⁽¹⁰⁾

$$y_{q0} = \sqrt{2} \frac{i}{n} \rho_{r1} \sin(\mathbf{k}I_{r1} - \phi_c) \exp(ir^1 \mathbf{q}) \tag{4.6}$$

$$y'_{q0} = \sqrt{2} \frac{i}{n} R_{r1} \sin(\phi_{r1} - \phi_0) \exp(ir^1 \mathbf{q})$$

In the vicinity of the arbitrary point chosen on the attractor ($\forall \mathbf{r}, \rho_r = R_c, \mathbf{k}I_r = \phi_c, R_r = R_0, \phi_r = \phi_0$), these expressions reduce to

$$y_{q0} = \sqrt{2} \frac{i}{n} R_c (\mathbf{k}I_{r1} - \phi_c) \exp(ir^1 \mathbf{q}) \tag{4.7}$$

$$y'_{q0} = \sqrt{2} \frac{i}{n} R_0 (\phi_{r1} - \phi_0) \exp(ir^1 \mathbf{q})$$

As a result, the variables $\{\mathbf{k}I_r - \phi_c\}$ and $\{\phi_r - \phi_0\}$ as their linear combinations $\{y_{q0}\}$ and $\{y'_{q0}\}$ have a Gaussian distribution which can be written

$$P(\{y_{q0}\}, \{y'_{q0}\}) = Z \exp\left(-nN \sum_{\mathbf{q}} \lambda_{q0} |y_{q0}|^2\right) \times Z' \exp\left(-nN \sum_{\mathbf{q}} \lambda'_{q0} |y'_{q0}|^2\right) \tag{4.8}$$

where Z and Z' are normalization constants.

Substituting for y_{q_0} and y'_{q_0} from Eq. (4.7) into Eq. (4.8), we obtain the probability of the "phase variables" $\phi_r, \mathbf{k}I_r$

$$P(\{\mathbf{k}I_r\}, \{\phi_r\}) = P_I P_\phi \quad (4.9)$$

with

$$P_\phi = Z' \exp \left[-nN \sum_q \lambda'_{q_0} \frac{2}{n^2} R_0^2 |(\phi_{r^1} - \phi_0) \exp(ir^1 \mathbf{q})|^2 \right] \quad (4.10)$$

$$P_I = Z \exp \left[-nN \sum_q \lambda_{q_0} \frac{2}{n^2} R_c^2 |(\mathbf{k}I_{r^1} - \phi_c) \exp(ir^1 \mathbf{q})|^2 \right] \quad (4.11)$$

Using the expressions for λ_{q_0} and λ'_{q_0} given in Table III, assuming a continuous description of the space dimensionless variable \mathbf{r} , and performing the summation over \mathbf{q} yields

$$P_\phi = Z' \exp \left[-N \frac{2(-\eta_1)}{M_{11}} R_0^2 \int (\nabla \phi_r)^2 d^d \mathbf{r} \right] \quad (4.12)$$

$$P_I = Z \exp \left\{ -N \frac{2(-\theta_c)}{H_{m, \bar{m}_c}} R_c^2 \int \mathbf{k}I_r [4(\mathbf{k} \cdot i \nabla_r)^2 + (\nabla_r \cdot \nabla_r)^2] \mathbf{k}I_r d^d \mathbf{r} \right\} \quad (4.13)$$

Thus, the two kinds of phase variables $\{\phi_r\}$ and $\{\mathbf{k}I_r\}$ have no cross-correlations and each of them has a Gaussian distribution whose correlations may be determined analytically. This Gaussian distribution remains invariant by any homogeneous translation of the phase variable. The same type of probability has already been derived in a similar way⁽¹⁰⁾ or through a Langevin method⁽⁹⁾ for a simple Hopf bifurcation. Our technique proves to be equally suitable for a symmetry-breaking bifurcation and even for all the attractors issuing from the degenerate situation described in the present work.

Now, to express the spatial correlation of the order parameters σ_r^H and σ_r^{SB} around the new spatial and/or temporal structures, we have to compute the following covariances

$$\begin{aligned} |\langle \sigma_r^H \cdot \sigma_r^{H*} \rangle| &= \langle R_r R_r \exp[i(\phi_r - \phi_r)] \rangle \\ &\simeq R_0^2 |\langle \exp[i(\phi_r - \phi_r)] \rangle| \\ |\langle \sigma_r^{SB} \cdot \sigma_r^{SB*} \rangle| &= \langle \rho_r \rho_r \exp[i(\mathbf{k} \cdot I_r - \mathbf{k} \cdot I_r)] \rangle \\ &\simeq R_c^2 |\langle \exp[i(\mathbf{k} \cdot I_r - \mathbf{k} \cdot I_r)] \rangle| \\ |\langle \sigma_r^H \cdot \sigma_r^{SB*} \rangle| &= \langle R_r \rho_r \exp[i(\phi_r - \mathbf{k} \cdot I_r)] \rangle \\ &\simeq R_0 R_c |\langle \exp[i(\phi_r - \mathbf{k} \cdot I_r)] \rangle| \end{aligned} \quad (4.14)$$

where we have neglected the fluctuations of R_r and ρ_r around R_0 and R_c (compared to the “most fluctuating” variables ϕ_r and $\mathbf{k}I_r$). Mean values of the form $\langle \exp[i(X_r - X_r')] \rangle$ can be considered as the values of the generating function $G(\{v_r\}) = \langle e^{v \cdot X} \rangle$ for $v_r = i, v_r = -i, v_{r' \neq r, r'} = 0$. For a Gaussian distribution of $\{X_r\}$, this generating function has a simple form, which leads to the following results (see, e.g., ref. 18):

$$\begin{aligned} |\langle \exp[i(X_r - X_r')] \rangle| &= \exp[-\langle (X_r - X_r')^2 \rangle / 2] \\ |\langle \exp(iX_r) \rangle| &= \exp[-\langle (X_r)^2 \rangle / 2] \end{aligned} \tag{4.15}$$

This allows us to express the covariances (4.14) in terms of the Gaussian covariances (4.12), (4.13) of $\{\phi_r\}$ and $\{\mathbf{k} \cdot I_r\}$. The latter are computed by coming back to the Fourier transform variables. We obtain in a d -dimensional space

$$\begin{aligned} F_d(\mathbf{r}, \mathbf{r}')^H &= |\langle \sigma_r^H \cdot \sigma_{r'}^H \rangle| \\ &= R_0^2 \exp\left(-\frac{M_{11}}{N(-4\eta_1)R_0^2} \int \frac{1 - \cos \mathbf{q}(\mathbf{r} - \mathbf{r}')}{q^2} \frac{d^d \mathbf{q}}{(2\pi)^d}\right) \\ F_d(\mathbf{r}, \mathbf{r}')^{SB} &= |\langle \sigma_r^{SB} \cdot \sigma_{r'}^{SB} \rangle| \\ &= R_c^2 \exp\left(-\frac{H_{m, \bar{m}_c}}{N(-4\theta_c)R_c^2} \int \frac{1 - \cos \mathbf{q}(\mathbf{r} - \mathbf{r}')}{4(\mathbf{k} \cdot \mathbf{q})^2 + (\mathbf{q} \cdot \mathbf{q})^2} \frac{d^d \mathbf{q}}{(2\pi)^d}\right) \\ |\langle \sigma_r^H \cdot \sigma_{r'}^{SB} \rangle| &= |\langle \sigma_r^H \rangle| |\langle \sigma_{r'}^{SB} \rangle| = 0 \end{aligned} \tag{4.16}$$

The integrals in Eqs. (4.16) depend on the dimensionality d of the system. The domain of integration in the \mathbf{q} space has to be limited to the domain of validity of our local expansion.

On one hand, we must impose

$$\theta_c(K_{\mathbf{q} \pm \mathbf{k}} - K_c)^2 < \eta_c, \quad \eta_1 K_{\mathbf{q}} < \eta_0 \tag{4.17}$$

We will denote by q_M the maximum value of the modulus $|\mathbf{q}|$ which satisfies (4.17). The quantity $v = 2\pi/q_M$ is the smallest wavelength corresponding to the most inhomogeneous mode incorporated in our description.

On the other hand, the modulus $|\mathbf{q}|$ is limited to a minimum value $q_m = 2\pi/n_1$ (n_1 is the number of cells in one direction) corresponding to the first nonhomogeneous mode appearing in the probability distribution (4.10), (4.11).

For all values of $|\mathbf{r} - \mathbf{r}'|$ such that $|\mathbf{r} - \mathbf{r}'| q_m$ remains small while $|\mathbf{r} - \mathbf{r}'| q_M$ remains large, that is, for

$$v \ll |\mathbf{r} - \mathbf{r}'| \ll n_1 \tag{4.18}$$

Table VI. The Spatial Correlation Function $F_d(\mathbf{r}, \mathbf{r}')^H$ for a System of Dimensionality d ($d \in \{1, 2, 3\}$) and for^a

$$v = 2\pi/q_M \ll |\mathbf{r} - \mathbf{r}'| \ll n_1 = 2\pi/q_m$$

$F_d(\mathbf{r}, \mathbf{r}')^H$	
$d = 1$	$R_0^2 \exp \left[-\frac{1}{N} \frac{M_{11}}{4(-\eta_1)R_0^2} \frac{ \mathbf{r} - \mathbf{r}' }{2} (1 + \varepsilon_1^M + \varepsilon_1^m) \right]$
$d = 2$	$R_0^2 \exp \left[-\frac{1}{N} \frac{M_{11}}{4(-\eta_1)R_0^2} \frac{1}{2\pi} \left(\ln \frac{ \mathbf{r} - \mathbf{r}' }{2} q_M + C + \varepsilon_2^M + \varepsilon_2^m \right) \right]$
$d = 3$	$R_0^2 \exp \left[-\frac{1}{N} \frac{M_{11}}{4(-\eta_1)R_0^2} \frac{1}{2\pi^2} \frac{1}{ \mathbf{r} - \mathbf{r}' } \left(\mathbf{r} - \mathbf{r}' q_M - \frac{\pi}{2} + \varepsilon_3^M + \varepsilon_3^m \right) \right]$

^a The results are valid for the three types of structures characterized by the values (3.7), (3.8), or (3.9) of R_0 , ε_d^M and ε_d^m tend to zero with $|\mathbf{r} - \mathbf{r}'|^{-1} q_M^{-1}$ and $|\mathbf{r} - \mathbf{r}'| q_m$, respectively. C is Euler's constant.

Table VII. The Spatial Correlation Function $F_d(\mathbf{r}, \mathbf{r}')^{SB}$ for a System of Dimensionality d ($d \in \{1, 2, 3\}$) and for^a

$$v = 2\pi/q_M \ll |\mathbf{r} - \mathbf{r}'| \ll n_1 = 2\pi/q_m$$

$F_d(\mathbf{r}, \mathbf{r}')^{SB}$	
$d = 1$	$F_1(\mathbf{r}, \mathbf{r}')^{SB} \simeq R_c^2 \exp \left[-\frac{1}{N} \frac{H_{m_c \bar{m}_c}}{(-4\theta_c)R_c^2} \frac{1}{4k^2} \frac{ \mathbf{r} - \mathbf{r}' }{2} (1 + \varepsilon_1^M + \varepsilon_1^m) \right]$
$d = 2$	$F_2(\mathbf{r}, \mathbf{r}')^{SB} < R_c^2 \exp \left[-\frac{1}{N} \frac{H_{m_c \bar{m}_c}}{(-4\theta_c)R_c^2} \frac{1}{4k^2} \frac{1}{2\pi} \left(\ln \frac{ \mathbf{r} - \mathbf{r}' }{2} q_M + C + \varepsilon_2^M + \varepsilon_2^m \right) \right]$
$d = 3$	$F_3(\mathbf{r}, \mathbf{r}')^{SB} = R_c^2 \exp \left\{ -\frac{1}{N} \frac{H_{m_c \bar{m}_c}}{(-4\theta_c)R_c^2} \frac{1}{2k} \frac{1}{8\pi} [\ln 2 \mathbf{r} - \mathbf{r}' \mathbf{k} + C + \varepsilon_3^M + \varepsilon_3^m + E_1(2 \mathbf{r} - \mathbf{r}' \mathbf{k})] \right\}$ $(\mathbf{r} - \mathbf{r}') // \mathbf{k}$

^a The results are valid for the three types of structures characterized by the values (3.7), (3.8), or (3.9) of R_c . ε_d^M and ε_d^m tend to zero with $|\mathbf{r} - \mathbf{r}'|^{-1} q_M^{-1}$ and $|\mathbf{r} - \mathbf{r}'| q_m$, respectively. C is Euler's constant. $E_1(2|\mathbf{r} - \mathbf{r}'| |\mathbf{k}|)$ is the exponential integral,⁽¹⁹⁾ which tends to zero for large arguments.

the integrals in Eqs. (4.16) exhibit simple $|\mathbf{r} - \mathbf{r}'|$ dependences. Tables VI and VII give the spatial dependence of the correlation functions for $d \in \{1, 2, 3\}$. According as R_0 and R_c take the values (3.7), (3.8), or (3.9), these results give the spatial correlations of the three types of previously described structures.

We thus conclude that, in *one-dimensional* systems, the spatial dispersion of the "phase variables" $\langle (\phi_{\mathbf{r}} - \phi_{\mathbf{r}'})^2 \rangle$ or $\langle (\mathbf{k}I_{\mathbf{r}} - \mathbf{k}I_{\mathbf{r}'})^2 \rangle$ increases *linearly* with the distance $|\mathbf{r} - \mathbf{r}'|$. The uncorrelated values of the phase at two distant points smear the structure and compromise its observability.

This phenomenon becomes smoother but remains in *two-dimensional* systems, where the phase dispersion increases as the *logarithm* of the distance. The same result (logarithmic dependence) is observed for $\langle (\mathbf{k}I_{\mathbf{r}} - \mathbf{k}I_{\mathbf{r}'})^2 \rangle$ even in *three-dimensional* systems. In particular, the pure spatial structure (3.7) (built on a single wave vector) is subject to this spatial dispersion of the phase and is expected to be smeared in large *three-dimensional* systems.

The case of homogeneous temporal oscillations [attractor (3.8)] in *three-dimensional* systems is the only one for which the correlation function does not decrease rapidly with the distance $|\mathbf{r} - \mathbf{r}'|$. Actually, the spatial dispersion $\langle (\phi_{\mathbf{r}} - \phi_{\mathbf{r}'})^2 \rangle$ is, in this case, quasi-independent of $|\mathbf{r} - \mathbf{r}'|$. According to this result, a temporal oscillation is expected to be observable in a macroscopic system of dimensionality three.

5. CONCLUSION

We have given a first approach to highly degenerate codimension-two bifurcations, where the large spatial extension of the system is responsible for the quasi-infinite dimension of the center manifold. The normal form of the deterministic system reduced to these critical variables has been derived, as well as the expression for the bifurcating attractors, including spatiotemporal structures which appear through secondary bifurcations specific to the degenerate codimension-two bifurcation under consideration. From the stochastic point of view, we have shown that near criticality the stochastic potential can be split into the sum of a quadratic term with respect to noncritical variables (Gaussian distribution) with a minimum depending on the critical variables as seen in Eqs. (2.20), (2.22), and a more complex term U_{cr} depending only on the critical variables. The noncritical distribution width is distinguishably narrower than that of the critical distribution: this corresponds to the deterministic idea of rapid decay of the noncritical (fast) modes, which then follow adiabatically the critical (slow) modes. This result can be understood as a stochastic equivalent of the center manifold reduction.

The *critical potential* U_{cr} has been shown to be *nonanalytical* in the general case. More specifically, we have shown that the p -order derivatives $U^{(p)}$ of the potential ($p \geq 4$) at the fixed point with respect to a *resonant* set of critical variables containing at least two distinct critical modes are *singular* at the bifurcation point unless three specific conditions are imposed on the kinetic and diffusion constants of the system. This situation is characteristic of degenerate bifurcations of a fixed point, which can be of two types. The first type consists of the bifurcations of codimension greater than one for a uniform system, e.g., the coalescence of two Hopf bifurcations.⁽¹⁵⁾ The second type of degeneracy arises for a system of large spatial extension including diffusion processes, e.g., a Hopf bifurcation in a distributed system. The complex bifurcation under consideration presents both types of degeneracy.

Under the three above-mentioned conditions, we have shown [see Eq. (2.45)] that the critical potential reduces at dominant order near criticality to a fourth-order polynomial with coefficients depending only on the real part of the deterministic normal form coefficients [Eq. (A.4)] and on two stochastic coefficients also associated with the fundamental resonances of the system at criticality. A fundamental consequence is that, due to intrinsic symmetry properties of the bifurcation under consideration and to the periodic boundary conditions, the reduced quartic potential presents both phase and translational invariance. We wish to emphasize the fact that this "normal form" of the critical potential and its symmetry properties are independent of the particular choice of local coordinates around the fixed point: in particular, a Poincaré normal form representation of the system in which the deterministic flow itself presents the above-mentioned invariance properties is not a prerequisite.

As shown in Section 3, the quartic critical potential contains all the information concerning the deterministic system near criticality, the attractors and their bifurcations following from the discussion of the extrema of U_{cr} . Moreover, it can be easily shown that U_{cr} provides a Lyapunov function for the attractors of the deterministic system, since it can only decrease along deterministic trajectories until it reaches an attractor and remains constant.

Beyond the comparison with deterministic results, our essential aim is to bring out the additional information given by the stochastic approach. In this respect, the main point is that due to internal fluctuations the existence of a given attractor does not necessarily imply that the corresponding structure is macroscopically observable. When the normal form of a dynamical system near a bifurcation point is invariant with respect to a "cyclic" variable ϕ , then bifurcating attractors consist of continuous families of asymptotic states obtained by varying the initial value

of ϕ . Examples can be given here of phase invariance in the case of temporal oscillations (limit cycle), translational invariance in the case of spatial structures, and both phase and translational invariance in the case of spatiotemporal structures. In a system of large size, the macroscopic observation of such a structure depends on the ability of the system to impose the same value of ϕ at any point of the space. This property depends essentially on the *range of the spatial correlations of the fluctuations* around the attractor. These correlation functions are derived from the quartic critical potential and it is shown that no long-range order can be sustained in low-dimensional systems undergoing such a degenerate bifurcation. More specifically, we show that limit cycles and spatiotemporal structures are destroyed by inhomogeneous fluctuations for large systems with $d = 1, 2$ and that spatial and spatiotemporal structures built on a single wave vector cannot be observed in a large three-dimensional system.

The main motivation for our stochastic local expansion method around the reference point is to draw as far as possible a parallel with the deterministic normal form approach to local bifurcations, which is based on the reduction of the flow to a finite jet around the fixed point. According to deterministic analysis, different topological types of bifurcation diagrams can be defined near criticality, depending on the signs of a finite number of coefficients derived from the normal form of the flow (in the present case, $A, B_1, u_1, v_1, 24u_1v_1 - AB_1, \eta_1u_1 + \theta_1u_1, \dots$). Because of the conditions of validity of the quartic potential, some of these topological types are excluded from our stochastic local analysis, corresponding, not so surprisingly, to the most complex behaviors which can lead to chaos. However, we wish to emphasize that although the constraints under which our results are derived may seem to be very drastic, they actually correspond to a *generic* situation in the sense that any small deviation from the imposed conditions does not modify the topological structure of the bifurcation diagram. Thus, we can expect our stochastic analysis to give a general description of the whole class of systems corresponding to the same topological types.

Nevertheless, our final aim is of course to explore all the possible topological types of the system. In the general case when the local expansion of U_{cr} around the fixed point breaks down near criticality, we still expect that a unique, normalizable, everywhere two-times differentiable stationary solution of the Hamilton–Jacobi equation exists. An alternative approach, based on straightforward quadratic local expansions of this solution *about the bifurcating attractors*, should enable us to explore secondary bifurcations by which the limit cycle is destabilized through inhomogeneous fluctuations and tertiary bifurcations of the spatiotemporal structures leading to biperiodic spatiotemporal structures. For a system of

large spatial extension, both deterministic fluctuations and random fluctuations of the *phase variables* have been shown to play a predominant role in the possible destabilization of the limit cycle. For this reason, it seems natural to seek a systematic reduction procedure, extending the ideas of phase dynamics⁽¹⁶⁾ to our stochastic approach; work in this direction is in progress.

APPENDIX

We give here the normal form of the deterministic equations describing the evolution of the concentrations in a reaction-diffusion system of large size near the codimension-two bifurcation of interest (interaction of a Hopf bifurcation and a symmetry-breaking bifurcation).

Considering the deterministic equations [see Eq. (2.4)]

$$\frac{dx_j}{dt} = \dot{x}_j = H_j(\{x_j\}) \quad (\text{A.1})$$

we switch to the representation σ_l , defined by Eq. (2.6), in which the linear stability operator H_j^l at the reference state \bar{x}_α is diagonal. We obtain the following system of ordinary differential equations:

$$\dot{\sigma}_l = \omega_l \sigma_l + \frac{1}{2!} H_l^{l^1 l^2} \sigma_{l^1} \sigma_{l^2} + \frac{1}{3!} H_l^{l^1 l^2 l^3} \sigma_{l^1} \sigma_{l^2} \sigma_{l^3} + \dots \quad (\text{A.2})$$

Generalizing to quairesonance (2.15) the theory of normal forms,⁽¹⁾ we carry out a nonlinear change of variables

$$\sigma_l = \xi_l + \frac{1}{2!} P_l^{l^1 l^2} \xi_{l^1} \xi_{l^2} + \frac{1}{3!} Q_l^{l^1 l^2 l^3} \xi_{l^1} \xi_{l^2} \xi_{l^3} \quad (\text{A.3})$$

(where $P_l^{l^1 l^2}$ and $Q_l^{l^1 l^2 l^3}$ are functions of the characteristics of the reaction-diffusion model) in order to eliminate in Eq. (A.2) as many nonlinearities as possible. The equations of evolution of the critical variables ξ_{l_0} associated with small real part eigenvalues ω_{l_0} may be decoupled from the evolution of noncritical variables and reduce to

$$\begin{aligned} \dot{\xi}_{\mathbf{m}_0 0} = & \omega_{\mathbf{m}_0 0} \xi_{\mathbf{m}_0 0} - \frac{1}{2} \gamma \delta(\bar{\mathbf{m}}_0 + \mathbf{m}_0^1 + \mathbf{m}_0^2) \xi_{\mathbf{m}_0^1} \xi_{\mathbf{m}_0^2} \\ & - 2v_1 \delta(\bar{\mathbf{m}}_0 + \mathbf{m}_0^1 + \mathbf{m}_0^2 + \mathbf{m}_0^3) \xi_{\mathbf{m}_0^1} \xi_{\mathbf{m}_0^2} \xi_{\mathbf{m}_0^3} \\ & - \frac{1}{2} A \delta(\bar{\mathbf{m}}_0 + \mathbf{m}_0^1 + \mathbf{q}^1 + \mathbf{q}^2) \xi_{\mathbf{m}_0^1} \xi_{\mathbf{q}^1} \xi_{\mathbf{q}^2} \end{aligned}$$

$$\begin{aligned}
 \dot{\xi}_{\mathbf{q}1} &= \omega_{\mathbf{q}1} \xi_{\mathbf{q}1} - (u_1 + iu_2) \delta(\bar{\mathbf{q}} + \mathbf{q}^1 + \mathbf{q}^2 + \mathbf{q}^3) \xi_{\mathbf{q}1} \xi_{\mathbf{q}2} \xi_{\mathbf{q}3} \\
 &\quad - \frac{1}{4} B \delta(\bar{\mathbf{q}} + \mathbf{q}^1 + \mathbf{m}_0^1 + \mathbf{m}_0^2) \xi_{\mathbf{q}1} \xi_{\mathbf{m}_0^1} \xi_{\mathbf{m}_0^2} \\
 \dot{\xi}_{\mathbf{q}\bar{1}} &= \omega_{\mathbf{q}\bar{1}} \xi_{\mathbf{q}\bar{1}} - (u_1 - iu_2) \delta(\bar{\mathbf{q}} + \mathbf{q}^1 + \mathbf{q}^2 + \mathbf{q}^3) \xi_{\mathbf{q}\bar{1}} \xi_{\mathbf{q}2} \xi_{\mathbf{q}3} \\
 &\quad - \frac{1}{4} B^* \delta(\bar{\mathbf{q}} + \mathbf{q}^1 + \mathbf{m}_0^1 + \mathbf{m}_0^2) \xi_{\mathbf{q}\bar{1}} \xi_{\mathbf{m}_0^1} \xi_{\mathbf{m}_0^2}
 \end{aligned} \tag{A.4}$$

where every coefficient depends on the characteristics of the reaction-diffusion model.

Note that the fourth-order expansion of the stochastic potential (2.45) depends on the real parts of the coefficients of the normal form. In particular, the stochastic potential U_{cr}^{SB} [see Eq. (2.23)] associated with a symmetry-breaking bifurcation^(6,7) depends on γ and v_1 . The u_1 appears in the expansion (2.30) of the stochastic potential U_{cr}^H associated with a Hopf bifurcation.⁽¹⁰⁾ The interaction term U_{cr}^I [see Eq. (2.42)] depends on A .

Let us give here the expressions of A and B as functions of the characteristics of the chemical model:

$$\begin{aligned}
 A &= -2 \left[\frac{M_0^{01} M_1^{11} - M_0^{01} M_1^{11}}{i\theta_0} - \frac{M_0^{1\phi} M_\phi^{10}}{\omega_{\mathbf{m}_c\phi} + i\theta_0} - \frac{M_0^{1\phi} M_\phi^{10}}{\omega_{\mathbf{m}_c\phi} - i\theta_0} + M_0^{110} \right] \\
 B &= -2 \left[\frac{-M_1^{00} M_1^{11} + M_1^{00} M_1^{11} + 2M_1^{00} M_0^{01}}{i\theta_0} - \frac{2M_1^{0\phi} M_\phi^{01}}{\omega_{\mathbf{m}_c\phi} - i\theta_0} + M_1^{100} \right]
 \end{aligned} \tag{A.5}$$

where $M_\beta^{\beta'\beta''}$ and $M_\beta^{\beta'\beta''\beta'''}$ are derivatives of the first transition moments of the chemical process. The two possible values of β are denoted by 0 and ϕ when they refer to the variables $\sigma_{\mathbf{m}_0\beta}$ or by 1 and $\bar{1}$ when they refer to $\sigma_{\mathbf{q}\beta}$. For example, we have

$$M_\phi^{01} = (c_{\mathbf{m}=\mathbf{m}_c}^{-1})_{\beta=\phi}^{\alpha^1} M_{\alpha^1}^{\alpha^2\alpha^3} (c_{\mathbf{m}=\mathbf{m}_c})_{\alpha^2=0}^{\beta'} (c_{\mathbf{q}=\mathbf{0}})^{\beta''} = \bar{1} \tag{A.6}$$

where $(c_{\mathbf{m}}^{-1})_\beta^\alpha$ is the inverse matrix of $(c_{\mathbf{m}})_{\alpha}^\beta$ defined by Eq. (2.8).

We now look for solutions of Eqs. (A.4) in the form

$$\begin{aligned}
 \xi_{\mathbf{m}_0\mathbf{k}} &= R_c e^{i\phi_c} \delta_{\mathbf{m}_0,\mathbf{k}} + R_c e^{-i\phi_c} \delta_{\mathbf{m}_0,-\mathbf{k}} \\
 \xi_{\mathbf{q}1} &= R_0 e^{i\phi_0} \delta_{\mathbf{q}0}, \quad \xi_{\mathbf{q}\bar{1}} = R_0 e^{-i\phi_0} \delta_{\mathbf{q}0}
 \end{aligned} \tag{A.7}$$

The radial and phase variables then satisfy

$$\dot{R}_c = \eta_c R_c - 6v_1 R_c^3 - \frac{1}{2} A R_c R_0^2, \quad \dot{R}_0 = \eta_0 R_0 - u_1 R_0^3 - \frac{1}{2} B_1 R_0 R_c^2 \tag{A.8}$$

$$\dot{\phi}_c = o(\eta_0^2, \eta_c^2), \quad \dot{\phi}_0 = \theta_0 - u_2 R_0^2 - \frac{1}{2} B_2 R_c^2 \tag{A.9}$$

The equations of evolution of the radii are decoupled from the evolution of the phases. Note that Eqs. (A.9) state that, at dominant order, ϕ_c is con-

stant and ϕ_0 is proportional to time t . System (A.8) admits four stationary solutions. The extrema of the stochastic potential U_{cr} given by Table I coincide with these stationary solutions.

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