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Stochastic Analysis of a Hopf Bifurcation: Master Equation Approach

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The effect of inhomogeneous fluctuations in a reaction-diffusion system exhibiting a Hopf bifurcation is analyzed using the master equation approach. A Taylor expansion of the logarithm of the stationary probability, known as the stochastic potential, is calculated. This procedure displays marked analogies with the theory of normal forms. The critical potential, reduced to its local expansion around an arbitrary point of the limit cycle, brings out the essential role played by the phase of the oscillating variables. A comparison with the Langevin analysis of Walgraef *et al.* [*J. Chem. Phys.* **78**(6):3043 (1983)] is performed.

KEY WORDS: Multivariate master equation; fluctuations; Hopf bifurcation; limit cycle; correlations.

1. INTRODUCTION

The analysis of nonlinear nonequilibrium systems in the vicinity of bifurcation points of new branches of solutions has attracted a great deal of attention in the last years. At the level of deterministic analysis, considerable progress has been achieved in the study of the various kinds of attractors using such tools as perturbation expansions or the theory of normal forms.(1,2) At the level of stochastic analysis, the master equation has been used to analyze bifurcations to steady states without symmetry breaking,(3–5) to limit cycles in a zero-dimensional description,(6–8) and to steady states involving space symmetry breaking.(9,10) On the other hand the techniques of critical dynamics, based on the addition of random Langevin forces in the phenomenological equations, has proved very fruitful to sort out qualitative results on the behaviour of the fluctuations near

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the principal bifurcations,(11,12) including the effect of inhomogeneous fluctuations near the onset of a limit cycle.(13)

Our principal goal in the present work is to analyze the effect of inhomogeneous fluctuations in a reaction-diffusion system exhibiting a Hopf bifurcation, using the master equation approach. In contrast to the Langevin approach, we incorporate from the very beginning information concerning the elementary processes going on in the system. We propose a general procedure allowing one to determine the system's attractors, which turns out to display marked analogies with the theory of normal forms.(2) In addition to normal form analysis however, our method takes into account the fluctuations around the attractor. Previous analysis(4,12) have shown that fluctuations can indeed play an important role near bifurcation points, especially in low-dimensional systems.

The procedure is based on the idea that the attractors of a deterministic dynamical system, including those representing *nonstationary* states like limit cycles, correspond to the extrema of the *stationary* solution of the underlying master equation. This simplifies considerably the analysis.

Following previous work by Lemarchand and Nicolis,(10,14) we write the master equation for a general reaction-diffusion system and derive a Hamilton–Jacobi type of equation for the logarithm of the stationary probability U, which can be regarded as *a stochastic potential.*

The Taylor expansion of U around an extremum is then expressed in the representation provided by the eigenfunctions of the linear stability operator. General expressions of the successive derivatives up to the fourth, calculated at the homogeneous steady state are obtained independently of the nature of the bifurcation.

In Section 2, we apply this general expression of U to the case of a Hopf bifurcation, and estimate the orders of magnitude of the different terms. This leads to a separation of U into two parts: a fourth-order polynomial U_{cr}, relative to the critical modes, and a second order one, associated with a Gaussian probability distribution for the noncritical modes. We compare the coefficients of U_{cr} calculated in the case of the Brusselator with the results found by Walgraef *et al.*(12)

The expression of the critical potential U_{cr} brings out quite naturally the essential role played by the phase of the oscillating variables. The equation of the homogeneous limit cycle is first determined by looking for the states of the phase space for which the gradient of the stochastic potential vanishes. The expression of U_{cr} is then reduced to its local expansion around an arbitrary point of the limit cycle (Section 3).

The matrix of the quadratic form in this expansion displays one vanishing eigenvalue associated with an eigenvector pointing in a direction tangent to the limit cycle. In a system of large spatial extension, there is in fact a large number of such eigenvalues accumulating to zero. This leads us to define two kinds of variables, according to whether they are associated with vanishing or nonvanishing eigenvalues. They correspond, in the deterministic analysis, to the slow and fast modes connected, respectively, to the phase and amplitude of the oscillating variables. In the representation diagonalizing the quadratic form, the local expansion of U_{cr} can then be expressed as a sum of two terms, which amounts to factorizing the probability in two functions of the two kinds of variables. The fluctuations of the fast variables have a Gaussian distribution around a state linked to the slow modes by a relation expressing an adiabatic elimination process. The important result is that the probability function of the slow variables also reduces to a Gaussian distribution, which permits the calculation of the spatial correlation of the phase fluctuations. This result, previously derived by Walgraef *et al.*(13) through a Langevin-equation approach, in the case of the Brusselator model, implies the destruction of long-range order through phase fluctuations in low-dimensional systems.

2. THE STOCHASTIC POTENTIAL NEAR A HOPF BIFURCATION

The reaction-diffusion model consists of a set of chemically active constituents in a volume $\mathscr{V}$ in a d-dimensional space. This space is divided into n submacroscopic cells. The numbers of cells along each axis are denoted by $n_1, n_2, ..., n_d$ such that $n_1 \times n_2 \times \cdots \times n_d = n$. A vector $r = (r_1, r_2, ..., r_d)$ with integer components locates a given cell.

The number of particles of species α in cell r will be denoted $X_{r\alpha}$. The kinetic characteristics of the chemical reactions are the following:

$\bar{\nu}_{\rho\alpha}$, order of the ρth reaction with respect to X_α;

$\nu_{\rho\alpha}$, stoichiometric coefficient of X_α in the ρth reaction ($\nu_{\rho\alpha} > 0$ for particles formed as a result of the reaction and $\nu_{\rho\alpha} < 0$ for particles disappearing as a result of the reaction);

k_ρ, kinetic constant of the ρth reaction expressed in $(\text{sec})^{-1}(\text{liter})^{\Sigma_\alpha \bar{\nu}_{\rho\alpha} - 1}$ units.

These constants may include externally controlled concentrations which can be regarded as bifurcation parameters. 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 $\mathscr{D}_\alpha$ through

$$D_\alpha (\Delta l)^2 \sim \mathscr{D}_\alpha$$

The usual stochastic description(10,14,15) of the chemical reactions as birth and death processes and of diffusion as random walk between adjacent cells leads to the multivariate master equation for the probability distribution $P(\{X_{r\alpha}\})$

$$\begin{aligned}\frac{dP}{dt} = {} & \sum_{\rho} k_{\rho}(\Delta\mathscr{V})^{1-\Sigma_{\alpha}\bar{\nu}_{\rho\alpha}} \\ & \times \sum_{r} \left[\left(\prod_{\alpha} \frac{(X_{r\alpha}-\nu_{\rho\alpha})!}{(X_{r\alpha}-\bar{\nu}_{\rho\alpha}-\nu_{\rho\alpha})!} \right) P(\{X_{r\alpha}-\nu_{\rho\alpha}\}) - \left(\prod_{\alpha} \frac{X_{r\alpha}!}{(X_{r\alpha}-\bar{\nu}_{\rho\alpha})!} \right) P \right] \\ & + \sum_{\alpha} D_{\alpha} \sum_{ra} ((X_{r\alpha}+1)\, P(X_{r\alpha}+1, X_{(r+a)\alpha}-1) - X_{r\alpha} P) \qquad (2.1)\end{aligned}$$

$\Delta\mathscr{V}$ is the volume of one cell, and a denotes the first neighbors of cell r. Only the values of the arguments of P which differ from $\{X_{r\alpha}\}$ are explicitly indicated.

Denoting by $\mathscr{N}$ the total number of particles in the system, one defines the stochastic potential $U(\{x_{r\alpha}\}, t)$ by

$$P(\{X_{r\alpha}\}, t) = e^{-S-\mathscr{N}U(\{x_{r\alpha}\},t)}$$

where S stands for the normalization factor; $U(\{x_{r\alpha}\}, t)$ is considered a continuous function of the reduced variables

$$x_{r\alpha} = \frac{X_{r\alpha}}{N}$$

where $N = \mathscr{N}/n$ is the mean number of particles in a cell.

Substituting this expression of P in the master equation and expanding in terms of $1/N$, one obtains, to zeroth order, a Hamilton–Jacobi type of equation$^{(16,17)}$

$$-\frac{1}{\mathscr{N}}\frac{dS}{dt} - \frac{\partial U}{\partial t} = H\left(\{x_j\}, \left\{ \frac{\partial U}{\partial x_j} \right\} \right) \qquad (2.2)$$

where we have used the contracted notation $(r\alpha) = j$ and the "Hamiltonian" H has an explicit form in terms of the kinetic characteristics of reactions and diffusion.

The starting point of the analysis is to seek for solutions of Eq. (2.2) in the form of a local expansion around an extremum of the stochastic potential, or more generally a point $\{\bar{x}_j(t)\}$, defined by the vanishing of the first derivatives of U. We refer to Refs. 10 and 14 for the detailed calculation of the successive derivatives of U. Specifically, one obtains from the equation of the first derivatives of U at $\{\bar{x}_j(t)\}$ a condition on $\bar{x}_j(t)$ which reflects the vanishing of all these derivatives at any time. This condition is merely the deterministic equation

$$\frac{d\bar{x}_j(t)}{dt} = H_j$$

where the kinetic polynomial H_j appears as the derivative of the Hamiltonian H with respect to $\partial U/\partial x_j$, evaluated at the reference state: $\{x_j = \bar{x}_j(t),\ \partial U/\partial x_j = 0\}$. Equivalently, H_j can be viewed as the first transition moment of the master equation.

Our aim is to exhibit the role of the fluctuations around the deterministic trajectories, when the externally controlled parameters are chosen in a neighborhood of a bifurcation point. The stability properties of these trajectories depend on the eigenvalues ω_l of the linear stability operator $H_j^{j'}$ evaluated at the deterministic state $\{\bar{x}_j(t)\}$. They obey the following equation

$$\sum_{j'} H_j^{j'} C_{j'}^l = \omega_l C_j^l$$

where C^l is the eigenvector associated with ω_l, and $H_j^{j'}$ can be viewed as the derivative of the Hamiltonian H with respect to x_j and $\partial U/\partial x_{j'}$, calculated at $\{x_j\} = \{\bar{x}_j(t)\}$ and $\{\partial U/\partial x_{j'}\} = 0$.

For a reaction diffusion system, $H_j^{j'} = H_{r\alpha}^{r'\alpha'}$ is a sum of two parts related, respectively, to the chemical and diffusion processes. It reads

$$H_{r\alpha}^{r'\alpha'} = M_\alpha^{\alpha'}\delta(r-r') - D_\alpha \delta(\alpha-\alpha') \sum_a [\delta(r+a-r') - \delta(r-r')]$$

where $M_\alpha^{\alpha'}$ is the linearized operator of the chemical kinetics or equivalently the first derivative of the first chemical transition moment of the master equation.

For periodic boundary conditions, the components of the eigenvectors C^l of the operator $H_j^{j'}$ are given by

$$C_j^l = C_{r\alpha}^{m\beta} = (c_m)_\alpha^\beta\, e^{ir\cdot m}$$

Note that the vector m plays the role of a Fourier variable. The coefficients $(c_m)_\alpha^\beta$ are the eigenvectors of a simple matrix in the space of chemical concentrations. They obey the following equation:

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

with $K_m = \sum_a (1 - e^{ia\cdot m})$.

Now, the equation of the higher derivatives of U can be expressed in terms of the higher derivatives of the transition moments denoted by $M_{\alpha\alpha'\cdots}^{\alpha''\alpha'''\cdots}$ for the chemical processes and $H_{jj'\cdots}^{j''j'''\cdots}$ for the reaction-diffusion processes. These equations are more easily solved in the representation which diagonalizes $H_j^{j'}$. In the new variables $\sigma_l = \sigma_{m\beta}$, defined by

$$\xi_j = C_{\mathrm{j}}^{l^1} \sigma_{l^1} \qquad \text{where} \quad \xi_j = x_j - \bar{x}_j(t)$$

the expansion of U takes the following form:

$$U(\{\sigma_l\}) = \frac{1}{2!} U^{l^1 l^2} \sigma_{l^1} \sigma_{l^2} + \frac{1}{3!} U^{l^1 l^2 l^3} \sigma_{l^1} \sigma_{l^2} \sigma_{l^3} + \frac{1}{4!} U^{l^1 l^2 l^3 l^4} \sigma_{l^1} \sigma_{l^2} \sigma_{l^3} \sigma_{l^4} + \cdots \tag{2.3}$$

We adopt the convention that all repeated indices with numerical exponents $l^1, l^2,\ldots$ are summed over. The coefficients $U^{ll'\cdots}$ denote the derivatives of U with respect of $\sigma_l, \sigma_{l'},\ldots$. The expansion does not contain first-order terms because of the definition of the reference state, nor a constant term, which can always be incorporated in the normalization constant.

As pointed out in the Introduction, the effect of fluctuation on the deterministic attractor may be reduced to the study of the stationary solution of the master equation. This solution is obtained by solving the stationary equations of the successive derivatives of U when the reference state $\{\bar{x}_j(t)\}$ is a *homogeneous stationary* state $\bar{x}_j(t) = \bar{x}_{r\alpha}(t) = \bar{x}_\alpha$ for all t and r. The coefficients of Eq. (2.3) have then the following expressions:

$$U_{ll'}^{-1} = -\frac{H_{ll'}}{\omega_l + \omega_{l'}} \tag{2.4a}$$

$$U^{ll'l''} = \frac{U^{ll^1} U^{l'l^2} U^{l''l^3}}{\omega_{l^1} + \omega_{l^2} + \omega_{l^3}} \left[\mathscr{S}_{l^1 l^2 l^3} \{ H_{l^1}^{l^4 l^5} U_{\bar{l}^4 l^2}^{-1} U_{\bar{l}^5 l^3}^{-1} + H_{l^1 l^2}^{l^4} U_{\bar{l}^4 l^3}^{-1} \} + H_{l^1 l^2 l^3} \right] \tag{2.4b}$$

$$\begin{aligned} U^{ll'l''l'''} &- \mathscr{S}_{ll'l''l'''} \{ U^{ll'l^1} U_{\bar{l}^1 l^2}^{-1} U^{l^2 l'' l'''} \} \\ &= \frac{U^{ll^1} U^{l'l^2} U^{l''l^3} U^{l'''l^4}}{\omega_{l^1} + \omega_{l^2} + \omega_{l^3} + \omega_{l^4}} \left[\mathscr{S}_{l^1 l^2 l^3 l^4} \{ -H_{l^1}^{l^5 l^6} U_{\bar{l}^5 l^2}^{-1} U^{l^7 l^8 l^9} U_{\bar{l}^7 l^3}^{-1} U_{\bar{l}^8 l^4}^{-1} U_{\bar{l}^9 l^6}^{-1} \right. \\ &\quad - H_{l^1 l^2}^{l^5} U^{l^6 l^7 l^8} U_{\bar{l}^6 l^3}^{-1} U_{\bar{l}^7 l^4}^{-1} U_{\bar{l}^8 l^5}^{-1} + H_{l^1}^{l^5 l^6 l^7} U_{\bar{l}^5 l^2}^{-1} U_{\bar{l}^6 l^3}^{-1} U_{\bar{l}^7 l^4}^{-1} \\ &\quad \left. + H_{l^1 l^2}^{l^5 l^6} U_{\bar{l}^5 l^3}^{-1} U_{\bar{l}^6 l^4}^{-1} + H_{l^1 l^2 l^3}^{l^5} U_{\bar{l}^5 l^4}^{-1} \} + H_{l^1 l^2 l^3 l^4} \right] \end{aligned} \tag{2.4c}$$

where the quantities $H_{jj'\cdots}^{j''j'''\cdots}$ are intensive (independent of n). The symmetrization symbol $\mathscr{S}_{jj'\cdots}$ applied to a function of the indices $j, j',\ldots$

represents the sum of all *distinct* terms obtained by permutation of these indices. This system of equations has already been studied in the case of a bifurcation to steady states.(10,14) Our aim here is to apply it to a Hopf bifurcation.

In the vicinity of a bifurcation of time-dependent solutions, the spectrum of the linear operator $H_j^{j'}$ is characterized by a set of complex conjugate eigenvalues $\omega_{m\beta}$, $\omega_{m\beta}$ with a small real part.

In a general reaction-diffusion system of p variables ($p > 2$), the first instability will generally appear for a nonvanishing value of K_m, corresponding to a preferred spatial periodicity. However, we are interested primarily in reaction diffusion systems of *two* chemical components. In this case, the first instability appears for $K_q = 0$ and the critical modes are characterized by values of $m = q$ so that K_q is small. The corresponding critical eigenvalues ω_{l_0} may be written

$$\omega_{l_0} = \omega_{q\beta} = \eta_q + i\beta\theta_q$$

with, to the first order in K_q:

$$\eta_q = \eta_0 + \eta_1 K_q, \qquad \theta_q = \theta_0 + \theta_1 K_q$$

and where β can only take the two values 1 and -1 (or $\bar{1}$). The noncritical eigenvalues are denoted by ω_{l_ϕ}.

In order to exhibit the simplifications arising in Eqs. (2.4), when the system operates in the vicinity of a Hopf bifurcation, the central quantity which has to be analyzed is the coefficient of the quadratic term $U^{ll'}$ in the expansion (2.3).

2.1. Evaluation of the Quadratic Terms

We first examine the elements of the inverse matrix $U_{\bar{l}\bar{l}'}^{-1}$ which reduce at the dominant order to

$$U_{q\beta q'\beta'}^{-1} = -\delta(q + q') \frac{M_{\beta\beta'}}{\omega_{\bar{q}\bar{\beta}} + \omega_{\bar{q}'\bar{\beta}'}} \tag{2.5}$$

where $\bar{q} = -q \mod 2\pi$.

If $(q', \beta') = (\bar{q}, \bar{\beta})$, the denominator in the expression of $U_{q\beta\bar{q}\bar{\beta}}^{-1}$ goes to zero, and this element diverges as one approaches the bifurcation point. All the other elements $U_{\bar{l}\bar{l}'}^{-1}$ are supposed to remain finite. This implies that all elements $U^{ll'}$ are small and given by

$$U^{q\beta q'\beta'} = \delta(q + q')[\varepsilon_{q\beta}\delta(\beta + \beta') + O(\varepsilon_{q\beta}^2)]$$

with $\varepsilon_{q\beta} = -2(\eta_0 + \eta_1 K_q)/M_{\beta\bar{\beta}}$, $\varepsilon_{q\beta} = \varepsilon_{q\beta} = \varepsilon_q$.

Hence the quadratic terms separate in two groups:

$$\tfrac{1}{2}U^{l^1l^2}\sigma_{l^1}\sigma_{l^2}=\tfrac{1}{2}\varepsilon_{l_0^1}\sigma_{l_0^1}\sigma_{\bar{l}_0^1}+\tfrac{1}{2}U^{l_\phi^1l_\phi^2}\sigma_{l_\phi^1}\sigma_{l_\phi^2}$$

where use has been made of the property $\sigma_{\bar{l}_0}=\sigma_{l_0}^*$, $\sigma_{l_0}^*$ being the complex conjugate of σ_{l_0}.

2.2. Orders of Magnitude of the Various Terms

If all $\omega_{m\beta}$'s are negative and finite, there is no need to go further than the quadratic term. But if some eigenvalue becomes small, the probability of the states for which the noncritical components are small and such that

$$|\sigma_{l_\phi}|\lesssim(\varepsilon_{l_0})^{1/2}\,|\sigma_{l_0}| \tag{2.6}$$

must include higher-order terms. Cubic and quartic terms become therefore significant provided they have the same order of magnitude as the quadratic ones.

Comparing first quadratic and quartic terms, we see that, for nonsmall $U^{l_0l_0'l_0''l_0'''}$, they are comparable if $|\sigma_{l_0}|=O((\varepsilon_{l_0})^{1/2})$, which defines a small neighborhood of the bifurcation point.

Coming next to the cubic terms, we see, from the above argument, that one way that these terms can be comparable to the quadratic and quartic ones is

$$U^{l_0l_0'l_0''}=O((\varepsilon_q)^{1/2})$$

The only other way cubic terms can influence the stochastic potential is through the presence of one noncritical mode:

$$U^{l_0l_0'l_\phi}\sigma_{l_0}\sigma_{l_0'}\sigma_{l_\phi}$$

provided that $U^{l_0l_0'l_\phi}$ is of $O(1)$ and σ_{l_ϕ} of $O(\varepsilon_q)$. Summarizing, we can write the following stochastic potential:

$$\begin{aligned}U={}&\frac{1}{2!}\,\varepsilon_{l_0^1}\sigma_{l_0^1}\sigma_{\bar{l}_0^1}+\frac{1}{3!}\,U^{l_0^1l_0^2l_0^3}\sigma_{l_0^1}\sigma_{l_0^2}\sigma_{l_0^3}+\frac{1}{4!}\,U^{l_0^1l_0^2l_0^3l_0^4}\sigma_{l_0^1}\sigma_{l_0^2}\sigma_{l_0^3}\sigma_{l_0^4}\\&+\frac{1}{2}\,U^{l_\phi^1l_\phi^2}\sigma_{l_\phi^1}\sigma_{l_\phi^2}+\frac{1}{2}\,U^{l_0^1l_0^2l_\phi^1}\sigma_{l_0^1}\sigma_{l_0^2}\sigma_{l_\phi^1}\end{aligned} \tag{2.7}$$

2.3. Evaluation of the Cubic Terms

We first evaluate the third derivative of U with respect to three critical variables. On examining the orders of magnitude of the various terms of

Eq. (2.4b) for $l = q\beta$, $l' = q'\beta'$, $l'' = q''\beta''$, we see that $U^{q\beta q'\beta' q''\beta''}$ reduces at the dominant order to

$$U^{q\beta q'\beta' q''\beta''} = \delta(q + q' + q'')\, \frac{\varepsilon_q M_{\beta}^{\beta'\beta''} + \varepsilon_{q'} M_{\bar{\beta}'}^{\beta\beta''} + \varepsilon_{q''} M_{\bar{\beta}''}^{\beta\beta'}}{-i(\beta + \beta' + \beta'')\,\theta_0} \tag{2.8}$$

where $M_{\beta}^{\beta'\beta''}$ are the second derivatives of the first chemical transition moment.

As the denominator never vanishes, this quantity is of order ε_q, and can be therefore neglected.

There is thus no need to introduce a second smallness parameter in addition to the bifurcation one, as in the case of stationary space periodic solutions.

Coming next to the third derivative with respect to only one noncritical variable $\sigma_{m\beta}$, we see that $U^{m\beta q\beta q'\beta'}$ vanishes owing to the Kronecker delta $\delta(q + q' + m)$.

Summarizing, the stochastic potential contains no cubic terms at the dominant order.

Note that in a system of more than two components, this property is not preserved because of the existence of noncritical values of β.

2.4. Evaluation of the Quartic Terms

The coefficients of the quartic terms are deduced from Eq. (2.4c) for $l = l_0$, $l' = l'_0$, $l'' = l''_0$, $l''' = l'''_0$; at the dominant order, they reduce to

$$\begin{aligned} U^{l_0 l'_0 l''_0 l'''_0} = {} & \frac{\varepsilon_{l_0} \varepsilon_{l'_0} \varepsilon_{l''_0} \varepsilon_{l'''_0}}{\omega_{\bar{l}_0} + \omega_{\bar{l}'_0} + \omega_{\bar{l}''_0} + \omega_{\bar{l}'''_0}} \\ & \times \mathscr{S}_{l_0 l'_0 l''_0 l'''_0} \left\{ -H_{\bar{l}_0}^{l'_0 l^1_0} \frac{U^{l''_0 l'''_0 \bar{l}^1_0}}{\varepsilon_{l'_0} \varepsilon_{l''_0} \varepsilon_{l'''_0} \varepsilon_{l^1_0}} + H_{\bar{l}_0}^{l'_0 l''_0 l'''_0} \frac{1}{\varepsilon_{l'_0} \varepsilon_{l''_0} \varepsilon_{l'''_0}} \right\} \end{aligned} \tag{2.9}$$

The only nonvanishing quartic terms are such that, at the bifurcation point,

$$\omega_{\bar{l}_0} + \omega_{\bar{l}'_0} + \omega_{\bar{l}''_0} + \omega_{\bar{l}'''_0} = O(\varepsilon_{l_0}) \tag{2.10}$$

This implies

$$\beta + \beta' + \beta'' + \beta''' = 0$$

It is instructive to point out the analogy between the choice of the dominant terms in the expansion of the stochastic potential and the reduction of the corresponding deterministic equations into a normal form. In this later case, it turns out that the only nonlinear terms that cannot be eliminated are associated to "resonant eigenvalues" ω_s for which there exists $m_i \in \mathbb{N}$, $(i \in \{1,..., k\})$ and $s \in \{1,..., k\}$ such that

$$\omega_s = \sum_{i=1}^{k} m_i \omega_i \qquad \text{with} \quad \sum_{i=1}^{k} m_i \geqslant 2$$

ω_i being the ith eigenvalue of $H_j^{j'}$, calculated at the bifurcation.

Here the relation $\omega_{0\beta} = \omega_{0\beta} + p(\omega_{0\beta} + \omega_{0\beta})$ verified at the bifurcation point, defines the resonances for all $p \geqslant 1$. The first resonance appears at $p = 1$ and is of order 3. Owing to the nonexistence of resonance of order 2, the terms of order 2 can be eliminated from the deterministic equation in agreement with our result that terms of order 3 are absent from the stochastic potential.

Actually for values of the smallness parameter η_0 slightly different from zero, and for a large system, the first resonance relation is replaced by

$$\omega_{q\beta} - \omega_{q'\beta} - \omega_{q''\beta} - \omega_{q'''\bar{\beta}} = O(\varepsilon_q)$$

or

$$\omega_{q\beta} + \omega_{q'''\beta} + \omega_{q'\beta} + \omega_{q''\bar{\beta}} = 0$$

This is exactly the condition (2.10).

We choose to write the term $\beta = 1$, $\beta' = 1$, $\beta'' = \bar{1}$, $\beta''' = \bar{1}$ for which

$$\begin{aligned}
&\omega_{\bar{q}\bar{1}} + \omega_{\bar{q}'\bar{1}} + \omega_{\bar{q}''1} + \omega_{\bar{q}'''1} \\
&\qquad = 4\eta_0 + \eta_1(K_q + K_{q'} + K_{q''} + K_{q'''}) + i\theta_1(-K_q - K_{q'} + K_{q''} + K_{q'''})
\end{aligned}$$

We denote

$$K = K_q + K_{q'} + K_{q''} + K_{q'''}, \qquad \underline{K} = -K_q - K_{q'} + K_{q''} + K_{q'''}.$$

Equation (2.9) is simplified by using the result previously obtained for $U^{l_0'' l_0''' \bar{l}_0^1}$ [Eq. (2.8)] and the expressions of $H_{l_0}^{l_0' l_0^1}$ and $H_{l_0}^{l_0' l_0'' l_0'''}$ in terms of the derivatives of the chemical transition moments. We thus obtain

$$\begin{aligned}
&U^{q1q'1q''\bar{1}q'''\bar{1}} \\
&\quad = -\frac{2\delta(q + q' + q'' + q''')}{M_{1\bar{1}}(4\eta_0 + \eta_1 K + i\theta_1 \underline{K})} \left\{ \left(\operatorname{Re} M_1^{11\bar{1}} - \frac{1}{\theta_0} \operatorname{Im} M_1^{11} M_1^{1\bar{1}} \right) (4\eta_0 + K\eta_1) \right. \\
&\quad\quad \left. + i\eta_1 \underline{K} \left[\operatorname{Im} M_1^{11\bar{1}} - \frac{1}{\theta_0} \left(\frac{1}{3} |M_1^{11}|^2 + 2 |M_1^{1\bar{1}}|^2 - \operatorname{Re} M_1^{11} M_1^{1\bar{1}} \right) \right] \right\}
\end{aligned}$$

Introducing the following notation:

$$u_1 = \operatorname{Re} M_1^{11\bar{1}} - \frac{1}{\theta_0} \operatorname{Im} M_1^{11} M_1^{1\bar{1}}$$

$$u_2 = \operatorname{Im} M_1^{11\bar{1}} - \frac{1}{\theta_0} \left(\frac{1}{3} |M_{\bar{1}}^{11}|^2 + 2\,|M_1^{1\bar{1}}|^2 - \operatorname{Re} M_1^{11} M_1^{1\bar{1}} \right)$$

$$u(q, q', q'', q''') = u_1 + \frac{i\underline{K}(\eta_1 u_2 - \theta_1 u_1)}{4\eta_0 + \eta_1 K + i\theta_1 \underline{K}}$$

we finally find

$$U^{q1q'1q''\bar{1}q'''\bar{1}} = \frac{-2}{M_{1\bar{1}}} \delta(q + q' + q'' + q''')\, u(q, q', q'', q''')$$

Note the properties of u:

$$u(q, q', q'', q''') = u(|q|, |q'|, |q''|, |q'''|)$$

$$u(q, q', q'', q''') = u^*(q'', q''', q, q')$$

where * means complex conjugate.

2.5. Stochastic Potential

We are now in position to write the stochastic potential in the reduced form:

$$\begin{aligned} U = &-\frac{2}{M_{1\bar{1}}} \left[\sum_q (\eta_0 + \eta_1 K_q)\, \sigma_{q1} \sigma_{\bar{q}\bar{1}} \right. \\ &\left. + \frac{6}{4!} \sum_{qq'q''q'''} \delta(q + q' + q'' + q''')\, u(q, q', q'', q''')\, \sigma_{q1} \sigma_{q'1} \sigma_{q''\bar{1}} \sigma_{q'''\bar{1}} \right] \\ &+ \frac{1}{2} U^{m^1\beta^1 m^2\beta^2} \sigma_{m^1\beta^1} \sigma_{m^2\beta^2} \end{aligned} \tag{2.11}$$

with $m^1 \neq q^1$ and $m^2 \neq q^2$.

The potential appears as a sum of two well-separated parts: a fourth-order polynomial U_{cr} relative to the critical variables and a second-order one, associated to a Gaussian distribution for the noncritical modes. It follows that the probability of the noncritical modes is a Gaussian distribution centered on the stationary state $\sigma_l = 0$, independently of the critical variables. This property justifies the validity of the rotating wave approximation used in Refs.13 and 19 to eliminate the noncritical modes.

2.6. Illustration

The above results will now be applied to the trimolecular model, the so-called Brusselator, corresponding to the following reaction scheme:

$$\begin{aligned} A &\to X \\ 2X + Y &\to 3X \\ B + X &\to Y + D \\ X &\to E \end{aligned}$$

Using standard notations, the eigenvalues of the linear stability operator verify the following matrix equation:

$$\begin{pmatrix} B - 1 - K_q D_1 - \omega_{q\beta} & A^2 \\ -B & -A^2 - K_q D_2 - \omega_{q\beta} \end{pmatrix} \begin{pmatrix} (c_q)_1^\beta \\ (c_q)_{\bar{1}}^\beta \end{pmatrix} = 0$$

At the first order in K_q and $B - B_c = B - (A^2 + 1)$, the eigenvalues reduce to

$$\omega_{q\beta} = \eta_0 + \eta_1 K_q + i\beta(\theta_0 + \theta_1 K_q)$$

with

$$\eta_0 = \frac{B - B_c}{2}, \qquad \eta_1 = -\frac{D_1 + D_2}{2}$$

$$\theta_0 = A, \qquad \theta_1 = \frac{A}{2}(D_1 - D_2)$$

The eigenvector matrix, evaluated at the lowest order, is independent of K_q, and is chosen as

$$[(c_q)_\alpha^\beta]_{\alpha \in \{1, \bar{1}\}, \beta \in \{1, \bar{1}\}} = \begin{pmatrix} -A & -A \\ A - i & A + i \end{pmatrix}$$

The coefficients necessary to write the critical potential U_{cr} are then

$$M_{1\bar{1}} = \frac{A^2 + 1}{A}$$

$$u_1 = -(A^2 + 2)$$

$$u_2 = -\frac{4A^4 - 7A^2 + 4}{3A}$$

The resulting stochastic potential is in agreement with the generalized potential $\mathscr{F}$ found by Walgraef *et al.*[12] with a Langevin equation approach, after a rescaling of the variables $\sigma_{q\beta}$. In addition, the comparison allows us to evaluate the strength of the random force Γ:

$$\Gamma = M_{1\bar{1}} = \frac{A^2+1}{A}$$

3. SEPARATION OF PHASE AND RADIAL VARIABLES IN THE PROBABILITY DISTRIBUTION

Our next step is to express the critical part of the stochastic potential around the stationary states, in a form exhibiting separately the contributions of the phase and of the amplitude of the oscillating variables.

In contrast with the process of adiabatic elimination used in Ref. 13 which ignores *a priori* the fluctuations of the fast variables, the analysis of the stochastic potential will allow us to calculate the probability associated to the slow as well as fast variables.

To this end, we seek for a local expansion of the critical part of the potential around a state for which all the first derivatives of U_{cr} vanish:

$$\frac{\partial U_{\text{cr}}}{\partial \sigma_{q\beta}} = 0 \qquad \text{for all } \sigma_{q\beta} \tag{3.1}$$

From Eq. (2.11), Eq. (3.1) reads

$$\frac{\partial U_{\text{cr}}}{\partial \sigma_{q1}} = -\frac{2}{M_{1\bar{1}}}\Bigg[(\eta_0+\eta_1 K_q)\,\sigma_{\bar{q}\bar{1}} + \frac{1}{2}\sum_{q'q''q'''} \delta(q+q'+\bar{q}''+\bar{q}''')\,u(q,q',q'',q''')\,\sigma_{q'1}\sigma_{\bar{q}''\bar{1}}\sigma_{\bar{q}'''\bar{1}}\Bigg] = 0$$

$$\frac{\partial U_{\text{cr}}}{\partial \sigma_{\bar{q}\bar{1}}} = -\frac{2}{M_{1\bar{1}}}\Bigg[(\eta_0+\eta_1 K_q)\,\sigma_{q1} + \frac{1}{2}\sum_{q'q''q'''} \delta(q+q'+\bar{q}''+\bar{q}''')\,u(q,q',q'',q''')\,\sigma_{\bar{q}'\bar{1}}\sigma_{q''1}\sigma_{q'''1}\Bigg] = 0$$

These two equations are complex conjugate, and admit the homogeneous solution:

$$\sigma_{q1} = \sigma_0\delta_{q0}, \qquad \sigma_{\bar{q}\bar{1}} = \sigma_0^*\delta_{q0} \tag{3.2}$$

where σ_0 obeys

$$\eta_0\sigma_0+\tfrac{1}{2}u_1\sigma_0^*\sigma_0\sigma_0=0 \tag{3.3}$$

with

$$u_1=u(0,0,0,0)$$

The nontrivial solution of Eq. (3.3) defines a homogeneous limit cycle of radius:

$$R_0=|\sigma_0|=\left(\frac{-2\eta_0}{u_1}\right)^{1/2} \tag{3.4}$$

Any point on the limit cycle is fixed by

$$\sigma_0=R_0e^{i\varphi_0} \tag{3.5}$$

where φ_0 is the phase.

For all points of this limit cycle, the probability is constant, at the dominant order.

3.1. Local Description Around a Point of the Limit Cycle

We look now for a local expansion V of the critical potential around any point of a given limit cycle defined by the value of η_0.

The first step of the calculation consists in writing the second derivatives of the critical potential U_{cr} found in Section 2, and in evaluating them at the homogeneous state defined by Eq. (3.3) and denoted by index 0. We obtain

$$\left.\frac{\partial^2U_{\text{cr}}}{\partial\sigma_{q1}\,\partial\sigma_{q'1}}\right|_0=-\frac{1}{M_{1\bar{1}}}\,\delta(q+q')\,u(q,q,0,0)\,\sigma_0^*\sigma_0^*$$

$$\left.\frac{\partial^2U_{\text{cr}}}{\partial\sigma_{\bar{q}\bar{1}}\,\partial\sigma_{\bar{q}'\bar{1}}}\right|_0=-\frac{1}{M_{1\bar{1}}}\,\delta(q+q')\,u^*(q,q,0,0)\,\sigma_0\sigma_0$$

$$\left.\frac{\partial^2U_{\text{cr}}}{\partial\sigma_{q1}\,\partial\sigma_{\bar{q}'\bar{1}}}\right|_0=-\frac{2}{M_{1\bar{1}}}\,\delta(q-q')[\eta_0+\eta_1K_q+u(q,0,q,0)\,\sigma_0\sigma_0^*]$$

To facilitate the writing of the quadratic part $V^{(2)}$ of the expansion, we introduce the shorthand notation:

$$\begin{aligned}
u(q,q,0,0)&=u_1+u_q\\
\eta_0+\eta_1K_q&=\eta_q\\
\sigma_{q1}-\sigma_0\delta_{q0}&=\zeta_{q1}\\
\sigma_{q\bar{1}}-\sigma_0^*\delta_{q0}&=\zeta_{q\bar{1}}
\end{aligned} \tag{3.6}$$

This gives

$$V^{(2)} = -\frac{2}{M_{1\bar{1}}}\sum_{qq'}\delta(q+q')\left[\frac{u_1+u_q}{4}\sigma_0^*\sigma_0^*\zeta_{q1}\zeta_{q'1}\right.$$
$$\left.+\frac{1}{2}(\eta_q+u_1\sigma_0\sigma_0^*)(\zeta_{q1}\zeta_{q'\bar{1}}+\zeta_{q\bar{1}}\zeta_{q'1})+\frac{u_1+u_q^*}{4}\sigma_0\sigma_0\zeta_{q\bar{1}}\zeta_{q'\bar{1}}\right]$$

Using the conjugation relations between the variables to eliminate $\zeta_{\bar{q}1}$ and $\zeta_{\bar{q}\bar{1}}$, we can write $V^{(2)}$ as a Hermitian form:

$$V^{(2)} = -\frac{2}{M_{1\bar{1}}}\sum_{q}\left[\frac{u_1+u_q}{4}\sigma_0^*\sigma_0^*\zeta_{q1}\zeta_{q\bar{1}}^*\right.$$
$$\left.+\frac{1}{2}(\eta_q+u_1\sigma_0\sigma_0^*)(\zeta_{q1}\zeta_{q1}^*+\zeta_{q\bar{1}}\zeta_{q\bar{1}}^*)+\frac{u_1+u_q^*}{4}\sigma_0\sigma_0\zeta_{q\bar{1}}\zeta_{q1}^*\right] \tag{3.7}$$

The matrix associated to $(-M_{1\bar{1}}/2)\, V^{(2)}$ is simplified by using the value of σ_0 [Eq. (3.5)]:

$$(\zeta_{q1}^*\zeta_{q\bar{1}}^*)\begin{pmatrix}\frac{1}{2}(-\eta_0+\eta_1 K_q) & -\frac{1}{2}\eta_0\frac{u_1+u_q^*}{u_1}e^{2i\varphi_0}\\ -\frac{1}{2}\eta_0\frac{u_1+u_q}{u_1}e^{-2i\varphi_0} & \frac{1}{2}(-\eta_0+\eta_1 K_q)\end{pmatrix}\begin{pmatrix}\zeta_{q1}\\ \zeta_{q\bar{1}}\end{pmatrix} \tag{3.8}$$

Our next step is the diagonalization of this matrix, and the separation of the two eigenvalues according to their different order of magnitude. The resulting eigenvalues are

$$\lambda_q = \frac{1}{2}\left(-\eta_0+\eta_1 K_q \pm \eta_0\left|1+\frac{u_q}{u_1}\right|\right) \tag{3.9}$$

As we are only concerned with the vicinity of the homogeneous reference state σ_0, K_q may be regarded as a suitable smallness parameter which controls our expansion. Let us recall that the smallness of η_0 guarantees the validity of the previous expansion of U [cf. Eq. (2.11)], but here we suppose to be far enough from the critical point, so that $\eta_0 \gg \eta_1 K_q$. This allows the following expansion of the eigenvalues in powers of K_q/η_0:

$$\begin{aligned}\lambda_{q0} &= \frac{\eta_1 K_q}{2} + O\left(\left(\frac{K_q}{\eta_0}\right)^2\right)\\ \lambda_{q1} &= -\eta_0 + \frac{1}{2}\eta_1 K_q + O\left(\left(\frac{K_q}{\eta_0}\right)^2\right)\end{aligned} \tag{3.10}$$

The important point is the different order of magnitude between the two kinds of eigenvalues. If we adopt the terminology of the adiabatic elimination process, the finite eigenvalue λ_{q1} will be associated to the so-called fast modes, whereas the small ones λ_{q0} to the slow modes. In particular, for $q=0$, λ_{q0} reduces to zero, and the corresponding eigenvector defines a direction tangent to the homogeneous limit cycle.

Note that, for any two-component chemical system, $\eta_1 = -(D_1+D_2)/2$ is negative while η_0 is positive above the bifurcation point: Thus, all the eigenvalues $(-2/M_{1\bar{1}})\,\lambda_{q0}$ and $(-2/M_{1\bar{1}})\,\lambda_{q1}$ are positive or vanish. This proves that the probability $P=e^{-\mathcal{N}V}$ is maximal at any point of the limit cycle, which is then an attractor.

To express $V^{(2)}$ in the new coordinates denoted by y_{q0} and y_{q1}, we choose the following normalized eigenvector matrix:

$$P=\begin{pmatrix} \frac{1}{\sqrt{2}}e^{i\varphi_0} & \frac{1}{\sqrt{2}}e^{i\varphi_0} \\ -\frac{1}{\sqrt{2}}e^{-i\varphi_0} & \frac{1}{\sqrt{2}}e^{-i\varphi_0} \end{pmatrix} \tag{3.11}$$

The relations between the new coordinates and the old ones are then

$$\begin{aligned} \zeta_{q1} &= \frac{1}{\sqrt{2}} e^{i\varphi_0}(y_{q0}+y_{q1}) \\ \zeta_{q\bar{1}} &= \frac{1}{\sqrt{2}} e^{-i\varphi_0}(-y_{q0}+y_{q1}) \end{aligned} \tag{3.12}$$

This leads to the following expression of the quadratic form:

$$V^{(2)} = -\frac{2}{M_{1\bar{1}}}\sum_q \lambda_{q0}\,|y_{q0}|^2 + \lambda_{q1}\,|y_{q1}|^2 \tag{3.13}$$

The states for which the fast components are small and such that

$$|y_{q1}|^2 \approx \frac{\eta_1 K_q}{\eta_0}\,|y_{q0}|^2 \tag{3.14}$$

are not described adequately by $V^{(2)}$. For these states, it is necessary to evaluate the orders 3 and 4. We thus evaluate next the dominant part of $V^{(3)}$ and $V^{(4)}$. The situation is analogous to the one described in Section 2, which had led to the separation between critical and noncritical terms. Let us examine the third order. We find in the old coordinates

$$V^{(3)} = -\frac{2}{M_{1\bar{1}}} \sum_{qq'q''} \delta(q+q'+q'')[u(q,q',q'',0)\,\sigma_0^*\zeta_{q1}\zeta_{q'1}\zeta_{q''\bar{1}} \\ + u(q,0,q',q'')\,\sigma_0\zeta_{q1}\zeta_{q'\bar{1}}\zeta_{q''\bar{1}}]$$

Carrying out the change of coordinates defined by Eq. (3.12) and looking for the dominant contributions, we first observe that the coefficient of $y_{q0}\,y_{q'0}\,y_{q''0}$ is of $O(K_q/\eta_0)$. Consequently, $V^{(3)}$ reduces at the dominant order to

$$V^{(3)} = -\frac{2}{M_{1\bar{1}}}\frac{1}{2}(-u_1\eta_0)^{1/2} \sum_{qq'q''} \delta(q+q'+q'')\, y_{q0}\,y_{q'0}\,y_{q''1} \tag{3.15}$$

From relation (3.14), the order of magnitude of this contribution is comparable to the one of $V^{(2)}$ if $y_{q0} \approx (K_q)^{1/2}$. It is then of $O(K_q^2)$.

Coming next to the fourth-order term $V^{(4)}$, we find at the dominant order K_q^2

$$V^{(4)} = -\frac{2}{M_{1\bar{1}}}\frac{u_1}{16} \sum_{qq'q''q'''} \delta(q+q'+q''+q''')\, y_{q0}\,y_{q'0}\,y_{q''0}\,y_{q'''0} \tag{3.16}$$

Summarizing, we can write the following local expansion:

$$V = V^{(0)} - \frac{2}{M_{1\bar{1}}}\left[\sum_q \frac{1}{2}\eta_1 K_q\,|y_{q0}|^2 - \eta_0\,|y_{q1}|^2 \right. \\ + \frac{1}{2}(-u_1\eta_0)^{1/2} \sum_{qq'q''} \delta(q+q'+q'')\, y_{q0}\,y_{q'0}\,y_{q''1} \\ \left. + \frac{u_1}{16} \sum_{qq'q''q'''} \delta(q+q'+q''+q''')\, y_{q0}\,y_{q'0}\,y_{q''0}\,y_{q'''0}\right] \tag{3.17}$$

where $V^{(0)}$ is a constant depending on the reference state σ_0, and will hereafter be ignored.

Taking advantage of the conjugation relations between the new variables defined in Eq. (3.12), namely:

$$y_{q0}^* = -y_{\bar{q}0}, \qquad y_{q1}^* = y_{\bar{q}1}$$

we further transform the expression of V in order to separate the contributions of the fast and slow variables, by using the same method as in Section 2. We obtain

$$\begin{aligned} V = -\frac{2}{M_{1\bar{1}}} \sum_q \Bigg[& \frac{1}{2} \eta_1 K_q \, |y_{q0}|^2 \\ & - \eta_0 \left(y_{q1} - \frac{1}{4} \left(\frac{-u_1}{\eta_0} \right)^{1/2} \sum_{q'q''} \delta(q' + q'' - q) \, y_{q'0} \, y_{q''0} \right) \\ & \times \left(y^*_{q1} - \frac{1}{4} \left(\frac{-u_1}{\eta_0} \right)^{1/2} \sum_{q'''q''''} \delta(q''' + q'''' - q) \, y^*_{q'''0} \, y^*_{q''''0} \right) \Bigg] \end{aligned} \tag{3.18}$$

We first observe that the probability of the fast variables is a multivariate Gaussian distribution centered in

$$y_{q1} = \frac{1}{4} \left(\frac{-u_1}{\eta_0} \right)^{1/2} \sum_{q'q''} \delta(q' + q'' - q) \, y_{q'0} \, y_{q''0} \tag{3.19}$$

This provides the justification of the adiabatic elimination of the fast variables used in Ref. 13. Note that the quartic terms in V are entirely included in the above Gaussian distribution. As a result, the probability of the slow variables also reduces to a Gaussian distribution.

3.2. Description in Terms of Phase and Amplitude Variables

Polar coordinates are especially appropriate to describe the potential expanded around a point of the limit cycle. We thus introduce the phase and radial variables by

$$\begin{aligned} \sigma_{q1} &= \frac{1}{n} R_{r^1} \exp[i(\varphi_{r^1} + r^1 q)] \\ \sigma_{q\bar{1}} &= \frac{1}{n} R_{r^1} \exp[i(-\varphi_{r^1} + r^1 q)] \end{aligned} \tag{3.20}$$

where R_r and φ_r appear, respectively, as modulus and argument of the inverse Fourier transform of σ_{q1}. Using the relations between the variables $y_{q\beta}$ and $\zeta_{q\beta}$ on one hand [Eq. (3.12)], $\zeta_{q\beta}$ and $\sigma_{q\beta}$ on the other hand [Eq. (3.6)], one can express $y_{q\beta}$ in terms of the new phase and radial variables as

$$\begin{aligned} y_{q0} &= \sqrt{2} \, \frac{i}{n} R_{r^1} \sin(\varphi_{r^1} - \varphi_0) \, e^{ir^1 q} \\ y_{q1} &= \sqrt{2} \, \frac{1}{n} (R_{r^1} \cos(\varphi_{r^1} - \varphi_0) - R_0) \, e^{ir^1 q} \end{aligned} \tag{3.21}$$

The conditions of validity of our local expansion V around a point of the homogeneous limit cycle defined by Eqs. (3.2)–(3.5), imply that the phase φ_r and the radius R_r remain close to φ_0 and R_0. Under these conditions, Eqs. (3.21) become at the dominant order

$$\begin{aligned} y_{q0} &= \sqrt{2}\,\frac{i}{n}\,R_0(\varphi_{r^1}-\varphi_0)\,e^{ir^1q} \\ y_{q1} &= \sqrt{2}\,\frac{1}{n}\,(R_{r^1}-R_0)\,e^{ir^1q} \end{aligned} \tag{3.22}$$

In terms of φ_r and R_r, the potential V reduces to

$$V=-\frac{2}{M_{1\bar{1}}}\left[\sum_q \frac{\eta_1 K_q}{n^2}\,R_0^2\,|(\varphi_{r^1}-\varphi_0)\,e^{ir^1q}|^2-\frac{2}{n^2}\,\eta_0\,|(R_{r^1}-R_0)\,e^{ir^1q}|^2\right]$$

This allows us to speak of the probability of the phase:

$$P_\varphi=e^{-\mathcal{N}V_\varphi}$$

with

$$V_\varphi=-\frac{2}{M_{1\bar{1}}}\sum_q \frac{\eta_1}{n^2}\,K_q R_0^2\,|(\varphi_{r^1}-\varphi_0)\,e^{ir^1q}|^2 \tag{3.23}$$

We thus conclude that phase and radial variables can be separated, and behave, respectively, as the slow and fast modes.

Note that the summation over q can be explicitly carried out, and lead, by switching to a continuous description of the space variable r, to

$$V_\varphi=-\frac{2}{M_{1\bar{1}}}\,\frac{\tilde{\eta}_1}{\mathscr{V}}\,R_0^2\int[\nabla(\varphi_r-\varphi_0)]^2\,dr \tag{3.24}$$

with $\tilde{\eta}_1=(\mathscr{D}_1+\mathscr{D}_2)/2$, where $\mathscr{D}_i$ is the Fick coefficient of the component i.

As mentioned earlier the validity of this expression is limited to small values of $\varphi_r-\varphi_0$. However, Eq. (3.24) shows that V_φ is independent of the phase φ_0 of the arbitrary reference state on the limit cycle ($\nabla\varphi_0=0$). Hence, the same form of V_φ can be used to express the phase probability of any state in the neighborhood of the whole limit cycle. It reads

$$P_\varphi=\exp\left(\frac{2}{M_{1\bar{1}}}\,\tilde{\eta}_1 R_0^2\int[\nabla\varphi_r]^2\,dr\right)$$

This result is in agreement with the probability found in Ref. 13, and allows for the calculation of the correlation function of the phase.

Walgraef *et al.*(13) show that the latter vanishes for infinitely extended low-dimensial systems. This compromises the possibility of sustaining oscillations in one- or two-dimension systems.

Note that the deterministic analysis alone could never lead to such a conclusion.

4. CONCLUSION

The stochastic potential analysis, which has been applied to a Hopf bifurcation in the present paper, provides a general method of investigation of the instabilities in nonequilibrium systems. Indeed a great variety of singular situations arising through bifurcations can be described by a systematic study of the Taylor expansion of the stochastic potential U in the vicinity of a homogeneous reference state.

The single linear change of variables which diagonalizes the linear operator of the deterministic equations is enough to simplify this expansion of U. The resulting form exhibits the singularities arising when the system operates in a region of the parameter space close to one or several bifurcations. By a simple estimate of the order of magnitude of the various terms in this expansion, one is able first to separate critical from noncritical modes and second to reduce the critical part to the so-called "resonant" terms in the terminology of the theory of normal form. It is worth pointing out that the expansion thus obtained for the critical stochastic potential is equivalent to one particular unfolding of the normal form, imposed by the specific reaction-diffusion model.

Moreover the determination of the states for which the gradient of the stochastic potential vanishes leads to a set of singular points which contains the fixed points as well as the other nearby attractors of the vector field.

In addition to the deterministic properties deduced from the analysis mentioned above, the stochastic potential provides information on the fluctuations around the attractor. In the case of the Hopf bifurcation these fluctuations turn out to play a major role in the destruction of long-range order in low-dimensional systems as known in Ref. 13. In the present paper, this result is deduced from a systematic analysis of the local properties of the critical stochastic potential.

We expect that this method is generalizable to more complicated cases like degenerate bifurcations. The deterministic study of such a situation has been carried out by Guckenheimer(18) for finitely extended systems for which the linear operator displays, at the bifurcation point, a simple zero eigenvalue and a pair of purely imaginary ones.

We intend to report of these problems in a forthcoming publication.

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