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On the Formation of Dissipative Structures in Reaction-Diffusion Systems

——*Reductive Perturbation Approach*——

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A unified viewpoint on the dynamics of spatio-temporal organization in various reaction-diffusion systems is presented. A dynamical similarity law attained near the instability points plays a decisive role in our whole theory. The method of reductive perturbation is used for extracting a scale-invariant part from original macroscopic equations of motion. It is shown that in many cases the dynamics near the instability point is governed by the time-dependent Ginzburg-Landau equation with coefficients which are in general complex numbers. An important effect of the imaginary parts of these coefficients on the stability of a spatially uniform limit cycle against inhomogeneous perturbation is also discussed.

§ 1. Introduction

Macroscopic spatio-temporal organization of matter is a most striking and attractive feature met in far-from-equilibrium situations.[1] In chemical and biochemical kinetics, a large number of theoretical models which can explain, to some extent, each individual phenomenon has been proposed,[1),2)] yet a unified viewpoint which is of conceptual as well as practical importance seems still lacking. The present paper aims to find out a universal feature emerging near the instability points in a fairly wide class of nonlinear chemical kinetics. Specifically, a universal form of macroscopic equation of motion valid near the instability points will be found. Such an equation also serves as an idealized model for a self-organized system which is not necessarily near the instability point.

Landau's classical theory of the second-order phase transition or more advanced scaling theory[3] tells us that the macroscopic properties of seemingly quite different systems may be described near the critical point with an identical equation if one makes an appropriate scaling of some physical quantities. Such a great simplicity of description is entirely due to the existence of a small parameter, i.e., the measure of deviation from the critical point. In other words, the role of this small parameter is to single out a few relevant variables or parameters from many ones, thus leading to a scale-invariance or a similarity property. In far-from-thermal equilibrium one often finds a similar situation near the catastrophe of a certain phase. It should be remenbered, however, that our main concern in the present paper is *not* to discuss the critical phenomena in its ordinary sense but to find out a simple description near the instability point for primary understanding of the formation of space-time structure from a unified viewpoint.

We shall make a reduction of macroscopic equation of motion including a small parameter by means of reductive perturbation.[4] The present paper may be regarded as an extension of previous work,[5] where the Prigogine-Lefever-Nicolis model[1] for nonlinear chemical kinetics was studied, to a general two-component system under chemical reaction and diffusion.

As was discussed by Tomita et al.,[6] possible types of instability in a system with a couple of degrees of freedom may be classified into two groups, namely, the soft mode instability and the hard mode instability. In the former case a single eigenmode becomes unstable and its eigenvalue vanishes at the marginal situation. In the latter case a couple of modes becomes unstable simultaneously; The real parts of their eigenfrequencies do not vanish at the marginal situation, thus leading to a time order beyond instability. It will turn out that the usefulness of such a classification is essentially unchanged when the spatial inhomogeneity is introduced. A most important conclusion from the present paper is that the dynamics near the *hard* mode instability point can be described with a generalized form of the time-dependent Ginzburg-Landau equation (abbreviated as the TDGL equation hereafter) for a complex field $W \equiv \rho e^{i\varphi}$ which has a certain relation with concentration fields. This equation has the form

$$\frac{\partial W}{\partial T} = (\pm 1 + ic_0) W + (1 + ic_1) \nabla_{\boldsymbol{R}}^2 W - (1 + ic_2) |W|^2 W, \qquad (1\cdot1)$$

where T and $\boldsymbol{R}$ are time and position vector each under an appropriate scaling, and c_i are real constants. In the case of soft mode instability, a certain restriction on the model should be required in order that the reductive perturbation may be applicable. At least for one-dimensional space order, which may be categorized as a soft mode instability, the ordinary TDGL equation where all c_i are vanishing in $(1\cdot1)$, has been found.

The existence of c_i in the case of hard mode instability implies that the two kinds of degrees of freedom, i.e., ρ and φ, are equally important. This is in contrast to the case of soft mode instability where the equation may be expressed only in terms of ρ. By introducing an appropriate rotating frame of reference, one may eliminate c_0 from $(1\cdot1)$, but c_1 and c_2 cannot be eliminated. The role of these uneliminated constants turns out crucial. In fact, when a certain condition is fulfilled by c_1 and c_2, the spatially uniform limit cycle becomes unstable against inhomogeneous perturbation. Such an instability may be expected to lead to a new type of organized phase, though the detailed analysis will not be given in the present paper.

§ 2. Linear stability theory of a reaction-diffusion system

A macroscopic equation of motion for a multi-component state variable $\boldsymbol{X}(\boldsymbol{r}, t)$

in a system under chemical reaction and diffusion may be expressed as

$$\frac{\partial}{\partial t}\boldsymbol{X}-\hat{D}\nabla_r^2\boldsymbol{X}=\boldsymbol{F}(\boldsymbol{X}), \tag{2·1}$$

where $\boldsymbol{F}(\boldsymbol{X})$ is in general a nonlinear function of $\boldsymbol{X}$ and $\hat{D}$ is a diagonal matrix formed by diffusion constants. The diffusion constants together with the other parameters included in $\boldsymbol{F}$ define a parameter space. We assume that Eq. (2·1) has at least one stationary, spatially uniform and asymptotically stable solution $\boldsymbol{X}_0$ in some physical region of the parameter space.

Let us now restrict our consideration to two-component system:

$$\boldsymbol{X}\equiv\begin{pmatrix} X \\ Y \end{pmatrix}, \tag{2·2}$$

$$\hat{D}\equiv\begin{pmatrix} D_x & 0 \\ 0 & D_y \end{pmatrix}. \tag{2·3}$$

Putting

$$\boldsymbol{X}=\boldsymbol{X}_0+\boldsymbol{x}, \tag{2·4}$$

where

$$\boldsymbol{x}\equiv\begin{pmatrix} x \\ y \end{pmatrix},$$

Eq. (2·1) may now be rewritten in the form

$$\hat{\Gamma}\boldsymbol{x}=\boldsymbol{G}(\boldsymbol{x}), \tag{2·5}$$

where

$$\hat{\Gamma}\left(-i\frac{\partial}{\partial t}, -i\nabla_r\right)\equiv\begin{pmatrix} \frac{\partial}{\partial t}+K_{xx}-D_x\nabla_r^2 & K_{xy} \\ K_{yx} & \frac{\partial}{\partial t}+K_{yy}-D_y\nabla_r^2 \end{pmatrix}, \tag{2·6}$$

and $\boldsymbol{G}$ is a quantity including only nonlinear terms in $\boldsymbol{x}$. We put

$$\boldsymbol{G}\equiv\begin{pmatrix} P \\ Q \end{pmatrix} \tag{2·7}$$

and introduce a notation R which will frequently be used below for representing either P or Q. The quantity R may in general be expanded as

$$R=\boldsymbol{x}^t\hat{R}_2\boldsymbol{x}+\boldsymbol{x}^t\hat{R}_3\hat{x}\boldsymbol{x}+\text{higher order terms in } \boldsymbol{x}, \tag{2·8}$$

where t indicates a transpose; $\hat{R}_2$ and $\hat{R}_3$ are 2×2 coefficient matrices and

$$\hat{x}=\begin{pmatrix} x & 0 \\ 0 & y \end{pmatrix}. \tag{2·9}$$

Let us now study the stability of the state $\boldsymbol{x}=0$ against infinitesimal perturbation. Introducing an operator $\hat{L}$ by

$$\hat{L}\left(-i\frac{\partial}{\partial t}, -i\nabla_r\right)=\begin{pmatrix} \frac{\partial}{\partial t}+K_{yy}-D_y\nabla_r^2 & -K_{xy} \\ -K_{yx} & \frac{\partial}{\partial t}+K_{xx}-D_x\nabla_r^2 \end{pmatrix}, \tag{2·10}$$

Eq. (2·5) may be rewritten as

$$\mathcal{L}\boldsymbol{x}=\hat{L}\boldsymbol{G}, \tag{2·11}$$

where

$$\mathcal{L}\left(-i\frac{\partial}{\partial t}, -i\nabla_r\right)=\det\hat{\Gamma}. \tag{2·12}$$

Let us consider for simplicity an infinitely large medium. Retaining only the linear part in $\boldsymbol{x}$, and setting $\boldsymbol{x}\propto\exp(i\omega t+i\boldsymbol{kr})$, one finds the dispersion equation

$$\mathcal{L}(\omega,\boldsymbol{k})=\omega^2+i\alpha\omega+\beta=0, \tag{2·13}$$

or

$$\omega_\pm=\frac{i}{2}\{-\alpha\pm\sqrt{\alpha^2+4\beta}\}, \tag{2·14}$$

where

$$\alpha(k)=-\mathrm{tr}\,\hat{\Gamma}(0,\boldsymbol{k})=-\{K_{xx}+K_{yy}+(D_x+D_y)k^2\} \tag{2·15}$$

and

$$\beta(k)=-\det\hat{\Gamma}(0,\boldsymbol{k})=-(K_{xx}+D_xk^2)(K_{yy}+D_xk^2)+K_{xy}K_{yx}. \tag{2·16}$$

An instability occurs when the condition "$\alpha, \beta<0$ for all k" becomes violated. According to Tomita et al.,[6] one has two possibilities for the occrrence of an instability:

Case A (soft mode instability)

This type of instability occurs when β vanishes for a certain wave number k_c while α remains negative. It is obvious that $\omega_-(k_c)$ vanishes at the marginal situation. The wave number k_c is the one which maximizes β, that is,

$$k_c^2=\mathrm{Max}[0,\ -(K_{xx}D_y+K_{yy}D_x)/2D_xD_y]. \tag{2·17}$$

The critical condition thus becomes

$$(K_{xx}D_y-K_{yy}D_x)^2+4K_{xy}K_{yx}D_xD_y=0, \quad (k_c\neq0) \tag{2·18}$$

$$K_{xx}K_{yy}-K_{xy}K_{yx}=0. \quad (k_c=0) \tag{2·19}$$

The case $k_c\neq0$ is called the Turing instability[1] which leads to a dissipative space structure.

Case B (hard mode instability)

This type of instability occurs when α vanishes for a certain wave number while β remains negative. Since the diffusion constants are non-negative, the critical wave number is zero. Hence at the marginal situation we have

$$K_{xx}+K_{yy}=0 \tag{2·20}$$

and the eigenvalues are

$$\omega_{\pm}=\pm i\omega_0\,, \tag{2·21}$$

where

$$\omega_0=\sqrt{K_{xx}K_{yy}-K_{xy}K_{yx}}\,. \tag{2·22}$$

Clearly, two modes become unstable simultaneously in this case. The non-vanishing frequency at the marginal situation leads to a temporal organization beyond the instability point.

§ 3. Reduction of evolution equation

In this section we shall make a reduction of Eq. (2·5) near the instability points. The two cases A and B may be treated in a parallel way. As to the former case, however, we shall restrict ourselves to the one-dimensional Turing instability because the applicability of the present method to the other kinds of soft mode instability seems questionable as we shall discuss at the end of this section.

Let us suppose that an instability occurs when a set of parameters $\boldsymbol{\lambda}\equiv(K_{\alpha\beta}, D_\alpha, \cdots)$ takes the value $\boldsymbol{\lambda}_0$. Near the instability point we put

$$\boldsymbol{\lambda}=\boldsymbol{\lambda}_0+\varepsilon^2\boldsymbol{\lambda}_1\,, \tag{3·1}$$

where ε is a small quantity. To save notations, let $(K_{\alpha\beta}, D_\alpha, \cdots)$ represent $\boldsymbol{\lambda}_0$. Thus, near the instability point, we have only to make the following substitution in (2·5):

$$\begin{pmatrix} K_{\alpha\beta} \\ D_\alpha \\ \vdots \end{pmatrix} \to \begin{pmatrix} K_{\alpha\beta} \\ D_\alpha \\ \vdots \end{pmatrix} + \varepsilon^2 \begin{pmatrix} \gamma_{\alpha\beta} \\ d_\alpha \\ \vdots \end{pmatrix}. \tag{3·2}$$

The reduction scheme presented below is quite similar to the one adopted by Newell and Whitehead[7] in the problem of the Bénard convection. From the expression for the eigenvalue ω one may easily find that the new characteristic time- and length-scales of the order ε^{-2} and ε^{-1}, respectively, appear near the instability point of the type either A or B. Therefore, it is natural to describe the dynamics in terms of the new variables T and $\boldsymbol{R}$ defined by

$$T=\varepsilon^2\xi t \qquad (\xi>0) \tag{3·3}$$

and

$$\boldsymbol{R}=\varepsilon\chi\boldsymbol{r}. \qquad (\chi=\text{real}) \tag{3·4}$$

Here ξ and χ are constants which will be chosen later in such a way that the final equation of motion may take a simplest form. To make a unified treatment of the cases A and B, it is convenient to introduce a notation f_ν by

$$f_\nu=\begin{cases} e^{i\nu k_c r} & \text{for Case } A, \\ e^{i\nu\omega_0 t} & \text{for Case } B. \end{cases} \tag{3·5}$$

Then, in either case one has from the linear theory

$$\boldsymbol{x}=\boldsymbol{\alpha}f_1+\boldsymbol{\alpha}^*f_{-1} \tag{3·6}$$

at the instability point, where $\boldsymbol{\alpha}$ is a constant vector with a certain orientation but an arbitrary length. As one goes slightly beyond the instability point, the expression (3·6) should be modified due to the appearance of slow variation in space and time such as the one described in terms of $\boldsymbol{R}$ and T. This effect may be taken into account by regarding $\boldsymbol{\alpha}$ as a function of $\boldsymbol{R}$ and T. Equation (3·6) should further be modified due to the small contribution from harmonics other than $f_{\pm1}$. On the other hand, the characteristic amplitude of $\boldsymbol{x}$ near the instability point is expected to be of order ε by the analogy of the classical theory of the second-order phase transition. From these arguments we expect that $\boldsymbol{x}$ may be expanded near the instability point as

$$\boldsymbol{x}=\sum_{n=1}^{\infty}\varepsilon^n\boldsymbol{x}_n, \tag{3·7}$$

where

$$\boldsymbol{x}_n=\sum_{\nu=-\infty}^{\infty}\boldsymbol{x}_n^{(\nu)}(T,\boldsymbol{R})f_\nu. \tag{3·8}$$

Accordingly, the operators $\partial/\partial t$ and $\nabla_{\boldsymbol{r}}$ appearing in (2·5) may be replaced by

$$\frac{\partial}{\partial t}+\varepsilon^2\xi\frac{\partial}{\partial T} \tag{3·9}$$

and

$$\nabla_{\boldsymbol{r}}+\varepsilon\chi\nabla_{\boldsymbol{R}}, \tag{3·10}$$

respectively. Substituting (3·2), (3·7), (3·9) and (3·10) into $\hat{\Gamma}$, $\mathcal{L}$, $\hat{L}$ and $\boldsymbol{G}$, one may express these quantities in the following expansion forms.

$$\hat{\Gamma}=\hat{\Gamma}_0+\varepsilon\hat{\Gamma}_1+\cdots, \tag{3·11a}$$

$$\mathcal{L}=\mathcal{L}_0+\varepsilon\mathcal{L}_1+\varepsilon^2\mathcal{L}_2+\cdots, \tag{3·11b}$$

$$\hat{L}=\hat{L}_0+\varepsilon\hat{L}_1+\cdots, \tag{3·11c}$$

$$\boldsymbol{G}=\varepsilon^2\boldsymbol{G}_2+\varepsilon^3\boldsymbol{G}_3+\cdots. \tag{3·11d}$$

The explicit forms of a few terms in each expansion are as follows:

$$\hat{\Gamma}_0=\begin{pmatrix} \dfrac{\partial}{\partial t}+K_{xx}-D_x\nabla_{\boldsymbol{r}}^2 & K_{xy} \\ K_{yx} & \dfrac{\partial}{\partial t}+K_{yy}-D_y\nabla_{\boldsymbol{r}}^2 \end{pmatrix}, \tag{3·12}$$

$$\mathcal{L}_0=\left(\frac{\partial}{\partial t}+K_{xx}-D_x\nabla_{\boldsymbol{r}}^2\right)\left(\frac{\partial}{\partial t}+K_{yy}-D_y\nabla_{\boldsymbol{r}}^2\right)-K_{xy}K_{yx}, \tag{3·13}$$

$$\mathcal{L}_1=-2\chi\nabla_{\boldsymbol{r}}\nabla_{\boldsymbol{R}}\left\{D_y\left(\frac{\partial}{\partial t}+K_{xx}\right)+D_x\left(\frac{\partial}{\partial t}+K_{yy}\right)-2D_xD_y\nabla_{\boldsymbol{r}}^2\right\}, \tag{3·14}$$

$$\mathcal{L}_2=\xi\mathcal{A}_1\frac{\partial}{\partial T}-\mathcal{A}_2-\chi^2\mathcal{A}_3\nabla_{\boldsymbol{R}}^2, \tag{3·15}$$

where

$$\mathcal{A}_1=2\frac{\partial}{\partial t}+K_{xx}+K_{yy}-(D_x+D_y)\nabla_{\boldsymbol{r}}^2, \tag{3·16}$$

$$\mathcal{A}_2=-\left\{\left(\frac{\partial}{\partial t}+K_{xx}-D_x\nabla_{\boldsymbol{r}}^2\right)(\gamma_{yy}-d_y\nabla_{\boldsymbol{r}}^2)+\left(\frac{\partial}{\partial t}+K_{yy}-D_y\nabla_{\boldsymbol{r}}^2\right)\right.$$
$$\left.\times(\gamma_{xx}-d_x\nabla_{\boldsymbol{r}}^2)-(K_{yx}\gamma_{xy}+K_{xy}\gamma_{yx})\right\} \tag{3·17}$$

and

$$\mathcal{A}_3=D_y\left(\frac{\partial}{\partial t}+K_{xx}\right)+D_x\left(\frac{\partial}{\partial t}+K_{yy}\right)-6D_xD_y\nabla_{\boldsymbol{r}}^2; \tag{3·18}$$

$$\hat{L}_0=\begin{pmatrix} \dfrac{\partial}{\partial t}+K_{yy}-D_y\nabla_{\boldsymbol{r}}^2 & -K_{xy} \\ -K_{yx} & \dfrac{\partial}{\partial t}+K_{xx}-D_x\nabla_{\boldsymbol{r}}^2 \end{pmatrix}, \tag{3·19}$$

$$\hat{L}_1=-2\chi\nabla_{\boldsymbol{r}}\nabla_{\boldsymbol{R}}\begin{pmatrix} D_y & 0 \\ 0 & D_x \end{pmatrix}, \tag{3·20}$$

$$\boldsymbol{G}_{2,3}=\begin{pmatrix} P_{2,3} \\ Q_{2,3} \end{pmatrix}, \tag{3·21}$$

where

$$R_2=\boldsymbol{x}_1{}^t\hat{R}_2\boldsymbol{x}_1 \tag{3·22}$$

and

$$R_3=2\boldsymbol{x}_1{}^t\hat{R}_2\boldsymbol{x}_2+\boldsymbol{x}_1{}^t\hat{R}_3\hat{x}_1\boldsymbol{x}_1. \tag{3·23}$$

For the sake of brevity, we further introduce some notations. Let $\boldsymbol{G}_n{}^{(\nu)}$ and $R_n{}^{(\nu)}$ denote the coefficients of expansion of $\boldsymbol{G}_n$ and R_n in various harmonics:

$$\boldsymbol{G}_n=\sum_\nu \boldsymbol{G}_n{}^{(\nu)}f_\nu, \tag{3·24}$$

$$R_n=\sum_\nu R_n^{(\nu)}f,\tag{3·25}$$

Regarding $\mathcal{L}_n$ as a function of $-i(\partial/\partial t)$ and $-i\nabla_r$ like

$$\mathcal{L}_n=\mathcal{L}_n\left(-i\frac{\partial}{\partial t},\ -i\nabla_r\right),\tag{3·26}$$

let us further define a quantity $\mathcal{L}_n(\nu)$ by

$$\mathcal{L}_n(\nu)=\begin{cases}\mathcal{L}_n(0,\ \nu k_c) & \text{for Case } A,\\ \mathcal{L}_n(\nu\omega_0,\ 0) & \text{for Case } B.\end{cases}\tag{3·27}$$

In a similar manner we define quantities $\hat{L}_n(\nu)$, $\hat{\Gamma}_n(\nu)$ and $\mathcal{A}_j(\nu)$.

In the lowest order in ε, Eq. (2·5) reduces to

$$\hat{\Gamma}_0\boldsymbol{x}_1=0,\tag{3·28}$$

which yields the neutral solution

$$\boldsymbol{x}_1^{(1)}=x_1^{(-1)*}=\boldsymbol{a}\zeta W(T,\ \boldsymbol{R}),$$
$$\boldsymbol{x}_1^{(\nu)}=0 \text{ for } \nu\neq\pm1,\tag{3·29}$$

where

$$\boldsymbol{a}=\begin{pmatrix}1\\a\end{pmatrix},$$

$$a=-\hat{\Gamma}_0(1)_{xx}/\hat{\Gamma}_0(1)_{xy}=-\hat{\Gamma}_0(1)_{yx}/\hat{\Gamma}_0(1)_{yy}$$
$$=\hat{L}_0(1)_{yy}/\hat{L}_0(1)_{xy}=\hat{L}_0(1)_{yx}/\hat{L}_0(1)_{xx},\tag{3·30}$$

and ζ is, in general, a parameter of complex number; the role of ζ is similar to that of ξ and χ. What we have to do below is to find an evolution equation for W which determines the dynamics near the instability point.

The second- and third-order balance equations from (2·11) are given by

$$\mathcal{L}_0\boldsymbol{x}_2+\mathcal{L}_1\boldsymbol{x}_1=\hat{L}_0\boldsymbol{G}_2\tag{3·31}$$

and

$$\mathcal{L}_0\boldsymbol{x}_3+\mathcal{L}_1\boldsymbol{x}_2+\mathcal{L}_2\boldsymbol{x}_1=\hat{L}_0\boldsymbol{G}_3+\hat{L}_1\boldsymbol{G}_2,\tag{3·32}$$

respectively. Equation (3·31) enables us to express $\boldsymbol{x}_2$ in terms of W as follows. Putting (3·29) into (3·22), one finds

$$R_2^{(0)}=2|\zeta|^2|W|^2\boldsymbol{a}^{*t}\hat{R}_2\boldsymbol{a}\tag{3·33}$$

and

$$R_2^{(2)}=R_2^{(-2)*}=\zeta^2W^2\boldsymbol{a}^t\hat{R}_2\boldsymbol{a}.\tag{3·34}$$

These equations together with the fact that $\mathcal{L}_1\boldsymbol{x}_1=0$ are sufficient to obtain the following expression for $\boldsymbol{x}_2^{(\nu)}$ from (3·31):

$$x_2^{(0)}=2|\zeta|^2\mathcal{L}_0(0)^{-1}\hat{L}_0(0)|W|^2\begin{pmatrix} a^{*t}\hat{P}_2 a \\ a^{*t}\hat{Q}_2 a \end{pmatrix}, \tag{3·35}$$

$$x_2^{(2)}=x_2^{(-2)*}=\zeta^2\mathcal{L}_0(2)^{-1}\hat{L}_0(2)W^2\begin{pmatrix} a^t\hat{P}_2 a \\ a^t\hat{Q}_2 a \end{pmatrix} \tag{3·36}$$

and

$$x_2^{(\nu)}=0 \text{ if } \nu\neq 0,\ \pm 2,\ \pm 1. \tag{3·37}$$

The quantity $x_2^{(\pm1)}$ cannot be determined at this stage, which does not cause any trouble as one sees below.

Let us consider the ε^3-balance equation (3·32) from which the equation for W may be determined. As to the fundamental wave component, Eq. (3·32) separately yields the balance equation

$$\mathcal{L}_2(1)x_1^{(1)}=\hat{L}_0(1)G_3^{(1)}, \tag{3·38}$$

or, written explicitly,

$$\mathcal{L}_2(1)x_1^{(1)}=\hat{L}_0(1)_{xx}P_3^{(1)}+\hat{L}_0(1)_{xy}Q_3^{(1)} \tag{3·39}$$

and

$$\mathcal{L}_2(1)y_1^{(1)}=\hat{L}_0(1)_{yx}P_3^{(1)}+\hat{L}_0(1)_{yy}Q_3^{(1)} \tag{3·40}$$

where we have used the facts that $\mathcal{L}_0(1)=\mathcal{L}_1(1)=0$ and that G_2 cannot yield fundamental wave. From the neutral solution (3·29) and the first nonlinear correction (3·35)∼(3·37) one obtains

$$\begin{aligned}R_3^{(1)}=|\zeta|^2\zeta|W|^2W\Bigg[&4\mathcal{L}_0^{-1}(0)a^t\hat{R}_2\hat{L}_0(0)\begin{pmatrix} a^{*t}\hat{P}_2 a \\ a^{*t}\hat{Q}_2 a \end{pmatrix}\\ &+2\mathcal{L}_0^{-1}(2)a^{*t}\hat{R}_2\hat{L}_0(2)\begin{pmatrix} a^t\hat{P}_2 a \\ a^t\hat{Q}_2 a \end{pmatrix}\\ &+2a^t\hat{R}_3\hat{a}a^*+a^{*t}\hat{R}_3\hat{a}a\Bigg],\end{aligned} \tag{3·41}$$

where

$$\hat{a}\equiv\begin{pmatrix} 1 & 0 \\ 0 & a \end{pmatrix}. \tag{3·42}$$

Substituting (3·29) and (3·41) into (3·39), we finally find

$$\mathcal{L}_2(1)W=-|\zeta|^2 g|W|^2W, \tag{3·43}$$

where

$$g=-\Bigg[4\mathcal{L}_0^{-1}(0)a^t\hat{M}_2\hat{L}_0(0)\begin{pmatrix} a^{*t}\hat{P}_2 a \\ a^{*t}\hat{Q}_2 a \end{pmatrix}$$

$$+2\mathcal{L}_0^{-1}(2)\boldsymbol{a}^{*t}\widehat{M}_2\widehat{L}_0(2)\begin{pmatrix}\boldsymbol{a}^t\widehat{P}_2\boldsymbol{a}\\ \boldsymbol{a}^t\widehat{Q}_2\boldsymbol{a}\end{pmatrix}$$

$$\left.+2\boldsymbol{a}^t\widehat{M}_3\hat{\boldsymbol{a}}\boldsymbol{a}^*+\boldsymbol{a}^{*t}\widehat{M}_3\hat{\boldsymbol{a}}\boldsymbol{a}\right], \tag{3·44}$$

$$\widehat{M}_{2,3}\equiv\widehat{L}_0(1)_{xx}\widehat{P}_{2,3}+\widehat{L}_0(1)_{xy}\widehat{Q}_{2,3}\,. \tag{3·45}$$

It is easy to confirm that Eq. (3·40) yields an equation identical to (3·43). To see this one has only to prove

$$\widehat{M}_{2,3}=a^{-1}\widehat{N}_{2,3}\,, \tag{3·46}$$

where

$$\widehat{N}_{2,3}=\widehat{L}_0(1)_{yx}\widehat{P}_{2,3}+\widehat{L}_0(1)_{yy}\widehat{Q}_{2,3}\,. \tag{3·47}$$

The equality (3·46) directly follows from Eq. (3·30) and the fact that

$$\det\widehat{L}_0(1)=\det\widehat{\Gamma}_0(1)=0\,. \tag{3·48}$$

By the use of (3·15)∼(3·18) one may rewrite Eq. (3·43) in the form

$$\frac{\partial W}{\partial T}=\xi^{-1}\{\tilde{\gamma}+\chi^2\widetilde{D}\nabla_{\boldsymbol{R}}^2-|\zeta|^2\tilde{g}|W|^2\}W\,. \tag{3·49}$$

Here $\mathrm{Re}\,\tilde{\gamma}>0$ and $\mathrm{Re}\,\tilde{\gamma}<0$ correspond to the post-critical and sub-critical situations, respectively. The coefficients $\tilde{\gamma}$, $\widetilde{D}$ and $\tilde{g}$ have the following expressions for respective types of instability.

Case A

$$\tilde{\gamma}=-\{(K_{xx}+D_xk_c^2)(\gamma_{yy}+d_yk_c^2)+(K_{yy}+D_yk_c^2)(\gamma_{xx}+d_xk_c^2)$$
$$-(K_{yx}\gamma_{xy}+K_{xy}\gamma_{yx})\}/\{K_{xx}+K_{yy}+(D_x+D_y)k_c^2\}, \tag{3·50}$$

$$\widetilde{D}=-2(D_yK_{xx}+D_xK_{yy})/\{K_{xx}+K_{yy}+(D_x+D_y)k_c^2\}>0\,, \tag{3·51}$$

$$\tilde{g}=g/\{K_{xx}+K_{yy}+(D_x+D_y)k_c^2\}. \tag{3·52}$$

The quantities appearing in g are given as follows.

$$\mathcal{L}_0(0)=K_{xx}K_{yy}-K_{xy}K_{yx}\,, \tag{3·53}$$

$$\mathcal{L}_0(2)=(K_{xx}+4D_xk_c^2)(K_{yy}+4D_yk_c^2)-K_{xy}K_{yx}\,, \tag{3·54}$$

$$\widehat{L}_0(0)=\begin{pmatrix}K_{yy} & -K_{xy}\\ -K_{yx} & K_{xx}\end{pmatrix}, \tag{3·55}$$

$$\widehat{L}_0(2)=\begin{pmatrix}K_{yy}+4D_yk_c^2 & -K_{xy}\\ -K_{yx} & K_{xx}+4D_xk_c^2\end{pmatrix}, \tag{3·56}$$

$$\widehat{M}_{2,3}=(K_{yy}+D_yk_c^2)\widehat{P}_{2,3}-K_{xy}\widehat{Q}_{2,3}\,, \tag{3·57}$$

$$a=-\frac{K_{xx}+D_xk_c^2}{K_{xy}}=-\frac{K_{yx}}{K_{yy}+D_yk_c^2}\,. \tag{3·58}$$

Case B

$$\tilde{\gamma}=-\frac{1}{2}(\gamma_{xx}+\gamma_{yy})+\frac{i}{2\omega_0}\{K_{xx}(\gamma_{yy}-\gamma_{xx})$$

$$-(K_{xy}\gamma_{yx}+K_{yx}\gamma_{xy})\}, \tag{3·59}$$

$$\tilde{D}=D_{+}+\frac{iK_{xx}}{\omega_0}D_{-}, \tag{3·60}$$

$$D_{\pm}\equiv\frac{1}{2}(D_x\pm D_y), \tag{3·61}$$

$$\tilde{g}=-ig/2\omega_0; \tag{3·62}$$

$$\mathcal{L}_0(2)=(2i\omega_0+K_{xx})(2i\omega_0+K_{yy})-K_{xy}K_{yx}, \tag{3·63}$$

$$\hat{L}_0(2)=\begin{pmatrix} 2i\omega_0+K_{yy} & -K_{xy} \\ -K_{yx} & 2i\omega_0+K_{xx} \end{pmatrix}, \tag{3·64}$$

$$\hat{M}_{2,3}=(i\omega_0+K_{yy})\hat{P}_{2,3}-K_{xy}\hat{Q}_{2,3}, \tag{3·65}$$

$$a=-\frac{i\omega_0+K_{xx}}{K_{xy}}=-\frac{K_{yx}}{i\omega_0+K_{yy}}. \tag{3·66}$$

By definition the quantities $\mathcal{L}_0(0)$ and $\hat{L}_0(0)$ have expressions identical to (3·53) and (3·55) respectively.

The expressions above show that all the coefficients in *Case A* are real while they are complex in *Case B*. This is because in *Case A* the operator ∇_r appears only through ∇_r^2 giving the real contribution k_c^2 in contrast to *Case B* where the operation $\partial/\partial t$ yields the imaginary contribution $i\omega_0$.

The condition $\mathrm{Re}\,\tilde{g}>0$ is necessary in order that the amplitude W may not diverge. If this condition is violated, the phase transition will be of a discontinuous type and the present method is not applicable. Assuming $\mathrm{Re}\,\tilde{g}>0$ and choosing the parameters ξ, χ and ζ as

$$\xi=|\mathrm{Re}\,\tilde{\gamma}|, \tag{3·67}$$

$$\chi^2=|\mathrm{Re}\,\tilde{\gamma}|/\mathrm{Re}\,\tilde{D} \tag{3·68}$$

and

$$|\zeta|^2=|\mathrm{Re}\,\tilde{\gamma}|/\mathrm{Re}\,\tilde{g}, \tag{3·69}$$

one may further reduce Eq. (3·49) to

$$\frac{\partial W}{\partial T}=\{(\pm 1+ic_0)+(1+ic_1)\nabla_R^2-(1+ic_2)|W|^2\}W, \tag{3·70}$$

where

$$c_0=\mathrm{Im}\,\tilde{\gamma}/|\mathrm{Re}\,\tilde{\gamma}|, \tag{3·71}$$

$$c_1=\mathrm{Im}\,\tilde{D}/\mathrm{Re}\,\tilde{D} \tag{3·72}$$

and

$$c_2 = \mathrm{Im}\,\tilde{g}/\mathrm{Re}\,\tilde{g}\,. \tag{3·73}$$

In *Case A* we have $c_0=c_1=c_2=0$. In *Case B* one may eliminate c_0 by transforming Eq. (3·70) into the equation for $\widetilde{W}$ defined by

$$\widetilde{W} = We^{-ic_0 T}\,. \tag{3·74}$$

That is, one gets

$$\frac{\partial \widetilde{W}}{\partial T} = \{\pm 1 + (1+ic_1)\nabla_{\boldsymbol{R}}^2 - (1+ic_2)\,|\widetilde{W}|^2\}\,\widetilde{W}\,. \tag{3·75}$$

Finally we give the reason why we have excluded from our consideration the cases of two- and three-dimensional space order and also the case in which a soft mode with $k=0$ becomes unstable. In two- and three-demensions the critical wave vector $\boldsymbol{k}_c$ has an arbitrariness in its orientation. In particular, one may always find a set of critical wave vector satisfying the relation

$$\boldsymbol{k}_{c1} + \boldsymbol{k}_{c2} + \boldsymbol{k}_{c3} = 0\,. \tag{3·76}$$

This fact makes the ε^2 balance equation (3·31) meaningless because the fundamental wave, e.g., $e^{-i\boldsymbol{k}_{c1}\boldsymbol{r}}$, appears on the right-hand side through the product of the neutral solutions corresponding to $\boldsymbol{k}_{c2}$ and $\boldsymbol{k}_{c3}$ while on the left-hand side the fundamental wave cannot appear due to the operation of $\mathcal{L}_0$ and $\mathcal{L}_1$. In the case of soft mode instability with $k_c=0$ one meets with the same kind of contradiction. When only a couple of order parameters corresponding to $\pm\boldsymbol{k}_c$ is essential to the emergence of space order, the above difficulty may be avoided since the problem then becomes essentially the same as that for one dimension.

§ 4. Some remarks

So far we have concentrated on deriving the reduced dynamical equations which are valid near the points of the two types of instability, namely, the time order and the one-dimensional space order. We have restricted ourselves to a two-component system so that the coefficients in the final equation (3·49) may be expressed explicitly in terms of the parameters appearing in the original equation (2·5). It is almost obvious, however, that such a restriction is not essential to our qualitative conclusion. In fact, from the analysis of a simplified model of n-component controlled biochemical system with negative feedback, we have obtained completely the same type of equation as (3·70), which will be reported elsewhere.

Equation (3·70) may be regarded as an idealized model for a self-organized system. The case of the temporal organization is of particular interest. In this case Eq. (3·70) describes the motion of an infinite number of self-sustained oscillators coupled to each other through the diffusion term with a complex diffusion coefficient. The motion of each nonlinear oscillator in the absence of the coupling is quite simple just as the motion of a harmonic oscillator. Such a simplified picture will surely provide a starting point with which the macroscopic theory of a wide variety of temporally organized system may be developed.

Finally we shall give a comment on an important role possibly played by the coefficients c_1 and c_2 in (3·75). Only the post-critical situation will be concerned below. Let us put

$$W=\rho e^{i\varphi}. \tag{4·1}$$

One may easily find that Eq. (3·75) has the spatially uniform solution

$$\rho_0^2=1, \quad \varphi_0=-c_2T, \tag{4·2}$$

which represents a limit cycle. That is, the concentrations X and Y which are uniform in space make a self-sustained oscillation about the point $\boldsymbol{X}_0$. We have now to ask whether this solution is stable against a spatially non-uniform perturbation. Assuming the space-time dependence of the perturbation around (4·2) as $\exp(i\Omega T+i\boldsymbol{KR})$, we find from the linearized perturbation equation, that

$$\Omega_\pm=i\{(1+K^2)\pm\sqrt{D}\}, \quad D=(1+K^2)^2-2(1+c_1c_2)K^2-(1+c_1^2)K^4. \tag{4·3}$$

It is clear from this expression that the solution (4·2) is unstable if

$$1+c_1c_2<0 \tag{4·4}$$

since $i\Omega_-$ becomes positive for sufficiently small K. It may easily be confirmed from the results of previous work[5] that the condition (4·4) can actually be satisfied by the Prigogine-Lefever-Nicolis model. Beyond the instability a certain spatial pattern which is oscillating in time is expected to appear. However, the detailed analysis of this problem will be given elsewhere.

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