MATHEMATICAL MODELING OF BIOSYSTEMS

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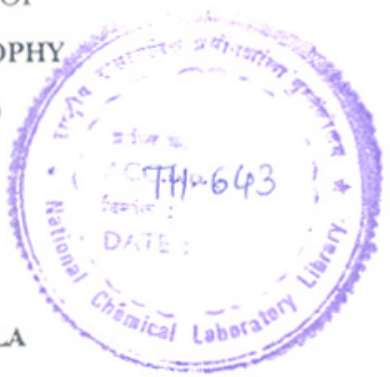

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Certified that the work incorporated in the thesis " **Mathematical Modeling of Biosystems**" submitted by Rajani R. Pillutla was carried out by the candidate under my supervision. Such materials as has been obtained from other sources has been duly acknowledged in the thesis.

B. D. Kulkarni

Research Guide

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CONTENTS

CHAPTER I

INTRODUCTION

1.1 Introduction

Research endeavors aimed at constructing a rational basis for understanding how living organisms function and interact have a long history (Hinshelwood, 1946). However, the complexity of living systems with their multitude of hierarchies in time, space as well as structures, makes the task of understanding biological processes in terms of physico-chemical principles, a formidable one. Nevertheless, interdisciplinary areas aimed at such studies are by now well established and the importance of the contribution from these fields to biology cannot be emphasized enough. Mathematical models in particular have produced spectacular and exciting results. Several treatises, on the application of mathematical models in biology are found in literature (Gold, 1977, Segel, 1981).

One of the most striking and intriguing aspects of natural phenomena is that, complex systems involving a large number of units can interact in a cooperative way to form and maintain spatio-temporal structures and can show self-organization at various levels (Eigen, 1971, Peacocke, 1983). This observation was seen to be at variance with the second law of thermodynamics, which states that all natural process should evolve with a net increase in entropy. Hence the need arose for new conceptual tools and ways of analyzing and modelling biological complexity. In the beginning of this century, attempts were made to extend the principles of thermodynamics to irreversible processes. A striking result of these attempts, pioneered by Ilya Prigogine of the Brussels School, was to prove that transition to ordered behavior can occur in systems far from equilibrium and which are open to exchange of matter and energy (Nicolis and Prigogine, 1977). Systems which can give rise to and maintain order at the cost of constant flow of matter and energy are known as dissipative systems and the ordered structures so formed, as dissipative structures. It is easy to see that biological systems are indeed dissipative systems, since they consist of a large number of interacting subunits, each operating far from equilibrium. The patterns observed in these dissipative systems

generally arise due to the fact that natural laws are inherently nonlinear. The study of these systems takes us from the classical thermodynamic approach to another universal discipline, *synergetics*. A brief overview of this newly emerging field is given in the next section.

Another major contribution to understanding biological organization has come from the field of chemical kinetics. In particular reactions involving feedback and autocatalysis are found to be prevalent in many biological systems. To explain the origin of oscillations in biological systems, many mechanisms have been proposed and modelled in terms of first order nonlinear ordinary differential equations (Peacocke, 1983). Beginning with Turing's classical work which includes diffusional effects, many reaction-diffusion models have also been proposed and described by partial differential equations. An important feature of these models is that, their solutions are capable of showing great sensitivities to variations in parameters, implying that for a small change in one of the parameters, the system loses its stability and evolves to a new state.

Thus bifurcation, and the behavior of the system near the instability point, are of prime importance to any system showing self-organization. In the present thesis, we study both these aspects for two systems of biological interest, namely, the exponential autocatalysis model and the substrate-inhibition kinetic model. The exponential autocatalysis model takes into account the exponential dependance of the rate constant on one of the reaction intermediates. This model is helpful in understanding many biological processes, significantly, the allosteric enzymatic reaction mechanisms. The substrate-inhibition kinetic model, is also known as the Seelig's oscillator. It is shown to produce oscillatory solutions and wave patterns in realistic parameter regimes. In order to obtain the bifurcation maps of these systems we resort to the well established linear stability analysis and for the second aspect we turn to synergetics.

1.2 From Thermodynamics to Synergetics

By definition synergetics means a study of the cooperation of subsystems of a larger system which produces macroscopic spatial, temporal and functional structures (Haken, 1989).

Such systems are found in diverse fields of science and they all show similar behavior, especially near the instability points in the system, irrespective of the field to which they belong. Thus we find typical examples of synergetic systems in lasers, hydrodynamics, solid state physics, neural networks, mechanical engineering, certain chemical reactions, morphogenesis, behavioral biology, evolution of living systems, population dynamics and even economics and sociology.

One of the major concepts underlying synergetics is the application of the *slaving principle* near the bifurcation points. This principle basically claims the responsibility of eliminating a large number of rapidly decaying modes. Further the decaying modes which can be eliminated are known as the *slaved parameters* and the linearly growing modes as the *order parameters*. Having eliminated the fast modes, we can now describe the system behavior in terms of a simple evolution equation, known as the *Ginzburg-Landau equation*. Much of the work presented in this thesis involves this equation with a view to apply it to biologically relevant schemes. The two models referred to earlier have been investigated for their bifurcation behavior, the possible dynamic features that can arise in these systems and the stability of these dynamic features. The mathematical methods employed in this study are the linear stability analysis, the multi-time scale theory, the reductive and singular perturbation techniques. Extensive numerical analysis has also been carried out wherever necessary. In the next section we give a more detailed outline of the contents of the present thesis.

1.3 Contents

In Chapter II an extensive bifurcation analysis of the exponential autocatalysis model has been carried out, with a view to obtain its rich bifurcation behavior. In this chapter, the linear stability analysis of this model is carried out in the absence of diffusion. Further on the basis of the properties of the Jacobian matrix of the system, the equations for the locus of two types of instabilities have been derived. A thorough numerical analysis has been carried out to scan the different parameter regimes for the possible behavior. The results for the global

stability behavior based on these conditions have also been presented and discussed.

Biological processes are known to involve a vast number of reactions which evolve on widely different time scales. In such situations one can obtain an approximation of the asymptotic behavior by eliminating the fast reactions. The principles of the multi-time scale theory presented in Chapter III allow us to separate the fast and slow variables and further to construct the asymptotic behavior of the system and analyze the stability of the evolving structures. In the third chapter we apply the two-time scales (singular perturbation) to the exponentially autocatalyzed reaction-diffusion model, for a defined set of initial and boundary conditions. The global nonuniform steady patterns and limit cycle behavior are obtained and the stability of these structures is analyzed.

In Chapter IV we make use of the principles of synergetics with view to obtaining a simplified description of reaction-diffusion system near the Hopf-bifurcation point, with the help of the Ginzburg-Landau equation. The method of deducing this equation form is presented and demonstrated for the specific case of exponential autocatalysis. This chapter serves as the beginning point for the analysis carried out in the rest of the chapters.

In the Chapter V the possibility of the application of the GL equation to the Seelig's oscillator, in the realistic parameter regimes is explored. The instability conditions for two well known types of instabilities are derived. Further we derive the GL equation for this model and analyze the system behavior near the two instability points, mentioned above. To this end, an extensive numerical analysis of the GL equation is carried out in these two regions. Some interesting results of this analysis are presented and discussed in detail.

It is well recognized that in systems operating far from equilibrium fluctuations play an important role in bringing about order. In particular, the fluctuations are seen to amplify near the critical points in the system. Therefore it becomes necessary to investigate effect of fluctuations near these points. In the Chapter VI we obtain an equation of the form of the GL equation for systems evolving in presence of external noise. Here, we make use of the

Langevin-equation approach to include the effect of noise. This equation can be easily extended to a general class of fluctuations and is crucial to the analysis of a variety of interesting situations. The point of importance of the present chapter is that, here we extend the principles of derivation of the GL equation to stochastic systems in a simple way. This method is demonstrated for the case of substrate-inhibition kinetics.

Another application of the reduction principles of the previous chapters is seen in the case of weakly coupled oscillators. When the coupling is in the form of diffusion then it is seen that the behavior of the coupled system can be obtained in terms of a non-linear phase diffusion equation. In Chapter VII we aim at deducing this equation and extending it to the case of Ginzburg-Landau oscillators. The condition for the stability of the limit cycle solutions against diffusional effects is obtained in terms of the constants of the GL equation. In this chapter, the stability of the limit cycle solutions to the substrate-inhibition model is investigated, with the help of the non-linear phase diffusion equation.

Chapter VIII while does not prevent any new result, takes a global view of the GL equation, with a view to critically analyzing its content and meaning. The physical relevance of the constants of the GL equation is also discussed in detail. A brief account of the applications of the GL equation to various other fields is presented with a view to bring forth the universality of this equation and its capacity to show a rich and varied behavior in different regimes. Finally the scope for the application of this equation to some biological problems is discussed.

Chapter IX concludes the thesis giving a brief summary of the work presented, and the results obtained.

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CHAPTER II

CHEMICAL INSTABILITIES AND BIFURCATIONS IN EXPONENTIAL AUTOCATALYSIS MODEL

The linear stability analysis of an autocatalytic model, termed as exponential autocatalysis has been carried out, with a view to obtain its rich bifurcation behavior. Further on the basis of the properties of the Jacobian matrix of the system, the equations for the locus of two types of instabilities have been derived. The numerical results for the global stability behavior based on these conditions have also been presented and discussed.

2.1 Introduction

Bifurcation implies, a qualitative change in the behavior of the system, as a consequence of a small change in some parameter. In systems undergoing bifurcations, we find that successive instabilities lead to highly ordered structures. A significant example is found in the field of developmental biology (Bonner, 1974); here the central problem is, how do cells differentiate from a single cell type to form tissues, organs, and patterns and eventually form the highly ordered structures that living beings are. Population dynamics, one of the oldest branches of mathematical biology also deals with various bifurcation phenomena (May, 1974). Other important examples are found in the study of metabolic processes. Among these, glycolytic oscillations, oscillations in photosynthesis, circadian rhythms and oscillating membrane phenomena are some well studied examples (Hess and Boiteux, 1971). For an useful review of bifurcations in chemically and biochemically reacting systems, one can refer to Peacocke (1983), and the references cited therein. Thus we find that bifurcation phenomena are found in a variety of biological systems, and their implications are profound and interesting. The bifurcation theories for systems showing temporal organization (ODEs) are relatively well established (Minorsky, 1962, Murray, 1977, Segel, 1981, Eckmann, 1981, Hofstadter, 1981).

Turing in his seminal paper had suggested that morphogenesis can be understood as a reaction diffusion process (Turing, 1952), which forms the basis of the present day theories of self-organization and pattern formation. His theory leads to the formulation of the problem in terms of partial differential equations (PDEs). The questions regarding existence, and stability of solutions to such systems, and hence a need for renewed mathematical analysis arise naturally. The application of bifurcation theories to such systems have been reported by various authors (Joseph and Sattinger, 1972, Auchmuty and Nicolis, 1975, 1976, and Herchkowitz-Kaufmann, 1975). Owing to the advances in the systematic formulation of bifurcation theories, studies in instabilities and spatio temporal patterns in chemical and biological systems has emerged as an active area of research. We find numerous examples of applications of bifurcation theories to ODEs and PDEs (Nicolis, 1971, 1975, Ortoleva and Ross, 1975, Hess *et al*, 1975, Higgins,

1976, Fairen and Velarde, 1979, Schiffmann, 1980, Murray, 1981, Devreotes *et al*, 1983, Muller *et al*, 1985, Tyson and Keener, 1987 and Luo and Epstein, 1991). Recently, Turing patterns have been shown to occur in a chemical reactor (Castets *et al*, 1990), the theoretical framework for which has also been given (Lengyel and Epstein, 1991). Application of bifurcation theories in the analysis of models for pattern formation during cellular development and differentiation have also becone popular (Gerhardt *et al*, 1990a,b,c).

As already discussed, bifurcation analyses of various models for many physical, chemical, and biological systems have been attempted in the past. In the present chapter, we undertake the bifurcation analysis of an alternate form of autocatalysis, the exponential autocatalysis. This model will be discussed in detail in the following section. The linear stability analysis of this model has been carried out in absence of diffusion, to bring out its rich bifurcation behavior and identify the regions for the occurence of the chemical instabilities. The stability analysis of this model will be presented in Chapter IV.

2.2 The Model

Kinetic coupling between reactions can give various forms of feedback, leading to the appearance of a variety of dynamical features in chemically reacting systems (Frank, 1978). In open systems, autocatalysis and autoinhibition can produce positive or negative feedback effects. A systemic feedback is said to occur when a reaction product influences the rate constants of one of the steps leading to its own formation. When the rate constant increases under the influence of the product, then it is known as systemic autocatalysis. Such a mechanism is seen to be operating in reactions involving allosteric enzymes, or in the binding of O_2 to heamoglobin.

The exponential autocatalysis model originally proposed by RaviKumar *et al* (1984) is given as,

$$X \rightarrow Y \rightarrow Z$$

where the product Y shows a systemic autocatalytic effect on the rate constant k, such that k increases exponentially with Y. Thus it is also refered to as the exponential autocatalytic scheme. This rate form was proposed mainly to explain the strong nonlinearity, resemblance to Semenov type of law, and analogy to Arrhenius type of rate constant dependance found in certain systems. This rate form has wide applications in several biochemical systems as also in explaining the phenomena in diverse chemical and combustion type of reactions. The exponential autocatalysis has revealed the existence of multiplicity and oscillatory behavior under homogeneous conditions (RaviKumar *et al*, 1984). The exponential autocatalysis has received acceptance as a general model for class of reaction - diffusion systems (Bar Eli, 1984) and results obtained by using conventional autocatalysis such as the one used in Brusselator type models compare well with this model system. More recently the scheme in presence of diffusion was analysed with a view to establish bounds on the steady state solutions (Inamdar, 1990). The conditions for the existence of nonuniform solutions in the form of dissipative structures have also been derived analytically (Inamdar, 1990). In the following chapter, the global nonuniform patterns and limit cycle have been constructed for this model using multi-time scale analysis. In Chapter IV, the behavior near the Hopf bifurcation point has been derived using the reductive perturbation to obtain the description in terms of Ginzburg-Landau equation.

2.3 Linear Stability Analysis

The temporal kinetic scheme of the exponential autocatalysis is governed by the following equations,

$$\frac{dx}{dt} = x_o - x - Da_1 x \exp(\alpha y) \qquad (2.3.1a)$$

$$\frac{dy}{dt} = y_o - y + Da_1 x \exp(\alpha y) - Da_2 y \qquad (2.3.1b)$$

The steady state solutions of these equations are represented as (x_s, θ) and can be obtained as,

$$\exp(\alpha\theta) = \frac{(x_o - x_s)}{x_s Da_1}, \qquad \theta = \frac{x_o + y_o - x_s}{1 + Da_2} \tag{2.3.2}$$

Using Eq. (2.3.2), the steady state equation in terms of θ can be expressed in the form of a transcendental equation as,

$$Da_1 e^{\alpha\theta} - \frac{x_o}{x_o + y_o - \theta(1 + Da_2)} + 1 = 0 \tag{2.3.3}$$

To obtain the conditions for the occurrence of chemical instabilities we write, the Jacobian matrix of the system in Eq. (2.3.1) as,

$$A = \begin{pmatrix} -1 - Da_1 e^{\alpha\theta} & -\alpha x_s Da_1 e^{\alpha\theta} \\ Da_1 e^{\alpha\theta} & -(1 + Da_2) + \alpha x_s Da_1 e^{\alpha\theta} \end{pmatrix} \tag{2.3.4}$$

where the steady state relations given by Eq. (2.3.2) are used. The trace (T) and determinant (D) of this matrix are identified as,

$$T = (Da_1 e^{\alpha\theta})^2 + (Da_2 + 3 - \alpha x_o) e^{\alpha\theta} + (Da_2 + 2) \tag{2.3.5}$$

$$D = [(1 + Da_2)(1 + \alpha\theta) - \alpha(x_o + y_o)] Da_1 e^{\alpha\theta} + (1 + Da_2) \tag{2.3.6}$$

The characteristic polynomial for determining the eigenvalues for this two dimensional system is obtained from Eq. (2.3.4) as,

$$\lambda^2 - T\lambda + D = 0 \tag{2.3.7}$$

We choose Da_1 as the bifurcation parameter, since it contains externally controlled flow term and we express all our results in terms of this parameter. The critical value of Da_1 can be obtained by putting trace T equal to zero.

The condition for the secondary bifurcation (Kubicek and Marek, 1983) to occur is given as $\mathrm{Det}(\overline{A}) = 0$. This is stated as,

$$\overline{A} = \begin{pmatrix} \frac{\partial f_1}{\partial x} & \frac{\partial f_1}{\partial Da_1} \\ \frac{\partial f_2}{\partial x} & \frac{\partial f_2}{\partial Da_1} \end{pmatrix} = \begin{pmatrix} -1 - Da_1 e^{\alpha\theta} & -x_s e^{\alpha\theta} \\ Da_1 e^{\alpha\theta} & x_s e^{\alpha\theta} \end{pmatrix} \qquad (2.3.8)$$

and gives us the limit point for the bifurcation curve in the $Da_1 - Da_2$ plot. One solution to Eq. (2.3.8) results in $e^{\alpha\theta} = 0$, giving $\theta = -\infty$, which is unrealistic. Another solution yields,

$$\theta = \frac{x_o + y_o}{1 + Da_2} \qquad (2.3.9)$$

Here we would like to give the definitions of some terms based on the maginitudes and signs of T and D appearing in Eq. (2.3.7) and which are useful in the characterization of the regions we shall obtain later.

Saddle point: If $D < 0$, then, regardless of the sign of T, real parts of two eigen values have opposite signs. This means the eigenfunction associated with the positive eigen value grows exponentially in time, while the eigenfunction associated with the negative eigenvalue decays exponentially to zero. In this case, the steady state point is called saddle point.

Nodes: If $\Delta\ (= T^2 - 4D) > 0$, with $D > 0$, then both eigenvalues have the same sign, the sign of T. The steady state point then is called a node. A node can be stable or unstable. If $T > 0$, all nonzero solutions grow exponentially with time, so the steady state point is called unstable node.If $T < 0$, then both eigen values are negative. All the solutions the decay exponentially to steady state point. In this case steady state point is called stable node.

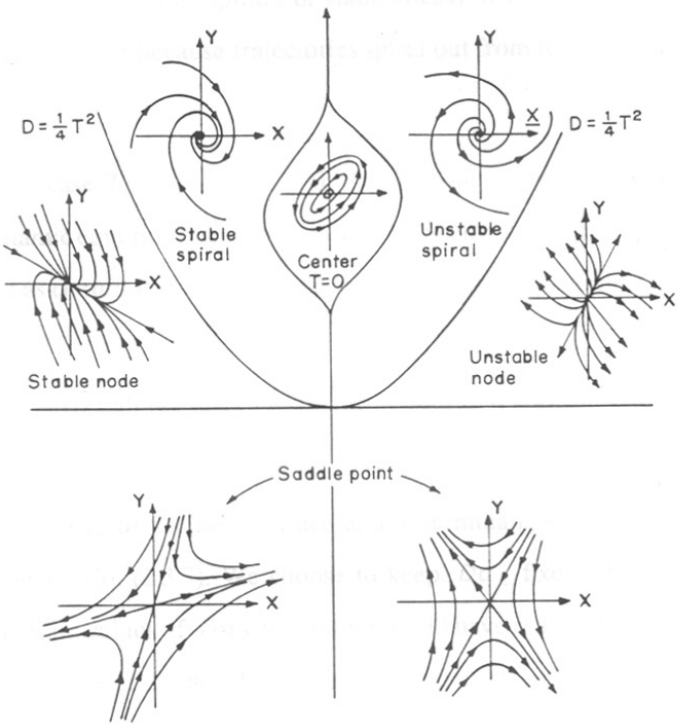

Fig. 2.1 : Diagram summarizing the various possible behaviors for a second order ODE

Spirals: If $\Delta < 0$, then the eigenvalues occur as a complex conjugate pair and the genaral solution oscillates with an amplitude envelopethat grows or decays as does exp $(Tt/2)$. Unless $T = 0$, the solution trajectories are spirals. If $T < 0$, the trajectories spiral to the steady state point, which is then called a stable spiral (or stable focus). If $T > 0$, then steady state is called an unstable spiral (or focus) because trajectories spiral out from it.

Center: In the case $T = 0$, and $D > 0$, the eigenvalues are purely imaginary and the solution of the dynamic equation becomes periodic in t. Such a steady state point is an imporatnt one and is termed as center. It is neutrally stable, and the solutions neither run away from, nor approach such a point.

Fig. 2.1 summarizes all the possible situations discussed above.

2.4 Chemical Instability results

Eq. (2.3.5) and (2.3.6) define the trace and determinant which help us to obtain the eigenvalues by solving Eq. (2.3.7). We choose to keep $D\alpha_2$ fixed while varying $D\alpha_1$ and observe that for certain values of $D\alpha_2$ the system would have real negative eigenvalues which coalesce as $D\alpha_1$ is increased. They then depart into a pair of eigenvalues which start moving in the complex plane, and for $D\alpha_1 = D\alpha_{1H}(D\alpha_2)$ they cross the imaginary axis which means that the steady state becomes unstable marking the onset of instability. Hard instability occurs when the real part of two complex eigenvalues becomes zero. On the other hand soft instability occurs when the imaginary part of a critical eigenvalue becomes zero. Thus for soft instability the circle traced out by the eigenvalues in the complex plane, lies entirely on the left of the imaginary axis. Along the instability line, for hard instability $D\alpha_{1H} = D\alpha_{1H}(D\alpha_2)$ in the $D\alpha_1 - D\alpha_2$ plane, the relaxation rate is purely imaginary (Daido and Tomita, 1979, Richter *et al*, 1980 and Richter *et al*, 1981). Equivalently one may state the condition for the occurrence of such instability as trace of the Jacobian matrix becoming zero with positive determinant.

This condition, valid for subcritical Hopf, also holds good for supercritical Hopf. Now, if the eigenvalues are given by $\lambda^* = \pm i\omega_H$, then the finite amplitude oscillation is characterized by a frequency ω_H, given by,

$$\omega_H = \sqrt{\Omega_1 \Omega_2 e^{\alpha\theta} - 1} \tag{2.4.1}$$

where Ω_1 is obtained by eliminating Da_1 from the steady state Eq. (2.3.3) and vanishing trace Eq. (2.3.5), while Ω_2 is extracted from the same equations by eliminating Da_2. These results are represented by two quadratic equations as below and Ω_1 and Ω_2 are the roots of these equations respectively.

$$Da_2^2\theta(1-\alpha\theta) + \{2\theta - (x_o + y_o) + \alpha\theta(x_o + 3y_o) - 2\alpha\theta^2\} Da_2 + \{[\alpha(x_o + y_o) + 1]\theta - \alpha y_o(x_o + y_o + 1) - 2x_o\} = 0 \tag{2.4.2}$$

$$\alpha(x_o + y_o)\theta e^{2\alpha\theta} Da_1^2 + \{e^{\alpha\theta}[\theta\alpha(x_o + y_o) - \theta(1 + y_o) - 1] - (x_o + y_o)\} Da_1 - (1 + y_o) = 0 \tag{2.4.3}$$

The curve characterized by the vanishing trace condition starts at $Da_2 = 0$, where θ_H and Da_{1H} using Eq. (2.3.3) and (2.3.5) are given by,

$$Da_{1H} = \frac{\theta - y_o}{x_o + y_o - \theta} e^{-\alpha\theta} \tag{2.4.4}$$

$$\theta_H = \frac{(1 + \alpha x_o + 2\alpha y_o) \pm \sqrt{\alpha^2 x_o^2 - 6\alpha x_o + 1}}{2\alpha} \tag{2.4.5}$$

This curve ends at a limit point in the $Da_1 - Da_2$ plane at $Da_2 = Da_{2,\max}$. The corresponding expressions for θ_H,

$$\alpha(1 + \Omega_1)^2\theta_H^2 - \theta_H\{\alpha(x_o + y_o) + \alpha y_o + (1 + \Omega_1)\} + \{[\alpha(x_o + y_o) - 1]y_o - (\Omega_1 + 2)(x_o + y_o)\} = 0 \tag{2.4.6}$$

where Ω_1 is the root of Eq. (2.4.2), and $Da_{1,max}$ is obtained from Eq. (2.4.3).

2.5 Numerical Method

To obtain the bifurcation diagram we have simultaneously solved Eq. (2.3.3), and steady state equation in terms of θ, along with one of the three conditions viz. $T = 0$, $D = 0$ and $\Delta = 0$, making use of Eq. (2.3.2) to reduce the equations into a single variable form. For this purpose the parameter Da_1 was expressed as,

$$Da_1 = \left\{ \frac{x_o}{x_o + y_o - \theta(1 + Da_2)} - 1 \right\} \exp(-\alpha\theta) \qquad (2.5.1)$$

using Eq. (2.3.3). For $T = 0$ and $\Delta = 0$, θ was kept as the single variable, whereas for the case $D = 0$, the equations were reduced in terms of x_s to avoid numerical instability. To obtain the roots of the function (T, D and Δ equal to zero), the bisection method was employed. The condition stated in Eq. (2.3.9) obtained from Eq.(2.3.8) for secondary bifurcation is the limit point of the curve $D = 0$ in the bifurcation diagram. This condition has been made use of in avoiding the numerical overflow while reaching the limit point in the numerical solution for obtaining curve $D = 0$.

2.6 Results and Discussion

The linear stability analysis of the exponential autocatalysis reveals many interesting features, which are elucidated in the following. Fig. 2.2a shows in the $Da_1 - Da_2$ plane the various curves which satisfy $T=0$, $D=0$ and $\Delta = T^2 - 4D = 0$. For the sake of clarity some portions of this figure have been magnified and shown as Fig. 2.2b and 2.2c. It is interesting to note that the transcendental nature of $\Delta = 0$ gives rise to four real roots in certain region of $Da_1 - Da_2$ phase plane while $T = 0$ and $D = 0$ give rise to two distinct roots. The curves representing the three conditions intersect each other at various points, enclosing certain regions in the parameter space. These various regions formed in the bifurcation diagram are discussed

below and their classification is presented in Table 1. Referring to Fig. 2.2a, 2.2b and 2.2c, we notice that in all there are four different basic types of regions observed within the boundaries described by three conditions mentioned above.

Region I, essentially a region of stability consists of a stable node (two real negative eigenvalues), and/or a stable focus (or spiral, two complex eigenvalues with negative real part). On the other hand, region II consists of an unstable node and unstable spiral (or focus, real part of complex eigenvalues being positive). Region III consists of multiplicity (three) solutions with the middle solution always being a saddle point with upper and lower steady states as either a stable or unstable focus (spiral) or node. All such eight combinations have been found to exist in the bifurcation diagram. Region IV contains multiplicity of two solutions given by an unstable focus and an unstable solution.

The boundary line with $D = 0$ is characterized by a pair of real eigenvalues with one being equal to trace and the other a simple zero eigenvalue. So the stability of the solution is determined by the sign of trace. The line characterized by $T = 0$ with $D < 0$ possesses two pure imaginary complex eigenvalues, and each such steady state solution represents a center point. These solutions can be called as neutrally stable solutions with nonvanishing finite imaginary parts. The curve having the discriminant $\Delta = 0$ and nonzero trace, is a locus of solutions with only eigenvalue equal to half the trace. So it follows that the stability of such solutions is governed by the sign of trace. We may add to our discussion that when the curves $T = 0$ and $D = 0$ cross each other, the intersection point is where homoclinic bifurcation can occur.

The stability exchange takes place when the trace and determinant change signs. For $T<0$ and $D>0$ the solution is termed as stable. As trace changes sign from negative to positive, the system crosses through the Hopf bifurcation point at $T = 0$ and later goes into the limit cycle region. Another possible bifurcation occurs as trace remains negative and determinant changes sign from positive to negative. The system with $T>0$ and $D>0$ *can also bifurcate to a state where* determinant changes sign, with trace remaining positive. The last two states give rise to unstable

states and no further possibility of exchange exists between them. The two types of chemical instabilities occur when following conditions are met: when a stable steady state solution identified by the condition $T < 0$ and $D > 0$, changes as, T -> -0 with determinant sign remaining unchanged, one type of instability sets in, whereas the other type of instability occurs when D -> +0 and trace remains negative. In other words this also implies that the circle traced out by the eigenvalues in the Iml -Re l plane as the bifurcating parameter changes, lies entirely to the left half of the imaginary axis. A typical solution diagram (x_s versus $D\alpha_1$) obtained using the DERPAR routine (Kubicek and Marek,1983) is shown in Fig. 2.3. It is seen that bistability is predominant, in the parameter region where a solution for the condition determinant equal to zero, exists.

Table 1

Classification of Regions obtained in the Bifurcation Diagram

Type of Region	Region	Sub-region	Condition			Remarks
Unique stable state.	I	a	T<0	D>0	Δ>0	Stable node with two negative real eigenvalues.
	I	b	T<0	D>0	Δ<0	Two complex eigenvalues with real negative part. A stable focus.
Unique unstable state.	II	a	T>0	D>0	Δ>0	Unstable node with two positive real eigenvalues.
	II	b	T>0	D>0	Δ<0	Two complex eigenvalues with real positive part. An unstable focus.
Region of multiplicity. Three solutions	III	a	T<0	D>0	Δ>0	Stable node - saddle point - stable node.
			T<0	D<0	Δ>0	
			T<0	D>0	Δ>0	
	III	b	T<0	D>0	Δ>0	Stable node - saddle point - stable focus.
			T<0	D<0	Δ>0	
			T<0	D>0	Δ<0	
	III	c	T<0	D>0	Δ<0	Stable focus - saddle point - stable node.
			T>0	D<0	Δ>0	
			T<0	D>0	Δ>0	
	III	d	T<0	D>0	Δ<0	Stable focus - saddle point - stable focus.
			T<0	D<0	Δ>0	
			T<0	D>0	Δ<0	
	III	e	T>0	D>0	Δ>0	Unstable node - saddle point - stable focus.
			T>0	D<0	Δ>0	
			T<0	D>0	Δ<0	
	III	f	T>0	D>0	Δ<0	Unstable focus - saddle point - stable focus.
			T>0	D<0	Δ>0	
			T<0	D>0	Δ<0	

Type of Region	Region	Sub-region	Condition			Remarks
	III	g	$T>0$	$D>0$	$\Delta<0$	Unstable focus - saddle point - stable node.
			$T>0$	$D<0$	$\Delta>0$	
			$T<0$	$D>0$	$\Delta>0$	
	III	h	$T>0$	$D>0$	$\Delta>0$	Unstable node - saddle point - unstable focus.
			$T>0$	$D<0$	$\Delta>0$	
			$T>0$	$D>0$	$\Delta<0$	
Region of multiplicity. Two solutions	IV	-	$T>0$	$D>0$	$\Delta<0$	Unstable focus - saddle point.
			$T<0$	$D<0$	$\Delta>0$	
Curve with $\Delta=0$	-	-	$T<0$	$D>0$	$\Delta=0$	Negative eigenvalues equal to half the trace.
			$T>0$	$D>0$	$\Delta=0$	Positive eigenvalues equal to half the trace.
Curve with $T=0$	-	-	$T=0$	$D<0$	$\Delta>0$	Saddle point.
			$T=0$	$D>0$	$\Delta<0$	Two pure complex eigenavlues, existence of a center.
Curve with $D=0$	-	-	$T>0$	$D=0$	$\Delta>0$	One positive eigenvalue equal to trace and one simple zero eigenvalue.
			$T<0$	$D=0$	$\Delta>0$	One negative eigenvalue equal to trace and one simple zero eigenvalue.

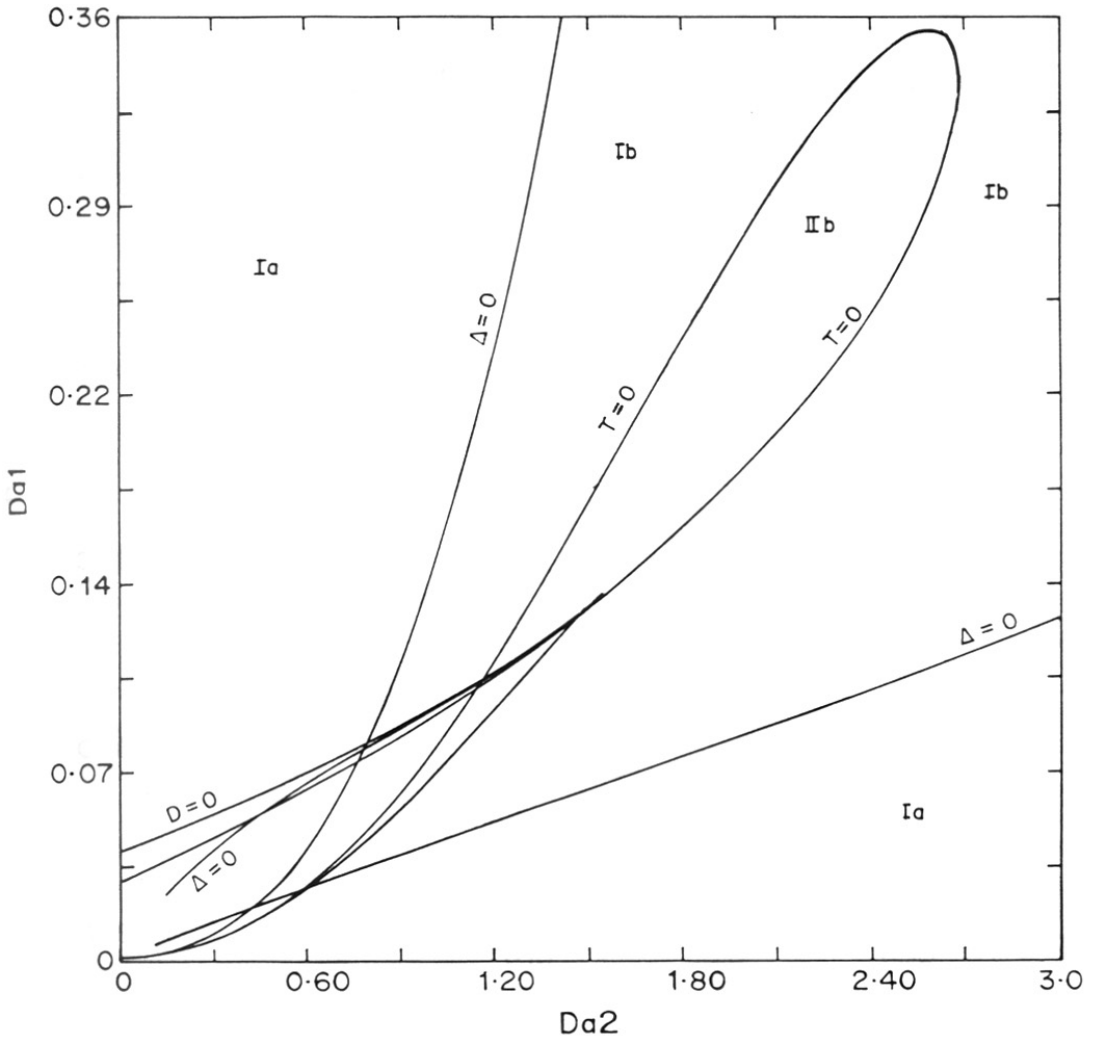

Fig. 2.2a : Bifurcation Diagram ($\alpha = 20$, $x_0 = 0.5$ $y_0 = 0.0$)

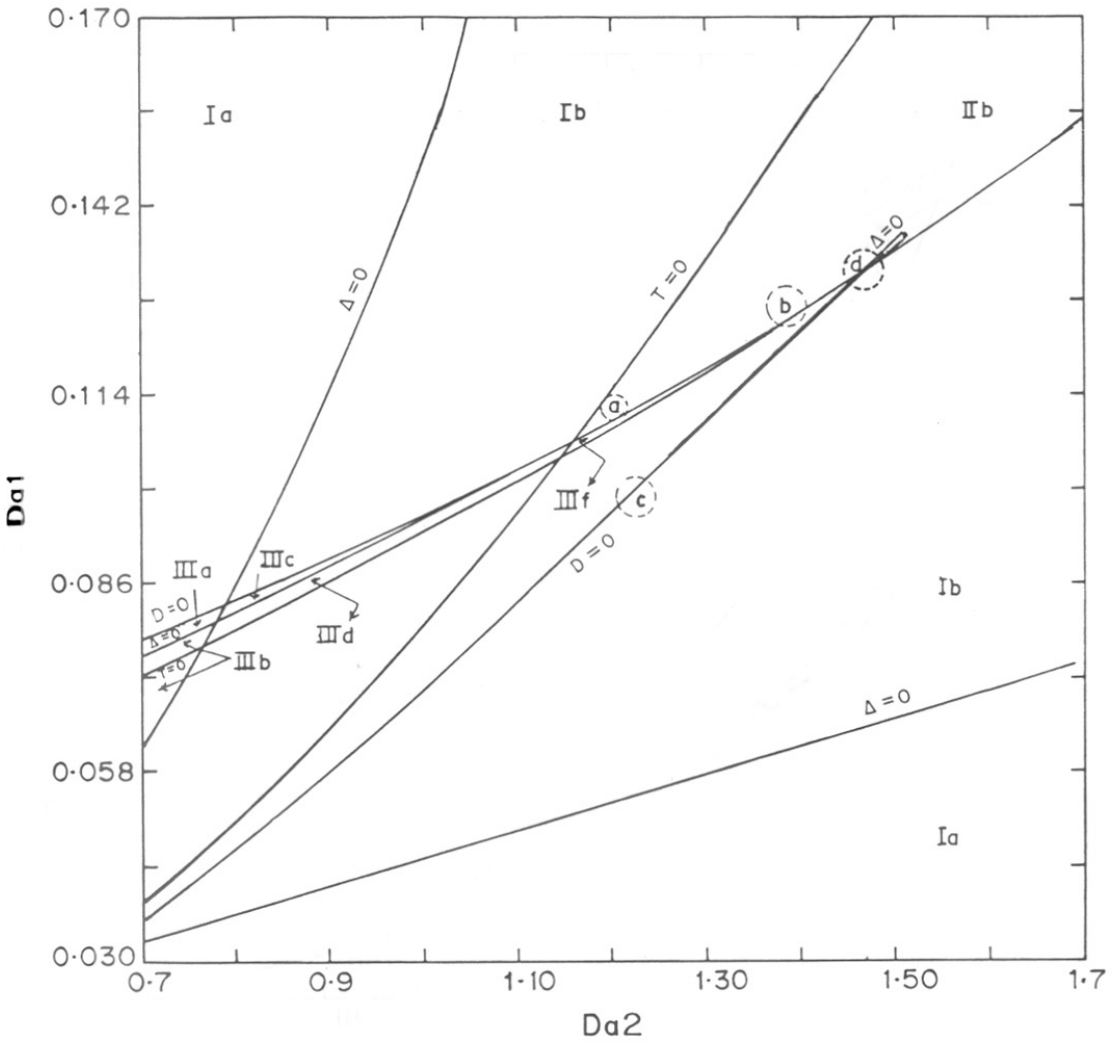

Fig. 2.2b : Magnified part of the bifurcation map for $Da_2 = 0.7 - 1.7$

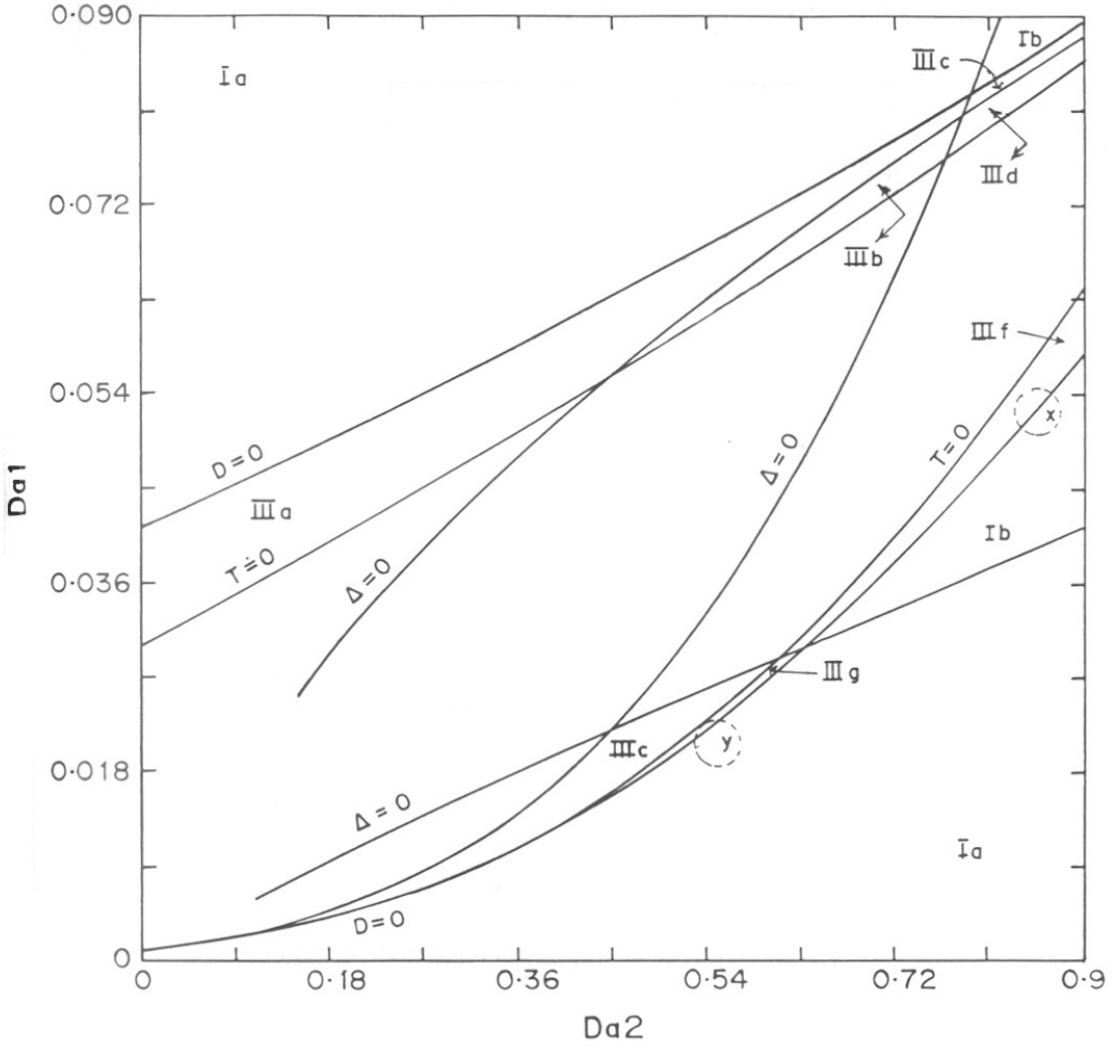

Fig 2.2c : Magnified part of the bifurcation map for $a\ Da_2 = 0.0 - 0.9$

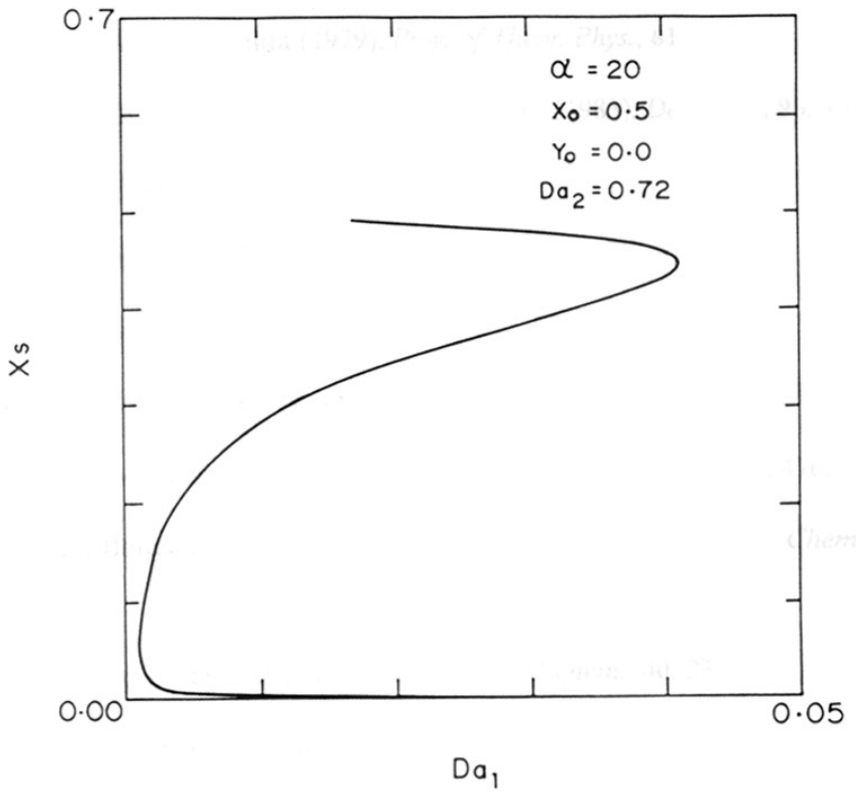

Fig 2.3 : Solution Diagram showing bistability

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CHAPTER III

MULTI-TIME SCALE ANALYSIS OF EXPONENTIAL AUTOCATALYSIS

In this chapter we employ the two-time scales (singular perturbation) method to construct the limit cycle and global nonuniform steady patterns for the exponential autocatalysis model, for a defined set of initial and boundary conditions. Here the analysis is carried out in presence of diffusion. The stability of the nonlinear structures that appear, is also analyzed.

3.1 Introduction

Scientists and engineers are often faced with problems which cannot be solved exactly and hence resort to approximate methods becomes necessary. One such problem is that of the interplay between mechanisms evolving on vastly different time scales, which is a commonly occurring phenomenon in nature. Often in such situations, the equations governing the dynamics of the system can be considerably simplified by eliminating the fast variables, and obtaining an approximate description, entirely in terms of the slow variables. The simplified slow description is usually the one we know from classical macroscopic laws of the concerned field. For example the hydrodynamic equations are obtained from the Boltzmann equation by eliminating all the variables except the local density, momentum and energy; Ohm's laws are derived based on the fact that the field varies much slowly compared with the rapid motion of the electrons; kinetic laws are obtained after the elimination of short lived intermediates in the reaction.

In biological systems, there is a definite hierarchy of processes, occurring in widely different time scales. At the lower end are the elementary life supporting reactions, while at the other end are genetic regulations and evolutionary processes. For a useful account of biological significance of time hierarchies and some methods of approximating their behavior, one can see the review by Heinrich *et al* (1977).

The approximations, leading to the description of the system in terms of the slow variables is conventionally called 'adiabatic elimination of fast variables' , 'perfect delay convention', or the 'slaving principle'. We shall discuss this principle a little more in detail in Chapter IV.

The first task one needs to perform before systematically eliminating the fast variables is the identification of the fast and slow variables. Unfortunately there is no rigorous theory for this purpose. However, after having acquired some fundamental understanding about the system in question, one can obtain a set of differential equations involving a small parameter, ϵ . Having obtained such a set of equations, we aim to find an expansion in successive powers of ϵ , known as the perturbation expansion. In a perturbation series, successive terms of higher

order ϵ account successively for more details of the influence of the fast variables on the system dynamics. The lowest order equation in ϵ represents the behavior of the system in absence of the influence from the fast variables.

Multi-time scale theory is a term applied to a class of methods which introduce in a systematic way, several time scales into the original equations. The simplest way of doing this is, to assume that the system variables, **u** vary not only with t, but also with t_n, where t_n are defined as, $t_n = \epsilon^n t$, i.e.,

$$t_1 = \epsilon t \ ; \ t_2 = \epsilon^2 t \ldots\ldots$$

Thus t_n represent different time scales, and since ϵ is a small parameter, we can immediately see that t is a fast scale where as t_1, t_2,..... represent successively slower scales. Using the chain rule, the time derivative in the original equations becomes,

$$\frac{d}{dt} = \frac{\partial}{\partial t} + \epsilon \frac{\partial}{\partial t_1} + \epsilon^2 \frac{\partial}{\partial t_2} + \ldots\ldots\ldots$$

In the present chapter, we exploit the fact that around some critical value of a control parameter, γ_c, where there is a qualitative change in the stability of system, the deviations from the original state vary on a slow scale, and that the slowness parameter is derived based on the distance from the critical value. Thus different time scales are inherent in the system. Here we employ the two-time scales (singular perturbation) method to construct the limit cycle and global nonuniform steady patterns that appear in this reaction-diffusion system for a defined set of initial and boundary conditions. The stability of these nonlinear structures is also analyzed.

3.2 The Model

The reaction-diffusion system representing the exponential autocatalysis model discussed in the last chapter is given by the following set of equations.

$$\frac{\partial X}{\partial t} = D_1 \Delta X + x_o - X - Da_1 X \exp(\alpha Y) \qquad (3.2.1a)$$

$$\frac{\partial Y}{\partial t} = D_2 \Delta Y + y_o - Y + Da_1 X \exp(\alpha Y) - Da_2 Y \qquad (3.2.1b)$$

where the operator $\Delta = \partial^2/\partial r^2$.

The steady state homogeneous solution to system in Eq. (3.2.1) is given as,

$$\exp(\alpha\theta) = \frac{(x_o - x_s)}{x_s Da_1}, \qquad \theta = \frac{x_o + y_o - x_s}{1 + Da_2} \qquad (3.2.2)$$

where x_s and θ are the steady state values of X, Y respectively.

The existence of this solution in Eq. (3.2.2) depends upon the boundary conditions. In the present case, we assume the concentrations to be fixed at the boundaries i.e. Dirichlet condition. This boundary condition is given as,

$$X(0,t) = X(1,t) = x_s$$

$$Y(0,t) = Y(1,t) = \theta, \qquad \text{for} \quad t > 0 \qquad (3.2.3)$$

All the calculations have been carried out for a one-dimensional system. To make this a well-posed problem, we add the following initial conditions,

$$X(r,0) = X_{in}(r) = x_o, \qquad (3.2.4a)$$

$$Y(r,0) = Y_{in}(r) = y_o \qquad (3.2.4b)$$

Assuming the initial conditions x_0 and y_0 to be non-negative, there exists a non-negative pair $(X(r,t), Y(r,t))$ of solutions of the system defined for $0 \le r \le 1$ and $0 \le t < \infty$. These solutions are infinitely differentiable functions of both r and t on $(0,1)X(0,\infty)$.

Defining deviations from steady state as

$u = \begin{pmatrix} x \\ y \end{pmatrix}$,

$$X = x + x_s, \qquad Y = y + \theta,$$

which obey homogeneous boundary conditions, and using the linearization of the exponential term

$$\exp(\alpha y) = 1 + \alpha y,$$

results in following evolution equations,

$$\frac{\partial x}{\partial t} = D_1 \Delta x - (1 + Da_1 e^{\alpha\theta}) x - (\alpha Da_1 x_s e^{\alpha\theta}) y - \alpha Da_1 e^{\alpha\theta} xy \qquad (3.2.5a)$$

$$\frac{\partial y}{\partial t} = D_2 \Delta y + Da_1 e^{\alpha\theta} x + (\alpha x_s Da_1 e^{\alpha\theta} - Da_2 - 1) y + \alpha Da_1 e^{\alpha\theta} xy \qquad (3.2.5b)$$

The boundary and initial conditions, in terms of the deviation variables, are,

$$x(0,t) = x(1,t) = y(0,t) = y(1,t) = 0, \qquad t \geq 0 \qquad (3.2.6)$$

and,

$$x(r,0) = X_{in}(r) - x_s, \qquad (3.2.7)$$

$$y(r,0) = Y_{in}(r) - \theta, \qquad 0 \leq r \leq 1 \qquad (3.2.8)$$

Introducing $\eta = D_1 / D_2$, , and $D = D_2$ for any parameter $\gamma = (\alpha, x_0, y_0, Da_1, Da_2, D, \eta)$, the linear differential operator can be written as,

$$L(\gamma) = \begin{pmatrix} \eta D\Delta - (1 + Da_1 e^{\alpha\theta}) & -\alpha x_s Da_1 e^{\alpha\theta} \\ Da_1 e^{\alpha\theta} & D\Delta + \alpha x_s Da_1 e^{\alpha\theta} - (1 + Da_2) \end{pmatrix} \qquad (3.2.9)$$

The nonlinear function, $N(\gamma, u)$ is represented as,

$$N(\gamma, u) = \begin{pmatrix} -\alpha Da_1 e^{\alpha\theta} xy \\ \alpha Da_1 e^{\alpha\theta} xy \end{pmatrix} \qquad (3.2.10)$$

So, the original Eq. (3.2.5) becomes,

$$u_t = L(\gamma)u + N(\gamma, u) \tag{3.2.11}$$

We are now interested in finding out the asymptotic solutions of Eq. (3.2.11) for $t \to \infty$ which are nontrivial solutions $u \neq 0$ with a boundary condition described in Eq. (3.2.6).

The sufficient condition for instability with respect to boundary condition (3.2.6) is that the solution $u = 0$ be unstable to small disturbances. Hence, the linearized form of Eq. (3.2.11),

$$\left[\frac{\partial}{\partial t} - L(\gamma)\right] u = 0, \tag{3.2.12}$$

would have a nontrivial solution for the specified boundary condition.

The solution to Eq. (3.2.12) can be given as,

$$u(r, t) = \Xi(r)e^{\lambda t} \tag{3.2.13}$$

where $\Xi(r) = (\xi(r), \kappa(r))^T$ corresponds to a solution to space dependent part, and λ is the eigenvalue for the time-dependent part. Then the eigenvalue problem to steady state version of Eq. (3.2.11) can be written as,

$$[L(\gamma) - \lambda I]\ \Xi(r) = 0 \tag{3.2.14}$$

The solution to Eq. (3.2.12) then becomes,

$$u(r, t) = \sum_{n=1}^{\infty} c_n e^{\lambda_n t} \Xi_n(r) \tag{3.2.15}$$

The eigenfunctions for any wavenumber n, can be written for the Dirichlet problem as, $\Xi(r) = \begin{pmatrix} \xi_n(r) \\ \kappa_n(r) \end{pmatrix}$, where,

$$\xi_n(r) = \sin n\pi r, \qquad \kappa_n(r) = M_n \sin n\pi r \tag{3.2.16}$$

Using Eqs. (3.2.9) and (3.2.14), we can write the characteristic equation in terms of trace $Tr(\gamma, n)$ and determinant $Det(\gamma, n)$ as,

$$\lambda_n^2 - Tr(\gamma, n)\lambda_n + Det(\gamma, n) = 0, \qquad (3.2.17)$$

The trace and determinant expressions are given as,

$$Tr(\gamma, n) = (\alpha x_s D a_1 e^{\alpha\theta} - D a_2 - 1) - (1 + D a_1 e^{\alpha\theta}) - n^2\pi^2 D(1+\eta) \qquad (3.2.18)$$

$$Det(\gamma, n) = (n^2\pi^2 D)^2\eta - n^2\pi^2 D[\eta(\alpha x_s D a_1 e^{\alpha\theta} - D a_2 - 1) - (1 + D a_1 e^{\alpha\theta})]$$
$$-[\alpha x_s D a_1 e^{\alpha\theta} - D a_2 - 1](1 + D a_1 e^{\alpha\theta}) + \alpha x_s (D a_1 e^{\alpha\theta})^2 \qquad (3.2.19)$$

The eigenvalues are then obtained from Eq. (3.2.17) as,

$$2\lambda_n^* = [[D a_1 e^{\alpha\theta}(\alpha x_s - 1) - (D a_2 + 2)] - n^2\pi^2 D(1+\eta)] \pm \{(n^2\pi^2 D)^2(1-\eta)^2$$
$$+ 2(1-\eta)n^2\pi^2 D[D a_2 - D a_1 e^{\alpha\theta}(\alpha x_s + 1)] + D a_2^2$$
$$+ D a_1 e^{\alpha\theta}[(\alpha x_s - 1)^2 D a_1 e^{\alpha\theta} - 2 D a_2(\alpha x_s + 1)]\}^{1/2} \qquad (3.2.20)$$

Substituting for Ξ from Eq. (3.2.16) in (3.2.14), the eigenfunctions can be obtained in terms of the eigenvalues as,

$$\lambda_n^* + (1 + D a_1 e^{\alpha\theta}) + n^2\pi^2\eta D + \alpha x_s D a_1 e^{\alpha\theta} M_n^* = 0 \qquad (3.2.21)$$

From Eq. (3.2.14) and (3.2.16), we also obtain

$$M_n^+ M_n^- = 1/\alpha x_s \qquad (3.2.22)$$

Note that, the eigenvalues have negative real part if and only if $Tr(L(\gamma)) < 0$ and $Det(L(\gamma)) > 0$, in which case the solution is linearly stable. If either $Tr(L(\gamma)) > 0$ or $Det(L(\gamma)) < 0$, then the solution is linearly unstable. If $Det(L(\gamma))$ changes sign, an exchange of stability takes place as one eigenvalue of $L(\gamma)$ changes sign. This results in bifurcation of steady state solution branches. If $Det(L(\gamma)) > 0$ and $Tr(L(\gamma))$ changes sign, exchange of

stability occurs as the real part of the eigenpair of $L(\gamma)$ changes sign. This corresponds to Hopf bifurcation, which generates a nontrivial branch of periodic solutions. However, if $Det(L(\gamma)) < 0$ when $Tr(L(\gamma))$ changes sign, no bifurcation occurs, and hence there is no exchange of stability. This is depicted in Fig. (3.1).

In this present study, we are interested in analyzing the possible modes through which instability sets in ending up with Hopf bifurcation. This can happen in two ways.

(i)At some $\gamma = \gamma_c$, an eigenvalue $\lambda^*_{n_c}$ crosses the imaginary axis with nonvanishing imaginary part. This case is in accordance with the conditions, that for critical value of parameter γ_c, and for any wave number n if trace is negative and determinant is nonnegative, the solution is stable. For the critical value of wave number n, we may have a vanishing trace condition, leading to Hopf bifurcation which is the onset of instability. To find the critical value n_c we then put the trace derivative $dTr(\gamma, n)/dn|_{n=n_c}$ equal to zero. This yields the result $n_c = 1$. Substituting for this critical value of n in the $Tr(\gamma, n) = 0$ condition we obtain the locus of points corresponding to neutral stability $\left(\mathrm{Re}\,\lambda^*_{n_c} = 0\right)$ in the plane Da_1, Da_2 as,

$$Da_1 e^{\alpha\theta}(\alpha x_s - 1) - (Da_2 + 2) = \pi^2 D(1+\eta) \qquad (3.2.23)$$

(ii)At $\gamma = \gamma_c$, the only value of n_c that crosses the imaginary axis from negative to positive has vanishing imaginary part. This means that at critical value of wave number n_c trace is negative and determinant is zero, while for other values of n the solution is stable as trace is again negative and determinant is non-negative. Then the critical value of wave number is obtained by putting determinant derivative $dDet(\gamma, n)/dn|_{n=n_c}$ equal to zero. This gives,

$$n_c = \left|\left|\pi^{-1} D^{-1/2} \eta^{-1/4} \{(1+Da_2) + Da_1 e^{\alpha\theta}[(1+Da_2) - \alpha x_s]\}^{1/4}\right|\right| \qquad (3.2.24)$$

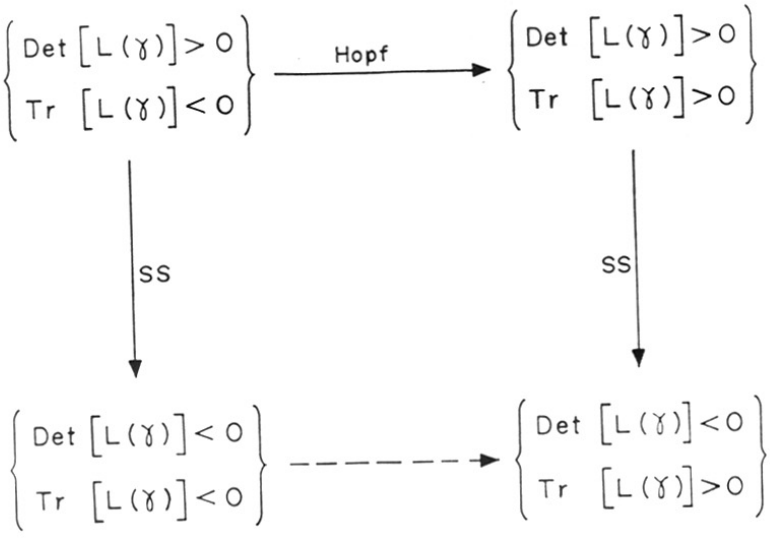

Fig. 3.1 : Stability Exchange Diagram; If $Det(L(\gamma)) > 0$ when $Tr(L(\gamma))$ changes sign Hopf bifurcation occurs; However if $Det(L(\gamma)) < 0$ when $Tr(L(\gamma))$ changes sign, there is no exchange of stability and no bifurcation, which is depicted by the broken line; SS denotes bifurcation from one steady state to another.

The locus of neutrally stable states obtained by substituting the value of n_c from Eq. (3.2.24) in the $Det(\gamma, n) = 0$ condition,

$$\{\eta[(1 + Da_2) + Da_1 e^{\alpha\theta}[(1 + Da_2) - \alpha x_s]]\}^{1/2} = \frac{\eta[\alpha x_s Da_1 e^{\alpha\theta} - (1 + Da_2)] - (1 + Da_1 e^{\alpha\theta})}{2} \quad (3.2.25)$$

Inserting Eq. (3.2.25) into the condition $Tr(L(\gamma_c, n_c)) < 0$, we obtain an inequality as,

$$(1 - \eta)\ [\eta[\alpha x_s Da_1 e^{\alpha\theta} - (1 + Da_2)] + (1 + Da_1 e^{\alpha\theta})] < 0 \quad (3.2.26)$$

and from the sufficiency condition of minimum $Det(\gamma, n)$ one obtains,

$$1 < \eta, \quad \text{or} \quad D_1 > D_2 \quad (3.2.27)$$

Fig. (3.2) depicts for some specific values of Da_1 the linear stability diagram in the neighborhood of $u = 0$. It should be noted that in this work the diffusion plays the destabilizing role, where the mixing in a stirred vessel is very poor.

3.3 Multiple time scale analysis

In this section, we would apply the technique of multiple time scale to obtain the global nonuniform steady patterns. The multiple time scale analysis takes advantage of the existence of slow and fast time scales, inherent in the system to construct an asymptotic solution. The method has been extensively employed and illustrated in the literature (Newell and Whitehead, 1969; Nayfeh, 1973; Ortelova and Ross, 1974; Bender and Orszag, 1978; Bonilla and Velarde, 1979; Keener, 1982; Ramakrishna and Amundson, 1985; van Kampen, 1985).

To construct the nonuniform steady solution that branches at $Da_1 = Da_{1c}$, in region III-b of Fig.(3.2), we see that in terms of a small expansion parameter ϵ the perturbations upon the trivial fixed point $x = y = 0$ can be arbitrarily written as,

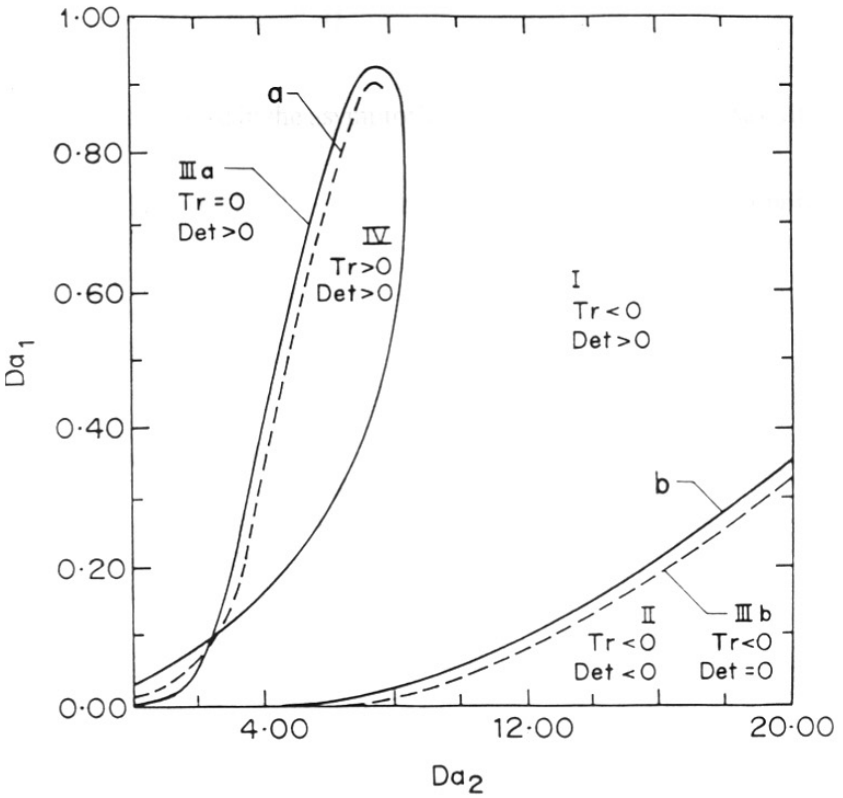

Fig. 3.2 : Stability diagram in the neighborhood of the homogeneous steady state [Eq. (3.2.2)]. Region I is of stability. Region III-a [Eq. (3.2.22)] and III-b [Eq. (3.2.24)] contains the unstable zone between the solid line and dotted line. In region III-a along a there is bifurcation to limit cycle behavior. In region III-b, along b spatial dissipative structures can occur. In region IV limit cycle behavior is expected.

$$x(r,0) = h(r,\epsilon); \qquad h_\epsilon(r,0) = \left(\frac{\partial h(r,\epsilon)}{\partial \epsilon}\right)_{\epsilon=0}, \tag{3.3.1a}$$

$$y(r,0) = g(r,\epsilon); \qquad g_\epsilon(r,0) = \left(\frac{\partial g}{\partial \epsilon}\right)_{\epsilon=0}, \tag{3.3.1b}$$

$$h(r,0) = g(r,0) = 0 \tag{3.3.1c}$$

The two time scales used in the asymptotic analysis are defined as, a fast time scale $\bar{t} = t$, and a slow scale $\tau = [Da_1(\epsilon) - Da_{1c}]t$. Now we define following expansions for the variables x and y,

$$x(r,t,\tau) \cong \sum_{i=1}^{\infty} \epsilon^i x_i(r,t,\tau), \qquad y(r,t,\tau) \cong \sum_{i=1}^{\infty} \epsilon^i y_i(r,t,\tau) \tag{3.3.2}$$

Note that the equation is exact as the series expands to all powers of ϵ and the corresponding expansions for initial and boundary conditions as,

initial condition:

$$x_j(r,0,0) = \frac{1}{j!}\frac{\partial^j h(r,0)}{\partial \epsilon^j} \tag{3.3.3a}$$

$$y_j(r,0,0) = \frac{1}{j!}\frac{\partial^j g(r,0)}{\partial \epsilon^j} \tag{3.3.3b}$$

boundary condition:

$$x_j(0,t,\tau) = x_j(1,t,\tau) = y_j(0,t,\tau) = y_j(1,t,\tau) = 0 \tag{3.3.3c}$$

Also the expansion for the bifurcation parameter Da_1, assuming it to be analytic in ϵ neighborhood of Da_{1c} can be written as,

$$Da_1(\epsilon) = Da_{1c} + Da_1'(0)\epsilon + \frac{1}{2}Da_1''(0)\epsilon^2 + O(\epsilon^3) \tag{3.3.4}$$

In terms of these expansions, $L(\gamma)$ and $N(\gamma, u)$ become,

$$L(Da_1) = \begin{pmatrix} \eta D\Delta - (1 + Da_{1c}e^{\alpha\theta}) & -\alpha x_s Da_{1c}e^{\alpha\theta} \\ Da_{1c}e^{\alpha\theta} & D\Delta + \alpha x_s Da_{1c}e^{\alpha\theta} - Da_2 - 1 \end{pmatrix} + \epsilon \begin{pmatrix} -Da_1'(0)e^{\alpha\theta} & -\alpha x_s Da_1'(0)e^{\alpha\theta} \\ Da_1'(0)e^{\alpha\theta} & \alpha x_s Da_1'(0)e^{\alpha\theta} \end{pmatrix} + \frac{1}{2}\epsilon^2 \begin{pmatrix} -Da_1''(0)e^{\alpha\theta} & -\alpha x_s Da_1''(0)e^{\alpha\theta} \\ Da_1''(0)e^{\alpha\theta} & \alpha x_s Da_1''(0)e^{\alpha\theta} \end{pmatrix} \quad (3.3.5)$$

and,

$$N(Da_1, u) = \epsilon^2 \begin{pmatrix} -\alpha e^{\alpha\theta} Da_{1c} x_1 y_1 \\ \alpha e^{\alpha\theta} Da_{1c} x_1 y_1 \end{pmatrix} + \epsilon^3 \begin{pmatrix} -\alpha e^{\alpha\theta}[Da_{1c}(x_1 y_2 + x_2 y_1) + Da_1'(0) x_1 y_1] \\ \alpha e^{\alpha\theta}[Da_{1c}(x_1 y_2 + x_2 y_1) + Da_1'(0) x_1 y_1] \end{pmatrix} \quad (3.3.6)$$

and the derivative term becomes,

$$\frac{\partial}{\partial t} = \frac{\partial}{\partial \tilde{t}} + \left[Da_1'(0)\epsilon + \frac{1}{2} Da_1''(0)\epsilon^2 + O(\epsilon^3) \right] \frac{\partial}{\partial \tau} \quad (3.3.7)$$

Here onwards, the tilde ~ on t will be dropped.

From Eqs. (3.2.12) and (3.3.5), collecting terms of equal powers of ϵ we obtain following linear equations,

$$L\begin{pmatrix} x_1 \\ y_1 \end{pmatrix} \equiv \begin{pmatrix} \frac{\partial}{\partial t} - [\eta D\Delta - (1 + Da_{1c}e^{\alpha\theta})] & \alpha x_s Da_{1c}e^{\alpha\theta} \\ -Da_{1c}e^{\alpha\theta} & \frac{\partial}{\partial t} - [D\Delta + \alpha x_s Da_{1c}e^{\alpha\theta} - (1 + Da_2)] \end{pmatrix} \begin{pmatrix} x_1 \\ y_1 \end{pmatrix} = 0 \quad (3.3.8)$$

$$L\begin{pmatrix} x_2 \\ y_2 \end{pmatrix} = \begin{pmatrix} -Da_1'(0)\dfrac{\partial x_1}{\partial \tau} + x_1(-Da_1'(0)e^{\alpha\theta}) + y_1(-\alpha x_s Da_1'(0)e^{\alpha\theta}) - x_1 y_1 \alpha e^{\alpha\theta} Da_{1c} \\ -Da_1'(0)\dfrac{\partial y_1}{\partial \tau} + x_1(Da_1'(0)e^{\alpha\theta}) + y_1(\alpha x_s Da_1'(0)e^{\alpha\theta}) + x_1 y_1 \alpha e^{\alpha\theta} Da_{1c} \end{pmatrix} \tag{3.3.9}$$

$$L\begin{pmatrix} x_3 \\ y_3 \end{pmatrix} = \begin{pmatrix} -Da_1'(0)\dfrac{\partial x_2}{\partial \tau} - \dfrac{1}{2}Da_1''(0)\dfrac{\partial x_1}{\partial \tau} + x_2(-Da_1'(0)e^{\alpha\theta}) + y_2(-\alpha x_s Da_1'(0)e^{\alpha\theta}) \\ + \dfrac{1}{2}x_1(-Da_1''(0)e^{\alpha\theta}) + \dfrac{1}{2}y_1(-\alpha x_s Da_1''(0)e^{\alpha\theta}) \\ - \alpha e^{\alpha\theta}[Da_{1c}(x_1 y_2 + x_2 y_1) + Da_1'(0)x_1 y_1] \\ -Da_1'(0)\dfrac{\partial y_2}{\partial \tau} - \dfrac{1}{2}Da_1''(0)\dfrac{\partial y_1}{\partial \tau} + x_2(Da_1'(0)e^{\alpha\theta}) + y_2(\alpha x_s Da_1'(0)e^{\alpha\theta}) \\ + \dfrac{1}{2}x_1(Da_1''(0)e^{\alpha\theta}) + \dfrac{1}{2}y_1(\alpha x_s Da_1''(0)e^{\alpha\theta}) \\ + \alpha e^{\alpha\theta}[Da_{1c}(x_1 y_2 + x_2 y_1) + Da_1'(0)x_1 y_1] \end{pmatrix} \tag{3.3.10}$$

The solution of Eq. (3.3.8) is,

$$\begin{pmatrix} x_1(r,t,\tau) \\ y_1(r,t,\tau) \end{pmatrix} = \mathrm{Re}\sum_{n=1}^{\infty}\left\{c_n^+(\tau)e^{\lambda_n^+ t}\Xi_n^+(r) + c_n^-(\tau)e^{\lambda_n^- t}\Xi_n^-(r)\right\} \tag{3.3.11}$$

Here the dominant eigenvalue is $\lambda_{n_c}^+ = 0$, while the eigenmodes corresponding to all other eigenvalues decay exponentially with *t*. Eq. (3.3.11) therefore reduces to,

$$\begin{pmatrix} x_1(r,t,\tau) \\ y_1(r,t,\tau) \end{pmatrix} = c_{n_c}^+(\tau)\Xi_{n_c}^+(r) + (\text{e.d.t}) \tag{3.3.12}$$

where (e.d.t.) denotes exponentially decaying terms.

The coefficients $c_{n_c}^+(0)$ can be obtained using Eqs. (3.3.1), (3.3.2),

$$c_n^*(0) = \frac{<\hat{\Xi}_n^* | \begin{pmatrix} h_\epsilon(r,0) \\ g_\epsilon(r,0) \end{pmatrix}>}{(1-\alpha x_s M_n^{*2})<\hat{\Xi}_n^* | \Xi_n^*>} \tag{3.3.13}$$

Here we have made use of the fact that if an orthogonal set $\Xi_n^*(r)$ is defined in F, then for any arbitrary function $f(r)$ belonging to F we have an expansion,

$$f(r) = \sum_{n=1}^{\infty} (\beta_n^+ \Xi_n^+(r) + \beta_n^- \Xi_n^-(r)) \tag{3.3.14}$$

where,

$$\beta_n^* = \frac{2<\hat{\Xi}_n^* | f>}{(1-\alpha x_s M_n^{*2})} \tag{3.3.15}$$

In Eq. (3.3.13) $\hat{\Xi}_n^*(r)$ is the eigen vector of the adjoint operator $\hat{L}(\gamma)$ of $L(\gamma)$.

$$\hat{\Xi}_n^*(r) = \begin{pmatrix} \sin n\pi r \\ N_n^* \sin n\pi r \end{pmatrix} \tag{3.3.16}$$

Solving the corresponding eigenvalue problem,

$$N_n^* = -\alpha x_s M_n^{**} \tag{3.3.17}$$

In this derivation we have made use of the linear operator property that if the eigenvalues λ_n^* of a given linear operator are complex, then the eigenvector $M_n^- = M_n^{+*}$, where * denotes complex conjugation.

If F is the space of analytic functions $u(r) = \begin{pmatrix} x(r) \\ y(r) \end{pmatrix}$ such that

$u(0) = u(1) = 0$, then, the inner product is defined as,

$$<u|\bar{u}> = \int_0^1 \{x^*(r)\bar{x}(r)+y^*(r)\bar{y}(r)\}dr \qquad (3.3.18)$$

This gives,

$$<\hat{\Xi}_m^*|\Xi_n^*> = \frac{1}{2}(1-\alpha x_s M_m^* M_n^*)\delta_{n,m}$$

$$<\hat{\Xi}_m^*|\Xi_m^*> = \frac{1}{2} \qquad (3.3.19)$$

and,

$$<\hat{\Xi}_n^*|\Xi_n^*> = 0 \qquad (3.3.20)$$

Making use of Eq. (3.3.14) - (3.3.20) in evaluating (3.3.13), we obtain,

$$c_n^*(0) = 2\int_0^1 \{\sin n\pi r(h_\epsilon(r,0)-\alpha x_s M n^* g_\epsilon(r,0))dr\} \qquad (3.3.21)$$

Thus, constants c_n^* are directly expressed in terms of the initial condition (3.3.1). Using the definition of Fredholm alternative the coefficient $c_{n_c}^*(\tau)$ can be obtained from the ϵ^2 equation in the set of Eqs. (3.3.8-3.3.10). It is convenient to introduce the following average which is useful when we take the limit $t \to \infty$.

$$<<\hat{\Xi}_{n_c}^*|f>> = \lim_{T\to\infty}\frac{1}{T}\int_0^T <\hat{\Xi}_{n_c}^*|f>dt \qquad (3.3.22)$$

where f is some arbitrary function in this equation. All products of $\hat{\Xi}_{n_c}^*$ with e.d.t. then vanish according to this definition.

Applying the average introduced in Eq. (3.3.22) to Eq. (3.3.21) we obtain,

$$\int_0^1 \{\sin^2 n_c\pi r\left\{-Da_1'(0)\frac{\partial c_{n_c}^*(\tau)}{\partial\tau}\left(1-\alpha x_s M_{n_c}^{*2}\right)+Da_1'(0)e^{\alpha\theta}c_{n_c}^*(\tau)\left(-\left(1+\alpha x_s M_{n_c}^*\right)^2\right)\right\}$$

$$+c_{n_c}^{*2}\sin^3 n_c\pi r\left(\alpha e^{\alpha\theta}Da_{1c}\right)\left\{-M_{n_c}^*-\alpha x_s M_{n_c}^{*2}\right\}\}dr=0 \qquad (3.3.23)$$

Note that $\int_0^1 \sin^3 n_c\pi r\,dr = (4/3n_c\pi)$ when n_c is odd

$=0$ when n_c is even

$$Da_1'(0)\frac{dc_{n_c}^*(\tau)}{d\tau}\left(1-\alpha x_s M_{n_c}^{*2}\right)=Da_1'(0)e^{\alpha\theta}c_{n_c}^*(\tau)\left\{-\left(1+\alpha x_s M_{n_c}^*\right)^2\right\}$$

$$+(4/3n_c\pi)c_{n_c}^{*2}\alpha e^{\alpha\theta}Da_{1c}\left(-M_{n_c}^*-\alpha x_s M_{n_c}^{*2}\right) \qquad (3.3.24)$$

We obtain the value of $M_{n_c}^*$ from Eq. (3.2.21) and the fact that at the critical point, the eigenvalue is real and zero.

$$M_{n_c}^* = -\frac{1}{2\alpha x_s Da_1 e^{\alpha\theta}}\{\eta[\alpha x_s Da_1 e^{\alpha\theta}-(1+Da_2)]+(1+Da_1 e^{\alpha\theta})\} \qquad (3.3.25)$$

$$\eta = \alpha x_s M_{n_c}^{**} \qquad (3.3.26)$$

From Eq. (3.3.9) and Eq. (3.3.24) - (3.3.26) we obtain,

$$\frac{1}{2}Da_1'(0)\frac{dc_{n_c}^*(\tau)}{d\tau}(1-\eta) = \frac{Da_1'(0)}{4}e^{\alpha\theta}c_{n_c}^*(\tau)$$

$$\times\left\{\frac{2[\eta(\alpha x_s Da_1 e^{\alpha\theta}-(1+Da_2))+(1+Da_1 e^{\alpha\theta})]-2Da_1 e^{\alpha\theta}(1+\alpha x_s\eta)}{Da_1 e^{\alpha\theta}}\right\}$$

$$+\ c_{n_c}^{*2}<\hat{\Xi}_{n_c}^*|\left[\frac{1}{2}\frac{\partial^2}{\partial\epsilon^2}N(\gamma,u)\right]_{\epsilon=0}> \qquad (3.3.27)$$

where,

$$<\hat{\Xi}^{*}_{n_c}|\left[\frac{1}{2}\frac{\partial^2}{\partial\epsilon^2}N(\gamma,u)\right]_{\epsilon=0}> = \left\langle \frac{\alpha e^{\alpha\theta} Da_{1c}[\eta(\alpha x_s Da_1 e^{\alpha\theta} - (1+Da_2)) + (1+Da_1 e^{\alpha\theta}) - 2\alpha x_s \eta Da_1 e^{\alpha\theta}]}{2\alpha x_s Da_1 e^{\alpha\theta} \frac{3}{4} n_c \pi} \right\rangle$$

when n_c is odd

$$= 0$$

when n_c is even

(3.3.28)

When n_c is odd and $<\hat{\Xi}^{*}_{n_c}|\left[\frac{1}{2}\frac{\partial^2}{\partial\epsilon^2}N(\gamma,u)\right]_{\epsilon=0}> \neq 0$, then we have,

$$\frac{dc^{*}_{n_c}(\tau)}{d\tau} = \nu\left[1 - \frac{c^{*}_{n_c}(\tau)}{c^{*}_{n_c}(\infty)}\right]c^{*}_{n_c}(\tau) \qquad (3.3.29a)$$

where,

$$\nu = \frac{1-\eta(1+Da_2)}{Da_1(1-\eta)} \qquad (3.3.29b)$$

$$c^{*}_{n_c}(\infty) = -\frac{3n_c\pi Da_1'(0)x_s}{4} \times \frac{1-\eta(1+Da_2)}{Da_{1c}[1+Da_1 e^{\alpha\theta}(1-\alpha x_s\eta)-\eta(1+Da_2)]} \qquad (3.3.30)$$

Integrating Eq. (3.3.29a) we obtain,

$$c^{*}_{n_c}(\tau) = \frac{c^{*}_{n_c}(0)c^{*}_{n_c}(\infty)e^{\nu\tau}}{c^{*}_{n_c}(\infty) - c^{*}_{n_c}(0)(1-e^{\nu\tau})} \qquad (3.3.31)$$

From Eqs. (3.3.11) and (3.3.31) and after substituting for, $M^{*}_{n_c}$ from Eq. (3.3.25), we obtain to first order in ϵ.

$$\epsilon\begin{pmatrix} x \\ y \end{pmatrix} \cong \epsilon c^{+}_{n_c}(\infty)c^{+}_{n_c}(0)\exp\left[\frac{-xy(Da_{1c}-Da_1)t}{c^{+}_{n_c}(\infty)-c^{+}_{n_c}(0)\left[1-\exp[-\nu(Da_{1c}-Da_1)t]\begin{pmatrix}1\\ M^{+}_{n_c}\end{pmatrix}\sin n_c\pi r\right]}\right]$$

$$+\epsilon c^{-}_{n_c}(0)e^{\lambda^{-}_{n_c}t}\begin{pmatrix}1\\ M^{-}_{n_c}\end{pmatrix}\sin n_c\pi r+\epsilon \mathrm{Re}\sum_{n\neq n_c}^{\infty} c^{+}_{n}(0)e^{\lambda^{+}_{n}t}\begin{pmatrix}1\\ M^{+}_{n}\end{pmatrix}\sin n\pi r+O(\epsilon^2) \tag{3.3.32}$$

if the trivial solution is to be asymptotically stable for $Da_1 > Da_{1c}$. To have such a case $c^{+}_{n_c}(\tau)=0$ for $t\to\infty$. This is obtained by imposing a condition,

$$\nu\tau = \nu(Da_1-Da_{1c})t < 0 \quad \text{at} \quad t\to\infty. \tag{3.3.33}$$

It follows from above that,

$$\nu = \frac{1-\eta(1+Da_2)}{Da_1(1-\eta)} < 0 \tag{3.3.34}$$

Since $\eta > 1$, we have, $\eta(1+Da_2) < 1$.

Eq. (3.3.34) can also be stated as follows :

$$\frac{\partial L(\gamma_c)}{\partial\epsilon} = \begin{pmatrix} -Da_1'(0)e^{\alpha\theta} & -\alpha x_s Da_1'(0)e^{\alpha\theta} \\ Da_1'(0)e^{\alpha\theta} & \alpha x_s Da_1'(0)e^{\alpha\theta}\end{pmatrix} \tag{3.3.35a}$$

Then, using Eq. (3.3.5), it can be shown that,

$$<\hat{\Xi}^{+}_{n_c}|\left[\frac{\partial L(\gamma_c)}{\partial\epsilon}\right]_{\epsilon=0}\Xi^{+}_{n_c}> = \frac{(1-\eta)\nu}{2} \tag{3.3.35b}$$

Hence, equivalently Eq. (3.3.34) can be stated as,

$$<\hat{\Xi}^{+}_{n_c}|\left[\frac{\partial L(\gamma_c)}{\partial\epsilon}\right]_{\epsilon=0}\Xi^{+}_{n_c}> < 0. \tag{3.3.36}$$

In Eq. (3.3.35b) and (3.3.36) we have on the left hand side, an inner product between $\hat{\Xi}_{n_c}$ and the vector obtained by the operation of $\frac{\partial L(\gamma_c)}{\partial\epsilon}]_{\epsilon=0}$ over Ξ_{n_c}.

For the sake of simplicity, we choose $Da_1'(0) = 1$ in Eq. (3.3.4). This gives,

$$Da_1 - Da_{1c} = \epsilon + O(\epsilon^2) \tag{3.3.37a}$$

To first order in ϵ, we then have the solutions x and y as,

$$x \cong c_{n_c}^{*}(\tau)(Da_1 - Da_{1c})\sin n_c \pi r$$

and

$$y \cong c_{n_c}^{*}(\tau)M_{n_c}^{*}(Da_1 - Da_{1c})\sin n_c \pi r \tag{3.3.37b}$$

If $c_{n_c}^{*}(0)$ and $c_{n_c}^{*}(\infty)$ have the same sign, with $Da_1 < Da_{1c}$, then as $t \to \infty$, the following asymptotic state will be reached.

$$\begin{pmatrix} x(r) \\ y(r) \end{pmatrix} \sim \begin{pmatrix} x_s \\ \theta \end{pmatrix} + c_{n_c}^{*}(\infty)\begin{pmatrix} 1 \\ M_{n_c}^{*} \end{pmatrix}(Da_{1c} - Da_1)\sin n_c \pi r + O[(Da_{1c} - Da_1)^2] \tag{3.3.38}$$

In more explicit terms, this becomes,

$$\begin{pmatrix} x(r) \\ y(r) \end{pmatrix} \sim \begin{pmatrix} x_s \\ \theta \end{pmatrix} + \left\{ \frac{3n_c \pi Da_1'(0)x_s}{4} \times \frac{1 - \eta(1 + Da_2)}{Da_1[1 + Da_1 e^{\alpha\theta}(1 - \alpha x_s \eta) - \eta(1 + Da_2)]} \right\} \times \begin{pmatrix} 1 \\ M_{n_c}^{*} \end{pmatrix}(Da_{1c} - Da_1)\sin n_c \pi r + O[(Da_{1c} - Da_1)^2] \tag{3.3.39}$$

The derivation assumes that the signs of $c_n^{*}(0)$ and $c_n^{*}(\infty)$ are similar. In the instance the signs of these differ, the denominator in Eq. (3.3.31) vanishes for a time interval of the order of $[\vee(Da_{1c} - Da_1)]^{-1}$. The solution after this time goes out of the ϵ region. Also, when Da_1 is slightly larger than Da_{1c} we would obtain the same equation for x and y; however, the

solution is now unstable and the neighboring concentration profiles diverge with time. The initial perturbation for this case with the signs of $c^*_{n_c}(0)$ and $c^*_{n_c}(\infty)$ different, will decay and the solution will culminate into the trivial asymptotically stable point.

The calculation of $c^*_n(\tau)$ for the case when n_c is even requires us to consider the next higher order Eq. (3.3.10). After some algebraic manipulations we then have,

$$\left[\frac{\partial}{\partial t} - L(\gamma_c)\right]\begin{pmatrix} x_2 \\ y_2 \end{pmatrix} = \begin{pmatrix} -\alpha e^{\alpha\theta} Da_{1c} M^*_{n_c} \\ \alpha e^{\alpha\theta} Da_{1c} M^*_{n_c} \end{pmatrix} c^{*2}_{n_c} \sin^2 n_c \pi r + (e.d.t.) \qquad (3.3.40)$$

Knowing that,

$$L(\gamma_c)\Xi^*_n(r) = \lambda^*_n(r) \qquad (3.3.41)$$

the particular solution to Eq. (3.3.40) is written as,

$$u_2^{ps} = \sum_{n \neq n_c} \beta^*_n(\tau)\Xi^*_n(r) \qquad (3.3.42)$$

the Eqs. (3.3.40) and (3.3.41) give,

$$\sum_{n \neq n_c} \beta^*_n(\tau)\lambda^*_n\Xi^*_n(r) = \begin{pmatrix} 1 \\ -1 \end{pmatrix} M^*_{n_c} \alpha e^{\alpha\theta} Da_{1c} c^{*2}_{n_c} \sin^2 n_c \pi r + (e.d.t.) \qquad (3.3.43)$$

Using Eq. (3.3.15), we get the result,

$$\beta^*_n(\tau) = \begin{Bmatrix} \rho^*_n c^{*2}_{n_c}(\tau) & \text{for odd } n \\ 0 & \text{for even } n \end{Bmatrix} \qquad (3.3.44)$$

where,

$$\rho^*_n = \frac{8n_c^2}{n(n^2 - 4n_c^2)\pi\delta^*_n} \frac{Da_{1c}(1 + \alpha x_s M^*_n)\{\eta[\alpha x_s Da_1 e^{\alpha\theta} - (1 + Da_2)] + (1 + Da_1 e^{\alpha\theta})\}}{2x_s Da_1(1 - \alpha x_s M^{*2}_n} \qquad (3.3.45)$$

The general solution of Eq. (3.3.40) then reduces to,

$$u_2 = b_{n_c}^{+}(\tau)\Xi_{n_c}^{+}(r) + c_{n_c}^{+2}(\tau)\Omega(r) + (e.d.t.) \tag{3.3.46}$$

where,

$$\Omega(r) = \begin{pmatrix} \omega(r) \\ \xi(r) \end{pmatrix} = \sum_{\substack{n \neq n_c \\ n \text{ is odd}}} \rho^{*}\Xi_{n}^{*}(r) \tag{3.3.47}$$

Substituting Eqs. (3.3.46) and (3.3.47) into Eq. (3.3.10) we obtain,

$$\left[\frac{\partial}{\partial t} - L(\gamma_c)\right]u_3 = -\frac{1}{2}Da_1''(0)\frac{\partial u_1}{\partial \tau} + L_{II}(\gamma_c)u_1 + N_{III}(\gamma_c, u_1, u_2) \tag{3.3.48}$$

where,

$$L_{II} = \left[\frac{1}{2}\frac{\partial^2 L(\gamma_c)}{\partial \epsilon^2}\right]_{\epsilon=0} = \begin{pmatrix} -1 & -\alpha x_s \\ 1 & \alpha x_s \end{pmatrix}\frac{Da_1''(0)}{2}e^{\alpha\theta} \tag{3.3.49a}$$

and

$$N_{III}(\gamma_c, u_1, u_2) = \begin{pmatrix} -1 \\ 1 \end{pmatrix}\alpha e^{\alpha\theta}Da_{1c}(x_1 y_2 + x_2 y_1) \tag{3.3.49b}$$

Multiplying Eq. (3.3.48) with $\hat{\Xi}_{n_c}^{+}(r)$ and applying Fredholm alternative with Eq. (3.3.22), the result is,

$$\frac{Da_1''(0)}{2}\frac{dc_{n_c}^{+}}{d\tau} = \frac{Da_1''(0)}{2}\nu c_{n_c}^{+} - \beta\nu c_{n_c}^{+3} \tag{3.3.50}$$

where,

$$\nu = \frac{1 - \eta(1 + Da_2)}{Da_1(1 - \eta)} \tag{3.3.51}$$

and,

$$\beta = \frac{-Da_{1c}\{\alpha x_s \xi(r)[Da_1 e^{\alpha\theta}(\alpha x_s \eta - 1) - \eta(1 + Da_2) + 1]\}}{2(1 - \eta(1 + Da_2))}$$

$$+ \frac{-Da_{1c}\{\omega(r)[1 + Da_1 e^{\alpha\theta} - \eta[(1 + Da_2) + \alpha x_s Da_1 e^{\alpha\theta}]]\}}{2(1 - \eta(1 + Da_2))} \qquad (3.3.52)$$

In the limit $\tau \to \infty$ one notices that,

$$c^{+}_{n_c}(\infty) = \pm[Da_1''(0)/2\beta]^{1/2} \qquad (3.3.53)$$

Therefore Eq. (3.3.50) using Eq. (3.3.53) becomes,

$$\frac{dc^{+}_{n_c}}{d\tau} = \nu c^{+}_{n_c}\left[1 - \frac{c^{+2}_{n_c}(\tau)}{c^{+2}_{n_c}(\infty)}\right] \qquad (3.3.54)$$

Integrating Eq. (3.3.54) we obtain,

$$c^{+}_{n_c}(\tau) = |c^{+}_{n_c}(\infty)|c^{+}_{n_c}(0)e^{\nu\tau}\left[c^{+2}_{n_c}(0)(e^{2\nu\tau} - 1) + c^{+2}_{n_c}(\infty)\right]^{-1/2} \qquad (3.3.55)$$

It is interesting to note that depending on the positive or negative sign of $c^{+}_{n}(0)$, the solution $c^{+}_{n}(\tau)$ goes to $|c^{+}_{n}(\infty)|$ or $-|c^{+}_{n}(\infty)|$. The dissipative structure at $t \to \infty$ therefore depends only on the sign of the initial conditions. The asymptotic expansion of the solution in this case gives,

$$\begin{pmatrix} x(r,t,\epsilon) \\ y(r,t,\epsilon) \end{pmatrix} = \begin{pmatrix} x_s \\ \theta \end{pmatrix} + \epsilon \begin{pmatrix} 1 \\ M^{+}_{n_c} \end{pmatrix} c^{+}_{n_c}(\tau)\sin n_c \pi r$$

$$+ \epsilon c^{-}_{n_c}(0)\begin{pmatrix} 1 \\ M^{+}_{n_c} \end{pmatrix} e^{\lambda^{+}_{n_c} t}\sin n_c \pi r + \epsilon \sum_{n \neq n_c}^{\infty} c^{*}_{n} e^{\lambda^{*}_{n} t}\begin{pmatrix} 1 \\ M^{*}_{n} \end{pmatrix}\sin n\pi r \qquad (3.3.56)$$

As $t \to \infty$,

$$\begin{pmatrix} x(r) \\ y(r) \end{pmatrix} \cong \begin{pmatrix} x_s \\ \theta \end{pmatrix} \pm \left(\frac{Da_1 - Da_{1c}}{\beta}\right) \times \begin{pmatrix} 1 \\ M^{+}_{n_c} \end{pmatrix}\sin n_c \pi r + O(|Da_1 - Da_{1c}|) \qquad (3.3.57)$$

Conclusively, we can say in the end that, when $Da_1 > Da_{1c}$, then the trivial solution is asymptotically stable, and vice-versa in the case of odd n_c.

3.4 Stability Analysis of Limit Cycle

We shall begin with the neutral stability curve given by Eq. (3.2.23),

$$Da_1 e^{\alpha\theta}(\alpha x_s - 1) - (Da_2 + 2) = \pi^2 D(1+\eta) \tag{3.4.1}$$

and note that the critical eigenvalue from Eq. (3.2.20) is,

$$\lambda_1^* = \pm i\left\{(Da_1 e^{\alpha\theta})^2 \alpha x_s - (\pi^2 D\eta + 1)^2\right\}^{1/2} \tag{3.4.2}$$

From Eq. (3.2.21), we have,

$$M_1^+ = \frac{-[\pm i\omega + (1 + Da_1 e^{\alpha\theta}) + \pi^2 D\eta]}{\alpha x_s Da_1 e^{\alpha\theta}} \tag{3.4.3}$$

We assume the solution to Eq. (3.3.8) with $n_c = 1$, as,

$$u_1(r,t,\tau) = \mathrm{Re}\{c_1^+(\tau)e^{i\omega t}\Xi_1^+(r) + c_1^-(\tau)e^{-i\omega t}\Xi_1^-(r)\} + (e.d.t) \tag{3.4.4}$$

The above equation contains two coefficients, which are unknown. It would be appropriate to define a new coefficient as follows,

$$c_1(\tau) = \frac{1}{2}[c_1^+(\tau) + c_1^{-*}(\tau)] \tag{3.4.5}$$

In addition, we have, $M_1^- = M_1^{+*}$ and $e^{-i\omega t}\Xi_1^-(r) = [e^{i\omega t}\Xi_1^+(r)]^*$ we can then write,

$$u_1(r,t,\tau) = c_1(\tau)e^{i\omega t}\Xi_1^+(r) + c.c. + (e.d.t.) \tag{3.4.6}$$

where *c.c.* stands for complex conjugate

The initial condition for $c_1(\tau)$ just as in Eq. (3.3.21) is given by,

$$c_1(0) = \frac{2\int_0^1 \{h_\epsilon(r,0) - \alpha x_s M_1^* g_\epsilon(r,0)\} \sin \pi r \, dr}{(1 - \alpha x_s M_1^{*2})} \tag{3.4.7}$$

Substitution of Eq. (3.4.4) into Eq. (3.3.9) gives,

$$\left[\frac{\partial}{\partial t} + (1 + Da_1 e^{\alpha\theta}) - \eta D\Delta\right] x_2 - \alpha x_s Da_{1c} e^{\alpha\theta} y_2$$

$$= \{-Da_1'(0)\sin \pi r (c_1' e^{i\omega t} + c.c.) + (c_1 e^{i\omega t} + c.c.)\sin \pi r[-Da_1'(0)e^{\alpha\theta}]$$

$$+ (c_1 e^{i\omega t} M_1^* + c.c.)\sin \pi r(-\alpha x_s Da_1'(0)e^{\alpha\theta}) + \sin^2 \pi r[(M_1^* + M_1^{**})|c_1|^2$$

$$+ c_1^2 e^{2i\omega t} M_1^* + c.c.](-\alpha e^{\alpha\theta} Da_{1c})\} + (e.d.t) \tag{3.4.8}$$

and,

$$\left[\frac{\partial}{\partial t} - [\alpha x_s Da_{1c} e^{\alpha\theta} - (1 + Da_2)] - D\Delta\right] y_2 + Da_{1c} e^{\alpha\theta} x_2 =$$

$$\{-Da_1'(0)\sin \pi r (c_1' M_1^* e^{i\omega t} + c.c.) + (c_1 e^{i\omega t} + c.c.)\sin \pi r (Da_1'(0)e^{\alpha\theta})$$

$$(c_1 e^{i\omega t} M_1^* + c.c.)\sin \pi r (\alpha x_s Da_1'(0)e^{\alpha\theta})$$

$$+ \sin^2 \pi r[(M_1^* + M_1^{**})|c_1|^2 + c_1^2 e^{2i\omega t} M_1^* + c.c.](\alpha e^{\alpha\theta} Da_{1c})\} + (e.d.t.) \tag{3.4.9}$$

Now, defining an average,

$$\ll [\hat{\Xi}_1^*] | f \gg = \lim_{T\to\infty} \frac{1}{T} \int_0^T < \hat{\Xi}_1^* | f > e^{i\omega t} dt \tag{3.4.10}$$

and using Fredholm alternative one sees that $Da_1'(0) = 0$, if c_1 is nonvanishing. Now, for the eigenvalue problem for the operator $L(u_2)$ we have,

$$\sum_{n\neq n_c} \beta_n^*(\tau)\lambda_n^*\Xi_n^*(r)$$

$$= \begin{pmatrix} \alpha e^{\alpha\theta} D a_{1c}[(M_1^* + M_1^{**})|c_1|^2 + c_1^2 e^{2i\omega t} M_1^* + c.c.] \\ -\alpha e^{\alpha\theta} D a_{1c}[(M_1^* + M_1^{**})|c_1|^2 + c_1^2 e^{2i\omega t} M_1^* + c.c.] \end{pmatrix} \sin^2 \pi r + (e.d.t.) \quad (3.4.11)$$

Applying Eq. (3.3.15) we obtain,

$$\beta_n^* = \frac{<\hat{\Xi}_n^* | f(r)>}{<\hat{\Xi}_n^* | \Xi_n^*>}$$

$$= -\frac{2<\begin{pmatrix} \sin \pi r \\ -\alpha x_s M_n^{**} \sin n\pi r \end{pmatrix} | \begin{pmatrix} \alpha e^{\alpha\theta} D a_{1c}[(M_1^* + M_1^{**})|c_1|^2 + c_1^2 e^{2i\omega t} M_1^* + c.c.] \\ -\alpha e^{\alpha\theta} D a_{1c}[(M_1^* + M_1^{**})|c_1|^2 + c_1^2 e^{2i\omega t} M_1^* + c.c.] \end{pmatrix} \sin^2 \pi r >}{(1 - \alpha x_s M_n^{*2})}$$

(3.4.12)

Integration of the system in Eq. (3.4.8) and (3.4.9) and the relation $\int_0^1 \sin n\pi r \sin^2 m\pi r\, dr = -4m^2/n^2(n^2-4m^2)$ yields,

$$u_2(r,t,\tau) = b_1(\tau)e^{i\omega t}\Xi_1^*(r) + c.c. + c_1^2(\tau)e^{2i\omega t}\Omega(r) + c.c.$$
$$+ |c_1(\tau)|^2[\Omega(r) + c.c.]_{\omega=0} + (e.d.t.) \quad (3.4.13)$$

where,

$$\Omega(r) = \begin{pmatrix} \omega(r) \\ \xi(r) \end{pmatrix} = \sum_{\substack{n=3 \\ n \text{ is odd}}}^{\infty} \frac{\rho_n^*}{2i\omega - \lambda_n^*} \times \{M_1^*(1 + \alpha x_s M_n^*)\}\Xi_n^*(r) \quad (3.4.14)$$

and,

$$\rho_n^* = \frac{-8}{n(n^2-4)}[1 - \alpha x_s M_n^{*2}], \quad (3.4.15)$$

$$[\Omega(r)]_{\omega=0} = \sum_{\substack{n=3 \\ n \text{ is odd}}}^{\infty} -\frac{\rho_n^*}{\lambda_n^*}\{M_1^*(1 + \alpha x_s M_n^*)\}\Xi_n^*(r) \quad (3.4.16)$$

To obtain $c_1(\tau)$, we substitute for u_1 and u_2 from Eq. (3.4.6) and (3.4.13) respectively into Eq. (3.3.10), with $Da_1'(0) = 0$. Multiplication of the result with $\hat{\Xi}_1^*(r)$ and using the identity in Eq. (3.4.10) as before, yields following differential equation,

$$\frac{Da_1''(0)}{2}\frac{\partial c_1^*}{\partial \tau} = \frac{Da_1''(0)}{2}\nu c_1^* + |c_1|^2 c_1^* \kappa \qquad (3.4.17)$$

where,

$$\nu = \frac{-e^{\alpha\theta}(1+\alpha x_s M_1^{**})^2}{(1-\alpha x_s M_1^{**2})} \qquad (3.4.18)$$

and,

$$\kappa = \frac{-2\int_0^1 \sin^2\pi r\, dr\, \alpha e^{**} Da_{1c}[\xi^*(r)+[\xi(r)+c.c.]_{\omega=0}+M_1^*\omega^*(r)+M_1^{**}[\omega(r)+c.c.]_{\omega=0}]\times\{1+\alpha x_s M_1^{**}\}}{(1-\alpha x_s M_1^{**2})} \qquad (3.4.19)$$

Writing, $c_1(\tau) = c(\tau)e^{-i\beta(\tau)}$ where $c(\tau)$ and $\beta(\tau)$ are yet to be specified, and then separating the real and imaginary parts in Eq. (3.4.17) gives us,

$$\frac{Da_1''(0)}{2}\frac{dc}{d\tau} = \frac{Da_1''}{2}c\,\mathrm{Re}\,\nu + c^3\,\mathrm{Re}\,\kappa \qquad (3.4.20a)$$

$$\frac{Da_1''(0)}{2}\frac{d\beta}{d\tau} = \frac{Da_1''(0)}{2}\mathrm{Im}\,\nu + c^2\,\mathrm{Im}\,\kappa \qquad (3.4.20b)$$

From Eq. (3.4.20a), as $\tau \to \infty$ we can write,

$$c(\infty) = \left[-\frac{Da_1''(0)}{2}\frac{\mathrm{Re}\,\nu}{\mathrm{Re}\,\kappa}\right]^{1/2} \qquad (3.4.21)$$

Using Eq. (3.4.21), Eq. (3.4.20b) can be rewritten as,

$$\frac{dc}{d\tau} = \mathrm{Re}\nu\left[1-\frac{c^2}{c(\infty)^2}\right]c \tag{3.4.22}$$

The solution to this equation is,

$$c(\tau) = \frac{c(0)c(\infty)e^{\mathrm{Re}\nu\tau}}{\{c(\infty)^2+c(0)^2[e^{2\mathrm{Re}\nu\tau}-1]\}^{1/2}} \tag{3.4.23}$$

The solution to unknown phase can be written using Eqs. (3.4.20b) and (3.4.23),

$$\beta(\tau) = \beta(0)+\tau\mathrm{Im}\nu+\frac{2\mathrm{Im}\kappa}{Da_1''(0)}\int_0^\tau c^2(s)ds \tag{3.4.24}$$

which for large values of time becomes,

$$\beta(\tau) \cong \tau\left\{\mathrm{Im}\nu-\frac{\mathrm{Im}\kappa\mathrm{Re}\nu}{\mathrm{Re}\kappa}\right\} \tag{3.4.25}$$

Finally, to first order in ϵ, the following result is obtained

$$\begin{aligned} u(r,t,\epsilon) \cong\ & \epsilon\frac{2c(0)c(\infty)\exp[-(Da_{1c}-Da_1)\mathrm{Re}\nu t]}{\{c(\infty)^2+c(0)^2\{\exp[-2(Da_{1c}-Da_1)\mathrm{Re}\nu t]-1\}\}^{1/2}} \\ & \times\left(-\frac{1}{\dfrac{2[1+Da_1e^{\alpha\theta}+\pi^2D\eta]}{\alpha x_s Da_1e^{\alpha\theta}}}\right)\times\cos[\omega t-\beta(Da_1-Da_{1c})t] \\ & +\epsilon\mathrm{Re}\sum_{n=2}^{\infty}c_n^*(0)\times e^{\lambda_n^*t}\Xi_n^*(r)+O(\epsilon^2) \end{aligned} \tag{3.4.26}$$

Eq. (3.4.26) reduces to following form as $t\to\infty$,

$$\begin{aligned} \begin{pmatrix}x(r,t)\\ y(r,t)\end{pmatrix} \cong & \begin{pmatrix}x_s\\ \theta\end{pmatrix}+2\left[\frac{\mathrm{Re}\nu}{\mathrm{Re}\kappa}(Da_{1c}-Da_1)\right]^{1/2}\left(-\frac{1}{\dfrac{2[1+Da_1e^{\alpha\theta}+\pi^2D\eta]}{\alpha x_s Da_1e^{\alpha\theta}}}\right)\times\sin\pi r \\ & \times\cos\left[\omega+(Da_{1c}-Da_1)\left(\mathrm{Im}\nu-\frac{\mathrm{Im}\kappa\mathrm{Re}\nu}{\mathrm{Re}\kappa}\right)\right]t+O(|Da_1-Da_{1c}|) \end{aligned} \tag{3.4.27}$$

3.5 Results and Discussion

The present study employs the two-time scale method to obtain the limit cycle and global nonuniform solutions for an exponentially autocatalyzed reaction-diffusion system. Sufficient condition for the steady uniform distribution of reactants in the presence of diffusion is established and stability of such states are examined. Global nonuniform solutions depending on whether n_c, the critical wave number, is even or odd, are then constructed and given respectively by Eqs. (3.3.32) and (3.3.56). Conditions under which the dissipative structures are asymptotically stable or when the inhomogeneous steady state solutions lose their stability are also identified. In a similar fashion Eq. (3.4.26) describe the limit cycle solution, the stability of which depends on whether $D\alpha_1$ exceeds $D\alpha_{1c}$ or not. In addition, we observe that, for sufficiently large values of diffusion parameters the limit cycle may not exist.

The important feature of the method of multiple time scales is that in addition to allowing us to construct the nonuniform and limit cycle solutions, it affords information on their stability. The detailed account of the evolution of initial disturbances upon the trivial steady state of the system is thus possible.

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CHAPTER IV

ANALYSIS OF A REACTION-DIFFUSION SYSTEM NEAR HOPF BIFURCATION POINT

The formation of complex spatio-temporal structures in reaction-diffusion systems, can be attributed to the existence of many degrees of freedom simultaneously. Studies reveal that near the critical points, like the Hopf bifurcation point, the system possesses only a few variables that vary on slow time scales while the others varying on fast time scales get eliminated in a projected description. Small amplitude oscillations near the Hopf bifurcation point can then be described in terms of simple evolution equation which acquires a universal form, known as Ginzburg-Landau equation. In the present chapter we aim at deducing this equation form and showing that it is a special case of the more general Schrodinger's equation.

4.1 Introduction

In the last chapter we saw the usefulness of the multi-time scale theory and the redutive perturbation techniques in obtaining the limit cycle and global nonuniform steady patterns in a reaction-diffusion system. In the present chapter we demonstrate yet another application of these, in deducing the behavior near the Hopf bifurcation point. The Hopf bifurcation point is a point at which the original steady state turns unstable and starts performing periodic motion. For various mathematical aspects of Hopf bifurcation one may refer to the book by Marsden and McCracken (1976). Closer to the onset of the periodic behavior all systems, despite their grossly manifested behavior exhibit similar manners.

Reaction-diffusion equations have often been likened to thermodynamic cooperative fields, which are composed of a large number of subfields such as atoms, molecules and magnetic spins. In this context we can look upon reaction-diffusion systems as being composed of a number of local reacting units, which are coupled diffusionally. The local systems are defined as those in which there are no diffusional constraints. It is important to note that in the reaction-diffusion system, each sub-unit can operate in far-from equilibrium conditions, and thus is capable of showing a rich variety of features. In the study of such systems which are composed of a large number of interacting units, we expect to find an equally large number of degrees of freedom coming into play. This is where the slaving principle comes to our rescue. Basically,the slaving priciple claims the possibilty of eliminating a large number of rapidly decaying degrees of freedom. This principle becomes all the more important near the bifurcation points where the system experiences a qualitative change in dynamical behavior. The usefulness of the slaving principle has been rightly emphasized and amply demonstrated by Haken (1983a,b, 1989).

To clarify some basic ideas, let us consider a set of first order differential equations, depicting a reaction scheme. Using the vector notation,

$$\frac{dX}{dt} = F(X;\mu)$$

The steady state solution to such a sytem is an exponential function given as,

$$\begin{pmatrix} x_1(t) \\ . \\ . \\ . \\ x_n(t) \end{pmatrix} = b_1 \begin{pmatrix} c_1^1 \\ . \\ . \\ . \\ c_n^1 \end{pmatrix} \exp(\lambda_1 t) + b_2 \begin{pmatrix} c_1^2 \\ . \\ . \\ . \\ c_n^2 \end{pmatrix} \exp(\lambda_2 t) + \ldots . b_n \begin{pmatrix} c_1^n \\ . \\ . \\ . \\ c_n^n \end{pmatrix} \exp(\lambda_n t)$$

where $(b_1 \ldots b_n)$ are known as the eigenmodes. Close to the instability point, certain eigenmodes are stable and grow in the linear stability analysis, while the others tend to become unstable and decay. The stable modes are known as the order parameters of the system, and the unstable ones as slaved ones. The slaved parametes can be eliminated and explicitly expressed in terms of the order parameters. In a geometrical sense, the decay of all but the stable eigenmodes means that, according to linear approximation, any trajectory starting near the critical point, is led to a few dimensional surface in the phase space. This surface, known as the center manifold, contains the steady state point, and is specified by the stable eigenmodes. Note that at points other than the critical ones, the trajectories are led to an n dimensional space, spanned by all the eigenvectors. In case of a Hopf bifurcation point, the center manifold is two dimensional.

Thus small amplitude oscillations near the Hopf bifurcation point, allow us to describe the behavior of the system in terms of a simple evolution equation, which acquires a universal form. This form known as the Ginzburg-Landau equation (GL equation), is a well-studied equation in physics and mathematics literature (Newell, 1988, Doering et. al., 1988, Ghidaglia and Heron, 1987, Stuart and Diprima, 1978, Hocking and Stewartson, 1972, Kramer and Zimmerman, 1985, Holmes, 1986 Landman, 1987 and Doelman 1989).

In the present chapter we deduce the GL equation for the exponential autocatalysis model. The mathematical tools employed in the derivation of the GL equation are the reductive perturbation methods (Tanuiti, 1968, Newell and Whitehead, 1969, Kuramoto, 1984).

4.2 Derivation of the Ginzburg Landau Equation

For a general homogeneous equation such as,

$$\frac{dX}{dt} = F(X, \mu) \tag{4.2.1}$$

where **X** is a vector representing concentration of chemical species and μ is some control paramter, we shall begin by expressing Eq.(4.2.1) in a deviational form $u = X - X_0$, where X_0 is a steady state solution of Eq.(4.2.1), and expanding it in a Taylor series as,

$$\frac{du}{dt} = Lu + Muu + Nuuu + \ldots. \tag{4.2.2}$$

where,

$$L_{ij} = \frac{\partial F_i(X_o)}{\partial X_{o_j}} \tag{4.2.3a}$$

$$(Muu)_i = \sum_{j,k} \frac{1}{2!} \frac{\partial^2 F_i(X_o)}{\partial X_{o_j} \partial X_{o_k}} u_j u_k \tag{4.2.3b}$$

$$(Nuuu)_i = \sum_{j,k,l} \frac{1}{3!} \frac{\partial^3 F_i(X_o)}{\partial X_{o_j} \partial X_{o_k} \partial X_{o_l}} u_j u_k u_l \tag{4.2.3c}$$

At criticality condition, when a pair of eigenvalue crosses the imaginary axis, Hopf bifurcation occurs.

Referring to Eq. 2, near criticality, the operators and variables involved can be expanded using the Poincáre-Linstédt series, in powers of μ where μ is defined as $(\mu - \mu_c)/\mu_c$. However it is more convenient to define a small parameter ϵ as $\epsilon^2 \chi = \mu$, where $\chi = \mathrm{sgn}\,\mu$.

$$L = L_0 + \chi\epsilon^2 L_1 + \epsilon^4 L_2 + \ldots. \tag{4.2.4a}$$

$$\lambda = \lambda_0 + \chi\epsilon^2 \lambda_1 + \epsilon^4 \lambda_2 + \ldots. \tag{4.2.4b}$$

$$u = \epsilon u_1 + \epsilon^2 u_2 + \ldots. \tag{4.2.4c}$$

$$M = M_0 + \chi\epsilon^2 M_1 + \epsilon^4 M_2 + \ldots. \tag{4.2.4d}$$

$$N = N_0 + \chi\epsilon^2 N_1 + \epsilon^4 N_2 + \ldots. \tag{4.2.4e}$$

Notice that the eigenvalues are also expanded in power series and the λ_v in general are complex, and can be represented as, $\lambda_v = \sigma_v + \omega_v$.

We shall now define the left and right eigenvectors of L_0 corresponding to the eigenvalue $\lambda_0 (= \pm i\omega_0)$, as

$$\text{Right eigenvector } L_0 U = \lambda_0 U \qquad \text{Left eigenvector } U^* L_0 = \lambda_0 U^* \tag{4.2.5a}$$

$$L_0 \bar{U} = \lambda_0 \bar{U} \qquad \bar{U}^* L_0 = \lambda_0 \bar{U}^* \tag{4.2.5b}$$

The right and left eigenvectors U and U^* satisfy a relation, $U^* \bar{U} = \bar{U}^* U = 0$,

and are normalized as, $U^* U = \bar{U}^* \bar{U} = 1$.

Note that the eigenvalues λ_0, λ_1 are given as, $\lambda_0 = i\omega_0 = U^* L_0 U$, $\lambda_1 = \sigma_1 + i\omega_1 = U^* L_1 U$.

The value of ω_0 is given as

$$\omega_0 = \left\{ \frac{(1 + Da_2)^2 - \alpha x_s}{(\alpha x_s - 1)} \right\}^{1/2}$$

Introducing scaling for the time via,

$$\tau = \epsilon^2 t, \tag{4.2.6a}$$

$$\frac{d}{dt} \rightarrow \frac{\partial}{\partial t} + \epsilon^2 \frac{\partial}{\partial \tau} \tag{4.2.6b}$$

and substituting Eq. (4.2.6b) into Eq. (4.2.2) gives,

$$\left(\frac{\partial}{\partial t} + \epsilon^2 \frac{\partial}{\partial \tau} - L_0 - \epsilon^2 \chi L_1 - \ldots \right) (\epsilon u_1 + \epsilon^2 u_2 + \ldots)$$

$$= \epsilon^2 M_0 u_1 u_1 + \epsilon^3 (2 M_0 u_1 u_2 + N_0 u_1 u_1 u_1) + O(\epsilon^4) \tag{4.2.7}$$

Equating coefficients of equal powers of ϵ in Eq. (4.2.7), we obtain,

$$\left(\frac{\partial}{\partial t} - L_0\right) u_\nu = B_\nu, \qquad \nu = 1,2,\ldots \tag{4.2.8}$$

The first few **B** are,

$$B_1 = 0, \tag{4.2.9a}$$

$$B_2 = M_0 u_1 u_1, \tag{4.2.9b}$$

$$B_3 = -\left(\frac{\partial}{\partial \tau} - \chi L_1\right) u_1 + 2M_0 u_1 u_2 + N_0 u_1 u_1 u_1 \tag{4.2.9c}$$

For the system of linear homogeneous Eqs. (4.2.8) we can write a solvability condition,

$$\int_0^{2\pi/\omega_0} U^* . B_\nu e^{-i\omega_0 t} dt = 0 \tag{4.2.10}$$

Since, u_ν are 2π -periodic functions of $\omega_0 t$, the solvability condition in Eq. (4.2.10) finally reduces to,

$$U^* . B_\nu^{(1)}(\tau) = 0 \tag{4.2.11}$$

Then, for $\nu = 1$, we have a neutral solution as,

$$u_1(t,\tau) = W(\tau) U e^{i\omega_0 t} + \text{c.c.} \tag{4.2.12}$$

where c.c. stands for complex conjugate and $W(\tau)$ is some complex amplitude yet to be specified.

Using the solvability condition and neutral solution, for $\nu = 2$, we obtain an expression for u_2

$$u_2 = V_+ W^2 e^{2i\omega_0 t} + V_- \overline{W}^2 e^{-2i\omega_0 t} + V_0 |W|^2 + \nu_0 u_1 \tag{4.2.13}$$

where,

$$V_+ = \overline{V}_- = -(L_0 - 2i\omega_0)^{-1} M_0 U U \tag{4.2.14a}$$

$$V_0 = -2L_0^{-1} M_0 U \overline{U} \tag{4.2.14b}$$

The constant v_0 cannot be determined at this stage, but is not required in the present analysis. Now writing the solvability condition for $v=3$, and knowing $\mathbf{u}_2$, we obtain following form,

$$\frac{\partial W}{\partial \tau} = \chi\lambda_1 W - g|W|^2 W \tag{4.2.15}$$

where the complex variable g is given as,

$$g = g' + ig'' = -2U^* M_0 U V_0 - 2U^* M_0 \bar{U} V_* - 3U^* N_0 U U \bar{U} \tag{4.2.16}$$

Equation (4.2.15) is known as Stuart Landau equation and effectively describes a nonlinear oscillator.

Defining the amplitude **R** and the phase Θ via $W = R\exp(i\Theta)$, we obtain a non-trivial solution,

$$R = R_s, \qquad \Theta = \tilde{\omega} t + \text{const.} \tag{4.2.17a}$$

$$R_s = \sqrt{\sigma_1 / |g'|}, \qquad \tilde{\omega} = \chi(\omega_1 - g'' R_s^2) \tag{4.2.17b}$$

which appears only in the supercritical region (*soft excitation*) ($\chi > 0$) for positive g' and the subcritical region (*hard excitation*) for negative g'. The bifurcating solution shows a perfectly smooth circular motion in the complex **W** plane. Hence, in the end, one can write an expression for the original vector **X** approximately as,

$$\tilde{X} = X_0 + \epsilon \mathbf{u}_1 = X_0 + \epsilon\left\{ U R_s \exp\left[i\left(\omega_0 + \epsilon^2 \tilde{\omega} \right) t \right] + \text{c.c.} \right\} \tag{4.2.18}$$

which describes a finite amplitude elliptic orbital motion in the critical eigenplane.

Now extending the above analysis for the reaction-diffusion system, we have an additional term accounting for the diffusion as,

$$\frac{\partial \mathbf{u}}{\partial t} = (L + D\nabla^2)\mathbf{u} + \mathbf{Muu} + \mathbf{Nuuu} + \ldots. \tag{4.2.19}$$

Allowing for slowly varying space coordinate apart from two time scales t and τ characterized by the slowness parameter ($\epsilon = |\mu|^2$), the extra space dependence of ϵ is embedded in a scaled coordinate defined by, $s = \epsilon r$. Also, transforming the Laplacian as, $\nabla \to \epsilon \nabla_s$, we have,

$$\left(\frac{\partial}{\partial t} + \epsilon^2 \frac{\partial}{\partial \tau} - \epsilon^2 D \nabla_s^2 - L_0 - \epsilon^2 \chi L_1 - \ldots\right) \; (\epsilon u_1 + \epsilon^2 u_2 + \ldots)$$

$$= \epsilon^2 M_0 u_1 u_1 + \epsilon^3 (2 M_0 u_1 u_2 + N_0 u_1 u_1 u_1) + O(\epsilon^4) \qquad (4.2.20)$$

With the additional term for the transformed Laplacian and s dependence, the remaining conditions can be rewritten, and we finally obtain the reduced equation as,

$$\frac{\partial W}{\partial \tau} = \chi \lambda_1 W + d \nabla_s^2 W - g |W|^2 W \qquad (4.2.21)$$

where d is generally a complex number defined as,

$$d = d' + i d'' = U^* D U \qquad (4.2.22)$$

Further using the transformation,

$$(\tau, s, W) \to (\sigma^{-1} \tau, \sqrt{d'/\sigma_1}\, s, \sqrt{\sigma_1 / |g'|}\, W) \qquad (4.2.23)$$

and rewriting s and t as r and t, Eq. (4.2.21) reduces to a more convenient form,

$$\frac{\partial W}{\partial t} = (1 + i c_0) W + (1 + i c_1) \nabla^2 W - (1 + i c_2) |W|^2 W \qquad (4.2.24)$$

where,

$$c_0 = \omega_1 / \sigma_1, \quad c_1 = d''/d', \quad c_2 = g''/g' \qquad (4.2.25)$$

and the bifurcation has been assumed to be supercritical. Subsequently, the transformation $W \to W \exp(i c_0 t)$ eliminates c_0 and reduces Eq. (4.2.24) to the form,

$$\frac{\partial W}{\partial t} = W + (1 + i c_1) \nabla_x^2 W - (1 + i c_2) |W|^2 W \qquad (4.2.26)$$

This form can be identified as a special case of the more general Schrodinger's equation. In the next section we shall apply this technique to the case of reaction-diffusion scheme representing exponential autocatalysis.

4.3 GL equation for exponential autocatalysis model

Following the general procedure outlined in section 4.2, we obtain the following specific results for a case of exponential autocatalysis. Beginning with Eqs. (4.2.3) and (4.2.4) the operators L_0 and L_1 can be identified as,

$$L_0 = \frac{1}{(\alpha x_s - 1)} \begin{bmatrix} -(\alpha x_s + Da_2 + 1) & -\alpha x_s (Da_2 + 2) \\ Da_2 + 2 & \alpha x_s + Da_2 + 1 \end{bmatrix} \qquad (4.3.1a)$$

$$L_1 = \frac{Da_2 + 2}{\alpha x_s - 1} \begin{bmatrix} -1 & -\alpha x_s \\ 1 & \alpha x_s \end{bmatrix} \qquad (4.3.1b)$$

Further defining A and B as,

$$A = \frac{-(\alpha x_s + Da_2 + 1)}{\alpha x_s (Da_2 + 2)}, \qquad B = \frac{\alpha x_s - (1 + Da_2)^2}{\omega_o \alpha x_s (Da_2 + 2)} \qquad (4.3.2)$$

we obtain the eigenvectors as solutions to Eqs. (4.2.5a) and (4.2.5b),

$$U = \begin{pmatrix} 1 \\ (A + iB) \end{pmatrix} \qquad \bar{U} = \begin{pmatrix} 1 \\ (A - iB) \end{pmatrix} \qquad (4.3.3a)$$

$$U^* = \frac{1}{2B} (B + iA, \; -i) \qquad \bar{U}^* = \frac{1}{2B} (B - iA, \; i) \qquad (4.3.3b)$$

The eigenvalue λ_1 defined as, $\lambda_1 = \sigma_1 + i\omega_1$, gives us,

$$\sigma_1 = \frac{-(Da_2 + 2)}{2B(\alpha x_s - 1)} [B(1 - \alpha x_s)] \qquad (4.3.4a)$$

$$\omega_1 = \frac{-(Da_2 + 2)}{2B(\alpha x_s - 1)} [\alpha x_s (B^2 + A^2) + A(\alpha x_s + 1) + 1] \qquad (4.3.4b)$$

Also, the parameter d defined in Eq. (4.2.22) is given by,

$$d' = \frac{1}{2} (D_1 + D_2) \qquad d'' = \frac{A}{2B} (D_1 - D_2) \qquad (4.3.5)$$

The vectors **M** and **N** are given as :

The vectors **M** and **N** are given as :

$$\mathbf{M}_0 = \frac{\alpha(Da_2+2)}{2(\alpha x_s-1)} \begin{pmatrix} 0 & -1 & -1 & 0 \\ 0 & 1 & 1 & 0 \end{pmatrix} \tag{4.3.6}$$

and $\mathbf{N} = 0$,

while the vectors $\mathbf{V}_+$ and $\overline{\mathbf{V}}_-$, given by Eq. (4.2.14a), are,

$$\mathbf{V}_+ = \overline{\mathbf{V}}_- = \frac{\alpha(Da_2+2)(A+iB)}{\eta} \begin{pmatrix} (\alpha x_s + Da_2 + 1) - 2i\omega_o - \alpha x_s(Da_2+2) \\ -(Da_2+2) + (\alpha x_s + Da_2 + 1) + 2i\omega_o \end{pmatrix} \tag{4.3.7a}$$

where

$$\eta = \{\alpha x_s(Da_2+2)^2 - [(\alpha x_s + Da_2 + 1)^2 + 4\omega_o^2]\} \tag{4.3.7b}$$

The vector $\mathbf{V}_0$ given in Eq. (4.2.14b) becomes,

$$\mathbf{V}_0 = \frac{2A\alpha(Da_2+2)}{\{(\alpha x_s-1)[(1+Da_2)^2-\alpha x_s]\}} \begin{pmatrix} (\alpha x_s + Da_2 + 1) - \alpha x_s(Da_2+2) \\ -(Da_2+2) + (\alpha x_s + Da_2 + 1) \end{pmatrix} \tag{4.3.8}$$

The term **g** in Eq. (4.2.16), can now be written as,

$$g' = \frac{[\alpha(Da_2+2)]^2}{2B(\alpha x_s-1)(\Omega_1^2 - 4\Omega_1\omega_0^2)}(B\Omega_2 - \Omega_3(A+1)) \tag{4.3.9a}$$

$$g'' = \frac{[\alpha(Da_2+2)]^2}{2B(\alpha x_s-1)(\Omega_1^2 - 4\Omega_1\omega_0^2)}(\Omega_2(A+1) + B\Omega_3) \tag{4.3.9b}$$

where,

$$\Omega_1 = (\alpha x_s-1)[(1+Da_2)^2 - \alpha x_s] \tag{4.3.10a}$$

$$\begin{aligned} \Omega_2 = {} & (\Omega_1 - 4\omega_0^2)(2A(\alpha x_s-1) - 2A^2(\alpha x_s-1)(1+Da_2)) \\ & + \Omega_1[A((\alpha x_s-1) - A(\alpha x_s-1)^2(1+Da_2) - 2B\omega_0) - \omega_0 \\ & + B(\alpha x_s-1)^2(1+Da_2) - 2A\omega_0)] \end{aligned} \tag{4.3.10b}$$

$$\begin{aligned} \Omega_3 = {} & -(\Omega_1 - 4\omega_0^2)(-2AB(1-\alpha x_s)^2(1+Da_2)) \\ & + \Omega_1[B((\alpha x_s-1) - A(\alpha x_s-1)^2(1+Da_2) - 2B\omega_0) \\ & + A(2\omega_0 + B(\alpha x_s-1)^2(1+Da_2) - 2A\omega_0)] \end{aligned} \tag{4.3.10c}$$

$$c_o = \frac{\alpha x_s(B^2+A^2)+A(\alpha x_s+1)+1}{B(1-\alpha x_s)} \qquad (4.3.11a)$$

$$c_1 = \frac{B(\gamma^2-1)}{A(\gamma^2+1)} \qquad (4.3.11b)$$

$$c_2 = \frac{\Omega_2(A+1)+B\Omega_3}{B\Omega_2-\Omega_3(A+1)} \qquad (4.3.11c)$$

where,

$$\gamma = \sqrt{D_1/D_2} \qquad (4.3.12)$$

4.4 Results and Discussion

The constants c_0, c_1 and c_2 as obtained above define the Ginzburg-Landau Eq. (4.2.24) for the exponentially autocatalysed reaction-diffusion system. Evaluation of these constants and therefore that of the Ginzburg-Landau equation is central to the any further development such as obtaining the plane waves, rotating waves, turbulence and entrainment phenomena in discrete oscillators. The GL equation is capable of showing a variety of solutions, and is shown to be of great practical importance in the self organization phenomena. Having derived the GL equation for the exponential autocatalysis model (or for any other system of interest), the conditions for the occurrence of the various types of solutions, to the GL equation, in the realistic parameter regimes can also be easily derived. However, we will not elaborate upon this point, here. The central point of importance of the work presented in this chapter is the derivation of the GL equation and the evaluation of the constants c_0, c_1 and c_2.

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CHAPTER V

ANALYSIS OF SUBSTRATE-INHIBITION KINETIC MODEL WITH THE HELP OF GINZBURG LANDAU EQUATION

A simple biochemically feasible mechanism which relies on substrate-inhibition kinetics, also known as Seelig's is analysed near the points of instability, with the help of the GL equation. To this end the instability conditions for two well known types of instabilities are derived. Further we derive the GL equation for this model and obtain the numerical solutions to this equation in the realistic parameter regimes.

5.1 Introduction

The spontaneous formation of well organized structures is a commonly observed phenomenon in all fields of science. In biology, origin of information, formation of patterns during the development of embryo, well developed coat marks in mammals, regeneration of organs, patterning during cellular aggregation are some well studied problems (Melton 1991, Davidenko *et al*, 1991, Hanyu and Matsumoto, 1991, Shapiro and Trubatch, 1991, Siegert and Weijer, 1991, Steinbock *et al*, 1991 and Lechleiter, 1991). Following Turing's classical work, reaction-diffusion mechanisms have been widely claimed to be active in pattern generation (Meinhardt, 1982). A simple oscillatory mechanism which relies on substrate-inhibition kinetics was proposed by Seelig (1976a,b). This model is claimed to be chemically more feasible and realistic than the Brusselator and the Oregonator models. Murray and Mimura (1978) have studied this model in presence of diffusion, with a view to analyze the diffusional instabilities. This model has also been studied in the context of transmission of chemical signals from one part of an organism to another (Britton and Murray, 1979). Mimura and Murray pointed out that beyond the diffusional instability point, the mode of the finite amplitude spatial structure is equal to the mode of the linearly growing mode in the stability analysis. This shows that near the instability points the system behavior can be described in terms of the linearly growing modes, or in other words the order parameters. Thus we can analyze the present model with the help of the GL equation. In the present chapter we study two types of instabilities that can arise in the system and derive the conditions for their occurrence. Further we derive the GL equation for this model with a view to study the evolution of the system near these instability points and obtain the numerical solutions to this equation in the realistic parameter regimes.

5.2 The Model

The overall reaction described by $X + Y = P + Q$ is effected by an enzyme M. The substrates X and Y are supplied to the cell by constant fluxes J_1 and J_2 respectively and

X is constantly removed by a first order reaction efflux. The enzyme substrate complex MX further combines with Y to give the final products. At high concentrations the substrate X inhibits the enzyme by forming an inert complex MX_2. The governing equations for this reaction mechanism are given as,

$$\frac{dX}{dt} = J_1 - k_0 X - R(X,Y) \tag{5.2.1}$$

$$\frac{dY}{dt} = J_2 - R(X,Y) \tag{5.2.2}$$

where,

$$R(X,Y) = \frac{\{k_{-1}.M_0.(k_1X/k_{-1}).(k_3Y/k_{-1})\}}{\{1+(k_1X/k_{-1})+(k_3Y/k_{-1})+(k_2k_{-1}/k_{-2}k_1).(k_1.X/k_{-1})^2\}}$$

After appropriate non dimensionalization Eq. (5.2.1) and (5.2.2) can be written as

$$\frac{dx}{dt} = j_1 - x - \beta.r(x,y) = f_1 \tag{5.2.3}$$

$$\frac{dy}{dt} = j_2 - \gamma.r(x,y) = f_2 \tag{5.2.4}$$

where,

$$r(x,y) = \frac{xy}{1+x+y+kx^2}$$

The steady state solutions for this system are

$$x_s = \frac{\gamma j_1 - \beta j_2}{\gamma} \qquad\qquad y_s = \frac{j_2(1+x_s+kx_s^2)}{\gamma x_s - j_2}$$

After substituting for the value of x_s in the expression for y_s we further get,

$$y_s = \frac{j_2\{k\gamma^2 j_1^2 + j_1\gamma a_3 + a_4\}}{\gamma^2(\gamma j_1 + a_2)} \tag{5.2.5}$$

where,

$$a_1 = -\beta j_2 \qquad\qquad a_3 = \gamma + 2ka_1$$

$\alpha_2 = -(\beta + 1) j_2 = \alpha_1 - j_2$ $\alpha_4 = (k\alpha_1^2 + \gamma\alpha_1 + \gamma^2)$

In order to get realistic solutions for x_s and y_s, we impose the condition, $\gamma j_1 + \alpha_1 > 0$ on the parameter values.

Defining the deviations as $u = x - x_s$, and $v = y - y_s$, the deviation equations can be written as,

$$\frac{du}{dt} = -u + \left\{ \frac{(j_1 - x_s)(u + v + ku^2 + 2kux_s) - \beta(x_s v + y_s u + uv)}{(1 + x_s + y_s + kx_s^2) + (u + v + ku^2 + 2kux_s)} \right\} \tag{5.2.6}$$

$$\frac{dv}{dt} = \left\{ \frac{j_2(u + v + ku^2 + 2kux_s) - \gamma(x_s v + y_s u + uv)}{(1 + x_s + y_s + kx_s^2) + (u + v + ku^2 + 2kux_s)} \right\} \tag{5.2.7}$$

The reaction-diffusion equations corresponding to (5.2.3), and (5.2.4) are,

$$\frac{\partial x}{\partial t} = D_1 \frac{\partial^2 x}{\partial r^2} + j_1 - x - (\beta xy / 1 + x + y + kx^2) \tag{5.2.8}$$

$$\frac{\partial y}{\partial t} = D_2 \frac{\partial^2 y}{\partial r^2} + j_2 - (\gamma xy / 1 + x + y + kx^2) \tag{5.2.9}$$

where D_1, and D_2 are the diffusivities of the species x and y respectively. The corresponding deviation equations are,

$$\frac{\partial u}{\partial t} = D_1 \frac{\partial^2 u}{\partial r^2} - u + \left\{ \frac{(j_1 - x_s)(u + v + ku^2 + 2kux_s) - \beta(x_s v + y_s u + uv)}{(1 + x_s + y_s + kx_s^2) + (u + v + ku^2 + 2kux_s)} \right\} \tag{5.2.10}$$

$$\frac{\partial v}{\partial t} = D_2 \frac{\partial^2 v}{\partial r^2} + \left\{ \frac{j_2(u + v + ku^2 + 2kux_s) - \gamma(x_s v + y_s u + uv)}{(1 + x_s + y_s + kx_s^2) + (u + v + ku^2 + 2kux_s)} \right\} \tag{5.2.11}$$

The deviations are assumed to be proportional to $e^{(iqr + \lambda t)}$, where q represents a wave number, and λ, the eigenvalue.

The Jacobian of the system can then be written as,

$$J = \left(\frac{1}{1 + x_s + y_s + kx_s^2} \right) \begin{bmatrix} -(1 + x_s + y_s + kx_s^2) - \beta y_s + (1 + 2kx_s)(j_1 - x_s) & -\beta x_s + j_1 - x_s \\ -\gamma y_s + (1 + 2kx_s) j_2 & -\gamma x_s + j_2 \end{bmatrix} \tag{5.2.12}$$

At this point we define the following terms for convenience

$$F_1 = \frac{\gamma^2(\gamma j_1 + a_2)}{(\gamma j_1 + a_1)(k\gamma^2 j_1^2 + j_1\gamma a_3 + a_4)} \qquad F_4 = (\gamma j_1 + a_2)$$

$$F_2 = y_s \qquad F_5 = \frac{(-a_5 j_1^3 + a_6 j_1^2 + a_7 j_1 + a_8)}{\gamma^2 F_4}$$

$$F_3 = \frac{\gamma + 2k(\gamma j_1 + a_1)}{\gamma}$$

where,

$$a_5 = k\gamma^3 \qquad a_7 = -\gamma(a_1(a_3 + 2ka_2) + a_4)$$

$$a_6 = -(2k\gamma^2 a_1 + \gamma^2 a_3) \qquad a_8 = -a_1 a_2 a_3$$

we can now write Eq. (5.2.12) as

$$J = F_1 \begin{bmatrix} \frac{\beta}{\gamma} j_2 F_3 - \beta F_2 - \frac{1}{F_1} & -\frac{\beta}{\gamma}(\gamma j_1 + a_2) \\ j_2 F_3 - \gamma F_2 & -(\gamma j_1 + a_2) \end{bmatrix} \tag{5.2.13}$$

We now analyze the stability of the steady state (x_s, y_s) by linearizing Eq. (5.2.10) and (5.2.11) in u and v.

$$\lambda^2 - \lambda A(q) + B(q) = 0 \tag{5.2.14}$$

where,

$$A(q) = \{q^2(D_1 + D_2) - (F_5 - F_4)\} \tag{5.2.15a}$$

$$B(q) = \{q^4 D_1 D_2 - q^2(D_2 F_5 - D_1 F_4 + (F_5/F_1)\} \tag{5.2.15b}$$

In the analysis that follows we chose j_1 as the control parameter, since it can be externally controlled and we express the conditions for occurrence of instability in terms of this parameter. Two types of instability, and the conditions for their occurrence have been discussed by Kuramoto (1984). Type I instability is a kinetic instability, which occurs when $A(q_c) = 0$ alongwith $q_c = 0$. The subscript 'c' denotes the critical condition as usual. This condition can be stated in terms of j_{1c} as

$$a_5 j_{1c}^3 - a_9 j_{1c}^2 - a_{10} j_{1c} - a_{11} = 0 \qquad (5.2.16)$$

where,

$$a_9 = a_6 - \gamma^4 \qquad a_{10} = a_7 - 2\gamma^3 a_2 \qquad a_{11} = a_8 - \gamma^2 a_2^2$$

The condition stated in Eq. (5.2.16) implies that for all $j_1 > j_{1c}$ the system evolves to an oscillatory state.

The critical value $j_1 = j_{1c}'$, for the occurrence of type II instability can be obtained from the set of conditions $B(q)_c = 0$ and $dB(q_c)/dq_c = 0$ in Eq. (5.2.14). We find that this set of conditions gives type II instability condition as,

$$(D_2 F_{4c}')^2 + (D_1 F_{5c}')^2 - 2D_1 D_2 F_{6c}' = 0 \qquad (5.2.17)$$

where,

$$F_6 = (a_5 j_1^3 + a_{12} j_1^2 + a_{13} j_1 + a_{14})/(\gamma^2 F_4)$$

and,

$$a_{12} = \gamma^2 a_3 \qquad a_{13} = \gamma(a_1 a_3 + a_4 - 2k a_1 a_2) \qquad a_{14} = a_1(2a_4 - a_2 a_3)$$

This further leads to the statement of the type II instability condition as,

$$j_{1c}'^6(D_2^2 b_1) + j_{1c}'^5(D_2^2 b_2 - 2D_1 D_2 b_3) + j_{1c}'^4(D_2^2 b_4 + D_1^2 b_5 - 2D_1 D_2 b_6)$$
$$+ j_{1c}'^3(D_2^2 b_7 + D_1^2 b_8 - 2D_1 D_2 b_9) + j_{1c}'^2(D_2^2 b_{10} + D_1^2 b_{11} - 2D_1 D_2 b_{12})$$
$$+ j_{1c}'(D_2^2 b_{13} + D_1^2 b_{14} - 2D_1 D_2 b_{15}) + (D_2^2 b_{16} + D_1^2 b_{17} - 2D_1 D_2 b_{18}) = 0 \qquad (5.2.18)$$

where,

$$b_1 = a_5^2 \qquad b_{10} = a_7^2 + 2a_6 a_8$$
$$b_2 = -2a_5 a_6 \qquad b_{11} = 6\gamma^6 a_2^2$$
$$b_3 = \gamma^4 a_5 \qquad b_{12} = \gamma^2(\gamma^2 a_{14} + 2\gamma a_2 a_{13} + a_2^2 a_{12})$$
$$b_4 = a_6^2 - 2a_5 a_7 \qquad b_{13} = 2a_7 a_8$$
$$b_5 = \gamma^8 \qquad b_{14} = 4(\gamma a_2)^3$$
$$b_6 = \gamma^3(\gamma^2 a_{12} + 2a_2 a_5) \qquad b_{15} = \gamma^2 a_2(2\gamma a_1 + a_2 a_{13})$$

$b_7 = 2(a_6 a_7 - a_5 a_8)$ $\qquad$ $b_{16} = a_8^2$

$b_8 = 4\gamma^7 a_2$ $\qquad$ $b_{17} = (\gamma a_2)^4$

$b_9 = \gamma^2(\gamma^2 a_{13} + 2\gamma a_2 a_{12} + a_2 a_5)$ $\qquad$ $b_{18} = a_{14}(\gamma a_2)^2$

Equation (5.2.4) denotes the diffusive instability condition , which is better known in literature as the Turing instability condition. Beyond j_{1c}' , the system can show interesting spatio-temporal patterns. Some interesting results obtained for the present model are discussed in Section.5.5. The conditions for the two types of instabilities discussed above are shown in Fig. 5.1 - 5.4 where j_{1c}, and j_{1c}' are mapped against j_2 , for a fixed set of the other parameters. In the regions where type I instability sets in before type II, the predominant feature of the system is spatially uniform oscillations, which however, can become unstable due to the diffusional effects, depending upon the magnitudes of D_1 and D_2 . This in turn is reflected in the magnitudes of c_1 and c_2 of the GL equation. This point has been further elaborated in Chapter VII.

The characteristic frequency of the system, ω_0 is obtained from Eq. (5.2.14), noting that at the onset of oscillations , λ becomes purely imaginary and $\lambda = \pm i\omega_0$

$$\omega_0 = \left\{(a_5 j_1^3 + a_{15} j_2^2 + a_{16} j_1 + a_{17})^{1/2}\right\}/\gamma \qquad (5.2.19)$$

where, $a_{15} = \gamma(k a_1 + a_3)$ $\qquad$ $a_{16} = \gamma(a_1 a_3 + a_4)$ $\qquad$ $a_{17} = a_1 a_4$

5.3 Derivation of GL equation for the substrate inhibition kinetic model

Following the procedure outlined in the previous chapter we shall now define the GL equation for the present model. We define the parameter μ as, $\mu = (j_1 - j_{1c}/j_{1c})$, and ϵ as $\epsilon^2 \chi = \mu$. Using these definitions in developing the perturbation series in powers of μ , we obtain the operators L_0 and L_1 .

$$\mathbf{L}_0 = F_7 \begin{bmatrix} l_{01} & -\beta l_{02} \\ l_{03} & -\gamma l_{02} \end{bmatrix} \tag{5.3.1}$$

$$\mathbf{L}_1 = F_7 j_{1c} \left\{ \begin{bmatrix} l_{11} & -\beta l_{12} \\ l_{13} & -\gamma l_{12} \end{bmatrix} - F_8 L_0 \right\} \tag{5.3.2}$$

where

$F_7 = (1/\gamma\omega_0)^2$ $F_8 = (3a_5 j_{1c}^2 + 2a_{15} j_{1c} + a_{16})$

$l_{01} = (-a_5 j_{1c}^3 + a_6 j_{1c}^2 + a_7 j_{1c} + a_8)$ $l_{11} = (-3a_5 j_{1c}^2 + 2a_6 j_{1c} + a_7)$

$l_{02} = \gamma(\gamma^2 j_{1c}^2 + l_4 j_{1c} + l_5)$ $l_{12} = \gamma(2\gamma^2 j_{1c} + l_4)$

$l_{03} = (l_1 j_{1c}^2 + l_2 j_{1c} + l_3)$ $l_{13} = (l_1 j_{1c} + l_2)$

and

$l_1 = 2k\gamma^3 j_2$ $l_2 = 2k\gamma^2 j_2 a_2$ $l_3 = \gamma j_2(a_2 a_3 - a_4)$ $l_4 = 2\gamma a_2$ $l_5 = a_2^2$

Note that according to Eq.(5.2.14) we have

$$l_{01} - \gamma l_{02} = 0 \tag{5.3.3}$$

Defining M and N according to Eq. (4.2.3), we obtain

$$M_0 = \frac{1}{2} F_9 \begin{bmatrix} \beta m_{01} & \beta m_{02} & \beta m_{02} & \beta m_{03} \\ \gamma m_{01} & \gamma m_{02} & \gamma m_{02} & \gamma m_{03} \end{bmatrix} \tag{5.3.4}$$

where,

$$F_9 = \left(\frac{\gamma^4(\gamma^2 j_{1c}^2 + l_4 j_{1c} + l_5)}{j_{1c}^6 m_1 + j_{1c}^5 m_2 + j_{1c}^4 m_3 + j_{1c}^3 m_4 + j_{1c}^2 m_5 + j_{1c} m_6 + m_7} \right.$$

$m_{01} = (2j_2/\gamma^2 F_4)\{2a_5 j_{1c}^3 + m_8 j_{1c}^2 + m_9 j_{1c} + m_{10}\}$

$m_{02} = \{k(\gamma^2 j_{1c}^2 + 2\gamma a_1 j_{1c} + a_1^2) - \gamma^2\}/\gamma$

$m_{03} = 2F_4$

and,

$m_1 = a_5^2$ $m_5 = a_{16}^2 + a_{15} a_{17}$ $m_9 = 2k\gamma(\gamma a_3 + a_4)$

$m_2 = 2a_5 a_{15}$ $m_6 = 2a_{16} a_{17}$ $m_{10} = a_3 a_4$

$m_3 = a_{15}^2 + a_5 a_{16}$ $m_7 = a_{17}^2$

$m_4 = 2(a_{15}a_{16} + a_5 a_{17})$ $\quad m_8 = k a_3 \gamma^2$

Note that in the derivation of M_0 the $(1/(1 + x_s + y_s + k x_s^2))^4$ terms have been neglected, assuming that their contribution, compared to the other terms is negligible. Arguing on the same lines, N_0 can shown to reduce to zero. The left and right eigenvectors corresponding to the eigenvalue $\lambda_0 (= \pm i\omega_0)$ are,

$$\mathrm{U} = \begin{pmatrix} 1 \\ (P_1 + iQ) \end{pmatrix} \qquad \bar{\mathrm{U}} = \begin{pmatrix} 1 \\ (P_1 - iQ) \end{pmatrix} \tag{5.3.5a}$$

$$\mathrm{U}^* = \frac{1}{2Q}(Q - iP_2, \; -i) \qquad \bar{\mathrm{U}}^* = \frac{1}{2Q}(Q + iP_2, \; i) \tag{5.3.5b}$$

where $P_1 = \gamma/\beta \quad P_2 = -(l_{01}/\beta l_{02})$ and, $Q = (\gamma l_{01} - \beta l_{03})/\beta\gamma^2\omega_0^3$

Note that $P_1 + P_2 = 0$ following Eq.(5.3.3)

The vectors V_- and $\bar{\mathrm{V}}_-$, given by Eq.(4.2.14a) are

$$\mathrm{V}_- = \bar{\mathrm{V}}_- = \frac{v_1 F_9}{2(\eta + 4F_7\omega_0^2)} \begin{pmatrix} -\beta(\gamma l_{02} + 2i\omega_0) + \beta\gamma l_{02} \\ -\beta l_{03} + \gamma(l_{01} - 2i\omega_0) \end{pmatrix} \tag{5.3.6}$$

where,

$\eta_1 = l_{02}(\gamma l_{01} - \beta l_{02})$

$v_1 = m_{01} + 2m_{02}(P_1 + iQ) + m_{03}(P_1 + iQ)^2$

The vector V_0 given in Eq.(4.2.14b) is

$$\mathrm{V}_0 = \frac{F_9}{\eta_1} \begin{pmatrix} 0 \\ v_2 v_3 \end{pmatrix} \tag{5.3.7}$$

where,

$v_2 = (\gamma l_{01} - \beta l_{03})$

$v_3 = (m_{01} + 2m_{02}P_1 + m_{03}(P_1^2 + Q^2))$

The complex variable g in Eq.(4.2.16), can now be written as

$$g = g' + g'' = (-F_9 R_2/2Q)\{(\beta Q R_7 + I_7(\gamma + \beta P_2)) + i(\beta Q I_7 - R_7(\gamma + \beta P_2))\} \tag{5.3.8}$$

where,

$\eta_2 = 2(\eta_1 + 4\omega_0^2)$

$R_1 = m_{01} + 2m_{02}P_1 + m_{03}(P_1^2 - Q^2)$

$I_1 = 2Q(m_{02} + m_{03}P_1)$

$R_2 = \dfrac{F_9}{F_7 \eta_1 \eta_2}$

$R_3 = 2\eta_1 I_1 \omega_0 \beta$

$I_3 = -2\eta_1 R_1 \omega_0 \beta$

$R_4 = \eta_1(R_1 v_2 + I_1(2\omega_0\beta)) + v_2 v_3 \eta_2$

$I_4 = \eta_1(I_1 v_2 - R_1(2\omega_0\beta))$

$R_5 = \eta_1(2\omega_0\beta(-R_1 Q + I_1 P_1))$

$I_5 = -\eta_1(2\omega_0\beta(I_1 Q + R_1 P_1))$

$R_6 = \eta_1(R_1(P_1 v_2 - 2\omega_0 \gamma Q) + I_1(Q v_2 + 2\omega_0 \gamma v_2)) + P_1 v_2 v_3 \eta_2$

$I_6 = (I_1(P_1 v_2 - 2\omega_0 \gamma Q) - R_1(Q v_2 + 2\omega_0 \gamma v_2)) + Q v_2 v_3 \eta_2$

$R_7 = (m_{01} R_3 + m_{02}(R_4 + R_5) + m_{03} R_6)$

$I_7 = (m_{01} I_3 + m_{02}(I_4 + I_5) + m_{03} I_6)$

The eigenvalue λ_1 gives us the values of σ_1 and ω_1 as,

$$\sigma_1 = \frac{F_7 j_{1c}}{2Q}\ (Q(L_{11} - \beta L_{12}(P_1 + P_2) - \gamma L_{12})) \tag{5.3.9a}$$

$$\omega_1 = \frac{F_7 j_{1c}}{2Q}\ (P_2(\beta L_{12} P_1 - L_{11}) - (Q^2 \beta L_{12} + L_{13} + \gamma L_{12} P_1)) \tag{5.3.9b}$$

where,

$L_{11} = F_7(l_{11} - F_7 F_8 l_{01})$ $\quad L_{12} = F_7(l_{12} - F_7 F_8 l_{02})$ $\quad L_{13} = F_7(l_{13} - F_7 F_8 l_{03})$

The parameter d defined in Eq. (4.2.22) is

$$d' = \frac{1}{2}Q\ (D_1 + D_2) \qquad d'' = \frac{1}{2Q}\ (D_1 P_2 + D_2 P_1) \tag{5.3.10}$$

Finally the constants in the GL equation c_0, c_1 and c_2, defined in Eq. (4.2.25) are found to be

$$c_0 = \frac{-(P_2(L_{11}-\beta L_{12}P_1)+Q(\beta L_{12}Q)+(L_{13}-\gamma L_{12}P_1))}{(Q(L_{11}-\beta L_{12}P_1)-L_{12}Q(\beta P_2+\gamma))} \quad (5.3.11a)$$

$$c_1 = -\left(\frac{D_1P_2+D_2P_1}{Q(D_1+D_2)}\right) \quad (5.3.11b)$$

$$c_2 = \frac{(\beta Q I_7 - R_7(\gamma+\beta P_2))}{(\beta Q R_7 + I_7(\gamma+\beta P_2))} \quad (5.3.11c)$$

5.4 Numerical Solution to the GL equation

In the present study we make use of the method of lines after discretizing the space dimension using orthogonal collocation. The original equation is split in to its real and imaginary components and solved simultaneously with the appropriate initial and boundary conditions. Hence, the original infinite dimensional system is replaced by a 2 N dimensional system where N is the number of space discretization points in [0,1]. No appreciable change was noted using orthogonal collocation on two finite elements. Hence, we confine our study to the method of lines with twelve-point collocation.

Equations (5.2.16) and (5.2.18) are solved in order to demarcate the regions of occurrence of type I and type II instabilities. We find that the solution to the GL equation shows a remarkable difference in its behavior in these two regimes. We have chosen representative parameters for these two zones and computed the corresponding values of GL constants namely, c_0, c_1 and c_2 from Eq. (5.3.11). Having obtained the values of c_0, c_1 and c_2 we go on to solve the transformed GL equation (4.2.26). Some of the interesting results obtained are discussed in the next section. The parameter values are chosen based on the analysis carried out by Mimura and Murray (1978).

5.5 Results and Discussion

Figure 5.1 and 5.2 show that a multiplicity of the instability conditions can exist in certain parameter regimes. Figure 5.1 shows the results for $\beta = 44.0$ whereas Fig. 5.2 presents the results for $\beta = 20.0$. The other parameters are $\gamma = 0.984$, $D_1 = 0.005$, $D_2 = 1.5$. In Fig.

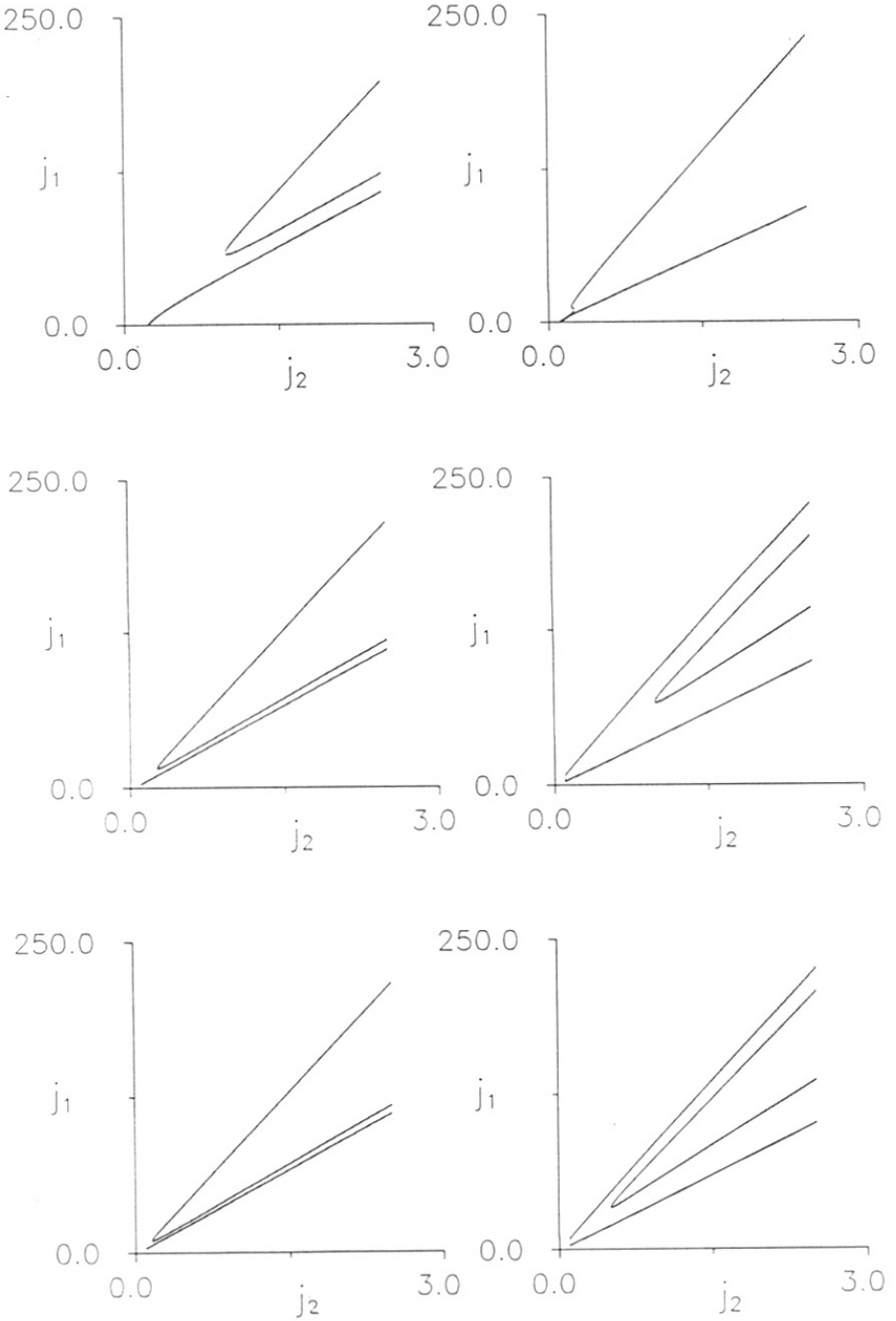

Fig. 5.1 : a, c and e: Occurrence of type I instability for $k = 0.1, 0.5, 1$ respectively; b, d and f show the type II instability for the same parameter space; ($\beta = 44$).

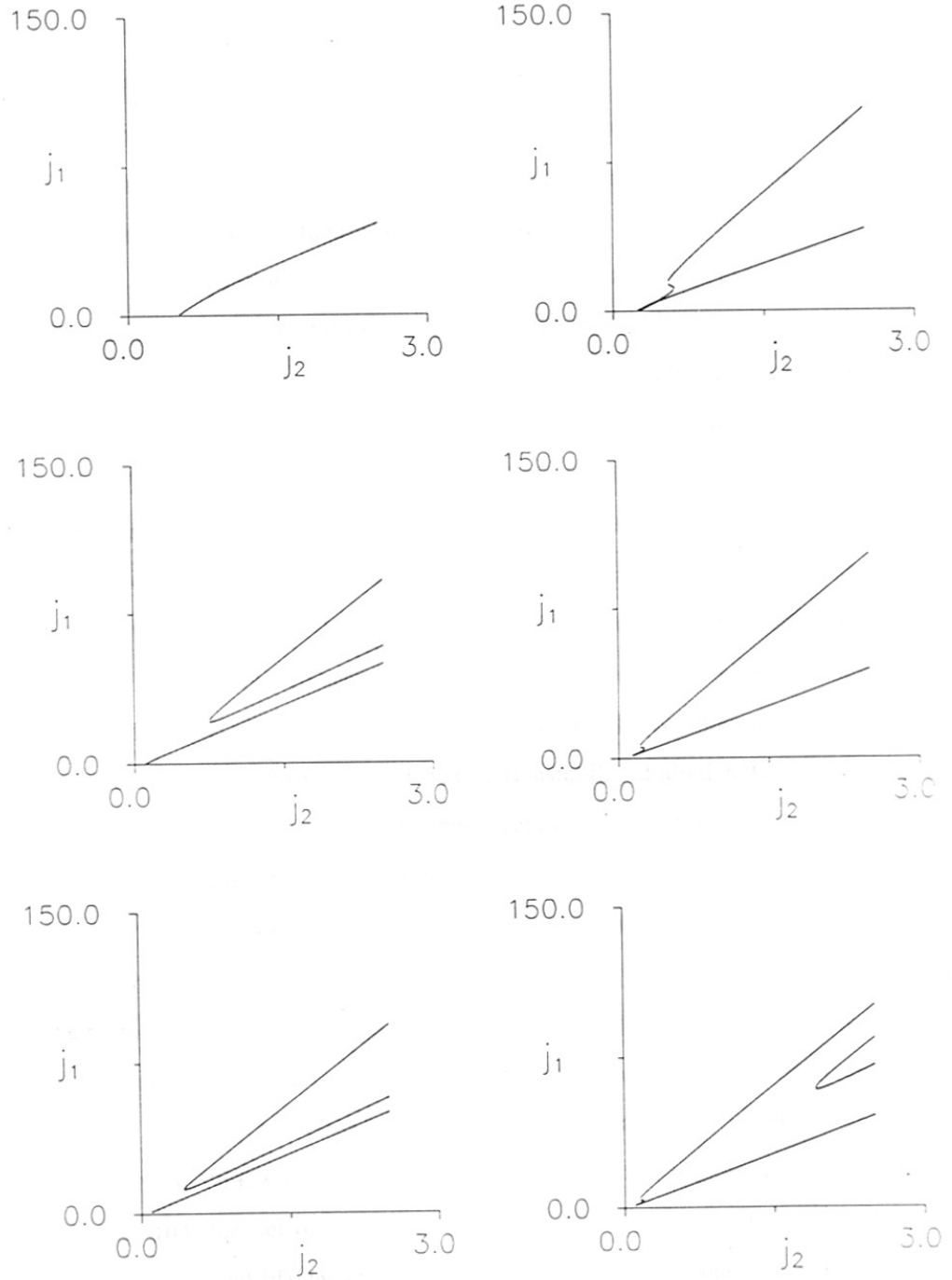

Fig. 5.2 : Occurrence of type I and type II instability conditions for $\beta = 20$: The type of instability and the values of k are as discussed in Fig. 5.1.

5.1 and 5.2 (a) and (b) the value of $k = 0.1$ while for (c) and (d) $k = 0.5$ and for (e) and (f) $k = 1.0$. The figures clearly indicate that for certain parameter values type I instability occurs before type II and vice versa. In the regions where type I instability occurs before type II the possibility of diffusion driven instabilities is eliminated.

Fig. 5.3 depicts the behavior of the solution in the region in which type I precedes type II. The three-dimensional plot shows that the solution is stable in time as well as in space. The values of c_1 and c_2 for this case are 0.78361 and 0.02367 respectively. The stability of the solution over long time periods was investigated by studying the evolution of the real part of the solution (Re(W)) for short as well as long time periods (Fig. 5.3 (a) and 5.3 (b)). In order to project the features appearing in three dimensions, on a two-dimensional plane, constant amplitude lines are projected in the time-space plane. Such a plot referred to as the contour plot, is shown in Fig. 5.3 (c) for the case 5.3 (b). In this region the most predominant feature of the system is spatially homogeneous oscillations, which can however become unstable due to the diffusional effects. This type of diffusional instability is known as phase instability (Kuramoto, 1984) and is different from the type II instability studied in this chapter. The condition for the phase instabilty is studied in more detail in Chapter VII.

Figure 5.4 depicts the short time behavior of the amplitude in the multiplicity regime referred to earlier. By multiplicity of the instability condition we mean that for a particular set of the other parameter values, there are multiple values of j_1 for which an instability can occur. In the region chosen here, the type II instability has already set in so that we see a distinct spatial inhomogenity. Figure 5.4 (a) and (b) show the evolution of the Re(W) and Im(W) for the case $j_1 = 39.8$. The other parameter values are $j_2 = 0.99; \beta = 44.0; \gamma = 0.984; k = 0.1; D_1 = 0.005;$ and $D_2 = 1.5;$. The values of c_1 and c_2 computed from this set of parameter values are 7.7×10^{5} and 1.07×10^{-2} respectively. Figures 5.4 (c) and (d) depict the case $j_1 = 57.2$, for the same values of the other parameters. The values of c_1 and c_2 change to 6328.0 and 14.08 respectively. In Fig. 5.4 (d) and (e) the values of j_1, c_1 and c_2 are 60.1, 4652.0 and -4.1828 respectively. Over

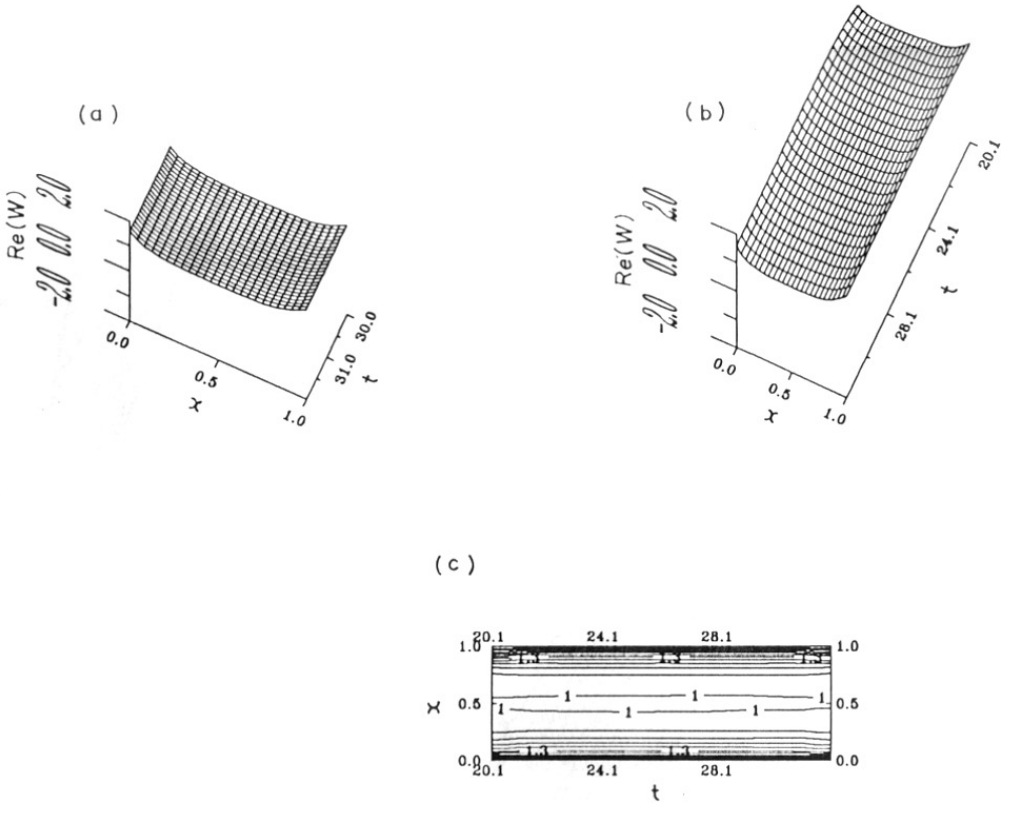

Fig. 5.3 : Space-time variation of the amplitude; a, b are 3-dimensional plots of the variation in the real part of the amplitude on short and long time scales respectively; c shows the contour plot (see text) for the real part.

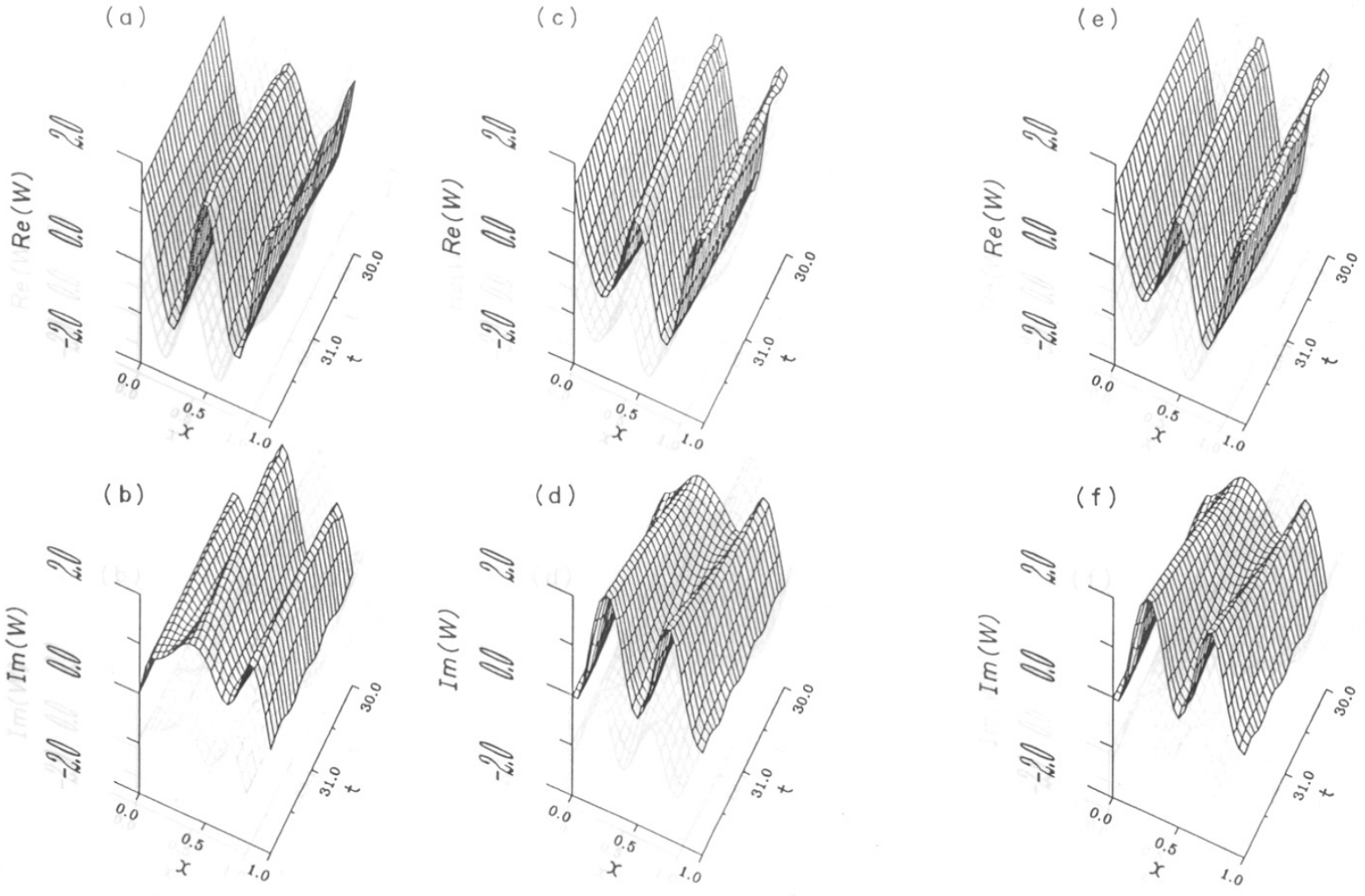

Fig. 5.4 : Space-time variation of the amplitude (on a short time scale) in a multiplicity regime of j_1 ; a, b show the real and imaginary parts for $j_1 = 39.8$; for c, d $j_1 = 57.2$ and e, f $j_1 = 60.1$.

Fig. 5.5 : Space-time variation of the amplitude on a long time scale for the cases discussed in Fig. 5.4.

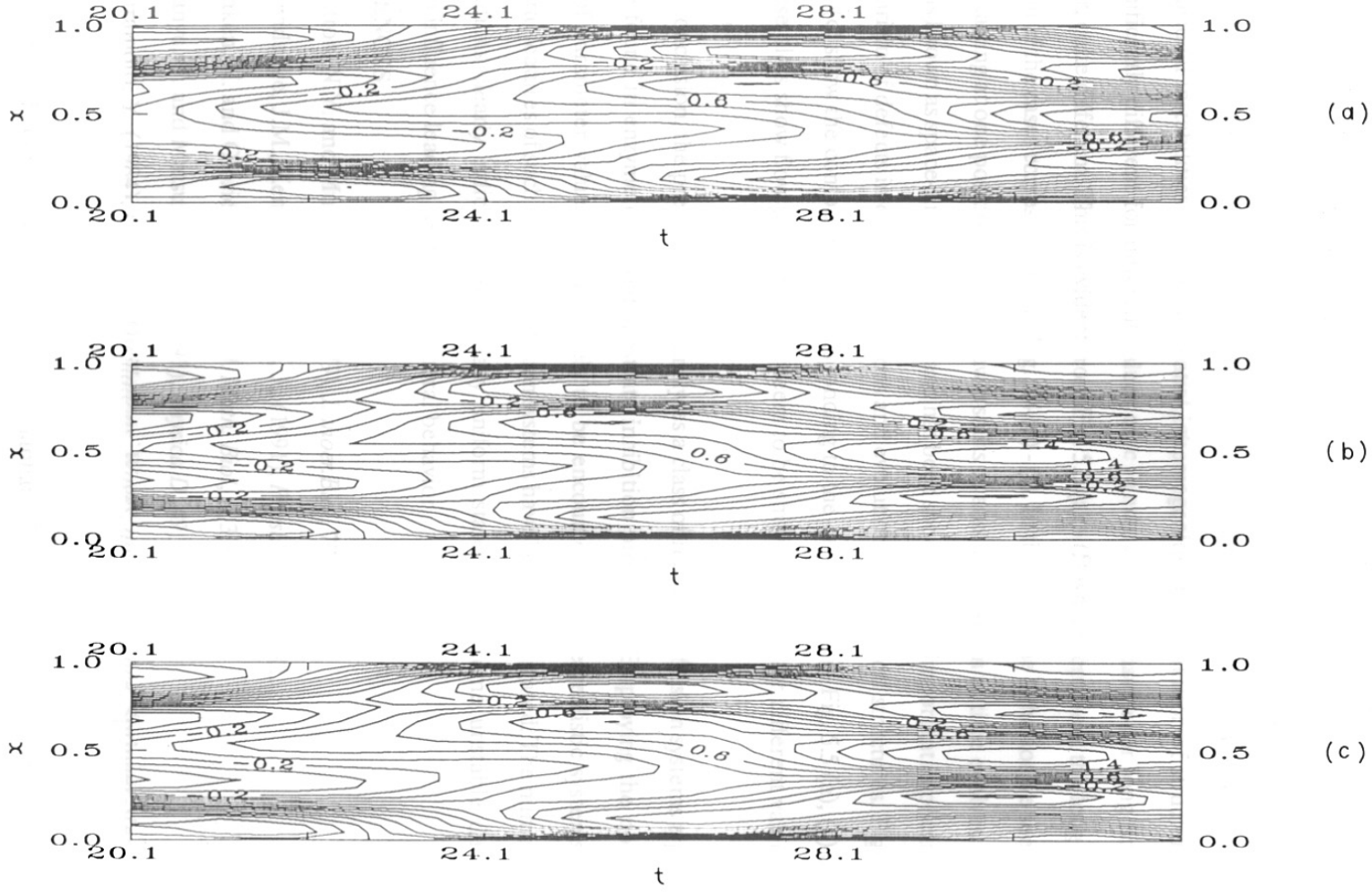

Fig. 5.6 : Contour plots for the three cases depicted in Fig. 5.5.

short time periods we see a structure similar to that of a roll-type pattern (Haken and Olbrich, 1978) which is not significantly different for different values of c_1 and c_2. From the relatively high value of c_1 we can infer that we are operating in reaction controlled regime and although the diffusional instabilities give rise to spatial inhomogenities the evolution of structures is not significantly different for different values of c_1 and c_2. The behavior for long time is however, quite different. This is evident from Fig. 5.5 (a) - (f) which represent the long time evolution of the cases corresponding to Fig. 5.4 (a) - (f). In the long time behavior we see that a wave starting at one position in space slowly shifts to another position as time evolves. This feature is known as space-time dislocation and has been shown to have important implications for the origin of defects in crystal formations and evolution of geological structures. Figures 5.6 (a) - (c) show the contour plots corresponding to the cases shown in Fig. 5.5 (a), (c) and (e). These plots show flow patterns which seem to be shifting in time for different values of c_1 and c_2.

In conclusion, the present chapter analyses a class of reaction-diffusion systems typical of those found in enzyme catalyzed substrate inhibition processes. Employing the normal ranges of parameter values that are likely to be encountered in the realistic systems the behavioral features of such reaction-diffusion systems have been investigated. The numerical computations clearly indicate that spatially uniform states can become unstable due to diffusion influence leading to spatial-temporal behavior.

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CHAPTER VI

DERIVATION OF GINZBURG LANDAU EQUATION IN THE PRESENCE OF EXTERNAL NOISE

Fluctuations are inherent to natural systems and it is seen that they can amplify at critical points, leading the system to a state of higher order. In the present chapter we obtain an equation of the form of the GL equation to describe the evolution of the system in presence of external fluctuations. This equation can be easily extended to a general class of fluctuations and is crucial to the analysis of a variety of interesting situations. The point of importance of the present chapter is that, here we extend the principles of derivation of the GL equation to stochastic systems in a simple way.

6.1 Introduction

Fluctuations are inherent in all natural systems and they are known to bring about evolution of order in many systems. Typical examples include lasers, hydrodynamics and mutations in living organisms. It is seen that fluctuations can amplify at critical points, allowing a new structure to supercede the old. The concept of order through fluctuations was well demonstrated by Nicolis and Prigogine (1977). Many attempts have been made in the past to analyze the transition from one state to another within the framework of the stochastic theory. Two types of stochastic approaches are generally followed in the analysis of fluctuations, leading to two types of equations *viz.*, Fokker-Planck equation and the other to Langevin equation. A given Langevin equation can always be converted into an equivalent Fokker-Planck equation.

Many authors have studied the evolution of nonequilibrium systems, near the critical points in presence of fluctuations (Graham and Haken, 1970, Haken, 1983,1975, Nitzan *et al*, 1974a,b, Gardiner *et al*, 1976 and Wunderlin and Haken,1975). In such systems, the various features can be attributed to the existence of slow modes near the transition points. Thus we come back to the slaving principle and principles of synergetics (Haken, 1989). Most of the studies outlined here make use of the master equation and the non-linear Fokker-Planck equation.

In the present chapter we analyze the substrate-inhibition kinetic model, near its critical points, in the presence of fluctuations, with a view to obtain an evolution equation in the form of a GL equation. The fluctuations are taken into account, using a Langevin-equation approach. The point of importance of the present chapter is that, here we extend the principles of derivation of the GL equation to stochastic systems in a simple way. The final equation obtained is qualitatively similar to the case without fluctuations, but is quantitatively different.

6.2 Substrate-Inhibition Kinetic Model with External Fluctuations

In the present study we incorporate external fluctuations into the governing equations by assuming that the parameter j_1 fluctuates according to $\overline{j_1} + \zeta(t)$. Here, $\overline{j_1}$ is the mean value of j_1, and $\zeta(t)$ represents the external noise, described by an Ornstein-Uhlenbeck process with zero mean and finite correlation given by

$$<\zeta(t)\zeta(t')> = \left(\frac{D}{\tau}\right)\exp-\{t-t'/\tau\} \qquad t>t' \tag{6.2.1}$$

The Langevin equations corresponding to the model equations Eq. (5.2.3) and (5.2.4) become

$$\frac{dx}{dt} = (\overline{j_1} + \zeta) - x - \beta r(x,y) = f_1 \tag{6.2.2a}$$

$$\frac{dy}{dt} = j_2 - \gamma . r(x,y) = f_2 \tag{6.2.2b}$$

The steady state solutions can then be written as

$$x_s = \frac{\gamma(\overline{j_1} + \zeta) - \beta j_2}{\gamma} \qquad y_s = \frac{j_2\{k\gamma^2(\overline{j_1} + \zeta)^2 + (\overline{j_1} + \zeta)\gamma a_3 + a_4\}}{\gamma^2(\gamma(\overline{j_1} + \zeta) + a_2)} \tag{6.2.3}$$

where, the definitions of the terms $a_1 - a_4$ remain the same as in Chapter V. The deviation equations change accordingly to

$$\frac{du}{dt} = -u + \left\{\frac{((\overline{j_1} + \zeta) - x_s)(u + v + ku^2 + 2kux_s) - \beta(x_s v + y_s u + uv)}{(1 + x_s + y_s + kx_s^2) + (u + v + ku^2 + 2kux_s)}\right\} \tag{6.2.4a}$$

$$\frac{dv}{dt} = \left\{\frac{j_2(u + v + ku^2 + 2kux_s) - \gamma(x_s v + y_s u + uv)}{(1 + x_s + y_s + kx_s^2) + (u + v + ku^2 + 2kux_s)}\right\} \tag{6.2.4b}$$

The corresponding reaction-diffusion equations are

$$\frac{\partial x}{\partial t} = D_1 \frac{\partial^2 x}{\partial r^2} + (\overline{j_1} + \zeta) - x - (\beta xy / 1 + x + y + kx^2) \tag{6.2.5a}$$

$$\frac{\partial y}{\partial t} = D_2 \frac{\partial^2 y}{\partial r^2} + j_2 - (\gamma x y / 1 + x + y + k x^2) \qquad (6.2.5b)$$

In the analysis presented in this chapter we conform to the assumption that the order of magnitudes of the macroscopic variables of the systems and the noise variable are widely different. In other words ζ is of the order $O(\epsilon^2)$, where ϵ is an arbitrary small parameter. The Jacobian of the system can then be expressed as

$$J = F_1 \begin{bmatrix} \frac{\beta}{\gamma} j_2 F_3 - \beta F_2 - \frac{1}{F_1} & -\frac{\beta}{\gamma} F_4 \\ j_2 F_3 - \gamma F_2 & -F_4 \end{bmatrix} \qquad (6.2.6)$$

where the definitions of the terms $F_1 - F_5$ are

$$F_1 = \gamma^2 (\gamma(\overline{j_1} + \zeta) + a_2) / (\gamma(\overline{j_1} + \zeta) + a_1)\left(k\gamma^2 \overline{j_1}^2 + (2k\gamma^2\zeta + \gamma a_3)\overline{j_1} + (\gamma a_3 \zeta + a_4)\right)$$

$$F_2 = y_s$$

$$F_3 = (\gamma + 2k(\gamma(\overline{j_1} + \zeta) + a_1))/\gamma$$

$$F_4 = (\gamma(\overline{j_1} + \zeta) + a_2)$$

$$F_5 = \left(-a_5 \overline{j_1}^3 + (-3a_5\zeta + a_6)\overline{j_1}^2 + (2a_6\zeta + a_7)\overline{j_1} + (a_7\zeta + a_8)\right)/\gamma^2 F_4 \qquad (6.2.7)$$

and the terms $a_5 - a_8$ are defined according to Chapter V.

Following the usual stability analysis procedure, we obtain the conditions for the occurrence of type I and type II instabilities as

$$a_5 j_{1c}^3 + (3a_5\zeta - a_9) j_{1c}^2 - (2a_9\zeta + a_{10}) j_{1c} - (a_{10}\zeta + a_{11}) = 0 \qquad (6.2.8)$$

$$j_{1c}'^{6}(D_2^2 b_1)+j_{1c}'^{5}(D_2^2 b_2-2D_1D_2 b_3+6\zeta)+j_{1c}'^{4}(D_2^2 b_4+D_1^2 b_5-2D_1D_2 b_6+5\zeta)$$
$$+j_{1c}'^{3}(D_2^2 b_7+D_1^2 b_8-2D_1D_2 b_9+4\zeta)+j_{1c}'^{2}(D_2^2 b_{10}+D_1^2 b_{11}-2D_1D_2 b_{12}+3\zeta)$$
$$+j_{1c}'(D_2^2 b_{13}+D_1^2 b_{14}-2D_1D_2 b_{15}+2\zeta)+(D_2^2 b_{16}+D_1^2 b_{17}-2D_1D_2 b_{18}+\zeta) = 0 \qquad (6.2.9)$$

Once again the definitions of the terms $a_9 - a_{14}$ and $b_1 - b_{18}$ are carried over from Chapter V. The characteristic frequency of the system ω_0 assumes a new value in the presence of fluctuations. This is given as

$$\omega_0 = \{(a_5\overline{j_1}^3+(3a_5\zeta+a_{15})\overline{j_1}^2+(2a_{15}\zeta+a_{16})\overline{j_1}+(a_{16}\zeta+a_{17})^{1/2}\}/2 \qquad (6.2.10)$$

6.3 Derivation of G-L Equation

For the analysis of the system near the critical points we define the parameter μ as, $\mu = \overline{j_1} - j_{1c}/j_{1c}$, and ϵ as $\epsilon^2\chi = \mu$. This definition gives

$$j_1 = (\overline{j_1}+\zeta) = j_{1c}(\mu+1)+\zeta \qquad (6.3.1)$$

As has been already mentioned, ζ is of order of $O(\epsilon^2)$. Here we define another smallness parameter ϵ', such that $\zeta = \zeta_0\epsilon'^2\chi$. We further define ψ by the following relationship

$$\epsilon'^2\chi/\epsilon^2\chi = \psi \qquad (6.3.2)$$

In addition we assume that the correlation time of the noise is much longer than the characteristic time of system and that the fluctuations evolve on the time scale τ. Although this assumption for the fluctuations is used somewhat less frequently, it is justified in the present case. The physical basis for this is as follows : though the supply of the species X and Y to the cell are supposed to be externally controlled parameters, this is not strictly so. No cell can operate in isolation but forms a part of a large network of cells coupled through

various forms of feedback. Thus the substrates of a reaction in one cell are actually products of another and any fluctuation in the production of the species X at one point in the network is reflected in the parameter j_1 only after a definite time delay.

Using these definitions we obtain L_0, L_1, M_0, and other quantities required for the derivation of the G-L equation. We find that L_0 still subscribes to the relation given in Eq. (5.3.1), whereas L_1 changes to

$$L_1 = F_7 j_{1c}(1+\zeta_0\psi)\left\{ \begin{bmatrix} l_{11} & -\beta l_{12} \\ l_{13} & -\gamma l_{12} \end{bmatrix} - F_8 L_0 \right\} \qquad (6.3.3)$$

with the definitions of all the terms remaining the same as in Chapter V. Similarly, we find that the equations describing the quantities M_0 and N_0 also remain the same as in the case without fluctuations. Further, this implies that the GL constants c_0, c_1 and c_2 can still be given by Eq. (5.3.11). However, note that the linear operator L_0 and hence the corresponding eigenvectors show an explicit dependence on ω_0, which changes considerably in the presence of fluctuations (see Eq. (6.2.10)).

The resulting GL equation in presence of fluctuations can now be solved coupled with the evolution equation for the fluctuations, Eq. (6.2.1). Note that according to our assumption, both W and ζ evolve on the same time scale, so that we do not need any further assumptions in solving this coupled set of equations.

6.4 Results and Discussion

In the present chapter, we have developed an evolution equation for the substrate-inhibition kinetic model in presence of fluctuations, near a critical point, based on some simple assumptions. Most of the previous studies in this direction, make use of a Fokker-Planck equation approach to include noise. Here, we approach the problem in a simple way leading to an equation which is essentially of the same form as the GL equation and can be solved easily using numerical methods such as the one employed in Chapter V. The method

presented in this chapter can also be extended to include different types of noise, investigate the effects of noise on coupled oscillators, changes in the entrainment conditions and so on. In particular it would be interesting to study the conditions for the stability of limit cycle solution in presence of external noise and see how the diffusional effects and external noise cooperate or counter each other and obtain the conditions under which one dominates the other. The method presented here is crucial to all such analysis.

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CHAPTER VII

ANALYSIS OF DIFFUSIONALLY COUPLED OSCILLATORS

Weakly coupled oscillators can be described in terms of a non-linear phase diffusion equation. In the present chapter we aim at deducing this equation and extending it to the case of Ginzburg-Landau oscillators. The condition for the stability of the orbital shape is also derived. The stability of the limit cycle solution to the GL equation is tested for the case of substrate-inhibition kinetics, with the help of this condition. Numerical analysis of the phase diffusion equation is also presented for the stable oscillatory regime of this model.

7.1 Introduction

Coupling of nonlinear functional units, such as individual cells following nonlinear kinetics, can lead to a multitude of dynamic features. Indeed, the study of diffusionally coupled cells, has served to understand many self-organization phenomena in physical, chemical and biological systems, in the past (Pavlidis, 1973, Schreiber and Marek 1982, Kubicek and Marek, 1983, Kaneko, 1989, Hadley and Weisenfold, 1989). Study of coupled nonlinear oscillators in particular, has aroused much interest, because such systems can be envisaged in many biologically significant phenomena, such as synchronization and onset of collective oscillations (Aronson *et al*, 1989, Ermentrout, 1989 and Silva *et al*, 1991). Analysis of coupled oscillators can be a formidable task, since the number of degrees of freedom, involved is bound to multiply. However, for weakly coupled oscillators, the dynamics can be greatly simplified, and by looking upon diffusion as an external perturbation, they become amenable to simple perturbation techniques. By weak coupling, we mean that diffusion terms are small compared to the reaction terms.

For systems, showing strictly periodic behavior, as $t \to \infty$, one may assign an arbitrary scalar value to each point associated with the oscillator. This scalar quantity, known as *phase* can be used to approximate the state of the oscillator. Thus the dynamics of a system of N discrete oscillators (of atleast $2N$ degrees of freedom), can be reduced to N coupled ODEs for N phase variables. Moreover, it is seen that weak perturbations generally produce a long time scale in the dynamics compared to the period of the original oscillator. Such a clear separation in time scales enables us to average the rapidly varying oscillating quantities of the original equations, for the slowly varying phases. Though phase is an arbitrarily defined quantity, it can yield important information for the coupled systems. For instance we find that the phase difference between two coupled oscillators, as a function of t, tells us whether they are mutually entrained or not; further if the deviation of the oscillator from its natural closed orbit is very small, then the evolution of various dynamic features can be approximated by the evolution of phases with t.

A phase description for populations of oscillators was first given by Winfree (1967). This theory was further developed, and suitably modified for specific cases, by various authors (Ortoleva and Ross, 1973, 1974, Kuramoto and Tsuzuki, 1976, Kuramoto and Yamada, 1976, Neu, 1979a, b, Neu, 1980, Kuramoto, 1984, Ermentrout and Kopell, 1984, Ermentrout, 1985, Sakaguchi *et al*, 1987, Daido, 1988, Strogatz and Mirollo, 1988, Sakaguchi *et al*, 1988, Ermentrout, 1989 and Kuramoto, 1991).

In chapter IV we saw that near the Hopf bifurcation point, the GL equation describes the behavior of a field of oscillators. In the present chapter, we show that when such oscillators are weakly coupled through diffusion, the system behavior can be described by the phases of the oscillators, through a non-linear phase diffusion equation.

7.2 Phase description: A case of single oscillator

In this section, we aim to illustrate how the phase can describe the perturbed motion of a single oscillator. Once the perturbation method is formulated, it can be extended to the case where, the perturbation is interpreted in terms of diffusion. This will be done in the following section with a view to derive the phase diffusion equation.

Let $X_0(t)$ denote a stable periodic solution to a system of ODEs, Eq. (4.2.1)

$$\frac{dX_0}{dt} = F(X_0) \qquad X_0(t+\tau) = X_0(t) \tag{7.2.1}$$

where, T is the time period.

If C represents the closed orbit corresponding to the periodic solution $X_0(t)$, then we can associate a certain value of phase ϕ to each $X \in C$, such that the motion along C produces a constant increase in ϕ, or for convenience,

$$\frac{d\phi(X)}{dt} = 1 \ , \qquad X \in C \tag{7.2.2}$$

All the perturbation analysis of the present chapter is carried out around the periodic solution, $X_0(t)$. Let us introduce a small perturbation as,

$$\frac{dX}{dt} = F(X) + \epsilon p(X) \tag{7.2.3}$$

The term $\epsilon p(X)$ generally depends on X, and denotes a general class of perturbations.

From Eq. (7.2.2), we see that the quantity ϕ is defined only on C. However, for perturbed systems, we would like to extend this definition to the vicinity of C. Since the limit cycle solution is a stable one, and since the perturbation is weak, the system still shows a periodic behavior, but with a different T. Thus we can imagine a circular tube enveloping C, as denoting the space in which the perturbed solution can be found. The tube is thin to the extent that the perturbation is weak. In this circular tube, we find two dimensional surfaces, known as isochrons, such that all the points lying on a given isochron have the same value of ϕ. Thus each point in C, belongs to a single two dimensional surface, say G. This is depicted in Fig. (7.1). It follows that, if a given point belongs to G, then its phase will remain the same whether it belongs to C or not. Thus, we can extend the definition of ϕ, beyond C as,

$$\frac{d\phi(X)}{dt} = 1 \quad , \qquad X \in G \tag{7.2.4}$$

Using the chain rule, we have,

$$\frac{d\phi(X)}{dt} = \text{grad}_X \phi \cdot \frac{dX}{dt} \tag{7.2.5}$$

so that using (7.2.4) and (4.2.1), we obtain,

$$\text{grad}_X \phi \, F(X) = 1 \qquad X \in G \tag{7.2.6}$$

It is also clear that for an unperturbed system, any point near C and belonging to an isochron $I(\phi)$, will always be found on the same isochron, but closer and closer to C as time increases. For the perturbed system,

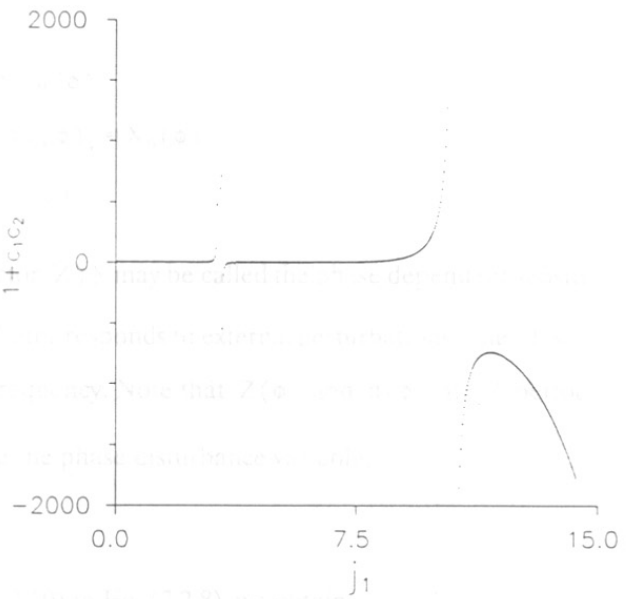

Fig. 7.1 : Mapping of the stability parameter α *vs* j_1 .

$$\frac{d\phi(X)}{dt} = \mathrm{grad}_x \phi[F(X)+\epsilon p(X)]$$

$$= 1 + \epsilon \mathrm{grad}_x \phi \cdot p(X) \qquad (7.2.7)$$

Although the specification of ϕ is insufficient to describe the position of the state point X on $I(\phi)$, from the above discussion we atleast know that it is close to $X_0(\phi)$. Thus we can replace X in Eq. (7.2.7) with $X_0(\phi)$.

$$\frac{d\phi}{dt} = 1 + \epsilon \Omega(\phi) \qquad (7.2.8)$$

where,

$$\Omega(\phi) = Z(\phi) \cdot \pi(\phi)$$
$$Z(\phi) = (\mathrm{grad}_x \phi)_x = X_0(\phi) \qquad (7.2.9)$$
$$\pi(\phi) = p(X_0(\phi))$$

Here, the vector $Z)\phi$ may be called the phase dependent sensitivity, as it measures how sensitively the oscillator responds to external perturbations. The r.h.s. of Eq. (7.2.8), represents an instantaneous frequency. Note that $Z(\phi)$ and $\pi(\phi)$ ate T-periodic functions of ϕ.

Here, we introduce the phase disturbance variable,

$$\phi = t + \psi \qquad (7.2.10)$$

Substituting Eq. (7.2.10) in Eq. (7.2.8), we obtain

$$\frac{d\psi}{dt} = \epsilon \Omega(t + \psi) \qquad (7.2.11)$$

This equation shows that ψ is a slow variable, and hardly changes during the period T. Then we can use time averaging to obtain a frequency change.

$$\frac{d\psi}{dt} = \epsilon\omega = \epsilon \frac{1}{\tau} \int_0^{\tau} \Omega(t)dt \qquad (7.2.12)$$

Using the procedure outlined in this section, we go on to derive the nonlinear phase diffusion equation.

7.3 Nonlinear phase diffusion equation

Reaction-diffusion equations may be written in the form of Eq. (7.2.2) if p is interpreted as a Laplacian operator multiplied by the matrix D.

$$\epsilon p = D\nabla^2 \tag{7.3.1}$$

Applying Eq. (7.3.1) to Eq. (7.2.8) and setting $\epsilon = 1$, we get

$$\frac{\partial \phi}{\partial t} = 1 + \epsilon\Omega(\phi)$$

$$1 + \left\{(\text{grad}_X \phi)_{x=X_0(\phi)} \quad D(\nabla^2)_{x=X_0(\phi)}\right\} \tag{7.3.2}$$

We now write,

$$D\frac{\partial^2 X_0(\phi)]}{\partial r^2} = D\frac{\partial}{\partial r}\left(\frac{\partial \phi}{\partial r}\frac{dX_0(\phi)}{d\phi}\right)$$

$$= D\left\{\nabla^2\phi \cdot \frac{dX_0(\phi)}{d\phi} + (\nabla\phi)^2 \cdot \frac{d^2X_0}{d\phi^2}\right\} \tag{7.3.3}$$

Substitution of Eq. (7.3.3) in Eq. (7.3.2), alongwith Eq. (7.2.9) gives,

$$\frac{\partial \phi}{\partial t} = 1 + \Omega^{(1)}\phi\nabla^2\phi + \Omega^{(2)}(\phi)(\nabla\phi)^2 \tag{7.3.4}$$

where,

$$\Omega^{(1)}(\phi) = Z(\phi) \cdot D \cdot \frac{dX_0(\phi)}{d\phi}$$

$$\Omega^{(2)}(\phi) = Z(\phi) \cdot D \cdot \frac{d^2X_0(\phi)}{d\phi^2} \tag{7.3.5}$$

In order to obtain the above terms, we need to do the stability analysis around the periodic solution. For this purpose, we have to consider the Floquet theory for the stability of periodic system, in detail. Let us denote a deviation from the periodic solution $X_0(t)$ as,

$$X(t) = X_0(t) + \mathbf{u}(t) \tag{7.3.6}$$

Linearization about $X_0(t)$ leads to,

$$\frac{d\mathbf{u}}{dt} = \mathsf{L}(t)\,\mathbf{u} \tag{7.3.7}$$

where, L is a $2x2$ T-periodic matrix with its ij th element given by $L_{ij} = \partial F_i(X_0(t))/\partial X_{0j}(t)$. The general solution to this equation can be expressed as,

$$\mathbf{u}(t) = \mathsf{S}(t)\,e^{\Lambda t}\,\mathbf{u}(0) \tag{7.3.8}$$

Here, $S(t)$ is again a $2x2$ T-periodic matrix with the initial condition $S(0) = 1$, and Λ is some time independent matrix. According to the Floquet theory, it is then possible, to define an eigenvalue problem for Eq. (7.3.7), just as in the steady state case, excepting for the fact that all the variables and operators are also T-periodic. In this case, the eigenvalues of the matrix Λ decide the stability of the solution. Let $\mathbf{u}_l$, and $\mathbf{u}_l^*$ denote the right and left eigenvectors, corresponding to the eigenvalues λ_l of Λ. Then,

$$\Lambda\,\mathbf{u}_l = \lambda_l\,\mathbf{u}_l \qquad \mathbf{u}_l^*\,\Lambda = \lambda_l\,\mathbf{u}_l^* \,, \qquad l = 0, 2, \ldots, n \tag{7.3.9a}$$

$$\mathbf{u}_l^*\,\mathbf{u}_m = \delta_{lm} \,, \qquad l = m = 0, 2, \ldots, n \text{ and },$$
$$= 0 \,, \qquad l \neq m \tag{7.3.9b}$$

Since $X_0(t)$ is assumed to be stable, no eigenvalues have a positive real part, and there is one special eigenvalue which is identically zero. Let λ_0 denote the zero eigenvalue, and $\mathbf{u}_0$ its corresponding eigenvector. Then we have following relation,

$$\Lambda\,\mathbf{u}_o = 0 \tag{7.3.10}$$

ow by differentiating Eq. (7.2.1), we have,

$$\frac{d}{dt}\left(\frac{dX_0(t)}{dt}\right) = \frac{d}{dt}(F(X_0))$$

$$= L(t)\left(\frac{dX_0(t)}{dt}\right) \qquad (7.3.11)$$

Thus from Eq. (7.3.11) we can see that $(dX_0(t)/dt)$ is a particular solution of Eq.(7.3.7). Then using Eq. (7.3.8) we can write,

$$\left(\frac{dX_0(t)}{dt}\right)_t = S(t)\, e^{\Lambda t}\left(\frac{dX_0(t)}{dt}\right)_{t=0} \qquad (7.3.12)$$

Further, the zero eigenvector of Eq. (7.3.10) can be taken as,

$$u_0 = \left(\frac{dX_0(t)}{dt}\right)_{t=0} \qquad (7.3.13)$$

Substituting in Eq. (7.3.12), we get,

$$\left(\frac{dX_0(t)}{dt}\right)_t = u_t = S(t)\, e^{\Lambda t}\, u_0$$

Linearizing $e^{\Lambda t}$ as $(I + \Lambda t)$ and making use of Eq. (7.3.10), we obtain,

$$S(t)\, u_0 = u_t \qquad (7.3.14)$$

In order to evaluate $Z(\phi)$ we imagine a two-dimensional space $T(\phi)$ tangent to the isochron $I(\phi)$ at $X(\phi)$ such that any small vector $u(t)$ lying on $T(\phi)$ and denoting $X(t) - X_0(t)$ reduces to zero as $t \to \infty$. From Eq. (7.3.8) it is evident that $u(t)$ can reduce to zero only if $u(0)$ is free of the zero-eigenvector component. Thus it means that T is nothing but the eigenspace spanned by all the eigenvectors but the zero eigenvector. Further $Z(\phi)$ is normal to $I(\phi)$ and hence $T(\phi)$. This fact combined with Eq. (7.3.9b), leads to the relation

$$Z(0)\, u_t = 0\,, \qquad l \neq 0 \qquad (7.3.15)$$

Comparing with Eq. (7.3.9b), we see that $Z(0)$ is proportional to $\mathbf{u}_0^*$ and for the sake of convenience, may be taken to be 1. Thus,

$$\mathbf{u}_0^* = Z(0) \tag{7.3.16}$$

From Eq. (7.3.9b) and Eq. (7.3.16), we have,

$$Z_0\left(\frac{dX_0(t)}{dt}\right)_{t=0} = 1$$

or more generally,

$$Z(t)\left(\frac{dX_0(t)}{dt}\right) = 1 \tag{7.3.17}$$

From Eq. (7.3.14) and Eq. (7.3.17) we have,

$$Z(t) = \left(\frac{dX_0(t)}{dt}\right)^{-1} = [S(t)\mathbf{u}_0]^{-1}$$

Again, using Eq. (7.3.9b), we can write,

$$Z(t) = \mathbf{u}_0^* S^{-1}(t) \tag{7.3.18}$$

Obtaining $(dX_0(t)/dt)$ from Eq. (7.3.14) and $Z(t)$ from Eq. (7.3.18), we can express the terms used in Eq. (7.2.9) and Eq. (7.3.5) as,

$$\Omega(\phi) = \mathbf{u}_0^* S^{-1}(\phi)\pi(\phi) \tag{7.3.19a}$$

$$\Omega^{(1)}(\phi) = \mathbf{u}_0^* S^{-1}(\phi)DS(\phi)\mathbf{u}_0 \tag{7.3.19b}$$

$$\Omega^{(2)}(\phi) = \mathbf{u}_0^* S^{-1}(\phi)D\frac{dS(\phi)}{d\phi}\mathbf{u}_0 \tag{7.3.19c}$$

In terms of the slow variable ψ, Eq. (7.3.4) can be transformed as,

$$\frac{\partial \psi}{\partial t} = \Omega^{(1)}\nabla^2\psi + \Omega^{(2)}(t+\psi)(\nabla\psi)^2 \tag{7.3.20}$$

Doing the time averaging as in the last section, we obtain,

$$\frac{\partial \psi}{\partial t} = \alpha \nabla^2 \psi + \beta (\nabla \psi)^2 \quad , \text{ or}$$

$$\frac{\partial \phi}{\partial t} = 1 + \alpha \nabla^2 \phi + \beta (\nabla \phi)^2 \tag{7.3.21}$$

where,

$$\alpha = \frac{1}{T}\int_0^T \Omega^{(1)}(t)dt \ , \qquad \beta = \frac{1}{T}\int_0^T \Omega^{(2)}(t)dt \tag{7.3.22}$$

In the next section we evaluate the quantities $S(\phi)$, α and β for the case of the GL model.

7.4 The Ginzburg-Landau oscillators.

The GL equation can be expressed as a two component reaction-diffusion system. The diffusion matrix D is different from the diffusion matrices of ordinary reaction-diffusion systems, in that it involves an antisymetric part.

$$D = \begin{pmatrix} 1 & -c_1 \\ c_1 & 1 \end{pmatrix} \tag{7.4.1}$$

We start with the Stuart-Landau equation which effectively describes a diffusionless oscillatory system. Then we go on to introduce the diffusion terms as a weak external perturbation through the matrix D.

$$\frac{dW}{dt} = (1 + ic_0)W - (1 + ic_2)|W|^2 W \tag{7.4.2}$$

Separating the real and imaginary parts, we obtain,

$$\frac{dX}{dt} = X - c_0 Y - (X - c_2 Y)(X^2 + Y^2)$$

$$\frac{dY}{dt} = Y + c_0 X - (Y + c_2 X)(X^2 + Y^2) \tag{7.4.3}$$

The periodic solution of Eq. (7.4.2), $W_0(t)$, is expressed in terms of the corresponding real and imaginary parts as,

$$W_0(t) = X_0(t) + iY_0(t) = A_0 \exp(i\omega_0 t) \tag{7.4.4}$$

Substituting for $W_0(t)$ as $A_0 \exp(i\omega_0 t)$, in Eq. (7.4.2), and putting $A_0 = 1$, we obtain

$$\omega_0 = c_0 - c_2 \tag{7.4.5}$$

Let $w(t)$ denote a disturbance variable defined as follows

$$W(t) = W_0(t)(1 + w(t)) \tag{7.4.6}$$

Substituting Eq. (7.4.6) in Eq. (7.4.2) and linearizing in $w(t)$, we obtain

$$\frac{dw}{dt} = -(1 + ic_2)(w + \bar{w}) \tag{7.4.7}$$

Separating $w(t)$ into real and imaginary parts as, $w = \zeta + i\eta$, Eq. (7.4.7) may be expressed as

$$\frac{d}{dt}\begin{pmatrix}\zeta \\ \eta\end{pmatrix} = \Lambda \begin{pmatrix}\zeta \\ \eta\end{pmatrix} \tag{7.4.8}$$

where Λ is yet to be defined. From Eq. (7.4.7) we have

$$\frac{d\zeta}{dt} + i\frac{d\eta}{dt} = -2\zeta - i(2c_2\zeta) \tag{7.4.9}$$

and from Eq. (7.4.9), we have

$$\frac{d\zeta}{dt} = \Lambda_1 \zeta + \Lambda_2 \eta$$

$$\frac{d\eta}{dt} = \Lambda_3 \zeta + \Lambda_4 \eta \tag{7.4.10}$$

Solving Eq. (7.4.10) and Eq. (7.4.11), together we obtain Λ as,

$$\Lambda = -2\begin{pmatrix} 1 & 0 \\ c_2 & 0 \end{pmatrix} \tag{7.4.12}$$

Equation (7.4.9) can now be integrated as,

$$\begin{pmatrix} \zeta(t) \\ \eta(t) \end{pmatrix} = e^{\Lambda t}\begin{pmatrix} \zeta(0) \\ \eta(0) \end{pmatrix} \tag{7.4.13}$$

In order to obtain $\mathbf{u}_0$, $\mathbf{u}_0^*$ and $S(\phi)$ we have to perform the stability analysis of Eq. (7.4.3) around the periodic solution $(X_0(t), Y_0(t))$. To do this we linearize Eq. (7.4.3) in the deviational variables $(x, y) = (X - X_0(t), Y - Y_0(t))$, and find their solutions of the deviational variables in the form Eq. (7.3.8).

Making use of the relation $W = X + iY$, Eq. (7.4.6), (7.4.8) and (7.4.14), we obtain the relation between, (x, y) and (ζ, η) as

$$\begin{pmatrix} x(t) \\ y(t) \end{pmatrix} = S(t)\begin{pmatrix} \zeta(t) \\ \eta(t) \end{pmatrix}$$

where

$$S(t) = \begin{pmatrix} \cos(\omega_0 t) & -\sin(\omega_0 t) \\ \sin(\omega_0 t) & \cos(\omega_0 t) \end{pmatrix} \tag{7.4.14}$$

Making use of the relation, $\begin{pmatrix} x(0) \\ y(0) \end{pmatrix} = \begin{pmatrix} \zeta(0) \\ \eta(0) \end{pmatrix}$, Eq. (7.4.13)can be shown to be equivalent to

$$\begin{pmatrix} x(t) \\ y(t) \end{pmatrix} = S(t)e^{\Lambda}\begin{pmatrix} x(0) \\ y(0) \end{pmatrix} \tag{7.4.15}$$

This is the same form as Eq. (7.3.8). Hence the eigenvalues of Λ decide the stability of the periodic solution. The eigenvalues are obtained as $\lambda_0 = 0$ and $\lambda_1 = -2$. The corresponding eigenvectors are,

$$\mathbf{u}_0 = \omega_0 \begin{pmatrix} 0 \\ 1 \end{pmatrix} \quad ; \quad \mathbf{u}_0^* = \omega_0^{-1}(-c_2, 1) \tag{7.4.16}$$

where w_0 is needed for consistency with Eq. (7.3.13). From Eq. (7.3.9a,b) we get,

Further, for $\lambda = -2$,

$$\mathbf{u}_1 = \begin{pmatrix} 1 \\ c_2 \end{pmatrix} \quad ; \quad \mathbf{u}_1^* = (1, 0) \tag{7.4.17}$$

We can now calculate the terms defined in Eq. (7.3.3) as follows

$$\Omega^{(1)} = \omega_0^{-1}(-c_2, 1) \begin{pmatrix} \cos(\omega_0 t) & \sin(\omega_0 t) \\ -\sin(\omega_0 t) & \cos(\omega_0 t) \end{pmatrix} \begin{pmatrix} 1 & -c_1 \\ c_1 & 1 \end{pmatrix} \begin{pmatrix} \cos(\omega_0 t) & -\sin(\omega_0 t) \\ \sin(\omega_0 t) & \cos(\omega_0 t) \end{pmatrix} \mathbf{u}_0$$

$$= (1 + c_1 c_2) \tag{7.4.18a}$$

Similarly, for $\Omega^{(2)}$ we get

$$\Omega^{(2)} = \omega_0 (c_2 - c_1) \tag{7.4.18b}$$

Thus the nonlinear phase diffusion equation for the GL model takes the explicit form

$$\frac{\partial \psi}{\partial t} = (1 + c_1 c_2) \quad \nabla^2 \psi + \omega_0 (c_2 - c_1)(\nabla \psi)^2 \tag{7.3.19}$$

For the unperturbed GL equation we can define the amplitude R and phase Φ via the relation, $W = R \exp(i\Phi)$ and from Eq. (7.4.4) we have $d\Phi/dt = \omega_0$. On the other hand from Eq. (7.4.19) $d\psi/dt = 0$ for an unperturbed orbit. This implies a relation $\psi = \omega_0 (t + \psi)$ for a weak perturbation. It follows that whenever diffusional coupling is weak, or the orbital deformation due to diffusion is negligible, the GL equation is contracted to

$$\frac{\partial \Phi}{\partial t} = \omega_o + (1 + c_1 c_2) \nabla^2 \Phi + \omega_o (c_2 - c_1)(\nabla \Phi)^2 \qquad (7.4.20)$$

after putting $R = 1$. Equation (7.4.10) is known as the phase diffusion equation and the parameter α as the phase diffusion coefficient. Equation (7.4.20) breaks down if $\alpha < 0$. This condition implies that in an oscillatory system spatially uniform oscillations can become unstable in presence of diffusion with respect to variations in phase. This is known as phase instability, and is distinctly different from the Turing type of diffusional instability discussed in Chapter V.

In the present study the stability of the GL equation has been studied with $\alpha > 0$ as well as with $\alpha < 0$. Further the phase diffusion equation has also been studied for the case $\alpha > 0$. Through out this analysis we are subject to the condition that type I instability precedes type II instability, in order to eliminate the possibility of the diffusional instabilities setting in. The numerical procedure employed is the same as in chapter V. The results obtained are discussed in the next section.

7.5 Results and Discussion

The parameters in the substrate-inhibition have been investigated for the occurrence of the condition $\alpha < 0$. Fig. 7.1 shows the mapping of the parameter α against j_1 for fixed values of other parameters. This figure shows that this condition can occur for a wide range of realistic parameter values. In order to ensure that type I instability occurs before type II, we have solved the conditions for type I and type II instabilities numerically. Fig 7.2 and 7.3 depict the short and long time behaviors respectively of the GL equation in three distinct regimes $\alpha > 0 (c_1 = 0.7836, c_2 = 0.0236)$, $\alpha \sim 0 (c_1 = 3.1727, c_2 = -0.5167)$ and $\alpha \ll 0 (c_1 = 107.6, c_2 = -2.3779)$. From the figures it is clear that the solution to the GL equation is unstable even on very short time scales for the case $\alpha \ll 0$. For the cases $\alpha > 0$ $\alpha \sim 0$ the solution is considerably stable even for long time periods. The contour maps of the long time behavior of these three cases, shown if Fig 7.4 clearly indicate that when $\alpha \ll 0$ a spatio-temporal chaotic behavior sets in. Further, the phase diffusion equation was integrated

Fig. 7.2 : Space-time variation of the amplitude (on a short time scale) for different values of α ; a, b show the real and imaginary parts for $\alpha \sim 0$; for c, d $\alpha > 0$ and e, f $\alpha \ll 0$.

Fig. 7.3 : Long time behavior of amplitude in space for the cases discussed in Fig. 7.2.

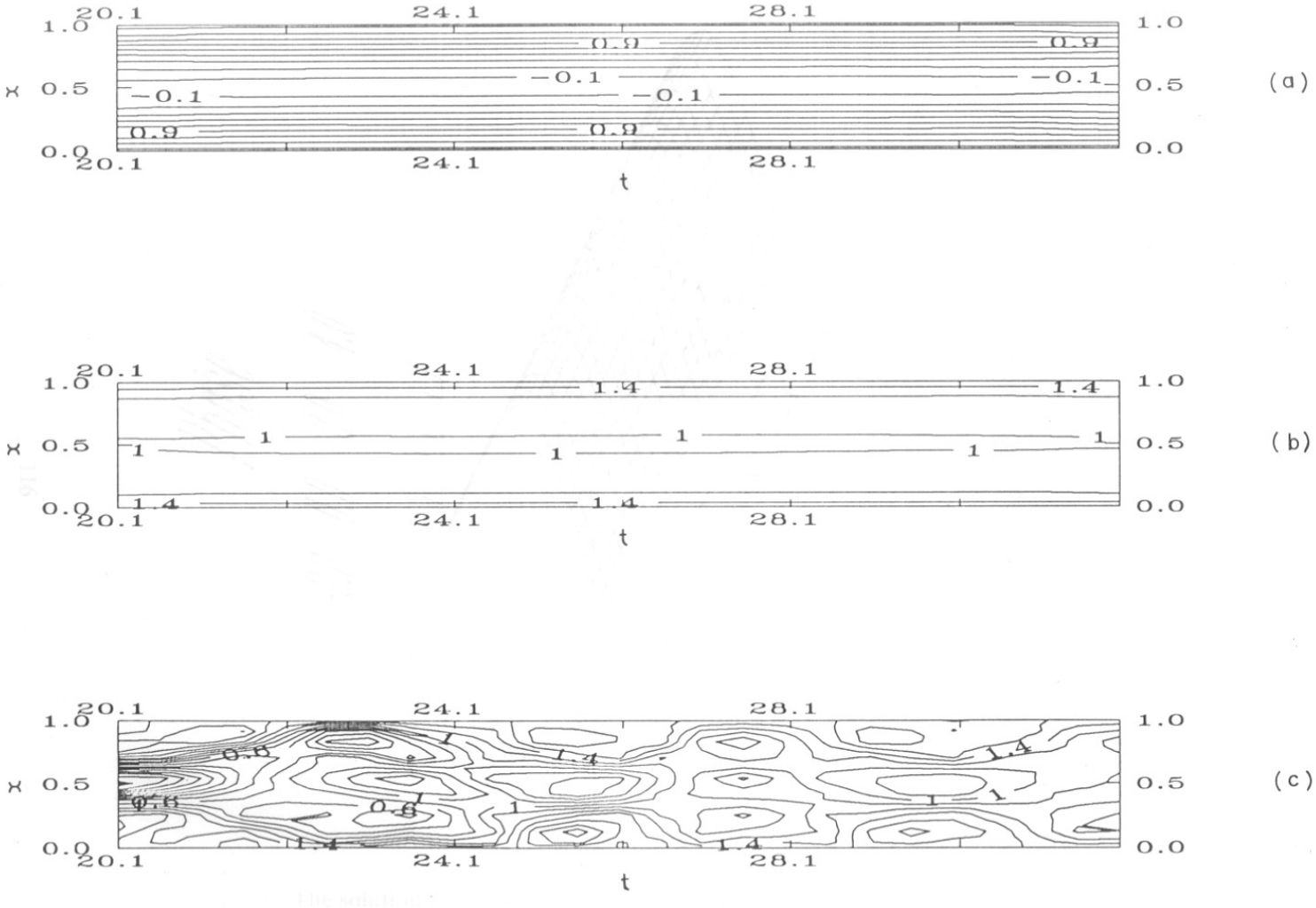

Fig. 7.4 : Contour plots for the three cases depicted in Fig. 7.3.

Fig. 7.5 : The solution to the phase diffusion equation for $\alpha > 0$.

using the same numerical procedures and periodic conditions. Figure 7.5 shows that the behavior of the GL equation can be effectively described through the phase diffusion equation, when $\alpha > 0$. For the case $\alpha < 0$ the equation cannot be integrated due to numerical instabilities.

The phase diffusion equation derived in this equation is central to the study of coupled oscillators. It has been shown to be useful in the studies of oscillators subject to periodic forcing, entrainment or synchronization of two or more oscillators, onset of collective oscillations, and also in the studies of populations of oscillators. The concepts of mutual synchronization and entrainment have proved to be useful in understanding chemical wave propagation in reaction-diffusion systems, origin of expanding target patterns and rotating spiral waves. In addition, chemical turbulence phenomena can also be effectively studied with the help of the phase diffusion equation.

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CHAPTER VIII

A CRITICAL ANALYSIS OF THE GINZBURG LANDAU EQUATION

The present chapter attempts to give an insight into the GL equation, in terms of the physical parameters of the reaction-diffusion systems. The physical relevance of the constants of the GL equation, c_1 and c_2 is discussed in detail. A brief literature survey of the applications of the GL equation to various fields is presented with a view to bring forth the universality of this equation and its capacity to show a rich and varied behavior in different regimes. Finally the scope for the application of this equation to some biological problems is discussed.

8.1 Introduction

In the introductory remarks of the meeting on 'Theories of Biological Pattern Formation', J. D. Murray had observed that a remark such as "It's probably a second order Hopf bifurcation in the p.d.e parameter space" does not have the biologists on the edge of their seats unless to leave (Murray, 1981). The usefulness of a mathematical theory to biology or any other field is complete, only when all the quantities involved can be effectively described in terms of realistic parameters of the system. Although the GL equation has become increasingly popular in the study of self-organization phenomena, we find that the studies are as yet, confined mostly to the general area of physics such as, hydrodynamics, binary mixtures and optics and the discussions given in these studies are somewhat abstract. In the present chapter we aim to give a critical analysis of the GL equation, its applicability in different parameter regimes, the physical meaning of the GL constants, and some potential applications of this equation to some problems of biological interest.

8.2 A critical analysis of the GL equation

In the present thesis we have often referred to the ubiquitous biological oscillations. Conditions leading to the bifurcation to a limit cycle behavior have been thoroughly studied by many authors and the further studies on bifurcations from the limit cycles leading to other dynamical complexities and chaos, is an interesting area of research. A remarkable feature about the biological systems however, is the stability they show even in face of many degrees of freedom and their capacity for self-organization through interacting subunits. Thus of greater importance to biological systems is the understanding of how the limit cycle oscillators interact with each other in a cooperative way leading to self-organization phenomena. The GL equation serves this purpose well, since it describes a field of oscillators which are diffusionally coupled.

As we have already seen the derivation of the GL equation is based on the slaving principle, which assumes that at critical points the number of degrees of freedom can be reduced to just a few. This brings us to the question of the regions of applicability of this principle. In this context, we see that the system size is a very important criterion. In Chapter IV we saw that the slaving principle forms the basis for the derivation of the GL equation. According to this principle, we can eliminate the fast modes, near the critical points, in order to get a simplified description. We see that for reaction-diffusion systems, wherein the description comes out in terms of PDEs, the number of degrees of freedom is infinite. In such cases, the reduction in the number of degrees of freedom depends upon the ratio of the time scales on which the slow and the fast modes evolve: τ_0 and τ_1 respectively. If $\tau_1/\tau_0 \ll 1$, then it implies that the slaving principle can be applied effectively. This condition can be stated alternately as $\mu \ll \zeta^{-2}$, where ζ is the system size and μ is the distance from the critical value of the bifurcation parameter. Once μ and ζ become comparable then the number of degrees of freedom coming into play becomes very large, and then the stability of the bifurcating solution to the homogeneous systems also becomes questionable. The condition stated above, gives rise to three distinct regions in the parameter space, *viz.*, $\mu \ll \zeta^{-2}$, $\mu \sim \zeta^{-2}$ and $\mu > \zeta^{-2}$. In the first two cases, the system size is very large, and the number of degrees of freedom is finite. In these regions, we find that the interplay of spatial modes can lead to pattern formation. In the third region, the system size is small and there are a few effective degrees of freedom, giving rise to a possibility of successive bifurcations, leading to chaos.

8.3 The Physical Significance of The GL Constants

For a reaction-diffusion system, the diffusion terms are incorporated through the diffusion matrix D. In a given reaction, if only one of the reacting species is diffusing, then

D reduces to a scalar quantity then the parameter d defined in Eq. (4.2.22) is a real positive number, which implies that the constant c_1 becomes zero. Thus, c_1 is a measure of the deviation of D from a scalar quantity. On the other hand c_2 is a measure of how the frequency of the individual oscillators depends on their amplitude. This is because, c_2 is derived from g (defined in Eq. (4.2.16)), which is a measure of the nonlinearity of the system. A positive value of g means that the bifurcation is supercritical and the solution is a stable limit cycle, whereas a negative value of g implies a subcritical bifurcation. A large value of c_1 compared to that of c_2 leads us to a reaction controlled regime, whereas the situation with large c_2 and small c_1 leads us to a diffusion controlled regime.

When c_1 and c_2 reduce to zero, then we obtain non oscillatory type of solutions leading to symmetry breaking instabilities, such as those found in Benard convection and Taylor vortices. On the other hand very large values of c_1 and c_2 indicate that the system is operating in a dissipationless regime. In this regime, the GL equation can be reduced to a nonlinear Schrodinger equation, which is a well known solition producing system. Solitons can travel without change of shape and at speeds faster than pure diffusion processes and can pass through cell walls to give rise to a cascade of reactions. Such a phenomenon clearly provides a basis for transmission of chemical signals, and cell to cell communication.

Further, we find that in regions of moderately low values of c_1 and c_2 we find a plethora of dynamical features such as traveling waves, periodic wave trains, rotating spiral waves, kinks and pulses, some of which are discussed in the next section.

8.4 Some Practical Applications of GL equation

In the recent years, the GL equation has become very popular in the studies of self-organization phenomena in dissipative systems (A. C. Newell, 1988). This is due to the

fact that this simple equation can show a wide range of dynamical features, ranging from simple oscillatory behavior to turbulence. Though the GL equation was first derived for problems arising in hydrodynamics, and optics (Newell and Whitehead, 1969 and Haken 1975) it has been extended to the general class of reaction-diffusion systems (Kuramoto, 1984). Physics literature is replete with studies on the GL equation aimed at obtaining different possible solutions to this equation. For instance, periodic wave solutions, and their stability investigated by were obtained by Stuart and DiPrima (1978); slowly varying waves were studied by Bernhoff (1988); Slow time periodic, bursting, quasiperiodic, homoclinic and heteroclinic solutions were also shown to exist (Doelman, 1989 and the references therein). Moon *et al* (1983) have shown transition to chaos in the GL equation, and the dimensions of the chaotic attractor were estimated by Ghidaglia and Heron (1987) and Doering *et al* (1988). The GL equation in the weak-dissipation regime, *i.e.*, in the near-nonlinear-Schrodinger regime has been transformed to a three dimensional model, which, for some parameters coincides with the famous Lorenz model (Malomed and Nepomnyashchy, 1990a). The latter is an established chaos generating model.

The GL equation, in its most commonly used form, shows that the supercritical bifurcating solution is stable and the subcritical one is unstable. However, when higher order terms are taken into consideration, the subcritical bifurcating solutions become important. The study of pulses, fronts, and wave trains for the GL equation, in the subcritical regime has received wide attention, since the discovery of stable pulses by Thual and Fauve (1988). Subcritical Hopf bifurcations, which generate stable localized waves such as solitary waves, widely observed in hydrodynamics, were studied by Fauve and Thual (1990). Pulses and wave fronts which appear in binary-fluid convection, plane Poiseuille flow and Taylor-Couette flow, were studied effectively by van Saarloos and Hohenberg (1990). Malomed and

Nepomnyashchy (1990b) have shown that in a subcritical range, a stable trivial solution can coexist with a stable traveling wave giving rise to the possibility of kinks and solitons for the GL equation. Small-amplitude periodic, stable quasiperiodic, and chaotic solutions were obtained in the same range, which can possibly explain the dispersive chaos found in binary fluid mixtures (Schopf and Kramer, 1991).

In addition, the GL equation can be helpful in understanding the systems of coupled oscillators and the features arising therein, through the phase diffusion equation.

Finally we would like to comment that the GL equation is a universal equation, and its field of application is very wide. Indeed it can prove useful for the description of any system that comes under the perview of synergetics. In biology, it has so far been used only in morphogenesis and developmental biology. Some of the other fields of potential application of the GL equation are neural networks, evolution, population dynamics, metabolic processes, behavioral biology, pattern formation and pattern recognition.

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CHAPTER IX

CONCLUSIONS

Conclusions

The central theme of the present thesis is the application of some advanced analytical and semi-analytical methods to the analysis of biological systems, with a view to gain an insight into the processes underlying the complex behavior of these systems. The emphasis has mainly been on the methods of reducing the number of degrees of freedom near the critical points in the system. Two specific models of biological significance have been considered, in the present analysis: the exponential autocatalysis and the Seelig's oscillator.

In the first chapter an extensive bifurcation analysis of the exponential autocatalysis has been carried out. The emphasis has been on obtaining the conditions for occurrence of known bifurcation features like the hard mode instability. A complete bifurcation map has been drawn for this model in order to characterize the possible behavior for different parameter regimes. The results obtained indicate that the model is capable of rich dynamic behavior. A variety of features like multiplicity, unstable and stable oscillatory solutions, unique stable and unstable states, saddle points and points of homoclinic bifurcation are obtained.

In the next chapter we exploit the presence of two different time scales inherent in this system, to eliminate the fast variables. The system is analyzed in the presence of diffusion. Global uniform solutions depending on whether the critical wave number is odd or even, are constructed. Conditions under which these dissipative structures are stable are also obtained. Further the conditions describing the evolution of a limit cycle and conditions for its stability are also obtained. Time heirarchies are known to play an important role biological processes and a method of analysis presented here can prove to be useful in approximating the behavior of such processes.

Much of the analysis presented in the rest of the thesis concentrates on simplifying the behavior of complex systems near bifurcation points, in particular the Hopf bifurcation point.

Chapter IV deduces a simple universal equation, which serves this purpose. The method of deduction is illustrated through the example of the exponential autocatalysis. This equation is central to any further analysis of the spatio-temporal evolution of the reaction-diffusion systems. The spatio-temporal features arising in the substrate-inhibition kinetic model are obtained using the GL equation. For this purpose, an extensive numerical analysis of the GL equation is carried out in the neighborhood of the onset of the kinetic and the diffusional instabilities. Further, the stability of the structures arising is tested by studying the evolution of the amplitude of the GL equation for short as well as long times. The results indicate that the predominant feature in the neighborhood of the first type of instability is spatially uniform oscillations. However, if the diffusional instabilities set in before the onset of spatially uniform oscillations, then spatial inhomogenities start appearing. The stabilities of these structures are also studied with respect to the GL constants. The results obtained indicate that in a reaction controlled regime the value of the constant c_1 does not affect the system behavior. Certain interesting features like the space-time dislocation of structures arise for certain parameter values.

Further, the evolution of the system under the influence of noise is studied for the case of substrate-inhibition kinetics. A simple equation of the form of the GL equation is obtained. This method of analysis of fluctuations can easily be extended to a general class of fluctuations, to analyze a variety of situations.

Next, the perturbation methods have been extended to the analysis of coupled oscillators. Here, we encounter a second type of diffusional instability, known as the phase instability. A nonlinear phase description equation is derived for this purpose. With the help of this equation, the stability of the uniform oscillatory solutions to the GL equation for the substrate-inhibition case is analyzed. We find that for certain realistic parameter values, the uniform oscillations

become unstable to due to diffusional effects and spatio-temporal chaos can occur.

In conclusion the present thesis aims at extending some advanced mathematical methods to problems of biological interest with a view to investigate the conditions for the occurrence of various types of dynamical features and the evolution of the system in some asymptotic regions.

List of Publications :

1.	Diffusive instability near Hopf bifurcation for exponentially autocatalyzed reaction-diffusion system	S. R. Inamdar, P. Rajani & B.D.Kulkarni.	*J. Phys. A Math. Gen.* **23**, 1990, L1293
2.	Multi-timescale approach to analysis of exponential autocatalysis: limit cycle and global non-uniform steady patterns	S. R. Inamdar, P. Rajani & B.D.Kulkarni.	*J. Phys. A Math. Gen.* **24**, 1991, 2539
3.	Chemical instabilities and bifurcations in Encillator	S. R. Inamdar, P. Rajani & B.D.Kulkarni.	*J. Phys. Chem.* **95**, 1991, 3422
4.	Bursting solutions for cubic autocatalysis in a continuous stirred tank reactor with recylce and time delay.	Parkash Badola, P. Rajani, V.RaviKumar & B.D.Kulkarni.	*J. Phys. Chem.* **95**, 1991, 2939
5.	Reduction of reaction-diffusion system to Schrodinger-like equation	S.R. Inamdar, P. Rajani & B.D.Kulkarni.	*AIChE* 1991, Annual meeting (accepted)

6.	Spatio-temporal Chaos in Reaction Diffusion systems	P. Rajani, J.K.Bandyopadhyay S.R. Inamdar and B.D. Kulkarni.	(Under preparation)
7.	Ginzberg-Landau equation for Reaction- diffusion systems	P. Rajani, S.R. Inamdar and B.D. Kulkarni.	(Under preparation)