The Analytical and Numerical Analysis of a Model of a Chemical Oscillator

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September 1989
This thesis is submitted for the award M Sc and is based on the candidates own work

For my parents

Acknowledgements

I wish to express my gratitude to Dr David Reynolds for his endless patience and constant enthuasism shown throughout the duration of this research Without his enduring optimism in the face of seemingly insolvable problems, much of this work would not have been completed

I also wish to thank Joe Crean and Colm Mc Guinness for their help in the typing and proof reading of the text, and for the use of Colm's Graph program

Abstract

This thesis concerns the analysis of the Exlpodator model for a Belousov-Zhabotinskii type oscillating chemical reaction The chemical kinetics of the reaction is discussed in detail and a system of kinetic equations, the Explodator, modelling the system is derived The equations are reduced to the system of non-dimensionalised equations

$$\begin{aligned} x_1 &= 2\mu_2 + x_1(1 - 3\mu_3) - x_1x_2 - 3\mu_1x_1^2, \\ x_2 &= \mu_4 - \beta x_2 + 3\alpha x_3 - x_1x_2, \\ x_3 &= \mu_3 - 2\alpha x_3 + x_1x_2 + \mu_1x_1^2 \end{aligned}$$

The existence for all time and boundedness of solutions of the Explodator are proved It is also proved that any trajectory solution which starts in the positive octant subsequently remains in it and that the model has a unique equilibrium point in the positive octant for a wide range of parameter values

The theory of Hopf bifurcation is introduced Stability is defined and the Hopf bifurcation theorem is explained The stability properties of the equilibrium solutions are examined A result is then proved that gives simple necessary and sufficient conditions in terms of the kinetic parameters, for an equilibrium point of the system to a be Hopf bifurcation point, and thus for there to be a family of limit cycle solutions AUTO, a software package for continuation and bifurcation problems in ordinary differential equations, is used to solve the system and to determine the stability of the periodic solutions The numerical solutions of the model agree very well with the chemical kinetics of the reaction and mathematical theory

Centre Manifold theory is used to reduce the model to a two-dimensional system with the same stability properties as the full system AUTO is then used to verify that the linear stability of the stationary solutions of the reduced system agree with that of the solutions of the full model

Contents

Chapter 1

Introduction

1.1 Oscillating Chemical Reactions

Over the past twenty years there has been a large amount of interest in biological and chemical systems which can sustain temporal and spatial oscillations In the field of chemical oscillators, the Belousov-Zhabotinskii reaction is one of the most widely studied chemical reactions of recent years It has been examined by a wide variety of scientists, mathematicians and engineers This interest is due to the fact that it is easily carried out and, although chemically complicated, it is still simple compared with examples of oscillating processes arising from biology The Belousov-Zhabotinskii reaction, which is the name given to the cerium ion catalysed oxidation of malonic acid in a sulphuric acid medium, has some very unusual properties It exhibits temporal oscillations in the concentration of several of the species present in the reaction mixture In the presence of an indicator, these oscillations are seen by the reagent periodically changing colour between blue and red When the reagent is spread thinly, circular chemical waves propagate outwards from a centre The waves are blue and they travel through a red background

Several systems of first order non-linear differential equations have been proposed as models for the Belousov-Zhabotinskii reaction, the best known being the Oregonator model [12] Mathematicians became interested in the Belousov-Zhabotinskii reaction because these models provide a new field in which to apply modern methods of nonlinear differential equations

1.2 Problem Statement

The Explodator is a model not only for the Belousov-Zhabotinskii reaction, but for many other chemical oscillators It consists of the *Explodator core* and one or more

Limitation reactions The core is not changed, but different limitation reactions are included for different oscillating systems The Explodator Core consists of the four reaction steps

$$\begin{aligned} A + X &\longrightarrow (1+a)X, \\ X + Y &\longrightarrow Z, \\ Z &\longrightarrow (1+b), \\ Y &\longrightarrow \text{Products}, \end{aligned}$$

where A is an initial reactant, X, Y and Z are intermediate species and a and b are positive constants whose value lies between zero and one The Explodator core on its own will not produce an oscillating scheme At least one limitation reaction must be included in the model to ensure that the consumption of the intermediate species is exceeded by production in the net process

The oscillating scheme examined in this thesis is a Belousov-Zhabotinskii type reaction It is an oxalic acid substrate system, where elementary bromine produced as a by-product of the reaction is removed by a stream of an inert gas For this system Noszticzius *et al* [23] proposed four limitation reactions and stated that the inclusion of any one of these limitation reactions with the core produces an oscillating scheme Since each of the limitation reactions expresses part of the underlying chemical mechanism, we include all of them here, and consider the *full Explodator model* This gives the following system of non-dimensionalised equations

$$\begin{aligned} x_1 &= 2\mu_2 + x_1(1 - 3\mu_3) - x_1 x_2 - 3\mu_1 x_1^2 \\ x_2 &= \mu_4 - \beta x_2 + 3\alpha x_3 - x_1 x_2 \\ x_3 &= \mu_3 - 2\alpha x_3 + x_1 x_2 + \mu_1 x_1^2 \end{aligned} \tag{1 1}$$

where x_1, x_2, x_3 are scaled concentrations of the intermediate species and α, β, μ_1, μ_2, μ_3, and μ_4 are functions of the rates of reaction

1.3 Basic Concepts

If the model is to realistically mirror a chemical oscillator, the model must reflect the properties of the chemical system x_1, x_2, x_3 are scaled concentrations of the intermediate species and should therefore each be positive Reproducible chemical oscillations must have some stabilising mechanism which drives the system into a stable closed orbit Mathematically this means that (1 1) must have a *stable limit cycle* solution A stable limit cycle Γ is a closed periodic orbit in phase space such that every trajectory which begins sufficiently near to Γ is attracted to it

The main tool which we shall use to show that (1 1) has a periodic orbit is the *Hopf bifurcation theorem* It considers the situation where an equilibrium point of a system exchanges stability as a parameter crosses a critical value The theorem provides conditions which guarantee that there is a family of periodic orbits emanating from this equilibrium point There is therefore a qualitative change in the phase space as this critical parameter value is traversed At such a critical parameter value the equilibrium point is called a *Hopf bifurcation point*

1.4 Thesis Outline

Chemical aspects of an oscillating chemical reaction are dealt with in Chapter 2 The chapter begins with a historical outline of the work carried out on the Belousov-Zhabotinskii reaction and a brief review of the best known model for the reaction, the FKN model Due to problems arising from the difficulty of modelling such a chemically complex system, a heterogenous Belousov-Zhabotinskii type reaction is then introduced and its chemical mechanism is discussed The Explodator is then suggested as a model for the reaction and the system (1 1) is derived

The existence for all time and boundedness of solutions of (1 1) are proved in Chapter 3 We also prove that any trajectory of (1 1) which starts in the positive octant subsequently remains in it Then it is shown that (1 1) has a unique equilibrium point in the positive octant for a wide range of parameter values Finally the chapter reviews the work carried out by other authors, the behaviour of solutions both in the absence of any limiting reactions and with the inclusion of only one limiting reaction

The theory of Hopf bifurcation is introduced in Chapter 4 Stability is defined and a version of the Hopf bifurcation theorem is explained To illustrate the application of this theorem, a simple system exhibiting limit cycle solutions is discussed

The stability properties of the equilibrium solutions of (1 1) are examined A result is then proved that gives simple necessary and sufficient conditions in terms of the kinetic parameters, for an equilibrium point of (1 1) to a be Hopf bifurcation point, and thus for there to be a family of limit cycle solutions It is a triumph that such a simple theorem has been found, because the large number of parameters in (1 1) make hand manipulations almost impossible The symbolic manipulator MACSYMA did not help either The result enables all the single limiting reactions to be discussed in detail

AUTO, a software package for continuation and bifurcation problems in ordinary differential equations, was used to solve (1 1) and to determine the stability

of the periodic solutions The numerical solutions of (1 1) agreed very well with the chemical mechanism described in Chapter 2 and the mathematical theory developed in Chapter 4

The complexity of the manipulations involved precluded determining the stability of the limit cycles using Hopf's theory Therefore Centre Manifold theory is used in Chapter 5 to reduce (1 1) to a two-dimensional system with the same stability properties as (1 1), in a limited parameter range This also required extensive calculations [1] AUTO was then used to verify that the linear stability of the stationary solutions of the reduced system agreed with that of the solutions of (1 1)

[1]The calculations in this chapter were initially done by hand and checked using the symbolic manipulator MACSYMA

Chapter 2

The Belousov-Zhabotinskii Reaction

2.1 Historical Outline

Oscillating or periodic phenomena are common to many areas of physics, biology and astronomy Examples of oscillating processes include the orbits of planets, the motion of pendulums and the biological clocks that govern our internal organs Until the mid twentieth century, chemists believed that the existence of chemical reactions which exhibit temporal or spatial oscillations was prohibited by the Second Law of Thermodynamics This law states that the entropy of the universe tends to increase Applied to chemical reactions the principle states that a closed chemical system at constant temperature and pressure must continuously approach an ultimate equilibrium state That is, if two substances react to form a third substance, it is expected that the reaction will continue steadily until the reactants are exhausted or an equilibrium is reached

In 1958 a Russian chemist, B P Belousov [1], accidentally discovered a system which seemed to defy the second law of thermodynamics He noticed that if citric acid and sulphuric acid are dissolved in water with potassium bromide and a cerium salt, the colour of the mixture changes periodically from colourless to pale yellow Although accounts of reactions such as this had been reported before these were mainly dismissed as non reproducible phenomena Belousov's reaction differed because it was easily reproduced In 1964, A M Zhabotinskii [31] began a systematic study of Belousov's reaction He modified the reaction by adding an indicator which produced a more dramatic colour change He also discovered that if a thin layer of the reagent is left undisturbed blue dots appear which spread out a pattern of spiral bands of alternate colour As a result of Zhabotinskii's work the reaction is now commonly called *The Belousov-Zhabotinskii Reaction*

There was increased interest in such reactions as a result of the work of Prigogine [26] Prigogine was the first to point out that oscillations are in fact possible for some systems provided they are far enough from equilibrium In such systems it is the concentrations of the intermediate and catalyst species that oscillate not the initial and final species For his work in this area Prigogine recieved the Nobel Prize for chemistry in 1977

Although the Belousov-Zhabotinskii reaction became well known as a result of Zhabotinskii's work, very little was known about the chemical mechanism of the reaction In 1972, R Field, E Koros and R Noyes [11] produced a detailed reaction mechanism which was widely accepted and is commonly called the FKN model Since then the FKN model and it's skeletonised version, the Oregonator [12], has served as a basis for study in the area of chemical oscillations J Tyson [29] produced an extensive review of the work done on the Oregonator in 1976

In 1984, Z Noszticzius, H Farkas and Z A Schelly [23] published a paper which proposed an alternative skeleton model due to experimental facts which emerged that were difficult to explain with the Oregonator [20,21] One of the problems with the model was due to the kinetic parameter F The kinetic behaviour of the model depends critically on F When $F < 1/4$ oscillations do not occur in the Oregonator model However the reactions included in the model would only produce an F value of less than $1/4$ The model suggested by Noszticzius *et al* does not include such a parameter

2.2 The Field, Köros and Noyes Model

The FKN model may be summarised in the five main steps

$$Br^- + HBrO_2 + H^+ \longrightarrow 2HOBr, \quad (2\,1)$$

$$Br^- + BrO_3^- + 2H^+ \longrightarrow HBrO_2 + HOBr, \quad (2\,2)$$

$$2Ce^{3+} + BrO_3^- + HBrO_2 + 3H^+ \longrightarrow 2Ce^{4+} + 2HBrO_2 + H_2O, \quad (2\,3)$$

$$2HBrO_2 \longrightarrow HOBr + BrO_3^- + H^+, \quad (2\,4)$$

$$nCe^{4+} + BrOx + Ox \longrightarrow nCe^{3+} + Br^- + \text{oxidised organic species}, \quad (2\,5)$$

where $BrOx$ and Ox represent brominated and unbrominated organic species respectively

The oxidation of organic species by Ce^{4+}, described by reaction 2 5, is extremely complex when the organic substrate is malonic acid As a result the model contains an unknown parameter which depends on the stoichiometry of

reaction 2 5 This problem may be avoided by finding an organic substrate with limited mechanistic possibilities for oxidation with Ce^{4+}

2.3 Heterogenous BZ Oscillators

Noszticzius and Bodiss [22] found that if oxalic acid is used as the organic substrate and the Br_2 produced is removed by bubbling an inert gas stream through the reaction mixture, a heterogenous type Belousov-Zhabotinskii (BZ) type reaction occurs This reaction proceeds in two stages in which different reactions are dominant For the purpose of the model we will use square brackets to denote the concentrations of the chemical species

Stage I

In aqueous solution bromic acid is a strong acid and a good oxidising agent It is reduced by oxalic acid to produce bromous acid, CO_2 and water according to

$$BrO_3 + (COOH)_2 \longrightarrow HOBr + 2CO_2 + H_2O$$

The cycle starts with the autocatalytic growth of $HBrO_2$ and the subsequent oxidation of Ce^{3+}

$$HBrO_3 + HBrO_2 + 2H^+ + 2Ce^{3+} \longrightarrow 2HBrO_2 + H_2O + 2Ce^{4+}$$

During this growth period $[Br^+]$ and $[HOBr]$ are low or negligible However as $[HBrO_2]$ starts to increase some $HOBr$ appears due to the reaction

$$2HBrO_2 \longrightarrow HOBr + HBrO_3$$

After a delay, large amounts of $HOBr$ are produced The $HOBr$ in turn produces Br_2 by the reaction

$$2HOBr + (COOH)_2 \longrightarrow Br_2 + 2CO_2 + 2H_2O$$

Eventually $[Br_2]$ reaches a high enough value so that Br_- is in equilibrium with the bromine

$$Br_2 + H_2O \rightleftharpoons HOBr + Br^- + H^+$$

It is because of this step that it and similar Belousov-Zhabotinskii oscillators are called bromine hydrolysis controlled oscillators However since the equilibrium in the hydrolysis lies well to the left ($k_{forward} = 110sec^{-1}, k_{reverse} = 8 \times 10^9 M^{-2}sec^{-1}$)[10, Ch 26], $HOBr$ is consumed in such large amounts that its consumption exceeds its autocatalytic growth At this point $[HBrO_2]$ reaches it maximum value and starts to fall Stage II then takes over

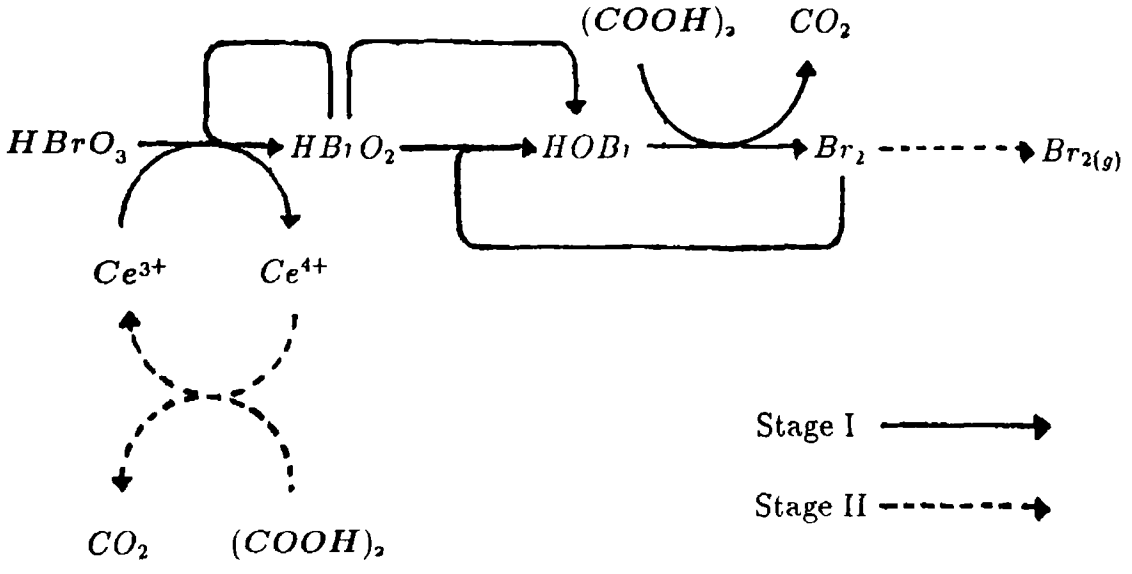

Figure 2 1 Schematic representation of the heterogenous Ce^{3+}-BrO_3^--Oxalic acid system Initial reactants are bolded in the diagram

Stage II

At this point $[Br_2]$ is still increasing due to accumulated $HOBr$ The Br_2 must be removed by some physical or chemical process Without such a process the oscillations would not occur In this case the Br_2 is removed by bubbling nitrogen gas through the reaction mixture The physical removal of Br_2 can be regarded as the chemical process

$$Br_2 \longrightarrow Br_{2(g)}$$

First the $[HOBr]$ and then the $[Br_2]$ start to decrease Due to the low $[HOBr]$, oxalic acid reacts instead to reduce Ce^{4+} according to

$$2Ce^{4+} + (COOH)_2 \longrightarrow 2CO_2 + 2H^+ + 2Ce^{3+}$$

Once $[Br_2]$ and $[HOBr]$ have become sufficiently low stage I can again take over

We may represent the reactions schematically by Figure 2 1 In Figure 2 2 we see how the Explodator model replicates how the concentrations of the intermediate species vary with time

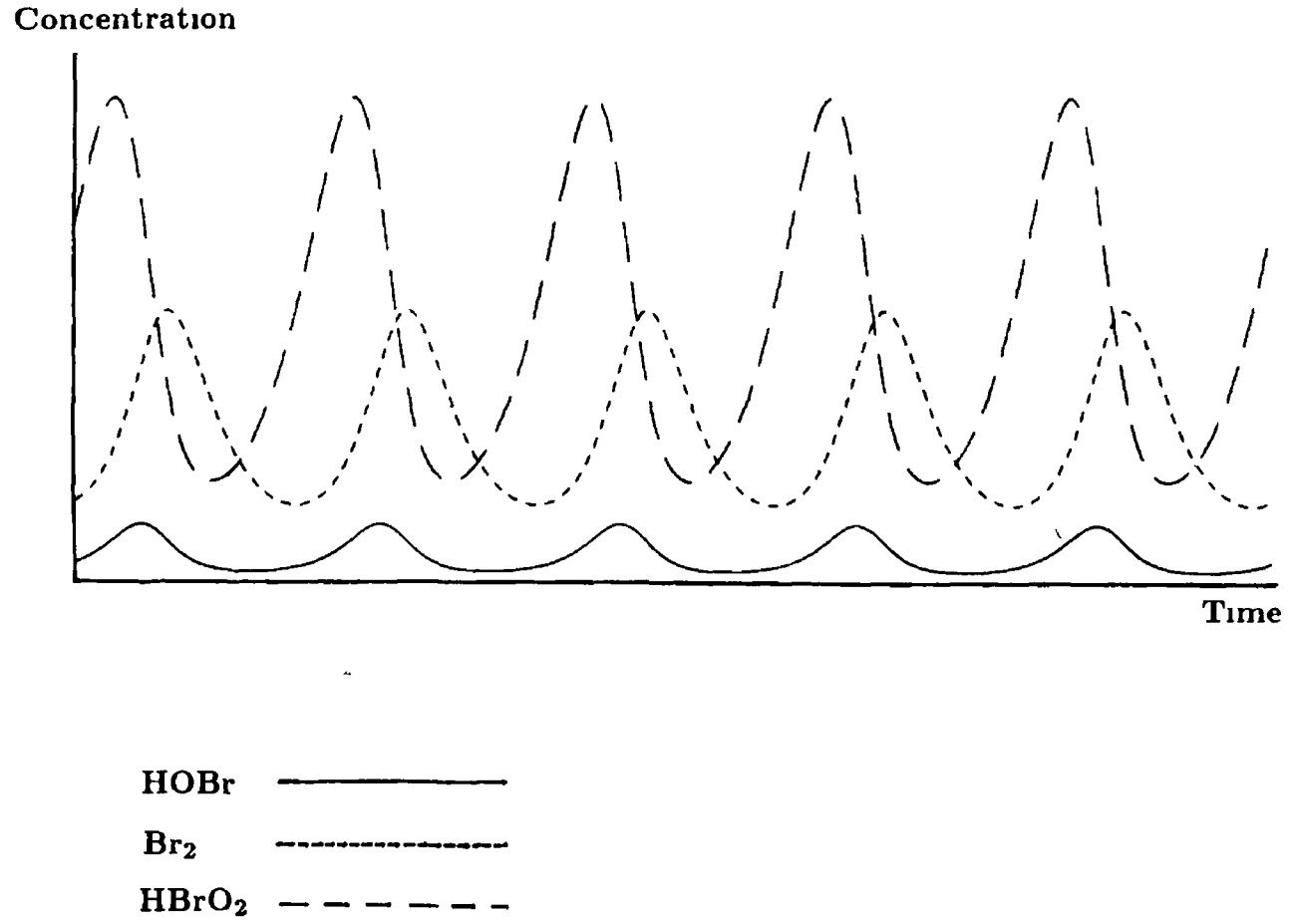

Figure 2 2 Variation in the concentration of the intermediate species with time (see section 4 4) During stage I, $HBrO_2$ increases until it reaches its maximum value, this produces an increase in the concentration of $HOBr$ and Br_2 However, once it has reached its peak, the concentration of $HBrO_2$ starts to decrease and after a delay the concentrations of the other species decay until the reach their minimum value- The cycle then repeats

2.4 The Explodator Model for BZ type Oscillators.

Due to shortcomings in the FKN model, Noszticzius, Farkas and Schelly [23] proposed an alternative scheme based on the heterogenous Belousov-Zhabotinskii oscillator described above The *Explodator* is a scheme which not only models the Belousov-Zhabotinskii reaction, but also can be generalised to include the Bray-Liebhafsky reaction [2], the Briggs-Rauscher reaction [3] and their modifications

The *Explodator Core* consists of the four main steps

$$\begin{aligned} A + X &\longrightarrow (1+a)X, \\ X + Y &\longrightarrow Z, \\ Z &\longrightarrow (1+b)Y, \\ Y &\longrightarrow \text{products}, \end{aligned}$$

where X, Y and Z are intermediate species, A is an initial reactant and a and b are positive values less than one However the Explodator core alone will not yield an oscillating scheme [18] Other reactions must be included to ensure that the production of the intermediate species is limited by consumption These reactions are thus called the *Limitation* reactions The Explodator core for the oxalic acid oscillating system described above can be modelled by the four main reactions

$$HBrO_3 + HBrO_2 + 2H^+ + 2Ce^{3+} \longrightarrow 2HBrO_2 + H_2O + 2Ce^{4+}, \quad (2\,6)$$

$$HBrO_2 + Br_2 + H_2O \longrightarrow 3HOBr, \quad (2\,7)$$

$$2HOBr + (COOH)_2 \longrightarrow Br_2 + 2CO_2 + H_2O, \quad (2\,8)$$

$$Br_2 \longrightarrow Br_{2(g)} \quad (2\,9)$$

We want to write these equations in the form given above The initial reactants $(COOH)_2$, Ce^{3+}, $HBrO_3$ and H_2SO_4 are used up slowly and thus their concentrations are much higher than those of the intermediate species present Sulphuric acid is used as a source of hydrogen ions, which are buffered by the bisulphate ion H_2SO_4, therefore $[H^+]$ does not change appreciably during the reaction Hence for the purpose of the model $[(COOH)_2]$, $[Ce^{3+}]$, $[HBrO_3]$, $[H_2SO_4]$ and $[H^+]$ will be considered constant over a short time period

We now let $A = [HBrO_3]$, $B = [Br_{2(g)}]$, $X = [HBrO_2]$, $Y = [3HOBr]$ and $Z = [Br_2]$

Reaction 2 6

$$HBrO_3 + HBrO_2 + 2H^+ + 2Ce^{3+} \longrightarrow 2HBrO_2 + H_2O + 2Ce^{4+}$$

This reaction occurs in several steps, its rate determining step being the formation of the $BrO_2^{\bullet}$ radical

$$H^+ + BrO_3^- + HBrO_2 \xrightarrow{k_1} 2HBrO_2^{\bullet} + H_2O$$

Thus in our model we write reaction 2 6 as

$$A + X \xrightarrow{k_1} 2X \tag{2 10}$$

Reaction 2 7

$$HBrO_2 + Br_2 + H_2O \xrightarrow{k_2'} 3HOBr$$

This is a simple one step reaction and its rate law is given by

$$R = k_2'[HBrO_2][Br_2][H_2O]$$

Since H_2O is present in large quantities its concentration may be considered constant and the rate law becomes

$$R = k_2[HBrO_2][Br_2],$$

where $k_2 = k_2'[H_2O]$ So we may write the reaction as

$$X + Y \xrightarrow{k_2} Z \tag{2 11}$$

Reaction 2 8

$$2HOBr + (COOH)_2 \longrightarrow Br_2 + 2CO_2 + H_2O$$

This is a complex reaction and its rate law cannot be determined from the above equation However, Noszticzius *et al* [23] have written the model equation in the form

$$Y \xrightarrow{k_3} 3Z/2 \tag{2 12}$$

Reaction 2 9

$$Br_2 \xrightarrow{k_4} Br_{2(g)}$$

The removal of Br_2 from the system is a physical not a chemical process It requires the bubbling of an inert gas stream through the reaction mixture This process can be regarded as a first order reaction where k_4 is a function of the gas flow rate and reaction volume For our model the reaction is written as

$$Z \xrightarrow{k_4} B \tag{2 13}$$

To complete the model we must include at least one limitation reaction Noszticzius *et al* [23] suggest as limitation reactions

$$2HBrO_2 \longrightarrow HOBr + HBrO_3, \tag{2 14}$$
$$HBrO_3 + (COOH)_2 \longrightarrow HBrO_2 + 2CO_2, \tag{2 15}$$
$$HBrO_2 + (COOH)_2 \longrightarrow HOBr + 2CO_2 + H_2O, \tag{2 16}$$
$$Br_{2(g)} \longrightarrow Br_2 \tag{2 17}$$

Again we let $A = [HBrO_3]$, $B = [Br_{2(g)}]$, $X = [HBrO_2]$, $Y = [3HOBr]$ and $Z = [Br_2]$

Reaction 2 14

$$2HBrO_2 \xrightarrow{k_{L1}} HOBr + HBrO_3$$

This is an elementary reaction and so we may write it as

$$2X \xrightarrow{k_{L1}} Y/3 + A \tag{2 18}$$

Reaction 2 15

$$HBrO_3 + (COOH)_2 \xrightarrow{k'_{L2}} HBrO_2 + 2CO_2 + H_2O$$

The rate law for this reaction may be represented by

$$R = k'_{L2}[HBrO_3][(COOH)_2],$$

which may be written as

$$R = k_{L2}[HBrO_3]$$

where $k_{L2} = k'_{L2}[(COOH)_2]$ Since $[(COOH)_2]$ is considered constant for the model then k_{L2} is also constant Thus reaction 2 15 may be written as

$$A \xrightarrow{k_{L2}} X \tag{2 19}$$

Reaction 2 16

$$HBrO_2 + (COOH)_2 \xrightarrow{k'_{L3}} HOBr + 2CO_2 + H_2O$$

As before this may be written as

$$X \xrightarrow{k_{L3}} Y/3, \tag{2 20}$$

where $k_{L3} = k'_{L3}[(COOH)_2]$

Reaction 2 17

$$Br_{2(g)} \xrightarrow{k_{L4}} Br_2$$

This is the reverse reaction of 2 9 Symbolically it may be written as

$$B \xrightarrow{k_{L4}} Z, \tag{2 21}$$

where $k_{L4} = k_{-4}$

The four limitation reactions are now represented by

$$\begin{aligned} 2X &\longrightarrow Y/3 + A, \\ A &\longrightarrow X, \\ X &\longrightarrow Y/3, \\ B &\longrightarrow Z \end{aligned}$$

2.5 Mathematical Formulation of the Explodator.

The *Law of Mass Action* states that the rate of a reaction is proportional to the active concentration of the reactants In fact, the rate of a reaction is the product of the rate constant for the reaction and the concentrations of the reactants involved Thus, the rate of change of the concentration of an intermediate species is the sum of the rates of the reactions where the intermediate species is produced, minus the sum of the rates of the reactions where the species is consumed On applying this law to the Explodator core and the limitation reactions, we get

$$\begin{aligned} \frac{dX}{d\tau} &= k_1AX - k_2XY - k_{L1}X^2 + k_{L2}A - k_{L3}X, \\ \frac{dY}{d\tau} &= -k_2XY + 3k_3Z/2 - k_4Y + k_{L4}B, \\ \frac{dZ}{d\tau} &= k_2XY - k_3Z + k_{L1}X^2/3 + k_{L3}X/3 \end{aligned}$$

The derivation of these reactions is quite easy For equation (2 22), the second term on the righthand side comes from (2 11), the third term from (2 18) and the fourth from (2 19) The first term in (2 22) comes from (2 10) and is the sum of $2k_1AX - k_1AX$ since one unit of X is consumed in the reaction while two units are produced

To transform the equations to dimensionless form we make the substitutions

$$x_1(t) = \frac{k_2 X(\tau)}{k_1 A}, \quad x_2(t) = \frac{k_2 Y(\tau)}{k_1 A}, \quad x_3(t) = \frac{k_2 Z(\tau)}{k_1 A},$$

$$\alpha = \frac{k_3}{2k_1 A}, \qquad \beta = \frac{k_4}{k_1 A}, \qquad t = k_1 A \tau,$$

$$\mu_1 = \frac{k_{L1}}{3k_2}, \qquad \mu_2 = \frac{k_2 k_{L1}}{2k_1^2 A}, \qquad \mu_3 = \frac{k_{L3}}{3k_1 A},$$

$$\mu_4 = \frac{k_{L4} k_2 B}{(k_1 A)^2}$$

Under this transformation the equations become

$$\begin{aligned} x_1 &= 2\mu_2 + x_1(1 - 3\mu_3) - x_1 x_2 - 3\mu_1 x_1^2, \\ x_2 &= \mu_4 - \beta x_2 + 3\alpha x_3 - x_1 x_2, \\ x_3 &= \mu_3 x_3 - 2\alpha x_3 + x_1 x_2 + \mu_1 x_1^2 \end{aligned}$$

Chapter 3

Existence and Uniqueness

3.1 Existence and Boundedness of Solutions

Our model system has been represented by the system of differential equations

$$\begin{aligned} x_1 &= 2\mu_2 + x_1(1 - 3\mu_3) - x_1x_2 - 3\mu_1x_1^2, \\ x_2 &= \mu_4 - \beta x_2 + 3\alpha x_3 - x_1x_2, \\ x_3 &= \mu_3x_1 - 2\alpha x_3 + x_1x_2 + \mu_1x_1^2, \end{aligned} \tag{3 1}$$

where x_1, x_2 and x_3 are scaled concentrations of the intermediate species and must be positive Therefore we look for solutions of (3 1) which satisfy $x_1(t) \geq 0, x_2(t) \geq 0$ and $x_3(t) \geq 0$, on some time interval [0,T)

In order to give a result on the existence of solutions of (3 1) we define

$$Q = \{(x_1, x_2, x_3) | x_1 > 0, x_2 > 0, x_3 > 0\} \tag{3 2}$$

Q is a *globally invariant* set for (3 1) if for every ξ in Q, the unique solution $x(\ ,\xi)$ of (3 1) satisfying $x(0) = \xi$, exists on $[0, \infty)$ and $x(t) \in Q$ for all $t > 0$

We may write (3 1) in the form

$$x = f(x), \tag{3 3}$$

where $x = (x_1, x_2, x_3)$ and $f \quad \mathbf{R}^3 \rightarrow \mathbf{R}^3$ Let Ω be an open subset of $\mathbf{R}^3$ where $\partial\Omega$ and $\overline{\Omega}$ denote the boundary and closure of Ω respectively A point $x_0 \in \partial\Omega$ is called an *egress point* of Ω with respect to (3 1) if for some solution $x(t)$ satisfying $x(t) = x_0$, there exists $\varepsilon > 0$ such that $x(t) \in \Omega$ for $t_0 - \varepsilon < t < t_0$ A point $x_1 \in \partial\Omega$ is called a *nonegress point* if it is not an egress point

Lemma 3 1 *[13] Let $U(x)$ be a real valued function on a neighbourhood N of $x_0 \in \partial\Omega$ such that $x(t) \in \Omega \cap N$ if and only if $U(x) < 0$ Then a necessary and*

sufficient condition for x_0 to be a nonegress point is that $U(x) < 0$ for $x \in \Omega$, where $U(x) = (grad\ U)\ f(x)$

Theorem 3 2 *The set Q is a globally invariant set for (3 1)*

Proof We define S_1, S_2 and S_3 by

$$\begin{aligned} S_1 &= \{(x_1, x_2, x_3) | x_1 = 0, x_2 \geq 0, x_3 \geq 0\}, \\ S_2 &= \{(x_1, x_2, x_3) | x_1 \geq 0, x_2 = 0, x_3 > 0\}, \\ S_3 &= \{(x_1, x_2, x_3) | x_1 > 0, x_2 > 0, x_3 = 0\}, \end{aligned}$$

then $\partial Q = S_1 \cup S_2 \cup S_3$ Q is globally a invariant set for (3 1) if every $x(t) \in \partial Q$ is a nonegress point

Assume first that $\mu_2 > 0$ We consider S_1 and let $U_1(x) = -x_1$, therefore $U_1(x) < 0$ for $x(t) \in Q$ On S_1

$$\begin{aligned} U_1 &= (-1, 0, 0)\ (x_1, x_2, x_3) \\ &= -(2\mu_2 + x_1(1 - 3\mu_3) - x_1 x_2 - 3\mu_1 x_1^2)_{x_1=0} \\ &= -2\mu_2 < 0 \end{aligned}$$

Similarly we consider S_2 and let $U_2(x) = -x_2$, thus $U_2(x) < 0$ for $x(t) \in Q$ On S_2

$$\begin{aligned} U_2 &= (0, -1, 0)\ (x_1, x_2, x_3) \\ &= -(\mu_4 - \beta x_2 + 3\alpha x_3 - x_1 x_2)_{x_2=0} \\ &= -(\mu_4 + 3\alpha x_3) < 0 \end{aligned}$$

Finally on S_3 we let $U_3(x) = -x_3$ so that $U_3(x) < 0$ for $x(t) \in Q$

$$\begin{aligned} U_3 &= (0, 0, -1)\ (x_1, x_2, x_3) \\ &= -(\mu_3 x_1 - 2\alpha x_3 + x_1 x_2 + \mu_1 x_1^2)_{x_3=0} \\ &= -(\mu_3 x_1 + x_1 x_2 + \mu_1 x_1^2) < 0 \end{aligned}$$

Hence any trajectory with an initial value in Q, cannot cross the boundary ∂Q and thus remains in Q

We now consider the case when $\mu_2 = 0$ As before $U_2 < 0$ and $U_3 < 0$, thus any trajectory starting in Q can only leave through S_1 We define

$$\Sigma = \{(x_1, x_2, x_3) | x_1 = 0\}$$

such that $S_1 \subset \Sigma$ Consider the initial value problem

$$\begin{aligned} x_1 &= 0, & x_1(0) &= \xi_1, \\ x_2 &= \mu_4 - \beta x_2 + 3\alpha x_3, & x_2(0) &= \xi_2, \\ x_3 &= -2\alpha x_3, & x_3(0) &= \xi_3, \end{aligned}$$

which has a unique solution Solutions of (3 1) which are in Σ for some initial time are always in Σ Thus Σ is an invariant manifold (surface) By uniqueness a trajectory cannot leave Q and intersect S_1 thus no trajectory can leave Q through S_1

In a closed chemical system, such as the one we are examining, all concentrations must be bound variables of time, because no new molecules are introduced into the system

Suppose that x_0 is in Q, and let x be the unique solution of (3 1) satisfying $x(0) = x_0$ For $i = 1, 2, 3$, $x_i(t) > 0$ for all t in the maximal interval of existence $[0, T)$ of this solution If $T < \infty$, $x_j(t) \to \infty$ as $t \to T$ for some j In order to show that this does not occur, we introduce

$$v(t) = x_1(t) + 2x_2(t) + 3x_3(t)$$

It is easy to show from (3 1) that

$$v = 2(\mu_2 + \mu_4) + x_1 - 2\beta x_2 < 2(\mu_2 + \mu_4) + v,$$

on $[0, T)$ Hence

$$\frac{d}{dt}(v(t)e^{-t} < 2(\mu_2 + \mu_4)e^{-t},$$

and therefore

$$v(t) < 2(\mu_2 + \mu_4)(e^t - 1) + v(0)e^t, 0 \leq t < T$$

Thus $T = \infty$ □

3.2 Equilibrium Solutions

The equilibrium solutions of (3 1) are solutions of

$$\begin{aligned} 0 &= 2\mu_2 + x_1(1 - 3\mu_3) - x_1 x_2 - 3\mu_1 x_1^2, \\ 0 &= \mu_4 - \beta x_2 + 3\alpha x_3 - x_1 x_2, \\ 0 &= \mu_3 x_1 - 2\alpha x_3 + x_1 x_2 + \mu_1 x_1^2 \end{aligned}$$

We find that there are two equilibrium solutions defined by

$$\begin{aligned} x_1 &= \frac{\beta(1-3\mu_3)-\mu_2-\mu_4 \pm \chi}{1+6\beta\mu_1}, \\ x_2 &= (x_1+2\mu_2+2\mu_4)/2\beta, \\ x_3 &= \{x_1^2+x_1(2\mu_2+2\mu_4+\beta)+2\beta\mu_2\}/6\alpha\beta, \end{aligned} \tag{3 4}$$

where

$$\chi = \{(\beta-3\beta\mu_3-\mu_2-\mu_4)^2+4\beta\mu_2(1+6\beta\mu_1)\}^{1/2} \tag{3 5}$$

Only the positive root has chemical significance since any limit cycle solution surrounding an equilibrium point which is not in Q will involve negative concentrations of the intermediate species It is easily shown that there is a unique solution in Q if either

$$\mu_2 > 0 \text{ or } \beta(1-3\mu_3)-\mu_4 > 0 \tag{3 6}$$

We denote by ξ this steady state solution in Q where $\xi = \xi(\alpha, \beta, \mu_1, \mu_2, \mu_3, \mu_4)$

3.3 Absence of Periodic Solutions of the Explodator Core.

The Explodator core consists of the reaction steps (2 6) to (2 9) and may be reduced, by letting $\mu_i = 0$ for $i = 1, \ ,4$, to the following set of differential equations,

$$\begin{aligned} x_1 &= x_1 - x_1x_2, \\ x_2 &= -\beta x_2 + 3\alpha x_3 - x_1x_2, \\ x_3 &= -2\alpha x_3 + x_1x_2 \end{aligned} \tag{3 7}$$

This system has only one equilibrium point in Q, namely,

$$\xi(\alpha, \beta) = (2\beta, 1, \beta/\alpha), \tag{3 8}$$

which was shown by Noszticzius *et al* [23] to be always linearly unstable (see definition on page 20) The global analysis of solutions of (3 7) was accomplished by Kertesz [18] His results are embodied in the following theorems

Theorem 3 3 *There is a one dimensional manifold, η, in Q in which, solutions of (3 7) tend to ξ as $t \to \infty$*

We let $Q_0 = Q/\eta \cup \xi$

Theorem 3 4 *(i) Solutions of (3 7) in Q_0 are not bounded*

(ii) If $\alpha > 1$ then all trajectories in Q_0 oscillate around ξ as they approach infinity

(iii) If $\alpha < 1$, then after a finite number of oscillations the trajectories in Q_0 cease to oscillate and

$$\lim_{t\to\infty} x(t) = \infty,$$

where $x(t) = (x_1(t), x_2(t), x_3(t))$

3.4 The Explodator Model with one limiting reaction.

Noszticzius, Farkas and Schelly proposed that the incorporation of any one of the limiting reactions into the model results in a system which exhibits limit cycle oscillations By considering the rate constant of the included reaction as a bifurcation parameter, μ, they state that, when $\alpha > 1$, the following can be proved for each case

(i) There exists a unique equilibrium point $\xi(\mu)$ in the set Q for every μ in an interval $[0, \mu_1)$ where $\mu_1 > 0$

(ii) There exists a critical parameter value, μ_0, in the interval $(0, \mu_1)$ such that

$$\xi(\mu) \text{ is unstable for } 0 < \mu < \mu_0$$

and

$$\xi(\mu) \text{ is stable for } \mu_0 < \mu < \mu_1$$

(iii) At the critical value μ_0 a Hopf bifurcation takes place, near the critical value a limit cycle exists

The above statements were proved in detail only for the inclusion of the limiting reaction (2 15)

Chapter 4

Hopf Bifurcation

Since periodic changes in the concentrations of the intermediate species are observed in the Belousov-Zhabotinskii reaction we look for stable limit cycle solutions of (3 1) in Q Here we analytically and numerically apply the Hopf Bifurcation Theorem

4.1 Linear Stability and Hopf Bifurcation Theory

The system (3 1) can be written in the form

$$x = F(x, \alpha), \tag{4 1}$$

where the function $F \quad \mathbf{R}^3 \times \mathbf{R} \to \mathbf{R}^3$ is analytic The parameter α is specifically indicated and the parameters $\beta \geq 0, \mu_1 \geq 0, \mu_2 \geq 0, \mu_3 \geq 0, \mu_4 \geq 0$ are suppressed in this section It was shown in Section 3 2 that (4 1) has a family, $\{\xi(\alpha)\}$, of equilibrium points in Q

Let $F_x(x, \alpha)$ be the Jacobian matrix of $F(x, \alpha)$ The equilibrium point $(\xi(\alpha), \alpha)$ of (4 1) is *linearly stable* if $\mathrm{Re}\,\lambda < 0$ for every eigenvalue λ of $F_x(\xi(\alpha), \alpha)$ Similarly, the equilibrium point $(\xi(\alpha), \alpha)$ is *linearly unstable* if $\mathrm{Re}\,\lambda > 0$ for some eigenvalue λ of $F_x(\xi(\alpha), \alpha)$ The relationship between linear stability of equilibrium points and their asymptotic stability is explored, for example, in [13, Ch III] or[15, Ch VI]

The case of an *exchange of linear stability* at an equilibrium point $(\xi(\alpha_0), \alpha_0)$ is generically covered by the Hopf Bifurcation Theorem The result presented here is not based on Hopf's original result [14], but on [7,8] The treatment in [15, Chs VII & VIII] is closely related (x, α) is a periodic solution of (4 1) with period T

if, and only if, (u, α) is a 2π-periodic solution of

$$u + G(u, \alpha) = 0,$$

where

$$\begin{aligned} u(t) &= x(tT/2\pi) - \xi(\alpha), \\ G(u\ \alpha) &= -\frac{T}{2\pi} F(u + \xi(\alpha), \alpha) \end{aligned}$$

It is this formulation of the problem which is used in [7,8] However the results here are stated for (4 1)

Let $x_0 = \xi(\alpha_0)$, and

$$L_0 = F_x(x_0, \alpha_0) \tag{4 2}$$

We make the following assumptions

A1 $i\omega$ is an algebraically simple eigenvalue of L_0, and $in\omega$ is not an eigenvalue for $n = 0, 2, 4, \quad ,$

A2 The *crossing condition*

$$\mathrm{Re}\,\lambda(\alpha_0) \neq 0,$$

holds, where λ is an eigenvalue of $F_x(\xi(\alpha), \alpha)$ and the dash implies differentiation with respect to α

Our second assumption ensures that the eigenvalues cross the imaginary axis transversally Figure 4 1 shows a possible path of the particular pair of eigenvalues that satisfy $\lambda(\alpha_0) = \pm i\omega$ If all other eigenvalues have strictly negative real parts then Figure 4 1 illustrates a loss of stability

Theorem 4 1 (The Hopf Bifurcation Theorem) *There are analytic functions $\varepsilon \mapsto T(\varepsilon)$, $\varepsilon \mapsto \alpha(\varepsilon)$ and $\varepsilon \mapsto x(\varepsilon)$, defined on $(-\varepsilon_0, \varepsilon_0)$, for some $\varepsilon_0 > 0$ These functions have the following properties*

(i) $t \mapsto x(\varepsilon)(t)$ has period $T(\varepsilon)$ and is a solution of (4 1),

(ii) $\alpha(0) = \alpha_0$, $x(0) = x_0$ and $T(0) = 2\pi/\omega$,

(iii) there is a positive number η such that if (x_1, α_1) is a solution of (4 1) of period T_1, and $|T_1 - 2\pi/\omega| < \eta$, $|\alpha_1 - \alpha_0| < \eta$, and $|x_1(t)| < \eta$ for $0 \leq t \leq T_1$, then there is an $\varepsilon \in (-\varepsilon_0, \varepsilon_0)$ and $\theta \in [0, 2\pi)$ such that $T_1 = T(\varepsilon)$, $\mu_1 = \mu(\varepsilon)$, and $x_1(t) = x(\varepsilon)(t + \theta)$,

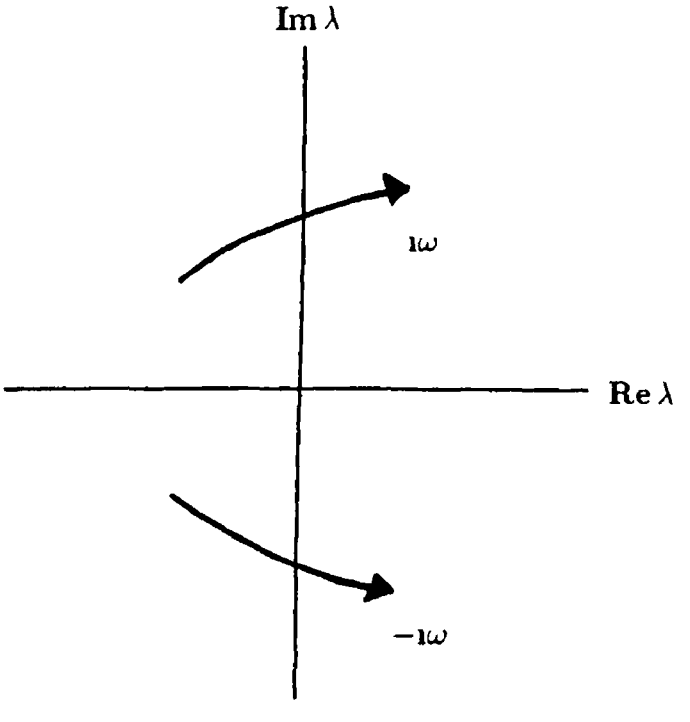

Figure 4 1 The transversal crossing of the imaginary axis for the pair of eigenvalues satisfying $\lambda(\alpha_0) = \pm i\omega$

(iv) the parameter ε can be chosen so that $\alpha(\varepsilon) = \alpha(-\varepsilon)$ and $T(\varepsilon) = T(-\varepsilon)$ for $0 < |\varepsilon| < \varepsilon_0$

If $\alpha \neq 0$, the analyticity of α requires that either $\alpha(\varepsilon) > \alpha_0$ or $\alpha(\varepsilon) < \alpha_0$ for all small $\varepsilon \neq 0$ These two cases are termed *supercritical* and *subcritical bifurcation* respectively

The linear mapping L_0 has a real eigenvalue ν, as well as $\pm i\omega$ We now determine the *linear stability* of the periodic orbits found in Theorem 4 1 under the assumption that $\nu < 0$ To do this, we review some material on linear equations with periodic coefficients For more details, see [13, Ch III] or [15, Ch VII]

Consider the linear equation

$$w = A(t)w, \tag{4 3}$$

where $A(t)$ $\mathbf{R}^3 \to \mathbf{R}^3$ has period T We denote by U the fundamental solution satisfying $U(0) = I$ and

$$U = A(t)U \tag{4 4}$$

Since $A(t)$ is T-periodic $U(t+T)$ is also a solution of (4 4) Therefore there exists a non-singular matrix C such that

$$U(t+T) = U(t)C \tag{4 5}$$

$C = U(T)$ is called the *monodromy matrix* The (possibly complex) eigenvalues of C are called *Floquet multipliers*, and κ is a *Floquet exponent* if $e^{\kappa T}$ is a Floquet multiplier κ is a Floquet exponent if, and only if, the equation

$$z = A(t)z - \kappa z \tag{4 6}$$

has a nontrivial T-periodic solution To see this consider a Floquet multiplier $e^{\kappa T}$ Then there is an eigenvector ψ of C such that

$$C\psi = e^{\kappa T}\psi \tag{4 7}$$

If $w(t) = U(t)\psi$, then w is a solution of (4 3) Moreover $z(t) = e^{-\kappa t}U(t)\psi$ solves (4 6) Since

$$\begin{aligned} z(t+T) &= e^{-\kappa(t+T)}U(t+T)\psi = e^{-\kappa t}e^{-\kappa T}U(t)U(T)\psi \\ &= e^{-\kappa t}U(t)\psi = z(t), \end{aligned}$$

z has period T Since z is continuous and periodic, it must be bounded Because $w(t) = e^{\kappa t}z(t)$, $w(t) \to 0$ exponentially as $t \to \infty$ if $\mathrm{Re}\,\kappa < 0$, and $w(t) \to \infty$ exponentially as $t \to \infty$ if $\mathrm{Re}\,\kappa > 0$ It is not hard to prove that $w = 0$ is a stable (asymptotically stable) solution of (4 3) if, and only if, every Floquet multiplier satisfies $\mathrm{Re}\,\kappa \leq 0$ ($\mathrm{Re}\,\kappa < 0$) If (4 3) has at least one multiplier satisfying $\mathrm{Re}\,\kappa > 0$, then $w = 0$ is unstable

We examine the solutions of the *variational equation*

$$w = A(t,\varepsilon)w, \tag{4 8}$$

where

$$A(t,\varepsilon) = F_x(x(\varepsilon)(t), \alpha(\varepsilon)) \tag{4 9}$$

Clearly $A(\ ,\varepsilon)$ has period $T(\varepsilon)$ The periodic solution $(x(\varepsilon), \alpha(\varepsilon))$ of (4 1) is said to be *linearly stable* if every Floquet multiplier of (4 8) satisfies $\mathrm{Re}\,\kappa \leq 0$ If (4 8) has at least one multiplier satisfying $\mathrm{Re}\,\kappa > 0$, then $(x(\varepsilon), \alpha(\varepsilon))$ is a *linearly unstable* solution of (4 1) For a result that relates the linear stability of $(x(\varepsilon), \alpha(\varepsilon))$ to *orbital stability with asymptotic phase*, see [13, Ch VI]

By differentiating (4 1), it is clear that $w = x$ is a nontrivial solution of (4 8) Thus 0 is a Floquet exponent of (4 8) for all $|\varepsilon| < \varepsilon_0$, $A(t,0) = L_0$ At $\varepsilon = 0$, the values of κ for which

$$z = L_0 z - \kappa z, \tag{4 10}$$

has nontrivial solutions are $\{\sigma(L_0) \pm in\omega \quad n = 0, 1, 2, \quad\}$ Thus $\kappa = 0$ is a double eigenvalue

Theorem 4 2 *Equation (4 8) has two Floquet exponents which approach* 0 *as* $\varepsilon \to 0$ *One is* 0 *and the other is* $\kappa(\varepsilon)$, *where* $\kappa \quad (-\varepsilon_1, \varepsilon_1) \to \mathbf{R}$ *is an analytic function and* $\kappa(0) = 0$ *Moreover there is a continuous function* $\chi \quad (-\varepsilon, \varepsilon) \to \mathbf{R}$ *such that*

$$\kappa(\varepsilon) = \varepsilon\alpha'(\varepsilon)\chi(\varepsilon), \tag{4 11}$$

and

$$\chi(0) = -Re\,\lambda'(0) \tag{4 12}$$

This result determines the sign of κ Equation (4 8) has the Floquet multipliers corresponding to exponents 0, $\kappa(\varepsilon)$ and another multiplier which must be in lefthand side of the complex plane Therefore the linear stability of the periodic solution $(x(\varepsilon), \alpha(\varepsilon))$ is determined by κ If $\mathrm{Re}\,\lambda'(0) < 0$ and $\varepsilon\alpha'(\varepsilon) < 0$, the periodic solution is linearly stable But if $\mathrm{Re}\,\lambda'(0) < 0$ and $\varepsilon\alpha'(\varepsilon) > 0$, the periodic solution is linearly unstable

4.2 An Example of the Application of the Hopf Bifurcation Theorem

As an illustration of the Hopf Bifurcation theorem consider the system

$$\begin{aligned} x_1 &= x_2 - x_1(x_1^2 + x_2^2 - \alpha), \\ x_2 &= -x_1 - x_2(x_1^2 + x_2^2 - \alpha), \end{aligned} \tag{4 13}$$

which may be written in the form

$$x = F(x, \alpha)$$

where $F \quad \mathbf{R}^2 \times \mathbf{R} \to \mathbf{R}^2$ This system has an equilibrium point at the origin and its linearisation at $(0,0)$ may be represented by

$$x = F_x(0, \alpha)x$$

where

$$F_x(0, \alpha) = \begin{pmatrix} \alpha & 1 \\ -1 & \alpha \end{pmatrix} \tag{4 14}$$

The eigenvalues of $F_x(0, \alpha)$ are $\lambda(\alpha) = \alpha \pm i$ When $\alpha = 0$, $F_x(0, \alpha)$ has purely imaginary eigenvalues and $\mathrm{Re}\,\lambda'(0) = 1$, thus the crossing condition holds Hence, by Theorem 4 1, there exists a periodic solution to (4 13) To explicitly see this,

we transform the equations to polar form by the change of variables $x_1 = r\cos\theta$ and $x_2 = r\sin\theta$, so that (4 13) becomes

$$\begin{aligned} \dot{r} &= r(\alpha - r^2), \\ \dot{\theta} &= -1 \end{aligned} \tag{4 15}$$

The general solution of (4 15) is

$$\begin{aligned} r(t) &= \begin{cases} \dfrac{r_0\alpha^{1/2}}{[r_0^2 + (\alpha - r_0^2)e^{-2\alpha t}]^{1/2}} & \text{for } \alpha \neq 0, \\ \dfrac{r_0}{[1 + 2tr_0]^{1/2}} & \text{for } \alpha = 0, \end{cases} \\ \theta(t) &= t - \theta_0, \end{aligned}$$

where $r_0 = r(0)$ and $\theta_0 = \theta(0)$ Hence, when $\alpha \neq 0$

$$\begin{aligned} x_1(t) &= \frac{r_0\alpha^{1/2}}{[r_0^2 + (\alpha - r_0^2)e^{-2\alpha t}]^{1/2}}\cos(\theta_0 - t), \\ x_2(t) &= \frac{r_0\alpha^{1/2}}{[r_0^2 + (\alpha - r_0^2)e^{-2\alpha t}]^{1/2}}\sin(\theta_0 - t), \end{aligned}$$

and when $\alpha = 0$

$$\begin{aligned} x_1(t) &= \frac{r_0}{[1 + 2tr_0]^{1/2}}\cos(\theta_0 - t), \\ x_2(t) &= \frac{r_0}{[1 + 2tr_0]^{1/2}}\sin(\theta_0 - t) \end{aligned}$$

We look at three separate cases

(i) When $\alpha < 0$ then $\dot{r} < 0$, $\dot{\theta} < 0$ and the equilibrium point is a stable focus Trajectories of (4 15) spiral inwards exponentially from the origin as $t \to \infty$ as shown in Figure 4 2

(ii) When $\alpha = 0$ then $\dot{r} < 0$, $\dot{\theta} < 0$ and trajectories spiral linearly towards the origin as $t \to \infty$, as shown in Figure 4 3

(iii) For $\alpha > 0$

$$\dot{r} \begin{cases} > 0 & \text{for } r_0 < \sqrt{\alpha} \\ = 0 & \text{for } r_0 = \sqrt{\alpha} \\ < 0 & \text{for } r_0 > \sqrt{\alpha} \end{cases} \tag{4 16}$$

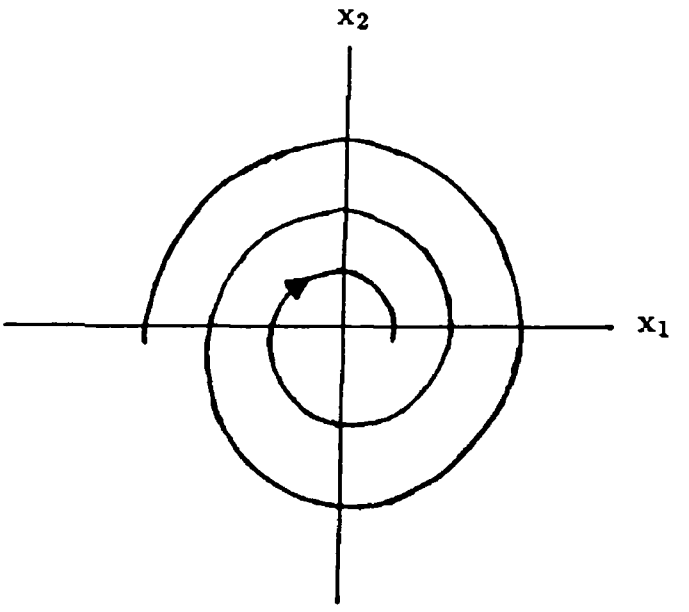

Figure 4 2 The phase portrait of (4) for $\alpha < 0$

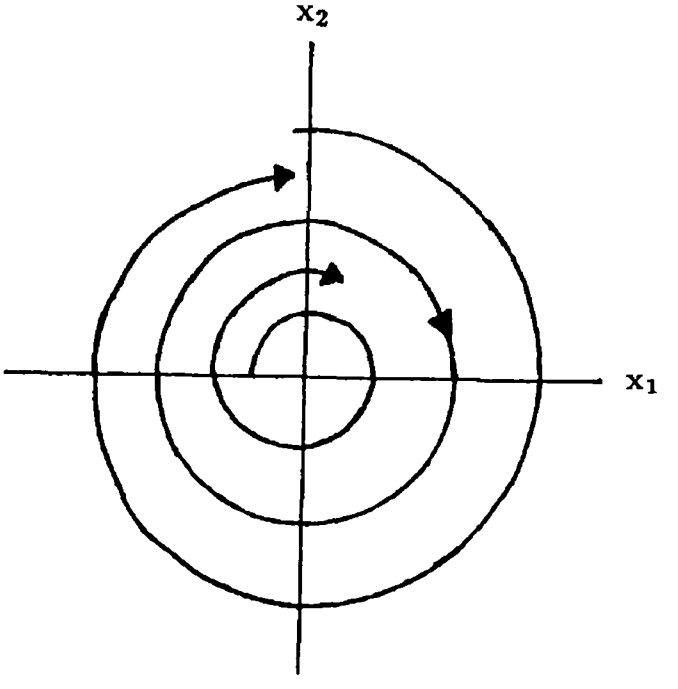

Figure 4 3 The phase portrait of (4) for $\alpha = 0$

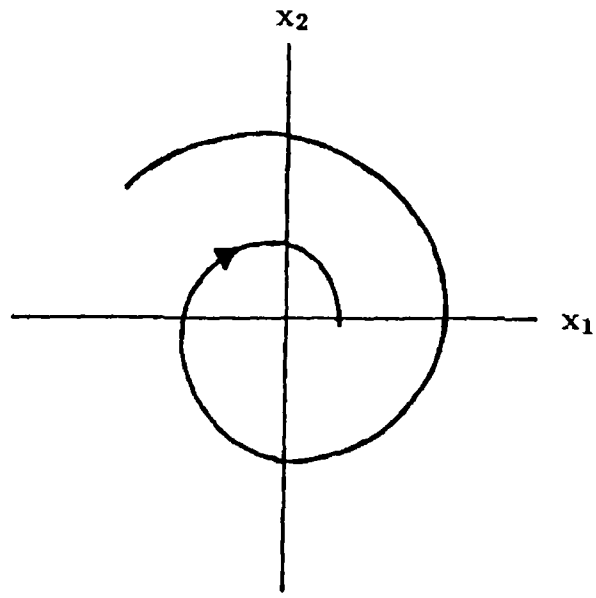

Figure 4 4 The phase portrait of system() for $\alpha < 0$

We can see that when $r_0 = \sqrt{\alpha}$ then

$$\begin{aligned} x_1 &= \sqrt{\alpha}\cos(\theta_0 - t), \\ x_2 &= \sqrt{\alpha}\sin(\theta_0 - t) \end{aligned}$$

This solution is the circle $x_1^2 + x_2^2 = \alpha$ and is periodic with period 2π Also, $r(t)$ approaches α as t approaches infinity Thus the origin is an unstable focus and all solutions which do not start at the origin tend towards the periodic solution $r = \sqrt{\alpha}$ as $t \to \infty$ as depicted in Figure 4 4

We can see that on passing through the critical value of $\alpha = 0$ the solutions undergo an exchange in linear stability and the phase portrait undergoes a qualitative change which results in the appearance of a limit cycle solution

The Hopf Bifurcation theorem is proved by Hale [13] using the Lyapunov-Schmidt procedure to reduce the problem to a 2 dimensional system Polar coordinates are then used, as in the illustration, to show the existence of a unique limit cycle solution

4.3 Hopf Bifurcation in the Full Explodator Model

The *Full Explodator Model* is the resulting system of equations on including all the limiting reactions in the model It has already been shown in section 3 2 that the full Explodator model has an equilibrium point $(\xi(\alpha), \alpha)$, defined by (3 4) The equilibrium point may be translated to the origin by performing the change of variable

$$y = x - \xi,$$

where

$$y = (y_1, y_2, y_3),$$

and

$$x = (x_1, x_2, x_3)$$

Under this change of variable (3 1) becomes

$$\begin{aligned} y_1 &= (1 - 3\mu_3 - 6\mu_1\xi_1 - \xi_2)y_1 - \xi_1 y_2 - 3\mu_1 y_1^2 - y_1 y_2, \\ y_2 &= -\xi_2 y_1 - (\xi_1 + \beta)y_2 + 3\alpha y_3 - y_1 y_2, \\ y_3 &= (\mu_3 + 2\mu_1\xi_1 + \xi_2)y_1 + \xi_1 y_2 - 2\alpha y_3 - \mu_1 y_1^2 + y_1 y_2, \end{aligned} \tag{4 17}$$

which may be written in the form

$$y = G(y, \alpha),$$

where $G \quad \mathbf{R}^3 \times \mathbf{R} \to \mathbf{R}^3$ The linearised stability of (4 17) is determined by the eigenvalues of the Jacobian matrix

$$G_y(0, \alpha) = \begin{pmatrix} 1 - 3\mu_3 - 6\mu_1\xi_1 - \xi_2 & -\xi_1 & 0 \\ -\xi_2 & -\xi_1 - \beta & 3\alpha \\ \mu_3 + 2\mu_1\xi_1 + \xi_2 & \xi_1 & -2\alpha, \end{pmatrix} \tag{4 18}$$

The eigenvalues are solutions of the cubic characteristic equation

$$\lambda^3 + a_2(\alpha)\lambda^2 + a_1(\alpha)\lambda + a_0(\alpha) = 0, \tag{4 19}$$

where

$$\begin{aligned} a_2(\alpha) &= \xi_2 + \xi_1(1 + 6\mu_1) + 3\mu_3 + \beta - 1 + 2\alpha, \\ a_1(\alpha) &= (2\alpha + \beta + \xi_1)(\xi_2 + 6\mu_1\xi_1 + 3\mu_3 - 1) - \alpha\xi_1 + 2\alpha\beta - \xi_1\xi_2, \\ a_0(\alpha) &= -2\alpha\beta(1 - 6\mu_1\xi_1 - 3\mu_3 - \xi_2) + \alpha\xi_1 \end{aligned}$$

By using the definitions of ξ_2 and χ given by (3 4) and (3 5) these equations may be reduced to the following

$$a_2(\alpha) = 2\alpha + C_0, \tag{4 20}$$
$$a_1(\alpha) = \alpha C_1 + C_2, \tag{4 21}$$
$$a_0(\alpha) = 2\alpha\chi \tag{4 22}$$

where

$$\begin{aligned} C_0 &= \beta + \xi_1 + \chi/\beta - \xi_1/2\beta, \\ C_1 &= 2(\chi/\beta - \xi_1/2\beta) + 2\beta - \xi_1, \\ C_2 &= (\beta + \xi_1)(\chi/\beta - \xi_1/2\beta) - \xi_1\xi_2 \end{aligned}$$

It is important to note that χ, C_0, C_1 and C_2 are independent of α In order to simplify further calculations it is necessary to look at the signs of the above expressions To do this we examine the signs of $\chi - \xi_1/2$ and $2\beta - \xi_1$

(i) From equation (3 5) we can see that

$$\chi \geq -\beta(1 - 3\mu_3) + \mu_2 + \mu_4, \tag{4 23}$$

therefore

$$\begin{aligned} \chi - \xi/2 &= \chi - \frac{\beta(1 - 3\mu_3) - \mu_2 - \mu_4 + \chi}{2(1 + 6\beta\mu_1)} \\ &\geq \chi - \frac{\chi}{1 + 6\beta\mu_1}, \\ &\geq 0 \end{aligned} \tag{4 24}$$

(ii) By (3 4) we see that ξ_1 satisfies the quadratic

$$(1 + 6\beta\mu_1)\xi_1^2 + 2\xi_1(3\beta\mu_3 - \beta + \mu_2 + \mu_4) = 4\beta\mu_2$$

Since $\xi_1 > 0$, $\mu_1 \geq 0$, $\mu_3 \geq 0$ and $\mu_4 \geq 0$ then

$$\xi_1^2 + 2\xi_1(\mu_2 - \beta) \leq 4\beta\mu_2 \tag{4 25}$$

By completing the square of the left hand side of (4 25) it is easy to see that

$$(\xi_1 + \mu_2 - \beta)^2 \leq (\mu_2 + \beta)^2,$$

and

$$\xi_1 + \mu_2 - \beta \leq \mu_2 + \beta$$

Therefore

$$\xi_1 \leq 2\beta \tag{4 26}$$

From these results it follows that

$$C_0 > 0, C_1 \geq 0 \tag{4 27}$$

A close examination of the inequalities shows that $C_1 = 0$ if and only if $\mu_i = 0$ for $i = 1, 2, 3, 4$ We now use the Routh-Hurwitz criterion to determine if solutions of (4 17) satisfy $\mathrm{Re}(\lambda) < 0$

Lemma 4 3 (The Routh-Hurwitz Criterion) *If*

$$P(z) = z^n + a_{n-1}z^{n-1} + \quad + a_1 z + a_0 \tag{4 28}$$

is a polynomial of real coefficients, let $D_1, D_2, \quad, D_n$ *denote the following determinants,*

$$D_1 = a_{n-1},$$

$$D_k = \begin{vmatrix} a_{n-1} & a_{n-3} & a_{n-5} & a_{n-(2k-1)} \\ 1 & a_{n-2} & a_{n-4} & a_{n-(2k-2)} \\ 0 & a_{n-1} & a_{n-3} & a_{n-(2k-3)} \\ 0 & 1 & a_{n-2} & a_{n-(2k-4)} \\ & & & \\ 0 & 0 & 0 & a_{(n-k)} \end{vmatrix}$$

where $k = 2, \quad, n$ *and* $a_{n-j} = 0$ *for* $j > n$ *The necessary and sufficient condition for the roots of*

$$P(z) = 0 \tag{4 29}$$

to lie in the half plane $Re\, z < 0$, *is that* $D_k > 0$ *for* $k = 1, \quad, n$

Thus the Routh-Hurwitz criteria for the linear stability of the solutions of (4 17) are

D1

$$a_2 > 0, \tag{4 30}$$

D2

$$\begin{vmatrix} a_2 & a_0 \\ 1 & a_1 \end{vmatrix} = a_2 a_1 - a_0 > 0 \tag{4 31}$$

and

D3

$$\begin{vmatrix} a_2 & a_0 & 0 \\ 1 & a_1 & 0 \\ 0 & a_2 & a_0 \end{vmatrix} = a_0(a_2a_1 - a_0) > 0$$

From (4 24) and (4 26), D1 always holds Also $\chi > 0$ implies that $a_0 > 0$ Hence $(\xi(\alpha), \alpha)$ is linearly stable if and only if

$$a_2a_1 - a_0 > 0 \tag{4 32}$$

Theorem 4 4 *Suppose that (3 6) holds Then*

(i) If $C_1 = 0$, $(\xi(\alpha), \alpha)$ is unstable for all $\alpha > 0$

(ii) If $C_1 > 0$, $C_2 \geq 0$ and $C_0C_1 - 2\chi \geq 0$, $(\xi(\alpha), \alpha)$ is stable for all $\alpha > 0$

(iii) If $C_1 > 0$ and $C_2 < 0$ there is a unique positive value α_0 such that

$$(\xi(\alpha), \alpha) \text{ is unstable for } 0 < \alpha < \alpha_0$$

and

$$(\xi(\alpha), \alpha) \text{ is stable for } \alpha_0 < \alpha$$

Proof A necessary and sufficient condition for $(\xi(\alpha), \alpha)$ to be linearly stable is that condition 4 32 holds Thus we examine the quadratic

$$q(\alpha) = a_2(\alpha)a_1(\alpha) - a_0(\alpha) \tag{4 33}$$

$$= 2\alpha^2C_1 + \alpha(C_0C_1 + 2C_2 - 2\chi) + C_0C_2 \tag{4 34}$$

Assume first that $C_1 = 0$ Then, as has been noted, $\mu_i = 0$ for $i = 1, 2, 3, 4$ An easy calculation shows that

$$C_0 = 3\beta, C_2 = -2\beta, \chi = \beta \tag{4 35}$$

Thus q becomes

$$q(\alpha) = -2\beta(3\beta + 2\alpha), \tag{4 36}$$

and $q(\alpha) < 0$ for $\alpha > 0$ Thus the equilibrium point is unstable

Suppose now that the hypotheses of (ii) hold Then q attains its minimum at a non positive value of α Since $C_0C_2 > 0$, it either has complex roots, two negative roots or a pair of real roots of opposite sign In every case $q(\alpha) > 0$ for all $\alpha > 0$

Assume now that the hypotheses (iii) hold Since $C_2C_0 < 0$, q has a unique positive root α_0 $(\xi(\alpha), \alpha)$ undergoes an exchange of linear stability at $\alpha = \alpha_0$ α_0 is given by

$$\alpha_0 = \frac{-C \pm \sqrt{C^2 - 8C_0C_1C_2}}{4C_1} \tag{4 37}$$

and $C = C_0C_1 + 2C_2 - 2\chi$ The theorem then follows from the graph of q □

Remark Theorem 4 4 is not exhaustive since we cannot prove that $C_0C_1 - 2\chi \geq 0$ if $C_1 > 0$ and $C_2 \geq 0$ However if $\mu_1 = \mu_2 = 0$ then $C_2 < 0$ Moreover if either (i) $\mu_1 > 0$ and $\mu_i = 0$ for $i = 2, 3, 4$ or (ii) $\mu_2 > 0$ and $\mu_i = 0$ for $i = 1, 3, 4$, it can be shown that $C_0C_1 - 2\chi \geq 0$ We now see if the crossing condition holds for the full Explodator When $\alpha = \alpha_0$ the characteristic equation may be written as

$$\lambda^3(\alpha_0) + \lambda^2(\alpha_0)a_2(\alpha_0) + \lambda(\alpha_0)a_1(\alpha_0) + a_1(\alpha_0)a_2(\alpha_0) = 0,$$

which, when factorised, becomes

$$(\lambda(\alpha_0) + a_2(\alpha_0))(\lambda^2(\alpha_0) + a_1(\alpha_0)) = 0$$

Thus there is a pair of complex conjugate eigenvalues which satisfy

$$\lambda(\alpha_0) = \pm i\sqrt{a_1(\alpha_0)}$$

and a third eigenvalue

$$\nu(\alpha_0) = -a_2(\alpha_0) < 0$$

and assumption (A1) required to apply the Hopf Bifurcation Theorem holds We now wish to verify that the crossing condition (A2) holds To find $\lambda'(\alpha_0)$ we return to look at the characteristic equation (4 19) Since the eigenvalues are simple, and hence differentiable, the derivative of each eigenvalue satisfies

$$\begin{aligned} 3\lambda^2(\alpha)\lambda'(\alpha) \;+\; & 2a_2(\alpha)\lambda'(\alpha)\lambda(\alpha) + a_2'(\alpha)\lambda(\alpha) \\ +\; & a_1(\alpha)\lambda'(\alpha) + a_1'(\alpha)\lambda(\alpha) + a_0'(\alpha) = 0, \end{aligned} \tag{4 38}$$

where the dash implies differentiation with respect to α Rearranging (4 38) yields

$$\lambda'(\alpha) = \frac{-\lambda(\alpha)\{a_2'(\alpha)\lambda(\alpha) + a_1'(\alpha)\} - a_0'(\alpha)}{3\lambda^2(\alpha) + 2a_2(\alpha)\lambda(\alpha) + a_1(\alpha)}$$

Evaluating the above expression at $\alpha = \alpha_0$ with $\lambda(\alpha_0) = i\sqrt{a_1(\alpha_0)}$ we find that

$$\lambda'(\alpha_0) = \frac{a_2'(\alpha_0)a_1(\alpha_0) - a_0'(\alpha_0) - i(\sqrt{a_1(\alpha_0)}a_1'(\alpha_0))}{-2a_1(\alpha_0) + i(2a_2(\alpha_0)\sqrt{a_1(\alpha_0)})}$$

and the crossing condition is determined by

$$\mathrm{Re}\,\lambda'(\alpha_0) = \frac{a_2'(\alpha_0)a_1(\alpha_0) + a_2(\alpha_0)a_1'(\alpha_0) - a_0'(\alpha_0)}{a_1(\alpha_0) + a_2^2(\alpha_0)}$$

Let

$$A(\alpha) = a_2'(\alpha)a_1(\alpha) + a_2(\alpha)a_1'(\alpha) - a_0'(\alpha),$$

then the crossing condition (A2) is satisfied if $A(\alpha_0) \neq 0$ From equations (4 20), (4 21) and (4 22) we find that

$$A(\alpha) = 4\alpha C_1 + 2C_2 + C_1C_0 - 2\chi$$

Since α_0 satisfies $q(\alpha_0) = 0$ then

$$\begin{aligned} A(\alpha_0) &= \frac{2\alpha_0^2 C_1 + \alpha_0(2C_2 + C_1C_0 - 2\chi)}{\alpha_0} + 2C_1\alpha_0 \\ &= -\frac{C_0C_2}{\alpha_0} + 2C_1\alpha_0 \end{aligned}$$

But we have already shown that $C_1 > 0$ and $C_0 \geq 0$, thus the crossing condition holds when $C_2 < 0$

Theorem 4 5 *The system of equations (4 17) has a family of periodic solutions when $C_2 < 0$ and $C_1 > 0$*

This result follows directly from theorem 4 4, the result above and the Hopf Bifurcation Theorem

4.4 Numerical Results

AUTO is an integrated collection of FORTRAN routines for continuation and bifurcation problems in ordinary differential equations The primary purpose of the package is the computation of branches of stable or unstable periodic solutions of

$$u(t) = f(u(t), \lambda),$$

where $u, f \in \mathbf{R}^n$ and λ denotes a free parameter The package also contains algorithms for computing steady state solutions and algorithms for the accurate determination of hopf bifurcations

The determination of branches of steady state solutions involves solving the algebraic system

$$f(u, \lambda) = 0$$

Thus the package contains continuation algorithms for general algebraic systems In addition there are a number of related continuations that can be useful in the analysis of (4 4) These include the computation of curves of limit points and curves of hopf bifurcation points For such computations λ will have two components

AUTO also contains an interactive graphics program PLAUT, which can produce bifurcation diagrams, to show the stability properties of the solutions, and, two and three dimensional plots of the periodic solutions found To illustrate bifurcation behaviour graphically, PLAUT uses symbols that distinguish between stable and unstable solutions A heavy continuous curve represents stable stationary solutions and unstable stationary solutions are indicated by dashed curves An open circle indicates an unstable periodic solution, a solid circle a stable solution These branches are continuous, the gaps between the dots do not indicate a break in the solution For every parameter in the corresponding range there is a periodic orbit A solid square marks a hopf bifurcation point Locally stable periodic solutions encircle unstable stationary solutions, thus the direction of the periodic solutions emanating from a hopf point is related to the stability properties of the solutions

In the bifurcation diagrams a quantity called the norm is used When dealing with stationary solutions of (4 4) the norm is simply the vector $l_2$norm, i e , for $u = (u_1, \quad , u_n)$ we let

$$\| u \| = \{\sum_{i=1}^{n} u_i^2\}^{1/2},$$

while for periodic solutions

$$\| u \| = (\frac{1}{T})^{1/2}\{\int_0^T \sum_{i=1}^{n} u_i^2(t)\}^{1/2}$$

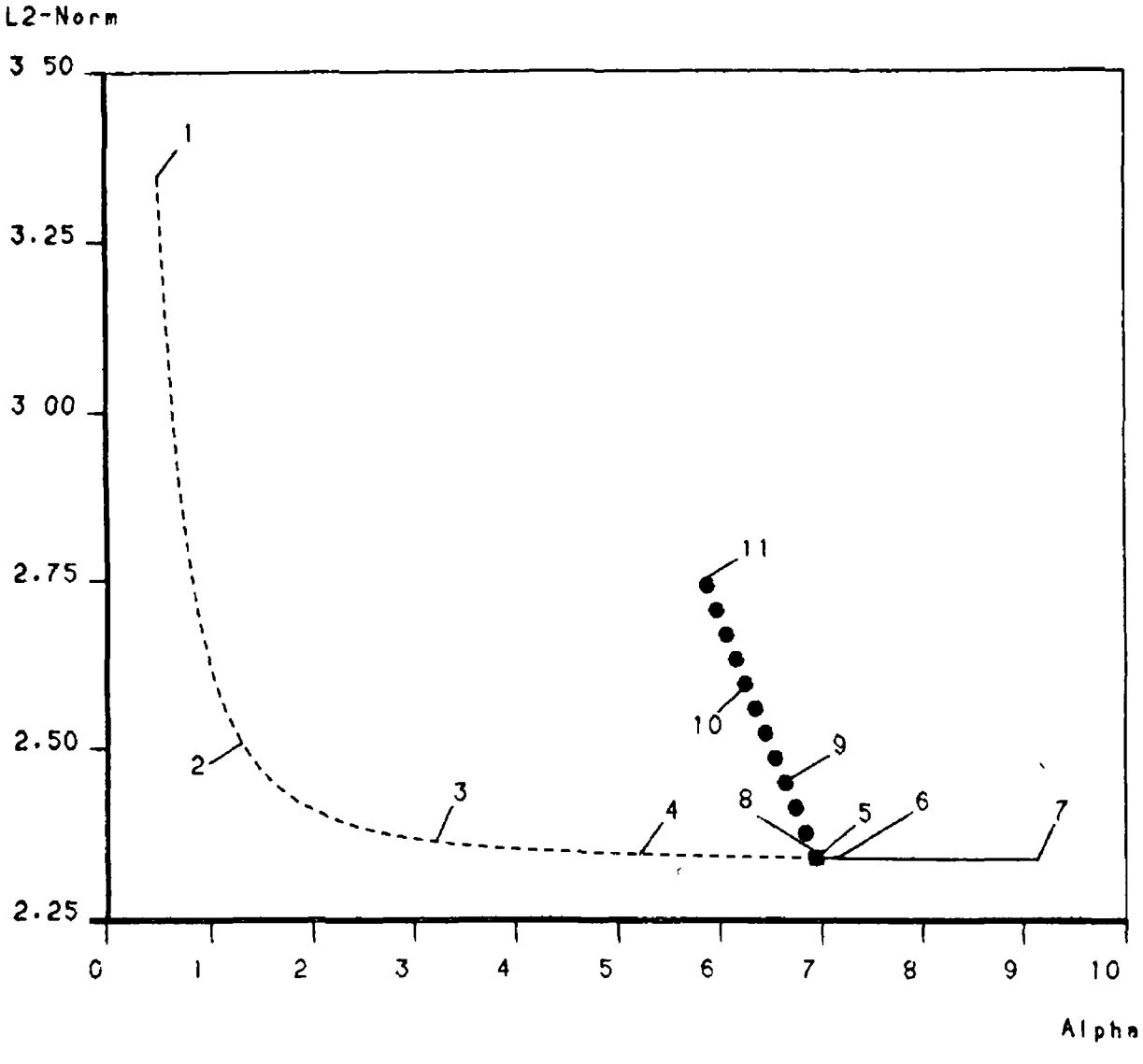

Figure 4 5 Bifurcation diagram for the full Explodator model with $\mu_1, \mu_3, \mu_4 = 0$ α is treated as the bifurcation parameter, with an initial value of $\alpha = 0\ 5$ The iteration is started with $\beta = 1\ 0$, $\mu_2 = 0\ 2$ and $\xi = (2\ 0, 1\ 2, 1\ 4)$ This system has a single branch of steady state solutions, with one Hopf Bifurcation point at $\alpha = 6\ 94$ This agrees with the positive value of α_0 found by using (4 35) There is a branch of stable periodic orbits emanating from the Hopf point, with an initial period $T = 6\ 29$ Plotting information was obtained for orbits at the marked points 8, 9, 10 and 11

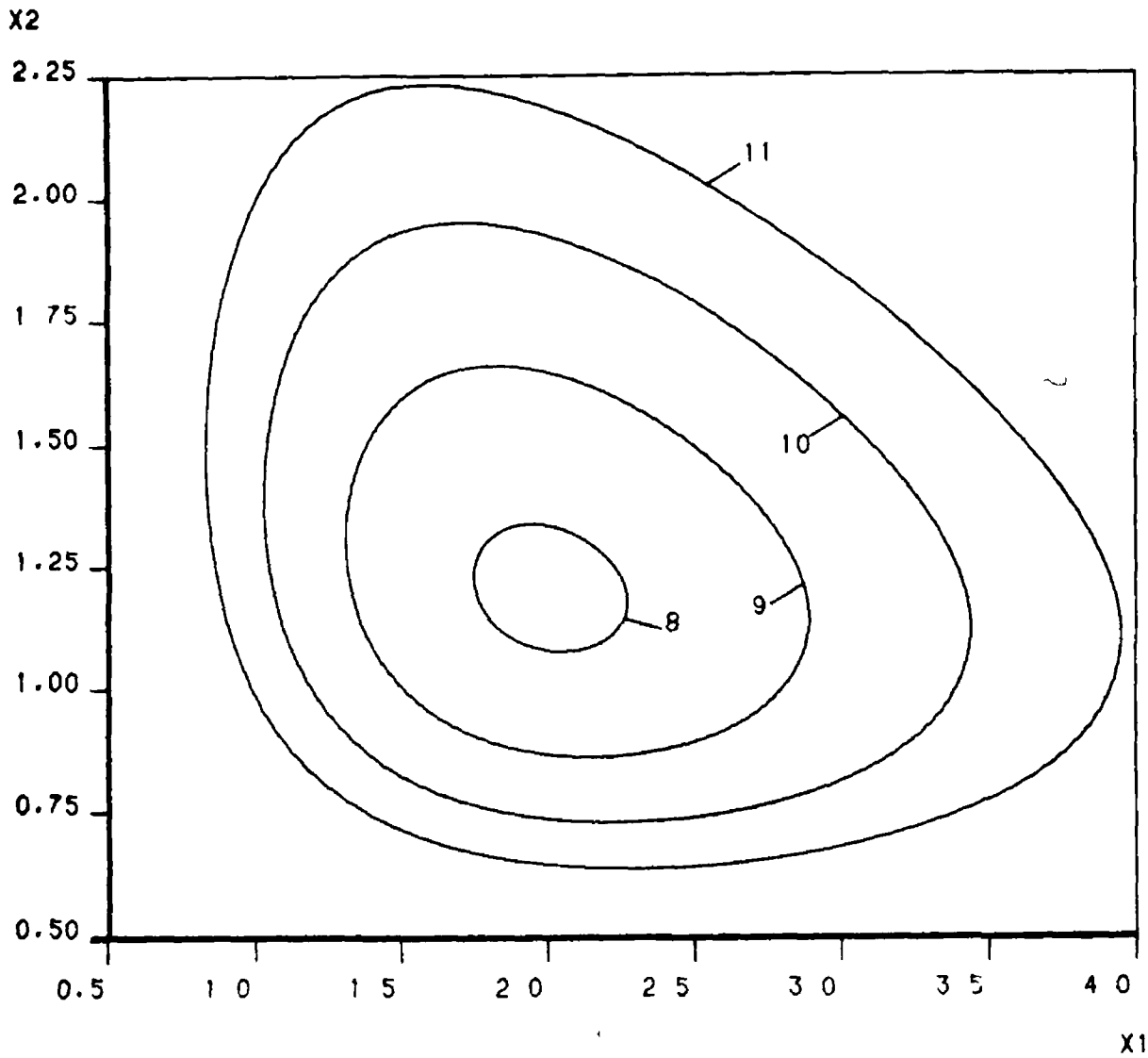

Figure 4 6 A 2-dimensional plot of the periodic orbits marked in figure 4 5 The parameter values and periods for these orbits are

orbit 8, $\alpha = 6\ 90$, $T = 6\ 37$,
orbit 9, $\alpha = 6\ 64$, $T = 6\ 43$,
orbit 10, $\alpha = 6\ 26$, $T = 6\ 52$,
orbit 11, $\alpha = 5\ 85$, $T = 6\ 63$

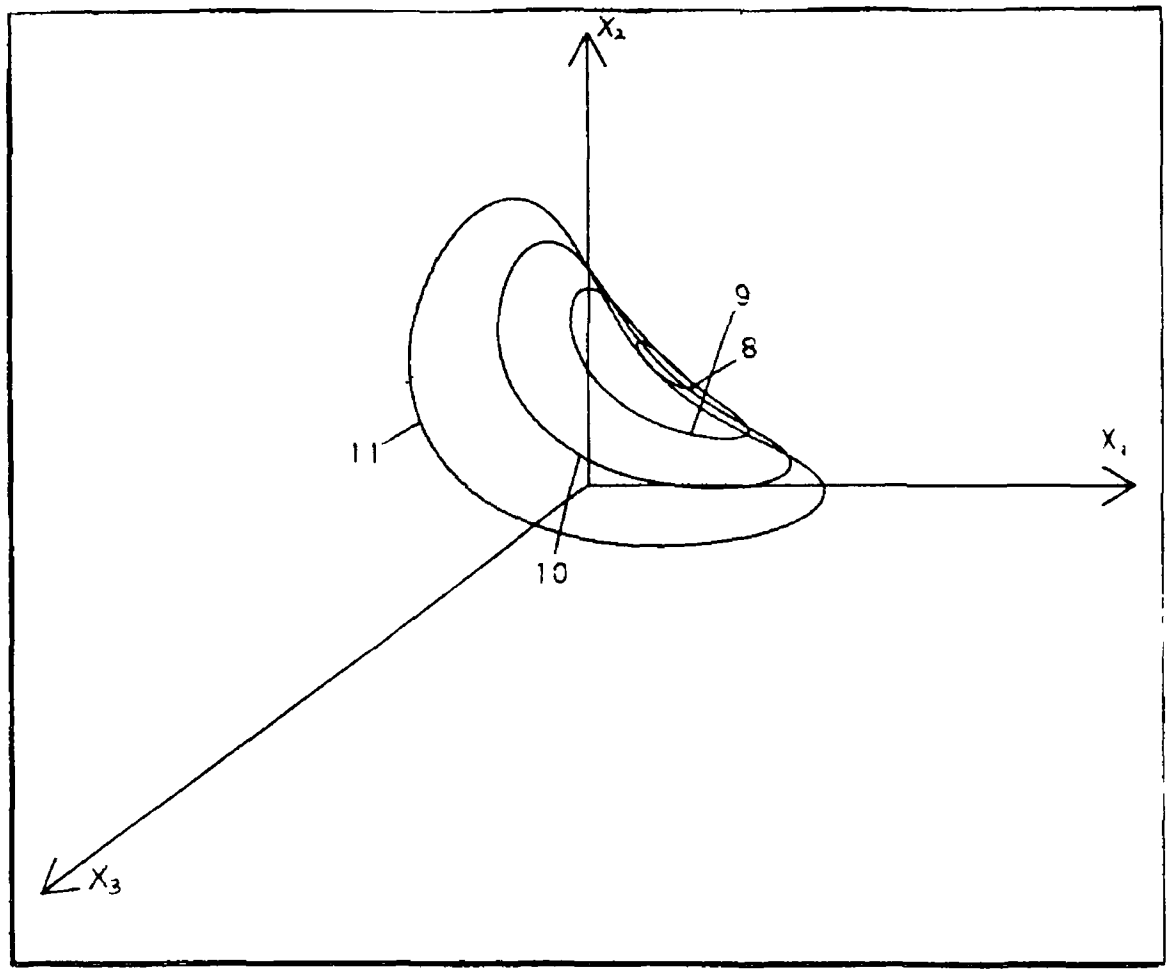

Figure 4 7 A 3-dimensional plot of the orbits marked in figure 4 5, with the projections on the planes marked by dotted lines A plot of x vs time obtained from orbit 11 may be seen in figure 2 2 The numerical solution is therefore consistent with the modelling assumptions made in Chapter 2 A local centre manifold can be detected in the vicinity of the equilibrium point

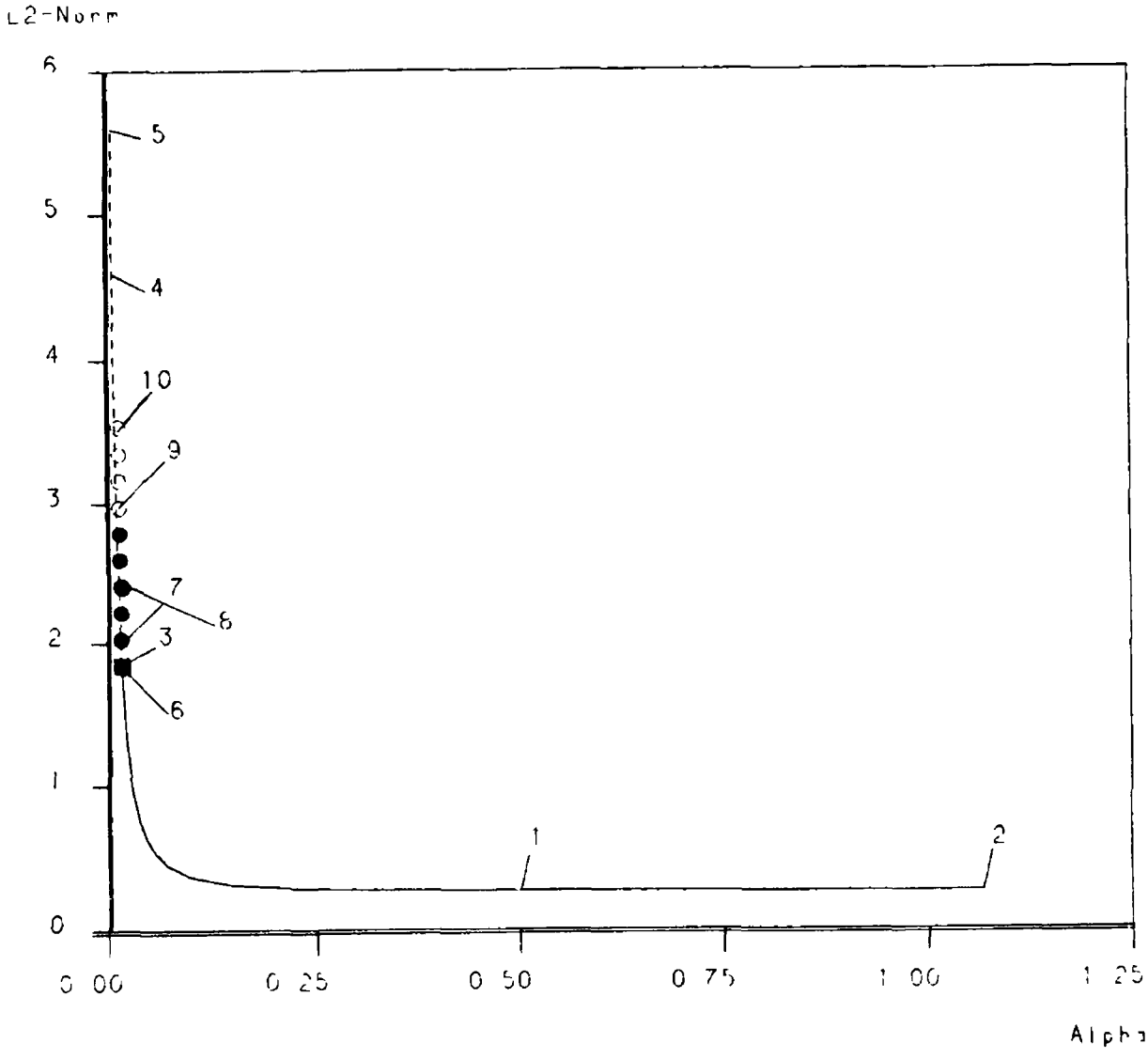

Figure 4 8 Bifurcation diagram for the full Explodator model with μ_1, $\mu_2 = 0$ α is treated as the bifurcation parameter, with an initial value of $\alpha = 0\ 5$ The iteration is started with $\beta = 1\ 0$, $\mu_3 = 0\ 25$, $\mu_4 = 0\ 2$ and $\xi = (0\ 1, 0\ 25, 0\ 05)$ This system has a single branch of steady state solutions, with one Hopf Bifurcation point at $\alpha = 1\ 38 \times 10^{-2}$ This agrees with the positive value of α_0 found by using (4 35) There is a branch of stable periodic orbits emanating from the Hopf point, with an initial period $T = 1\ 8 \times 10^2$ These orbits are initially stable Plotting information was obtained for orbits at the marked points 6, 7, 8, 9 and 10

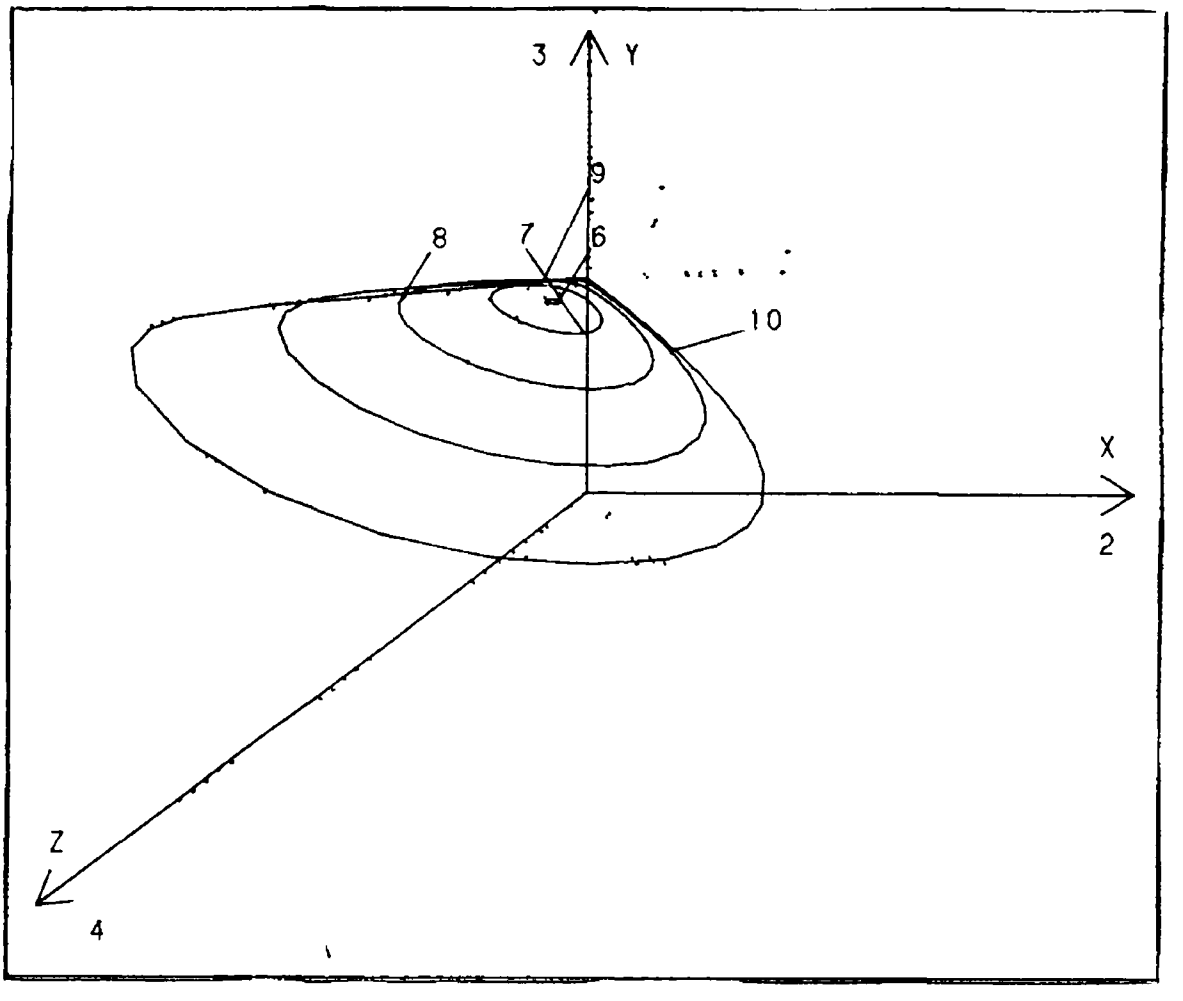

Figure 4 9 A 3-dimensional plot of the orbits marked in figure 4 8, with the projections on the planes marked by dotted lines The parameter values and periods for these orbits are A local centre manifold can be detected in the vicinity of the equilibrium point

The parameter values and periods for these orbits are

$$\begin{aligned}
\text{orbit 6,}\quad & \alpha = 1\,38 \times 10^{-2},\quad T = 1\,80 \times 10^{2},\\
\text{orbit 7,}\quad & \alpha = 1\,35 \times 10^{-2},\quad T = 1\,91 \times 10^{2},\\
\text{orbit 8,}\quad & \alpha = 1\,28 \times 10^{-2},\quad T = 2\,27 \times 10^{2},\\
\text{orbit 9,}\quad & \alpha = 1\,20 \times 10^{-2},\quad T = 2\,87 \times 10^{2},\\
\text{orbit 10,}\quad & \alpha = 1\,15 \times 10^{-2},\quad T = 3\,71 \times 10^{2},
\end{aligned}$$

Chapter 5

Centre Manifolds

The centre manifold theorem often provides us with a way of reducing the dimension of a system under consideration, and gives information regarding stability The method involves restricting attention to an invariant manifold (surface) to which all solutions in a neighbourhood of the equilibrium point are attracted exponentially In this chapter we will reduce the system of equations (3 1) to a 2-dimensional system

5.1 Invariant Manifolds and the Centre Manifold Theorem

Consider the compact form of equation (4 17)

$$y = G(y) \tag{5 1}$$

where $G \quad \mathbf{R}^{n+m} \to \mathbf{R}^{n+m}$ A set $S \subset \mathbf{R}^{n+m}$ is said to be a *local invariant manifold* for (5 1) if, for $y_0 \in S$, the solution $y(t)$ of (5 1) with $y(0) = y_0$ is in S for $|t| < T$, where $T > 0$ If we can always choose $T = \infty$, then we say that S is a *global invariant manifold*

Consider the system

$$\begin{aligned} x_1 &= Ax_1 + f(x_1, x_2), \\ x_2 &= Bx_2 + g(x_1, x_2), \end{aligned} \tag{5 2}$$

where $x_1 \in \mathbf{R}^n$, $x_2 \in \mathbf{R}^m$, f, $g \in C^2$ and A and B are constant matrices We make the following assumptions

C1 All the eigenvalues of A have zero real parts,

C2 All the eigenvalues of B have negative real parts

The case when the eigenvalues of B have nonzero real parts is covered by [5 ,16,17] [1] The situation examined here is that looked at by Carr [4]

Consider the linearised system

$$\begin{aligned} x_1 &= Ax_1, \\ x_2 &= Bx_2 \end{aligned} \tag{5 3}$$

Under assumption $C1$, the component, x_2 of the solution which corresponds to those eigenvalues with negative real parts will approach zero as t tends to infinity Hence the solutions (x_1, x_2) of (5 3) will approach the centre eigenspace [The centre eigenspace is the space spanned by the eigenvectors corresponding to those eigenvalues with zero real part] The centre manifold theorem tells us that this behaviour extends to the full non-linear system

Theorem 5 1 (Centre Manifold Theorem) *[4,16,17]*

Assume that $C1$, $C2$ hold and $f(x_1, x_2)$, $g(x_1, x_2)$ satisfy

$$\begin{aligned} f(0,0) &= 0 = g(0,0), \\ f'(0,0) &= 0 = g'(0,0), \end{aligned}$$

then there exists a C^2 function defined on $\{x \in \mathbf{R}^n \quad |x| < \delta\}$, such that

(i) $h(0) = 0, h'(0) = 0,$

(ii) the set

$$M_c = \{(x_1, x_2) | x_2 = h(x_1), x_1 \in \mathbf{R}^n\}$$

is an invariant manifold in $\mathbf{R}^{n+m}$ under the flow of (5 2),

The theorem tells us that there exists a surface (a Local Centre Manifold) which at the equilibrium point is tangent to the subspace spanned by the eigenvectors corresponding to those eigenvalues with zero real part The surface need only exist near the equilibrium point $(0,0)$ All solutions which start sufficiently close to the equilibrium point will tend asymptotically to a centre manifold The flow on the centre manifold is governed by the n-dimensional system

$$u = Au + f(u, h(u)) \tag{5 4}$$

Theorem 5 2 *[4]*

[1] We assume that no eigenvalues have positive real parts since in such a case the centre manifold will not be attractive as $t \to \infty$

(i) *Suppose that the zero solution of (5 4) is stable (asymptotically stable)(unstable) Then the zero solution of (5 2) is stable (asymptotically stable)(unstable)*

(ii) *Suppose that the zero solution of (5 4) is stable Let $(x_1(t), x_2(t))$ be a solution of (5 2) with $(x_1(0), x_2(0))$ sufficiently small Then there exists a solution $u(t)$ of (5 4) such that as $t \to \infty$*

$$\begin{aligned} x_1(t) &= u(t) + O(e^{-\gamma t}), \\ x_2(t) &= h(u(t)) + O(e^{-\gamma t}), \end{aligned}$$

where $\gamma > 0$ is a constant

5.2 Finding the Centre Manifold

We now discuss how to reduce a system to its centre manifold In order to obtain an approximate expression for the centre manifold we write x_2 as a function of x_1, and expand in a power series For every $\phi \quad \mathbf{R}^n \to \mathbf{R}^m$ where $\phi \in C^1$ with $\phi(0) = 0$ and $\phi'(0) = 0$, we define

$$(m\phi)(x_1) = D_x\phi(x_1)[Ax_1 + f(x_1, \phi(x_1))] - B\phi(x_1) - g(x_1, \phi(x_1))$$

From the second equation of (5 2) it is easy to see that

$$(mh)(x_1) = 0$$

Theorem 5 3 *[4]*
If

$$(m\phi)(x_1) = O(|x|^r) \text{ as } x_1 \to 0, \tag{5 5}$$

where $r > 1$, then

$$|h(x_1) - \phi(x_1)| = O(|x|^r) \text{ as } x_1 \to 0$$

We will now illustate this method by looking at the Limited Exploder Model

We look at the system of equations (3 1) and let $\mu_1 = \mu_3 = \mu_4 = 0$ Thus, $\xi = 2\beta$, $\xi_2 = 1 + \eta$ and

$$\begin{aligned} y_1 &= -\eta y_1 - 2\beta y_2 - y_1 y_2, \\ y_2 &= -(1+\eta) y_1 - 3\beta y_2 + 3\alpha y_3 - y_1 y_2, \\ y_3 &= (1+\eta) y_1 + 2\beta y_2 - 2\alpha y_3 + y_1 y_2, \end{aligned} \tag{5 6}$$

where $\eta = \mu_2/\beta$ The linearised form of (5 6) has eigenvalues whose real parts depend on the value of α However, we can write (5 6) in the equivalent suspended form

$$\begin{aligned} y_1 &= -\eta y_1 - 2\beta y_2 - y_1 y_2, \\ y_2 &= -(1+\eta) y_1 - 3\beta y_2 + 3\alpha y_3 - y_1 y_2, \\ y_3 &= (1+\eta) y_1 + 2\beta y_2 - 2\alpha y_3 + y_1 y_2, \\ \beta &= 0, \\ \eta &= 0 \end{aligned} \tag{5 7}$$

For this system, the terms ηy_1 and βy_2 are considered non-linear and the linearised form of (5 7) has as eigenvalues -2α, with multiplicity 1, and 0, with multiplicity 4

We now write (5 7) in the matrix form

$$y = My + N(y)$$

where $y = (y_1, y_2, y_3, \beta, \eta)$, M is a constant matrix and $N(y)$ contains all the terms quadratic in y We may write (5 2) in the form required by Theorem 5 1 by performing the change of variables $v = P^{-1}y$, where P is the transformation matrix corresponding to the eigenvalues found above Under this transformation (5 2) becomes

$$\begin{aligned} v &= Av + f(v, v_5), \\ v_5 &= -2\alpha v_5 + g(v, v_5), \end{aligned} \tag{5 8}$$

where $v = (v_1, v_2, v_3, v_4)$,

$$A = \begin{bmatrix} 0 & 0 & 0 & 0 \\ a & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix},$$

$$f(v, v_5) = \begin{bmatrix} -(v_2 - 3v_5)(v_1 + v_3/\alpha) \\ 3(v_2 - 3v_5)(v_1 + v_3/\alpha)/2 + v_1(\alpha + 3/2)v_4 + \alpha(v_2 - 3v_5) \\ 0 \\ 0 \end{bmatrix},$$

$$g(v, v_5) = (2\alpha + 1)[\alpha v_1 v_4 + (\alpha v_1 + v_3)(v_2 - 3\phi)]/2\alpha,$$

and $v_3 = \beta$, $v_4 = \eta$ By Theorem 5 1, the system (5 8) has a 4-dimensional centre manifold

$$v_5 = h(v_1, v_2, v_3, v_4)$$

Consider the power series approximation for h,

$$\begin{aligned}\phi(v,v_5) &= a_1v_1^2 + a_2v_1v_2 + a_3v_2^2 + a_4v_2v_3 + a_5v_3^2 \\ &+ a_6v_1v_3 + a_7v_4^2 + a_8v_1v_4 + a_9v_2v_4 + a_{10}v_3v_4 \qquad (5\ 9)\end{aligned}$$

From equations (5 5) and (5 8) we can see that

$$\begin{aligned}(m\phi)(v) &= \frac{\partial\phi}{\partial v_1}[-(v_2-3\phi)(v_1+v_3/\alpha)] \\ &+ \frac{\partial\phi}{\partial v_2}[3(v_2-3\phi)(v_1+v_3/\alpha)/2 + v_1(\alpha+3/2)v_4 + \alpha(v_2-3\phi)] \\ &+ 2\alpha\phi - (2\alpha+1)[\alpha v_1v_4 + (\alpha v_1+v_3)(v_2-3\phi)]/2\alpha\end{aligned}$$

Substituting (5 7) into this expression and neglecting cubic and higher order terms gives

$$\begin{aligned}(m\phi)(v) &= \alpha(a_2v_1^2 + 2a_3v_1v_2 + a_4v_1v_3 + a_9v_1v_4) \\ &= 2\alpha\phi - (2\alpha+1)(\alpha v_1v_4 + \alpha v_1, v_2 + v_2v_3)/2\alpha\end{aligned}$$

Equating the coefficients to zero, solving the ten resulting equations for a_i, $i = 1,10$, and substituting these values in (5 9) yields

$$\phi(v) = (\frac{1+2\alpha}{4\alpha})(-\frac{v_1^2}{2} + v_1v_2 + \frac{v_2v_3}{\alpha} - \frac{v_1v_3}{2\alpha} + v_1v_4)$$

Thus, by applying Theorem 5 3 we find that

$$h(v) = (\frac{1+2\alpha}{4\alpha})(-\frac{v_1^2}{2} + v_1v_2 + \frac{v_2\beta}{\alpha} - \frac{v_1\beta}{2\alpha} + v_1\eta) + O(|v|^3)$$

We may now substitute $h(v)$ into (5 4) and expand in order to obtain the approximate equations on the centre manifold Since β and η are constants, the equations on the centre manifold are reduced to the 2-dimensional system

$$\begin{aligned}u_1 &= A_{11}u_1 + A_{12}u_2 + B_{11}u_1^2 + B_{12}u_1u_2 + O(|u|^3), \\ u_2 &= A_{21}u_1 + A_{22}u_2 + B_{21}u_1^2 + B_{22}u_1u_2 + O(|u|^3), \qquad (5\ 10)\end{aligned}$$

where

$$\begin{aligned}A_{11} &= \eta\Phi - \frac{3\Omega\beta^2}{2\alpha}, \\ A_{12} &= \frac{\beta}{\alpha}\Phi,\end{aligned}$$

$$
\begin{aligned}
B_{11} &= 2\Phi + 1, \\
B_{12} &= 3\Omega(\eta - \frac{\beta}{\alpha}), \\
A_{21} &= (1+\eta)\alpha - \frac{3\gamma_2}{2}\eta\Phi - \frac{3\Omega\beta^2}{2\alpha}, \\
A_{22} &= -\frac{3}{2}\frac{\beta}{\alpha}\Phi, \\
B_{21} &= \alpha\Phi - \frac{3}{2}2\Phi + 1, \\
B_{22} &= 3\Omega\alpha(\frac{\eta}{2} - \frac{\beta}{\alpha}) - \frac{3}{2}(\eta - \frac{\beta}{\alpha}),
\end{aligned}
$$

and

$$
\begin{aligned}
\Omega &= \frac{1+2\alpha}{4\alpha}, \\
\Phi &= \frac{3\Omega\beta}{\alpha} - 1
\end{aligned}
$$

Although (5 10) is an approximation to the equations on the centre manifold, the stability properties of (5 6) will be contained in (5 10)

5.3 Numerical Results

AUTO is now used to find, and examine the stability of, periodic solutions of the full Explodator and the reduced system β is used as the bifurcation parameter in each case

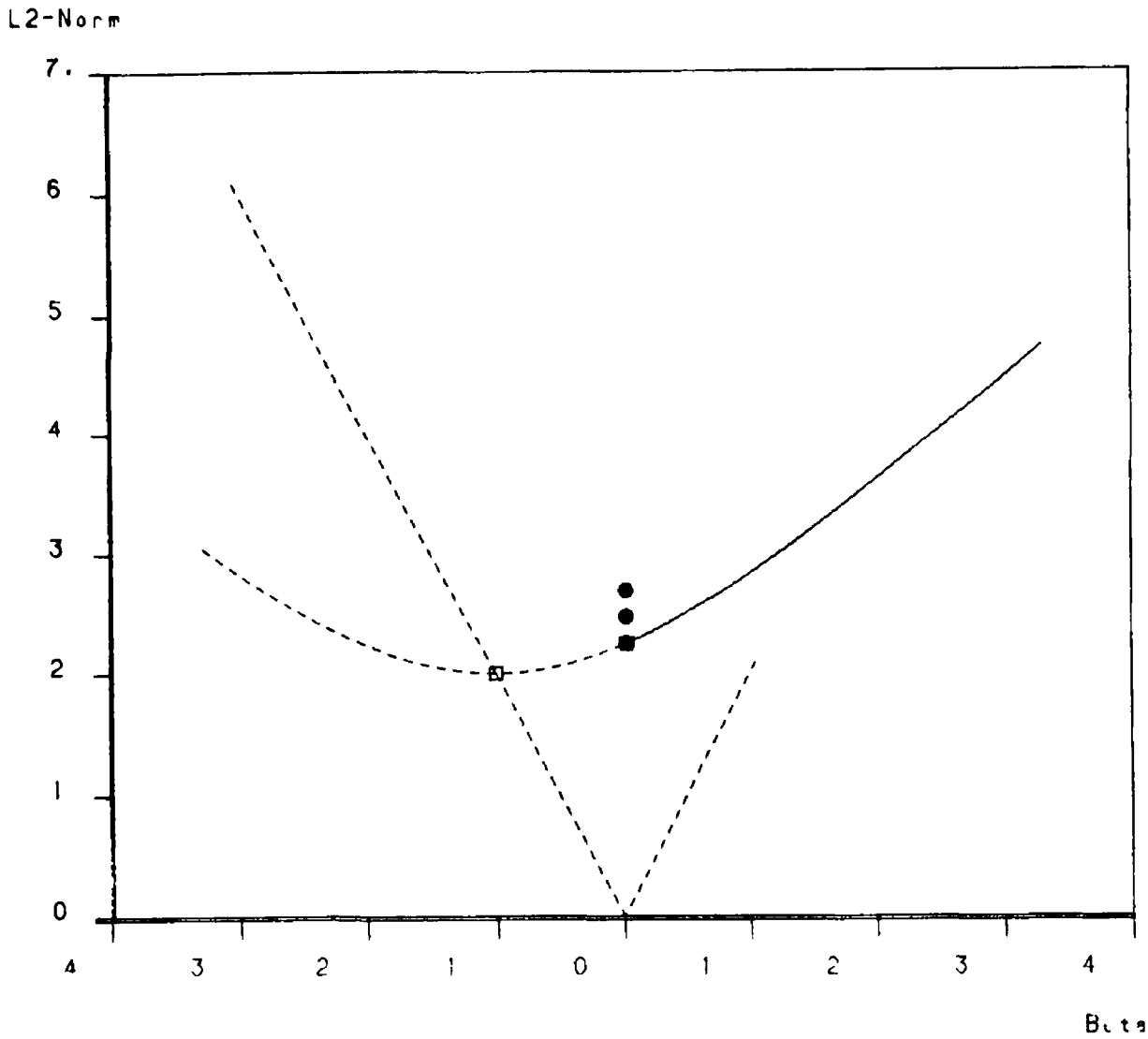

Figure 5 1 Bifurcation diagram for the full Explodator model with μ_1, μ_3, $\mu_4 = 0$ β is used as the bifurcation parameter, with an initial value of $\beta = 1\,0$ The iteration is started with $\alpha = 50$, $\mu_2 = 0$ and $\xi = (2, 1, 0\,02)$ The system has a bifurcation point at $\beta = -1$, which yields a branch of unstable stationary solutions On this branch either the parameter β is less than zero or the equilibrium point is not in the quadrant Q There is a hopf bifurcation point at $\beta = 2\,97 \times 10^{-2}$ with an emanating branch of stable periodic solutions Plotting information was obtained for several periodic solutons

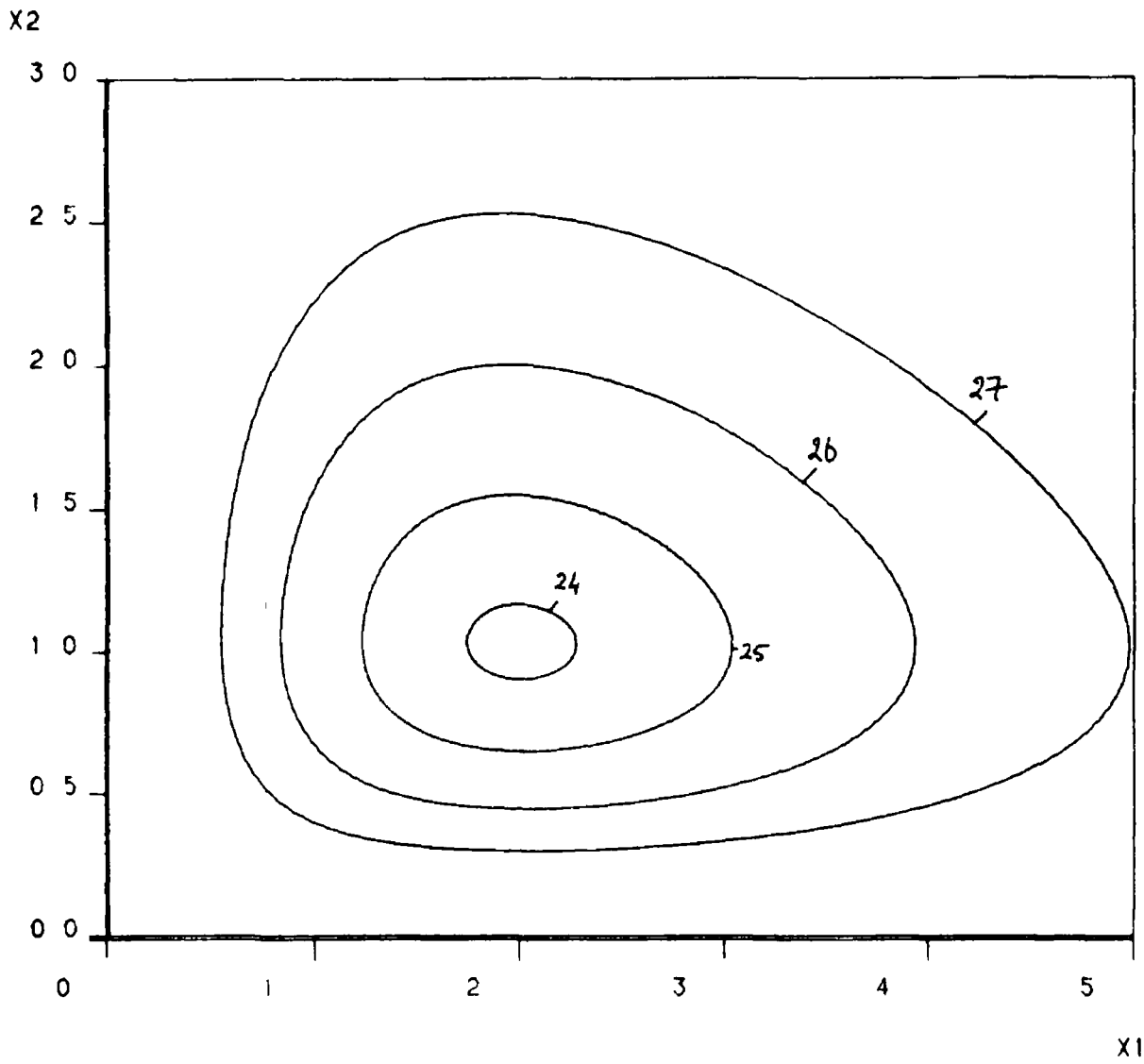

Figure 5 2 A 2-dimensional plot of the periodic orbits found for the full Exploda-tor model The parameter values and periods of the orbits are

orbit 24, $\beta = 2\,95 \times 10^2$, $T = 6\,229$,
orbit 25, $\beta = 2\,82 \times 10^2$, $T = 6\,38$,
orbit 26, $\beta = 2\,55 \times 10^2$, $T = 6\,57$,
orbit 27, $\beta = 2\,19 \times 10^2$, $T = 6\,86$

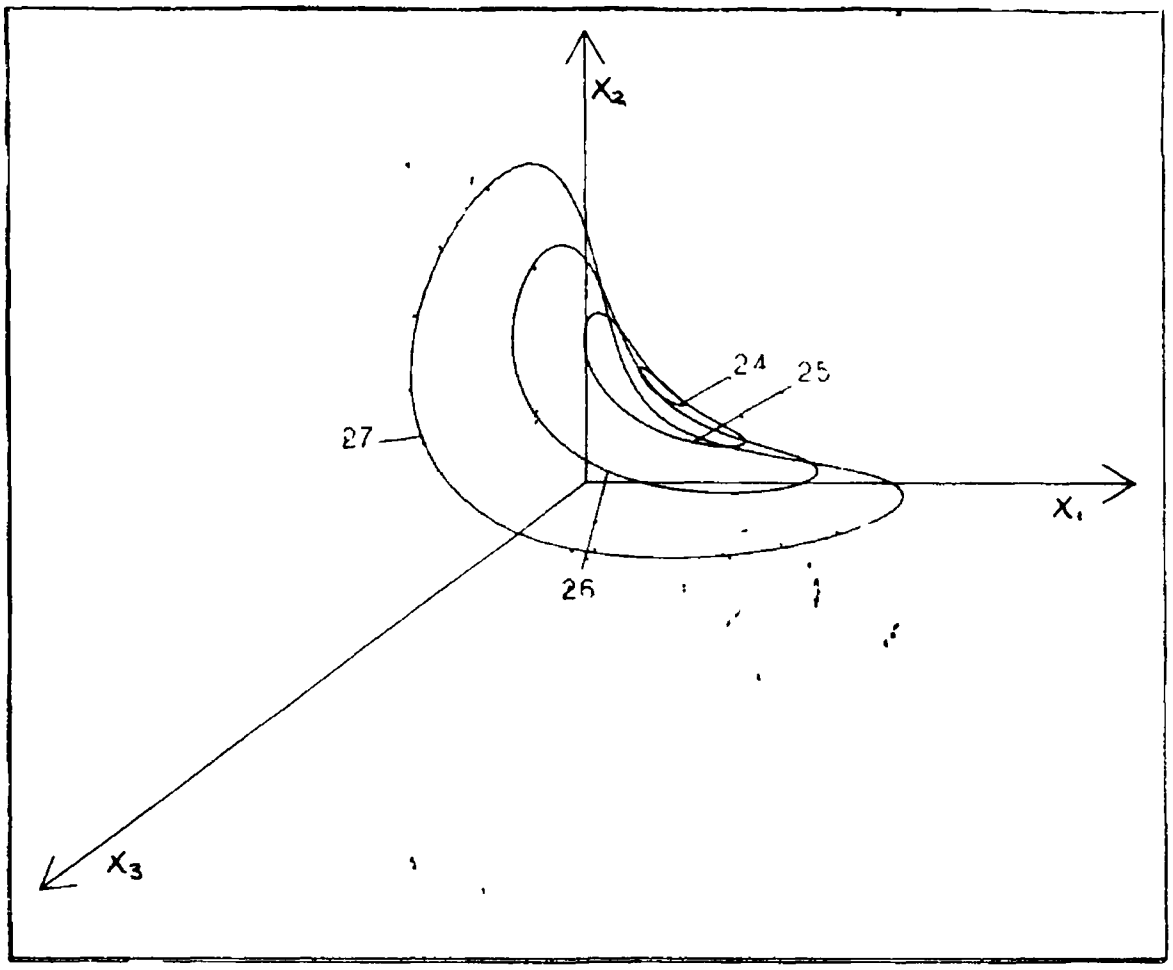

Figure 5 3 A 3-dimensional plot of the periodic orbits found for the full Exploda-tor model The projections on the planes are marked by dotted lines

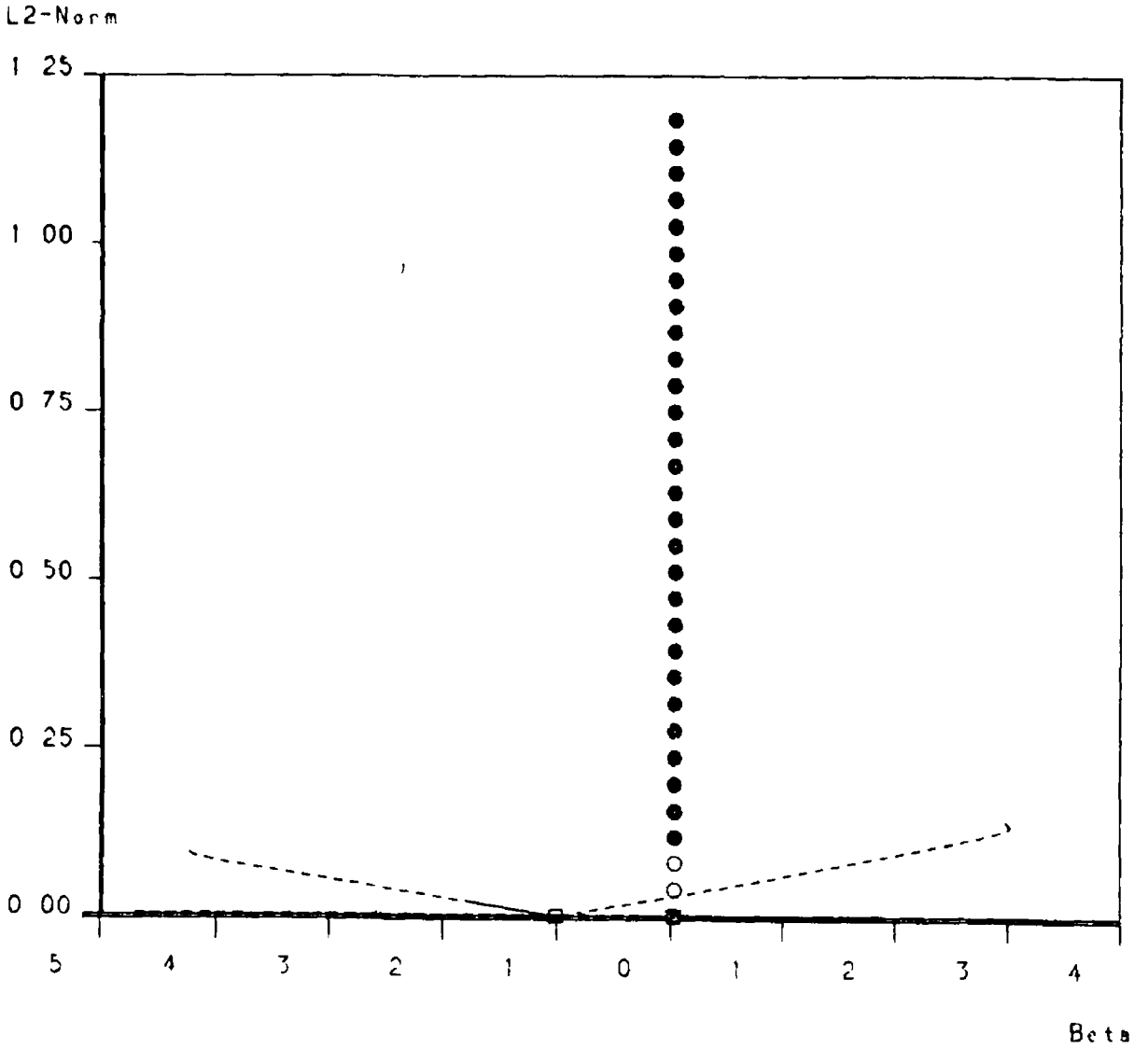

Figure 5 4 Bifurcation diagram for the reduced system β is used as the bifurcation parameter, with an initial value of $\beta = 1\,0$ The iteration was started with $\alpha = 50$, $\mu_2 = 0$ and $\xi = (0,0)$ The system has a bifurcation point at $\beta = -1$ which yields a second branch of stationary solutions There is a hopf bifurcation point at $\beta = 2\,97 \times 10^{-2}$ with an emanating branch of stable periodic solutions The stability of the first periodic solution found does not agree with that of the full model However, for this solution x is very small (e g x_3 is of the order of 10^{-34}) and since AUTO is implemented in double precision there is an unpredictibility associated with these results The stability of all other solutions found close to the origin agree in both systems The bifurcation points are identical for both systems Higher order terms in the approximation of the centre manifold become important outside a neighbourhood of the origin, so the validity of these numerical results is restricted Obviously, cubic and even quartic terms should be included if this restriction is to be weakened

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