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Algorithm of Calculation of Lyapounov Coefficients for Analysis of Chemical Autooscillations, as Applied to Calcite Crystallization Model[1]

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The numerical algorithm of calculation of Lyapounov coefficients ($\mathrm{L_k}$) *of any order is developed. The apparatus of analytical calculations is not used in this algorithm. The proposed algorithm is of use for usual computer languages and allows us to find the numerical value of* $\mathrm{L_k}$ *for any k and to make complete qualitative analyses of dynamic models on the plane.*

KEY WORDS: dynamic model, oscillatory zoning, qualitative analyses, limit cycle.

INTRODUCTION

Oscillatory zoning in minerals, as well as many other geological processes, is modeled quantitatively by a system of nonlinear differential equations, that takes account of mineral growth, diffusion, and mass continuity. The problem of whether mineral growth can oscillate autonomously with time reduces to finding the parameter values of the model under which those equations have periodic solutions. The numerical integrating of nonlinear dynamic model usually has the problems connected with instability or multiplicity of solutions. So, qualitative analysis of systems of this type is very important. It involves a linear instability analyses and a small-amplitude limit cycles bifurcation analyses. The first indicates the existence of limit cycles and the second talks about the maximum possible number of limit cycles and their stability. If the dynamic model has stable limit cycles, then the geological process, described by this model, has to be periodic. So, the problem of the number of stable limit cycles is significant in the field of oscillatory zoning in minerals (Wang and Merino, 1995). Lyapounov coefficients (L_k) are used to solve this problem. They are functions of parameters of dynamic system on the

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plane and too complex for calculation. There are completed formulas only for L_1, L_2 in the literature (Andronov and others, 1967; Bautin and Leontovich, 1976). However, Lyapounov coefficients L_k for $k > 2$ are often deficient. For example, in analyzing agate autooscillations (Bryxina and Sheplev, 1999), it was necessary to calculate Lyapounov coefficients for $k = 1, 2, 3$ and in analyzing a cubic system (Lloyd and Pearson, 1992), it was necessary to calculate Lyapounov coefficients for $k = 1, \ldots, 6$.

In our investigation we used the algorithm developed by Poincare when he solved the problem of distinctions between the center and the focus (Nemizkii and Stepanov, 1947). In this work the numerical algorithm that is developed is capable for the traditional programming languages and does not use the apparatus of analytical calculations on computer.

LYAPOUNOV COEFFICIENTS

We investigate a dynamic system on the plane

$$\frac{dx}{dt} = Q(x, y)$$
$$\frac{dy}{dt} = P(x, y) \tag{1}$$

and write

$$Q(x, y) = Q_1(x, y) + \cdots + Q_n(x, y)$$
$$P(x, y) = P_1(x, y) + \cdots + P_n(x, y)$$

where Q_k and P_k are homogeneous polynomials of degree k, whose coefficients can depend on some parameters. The steady states of system (1) are obtained by setting the time derivatives of the left side to zero, and solving for x and y, now designated as x_0, y_0:

$$Q(x_0, y_0) = P(x_0, y_0) = 0$$

We introduce small perturbations ξ and η around a steady state (x_0, y_0), and replace $x = x_0 + \xi$ and $y = y_0 + \eta$ into (1). We obtain

$$\frac{d\xi}{dt} = a\xi + b\eta + \varphi(\xi, \eta)$$
$$\frac{d\eta}{dt} = c\xi + d\eta + \psi(\xi, \eta) \tag{2}$$

A general solution to linear part of (2) has the following form:

$$\xi = c_1 e^{\lambda_1 t} + c_2 e^{\lambda_2 t}$$
$$\eta = c_1 \lambda_1 e^{\lambda_1 t} + c_2 \lambda_2 e^{\lambda_2 t}$$

where c_1 and c_2 are integration constants, and λ_1 and λ_2 are the roots of the characteristic equation $\lambda^2 - \sigma\lambda + \Delta = 0$. The behavior of system (2) near the steady state can be characterized by determinant Δ and trace σ of matrix of linear part of (2). According to Δ and σ, the steady states of the system are classified into (Bautin and Leontovich, 1976) (1) *saddle point*, if $\Delta < 0$ and σ is arbitrary; (2) *node point*, if $\Delta > 0$ and $(\sigma/2)^2 - \Delta > 0$; (3) *focus point*, if $\Delta > 0$ and $(\sigma/2)^2 - \Delta < 0$; (4) *limit cycle*, if $\Delta > 0$ and $\sigma = 0$. In cases 1–3, the steady state is stable if $\sigma < 0$ and unstable if $\sigma > 0$; case 4 requires special investigation of functions $\varphi(\xi, \eta)$ and $\psi(\xi, \eta)$. Let us eliminate t from system (2)

$$\frac{d\xi}{d\eta} = \frac{a\xi + b\eta + \varphi(x, y)}{c\xi + d\eta + \psi(x, y)} \tag{3}$$

The solution to Eq. (3) is curve $\xi(\eta)$, which is called a phase trajectory. A periodical solution of system (2) is a closed phase trajectory (limit cycle) of Eq. (3). All phase trajectories of Eq. (3) give phase portrait of dynamic system (1). So, the parameter values, when there are limit cycles, are interesting for us when we qualitatively analyze the model.

Let us turn to the polar coordinates (r, θ): $\xi = r\cos(\theta)$ and $\eta = r\sin(\theta)$ (Andronov and others, 1967). Then Eq. (3)

$$\frac{dr}{d\theta} = R(r, \theta), \tag{4}$$

where $R(r, \theta)$ is a periodic function with period 2π. Furthermore, $R(0, 0) = 0$, that is, $r = 0$ is the solution to Eq. (4). Consequently, function $R(r, \theta)$ can be presented as a power series

$$R(r, \theta) = rR_1(\theta) + r^2 R_2(\theta) + \cdots, \tag{5}$$

which will converge on the solution to Eq. (4) at any θ and reasonably small r.

The solution to Eq. (5) $r = f(\theta, r_0)$, where r_0 is the initial value of r at $\theta = 0$, can be also expanded in power series:

$$r = f(\theta, r_0) = u_1(\theta)r_0 + u_2(\theta)r_0^2 + \cdots \tag{6}$$

Substituting expansion (6) into Eq. (5) and equating the coefficients at like power r_0 to each other, we obtain recurrent differential equations for finding the functions $u_i(\theta)$:

$$\begin{aligned}
\frac{du_1}{d\theta} &= R_1(\theta) \\
\frac{du_2}{d\theta} &= R_1(\theta)u_2 + R_2(\theta)u_1^2 \\
\frac{du_3}{d\theta} &= R_1(\theta)u_3 + 2R_2(\theta)u_1u_2 + R_3(\theta)u_1^3 \\
&\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots
\end{aligned} \tag{7}$$

Providing that $r_0 = f(\theta = 0, r_0)$, we have the following initial values for the functions $u_i(\theta)$: $u_1(0) = 1$ and $u_i(0) = 0$ for $i > 1$. So, all functions $u_i(\theta)$ can be found sequentially from (7). The function

$$w = \frac{r - r_0}{r_0} = \alpha_2 r_0 + \alpha_3 r_0^2 + \alpha_4 r_0^3 + \alpha_5 r_0^4 + \cdots,$$

where $\alpha_i = u_i(2\pi)$ is called a rate of change phase trajectory of Eq. (4), and the coefficients α_i are the focal values. It is well known (Andronov and others, 1967) about the focal values that if $\Delta > 0$ and $\sigma = 0$, then $\alpha_2 = 0$; if $\sigma = 0$ and $\alpha_3 = 0$, then $\alpha_4 = 0$; if $\sigma = 0$, $\alpha_3 = 0$, and $\alpha_5 = 0$, then $\alpha_6 = 0$; and so on. The focal value α_3 at $\sigma = 0$ is called the first Lyapounov coefficient L_1; the focal value α_5 at $\sigma = 0$ and $\alpha_3 = 0$ is called the second Lyapounov coefficient L_2 and so on. If σ is sufficiently small and it is not equal to zero, the function w can be rewritten in the following form:

$$w = \sigma + L_1 r_0^2 + L_2 r_0^4 + L_3 r_0^6 + \cdots$$

The limit cycle stability is determined by the sign of w, that is, by the signs of Lyapounov coefficients, the calculation of which is difficult, but necessary at qualitative analyses of dynamic system on the plane.

DESCRIPTION OF ALGORITHM

We consider the system of differential equations of the following form:

$$\begin{aligned}
\frac{dX}{dt} &= aX + bY + q(X, Y) \\
\frac{dY}{dt} &= cX + dY + p(X, Y)
\end{aligned} \tag{8}$$

in which $q(x, y)$ and $p(x, y)$ are polynomials. As it follows from the theory of bifurcation (Nemitskii and Stepanov, 1947), system (8) has periodical solutions if its steady state is a limit cycle, that is, if the parameter values of this system satisfy the conditions

$$\sigma = a + d = 0 \quad \text{and} \quad \Delta = ad - bc > 0$$

Here σ is a trace of matrix of linear part of (8) and Δ is its determinant. Let us replace variables:

$$x = -\frac{aX + bY}{\sqrt{\Delta}} \qquad y = Y \qquad \tau = \sqrt{\Delta}t$$

System (8) becomes, in its canonical form,

$$\begin{aligned} \frac{dx}{d\tau} &= y + \sum_{i=2} q_i \\ \frac{dy}{d\tau} &= -x + \sum_{i=2} p_i \end{aligned} \tag{9}$$

in which p_i, q_i are also homogenous polynomials by powers of x and y:

$$p_i = \sum_{j=0}^{i} p_{ij} x^j y^{i-j}, \qquad q_i = \sum_{j=0}^{i} q_{ij} x^j y^{i-j} \tag{10}$$

and its coefficients p_{ij}, q_{ij} depend on the parameter values of the model. Let us consider that $p_{ij} = 0$ and $q_{ij} = 0$, if $j < 0$ or $j > i$, then the derivatives of p_i and q_i on x and y are written in the following form:

$$\begin{aligned} \frac{\partial p_i}{\partial x} &= p_{i,x} = \sum_{j=0}^{i} z_{ij} x^{j-1} y^{i-j} j & \qquad \frac{\partial q_i}{\partial x} &= q_{i,x} = \sum_{j=0}^{i} z_{ij} x^{j-1} y^{i-j} j \\ \frac{\partial p_i}{\partial y} &= p_{i,y} = \sum_{j=0}^{i-1} z_{ij} x^{j} y^{i-j-1} (i-j) & \qquad \frac{\partial q_i}{\partial y} &= q_{i,y} = \sum_{j=0}^{i-1} z_{ij} x^{j} y^{i-j-1} (i-j) \end{aligned}$$

Eliminating τ from system (9) and will give the following equation:

$$\frac{dy}{dx} = \frac{-x + \sum_{i=2} p_i}{y + \sum_{i=2} q_i}. \tag{11}$$

Search of periodic solutions of system (8) is same as the search of closed trajectories on phase plot (x, y), which satisfy Eq. (11). Let us represent a set of closed curves in the following form:

$$f(x, y) = x^2 + y^2 + f_3 + f_4 + \cdots + f_k + \cdots = \text{constant}, \tag{12}$$

where,

$$\begin{aligned}
f_3 &= f_{33}x^3 + f_{32}x^2y + f_{31}xy^2 + f_{30}y^3 \\
f_4 &= f_{44}x^4 + f_{43}x^3y + f_{42}x^2y^2 + f_{41}xy^3 + f_{40}y^4 \\
&\vdots \\
f_k &= \sum_{j=0}^{k} f_{kj}x^jy^{k-j} \\
&\vdots
\end{aligned}$$

and f_{ij} are unknown coefficients for the time being, although they can be found from the condition that the function $f(x, y)$ is the solution of Eq. (11). The total differential of Eq. (12) is equal to zero, that is,

$$df(x, y) = \frac{\partial f(x, y)}{\partial x}\,dx + \frac{\partial f(x, y)}{\partial y}\,dy = 0. \tag{13}$$

Taking $f(x, y)$ from the Eq. (12) we have

$$\frac{\partial f(x, y)}{\partial x} = 2x + \sum_{i=3} f_{i,x}, \qquad \frac{\partial f(x, y)}{\partial y} = 2y + \sum_{i=3} f_{i,y}$$

where

$$f_{i,x} = \frac{\partial f_i}{\partial x} = \sum_{j=0}^{i} f_{ij}x^{j-1}y^{i-j}j \qquad f_{i,y} = \frac{\partial f_i}{\partial y} = \sum_{j=0}^{i-1} f_{ij}x^jy^{i-j-1}(i-j) \tag{14}$$

Equation (13) can be written in the form of

$$\frac{dy}{dx} = -\frac{2x + \sum_{i=3} f_{i,x}}{2y + \sum_{i=3} f_{i,y}} \tag{15}$$

Comparing this equation with Eq. (11), we obtain

$$\frac{-x + \sum_{i=2} p_i}{y + \sum_{i=2} q_i} = -\frac{2x + \sum_{i=3} f_{i,x}}{2y + \sum_{i=3} f_{i,y}} \tag{16}$$

The terms of the second order cancel each other in Eq. (16). Canceling step-by-step the terms of the third, fourth, etc. orders, we can have some equations to find the coefficients f_{ij}. Using the following designations

$$a_0 = \sum_{i=3} \left(y f_{i,x} - x f_{i,y}\right) \qquad a_1 = \sum_{i=3} (2y p_{i-1} + 2x q_{i-1})$$
$$a_2 = \sum_{k=2} p_k \sum_{m=3} f_{m,y} \qquad a_3 = \sum_{k=2} q_k \sum_{m=3} f_{m,x} \tag{17}$$

Equation (16) becomes

$$a_0 + a_1 + a_2 + a_3 = 0 \tag{18}$$

Substitution of p_i, q_i, $f_{i,x}$, and $f_{i,y}$ into (17) gives

$$a_0 = \sum_{i=3} \left(\sum_{j=0}^{i} f_{i,j+1}(j+1) - f_{i,j-1}(i-j+1) \right) x^j y^{i-j}$$
$$a_1 = \sum_{i=3} \left(\sum_{j=0}^{i} (2p_{i-1,j} + 2q_{j-1,j-1}) \right) x^i y^{i-j}$$
$$a_2 = \sum_{i=4}^{\infty} \sum_{j=0}^{i} \left(\sum_{m=3}^{i-1} \sum_{n=\max(0,j-i-1+m)}^{\min(m-1,j)} f_{mn} p_{i+1-m,j-n}(m-n) \right) x^j y^{i-j}$$
$$a_3 = \sum_{i=4}^{\infty} \sum_{j=0}^{i} \left(\sum_{m=3}^{i-1} \sum_{n=\max(1,j-i+m)}^{\min(m,j+1)} q_{i+1-m,j+1-n} f_{mn} n \right) x^j y^{i-j}$$

We substitute the resulting expressions of a_0, a_1, a_2, and a_3 into (18) and set the coefficient at $x^i y^{i-j}$ equal to zero. We have for $i \geq 3$, $i \geq j \geq 0$:

$$f_{i,j+1}(j+1) - f_{i,j-1}(i-j+1) + c_{ij} = 0 \tag{19}$$

where

$$c_{ij} = 2(p_{i-1,j} + q_{i-1,j-1}) + \sum_{m=3}^{i-1} \left(\sum_{n=\max(0,j-i-1+m)}^{\min(m-1,j)} f_{mn} p_{i+1-m,j-n}(m-n) \right.$$
$$\left. + \sum_{n=\max(1,j-i+m)}^{\min(m,j+1)} q_{i+1-m,j+1-n} f_{mn} n \right) \tag{20}$$

Using index agreement (10), Eq. (20) can be simplified to the following form:

$$c_{ij} = 2(p_{i-1,j} + q_{i-1,j-1}) + \sum_{m=3}^{i-1}\sum_{n=0}^{j+1} f_{mn}[(m-n)p_{i+1-m,j-n} + nq_{i+1-m,j+1-n}] \tag{21}$$

The system of linear Eqs. (19) is divided into two independent subsystems with two-diagonal matrixes. If index i is odd, each of those subsystems is simultaneous. If index i is even ($i = 2k$), the first subsystem is overdetermined and the second one is undetermined. According to the algorithm (Nemytskii and Stepanov, 1947), in the first subsystem we take away the last equation, and in the second subsystem we add equation $f_{ii} = 0$. The condition of the Cronekera–Couppelly theorem of consistency of system can be expressed in the following form:

$$f_{i,i-1} = c_{ii}$$

The Lyapounov coefficient of index $k-1$ is

$$L_{k-1} = -f_{i,i-1} + c_{ii} \tag{22}$$

It is necessary to have all $f_{l,j}$, $0 \leq l < i$, to calculate c_{ij}. Formula (22) for $k = 2, 3, 4, 5, 6$ can be rewritten in the following form (the coefficients are given with a precision of positive factor):

$$L_1 = (3\ \ 1\ \ 3)(c_{40}\ \ c_{42}\ \ c_{44})^{\mathrm{T}}$$
$$L_2 = (5\ \ 1\ \ 1\ \ 5)(c_{60}\ \ c_{62}\ \ c_{64}\ \ c_{66})^{\mathrm{T}}$$
$$L_3 = (35\ \ 5\ \ 3\ \ 5\ \ 35)(c_{80}\ \ c_{82}\ \ c_{84}\ \ c_{86}\ \ c_{88})^{\mathrm{T}}$$
$$L_4 = (63\ \ 7\ \ 3\ \ 3\ \ 7\ \ 63)(c_{10,0}\ \ c_{10,2}\ \ c_{10,4}\ \ c_{10,6}\ \ c_{10,8}\ \ c_{10,10})^{\mathrm{T}}$$
$$L_5 = (231\ \ 21\ \ 7\ \ 5\ \ 7\ \ 21\ \ 231)(c_{12,0}\ \ c_{12,2}\ \ c_{12,4}\ \ c_{12,6}\ \ c_{12,8}\ \ c_{12,10}\ \ c_{12,12})^{\mathrm{T}}$$

CALCULATION OF L_1, L_2, L_3

Now we present the algorithm of calculation L_1, L_2, L_3 in some detail. Let us take $i = 3$ and take c_{3j} from (21):

$$c_{30} = 2p_{20} \qquad c_{31} = 2(p_{21} + q_{20}) \qquad c_{32} = 2(p_{22} + q_{21}) \qquad c_{33} = 2q_{22}$$

Systems (19) fall into two independent subsystems with the odd and even indices j:

$$\begin{pmatrix} 1 & 0 \\ -2 & 3 \end{pmatrix}\begin{pmatrix} f_{32} \\ f_{30} \end{pmatrix} = \begin{pmatrix} c_{33} \\ c_{31} \end{pmatrix} \qquad \begin{pmatrix} 1 & 0 \\ -2 & 3 \end{pmatrix}\begin{pmatrix} f_{31} \\ f_{33} \end{pmatrix} = -\begin{pmatrix} c_{30} \\ c_{32} \end{pmatrix}$$

Let us solve these systems and find f_{3j}, $j = 0, 1, 2, 3$. Let us take $i = 4$ and take c_{4j} from (21):

$$\begin{aligned}
c_{40} &= 3f_{30}p_{20} + f_{31}q_{20} + 2p_{30} \\
c_{41} &= 3f_{30}p_{21} + 2f_{31}p_{20} + f_{31}q_{21} + 2f_{32}q_{20} + 2p_{31} + 2q_{30} \\
c_{42} &= 3f_{30}p_{22} + 2f_{31}p_{21} + f_{31}q_{22} + f_{32}p_{20} + 2f_{32}q_{21} + 3f_{33}q_{20} \\
&\quad + 2p_{32} + 2q_{31} \\
c_{43} &= 2f_{31}p_{22} + f_{32}p_{21} + 2f_{32}q_{22} + 3f_{33}q_{21} + 2p_{33} + 2q_{32} \\
c_{44} &= f_{32}p_{22} + 3f_{33}q_{22} + 2q_{33}
\end{aligned}$$

We can see that c_{4j} depends on f_{3j}. For $i = 4$ systems (19) fall into two independent subsystems: The first subsystem is undetermined and the second one is overdetermined. According to the algorithm (Nemytskii and Stepanov, 1960), in the first subsystem we add equation $f_{44} = 0$, and in the second subsystem we take away the last equation. This condition Cronekera–Couppelly coincides with demand $L_1 = 0$ and we can find the first Lyapounov coefficient as

$$L_1 = 3c_{40} + c_{42} + 3c_{44}$$

So f_{4j} are found by solving the following systems:

$$\begin{pmatrix} 2 & 0 \\ -2 & 4 \end{pmatrix}\begin{pmatrix} f_{42} \\ f_{40} \end{pmatrix} = \begin{pmatrix} c_{43} \\ c_{41} \end{pmatrix} \qquad \begin{pmatrix} 1 & 0 \\ -3 & 3 \end{pmatrix}\begin{pmatrix} f_{41} \\ f_{43} \end{pmatrix} = -\begin{pmatrix} c_{40} \\ c_{42} \end{pmatrix}$$

Let us take $i = 5$ and take c_{5j} from (21):

$$\begin{aligned}
c_{50} &= 3f_{30}p_{30} + f_{31}q_{30} + 4f_{40}p_{20} + f_{41}q_{20} + 2p_{40} \\
c_{51} &= 3f_{30}p_{31} + 2f_{31}p_{30} + f_{31}q_{31} + 2f_{32}q_{30} + 4f_{40}p_{21} \\
&\quad + 3f_{41}p_{20} + f_{41}q_{21} + 2f_{42}q_{20} + 2p_{41} + 2q_{40} \\
c_{52} &= 3f_{30}p_{32} + 2f_{31}p_{31} + f_{31}q_{32} + f_{32}p_{30} + 2f_{32}q_{31} + 3f_{33}q_{30} \\
&\quad + 4f_{40}p_{22} + 3f_{41}p_{21} + f_{41}q_{22} + 2f_{42}p_{20} + 2f_{42}q_{21} + 3f_{43}q_{20} \\
&\quad + 2p_{42} + 2q_{41} \\
c_{53} &= 3f_{30}p_{33} + 2f_{31}p_{32} + f_{31}q_{33} + f_{32}p_{31} + 2f_{32}q_{32} + 3f_{33}q_{31} \\
&\quad + 3f_{41}p_{22} + 2f_{42}p_{21} + f_{42}q_{22} + f_{43}p_{20} + 3f_{43}q_{21} + 4f_{44}q_{20} \\
&\quad + 2p_{43} + 2q_{42}
\end{aligned}$$

$$
\begin{aligned}
c_{54} &= 2 f_{31} p_{33} + f_{32} p_{32} + 2 f_{32} q_{33} + 3 f_{33} q_{32} + 2 f_{42} p_{22} + f_{43} p_{21} \\
&\quad + 3 f_{43} q_{22} + 4 f_{44} q_{21} + 2 p_{44} + 2 q_{43} \\
c_{55} &= f_{32} p_{33} + 3 f_{33} q_{33} + f_{43} p_{22} + 4 f_{44} q_{22} + 2 q_{44}
\end{aligned}
$$

Systems (19) fall into two independent subsystems with the odd and even indices j:

$$
\begin{pmatrix} 1 & 0 & 0 \\ -4 & 3 & 0 \\ 0 & -2 & 5 \end{pmatrix} \begin{pmatrix} f_{54} \\ f_{52} \\ f_{50} \end{pmatrix} = \begin{pmatrix} c_{55} \\ c_{53} \\ c_{51} \end{pmatrix} \qquad \begin{pmatrix} 1 & 0 & 0 \\ -4 & 3 & 0 \\ 0 & -2 & 5 \end{pmatrix} \begin{pmatrix} f_{51} \\ f_{53} \\ f_{55} \end{pmatrix} = -\begin{pmatrix} 50 \\ c_{52} \\ c_{54} \end{pmatrix}
$$

Let us solve these systems and find f_{5j}, $j = 0, 1, 2, 3, 4, 5$. Let us take $i = 6$ and take c_{6j} from (21):

$$
\begin{aligned}
c_{60} &= 3 f_{30} p_{40} + f_{31} q_{40} + 4 f_{40} p_{30} + f_{41} q_{30} + 5 f_{50} p_{20} + f_{51} q_{20} + 2 p_{50} \\
c_{61} &= 3 f_{30} p_{41} + 2 f_{31} p_{40} + f_{31} q_{41} + 2 f_{32} q_{40} + 4 f_{40} p_{31} + 3 f_{41} p_{30} \\
&\quad + f_{41} q_{31} + 2 f_{42} q_{30} + 5 f_{50} p_{21} + 4 f_{51} p_{20} + f_{51} q_{21} + 2 f_{52} q_{20} \\
&\quad + 2 p_{51} + 2 q_{50} \\
c_{62} &= 3 f_{30} p_{42} + 2 f_{31} p_{41} + f_{31} q_{42} + f_{32} p_{40} + 2 f_{32} q_{41} + 3 f_{33} q_{40} \\
&\quad + 4 f_{40} p_{32} + 3 f_{41} p_{31} + f_{41} q_{32} + 2 f_{42} p_{30} + 2 f_{42} q_{31} + 3 f_{43} q_{30} \\
&\quad + 5 f_{50} p_{22} + 4 f_{51} p_{21} + f_{51} q_{22} + 3 f_{52} p_{20} + 2 f_{52} q_{21} + 3 f_{53} q_{20} \\
&\quad + 2 p_{52} + 2 q_{51} \\
c_{63} &= 3 f_{30} p_{43} + 2 f_{31} p_{42} + f_{31} q_{43} + f_{32} p_{41} + 2 f_{32} q_{42} + 3 f_{33} q_{41} \\
&\quad + 4 f_{40} p_{33} + 3 f_{41} p_{32} + f_{41} q_{33} + 2 f_{42} p_{31} + 2 f_{42} q_{32} + f_{43} p_{30} \\
&\quad + 3 f_{43} q_{31} + 4 f_{44} q_{30} + 4 f_{51} p_{22} + 3 f_{52} p_{21} + 2 f_{52} q_{22} + 2 f_{53} p_{20} \\
&\quad + 3 f_{53} q_{21} + 4 f_{54} q_{20} + 2 p_{53} + 2 q_{52} \\
c_{64} &= 3 f_{30} p_{44} + 2 f_{31} p_{43} + f_{31} q_{44} + f_{32} p_{42} + 2 f_{32} q_{43} + 3 f_{33} q_{42} \\
&\quad + 3 f_{41} p_{33} + 2 f_{42} p_{32} + 2 f_{42} q_{33} + f_{43} p_{31} + 3 f_{43} q_{32} + 4 f_{44} q_{31} \\
&\quad + 3 f_{52} p_{22} + 2 f_{53} p_{21} + 3 f_{53} q_{22} + f_{54} p_{20} + 4 f_{54} q_{21} + 5 f_{55} q_{20} \\
&\quad + 2 p_{54} + 2 q_{53} \\
c_{65} &= 2 f_{31} p_{44} + f_{32} p_{43} + 2 f_{32} q_{44} + 3 f_{33} q_{43} + 2 f_{42} p_{33} + f_{43} p_{32} \\
&\quad + 3 f_{43} q_{33} + 4 f_{44} q_{32} + 2 f_{53} p_{22} + f_{54} p_{21} + 4 f_{54} q_{22} + 5 f_{55} q_{21} \\
&\quad + 2 p_{55} + 2 q_{54} \\
c_{66} &= f_{32} p_{44} + 3 f_{33} q_{44} + f_{43} p_{33} + 4 f_{44} q_{33} + f_{54} p_{22} + 5 f_{55} q_{22} + 2 q_{55}
\end{aligned}
$$

For $i = 6$, systems (19) fall into two independent subsystems too, but the first subsystem is undetermined and the second one is overdetermined. According to the algorithm (Nemytskii and Stepanov, 1960), in the first subsystem we add equation $f_{66} = 0$. In the second subsystem we take away the last equation. This condition Cronekera–Couppelly coincides with demand $L_2 = 0$ (at condition: $L_1 = 0$) and we can find the second Lyapounov coefficient as

$$L_2 = 5c_{60} + c_{62} + c_{64} + 5c_{66}$$

So f_{6j} are found by sloving the following systems:

$$\begin{pmatrix} 2 & 0 & 0 \\ -4 & 4 & 0 \\ 0 & -2 & 6 \end{pmatrix} \begin{pmatrix} f_{64} \\ f_{62} \\ f_{60} \end{pmatrix} = \begin{pmatrix} c_{65} \\ c_{63} \\ c_{61} \end{pmatrix} \qquad \begin{pmatrix} 1 & 0 & 0 \\ -5 & 3 & 0 \\ 0 & -3 & 5 \end{pmatrix} \begin{pmatrix} f_{61} \\ f_{63} \\ f_{65} \end{pmatrix} = -\begin{pmatrix} c_{60} \\ c_{62} \\ c_{64} \end{pmatrix}$$

Let us take $i = 7$ and take c_{7j} from (21):

$$\begin{aligned}
c_{70} &= 3 f_{30} p_{50} + f_{31} q_{50} + 4 f_{40} p_{40} + f_{41} q_{40} + 5 f_{50} p_{30} + f_{51} q_{30} \\
&\quad + 6 f_{60} p_{20} + f_{61} q_{20} + 2 p_{60} \\
c_{71} &= 3 f_{30} p_{51} + 2 f_{31} p_{50} + f_{31} q_{51} + 2 f_{32} q_{50} + 4 f_{40} p_{41} + 3 f_{41} p_{40} \\
&\quad + f_{41} q_{41} + 2 f_{42} q_{40} + 5 f_{50} p_{31} + 4 f_{51} p_{30} + f_{51} q_{31} + 2 f_{52} q_{30} \\
&\quad + 6 f_{60} p_{21} + 5 f_{61} p_{20} + f_{61} q_{21} + 2 f_{62} q_{20} + 2 p_{61} + 2 q_{60} \\
c_{72} &= 3 f_{30} p_{52} + 2 f_{31} p_{51} + f_{31} q_{52} + f_{32} p_{50} + 2 f_{32} q_{51} + 3 f_{33} q_{50} \\
&\quad + 4 f_{40} p_{42} + 3 f_{41} p_{41} + f_{41} q_{42} + 2 f_{42} p_{40} + 2 f_{42} q_{41} + 3 f_{43} q_{40} \\
&\quad + 5 f_{50} p_{32} + 4 f_{51} p_{31} + f_{51} q_{32} + 3 f_{52} p_{30} + 2 f_{52} q_{31} + 3 f_{53} q_{30} \\
&\quad + 6 f_{60} p_{22} + 5 f_{61} p_{21} + f_{61} q_{22} + 4 f_{62} p_{20} + 2 f_{62} q_{21} + 3 f_{63} q_{20} \\
&\quad + 2 p_{62} + 2 q_{61} \\
c_{73} &= 3 f_{30} p_{53} + 2 f_{31} p_{52} + f_{31} q_{53} + f_{32} p_{51} + 2 f_{32} q_{52} + 3 f_{33} q_{51} \\
&\quad + 4 f_{40} p_{43} + 3 f_{41} p_{42} + f_{41} q_{43} + 2 f_{42} p_{41} + 2 f_{42} q_{42} + f_{43} p_{40} \\
&\quad + 3 f_{43} q_{41} + 4 f_{44} q_{40} + 5 f_{50} p_{33} + 4 f_{51} p_{32} + f_{51} q_{33} + 3 f_{52} p_{31} \\
&\quad + 2 f_{52} q_{32} + 2 f_{53} p_{30} + 3 f_{53} q_{31} + 4 f_{54} q_{30} + 5 f_{61} p_{22} + 4 f_{62} p_{21} \\
&\quad + 2 f_{62} q_{22} + 3 f_{63} p_{20} + 3 f_{63} q_{21} + 4 f_{64} q_{20} + 2 p_{63} + 2 q_{62} \\
c_{74} &= 3 f_{30} p_{54} + 2 f_{31} p_{53} + f_{31} q_{54} + f_{32} p_{52} + 2 f_{32} q_{53} + 3 f_{33} q_{52} \\
&\quad + 4 f_{40} p_{44} + 3 f_{41} p_{43} + f_{41} q_{44} + 2 f_{42} p_{42} + 2 f_{42} q_{43} + f_{43} p_{41} \\
&\quad + 3 f_{43} q_{42} + 4 f_{44} q_{41} + 4 f_{51} p_{33} + 3 f_{52} p_{32} + 2 f_{52} q_{33} + 2 f_{53} p_{31}
\end{aligned}$$

$$+3f_{53}q_{32} + f_{54}p_{30} + 4f_{54}q_{31} + 5f_{55}q_{30} + 4f_{62}p_{22} + 3f_{63}p_{21}$$
$$+3f_{63}q_{22} + 2f_{64}p_{20} + 4f_{64}q_{21} + 5f_{65}q_{20} + 2p_{64} + 2q_{63}$$

$$\begin{aligned}
c_{75} = {} & 3f_{30}p_{55} + 2f_{31}p_{54} + f_{31}q_{55} + f_{32}p_{53} + 2f_{32}q_{54} + 3f_{33}q_{53} \\
& + 3f_{41}p_{44} + 2f_{42}p_{43} + 2f_{42}q_{44} + f_{43}p_{42} + 3f_{43}q_{43} + 4f_{44}q_{42} \\
& + 3f_{52}p_{33} + 2f_{53}p_{32} + 3f_{53}q_{33} + f_{54}p_{31} + 4f_{54}q_{32} + 5f_{55}q_{31} \\
& + 3f_{63}p_{22} + 2f_{64}p_{21} + 4f_{64}q_{22} + f_{65}p_{20} + 5f_{65}q_{21} + 6f_{66}q_{20} \\
& + 2p_{65} + 2q_{64} \\
c_{76} = {} & 2f_{31}p_{55} + f_{32}p_{54} + 2f_{32}q_{55} + 3f_{33}q_{54} + 2f_{42}p_{44} + f_{43}p_{43} \\
& + 3f_{43}q_{44} + 4f_{44}q_{43} + 2f_{53}p_{33} + f_{54}p_{32} + 4f_{54}q_{33} + 5f_{55}q_{32} \\
& + 2f_{64}p_{22} + f_{65}p_{21} + 5f_{65}q_{22} + 6f_{66}q_{21} + 2p_{66} + 2q_{65} \\
c_{77} = {} & f_{32}p_{55} + 3f_{33}q_{55} + f_{43}p_{44} + 4f_{44}q_{44} + f_{54}p_{33} + 5f_{55}q_{33} \\
& + f_{65}p_{22} + 6f_{66}q_{22} + 2q_{66}
\end{aligned}$$

Systems (19) fall into two independent subsystems with the odd and even indices j:

$$\begin{pmatrix} 1 & 0 & 0 \\ -4 & 3 & 0 \\ 0 & -2 & 5 \end{pmatrix} \begin{pmatrix} f_{54} \\ f_{52} \\ f_{50} \end{pmatrix} = \begin{pmatrix} c_{55} \\ c_{53} \\ c_{51} \end{pmatrix} \qquad \begin{pmatrix} 1 & 0 & 0 \\ -4 & 3 & 0 \\ 0 & -2 & 5 \end{pmatrix} \begin{pmatrix} f_{51} \\ f_{53} \\ f_{55} \end{pmatrix} = -\begin{pmatrix} c_{50} \\ c_{52} \\ c_{54} \end{pmatrix}$$

Let us slove these systems and find f_{5j}, $j = 0, 1, 2, 3, 4, 5$. Let us take $i = 8$ and take c_{8j} from (21) only for even j, since only they are needed for L_3:

$$\begin{aligned}
c_{80} = {} & 3f_{30}p_{60} + f_{31}q_{60} + 4f_{40}p_{50} + f_{41}q_{50} + 5f_{50}p_{40} + f_{51}q_{40} \\
& + 6f_{60}p_{30} + f_{61}q_{30} + 7f_{70}p_{20} + f_{71}q_{20} + 2p_{70} \\
c_{82} = {} & 3f_{30}p_{62} + 2f_{31}p_{61} + f_{31}q_{62} + f_{32}p_{60} + 2f_{32}q_{61} + 3f_{33}q_{60} \\
& + 4f_{40}p_{52} + 3f_{41}p_{51} + f_{41}q_{52} + 2f_{42}p_{50} + 2f_{42}q_{51} + 3f_{43}q_{50} \\
& + 5f_{50}p_{42} + 4f_{51}p_{41} + f_{51}q_{42} + 3f_{52}p_{40} + 2f_{52}q_{41} + 3f_{53}q_{40} \\
& + 6f_{60}p_{32} + 5f_{61}p_{31} + f_{61}q_{32} + 4f_{62}p_{30} + 2f_{62}q_{31} + 3f_{63}q_{30} \\
& + 7f_{70}p_{22} + 6f_{71}p_{21} + f_{71}q_{22} + 5f_{72}p_{20} + 2f_{72}q_{21} + 3f_{73}q_{20} \\
& + 2p_{72} + 2q_{71} \\
c_{84} = {} & 3f_{30}p_{64} + 2f_{31}p_{63} + f_{31}q_{64} + f_{32}p_{62} + 2f_{32}q_{63} + 3f_{33}q_{62} \\
& + 4f_{40}p_{54} + 3f_{41}p_{53} + f_{41}q_{54} + 2f_{42}p_{52} + 2f_{42}q_{53} + f_{43}p_{51} \\
& + 3f_{43}q_{52} + 4f_{44}q_{51} + 5f_{50}p_{44} + 4f_{51}p_{43} + f_{51}q_{44} + 3f_{52}p_{42}
\end{aligned}$$

$$
\begin{aligned}
&+2f_{52}q_{43}+2f_{53}p_{41}+3f_{53}q_{42}+f_{54}p_{40}+4f_{54}q_{41}+5f_{55}q_{40}\\
&+5f_{61}p_{33}+4f_{62}p_{32}+2f_{62}q_{33}+3f_{63}p_{31}+3f_{63}q_{32}+2f_{64}p_{30}\\
&+4f_{64}q_{31}+5f_{65}q_{30}+5f_{72}p_{22}+4f_{73}p_{21}+3f_{73}q_{22}+3f_{74}p_{20}\\
&+4f_{74}q_{21}+5f_{75}q_{20}+2p_{74}+2q_{73}\\
c_{86}=\;&3f_{30}p_{66}+2f_{31}p_{65}+f_{31}q_{66}+f_{32}p_{64}+2f_{32}q_{65}+3f_{33}q_{64}\\
&+3f_{41}p_{55}+2f_{42}p_{54}+2f_{42}q_{55}+f_{43}p_{53}+3f_{43}q_{54}+4f_{44}q_{53}\\
&+3f_{52}p_{44}+2f_{53}p_{43}+3f_{53}q_{44}+f_{54}p_{42}+4f_{54}q_{43}+5f_{55}q_{42}\\
&+3f_{63}p_{33}+2f_{64}p_{32}+4f_{64}q_{33}+f_{65}p_{31}+5f_{65}q_{32}+6f_{66}q_{31}\\
&+3f_{74}p_{22}+2f_{75}p_{21}+5f_{75}q_{22}+f_{76}p_{20}+6f_{76}q_{21}+7f_{77}q_{20}\\
&+2p_{76}+2q_{75}\\
c_{88}=\;&f_{32}p_{66}+3f_{33}q_{66}+f_{43}p_{55}+4f_{44}q_{55}+f_{54}p_{44}+5f_{55}q_{44}\\
&+f_{65}p_{33}+6f_{66}q_{33}+f_{76}p_{22}+7f_{77}q_{22}+2q_{77}
\end{aligned}
$$

For $i=8$, systems (19) fall into two independent subsystems too, but the first subsystem is undertermined and the second one is overdetermined. According to the algorithm (Nemytskii and Stepanov, 1960), in the first subsystem we add equation $f_{88}=0$. In the second subsystem, we take away the last equation. This condition of Cronekera–Couppelly coincides with demand of $L_3=0$ (at condition: $L_1=0$ and $L_2=0$) and we can determine the third Lyapounov coefficient as

$$L_3=35c_{80}+5c_{82}+3c_{84}+5c_{86}+35c_{88}$$

EXAMPLE

The growth of calcite from an aqueous solution containing trace elements has been modeled by Wang and Merino (1992). The linear stability analyses of steady states of dynamic model is made in their paper, and the periodical solutions have been found for specific parameter values. To complete qualitative analyses of this model we have to take into account its nonlinear terms. This has been done in another paper by Bryxina and Sheplev (1997), where they lead the model of Wang and Merino to the system:

$$
\begin{aligned}
\frac{dX}{d\tau}&=-X-Y-F(X,Y)\\
\frac{dY}{d\tau}&=apX+p\left(1-\frac{\rho}{\rho_1 b}\right)Y+pF(X,Y)'
\end{aligned}
\tag{23}
$$

where,

$$F(X,Y) = sY^2 + aXY + asXY^2$$
$$\rho = 1 + b + \beta b^2 \qquad \rho_1 = 1 + 2\beta b \qquad p = \frac{\rho}{\rho_1} Ba(1-a)$$
$$\omega^2 = ap - 1 \qquad s = \frac{\beta\rho}{\rho_1^2}$$

Here X and Y are small perturbations of the concentrations of aqueous species around steady-state values, and a, b, β, and B are the parameters of the model depending on diffusion coefficients of species in the solution, on kinetic parameters, and on reaction rate; all of them are dimensionless. By substituting

$$X = -\frac{r}{p}\left(u + \frac{\pi}{\omega} v\right) \qquad Y = r\left(u + \frac{\alpha}{\omega} v\right) \qquad \tau = \omega t$$

where

$$r = \frac{1}{\sqrt{as}} \qquad \pi = p - 1 \qquad \alpha = 1 - a \qquad \mu = \sqrt{\frac{a}{s}}$$

We bring Eqs. (23) to their canonical form:

$$\frac{dv}{dt} = u \qquad \frac{du}{dt} = -v + P(u,v) \qquad P(u,v) = \sum_{i=2}^{3}\left(\sum_{j=0}^{i} p_{ij} u^i v^{i-j}\right), \quad (24)$$

where

$$p_{20} = \left(\frac{p}{\mu} - \mu\right) \Big/ \omega \qquad p_{21} = \left(\frac{2\alpha p}{\mu} - (\alpha + \pi)\mu\right) \Big/ \omega^2$$
$$p_{22} = \left(\frac{\alpha^2 p}{\mu} - \alpha\pi\mu\right) \Big/ \omega^3$$
$$p_{30} = -1/\omega \qquad p_{31} = -(2\alpha + \pi)/\omega^2 \qquad p_{32} = -\alpha(\alpha + 2\pi)/\omega^3$$
$$p_{33} = -\alpha^2\pi/\omega^4 \quad (25)$$

The determinant and trace of matrix of the linear part of (23) are given as

$$\Delta = \omega^2 - \sigma, \quad \sigma = -1 + p\left(1 - \frac{\rho}{\rho_1 b}\right)$$

If $\Delta > 0$ and $\sigma = 0$, and using the above described algorithm for system (23), we shall obtain the analytical expressions for L_1 and L_2 in terms of parameters of the model:

$$
\begin{aligned}
L_1 &= a^2 - a(-2s^2p^2 - sp^2 + s + p) - 2s(sp^2 - p + 1) \\
L_2 &= a(a(a(a(p(-a + s(p(5s - 4) - 3) + 5) - 3) + p(p(p(s(p(10s^2 - 1) \\
&\quad + s(-8s - 9) + 4) + 1) + s(6s + 7) - 5) + s(-11s - 8) + 3) + 12s) \\
&\quad + p(p(p(2ps^2(2s(3s + 1) + 3) + s(s(10s(-2s - 3) - 17) - 6)) \\
&\quad + 2s(2s(3s + 4) + 11)) + s(s(10s + 17) - 29)) + 3s(-3s + 2)) \\
&\quad + p(p(p(4ps^3(-5s - 3) + 12s^2(s(3s + 4) + 1)) + 2s^2(-23s - 22)) \\
&\quad + 2s^2(6s + 23)) - 18s^2) + p(p(8ps^3(s(p - 2) - 1) + 24s^3) - 16s^3)
\end{aligned}
$$

and for L_3 in terms of p_{ij} from (25):

$$
\begin{aligned}
L_3 &= 174p_{20}^5 p_{21} + 2p_{20}^4(349p_{21}p_{22} + 261p_{30}) + 2p_{20}^3\big(p_{21}\big(472p_{22}^2 - 85p_{31} \\
&\quad - 164p_{33}\big) + 1047p_{22}p_{30}\big) + p_{20}^2\big(200p_{21}^2p_{30} + p_{21}p_{22}\big(490p_{22}^2 - 337p_{31} \\
&\quad - 818p_{33}\big) + 3p_{30}\big(944p_{22}^2 + 4p_{31} + 33p_{33}\big)\big) + 2p_{20}\big(150p_{21}^2p_{22}p_{30} \\
&\quad + p_{21}\big(35p_{22}^4 - p_{22}^2(101p_{31} + 280p_{33}) + 3\big(14p_{33}^2 + 2p_{31}(p_{31} + 3p_{33}) \\
&\quad + 39p_{30}^2\big)\big) + 21p_{22}p_{30}\big(35p_{22}^2 - 6p_{33}\big)\big) + 2p_{21}^2p_{30}\big(50p_{22}^2 - 3p_{31} \\
&\quad - 12p_{33}\big) + p_{21}p_{22}\big(35p_{22}^2(-p_{31} - 2p_{33}) + 3\big(35p_{33}^2 \\
&\quad + 9p_{33}p_{31} + 2\big(42p_{30}^2 + p_{31}^2\big)\big)\big) + 3p_{30}\big(70p_{22}^4 \\
&\quad + p_{22}^2(8p_{31} - 105p_{33}) + 3\big(p_{31}(p_{31} + 5p_{33}) - 6p_{30}^2\big)\big)
\end{aligned}
$$

We have four quantities: σ, L_1, L_2, and L_3. The first depends on the linear terms of (23), and the others depend on the nonlinear terms of (23). All of them are connected with four parameters of the model. When we take into consideration only the linear terms of the model, we have one limitation on the parameters, when there are periodical solutions, that is $\sigma = 0$. When we take into consideration the nonlinear terms too, we have more limitations on the parameters of the model, that is $L_k = 0$. So, there are more stringent restrictions on the parameter values of the model, when periodic solution exists accounting for the nonlinear terms of the model.

System (23) has four parameters: a, b, β, and B. The calculations of σ, L_1, L_2, and L_3 have shown that there are parameter values at which $\sigma = 0$, $L_1 = 0$,

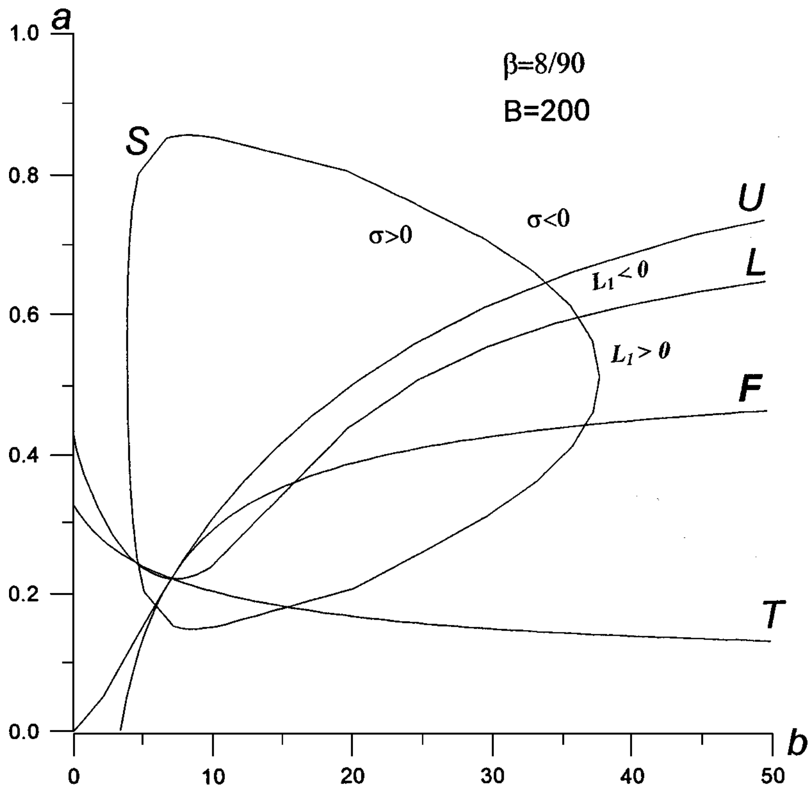

Figure 1. Behavior diagram *a–b* for $\beta = 8/90$. Curve *U* is the boundary of the region in unique steady state of system (23); curve *F* is the line of the two-fold steady states; curve *T* is the line of triple steady states; curve *S* is the line $\sigma = 0$ at $B = 200$; curve *L* is the line $L_1 = 0$.

and $L_2 = 0$, but there are no parameter values at which $L_3 = 0$. The value of L_3 is smaller than zero at any parameter value of the model. So, Lyapounov coefficients L_k for $k > 3$ are not necessary for this model.

Several important curves for parameter $\beta = 8/90$ and $B = 200$ are indicated in coordinates (a, b) in Figure 1 (at other β, the qualitative picture is the same): *U* is a line of unique steady states, *F* is a line of multiple steady states, *T* is a line of triple steady states, *S* is a line where $\sigma = 0$, and *L* is a line where the first Lyapounov coefficient is equal to zero, that is, $L_1 = 0$. The division of area of parameters β–b into subareas of constant sign of the factor L_2 is indicated in Figure 2. Using the possible sign changing of σ, L_1, and L_2, all phase portraits are found in Bryxina and Sheplev (1997) for this model. If there are parameter values when σ, L_1, L_2, and L_3 are small, but not equal to zero, and if $\sigma > 0$, $L_1 < 0$, $L_2 > 0$, and $L_3 < 0$, then there will be a phase portrait with three limit cycles (stable, unstable, and stable) around

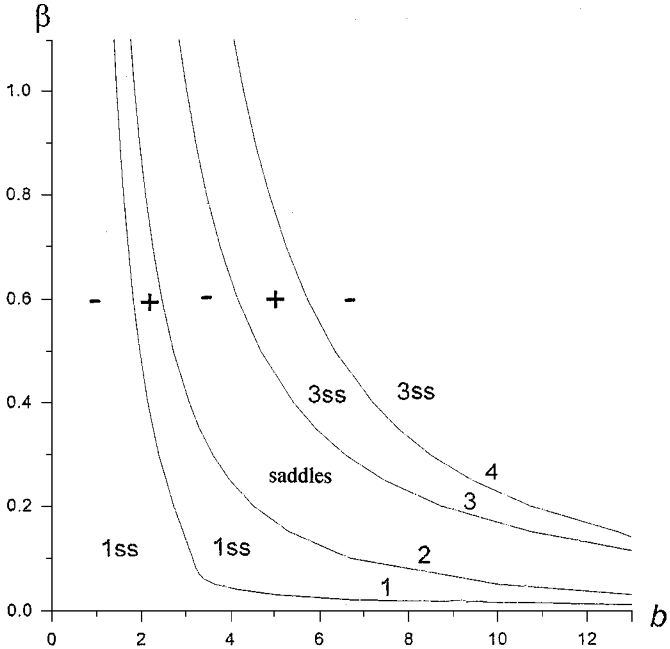

Figure 2. Behavior diagram β–b. Curves 1, 2, 3, 4 are the lines $L_2 = 0$; the region has sign (+) if $L_2 > 0$ and the region has sign (−) if $L_2 < 0$; the area of saddles separates the area (1ss) of unique steady states from the multiplicity area (3ss).

the unstable steady state. The existence of stable limit cycles of the dynamic model means that this model has periodic solutions, and, consequently, it can describe oscillatory zoning of trace elements in calcite growing from an aqueous solution.

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