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Solution of the periodic Belousov-Zhabotinsky reaction using a closed-loop mechanism

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Abstract: *Belousov-Zhabotinsky* reaction generates self-organized oscillatory pattern which is common in biological systems, synergistic study of oscillatory patterns will assist understanding and modeling complex processes in biological sphere. However, the analytical solution of a self-oscillator is difficult because the system exhibits nonlinear dynamics. In this study, a frequency domain analysis of Hopf bifurcation based on a closed-loop representation is addressed. For better understanding of the closed-loop mechanism, a Laplace-Borel transform is implemented, which is proved to be effective in identifying the coefficients of the harmonics.

Keywords: generalized Nyquist criterion, Laplace-Borel transform, describing function approach.

1. INTRODUCTION

Some processes (e.g., *Belousov–Zhabotinsky* (BZ) reaction) don't follow principles (e.g., Onsager reciprocal relation) of classic thermodynamics. I. Prigogine (1978) claimed that when open systems are far-from-equilibrium and nonlinear, structurally organized patterns would form out of symmetric-breaking bifurcations; and such open systems are named as the dissipative structure systems. Since the BZ reaction is far-from-equilibrium, the dynamics of the system does not obey the Onsager reciprocal relations and symmetry breaking bifurcation may cause the system to generate self-organized patterns. Fig. 1 depicts the generation of the self-oscillatory waveform observed in experiments.

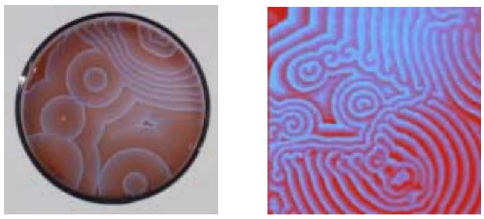

Fig. 1. The snapshot (and amplification) of the diffusive BZ reaction, which can generate waveform of color change from blue to red periodically (http://hopf.chem.brandeis.edu/anatol.htm).

By the dissipative structure theory, the BZ reaction is viewed as an open system with constant negative entropy consumption, and a portion of the overall reaction entropy-change is consumed for the maintenance of the periodic color-change structure. The reaction kinetics for the Oregonator (Field et al., 1974) model is given as follows,

$$
\begin{aligned}
A + Y &\rightarrow X + P & r_1 &= k_1 AY \\
X + Y &\rightarrow 2P & r_2 &= k_2 XY \\
A + X &\rightarrow 2X + 2Z & r_3 &= k_3 AX \\
2X &\rightarrow A + P & r_4 &= k_4 X^2 \\
B + Z &\rightarrow 0.5 f\, X & r_5 &= k_5 BZ
\end{aligned}
$$

Species identification with respect to the Field-Koros-Noyes, (FKN) mechanism (Field, 1972) are $X = HBrO^2$, $Y = Br^-$, $Z =$ Ce(IV), $A = BrO^3$, $B =$ Organic species, $P =$ HOBr, and f is an adjustable stoichiometric factor. The reactant species A and B are normally presented in much higher concentrations than the dynamic intermediate species X, Y and Z, and are assumed to be constant on the time scale of a few oscillations. The oscillatory exchange of the intermediates causes Z to vary between Ce(IV) and Ce(III) back and forth, and with the presence of the ferroin indicator, the media would change color between blue and red repeatedly, indicating a periodically dissipative system, as shown in Fig. 2.

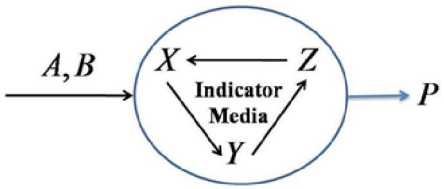

Fig. 2. Schematic of the periodically dissipative structure.

From the viewpoint of a dynamic system, the nonlinearity of the intermediate terms (X, Y, Z) brings about a Hopf bifurcation where increasing one of the parameters beyond the critical point may cause a periodical colour-change waveform to emerge, and the model is given as follows,

$$\begin{cases} X' = k_1 AY - k_2 XY + k_3 AX - 2k_4 X^2 \\ Y' = -k_1 AY - k_2 XY + 0.5 f k_5 BZ \\ Z' = 2k_3 AY - 0.5 f k_5 BZ \end{cases} \quad (1)$$

Equation (1) can be normalized in dimensionless form by defining $x = X/X_0$, $y = Y/Y_0$, $z = Z/Z_0$, $\tau = T/T_0$, and the steady-state is given as a function of the reactants A and B,

$$X_0 = \frac{k_3 A}{2k_4}; Y_0 = \frac{k_3 A}{k_2}; Z_0 = \frac{(k_3 A)^2}{k_5 k_4 B}; T_0 = \frac{1}{k_5 B}$$

which is now rephrased as follows,

$$\begin{cases} \varepsilon \dfrac{dx}{d\tau} = qy - xy + x(1-x) \\ \delta \dfrac{dy}{d\tau} = -qy - xy + f z \\ \dfrac{dz}{d\tau} = x - z \end{cases} \quad (2)$$

where

$$\varepsilon = \frac{k_5 B}{k_3 A} = 0.04; \delta = \frac{k_4 k_5 B}{k_2 k_3 A} = 0.0004; q = \frac{2k_1 k_4}{k_3 k_4} = 0.04$$

It is clear that the periodic solution of the self-oscillatory pattern is relevant to the input parameter, i.e., the input entropy flux. Knowing how the period is related to the parameter change by varying the parameter (say, f) would be critically beneficial in identifying the characteristics of the self-oscillatory structure.

Our goal in this study is to develop mathematical methods to compute the period of limit cycle as a function of parametric changes. Often one can use a numerical continuation method (Zhai et al., 2017) or a shooting method (Doedel, 1981) to obtain the period of a limit cycle as a function of changes in parameters. It would be much more effective, however, to find analytical relationships as they would reveal the specific characteristics of the self-oscillatory system, i.e., the maintenance entropy flux of the self-oscillatory structure. Mees and Chua (1979) have proposed a method to solve the period of the limit cycle analytically based on the frequency domain Hopf bifurcation theory.

The BZ reaction, however, is a multivariable system, and, when the method above is applied, tensor operations render the calculations tedious and impractical. Since the BZ reaction is a highly nonlinear system, higher order harmonics (Allwright, 1977) may also be needed to approximate the oscillatory behaviour accurately, which would complicate the calculation process further.

In the current study, we propose to use the Laplace-Borel (LB) transfer function representation to express the feedback system. The LB transform is an extension of the Laplace transform to the nonlinear polynomial terms by an infinite series of iterated integrals, and the transformed system obeys the shuffle algebraic operation (Batigun, et al., 1997). Similar to Mees and Chua (1979), we propose a closed-loop mechanism to compute the properties of the oscillation. The approximation procedure is progressive and the residuals of each expansion are used to identify the parameters of the harmonics.

2. THE GENERATION OF SELF-OSCILLATION

2.1 Hopf bifurcation Preliminaries

From bifurcation theory, Hopf bifurcation is the birth of a limit cycle from equilibrium in dynamic systems composed of a set of ordinary differential equations (ODEs), i.e., a Hopf point is the critical point where a small smooth change made to the bifurcation parameter(s) causes the system to change suddenly from an equilibrium point to a self-sustained oscillation. Hence, the Hopf point is viewed as a limit cycle where the forcing amplitude is zero. How the properties (amplitude and frequency) of the limit cycle will be varying with changes of the parameter is the focus of our study, but first, we need to locate the Hopf bifurcation point that causes the generation of the limit cycle.

The geometrical idea behind the Hopf bifurcation is seen by observing how the phase portrait of a 2-dimensional system might alter as the parameter u is varied.

$$\begin{cases} x_1' = f_1(x_1, x_2, u) = \alpha(u) x_1 + \omega(u) x_2 + O(|\mathbf{x}|^2) \\ x_2' = f_2(x_1, x_2, u) = -\omega(u) x_1 + \alpha(u) x_2 + O(|\mathbf{x}|^2) \end{cases} \quad (3)$$

where the equilibrium of the state vector $[x_1, x_2]^T$ is at the origin. We truncate the 3rd-order terms via Taylor's theorem and the eigenvalues are $\lambda_{1,2} = \alpha \pm \omega$ i. By a linear transformation, Eq. (3) has the topological structure equal to the normal form,

$$\begin{cases} y_1' = \alpha(u) y_1 - y_2 + y_1(y_1^2 + y_2^2) \\ y_2' = y_1 + \alpha(u) y_2 + y_2(y_1^2 + y_2^2) \end{cases} \quad (4)$$

where the 2nd-order terms disappear. Now, the eigenvalues are $\lambda_{1,2} = \alpha \pm$ i. Introducing the complex variable, $z = x_1 + i\, x_2$, and the bar on the variable representing the complex conjugate, then, Eq. (4) is given as,

$$z' = (\alpha + i) z - z|z|^2 \quad (5)$$

Finally, by using the exponential representation, $z = \rho e^{i\theta}$, the system is given in the *polar* form as follows,

$$\begin{cases} \rho' = \rho(\alpha - \rho^2) \\ \theta' = 1 \end{cases} \quad (6)$$

The phase portrait of Eq. (6) as α passes through zero can be analyzed: the first formula has equilibrium point $\rho = 0$ for all values of α. The sign of α decides the stability at the origin. For $\alpha > 0$, the origin is not stable, but there is additional equilibrium point $\rho_0(\alpha) = \text{sqrt}(\alpha)$, combining the second formula, which describes a rotation with constant speed, an isolated closed orbit is generated. Then $\alpha = 0$ becomes the critical point.

Therefore, a Hopf bifurcation gives birth to a limit cycle from an equilibrium in dynamic systems generated by ODEs, when the equilibrium changes stability via a pair of purely imaginary eigenvalues. Note that the stability of the limit

cycle needs to be tested by the Lyapunov function; for higher-dimensional systems, the central manifold theorem can be applied to reduce to 2nd-order ones, given that the Jacobian of the system only have a pair of purely imaginary eigenvalues, while the rest are negative.

2.2 Detection of the Hopf point

Exact detection of the Hopf bifurcation point with a varying parameter u can be problematic, especially when the system is high dimensional and highly nonlinear. The bi-alternate product manipulation gives a numerical detection criterion for existence of a pair of purely imaginary eigenvalues.

Proposition 1: The function

$$\varphi_H(x,u) = \det(2 f_x(x,u) \Theta I_n)$$

vanishes at a Hopf bifurcation point, where "det" represents the determinant, Θ is the bi-alternate product.

To prove *Proposition 1*, the following theorem is needed,

Lemma 1 (Stephanos theorem): If A is a $n \times n$ matrix and have eigenvalues $\mu_1, \mu_2 \ldots \mu_n$, then $2A\Theta I_n$ have satisfies $(\lambda_i+\lambda_j)_{n>i>j>1}$.

Proof. The eigenvalues of a matrix are preserved by similarity transforms as follows (Govaerts, 2000),

$$(P\Theta P)(2A\Theta I_n)(P\Theta P)^{-1} = 2B\Theta I_n$$

where, B is the similar transform of A, $B = PAP^{-1}$. Hence, we can assume that A is in upper triangular form (by reducing to the Jordan form if necessary). Then the eigenvalues of A are its diagonal elements. By applying the following Eq. (7) below for the case of upper triangular matrix,

$$(2A\Theta I_n)_{(i,j)(k,l)} = a_{ik} \begin{cases} -a_{il} & if\ k = j \\ -a_{ik} & if\ k \neq i\ and\ l = j \\ a_{ii} + a_{jj} & if\ k = i\ and\ l = j \\ a_{jl} & if\ k = i\ and\ l \neq j \\ -a_{jk} & if\ l = i \\ 0 & else \end{cases} \quad (7)$$

It is easy to find out that $2A\Theta I_n$ is an upper matrix, and its diagonal elements are given as *Lemma 1* claims. Therefore, *Proposition 1* is proved.

3. CLOSED-LOOP REPRESENTATION OF A LIMIT CYCLE

3.1 Feedback equivalent of the nonlinear system

The frequency domain analysis of the Hopf bifurcation is based on the closed-loop representation of the nonlinear system. Suppose there is an n-dimensional autonomous system represented by a set of general differential equations, comprising a dynamical linear part and a memoryless nonlinear part $\mathbf{g}$,

$$\mathbf{x}' = A\mathbf{x} + B\mathbf{g}(C\mathbf{x}, \mu) \quad (8)$$

where A is an $n \times n$ matrix, B is an $n \times l$ matrix, C is an $m \times n$ matrix, and $\mathbf{g}$: $R^m \rightarrow R^l$. μ is the varying parameter which may appear in A, B and C. Equation (8) can be represented as a multi-loop feedback system, as shown in Fig. 3. Introducing an arbitrary $D \in R^{l \times m}$, then, Eq. (8) is rewritten as follows,

$$\mathbf{x}' = A\mathbf{x} + BDy + B[\mathbf{g}(C\mathbf{x}, \mu) - Dy] \quad (9)$$

where $\mathbf{y} = C\mathbf{x}$. We take the Laplace transform L of Eq. (9),

$$L\mathbf{e}(s) = -G(s,\mu) L\mathbf{u}(s) \quad (10)$$

This leads to,

$$G(s,\mu) = C[sI - (A + BDC)]^{-1} B$$
$$\mathbf{u} = \mathbf{g}(C\mathbf{x}, \mu) - Dy$$
$$\mathbf{y} = -\mathbf{e} = C\mathbf{x}$$

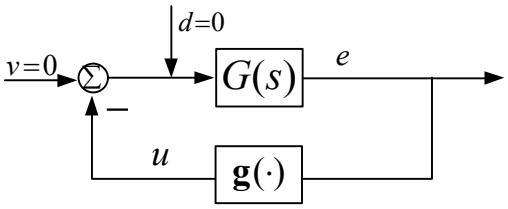

Fig. 3. The closed-loop equivalence of the dynamic system.

In practice, we choose $D = \mathbf{0}$ if $A \neq \mathbf{0}$, and $D = I_n$ if $A = \mathbf{0}$. The limit condition (s approaches zero) of the Laplace transform guarantees that the system has an equilibrium point $\hat{\mathbf{e}}(\mu)$ in the frequency domain when the Laplace variable is $s = 0$.

$$G(0,\mu)\mathbf{g}(\hat{\mathbf{e}}, \mu) = -\hat{\mathbf{e}} \quad (11)$$

By linearizing the feedback path in Fig.3 about the equilibrium point, where $J = \partial\mathbf{g}/\partial\mathbf{e}$ is the Jacobian matrix, we can apply the generalized Nyquist criterion (MacFarlane, 1977) to study the stability of the equilibrium solutions of this linear system. Analogous to the single-loop Nyquist theory, which may generate a self-oscillation when the *characteristic locus* passes through point (-1, 0), the multi-loop system evaluates the characteristic polynomials given as follows,

$$\begin{aligned} &\det\left|\lambda I_m - G(s,\mu)J(\mu)\right| \\ &\quad = h(\lambda, s, \mu) \\ &\quad = \lambda^t + a_{t-1}(s,\mu)\lambda^{t-1} + \mathrm{L}\ + a_1(s,\mu)\lambda + a_0(s,\mu) \\ &\quad = 0; \qquad t = \min(l, m) \end{aligned} \quad (12)$$

From the Hopf theorem, if we are to detect a point $\mu = \mu_0$ that has a pair of eigenvalues of the linearized time domain equations crossing the imaginary axis, the characteristic locus needs to move through -1+i 0 at a unique frequency $\omega_0 \neq 0$, and Eq. (12) has an eigenvalue -1+i 0 at $\mu = \mu_0$.

$$\begin{aligned} &\mathrm{Re}[h(-1, i\omega, \mu)] \equiv 0 \\ &\mathrm{Im}[h(-1, i\omega, \mu)] \equiv 0 \end{aligned} \quad (13)$$

One can detect the Hopf point of the system by combining Eqs. (11) and (13), but method in subsection 2.2 is applied in

this study because of the complex algebra encountered in solving Eqs. (11) and (13).

3.2 Analytical solution the of the limit cycle

In this section, we use the describing function approach to analyze the steady-state oscillations in nonlinear systems, which is an approximate tool to estimate the limit cycle parameters. A self-oscillatory system can be reformulated as a feedback system as shown in Fig. 3. The Nyquist criterion provides the necessary condition for the closed-loop system to generate oscillatory outputs, which is clear in the graphical form, as shown in Fig. 4. The intersection point satisfies the following condition:

$$\begin{aligned} & y = G(i\omega)u \approx -G(i\omega)N(\theta)y \\ & \Rightarrow G(i\omega) = -1/N(\theta) \end{aligned} \tag{14}$$

where $N(\theta)$ is the amplitude correlated approximation of the nonlinear term $\mathbf{g}$, and $N(\theta)$ is identified by the harmonic balance method.

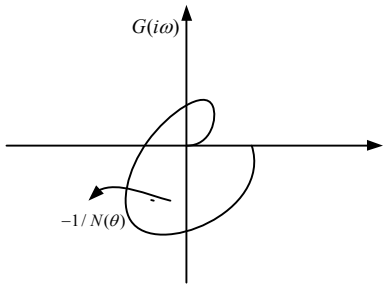

Fig. 4. The oscillation criterion on the Nyquist diagram.

Since the BZ reaction is a multivariable system, introducing harmonics will make the derivation quite tedious. Suppose a 2nd-harmonic has the form

$$\mathbf{e}(t) = \mathbf{\grave{e}} + \mathrm{Re}\sum_{k=0}^{2}\mathbf{E}^k \exp(ik\omega t) \tag{15}$$

Equating the input and output of the linear part gives

$$\mathbf{E}^k = -G(ik\omega)\mathbf{F}^k \tag{16}$$

where $\mathbf{E}^k$ are coefficients of the harmonics defined in Eq. (15) and $\mathbf{F}^k$ are the Fourier coefficients after the harmonics going through the memoryless nonlinear part $\mathbf{g}$.

By using the *tensor product operator*, Mees and Chua (1979) provided the relation of these two coefficients

$$\begin{aligned} \mathbf{F}^0 &\approx (D\mathbf{g})_{\grave{e}}\mathbf{E}^0 + \frac{1}{4}(D^2\mathbf{g})_{\grave{e}}\mathbf{E}^1 \otimes \bar{\mathbf{E}}^1 \\ \mathbf{F}^2 &\approx (D\mathbf{g})_{\grave{e}}\mathbf{E}^2 + \frac{1}{4}(D^2\mathbf{g})_{\grave{e}}\mathbf{E}^1 \otimes \mathbf{E}^1 \\ \mathbf{F}^1 &\approx (D\mathbf{g})_{\grave{e}}\mathbf{E}^1 + (D^2\mathbf{g})_{\grave{e}}[\mathbf{E}^0 \otimes \mathbf{E}^1 + \bar{\mathbf{E}}^1 \otimes \mathbf{E}^2] \\ &\quad + \frac{1}{8}(D^3\mathbf{g})_{\grave{e}}\mathbf{E}^1 \otimes \mathbf{E}^1 \otimes \bar{\mathbf{E}}^1 \end{aligned} \tag{17}$$

Substituting Eq. (16) to Eq. (17) and only retaining the $[1\omega]$ terms, the following relation is obtained

$$\begin{aligned} & [G(i\omega)J + 1]\mathbf{E}^1 = -G(i\omega)\mathbf{p}(\omega, \mathbf{E}^1) \\ & where: \end{aligned} \tag{18}$$

$$\mathbf{p} = (D^2\mathbf{g})_{\grave{e}}[\mathbf{E}^1 \otimes \mathbf{E}^0 + \frac{1}{2}\bar{\mathbf{E}}^1 \otimes \mathbf{E}^2] + \frac{1}{8}(D^3\mathbf{g})_{\grave{e}}\mathbf{E}^1 \otimes \mathbf{E}^1 \otimes \bar{\mathbf{E}}^1$$

By introducing the eigenvalue and the eigenvector, the nonlinear term $N(\theta)$ can be approximated to 2nd-order $1+\theta^2\zeta(\omega)$,

$$\overset{1}{\lambda}(i\omega) = -1 - \theta^2\mathbf{u}_1^T G(i\omega)\mathbf{p} \tag{19}$$

where $\overset{1}{\lambda}(i\omega)$ is the eigenvalue of Eq. (10) nearest to (-1+0*i*), and $\mathbf{u}$ is its left eigenvector. *A* is the amplitude.

3.3 Laplace-Borel representation of the close-loop

The solution method provided in subsection 3.2 requires extensive computing and is not straightforward. In practice, the frequency is computed by the following equation,

$$\mathrm{Re}[\overset{1}{\lambda}(i\omega)+1]\mathrm{Im}[\mathbf{u}_1^T G(i\omega)\mathbf{p}] - \mathrm{Re}[\mathbf{u}_1^T G(i\omega)\mathbf{p}]\mathrm{Im}[\overset{1}{\lambda}(i\omega)] = 0 \tag{20}$$

which could not be calculated directly, hence, a search algorithm is needed. Then, the amplitude is computed by the following equation

$$\theta^2 = -\frac{\mathrm{Re}[\overset{1}{\lambda}(i\omega)]+1}{\mathrm{Re}[\mathbf{u}_1^T G(i\omega)\mathbf{p}]} = -\frac{\mathrm{Im}[\overset{1}{\lambda}(i\omega)]}{\mathrm{Im}[\mathbf{u}_1^T G(i\omega)\mathbf{p}]} \tag{21}$$

The introduction of the vector representation in Eq. (17) for the multi-loop system is not easy to understand, and Eq. (17) may truncate out nonlinear polynomials higher than 3rd-order terms. Therefore, in this study, we propose to use the Laplace-Borel (LB) transfer function representation to express the closed-loop system. The LB transform is an extension of the Laplace transform to the nonlinear polynomial terms by an infinite series of iterated integrals, and the transformed system obeys the shuffle algebraic operation.

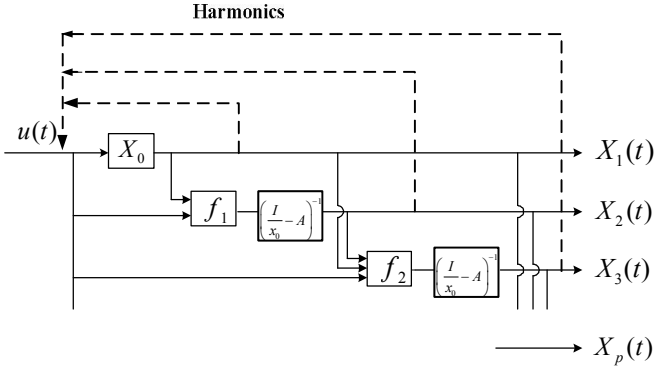

Fig. 5. Block diagram for the numerical computation of functional expansions.

The nonlinear system represented in Eq. (8) can be shifted to the x_0 domain by using the LB transform as follows,

$$X = X_0 + X_N$$
$$X_0 = \left(\frac{I}{x_0} - A\right)^{-1} bU \tag{22}$$
$$X_N = \left(\frac{I}{x_0} - A\right)^{-1} F(X, U, \text{C})$$

where x_0 is the transform operator, Also, Ш is the shuffle operator, X_0 and X_N are the linear and nonlinear parts of the system, respectively. The functional expansion (FEx) method approximates the analytical solution of the system which can be presented graphically as in Fig. 5. However, for a specific order of expansion $X_p(t)$, the approximated solution by the FEx method does not guarantee the solution to reach a closed cycle as time approaches infinity.

Similar to Fig. 3, we propose to close the system by adding the dashed lines to Fig. 4, and set $u(t) = 0$. By utilizing the shuffle algebra, each variable of the nonlinear part f_i (ith expansion) could be decoupled if the order of the harmonics is specified, which is especially attractive for multivariable systems. The approximation procedure is progressive and the residuals of each expansion are used to identify the parameters of the harmonics. Given that (1) the nonlinear blocks in Fig. 5 may introduce higher order harmonics than the corresponding inputs and (2) the output order of the harmonics is predictable if the structure of the system is given, the equation for $N(\theta)$ is explicitly known and the parameters of $N(\theta)$ can be identified by setting higher order residuals to zero. This method makes the calculation of $N(\theta)$ flexible for different systems and expendable to higher order approximations.

4. RESULTS AND DISCUSSION

Hopf point of the BZ model is detected using the criterion provided in *Proposition 1*. Since f in Eq. (2) is adjustable, numerical bifurcation analysis is implemented as shown in Fig. 6. Two Hopf points are detected, H1 is obtained when f = 0.522614, the equilibrium is (0.479129 0.521743 0.479129), and ω_1 = 3.4486; H2 is obtained when f = 2.253903, the equilibrium is (0.002071 1.625916 0.002071), and ω_1 = 2.60962.

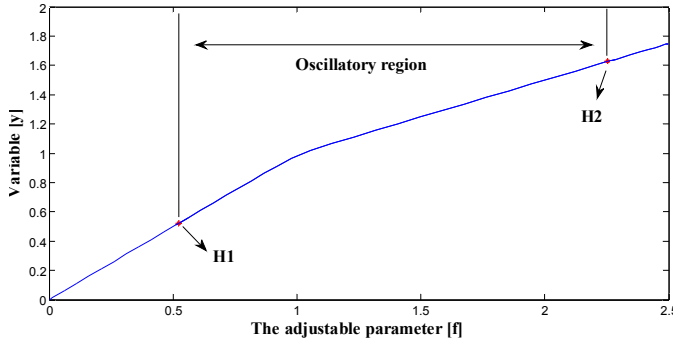

Fig. 6. Numerical continuation and bifurcation of the BZ reaction with varying f.

LB transforms of Eq. (2) and eliminating z gives the following formula, where the initial point is arbitrarily set as (0, 0, 0) because when the steady-periodic solution is concerned, dynamic transients led by the initial setting is neglected (Harris and Palazoglu, 1998):

$$\begin{cases} X = \frac{a_1}{\Delta}(-U_1) + \frac{b_1}{\Delta}(-U_2) = Z \\ Y = \frac{a_2}{\Delta}(-U_1) + \frac{b_2}{\Delta}(-U_2) \\ U_1 = \frac{1}{\varepsilon} X \text{C} X + \frac{1}{\varepsilon} X \text{C} Y \\ U_2 = \frac{1}{\delta} X \text{C} Y \end{cases} \tag{23}$$

where

$$\Delta = -\delta\varepsilon + (\delta - q\varepsilon - \delta\varepsilon)x_0 + (q + \delta - q\varepsilon)x_0^2 + (1+f)qx_0^3$$
$$a_1 = \delta\varepsilon x_0 + (q\varepsilon + \delta\varepsilon)x_0^2 + q\varepsilon x_0^3$$
$$b_1 = q\varepsilon x_0^2 + q\varepsilon x_0^3$$
$$a_2 = f\varepsilon x_0^3$$
$$b_2 = \delta\varepsilon x_0 - (\delta - \delta\varepsilon)x_0^2 - \delta x_0^3$$

Here X , Y and Z are the transform form of x , y and z, respectively, and $[U_1, U_2]^T$ is the transform of the input u in Fig. 3. The vector $\mathbf{e} = [X, Y]^T$ is defined as the harmonics similar to Eq. (15). Only 1st-order harmonics are identified for simplicity,

$$\begin{cases} X = e_1^0 + e_1^1(1 - i\omega x_0)^{-1} + \bar{e}_1^1(1 + i\omega x_0)^{-1} \\ Y = e_2^0 + e_2^1(1 - i\omega x_0)^{-1} + \bar{e}_2^1(1 + i\omega x_0)^{-1} \end{cases} \tag{24}$$

We caution that the offsets of X and Y are not equal to the equilibrium since a symmetry-breaking bifurcation may take place in this nonlinear system. The computation of the nonlinear part is implemented by the distributive property of the *shuffle product* operation,

$$\begin{aligned} &[F_1(x_0) + F_2(x_0)] \text{C} [F_3(x_0) + F_4(x_0)] \\ &= F_1(x_0) \text{C} F_3(x_0) + F_1(x_0) \text{C} F_4(x_0) \\ &\quad + F_2(x_0) \text{C} F_3(x_0) + F_2(x_0) \text{C} F_4(x_0) \end{aligned} \tag{25}$$

Substituting Eq. (24) into Eq. (23), $[U_1, U_2]^T$ is obtained,

$$U_1 = \frac{1}{\varepsilon}\left(\begin{aligned} &e_1^0 e_1^0 + 2e_1^1\bar{e}_1^1 + e_1^0 e_2^0 + e_1^1\bar{e}_2^1 + e_2^1\bar{e}_1^1 \\ &+ \frac{e_1^0 e_2^1 + e_2^0 e_1^1 + 2e_1^0 e_1^1}{1 - i\omega x_0} + \frac{e_1^0\bar{e}_2^1 + e_2^0\bar{e}_1^1 + 2e_1^0\bar{e}_1^1}{1 + i\omega x_0} \\ &+ \frac{e_1^1 e_2^1 + e_1^1 e_1^1}{1 - 2i\omega x_0} + \frac{\bar{e}_1^1\bar{e}_2^1 + \bar{e}_1^1\bar{e}_1^1}{1 + 2i\omega x_0} \end{aligned} \right) \tag{26}$$

$$U_2 = \frac{1}{\delta}\left(\begin{aligned} &e_1^0 e_2^0 + e_1^1\bar{e}_2^1 + e_2^1\bar{e}_1^1 \\ &+ \frac{e_1^0 e_2^1 + e_2^0 e_1^1}{1 - i\omega x_0} + \frac{e_1^0\bar{e}_2^1 + e_2^0\bar{e}_1^1}{1 + i\omega x_0} \\ &+ \frac{e_1^1 e_2^1}{1 - 2i\omega x_0} + \frac{\bar{e}_1^1\bar{e}_2^1}{1 + 2i\omega x_0} \end{aligned} \right)$$

The $[2\omega]$ are neglected, and $[U_1, U_2]^T$ is of the form of 1st-order harmonics, which is operated on by the transfer function $G(x_0 = 1/s)$. By neglecting the transient dynamics,

we can identify the coefficients **e** by comparing the outputs to $[X, Y]^T$ defined in Eq. (24). A harmonic pass through G will produce an amplification and an angle shift as time approaches infinity, while preserving the shape of the oscillation, taking $f = 0.53$ as example,

$$X = \frac{a_1}{\Delta}U_1 + \frac{b_1}{\Delta}U_2$$
$$Y = \frac{a_2}{\Delta}U_1 + \frac{b_2}{\Delta}U_2 \tag{27}$$

Combining Eq. (27) and Eq. (24) and truncating $[2\omega]$ and higher terms, we obtain the following relations

$$\begin{cases}
(1+f)e_1^0 = e_1^0e_1^0 + 2e_1^1\bar{e}_1^1 + 2e_1^0e_2^0 + 2e_1^1\bar{e}_2^1 + 2e_2^1\bar{e}_1^1 \\
(1+f)e_1^1 = e_1^0e_2^1 + e_2^0e_1^1 + 2e_1^0e_1^1 + e_1^0e_2^1 + e_2^0e_1^1 \\
(1+f)\bar{e}_1^1 = e_1^0\bar{e}_2^1 + e_2^0\bar{e}_1^1 + 2e_1^0\bar{e}_1^1 + e_1^0\bar{e}_2^1 + e_2^0\bar{e}_1^1 \\
e_2^0 = \dfrac{e_1^0e_1^0 + 2e_1^1\bar{e}_1^1 + e_1^0e_2^0 + e_1^1\bar{e}_2^1 + e_2^1\bar{e}_1^1}{(1+f)q/f} - \dfrac{e_1^0e_2^0 + e_1^1\bar{e}_2^1 + e_2^1\bar{e}_1^1}{(1+f)q} \\
e_2^1 = \dfrac{e_1^0e_2^0 + e_2^0e_1^1 + 2e_1^0e_1^1}{(1+f)q/f} - \dfrac{e_1^0e_2^1 + e_2^0e_1^1}{(1+f)q} \\
\bar{e}_2^1 = \dfrac{e_1^0\bar{e}_2^0 + e_2^0\bar{e}_1^1 + 2e_1^0\bar{e}_1^1}{(1+f)q/f} - \dfrac{e_1^0\bar{e}_2^1 + e_2^0\bar{e}_1^1}{(1+f)q}
\end{cases} \tag{28}$$

By the following transform relation,

$$\eta_1\cos(\omega t) + \eta_2\sin(\omega t) = \eta_-(1 - i\omega x_0)^{-1} + \eta_+(1 + i\omega x_0)^{-1}$$
$$where \qquad \eta_\pm = \frac{\eta_1 \pm i\eta_2}{2}$$

the 1st-order harmonics are given in the following explicit form,

$$\begin{cases} x = x_0 + A_1\cos(\omega t) + B_1\sin(\omega t) \\ y = y_0 + A_2\cos(\omega t) + B_2\sin(\omega t) \end{cases} \tag{29}$$

and numerical solution of the 6 nonlinear equations yields the coefficients, $x_0 = 0.23461$, $A_1 = -0.16772$, $B_1 = -0.00325$; $y_0 = 0.64100$, $A_2 = 0.00791$, $B_2 = -0.00153$. The equilibrium of (x, y) is (0.472147, 0.528749).

As suggested by Mees and Chua (1979), the frequency of the harmonics is computed by Eq. (19), where $[1\omega]$ terms are examined and the residual is proved to be $O(\theta^3)$.

As computing Eq. (20) to obtain ω is tedious, we will follow the iterative steps: (1) compute $\lambda(i\omega) = -1$ to get the initial guess $\omega_0 \in R^+$; (2) substitute ω_0 to $\mathbf{u}^T\ G\ (i\ \omega)\mathbf{p}$ and ω_1 is obtained by computing Eq. (20), (3) repeat step (2) till ω_i is convergent. Hence, the frequency is computed for $f = 0.53$ as 0.9053.

It is rather obvious that a similar formula can be obtained by minimizing the residual of Laplace-Borel expansion to higher order harmonics, which will be used to identify ω, and this is the next challenge.

5. CONCLUSIONS

In this paper, the time domain as well as frequency domain Hopf bifurcation theory are reviewed and implemented in the analysis of the oscillatory BZ reaction model. This paper also studied solution of the oscillator based on a closed-loop mechanism, and Laplace-Borel transform is introduced to identify the coefficients of the multi-dimensional harmonics.

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