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LETTER TO THE EDITOR

Diffusive instability near Hopf bifurcation for exponentially autocatalyzed reaction–diffusion system

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Abstract. The analysis of an exponentially autocatalyzed reaction–diffusion system near the Hopf bifurcation point has been carried out using a reductive perturbation approach to obtain a description in terms of the Ginzburg–Landau equation. The conditions for the occurrence of instability, in the presence and absence of diffusion, leading to Hopf bifurcation are also derived. The nature of the governing equations leads to multi-valued instability conditions and eventually results in more than one region in parameter space where instability of uniform oscillations due to diffusion is possible.

In this letter we shall consider an alternative form of autocatalysis where the product affects its own rate of formation through interactions with the rate constant in contrast to the normal autocatalysis where the rate is affected directly by the concentration of the product. The exponential autocatalysis has received acceptance as a general model for class of reaction–diffusion systems (Bar-Eli 1984a, b, c and 1985). The reaction scheme has been extensively analysed (i) to identify the regions of multiplicity and oscillations (Ravi Kumar *et al* 1984), (ii) to establish bounds on the steady-state solutions (Inamdar and Kulkarni 1990a), and (iii) for the occurrence of dissipative structures (Inamdar and Kulkarni 1990b, c). In this letter we shall reduce the reaction–diffusion equations to the form of a Ginzburg–Landau equation and thus open the reaction scheme to analysis in terms of features such as pattern formation, occurrence of chemical turbulence, existence of rotating and spiral waves and description in terms of nonlinear phase diffusion equations. The transcendental nature of the governing equations give rise to some new features such as the multiplicity of critical wavenumbers and more than one way of satisfying the different types of instability criteria that are not found in the conventional analysis of model systems.

This letter begins by presenting the results of linear stability analysis where the conditions for the occurrence of different types of instabilities leading to Hopf bifurcation are identified. The reaction–diffusion equations are then reduced to a generalized Ginzburg–Landau equation form using the reductive perturbation technique (Taniuti *et al* 1968, Taniuti 1974, Newell and Whitehead 1969, Kuramoto 1983). The results of reductive perturbation are then analysed and discussed.

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The reaction-diffusion equations of the exponentially autocatalyzed scheme are given as:

$$\frac{\partial x}{\partial t} = D_1 \frac{\partial^2 x}{\partial r^2} + x_0 - x - Da_1 x \exp(\alpha y) \tag{1a}$$

$$\frac{\partial y}{\partial t} = D_2 \frac{\partial^2 y}{\partial r^2} + y_0 - y + Da_1 x \exp(\alpha y) - Da_2 y. \tag{1b}$$

The homogeneous solutions of the system in (1a) and (1b) come out as

$$\exp(\alpha\theta) = \frac{x_0 - x_s}{x_s Da_1} \tag{2}$$

$$\theta = \frac{x_0 + y_0 - x_s}{(1 + Da_2)} \tag{3}$$

where x_s, θ are the steady state values of x and y respectively.

Defining the deviations as $x = u + x_s$, and $y = v + \theta$, the system of (1a) and (1b) along with (2), (3) reduces to the following equations after linearization of the nonlinear term $\exp(\alpha v) = (1 + \alpha v)$:

$$\frac{\partial u}{\partial t} = D_1 \frac{\partial^2 u}{\partial r^2} - (1 + Da_1\, e^{\alpha\theta})u - (\alpha Da_1 x_s\, e^{\alpha\theta})v - \alpha Da_1\, e^{\alpha\theta} uv \tag{4a}$$

$$\frac{\partial v}{\partial t} = D_2 \frac{\partial^2 v}{\partial r^2} + Da_1\, e^{\alpha\theta} u + (\alpha Da_1 x_s\, e^{\alpha\theta} - (1 + Da_2)v + \alpha Da_1\, e^{\alpha\theta} uv. \tag{4b}$$

Assuming that the deviations u and v follow the relation

$$u, v \propto \exp(\mathrm{i}\bar{q}r + \lambda t). \tag{5}$$

Equation (4a) and (4b) can be rewritten in terms of a linear matrix differential operator as

$$\frac{\partial}{\partial t}\begin{pmatrix} u \\ v \end{pmatrix} = \begin{pmatrix} -D_1 q^2 - (1 + Da_1\, e^{\alpha\theta}) & -\alpha Da_1 x_s\, e^{\alpha\theta} \\ Da_1\, e^{\alpha\theta} & -D_2 q^2 + \alpha x_s Da_1\, e^{\alpha\theta} - (1 + Da_2) \end{pmatrix}\begin{pmatrix} u \\ v \end{pmatrix} + \begin{pmatrix} -\alpha Da_1\, e^{\alpha\theta} uv \\ \alpha Da_1\, e^{\alpha\theta} uv \end{pmatrix}. \tag{6}$$

The characteristic polynomial can then be expressed as

$$\lambda^2 + \kappa(q)\lambda + \beta(q) = 0 \tag{7}$$

where

$$\kappa(q) = q^2(D_1 + D_2) + (Da_2 + 2) + Da_1\, e^{\alpha\theta}(1 - \alpha x_s) \tag{8a}$$

$$\beta(q) = D_1 D_2 q^4 - q^2\{D_1[\alpha x_s Da_1\, e^{\alpha\theta} - (1 + Da_2)] - D_2(1 + Da_1\, e^{\alpha\theta})\} + (1 + Da_2) - \alpha x_s Da_1\, e^{\alpha\theta} + (1 + Da_2) Da_1\, e^{\alpha\theta}. \tag{8b}$$

The condition that $\kappa(q)$ and $\beta(q)$ are non-negative for all q assures the stability of the steady state (x_s, θ). This stability condition can be violated in two different ways.

(i) Both $\kappa(q)$ and $\beta(q)$ remain positive for all q, except some where $\kappa(q) = 0$. This is referred to as type I instability and implies that,

$$\kappa(q) = 0 \qquad \text{with} \qquad q = 0. \tag{9}$$

This results in an equation for the critical value of the bifurcating parameter y_{0c} as

$$y_{0c}=(x_s-x_0)+\frac{(1+Da_2)}{\alpha}\ln\left(\frac{Da_2+2}{Da_1(\alpha x_s-1)}\right). \tag{10}$$

The logarithmic term in (10) is subject to the constraint

$$x_s>\frac{1}{\alpha}. \tag{11}$$

(ii) It is also possible that both $\kappa(q)$ and $\beta(q)$ remain positive for all q except where $\beta(q)$ vanishes. This is referred to as type II instability can be expressed as

$$\beta(q_c)=0 \qquad \text{and} \qquad \frac{\partial\beta(q_c)}{\partial(q_c)}=0 \tag{12}$$

and gives the following expressions for critical wavenumber q_c and y'_{0c}.

The critical wavenumber q_c is

$$\begin{aligned} q_c^2=&\frac{D_1[\alpha x_s Da_1\,\mathrm{e}^{\alpha\theta}-(1+Da_2)]-D_2(1+Da_1\,\mathrm{e}^{\alpha\theta})}{2D_1D_2}\\ &\pm\frac{1}{2D_1D_2}(\{D_1[\alpha x_s Da_1\,\mathrm{e}^{\alpha\theta}-(1+Da_2)]-D_2(1+Da_1\,\mathrm{e}^{\alpha\theta})\}^2\\ &-4D_1D_2[(1+Da_2)(1+Da_1\,\mathrm{e}^{\alpha\theta})-\alpha x_s Da_1\,\mathrm{e}^{\alpha\theta}])^{1/2} \end{aligned} \tag{13}$$

and the critical value y'_{0c} is

$$y'_{0c}=(x_s-x_0)+\frac{(1+Da_2)}{\alpha}\ln\left(\frac{z}{Da_1}\right) \tag{14}$$

where z is

$$\begin{aligned} z&=Da_1\,\mathrm{e}^{\alpha\theta}\\ &=\frac{-b'\pm\sqrt{b'^2-4a'c'}}{2a'} \end{aligned} \tag{15}$$

and the various constants are

$$a'=\frac{D_1}{D_2}(\alpha x_s)^2+\frac{D_2}{D_1}-2\alpha x_s \tag{16a}$$

$$b'=2\left(\frac{D_2}{D_1}-\frac{D_1}{D_2}\alpha x_s(1+Da_2)+(\alpha x_s-Da_2-1)\right) \tag{16b}$$

$$c'=\frac{D_1}{D_2}(1+Da_2)^2+\frac{D_2}{D_1}-2(1+Da_2). \tag{16c}$$

We have thus obtained the conditions ((10) and (14)) for the occurrence of type I and type II instability. Equations (10) and (14) are plotted in figure 1 where parametric maps of x_0 against y_0 are presented for a defined set of other parameters. The cases for $D_1=D_2$ and $D_1>D_2$ are shown separately. It is interesting to note that the transcendental nature of the governing equations gives rise to a set of two values for the critical concentration y_{0c}. Clearly the requirement that $\kappa(q)$ be positive for all q for the stability of the system requires that $y_0>y_{0c}$. The region below the line y_{0c}

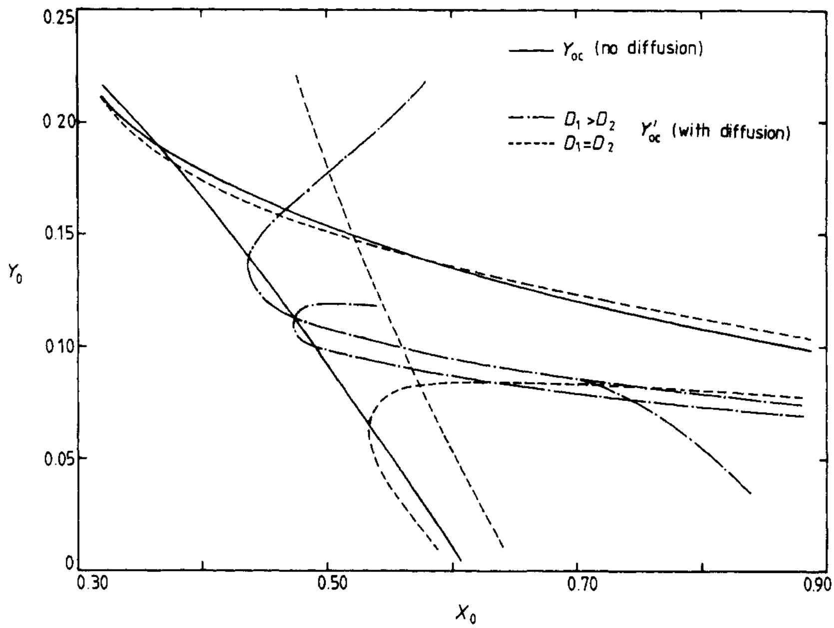

Figure 1. Instability criteria in the presence and absence of diffusion.

therefore shows type I instability. The choice of curve is decided by the steady state x_s and θ to which the system evolves, which in turn depends on the initial conditions specified. Of more interest, perhaps, is the condition (14) which shows a set of four possible values for the critical concentration y'_{0c} in a certain region. We can see in figure 1 that the two critical curves cut and cross each other at several places. In order that diffusion plays an important role we impose the condition $y_{0c} < y'_{0c}$ so that type II instability does not occur earlier than type I instability. The multi-valued nature of these criteria gives rise to interesting possibilities for satisfying this condition in many different ways.

Following the general procedure outlined in Kuramoto (1983), we shall now reduce the reaction–diffusion equation using reductive perturbation technique following the specific form of the Ginzburg–Landau equation.

$$\frac{\partial W}{\partial t} = (1+ic_0)W + (1+ic_i)\nabla_x^2 W - (1+ic_2)|W|^2 W \tag{17}$$

where

$$c_0 = \frac{\alpha x_s(B^2+A^2) + A(\alpha x_s+1)+1}{B(1-\alpha x_s)} \tag{18a}$$

$$c_1 = \frac{B(\gamma^2-1)}{A(\gamma^2+1)} \tag{18b}$$

$$c_2 = \frac{\Omega_2(A+1)+B\Omega_3}{B\Omega_2-\Omega_3(A+1)} \tag{18c}$$

and the other quantities are defined as

$$A = \frac{-(\alpha x_s + Da_2+1)}{\alpha x_s(Da_2+2)} \qquad B = \frac{\alpha x_s - (1+Da_2)^2}{\omega_0 \alpha x_s(Da_2+2)} \tag{19}$$

$$\Omega_1 = (\alpha x_s - 1)[(1+Da_2)^2 - \alpha x_s] \tag{20a}$$

$$\begin{aligned}\Omega_2 = (\Omega_1 - 4\omega_0^2)[2A(\alpha x_s - 1) - 2A^2(\alpha x_s - 1)^2(1+Da_2)] \\ + \Omega_1\{A[(\alpha x_s - 1) - A(\alpha x_s - 1)^2(1+Da_2) - 2B\omega_0] \\ - B(2\omega_0 + B(\alpha x_s - 1)^2(1+Da_2) - 2A\omega_0]\}\end{aligned} \tag{20b}$$

$$\begin{aligned}\Omega_3 = (\Omega_1 - 4\omega_0^2)[-2AB(1-\alpha x_s)^2(1+Da_2)] \\ + \Omega_1\{B[(\alpha x_s - 1) - A(\alpha x_s - 1)^2(1+Da_2) - 2B\omega_0] \\ + A[2\omega_0 + B(\alpha x_s - 1)^2(1+Da_2) - 2A\omega_0]\}.\end{aligned} \tag{20c}$$

The constants c_0, c_1 and c_2, as obtained above, define the Ginzburg–Landau equation for the exponentially autocatalyzed reaction-diffusion system. Evaluation of these constants and therefore of the Ginzburg–Landau equation is central to subsequent development such as obtaining the plane waves, rotating waves, turbulence and entrainment phenomena in discrete oscillators. The quantity $\beta(=1+c_1c_2)$, defined in terms of the constants appearing in the Ginzburg–Landau equation, is an important parameter of the uniform oscillation stability, such that if $\beta > 0$ implies stability and vice versa. However, the stability of uniform limit cycle oscillation is not guaranteed for an infinitely large system size.

In order to ensure that type II instability does not occur earlier than type I instability as the value of y_0 increases, we impose the condition $y_{0c} < y'_{0c}$, in terms of $\gamma[=(D_1/D_2)^{1/2}]$ as

$$F(\gamma) \equiv (A\gamma^4) + (B\gamma^2) + C = 0 \tag{21}$$

where

$$A = \left(\frac{\alpha x_s(Da_2+2)}{(\alpha x_s - 1)} - (1+Da_2)\right)^2 \tag{22a}$$

$$B = 2\left(\frac{(\alpha x_s - Da_2 - 1)(Da_2+2)}{\alpha x_s - 1} - \frac{\alpha x_s(Da_2+2)^2}{(\alpha x_s - 1)^2} - (1+Da_2)\right) \tag{22b}$$

$$C = \left(\frac{Da_2+2}{\alpha x_s - 1} + 1\right)^2. \tag{22c}$$

In the present instance, the parameter β defining the stability of the plane waves is used along with the condition describing the occurrence of type I instability prior to type II instability, $y_{0c} < y'_{0c}$, and the results are plotted in the form of γ against x_0 in figures (2*a*) and (2*b*). The conditions $\beta < 0$ or $\beta > 0$ and $F(\gamma) > 0$ and $F(\gamma) < 0$ are marked along the curves.

We note that for a given set of parameter values such as Da_1, Da_2, D_1, D_2 and α etc, (10) and (14) give rise to multiple values for y_{0c} and y'_{0c} and therefore various different possibilities by satisfying the requirement of $y_0 < y'_{0c}$. Two such possibilities are presented as figures (2*a*) and (2*b*). In both situations the conditions $\beta < 0$ for stability and $F(\gamma) > 0$ can be realized. The region $F(\gamma) > 0$ suggests that the onset of spatially uniform oscillations precede the spatially non-uniform ones. Furthermore if $\beta < 0$ then these oscillations are unstable. The exponentially autocatalyzed reaction-diffusion system thus shows the multiple existence of an instability condition of uniform oscillations due to diffusion.

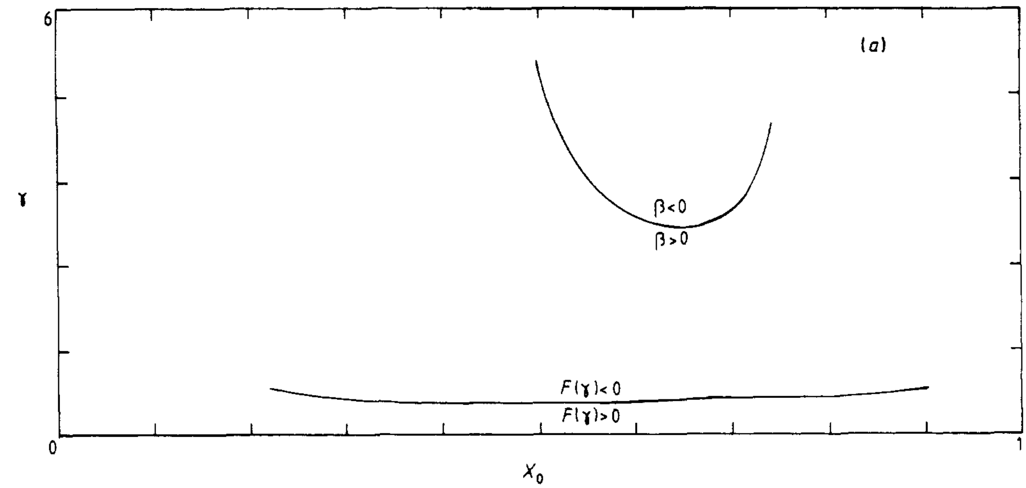

Figure 2a. Region in parameter space for uniform oscillations to become unstable due to diffusion.

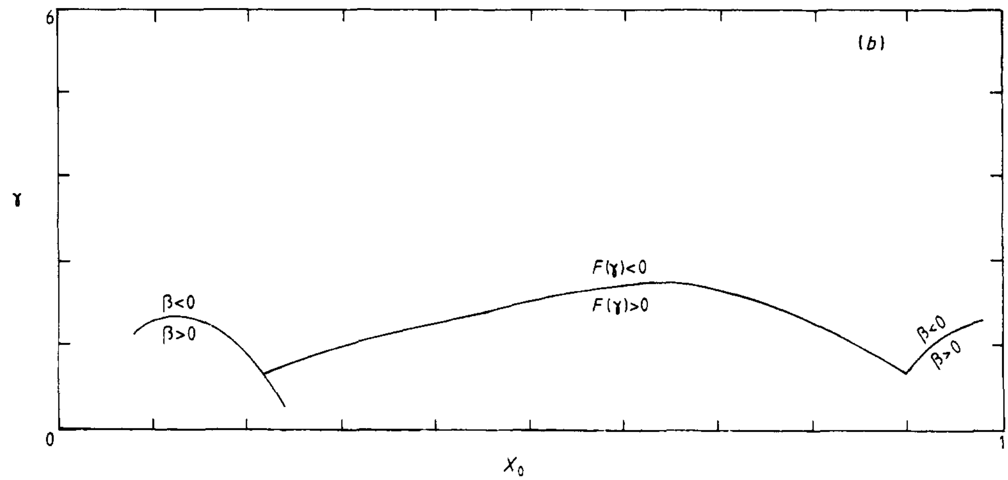

Figure 2b. Region in parameter space for uniform oscillations to become unstable due to diffusion.

To sum up, this letter derives the Ginzburg–Landau equation for the exponentially autocatalyzed reaction–diffusion system, and describes the behaviour of a system near the Hopf bifurcation analytically. The conditions derived explain in general the manner in which a phase transition in a reaction–diffusion system can occur.

References

Bar-Eli K 1984a *J. Phys. Chem.* **88** 3616
—— 1984b *Lett. Phys.* **119** 437
—— 1984c *J. Phys. Chem.* **88** 6174
Bar-Eli K and Reuveni S 1985 *J. Phys. Chem.* **89** 1329

Haken H 1975 *Z. Phys.* B **21** 105
Haken H 1983 *An Introduction: Nonequilibrium Phase Transitions and Self-Organisation in Physics, Chemistry and Biology* 3rd edn (Berlin: Springer)
Inamdar S R and Kulkarni B D 1990a *J. Phys. A: Math. Gen.* **23** L461
—— 1990b *Chem. Eng. J.* submitted
—— 1990c *Chem. Eng. J.* submitted
Kuramoto Y 1984 *Chemical Oscillations, Waves and Turbulence* (Berlin: Springer)
Newell A C and Whitehead J A 1969 *J. Fluid Mech.* **38** 279
Ravi Kumar V, Kulkarni B D and Doraiswamy L K 1984 *AIChE* **30** 649
Taniuti T and Wei C C 1968 *J. Phys. Soc. Japan* **24** 941
Taniuti T 1974 *Prog. Theor. Phys. Suppl.* **55** 1