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A model reaction diffusion system under spatial perturbation: theoretical and numerical investigation

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Abstract. We consider a model (Lengyel-Epstein) reaction-diffusion system under spatial parametric modulation and demonstrate the effect of resonance shift of the Hopf-Turing boundary. A systematic perturbative and numerical analysis shows that this shift may induce spatial inhomogeneity on an homogeneous stable state resulting in pattern formation.

PACS. 82.40.Ck Pattern formation in reactions with diffusion, flow and heat transfer – 47.54.+r Pattern selection; pattern formation – 87.10.+e General theory and mathematical aspects

1 Introduction

Pattern formation in two-dimensional reaction diffusion systems has been the subject of wide renewed interest over the last decade since the unambiguous experimental demonstration of Turing patterns in the early nineties [1–3]. Turing patterns affected by spatial inhomogeneities have been observed more recently in liquid crystals [4], catalytic surface reactions [5], porous media [6], biological self replication [7,8] and other systems [9]. The formation of these patterns are due to the interplay of self enhancement which causes local inhomogeneous growth and a long range inhibition restricting the effect of rapid diffusion. Turing patterns appear as symmetry breaking spatial structures arising when a difference in diffusion coefficients between the two species causes an initially homogeneous stable state to become unstable due to inhomogeneous perturbation. The homogeneous stable state often corresponds to an oscillatory state arising out of a Hopf bifurcation [10,11]. Hopf instability results in spatially homogeneous temporal oscillations and its relation to Turing instability is of great interest. Both instabilities have been observed experimentally in the case of the CIMA system through concentration variation of the colour indicator in the reactor [12,13]. The interaction between these instabilities [14] may take place either through a co-dimensional-two Turing-Hopf bifurcation, when the corresponding bifurcation parameter threshold values are equal [15,16] or due to different competing bifurcations of multiple stationary states [12–17]. A region of specific interest is the boundary between the associated Hopf (homogeneous oscillatory) region and the Turing (spatially periodic) region. The object of the present paper is to show that an external spatial parametric resonance on a reaction diffusion system may induce a transition from Hopf to Turing region leading to initiation of stable pattern formation. The spatial parametric modulations have been studied earlier in the context of convective instabilities in fluids [18–23] and quite extensively in a number of chemical [25–30] and optical [31] systems. Special attention has been paid to asymptotic states of Turing patterns as well as on growth dynamics. Among the chemical systems the photosensitivity of the CDIMA system offers excellent opportunity in this context to study the effect of spatiotemporal forcing which results in modulation and control of Turing pattern. Spatial forcing using illumination through gray-scaled masks [25,27] is also used for removing defects in order to produce ordered patterns [27]. Bearing in mind these experimental successes in forcing CDIMA/CIMA system, it may be pointed out that while the majority of the work focuses on controlling a developed Turing pattern by external forcing, it is worthwhile to explore how an external spatial forcing can create a pattern due to resonance in an otherwise homogeneous state. Secondly in contrast to these earlier works where the modulation is additive in nature we study the role of multiplicative forcing in the system. Thirdly, although the theoretical perturbative method has been determined previously [18] it was incomplete primarily in some ways. The Gierer-Meinhardt model and the variation of diffusion coefficient as analyzed to study resonance effect were theoretical and without any numerical support and are thus difficult to comprehend in an experimental situation. The parametric space chosen in the Lengyel-Epstein model in the present context is much more realistic and the efficiency of the perturbation method can be well-tested

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numerically as well as by experiment. The photosensitivity of the present chemical system can be utilized using illumination through masks with appropriate structure for periodic spatial modulations. With this end in view we carry out a systematic perturbation analysis of the problem to investigate the shift of the Hopf-Turing boundary due to a parametric resonance in a general perspective followed by a numerical simulation to corroborate the result of our perturbation analysis on Lengyel-Epstein model reaction-diffusion system [24].

2 A perturbative analysis of parametric resonance

To start with we consider a reaction diffusion system with activator and inhibitor characterized by their concentrations u and v, respectively as follows:

$$\frac{\partial u}{\partial t} = \nabla^2 u + f(u,v) \tag{1}$$

$$\frac{\partial v}{\partial t} = \sigma(d\nabla^2 v + bg(u,v)). \tag{2}$$

Here f and g are the reaction terms. d refers to the ratio of the diffusion coefficients of the two species $d = d_v/d_u$. σ is a scale parameter. b refers to a parameter to be modulated spatially in the dynamics.

For spatial modulation of b we consider $b = \bar{b_0}(1 + \epsilon \cos Kx)$ where ϵ is the amplitude of modulation around the spatial mean $\bar{b_0}$. K refers to the wave number of the modulation. ϵ is assumed to be small such that it can be treated as a perturbation parameter. Furthermore it is important to note that the appearance of b in the reaction term is such that it does not affect the homogeneous steady state u_0, v_0 of the reaction-diffusion system. Our aim here is to investigate the shift in the boundary for the formation of an inhomogeneous steady state. In the long time limit the time evolution of the spatial distribution leads to

$$\nabla^2 u + f(u,v) = 0 \tag{3}$$

$$d\nabla^2 v + bg(u,v) = 0. \tag{4}$$

With $u = u_0 + \delta u$ and $v = v_0 + \delta v$, where δu and δv are the perturbation around the steady state (u_0, v_0) we obtain

$$-\nabla^2 \delta u = f_u \delta u + f_v \delta v \tag{5}$$

$$-d\nabla^2 \delta v = b(g_u \delta u + g_v \delta v). \tag{6}$$

Here f_u, f_v and g_u, g_v terms correspond to partial derivatives of the functions f or g with respect to the variables u or v evaluated at steady state (e.g., $f_u = \frac{\partial f}{\partial u}|_{u=u_0}$). We now expand the perturbations δu, δv in series in ϵ along with $\bar{b_0}$ as $\delta u = \delta u_0 + \epsilon \delta u_1 + \epsilon^2 \delta u_2 + ...$, $\delta v = \delta v_0 + \epsilon \delta v_1 + \epsilon^2 \delta v_2 + ...$ and $\bar{b_0} = b_0 + \epsilon b_1 + \epsilon^2 b_2 + ...$ and put them in equations (5) and (6), obtaining after rearranging up to $O(\epsilon^2)$,

$$\begin{aligned} L_0 \begin{pmatrix} \delta u_0 \\ \delta v_0 \end{pmatrix} &+ \epsilon \left[L_0 \begin{pmatrix} \delta u_1 \\ \delta v_1 \end{pmatrix} \right. \\ &\left. - \begin{pmatrix} 0 \\ b_1 g_u \delta u_0 + b_1 g_v \delta v_0 + (g_u b_0 \delta u_0 + g_v b_0 \delta v_0)\cos Kx \end{pmatrix} \right] \\ &+ \epsilon^2 \left[L_0 \begin{pmatrix} \delta u_2 \\ \delta v_2 \end{pmatrix} \right. \\ &\left. - \begin{pmatrix} 0 \\ b_2 g_u \delta u_0 + b_2 g_v \delta v_0 + (g_u b_0 \delta u_1 + g_v b_0 \delta v_1)\cos Kx \end{pmatrix} \right] = 0 \end{aligned} \tag{7}$$

where

$$L_0 = \begin{pmatrix} -\nabla^2 - f_u & -f_v \\ -b_0 g_u & -d\nabla^2 - b_0 g_v \end{pmatrix}. \tag{8}$$

In the low order ($\epsilon = 0$) we have $L_0 \begin{pmatrix} \delta u_0 \\ \delta v_0 \end{pmatrix} = 0$, so that putting

$$\delta u_0 = \delta \bar{u}_0 \cos K_0 x \qquad \delta v_0 = \delta \bar{v}_0 \cos K_0 x \tag{9}$$

we obtain

$$({K_0}^2 - f_u)\delta \bar{u}_0 = f_v \delta \bar{v}_0 \tag{10}$$

and

$$-b_0 g_u \delta \bar{u}_0 + (d{K_0}^2 - b_0 g_v)\delta \bar{v}_0 = 0. \tag{11}$$

Equation (10) yields

$$\delta \bar{v}_0 = \frac{({K_0}^2 - f_u)}{f_v} \delta \bar{u}_0. \tag{12}$$

For the perturbation of the order $0(\epsilon)$ we have

$$L_0 \begin{pmatrix} \delta u_1 \\ \delta v_1 \end{pmatrix} = \begin{pmatrix} 0 \\ b_1 g_u \delta u_0 + b_1 g_v \delta v_0 + (g_u b_0 \delta u_0 + g_v b_0 \delta v_0)\cos Kx \end{pmatrix}. \tag{13}$$

Making use of (9) we rewrite (13) as

$$L_0 \begin{pmatrix} \delta u_1 \\ \delta v_1 \end{pmatrix} = (g_u \delta \bar{u}_0 + g_v \delta \bar{v}_0) \begin{pmatrix} 0 \\ b_1 \cos K_0 x + b_0 \cos K_0 x \cos Kx \end{pmatrix}. \tag{14}$$

From equation (14) it is clear that provided $K \neq 2K_0$ it would be solvable if $b_1 = 0$. For $K = 2K_0$ we have

$$L_0 \begin{pmatrix} \delta u_1 \\ \delta v_1 \end{pmatrix} = (g_u \delta \bar{u}_0 + g_v \delta \bar{v}_0) \times \begin{pmatrix} 0 \\ (b_1 + \frac{b_0}{2}) \cos (K - K_0)x + \frac{b_0}{2} \cos (K + K_0)x \end{pmatrix}. \tag{15}$$

Thus the removal of spurious resonances requires

$$b_1 = -\frac{b_0}{2}. \tag{16}$$

The boundary is thus shifted to $b_c = \bar{b}_0(1+\frac{\epsilon}{2})$ under resonance condition.

For $K \neq K_0$ we have to consider $b_1 = 0$. Therefore equation (14) reduces to

$$L_0\begin{pmatrix}\delta u_1\\ \delta v_1\end{pmatrix} = b_0\begin{pmatrix}0\\ (g_u\delta\bar{u}_0\cos K_0x + g_v\delta\bar{v}_0\cos K_0x)\cos Kx\end{pmatrix} \tag{17}$$

which using (12) can be rearranged further to obtain

$$L_0\begin{pmatrix}\delta u_1\\ \delta v_1\end{pmatrix} = b_0\begin{pmatrix}0\\ (g_u + \frac{(K_0{}^2-f_u)}{f_v}g_v)\delta\bar{u}_0\cos K_0x\cos Kx\end{pmatrix}. \tag{18}$$

Writing equation (18) more explicitly we have

$$(-\nabla^2 - f_u)\delta u_1 - f_v\delta v_1 = 0 \tag{19}$$

$$-b_0g_u\delta u_1 + (-d\nabla^2 - b_0g_v)\delta v_1 = \frac{\delta\bar{u}_0b_0}{2}\left(g_u + \frac{K_0{}^2-f_u}{f_v}g_v\right)(\cos k_+x + \cos k_-x) \tag{20}$$

where k_+ and k_- are given by $k_+ = K + K_0$ and $k_- = K - K_0$.

Assuming first order perturbation δu_1 and δv_1 to vary as δu_1 or $\delta v_1 \sim \cos k_+x$ we have from equations (19) and (20)

$$(k_+{}^2 - f_u)\delta u_1 - f_v\delta v_1 = 0 \tag{21}$$

$$-b_0g_u\delta u_1 + (dk_+{}^2 - b_0g_v)\delta v_1 = \frac{\delta\bar{u}_0b_0}{2}\left(g_u + \frac{K_0{}^2-f_u}{f_v}g_v\right)(\cos k_+x + \cos k_-x). \tag{22}$$

The above algebraic equations can be solved to yield for the contribution due to the k_+ term,

$$\delta u_1 = \frac{\delta\bar{u}_0b_0}{2}\frac{(f_vg_u + (K_0{}^2-f_u)g_v)}{\Delta_+}\cos k_+x \tag{23}$$

where $\Delta_+ = (k_+{}^2 - f_u)(dk_+{}^2 - b_0g_v) - b_0g_uf_v$.

For the actual solution of δu_1 we should incorporate both the k_+ and k_- terms so that we have

$$\delta u_1 = \frac{\delta\bar{u}_0b_0}{2}\left[f_vg_u + \left(K_0{}^2 - f_u\right)g_v\right]\left[\frac{\cos k_+x}{\Delta_+} + \frac{\cos k_-x}{\Delta_-}\right] \tag{24}$$

where $\Delta_- = (k_-{}^2 - f_u)(dk_-{}^2 - b_0g_v) - b_0g_uf_v$.

Similarly one obtains

$$\delta v_1 = \frac{\delta\bar{u}_0b_0}{2}\left[g_u + \frac{(K_0{}^2-f_u)}{f_v}g_v\right] \times\left[\frac{(k_+{}^2-f_u)\cos k_+x}{\Delta_+} + \frac{(k_-{}^2-f_u)\cos k_-x}{\Delta_-}\right]. \tag{25}$$

Having obtained the perturbative corrections up to $O(\epsilon)$ we look for the next order $O(\epsilon^2)$. To this end we start from

$$L_0\begin{pmatrix}\delta u_2\\ \delta v_2\end{pmatrix} = \begin{pmatrix}0\\ b_2g_u\delta u_0 + b_2g_v\delta v_0 + (g_ub_0\delta u_1 + g_vb_0\delta v_1)\cos Kx\end{pmatrix} + \text{non secular terms}. \tag{26}$$

Substituting the appropriate expressions for δu_0, δv_0, δu_1 and δv_1 in the secular part of the right hand side of the above equation we obtain the relevant non-zero element as

$$\begin{aligned} &-b_2\left[g_u\delta\bar{u}_0 + g_v\delta\bar{u}_0\frac{(K_0{}^2-f_u)}{f_v}\right]\cos K_0x \\ &-\frac{g_u}{2}b_0{}^2\cos Kx\delta\bar{u}_0\left[f_vg_u + \left(K_0{}^2-f_u\right)g_v\right] \times\left[\frac{\cos k_+x}{\Delta_+} + \frac{\cos k_-x}{\Delta_-}\right] \\ &-\frac{g_vb_0{}^2}{2f_v}\cos Kx\delta\bar{u}_0\left[f_vg_u + \left(K_0{}^2-f_u\right)g_v\right] \\ &\times\left[\frac{(k_+{}^2-f_u)\cos k_+x}{\Delta_+} + \frac{(k_-{}^2-f_u)\cos k_-x}{\Delta_-}\right]\ldots \end{aligned} \tag{27}$$

$\cos K_0x$ containing terms of the above expression give rise to the following relation

$$\begin{aligned} &-\frac{b_2}{f_v}\left(g_uf_v + g_v\left(K_0{}^2 - f_u\right)\right)\delta\bar{u}_0 \\ &-\frac{g_u}{4}b_0{}^2\delta\bar{u}_0\left[f_vg_u + \left(K_0{}^2-f_u\right)g_v\right]\left(\frac{1}{\Delta_+} + \frac{1}{\Delta_-}\right) \\ &-\frac{g_vb_0{}^2}{4f_v}\delta\bar{u}_0\left[f_vg_u + \left(K_0{}^2-f_u\right)g_v\right] \\ &\times\left[\frac{(k_+{}^2-f_u)}{\Delta_+} + \frac{(k_-{}^2-f_u)}{\Delta_-}\right] = 0. \end{aligned} \tag{28}$$

The above equation yields b_2 as

$$b_2 = -\frac{b_0{}^2}{4}\left[g_uf_v\left(\frac{1}{\Delta_+} + \frac{1}{\Delta_-}\right) + g_v\left(\frac{(k_+{}^2-f_u)}{\Delta_+} + \frac{(k_-{}^2-f_u)}{\Delta_-}\right)\right]. \tag{29}$$

The spatial modulation therefore shifts the boundary under parametric resonance $K = K_0$ as $b = b_0 + \epsilon^2b_2$ or explicitly we obtain

$$b = b_0\left[1 - \epsilon^2\frac{b_0}{4}\left[g_uf_v\left(\frac{1}{\Delta_+} + \frac{1}{\Delta_-}\right) + g_v\left(\frac{(k_+{}^2-f_u)}{\Delta_+} + \frac{(k_-{}^2-f_u)}{\Delta_-}\right)\right]\right]. \tag{30}$$

In what follows we now make use of the above relation to determine the boundary of instability leading to pattern formation under spatial parametric resonance.

3 A model system and numerical analysis

In order to illustrate the above perturbative analysis we now resort to a well known reaction-diffusion system, the Lengyl-Epstein model for chlorite-iodide-malonic acid reaction [24]. The typical equations in the two dimensional domain are given by

$$\frac{\partial u}{\partial t} = a - u - \frac{4uv}{1+u^2} + \frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} \tag{31}$$

$$\frac{\partial v}{\partial t} = \sigma b \left(u - \frac{uv}{1+u^2} \right) + \sigma d \left(\frac{\partial^2 v}{\partial x^2} + \frac{\partial^2 v}{\partial y^2} \right). \tag{32}$$

a, b and σ are dimensionless parameters containing kinetic parameters and initial concentrations of the reactants. The ratio of the diffusion coefficients d $(=D_{\mathrm{ClO_2}^-}/D_{\mathrm{I}^-})$ of the activator $(\mathrm{ClO_2}^-)$ and inhibitor (I^-) in the absence of the complexing agent starch is related to the effective ratio of the diffusion coefficients through σd. $u(x,y,t)$ and $v(x,y,t)$ are normalized dimensionless concentrations of the two reactants I^- and $\mathrm{ClO_2}^-$ respectively. The fixed point of the dynamical system is given by $u_{ss} = \frac{a}{5}$, $v_{ss} = 1 + \frac{a^2}{25}$. Thus the homogeneous steady state is independent of b, the critical parameter for studying parametric resonance in the present analysis. This well known model has been used widely for the study of pattern formation over a decade. We fix the experimentally admissible parameter values $a = 16.0$, $b = 1.5$ and $d = 1.5$ for our treatment. The other important parameter σ (which can be controlled experimentally by initial concentration of starch) the complexing agent in the Lengyl-Epstein model, plays an important part in determining the stability regions. In the absence of diffusion and spatial modulation the Hopf curve, below which one observes the stable oscillation, is given by

$$\sigma b = \frac{3a}{5} - \frac{25}{a}. \tag{33}$$

In Figure 1 we plot the Hopf curve (b vs. a) shown by dashed line for $\sigma = 5.6$. The reaction-diffusion system in the presence of diffusion on the other hand gives rise to Turing bifurcation in the absence of spatial modulation if

$$(3da^2 - 5ab - 125d)^2 = 100abd(25 + a^2). \tag{34}$$

This curve is shown as a solid line in Figure 1 and is independent of σ. The homogeneous stable steady state is unstable below this line. Thus as one increases σ the Hopf line shifts downwards gradually and once it crosses the Turing line, instability leading to formation of pattern arises. Spatial modulation of b under parametric resonance $K = K_0$ however leads to a situation where the applied perturbation shifts the Turing boundary line above the Hopf curve according to equation (30) (for the same σ value for which no such instability arises in absence of modulation). This results in a condition for pattern formation under parametric resonance with $\epsilon = 0.5$ and $K = 1.0$. This is shown by the dotted line in Figure 1.

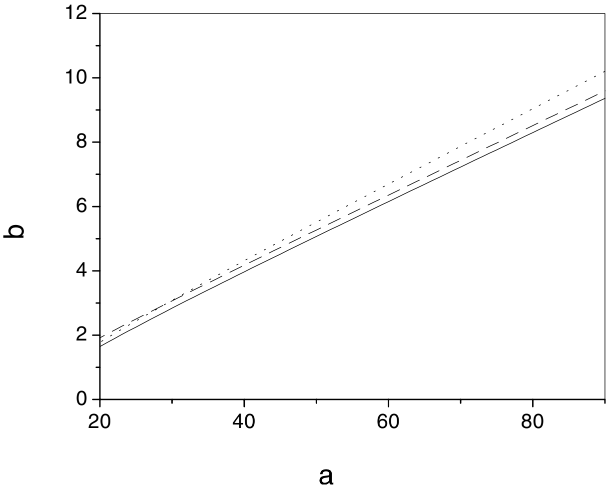

Fig. 1. b vs. a curve for $\sigma = 5.6$ and $d = 1.5$ are shown where the dashed line represents the Hopf bifurcation line (Eq. (33)), solid line represents the Turing bifurcation line (Eq. (34)) and the dotted line corresponds to the Turing bifurcation line under spatial resonance of the parameter b where $\epsilon = 0.5$ and $K = 1.0$ (Eq. (30)).

To realize this situation it is important to recall at this point the experimental spatial modulation of earlier studies [23,29,30]. Since the scaled parameter b essentially contains in addition to kinetic constants the initial concentration of iodine which is highly photosensitive, one may envisage the possibility of modulating this initial concentration by illumination through appropriate masks as I_2 starts reacting in the system in a spatially distributed manner. The mask, as we anticipate, should be designed as a cosine function capable of varying b $[= b_0(1+\cos Kx)]$ in the x-direction with wavelength K as $\frac{2\pi}{\lambda_f}$ where the choice of λ_f should be close to the wavelength associated with the Turing pattern, for study of resonance pattern formation. The control of b through photo illumination for spatial modulation is multiplicative in nature since b appears in equation (32) as a multiplicative kinetic term rather than an additive term as employed before [25,27]. This difference, we believe, is nontrivial from a kinetic or dynamic point of view so far as the effect of spatial modulation is concerned.

For an analysis of pattern formation under parametric resonance the calculations are performed using equations (31) and (32) by the explicit Euler method on a two dimensional grid 128 × 128 with grid spacings $\triangle x = \triangle y = 0.5$ and time step $\triangle t = 0.0005$ and zero flux boundary condition. The simulations are started with spatially random perturbation of $\sim 1\%$ around the steady state. The stationary patterns formed as shown in Figures 2–4 are highly sensitive to the value of perturbation parameter ϵ. We observe numerically that with increase of value of ϵ from 0.5 to 1.0 the spots go over to stripes along x-direction for the same set of parameter values used in Figure 1. The presence of parametric spatial modulation tends to remove the nodal structure along this direction. The domain of instability as noted from theoretical

Fig. 2. Stable spot pattern obtained for $a = 16.0$, $b = 1.5$, $d = 1.5$ and $\sigma = 5.6$ when the spatial perturbation on b is switched off.

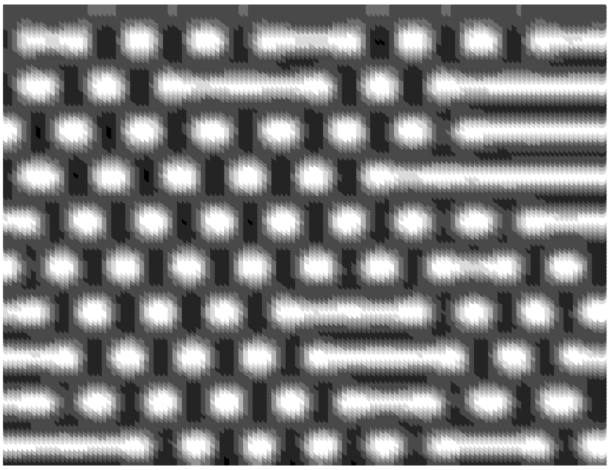

Fig. 3. Stable pattern obtained for $a = 16.0$, $b = 1.5$, $d = 1.5$ and $\sigma = 5.6$ when the spatial perturbation on b is switched on. Pattern with mixed stripes and spots obtained when spatial perturbation parameter $\epsilon = 0.5$.

Fig. 4. Stripe pattern obtained when spatial perturbation parameter $\epsilon = 1.0$ and all other parameter values are same as in Figure 3.

analysis of spatial perturbation is in accordance with numerical simulation results. The stability of the numerical results has been checked by using different space and time steps.

4 Conclusion

We have analyzed a reaction-diffusion system driven by spatial parametric modulation. It is shown that symmetry breaking instability leading to the formation of spatial structures may result, when the depth of modulation is critically controlled in the Hopf region under appropriate resonance conditions. The shift of the Turing bifurcation boundary is calculated perturbatively and analyzed numerically to study the formation of spatial patterns. In view of several recent experiments with temporal and spatial variation of light intensity, the spatial control of modulation in a chemical system where the parameters can be experimentally controlled appears to be important in the context of instability around the Hopf-Turing boundary and related issues in reaction-diffusion systems.

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