# Dynamics of Traveling Patterns under Spatio-Temporal Forcing

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化学反応下で相分離が起こる3 成分混合系に現れる伝播パターンに対して、一定の速度で伝播 する空間周期外力を加えた時のその構造の安定性、引き込み及び変調などを数値シュミレーショ ンで調べ、得られたダイナミクスを理解するため振幅方程式に基づく理論解析を行った。

# **1** INTRODUCTION

Various self-organized patterns emerge in non-equilibrium open systems. Spatio-temporal structures of these patterns have been studied extensively both experimentally and theoretically for many years [1, 2]. The effects of external forcing of these patterns have also been investigated with tremendous interest due to their potential application for nano/mesoscopic domain control in material sciences.

However, there are less extensive studies for the effects of external forcing of non-uniform systems having a spatially regular structure. Only recently, experiments and theoretical consideration have began for Turing patterns influenced by spatio-temporal forcing. For example, effects of illuminating light on a spatially periodic structure are investigated in chemically reacting systems [3, 4].

# 2 CHEMICALLY REACTIVE TERNARY MIXTURES

We employ the model system of phase separation undergoing chemical reactions introduced previously [5]. This mixture is composed of three chemical components A, B and C where A and B species tend to segregate each other and there is a cyclic chemical reaction

$$A \xrightarrow{\gamma_1} B \xrightarrow{\gamma_2} C \xrightarrow{\gamma_3} A \tag{1}$$

with the reaction rates  $\gamma_1$ ,  $\gamma_2$  and  $\gamma_3$ . We assume that other components are also involved in the chemical reaction, which are supplied to and removed from the system and their time scale is sufficiently rapid to be regarded as constant in both space and time. Therefore these components modify only the reaction rates.

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By introducing the local concentrations  $\psi_A, \psi_B$  and  $\psi_C$  of A, B and C components respectively, the time-evolution equations are given by [5]

$$\frac{\partial \psi}{\partial t} = \nabla^2 \frac{\delta F}{\delta \psi} + f(\psi, \phi), \qquad (2)$$

$$\frac{\partial \phi}{\partial t} = g(\psi, \phi), \tag{3}$$

where  $\psi = \psi_A - \psi_B$  and  $\phi = \psi_A + \psi_B$ . We have imposed the condition  $\psi_A + \psi_B + \psi_C = 1$  which is justified by the assumption of the uniformity of other chemical species as mentioned above. The free energy functional F is given by

$$F = \int d\mathbf{r} \left[ \frac{D}{2} (\nabla \psi)^2 - \frac{\tau}{2} \psi^2 + \frac{1}{4} \psi^4 \right],$$
 (4)

where D and  $\tau$  are positive constants. For simplicity, we have ignored  $(\nabla \phi)^2$  term and  $\phi$  dependence of  $\tau$ . The last terms in eqs. (2) and (3) arise from the chemical reaction (1) and are given, respectively, by

$$f(\psi,\phi) = -\left(\gamma_1 + \frac{\gamma_2}{2}\right)\psi - \left(\gamma_1 - \frac{\gamma_2}{2} + \gamma_3\right)\phi + \gamma_3,\tag{5}$$

$$g(\psi,\phi) = \frac{\gamma_2}{2}\psi - \left(\frac{\gamma_2}{2} + \gamma_3\right)\phi + \gamma_3.$$
(6)

It has been shown that the set of equations (2) and (3) has a motionless periodic solution and a traveling wave solution depending on the parameters [5].

The uniform stationary solution of eqs. (2) and (3) are readily obtained by putting f = g = 0. The linear stability analysis gives us the bifurcation diagram as shown in Fig. 1 [5].



 $\boxtimes$  1: Bifurcation diagram for the uniform stationary solution for D = 1,  $\gamma_1 = 0.3$  and  $\gamma_3 = 0.05$ . The full line and the dotted line are the Hopf-type bifurcation line and the Turing-type bifurcation line respectively.

### **3 NUMERICAL SIMULATIONS**

In order to study the behavior above the bifurcation lines we have carried out numerical simulations of eqs. (2) and (3) in one dimension. The Euler method is employed and a periodic boundary condition is imposed. A motionless periodic pattern appears in the region indicated by  $\times$  in Fig. 1 whereas a propagative wave pattern appears in the region +. The value of  $\tau$  at the Hopf-type bifurcation point for  $\gamma_2 = 0.16$  is  $\tau_c \approx 1.46$  at which the critical wave number is  $q_c \approx 0.9$  and the critical frequency  $\omega_c \approx 0.07$ . Hereafter we choose (by the initial condition) a wave traveling to the right without loss of generality.

The spatio-temporal forcing is added to the right hand side in eqs. (2) and (3) as

$$\Gamma(x,t) = \epsilon \cos(q_f x - \Omega t). \tag{7}$$

This is a sinusoidal force traveling to the right at the velocity  $\Omega/q_f$ . Here we suppose that the system is exposed through a periodically arrayed slits by illuminating light with an oscillating intensity. As a result, we assume that the reaction rate  $\gamma_3$  is modified such that  $\gamma_3 \rightarrow \gamma_3 + \Gamma$ . We have ignored a term  $\Gamma \phi$  arising from the  $\gamma_3 \phi$  term in eqs. (5) and (6) providing a sufficiently small forcing  $\epsilon$ .

Here we show only the results for  $q_f = q_c$ . The parameters are chosen as  $\tau = 1.6$  and  $\gamma_2 = 0.16$ . Figure 2 summarizes the behavior on the  $\Omega - \epsilon$  plane. The region indicated by + shows that the traveling wave is entrained with the external force so that it propagates at the velocity  $\Omega/q_f$ . When the frequency  $\Omega$  of the external forcing is far away from  $\omega_c$ , entrainment breaks down and the traveling wave is modulated. These respective behaviors are shown in the space-time plot of the concentration  $\psi$  in Fig. 3.



 $\boxtimes$  2: Phase diagram for  $\gamma_2 = 0.16$ ,  $\tau = 1.6$ and  $q_f = q_c$ . The solid line is the theoretically obtained stability limit of the unmodulated traveling wave having the same velocity as the external force.



 $\boxtimes$  3: Space-time plot of  $\psi$ . The value of  $\psi$ is large (small) for lighter (darker) regions. The parameters are  $\epsilon = 0.006, \Omega = 0.07, \epsilon = 0.005, \Omega = 0.01, \epsilon = 0.006, \Omega = 0.01$  and  $\epsilon = 0.006, \Omega = 0.11$ from top-left to bottom-right.

#### 4 THEORETICAL ANALYSIS

Now we perform a theoretical analysis to understand the dynamics for  $q_f = q_c$ . When the external forcing is not considered, the amplitude equation near the Hopf-type bifurcation has been derived from eqs. (2) and (3) [5]. When the external force is weak as in the present study, the amplitude equation is modified as

$$\frac{\partial W_R}{\partial t} = AW_R + B\frac{\partial^2 W_R}{\partial x^2} - g|W_R|^2 W_R + \epsilon' e^{i(\Omega - \omega_c)t},\tag{8}$$

where we have considered only a wave traveling to the right, whose amplitude is denoted by  $W_R$ . All the coefficients are complex, which we write as  $A = A_1 + iA_2$ . The coefficient  $\epsilon'$  is proportional to the strength  $\epsilon$  in eq. (7).

We have examined the stability of the uniform stationary solution of eq.(8), which is written in the form of  $W_R = Re^{i(\Omega - \omega_c)t}$ . The linear stability analysis of this solution gives us the eigenvalues

$$\lambda \simeq A_1 - 2g_1 Q^2 \pm i |\Omega - \omega_c^R|, \tag{9}$$

where we have used the approximations  ${\epsilon'_1}^2 + {\epsilon'_2}^2 \ll 1$  and  $|\Omega - \omega_c^R| >> 1$  with  $\omega_c^R = \omega_c + A_2 - g_2 Q^2$ . The solid lines in Fig. 2 are obtained in this way and are the stability limit of a completely entrained traveling wave. Note that the imaginary part of the eigenvalue is finite indicating that this is a Hopf bifurcation. This is consistent with the observed oscillatory modulation of traveling waves beyond the threshold. It is emphasized that the simulations for the stability limit agree quantitatively with the theory without any adjustable parameters.

To be summarized, we have investigated, for the first time, entrainment and modulation of a traveling wave under external forcing which depends on both space and time. We have also performed theoretical analysis based on the amplitude equation.

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