

Chemically driven convection can stabilize Turing patterns

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We study the effects of chemically driven convection on Turing structures. We consider a model where convective motion is generated by density gradients due to concentration variations of the Turing structures in a thin horizontally infinite layer of a solute reaction-diffusion system. We study this system with envelope equations and find that the coupling to chemically driven convection modifies the nonlinear coefficients in these equations. For a roll structure we show that this coupling can stabilize pure Turing patterns. An experimental setup to investigate these effects is proposed.

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In 1952 Turing [1] demonstrated that the competition of chemical reactions and diffusion can give rise to a rich variety of pattern forming instabilities. In particular, Turing showed that the wavelength of the resulting stationary patterns is independent of the characteristic lengths in the system, a feature that is qualitatively different from the case of many hydrodynamic instabilities for which the wavelength of the patterns is often given by the geometry of the setup. This is, for example, the case for Rayleigh-Bénard convection and for Taylor vortex flow. Apart from chemical systems this mechanism is also relevant to pattern forming biological systems [2] which were Turing's main motivation. Only quite recently experimental evidence for the existence of Turing structures [3,4] in comparatively simple chemical systems emerged. These structures were observed in reaction-diffusion systems embedded in a gel strip where one species of the reaction-diffusion system is effectively trapped in the gel to provide a big difference in the diffusion coefficients of activator and inhibitor which tends to increase the volume of parameter space where stationary instabilities can occur. Another reason for using gel as a medium for reaction-diffusion pattern forming systems is to avoid the development of convective instabilities which are believed to destroy the emerging Turing patterns. There seems to be no evidence for the existence of Turing patterns in an aqueous solution where convection is possible. But it has been shown that the gel does not seem to be crucial in providing a difference in the diffusion coefficients of the activator and inhibitor species [5]. This difference — for example in the chlorite-iodide-malonic-acid (CIMA) reaction — rather seems to depend on the concentration of starch molecules in which one of the reacting species can be trapped and diffuses therefore more slowly since starch is a fairly big molecule with a diffusion coefficient smaller than that of the usual species in water by one order of magnitude. Without the gel Turing-like patterns could still be observed [5]. In this case convective motion is possible, which leads to the question of how these Turing patterns will interact with the convective flow they induce via concentration or temperature gradients produced by the chemical reactions.

While for the case of oscillatory unstable or excitable reaction-diffusion systems there are numerous studies on the interactions of the oscillatory chemical patterns and convection in aqueous solutions [6–9], triggered by experimental

evidence [10,11], there has not been much work on the interaction of Turing structures and the convective flows they induce in aqueous solutions. The works published so far are mainly numerical studies of the problem in a restricted geometry [12]. There has, however, been some work on the interaction of Turing patterns and an imposed hydrodynamic flow [13,14], which was not driven by the Turing patterns. Here we investigate the emergence of Turing patterns under chemically driven convection close to the onset of the stationary instability using amplitude expansions.

To be definite, consider an isothermal solute reaction-diffusion system in a thin horizontally infinite layer of thickness L with free-free boundary conditions for the fluid motion and Dirichlet boundary conditions for the concentrations in the z -direction. The system is described by the following set of equations:

$$\partial_i v_i = 0, \quad (1)$$

$$\rho_0(\partial_i v_i + v_j \partial_j v_i) = -\partial_i p - \rho g \delta_{i3} + \nu \nabla^2 v_i, \quad (2)$$

$$\partial_t c_i + v_j \partial_j c_i - D_i \nabla^2 c_i = f_i(c_i), \quad (3)$$

where we assume incompressibility. ρ is a function of the concentrations of the two chemical species of the reaction-diffusion system, whereas the density ρ_0 , the viscosity ν , and the diffusivities D_i of the two chemical species c_i are assumed to be constant (generalized Boussinesq approximation). The functions $f_i(c_i)$ describe the kinetics of the chemical reaction of the two species which are assumed to be in general polynomials in c_1 and c_2 . As we will demonstrate in the following, density gradients that are created and maintained by the chemical reaction will create fluid motion because of the density dependent gravity term in the momentum equation for the fluid. This is somewhat similar to the case of a container heated from the sidewalls in thermal convection [15], where however the stress exerted on the fluid layer now is of an intrinsic nature.

We consider the stability of the stationary constant concentration solutions $(c_{10}, c_{20}, \vec{v} = \vec{0})$ to infinitesimal perturbations (c_1, c_2, \vec{v}) where we assume that the density ρ depends on the concentrations and their small perturbations as follows:

$$\rho(c_i) = \rho_0(c_{10}, c_{20}) + \alpha c_1 + \beta c_2. \quad (4)$$

The linear stability analysis shows that the system will undergo a stationary, e.g., a Turing instability, if the condition

$$\frac{(K_{11}D_2 - K_{22}D_1)^2}{4D_1D_2} = -K_{12}K_{21} \quad (5)$$

is fulfilled. Another requirement is that the system has not developed an oscillatory stability before the threshold for the stationary instability is reached. Here k_c is the wave number of the most unstable wavelength given by

$$k_c^2 = \frac{K_{11}D_2 + K_{22}D_1}{2D_1D_2} - \pi^2 \quad (6)$$

and the K_{ij} are the components of the Jacobian of the kinetic functions, $\partial f_i / \partial c_j$, for the chosen equilibrium concentrations (c_{10}, c_{20}) . These can be thought of as being controlled via temperature or the concentrations of basic ingredients in an experiment. There is a critical wave number if $k_c^2 > 0$ or otherwise the wave number that first becomes unstable is zero corresponding to an infinite wavelength. We shall be interested in finite wavelengths here. It is important to keep in mind that the critical wave number is of an intrinsic nature, determined by the details of the reaction-diffusion system and not by the size of the experimental setup as is typically the case for hydrodynamic systems, for example in Rayleigh-Bénard convection. In an experiment undertaken to check the results of this analysis, one must therefore match the system size, e.g., the thickness of the layer, with that intrinsic wavelength. Otherwise effective two dimensionality is no longer preserved and the system we are looking at becomes more complicated. An apparatus similar to the one investigated in this context has already been proposed [16].

To study the effect of convection on Turing structures we proceed to look at the example of roll-like structures. An extension of this analysis to hexagonal and square patterns as well as to their competition with roll patterns will be given elsewhere. We look at roll patterns in the weakly nonlinear regime close to the onset of the instability using standard perturbation expansion methods [17–19] to derive an envelope equation that describes the long time and large scale behavior of these roll-like solutions. In the first order of the perturbation expansion the solution for the concentration perturbation is

$$\begin{pmatrix} c_1 \\ c_2 \end{pmatrix} = W(X, Y, T) e^{ik_c x} \cos(\pi z) \begin{pmatrix} 1 \\ \eta \end{pmatrix} + \text{c.c.}, \quad (7)$$

$$\eta = \frac{D_1(k_c^2 + \pi^2) - K_{11}}{K_{12}}, \quad (8)$$

where we have introduced the amplitude of the roll-like solution which only depends on the slow time coordinate T and the rescaled horizontal coordinates (X, Y) whose dependence on these coordinates has to be determined by solving the equations for the higher orders in the perturbation expansion. The corresponding solutions for the flow velocities are

$$v_{x1} = ik_c \chi W(X, Y, T) e^{ik_c x} \sin(\pi z) + \text{c.c.}, \quad (9)$$

$$v_{y1} = 0, \quad (10)$$

$$v_{z1} = -\frac{k_c^2}{\pi} \chi W(X, Y, T) e^{ik_c x} \cos(\pi z) + \text{c.c.}, \quad (11)$$

the first nontrivial contribution to v_y ,

$$v_{y2} = \chi \partial_Y W(X, Y, T) e^{ik_c x} \sin(\pi z) + \text{c.c.}, \quad (12)$$

being of the next order in the perturbation expansion and where

$$\chi = \frac{g \pi (\alpha + \eta \beta)}{\nu (k_c^2 + \pi^2)^2}. \quad (13)$$

As one sees the first contributions to the velocity in the expansion are of the same order as the first contributions to the chemical concentration variations that drive convection. This is qualitatively different from the case of the oscillatory instability discussed in Ref. [20], where the first nontrivial contributions to the velocity were only of second order.

The resulting envelope equation for rolls is then given by

$$\begin{aligned} \partial_T W + \frac{D_1 D_2}{\delta} (\partial_Y^2 + 2ik_c \partial_X)^2 W \\ + (\gamma_{11} K_{22} + K_{11} \gamma_{22} - K_{12} \gamma_{21} - \gamma_{12} K_{21}) W / \delta = b_3 |W|^2 W, \end{aligned} \quad (14)$$

where

$$\delta = D_2(k_c^2 + \pi^2) - K_{22} + D_1(k_c^2 + \pi^2) - K_{11}. \quad (15)$$

The γ_{ij} measure the small deviations of the linear coefficients of the chemical reaction kinetics polynomials which control onset of the chemical instability and determine its threshold from their critical values into the Turing unstable region of the parameter space of the system in question. They are a measure for the strength of the chemical instability [18]. The cubic coefficient in the amplitude equation whose explicit form is rather lengthy and will therefore be given in a longer paper contains contributions from the convective term in the reaction-diffusion system, whereas the advection terms of the velocity equations do not contribute to the cubic coefficient as is the case for Rayleigh-Bénard convection [17,19]. It should be noted that this contribution to the cubic coefficient will always lower the value of the cubic coefficient and hence increase the stabilizing contributions to the cubic term in the envelope equation. This is due to the explicit mathematical form of the terms that describe the coupling of convection to the chemical concentrations and vice versa. They have the same form as in the case of the coupling of the temperature field to convection and vice versa in simple Rayleigh-Bénard convection where they result in the cubic coefficient which gets only one contribution from the advective term in the temperature equation always saturating the envelope equation for the roll solution [17,19]. Therefore convection enhances the stability of the roll solution. Namely, if convection results in a negative cubic coefficient where, without convection, there would be a positive cubic coefficient, a roll structure that would otherwise be unstable is stabilized, if convection is present, and therefore

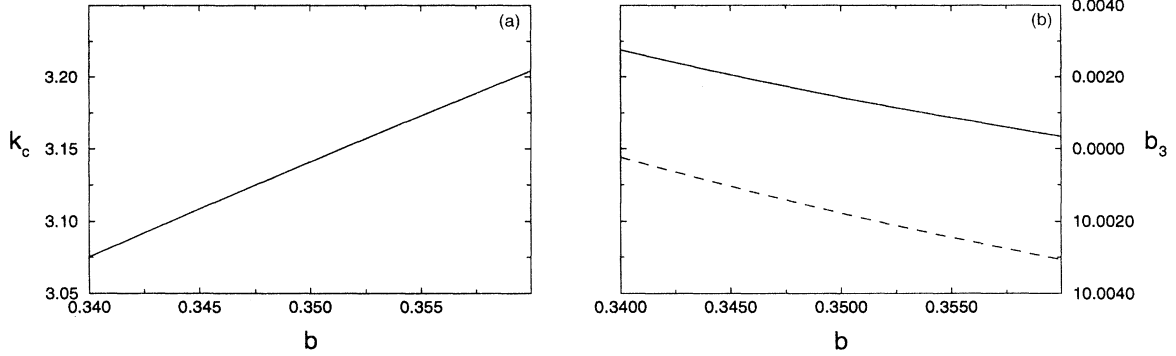


FIG. 1. (a) is a plot of the dimensionless critical wave number k_c as a function of b where $d=0.3$, $\gamma=80$, $Sc=2000$, $R_1=1 \times 10^{-4}$, and $R_2=0$. The critical wave number $k_c = \pi$ that matches the layer depth is reached at approximately $b=0.35$. (b) is a plot of the dimensionless cubic coefficient b_3 where the solid line denotes the case without convection and the dashed line the case with convection.

some other pattern that exists in the absence of convection might be replaced by the roll solution under convection.

To demonstrate this possibility we have calculated the coefficient in question for a particular model reaction-diffusion system — the Schnakenberg model [21,22]. Equations (1)–(3), when specialized to the Schnakenberg model and after suitable reduction of parameters, read

$$\partial_i v_i = 0, \quad (16)$$

$$\partial_t v_i + v_j \partial_j v_i - \nabla^2 v_i + \partial_i p = -Sc(R_1 c_1 + R_2 c_2) \delta_{i3}, \quad (17)$$

$$\partial_t c_1 + v_j \partial_j c_1 - \frac{1}{Sc} \nabla^2 c_1 = \frac{\gamma}{Sc} (a - c_1 + c_1^2 c_2), \quad (18)$$

$$\partial_t c_2 + v_j \partial_j c_2 - \frac{1}{dSc} \nabla^2 c_2 = \frac{\gamma}{Sc} (b - c_1^2 c_2), \quad (19)$$

where

$$R_1 = \frac{g\alpha L^3}{D_1 \nu} \left(\frac{k_2}{k_3} \right)^{1/2}, \quad R_2 = \frac{g\beta L^3}{D_1 \nu} \left(\frac{k_2}{k_3} \right)^{1/2}, \quad (20)$$

$$Sc = \frac{\nu}{D_1}, \quad d = \frac{D_1}{D_2}, \quad \gamma = \frac{L^2 k_2}{D_1}, \quad (21)$$

ν is the viscosity of the fluid in question, the parameter d is the ratio of the diffusion coefficients D_1 and D_2 . The R_i are somewhat analogous to the Rayleigh number and Sc is the Schmidt number associated with species 1. a and b are the suitably rescaled dimensionless pool species concentrations, which are controlled and held constant in an experiment and determine the type of instability the system can undergo. We introduced the thickness of the layer as relevant length scale. The k_i are the coefficients of the Schnakenberg model's chemical reaction terms.

The contributions of the convection to the cubic term in the amplitude equation can lead to the stabilization of a roll-type structure as can be seen in Fig. 1, where convection results in a shift of the cubic coefficient for the relevant parameter value $b=0.35$ towards a negative value which assures saturation of the roll amplitude to cubic order. Stabilization has been observed for different sets of parameters.

The parameters here are evaluated using $\nu=10^{-2}$ cm²/s, $D_1=5 \times 10^{-6}$ cm²/s and assuming $L \propto 1$ mm. This leads to $Sc=2000$ and we subsequently chose $\gamma=80$. The parameters d , R_1 , R_2 were varied in the calculations.

Figure 2 demonstrates a situation where the cubic coefficient is already negative, but becomes more negative when convection is taken into account. Furthermore, one can see that a contribution of the second species to the density variation ($R_2 \neq 0$) can reduce the effect, because the concentration c_2 is out of phase with the concentration c_1 ($\eta < 0$). But the effect is larger again, when R_2 is bigger than a certain value that leads to a vanishing contribution to the cubic coefficient. All this is due to the fact that the coupling parameters R_i appear in the relevant contribution to the cubic coefficient in the form $(R_1 + \eta R_2)^2$.

In conclusion we have demonstrated that Turing patterns

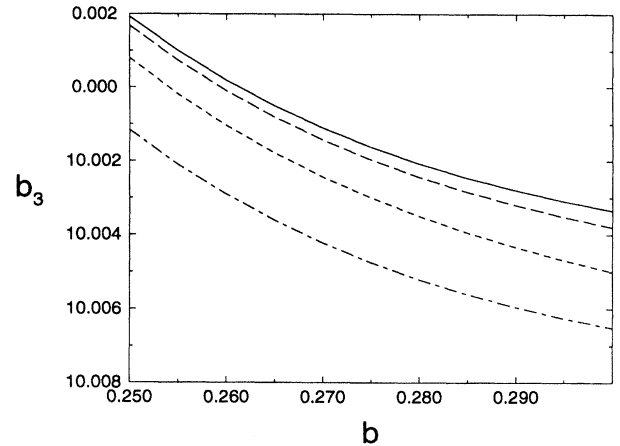


FIG. 2. This is a plot of the dimensionless cubic coefficient b_3 as a function of b where $d=0.5$, $\gamma=80$, $Sc=2000$, $R_1=1 \times 10^{-4}$. The critical wave number $k_c = \pi$ that matches the layer depth is reached at approximately $b=0.27$. The solid line denotes the case without convection. The short dashed line denotes the case with convection and $R_2=0$, whereas the long dashed line denotes the case with convection, but with $R_2=1.5 \times 10^{-5}$, and the dot dashed line denotes the case with convection with $R_2=1 \times 10^{-4}$. The partial cancellation of the effects of the two different concentrations is due to η being negative.

and the convection induced by them are of the same order in the perturbation expansion in the Schnakenberg model. Therefore convection is important already close to onset of the instability. The interaction of Turing patterns and convection results in the stabilization of roll patterns as compared to the situation when convection is absent, e.g., in Turing structures in gel strips. This result indicates that coexistence of chemically driven convection and Turing patterns driving them is possible at least in the weakly nonlinear regime.

We have chosen the geometry of the system we were looking at already in the spirit of what might be a promising experimental setup to study these effects. In matching the thickness of the fluid layer with the intrinsic wavelength of the chemical instability we arrive at a quasi-two-dimensional pattern forming system that is very similar to the situation encountered in Rayleigh-Bénard convection or in electrocon-

vection in liquid crystals. A layer much thicker than the critical intrinsic wavelength would result in three-dimensional Turing structures [23] whose interactions with convection are likely to be much more complicated and difficult to observe than the system described here. So matching the intrinsic wavelength of the system and the thickness of the apparatus we are using to study chemically driven convection in Turing patterns is essential to control the system and study it. We think that it is also more likely to observe ordered structures in the setup proposed here. To reduce experimental difficulties it might be interesting to start studying the interaction of Turing patterns and chemically driven convection in sol-like media where the strength of convective flows could be reduced to a level that is easier to control.

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