Patterns in reaction diffusion system

Boyce Tsang
Department of Physics, University of Illinois Urbana-Champaign

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Abstract

Reaction-Diffusion systems are important in the field of non-equilibrium phenomena with relevance to biological and synthetic pattern formation. While homogenous distribution of chemicals was always believed to be a stable state, the symmetry-breaking treatment by Turing on such systems in 1951 showed pattern formation could be more stable in certain cases. This paper reviews the treatment by Turing and the subsequent evidences for and against its application.

1 Introduction

1.1 Reaction Diffusion System

Reaction diffusion (**RD**) systems are characterized, in this article, as chemical systems with two active components that react with other components, and with different diffusion coefficient. Due to limitations in this paper, generalized systems with more components will not be discussed. Specifically, the concentration $u_1(\mathbf{r})$ and $u_2(\mathbf{r})$ are governed by the following **master equations** [1],

$$\begin{cases} \dfrac{\partial u_1}{\partial t} = r_1(u_1, u_2) + D_1 \nabla^2 u_1 & (1) \\ \dfrac{\partial u_2}{\partial t} = r_2(u_1, u_2) + D_2 \nabla^2 u_2 & (2) \end{cases}$$

where the following assumptions are made,

(i) The reaction terms, r_1 and r_2, are assumed to be a function of *present* and *local* concentration u_i only ($i = 1, 2$), but not explicitly on space and time.

(ii) the diffusion coefficient D_i do not explicitly depend on space and time. (Clearly, when $r_i \equiv 0$ then both equations are diffusion equations.)

These assumptions are justified when there is no pre-pattern in the system [1], apart from the assumption that r_i is a function of u_i only. It was a practical consideration that such assumption was difficult to hold for *closed system*, since chemical reactions often involves more than two components, and also catalysts would affect the reaction rates [1]. Therefore, the dependence of solution on boundary not only originates from the fact that Equation (1) and (2) are *coupled partial differential equations*. The validity of these equation itself introduced considerable experimental difficulties, which will be addressed in this paper [11]. It should be noted that such assumption is often valid only in *open systems*[11].

In equilibrium, u_1 and u_2 are homogeneous in space and time, then the master equations become $r_1 = r_2 = 0$. With further knowledge in r_1 and r_2, in principle the homogeneous solution $u_i(\mathbf{r}) = U_i$ can be found.

The aim of this paper is to explore symmetry breaking in RD systems beyond equilibrium thermodynamics. It was shown by Alan Turing in 1951 [2] that homogeneous solution may be, surprisingly, unstable subject to spatially periodic perturbation. Hence, even *without* pre-pattern, Turing patterns (periodic patterns from RD system) are possible in RD system.

Before introducing the classical treatment by Alan Turing, the motivation of studying such system and some background information will be presented.

1.2 Implication on pattern formation

The possibility of pattern formation without pre-pattern is important in both understanding in natural-occurring patterns and synthetic patterns.

Firstly, the pattern formation of biological beings, including fingerprints, strips on zebra and skin patterns on fishes, were not well quantitatively understood [3]. RD systems has been speculated to be an accurate model for such patterns, because the strips or the spots on the skins are not determined by structures underneath [4]. In fact, there are experiments aiming at using RD model to predict perturbation on animal skin by laser ablation [5], as shown in Fig. 1.

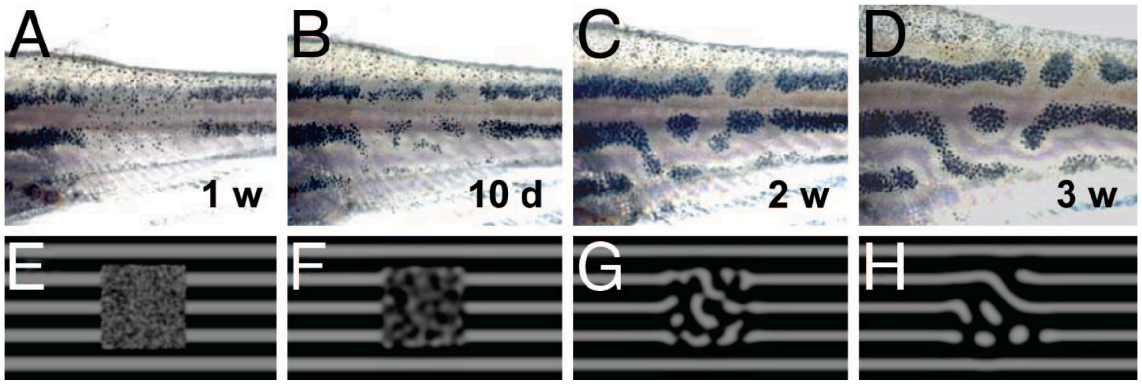

Figure 1: Time evolution of recovering of strips on zebra fish. The upper series (A to D) showed the regeneration of an actual zebra fish after laser ablation. The lower series (E to H) showed results from simulation. Adapted from [5]

However, it should be noted that the biological community remained conservative towards applying simplistic model such as Turing's treatment on complicated biological systems. Also, supporting literature was limited and justification was difficult.

Secondly, pattern formation is also of great importance in microstructure engineering. Popular deposition methods, in both industry and research, include sputtering and lithography [6]. In order to deposit desired structure on substrates, masks are often used to control to arrival sites modifying agent. With spontaneous pattern formation from RD system, it is possible to reduce complications such as diffraction and splashing from masks, and thus to achieve higher accuracy [7]. Also, the sensitivity of pattern formation to initial condition and environmental factors may be used in sensing application [7] and cellular engineering [8]. These advantages may be applied to different classes of devices such as cameras and antioxidant detection [7].

2 Turing Patterns

In 1951, Alan Turing published his only paper on biological pattern formation and opened the door to the investigations thereafter [1]. In the original paper [2], Turing analyzed the possible cases from Equation (1) and (2) for a simplified scenario. Instead of field equations, Turing discretized the system with N sites in one dimension. It was done to simplify the computation and it can treated as a circle of N cells in which reaction may take place, and intracellular exchange of chemical was possible [2]. Here the Turing's linear stability analysis will be studied with continuous coordinates x instead.

2.1 Linear stability analysis

In order to investigate whether uniform distribution was stable, Turing assumed the solution to be $u_i(x) = U_i$ for all x. Then, small perturbation in $\tilde{u}_i$ was introduced such that $u_i = U_i + \tilde{u}_i(x)$. Then r_i can be expanded in first order, with $r_i(U_1, U_2) = 0$, as follow,

$$r_i(u_1, u_2) = r_{i1}\tilde{u}_1 + r_{i2}\tilde{u}_2 \tag{3}$$

Now with the convention above, the *linearized* master equations read [1],

$$\frac{\partial \tilde{\mathbf{u}}}{\partial t} = \mathbf{R}\tilde{\mathbf{u}} + \mathbf{D}\frac{\partial^2 \tilde{\mathbf{u}}}{\partial x^2} \tag{4}$$

with $\tilde{\mathbf{u}} = \begin{pmatrix} \tilde{u_1} \\ \tilde{u_2} \end{pmatrix}$, $\mathbf{R} = \begin{pmatrix} r_{11} & r_{12} \\ r_{21} & r_{22} \end{pmatrix}$, $\mathbf{D} = \begin{pmatrix} D_1 & 0 \\ 0 & D_2 \end{pmatrix}$.

Assuming the perturbations are harmonic in space, the spatial variation was assumed to be e^{iqx}. Then, to allow this perturbation to grow, diminish, or oscillate, the temporal variation as assumed to be $e^{k_q t}$. Now the real component of k_q, $\Re(k_q)$ represents time evolution of the *amplitude* of the perturbation. If $\Re(k_q) < 0$, then the perturbation diminish with time and the system is then referred as *linearly stable*. On the other hand, if $\Re(k_q) > 0$, then perturbation will grow with time, at least when the linear approximation (Equation (3)) is valid. Therefore, it is crucial to find out the sign of $\Re(k_q)$ for different q. In principle this is straight forward since Equation (4) is linear, therefore differential operators become arithmetic multiplication. Requiring non-trivial solutions give

$$\det(\mathbf{R} - \mathbf{D}q^2 - k_q\mathbf{I}) = 0 \tag{5}$$

After some algebra, the condition for existence of finite q with positive $\Re(k_q)$ and $\Im(k_q) = 0$ is [1]

$$D_1 r_{22} + D_2 r_{11} > 2\sqrt{D_1 D_2 (r_{11} r_{22} - r_{12} r_{21})} > 0 \tag{6}$$

Note that, when $q \neq 0$ this is called *Turing instability* or *Turing bifurcation*.

2.2 Diffusion-induced instability

There is a specific type of instability that require non-zero D_i [9]. In order to choose such instability, Equation (5) with $\mathbf{D{=}0}$ can be used to solve for $\Re(k_q) < 0$ (stable solution). It can be shown that

$$r_{11} + r_{22} < 0 \tag{7}$$

Requiring both Equation (6) (Existence of instability) and Equation (7) (Stability without diffusion) to be true, r_{11} and r_{22} must have opposite signs [1]. Without loss of generality, from here it is assumed that $r_{11} > 0$ and $r_{22} < 0$. The allowed signs for $\mathbf{R}$ are $\begin{pmatrix} + & - \\ + & - \end{pmatrix}$ and

$\begin{pmatrix} + & + \\ - & - \end{pmatrix}$ [10]. In these cases, component 1 in the system is called *activator*, as the linear response of increased u_1 is positive. On the other hand, component 2 is called *inhibitor* in the literature [1]. The necessary condition for diffusion-induced instability is [1],

$$\frac{D_2}{D_1} > \frac{-r_{22}}{r_{11}} \tag{8}$$

2.3 Activator-inhibitor systems

One of the triumph of Turing's paper was the possibility for fluctuations to spontaneously break symmetry in uniform equilibrium state [3]. In this case, Inequality (8) was simple enough such that physical interpretation was possible.

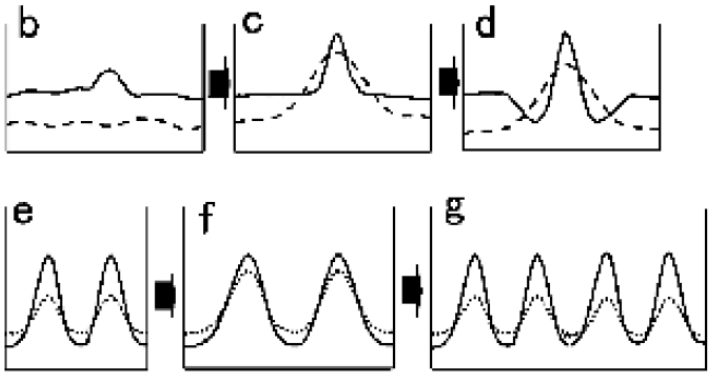

Figure 2: Schematic diagram (from b to f) of time evolution of activator-inhibitor system. Solid and dashed line represents the concentration of activator and inhibitor respectively. Note that in this case $\mathbf{R} = \begin{pmatrix} + & + \\ - & - \end{pmatrix}$. Adapted and extracted from [4]

In the figure above, a localized increase in activator resulted in increases in both components. Then, with condition (8), the inhibitor diffused faster and created a localized decrease around. Then such behaviour continues to establish a fluctuation in finite q. This is the core insight of Turing's analysis on RD system, that "local activation with long-range inhibition" [1] could lead to symmetry breaking patterns. Then, one of crucial question left was how exactly could experimentalist verify the analysis above. While results from experiments will be discussed in later sections, in below the subtle difficulties will be evaluated first.

2.4 Experimental Limitations

Although the analysis seemed promising to give spontaneous Turing patterns in many systems with two active components, only after about 40 years did experimentalists succeed in

observing Turing patterns in chlorite-iodide-malonic acid (CIMA) reaction [11]. The difficulties arise not only from condition (8), but also the master equation itself, reflected by the limited validity of assumption (i) at the beginning of the paper.

2.4.1 Supply of inactive chemical

It was assumed from Equation (1) and (2) that r_i are functions of concentration of active components u_1 and u_2 only. In reality, of course activators can not simultaneously increase both u_1 and u_2, as depicted in Figure (2), without consuming other chemicals. Therefore, continuous supply of *inactive* chemicals were needed [12], as depicted by Figure (3).

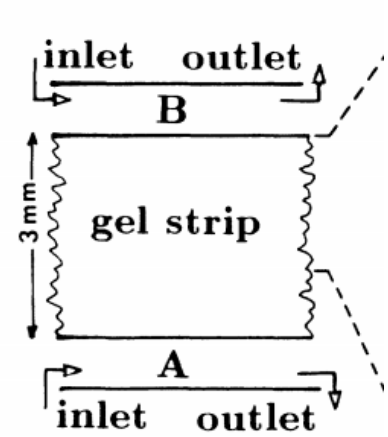

Figure 3: Experimental transport system to supply chemicals. Adapted and extracted from [11]

2.4.2 Limited difference in diffusivity

On the other hand, finding suitable chemical to satisfy condition (8) was not straightforward, as $\frac{-r_{22}}{r_{11}}$ may exceed 10 in general [1], and mobile ions with colour usually do not differ in diffusivity as much. In fact, the diffusivity ratio $\frac{D_2}{D_1}$ was only 1.07 in the CIMA experiment [12]. However, when starch was added to the solution, it binds with activator and slowed down its diffusion, render the effective diffusivity ratio much higher than usual.

2.5 Pattern selections and wave instability

The linear response analysis from Turing could not reveal the dynamics of pattern selection because Equation (4) would not be valid soon after deviation from uniform state. Therefore, concrete analysis with functional form of $r_i(u_1, u_2)$ are needed to solve the problem of *pattern selection*. Some popular choice of nonlinear system, such as the Brusselator, is given by [13],

$$\begin{cases} r_1(u_1, u_2) = a - (b+1)u_1 + u_1^2 u_2 \\ r_2(u_1, u_2) = bu_1 - u_1^2 u_2 \end{cases} \tag{9}$$

One could have followed the analysis above and the corresponding q for this system. Furthermore, with Equation (9), numerical integration is possible such that the dynamics of pattern selection can be studied. Due to length constraints such technical detail will be discussed here.

Also, Turing patterns are not the only possibility for symmetry breaking stable states. One could have chosen to tune parameters in r_i such that $\Re(k_q) > 0$ but $\Im(k_q) \neq 0$ at $q = 0$. This is then called *Hopf instability*, or *bifurcation*. As a result, the concentrations u_1 and u_2 remain uniform at a fixed time, but oscillate with time. It can be viewed as a symmetry breaking phenomena in temporal dimension but the spatial symmetry remained unbroken. Another studied instability, *wave instability* in literature generally refers the traveling wave solution.

3 Progress after Turing's work

3.1 Progress in realization of Turing patterns in chemical system

As stated before, direct observation of Turing patterns are first recorded in 1990 in CIMA system. Other than actual chemical systems, computer simulations was also instrumental to understanding of RD systems. Figure (4) shows possible patterns from simulations and only relatively simple patterns such as strips and hexagonal geometries were found [14].

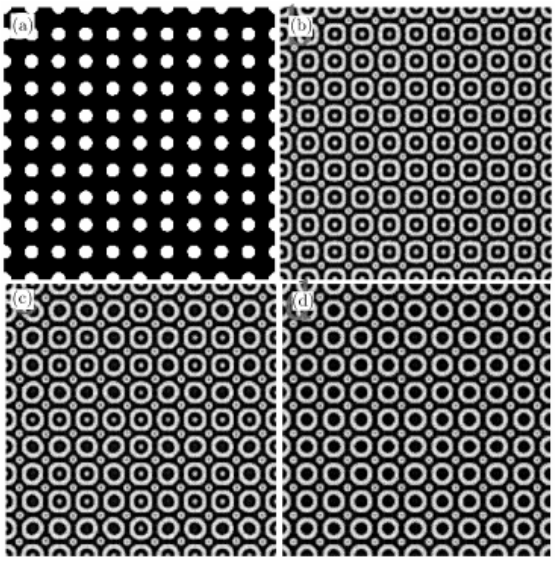

Figure 4: Square lattice formed by strips in orthogonal directions. Adapted and extracted from [14]

In order to generate complex patterns, illumination may be used to externally force the CIMA solution. This is because the reactions are sensitive to light [14]. It was known that, if

the illumination was modulated in time, then CIMA system would behave as *stirred* system, not allowing spatial variance. On the other hand, spatial periodic masks could be used to generate complex patterns [14].

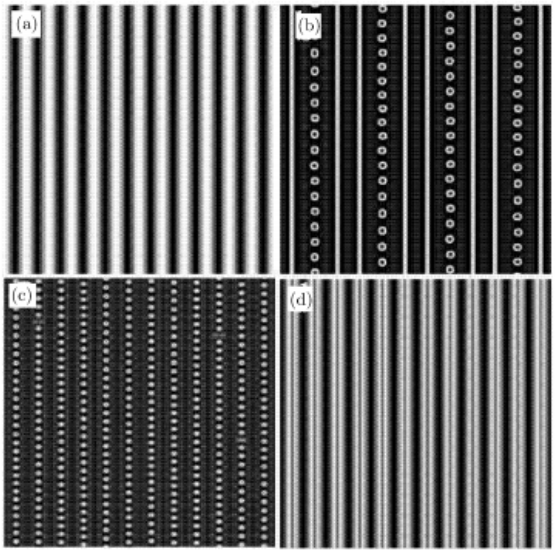

Figure 5: Strip patterns with spot inside. These patterns were possible with spatially modulated masks. Adapted and extracted from [14]

3.2 Progress in identifying Turing patterns in biological system

As stated at the beginning of this paper, one of the motivation of studying pattern formation is to understand how biological pattern is possible without pre-pattern. Of course, even when simulations with particular value of parameter reproduce some known biological patterns, it is not a direct evidence that RD equations are responsible for such patterns [3].

In fact, biologists community had question for years that how could complex biological system be modeled by simple mathematical equations [3]. However, recently it is possible to provide stronger evidence for RD system by predicting dynamics of biological patterns, as depicted by Figure 1, by higher computational capacity. On the other hand, using genetic techniques, it is now possible to study directly the dynamics of molecules by fluorescence particles [5].

3.3 Progress in theoretical works

3.3.1 Cross diffusion

In Equation (1) and (2), the diffusion was assumed to be independent of the other components. However, ion interactions and excluded volume effect may induce *cross diffusion*, in which a gradient in concentration of one component would induce flux of the other one [15].

It was equivalent to adding a $\nabla \cdot (D_{21} \nabla u_1)$ term to Equation (1) (similarly for Equation (2)). It was shown that even a relatively small value of D_{ij} $(i \neq j)$, the pattern formation dynamics could still be affected [15].

3.3.2 Interactions between Turing and Hopf instabilities

In earlier section, the calculation for Turing instability had been explicitly carried out. In principle, one could also solve for Hopf instability in similar manner. In fact, at the vicinity of Hopf instability, the oscillation is governed by complex Ginzburg-Landau equation [16], which also governs the order parameter of superconductor in equilibrium. In fact, there was experimental efforts in finding the parameters in Ginzburg Landau Equation [17].

When the parameters in r_i changes, the RD system is then unstable to different perturbations. Some recent theoretical works focused on the behaviour of systems when the parameters were near both instability. It was shown to be possible to form more complex patterns with interaction between instabilities [18].

4 Summary

In this paper, the master equation for studying reaction diffusion systems (Equation (1) and (2)) and its importance on pattern formation was introduced. Then the influential treatment by Turing was introduced by linear stability analysis. It was shown that Turing patterns are diffusion-induced instability, with the condition of "local activation with long-range inhibition" [1].

However, to study dynamics away from uniform states, such as pattern dynamics and pattern selections, tools beyond stability analysis have to be used. They are particularly crucial to settle disputes in applying RD system to explain pattern formation in nature. Also, recent theoretical progress allowed more complex patterns by considering additional terms or interactions between instabilities.

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