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Instability and pattern formation in reaction-diffusion systems: A higher order analysis

Heterogeneous and SelfOrganized Pacemakers in ReactionDiffusion Systems
Diffusion-induced periodic transition between oscillatory modes in amplitude-modulated patterns

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We study amplitude-modulated waves, e.g., wave packets in one dimension, overtarget spirals and superspirals in two dimensions, under mixed-mode oscillatory conditions in a three-variable reaction-diffusion model. New transition zones, not seen in the homogeneous system, are found, in which periodic transitions occur between local $1^N$-1 and $1^N$ oscillations. Amplitude-modulated complex patterns result from periodic transition between $(N-1)$-armed and N-armed waves. Spatial recurrence rates provide a useful guide to the stability of these modulated patterns. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4872215]

Modulated waves, i.e., waves with multiple wavelengths and/or amplitudes, are of interest in many disciplines. Their mechanistic origin and their stability are fertile areas of inquiry. Here we study a tritrophic predator-prey model as a reaction-diffusion system in mixed-mode oscillatory media. The introduction of diffusion induces new transition zones in the phase diagram, in which periodic switches occur between local $1^N$-1 and $1^N$ oscillations, and complex amplitude-modulated waves appear. Analysis using spatial recurrence rates suggests that the modulated waves are intermediate in stability between unmodulated waves and spatial turbulence. The diffusion-induced second-frequency oscillations between dynamical modes may shed light on the complexity of certain spatiotemporal patterns in nature.

I. INTRODUCTION

Spatiotemporal patterns are a trademark of spatially distributed nonlinear systems and have played an important role in recent investigations in physics, chemistry, biology, and ecology.1–5 Most studies, however, have focused on relatively simple dynamical systems whose local dynamics exhibit either excitability or simple periodic oscillation. The dynamical complexity in the multivariable systems that occur in nature often leads to the presence of multiple feedback loops, which can produce mixed-mode oscillations, period-doubled oscillations, or quasi-periodic oscillations via various bifurcations of periodic orbits.6 These complex oscillations and their associated instability-induced chaos, when they constitute the local dynamics of a reaction-diffusion system, can lead to a rich variety of complex spatiotemporal patterns. For instance, Hopf bifurcation of tip movement in spirals can result in the meandering of spiral cores.7–10 Period-doubling bifurcations can lead to deformed spirals that exhibit several types of defect lines.11–13

Mixed-mode oscillations, first observed in heterogeneous chemical reactions14 and most thoroughly characterized in the classic Belousov-Zhabotinsky reaction,15 have been widely investigated in experiments and simulations of chemical and biological nonlinear systems.16 In a multi-variable dynamical system, a fold bifurcation (saddle-node bifurcation) of periodic orbits can result in a series of mixed-mode oscillations, which are constituted by large amplitude oscillations combined with a number of small peaks, where the notation $L_1$ signifies L large and S small amplitude oscillations per cycle, e.g., $1^1,1^2,\ldots,1^N$ ($L=1, S=1, 2,\ldots, N$) mixed-mode oscillations. Of particular interest here, spatiotemporal patterns with amplitude modulation can be observed both in 1D and 2D simulations of mixed-mode oscillatory reaction-diffusion media. In previous works, amplitude-modulated patterns with superstructures, such as overtarget spirals and superspirals, have been reported.17,18 The aim of the present work is to examine the dynamical origin, stability and structure of modulated patterns generated in mixed-mode oscillatory media.

II. MODEL AND HOMOGENEOUS DYNAMICS

The three-variable dynamic model employed in this work, which was proposed by Hastings and Powell,19,20 is an extension of two-variable Rosenzweig-MacArthur model.21,22 This model can represent either a prey or a chemical reaction system.23 The reaction-diffusion model is written in dimensionless units as

$$
\frac{\partial u}{\partial t} = u - u^2 - \frac{a_1 u}{1 + b_1 u} v + D_u \nabla^2 u
$$

$$
\frac{\partial v}{\partial t} = \frac{a_1 u}{1 + b_1 u} v - \frac{a_2 v}{1 + b_2 v} w - d_1 v + D_v \nabla^2 v
$$

$$
\frac{\partial w}{\partial t} = \frac{a_2 v}{1 + b_2 v} w - d_2 w + D_w \nabla^2 w.
$$

[1]

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In ecological terms, $v$ corresponds to a species that preys upon $u$, and $w$ preys upon $v$. In a chemical interpretation, the system describes consecutive reactions, $u \rightarrow v \rightarrow w$, where $u$ is the substrate, and $v$ and $w$ are products or activators. The parameter values $a_1, b_1, a_2, b_2, d_1, d_2$ and the diffusion coefficients in the present paper are given in Table I. We assume that diffusion of $u$ may be neglected in comparison with that of the other two species, i.e., we take $D_u = 0$.

Peet et al.\textsuperscript{24} have analyzed the temporal behavior of a similar model for a three-level trophic food chain but with additional quadratic decay terms that represent intraspecies interaction of species $v$ and $w$. These additional terms, which may represent fighting or competition for resources within a species, limit the growth of, or “close,” the two higher trophic levels, a feature that makes them more realistic in an ecological context, though it may or may not be appropriate in a given chemical context. McGehee and Peacock-López\textsuperscript{25} studied the spatial behavior of a two-variable model with a quadratic decay term to close the predator level and found the existence of Turing patterns.

There exist two limit cycles in this dynamical model; each involves one positive and one negative feedback. The first corresponds to the relationship between the prey $u$ and the predator $v$. The second relationship describes the predator-superpredator ($v$-$w$) interaction. If the parameters are chosen so that $a_2/b_2 < d_2$, we have at all times

$$\frac{\partial w}{\partial t} = \frac{a_2 v}{1 + b_2 v} w - d_2 w < 0.$$  

(2)

In this case (and for some nearby parameter sets), the concentration of $w$ approaches zero, and the local dynamics of system (1) can be reduced to a two-variable subsystem:

$$\frac{\partial u}{\partial t} = \frac{a_1 u}{1 + b_1 u} v - d_1 v.$$  

(3)

As shown in Fig. 1(a), under these conditions only oscillations of components $u$ and $v$ are observed. For other parameter values, due to the coupling among feedback loops, attractors emerge that consist of mixed-mode oscillations with low-frequency (arising from the relationship between

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### TABLE I. Parameters in the reaction-diffusion model.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
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<tr>
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<tr>
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<td>0.1</td>
</tr>
<tr>
<td>$D_w$</td>
<td>0.1</td>
</tr>
</tbody>
</table>

---

FIG. 1. Lyapunov exponent phase diagram in the $(a_2-b_2)$ parameter space. Colors denote the chaotic regime (positive exponents), while gray areas correspond to periodic behavior (negative exponents). The system is divided into several dynamical regions by bifurcation curves: (a) simple oscillations in the $u$-$v$ plane; (b) simple three-variable oscillations; (c) $1^1$ oscillations; (d) period-2 ($1^1$) oscillations; (e) $1^2$ oscillations. Yellow lines and white lines are fold bifurcation curves between different mixed-mode oscillatory regions and period-doubling bifurcation curves, respectively.
v and w) and high frequency (small orbits in the u-v plane) components, as depicted in Figs. 1(c)–1(e).

In the absence of diffusion, this three-variable model displays several bifurcation routes to chaos.26,27 Figure 1 presents typical oscillatory behaviors in this model and summarizes the data as a Lyapounov exponent phase diagram in the $a_2$-$b_2$ parameter plane. Different color maps are employed for the positive (colors) and negative (gray) Lyapounov exponent regions. The system may lose its stability via two kinds of bifurcation: period-doubling and fold bifurcations. The bifurcation curves (constructed from the critical points where a Lyapounov exponent is zero) partition the parameter space into several dynamical regions, as seen in Fig. 1. The white lines are the period-doubling bifurcation curves, whereas the yellow lines are fold-bifurcation curves separating $1^{N-1}$ and $1^N$ mixed-mode oscillations.

III. SPATIOTEMPORAL BEHAVIOR: RESULTS AND DISCUSSION

A. 1-D traveling wave packets

We first carried out numerical simulations of traveling waves in one spatial dimension (1D) to elucidate the characteristic spatiotemporal structures in mixed-mode media. The 1D reaction-diffusion system had length $L = 3000$, time step $d_t = 0.01$, and spatial grid $d_s = 1.0$. The simulations utilized an explicit fourth-order Runge-Kutta algorithm. With smaller spatial grids and time steps, the results were essentially unchanged. Snapshots of component v are employed for the best visual presentation. The parameter values and diffusion coefficients in Table I were used in the 1D simulations.

We observed two distinct types of traveling waves in our simulations. The first are simple waves with constant amplitude, which typically occur in parameter regions far from the dynamical bifurcation curves. The second and more interesting form of traveling wave generally arises in the neighborhood of one of the fold bifurcation curves. As shown in Fig. 2(a), the heights of these traveling waves oscillate, forming packets of traveling waves (red line in Fig. 2(a)) on the basic wave. The time series in Fig. 2(b) indicates that the local dynamics at a point consists of oscillation packets, containing a mixture of $1^0$ and $1^1$ oscillations with different amplitudes. The space-time plot shown in Fig. 2(c) also illustrates that the pattern comprises two scales of traveling waves, which are marked by arrows. $K_1$ indicates the propagation of the basic traveling waves, and $K_2$ highlights the propagation of the amplitude modulation. We shall refer to such complex patterns with more than one wave number as modulated patterns.

B. 2-D amplitude-modulated patterns

We next extended our reaction-diffusion simulations to two spatial dimensions (2D), focusing on spiral waves, which are familiar in many biological, chemical and physical systems. We generated spirals by perturbing the homogeneous steady state with imposed orthogonal gradients of v and w at the central point of the system. We used an Euler integration algorithm with a spatial grid of 1.0 unit and integration time step 0.01. The patterns were essentially unchanged with a smaller time step and finer spatial grid. To further check the accuracy, we carried out simulations with an explicit fourth-order Runge-Kutta algorithm, and the same results were obtained.

We are interested here in the evolution of spatiotemporal patterns generated in mixed-mode oscillatory media with different types of local kinetics. As in the case of traveling waves in 1D, the evolution of 2D spiral patterns from $1^0$ to $1^1$ oscillatory media can be divided into two classes: stable spirals with phase-locked arms (single arm in $1^0$, twin-armed in $1^1$ oscillatory media) and amplitude-modulated spirals.

Again, we employed $a_2$ and $b_2$ as the control parameters with the other parameters given in Table I. As expected, we found only stable single-armed spiral waves with constant amplitude except in the spiral core when the homogeneous kinetics corresponded to simple $1^0$ oscillatory media. On increasing either $a_2$ or $b_2$ into the mixed-mode region, the phase locking across the spiral breaks down, and the amplitude of the spiral arm begins to oscillate. Meanwhile, an adjunct arm, caused by the small peak of the $1^1$ oscillations, also emerges and vanishes periodically. Figures 3(a) and 3(b)

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![Fig. 2. Traveling wave-packets in a $1^1$ mixed-mode oscillatory medium with $a_2 = 0.09$, $b_2 = 1.60$ (other parameters are given in Table I). (a) Traveling wave packets in space; red line shows the superstructure of amplitude modulation; (b) local dynamics of traveling wave packets; (c) space-time plot for modulated traveling waves. $K_1$ and $K_2$ indicate the propagation of the basic traveling waves and modulated structures, respectively.](image-url)
show a snapshot of this modulated spiral wave and the amplitudes of the wave arms from the spiral core to the boundary, respectively. For still larger $a_2$, the increasing modulation can lead to instability of the spiral waves, which causes breakage of the spiral far from the core. Alternatively, the amplitude modulation of the spiral arms may subside as the parameters shift away from the fold bifurcation curve between 10 and 11 oscillations, resulting in steady twin-armed spirals with 11 local oscillations.

We have recorded and analyzed the local dynamics of the modulated patterns. In the case of modulated spirals, the local dynamics periodically moves between 10 (red orbit) and 11 (black orbit) oscillatory trajectories, as shown in Fig. 3(c). The Poincaré map for the attractor forms a ring, which indicates that the system oscillates quasiperiodically. Similar to the case of the traveling wave packets shown in Fig. 2, the time series in this case also consists of 10 and 11 oscillations with periodically modulated amplitudes. Two fundamental frequencies can be observed in the Fast Fourier Transform (Fig. 3(d)) of the time series: $f_b$ is the frequency of the basic oscillations, and $f_m$ represents the frequency of the modulation.

C. Analysis of local dynamics in the reaction-diffusion system

To provide further insight into these modulated patterns in mixed-mode oscillatory media, we calculated the frequencies of local oscillations of the reaction-diffusion model in the $a_2$-$b_2$ parameter space. Figures 4(a) and 4(b) show phase diagrams of the fundamental frequencies, $f_b$ and $f_m$, in the $a_2$-$b_2$ plane. As the color maps indicate, the primary frequency $f_b$ decreases smoothly with both $a_2$ and $b_2$ (Fig. 4(a)), i.e., the reaction-diffusion behavior shows the same trends found in the point system.

As noted above, the second fundamental frequency, $f_m$, appears when amplitude-modulated spirals emerge in the system. The phase diagram of $f_m$ shown in Fig. 4(b) thus characterizes the regions of existence and the transitions between stable and modulated spiral waves. The white regions represent single frequency states ($f_m = 0$), whereas the color region corresponds to modulated spirals with two frequencies ($f_m \neq 0$). For comparison, the bifurcation curves calculated in homogenous media without diffusion are also shown in Fig. 4(b).

As we increase $b_2$, crossing the first fold bifurcation curve, we observe a supercritical region of 10 local oscillations, in which the dynamical behavior in homogenous media would be 11 oscillations. Due to the damping effect of diffusion, the reaction-diffusion system continues to support single-armed spirals (local 10 oscillations, $f_m = 0$) in this region. Next, a transition zone with $f_m \neq 0$ emerges between the steady 10 and 11 oscillatory regions. Here, the local kinetics periodically switches between 10 and 11 oscillations. These quasi-periodic local oscillations are associated with modulated spiral waves in the reaction-diffusion system. When the parameters are increased further to take the system far from the fold bifurcation curve, only twin-armed spirals with steady amplitude and 11 local dynamics can be obtained.
Notice also that a period-2 bifurcation curve (blue dashed line in Fig. 4(b)) is embedded in the transition zone. Previous studies have found that spiral waves can undergo period-doubling bifurcations and lose stability by generating defect lines.11–13 Here, instead of the defect lines, modulated patterns are generated as diffusion transforms the period-doubled oscillations into toroidal oscillations.

After the second fold bifurcation between 11 and 12 oscillations in the homogenous model, the damping effect of diffusion can prevent the emergence of 12 oscillations and generate a supercritical region of 11 oscillations. Although the local dynamics remain 11 oscillations, the spiral waves in some supercritical regions (a2 = 0.09–0.11) may lose stability and break up at larger b2. For b2 > 1.95, periodic switching between 11 and 12 oscillations occurs in the local dynamics, so a second fundamental frequency reappears in the phase diagram, as shown in Fig. 4, i.e., there is also a transition zone between 11 and 12 mixed-mode oscillations in reaction-diffusion media. However, the spiral waves are not stable in this transition zone for D1 = D2 = 0.1.

The ratio of diffusion coefficients, δ = D1/D2, was used as a control parameter in the three-variable model,17 which demonstrated that the local dynamics of the spatiotemporal system approach those of the homogenous system at lower values of δ. We decreased D1 to 0.01, i.e., δ = 0.1, and recalculated the system behavior in the a2-b2 parameter space. As shown in Fig. 5(c), the transition zone shrinks and shifts to the left, while a new zone of period-doubled oscillations emerges. These two behaviors are separated by a band of simple 11 oscillations. As illustrated in Fig. 5(a), we observe twin-armed spirals with defect lines in the zone of period-doubled oscillations. We see that the instability caused by the period-doubling bifurcation is distinct from the region of modulated waves. We conclude that the period-doubling bifurcation is not responsible for the modulated patterns in mixed-mode oscillatory media. Rather, the modulations are generated by a secondary bifurcation of the attractor associated with the fold bifurcation under suitable conditions.

Furthermore, the transition zone between 11 and 12 oscillations can be clearly observed in the case of δ = 0.1. As shown in Fig. 5(b), amplitude-modulated spirals with regular alternation of twin-arm waves and triplet-arm waves, which have local dynamics containing both 11 and 12 oscillations, can be generated in this region. Meanwhile, three-armed spirals with defect lines can also be obtained in the period-2 zone of the 12 oscillatory region. These modulated patterns between the 11 and 12 oscillatory regions suggest that similar transition zones arise between higher 1N-1 and 1N mixed-mode oscillations as well for appropriate diffusion parameters, where the amplitude-modulated complex patterns result from periodic transition between (N − 1)-armed and N-armed waves.

D. Spatial recurrence rates

Characterizing the structure and stability of complex spatiotemporal patterns is a challenging problem. One approach that has been suggested is the use of spatial recurrence rates.28,29 Recurrence analysis has been widely used in the analysis of time series or one-dimensional plots.30 We apply it here to assess whether it can yield insight into the patterns we observe.

For two-dimensional patterns, the recurrence matrices are

\[ R(i,j,1,2,j2) = \Theta(\varepsilon - ||v_i,j1 - v_i,j2||), \quad i_k,j_k \in (1,N), \]

where \( v(i,j) \) represents the concentration of component \( v \) for each site \((i,j)\) of the pattern; \( \Theta \) is the Heaviside step function; \( || \cdot || \) is a norm; and \( N \times N \) is the number of sites in the system. The binary recurrence matrices indicate the spatial recurrences that occur in the 2-D phase space of patterns within a tolerance \( \varepsilon \). \( R(i,j,1,2,j2) \) is a four-dimensional matrix, which does not lend itself to visualization. However, the recurrence rate (Rr) of a pattern can be calculated as a quantity that characterizes its spatiotemporal structure

\[ Rr = \frac{1}{N^2} \sum_{(i,j1,j2) = 1}^{N} R(i,j1,j2). \]

For homogeneous media, where all points in the system have the same phase, the recurrence rate is 1.0. Here we fix \( b_2 = 1.70 \) and vary \( a_2 \), monitoring the spatial recurrence rates corresponding to the modulation of spiral waves shown in Fig. 4(b). The emergence of modulated patterns is seen to reduce the recurrence rate significantly. Figure 6 shows that
amplitude-modulated spirals have smaller recurrence rates than simple spirals. The spatial recurrence rate reaches a minimum, which corresponds to the breaking of modulated spirals. $Rr$ then begins to increase again as we raise $a_2$, approaching its initial maximum (single-armed spirals at $a_2 = 0.08$) when the pattern consists of stable twin-armed spirals without amplitude modulation ($a_2 > 0.11$). The spatial recurrence rates are thus quite sensitive to the characteristics of the spatial pattern and reflect the distinction between simple and modulated structures.

**IV. CONCLUSIONS**

The model we have investigated contains one substrate inhibitor and two activators. The coupling between these species under homogeneous conditions generates several types of mixed-mode oscillations. This coupling is inevitably influenced by the effects of diffusion. Close to the fold bifurcation between $1^0$ and $1^1$ oscillations, the synchronization caused by diffusion can shrink the basin of stability of $1^1$ oscillations relative to that of $1^0$ oscillations. In the parameter regions further away from the bifurcation, diffusion can no longer suppress the fold bifurcation. A periodic switching between $1^0$ and $1^1$ oscillatory modes may then emerge, resulting in a secondary Hopf bifurcation, which generates local time series containing oscillation packets and spatial patterns with amplitude modulation. Still stronger modulation of the waves may destroy the stability of these patterns and lead to turbulence for some sets of parameters. The fraction of $1^1$ oscillations in the time series of local dynamics grows as the system moves away from the fold bifurcation curve into the $1^1$ oscillatory region. Finally, stable twin-armed spiral waves of $1^1$ appear. In our simulations, modulated patterns can also be found after the fold bifurcation curves between $1^1$ and $1^2$ oscillations if we reduce the ratio of diffusion coefficients, as indicated in Figs. 4 and 5. The above results support the notion that such transition states occur in regions between $1^{N-1}$ and $1^N$ mixed-mode oscillations for higher values of $N$ as well in reaction-diffusion systems when the diffusion coefficients are chosen appropriately.

We conclude that amplitude-modulated patterns composed of alternating $(N-1)$-armed and $N$-armed waves in spatially distributed mixed-mode oscillatory media result from periodic transitions between $1^{N-1}$ and $1^N$ local oscillations. We have identified such transition regions in the phase diagram for the chosen reaction-diffusion model and used recurrence rate analysis to characterize the various amplitude-modulated patterns.

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