Control of the Hopf-Turing transition by time-delayed global feedback in a reaction-diffusion system

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Application of time-delayed feedback is a practical method of controlling bifurcations in reaction-diffusion systems. It has been shown that for a suitable feedback strength, time delay beyond a threshold may induce spatiotemporal instabilities. For an appropriate parameter space with differential diffusivities of the activator-inhibitor species, delayed feedback may generate Turing instability via a Hopf-Turing transition, resulting in stationary patterns. This is explored by a theoretical scheme in a photosensitive chlorine dioxide–iodine–malonic acid reaction-diffusion system where the delayed feedback is externally tuned by photoillumination intensity. Our analytical results corroborate with direct numerical simulations.

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I. INTRODUCTION

The interaction of reaction and diffusion in a system far from equilibrium may generate various kinds of spatiotemporal instabilities, for example, stationary patterns, spirals, targets, traveling waves, solitons, and oscillons [1]. Among these, Turing patterns have been widely studied in the past several decades [2]. The underlying instability owes its origin to short-range activation and long-range diffusion of two interacting species obeying activator-inhibitor reaction kinetics, and the resulting symmetry-broken state is characterized by ordered structures or patterns. The far-from-equilibrium states are intrinsically guided by the nonlinearity in the kinetic terms and the diffusivities of the species present in the system. The first unambiguous experimental evidence of a convection-free Turing pattern [3] was reported in a thermodynamically open chlorite–iodide–malonic acid (CIMA) reaction-diffusion system. Since then, a lot of experimental and theoretical studies on Turing instabilities have been made in this system [4,5] and its variant chlorine dioxide–iodine–malonic acid reaction (CDIMA) [6,7].

Controlling spatial or spatiotemporal instabilities and pattern formation is a growing area of interest in this context. The influences of external perturbations such as light, electric field, magnetic field, and noise play major roles in controlling and modulating patterns when the parameter space of the dynamics is appropriately tuned [8–10]. For example, in the case of a reaction-diffusion system with ionic species, the electric field may affect mass transport, resulting in symmetry-breaking spatial structures [8,9]. For photosensitive reaction-diffusion systems, depending upon the photoillumination intensity, the excitation by light is very effective in inducing various types of spatial organizations and crossover between them [11,12]. Apart from electric fields and light, noise in both additive and multiplicative forms [13–16] has been used to induce ordered spatial structures. Another interesting area covering external effects on pattern-forming processes concerns the application of the forces generated by the system itself (i.e., feedback). The feedback can also be generated and manipulated externally in a system. A convenient method of feedback control is based on the use of appropriate time delay on the dynamical system. Since time delay is important in many natural systems, its effect can be crucial and instrumental in the system dynamics. An early attempt in this direction was made by Ott *et al.*, who employed input signals constructed from the difference between the current and past states [17]. Another simple and efficient scheme of time-delayed feedback control is given by Pyragas [18,19]. Delayed feedback and its modifications are widely used to control chaos and to stabilize unstable oscillations. This has been useful in controlling multiple rhythms in a self-sustained oscillator [20–22]. Control of pattern in the form of time-delayed feedback in spatially extended systems has been investigated in the past few years [23–27]. For example, recently, it has been shown that applying localized feedback at a few spatial locations can stabilize both uniform and pattern states. Local delayed feedback has been used to orient spatial patterns in various forms and to induce and manipulate spiral dynamics [28–30]. The majority of these concern the influence of delayed feedback on systems in pattern-forming regions for delayed feedback, revealing that the original patterns can be modulated or even suppressed by the delayed feedback. In this paper, we focus on the application of a global feedback control involving short time delays on a photosensitive CDIMA reaction-diffusion system. This model has remained a classic paradigm for both experimental and theoretical studies for a wide class of far-from-equilibrium phenomena over the years. Global feedback control implies that, based on the measurements of some global characteristics of the system, we may vary some parameter that determines the system behavior in the whole domain. Such type of control is easier to implement experimentally. An important attempt in this direction has been made in [23], where pattern formation in catalytic CO oxidation on a Pt surface has been observed by controlling chemical turbulence using global delayed feedback. In this paper, time-delayed feedback is externally imposed and manipulated by photoillumination intensity in the non-pattern-forming region where the system without the feedback is a spatially uniform state. A simple theoretical scheme is worked out to determine the instability criteria in the presence of delayed feedback, which ensures a Hopf-Turing transition due to the feedback initiating the formation of stationary patterns. Our theoretical analysis is corroborated by numerical simulations.

The paper is organized as follows: In the next section, we explore analytically the influence of time-delayed feedback on

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the photosensitive CDIMA system to determine the instability conditions. Our theoretical analysis is verified by numerical simulation of the spatially extended dynamical equations of the reaction-diffusion system. The paper is concluded in Sec. III.

II. ANALYSIS OF TIME-DELAYED FEEDBACK IN REACTION-DIFFUSION SYSTEM

A. The model and preliminary discussion

To demonstrate the effect of time-delayed feedback in controlling the pattern-forming process, we choose the modified Lengyel-Epstein model as a prototypical two-variable reaction-diffusion system. The reaction includes the illumination effect for the photosensitive CDIMA system:

$$\frac{\partial u}{\partial t} = a - u - \frac{4uv}{1+u^2} - \phi + \nabla^2 u, \tag{2.1}$$

$$\frac{\partial v}{\partial t} = \sigma \left[b \left(u - \frac{uv}{1+u^2} + \phi \right) + d\nabla^2 v \right]. \tag{2.2}$$

Here u and v are dimensionless concentrations of I^- and ClO_2^-, respectively. They are dimensionless parameters; a and b are related to kinetic parameters and are proportional to the concentration ratios $[CH_2(COOH)_2]/[ClO_2]$ and $[I_2]/[ClO_2]$, respectively, where $[ClO_2]$, $[I_2]$, and $[CH_2(COOH)_2]$ are in large excess. Variable d denotes the ratio of the diffusion coefficients, $d = [D_{ClO_2^-}]/[D_{I^-}]$; σ refers to the concentration of starch, which forms a complex with I_3^- such that $\sigma = 1 + K[S]$. Here K is the equilibrium constant for the starch-iodide complex and $[S]$ is the concentration of starch tri-iodide binding sites. The parameter σ is controlled by the concentration of starch. Here ϕ refers to the dimensionless rate of photochemical reaction, which is proportional to the external light intensity. The feedback is introduced by external illumination, which becomes a time-dependent quantity when feedback is present and has the form $\phi(t) = \phi_0 - P[v(t-\tau) - v(t)]$, where P is the feedback intensity of the control strategy, $v(t-\tau)$ denotes the delayed concentration with respect to present variables, τ is the effective delay time, and ϕ_0 is the reference light intensity, which is a constant. $\phi = \phi_0$ when there is no feedback in the system. We confine our treatment to short time delay for the present purpose.

It is well known from the linear stability analysis of the system that by varying the complexing agent (σ) one can adjust the Hopf curve in the $b-a$ parameter plane in such a way that it lies below the Turing bifurcation curve. The Turing curve is independent of the value of σ, and below it the homogeneous steady state is unstable to inhomogeneous perturbation. The region above the Hopf curve is homogeneously stable. Turing patterns arise in the region below the dashed Turing curve and above the solid Hopf curve. This can be seen from the inset in Fig. 1. Introduction of delayed feedback may give us another useful handle for further manipulation of the instability region between Hopf and Turing curves. We proceed to explore this in the next section.

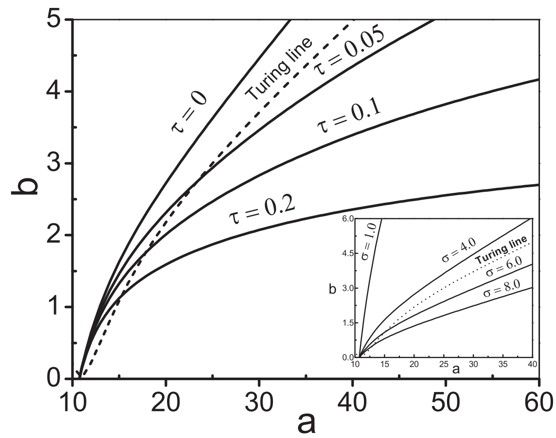

FIG. 1. Bifurcation diagram for $b-a$ parameter region for $P = 0.2$, $d = 1.6$, and $\phi_0 = 1.0$ and for fixed $\sigma = 4.0$. The solid curves denote Hopf bifucation curves. Inset figure represents $b-a$ bifurcation diagram for different values of σ in the absence of time-delayed feedback.

B. Theoretical analysis of the system under feedback control

In the presence of time-delayed feedback externally manipulated by the illumination intensity $\phi(t)$, the governing equations for the CDIMA system become

$$\begin{aligned}\frac{\partial u}{\partial t} &= a - u - \frac{4uv}{1+u^2} - \{\phi_0 - P[v(t-\tau) - v(t)]\} + \nabla^2 u \\ &= f(u,v) + P[v(t-\tau) - v(t)] + \nabla^2 u \end{aligned} \tag{2.3}$$

and

$$\begin{aligned}\frac{\partial v}{\partial t} &= \sigma \left[b \left(u - \frac{uv}{1+u^2} + \{\phi_0 - P[v(t-\tau) - v(t)]\} \right) + d\nabla^2 v \right] \\ &= g(u,v) - \sigma b P[v(t-\tau) - v(t)] + \sigma d \nabla^2 v, \end{aligned} \tag{2.4}$$

where

$$\begin{aligned} f(u,v) &= a - u - \frac{4uv}{1+u^2} - \phi_0, \\ g(u,v) &= \sigma \left[b \left(u - \frac{uv}{1+u^2} + \phi_0 \right) \right]. \end{aligned} \tag{2.5}$$

Assuming τ to be small, we replace $v(x,y,t-\tau)$ as $[v(x,y,t) - \tau \frac{\partial v(x,y,t)}{\partial t}]$ in Eqs. (2.3) and (2.4) and write it as

$$\frac{\partial u}{\partial t} = f(u,v) - P\tau \frac{\partial v(t)}{\partial t} + \nabla^2 u, \tag{2.6}$$

$$\frac{\partial v}{\partial t} = g(u,v) + \sigma b P \tau \frac{\partial v(t)}{\partial t} + \sigma d \nabla^2 v. \tag{2.7}$$

Now, by rearranging the above two equations, we finally obtain the following expressions:

$$\begin{aligned}\frac{\partial u}{\partial t} &= f(u,v) - \left(\frac{\tau P}{1 - \sigma b \tau P} \right) g(u,v) + \nabla^2 u \\ &\quad - \left(\frac{\tau P}{1 - \sigma b \tau P} \right) \sigma d \nabla^2 v, \end{aligned} \tag{2.8}$$

$$\frac{\partial v}{\partial t} = \left(\frac{1}{1 - \sigma b \tau P} \right) g(u,v) + \left(\frac{1}{1 - \sigma b \tau P} \right) \sigma d \nabla^2 v. \tag{2.9}$$

The homogeneous steady states of the dynamical system are the fixed points u_0 and v_0 defined by $f(u_0,v_0) = 0$, $g(u_0,v_0) = 0$, which is obtained as $u_0 = a/5 - \phi_0$ and $v_0 = a(1 + u_0^2)/5u_0$.

To see how the system responds when it is perturbed, we consider small spatiotemporal perturbations $\delta u(x,y,t)$ and $\delta v(x,y,t)$ around the homogeneous steady state (u_0,v_0) as follows:

$$\begin{aligned} u(x,y,t) &= u_0 + \delta u(x,y,t),\\ v(x,y,t) &= v_0 + \delta v(x,y,t). \end{aligned} \tag{2.10}$$

Linearizing the dynamical system around the steady state (u_0,v_0), we obtain

$$\begin{aligned} \frac{\partial(\delta u)}{\partial t} &= (f_u - \chi\tau P g_u + \nabla^2)\delta u\\ &\quad + (f_v - \chi\tau P g_v - \chi\tau P\sigma d\nabla^2)\delta v, \end{aligned} \tag{2.11}$$

$$\frac{\partial(\delta v)}{\partial t} = \chi g_u \delta u + \chi(g_v + \sigma d\nabla^2)\delta v. \tag{2.12}$$

Here $(\frac{1}{1-\sigma b\tau P})$ is abbreviated as χ.

By expressing spatiotemporal perturbation $\delta u(x,y,t)$ and $\delta v(x,y,t)$ in the form

$$\begin{aligned} \delta u(x,y,t) &= \delta u_0 e^{\lambda t}\cos k_x x \cos k_y y,\\ \delta v(x,y,t) &= \delta v_0 e^{\lambda t}\cos k_x x \cos k_y y, \end{aligned} \tag{2.13}$$

and inserting them into Eqs. (2.11) and (2.12), we obtain the following matrix equation for eigenvalues of the stability matrix.

$$\begin{pmatrix} (f_u - \chi\tau P g_u - k^2) - \lambda & (f_v - \chi\tau P g_v + \chi\tau P\sigma d k^2)\\ \chi g_u & \chi(g_v - \sigma d k^2) - \lambda \end{pmatrix} \times \begin{pmatrix} \delta u_0\\ \delta v_0 \end{pmatrix} = 0, \tag{2.14}$$

where $k^2 = k_x^2 + k_y^2$.

By expanding the Jacobian matrix, we get the following quadratic equation for the eigenvalues of the associated stability matrix:

$$\begin{aligned} &\lambda^2 - [f_u + \chi g_v - \chi\tau P g_u - (1 + \chi\sigma d)k^2]\lambda\\ &\quad + \chi[(f_u g_v - g_u f_v) - k^2(\sigma d f_u + g_v) + \sigma d k^4] = 0. \end{aligned} \tag{2.15}$$

We now insert the explicit form of χ in Eq. (2.15), and after a little rearrangement we obtain the following form:

$$\begin{aligned} &\lambda^2 - \frac{[f_u + g_v - \tau P(\sigma b f_u + g_u) - (1 + \sigma d - \tau P\sigma b)k^2]}{(1 - \tau P\sigma b)}\lambda\\ &\quad + \frac{[(f_u g_v - g_u f_v) - k^2(\sigma d f_u + g_v) + \sigma d k^4]}{(1 - \tau P\sigma b)} = 0. \end{aligned} \tag{2.16}$$

Our aim here is to find out the threshold value of the delay time for which the system with delayed feedback, which is otherwise stable with respect to homogeneous perturbation, becomes unstable. For the homogeneous case, Eq. (2.16) can be written in the form

$$\lambda^2 - A\lambda + B = 0, \tag{2.17}$$

where

$$A = \frac{[f_u + g_v - \tau P(\sigma b f_u + g_u)]}{(1 - \tau P\sigma b)}$$

and

$$B = \frac{(f_u g_v - g_u f_v)}{(1 - \tau P\sigma b)}.$$

The condition for stability of the homogeneous steady state for the system with delayed feedback is $A < 0$ and $B > 0$. B is positive if $(1 - \tau P\sigma b) > 0$ since $(f_u g_v - g_u f_v)$ is greater than 0 for homogeneous system. Now the condition $A < 0$ can be obtained when $[f_u + g_v - \tau P(\sigma b f_u + g_u)] < 0$ and $(1 - \tau P\sigma b) > 0$. This allows the range of values for τ for a fixed value of feedback strength determined by the following condition:

$$\frac{f_u + g_v}{P(\sigma b f_u + g_u)} < \tau < \frac{1}{P\sigma b}. \tag{2.18}$$

Now let us consider the effect of diffusion on the dynamical system and study how it destabilizes the homogeneous steady state of the system under delayed feedback to generate spatiotemporal instability, if any. We rewrite Eq. (2.16) in the form

$$\lambda^2 - Q\lambda + R = 0, \tag{2.19}$$

where

$$Q = \frac{[f_u + g_v - \tau P(\sigma b f_u + g_u) - (1 + \sigma d - \tau P\sigma b)k^2]}{(1 - \tau P\sigma b)}$$

and

$$R = \frac{[(f_u g_v - g_u f_v) - k^2(\sigma d f_u + g_v) + \sigma d k^4]}{(1 - \tau P\sigma b)}.$$

We now impose the condition of stability of homogeneous steady state ($f_u + g_v < 0$ and $f_u g_v - g_u f_v > 0$). This ensures the positivity of R when the ratio of the diffusion coefficients is unity ($d = 1.0$) with $\sigma = 1.0$. Since the numerator of R determines the Turing curve, which is found to be independent of τ and P, any delayed-feedback-induced instability in the presence of diffusion must depend on Q. Therefore, the condition of instability is $Q > 0$. At the same time, since the denominator of Q is always positive, the condition of instability reduces to the following inequality:

$$[f_u + g_v - \tau P(\sigma b f_u + g_u) - (2 - \tau P\sigma b)k^2] > 0. \tag{2.20}$$

This implies that the lower bound of τ must satisfy

$$\tau > \left[\frac{2k^2 - (f_u + g_v)}{P(bk^2 - bf_u - g_u)}\right] = \tau_c, \tag{2.21}$$

which further leads to the condition $k^2 > (f_u + g_u/b)$ and gives a measure of wavelength scale of the system when the symmetry is broken.

Therefore, for a fixed parameter set and feedback strength P, time delay beyond a critical threshold τ_c asserts a condition of instability of the homogeneous steady state of the system even if the ratio of the diffusion coefficients of the species is unity. However, it can be observed from the detailed numerical simulations of the partial differential equations carried out in the next section that this instability does not give rise to any spatial structures. When the ratio of diffusion coefficients of the two species (i.e., activator and inhibitor) is not unity ($d \neq 1$), then the system exhibits symmetry-breaking spatial structures. Hence, the condition for instability is actually

$$\tau > \frac{(1+\sigma d)k^2 - (f_u + g_v)}{P[\sigma b(k^2 - f_u) - g_u]} = \tau_c, \tag{2.22}$$

which finally gives the condition $k^2 > (f_u + g_u/\sigma b)$.

Hence, the conclusion is that for a suitable feedback strength, a time delay τ beyond the threshold value can generate spatiotemporal instability in the two-component activator-inhibitor system when the system without delayed feedback remains homogeneously stable. It can be shown that the Hopf bifurcation curve for the system in the presence of delayed feedback ($\tau \neq 0$) is given by

$$b = \frac{5u_0\left(1+u_0^2\right) + 4a\left(1-u_0^2\right)}{5\sigma\left[\tau P a\left(1-u_0^2\right) - u_0^2\right]}, \tag{2.23}$$

whereas the Turing bifurcation curve is given by

$$b = \frac{d}{5u_0^2}\Big[45u_0\left(1+u_0^2\right) - 4a\left(1-u_0^2\right) - 20\sqrt{u_0\left(1+u_0^2\right)\left[5u_0\left(1+u_0^2\right) - a\left(1-u_0^2\right)\right]}\Big], \tag{2.24}$$

where $u_0 = (a/5 - \phi_0)$.

Expression (2.24) reveals that the Turing bifurcation curve is unaffected by the presence of delayed feedback. Therefore,

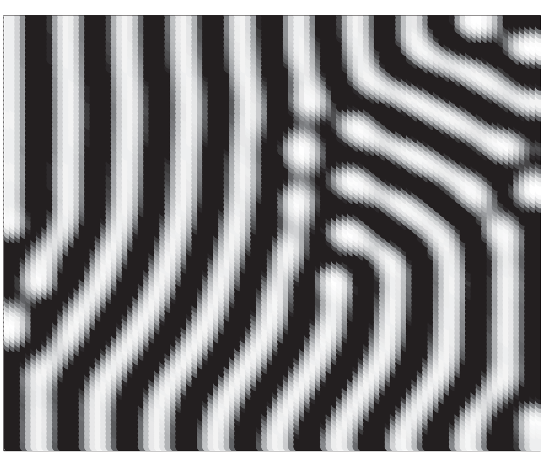

FIG. 2. Numerically simulated (in two-dimensional space; grid size 100×100 with $\Delta x = \Delta y = 0.50$ and $\Delta t = 0.0025$) delayed-feedback-induced spatial instability in a photosensitive CDIMA system for the parameters $a = 18.0$, $b = 1.5$, $P = 0.2$, $\phi_0 = 1.0$, $\sigma = 4.0$, $d = 1.6$, and $\tau = 0.2$ (delayed feedback in Hopf region). White corresponds to a higher concentration of iodide (u).

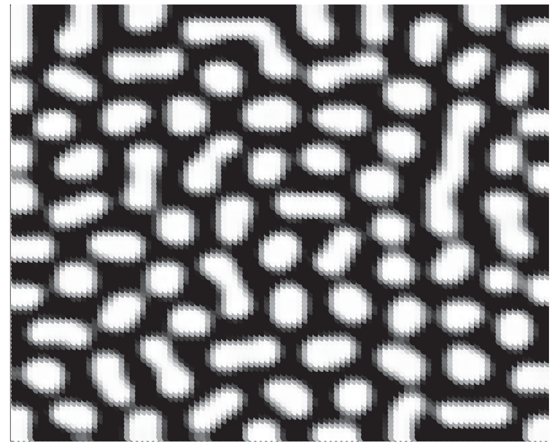

FIG. 3. Numerically simulated (in two-dimensional space; grid size 100×100 with $\Delta x = \Delta y = 0.50$ and $\Delta t = 0.0025$) delayed-feedback-induced spatial instability in a CDIMA system for the parameters $a = 36.0$, $b = 3.5$, $P = 0.2$, $\phi_0 = 1.0$, $\sigma = 4.0$, $d = 1.6$, and $\tau = 0.1$ (delayed feedback in Hopf region). White corresponds to a higher concentration of iodide (u).

it is noteworthy that Hopf bifurcation curve can be suitably adjusted in the b-a plane by the appropriate choice of time delay (τ) for a given feedback strength (P) so that it crosses the Turing curve. By adjusting the Hopf curve, the region of instability can be modified by proper tuning of the delay time. This is shown in Fig. 1 for the parameter set $\phi_0 = 1.0$, $d = 1.6$, $\sigma = 4.0$ and for a fixed feedback strength $P = 0.2$. By increasing the time delay τ, the Hopf curve crosses the Turing curve to give rise to a Turing pattern. The nature of the Hopf curve is such that it crosses the Turing curve for higher values of a and b. With increases of time delay, the Turing region expands for lower values of a and b (i.e., the Turing region is widened for larger τ).

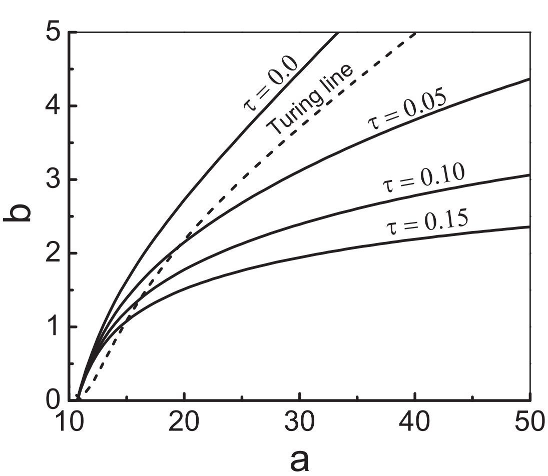

FIG. 4. Bifurcation diagram for $b - a$ parameter region for $P = 0.3$, $d = 1.6$, and $\phi_0 = 1.0$ and a fixed $\sigma = 4.0$; the solid curves denote Hopf bifucation curves.

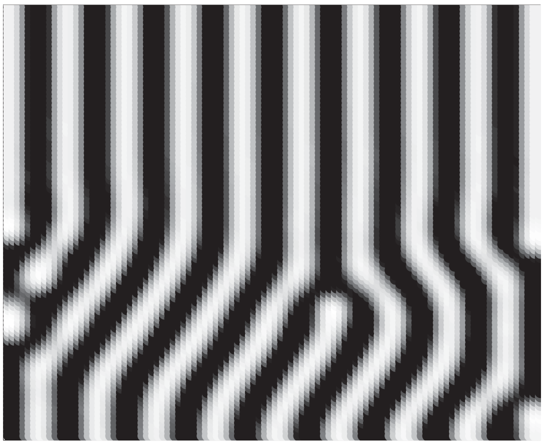

FIG. 5. Numerically simulated (in two-dimensional space; grid size 100 × 100 with $\Delta x = \Delta y = 0.50$ and $\Delta t = 0.0025$) delayed-feedback-induced spatial instability in a CDIMA system for the parameters $a = 18.0$, $b = 1.5$, $P = 0.3$, $\phi_0 = 1.0$, $\sigma = 4.0$, $d = 1.6$, and $\tau = 0.15$ (delayed feedback in Hopf region). White corresponds to a higher concentration of iodide (u).

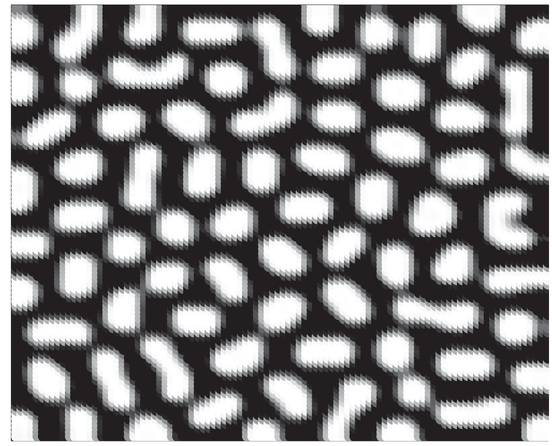

FIG. 6. Numerically simulated (in two-dimensional space; grid size 100 × 100 with $\Delta x = \Delta y = 0.50$ and $\Delta t = 0.0025$) delayed-feedback-induced spatial instability in a CDIMA system for the parameters $a = 36.0$, $b = 3.5$, $P = 0.3$, $\phi_0 = 1.0$, $\sigma = 4.0$, $d = 1.6$, and $\tau = 0.05$ (delayed feedback in Hopf region). White corresponds to a higher concentration of iodide (u).

C. Numerical results

In order to compare the analytical predictions from the aforesaid analysis, we carry out numerical simulations of the system under the influence of delayed feedback, using Eqs. (2.3) and (2.4), by the explicit Euler method with parameter set (experimentally admissible values) $a = 18.0$, $b = 1.5$, $d = 1.6$, $\phi_0 = 1.0$, and $P = 0.2$. The computations have been performed on a 100 × 100 array with grid spacing $\Delta x = \Delta y = 0.50$, time step $\Delta t = 0.0025$, and zero flux boundary condition. The simulations are started with spatially random perturbations of ∼1% around the steady state $u_0 = (a/5 - \phi_0)$, $v_0 = a(1 + u_0^2)/5u_0$ and for $\sigma = 4.0$, which is a Hopf region. The system without delay ($\tau = 0$) remains homogeneous in this parameter regime. The development of a typical delayed-feedback-induced pattern in the form of labyrinthine stripes for the above-mentioned parameter set is shown in Fig. 2 for $\tau = 0.2$. This clearly demonstrates a Hopf-Turing transition induced by delay (τ). It is also noted from the analytical treatment (Fig. 1) that the Turing region appears for much lower value of delay time τ in higher range of $b - a$ parameter space. In order to explore this observation further, we have numerically simulated the system with $a = 36.0$ and $b = 3.5$, with other parameters remaining the same and found that inhomogeneity in the form of spots arises for much lower τ (=0.1) value shown in Fig. 3.

It is pertinent to understand to what extent the dynamics is sensitive to the choice of P, the feedback strength. A simple consideration shows that a change in the strength of feedback to $P = 0.3$, gives rise to a Turing pattern with much lower value of time delay (τ). This can be seen from Fig. 4. It indicates that the Hopf curve crosses the Turing bifurcation curve for a smaller value of delay time in comparison to the corresponding situation for lower feedback strength. The results on variation of delay time are shown in the bifurcation diagram, Fig. 4. The numerical simulation of Eqs. (2.3) and (2.4) is further extended to the case for higher feedback strength $P = 0.3$. The results are shown in Figs. 5 and 6. The results on numerical simulations agree well with the predictions from the bifurcation diagram.

III. CONCLUSION

We have proposed a simple approach to study the effect of global delayed-feedback-induced instabilities in two-component reaction-diffusion systems. This method is particularly suitable for short time delay (τ), which is relevant for chemical systems as compared to biological systems. In the latter cases, where the delay time is much longer, one needs a more careful scrutiny by taking care of the joint effects of diffusion and delay [31]. The technique involves stability conditions based on transcendental equation where the relation between characteristic eigenvalue λ appears as a function of $e^{\lambda\tau}$. Our analysis reveals that for a suitable feedback strength, delay time can be an useful handle to control Hopf-Turing space. By tuning the value of delay time, the Hopf-bifurcation curve can be manipulated in such a way that it can give rise to symmetry-breaking spatial structures in the form of Turing patterns. The theoretical predictions have been tested on a chlorine dioxide–iodine–malonic acid system by numerical simulations. Although the present study of dynamical control of patterns by delayed feedback has been carried out on a specific model as a prototypical example, we believe that our results on delayed-feedback-induced instability are likely to be important for other reaction-diffusion systems.

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