



## Investigating the effect of circularly polarized electric field on spatially extended Gray-Scott model

Amitava Giri and Sandip Kar\*

Department of Chemistry, Indian Institute of Technology Bombay, Powai, Mumbai-400 076, India

E-mail: sandipkar@iitb.ac.in

Manuscript received online 22 April 2019, revised and accepted 15 May 2019

---

According to Turing's hypothesis, the spatially heterogeneous pattern emerges from a reaction-diffusion system due to the diffusion driven instability caused by the difference in diffusivities of the activator and inhibitor like reactant species. However, reaction-diffusion systems consisting of ionic reactants often lead to interesting spatiotemporal structures under the influence of constant external electric or magnetic field even in absence of diffusion driven instability. Unfortunately, only stripe like pattern arises in presence of such kind of external electric field. Herein, we explored the effect of circularly polarized electric field on a spatially extended model reaction-diffusion system (Gray-Scott model), and showed that under the influence of such kind of periodic perturbation, a diverse range of spatial patterns can be obtained both in presence and absence of diffusion driven instability. Interestingly, the long time limit spatiotemporal structures emerged under the presence of circularly polarized electric field are found to be evolving with time due to the intrinsic time dependent nature of the external perturbation. Thus, our numerical study provides a way to obtain diverse spatiotemporal patterns by circumventing the stringent condition of diffusion-driven instability, which can be easily verified by performing experiments with ionic reaction-diffusion system.

Keywords: Turing pattern, Gray-Scott model, electric field, periodic perturbation.

---

### Introduction

Spatiotemporal pattern formation in chemical and biological systems is an intriguing and exciting phenomena observed in nature<sup>1,2</sup>. Around the middle of the last century, mathematician Alan Turing first proposed the chemical theory of morphogenesis for spontaneous symmetry breaking during embryogenesis. He suggested that during early development of an embryo, appropriate interplay between reactions and diffusion of activator and inhibitor kind of chemical species caused the spontaneous symmetry breaking, and ultimately lead to stationary spatial structures. Importantly, these structures arise only when diffusion makes a homogeneous stable steady state into spatially inhomogeneous one. This phenomena was termed as diffusion driven instability<sup>3</sup>. Attainment of such a situation requires a significant difference between the diffusivities of the activator and inhibitor chemical species, which was difficult to achieve under laboratory condition<sup>4,5</sup>. Thus, it took about 40 years to prove Turing's idea about spatial pattern formation using a chemical system<sup>6</sup>.

The experimental verification of Turing's idea renewed the interest among the researchers to explore other reaction-diffusion systems both from experimental and theoretical standpoint to observe Turing kind of spatial patterns<sup>6-8</sup>. Nonetheless, creating a condition of wider difference of diffusivity between the inhibitor and activator, still remained to be a big challenge to overcome experimentally.

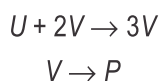
How to circumvent such a challenging experimental scenario? To answer this important question, efforts were made to overcome the stringent condition of diffusion driven instability by employing external perturbation in the form of magnetic field<sup>9,10</sup>, electric field<sup>11-13</sup>, light (for photosensitive reactions)<sup>14,15</sup> or by introducing relevant perturbations<sup>16-21</sup> in the corresponding reaction-diffusion systems. It has been found experimentally that most of the pattern forming reaction-diffusion systems are ionic in nature, and externally applied electric field can generate chemical waves in many such chemical systems<sup>22-26</sup>. In literature, it is reported that constant electric field perturbation can deform the existing Tur-

ing patterns obtained due to diffusion driven instability<sup>27–30</sup>, while the condition of wider difference of diffusivity of inhibitor and activator can be removed by employing external electric field under certain condition<sup>12,11,31</sup>. One disadvantage of employing such externally directed electric field is that the diversity in the range of spatiotemporal structure is lost. This is due to the fact that the directional field aligns the patterns in a specific direction after a while, and we mostly get stripe patterns of different form<sup>11,32</sup>. The obvious challenge is to obtain a more diverse range of spatial patterns even in presence of external perturbation, while subsiding the stringent condition on diffusivities of the reactants involved in different reaction-diffusion system.

In this article, we have investigated the effect of circularly polarized external electric field on a Gray-Scott like chemical reaction-diffusion system. In this regard, it is worth mentioning that Gray-Scott model system is basically an activation-substrate depletion<sup>33</sup> model system capable of manifesting different kind of spatial patterns under different parametric regime<sup>34</sup>, and there are chemical systems (like ferrocyanide-iodate-sulphite (FIS) reaction-diffusion system) that has been studied extensively experimentally and theoretically in terms of this model system<sup>32,34–36</sup>. Our endeavor here is to explore whether a similar range of diverse spatiotemporal patterns can be observed in absence of diffusion driven instability by simply employing circularly polarized external electric field or not. At the same time, we investigated the effect of the circularly electric field in presence of diffusion driven instability for the same system. Our numerical study demonstrates that in presence of a circularly polarized external electric field the Gray-Scott model system will be capable of demonstrating wide range of diverse spatiotemporal patterns in presence as well as in absence of diffusion driven instability.

### The Gray-Scott model

Here we have considered an autocatalytic reaction of two reactants  $U$  and  $V$  that follows Gray-Scott model kinetics.



The reactions are occurring in the above mentioned way in an open flow reactor, where  $U$  is constantly fed into the reactor, and the inert product is removed from the system. It is evident that the first reaction is the autocatalytic step, in which

the presence of  $V$  itself stimulates its own production. The second reaction is just a decay of  $V$  into product. That's why this reaction is called activation-substrate depletion kind of system, where  $V$  is playing role of activator and  $U$  is substrate. In literature, it is reported that the FIS-system mentioned earlier follows Gray-Scott kinetics<sup>35</sup>. This is an ionic reaction. Thus, if the reaction happens in presence of externally applied circularly polarized electric field, because of the ionic nature of the reactants, the spatiotemporal evolution of the reactants will be altered. To analyze the effect of such kind of electric field on the spatially extended Gray-Scott model, the corresponding reaction-diffusion equations in presence of externally applied (from two mutually perpendicular directions) circularly polarized electric field can be represented as;

$$\frac{\partial u}{\partial t} = f(u,v) + D_u \nabla^2 u + z_u D_u \left( E_x \frac{\partial u}{\partial x} + E_y \frac{\partial u}{\partial y} \right) \quad (1)$$

$$\frac{\partial v}{\partial t} = g(u,v) + D_v \nabla^2 v + z_v D_v \left( E_x \frac{\partial v}{\partial x} + E_y \frac{\partial v}{\partial y} \right) \quad (2)$$

Here  $f(u,v) = -uv^2 + F(1-u)$  and  $g(u,v) = uv^2 - (F+K)v$  are the non-dimensionalized reaction kinetics of  $U$  and  $V$ , where  $u(x,y,t)$  and  $v(x,y,t)$  are the two concentration variables of substrate ( $U$ ) and activator ( $V$ ) in the two dimensional space  $(x,y)$  considered.  $F$  and  $K$  are the dimensionless feed rate and rate constant, respectively. Rescaled diffusion coefficients of  $U$  and  $V$  are represented by  $D_u$  and  $D_v$ .  $E_x$  and  $E_y$  are the two applied circularly polarized electric field along  $x$  and  $y$  direction, where  $E_x = E_x^0 \cos(\omega t)$  and  $E_y = E_y^0 \cos(\omega t + \frac{3\pi}{2})$ .  $E_x^0$  and  $E_y^0$  are the rescaled electric field strength of the circularly polarized electric field along  $x$  and  $y$  direction. Along  $x$  and  $y$  direction, the applied electric field has a phase difference of  $\frac{3\pi}{2}$ , so basically we are applying a circularly polarized electric field to the spatially extended Gray-Scott model. Charges of the reactants  $U$  and  $V$  are  $z_u$  and  $z_v$ , respectively. Here we adopted the value of  $z_u$  and  $z_v$  from the FIS reaction system, where  $\text{HSO}_3^-$  is the substrate ( $U$ ) and  $\text{H}^+$  ion acts as catalyst ( $V$ )<sup>37</sup>.

First, we analyze the kinetic terms of eqs. (1) and (2) without considering diffusion and electric field related terms. Under long time limit, this system will give one or more than one steady states in different parametric regime. In every

parametric regime, the system shows the trivial steady state  $u_{ss}^0 = 1.0$  and  $v_{ss}^0 = 0.0$ , which is a linearly stable steady state. For other combinations of  $F$  and  $K$ , if  $F > 4(F + K)^2$  holds, then the system will have another two steady states:

$$u_{ss}^1 = \frac{1}{2} \frac{a}{c} - \sqrt{1 - \frac{4(F + K)^2}{F}}$$

$$v_{ss}^1 = \frac{1}{2} \frac{F}{F + K} \frac{a}{c} + \sqrt{1 - \frac{4(F + K)^2}{F}}$$

$$u_{ss}^2 = \frac{1}{2} \frac{a}{c} + \sqrt{1 - \frac{4(F + K)^2}{F}}$$

$$v_{ss}^2 = \frac{1}{2} \frac{F}{F + K} \frac{a}{c} - \sqrt{1 - \frac{4(F + K)^2}{F}}$$

The trivial steady state  $(u_{ss}^0, v_{ss}^0)$  is called ‘red state’ and  $(u_{ss}^1, v_{ss}^1)$  is called ‘blue state’, which may be stable or unstable<sup>36</sup>. In literature, attempts are made to obtain Turing kind of patterns either by perturbing the stable trivial steady state, or in the parametric regime where non-trivial ‘blue state’ exists. Pearson showed a wide variety of pattern can be obtained in Gray-Scott model in presence of diffusion driven instability, mostly by perturbing the trivial steady state by a strong local perturbation. In this report, we followed a numerical procedure similar to Pearson, while numerically simulating eqs. (1) and (2) in presence of circularly polarized external electric field along two mutually perpendicular directions in absence and presence of diffusion driven instability.

### Method

For numerical simulation of eq. (1) and eq. (2), we have employed explicit Euler method of integration under periodic boundary condition. The reaction-diffusion equations are discretized with a time step of 0.5 and with a spatial step size of 0.0098 along both  $x$  and  $y$  direction. The spatial mesh size for the simulation is  $256 \times 256$  (unless otherwise mentioned). In the parametric space, where the sole steady state is the trivial steady state  $(u_{ss}^0 = 1.0, v_{ss}^0 = 0.0)$ , we used the method followed by Pearson to perturb the system at the start of the simulation. Initially the whole system is placed on the trivial steady state  $(u_{ss}^0 = 1.0, v_{ss}^0 = 0.0)$  except the cen-

ter ( $20 \times 20$  mesh points) of the box. The center of the box is placed at a value of  $u = 0.5, v = 0.25$ . Now in order to break the square symmetry the initial values at the center, each grid points are perturbed with  $\pm 1\%$  random noise. For our numerical integration we have employed same methods for the parametric space, where the sole steady state is the trivial one. For other cases, where along with the trivial steady state another steady state usually coined as ‘blue state’ exits, we initially placed the entire grid with random noise around the steady state value of ‘blue state’, and performed our numerical simulation under the influence of circularly polarized external electric field in presence and absence of diffusion driven instability.

### Results and discussion

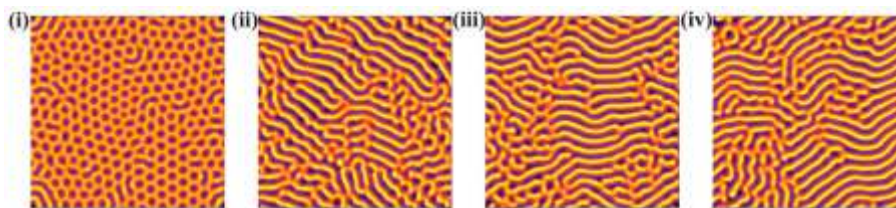
We began our analysis by numerically integrating eq. (1) and eq. (2) by considering the presence of diffusion driven instability. Our intention here is to unravel how the applied circularly polarized external electric field is going to affect the spatiotemporal patterns observed earlier by Pearson under different parametric domain of Gray-Scott model.

#### **Transformation of stationary patterns into time dependent spatial structures in presence of diffusion driven instability**

For the parameter value  $F = 0.038, K = 0.0585$ , along with the trivial steady state, there exists another steady state  $u_{ss} = 0.429, v_{ss} = 0.224$ , which is stable in absence of diffusion. Now, if the diffusivity of  $U$  is higher compared to the diffusivity of  $V$  ( $D_u = 2 \times 10^{-5}, D_v = 10^{-5}$ ), the stable steady state becomes unstable in presence of diffusion and ultimately leads to spot pattern (Fig.1(i)). Upon application of a circularly polarized electric field  $E_x^0 = E_y^0 = 10.0$ , this spot pattern gets transformed into complex labyrinth type spatial structure (Fig. 1(ii)-(iv)). For the numerical simulation we took  $\omega = 0.0015$ . Since the applied electric field is circularly polarized, the whole spatial structure seems to evolve with time (Vid1.mov, Supplementary file).

We get the similar observation (SFig. S1, Supplementary file) for another parametric space ( $F = 0.03, K = 0.05$ ). Here also the spot pattern gets transformed into complex labyrinth type structure, because of the interplay between diffusion driven instability and circularly polarized electric field induced instability.

Interestingly, in the regime, where only the trivial steady

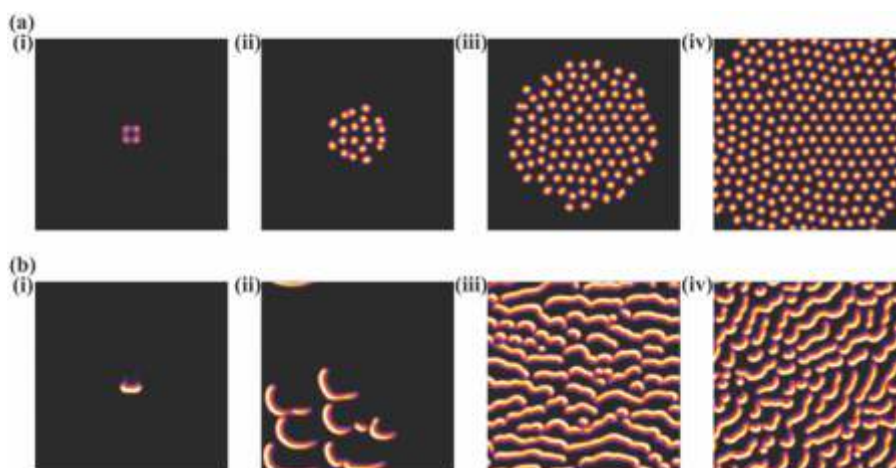


**Fig. 1.** Transformation of the spatiotemporal structure (in presence of diffusion driven instability) in presence of a circularly polarized electric field by perturbing the “blue state”. (i) The reaction-diffusion equations are simulated for 50000 time steps in absence of electric field. (ii)-(iv) Results are obtained in presence of electric field at different time points: (ii) 10000 time steps, (iii) 30000 time steps and (iv) 50000 time steps.

state exists for the parameter values of  $F = 0.035$ ,  $K = 0.065$ , one usually obtains a multiplicative spot pattern (Vid2a.mov, Supplementary file) that evolves slowly with time to produce a spatiotemporal structure as shown in Fig. 2a(i)-(iv). For numerical integration we use the same procedure as used by Pearson. When a bidirectional circularly polarized external electric field having amplitude of  $E_x^0 = E_y^0 = 25.0$ , ( $\omega = 0.0015$ ) is applied to the same system, the spots started colliding with each other Fig. 2b(i)-(iii) and gave rise to a mixture of spots and stripes (Vid2b.mov, Supplementary file), and eventually produced a mixed spot-stripe kind of spatiotemporally evolving structure Fig. 2b(iv). The time dependent nature of the external forcing made the spatial structures also time dependent (as shown in Fig. 2b).

### Circularly polarized electric field induces spiral pattern formation

The previous section demonstrates that circularly polarized external electric field can influence the spatial patterns already observed due to diffusion driven instability. Intriguingly, we explored another set of parameter values ( $F = 0.005$ ,  $K = 0.025$ ), where even in presence of difference in diffusivities between  $U$  and  $V$  ( $D_U = 2 \times 10^{-5}$ ,  $D_V = 10^{-5}$ ), the numerical simulation produces a homogeneous spatial solution, i.e. no spatiotemporal pattern is obtained. This implies that diffusion driven instability is not enough to create an inhomogeneous spatial solution in this instance. To make the system unstable enough, we apply a circularly polarized electric field having amplitude  $E_x^0 = E_y^0 = 5.0$  and  $\omega = 0.0015$ . Upon ap-

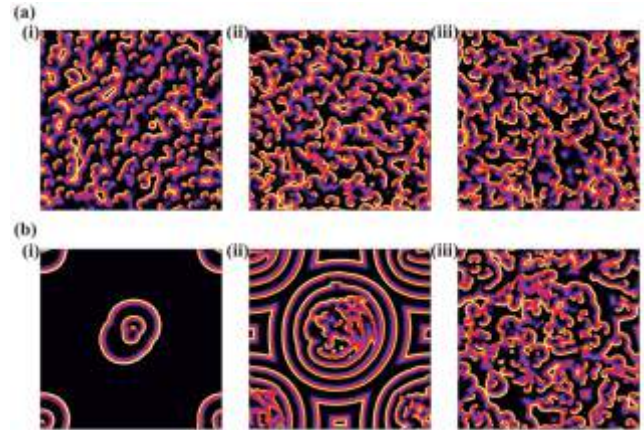


**Fig. 2.** Transformation of the spatiotemporal structure (in presence of diffusion driven instability) in presence of a circularly polarized electric field by perturbing the trivial steady state. (a) Evolution of spot pattern only because of diffusion driven instability for the parameter value  $F = 0.035$ ,  $K = 0.065$ . (b) Effect of electric field on this parameter value. The reaction diffusion equations are simulated for 50000 time steps for both the cases and at different time points, it is shown in the figure as follow: (i)  $t = 0$  time step, (ii)  $t = 10000$  time steps, (iii)  $t = 30000$  time steps and (iv)  $t = 50000$  time steps.

plication of such a circularly polarized external electric field, the system evolves toward a heterogeneous spatial regime (Fig. 3). Here we obtain an immature spiral pattern, where the spiral produced transiently collides with each other and annihilates each other. As mentioned in the previous section, here too the stationary spatial structures will not form due to the time dependent nature of the external forcing.

We further investigate whether the initial perturbation employed to initiate the numerical integration has any role in determining the finally obtained spatiotemporal structures or not? For that we give initial perturbation to the system in two different ways while initiating the numerical simulation. First, we initially perturb the whole grid with  $\pm 1\%$  random noise around the steady state of this parameter value ( $u_{ss} = 0.235$ ,  $v_{ss} = 0.127$ ). To look at the evolution of the perturbation in a better way, we take a larger grid ( $1024 \times 1024$ ) size. In this scenario, we have observed the evolution of immature spirals on the whole grid (shown in Fig. 3a). Second, we have employed almost similar procedure as mentioned by Pearson for simulating the trivial steady state. Since in this parameter value ( $F = 0.005$ ,  $K = 0.025$ ) along with the trivial one ( $u_{ss} = 1.0$ ,  $v_{ss} = 0.0$ ), another steady state also exists, we initially place the whole box on the steady state ( $u_{ss} = 0.235$ ,  $v_{ss} = 0.127$ ) except the center of the box ( $80 \times 80$ ). In the middle of the box we initially perturb the steady state with  $\pm 1\%$  random noise. We found that the evolution of the initial perturbation is different compared to the first case discussed earlier. Here the heterogeneous solution emerges from the four corners along with the center of the box.

From the center of the box and from the corners, ring like structures are appearing (Fig. 3b(i)). Eventually, with time spiral structures are emerging from the ring like structures (Fig. 3b(ii)). Fascinatingly, the rings coming from the center collide with the rings evolving from the corners, and the whole grid becomes full of immature spirals, where the spiral structures collide with its neighboring spirals (Fig. 3b(iii)). This clearly depicts that the initial perturbation is not dictating the ultimate spatial structures. Initial perturbation only can change the initial evolution of the heterogeneous solutions, but under long time limit the heterogeneous spatial structures become independent of the initial perturbation. To show the spatio-temporal evolution of the patterns as discussed in Fig. 3, we have provided additional video (Supplementary file,



**Fig. 3.** Circularly polarized electric field induced spatial spiral patterns (difference in diffusivities is present) and the effect of initial perturbation on the emergent spiral patterns. The patterns shown in (i)-(iii) for both (a) and (b) are at (i)  $t = 5000$  time steps, (ii)  $t = 15000$  time steps and (iii)  $t = 25000$  time steps. In (a) and (b), the initial perturbation employed to initiate the numerical simulation is different, but other conditions remain same.

Vid3a.mov and Vid3b.mov) files produced from the simulations to make the point clearer.

### ***Circularly polarized electric field induced spatial pattern formation in absence of diffusion driven instability***

In previous sections, we have shown interplay between diffusion-driven instability and circularly polarized electric field induced instability can give birth to wide variety of time dependent spatial structures. In literature, it has been recently shown that stationary bi-directional static electric field can induce instability even when the diffusivity ratio of the reactants is one ( $D_u = D_v = 2 \times 10^{-5}$ ), and stable stripe pattern can be obtained in various parametric space for Gray-Scott model system<sup>32</sup>. The stripe patterns were obtained because of the uni-directionality of the external forcing. Can we get wider variety of spatial structures in absence of diffusion driven instability just by applying external forcing? To answer this question, we employ the circularly polarized electric field on the spatially extended Gray-Scott model, by assuming that both the reactants have similar diffusivities ( $D_u = D_v = 2 \times 10^{-5}$ ). We have performed numerical simulation for the parameter value  $F = 0.038$ ,  $K = 0.0585$  for 50000 time steps in presence of circularly polarized electric field ( $E_x^0 = E_y^0 = 10.0$ )

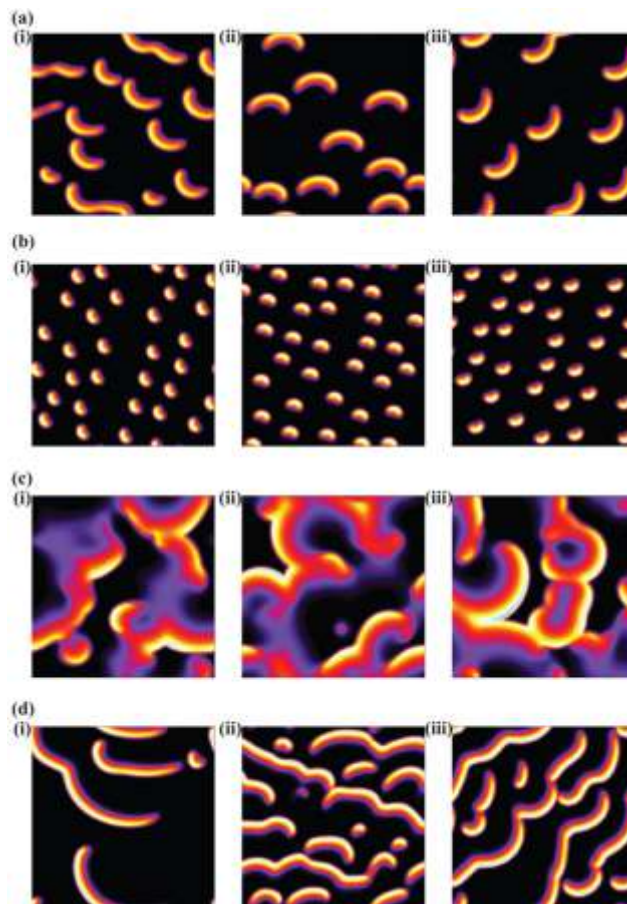
on a spatial grid of size  $256 \times 256$ . Initially,  $\pm 1\%$  random noise is being introduced to the whole system around the steady state ( $u_{ss} = 0.429$ ,  $v_{ss} = 0.224$ ). Results of the numerical simulation show that at the very beginning stripe like structure evolves, but later on, due to the periodic nature of the external forcing the stripe patterns break into a unique kind of spatial pattern ('half-moon' or 'elongated spot' shaped structures). Since the temporal dynamics of external electric field varies periodically in both  $x$  and  $y$  direction, the whole pattern rotates and during the rotation these elongated spots are coming close to each other and colliding with each other (Vid4a.mov, Supplementary file). In Fig. 4a(i)-(iii) the above mentioned patterns are shown at three different time points.

In a similar manner, upon application of a bi-directional circularly polarized external electric field ( $E_x^0 = E_y^0 = 25.0$ ,  $\omega = 0.0015$ ) on another parameter space ( $F = 0.063$ ,  $K = 0.06249$ ), where 'blue state' exists along with the trivial one, a heterogeneous spatiotemporal solution emerges (Fig. 4b(i)-(iii)) in absence of diffusion driven instability (here  $D_u = D_v = 2 \times 10^{-5}$ ).

Here also at the very beginning only stripe patterns evolve, but with time these stripes break into spots. Due to the circularly polarized nature of the electric field the spots rotate too (Vid4b.mov, Supplementary file). Fig. 4b depicts how with time the spots are changing their positions continuously.

At this point, we further examine what happens when we excite the Gray-Scott model system at the parametric ( $F = 0.005$ ,  $K = 0.025$ ) regime, where we got spiral kind of spatiotemporal pattern earlier (Fig. 3). Here we added the extra constraint that there is no difference in the diffusivities of the reactant species involved. We observed that even when the activator and substrate have the same diffusivities ( $D_u = D_v = 2 \times 10^{-5}$ ), we can get spiral pattern (Fig. 4(b)). For this we have to increase the amplitude of the circularly polarized electric fields ( $E_x^0 = E_y^0 = 10.0$ ) than the previous case (Fig. 3). Fig. 4(b) evidently shows that only applying circularly polarized external electric field ( $E_x^0 = E_y^0 = 10.0$ ,  $\omega = 0.0015$ ) can induce instability into the system and lead to spiral spatial structures in complete absence of diffusion driven instability.

Like constant bi-directional electric field, circularly polarized electric field can also induce instability in a Gray-Scott like system, where the sole steady state is the trivial one. In the parameter space  $F = 0.03$ ,  $K = 0.06$ , the sole steady is  $u_{ss} = 1.0$ ,  $v_{ss} = 0.0$ . For numerical simulation on this regime,



**Fig. 4.** Circularly polarized electric field induced pattern formation (in absence of diffusion driven instability) in Gray-Scott model even in absence of diffusion driven instability ( $D_u = D_v = 2 \times 10^{-5}$ ) at different values of  $F$  and  $K$ . (a) Patterns formed in presence of the circularly polarized electric field ( $E_x^0 = E_y^0 = 10.0$ ,  $\omega = 0.0015$ ) for the parameter value  $F = 0.038$ ,  $K = 0.0585$ . (b) Spot pattern formation for the parameter value  $F = 0.063$ ,  $K = 0.06249$  in presence of bi-directional ac electric field. The amplitude of ac electric field is  $E_x^0 = E_y^0 = 10.0$  and frequency,  $\omega = 0.0015$ . (c) Spiral pattern formed for the parameter value  $F = 0.005$ ,  $K = 0.025$ , where the strength of the time dependent electric field is  $E_x^0 = E_y^0 = 10.0$  and the frequency,  $\omega = 0.0015$ . (d) Patterns formed in the parameter space  $F = 0.03$ ,  $K = 0.06$  under the influence of a circularly polarized electric field ( $E_x^0 = E_y^0 = 25.0$ ,  $\omega = 0.0015$ ). For all the parameter sets described here the reaction-diffusion equations are simulated for 50000 time steps. Patterns shown in (i)-(iii) for (a)-(d) are at different time points: (i)  $t = 10000$  time steps, (ii)  $t = 30000$  time steps and (iii)  $t = 50000$  time steps.

we again employ Pearson's strategy described earlier. Results of the numerical simulations show the emergence of curved stripe type pattern at the beginning, and the patterns

start rotating with time. During rotation, one stripe collide with its neighbor and gives birth to a mixture of spots and stripe (Fig. 4d(i)-(iii)). The observations made in this section suggest that even with equal diffusivity of the reactants, circularly polarized electric field can give birth to wide variety of spatial structures. This is in stark contrast to the scenario of the spatial structures obtained under constant bi-directional electric field, which only produces stripe patterns<sup>11,32</sup>. The reason of showing wide variety of pattern in this situation lies on the periodic nature of applied electric field and the phase difference of the two circularly polarized electric field along  $x$  and  $y$  directions, respectively. Due to the phase difference, with time the amplitude of external forcing in both directions get modulated in different extent. Thus, the stripe like patterns usually obtained for constant bi-directional electric field give rise to other kinds of patterns under periodic variation of the applied circularly polarized fields.

## Conclusion

In early 1950's, Turing proposed an ingenious idea that in a reaction-diffusion system, the difference in diffusivities of the two different kind of reactant species can create an instability that in turn gives rise to spatiotemporal patterning in chemical and biological systems<sup>3</sup>. Experimentally, it is still a challenge to generate a Turing like system due to such stringent requirement of differences in diffusion coefficient of reactant species. In literature, it had been demonstrated that the diffusion driven instability can be supplemented by applying external electric field in reaction-diffusion systems involving ions. Application of constant electric field along one direction (or in both directions) drives the ions to align along a particular way, which always produces the stripe like pattern even in absence of diffusion driven instability. This has been well explored in literature from both theoretical and experimental standpoints<sup>11,27,28,32</sup>. Unfortunately, the spatial patterns formed under such cases are only limited to stripe like patterns. How to manufacture different types of spatial patterns by applying external forcing especially in absence of diffusion driven instability?

In this article, we addressed this question and showed that by employing circularly polarized external electric field in a Gray-Scott like reaction-diffusion system, one can obtain dynamically highly diverse spatiotemporal patterns. Importantly, we demonstrated that by applying a circularly po-

larized electric field, a wide variety of spatial structures can be obtained even when the two reactants have equal diffusivities (Fig. 4). Interestingly, the spatiotemporal structures attained after long time limit were still evolving with time (Vid4a-d.mov, Supplementary files) due to the time dependent nature of the applied circularly polarized electric fields along two perpendicular directions. The point to be noted here is that the spatiotemporal instability in this instance is mainly caused by the disparity in the phase difference of the circularly polarized electric fields along two mutually perpendicular directions. Not only that, this phase difference gets also reflected in the nature of revolutions (Vid4a-d.mov, Supplementary files) we observed in the long time limit spatiotemporal patterns as well. Due to this, the patterns formed are not getting aligned into a specified direction. As a results, the stripe patterns at the beginning break down into other kinds of mixed spatial structures with progression of time.

Additionally, we showed that by applying circularly polarized electric field, one can transform the stationary structures into time dependent one, even in presence of diffusion driven instability (Figs. 1–3). The applied circularly polarized electric field seemed to transform one type of spot pattern (originated from 'blue state') into complex labyrinth type of spatial structure (Fig. 1), while other type of spot pattern (originated from 'red state') is getting transformed into a mixture of spot and stripe (Fig. 2). This variety in the spatial structures obtained can be attributed to the diverse dynamical nature of the Gray-Scott model system, which shows various dynamical features under a range of parametric conditions of  $F$  and  $K$ <sup>33,38,39</sup>. Due to such dynamical diversity of the Gray-Scott system, for one set of parameter value the spot patterns get transformed into labyrinth, and for another set it is getting converted into mixture of spots and stripe. Our numerical simulations further revealed that where diffusion is not able to create enough instability to observe pattern formation, introduction of circularly polarized electric field (with relatively low amplitude) is enough to create spatially heterogeneous solution. In this context, we showed that the initial perturbation (employed to initiate the simulation) only can change the transient evolution of spatial heterogeneous solution seen at the beginning, but ultimately the initial perturbation will not have any role on the final form of the spatiotemporal patterns (Fig. 3).

In conclusion, our simulation studies unravel that circularly polarized electric field can produce a wide variety of spatial structures both in presence and absence of diffusion driven instability in a spatially extended Gray-Scott like system constituted with ionic reactants. Since, there are more than one kind of experimental systems available, which follow the Gray-Scott model system, one can verify our predictions experimentally. We strongly believe that application of such kind of circularly polarized electric field can provide a unique way out to circumvent the stringent Turing conditions, and at the same time will allow to realize complex spatiotemporal patterns emerging from ionic reaction-diffusion systems.

### Acknowledgements

Thanks are due to UGC for providing the UGC-CSIR-JRF fellowship (Ref. No: 19-06/2016(i)EU-V) to AG.

### Supplementary material

Supplementary video files for this work are available in the following link - <https://drive.google.com/open?id=1HG3KHtQx7KG6eF5wRyAZdSbsCRYC0BYt>.

### References

- J. D. Murray, "Mathematical Biology", Springer, Berlin, 1993.
- J. D. Murray, *Interface Focus*, 2012, **2**, 397.
- A. M. Turing, *Philos. Trans. R. Soc. London, Ser. B*, 1952, **237**, 37.
- P. K. Maini, K. J. Painter and H. N. P. Chau, *J. Chem. Soc., Faraday Trans.*, 1997, **93**, 3601.
- P. K. Maini, *Comptes Rendus - Biol.*, 2004, **327**, 225.
- I. Lengyel and I. R. Epstein, *Science*, 1991, **251**, 650.
- L. Yang, A. M. Zhabotinsky and I. R. Epstein, *Phys. Rev. Lett.*, 2004, **92**, 1.
- I. Berenstein, A. P. Muñozuri, L. Yang, M. Dolnik, A. M. Zhabotinsky and I. R. Epstein, *Phys. Rev. E - Stat. Nonlinear, Soft Matter Phys.*, 2008, **78**, 15.
- S. Dutta and D. S. Ray, *Phys. Rev. E - Stat. Nonlinear, Soft Matter Phys.*, 2007, **75**, 016205.
- S. S. Riaz, S. Banarjee, S. Kar and D. S. Ray, *Eur. Phys. J.*, 2006, **53B**, 509.
- S. S. Riaz, S. Kar and D. S. Ray, *J. Chem. Phys.*, 2004, **121**, 5395.
- W. Q. Chen, H. Zhang, H. P. Ying, B. W. Li and J. X. Chen, *J. Chem. Phys.*, 2007, **127**, 154708.
- B. W. Li, M. C. Cai, H. Zhang, A. V. Panfilov and H. Dierckx, *J. Chem. Phys.*, 2014, **140**, 184901.
- I. Berenstein, L. Yang, M. Dolnik, A. M. Zhabotinsky and I. R. Epstein, *J. Phys. Chem. A*, 2005, **109**, 5382.
- R. Nagao, I. R. Epstein and M. Dolnik, *J. Phys. Chem. A*, 2013, **117**, 9120.
- V. K. Vanag and I. R. Epstein, *Phys. Rev. E - Stat. Nonlinear, Soft Matter Phys.*, 2010, **81**, 1.
- V. Horvath, P. L. Gentili, V. K. Vanag and I. R. Epstein, *Angew. Chem. Int. Ed.*, 2012, **51**, 6878.
- S. Ghosh, S. Paul and D. S. Ray, *Phys. Rev. E*, 2016, **94**, 042223.
- R. Zhang, L. Yang, A. M. Zhabotinsky and I. R. Epstein, *Phys. Rev. E - Stat. Nonlinear, Soft Matter Phys.*, 2007, **76**, 1.
- X. J. Lu, L. Ren, Q. Y. Gao, Y. Y. Yang, Y. M. Zhao, J. Huang, X. L. Lv and I. R. Epstein, *J. Phys. Chem. Lett.*, 2013, **4**, 3891.
- L. Stucchi and D. A. Vasquez, *Phys. Rev. E - Stat. Nonlinear, Soft Matter Phys.*, 2013, **87**, 1.
- H. Sevcikova, M. Marek and S. C. Muller, *Science*, 1992, **257**, 951.
- D. Snita, H. Sevcikova and M. Marek, *J. Phys. Chem.*, 1996, **100**, 18740.
- S. L. Schmidt, *J. Chem. Phys.*, 1983, **79**, 5939.
- Á. Tóth and D. Horváth, *Chaos*, 2015, **25**, 064304.
- Z. Virányi, Á. Tóth and D. Horváth, *Phys. Rev. Lett.*, 2008, **100**, 2.
- T. Lin, S. M. Rubinstein, A. Korchev and D. A. Weitz, *Langmuir*, 2014, **30**, 12119.
- B. Schmidt, P. De Kepper and S. C. Müller, *Phys. Rev. Lett.*, 2003, **90**, 118302.
- M. Watzl and A. F. Münster, *J. Phys. Chem. A*, 1998, **102**, 2540.
- J. X. Chen, H. Zhang and Y. Q. Li, *J. Chem. Phys.*, 2009, **130**, 1.
- Z. Virányi, A. Szommer, Á. Tóth and D. Horváth, *Phys. Chem. Chem. Phys.*, 2004, **6**, 3396.
- A. Giri and S. Kar, *J. Chem. Phys.*, 2019, **150**, 094904.
- P. Gray and S. K. Scott, *Chem. Eng. Sci.*, 1983, **39**, 1087.
- J. E. Pearson, *Science*, 1993, **261**, 189.
- K. J. Lee, W. D. McCormick, J. E. Pearson and H. L. Swinney, *Nature*, 1994, **369**, 215.
- W. Mazin, K. E. Rasmussen, E. Mosekilde, P. Borckmans and G. Dewel, *Math. Comput. Simul.*, 1996, **40**, 371.
- V. Horváth, I. R. Epstein and K. Kustin, *J. Phys. Chem. A*, 2016, **120**, 1951.
- S. K. Scott, *Chem. Eng. Sci.*, 1987, **42**, 307.
- E. E. Sel'kov, *Eur. J. Biochem.*, 1968, **4**, 79.