

Chaotic behaviour induced by modulated illumination in the Lengyel-Epstein model under Turing considerations

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Abstract: The photosensitive CDIMA reaction was investigated using the Lengyel Epstein model modified to include the effect of external illumination. Different spatial patterns are exhibited under constant values of light, ranging from Turing Spots to Stripes for the minimum and maximum values of illumination, respectively. Moreover, by neglecting the diffusion, the system displays oscillations with a characteristic period that also depends on the illumination value. When illumination is set to periodically oscillate three different behaviors are observed. Namely, a regime exhibiting the period of the external forcing; another where there is a resonance between several periods of oscillations and a broad regime where the system demonstrates a chaotic-like behavior.

Keywords: Chaotic modeling, Lengyel-Epstein model, CDIMA photosensitive reaction, reaction-diffusion system.

1 Introduction

The reaction between chlorine dioxide, iodine and malonic acid (CDIMA reaction) is one of the most thoroughly studied oscillatory chemical systems [1, 2] both experimentally and numerically. This reaction constitutes a good prototype for studying complex dynamics, such as the symmetry-breaking, reaction diffusion Turing patterns [3]. Moreover, experiments performed by Epstein Group reported that CDIMA reaction presents a high sensitivity to visible light [4]. The photosensitivity opens the possibility to control the different patterns by using either temporal illumination (constant or periodical), spatial or spatiotemporal forcing [5, 6]. Specifically, the light forcing is able to induce a transition between patterns [7], suppress the structures [8] or introduce new localized patterns [9].

2 The Model and Simulations

We employed the Lengyel-Epstein model [10, 11] because it approaches to the true kinetics of the experiments and allows analytical calculations in good agreement, both quantitative and qualitative, with the experiments. This model consists of two coupled reaction-diffusion equations, once modified to take into account the illumination, as:



$$\frac{\partial u}{\partial t} = a - cu - 4 \frac{uv}{1+u^2} - \phi(t) + \nabla^2 u \quad (1)$$

$$\frac{\partial v}{\partial t} = \sigma \left[cu - \frac{uv}{1+u^2} + \phi(t) \right] + d \nabla^2 v \quad (2),$$

Here u and v are the dimensionless concentration for iodide (activator) and ClO_2^- ions (inhibitor), respectively; a , c and σ are dimensionless parameters of the chemical system; d is proportional to the ratio of the diffusion coefficients of the main species ($d = D_{\text{inhibitor}}/D_{\text{activator}}$). The parameter Φ plays the role of the illumination intensity. In this work the light sinusoidally varies with time according to:

$$\phi(t) = \frac{\phi_{\max} + \phi_{\min}}{2} + \frac{\phi_{\min} - \phi_{\max}}{2} \sin(\omega t) \quad (3)$$

The relevance of the above light forcing lies in the always positive value of Φ which can be tuned through a characteristic period of forcing ($\omega = 2\pi/T$). Whether we only considered the temporal evolution and without any spatial consideration, *i.e.* a OD-system, the model equations (1)-(2) are solved numerically by the Runge-Kutta method with a time step 0.001 time units (t. u.). In presence of diffusion the simulations were performed by a Dufort-Frankel model in addition to Dirichlet and Newman conditions:

$$u(\bar{r}, t)|_{t=t_0} = u_{ini} ; \hat{n} \nabla u|_{\partial\Omega} = 0 \quad (4)$$

$$v(\bar{r}, t)|_{t=t_0} = v_{ini} ; \hat{n} \nabla v|_{\partial\Omega} = 0 \quad (5)$$

In our simulations, for initial conditions we chose small perturbation (5%) of random values close to steady state:

$$u_0 = \frac{(a - 5\phi)}{c} \quad (6)$$

$$v_0 = \frac{(u_0 c + \phi)}{u_0} (1 + u_0^2) \quad (7)$$

The steady state go through a Hopf instability if

$$c_H < \sqrt{\frac{(3a/5 + \phi - \sigma)(a - 5\phi)^2}{25(a - \phi)}} \quad (8)$$

evolving into a homogeneous limit cycle characterized by a typical frequency.

The homogeneous steady state of the system may also undergo Turing instability when:

$$c_T < \frac{\beta - \sqrt{\beta^2 - 4\alpha\beta}}{2\alpha} \tag{9}$$

where

$$\alpha = 5^6 (d(a - \phi))^2, \tag{10}$$

$$\beta = 25(10d(a - \phi)(a - 5\phi)^2(d(3a - 5\phi) - 5)) + 2500d(a - 5\phi)^3 \tag{11}$$

$$\text{and } \gamma = (a - 5\phi)^4 (d(3a - 5\phi) - 5)^2 - 100d(a - 5\phi)^5 \tag{12}$$

The point where these two different instabilities coincide is the so-called codimension-two Turing-Hopf point (CTHP). By plotting in a parameter space the two parameters used to determine the different instabilities (C and Φ), we obtained that our range of study is located in a Subcritical Turing domain (see Figure 1). The relevance of such regime reside in the oscillations displayed when we study the system without spatial diffusion and the Turing patterns observed taking into account the diffusion.

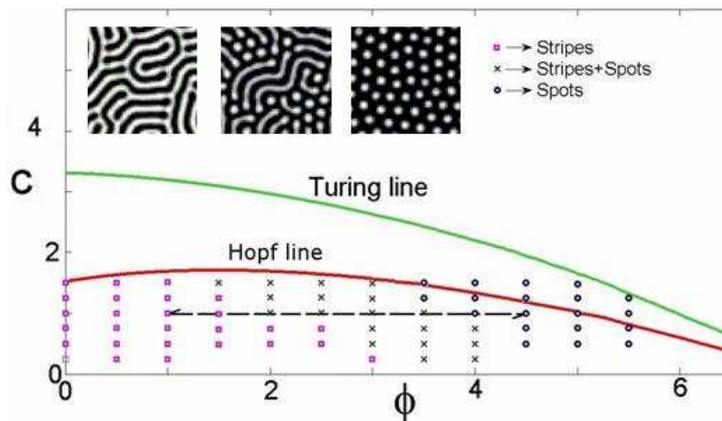


Fig. 1. C vs Φ phase portrait in a model of the CDIMA reaction-diffusion system with constant illumination. Fixed parameters in our simulations: $a= 36$, $c=1$, $\sigma=20$ and $d=1.027$. The dashed line corresponds to the range of parameters studied once that we introduce the modulated light forcing. Different stationary Turing patterns where obtained in our numerical simulations, \square Stripes, x mixture of stripes and spots and \circ spots by at constant values of the illumination.

It is important to note the different Turing patterns exhibited by Lengyel-Epstein model for the different values of the light as we shown in Figure 1. Thus, as we increase the illumination parameter control (Φ), the system evolves from a pure Stripe configuration to a pure hexagonal Spots regime going trough a mixture of both of them (see insets in Figure 1). We want to recall that each of these patterns was obtained for a constant value of illumination. The purpose of this

work is the analysis of the dynamics obtained when we modulated the light in a sinusoidal way in between the Stripe and Spots configuration.

By using a linear stability analysis of the model (1) - (2), we obtain the related dispersion curves as a function of the wavenumber (see Figure 2). We observed that although both instabilities (Turing and Hopf) coexist in our range of parameters, the predominant mode differs according to the value of Φ . For lower values of the illumination, the dispersion relationship presents a predominant Turing mode slightly influenced by Hopf. However, the maximum value of illumination, Φ_{\max} , demonstrates a clear resonance between the Turing and Hopf instability, where the last one became predominant. Increasing the parameter of control Φ in the relation dispersion makes the Turing regime to expand and shifts the most probable to higher values.

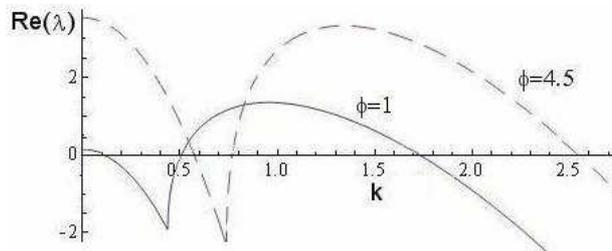


Fig. 2. Schematic dispersion relations displaying the interaction between the Turing and Hopf instabilities. The dispersion curves were analyzed for two different values of the illumination: Dash line (Φ_{\max}), solid line (Φ_{\min}).

We focused our study analysis in the two-variable model (1) – (2) in the absence of diffusion, i.e. we analyzed the temporal evolution of the 0D system. Thereby, the Lengyel-Epstein with a constant illumination presents an oscillatory solution with a characteristic period (figure 3a). By changing the illumination parameter in between the minimum (Φ_{\min}) and maximum (Φ_{\max}) values, the period of oscillation increases in the same way that Φ does (Figure 3b).

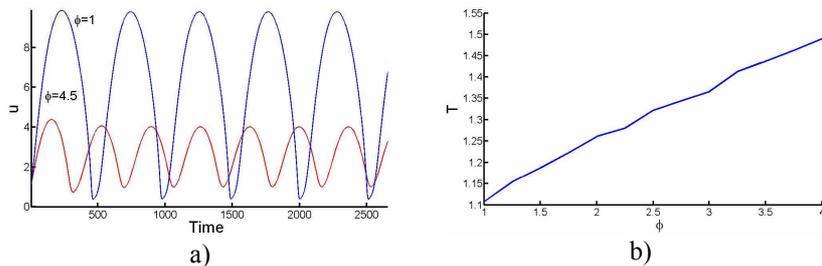


Fig. 3. Lengyel-Epstein model in presence of constant illumination. a) Oscillations profile for two forcing values (blue line corresponds to $\Phi_{\min}=1$. Red line corresponds to $\Phi_{\max}=4.5$). b) Dependence of the oscillation period with the illumination parameter

The modulation periodic of the external light, introduces a new parameter, the period of the forcing, and depending on its value the dynamics of the system show different responses. The amplitude of the oscillatory behavior, for both the activator and inhibitor dimensionless species, was considered as the key parameter in order to analyze the results. Moreover, we want to note that all the simulations were carried out for a narrow range of the period of the forcing ($1 < T < 1.06$). This fact enhances the susceptibility of the Lengyel-Epstein model to the illumination parameter. By plotting the amplitude of the activator for all the different values of the forcing, we differentiated three different regimes as we show in figure 4

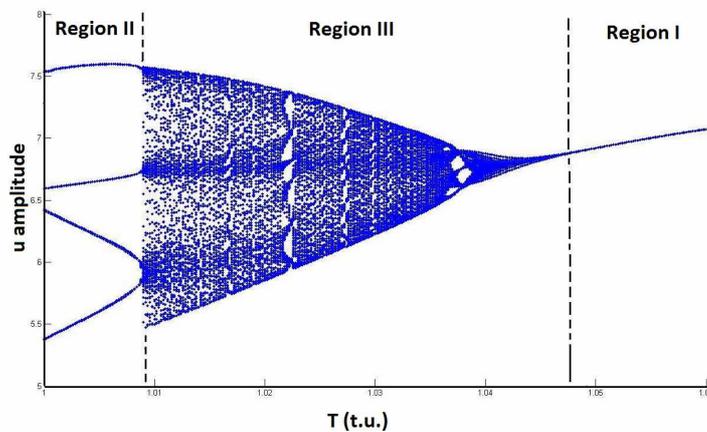


Fig. 4. Bifurcation diagram showing the values of activator's amplitude as the response of the period of illumination.

Region I. In the range of period of forcing $1.05 < T < 1.06$, the system demonstrates an oscillatory dynamic. The peculiarity of such sinusoidal behavior lies in the fact that the LE model exhibits a period of oscillation equal to the period of the forcing. All the oscillations of the system are performed with the same amplitude (In the example displayed in figure 5, the amplitude given by the limit cycle of figure 5.a). Fast Fourier transform was used to verify the presence of a unique period of oscillation (Figure 5b).

Region II. By forcing the system with a period of illumination within this regime, the limit cycle described by the system is splitted into three amplitudes of oscillation, as can be seen in the example of figure 5c. Moreover, the LE model does not present oscillations with the frequency of the forcing, but rather, it exhibits periods close to this value (see Figure 5d).

Region III. For a broad range of forcing periods, the system oscillates showing a chaotic behavior in the oscillation amplitudes, as we show in the path traced in

the phase space (see Figure 5e). The analysis of the period of oscillations followed by FFT also corroborates such results (Figure 5f).

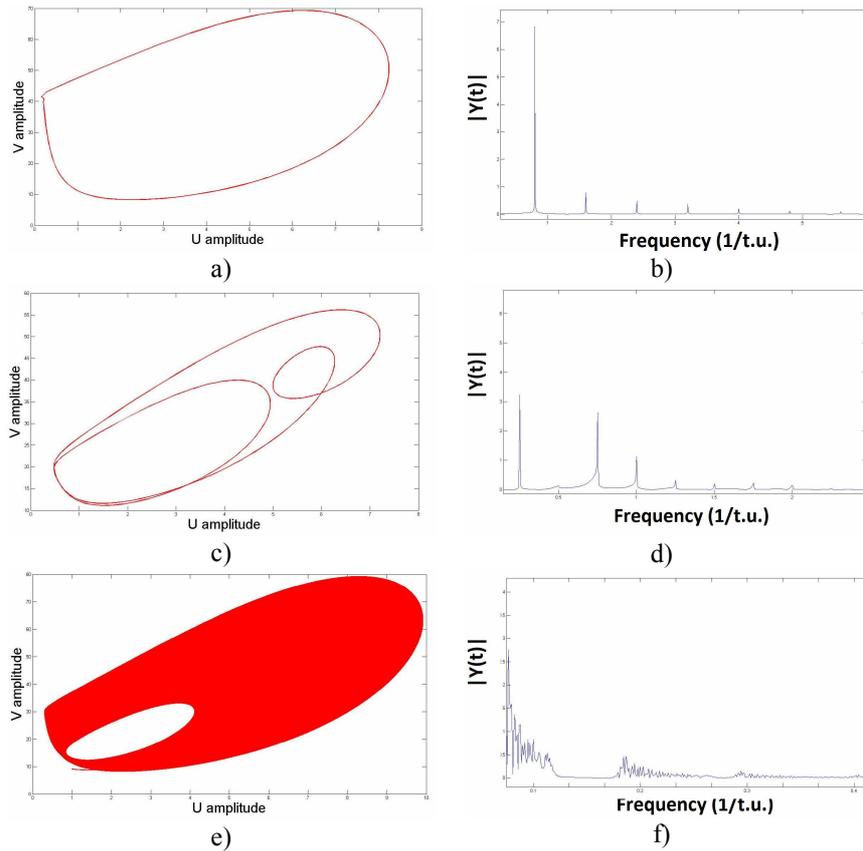


Figure 5. System responses for different frequencies of forcing. Left panels: Phase space exhibiting the associated limit cycles. Right panels: Fast Fourier Transform of the response signal showing the most significant peaks. Frequency of forcing: 1.05 (cases a, b), 1.009 (cases c, d) and 1.035 1/t.u. (cases e, f)

3 Conclusions

The Lengyel-Epstein model was modified to include the photosensitivity as an external forcing. Our work deals with a time-periodic (always positive and non-zero) illumination restricted between a maximum and minimum that displays a sort of subcritical Turing instability. Moreover, the wealth of the system allow us to observe oscillations (which depend linearly on the forcing), when we analyze the system without spatial considerations, and also Turing patterns

(going from Stripes to Spots as we increase the illumination parameter), whether the diffusion takes place.

Although our analysis concerns to a narrow range of forcing periods, we obtain three regimes with different dynamics. For higher periods, the system oscillates with the period induced by the forcing. However, for higher values of the illumination period, the system splits into different periods. Under intermediate periods of oscillations, the Lengyel-Epstein presents a broad range of parameters with a chaotic-like behavior.

These results enhance the high sensitivity of LE model under applied forcing and also open the possibility to perform a more carefully simulations under a broad range of illumination frequencies, where these kind of resonant dynamics are expected. Furthermore, these results suggest that applied waveform forcing can induce exciting spatiotemporal complex patterns once we take into account the spatial diffusion.

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