Harmonic vibration on a reactive fluid at boundary layer regime modifies the Turing instability

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Abstract. Studies of Turing instability in a reactive fluid, known as BZ-AOT microemulsion, under external harmonic vibration, show modifications on pattern formation. The characteristics of the reactor and forcing prevent flows to occur, and therefore the transport phenomena can only be due to microscopic diffusion. In order to understand this problem, we combine the reaction-diffusion model with a mesoscopic model. The performed linear stability analysis and numerical simulations show, on the one hand, that the forcing increases the wavelength of the patterns and, on the other hand, that the system is unable to exhibit any pattern for sufficiently large forcings thus yielding to a state of homogeneous concentration.

1. Introduction

Extensive studies have been performed in order to understand the formation mechanisms of the elegant patterns that are ubiquitous in Nature [1, 2], i.e. striped tiger skin or the sea-shells forms. Turing instability [3] represents a good mechanism to reproduce the pattern formation from a physicochemical perspective. Turing structures are stationary patterns that may appear when the propagating chemical waves are combined with the effects of diffusion in a medium. In this paper, we will consider the numerical and theoretical aspects of Turing instability obtained in a compartmented reaction-diffusion system (BZ-AOT system), by keeping in mind the location of the problem within the experimental context where these results can be applied.

The Belousov-Zhabotinsky reaction [4, 5] can be confined inside a water-in-oil microemulsion using AOT (sodium bis(2-ethylhexyl) sulfosuccinate) as the stabilizing amphiphile (BZ-AOT system). This closed system demonstrates a rich variety of dynamics, which are impossible to obtain by the classical BZ reaction [6, 7], such as Turing patterns. The BZ-AOT reaction involves a lot of elementary reactions [8]; however, the system dynamics is related to two main characteristics; on one hand, the antagonistic and competitive behaviour between some species of the reaction (known as activator and inhibitor), and on the other hand, their different diffusion rates. This difference in the diffusion coefficients lies in the different location of these antagonistic species. While the activator resides within the AOT microemulsion and diffuses as a micelle, the inhibitor is able to diffuse through the oil phase due to its nonpolar nature.

Nevertheless, in Nature, pattern formation usually occurs in presence of external perturbations [9-13]; thus, it is necessary to develop a more general description, in order to understand the transport phenomena and their impact in pattern formation mechanisms. In this work, we develop a numerical study in which the dynamics of pattern formation will be modified by introducing a harmonic temporal perturbation vertically to a two-dimensional BZ-AOT system [14, 15], so that, convection is
not allowed due to physical constrains. This paper is organized as follows: in Sec. 2 we present the reaction-diffusion model, as well as a discussion establishing the kind of transport mechanism within our system. Sec. 3 deals with the mesoscopic treatment of the harmonic perturbation and how it affects the diffusion processes. The characteristics of Turing pattern formation are described in Sec. 4, by a linear stability analysis of the reaction-diffusion equations. In Sec. 5 we present numerical simulations of the pattern formation showing how the harmonic perturbation modifies the characteristics of the Turing structures. Finally, in Sec. 6 we present the conclusions and discussion.

2. Model equations

The problem we want to address is schematically depicted in Fig. 1. We consider a liquid pattern-forming system (BZ-AOT system) located within a closed reactor with a small vertical dimension (marked with L in figure). The whole system is bounded by glass boundaries that actually play a crucial role in determining the fluid dynamics as we will discuss below. The system described is periodically forced (up to 2.4 cm in amplitude and 100 Hz in frequency) in the vertical direction thus, the actual gravity affecting the reaction is periodically modulated with time.

The phenomenology of pattern formation including Turing instability can be studied through the Reynolds theorem. Turing structures are due to singular space-temporal evolution of the concentration of each species. By using this theorem we obtain the variation on the species concentration per unit volume in the presence of sources and flows:

\[
\frac{\partial c_i}{\partial t} + \nabla \cdot \Phi_i - S_i(c_1, \ldots, c_n) = 0; \quad \Phi_i = -D_i \nabla c_i + c_i v_i
\]

where \(i=1,2,\ldots,n\) is the specie index and \(D\) is the diffusion tensor; \(c_i\), \(S_i\), \(\Phi_i\) and \(v_i\) are the terms associated with the concentration, the sources, the flows and the velocity field of the \(i\) specie, respectively.

![Encapsulated BZ-AOT System](image)

Figure 1. Axial section of the cylindrical reactor.

Taking into account the fact that the z-dimension of our system is much smaller than the global displacements of the reactor and, on the other hand, much larger than the mean free path between collisions for the particles of the reaction, it is possible to demonstrate (see below) that the convection terms in this equations are neglected; thus, only diffusion transport is present in the problem. For the source terms in eq. (1) we use the model described in [16] that it is commonly accepted to describe accurately the mechanism of the BZ-AOT system,

\[
\frac{\partial c_1}{\partial t} = \frac{1}{\epsilon} \left[ f c_2 \frac{q - c_1}{q + c_1} + c_1 \frac{1-mc_2}{1-mc_2 + \epsilon_1} - c_1^2 \right] + D_1 \nabla^2 c_1
\]

\[
\frac{\partial c_2}{\partial t} = c_1 \frac{1-mc_2}{1-mc_2 + \epsilon_2} - c_2 + D_2 \nabla^2 c_2
\]
where \(c_1\) and \(c_2\) are the concentrations of the main species competing in the pattern formation mechanism known as activator and inhibitor, respectively, and \(D_1, D_2\) are their corresponding diffusion coefficients. The parameters \(f, m, q, \varepsilon_1\) and \(\varepsilon\) depend on the reaction rates and they appear as a result of applying the mass action law on the chemical equations and a posteriori dimensionless analysis [17]. Appropriated values to model the BZ-AOT systems are \(f=1.85\), \(m=10\), \(q=0.004\), \(\varepsilon_1=0.02\) and \(\varepsilon=0.4\) [15-17]. In addition, it is necessary to consider, a difference of one order, at least, between the inhibitor and activator diffusion coefficients in order to produce Turing structures in this model, i.e. \(D_1 = 1\) and \(D_2 = 30\). This difference can be understood because of the different locations of both species; the activator remains inside of AOT nanodroplets and diffuses with the micelle. Meanwhile, the inhibitor (a nonpolar molecular compound) is able to diffuse through the oil phase and this process is much faster than that of the activator.

Furthermore, to complete the model we add the following Dirichlet and Newman conditions:

\[
\begin{align*}
\left. c_1(\vec{r},t) \right|_{\partial \Omega} &= c_{1,0}, & \hat{n} \cdot \nabla c_1 \bigg|_{\partial \Omega} &= 0 \\
\left. c_2(\vec{r},t) \right|_{\partial \Omega} &= c_{2,0}, & \hat{n} \cdot \nabla c_2 \bigg|_{\partial \Omega} &= 0
\end{align*}
\]  

(4)

Let us analyze now in detail the deep mechanism of the problem here presented that justify the absence of convection in our system. First, we proceed to estimate the boundary layer thickness, \(\delta\). Initially, we need to know the Reynolds number of the flow (in case it would exist). By supposing that a flow is present in the system, we may estimate the Reynolds number, \(Re\):

\[
Re = \frac{\rho \omega A L}{\mu} < 7
\]

(5)

here \(\rho\) (854.3 kg/m\(^3\)) and \(\mu\) (0.002 Pa·s) are the density and the dynamic viscosity of the BZ-AOT microemulsion, respectively; \(v=\omega A\) (0.2 m/s) is the typical velocity of oscillating flow at maximum forcing and \(L\) (80 \(\mu\)m) is the thickness of the reactor [7, 15]. The low value of \(Re\) demonstrates that the flow is laminar. Thus, we may estimate the boundary layer thickness by using the Blasius boundary layer model [18]:

\[
\delta = \frac{4.9d}{\sqrt{Re}} \approx 3.58 \text{ cm}
\]

(6)

where \(d\) (3.5 cm) is the diameter of the reactor (see figure 1). The boundary layer thickness in the worst case is several orders of magnitude larger than the thickness of the reactor (80 \(\mu\)m). Consequently, it is appropriate to consider that the flows are heavily influenced by the boundaries, so convective phenomena are strongly inhibited. And, thus, our system is within the boundary layer regime and we can assume that the transport mechanisms are only due to diffusive processes.

3. Mesoscopic description

As no convective flows are allowed in the system due to the mechanical restrictions, as described in the previous section, the only transport mechanism for the molecules is molecular diffusion, thus, the continuum hypothesis can be applied [19]. We developed a microscopic description, based on the statistical mechanics, in order to analyze the influence of the vertical forcing on the molecular diffusion coefficients.

The particle position is composed by the sinusoidal motion of the forcing plus the motion of the particles inside of the reactor, where the BZ-reaction takes places. Therefore, the position vector within the laboratory reference frame is:

\[
\vec{r} = x\hat{x} + y\hat{y} + z\hat{z} = x'\hat{x} + y'\hat{y} + (z' + A\sin(\omega t))\hat{z}
\]

(7)
where $x', y', z'$ is the position of the particles inside of the reactor. In the xy-plane, there are only thermal fluctuations, meanwhile in the z-direction there is also a sinusoidal forcing. We want to note that as the forcing amplitude ($A \sim 10^{-2} m$) is much greater than thermal fluctuations, i.e. $z' \ll Asin(\omega t)$, we assume that $z' \sim Asin(\omega t)$. Thus the resulting Hamiltonian, by neglecting variations in the gravitational contributions and introducing the vibration energy, is given by

$$H_{1,2} = \frac{\vec{p}_{1,2}^2}{2m_{1,2}} + m_{1,2} \omega \sqrt{A - z_{1,2}^2} + \frac{1}{2} m_{1,2} \omega^2 A^2$$

where $p_{1,2}$ and $m_{1,2}$ are the momentum and masses of the activator and inhibitor; $\omega$ and $A$ are the frequency and amplitude of the applied forcing. In addition, we assumed that the collisions are only between identical particles, i.e. 1-1 or 2-2. The molecules of different species, 1-2, do not collide with each other due to their different sizes and charges. Therefore, the canonical partition function associated with our system is:

$$Z_{1,2} = \frac{V}{\hbar^3} \left( \frac{2\pi m_{1,2}}{\beta} \right)^{3/2} \sum_{n=0}^\infty \left( \frac{-\beta m_{1,2} \omega^2 A^2}{2^n (2n+1) n!} \right)^n$$

here $V$ is the volume of the reactor, $\hbar$ is the Planck constant and $\beta = k_B T$ with $k_B$ as the Boltzmann constant and $T$ the temperature of the reactor. Thus, the momentum of our particles can be estimated and it happens to increase with the forcing parameters ($\omega$ and $A$) and with the mass of the particles ($m_{1,2}$):

$$\left\langle p_{1,2}^2 \right\rangle = \frac{m_{1,2}}{\beta} \left\{ 3 + \beta m_{1,2} \omega^2 A^2 \right\}$$

It is important to note that the momentum domain is always positive for all ranges of forcings, and also at zero forcing we recover the classical expression for free particles.

By statistical procedures [20], we estimate the collision number and the mean free path of our particles. With these results we obtain the following expression for the diffusion coefficients:

$$D_{1,2} = \frac{D_{1,2}^0 (\beta, m_{1,2})}{3^{3/2}} \left( 3 + \beta m_{1,2} \omega^2 A^2 \right)^{3/2}$$

where $D_{1,2}^0$ is the diffusion coefficient of the free particles given by:

$$D_{1,2}^0 (\beta, m_{1,2}) = \frac{1}{8 N_{1,2} \sigma_{1,2}} \sqrt{\frac{3}{\beta m_{1,2}}}$$

here $\sigma_{1,2}$ is the total cross section and $N_{1,2}$ is the number of particles per unit of volume for each species. It is important to note that to obtain simplified expressions in the calculations of the diffusion coefficients, we consider a first order approximation ($O(\omega^2 A^2)$) in the expansion of forcing term of the summations (as all forcings verify $\beta m_{1,2} \omega^2 A^2 << 1$). According with expression (11), the forcing on heavier particles is going to induce a larger effective diffusion coefficient. We want to emphasize that the diffusion coefficients $D_{1,2}$ also recover their classical value $D_{1,2}^0$ in absence of forcing.
4. Pattern formation

In order to understand the mechanism of Turing pattern formation, we perform a standard stability analysis [21] of the reaction-diffusion equations (4, 5). Thus, we test a general solution for chemical waves (concentration waves):

\[
\bar{c} = \begin{pmatrix} c_1 \\ c_2 \end{pmatrix} = \overline{C} \exp \left( \lambda t + i \frac{k r}{\sqrt{n}} \right) \tag{13}
\]

where \( \lambda \) is the eigenvalue, \( k \) is the wavenumber and \( n \) is the number of space dimensions. Through a linear stability analysis we obtain the following expression for the Turing eigenvalue:

\[
\lambda_{1,2} = \frac{1}{2} \left[ \text{tr}(J) - k^2 \text{tr}(D) \pm \sqrt{\left( k^2 \text{tr}(D) - \text{tr}(J) \right)^2 - 4p(k^2) \right] \tag{14}
\]

with,

\[
p(k^2) = k^4 |D| - k^2 \text{tr}(D^* J) + |J|, \tag{15}
\]

\[
D = \begin{pmatrix} D_1 & 0 \\ 0 & D_2 \end{pmatrix}; \quad D^* = \det(D) D^{-1} \tag{16}
\]

here \( J \) is the Jacobian matrix associated with the linearization. Turing patterns occur when the eigenvalue is positive, for some wavenumber. Then, only one wavenumber can generate the pattern with a characteristic wavelength. It is important to note that the final wavelength of the pattern comes from the wavenumber which provides the most unstable value for the eigenvalue, \( k_0 \). Thus, by maximizing the eigenvalue we obtain:

\[
\left. \frac{dp(k^2)}{dk} \right|_{k_0^2} = 0 \quad \Rightarrow \quad k_0^2 = \frac{\text{tr}(D^* J)}{2|D|} \geq 0 \tag{17}
\]

Figure 2 shows the set of wavenumbers\(^1\) that may produce Turing instability and, thus, Turing structures and how this region decreases as the forcing amplitude is increased. For larger values of the forcing (\( \omega' A^2 > 6.22 \text{m}^2\text{s}^{-2} \)) there are not wavenumbers that produce unstable eigenvalues. Moreover, the most unstable wavenumber, \( k_0 \), is shifted to lower values when the forcing is applied. These results suggest that the Turing wavelength (\( A = 2\pi/k_0 \)) is increasing with \( \omega' A^2 \). Thus, we establish that the forcing modifies the configuration of the patterns (given by their wavelength) and it can even inhibit their formation.

\(^1\) \text{i.e. } k^2 \text{ such that } \text{Re} \left[ \lambda(k^2) \right] > 0.
5. Numeric Simulations

In order to verify the predictions of the previous theoretical model, we present numeric simulations of the reaction-diffusion equations (2) and (3). First of all, the effect of the forcing amplitude on the parameter space was analyzed. This phase diagram is plotted in figure 3 where the regions were Turing structures appear are plotted for the different values of the parameters $f$ and $m$. The main effect of increasing the forcing amplitude is that the Turing region shrinks until it completely disappears. This diagram reflects the tendency observed in figure 2; large values of the forcing makes the Turing region diminishes. The largest forcing amplitude considered in this figure ($A^2 \omega^2 = 15 \text{m}^2\text{s}^{-2}$) is marked in the figure with empty circles (denoted by region $T_{15}$ in the figure) and covers the smallest area in the diagram. On the other hand, when no forcing is considered ($A^2 \omega^2 = 0 \text{m}^2\text{s}^{-2}$, region $T_0$ in figure 3 and marked with dark spots) the range of parameters that yield to Turing patterns is maximum. Please note that as the forcing amplitude is increased, the Turing region is included into the previously calculated, i.e., the region for $A^2 \omega^2 = 0 \text{m}^2\text{s}^{-2}$ contains all other plotted regions (T3 to T15); the region for $A^2 \omega^2 = 3 \text{m}^2\text{s}^{-2}$ (called T3 in figure 3) contains all other Turing regions calculated for higher forcing amplitudes (T6 to T15); and so on. Also note that the symbol marking each point in the $f$-$m$ phase diagram shows the maximum forcing the system can experience still exhibiting Turing structures; if the symbol corresponds with T0 region then any forcing inhibits the appearance of structures. If the point in the phase diagram is marked with T3, then any forcing above $A^2 \omega^2 = 3 \text{m}^2\text{s}^{-2}$ inhibits Turing structures.

We considered a set of parameter values that we considered as standard in the following simulations. This is marked in figure 3 with an arrow and correspond with the following values, $f=1.85$, $m=10$, $q=0.004$, $\varepsilon_1=0.02$ and $\varepsilon=0$. In this case, Turing structures are observed for forcing amplitudes up to $A^2 \omega^2 = 6.22 \text{m}^2\text{s}^{-2}$. Above such threshold, i.e. $A^2 \omega^2 > 6.22 \text{m}^2\text{s}^{-2}$, the forcing changes the characteristic eigenvalue from unstable ($\text{Re}(\lambda)>0$) to stable ($\text{Re}(\lambda)<0$) and the system is not able to exhibit the Turing instability.

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$^2$ For example, for the point $f=1.9$ and $m=60$ (inside T0 region) in the phase diagram in fig 3, the maximum forcing that still exhibits Turing structures is $0 \text{m}^2\text{s}^{-2}$ so this region is very sensitivity to the forcing, however for the point $f=1.7$ and $m=39$ (inside T12 region), the maximum forcing supported is $12 \text{m}^2\text{s}^{-2}$.
Figure 3. Phase Diagram in the \( f-m \) space of the different Turing regions (T) corresponding with different forcing amplitudes (\( A^2 \omega^2 \)). Other model parameters are kept constant and equal to \( q=0.004 \), \( \varepsilon_1=0.02 \) and \( \varepsilon=0.4 \). T0, T3, T6, T9, T12 and T15 stands for the Turing regions for forcing amplitudes equal to 0 \( m^2s^{-2} \), 3 \( m^2s^{-2} \), 6 \( m^2s^{-2} \), 9 \( m^2s^{-2} \), 12 \( m^2s^{-2} \) and 15 \( m^2s^{-2} \) respectively. Note that the different Turing regions (corresponding with different values of the forcing amplitude) are overlapped, i.e., T0 region contains all T3 to T15 regions, T3 region contains regions T6 to T15, and so on (\( T0 \supset T3 \supset T6 \supset T9 \supset T12 \supset T15 \)).

Numerical simulations of the equations (2) and (3) were performed by using a three level explicit algorithm with a centered in space discretization [22]. The computational domain consists in a 300x300 grid elements with space and time steps values of 0.001 t.u. and 0.5 s.u., respectively. For initial conditions we set both variables with random initial concentration around the 5 percent of the steady-state concentrations (i.e. the fixed points of the equations (2) and (3)). We present in figure 4 a sequence of pictures of the 2D Turing pattern for different forcing amplitudes.

Figure 4. Two-dimensional Turing structures obtained with the model (4),(5) and (11). Parameters: \( f=1.85 \), \( q=0.004 \), \( m=10 \), \( \varepsilon=0.4 \) and \( \varepsilon_1=0.02 \). The value of the diffusion coefficients (\( D_v \) and \( D_u \)) depends on the applied forcing following the equation (11). Figure (a) shows the pattern at rest, i.e. \( \omega^2 A^2=0 \ m^2s^{-2} \). In figures (b) and (c) the system was forced with a magnitude of \( \omega^2 A^2=3.00 \ m^2s^{-2} \) and \( \omega^2 A^2=6.00 \ m^2s^{-2} \), respectively. In figure (d) the system is very close to the transition region (see figure 2), the forcing is \( \omega^2 A^2=6.22 \ m^2s^{-2} \). In figure (e) the forcing is \( \omega^2 A^2=9.00 \ m^2s^{-2} \) and the system is clearly out of the Turing region and patterns disappear totally.
Note that the different Turing patterns exhibit a different wavelength depending on the applied forcing (figures 4a-c). Thus, we obtain numerically an increase on the Turing wavelength, in a good agreement with the theoretical predictions. When the forcing is $\omega A^2 = 6.22 \text{ m}^2\text{s}^{-2}$ (figure 4d) the maximum of the eigenvalue is zero, the onset of Turing instability. For this forcing, the system evolves to a diffuse structure with a wavelength clearly not defined. Finally, with even larger forcings ($\omega A^2 > 6.22 \text{ m}^2\text{s}^{-2}$), the maximum of the eigenvalue is negative and the system, which is in a stable configuration, evolves to a uniform value of the concentration (figure 4e) determined by the fixed points.

6. Discussion and conclusions

In this work, we studied Turing pattern formation under external harmonic vibrations in the BZ-AOT active media. The characteristics of the system are such that convective flows are not allowed, and so, only diffusion processes can take place. In this case, the energy of the forcing is spent on increasing the internal degrees of freedom of the system, and, therefore, the molecular diffusion is modified producing macroscopic effects. Pattern structure is highly sensitive to the diffusion process. We determine the relationship between the modified diffusion coefficients (due to the presence of forcing) and the resulting pattern.

We developed a mesoscopic model, based on statistical mechanics, to determine the changes in the diffusion coefficients with the forcing. By coupling this model to the reaction-diffusion equations we calculate the threshold of Turing instability. Using a linear stability analysis we find a shortening in the set of wavenumbers that are able to produce Turing instability as the forcing increases. In addition, an increase in the forcing induces a shift of the most probable eigenvalue to lower values, and therefore the pattern wavelength increases. Moreover, large values of the forcing amplitude (up to $\omega A^2 = 6.22 \text{ m}^2\text{s}^{-2}$) produce stable values for the corresponding eigenvalues, and consequently, the system moves from the Turing regime into a uniform state of concentration.

Numerical simulations of the reaction-diffusion model, demonstrate a good agreement with the predictions. In figure 4 we show a Turing pattern with hexagonal arrangement and how the associated wavelength increases with the forcing until the Turing structure disappears.

The mesoscopic approach, that we present in this work, can be applied to other situations. We believe it may result helpful in understanding other systems where an external forcing modifies the transport mechanism through diffusion (in particular when the system is highly influenced by boundary layer effects).

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