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# A theory for one-dimensional asynchronous chemical waves

**A Bhattacharyay**

Dipartimento di Fisika 'G. Galilei', Università di Padova, Via Marzolo 8, 35131 Padova, Italy

E-mail: [arjit@pd.infn.it](mailto:arjit@pd.infn.it)

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## Abstract

We present a theory for an experimentally observed phenomenon of one-dimensional asynchronous waves. It has been proposed that two oppositely moving travelling wave states can localize an oscillatory core structure. A class of even and odd parity localized solution of the same system has been explicitly shown. It has been argued that a combination of these local and global states in an extended system can produce asynchronous waves. We also produce numerical results in support of our analytic predictions.

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One-dimensional asynchronous chemical waves (or 1D spirals) were first reported by J-J Perraud *et al* [1] in 1993. An explanation of this phenomenon had been put forward on the basis of bi-stability of Turing and Hopf states and non-variational effects [1]. Historically, in those days when the above-mentioned experiment was carried out, studying droplet of other states inside a global one under uniform conditions was a topic of tremendous interest. Presumably, being influenced by those new observations [2–5], the bi-stability of Turing and Hopf modes and their interaction was considered to be the basic underlying principle on which a possible explanation of experimentally observed 1D spirals was given. Such localized droplets of global states near a Hopf–Turing instability boundary have also been analysed in many subsequent papers [6–12]. However, a proper understanding of the true nature of the parity breaking core which generates asynchronous waves on opposite sides is still lacking. It is also important to have a proper understanding of this novel phenomenon of asynchronous waves because such a thing shows up near a Hopf instability threshold where behaviours of dynamical systems are quite universal. In the present paper we are going to demonstrate an alternative scenario for 1D spiral (asynchronous waves) generation. We are going to present our alternative theory on the basis of a complex Ginzburg–Landau-type amplitude equation (CGLE). We consider the simplest form of such a universal amplitude equation to demonstrate our results. In what follows, we argue that the global travelling wave solutions of a CGLE

can restrict some oscillatory odd-parity localized solutions of the linear CGLE from spreading out. When restricted to a small region, these localized solutions (of essentially the linear equation) do not get perturbed by the nonlinearity of the system and can stay longer. In return, the odd-parity oscillating core helps oppositely moving travelling waves on two sides of it remain asynchronous to each other. We also illustrate this simple mechanism of formation of asynchronous waves by numerical simulation of the CGLE.

Let us first present a short description of what people have already seen experimentally [1]. 1D spiral pattern was experimentally observed in CIMA (chlorite iodide malonic acid) reaction. In such a reaction one can control transition from Hopf (oscillatory) to Turing (stationary) states by tuning the concentration of the starch or the malonic acid. Similar transitions between a stationary periodic (Turing) and a travelling wave like Hopf mode were observed by lowering the starch concentration in the chemical reactor or by keeping a low starch concentration and then increasing the malonic acid concentration. Since it is difficult to change the starch concentration of the reactor, the malonic acid concentration had been used as the bifurcation parameter and was tuned. It had been observed that when the Hopf state takes up from the Turing state, very often there remain a few spots (considered to be reminiscent of the previous Turing state from the time-averaged concentration profile [1]) acting as the source of one-dimensional anti-synchronous wave trains. Bands of maximum intensity were spreading alternatively towards right and left of the central core with a time delay. At a particular moment of time these asynchronous wave trains look like a section of a 2D spiral on a line passing through its core and that is why the other name 1D spiral. The very basic and primary question that arises in connection is exactly who breaks the parity sitting in the middle and how. The experiment suggests the symmetry breaking agent is local and endogenous in nature [1]. Our purpose in this paper is to identify and show the localized structure that can break parity and put forward plausible mechanism as to how global asynchronous waves are supported by this localized structure.

The CGLE with a cubic nonlinearity looks like

$$\frac{\partial H}{\partial t} = \epsilon H + (D_r + iD_i) \frac{\partial^2 H}{\partial x^2} - (\beta_r + i\beta_i) |H|^2 H. \quad (1)$$

This is a simple amplitude equation at a Hopf instability threshold. Near the instability threshold, the dynamics of slow amplitude modes is universal. Since, the observed phenomenon of 1D spirals appears in a region of Hopf phase and close to the instability boundary we employ such an equation. Equation (1) has a one-parameter family of travelling wave solutions of the form  $H = H_0 e^{i(\omega t - kx)}$  where the amplitude and the frequency of oscillation are related as

$$|H|^2 = \frac{D_i \epsilon + D_r \omega}{D_i \beta_r - D_r \beta_i}. \quad (2)$$

Here,  $\epsilon$  is the bifurcation parameter—meaning that by tuning  $\epsilon$  one can cross the primary instability boundaries. It is important to note that equation (1) is invariant under a change in sign of  $H$ . So, the opposite parity states are solutions of this equation on the same footing. This equation is also invariant when  $H$  and  $x$  change sign together. So, one can always expect to have parity-inverted states on the two sides of the origin. At this point, we propose that such a pair of opposite parity travelling wave states are connected to a localized structure of appropriate form at the origin. The form of this localized entity will be revealed in what follows keeping in mind that it has to match with the bounding states which keeps it localized

The CGLE has the form of a linear Schrödinger equation when we neglect the nonlinear term. As we know a harmonic well potential localizes the solutions in a Gaussian envelop, we expect to have the same spatial solution from the linear CGLE. We neglect the nonlinear

part on the basis that we are after a localized solution of the form  $\frac{x}{\sqrt{b}} e^{-x^2/2b}$ . At large  $b$  and considering that travelling wave states of the full CGLE will help keep the central core bound in a small region at the origin. The CGLE (equation (1)) without nonlinear term can still have a solution  $H e^{i\omega t}$ . With this ansatz in place we can write equation (2) as

$$i\omega H + \frac{\partial H}{\partial t} = \epsilon H + (D_r + iD_i) \frac{\partial^2 H}{\partial x^2}.$$

Let us localize the solution of this linear equation with a Gaussian envelop as  $H = H e^{-\frac{x^2}{2b}}$ . With this consideration the above equation changes to

$$(D_r + D_i) \left[ \frac{\partial^2 H}{\partial x^2} e^{-\frac{x^2}{2b}} - \frac{2x}{b} \frac{\partial H}{\partial x} e^{-\frac{x^2}{2b}} + \frac{x^2 H}{b^2} e^{-\frac{x^2}{2b}} - \frac{H}{b} e^{-\frac{x^2}{2b}} \right] + (\epsilon - i\omega) H e^{-\frac{x^2}{2b}} = 0.$$

The Equations result by equating real and imaginary parts look like

$$D_r \left[ \frac{\partial^2 H}{\partial x^2} - \frac{2x}{b} \frac{\partial H}{\partial x} + \frac{x^2 H}{b^2} - \frac{H}{b} \right] + \epsilon H = 0$$

and

$$D_i \left[ \frac{\partial^2 H}{\partial x^2} - \frac{2x}{b} \frac{\partial H}{\partial x} + \frac{x^2 H}{b^2} - \frac{H}{b} \right] - \omega H = 0.$$

Now, considering the one that comes from the real parts and rearranging the terms we get

$$D_r \frac{\partial^2 H}{\partial x^2} - \frac{2D_r x}{b} \frac{\partial H}{\partial x} + \frac{D_r x^2}{b^2} H + \left( \epsilon - \frac{D_r}{b} \right) H = 0. \tag{3}$$

We are looking for a polynomial solution of the dimensionless amplitude  $H$ . So, nondimensionalize the length as  $z = x/\sqrt{b}$ , which is also suggested by the form of the above equation. We also drop the term containing  $x^2/b^2$  which is small for large  $b$  where  $\frac{x^2}{b^2} \sim \frac{1}{b}$ . Actually, our purpose is served with the expected Hermite polynomial solution of order unity since it has odd parity and we need not consider higher order terms. Thus, we get the equation

$$\frac{\partial^2 H}{\partial z^2} - 2z \frac{\partial H}{\partial z} + \frac{b}{D_r} \left( \epsilon - \frac{D_r}{b} \right) H = 0. \tag{4}$$

The linear equation (4) admits solutions which are Hermite polynomials and a solution of order zero is obtained when  $(\epsilon - \frac{D_r}{b})$  is equal to zero. When  $\epsilon - \frac{D_r}{b} = \frac{2D_r}{b}$  it admits a solution same as the Hermite polynomial of order unity. Now, we have explicitly got a small amplitude-localized asymmetric solution of the form  $\frac{x}{\sqrt{b}} e^{-x^2/2b}$  which oscillates with a frequency  $\omega$ . One important thing about this linear localized solution is that it can always have an arbitrary constant factor which can also keep it small apart from the requirement of large  $b$ . So, in a number of different ways one can justify the non-functioning of the nonlinear term for such a localized solution. Before we show the asynchronous wave solutions we consider the equation got from equating the imaginary terms of the CGLE. If both the equations are to have the same asymmetric localized spatial solutions we get a selection for the oscillation frequency given by

$$\omega = -\frac{\epsilon D_i}{D_r}. \tag{5}$$

Now, take the travelling wave state  $H_0 e^{i(\omega t - kx)}$  of the full CGLE (equation (1)) into consideration. This travelling wave state be present on the positive side of the  $x$ -axis (right-hand side of the core) having the localized core at the origin. The localized solution in

the middle sets the boundary condition on the left-hand side boundary of this travelling wave state. This can be done keeping in mind that the amplitude of the travelling wave solution is a function of the parameters of the system and in reality such a matching can always happen by tuning the parameters. Consider the  $\omega$  of the travelling wave to be the same as the oscillation frequency of the central asymmetric core. Now, putting the expression of  $\omega$  (equation (5)) of the oscillating core in that of the amplitude (equation (2)) of the travelling wave one gets

$$|H|^2 = \frac{D_i(\epsilon - \epsilon_l)}{D_i\beta_r - D_r\beta_i}, \quad (6)$$

where  $\epsilon_l$  is the local value of the bifurcation parameter where the core has formed. With the numerator of equation (6) positive one can say that  $D_i\beta_r > D_r\beta_i$  is the condition for frequency matching. It is also evident that for the existence of such a combination of the linear and nonlinear solutions a local variation in the bifurcation parameter is essential. In the results of numerical simulation it will be shown that with this local variation of the bifurcation parameter such a co-existence of linear and nonlinear states is more stable. It is important to note that in the actual experiment [1] the bifurcation parameter was malonic acid feeding rate. It is very difficult if not impossible to maintain exactly the same feeding rate along the whole length of the reactor. As a result there can always be localized fluctuations in the bifurcation parameter. These regions can develop into endogenous sources (or core) from which asynchronous waves generate.

Now, apply the boundary condition to the wave that moves in the positive  $x$ -direction from the point  $x = C$  (say), with the localized core extending upto the point  $x = C$ . The amplitude of the travelling wave ( $H_{0r}$ ) is given by

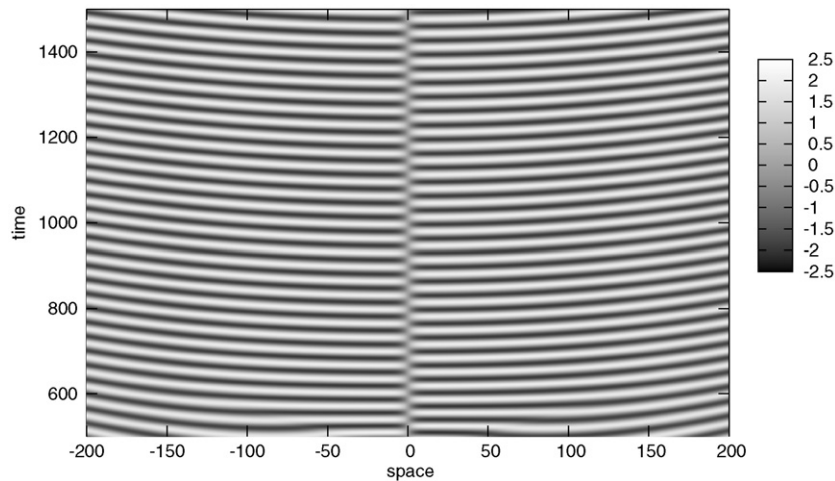
$$H_{0r} = \frac{C}{\sqrt{b}} e^{-C^2/2b+ikC}. \quad (7)$$

By matching a similar wave moving towards negative  $x$ -direction from near the left-hand side of the asymmetric localized core we will get its amplitude  $H_l$  as

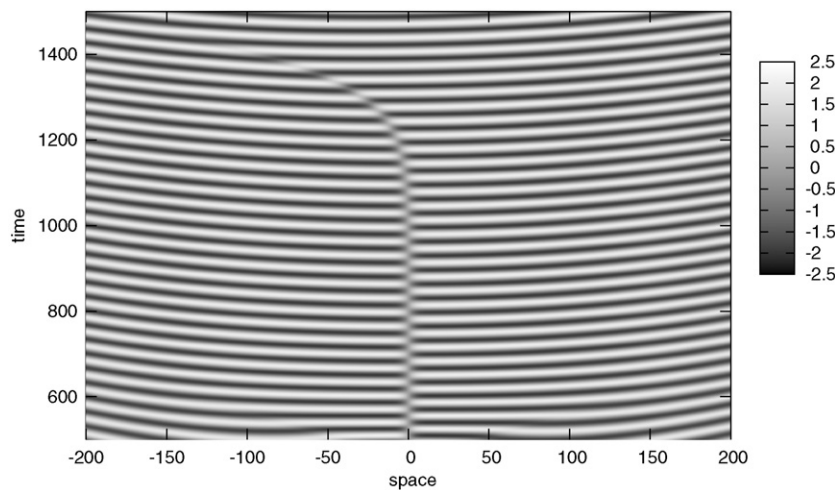
$$H_{0l} = -\frac{C}{\sqrt{b}} e^{-C^2/2b+ikC}. \quad (8)$$

The boundary conditions on the travelling wave states cause a clear phase difference of  $\pi$  in the left and the right moving waves due to obvious reasons ( $k$  and  $C$  are negative for the left moving wave). Apart from that phase difference everything else are identical on the both sides of the origin. This is actually no surprise given the fact that the CGLE is invariant under the inversions  $H = -H$  and  $x = -x$ . The  $e^{ikC}$  term in the amplitude ensures that the points  $x = \pm C$  are the origin of the travelling wave states and the oppositely moving waves exist beyond them. Simple matching of solutions shown here in order to demonstrate a phase inversion of travelling wave states might be simplistic in comparison with what happens in reality. Nevertheless, whatever be the detailed mechanism the symmetry suggests that the global wave states on the two sides of the core should remain parity inverted. The more important point here is the stability of such a combination. In what follows we establish the stability of such a system by numerical simulation.

To justify the analytic arguments we simulated the full nonlinear CGLE. A finite difference scheme (Crank–Nicolson formula) with implicit method of integration has been employed. The nonlinearity is tackled by the predictor corrector rule. The numerical integration has been performed on a linear lattice of 2001 points with a small parity breaking initial seed symmetrically at a very small region (five lattice points) on both sides of the centre of the lattice. We have employed no flux boundary conditions. Prominent asynchronous waves develop and

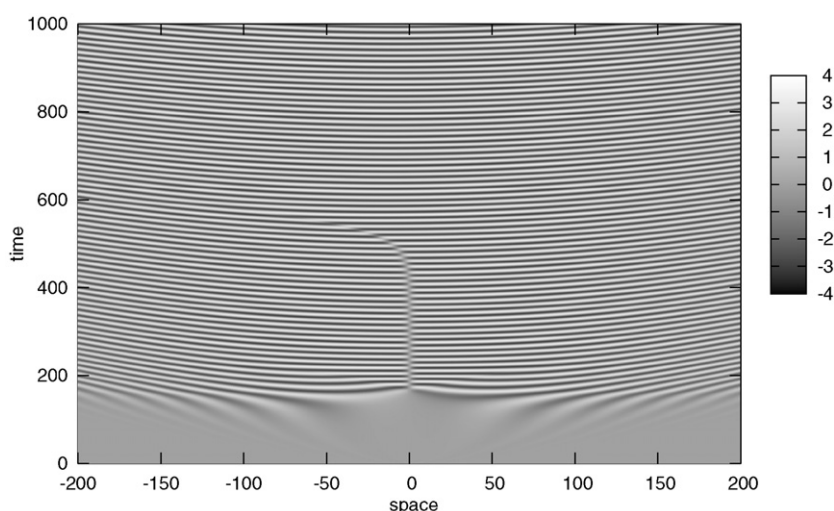


**Figure 1.** The concentration profile of the asynchronous wave generated from an oscillatory localized structure. The bifurcation parameter at a core region in the middle is 0.02 and elsewhere it is 0.05.



**Figure 2.** The concentration profile of the asynchronous wave generated from an oscillatory localized structure. The bifurcation parameter is 0.02 everywhere.

spread from asymmetric oscillatory core at the middle. Figure 1 shows the space time plot of the concentration waves asynchronously moving in the opposite sides. The parameter values at which the simulation has been done are  $(D_r, D_i) = (0.2, 2.0)$ ,  $(\beta_r, \beta_i) = (0.005, 0.05)$  and  $\epsilon = 0.02$  in a small core at the middle where the initial seed has been given. In the rest of the lattice  $\epsilon = 0.05$ . We see that asynchronous wave trains generated by the oscillatory core remains stable for a wide range of time. Similar patterns are observed on some range of the parameter values about that mentioned. Now, keeping all other parameters exactly the same if we make  $\epsilon = 0.02$  everywhere on the lattice we get the result shown in figure 2. Due to the action of the seed in the middle the oscillatory core is generated which in turn produces

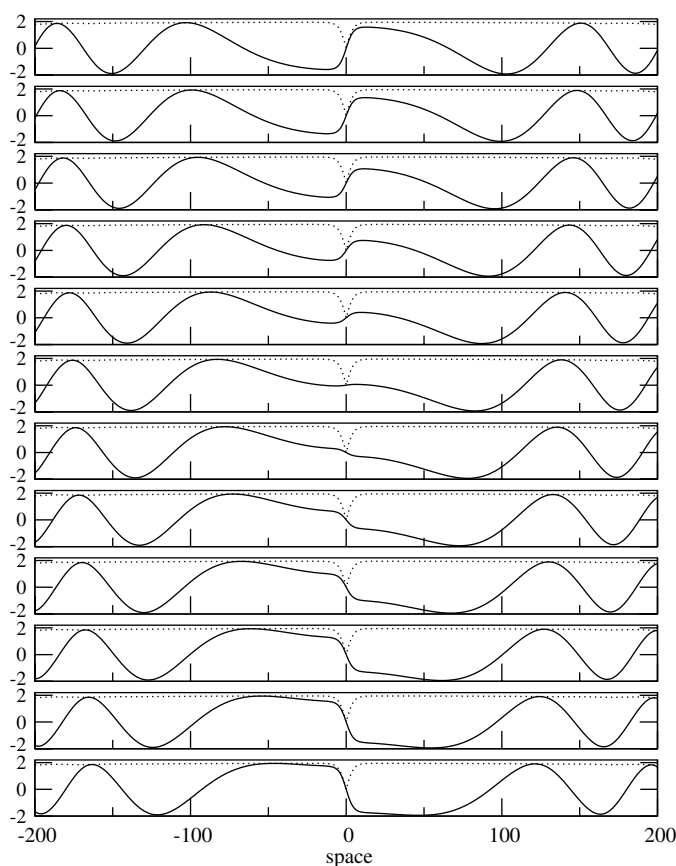


**Figure 3.** The concentration profile of the asynchronous wave generated from an oscillatory localized structure. The bifurcation parameter is 0.05 everywhere.

asynchronous waves, but these waves are dying out early. Figure 3 shows another plot at  $\epsilon = 0.05$  everywhere with all other parameters being the same. Here, the asynchronous wave dies out even early. It is important to note that in this case the  $\epsilon$  at the core is  $2\frac{1}{2}$  times that of figure 1. As a result the oscillation frequency of the central core structure should increase by that same factor (see equation (5)). Since, it is this core that generates the travelling waves, figure 3 has bright and dark lines more closely spaced which clearly demonstrate a larger frequency. Instead of having the same value of the  $\epsilon$  everywhere except in the middle figure 1 corresponds to smaller frequency waves. To demonstrate this frequency variation with the variation of the bifurcation parameter in the middle we have plotted figure 3 at the same time scales as the other ones.

To reveal the nature of the central core more clearly, we have plotted a few snapshots of the core part in figure 4. Here, we see almost a half period of its oscillation. In this figure time runs up-wards through the frames in steps of equal size (10 unit). The parameter values for figures 4 and 1 are the same. we can see an almost straight line graph (continuous) oscillating in the middle. The amplitude of oscillation is shown by the dotted curve which is every where almost constant except in the middle where it symmetrically goes to zero. This is quite expected for the form of localized solution predicted. As we have mentioned earlier, the simple matching of two solutions (equations (7) and (8)) may be simplistic. It is also difficult to identify exactly at which point (C) the local and global states meet and also determining the exact value of the parameter  $b$ .

We conclude by saying that an asymmetric localized oscillatory structure has been identified as the parity breaking agent for asynchronous one-dimensional chemical waves. Based on this observation we put forward an alternative theory. The present theory rests on the principle of matching and stabilizing localized linear solutions by nonlinear global states of a general nonlinear model in an extended system. The global states keep the central core localized in a small region and the central oscillatory core helps global states remain parity inverted. Localized fluctuations in the bifurcation parameter are needed to create and sustain such a phenomenon and thus the endogenous nature of the 1D spirals are explained. Such a



**Figure 4.** Snapshots of the central oscillatory region at equal time gaps.

situation is very general to our belief and can explain a lot of complex phenomenon as observed in extended nonlinear systems. The linear part with its characteristics symmetry and parity can interact with the nonlinear solutions to generate a range of complexities. The present theory also predicts the presence of synchronous waves originating from symmetric sources and sinks as the asymmetric ones on the same footing (corresponding to Hermite polynomials of even order). Even, the existence of higher order localized structures is quite plausible and it would be interesting to investigate experimentally and numerically the role played by these structures in an extended system.

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