

**Waving patterns: A general transition from stationary to moving forced Turing structures**

Igal Berenstein and Alberto P. Muñuzuri

*Group of Complex Systems, Universidade de Santiago de Compostela, E-15782, Santiago de Compostela, Spain*

(Received 18 April 2006; published 5 September 2006)

We perform experiments on the chlorine dioxide-iodine-malonic acid (CDIMA) reaction forced with light with a pattern of moving stripes in which the spatiotemporal behavior is the oscillating movement of stripes (waving). This behavior is seen for different relative wavelengths between stationary and moving patterns. Different velocities of forcing may produce different modes of relaxation of the pattern in order to get to the natural Turing wavelength. Zigzag or Eckhaus instabilities may affect the symmetry of the pattern but do not influence the waving movement of stripes.

DOI: [10.1103/PhysRevE.74.036202](https://doi.org/10.1103/PhysRevE.74.036202)

PACS number(s): 82.40.Ck

**I. INTRODUCTION**

Pattern formation is a subject of great interest in many fields [1]. In general, and in reaction-diffusion systems in particular, some very interesting patterns appear due to resonance [2]. Other striking phenomena can be observed when different instabilities interact, such as Turing and Hopf [3] and Turing and wave instability [4], to mention a few.

In order to be able to see resonant phenomena, very good control of parameters is needed, and this can be achieved by forcing the system in a controlled way. This way, it was possible to study resonant patterns in the Belousov-Zhabotinsky reaction [5], as well as spatial resonances in Turing patterns in the chlorine dioxide-iodine-malonic acid (CDIMA) reaction [6,7]. Forcing can also simulate interaction of different instabilities, and has been used to force Turing patterns with a train of waves [8–10,16]. These experiments have focused on the case where the wavelength of the moving stripes is the same as the wavelength of the Turing patterns (i.e., spatial resonance). As expected, for high velocities of the forcing, the pattern only perceives the forcing as a constant (in space and time) illumination, while for very low velocities, the pattern moves entrained with the forcing [8]. As for intermediate cases, several unexpected behaviors were observed, which include modulation of the amplitude of the pattern as well as modulation of the velocity at which it moves and a back and forth motion of the forced stripes [10]. In some cases, a switch in the type of pattern, from labyrinthine to hexagonal arrangement, is also observed [9].

In this paper, we inspect the effect of the wavelength of the forcing (compared to the Turing wavelength) and the velocity of the moving stripes.

The paper is organized in four sections. In Sec. II we give a brief description of the experimental setup and the experiments. In Sec. III the results are presented and Sec. IV gives the conclusions.

**II. EXPERIMENTAL PART**

The experiments are carried out using the chlorine dioxide-iodine-malonic acid (CDIMA) reaction, which is photosensitive [11,12]. The reaction takes place in an agarose gel (2%), 0.3 mm thick. The gel is separated from the feeding chamber by an anapore membrane (Whatman, pore

size 0.25  $\mu\text{m}$ , to hold the gel and to avoid stirring effects on the pattern) and a nitrocellulose membrane (Scheicher & Schuell, pore size 0.45  $\mu\text{m}$  for contrast). A more detailed explanation of the setup can be found in Ref. [10]. Three solutions are pumped constantly into the reactor. One contains iodine (Aldrich), a second one contains malonic acid (MA, Sigma) and polyvinyl alcohol (PVA, Aldrich, Mw 9000-10 000), and a third one containing chlorine dioxide. All three solutions are made in 10 mM sulfuric acid. The residence time is kept at  $260 \pm 10$  s and the temperature is set to 4 °C. Under these conditions, the reaction produces a labyrinthine Turing pattern. The concentrations used are  $[\text{MA}]_0 = 1.4$  mM,  $[\text{ClO}_2]_0 = 1$  mM, and  $[\text{I}_2]_0 = 0.4$  mM

Just as in Refs. [8–10], the experiments start by eliminating the existing pattern with homogeneous light of high intensity (2500 lx). Then a pattern of stripes with the forcing wavelength is imposed for 20 min after which the spatiotemporal forcing begins. Contrary to previous experiments, the pattern is not allowed to evolve freely to avoid disordering of the pattern due to transverse instabilities, or splitting of stripes [13,14]. The mean intensity of illumination used is 450 lx.

**III. RESULTS**

For very high velocity of forcing (in the order of several mm/min), the behavior is the same as for relaxation after imposing a pattern of stripes [14]: Splitting in two for imposed stripes with two times the Turing wavelength, splitting in two and then becoming zigzag patterns for imposed stripes with three times the natural wavelength, and two consecutive splittings for stripes with four times the wavelength of the unforced pattern. This indicates that the global illumination does not change the type of patterns that are observed.

For relatively high velocities of forcing (around 0.4 mm/h), the initial behavior is splitting of stripes, but in the case of imposed stripes with about four times the Turing wavelength, the splitting is into three stripes instead of two successive splittings [which can be seen in Fig. 1(b)]. For any chosen wavelength, what can be seen is that at times some stripes are more intense than others, but they do not move. This behavior is seen in Fig. 1(a) for imposed stripes with twice and four times [in Fig. 1(b)] the wavelength of the

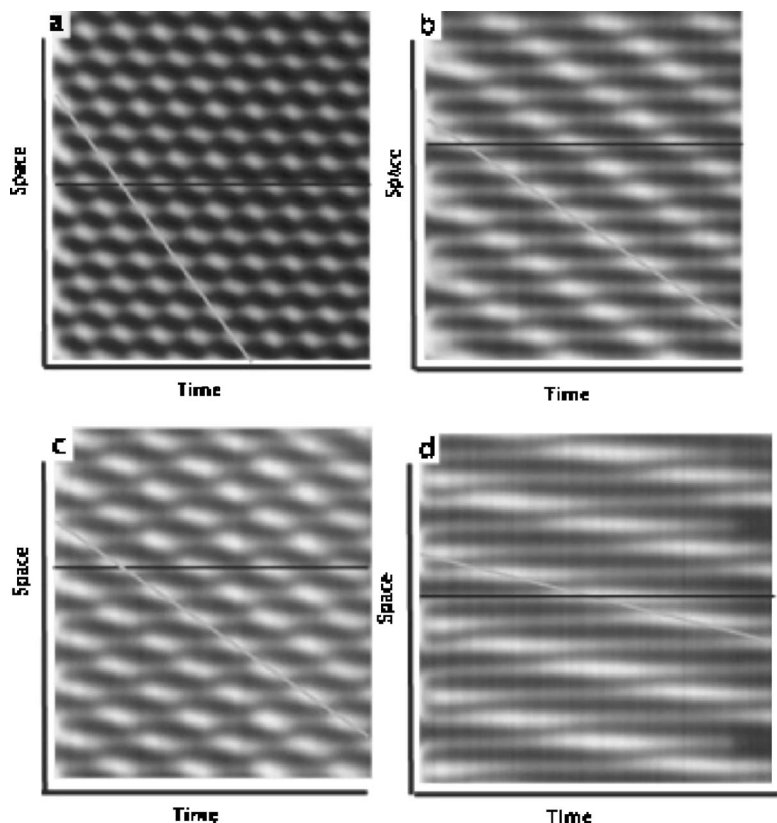


FIG. 1. Space-time plots of modulated and waving patterns. The relative wavelength of forcing with respect to the Turing wavelength is 2 [(a) and (c)] and 4 [(b) and (d)]. Space corresponds to 10 mm in all cases.

unforced Turing patterns. Notice that the average position remains constant.

For slower velocities (around 0.2 mm/h), the initial behavior is the same as seen for higher velocities, but in this case the stripes do move. The stripes that are forced by the moving bands (one out of each two or three) move along until they get too close to an unforced stripe, where the forc-

ing band changes the stripe that is forcing. This behavior is seen in Figs. 1(c) and 1(d). Figure 2 shows some of the patterns seen at different times. While in Fig. 2(a) and 2(b), we see stripes which preserve the distance between them; in Fig. 2(c) and 2(d) we see stripes that are closer to stripes on one side than in the other.

It is expected that for very low velocities of forcing, the pattern is going to move. However, as the wavelength of the forcing is higher than the wavelength of the pattern, the pattern does not get entrained by the forcing, but moves stepwise. This behavior is seen for a forcing with twice the Turing wavelength in Fig. 3. The behavior is almost as for the waving stripes, but the returning movement is a little slower and does not come back to its initial position.

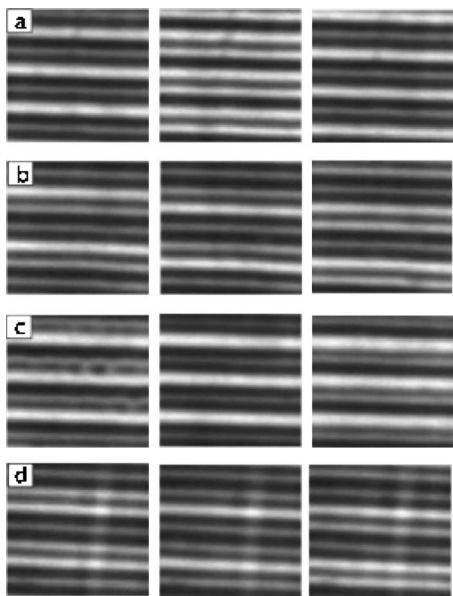


FIG. 2. Pictures of patterns corresponding with Fig. 1. The picture size is  $5 \times 5 \text{ mm}^2$ . Pictures taken 30 min (a), 1 h (b), and 3 h (c) and (d) apart.

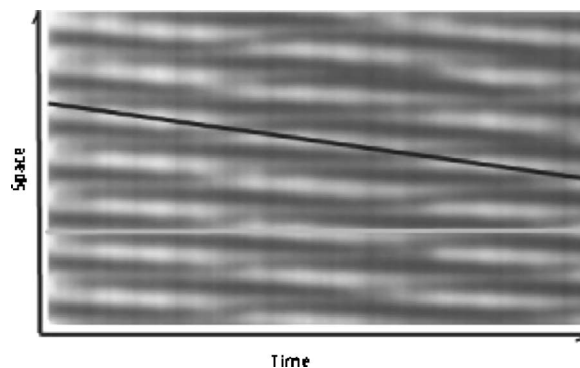


FIG. 3. Space-time plot for moving patterns with a relative wavelength of forcing of two times the Turing wavelength. Space corresponds to 10 mm.

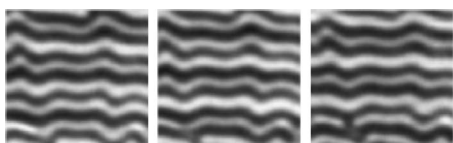


FIG. 4. Waving of zigzag patterns. Pictures taken 1 h apart. The relative wavelength of forcing is three times the Turing wavelength. The size of the pictures is  $5 \times 5 \text{ mm}^2$ .

Waving of stripes is also seen when the pattern relaxes to zigzag patterns (see Fig. 4), or when the pattern is distorted due to Eckhaus instability after splitting [13,14]. However, depending on the velocities of the moving stripes, the dynamics that lead to attaining the Turing wavelength may change, and for slow velocities and a relative wavelength of forcing of three times the Turing wavelength, the imposed stripes split in three rather than splitting in two and then becoming zigzags. All these behaviors are summarized in Fig. 5, where a phase diagram is shown.

IV. CONCLUSIONS

We can see that in general the transition from stationary to moving structures is smooth. For high forcing velocities, the pattern stays stationary but is modulated. As the velocity of forcing diminishes, the pattern starts to do a back and forth (waving) motion with very small amplitude (the distance that each stripe travels). As the forcing velocity slows even further, the amplitude of the traveling grows. At some point, the waving becomes asymmetric and the stripes move more in one direction (the direction of forcing) than the other. The modulation and waving of stripes is independent on transverse instabilities.

It is worth mentioning that the waving behavior is dependent on the interaction between neighboring stripes and is not seen in a system consisting on a single moving stripe where the Turing pattern can form [15].

How universal is this waving behavior? With these experimental conditions, in which the type of Turing structures corresponds to labyrinths, it seems to work for every wavelength used. However, when the forcing turns the pattern into a spot pattern, this back and forth motion is not observed [9,10]. Furthermore, when changing the wavelength of the forcing pattern to a higher value, a superposition of Turing hexagons and moving stripes is observed. (This behavior deserves special attention and will be treated elsewhere.)

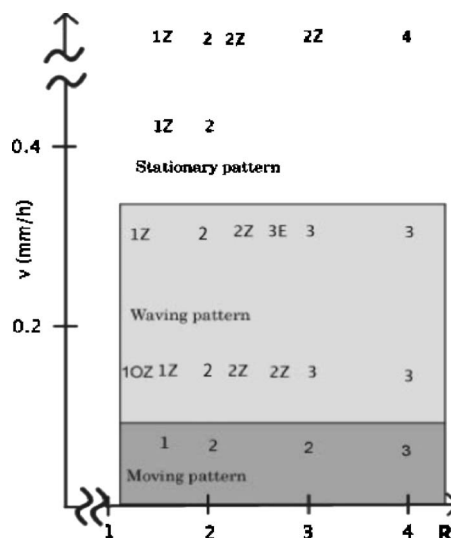


FIG. 5. Phase diagram. The number corresponds to the number of stripes that appear from each imposed stripe. Z corresponds to patterns that turn into zigzag patterns, while E corresponds to patterns that are distorted trough the Eckhaus instability. OZ corresponds to oscillating zigzag patterns.

The forcing velocity can alter the way the imposed stripes develop in order to attain the Turing wavelength. This is seen for relative forcing wavelength higher or equal to three times the Turing wavelength. This can be explained taking into account that with different width of forced stripes the splitting behavior changes [14]. For a relative wavelength of forcing of three times the natural wavelength, as the forcing stripe starts to move there are two things going on. On one hand, the stripe starts to split and one of the segments will move in the opposite direction of the forcing. On the other hand, the forcing band starts influencing a region in front of the stripe. These two events together generate a broad band that can split in three. The case of 4:1 forcing going to three stripes (instead of splitting twice) is easier to explain. The forcing bands are very broad and compensate for the wavelength mismatch that generates splitting in three.

ACKNOWLEDGMENTS

This work was supported by the MCyT (Spain) under Project No. FIS2004-03006 and Xunta de Galicia (Spain) under Project No. PGIDIT05PXIC20607PN.

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