Physics Letters A 179 (1993) 91–96
North-Holland

PHYSICS LETTERS A

Subcritical transitions to Turing structures

O. Jensen [1], V.O. Pannbacker [1], G. Dewel [2] and P. Borckmans [2]
Service de Chimie-Physique and Centre for Nonlinear Phenomena and Complex Systems, C.P. 231, Université Libre de Bruxelles, 1050 Brussels, Belgium

Received 4 March 1993; revised manuscript received 12 May 1993; accepted for publication 25 May 1993
Communicated by A.P. Fordy

The existence of localized structures is discussed within the framework of the pattern selection problem for a model for the chlorine dioxide–iodine–malonic acid reaction that represents a key to the understanding of the recently obtained Turing structures.

1. Introduction

Recent experiments [1,2] on the chlorite–iodide–malonic acid (CIMA) reaction have revealed the existence of stationary space periodic concentration patterns, so-called Turing structures [3], in open gel reactors. Lengyel and Epstein have suggested [4] that these patterns could arise because the iodine activator species forms a reversible complex of low mobility with the starch molecules used as color indicator for this reaction. They have also developed [5] a simple two-variable model that includes the three overall stoichiometric processes that lie at the heart of the mechanism of the CIMA reaction: the chlorine dioxide–iodine–malonic acid model.

The corresponding dimensionless reaction–diffusion equations allowing for the complex formation take the form

$$\sigma\frac{\partial u}{\partial t}=a-u-\frac{4uv}{1+u^2}+\nabla^2 u\,,$$

$$\frac{\partial v}{\partial t}=b\left(u-\frac{uv}{1+u^2}\right)+c\nabla^2 v\,, \qquad (1)$$

where u and v denote respectively the dimensionless iodide (I^-) and chlorite (ClO_2^-) concentrations; a and b are parameters related to feed concentrations of other reactants, whereas c represents the ratio of the molecular diffusion coefficients of the inhibitor (v) and activator (u): $c=D_{ClO_2^-}/D_{I^-}$, and σ, that is proportional to the starch concentration, accounts for the complex formation.

Linear stability analysis shows that for realistic values of the parameters, the unique homogeneous steady state (HSS), $u_s=a/5$ and $v_s=1+a^2/25$, may undergo a diffusion-driven instability, even when the diffusion coefficients of the activator and inhibitor are of the same order, if starch is present in sufficient amount. These results have been corroborated by numerical simulations of eq. (1) leading to Turing structures with a wavelength comparable to that observed experimentally [5]. Moreover steady concentration patterns have also been obtained in experiments using the chlorine dioxide–iodine–malonic acid–starch system for which the model described by eq. (1) has been specifically developed [6].

In this Letter the study of the pattern selection is pursued (see ref. [7], and references therein) as we report on preliminary results of this problem for the one- and two-dimensional (1D and 2D) chlorine dioxide–iodine–malonic acid system where subcritical bifurcations occur. It is also shown that the pinning of the front between the homogeneous steady state and a Turing pattern can give rise to a multiplicity of localized structures. The results we present were obtained by numerically integrating eq. (1) for $a=30$, $c=1.5$, $\sigma=8$ and by varying the control parameter b. In one dimension a semi-implicit Crank–

[1] Current address: Physics Laboratory III, Technical University of Denmark, DK-2800 Lyngby, Denmark.
[2] Senior Research Associates with the F.N.R.S. (Belgium).

0375-9601/93/$ 06.00 © 1993 Elsevier Science Publishers B.V. All rights reserved.

Nicolson method was used with no-flux boundary conditions. In two dimensions we respectively used an odd–even hopscotch method with periodic boundary conditions and an alternating direction implicit method with no-flux conditions.

2. One-dimensional systems

In one space dimension, on decreasing b, the HSS undergoes a *subcritical* bifurcation at $b=b_T$ giving rise to a spatially periodic state. As a result there is a region of bistability between the HSS and the Turing patterns (fig. 1). In absence of diffusion the HSS also undergoes a weakly *inverted* Hopf bifurcation at $b=b_H$ leading to oscillations of limit cycle type. When the starch concentration (i.e. σ) is sufficiently large we have $b_T > b_H$; the Turing threshold appears first and the stability domains of the patterns and the uniform temporal oscillations overlap. Because of this bistability and also the subcritical nature of its bifurcation, the Turing structures can exist over a wider range of parameter b than the so-called Turing space (i.e. $b_T \leqslant b \leqslant b_H$) deduced from the linear stability analysis of the HSS.

A large variety of inhomogeneous solutions may then be formed by spatial juxtaposition of these global states. We here describe only those that may appear in the subcritical region $b > b_T$.

The simplest consists of a front (kink) connecting a Turing pattern to the HHS (fig. 2a). Because in our simulations the hysteresis region is rather large this front is sharp and it can thus interact with the underlying Turing pattern leading to the pinning of the interface by the structure. As a result the front velocity is zero in a finite range of values of the parameter b ($p_{low} \leqslant b \leqslant p_{high}$ in fig. 1). Outside this *locking band* one observes a depinning transition; the structure invades the territory of the HSS for $b < p_{low}$ and the reverse situation occurs for $b > p_{high}$. In this process the velocity of the front is not uniform but executes jumps of one wavelength of the underlying structure (fig. 3). The time interval between successive jumps increases when the pinning band is approached from the outside (plateau behavior). This phenomenon of interface pinning results from non-

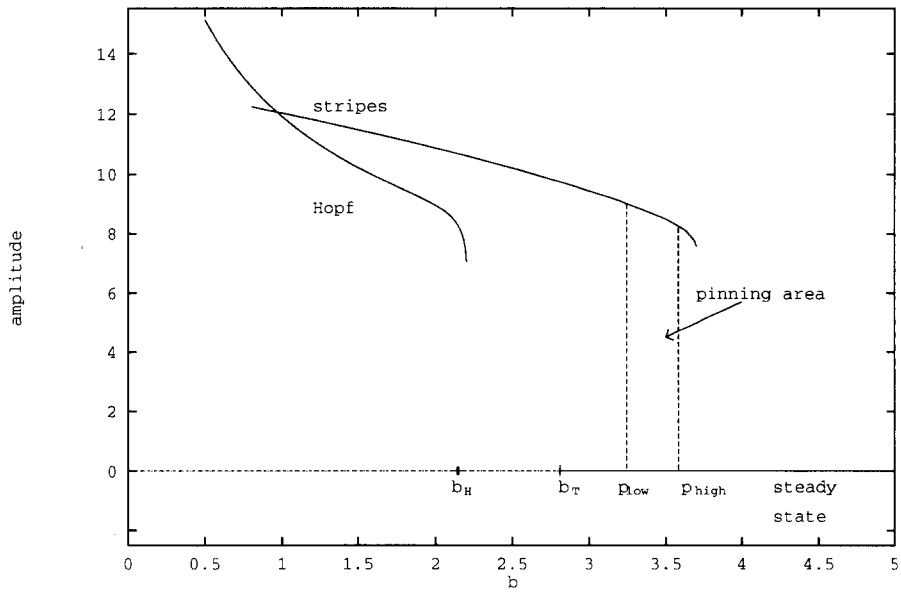


Fig. 1. Bifurcation diagram in one dimension obtained numerically for system (1) with $a=30$, $c=1.5$, $\sigma=8$. The peak-to-peak amplitude of u is represented as a function of parameter b. A Turing structure appears subcritically through an inverted pitchfork bifurcation at $b=b_T$. In the subcritical region where bistability occurs between the homogeneous steady reference state and the Turing structures, localized states may be found in the interval $p_{low} \leqslant b \leqslant p_{high}$. b_H represents the position of the Hopf bifurcation obtained from the linear stability analysis of the uniform reference state from which a branch of uniform time oscillations originates subcritically.

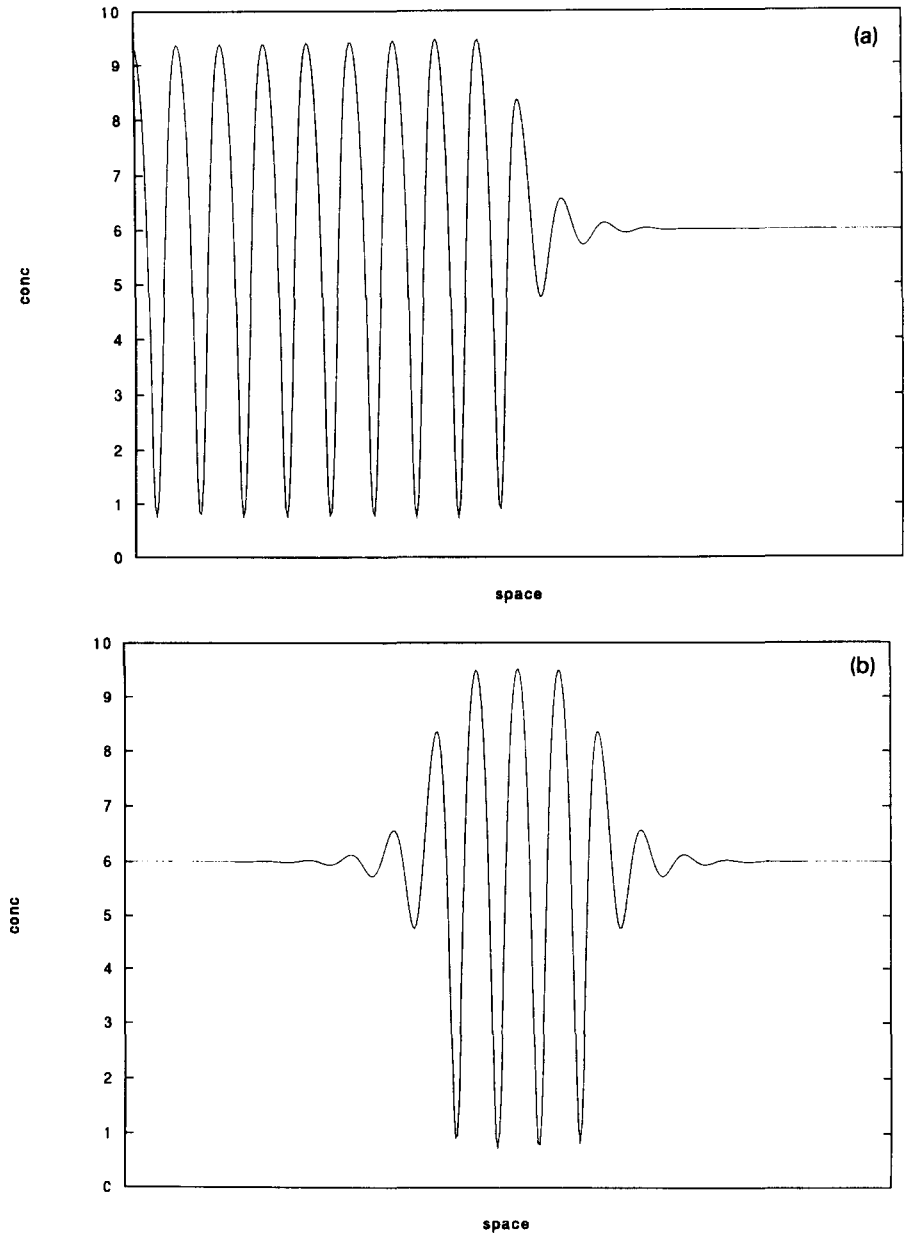


Fig. 2. (a) Pinned front of u between the homogeneous steady state and a 1D Turing structure for $b=3.4$ in the $p_{low} \leqslant b \leqslant p_{high}$ region. The other parameters are $a=30$, $c=1.5$, $\sigma=8$. (b) Stable (pinned) Turing droplet (concentration of u) localized in the background homogeneous steady state for $b=3.4$ in the $p_{low} \leqslant b \leqslant p_{high}$ region. All parameters ($a=30$, $c=1.5$, $\sigma=8$) have a uniform value over the whole system. In both cases no-flux conditions are taken at the boundaries.

adiabatic effects that are not contained in the standard amplitude equations of the weakly nonlinear theory [8]. Such interferences between a kink and a structure are frequent in condensed matter physics where they are known to dominate the dynamics of dislocations, charge density waves or domains (see references in ref. [9]). More recently pinning effects have been discussed in connection with convective

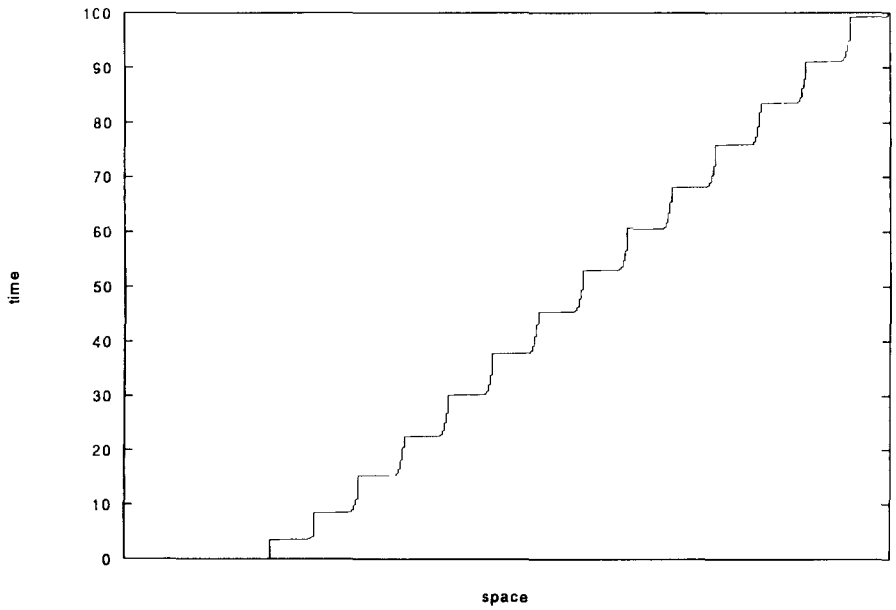


Fig. 3. Position of the front of u as in fig. 2a as a function of time when b is carried over ($b=3$) in the region $b_T \leqslant b \leqslant p_{low}$, outside the pinning band. The Turing structure is invading the domain of the homogeneous steady state with a nonuniform velocity.

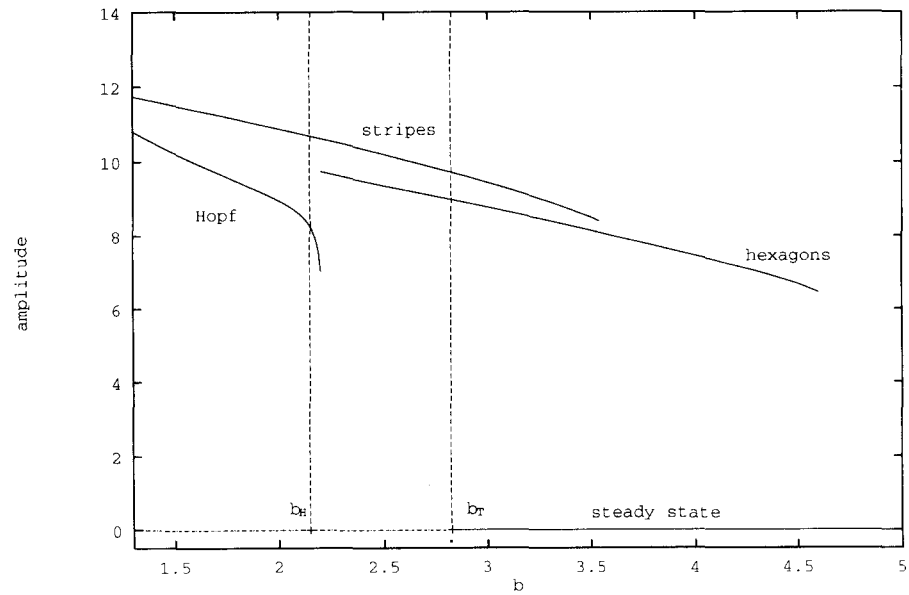


Fig. 4. Bifurcation diagram in two dimensions obtained numerically for system (1) with $a=30$, $c=1.5$, $\sigma=8$. The peak-to-peak amplitude of u is represented as a function of parameter b. Hexagons and stripes both appear subcritically respectively through a transcritical and inverted pitchfork bifurcation. A region exists where tristability occurs permitting many kinds of localized structures bringing couples of states into play and stabilized by pinning. A branch of uniform time oscillations originates, subcritically, from b_H.

instabilities in hydrodynamic systems [10]. A similar problem, the propagation failure in discrete arrays of bistable devices has also stimulated some interesting works [11–13].

These fronts then furnish the building blocks for the construction of coherent structures formed by *droplets* of one state embedded in the other. Our simulations have indeed produced localized Turing structures limited to a few wavelengths appearing in an otherwise uniform background (fig. 2b). Depending on the initial conditions, such localized states with different numbers of wavelengths have been obtained for the same values of the parameters. This multistability is obviously the consequence of the domain wall pinning effect. The width of the locking band is indeed the same for the various localized patterns as for the front. The complementary coherent structure where a bubble of the HSS appears in a Turing background has also been obtained.

3. Two-dimensional systems

In 2D, the first structure to appear subcritically has hexagonal symmetry where the maxima of concentration of the activator form a triangular lattice. It is followed, on decreasing b and also subcritically, by stripes. In this subcritical region the system may thus exhibit tristability between the triangles, the stripes and the HSS (fig. 4).

As a result a large variety of 2D localized structures are possible. The pinning of a structure takes place when the wavevectors characterizing the pattern are nearly perpendicular to the domain boundary with the HSS [14]. A hexagonal localized domain of hexagons embedded in the HSS is displayed in fig. 5a. Isolated patches of hexagons have also been obtained in the case of strong resonant forcing of oscillators [15] and in experiments on thermocapillary convection in the presence of localized heating [16].

In the region where they are both stable a band of stripes embedded in a hexagonal domain has also been observed (fig. 5b).

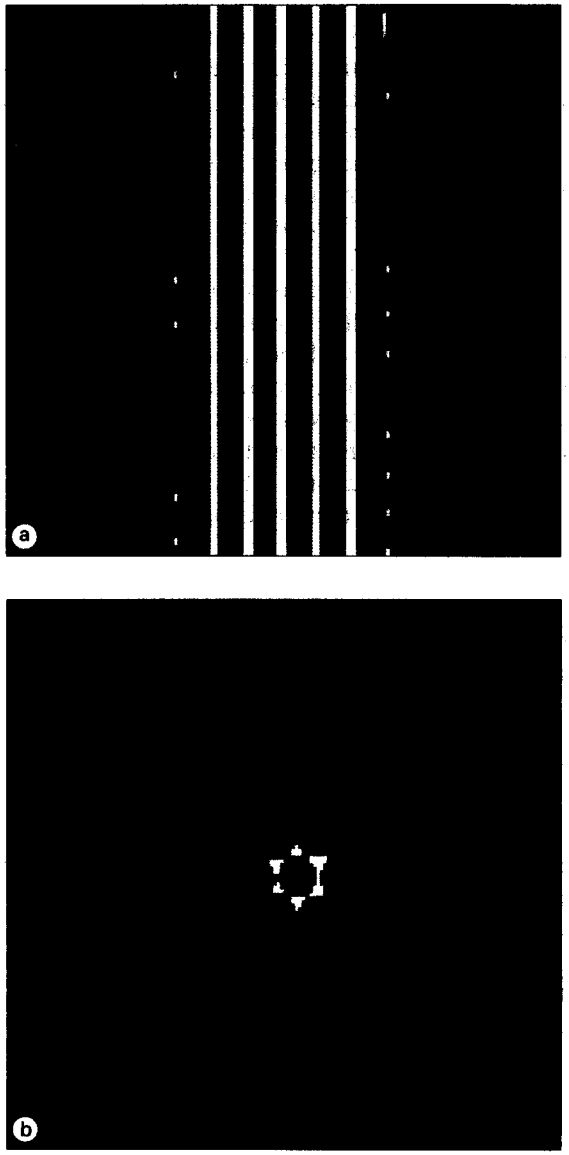


Fig. 5. (a) Localized 2D hexagonal pattern in the background of the homogeneous steady state ($a=30, c=1.5, \sigma=8, b=3.6$). (b) Localized 2D pattern where a droplet of stripes is embedded in the background of hexagons ($a=30, c=1.5, \sigma=8, b=3.6$). The grey scale correspond to the concentration of u between the absolute minimum (black) and maximum (white),

4. Conclusion

Until now experimental bifurcation diagrams [2,17,18] for chemical stationary patterns have failed

to exhibit subcritical solutions, even for patterns of hexagonal symmetry. Experimental evidence of some of the localized structures presented here would however point towards the existence of such a subcritical region. Localized structures, similar in nature, but where a stationary pattern is embedded in an oscillating background, when Turing and Hopf bifurcations interact, have on the other hand already been observed [19].

Let us also stress that the stabilization mechanism of the droplets studied here relies heavily on the intrinsic pinning effect of the fronts by the underlying structure. This is to be contrasted with other situations where stability owes its existence from the interaction between fronts mediated by so-called nonvariational contributions. It has indeed been shown that the absence of a minimizing potential function provides a stabilization mechanism for localized droplets [20].

Work is now in progress on the study of the growth mechanisms ("crystallisation") of such polygonal structures.

Acknowledgement

We would like to thank Professors G. Nicolis and E. Mosekilde for their interest in this work. Discussions with P. de Kepper, A. de Wit and S. Metens are acknowledged. This work was supported by the Erasmus Program B1028/13 of the EC and sponsored by Twinning contract no. SC1*-CT91-0706(TSTS) of the EC Science Program.

References

[1] V. Castets, E. Dulos, J. Boissonade and P. de Kepper, Phys. Rev. Lett. 64 (1990) 2953.
[2] Q. Ouyang and H.L. Swinney, Nature 352 (1991) 610.
[3] A. Turing, Philos. Trans. R. Soc. B 237 (1952) 37.
[4] I. Lengyel and I. Epstein, Proc. Natl. Acad. Sci. (USA) 89 (1992) 3977.
[5] I. Lengyel and I. Epstein, Science 251 (1991) 650.
[6] I. Epstein, I. Lengyel, S. Kadar, M. Kagan and M. Yokoyama, Physica A 188 (1992) 26.
[7] P. Borckmans, A. de Wit and G. Dewel, Physica A 188 (1992) 137.
[8] Y. Pomeau, Physica D 23 (1986) 3.
[9] M. Peyrard and M.D. Kruskal, Physica D 14 (1984) 88.
[10] D. Bensimon, B.I. Shraiman and V. Croquette, Phys. Rev. A 38 (1988) 5461.
[11] J. Keener, SIAM J. Appl. Math. 47 (1987) 556.
[12] J.P. Laplante and T. Erneux, Physica A 188 (1992) 89.
[13] H. Willebrand, C. Radehaus, F.-J. Niedernostheide, R. Dohmen and H.-G. Purwins, Phys. Lett. A 149 (1990) 131.
[14] B.A. Malomed, A.A. Nepomnyashchy and M.I. Tribelsky, Phys. Rev. A 42 (1990) 7244.
[15] P. Coullet and K. Emilsson, Physica A 188 (1992) 190.
[16] A.V. Gaponov-Grekhov, A.S. Lomov, G.V. Osipov and M.I. Rabinovich, in: Nonlinear waves 1: dynamics and evolution, eds. A.V. Gaponov-Grekhov, M.I. Rabinovich and J. Engelbrecht, Research reports in physics (Springer, Berlin, 1989) p. 65.
[17] P. de Kepper, V. Castets, E. Dulos and J. Boissonade, Physica D 49 (1991) 161.
[18] R.D. Vigil, Q. Ouyang and H. Swinney, Physica A 188 (1992) 17.
[19] J.J. Perraud, E. Dulos, P. de Kepper, A. de Wit, G. Dewel and P. Borckmans, in: Spatio-temporal organization in nonequilibrium systems, eds. S.C. Müller and T. Plesser (Projekt-Verlag, Dortmund, 1992) p. 205.
[20] O. Thual and S. Fauve, J. Phys. (Paris) 49 (1988) 1829.