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# Pattern selection in bistable systems

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**Abstract.** – Pattern selection in reaction-diffusion systems exhibiting bistability of homogeneous steady states is discussed. In agreement with recent experimental results, we obtain new bifurcation diagrams involving large-amplitude structures that arise from the coupling of the spatial critical modes with a quasi-neutral homogeneous mode.

An ever-increasing set of chemical and physical systems give rise to macroscopic patterns, dissipative structures, resulting from diffusive instabilities. They range from chemical reactors [1]-[3] (Turing structures) to electron-hole plasmas [4], semiconductor devices [5], gas discharges [6], materials irradiated by energetic particles or light [7], optics [8], ... Experiments recently revealed that the same sequence of structures occurs in these physically dissimilar systems when they exhibit bistability between homogeneous steady states (h.s.s.). [9]-[12]. We here analyze the formation and selection of these diffusion-driven patterns. It is indeed amazing to note that such studies for multistable systems are not very advanced although scores of works have been devoted to the properties of the fronts linking two h.s.s. that may also arise [4], [13]-[15]. We show that pattern selection is dominated by the coupling of the active spatial modes with a quasi-neutral homogeneous mode generated by the bifurcations inducing the bistability. We also discuss the existence of isolated branches of patterned solutions even when the h.s.s. are stable with respect to non-uniform perturbations.

We consider a general two-variable reaction-diffusion model characterized by the competition between an activator $u$ that stimulates its own variations and a controlling inhibitor $v$:

$$\begin{cases} \dfrac{\partial u}{\partial t} = f(u,v;a,\gamma) \; + \; \nabla^2 u \,, \\ \dfrac{\partial v}{\partial t} = \epsilon g(u,v;a,\gamma) \; + \; \delta \nabla^2 v \,, \end{cases} \tag{1}$$

where $f$ and $g$ describe the non-linear kinetics, $\delta = D_v/D_u$ is the ratio of the diffusion coefficients and $\epsilon = T_u/T_v$ that of the characteristic time scales of the two fields. In the following we consider cases where $\epsilon$ is such that a Hopf bifurcation never comes into play, in agreement with the experimental conditions in [9]-[12]. We distinguish two parameters $a$ and $\gamma$ that control the relative position and the number of intersections between the nullclines

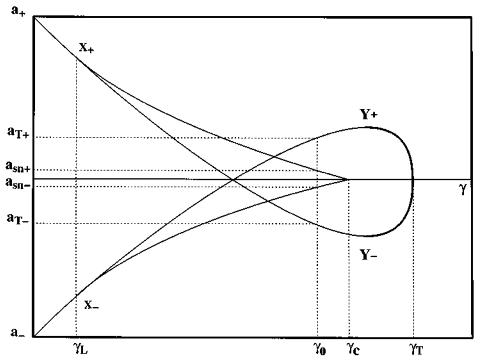


Fig. 1. – Schematic phase space $(a, \gamma)$ for the FitzHugh-Nagumo model (for fixed $\epsilon$ and $\delta$ in the absence of Hopf bifurcations). The locus of the homogeneous saddle-node bifurcations is represented by the curve $(a_+\gamma_c a_-)$ originating from the cusp $\gamma_c$. The locus of Turing instabilities is given by $(x_+\gamma_T x_-)$.

$(f = g = 0)$. When $a = a_c$ and $\gamma = \gamma_c$, the system exhibits a *critical point* (cusp). Around this point the model possesses either one or three h.s.s. Bistability emerges through two saddle-node bifurcations at $a = a_{sn\pm}$, creating a hysteresis loop based on the two states $(u_\pm, v_\pm)$ stable with respect to uniform perturbations. The intermediate unstable h.s.s. is denoted by $(u_0, v_0)$. From the bifurcation diagram of the homogeneous states, there results that parameter $\gamma$ is similar to the temperature and $a$ to the external field in the study of equilibrium phase transitions. The properties of the system for $a = 0$ were studied previously [16]. When the spatial processes are taken into account, diffusive instabilities may appear on the stable h.s.s. at $a = a_{T\pm}$, respectively on $u_+$ and $u_-$. This information is summarized in fig. 1, where, for given $\epsilon$ and $\delta$, we have represented the locus of the homogeneous saddle-node bifurcations $(a_+\gamma_c a_-)$ together with that of the diffusive instabilities $(x_+\gamma_T x_-)$ in the $(a, \gamma)$ parameter space. For a given $\gamma = \gamma_0$, these loci determine $a = a_{sn\pm}$ and $a = a_{T\pm}$.

We study the system in the vicinity of the critical point and essentially discuss the situation of nascent bistability. Our analysis relies on the fact that the homogeneous perturbations about the uniform states are quasi-neutral. As a consequence, a zero mode is active and must be taken into account in the dynamics since it will affect the amplitudes and the stability of the patterns. We therefore approximate the concentration fields $\mathbf{c} = \begin{pmatrix} u \\ v \end{pmatrix}$ by a linear superposition of these active modes:

$$\mathbf{c} = \mathbf{e}_1 A_0 + \left[\mathbf{e}_2 \sum_{i=1}^{m} A_i \exp[i\mathbf{q}_i \cdot \mathbf{r}] + \text{c.c.}\right], \tag{2}$$

where $(|\mathbf{q}_i| = q_c)$, $\mathbf{e}_1$ and $\mathbf{e}_2$ are the eigenvectors of the Jacobian matrix of the homogeneous and distributed systems. In 2D, for the case of the resonant patterns ($m = 3$ with $\mathbf{q}_1 + \mathbf{q}_2 + \mathbf{q}_3 = 0$), the reduction to a center unstable manifold [17] leads to coupled amplitude equations:

$$\begin{cases} \partial_t A_0 = h(A_0) + \alpha \sum_{i=1}^{3} |A_i|^2 - \beta_1 \sum_{i=1}^{3} |A_i|^2 A_0 - \\ \qquad - \beta_2 (A_1^* A_2^* A_3^* + A_1 A_2 A_3), \\ \partial_t A_1 = \mu A_1 + \alpha [A_0 A_1 + A_2^* A_3^*] - g_1 |A_1|^2 A_1 - \\ \qquad - g_2 [|A_2|^2 + |A_3|^2] A_1 - g_3 A_1 A_0^2 - g_4 A_0 A_2^* A_3^* \end{cases} \tag{3}$$

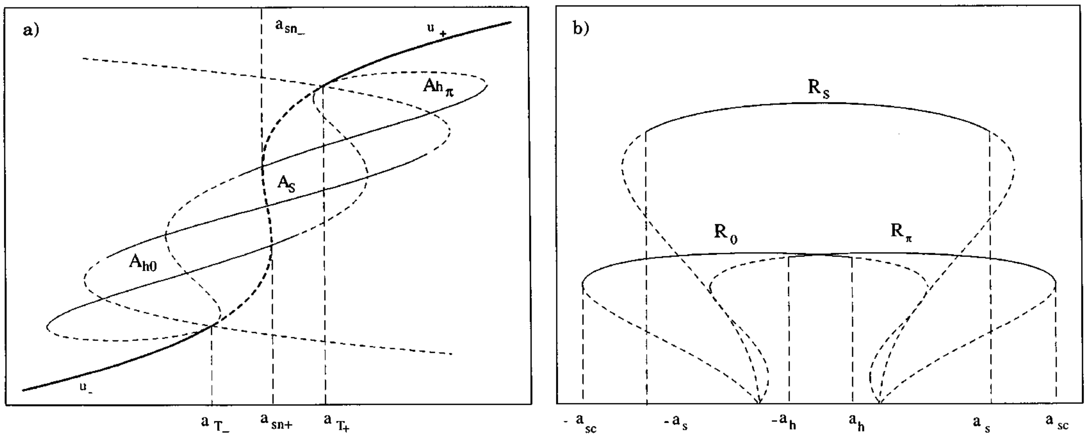


Fig. 2. – Bifurcation diagram in the bistable region for the FHN model with $a$ as parameter. *a*) Amplitudes of the homogeneous components of the stripe ($A_{\mathrm{s}}$) and hexagonal ($A_{h\pi}, A_{h_0}$) patterns. The homogeneous steady states are shown by the thicker lines. *b*) Moduli of the amplitudes of the corresponding patterned components of the same structures ($R_{\mathrm{s}}$: stripes, $R_0$ and $R_\pi$: hexagons). Stable states are in full lines. The $h_0$ and $h_\pi$ are, respectively, stable in the ranges $[-a_{\mathrm{sc}},\ a_h]$ and $[-a_h, a_{\mathrm{sc}}]$ with $a_h = (r/51)^{1/2}[74r^2/51 - q_{\mathrm{c}}^4]$ and $r = q_{\mathrm{c}}^4 - (\gamma - 1)$. The stripes are stable in the interval B $[-a_{\mathrm{s}}, a_{\mathrm{s}}]$, where $a_{\mathrm{s}} = (r/15)^{1/2}[2r^2/3 + q_{\mathrm{c}}^4]$.

and cyclic permutations (c.p.) of the subscripts for the equations of $A_2$ and $A_3$; we consider only the case of a cubic non-linearity saturation ($\beta_1,\ \beta_2,\ g_3,\ g_4$ are positive), $h(A_0)$ is a cubic function of $A_0$. The growth rate $\mu$ of the critical mode is computed from the linear stability analysis. The amplitude equations admit the following classes of solutions:

- Homogeneous solutions given by $h(A_0) = 0$ with $A_1 = A_2 = A_3 = 0$ corresponding to the reference states (h.s.s.).

- Patterned solutions for which the amplitudes of both types of modes are different from zero. They may exhibit either smectic ($A_0 \neq 0,\ |A_1| \neq 0,\ |A_2| = |A_3| = 0$ and c.p.) or hexagonal ($A_0 \neq 0,\ |A_1| = |A_2| = |A_3| \neq 0$) symmetries.

In the smectic case, eqs. (3) admit three solutions for the homogeneous component, $A_0$, represented by the inverted $S$ branch labelled $A_{\mathrm{s}}$ in fig. 2 *a*). Surprisingly, in contrast with the case of the h.s.s., it is only the intermediate part of the branch that is stable. The corresponding amplitudes of the inhomogeneous components $R_{\mathrm{s}} = |A_1|$ are given in fig. 2 *b*).

Solutions of hexagonal symmetry ($A_0 = A,\ A_i = R\exp[i\phi_i],\ i = 1\ ,2,\ 3$) are determined by the total phase $\Phi = \phi_1 + \phi_2 + \phi_3$ of the resonant modes. According to the sign of $v = g_4 A - \alpha$, the stable stationary value of $\Phi$ is zero ($v < 0$) or $\pi$ ($v > 0$). These values correspond respectively to a triangular ($h_0$) or a honeycomb ($h_\pi$) lattice. For each value of $\Phi$, we again find a triplet of solutions as shown in fig. 2 *a*) by the two inverted $S$ labelled $A_{h0}$ and $A_{h\pi}$ linking the two Turing instability points. Again the intermediate parts are the stable ones. The corresponding inhomogeneous components are also represented in fig. 2 *b*).

For the sake of concreteness, the pattern selection problem was analyzed on a FitzHugh-Nagumo–type model (FHN) defined by $f = u - u^3 - v,\ g = (\gamma u - v - a)$. Then $\gamma_{\mathrm{c}} = 1$, $a_{\mathrm{c}} = 0$ and $\alpha = 0$. The analytic bifurcation diagrams in 2D for the moduli of the homogeneous and the patterned modes are given in fig. 2 with the relative stability of the striped and

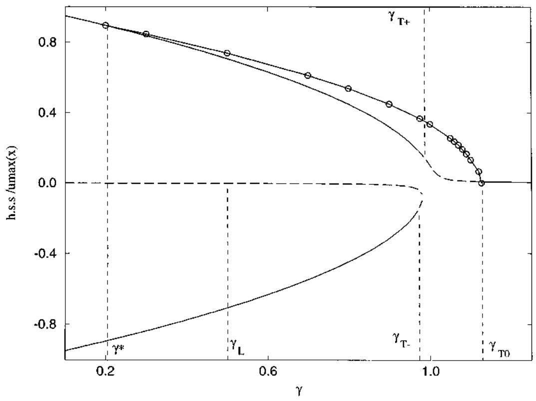


Fig. 3. – Bifurcation diagram, in 1D, with $\gamma$ as parameter for $a = 0.001$ ($\epsilon = 2, \delta = 4$). The homogeneous states exhibit an imperfect pitchfork bifurcation as $a \neq 0$. A branch ($T$) of stable stripes originates from the Turing instability at $\gamma_{\mathrm{T0}}$ and extends up to $\gamma^*$ where the maximum of the amplitude of the pattern, $u_{\max}$, becomes equal to the value of the homogeneous steady state. No stable patterns are generated by the Turing bifurcations at $\gamma_{\mathrm{T}\pm}$. $\gamma_{\mathrm{L}}$ is the limit of existence of Turing bifurcations on $u_+$ and $u_-$ (see fig. 1).

hexagonal patterns. These bifurcation diagrams are in excellent agreement with the results of our numerical integration of this FHN model. On varying the parameter $a$, one observes the sequence *h.s.s.* $\leftrightarrow$ *hexagons* 0 ($\pi$) $\leftrightarrow$ *stripes* $\leftrightarrow$ *hexagons* $\pi$ (0) $\leftrightarrow$ *h.s.s.* This succession is typical of bistable systems and is experimentally observed in a large diversity of systems [9]-[12].

The property distinguishing the behaviour of bistable systems, with respect to strictly monostable systems such as the Brusselator, arises from the effects of the homogeneous mode that acts globally on the dynamics through its resonant coupling (characterized by the coefficient $g_4$) with the critical modes. It ensures a smooth connection between the Turing bifurcation branches emerging from $a_{\mathrm{T}\pm}$ thereby giving rise to patterns whose amplitudes are of the order of the difference in amplitudes between $(u_+,\ v_+)$ and $(u_-,\ v_-)$. Such *large-amplitude* patterns cannot be described by applying the standard weakly non-linear analysis around $a_{\mathrm{T}+}$ or $a_{\mathrm{T}-}$. The $g_4$ pseudoquadratic coupling induces large subcriticality of the $h_0$ and $h_\pi$ hexagons respectively to the left of $a_{\mathrm{T}-}$ and to the right of $a_{\mathrm{T}+}$. The stripes also appear subcritically because of the presence of the cubic term proportional to $g_3$. As a result large domains of coexistence between hexagons, stripes and the h.s.s. can be observed. In these regions stable stationary *localized* structures [18] like hexagonal domains of $h_0$ or $h_\pi$ embedded in the uniform state can be obtained numerically. Spatial coexistence of structures of different symmetries is also possible, for instance both types of hexagons may coexist [16].

Surprisingly, we have also numerically obtained an *isolated* branch of stable patterns when no diffusive instability occurs on the h.s.s. $(u_\pm, v_\pm)$, *i.e.* when $\delta f_u \mid_\pm + g_v \mid_\pm < 0$. For the FHN model this relation requires $\gamma < \gamma_{\mathrm{L}} = \epsilon/\delta$. The origin of this branch can be apprehended if one considers the full bifurcation picture that extends in the two-parameter space $(a, \gamma)$ (fig. 1). Indeed the family of patterned solutions emanating from the diffusive bifurcations on the arc $Y_+\ \gamma_{\mathrm{T}}\ Y_-$ extends beyond $\gamma_{\mathrm{L}}$. This is exemplified in fig. 3 representing an amplitude bifurcation diagram with $\gamma$ as parameter when $Y_- < a < Y_+$. As $a \neq 0$, the reference h.s.s., at large $\gamma$, undergoes an imperfect pitchfork bifurcation. Of the three existing Turing instabilities at $\gamma_{\mathrm{T0}}$, $\gamma_{\mathrm{T}+}$ and $\gamma_{\mathrm{T}-}$, only $\gamma_{\mathrm{T0}}$ (belonging to arc $Y_+\ \gamma_{\mathrm{T}}\ Y_-$ ) gives rise to a branch $T$ of stable stripes (in 1D for simplicity) extending below $\gamma_{\mathrm{c}}$. Now, as one varies $a$, each Turing structure belonging to $T$ will generate, for every value of $\gamma (< \gamma_{\mathrm{L}})$, a branch of patterns disconnected from both $u_+$ and $u_-$ in fig. 2-type bifurcation diagrams as no Turing instability remains. Nevertheless the $T$ branch itself terminates at $\gamma^*(<\gamma_{\rm L})$, when the maximum ($a>0$) (respectively, minimum ($a<0$)) value of its amplitude becomes equal to that of the homogeneous steady state. Therefore for $\gamma<\gamma^*(a)$, $u_\pm$ are the only stable states to remain.

Such an isolated branch is reminiscent of the recent experimental observations in the ferrocyanide-iodate-sulfite chemical reaction [19]. There, labyrinthine-type patterns, similar to some of the structures generated by numerical simulations of the FHN model, have been obtained by local finite perturbations applied to an h.s.s. Their formation can be understood in terms of the morphological instability of fronts connecting the two h.s.s. [13], [20]. In the framework of non-local gradient systems, the representation of such a dynamics by an interface equation between domains of different composition has been proposed [21]. We have also often numerically observed situations where front instabilities come into play in the generation of hexagonal patterns. During a first stage, a flower-like pattern develops due to the alluded morphological instability. In a second stage, it then disorganizes slowly into a hexagonal pattern by breaking the flower petals. A similar scenario has been observed recently in a variant of the chlorite-iodide-malonic acid reaction in the presence of a high value of the concentration of the indicator [22].

In conclusion, in the presence of bistability, the pattern selection problem must take into account the resonant coupling of a homogeneous mode with the critical modes, in order to describe the sequence of large-amplitude structures that are observed in the experiments [9]-[12]. We have shown that pattern selection is controlled by the emergence of the stable intermediate branch of the homogeneous component of the patterned solutions. Similar results are obtained on the other side of, but near, the cusp when the system is monostable. There, however, the coexistence between the two types of hexagons disappears. When quadratic interactions are taken into account ($\alpha\neq 0$), the $a\to -a$ symmetry of fig. 1 is broken and other bifurcation behaviours such as restabilization of patterns of same symmetries will be possible as obtained previously in the generalized Swift-Hohenberg model [16] and a chemical system [23].

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## REFERENCES

[1] Turing A. M., *Philos. Trans. Soc. London B*, **237** (1952) 37.

[2] Castets V., Dulos E., Boissonade J. and De Kepper P., *Phys. Rev. Lett.*, **64** (1990) 2953.

[3] Kapral R. and Showalter K. (Editors), *Chemical Waves and Patterns* (Kluwer Academic Publishers, Dordrecht) 1994.

[4] Kerner B. S. and Osipov V. V., *Autosolitons* (Kluwer Academic Publishers, Dordrecht) 1994.

[5] Schöll E., *Nonequilibrium Phase Transitions in Semiconductors* (Springer-Verlag, Berlin) 1987.

[6] Radehaus C., Willebrand H., Dohmen R., Niedernostheide F.-J., Bengel G. and Purwins H.-G., *Phys. Rev. A*, **45** (1992) 2546.

[7] Emelyanov V. I., *Laser Physics*, **2** (1992) 389.

[8] LUGIATO L. (Editor), *Nonlinear Optical Structures, Patterns, Chaos*, special issue of *Chaos, Solitons Fractals*, **4**, No. 8-9 (1994).

[9] ACKEMANN T., LOGVIN YU. A., HEUERAND A. and LANGE W., *Phys. Rev. Lett.*, **75** (1995) 3450.

[10] BREAZEAL W., FLYNN K. M. and GWINN E. G., *Phys. Rev. E*, **52** (1995) 1503.

[11] FIRTH W. J., SCROGGIE A. J. and MCDONALD G. S., *Phys. Rev. A*, **46** (1992) 3609.

[12] AHLERS G., BERGE L. I. and CANNELL D. S., *Phys. Rev. Lett.*, **70** (1993) 2399.

[13] KURAMOTO Y., *Chemical Oscillations, Waves and Turbulence* (Springer-Verlag, Berlin) 1984.

[14] HORVATH D., PETROV V., SCOTT S. K. and SHOWALTER K., *J. Chem. Phys.*, **98** (1993) 6332.

[15] PISMEN L., *J. Chem. Phys.*, **71** (1979) 462.

[16] DEWEL G., MÉTENS S., HILALI M'F., BORCKMANS P. and PRICE C. B., *Phys. Rev. Lett.*, **74** (1995) 4647.

[17] ROBERTS A. J., *SIAM J. Math. Anal.*, **16** (1985) 1243.

[18] HILALI M'F., MÉTENS S., BORCKMANS P. and DEWEL G., *Phys. Rev. E*, **51** (1995) 2046.

[19] LEE K., MCCORMICK W. D., OUYANG Q. and SWINNEY H. L., *Science*, **261** (1993) 192.

[20] HAGBERG A. and MERON E., *Phys. Rev. Lett.*, **72** (1994) 2494.

[21] PETRICH D. M. and GOLDSTEIN R. E., *Phys. Rev. Lett.*, **72** (1994) 1120.

[22] DE KEPPER P., private communication.

[23] DUFIET V. and BOISSONADE J., *J. Chem. Phys.*, **96** (1991) 664.