EUROPHYSICS LETTERS 1 July 1998

Europhys. Lett., **43** (1), pp. 35-40 (1998)

Phase instabilities in hexagonal patterns(*)

B. Echebarria and C. Pérez-García

Departamento de Física y Matemática Aplicada, Facultad de Ciencias
Universidad de Navarra - E-31080 Pamplona, Navarra, Spain

(received 6 October 1997; accepted in final form 20 May 1998)

PACS. 47.54+r – Pattern selection; pattern formation.
PACS. 47.20Ky – Nonlinearity (including bifurcation theory).
PACS. 42.65Sf – Dynamics of nonlinear optical systems; optical instabilities, optical chaos and complexity, and optical spatio-temporal dynamics.

Abstract. – The general form of the amplitude equations for a hexagonal pattern including spatial terms is discussed. At the lowest order we obtain the *phase equation* for such patterns. The general expression of the diffusion coefficients is given and the contributions of the new spatial terms are analysed in this paper. From these coefficients the phase stability regions in a hexagonal pattern are determined. In the case of Bénard-Marangoni instability our results agree qualitatively with numerical simulations performed recently.

Several systems out of equilibrium exhibit hexagonal patterns. Historically, the cellular patterns reported by Bénard almost a century ago were the first nonequilibrium system showing this planform [1]. More recently, hexagonal patterns were obtained in front solidification [2], in Rayleigh-Bénard convection with non-Boussinesquian effects [3], in Faraday crispation [4], in a nonlinear Kerr medium [5], in a liquid-crystal valve device [6], in chemical Turing patterns [7], in ferrofluids [8] and in vibrating granular layers [9]. Although the physical mechanism responsible for these patterns is different in each system, they can be described within a common framework. A hexagonal pattern can be seen as the superposition of three systems of rolls at $2\pi/3$ rad, so that the resonance condition $\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3 = 0$ is satisfied. The main aim of this paper is to discuss the form of the evolution equations of the amplitudes of those three modes, *i.e.* the so-called *amplitude equations*, as well as the most general linear *phase equation* for a hexagonal pattern and the corresponding stability regions.

From symmetry arguments one can deduce [10] that the normal form for the amplitude of the modes forming the hexagonal pattern is

$$\partial_t A_1 = \epsilon A_1 + \alpha_0 \bar{A}_2 \bar{A}_3 - \gamma(|A_2|^2 + |A_3|^2)A_1 - |A_1|^2 A_1 \,. \quad (1)$$

(The overbar denotes complex conjugation. The equations for A_2 and A_3 are obtained by rotating the subindices.) Spatial variations can be included following the Newell-Whitehead technique (see ref. [11]). Up to the third order in the amplitudes the linear spatial variations of

(*) Dedicated to Prof. J. Casas-Vázquez on the occasion of his 60th birthday.

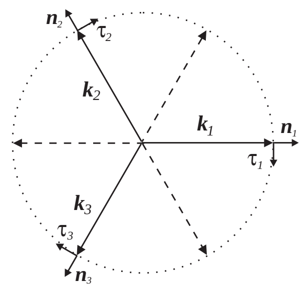

Fig. 1. – Unitary vectors: $\mathbf{n}_i$ parallel and $\boldsymbol{\tau}_i$ perpendicular to the wave numbers in a hexagonal pattern.

each system of rolls is in the form $(\hat{\mathbf{n}}_1 \cdot \nabla)^2 A_1$, a term that must be added to eq. (1). Here $\hat{\mathbf{n}}_1$ indicates a unitary vector in the direction of the first system of rolls. Until recently, as in the case of a pattern of rolls, only this term was considered. Brand [12] discussed the possibility of including terms in the form $(A\nabla A)$. Considering the hexagonal symmetries (reflections on X-axis and Y-axis, and $2\pi/3$ rad rotations) this author deduced that a term in the form

$$i\beta_1[\bar{A}_3(\hat{\mathbf{n}}_2 \cdot \nabla)\bar{A}_2 + \bar{A}_2(\hat{\mathbf{n}}_3 \cdot \nabla)\bar{A}_3] \tag{2}$$

(β_1 is a real coefficient) could also be added to eq. (1). However, as noticed by several authors [13, 14], in the same order another term can appear, namely

$$i\beta_2[\bar{A}_3(\hat{\mathbf{n}}_3 \cdot \nabla)\bar{A}_2 + \bar{A}_2(\hat{\mathbf{n}}_2 \cdot \nabla)\bar{A}_3] \tag{3}$$

(again β_2 is a real coefficient). It can easily be seen that this term also remains invariant under the hexagonal group transformations [15].

Then, for perturbations up to the third order a generalized amplitude equation that accounts for spatial variations in a hexagonal pattern is

$$\begin{aligned}\partial_t A_1 = {} & \epsilon A_1 + (\hat{\mathbf{n}}_1 \cdot \nabla)^2 A_1 + \alpha_0 \bar{A}_2 \bar{A}_3 + \\ & + i\beta_1[\bar{A}_3(\hat{\mathbf{n}}_2 \cdot \nabla)\bar{A}_2 + \bar{A}_2(\hat{\mathbf{n}}_3 \cdot \nabla)\bar{A}_3] + i\beta_2[\bar{A}_3(\hat{\mathbf{n}}_3 \cdot \nabla)\bar{A}_2 + \bar{A}_2(\hat{\mathbf{n}}_2 \cdot \nabla)\bar{A}_3] - \\ & - \gamma(|A_2|^2 + |A_3|^2)A_1 - |A_1|^2 A_1 \,.\end{aligned} \tag{4}$$

The gradient terms in eqs. (2) and (3) are a consequence of quadratic resonances and therefore their influence is essentially two-dimensional. (It should be noticed that, at the same order, terms in the form $|A|^2\nabla A$ could be included, but for the sake of simplicity we will not consider them in this paper). To gain some physical insight into the problem, it is useful to express the derivatives in eq. (3) in terms of the unitary vectors of the corresponding mode, *i.e.* $\hat{\mathbf{n}}_2 = -\frac{1}{2}\hat{\mathbf{n}}_3 + \frac{\sqrt{3}}{2}\hat{\boldsymbol{\tau}}_3$ in the first term and $\hat{\mathbf{n}}_3 = -\frac{1}{2}\hat{\mathbf{n}}_2 - \frac{\sqrt{3}}{2}\hat{\boldsymbol{\tau}}_2$ in the second, where $\hat{\boldsymbol{\tau}}_i$ stands for the unitary vectors perpendicular to the direction of the wave number of the corresponding system of rolls (see fig. 1). Notice that the gradient terms (eqs. (2) and (3)) can be added to give

$$i\alpha_1[\bar{A}_3(\hat{\mathbf{n}}_2 \cdot \nabla)\bar{A}_2 + \bar{A}_2(\hat{\mathbf{n}}_3 \cdot \nabla)\bar{A}_3] + i\alpha_2[\bar{A}_2(\hat{\boldsymbol{\tau}}_3 \cdot \nabla)\bar{A}_3 - \bar{A}_3(\hat{\boldsymbol{\tau}}_2 \cdot \nabla)\bar{A}_2] \tag{5}$$

with $\alpha_1 = \beta_1 - \frac{1}{2}\beta_2$ and $\alpha_2 = \frac{\sqrt{3}}{2}\beta_2$. Terms in this form have been discussed by Gunaratne *et al.* [13] for chemical reactions, Kuznetsov *et al.* [14] for Rayleigh-Bénard convection and Bragard and Golovin *et al.* [16] for Bénard-Marangoni convection. A term with the coefficient α_1 accounts for distortions in the direction of the rolls and it therefore corresponds to *dilatations* of hexagons (it slightly changes the volume in Fourier space), while the terms with α_2 account

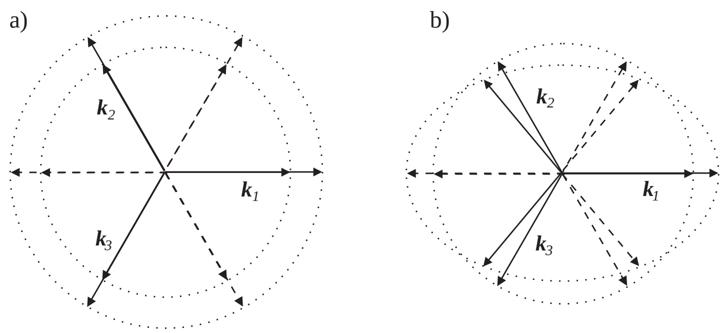

Fig. 2. – a) Dilatations and b) distortions of a hexagonal pattern.

for *distortions* of the hexagonal form. In fig. 2 we represent the action of these two terms in Fourier space. As discussed by Kuznetsov *et al.* [14], α_0 and α_1 vanish when the bifurcation that leads to the hexagonal pattern is supercritical, while in the subcritical case the three coefficients α are, in general, different from zero. The term with α_1 will stabilize patterns with $|k| \neq |k_c|$ while the term with α_2 would stabilize nonequilateral hexagons.

With the transformation in eq. (5), eq. (4) becomes

$$\begin{aligned}\partial_t A_1 = {}& \epsilon A_1 + (\hat{\mathbf{n}}_1 \cdot \nabla)^2 A_1 + \alpha_0 \bar{A}_2 \bar{A}_3 + \\ & + i\alpha_1[\bar{A}_3(\hat{\mathbf{n}}_2 \cdot \nabla)\bar{A}_2 + \bar{A}_2(\hat{\mathbf{n}}_3 \cdot \nabla)\bar{A}_3] + i\alpha_2[\bar{A}_2(\hat{\boldsymbol{\tau}}_3 \cdot \nabla)\bar{A}_3 - \bar{A}_3(\hat{\boldsymbol{\tau}}_2 \cdot \nabla)\bar{A}_2] - \\ & - \gamma(|A_2|^2 + |A_3|^2)A_1 - |A_1|^2 A_1 \,. \end{aligned} \tag{6}$$

Now let us consider solutions with a wave number k slightly different from k_c, *i.e.* $A_i = \hat{A}_i e^{i\mathbf{q}_i \cdot \mathbf{r}}$ with $\mathbf{q}_i = \mathbf{k}_i - \mathbf{k}_c$. We are interested in homogeneous and stationary solutions of the last equation in the form of hexagons $\hat{A}_1 = \hat{A}_2 = \hat{A}_3 = H \neq 0$ for which the amplitude must be

$$H = \frac{(\alpha_0 + 2q\alpha_1) + \sqrt{(\alpha_0 + 2q\alpha_1)^2 + 4(\epsilon - q^2)(1 + 2\gamma)}}{2(1 + 2\gamma)} \,. \tag{7}$$

The stability of this solution is determined considering perturbations in the form $A_i = He^{iq_i \cdot x_i}(1 + r_i + i\phi_i)$, where r_i is the amplitude and ϕ_i is the phase of the perturbation. After introducing these perturbations in eq. (6) and linearizing, one arrives at the following system of equations:

$$\begin{aligned}\partial_t r_1 = {}& \partial_1^2 r_1 - 2q\partial_1\phi_1 + (\alpha_0 + 2q\alpha_1)H(r_2 + r_3 - r_1) + H\left(\alpha_1 + \frac{\alpha_2}{\sqrt{3}}\right)(\partial_2\phi_2 + \partial_3\phi_3) + \\ & + \alpha_2 H(\partial_3\phi_2 + \partial_3\phi_3) - 2H^2 r_1 - 2\gamma H^2(r_2 + r_3) \,, \end{aligned} \tag{8}$$

$$\begin{aligned}\partial_t \phi_1 = {}& 2q\partial_1 r_1 + \partial_1^2\phi_1 - (\alpha_0 + 2q\alpha_1)H(\phi_1 + \phi_2 + \phi_3) + H\left(\alpha_1 + \frac{\alpha_2}{\sqrt{3}}\right)(\partial_2 r_2 + \partial_3 r_3) + \\ & + \frac{2}{\sqrt{3}}\alpha_2 H(\partial_2 r_3 + \partial_3 r_2) \,, \end{aligned} \tag{9}$$

where we have used the following notation: $\partial_i = \hat{\mathbf{n}}_i \cdot \nabla$. We assume that the amplitudes r_i and the total phase $\Phi = \phi_1 + \phi_2 + \phi_3$ are fastly decaying variables and therefore they can be eliminated adiabatically. As a result, the dynamics is dominated by two of the phases. Instead of using ϕ_2 and ϕ_3 one can take a vector $\vec{\phi} = [-(\phi_2 + \phi_3), \frac{1}{\sqrt{3}}(\phi_2 - \phi_3)]$ and the resulting equation will have the most general form of a linear diffusion equation in 2D,

$$\partial_t \vec{\phi} = D_\perp \nabla^2 \vec{\phi} + (D_\parallel - D_\perp)\nabla(\nabla \cdot \vec{\phi}) \,. \tag{10}$$

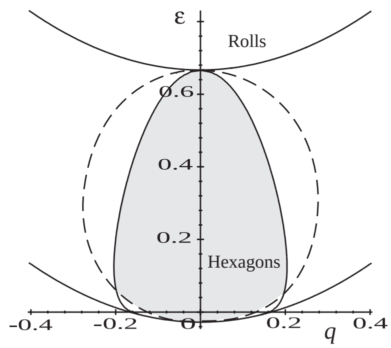

Fig. 3. – Stability region of hexagonal cells with $\alpha_1 = \alpha_2 = 0$. Notice that this region is symmetrical with respect to the vertical axis. The phase instabilities are represented by a solid ($D_\parallel = 0$) and a dashed line ($D_\perp = 0$).

This is the linear *phase equation* of a pattern of hexagons. The form of the coefficients in this equation has been chosen by analogy with that in the wave equation in an elastic solid [17]. (The velocity of the transversal waves c_t corresponds here to $D_\perp$, while the velocity for the longitudinal waves c_l is replaced by $D_\parallel$.) Using this analogy, we split the phase $\vec{\phi}$ into a longitudinal part $\vec{\phi}_\mathrm{l}$ and a transversal $\vec{\phi}_\mathrm{t}$, that satisfy $\nabla \times \vec{\phi}_\mathrm{l} = 0$ (rhomboidal phase perturbations) and $\nabla \cdot \vec{\phi}_\mathrm{t} = 0$ (rectangular phase perturbations). It can be proved straightforwardly that these components satisfy

$$\partial_\mathrm{t} \vec{\phi}_\mathrm{l} = D_\parallel \nabla^2 \vec{\phi}_\mathrm{l}, \qquad \partial_\mathrm{t} \vec{\phi}_\mathrm{t} = D_\perp \nabla^2 \vec{\phi}_\mathrm{t}. \tag{11}$$

A linear stability analysis of these equations shows that the system is stable to phase perturbations provided that $D_\perp > 0$ and $D_\parallel > 0$. After tedious calculations one arrives to a general expression of these coefficients:

$$D_\perp = \frac{1}{4} - \frac{q^2}{2u} + \frac{H^2}{8u}(\alpha_1 - \sqrt{3}\alpha_2)^2\,, \tag{12}$$

$$D_\parallel = \frac{3}{4} - \frac{q^2(4u+v)}{2uv} + \frac{H^2}{8u}(\alpha_1 - \sqrt{3}\alpha_2)^2 - \frac{H^2\alpha_1}{v}(\alpha_1 + \sqrt{3}\alpha_2) + + \frac{Hq}{v}(3\alpha_1 + \sqrt{3}\alpha_2) \tag{13}$$

with the relationships

$$u = H^2(1-\gamma) + (\alpha_0 + 2\alpha_1 q)H > 0\,, \tag{14}$$

$$v = 2H^2(1+2\gamma) - (\alpha_0 + 2\alpha_1 q)H > 0\,. \tag{15}$$

The curves $D_\perp = 0$ and $D_\parallel = 0$ determine the stability of a regular hexagonal pattern to rhomboid and rectangular phase perturbations, respectively, while $u = 0$ determines the region where the hexagons are unstable to amplitude perturbations. Let us mention that we have assumed that the homogeneous stationary solution H is positive, so $\alpha_0 + 2q\alpha_1 > 0$. Otherwise, the hexagons become unstable to a global phase change from 0 to π (up-hexagons to down-hexagons). It is interesting to examine the influence of the different parameters in phase stability. We first consider the particular case in which the nonlinear spatial terms are absent ($\alpha_1 = \alpha_2 = 0$). This case has been considered by several authors [18–20]. The results are given in fig. 3 in a (q, ϵ) representation, where the shaded area corresponds to the stability region of the hexagonal pattern. Notice that due to the symmetry $q \to -q$ in the diffusion coefficients

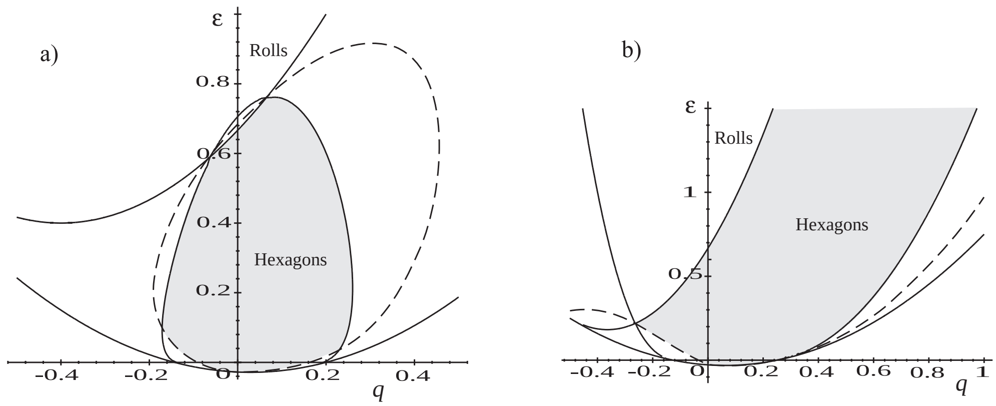

Fig. 4. – Stability region of a hexagonal pattern when all spatial terms are included. a) Closed case ($\alpha_0 = 1, \alpha_1 = \alpha_2 = 0.5, \gamma = 4$). b) Open case ($\alpha_0 = 1, \alpha_1 = 1, \alpha_2 = 2.5, \gamma = 4$).

and in u and v this figure is symmetrical with respect to the vertical axis. Near threshold rhomboidal phase perturbations destabilize the pattern ($D_\parallel = 0$), but for higher supercritical conditions the pattern becomes unstable by rectangular phase perturbations ($D_\perp = 0$). These two curves intersect at the values of q that correspond to the conditions $D_\perp = D_\parallel = 0$, *i.e.* $q = \pm\frac{\alpha_0}{2\gamma}\sqrt{(1+\gamma)/2}$. (For $(\alpha_1, \alpha_2) \neq 0$ these points are quantitatively but not qualitatively modified.) Both curves are tangent at $q = 0$ with the upper amplitude stability curve $u = 0$.

From eqs. (13)-(15) one can deduce the following symmetry properties:

$$H(\alpha_1, q) = H(-\alpha_1, -q); \quad u(\alpha_1, q) = u(-\alpha_1, -q); \quad v(\alpha_1, q) = v(-\alpha_1, -q)\,, \qquad (16)$$
$$D_\parallel(\alpha_1, \alpha_2, q) = D_\parallel(-\alpha_1, -\alpha_2, -q); \quad D_\perp(\alpha_1, \alpha_2, q) = D_\perp(-\alpha_1, -\alpha_2, -q).$$

These symmetry expressions imply that the stability curves for a particular value of (α_1, α_2) become reflected in a (q, ϵ) representation with respect to the vertical axis under the transformation $\alpha_1 \to -\alpha_1$, $\alpha_2 \to -\alpha_2$.

The stability curves are qualitatively modified when the gradient terms are present. In general, for $(\alpha_1, \alpha_2) \neq 0$ the stability regions are no longer symmetrical with respect to the ϵ-axis. An example of this situation is given in fig. 4a). Several cases are possible. We see that for $(\alpha_1, \alpha_2) > 0$ the phase instability curves ($D_\parallel = 0, D_\perp = 0$) are decentered to the right, while the minimum of amplitude instability curve $u = 0$ is decentered to the left in the (q, ϵ) plane. The phase instability and the amplitude instability curves are tangent at $q = 0$ when the two conditions $D_\perp = 0$ and $u = 0$ are met simultaneously. This leads to the condition $\alpha_1 = \sqrt{3}\alpha_2$. (Notice that the two phase instability curves are tangent at the same point). But in many cases the phase and amplitude instability curves intersect at two points, namely

$$q = -\frac{\alpha_0(\alpha_1 - \sqrt{3}\alpha_2)}{2[\alpha_1(\alpha_1 - \sqrt{3}\alpha_2)) \pm (\gamma - 1)]}\,. \qquad (17)$$

However, when the following condition $|\alpha_1(\alpha_1 - \sqrt{3}\alpha_2)| \geq (\gamma - 1)$ is satisfied the curve $D_\perp = 0$ is not closed and the phase and amplitude instability curves do not intersect themselves in the $q \geq 0$ quadrant. (Such a situation is represented in fig. 4b).)

When convection is due to surface-tension variations with the temperature (Bénard-Marangoni (BM) convection) the condition $\alpha_1 > 0$ is satisfied [21]. Two examples of the the phase stability region for $(\alpha_1, \alpha_2) \geq 0$ are given in fig. 3. We notice that the stability regions in figs. 3a) and 3b) are in qualitative agreement with the numerical results obtained by

Bestehorn [22] for the BM problem. We shall mention that in experiments performed by Koschmieder [23] the number of cells increases (q increases) with the supercritical heating in BM convection. The corresponding values are fitted quite well with the line of maximal growth rate obtained numerically by Bestehorn [22]. (This line remains inside the phase stability region.) This asymmetry could explain why in that system the transition between hexagons and rolls is not observed near threshold.

From the general form of the amplitude equations we derived the *phase equation* for a hexagonal pattern which is formally similar to the wave equation in an elastic solid. The expressions of the coefficients in this equation allow to determine the stability diagram. Unfortunately, experimental results on these phase instabilities are not available yet. We hope that the present results will suggest new experiments to study the wave number selection mechanisms in hexagonal patterns in different physical systems.

***

Fruitful discussions with R. Hoyle (Cambridge), M. Bestehorn (Stuttgart, Cottbus), A. G. Golovin and A. E. Nuz (Haifa) are gratefully acknowledged. This work was partially financed by the DGICYT (Spanish Government) under grants PB95-0578 and by the PIUNA (Universidad de Navarra). One of us (BE) acknowledges the Basque Government for a fellowship (BFI95.035).

REFERENCES

[1] Bénard H., *Rev. Gen. Sci. Pure Appl.*, **11** (1900) 1261.
[2] Wollkind D. J., Sriranganathan R. and Oulton D. B., *Physica D*, **12** (1984) 215.
[3] Ciliberto S., Pampaloni E. and Pérez-García C., *Phys. Rev. Lett.*, **61** (1988) 1198.
[4] Edwards W. S. and Fauve S., *J. Fluid Mech.*, **278** (1994) 123.
[5] D'Alessandro G. and Firth W. J., *Phys. Rev. Lett.*, **66** (1992) 2597.
[6] Pampaloni E., Ramazza P. L., Residori S. and Arecchi F. T., *Europhys. Lett.*, **24** (1993) 587.
[7] de Kepper P., Castets V., Dulos E. and Boissonade J., *Physica D*, **49** (1991) 161; Ouyang Q. and Swinney H. L., *Chaos*, **1** (1991) 411.
[8] Gailitis A., *J. Fluid Mech.*, **82** (1987) 401; Abou B. and Wesfreid J. E., in preparation.
[9] Melo F., Umbanhoward P. and Swinney H. L., *Phys. Rev. Lett.*, **95** (1995) 3838.
[10] Golubitsky M., Swift J. W. and Knobloch E., *Physica D*, **10** (1984) 249.
[11] Newell A. C., Passot T. and Lega J., *Ann. Rev. Fluid Mech.*, **25** (1993) 399.
[12] Brand H., *Prog. Theor. Phys. Suppl.*, **99** (1989) 442.
[13] Gunaratne G., Ouyang Q. and Swinney H. L., *Phys. Rev. E*, **50** (1994) 2802.
[14] Kuznetsov E. A., Nepomnyashchy A. A. and Pismen L. M., *Phys. Lett. A*, **205** (1995) 261.
[15] As discussed in ref. [13], terms in the form $i(\bar{A}_{i+1}\nabla^2\bar{A}_{i+1})/2q_c$ and $i(\nabla\bar{A}_{i+1})\cdot(\nabla\bar{A}_{i+2})/q_c$ must be added to eqs. (2) and (3) to render the resulting equation rotationally invariant. As we restrict our analysis up to third order in the amplitude we will not consider these kinds of terms.
[16] Bragard J., PhD Thesis, Universidad Complutense de Madrid (1996), unpublished; Golovin A. A., Nepomnyashchy A. A. and Pismen L. M., *J. Fluid Mech.*, **25** (1997) 317.
[17] Landau L. D. and Lifshitz E. M., *Teoría de la Elasticidad* (Edit. Reverté, Barcelona) 1969.
[18] Lauzeral J., Metens S. and Walgraef D., *Europhys. Lett.*, **24** (1993) 707.
[19] Hoyle R., *Appl. Math. Lett.*, **9** (1995) 81.
[20] Sushchik M. M. and Tsimring L. S., *Physica D*, **74** (1994) 90.
[21] Echebarria B. and Pérez-García C., in preparation.
[22] Bestehorn M., *Phys. Rev. E*, **48** (1993) 3622.
[23] Koschmieder L. and Switzer D. W., *J. Fluid Mech.*, **240** (1992) 533.