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Research Workshop on Condensed Matter Physics
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MINIWORKSHOP ON
PATTERN FORMATION AND SPATIO-TEMPORAL CHAOS
28 JULY - 8 AUGUST 1997

"Pattern selection and spatiotemporal dynamics in reaction - diffusion systems" Part I

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These are preliminary lecture notes, intended only for distribution to participants.

A / Introduction - experiments

B / Pattern selection in 2D reaction - diffusion systems

① Chemical reaction - diffusion mode[l]

② amplitude equation - perturbation expansion
- stripes
- rhombs
- hexagons

③ bifurcation diagrams

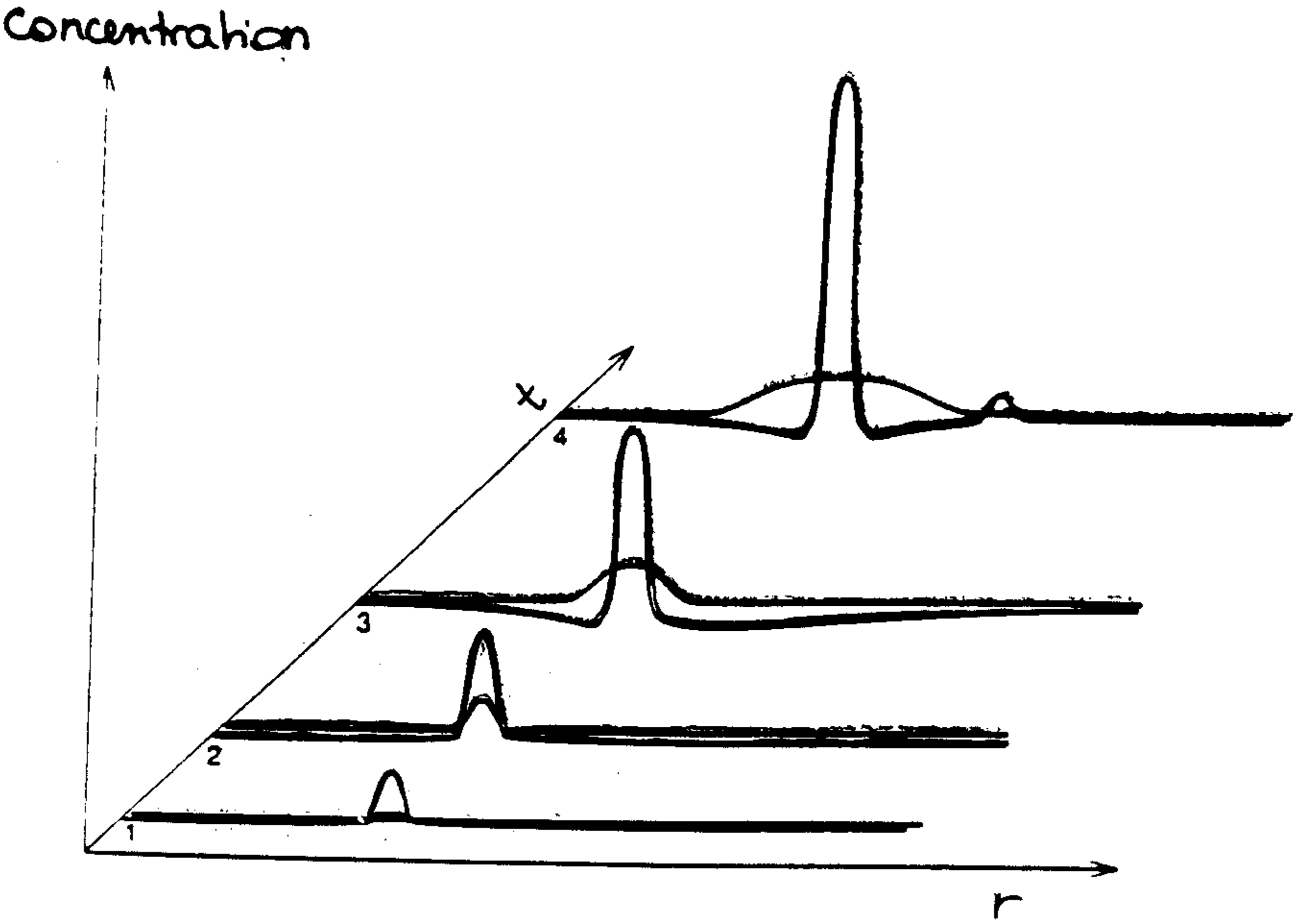
C / Turing - Hopf interaction

① Turing - Hopf mixed mode

② Bistability and localized structures

③ Subcritical instabilities

$$D_Y > D_X$$



<u>Activator</u> X : species involved in an autocatalytic reaction

<u>Inhibitor</u> Y : species that slows down the preceding activation step

Turing instability

Reaction - diffusion driven systen

$$\begin{cases} \dfrac{\partial X}{\partial t} = f(X,Y) + D_x \nabla^2 X \\ \dfrac{\partial Y}{\partial t} = g(X,Y) + D_y \nabla^2 Y \end{cases}$$

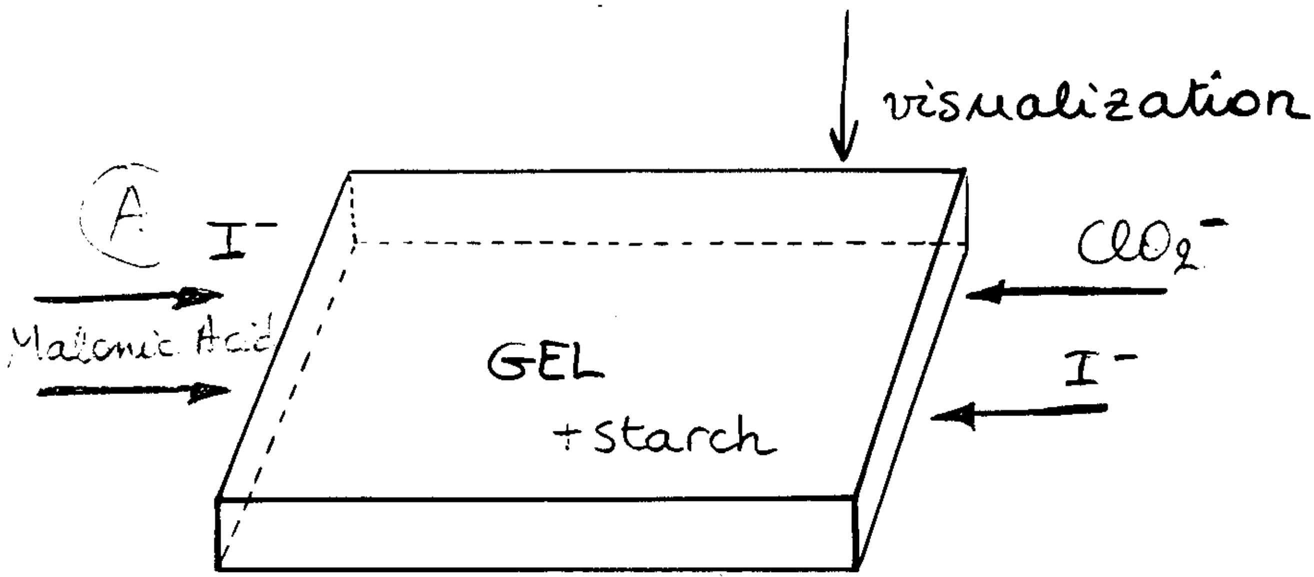
- .) <u>no diffusion</u> → stable uniform steady stat
- .) <u>diffusion</u> → the coupling between non linear kinetics + diffusion processes may induce a spatial symmetry breaking instabili (if $D_x \neq D_y$) leading to stationary periodic concentrat patterns ≡ dissipative structures

Intrinsic λ (Turing, 1952)

A. Turing: Phil. Trans. R. Soc. Lond., <u>237B</u>, 37 (1952)

A / Experiments

P. De Kepper, E. Dulos, J.-J. Perraud
C.R.P.P. - Bordeaux.



CIMA reaction

oscillations in time

when

$D(ClO_2^-) \simeq D(I^-)$

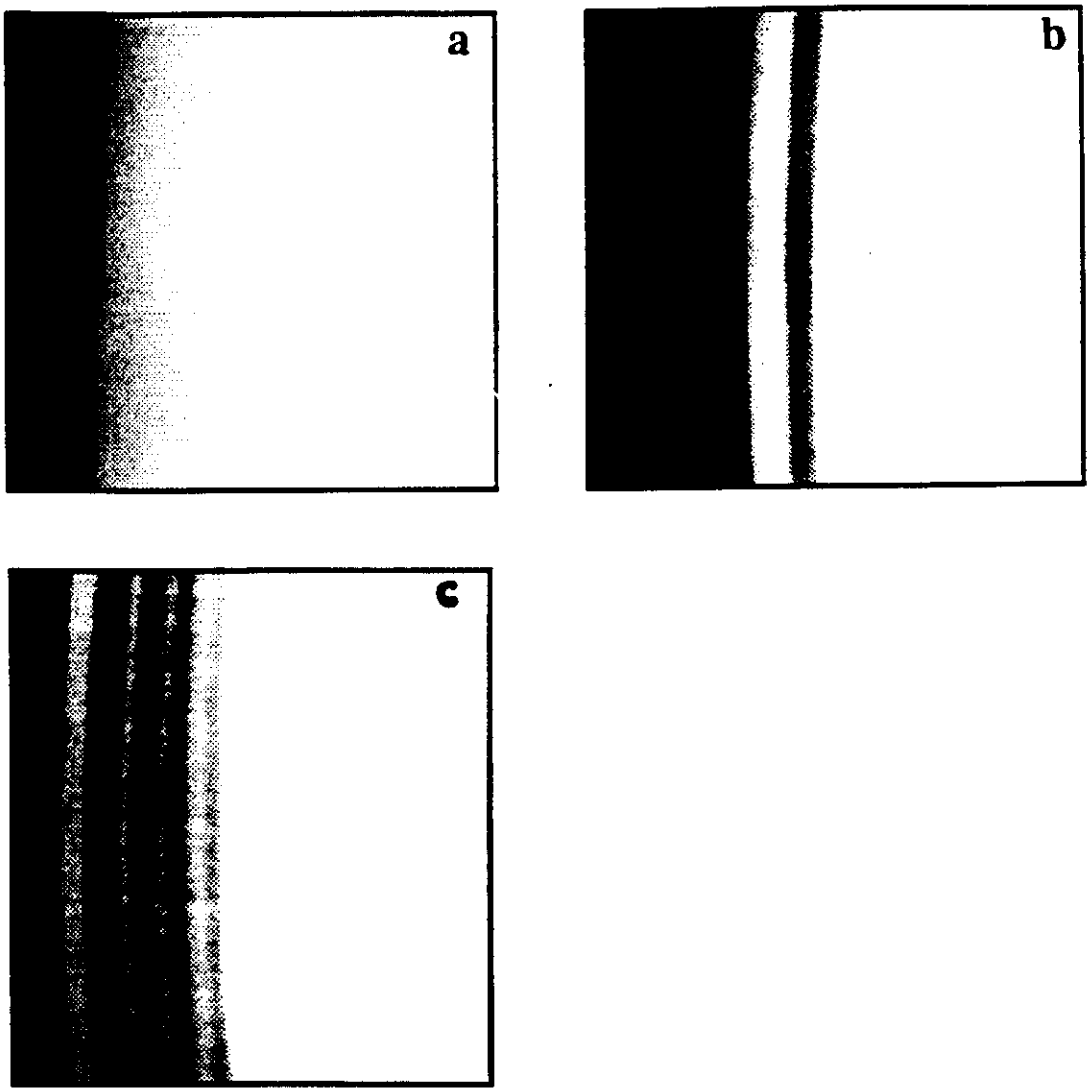
Hopf instability

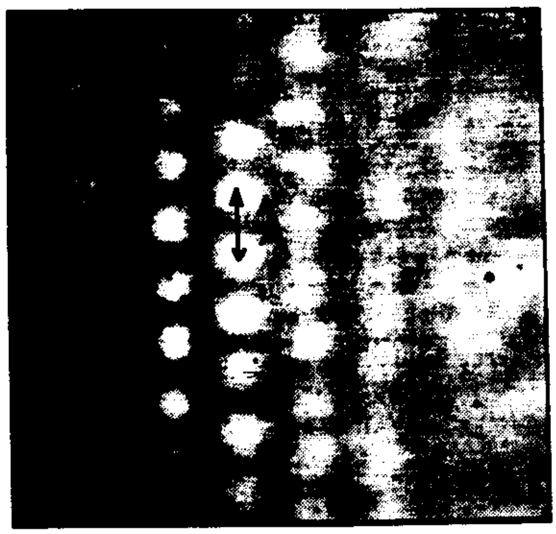
stationary patterns

oscillations in space

$D(ClO_2^-) \gg D(I^-)$

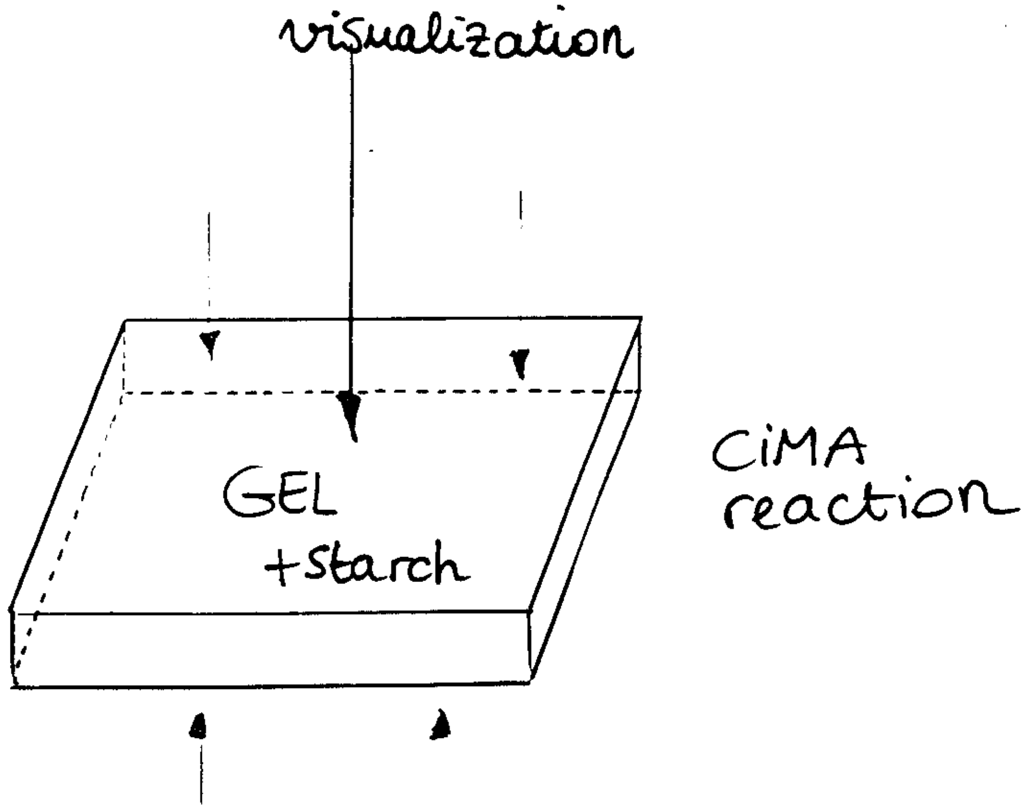
Turing instability





V. Castets et al., Phys. Rev. Letters, 64, 2953 1990

1991 : Q. Ouyang & H. Swinney
(Austin – Texas)



Quasi – 2D structures

Q. Ouyang & H. Swinney, Nature, 352, 610, 1991

(a) (b) (c) (d) (e) (f)

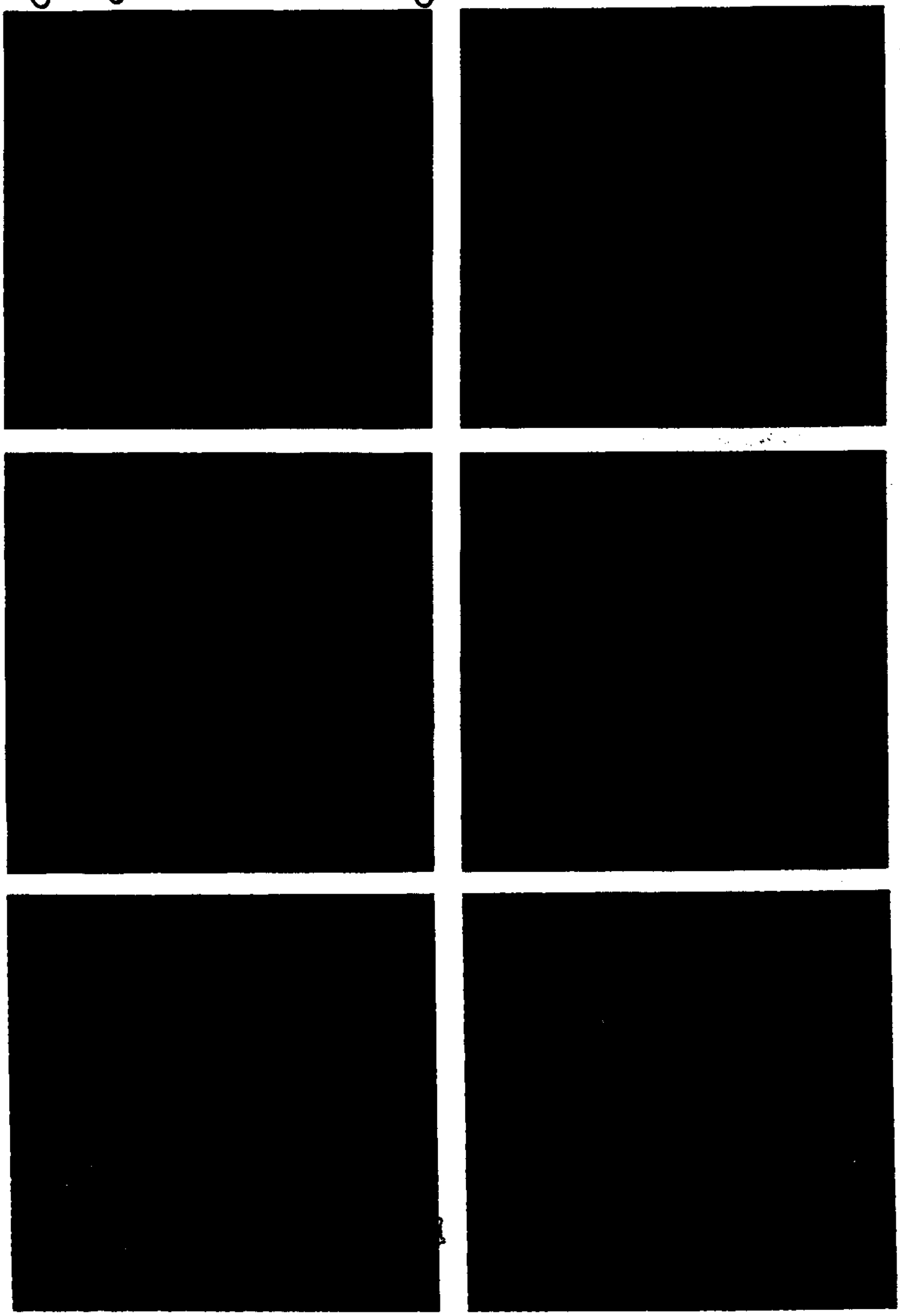
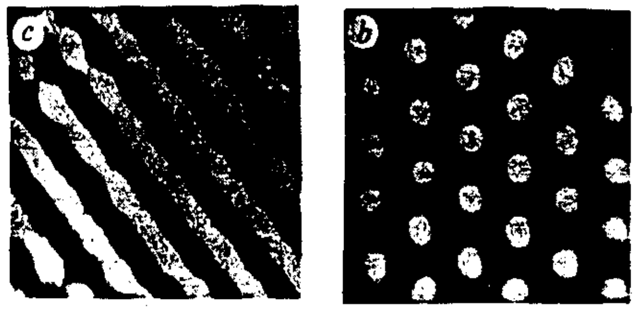
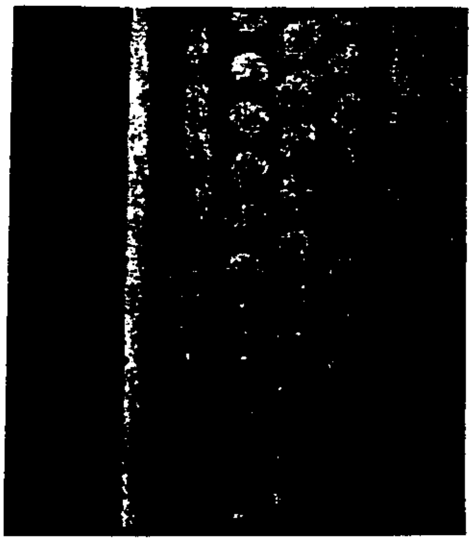


FIG. 3. Chemical patterns obtained with a continuously fed open spatial reactor: (a) transient honeycomb; (b) and (e), hexagons; (c) and (f), stripes; (d) mixed state. The bar beside each picture represents 1.0 mm. The blue and yellow colors of the image correspond, respectively, to the reduced and oxidized states of the system. The concentrations in compartments A and B were: $[I^-]_0$, $[CH_2(COOH)_2]_0^B$, $[ClO_2^-]_0^A$ (in mM) in (a), (b), and (d), 3.5, 8.3, 18.0; in (c), 5.0, 8.3, 18.0; in (e), 3.0, 9.0, 12.0; in (f), 3.0, 11.0, 18.0. The control parameters common for all these patterns were: $[Na_2SO_4]_0 = 4.5$ mM, $[H_2SO_4]_0^A = 0.5$ mM, $[H_2SO_4]_0^B = 8.5$ mM, temperature $= 5.6\,^{\circ}C$.

Ouyang & Swinney, Nature 352 (1991)

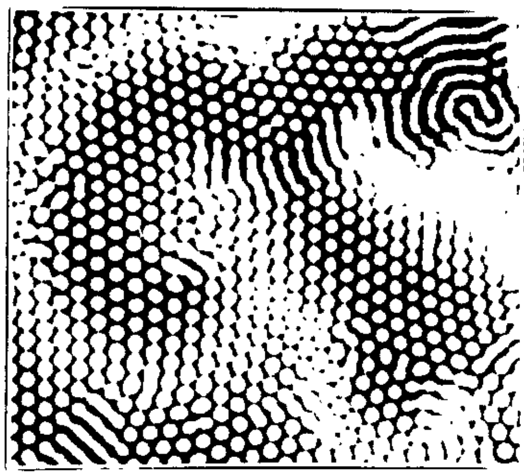
bifurcation diagrams





Castets et al. PRL 64 (199[illegible]

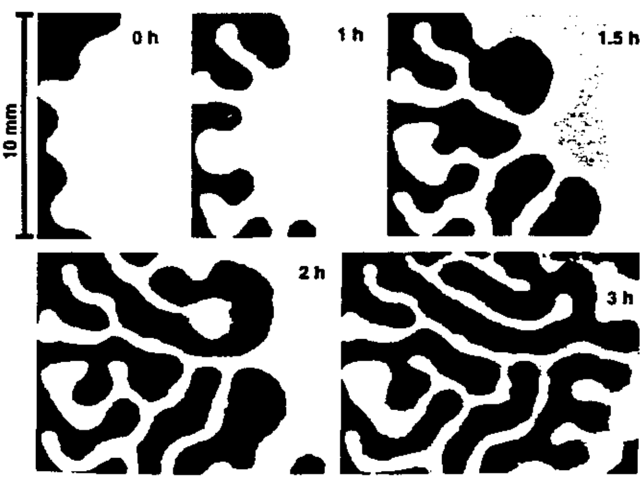
- 2D-3D
- ramps
- confinement



De Kepper et al. Int. J. Bif & Chao[illegible] 4 (1994)

- Spatiotemporal behavior
- Interaction betw[illegible] instabilities

- labyrinthine patterns
- morphological instabilities of fronts



Lee et al., Science 261 (1993)

Recently, lots of experiments have been devoted to the study of patterns in reaction - diffusion systems

⇝ questions :

- possible bifurcation diagrams
- role of bistability
- dimensionality : 2D - 3D ?
- ramps of concentration
- confinement
- spatiotemporal behavior
- nonstandard structures
- growth of structures
- labyrinthine patterns
- morphological instabilities of fronts

B/ Pattern selection in 2D reaction - diffusion systems

General form:

$$\partial_t U = f(U) + \underline{\underline{D}} \nabla^2 U$$

"reactions" → nonlinearities

- limit cycle (temporal oscillations)
- bistability
- excitability
- chaos

diffusion

- spatial structure
- waves
- spirals

Examples

. chemical systems
. charge dynamics in semiconducto
. gas discharge devices
. heterogeneous catalysis
. materials irradiated by particles or ligh
....

1) Chemical reaction - diffusion mod

$$A \longrightarrow X$$
$$X + B \longrightarrow Y + D$$
$$2X + Y \longrightarrow 3X$$
$$X \longrightarrow E$$

Brusselator

→ equations of evolution

$$\begin{cases} \partial_t X = A - (B+1)X + X^2 Y + D_X \nabla^2 X \\ \partial_t Y = BX - X^2 Y + D_Y \nabla^2 Y \end{cases}$$

Homogeneous steady state

$$\begin{cases} \partial_t X = 0 \\ \partial_t Y = 0 \end{cases} \longrightarrow \boxed{\begin{array}{l} X_S = A \\ Y_S = B/A \end{array}}$$

Linear stability analysis

$$\begin{cases} X = X_S + x \\ Y = Y_S + y \end{cases}$$

$$\Rightarrow \begin{cases} \partial_t x = (B-1)x + A^2 y + D_x \nabla^2 x \\ \partial_t y = -Bx - A^2 y + D_y \nabla^2 y \end{cases}$$

or also $\quad \partial_t \underline{u} = \underline{\underline{L}}\, \underline{u}$

with $\quad \underline{u} = \begin{pmatrix} x \\ y \end{pmatrix} \qquad \underline{\underline{L}} = \begin{pmatrix} B-1+D_x\nabla^2 & A^2 \\ -B & -A^2+D_y\nabla^2 \end{pmatrix}$

In infinite systems $\quad \boxed{\underline{u} \sim \underline{u}_0\, e^{\omega t}\, e^{i\underline{k}\cdot\underline{r}}}$

There exists a nontrivial solution $\underline{u}_0$ only if

$$\left| \underline{\underline{L}} - \omega \underline{\underline{I}} \right| = 0$$

Characteristic equation

<u>dispersion relation</u>

$$\omega^2 - \left(B-1-A^2-(D_x+D_y)k^2\right)\omega + A^2 + \left[A^2 D_x - (B-1)D_y\right]k^2 + D_x D_y k^4 =$$

or also in short

$$\omega^2 - T\omega + \Delta = 0$$

with $\quad T = \omega_1 + \omega_2$

$\qquad\quad \Delta = \omega_1 \omega_2$

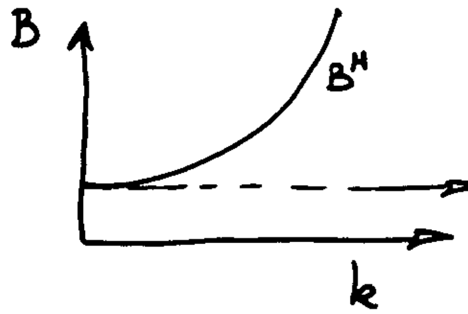
Hopf bifurcation

Pair of complex conjugated roots

$\mathrm{Re}(\omega_{1,2}) = 0 \longrightarrow T = 0$

$$\Downarrow$$

$$B^H = 1 + A^2 + (D_x + D_y)k^2$$



$$\boxed{B_c^H = 1 + A^2}$$ for $k = 0$

Homogeneous temporal oscillations with frequency $\omega = A$

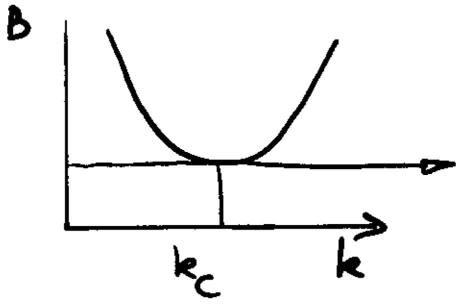
Turing bifurcation

2 Real roots. One of them vanishes

$\longrightarrow \Delta = 0$

$$\Downarrow$$

$$B^T = \frac{1}{D_y k^2}\left[A^2 + (A^2 D_x + D_y)k^2 + D_x D_y k^4\right]$$



$$\boxed{B_c^T = \left(1 + A\sqrt{\frac{D_x}{D_y}}\right)^2}$$

$$\longrightarrow k_c^2 = \frac{A}{\sqrt{D_x D_y}}$$

- The first instability to occur is the Turing one if $B_c^T < B_c^H$ i.e.:

$$\left(1 + A\sqrt{\frac{D_x}{D_y}}\right)^2 < 1 + A^2$$

Possible only if $\underline{\underline{D_x < D_y}}$

- The thresholds of the two instabilities coïncide at the codimension-two Turing-Hopf point such that $B_c^T = B_c^H$

Occurs when $$\frac{D_x}{D_y} = \left(\frac{\sqrt{1+A^2}-1}{A}\right)^2$$

2) amplitude equations

Beyond the bifurcation point, all modes with $|k| = k_c$ may grow (orientation degeneracy)

$$\rightarrow \quad \underline{c} = \underline{c}_0 + \sum_{i=1}^{m} \left(A_i \, e^{i \underline{k}_i \cdot \underline{r}} + c.c. \right)$$

→ Patterns characterized by m pairs of wavevectors

One could expect the formation of planforms with any type of symmetry

The non linear competition between the active modes select some planforms

↓

weakly non linear analysis to derive the evolution equations for $A(t)$

perturbation expansion

Non linear evolution equation for the fluctuations $\underline{u} = \underline{c} - \underline{c}_0$:

$$\frac{\partial \underline{u}}{\partial t} = \underline{\underline{L}}\ \underline{u} + N(\underline{u}, \underline{u})$$

In the vicinity of the bifurcation point we look for

$$\underline{u} = \varepsilon\, \underline{u}_1 + \varepsilon^2\, \underline{u}_2 + \varepsilon^3\, \underline{u}_3 + \dots$$

where ε is related to the distance from the critical point through the expansion of the control paramete

$$B = B_c + \varepsilon B_1 + \varepsilon^2 B_2 + \varepsilon^3 B_3 + \dots$$

$$\downarrow$$

at different orders in ε : set of linear equations. The solvability conditions of these equations lead to a set of ODEs which are the amplitude equations.

Example on the Bruxelator

$$\begin{cases} \partial_t X = A - (B+1)X + X^2 Y + D_x \nabla^2 X \\ \partial_t Y = BX - X^2 Y + D_y \nabla^2 Y \end{cases}$$

$$\begin{pmatrix} X \\ Y \end{pmatrix} = \begin{pmatrix} X_s \\ Y_s \end{pmatrix} + \begin{pmatrix} x \\ y \end{pmatrix}$$

$\rightarrow$ non linear evolution equations for x, y

$$\begin{cases} \partial_t x = (B-1)x + A^2 y + \frac{B}{A} x^2 + 2Axy + x^2 y + D_x \nabla^2 x \\ \partial_t y = -Bx - A^2 y - \frac{B}{A} x^2 - 2Axy - x^2 y + D_y \nabla^2 y \end{cases}$$

Perturbation expansion

$\rightarrow$ $B = B_c + \varepsilon B_1 + \varepsilon^2 B_2 + \dots$

$\rightarrow$ $\begin{pmatrix} x \\ y \end{pmatrix} = \varepsilon \begin{pmatrix} x_1 \\ y_1 \end{pmatrix} + \varepsilon^2 \begin{pmatrix} x_2 \\ y_2 \end{pmatrix} + \varepsilon^3 \begin{pmatrix} x_3 \\ y_3 \end{pmatrix} + \dots$

or also $\underline{u} = \varepsilon \underline{u}_1 + \varepsilon^2 \underline{u}_2 + \varepsilon^3 \underline{u}_3$

$\rightarrow$ $\partial_t = \partial_{t_0} + \varepsilon \partial_{\tau_1} + \varepsilon^2 \partial_{\tau_2} + \dots$

$\rightarrow$ $\partial_x = \partial_{x_0} + \varepsilon \partial_{x_1} + \varepsilon^2 \partial_{x_2} + \dots$

$\rightarrow$ $\partial_y = \partial_{y_0} + \varepsilon^{1/2} \partial_{y_1} + \varepsilon \partial_{y_2} + \dots$

$\boxed{\text{Order } \varepsilon^1}$ $\boxed{\left(\partial_{\tau_0} - \underline{\underline{L}}_c\right)\begin{pmatrix} x_1 \\ y_1 \end{pmatrix} = 0}$

where $\underline{\underline{L}}_c$ is the linear operator appearing in the linear stability analysis computed for $B = B_c$ and $k = k_c$:

$$\longrightarrow \underline{\underline{L}}_c = \begin{pmatrix} B_c - 1 + D_x \nabla_0^2 & A^2 \\ -B_c & -A^2 + D_y \nabla_0^2 \end{pmatrix}$$

$\longrightarrow$ $\underline{u}_1 = \begin{pmatrix} x_1 \\ y_1 \end{pmatrix}$ is proportional to the eigenmodes of $\underline{\underline{L}}_c$ corresponding to a zero eigenvalue

i.e. : $$\boxed{\underline{u}_1 = \sum_{j=1}^{m} \left(W_i\, e^{i\underline{k}_j \cdot \underline{r}} + cc\right)\begin{pmatrix} \omega_x \\ \omega_y \end{pmatrix}}$$

with $|\underline{k}_j| = k_c$

where $$\underline{w} = \begin{pmatrix} 1 \\ -\frac{\eta}{A}(1 + A\eta) \end{pmatrix} e^{i\underline{k}_c \cdot \underline{r}} = \begin{pmatrix} \omega_x \\ \omega_y \end{pmatrix} e^{ik}$$

is the right eigenvector of $\underline{\underline{L}}_c$

$$\eta = \sqrt{\frac{D_x}{D_y}}$$

NB: Definition of the scalar product

The left eigenvectors of $\underline{\underline{L}}_c$ are:

$$\underline{v} = \left(1, \frac{A\eta}{1+A\eta}\right) e^{i\underline{k}_c \cdot \underline{r}} = (v_x, v_y)\, e^{i\underline{k}_c}$$

Hence, we define the scalar product as

$$\langle \underline{v} | \underline{w} \rangle = \frac{1}{V}\int_V d\underline{r}\ (v_x^*, v_y^*)\, e^{-i\underline{k}_c \cdot \underline{r}} \cdot \begin{pmatrix} w_x \\ w_y \end{pmatrix} e^{i\underline{k}_c' \cdot \underline{r}}$$

where $*$ means complex conjugat

As $\displaystyle\int_V d\underline{r}\ e^{i\underline{n}\cdot\underline{r}} = 0$ for $\underline{n} \neq \underline{0}$

the only contribution comes from resonant terms such that $\underline{k}_c' - \underline{k}_c = \underline{0}$

Hence there remains:

$$\frac{1}{V}\int_V d\underline{r}\ (v_x^*, v_y^*)\begin{pmatrix} w_x \\ w_y \end{pmatrix} = v_x^* w_x + v_y^* w_y$$

$$= 1 - \eta^2 = \frac{D_y - D_x}{D_y}$$

~~Example~~ Order ε^2: Stripes in 1D system

→ one pair of wavevectors

$$\underline{u}_1 = \begin{pmatrix} x_1 \\ y_1 \end{pmatrix} = \begin{pmatrix} w_x \\ w_y \end{pmatrix} \left(W e^{ik_c r} + W^* e^{-ik_c r} \right)$$

Order ε^2

$$\boxed{\left(\partial_{\tau_0} - \underline{\underline{L}}_c\right)\begin{pmatrix} x_2 \\ y_2 \end{pmatrix} = \begin{pmatrix} I_{2x} \\ I_{2y} \end{pmatrix} = \underline{I}_2}$$

$$\begin{pmatrix} I_{2x} \\ I_{2y} \end{pmatrix} = \left[\frac{B_c}{A} x_1^2 + 2A x_1 y_1 \right] \begin{pmatrix} +1 \\ -1 \end{pmatrix}$$

$$+ \left[-\partial_{\tau_1} + \begin{pmatrix} B_1 + 2D_x \nabla_0 \nabla_1 & 0 \\ -B_1 & 2D_y \nabla_0 \nabla_1 \end{pmatrix} \right] \begin{pmatrix} x_1 \\ y_1 \end{pmatrix}$$

→ at higher orders, we have nonhomogeneous linear equations such as $(\partial_{\tau_0} - \underline{\underline{L}}_c)\underline{u}_i = \underline{I}_i$

The operator $(\partial_{\tau_0} - \underline{\underline{L}}_c)$ is not invertible as it has a zero eigenvalue

→ $\exists$ solution $\underline{u}_2$ only if $\underline{I}_2$ is orthogonal to the kernel of $(\partial_{\tau_0} - \underline{\underline{L}}_c)^+$

or $\langle \underline{v} | I_2 \rangle = 0$ — Solvability condition

Keeping only the resonant terms, we have then

$$\begin{cases} v_x I_{2x}^{+} + v_y I_{2y}^{+} = 0 \\ v_x I_{2x}^{-} + v_y I_{2y}^{-} = 0 \end{cases}$$

where $I^{+}_{2x(y)}$ and $I^{-}_{2x(y)}$ are the coefficient of the terms in $\exp(+i\underline{k}_c\underline{r})$ and $\exp(-i\underline{k}_c\underline{r})$ in $I_{2x(y)}$ respectively.

These conditions are satisfied if

$\boxed{B_1 = 0} \longrightarrow \varepsilon \sim \sqrt{B - B_c}$

super-critical bifurcation

$$\boxed{\begin{array}{l} \partial_{\tau_1} W = 0 \\ \partial_{\tau_1} W^* = 0 \end{array}}$$

No dynamical equation for A

$\longrightarrow$ go up to order ε^3

$\longrightarrow$ first we need to know $\begin{pmatrix} x_2 \\ y_2 \end{pmatrix} = \underline{u}_2$

As $(\partial_{T_0} - \underline{L}_c)\underline{u}_2 = \underline{I}_2$ and $\underline{I}_2 \sim x_1^2$ or $x_1 y_1$

$\underline{u}_2$ must be of the form

$$\boxed{\underline{u}_2 = \begin{pmatrix} x_2 \\ y_2 \end{pmatrix} = \begin{pmatrix} a_0 \\ b_0 \end{pmatrix} + \begin{pmatrix} a_1 \\ b_1 \end{pmatrix} e^{ik_c r} + \begin{pmatrix} a_2 \\ b_2 \end{pmatrix} e^{2ik_c r} + \begin{pmatrix} a_1^* \\ b_1^* \end{pmatrix} e^{-ik_c r} + \begin{pmatrix} a_2^* \\ b_2^* \end{pmatrix} e^{-2ik_c}}$$

We find

$$a_0 = 0$$

$$b_0 = -\frac{2}{A^3}\left(1 - A^2\eta^2\right) WW^*$$

$$a_2 = \frac{4}{9} \frac{(1 - A^2\eta^2)}{A^2\eta} W^2$$

$$b_2 = -\frac{1}{9} \frac{(1 - A^2\eta^2)(1 + 4A\eta)}{A^3} W^2$$

$$a_1 + \frac{A}{\eta(1 + A\eta)} b_1 = \frac{-2i\sqrt{D_x D_y}}{A(1 + A\eta)} k_c \nabla_1 W$$

$\boxed{\text{Order } \varepsilon^3}$ $$(\partial_{\tau_0} - \underline{\underline{L}}_c)\begin{pmatrix} x_3 \\ y_3 \end{pmatrix} = \begin{pmatrix} I_{3x} \\ I_{3y} \end{pmatrix} = \underline{I}_3$$

with $$\begin{pmatrix} I_{3x} \\ I_{3y} \end{pmatrix} = \left[-\partial_{\tau_2} + \begin{pmatrix} B_2 + D_x \nabla_1^2 & 0 \\ -B_2 & D_y \nabla_1^2 \end{pmatrix} \right] \begin{pmatrix} x_1 \\ y_1 \end{pmatrix}$$

$$+ \begin{pmatrix} 2D_x \nabla_0 \nabla_1 & 0 \\ 0 & 2D_y \nabla_0 \nabla_1 \end{pmatrix} \begin{pmatrix} x_2 \\ y_2 \end{pmatrix}$$

$$+ \left(\frac{2B_c}{A} x_1 x_2 + 2A(x_1 y_2 + x_2 y_1) + x_1^2 y_1 \right) \begin{pmatrix} +1 \\ -1 \end{pmatrix}$$

or also in short $(\partial_{\tau_0} - \underline{\underline{L}}_c)\, \underline{u}_3 = \underline{I}_3$

Solvability condition $\langle \underline{v} | \underline{I}_3 \rangle = 0$

Keeping only the resonant terms:

$$\begin{cases} v_x I_{3x}^{+} + v_y I_{3y}^{+} = 0 \\ v_x I_{3x}^{-} + v_y I_{3y}^{-} = 0 \end{cases}$$

The first condition gives

$$\boxed{(1-\eta^2)\,\partial_{\tau_2} W = \frac{B_2}{1+A\eta} W + \frac{4D_x}{1+A\eta}\nabla_1^2 W - \frac{1}{9A^3\eta}\left(-8A^3\eta^3+5A^2\eta^2+38A\eta-8\right)|W|^2 W}$$

B_2 is unknown $\longrightarrow$ multiplying both members by $\varepsilon^2/(1-\eta^2)$ and taking

$$\varepsilon^2 \tau_2 = \partial_\tau \qquad T = \varepsilon W$$

$$\varepsilon^2 B_2 = B - B_c$$

$$\varepsilon^2 \nabla_1^2 = \nabla^2$$

we have

$$\boxed{\partial_\tau T = \mu T - g|T|^2 T + \mathcal{D}\nabla^2 T}$$

with

$$\mu = \frac{1+A\eta}{1-\eta^2}\left(\frac{B-B_c}{B_c}\right)$$

$$g = \frac{-8A^3\eta + 5A^2\eta^2 + 38A\eta - 8}{9A^3\eta(1-\eta^2)}$$

$$\mathcal{D} = \frac{4D_x(1+A\eta)}{B_c(1-\eta^2)}$$

The 2^{d} condition gives the c.c. of this equati

Rhombs

$\longrightarrow$ 2 pairs of wavevectors

$$\underline{u}_1 = \begin{pmatrix} x_1 \\ y_1 \end{pmatrix} = \begin{pmatrix} w_x \\ w_y \end{pmatrix} \left[W_1 e^{i\underline{k}_1 \cdot \underline{r}} + W_2 e^{i\underline{k}_2 \cdot \underline{r}} + cc \right]$$

with $|\underline{k}_1| = |\underline{k}_2| = k_c$

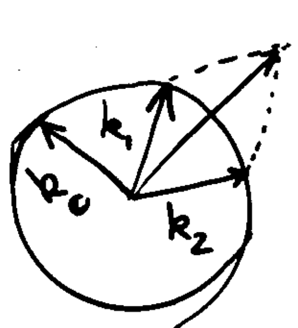
let's neglect the spatial dependence of W_i

$\boxed{\text{Order } \varepsilon^2}$

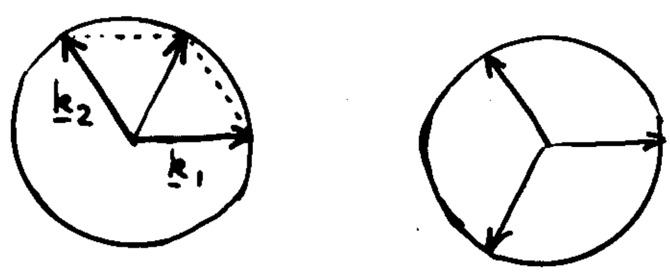
$$(\partial_{T_0} - \underline{\underline{L}}_c)\begin{pmatrix} x_2 \\ y_2 \end{pmatrix} = \left(B_1 x_1 + \frac{B_c}{A} x_1^2 + 2A x_1 y_1\right)\begin{pmatrix} +1 \\ -1 \end{pmatrix} = \begin{pmatrix} I_{2x} \\ I_{2y} \end{pmatrix}$$

$\sim e^{i\underline{k}_c \cdot \underline{r}}$

$\sim e^{2i\underline{k}_1 \cdot \underline{r}}, \underbrace{e^{2i\underline{k}_2 \cdot \underline{r}}, e^{i(\underline{k}_1 + \underline{k}_2)}}_{\text{NOT RESONANT}}$



$\theta = 120°$



$\underline{k}_1 + \underline{k}_2 \neq \underline{k}_c$

not resonant

$\longrightarrow$ RHOMBS

$\underline{k}_1 + \underline{k}_2 = \underline{k}_c$

HEXAGONS

$\longrightarrow$ resonance excites 3rd wavenumber

For rhombs, the only resonant contribution at order ε^2 comes from $B_1 x_1$

$\rightarrow \langle \underline{v} | \underline{I}_2 \rangle = 0$ if $\boxed{B_1 = 0}$

At order ε^2, the solution is of the form

$$\begin{pmatrix} x_2 \\ y_2 \end{pmatrix} = \begin{pmatrix} 0 \\ b_0 \end{pmatrix} + \left[\begin{pmatrix} a_{11} \\ b_{11} \end{pmatrix} e^{2i\underline{k}_1 \cdot \underline{r}} + \begin{pmatrix} a_{22} \\ b_{22} \end{pmatrix} e^{2i\underline{k}_2 \cdot \underline{r}} \right.$$

$$\left. + \begin{pmatrix} a_{12} \\ b_{12} \end{pmatrix} e^{i(\underline{k}_1 + \underline{k}_2)\underline{r}} + \begin{pmatrix} a_{1,-2} \\ b_{1,-2} \end{pmatrix} e^{i(\underline{k}_1 - \underline{k}_2)\underline{r}} + cc \right]$$

$\boxed{\text{Order } \varepsilon^3}$

$$\left(\partial_{\tau_0} - \underline{\underline{L}}_c\right) \underline{u}_3 = \underline{I}_3$$

Solvability condition $\langle \underline{v} | \underline{I}_3 \rangle$

$$\leadsto \boxed{\partial_\tau T_1 = \mu T_1 - g |T_1|^2 T_1 - g_{ND}(\theta) |T_2|^2 T_1}$$

+ same type of equation for T_2

with $T_1 = \varepsilon W_1$

$\partial_\tau = \varepsilon^2 \partial_{\tau_2}$

$B - B_C = \varepsilon^2 B_2$

RHOMBS

$$\mu = \frac{1+A\eta}{1-\eta^2}\left(\frac{B-B_c}{B_c}\right)$$

$$g = \frac{-8A^3\eta^3 + 5A^2\eta^2 + 38A\eta - 8}{9A^3\eta\,(1-\eta^2)}$$

$$g_{ND}(\theta) = \frac{2(2+A\eta)}{A^2(1-\eta^2)} - \frac{8(1-A\eta)}{A^2(1-\eta^2)}\cdot\left[\frac{2(1+A\eta-A^2\eta^2)}{A\eta\,(1-4\cos^2\theta)^2} - \frac{(1+4\cos^2\theta)}{(1-4\cos^2\theta)}\right]$$

HEXAGONS

→ 3 pairs of wavevectors

$$\underline{u}_1 = \begin{pmatrix} x_1 \\ y_1 \end{pmatrix} = \begin{pmatrix} \omega_x \\ \omega_y \end{pmatrix} \left[W_1 e^{i\underline{k}_1\cdot\underline{r}} + W_2 e^{i\underline{k}_2\cdot\underline{r}} + W_3 e^{i\underline{k}_3\cdot\underline{r}} + c.c. \right]$$

with $|\underline{k}_1| = |\underline{k}_2| = |\underline{k}_3| = k_c$

Order ε^2

$$(\partial_{T_0} - \underline{\underline{L}}_c)\begin{pmatrix} x_2 \\ y_2 \end{pmatrix} = \left(B_1 x_1 + \frac{B_c}{A} x_1^2 + 2A x_1 y_1 \right)\begin{pmatrix} +1 \\ -1 \end{pmatrix} = \begin{pmatrix} I_{2x} \\ I_{2y} \end{pmatrix}$$

give terms like $e^{i(\underline{k}_1+\underline{k}_2)\underline{r}} = e^{-i\underline{k}_3\cdot\underline{r}}$

Solvability condition $\langle \underline{v} | \underline{I}_2 \rangle = 0$

or $v_x I_{2x}^{+} + v_y I_{2y}^{+} = 0$

As $I_{2x}^{+} = -I_{2y}^{+} \rightarrow (v_x - v_y)\, I_{2x}^{+} = 0$

or

$$\boxed{B_1 W_1 + \frac{2}{A}(1 - A^2 \eta^2)\, W_2^{*} W_3^{*} = 0}$$

$B_1 \neq 0$

→ subcritical bif.

We find next

$$\begin{pmatrix} x_2 \\ y_2 \end{pmatrix} = \begin{pmatrix} a_0 \\ b_0 \end{pmatrix} + \left[\begin{pmatrix} a_{11} \\ b_{11} \end{pmatrix} e^{2i\underline{k}_1\underline{r}} + \begin{pmatrix} a_{22} \\ b_{22} \end{pmatrix} e^{2i\underline{k}_2} \right.$$

$$+ \begin{pmatrix} a_{33} \\ b_{33} \end{pmatrix} e^{2i\underline{k}_3\underline{r}} + \begin{pmatrix} a_{1,-2} \\ b_{1,-2} \end{pmatrix} e^{i(\underline{k}_1-\underline{k}_2)\underline{r}} + \begin{pmatrix} a_{1,-3} \\ b_{1,-3} \end{pmatrix} e^{i(\underline{k}_1-\underline{k}_3)\underline{r}} + \begin{pmatrix} a_{2,-3} \\ b_{2,-3} \end{pmatrix} e^{i(\underline{k}_2-\underline{k}_3)} +$$

$\boxed{\text{Order } \varepsilon^3}$ Solvability condition

$$\boxed{(1-\eta^2)\frac{\partial W_1}{\partial \tau_2} = \frac{1}{(1+A\eta)}\left[B_2 W_1 + \frac{2B_1}{A} W_2^* W_3^* - g'|W_1|^2 W_1 - h'(|W_2|^2 + |W_3|^2) W_1\right]}$$

Summing the solvability conditions of order ε^2 and order ε^3, we have

$$\varepsilon^3(1-\eta^2)\frac{\partial W_1}{\partial \tau_2} = \frac{1}{(1+A\eta)}\big(\overbrace{\varepsilon B_1 + \varepsilon^2 B_2}^{\sim (B-B_c)}\big)\varepsilon W_1$$

$$+ \left\{\frac{2}{A}(1-A\eta) + \frac{2\varepsilon B_1}{A(1+A\eta)}\right\}\varepsilon^2 W_2^* W_3^*$$

$$- \frac{g'}{(1+A\eta)}\varepsilon^3 |W_1|^2 W_1 - \frac{h'}{(1+A\eta)}\varepsilon^3(|W_2|^2+|W_3|^2) W_1$$

Using the usual scaling, we get

with $\partial_t = \varepsilon^2 \partial_{\tau_2}$

$T_i = \varepsilon W_i$:

$$\boxed{\partial_\tau T_1 = \mu T_1 + v\, T_2^* T_3^* - g\,|T_1|^2 T_1 - h\left(|T_2|^2 + |T_3|^2\right)}$$

+ eq. for T_2 & T_3 obtained by permutation of indices

$$\mu = \frac{1 + A\eta}{1 - \eta^2} \left(\frac{B - B_c}{B_c} \right)$$

$$v = \frac{2(1 - A\eta)}{A(1 - \eta^2)} + \frac{2}{A}\mu$$

$$g = \frac{-8A^3\eta^3 + 5A^2\eta^2 + 38A\eta - 8}{9A^3\eta(1-\eta^2)}$$

$$h = \frac{-3A^3\eta^3 + 7A^2\eta^2 + 5A\eta - 3}{9A^3\eta(1-\eta^2)}.$$

3/ Bifurcation diagram

→ study of the stability of the different solutions of the amplitude e

A/ Stripes

$$\partial_t T = \mu T - g |T|^2 T$$

$$g > 0$$

let's separate the real amplitude R and the phase θ of the structure by writing

$$T = R e^{i\theta}$$

$$\longrightarrow \begin{cases} \partial_t R = \mu R - g R^3 \\ \partial_t \theta = 0 \end{cases} \longrightarrow \theta = cst$$

The different consta relate to a translation of the pattern.

Real part → 2 stationary states

$$R_S = 0 \quad \text{or} \quad R_S = \sqrt{\frac{\mu}{g}}$$

Stability Let's take $R = R_S + \delta R$

$$\rightarrow \partial_t \delta R = (\mu - 3 R_S^2 g)\, \delta R$$

$R_S = 0 \longrightarrow$ unstable for $\mu > 0$

$R_S = \sqrt{\frac{\mu}{g}} \longrightarrow \partial_t \delta R = -2\mu \delta R \longrightarrow$ stable for $\mu >$

B/ Rhombs

$$\begin{cases} \partial_t T_1 = \mu T_1 - g|T_1|^2 T_1 - g_{ND} |T_2|^2 T_1 \\ \partial_t T_2 = \mu T_2 - g|T_2|^2 T_2 - g_{ND} |T_1|^2 T_2 \end{cases}$$

Let's take $T_j = R_j e^{i\theta_j}$

$$\rightarrow \begin{cases} \partial_t R_j = \mu R_j - g R_j^3 - g_{ND} R_k^2 R_j \\ \partial_t \theta_j = 0 \end{cases}$$

$\hookrightarrow \theta_j = ct$

Real part $\rightarrow$ 3 stationary states

1) $R_1 = R_2 = 0$
2) $R_1 = R_S \quad R_2 = 0 \rightarrow$ stripes
3) $R_1 = R_2 = R_R = \sqrt{\dfrac{\mu}{g+g_{ND}}}$

This solution exists for $\mu > 0$ if $(g+g_{ND}) >$
" " $\mu < 0$ if $(g+g_{ND}) <$

Stability .) identical perturbations

$$R_1 = R_2 = R_R + \delta R$$

$$\rightarrow \partial_t \delta R = -2(g+g_{ND})\,\delta R \cdot R_R^2$$

$\rightarrow$ $\boxed{\text{rhombs stable for } \mu > 0}$

•) <u>different perturbations</u>

$$\begin{cases} R_1 = R_R + \delta R_1 \\ R_2 = R_R + \delta R_2 \end{cases}$$

$$\longrightarrow \quad \partial_t \begin{pmatrix} \delta R_1 \\ \delta R_2 \end{pmatrix} = -2R_R^2 \begin{pmatrix} g & g_{ND} \\ g_{ND} & g \end{pmatrix} \begin{pmatrix} \delta R_1 \\ \delta R_2 \end{pmatrix}$$

$$\longrightarrow \quad \omega^2 - 2g\omega + g^2 - g_{ND}^2 = 0$$

unstable if $\Delta = \omega_1 \omega_2 < 0$ i.e. $g^2 - g_{ND}^2 < 0$

$\longrightarrow$ $\boxed{\text{rhombs unstable if } g < g_{ND}}$

C/ Stability of stripes vs rhombs

If $\mu > 0$: stripes and rhombs are both possible solutions

$\longrightarrow$ to know which one will be favored, let's study the stabilit[y] of stripes with regard to perturbati[ons] favoring rhombs

$$\longrightarrow \quad \begin{cases} R_1 = R_S + \delta R_1 \\ R_2 = \delta R_2 \end{cases} \qquad \text{with } R_S = \sqrt{\frac{\mu}{g}}$$

$$\longrightarrow \quad \begin{cases} \partial_t \delta R_1 = -2\mu\, \delta R_1 \\ \partial_t \delta R_2 = \mu \left(\frac{g - g_{ND}}{g}\right) \delta R_2 \end{cases} \longrightarrow \boxed{\text{stripes stable if } g < g_{ND}}$$

$\Rightarrow$ stripes and rhombs are mutually exclusive.

D/ Stability of hexagons

$$\partial_t T_1 = \mu T_1 + v T_2^* T_3^* - g|T_1|^2 T_1 - h(|T_2|^2 + |T_3|^2)$$

+ same equation for T_2 & T_3

Let's take $T_j = R_j e^{i\theta_j}$

a) phase

$\Rightarrow$ We end up with the following equation for $\theta = \theta_1 + \theta_2 + \theta_3$:

$$\partial_t \theta = -v \left[\overbrace{\frac{R_1^2 R_2^2 + R_1^2 R_3^2 + R_2^2 R_3^2}{R_1 R_2 R_3}}^{C>0} \right] \sin\theta$$

Stationary solution : $\boxed{\theta = 0 \quad \text{or } \pi}$

stability $\quad \sin(\theta_s + \delta\theta) = \sin\theta_s + \delta\theta \cos\theta_s$

$\rightarrow \partial_t \delta\theta = -v\, C\, \delta\theta \cos\theta_s$

If
$$\boxed{\begin{array}{lll} \theta_s = 0 & \longrightarrow & \text{stable if } v > 0 \\ \theta_s = \pi & \longrightarrow & \text{stable if } v < 0 \end{array}}$$

b) Real part

$$\partial_t R_1 = \mu R_1 + |v|\, R_2 R_3 - g R_1^3 - h(R_2^2 + R_3^2) R_1$$

Stationary states:

1) homogeneous $\quad R_1 = R_2 = R_3 = 0$
2) stripes
3) Mixed solution $\quad R_1 \neq R_2 = R_3$

always unstable

4) hexagons $\quad R_1 = R_2 = R_3 = R$

where R is solution of

$$-R\left[(g+2h) R^2 - |v| R - \mu\right] = 0$$

or $\quad -R\,(R - R_+)(R - R_-) = 0$

with $\boxed{R_\pm = \dfrac{|v| \pm \sqrt{v^2 + 4\mu(g+2h)}}{2(g+2h)}}$

R must be real $\longrightarrow$ hexagons exist only if

$$\boxed{\mu > \mu_+ = \frac{-v^2}{4(g+2h)}}$$

μ_+ is negative

as hexagons appear subcritical

c) Stability •) identical perturbations

Let's take $R = R_+ + \delta R$ into $-R(R-R_+)(R-R_-)$e.

$$\longrightarrow \partial_t \delta R = -R_+ \underbrace{(R_+ - R_-)}_{0} \delta R$$

$$\longrightarrow \boxed{R_+ \text{ is stable}}$$

Similarly, we show that R_- is unstabl

•) different perturbations

$$\begin{cases} R_1 = R_+ + \delta R_1 \\ R_2 = R_+ + \delta R_2 \\ R_3 = R_+ + \delta R_3 \end{cases}$$

$$\longrightarrow \boxed{\text{hexagons unstable for } \mu > \mu_H}$$

where $\mu_H = \dfrac{v^2(2g+h)}{(g-h)^2}$

E/ Stability of Stripes vs hexagons

If $\mu > 0$: stripes and hexagons coexist

$\longrightarrow$ study of their relative stability

Let's take $\begin{cases} R_1 = R_S + \delta R_1 \\ R_2 = \delta R_2 \\ R_3 = \delta R_3 \end{cases}$ with $R_S = \sqrt{\frac{\mu}{g}}$

$\rightsquigarrow \cdots \rightsquigarrow$ $\boxed{\text{stripes stable for } \mu > \mu_B}$

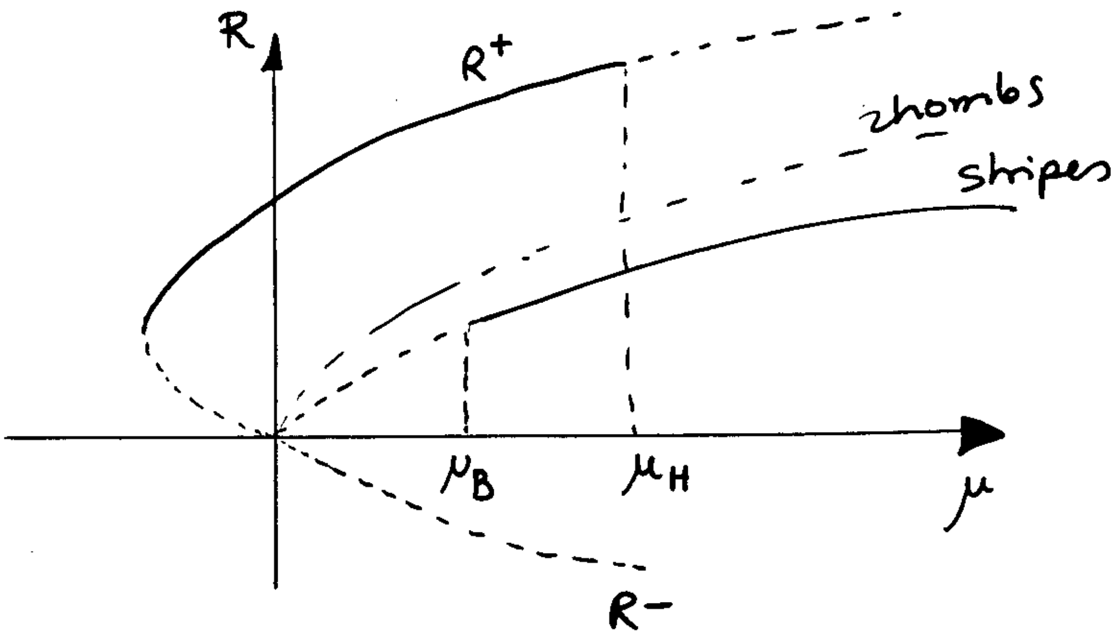
with $\mu_B = \dfrac{v^2 g}{(g-h)^2}$

Summary

Stripes
- supercritical : stable for $\mu > 0$
- stable versus rhombs if $g < g_{ND}$
- stable versus hexagons if $\mu_B < \mu$

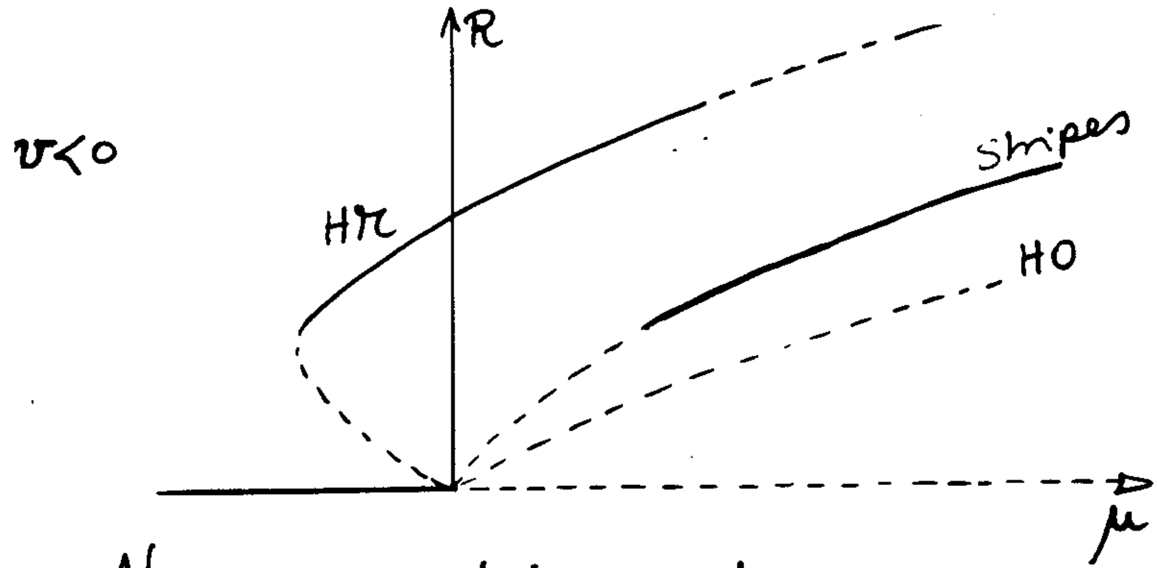
rhombs
- supercritical
- unstable if $g < g_{ND}$

hexagons
- subcritical → exist for $\mu_+ < \mu$ with μ_+ being negative
- unstable versus stripes if $\mu_H < \mu$
- HO if $v > 0$
  HR if $v < 0$
- 2 branches : H_+ stable
  H_- unstable

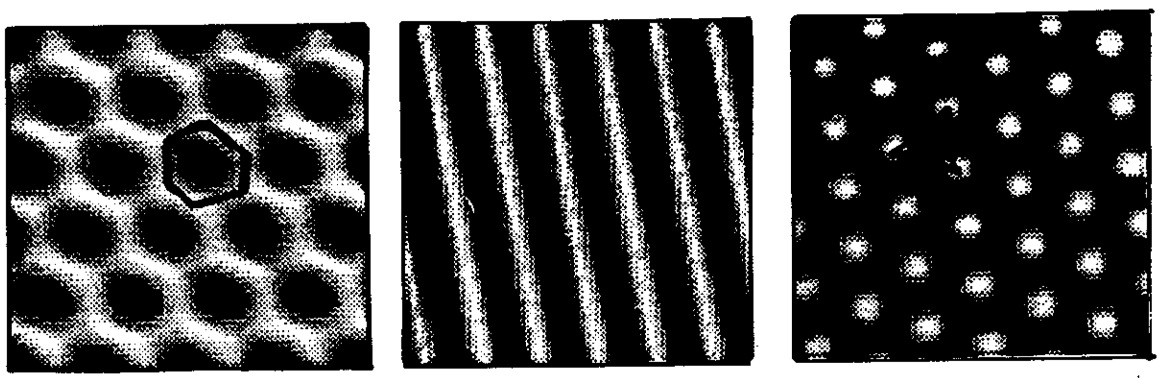


2D

$m = 1$ stripes
$m = 3$ hexagons



Non-symmetric systems



$H\pi$ $(v<0)$ Stripes HO $(v>0)$

Observed in several RD models

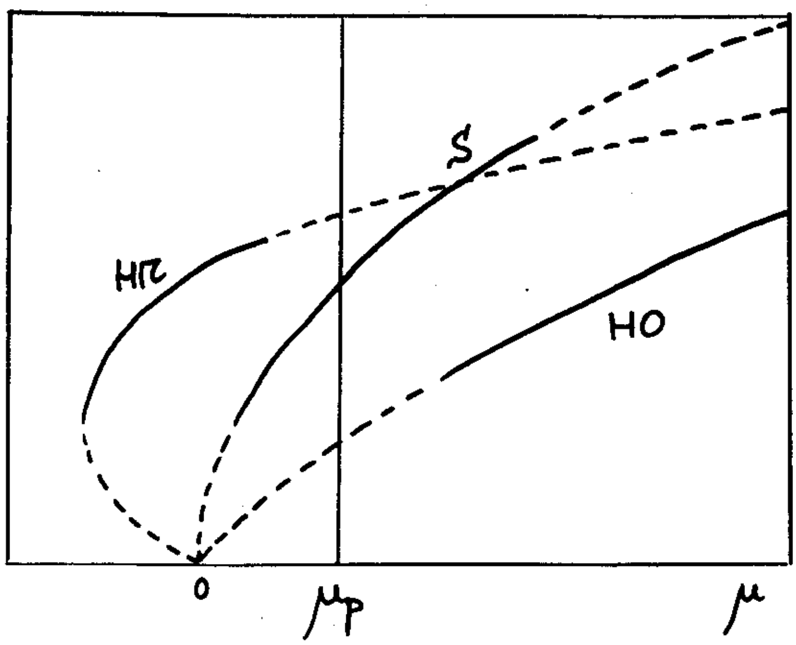
- Brusselator
- Schnakenberg
- Lengyel-Epstein

c) Large B regions

renormalization of v:

$$v' = v + \frac{2}{A}\mu = \frac{2}{A}\left[\frac{1-AM}{1+AM}\right] + \frac{2}{A}\mu$$

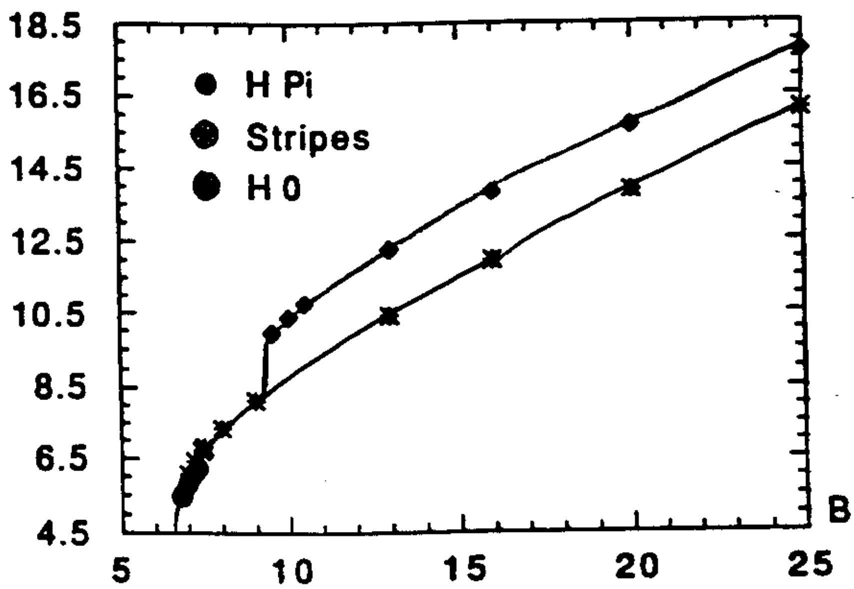
•) $v<0$: Modification of the bifurcation diagram because v' changes sign at μ_p



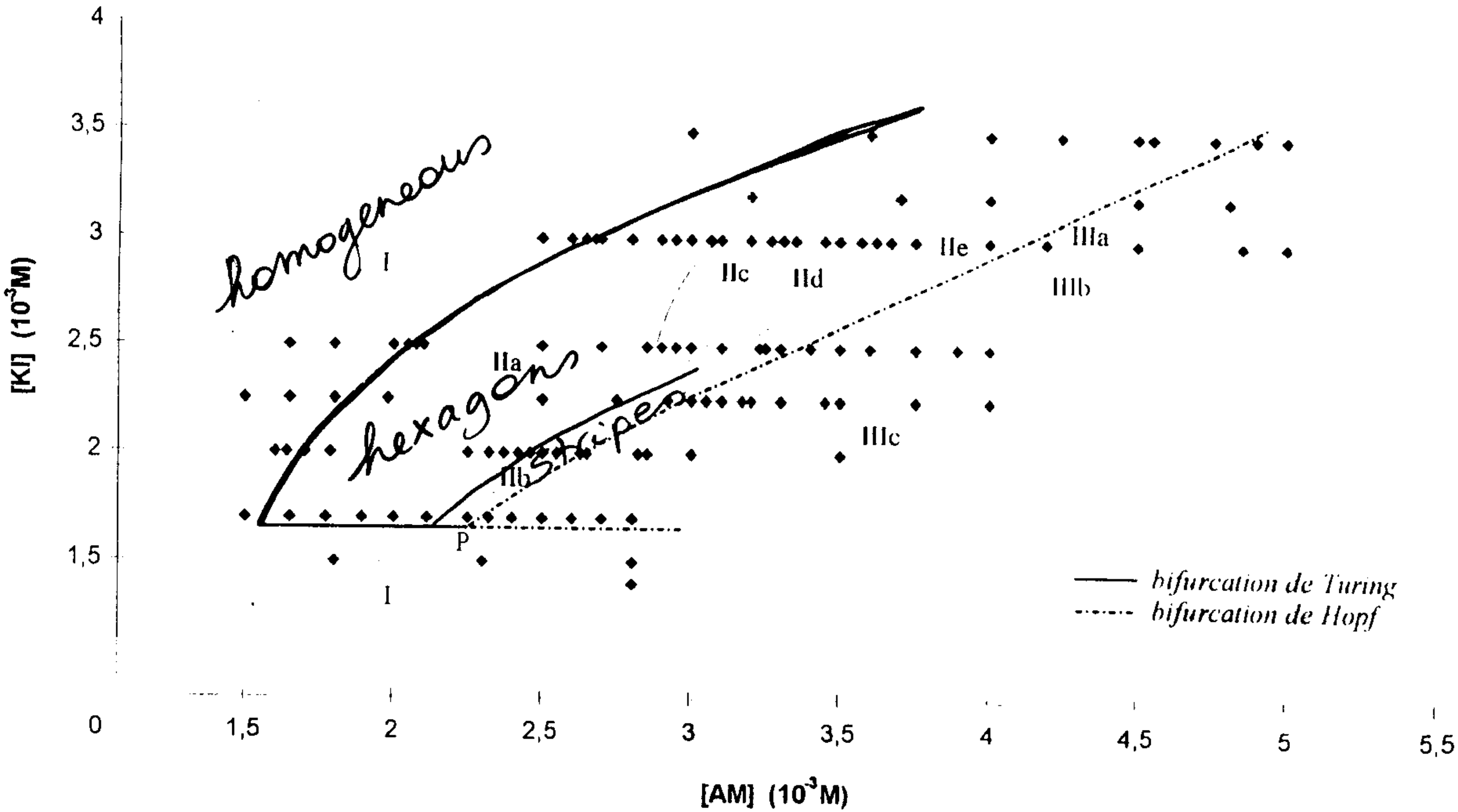
•) $v>0$: No important change HO, S

High Values of the control parameter

⇒ re-entrant hexagons



- Also obtained by V. Dufiet & J. Boissonade on the Schnackenberg model (J. Chem. Phys - to be published)
- E. Buzano et M. Golubitsky : Phil. Trans. R. Soc. London, A308, 617 (1983)



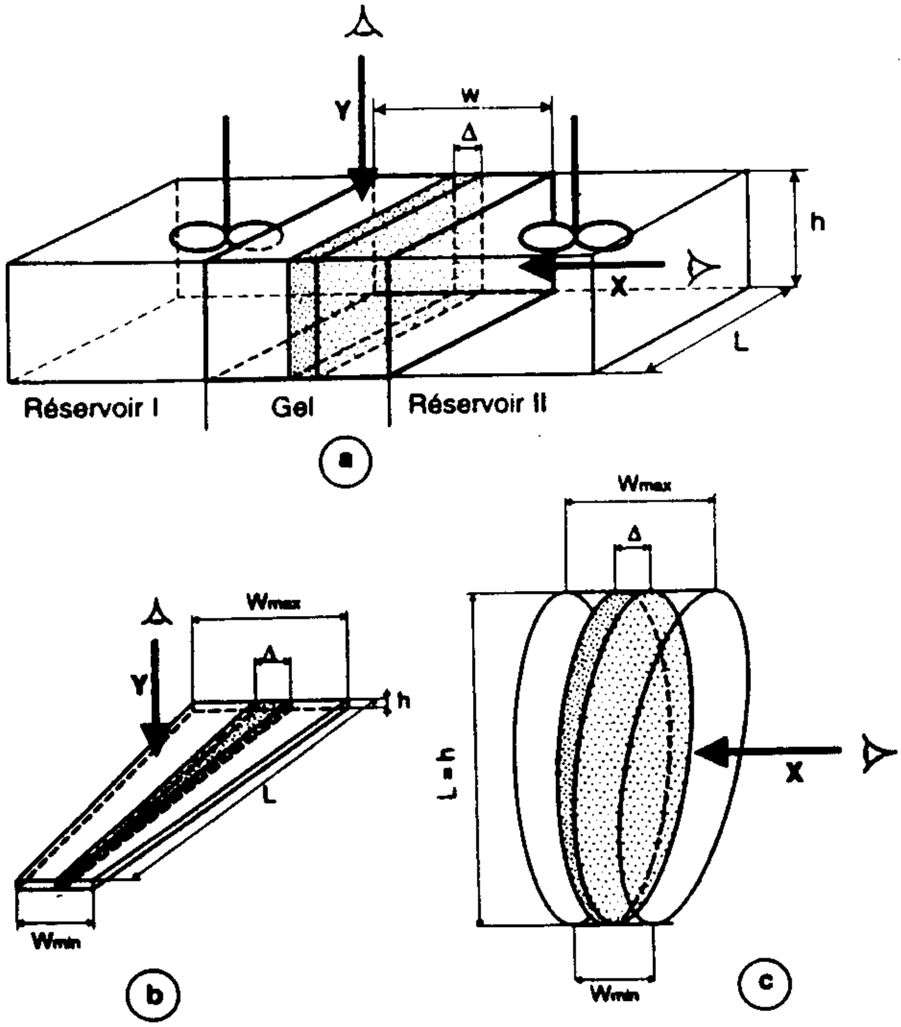
I *état homogène non-structuré*

II *structures de Turing* (IIa: *hexagones*, IIb: *bandes*, IIc: *triangles*, *IId: hexa-bandes*, *IIe: structures complexes)*

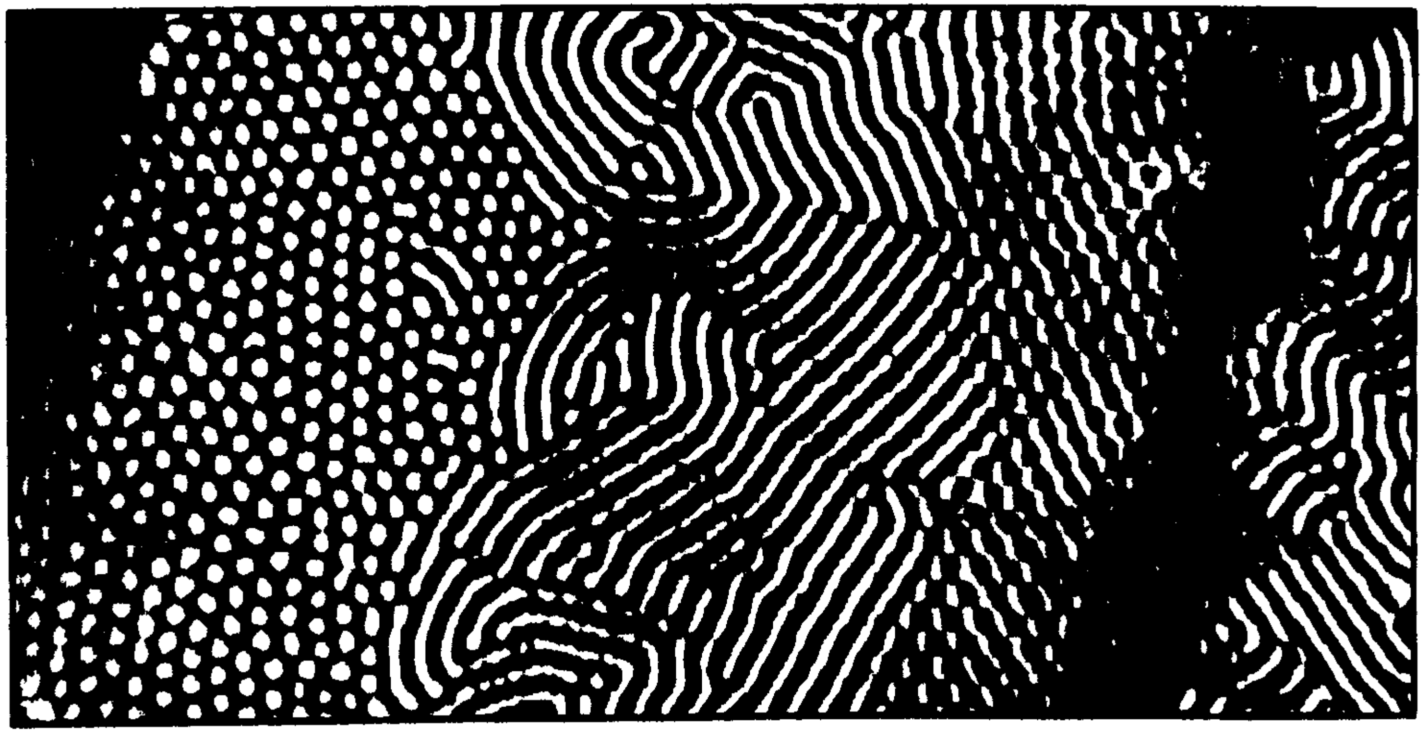
III *structures spatio-temporelles (IIIa: superposition Turing-Hopf, IIIb: interaction Turing-Hopf, IIIc: ondes)*

PhD thesis of Beata Rudovics

CIMA reaction


Y
w
Δ
h
X
L
Réservoir I
Gel
Réservoir II
a
Wmax
Δ
Y
h
L
Wmin
b
Wmax
Δ
L=h
X
Wmin
c

E. Dulos, P. Davies, B. Rudovics, P. De Kepper,
Physica D **98**, 53 (1996)



increase of the
control parameter