Spatiotemporal chaos and quasipatterns in coupled reaction–diffusion systems

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R., Silber & Skeldon (2012), *Phys. Rev. Lett.*, **108** 074504
Skeldon & R. (2015), *J. Fluid Mech.*, **777** 604–632
Castelino, Ratliff, R., Subramanian & Topaz (2020), *Physica D*, **209** 132475

Support from the EPSRC and Leverhulme Trust.

Workshop on PDEs describing far-from-equilibrium open systems, "Prague", September 2020

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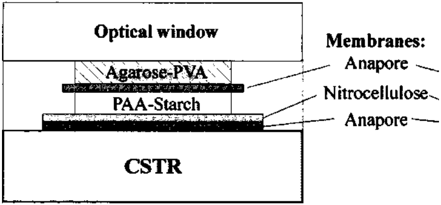
Pattern formation examples I

Patterns in animal coat markings: one length scale

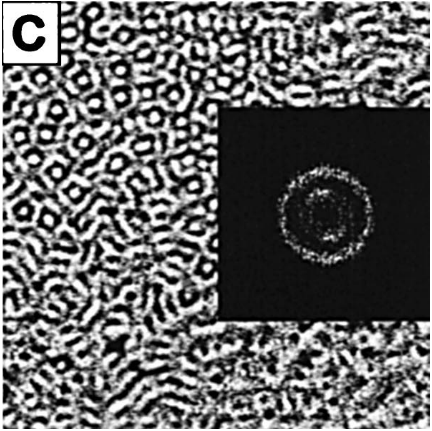
Pattern formation examples II

Patterns with two length scales:

Two-layer Turing (reaction–diffusion) patterns:



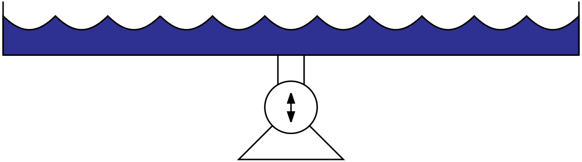
Patterns with different length-scales ($0.46\,\mathrm{mm}$ and $0.25\,\mathrm{mm}$) in the two layers are diffusively coupled. Chemistry: chlorine dioxide–iodine–malonic acid (CDIMA).

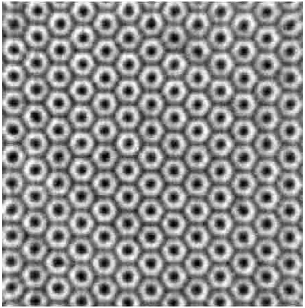


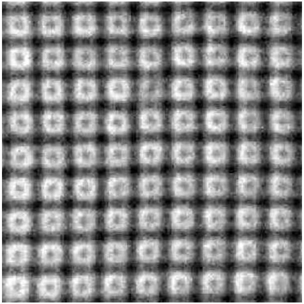
Berenstein et al. (2004)

Pattern formation examples III

Faraday (surface) wave experiment:







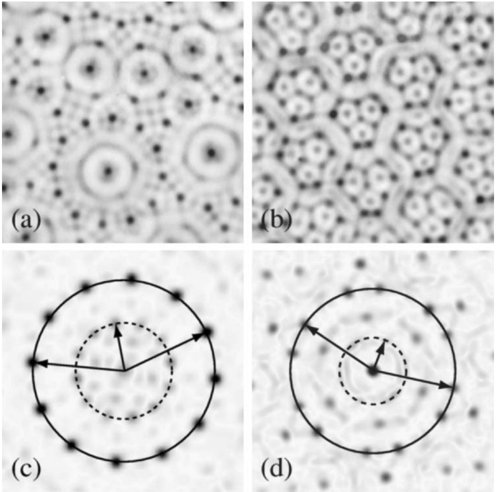
Arbell & Fineberg (2002)

Pattern formation examples IV

Left: 12-fold quasipattern. The two circles in Fourier space have radius ratio $0.52 \approx \frac{1}{2}(\sqrt{6} - \sqrt{2}) = 2\cos(75°)$

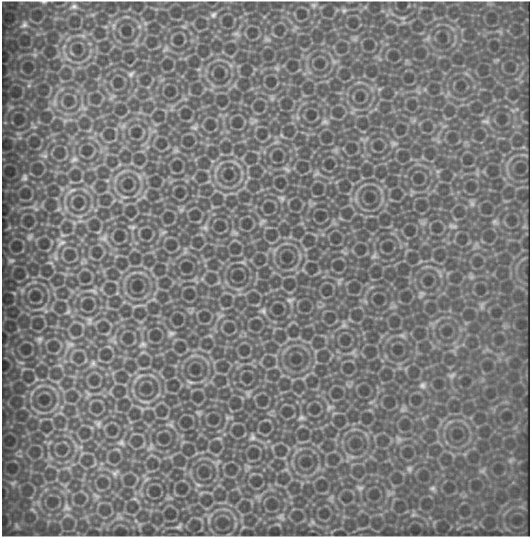
Right: superlattice pattern. The two circles in Fourier space have radius ratio $0.38 \approx 1/\sqrt{7}$

There are two length scales apparent in the Fourier power spectra.



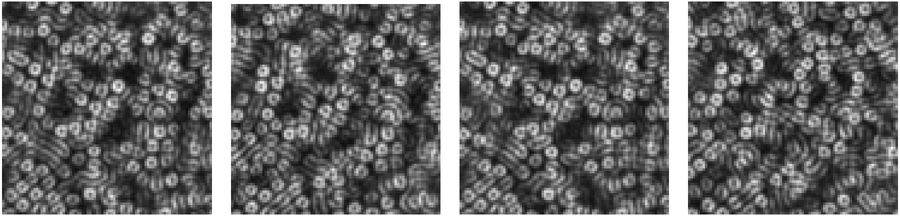
Ding & Umbanhowar (2006)

Pattern formation examples V



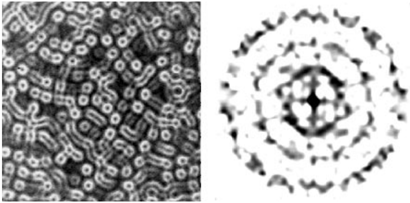
Kudrolli, Pier & Gollub (1998)

Pattern formation examples VI



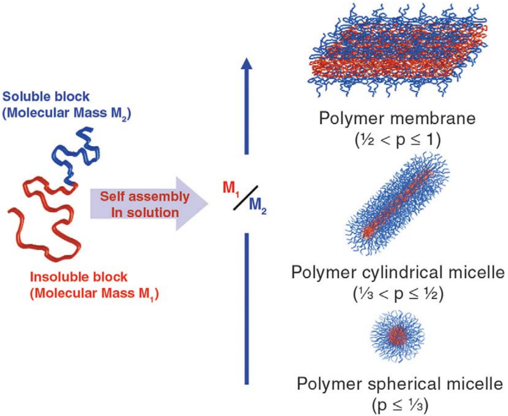
Epstein & Fineberg (2005)

Spatiotemporal chaos: "... continually evolving irregular domains of patterns with differing spatial orientations." (movie)



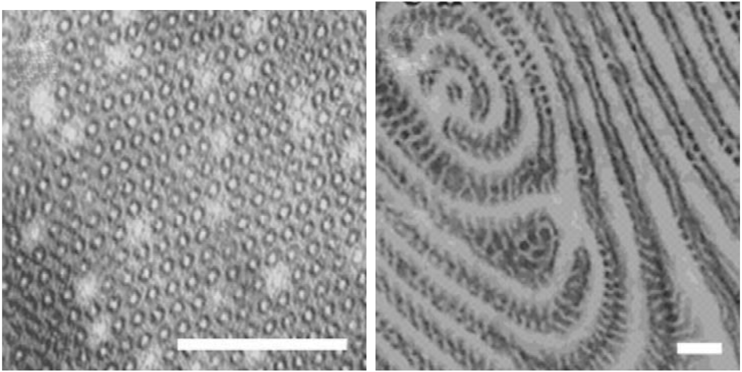
Pattern formation examples VII

Micelles formed from (for example) branched polymers or block co-polymers can make a stiff inner hydrophobic polymer core surrounded by a corona of hydrophillic polymer chains with a varying degree of flexibility.



Smart et al. (2008)

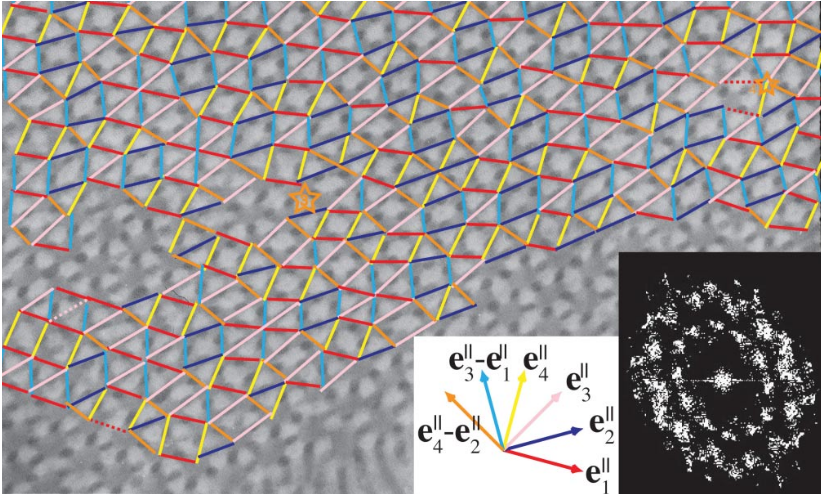
Pattern formation examples VIII



Smart et al. (2008)

Cubic micellar phase formed by poly(ethylene oxide)-poly(ethyl ethylene) in an epoxy network; disordered lamellar phase formed by poly(styrene)-block-poly(butadiene)-block-poly-(methyl methacrylate) in an epoxy network

Pattern formation examples IX



Hayashida et al. (2007)

Two-dimensional 12-fold quasicrystal formed by a polyisoprene/polystyrene/poly(2-vinylpyridine) star polymer.

Pattern formation examples X

Summary:

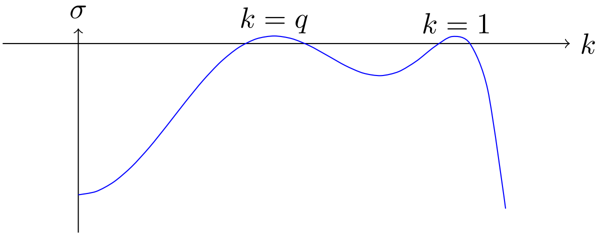
- Complex disordered patterns (Turing, soft matter)
- Spatiotemporal chaos (Faraday)
- Quasipatterns (Faraday, soft matter)

Common connection:

- Nonlinear interactions between modes with different length scales.

Two length scales: linear theory I

Consider waves with wavenumbers $k = 1$ and $k = q$ $(q < 1)$ becoming unstable, with growth rates μ and ν respectively:

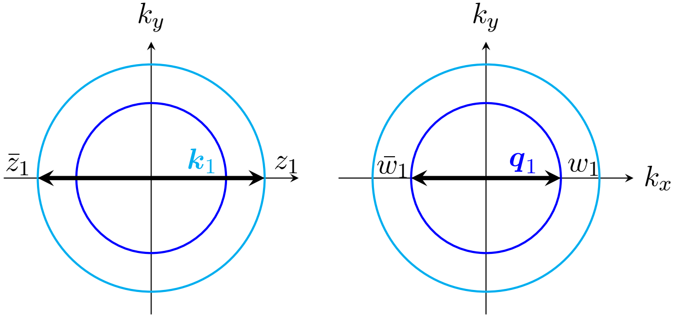


At onset, the pattern $U(x, y, t)$ will contain a combination of eigenfunctions: Fourier modes $e^{i\boldsymbol{k}\cdot\boldsymbol{x}}$ with $|\boldsymbol{k}| = q$ or $|\boldsymbol{k}| = 1$:

$$U = \sum_{\boldsymbol{q}_j} w_j(t) e^{i\boldsymbol{q}_j\cdot\boldsymbol{x}} + \sum_{\boldsymbol{k}_j} z_j(t) e^{i\boldsymbol{k}_j\cdot\boldsymbol{x}}$$

Two length scales: linear theory II

From the multitude, focus on one wave from each of the two circles: $z_1 e^{i\boldsymbol{k}_1\cdot\boldsymbol{x}}$ and $w_1 e^{i\boldsymbol{q}_1\cdot\boldsymbol{x}}$, as well as complex conjugates:

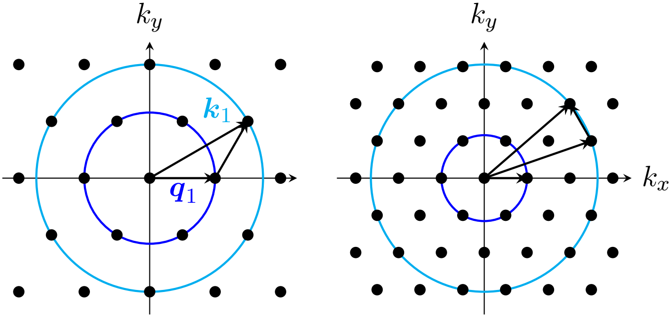


and the evolution of the amplitudes z_1 and w_1 will governed by:

$$\dot{z}_1 = \mu z_1, \qquad \dot{w}_1 = \nu w_1$$

Two length scales: choice of vectors I

At this stage, one would usually choose a set of wavevectors, appropriate for the pattern of interest:

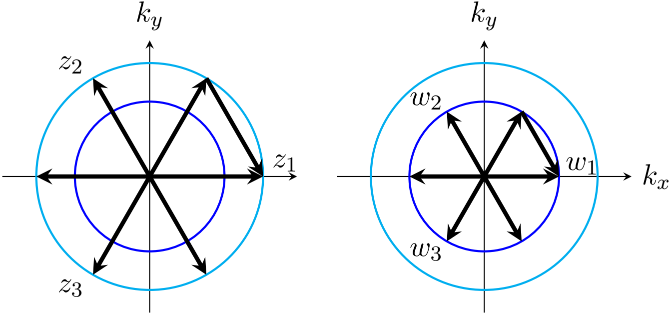


$q = 1/\sqrt{3}$ $\qquad$ $q = 1/\sqrt{7}$

While this is appropriate for $q < \frac{1}{2}$ (superlattice patterns), when $q > \frac{1}{2}$, the story is more interesting...

Two length scales: nonlinear theory I

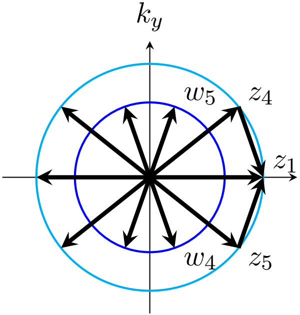
Products of waves lead to sums of wave vectors. Expanding in a power series in the small amplitude of the waves, at second order, there will be contributions from all possible three-wave interactions. The simplest interactions involve modes at 60°:

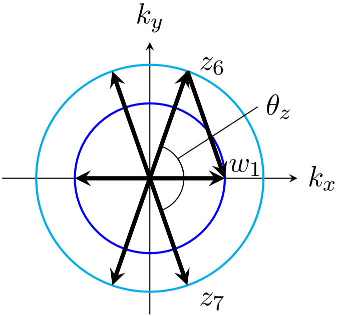


$$\dot{z}_1 = \mu z_1 + Q_{zh}\bar{z}_2\bar{z}_3, \qquad \dot{w}_1 = \nu w_1 + Q_{wh}\bar{w}_2\bar{w}_3$$

Two length scales: nonlinear theory II

Two waves on the outer circle can couple to a wave on the inner circle: $\boldsymbol{k}_6 + \boldsymbol{k}_7 = \boldsymbol{q}_1$, defining $\theta_z = 2\arccos(q/2)$.





$\dot{z}_1 = \cdots + Q_{zw}(z_4 w_4 + z_5 w_5), \qquad \dot{w}_1 = \cdots + Q_{zz} z_6 z_7$

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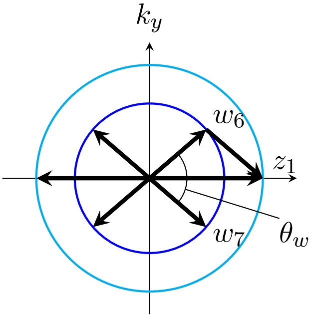
Two length scales: nonlinear theory III

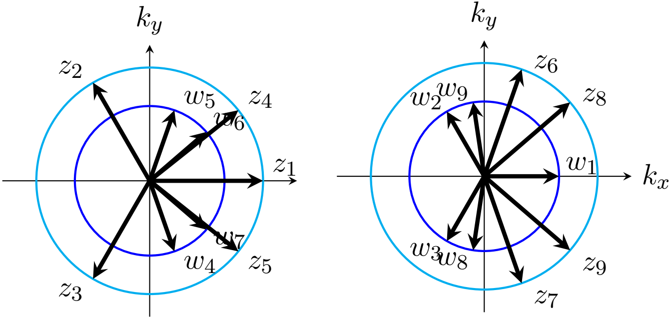
Two waves on the inner circle can couple to a wave on the outer, provided $q \geq \frac{1}{2}$: $\boldsymbol{q}_6 + \boldsymbol{q}_7 = \boldsymbol{k}_1$, defining $\theta_w = 2\arccos(1/2q)$.



$\dot{z}_1 = \cdots + Q_{ww} w_6 w_7, \qquad \dot{w}_1 = \cdots + Q_{wz}(w_8 z_8 + w_9 z_9)$

Two length scales: nonlinear theory IV

Putting it all together: there are 8 modes that couple to each of z_1 and w_1:

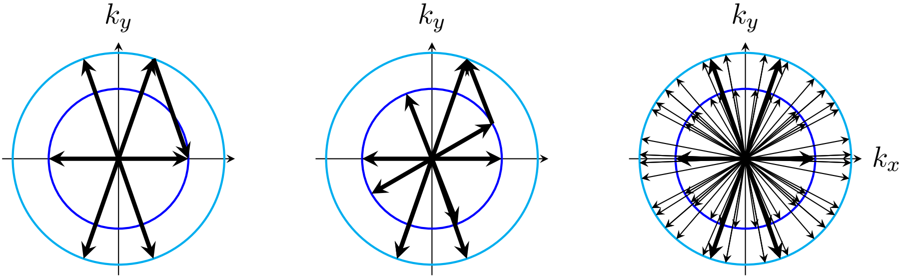


$$\dot{z}_1 = \mu z_1 + Q_{zh}\bar{z}_2\bar{z}_3 + Q_{zw}(z_4 w_4 + z_5 w_5) + Q_{ww} w_6 w_7,$$

$$\dot{w}_1 = \nu w_1 + Q_{wh}\bar{w}_2\bar{w}_3 + Q_{zz} z_6 z_7 + Q_{wz}(w_8 z_8 + w_9 z_9)$$

Two length scales: nonlinear theory V

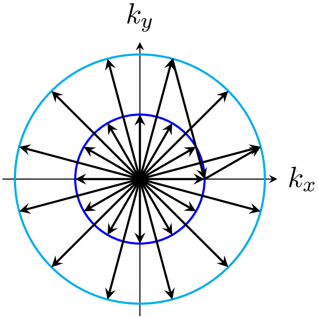
However, each z mode we've introduced couples to 8 other modes, and each w mode we've introduced couples to 8 other modes, and so on: an infinite number of modes can be generated:



Here, $q = 0.66$, $\theta_z = 141.4^\circ$, $\theta_w = 81.5^\circ$.

At cubic order, all modes couple to all other modes.

Two length scales: nonlinear theory VI

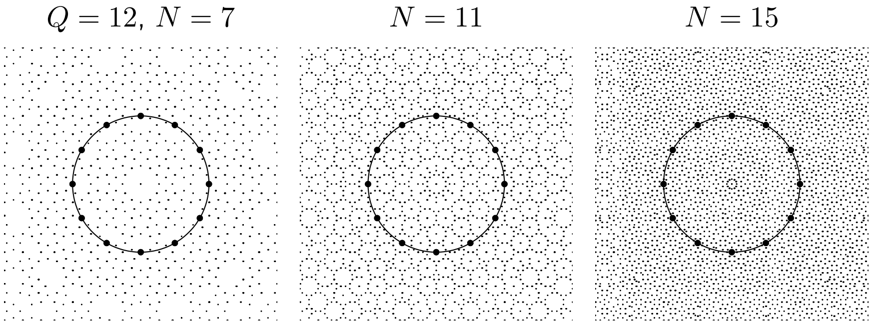


For $q = \frac{1}{2}(\sqrt{6} - \sqrt{2})$ ($\theta_z = 150^\circ$, $\theta_w = 30^\circ$), these interactions lead to a finite number of waves

This is the only q for which a finite number of waves will form a closed set under three-wave interaction in two dimensions, suggesting why 12-fold quasipatterns are the most common in 2D

Two length scales: nonlinear theory VII

However, these 12 vectors form a quasilattice:



which leads to the problem of small divisors:

- Standard results (e.g., Equivariant Branching Lemma) cannot be used
- Weakly nonlinear theory diverges, Nash–Moser Theorem needed
- See R & R (2003), R & Silber (2009), Iooss & R (2010), Braaksma *et al.* (2017), Iooss (2019)

Three-wave interactions I

How to make progress? Pull out one of the basic three-wave interactions, two outer vectors coupling to an inner:

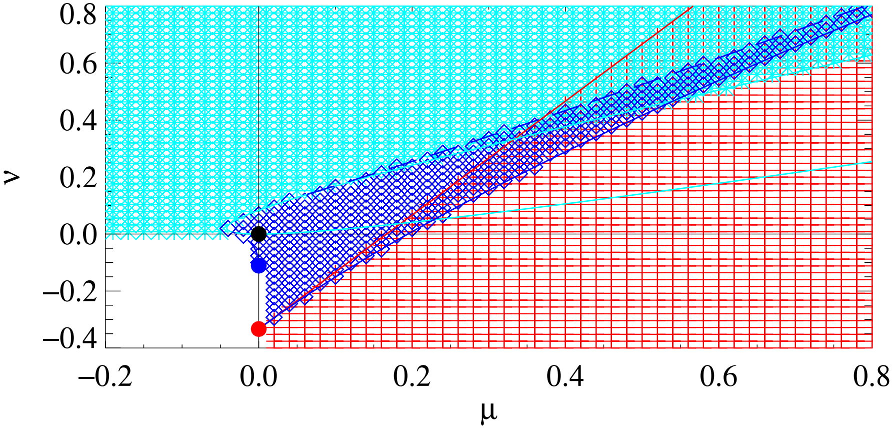
We illustrate using:

$$\begin{aligned}
\dot{z}_1 &= \mu z_1 + Q_{zw}\bar{z}_2 w_1 - (3|z_1|^2 + 6|z_2|^2 + 6|w_1|^2)z_1 \\
\dot{z}_2 &= \mu z_2 + Q_{zw}\bar{z}_1 w_1 - (6|z_1|^2 + 3|z_2|^2 + 6|w_1|^2)z_2 \\
\dot{w}_1 &= \nu w_1 + Q_{zz} z_1 z_2 - (6|z_1|^2 + 6|z_2|^2 + 3|w_1|^2)w_1
\end{aligned}$$

The outcome depends on the product of quadratic coefficients $Q_{zw}Q_{zz}$. Typically (Cf Porter & Silber 2004):

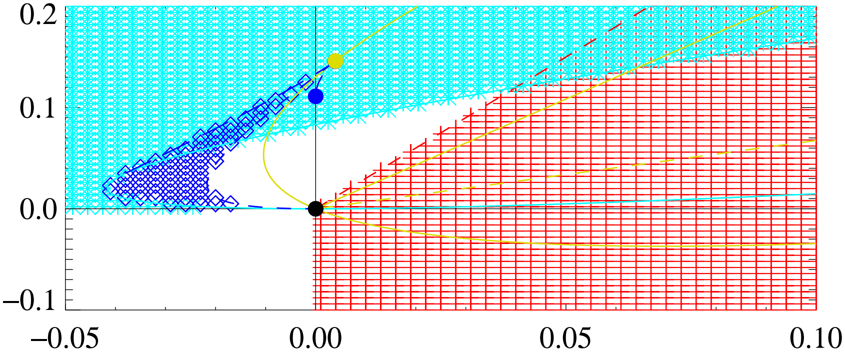
- Positive $Q_{zw}Q_{zz}$: stable steady stripes, or stable rhombs (mixed z and w);
- Negative $Q_{zw}Q_{zz}$: stable steady stripes or rhombs, or time-dependent competition between z and w modes.
- Same conclusion for any of the three-wave interactions.

Three-wave interactions II



Positive $Q_{zw}Q_{zz}$: stable steady z (red) or w (cyan) stripes, or stable rhombs (blue), which are mixed z and w.

Three-wave interactions III



Negative $Q_{zw}Q_{zz}$: stable steady z or w stripes, some stable rhombs (blue), or time-dependent competition between z and w modes (empty area). (Cf Porter & Silber 2004.)

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Three-wave interactions IV

With multiple three-wave interactions, we hypothesise, with $q > \frac{1}{2}$:

- We expect to find steady complex patterns or spatiotemporal chaos, according to the signs of $Q_{zw}Q_{zz}$ and $Q_{wz}Q_{zz}$.
- If $Q_{zw}Q_{zz}$ and $Q_{wz}Q_{zz}$ are both negative, we expect to see greater time dependence.
- With $q = \frac{1}{2}(\sqrt{6} - \sqrt{2}) = 0.5176$ we expect steady or time-dependent 12-fold quasipatterns, according to the signs of $Q_{zw}Q_{zz}$ and $Q_{wz}Q_{zz}$.

and with $q < \frac{1}{2}$:

- We expect to find steady complex patterns or spatiotemporal chaos, according to the sign of $Q_{zw}Q_{zz}$.

Examples of this “rule of thumb”: Two-layer Turing (reaction–diffusion) patterns (Castelino et al., 2020), Faraday waves (R & Skeldon 2015), soft matter crystalisation (Subramanian, Archer, Knobloch, Ratliff & R, 2013–2020), model PDE (R, Silber & Skeldon 2012), . . .

Two-layer Turing patterns I

The Brusselator is a simple example of a Turing (reaction–diffusion) system:

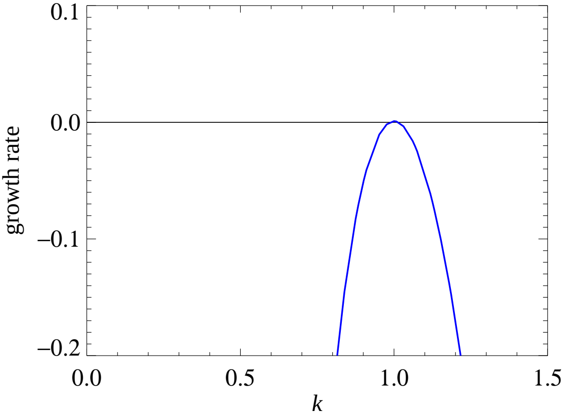
$$\frac{\partial U}{\partial t} = (B-1)U + A^2V + D_U\nabla^2 U + \frac{B}{A}U^2 + 2AUV + U^2V,$$
$$\frac{\partial V}{\partial t} = -BU - A^2V + D_V\nabla^2 V - \frac{B}{A}U^2 - 2AUV - U^2V,$$

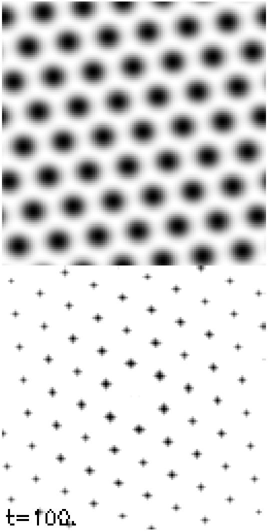
where:

- $U(x, y, t)$ and $V(x, y, t)$ represent chemical concentrations
- A and B are parameters ($A = 3$ and $B = 9$)
- D_U and D_V are diffusion constants
- Hopf ($k = 0$) and pitchfork ($k \neq 0$) instabilities are possible
- The usual nontrivial equilibrium has been moved to the origin
- Link: Castelino et al., 2020

Two-layer Turing patterns II

Typical Turing pattern: $D_U = 1.99833$ and $D_V = 4.50875$, 8×8 box





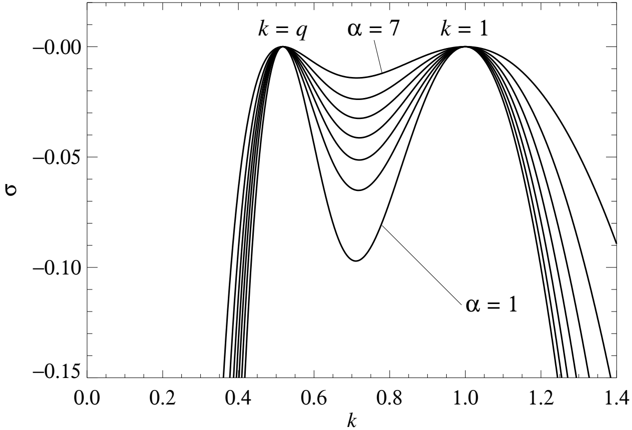
Two-layer Turing patterns III

Two layer model (Yang *et al.* 2002, Catlla *et al.* 2012):

$$
\begin{aligned}
\frac{\partial U_1}{\partial t} &= (B-1)U_1 + A^2 V_1 + D_{U_1}\nabla^2 U_1 + \alpha(U_2 - U_1) + \text{NLT},\\
\frac{\partial V_1}{\partial t} &= -BU_1 - A^2 V_1 + D_{V_1}\nabla^2 V_1 + \beta(V_2 - V_1) + \text{NLT},\\
\frac{\partial U_2}{\partial t} &= (B-1)U_2 + A^2 V_2 + D_{U_2}\nabla^2 U_2 + \alpha(U_2 - U_1) + \text{NLT},\\
\frac{\partial V_2}{\partial t} &= -BU_2 - A^2 V_2 + D_{V_2}\nabla^2 V_2 + \beta(V_2 - V_1) + \text{NLT},
\end{aligned}
$$

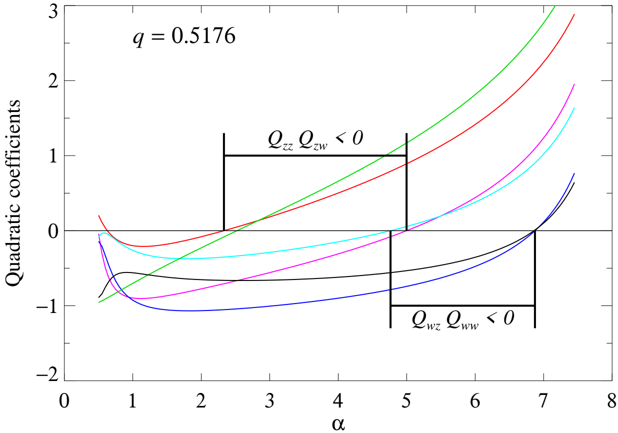
- $U_{1,2}$ and $V_{1,2}$ are concentrations in each layer
- Same A and B and nonlinear terms (NLT) as before
- The diffusion coefficients are not the same in each layer
- The α and β terms couple the two layers
- Linear theory: 4×4 matrix, solve for the D's

Two-layer Turing patterns IV



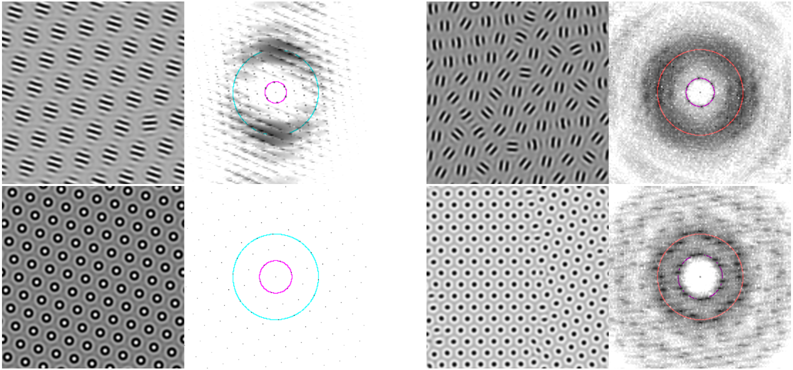
Dispersion relation: the largest eigenvalue $\sigma(k)$, for $q = \sqrt{2-\sqrt{3}} = 0.5176$, $\beta = 1$, and $\alpha = 1, 2, \ldots, 7$.

Two-layer Turing patterns V



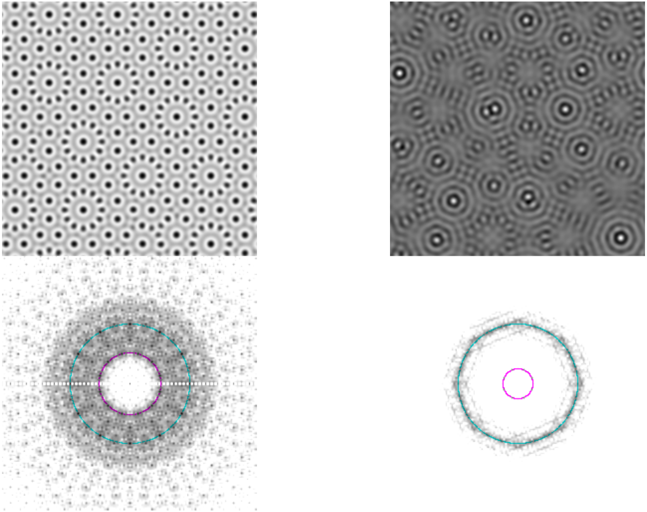
Weakly nonlinear theory for $q = 0.5176$ and $\beta = 1$: Q_{zh} (green), Q_{wh} (black), Q_{zz} (red), Q_{zw} (magenta), Q_{wz} (cyan), Q_{ww} (blue), with $Q_{zw}Q_{zz} < 0$ for $2.3 < \alpha < 5.0$, and $Q_{wz}Q_{ww} < 0$ for $4.8 < \alpha < 6.9$.

Two-layer Turing patterns: steady I



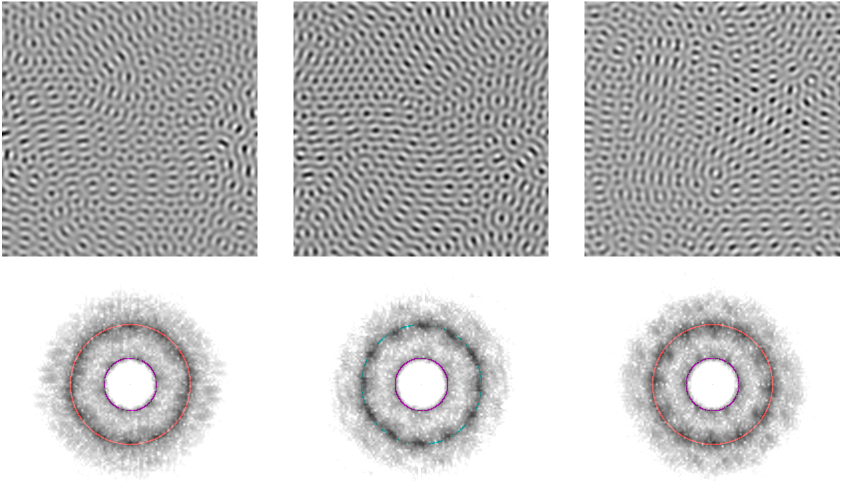
Steady patterns: $q = 0.2500,\ 0.3300,\ 0.3780,\ 0.5176$, with $\alpha = \beta = 1$, all quadratic coefficients the same sign, linear growth rates $\mu = \nu = 0.01/\sqrt{2}$, 30×30 domain with periodic boundary conditions. One chemical field is shown along with its power spectrum.

Two-layer Turing patterns: steady II



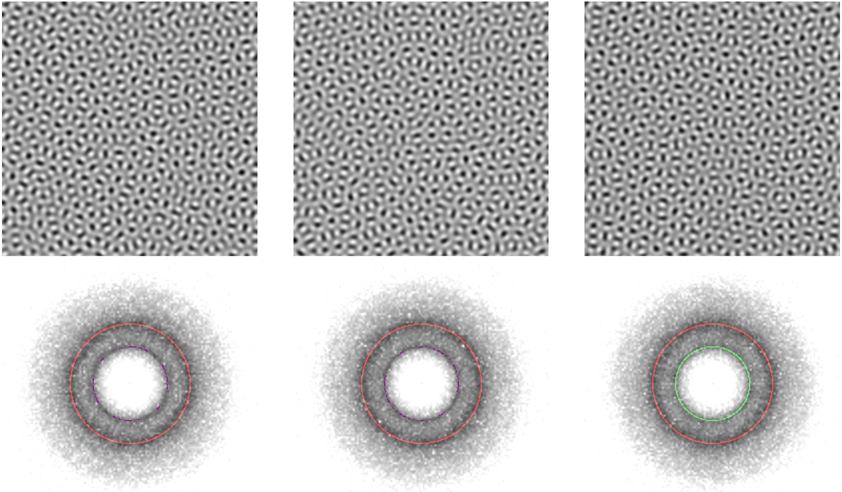
Steady quasipattern approximants: $q = 0.5176$ (12-fold), 0.2500 (8-fold), with $\alpha = \beta = 1$, different choices of linear growth rates.

Two-layer Turing patterns: time dependent I



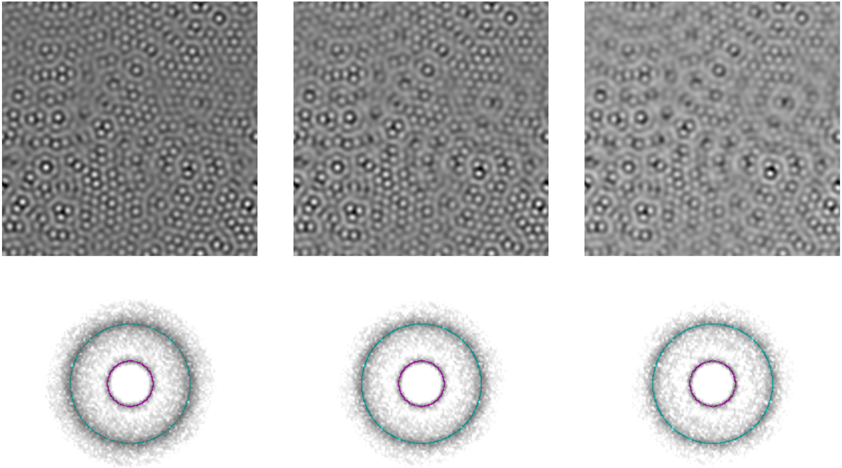
Spatio-temporal chaos: $q = 0.4400$, $\alpha = 2$, $\beta = 1$, $Q_{zw}Q_{zz} < 0$. In an 8×8 domain, the dynamics is much simpler. Links: paper, movies.

Two-layer Turing patterns: time dependent II



Spatio-temporal chaos: $q = 0.6180$, $\alpha = 3$, $\beta = 1$, $Q_{zw}Q_{zz} < 0$, $Q_{wz}Q_{ww} > 0$.

Two-layer Turing patterns: time dependent III



Spatio-temporal chaos: $q = 0.3780$, $\alpha = 3$, $\beta = 1$, $Q_{zw}Q_{zz} < 0$, larger linear growth rates.

Conclusions

- If the ratio of wavenumbers q is between $\frac{1}{2}$ and 1, mode interactions in both directions must be taken in to account.
- Most values of q in this range lead to the possibility of generating an infinite number of interacting waves. The exception is $q = 0.5176$, associated with 10- and 12-fold quasipatterns.
- Even $q < \frac{1}{2}$, with only a single direction of mode interactions, turns out to produce interesting patterns.
- The outcome of the mode interactions will be influenced by the signs of the quadratic coefficients, with time-dependence (and spatiotemporal chaos) most likely in the case of (both pairs of) quadratic coefficients with opposite sign.
- Large domains are needed to see spatiotemporal chaos.
- Steady patterns with an "infinite" set of wavevectors are elusive.
- Is this mechanism responsible for complex behaviours seen in experiments?
- Further work in progress.