Coherent Structures Emerging from Turbulence in the Nonlocal Complex Ginzburg-Landau Equation

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Outline

- Model of a spatially extended electrochemical system with nonlocal coupling

- A general mathematical model: The Nonlocal Complex Ginzburg-Landau Equation (NCGLE)

- Investigation of found patterns in real/Fourier space

\[
\frac{\partial_t W}{=}?
\]
Basic experimental setup
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\[ I_{\text{react}} = f(\phi_{DL}) \]
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Local charge balance

\[
C \frac{\partial \phi_{DL}}{\partial t} = -i_F - \sigma \frac{\partial \phi}{\partial z} \bigg|_{z=z_{WE}}
\]

\(\phi_{DL}\): potential drop across the interface

\(\phi\): potential in the electrolyte

\(\Delta \phi = 0\)

double layer

electrolyte

WE

CE
Geometry of the experimental setup

With a 1D ring electrode, the Laplace equation must only be solved in two dimensions:

$$\Delta \phi(x, z) = 0$$

with the boundary conditions

$$\phi(x, 0) = U - \phi_{DL}(x)$$
$$\phi(x, w) = 0$$

where \( \phi(x, z) \) is periodic in \( x \):

$$\phi(x + L, z) = \phi(x, z)$$

Nonlocality depends on the aspect ratio \( \beta = \frac{w}{L} \) is of the cell.
Derivation of the nonlocal coupling term

$\phi(x, z)$ can be obtained from $\phi_{DL}$ by using a Green function

$$
\phi(x, z) = \int_{WE} G(x - x', z)(U - \phi_{DL}(x'))dx'
$$

where $G(x - x', z)$ depends on the geometry.
Derivation of the nonlocal coupling term

\( \phi(x, z) \) can be obtained from \( \phi_{DL} \) by using a Green function

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\phi(x, z) = \int_{WE} G(x - x', z)(U - \phi_{DL}(x'))dx'
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where \( G(x - x', z) \) depends on the geometry. Inserting \( \phi(x, y) \) into

\[
C \frac{\partial \phi_{DL}}{\partial t} = -i_F - \sigma \frac{\partial \phi}{\partial z} \bigg|_{z = z_{WE}}
\]

and separating uniform and non-uniform terms yields:

\[
C \frac{\partial \phi_{DL}}{\partial t} = f(\phi_{DL}, c) - \sigma \int_{WE} H_\beta(|x - x'|) [\phi_{DL}(x') - \phi_{DL}(x)] dx'
\]
Derivation of the nonlocal coupling term

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\]

where

\[
H_\beta(|x - x'|) = \frac{\pi}{4 \beta^2 \sinh^2 \left( \frac{\pi |x - x'|}{2 \beta} \right)} + \frac{\delta(|x - x'|)}{\beta}
\]
Experiments\textsuperscript{1}: H$_2$-oxidation in the presence of poisons

Global current (upper part) and spatiotemporal evolution of $\phi_{DL}$ for three different external voltages.

Phase(a) and amplitude(b) representations of spatiotemporal data for local coupling exhibiting \textit{defect turbulence}.

Defect density over voltage for small ("local") and large ("nonlocal") coupling range.

\textsuperscript{1}[H. Varela, C. Beta, A. Bonnefont, K. Krischer PHYS. REV. LETT. 94 174104 (2005)]
The complex Ginzburg-Landau equation (CGLE)

Turbulence can be described nicely in the framework of the complex Ginzburg-Landau Equation

\[ \partial_t W = W + (1 + ic_1) \partial_x^2 W - (1 + ic_2)|W|^2W \]

- general model for reaction-diffusion systems near a supercritical Hopf-Bifurcation (permitting stable oscillations).
- the complex quantity \( W(x, t) = |W|(x, t)e^{i\phi(x,t)} \) describes amplitude and phase of the oscillations near the Hopf bifurcation
- \( c_1 \) and \( c_2 \) depend on the original reaction-diffusion system
Turbulence in the CGLE

Simulation taken from David Winterbottom's site: http://codeinthehole.com/tutorials/cgl/images/defturbabs.jpg
The nonlocal complex Ginzburg-Landau equation (NCGLE)

The CGLE is not applicable to a non-diffusive, nonlocal spatial coupling:

\[ \partial_t C \phi_{DL} = f(\phi_{DL}, c) - \sigma \int_{WE} H_\beta(|x - x'|) [\phi_{DL}(x') - \phi_{DL}(x)] \, dx' \]

\[ \partial_t c = g(\phi_{DL}, c) \]

\[2\] [V. García-Morales, K. Krischer, PHYS. REV. LETT. 100, 054101 (2008)]
The nonlocal complex Ginzburg-Landau equation (NCGLE)\(^2\)

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\[
\partial_t C\phi_{DL} = f(\phi_{DL}, c) - \sigma \int_{WE} H_{\beta}(|x - x'|) [\phi_{DL}(x') - \phi_{DL}(x)] \, dx'
\]

\[
\partial_t c = g(\phi_{DL}, c)
\]

However, near the onset of oscillations, the system can be mapped to:

**NCGLE**

\[
\partial_t W = W + (1 + ic_1) \int_{WE} H_{\beta}(|x - x'|) [W(x') - W(x)] \, dx' - (1 + ic_2)|W|^2W
\]

- nonlocal integral operator replaces local \(\partial_x^2\)
- additional parameter \(\beta\) adjusts the coupling range
- for \(\beta \rightarrow 0\), the NCGLE becomes the CGLE

\(^2\) [V. García-Morales, K. Krischer, PHYS. REV. LETT. 100, 054101 (2008)]
Defect turbulence in the NCGLE

Left: $|W|$ in the turbulent regime of the CGLE ($c_1 = -3$, $c_2 = 1$) for $\beta = 0.2$. Right: $|W|$ in the turbulent regime for $\beta = 1.0$ Blue means small amplitude $|W|$, red means high $|W|$. 

large coupling range $\Leftrightarrow$ small defect density
Spatiotemporal patterns observed in the NCGLE

patterns in the turbulent regime of the CGLE \((c_1 = -3, c_2 = 1)\), for a large coupling range \(\beta\), after a transient of 500s (initial condition was a delta peak).
Spectral decomposition of the NCGLE

Inserting the Fourier decomposition of $W$

$$W(x, t) = \sum_{n=-\infty}^{\infty} W_n(t) e^{i2\pi nx/L}$$

in the NCGLE

$$\partial_t W = W + (1 + ic_1) \int_{WE} H_\beta(|x - x'|) [W(x') - W(x)] dx' - (1 + ic_2)|W|^2 W.$$

and comparing coefficients yields:

**NCGLE in Fourier space**

$$\partial_t W_n = \left(1 + (1 + ic_1) \left[-\frac{2\pi n}{L} \coth \left(\frac{2\pi n \beta}{L}\right) + \frac{1}{\beta}\right]\right) W_n$$

$$- (1 + ic_2) \sum_{j-k+l=n} W_j W_k W_l$$
Patterns with $D_2$ symmetry

$W_n = 0$ for odd $n$,
$W_n = W_{-n}$ for even $n$
Patterns with alternating symmetries

In $\mathbb{Z}_2$: $W_n = W_{-n}$ for odd $n$,

In $\tilde{\mathbb{Z}}_2$: $W_n = -W_{-n}$ for odd $n$
Phase decoupling

Phase decoupling reduces the homogeneous oscillation to a steady state:

\[ W_n = w_n e^{i \phi(t)} \]

where \( \phi(t) \) is the phase of the homogeneous mode (therefore \( w_0 \) is real). For the time evolution of \( w_n \) we obtain:

\[ \dot{w}_n = \left( 1 + (1 + ic_1) \left[ \frac{1}{\beta} - \frac{2\pi n}{L} \coth \left( \frac{2\pi n\beta}{L} \right) \right] \right) w_n - (1 + ic_2) \sum_{j-k+l=n} w_j \overline{w}_k w_l - i \dot{\phi} w_n \]

with

\[ \dot{\phi} = \Im \left( -\frac{1 + ic_2}{w_0} + \sum_{j-k+l=0} w_j \overline{w}_k w_l \right) \]
Bifurcation analysis in a $D_2$-symmetric subspace

Five variables: $a_{-2}, b_{-2}, a_0, a_2, b_2$
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Bifurcation analysis in a $\mathbb{Z}_2$-symmetric subspace

Nine variables: $a_{-2}, b_{-2}, a_{-1}, b_{-1}, a_0, a_1, b_1, a_2, b_2$

In the $\mathbb{Z}_2$ subspace, $\xi_1$ is unstable and $\xi_2$ is stable.
In the $\tilde{\mathbb{Z}}_2$, $\xi_1$ is stable and $\xi_2$ is unstable.
Heteroclinic connection of stationary states

There are four paths connecting $\xi_1$ and $\xi_2$ which seem to be chosen erratically. After the Hopf bifurcation, this scenario becomes a heteroclinic connection of limit cycles.
Summary

- Limit cycle
- Heteroclinic connection of limit cycles
- Heteroclinic connection of fixed points
- Two Hopf bifurcations
- Double saddle loop bifurcation

Hölzel, García Morales, Krischer (TUM)
Conclusions

- Spatiotemporal pattern formation in electrochemical systems can be modelled by the NCGLE near the onset of oscillations.
- Both turbulent and coherent behavior can be described in this general frame.
- Complex dynamical behavior of the full model is governed by the dynamics and bifurcations in low dimensional subsystems.
Location of heteroclinic orbits in $c_1$-$c_2$-space