Chirality in soft matter: from out-of-equilibrium physics to non-linear optics

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Univerza v Ljubljani
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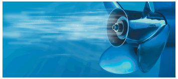
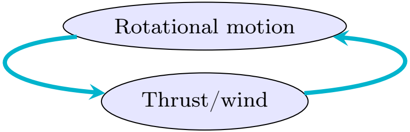
MARIE CURIE ACTIONS

Outline

Chirality in everyday life

- Chiral object: distinguishable from its mirror image.
- A common example: propeller.





- Without chirality, this conversion is not possible.

Chirality in soft matter: the cholesteric phase

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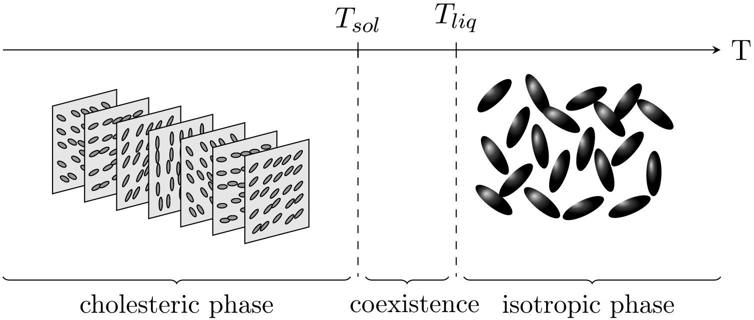
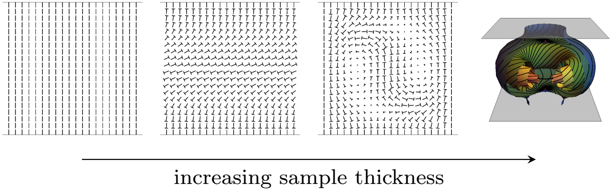


Confining cholesterics between two plates

- Surface constraint: molecules must be normal to the confining surface



P. J. Ackerman et al. *Scientific Reports*, 2, 2012

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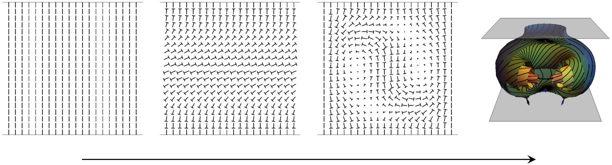
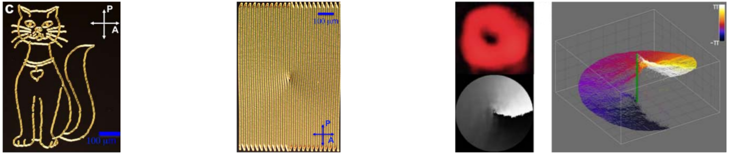


increasing sample thickness

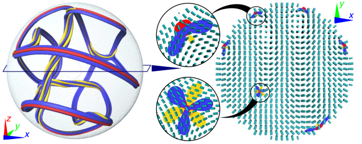
- Arbitrary shapes can be written!



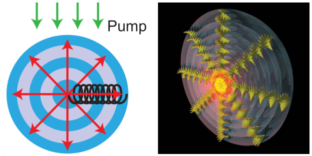
P. J. Ackerman et al. *Scientific Reports*, 2, 2012

Confining cholesterics inside droplets

Topological zoo of free standing knots



Lasing in a cholesteric droplet: an omnidirectional microscopic coherent light source

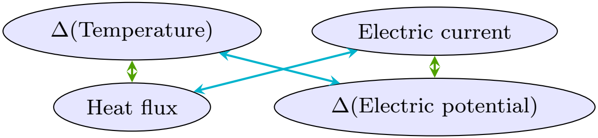


D. Seč, S. Čopar, and S. Žumer. *Nature Communications*, 5:3057, 2014

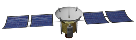
M. Humar. *Liquid Crystals*, 43:1937–1950, 2016

Other aspects of chirality in soft matter

Cross-coupling effects in out-of-equilibrium systems:



Applications:

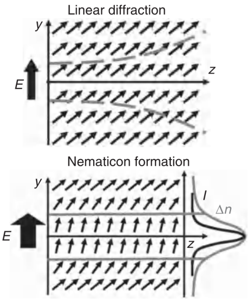



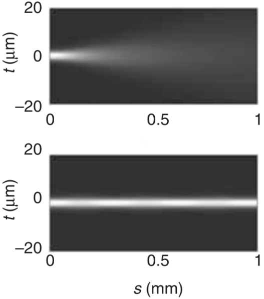
Problematic

Role of chirality in confined liquid-crystal systems submitted to a temperature gradient?

Other aspects of chirality in soft matter

Non-linear optical response of liquid crystal systems:





Increasing beam power

G. Assanto. *Nematicons.* John Wiley & Sons, 2013

Problematic

Role of chirality in the non-linear optical response of a confined cholesteric?

Outline

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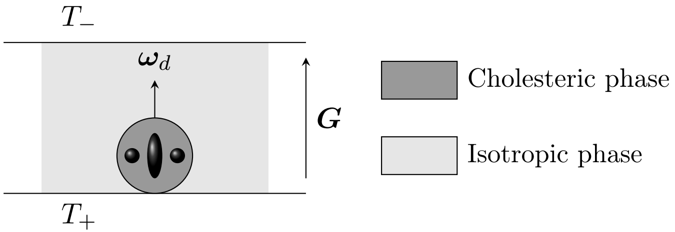
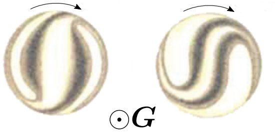
First observations by Lehmann





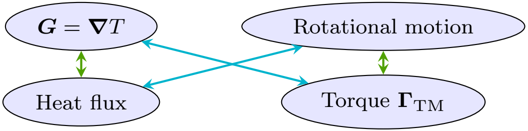
Lehmann, 1900:

- coexistence of cholesteric droplets with the isotropic fluid
- rotation of the droplets internal texture when heated from below

O. Lehmann. *Annalen der Physik*, 307:649–705, 1900

Leslie interpretation of the Lehmann experiment

First explanation by Leslie in 1968:



- Existence, in a cholesteric phase, of a torque on the director: $\boldsymbol{\Gamma}_{\mathrm{TM}} = \nu \, \boldsymbol{n} \times [\boldsymbol{n} \times \boldsymbol{G}]$, with ν the Leslie thermomechanical coefficient.

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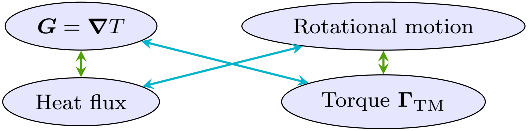

- Existence, in a cholesteric phase, of a torque on the director: $\boldsymbol{\Gamma}_{\mathrm{TM}} = \nu\, \boldsymbol{n} \times [\boldsymbol{n} \times \boldsymbol{G}]$, with ν the Leslie thermomechanical coefficient.
- As in a wind turbine, essential role of the chirality: no rotation predicted in a nematic phase.

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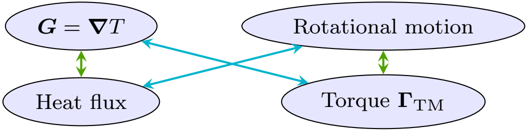


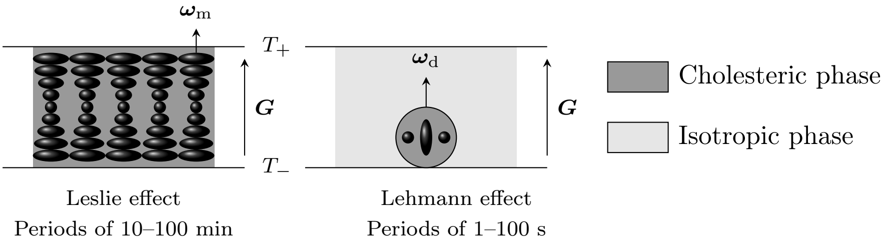
Leslie paradigm

The rotation of the texture in the Lehmann experiment is due to the Leslie thermomechanical torque $\boldsymbol{\Gamma}_{\mathrm{TM}}$

F. M. Leslie. *Proceedings of the Royal Society A*, 307:359–372, 1968

Lehmann vs. Leslie experiment

Oswald & Dequidt, 2008-2014:



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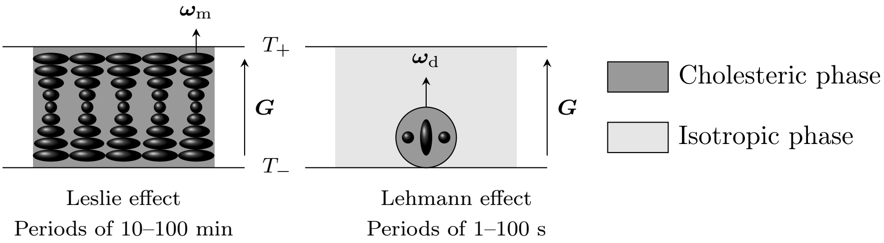

ω_d and ω_m sometimes of opposite signs!

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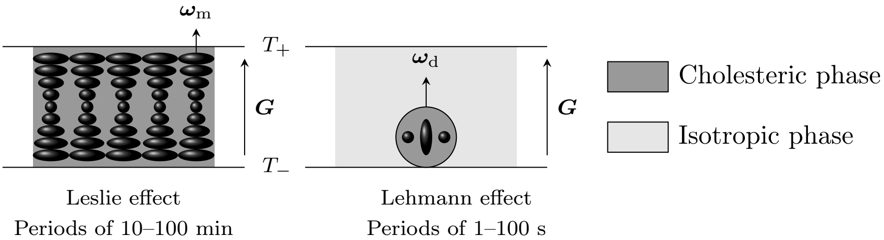

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Leslie effect $\neq$ Lehmann effect?

P. Oswald and A. Dequidt. *Physical Review Letters*, 100:217802, 2008

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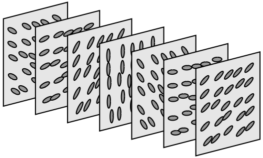
Rotation because of the microscopic or macroscopic chirality?

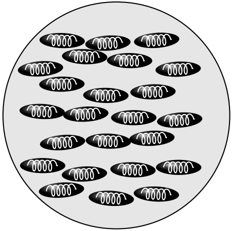
- microscopic chirality ⇔ chiral molecules

- macroscopic chirality ⇔ twisted texture (helix in at least one direction)



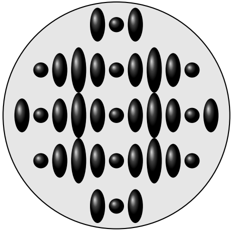
What is causing the rotation of the Lehmann droplets

Possible tests:



{ chiral molecules ↔ cholesteric
no macroscopic twist (compensated)

Thermal gradient ⇒ no rotation



{ no chiral molecules ↔ nematic
macroscopic twist

Thermal gradient ⇒ rotation?

Question

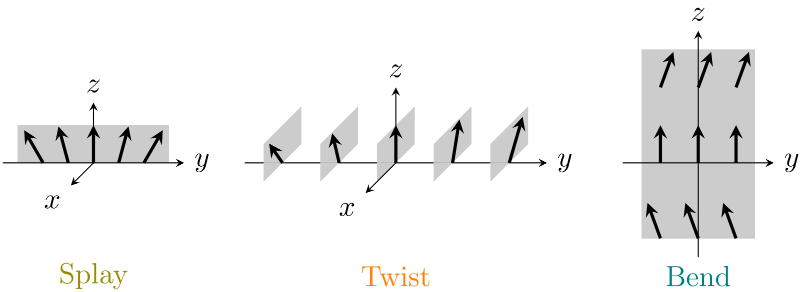
Can we observe the Lehmann effect in droplets of a **nematic achiral phase** with a **chiral director field**?

Outline

Elastic deformations in a nematic phase

Frank-Oseen elastic energy:

$$F[\boldsymbol{n}] = \int_V \frac{\mathrm{d}V}{2} \left(K_1 \left[\nabla \cdot \boldsymbol{n}\right]^2 + K_2 \left[\boldsymbol{n} \cdot \nabla \times \boldsymbol{n}\right]^2 + K_3 \left[\boldsymbol{n} \times \nabla \times \boldsymbol{n}\right]^2 \right)$$



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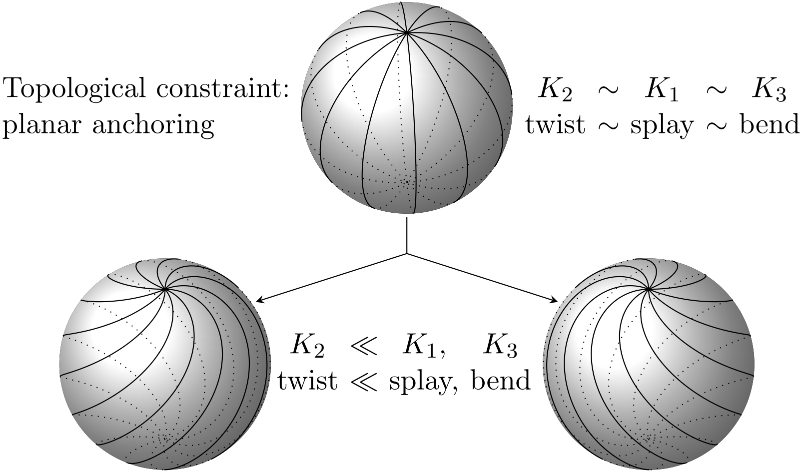
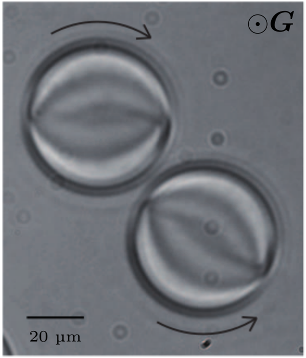
Stability of bipolar configuration



R. D. Williams. *Journal of physics A: mathematical and general*, 19:3211, 1986

Rotation of twisted bipolar droplets



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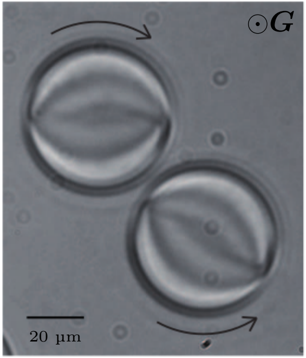

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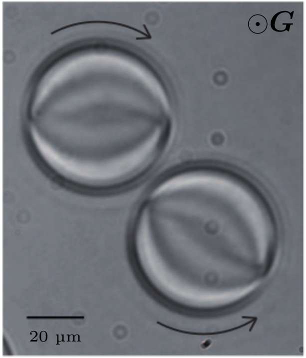

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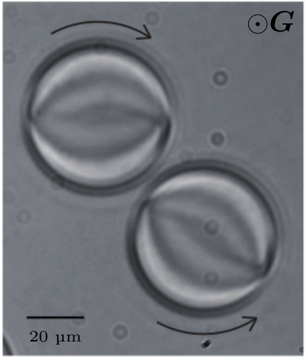

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Rotation only due to the twist of the director field

Outline

Concluding remarks for the Lehmann effect

- Lehmann effect in an achiral phase with a twisted director field:

The Lehmann effect is only due to the chirality of the director field

$\Downarrow$

The Leslie thermomechanical model cannot explain alone the Lehmann effect

P. Oswald, A. Dequidt, and G. Poy. *Liquid Crystals Reviews*, 7:142–166, 2019

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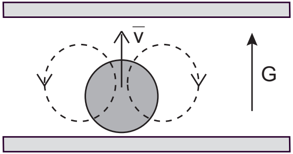
- What about other theoretical model? What is the “right” explanation?



Melting-growth model: a gradient of impurity drives the molecules upward inside the droplet while the droplet interface stays fixed

P. Oswald, A. Dequidt, and G. Poy. *Liquid Crystals Reviews*, 7:142–166, 2019

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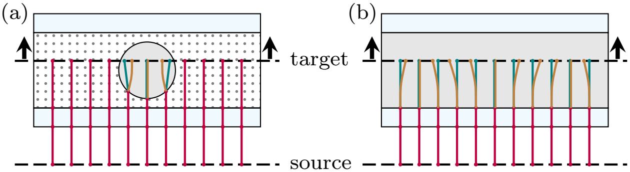
Nemaktis: an easy-to-use open-source platform including tools for light propagation in arbitrary birefringent media.

Outline

Hamiltonian ray-tracing and energy transport

$$\frac{\mathrm{d}\eta}{\mathrm{d}\bar{s}} = \{\boldsymbol{\eta}, \mathcal{H}\}$$

$$\boldsymbol{\eta} \equiv (\boldsymbol{x}, \boldsymbol{p})$$



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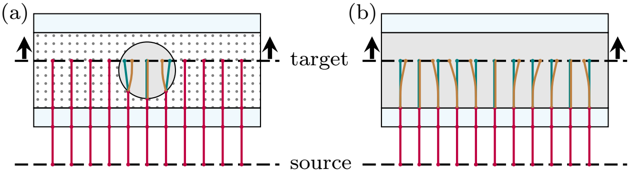
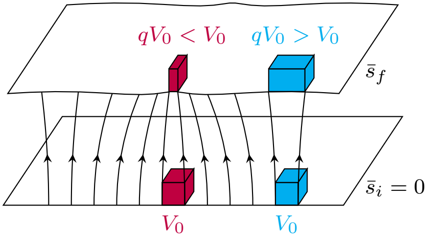
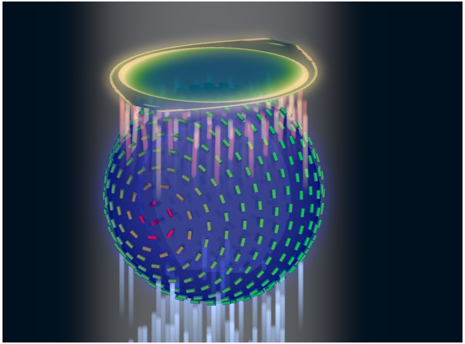
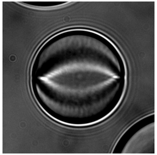


$\mathcal{F}^{(\alpha)} = n_{\text{eff}}\sqrt{q}\,E$ conserved along a ray

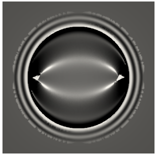


Application to bright-field microscopy





exp.



sim.

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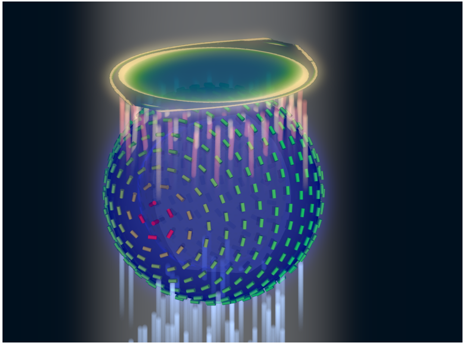

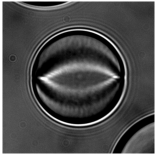

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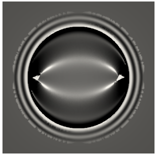

sim.

Advantage: access to ray geometry and natural eigenmodes
Disadvantage: Mauguin regime, caustics

Outline

Physics-based splitting of the wave equation

- Wave-equation in anisotropic media: $\left[\partial_k\partial_k\delta_{ij} - \partial_i\partial_j + k_0^2\epsilon_{ij}\right] E_j = 0$

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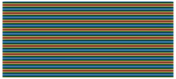
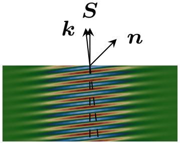
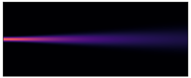
- What's inside $\boldsymbol{\mathcal{P}}$?



Phase op. $\boldsymbol{K} \sim k_0^2\boldsymbol{\epsilon}$

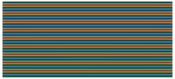


Walkoff op. $\boldsymbol{W} \sim (\boldsymbol{\epsilon}\,\boldsymbol{u}_z) \otimes \boldsymbol{\nabla}_\perp$

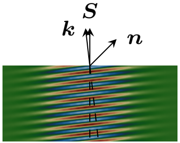


Diffraction op. $\boldsymbol{D} \sim \boldsymbol{\Delta}_\perp$

Physics-based splitting of the wave equation



Phase op. $\boldsymbol{K} \sim k_0^2 \boldsymbol{\epsilon}$



Walkoff op. $\boldsymbol{W} \sim (\boldsymbol{\epsilon}\, \boldsymbol{u}_z) \otimes \boldsymbol{\nabla}_\perp$

Diffraction op. $\boldsymbol{D} \sim \boldsymbol{\Delta}_\perp$

General expression for $\boldsymbol{\mathcal{P}}$:

$$\boldsymbol{\mathcal{P}} = i\, \boldsymbol{W} + \sqrt{\boldsymbol{K} + \boldsymbol{D}} + \mathcal{O}\left(\delta\epsilon^2\right)$$

Explicit solution for the transverse optical field:

$$\boldsymbol{E}_\perp\big|_{z_2} = \exp\left\{ i \int_{z_1}^{z_2} \boldsymbol{\mathcal{P}} \mathrm{d}z \right\} \boldsymbol{E}_\perp\big|_{z_1}$$

Typical application: polarised micrographs simulation

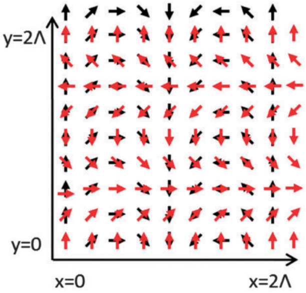
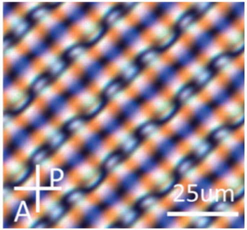
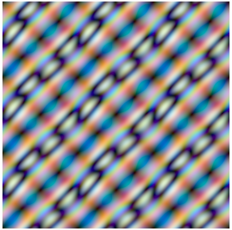


Photo-patterned sample:
I. Nys, J. Beeckman and K. Neyts, Soft Matter **11**, 2015



exp.



sim. (BPM)

Outline

Nemaktis is open-source

- The open-source package includes:
  - Low-level simulation backends (C++, python)
  - An easy-to-use high-level interface (python)
  - A graphical interface for micrographs simulation

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- Only Windows and Linux package for now (Mac should be supported in the future)

Outline

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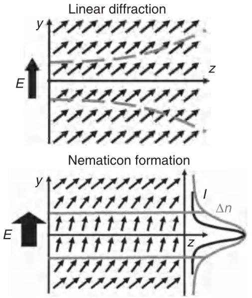
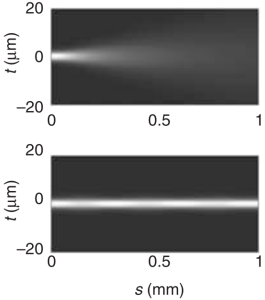
Motivations

Spatial light solitons in liquid crystals: nematicons



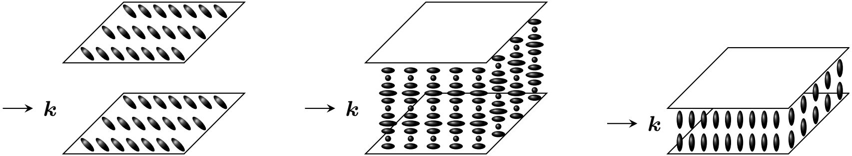


Increasing beam power

G. Assanto. *Nematicons.* John Wiley & Sons, 2013

Motivations

Studied systems in the past 20 years:



thick samples with planar $\boldsymbol{n}$

thick samples with cholesteric helix

thin samples with homeotropic $\boldsymbol{n}$

What about confined chiral systems?

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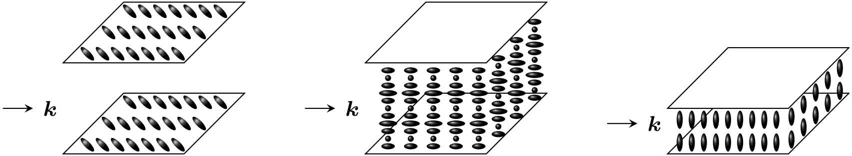

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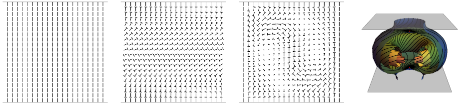
What about confined chiral systems?

Motivations

What makes frustrated cholesteric (FCLC) an interesting system:

- Metastability for carefully chosen values of d/P



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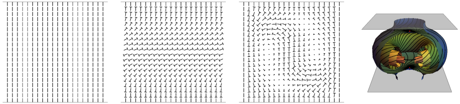
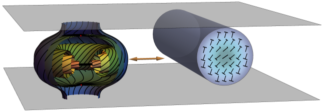


- Rich possibilities of interaction between light beams and topological solitons.



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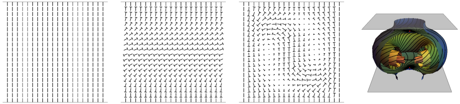

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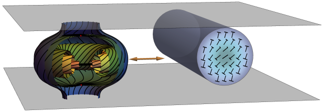

Problematic

Can we generate light solitons in frustrated cholesteric?

Outline

Orientational elasticity and non-linear interactions

Free energy of the liquid crystal phase:

$$F[\boldsymbol{n}, \boldsymbol{E}] = \int_V \mathrm{d}V \left[f_{\mathrm{F}}(\boldsymbol{n}, \nabla \boldsymbol{n}) - \frac{\epsilon_0 \epsilon_a \left| \boldsymbol{n} \cdot \boldsymbol{E} \right|^2}{4} \right]$$

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Non-linear iterative scheme:

- $\boldsymbol{E}_{k+1}$: BPM solution with $\boldsymbol{\epsilon} = \epsilon_\perp \mathbf{I} + \epsilon_a \boldsymbol{n}_k \otimes \boldsymbol{n}_k$
- $\boldsymbol{n}_{k+1} = \boldsymbol{n}_k + \mu \frac{\delta F}{\delta \boldsymbol{n}} \left[\boldsymbol{n}_k, \boldsymbol{E}_{k+1}\right]$

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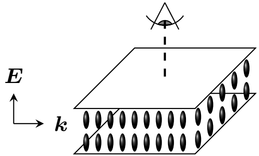
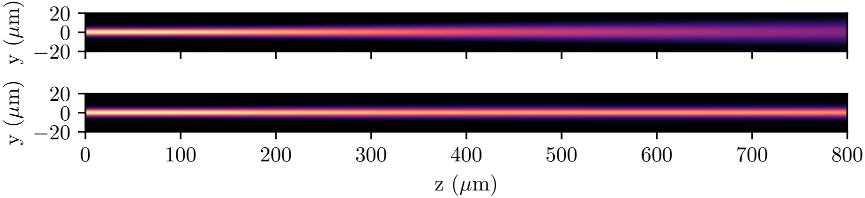
Typical running time for a mesh of 3×10^6 points: **4 s / step**
(Full resolution of Maxwell equations for the same mesh: **∼ 1 h**)

Optical fields structure

Top view of the thickness-averaged intensity:



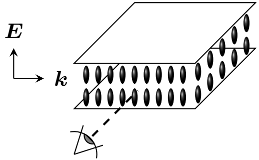


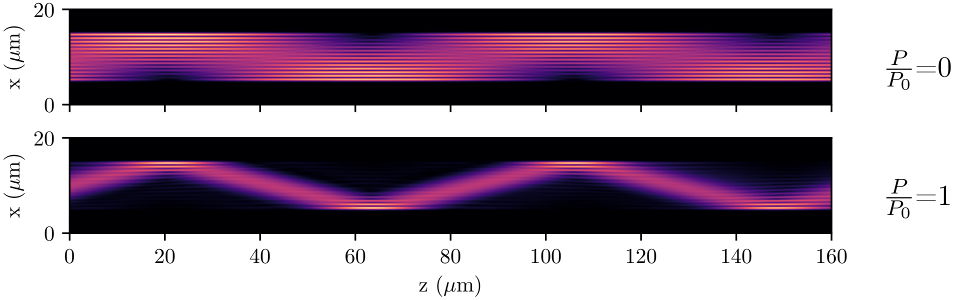
$\frac{P}{P_0} = 0$

$\frac{P}{P_0} = 1$

Optical fields structure

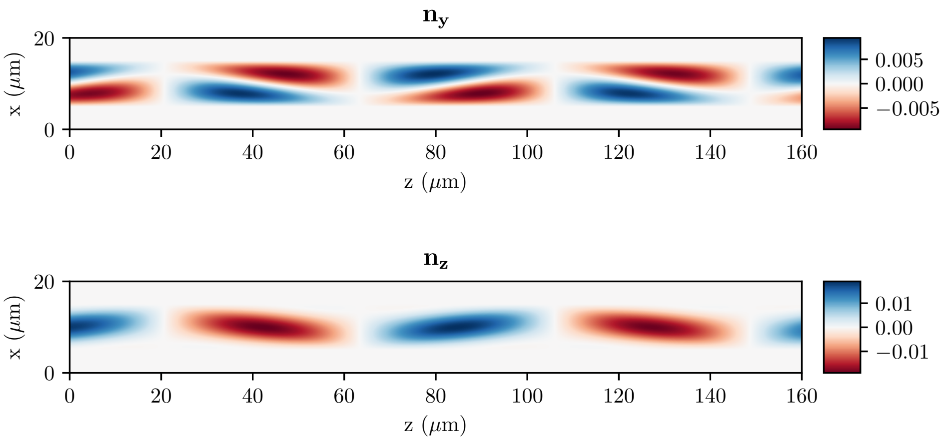
Side view of the field amplitude:





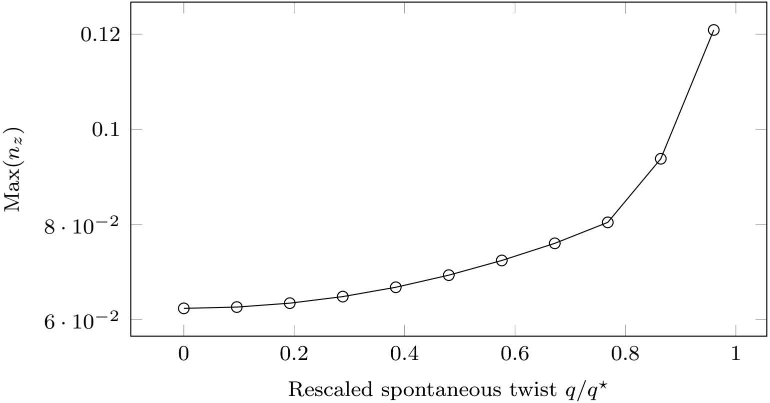
Non-linear response

Side view of the director field:



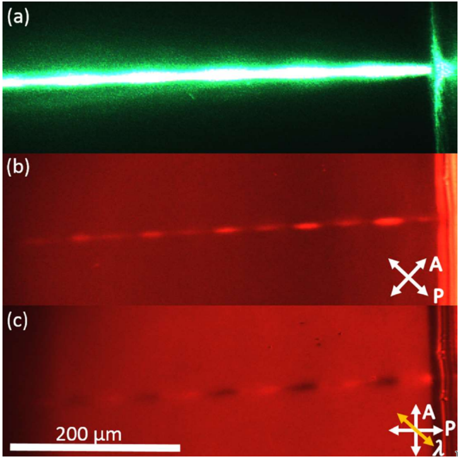
Non-linear response

Chirality-enhanced Kerr response:



Comparison with experiments

Scattered light and polarised optical micrographs (I. Smalyukh group):



Outline

Summary

- It is possible to generate solitons in confined cholesteric system, with:
  - ⋆ "bouncing" beam between the sample plates
  - ⋆ periodic reorientation along the beam axis
  - ⋆ chirality-enhanced Kerr response

Summary

- It is possible to generate solitons in confined cholesteric system, with:
  - ⋆ "bouncing" beam between the sample plates
  - ⋆ periodic reorientation along the beam axis
  - ⋆ chirality-enhanced Kerr response
- To be explored:
  - ⋆ Superposition of normal and transverse polarisations
  - ⋆ Interaction with topological solitons

Thank you for your attention!