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

Guilhem Poy

Faculty of Physics and Mathematics, Ljubljana

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Univerza v Ljubljani
Fakulteta za matematiko in fiziko

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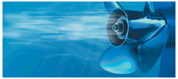
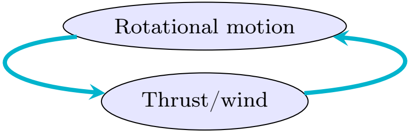
MARIE CURIE ACTIONS

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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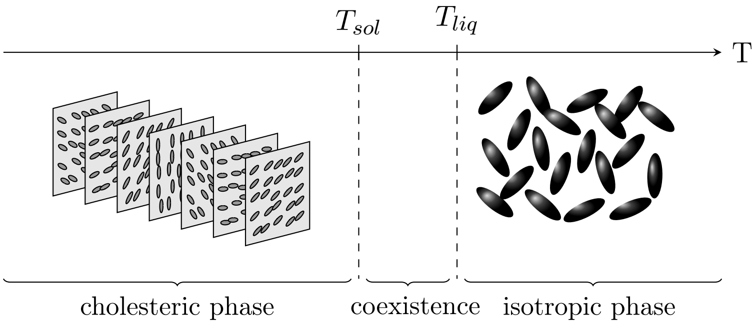
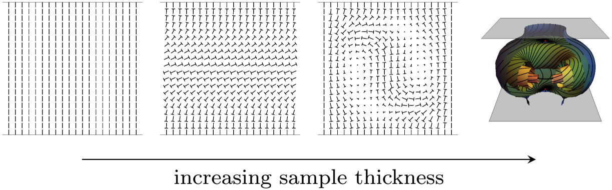


Confining cholesterics between two plates

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



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

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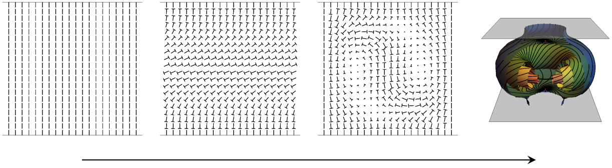
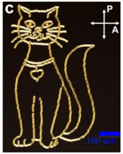
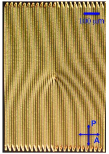
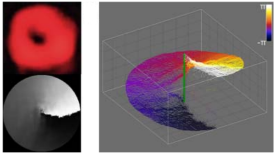


increasing sample thickness

- Arbitrary shapes can be written!



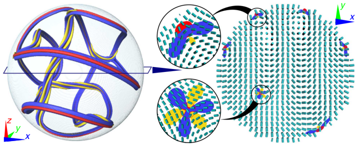




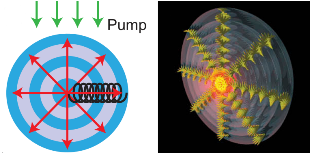
P. J. Ackerman et al. *Scientific Reports*, 2, 2012

Confining cholesterics inside droplets

Topological zoo of free standing knots



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

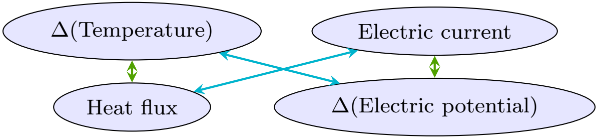


D. Seč, S. Čopar, and S. Žumer. *Nature Communications*, 5:3057, 2014

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

Other aspects of chirality in soft matter

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



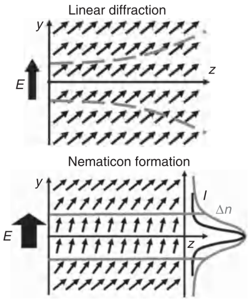
Applications:

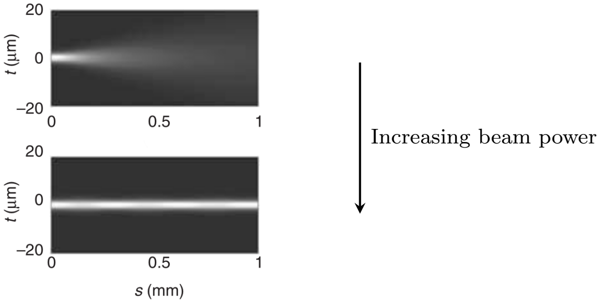
Problematic

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:





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

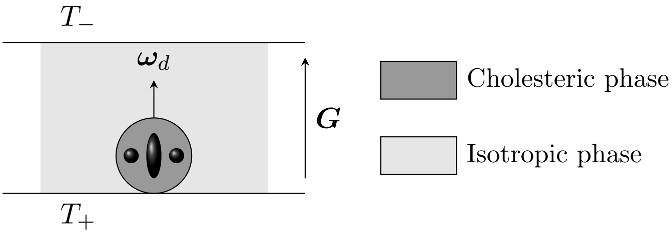
Problematic

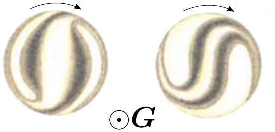
Role of chirality in the non-linear optical response of a confined cholesteric?

Outline

Outline

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

What is causing the rotation of the Lehmann droplets

Rotation because of the microscopic or macroscopic chirality?

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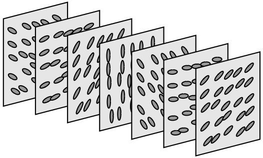
Rotation because of the microscopic or macroscopic chirality?

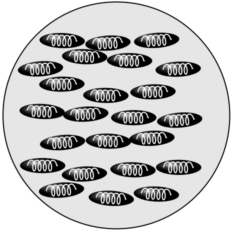
- microscopic chirality ⇔ chiral molecules

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



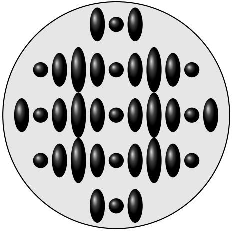
What is causing the rotation of the Lehmann droplets

Possible tests:



$\begin{cases} \text{chiral molecules} \leftrightarrow \text{cholesteric} \\ \text{no macroscopic twist (compensated)} \end{cases}$

Thermal gradient $\Rightarrow$ no rotation



$\begin{cases} \text{no chiral molecules} \leftrightarrow \text{nematic} \\ \text{macroscopic twist} \end{cases}$

Thermal gradient $\Rightarrow$ rotation?

Question

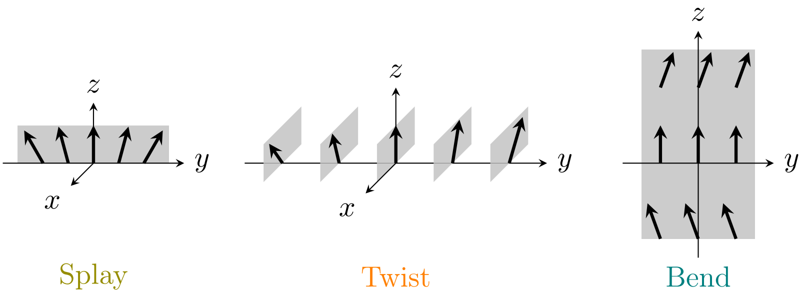
Can we observe the Lehmann effect in droplets of a **nematic achiral phase** with a **chiral director field**?

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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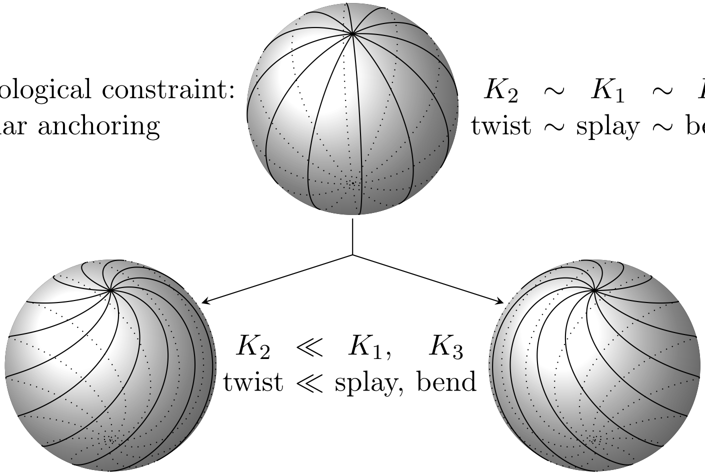
Stability of bipolar configuration

Topological constraint:
planar anchoring



$K_2 \sim K_1 \sim K_3$
twist $\sim$ splay $\sim$ bend

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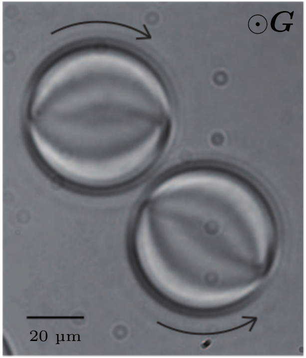
R. D. Williams. *Journal of physics A: mathematical and general*, 19:3211, 1986

Rotation of twisted bipolar droplets



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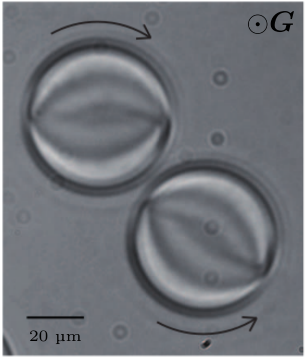

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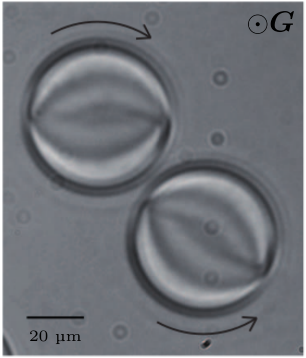

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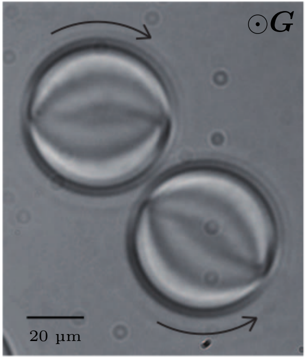

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Rotation only due to the twist of the director field

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1. Introduction

2. Lehmann effect: an out-of-equilibrium effect in chiral liquid crystal droplets
   - Problematic
   - Lehmann effect in nematic droplets
   - Summary

3. Interlude: light propagation in anisotropic media

4. Role of chirality in the non-linear reponse of a confined cholesteric

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

J. Ignés-Mullol, G. Poy, and P. Oswald. *Physical Review Letters*, 117:057801, 2016

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

Concluding remarks for the Lehmann effect

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

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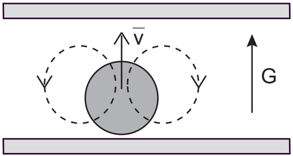
- What is the "right" mechanism behind the Lehmann effect?



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

J. Ignés-Mullol, G. Poy, and P. Oswald. *Physical Review Letters*, 117:057801, 2016

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Motivations

- Recent advances in LC-based light application: tunable microresonators, micro-optical elements, diffraction gratings...

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- Simulation tools for light propagation:
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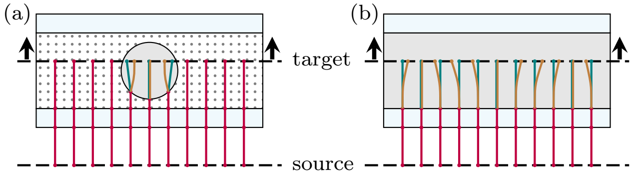
Need for advanced light propagation code, if possible open-source

First approach: 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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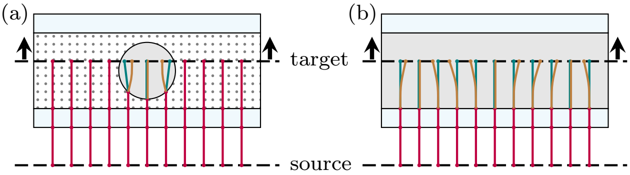
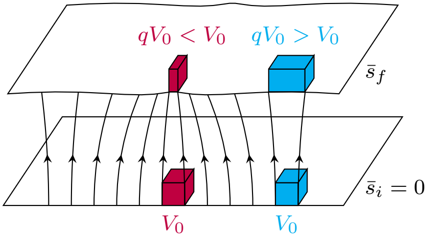


$\mathcal{F}^{(\alpha)} = n_{\text{eff}}\,\sqrt{q}\,E$ conserved along a ray



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Second approach: physics-based splitting of the wave equation

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

G. Poy and S. Žumer. *Optics Express*, 28:24327, 2020

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$$i\partial_z \boldsymbol{E}_\perp = -\boldsymbol{\mathcal{P}}\boldsymbol{E}_\perp$$

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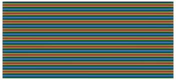
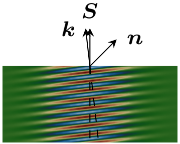
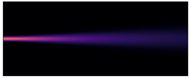
- What's inside $\boldsymbol{\mathcal{P}}$?



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

G. Poy and S. Žumer. *Optics Express*, 28:24327, 2020

Nemaktis: an open-source package for polarised microscopy

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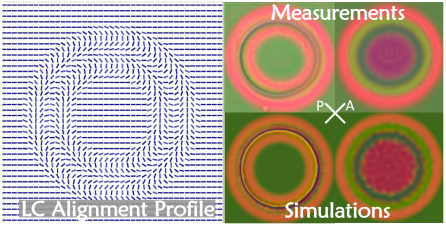
- Closed-source BPM code for advanced uses: wide-angle beam deflection, non-linear optics, etc.



B. Berteloot, I. Nys, G. Poy, J. Beeckman, and K. Neyts. *Soft Matter*, 16:4999, 2020

Outline

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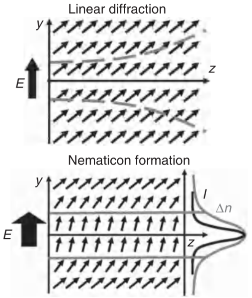
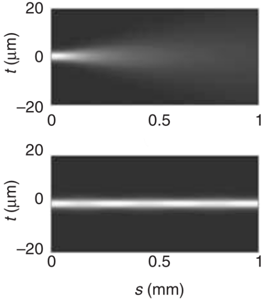
Motivations

Spatial light solitons in liquid crystals: nematicons



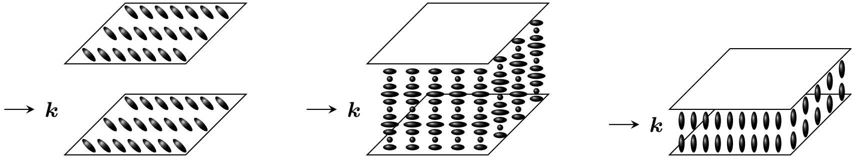


Increasing beam power

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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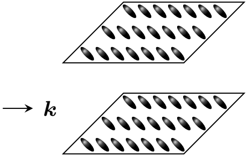

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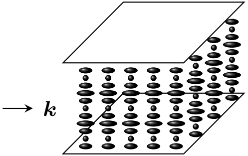

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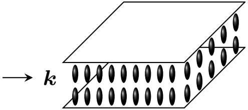

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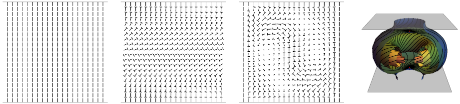
What about confined chiral systems?

Motivations

What makes frustrated cholesteric (FCLC) an interesting system:

- Metastability for carefully chosen values of d/P



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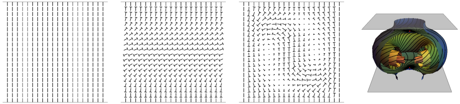
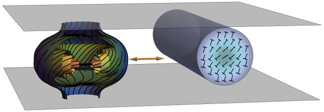


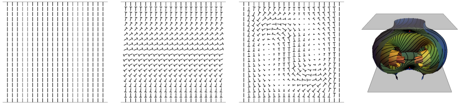
- Rich possibilities of interaction between light beams and topological solitons.



Motivations

What makes frustrated cholesteric (FCLC) an interesting system:

- Metastability for carefully chosen values of d/P



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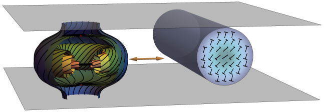

Problematic

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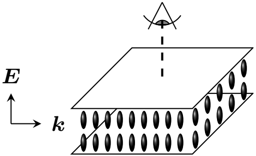
Typical running time for a mesh of 3×10^6 points: **4 s / step**
(Full resolution of Maxwell equations for the same mesh: **∼ 1 h**)

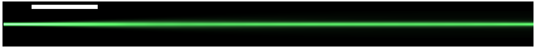
Top-view observations



Top view of the thickness-averaged laser intensity (simulation):

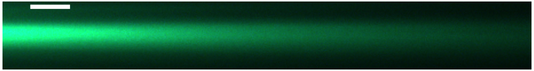
Linear optical regime

Non-linear optical regime

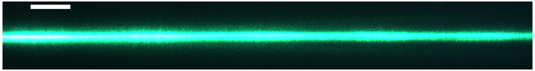


Top view of the scattered laser light (experiments):

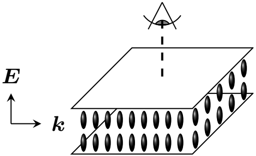
Linear optical regime



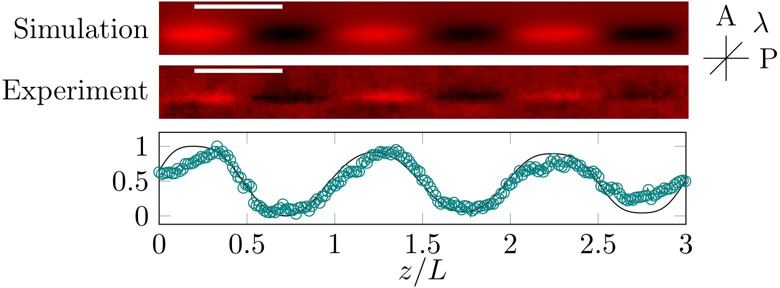
Non-linear optical regime



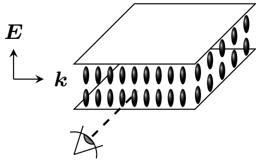
Top-view observations



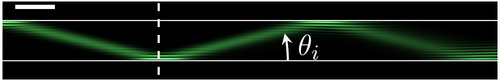
Top view polarised optical micrograph:



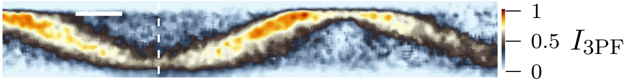
Why is there a periodic molecular reorientation?



Side slice of beam intensity (simulation):



Side slice of 3PF signal (experiment):



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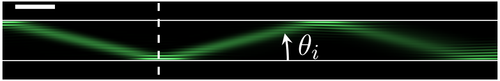

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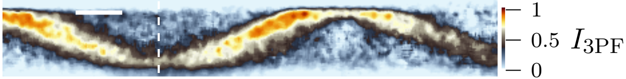
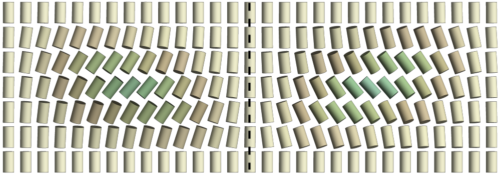
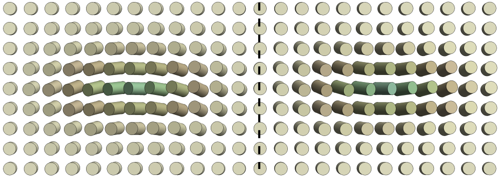


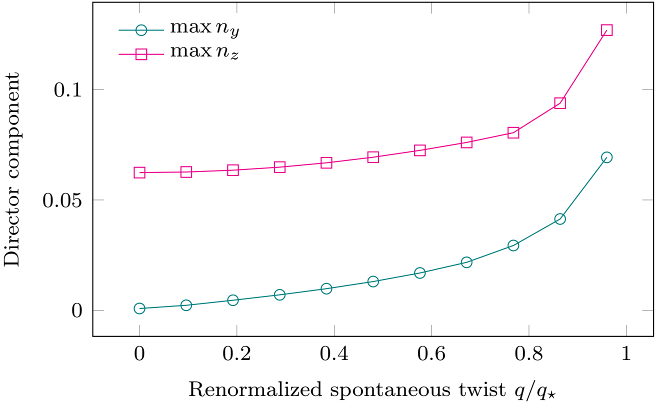
Side slice of director field



Mid-sample slice of director field



Chirality-enhanced non-linear optical response



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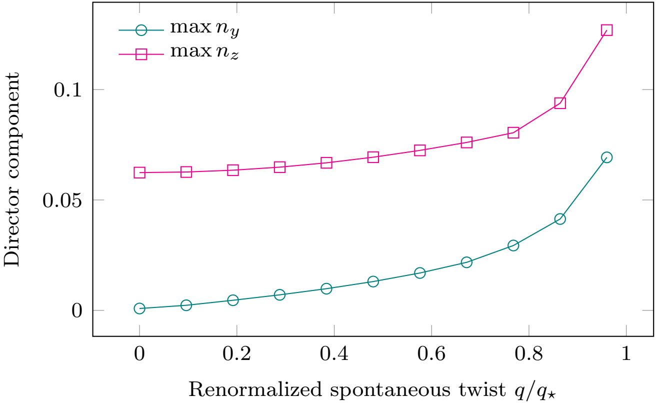


⇒ Potential for low-power non-linear optical photonics devices (e.g. active lenses)

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

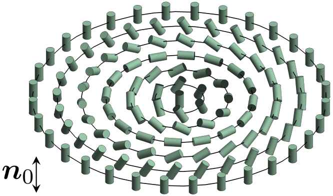
G. Poy, A. J. Hess, I. I. Smalyukh, and S. Žumer. *Physical Review Letters*, 125:077801, 2020

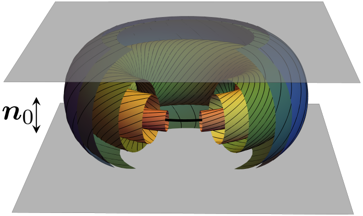
Summary

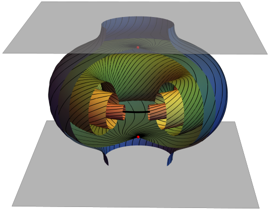
- It is possible to generate solitons in confined cholesteric system, with:
  - "bouncing" beam between the sample plates
  - periodic reorientation along the beam axis
  - chirality-enhanced Kerr response
- To be explored:
  - Superposition of normal and transverse polarisations (spin-orbit solitons)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

G. Poy, A. J. Hess, I. I. Smalyukh, and S. Žumer. *Physical Review Letters*, 125:077801, 2020

Towards topological optomechanics







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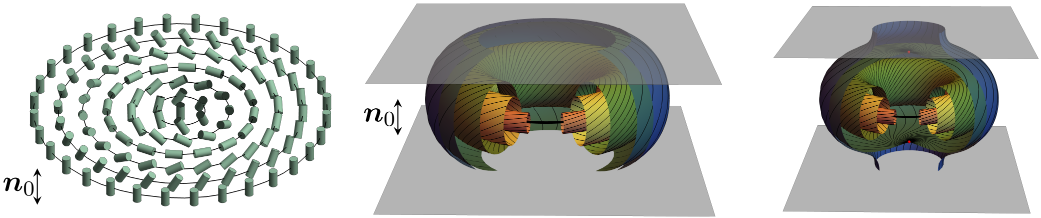
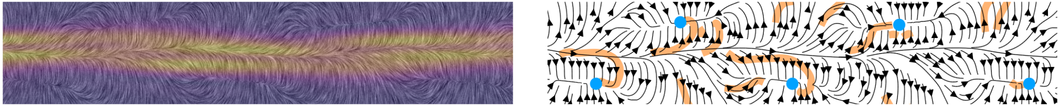
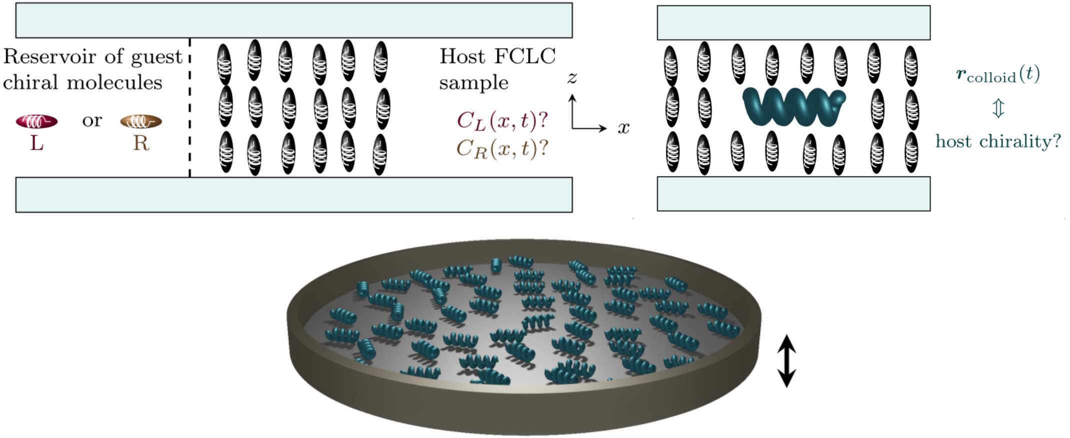




CNRS{5}: Diffusive transport properties in chiral guest-host systems

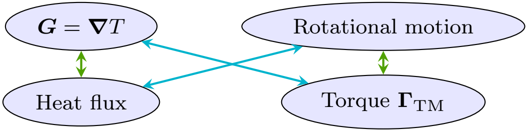


Avertisement

Thank you for your attention!

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.

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

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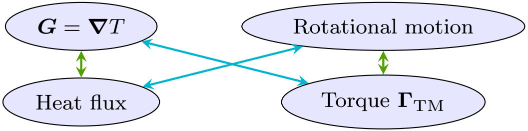

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  $\boldsymbol{\Gamma}_{\mathrm{TM}} = \nu\, \boldsymbol{n} \times [\boldsymbol{n} \times \boldsymbol{G}]$, with ν the Leslie thermomechanical coefficient.
- As in a wind turbine, essential role of the chirality:
  no rotation predicted in a nematic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

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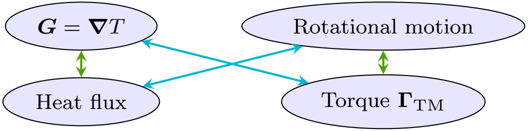


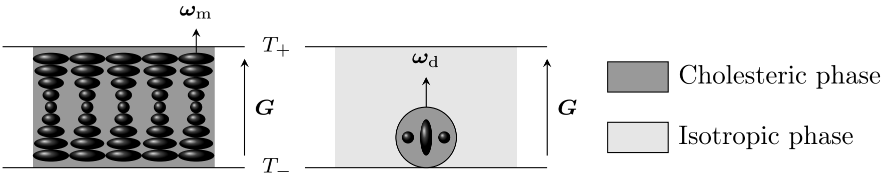
Leslie paradigm

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:



Leslie effect
Periods of 10–100 min

Lehmann effect
Periods of 1–100 s

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

P. Oswald. *Europhysics Letters*, 108:36001, 2014

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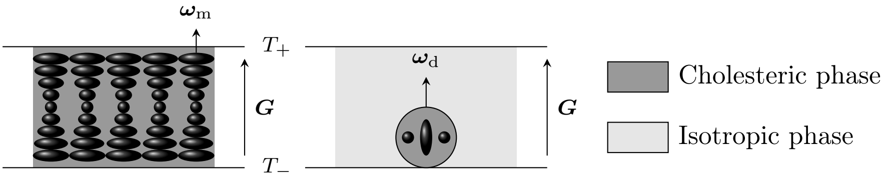

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ω_d and ω_m sometimes of opposite signs!

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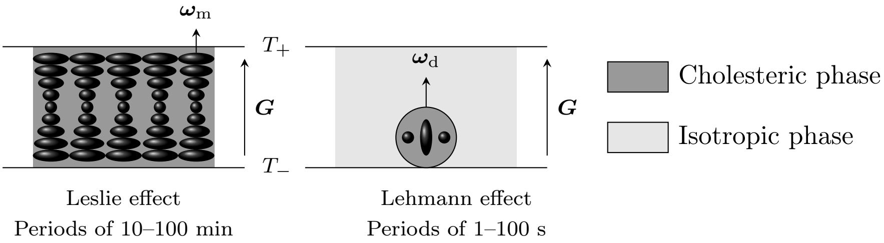

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Leslie effect $\neq$ Lehmann effect?

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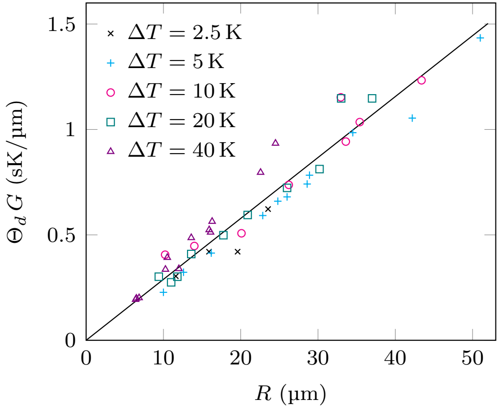
P. Oswald. *Europhysics Letters*, 108:36001, 2014

Rotation periods of SSY droplets



- Angular velocity $\omega_d = 2\pi/\Theta_d$ proportional to G.

J. Ignés-Mullol, G. Poy, and P. Oswald. *Physical Review Letters*, 117:057801, 2016

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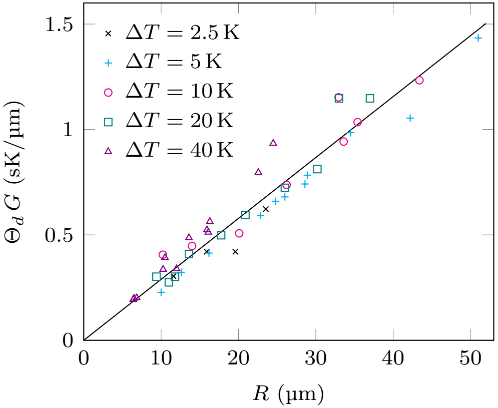

- Angular velocity $\omega_d = 2\pi/\Theta_d$ proportional to G.
- Period Θ_d proportional to the radius R.

J. Ignés-Mullol, G. Poy, and P. Oswald. *Physical Review Letters*, 117:057801, 2016