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

Guilhem Poy

Faculty of Physics and Mathematics, Ljubljana

October 16, 2020
Outline

1 Introduction

2 Lehmann effect: an out-of-equilibrium effect in chiral liquid crystal droplets

3 Interlude: light propagation in anisotropic media

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

- Nematic liquid crystal: no positional order, mean molecular orientation $\mathbf{n}$
Chirality in soft matter: the cholesteric phase

- Nematic liquid crystal: no positional order, mean molecular orientation $n$
- Nematic phase + chiral molecules: cholesteric phase.
Chirality in soft matter: the cholesteric phase

- Nematic liquid crystal: no positional order, mean molecular orientation $\mathbf{n}$
- Nematic phase + chiral molecules: cholesteric phase.
- Effect of chirality: helix structure for the director vector field $\mathbf{n}$. 
Chirality in soft matter: the cholesteric phase

- Nematic liquid crystal: no positional order, mean molecular orientation $\mathbf{n}$
- Nematic phase + chiral molecules: cholesteric phase.
- Effect of chirality: helix structure for the director vector field $\mathbf{n}$.

\[
\begin{array}{c}
T_{\text{sol}} \quad T_{\text{liq}} \\
\text{cholesteric phase} & \text{coexistence} & \text{isotropic phase}
\end{array}
\]
Confining cholesterics between two plates

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

![Diagram of cholesterics](image)

increasing sample thickness

P. J. Ackerman et al. *Scientific Reports, 2, 2012*
Confining cholesterics between two plates

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

- 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


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

Δ(Temperature) → Heat flux → Electric current → Δ(Electric potential)

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:


Problematic

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

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 response of a confined cholesteric
Outline

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 response of a confined cholesteric
Lehmann effect

Problematic

First observations by Lehmann

Lehmann, 1900:
- coexistence of cholesteric droplets with the isotropic fluid
- rotation of the droplets internal texture when heated from below

What is causing the rotation of the Lehmann droplets

Rotation because of the microscopic or macroscopic chirality?
What is causing the rotation of the Lehmann droplets

Rotation because of the microscopic or macroscopic chirality?

- microscopic chirality $\Leftrightarrow$ chiral molecules
What is causing the rotation of the Lehmann droplets?

Rotation because of the microscopic or macroscopic chirality?

- microscopic chirality $\Leftrightarrow$ chiral molecules
- macroscopic chirality $\Leftrightarrow$ twisted texture (helix in at least one direction)
What is causing the rotation of the Lehmann droplets?

Possible tests:

\[
\begin{align*}
\{ & \text{chiral molecules} \leftrightarrow \text{cholesteric} \\
& \text{no macroscopic twist (compensated)} \\
\} \\
\Rightarrow & \text{Thermal gradient} \Rightarrow \text{no rotation}
\end{align*}
\]

\[
\begin{align*}
\{ & \text{no chiral molecules} \leftrightarrow \text{nematic} \\
& \text{macroscopic twist} \\
\} \\
\Rightarrow & \text{Thermal gradient} \Rightarrow \text{rotation?}
\end{align*}
\]

Question

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

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 response of a confined cholesteric
Elastic deformations in a nematic phase

Frank-Oseen elastic energy:

$$F[n] = \int_V \frac{dV}{2} \left( K_1 \left[ \nabla \cdot n \right]^2 + K_2 \left[ n \cdot \nabla \times n \right]^2 + K_3 \left[ n \times \nabla \times n \right]^2 \right)$$
How to obtain twist

Two possible origins for a twisted director field:
How to obtain twist

Two possible origins for a twisted director field:

- action of a chiral interaction potential between molecules:
  \[ F[n] \to F[n] + \int_V dV \ K_2 \ q \ [n \cdot \nabla \times n] \]
How to obtain twist

Two possible origins for a twisted director field:

- action of a **chiral interaction potential** between molecules:
  - \( F[n] \rightarrow F[n] + \int_V dV \ K_2 \ q \ [n \cdot \nabla \times n] \)
  - pertinent only in cholesteric phase
How to obtain twist

Two possible origins for a twisted director field:

- action of a **chiral interaction potential** between molecules:
  - \( F[n] \rightarrow F[n] + \int_V dV \ K_2 \ q \ [n \cdot \nabla \times n] \)
  - pertinent only in cholesteric phase

- action of a **topological constraint** on the LC domain surface:
  - \( F[n] \rightarrow F[n] + \int_S dS \ \gamma(n) \), with \( \gamma \) the anchoring energy
How to obtain twist

Two possible origins for a twisted director field:

- **action of a chiral interaction potential** between molecules:
  \[ F[n] \rightarrow F[n] + \int_V dV \, K_2 \, q \, [n \cdot \nabla \times n] \]
  - pertinent only in cholesteric phase

- **action of a topological constraint** on the LC domain surface:
  \[ F[n] \rightarrow F[n] + \int_S dS \, \gamma(n), \text{ with } \gamma \text{ the anchoring energy} \]
  - pertinent both in nematic and cholesteric phases
Stability of bipolar configuration

Topological constraint:
planar anchoring

\[ K_2 \sim K_1 \sim K_3 \]
twist \sim splay \sim bend

\[ K_2 \ll K_1, K_3 \]
twist \ll splay, bend

Rotation of twisted bipolar droplets

- Lyotropic chromonic nematic used: water + 30% SSY
  
  \[
  \frac{K_2}{K_1} \simeq 0.16, \quad \frac{K_2}{K_3} \simeq 0.12
  \]
Rotation of twisted bipolar droplets

- Lyotropic chromonic nematic used: water + 30% SSY
  \( K_2/K_1 \simeq 0.16, \ K_2/K_3 \simeq 0.12 \)

- **Achiral phase**, with random handedness of the twist inside the droplets
Rotation of twisted bipolar droplets

- Lyotropic chromonic nematic used: water + 30% SSY
  \( K_2/K_1 \simeq 0.16, \ K_2/K_3 \simeq 0.12 \)

- **Achiral phase**, with random handedness of the twist inside the droplets

- The sign of twist fixes the sign of the angular velocity \( \Rightarrow \) two senses of rotation
Rotation of twisted bipolar droplets

- Lyotropic chromonic nematic used: water + 30% SSY
  \( K_2/K_1 \simeq 0.16, \ K_2/K_3 \simeq 0.12 \)

- **Achiral phase**, with random handedness of the twist inside the droplets

- The sign of twist fixes the sign of the angular velocity ⇒ **two senses of rotation**

Rotation only due to the twist of the director field
Outline

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 response 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


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

- 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


Outline

1. Introduction

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

3. Interlude: light propagation in anisotropic media

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

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

- Recent advances in LC-based light application: tunable microresonators, micro-optical elements, diffraction gratings...
- Simulation tools for light propagation:
  - Jones method (fast but inaccurate, easy to code)
  - Finite Difference Time Domain (accurate but slow, open-source, complex to use)
  - Other methods (in-house implementation)
Motivations

- Recent advances in LC-based light application: tunable microresonators, micro-optical elements, diffraction gratings...
- Simulation tools for light propagation:
  - Jones method (fast but inaccurate, easy to code)
  - Finite Difference Time Domain (accurate but slow, open-source, complex to use)
  - Other methods (in-house implementation)

Need for advanced light propagation code, if possible open-source
First approach: Hamiltonian ray-tracing and energy transport

\[ \frac{d\eta}{ds} = \{\eta, \mathcal{H}\} \]
\[ \eta \equiv (x, p) \]

First approach: Hamiltonian ray-tracing and energy transport

\[ \frac{d\eta}{d\bar{s}} = \{\eta, \mathcal{H}\} \]
\[ \eta \equiv (x, p) \]

\[ J^{(\alpha)} = n_{\text{eff}} \sqrt{q} E \text{ conserved along a ray} \]

Second approach: physics-based splitting of the wave equation

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

Second approach: physics-based splitting of the wave equation

- Wave-equation in anisotropic media: \[ [\partial_k \partial_k \delta_{ij} - \partial_i \partial_j + k_0^2 \epsilon_{ij}] E_j = 0 \]
- After eliminating \( E_z \) and keeping only forward modes:

\[ i \partial_z E_\perp = -\mathcal{P} E_\perp \]

Second approach: physics-based splitting of the wave equation

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

- After eliminating \( E_z \) and keeping only forward modes:
  \[ i \partial_z E_{\perp} = -\mathcal{P} E_{\perp} \]

- What’s inside \( \mathcal{P} \)?

  - Phase op. \( K \sim k_0^2 \epsilon \)
  - Walkoff op. \( W \sim (\epsilon u_z) \otimes \nabla_{\perp} \)
  - Diffraction op. \( D \sim \Delta_{\perp} \)

Nemaktis: an open-source package for polarised microscopy

- 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

Where to find it: search Nemaktis on github.com or google.
Nemaktis: an open-source package for polarised microscopy

- 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
- Where to find it: search Nemaktis on github.com or google.
Nemaktis: an open-source package for polarised microscopy

- 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

- Where to find it: search Nemaktis on github.com or google.
- Closed-source BPM code for advanced uses: wide-angle beam deflection, non-linear optics, etc.

Outline

1 Introduction

2 Lehmann effect: an out-of-equilibrium effect in chiral liquid crystal droplets

3 Interlude: light propagation in anisotropic media

4 Role of chirality in the non-linear response of a confined cholesteric
   - Motivations
   - Light solitons in frustrated cholesteric
   - Summary
Outline

1. Introduction

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

3. Interlude: light propagation in anisotropic media

4. Role of chirality in the non-linear response of a confined cholesteric
   - Motivations
   - Light solitons in frustrated cholesteric
   - Summary
Motivations

Spatial light solitons in liquid crystals: nematicons

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

Motivations

Studied systems in the past 20 years:

- Thick samples with planar $n$
- Thick samples with cholesteric helix
- Thin samples with homeotropic $n$

What about confined chiral systems?
Motivations

Studied systems in the past 20 years:

- Thick samples with planar $n$
- Thick samples with cholesteric helix
- Thin samples with homeotropic $n$

What about confined chiral systems?
Motivations

What makes frustrated cholesteric (FCLC) an interesting system:

- Metastability for carefully chosen values of $d/P$
Motivations

What makes frustrated cholesteric (FCLC) an interesting system:

- Metastability for carefully chosen values of $d/P$
- 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 $d/P$
- Rich possibilities of interaction between light beams and topological solitons.

Problematic

Can we generate light solitons in frustrated cholesteric?
Outline

1. Introduction

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

3. Interlude: light propagation in anisotropic media

4. Role of chirality in the non-linear response of a confined cholesteric
   - Motivations
   - Light solitons in frustrated cholesteric
   - Summary
Orientational elasticity and non-linear interactions

Free energy of the liquid crystal phase:

\[
F[n, E] = \int_V \mathrm{d}V \left[ f_F(n, \nabla n) - \frac{\epsilon_0 \epsilon_a |n \cdot E|^2}{4} \right]
\]
Orientational elasticity and non-linear interactions

Free energy of the liquid crystal phase:

\[ F[n, E] = \int_V dV \left[ f_F(n, \nabla n) - \frac{\epsilon_0 \epsilon_a |n \cdot E|^2}{4} \right] \]

Non-linear iterative scheme:

- \( E_{k+1} \): BPM solution with \( \epsilon = \epsilon_\perp I + \epsilon_a n_k \otimes n_k \)
- \( n_{k+1} = n_k + \mu \frac{\delta F}{\delta n} [n_k, E_{k+1}] \)
Orientational elasticity and non-linear interactions

Free energy of the liquid crystal phase:

$$F[n, E] = \int_V dV \left[ f_F(n, \nabla n) - \left( \epsilon_0 \epsilon_a |n \cdot E|^2 \right) \right]$$

Non-linear iterative scheme:

- $E_{k+1}$: BPM solution with $\epsilon = \epsilon_\perp I + \epsilon_a n_k \otimes n_k$
- $n_{k+1} = n_k + \mu \frac{\delta F}{\delta n} [n_k, E_{k+1}]$

Typical running time for a mesh of $3 \times 10^6$ points: $4$ s/step

(Full resolution of Maxwell equations for the same mesh: $\sim 1$ 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:

Simulation

Experiment
Why is there a periodic molecular reorientation?

Side slice of beam intensity (simulation):

Side slice of 3PF signal (experiment):

$1$

$0.5$

$I_{3PF}$

$0$
Why is there a periodic molecular reorientation?

Side slice of beam intensity (simulation):

Side slice of 3PF signal (experiment):

Side slice of director field

Mid-sample slice of director field
Chirality-enhanced non-linear optical response

\[
\begin{align*}
\text{Renormalized spontaneous twist } & \frac{q}{q^*} \\
\text{Director component } & \max n_y \quad \max n_z
\end{align*}
\]

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

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

1. Introduction
2. Lehmann effect: an out-of-equilibrium effect in chiral liquid crystal droplets
3. Interlude: light propagation in anisotropic media
4. Role of chirality in the non-linear response of a confined cholesteric
   - Motivations
   - Light solitons in frustrated cholesteric
   - Summary
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

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

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)

Towards topological optomechanics
Towards topological optomechanics
CNRS\{5\}: Diffusive transport properties in chiral guest-host systems
Realization and Application of Topological Defect Patterns in Soft and Living Matter

Guest Editors
Dr. Simon Čopar, Dr. Guilhem Poy, Prof. Dr. Anupam Sengupta

Deadline
01 June 2021
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:
  \[ \Gamma_{\text{TM}} = \nu \mathbf{n} \times [\mathbf{n} \times \mathbf{G}] \], with \( \nu \) the Leslie thermomechanical coefficient.

Summary

Leslie interpretation of the Lehmann experiment

First explanation by Leslie in 1968:

- Existence, in a cholesteric phase, of a torque on the director:
  \[ \Gamma_{TM} = \nu \mathbf{n} \times [\mathbf{n} \times \mathbf{G}] \], with \( \nu \) the Leslie thermomechanical coefficient.
- As in a wind turbine, essential role of the chirality:
  no rotation predicted in a nematic phase.

Leslie interpretation of the Lehmann experiment

First explanation by Leslie in 1968:

\[ G = \nabla T \]

Heat flux \hspace{1cm} Rotational motion

\[ \Gamma_{TM} \]

Torque

Leslie paradigm

The rotation of the texture in the Lehmann experiment is due to the Leslie thermomechanical torque \( \Gamma_{TM} \)

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

Summary

Lehmann vs. Leslie experiment

Oswald & Dequidt, 2008-2014:

\[
\begin{align*}
\omega_m & \quad T_+ \\
G & \quad T_- \\
\omega_d & \quad \text{Cholesteric phase} \\
& \quad \text{Isotropic phase}
\end{align*}
\]

Leslie effect
Periods of 10–100 min

Lehmann effect
Periods of 1–100 s

\(G\omega_m\) and \(G\omega_d\) sometimes of opposite signs!

Leslie effect \(\neq\) Lehmann effect?


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

**Summary**

Lehmann vs. Leslie experiment

Oswald & Dequidt, 2008-2014:

Leslie effect
Periods of 10–100 min

\[ T^- \quad G \quad T^+ \]

\[ \omega_m \]

Cholesteric phase

Isotropic phase

Lehmann effect
Periods of 1–100 s

\[ G \]

\[ \omega_d \]

\( \omega_d \) and \( \omega_m \) sometimes of opposite signs!


Lehmann vs. Leslie experiment

Oswald & Dequidt, 2008-2014:

\[ \omega_d \text{ and } \omega_m \text{ sometimes of opposite signs!} \]

**Leslie effect ≠ Lehmann effect?**


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

Summary

Rotation periods of SSY droplets

\[ \Theta_d G (sK/\mu m) \]

- \( \Delta T = 2.5 \text{ K} \)
- \( \Delta T = 5 \text{ K} \)
- \( \Delta T = 10 \text{ K} \)
- \( \Delta T = 20 \text{ K} \)
- \( \Delta T = 40 \text{ K} \)

Angular velocity \( \omega_d = 2\pi/\Theta_d \)

proportional to \( G \).

Rotation periods of SSY droplets

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