On the pertinence of the thermomechanical model in the Lehmann rotation of cholesteric and nematic droplets

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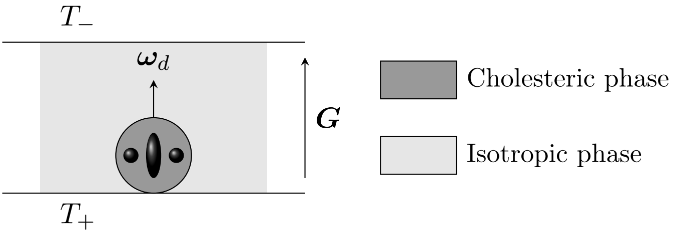
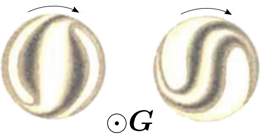
Outline

Outline

First observations by Lehmann



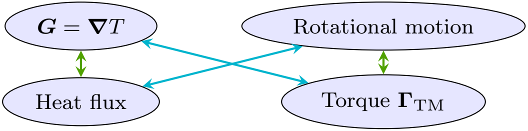


Lehmann, 1900:

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

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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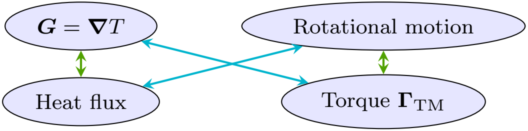

- Existence, in a cholesteric phase, of a torque on the director: $\boldsymbol{\Gamma}_{\mathrm{TM}} = \nu\, \boldsymbol{n} \times [\boldsymbol{n} \times \boldsymbol{G}]$, with ν the Leslie thermomechanical coefficient.
- As in a wind turbine, essential role of the chirality: no rotation predicted in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

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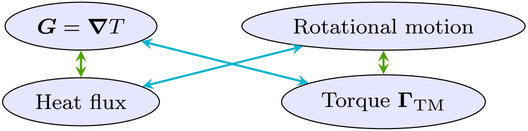
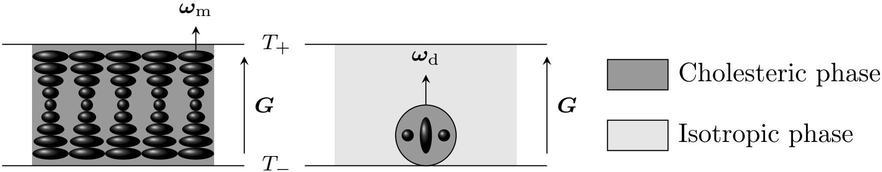


Leslie paradigm

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

Lehmann vs. Leslie experiment

Oswald & Dequidt, 2008-2014:



Leslie effect
Periods of 10–100 min

Lehmann effect
Periods of 1–100 s

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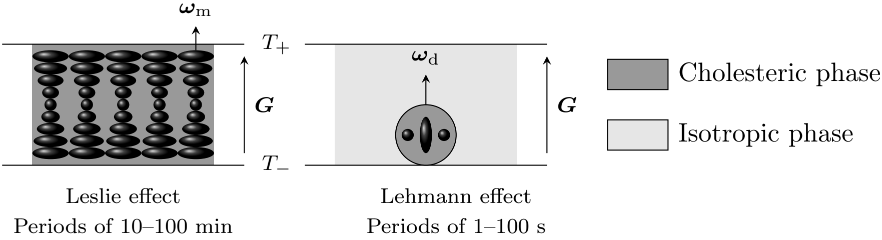

- Measurement of ω_m gives a value for the thermomechanical coefficient of Leslie ν.

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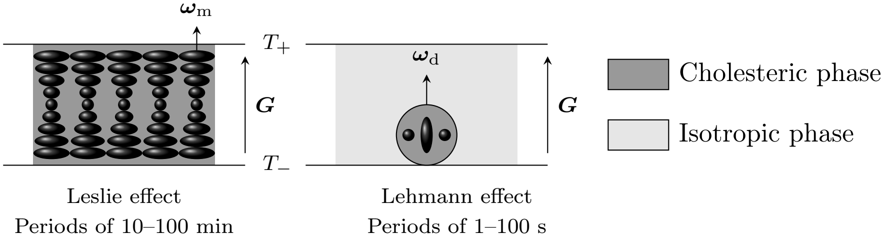

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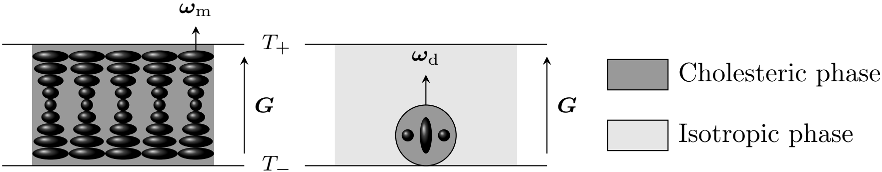

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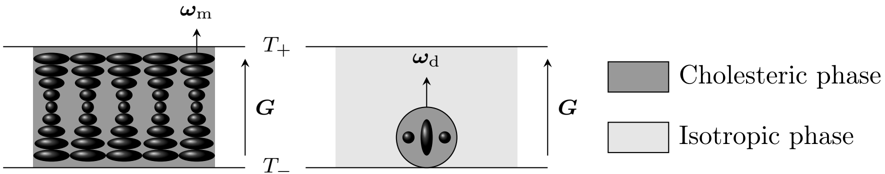
- Measurement of ω_m gives a value for the thermomechanical coefficient of Leslie ν.
- The value of ν is $10 - 1000$ too small to explain the order of magnitude of ω_d.
- ω_d and ω_m sometimes of opposite signs!

Lehmann vs. Leslie experiment



Leslie effect
Periods of 10–100 min

Lehmann effect
Periods of 1–100 s

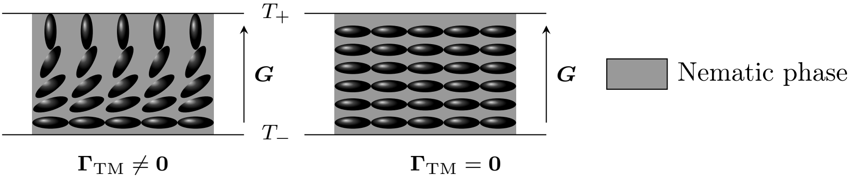
Leslie effect $\neq$ Lehmann effect: the Leslie paradigm must be abandoned.

Outline

Akopyan and Zel'dovich couplings

Akopyan & Zel'dovich, 1984:

- Generalization of $\boldsymbol{\Gamma}_{\mathrm{TM}}$ with terms of the type $\xi\,(\nabla n)\,G$.
- Terms in ξ are allowed both in nematics and cholesterics.



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To be explored

- Clarification on the existence of these terms.
- Can we explain the Lehmann effect with these effects?
- Can we observe the Lehmann effect in twisted nematic droplets?

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Derivation of the phenomenological equations

- Write down the entropy production:

$$T\mathring{\sigma} = \boldsymbol{j}^{\alpha} \cdot \boldsymbol{f}^{\alpha} + \boldsymbol{j}^{\beta} \cdot \boldsymbol{f}^{\beta}.$$

α and β relate to the behaviour under $t \to -t$

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$$\boldsymbol{j}^\alpha = \boldsymbol{L}^{\alpha\alpha}\,\boldsymbol{f}^\alpha + \boldsymbol{L}^{\alpha\beta}\,\boldsymbol{f}^\beta, \qquad \boldsymbol{j}^\beta = \boldsymbol{L}^{\beta\alpha}\,\boldsymbol{f}^\alpha + \boldsymbol{L}^{\beta\beta}\,\boldsymbol{f}^\beta$$

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- Onsager reciprocity relations:

$$\boldsymbol{L}^{\alpha\alpha} = \left[\boldsymbol{L}^{\alpha\alpha}\right]^{\mathsf{T}}, \qquad \boldsymbol{L}^{\beta\beta} = \left[\boldsymbol{L}^{\beta\beta}\right]^{\mathsf{T}}, \qquad \boldsymbol{L}^{\alpha\beta} = -\left[\boldsymbol{L}^{\beta\alpha}\right]^{\mathsf{T}}.$$

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- Curie principle: compatibility with the symmetries of the phase

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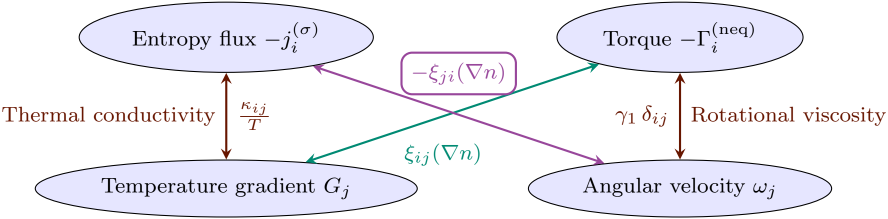

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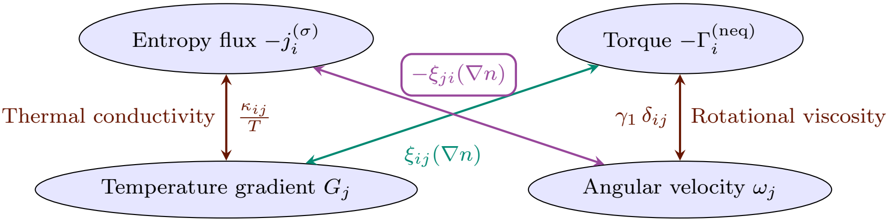

This system respects the Onsager reciprocity relations.

Simplified version of the phenomenological equations

$$\Gamma_i^{\mathrm{TM}} = \xi_{ij}(\nabla n)\, G_j$$

- Tensorial expression of $\xi_{ij}(\nabla n)$ quite complicated:

$$\begin{aligned} \xi_{ij}(\nabla n) = -\left[\nu + \bar{\xi}_2\left(\epsilon_{kpq}\, n_k\, n_{q,p}\right)\right] \delta_{ij}^{\perp} + \bar{\xi}_1\, n_{l,l}\, n_k\, \epsilon_{ikj} \\ + \bar{\xi}_3\left(\epsilon_{ikp}\, n_k\, n_q\, n_{p,q}\right) n_j + \bar{\xi}_4\, \epsilon_{ikp}\, n_k\left(n_{j,p} - n_{p,j}\right). \end{aligned}$$

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- We assume a simplified form for the torque $\mathbf{\Gamma}^{\mathrm{TM}}$ ($\bar{\xi}_i = \xi$):

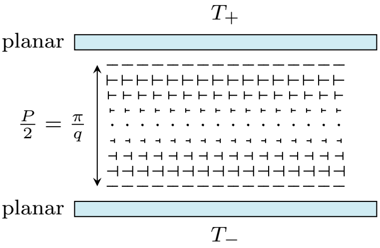
$$\mathbf{\Gamma}^{\mathrm{TM}} = \nu\,\boldsymbol{G}^{\perp} + \xi\,(\boldsymbol{G}\cdot\boldsymbol{\nabla})\,\boldsymbol{n}$$

  - ⋆ ν: Leslie effect, allowed only in cholesterics.
  - ⋆ ξ: Akopyan & Zel'dovich effect, allowed both in nematics and cholesterics.

Outline

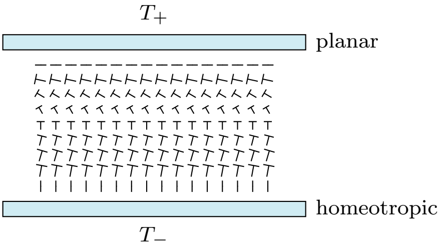
Translationally invariant configurations (TIC)

Planar TIC



$$\omega_p = -\bar{\nu}\, J_\nu[\boldsymbol{n}]\, \Delta T, \text{ with } \bar{\nu} \equiv \nu - \xi\, q$$

Mixed TIC



$$\omega_m = -\left(\bar{\nu}\, J_\nu[\boldsymbol{n}] + \xi\, q\, J_\xi[\boldsymbol{n}]\right) \Delta T$$

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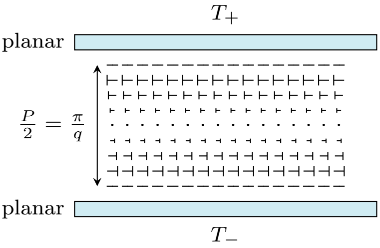

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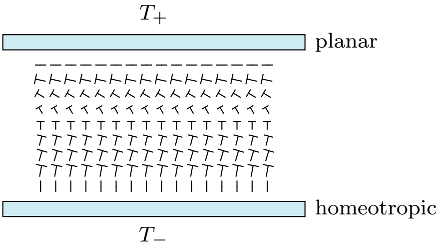
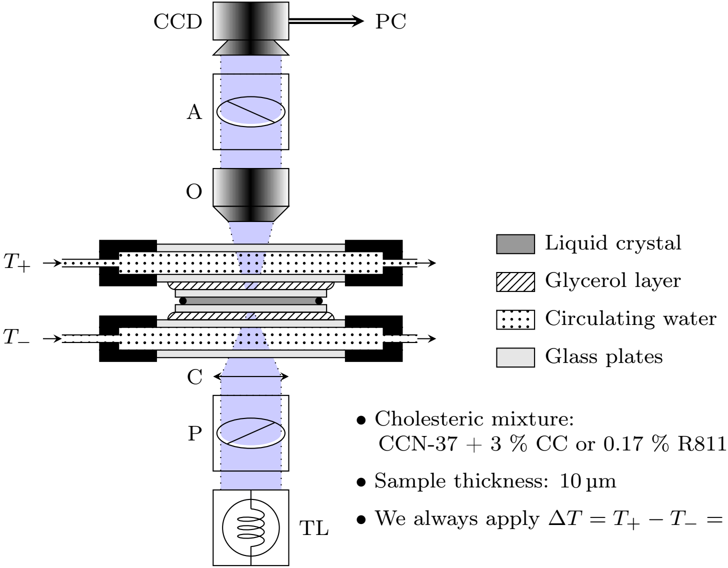


$\omega_m = -\left(\bar{\nu}\, J_\nu[\boldsymbol{n}] + \xi\, q\, J_\xi[\boldsymbol{n}]\right) \Delta T$

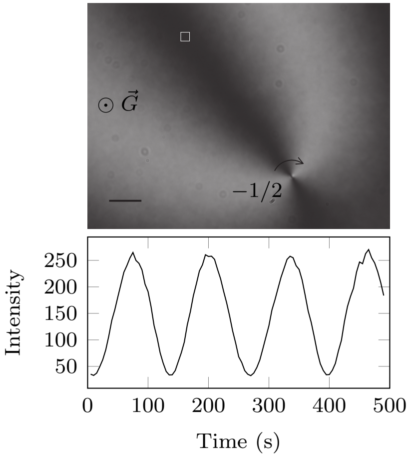
$\omega_p \neq \omega_m$ in general $\Rightarrow$ we can deduce $(\bar{\nu}, \xi)$ from (ω_p, ω_m).

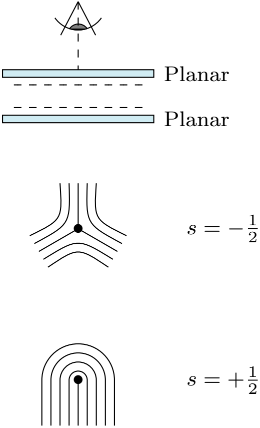
Experimental setup



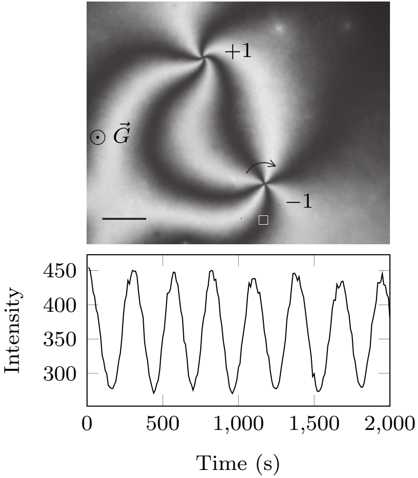
- Cholesteric mixture: CCN-37 + 3 % CC or 0.17 % R811 ($P \approx 63\,\mu m$).
- Sample thickness: 10 µm
- We always apply $\Delta T = T_+ - T_- = 40\,K$.

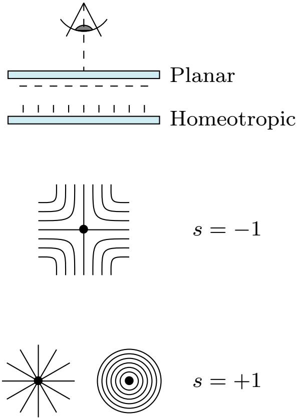
Rotation of the planar TIC



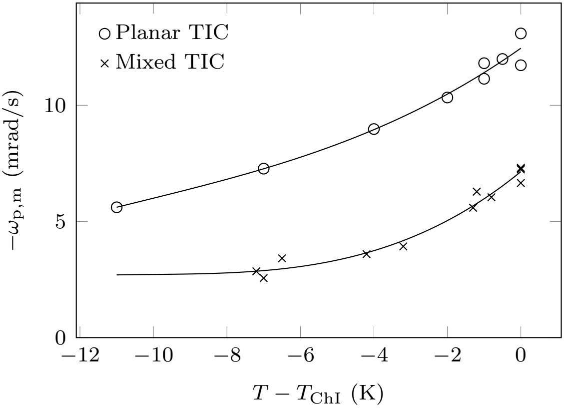


Rotation of the mixed TIC





Angular velocities with a mixture of CCN-37 + 3 % CC



$\omega_p \neq \omega_m \Rightarrow$ we can measure $\bar{\nu}$ and ξ.

Final results

From our theoretical model, we calculate just below T_{ChI}:

	CC	R811
$\bar{\nu}/q$ (fN/K)	11 ± 1	3 ± 1
ξ (fN/K)	-35 ± 17	-25 ± 17

$$\Rightarrow (\bar{\nu}/q)_{\mathrm{CC}} \neq (\bar{\nu}/q)_{\mathrm{R811}} \text{ and } \xi_{\mathrm{CC}} \approx \xi_{\mathrm{R811}}.$$

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- We have confirmed theoretically and experimentally the existence of the Akopyan & Zel'dovich coupling.
- Typical order of magnitude of 10 fN/K.
- $\bar{\nu}/q$ depends on the chiral dopant, contrary to ξ.

Outline

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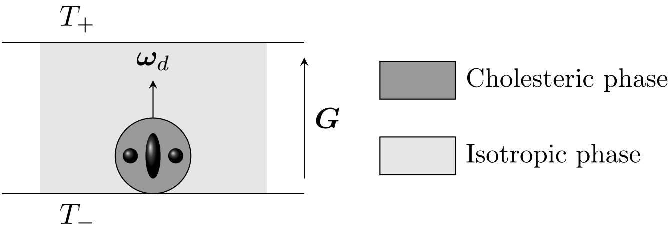
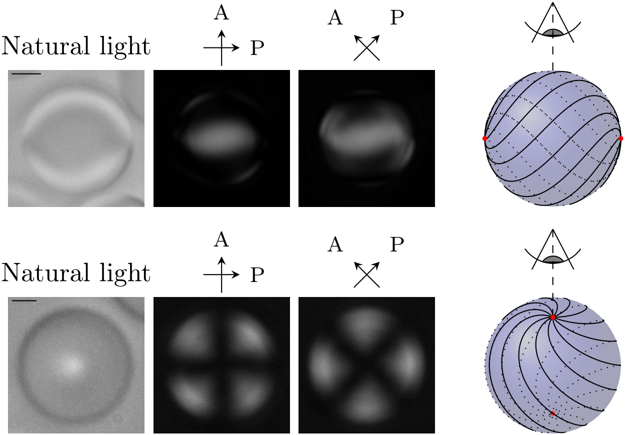
A quick reminder



Questions

- What is the texture inside the droplets?
- Is there a scaling law for the angular velocity ω_d?

Optical micrographs



CCN-37 + R811 or CC: planar anchoring at the droplet interface

Numerical minimization of the free energy

- Unit director field: $\boldsymbol{n}_s = \underset{\boldsymbol{n},\ |\boldsymbol{n}|=1}{\operatorname{argmin}}\ F[\boldsymbol{n}]$

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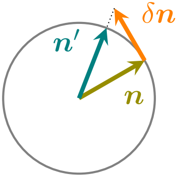
$$F[\boldsymbol{n}] \to f(\boldsymbol{N}) \text{ with } \boldsymbol{N} = \begin{pmatrix} \boldsymbol{n}_1 \\ \vdots \\ \boldsymbol{n}_M \end{pmatrix}$$

- Iterative minimization:
  - ⋆ $\boldsymbol{N}_{(k)}$ verifying $\boldsymbol{n}_\beta \cdot \boldsymbol{n}_\beta = 1$
  - ⋆ $\boldsymbol{N}_{(k+1)} = \mathcal{P}\left(\boldsymbol{N}_{(k)} + \boldsymbol{\delta N}\right)$, where $\mathcal{P}$ is the normalization operation $\boldsymbol{n}_\beta \to \boldsymbol{n}_\beta / |\boldsymbol{n}_\beta|$
  - ⋆ $\boldsymbol{\delta N}$ found with the truncated conjugate gradient algorithm (trust region strategy)



Essential properties of this algorithm

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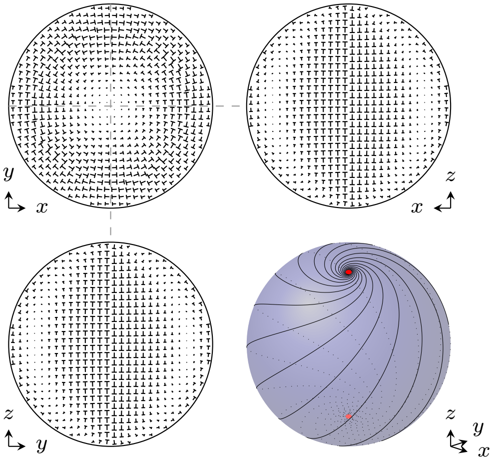
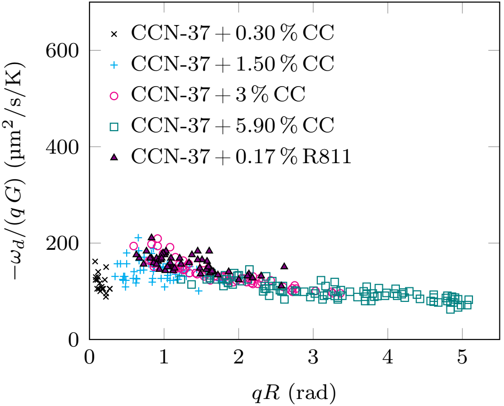
- Quadratic convergence near the minimum
- Unit director field at each step: only $2M$ degree of freedoms in 3D

Results for a typical droplet of CCN-37



$R = 19\,\mu\text{m}$, $P = 30\,\mu\text{m}$, $l_a = 0.82\,\mu\text{m}$ (planar anchoring)

Angular velocities



- $-\frac{2\pi}{\omega_d} \approx 20 - 2000\,\text{s}.$

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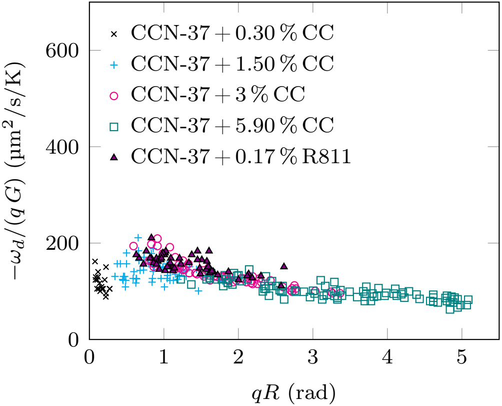

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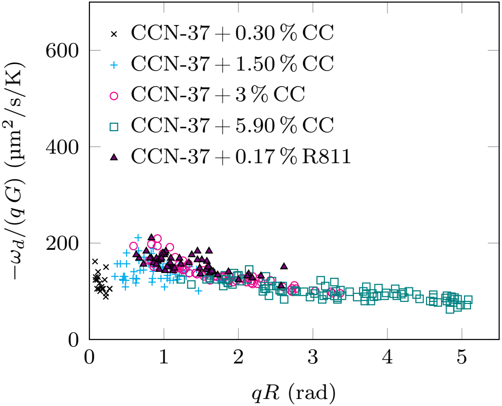

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Data obtained with a different chiral dopant rescale on the same master curve.

New problematic

Rotation because of the microscopic or macroscopic chirality?

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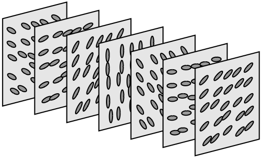
- microscopic chirality $\Leftrightarrow$ chiral molecules

New problematic

Rotation because of the microscopic or macroscopic chirality?

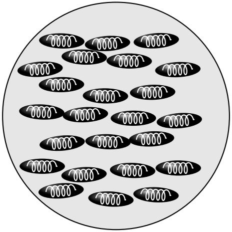
- microscopic chirality ⇔ chiral molecules

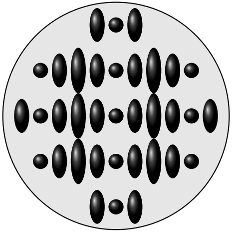
- macroscopic chirality ⇔ twisted texture (helix in at least one direction)



New problematic

Possible tests:





$\left\{\begin{array}{l}\text{chiral molecules} \leftrightarrow \text{cholesteric} \\ \text{no macroscopic twist (compensated)}\end{array}\right.$

Thermal gradient ⇒ no rotation

$\left\{\begin{array}{l}\text{no chiral molecules} \leftrightarrow \text{nematic} \\ \text{macroscopic twist}\end{array}\right.$

Thermal gradient ⇒ rotation?

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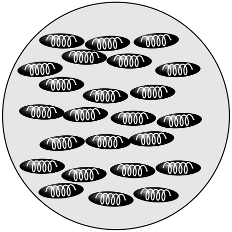
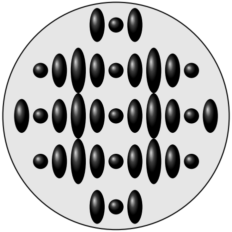


chiral molecules ↔ cholesteric
no macroscopic twist (compensated)

Thermal gradient ⇒ no rotation



no chiral molecules ↔ nematic
macroscopic twist

Thermal gradient ⇒ rotation?

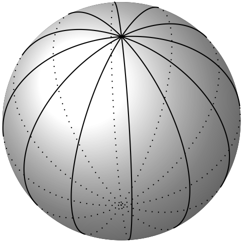
Question

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

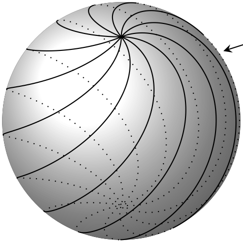
Outline

Stability of bipolar configuration

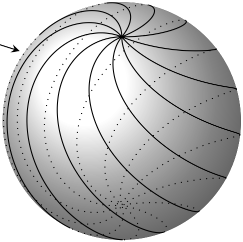
Topological constraint:
planar anchoring



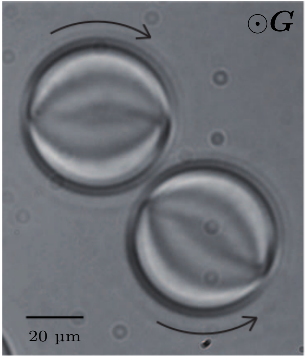
$K_2 \sim K_1 \sim K_3$
twist ∼ splay ∼ bend



$K_2 \ll K_1, \quad K_3$
twist ≪ splay, bend



Rotation of twisted bipolar droplets



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

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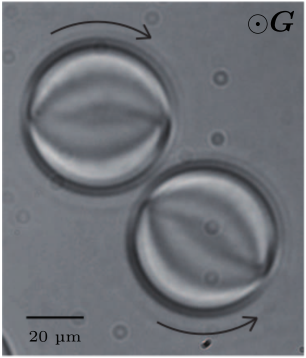

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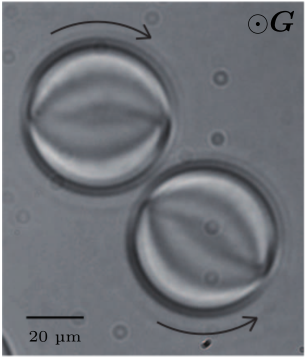

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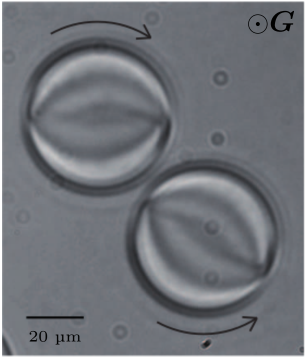

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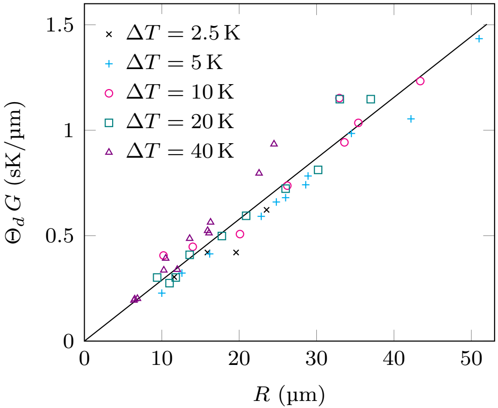
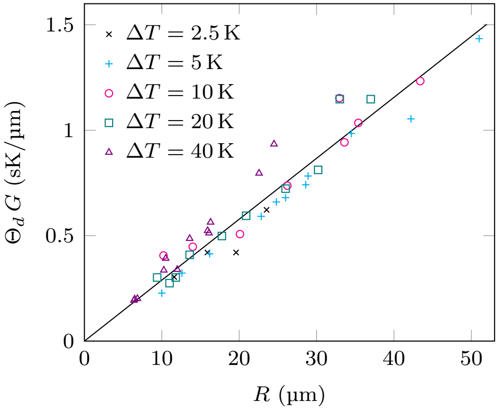
Rotation only due to the twist of the director field

Rotation periods



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

Rotation periods



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

Outline

Thermomechanical model of the Lehmann effect

- Rotation "in block" of the texture without flow:

$$\frac{\mathrm{D}}{\mathrm{D}\Theta}\Big(F[\boldsymbol{n}]\Big) = 0$$

D/DΘ: change rate associated with a rotation of the texture.

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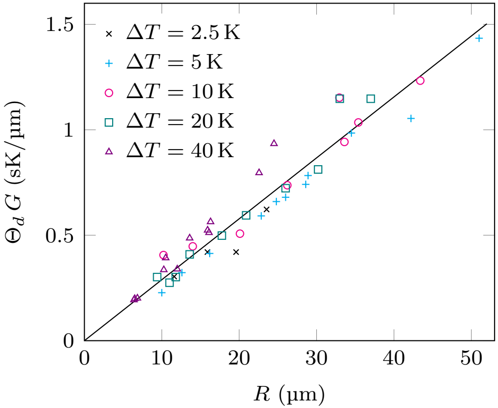
- Using the torque equation $\boldsymbol{\Gamma}^{(\mathrm{E})} + \boldsymbol{\Gamma}^{(\mathrm{V})} + \boldsymbol{\Gamma}^{(\mathrm{TM})} = \mathbf{0}$, we can find a prediction for the angular velocity of the droplet.

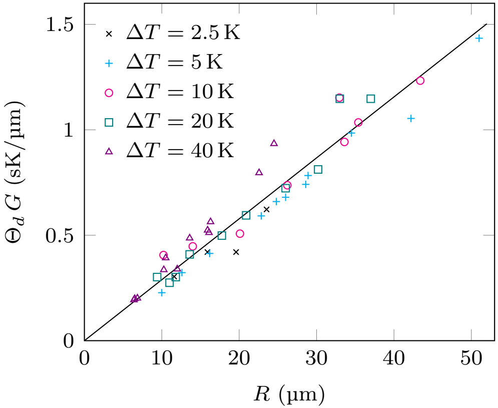
Results with nematic droplets of water + 30 % SSY



- Theoretical prediction:

$$\Theta_d\,G = \frac{R}{\xi\,L_\xi[\boldsymbol{n}]}$$

Results with nematic droplets of water + 30 % SSY



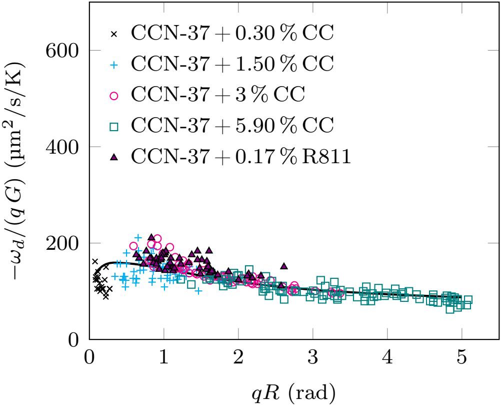
- Theoretical prediction:

$$\Theta_d\,G = \frac{R}{\xi\,L_\xi[\boldsymbol{n}]}$$

- Numerical fit: $(\xi)_{\mathrm{fit}} \approx 70\,\mathrm{pN/K}$
- Measured value in CCN-37 below T_{ChI}: $\xi \approx 30\,\mathrm{fN/K}$

Qualitative agreement. Quantitative agreement?

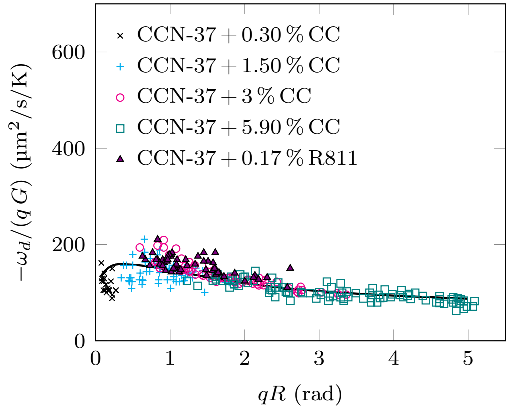
Results with cholesteric droplets of CCN-37



- Theoretical prediction:

$$\frac{\omega_d}{q\,G} = [\bar{\nu}/q]\,L_\nu[\boldsymbol{n}] + \xi\,L_\xi[\boldsymbol{n}]$$

Results with cholesteric droplets of CCN-37



- Theoretical prediction:

$$\frac{\omega_d}{q\,G} = [\bar{\nu}/q]\,L_\nu[\boldsymbol{n}] + \xi\,L_\xi[\boldsymbol{n}]$$

- Numerical fit:
  $(\bar{\nu}/q)_{\text{fit}} \approx 1.7\,\text{pN/K}$
  $(\xi)_{\text{fit}} \approx -3\,\text{pN/K}$
  $\Rightarrow$ values 100 bigger than those measured below T_{ChI} ($\sim 10\,\text{fN/K}$).

Qualitative and quantitative disagreement.

Outline

Main results

- Experimental and theoretical confirmation of the existence of the (corrected) Akopyan & Zel'dovich coupling terms.

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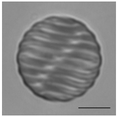
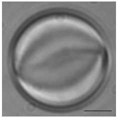
- The Lehmann effect can be observed in twisted nematic droplets.
- The Leslie, Akopyan & Zel'dovich thermomechanical terms cannot explain the Lehmann effect.

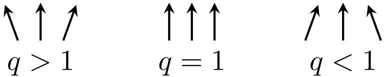
Postdoc project

Simulation of natural and polarized light micrographs of nematic and cholesteric droplets.



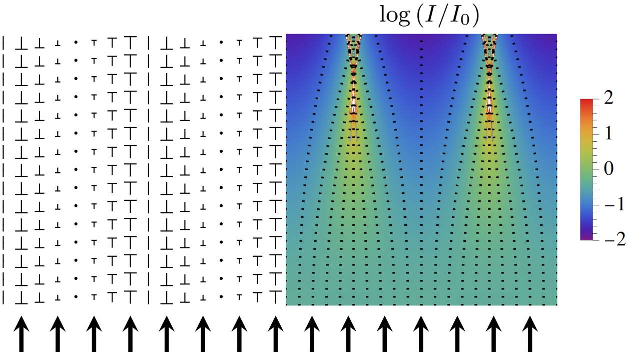


- Hamiltonian ray-tracing method: $\dot{\boldsymbol{\eta}} = \boldsymbol{\Omega} \cdot \boldsymbol{\nabla}_{\boldsymbol{\eta}} \left(H^{e,o} \right)$, with $\boldsymbol{\eta} = (\boldsymbol{r}, \boldsymbol{k})$.
- Conserved quantities along a ray: $\left(\sqrt{q}\, n_{\text{eff}}\, E \right)$ and $\left(\sqrt{q}\, B \right)$, with q the geometrical spreading.



Proof of concept

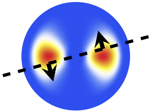
Deviation of extraordinary rays in a cholesteric slab:



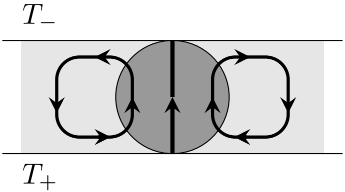
Thank you!

In the future...

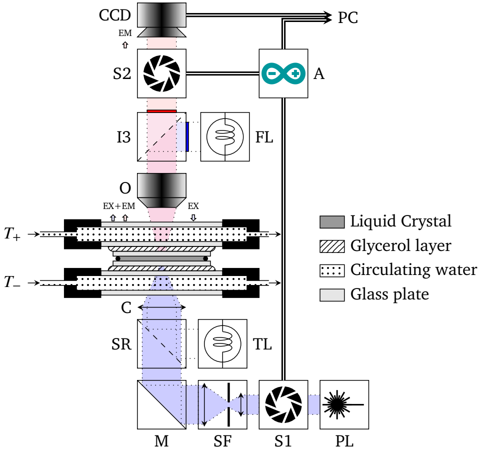
- Why the droplets stay spherical in a temperature gradient?
- Importance of the theoretical model of A. Dequidt in the Lehmann effect?

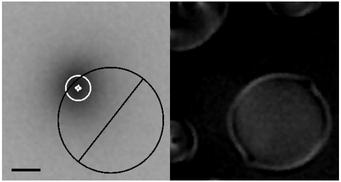


- Convective rolls? Marangoni and/or thermohydrodynamic couplings?



Photobleaching experiment





- LC mixture doped with fluorescent molecules
- Gaussian beam of a laser focalized near a rotating droplet