Mathematical Models for Chromonic Liquid Crystals

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Summary

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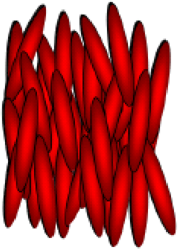

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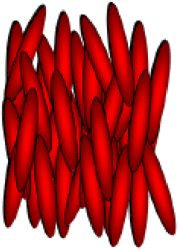

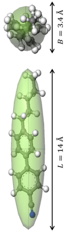

molecular height $\sim 1\,\mathrm{nm}$

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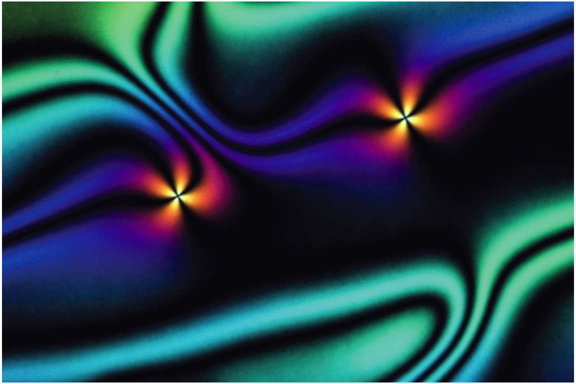
- Defects are ***optically*** detectable.



Courtesy of O.D. Lavrentovich

early statistical theories

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- Ericksen (1961): first general system of balance laws.
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W is ***even***

$$W(-\boldsymbol{n}, -\nabla\boldsymbol{n}) = W(\boldsymbol{n}, \nabla\boldsymbol{n})$$

Frank's formula

The most general frame-indifferent and even function W that is at most ***quadratic*** in $\nabla \boldsymbol{n}$ was obtained by Frank (1958),

$$W_{\mathrm{F}}(\boldsymbol{n}, \nabla \boldsymbol{n}) = \frac{1}{2} K_{11} (\operatorname{div} \boldsymbol{n})^2 + \frac{1}{2} K_{22} (\boldsymbol{n} \cdot \operatorname{curl} \boldsymbol{n})^2 + \frac{1}{2} K_{33} |\boldsymbol{n} \times \operatorname{curl} \boldsymbol{n}|^2 + K_{24} \left(\operatorname{tr}(\nabla \boldsymbol{n})^2 - (\operatorname{div} \boldsymbol{n})^2 \right)$$

K_{ij} Frank's elastic constants

K_{11} splay constant

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Ericksen's inequalities

$$W_{\mathrm{F}}(\boldsymbol{n}, \nabla \boldsymbol{n}) \geqq 0 \quad \text{a.e. } \forall\ \boldsymbol{n} \in H^1(\mathscr{B};\mathbb{S}^2) \text{ iff}$$

$$K_{33} \geqq 0, \quad K_{22} \geqq K_{24}, \quad K_{11} \geqq K_{24} \geqq 0$$

Ericksen (1966)

Elementary Distortion Modes

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distortion decomposition

$$\nabla \boldsymbol{n} = -\boldsymbol{b} \otimes \boldsymbol{n} + \frac{1}{2} T \mathbf{W}(\boldsymbol{n}) + \frac{1}{2} S \mathbf{P}(\boldsymbol{n}) + \mathbf{D}$$

$S := \operatorname{div} \boldsymbol{n}$ splay scalar

$T := \boldsymbol{n} \cdot \operatorname{curl} \boldsymbol{n}$ twist pseudoscalar

$\boldsymbol{b} := \boldsymbol{n} \times \operatorname{curl} \boldsymbol{n}$ bend vector

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octupolar splay

$$\mathbf{D} = q(\boldsymbol{n}_1 \otimes \boldsymbol{n}_1 - \boldsymbol{n}_2 \otimes \boldsymbol{n}_2)$$

q ***positive*** eigenvalue of $\mathbf{D}$

identity

$$2q^2 = \mathrm{tr}(\nabla \boldsymbol{n})^2 + \frac{1}{2}T^2 - \frac{1}{2}S^2$$

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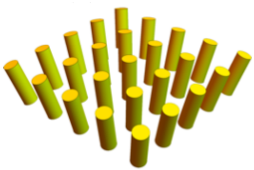
$$B^2 := \boldsymbol{b} \cdot \boldsymbol{b}$$

Mode illustration

The four independent modes can be illustrated pictorially.

Selinger (2021)

splay mode



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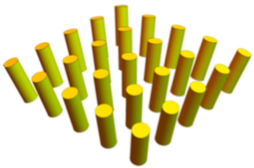

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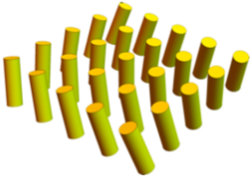
(double) twist mode



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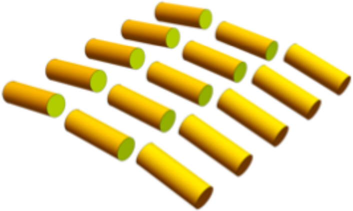

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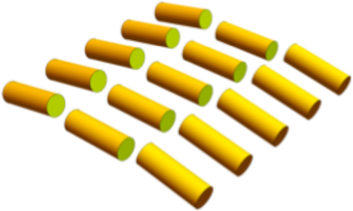

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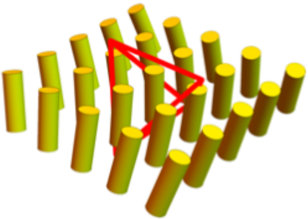

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$\boldsymbol{n}_\perp := \mathbf{N}\boldsymbol{n}$ unit vector orthogonal to $\boldsymbol{n}$
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- Only ***hyperbolic*** geometries can host ***uniform*** distortions in 2D.

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lost in space

For such a field, we could not tell ***where we are*** in space only by sampling the local nematic distortion.

3D Euclidean space

There are only ***two families*** of possible uniform distortions that fill 3D Euclidean space:

$$S = 0, \quad T = 2q, \quad b_1 = b_2 = b$$
$$S = 0, \quad T = -2q, \quad b_1 = -b_2 = b$$

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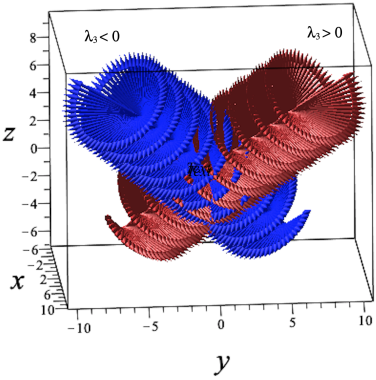
heliconical fields

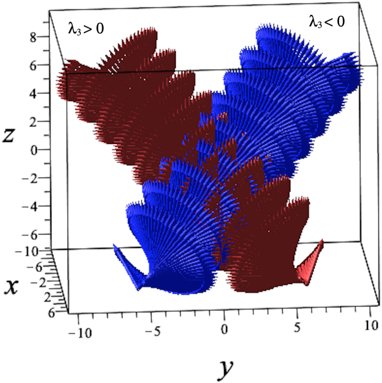
The director $\boldsymbol{n}$ makes a constant ***conical*** angle θ with the ***axis*** of a ***helix*** with ***pitch*** p:

$$\cos\theta = \frac{|b|}{\sqrt{b^2 + 2q^2}}$$

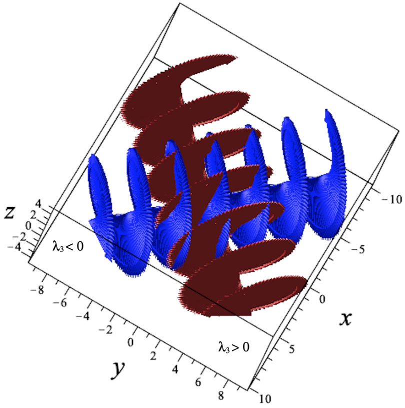
$$p = \frac{2\pi}{|\lambda_3|} \qquad \lambda_3 = \pm\left(2q + \frac{b^2}{q}\right)$$



$b/q = -1$



$b/q = 1$



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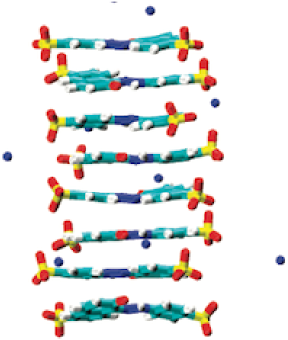
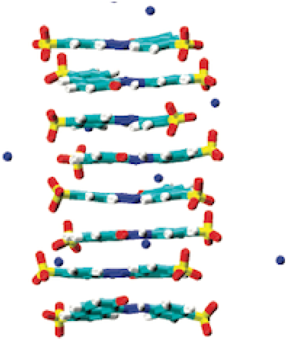


Chami & Wilson (2010)

Chromonic Liquid Crystals

Chromonic liquid crystals are ***lyotropic***. They are composed of ***plank-like*** molecules with a poly-aromatic core and polar peripheral groups, aggregated in columnar ***stacks***.



Chami & Wilson (2010)

molecular diameter:
$1 - 2\,\mathrm{nm}$
columnar height:
$10 - 100\,\mathrm{nm}$

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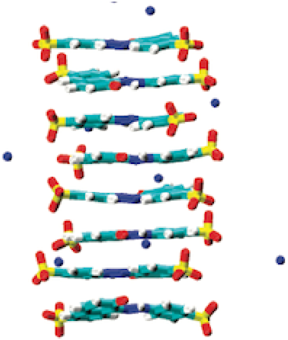

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Chami & Wilson (2010)

Chromonics are formed by certain ***dyes***, ***drugs***, and short ***nucleic-acid*** oligomers in ***aqueous*** solutions.

Anomalous Ground State

When subject to ***degenerate*** planar anchoring conditions on the lateral boundary $\partial\mathscr{B}$ of a ***cylinder***,

$$\boldsymbol{n} \cdot \boldsymbol{\nu} \equiv 0$$

$\boldsymbol{\nu}$ outer unit normal to $\partial\mathscr{B}$

the director does ***not*** spontaneously acquire the ***uniform alignment***, but tend to take on either of the two ***double twists*** compatible with symmetry.

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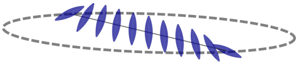
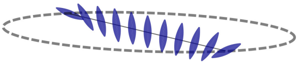
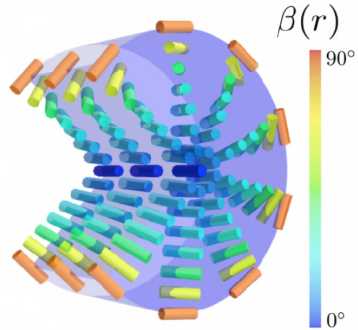




escaped-twist fields



Davidson, Kang, Jeong, Still, Collings, Lubensky, & Yodh (2015)

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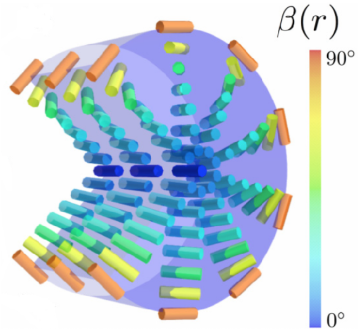

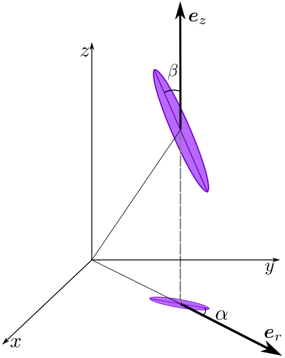

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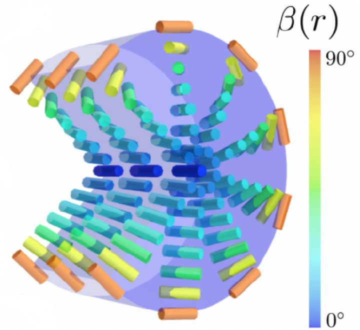

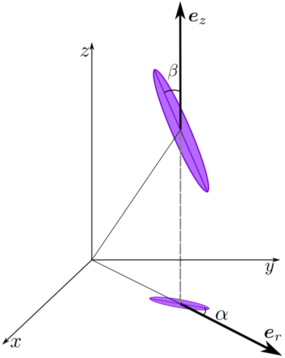

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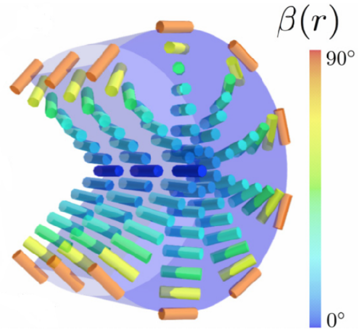

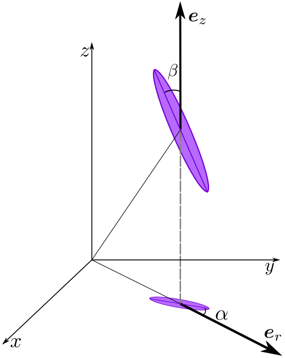

DAVIDSON, KANG, JEONG, STILL, COLLINGS, LUBENSKY, & YODH (2015)

SSY @25°C, $\phi = 0.18$:

$K_{11} = 4.3\,\mathrm{pN}$ $K_{22} = 0.7\,\mathrm{pN}$ $K_{33} = 6.1\,\mathrm{pN}$ $K_{24} = 15.7\,\mathrm{pN}$

ZHOU, NASTISHIN, OMELCHENKO, TORTORA, NAZARENKO, BOIKO, OSTAPENKO, HU, ALMASAN, SPRUNT, GLEESON, & LAVRENTOVICH (2012)

violation of Ericksen's inequality $K_{24} > K_{22}$

reduced free-energy functional

In cylindrical coordinates (r, ϑ, z), we set

$$\boldsymbol{n} = \sin\beta(r)\boldsymbol{e}_\vartheta + \cos\beta(r)\boldsymbol{e}_z \quad \text{and} \quad \alpha = \frac{\pi}{2}$$

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Then

$$\mathcal{F}[\beta] := \frac{\mathscr{F}_{\mathrm{F}}[\boldsymbol{n}]}{2\pi K_{22} L} = \int_0^1 \left(\frac{\rho\beta'^2}{2} + \frac{1}{2\rho}\cos^2\beta\sin^2\beta + \frac{k_3}{2\rho}\sin^4\beta \right) \mathrm{d}\rho$$
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$$k_3 := \frac{K_{33}}{K_{22}} \quad k_{24} := \frac{K_{24}}{K_{22}} \quad \text{with} \quad K_{22} > 0$$

$$\rho := \tfrac{r}{R}$$

R radius of the cylinder

L height of the cylinder

equilibrium distortions

For $k_{24} > 1$,

$$\beta_{\mathrm{ET}}(\rho) = \arctan\left(\frac{2\sqrt{k_{24}(k_{24}-1)}\rho}{\sqrt{k_3}\left[k_{24}-(k_{24}-1)\rho^2\right]}\right)$$

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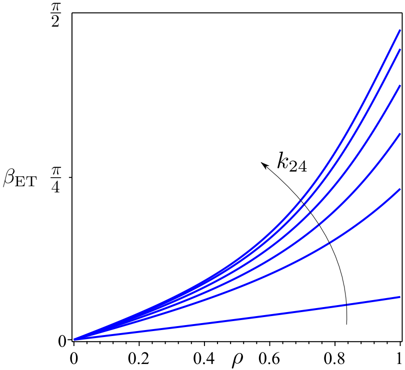

$k_3 = 10 \quad k_{24} = 1 \to 22$

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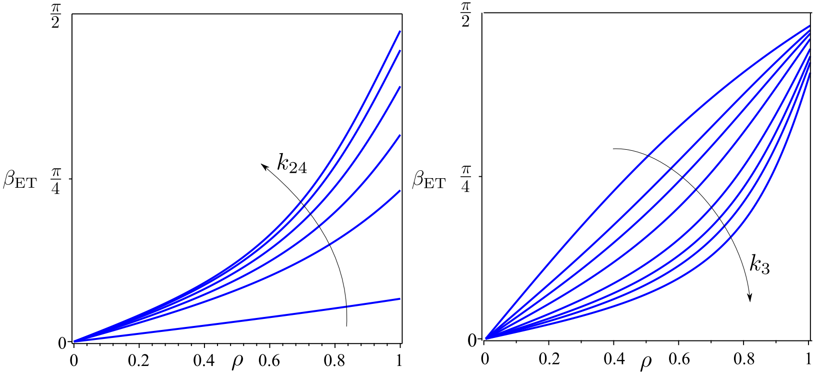

$k_3 = 10 \quad k_{24} = 1 \to 22$ $\qquad$ $k_{24} = 10 \quad k_3 = 1 \to 30$

role of boundary conditions

Degenerate planar anchoring conditions save the day (and ground state), as the K_{24}-integral in $\mathscr{F}_{\mathrm{F}}$ can be given the form

$$-K_{24}\int_{\partial\mathscr{B}}\left(\kappa_1 n_1^2+\kappa_2 n_2^2\right)\mathrm{d}A$$

κ_i principal curvatures of $\partial\mathscr{B}$

n_i components of $\boldsymbol{n}$ along the principal direction of curvature

Koning, van Zuiden, Kamien, & Vitelli (2014)

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distortion characteristics

For $\beta=\beta_{\mathrm{ET}}$,

$$\begin{aligned}
T &= \frac{1}{r}\sin\beta\cos\beta(\sqrt{1+\tan^2\beta}+1)\\
q &= \frac{1}{2r}|\sin\beta|\cos\beta(\sqrt{1+\tan^2\beta}-1)\\
b_1 &= -\frac{1}{\sqrt{2}r}\sin^2\beta=-b_2
\end{aligned}$$

along the axis

$$S = q = b_1 = b_2 = 0 \quad T = \pm \frac{4\sqrt{k_{24} - 1}}{R\sqrt{k_3 k_{24}}}$$

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- Can we really get away with a violation to Ericksen's inequality?

Long & Selinger (2022)

Local Stability

Both ***escaped-twist*** field are ***locally stable***, as can be shown elaborating on the following general formula.

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second variation

$$
\begin{aligned}
\delta^2\mathscr{F}_{\mathrm{F}}(\boldsymbol{n})[\boldsymbol{v}] = \int_{\mathscr{B}}\Big\{ & (K_{11}-2K_{24})\left[(\operatorname{div}\boldsymbol{v})^2 - v^2(\operatorname{div}\boldsymbol{n})^2 - (\operatorname{div}\boldsymbol{n})\,\boldsymbol{n}\cdot\nabla v^2\right] \\
& + K_{22}\Big[(\boldsymbol{v}\cdot\operatorname{curl}\boldsymbol{n}+\boldsymbol{n}\cdot\operatorname{curl}\boldsymbol{v})^2 \\
& + 2(\boldsymbol{n}\cdot\operatorname{curl}\boldsymbol{n})(\boldsymbol{v}\cdot\operatorname{curl}\boldsymbol{v}-v^2\boldsymbol{n}\cdot\operatorname{curl}\boldsymbol{n})\Big] \\
& + K_{33}\Big[\,|\boldsymbol{v}\times\operatorname{curl}\boldsymbol{n}+\boldsymbol{n}\times\operatorname{curl}\boldsymbol{v}|^2 \\
& + (\boldsymbol{n}\times\operatorname{curl}\boldsymbol{n})\cdot(\boldsymbol{v}\times\operatorname{curl}\boldsymbol{v}-2v^2\boldsymbol{n}\times\operatorname{curl}\boldsymbol{n}-\nabla v^2)\Big] \\
& + 2K_{24}\left[\operatorname{tr}(\nabla\boldsymbol{v})^2 - v^2\operatorname{tr}(\nabla\boldsymbol{n})^2 + \boldsymbol{n}\times\operatorname{curl}\boldsymbol{n}\cdot\nabla v^2\right]\Big\}\,\mathrm{d}V
\end{aligned}
$$

$$\boldsymbol{v}\cdot\boldsymbol{n}=0$$

Paparini & Virga (2022)

Drop Paradoxes

In ***rigidly confined*** systems subject to ***degenerate planar*** anchoring conditions, the free-energy functional $\mathscr{F}_{\mathrm{F}}$ is well-behaved, despite the ***violation*** of Ericksen's inequality.

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free-boundary problems

However, the violation of Ericksen's inequality would have noxious consequences in ***chromonic*** droplets surrounded by their ***isotropic phase***.

free-energy functional

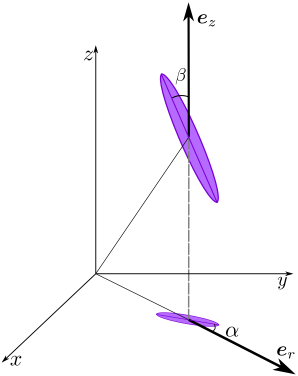
$$\mathscr{F}[\boldsymbol{n},\mathscr{B}] := \int_{\mathscr{B}} W_{\mathrm{F}}(\boldsymbol{n},\nabla\boldsymbol{n})\,\mathrm{d}V + \gamma A(\partial\mathscr{B})$$

γ surface tension
A area measure

isoperimetric constraint

$$V(\mathscr{B}) = V_0$$

director and tactoid representations



$$\boldsymbol{n} = \cos\alpha(z)\sin\beta(\rho)\boldsymbol{e}_r + \sin\alpha(\rho)\sin\beta(\rho)\boldsymbol{e}_\vartheta + \cos\beta(\rho)\boldsymbol{e}_z$$

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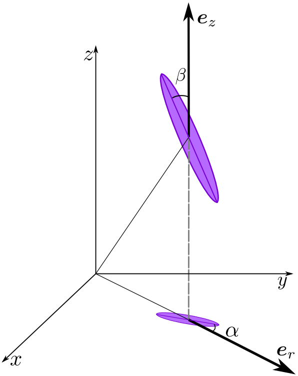

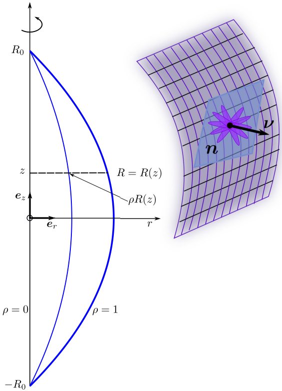

$$\boldsymbol{n} = \cos\alpha(z)\sin\beta(\rho)\boldsymbol{e}_r + \sin\alpha(\rho)\sin\beta(\rho)\boldsymbol{e}_\vartheta + \cos\beta(\rho)\boldsymbol{e}_z$$

$$\boldsymbol{\nu} = \frac{\boldsymbol{e}_r - R'\boldsymbol{e}_z}{\sqrt{1+R'^2}}.$$

$$\cos\alpha(z) = \frac{R'}{\tan\beta(1)}$$

minimizing sequence

Letting $\beta = \beta_{\mathrm{ET}}$, in a wide class of shapes, we estimate

$$\mathcal{F} := \frac{\mathscr{F}[\boldsymbol{n}, \mathscr{B}]}{2\pi K_{22} R_{\mathrm{e}}} \leqq \mu \mathcal{F}_{\mathrm{ET}}[\beta_{\mathrm{ET}}] + \sqrt{\frac{8}{3}} \upsilon \sqrt{\mu} + \mathcal{O}\left(\frac{1}{\sqrt{\mu}}\right)$$

Paparini & Virga (2022)

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PAPARINI & VIRGA (2022)

$\mathcal{F}_{\mathrm{ET}}$ dimensionless free energy stored in a ***cylinder***

$\mu := \frac{R_0}{R_{\mathrm{e}}}$ dimensionless tactoid ***height***

R_{e} equivalent ***radius*** (of the sphere of volume V_0)

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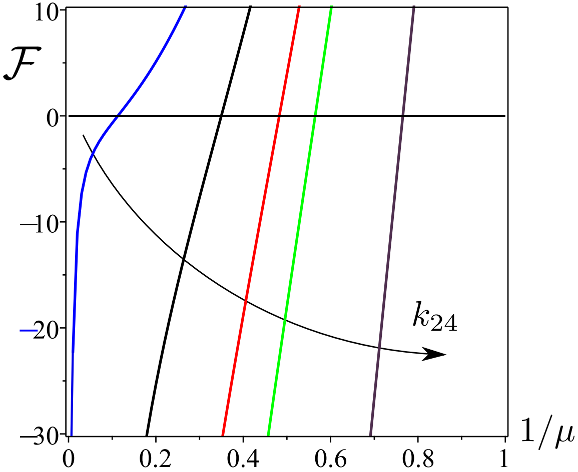
but

For $K_{24} > K_{22}$,

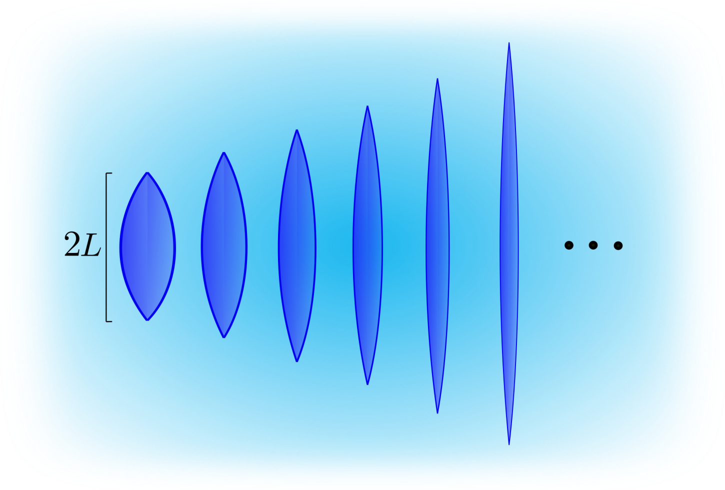
$$\mathcal{F}_{\mathrm{ET}}[\beta_{\mathrm{ET}}] < 0$$

. . . and so



$k_{11} = k_{33} = 30 \quad k_{24} = 1 \to 22$

...which means that



$\mu = \frac{L}{R_e}$

PAPARINI & VIRGA (2022)

disintegration paradox

Confining the drop would not save it from disintegration, as for μ sufficiently large,

$$\mathcal{F}_n \approx \mathcal{F}_{\mathrm{ET}}[\beta_{\mathrm{ET}}]\mu 2^n \to -\infty \quad \text{as} \quad n \to \infty$$

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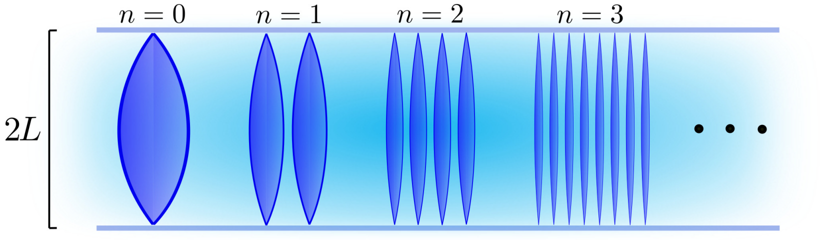

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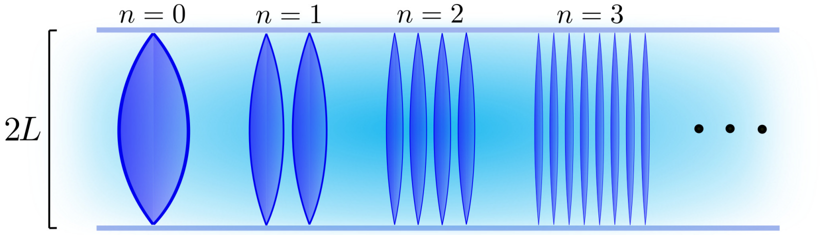

remark

None of these drop instabilities has been observed experimentally so far (and they are ***unlikely*** to be observed in the future).

Quartic Twist Theory

A possible way out (admittedly, not the only one) would be to correct Frank's curvature energy density with a ***quartic*** term,

$$W_{\mathrm{chr}} = \frac{(K_{11} - K_{24})}{2} S^2 + \frac{(K_{22} - K_{24})}{2} T^2 + 2K_{24} q^2 + \frac{K_{33}}{2} B^2 + \underbrace{\frac{K_{22} a^2}{4}}_{K_{44}} T^4$$

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This theory would induce an intrinsic, ***degenerate*** double twist $\pm T_0$ in the ***ground state***, still ***incompatible*** with a ***uniform*** extension in space, and thus condemned to ***frustration***,

$$S = B = q = 0, \quad T = \pm T_0 := \pm \frac{1}{a} \sqrt{\frac{(K_{24} - K_{22})}{K_{22}}}$$

Conclusions

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- Would the critical dimension of the ***singular set*** be affected by the ***quartic twist*** term?
- It is nearly needless to say that no ***dynamical theory*** is available specifically for ***chromonics***; neither can we predict what role would play in it the proposed quartic twist energy.

Acknowledgements

Collaboration

S. Paparini

Discussion

O. D. Lavrentovich

P. Palffy-Muhoray

J. V. Selinger

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