

Liquid Crystals: Lecture 1

Basic properties

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Acknowledgements

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The lectures are based on the book by
Maurice Kleman and O. D. Lavrentovich
“Soft Matter Physics: An Introduction” (Springer, 2003)

Content: Lecture 1

☐ Types of liquid crystalline order

- Nematic
- Cholesteric and blue phases
- Twist bend nematic
- Smectic A
- Lyotropic and Chromonic LCs

☐ Basic Physics

- Dielectric anisotropy
- Surface anchoring
- Elasticity
- Frederiks effect and modern LCDs

☐ Optics

- Birefringence
- Polarizing microscopy: 2D imaging
- Fluorescence Confocal Polarizing microscopy: 3D imaging

Content: Lecture 2

□ Topological defects and droplets

- Disclinations in uniaxial nematics
 - Singular
 - Nonsingular
- Homotopy classification
- Drops and Conservation laws of topological defects
- Cholesteric droplets, Dirac monopole
- Chromonic tactoids
 - Broken chiral symmetry
 - Wulff construction for liquid crystals

□ Lamellar phases

- Free energy density for weak and strong perturbations
- Long-range character of deformations; undulations
- Focal conic domains

Content: Lecture 3

- Dynamics of director realignment
 - Anisotropy of viscosity
 - Coupling of director reorientation and flow
- Statics of colloids in nematic LC
 - Levitation
- Dynamics of colloids in nematic LC
 - Brownian motion
 - LC-enabled electrophoresis
 - LC with patterned orientation as an active medium
- Living LC
 - Swimming bacteria in LC; individual and collective effects

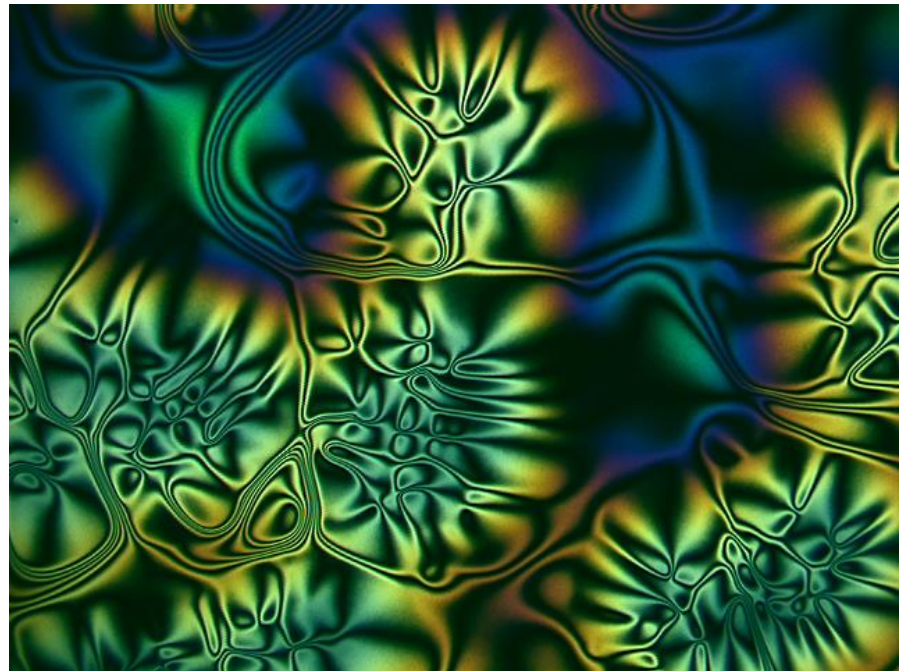
Liquid crystal:

a state of matter with long-range orientational order and

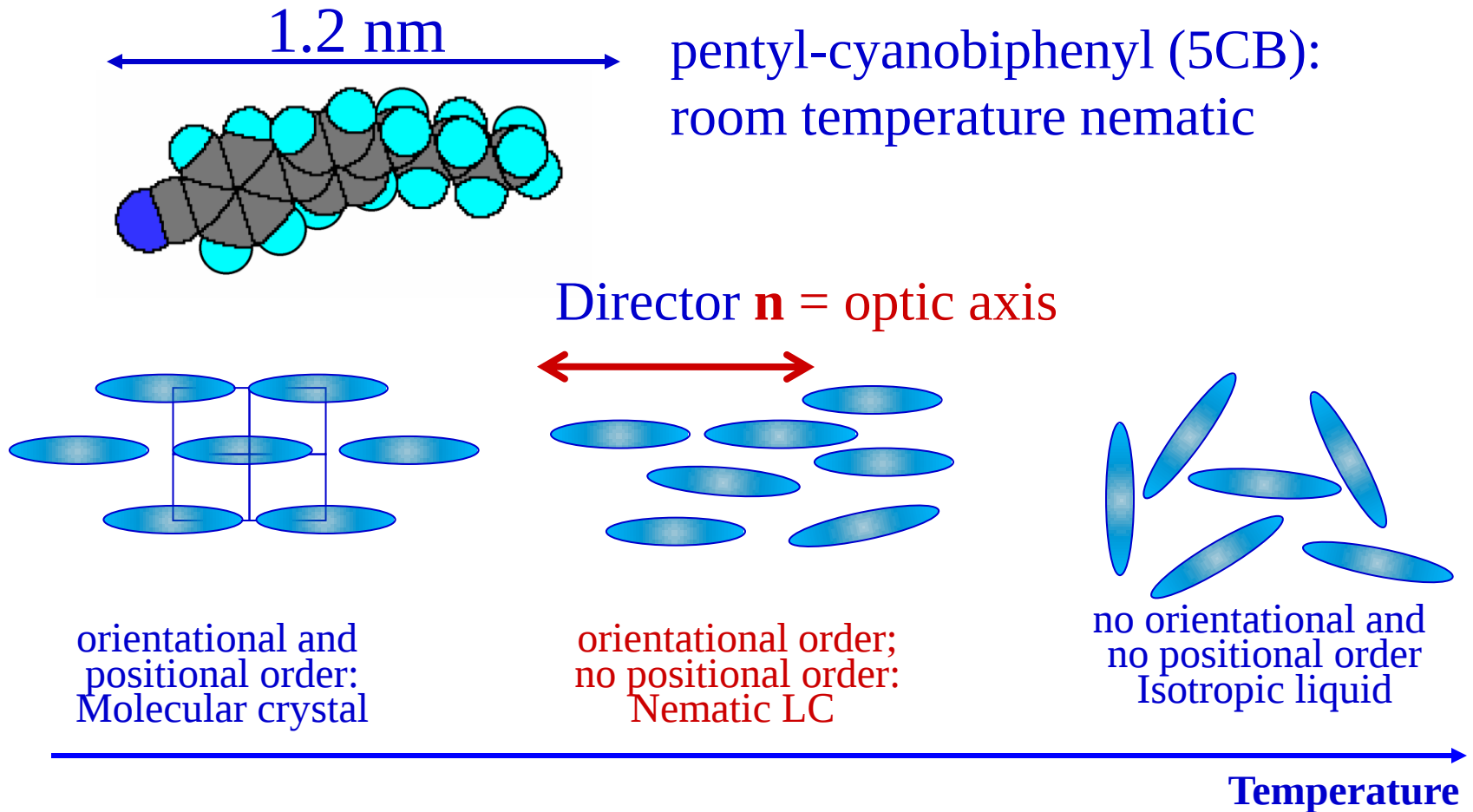
- complete (**nematic**)
- partial (**smectics, columnar phases**)

absence of long-range positional order of “building units”
(molecules, viruses, aggregates, etc.)

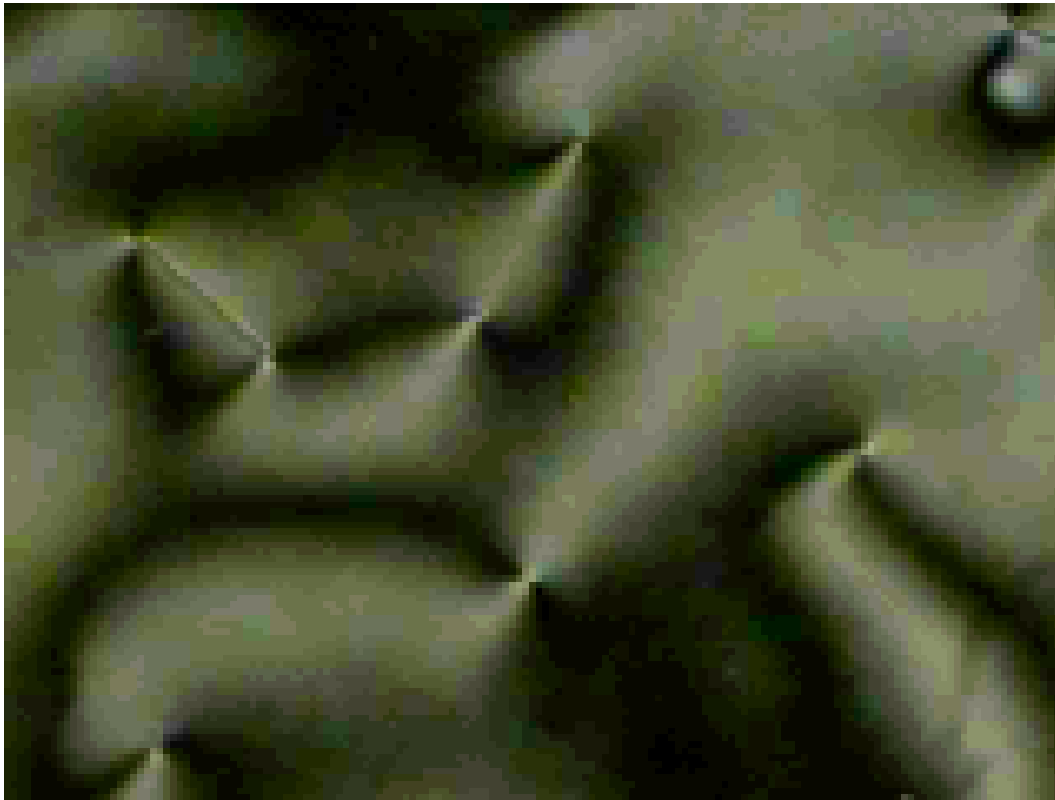
Our goal: Develop an intuitive understanding of what kind of new physics the orientational order brings to soft matter



Crystals, liquid crystals, liquids

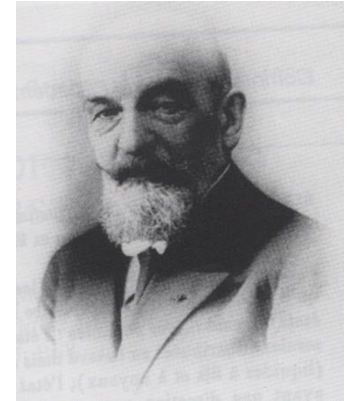


Nematic LC: $\nu\epsilon\mu\alpha$ =thread; aka “disclination”



50 microns $\longleftrightarrow$

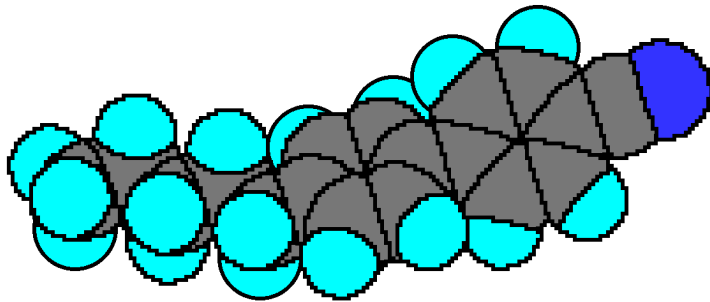
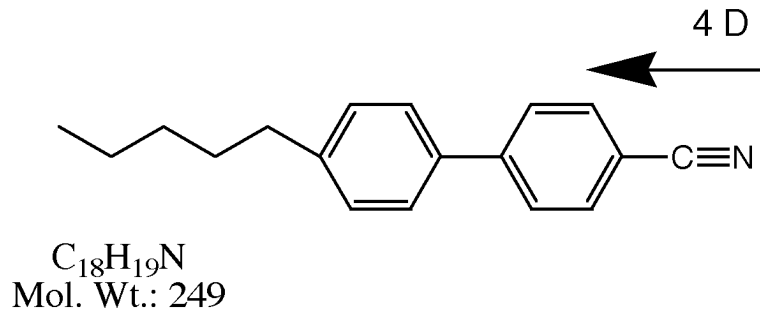
Nematic droplet-pancake sheared between two glass plates; polarizing optical microscope



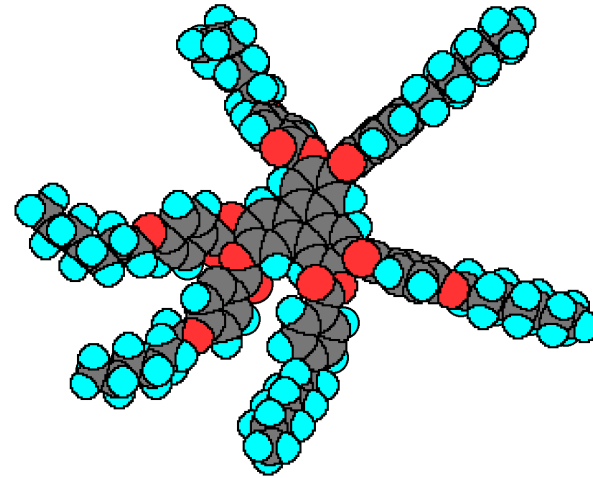
1922, G. Friedel:

Named “nematics”, the simplest LC, after observing linear defects, $\nu\epsilon\mu\alpha$ =thread, under a polarized light microscope

Nematic LC: Calamitic and Discotic

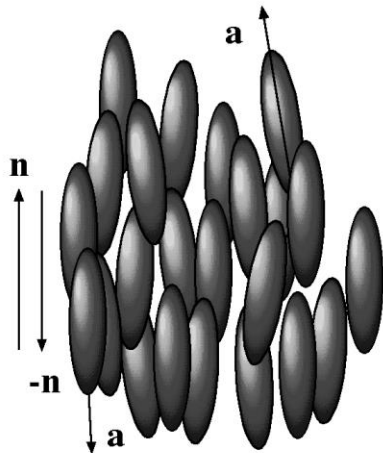


solid $\xleftrightarrow{22.5^{\circ}C}$ nematic $\xleftrightarrow{35^{\circ}C}$ isotropic

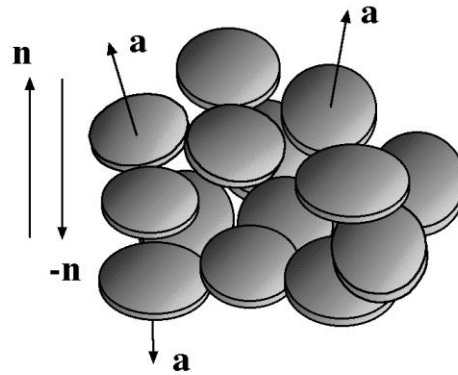


solid $\xleftrightarrow{168^{\circ}C}$ discotic nematic $\xleftrightarrow{253^{\circ}C}$ isotropic

Nematic LC: Calamitic and Discotic. Biaxial?

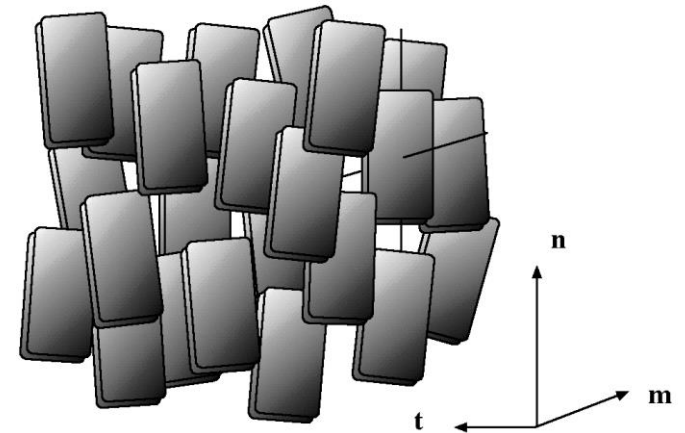


**Uniaxial
calamitic**



**Uniaxial
discotic**

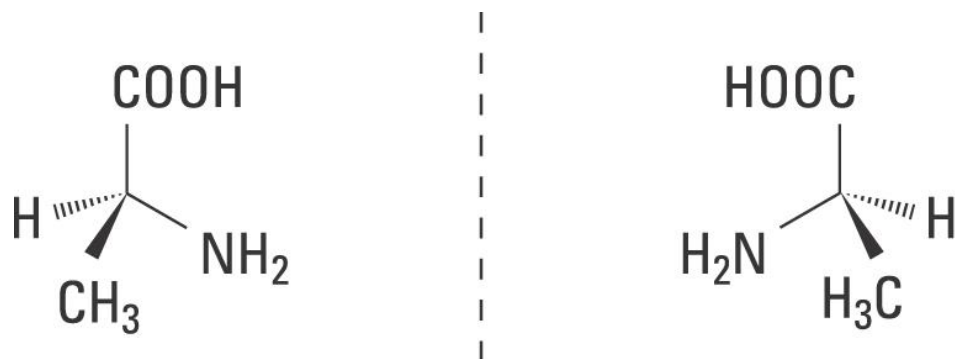
Existence established



Biaxial

Existence debated,
Uniaxial N often mimics biaxiality

Chiral molecule (does not overlap with its chiral image)

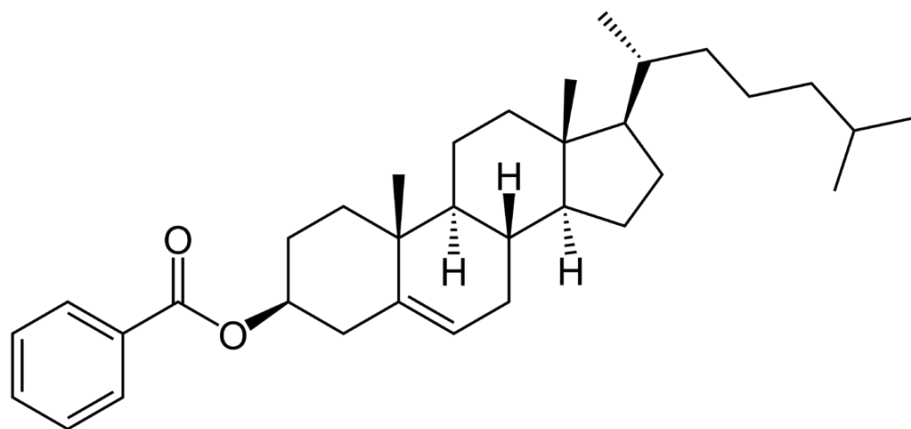


Carbon atom with 4 different attachments

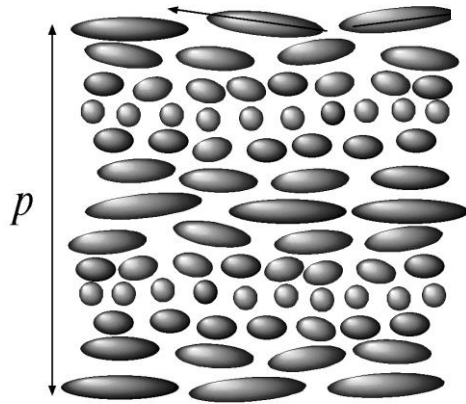
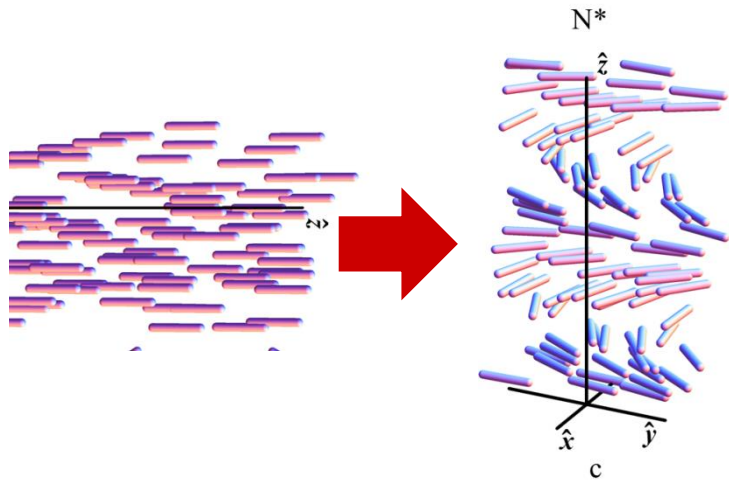
(*S*)-alanine mirror plane (*R*)-alanine

Cholesterol benzoate: Rod-like molecule with a chiral C atom;

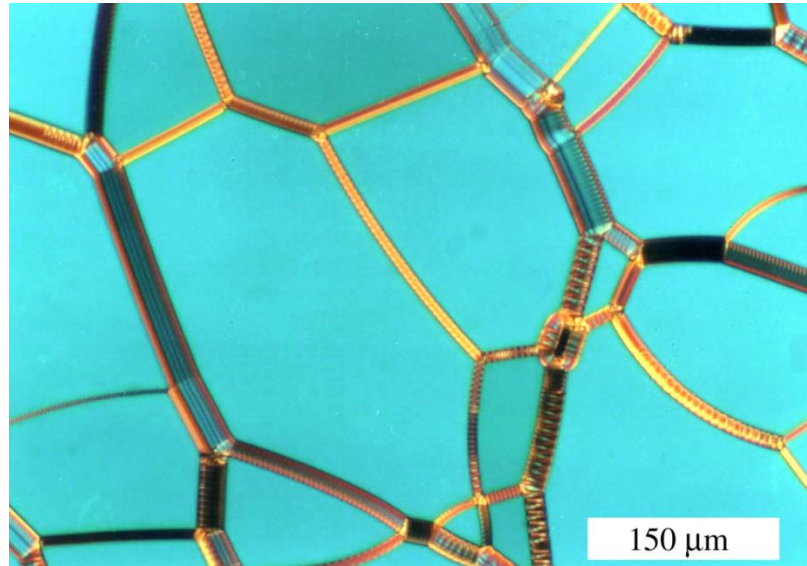
A similar cholesterol derivative was the subject of the first known publication on LCs, reporting double melting point and selective reflection of light, J. Planer, Ann Chem Pharm **118**, 25 (1861)



Add chiral molecules to nematic and obtain a cholesteric:



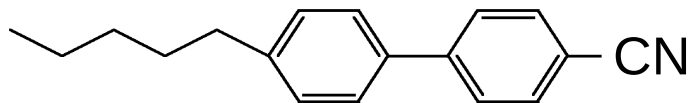
pitch $\sim 0.5 \mu\text{m}$



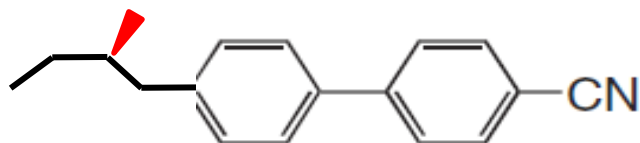
Where do the colors come from?

Bragg reflection at the periodic structure with period close to the wavelength of visible light
Why $P \gg$ molecular scale? Weakness of chiral contribution to the intermolecular potential, see Harris et al, Rev Mod Phys **71**, 1745 (1999)

Comparative chemistry

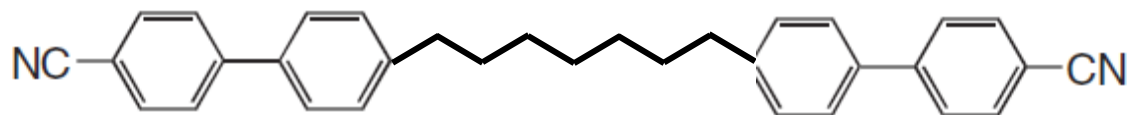


Nematic 5CB molecule

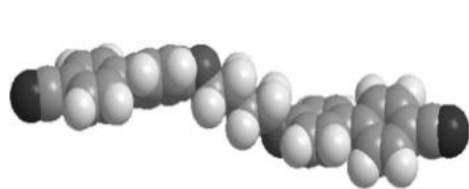


Cholesteric CB15 molecule

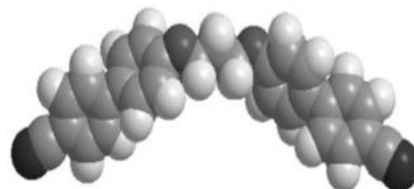
What would happen when two CB molecules are connected by a flexible aliphatic chain and form a “bimesogen” or “dimer”?



Answer: depends on odd-even character of aliphatic chain



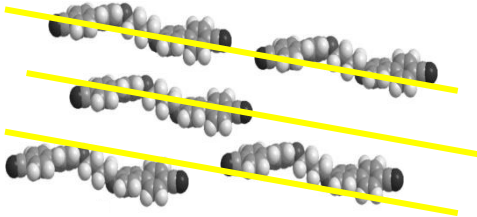
even number m of CH₂ groups



odd number m of CH₂ groups

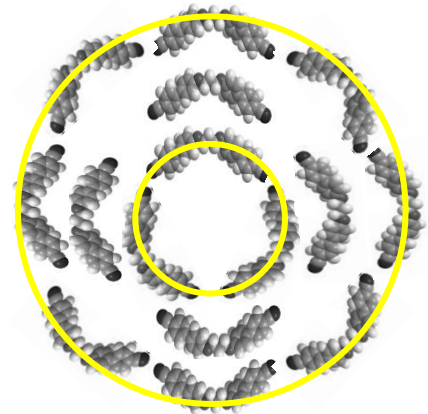
How can we pack these molecules in space?

Packing bimesogens

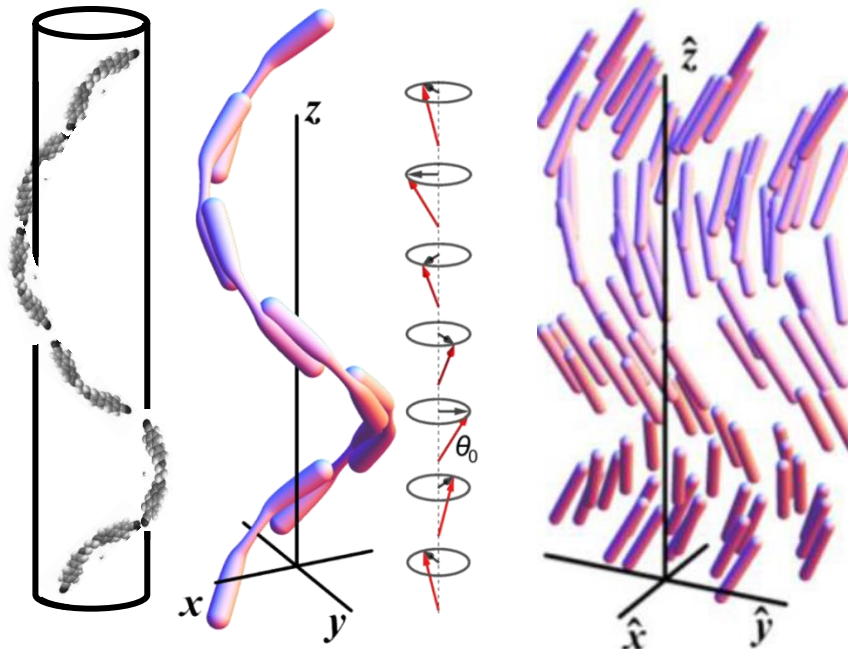


Even m , rod-like molecules:
Easy! Nematic!

Odd m , bent shape:
Difficult...cannot
sustain uniform bend in
2D...



Go to 3D:
Uniform bend
is achieved
through twist!



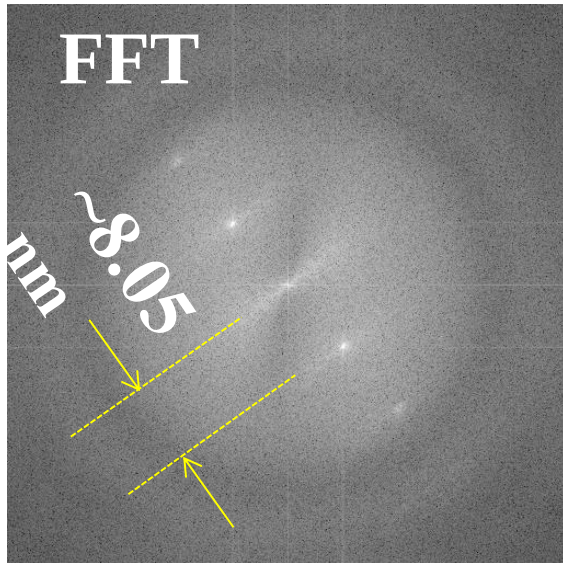
Predictions:

- R.B. Meyer (1973, Les Houches)
- R. Kamien, *J. Phys II* **6**, 461 (1996)
- I. Dozov, *EPL* **56**, 247 (2001)
- J. Selinger et al, *PRE* **87**, 052503 (2013)
- E. Virga, *PRE* **89** 052502 (2014)

$$\hat{\mathbf{n}} = (\sin \theta_0 \cos tz, \sin \theta_0 \sin tz, \cos \theta_0)$$

Freeze Fracture TEM, “planar” fractures

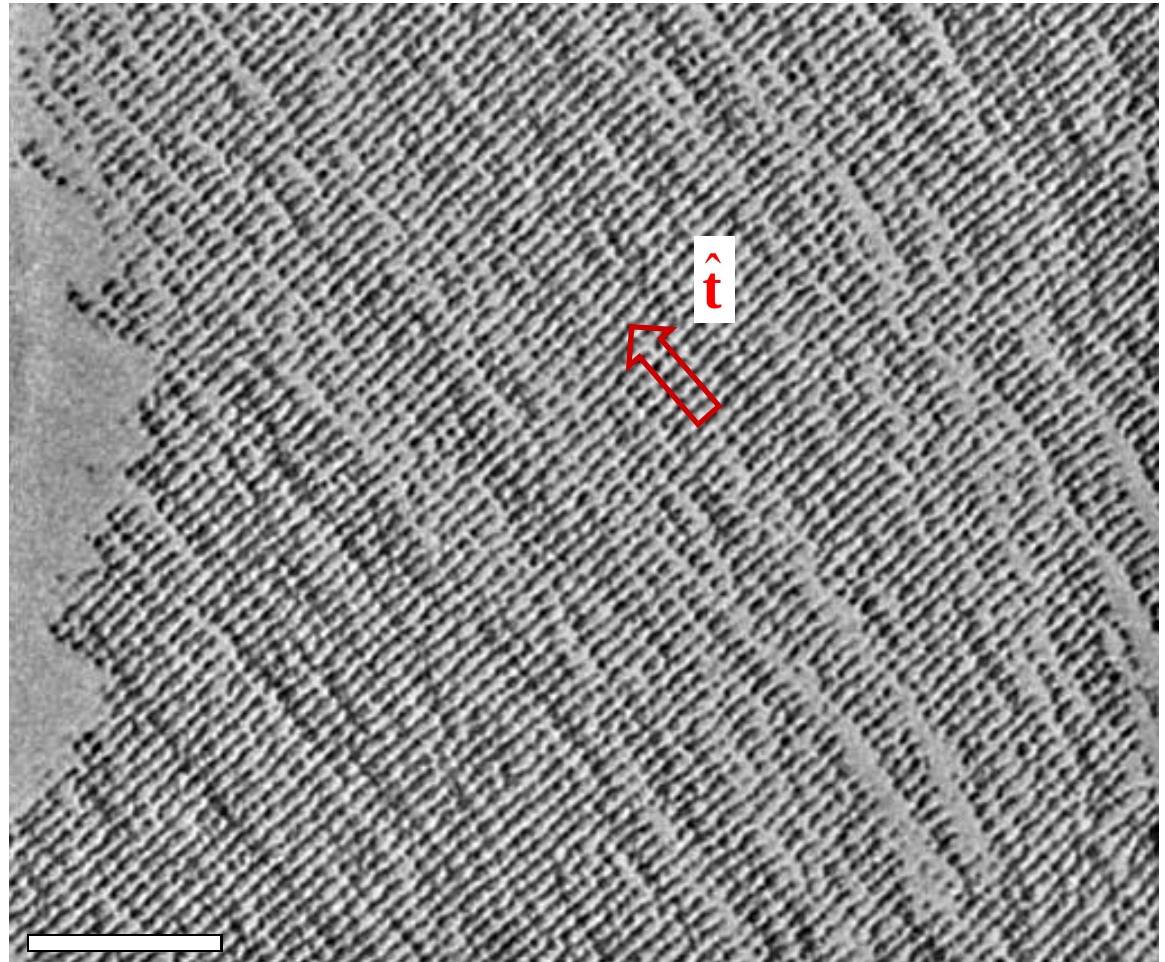
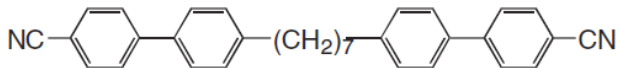
CB7CB quenched from the “X” temperature range and viewed under TEM



Period 8.05 nm

(molecular scale!!! Frozen rotations?)

Not visible under regular optical microscope; need an electron microscope)

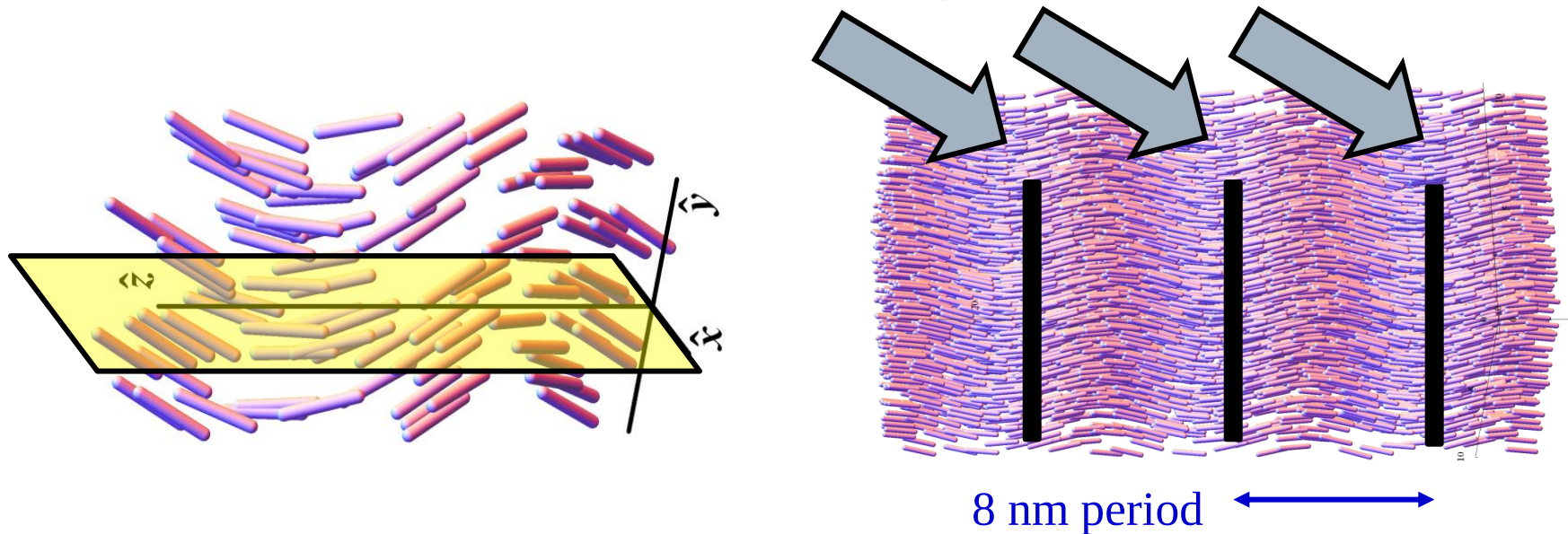


Chen, Walba, Clark et al, PNAS **110**, 15931 (2013)

Borshch, Gao et al, Nature Comm **4**, 2635 (2013)

Freeze Fracture TEM, “planar” fractures

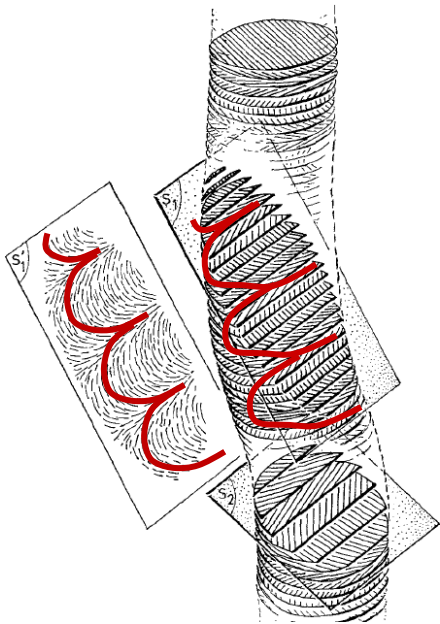
Freeze-Fractures in TEM: most likely to occur parallel to the long axes of molecules, thanks to the lowest molecular density



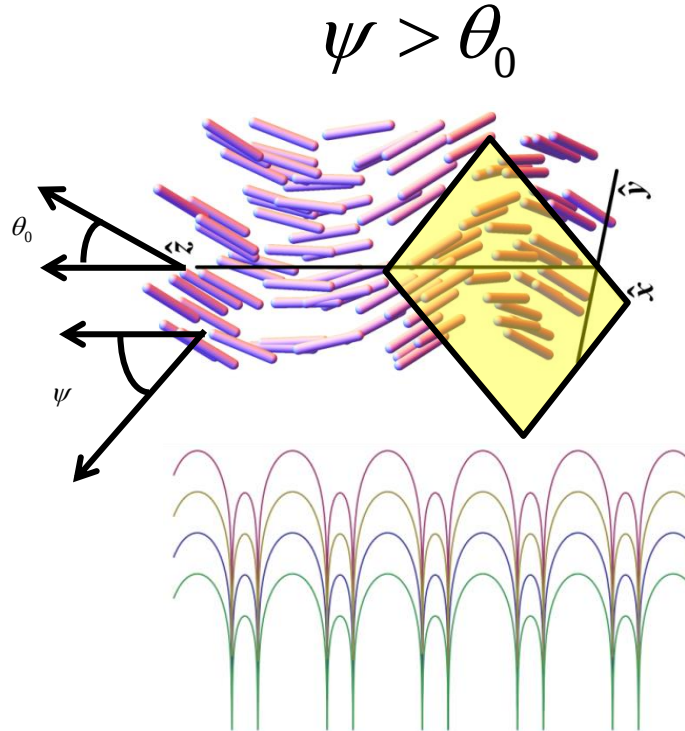
The 8 nm period is not visible in X-ray studies; thus there is no modulation of density

How do we know that the structure is indeed twist-bend as opposed to a simple cholesteric, or, say, splay-bend, also predicted to exist?

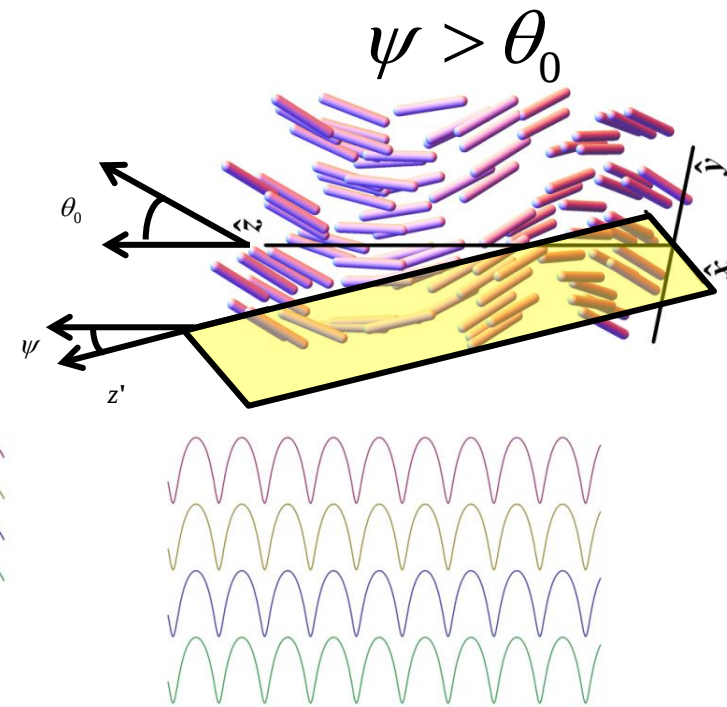
Tilted fractures: Bouligand arches are different in Ch and N_{tb}



Cholesteric:
Bouligand
arches are
symmetric

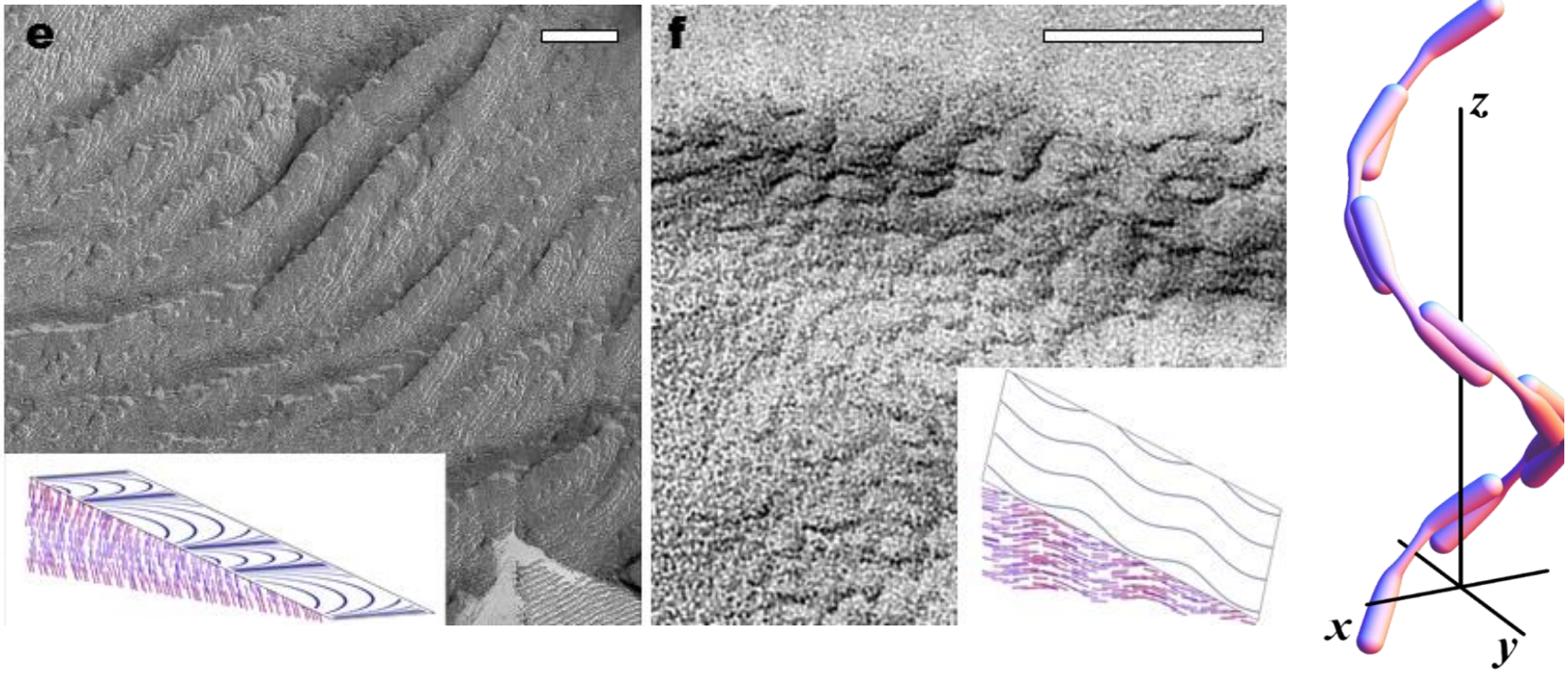


N_{tb} : Bouligand arches
are either asymmetric or
incomplete



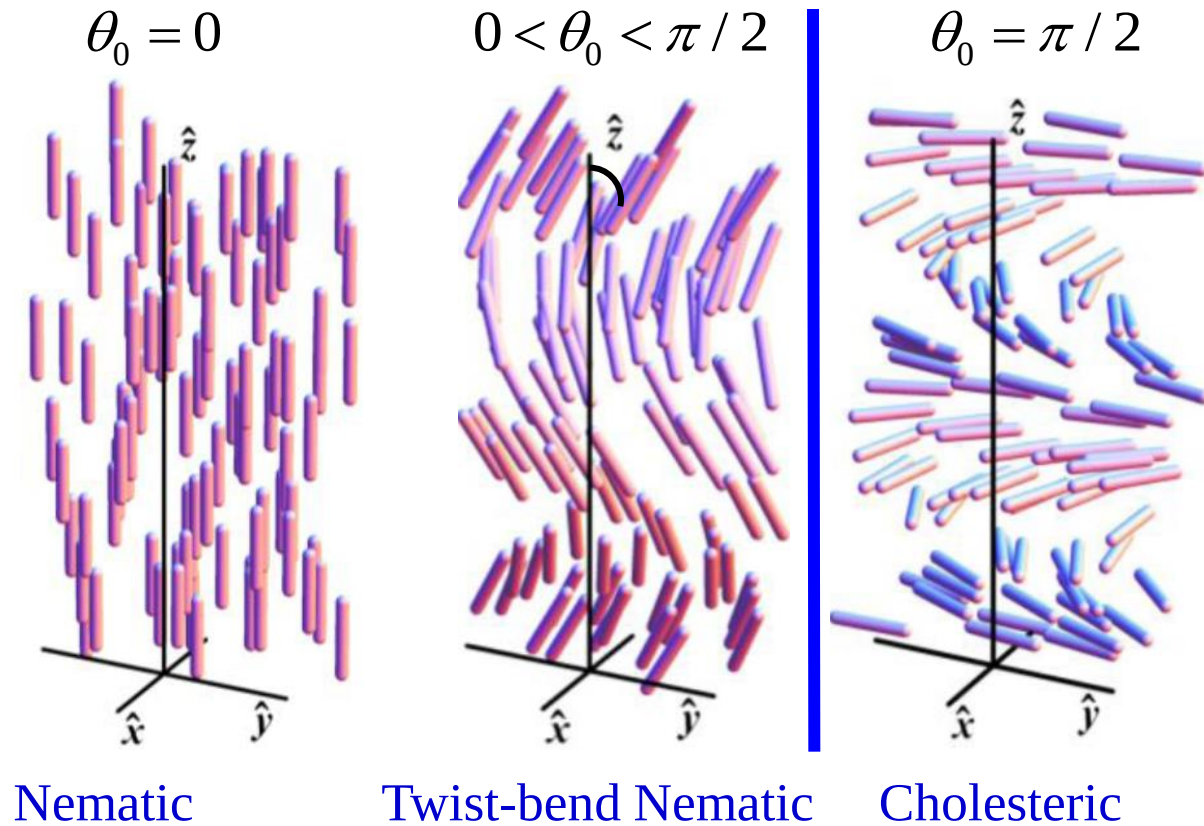
$$x_0 = \frac{\ln \left| 2 \cos \psi \sin \theta_0 \left[\sin(t_{tb} y' \sin \psi) - \tan \psi \cot \theta_0 \right] \right|}{t_{tb} \sin \psi \cos \psi}$$

FF TEM, Asymmetric Bouligand arches



Both types of N_{tb} asymmetric Bouligand arches are observed

Nematic and Cholesteric are merely two point ends of The Twist-Bend Nematic World...



$$\hat{\mathbf{n}} = (\sin \theta_0 \cos \varphi, \sin \theta_0 \sin \varphi, \cos \theta_0)$$

θ_0 : molecular tilt angle

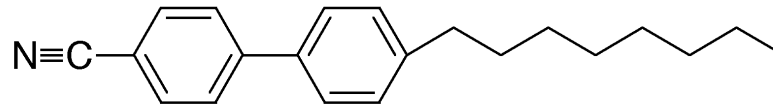
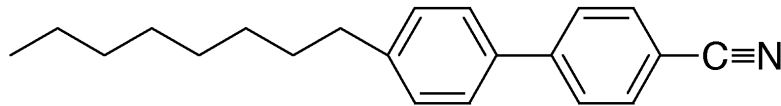
$$\varphi = tz$$

$$t = 2\pi / P$$

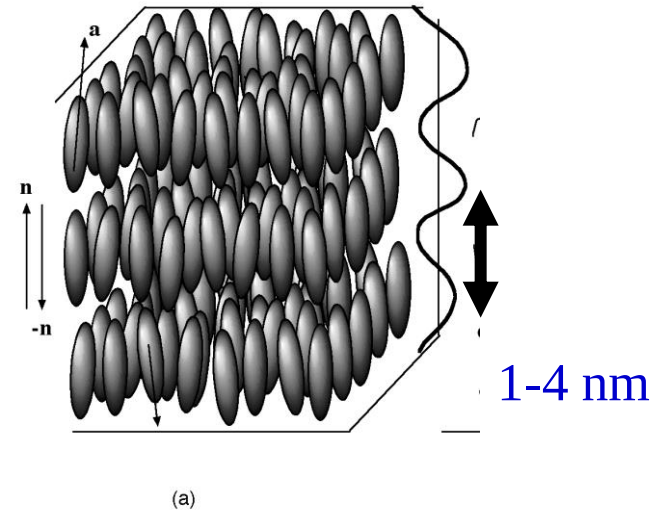
Add a 1D positional order to obtain a smectic...

Smectic A (SmA)

Periodic modulation of density (verified by X-ray experiments), unlike in Ntb phase



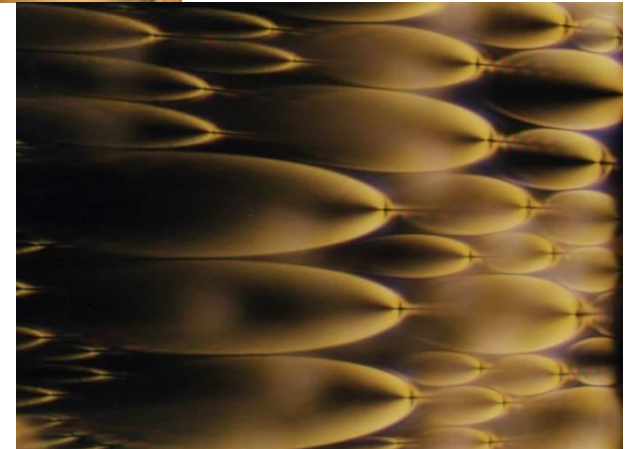
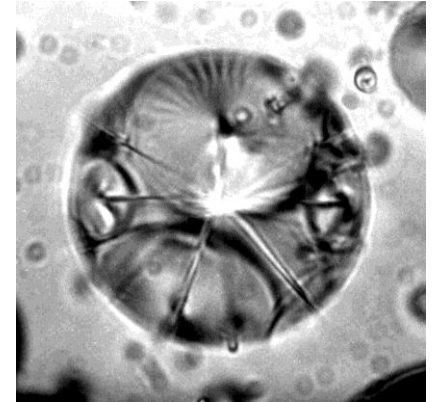
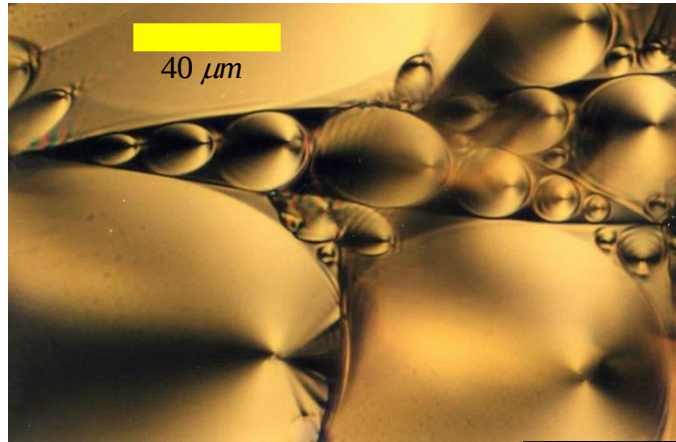
C₂₁H₂₅N
Mol. Wt.: 291



solid $\xleftrightarrow{24^{\circ}\text{C}}$ smectic A $\xleftrightarrow{34^{\circ}\text{C}}$ nematic $\xleftrightarrow{42.6^{\circ}\text{C}}$ isotropic

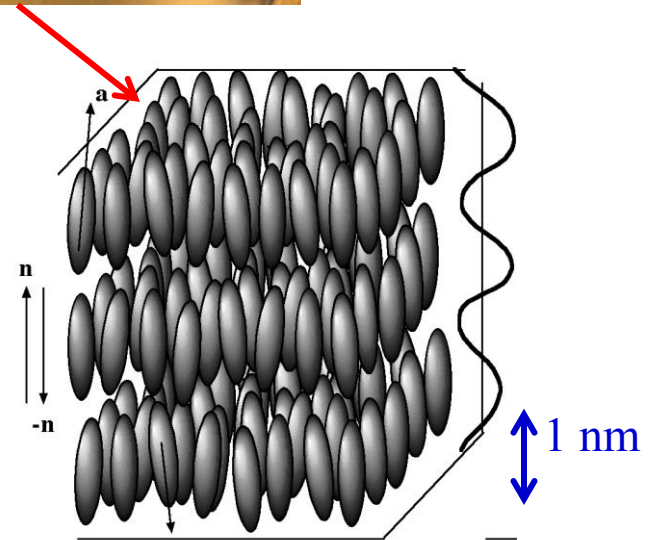
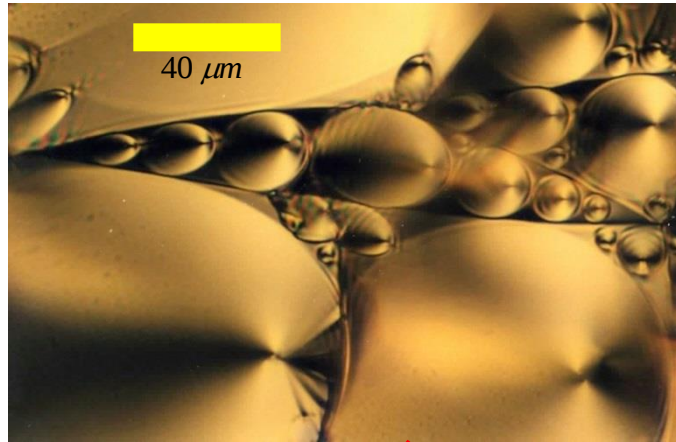
Smectics and focal conic domains

1910, G. Friedel, F. Grandjean:
Deciphered SmA structure
from observation of
focal conic domains;
X-ray was not available

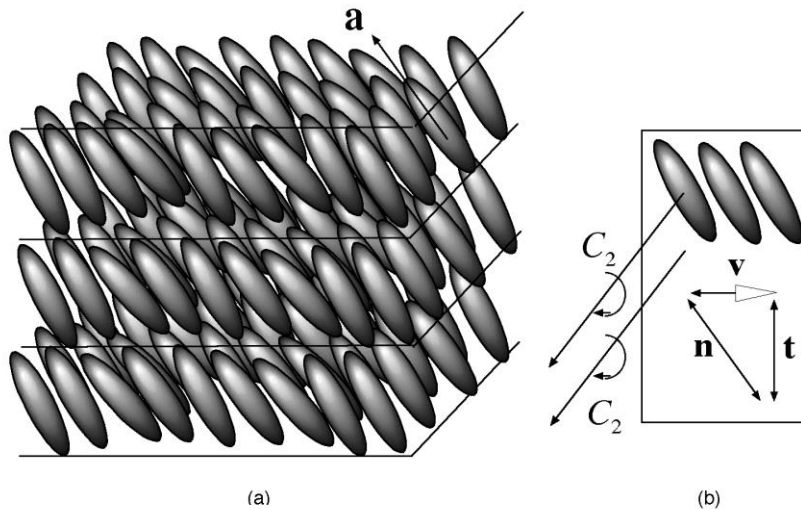


Smectics: Optical Microscopy at its best

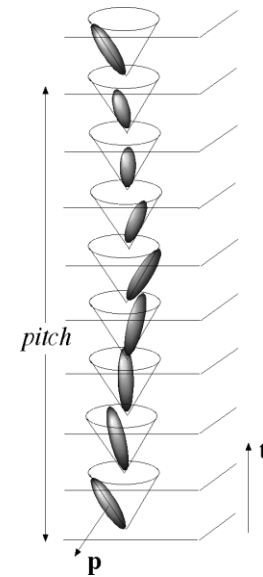
1910, G. Friedel, F. Grandjean:
Deciphered SmA structure
from observation of
focal conic domains;
X-ray was not available,
but the mere fact of existence of
ellipses and hyperbolae led to a
correct conclusion: SmA is a
system of equidistant flexible
fluid layers, i.e, a 1D periodic
structure



Smectics A, C, C*, etc



SmC



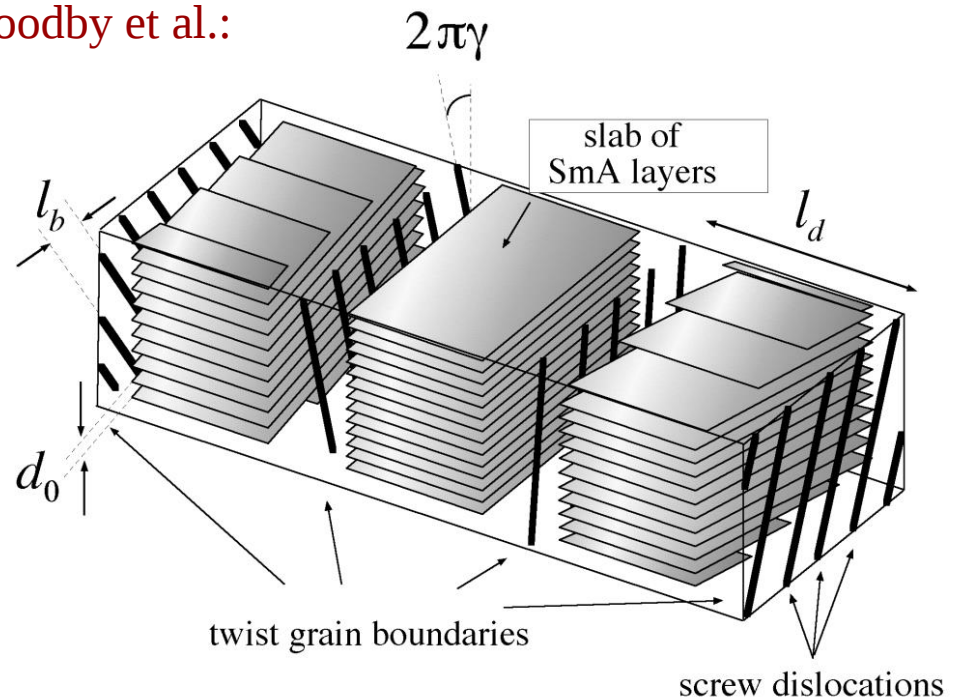
SmC*

Smectic A: Molecules normal to the layers; Smectic C; molecules are tilted
Chiral smectic C; molecules are tilted and follow an oblique helicoid

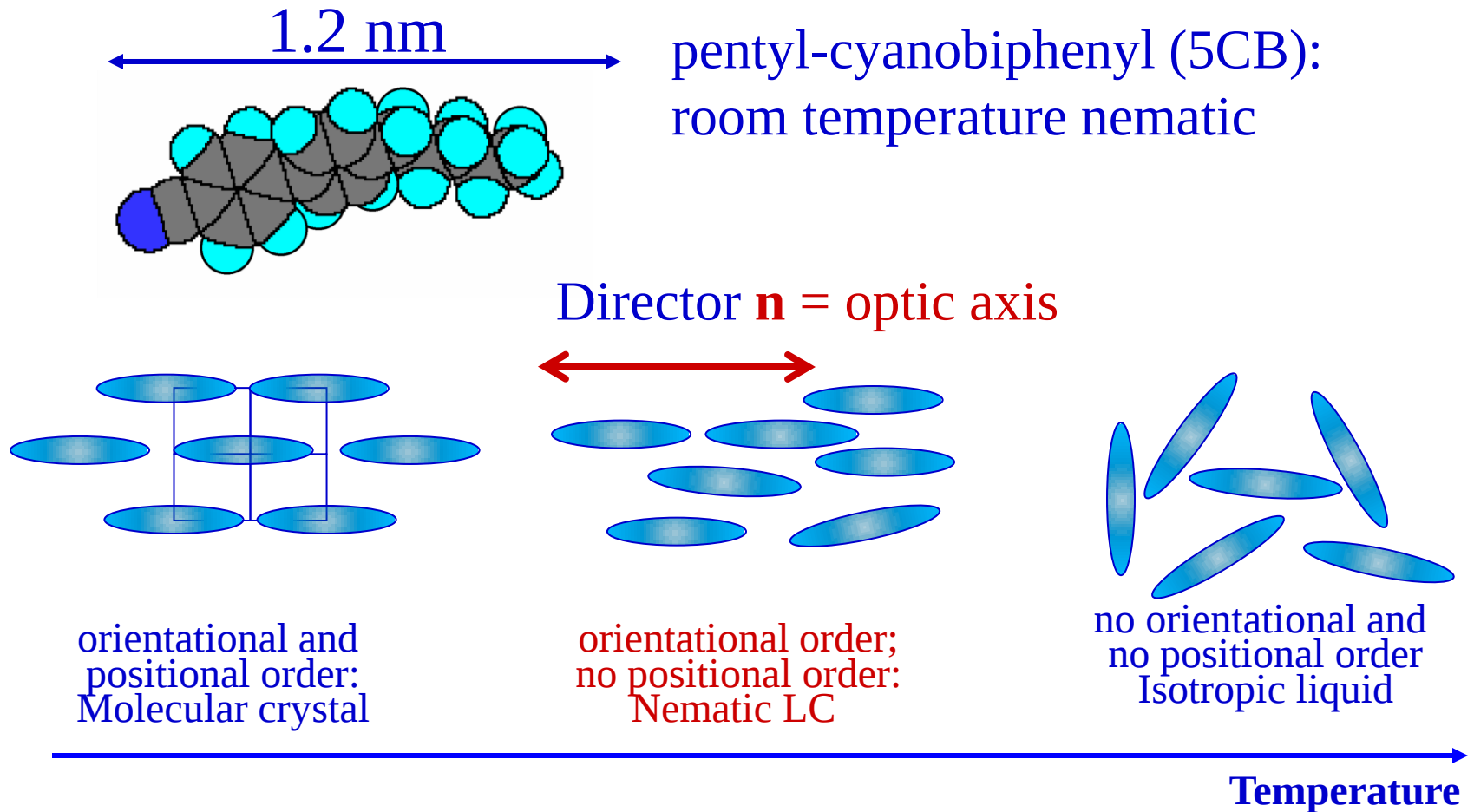
Twist Grain Boundary Phases

Combination of smectics and cholesterics; formed by chiral molecules; sophisticated analog of Abrikosov phase in superconductors; smectic is penetrated by a lattice of screw dislocations that allows the smectic to twist

1989, Renn, Lubensky, Pindak, Goodby et al.:

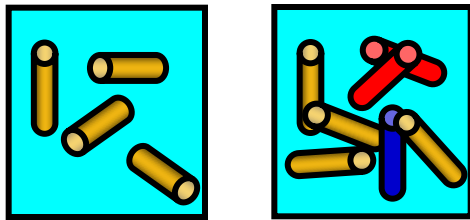


Crystals, thermotropic liquid crystals, liquids



Lyotropic Liquid Crystals: Power of Entropy

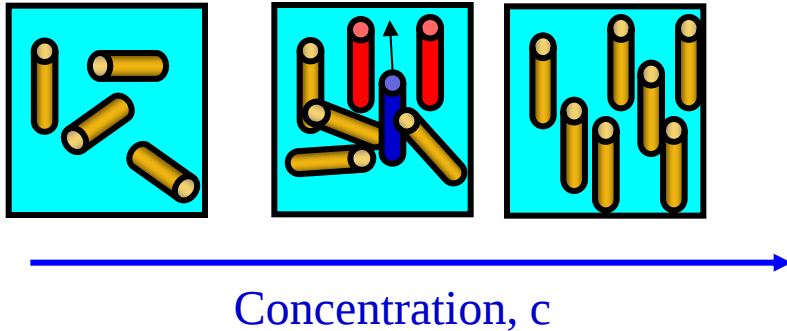
Onsager (1949): Nematic order in solution of long thin rods, thanks to translational vs orientational entropy trade-off



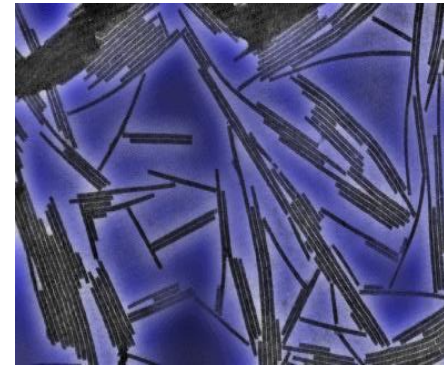
Concentration, c

Lyotropic Liquid Crystals: Power of Entropy

Onsager (1949): Nematic order in solution of long thin rods, thanks to translational vs orientational entropy trade-off



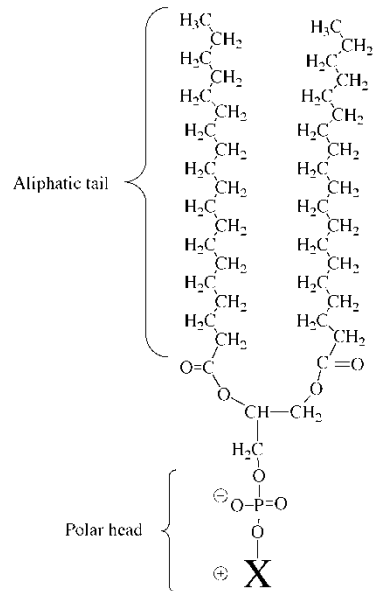
Phase diagram does not depend on temperature



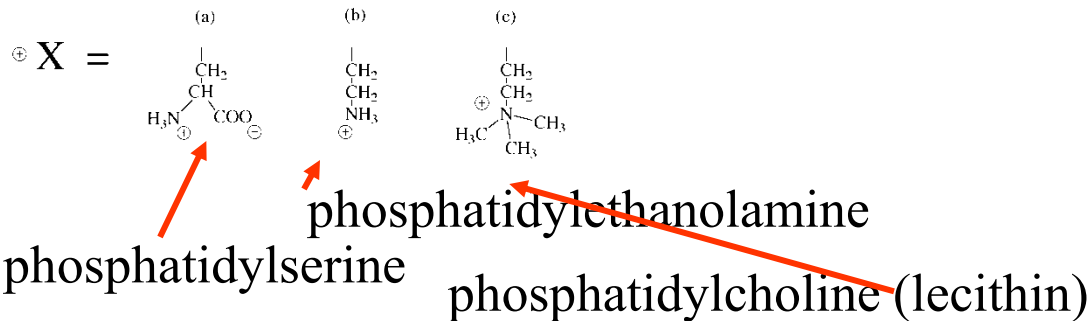
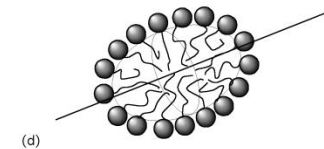
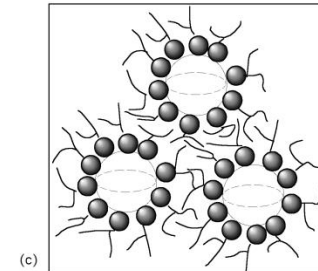
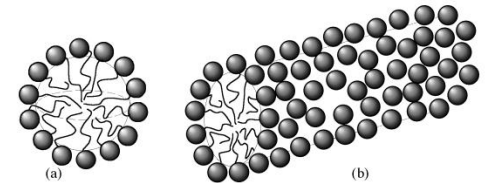
Tobacco mosaic virus (TMV)

J. Bernal and I. Fankuchen, J. Gen. Physiol. (1941): tactoids as N nuclei in tobacco mosaic virus dispersions (1939: First images of TMVs)

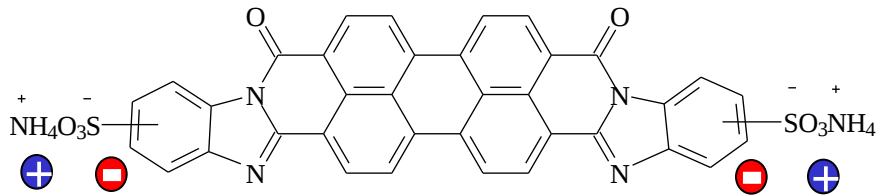
Molecular structure of phospholipids



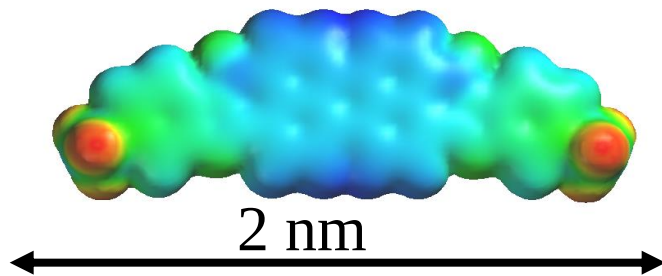
Molecular structure of phospholipids. The number of carbon atoms in the aliphatic chains varies, usually between 16 and 20. Two chains might be of different length.



Lyotropic Chromonic LCs: special case



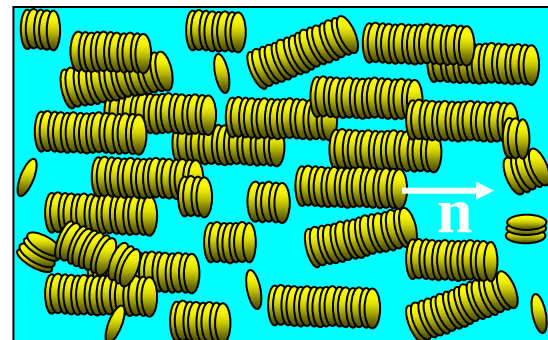
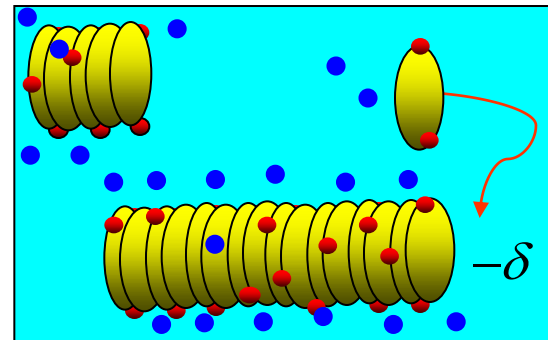
Violet 20



Chromonic molecules:

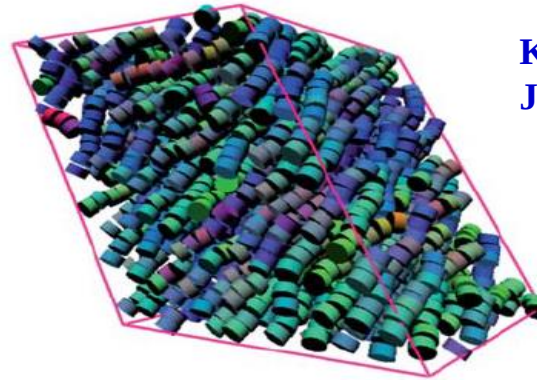
1. Rigid plank-like polyaromatic core
2. Ionic groups at periphery

Mechanism of LC formation: through aggregation: balance of entropy and association energy δ ; average length of aggregates $\bar{L} \propto \sqrt{c} \exp \frac{\delta}{2k_B T}$

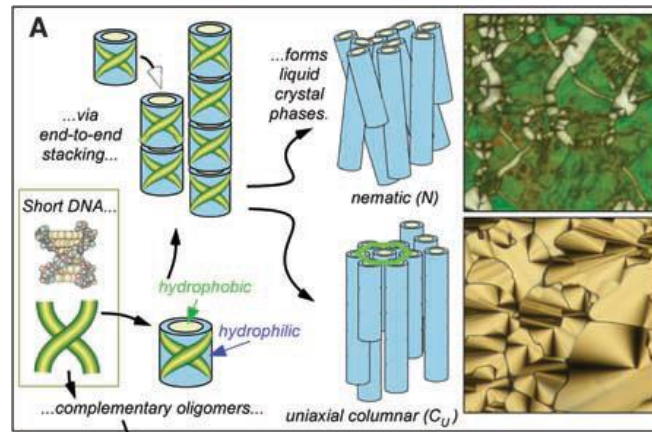


Lyotropic Chromonic LCs: special case

- ❑ LCLCs=Mesomorphic phases (N and Col) resulting from 1D non-covalent molecular self-assembly
- ❑ Common occurrence in dyes, drugs, proteins, nucleic acids
- ❑ Similar to living polymers, wormlike micelles of surfactants
- ❑ Promising applications as sensing material, in optical components, organic semiconductors, alignment of nanotubes, assembly of nanorods, etc.



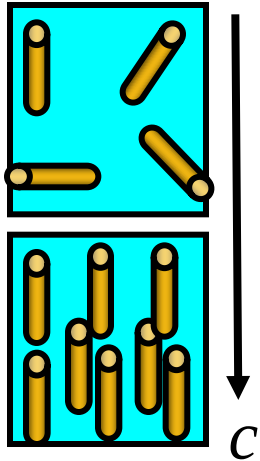
Kuriabova, Betterton, Glaser,
J. Mat. Chem. 20,10366 (2010)



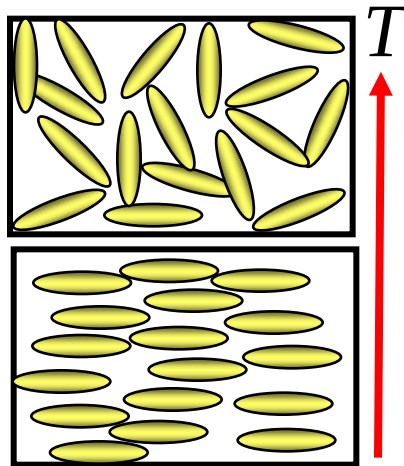
Nakata, Clark et al. Science 318, 1276
(2007)

Lyotropic Chromonic LCs: special case

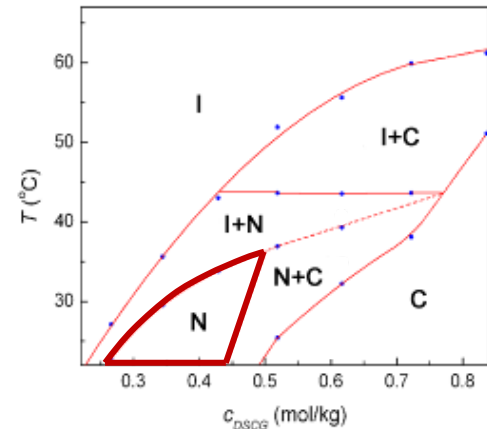
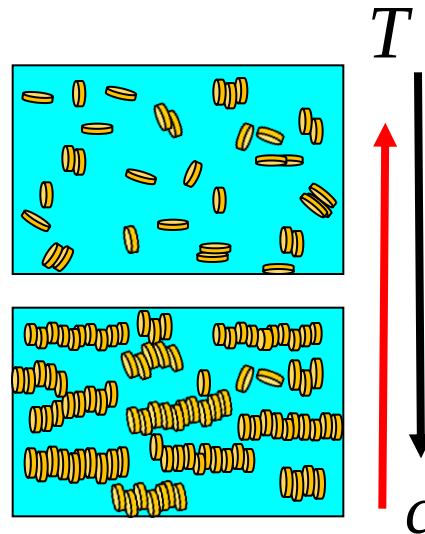
Onsager systems:
same rods,
athermal,
aligned at high c



Thermotropic LC (in your
displays): same molecules,
 $c = \text{const}$, aligned at low T



Chromonics :
controlled by both c
and T



Columnar phase: 2D positional order of columns (aggregates)



Nuclei of columnar phase in isotropic fluid: Bend deformations that preserve the equidistance of 2D lattices
Toroids rather than spheres

Disodium cromoglycate + water + PEG

Content

☐ Types of liquid crystalline order

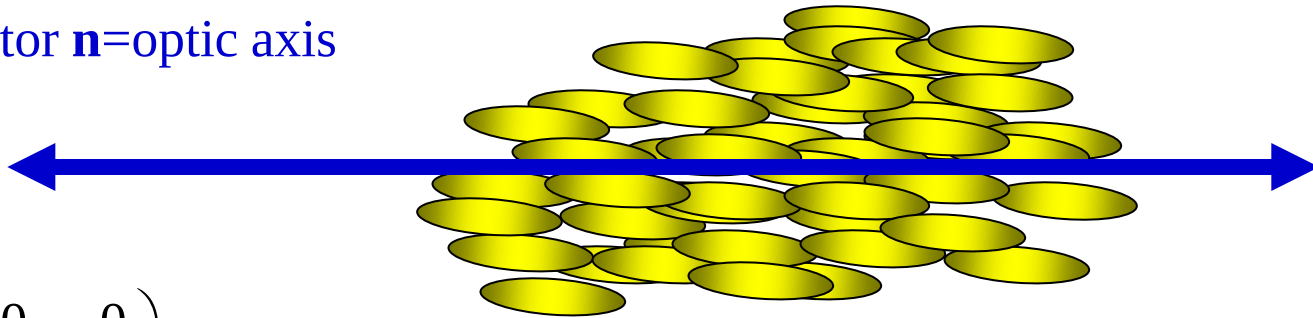
- Nematic
- Cholesteric and blue phases
- Twist bend nematic
- Smectic A
- Lyotropic and Chromonic LCs

☐ Basic Physics

- Dielectric anisotropy
- Surface anchoring
- Elasticity
- Frederiks effect and modern LCDs

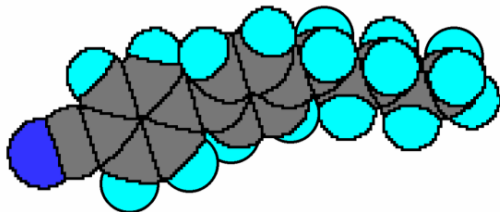
Anisotropy of uniaxial nematic

Director $\mathbf{n}$ =optic axis



$$\underline{\underline{\epsilon}} = \begin{pmatrix} \epsilon_{\perp} & 0 & 0 \\ 0 & \epsilon_{\perp} & 0 \\ 0 & 0 & \epsilon_{\parallel} \end{pmatrix}$$

Anisotropy of all properties: Optical, Dielectric, Diamagnetic, Electric conductivity, Elasticity, Viscosity, etc.



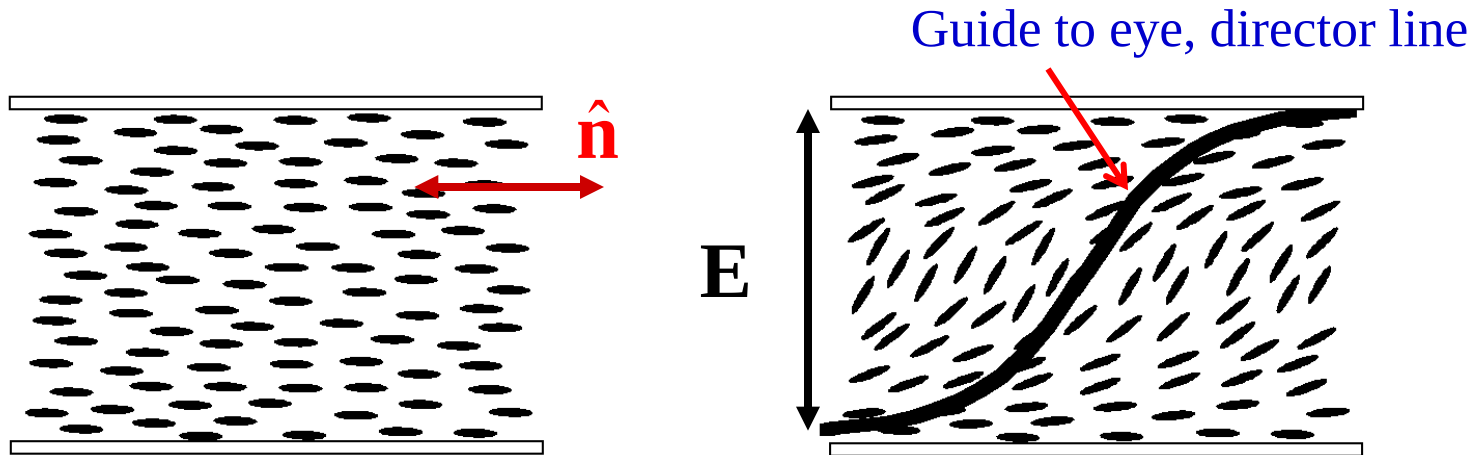
pentyl-cyanobiphenyl (5CB)

$$\epsilon_a = \epsilon_{\parallel} - \epsilon_{\perp} \approx 14 - 4 = 10$$

$$\Delta n = n_e - n_o \approx 1.7 - 1.5 \approx 0.2$$

Optic axis: Easily deformable by E-field: Frederiks effect

Field-induced reorientation of LC optic axis caused by diamagnetic or dielectric anisotropy: Frederiks effect (Leningrad, USSR, 1920-1930ies);



Optical response to the electric field puts liquid crystals at the heart of modern informational displays technologies

Frederiks effect: Field reorients LC

1927, Vsevolod Frederiks and Antonina Repiova

Magnetic field reorients LC

1934, V. Frederiks and V. Tsvetkov: Electric field reorients LC

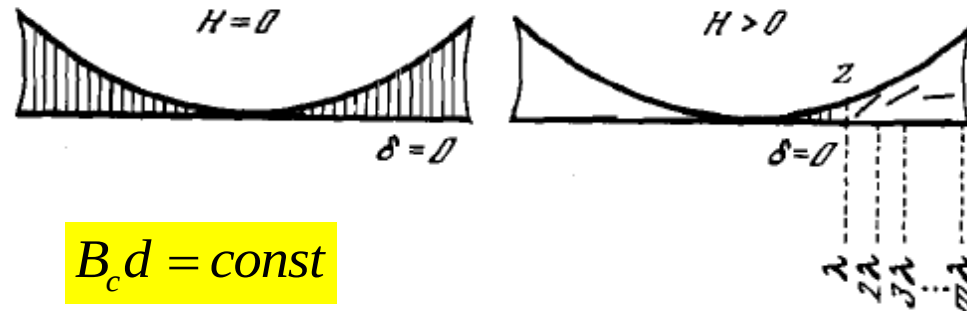
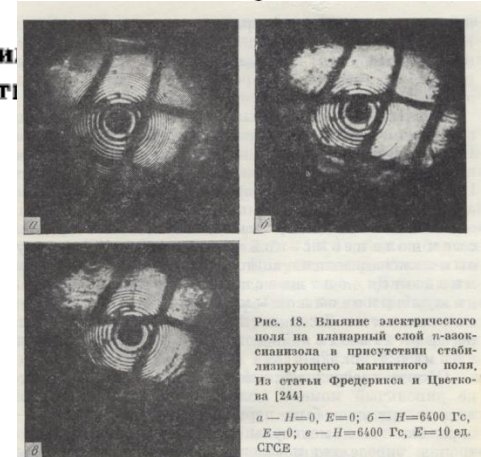


Рис. 14. Схема эксперимента Репёвой и Фредерикс. Влияние магнитного поля на ориентацию нематика. Из статьи Репёвой [218]

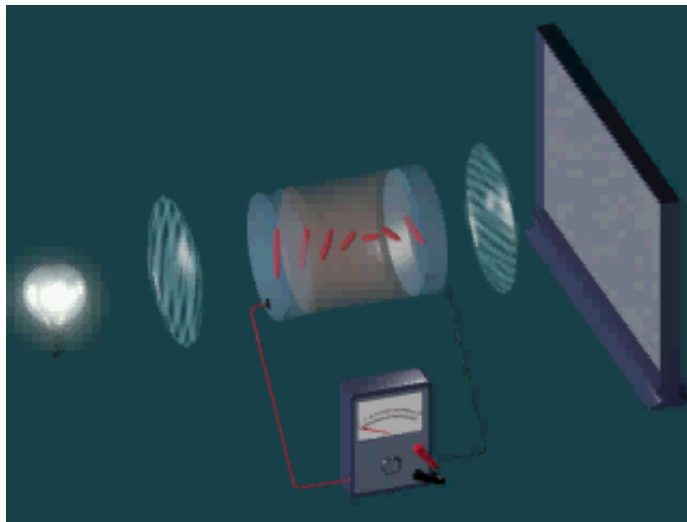


Field effects = LC displays

1969, Kent, Ohio: James Fergason
patents Twisted nematic cell, the first field-operated LCD
(M. Schadt and W. Helfrich filed a similar patent in Europe and
also published an article)



1934-2008

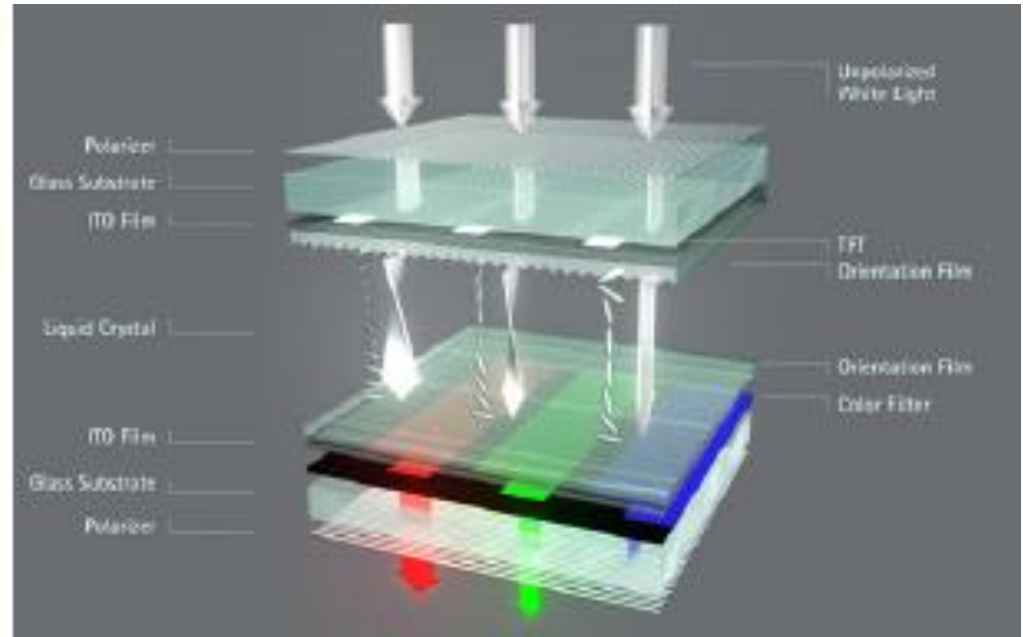
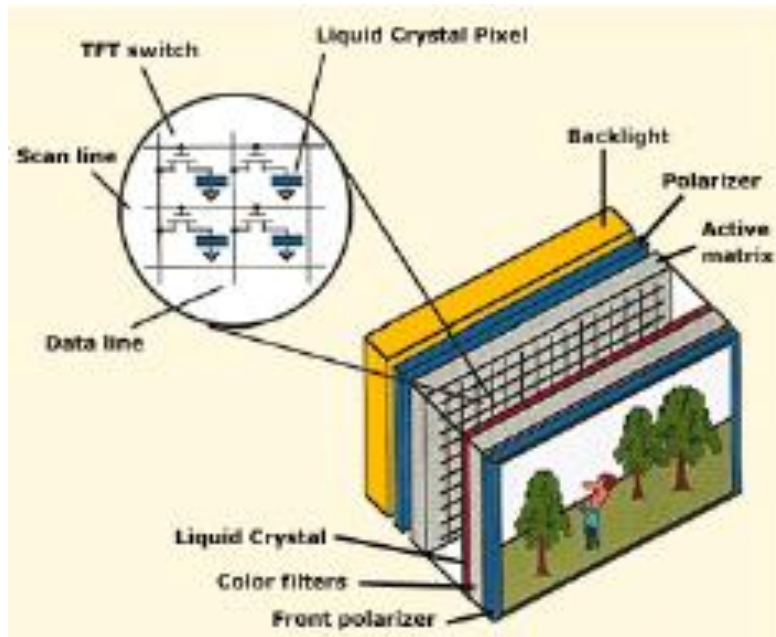


2006: Lemelson-MIT \$500,000 award



First product,
Fergason's ILIXCO (Kent, 1970)

LC displays: Control of polarized light



Dielectric anisotropy and birefringence of LC enabled revolution in informational portable displays



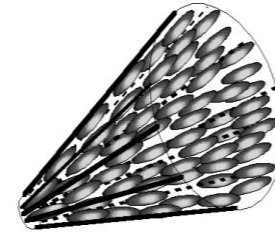
3.5 billion LCD panels sold in 2014 (all sizes)

Elasticity vs. Anchoring

□ Elasticity: Director gradients cost energy

$$f = \frac{1}{2} K \left(\frac{\partial n_i}{\partial x_j} \right)^2 \sim \frac{K}{L^2}$$

$$F = \int f dV \sim KL$$

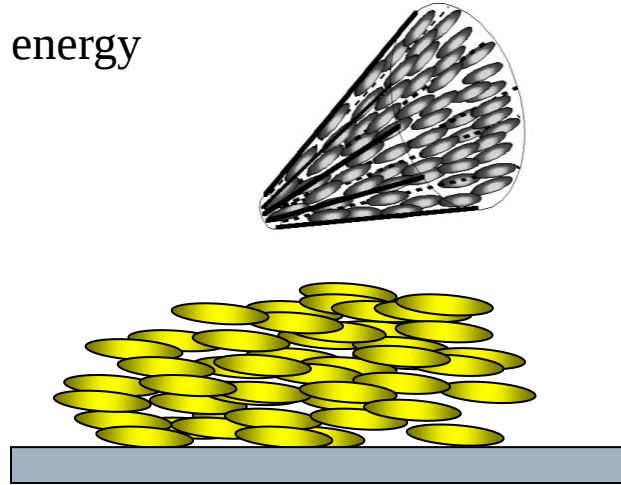


Elasticity vs. Anchoring

- Elasticity: Director gradients cost energy

$$f = \frac{1}{2} K \left(\frac{\partial n_i}{\partial x_j} \right)^2 \sim \frac{K}{L^2}$$

- $F = \int f dV \sim KL$
□ Surface anchoring



Elasticity vs. Anchoring

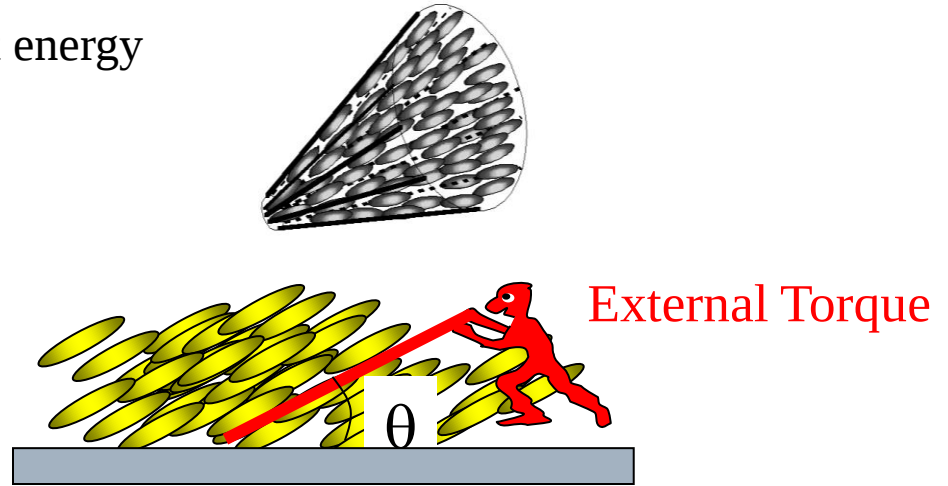
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$$F = \int f dV \sim KL$$

- Surface anchoring

$$f_s \approx \frac{1}{2} W \theta^2; F_s \sim WL^2$$



Elasticity vs. Anchoring

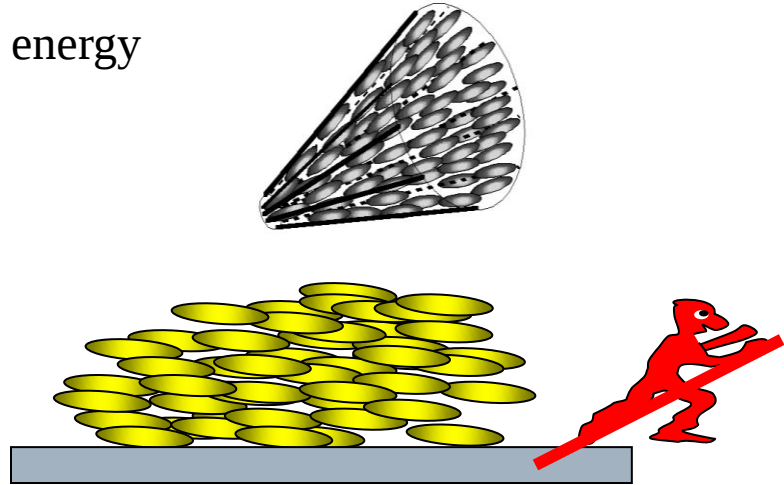
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□ Surface anchoring

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Elasticity vs. Anchoring

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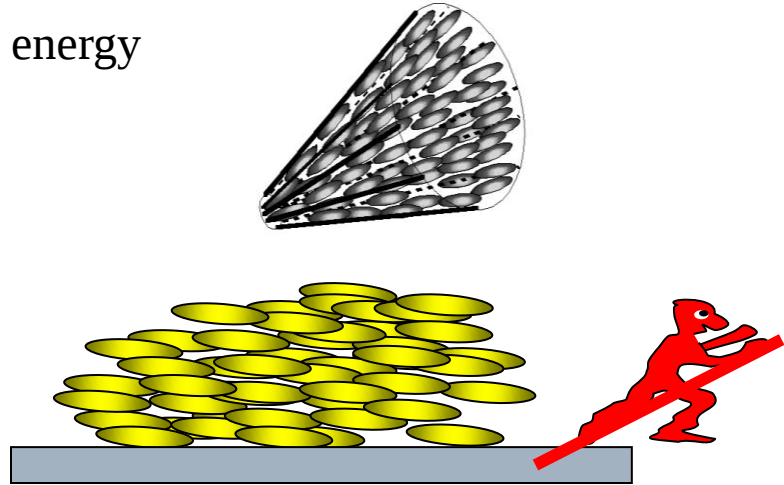
$$F = \int f dV \sim KL$$

- Surface anchoring

$$f_s \approx \frac{1}{2} W \theta^2; F_s \sim WL^2$$

- Anchoring extrapolation length

$$\xi = K / W$$



Material parameters: orders of magnitude

$K - ?$ $W - ?$ $\xi - ?$

Elastic constants $K \sim \frac{k_B T_{NI}}{a} \sim \frac{5 \times 10^{-21} \text{ J}}{10^{-9} \text{ m}} \sim 5 \text{ pN}$

Surface tension $\sigma \sim \frac{k_B T}{a^2} \sim \frac{5 \times 10^{-21}}{10^{-18}} \sim 10^{-2} \text{ J/m}^2$

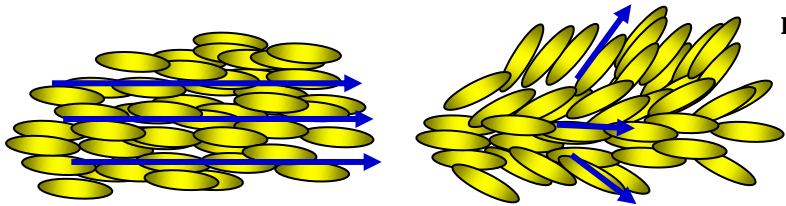
Anisotropy of surface tension: Intuition fails; experiments say

$$W \sim (10^{-3} \div 10^{-6}) \text{ J/m}^2$$

(weak as compared to surface tension)

 $\xi = \frac{K}{W} = \frac{10^{-11} \text{ N}}{10^{-3} \div 10^{-6} \text{ J/m}^2} \sim 10 \text{ nm} \div 10 \mu\text{m} \gg \text{molecular size } a$

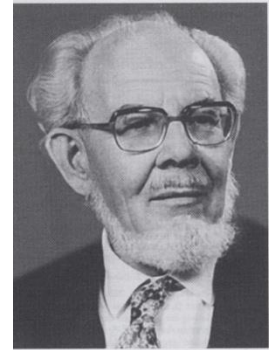
Elasticity of N: Oseen, 1933, Frank, 1958



$$\hat{\mathbf{n}} = \hat{\mathbf{n}}(x, y, z)$$

Elastic free energy density-?

$$f = f_0 + \alpha_{ij} \frac{\partial n_i}{\partial x_j} + \beta_{ijkl} \left(\frac{\partial n_i}{\partial x_j} \right)^2 + \dots$$



Requirements: $\left| \frac{\partial n_i}{\partial x_j} \right| \ll \frac{1}{\text{molecular size}}$

- $\mathbf{n}$ and $\mathbf{n}$ are invariant;
- central inversion about any point;
- invariance under any rotation around $\mathbf{n}$.

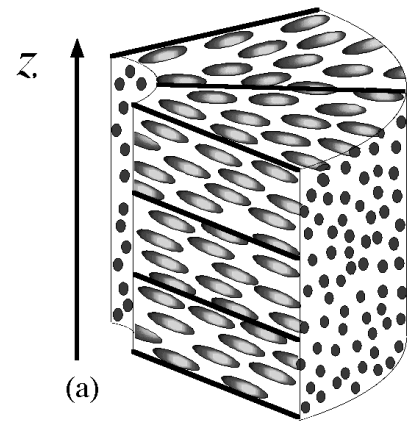
Two scalar invariants linear in derivatives: $\text{div } \hat{\mathbf{n}}$ $\hat{\mathbf{n}} \cdot \text{curl } \hat{\mathbf{n}}$

Invariant and Quadratic in derivatives: $(\text{div } \hat{\mathbf{n}})^2$ $(\hat{\mathbf{n}} \cdot \text{curl } \hat{\mathbf{n}})^2$ $(\text{curl } \hat{\mathbf{n}})^2$

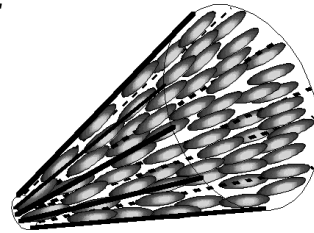
$$(\text{curl } \hat{\mathbf{n}})^2 = (\hat{\mathbf{n}} \times \text{curl } \hat{\mathbf{n}})^2 + (\hat{\mathbf{n}} \cdot \text{curl } \hat{\mathbf{n}})^2 \quad \dots \text{thus can also be } (\mathbf{n} \times \text{curl } \mathbf{n})^2$$

Frank-Oseen elastic free energy density:

$$f_{FO} = \underbrace{\frac{1}{2} K_1 (\text{div } \hat{\mathbf{n}})^2}_{\text{splay}} + \underbrace{\frac{1}{2} K_2 (\hat{\mathbf{n}} \cdot \text{curl } \hat{\mathbf{n}})^2}_{\text{twist}} + \underbrace{\frac{1}{2} K_3 (\hat{\mathbf{n}} \times \text{curl } \hat{\mathbf{n}})^2}_{\text{bend}}$$



$$\text{div} \mathbf{n} = \frac{1}{r}$$



$$\text{div} \mathbf{n} = \frac{2}{r}$$

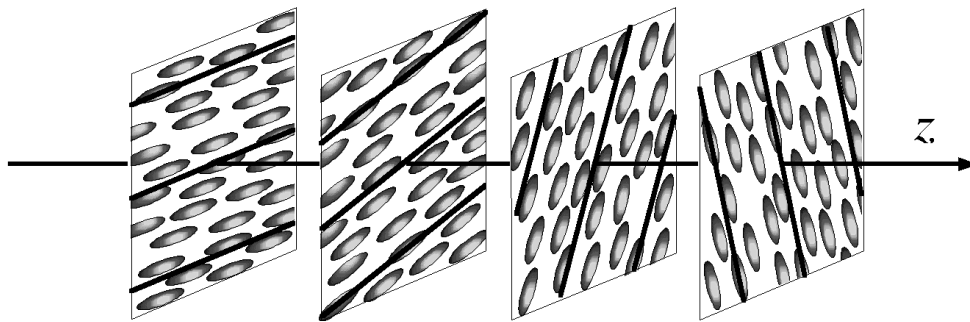
$$\left(n_x = \frac{x}{r}, \quad n_y = \frac{y}{r}, \quad n_z = \frac{z}{r} \right)$$

2D and 3D splay

(a)

(b)

$$(n_x = \cos \varphi, \quad n_y = \sin \varphi, \quad n_z = 0)$$

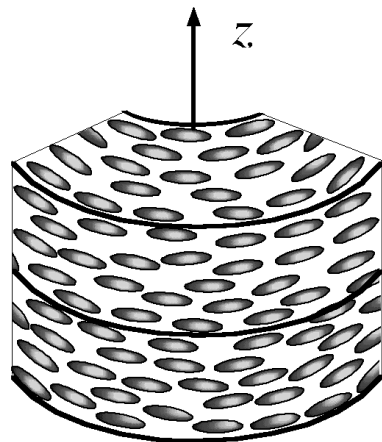


(c)

Twist (one-directional)

$$(n_x = \cos qz, \quad n_y = \sin qz, \quad n_z = 0)$$

$$q = \frac{\alpha}{d} = \frac{2\pi}{p} \quad q = -\mathbf{n} \cdot \text{curl} \mathbf{n}$$



(d)

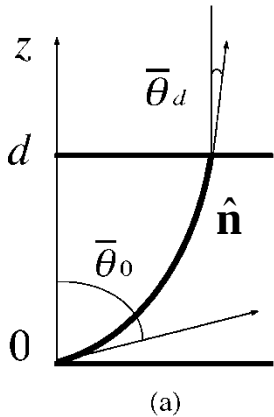
Bend

$$\mathbf{n} \times \text{curl} \mathbf{n}$$

$$(n_x = \sin \varphi, \quad n_y = \cos \varphi, \quad n_z = 0)$$

Elasticity vs Anchoring: Hybrid-Aligned Nematic Film

1. Infinitely strong anchoring



Fixed boundary conditions $\theta(z=0) = \bar{\theta}_0$ $\theta(z=d) = \bar{\theta}_d$

Problem: Find the director dependence on z in equilibrium

Assumption #1: $\hat{\mathbf{n}} = \{n_x, n_y, n_z\} = \{\sin \theta(z), 0, \cos \theta(z)\}$

$$f_{FO} = \frac{1}{2} K_1 \left(\sin \theta \frac{d\theta}{dz} \right)^2 + \frac{1}{2} K_3 \left(\cos \theta \frac{d\theta}{dz} \right)^2$$

Assumption #2: $K_1 = K_3 = K \quad \longrightarrow \quad f_{FO} = \frac{1}{2} K \left(\frac{d\theta}{dz} \right)^2$

The problem reduces to finding $\theta(z)$ that minimizes the integral $F_{FO} = \frac{1}{2} K \int_{z=0}^{z=d} \left(\frac{d\theta}{dz} \right)^2 dz$

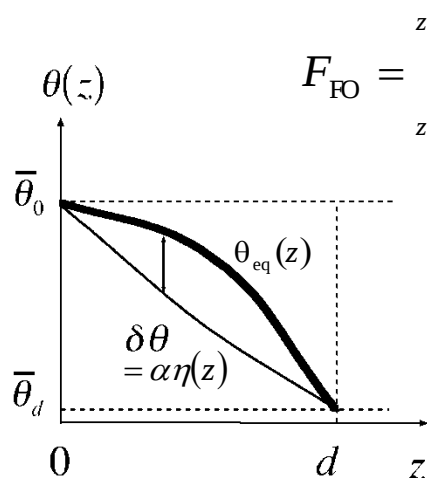
Euler-Lagrange eq. (next slide gives the outline, home assignment if you do not know it yet):

$$\frac{\partial f_{FO}}{\partial \theta} - \frac{d}{dz} \frac{\partial f_{FO}}{\partial \theta'} = 0 \Rightarrow \frac{d^2 \theta}{dz^2} = 0$$

$$\theta(z) = \bar{\theta}_0 - \frac{(\bar{\theta}_0 - \bar{\theta}_d)z}{d}$$

$$F_{FO} = \frac{1}{2} K \frac{(\bar{\theta}_0 - \bar{\theta}_d)^2}{d}$$

Euler-Lagrange equation for 1D problem, fixed boundary conditions (leisure time reading)



$$F_{\text{FO}} = \int_{z=0}^{z=d} f_{\text{FO}}[\theta, \theta', z] dz$$

$$F_{\text{FO}} = \frac{1}{2} K \int_{z=0}^{z=d} (\theta')^2 dz$$

?

$$\theta(z) = \theta_{\text{eq}}(z) + \alpha \eta(z)$$

where $\eta(z)$ is such that

$$\eta(z=0) = \eta(z=d) = 0$$

$$F[\theta(z)] = \int_0^d f[\theta_{\text{eq}}(z) + \alpha \eta(z), \theta'_{\text{eq}}(z) + \alpha \eta'(z), z] dz$$

Condition of the extremum: $\left[\frac{\partial F(\alpha)}{\partial \alpha} \right]_{\alpha=0} = 0$

$$\frac{\partial F(\alpha)}{\partial \alpha} = \int_0^d \left[\frac{\partial f}{\partial \theta} \frac{\partial \theta}{\partial \alpha} + \frac{\partial f}{\partial \theta'} \frac{\partial \theta'}{\partial \alpha} \right] dz = \int_0^d \left[\frac{\partial f}{\partial \theta} \eta(z) + \frac{\partial f}{\partial \theta'} \frac{d\eta(z)}{dz} \right] dz$$

$$\int_0^d \frac{d\eta(z)}{dz} \frac{\partial f}{\partial \theta'} dz = \underbrace{\eta(z) \frac{\partial f}{\partial \theta'} \Big|_{z=0}^{z=d}}_0 - \int_0^d \eta(z) \frac{d}{dz} \frac{\partial f}{\partial \theta'} dz$$

$$\int_0^d \left[\frac{\partial f}{\partial \theta} - \frac{d}{dz} \frac{\partial f}{\partial \theta'} \right] \eta(z) dz = 0 \quad \text{or}$$

$$\int_0^d \left[\frac{\partial f}{\partial \theta} - \frac{d}{dz} \frac{\partial f}{\partial \theta'} \right] \delta \theta dz = \alpha \left[\frac{\partial F(\alpha)}{\partial \alpha} \right]_{\alpha=0} = \delta F = 0$$



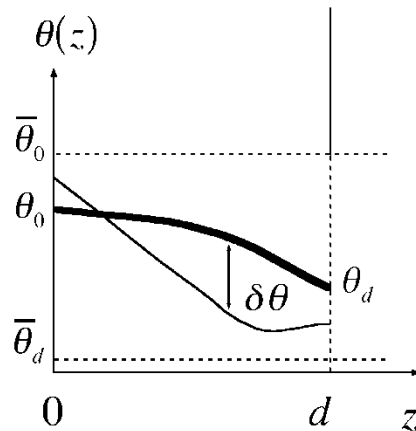
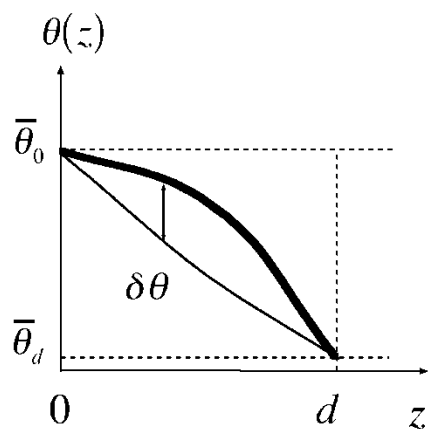
$$\frac{\partial f}{\partial \theta} - \frac{d}{dz} \frac{\partial f}{\partial \theta'} = 0$$

Euler-Lagrange equation for 1D problem; its solution

$\theta = \theta(z, c_1, c_2)$ has 2 constants of integration defined from the boundary conditions, for example,

$$\theta(z=0) = \bar{\theta}_0 \quad \text{and} \quad \theta(z=d) = \bar{\theta}_d$$

Euler-Lagrange equation for 1D problem, soft boundary conditions (leisure time reading)



$$F = \int_0^d f[\theta, \theta', z] dz + f_{s0}(\theta_0 - \bar{\theta}_0) + f_{sd}(\theta_d - \bar{\theta}_d)$$

$$\delta\theta = \theta(z) - \theta_{eq}(z) = \alpha\eta(z)$$

$\eta(z)$ is not necessarily 0 at the boundaries!

$$\frac{df_{s0}(\theta_0)}{d\alpha} = \frac{d}{d\alpha} f_{s0}[\theta_{0,eq} + \alpha\eta(z=0)] = \eta(0) \frac{df_{s0}}{d\theta_0}$$

(a)



$$\frac{\partial F(\alpha)}{\partial \alpha} = \int_0^d \left[\frac{\partial \mathcal{F}}{\partial \theta} \frac{\partial \theta}{\partial \alpha} + \frac{\partial \mathcal{F}}{\partial \theta'} \frac{\partial \theta'}{\partial \alpha} \right] dz + \frac{df_{s0}}{d\theta_0} \eta(0) + \frac{df_{sd}}{d\theta_d} \eta(d)$$

$$\int_0^d \frac{d\eta(z)}{dz} \frac{\partial \mathcal{F}}{\partial \theta'} dz = \eta(z) \frac{\partial \mathcal{F}}{\partial \theta'} \Big|_{z=0}^{z=d} - \int_0^d \eta(z) \frac{d}{dz} \frac{\partial \mathcal{F}}{\partial \theta'} dz$$



$$\int_0^d \left[\frac{\partial \mathcal{F}}{\partial \theta} - \frac{d}{dz} \frac{\partial \mathcal{F}}{\partial \theta'} \right] \eta(z) dz + \left[-\frac{\partial \mathcal{F}}{\partial \theta'} + \frac{df_{s0}}{d\theta} \right]_{z=0} \eta(0) + \left[\frac{\partial \mathcal{F}}{\partial \theta'} + \frac{df_{sd}}{d\theta} \right]_{z=d} \eta(d) = 0$$



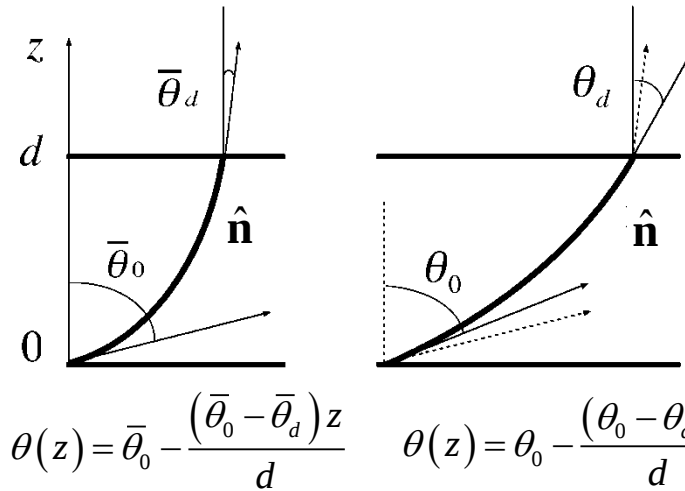
$$\frac{\partial \mathcal{F}}{\partial \theta} - \frac{d}{dz} \frac{\partial \mathcal{F}}{\partial \theta'} = 0$$

Euler-Lagrange equation for 1D problem; its solution $\theta = \theta(z, c_1, c_2)$ has 2 constants of integration defined from the boundary conditions

$$\left[-\frac{\partial \mathcal{F}}{\partial \theta'} + \frac{df_{s0}}{d\theta} \right]_{z=0} = 0 \quad \text{and} \quad \left[\frac{\partial \mathcal{F}}{\partial \theta'} + \frac{df_{sd}}{d\theta} \right]_{z=d} = 0$$

Elasticity vs Anchoring: Hybrid-Aligned Nematic Film

2. Finite (weak) anchoring



$$f_{FO} = \frac{1}{2} K \left(\frac{d\theta}{dz} \right)^2 \quad f_{s0} = \frac{1}{2} W_0 (\theta_0 - \bar{\theta}_0)^2 \quad f_{sd} = \frac{1}{2} W_d (\theta_d - \bar{\theta}_d)^2$$

Problem: Find the director vs z in equilibrium with new boundary conditions:

$$\left[-\frac{\partial f_{FO}}{\partial \theta'} + \frac{df_{s0}}{d\theta} \right]_{z=0} = 0 \quad \left[\frac{\partial f_{FO}}{\partial \theta'} + \frac{df_{sd}}{d\theta} \right]_{z=d} = 0$$



$$K(\theta_0 - \theta_d) + W_0 d (\theta_0 - \bar{\theta}_0) = 0$$



$$K(\theta_d - \theta_0) + W_d d (\theta_d - \bar{\theta}_d) = 0$$

$$\theta_0 = \bar{\theta}_0 - \frac{(\bar{\theta}_0 - \bar{\theta}_d) L_0}{d + L_0 + L_d}$$

$$\theta_d = \bar{\theta}_d + \frac{(\bar{\theta}_0 - \bar{\theta}_d) L_d}{d + L_0 + L_d}$$

$$L_0 = K / W_0$$

$$L_d = K / W_d$$

$$F_{FO} = \frac{1}{2} K \frac{(\bar{\theta}_0 - \bar{\theta}_d)^2}{d + L_0 + L_d}$$

Finite anchoring makes the director gradients weaker and the energy smaller, effectively increasing the cell thickness

Summary of hybrid aligned N film

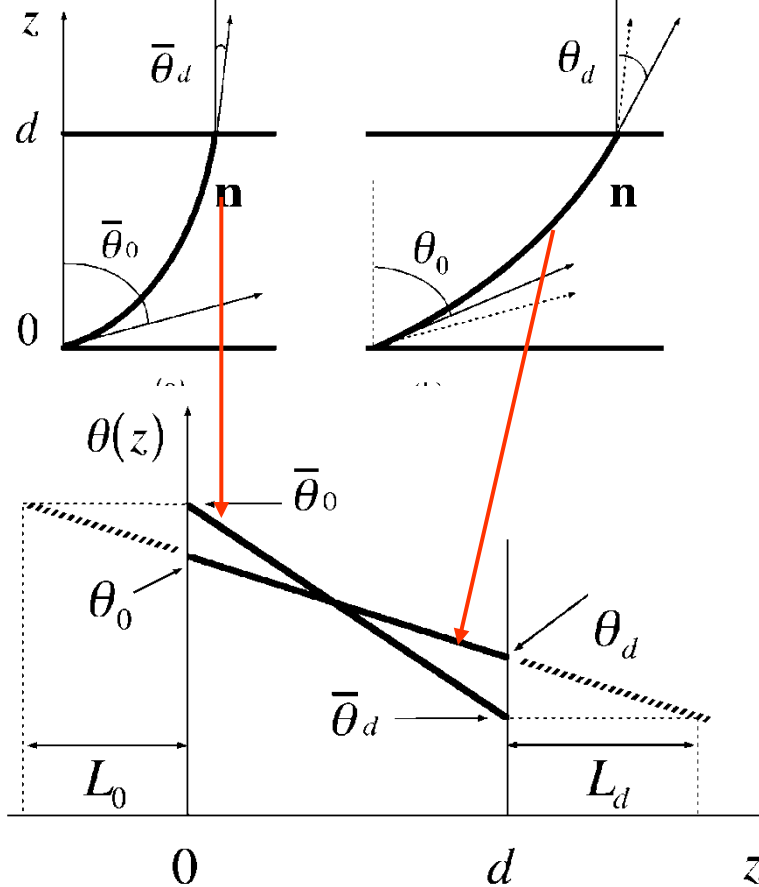
$$\theta(z) = \bar{\theta}_0 - \frac{(\bar{\theta}_0 - \bar{\theta}_d)z}{d}$$

$$\theta(z=0) = \bar{\theta}_0$$

$$\theta(z=d) = \bar{\theta}_d$$

$$F_{FO} = \frac{1}{2} K \frac{(\bar{\theta}_0 - \bar{\theta}_d)^2}{d}$$

Infinitely strong Finite anchoring



$$\theta(z) = \bar{\theta}_0 - \frac{(\bar{\theta}_0 - \bar{\theta}_d)z}{d}$$

$$\bar{\theta}_0 = \bar{\theta}_0 - \frac{(\bar{\theta}_0 - \bar{\theta}_d)L_0}{d + L_0 + L_d}$$

$$\bar{\theta}_d = \bar{\theta}_d + \frac{(\bar{\theta}_0 - \bar{\theta}_d)L_d}{d + L_0 + L_d}$$

$$F_{FO} = \frac{1}{2} K \frac{(\bar{\theta}_0 - \bar{\theta}_d)^2}{d + L_0 + L_d}$$

$$d \rightarrow d + L_0 + L_d$$

$$L_d = K / W_d$$

L: anchoring extrapolation length

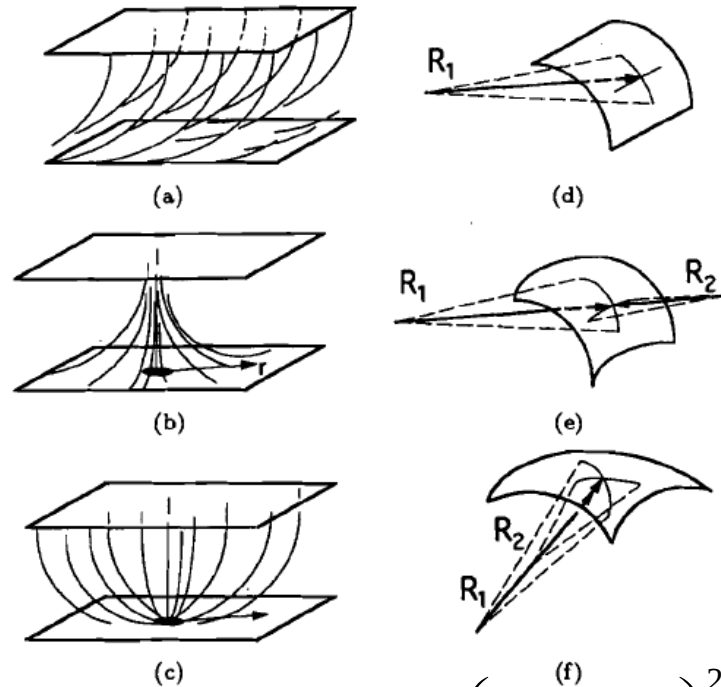
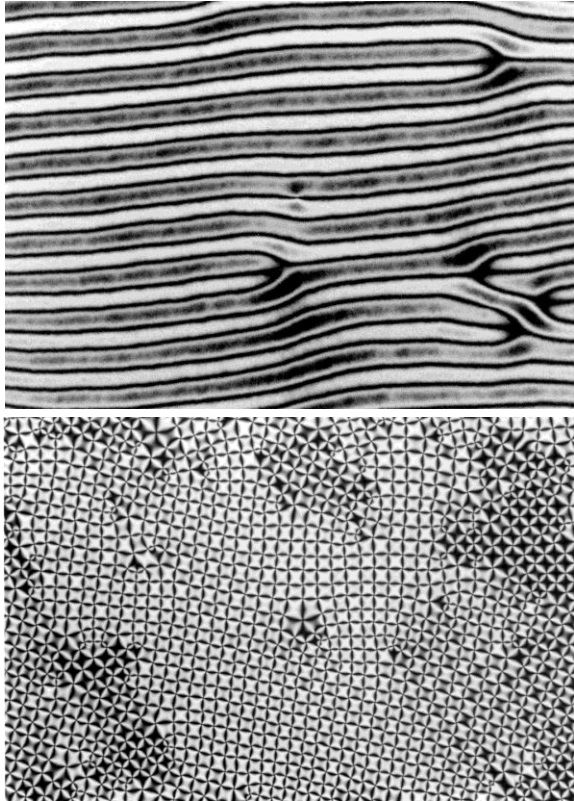
$$F_{FO} \sim Kl$$

$$F_{anchoring} \sim Wl^2$$

NB: at large scales, surface anchoring takes over, enslaving the director field, the director follows the “easy axis” prescribed by surface anchoring potential

End of story for hybrid aligned films?

No. At submicron thicknesses, the director is unstable w.r.t. in-plane deformations (similar to buckling)



$$\frac{1}{2} K_1 (\text{div} \hat{\mathbf{n}})^2 \sim \frac{1}{2} K_1 \left(\frac{1}{R_1} + \frac{1}{R_2} \right)^2$$

$$K_{24} \text{div} (\hat{\mathbf{n}} \cdot \text{div} \hat{\mathbf{n}} + \hat{\mathbf{n}} \times \text{curl} \hat{\mathbf{n}}) = \frac{2K_{24}}{R_1 R_2}$$

$$\frac{1}{R_1 R_2} = \frac{1}{2} \text{div} (\hat{\mathbf{n}} \cdot \text{div} \hat{\mathbf{n}} + \hat{\mathbf{n}} \times \text{curl} \hat{\mathbf{n}})$$

Free Energy of a Nematic in an External Field

Dielectric case (no ions, no flexoelectric/surface polarization)

Energy density should depend on two vectors, the field and the director

$$\mathbf{E} \quad \hat{\mathbf{n}} = -\hat{\mathbf{n}}$$

$$\underline{\underline{\varepsilon}} = \begin{pmatrix} \varepsilon_{\perp} & 0 & 0 \\ 0 & \varepsilon_{\perp} & 0 \\ 0 & 0 & \varepsilon_{\parallel} \end{pmatrix}$$

Electric displacement: $D_i = \varepsilon_0 \varepsilon_{ij} E_j$

Energy density (x 2): $\mathbf{E} \cdot \mathbf{D} = \varepsilon_0 \varepsilon_{\perp} E^2 + \varepsilon_0 \varepsilon_a (\mathbf{n} \cdot \mathbf{E})^2 \quad \varepsilon_a = \varepsilon_{\parallel} - \varepsilon_{\perp}$

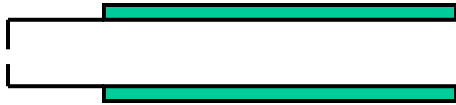
 $f = f_{FO} - \frac{1}{2} \varepsilon_0 \varepsilon_a (\hat{\mathbf{n}} \cdot \mathbf{E})^2$

$$f = f_{FO} - \frac{1}{2} \mu_0^{-1} \chi_a (\hat{\mathbf{n}} \cdot \mathbf{B})^2 \quad \mu_0 = 4\pi \times 10^{-7} \text{ Henry / m}$$

Free Energy of a Nematic in an External Field

Dielectric case (no ions, no flexoelectric/surface polarization)

U=const



$$F_{FO} = \int f_{FO} dV$$

$$F_E = \int f_E dV = \frac{1}{2} \int \mathbf{E} \cdot \mathbf{D} dV$$

The energy of the electric field and its change:

$$\delta F_E = \frac{1}{2} \int \mathbf{E} \cdot \delta \mathbf{D} dV$$

Which leads to a change in the surface charge density by $(-\delta D_z)$

When $\mathbf{n}$ reorients, surface charge density changes; to keep the voltage constant at the electrodes, one needs to supply energy from the electric source:

$$\delta F_G = \iint_A \psi \delta D_z dA = \int \text{div}(\psi \delta \mathbf{D}) dV$$

Electric potential

$$\text{div}(\psi \delta \mathbf{D}) = \psi \text{div} \delta \mathbf{D} + \delta \mathbf{D} \cdot \nabla \psi$$

$$\mathbf{E} = -\nabla \psi$$

$$\delta F_G = -\int \mathbf{E} \cdot \delta \mathbf{D} dV = -2\delta F_E$$

The minimum of the total bulk free energy is achieved when $\delta F_{FO} + \delta F_E + \delta F_G = \delta(F_{FO} - F_E) = 0$

$$\mathbf{D} = \epsilon_0 \epsilon_{\perp} \mathbf{E}_{\perp} + \epsilon_0 \epsilon_{\parallel} \mathbf{E}_{\parallel} = \epsilon_0 \epsilon_{\perp} \mathbf{E} + \epsilon_0 \epsilon_a (\mathbf{E} \cdot \mathbf{n}) \mathbf{n}$$

$$\mathbf{E} \cdot \mathbf{D} = \epsilon_0 \epsilon_{\perp} E^2 + \epsilon_0 \epsilon_a (\mathbf{n} \cdot \mathbf{E})^2$$

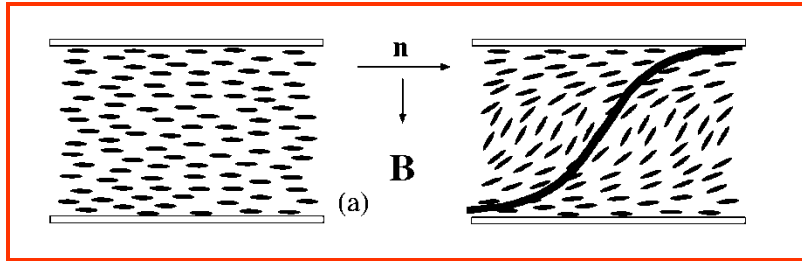


$$f = f_{FO} - \frac{1}{2} \epsilon_0 \epsilon_a (\hat{\mathbf{n}} \cdot \mathbf{E})^2$$

$$f = f_{FO} - \frac{1}{2} \mu_0^{-1} \chi_a (\hat{\mathbf{n}} \cdot \mathbf{B})^2$$

$$\mu_0 = 4\pi \times 10^{-7} \text{ Henry / m}$$

Splay Frederiks Transitions



$$\theta=0 \text{ at } z=0, z=d$$

$$f = \frac{1}{2} K_1 (\text{div} \hat{\mathbf{n}})^2 - \frac{1}{2} \mu_0^{-1} \chi_a (\mathbf{B} \cdot \hat{\mathbf{n}})^2$$

$$\angle \theta \quad \{n_x, n_y, n_z\} = \{\cos \theta(z), 0, \sin \theta(z)\}$$

$$f = \frac{1}{2} K_1 \cos^2 \theta \left(\frac{d\theta}{dz} \right)^2 - \frac{1}{2} \mu_0^{-1} \chi_a B^2 \sin^2 \theta$$

Assuming deviations are small:

$$f = \frac{1}{2} K_1 \left(\frac{d\theta}{dz} \right)^2 - \frac{1}{2} \mu_0^{-1} \chi_a B^2 \theta^2$$

E-L equation:

$$\xi^2 \frac{d^2 \theta}{dz^2} + \theta = 0 \quad \xi = \frac{1}{B} \sqrt{\frac{K_1}{\mu_0^{-1} \chi_a}}$$

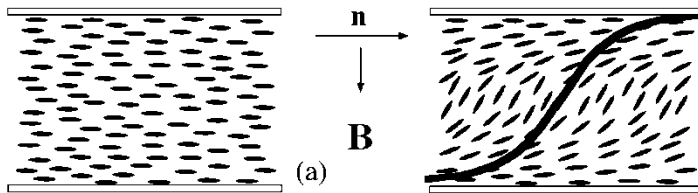
General solution:

$$\theta = a_1 \cos \frac{z}{\xi} + a_2 \sin \frac{z}{\xi}$$

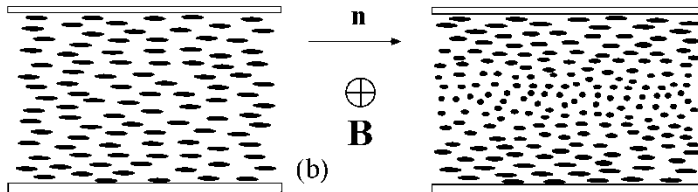
Boundary conditions yield $a_1 = 0$ and $d / \xi = n\pi$

Non-trivial solution at $B > B_c = \frac{\pi}{d} \sqrt{\frac{K_1}{\mu_0^{-1} \chi_a}}$, the critical field for the Frederiks transition

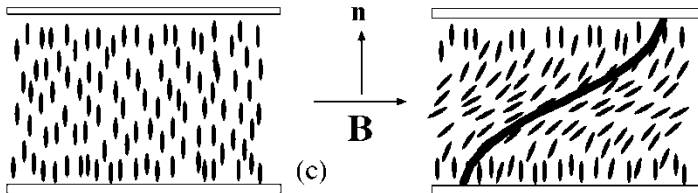
Three basic geometries of Frederiks effect



$$B_c = \frac{\pi}{d} \sqrt{\frac{K_1}{\mu_0^{-1} \chi_a}} \quad \text{Splay deformation}$$



$$B_c = \frac{\pi}{d} \sqrt{\frac{K_2}{\mu_0^{-1} \chi_a}} \quad \text{Twist deformation}$$



$$B_c = \frac{\pi}{d} \sqrt{\frac{K_3}{\mu_0^{-1} \chi_a}} \quad \text{Bend deformation}$$

Electric field case can be treated similarly

Heliconical director in electric field

Volume 12, Number 9

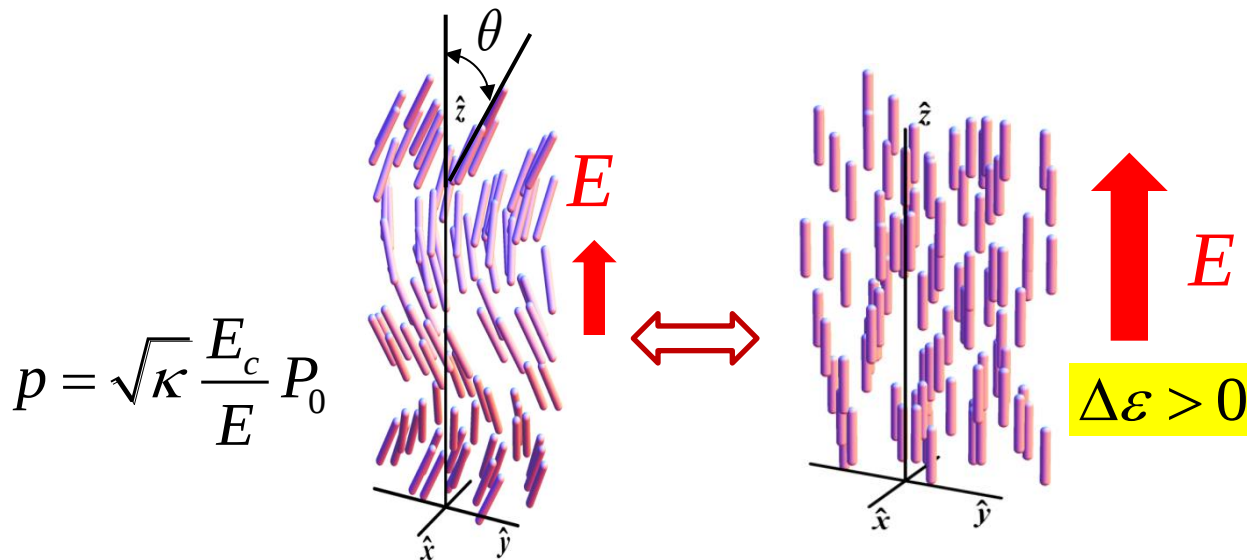
APPLIED PHYSICS LETTERS

1 May 1968

EFFECTS OF ELECTRIC AND MAGNETIC FIELDS ON THE STRUCTURE OF CHOLESTERIC LIQUID CRYSTALS*

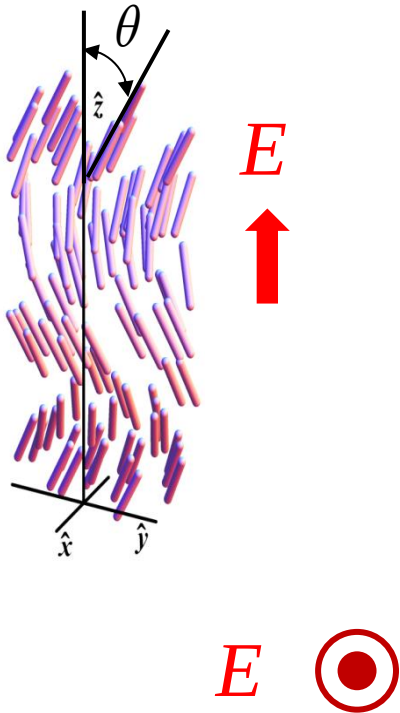
Robert B. Meyer

A cholesteric with a small bend-twist ratio $K_3/K_2 = \kappa \ll 1$ and $\Delta\epsilon > 0$ adopts an oblique helicoidal shape in an electric field with the field-dependent pitch:



Bimesogens: Ideal for electrically induced twist bend, since the bend constant is very small

Color tuning: Vertical field



J. Xiang et al, Advanced Materials, 3014 (2015)

Liquid Crystals. Lecture 1.2

Optics

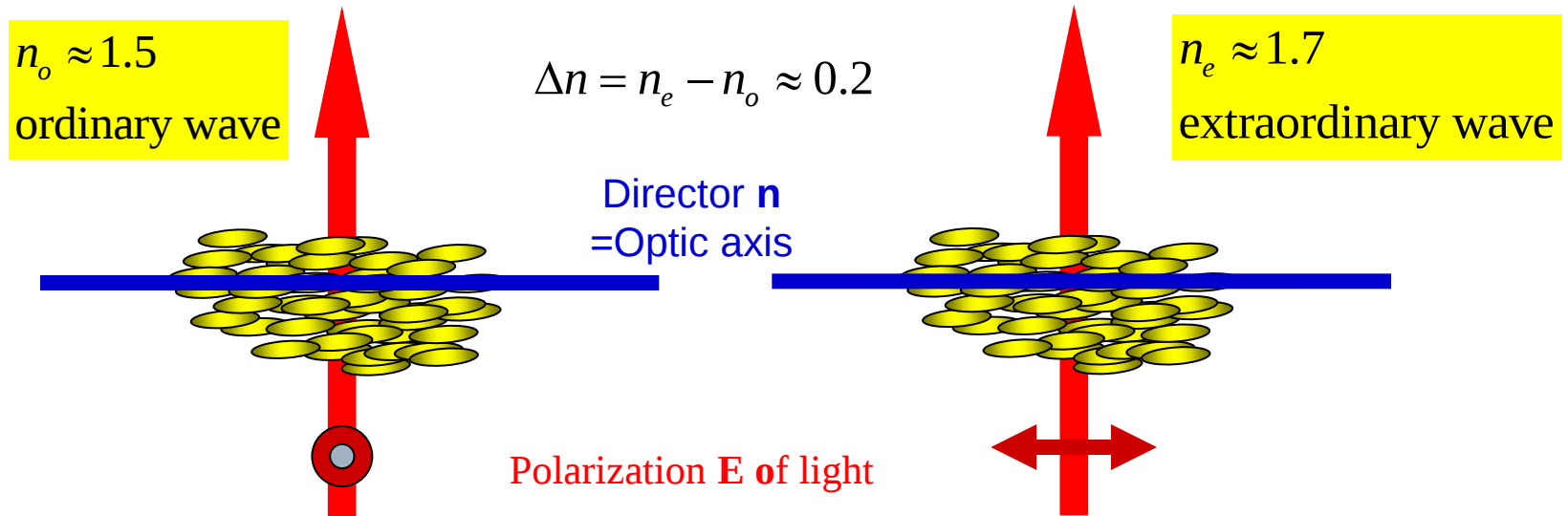
Oleg D. Lavrentovich

Support: NSF



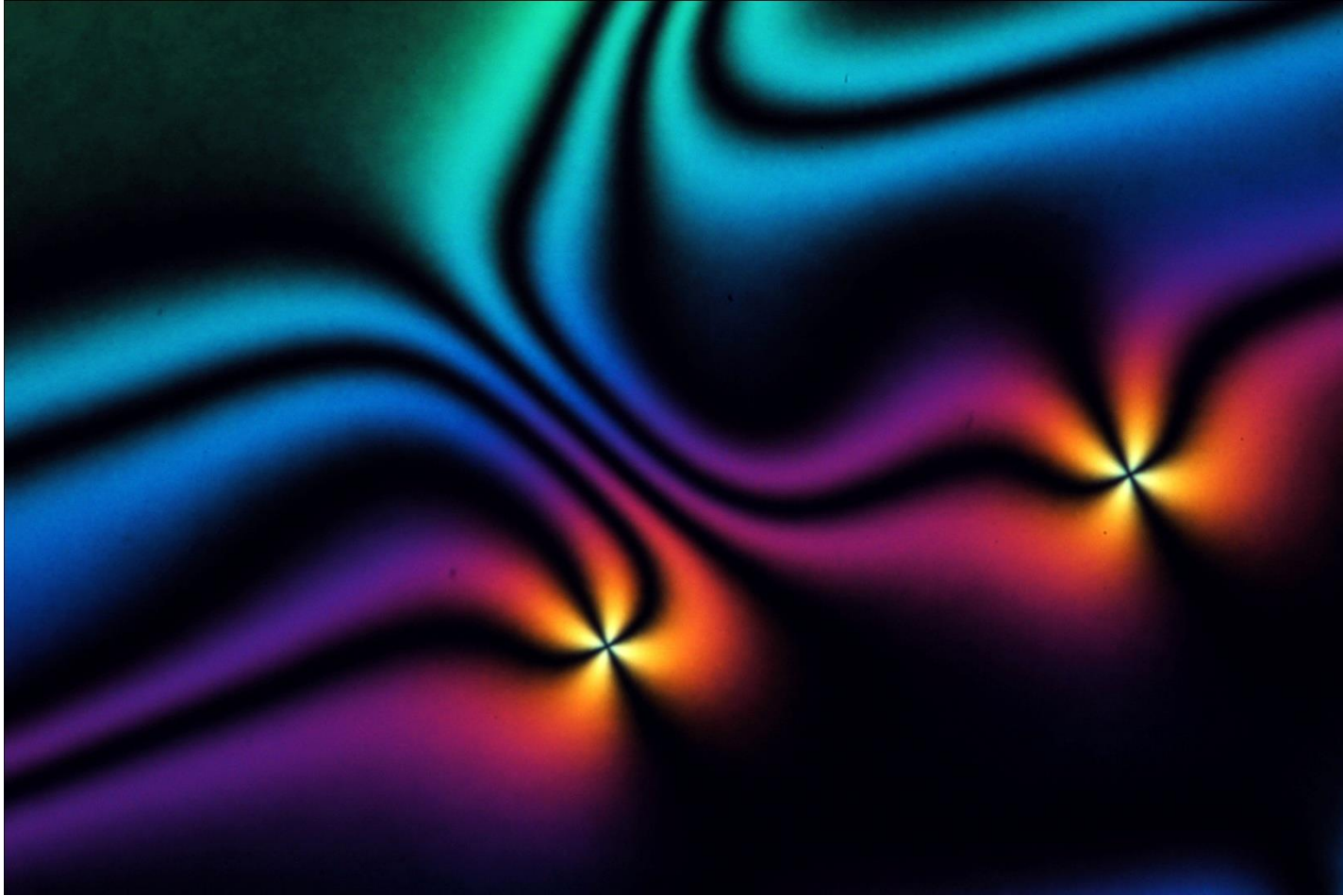
Boulder School for Condensed Matter and Materials Physics,
Soft Matter In and Out of Equilibrium,
6-31 July, 2015

LCs: Birefringent materials



Birefringence: Double refraction of light in an ordered material, manifested through dependence of refractive indices on polarization of light

Birefringence revealed through pair of polarizers: textures and LCDs



LCs: Ordinary and Extraordinary waves

El. field	Mag. field strength	Mag. field induction	El. displacement
$\nabla \times \mathbf{E} = -\partial \mathbf{B} / \partial t$	$\nabla \times \mathbf{H} = \partial \mathbf{D} / \partial t$	$\nabla \cdot \mathbf{B} = 0$	$\nabla \cdot \mathbf{D} = 0$
$\bar{\bar{\epsilon}} = \begin{pmatrix} \epsilon_{\perp} & 0 & 0 \\ 0 & \epsilon_{\perp} & 0 \\ 0 & 0 & \epsilon_{\parallel} \end{pmatrix}$	$D_i = \epsilon_0 \epsilon_{ij} E_j$	$B_i = \mu_0 (\delta_{ij} + \chi_{ij}) H_j$	

Light propagation in a homogeneous medium

Consider a plane monochromatic wave: $\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_0 \exp(i\mathbf{k} \cdot \mathbf{r} - i\omega t)$, $\mathbf{H}(\mathbf{r}, t) = \dots$

$$\mathbf{k} \times \mathbf{E} = \omega \mathbf{B} \qquad -\mathbf{k} \times \mathbf{H} = \omega \mathbf{D} \qquad \mathbf{k} \cdot \mathbf{D} = 0 \qquad \mathbf{k} \cdot \mathbf{H} = 0$$

Eliminating mag. field $\mathbf{H}$: $\omega^2 \mu_0 \mathbf{D} = k^2 \mathbf{E} - \mathbf{k}(\mathbf{E} \cdot \mathbf{k})$ and using constitutive eq. for $\mathbf{D}$:

 $\omega^2 \mu_0 \epsilon_0 \epsilon_{ij} E_j = k^2 E_i - k_i (E_j k_j)$

$$(N^2 \delta_{ij} - N_i N_j - \epsilon_{ij}) E_j = 0 \qquad \text{Refractive index "vector"} \quad \mathbf{N} = \mathbf{k} \frac{1}{\omega \sqrt{\epsilon_0 \mu_0}} \propto \frac{c}{v}$$

Fresnel equation; Propagation of Light in LC

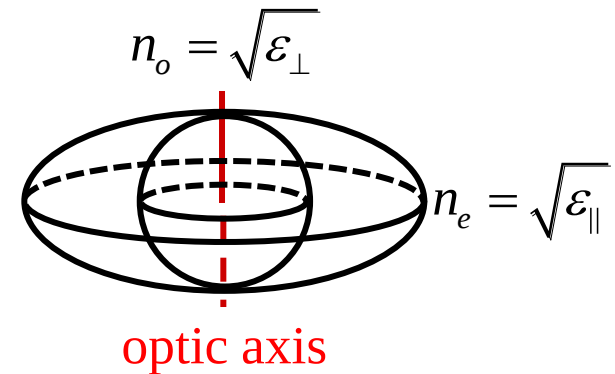
$$\left(N^2 \delta_{ij} - N_i N_j - \varepsilon_{ij}\right) E_j = 0$$

The three homogeneous equations have a nontrivial solution only if the determinant of coefficients vanishes (**Fresnel equation**):

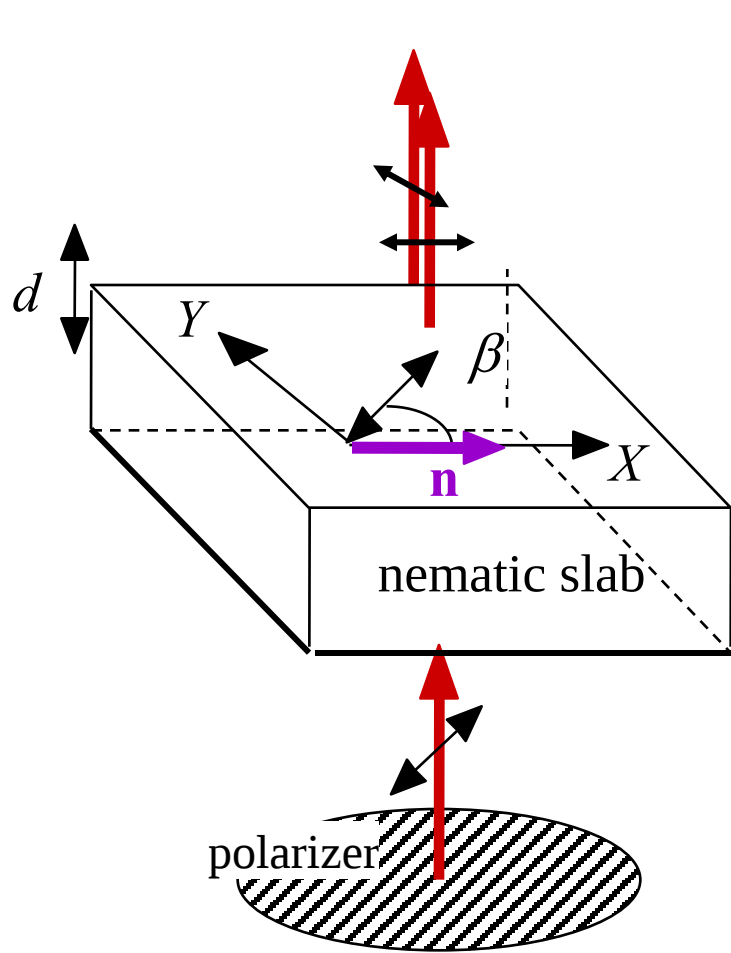
$$\text{Det}(\mathbf{k}, \omega) = \left(N^2 - \varepsilon_{\perp}\right) \left[\varepsilon_{\parallel} N_z^2 + \varepsilon_{\perp} \left(N_x^2 + N_y^2\right) - \varepsilon_{\parallel} \varepsilon_{\perp} \right] = 0$$

Two waves: ordinary, with $N = n_o = \sqrt{\varepsilon_{\perp}}$ and extraordinary, with refractive index that depends on the angle between the wave-vector and the optic axis:

$$N = n_{e,\text{eff}} = \frac{n_o n_e}{\sqrt{n_e^2 \cos^2 \theta + n_o^2 \sin^2 \theta}}$$



Polarizing microscopy: Principle



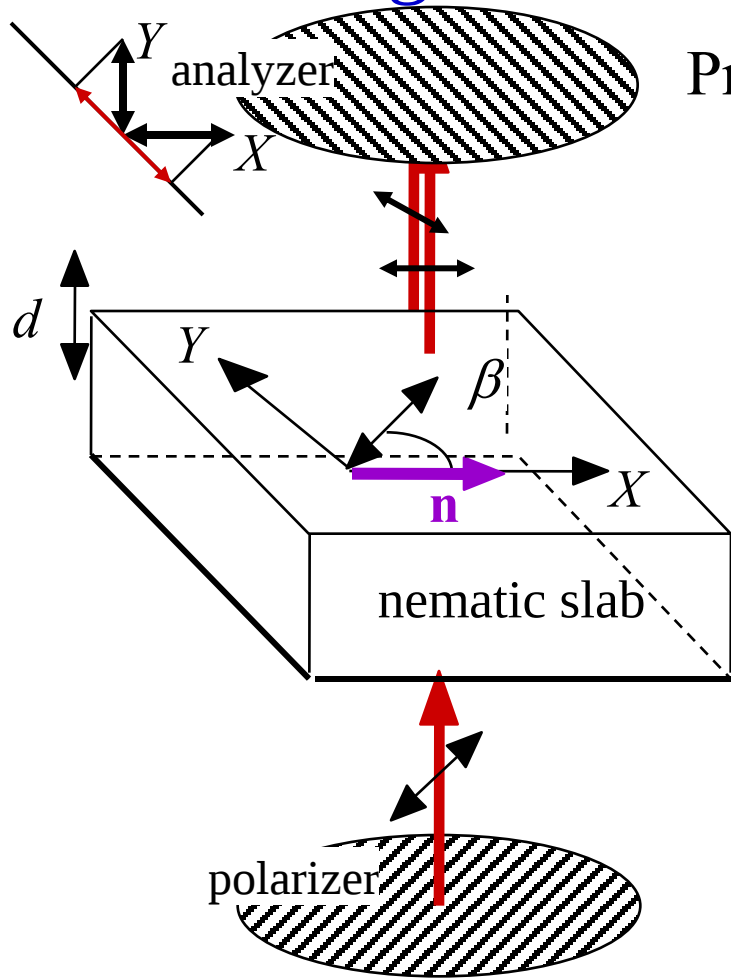
$$\text{Exit : } \begin{cases} A \cos \beta \cos \left(\omega t - \frac{2\pi}{\lambda_0} n_o d \right) \\ A \sin \beta \cos \left(\omega t - \frac{2\pi}{\lambda_0} n_e d \right) \end{cases}$$

$$\text{Phase difference } \Delta \varphi = \frac{2\pi d}{\lambda_0} (n_e - n_o)$$

$$\text{Entry : } \begin{cases} A \cos \beta \\ A \sin \beta \end{cases}$$

$$\text{Time to propagate : } \begin{cases} \frac{n_o d}{c}, \text{ ordinary} \\ \frac{n_e d}{c}, \text{ extraordinary} \end{cases}$$

Polarizing microscopy: Principle



Projected onto analyzer' direction:

$$\begin{cases} A_1 = A \sin \beta \cos \beta \cos \left(\omega t - \frac{2\pi}{\lambda_0} n_o d \right) \\ A_2 = A \sin \beta \cos \beta \cos \left(\omega t - \frac{2\pi}{\lambda_0} n_e d \right) \end{cases}$$

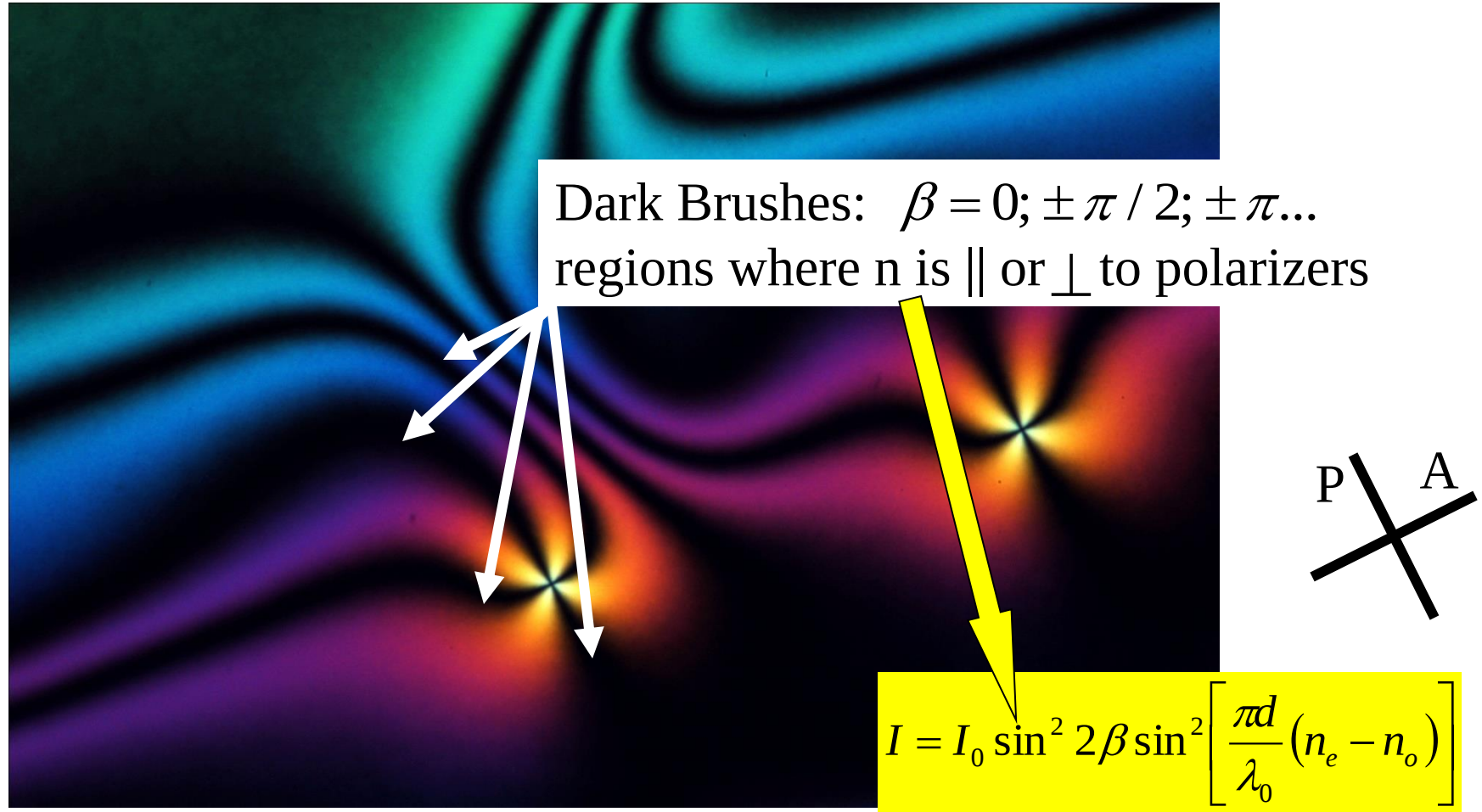
Resulting vibration: $\bar{A} \cos(\omega t + \bar{\varphi})$

with amplitude

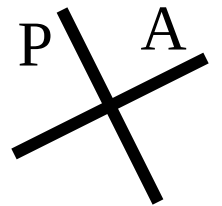
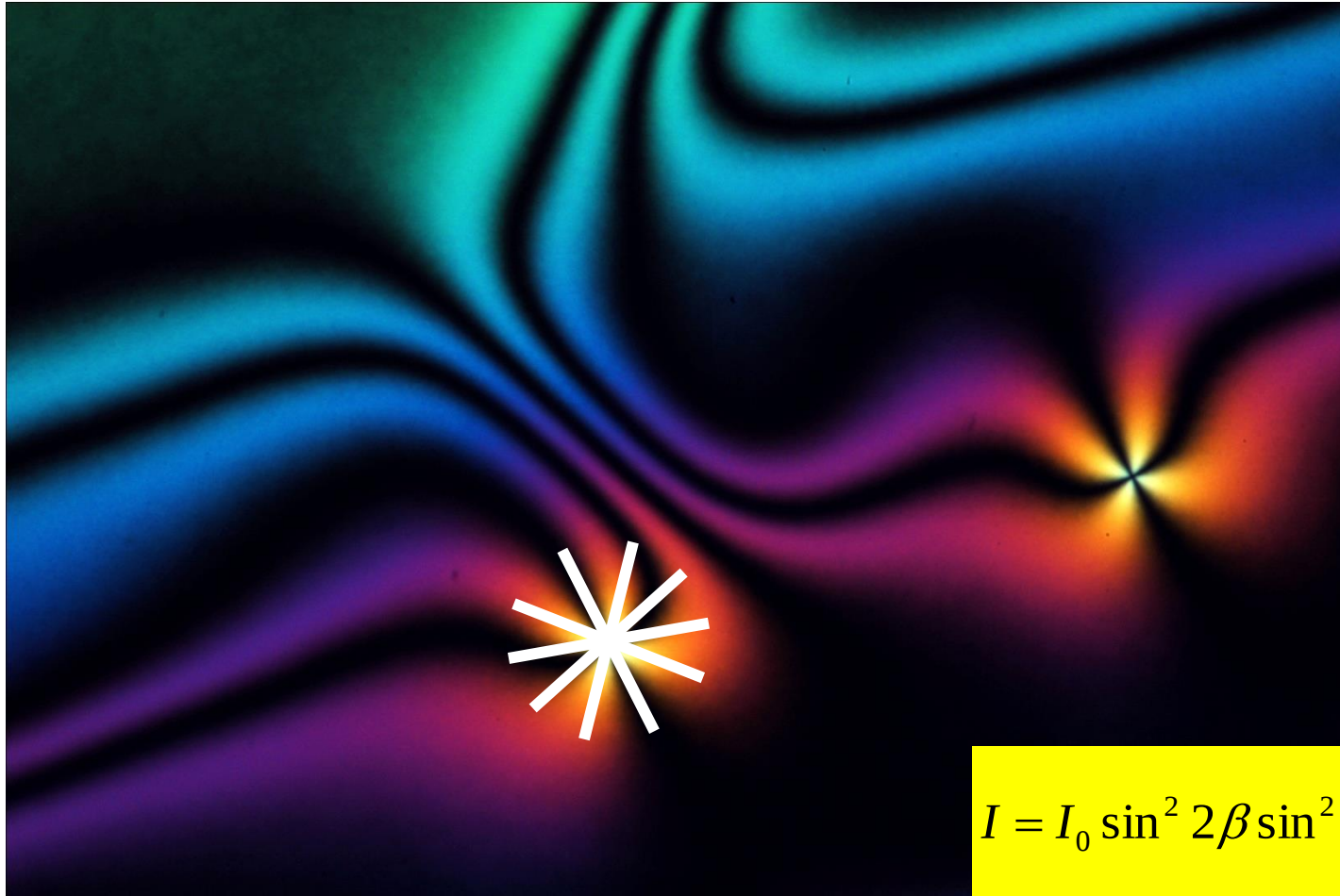
$$\bar{A}^2 = A_1^2 + A_2^2 + 2A_1A_2 \cos \Delta\phi$$

$$I = I_0 \sin^2 2\beta \sin^2 \left[\frac{\pi d}{\lambda_0} (n_e - n_o) \right]$$

Polarizing microscopy: Principle

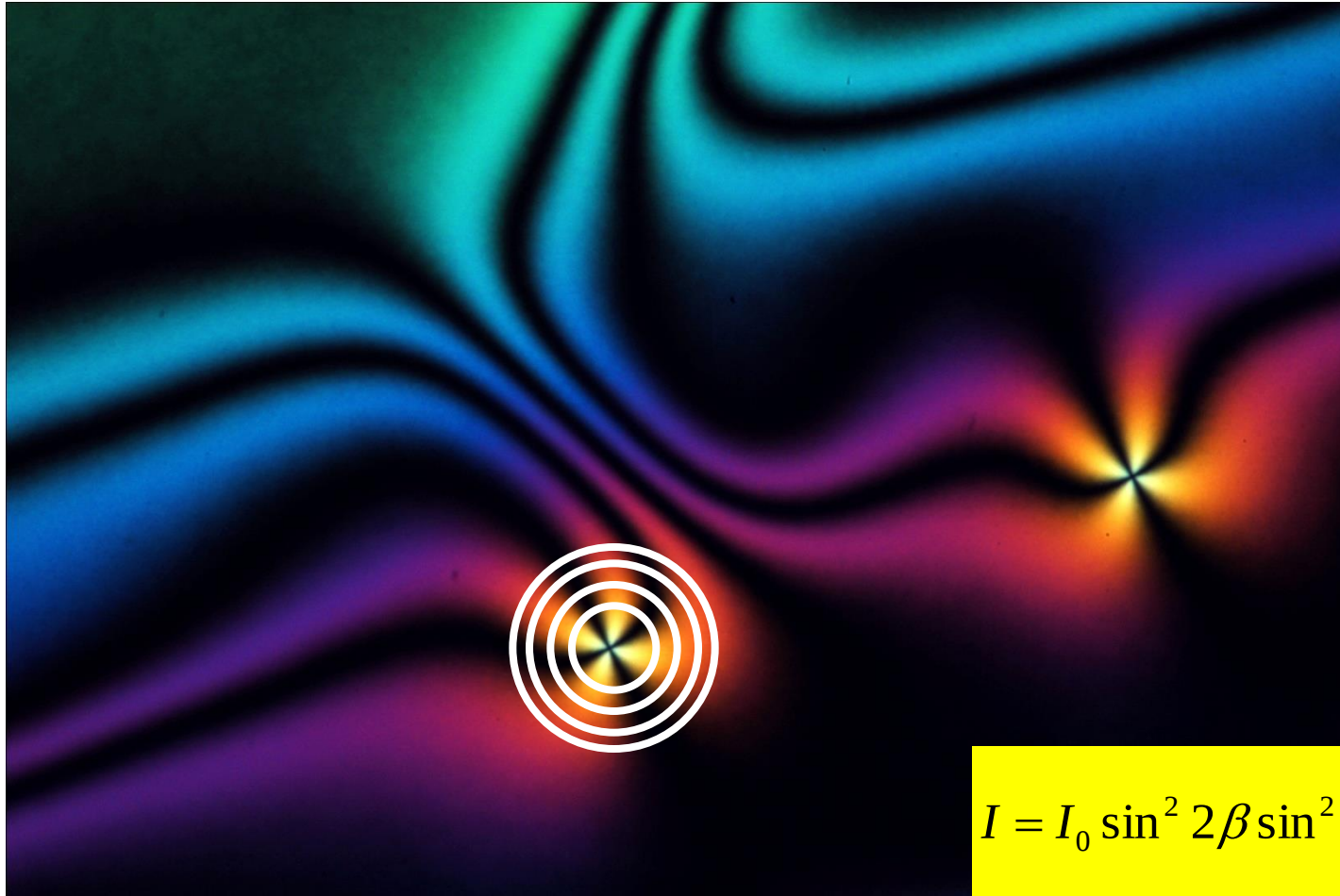


Polarizing microscopy: Degeneracy of two director fields



$$I = I_0 \sin^2 2\beta \sin^2 \left[\frac{\pi d}{\lambda_0} (n_e - n_o) \right]$$

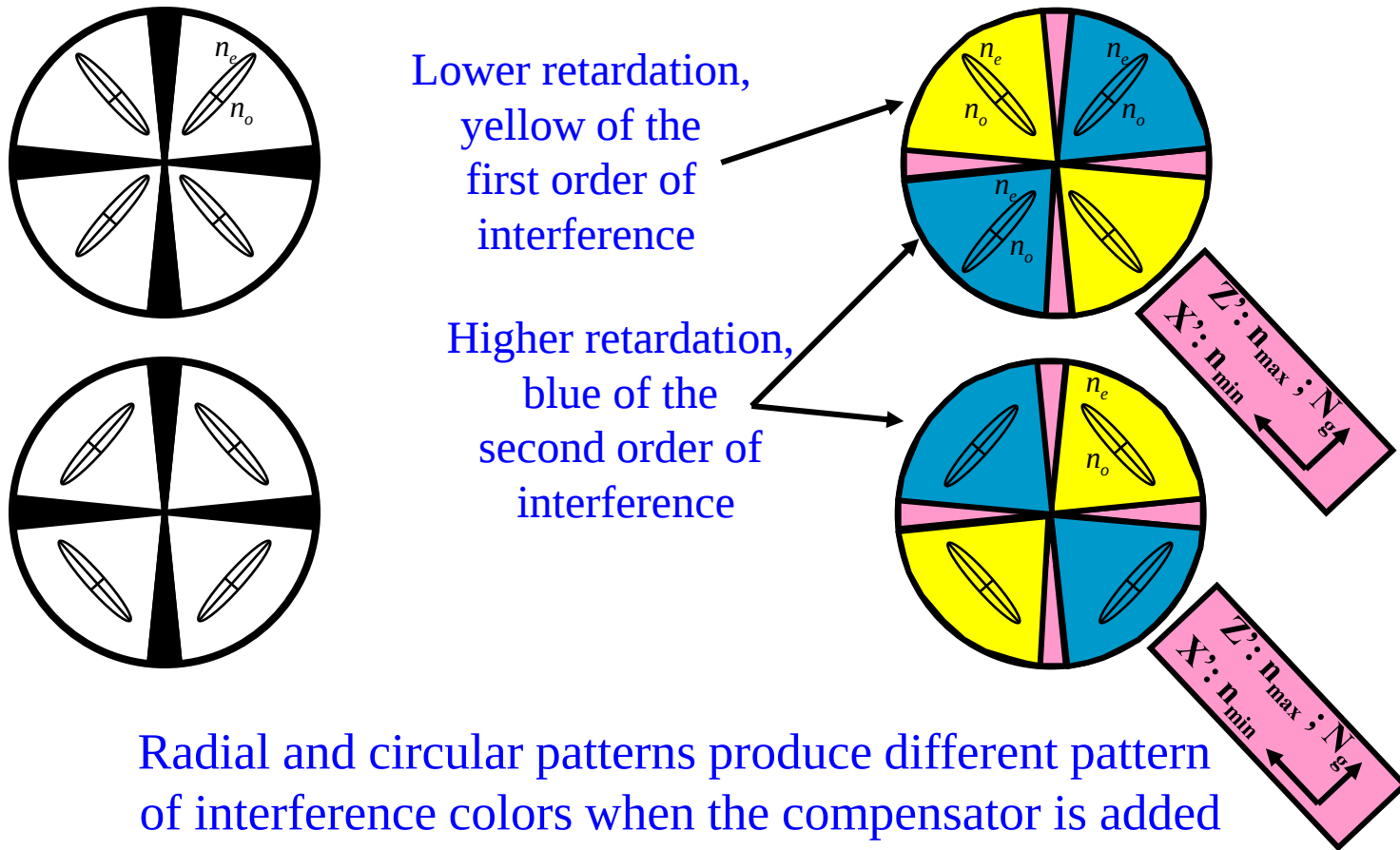
Polarizing microscopy: Degeneracy of two director fields: Radial or Circular?



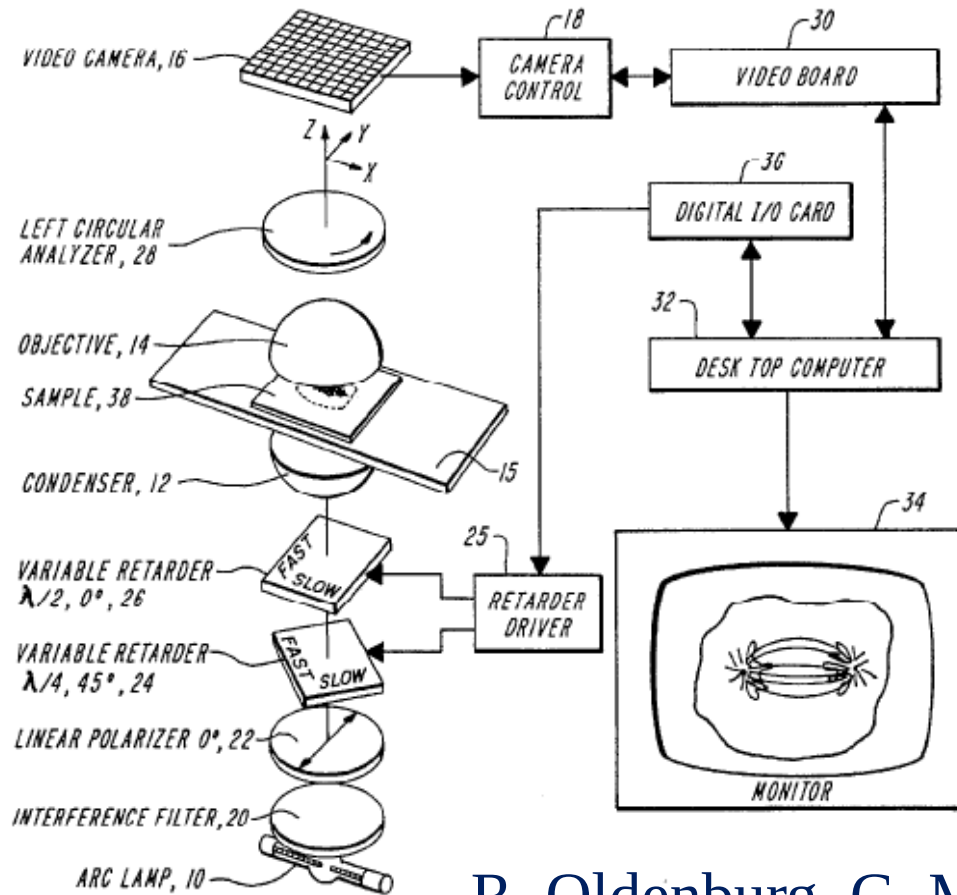
$$I = I_0 \sin^2 2\beta \sin^2 \left[\frac{\pi d}{\lambda_0} (n_e - n_o) \right]$$

PM problem: Two orthogonal $\mathbf{n}$ fields

Solution: optical compensator (quartz wedge, Red plate, etc.)

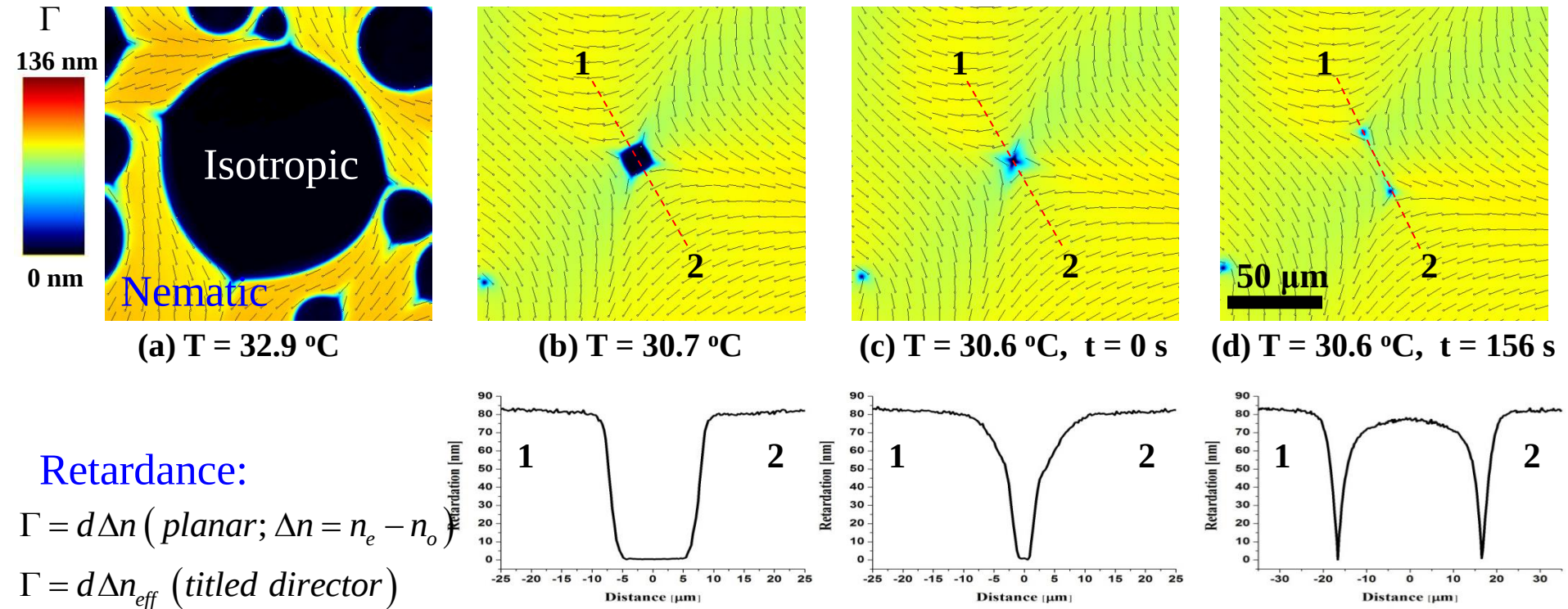


Ultimate Compensator: LC PolScope



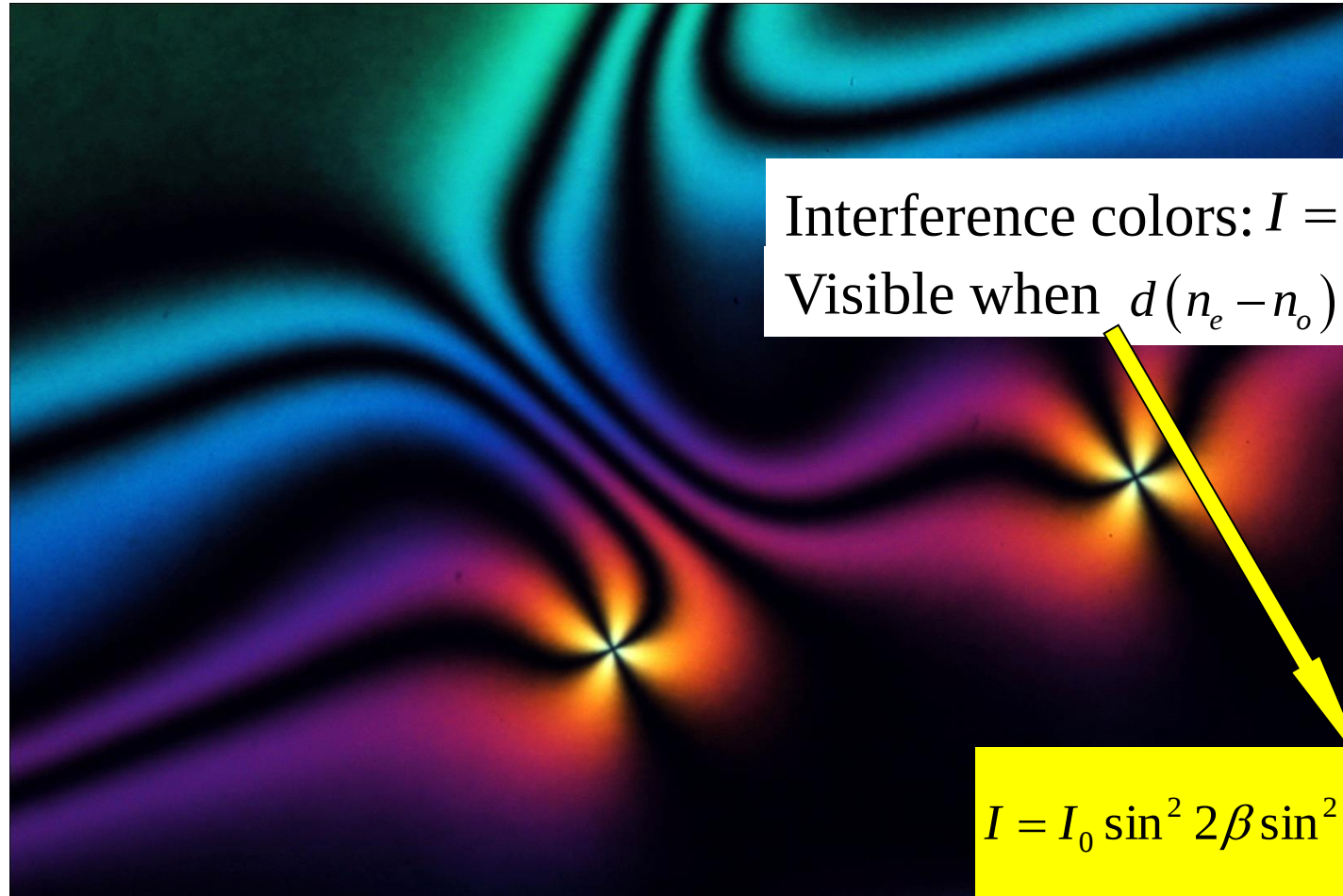
R. Oldenburg, G. Mei, US Patent 5,521,705

LC PolScope image of chromonic: shows both the in-plane director and retardance



PolScope creates a map of the orientation of the optic axis in the sample and of the local value of the optical phase retardation;

Polarizing microscopy

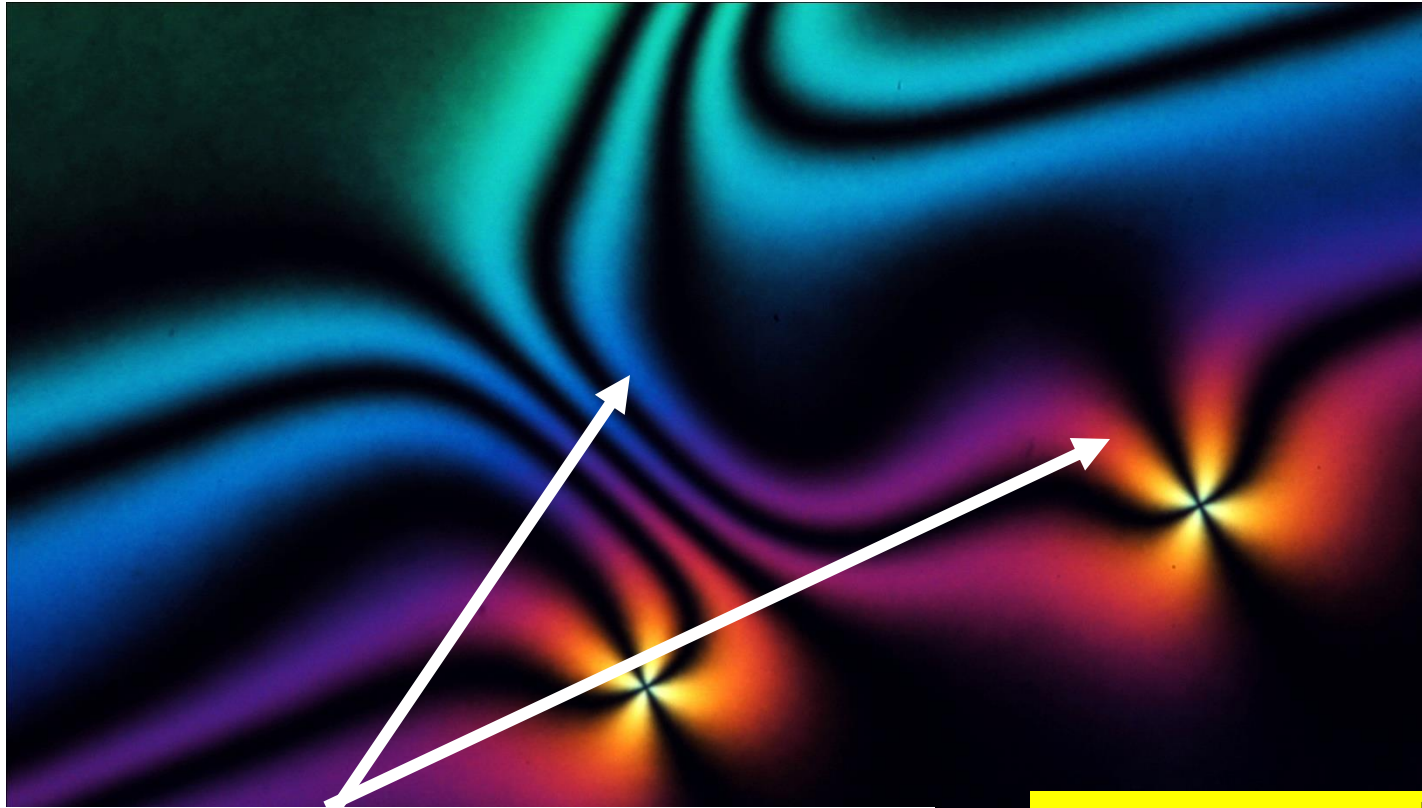


Interference colors: $I = I[\lambda_0]$

Visible when $d(n_e - n_o) \sim (1-3)\lambda_0$

$$I = I_0 \sin^2 2\beta \sin^2 \left[\frac{\pi d}{\lambda_0} (n_e - n_o) \right]$$

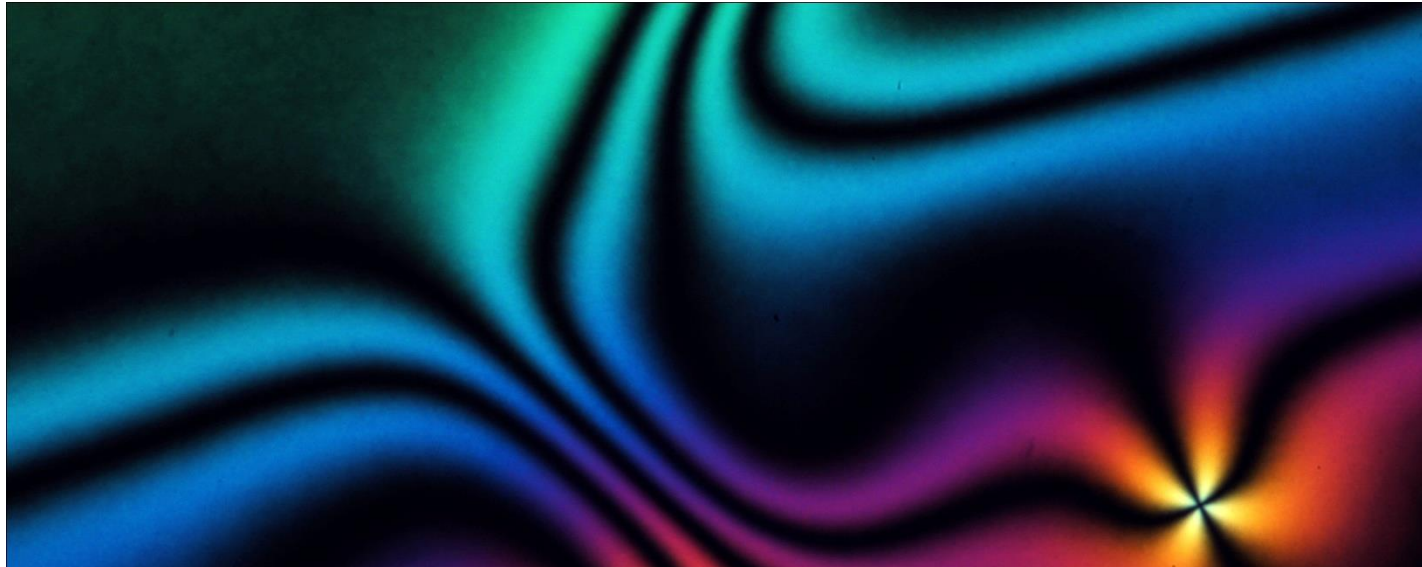
Limitation: 2D image



Why the interference colors change?

$$I = I_0 \sin^2 2\beta \sin^2 \left[\frac{\pi d}{\lambda_0} (n_e - n_o) \right]$$

Limitation: 2D image

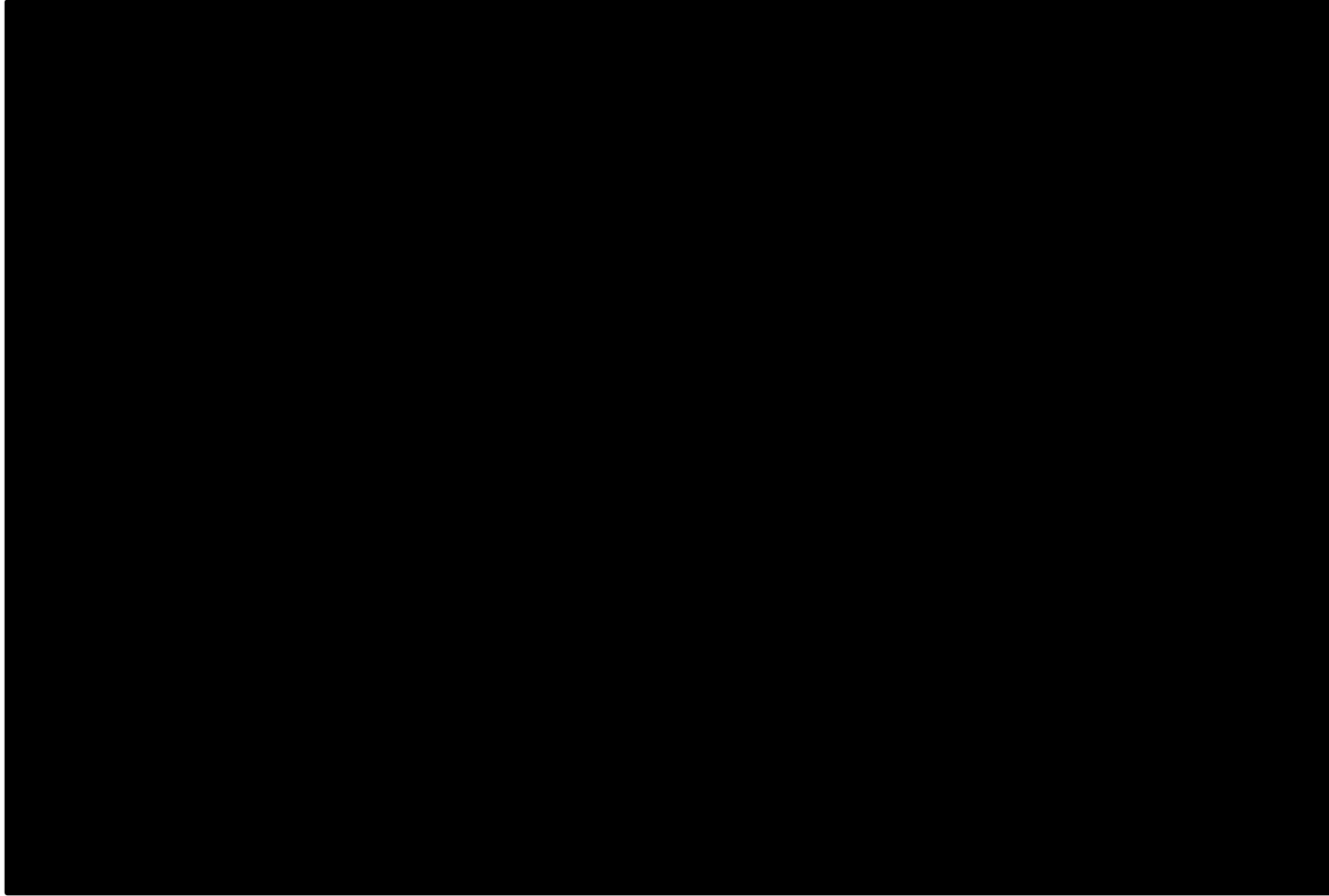


Derived in approximation of planar director, $\hat{\mathbf{n}} = (n_x, n_y, 0)$
and constant thickness d


$$I = I_0 \sin^2 2\beta \sin^2 \left[\frac{\pi d}{\lambda_0} (n_e - n_o) \right]$$

BTW!

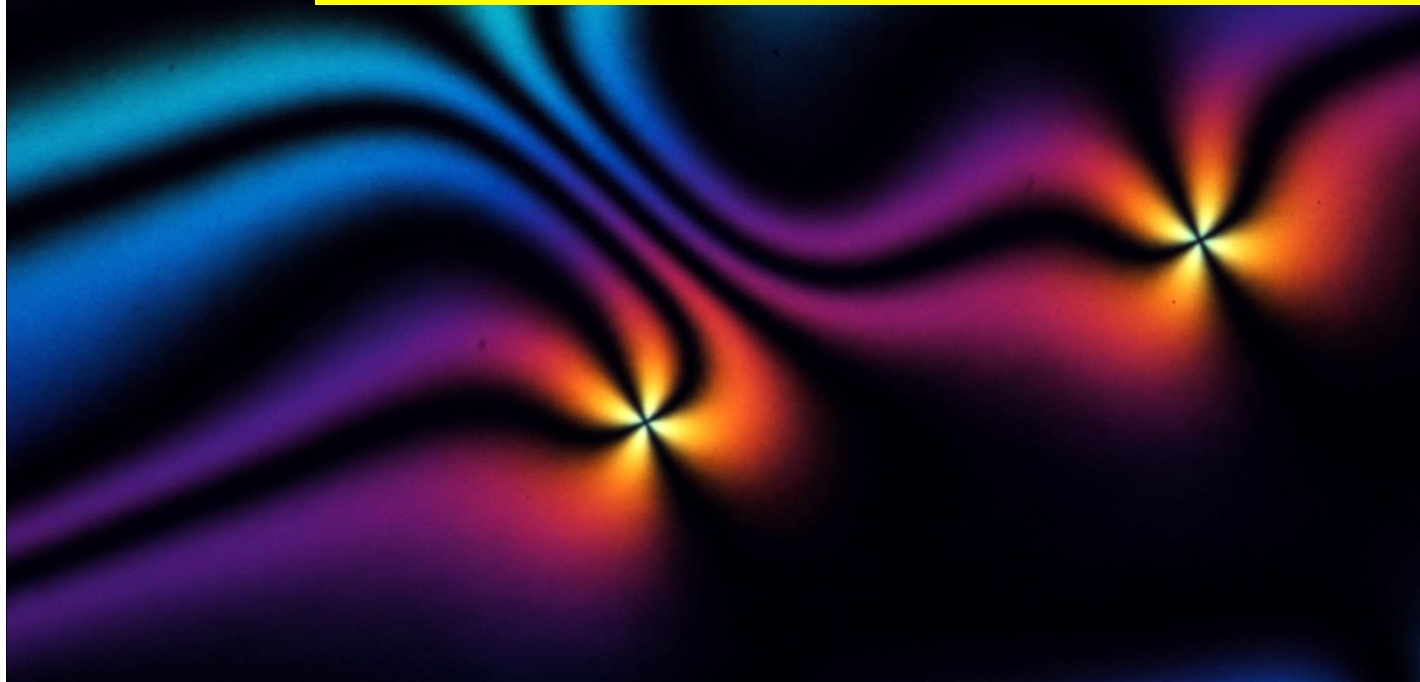
When $\{n_x, n_y, n_z\} = \{0, 0, 1\}$, how does the texture look like?



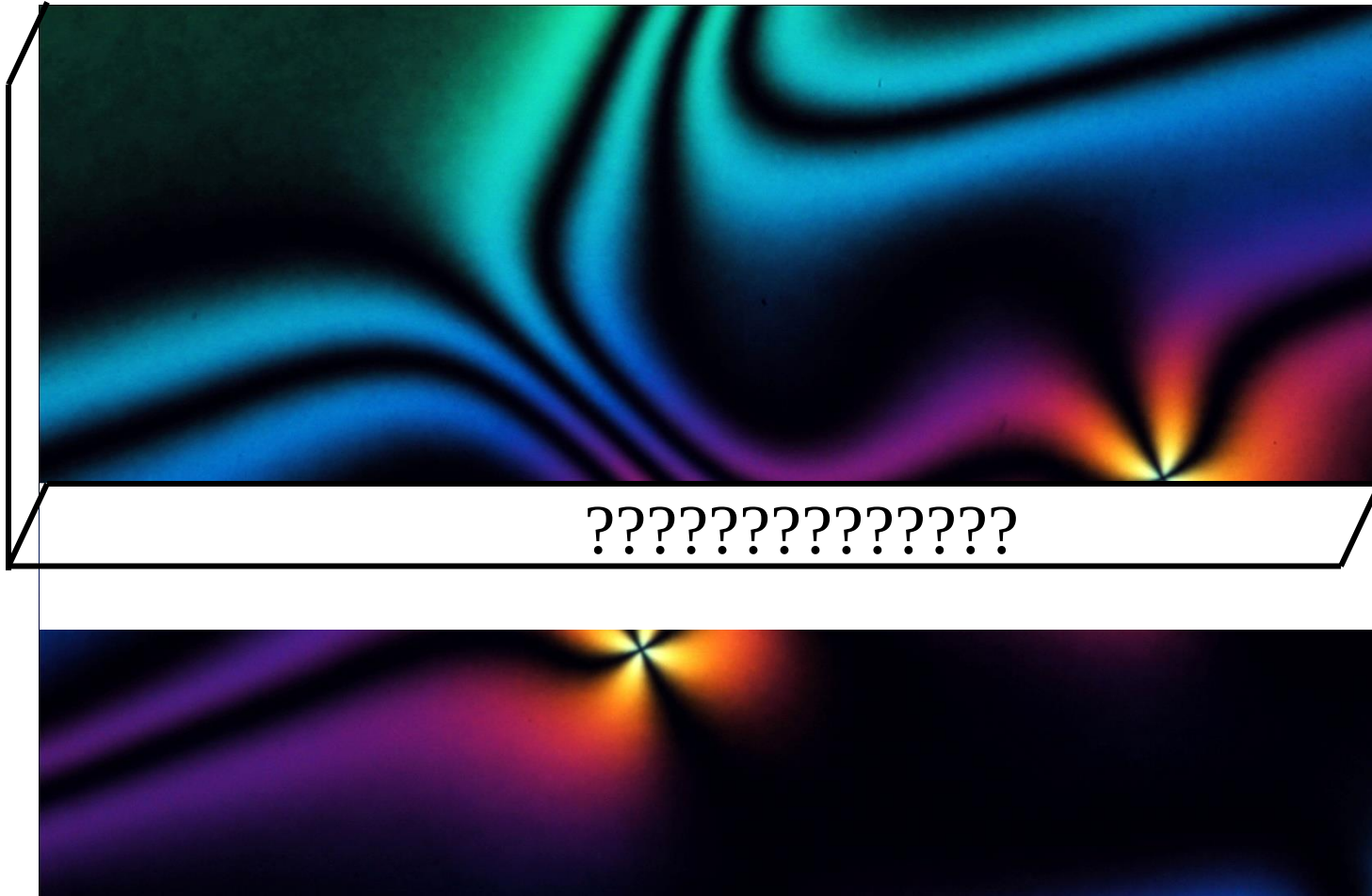
Limitation: 2D image

When $\mathbf{n} = \mathbf{n}(x, y, z)$ and $d = d(x, y)$, one deals with:

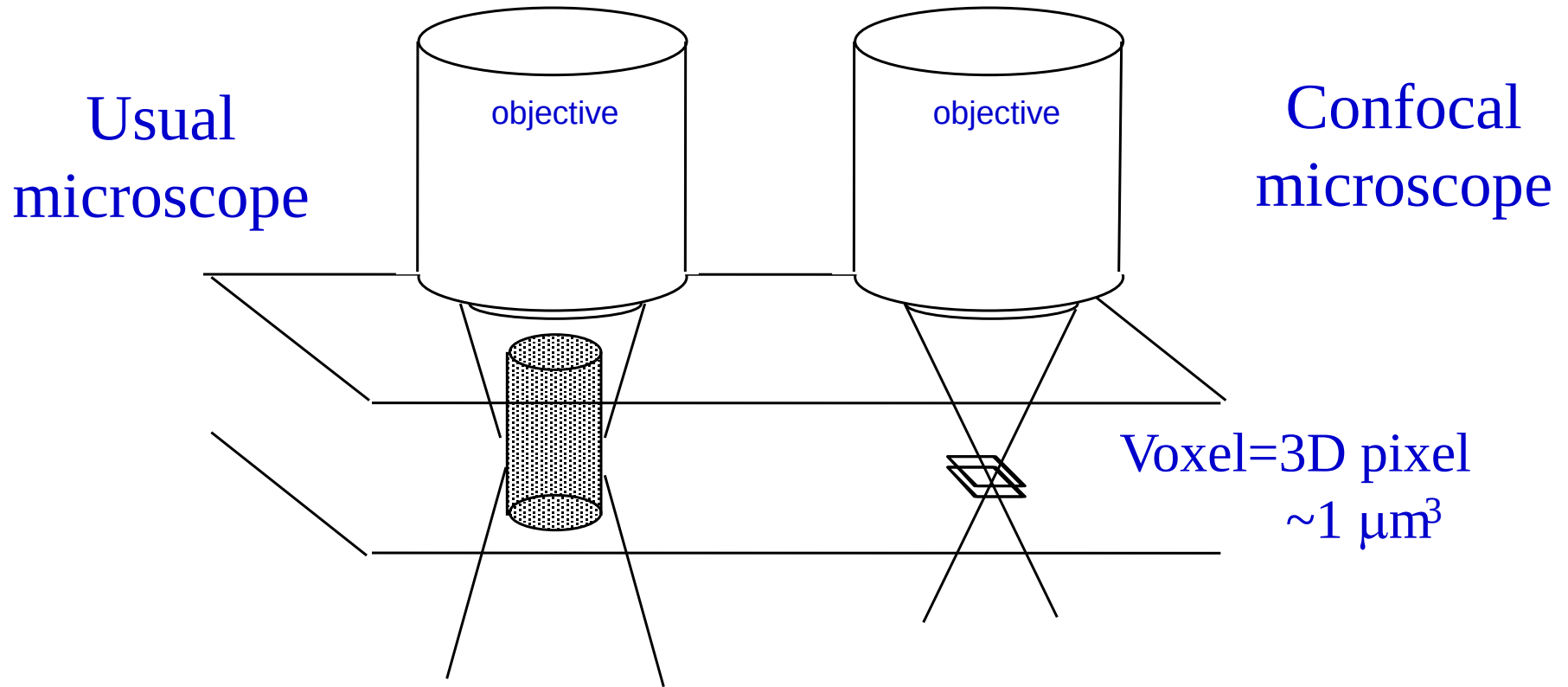
$$I = I_0 \sin^2 2\beta \sin^2 \frac{\pi}{\lambda} \int_0^d f[\mathbf{n}(x, y, z), n_{\text{eff}}(x, y, z), d(x, y)] dz$$



Limitation: 2D image

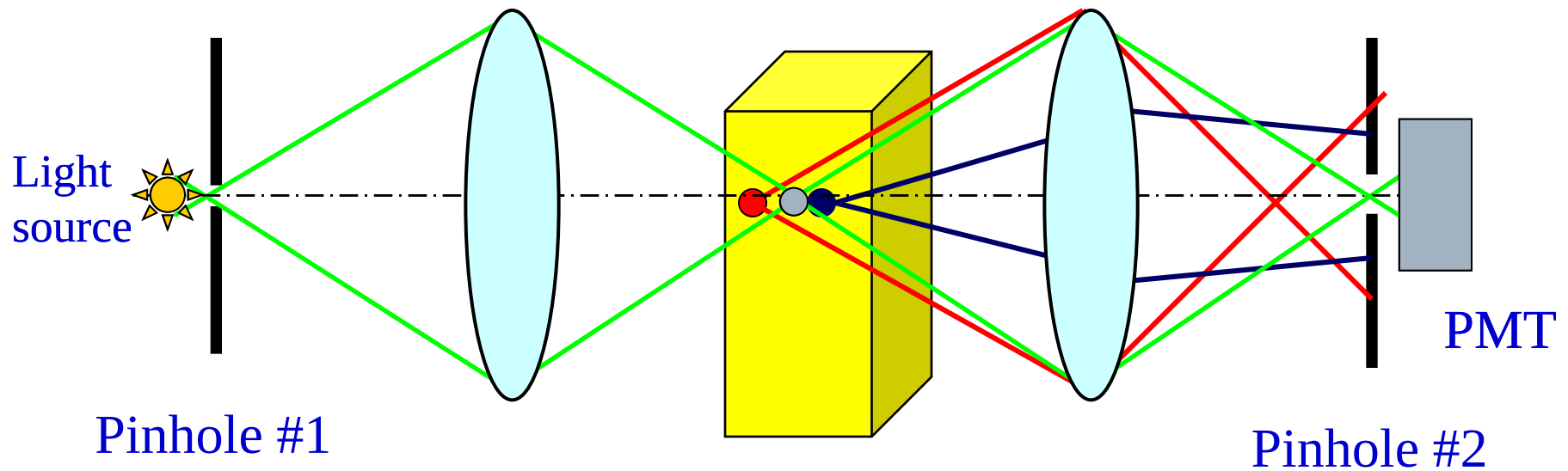


Confocal Microscopy: 3D image of 3D



Confocal Microscopy: Principle

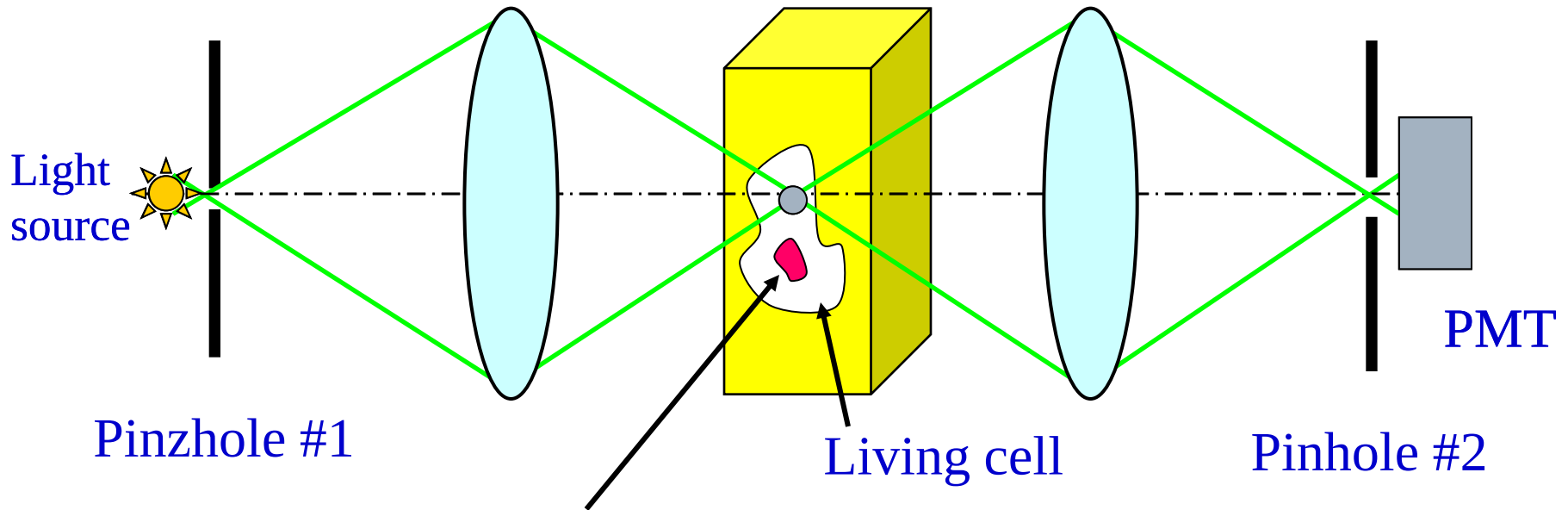
(Minsky, 1957)



→ Scanning → Computer → 3D image

Fluorescence Confocal Microscopy

1980 M. Petran and A. Boyde



Fluorescent tag increases contrast of tissues



3D image of **concentration (positional distribution)** of fluorescent dopant....

....but we are interested **in orientations**
rather than in **concentrations...**

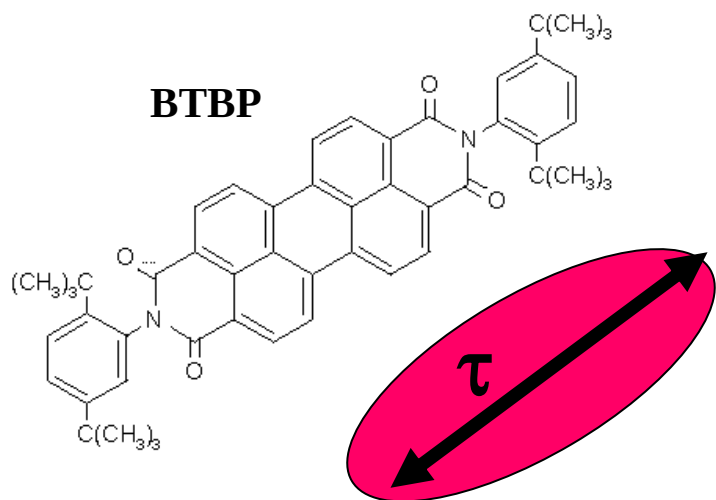


Fluorescence Confocal **Polarizing** Microscopy

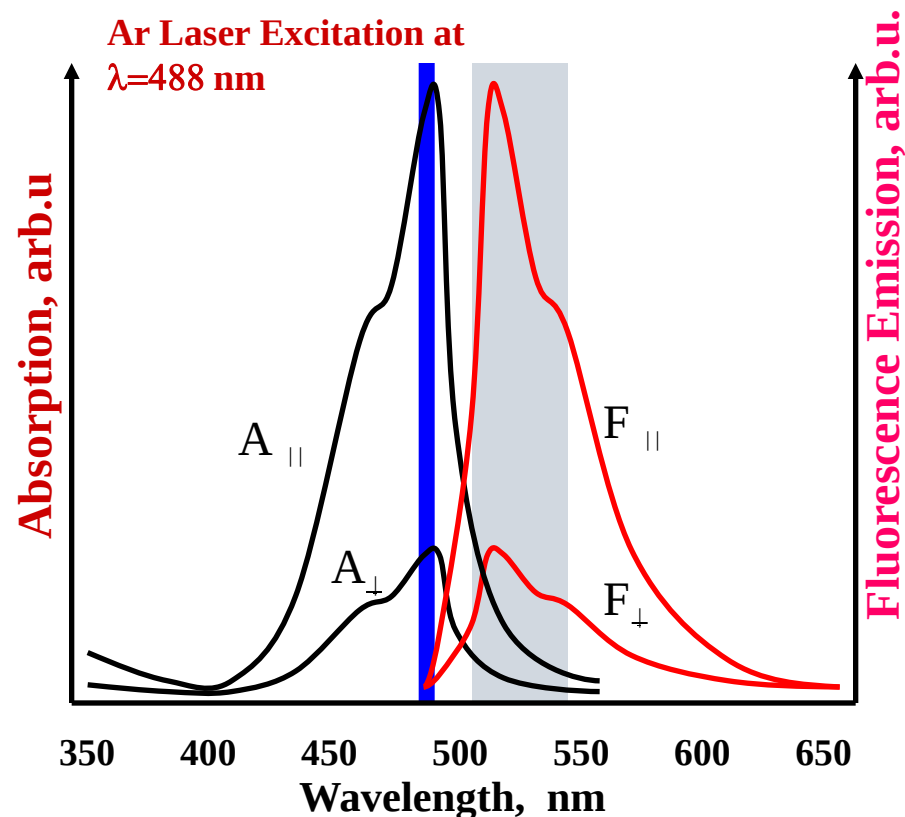
Two distinctive features:

1. Anisometric fluorescent dye aligned by LC
2. Polarized light

FCPM: Fluorescent anisometric dyes

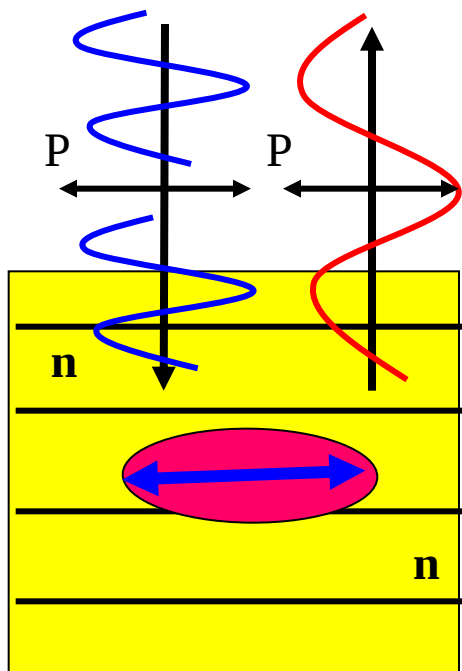


N,N'-Bis(2,5-di-tert-butylphenyl)-3,4,9,10-perylenedicarboximide)
BTBP

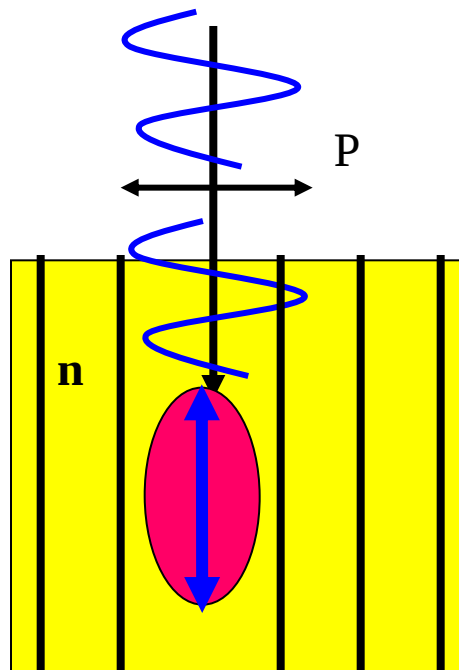


0.01 % of BTBP in ZLI-3412

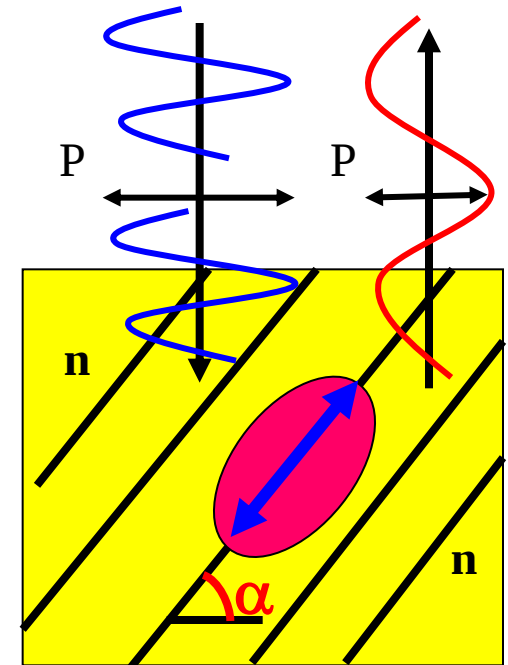
FCPM: Anisotropic Fluorescence



maximum signal



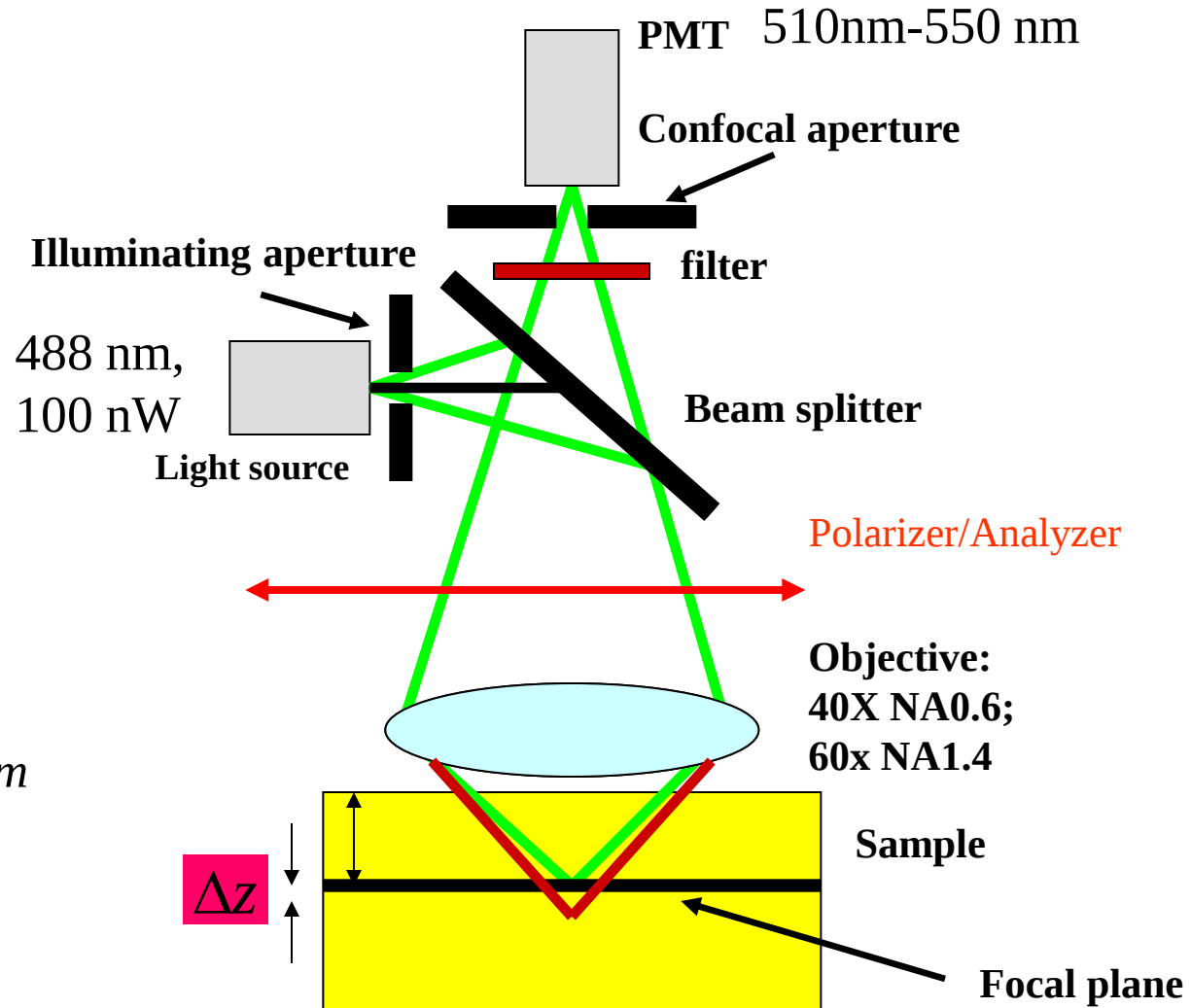
minimum signal



$$I \sim \cos^4 \alpha$$

Fluorescence signal = f (orientation of dye)

FCPM: Anisotropic Fluorescence

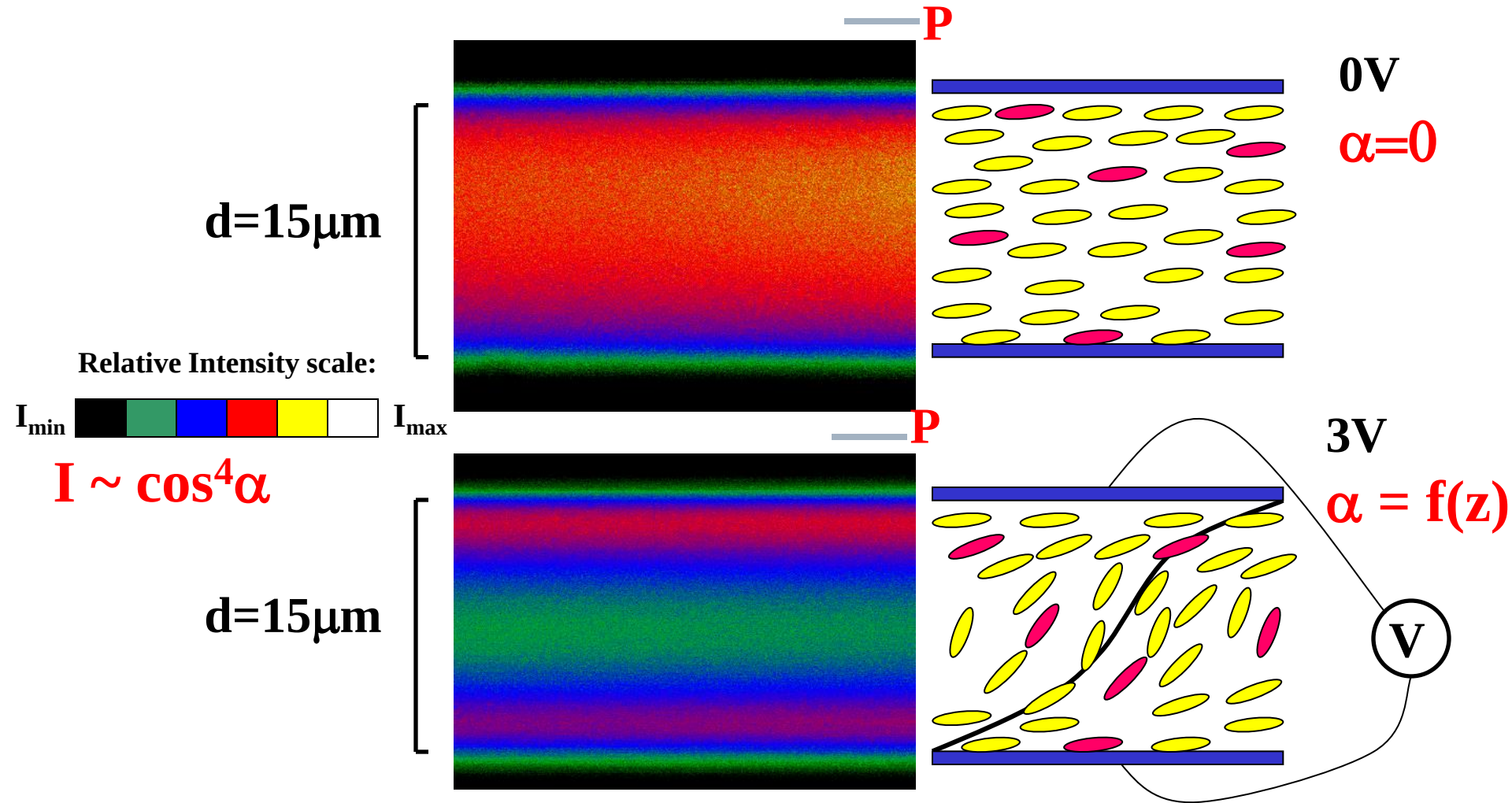


$$\Delta z \approx |n_e - n_o| \cdot z / \bar{n}$$

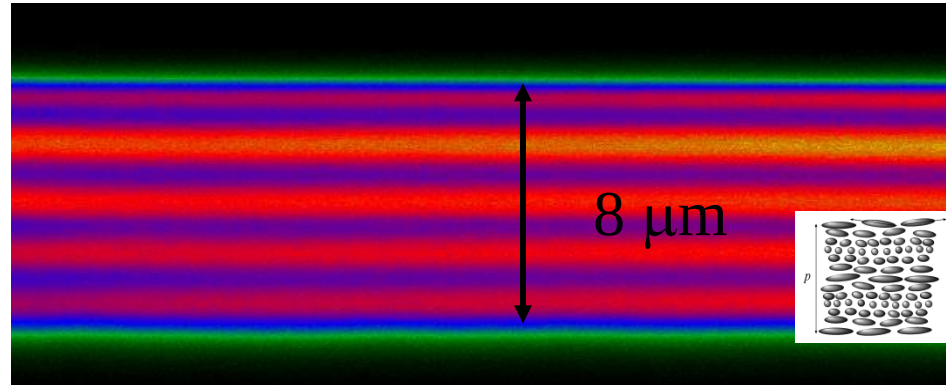
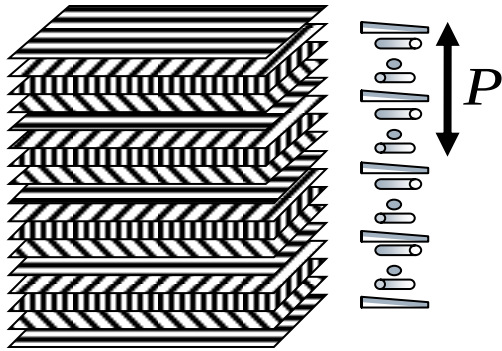
$$|n_e - n_o| / \bar{n} \approx 0.1; z \approx 10 \mu m$$

$$\Delta z \sim 1 \mu m$$

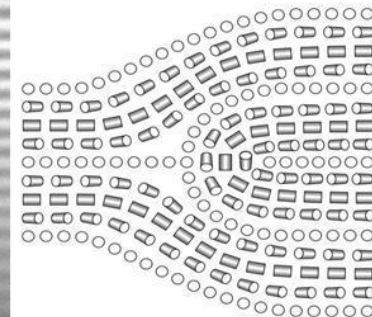
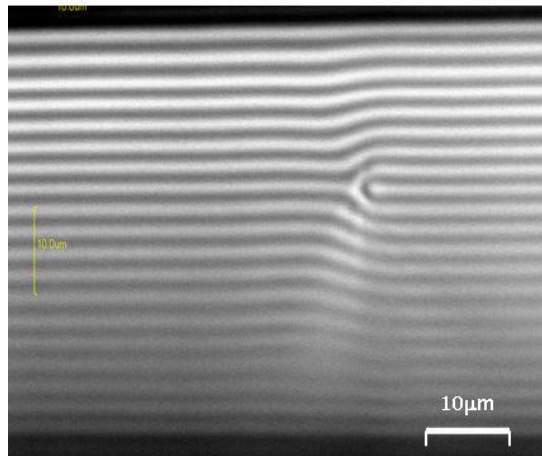
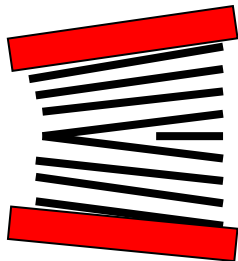
FCPM: Frederiks Effect



FCPM: Planar Cholesteric



$P\Delta n/2\lambda < 1$ (to avoid the Mauguin regime)



Summary: What have you learned

□ Liquid crystals: Orientationally ordered media

- Thermotropic (t-driven), lyotropic (c-driven), chromonic (t,c-driven)
- Uniaxial nematic, twist bend, cholesteric, smectic, columnar, dramatic dependence on molecular structure...

□ Orientational elasticity vs surface anchoring

- Frank elastic constants ~ 5 pN
- Equilibrium director defined by boundary conditions (anchoring), elasticity, and external field
- As the system become larger, anchoring imposes stronger restrictions on the director
- Frederiks transitions @ heart of modern LCDs

□ Optics

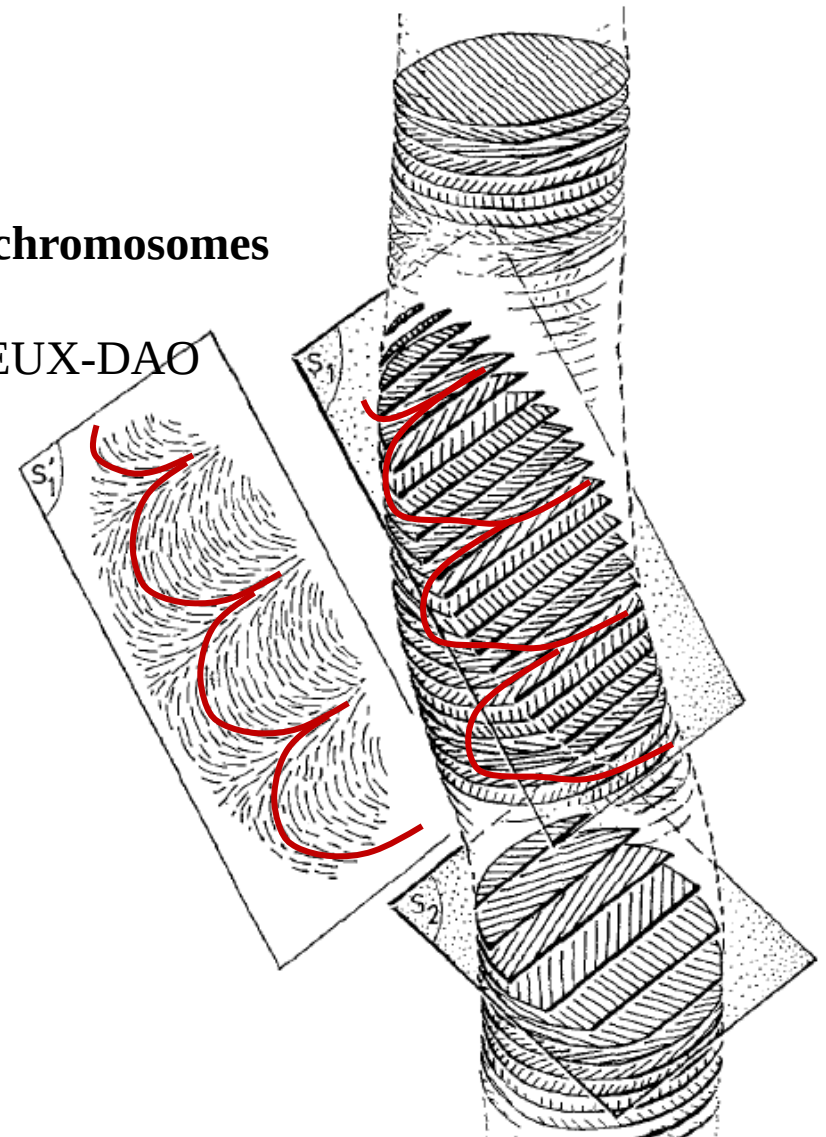
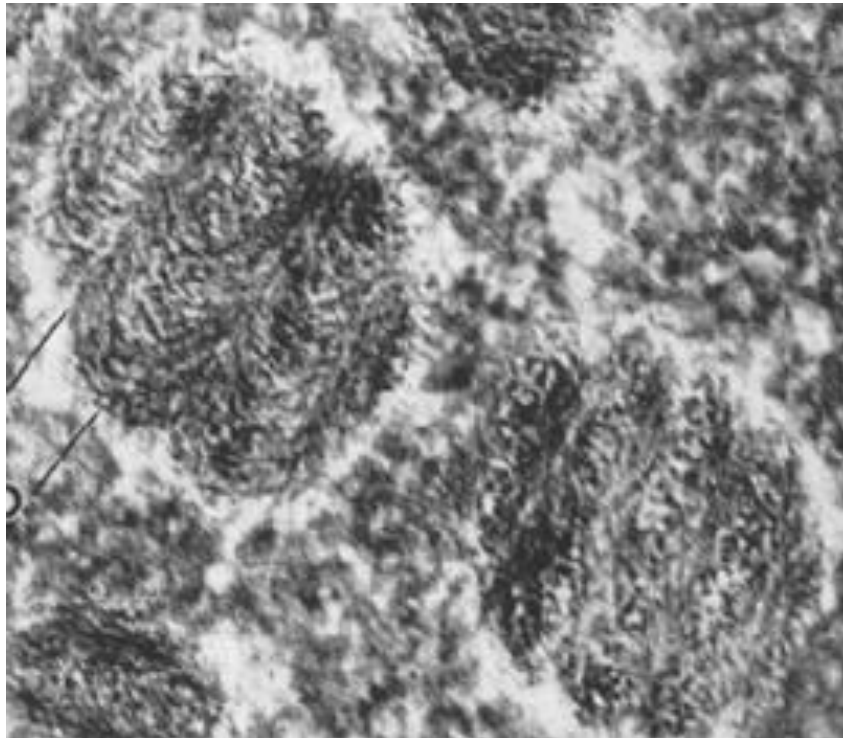
- LCs are birefringent; ordinary and extraordinary waves
- Polarizing microscope: 2D image of 3D sample
- Fluorescence confocal polarizing microscopy: 3D image of 3D orientational order

Cholesteric structure of DNA in chromosomes: Bouligand arches

Chromosoma (Berl.) 24, 251--287 (1968)

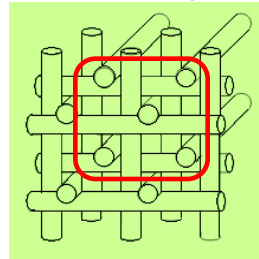
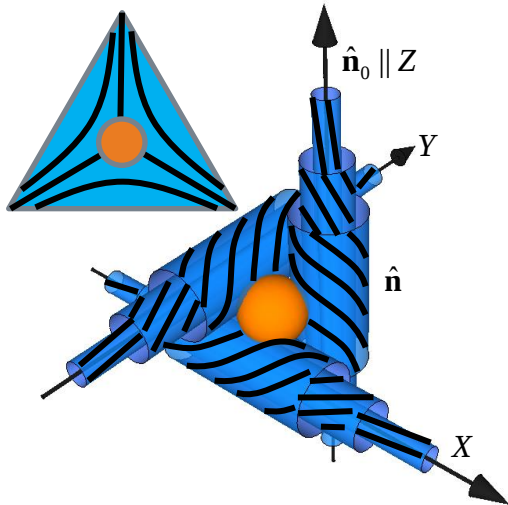
**La structure fibrillaire et l'orientation des chromosomes
chez les Dinoflagellés**

Y. BOULIGAND, M.-O. SOYER et S. PUISEUX-DAO



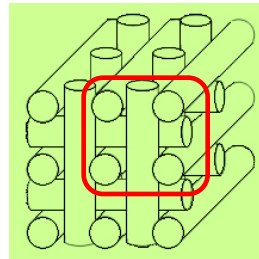
Colesteric: 1D twist; Blue phases: 3D twist

Double twisted cylinders stabilized by a 3D network of topological defects –

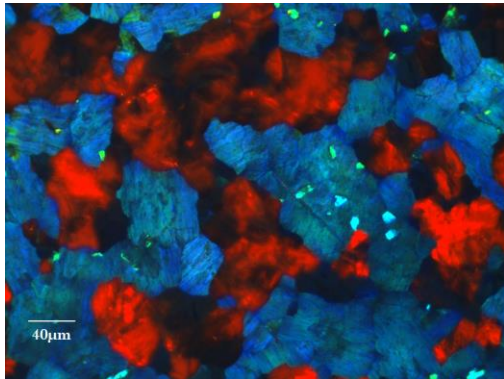


BPI

100 nm



BPII



Polymer mesh formed in BPII
Can be refilled with N for
electro-optic applications
J. Xiang et al, APL **103**, 051112
(2013)

Main problem for applications:
Temperature range of BPs is narrow,
 $\sim 1^\circ\text{C}$

One approach: Polymerization
H. Kikuchi et al, Nature Mat. **1**, 64
(2002)

