

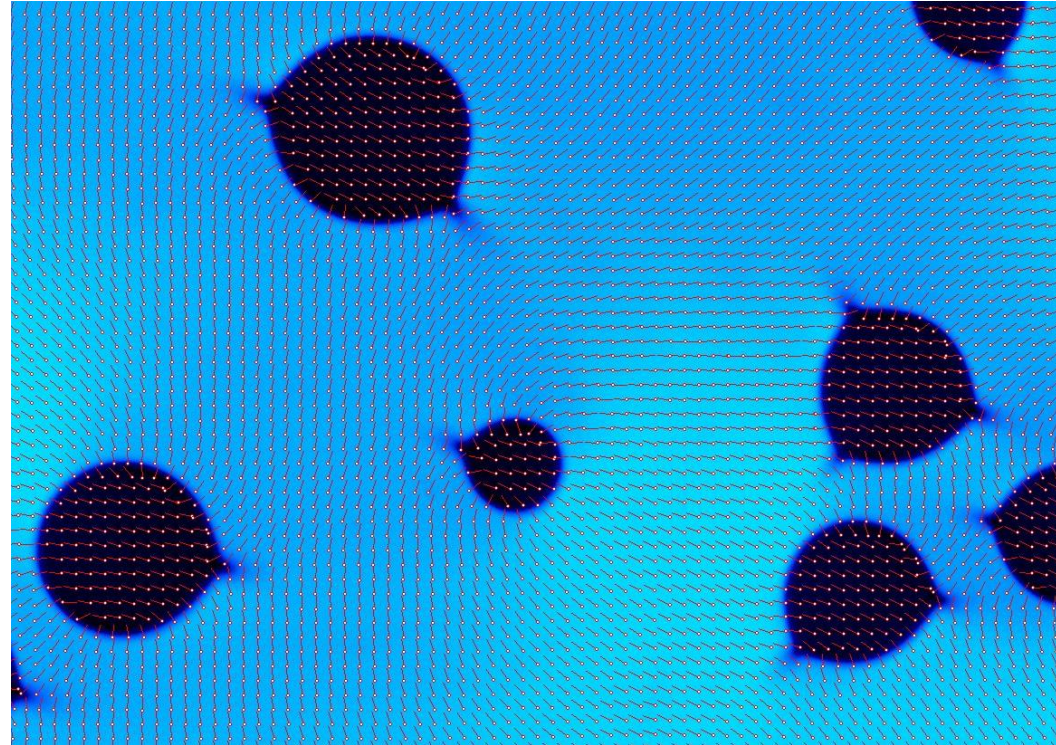
# Liquid crystals: Lecture 2

## Topological defects, Droplets and Lamellar phases

Oleg D. Lavrentovich

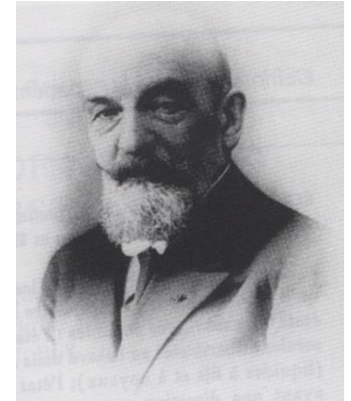
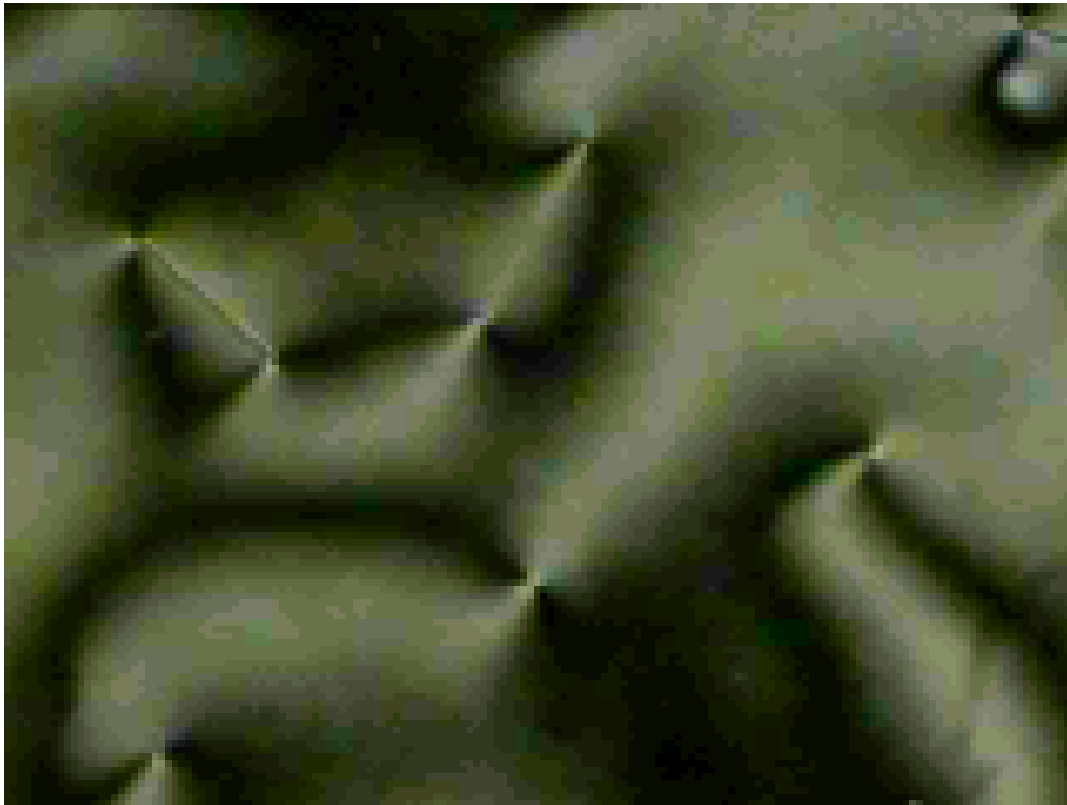
Support: NSF

Liquid Crystal Institute  
Kent State University, Kent, OH



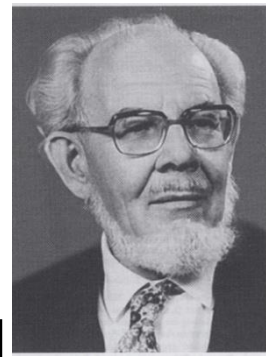
Boulder School for Condensed Matter and Materials Physics,  
Soft Matter In and Out of Equilibrium,  
6-31 July, 2015

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



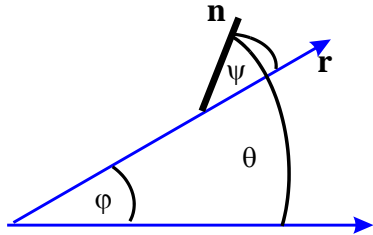
1922, G. Friedel:  
Named “nematics”, the simplest  
LC, after observing linear defects,  
 $\nu\epsilon\mu\alpha$ =thread, under a polarized  
light microscope

# Frank's model of disclinations in N



The simplest form (one constant) approximation:

$$f = \frac{1}{2} K \left[ (\text{div} \hat{\mathbf{n}})^2 + (\text{curl} \hat{\mathbf{n}})^2 \right]$$

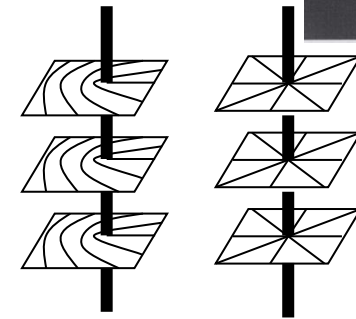


$$\{n_r, n_\phi, n_z\} = \{\cos \psi(\phi), \sin \psi(\phi), 0\}$$

$$\text{div} \hat{\mathbf{n}} = \frac{1}{r} \frac{dn_\phi}{d\phi} + \frac{n_r}{dr} = \frac{\cos \psi}{r} \left( 1 + \frac{d\psi}{d\phi} \right)$$

$$\text{curl}_z \hat{\mathbf{n}} = -\frac{1}{r} \frac{dn_r}{d\phi} + \frac{n_\phi}{r} = \frac{\sin \psi}{r} \left( 1 + \frac{d\psi}{d\phi} \right)$$

$$F_{\text{length}} = \frac{1}{2} K \int_S \left( 1 + \frac{\partial \psi}{\partial \phi} \right)^2 \frac{1}{r^2} r d\phi dr = \frac{1}{2} K \int_{\phi=0}^{\phi=2\pi} \left( 1 + \frac{\partial \psi}{\partial \phi} \right)^2 d\phi \int_{r=0}^{r=R} \frac{dr}{r}$$



$k=1/2$

$k=1$

Euler-Lagrange equation  $\Rightarrow \frac{\partial^2 \psi}{\partial \phi^2} = 0 \Rightarrow \psi = A\phi + c$

$$\oint d\psi = 2\pi k \Rightarrow A = 0, \pm 1/2, \pm 1, \dots \quad (n_r, n_\phi) = (\cos[\phi(k-1) + c], \sin[\phi(k-1) + c]) \quad (n_x, n_y) = (\cos[\phi k + c], \sin[\phi k + c])$$

$$F_{\text{length}} = \pi k^2 K \int_{r=0}^{r=R} \frac{dr}{r} = \pi k^2 K \ln \frac{R}{r_{\text{core}}} + F_{\text{core}}$$

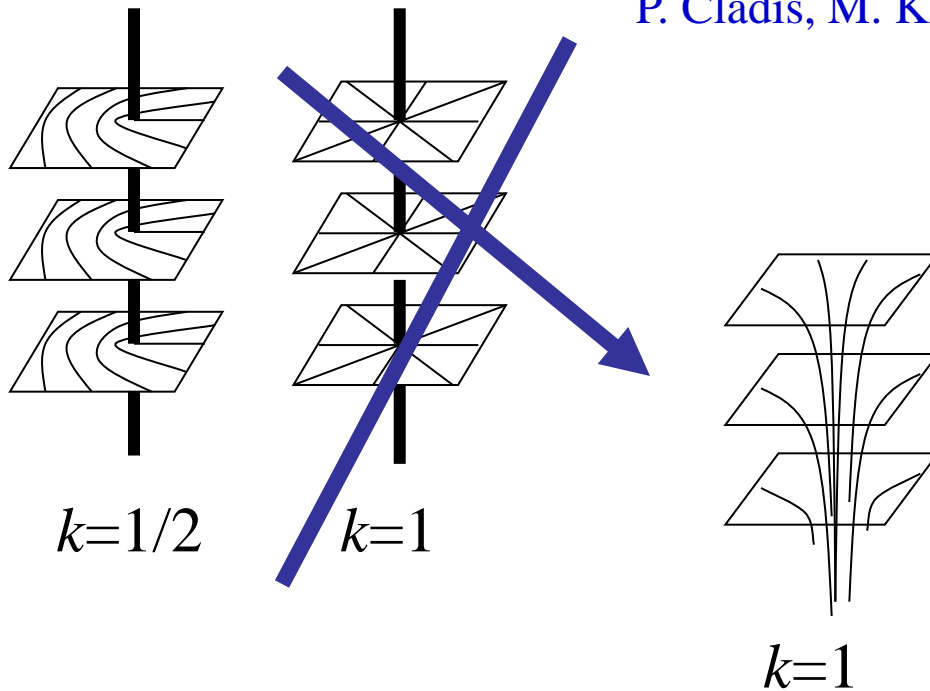
$$F_{\text{core}} = k_B (T_{NI} - T) \times \pi r_{\text{core}}^2 \times \rho N_A / M$$

Energy per degree of freedom      # degrees of freedom

$$r_{\text{core}} = k \sqrt{\frac{MK}{\rho N_A k_B (T_{NI} - T)}} \sim \text{few molecular lengths}; F_{\text{core}} \sim \pi k^2 K$$

# Escape into the 3<sup>rd</sup> dimension

P. Cladis, M. Kleman (1972), R.B. Meyer (1972)

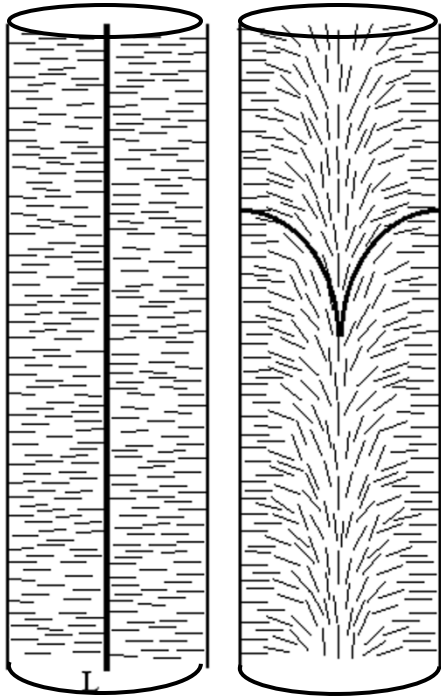




# Escape into the 3<sup>rd</sup> dimension

$$n_r = \cos\chi(r), \quad n_\varphi = 0, \quad n_z = \sin\chi(r)$$

$$f = \frac{1}{2} K \left[ (\text{div}\hat{\mathbf{n}})^2 + (\text{curl}\hat{\mathbf{n}})^2 \right]$$



(a)

$$\chi(r=R) = 0$$

$$\chi(r=0) = \pi/2$$

$$\text{div}\hat{\mathbf{n}} = \frac{1}{r} \frac{d(rn_r)}{dr} = -\sin\chi \frac{d\chi}{dr} + \frac{\cos\chi}{r} \quad \text{curl}_\varphi\hat{\mathbf{n}} = -\frac{dn_z}{dr} = -\cos\chi \frac{d\chi}{dr}$$

$$F_{\text{length}} = \frac{1}{2} K \int_{\varphi=0}^{\varphi=2\pi} d\varphi \int_{r=0}^{r=R} \left[ \left( \frac{\partial\chi}{\partial r} \right)^2 + \frac{\cos^2\chi}{r^2} - \frac{1}{r} \sin 2\chi \frac{d\chi}{dr} \right] r dr \quad r \rightarrow \exp \xi$$

$$F_{\text{length}} = \pi K \int_{r=0}^{r=R} \left[ \left( \frac{\partial\chi}{\partial \xi} \right)^2 + \cos^2\chi - \sin 2\chi \frac{d\chi}{d\xi} \right] d\xi$$

Euler-Lagrange equation

$$\frac{\partial^2 \chi}{\partial \xi^2} = -\cos\chi \sin\chi$$

$$\Rightarrow \left( \frac{\partial\chi}{\partial \xi} \right)^2 = \cos^2\chi + \text{const}; \quad \left. \frac{\partial\chi}{\partial \xi} \right|_{r \rightarrow 0} \rightarrow 0 \Rightarrow \text{const} = 0$$

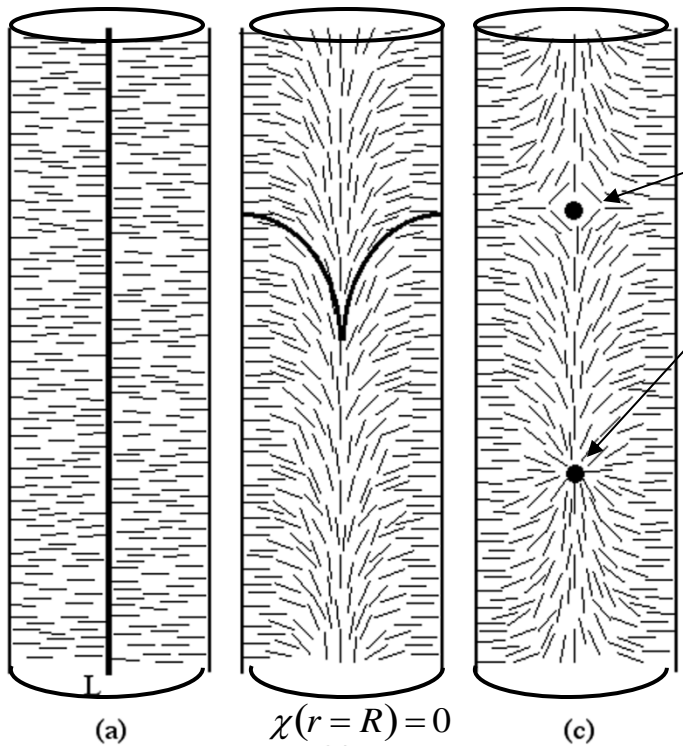
$$\frac{\partial\chi}{\partial \xi} = -\cos\chi \Rightarrow \int_r^R \frac{dy}{y} = -\int_\chi^0 \frac{dp}{\cos p} \quad \chi = 2 \arctan \left( \frac{R-r}{R+r} \right)$$

$$F_{\text{length}}^{\text{escaped}} = 3\pi K$$

$$F_{\text{length}}^{\text{singular}} = \pi K \ln \frac{R}{r_{\text{core}}} + F_{\text{core}}$$

Escape is preferred when  $R > 10r_{\text{core}}$

# Escape into the 3<sup>rd</sup> dimension



Point defects in the nematic bulk  
-hedgehogs

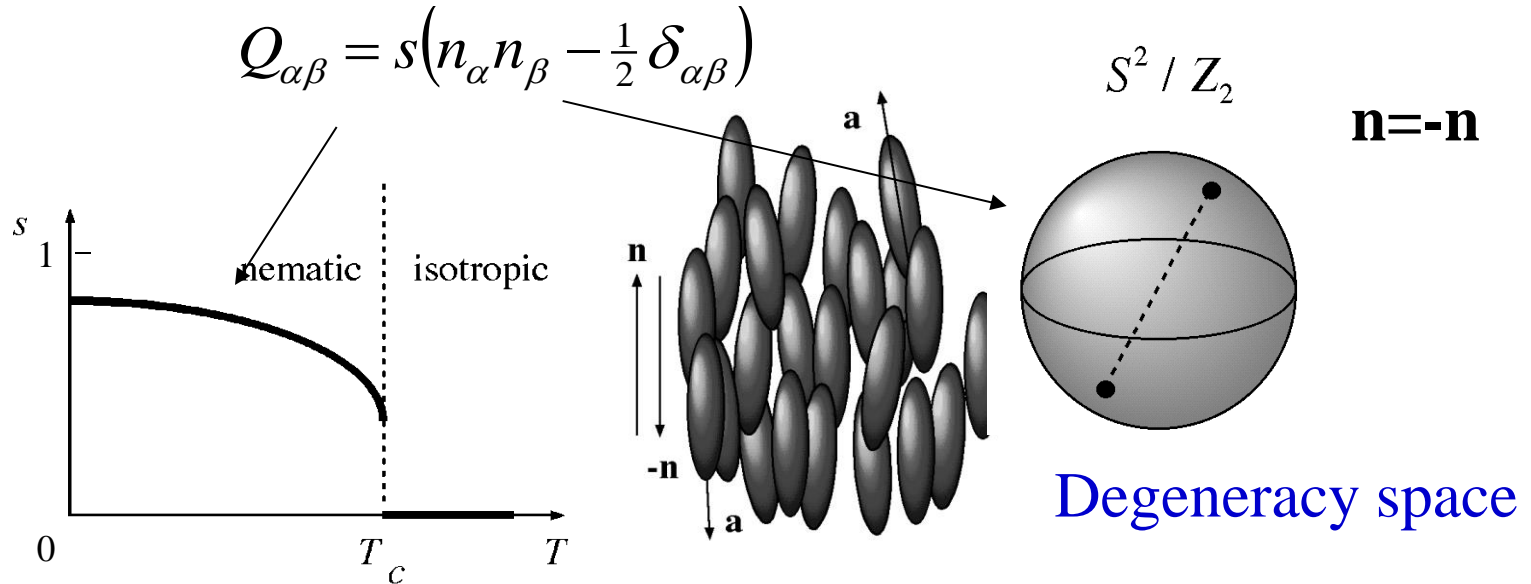
(a)

$$\chi(r=R)=0$$

(c)

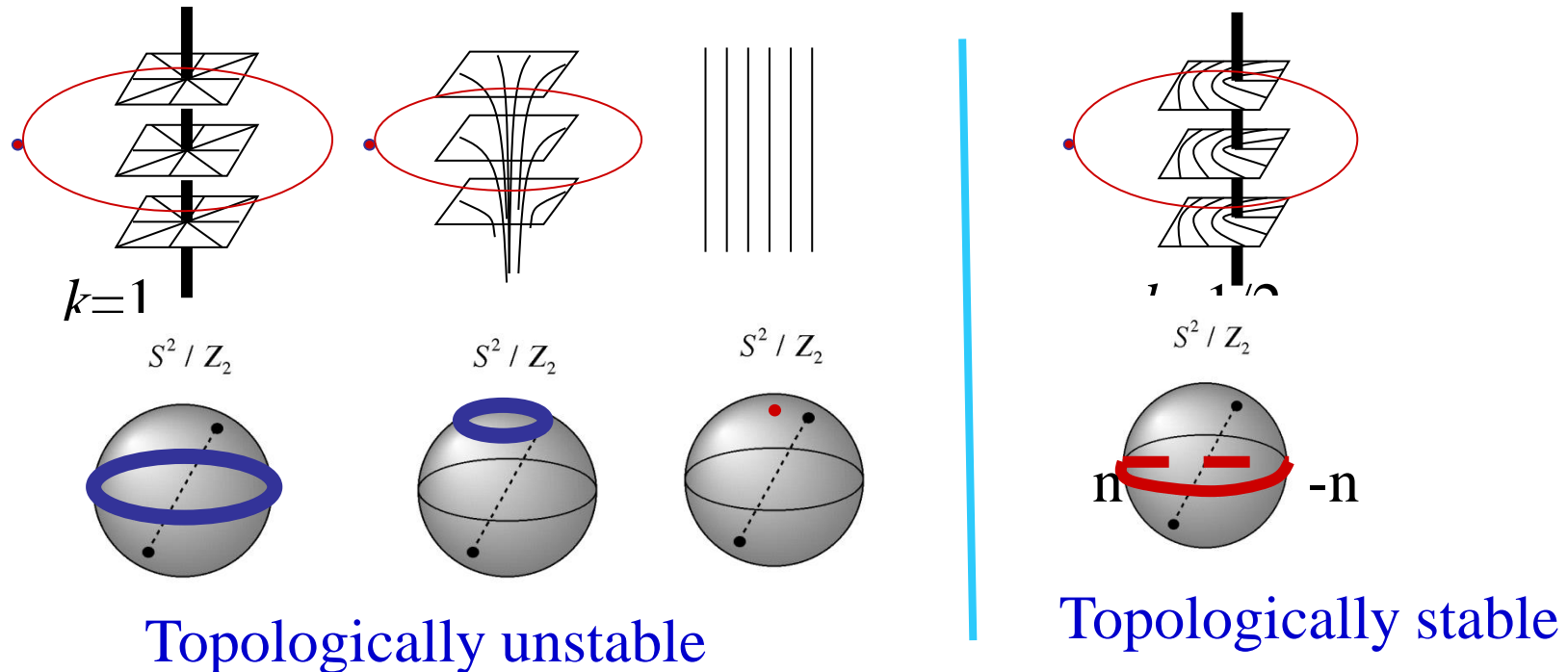
$$\chi(r=0)=\pi/2$$

# Homotopy classification: Uniaxial nematic $N_u$



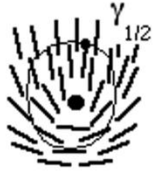
Topological stability is established by mappings from real space onto the degeneracy (or order parameter) space

# Homotopy classification: Lines in $N_u$



Topologically stable defect: A non-uniform configuration of the order parameter that cannot be reduced to a uniform state by a continuous transformation. In practical terms, to destroy a topological effect, one needs an energy exceeding the self energy of the defects by many orders of magnitude; e.g. melt the entire sample.

# Homotopy classification: Lines in $N_u$

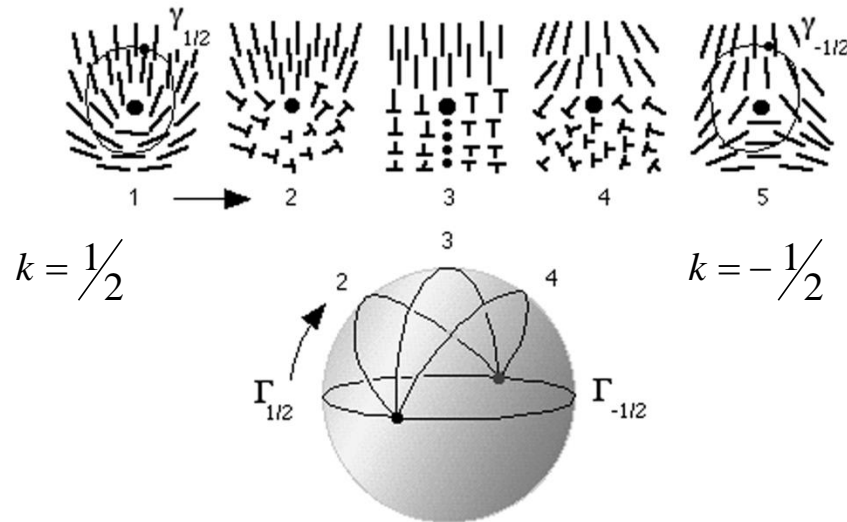


$$k = \frac{1}{2}$$



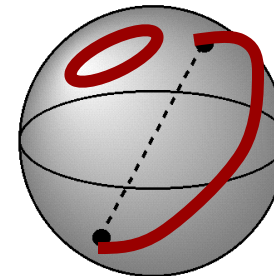
$$k = -\frac{1}{2}$$

# Homotopy classification: Lines in $N_u$



Disclinations in  $N$  are described by the 1<sup>st</sup> homotopy group, comprised of two elements

$$\pi_1(S^2 / Z_2) = Z_2 = (0, 1/2)$$



All semi-integer disclinations are topologically equivalent to each other and can be smoothly transformed one into another

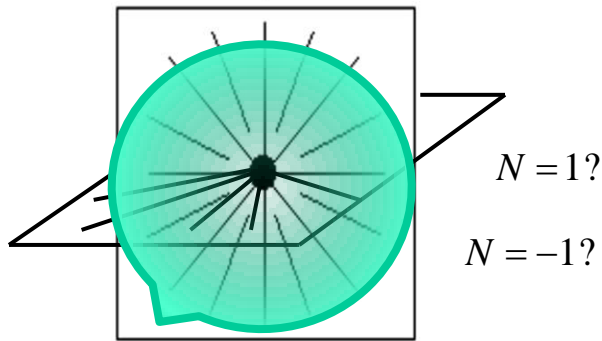


# Homotopy classification: Points in 3D $N_u$

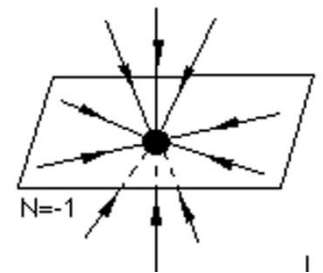
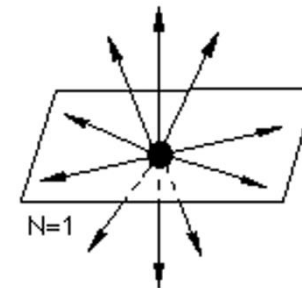
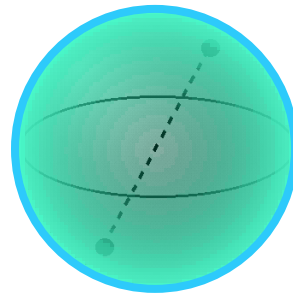
Hedgehogs (point defects) in uniaxial N

$$\pi_2(S^2 / Z_2) = N = 0, \pm 1, \pm 2, \dots$$

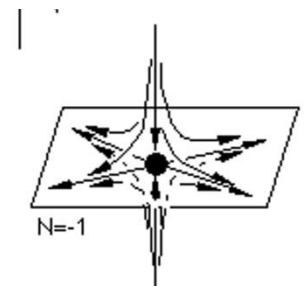
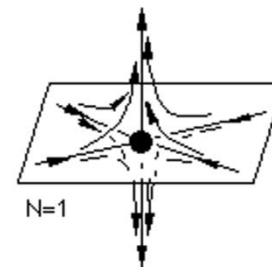
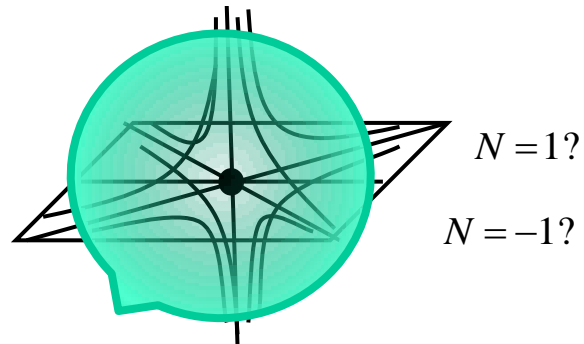
Radial hedgehog



$S^2 / Z_2$



Hyperbolic hedgehog



# Topological charges of points, vector fields, t-space

t-dimensional vector field:

$$\mathbf{n} = (n^1, n^2, \dots, n^t)$$

(t-1)-coordinates  
specified on the sphere  
around the defect

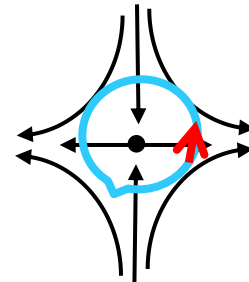
$$(u^1, u^2, \dots, u^{t-1})$$

$$N^{(t)} = \frac{1}{\Omega} \int_{S^{t-1}} \begin{vmatrix} n^1 & \dots & \dots & n^t \\ \frac{\partial n^1}{\partial u^1} & \dots & \dots & \frac{\partial n^t}{\partial u^1} \\ \dots & \dots & \dots & \dots \\ \frac{\partial n^1}{\partial u^{t-1}} & \dots & \dots & \frac{\partial n^t}{\partial u^{t-1}} \end{vmatrix} du^1 \dots du^{t-1}$$

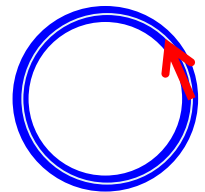
Definition of t-dimensional topological charge:  
E. Dubrovin et al, Modern Geometry, Springer, 1984

Point defects in 2D:

$$N^{(2)} \equiv k = \frac{1}{2\pi} \int_{loop} \left( n^1 \frac{dn^2}{dl} - n^2 \frac{dn^1}{dl} \right) dl = \pm 1, \pm 2, \dots$$

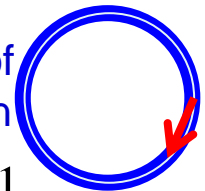


$k = 1$

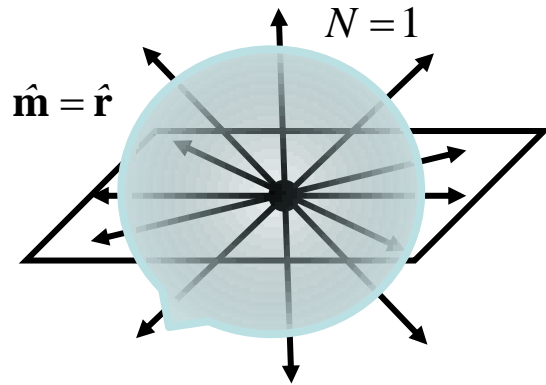


Circle of all  
possible  
orientations of  
magnetization

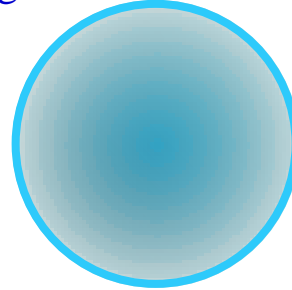
$k = -1$



# Point defects in 3D: Ferromagnetic vs Nematic



Sphere of all possible orientations of magnetization



Topologically charge: how many times the magnetization vector goes through all possible orientations

$$N^{(3)} = \frac{1}{4\pi} \int_{\sigma} \hat{\mathbf{m}} \left[ \frac{\partial \hat{\mathbf{m}}}{\partial u} \times \frac{\partial \hat{\mathbf{m}}}{\partial v} \right] dudv$$

Topologically stable point defect; to remove it, one needs to destroy ferromagnetic order on the entire line

M. Kleman, Phil. Mag. **27** 1057 (1973).

N. Mermin et al PRL 36, 594 (1976)

$$\hat{\mathbf{m}}(u, v) = \{ \sin \theta(u, v) \cos \varphi(u, v); \sin \theta(u, v) \sin \varphi(u, v); \cos \theta(u, v) \}$$

$$N^{(3)} = \frac{1}{4\pi} \int_{\sigma} \left( \frac{\partial \theta}{\partial u} \frac{\partial \varphi}{\partial v} - \frac{\partial \theta}{\partial v} \frac{\partial \varphi}{\partial u} \right) \sin \theta dudv \quad \hat{\mathbf{m}} = \hat{\mathbf{r}} \quad N = 1$$

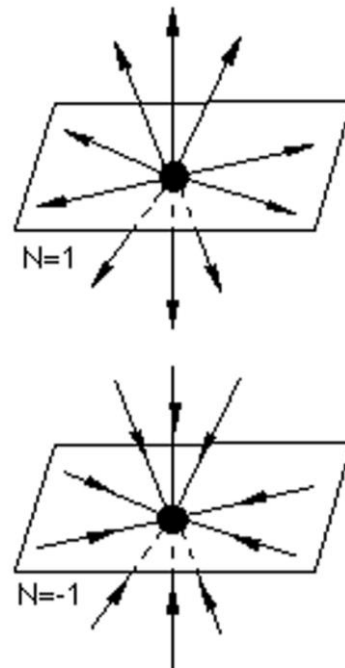
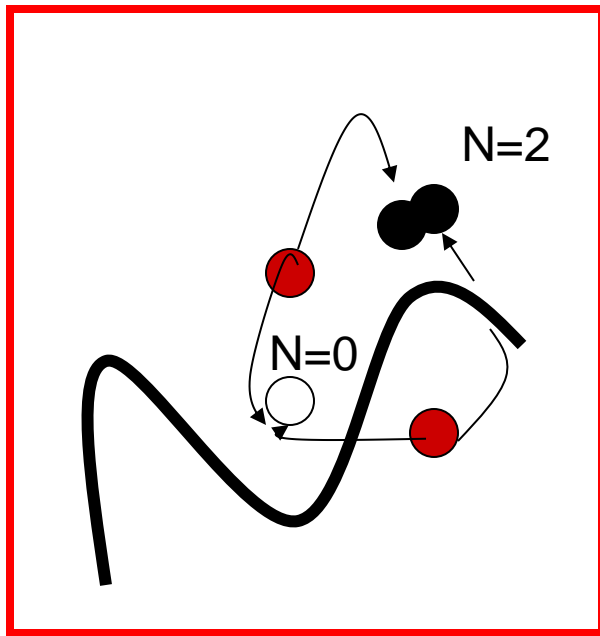
Uniaxial nematic

$$N^{(3)} = \frac{1}{4\pi} \int_{\sigma} \hat{\mathbf{n}} \left[ \frac{\partial \hat{\mathbf{n}}}{\partial u} \times \frac{\partial \hat{\mathbf{n}}}{\partial v} \right] dudv = \pm 1$$

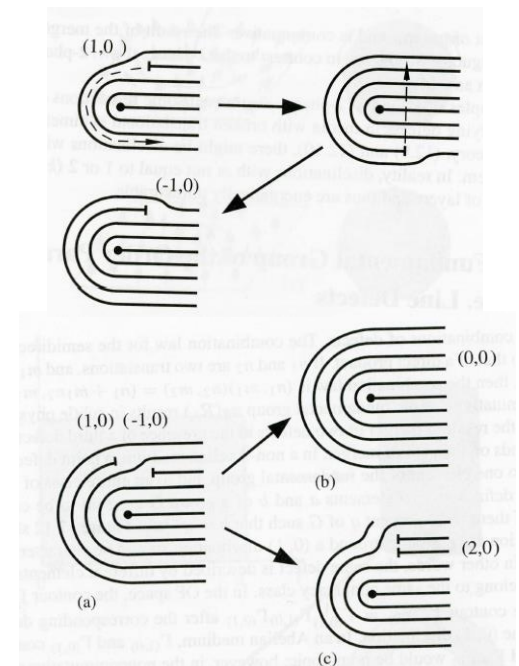
# Homotopy: Points in 3D $N_u$

Result of merger of 2 hedgehogs in a uniaxial  $N$  in presence of a disclination depends on the pathway of merger

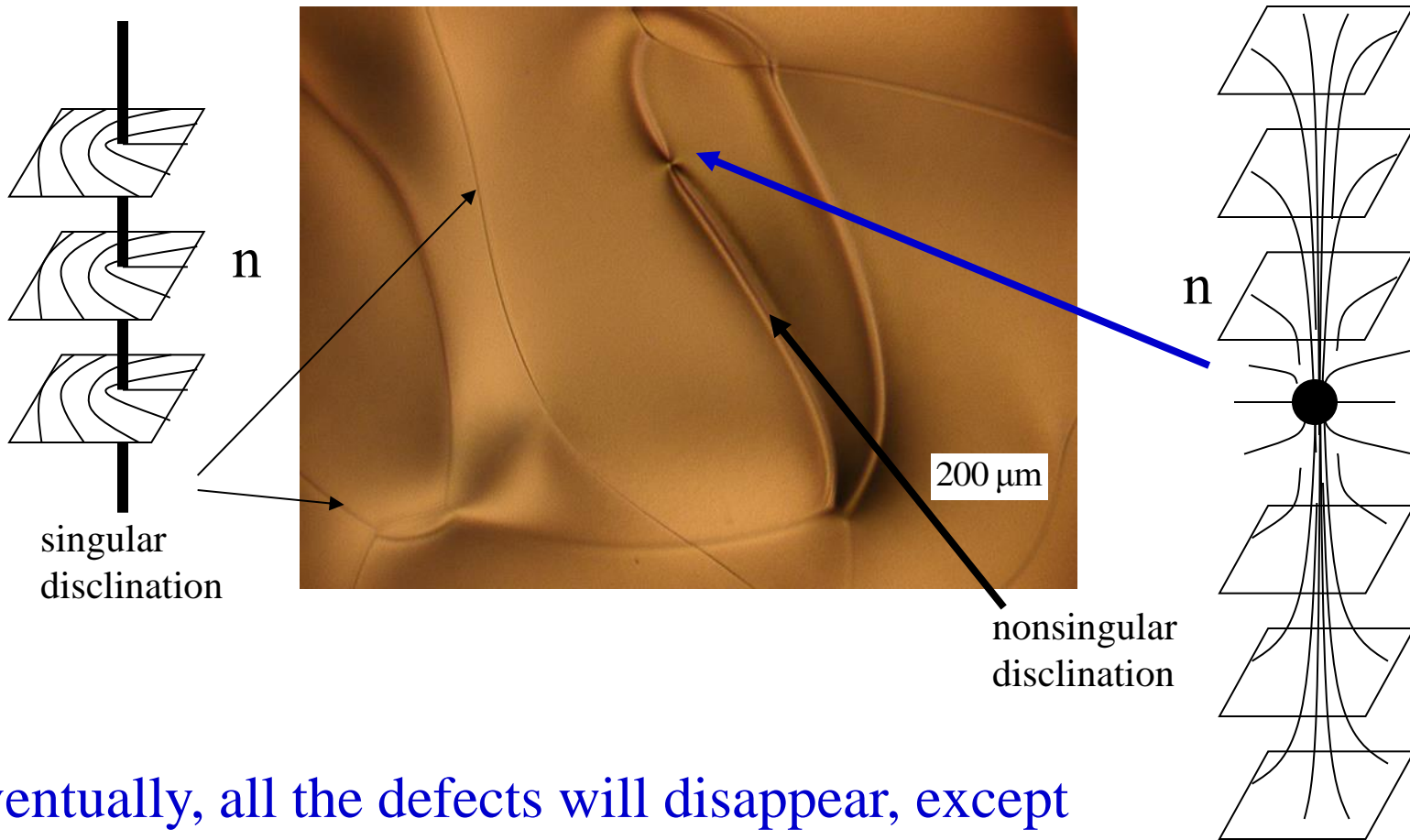
$$N + N = \begin{cases} 2N \\ 0 \end{cases}$$



Similar example for dislocations in presence of disclinations:



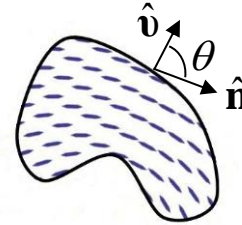
# Typical texture of a (thick) $N_u$



Eventually, all the defects will disappear, except maybe one line and one point defect, mostly through annihilation, as  $\frac{1}{2} + \frac{1}{2} = 0$  and  $1 + 1 = 0$ !

# Defects in equilibrium: LC droplets

The structure is determined by the balance of anisotropic surface tension and internal elasticity



□ Anisotropic surface energy  $F_{surface} = \sigma_o + f(\hat{\mathbf{n}} \cdot \hat{\mathbf{v}})$   
 $f(\hat{\mathbf{n}} \cdot \hat{\mathbf{v}}) = W_2 (\hat{\mathbf{n}} \cdot \hat{\mathbf{v}})^2 = W_2 \cos^2 \theta$  Rapini-Papoular surface anchoring potential

S. Faetti et al, PRA **30**, 3241 (1984): N-I thermotropic interface is weakly anisotropic:

$$\sigma_o \sim 10^{-5} \text{ J/m}^2; W_2 \sim 10^{-6} \text{ J/m}^2; W_2 / \sigma_o \sim 0.1 - 0.01$$

□ Elasticity:

$$F_{elastic} \sim KR \quad K_i \sim 5 \text{ pN} \quad R \geq 10 \text{ } \mu\text{m} \Rightarrow$$

$$\frac{\sigma_o R^2}{KR} \geq 10; \quad \frac{W_2 R^2}{KR} \geq 1$$

The droplets of thermotropic N in isotropic melt are spherical and contain defects to satisfy surface anchoring conditions

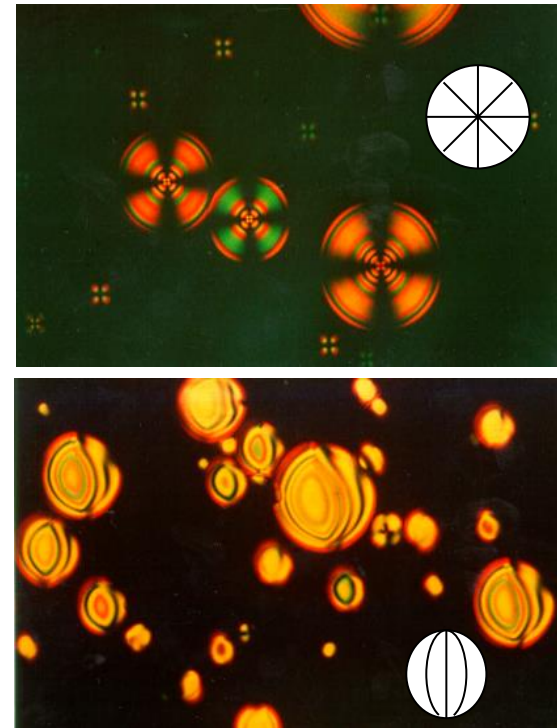
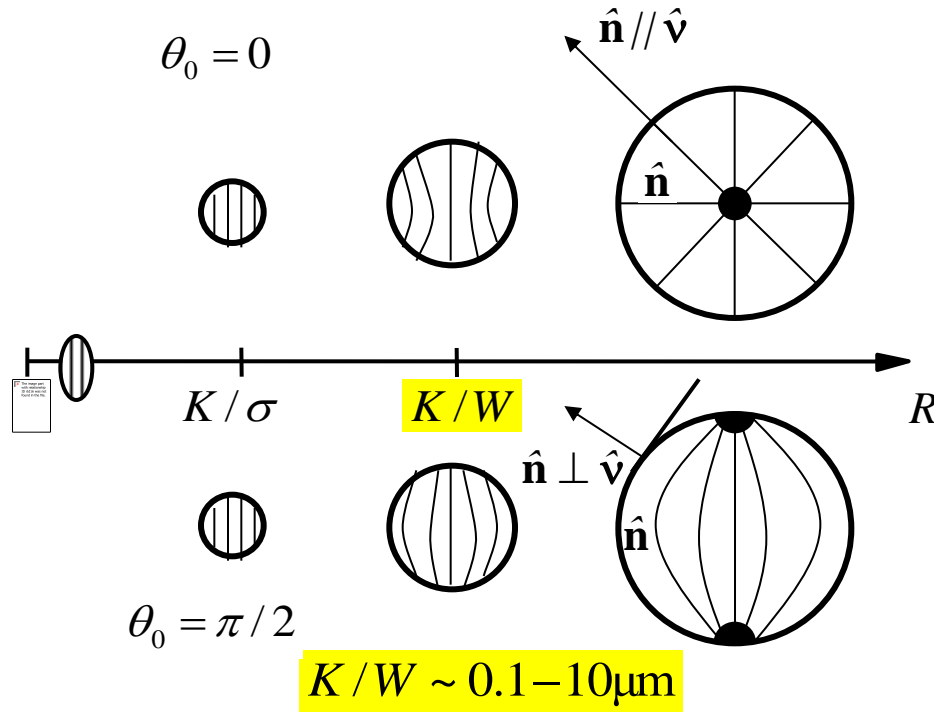


# Defects in equilibrium: LC droplets

Balance of elasticity and surface anchoring

$$F_{elastic} \sim KR \quad F_{anchoring} \sim WR^2$$

leads to the following expectation for scaling behavior:



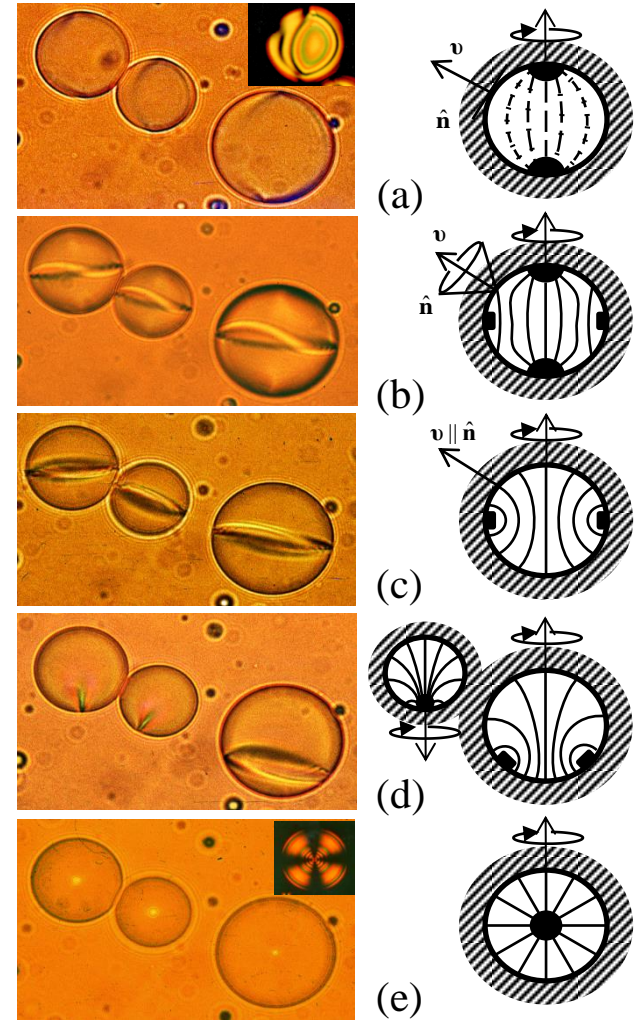
Defects correspond to the equilibrium state of the (large) system

# Defects in equilibrium: LC droplets

N droplets in glycerine with temperature varied anchoring axis; topological dynamics of boojums, disclination loops and hedgehogs

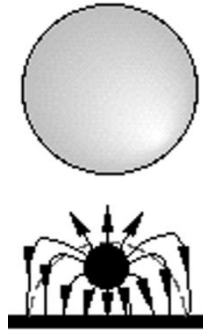
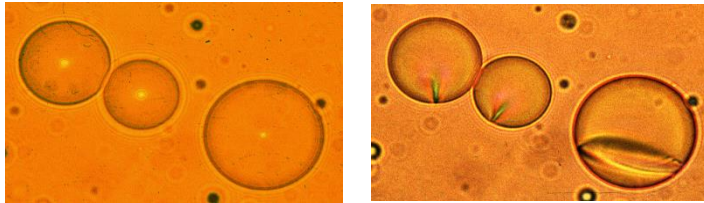
NB: Defects correspond to the equilibrium state of the system; they help to minimize the sum of the anisotropic surface tension and bulk energy.

Do we really want to minimize the energy for each and every surface angle?!

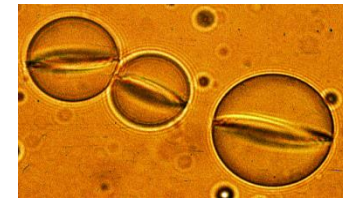


# Topological dynamics of defects in LC drops

Bulk point defect hedgehog:  $N = 1$



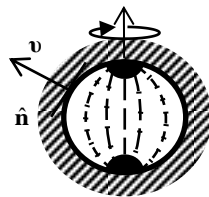
Hedgehog spreadable into a disclination loop:



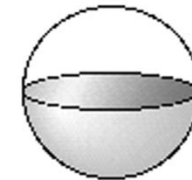
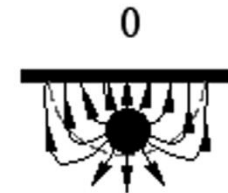
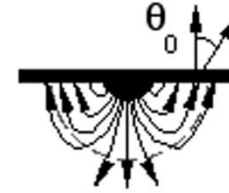
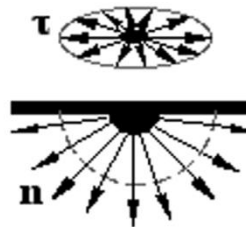
$N = 1$

Replace director with a vector

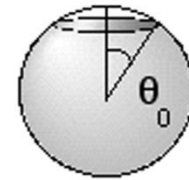
Surface point defect boojum: two topological charges, 2D and 3D;



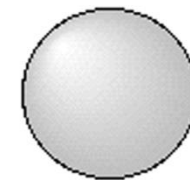
$k = 1$



$C = 1/2$



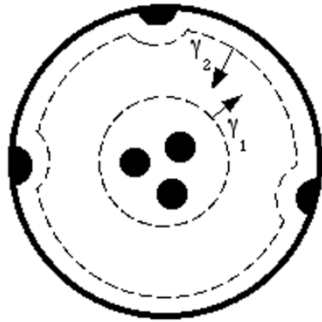
$C = \cos^2 \frac{\theta_0}{2}$



$C = 1$

$$C_{k,N} = \frac{1}{4\pi} \int_{\sigma} d\theta d\phi \mathbf{n} \left[ \frac{\partial \mathbf{n}}{\partial \theta} \times \frac{\partial \mathbf{n}}{\partial \phi} \right] = \frac{k}{2} (\mathbf{n} \cdot \mathbf{v} - 1) + N$$

# Topological dynamics of defects in LC drops



$$\sum_{i=1}^b C_{k_i, N_i} + C_s + \sum_{j=b+1}^{h+b} N_j = \left( -1 + \frac{1}{2} \sum_{i=1}^b k_i \right) (\mathbf{n} \cdot \mathbf{v} - 1) + \sum_{j=b+1}^{h+b} N_j - 1 = 0$$

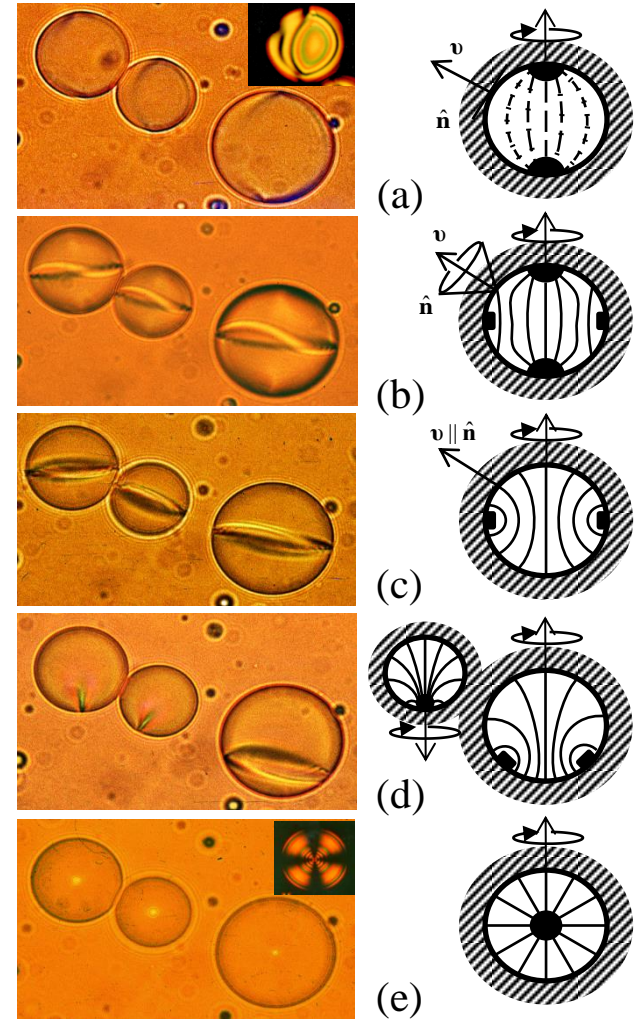
Poincare theorem: conservation law for vector fields tangential to the surface

$$\sum_i^b k_i = E$$

Gauss theorem: conservation law for vector fields perpendicular to the surface

$$\sum_{j=1}^{h+b} N_j = E / 2$$

Euler characteristic for sphere  $E = 2$

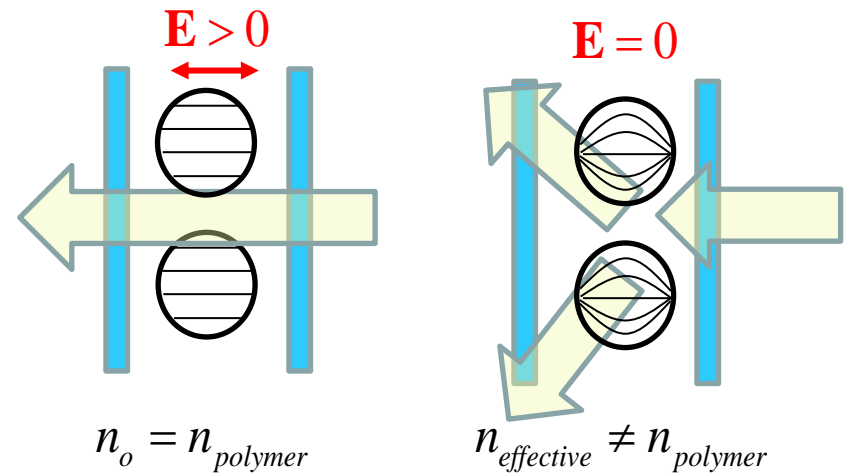




# Applications of LC drops



Privacy windows:  
Polymer Dispersed Liquid Crystals  
(JW Doane et al, LCI, Kent)

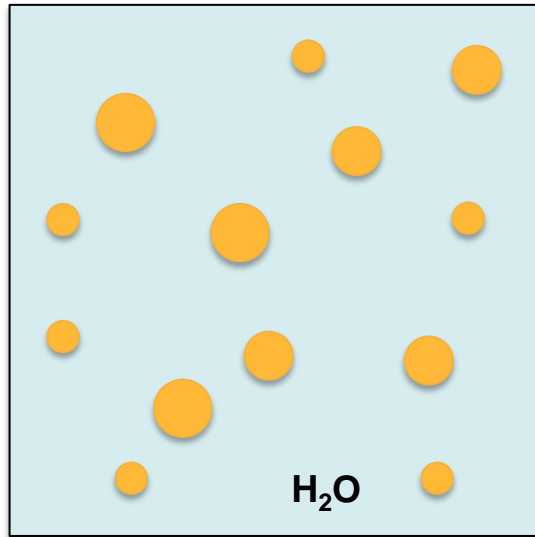


# Droplets as Biosensors:

Abbott and Lynn (UW-Madison)

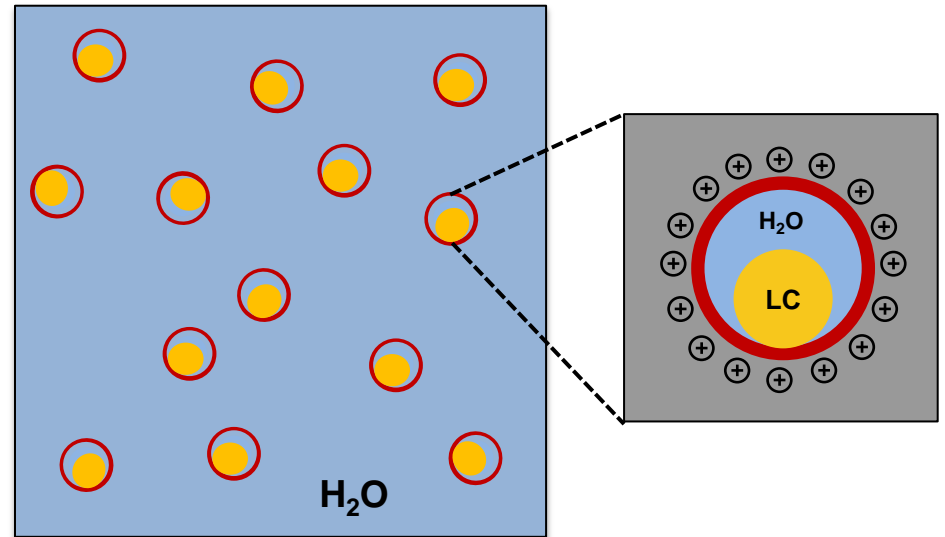
## Microscale LC Droplets

(Oil-in-Water Emulsion)



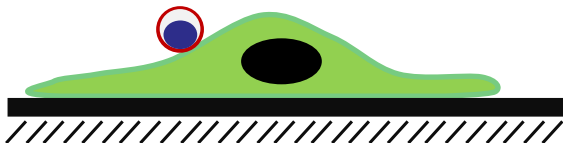
## 'Caged' LC Droplets

(Droplets in Polymer Capsules)

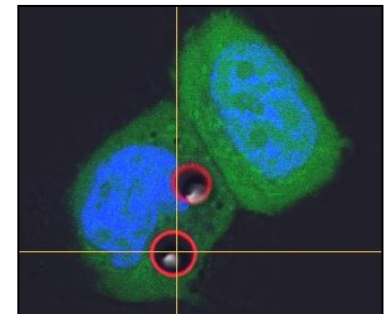
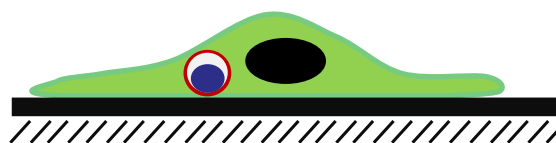


## Sensing in Biological Environments:

Immobilized LC Droplets



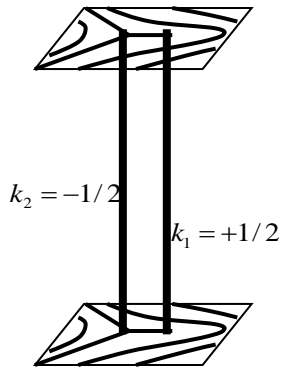
Internalized LC Droplets







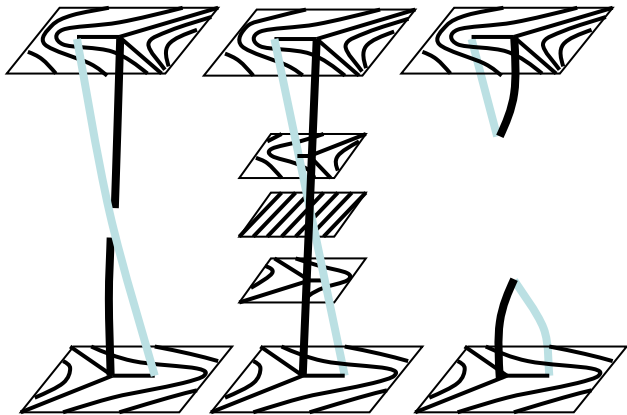
# Reconnection of disclinations in $N_u$



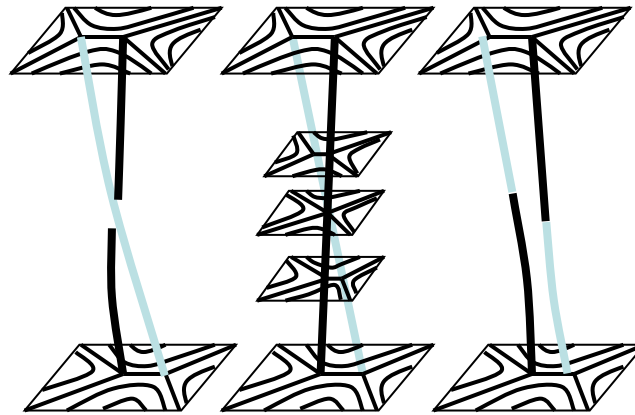
Disclinations are not “material” lines and can cross each other.

Two disclinations connecting opposite plates of a nematic cell; plates are twisted; disclination ends reconnect

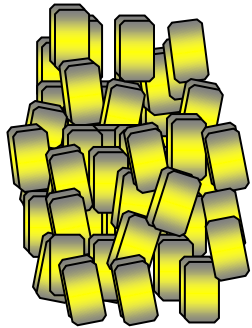
$k_1 = +1/2$   $k_2 = -1/2$



$k_1 = -1/2$   $k_2 = -1/2$



# Reconnection of disclinations in biaxial $N_{bx}$

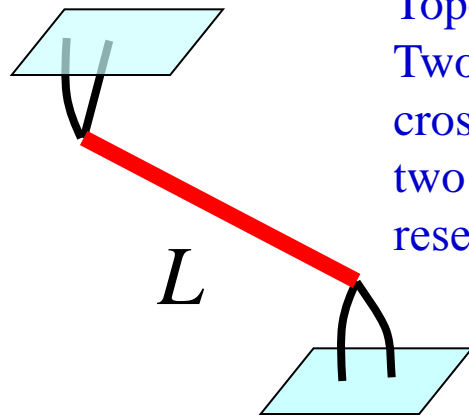
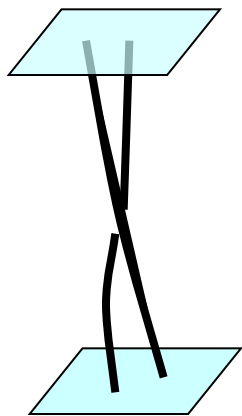


Degeneracy space: Solid sphere; each point describes a state of three directors,  $n, m, l$ . In biaxial nematics, the strength 1 disclinations cannot escape (strength 2 can). There are three different classes of  $1/2$  disclinations with  $k$  semi-integer and one with  $k=1$

$k = 1$  does not escape!



$$\pi_1(S^3 / D_2) = \text{Quaternion units} = (0; 1; 1/2_x; 1/2_y; 1/2_z)$$

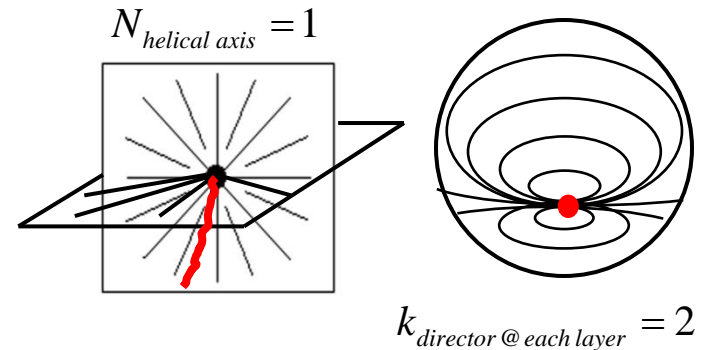


Topological test for biaxiality of a nematic:  
Two disclinations  $1/2$  belonging to different classes cannot cross each other, the trace is a third line of strength 1; the two separated disclinations interact through a potential that resembles interaction of ... Quarks!

$$U \propto KL$$

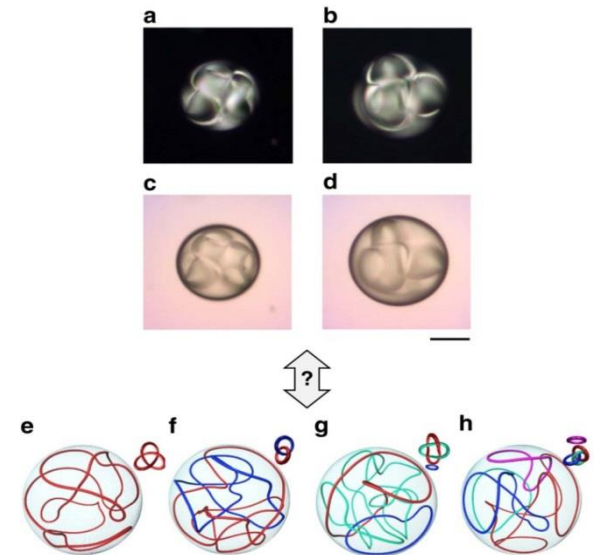
# Cholesteric drops and Dirac monopole

Two orthogonal vector fields: helical axis and director (magnetic field and vector-potential)



Point hedgehog in helical axis (magnetic) field and an attached disclination (Dirac string) in the director field

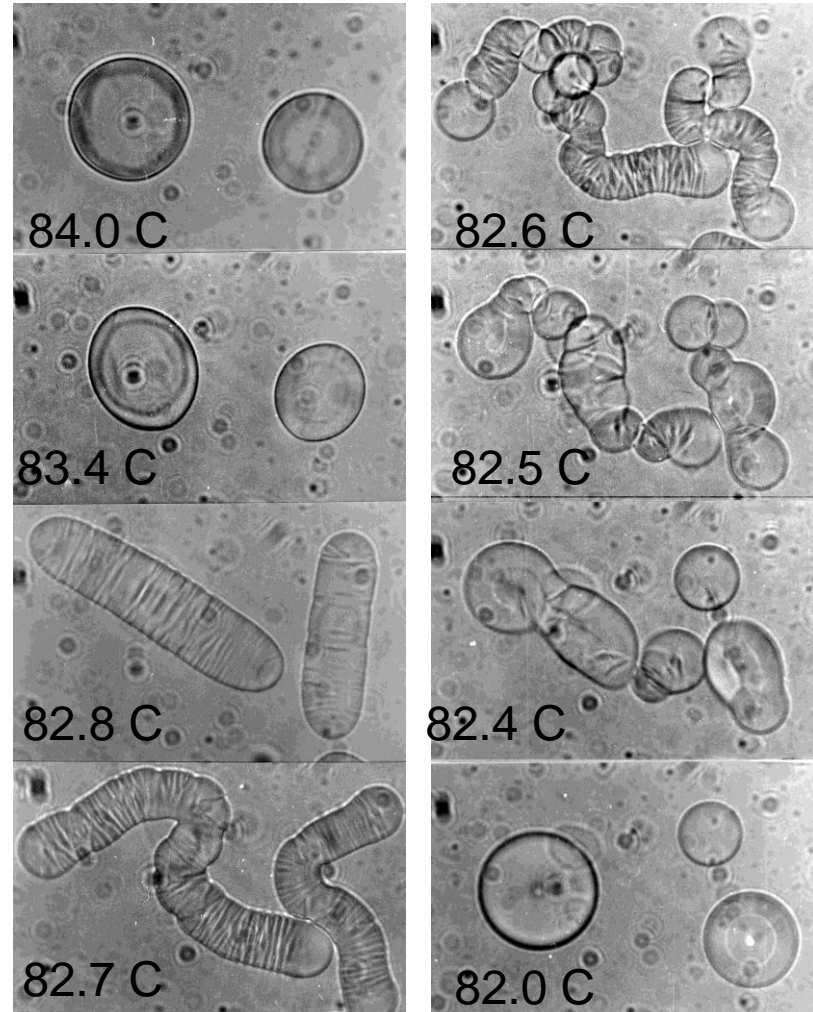
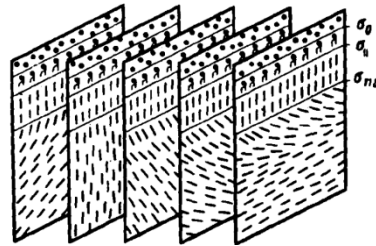
P. Dirac, Proc. Roy. Soc. (London) A133, 60 (1931)  
 Ch droplet, called Robinson spherulite or Frank-Price structure,  
 C. Robinson et al, Disc. Faraday Soc. 25, 29 (1958);  
 Kurik et al, JETP Lett **35**, 444 (1982)



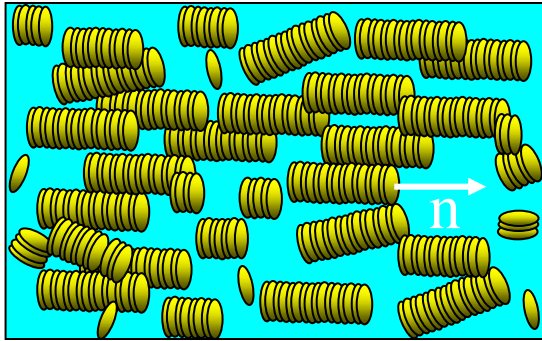
T. Orlova et al, Nat. Comm. **6**, 7603 (2015)

# Rare exception of round cohesive droplets: Ch-SmA phase transition

Ch droplets in glycerine+lecithin; cooling down leads to extended shapes, then nucleation of spherical SmA sites; the process often results in division of droplets (Nastishin et al Sov Phys JETP Lett 1984; EurophysLett 1990)



# (LC)<sup>2</sup>: Lyotropic Chromonic Liquid Crystals



$$\sigma_o \sim \alpha \frac{k_B T}{LD} \sim (10^{-7} - 10^{-6}) \text{ J/m}^2$$

$$W \sim 10^{-5} \text{ J/m}^2$$

Model of surface tension: P. van der Schoot *J. Phys. Chem B* **103**, 8804 (1999)

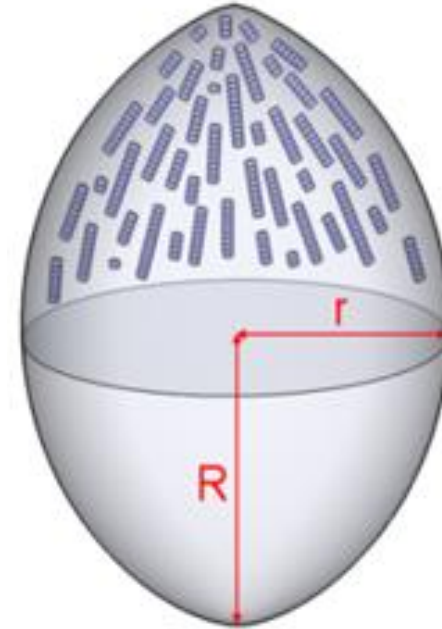
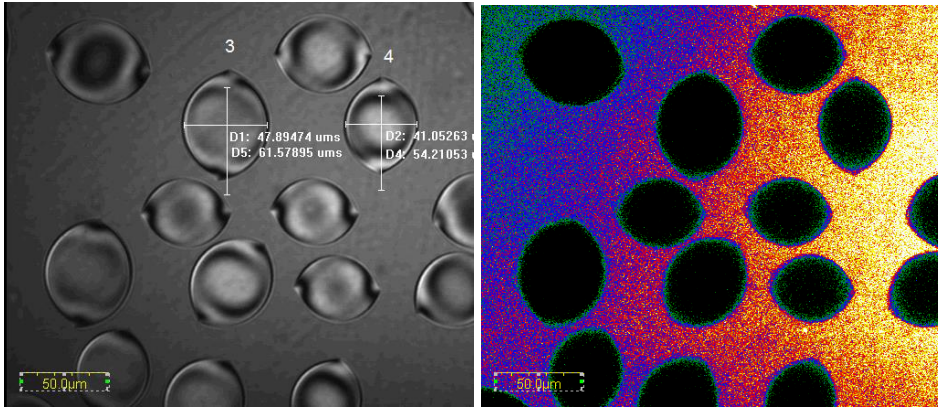
Lyotropic I-N interface might be strongly anisotropic and be influenced by elasticity



J. Bernal and I. Fankuchen, *J. Gen. Physiol.* (1941): tactoids as N nuclei in tobacco mosaic virus dispersions

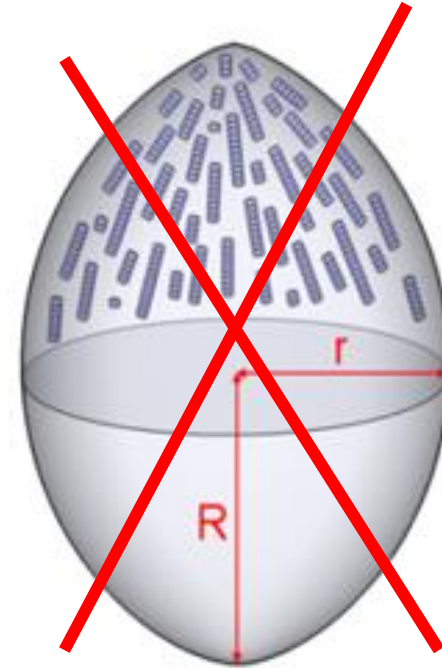
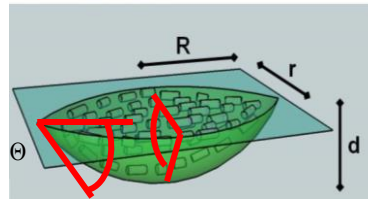
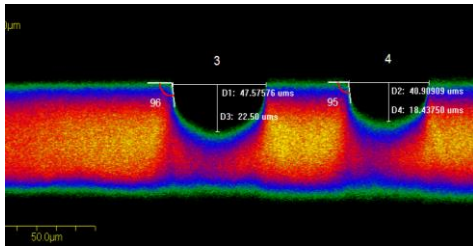
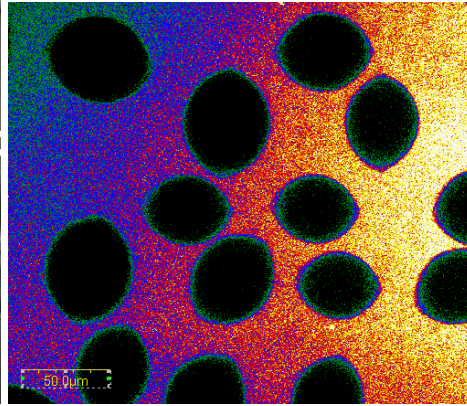
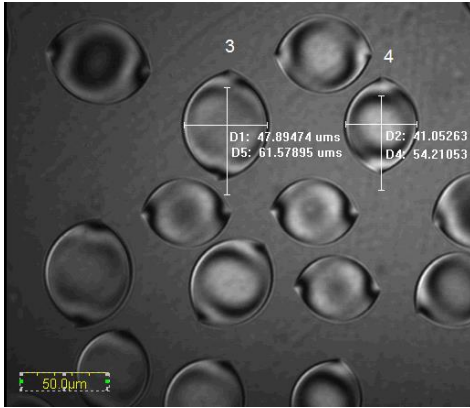


# Droplets of chromonic N in isotropic melt (tactoids)



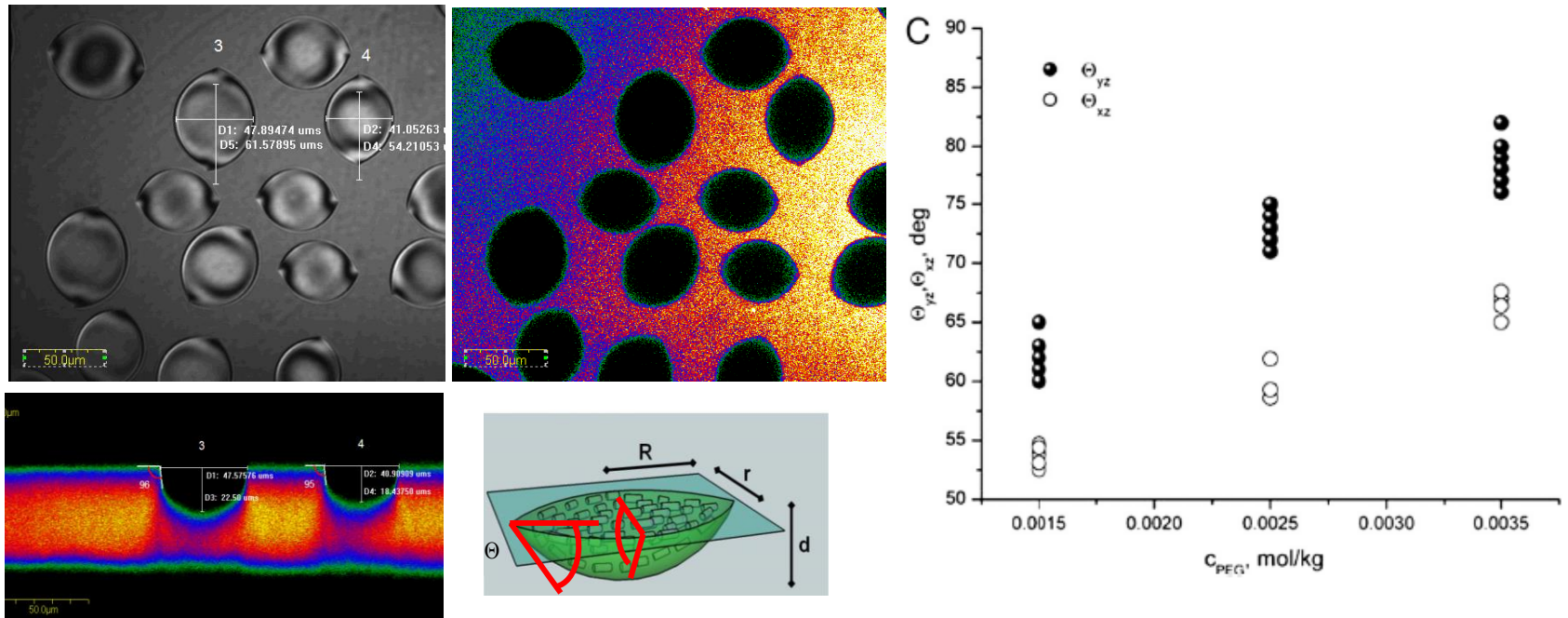


# Surprise #1: Surface-located, not bulk



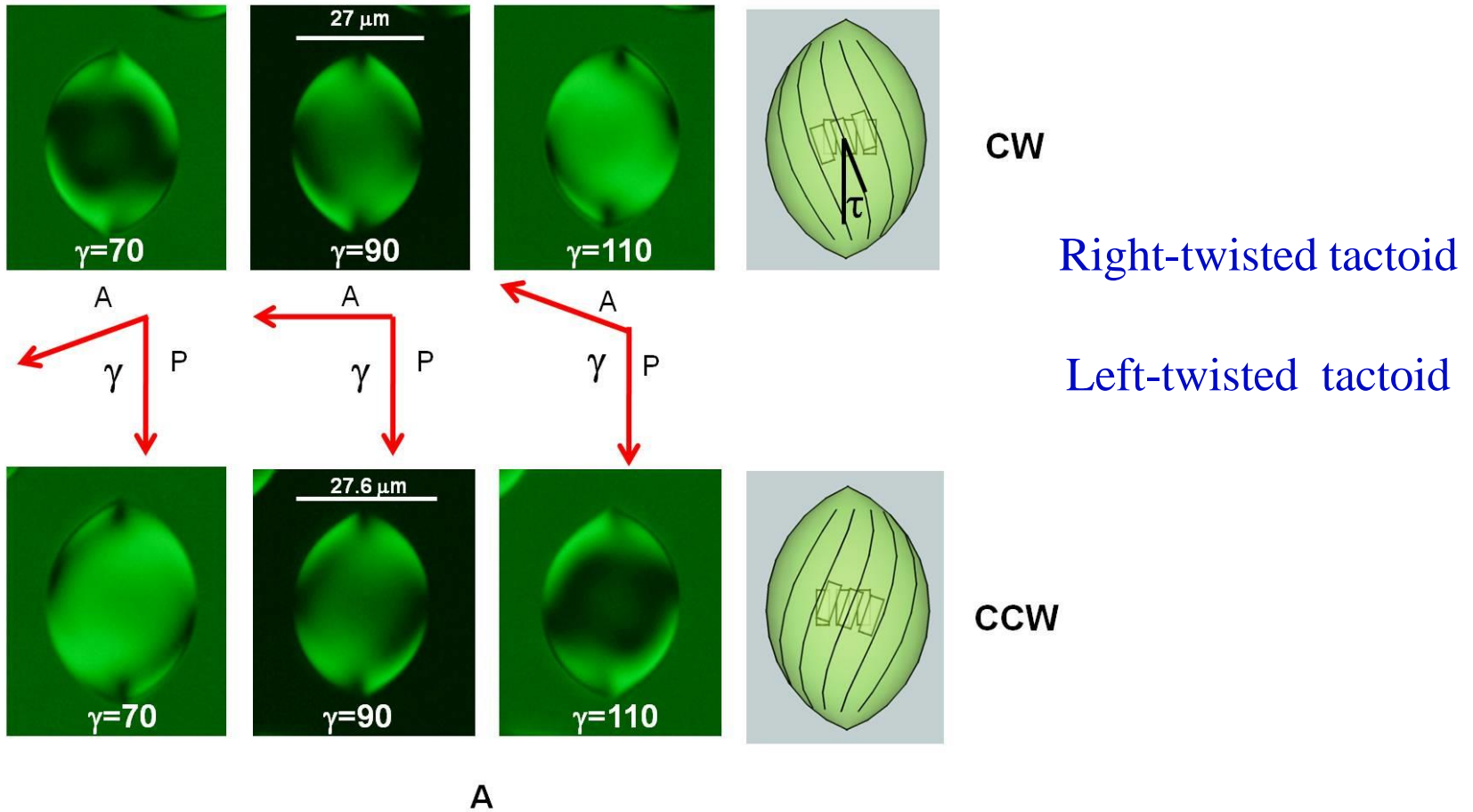
Vertical cross-section image;  
fluorescent confocal  
microscopy

# Surprise #2 (mild): Contact angle changes along the perimeter

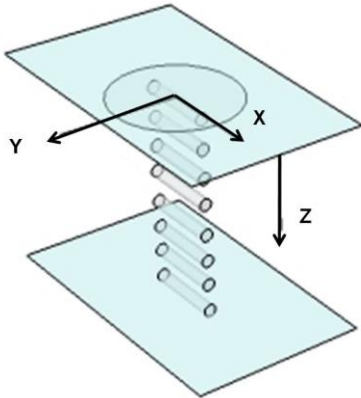


Vertical cross-section image;  
fluorescent confocal  
microscopy

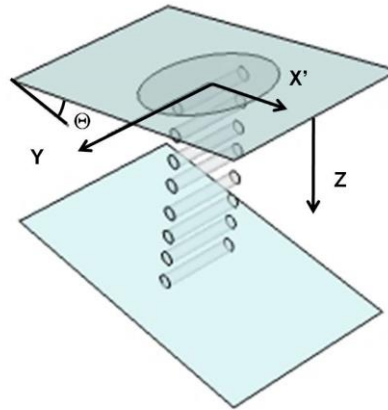
# Surprise #3: Twist



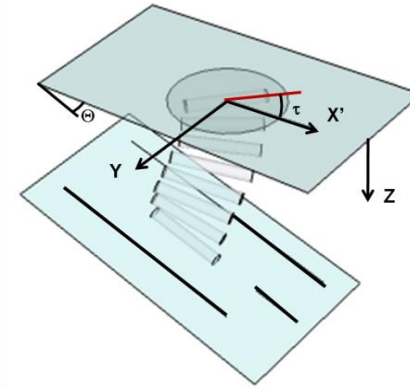
# Mechanism: “Geometrical” anchoring+large $K_1/K_2$



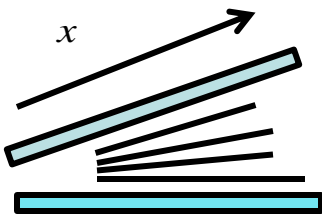
Nematic between two isotropic parallel boundaries:  
Degenerate in-plane orientation



One plate tilts: alignment perpendicular to the thickness gradient is the only one without distortions; other directions cause splay



One plate tilts, the other sets “physical anchoring” say, along the long axis of the tactoid’s footprint: balance of twist and splay establishes a twist angle  $\tau$



# Mechanism: “Geometrical” anchoring+large $K_1/K_2$

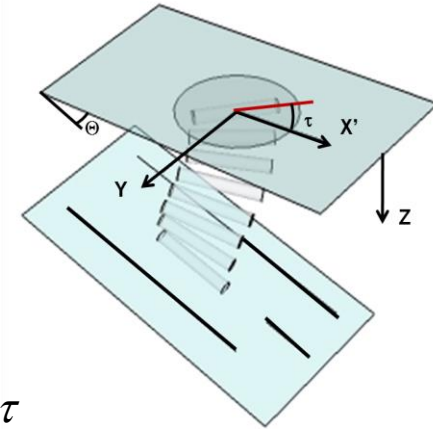
Elastic energy of a tilted element:

$$f \approx K_1 \left( \frac{\partial \theta}{\partial z} \right)^2 + K_2 \left( \frac{\partial \varphi}{\partial z} \right)^2$$

Bulk equilibrium:

$$\frac{\partial^2 \theta}{\partial z^2} = 0; \frac{\partial^2 \varphi}{\partial z^2} = 0$$

$$\theta(z=d) = -\arcsin(\sin \Theta \cos \tau) \approx \Theta(1 - \tau^2/2); \varphi(z=d) \approx \tau$$



Twist  
deformations  
reduce the cost  
of splay  
deformations

$$F \approx \frac{K_1}{2d} \Theta^2 (1 - \tau^2) + \frac{K_2}{2d} \tau^2$$

Condition for the twist:  $K_2 / K_1 < \Theta^2$

Easy to fulfill as in  
chromonics,  $K_2 / K_1 \sim 0.1 - 0.03$

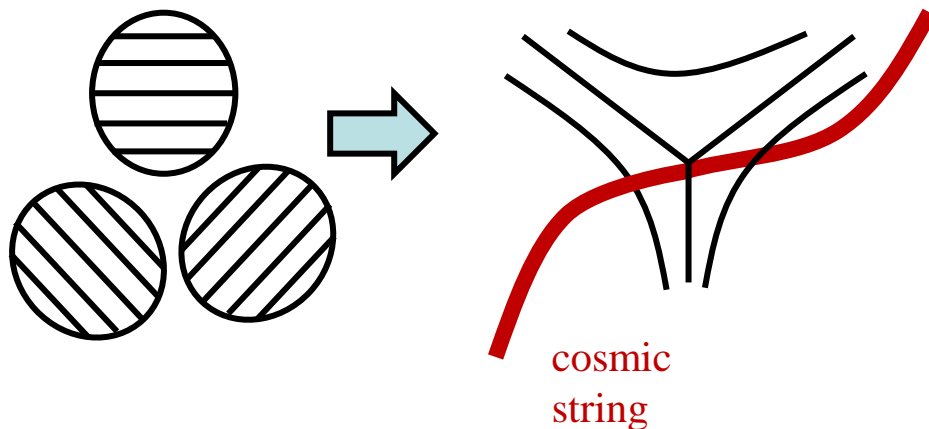
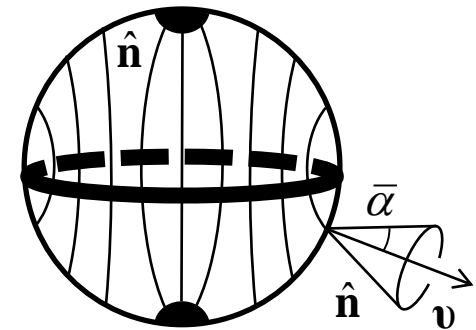
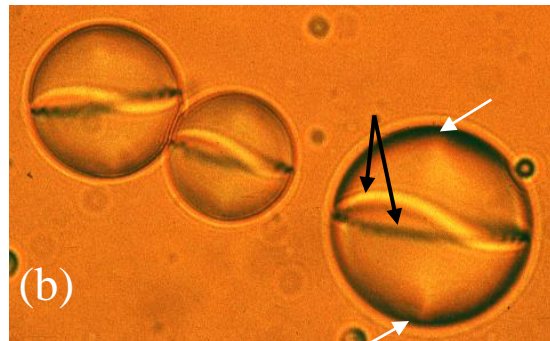
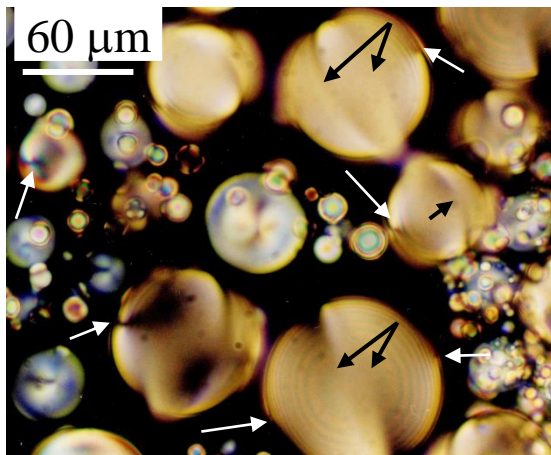
S. Zhou et al., *Soft Matter* **10**, 6571 (2014)

Twisted tactoids: An example of chiral symmetry breaking in a molecularly non-chiral system; only spatial confinement and elastic anisotropy are needed to produce macroscopic chiral purity.



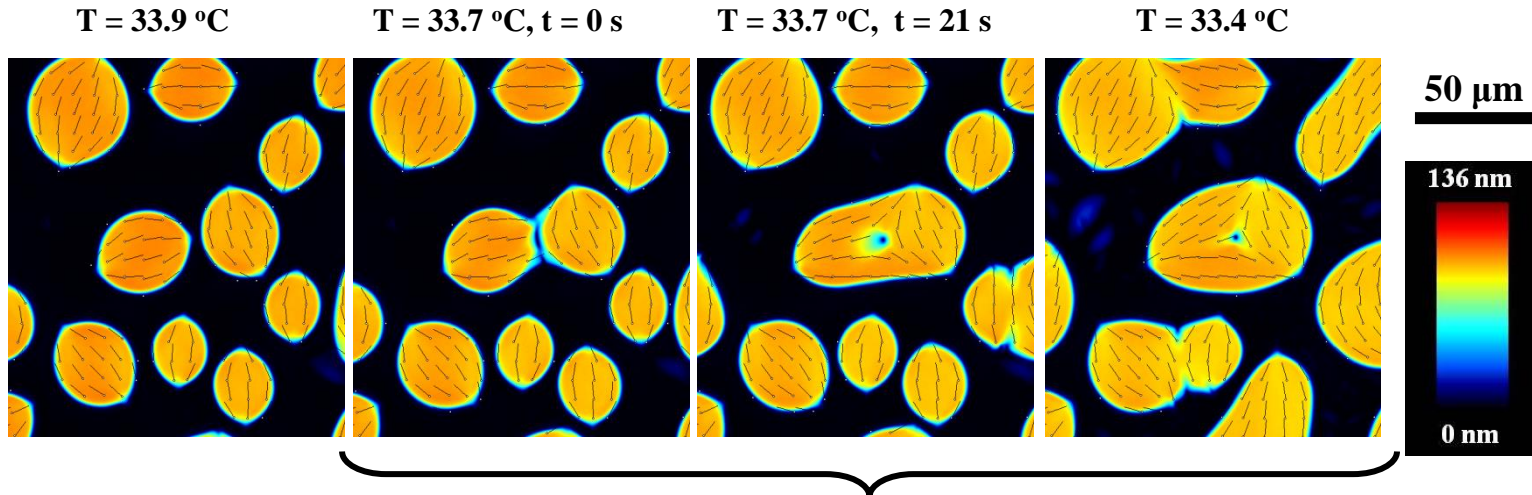
# 2D tactoids and Kibble mechanism

Isotropic-Nematic transition: Anchoring-induced topological defects in each and every nuclei of the N phase, as long as it is large enough,  $R > K / W$



Kibble (1976) Model of formation of cosmic domains and strings

# 2D tactoids and Kibble mechanism in (LC)<sup>2</sup>

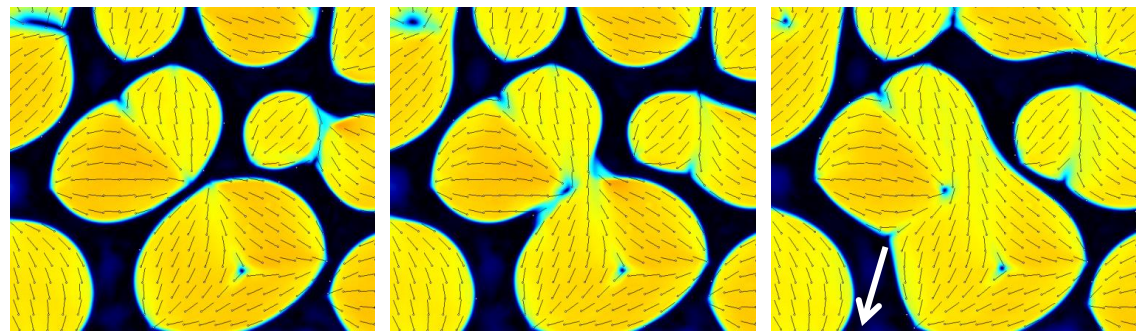


Anchoring-produced surface defects-booiums

Kibble-like production of disclinations as a result of tactoids merger

Conservation law for positive and negative cusps:

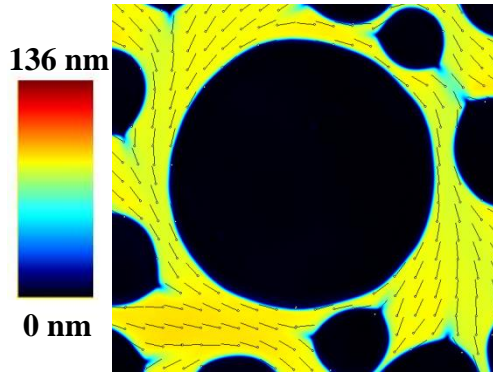
$$c^+ - c^- = 2 \left( 1 - \sum_k^n m_k \right)$$



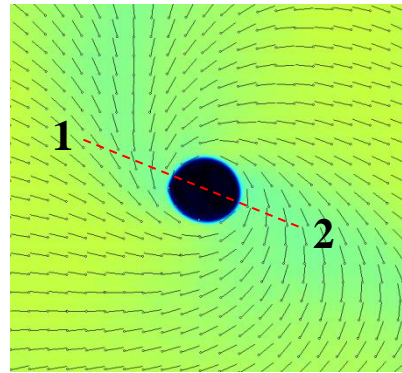
negative cusp



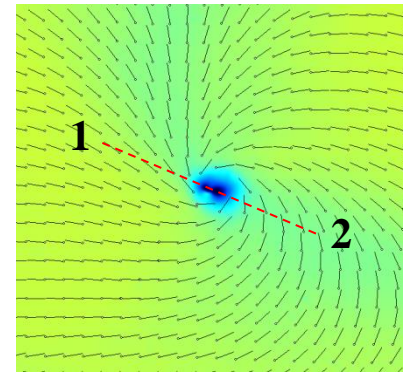
# 2D tactoids and k=1 disclinations



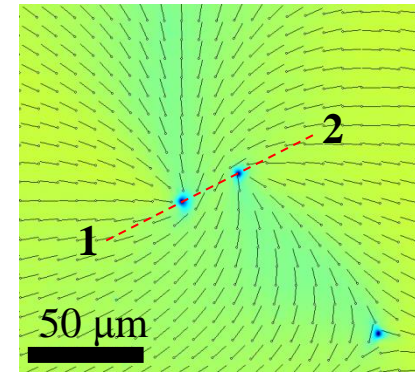
(a) T = 32.7 °C



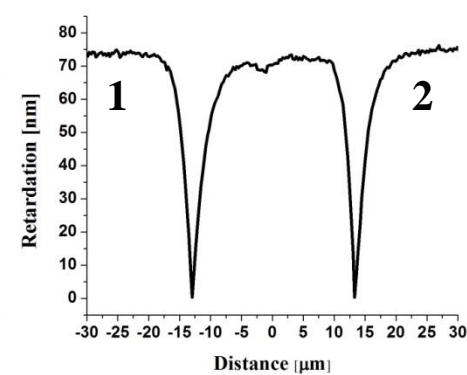
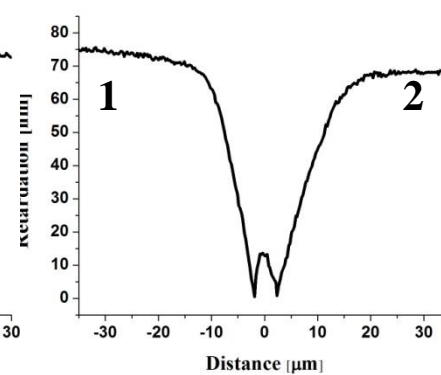
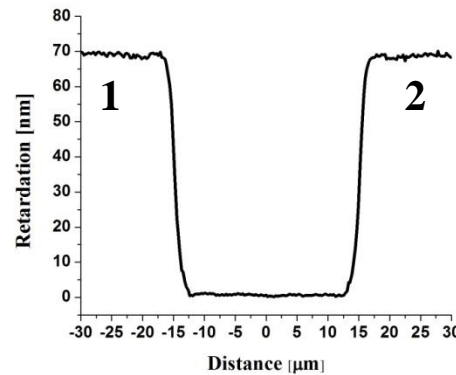
(b) T = 29.8 °C



(c) T = 29.7 °C, t = 0 s



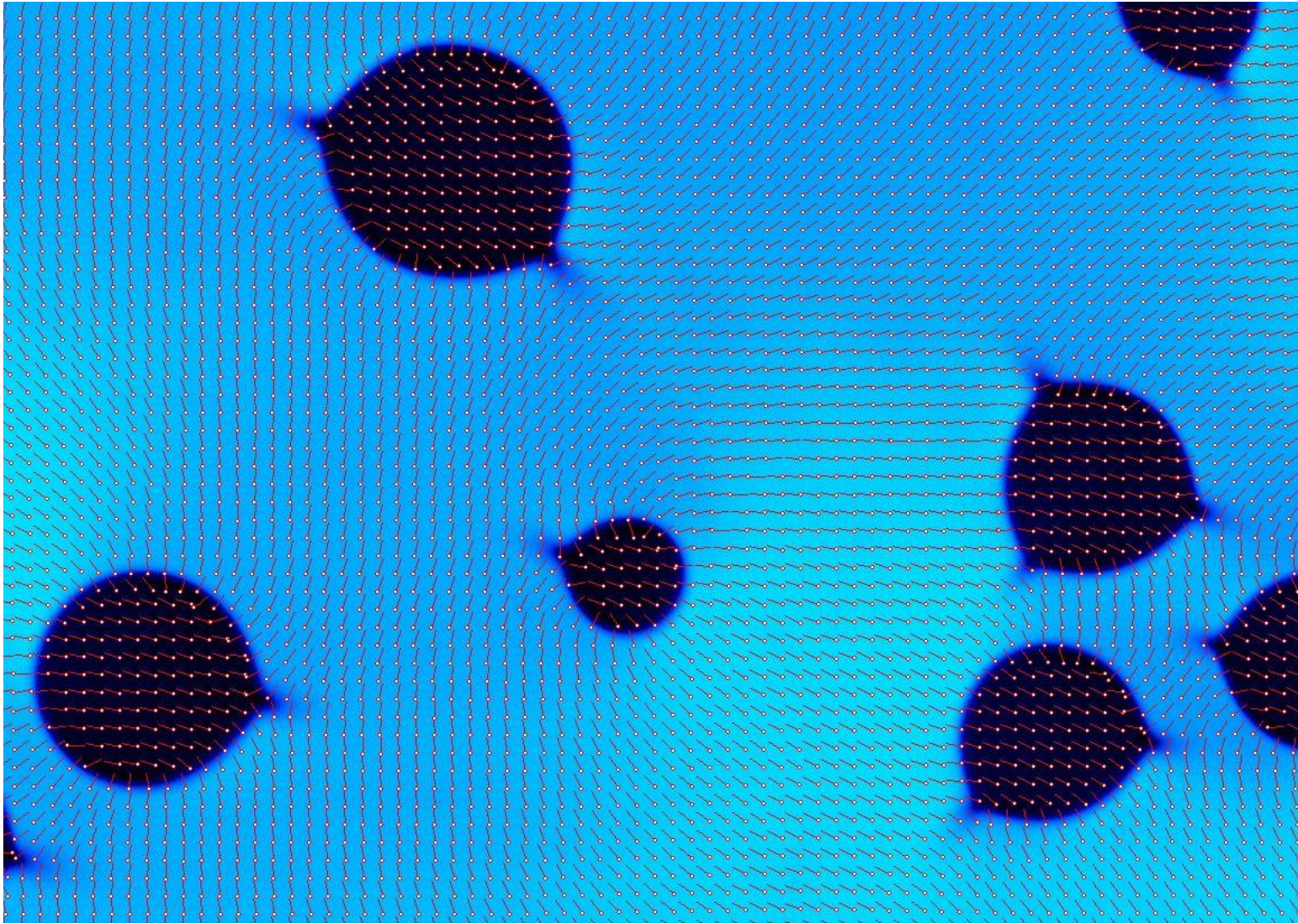
(d) T = 29.7 °C, t = 603 s



$$f_1 - f_{pair} = \frac{\pi K}{2} \ln \frac{L}{\sqrt{2}r_1} + \sqrt{2}\pi r_1 \sigma_0 (\sqrt{2} - 2 - w)$$



Drops of isotropic phase in N environment with distorted director: A balance of surface tension, anchoring, and elasticity



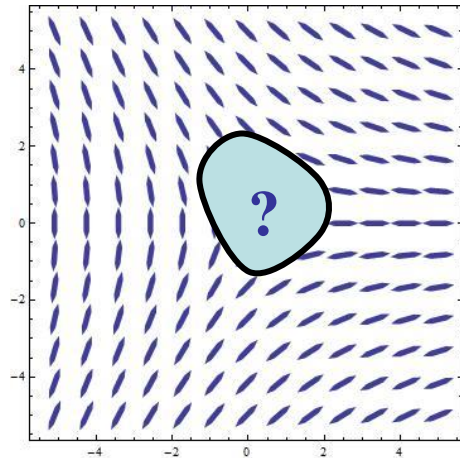
# Equilibrium shape+director?

Difficult problem, requires to minimize both the anisotropic surface energy and elastic interior/exterior

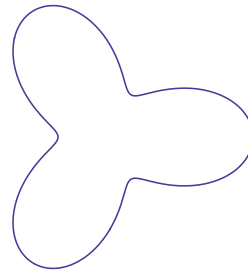
$$\iiint_V f_{FO}(\hat{\mathbf{n}}(\mathbf{r}))dV + \iint_s \left[ \sigma + W(\hat{\mathbf{n}} \cdot \mathbf{v})^2 \right] dS \rightarrow \min$$

First step: Assume “infinite” elasticity (frozen director); then calculate the equilibrium shape of the I tactoid at the core, using the angular dependence of the surface tension for each disclination

$$\sigma(\theta) = \sigma_0 \left\{ 1 + w \cos^2 \left[ (k-1)\theta \right] \right\}$$



$$k = -1/2$$



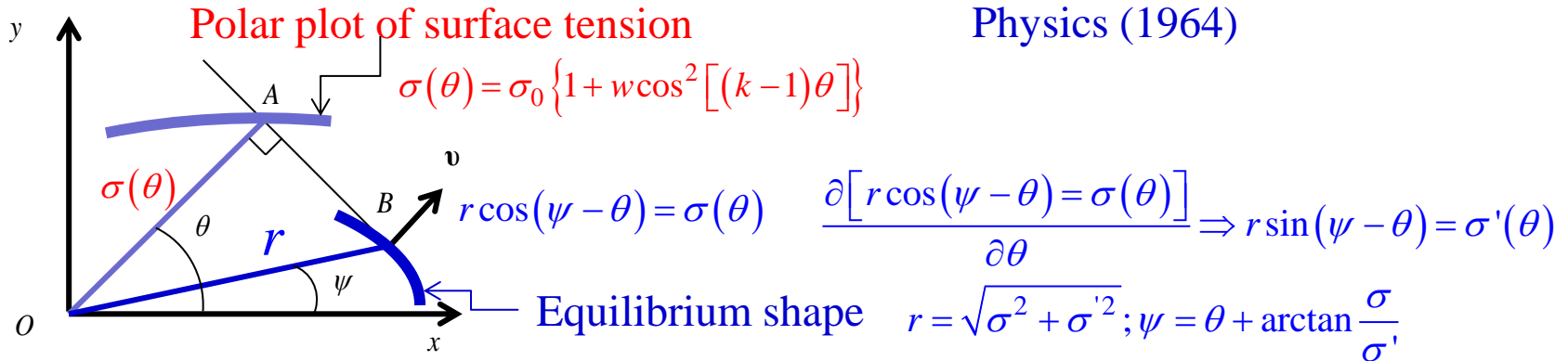
$$w = 2$$

Polar plot of surface tension of an isotropic island inside a nematic region representing a disclination of  $-1/2$  strength;  
Problem: Find the shape that minimizes the anisotropic surface tension



# Equilibrium shape by Wulff construction for distorted director

Wulff construction for crystals:  
L. Landau, E. Lifshits, Statistical  
Physics (1964)



Radius of curvature:  $R = \sqrt{r'^2 + r^2 \psi'^2} = \sigma + \sigma''$

Round shape, all orientations of I-N interface:  $\sigma + \sigma'' > 0$

Missing orientations and cusps:  $\sigma + \sigma'' < 0$

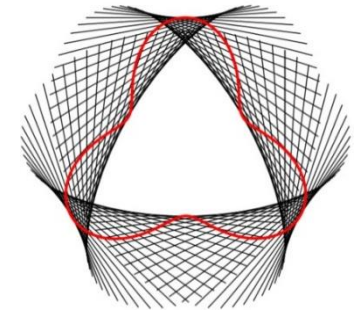
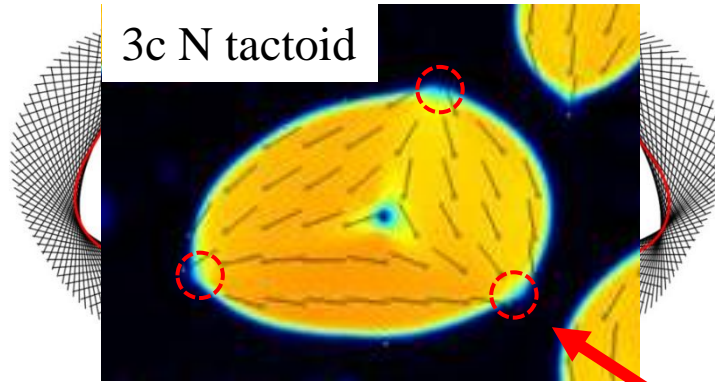
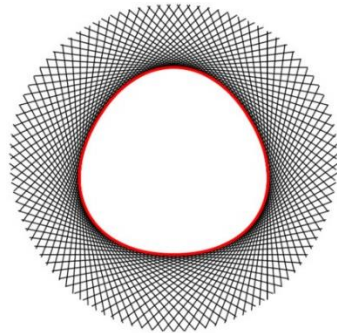
$$1 + w \left[ 1 - 4(k-1)^2 \right] \cos^2 (k-1)\psi + 2(k-1)^2 w < 0$$



# Wulff construction by Mathematica

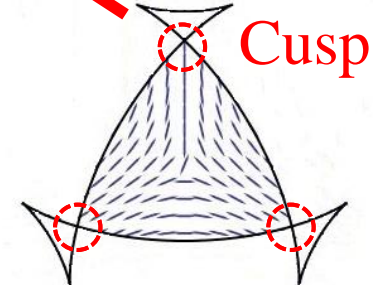
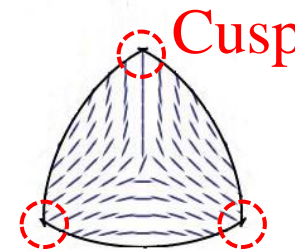
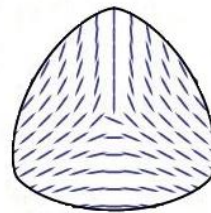
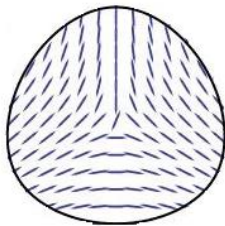
Draw the tangent lines at each point in a polar plot of  $\sigma(\theta)$

Wulff  
Construction



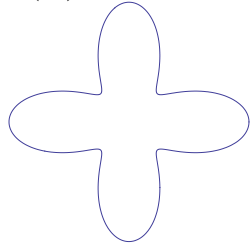
The interior envelope describes the shape in equilibrium

Shape of  
Domain

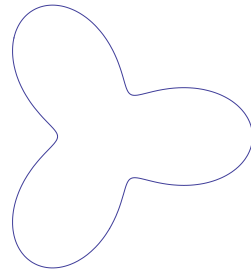


# Equilibrium shape by Wulff construction for distorted director

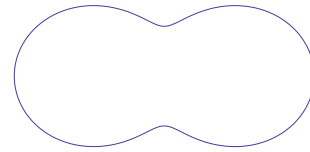
$\sigma(\psi)$



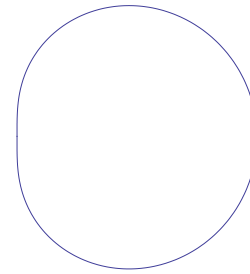
$k = -1; w = 2$



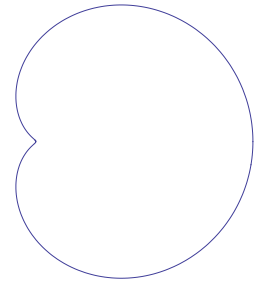
$k = -1/2; w = 2$



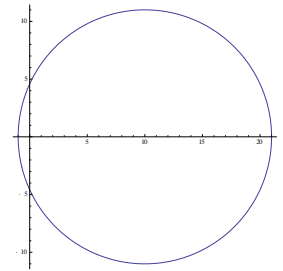
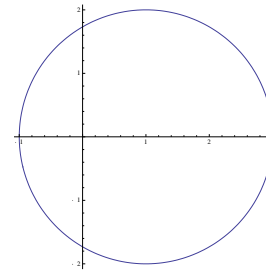
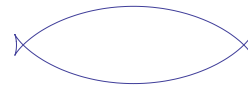
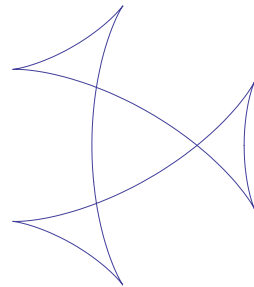
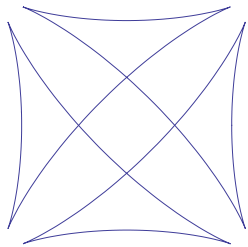
$k = 0; w = 2$



$k = 1/2; w = 2$



$k = 1/2; w = 20$



Missing orientations and cusps:  $1 + w \left[ 1 - 4(k - 1)^2 \right] \cos^2(k - 1)\psi + 2(k - 1)^2 w < 0$

Never the case for  $k=1/2$  and  $k=1$ ; for  $k=0$ ,  $w_c=1$ ; for  $k=-1/2$ ,  $w_c=2/7$ ; for  $k=-1$ ,  $w_c=1/7$

# Summary/What have you learned

- ❑ Disclinations: Frank model, line energy  $\sim \ln$  of size,  $\sim k^2$
- ❑ Integer disclinations: Escape into the third dimension
- ❑ Semi-integer: Stable
- ❑ Homotopy classification: A natural language to describe defects in any medium, LCs and superfluid He-3 including
- ❑ Surface anchoring: Controls topology and energy of defects;
- ❑ Defects occur as equilibrium features in LC droplets, ...Including the nuclei during the phase transition from the isotropic phase
- ❑ Cholesteric: Dirac monopoles
- ❑ Chromonic droplets: Spontaneous chiral symmetry broken; shape is strongly dependent on director deformations, the problem of full energy (elastic+surface tension+anchoring) minimization not solved yet



# Liquid crystals: Lecture 2.2

## Lamellar phases

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Oleg D. Lavrentovich

Support: NSF



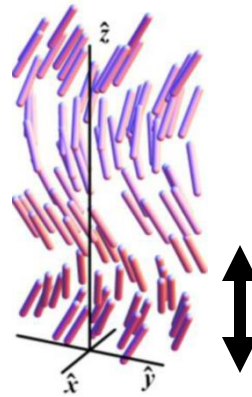
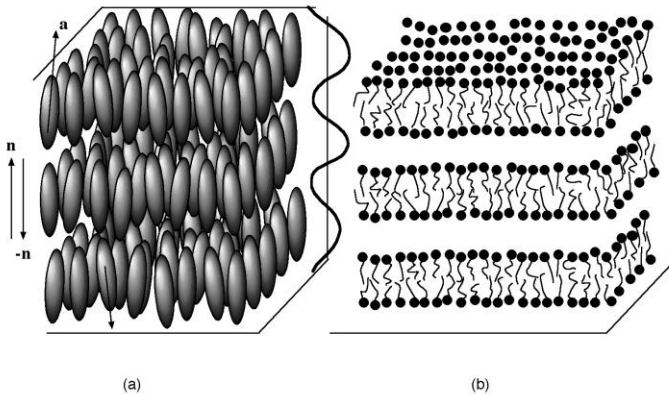
Boulder School for Condensed Matter and Materials Physics,  
Soft Matter In and Out of Equilibrium,  
6-31 July, 2015

# Content

## **Lamellar Phases**

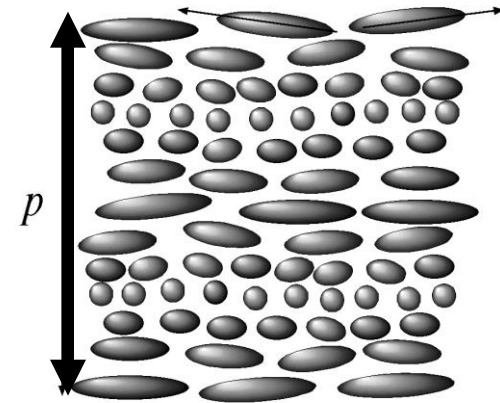
1. Free Energy Density
2. Weak distortions: Dislocations, Undulations
3. Strong distortions: Focal conic domains, grain boundaries

# Lamellar phases



1-10 nm

>0.1  $\mu\text{m}$



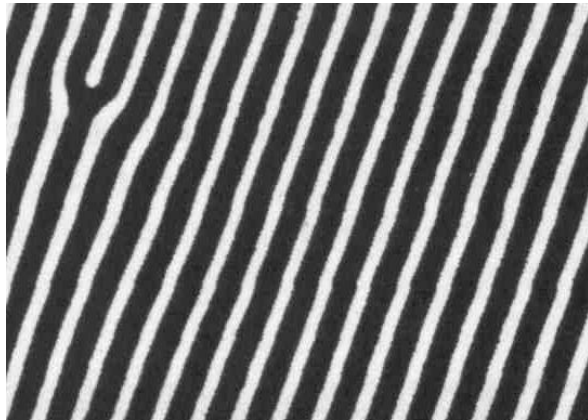
Smectics A  
(thermotropic)

Smectic A  
(lyotropic)

Twist-bend  
nematic

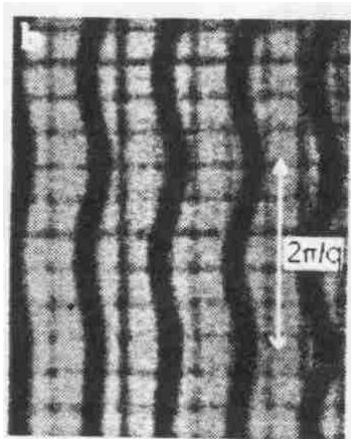
Cholesteric  
(chiral N)

# Weak perturbations: Dislocations, undulations



Stripe magnetic domain

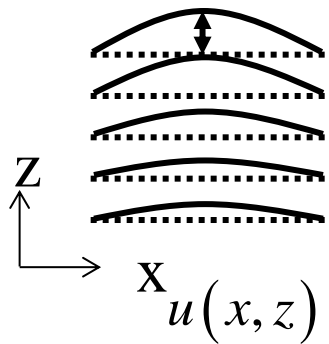
(M. Seul et. al., P.R.L., **68**, 2460 (1992))



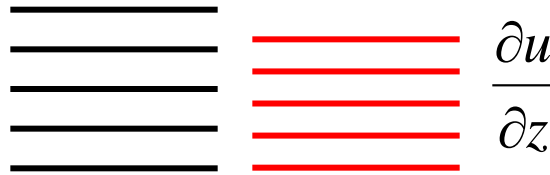
Layered structure in magnetic fluid

(C. Flament et. al., Europhys. Lett., **34**, 225 (1992))

# Elasticity of lamellar phase; 1D translational order

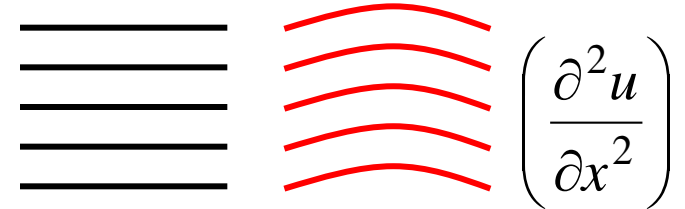


**Compression/dilation:**



$$\frac{\partial u}{\partial z}$$

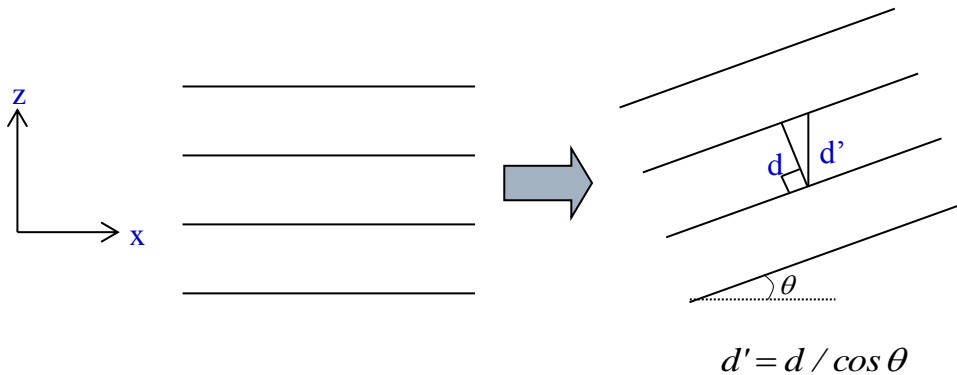
**Curvature:**



$$\left( \frac{\partial^2 u}{\partial x^2} \right)$$

$$f = \frac{1}{2} B \left( \frac{\partial u}{\partial z} \right)^2 + \frac{1}{2} K \left( \frac{\partial^2 u}{\partial x^2} \right)^2$$

**Correction to make the model invariant w.r.t. uniform rotations:**



$$\frac{\partial u}{\partial z} - \frac{1}{2} \left( \frac{\partial u}{\partial x} \right)^2$$



# Elasticity of lamellar phase; 1D translational order

Energy density  $f = \frac{1}{2} B \left\{ \frac{\partial u}{\partial z} - \frac{1}{2} \left( \frac{\partial u}{\partial x} \right)^2 \right\}^2 + \frac{1}{2} K \left( \frac{\partial^2 u}{\partial x^2} \right)^2$

$$[B] = \left[ \frac{J}{m^3} \right] \quad [K] = \left[ \frac{J}{m} = N \right]$$

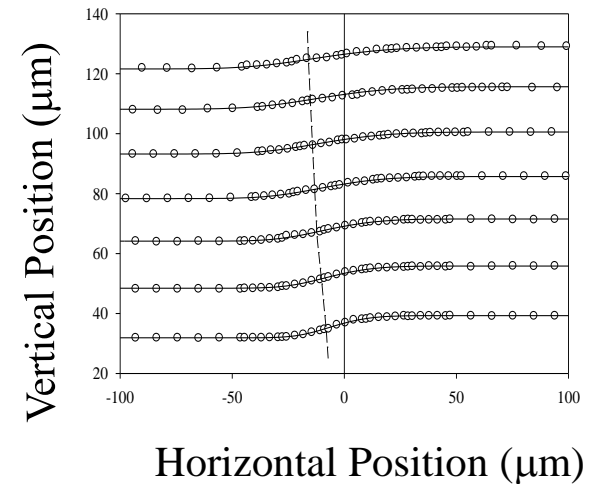
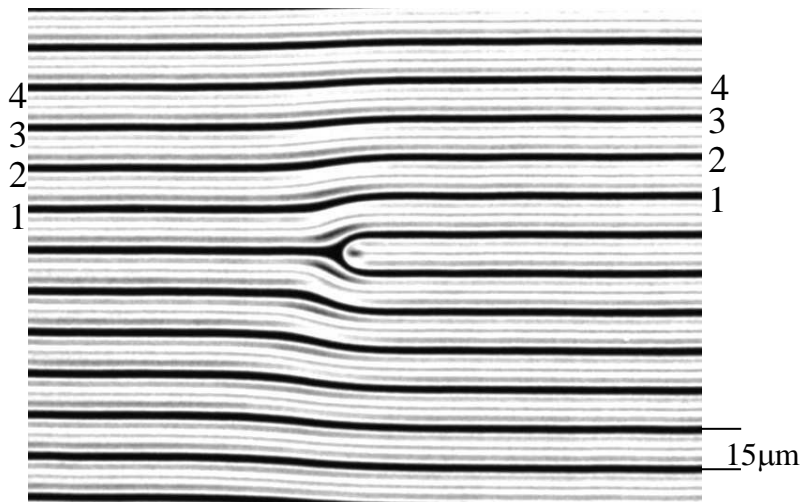
**Material length**  $\lambda = \sqrt{\frac{K}{B}} \sim \text{period?}$

# Elasticity of Smectics: Dislocation

$$u(x, y) = 2\lambda \ln \left\{ 1 + \frac{\exp(b/4\lambda) - 1}{2} \left[ 1 + \operatorname{erf} \left( \frac{x}{2\sqrt{\lambda z}} \right) \right] \right\}$$

Brener and Marchenko PRE'99

By fitting  $u(x, z)$ , one can measure  $\lambda = \sqrt{\frac{K}{B}}$



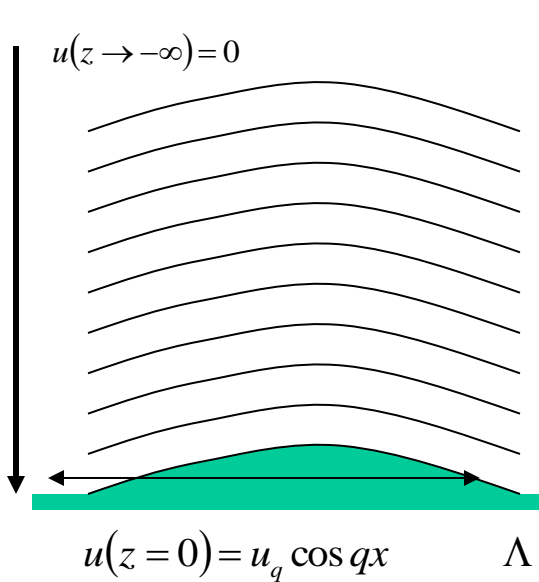
$period = 14.9 \mu m$

$\lambda = 2.7 \mu m$

$\lambda \approx 0.2 \times period$

# Long range effect of layers deformations

$$u(x, z) \quad \text{E-L equation} \quad K \frac{\partial^4 u}{\partial x^4} - B \frac{\partial^2 u}{\partial z^2} = 0$$



Look for solution of the type:  $u = u_q \cos qx \exp\left(\frac{z}{L}\right)$

$$u(z = 0) = u_q \cos qx \quad u(z \rightarrow -\infty) = 0$$

$$K_1 q^4 - B \frac{1}{L^2} = 0 \quad \rightarrow \quad L = \frac{1}{q^2} \sqrt{\frac{B}{K}} \propto \frac{\Lambda^2}{\lambda}$$

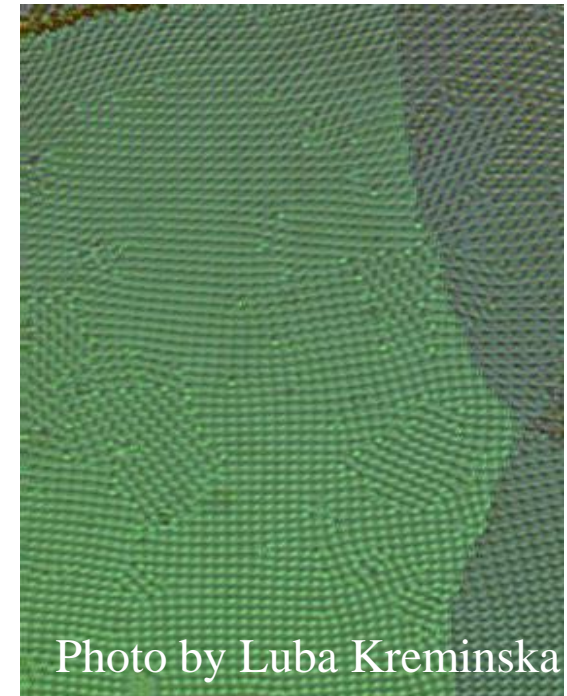
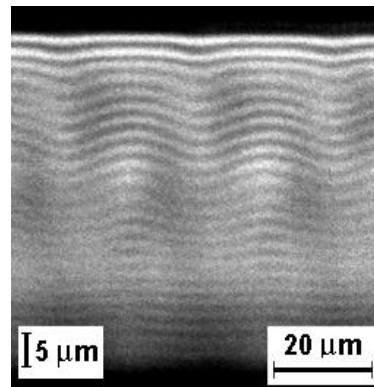
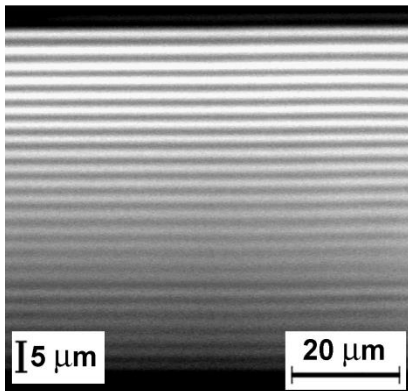
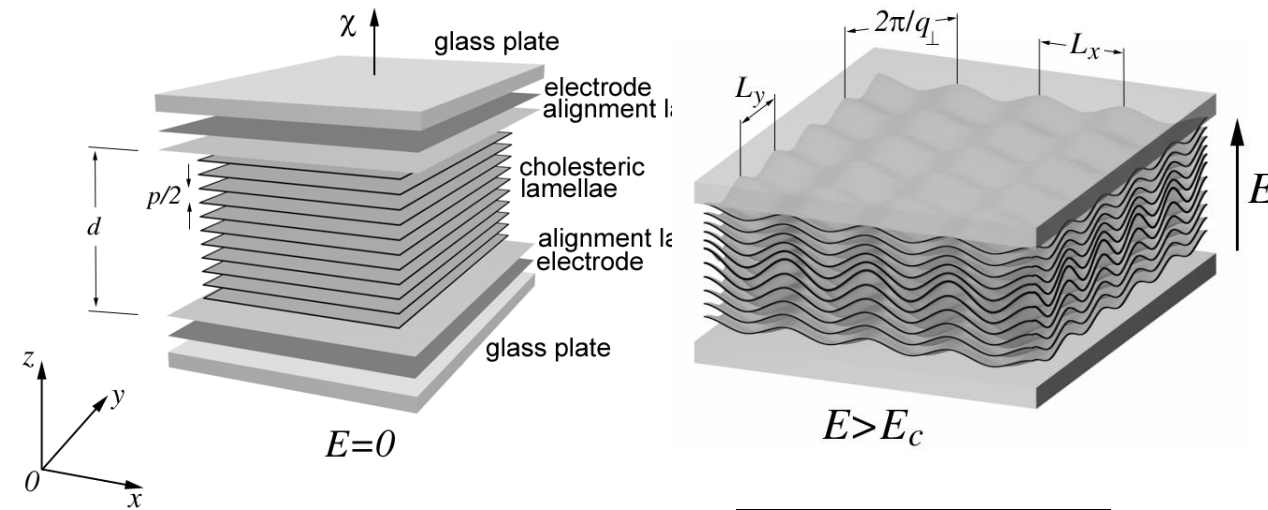
$$L \equiv \frac{\text{macroscopic scale}^2}{\text{microscale}} = \Lambda \left( \frac{\Lambda}{\lambda} \right) \gg \Lambda$$

Saint-Venant principle  $L \sim \Lambda$  is not applicable to SmA materials;

Smectics: A good model of “la Princesse sur la graine de pois”

If a pea is 1 mm, layer thickness 10 nm, then the pea is felt over 100 m!

# Undulations in Cholesteric caused by E field



Senyuk et al PRE 74, 011712 (2006)

# Undulations in Cholesteric (2D)

$$f = \frac{1}{2} K \left( \frac{\partial^2 u}{\partial x^2} \right)^2 + \frac{1}{2} B \left\{ \frac{\partial u}{\partial z} - \frac{1}{2} \left( \frac{\partial u}{\partial x} \right)^2 \right\}^2 - \frac{1}{2} \varepsilon_0 \varepsilon_a E^2 \left( \frac{\partial u}{\partial x} \right)^2$$

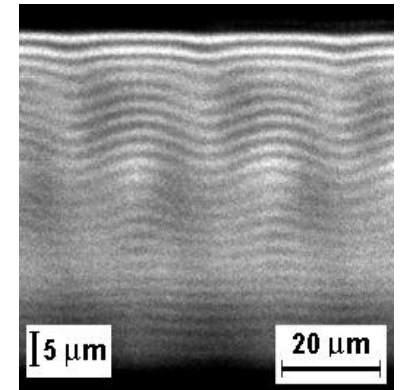
$$f_{\text{anchoring}} = \frac{1}{2} W \left[ \left( \frac{\partial u}{\partial x} \right)^2 \right]_{z=0,d}$$

$$u(x, z) = u_0 \phi(z) \sin q_x x \quad \Rightarrow \quad \begin{cases} B\phi'' - q_x^2 (Kq_x^2 - \varepsilon_0 \varepsilon_a E^2) \phi = 0 \\ B\phi' \pm Wq_x^2 \phi = 0 \Big|_{z=\pm a/2} \end{cases}$$

$$\phi = \text{const} \times \cos q_z z$$

$$\left\{ \begin{array}{l} q_z = q_x \sqrt{\kappa - \lambda^2 q_x^2} \\ W = \frac{B \sqrt{\kappa - \lambda^2 q_x^2}}{q_x \cot \left( \frac{dq_x}{2} \sqrt{\kappa - \lambda^2 q_x^2} \right)} \end{array} \right. \quad \kappa = \varepsilon_0 \varepsilon_a E^2 / B \quad \Rightarrow \quad E_c \approx \sqrt{\frac{2\pi K}{\varepsilon_0 \varepsilon_a \lambda a}}$$

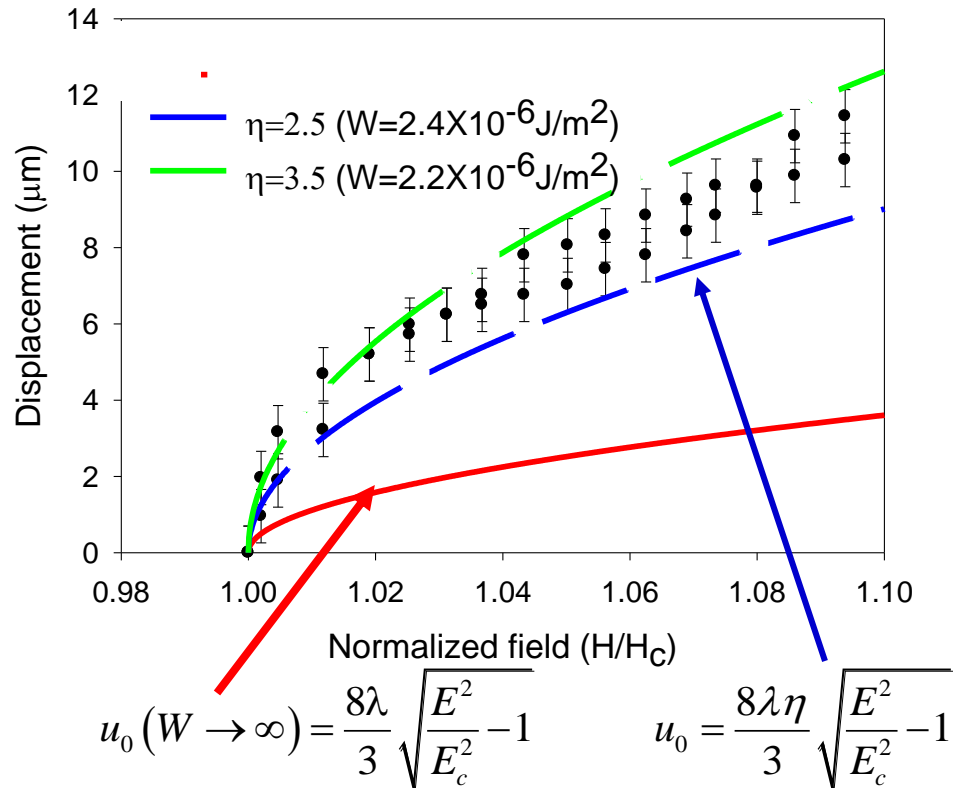
$$u_0 (W \rightarrow \infty) = \frac{8\lambda}{3} \sqrt{\frac{E^2}{E_c^2} - 1} \quad u_0 = \frac{8\lambda\eta}{3} \sqrt{\frac{E^2}{E_c^2} - 1}$$



3D version: Senyuk et al PRE 74, 011712 (2006)

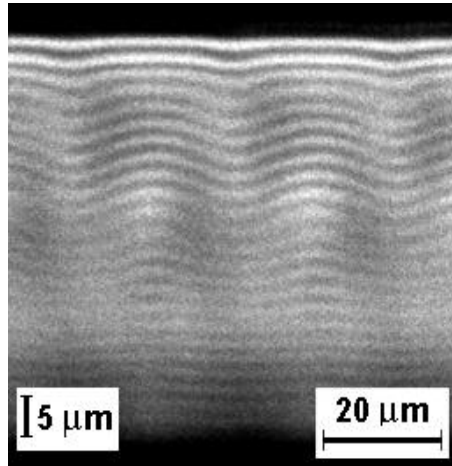


# Undulations in Cholesteric (2D)

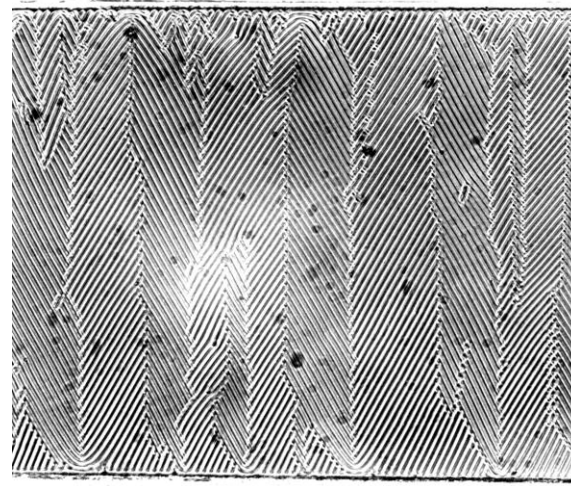


Ishikawa et al PRE 63, 030501(R) (2001)

# Undulations in Cholesteric above threshold

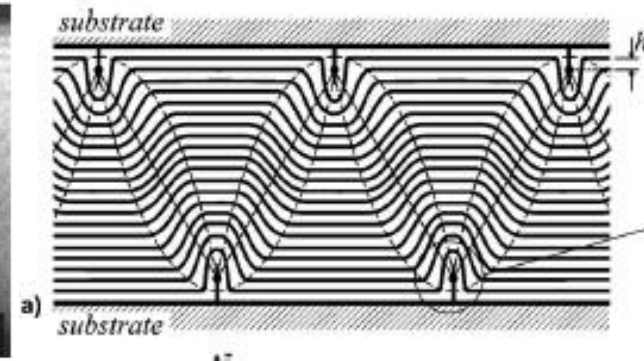
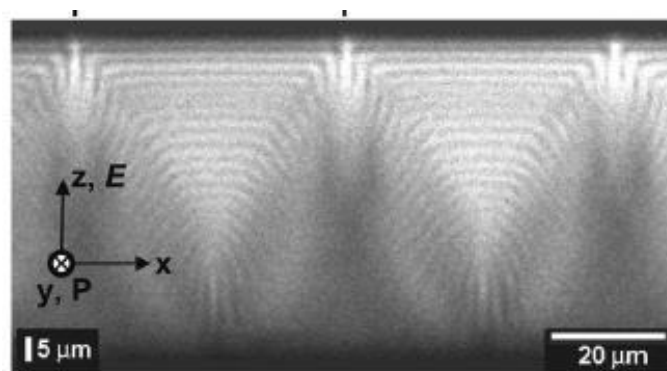


Immediately above the threshold

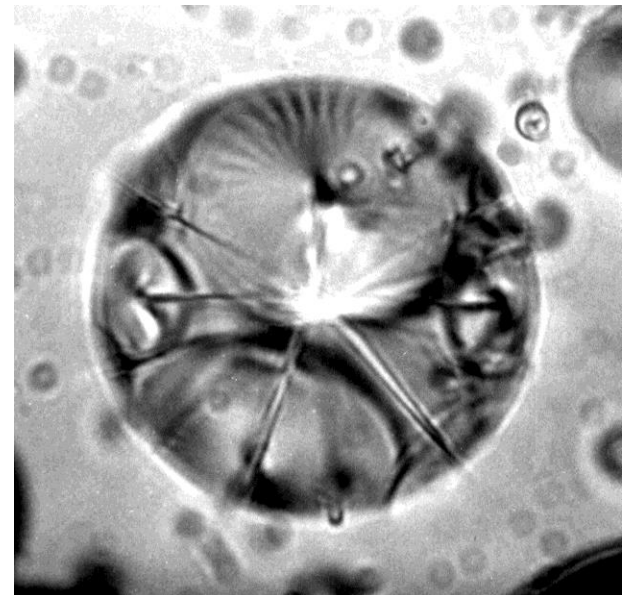
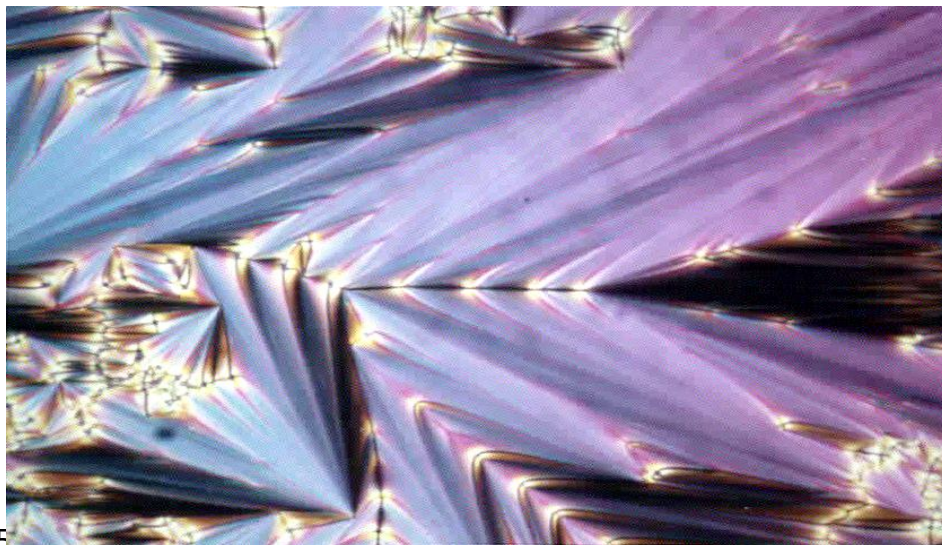
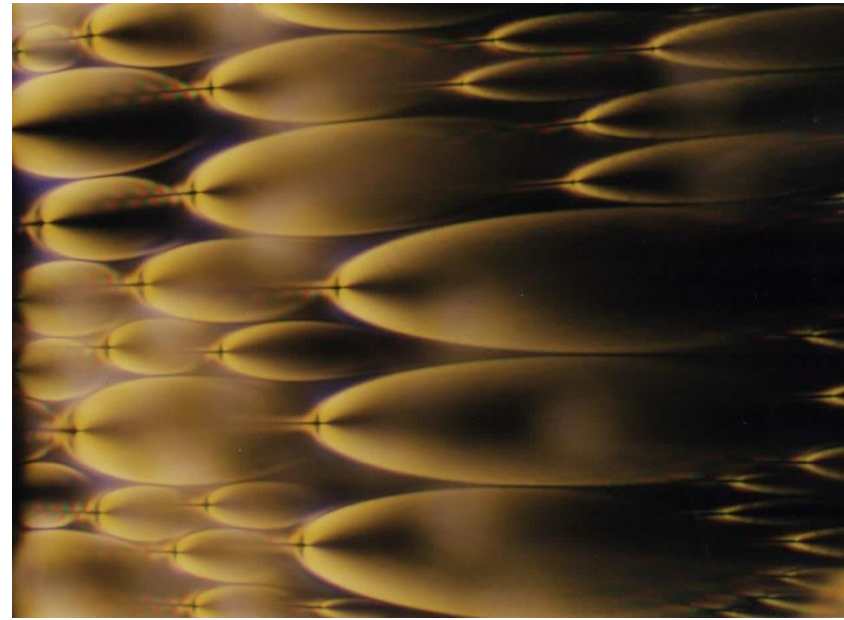
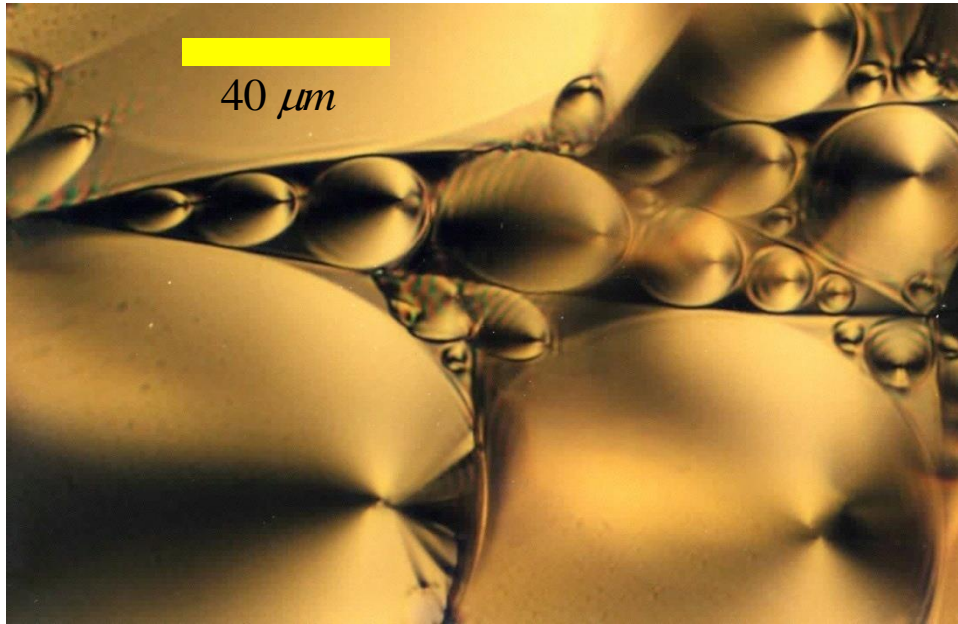


Well above the threshold, 2D cell; broken surface anchoring

Well above the threshold, 3D, Anchoring takes over, forcing parabolic domain walls



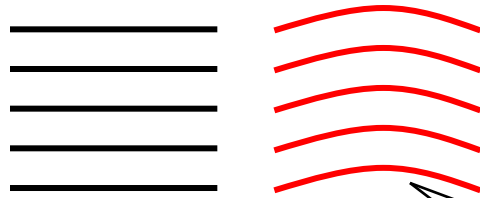
# Strong perturbations: Focal conic domains



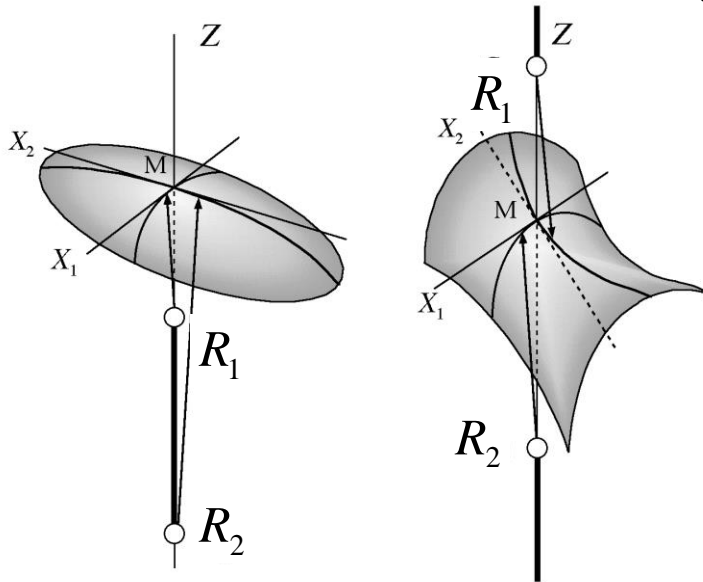
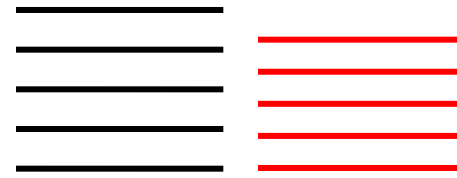


# Elasticity of Smectics: Strong distortions

**Curvature:**



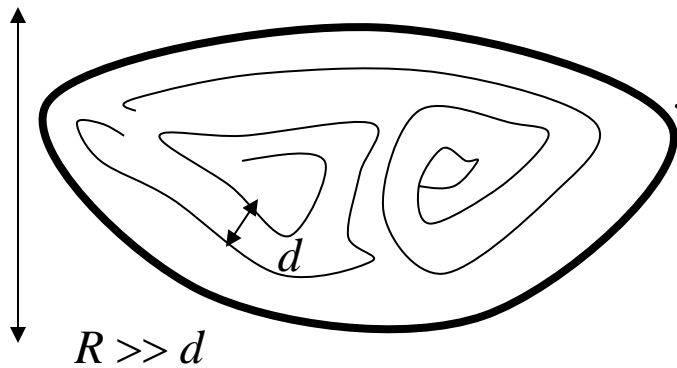
**Compression/Dilation:**



$$f = \frac{1}{2} K \left( \frac{1}{R_1} + \frac{1}{R_2} \right)^2 + \bar{K} \frac{1}{R_1 R_2} + \frac{1}{2} B \left( \frac{d}{d_0} - 1 \right)^2$$

principal radii

# Elasticity of Smectics



$$f = f_{curv} + f_{dilat} = \frac{1}{2} K \left( \frac{1}{R_1} + \frac{1}{R_2} \right)^2 + \bar{K} \left( \frac{1}{R_1 R_2} \right) + \frac{1}{2} B \left( \frac{d - d_0}{d_0} \right)^2$$

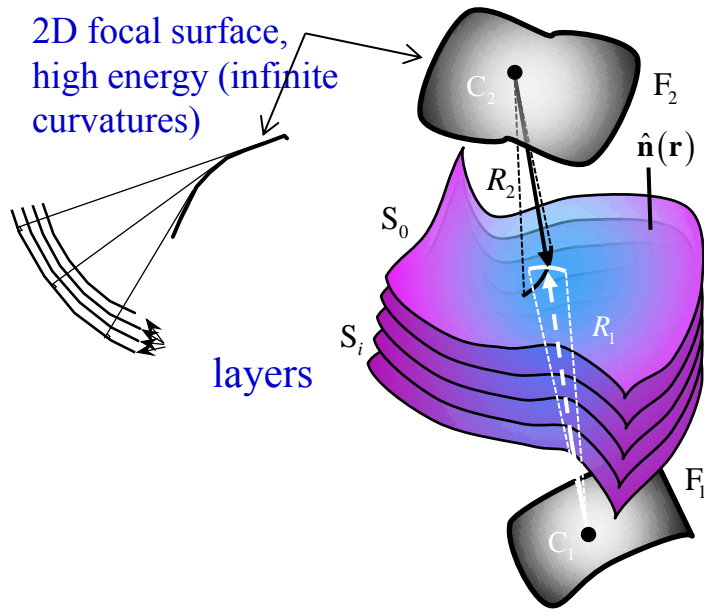
$$\lambda = \sqrt{\frac{K}{B}} \sim d_0$$

$$\frac{F_{curv}}{F_{dilat}} \sim \frac{KR}{BR^3} \sim \left( \frac{\lambda}{R} \right)^2 \ll 1 \quad \text{if } R \gg \lambda$$

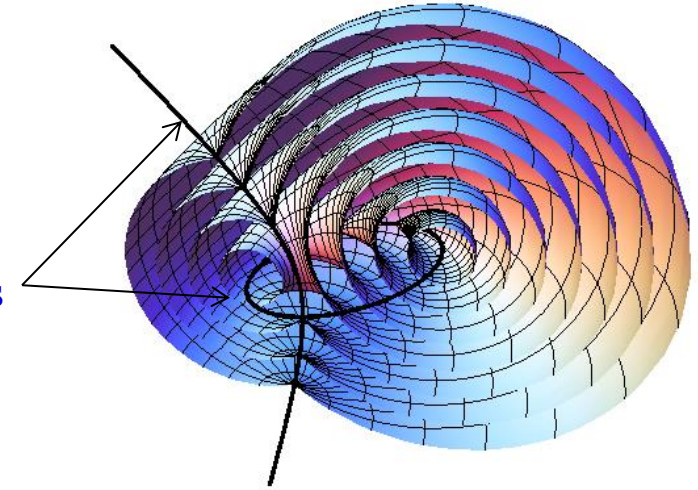


At large scales of deformations,  
the curved layers are equidistant

# Curved smectic layers: Dupin Cyclides



2D focal surfaces shrink into lines



To reduce the energy, focal surfaces in SmA degenerate into 1D focal lines, that can be only of three types:

- (a) circle and straight line, or
- (b) confocal ellipse and hyperbola,
- (c) two confocal parabolae

producing a Focal Conic Domain (FCD); the smectic layers are **Dupin cyclides**

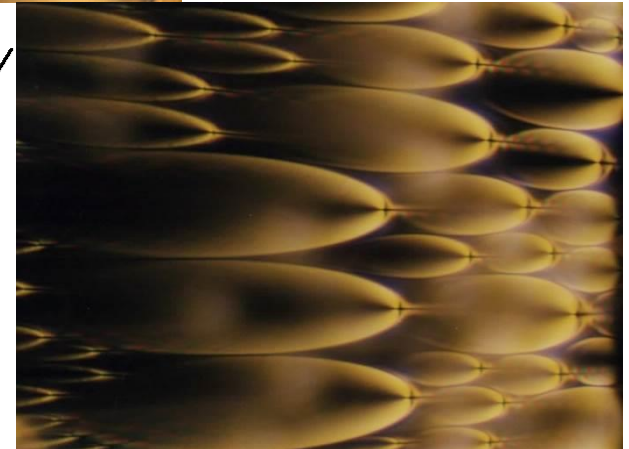
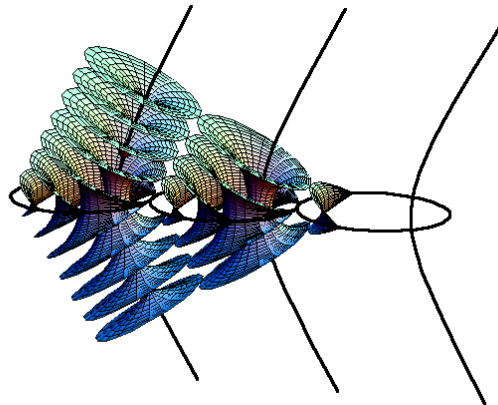
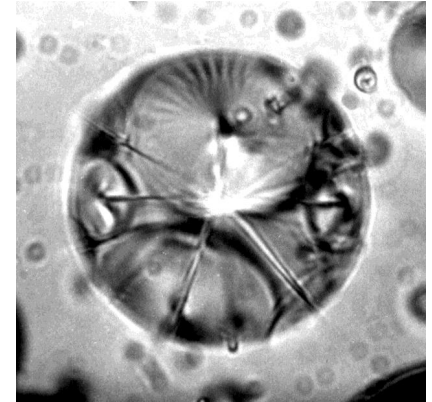
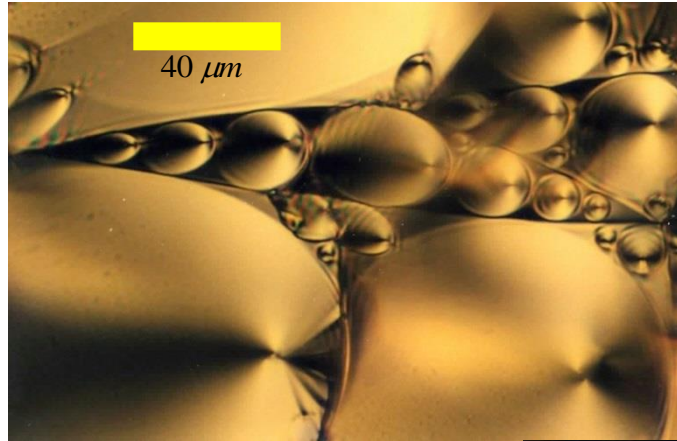
C.P. Dupin, Applications de Géométrie (Paris, 1822)

J. Maxwell, On the cyclide, Q. J. Pure Appl. Math **9**, 111 (1868)

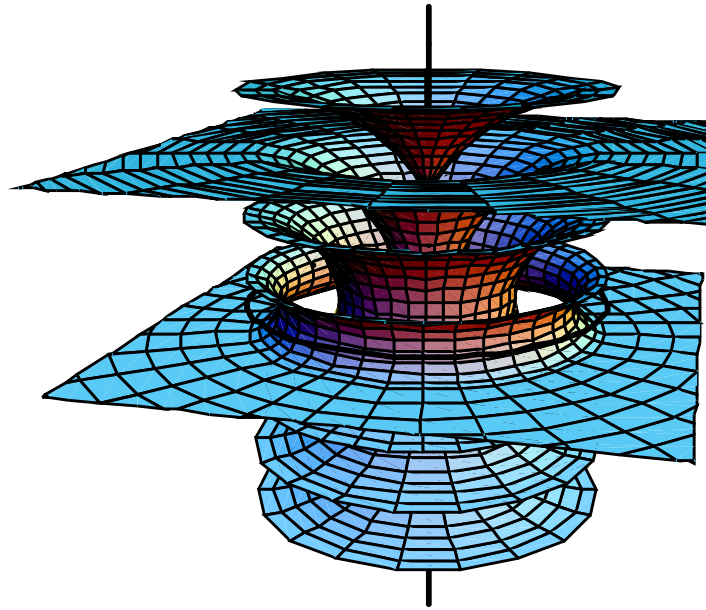
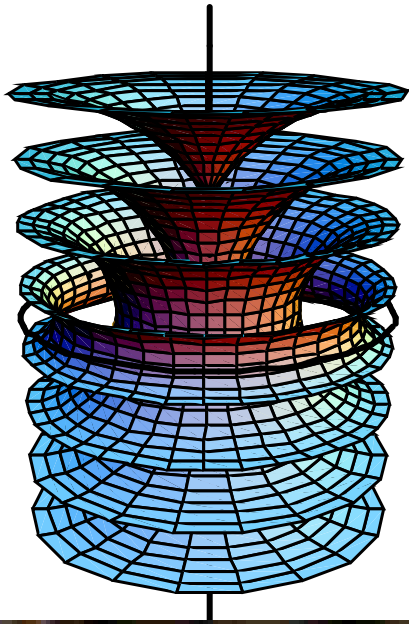


# Smectics and focal conic domains

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



# Toroidal Focal Conic Domains

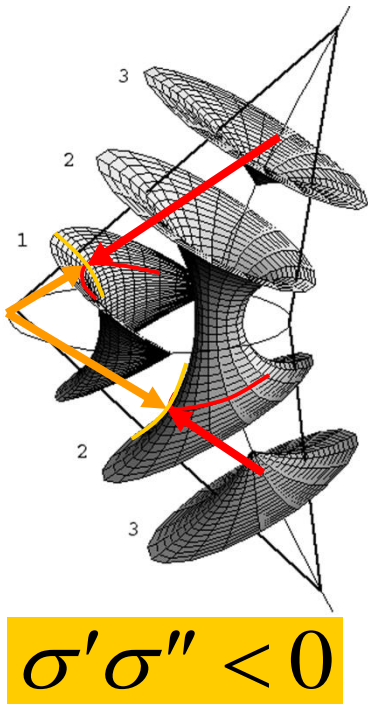


Smoothly fits into the system of parallel layers

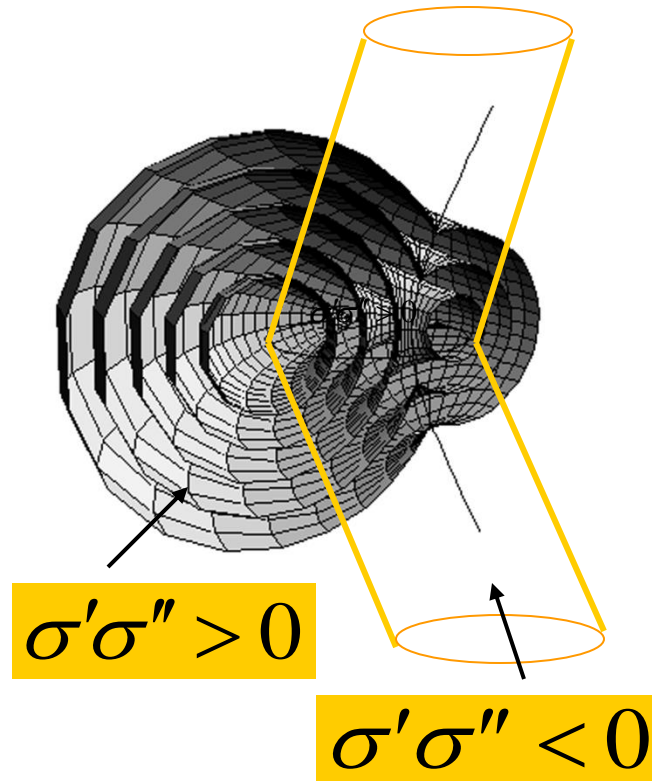


# Classification of FCDs by Gaussian curvature

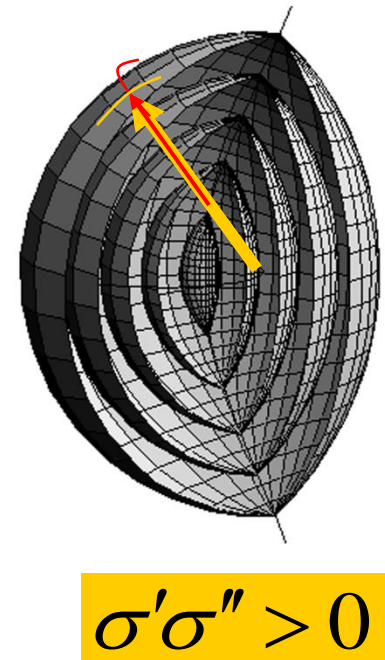
FCD-I



FCD-III

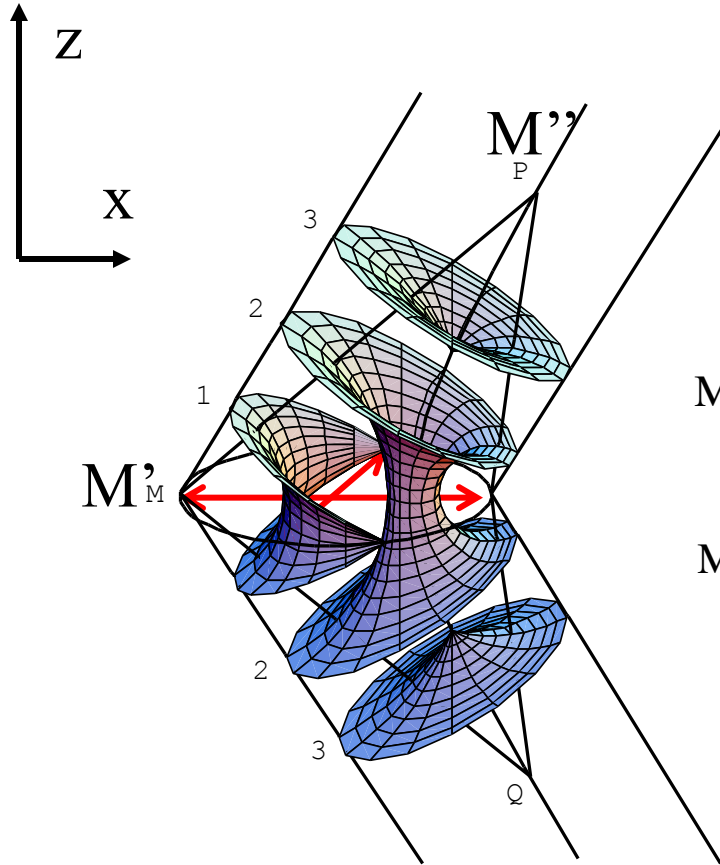


FCD-II



How to describe the FCD analytically?

# Curvilinear coordinates $r, u, v$



Ellipse

$$z=0, \quad \frac{x^2}{a^2} + \frac{y^2}{b^2} = 1$$

Hyperbola

$$y=0, \quad \frac{x^2}{a^2 - b^2} - \frac{z^2}{b^2} = 1$$

Parametrization of conics with coordinates  $u$  and  $v$  defined within the cyclides

$$M' \begin{cases} x' = a \cos u, \\ y' = b \sin u, \end{cases} \quad 0 \leq u \leq 2\pi,$$

$$M'' \begin{cases} x'' = \pm c \cosh v \\ z'' = b \sinh v \end{cases} \quad -\infty \leq v < \infty, \quad c^2 = a^2 - b^2$$

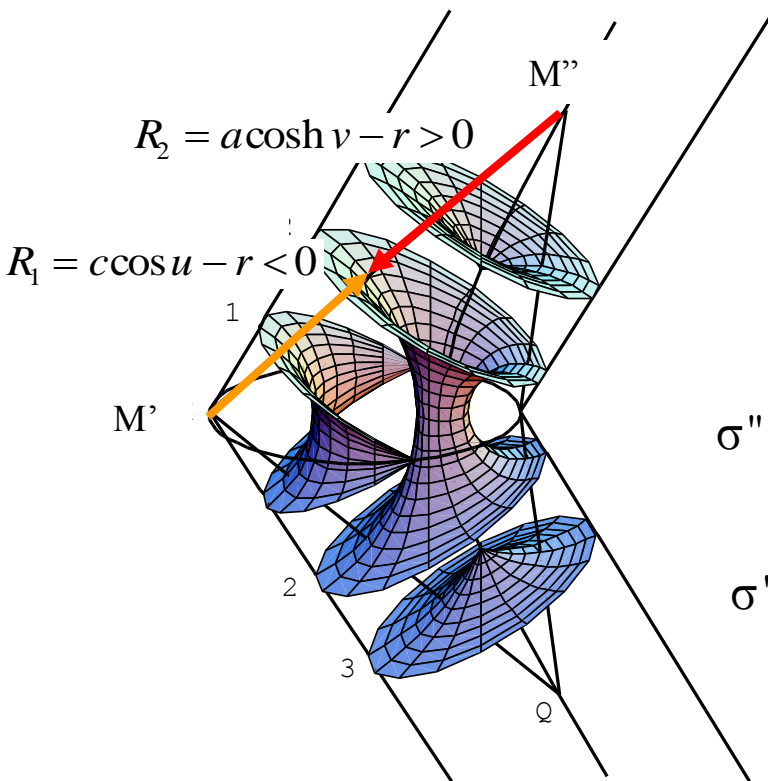
$$M''M' = a \cosh v - c \cos u$$

# Curvature energy functional

Let us introduce the third coordinate  $r$  that “counts” the layers

$$M''M' = a \cosh v - c \cos u = R_2 - R_1 = a \cosh v - r + r - c \cos u$$

$$R_2 = a \cosh v - r > 0 \quad R_1 = c \cos u - r < 0$$



$$F_{curv} = \int_{FCD} \left[ \frac{1}{2} K \left( \frac{1}{R_1} + \frac{1}{R_2} \right)^2 + \bar{K} \frac{1}{R_1 R_2} \right] dV$$

$$\sigma'' = \frac{1}{R_2} = \frac{1}{a \cosh v - r} > 0$$

$$\sigma' = \frac{1}{R_1} = \frac{1}{c \cos u - r} < 0$$

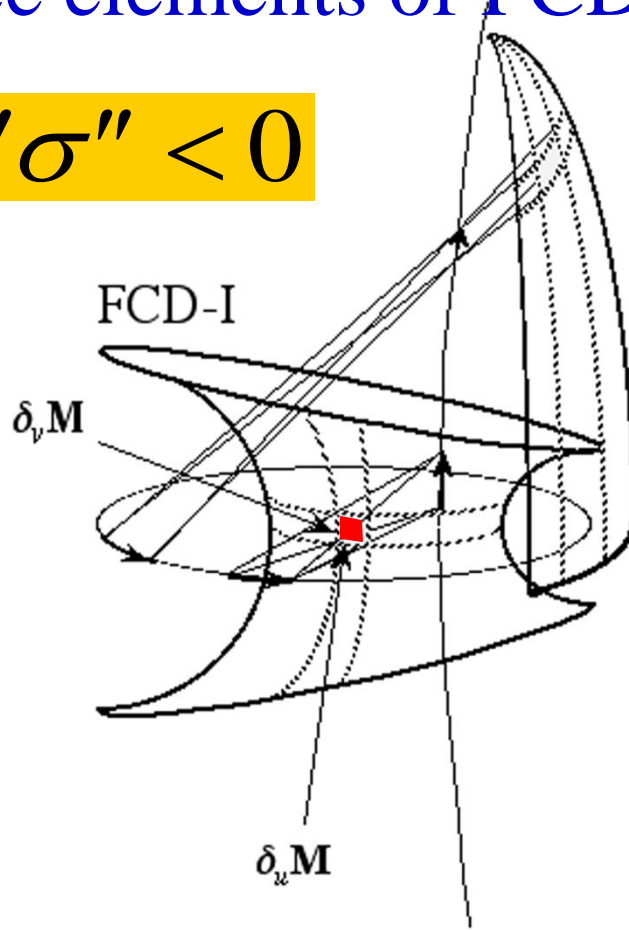
$$dV = dr \times d\Sigma = dr \times \frac{b^2 |\sigma' \sigma''|}{(\sigma' - \sigma'')^2} du dv$$



# Surface elements of FCD I and FCD II

$$\sigma' \sigma'' < 0$$

$$\sigma' \sigma'' > 0$$



$$d\Sigma(r) = AB du dv$$

$$A du = |\delta_u \mathbf{M}| = \left| \delta_u \left( \frac{r'}{\Delta} \mathbf{M}' \mathbf{M}'' \right) \right| = \pm \frac{b \sigma'}{\sigma' - \sigma''} du,$$

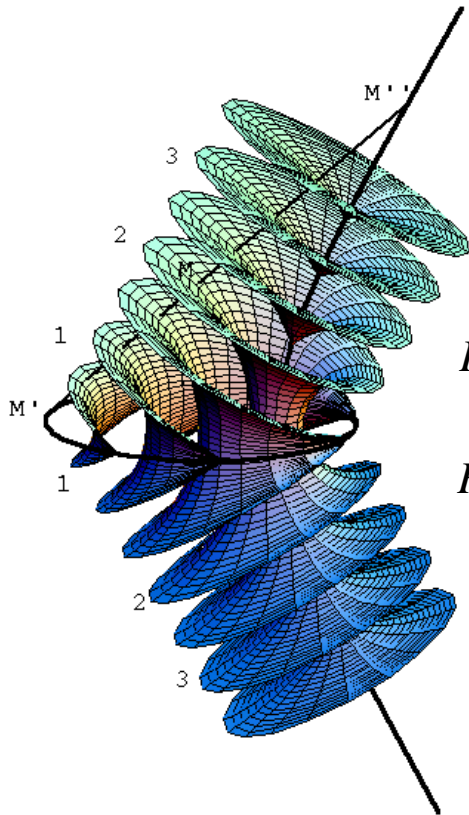
$$\Delta = r'' - r'$$

$$\mathbf{M}' \mathbf{M}'' = |a \cosh v - c \cos u| = |e^x - e^{-1} x'|$$

$$B dv = \pm \frac{b \sigma'}{\sigma' - \sigma''} dv$$

$$d\Sigma(r) = \frac{b^2 |\sigma' \sigma''|}{(\sigma' - \sigma'')^2} du dv$$

# Curvature energy of FCD-I



$$f = \frac{1}{2} K(\sigma' + \sigma'')^2 + \bar{K}\sigma'\sigma'' \quad F_{curv} = \int_{FCD} f \frac{b^2 |\sigma'\sigma''|}{(\sigma' - \sigma'')^2} dudvdr$$

$$f = \frac{1}{2} K(\sigma' - \sigma'')^2 + (2K + \bar{K})\sigma'\sigma'' \quad F_{curv} = F_1 + F_2$$

$$F_1 = -\frac{1}{2} K(1 - e^2) a \int \frac{du dv d\rho}{(e \cos u - \rho)(\cosh v - \rho)}$$

$$F_2 = -(2K + \bar{K})(1 - e^2) a \int \frac{du dv d\rho}{(\cosh v - e \cos u)^2}$$

$$\rho = r / a$$

$$r_{cutoff} = a \cosh v - r_c$$

$$r_{cutoff} = c \cos u + r_c$$

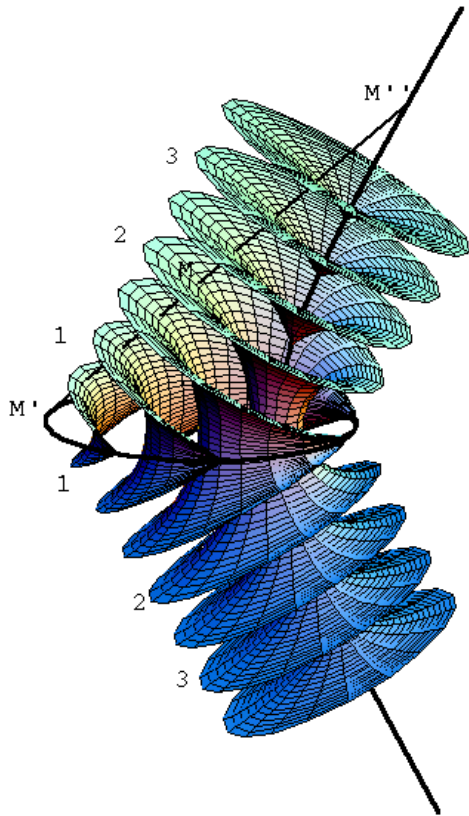
Mathematica©  
cannot do it....

substitutions such as

$$\frac{\cosh^2 v - e^2}{1 - e^2} = \frac{1}{\cos^2 x}$$

lead to the desired result...

# Curvature energy of FCD-I

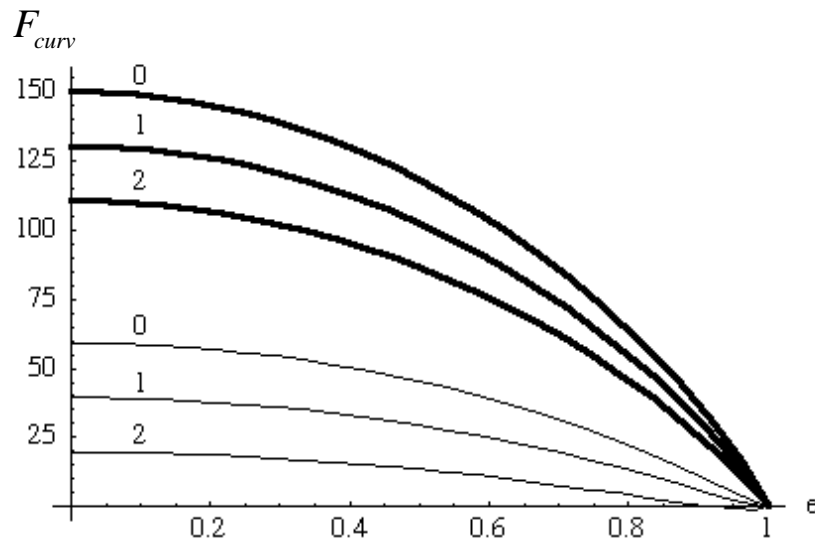


$$F_{curv} = 4\pi a(1-e^2) \mathbf{K}(e^2) \left[ K \left( \ln \frac{2a\sqrt{1-e^2}}{r_c} - 2 \right) - \bar{K} \right] + F_{core}$$

$$F_{core} \approx 8aK \mathbf{E}(e^2)$$

valid for any  $0 \leq e < 1$

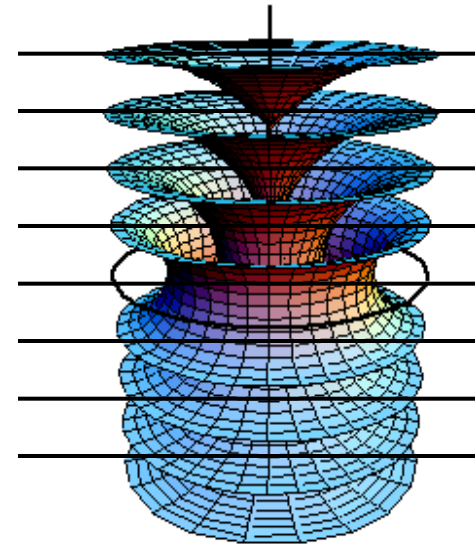
$\mathbf{K}(x), \mathbf{E}(x)$  complete elliptic integrals of the first and second kind



# Circular FCDs-I

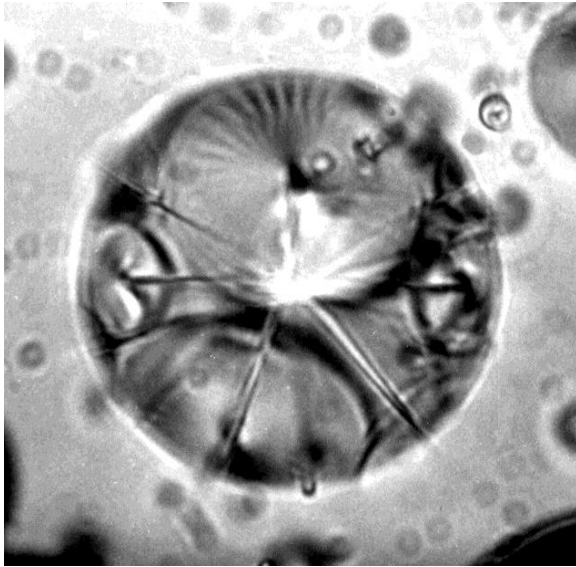
$$F = 2\pi^2 a \left( K \ln \frac{a}{r_c} + K (\ln 2 - 2) - \bar{K} \right)$$

$$\begin{aligned} a &= b \\ e &= 0 \\ \omega &= 0 \end{aligned}$$

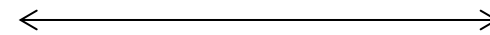
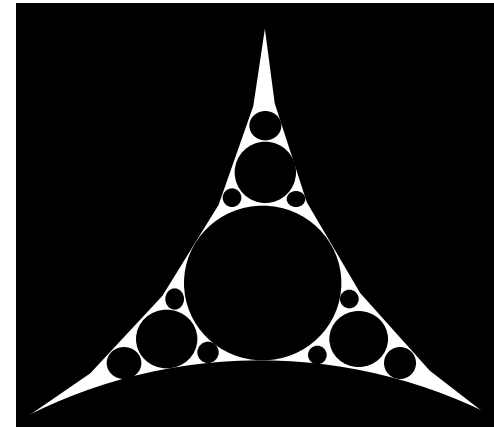


Home assignment: Derive  $F$  for a circular FCD

# Anchoring-Controlled FCD Assembly



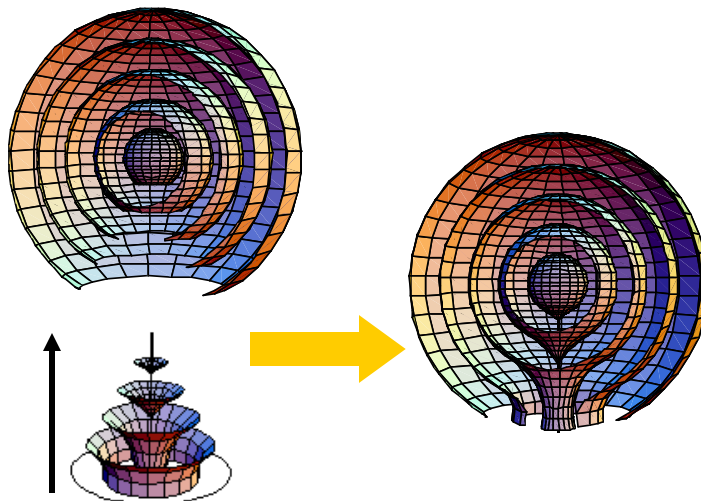
Apollonius  
filling



L, sample size

Black: tangential anchoring

White: Normal anchoring

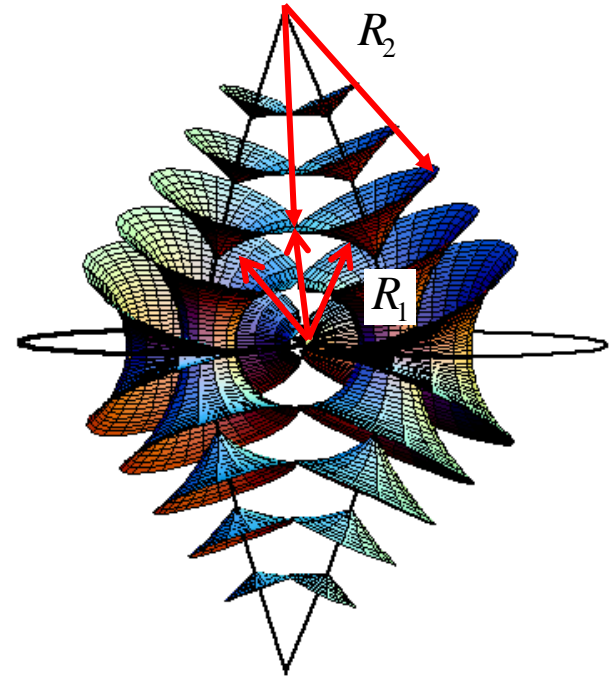
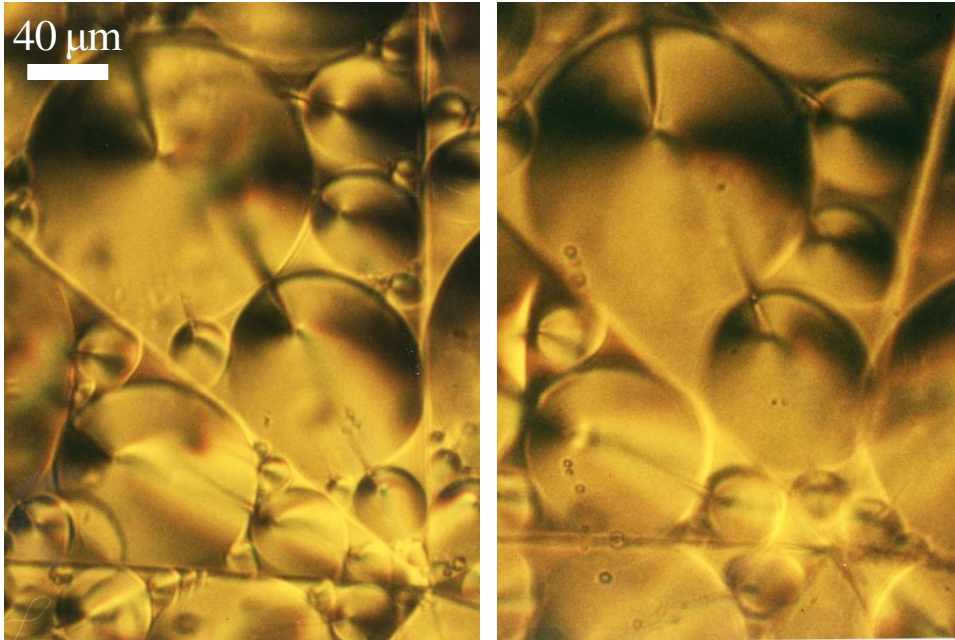


$$F(b) \sim Kb - \Delta\gamma b^2$$

$$\frac{\partial F}{\partial b} = 0 \Rightarrow b^* \sim K / \Delta\gamma \gg \lambda$$

The smallest FCDs  
are macroscopic

# Associations of FCDs

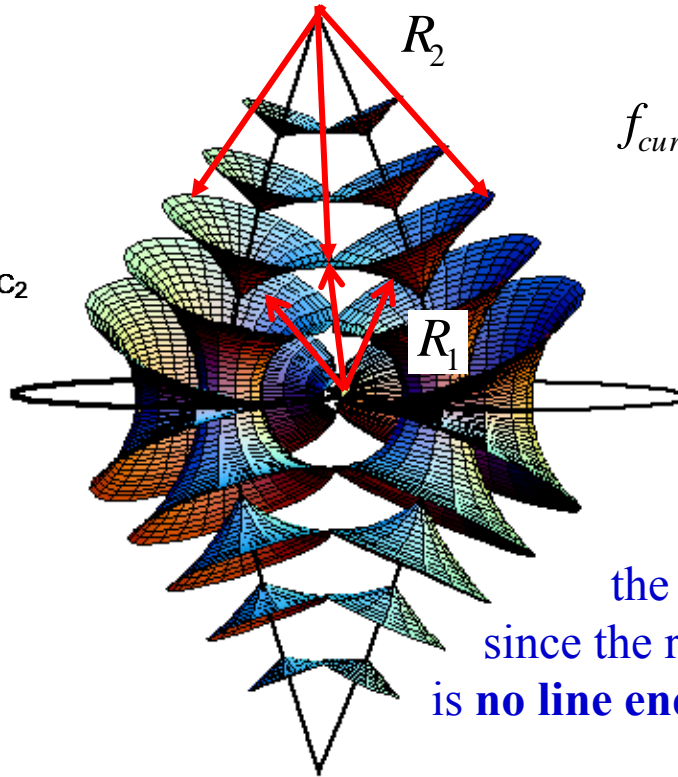
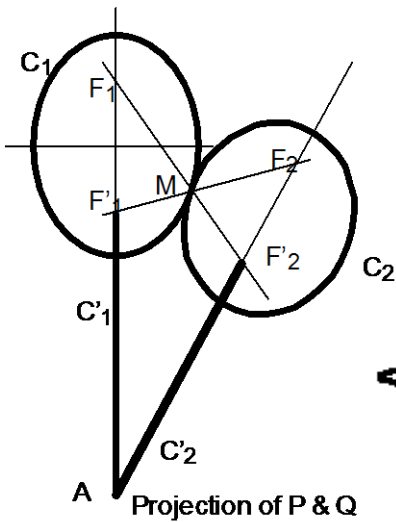


FCDs form families with common apex; Bragg, Nature (1934)



# Law of Corresponding Cones

Friedel, 1922: When two coplanar ellipses are in contact at M, the two corresponding hyperbolae have two points of intersection P and Q and the domains have two generatrices MP and QM of contact.



$$f_{curv} = \frac{1}{2} K \left( \frac{1}{R_1} + \frac{1}{R_2} \right)^2 + \bar{K} \frac{1}{R_1 R_2}$$

At the line of contact of two FCDs, the curvature energy densities are equal since the radii of curvature are the same; there is **no line energy** at the line of contact

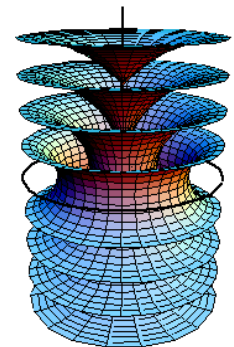
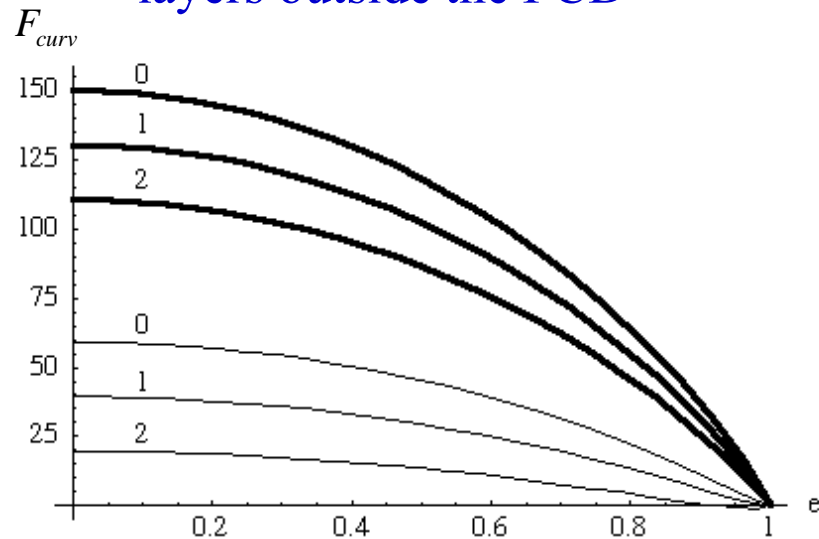
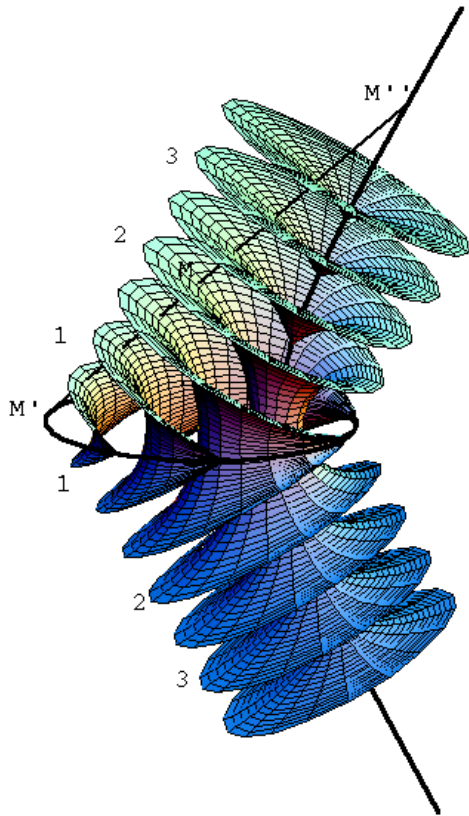
# Curvature energy of FCD-I

$$F_{curv} = 4\pi a(1-e^2) \mathbf{K}(e^2) \left[ K \left( \ln \frac{2a\sqrt{1-e^2}}{r_c} - 2 \right) - \bar{K} \right] + F_{core}$$

$$F_{core} \approx 8aK \mathbf{E}(e^2)$$

valid for any  $0 \leq e < 1$

Energy decreases as eccentricity increases.  
However eccentricity is not a minimization parameter as it is often fixed by the geometry of layers outside the FCD



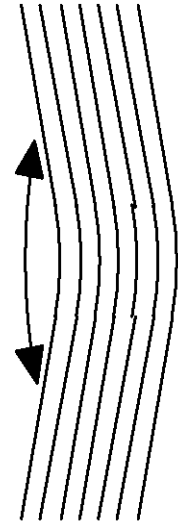
# Grain Boundaries in SmA



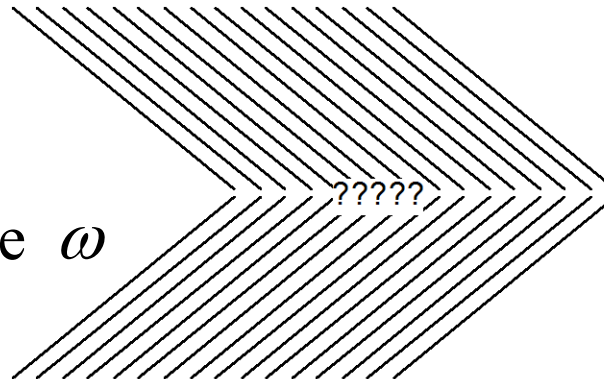
curvature wall

$$\omega \rightarrow \pi/2$$

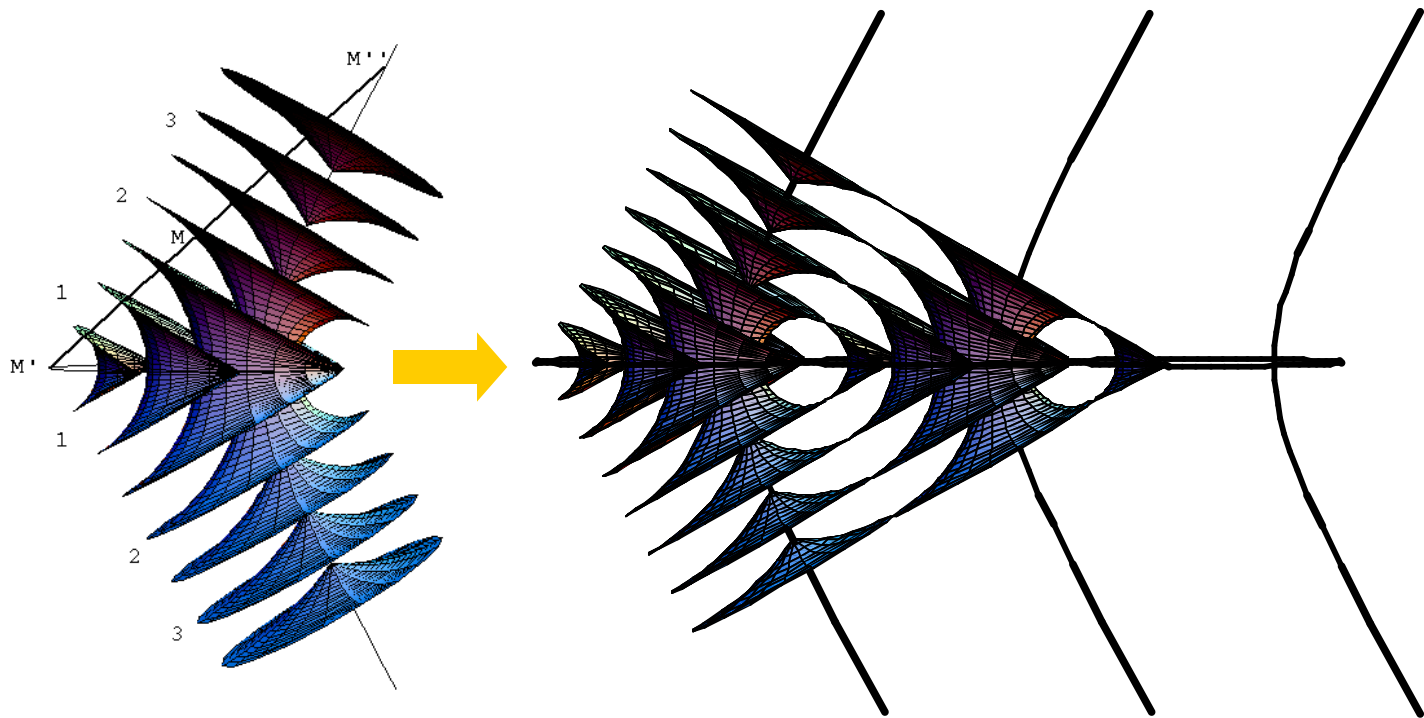
de Gennes, 1970



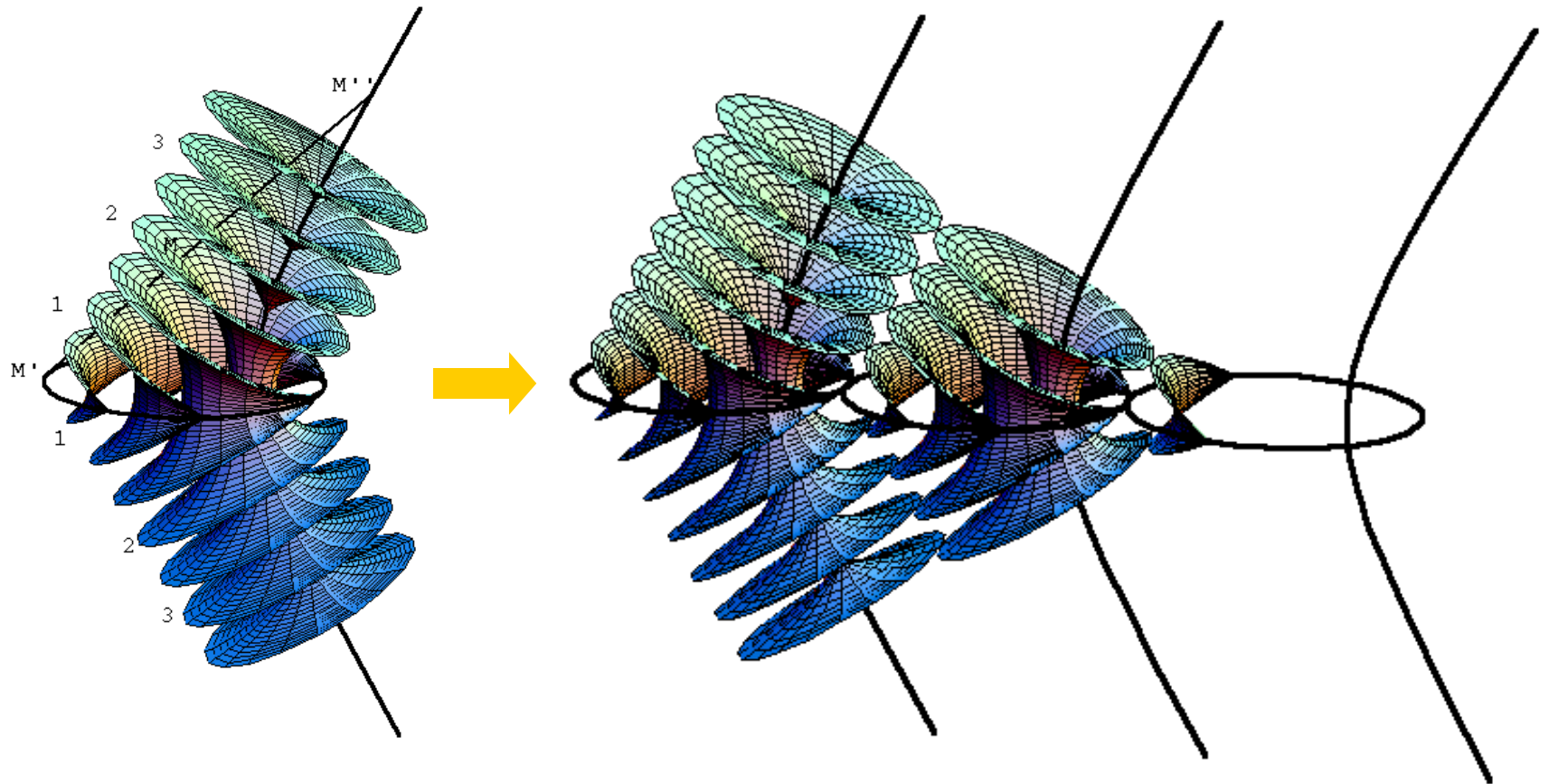
FCDs:  
intermediate  $\omega$

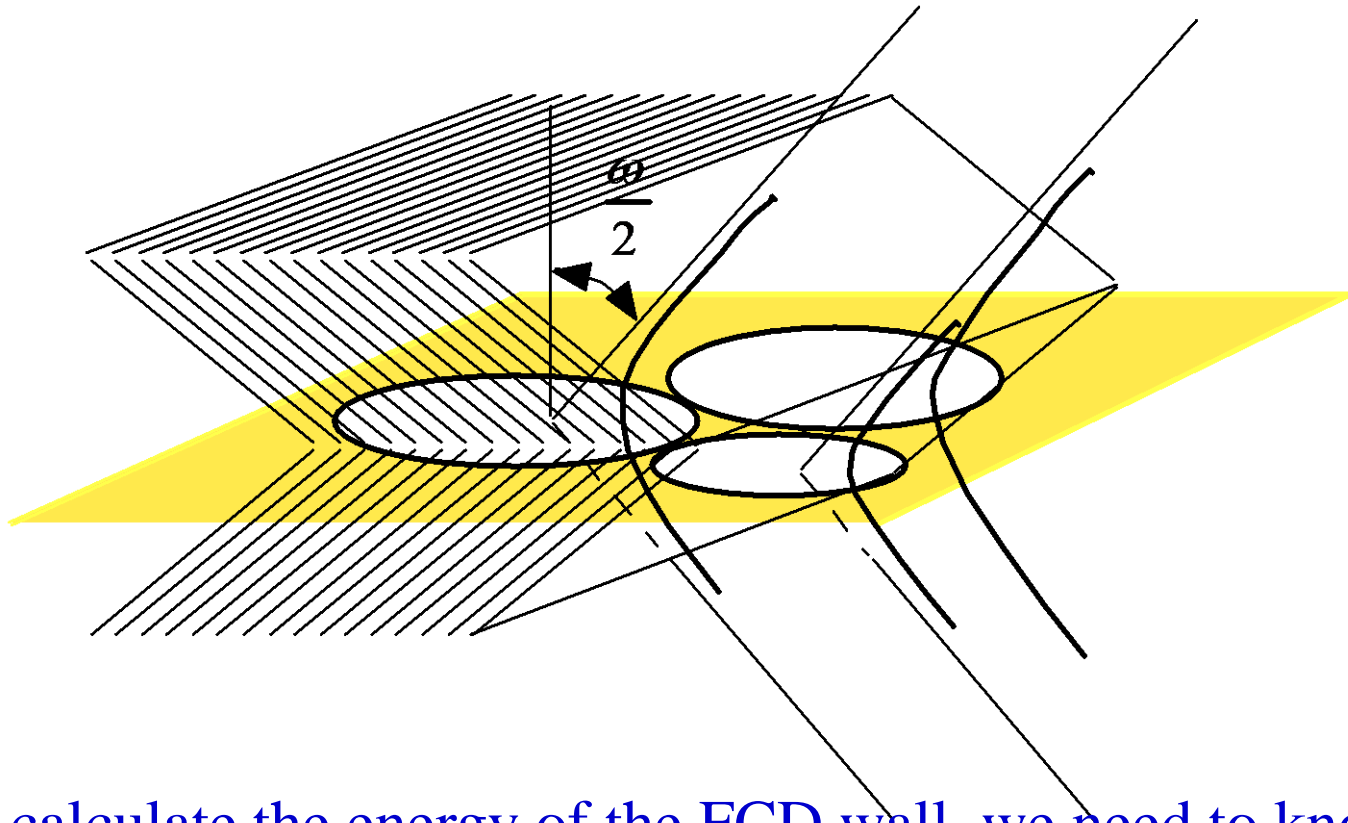


# FCD-filled Grain Boundary-Ist hint



# FCD-filled Grain Boundary-Ist hint



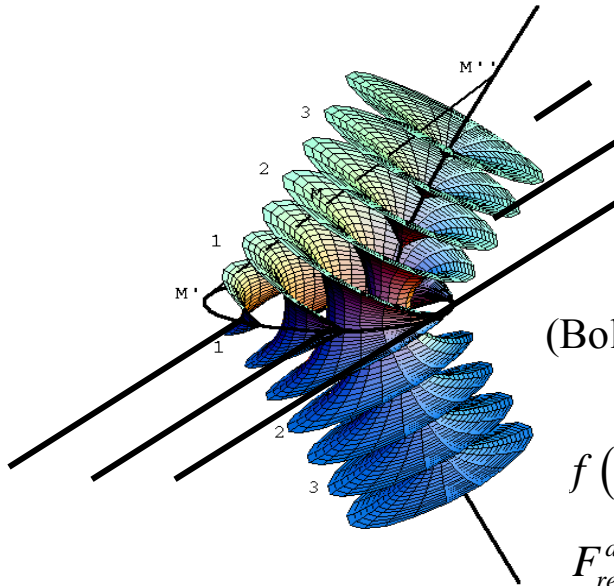


To calculate the energy of the FCD wall, we need to know:

- FCD energy (curvatures and defect cores)
- Spatial filling pattern, size distribution
- The energy of residual areas



# Residual Areas with Dislocations

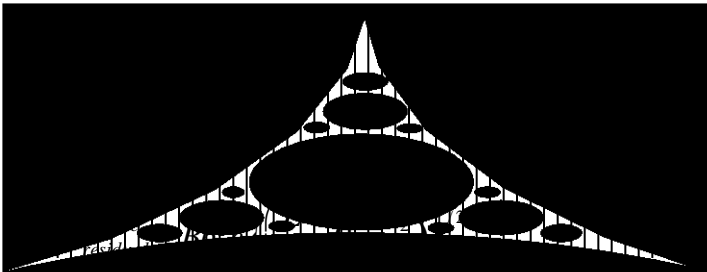


Dislocations with  
total Burgers vector  
 $b_u = 2ae$

(Boltenhagen et al, J. Physique (1991))

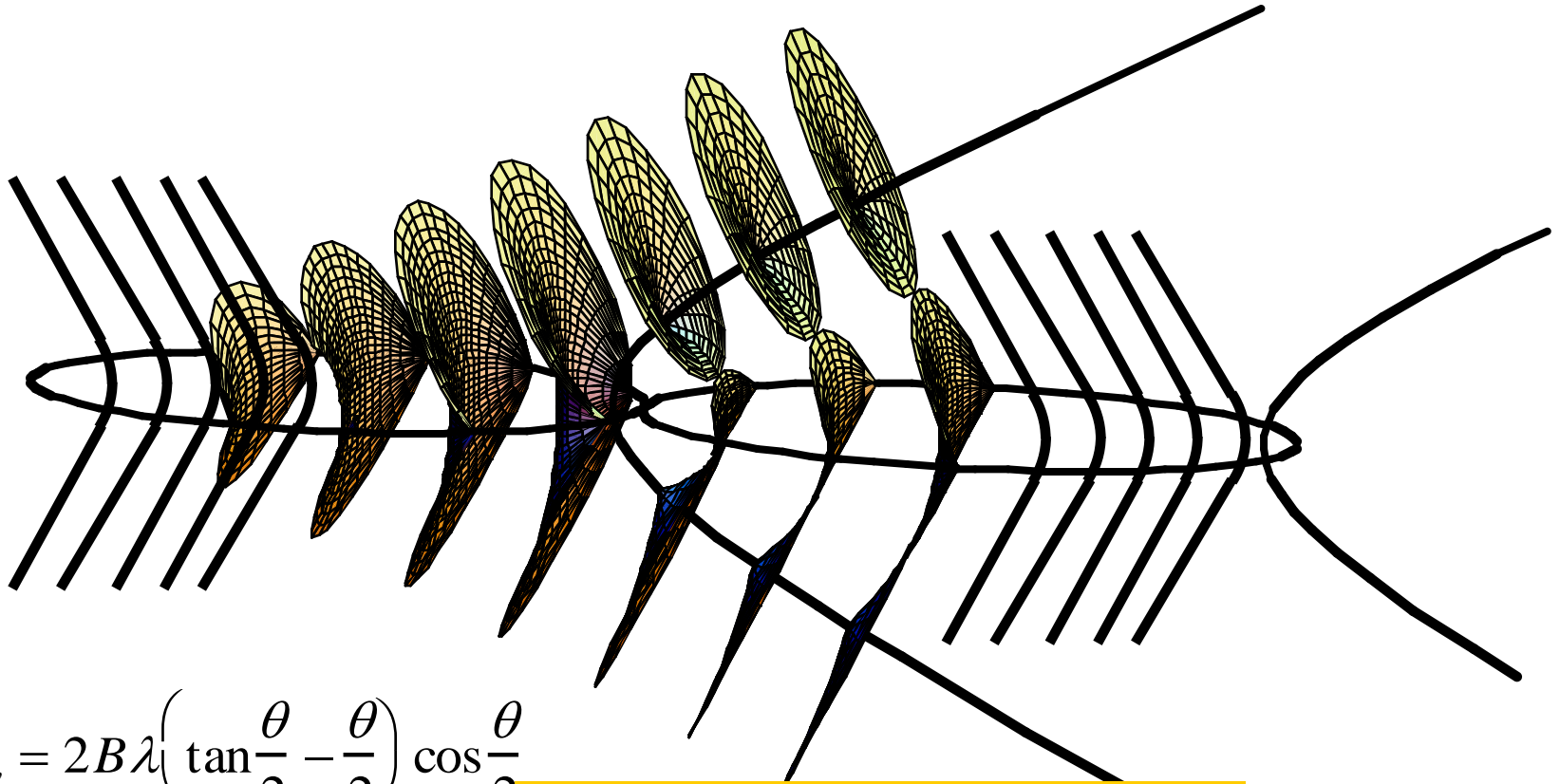
$$f \text{ (per unit length)} \sim Bb_u\lambda \sim Bae\lambda$$

$$F_{residual}^{disloc} = f \times b \times \text{Number of gaps} \sim Bae\lambda \times b \times \frac{\Sigma(b)}{ab} \sim Be\lambda\Sigma(b)$$



$$F_{residual}^{disloc} \sim Be\lambda b^2 \left(\frac{L}{b}\right)^n (1-e^2)^{-1/2}$$

# Residual Areas with Curvatures

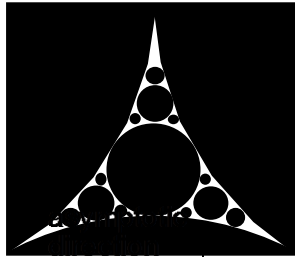


$$\sigma_{curv} = 2B\lambda \left( \tan \frac{\theta}{2} - \frac{\theta}{2} \right) \cos \frac{\theta}{2}$$

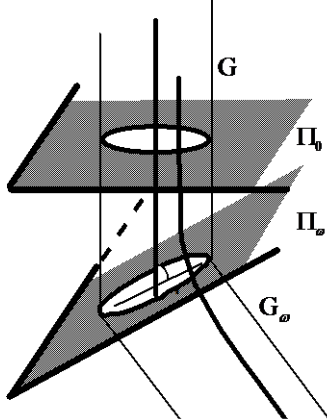
C. Blanc, M. Kleman, 1999

$$F_{resid}^{curv} \sim 2Bb^2\lambda \left[ 1 - \frac{e \operatorname{Arccos} e}{\sqrt{1-e^2}} \right] \left( \frac{L}{b} \right)^n$$

# From Circles to Ellipses



$$g(b) \sim \left(\frac{L}{b}\right)^n \quad P(b) \sim \left(\frac{L}{b}\right)^n b \quad \Sigma(b) \sim \left(\frac{L}{b}\right)^n b^2 \quad n \approx 1.32$$

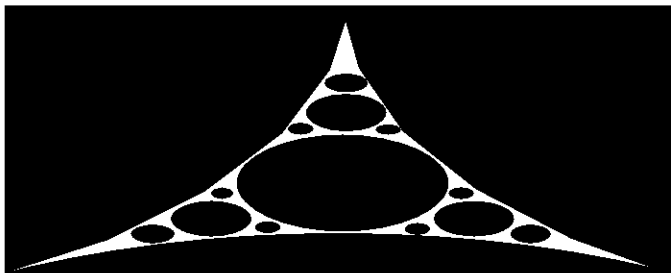


Projective properties of conic sections lead to:

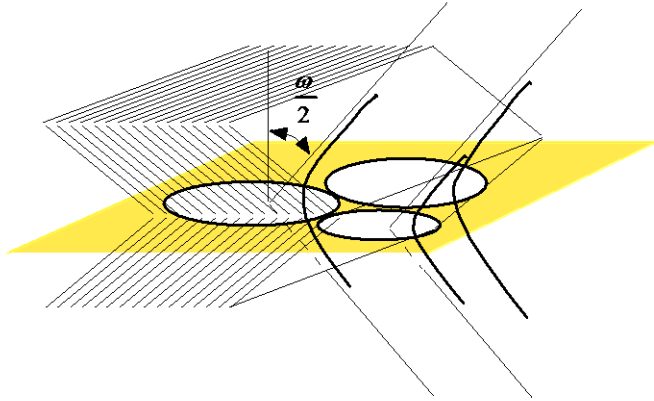
$$g(b) \sim \left(\frac{L}{b}\right)^n \quad n \approx 1.32$$

$$P(b) \sim \frac{\mathbf{E}(e^2)}{\sqrt{1-e^2}} \times \left(\frac{L}{b}\right)^n b$$

$$\Sigma(b) \sim \frac{1}{\sqrt{1-e^2}} \times \left(\frac{L}{b}\right)^n b^2$$



# FCD wall with Dislocations



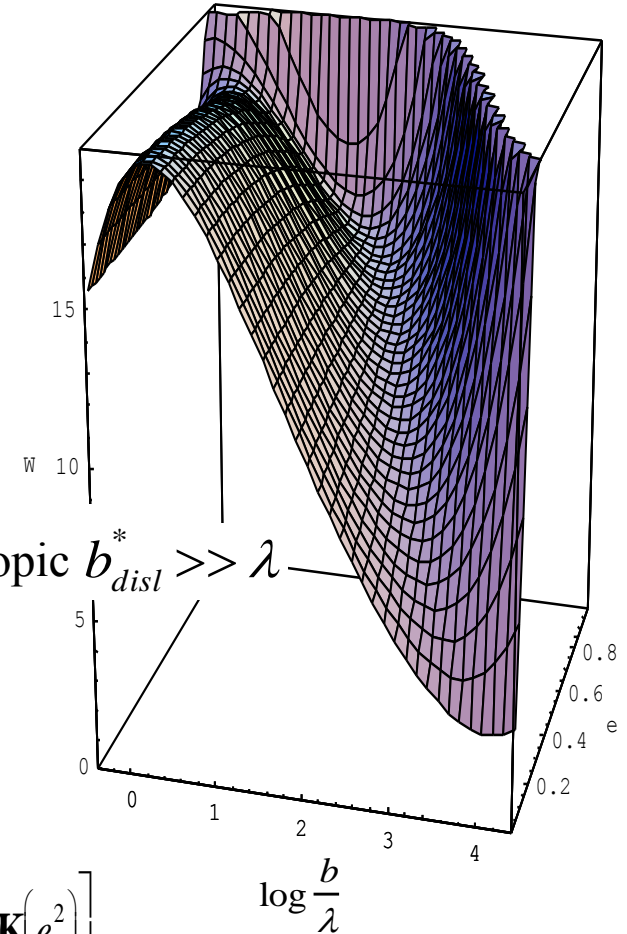
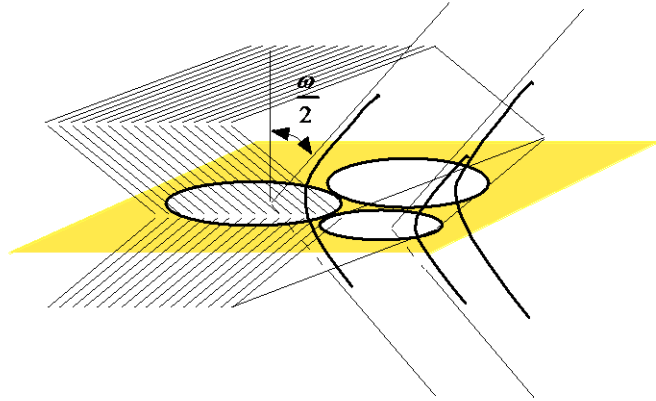
$$F = F_{bulk} + F_{core} + F_{residual}^{disloc}$$

$$F_{bulk} = - \int_{x=b}^L dg(x) f_{bulk}(x) \sim \alpha_b K_1 (1-e^2)^{1/2} \mathbf{K}(e^2) \int_b^L n \left[ \ln 2 \frac{x}{\lambda} - 2 \right] \left( \frac{L}{x} \right)^n dx$$

$$F_{core} = - \int_{x=h}^L dg(x) f_{core}(x) \sim \alpha_c K_1 (1-e^2)^{-1/2} \mathbf{E}(e^2) \int_b^L n \left( \frac{L}{x} \right)^n dx$$

$$F_{residual}^{disloc} = Be\lambda b^2 \left( \frac{L}{b} \right)^n (1-e^2)^{-1/2}$$

# FCD wall with Dislocations



$$\partial(F_{bulk} + F_{core} + F_{resid}) / \partial b = 0$$

$$\frac{b_{disl}^*}{\lambda} \sim \frac{1}{e} \frac{n}{2-n} \left\{ \alpha_b (1-e^2) \mathbf{K}(e^2) \left[ \ln \frac{2b_{disl}^*}{\lambda} - 2 \right] + \alpha_c \mathbf{E}(e^2) \right\}$$

$$\sigma_{FCD}^{disl}(e) \sim \frac{1}{(n-1)L^2} \left( \frac{L}{\lambda} \right)^n B \lambda^3 \left( \frac{\lambda}{b_{disl}^*} \right)^{n-1} (1-e^2)^{-1/2} \left[ e \frac{b_{disl}^*}{\lambda} + \alpha_b \frac{n}{n-1} (1-e^2) \mathbf{K}(e^2) \right]$$

# Grain boundaries with FCDs

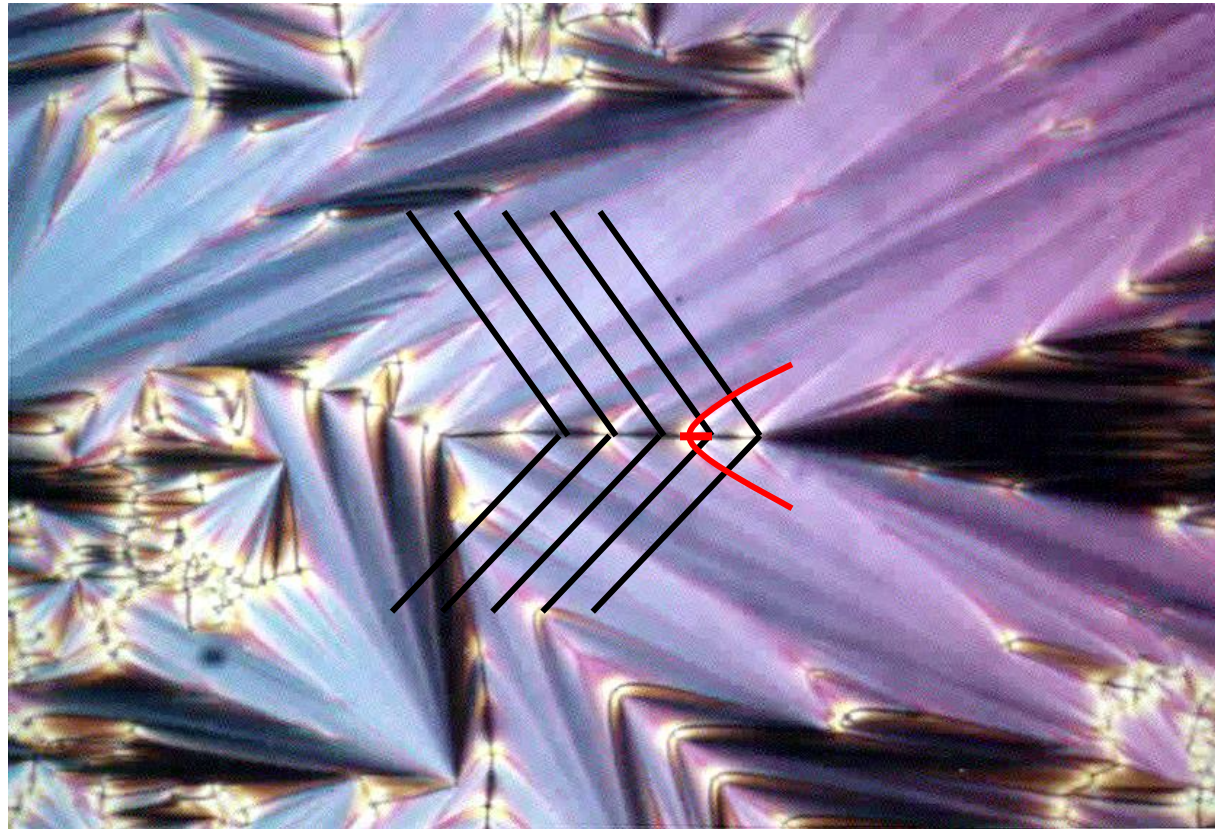
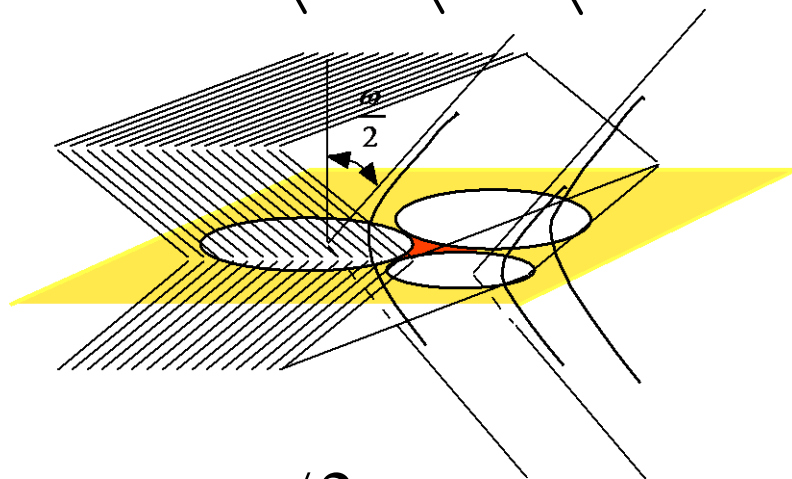
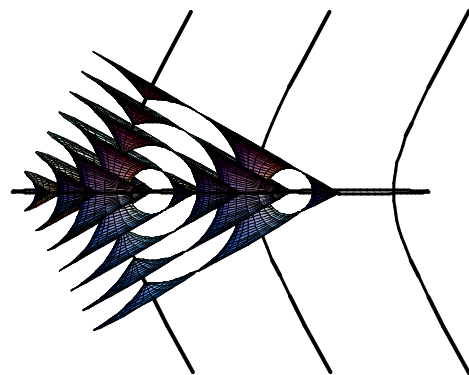


Photo: Claire Meyer



# Domain walls in SmA



$$\omega = 0$$

$$\omega = \pi/2$$

$$\omega = \pi$$

dislocations

FCDs, gaps with  
dislocations

FCDs, gaps with  
curvatures

curvatures

# Conclusions/What have you learned

## □ Lamellar phases

- Both compressibility and curvatures are generally important in weak elastic deformation, such as dislocations, surface perturbations, undulations
- Strong deformations such as focal conic domains are described sufficiently well by the curvature term; layer thickness is preserved everywhere except at singular lines (confocal pairs) that are remnants of singular focal surfaces
- Observation of focal conic domains led to correct identification of smectics as 1D periodic stacks of fluid 2D layers
- Focal conic domains participate in relaxation of surface anchoring and grain boundaries