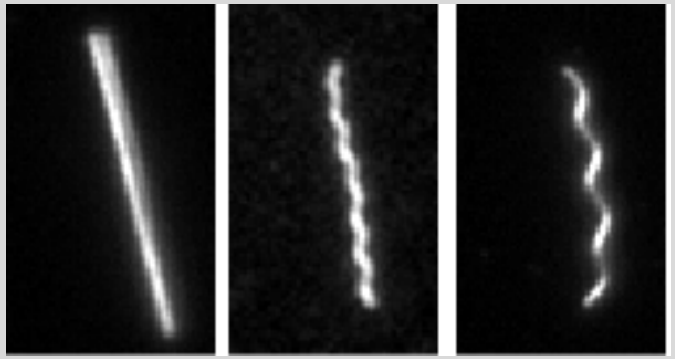
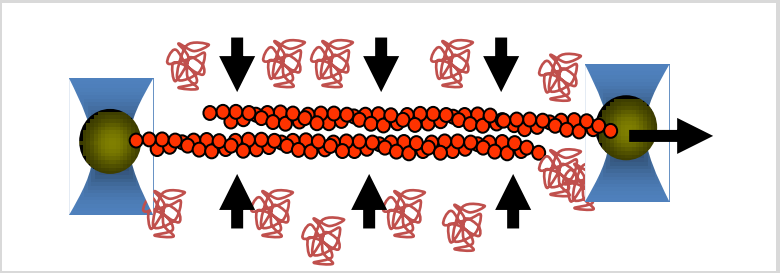
Self-assembly through chiral control of interfacial tension

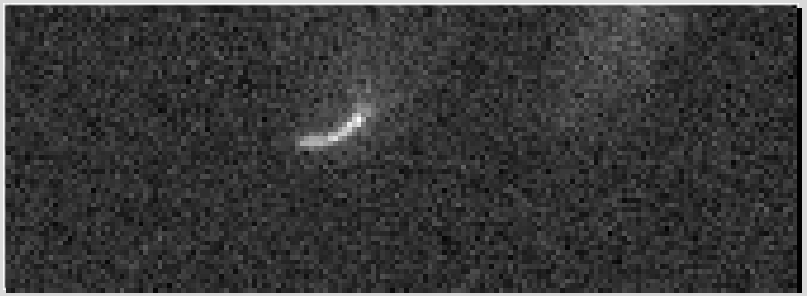
Zvonimir Dogic
Department of Physics
Brandeis University

Dynamics and assembly of helical filaments

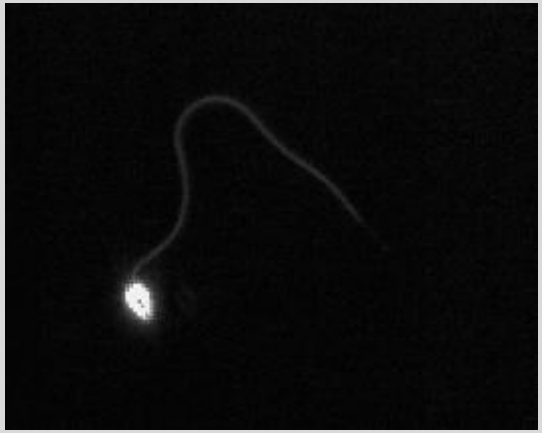


Sliding friction between biopolymers

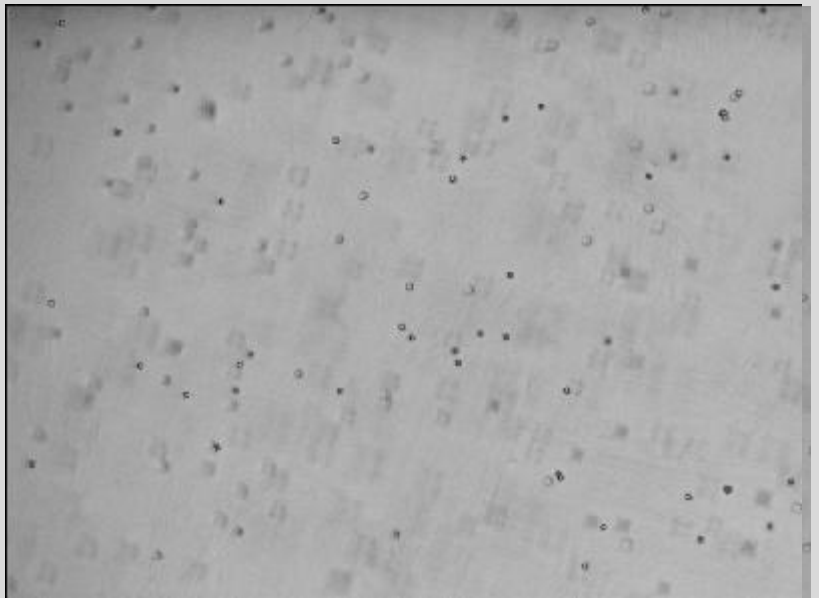




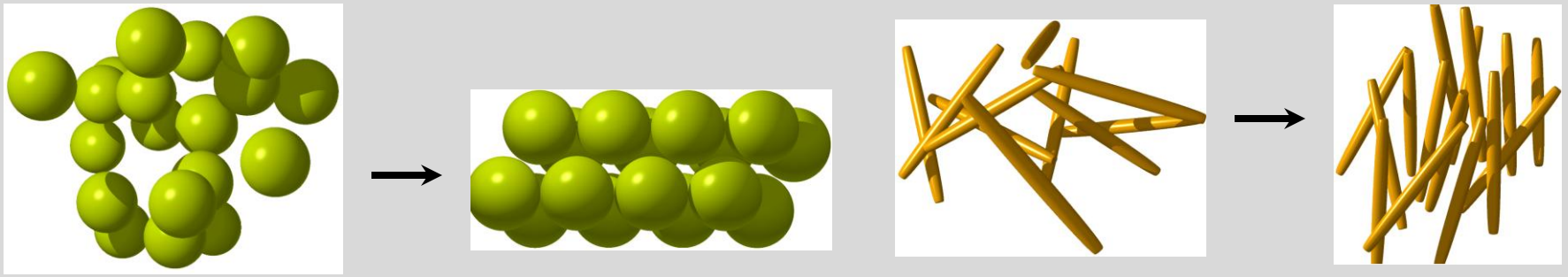
Flagellar beating *in vivo* and *in vitro*



In Vitro cytoplasmic Streaming



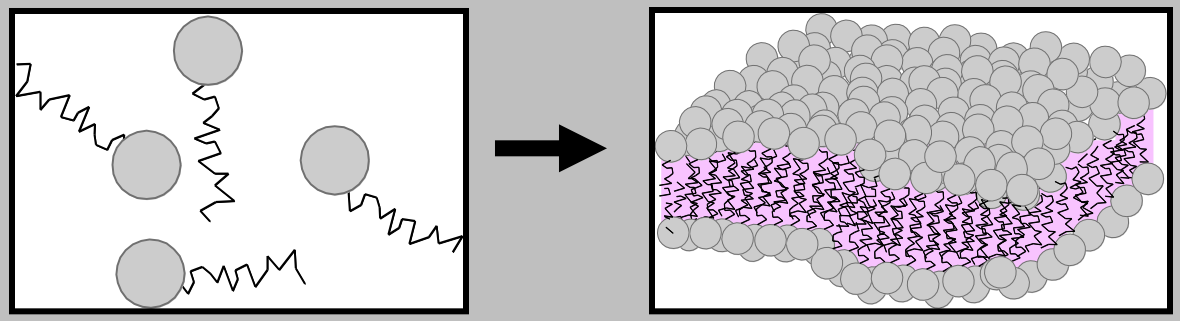
Colloidal spheres/rods → colloidal crystals, liquid crystals …



liquid crystal isotropic nematic

Assembly of 3D structures !

Amphiphilic self-assembly → self-limited structures, i.e. 2D membranes, 1D wormlike micelles, spherical micelles …



Assembly of self-limited structures from homogenous particles?

Entropy and chirality!

Acknowledgements

Mike Hagan
Brandeis University

Robert Meyer
Brandeis University

Daniela Nicastro
Brandeis University

Rudolf Oldenbourg
MBL

Robert Pelcovits
Brown University

Thomas Gibaud
Brandeis University

Mark Zakhary
Brandeis University

Edward Barry
Brandeis University

MRSEC
Materials Research Science Engineering Center
Brandeis Univeristy
NSF

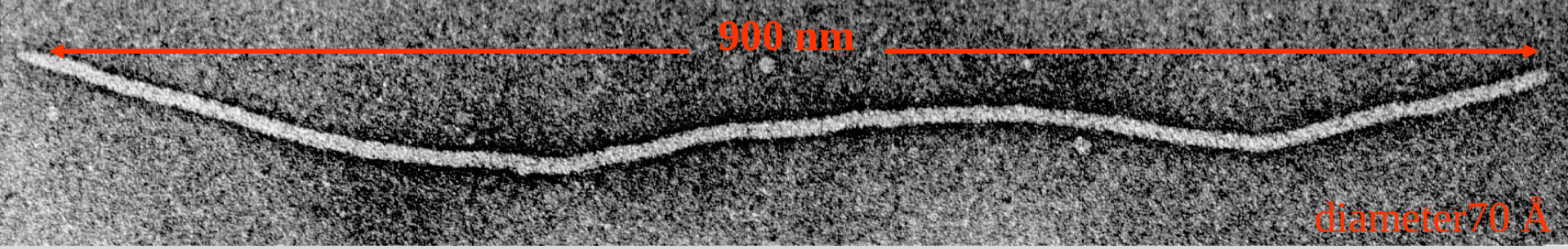
Outline

Part I: Entropy drive self-assembly of 2D non-amphiphilic membranes

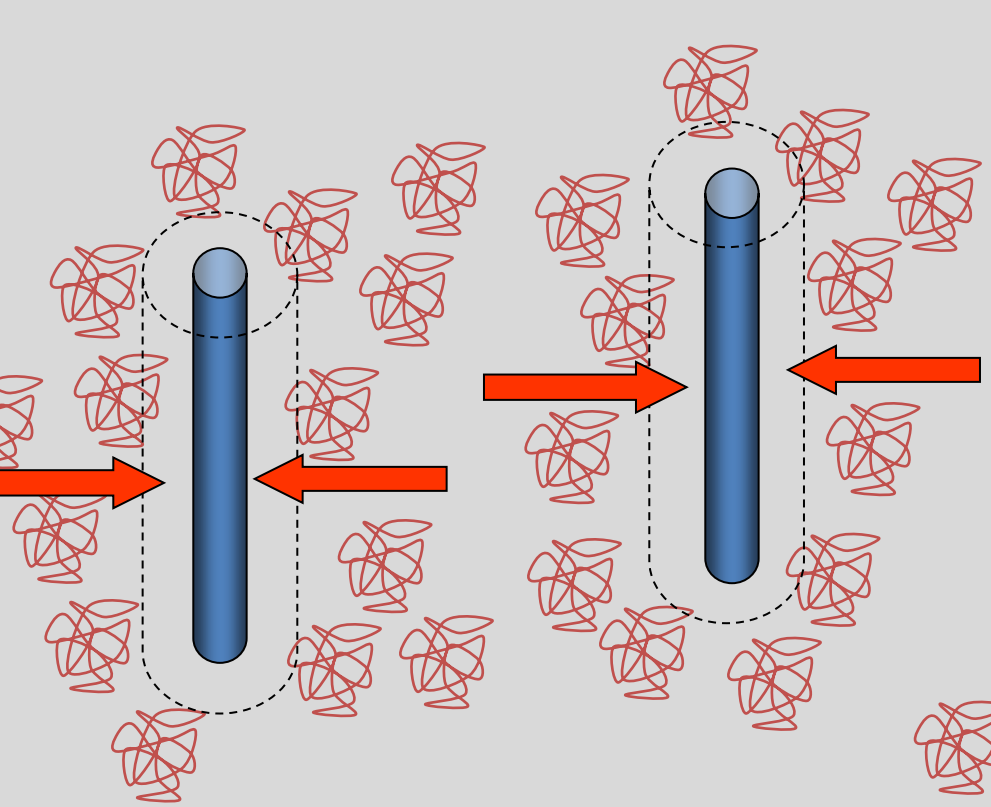
Part II: Structure and energetics of an exposed edge -> chiral control of line tension

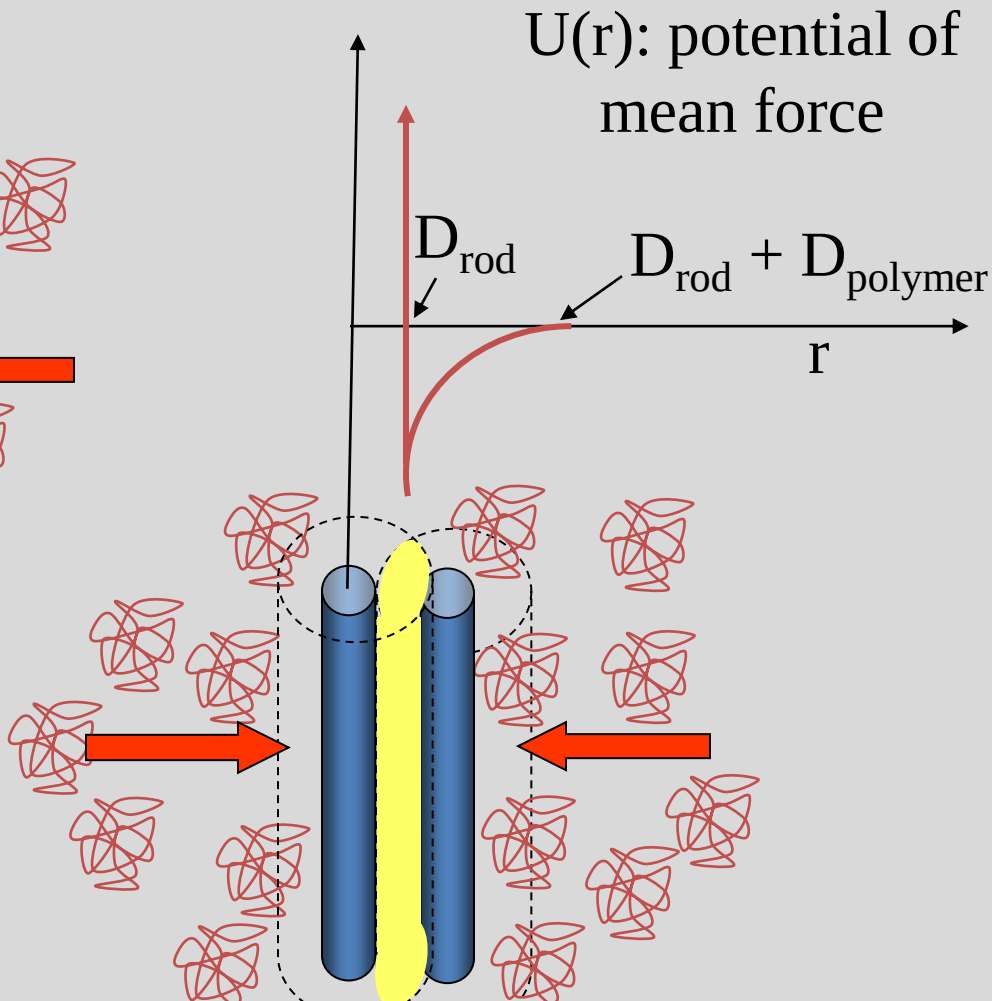
Part III: Structure and interactions of π-wall defect in 2D membranes

Part I:
Entropic self-assembly of 2D membranes



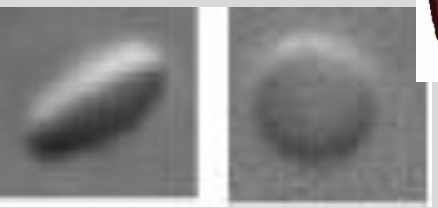
Depletion: Attraction through Repulsion



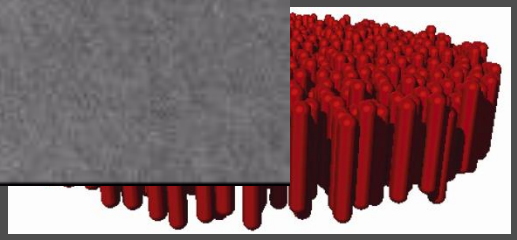


entropy induced effect

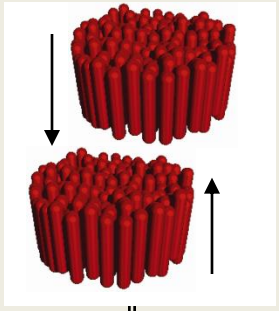
monolayer nuclei

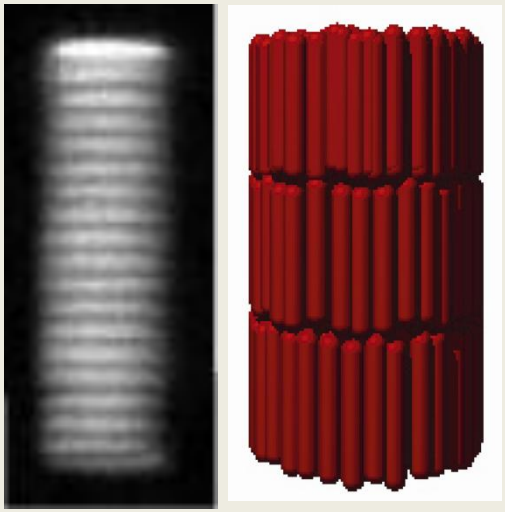


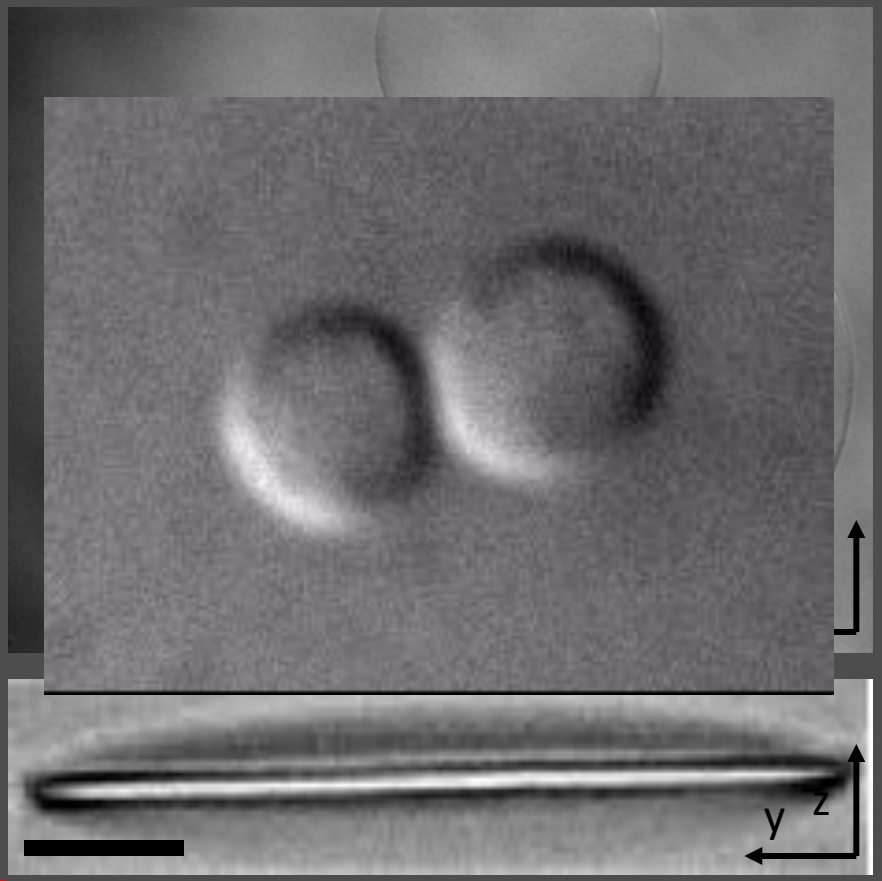
membranes



I. smectic filaments

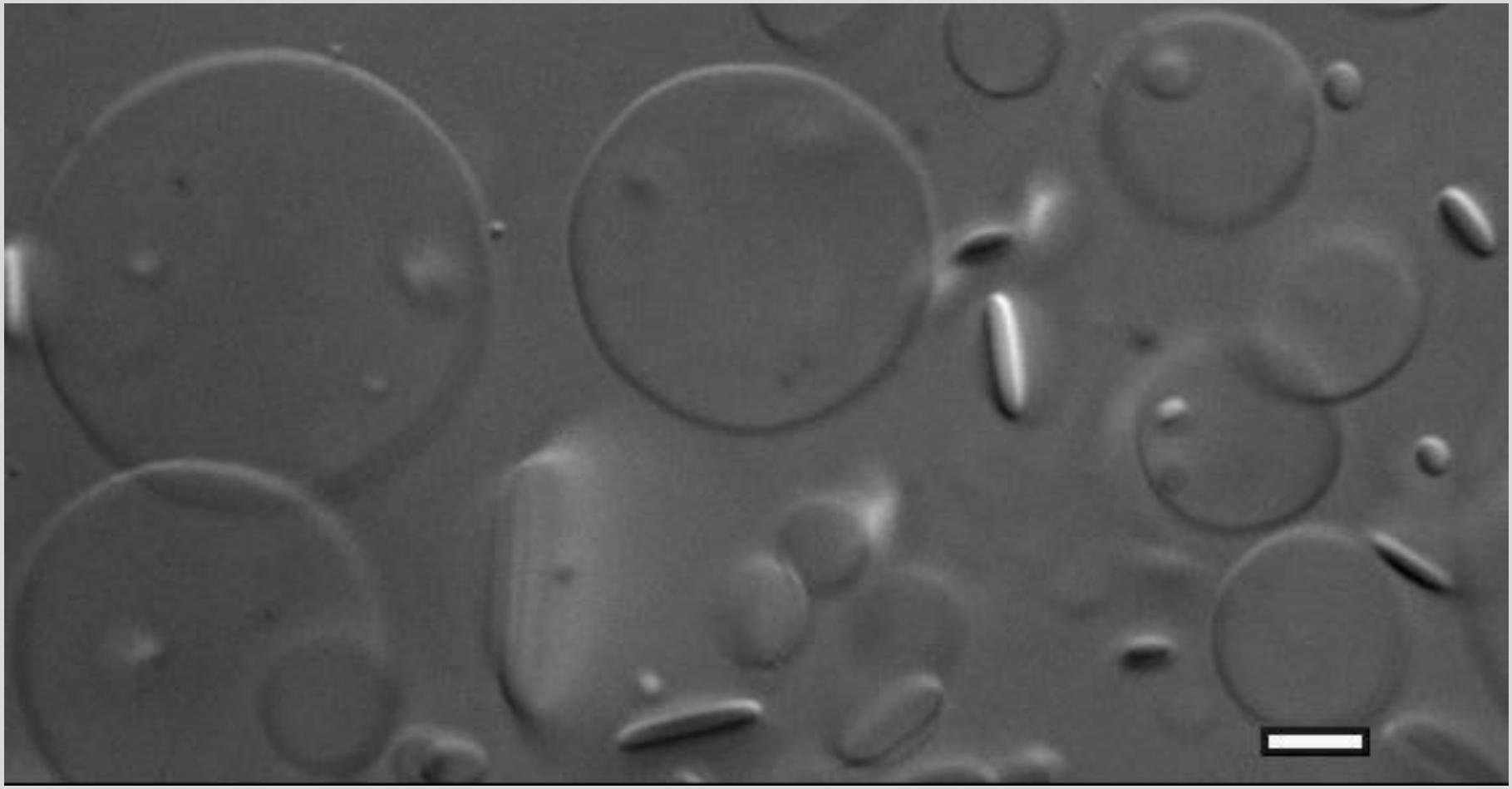




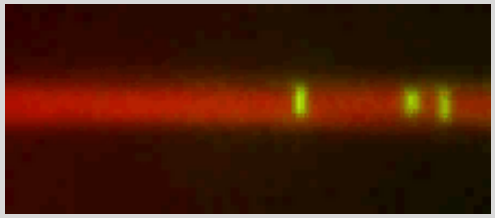


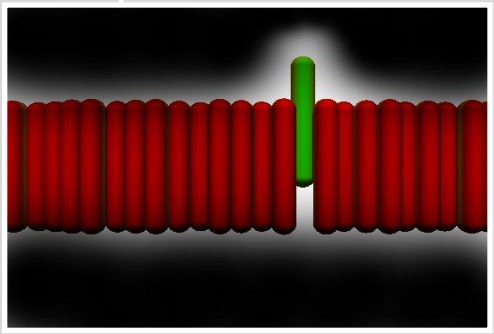
Schilling and Frenkel, Phys. Rev E 2003

Colloidal Membranes

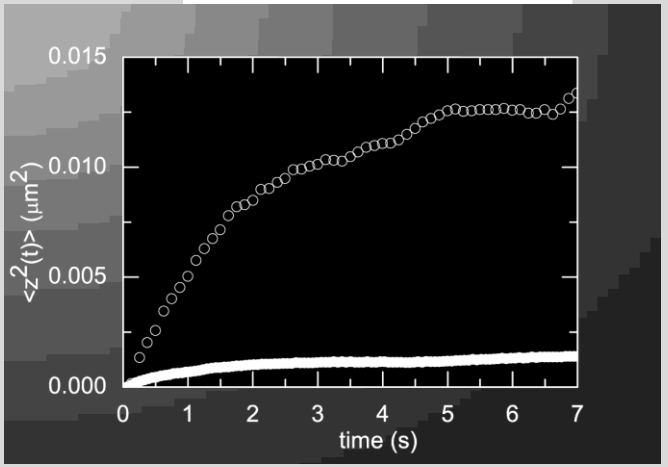


Colloidal Membranes





Molecular Protrusions

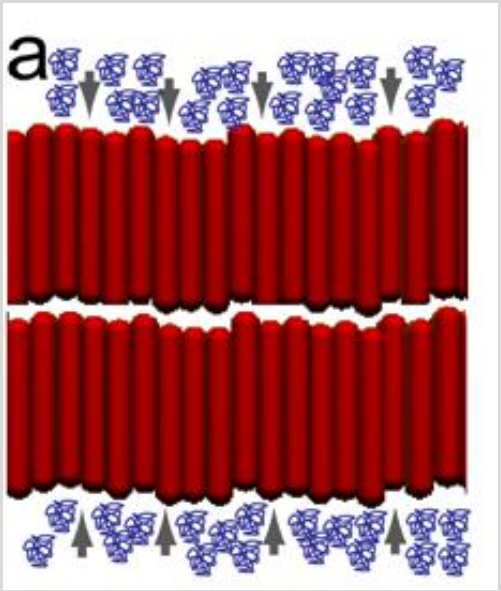


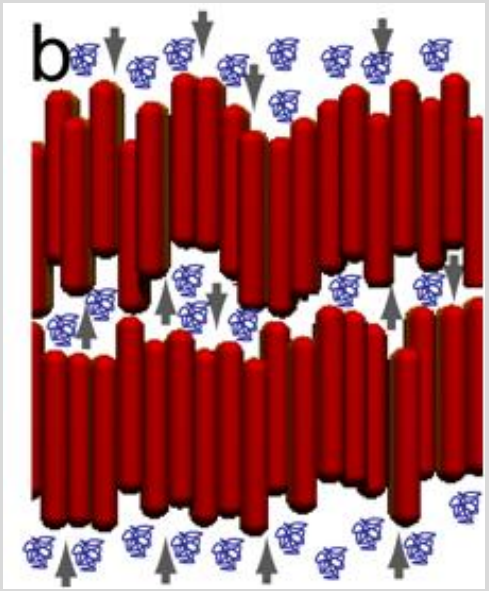
Protrusion Fluctuations suppressed in a stack

→**high polymer concentration**

smooth surface

attractive interactions





→ **low polymer concentrations**

rough surface

repulsive interactions

Conclusions
Part I:
Assembly of colloidal membranes

→ **continuum properties analogous to lipid bilayers**

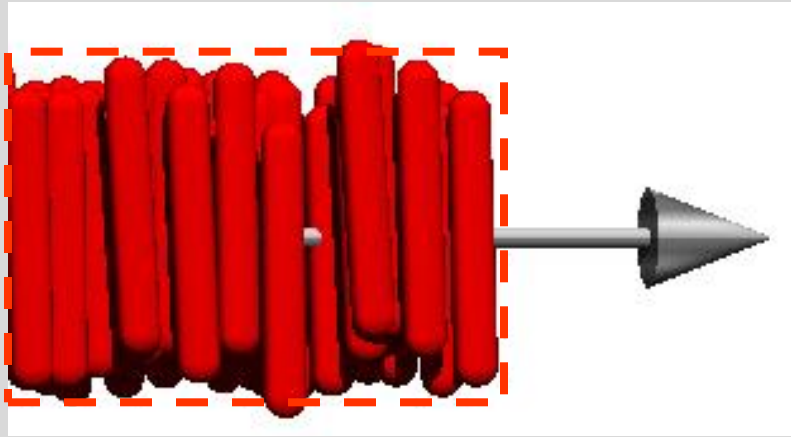
→ **self-assembly of 2D surface from homogeneous molecules**

→ **entropy driven phenomena**

Part II:
edge of colloidal membranes

Structure of the membrane's edge (achiral rods)

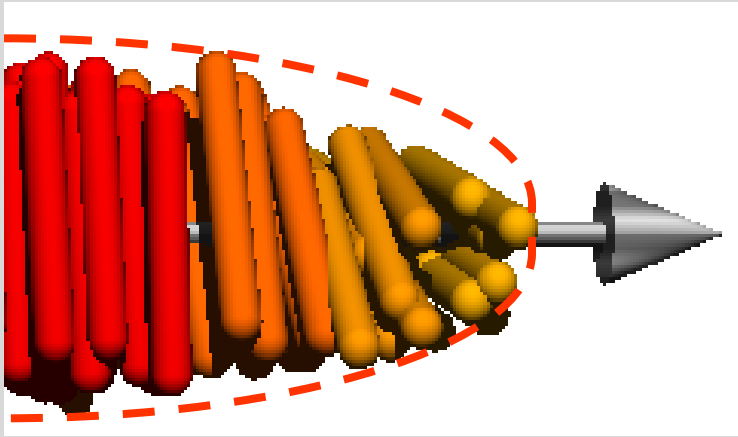
Edge configuration 1



→ large interfacial area

→ no elastic distortion

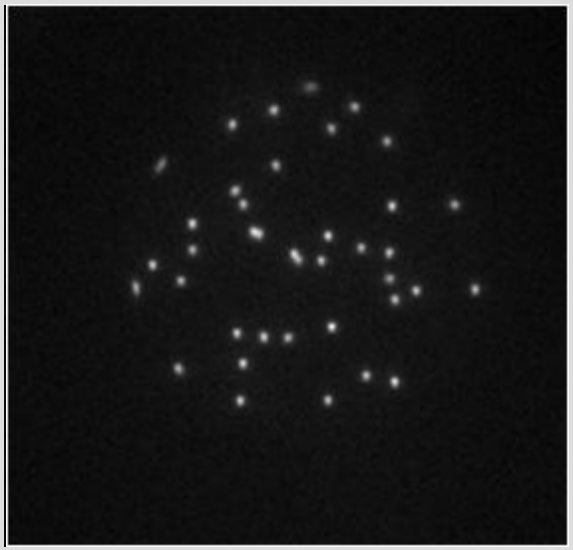
Edge configuration 2



→ small interfacial area

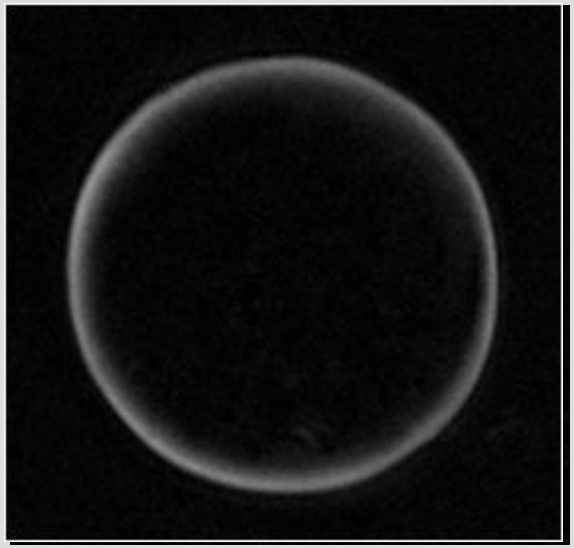
→ large elastic distortion

Birefringence map of colloidal membranes



fluorescence microscopy

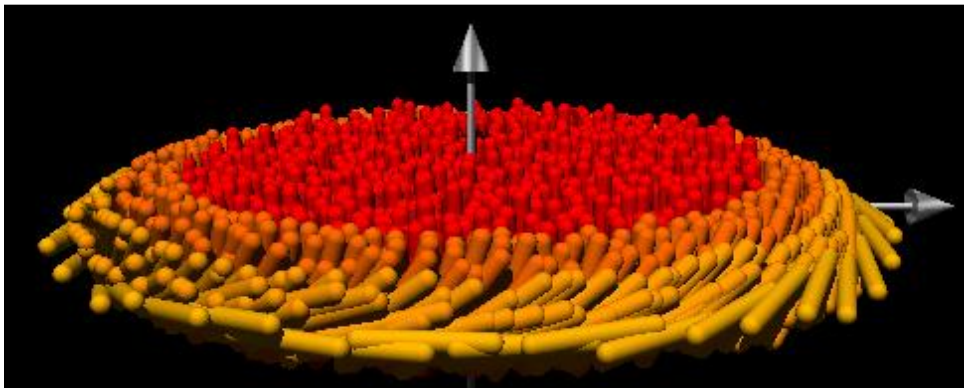
molecular lengthscale



Quantitative PolScope

director field

LC-PolScope – pixel brightness corresponds to 2D projection of the local birefringence



Twist penetration length in 2D membranes

1. **Theoretical analysis – half plane**

θ(x) – local tilt of the molecules

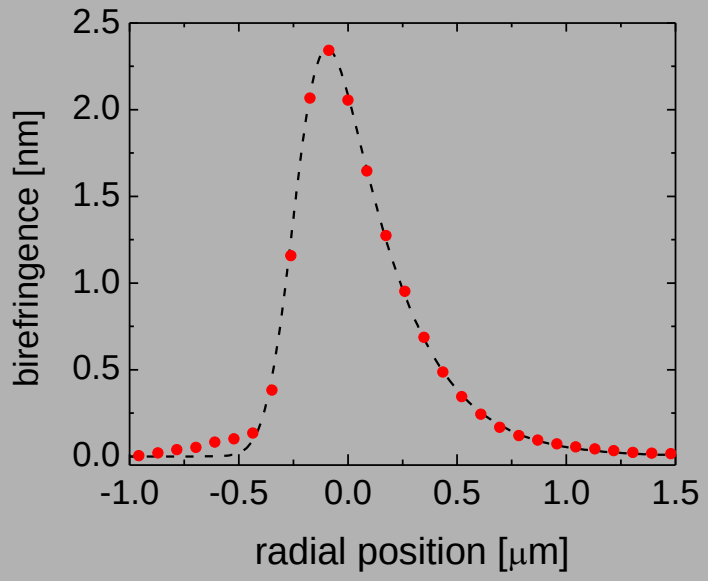
$$F = \int_0^{\infty} \left[K_{22} \left(\frac{d\theta}{dx} \right)^2 + C \sin^2 \theta \right] dx$$

$$\theta(x) = 2\,\mathrm{ArcTan}[\tan(\theta_O) e^{-x/\xi}]$$

$$\xi = \sqrt{\frac{K_{22}}{C}}$$

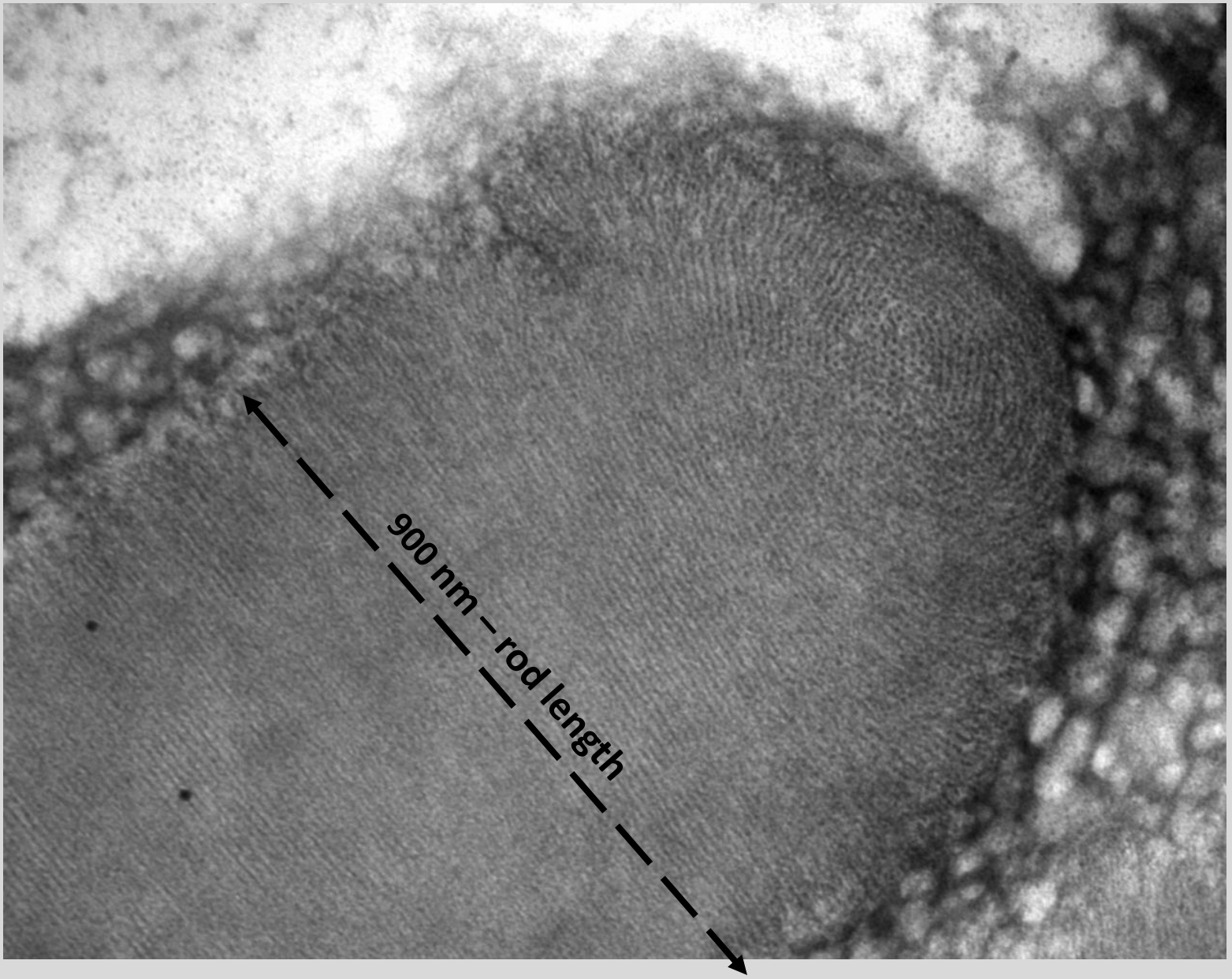
- ξ is analogous to London penetration depth in superconductors (deGennes)

2. Theory & Experiment

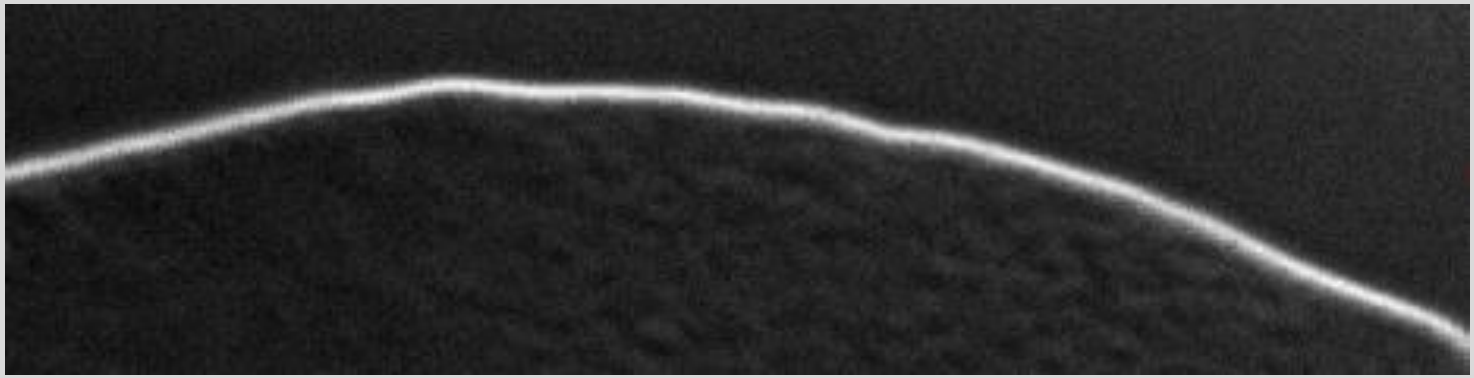


- twist penetration length 0.48 μm

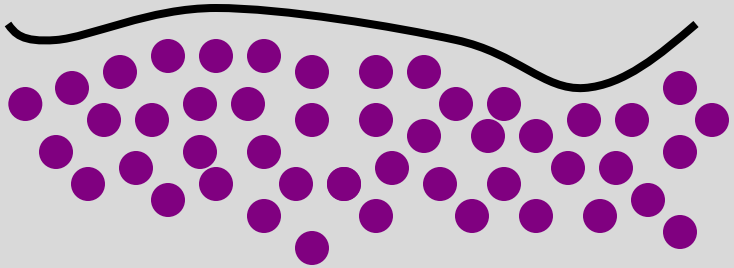
Electron microscopy of membrane's edge



Fluctuations of the edge → line tension



$h(x)$

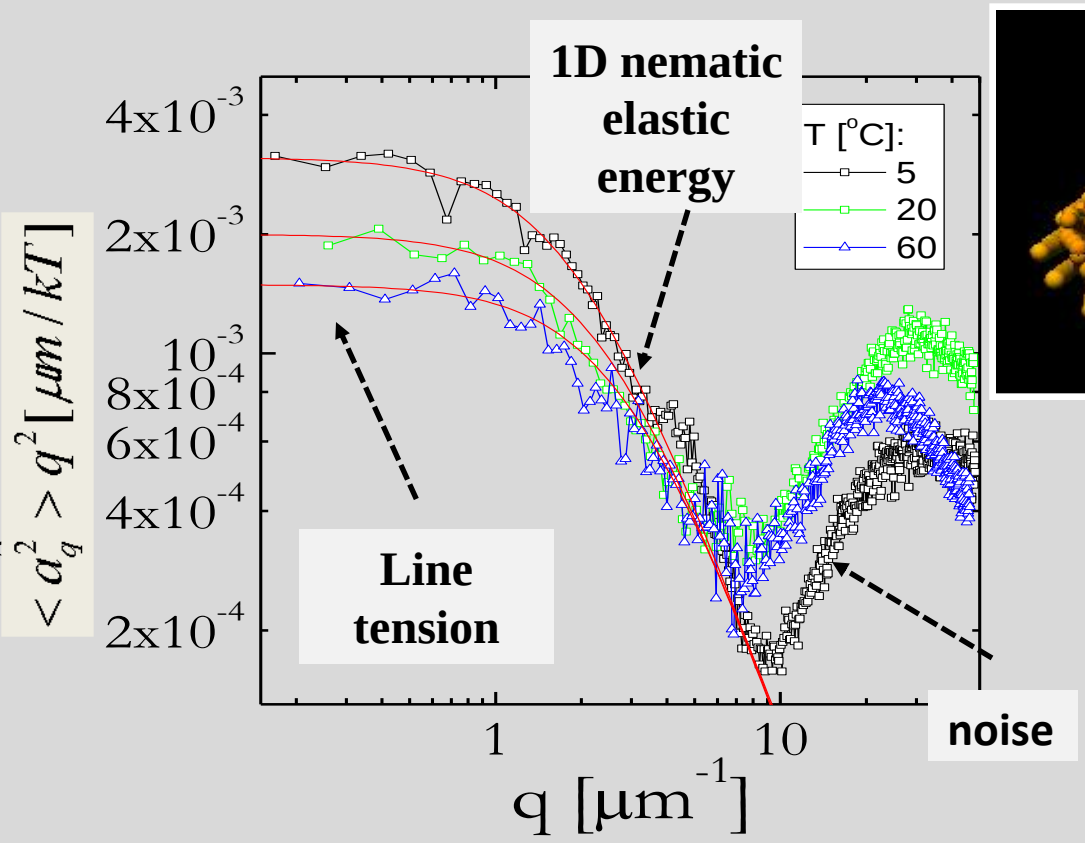


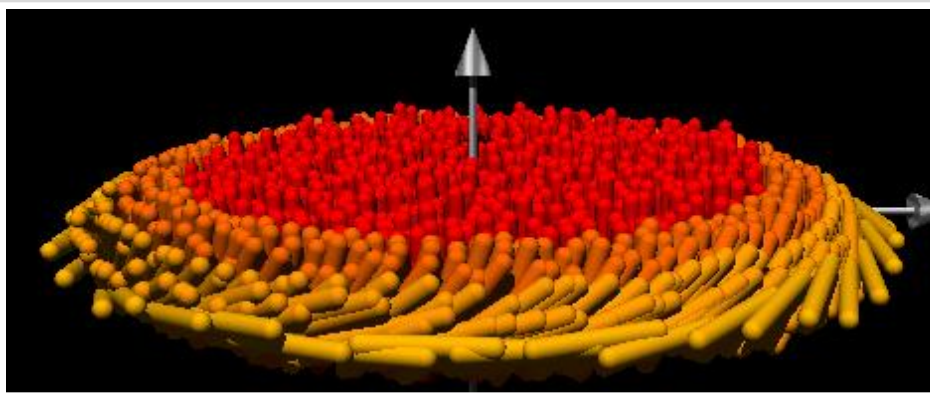
$$h(x) = \sum_q a_q \cos(qx)$$

$$F_L = \gamma \int dL \approx \gamma \int \left(1 + \frac{1}{2} h'(x)^2\right) dx$$

$$\left\langle a^2{}_q \right\rangle q^2 = \frac{k_B T}{\gamma}$$

Fluctuations spectrum of membrane's edge





$$\langle a^2_q \rangle q^2 = \frac{k_B T}{c_1 + c_2 q^2}$$

line tension

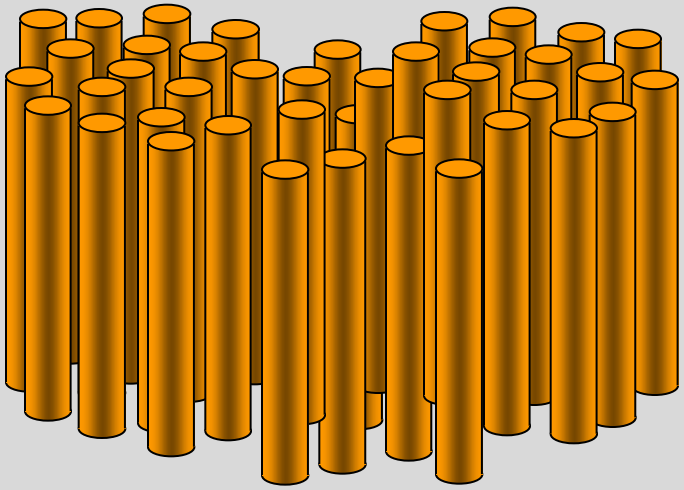
frank elastic constant

Small q limit
Fluctuations dominated by surface tensions

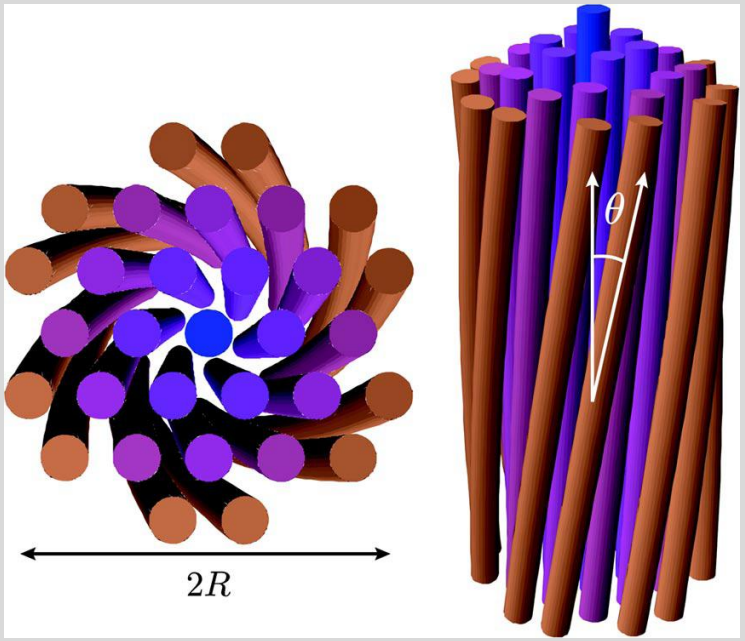
large q limit
fluctuations dominated by bending elasticity

Fluctuation spectrum yields line tension and the Frank elastic constant

Formation of a monolayer is incompatible with chirality



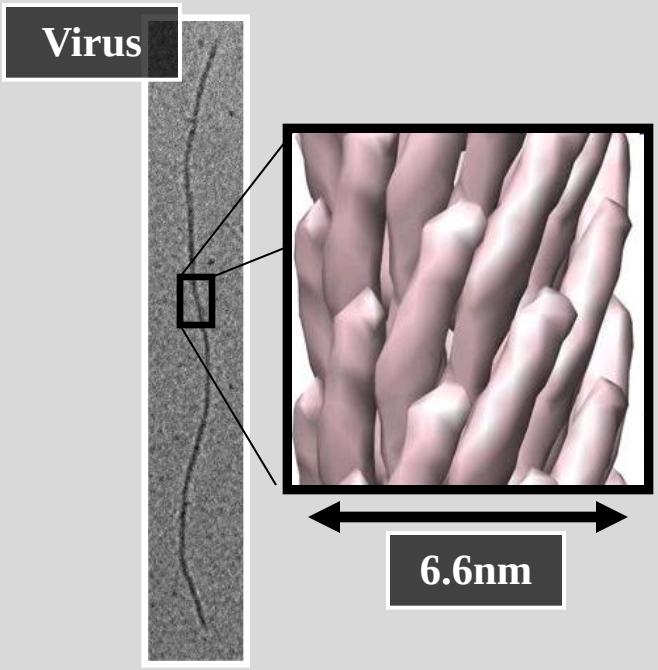
achiral monolayers
indefinete growth

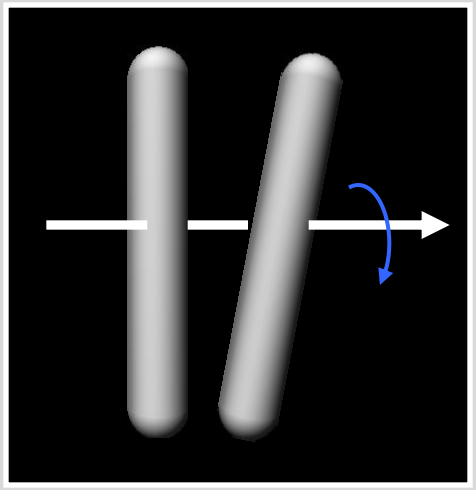


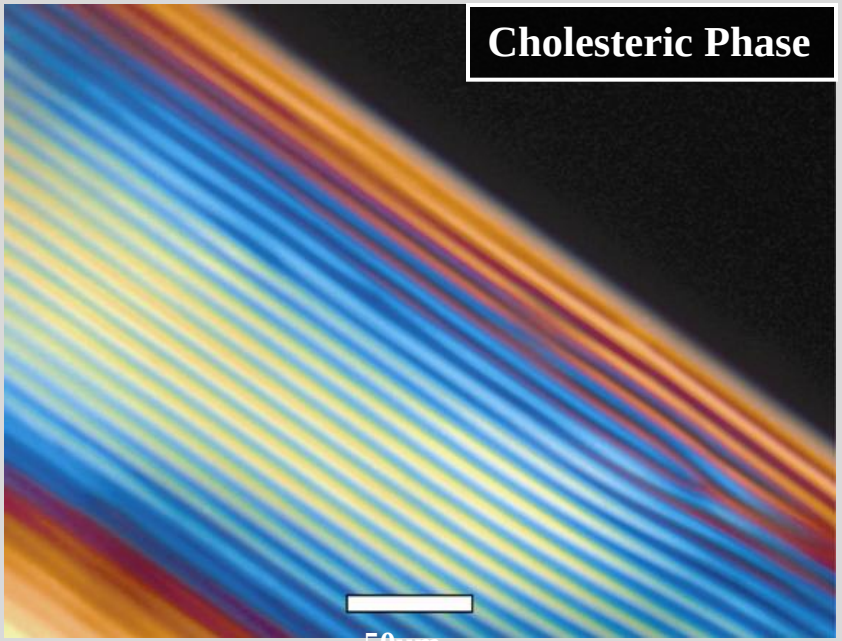
Monolayers with chiral rods
frustration

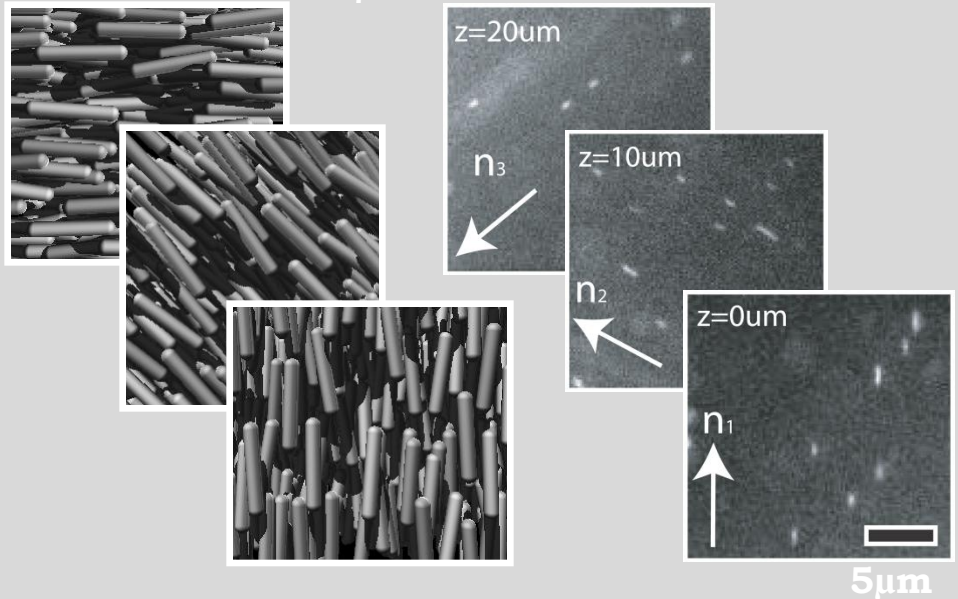
Grason and Bruinsma PRL, 2007
Selinger, Spector and Schnur, J. Chem Phys B 2001

Filamentous viruses can be chiral





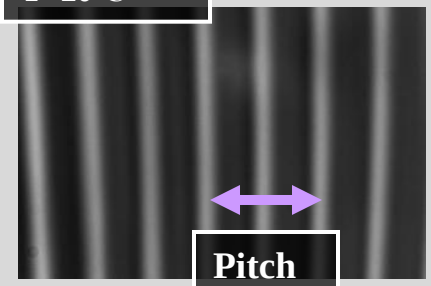




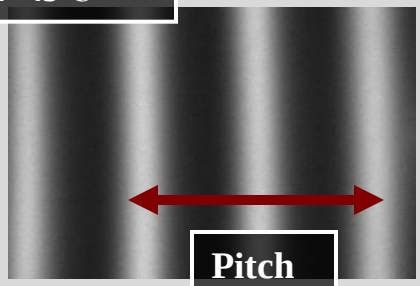
Chirality is temperature dependent

50µm

T=20˚C

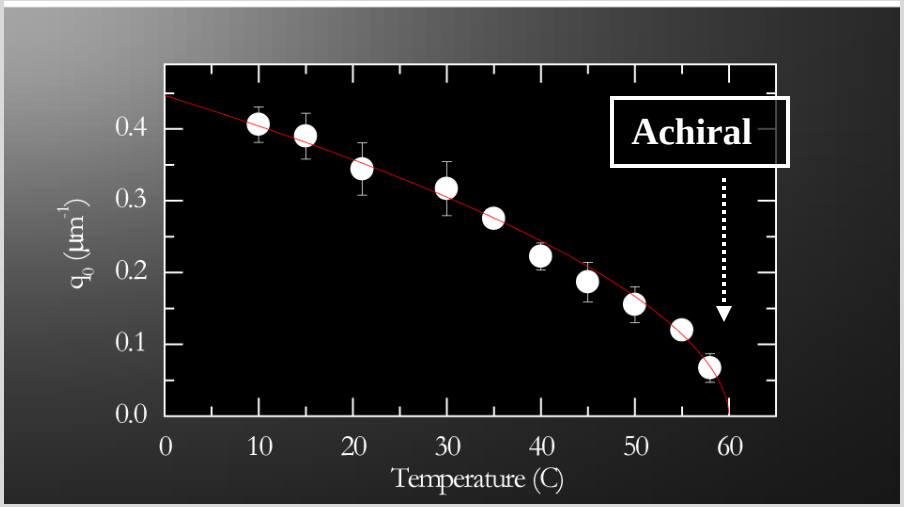


T=45˚C



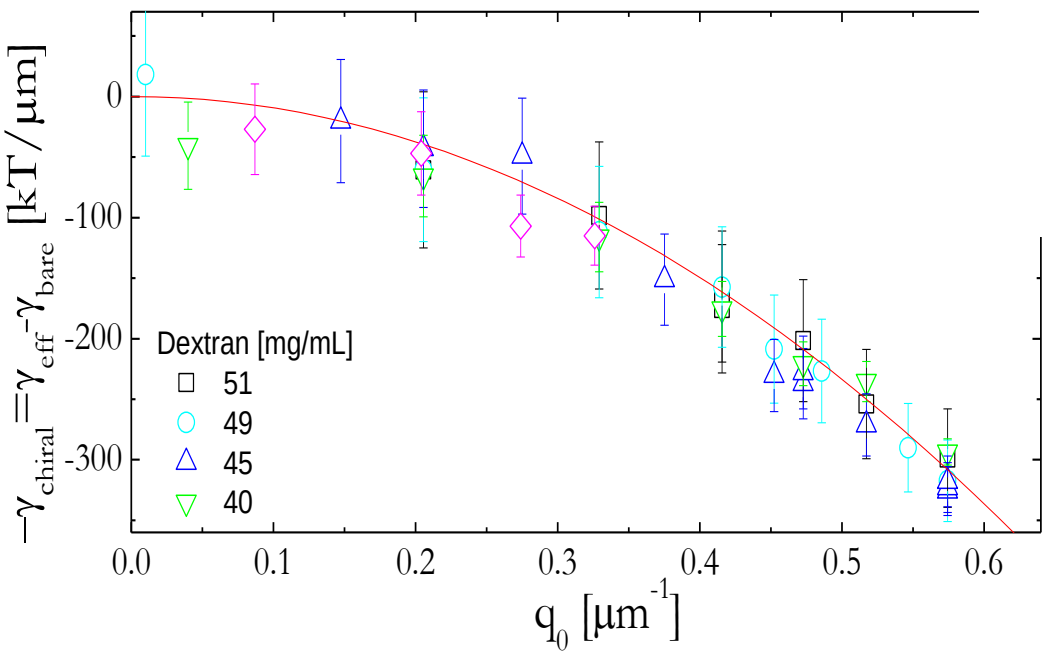
$$q_o = \frac{2\pi}{Pitch}$$

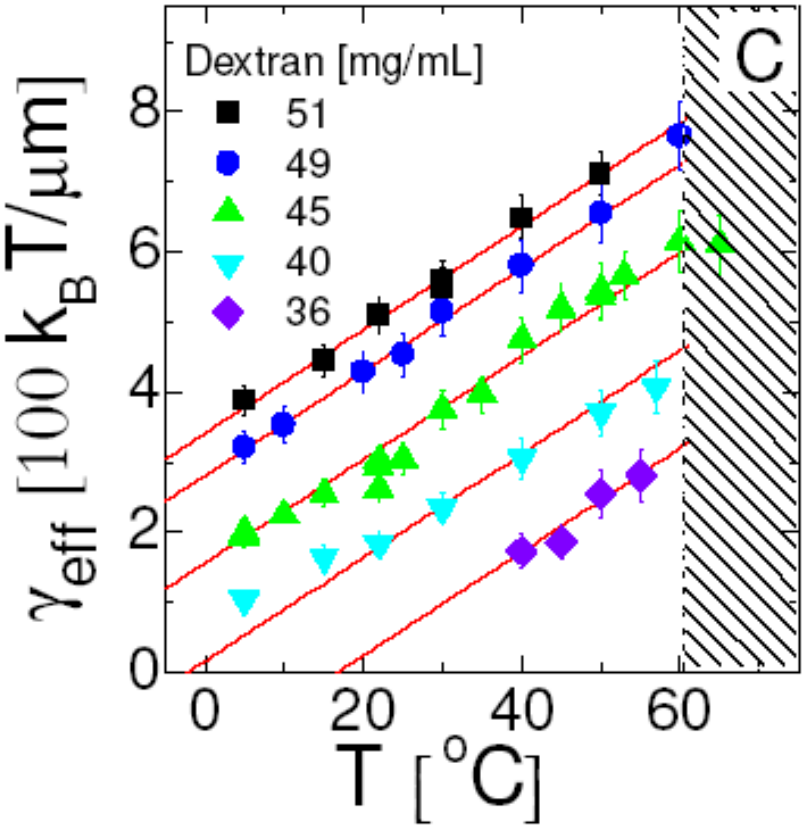
5µm



Chirality controls line tension !

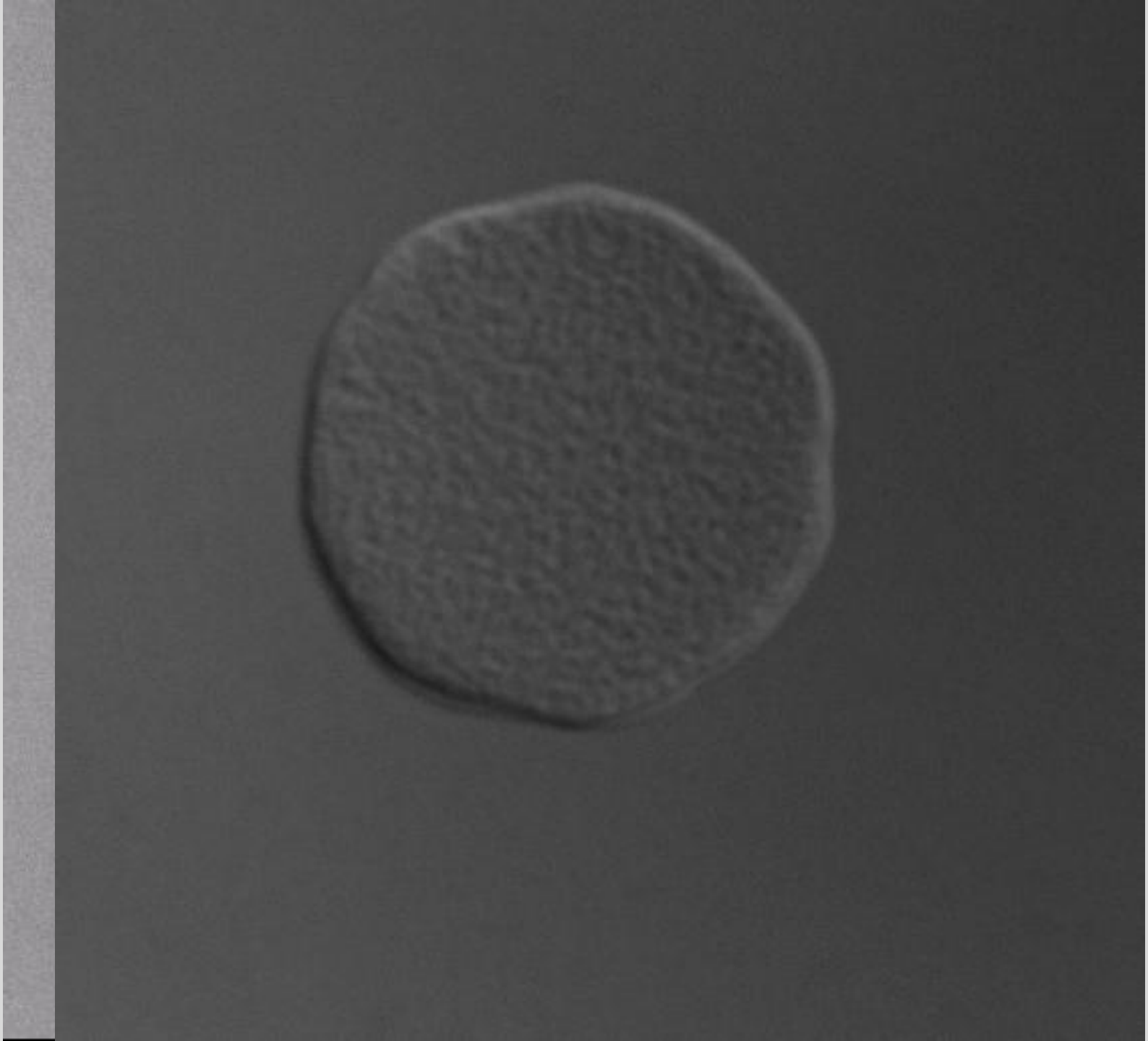
$$\gamma_{eff} \approx \gamma_{bare} - \gamma_{chiral}$$





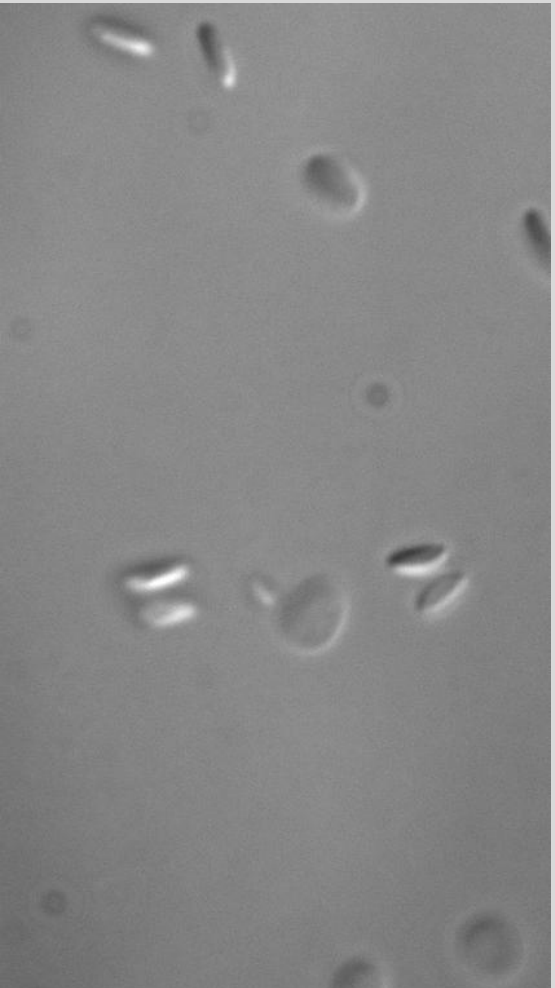
γ_{chiral} independent of dextran concentration

Can γ_{efff} be zero?

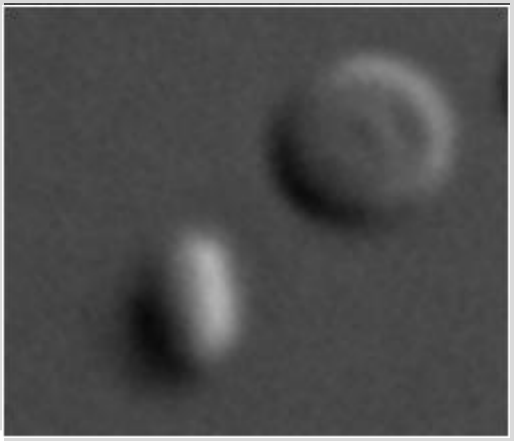


Self-assembly and growth of ribbons

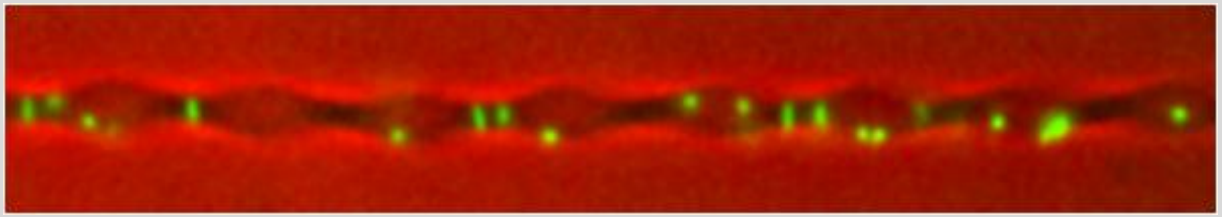
1. Metastable self-limited disks

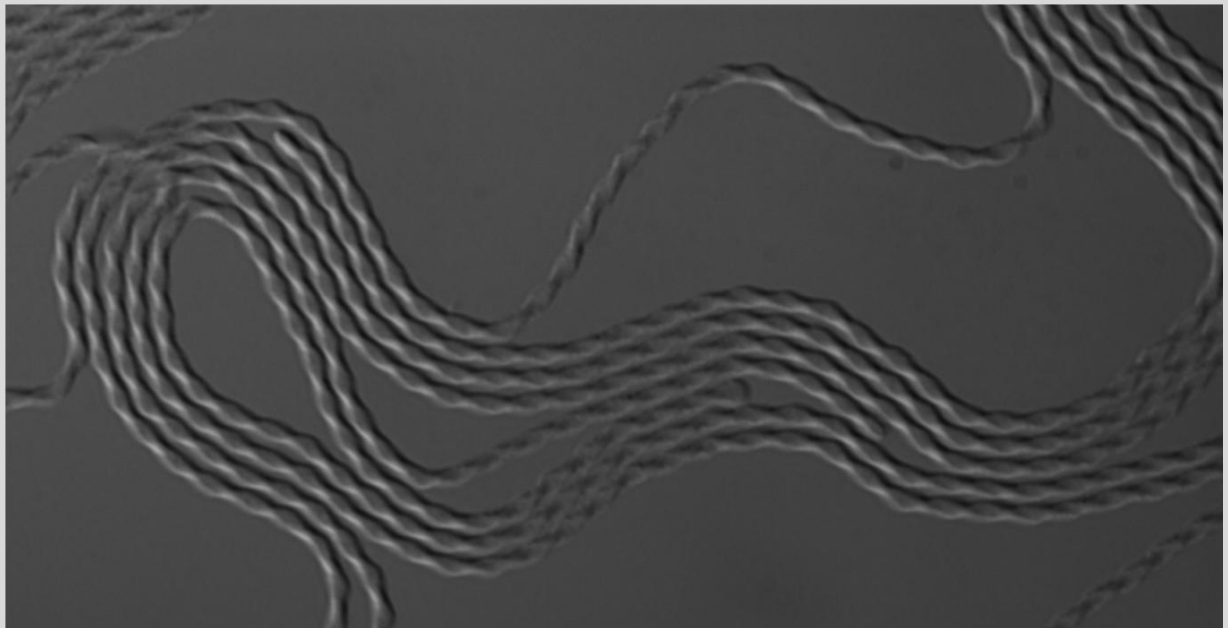


2. Coalescence of disks into ribbons

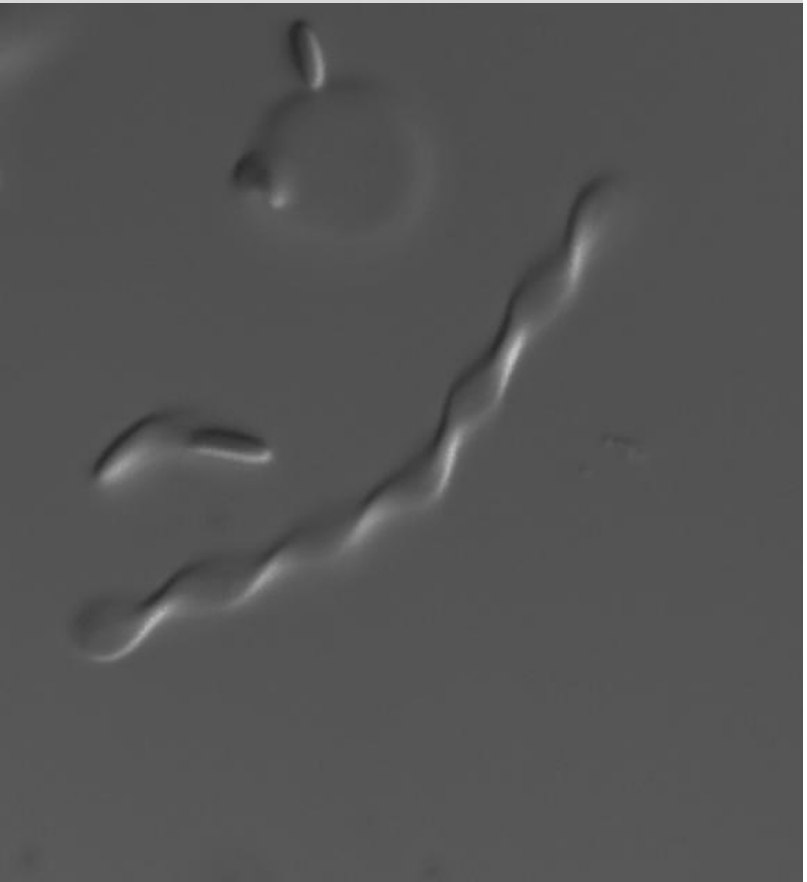


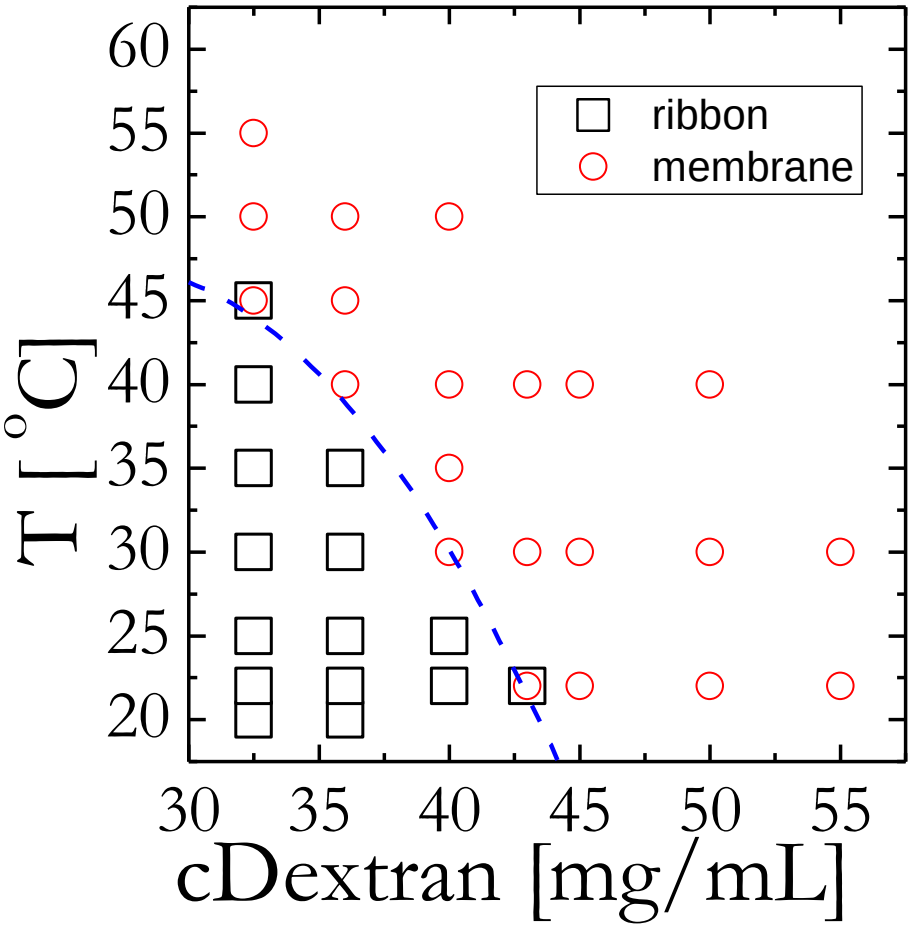
3. Growth of long ribbons





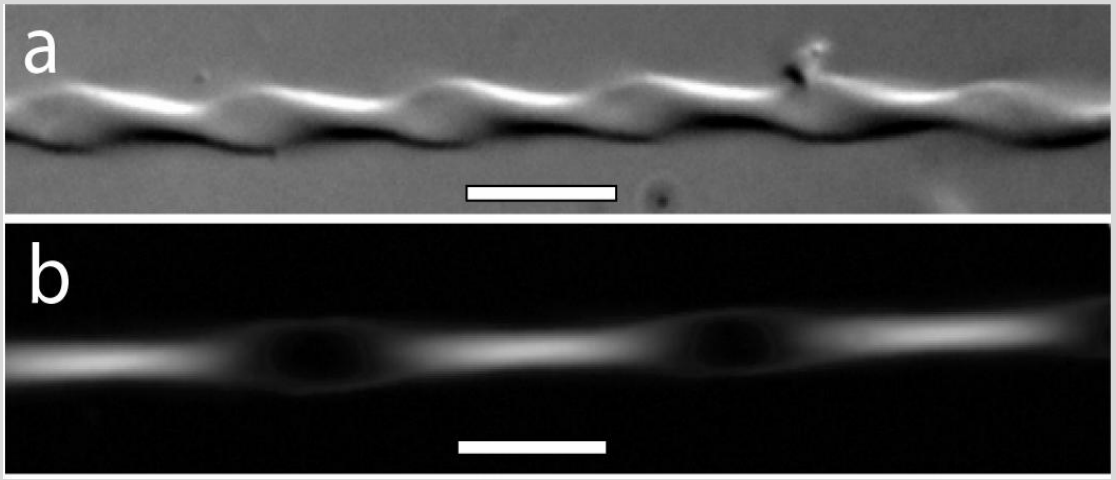
Phase diagram: 2D disks vs. 1D twisted ribbons



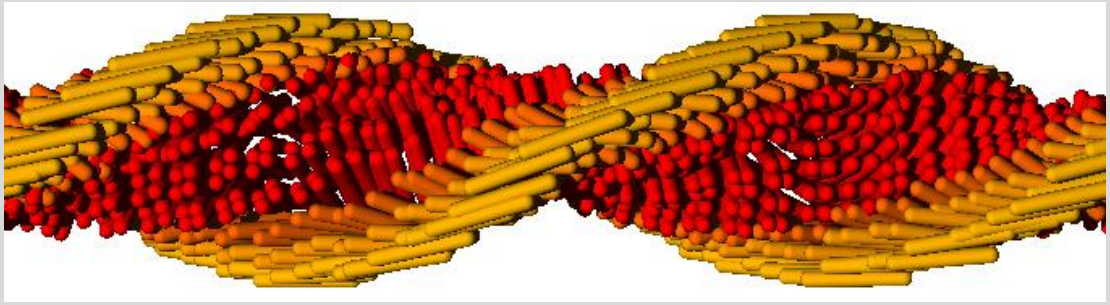


director field is inhomogeneous

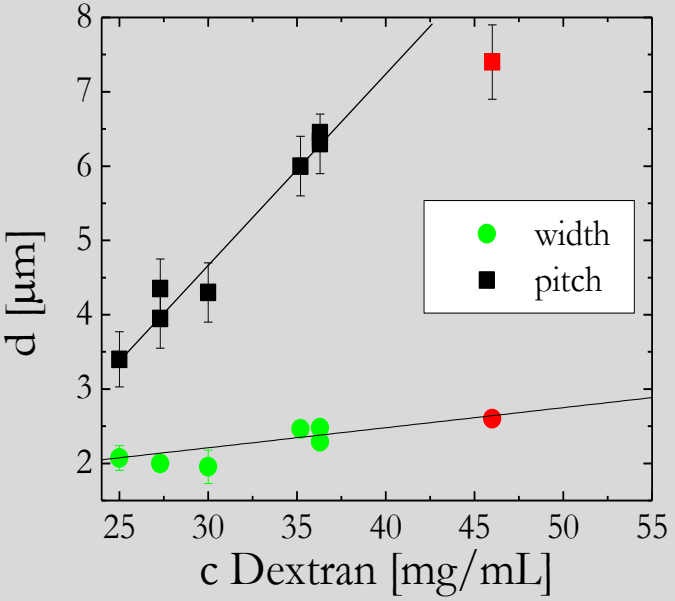
DIC image



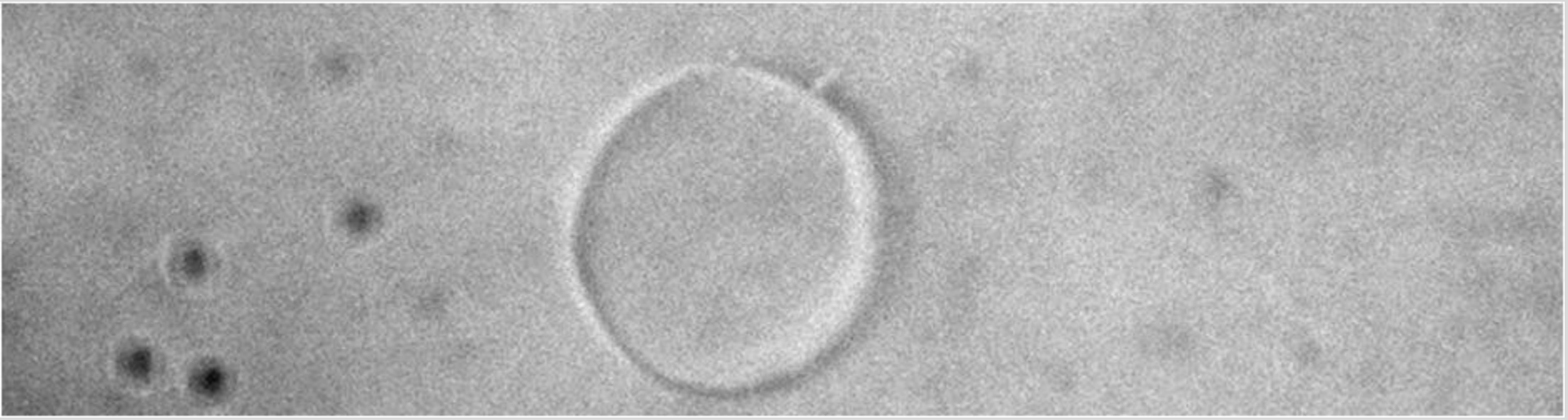
PolScope image



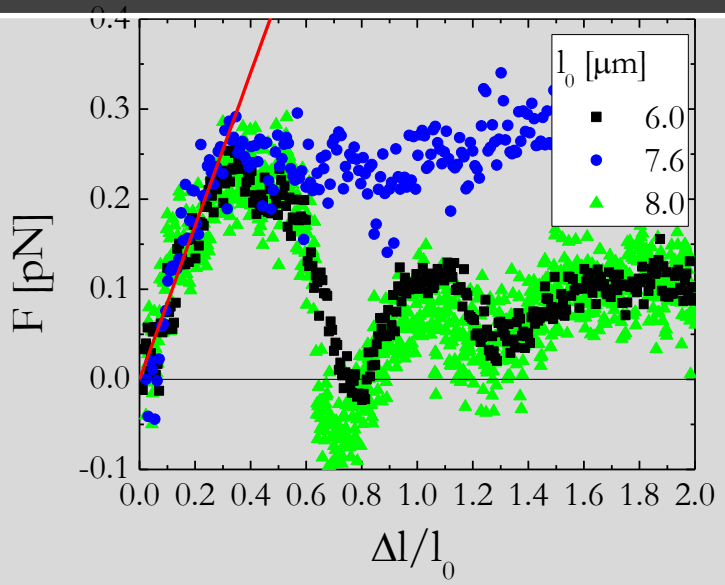
Pitch and width of twisted ribbons



Metastable ribbons

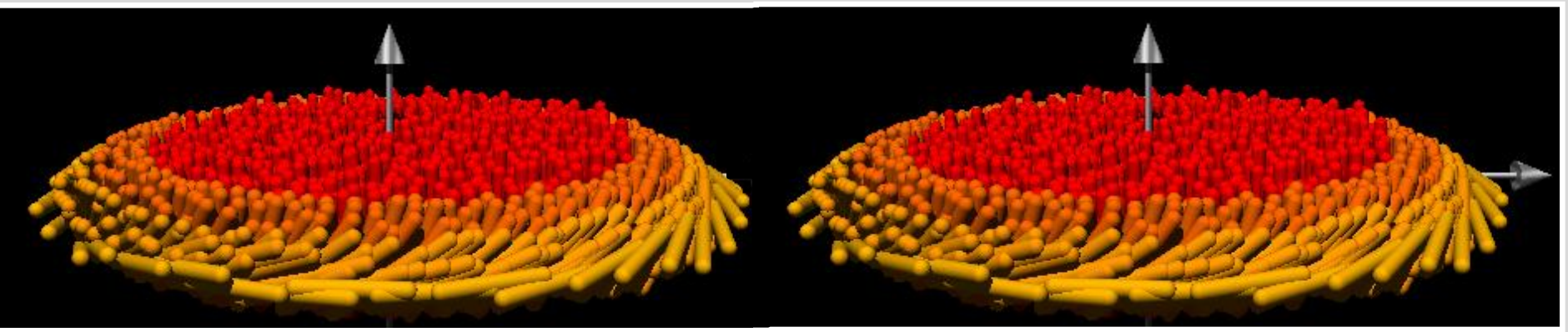


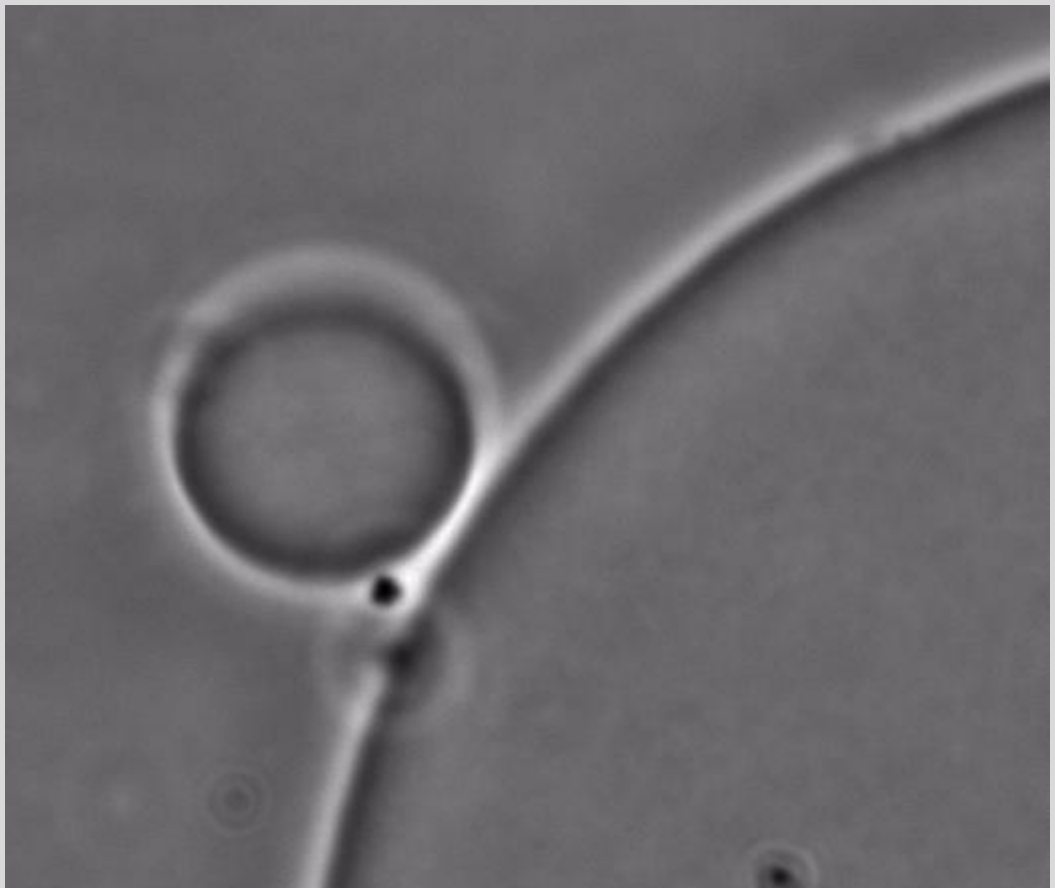
Force-extension curve

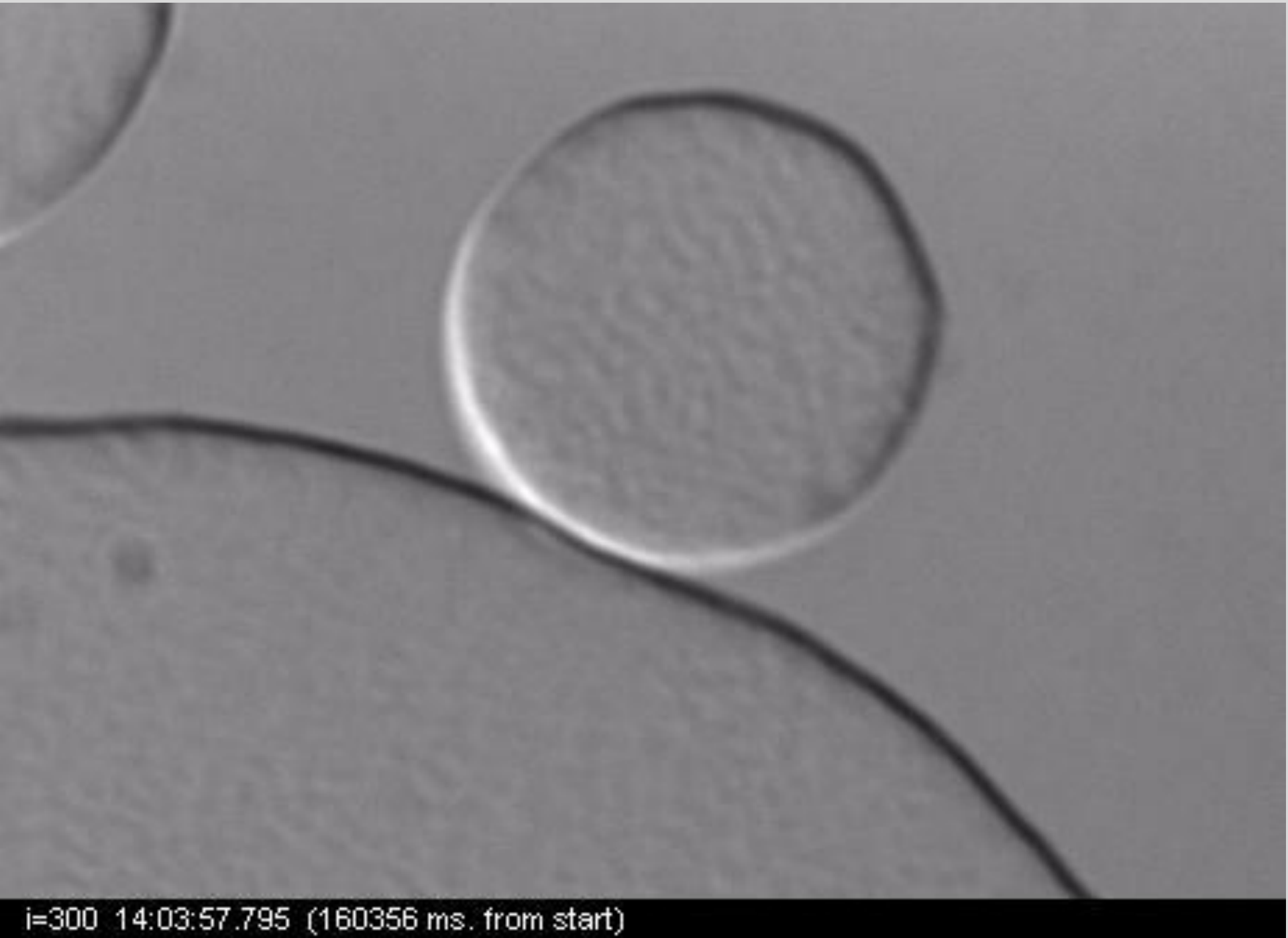


Part III:
defects in colloidal membranes

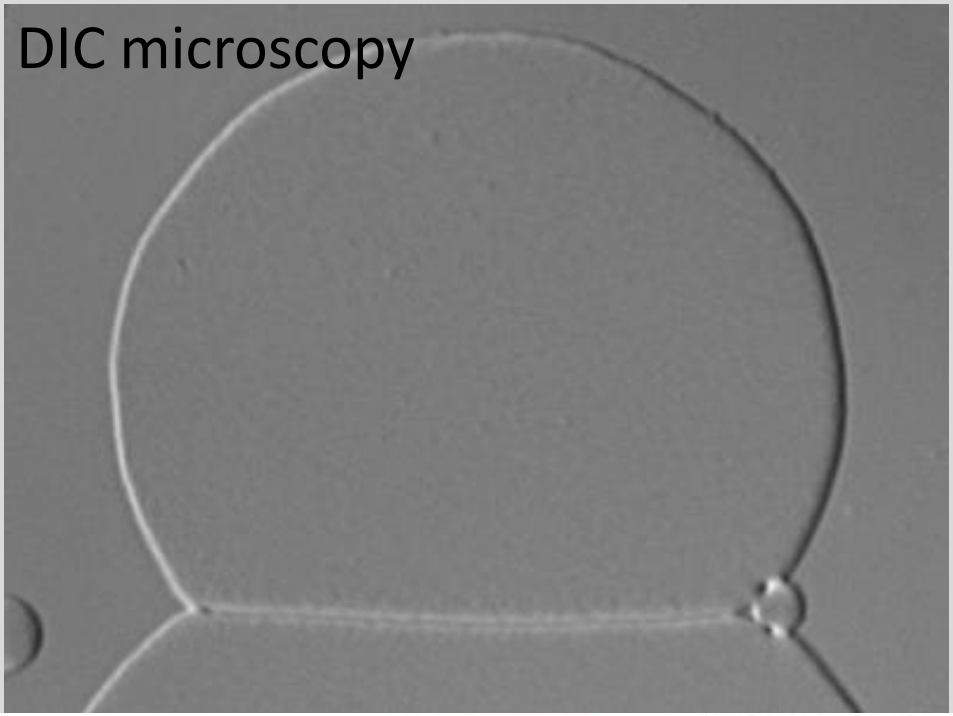
Disk coalescence is chirality dependent

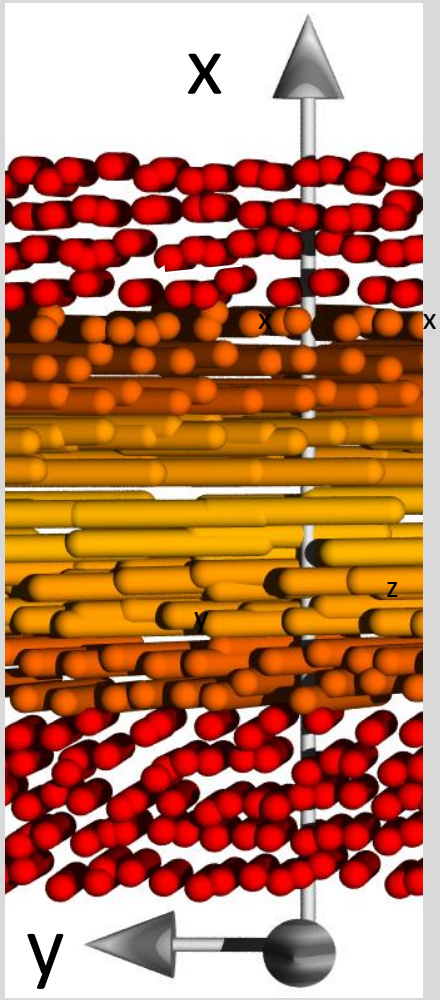


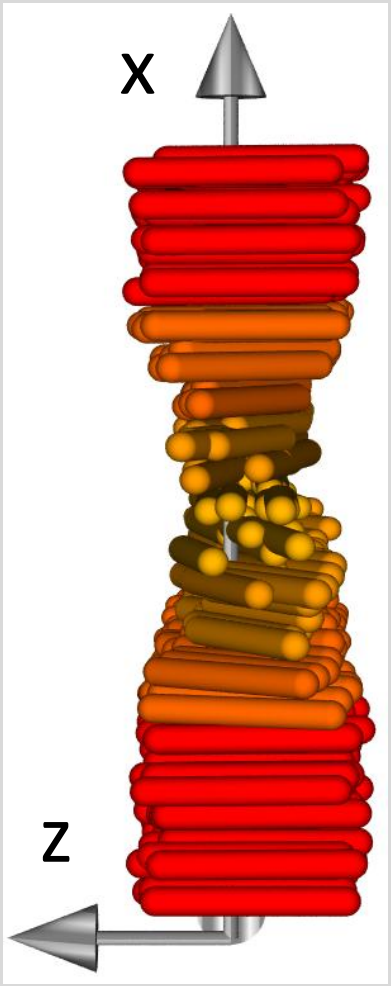


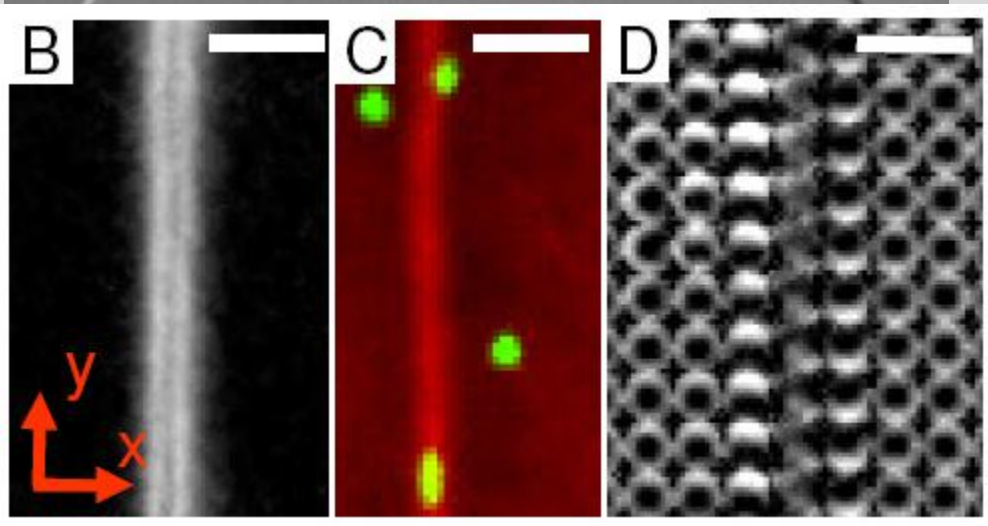

i=300 14:03:57.795 (160356 ms. from start)

Defects in chiral membranes: π twist walls





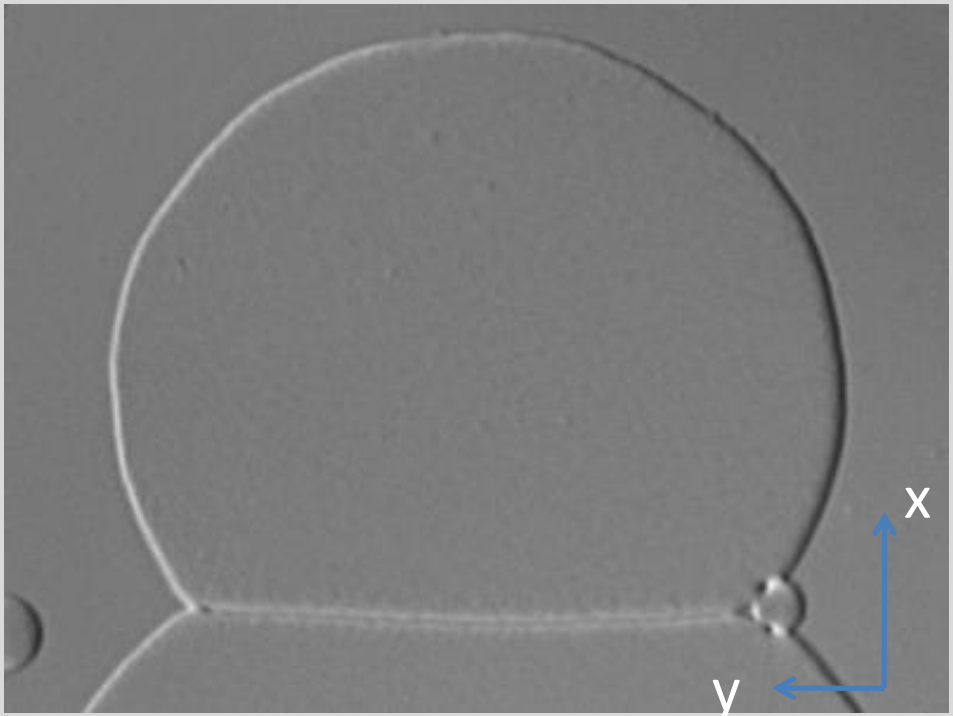




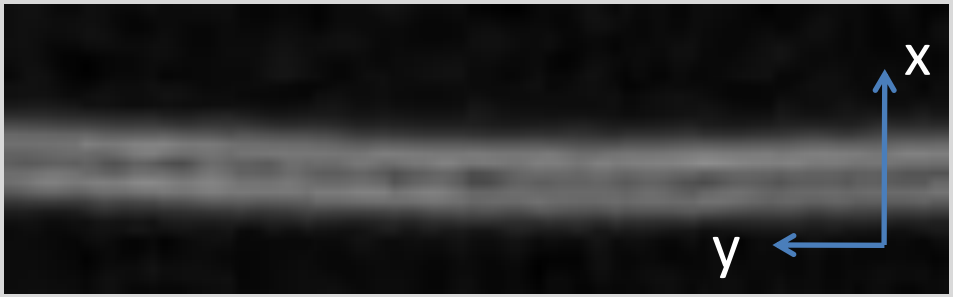
2D PolScope fluorescence 3D PolScope

core of the π wall is 1D nematic

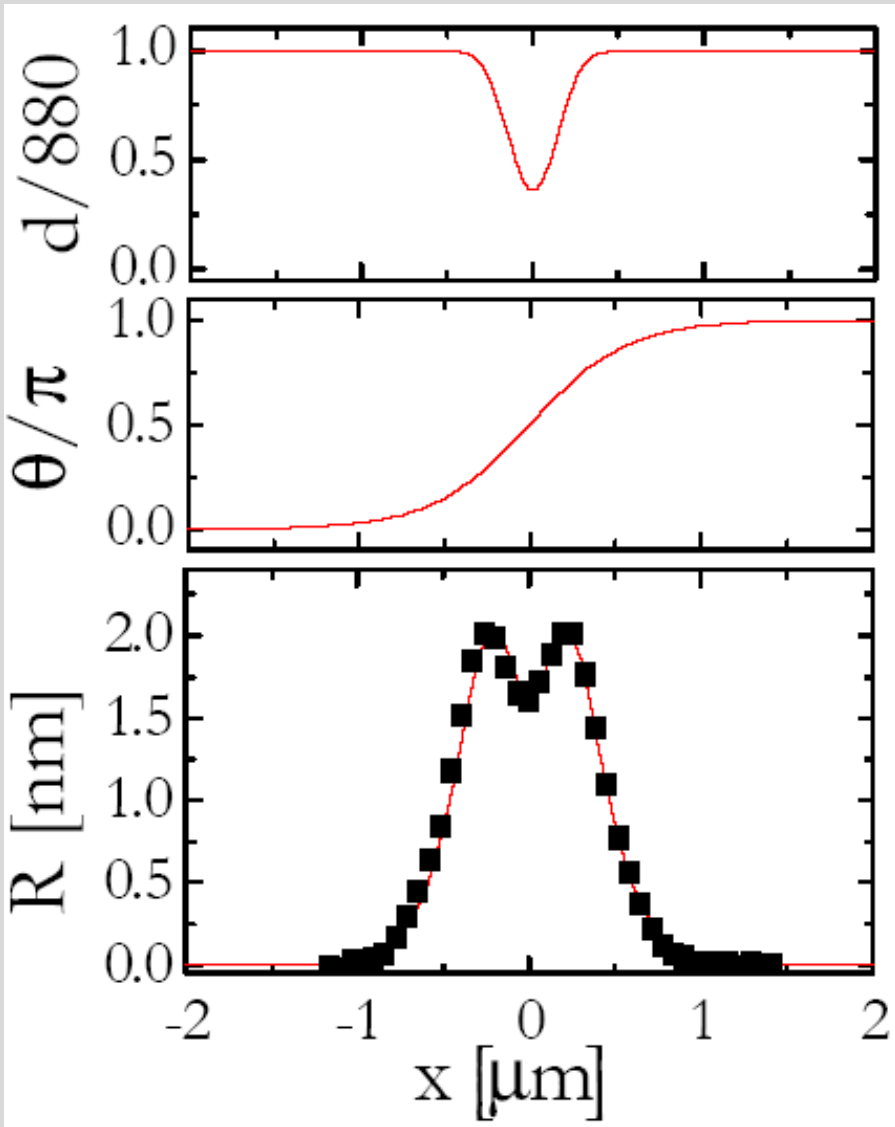
Defects in chiral membranes: π twist walls



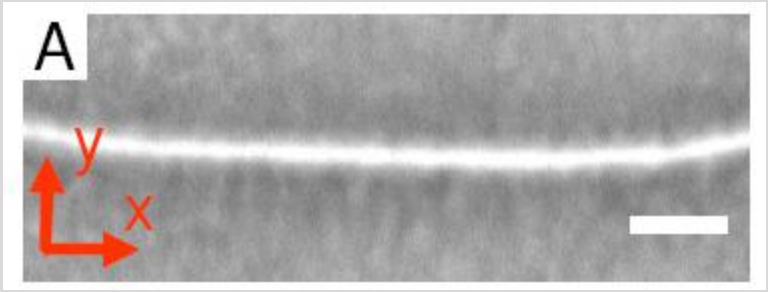
DIC microscopy

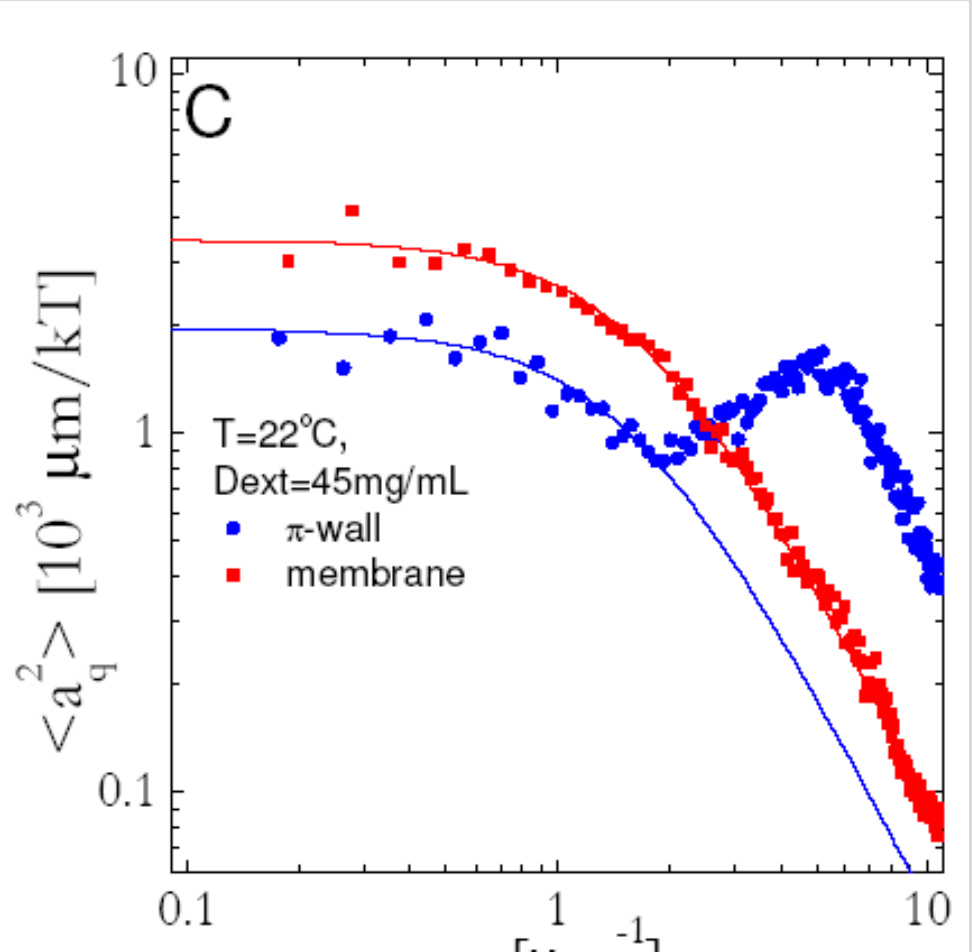


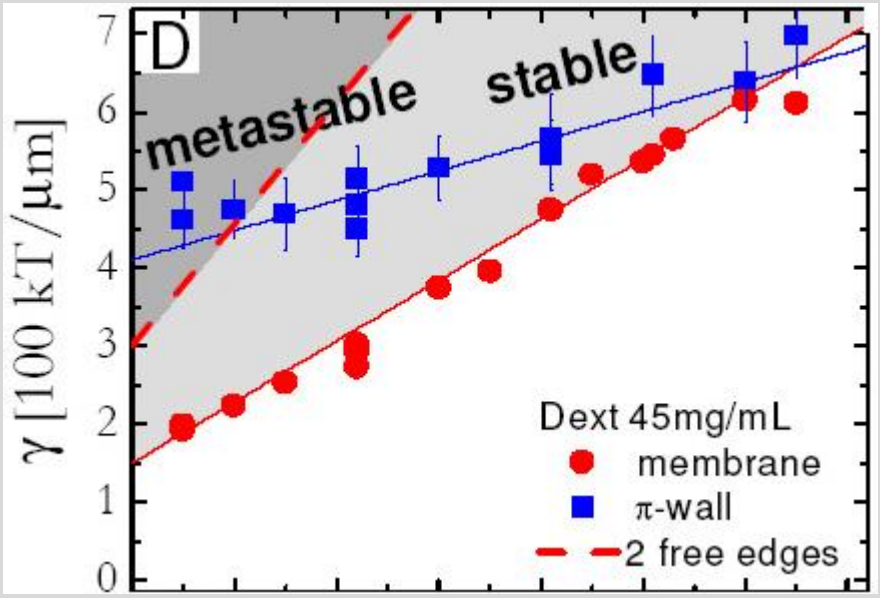
LC PolScope

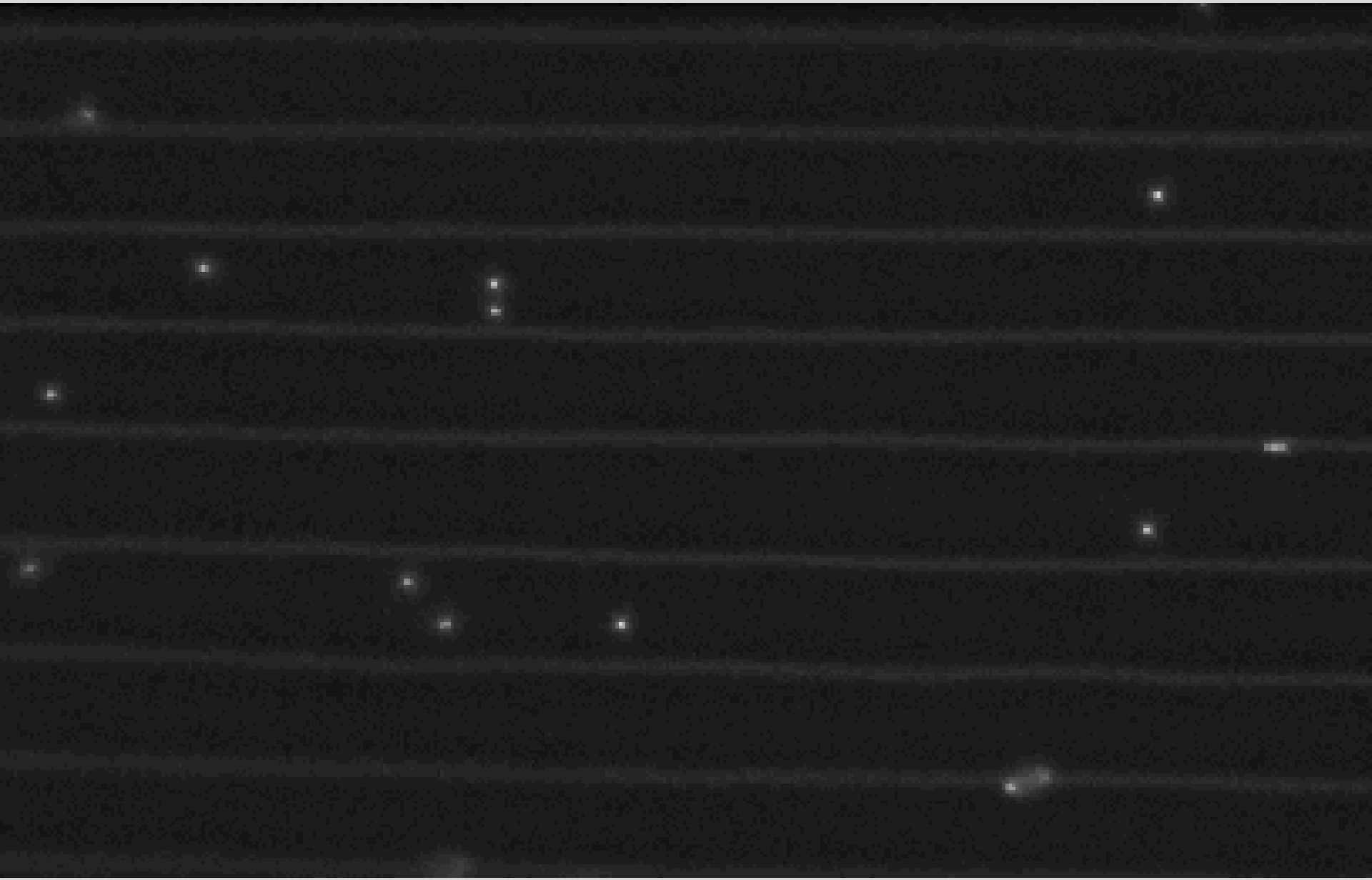


Line tension of π walls

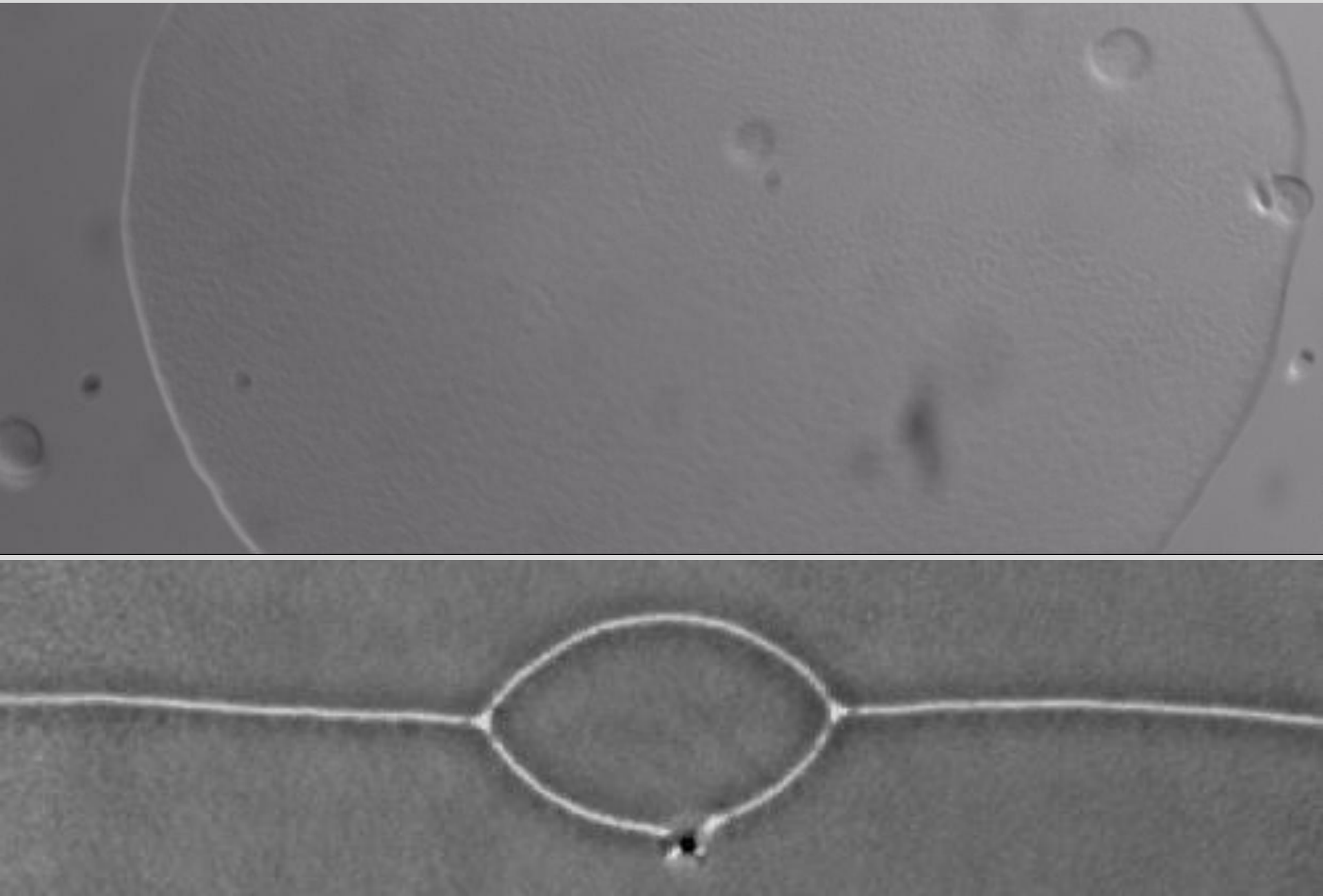


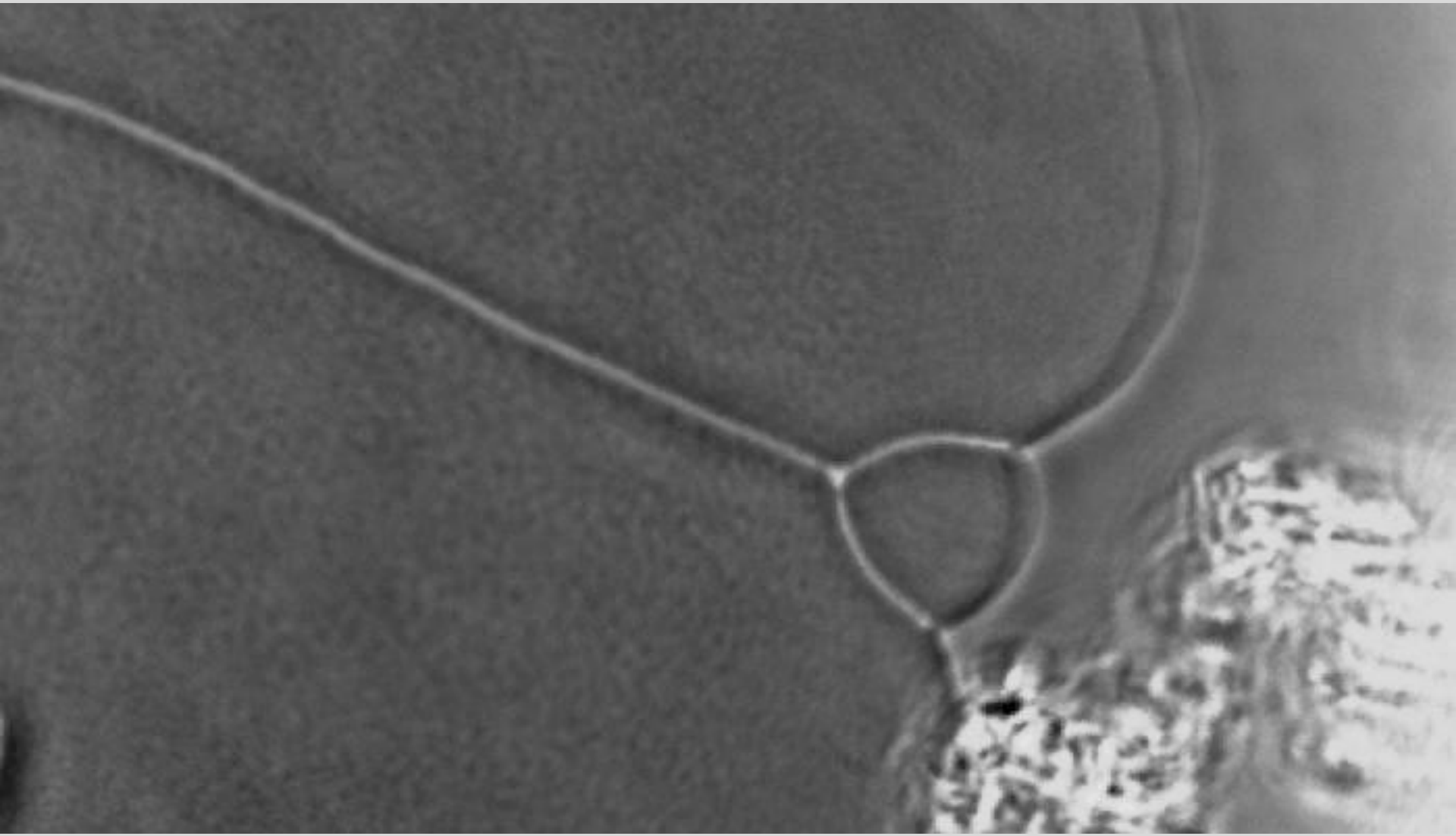




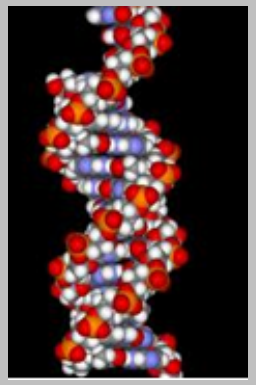


Creating π-walls with laser tweezers



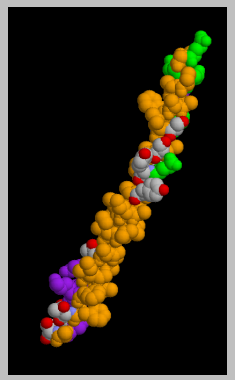


Hierarchical self-assembly of filamentous viruses



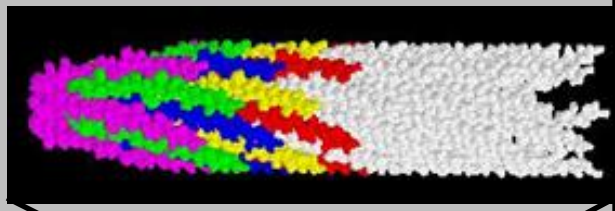
DNA

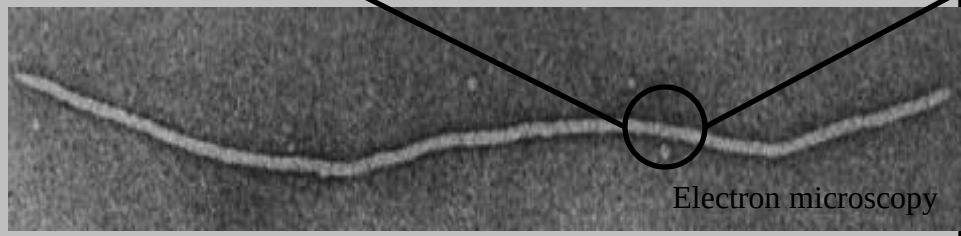
+



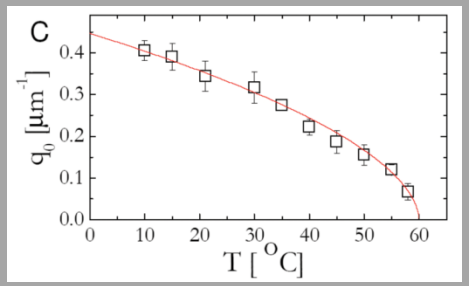
major coat protein

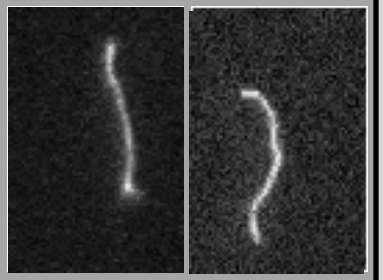
Monodisperse filamentous viruses





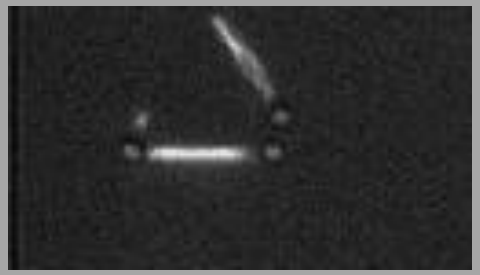
Flexibility, aspect ratio, chirality …

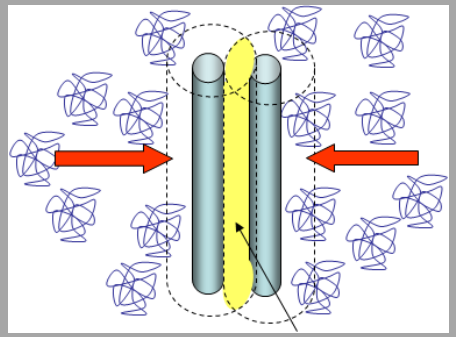




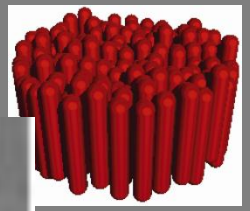
10 μm 2.5 μm

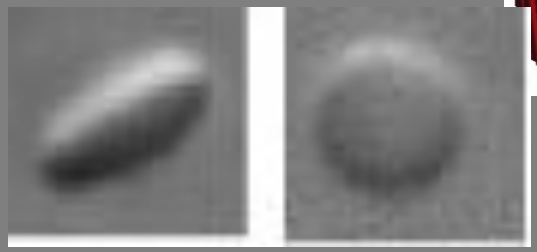
Attractive interactions



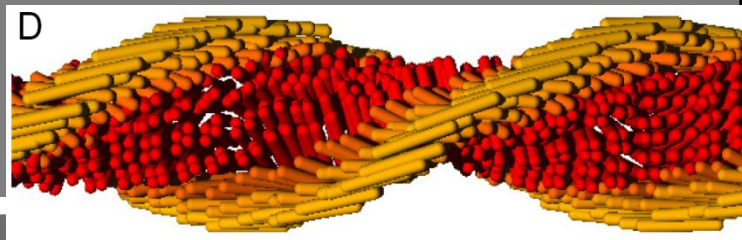


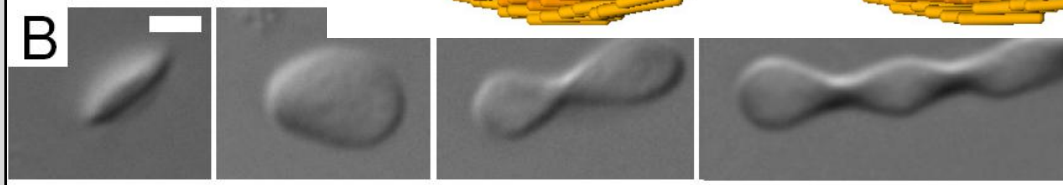
Mesoscopic self-assembled disks



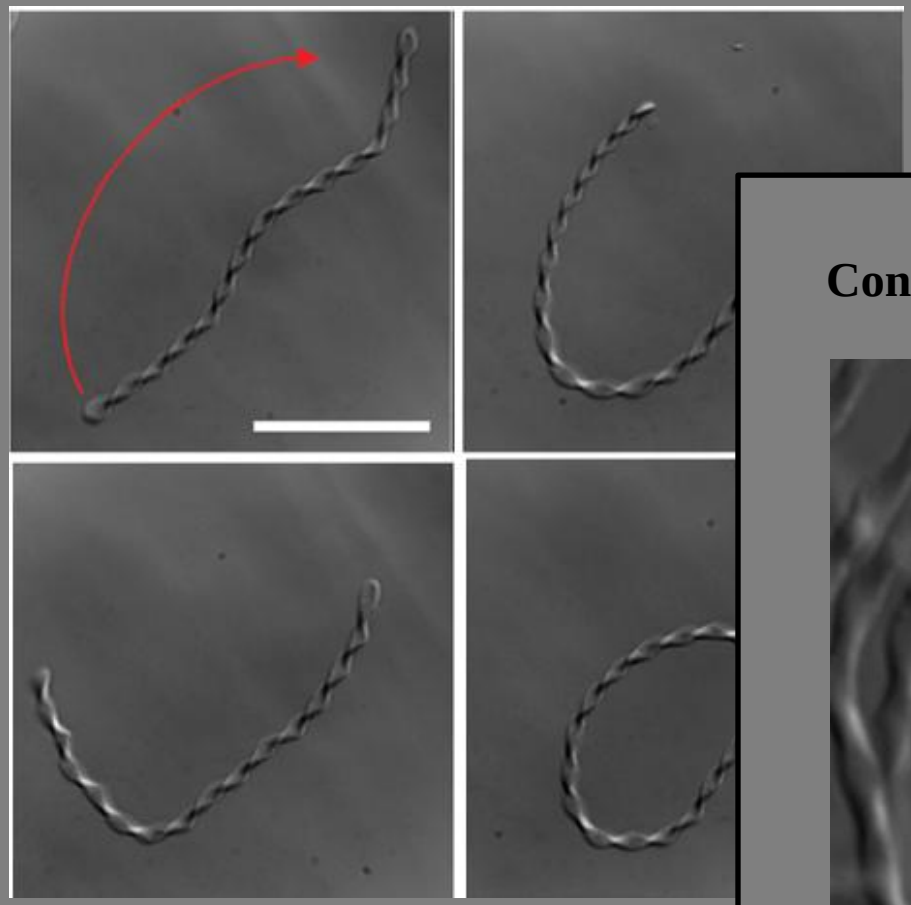


Twissted ribbons

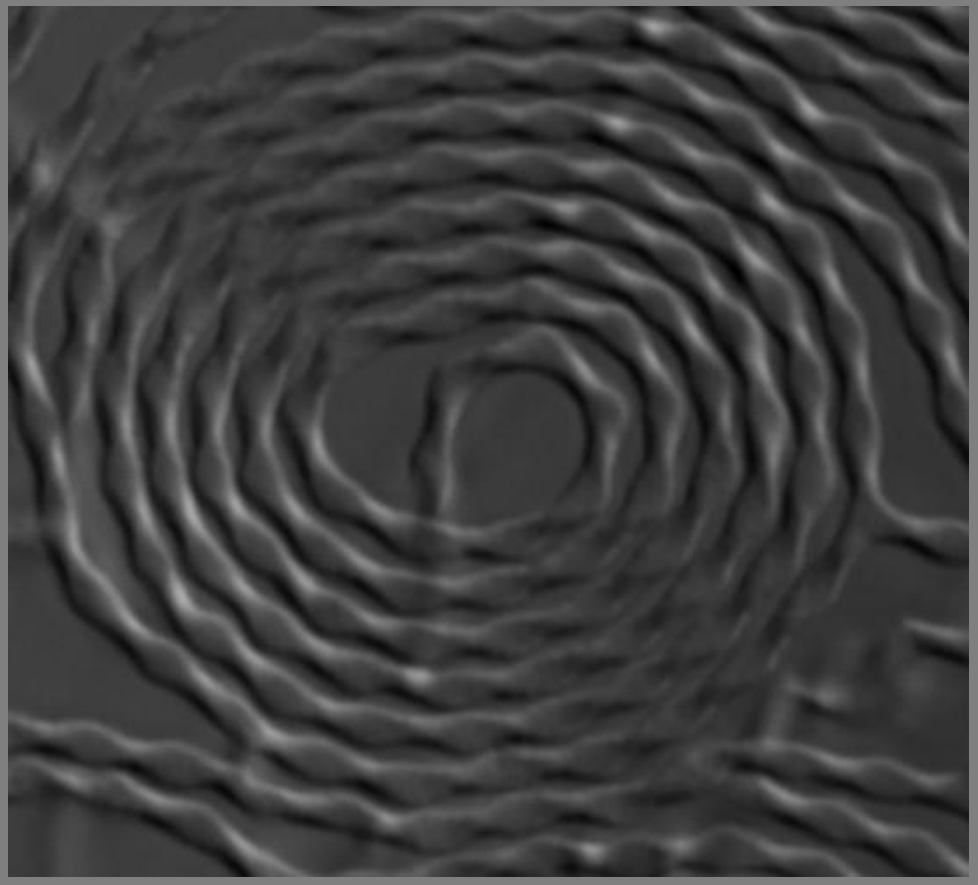




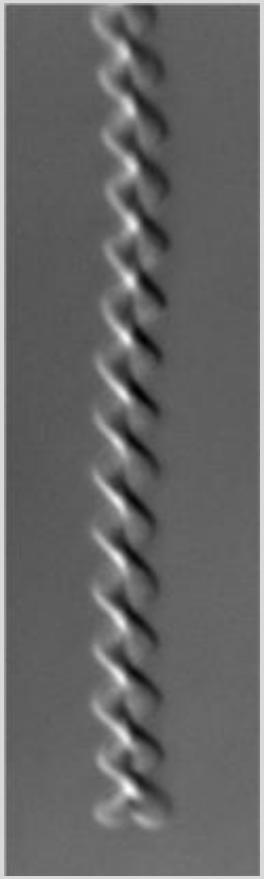
Assembly of ring polymers

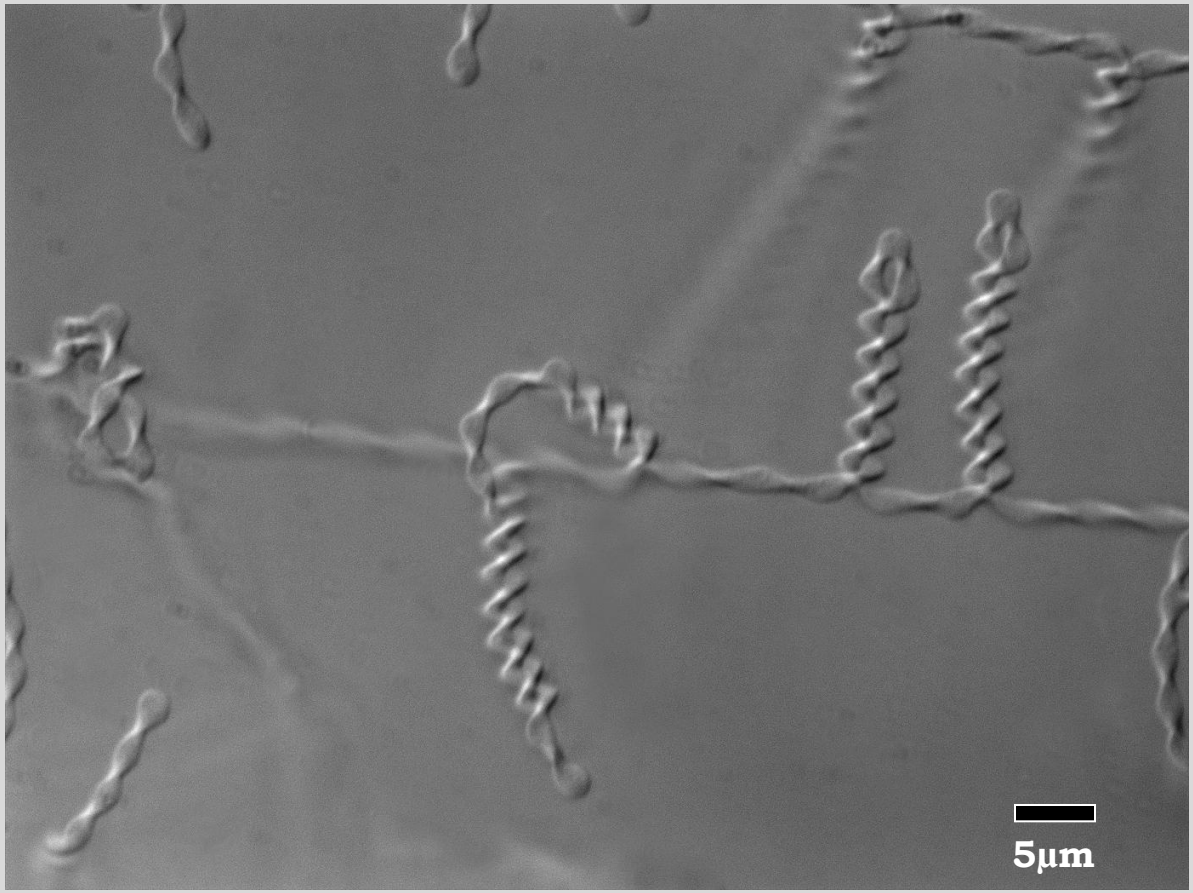


Condensation of twisted ribbons into torroids



Double Twisted Helices





Acknowledgements

Mike Hagan
Brandeis University

Robert Meyer
Brandeis University

Robert Pelcovits
Brown University

Rudolf Oldenbourg
MBL

Daniela Nicastro
Brandeis University

Thomas Gibaud
Brandeis University

Mark Zakhary
Brandeis University

Edward Barry
Brandeis University

MRSEC
Materials Research Science Engineering Center
Brandeis Univeristy
NSF