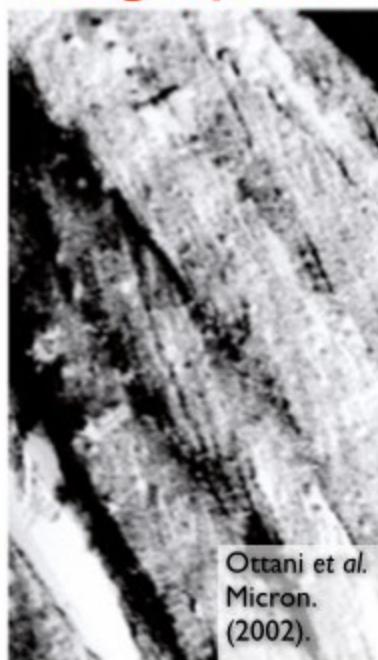
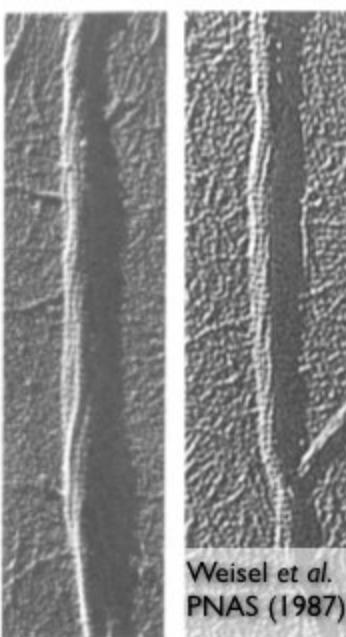


Geometric frustration in twisted filament assemblies: Non-euclidean packing & morphology of self-limiting bundles

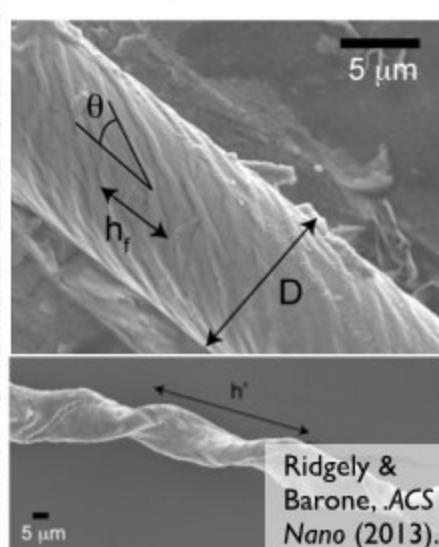
collagen fibrils



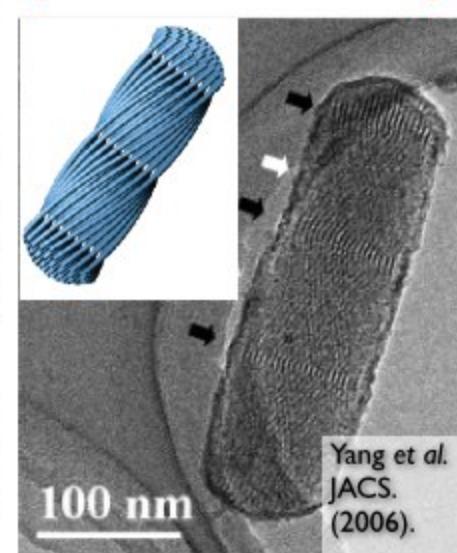
fibrin bundles



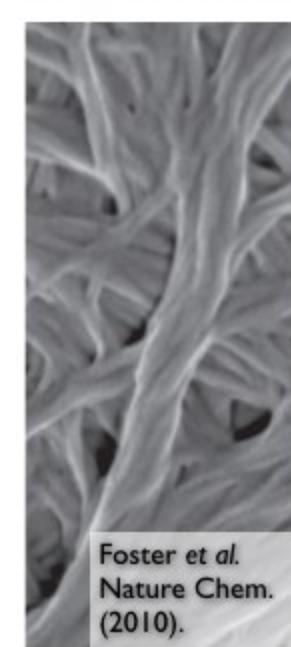
amyloid fibrils



**mesoporous silica
(columnar micelles)**



organogel fibers



Isaac Bruss¹, Amir Azadi², Doug Hall¹ & **Gregory M. Grason¹**

¹Department of Polymer Science & Engineering; ²Department of Physics
University of Massachusetts Amherst

<http://www.pse.umass.edu/ggrason>

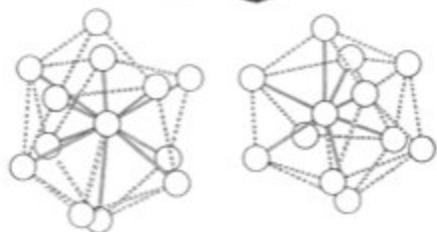


NSF CAREER DMR 09-55760; Alfred P. Sloan Foundation;
UMass Center for Hierarchical Manufacturing (NSF NSEC)



Models of matter: Spheres from Kepler to colloids

random-close packing:
from dense liquids to glassiness



Bernal, Nature (1960).

N-body cluster geometry
& viral expansion



2-sphere

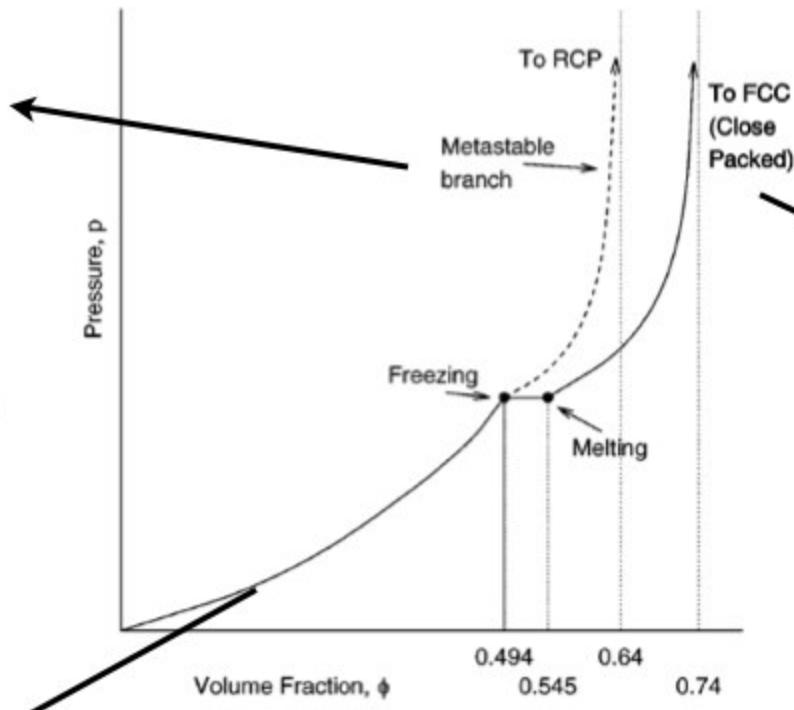


3-sphere



4-sphere

equation of state hard spheres

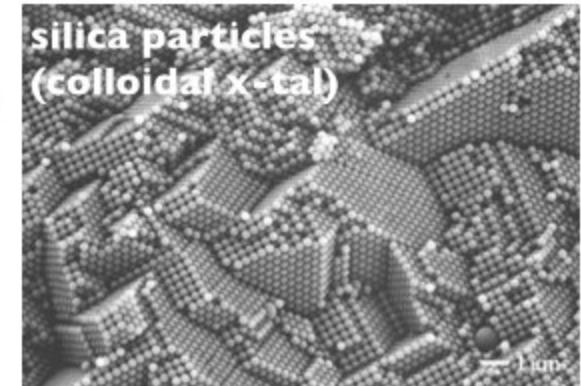
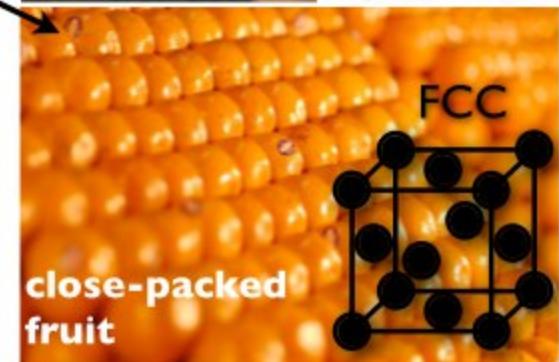
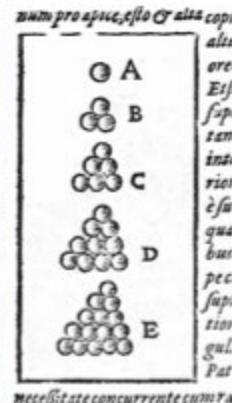


Rintoul & Torquato, PRL (1996).

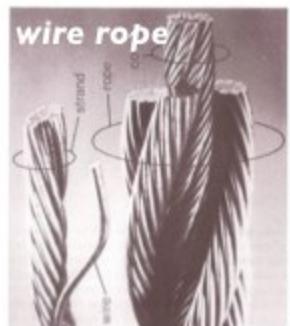
sphere packing, optimal lattices
& crystallization



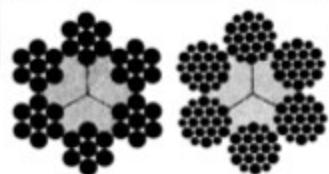
Kepler (1611)



Models of matter: Filamentous matter from Galileo to nanomaterials



macroscale,
manufactured
materials



(~0.1-1cm
diameter)



A spun nanotube yarns

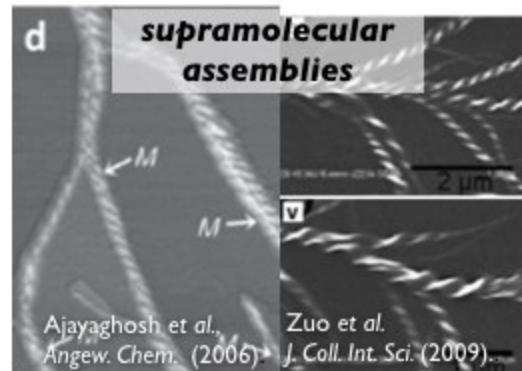
(~1 micron diameter)

Zhang, Atkinson & Baughman,
Science (2004).



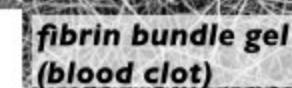
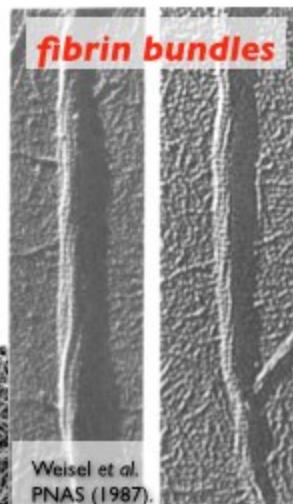
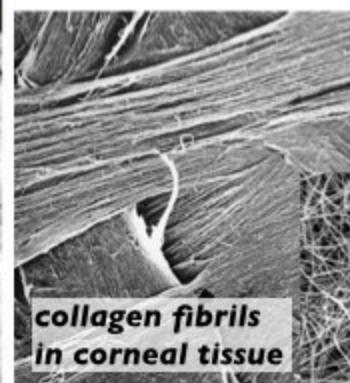
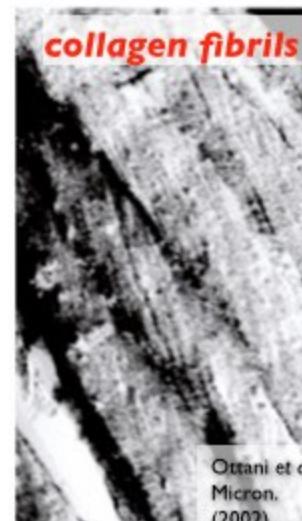
"But in the case of the rope, the very act of twisting causes the threads to bind one another in such a way that when the rope is stretched with a great force, the fibers break rather than separate from each other."

Galileo, Strength of Materials (1638)



nanoscale self-assembled
materials

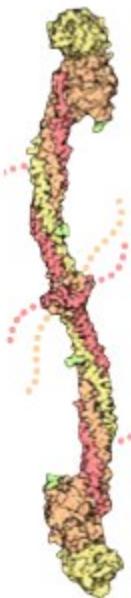
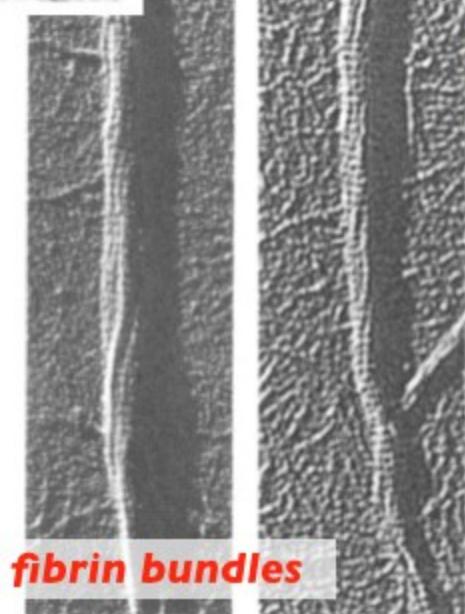
(~10-1000 nm "rope" diameter;
~1-10 nm "filament" diameter)



Structure & assembly of twisted, cohesive bundles



Weisel et al.
PNAS (1987).



Motivations:

- 1) “Self-twisting” filament bundle: common structural motif of biofilament assemblies
- 2) Twist: Simplest, non-trivial example of geometrically-nonlinear coupling between filament tilt and spacing

R - radius

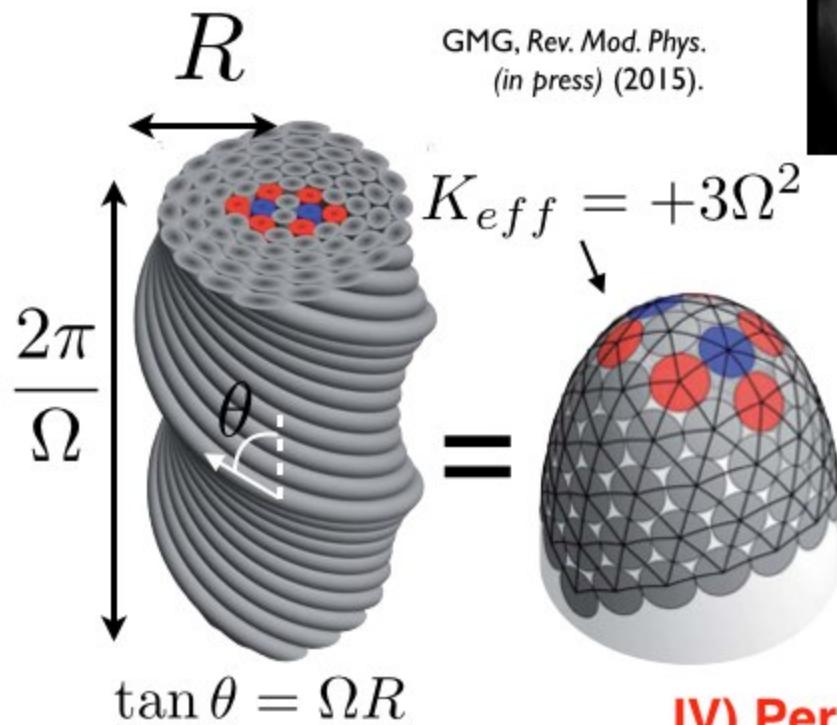
Ω - helical rotation rate ($2\pi/\text{pitch}$)

$$\tan \theta = \Omega R$$

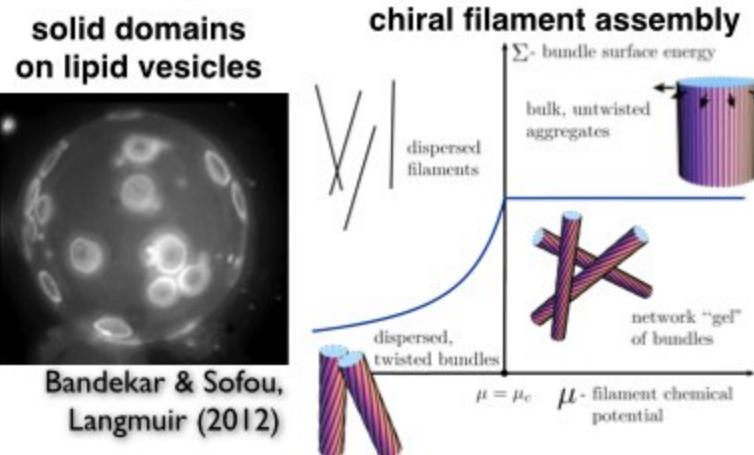
θ - helical tilt angle

Twisted bundles: non-Euclidean geometry & anomalous assembly

I) Non-euclidean metric geometry of bundles



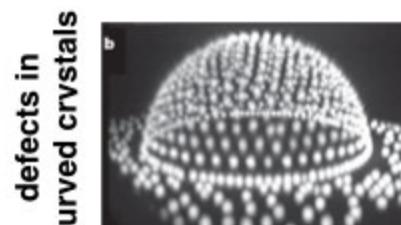
GMG, Rev. Mod. Phys.
(in press) (2015).



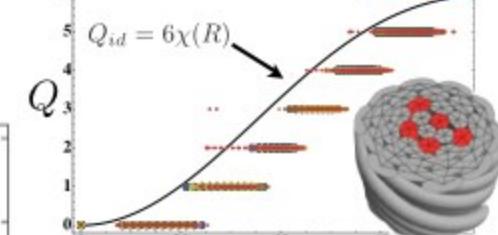
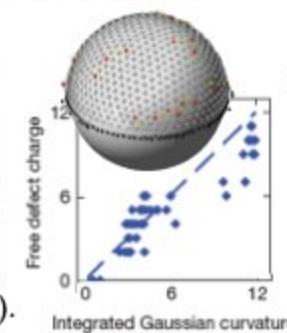
II) Self-limiting Assembly

GMG & Bruinsma,
PRL (2007); GMG,
PRE (2009)

III) Topological Defects

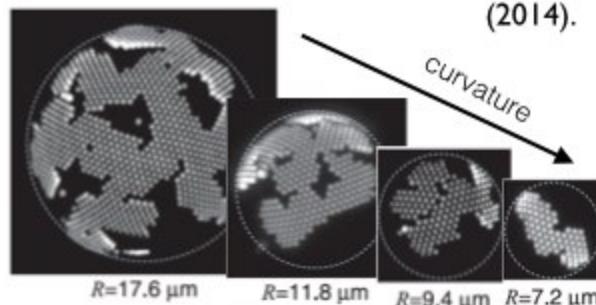
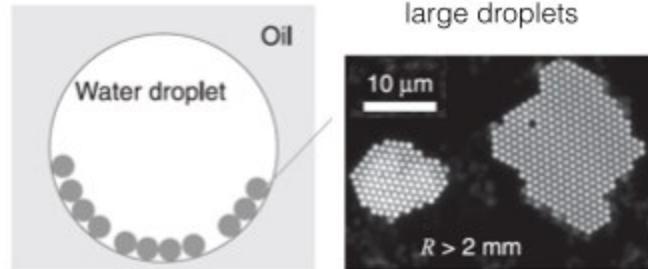


Irvine et al., Nature (2010).

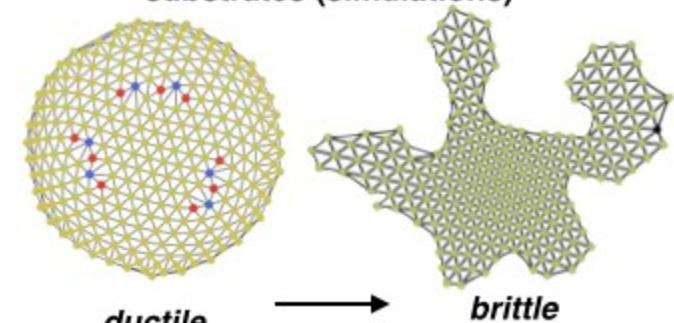


IV) Perimeter Instability & Anisotropic Domains

colloidal crystals on spherical droplets



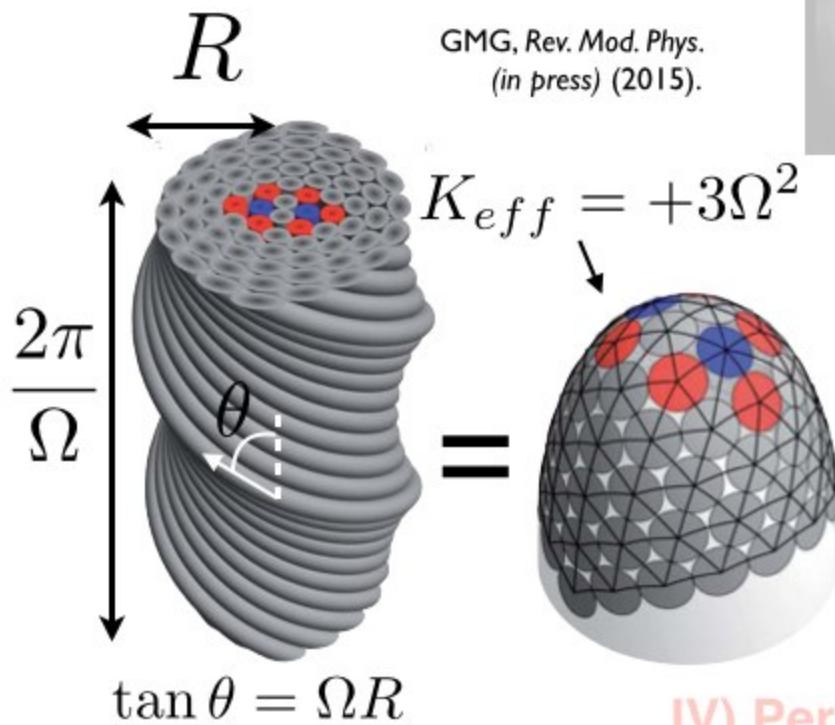
cohesive membranes on spherical substrates (simulations)



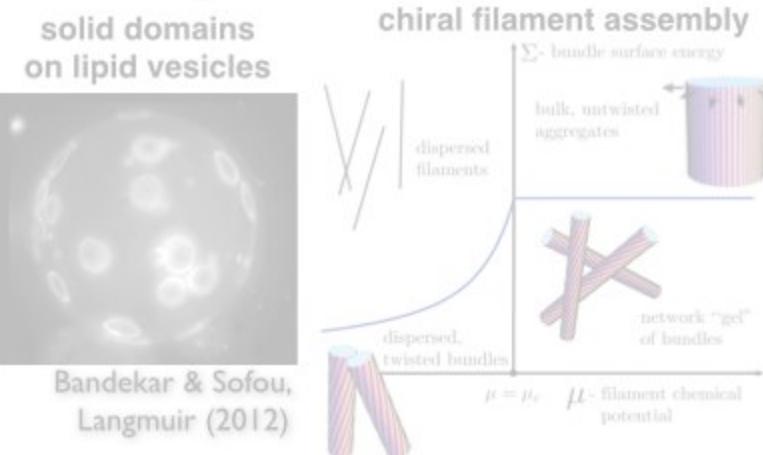
Amir Azadi, to be published.

Twisted bundles: non-Euclidean geometry & anomalous assembly

I) Non-euclidean metric geometry of bundles



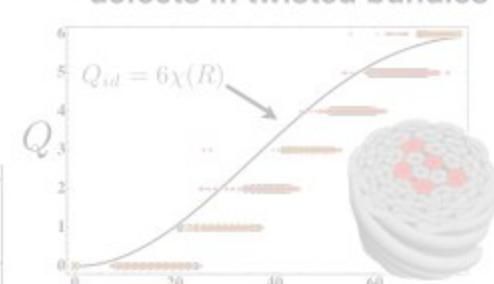
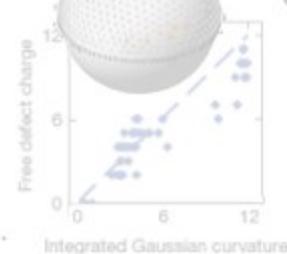
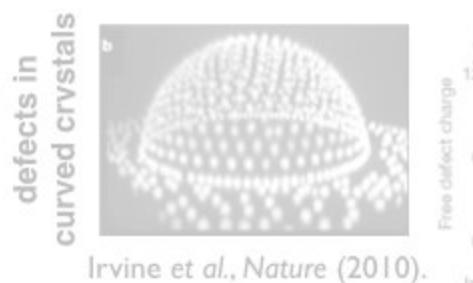
GMG, Rev. Mod. Phys.
(in press) (2015).



II) Self-limiting Assembly

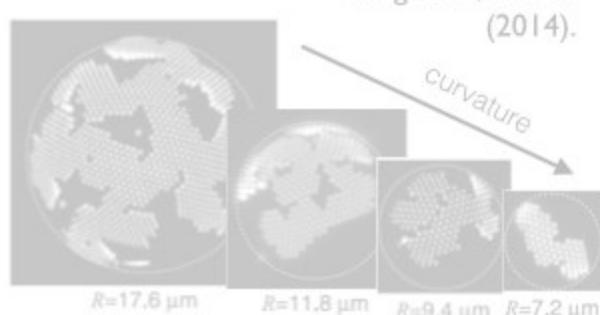
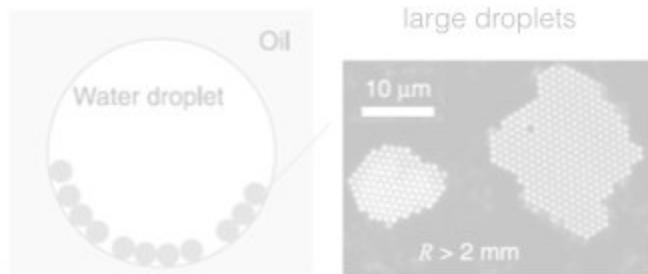
GMG & Bruinsma,
PRL (2007); GMG,
PRE (2009)

III) Topological Defects

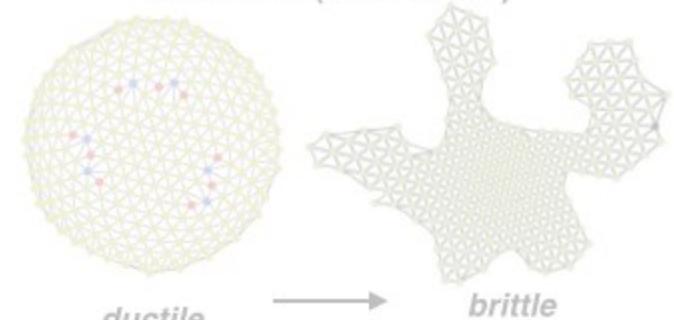


IV) Perimeter Instability & Anisotropic Domains

colloidal crystals on spherical droplets

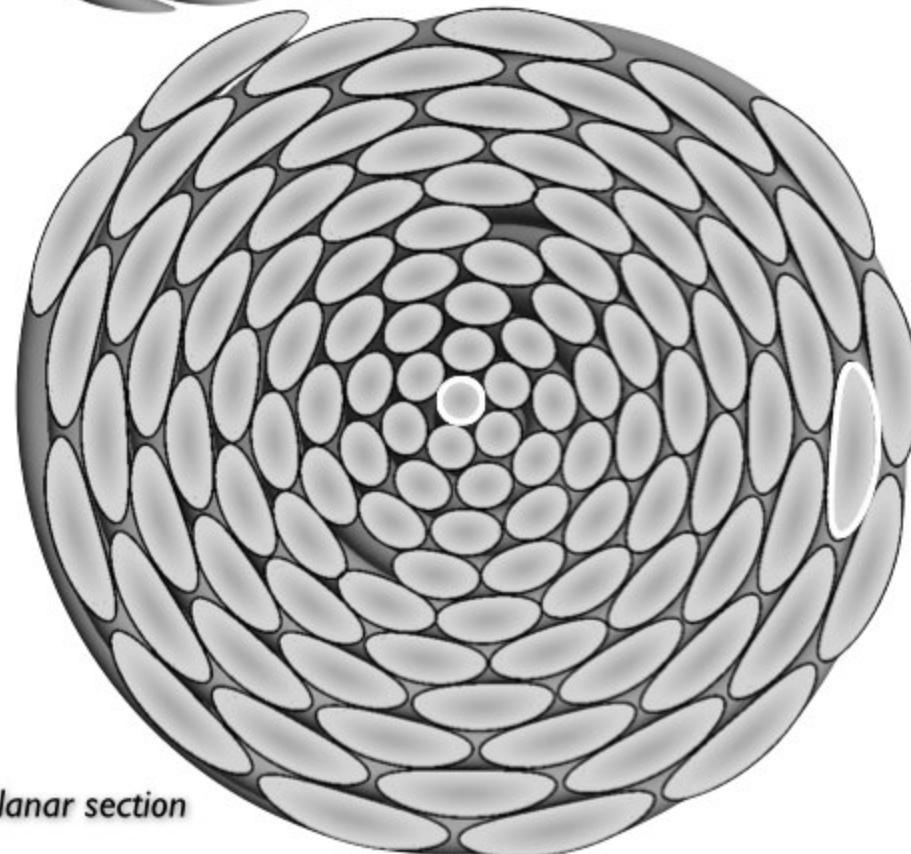
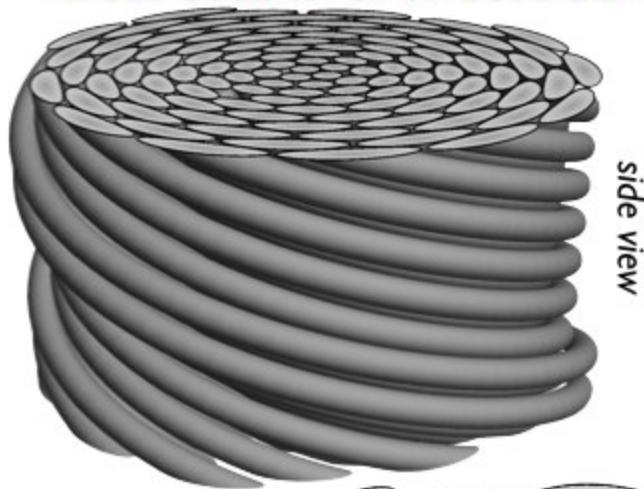


cohesive membranes on spherical substrates (simulations)

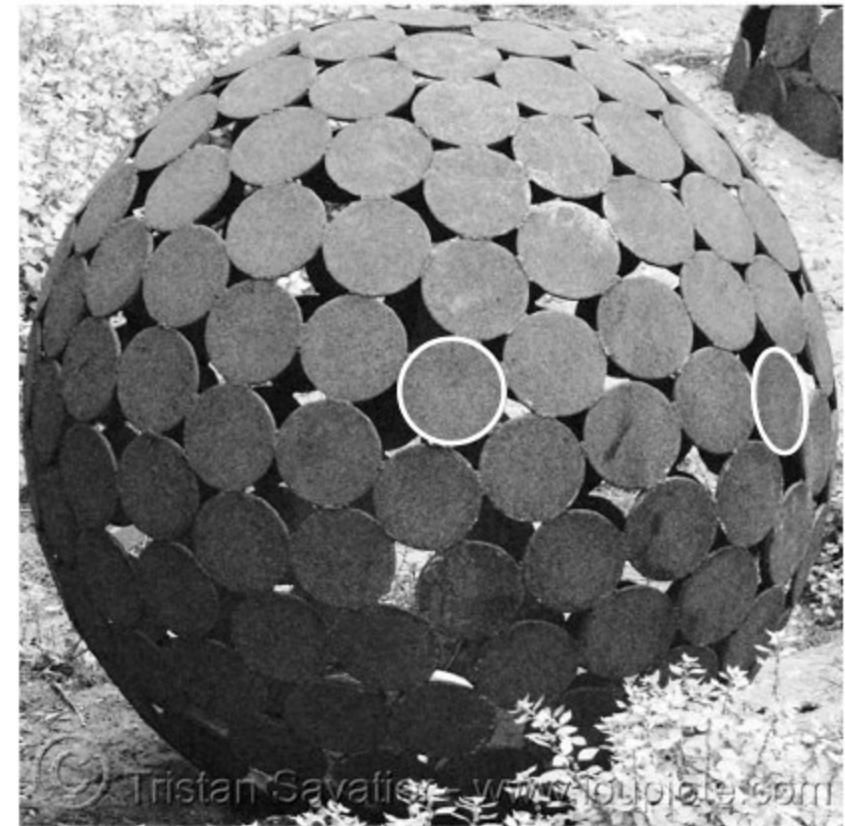


Mapping frustration in filament packing

cross-section of twisted bundle

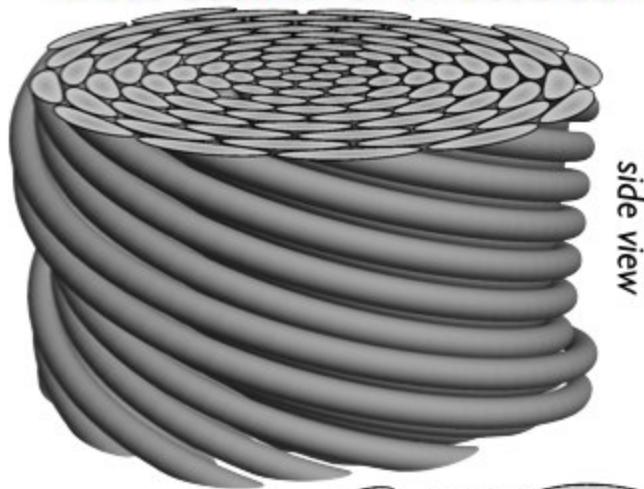


packing discs on sphere
(orthographic projection)



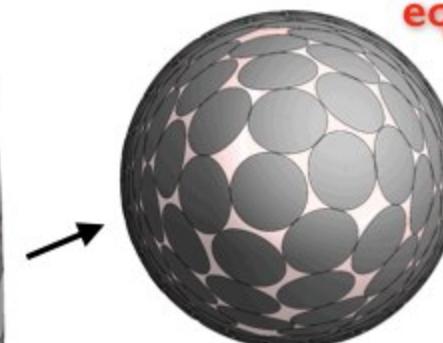
Mapping frustration in filament packing

cross-section of twisted bundle

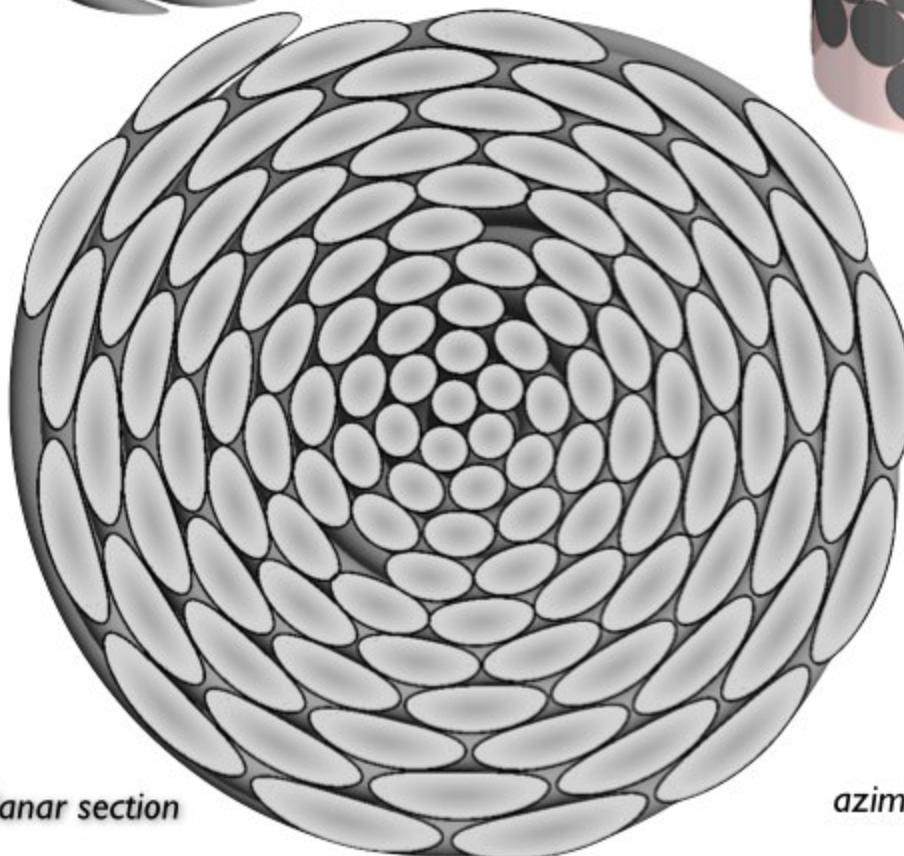


side view

packing discs on a “bundle-equivalent dome”

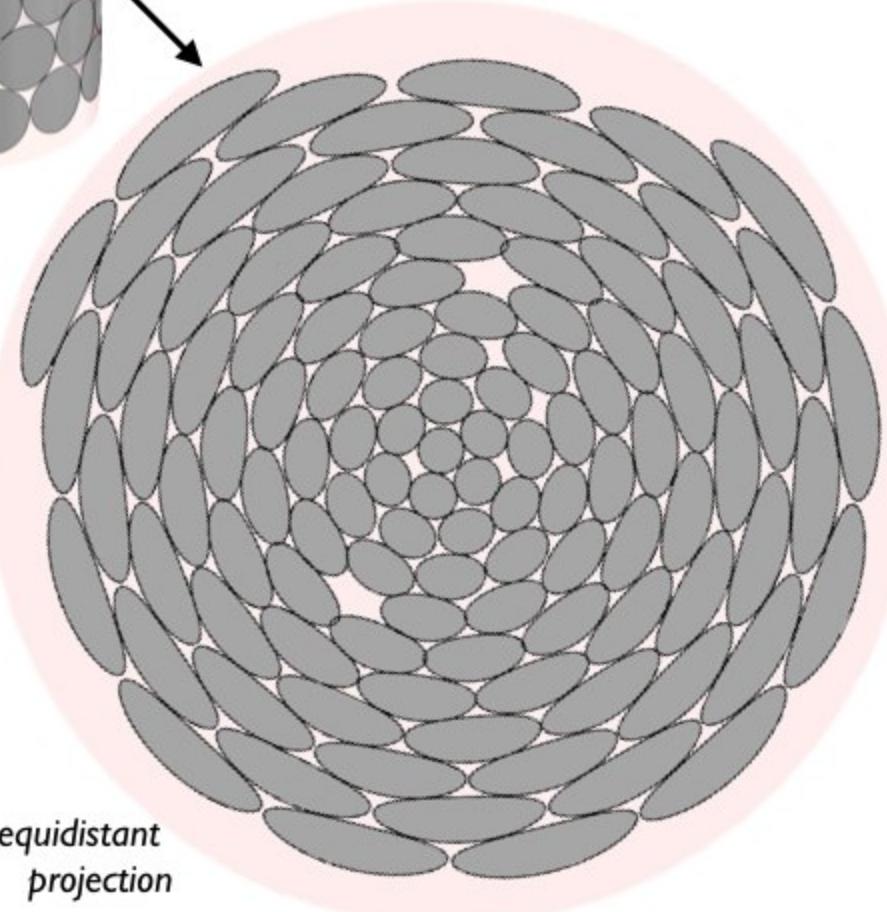


orthographic projection (“from above”)

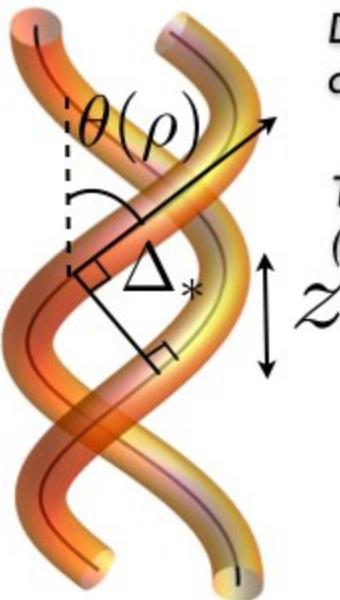


planar section

azimuthal equidistant
projection



Mapping frustration in filament packing



Distance of closest approach: $\Delta_* = \min_z [\Delta(z)]$

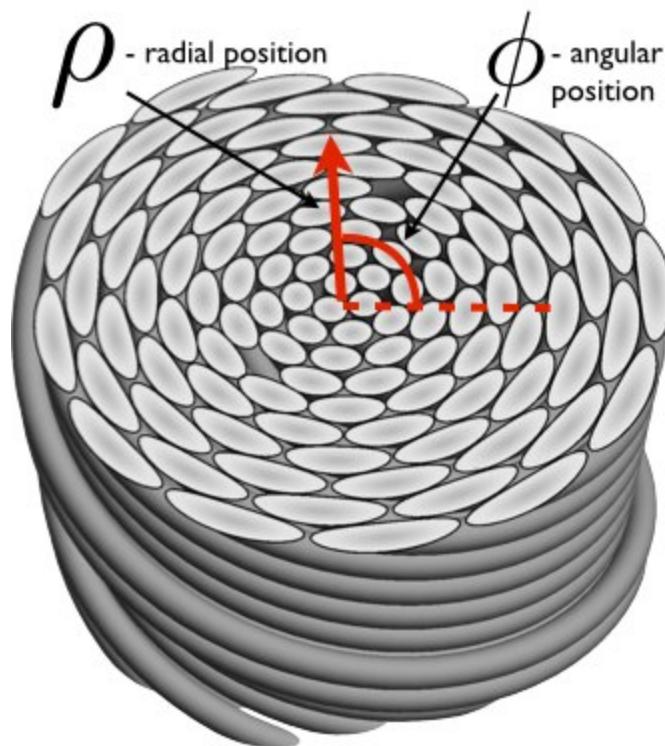
Tilt angle (local): $\sin \theta(\rho) = \frac{\Omega \rho}{\sqrt{1 + (\Omega \rho)^2}}$

Distance between helical curves:

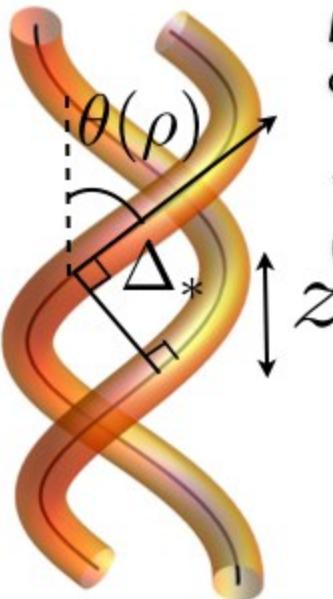
$$\Delta^2(z) = \rho_1^2 + \rho_2^2 - 2\rho_1\rho_2 \cos(\Omega z + \phi_1 - \phi_2) + z^2$$

Metric geometry of bundles:

$$\lim_{\delta\phi \rightarrow 0, \delta\rho \rightarrow 0} \Delta_*^2 \equiv ds^2 = (\delta\rho)^2 + \Omega^{-2} \sin^2 \theta(\rho) (\delta\phi)^2$$



Mapping frustration in filament packing



Distance of closest approach: $\Delta_* = \min_z [\Delta(z)]$

Tilt angle (local): $\sin \theta(\rho) = \frac{\Omega \rho}{\sqrt{1 + (\Omega \rho)^2}}$

Distance between helical curves:

$$\Delta^2(z) = \rho_1^2 + \rho_2^2 - 2\rho_1\rho_2 \cos(\Omega z + \phi_1 - \phi_2) + z^2$$

Metric geometry of bundles:

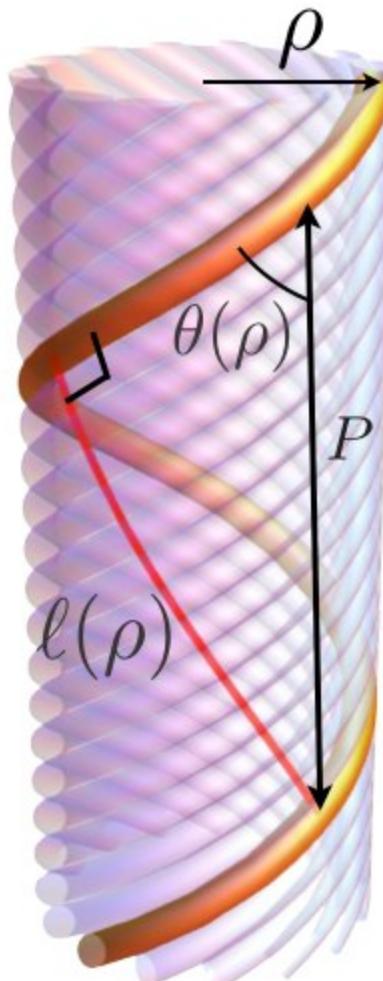
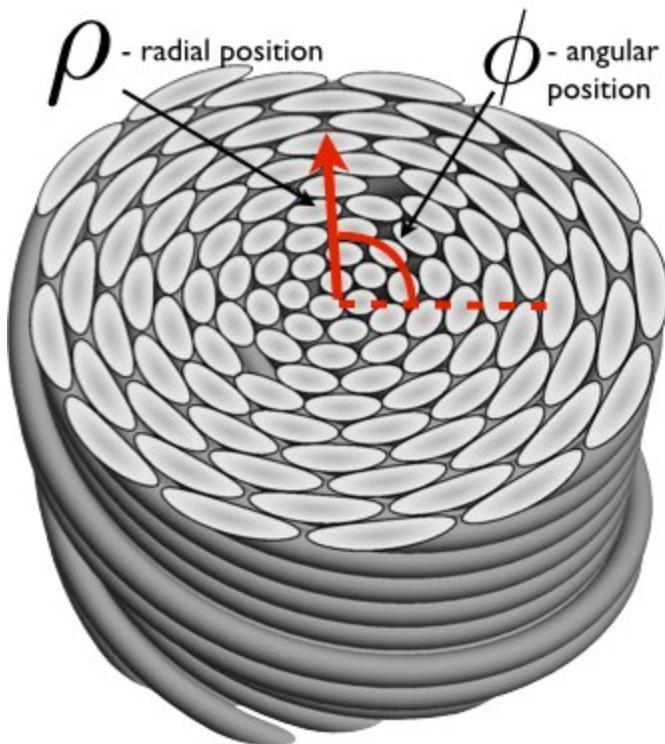
$$\lim_{\delta\phi \rightarrow 0, \delta\rho \rightarrow 0} \Delta_*^2 \equiv ds^2 = (\delta\rho)^2 + \Omega^{-2} \sin^2 \theta(\rho) (\delta\phi)^2$$

Perimeter: space available in bundle @ ρ

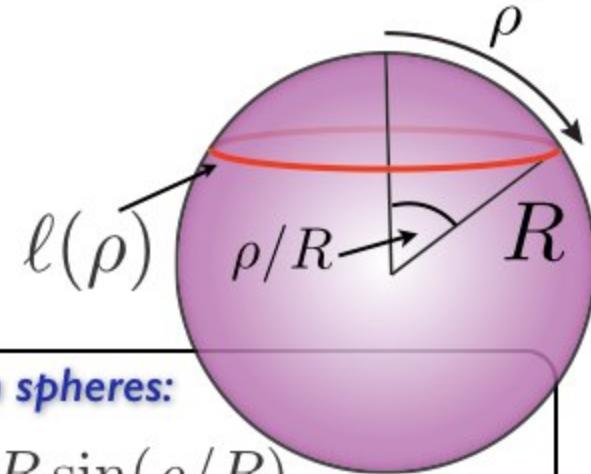
$$\ell(\rho) = P \sin \theta(\rho)$$

less than
planar packing!

$$\simeq 2\pi\rho \left(1 - \frac{\Omega^2}{2}\rho^2\right)$$



$$P = \frac{2\pi}{\Omega}$$

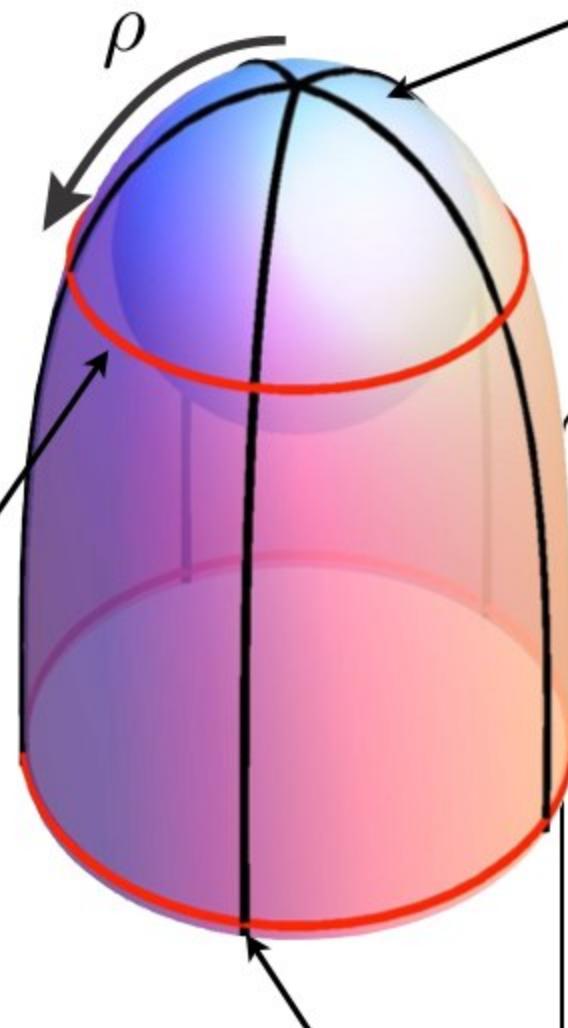
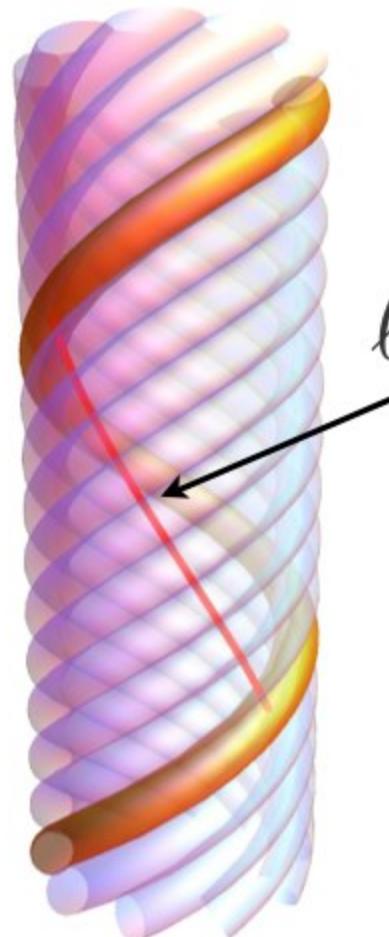
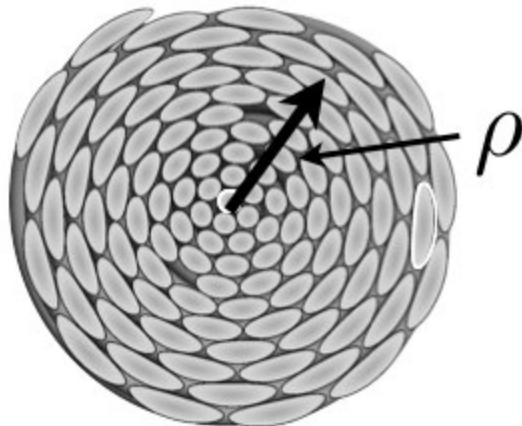


Latitudes on spheres:

$$\ell(\rho) = 2\pi R \sin(\rho/R)$$

$$\simeq 2\pi\rho \left(1 - \frac{1}{6R^2}\rho^2\right)$$

Frustration in filament packing: hidden geometry

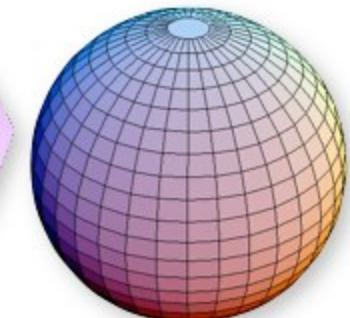
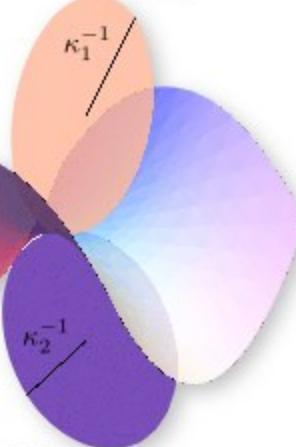


Twisted bundle packing is equivalent to packing on curved surface!

$$\text{spherical radius} = \frac{\Omega^{-1}}{\sqrt{3}}$$
$$K_G = \frac{3\Omega^2}{[1 + (\Omega\rho)^2]^2}$$

Gaussian curvature of “dual surface” to twisted filament bundle

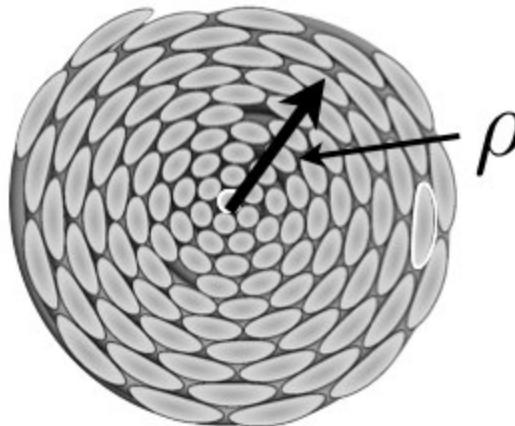
$$K_G = \kappa_1 \kappa_2$$



$$K_G < 0$$

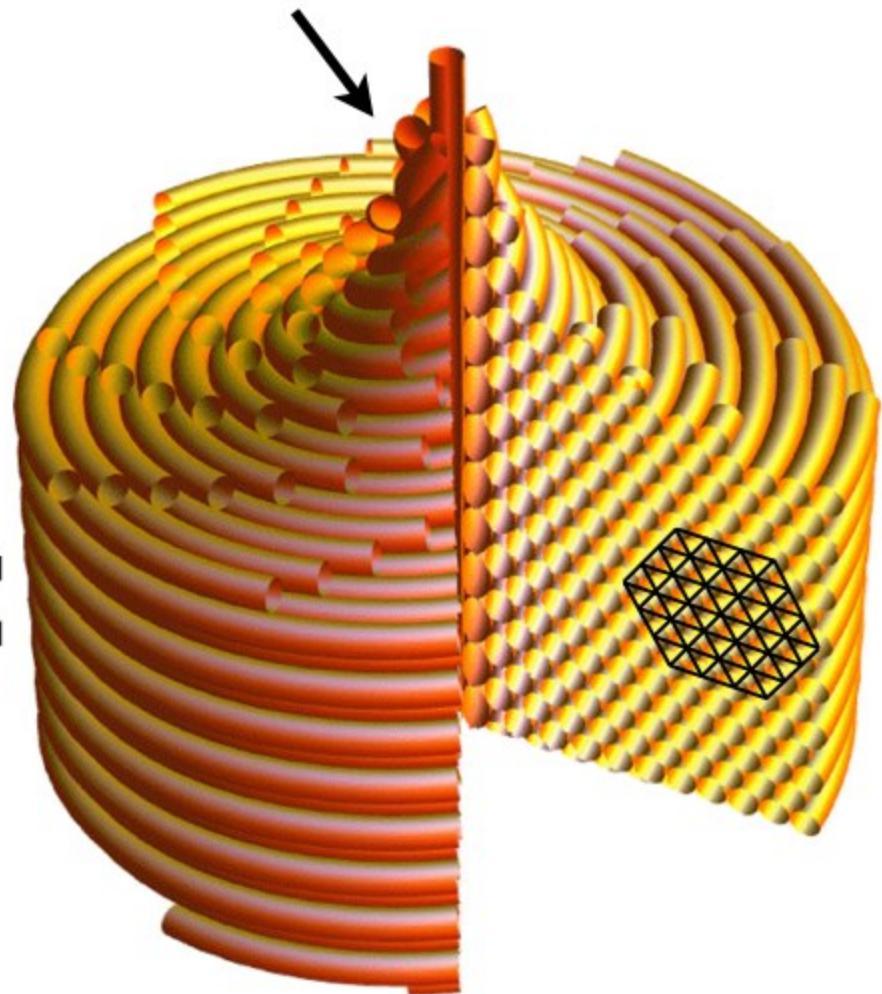
$$K_G > 0$$

Frustration in filament packing: hidden geometry



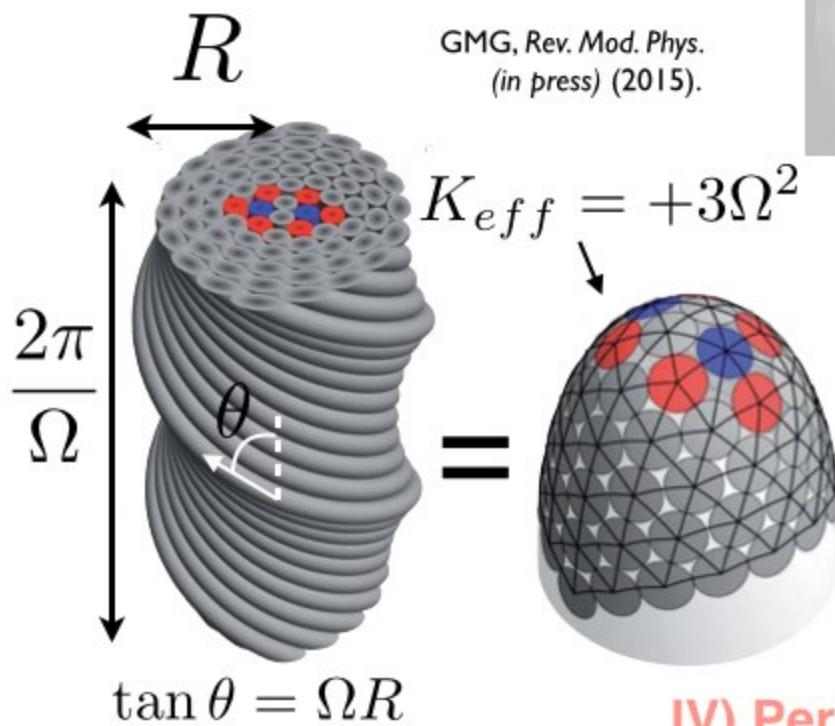
=

Perfect, regular packing
is **frustrated** (disrupted) at
the central core of twisted
bundles

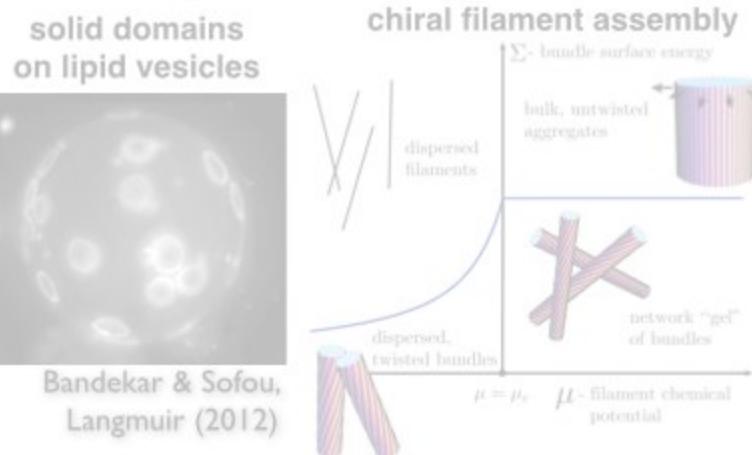


Twisted bundles: non-Euclidean geometry & anomalous assembly

I) Non-euclidean metric geometry of bundles



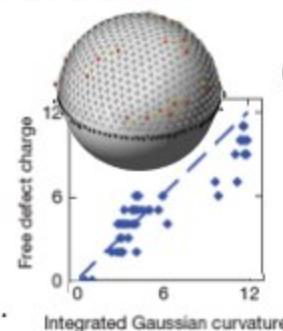
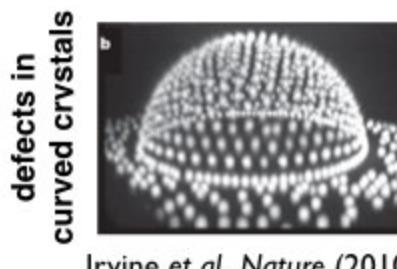
GMG, Rev. Mod. Phys.
(in press) (2015).



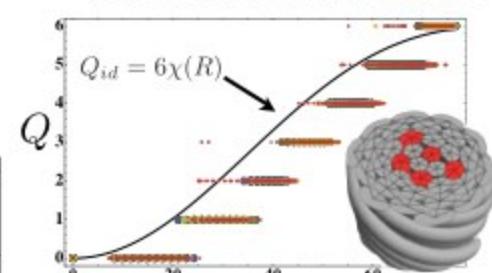
II) Self-limiting Assembly

GMG & Bruinsma,
PRL (2007); GMG,
PRE (2009)

III) Topological Defects

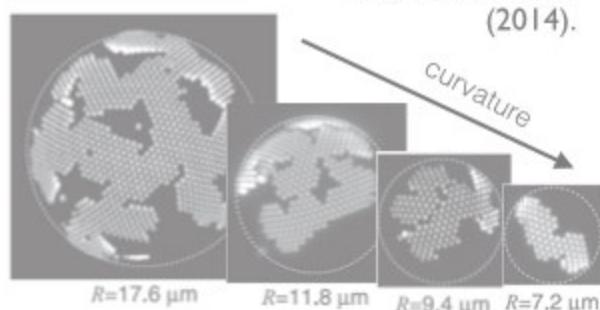
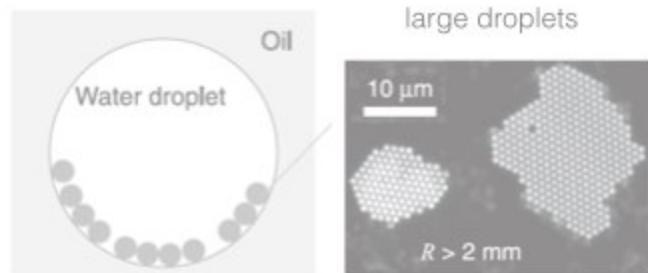


defects in twisted bundles

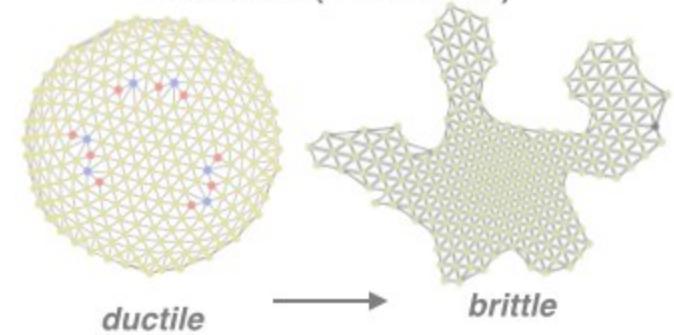


IV) Perimeter Instability & Anisotropic Domains

colloidal crystals on spherical droplets



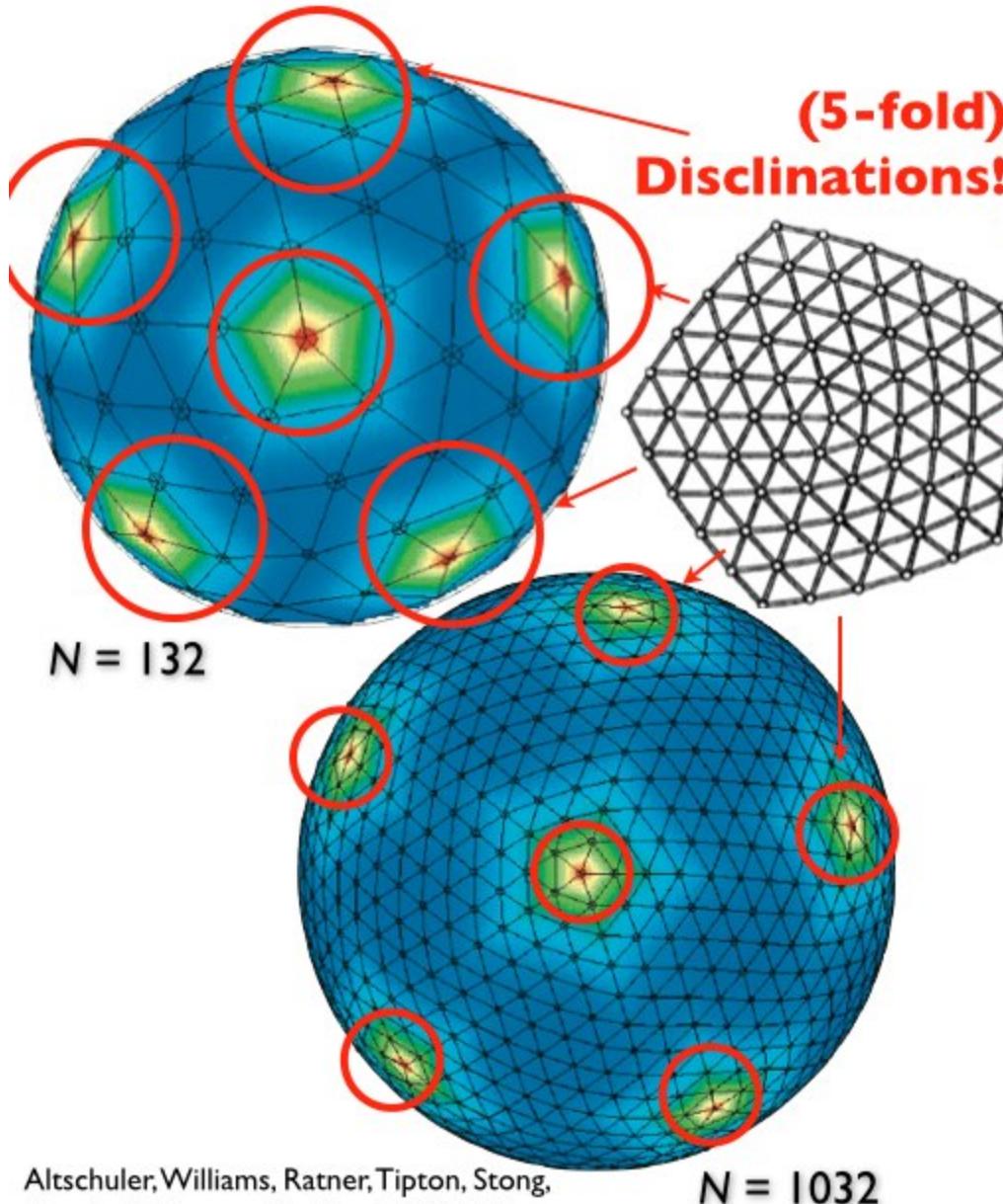
cohesive membranes on spherical substrates (simulations)



Amir Azadi, to be published.

Spherical Crystallography: Generalized Thomson Problem

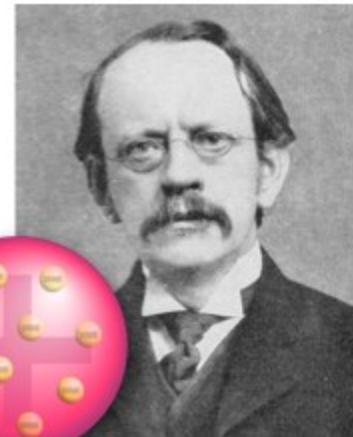
Repulsive particles on spheres



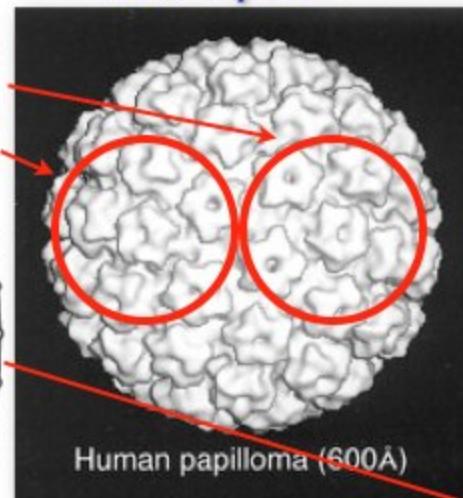
Altschuler, Williams, Ratner, Tipton, Stong,
Dowla & Wooten, *Phys Rev Lett* (1999)

$N = 1032$

“plum pudding”
model:
electronic “corpuscles”
on a charged sphere
Thomson, *Phil. Mag.* (1904).

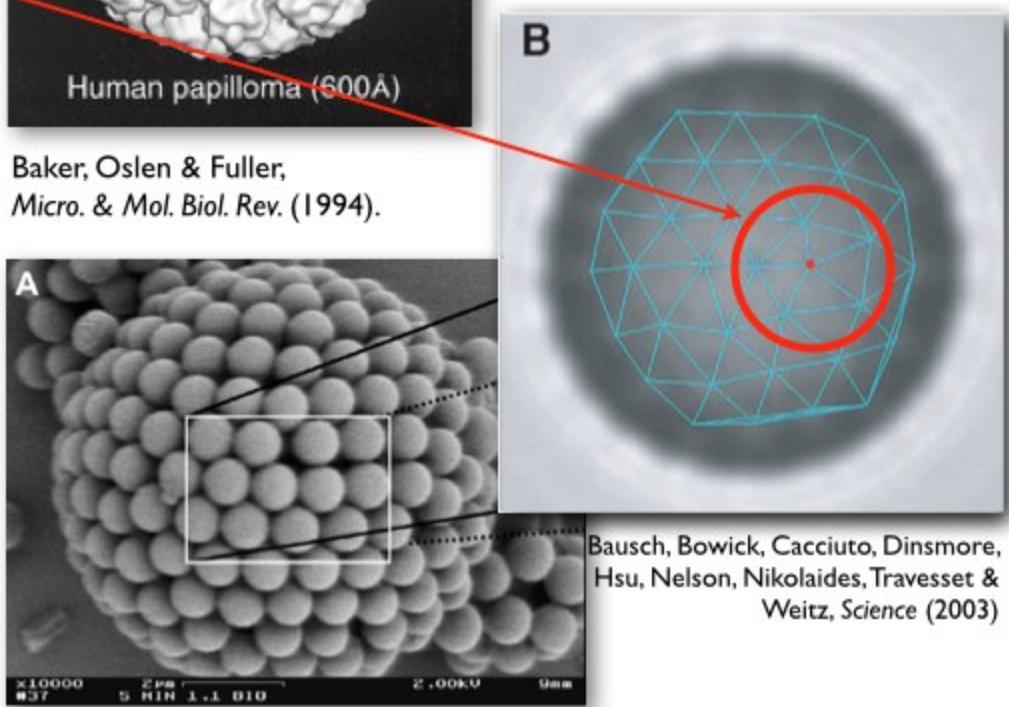


Viral capsids



Baker, Oslen & Fuller,
Micro. & Mol. Biol. Rev. (1994).

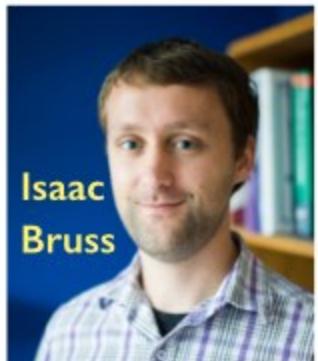
Colloidsomes



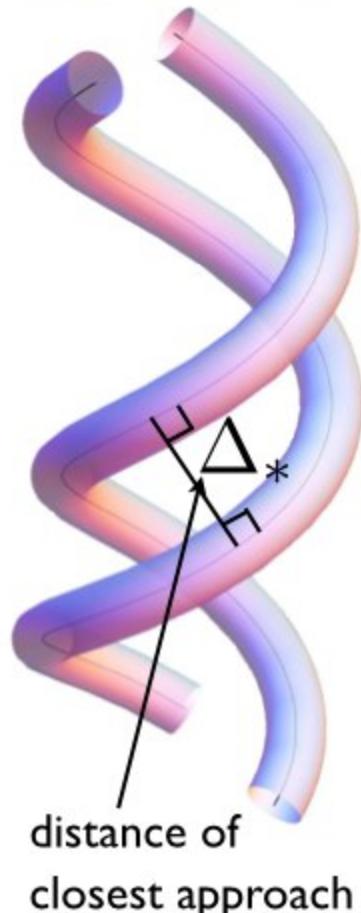
Bausch, Bowick, Cacciuto, Dinsmore,
Hsu, Nelson, Nikolaides, Travesset &
Weitz, *Science* (2003)

Dinsmore, Hsu, Nikolaides, Marquez
& Weitz, *Science* (2002)

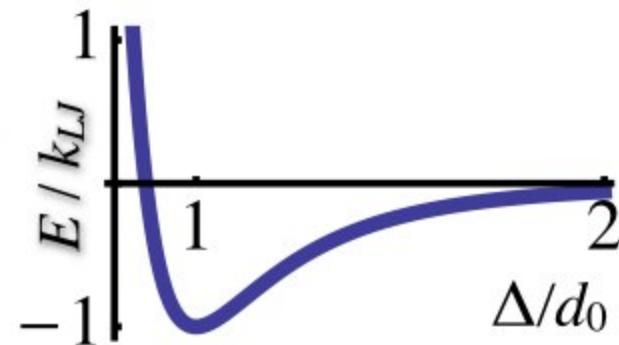
Ground States of Twisted Bundles: Discrete Model



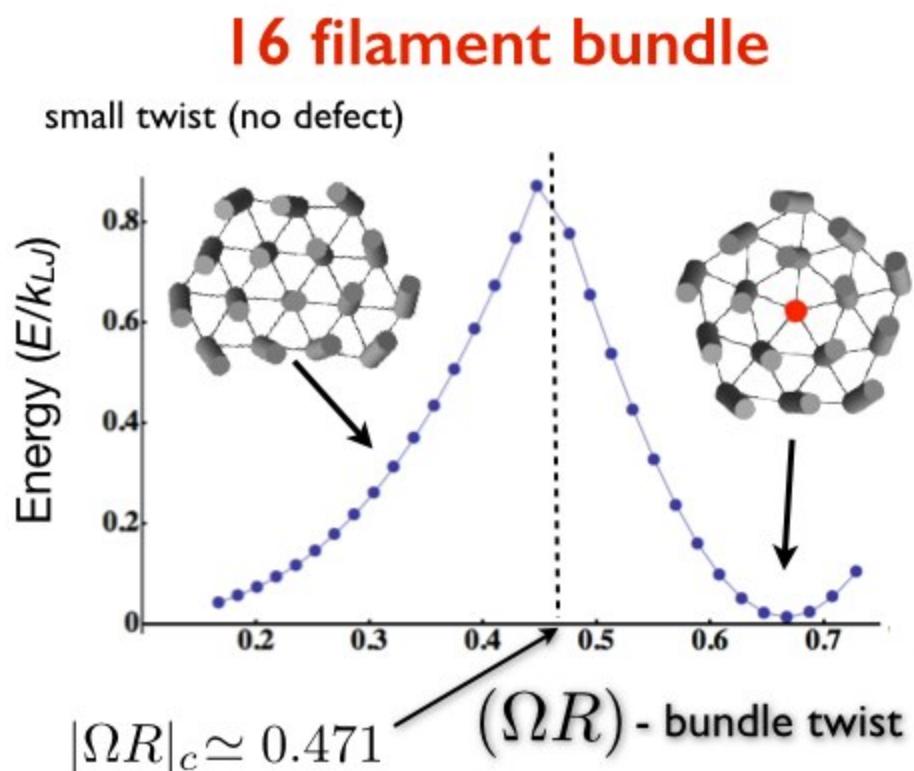
Method: (numerically) minimize 2D cross-section of N filament bundles of fixed twist interacting via attractive, pair-wise forces



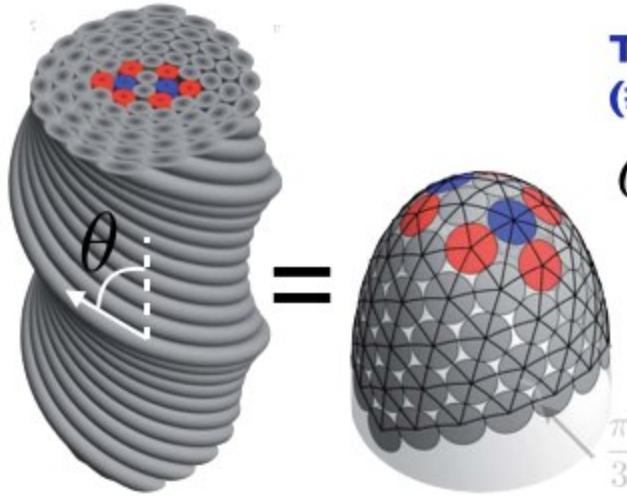
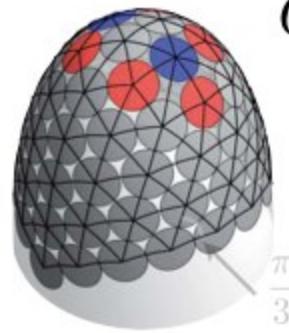
$$E_{LJ} = \frac{k_{LJ}}{2} \left(\frac{5d_o^{12}}{6\Delta^{11}} - \frac{11d_o^6}{6\Delta^5} \right)$$



Pair-wise adhesive energy
("Curva-Lennard Jones")



Defects in Ground States of Small- N Bundles

 $=$ 

Total disclination “charge”
(# 5’s - #7’s):

$$Q = \sum_n (n - 6)V_n$$

of n -fold
coordinated filaments

$\frac{\pi}{3} + \Delta\theta_b$
“equilateral excess”

Generalized Euler-Poincare formula
(triangulation w/open boundary):

$$6 \int dA K_G = 2\pi Q + \sum_b \cancel{\Delta\theta_b}$$

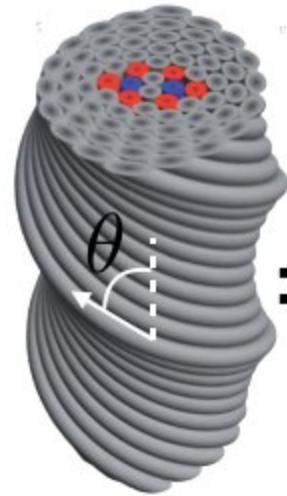
distortion
@ boundary

“Net neutral” disclination charge:

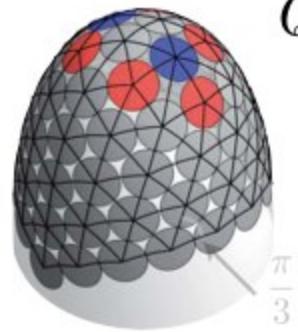
$$Q_{id} = \frac{3}{\pi} \int dA K_{\text{eff}} = 6(1 - \cos^3 \theta)$$

bundle twist angle

Defects in Ground States of Small- N Bundles



=



Total disclination "charge"
(# 5's - #7's):

$$Q = \sum_n (n - 6) V_n$$

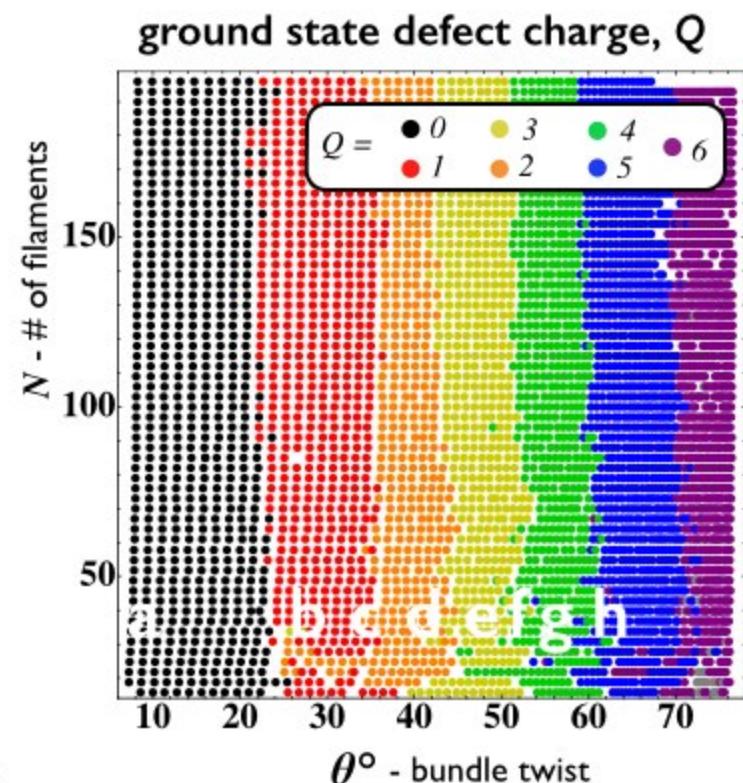
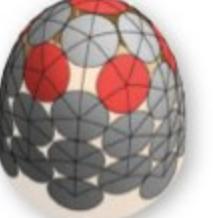
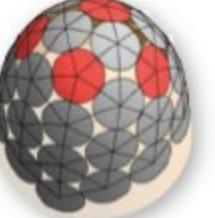
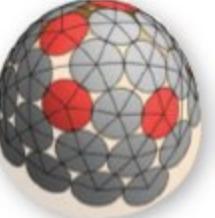
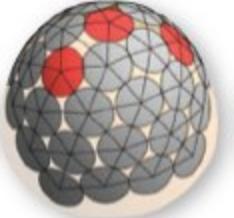
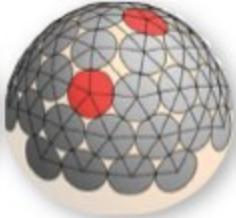
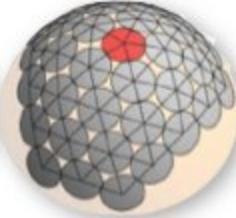
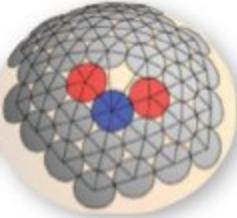
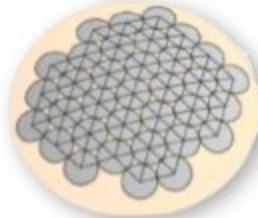
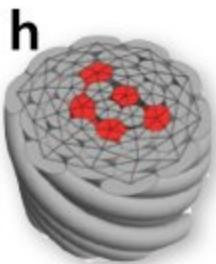
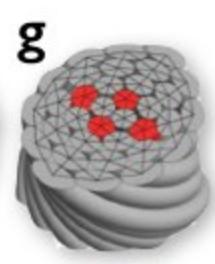
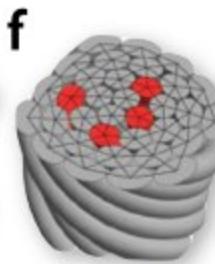
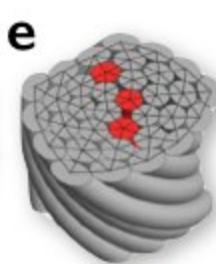
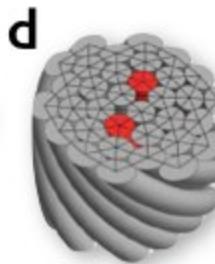
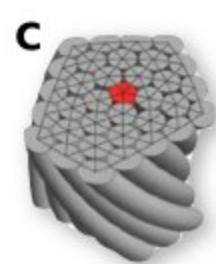
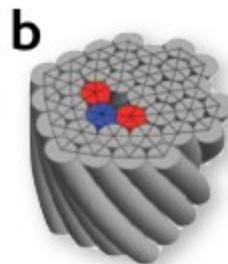
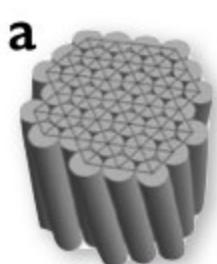
of n -fold
coordinated filaments

$\frac{\pi}{3} + \Delta\theta_b$
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"Net neutral" disclination charge:

$$Q_{id} = \frac{3}{\pi} \int dA K_{\text{eff}} = 6(1 - \cos^3 \theta)$$

bundle twist angle

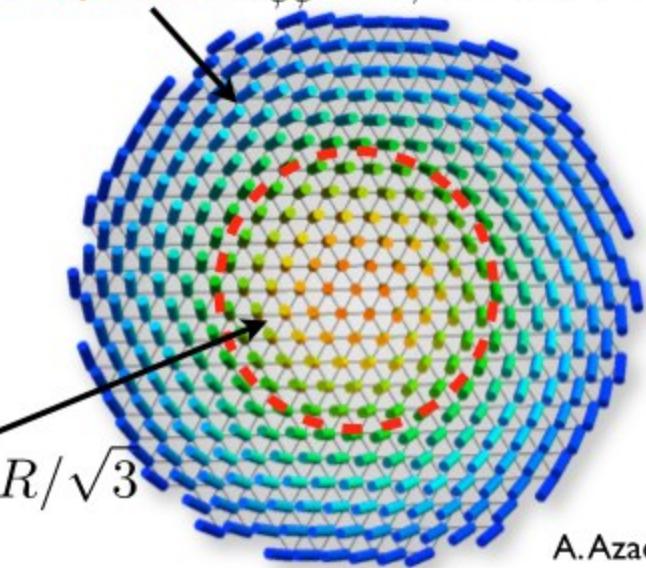


Large- N Bundles: Multi-dislocation ground states



twist-induced stresses:

compression: $\sigma_{\phi\phi} < 0$; $\rho > R/\sqrt{3}$

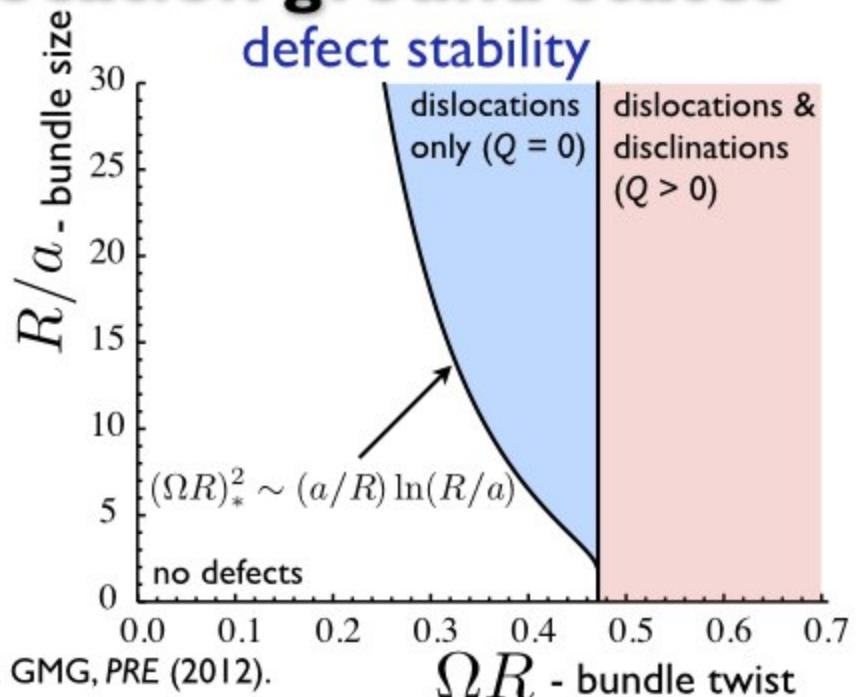
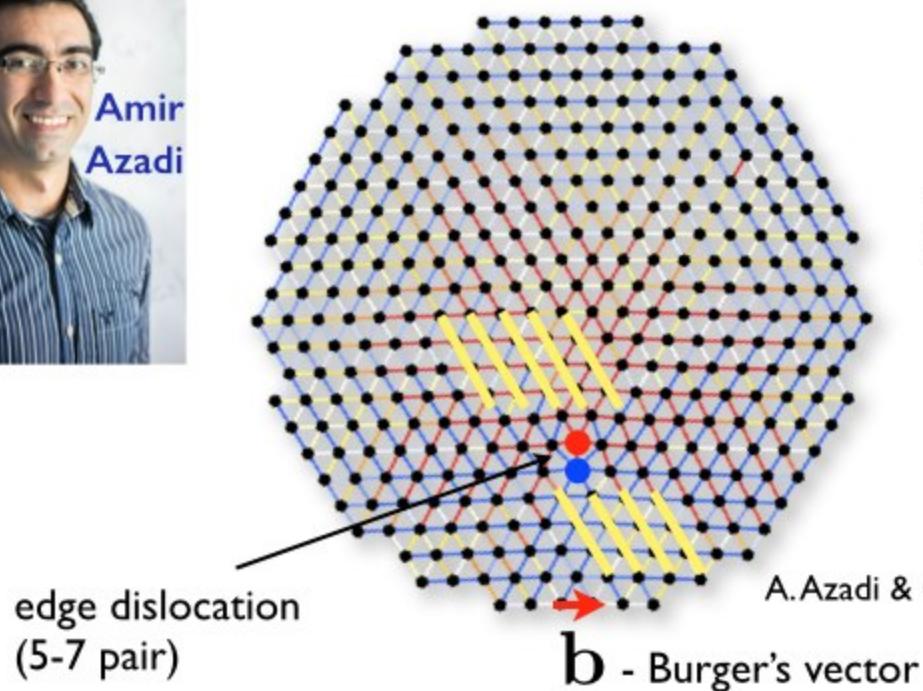
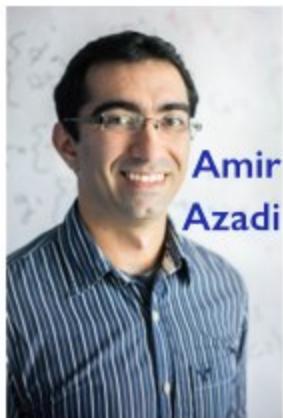


tension:

$\sigma_{\phi\phi} > 0$; $\rho < R/\sqrt{3}$

A.Azadi & GMG, PRE (2012).

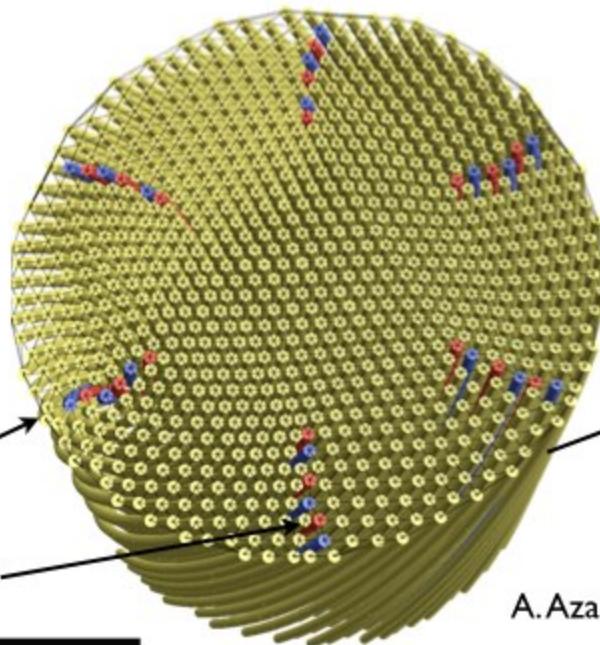
Large- N Bundles: Multi-dislocation ground states



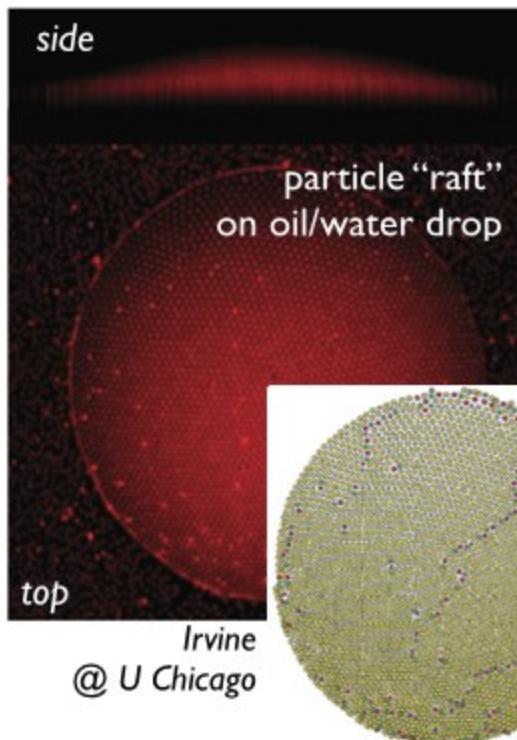
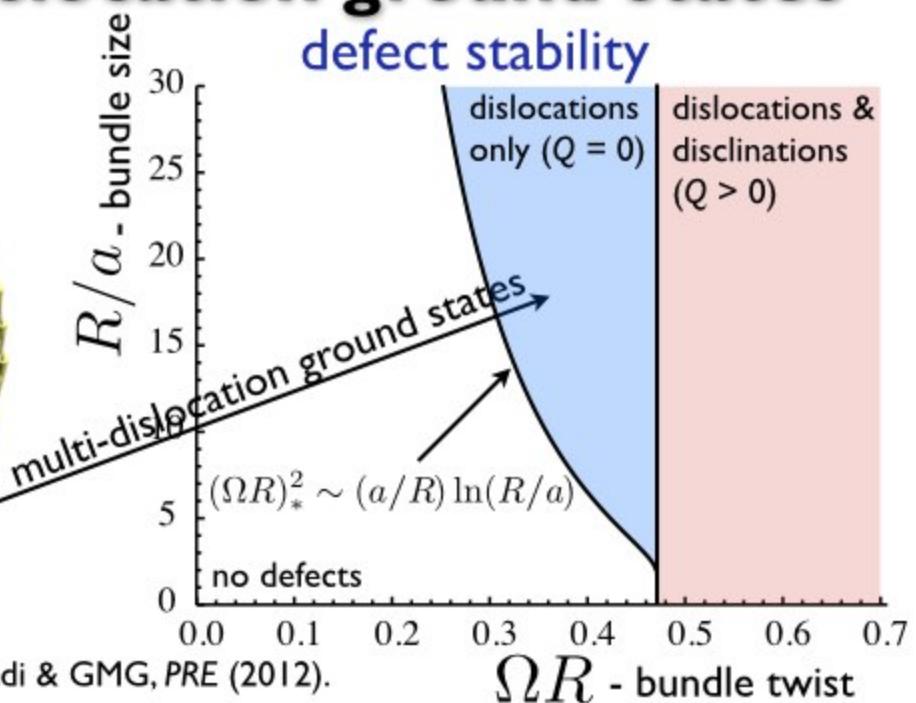
Large- N Bundles: Multi-dislocation ground states



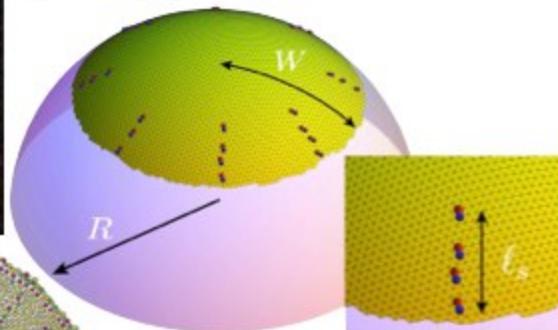
low-angle grain boundary “scars”



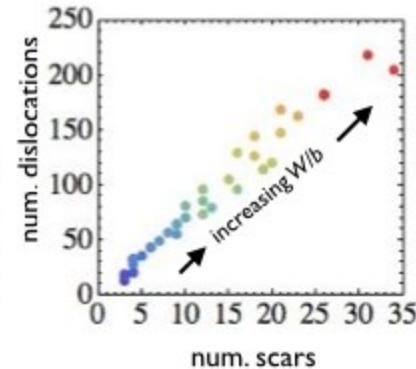
A. Azadi & GMG, PRE (2012).



Optimal symmetry of multi-dislocation (scar) patterns?

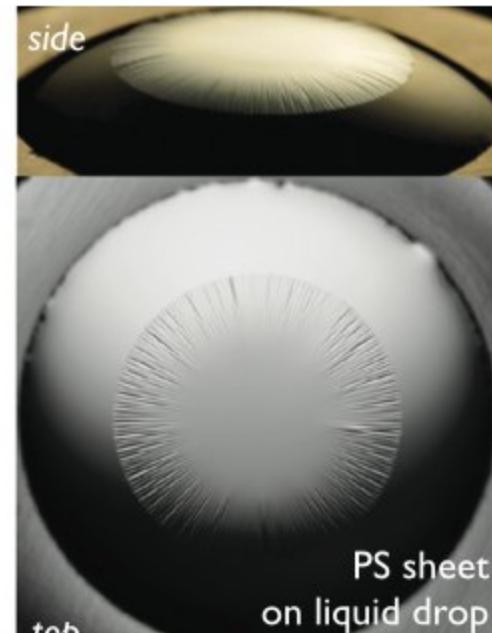


A. Azadi & GMG, PRL (2014).



Ground states of surface confined assemblies: “elastic” wrinkle vs. “plastic” defect patterns?

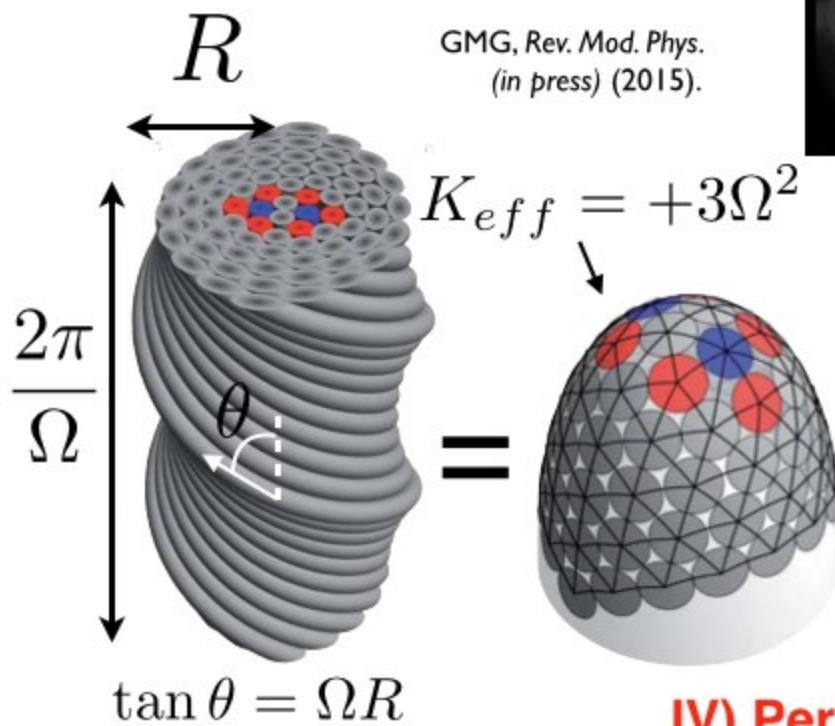
GMG & Davidovitch, PNAS (2013).



Menon @ UMass

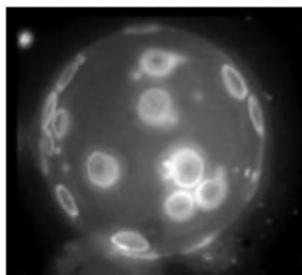
Twisted bundles: non-Euclidean geometry & anomalous assembly

I) Non-euclidean metric geometry of bundles



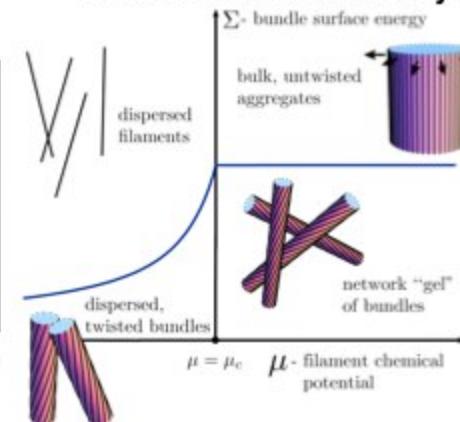
GMG, Rev. Mod. Phys.
(in press) (2015).

solid domains
on lipid vesicles



Bandekar & Sofou,
Langmuir (2012)

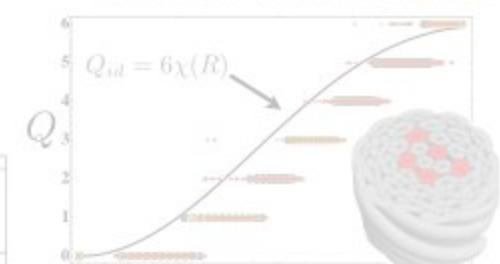
chiral filament assembly



II) Self-limiting Assembly

GMG & Bruinsma,
PRL (2007); GMG,
PRE (2009)

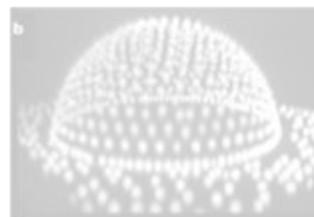
defects in twisted bundles



Bruss & GMG, PNAS
(2012); Soft Matter (2013).

III) Topological Defects

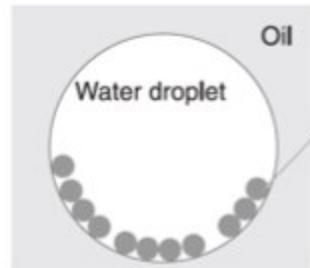
defects in
curved crystals



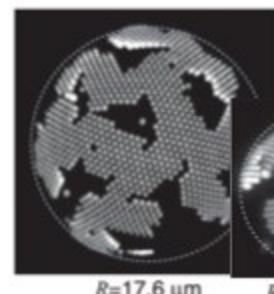
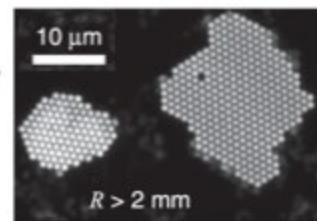
Irvine et al., Nature (2010).

IV) Perimeter Instability & Anisotropic Domains

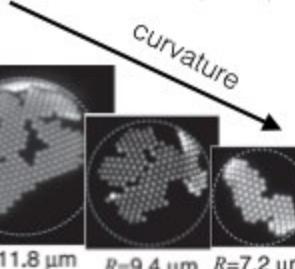
colloidal crystals on spherical droplets



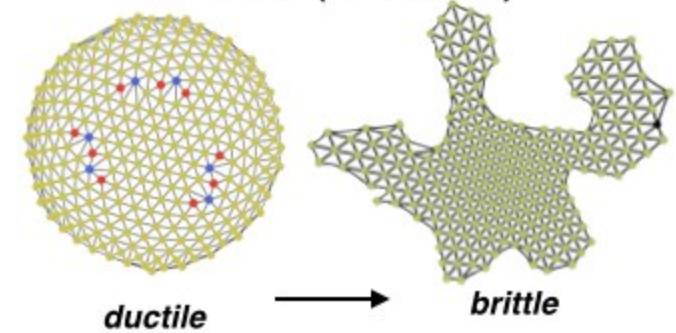
large droplets



Meng et al., Science
(2014).



cohesive membranes on spherical
substrates (simulations)



Amir Azadi, to be published.

Elastic Perimeter Instability of Curved Crystals

EUROPHYSICS LETTERS

1 April 2005

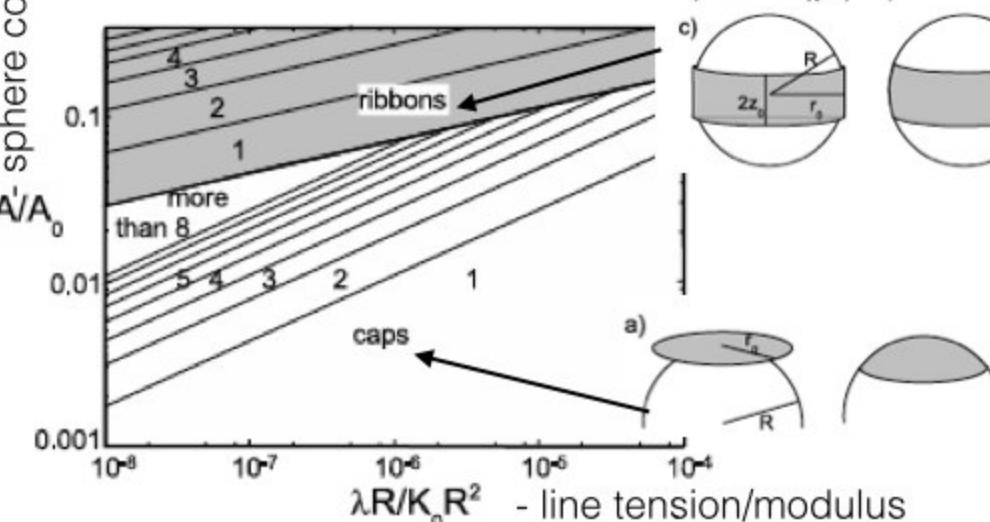
Europ. Lett., 70 (1), pp. 136–142 (2005)

DOI: 10.1209/epl/i2004-10464-2

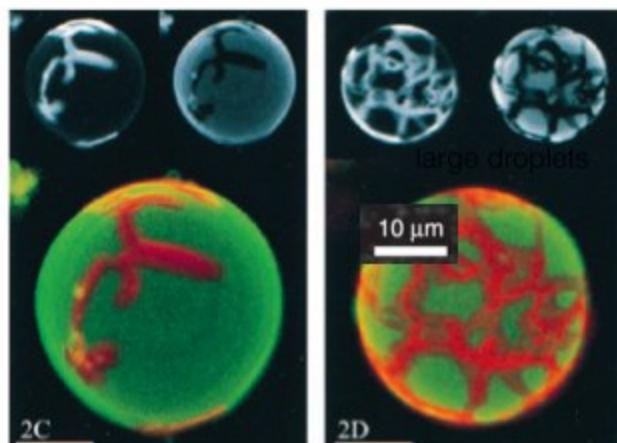
Shapes of crystalline domains on spherical fluid vesicles

S. SCHNEIDER and G. GOMPPER

Institut für Festkörperforschung - Forschungszentrum Jülich, D-52095 Jülich, Germany



lipid solid-fluid coexistence on GUVS

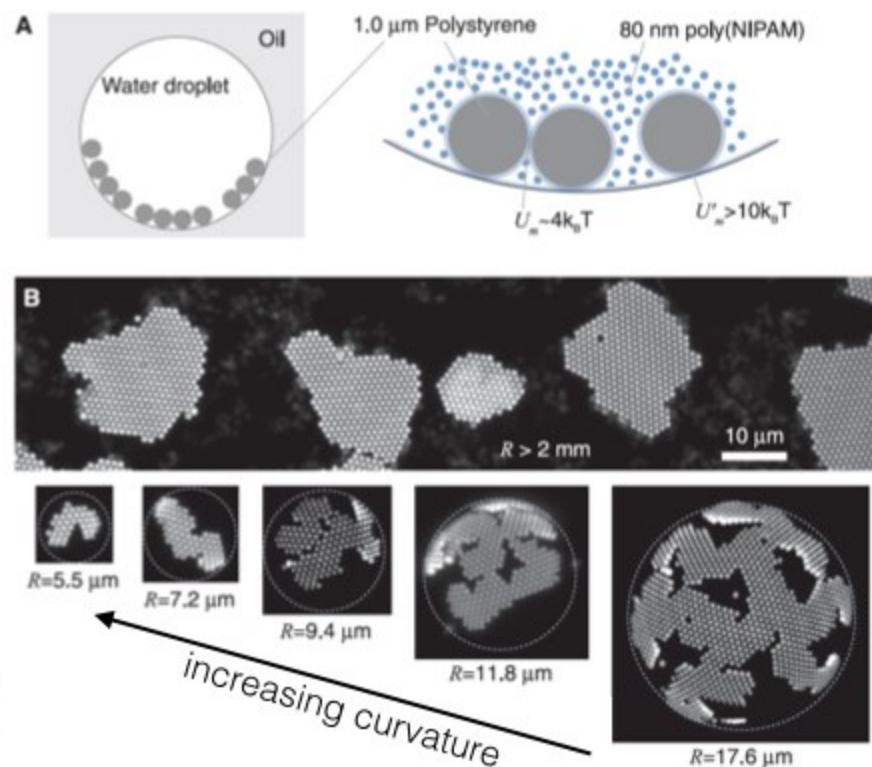


Webb *et al.*
PNAS (1999).

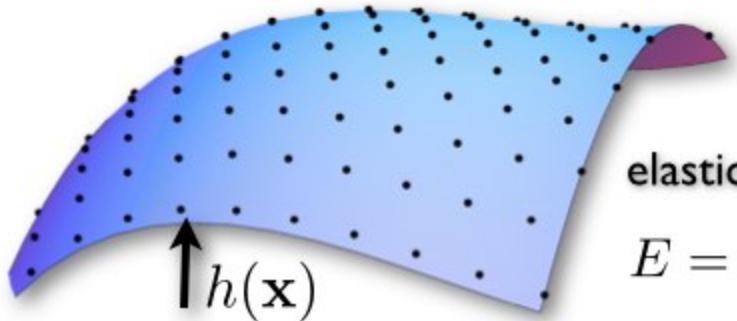
Elastic Instability of a Crystal Growing on a Curved Surface

Guangnan Meng,¹ Jayson Paulose,² David R. Nelson,^{3,2} Vinodhan N. Manoharan^{2,1*}

7 FEBRUARY 2014 VOL 343 SCIENCE



Curved Membranes vs. Twisted Bundles: Elasticity



membrane displacement

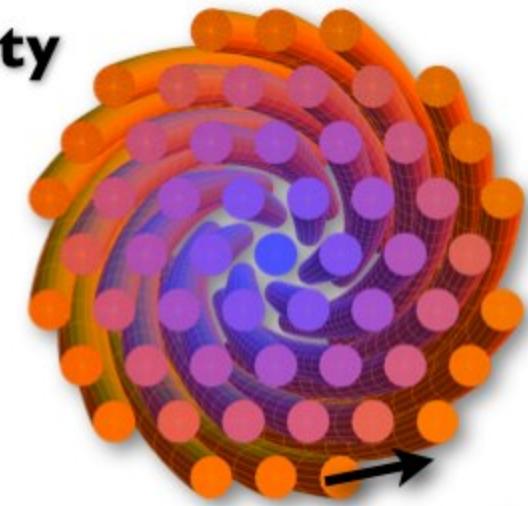
in-plane stress:

$$\sigma_{ij} = \lambda \delta_{ij} u_{kk} + 2\mu u_{ij}$$

elastic energy:

$$E = \frac{1}{2} \int dV \sigma_{ij} u_{ij} ; \quad Y = \frac{4\mu(\lambda + \mu)}{\lambda + 2\mu}$$

Young's modulus:



non-linear strain

$$u_{ij} = \frac{1}{2} (\partial_i u_j + \partial_j u_i + \partial_i h \partial_j h)$$

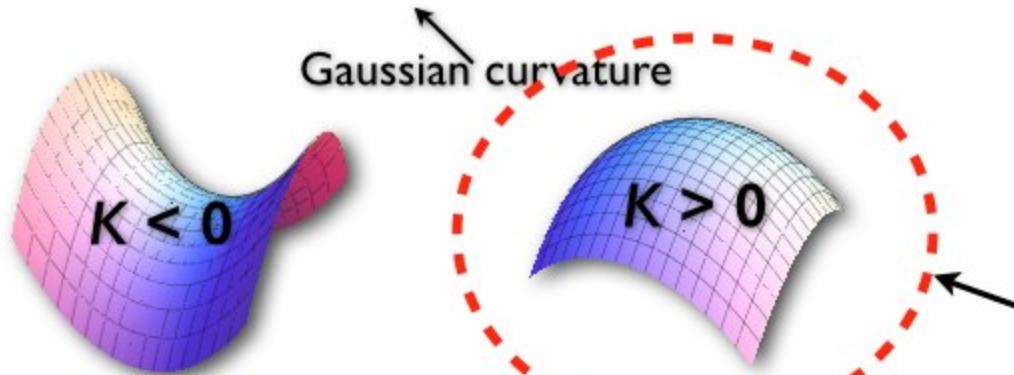
$$u_{ij}^\perp = \frac{1}{2} (\partial_i u_j + \partial_j u_i - t_i t_j)$$

Compatibility relation

$$\frac{\nabla_\perp^2 \sigma_{ii}}{Y} = -(\partial_x^2 h)(\partial_y^2 h) + (\partial_x \partial_y h)^2 \\ = -K$$

$$\frac{\nabla_\perp^2 \sigma_{ii}}{Y} = -\frac{1}{2} [(\partial_x t_y)^2 + (\partial_y t_x)^2 - 2\partial_x \partial_y (t_x t_y)]$$

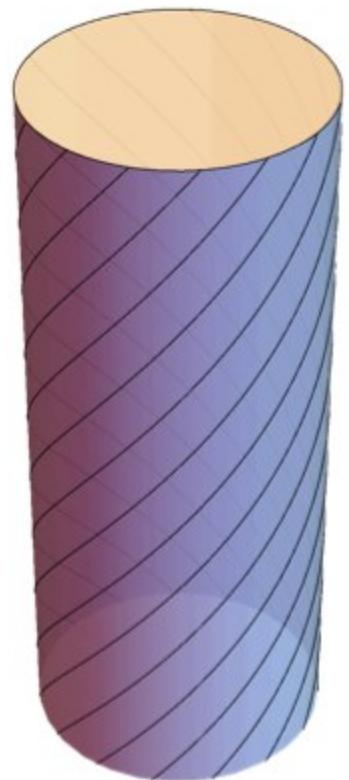
$$\equiv -K_{eff} = -3\Omega^2$$



Bundle twist generates interfilament stress identical to **positive** Gaussian curvature in 2D membranes

Isotropic, Twisted Cylindrical Bundles: Thermodynamics

<i>intra-filament (bending) elasticity</i>	<i>inter-filament (packing) elasticity</i>	<i>surface energy (cohesion)</i>	
$\frac{B\rho_0}{2} \int dV \kappa^2(r)$	$\frac{1}{2} \int dV \sigma_{ij} u_{ij}$	$\Sigma(2\pi RL)$	R - bundle radius
$\kappa(r) \simeq \Omega^2 r$ filament curv.	$\sigma_{ii} \approx -Y(\Omega R)^2$ geometric strain		Ω - twist ($2\pi/\text{pitch}$)



Isotropic, Twisted Cylindrical Bundles: Thermodynamics

intra-filament
(bending) elasticity

inter-filament
(packing) elasticity

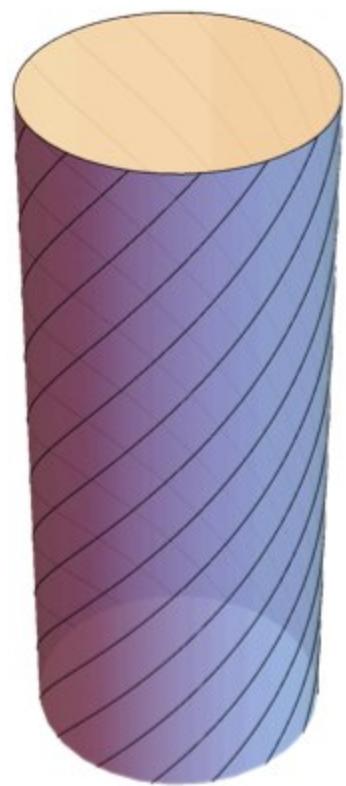
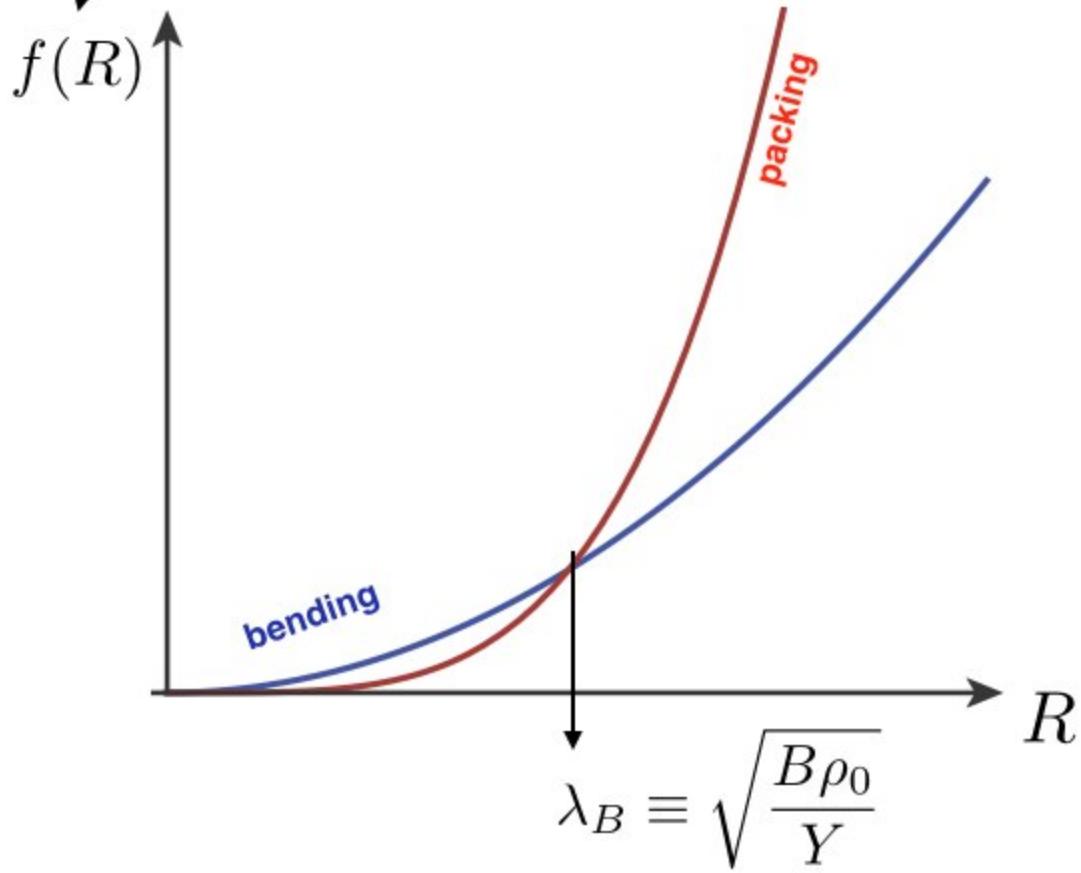
surface energy
(cohesion)

R - bundle radius

Ω - twist ($2\pi/\text{pitch}$)

$$f(R) = \frac{B\rho_0}{4}\Omega^4 R^2 + \frac{3Y}{128}(\Omega R)^4 + \frac{2\Sigma}{R}$$

free-energy density



Isotropic, Twisted Cylindrical Bundles: Thermodynamics

*intra-filament
(bending) elasticity*

*inter-filament
(packing) elasticity*

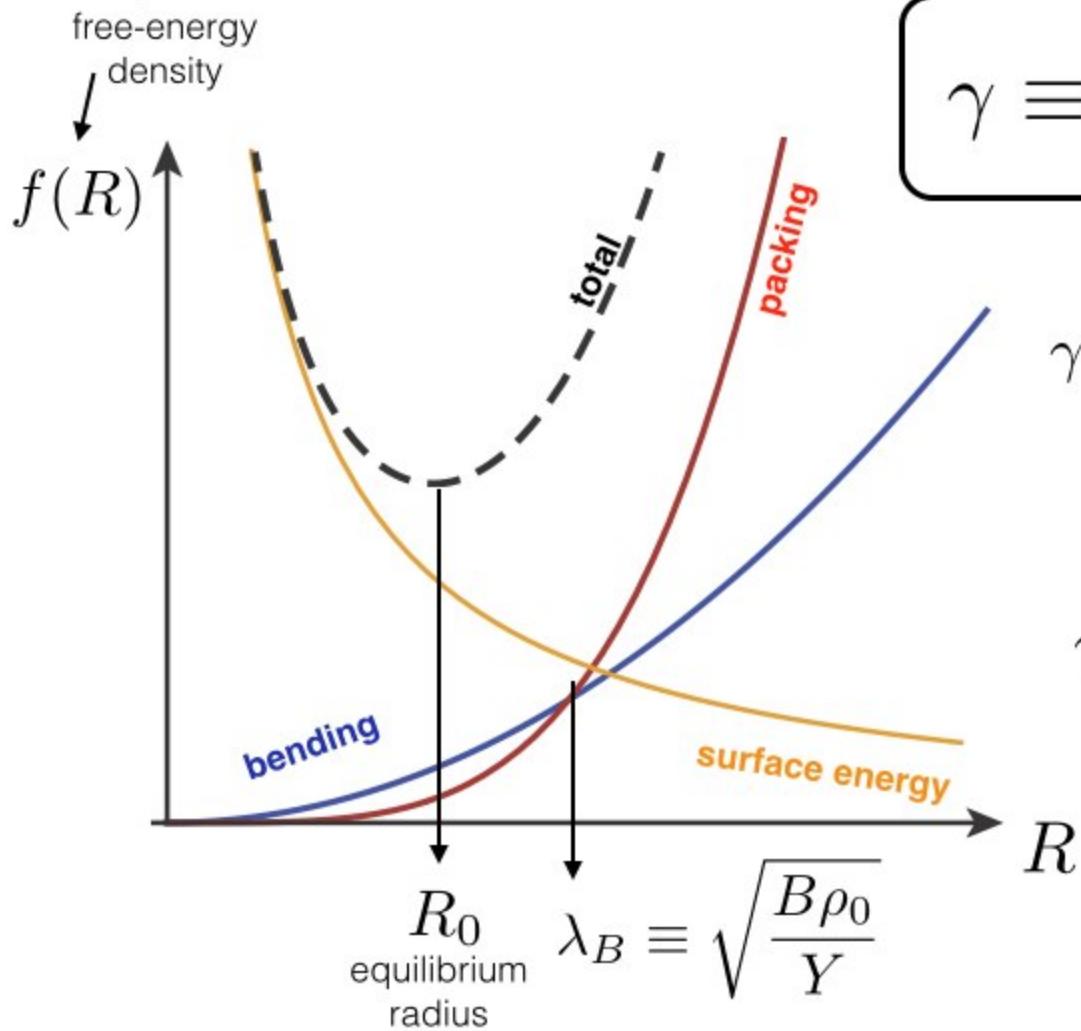
*surface energy
(cohesion)*

R - bundle radius

Ω - twist ($2\pi/\text{pitch}$)

$$f(R) = \frac{B\rho_0}{4}\Omega^4 R^2 + \frac{3Y}{128}(\Omega R)^4 + \frac{2\Sigma}{R}$$

$$\gamma \equiv (R_0/\lambda_B)^2 = \frac{\text{"packing" cost}}{\text{bending cost}}$$

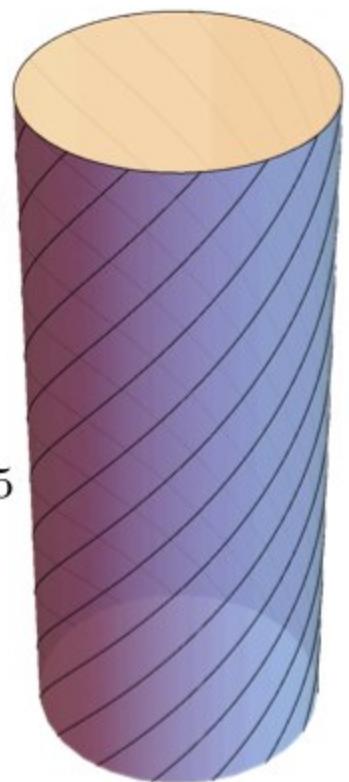


$\gamma \ll 1$: **bending** limited

$$R_0 \sim (\Sigma P^4 / B)^{1/3}$$

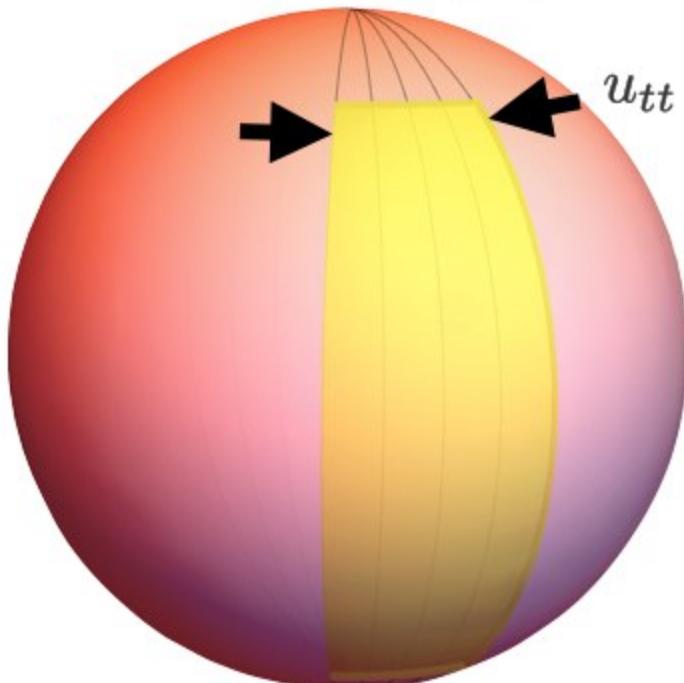
$\gamma \gg 1$: **packing** limited

$$R_0 \sim (\Sigma R^4 / Y)^{1/5}$$



Anisotropic, Helical “Tapes”: Inter- vs. Intra-filament Strain

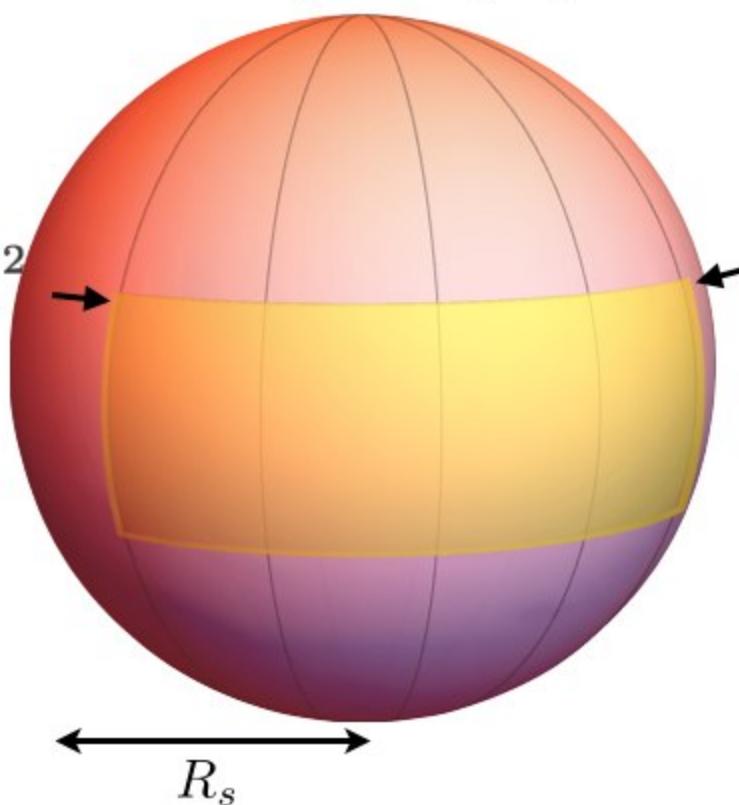
width preserving map



$$u_{tt} \approx -(w/R_s)^2$$

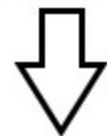
strain grows with wide dimension \rightarrow **large elastic energy**

thickness preserving map



$$u_{ww} \approx -(t/R_s)^2$$

strain grows with thin dimension



low elastic energy



Anisotropic, Helical “Tapes”: Inter- vs. Intra-filament Strain

<i>intra-filament (bending) elasticity</i>	<i>inter-filament (packing) elasticity</i>	<i>surface energy (cohesion)</i>
$f(w \gg t) \simeq \frac{B\rho_0}{24}\Omega^4 w^2 + \frac{Y}{160}(\Omega t)^4 + 2\Sigma\left(\frac{1}{w} + \frac{1}{t}\right)$	$\kappa \approx \Omega^2 w$	$u_{ww} \approx -(t/R_s)^2$

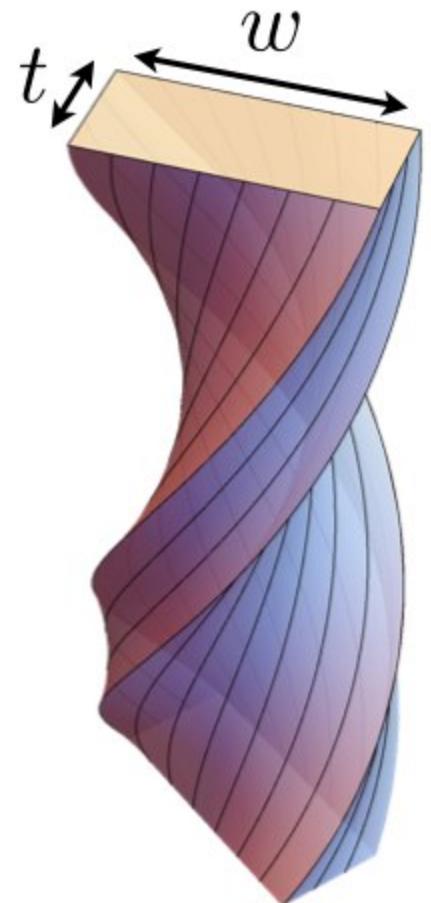
tape width: **bending** limited

$$w_0 = 24^{1/3} \left(\frac{\Sigma}{\Omega^4 B \rho_0} \right)^{1/3}$$

equilibrium dimensions

tape thickness: **packing** limited

$$t_0 = 80^{1/5} \left(\frac{\Sigma}{\Omega^4 Y} \right)^{1/5}$$

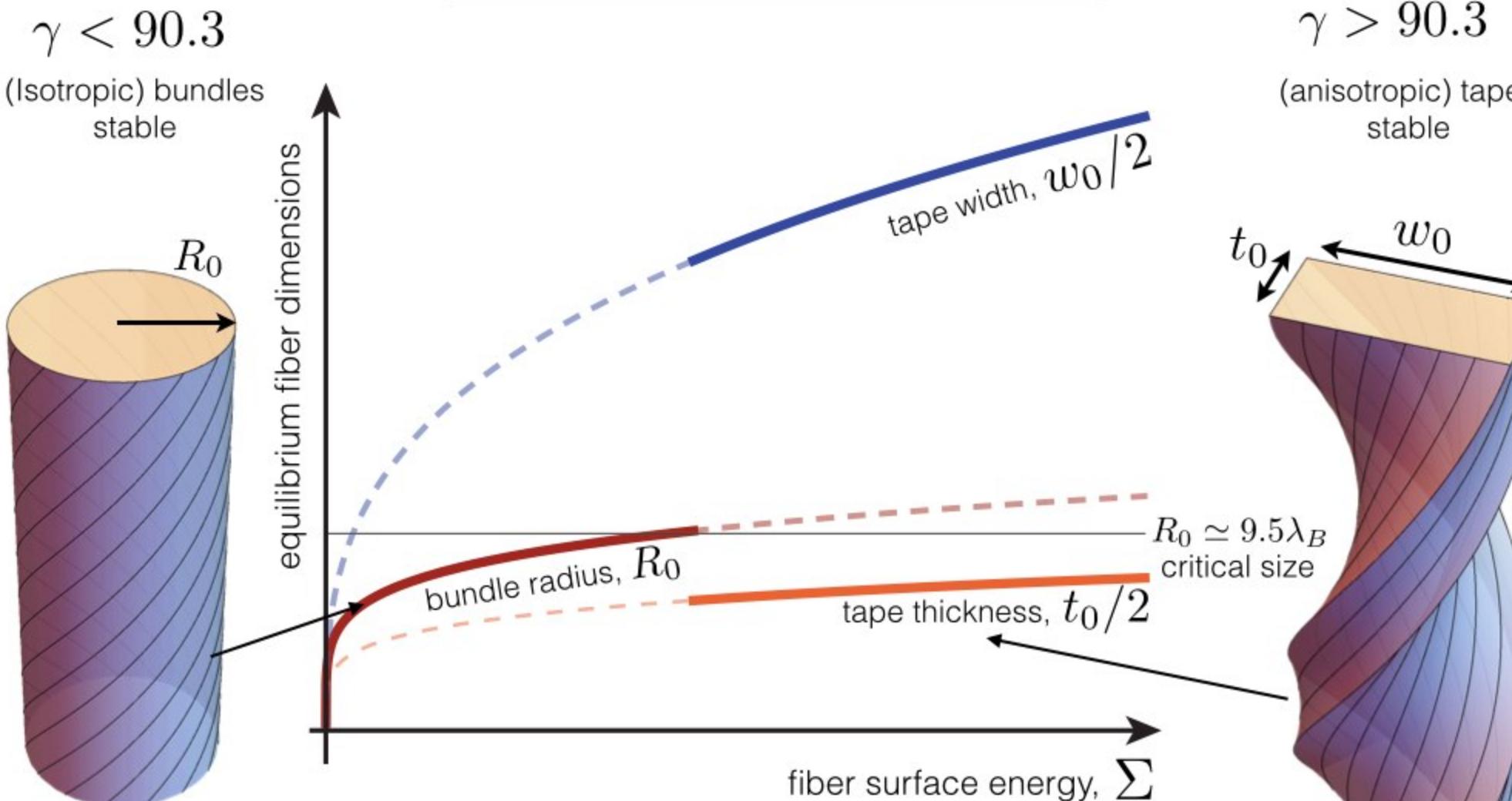
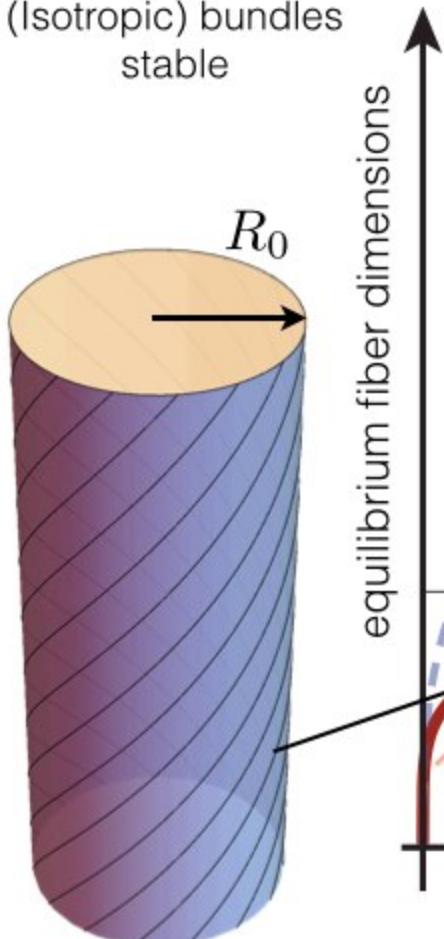


From Bundles to Tapes: Critical Bundle Size

$$\gamma \equiv (R_0/\lambda_B)^2 = \frac{\text{"packing" cost}}{\text{bending cost}}$$

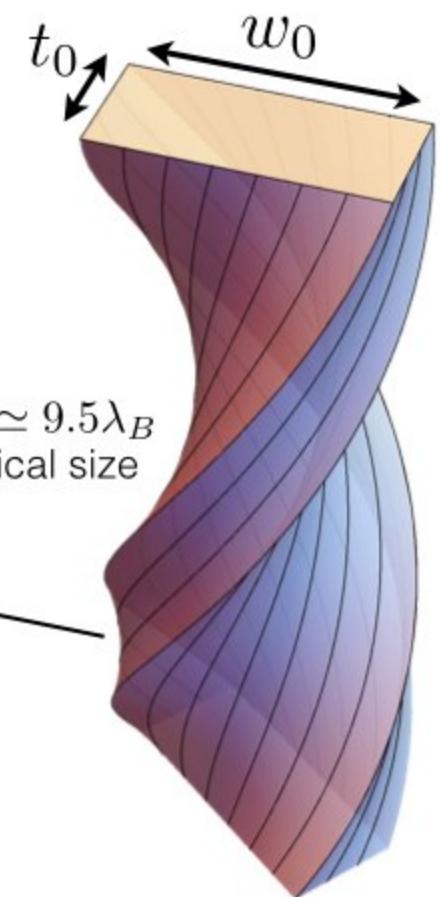
$\gamma < 90.3$

(isotropic) bundles
stable



$\gamma > 90.3$

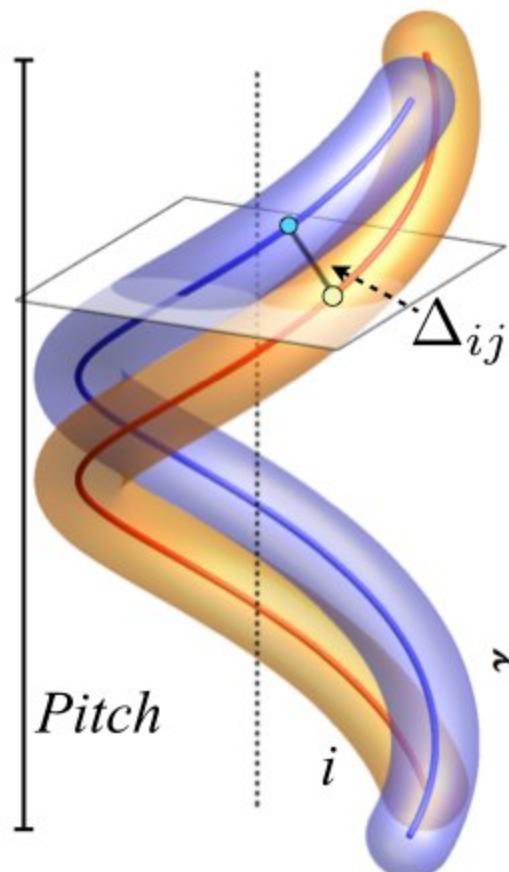
(anisotropic) tapes
stable



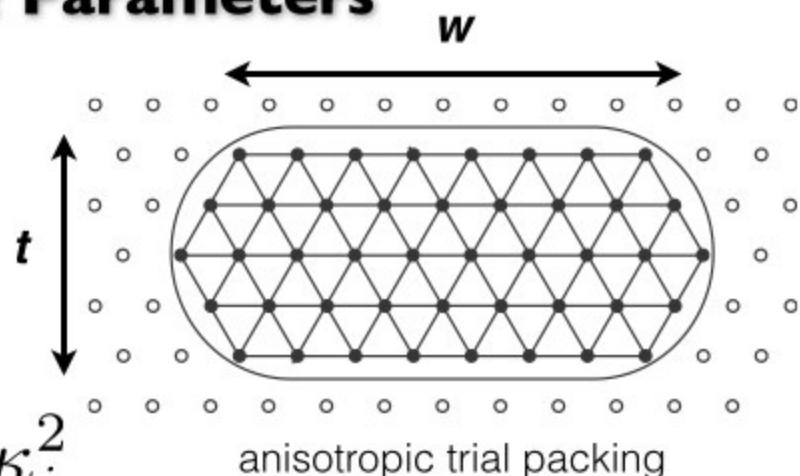
Discrete filament simulations: Model & Parameters



Doug Hall (UMass)



Method: (numerically) optimize 2D cross-section and N of filament bundles of fixed twist interacting via attractive, pair-wise forces



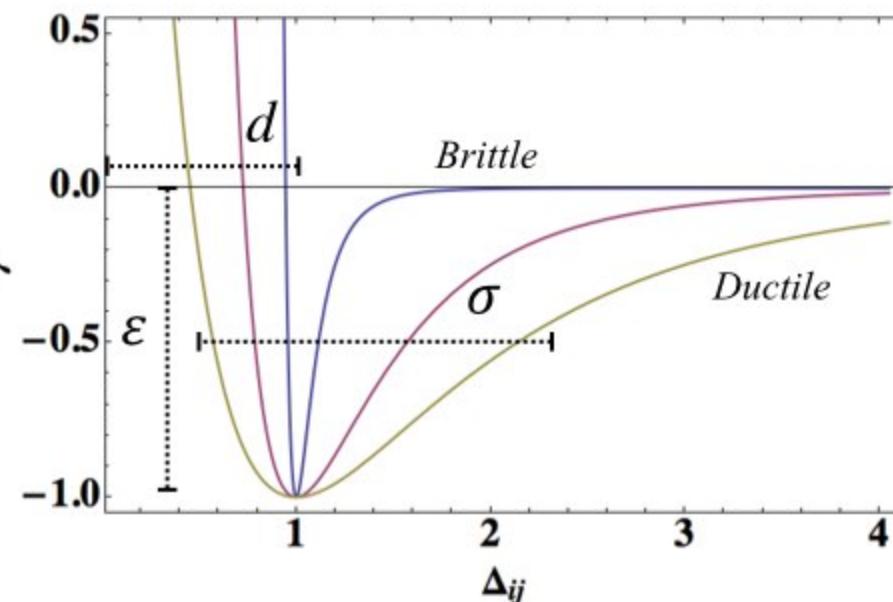
$$E_{Bend} = \frac{B}{2} L \sum_i \kappa_i^2$$

Curvature

$$E_{co} = \sum_{ij} L_{ij} \gamma(\Delta_{ij})$$

Length

Energy per unit length



$$\lambda_B \equiv \sqrt{\frac{B\rho_0}{Y}}$$

$$\propto (B/\epsilon)^{1/2} (d/\sigma)$$

$$\lambda_S \equiv \Sigma/Y$$

$$\propto d(\sigma/d)^2$$

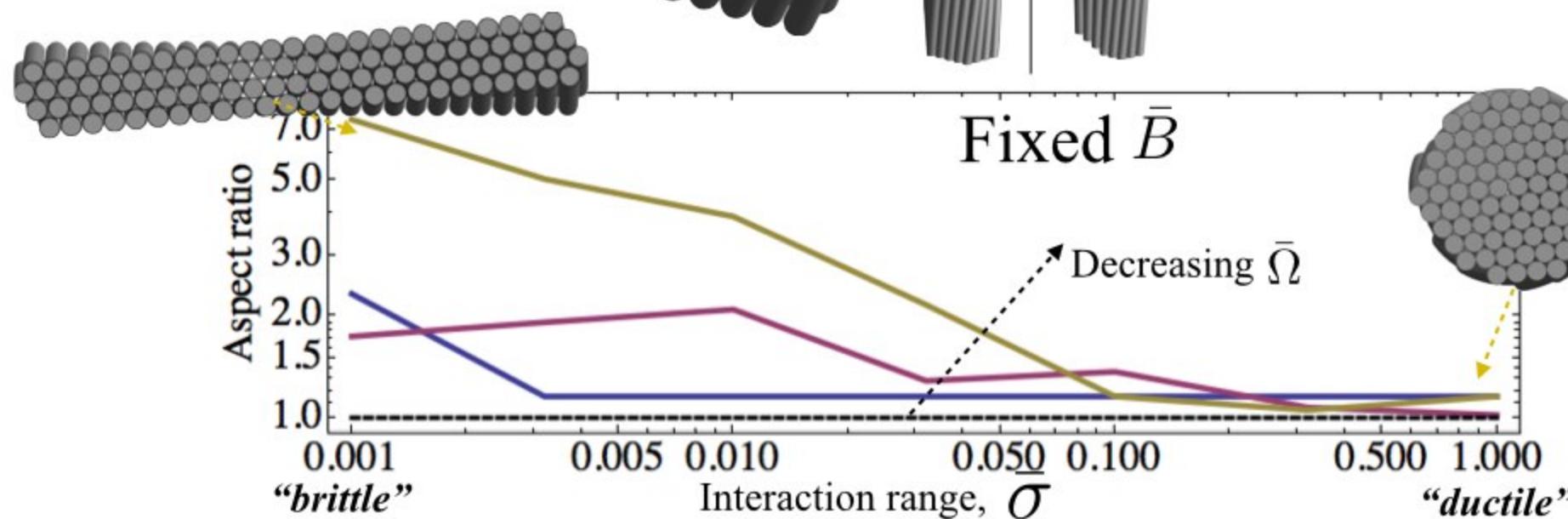
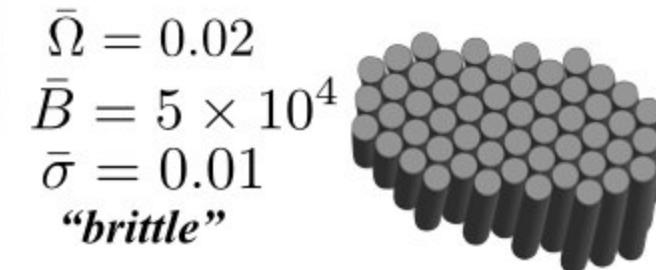
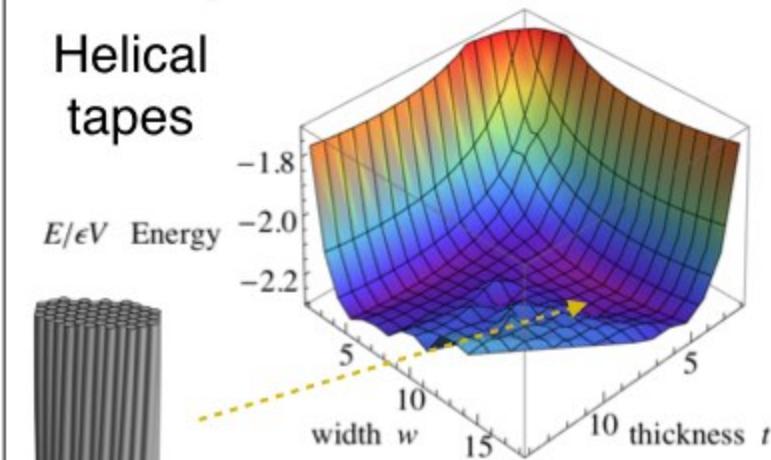
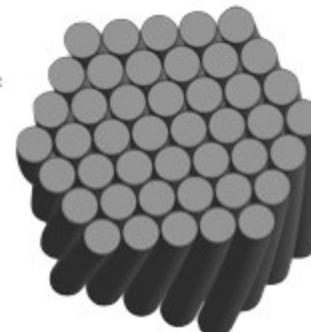
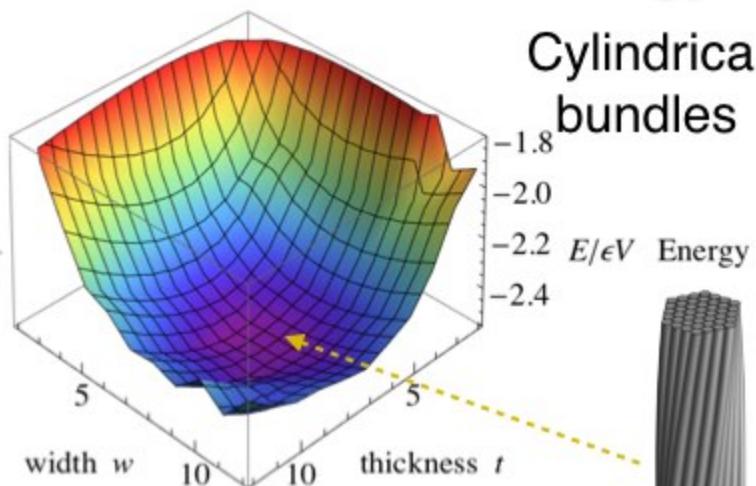
Discrete filament simulations: Energy Landscapes & Phase Diagram

scaled parameters:

$$\bar{B} = \frac{B}{\epsilon d^2}$$

$$\bar{\sigma} = \frac{\sigma}{d}$$

$$\bar{\Omega} = 2\pi d/\text{Pitch}$$



Twisted Amyloid Tapes: From Mesoscale Dimensions to “Molecular” Scale Parameters

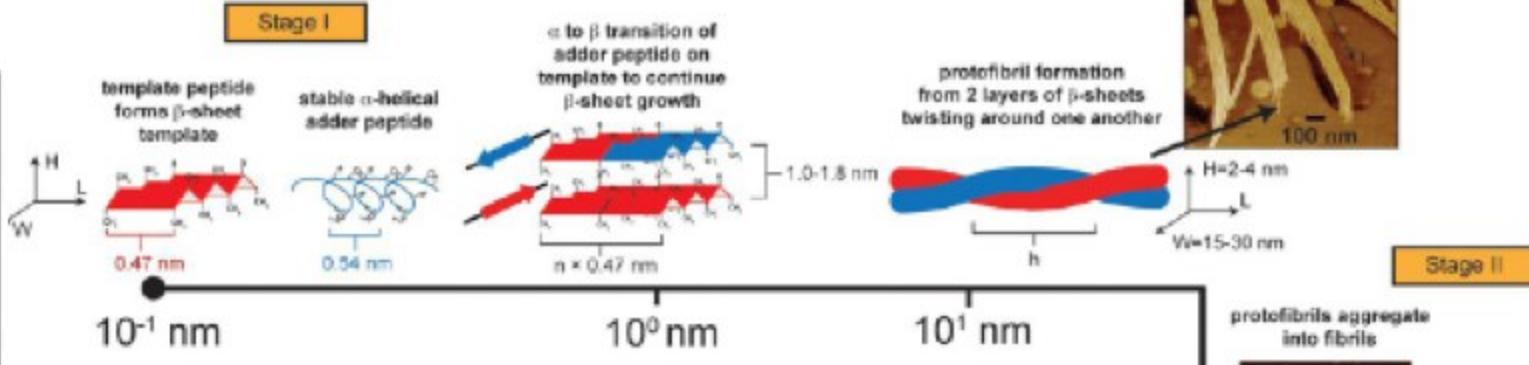
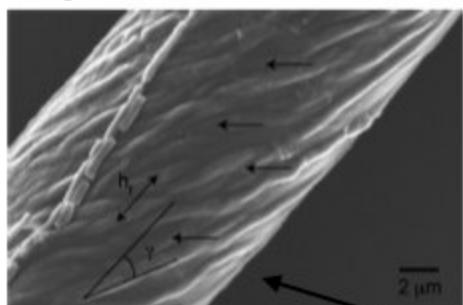
Evolution of the Amyloid Fiber over Multiple Length Scales

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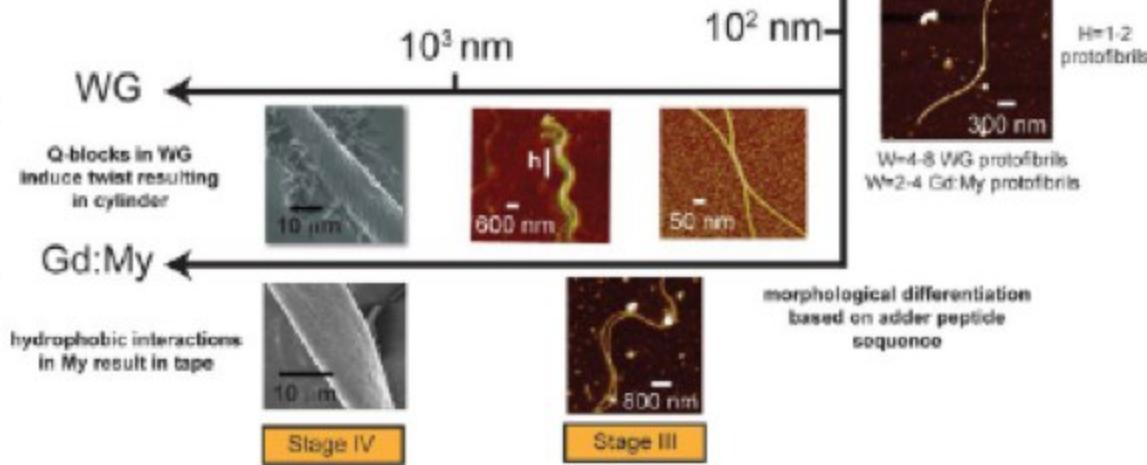
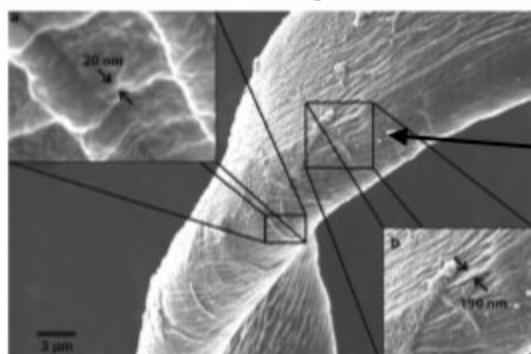
Devin M. Ridgley and Justin R. Barone*

Biological Systems Engineering Department, Virginia Tech, 303 Seitz Hall, Blacksburg, Virginia 24061, United States

cylindrical fiber

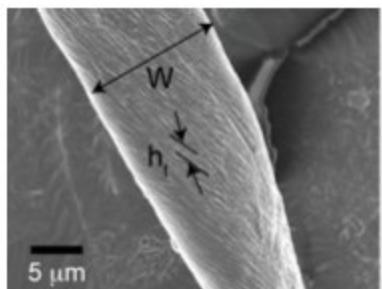


helical tape

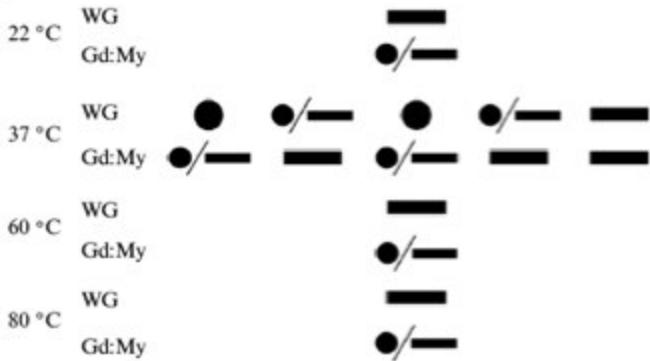
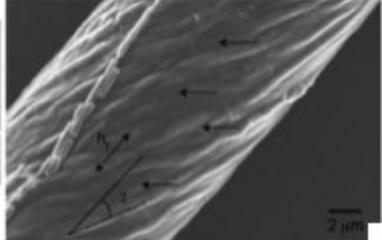
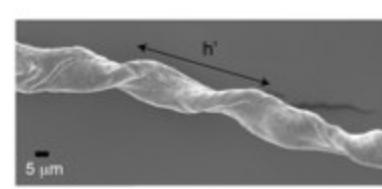
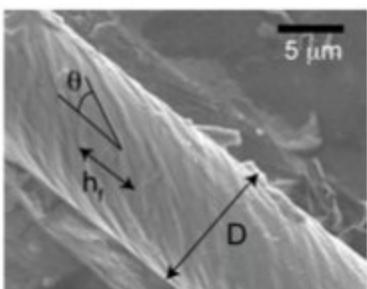


Twisted Amyloid Tapes: From Mesoscale Dimensions to “Molecular” Scale Parameters

helical tapes



cylindrical bundles



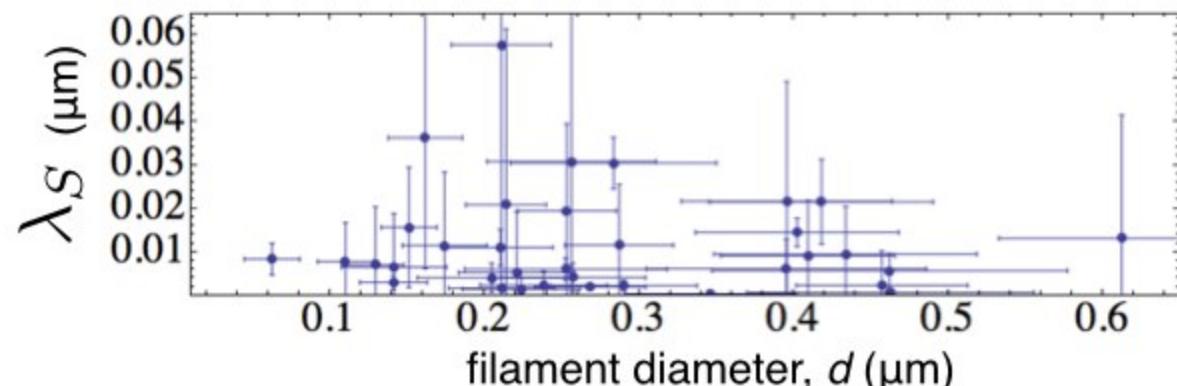
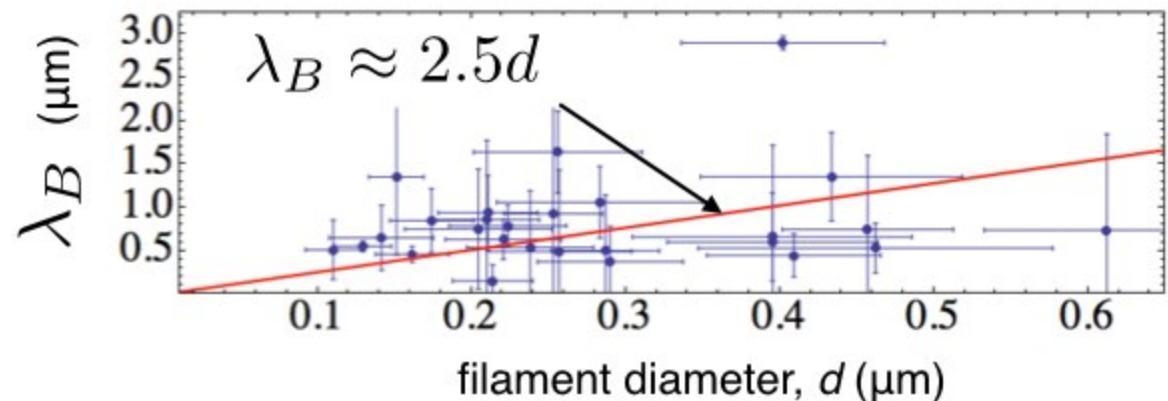
Ridgely & Barone, Soft Matter (2012).

predicted tape dimensions:

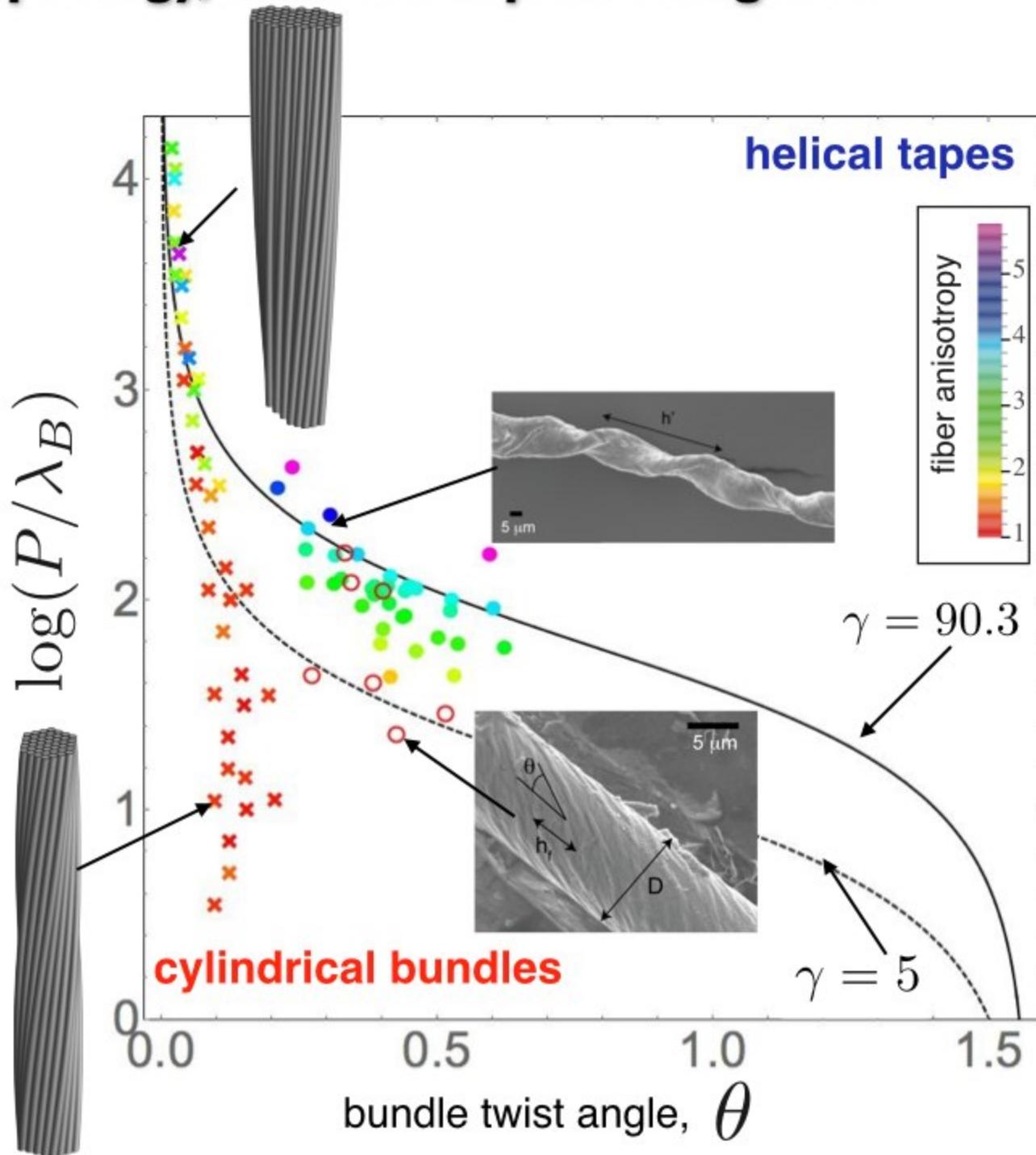
$$w_0 = 24^{1/3} \left(\frac{\Sigma}{\Omega^4 B \rho_0} \right)^{1/3} \quad t_0 = 80^{1/5} \left(\frac{\Sigma}{\Omega^4 Y} \right)^{1/5}$$

characteristic lengths (material dependent):

$$\lambda_B \equiv \sqrt{\frac{B \rho_0}{Y}} = \sqrt{\frac{3t_0^5}{10w_0^3}} \quad \lambda_S \equiv \Sigma/Y = \frac{\Omega^4 t_0^5}{80}$$



Fiber morphology, a universal phase diagram?



Hall, Bruss, Barone,
GMG, in preparation.

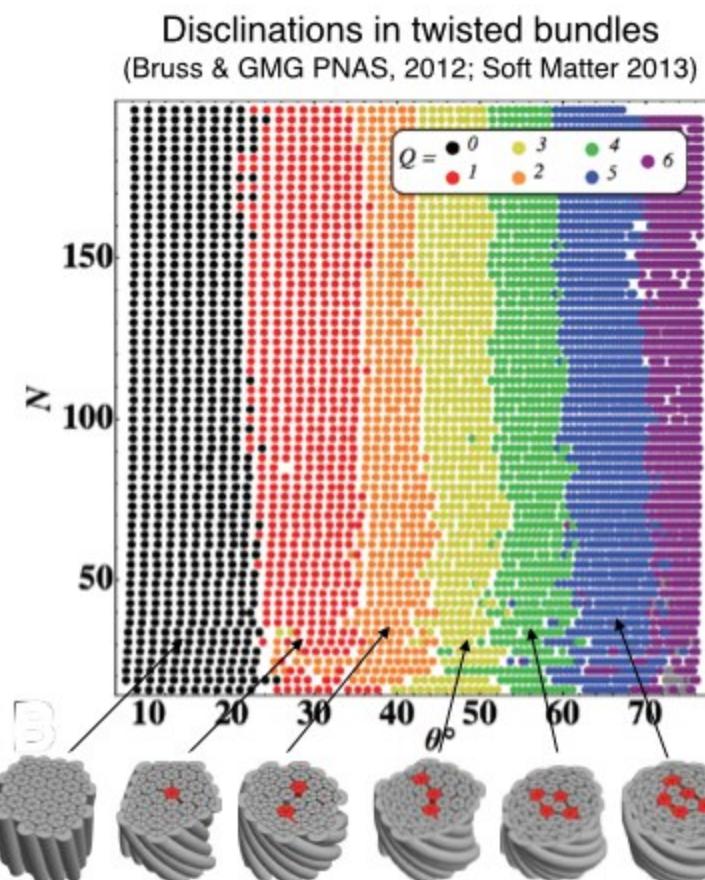
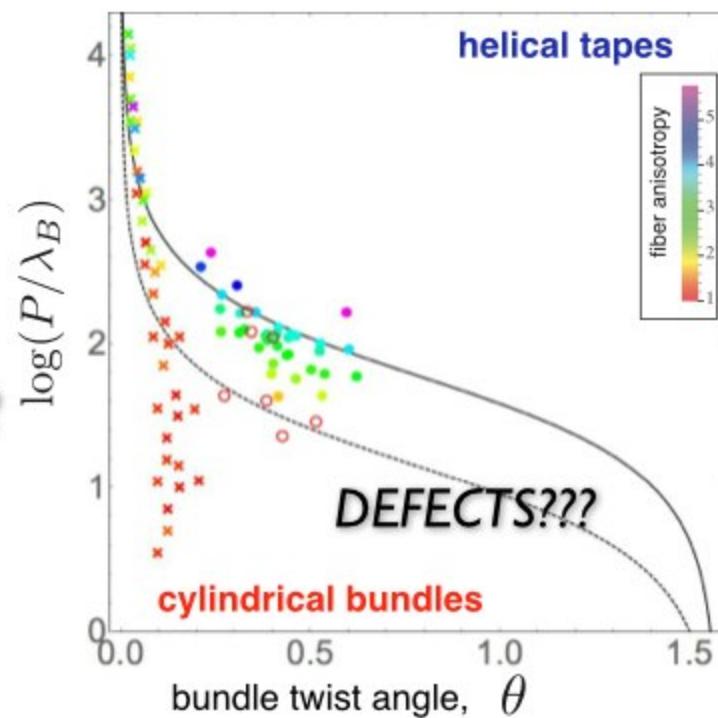
Summary (Frustration & Morphology Selection):

- 1) Geometric strains drive anisotropy in fiber sections above critical size
- 2) Tape width/thickness dimensions are selected by intra-filament (bending)/intra-filament (packing) elasticity
- 3) Mesoscale dimensions directly quantify microscopic parameters (inter-filament elasticity; intra-filament elasticity; inter-fil. cohesion)

Open questions:

Stiff filaments at large twist?

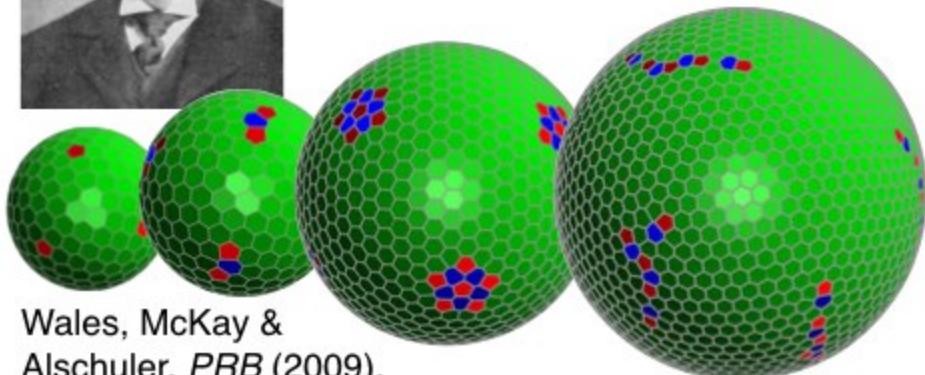
How do mitigate bundle/tape transition?



Geometry of Filamentous Matter:



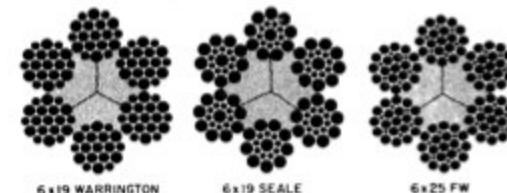
Optimal packing on spheres:
Thomson Problem (1904)



Wales, McKay &
Alschuler, PRB (2009).



Optimal packing in ropes:
“Roebling Problem” (~1849)



$\theta = 33^\circ; N = 34$



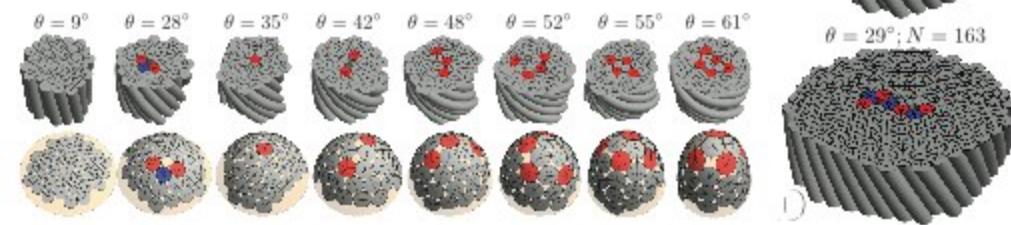
$\theta = 29^\circ; N = 46$



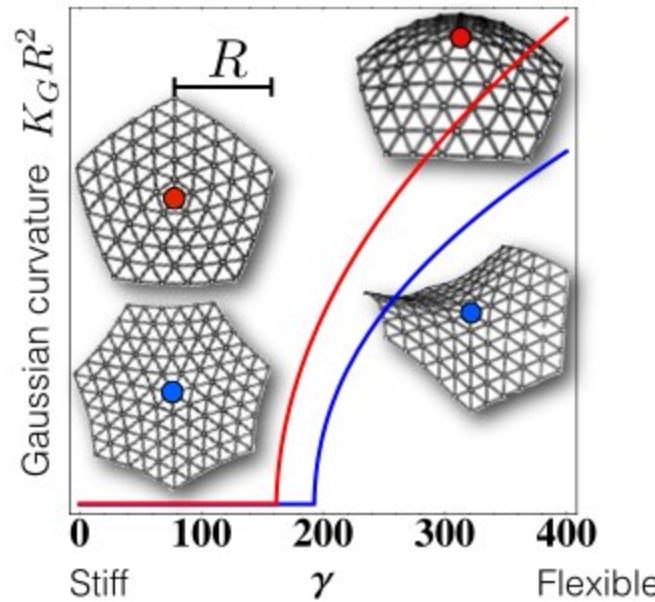
$\theta = 29^\circ; N = 94$



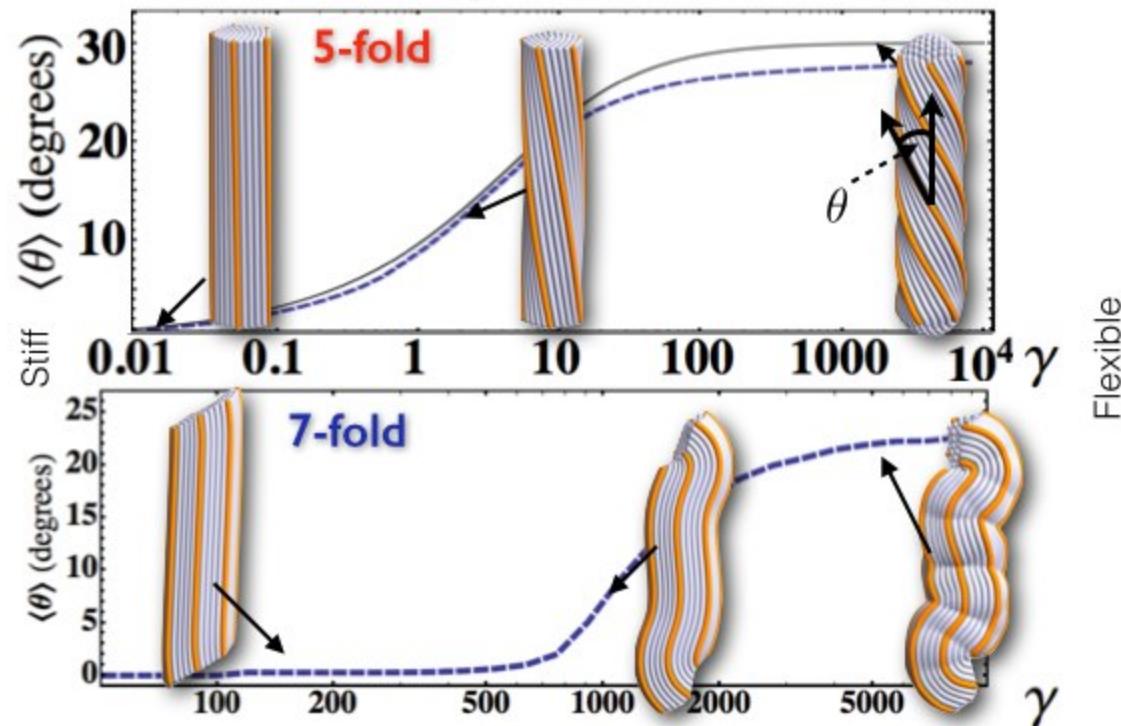
$\theta = 29^\circ; N = 163$



Defect-induced buckling of
2D x-tals: Seung & Nelson (1988)



Defect-induced buckling of bundles: Bruss *in preparation*



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