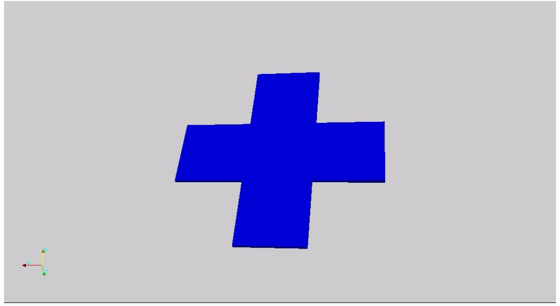
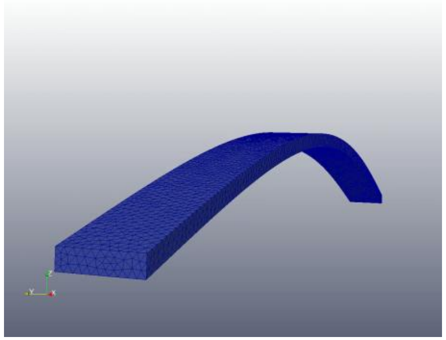
Modeling liquid crystal elastomers: from auto-origami to light-driven autonomous soft robotics





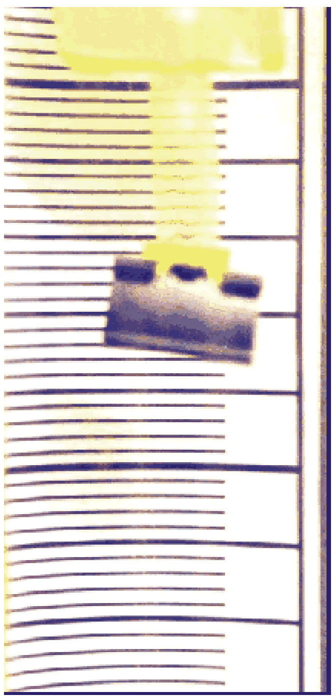
Robin L. B. Selinger

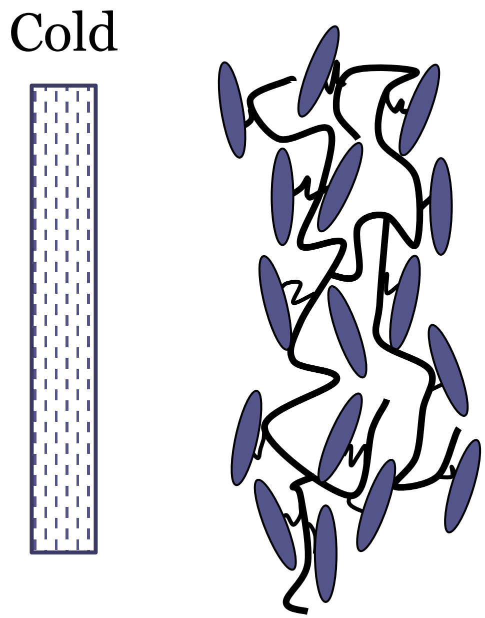
LIQUID CRYSTAL INSTITUTE

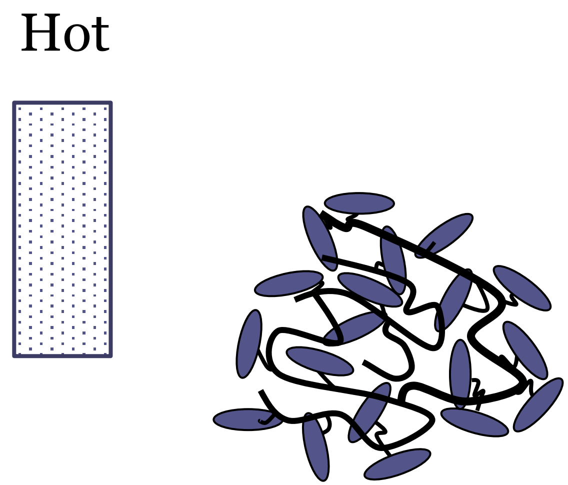
KENT STATE UNIVERSITY

NSF-DMR 1409658, and NSF CMMI-1436565, NSF-CMMI -1663041

Liquid crystal elastomers: programmable shape-morphing materials







Rod-like liquid crystal mesogens bonded to polymer
...on side-chains or in the main chain

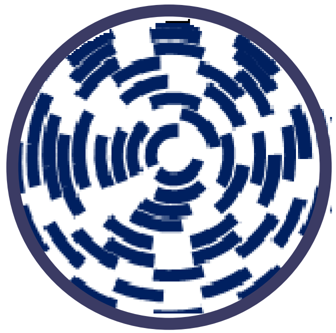
Local average molecular orientation of liquid crystal defines the **nematic director**

Heating reduces nematic order→ induces strain

UNIVERSITY OF CAMBRIDGE

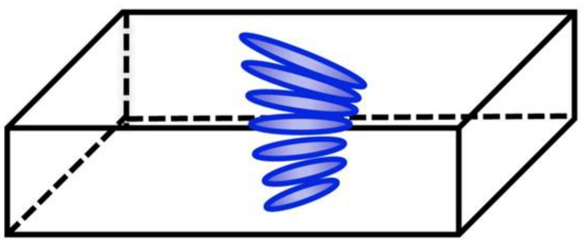
E. M. Terentjev

Non-uniform nematic director encodes complex shape change

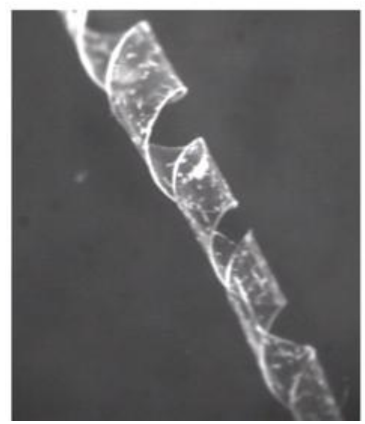


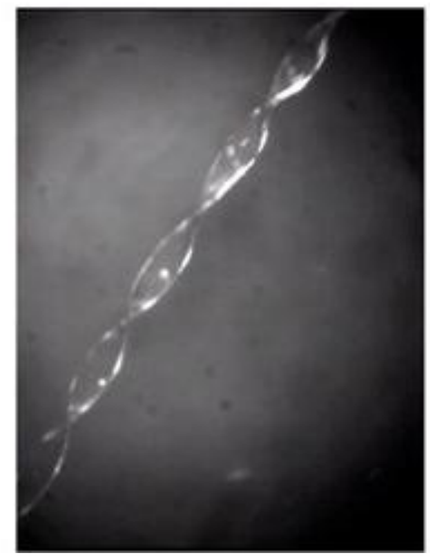
Disk → cone

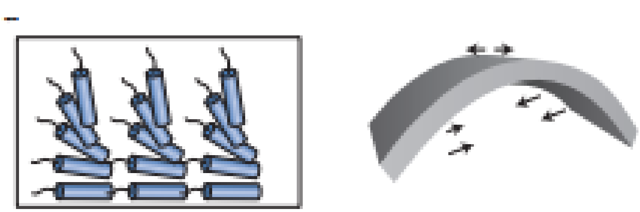
De Haan et al
Angewandte Chemie
2012



Twisted or splayed director
→ spirals, helicoids, curves

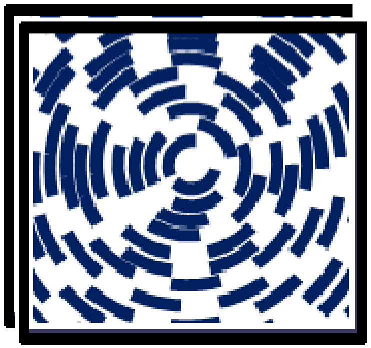






Y. Sawa, F. Ye, K. Urayama, T. Takigawa,
V. Gimenez-Pinto, RLB Selinger and JV Selinger
PNAS 2011

Ways to encode memory: "Blueprinting"



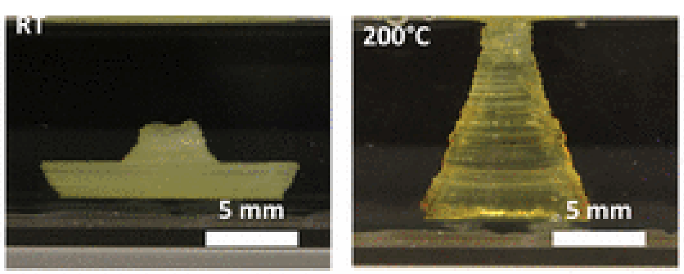
OLD: Form polymer between patterned substrates that impose surface anchoring

- Director uniform through thickness; or with twist or splay
- Shape transformations: 2-D to 3D

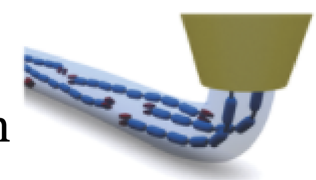
NEW: "4-D printing" recently developed by…

- Taylor Ware – ACS Applied Materials & Interfaces 2017
- Dirk Broer – Macromolecular Rapid Communications 2017
- TJ White and Jennifer Lewis—Advanced Materials 2018

Ware 2017



Director set by direction of extrusion

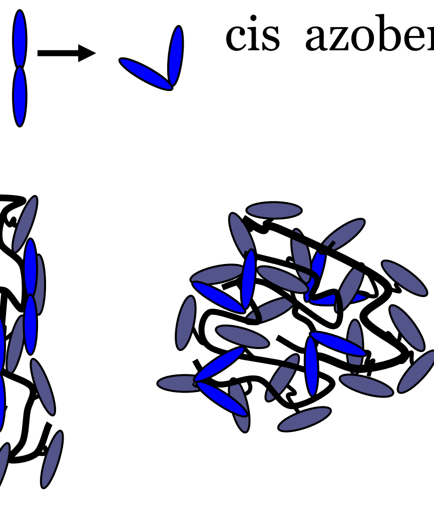


Shape transforms from one 3D shape to another

Inducing motion with light

Nathalie Katsonis
Univ of Twente

trans



cis azobenzene derivative

Change of nematic order produces light-driven mechanical strain

Trans→ Cis photoactuation

Cis→Trans relaxation

Each may be slow or fast, depending on which azo-compound is used, light wavelength and intensity

How to model shape change? Often too complex for analytical solution.

Finite element elastodynamics with a tetrahedral mesh

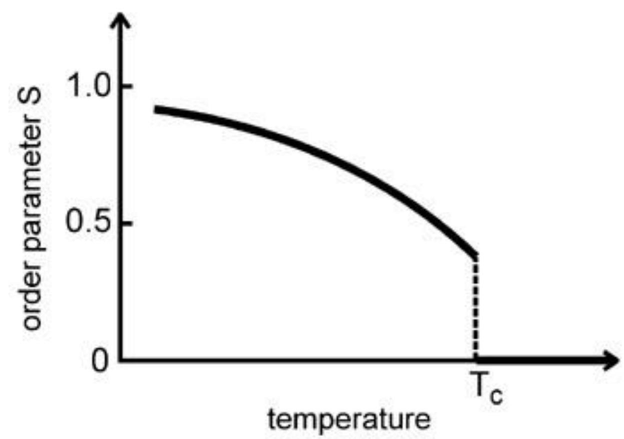
$$H = \frac{1}{2}\sum_t V^t C_{ijkl}\varepsilon^t_{ij}\varepsilon^t_{kl} - \alpha\sum_t V^t\varepsilon^t_{ij}(S^t - S^0)\frac{1}{2}\left(3n^t_i n^t_j - \delta_{ij}\right) + \frac{1}{2}\sum_p m_p v_p^2$$

Elastic potential energy
(function of Green-Lagrange strain)
Summed over elements

Coupling between
strain and nematic
order
Summed over
elements

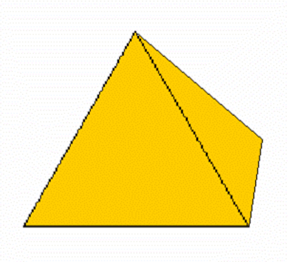
Kinetic energy
Summed over
nodes

Change in *scalar order parameter S(T)* describes loss of orientational order

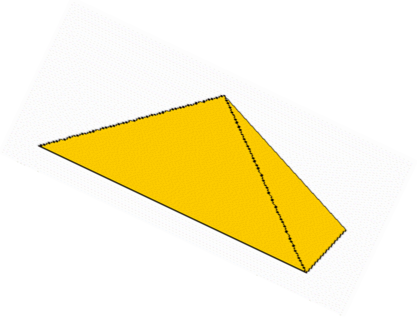


Each tetrahedral element has a REFERENCE state (positions of nodes if free of external stress→ fully relaxed)

and a CURRENT state (defined by current node positions)



Reference state:
Relaxed, in mechanical equilibrium



Current state
Stretched, sheared, translated, rotated…

<u>This approach can be adapted</u> to model materials that undergo "training" via evolution of the reference state as a function of mechanical stress → e.g. via plastic deformation

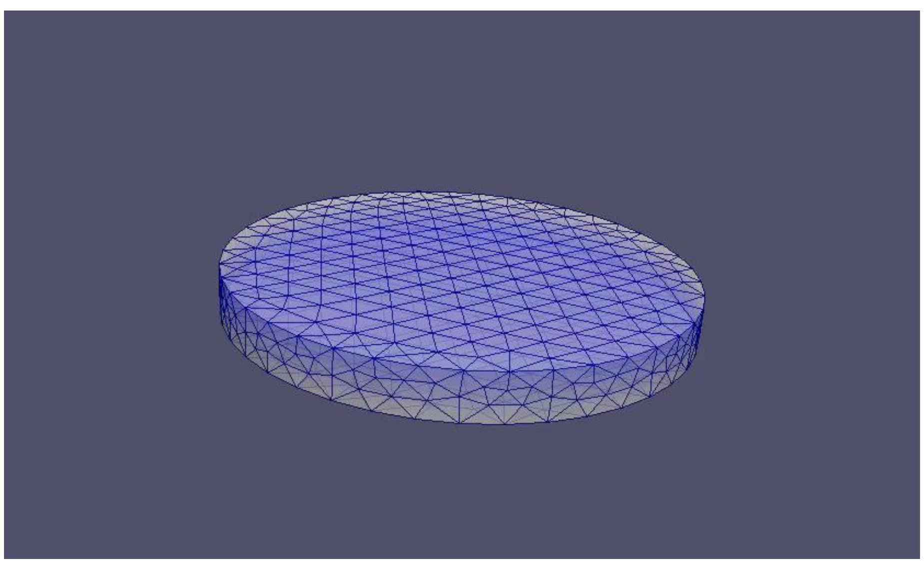
Finite Element Elastodynamics Simulation (FEM)

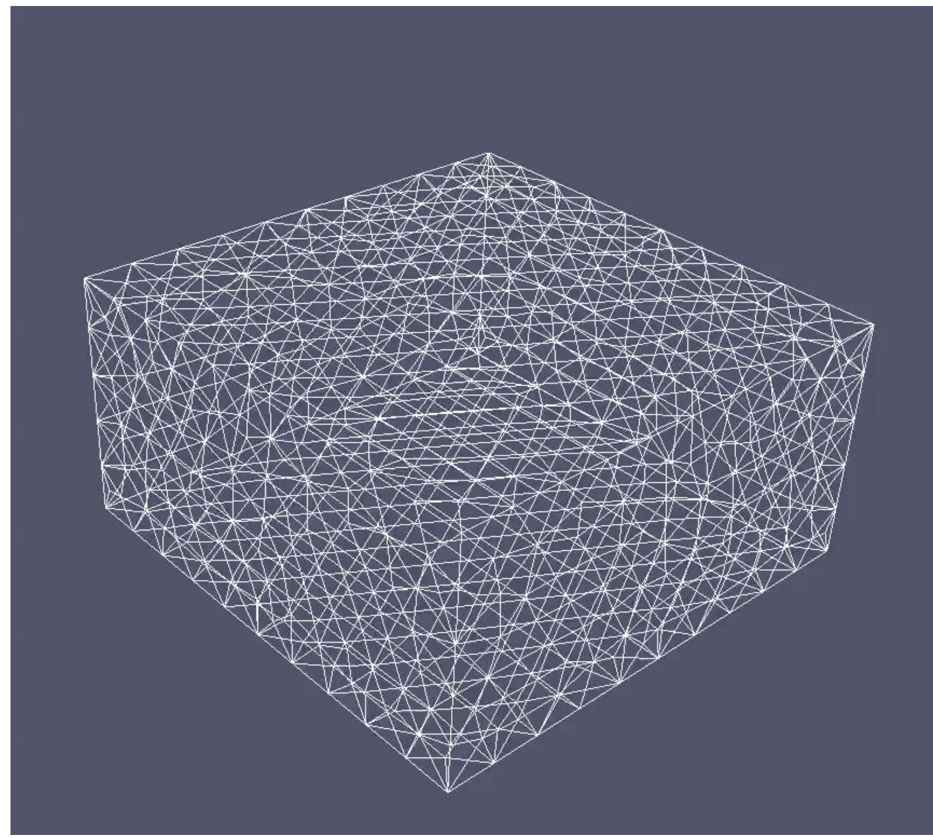
Elastodynamics: Mesh nodes move via $f = ma$

$$m\ddot{x}_n = -\frac{\partial F_n}{\partial r_n}$$ → **Force on each node**

Home-made code implemented in CUDA for GPU-enabled computer

Fast nonlinear
3-d elastodynamics

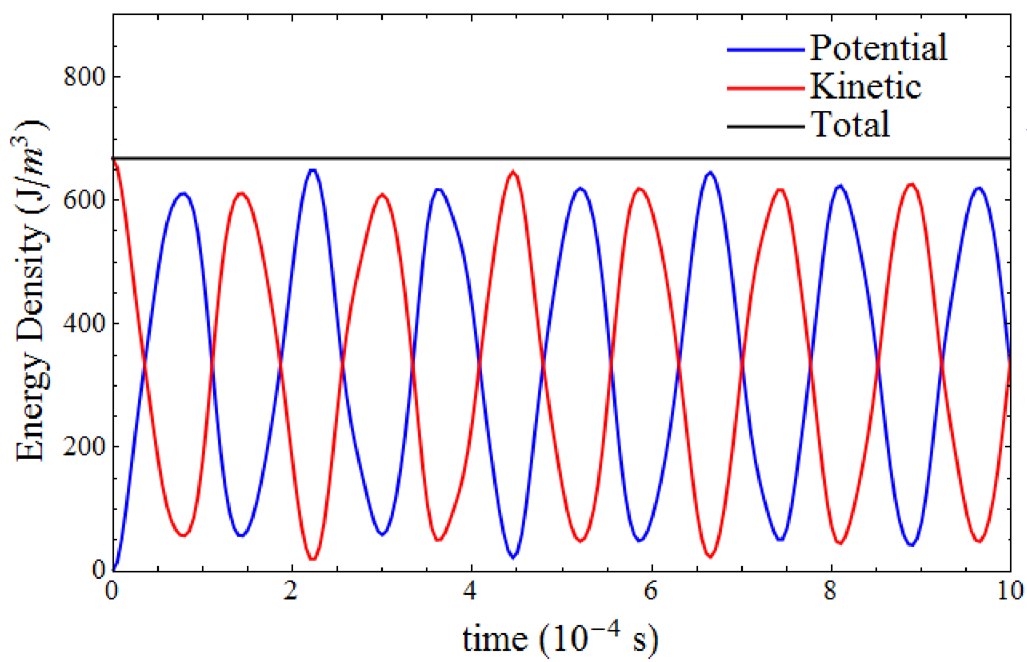




Code runs at 700 frames/second

Without dissipative forces added, kinetic+potential energy is well conserved

Must add friction/dissipation to relax to mechanical equilibrium



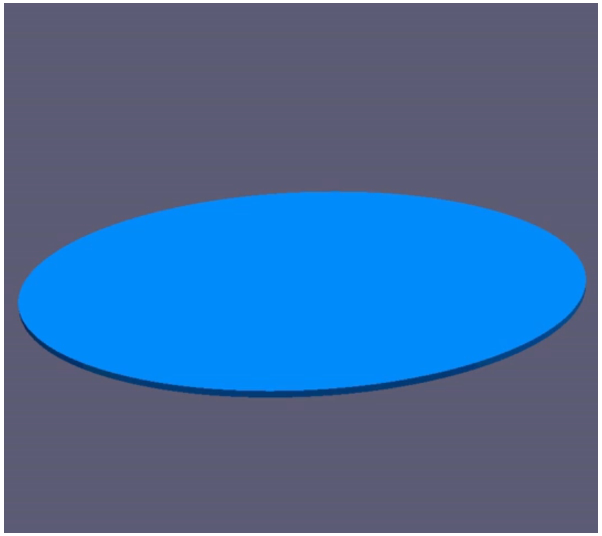
Sum of kinetic+potential energy is constant

Typical size for simulations:
50K nodes,
300K elements

Nematic director is "programmed" when the polymer is cross-linked, encodes not just initial and final states, but a whole choreography of motion. Some simple cases…

frontiers in Materials

A Konya, V Gimenez-Pinto, RLBS, 2016



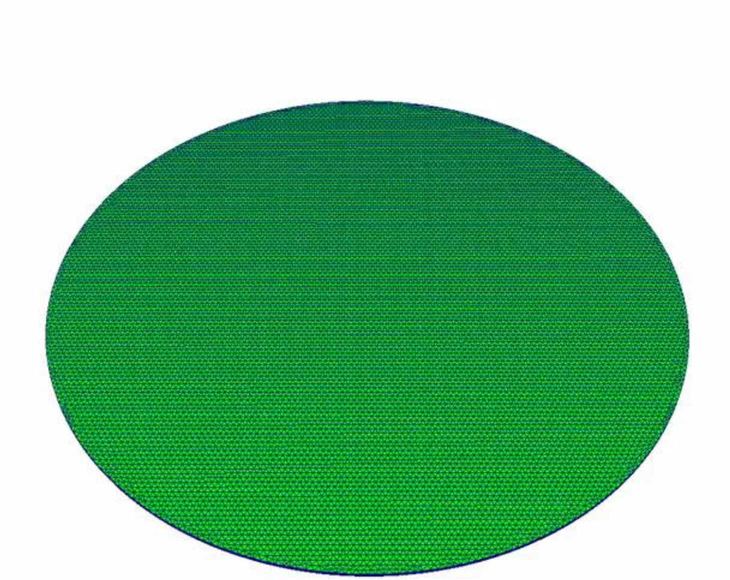
Degenerate final state…
disk can pop up or down

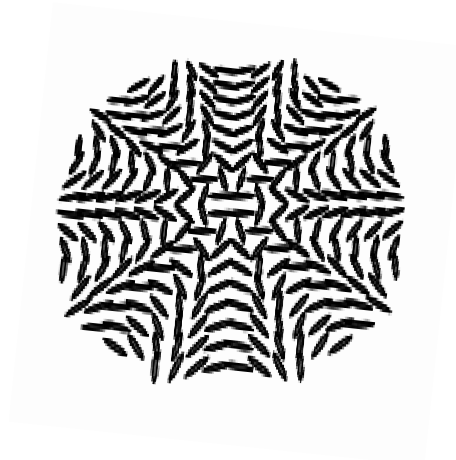
frontiers
in Materials

A Konya, V Gimenez-Pinto, RLBS, 2016

Pop-through → hysteresis

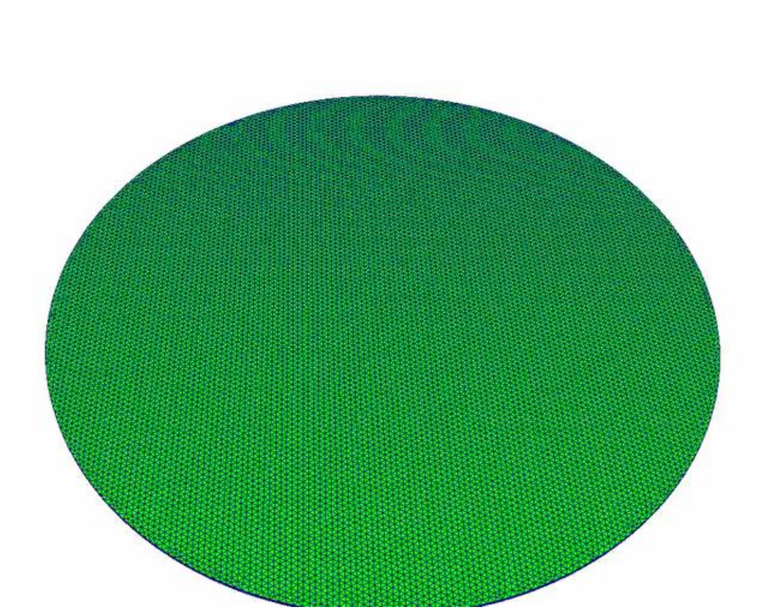
Degenerate final states can include both stable and metastable configurations





-4 defect

Heat uniformly…
Up-down symmetry→
asymmetic, metastable state



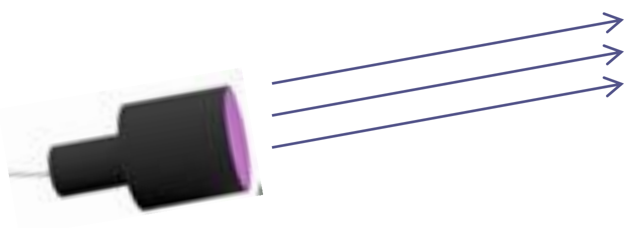
Heat from one side→
breaks up/down symmetry

Reaches symmetric ground state

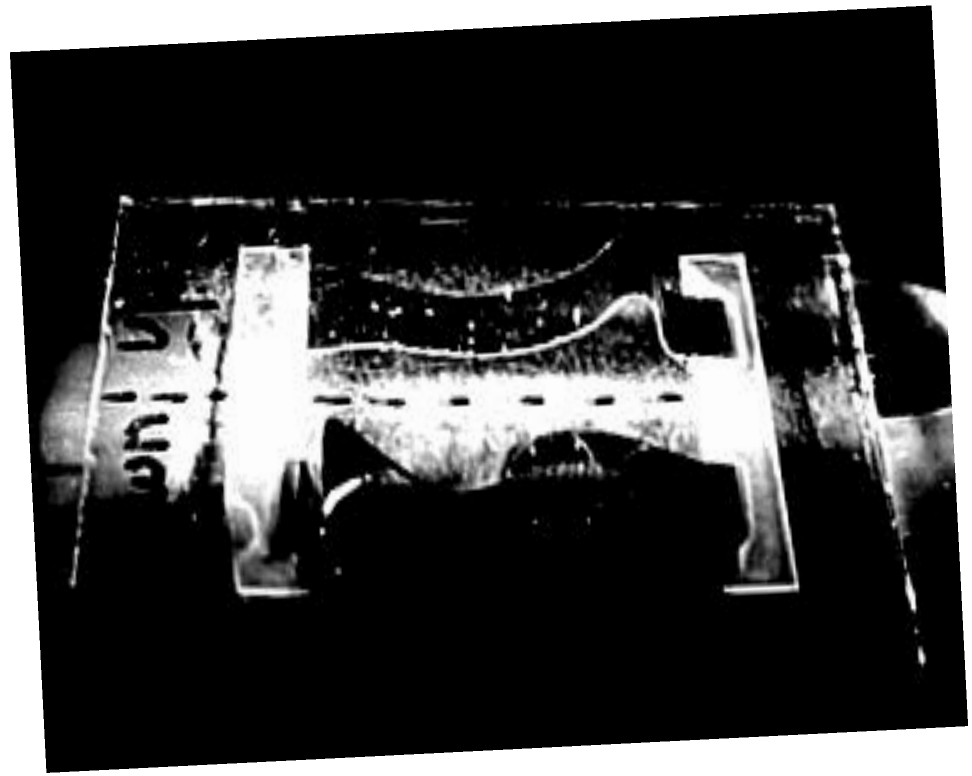
frontiers in Materials

A Konya, V Gimenez-Pinto, RLBS, 2016

Nematic director can be programmed to enable photoactuated wave motion



UV light from left side
at oblique angle



Experiment by Anne Helene Gelebart, Dirk Broer, and coworkers
Thin ribbon with splay director: planar on one side, homeotropic on the other

Anne Hélène
Gélébart

Dirk Broer

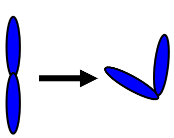
Blueprinted nematic polymer film with azobenzene derivative

Director: Planar on top

Homeotropic on bottom

photoactuation

trans

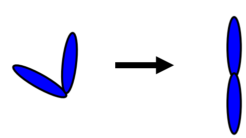


cis

under UV light

thermal relaxation

cis



trans

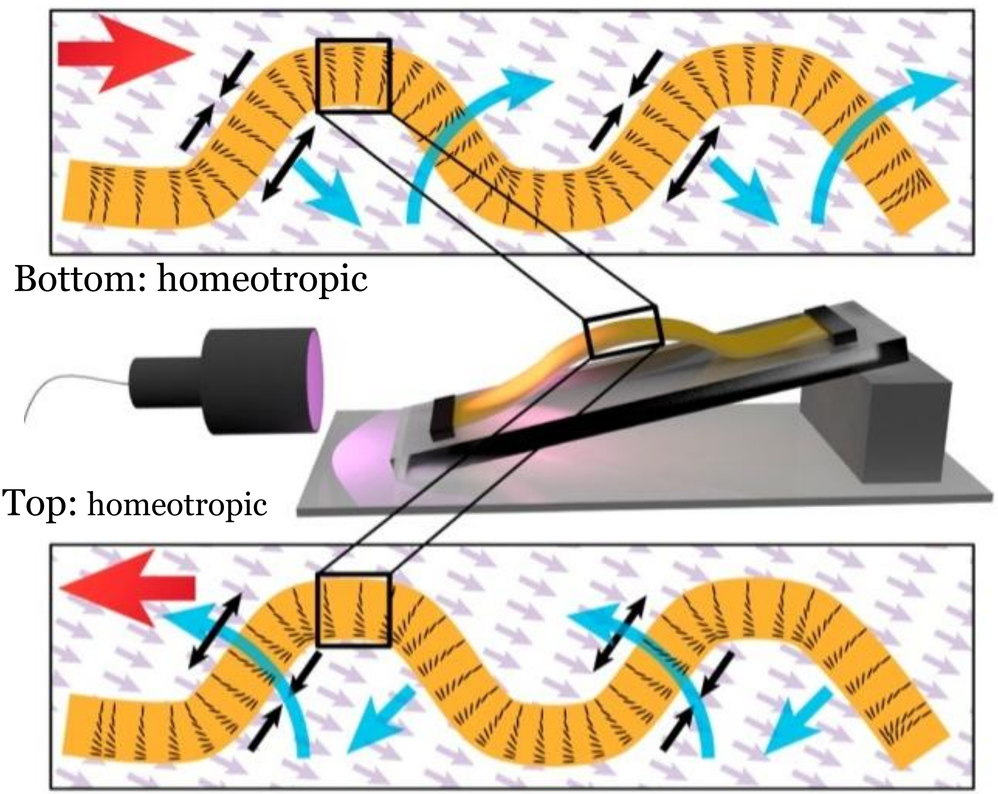
when light is removed

Both transitions take
less than 1 sec

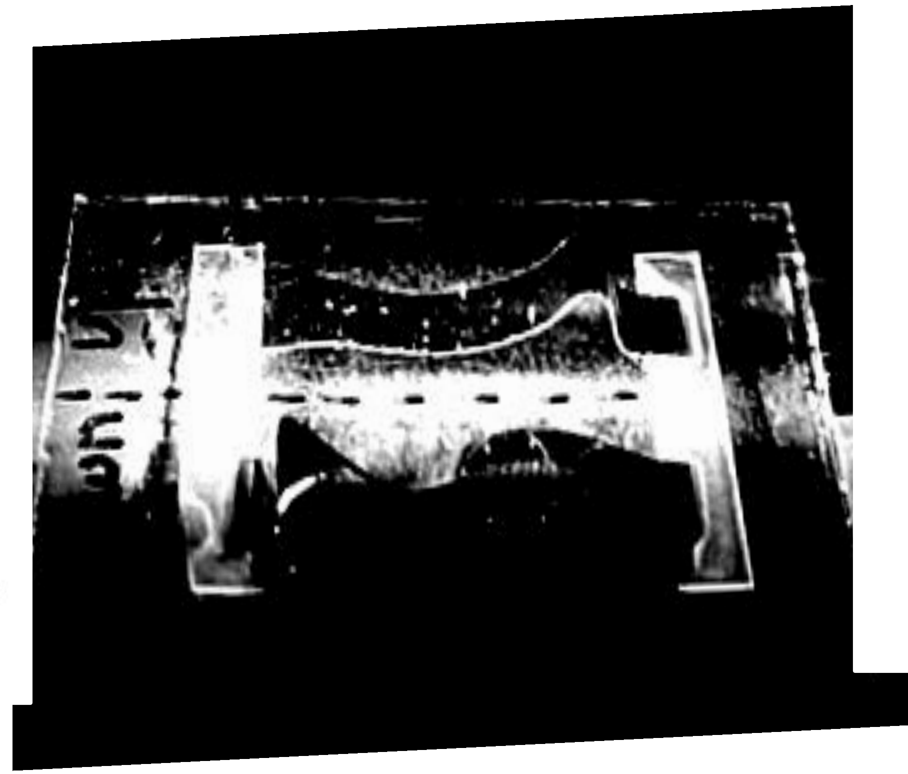
Anne Hélène
Gélébart

Dirk Broer

Top: planar ... waves move away from the light source



Bottom: planar... waves move toward the light source



UV light from left side

Questions:

What produces continuous photoactuated oscillation?

Why does the direction of wave motion change with orientation?

Clues:

→ Oscillation occurs ONLY with splay director geometry
… Not uniform planar, not uniform homeotropic

→ Oscillation only occurs with oblique light angle (e.g. 10° to the horizontal)
… self-shadowing plays an important role

Dynamics matter, motion is not overdamped…

Changes in S are not uniform in the sample…depend on local light exposure and vary with time as the material self-shadows

Assumptions in our model:

S = Nematic scalar order parameter in each tetrahedral volume element

S varies between upper and lower limits according to light exposure

Upper limit= highest degree of nematic order, all azo-dye in trans state

Lower limit = lowest degree of nematic order, maximum of azo-dye in cis state

Perform **ray tracing** to figure out which surface volume elements are illuminated

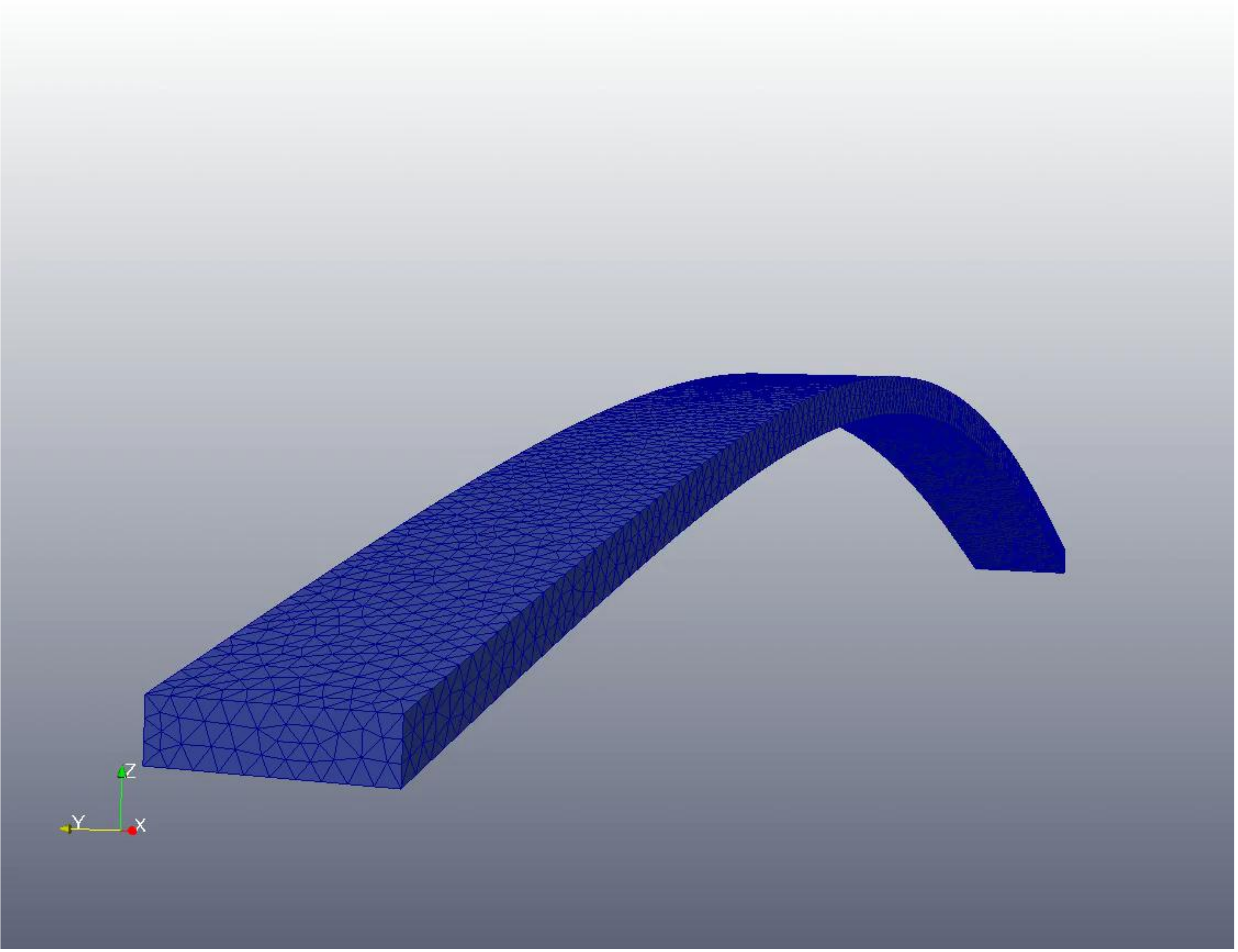
Photoactuation:

S drops linearly in time when material illuminated, reaches equilibrium in ~ 1 sec

Thermal Relaxation:

S *i*ncreases linearly in time when material shadowed, reaches equilibrium in ~1 sec

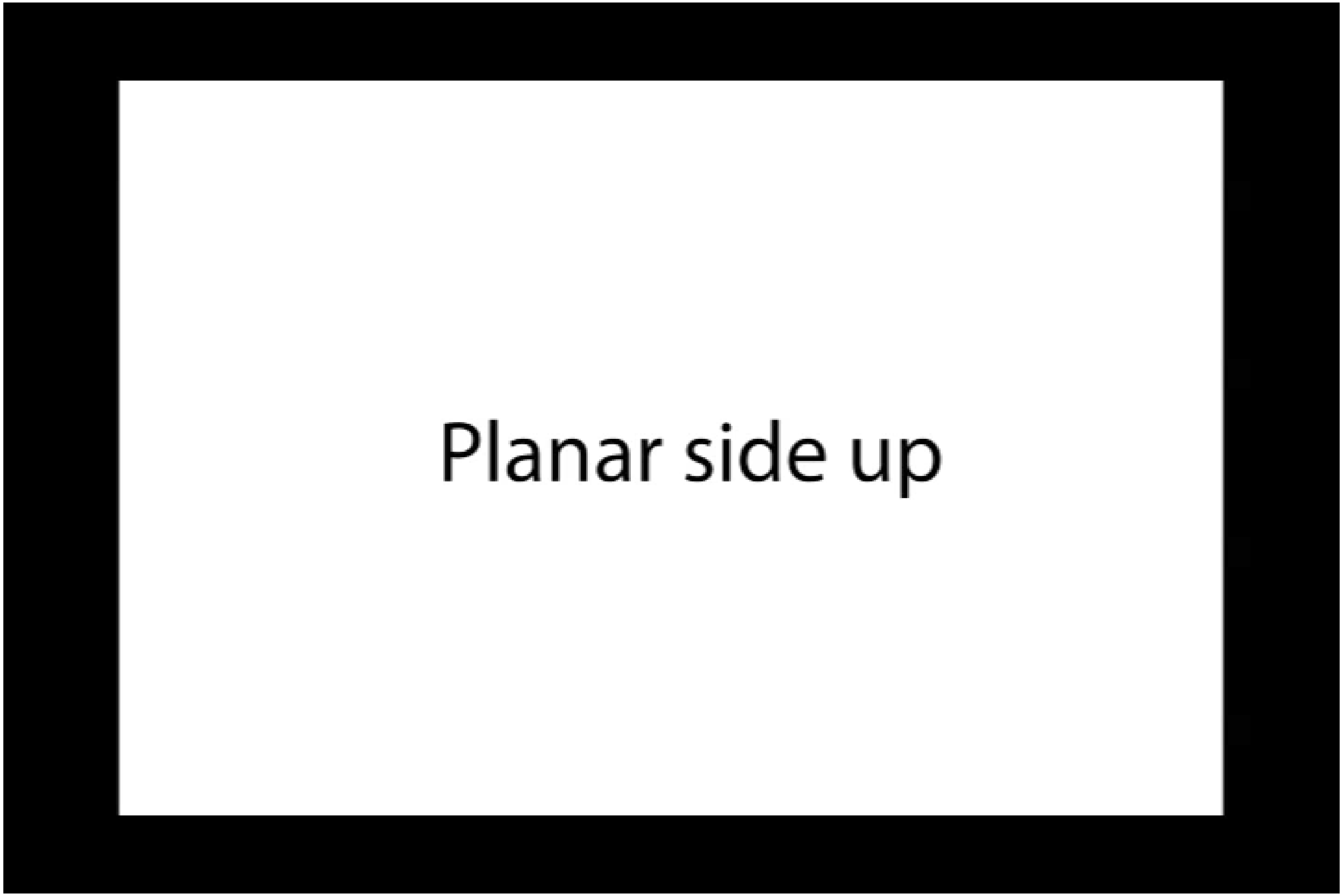
Rather simplified model ...



Michael Varga
Kent State University

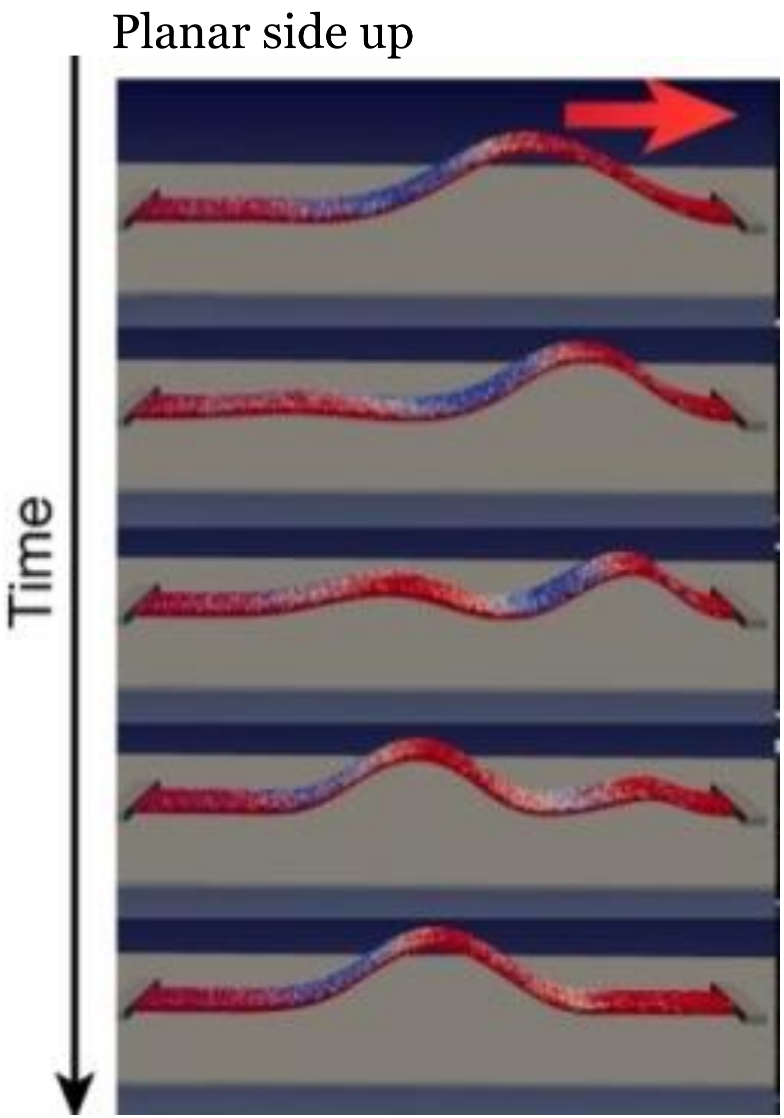
Planar side up: when illuminated, top shrinks along the long axis and expands weakly along the other two axes… light-exposed area curves downwards.

Comparison: Planar side up model and experiment



Time step: $2 \times 10{-}5$ s, 2×10^{6} t ime steps → 40 seconds of motion
Execution: 25 min on a single CPU with GPU acceleration
Tetrahedral mesh: 5,717 nodes and 19,478 volume elements

Comparison: Homeotropic side up model and experiment



Pop-through instability drives wave regeneration

Light shines from the left and strikes the film. Local order parameter decreases in area shown in blue.

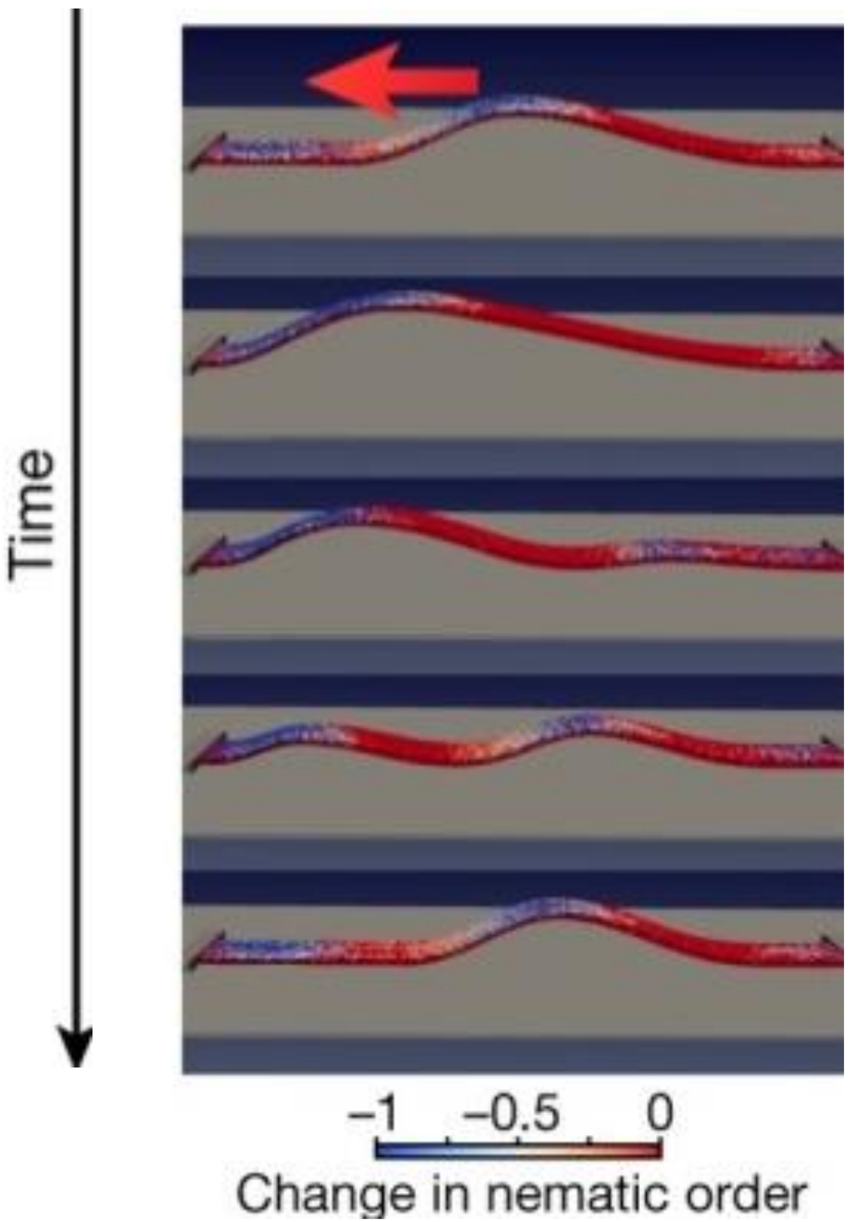
There, the top surface of the film contracts along its long axis and the film curves downwards, pushing crest toward the right.

Crest reaches end of the sample; its motion halts

New crest begins to form near the other end, grows to a critical size

Pop-through transition: elastic energy in previous crest is released into the new crest. Process repeats…

Homeotropic side up



Illuminated area in blue has reduced nematic order; homeotropic surface shrinks along surface normal, inducing upward curvature.

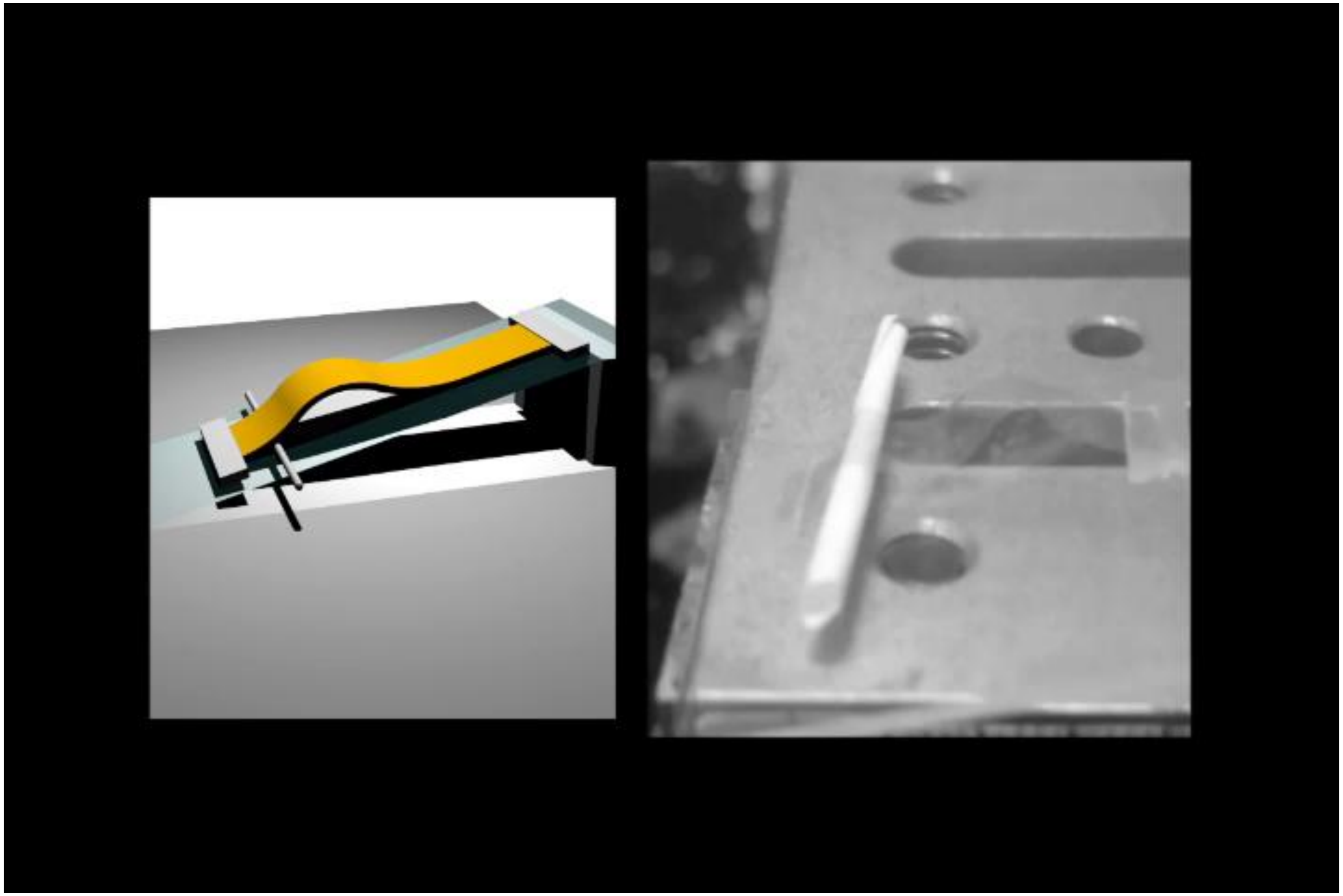
Resulting stress pulls the crest of the bump towards the left, towards the light source

It arrests when it reaches the clamped end and motion stops.

Shadowed region relaxes towards equilibrium; crest shrinks until light passes over it, nucleating a new crest to the right.

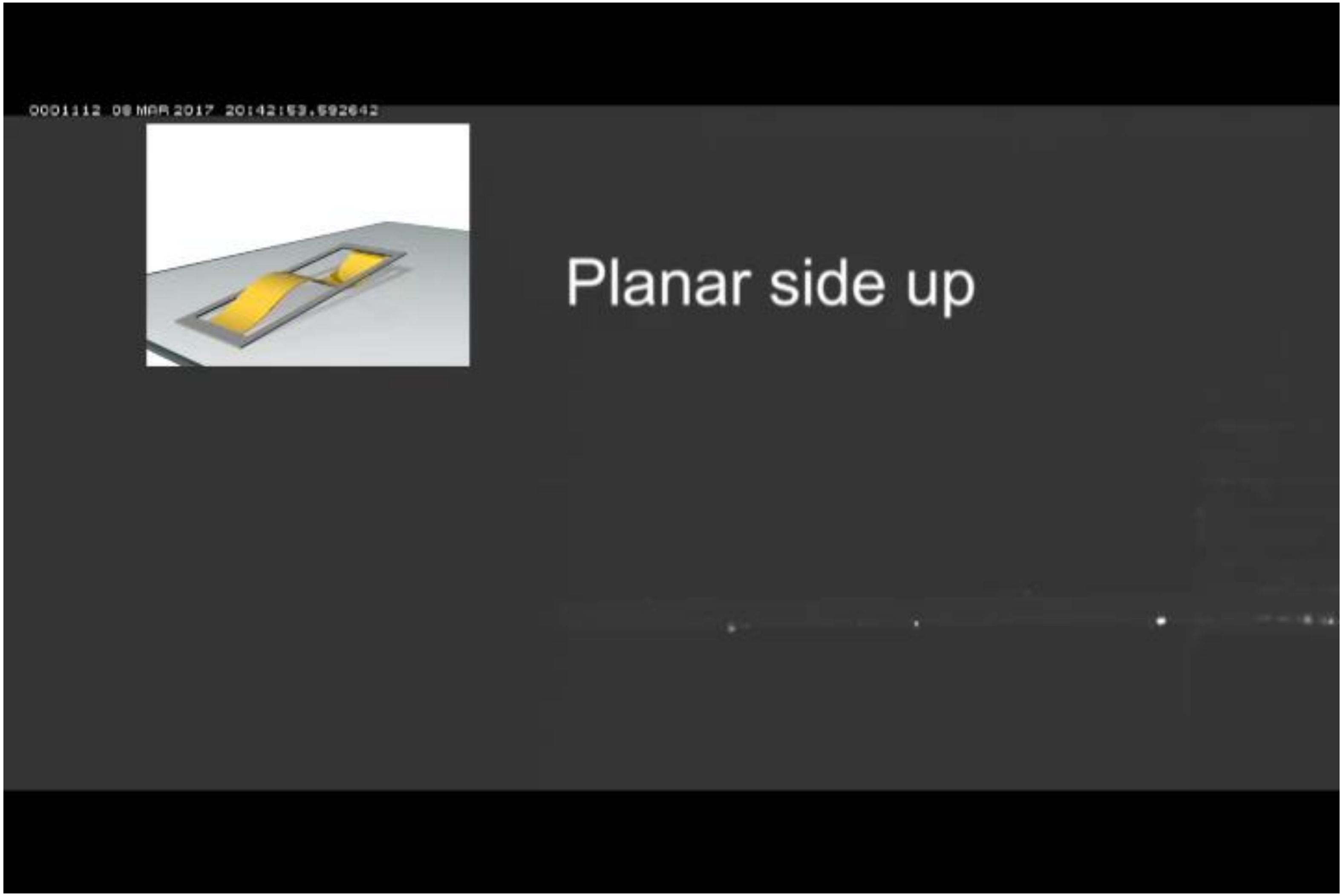
Time delay while the new crest forms, then a pop-through transition

What can this device potentially accomplish?



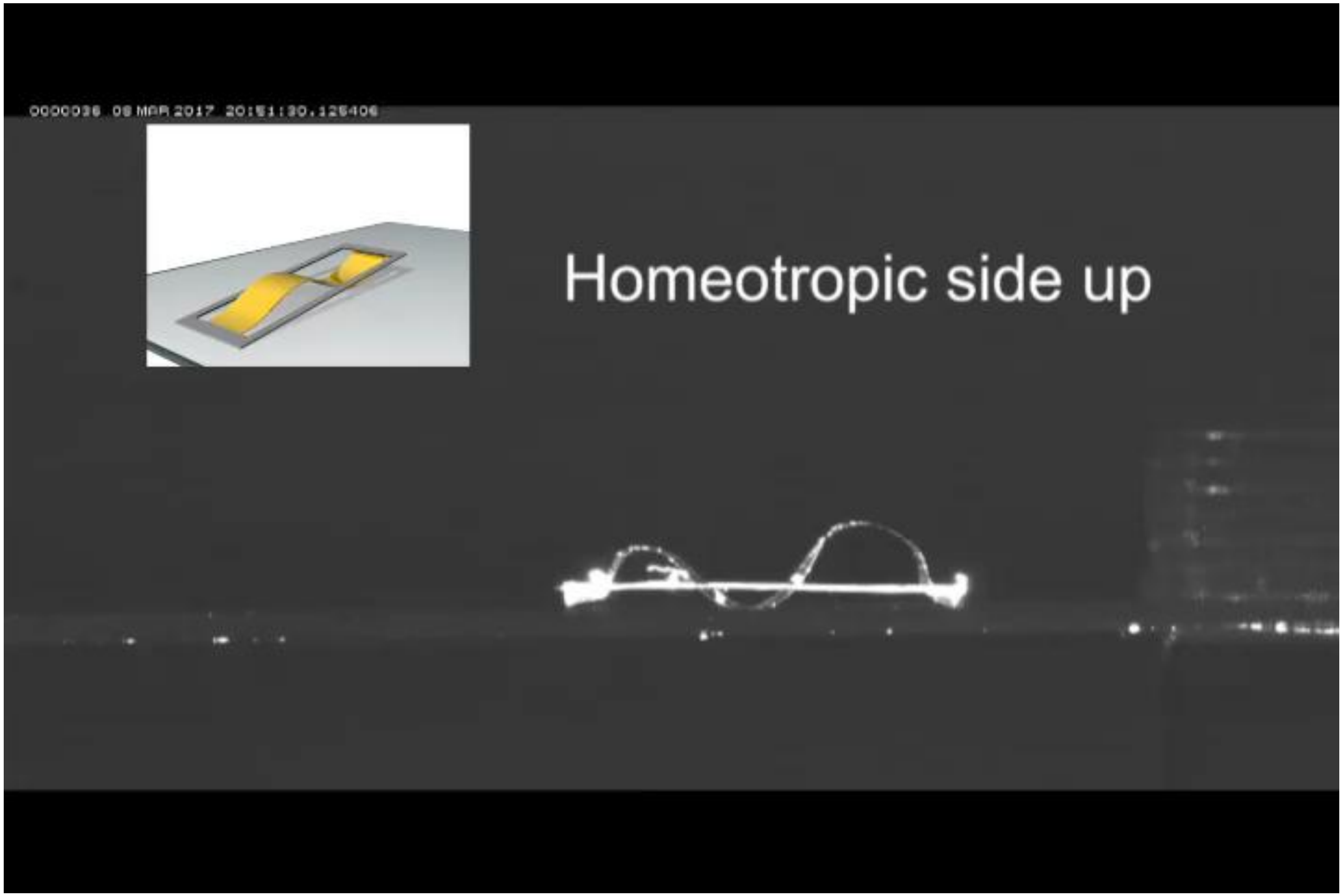
Photoactuated autonomous locomotion:

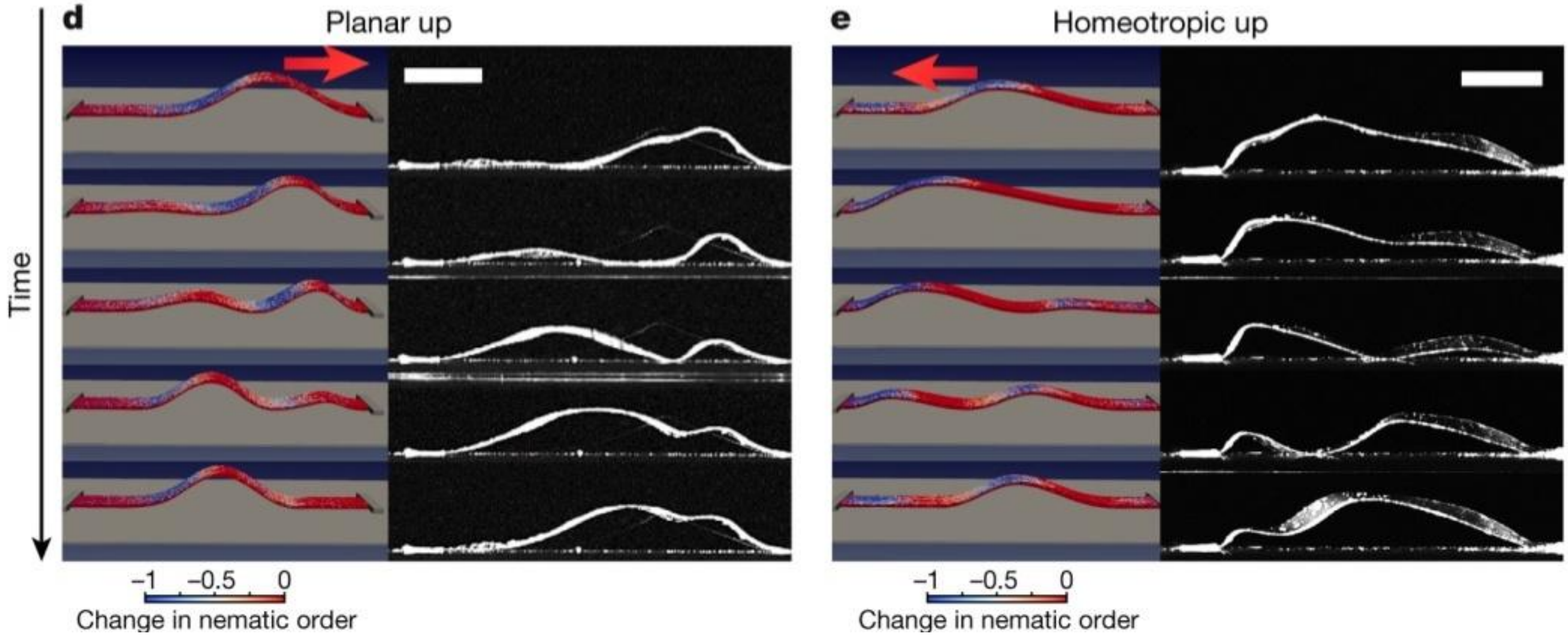
Planar side up ... Moves away from light source



Photoactuated autonomous locomotion:

Homeotropic side up ... Moves toward light source





Anne Helene Gelebart, Dirk Jan Mulder, Michael Varga, Andrew Konya, Ghislaine Vantomme, E. W. Meijer, Robin L. B. Selinger, and Dirk J. Broer, "Making waves in a photoactive polymer film," ***Nature*** v. 546, p. 632, 29 JUNE 2017

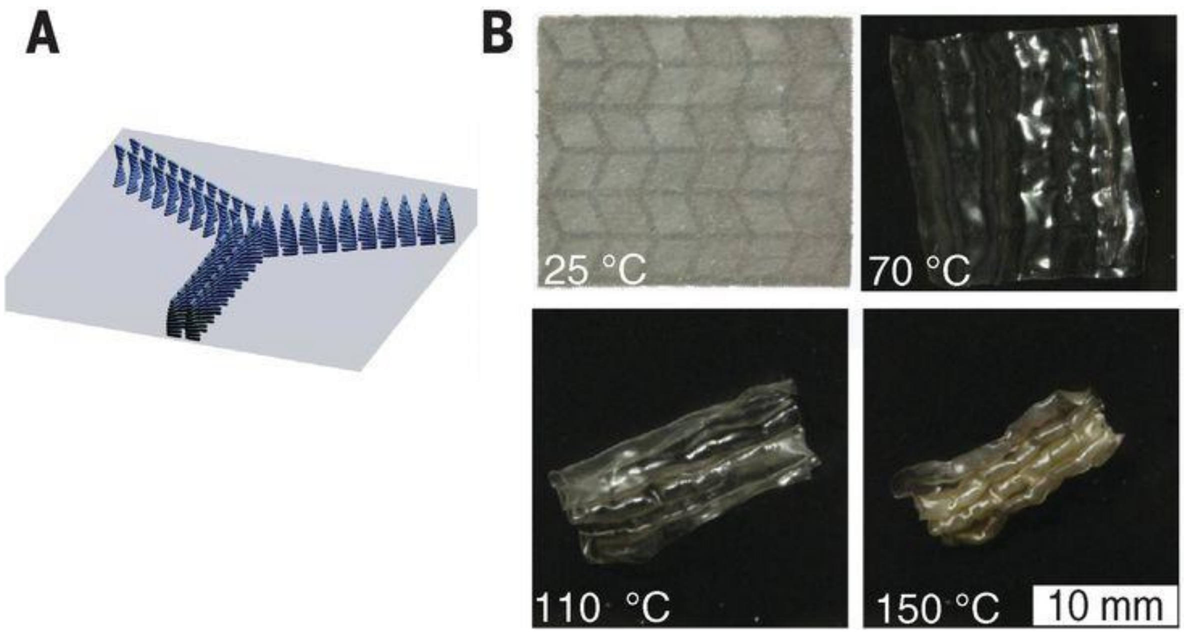
Link to paper and videos at **tiny.cc/rselinger2018**

nature

Shape change includes origami-like folding, slits and irises that open/close

Taylor Ware et al Science 2015

Origami-inspired actuators.(A) Schematic of an edge portion of the Miura-ori pattern with a localized twisted nematic region bounded by unordered regions.

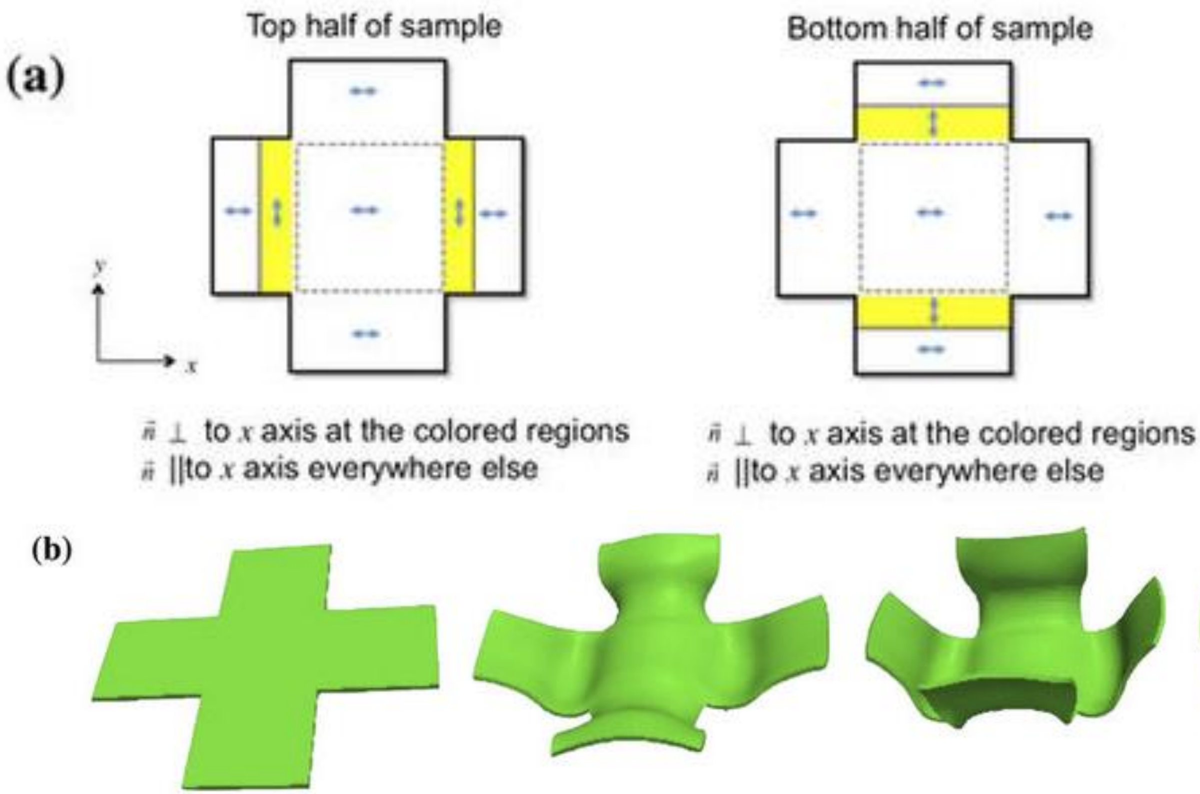


Taylor H. Ware et al. Science 2015;347:982-984

Science AAAS

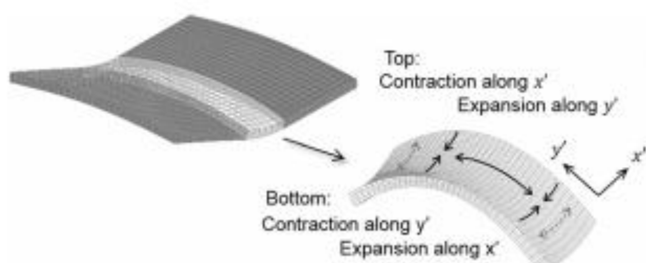
Hinges are narrow regions with director twist…

Can narrow twisted domains create sharp folds? Not very well!



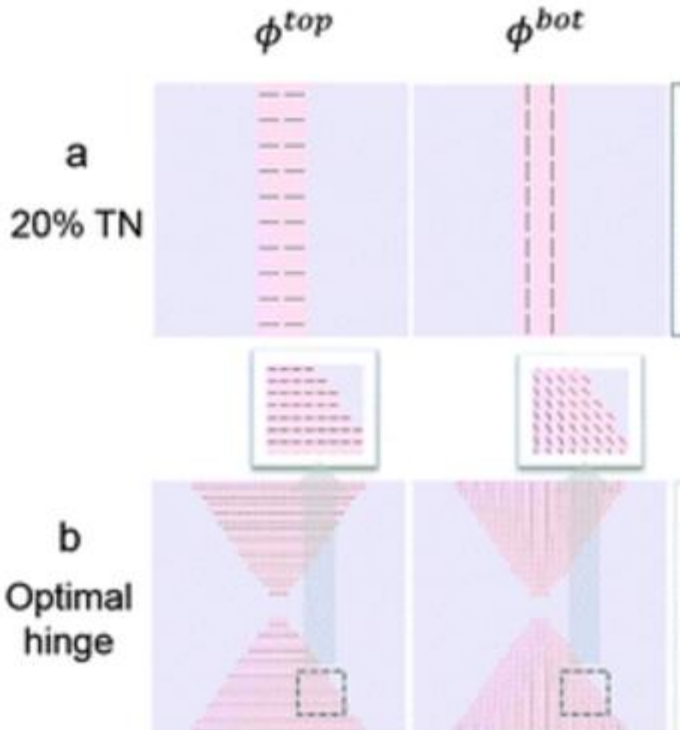
V Gimenez-Pinto, F. Ye, B. Mbanga, JV Selinger, RLB Selinger Scientific Reports 2017

‘anticlastic’ bend

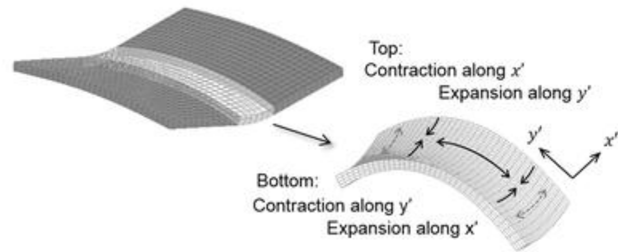


K. Fuchi et al Soft Matter 2015

How to design a sharp hinge for origami? Fuchi et al propose a structure



Twisted nematic hinge creates saddle curvature



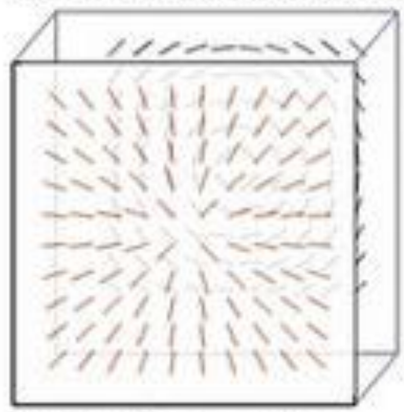
Topology optimization for the design of folding liquid crystal elastomer actuators†

Kazuko Fuchi,‡[ab] Taylor H. Ware,§‡[cd] Philip R. Buskohl,[c] Gregory W. Reich,[a] Richard A. Vaia,[c] Timothy J. White[c] and James J. Joo*[a]

Soft Matter 2015

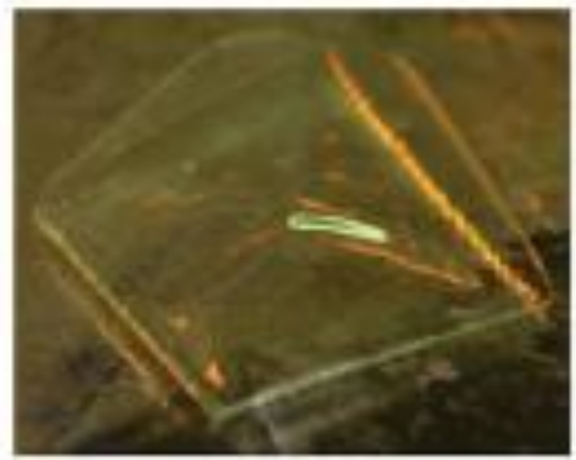
PROBLEM: This optimized solution isn't workable if spacing between folds is small compared to their length

Twisted director –square sample with a +1 defect



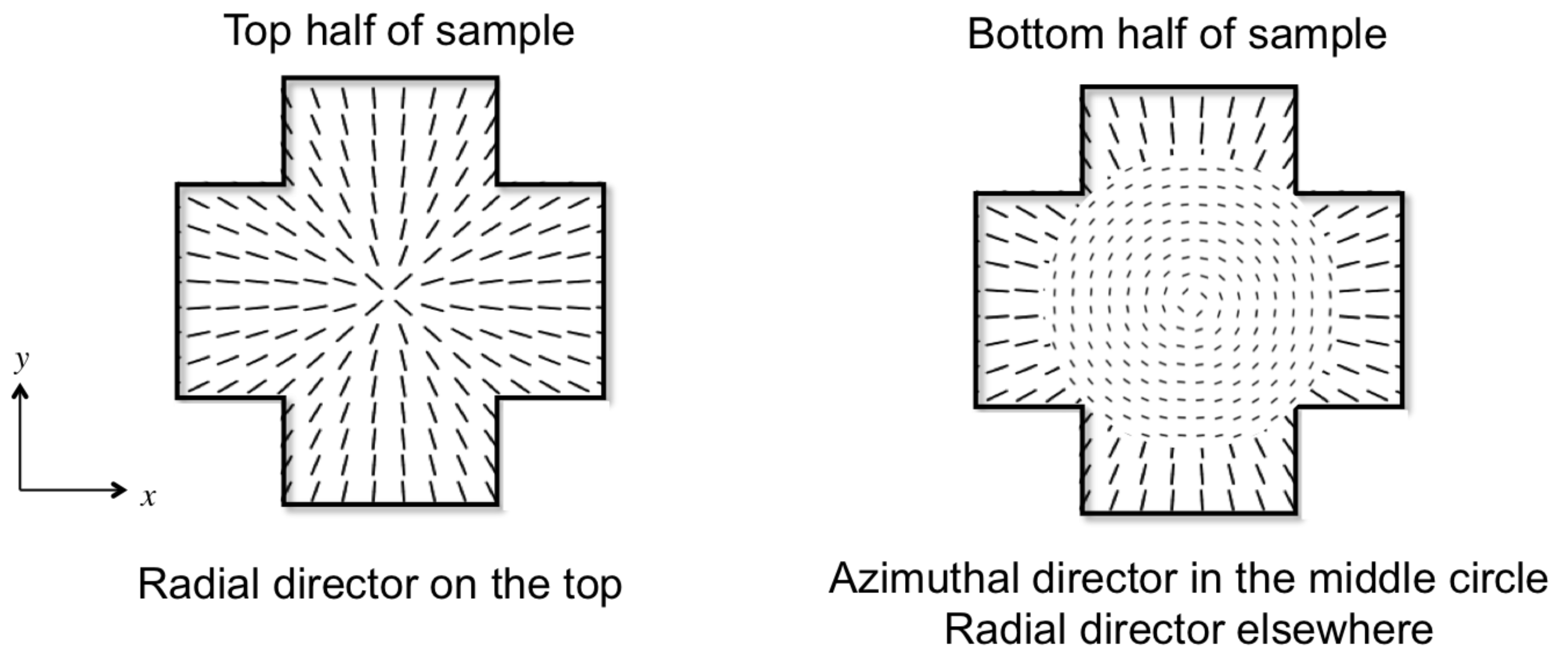
Twist through the thickness
…add chiral dopant to keep twist direction uniform

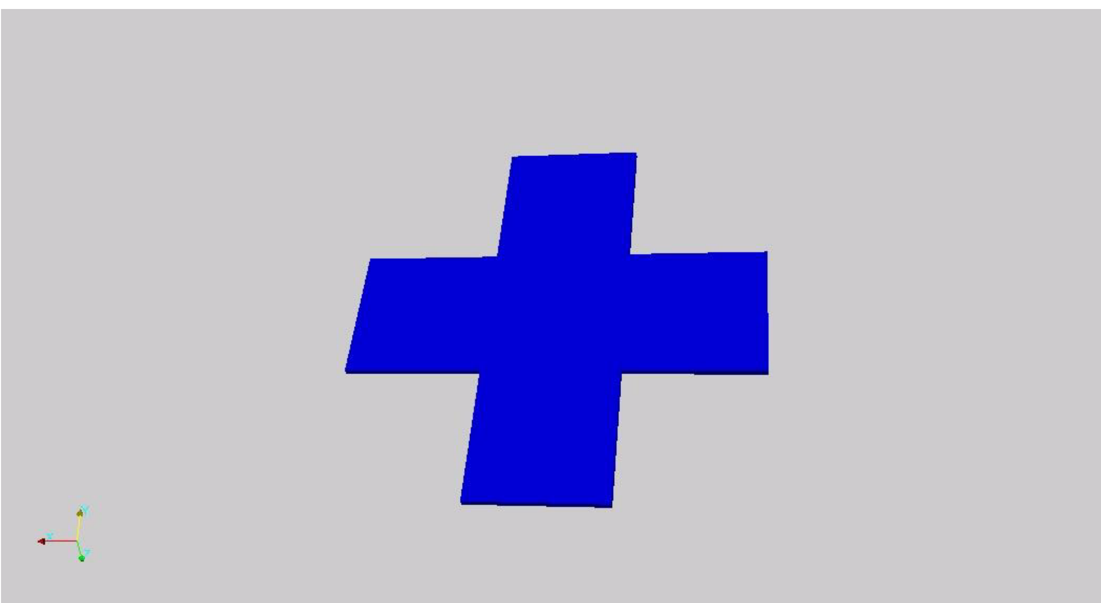
Azimuthal + radial = "Radimuthal"…. Term coined by Laurens de Haan



“Radimuthal” disk: makes sharp folds

Predicted by theory of Carl Modes and Mark Warner (C Modes, PRE 2012)



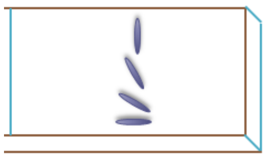


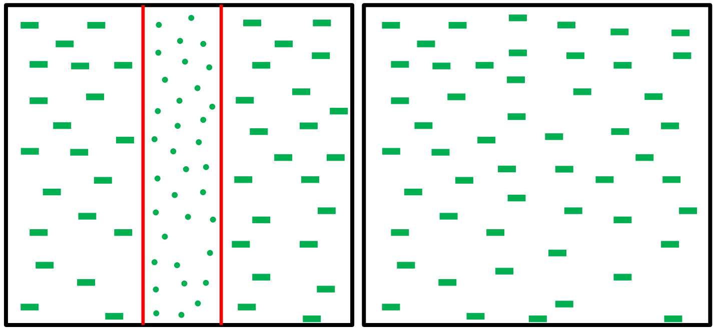
"Radimuthal" box

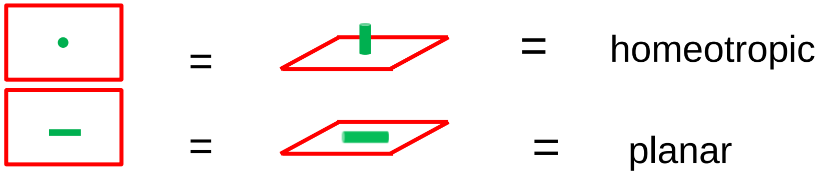
V Gimenez-Pinto et al
Scientific Reports 2017

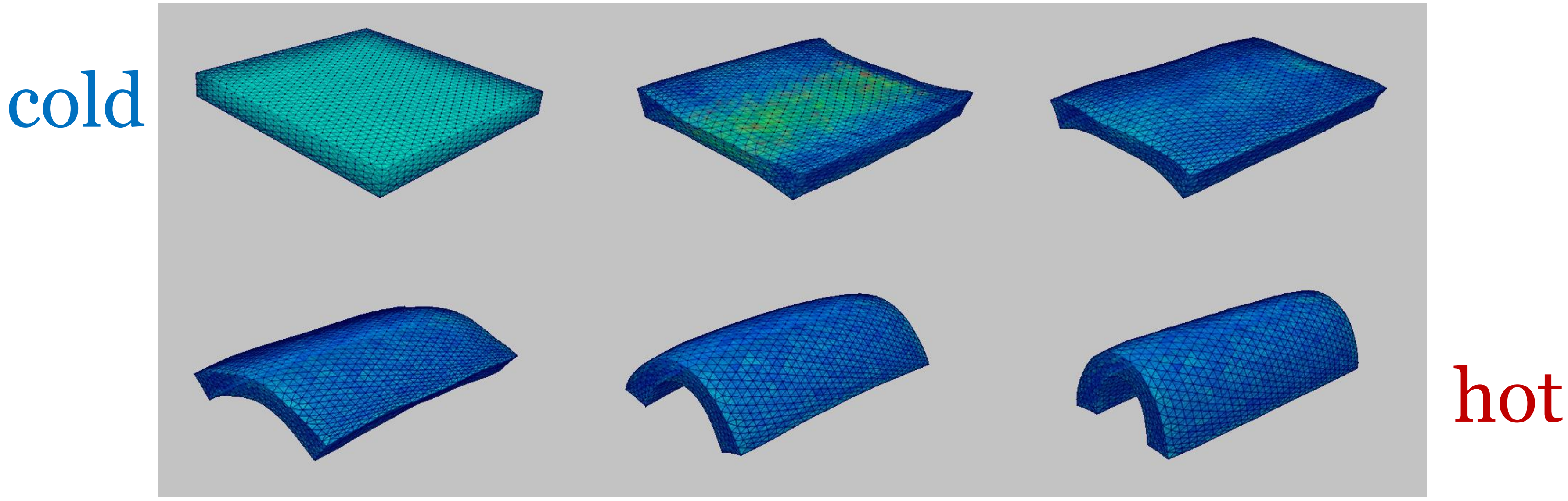
Splay hinge design:

Homeotropic anchoring on top



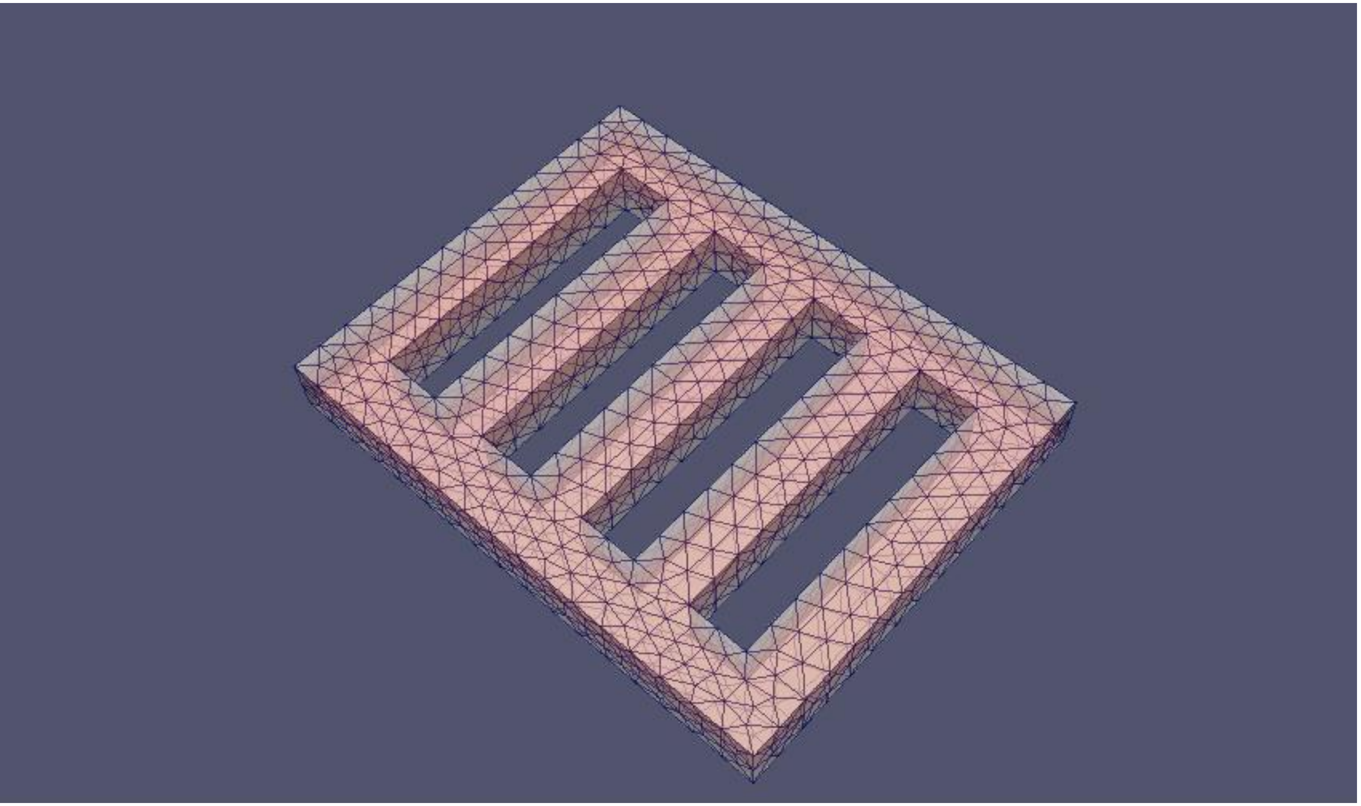




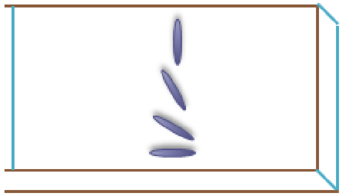


“Kirigami” approach: cut-outs accommodate strain in splay hinge

Cut-outs accommodate transverse strain



Cross beams: homeotropic on top, planar on bottom

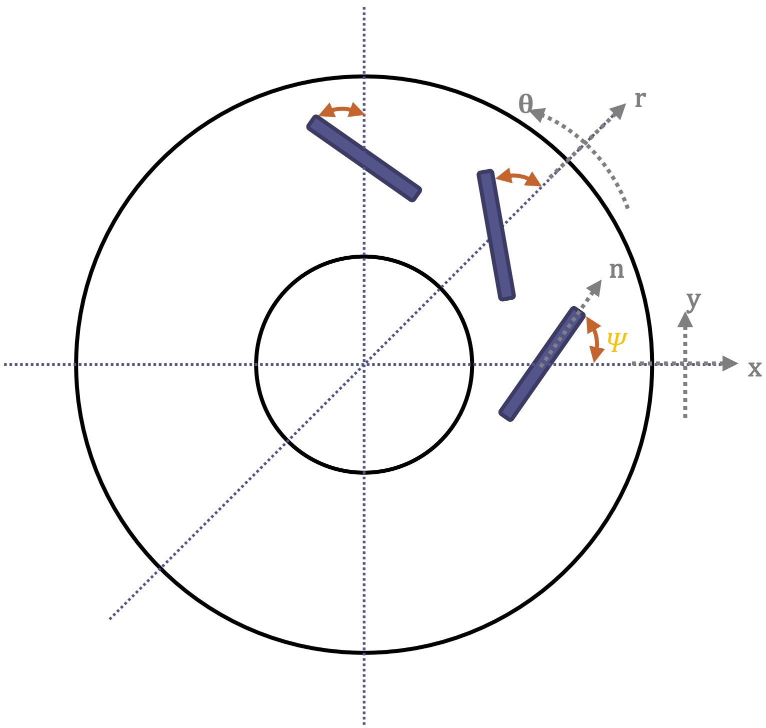


Long edges: isotropic

frontiers in Materials

A Konya, V Gimenez-Pinto, RLBS, 2016

Modeling an iris…annulus with director at an angle to radial; clamp outer edge

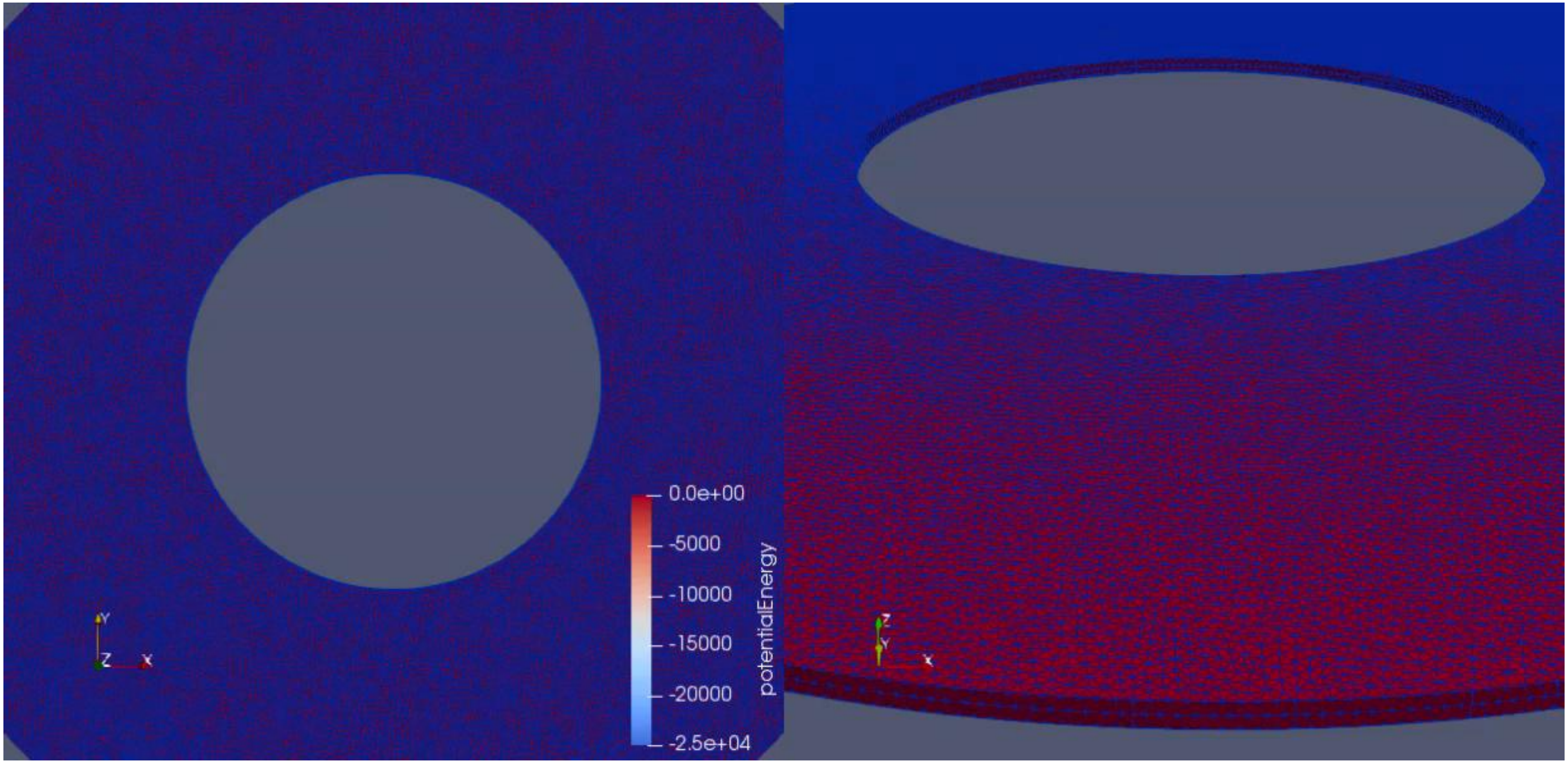


Initial (nematic) director field: n(Φ) = (cos Φ, Sin Φ,0)
at (x,y,z)
$\boldsymbol{\theta}$ = arctan(y/x)
$\Phi = \boldsymbol{\theta} + \Psi$

Opening the iris on heating: if director at an angle to radial direction, actuate with both dilation and rotation

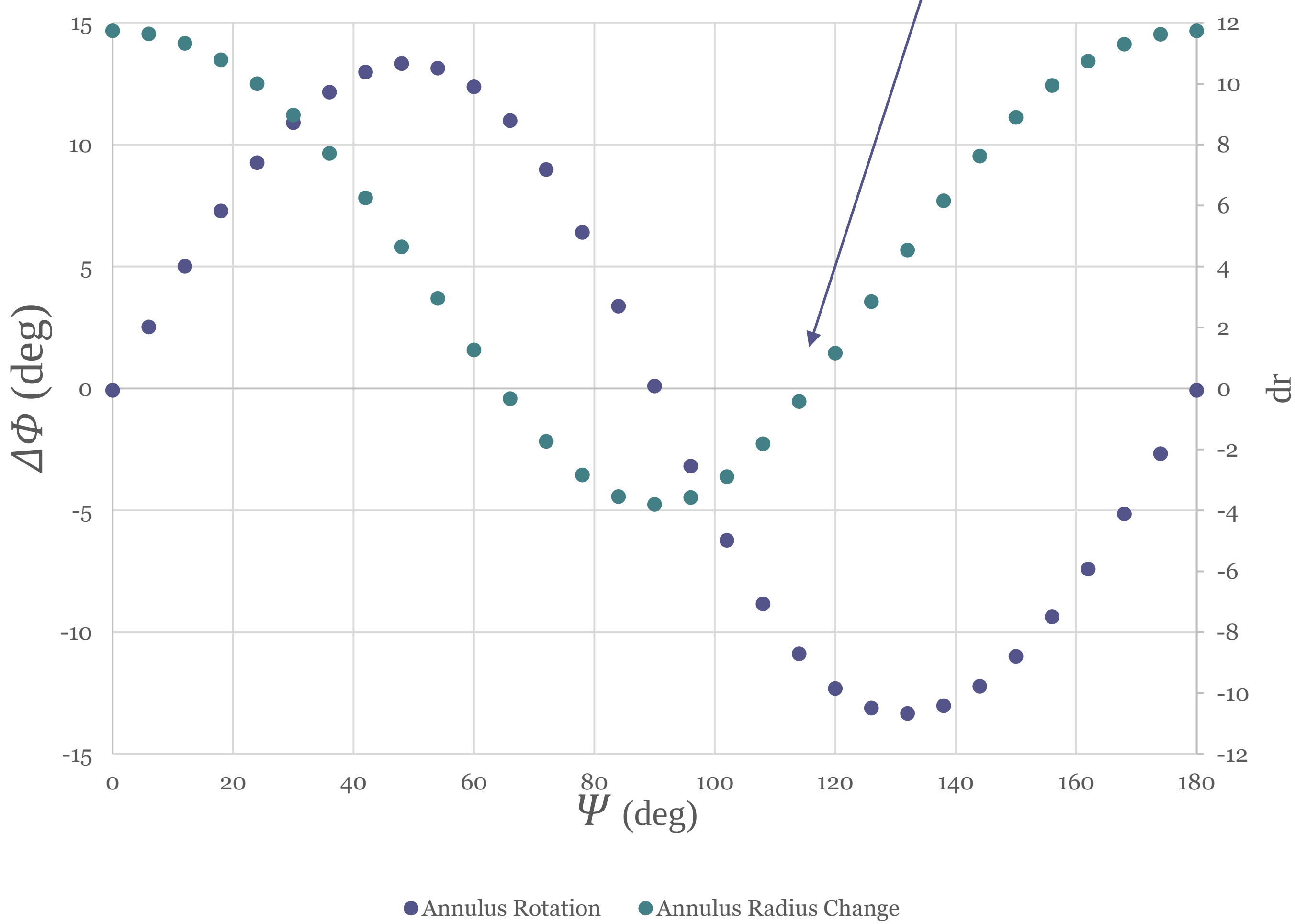
Director is 30deg from radial
30,000 time steps

Mesh:
nodes:88,816
tetrahedra: 315,090

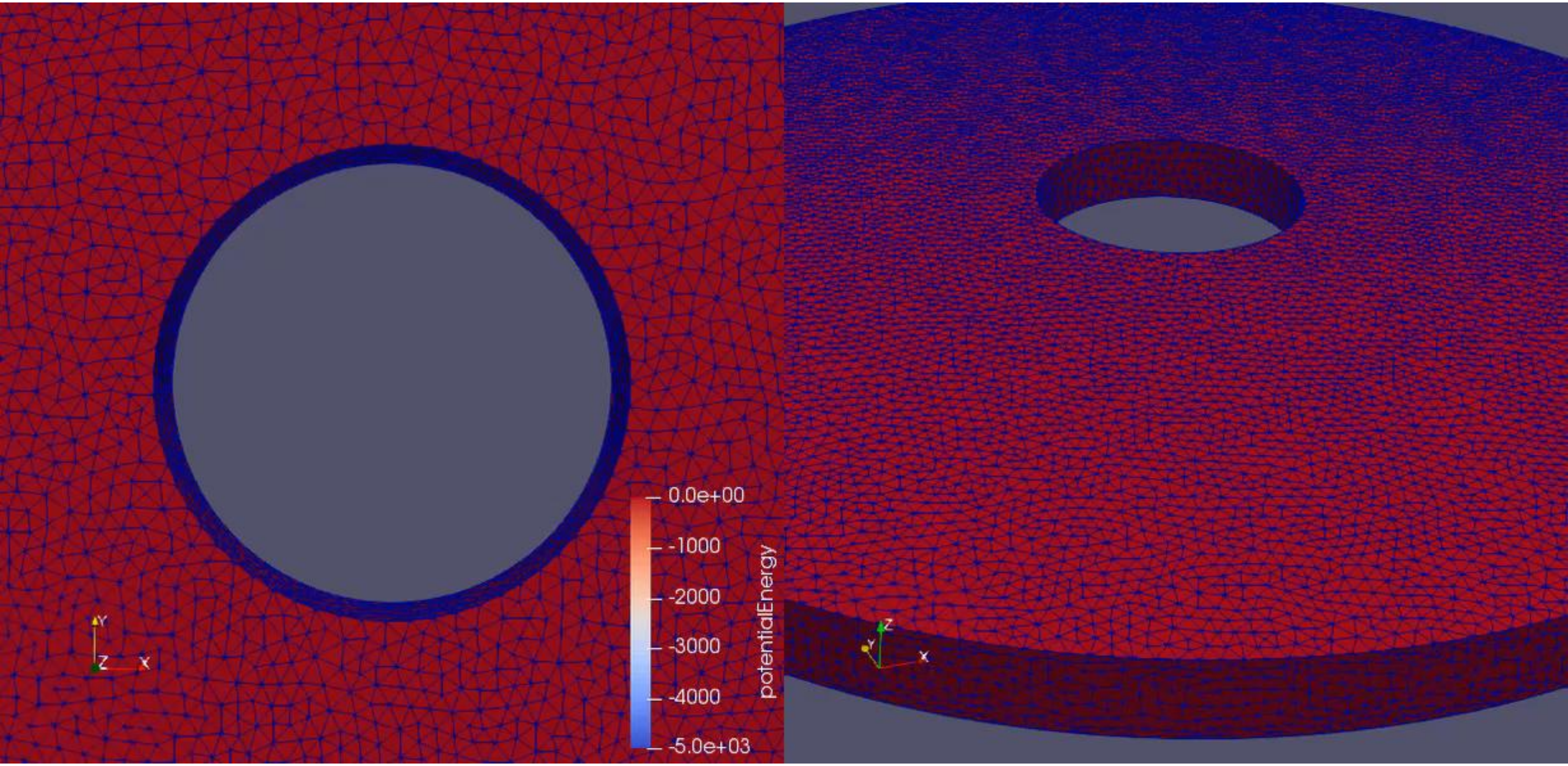


Rotation and dilation

At special angle, no dilation…
pure rotation!



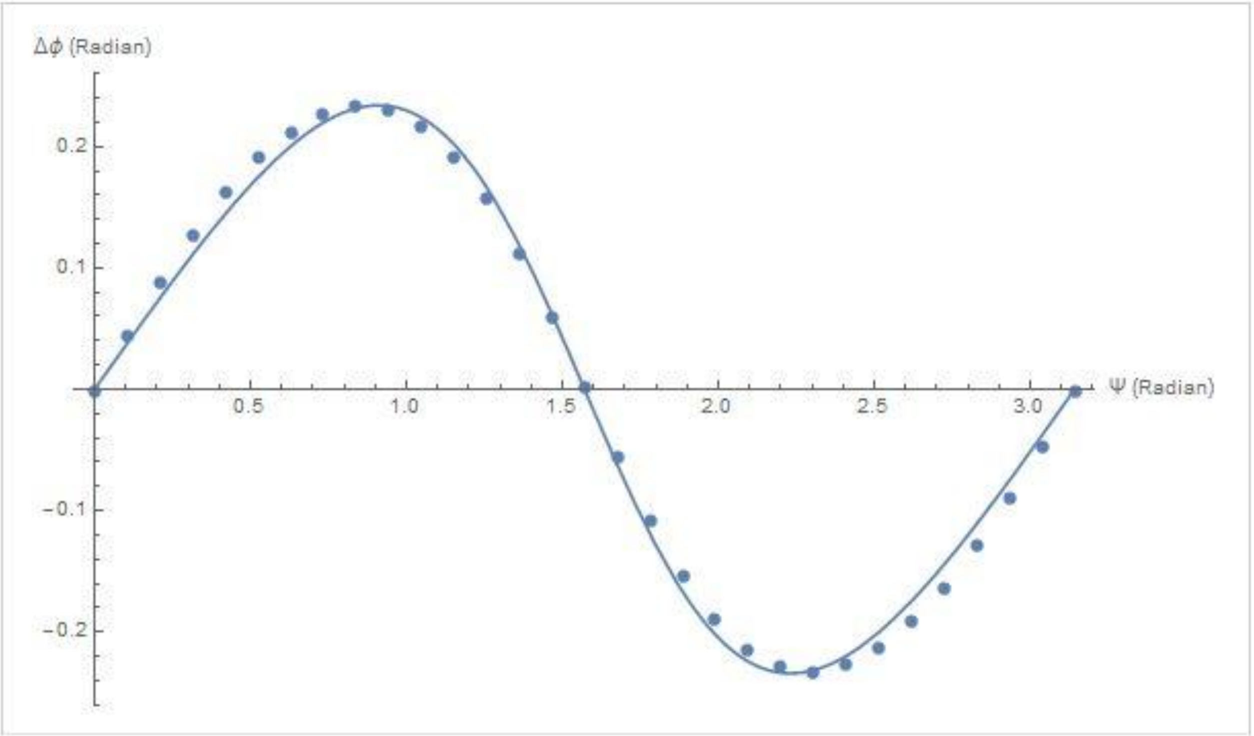
Pure rotation→ Torsional actuator



Analytical solution by Hillel Aharoni

$$\Delta\phi = \frac{\epsilon \sin(2\theta)}{1 + \epsilon \cos(2\theta)} \log\left(\frac{r_{out}}{r_{in}}\right)$$

$$\epsilon = \frac{\lambda^{-2\nu} - \lambda^2}{\lambda^{-2\nu} + \lambda^2}.$$

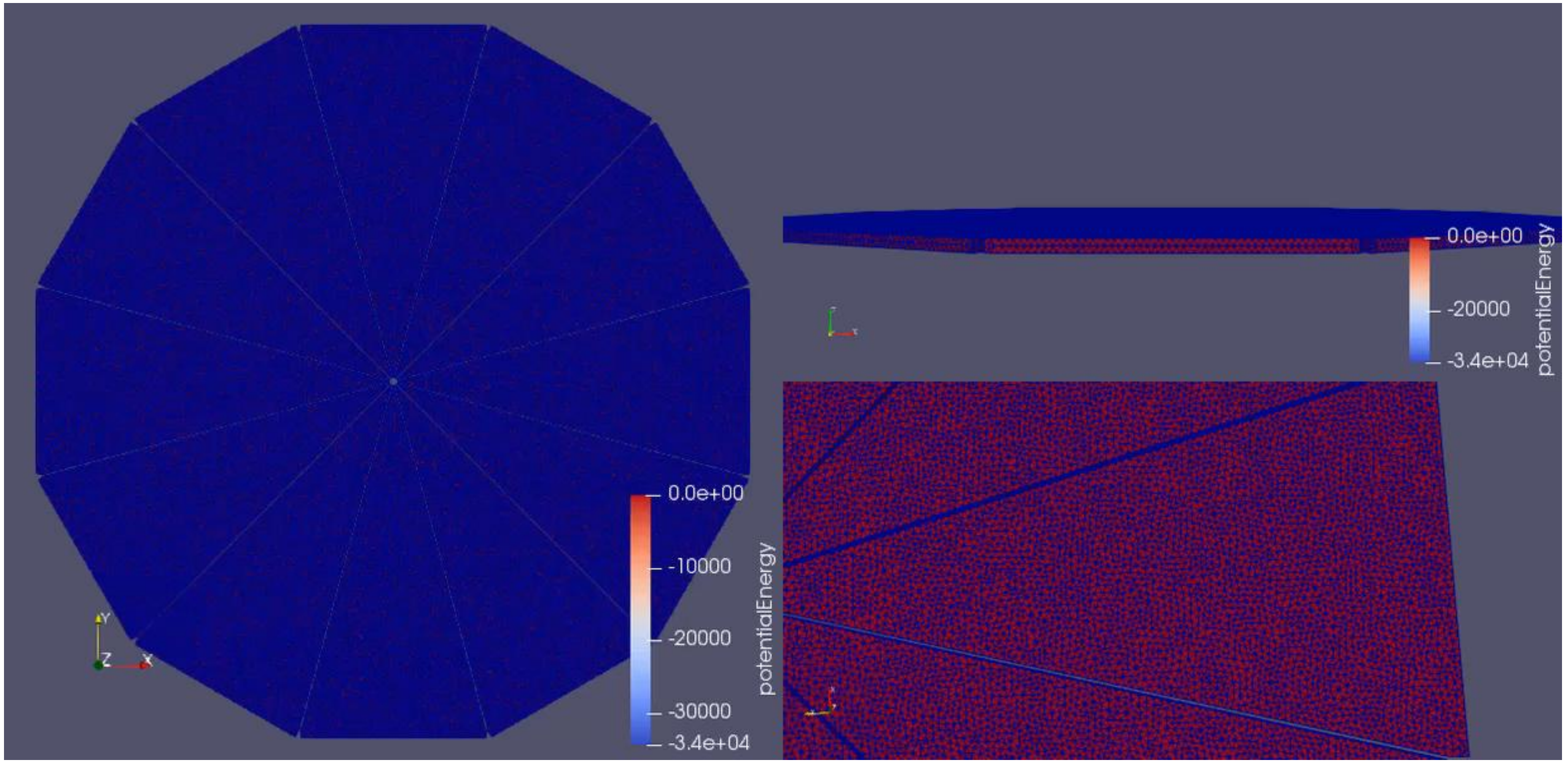


Input
$R_{out}(r_{out})$=100
$R_{in}(r_{in})$=40
υ=0.49

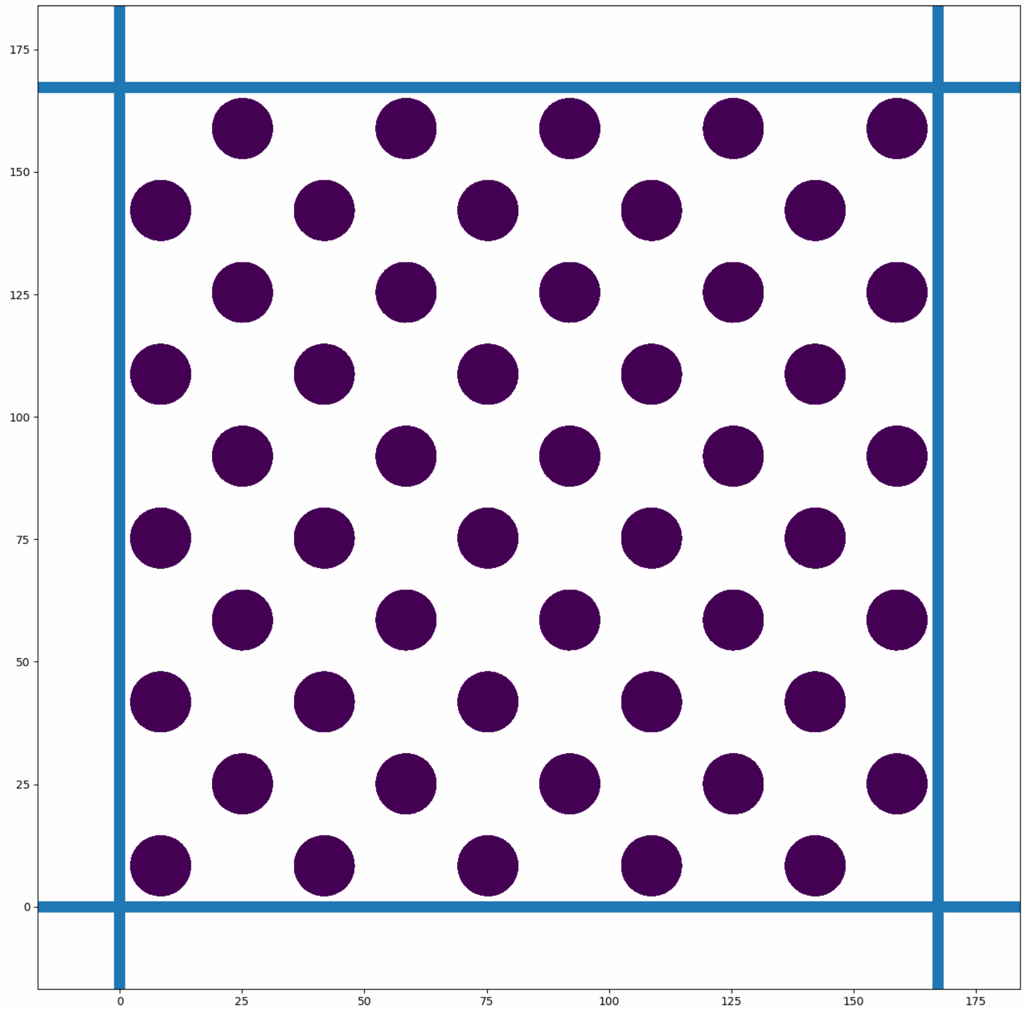
Matched
λ=0.844

λ measured at Ψ=0, where there is no rotation, is $(R_{out}\text{-}dr)/(R_{out})$ = 0.805. Plot above was obtained after matching λ=0.844

Another iris design …



Design studied in experiments by Priimagi, Advanced Materials 2017



Using FEM to model deformable colloids

Preliminary 2D results…

RLBS Collaboration with:

Manuel Valera-Slippery Rock
Craig Maloney –Northeastern

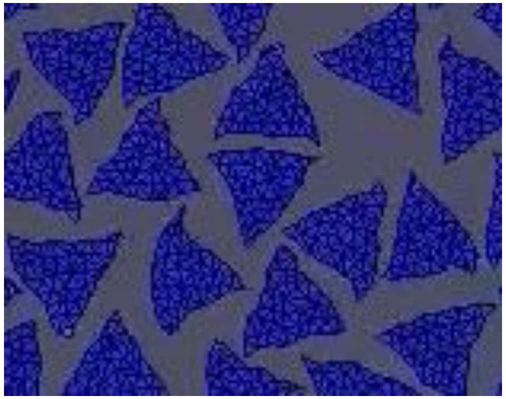
Can add…
- swelling effects
- internal dissipation
- friction
- hydrodynamic interactions

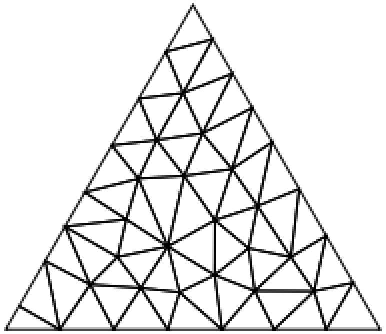
→ Implement in 3D with CUDA

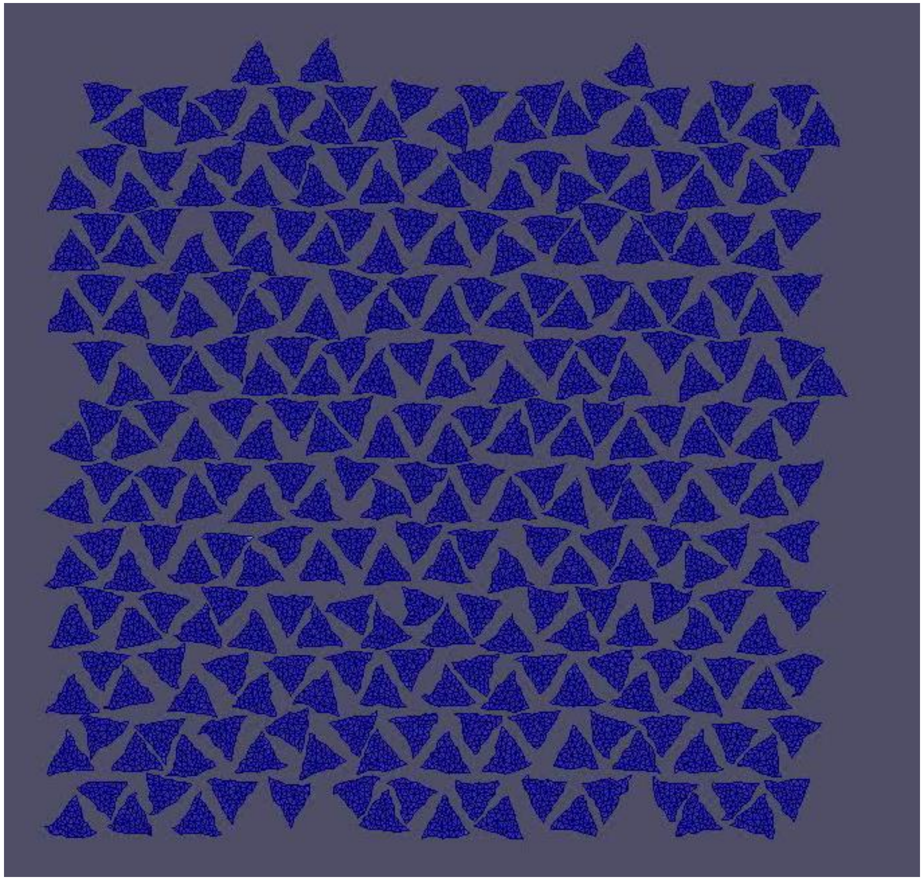
Possible application: visualize force chains in flexible granular media, different grain shapes

Particles don't have to be disks…

Could model any particle shape in 2D or 3D

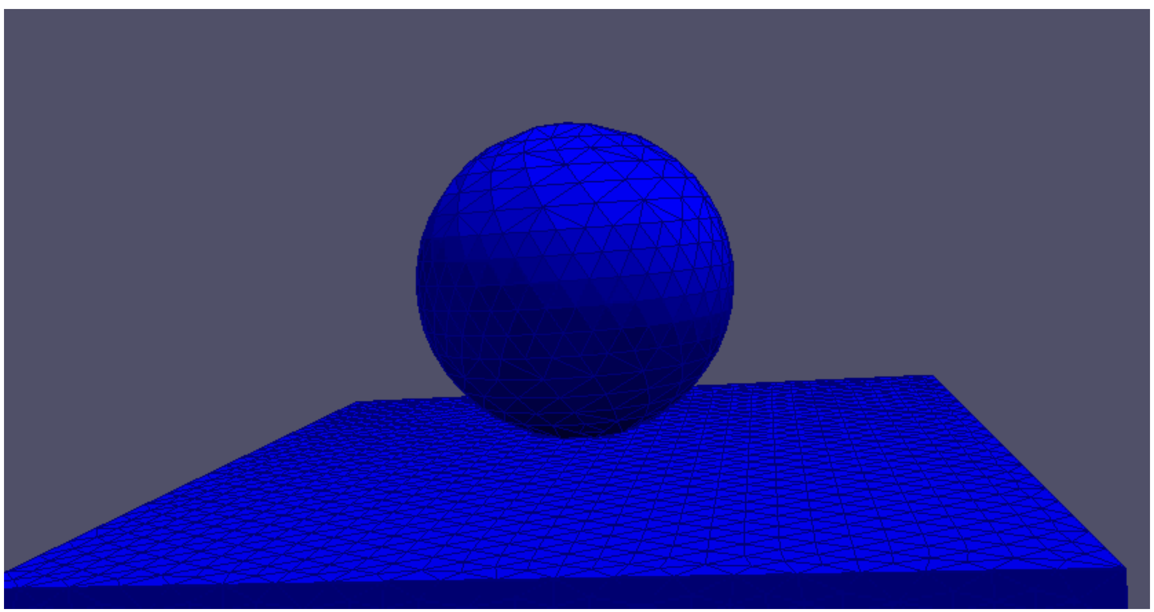






Repulsive forces on surface nodes
prevent objects from passing through each other

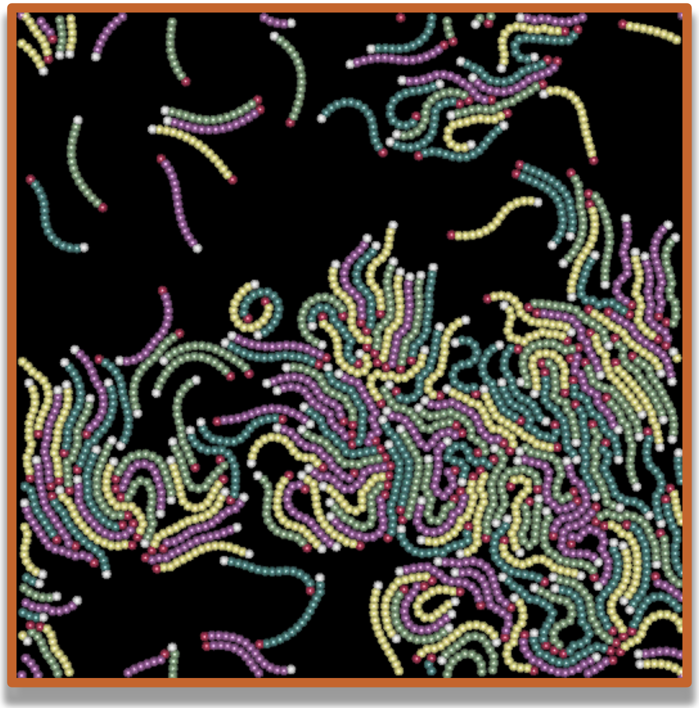
Surface node interactions → bumpy equipotential surfaces



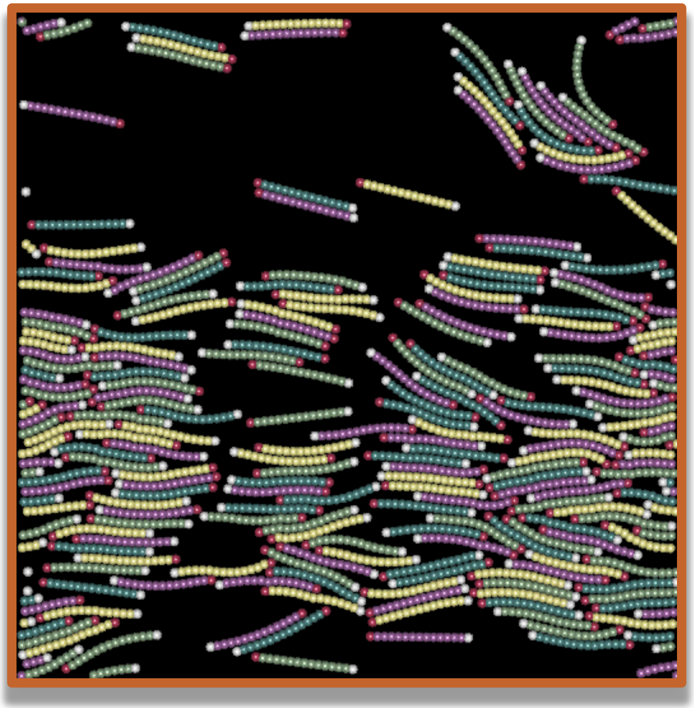
Other fun things we've been working on....

(if time allows)

Active matter: flexible self-propelled fibers

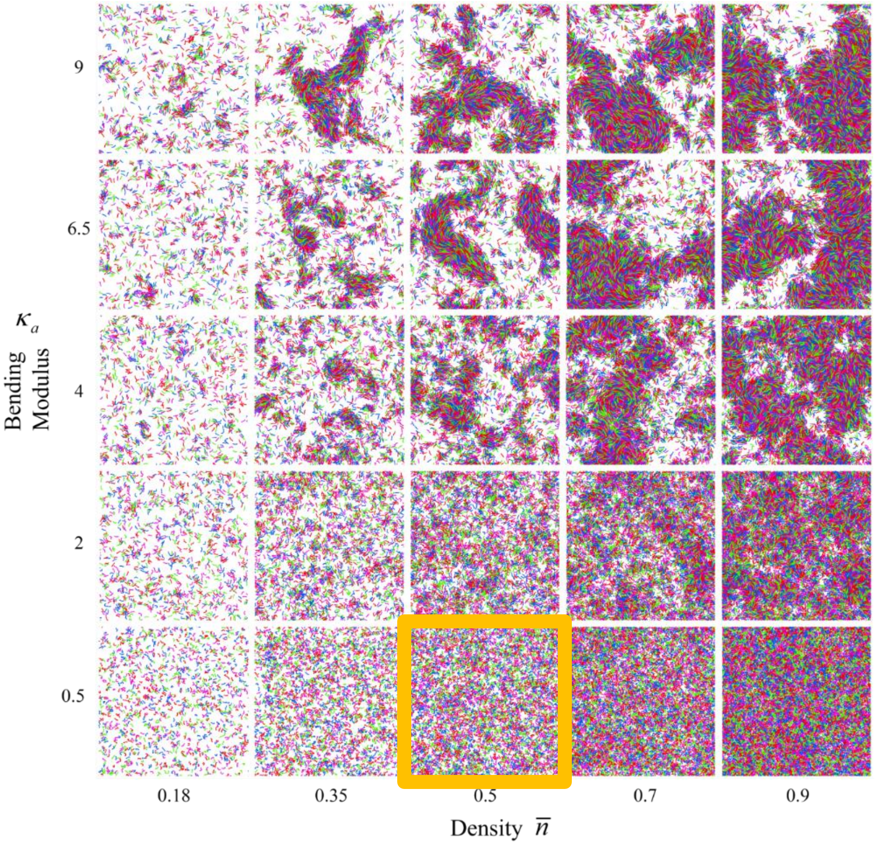


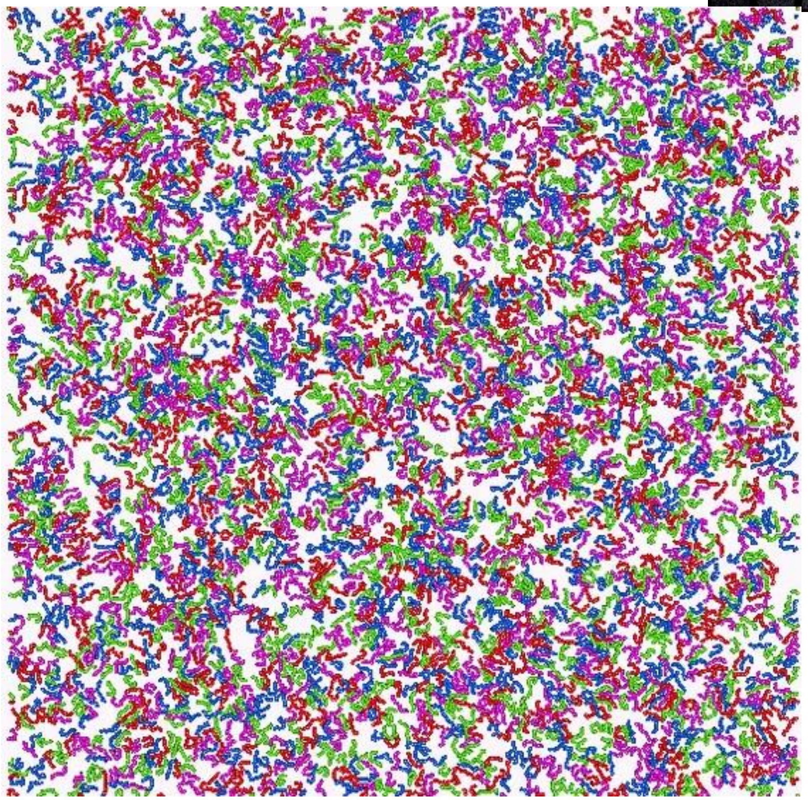
Lower bending modulus

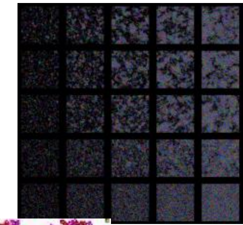


Higher bending modulus

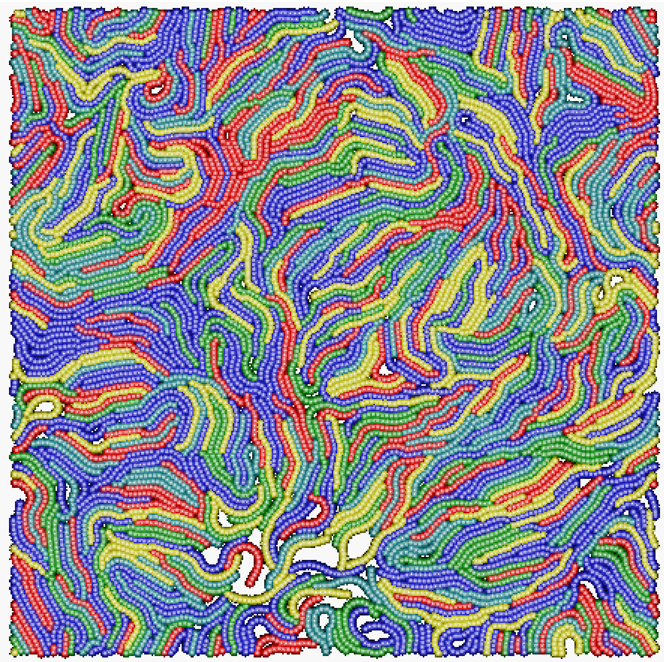
Phase diagram: bending modulus vs density



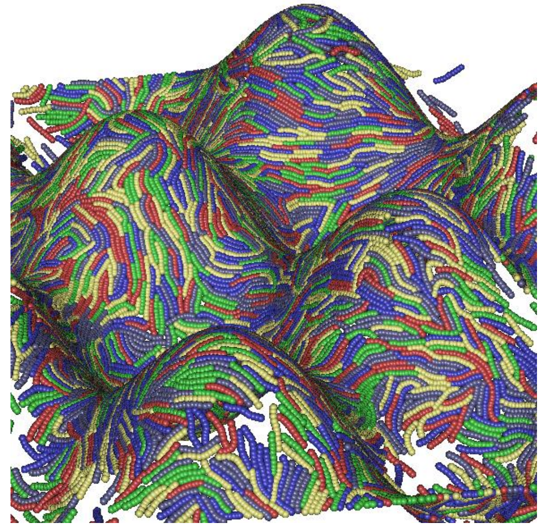


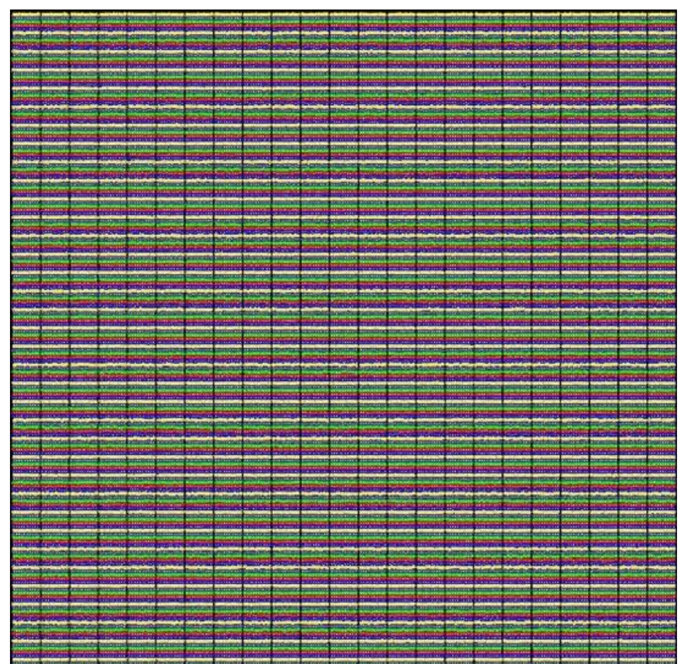


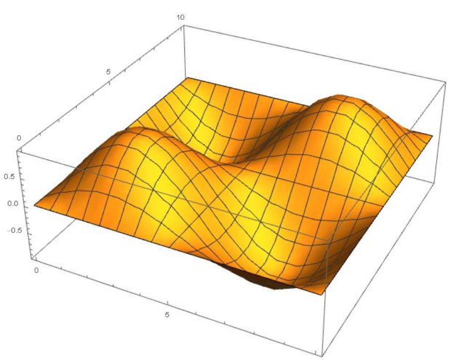
Flexible Active Filaments on Curved Surfaces

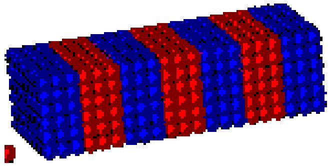


Flexible active filaments







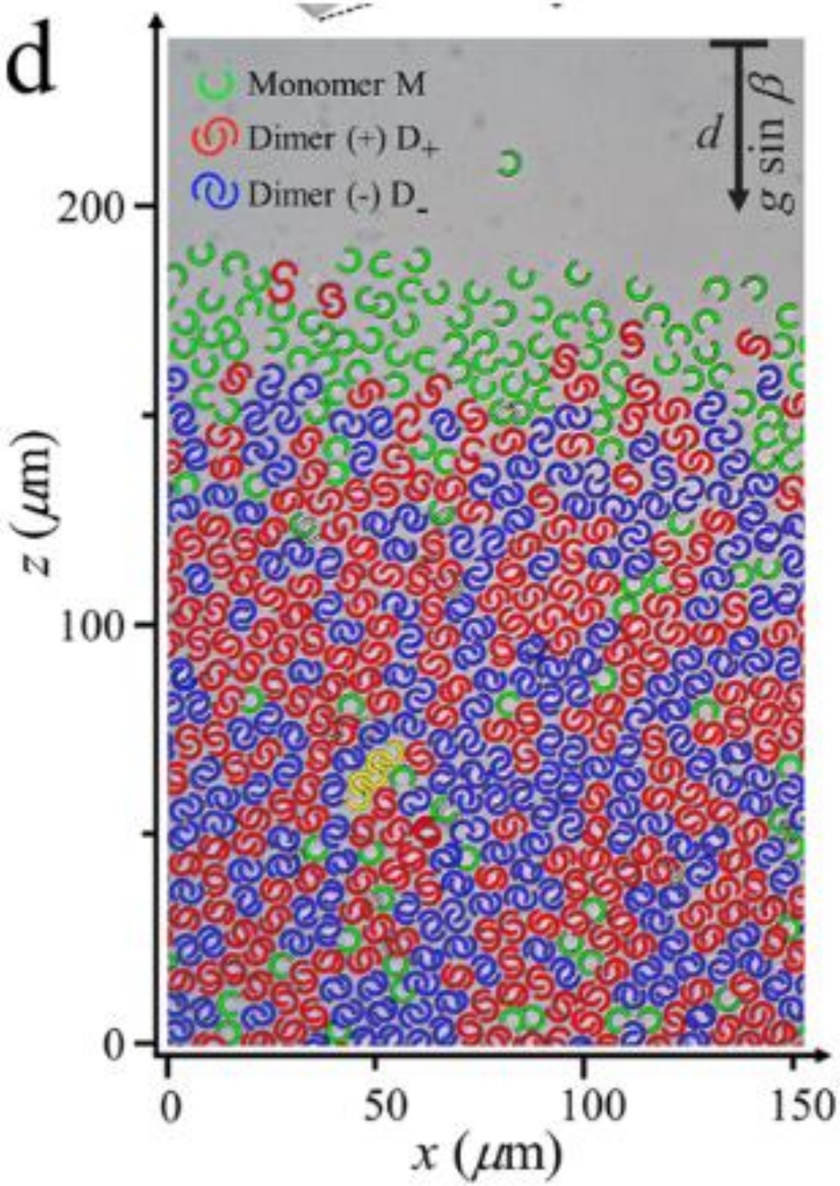


More detailed models of soft active particles using FEM elastodynamics

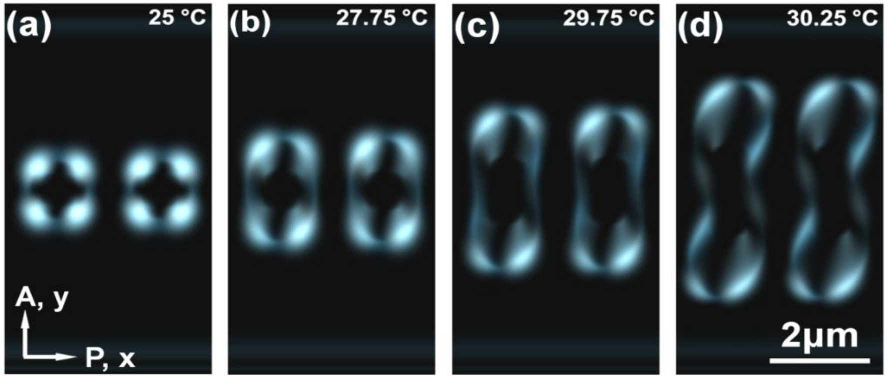
… animal locomotion?

C-shaped colloids → chiral dimers: Spontaneous formation of homochiral domains

Experiments by PY Wang and Tom Mason, JACS 2015



Simulation by Dong Li and RLBS



"Cooking **Skyrmions** until they pop"

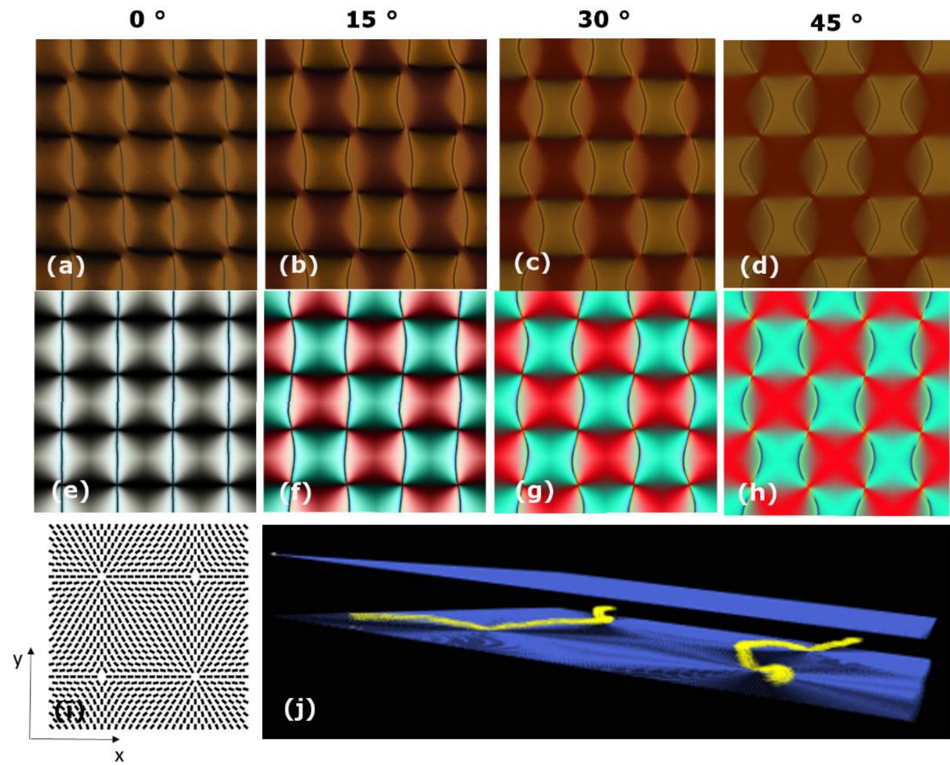
Bubble defects in cholesteric LC's confined in microfluidic channels expand when heated

Simulations
by Sajedeh Afghah

Cholesteric liquid crystals in rectangular microchannels: skyrmions and stripes

Yubing Guo,† Sajedeh Afghah,† Jie Xiang,† Oleg D. Lavrentovich, Robin L. B. Selinger and Qi-Huo Wei*

Soft Matter, 2016



Nematic between substrates with surface-patterned point defects
Collaboration with Qi-Huo Wei

Collaborators…

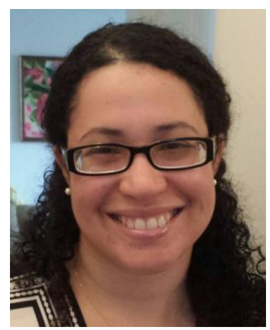
Anne Hélène Gélébart and Dirk Broer
TU Eindhoven

Jonathan Selinger
Kent State

Manuel Valera
Slippery Rock University

Andrew Konya

Michael Varga



Vianney Gimenez-Pinto

Youssef Golestani

Current and former students…

Conclusions

Liquid crystal elastomers encode complex shape actuation trajectories

- Simulation studies of the forward problem: for a given sample shape and director field, calculate trajectory
  - →Degeneracy, metastable states, snap-through, hysteresis
  - → Design motifs for actuators and simple machines
- Director field can encode photoactuated wave motion
- Origami folding: designing the best fold
- Simple machines: iris, torsion actuator

- Preliminary studies of flexible colloids

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References: tiny.cc/selinger2018