Biomimetic 4D Printing:

Programmable Shape Shifting 3D Printed Polymer Gels

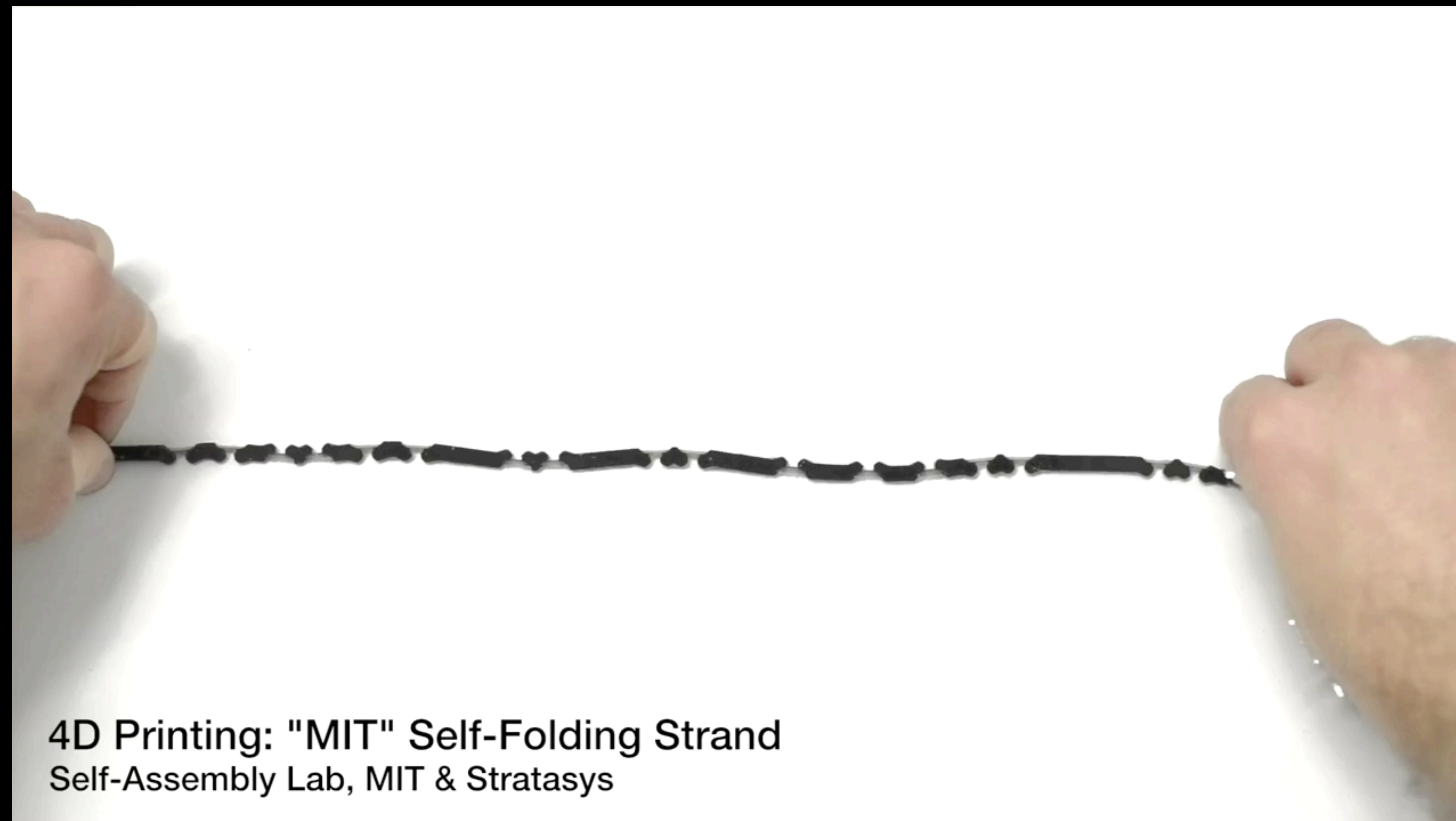
Sabetta Matsumoto (she/her/hers) Georgia Institute of Technology

3rd Edwards Symposium ~ 7 Sept 2018

Shape-morphing Systems

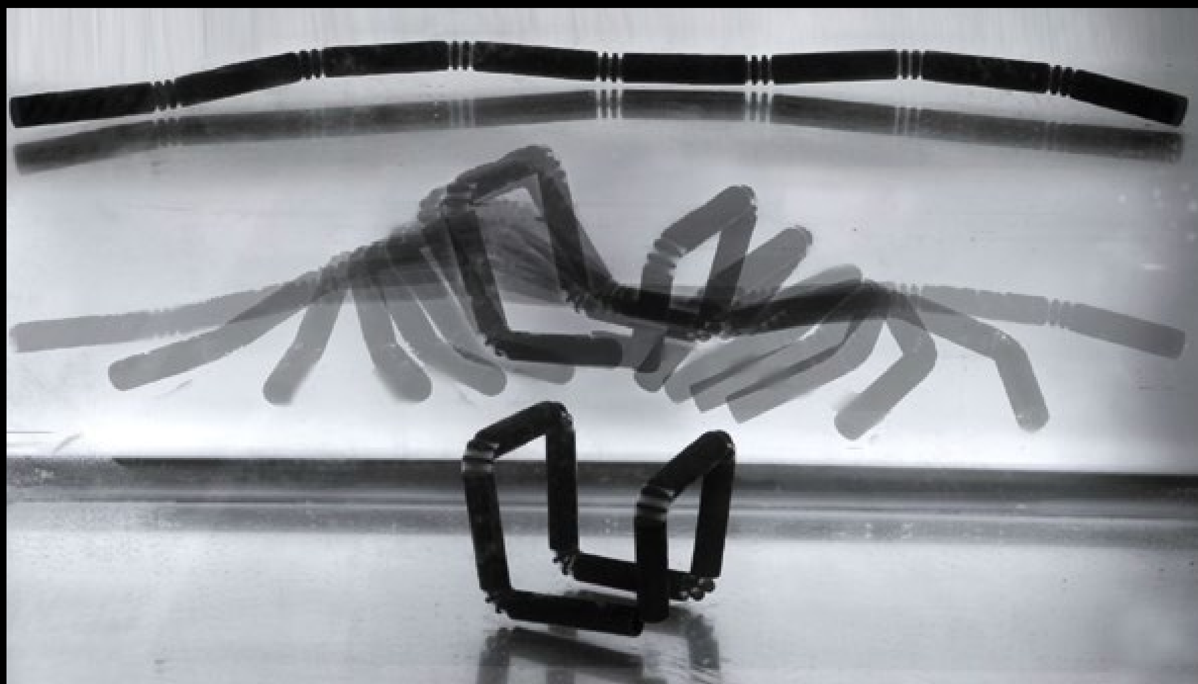
Shape-morphing Systems

4D Printing


4D Printing: "MIT" Self-Folding Strand
Self-Assembly Lab, MIT & Stratasys

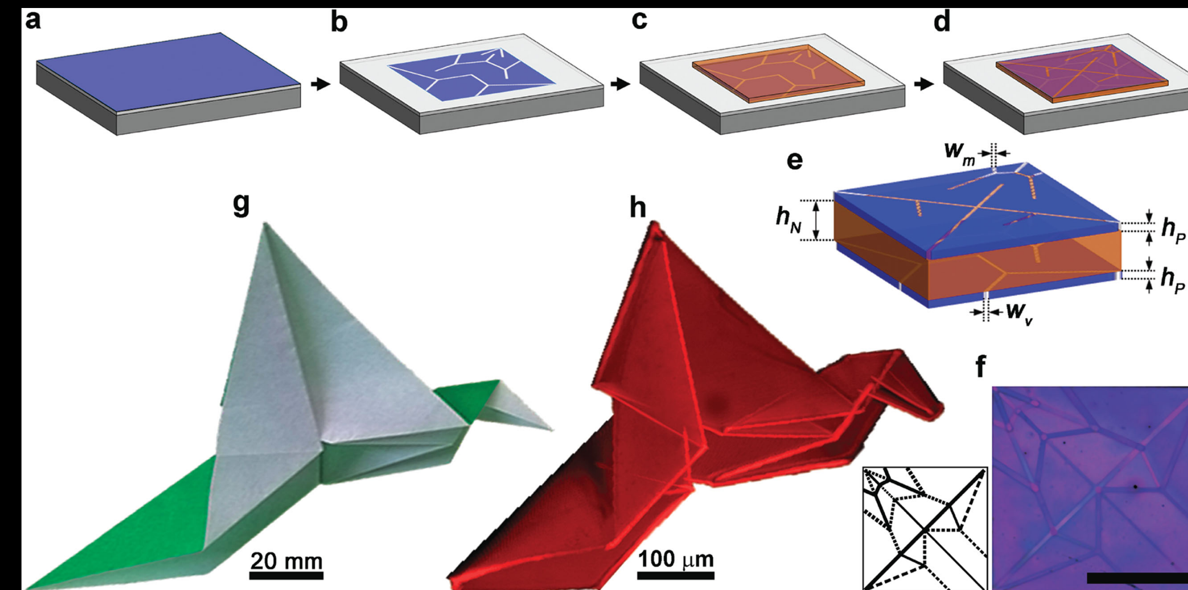
Shape-morphing Systems

Mechanical Hinges



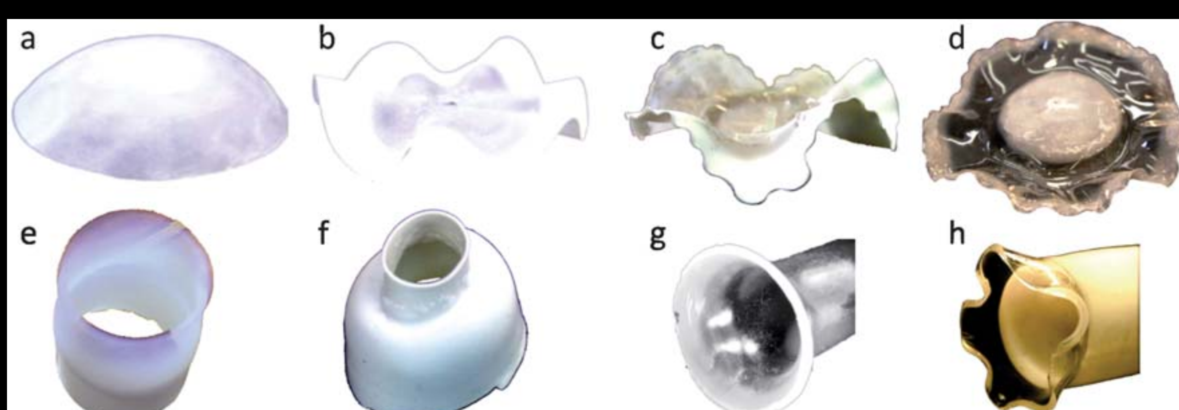
Tibbits, *Arch. Design* **84** 116 (2014)

Origami



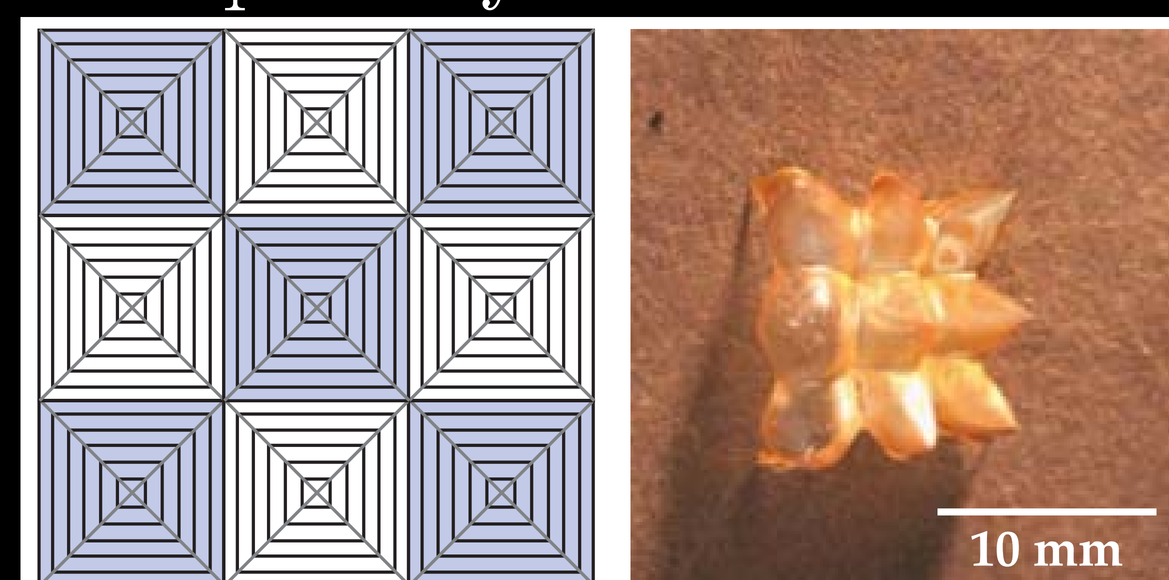
Na, *et al. Adv. Mat.* **27** 79 (2015)

Swelling Hydrogels



Sharon & Efrati, *Soft Matter* **6** 5693 (2010)

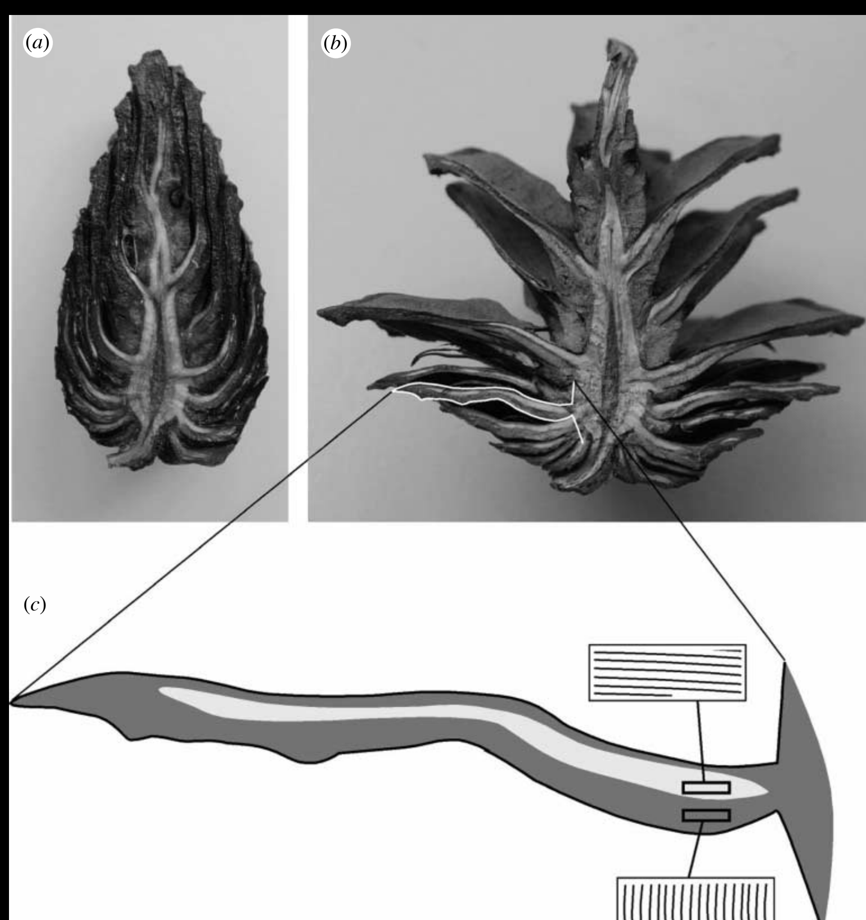
Liquid Crystal Elastomers



Modes, *et al. Phys. Today.* **69** 32 (2016)
Ware, et al. Science 347, 982 (2015)

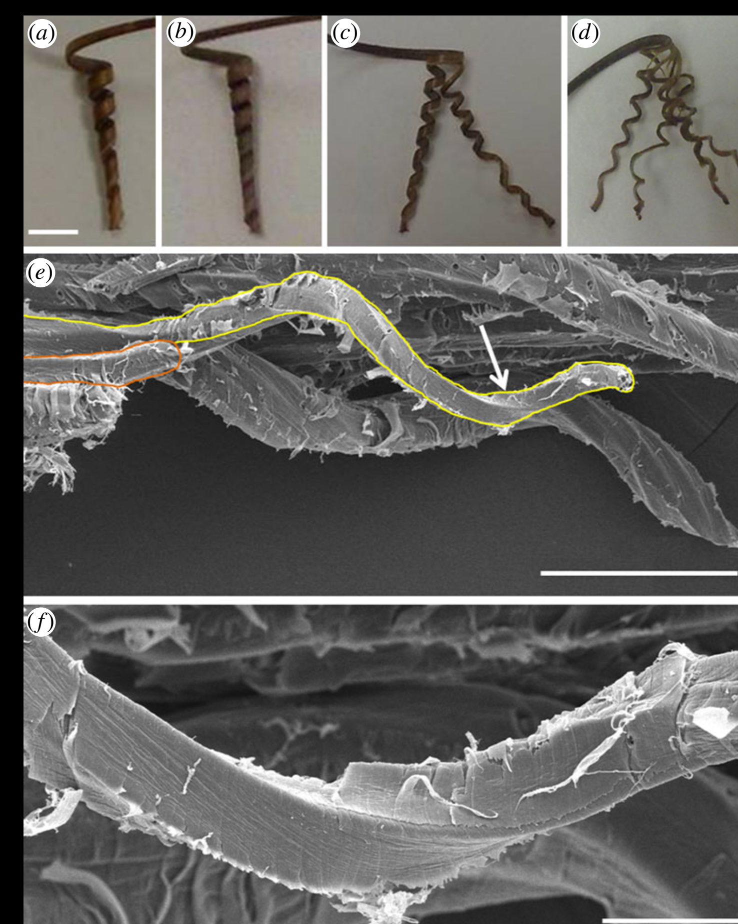
Hygroscopic Motion

Pine Cone



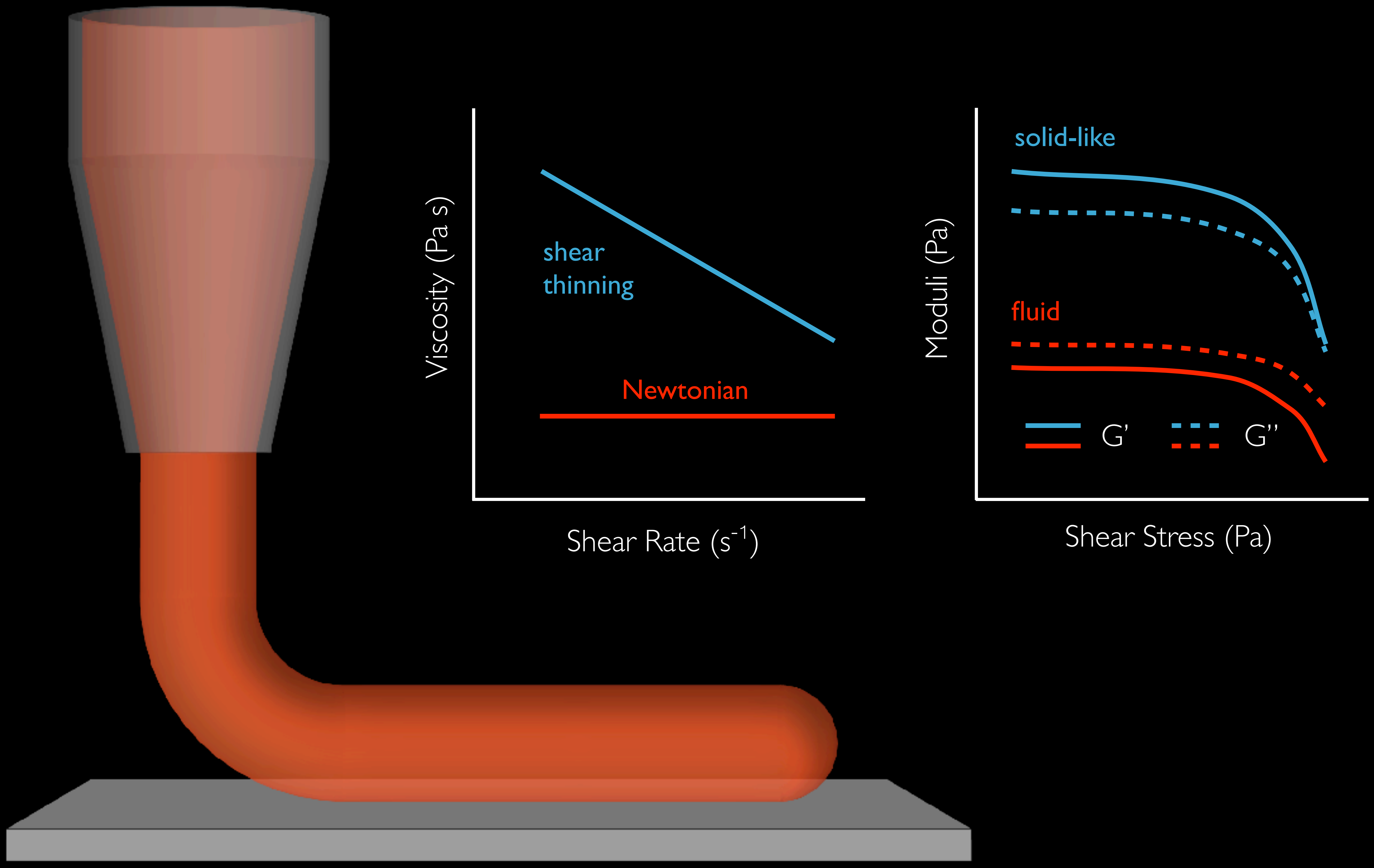
Burgert & Fratzl, *Phil. Trans. R. Soc. A* **367** 1541 (2009)

Erodium Awn



Abraham, *et al. J. R. Soc. Interface* **9** 640 (2012)

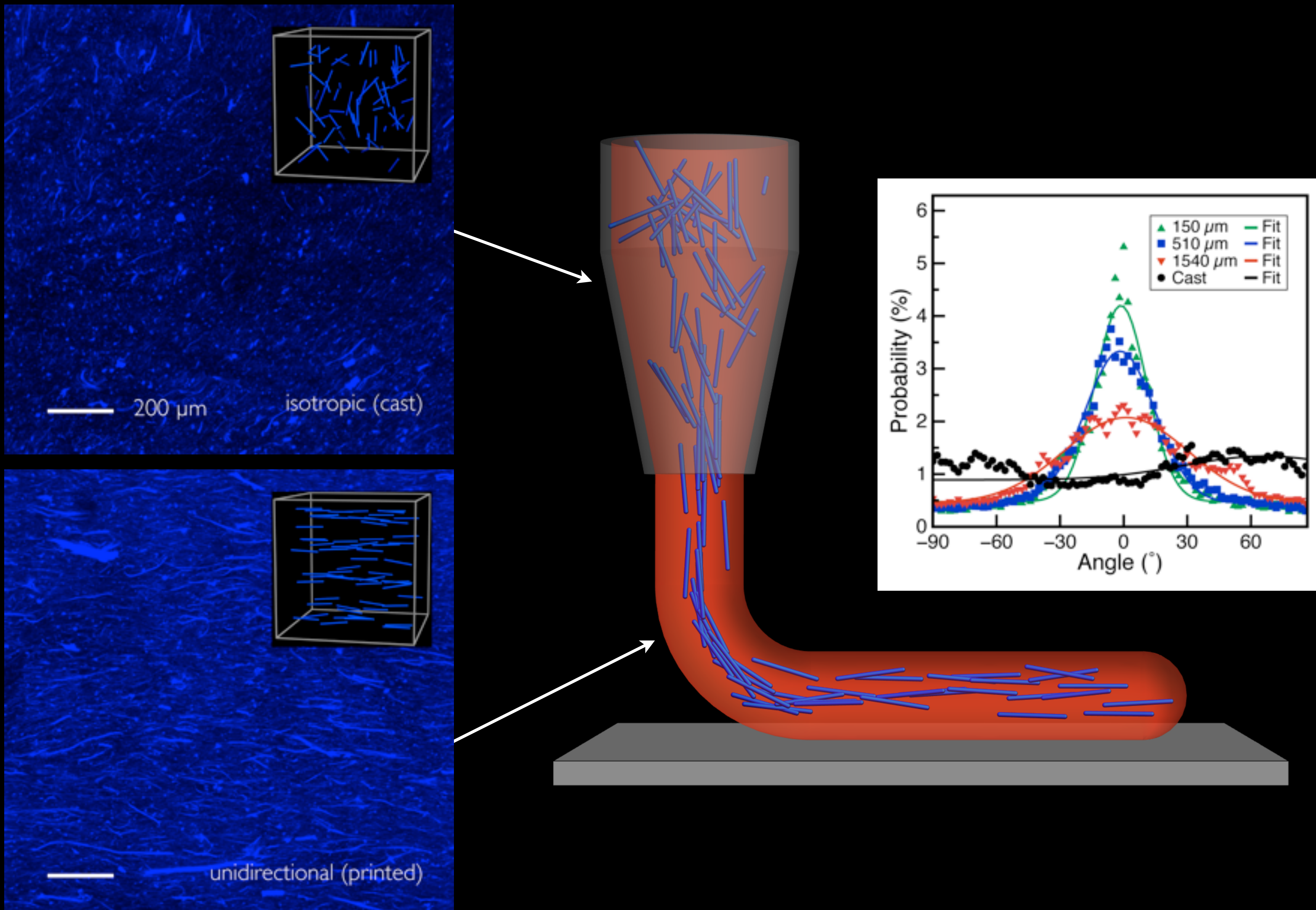
3D Printing



A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

Our Ink

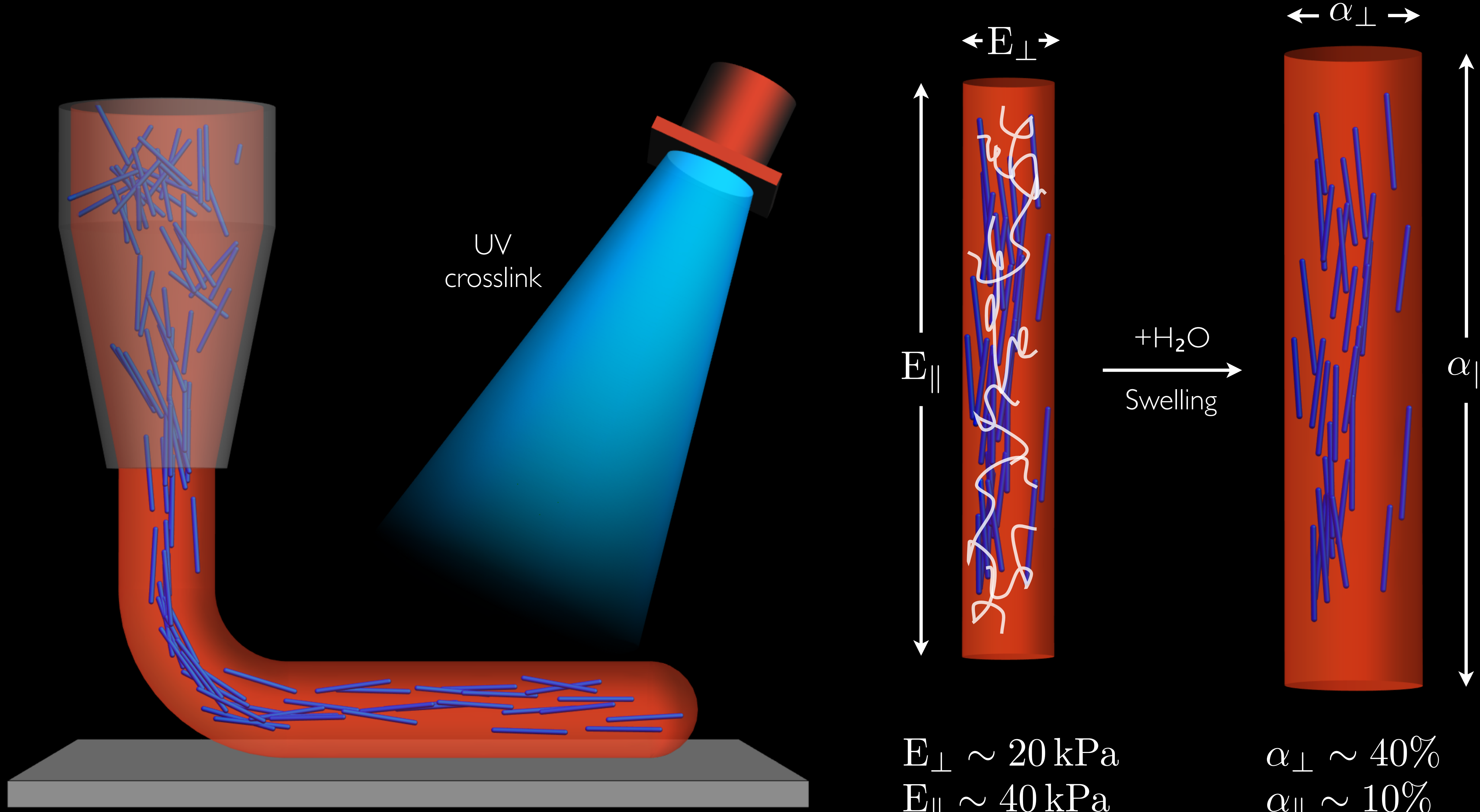
Cellulose Nanofibrils + Acrylamide Monomers + Clay = Composite Ink



A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

Our Ink

Cellulose Nanofibrils + Acrylamide Monomers + Clay = Composite Ink



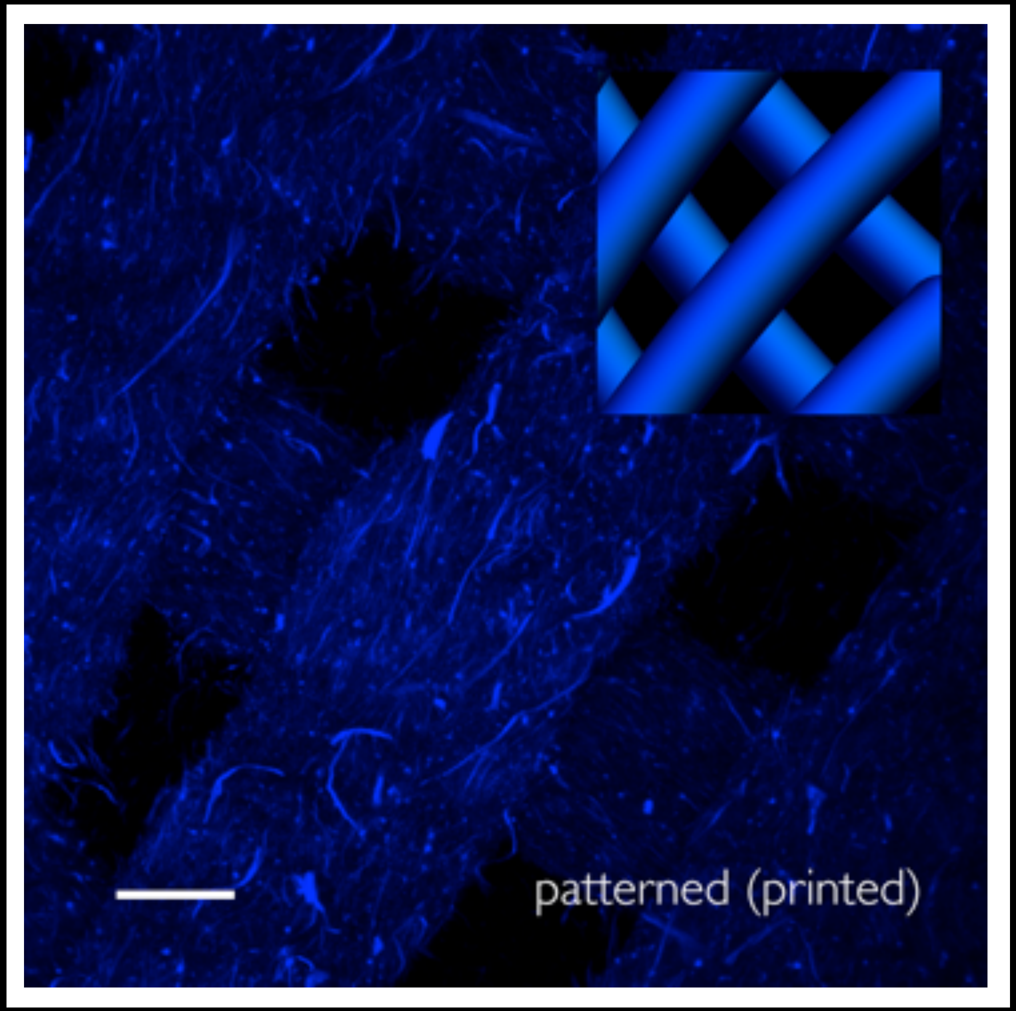
$E_\perp \sim 20\,\mathrm{kPa}$
$E_\parallel \sim 40\,\mathrm{kPa}$

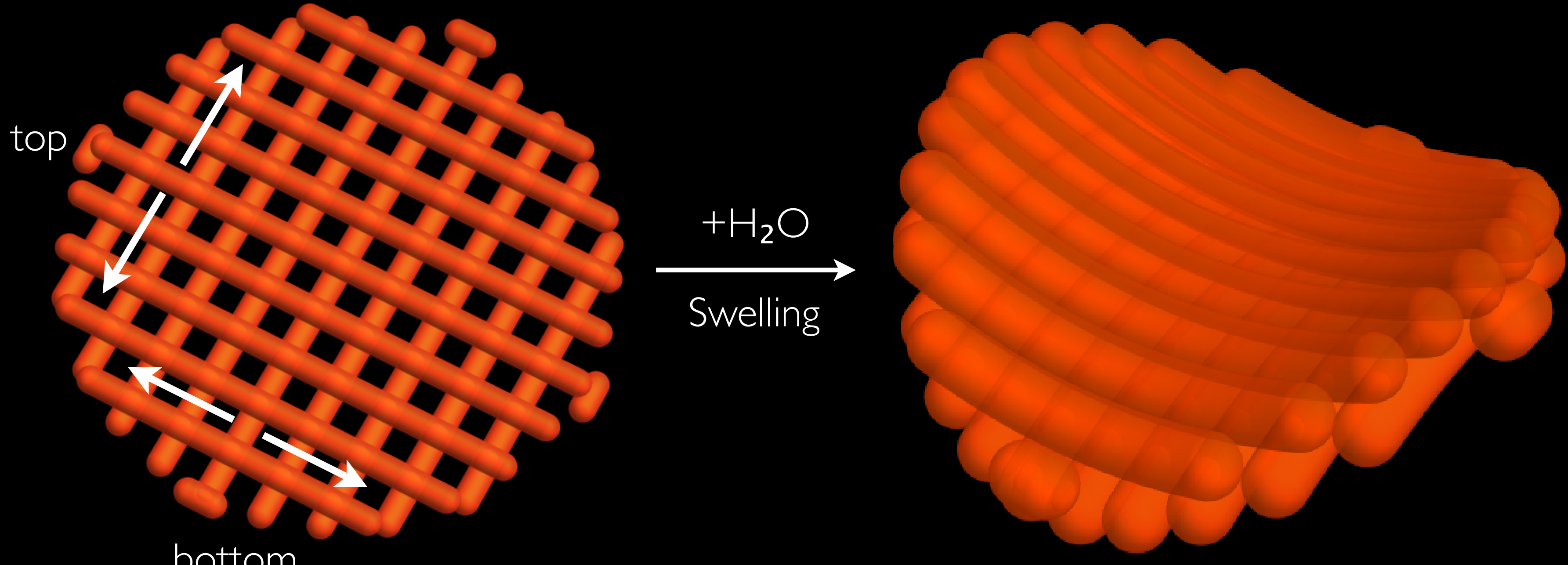
$\alpha_\perp \sim 40\%$
$\alpha_\parallel \sim 10\%$

Elastic Anisotropy leads to swelling anisotropy

A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

Encoding Local Anisotropy



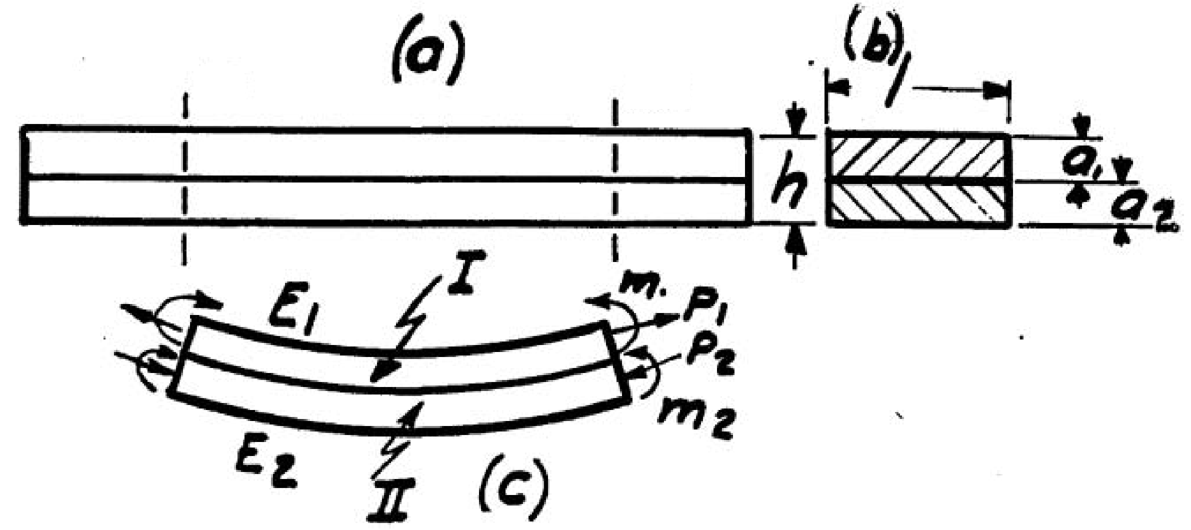


A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

Bi-Metallic Strips

1. Equilibrium Condition

"Due to the fact that there are not external forces acting on the strip, all forces acting over any cross-section of the strip must be in equilibrium"


ANALYSIS OF BI-METAL THERMOSTATS

BY S. TIMOSHENKO

FIG. 1. *Deflection of a bi-metal strip while uniformly heated.*

Stress from fictitious force

$$\sigma = \frac{\mathrm{P}^{\mathrm{eff}}}{h}$$

Bending Moment from stress

$$M^{\mathrm{tot}} = \int z\sigma dz = \frac{\mathrm{P}^{\mathrm{eff}} h}{2}$$

Bending Moment from curvature

$$\begin{aligned} M^{\mathrm{tot}} &= \int_{-a_2}^{a_1} z\mathrm{E}(z)\varepsilon dz \\ &= \kappa \mathrm{E}_1 \int_0^{a_1} z^2 dz + \kappa \mathrm{E}_2 \int_{-a_2}^{0} z^2 dz \\ &= \frac{\kappa}{3}\left(\mathrm{E}_1 a_1^3 + \mathrm{E}_2 a_2^3\right) \end{aligned}$$

Moment-stress relationship

$$\sigma^{\mathrm{tot}} = \frac{M^{\mathrm{tot}}}{2h}$$

Timoshenko, *J. Opt. Soc. Am.* **11** 233 (1925)

Bi-Metallic Strips

2. Compatibility Condition

"On the bearing surface of both metals the unit elongation occurring in the longitudinal fibres of metals (1) and (2) must be equal."

$$\varepsilon^{(1)} = \varepsilon^{(2)}$$

Strain from swelling $\varepsilon^{\mathrm{s}} = \alpha$

Strain from curvature $\varepsilon^{\mathrm{s}} = z\kappa$

Strain from stress $\varepsilon = \mathrm{E}^{-1}\sigma^{\mathrm{tot}} = \frac{1}{\mathrm{E}}\frac{M^{\mathrm{tot}}}{2h}$

ANALYSIS OF BI-METAL THERMOSTATS

By S. Timoshenko

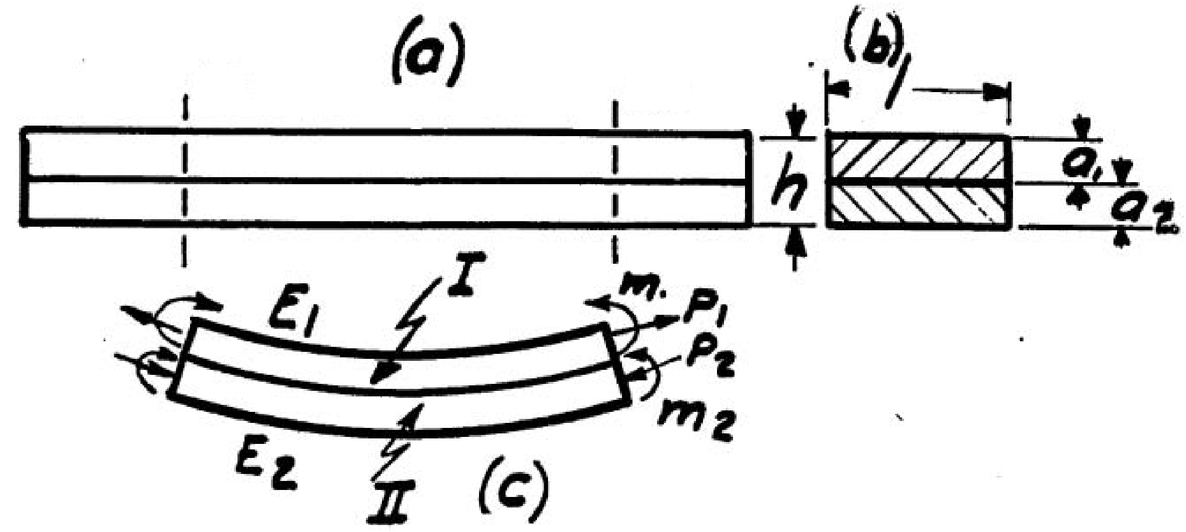


Fig. 1. *Deflection of a bi-metal strip while uniformly heated.*

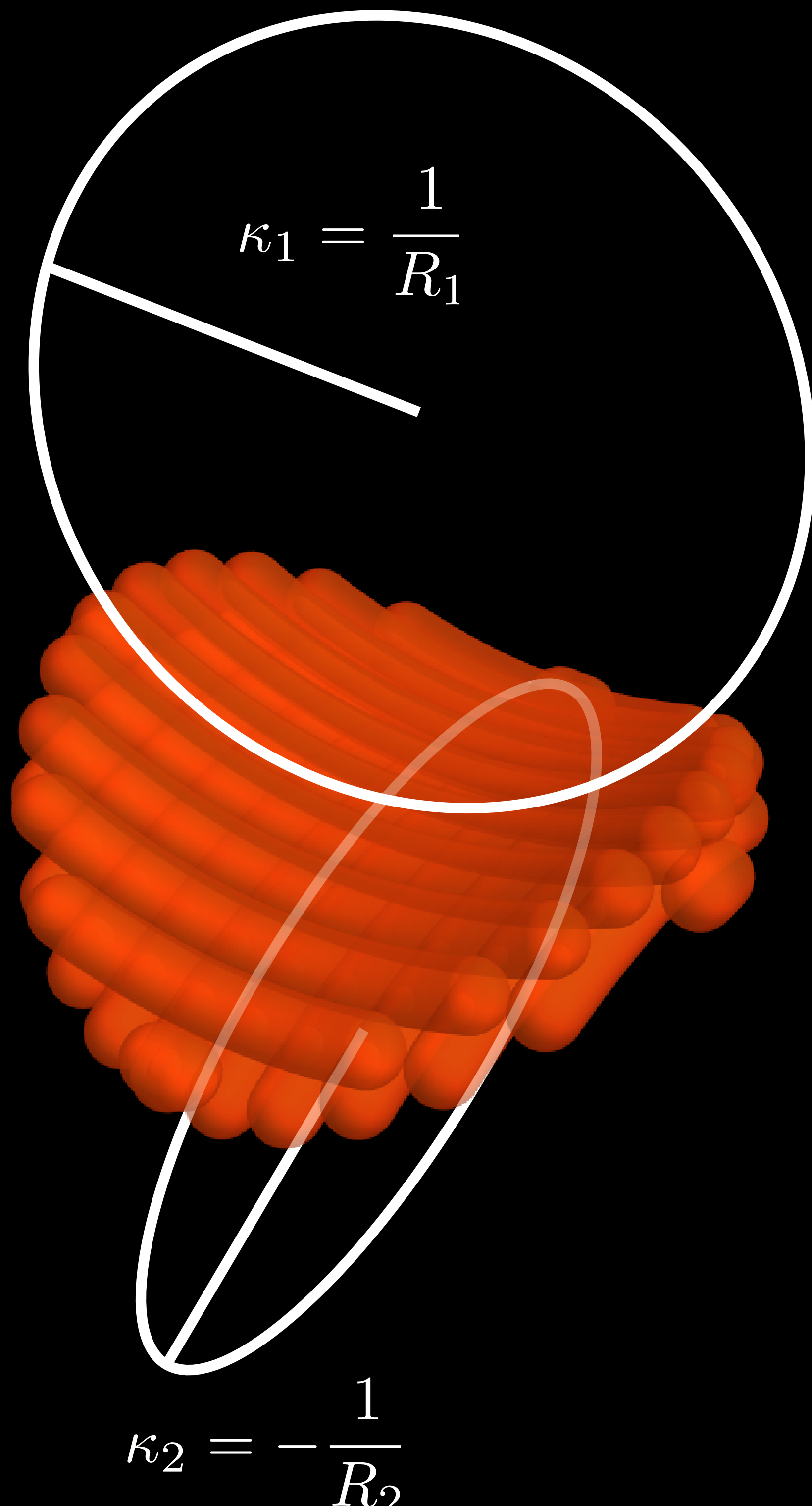
$$\frac{1}{a_1}\int_0^{a_1}\left(\varepsilon^{\mathrm{s}(1)} + \varepsilon^{\mathrm{e}(1)}(z) + \frac{\sigma^{\mathrm{tot}}}{\mathrm{E}_1}\right)dz = \frac{1}{a_2}\int_{-a_2}^{0}\left(\varepsilon^{\mathrm{s}(2)} + \varepsilon^{\mathrm{e}(2)}(z) + \frac{\sigma^{\mathrm{tot}}}{\mathrm{E}_2}\right)dz$$

$$\alpha_1 + \frac{a_1\kappa}{2} + \frac{1}{\mathrm{E}_1 a_1}\frac{M^{\mathrm{tot}}}{2h} = \alpha_2 - \frac{a_2\kappa}{2} - \frac{1}{\mathrm{E}_2 a_2}\frac{M^{\mathrm{tot}}}{2h}, \quad M^{\mathrm{tot}} = \frac{\kappa}{3}\left(\mathrm{E}_1 a_1^3 + \mathrm{E}_2 a_2^3\right)$$

$$\kappa = \frac{6(\alpha_2 - \alpha_1)(1+m)^2}{h\left(3(1+m)^2 + (1+mn)\left(m^2 + \frac{1}{mn}\right)\right)}, \quad m = \frac{a_1}{a_2},\ n = \frac{\mathrm{E}_1}{\mathrm{E}_2}$$

Timoshenko, *J. Opt. Soc. Am.* **11** 233 (1925)

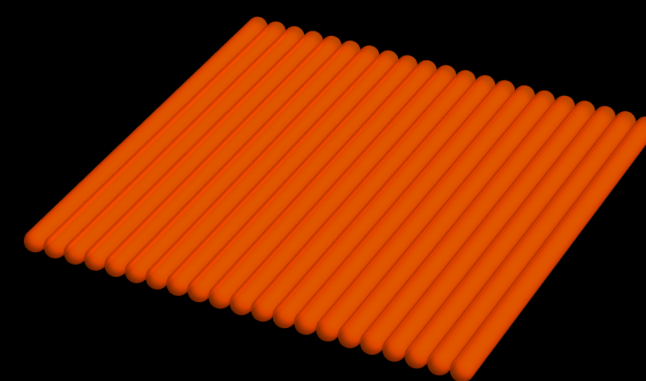
A Brief Primer on Curvature



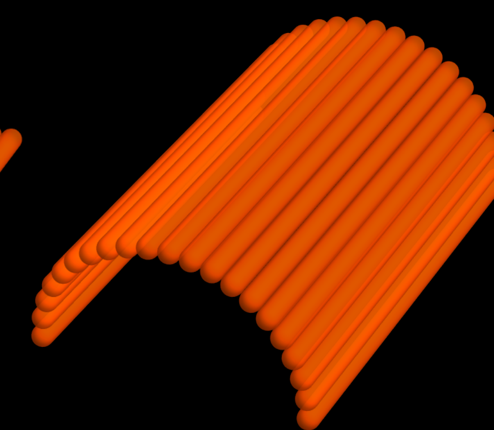
Mean (Extrinsic) Curvature: $H = \frac{1}{2}(\kappa_1 + \kappa_2)$

Bending energy

H=0



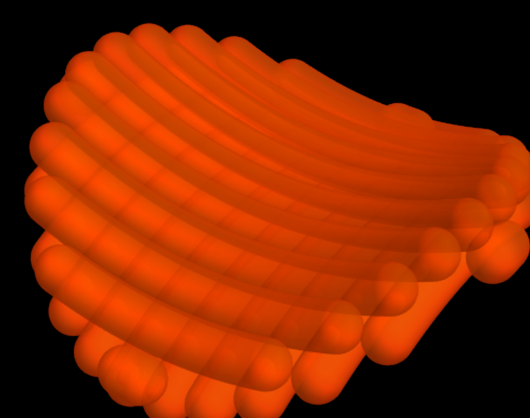
H<0



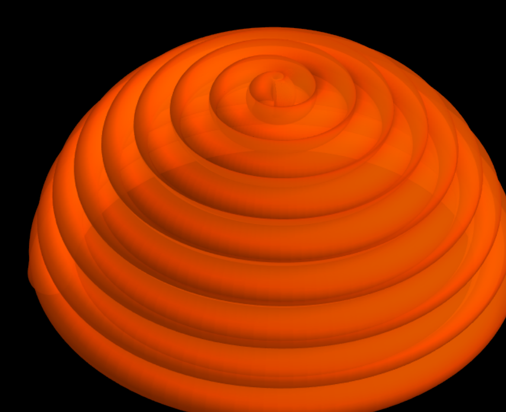
Gaussian (Intrinsic) Curvature: $K = \kappa_1 \kappa_2$

Stretching energy

K<0



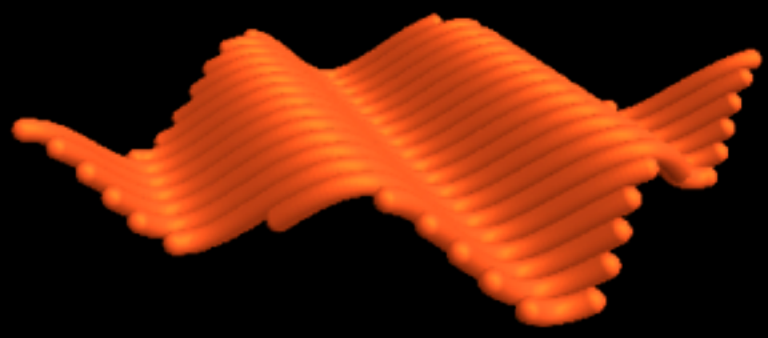
K>0



A Geometric Model

$$g(x,y) = R[\theta(x,y)] \begin{bmatrix} \alpha_\parallel & 0 \\ 0 & \alpha_\perp \end{bmatrix} R^T[\theta(x,y)]$$

Gauss's Theorema Egregium



GENERAL INVESTIGATIONS

OF

CURVED SURFACES

BY

KARL FRIEDRICH GAUSS

PRESENTED TO THE ROYAL SOCIETY, OCTOBER 8, 1827

Thus the formula of the preceding article leads of itself to the remarkable

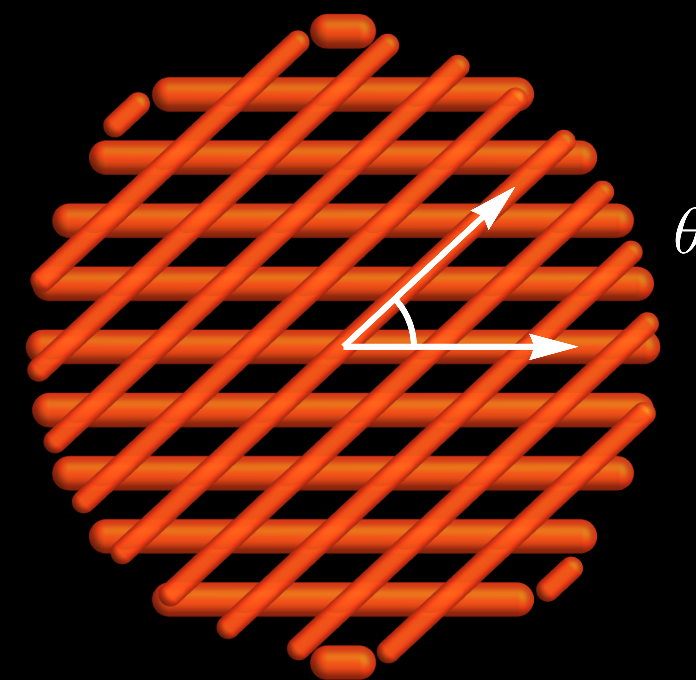
THEOREM. *If a curved surface is developed upon any other surface whatever, the measure of curvature in each point remains unchanged.*

$$K(x,y) = K\left(g, \partial_x g, \partial_y g, \partial_{xx} g, \partial_{xy} g, \partial_{yy} g\right)$$

$$K(x,y) = (\alpha_\parallel - \alpha_\perp)\left[\frac{(\phi^2-1)\phi_{xy} - \phi(\phi_{xx} - \phi_{vv})}{(\phi^2+1)^2} + \frac{(3\phi^2-1)(\phi_x^2 - \phi_y^2) - 2\phi(\phi^2-3)\phi_x\phi_y}{(\phi^2+1)^3}\right]$$

Aharoni, Sharon, Kupferman, *PRL* **113** 257801 (2014)

The Model



Theory of Anisotropic Plates and Shells

Curvature in Monge Gauge $\kappa_{ij} = \partial_i \partial_j H(x,y)$

Swelling Strain $\varepsilon^{\mathrm{s}} = \begin{bmatrix} \alpha_{\parallel} & 0 \\ 0 & \alpha_{\perp} \end{bmatrix}$

Elastic Strain $\varepsilon^{\mathrm{e}}_{ij} = -z\kappa_{ij}$

Strain Tensor $\varepsilon = \varepsilon^{\mathrm{s}} + \varepsilon^{\mathrm{e}}$ $\quad \varepsilon_{ij}(\theta) = R_{im}(\theta)\varepsilon_{mn}R^T_{jn}(\theta)$

Elastic Modulus Tensor $\mathrm{E}_{ijkl}(\theta) = R_{im}(\theta)R_{kp}(\theta)\mathrm{E}_{mnpq}R^T_{jn}(\theta)R^T_{lq}(\theta)$

Stress-Strain Relation $\sigma_{ij} = \mathrm{E}_{ijkl}\varepsilon^{\mathrm{e}}_{kl}$

Bending Moments
$$M_{ij} = \int_{-a_2}^{a_1} z\sigma_{ij}dz = -\int_{-a_2}^{a_1} z^2\mathrm{E}_{ijkl}\kappa_{kl}dz$$
$$= -\int_0^{a_1}\mathrm{E}_{ijkl}(0)\kappa_{kl}z^2dz - \int_{-a_2}^{0}\mathrm{E}_{ijkl}(\theta)\kappa_{kl}z^2dz$$

1. Equilibrium Condition

"Due to the fact that there are not external forces acting on the strip, all forces acting over any cross-section of the strip must be in equilibrium"

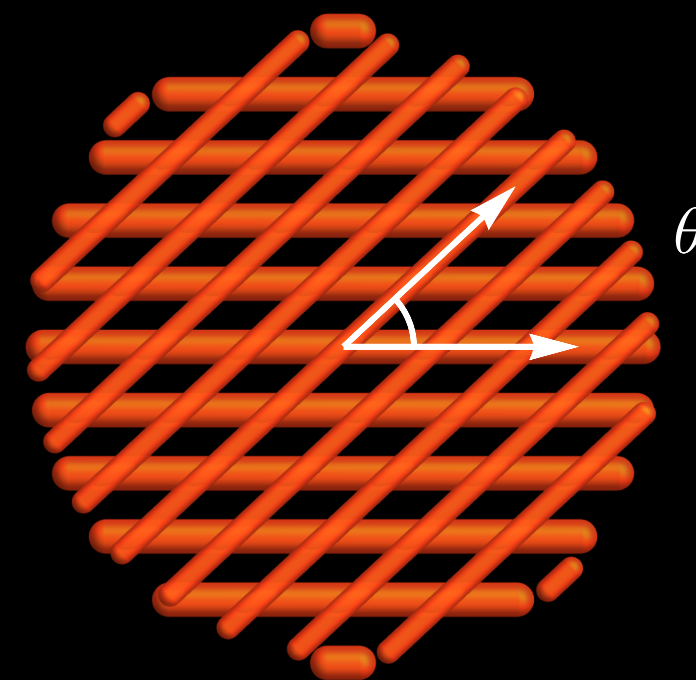
2. Compatibility Condition

"On the bearing surface of both metals the unit elongation occurring in the longitudinal fibres of metals (1) and (2) must be equal."

A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

The Model

Theory of Anisotropic Plates and Shells



Curvature in Monge Gauge $\kappa_{ij} = \partial_i \partial_j H(x, y)$

Swelling Strain $\varepsilon^{\mathrm{s}} = \begin{bmatrix} \alpha_{\parallel} & 0 \\ 0 & \alpha_{\perp} \end{bmatrix}$

Elastic Strain $\varepsilon^{\mathrm{e}}_{ij} = -z\kappa_{ij}$

Strain Tensor $\varepsilon = \varepsilon^{\mathrm{s}} + \varepsilon^{\mathrm{e}}$ $\quad \varepsilon_{ij}(\theta) = R_{im}(\theta)\varepsilon_{mn}R^T_{jn}(\theta)$

Elastic Modulus Tensor $\mathrm{E}_{ijkl}(\theta) = R_{im}(\theta)R_{kp}(\theta)\mathrm{E}_{mnpq}R^T_{jn}(\theta)R^T_{lq}(\theta)$

Stress-Strain Relation $\sigma_{ij} = \mathrm{E}_{ijkl}\varepsilon^{\mathrm{e}}_{kl}$

Bending Moments

$$M_{ij} = \int_{-a_2}^{a_1} z\sigma_{ij}dz = -\int_{-a_2}^{a_1} z^2 \mathrm{E}_{ijkl}\kappa_{kl}dz$$

$$= -\int_0^{a_1} \mathrm{E}_{ijkl}(0)\kappa_{kl}z^2 dz - \int_{-a_2}^{0} \mathrm{E}_{ijkl}(\theta)\kappa_{kl}z^2 dz$$

$$\frac{1}{\alpha_1}\int_0^{a_1}\left(\varepsilon^{(1)}_{ij} + \frac{\mathrm{E}^{-1}_{ijkl}}{a_1}M_{kl}(\theta)\right)dz = \frac{1}{\alpha_2}\int_{-a_2}^{0}\left(\varepsilon^{(2)}_{ij}(\theta) + \frac{\mathrm{E}^{-1}_{ijkl}(\theta)}{a_2}M_{kl}(\theta)\right)dz$$

A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

The Model

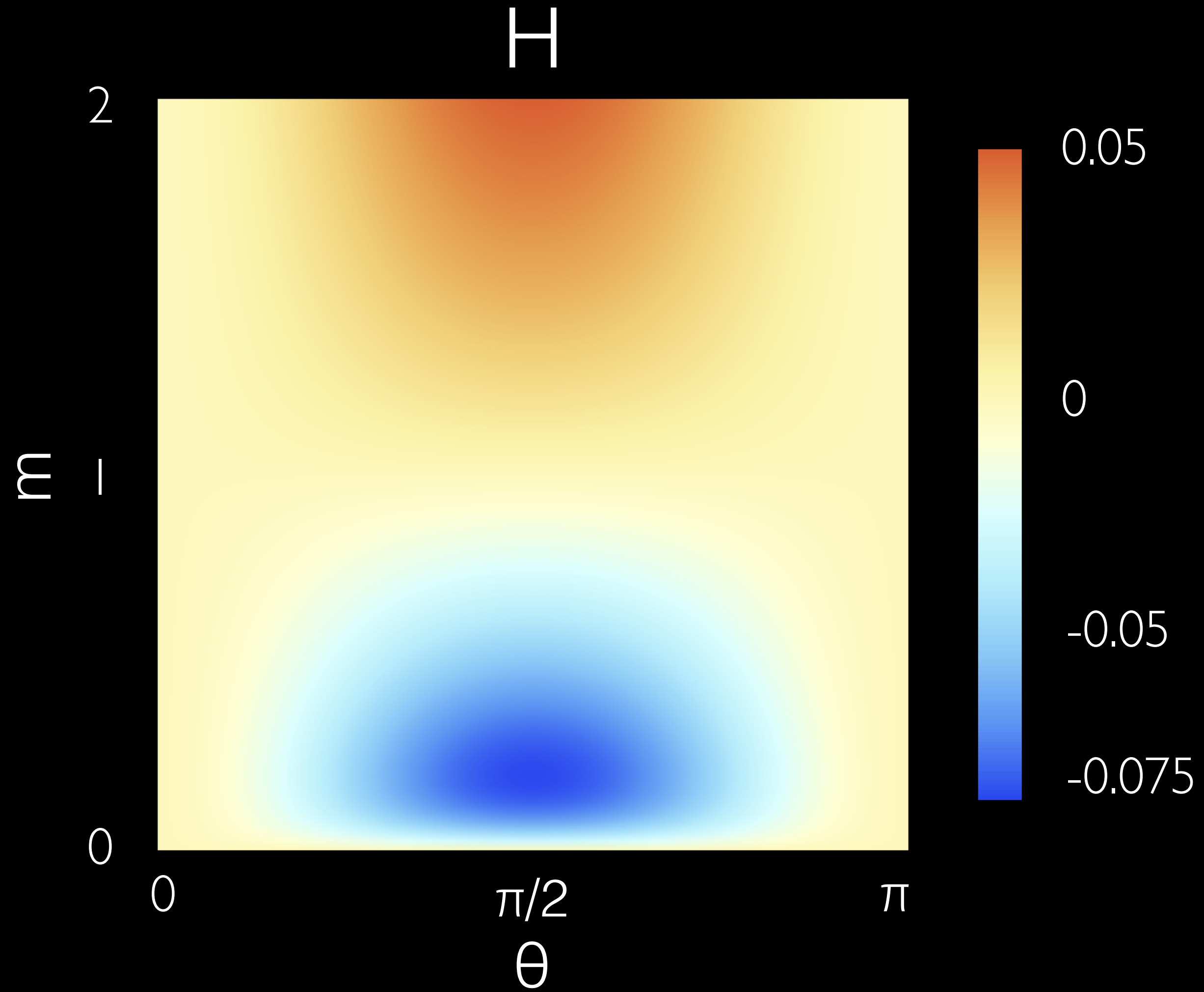
$$\frac{1}{\alpha_1}\int_0^{a_1}\left(\varepsilon_{ij}^{(1)} + \frac{\mathrm{E}_{ijkl}^{-1}}{a_1}M_{kl}(\theta)\right)dz = \frac{1}{\alpha_2}\int_{-a_2}^{0}\left(\varepsilon_{ij}^{(2)}(\theta) + \frac{\mathrm{E}_{ijkl}^{-1}(\theta)}{a_2}M_{kl}(\theta)\right)dz$$

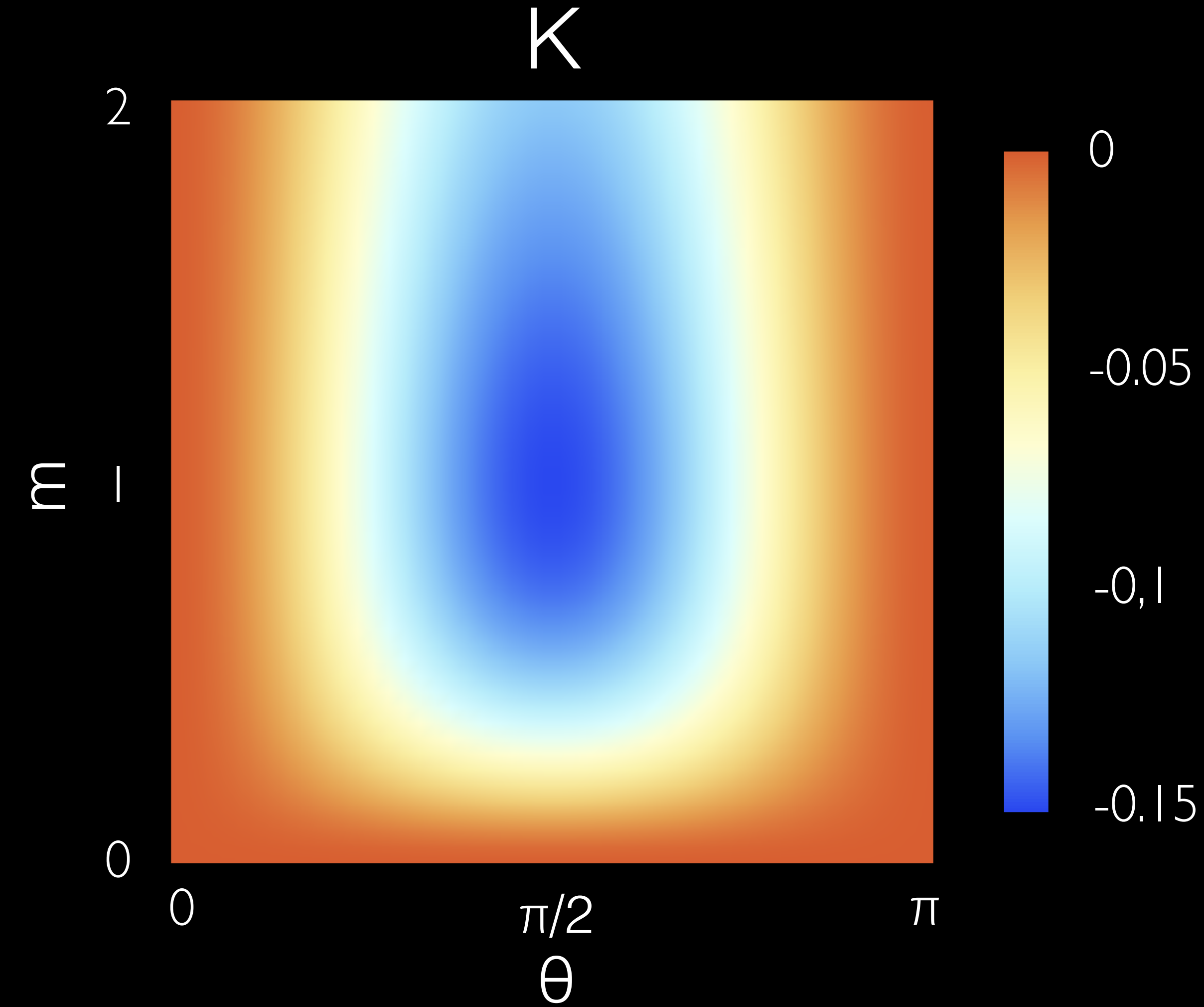
Given: $\alpha_\parallel, \alpha_\perp, \mathrm{E}_{ijkl}, a_1, a_2, \theta$ Solve for: κ_{ij}

$$H = \frac{\alpha_\perp - \alpha_\parallel}{h}\frac{c_1\sin^2(\theta)}{c_2 - c_3\cos(2\theta) + m^4\cos(4\theta)}$$

$$K = -\frac{(\alpha_\perp - \alpha_\parallel)^2}{h^2}\frac{c_4\sin^2(\theta)}{c_5 - c_6\cos(2\theta) + m^4\cos(4\theta)}$$

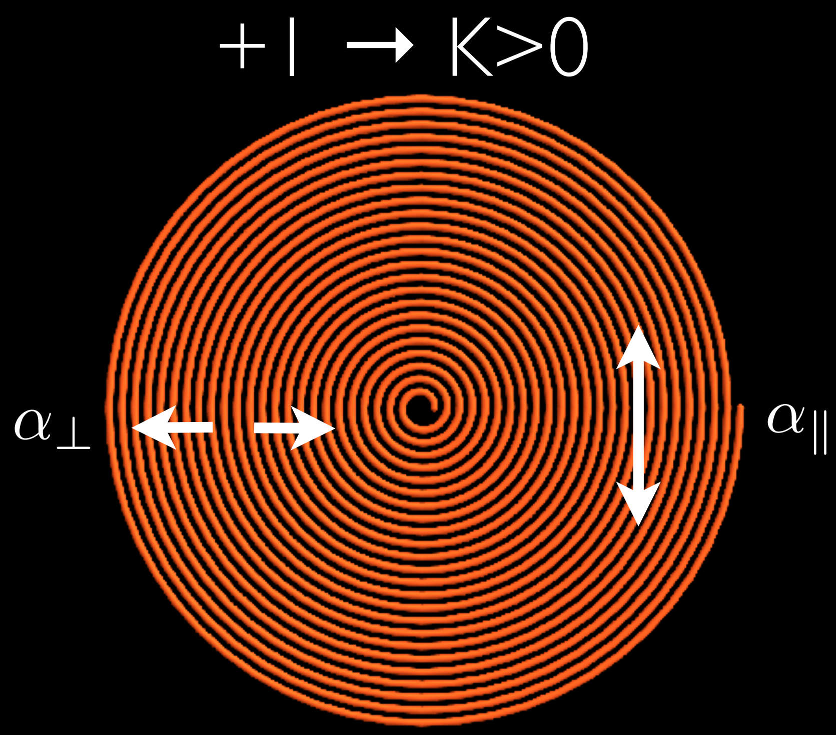
$$, \quad c_i = c_i(\mathrm{E}^{(1)}, \mathrm{E}^{(2)}, \mathrm{m} = \mathrm{a}_1/\mathrm{a}_2)$$

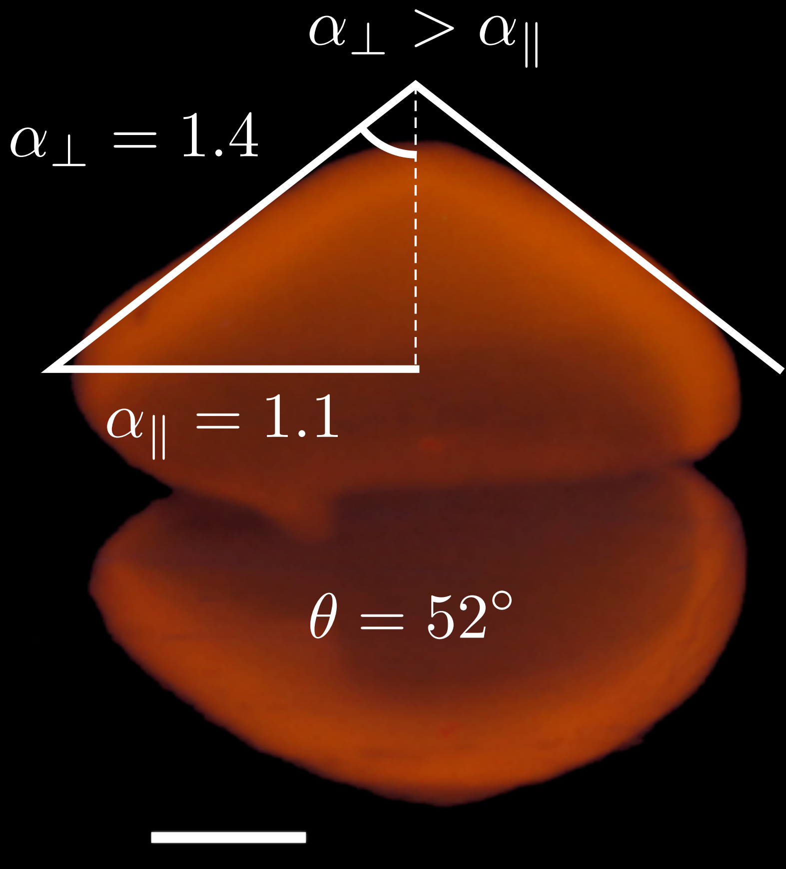




A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

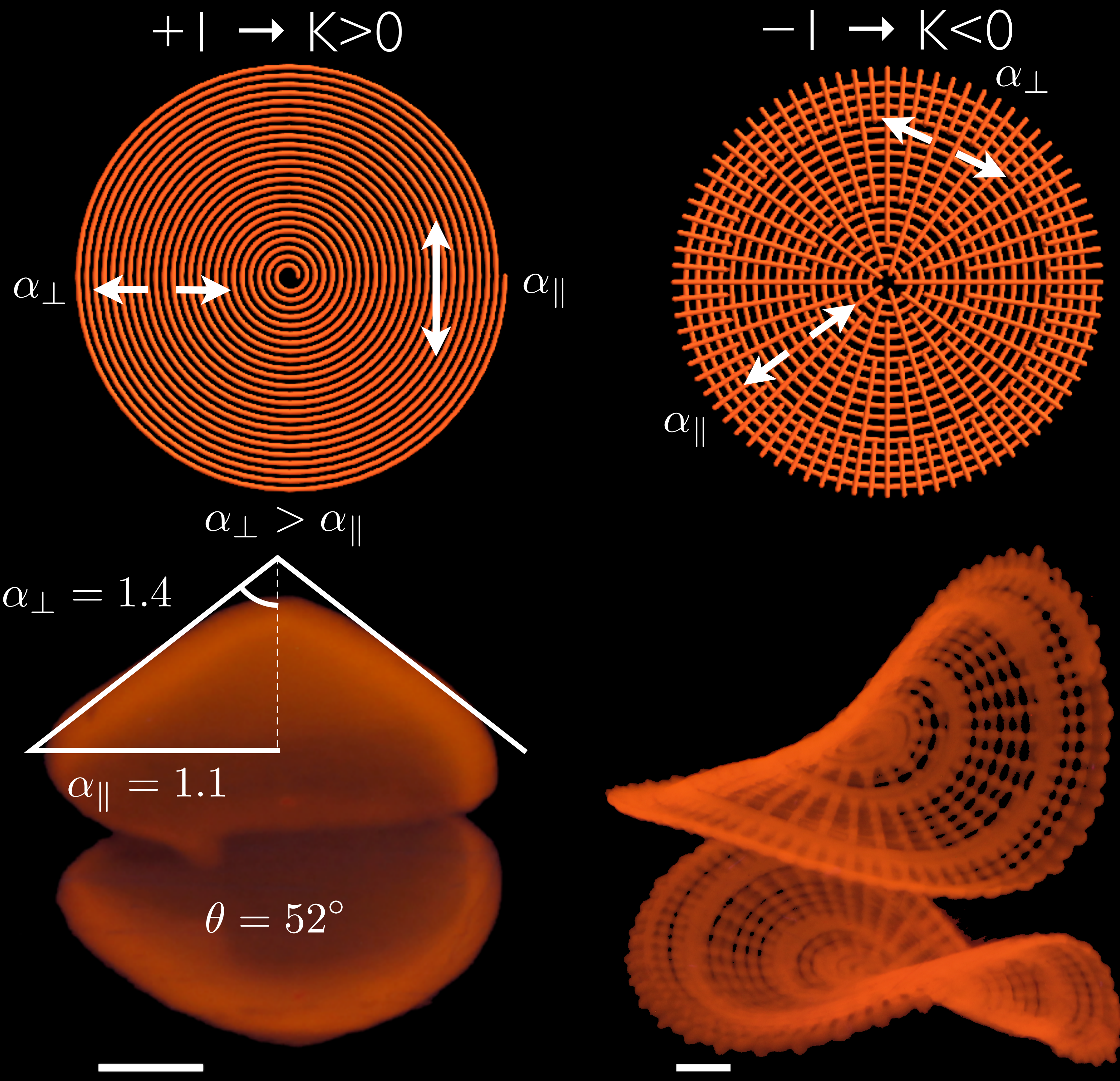
Disclinations and Gaussian Curvature





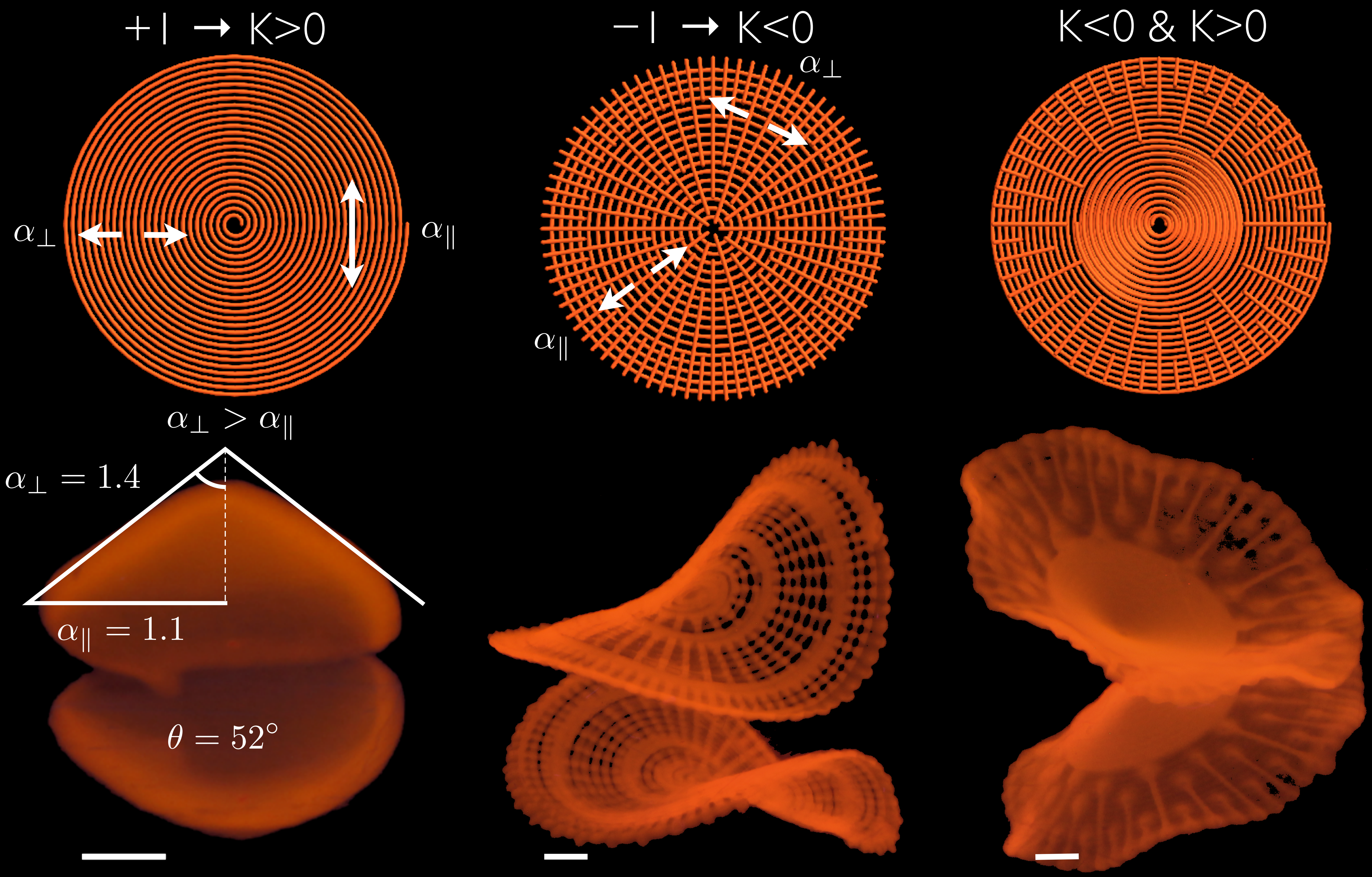
A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

Disclinations and Gaussian Curvature



A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

Disclinations and Gaussian Curvature

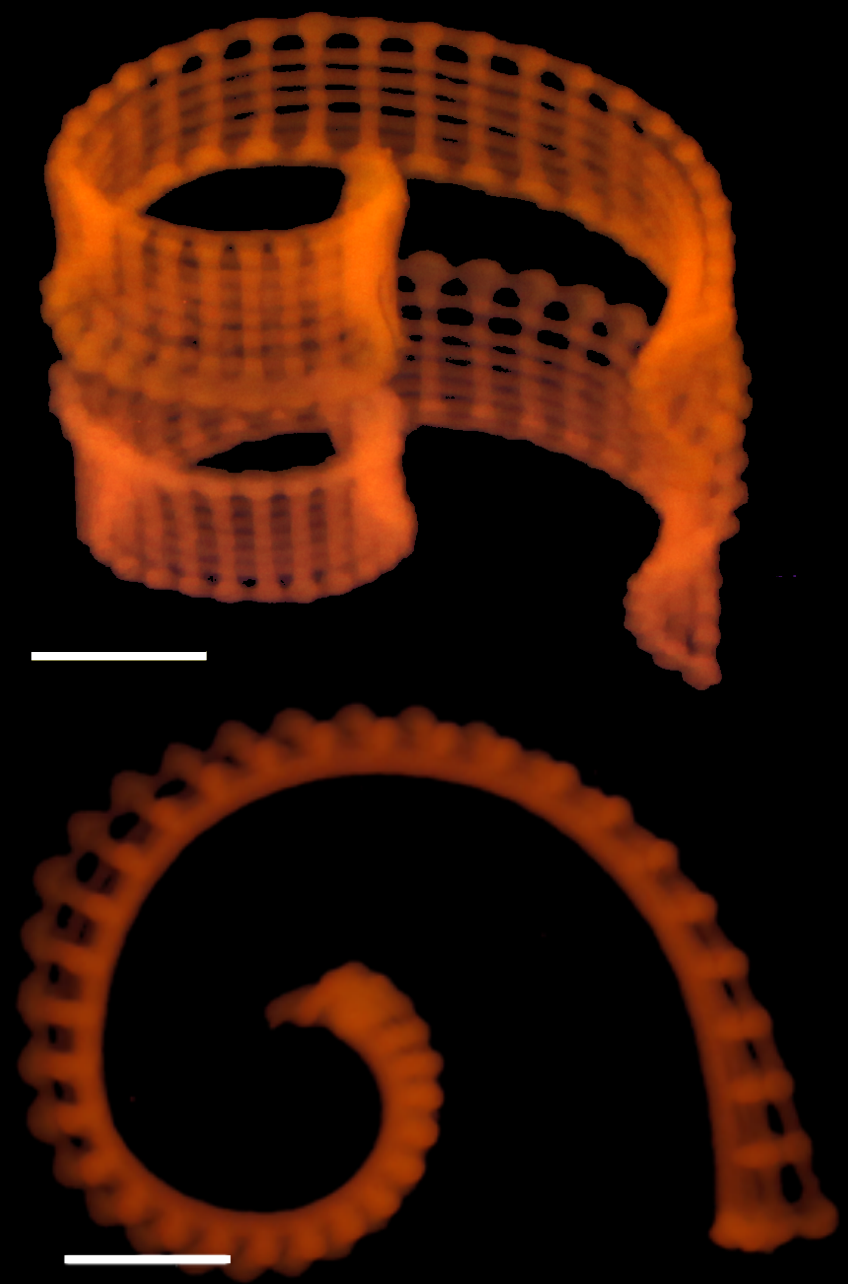


A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

Controlling Mean Curvature

Bilayers control the **sign** of mean curvature

Thickness controls its **magnitude**

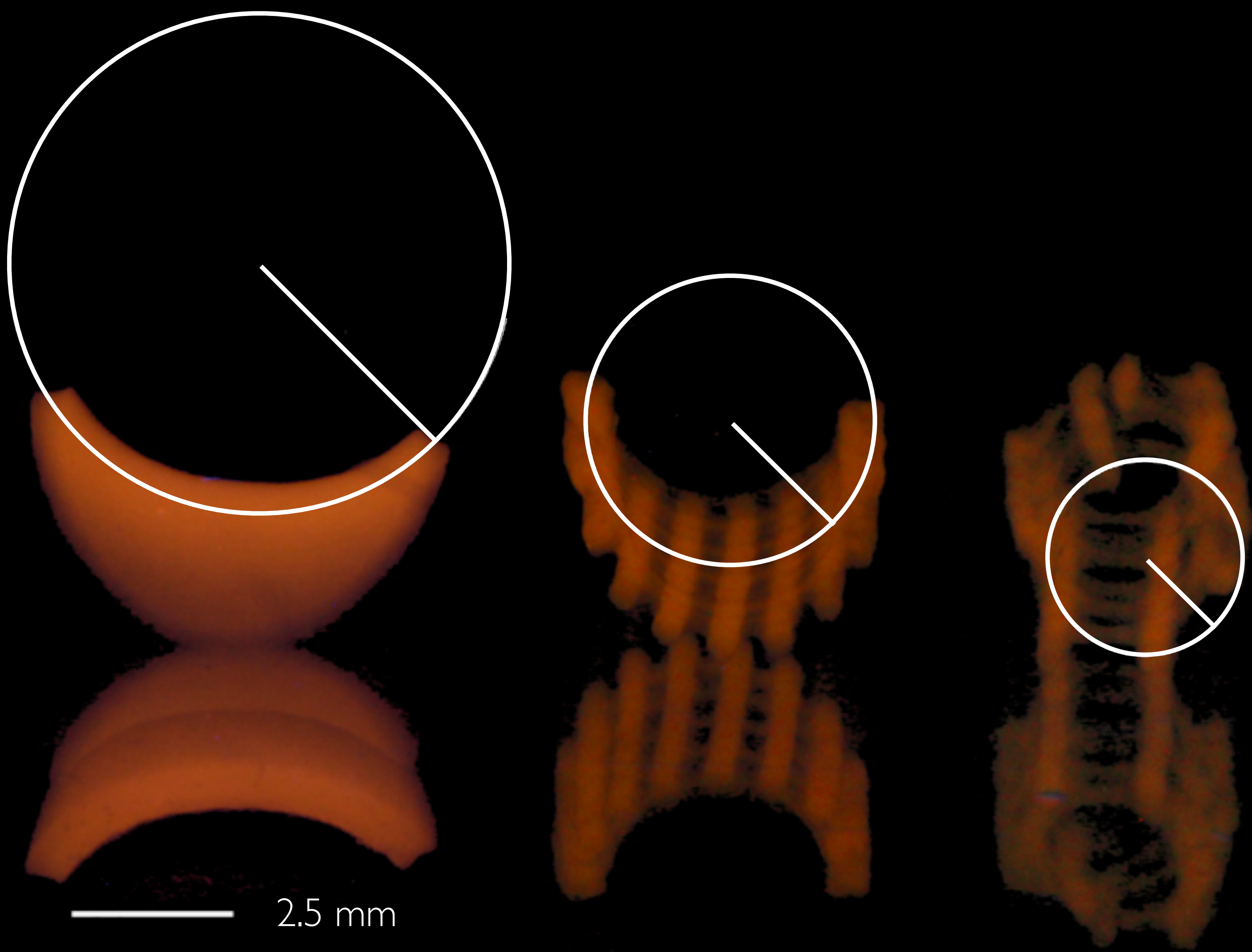


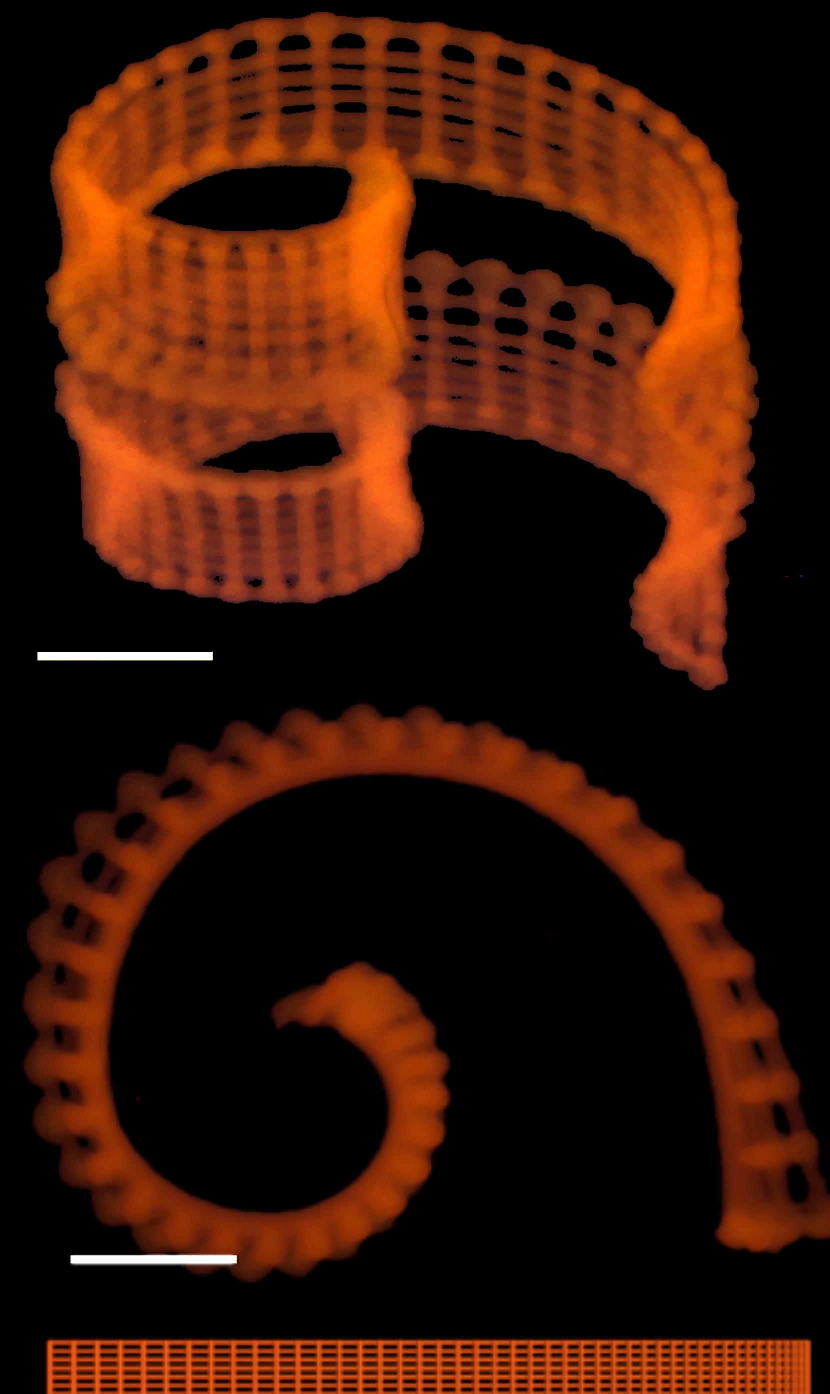
A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

Controlling Mean Curvature

$\kappa = 3/2(\alpha_1-\alpha_2)/h = 0.45/h \text{ mm}^{-1}$

h = 1.25 mm | h = 0.75 mm | h = 0.5 mm





predicted

$\kappa = 0.36 \text{ mm}^{-1}$ | $\kappa = 0.6 \text{ mm}^{-1}$ | $\kappa = 0.9 \text{ mm}^{-1}$

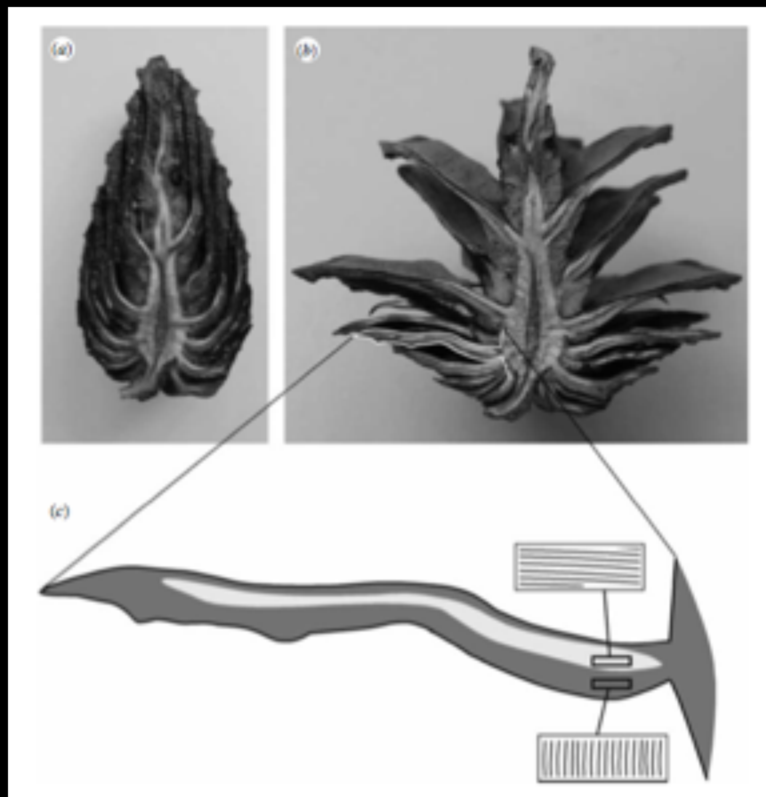
measured

$\kappa = 0.34 \text{ mm}^{-1}$ | $\kappa = 0.61 \text{ mm}^{-1}$ | $\kappa = 0.85 \text{ mm}^{-1}$

A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

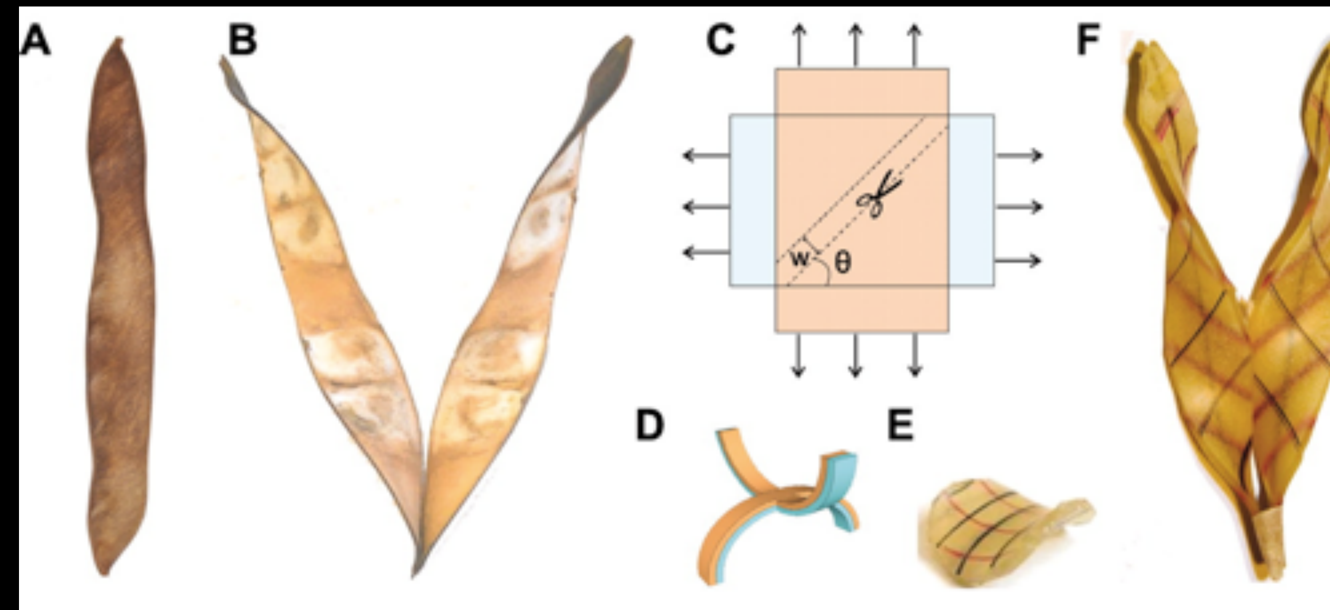
Orthogonal Bilayers: Two Morphologies

Pine Cone



Burgert & Fratzl, *Phil. Trans. R. Soc. A* **367** 1541 (2009)

Bauhinia Seed Pod



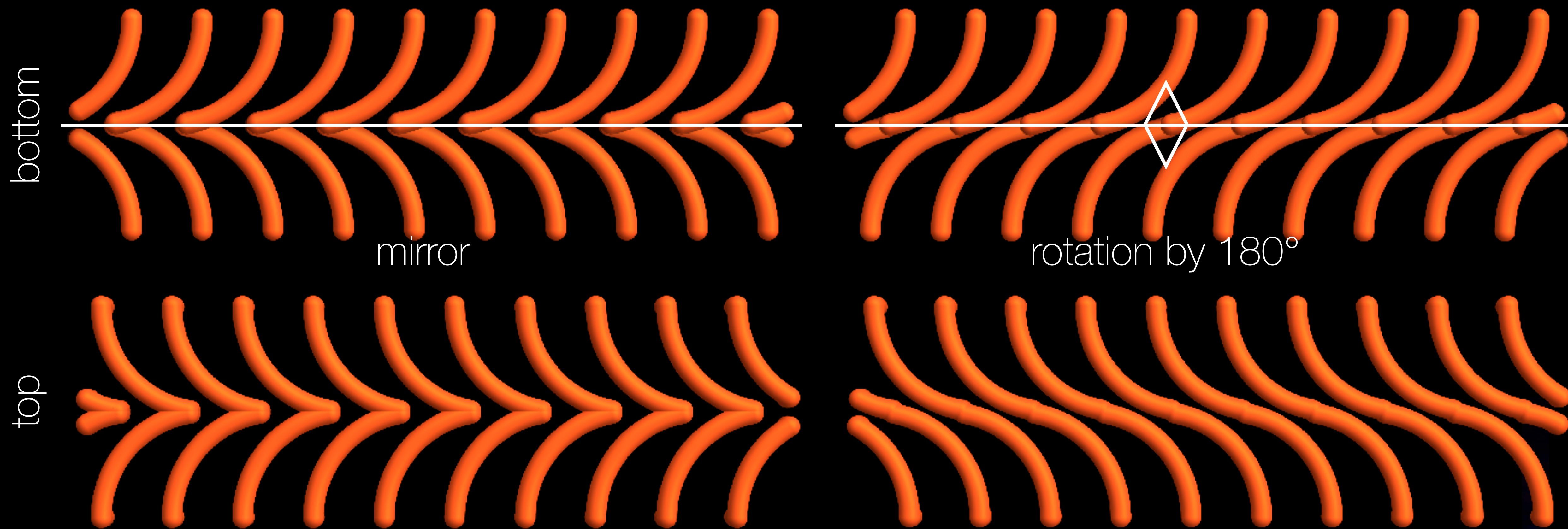
S. Armon, E. Efrati, R. Kupferman, E. Sharon, **333** 1726 Science (2011)

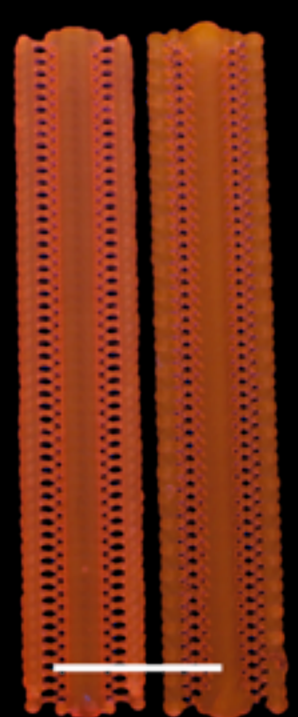
Bottom Layer: 0°
Top Layer: 90°

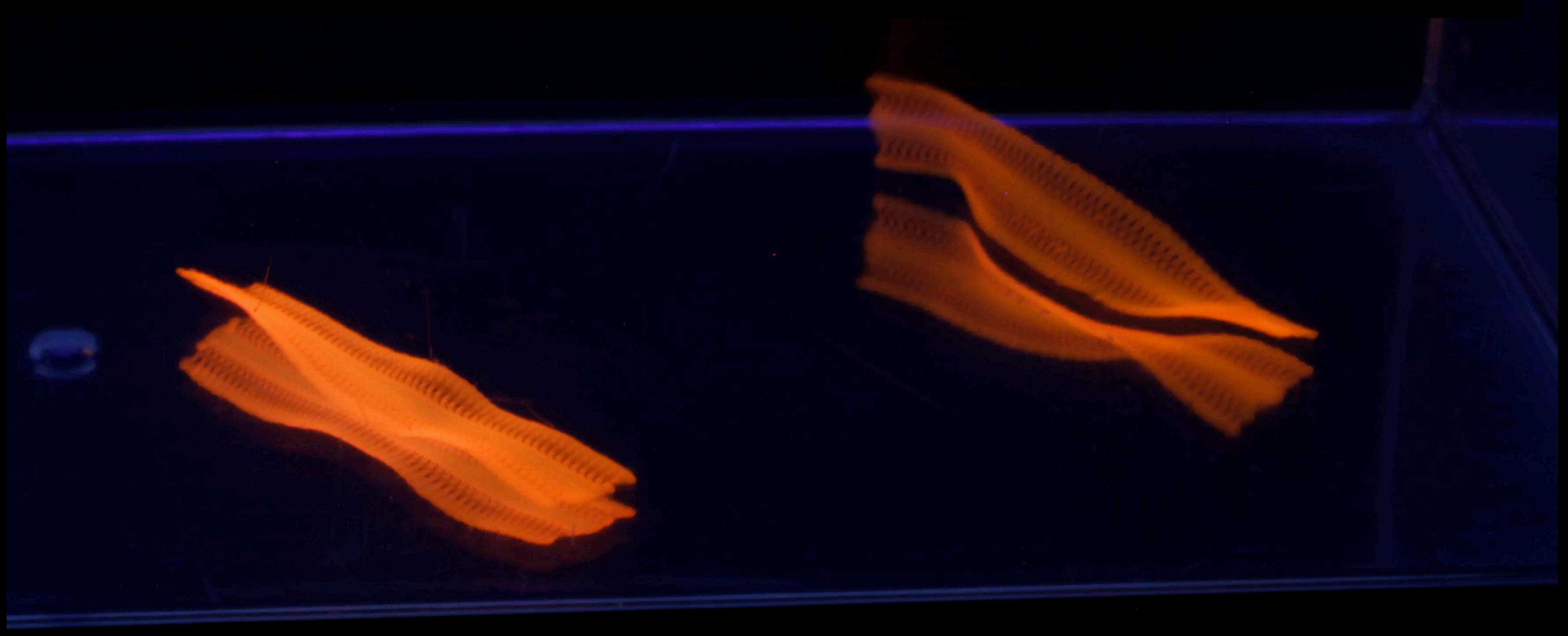
Bottom Layer: -45°
Top Layer: 45°

A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

To Twist or Not To Twist, That is the Question

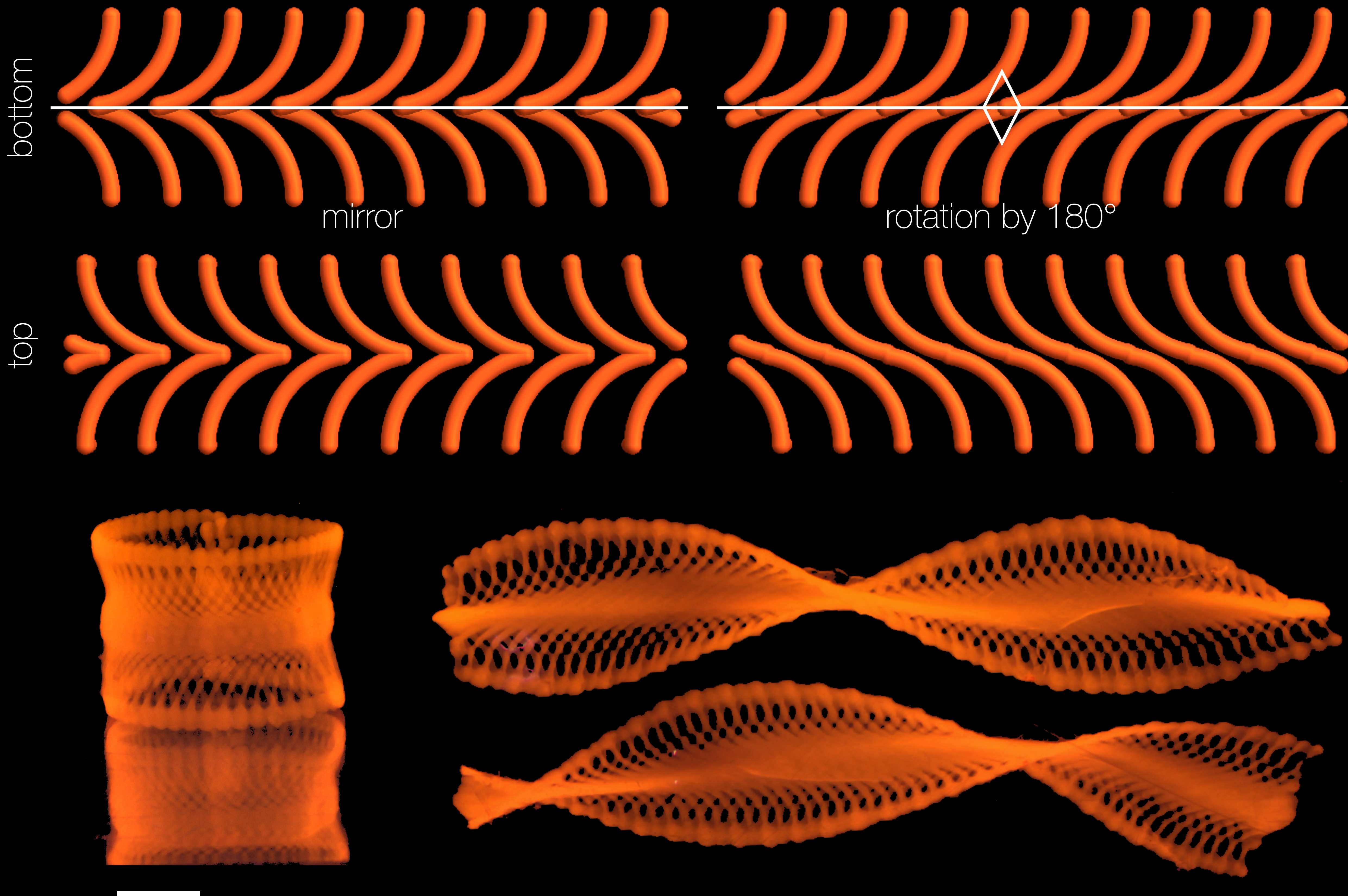






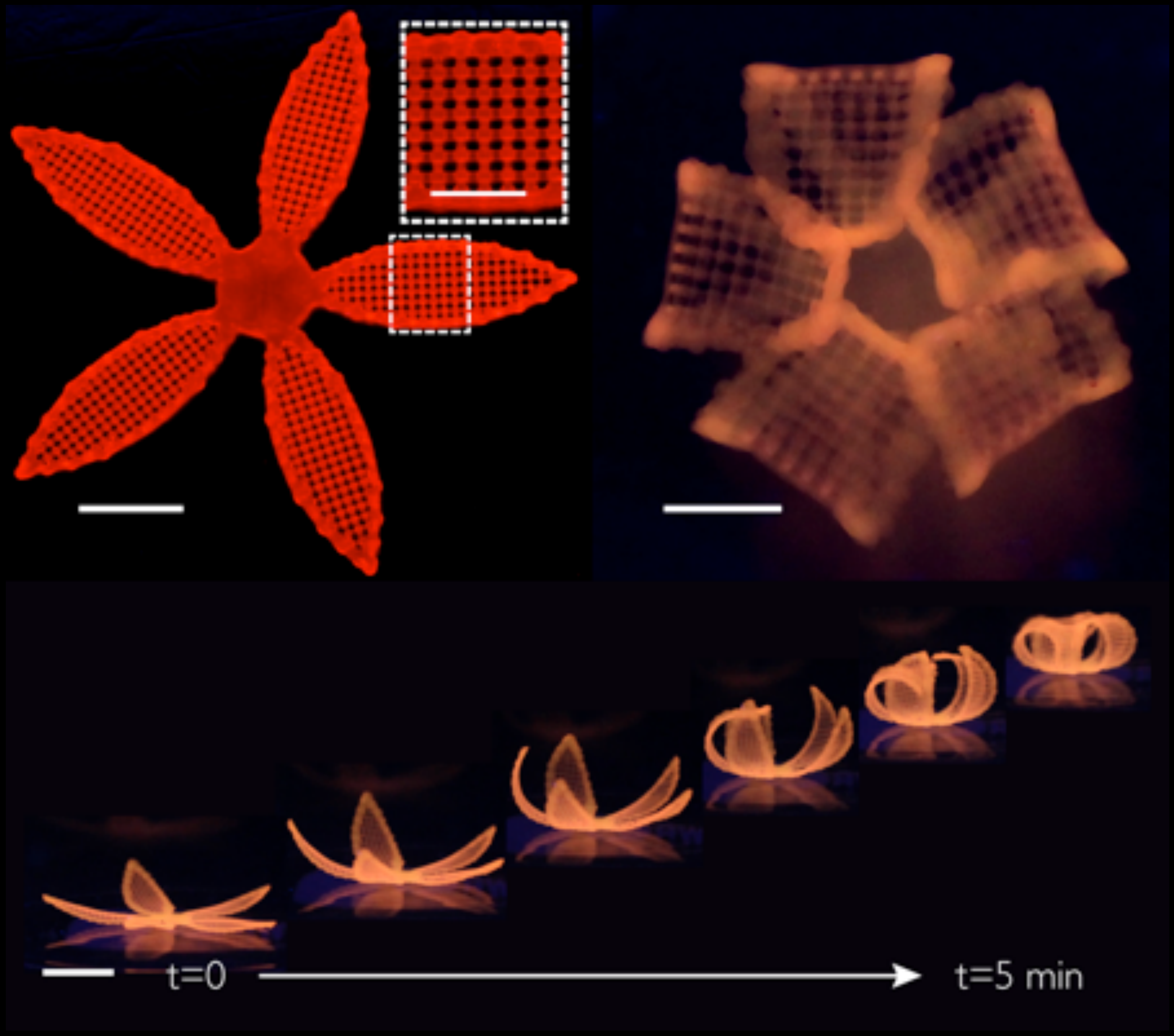
A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

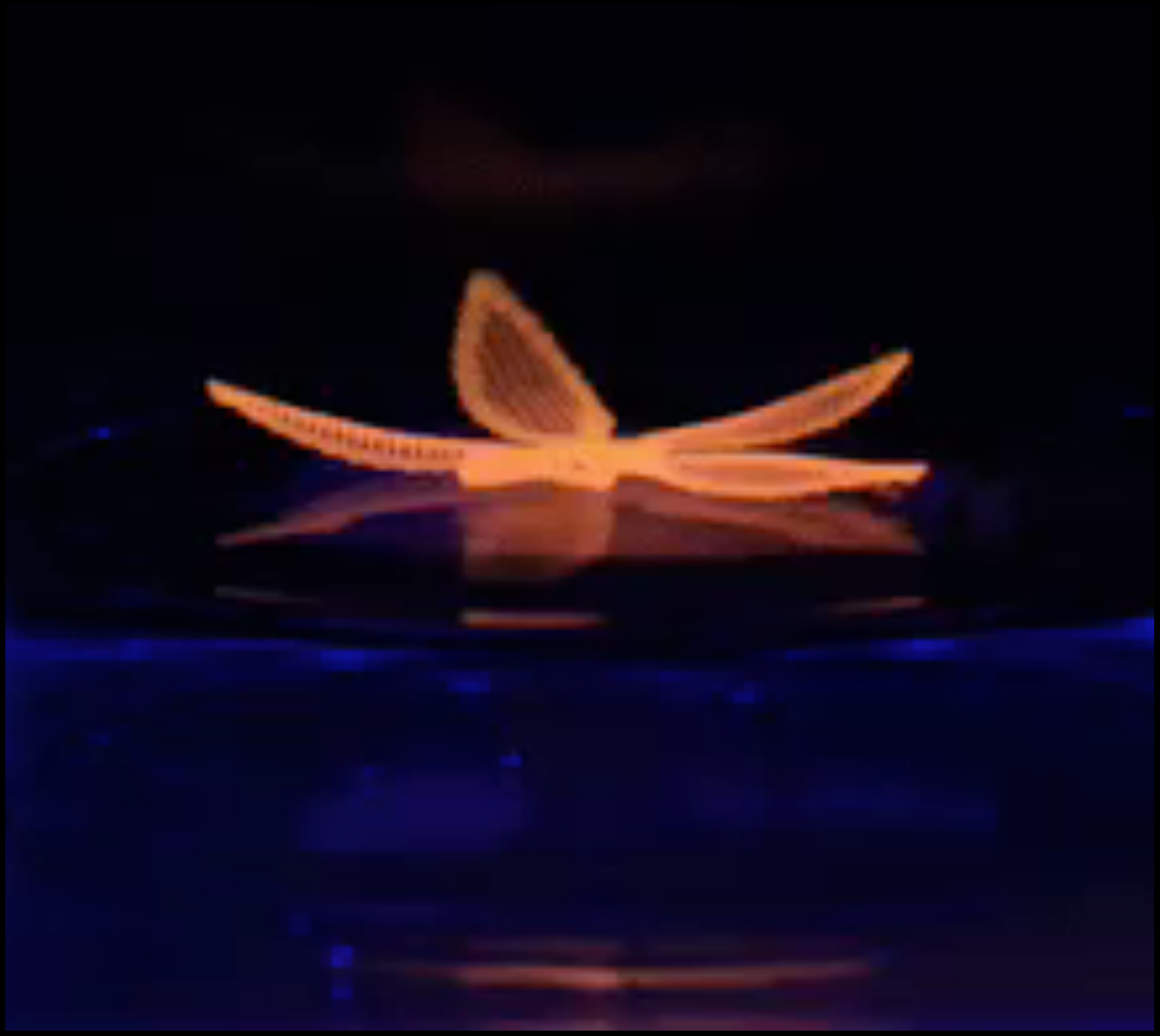
To Twist or Not To Twist, That is the Question



A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

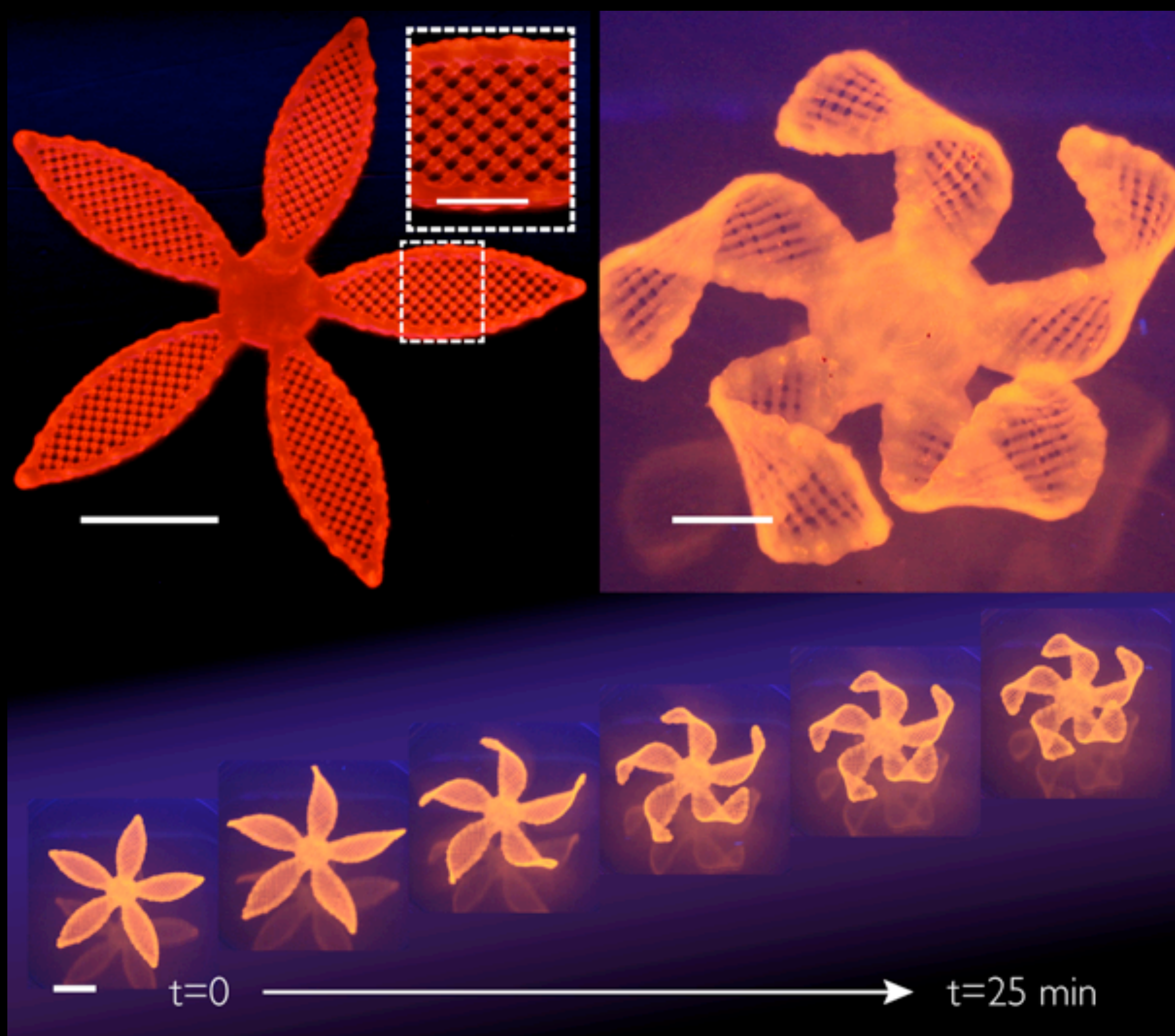
Forty 4D Folding Flowers





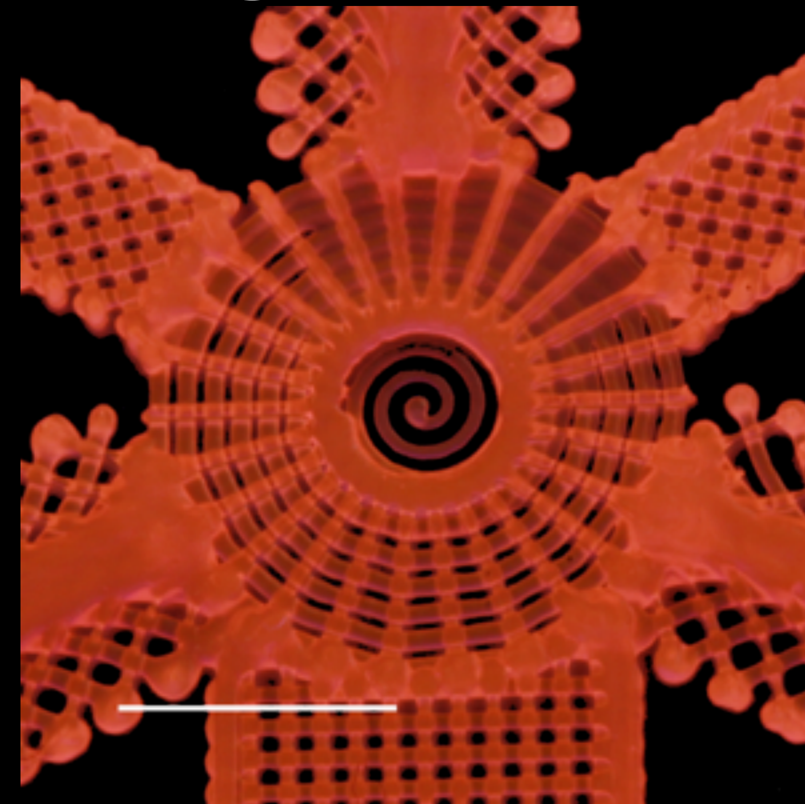
A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

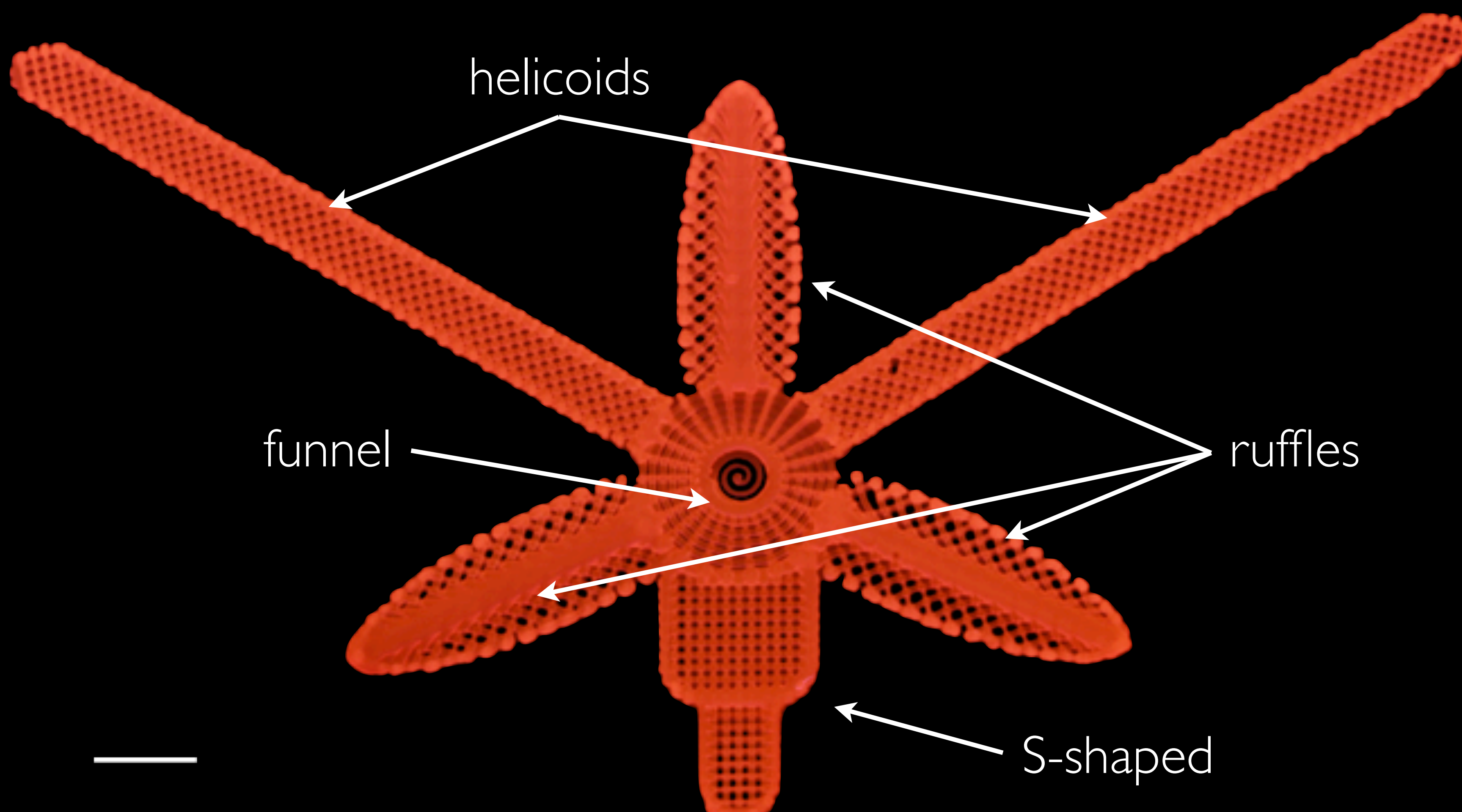
Forty 4D Folding Flowers



A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

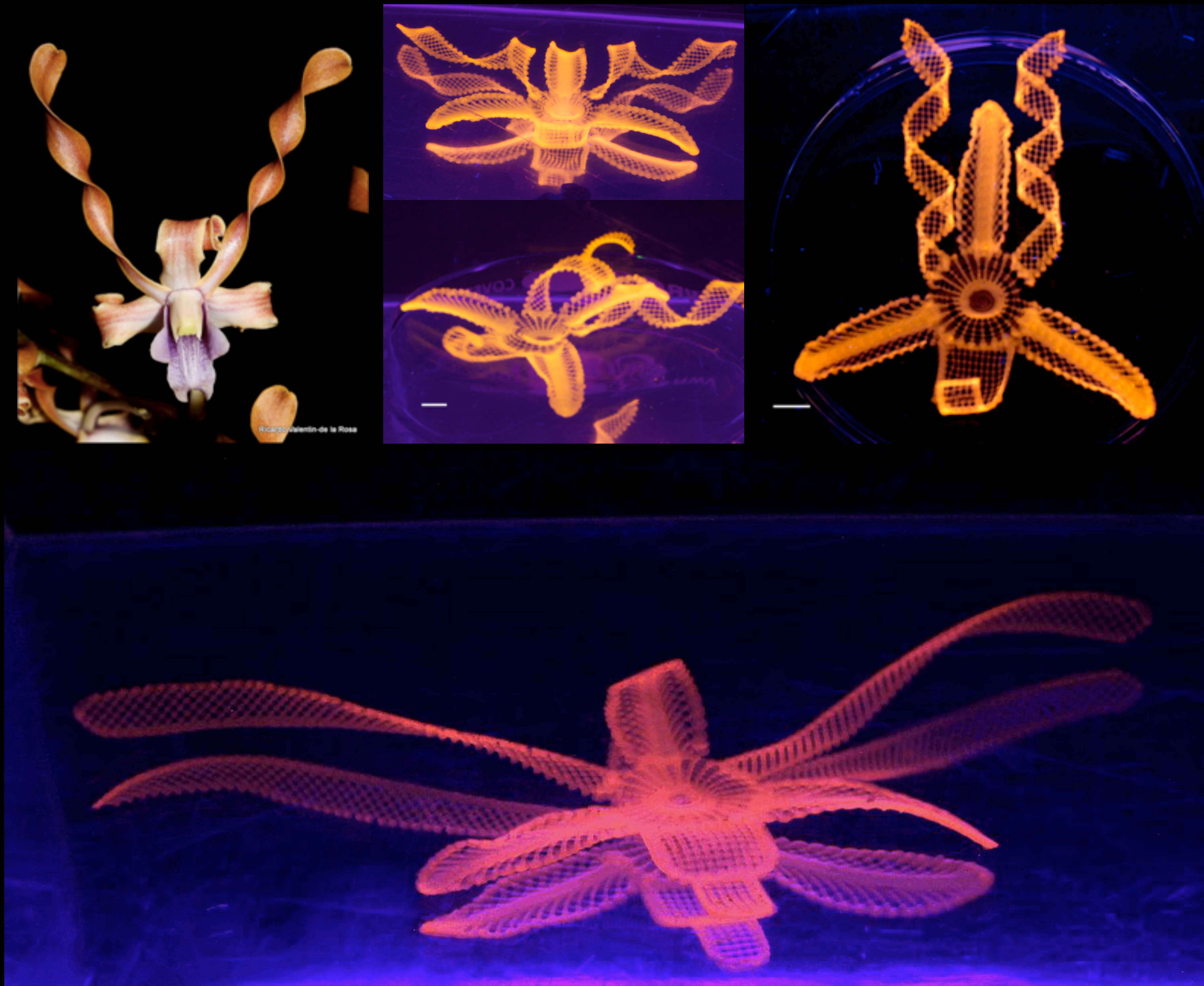
Forty 4D Folding Flowers





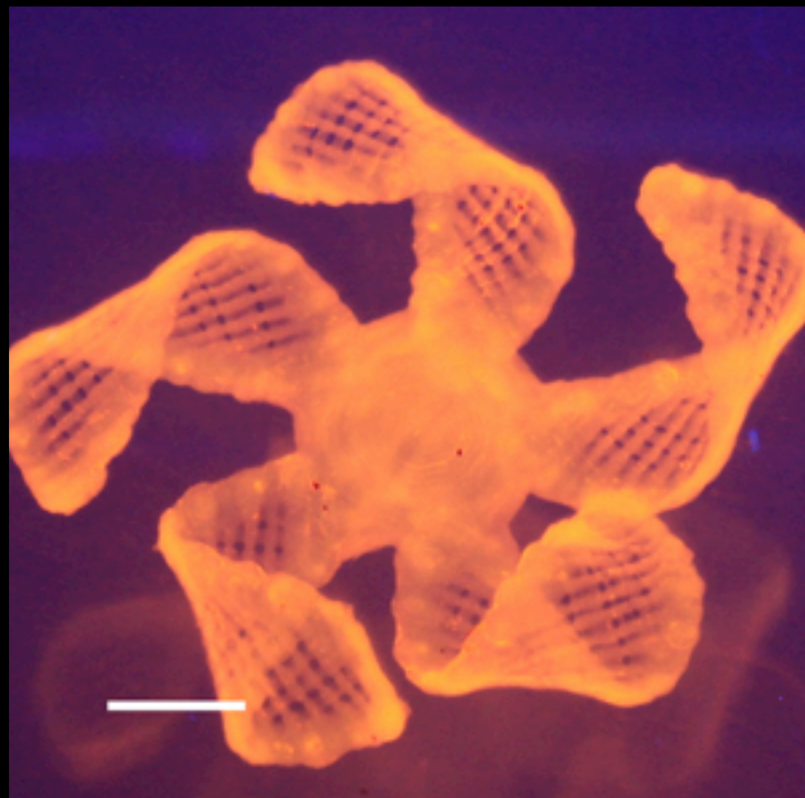
A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

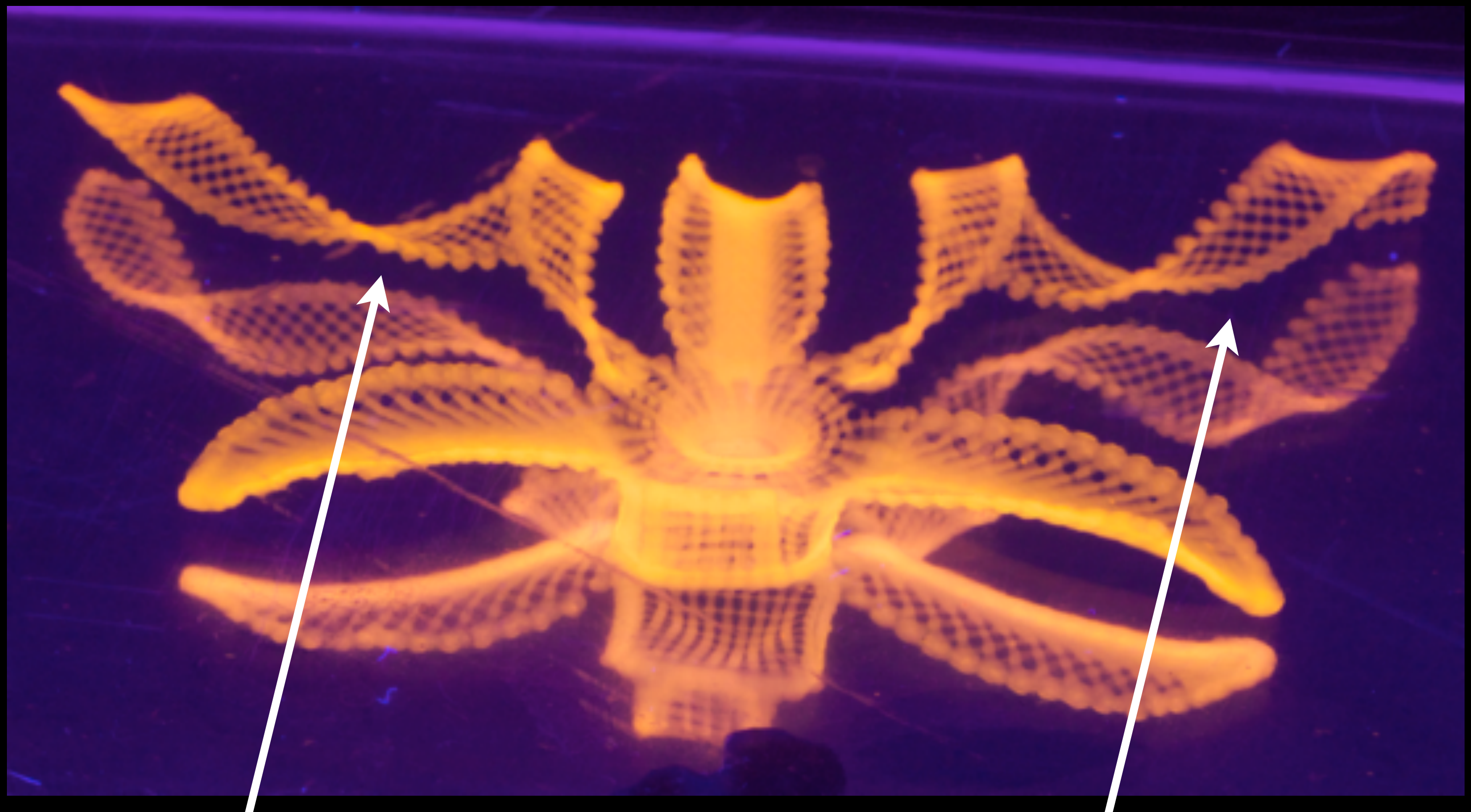
Forty 4D Folding Flowers



A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

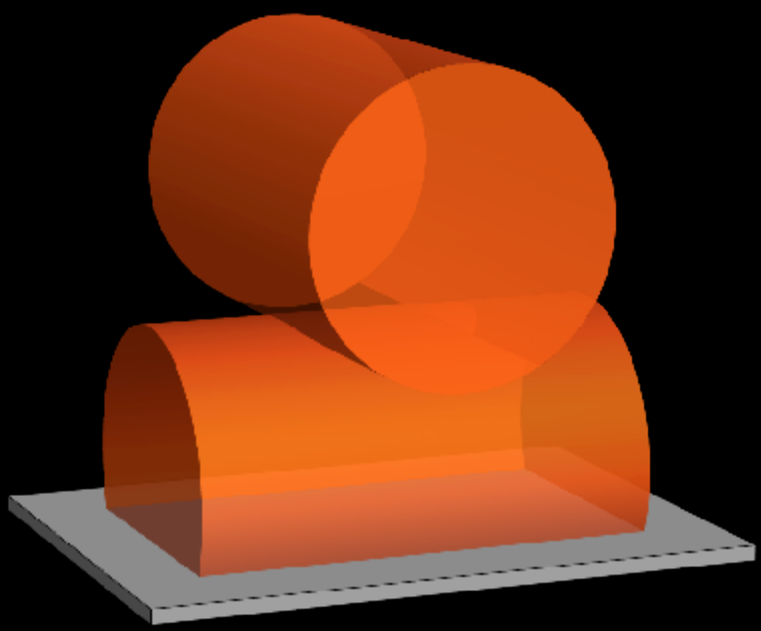
Left-handed or Right-handed?

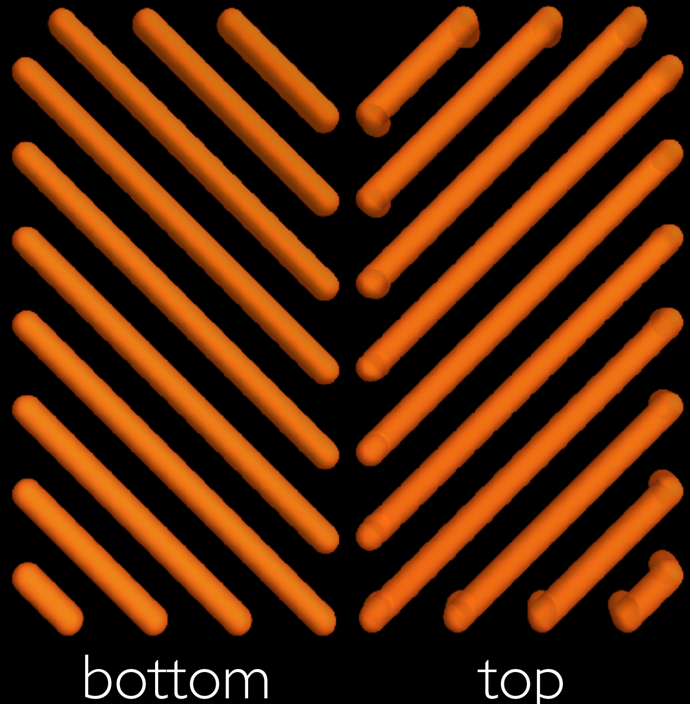


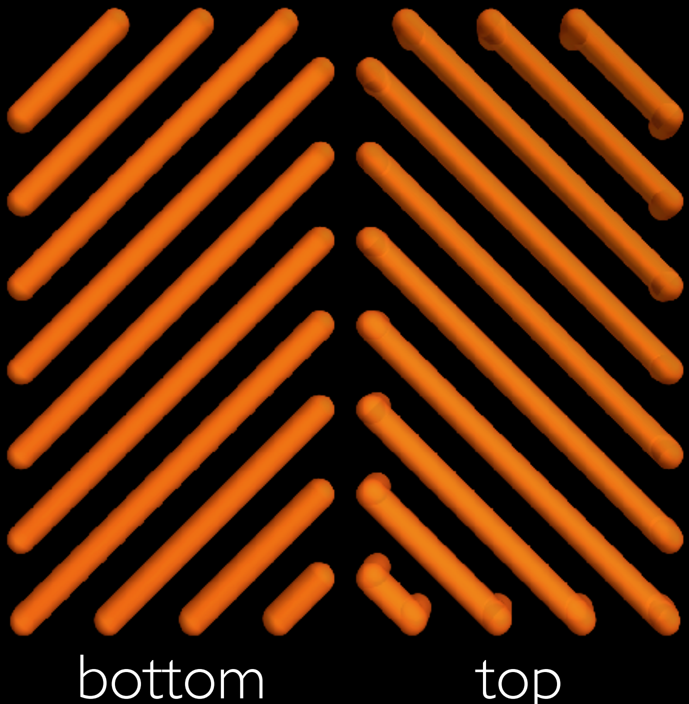


Left-handed

Right-handed



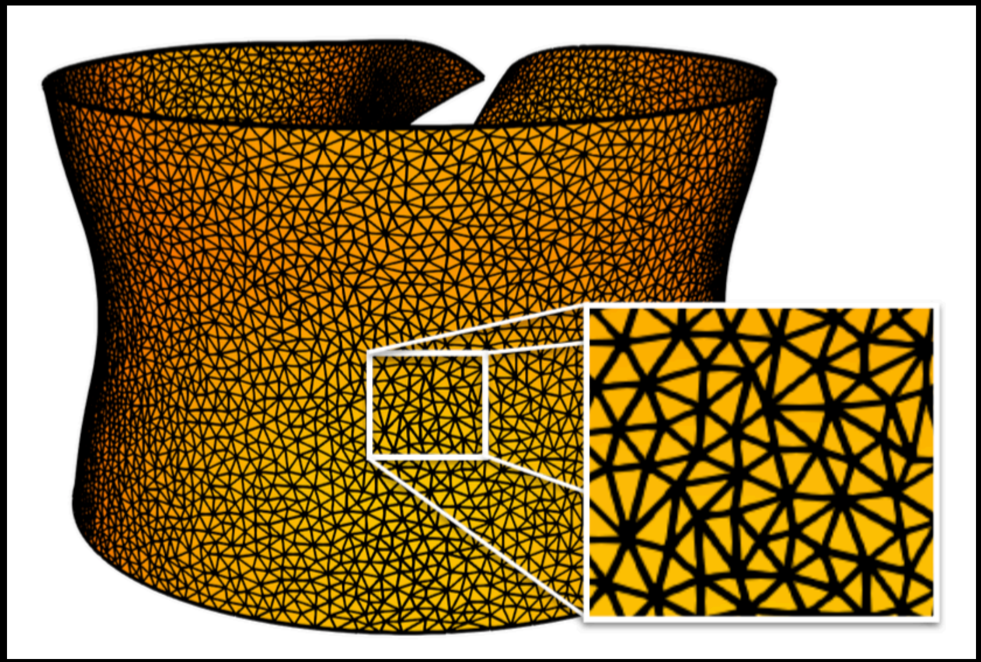


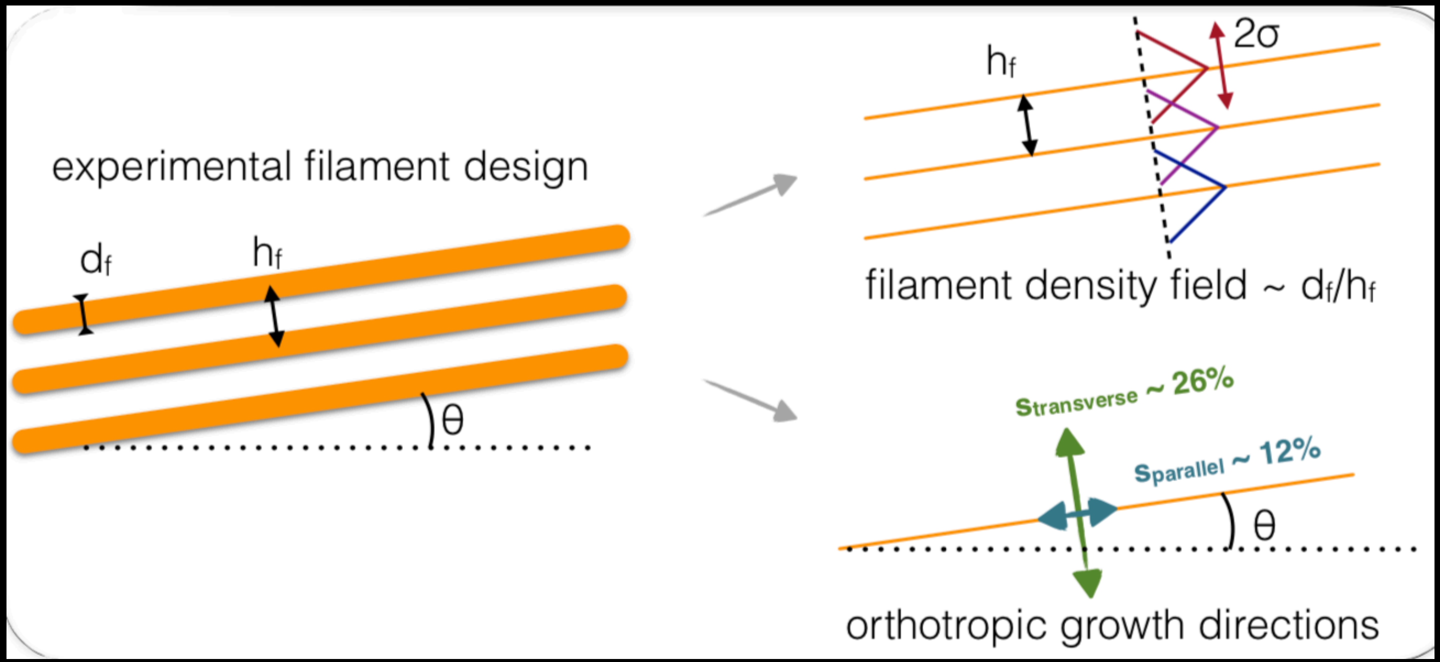


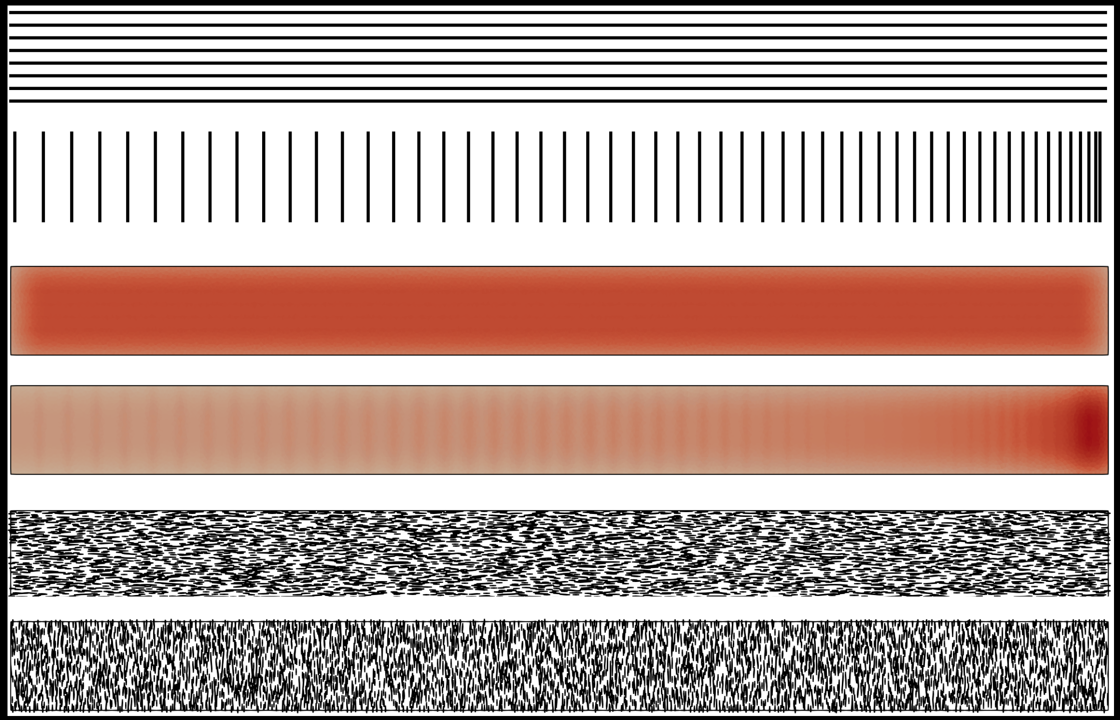
A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

The Simulations

Geometric Elastic Energy: $$E = \frac{1}{2}\int_{\Omega}\left[\frac{h}{4}\left(g_{ij}-\bar{g}_{ij}\right)^2 + \frac{h^3}{12}\left(b_{ij}-\bar{b}_{ij}\right)^2\right]$$

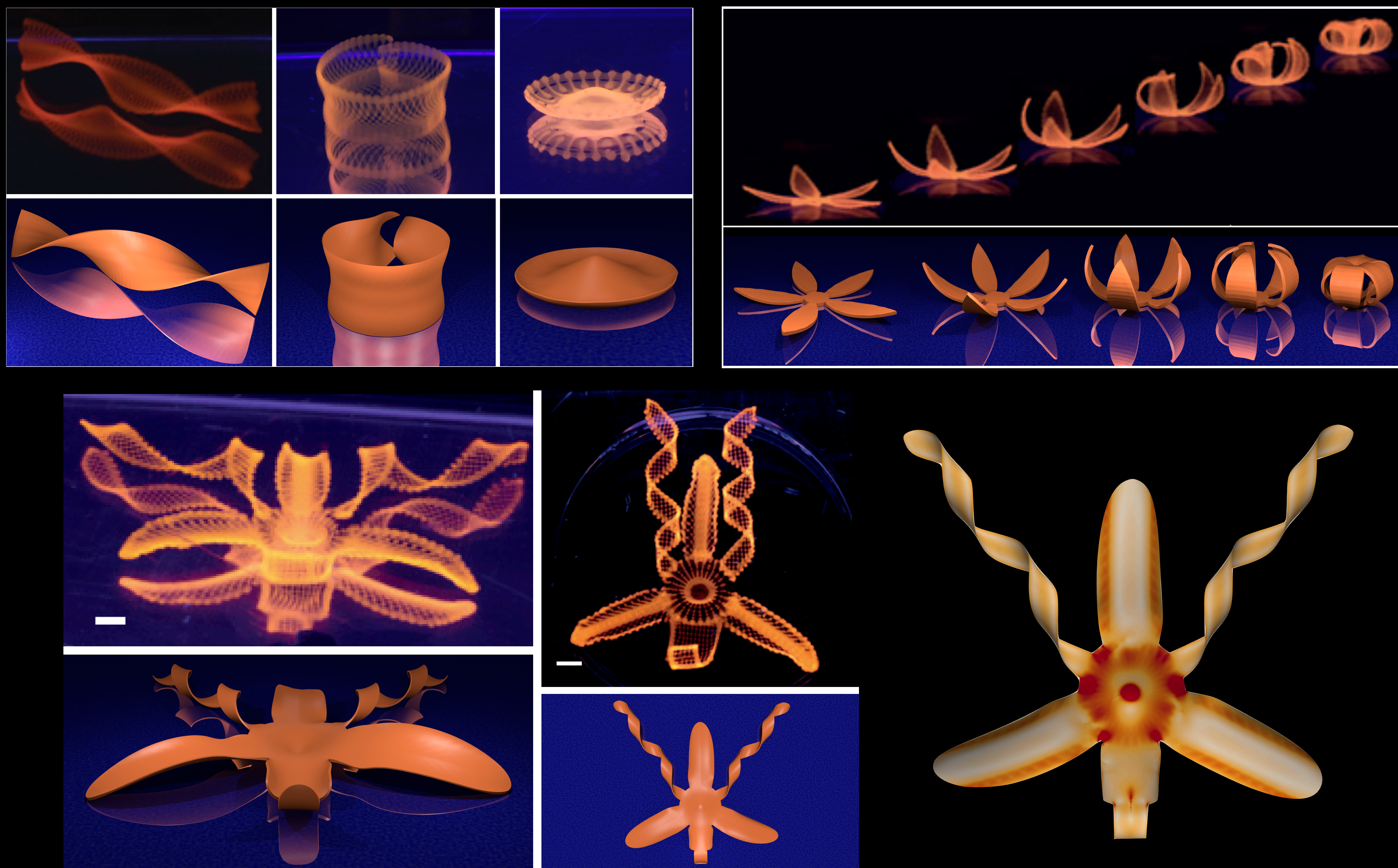






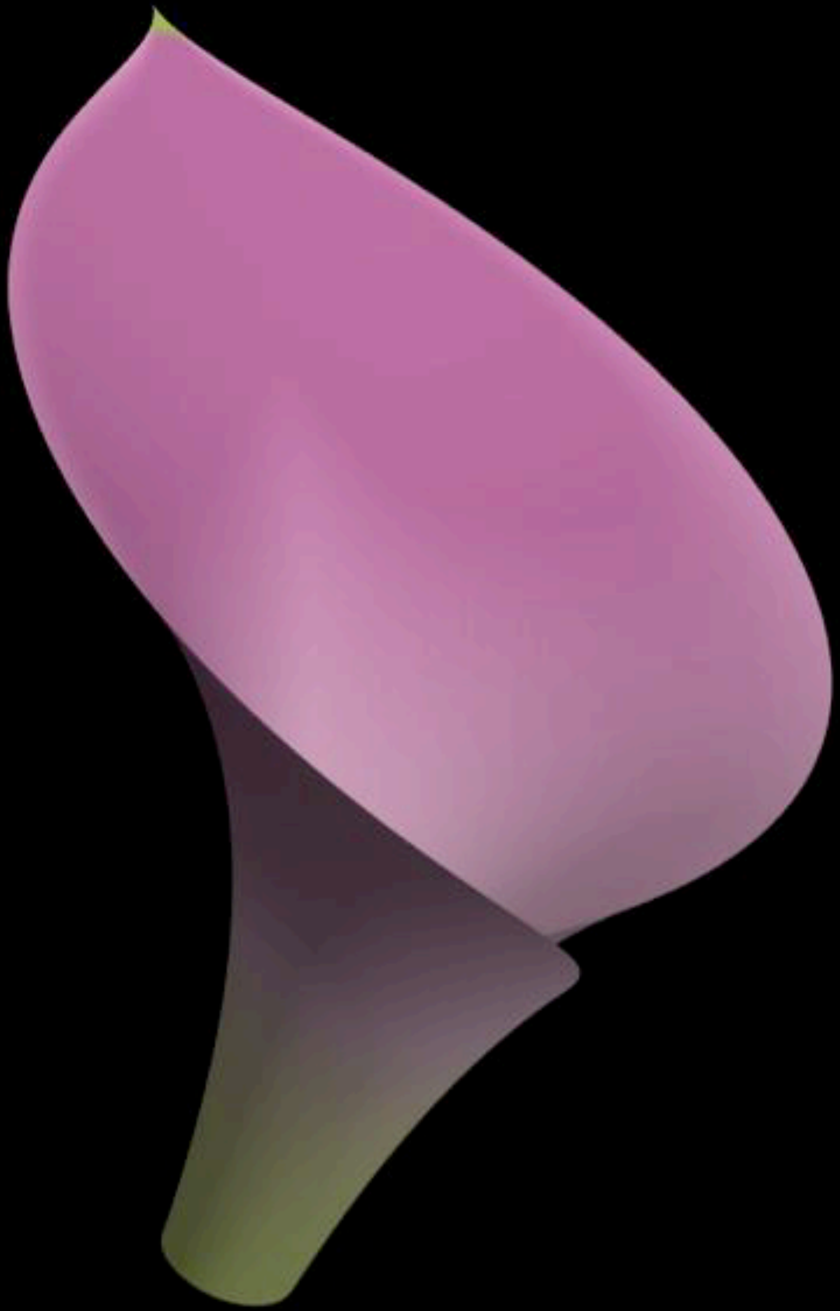
W. van Rees, **EAM**, A.S. Gladman, J.A. Lewis, and L. Mahadevan, *in preparation* 2018.

The Simulations: Comparison to Experiments



W. van Rees, **EAM**, A.S. Gladman, J.A. Lewis, and L. Mahadevan, *in preparation* 2018.

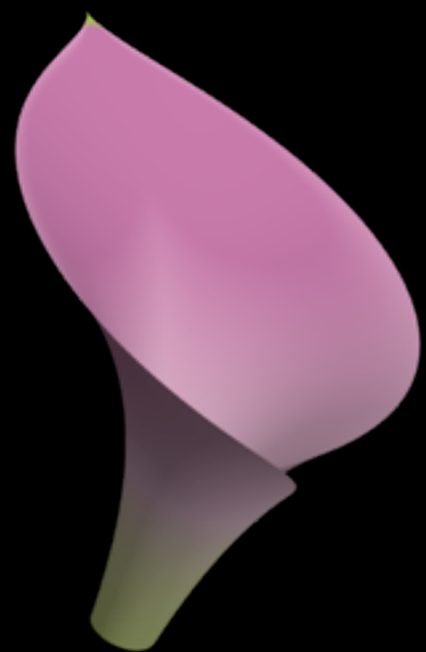
The Inverse Problem

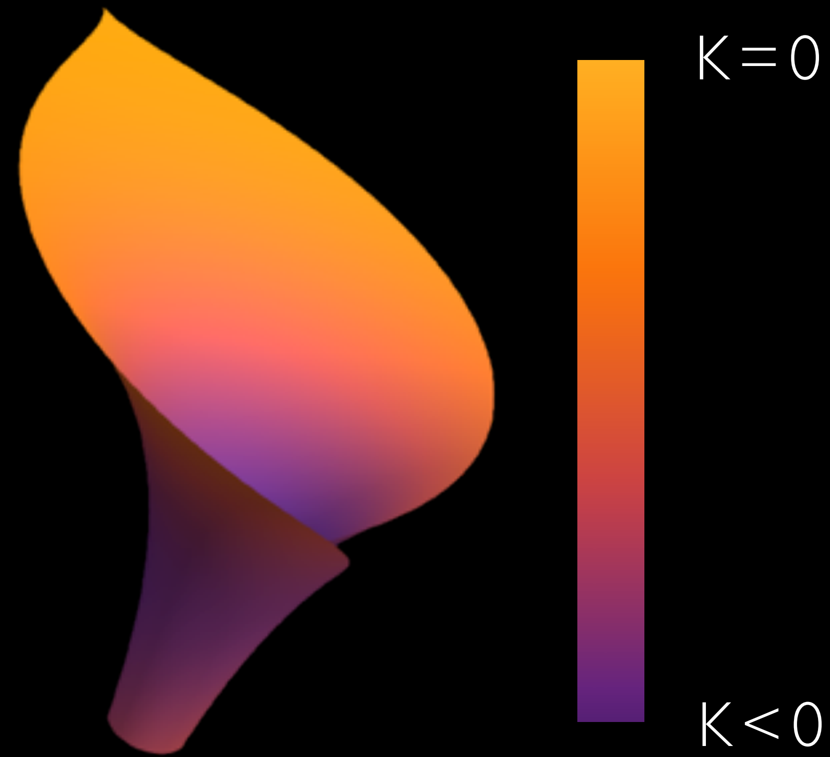


$$\Phi(u,v) = \left\{ \frac{e^{\frac{4u}{8+u}}}{2}\cos(v), \frac{e^{\frac{4u}{8+u}}}{2}\sin(v), \frac{u}{1+u/8} \right\}$$

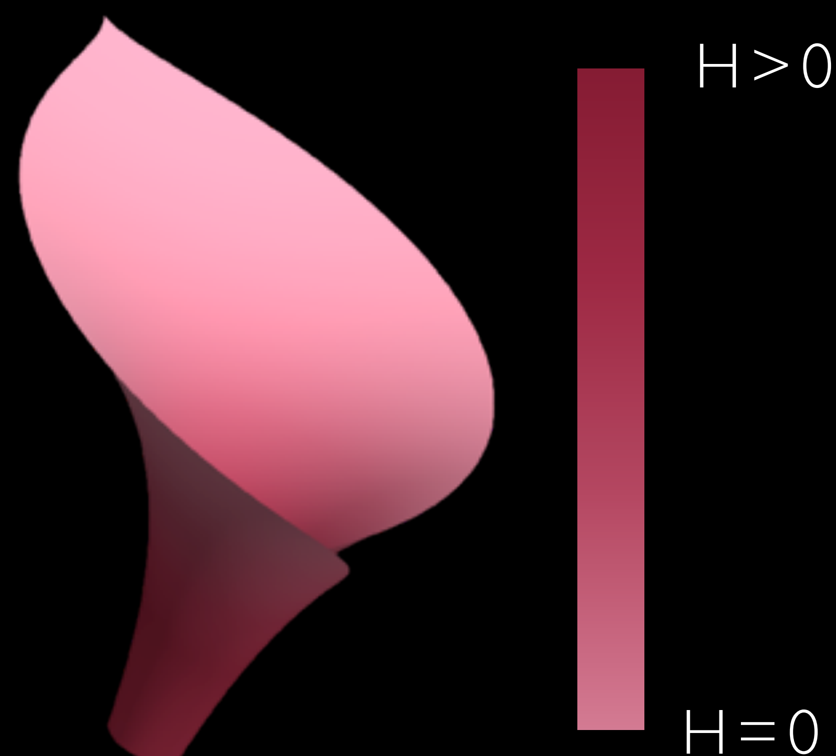
A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

The Inverse Problem





$$K = -\frac{64}{\left(e^{\frac{8u}{u+8}} + 16\right)^2}$$



$$H = \frac{128 e^{-\frac{4u}{u+8}}}{\left(e^{\frac{8u}{u+8}} + 16\right)^{3/2}}$$

$$\begin{aligned} X(u,v) &= \frac{e^{\frac{4u}{8+u}}}{2}\cos(v) \\ Y(u,v) &= \frac{e^{\frac{4u}{8+u}}}{2}\sin(v) \\ Z(u,v) &= \frac{u}{1+u/8} \end{aligned}$$

A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

Programming Local Curvatures

$$H = \frac{\alpha_\perp - \alpha_\parallel}{h} \frac{c_1 \sin^2(\theta)}{c_2 - c_3 \cos(2\theta) + m^4 \cos(4\theta)}, \quad K = -\frac{(\alpha_\perp - \alpha_\parallel)^2}{h^2} \frac{c_4 \sin^2(\theta)}{c_5 - c_6 \cos(2\theta) + m^4 \cos(4\theta)}$$

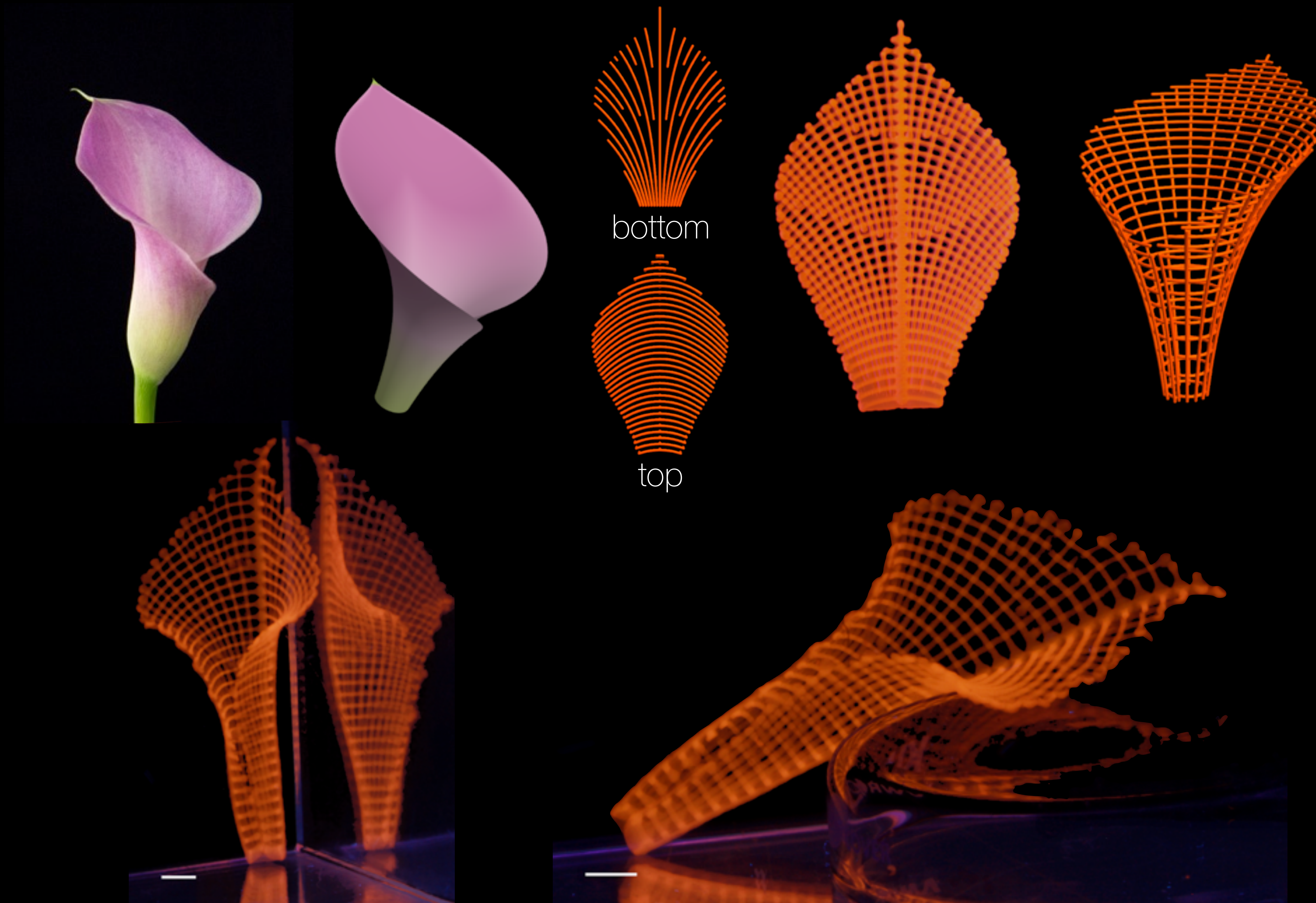
Given: $H,\ K,\ \alpha_\parallel,\ \alpha_\perp,\ \mathrm{E}^{(1)},\ \mathrm{E}^{(2)}$ Solve for: $\theta,\ m = a_1/a_2$

bottom

top

A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

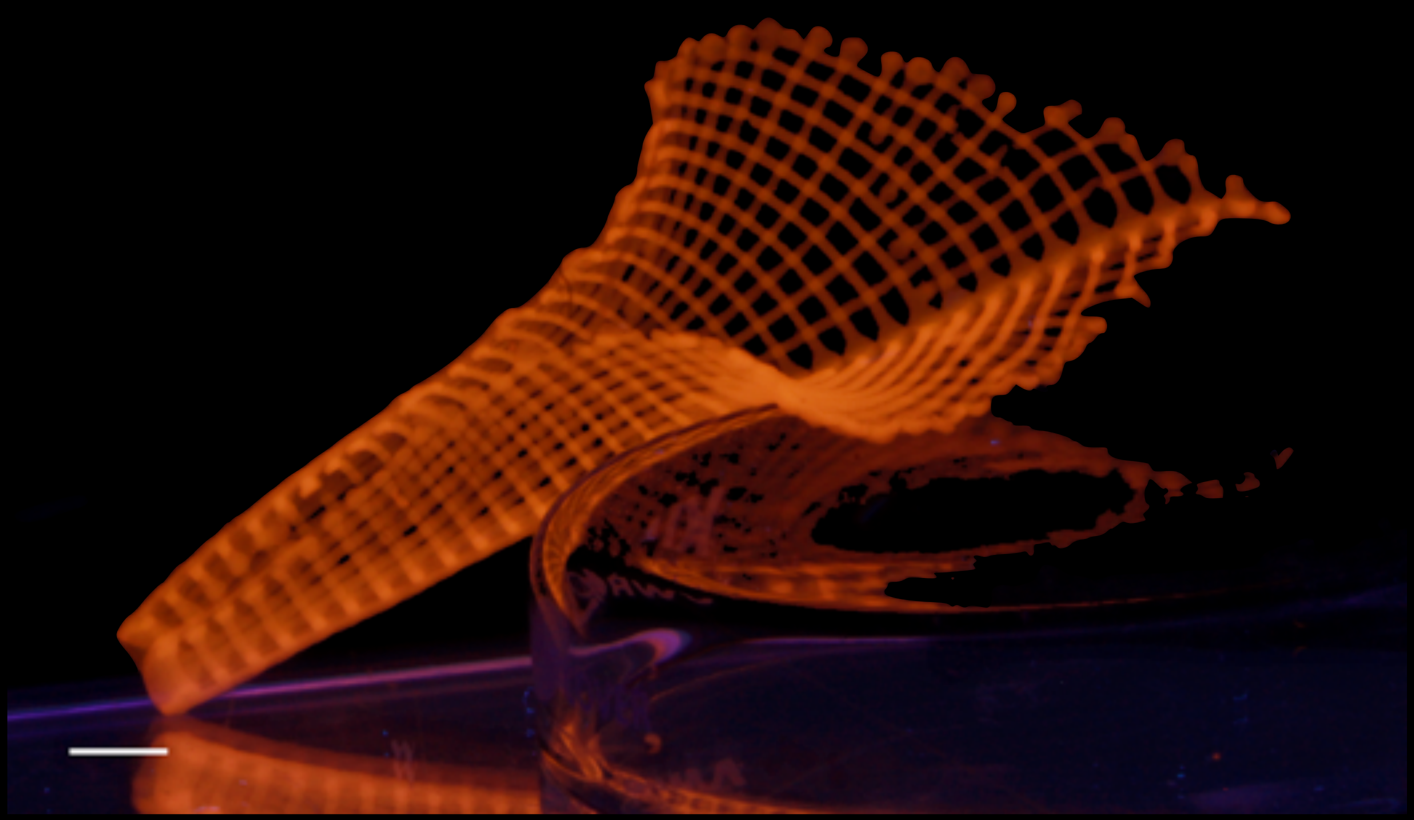
Programming Local Curvatures



A. S. Gladman, **EAM**, R. Nuzzo, L. Mahadevan, and J. Lewis, *Nat. Mater.* **15** (413) 2016.

Conclusions and Future Directions

- 3D printing hydrogel ink + cellulose nanofibrils simultaneously encodes anisotropy in swelling and elastic modulus. Complexity is free with additive manufacturing techniques.
- Local swelling anisotropy in a bilayer system generates curvature.
- Elasticity theory of anisotropic plates and shells allows us to predict mean and Gaussian curvatures.
- The inverse problem: How may we design print paths associated with specific target surfaces?
- Platform technology can be used with multi-stimuli responsive inks: light, temperature, electric field, hydration.



Acknowledgements

- **Dr. A. Sydney Gladman** - Harvard SEAS
- **Dr. Wim van Rees** - MIT Mechanical Engineering
- Prof. L. Mahadevan - Harvard SEAS
- Prof. Jennifer Lewis - Harvard SEAS
- NSF MRSEC DMR 14-20570
- NSF DMREF 15-33985
- Army Research Office Award W911NF-13-0489

Thank you!