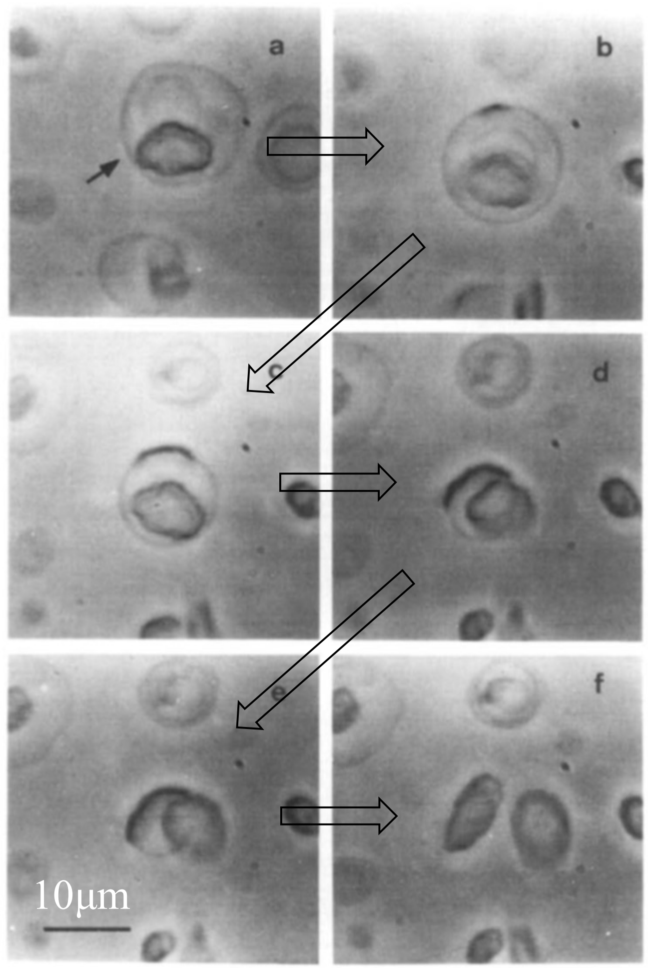
What is the physics of thin flexible sheets with a shear modulus?

Example 1: partially polymerized diacetylenic phospholipid vesicles

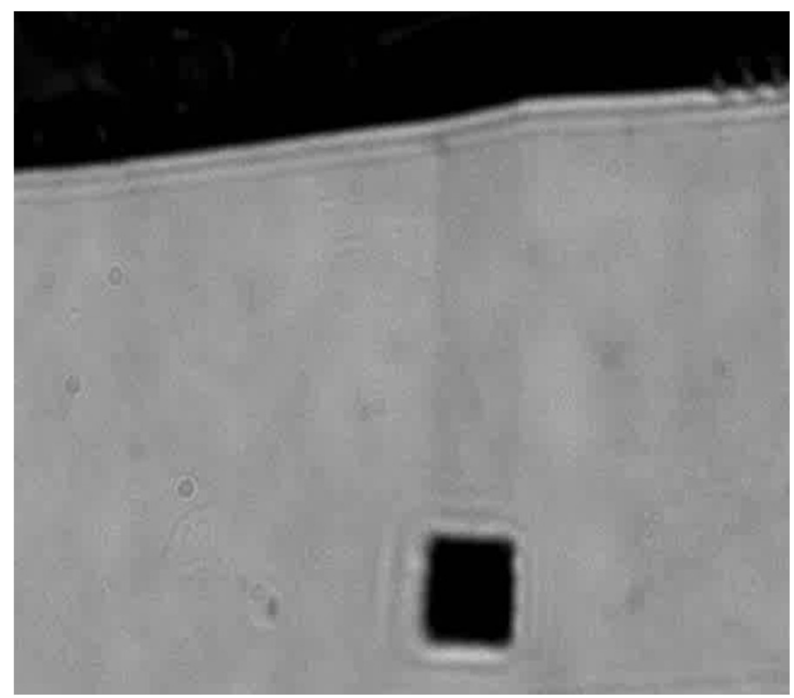
After UV polymerization, reversible wrinkling transition at 18°C obtained by cooling from 43°C. This vesicle contains additonal lyposomes including an already wrinkled one. time ~30s



D. Bensimon et al. Physica A194, 190 (1993)

Example 2: Graphene!

M. Blees, P. McEuen et al., Nature 524, 204 (2015)



Statistical Mechanics of Sheets, Shells and Cylinders

Statistical mechanics of thin plates

--nonlinear bending and stretching energies

--vK = Föppl-von Karman number = $YR^2/\kappa >> 1$

--strongly scale-dependent elastic parameters

The physics of dilations in fluctuating sheets

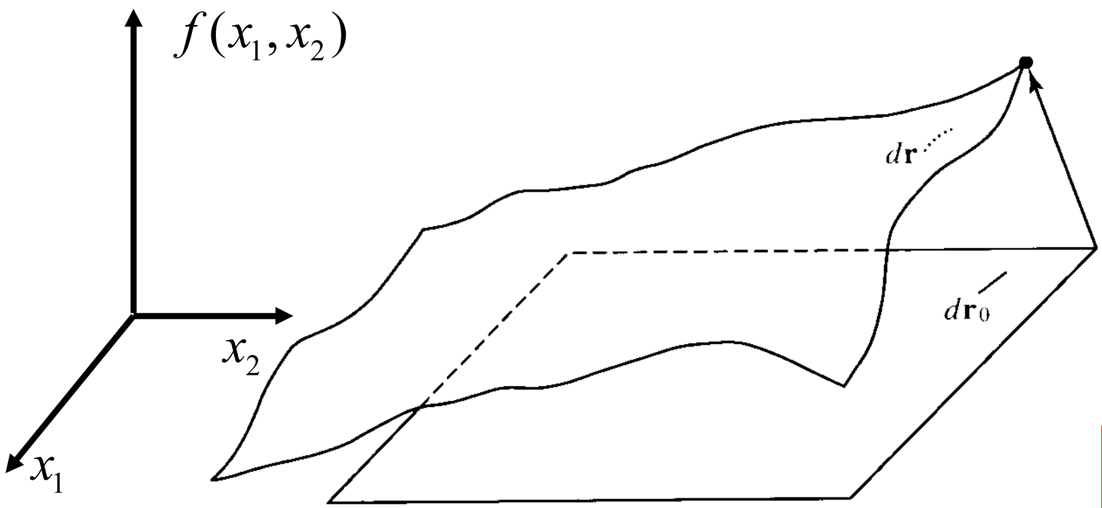
"dilations" are localized regions of positive or negative Gaussian curvature. These mechanical analogs of Ising spins, can order in a fluctuating background of flexural phonons

Abby Plummer
Paul Hanakata

Curvature matters: thermally excited spherical shells & cylinders differ from flat sheets!

Jayson Paulose
Andrej Kosmrlj

Thin plate theory: the Foeppl-von Karman equations (1904)



$$\vec{r}(x_1,x_2) = \vec{r}_0 + \begin{pmatrix} f(x_1,x_2) \\ u_1(x_1,x_2) \\ u_2(x_1,x_2) \end{pmatrix}$$

$$dr^2 = dr_0^2 + 2u_{ij}dx_i dx_j$$

$$u_{ij}(\vec{x}) = \frac{1}{2}\left[\frac{\partial u_i(\vec{x})}{\partial x_j} + \frac{\partial u_j(\vec{x})}{\partial x_i} + \frac{\partial f(\vec{x})}{\partial x_i}\frac{\partial f(\vec{x})}{\partial x_j}\right]$$

$$E = \frac{1}{2}\int d^2x\, [\underbrace{\kappa(\nabla^2 f(\vec{x}))^2}_{\text{bending energy}} + \underbrace{2\mu u_{ij}^2(\vec{x}) + \lambda u_{kk}^2(\vec{x})}_{\text{stretching energy}}]$$

bending energy

stretching energy

$\kappa =$ bending rigidity

$\mu =$ shear modulus

$\mu+\lambda =$ bulk modulus

Flexural phonons create a matrix "vector potential" and can escape softly into the 3rd dimension…

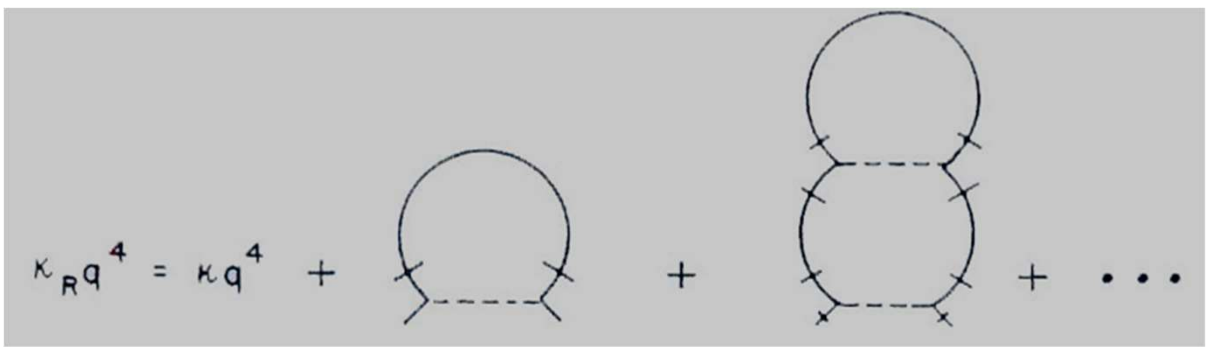
Statistical mechanics: thermally excited membranes *(L. Peliti & drn)*

Tracing out in-plane phonon degrees of freedom yields a massless nonlinear field theory at a critical point

$$F_{\text{eff}} = -k_B T \ln\left(\int D\{u_x(x,y)\} \int D\{u_y(x,y)\} e^{-E/k_B T} \right) \qquad \mathrm{Y} = \frac{4\mu(\mu+\lambda)}{2\mu+\lambda} = \text{Young's modulus}$$

$$F_{eff} = \frac{1}{2}\kappa \int d^2x \left[(\nabla^2 f)^2 \right] + \frac{1}{4} Y \int d^2x \left[P_{ij}^T (\partial_i f \partial_j f) \right]^2 \equiv F_0 + F_1; \quad P_{ij}^T = \delta_{ij} - \frac{\partial_i \partial_j}{\nabla^2}$$

Assume $k_B T/\kappa << 1$, and do low temperature perturbation theory…



$$\kappa_R(q) = \kappa + \frac{k_B T Y}{8} \int \frac{d^2k}{(2\pi)^2} \frac{[\hat{q}_i P_{ij}^T(\vec{k}) \hat{q}_j]^2}{\kappa |\vec{q} + \vec{k}|^4} + \ldots.$$

$$\lim_{q \to 0} \kappa_R(q) \approx \kappa[1 + 3(vK) k_B T / (32\pi^3 \kappa) + \ldots]$$

$vK = YL^2/\kappa \sim (L/h)^2 \gg 1$

L = membrane size

h = membrane thickness

Self-consistent bending rigidity, $\kappa_R(q) \sim 1/q$, diverges as $q \to 0$!

Renormalization Group for Thermally Excited Sheets (example of "self-organized criticality")

L. Peliti & drn (~1987)
J. Aronovitz and T. Lubenksy (~1988)
P. Le Doussal and L. Radzihovsky 1992 & Ann. Phys. ***39****, 340 (2018)*

$$E = \frac{1}{2}\int d^2x[\kappa(\nabla^2 f(\vec{x}))^2 + 2\mu u_{ij}^2(\vec{x}) + \lambda u_{kk}^2(\vec{x})]$$

$$u_{ij}(\vec{x}) = \frac{1}{2}\left[\frac{\partial u_i(\vec{x})}{\partial x_j} + \frac{\partial u_j(\vec{x})}{\partial x_i} + \frac{\partial f(\vec{x})}{\partial x_i}\frac{\partial f(\vec{x})}{\partial x_j}\right]$$

$$Z = \int \mathcal{D}\vec{u}(x_1,x_2)\int \mathcal{D}f(x_1,x_2)\exp(-E/k_BT)$$

define running coupling constants....

$$\bar{\mu}(l) = k_BT\mu a_0^2/\kappa^2; \quad \bar{\lambda}(l) = k_BT\lambda a_0^2/\kappa^2$$

scale dependent Young's modulus

$$Y(l) = \frac{4\mu(l)[\mu(l)+\lambda(l)]}{2\mu(l)+\lambda(l)}$$

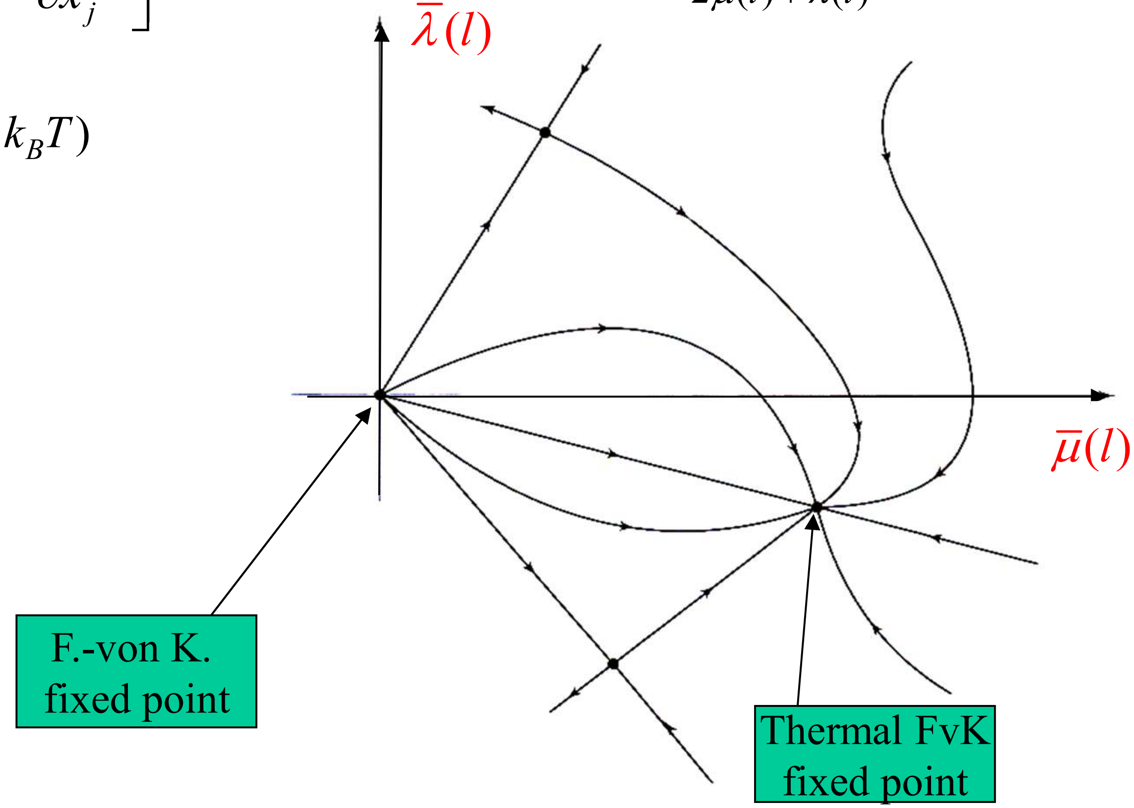
$$\kappa_R(l) \approx \kappa(l/l_{th})^{\eta}$$

$$Y_R(l) \approx Y(l_{th}/l)^{\eta_u}$$

$$\eta \approx 0.82, \quad \eta_u \approx 0.36$$

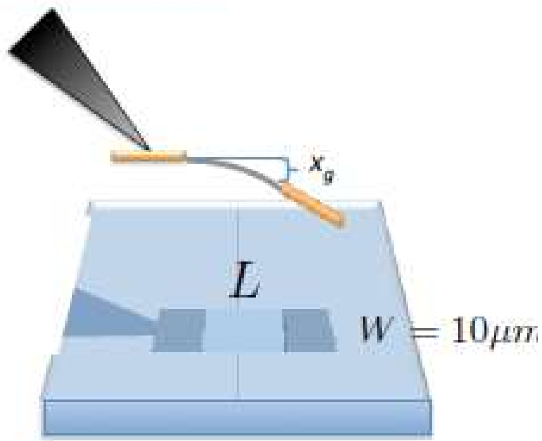
Thermal fluctuations dominate whenever $L > l_{th}$

$$l_{th} \approx \sqrt{\kappa^2/(k_BTY)}$$



Measured Bending rigidity of graphene membranes

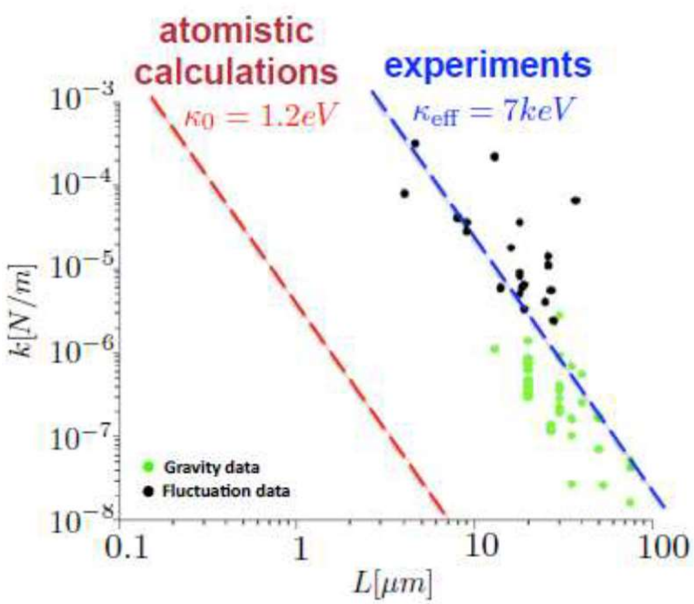
M. Blees et al., Nature 524, 204 (2015); T = 300ºK



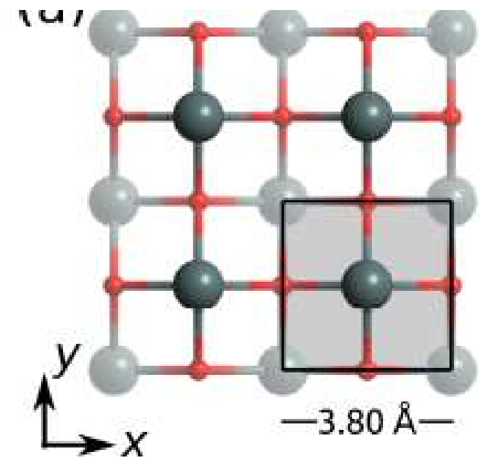
Bending rigidity enhanced ~4000 fold; agrees with

$$\kappa_R(l) \approx \kappa (W / l_{th})^{0.8}$$

(l_{th} ~ 0.2nm for graphene)
Scale-dependent bending rigidity!

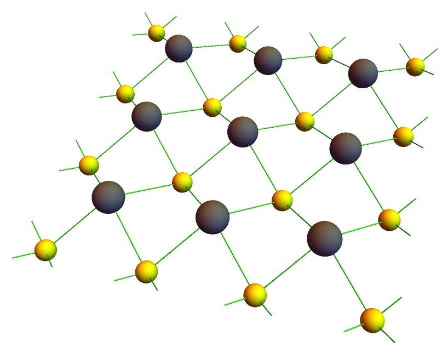


What about more complex "metamaterials"



Tin oxide SnO "AFM"

Seixas, et al., PRL, **116** (2016): 206803



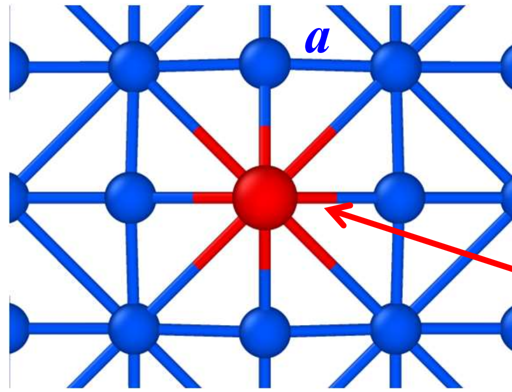
Lead Sulfide PbS "FM"

Hanakata, et al. *PR B***96** (2017): 161401

Dilational impurities hosted on a square lattice

$$E = \frac{k}{2}\sum_{<i,j>} \left| |\boldsymbol{r}_i - \boldsymbol{r}_j| - a_{ij} \right|^2 + \kappa \sum_{\alpha,\beta} (1 - \boldsymbol{n}_\alpha \cdot \boldsymbol{n}_\beta)$$

(n_α and n_β are normals to neighboring triangular plaquettes)



***a*(1+δ)**

Abigail Plummer, & drn, Phys. Rev E**102,** 033002 (2020)

Finite temperature simulations by Paul Hanakata

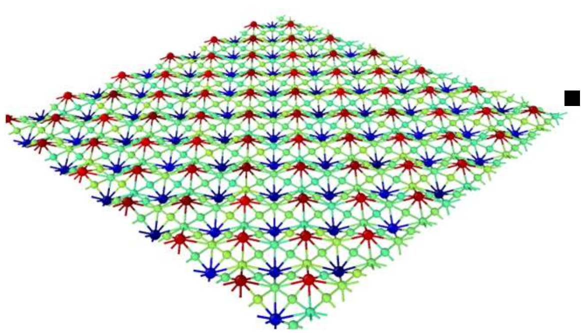
Regular nodes - Equilibrium bond length $r_e=a$
Impurities - Equilibrium bond length $r_e=a_{ij}(1+\delta)$
Spacing between impurities ***2a***

$\gamma = \frac{Y\Omega_0}{\kappa} =$ local vK number

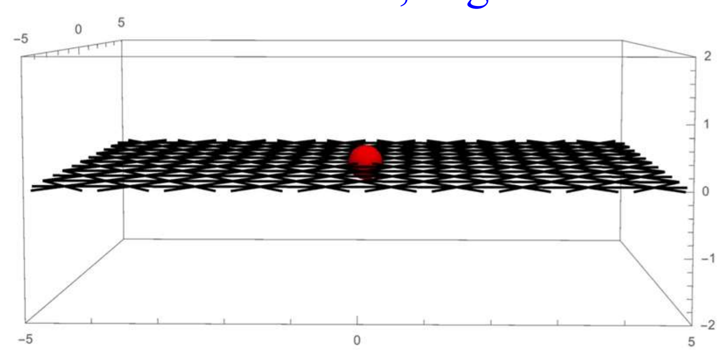
$\Omega_0 = 4a^2\delta =$ area of dilation

$\delta < 0 \leftrightarrow$ 'stitch', negative dilation

AFM ground state at *zero* temperature

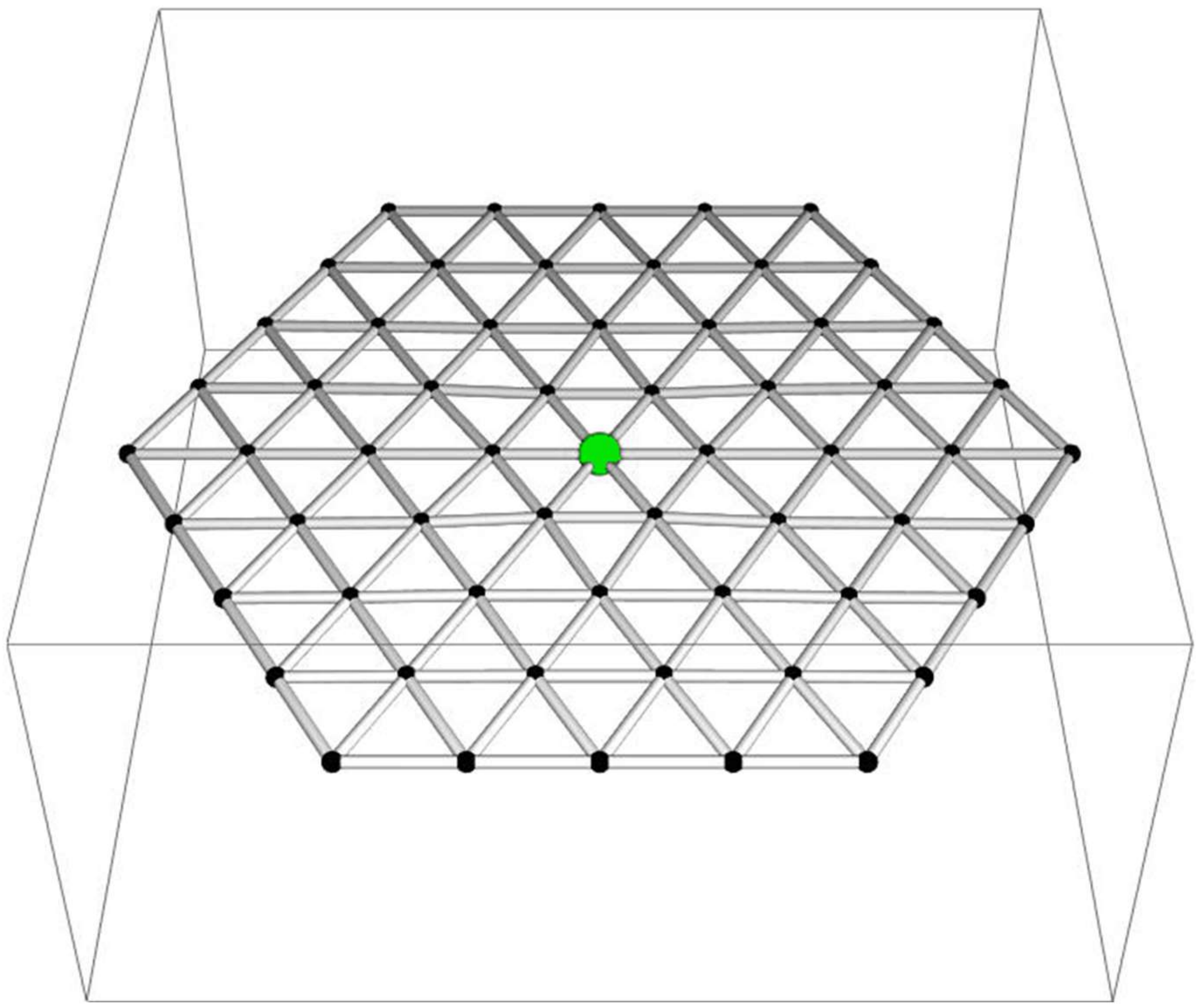


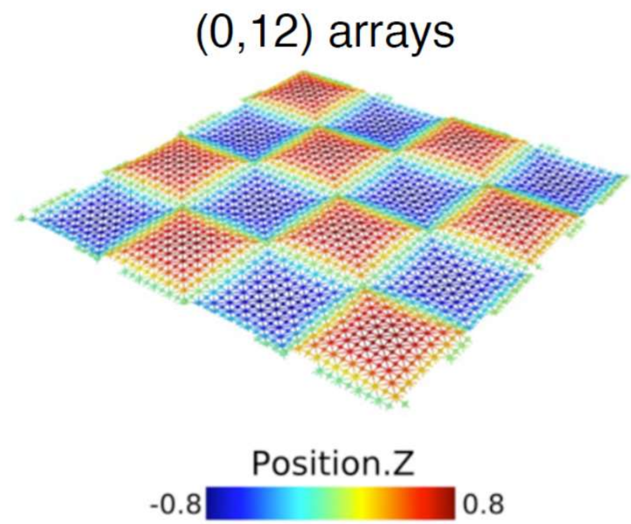
- Mechanical analog of an Ising <u>anti</u>ferromagnet….



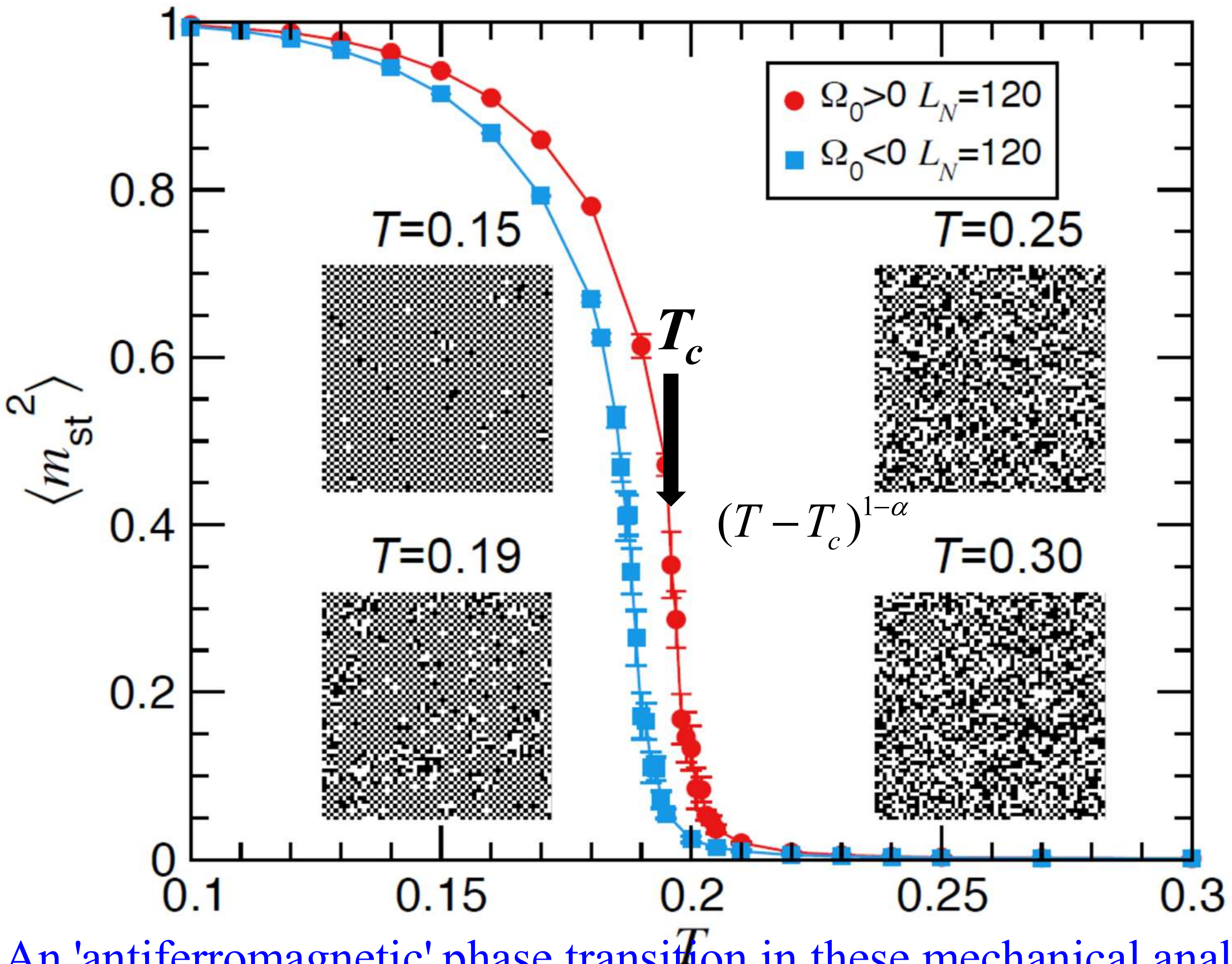
Increasing γ → mechanical analog of an Ising spin

Positive and negative dilations distort the host lattice in different ways

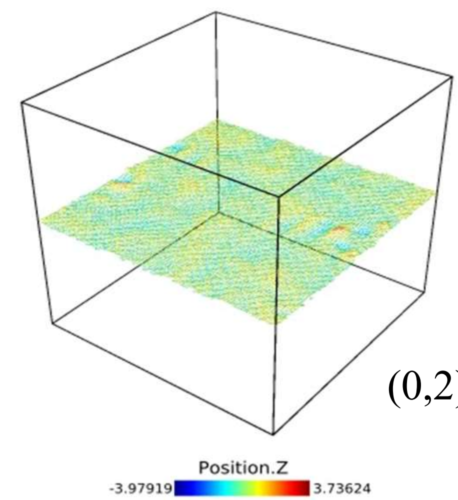




Both positive and negative dilation lattices exhibit an “antiferromagnetic” phase transition

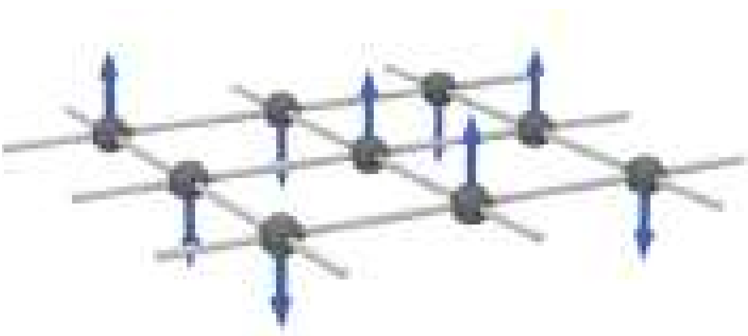


Paul Hanakata & Abby Plummer



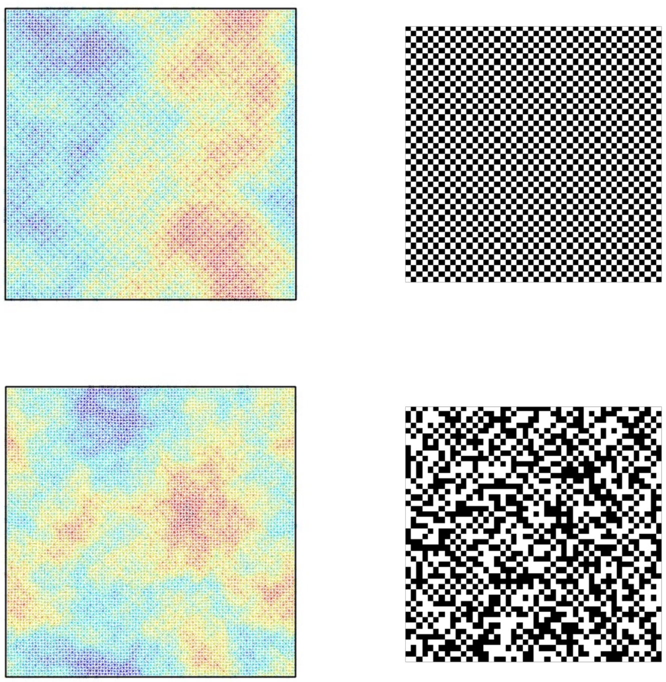
(0,2) PBC

An 'antiferromagnetic' phase transition in these mechanical analogs of Ising spins occurs at $k_B T_c \approx 0.2\kappa$ ($T_c = 800^o$ K), riding on a fluctuating background of flexural phonons



Couple the local squared staggered magnetization $m_{st}(x)$ to the strain field…

Top view | "Spins"



Staggered magnetization:

$$M_s = \sum_{i}^{N_I} (-1)^{(x_i+y_i)} s_i$$

$$H = H_{elastic} + H_{\text{Ising}} + H_{coupling}$$

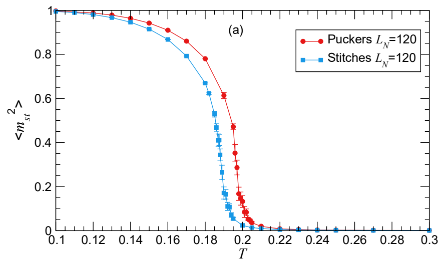
$$H_{elastic} = \frac{1}{2}\int d^2x \; [\kappa(\nabla^2 f(\vec{x}))^2 \;+\; 2\mu u_{ij}^2(\vec{x}) + \lambda u_{kk}^2(\vec{x})]$$

$$u_{ij}(\vec{x}) = \frac{1}{2}\left[\frac{\partial u_i(\vec{x})}{\partial x_j} + \frac{\partial u_j(\vec{x})}{\partial x_i} + \frac{\partial f(\vec{x})}{\partial x_i}\frac{\partial f(\vec{x})}{\partial x_j}\right]$$

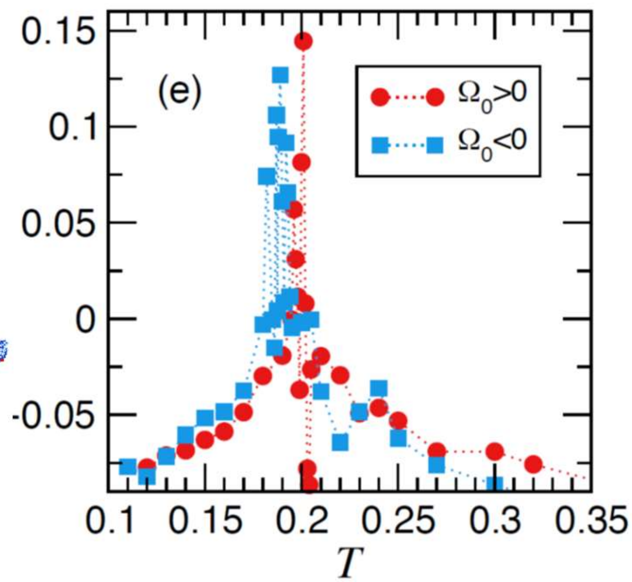
$$H_{\text{Ising}} = \int d^2x \; [\frac{1}{2}|\vec{\nabla} m_{st}|^2 \;+\; \frac{1}{2} r m_{st}^2 + u m_{st}^4], \quad r = T - T_c^0$$

$$H_{coupling} = g\int d^2x \; m_{st}^2(\vec{x}) u_{kk}(\vec{x}), \; g > 0$$

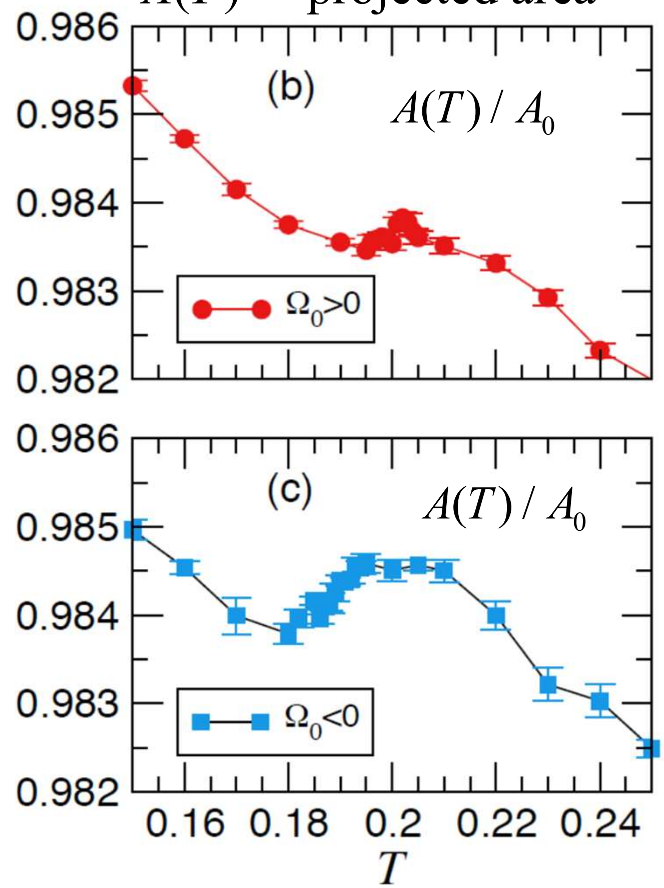
Anomalous thermal expansion when dilations disorder

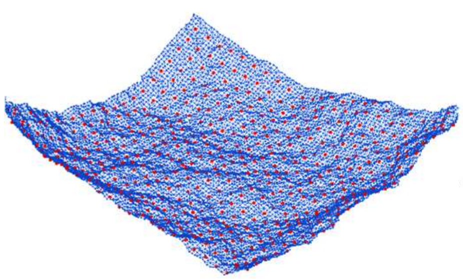


$$\alpha_T = \frac{1}{A_0}\frac{dA(T)}{dT} = \text{ coeff. of thermal exp.}$$



$A(T) =$ projected area





→ the disordering of the AFM dilations near T_c causes the membrane to swell.

$$A(T)/A_0 = -\underbrace{\frac{1}{2}\sum_q q^2 \left\langle |f(q)|^2 \right\rangle}_{\textit{shrinkage}} - \underbrace{\frac{g}{\mu+\lambda}\left\langle m_{st}^2 \right\rangle}_{\textit{expansion}}$$

Trace out the in-plane phonons to get an Ising order parameter coupled to the flexural phonons

$$H_{eff} = \frac{1}{2}\kappa\int d^2x\left[(\nabla^2 f)^2\right] + \frac{1}{8}Y\int d^2x\left[P_{ij}^T(\partial_i f\partial_j f)\right]^2 \text{ (flexural phonons)}$$

$$+\int d^2x\,\left[\frac{1}{2}|\vec{\nabla}m_{st}|^2 + \frac{1}{2}rm_{st}^2 + \tilde{u}m_{st}^4\right] + \frac{v}{A}\left(\int d^2x m_{st}^2\right)^2 \text{ (2d ising model)}$$

$$+w\int d^2x\; m_{st}^2 P_{ij}^T(\partial_i f\partial_j f) \qquad \text{(cross coupling)}$$

$$Y = \frac{4\mu(\mu+\lambda)}{2\mu+\lambda} \quad \tilde{u} = u - \frac{g^2}{2(2\mu+\lambda)} \quad v = \frac{-g^2\mu}{2(\mu+\lambda)(2\mu+\lambda)} \quad w = \frac{g\mu}{2\mu+\lambda}$$

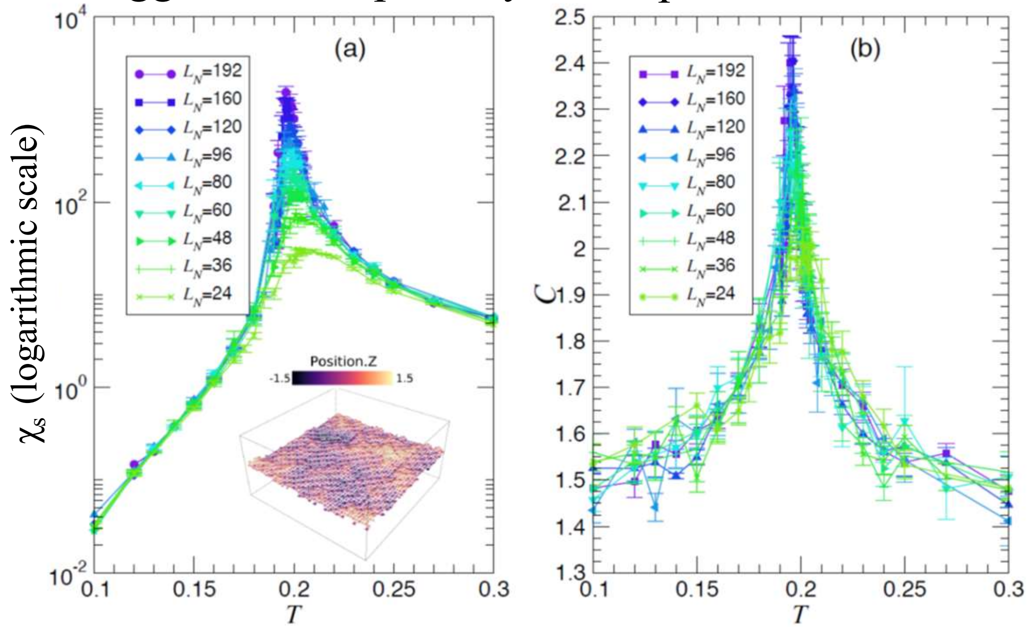
The term $w\int d^2x\; m_{st}^2 P_{ij}^T(\partial_i f\partial_j f)$ produces a long range interaction between $m_{st}^2(\vec{\mathrm{x}})$ and the local Gaussian curvature $G(\vec{x}) = \det\left(\frac{\partial^2 f}{\partial x_i \partial x_j}\right)$ of the flexural phonons

$$w\int d^2x\; m_{st}^2 P_{ij}^T(\partial_i f\partial_j f) = w\int d^2x\int d^2x' m_{st}^2(\vec{\mathrm{x}})\ln|\vec{x}-\vec{x}'|\,G(\vec{x}'), \quad G(\vec{x}') = \det\left(\frac{\partial^2 f(\vec{x})}{\partial x'_i\,\partial x'_j}\right)$$

$dw(\ell)/d\ell \approx \frac{19}{20}w(\ell)$ (w strongly perturbs Onsager's 2d Ising solution!)

The Ising ordering of dilations coupled to flexural phonons *may* be in a new universality class…

staggered susceptibility   specific heat



$$\chi = \frac{1}{N_I k_B T}\langle M_s^2\rangle - \langle|M_s|\rangle^2$$

$$C_V = \frac{1}{N k_B T^2}(\langle E^2\rangle - \langle E\rangle^2)$$

$L =$ system size

$\chi_{\max}(T = T_c) \propto L^{\gamma/\nu}$,

$\gamma/\nu \approx 1.70 \pm 0.06$

$L =$ system size

$C_V^{\max}(T = T_c) \propto L^{\alpha/\nu}$,

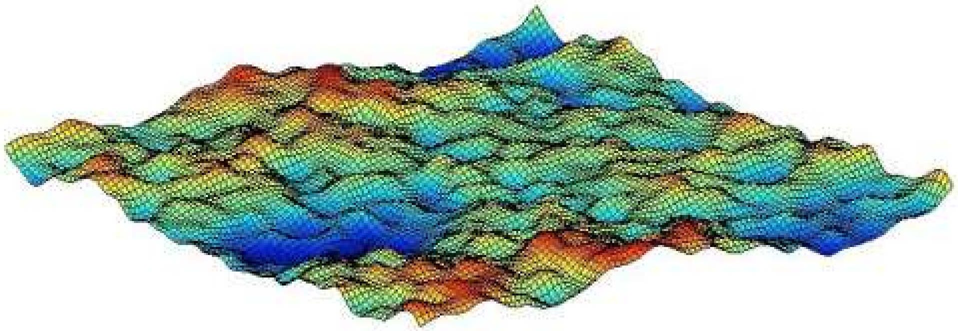
$\alpha/\nu \approx 0.07 \pm 0.02$

flexural Ising model	Onsager
$\alpha/\nu = 0.071 \pm 0.017$	0(log)
$\gamma/\nu = 1.712 \pm 0.062$	7/4=1.75

Our measured critical exponents for this highly compressible Ising model coupled to flexural phonons are several standard deviation away from Onsagers famous exact solution of the incompressible Ising model in two dimensions.

Can anyone calculate them?

Curvature matters: The statistical mechanics of spherical and cylindrical shells seem have striking differences in the themodynamic limit of large size!



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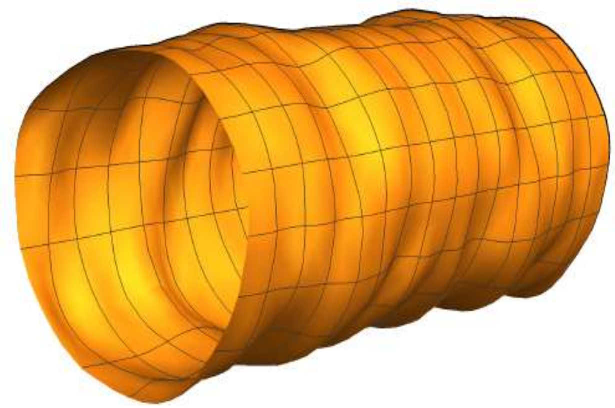

What is is the physics of thin, fluctuating shells and cylinders?

Do they collapse when sufficiently large?

(A. Kosmrlj & drn)

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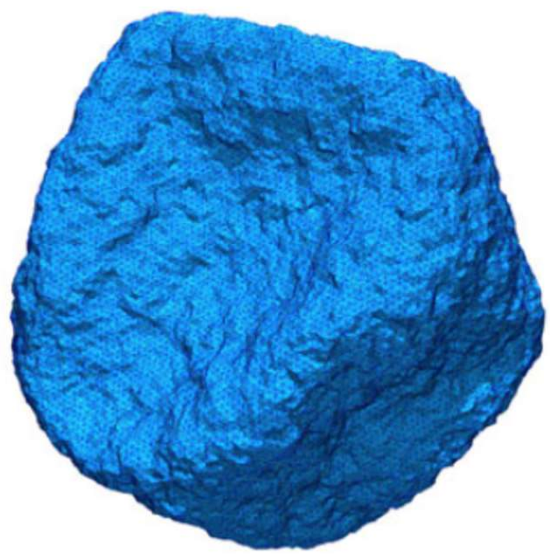


S. Komura, Shigeyi and R. Lipowsky. "Fluctuations and stability of polymerized vesicles." *Journal de Physique II* 2, no. 8 (1992): 1563-1575,

Statistical mechanics of thin spherical shells and spherocylinders…

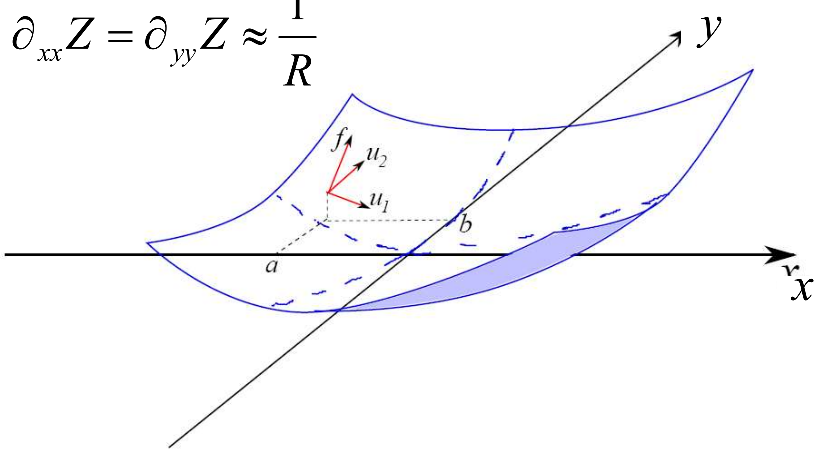
Evacuated tanker car crushed by atmospheric pressure; what happens on the nanoscale?

https://www.youtube.com/watch?v=Zz95_VvTxZM

Initial shape: $\begin{cases} z = Z(x,y) \\ z = \sqrt{R^2 - x^2 - y^2} \end{cases}$

To study thermal deformations of spherical shells, we use shallow shell theory….

$$\partial_{xx} Z = \partial_{yy} Z \approx \frac{1}{R}$$



$$\begin{pmatrix} x \\ y \\ Z(x,y) \end{pmatrix} \to \begin{pmatrix} x + u_x(x,y) - f(x,y)\partial_x Z(x,y) \\ y + u_y(x,y) - f(x,y)\partial_y Z(x,y) \\ Z(x,y) + f(x,y) \end{pmatrix}$$

$$E = \frac{1}{2}\int d^2x [\kappa(\nabla^2 f(\vec{x}))^2 + 2\mu u_{ij}^2(\vec{x}) + \lambda u_{kk}^2(\vec{x})]$$

$$ds'^2 = ds^2 + 2u_{ij} dx_i dx_j$$

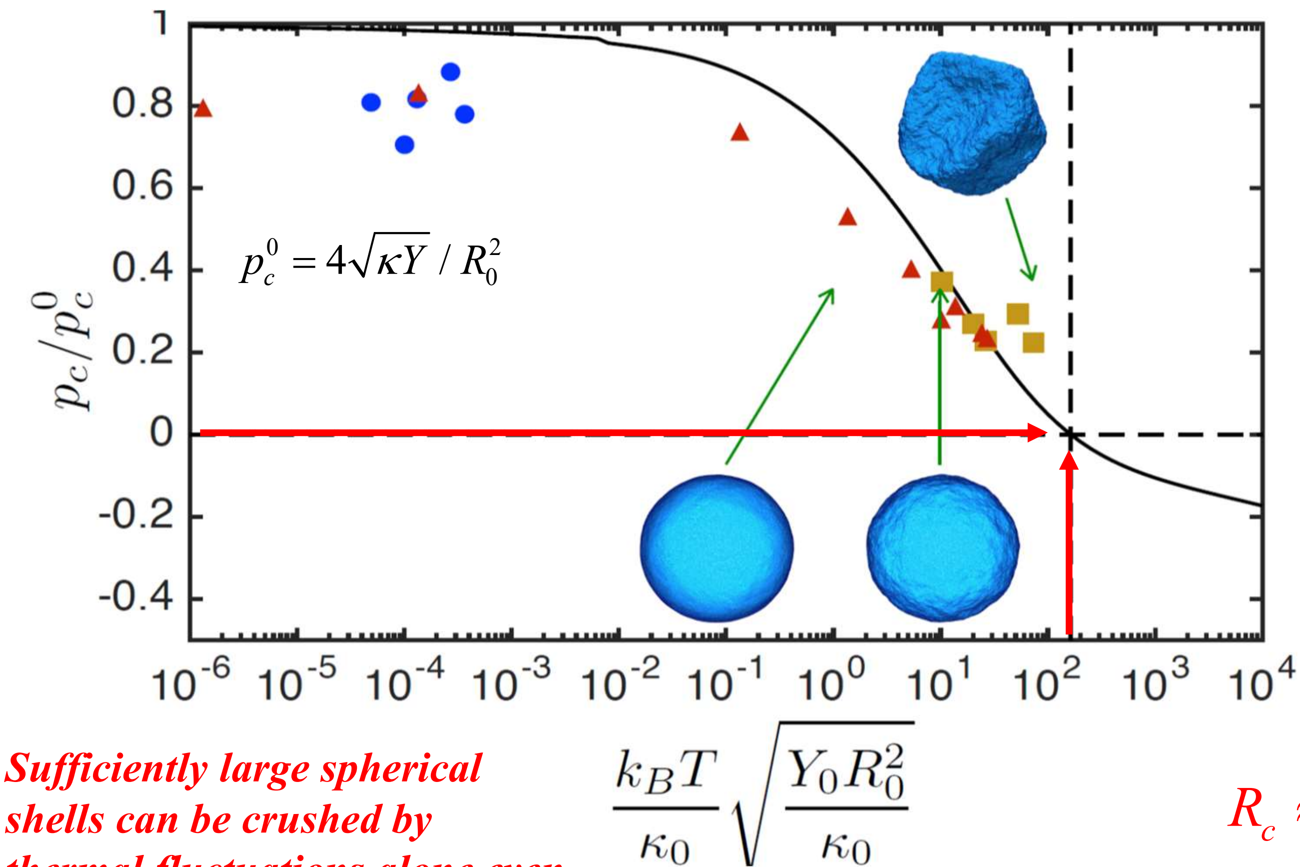
$$u_{ij}(\vec{x}) = \frac{1}{2}\left[\frac{\partial u_i(\vec{x})}{\partial x_j} + \frac{\partial u_j(\vec{x})}{\partial x_i} + \frac{\partial f(\vec{x})}{\partial x_i}\frac{\partial f(\vec{x})}{\partial x_j}\right]$$

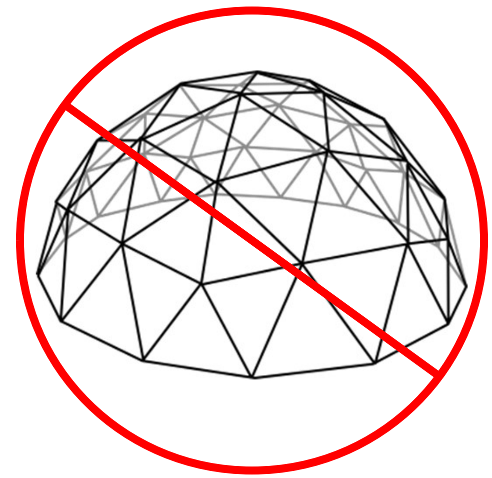
$$u_{ij}(\vec{x}) = \frac{1}{2}\left[\frac{\partial u_i(\vec{x})}{\partial x_j} + \frac{\partial u_j(\vec{x})}{\partial x_i} + \frac{\partial f(\vec{x})}{\partial x_i}\frac{\partial f(\vec{x})}{\partial x_j} - \delta_{ij}\frac{f}{R}\right]$$

Paulose et al. PNAS **109,** 19551 (2012); *Kosmrlj & drnPhysical Review X* 7 011002 (2017)

Thermalized Shells with $\kappa_R(l) \approx \kappa(l/l_{th})^{0.8}$, $Y_R(l) \approx Y(l_{th}/l)^{0.4}$

Spherical shells, even at zero microscopic pressure, are crushed when the thermally renormalized pressure exceeds the critical buckling threshold





Amorphous domes with R > R_c will collapse

Sufficiently large spherical shells can be crushed by thermal fluctuations alone even when p=0

$$R_c \approx 160 \frac{\kappa}{k_B T}\sqrt{\frac{\kappa}{Y}} \sim \frac{Eh^4}{k_B T}$$

($R_c \approx 160nm$ for graphene)

J. Paulose et al., PNAS **109**, 19551 (2012)

A, Kosmrlj and drn, Phys. Rev. X**7** 011002 (2017)

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Statistical mechanics of thin plates

Questions?

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