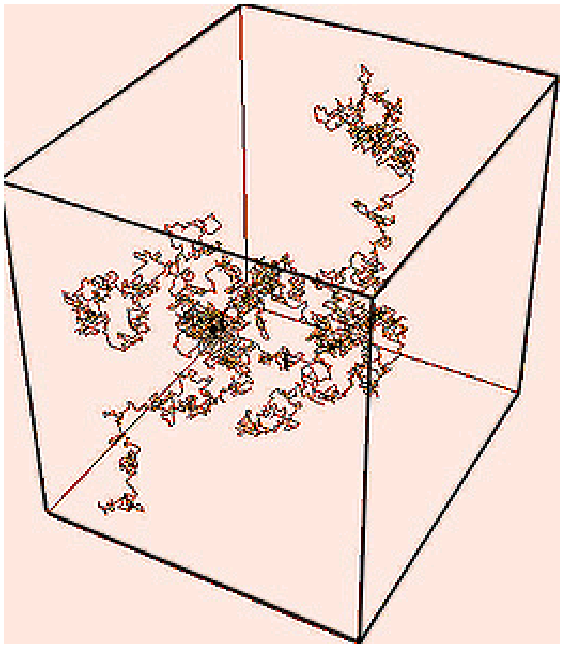
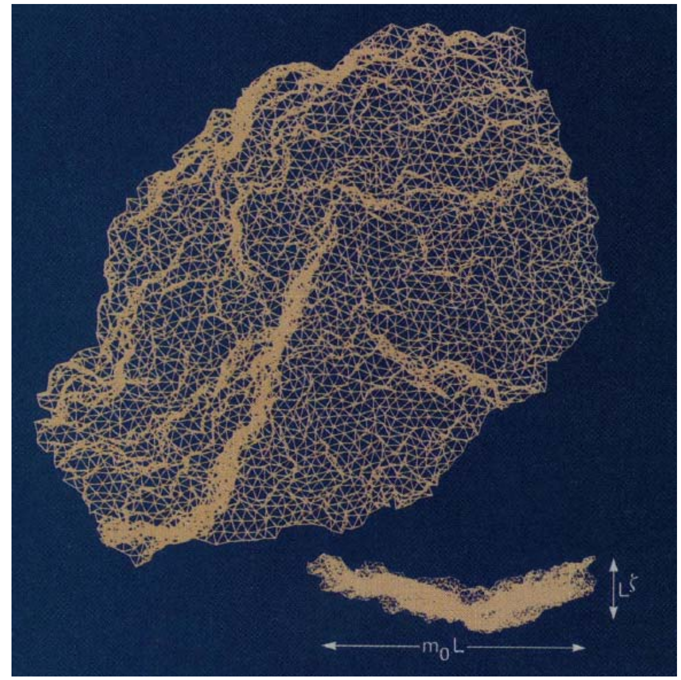
Ancient History: from linear polymers to tethered surfaces

By the 1990's, theories of linear polymer chains in a good solvent were generalized to treat the statistical mechanics of flexible sheet polymers





F. Abraham and drn, Science 249, 393 (1990)

sheet polymer = soft interface permeable to solvent molecules on either side…

- Remarkably, "tethered surfaces" with a shear modulus are able to resist thermal crumpling and exhibit a low temperature, "wrinkled" flat phase…
- A continuous broken symmetry --long range order in the surface normals-- arises in two dimensions (violates Mermin-Wagner-Hohenberg theorem)

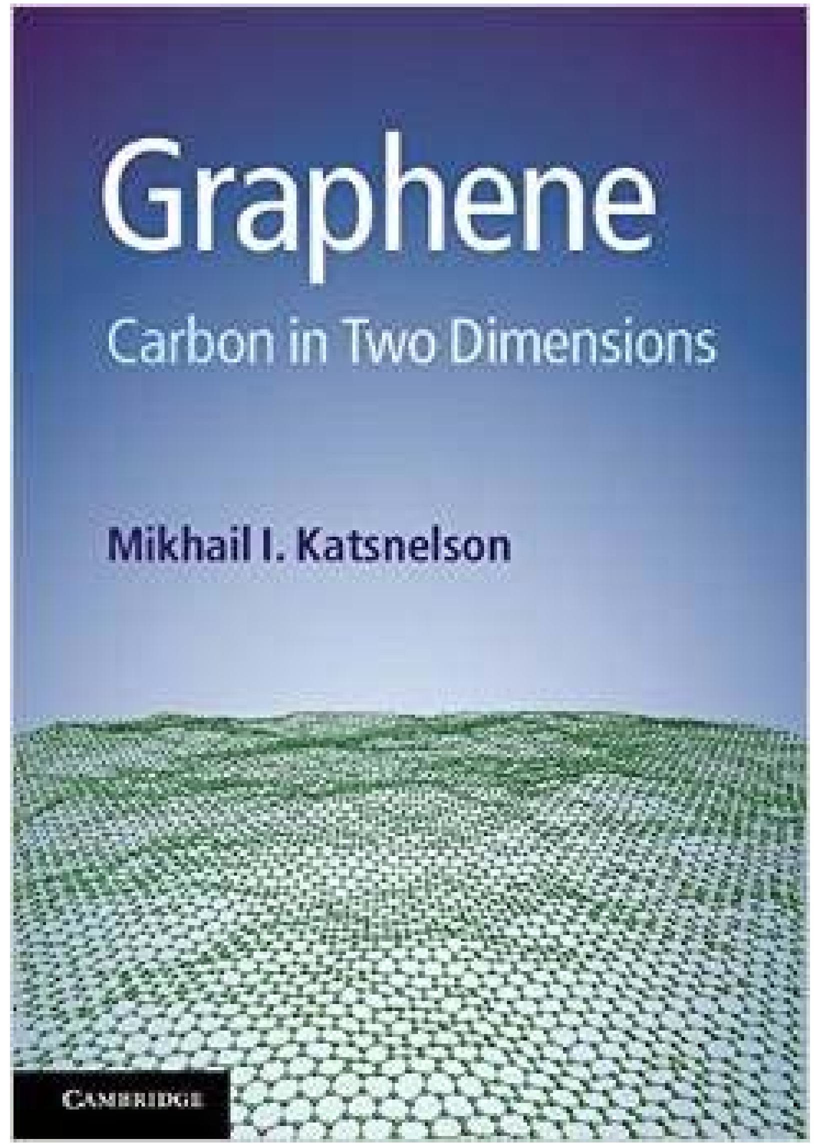
Lots of recent interest in the flat phase among graphene theorists, but (until recently) not many experiments….

Influence of out-of-plane phonons on electronic properties:

1. E. Mariani and F. von Oppen, Phys. Rev. Lett. 100, 076801 (2008).
2. K. S. Tkhonov, W. L. Z. Zhao and A. M. Finkel'stein, Phys. Rev. Lett. 113, 076601 (2014).

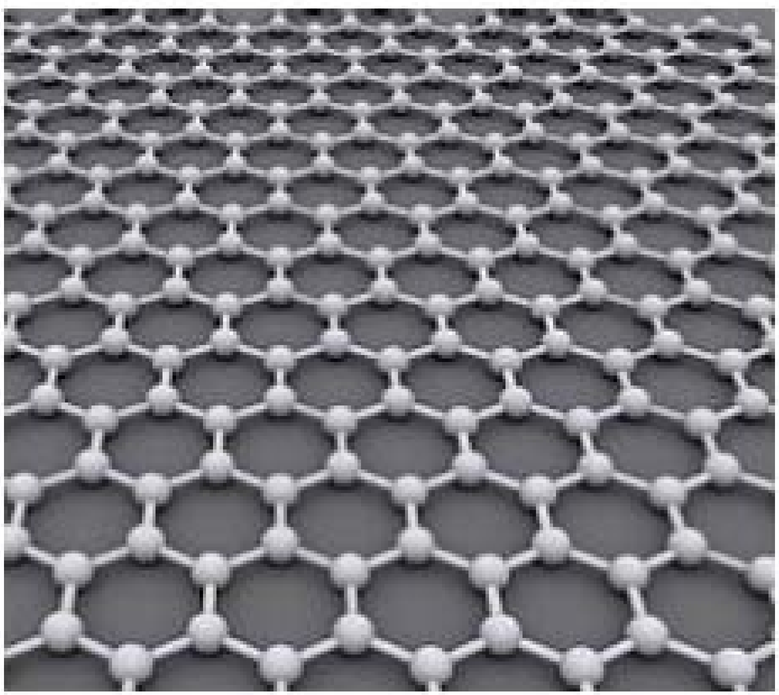
Quantum effects at low temperatures:

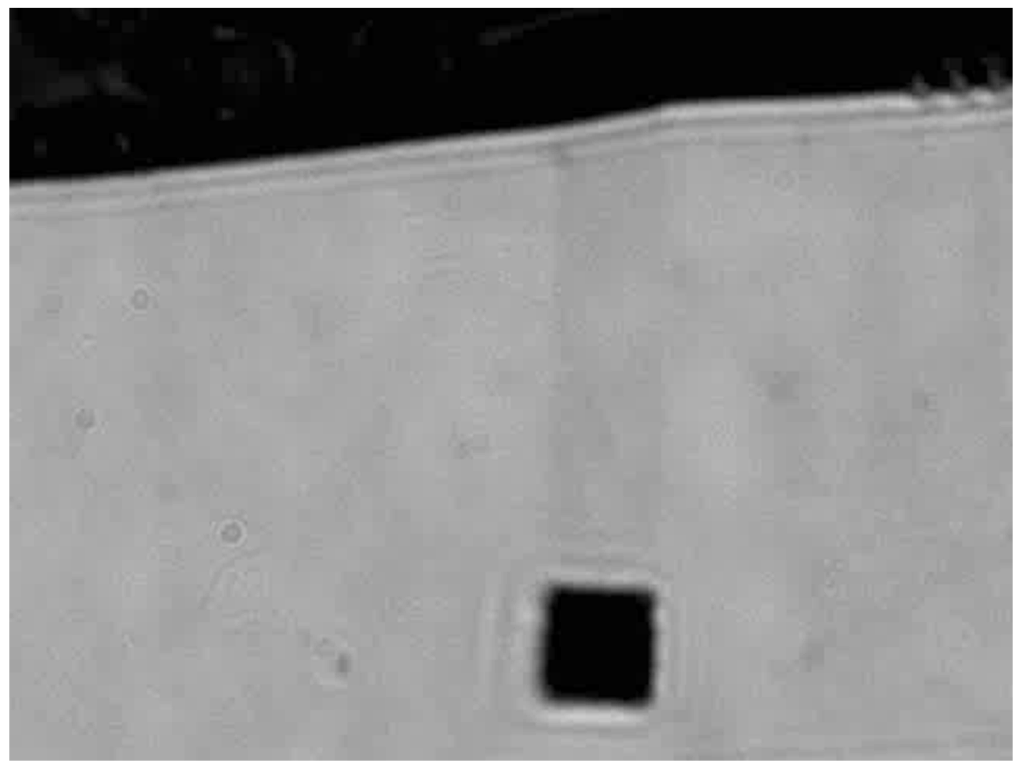
1. E. I. Kats and V. V. Lebedev, Phys. Rev. B89, 125433 (2014).
2. B. Amorim, R. Roldan, E. Cappelluti, F. Guinea, A. Fasolino and M. I. Katsnelson, Phys. Rev. B 89, 224307 (2014)



Experiments of the McEuen group Cornell: **"Single molecule polymer physics" for graphene**

graphene





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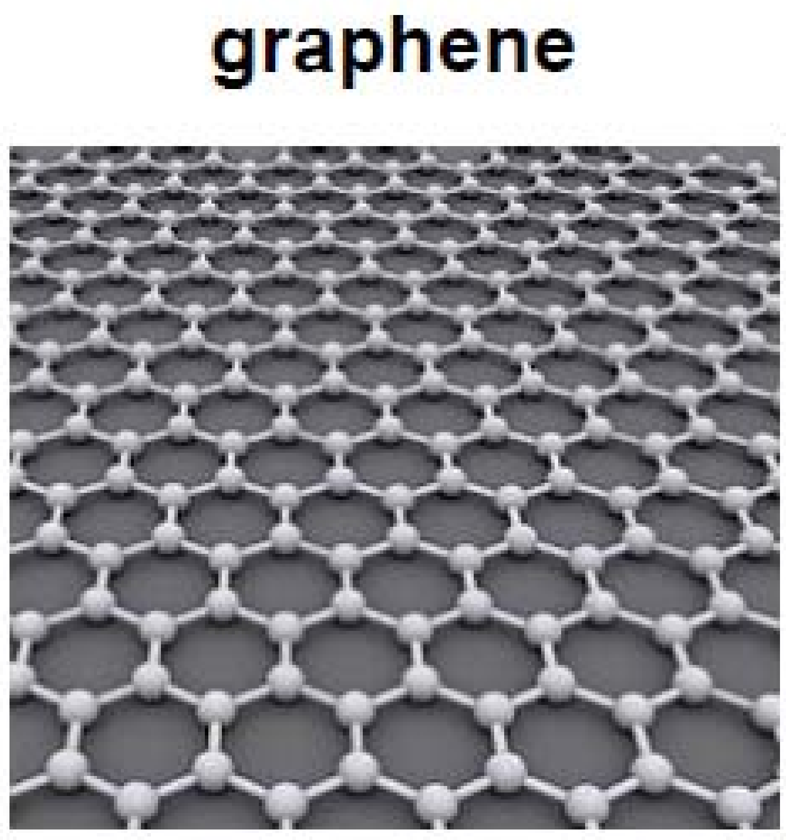
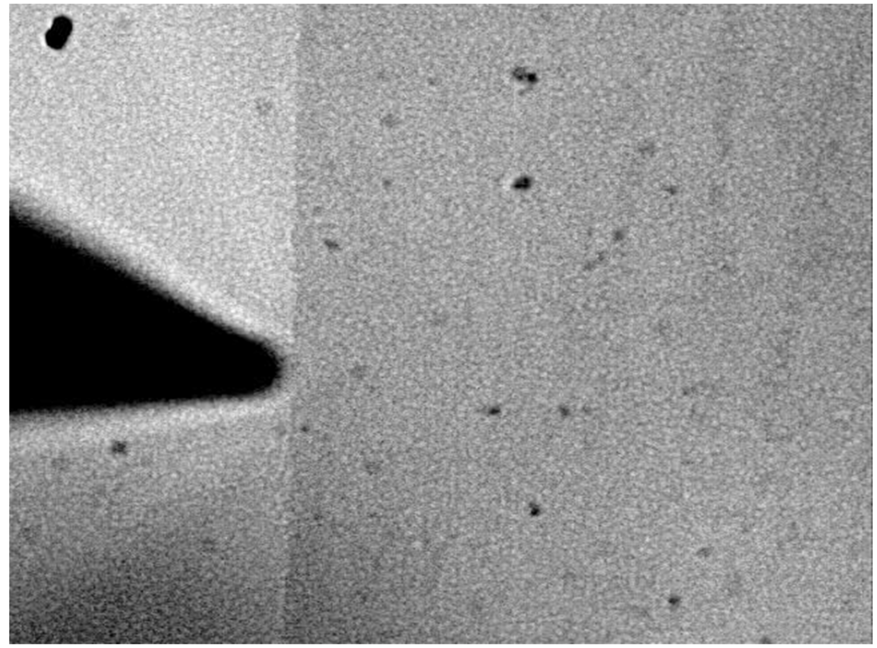
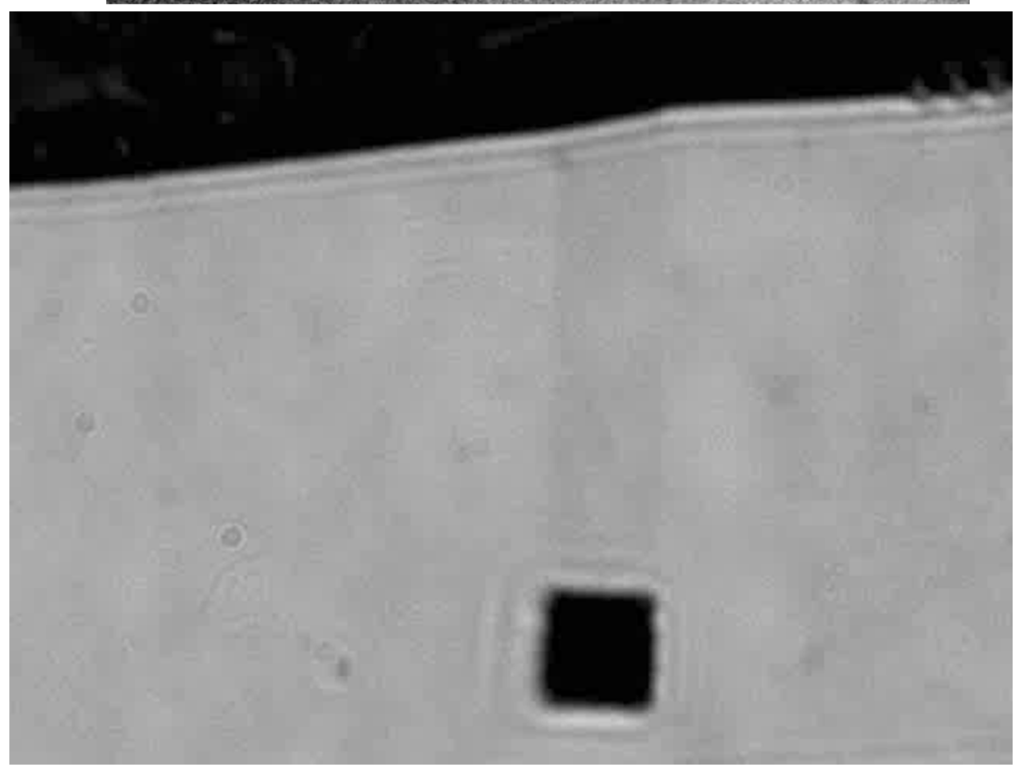






Critical phenomena without critical points: Theory of free-standing graphene ribbons

Luca Peliti
Mehran Kardar
Yantor Kantor

Nonlinear equations of thin plate theory

--nonlinear bending and stretching energies

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

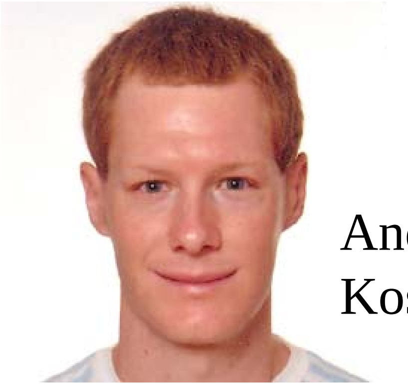
Recent experiments:
Paul McEuen group
(Cornell)

Remarkable effect of thermal fluctuations

-- entire low temperature flat phase characterized by critical fluctuations; "self-organized criticality"

--strongly scale-dependent bending elastic parameters

Recent theory:
Mark Bowick
Rastko Sknepnek &



Andrej
Kosmrlj

Graphene (and BN, MoS_2, WS_2, …?)

-- vK ~ 10^{13}*! "Moore's Law limit of thinness…*

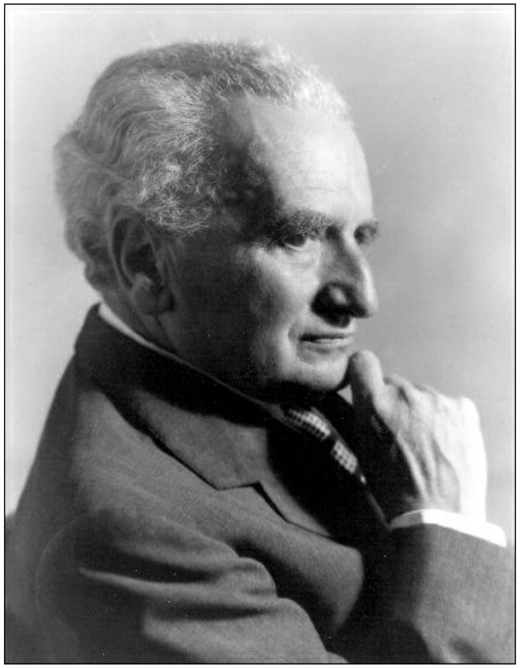
--Thermal fluctuations dominate for $l > l_{th} = 0.15nm$*…*

-- Bending rigidity at room temperature enhanced 6000-fold

-- Anomalous properties of ribbons, crumpling transition, etc.

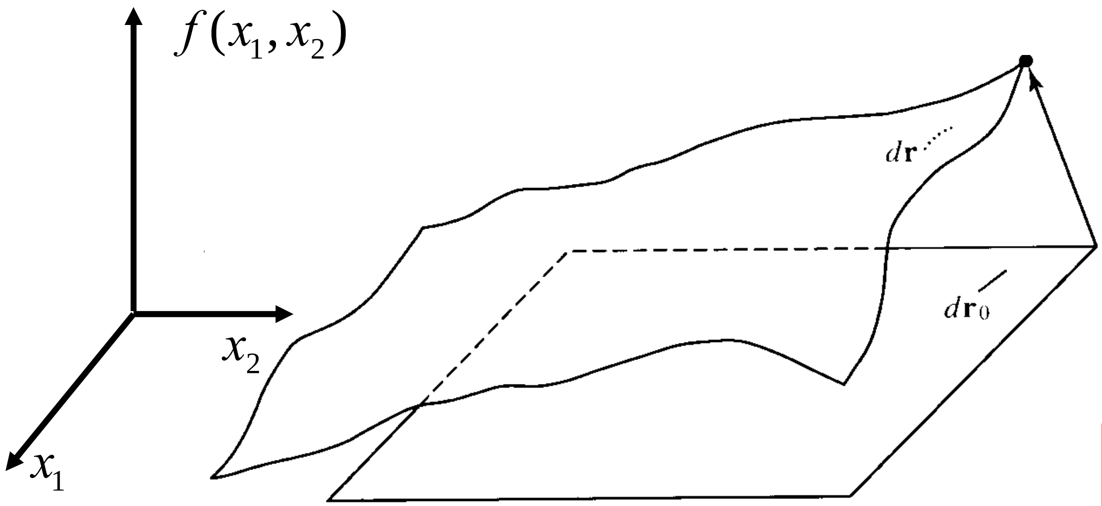
Truly ancient history: in 1904, Föppl & von Kármán studied large deflections of elastic plates

August Föppl (1854-1924) Pioneer of elasticity theory



Theodore von Kármán (1881-1963) Hungarian-American physicist & aeronautical engineer

To study deformed surfaces, expand about a flat reference state…



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

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

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

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

$\kappa =$ bending rigidity

$\mu =$ shear modulus

$\mu + \lambda =$ bulk modulus

Flexural phonons can escape softly into the 3rd dimension…

Nonlinear Föppl -von Karman Equations (1905)

$$\partial_i \sigma_{ij} = 0 \;\Rightarrow\; \sigma_{ij}(\vec{x}) = 2\mu u_{ij}(\vec{x}) + \lambda \delta_{ij} u_{kk}(\vec{x}) \equiv \varepsilon_{im}\varepsilon_{jn}\partial_m \partial_n \chi(\vec{x})$$

$\chi(\vec{x}) =$ Airy stress function

Bending modes $f(\vec{x})$ coupled to stretching modes $\vec{u}(\vec{x})$;

Minimize energy over $f(\vec{x})$ and $\chi(\vec{x})$...

$$\kappa \nabla^4 f = \frac{\partial^2 \chi}{\partial y^2}\frac{\partial^2 f}{\partial x^2} + \frac{\partial^2 \chi}{\partial x^2}\frac{\partial^2 f}{\partial y^2} - 2\frac{\partial^2 \chi}{\partial x \partial y}\frac{\partial^2 f}{\partial x \partial y}$$

Young's modulus $Y = \dfrac{4\mu(\mu+\lambda)}{2\mu+\lambda}$

$$\frac{1}{Y}\nabla^4 \chi = -\frac{\partial^2 f}{\partial x^2}\frac{\partial^2 f}{\partial y^2} + \left(\frac{\partial^2 f}{\partial x \partial y}\right)^2 = \text{ Gaussian curvature}$$

bending rigidity κ

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bending rigidity κ

$\Rightarrow$ dimensionless "Foeppl-von Karman number" $\nu K = YL^2/\kappa \gg 1$ ($L =$ linear dimension)

(compare Reynold's number $\mathrm{Re} = uL/\nu$ in fluid mechanics)

$\Rightarrow$ resembles a simplified form of general relativity (developed 10 years later....)

$\Rightarrow$ exact solutions available only in very special cases

Applications: thin solid shells and structures

macroscopic (1cm - 10m)

plant
leaves

egg
shell

aluminum foil

pipes

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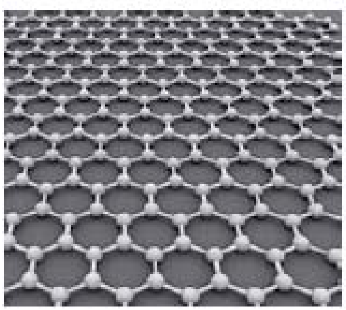
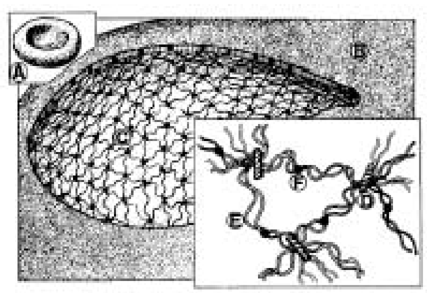
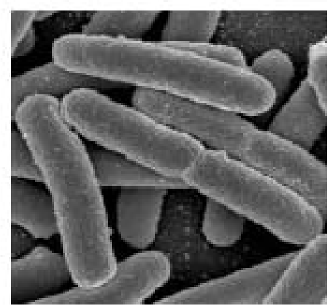
& microscopic (0.1nm - 1µm, but what about Brownian motion??)



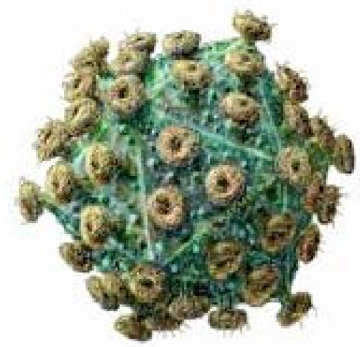
graphene



cell membrane
with cytoskeleton



bacterial
cell wall



viral
capsid

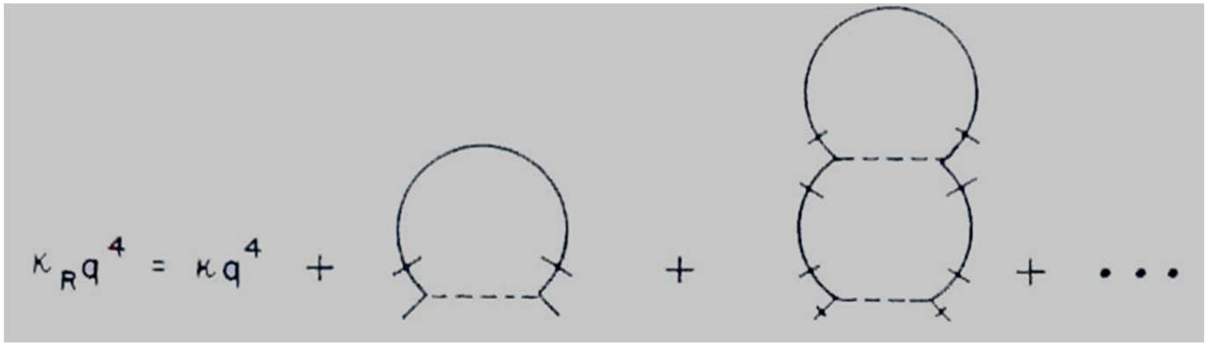
What About Thermally Excited Membranes? *(L. Peliti & drn)*

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

$$F_{\text{eff}} = -k_B T \ln\left(\int D\{u_x(x,y)\} \int D\{u_y(x,y)\} e^{-E/k_B T} \right)$$

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

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



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

$$\approx \kappa[1 + (vK) k_B T / (4\pi^3 \kappa) + \ldots]$$

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

L = membrane size

h = membrane thickness

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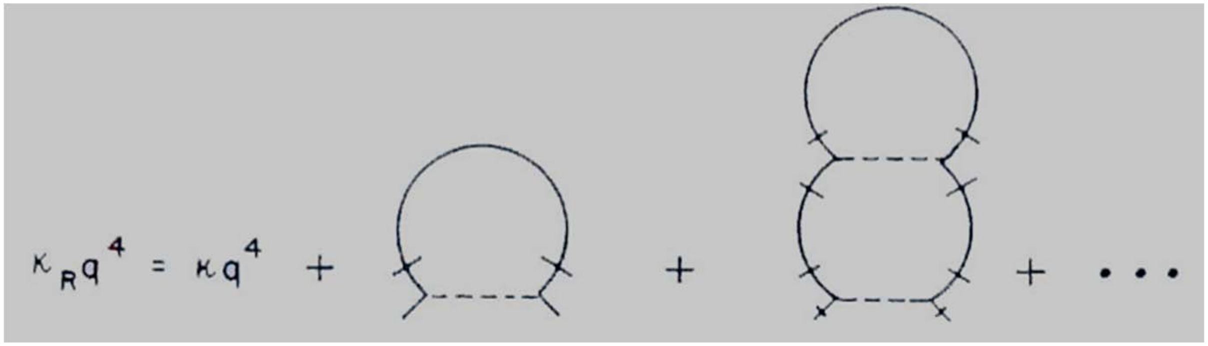

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$vK = YL^2/\kappa \approx (L/h)^2 \gg 1$

$L =$ membrane size

$h =$ membrane thickness

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

Renormalization Group for Thermally Excited Sheets

L. Peliti & drn (~1987)
J. Aronovitz and T. Lubenksy
P. Le Doussal and L. Radzihovsky

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

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

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

define running coupling constants....

$$\bar{\mu}(l) = k_B T \mu a_0^2 / \kappa^2; \qquad \bar{\lambda}(l) = k_B T \lambda a_0^2 / \kappa^2$$

scale dependent Young's modulus

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

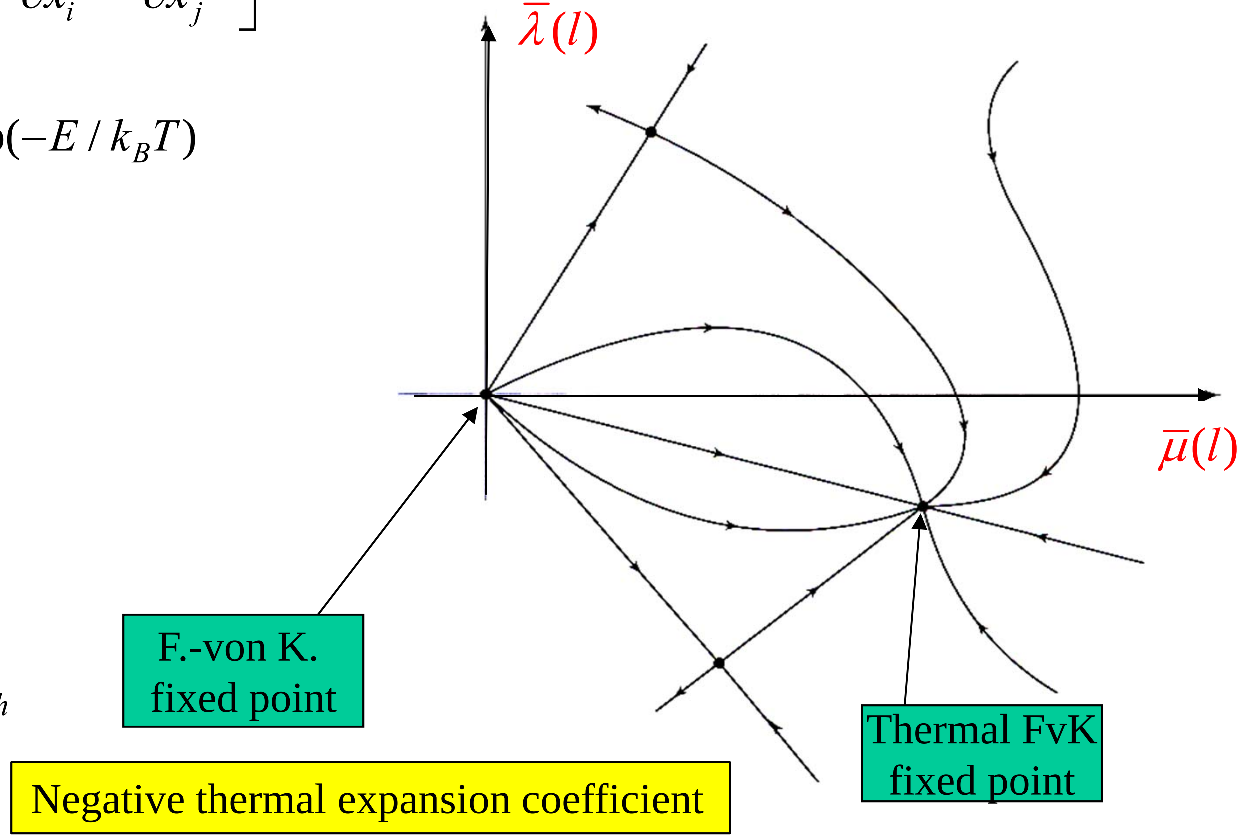
$$\kappa_R(l) \approx \kappa(l / l_{th})^{\eta}$$

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

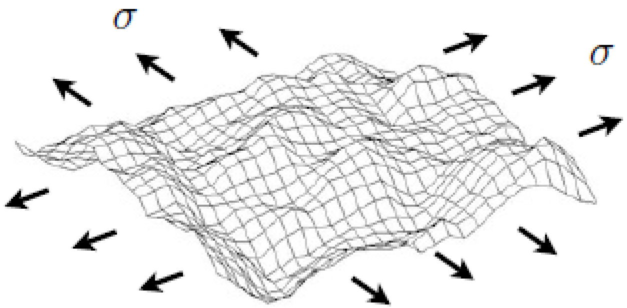
$$\eta \approx 0.82, \quad \eta_u \approx 0.36$$

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

$$l_{th} \approx \sqrt{\kappa^2 / (k_B T Y)}$$



Negative coefficient of thermal expansion and nonlinear stress strain curves



Out of plane fluctuations suppressed by an external tension σ

$$\langle |f(\mathbf{q})|^2 \rangle = \frac{k_B T}{\sigma q^2 + \kappa q^{4-\eta}/\ell_{\rm th}^{\eta}}$$

$$\eta \approx 0.82 \qquad \ell_{\rm th} \sim \kappa/\sqrt{k_B T Y}$$

E Guitter, F David, S Leibler, L Peliti, PRL **61**, 2949 (1988)

Membrane area extension

$$\left\langle \frac{\delta A}{A} \right\rangle \approx \underbrace{-\frac{k_B T}{4\pi\kappa}\left[\eta^{-1} + \ln(\ell_{\rm th}\Lambda)\right]}_{\text{shrinking due to fluctuations}} + \underbrace{\frac{k_B T}{4\pi\kappa}\left[\eta^{-1} - \frac{1}{2}\right]\left(\frac{\ell_{\rm th}^2 \sigma}{\kappa}\right)^{\eta/(2-\eta)}}_{\text{non-linear response like in critical phenomena}} \underbrace{+\frac{\sigma}{(\mu+\lambda)}}_{\text{material stretching}}$$

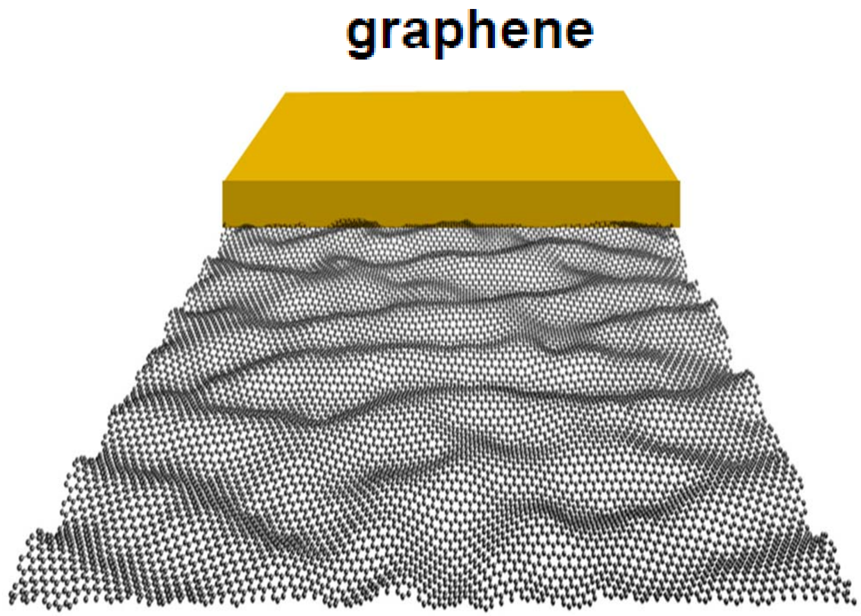
$$\eta/(2-\eta) \approx 0.7$$

For large tension, $\sigma \gtrsim k_B T Y/\kappa$,
thermal fluctuations become irrelevant!

Freely supported graphene is an ideal test bed…

Graphene is the ultimate 2D crystalline membrane:

- One atom thick
- Very stiff in-plane (Young's modulus Y = 500 GPa)



(atomistic calculations)

$\kappa_0 \approx 1.2\mathrm{eV} \approx 2 \times 10^{-19} J$

Extremely flexible!

R. Nicklow, N. Wakabayashi and H. G. Smith, PRB 5, 4951 (1972)

With graphene, we have reached the "Moore's Law" limit of large Foppl-von Karman numbers

$L = 10\mu,\ h = 3\ A,\ vK = 10^{13}$ *!!*

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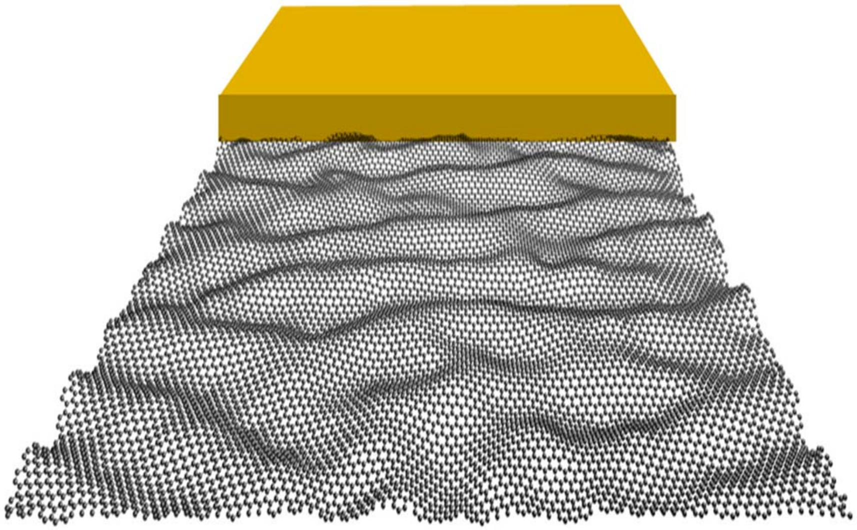

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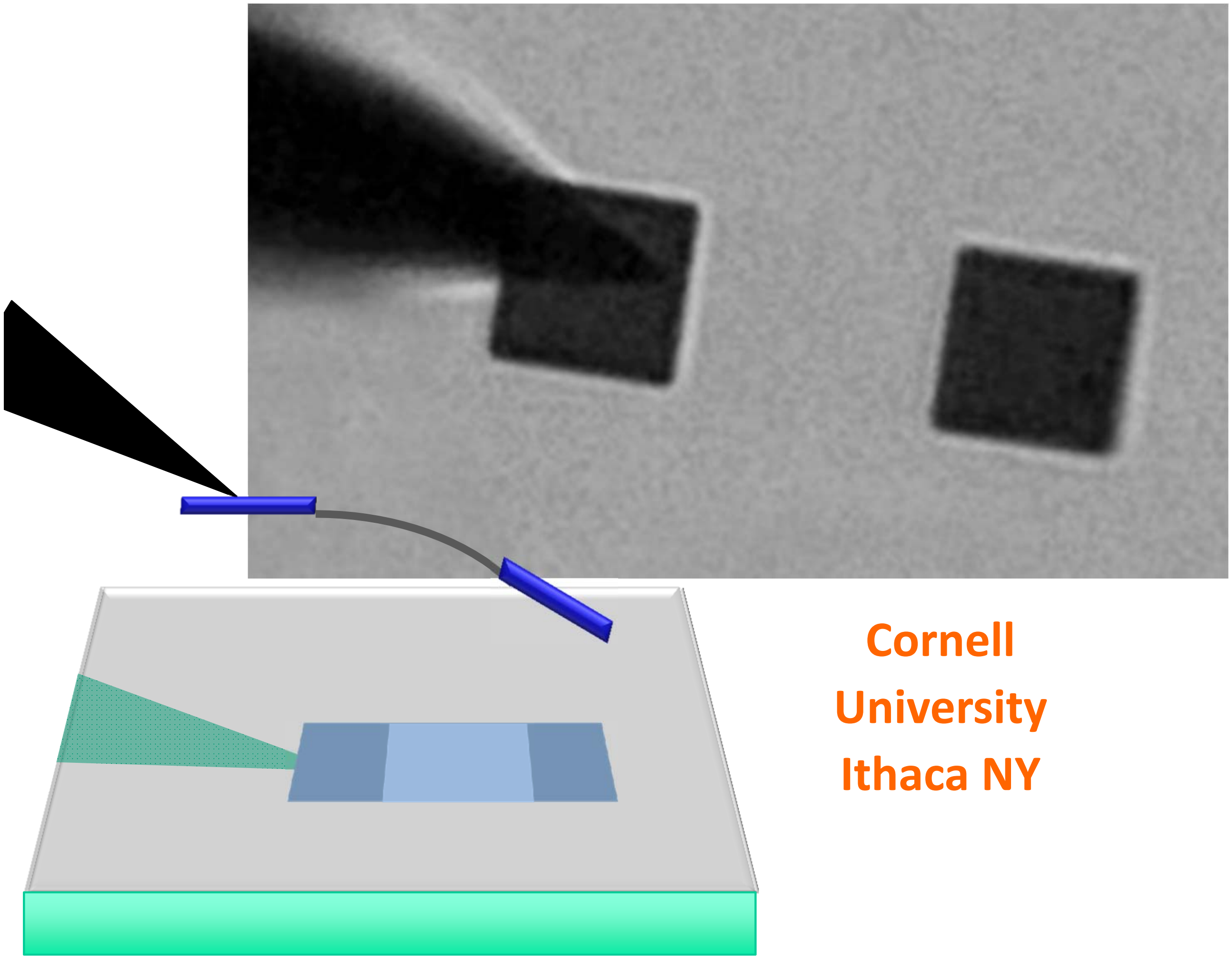
$$L = 10\mu,\ h = 3\ A,\ \ vK = 10^{13}\ !!$$

fluctuations dominate for $L > l_{th}$

$$l_{th} \approx \sqrt{\kappa^2 / (k_B TY)} \approx 0.2\text{nm!}$$

Graphene cantilever experiment



Cornell
University
Ithaca NY

Melina Blees, Arthur Barnard, Samantha Roberts, Josh Kevek, Alex Ruyack, Jenna Wardini, Peijie Ong, Aliaksandr Zaretski, Si Ping Wang, and Paul L. McEuen

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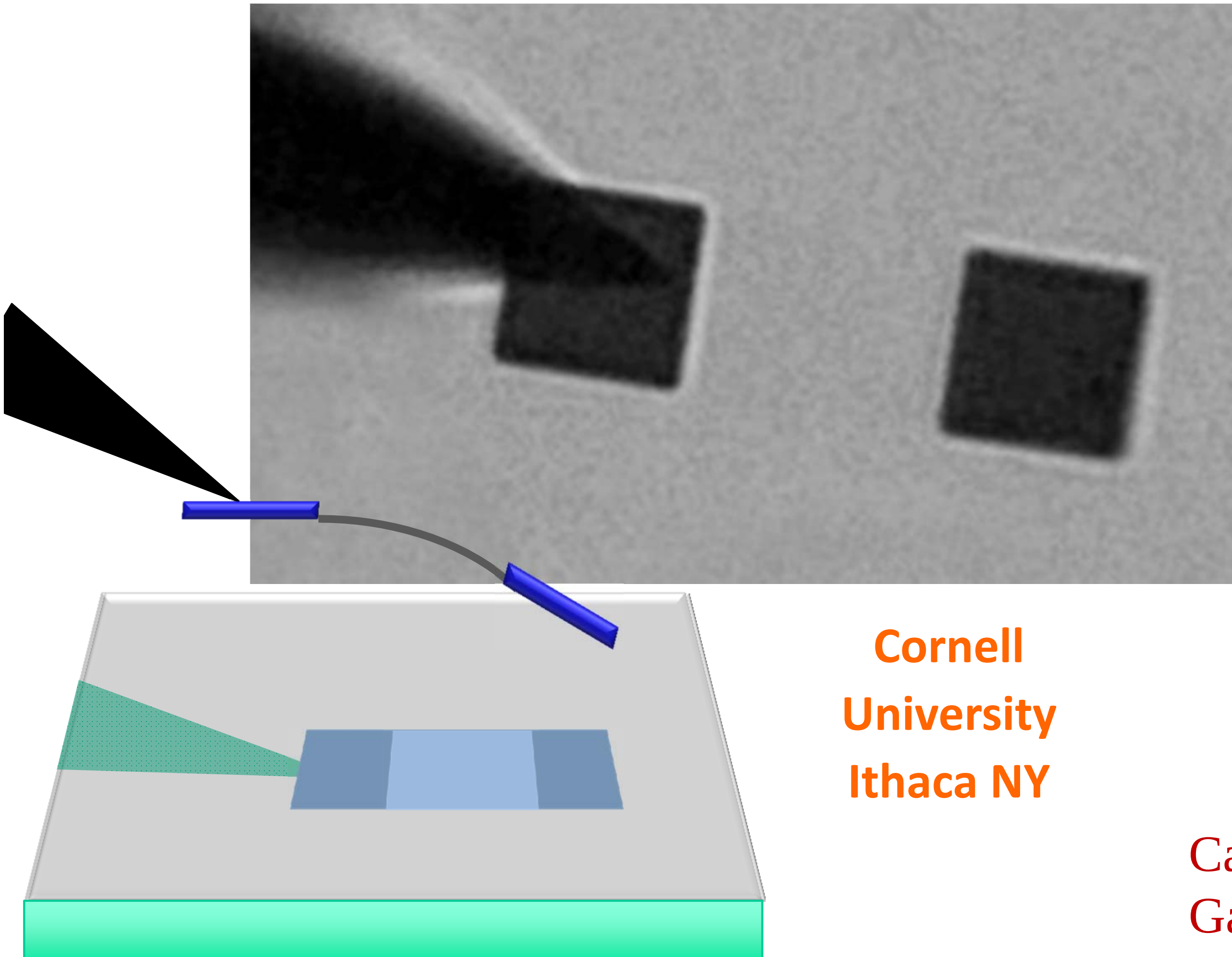


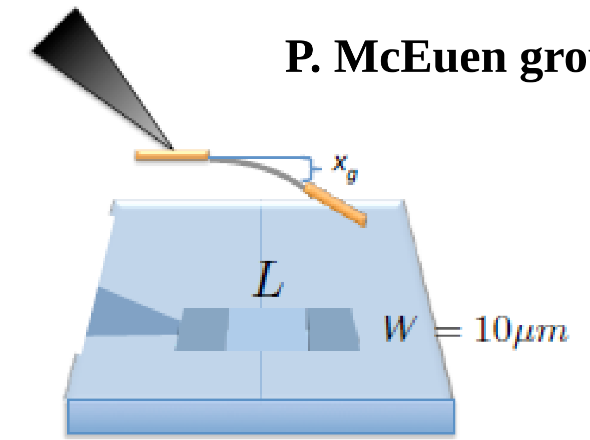
Cornell
University
Ithaca NY

Cantilever experiment
Galileo Galilei (1638)
"Discourses and Mathematical Demonstrations Relating to Two New Sciences"

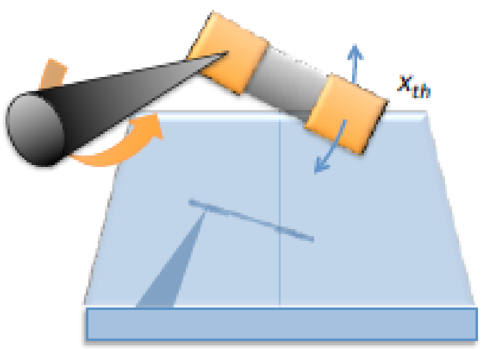
Melina Blees, Arthur Barnard, Samantha Roberts, Josh Kevek, Alex Ruyack, Jenna Wardini, Peijie Ong, Aliaksandr Zaretski, Si Ping Wang, and Paul L. McEuen

Bending rigidity of graphene membranes

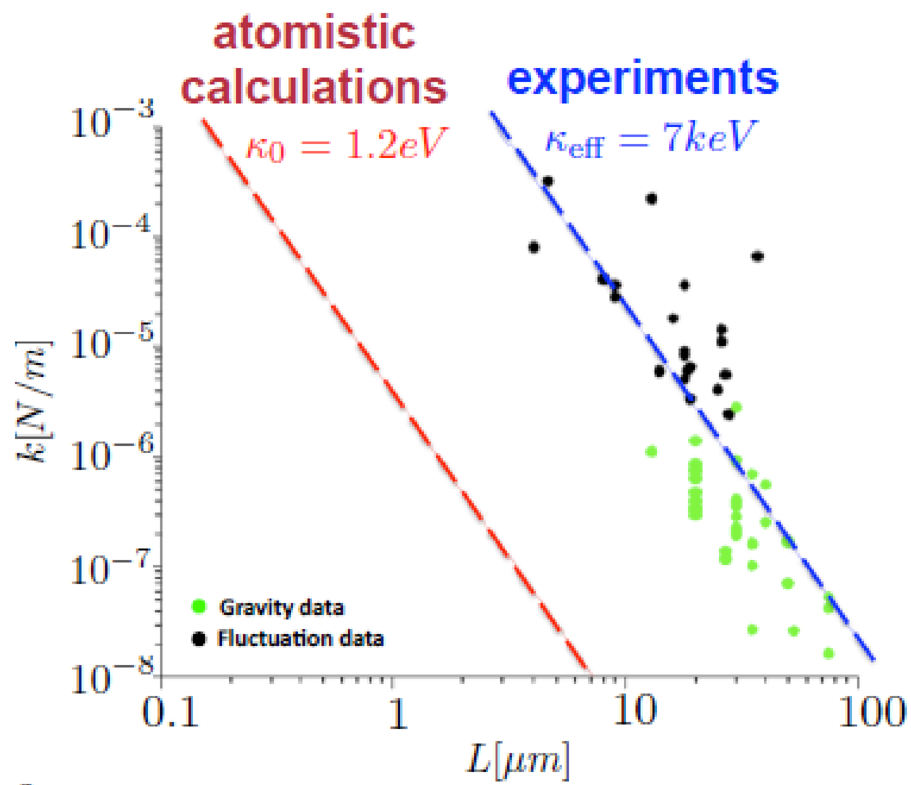
P. McEuen group, preprint



$$mg = kx_g$$



$$k_B T = \frac{1}{2} k x_{th}^2$$



$$WL \times \kappa_{\text{eff}} \times (x/L^2)^2 \sim (\kappa_{\text{eff}} W/L^3) x^2 = kx^2$$

plate area	bending rigidity	(curvature)^2	bending energy

$$k = 3\kappa_R W / L^3$$

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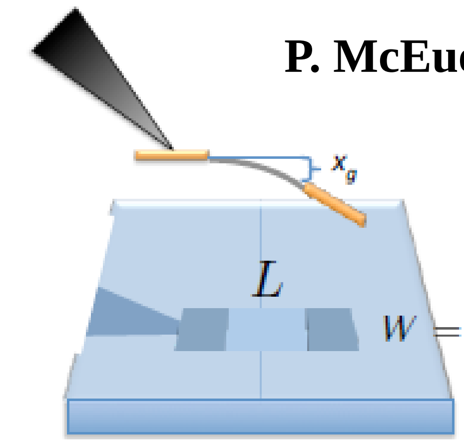
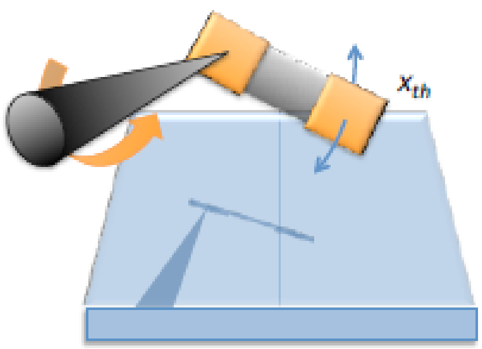


$W = 10\mu m$

$$mg = kx_g$$



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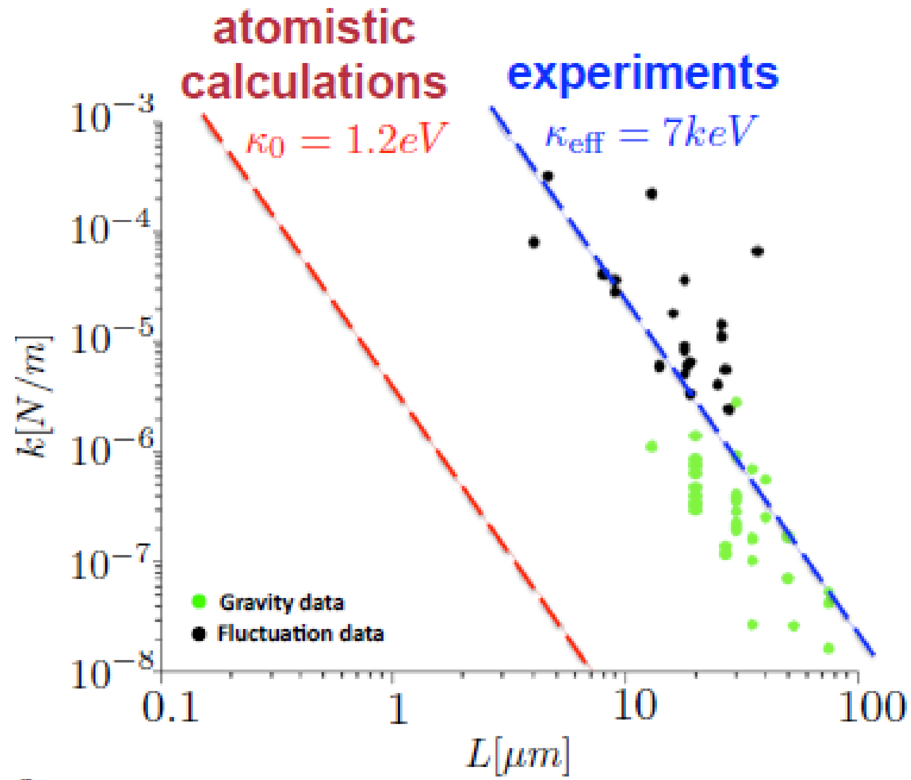

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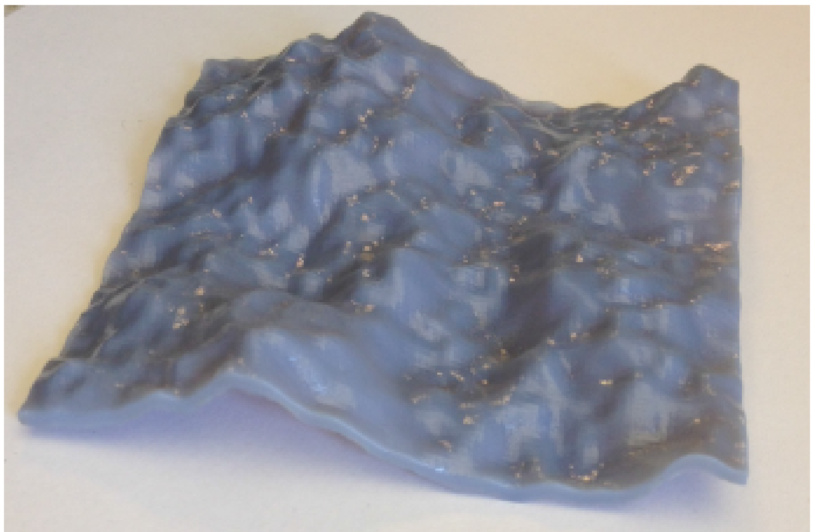
plate area	bending rigidity	(curvature)^2	bending energy

$$k = 3\kappa_R W / L^3$$

Bending rigidity enhanced 6000 fold.
Agrees with $\kappa_R(l) \approx \kappa(W/l_{th})^{0.8}$
($l_{th} \sim$ 0.2nm for graphene)

Frozen fluctuations in nearly flat membranes

A. Košmrlj and D. Nelson, PRE **88**, 012136 (2013)
A. Košmrlj and D. Nelson, PRE **89**, 022126 (2014)



frozen height fluctuations $h_{\text{eff}}^2 = \frac{1}{A}\int dA \, \langle h^2 \rangle$

thickness t

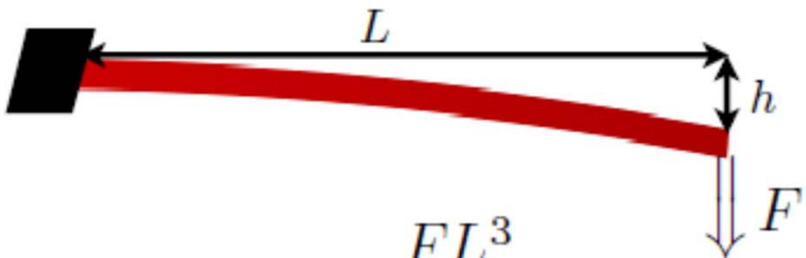
Linear response properties averaged over frozen disorder

$$\langle \kappa_{\text{eff}} \rangle / \kappa \sim \sqrt{(Y h_{\text{eff}}^2)/\kappa} \sim h_{\text{eff}}/t$$

$$\langle Y_{\text{eff}} \rangle / Y, \langle \mu_{\text{eff}} \rangle / \mu \sim \sqrt{\kappa/(Y h_{\text{eff}}^2)} \sim t/h_{\text{eff}}$$

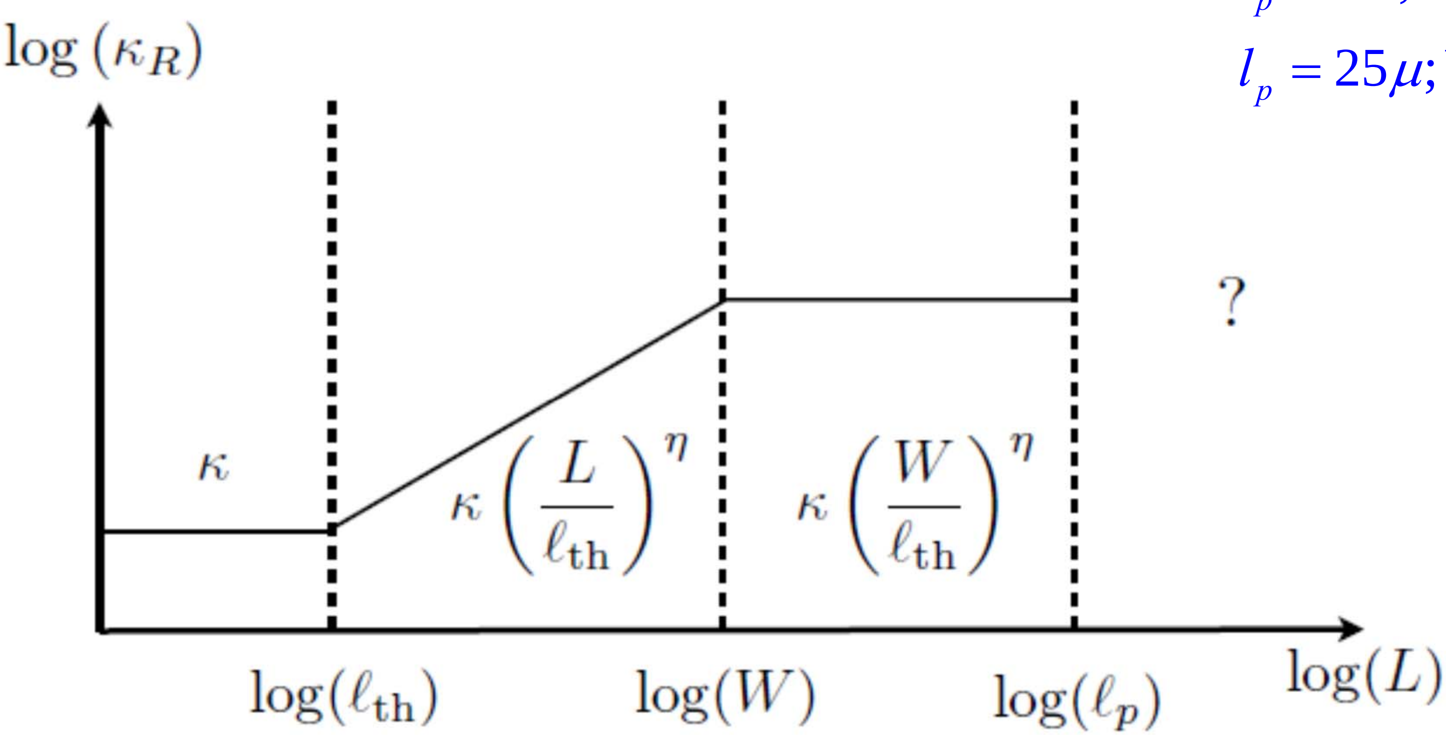
Effective bending rigidity for ribbons

L = ribbon length
W = ribbon width
l_p= *persistance length*



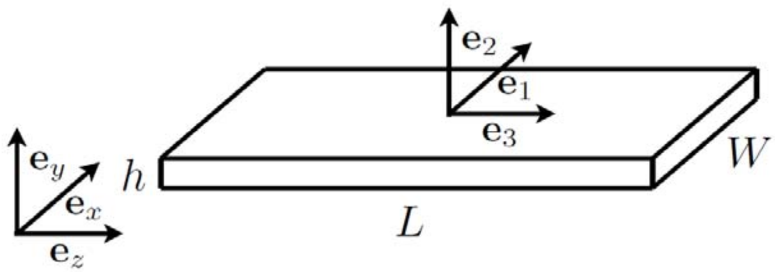
$$\langle h\rangle = \frac{FL^3}{3W\kappa_R(W,L)}$$

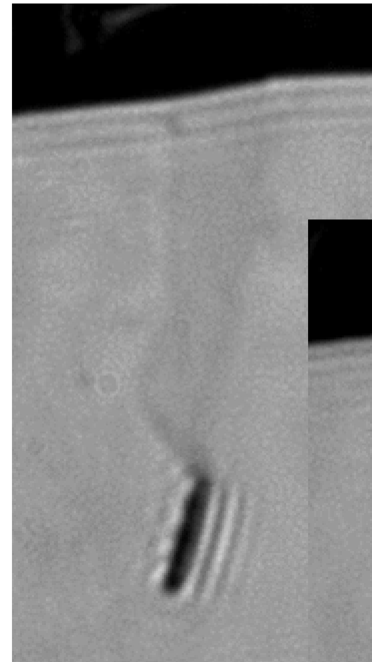
$l_p = 5m$; W = 10μ, but
$l_p = 25\mu$; W = 10nm!

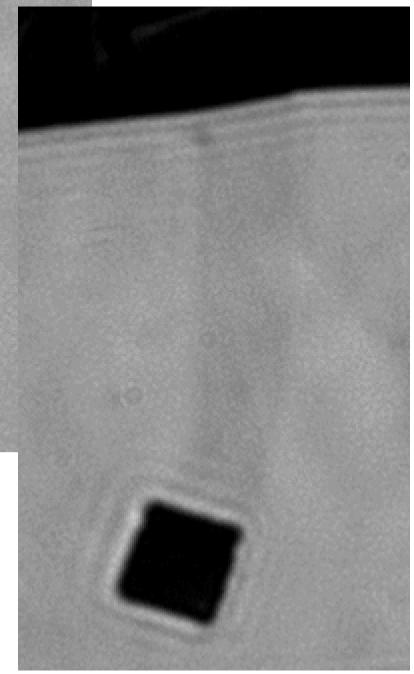


Theory of thermalized cantilever ribbons (A. Kosmrlj and drn)

$$\frac{d\mathbf{e}_i}{ds} = \mathbf{\Omega} \times \mathbf{e}_i$$







McEuen group

$$E = \int \frac{ds}{2}\left[A_1\Omega_1^2 + A_2\Omega_2^2 + C\Omega_3^2 \right] - Fz$$

$$A_1 = \kappa W(1-\nu^2)/12;\ \mathrm{C} = 2\kappa W(1-\nu)$$

$$A_2 = YW^3/12 \gg A_1, C$$

E = 2d Young's modulus
μ = 2d shear modulus
ν = 2d Poisson ratio

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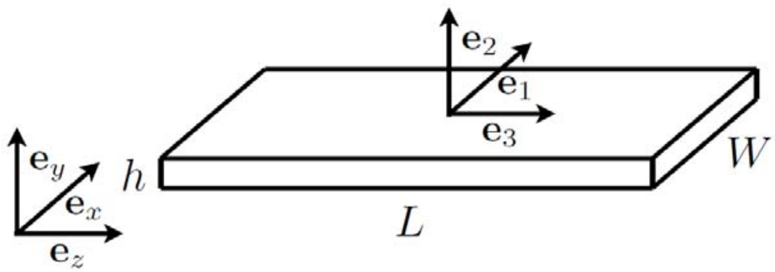

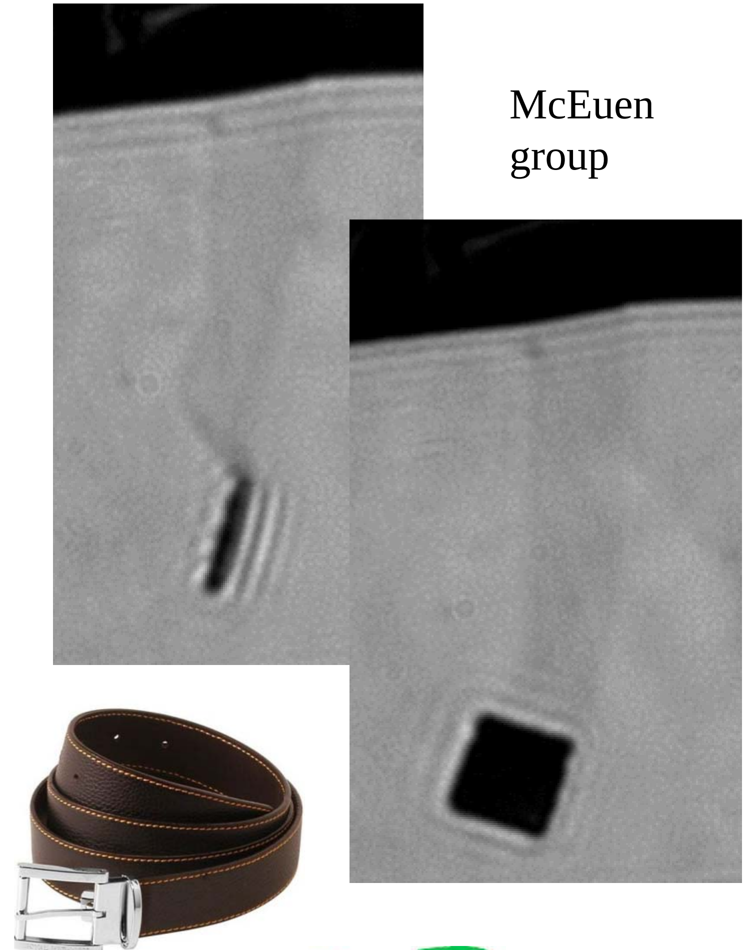

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Y = 2d Young's modulus
κ = 2d bending rigidity
ν = 2d Poisson ratio

Map 1d path integral statistical mechanics onto the quantum mechanics of a rigid rotor in an external gravitational field

Pulling and bending of ribbons of varying lengths

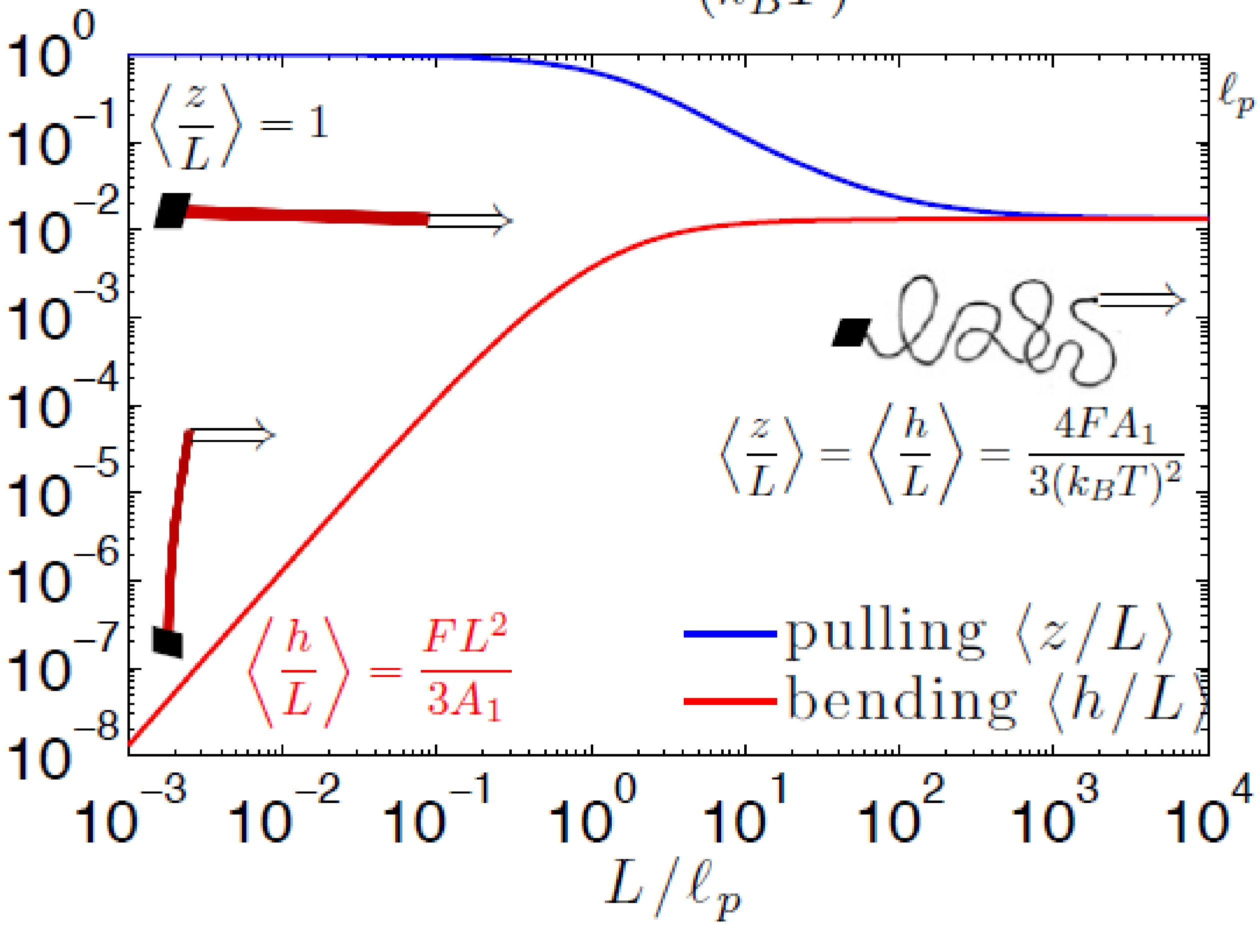
Fixed force, temperature and ribbon width

$$\frac{FA_1}{(k_BT)^2} = 0.01$$

$$C = A_1$$

$$A_2/A_1 \to \infty$$

$$\ell_p \approx \frac{2A_1}{k_BT} \sim \frac{\kappa W^{1+\eta}}{k_BT\ell_{\rm th}^{\eta}}$$

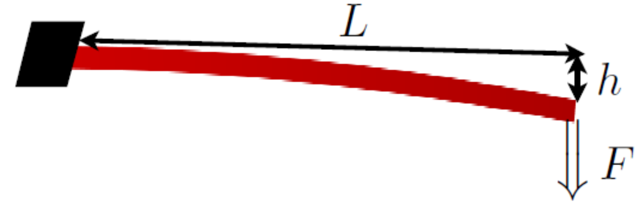


For long ribbons direction of pulling force is irrelevant!

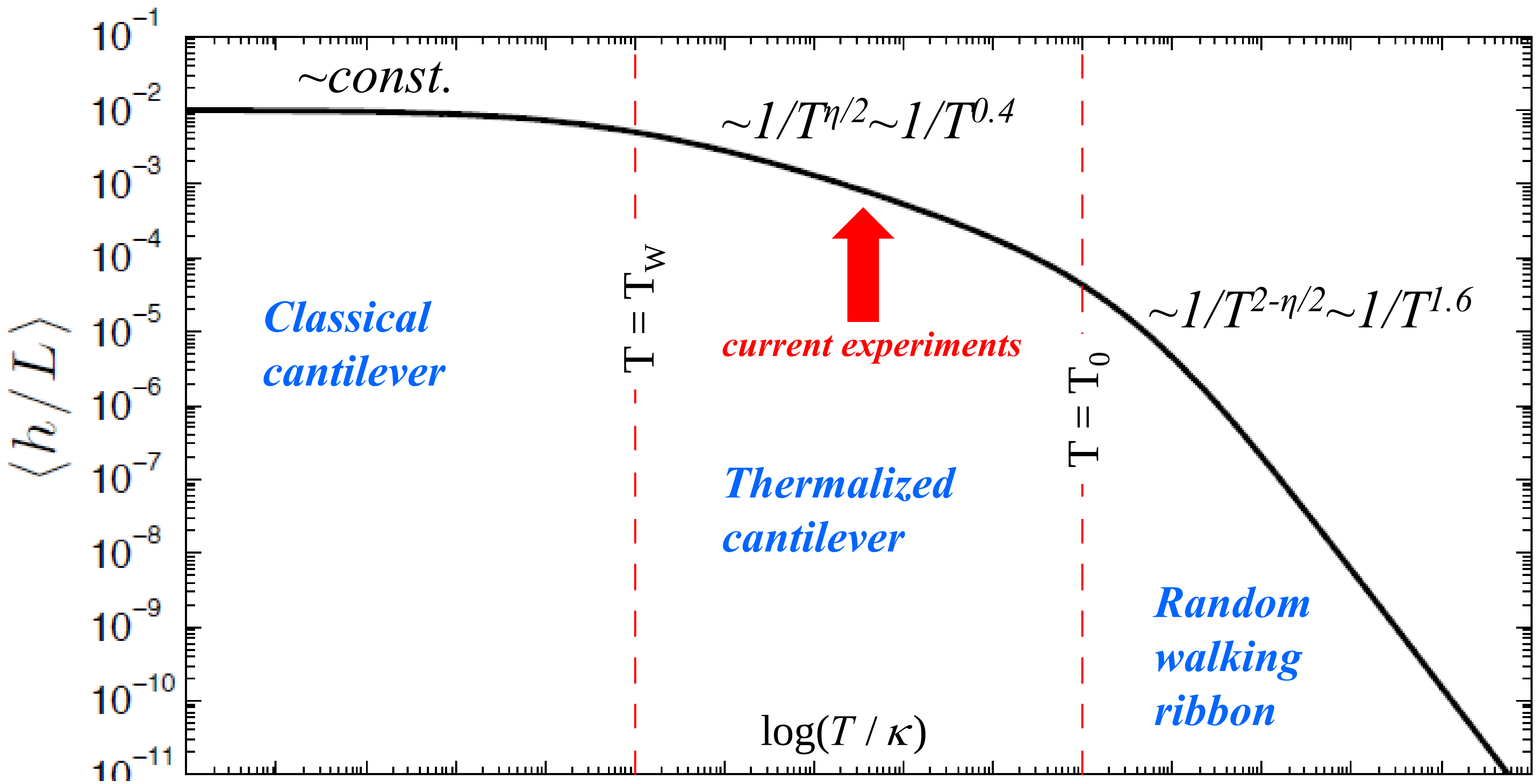
Temperature dependence of cantilever deflection

$$l_{th}(T) = \sqrt{4\pi^3\kappa^2 / (k_B T Y)}$$

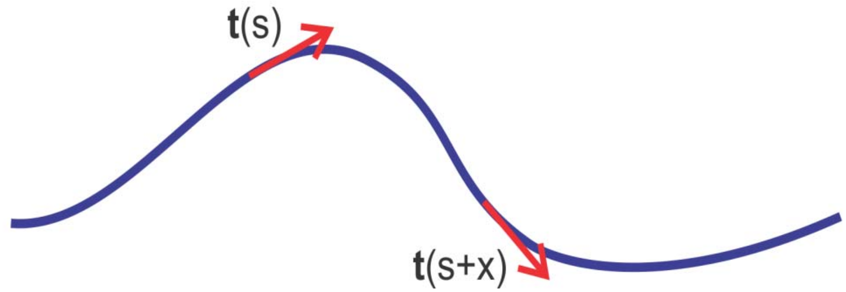
$$l_p(T) = \frac{\kappa}{k_B T}\left(\frac{W}{l_{th}(T)}\right)^{\eta} \quad W \gg l_{th}(T)$$

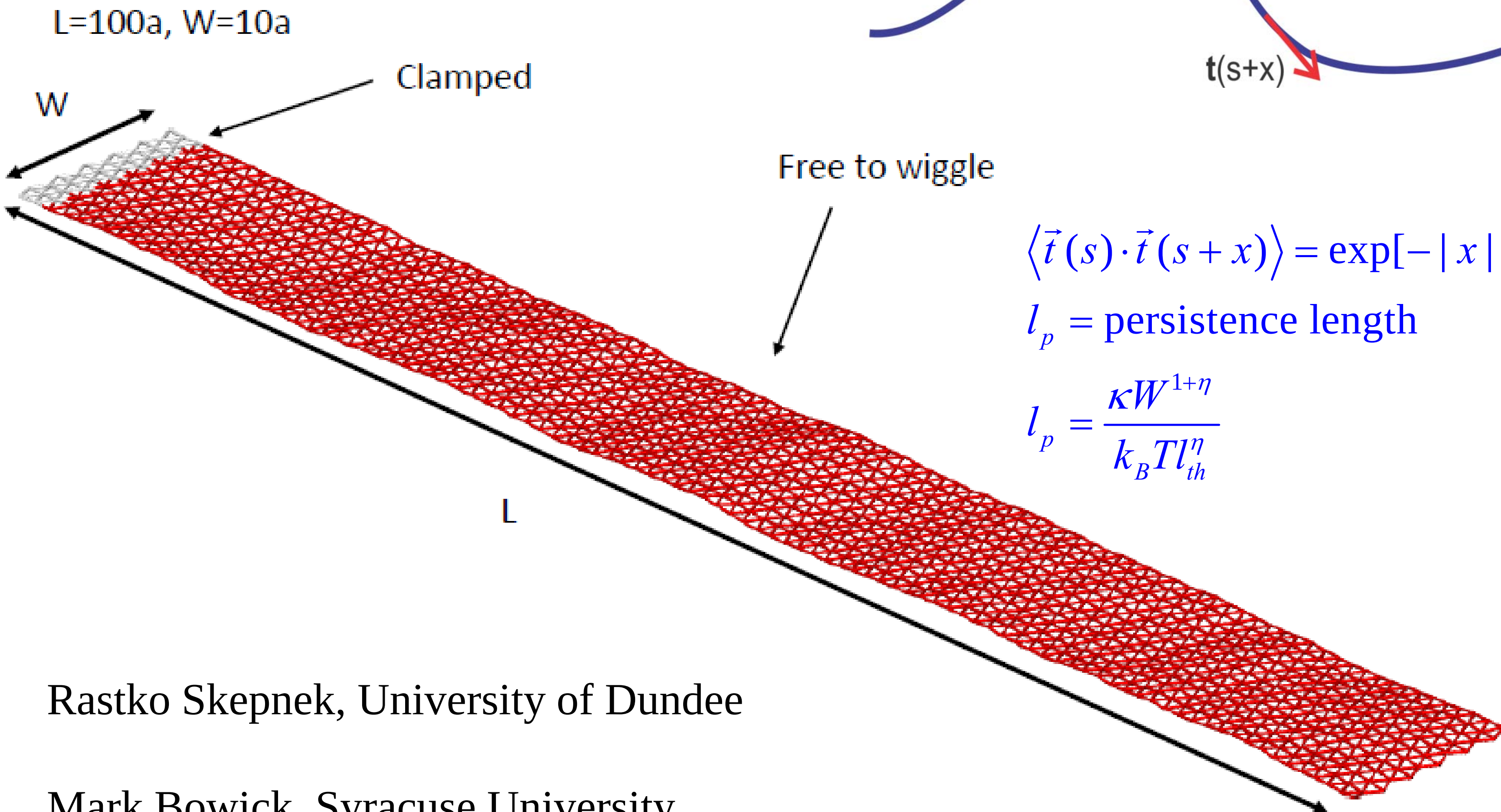


Above T_0, L exceeds the persistence length
Below T_W, the thermal length exceeds W



Computer simulations of graphene ribbons





$$\langle \vec{t}\,(s)\cdot\vec{t}\,(s+x)\rangle = \exp[-|\,x\,|\,/l_p]$$

l_p = persistence length

$$l_p = \frac{\kappa W^{1+\eta}}{k_B T l_{th}^{\eta}}$$

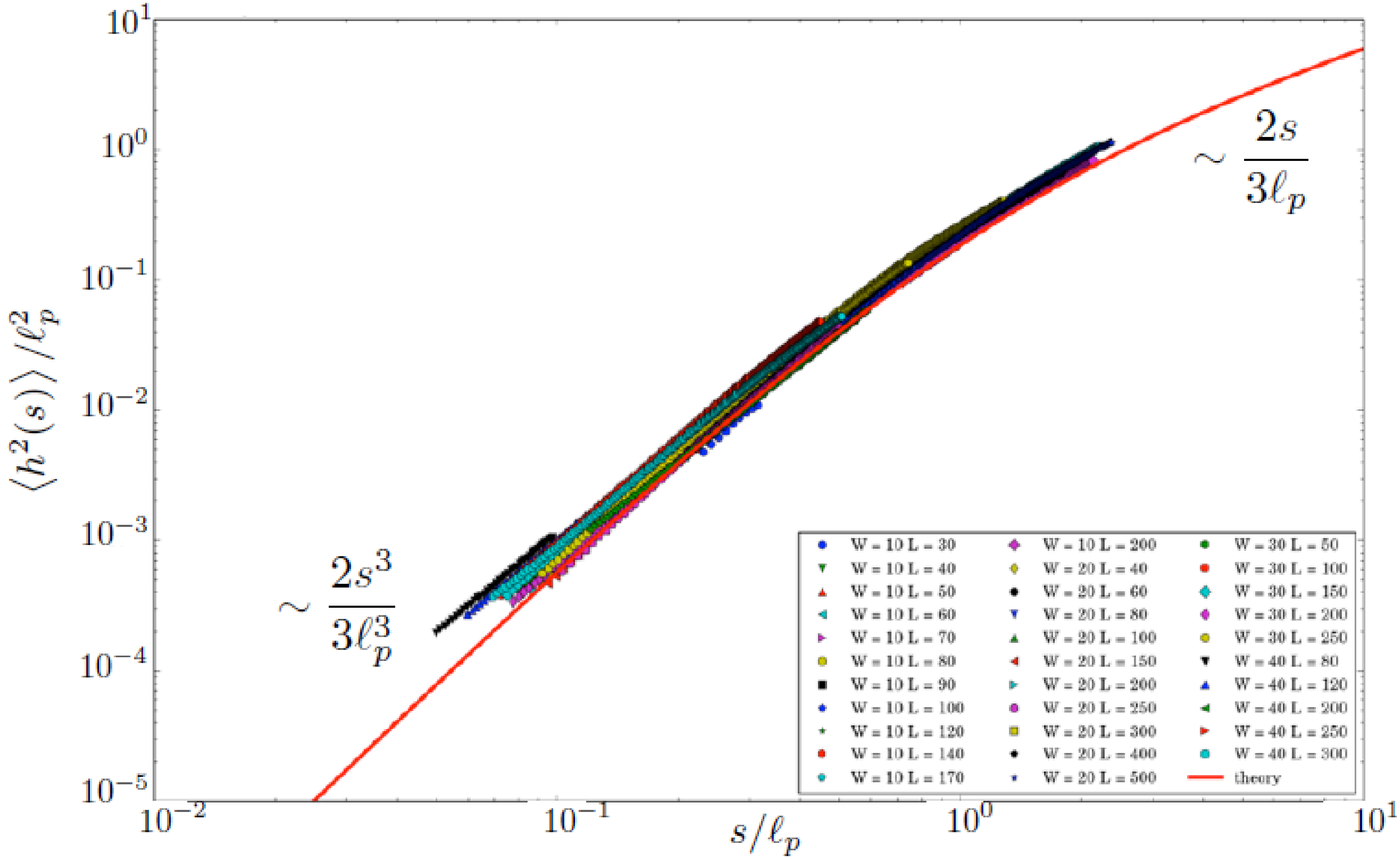
Rastko Skepnek, University of Dundee

Mark Bowick, Syracuse University

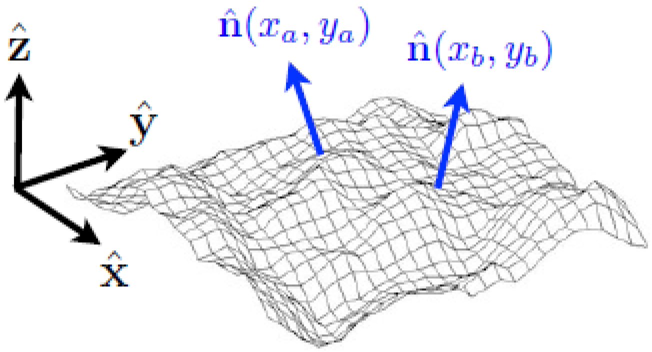
Molecular dynamics simulations of ribbons

Ribbon fluctuations from molecular dynamics simulations

Data collapse without adjustable parameters!



Future directions: flat vs. crumpled phases



$$\hat{\mathbf{n}}(x,y) = \frac{1}{\sqrt{1+(\nabla f)^2}} \begin{pmatrix} -\partial_x f \\ -\partial_y f \\ 1 \end{pmatrix}$$

$$\kappa_R(q) \sim \kappa/(q\ell_{\rm th})^\eta \qquad \ell_{\rm th} \sim \kappa/\sqrt{k_B T Y}$$

$$\eta \approx 0.82$$

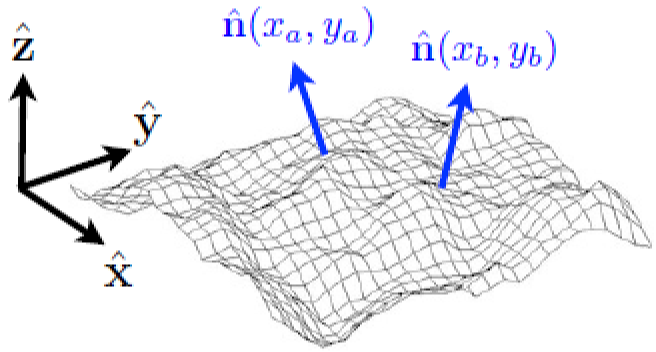
$$\langle \hat{\mathbf{n}}(\mathbf{r}_a)\cdot\hat{\mathbf{n}}(\mathbf{r}_b)\rangle = 1 - \int \frac{d^2\mathbf{q}}{(2\pi)^2}\frac{k_B T}{\kappa_R(q) q^2}\left[1 - e^{i\mathbf{q}\cdot(\mathbf{r}_b - \mathbf{r}_a)}\right]$$

$$\langle \hat{\mathbf{n}}(\mathbf{r}_a)\cdot\hat{\mathbf{n}}(\mathbf{r}_b)\rangle \approx 1 - \frac{k_B T}{2\pi\kappa}\left[\eta^{-1} + \ln\left(\frac{\ell_{\rm th}}{a_0}\right)\right] + C\frac{k_B T}{\kappa}\left(\frac{\ell_{\rm th}}{|\mathbf{r}_b - \mathbf{r}_a|}\right)^\eta$$

Normal-normal correlations approach constant value at large separation.

OR

Future directions: flat vs. crumpled phases



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Projected Area ~ L^2 low T

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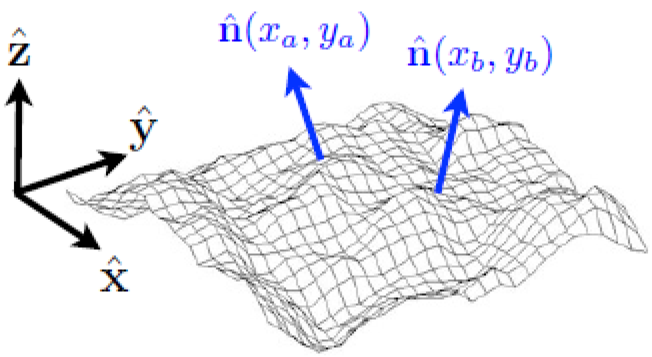

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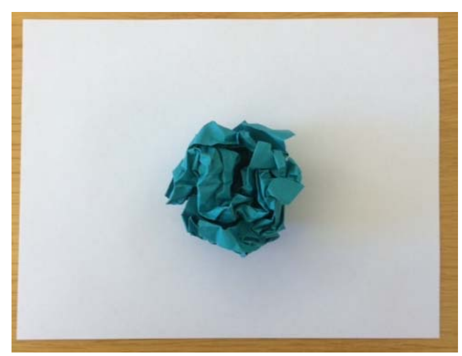
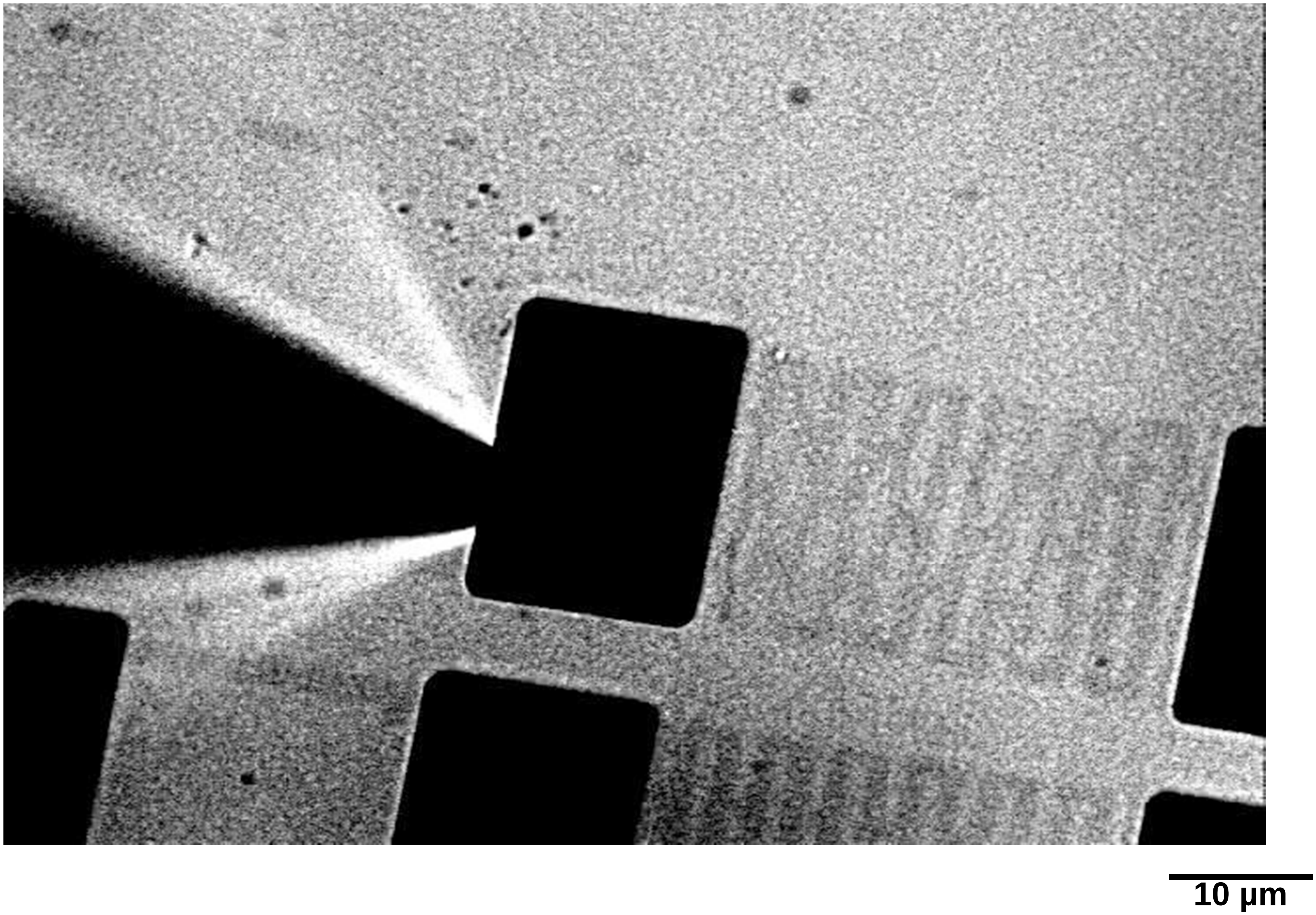
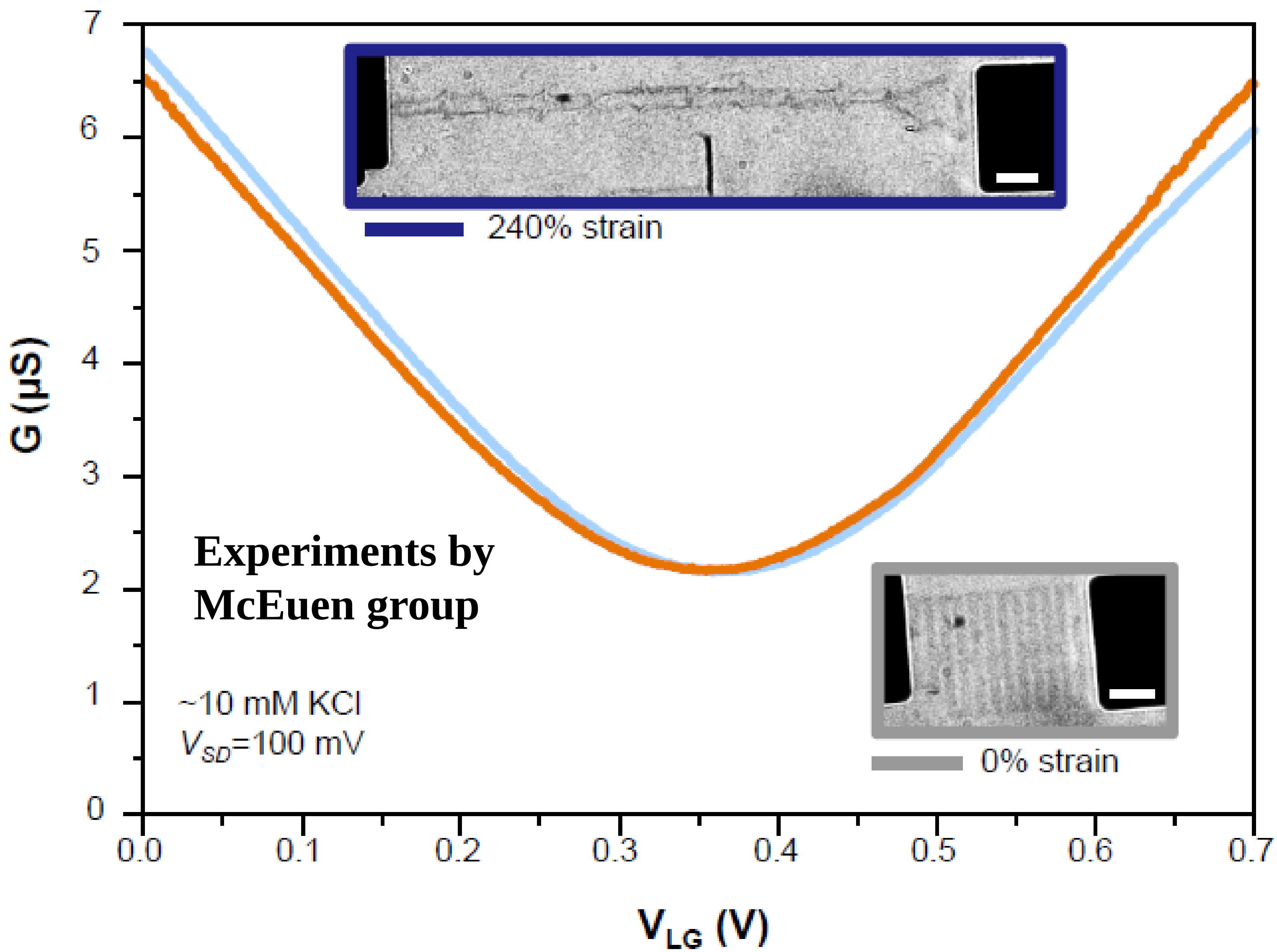


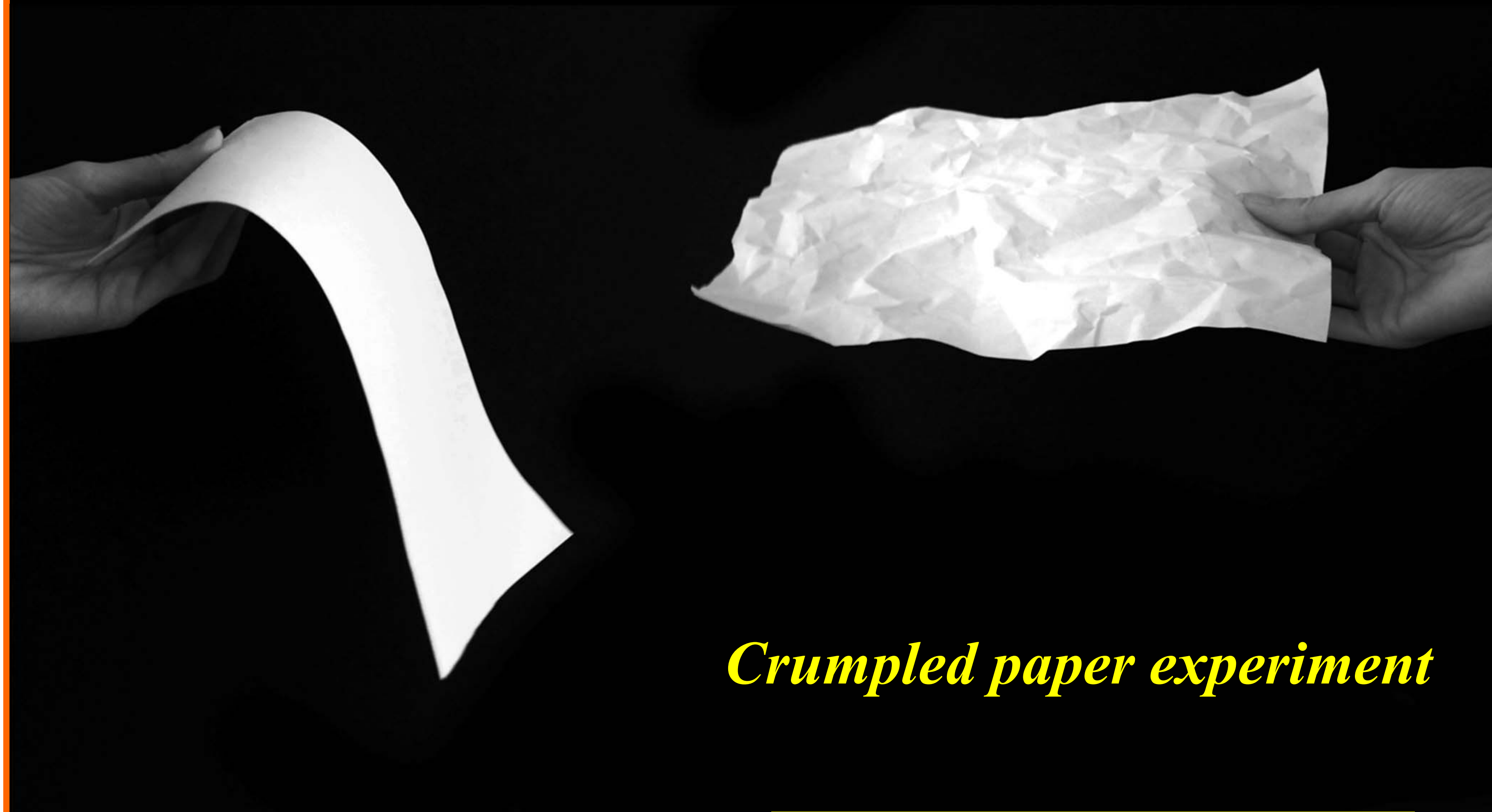
Projected area ~ $L^{8/5}$ high T

Future directions: new, atomically thin springs



Electrical conduction of graphene spring as a function of strain & gate voltage





Crumpled paper experiment

Thank you!!

Figure: Melina Blees, Cornell

Critical phenomena without critical points: Theory of free-standing graphene ribbons

Nonlinear equations of thin plate theory

--nonlinear bending and stretching energies

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

Remarkable effect of thermal fluctuations

-- entire low temperature flat phase characterized by critical fluctuations; "self-organized criticality"

--strongly scale-dependent bending elastic parameters

Graphene (and BN, MoS_2, WS_2 , …?)

-- $vK \sim 10^{13}$*! "Moore's Law limit of thinness…*

--Thermal fluctuations dominate for $l > l_{th} = 0.15nm$*…*

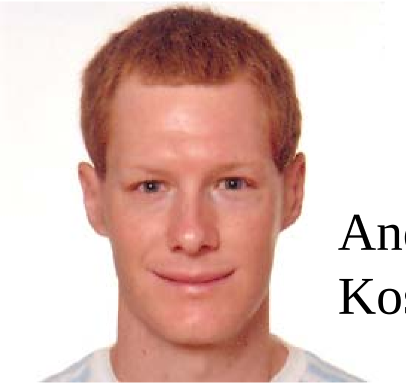
-- Bending rigidity at room temperature enhanced 6000-fold

-- Anomalous properties of ribbons, crumpling transition, etc.

Luca Peliti
Mehran Kardar
Yantor Kantor

Recent experiments:
Paul McEuen group (Cornell)

Recent theory:
Mark Bowick
Rastko Sknepnek &



Andrej Kosmrlj