Thermodynamics of quantum crystalline membranes

Bruno Amorim,
R. Roldán, E. Cappelluti, A. Fasolino, F. Guinea, M. I. Katsnelson

Instituto de Ciencia de Materiales de Madrid (ICMM)
Centro de Física da Universidade do Minho (CFUM)

Correlations, Criticality and Coherence in Quantum Systems
Évora, 6-10 October 2014

amorim.bac@icmm.csic.es

PRB **89**, 224307 (2014)

Fundação para a Ciência e a Tecnologia
MINISTÉRIO DA EDUCAÇÃO E CIÊNCIA

Summary

- Crystalline membranes
- 2D crystals as membranes
- Quantum effects
- Conclusions and current work

Crystalline membranes

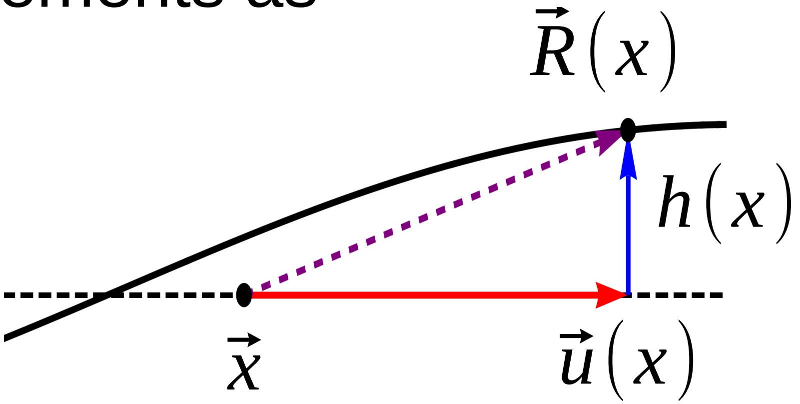
What is a crystalline membrane?

Membrane is a 2D object embedded in 3D space

A **crystalline** membrane resists shear stress

For a **flat membrane** we write displacements as

$$\vec{R}(x) = (\vec{x} + \vec{u}(x), h(x))$$



At quadratic level we have 3 modes

- In-plane longitudinal: $\omega_L(q) \sim q$
- In-plane transverse: $\omega_T(q) \sim q$
- Out-of-plane/flexural: $\omega_F(q) \sim q^2$

Existence of a flat phase

At harmonic level a flat phase is not stable against thermal fluctuations

- No positional order: $\langle h^2 \rangle \sim L^2$
- No orientational order: $\langle \delta \vec{n} \cdot \delta \vec{n} \rangle = \langle \partial h \cdot \partial h \rangle \sim \log(L)$

Flat phase is stabilized by anharmonic effects

[Nelson & Peliti J. Phys. '87]

Classical theory: Elastic energy = bending + stretching

[Landau & Lifshitz]

$$U = \frac{1}{2} \kappa (\partial^2 h)^2 + \frac{1}{2} \lambda \gamma_{ii}^2 + \mu \gamma_{ij} \gamma_{ij}$$

Relevant strain tensor: $\gamma_{ij} = \frac{1}{2} (\partial_i u_j + \partial_j u_i + \partial_i h \, \partial_j h)$

Anharmonic effects

Anharmonic effects change correlation functions:

$$\langle h_q h_{-q} \rangle \sim 1/q^{4-\eta} \qquad \langle u_q u_{-q} \rangle \sim 1/q^{2+\eta_u}$$

Anomalous elasticity: $\kappa(q) \sim q^{-\eta} \quad \lambda(q), \mu(q) \sim q^{\eta_u}$

Relation between exponents: $\eta_u + 2\eta = 4 - D$

[Aronovitz & Lubensky PRL '88]

Stabilization of flat phase $\langle \partial h \cdot \partial h \rangle \sim finite$

Problem is non-perturbative! Some theoretical values for *η*:

- ε-expansion: 0,96 [Aronovitz & Lubensky PRL '88]
- Self-consistent: 0,82 [Le Doussal & Radzihovsky PRL '92)]
- FRG: 0,849 [Kownacki & Mouhanna PRE '09]
- Monte Carlo: 0,85 [Los et al PRB '09]

Anharmonic effects

Experimental measurement of anomalous exponents

- X-ray and light scattering in **red blood cell membranes**:

$\eta \approx 0{,}65 \pm 0{,}10$ [Schmidt et al Science '93]

- X-ray scattering of **amphiphilic fields**:

$\eta \approx 0{,}7 \pm 0{,}2$ [Gourier et al PRL '97]

What about 2D crystals (graphene, hBN, MoS_2, WS_2, …)?

2D crystals as membranes

Anomalous elasticity

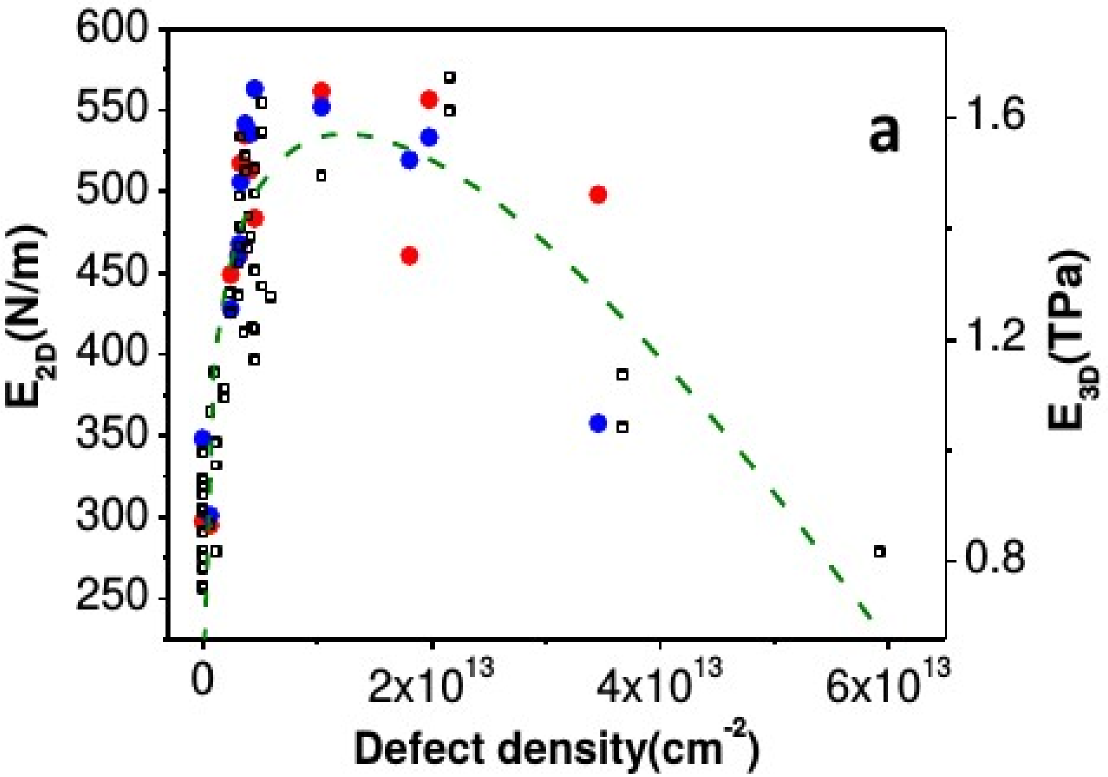
Graphene is usually advertised as "**strongest material ever**"

However, $\lambda(q), \mu(q) \sim q^{\eta_u}, \eta_u > 0$

No well defined elastic constants in the thermodynamic limit!

Data shows some decrease of "Young modulus" with sample size
[Lee et al Nano Lett '12]

"Young modulus" increases with defect concentration!
[López-Polín arXiv:1406.2131]



(Taken from López-Polín arXiv:1406.2131)

Thermal expansion

Usually described using **quasi-harmonic** approximation (QHA)

$$\alpha = \frac{k_B}{B_0} \frac{1}{V_0} \sum_{q,\lambda} \gamma_{q,\lambda} \frac{(\hbar \omega_{q,\lambda}/2 k_B T)^2}{\sinh(\hbar \omega_{q,\lambda}/2 k_B T)^2}$$

$$\gamma_{q,\lambda} = -\frac{\partial \log \omega_{q,\lambda}}{\partial \log V}$$

Grunneisen parameter

For the flexural mode: $\gamma_{q,F} = -(\lambda+\mu)/(2\kappa q^2)$

QHA predicts a divergent thermal expansion at any finite T!

$$\alpha \sim -k_B/(4\pi\kappa) \log(\Lambda L)$$

3rd Law of thermodynamics: $\alpha \rightarrow 0$ as $T \rightarrow 0$

Thermal expansion

Divergence of thermal expansion can be corrected at high T

$$\frac{1}{L} \rightarrow k_c = \sqrt{\frac{3 k_B T \, 4\mu(\lambda+\mu)}{16\pi\kappa^2(\lambda+2\mu)}}$$

[de Andres PRB '12]

However, classical result cannot be extended to $T \to 0$

Furthermore, graphene Debye temperature ~ 1000 K !

Quantum effects should still be important at room temperature

Quantum effects

Our aim

Realistic description of the mechanical properties of crystalline membranes requires a theory that includes:

- Quantum mechanical effects
- Non-perturbative treatment of anharmonic effects
- Works in the thermodynamic limit

Our aim

Realistic description of the mechanical properties of crystalline membranes requires a theory that includes:

- Quantum mechanical effects
- Non-perturbative treatment of anharmonic effects
- Works in the thermodynamic limit

On this talk

PRB **89**, 224307 (2014)

Model

Quantize Nelson & Peliti theory

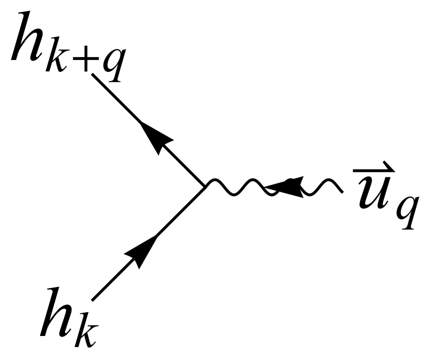
- Partition function

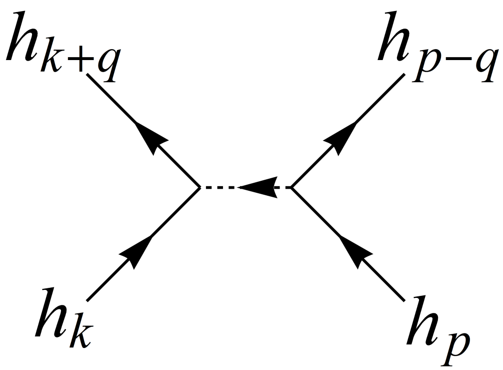
$$Z=\int D[h,\vec{u}]\,e^{-S_E}$$

with

$$S_E=\frac{1}{2}\int d\tau\, d^2x\left[\rho(\dot{h})^2+\rho(\dot{\vec{u}})^2+\kappa(\partial^2 h)^2+\lambda\gamma_{ii}^2+2\mu\gamma_{ij}\gamma_{ij}\right]$$

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





Perturbative calculation

Perturbative calculation of flexural mode self-energy

$$\langle h_q h_{-q}\rangle = \left(-\rho (i q_n)^2 + \kappa q^4 + \Sigma_q\right)^{-1}$$

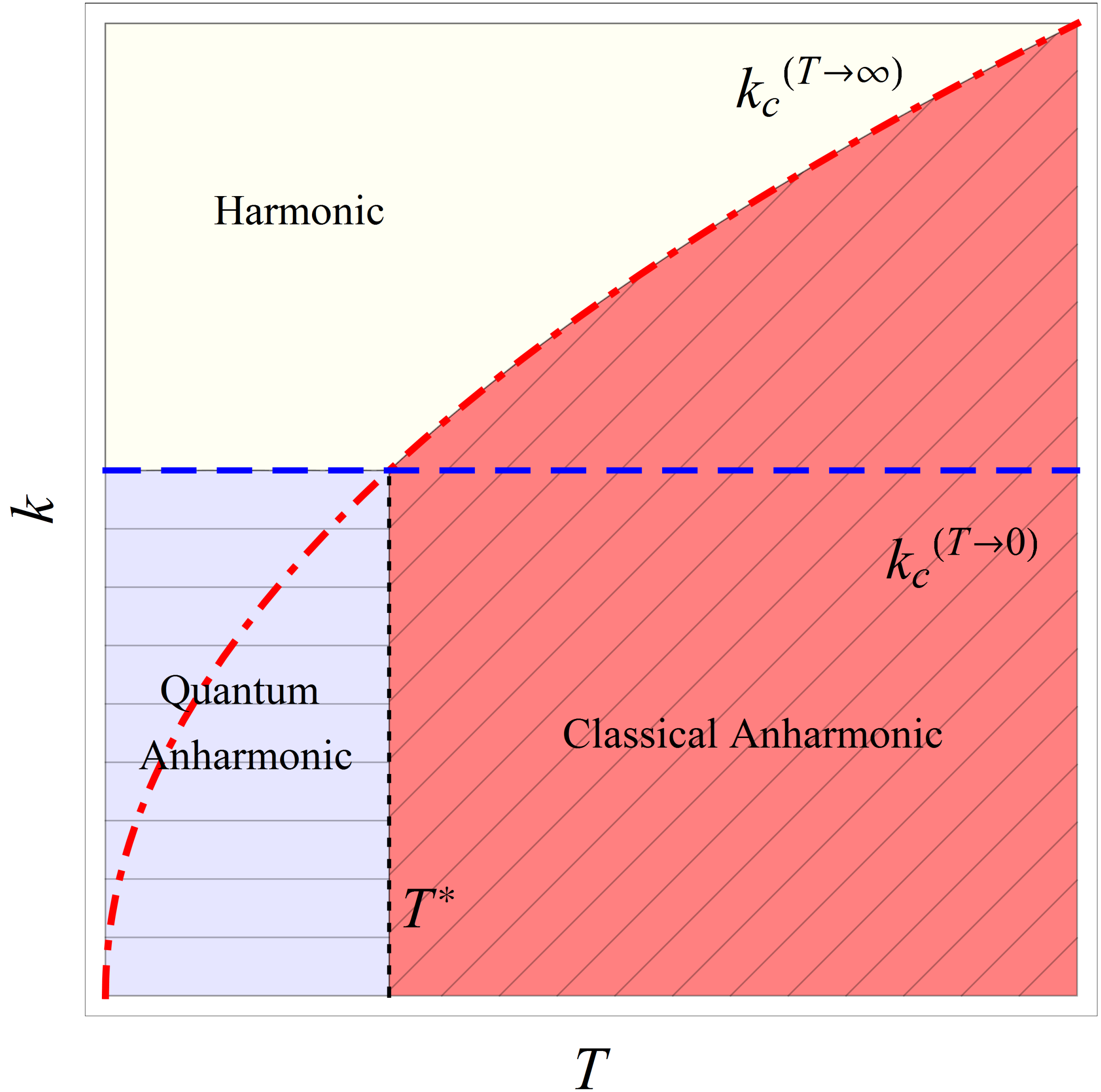
At high T: $$\Sigma_q \sim \frac{3 k_B T \mu(\lambda+\mu)}{4\pi(\lambda+2\mu)\kappa} q^2$$

At T=0: $$\Sigma_q \sim \frac{\hbar(\lambda+2\mu)^2}{8\pi\rho^{1/2}\kappa^{3/2}} f(\Lambda) q^2$$

Harmonic-to-anharmonic crossover: $\Sigma_q \sim \kappa q^4$

Classical-to-quantum crossover: $\lim_{T\to\infty} \Sigma_q \sim \lim_{T\to 0} \Sigma_q$

Perturbative calculation



For typical graphene values:

$$T^* \sim 70-90\,K$$

$$k_c^{(T\to\infty)} \sim 0{,}17\,\mathring{A}^{-1}$$

$$@\,300\,K$$

$$k_c^{(T\to 0)} \sim 0{,}1\,\mathring{A}^{-1}$$

Thermodynamics

Thermal expansion

Rigurously expressed as $\alpha = -\frac{1}{2}\frac{\partial}{\partial T}\langle \partial h \, \partial h \rangle$

Writing $\Sigma_q = \kappa k_c^{\eta} q^{4-\eta}$

We obtain at low T $\alpha \sim -\frac{k_B}{\kappa}\left(\frac{2\rho^{1/2} k_B T}{\hbar \kappa^{1/2} k_c^2}\right)^{2\eta/(4-\eta)}$

Becomes finite in the thermodynamic limit!

Thermodynamics

Specific heat

$$c_p = \left(\frac{\partial U}{\partial T} \right)_p$$

Total energy can be expressed using a **modified Migdal-Galitskii-Koltun sum**

$$U = \frac{1}{4\beta V} \sum_q \left(3\rho (i q_n)^2 + \kappa q^4 \right) \langle h_q h_{-q} \rangle + \frac{1}{\beta V} \sum_q \rho (i q_n)^2 \langle u^i_q u^i_{-q} \rangle$$

At low temperature $c_p \sim k_B k_c^2 \left(\frac{2\rho^{1/2} k_B T}{\hbar \kappa^{1/2} k_c^2} \right)^{4/(4-\eta)}$

Conclusions and current work

Conclusions and current work

Conclusions

- Anharmonic effects are fundamental to understand the physical properties of crystalline membranes
- Our calculation, hints that quantum fluctuations should be dominant bellow 70 K
- Anharmonic effects make thermal expansion finite

Current work

- Used model does not obbey rotational symmetry
- This is OK with classical theory. For quantum?
- Joint effect of thermal + quantum fluctuations
- Go beyond perturbative calculation