Quantum Monte Carlo for Carbon Nanotubes

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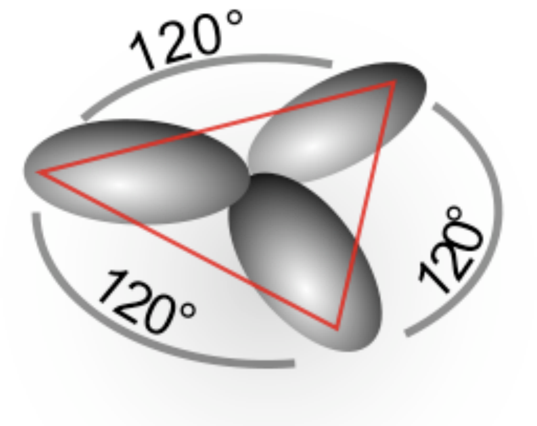
Outline

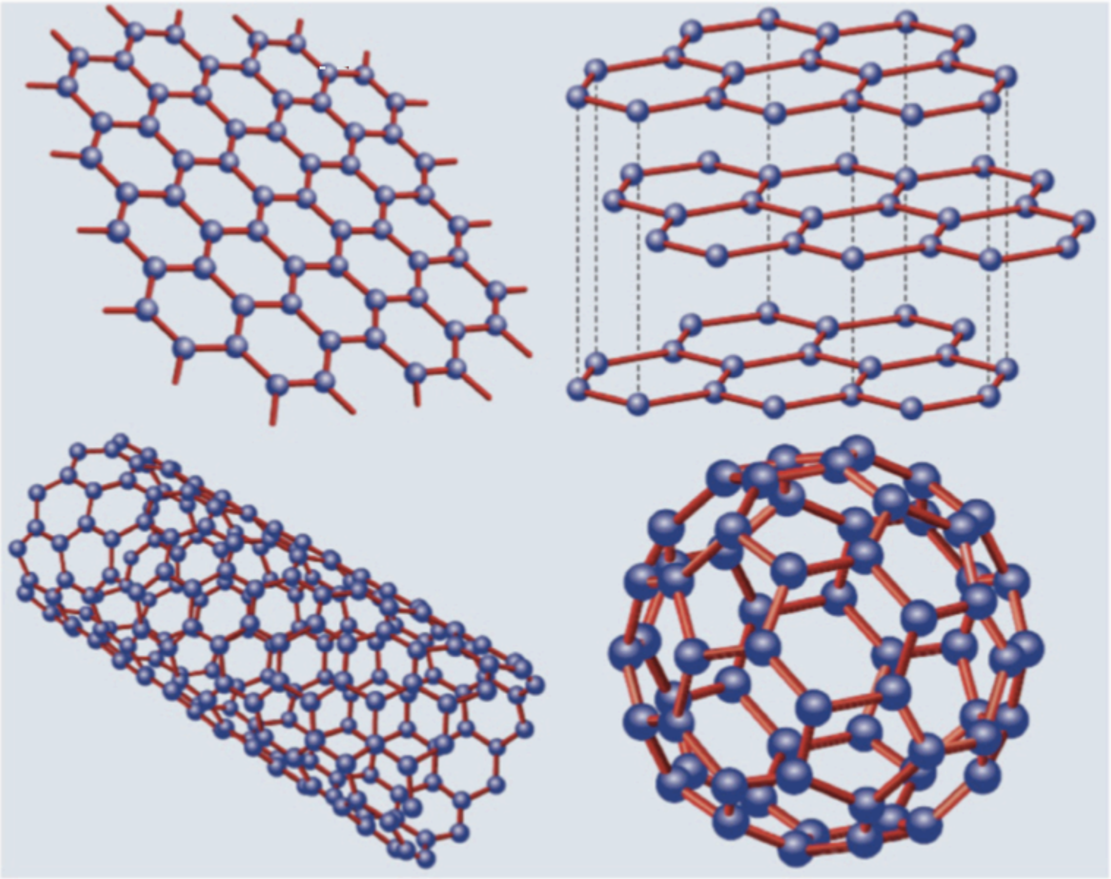
- Introduction: Dispersion relations in graphene and carbon nanotubes (non-interacting case)
- Quantum Monte Carlo formulations for hexagonal Hubbard theories (with electron-electron interactions)
- Correlators, bandgaps and dispersion relations in carbon nanotubes (with electron-electron interactions)
- Quantum Monte Carlo results for armchair nanotubes, experimental situation

Graphene as a “progenitor material” …



graphene
single graphite layer

graphite
stacked graphene



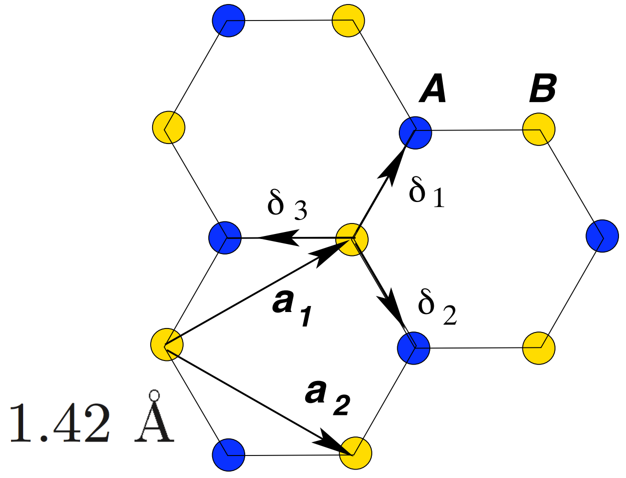
nanotube
rolled graphene

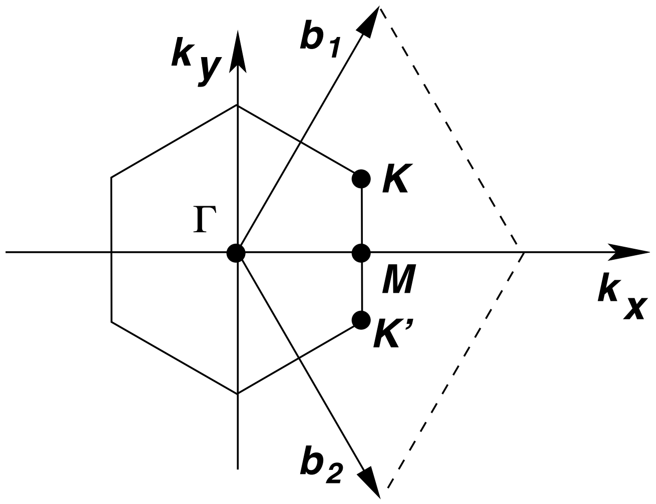
fullerene
wrapped graphene

Nearest-neighbor hopping description of graphene (non-interacting) …

$$\vec{a}_1 \equiv \left(\frac{3}{2}, \frac{\sqrt{3}}{2}\right) a\ ,$$

$$\vec{a}_2 \equiv \left(\frac{3}{2}, -\frac{\sqrt{3}}{2}\right) a$$





$$\vec{b}_1 \equiv \left(\frac{1}{3}, \frac{1}{\sqrt{3}}\right) \frac{2\pi}{a}\ ,$$

$$\vec{b}_2 \equiv \left(\frac{1}{3}, -\frac{1}{\sqrt{3}}\right) \frac{2\pi}{a}$$

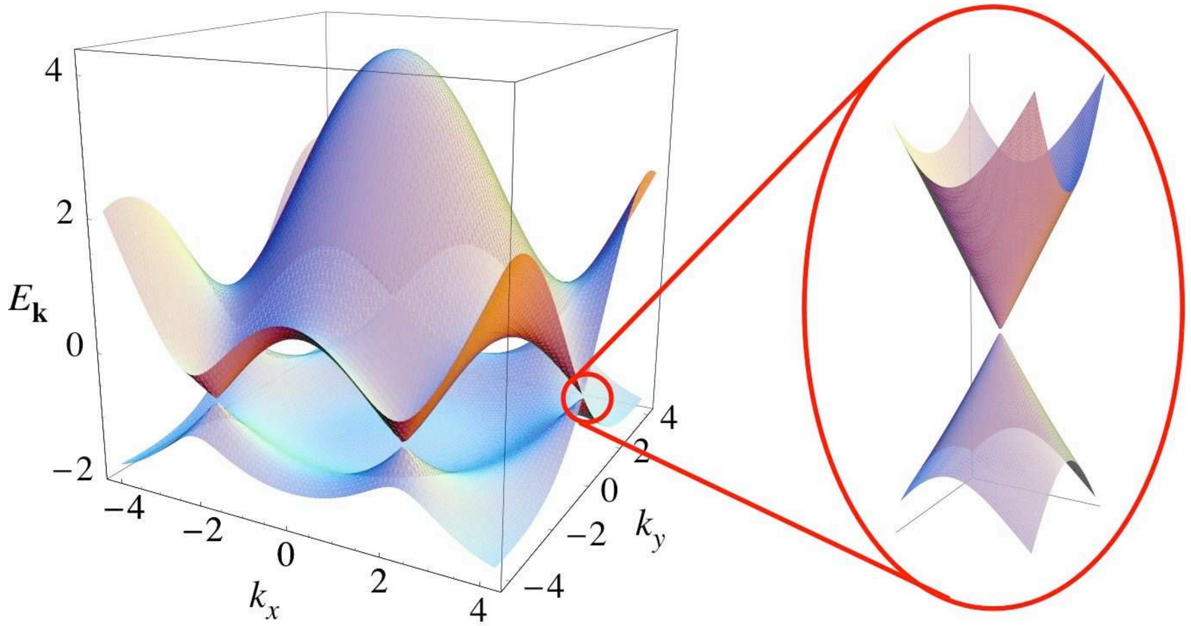
Tight-binding calculation of the dispersion relation …

$$H \equiv H_{tb}$$

$$\equiv -\kappa \sum_{\langle x,y\rangle,s} a^{\dagger}_{x,s} a_{y,s}$$

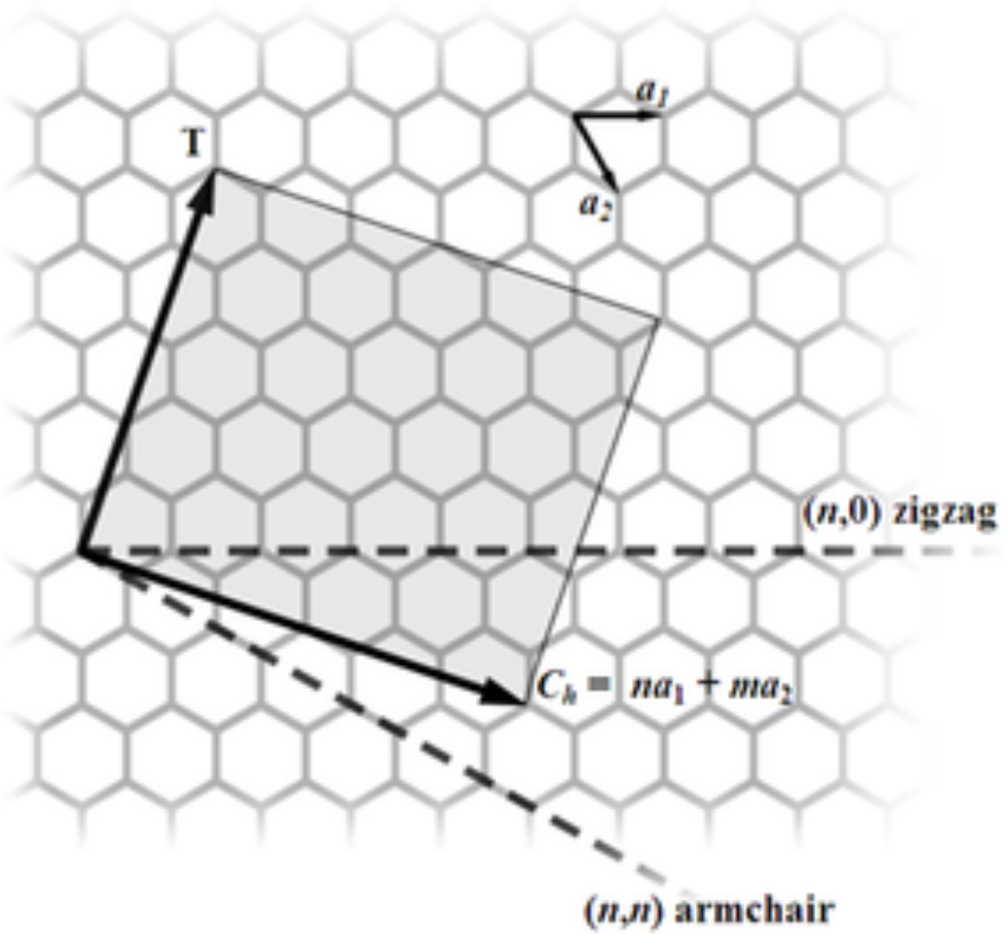
$$E(\vec{k}) = i\omega(\vec{k}) = \pm\kappa |f(\vec{k})|$$

$$f(\vec{k}) = e^{iak_x/\sqrt{3}} + 2e^{-iak_x/(2\sqrt{3})} \cos(ak_y/2)$$



Structure of carbon nanotubes, application of Quantum Monte Carlo …

Saito, Dresselhaus & Dresselhaus,
“Physical properties of carbon nanotubes”



Chiral vector:

$$\vec{C}_h \equiv n\vec{a}_1 + m\vec{a}_2$$

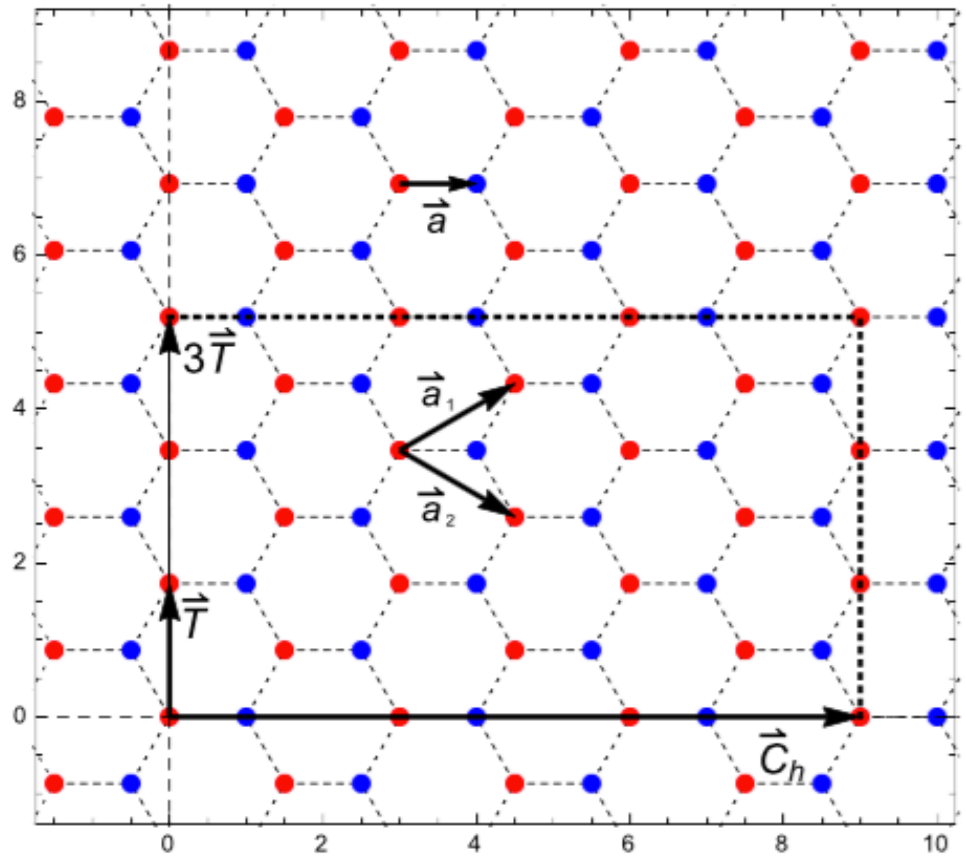
Translation vector:

$$\vec{T} \equiv t_1\vec{a}_1 + t_2\vec{a}_2$$

$$t_1 \equiv \frac{2m+n}{d_R}$$

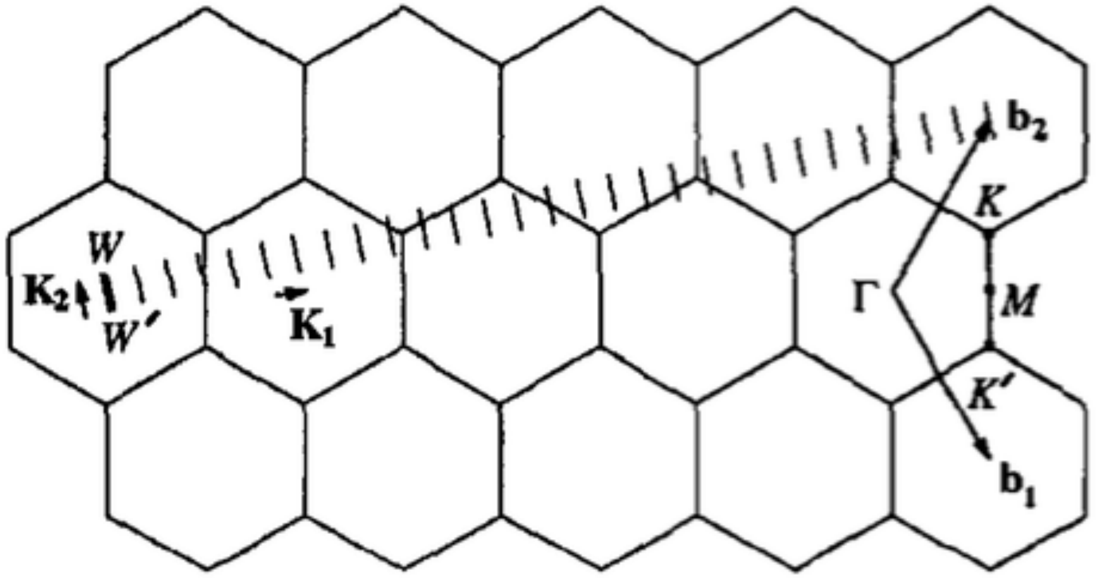
$$t_2 \equiv -\frac{2n+m}{d_R}$$

$$d_R \equiv \gcd(2m+n, 2n+m)$$



Chiral (4,2) nanotube - semiconducting …

Saito, Dresselhaus & Dresselhaus, "Physical properties of carbon nanotubes"



$$N_U = \frac{|\vec{C}_h \times \vec{T}|}{|\vec{a}_1 \times \vec{a}_2|}$$

$$\boldsymbol{K}_1 = \frac{1}{N}(-t_2\boldsymbol{b}_1 + t_1\boldsymbol{b}_2), \quad \boldsymbol{K}_2 = \frac{1}{N}(m\boldsymbol{b}_1 - n\boldsymbol{b}_2)$$

(4,2) w/ 3, 4, & 5 unit cells

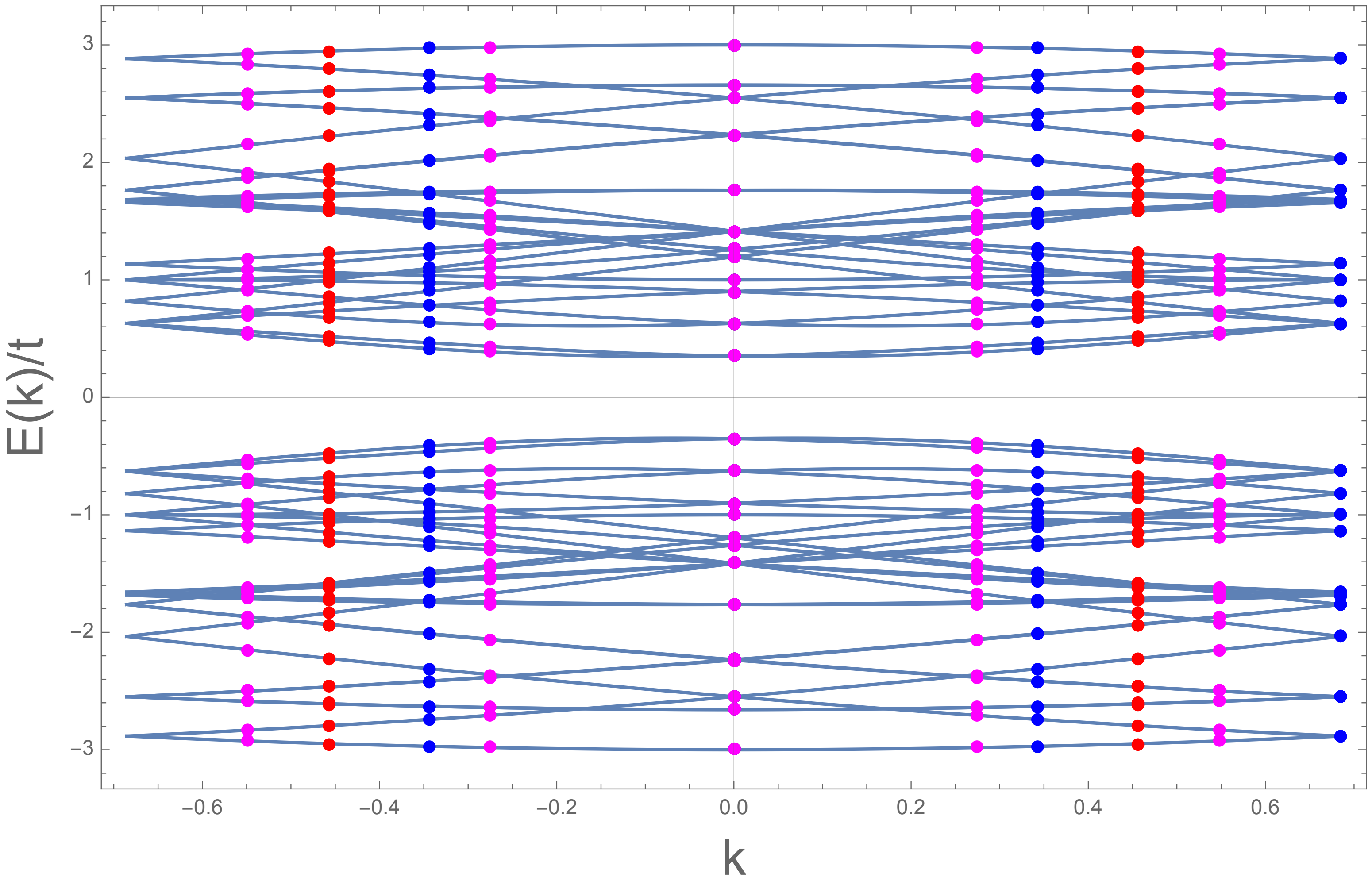
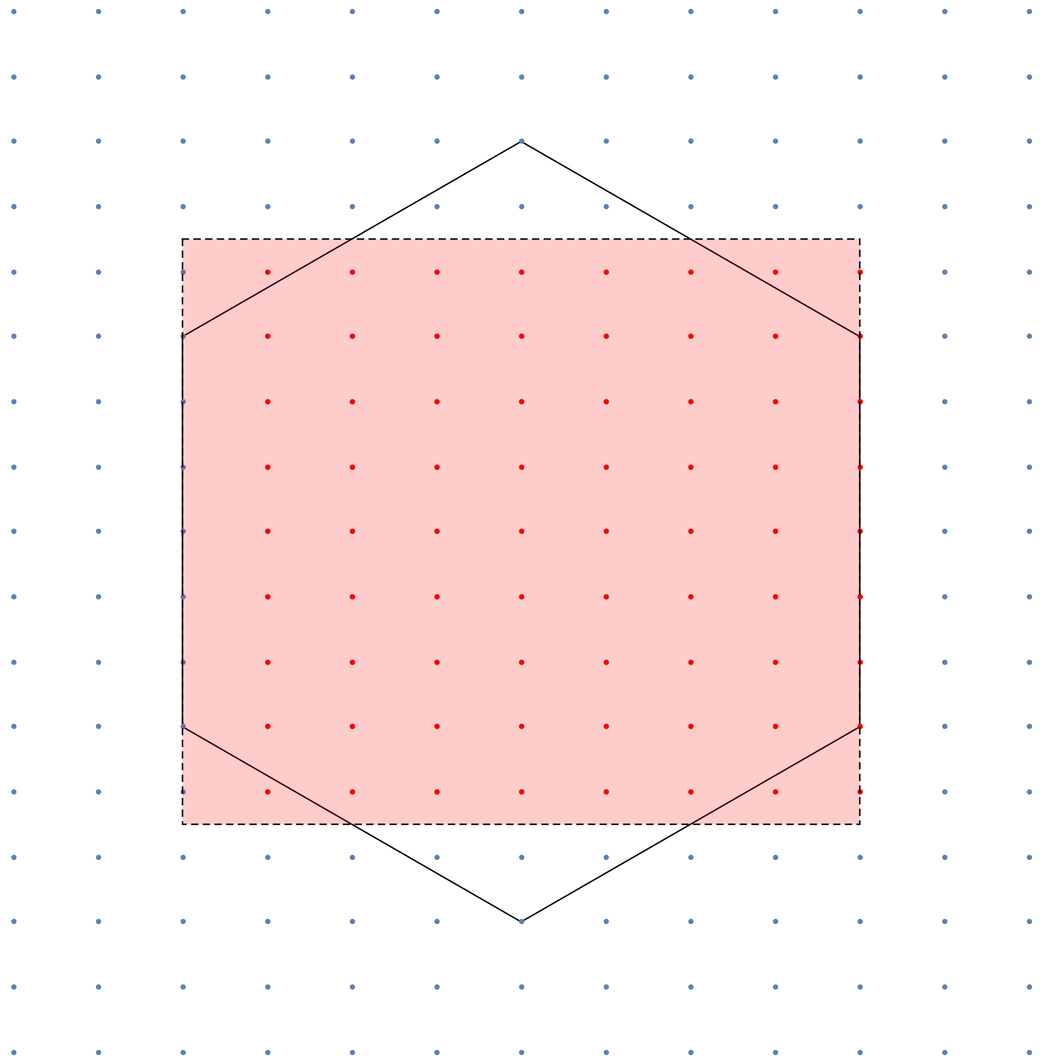
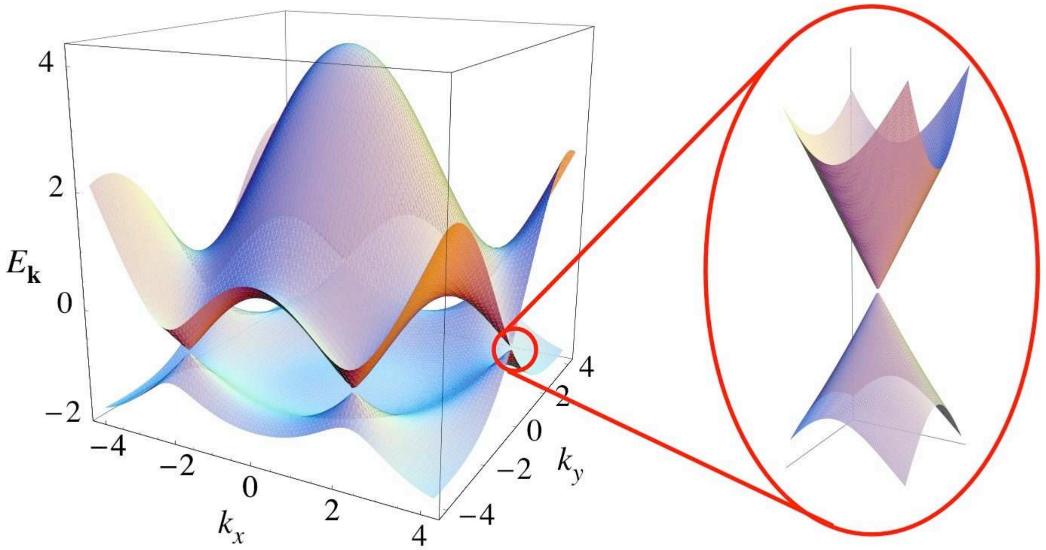


Figure by Thomas Luu

Armchair (4,4) nanotube - metallic ...





(4,4) w/ 6, 7, & 8 unit cells

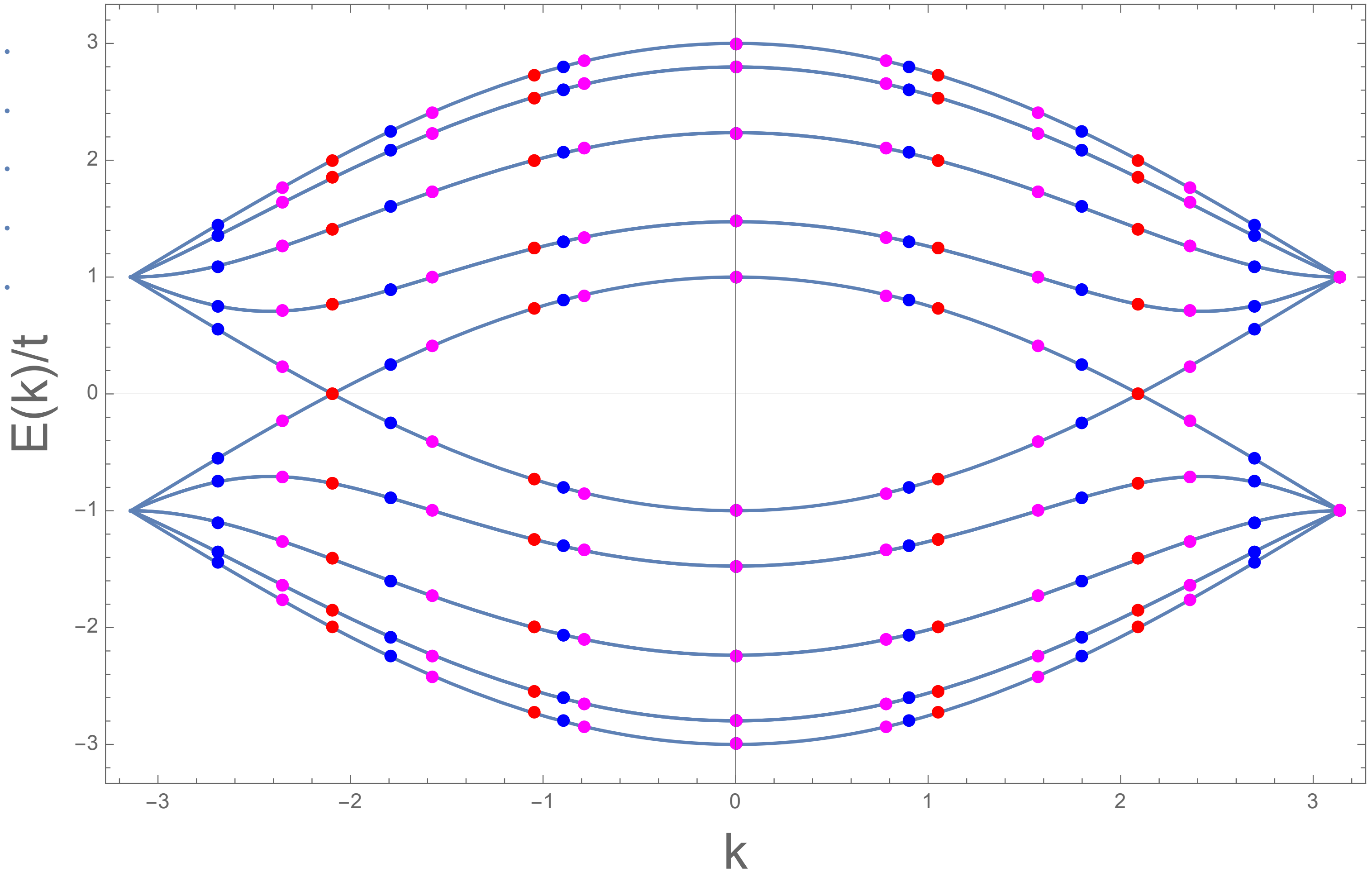


Figure by Thomas Luu

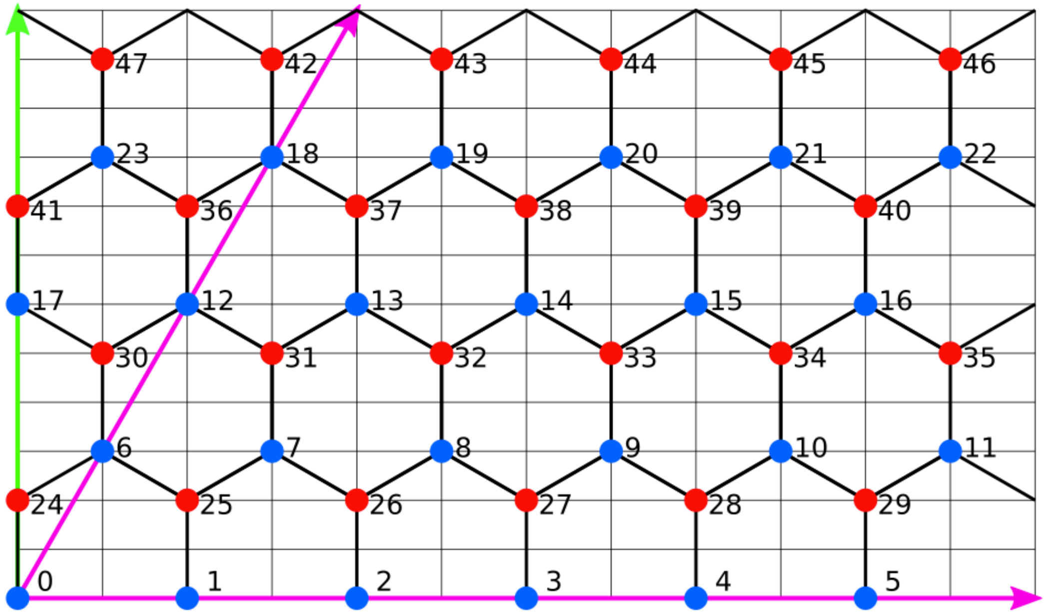
Graphene with electron-electron interactions, hexagonal Hubbard theory …

Paiva et al., Assaad et al., Sorella et al., Brower et al., Buividovich et al., von Smekal et al., …

$$H \equiv H_{tb} + H_I$$

$$\equiv -\kappa \sum_{\langle x,y\rangle,s} a^\dagger_{x,s} a_{y,s} + \frac{1}{2}\sum_{x,y} V_{x,y}\, q_x q_y$$

$$q_i \equiv a^\dagger_{i,\uparrow} a_{i,\uparrow} + a^\dagger_{i,\downarrow} a_{i,\downarrow} - 1$$



Introduce "hole operators" …

$$b^\dagger_{x,\downarrow} \equiv a_{x,\downarrow}, \qquad b_{x,\downarrow} \equiv a^\dagger_{x,\downarrow}$$

$$H = -\kappa \sum_{\langle x,y\rangle} \left(a^\dagger_{x,\uparrow} a_{y,\uparrow} - b^\dagger_{x,\downarrow} b_{y,\downarrow} \right) + \frac{1}{2}\sum_{x,y} V_{x,y}\, q_x q_y \qquad q_i = a^\dagger_{i,\uparrow} a_{i,\uparrow} - b^\dagger_{i,\downarrow} b_{i,\downarrow}$$

$$H = -\kappa \sum_{\langle x,y\rangle} \left(a^\dagger_x a_y + b^\dagger_x b_y \right) + \frac{1}{2}\sum_{x,y} V_{x,y}\, q_x q_y$$

Sign of hole operators changed on one sublattice

Operator expectation values,
Grassmann path integral, particles and holes …

Brower et al.,
Buividovich et al.,
von Smekal et al., …

$$\langle O(t)\rangle \equiv \frac{1}{Z}\mathrm{Tr}\left[O(t)e^{-\beta H}\right]$$

$$= \frac{1}{Z}\int \left[\prod_{\alpha} d\psi_\alpha^* d\psi_\alpha d\eta_\alpha^* d\eta_\alpha\right] e^{-\sum_\alpha(\psi_\alpha^*\psi_\alpha+\eta_\alpha^*\eta_\alpha)}\langle -\psi, -\eta|O(t)e^{-\beta H}|\psi,\eta\rangle$$

Euclidean time evolution
\beta = inverse temperature

$$e^{-\beta H} \equiv e^{-\delta H}e^{-\delta H}\cdots e^{-\delta H}$$

Subdivide Euclidean time
into N_t "slices"
\delta = \beta / N_t

Monte Carlo sampling, partition function …

$$Z = \mathrm{Tr}\left[e^{-\beta H}\right] =$$

$$\int \prod_{t=0}^{N_t-1}\left\{\left[\prod_{\alpha} d\psi_{\alpha,t}^* d\psi_{\alpha,t} d\eta_{\alpha,t}^* d\eta_{\alpha,t}\right] e^{-\sum_\alpha(\psi_{\alpha,t+1}^*\psi_{\alpha,t+1}+\eta_{\alpha,t+1}^*\eta_{\alpha,t+1})}\langle\psi_{t+1},\eta_{t+1}|e^{-\delta H}|\psi_t,\eta_t\rangle\right\}$$

Product of time slices,
anti-periodic boundary conditions

Hubbard-Stratonovich (HS) transformation …

$$\tilde{\kappa} \equiv \delta\kappa, \quad \tilde{V} \equiv \delta V, \quad \tilde{\phi} \equiv \delta\phi$$

$$\langle \psi_{t+1}, \eta_{t+1} | e^{-\delta H} | \psi_t, \eta_t \rangle = \langle \psi_{t+1}, \eta_{t+1} | e^{\delta\kappa \sum_{\langle x,y \rangle} \left(a_x^\dagger a_y + b_x^\dagger b_y \right) - \frac{1}{2} \sum_{x,y} \delta V_{x,y} q_x q_y} | \psi_t, \eta_t \rangle$$

$$\propto \int \prod_x d\tilde{\phi}_x \langle \psi_{t+1}, \eta_{t+1} | e^{\tilde{\kappa} \sum_{\langle x,y \rangle} \left(a_x^\dagger a_y + b_x^\dagger b_y \right) - \frac{1}{2} \sum_{x,y} [\tilde{V}]^{-1}_{x,y} \tilde{\phi}_x \tilde{\phi}_y + \sum_x i\tilde{\phi}_x q_x} | \psi_t, \eta_t \rangle$$

No quartic terms, but:
Path integral over "Auxiliary
field" degrees of freedom

Interactions encoded by "gauge links" …

$$\langle \psi_{t+1}, \eta_{t+1} | e^{-\delta H} | \psi_t, \eta_t \rangle = \int \prod_x d\tilde{\phi}_{x,t} \, e^{-\frac{1}{2} \sum_{x,y} [\tilde{V}]^{-1}_{x,y} \tilde{\phi}_{x,t} \tilde{\phi}_{y,t}}$$

$$\times \exp \left\{ \tilde{\kappa} \sum_{\langle x,y \rangle} \left(\psi^*_{x,t+1} \psi_{y,t} + \eta^*_{x,t+1} \eta_{y,t} \right) + \sum_x \left(e^{i\tilde{\phi}_{x,t}} \psi^*_{x,t+1} \psi_{x,t} + e^{-i\tilde{\phi}_{x,t}} \eta^*_{x,t+1} \eta_{x,t} \right) \right\} + \mathcal{O}(\delta^2)$$

We assumed here that exp(-\delta*H) is normal ordered
–> "discretization error" in \delta …

We can integrate over the Grassmann fields …

$$\begin{aligned} Z = \int \mathcal{D}\tilde{\phi}\mathcal{D}\psi^*\mathcal{D}\psi\mathcal{D}\eta^*\mathcal{D}\eta\, e^{-\frac{1}{2}\sum_{x,y,t}[\tilde{V}]^{-1}_{x,y}\tilde{\phi}_{x,t}\tilde{\phi}_{y,t}} \exp\Bigg\{ \tilde{\kappa} \sum_{\langle x,y\rangle,t} \left(\psi^*_{x,t+1}\psi_{y,t} + \eta^*_{x,t+1}\eta_{y,t}\right) \\ - \sum_{x,t} \left(\psi^*_{x,t+1}\left(\psi_{x,t+1} - e^{i\tilde{\phi}_{x,t}}\psi_{x,t}\right) + \eta^*_{x,t+1}\left(\eta_{x,t+1} - e^{-i\tilde{\phi}_{x,t}}\eta_{x,t}\right)\right) \Bigg\} \end{aligned}$$

Due to HS:
Quadratic dependence on
the Grassmann fields

Effective lattice action (similar to Lattice QCD) …

$$Z = \int \mathcal{D}\tilde{\phi}\, \det[M(\tilde{\phi})]\, \det[M^*(\tilde{\phi})] \exp\left\{ -\frac{1}{2} \sum_{x,y,t=0}^{N_t-1} [\tilde{V}]^{-1}_{x,y}\tilde{\phi}_{x,t}\tilde{\phi}_{y,t} \right\}$$

$$M(x,t;y,t';\tilde{\phi}) \equiv \delta_{x,y}\left(\delta_{t,t'} - e^{i\tilde{\phi}_{x,t'}}\delta_{t-1,t'}\right) - \tilde{\kappa}\,\delta_{\langle x,y\rangle}\delta_{t-1,t'}$$

“Fermion operator”

Monte Carlo probability weight ...

(note: should be positive definite!)

$$P(\tilde{\phi}) \equiv \frac{1}{Z} \det[M(\tilde{\phi})] \det[M^*(\tilde{\phi})] \exp\left\{ -\frac{1}{2} \sum_{x,y,t=0}^{N_t-1} [\tilde{V}]^{-1}_{x,y} \tilde{\phi}_{x,t} \tilde{\phi}_{y,t} \right\}$$

$$= \frac{1}{Z} \det[M(\tilde{\phi}) M^\dagger(\tilde{\phi})] \exp\left\{ -\frac{1}{2} \sum_{x,y,t=0}^{N_t-1} [\tilde{V}]^{-1}_{x,y} \tilde{\phi}_{x,t} \tilde{\phi}_{y,t} \right\},$$

Generate "ensembles" of configurations
with your favorite Monte Carlo algorithm!

Calculation of observables:

Metropolis, Hybrid Monte Carlo ...

$$\langle O \rangle \approx \frac{1}{N_{\rm cf}} \sum_{i=1}^{N_{\rm cf}} O[\tilde{\phi}_i]$$

Single quasiparticle propagator
–> quasiparticle spectrum

$$\langle a_x(\tau) a_y^\dagger(0) \rangle = \langle M^{-1}(x,\tau;y,0) \rangle \approx \frac{1}{N_{\rm cf}} \sum_{i=1}^{N_{\rm cf}} M^{-1}(x,\tau;y,0;\tilde{\phi}_i)$$

For the calculation of correlators:

Exploit the A/B sublattice structure of graphene ...

$$\Psi(x,t) = \begin{pmatrix} \Psi_A(x,t) \\ \Psi_B(x,t) \end{pmatrix} = \begin{pmatrix} \psi_{x,t} \\ \psi_{x+\vec{a},t} \end{pmatrix} \qquad \Phi(x,t) = \begin{pmatrix} \Phi_A(x,t) \\ \Phi_B(x,t) \end{pmatrix} = \begin{pmatrix} \tilde{\phi}_{x,t} \\ \tilde{\phi}_{x+\vec{a},t} \end{pmatrix}$$

Redefinition of electron and auxiliary fields

Block structure of the fermion matrix ...

$$M(x,t';y,t)\Psi(y,t) =$$

$$\begin{pmatrix} \delta_{x,y}\left(\delta_{t',t} - e^{i\Phi_A(x,t')}\delta_{t-1,t'}\right) & -\tilde{\kappa}\;\delta_{\langle x,y\rangle}\delta_{t-1,t'} \\ -\tilde{\kappa}\;\delta_{\langle x,y\rangle}\delta_{t-1,t'} & \delta_{x,y}\left(\delta_{t',t} - e^{i\Phi_B(x,t')}\delta_{t-1,t'}\right) \end{pmatrix} \begin{pmatrix} \Psi_A(y,t) \\ \Psi_B(y,t) \end{pmatrix}$$

Equivalent to the original M, but (x,y) now label cells

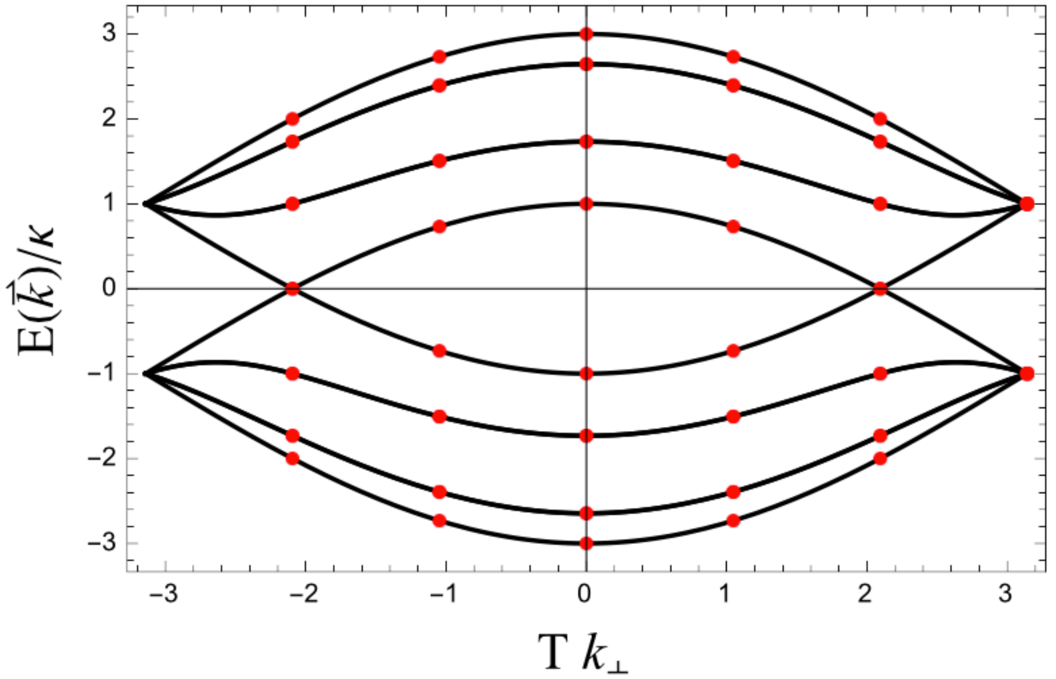
Correlators for arbitrary momentum modes:

Momentum projection …

$$G_\pm(\vec{k}_i,\tau) \equiv \langle a_\pm(\vec{k}_i,\tau) a_\pm^\dagger(\vec{k}_i,0)\rangle = \frac{1}{N^2}\sum_{\vec{x}_j,\vec{x}_k\in\{\vec{X}\}} e^{i\vec{k}_i\cdot(\vec{x}_j-\vec{x}_k)}\langle a_\pm(\vec{x}_j,\tau)a_\pm^\dagger(\vec{x}_k,0)\rangle$$

$$a_\pm^\dagger(\vec{x}) \equiv \frac{1}{\sqrt{2}}\begin{pmatrix} a_A^\dagger(\vec{x}) \\ \pm a_B^\dagger(\vec{x})\end{pmatrix} = \frac{1}{\sqrt{2}}\begin{pmatrix} a_{\vec{x}}^\dagger \\ \pm a_{\vec{x}+\vec{a}}^\dagger\end{pmatrix}$$

(3,3) nanotube, 6 unit cells



Linear combination of A/B correlators …

$$G_\pm(\vec{k}_i,\tau) = \frac{1}{2N^2}\sum_{\vec{x}_j,\vec{x}_k\in\{\vec{X}\}} e^{i\vec{k}_i\cdot(\vec{x}_j-\vec{x}_k)}\Big\{\langle M_{AA}^{-1}(\vec{x}_j,\vec{x}_k;\tau)\rangle + \langle M_{BB}^{-1}(\vec{x}_j,\vec{x}_k;\tau)\rangle$$

$$\pm\left(\langle M_{AB}^{-1}(\vec{x}_j,\vec{x}_k;\tau)\rangle + \langle M_{BA}^{-1}(\vec{x}_j,\vec{x}_k;\tau)\rangle\right)\Big\}$$

$$= \frac{1}{2}\left[G_{AA}(\vec{k}_i,\tau) + G_{BB}(\vec{k}_i,\tau) \pm \left(G_{AB}(\vec{k}_i,\tau) + G_{BA}(\vec{k}_i,\tau)\right)\right],$$

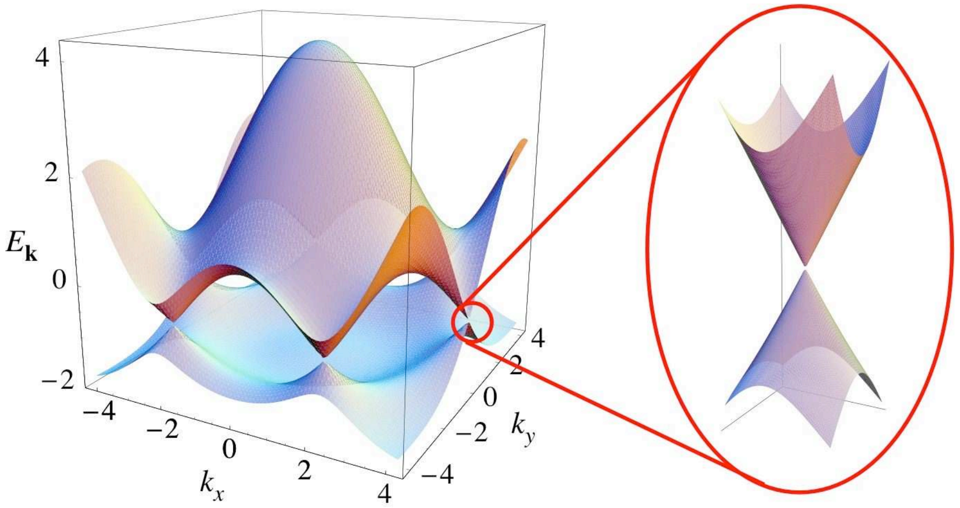
Non-interacting case:

Analytical solution possible …

$$G(\vec{k}_i,\tau) = \frac{1}{2\cosh(\omega(\vec{k}_i)\beta/2)} \begin{pmatrix} \cosh(\omega(\vec{k}_i)(\tau-\beta/2)) & e^{i\theta_{k_i}}\sinh(\omega(\vec{k}_i)(\tau-\beta/2)) \\ e^{-i\theta_{k_i}}\sinh(\omega(\vec{k}_i)(\tau-\beta/2)) & \cosh(\omega(\vec{k}_i)(\tau-\beta/2)) \end{pmatrix}$$

$$\equiv \begin{pmatrix} G_{AA}(\vec{k}_i,\tau) & G_{AB}(\vec{k}_i,\tau) \\ G_{BA}(\vec{k}_i,\tau) & G_{BB}(\vec{k}_i,\tau) \end{pmatrix},$$

$$\theta_{k_i} \equiv \tan^{-1}(\mathrm{Im}f(\vec{k}_i)/\mathrm{Re}f(\vec{k}_i))$$



$$G_\pm(\vec{k}_i,\tau) \equiv \frac{1}{2}\left[G_{AA}(\vec{k}_i,\tau) + G_{BB}(\vec{k}_i,\tau) \pm (G_{AB}(\vec{k}_i,\tau) + G_{BA}(\vec{k}_i,\tau))\right]$$

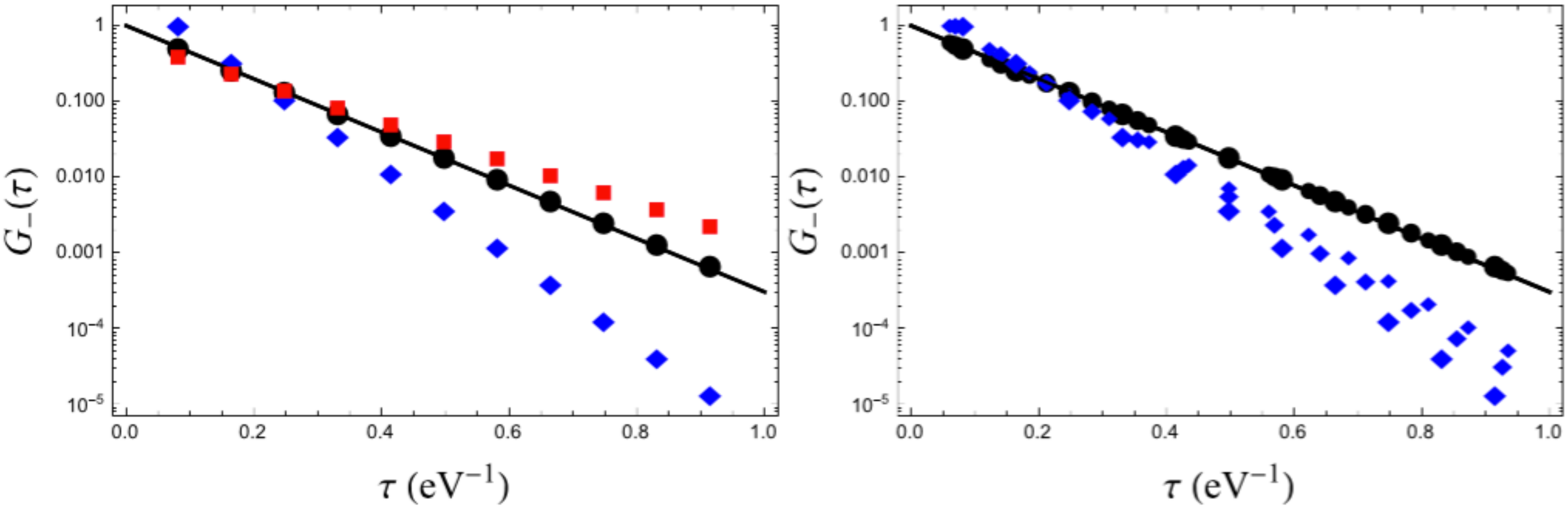
$$= \frac{1}{2\cosh(\omega(\vec{k}_i)\beta/2)}\left[\cosh(\omega(\vec{k}_i)(t-\beta/2)) \pm \cos(\theta_{k_i})\sinh(\omega(\vec{k}_i)(t-\beta/2))\right]$$

$$G_\pm(\vec{k}_i,\tau) \propto e^{\pm\omega(\vec{k}_i)\tau}$$

The dispersion relation can be obtained from the asymptotic behavior

Clever choice of time derivative:

—> Discretization error is greatly reduced



Blue diamonds —> forward time difference

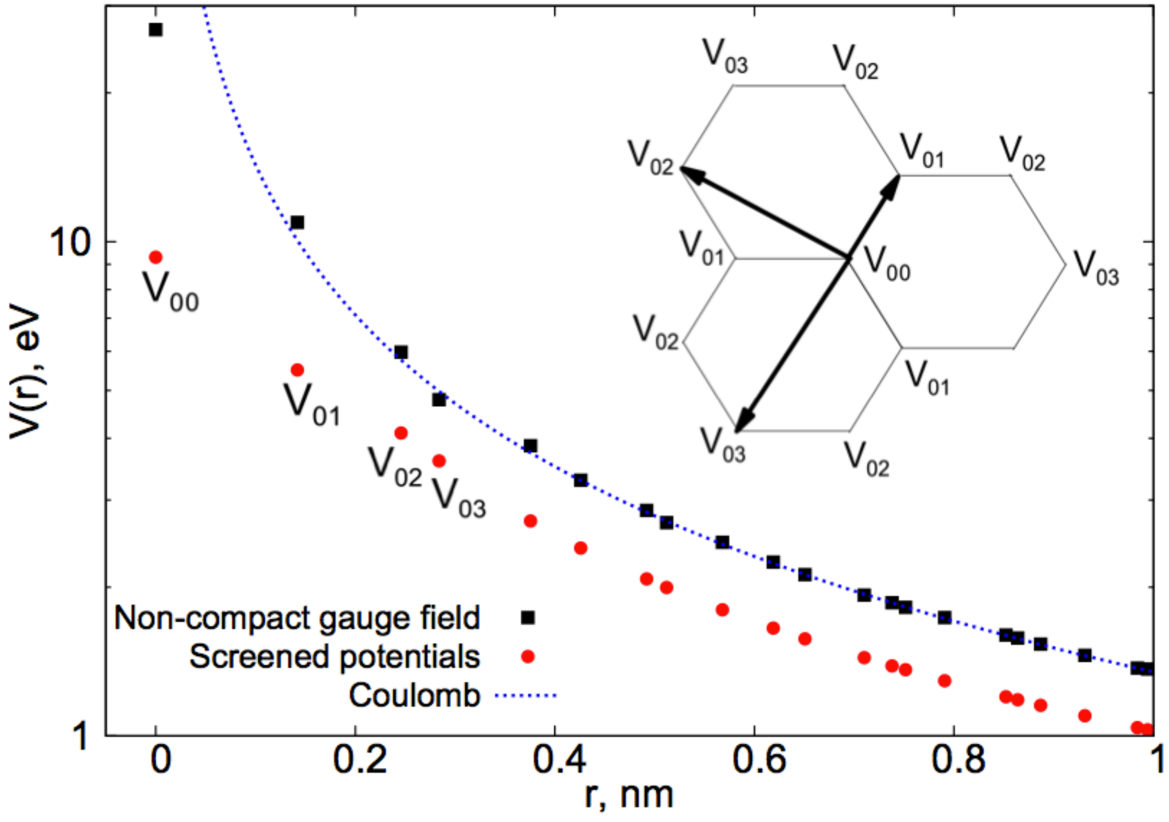
Red squares —> backward time difference

Black circles —> "mixed" time difference

$$M(x,t';y,t;\Phi) = \begin{pmatrix} \delta_{x,y}\left(e^{-i\Phi_A(x,t')}\delta_{t+1,t'} - \delta_{t,t'}\right) & -\tilde{\kappa}\ \delta_{\langle x,y\rangle}\delta_{t,t'} \\ -\tilde{\kappa}\ \delta_{\langle x,y\rangle}\delta_{t,t'} & \delta_{x,y}\left(\delta_{t',t} - e^{i\Phi_B(x,t')}\delta_{t-1,t'}\right) \end{pmatrix}$$

How do we obtain a realistic potential?

Note: long-range part modified by curvature …



We have so far used the screened Coulomb potential —> too strong?

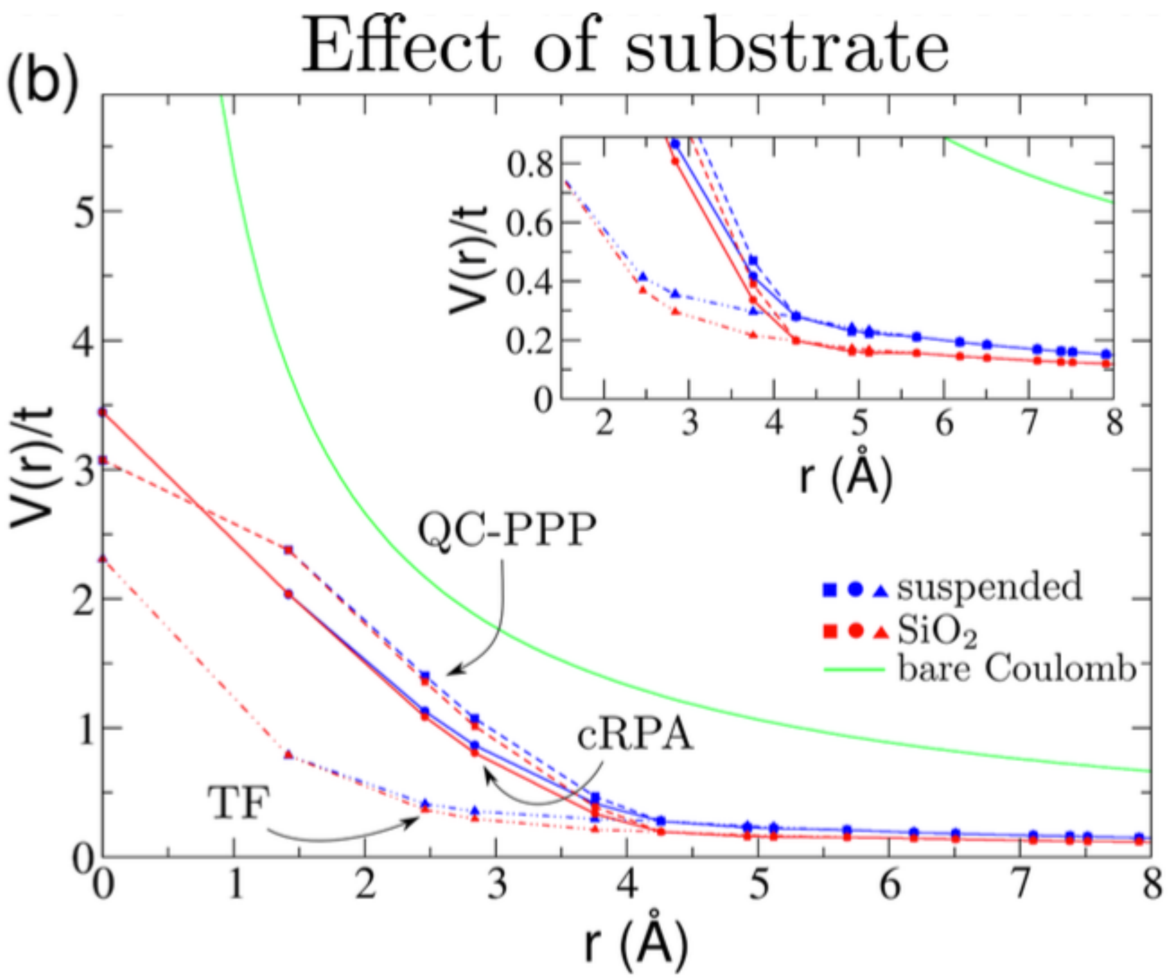
see talks by S. Adam & J. Rodrigues

T. O. Wehling et al.,
Phys. Rev. Lett. ***106****, (2011) 236805*

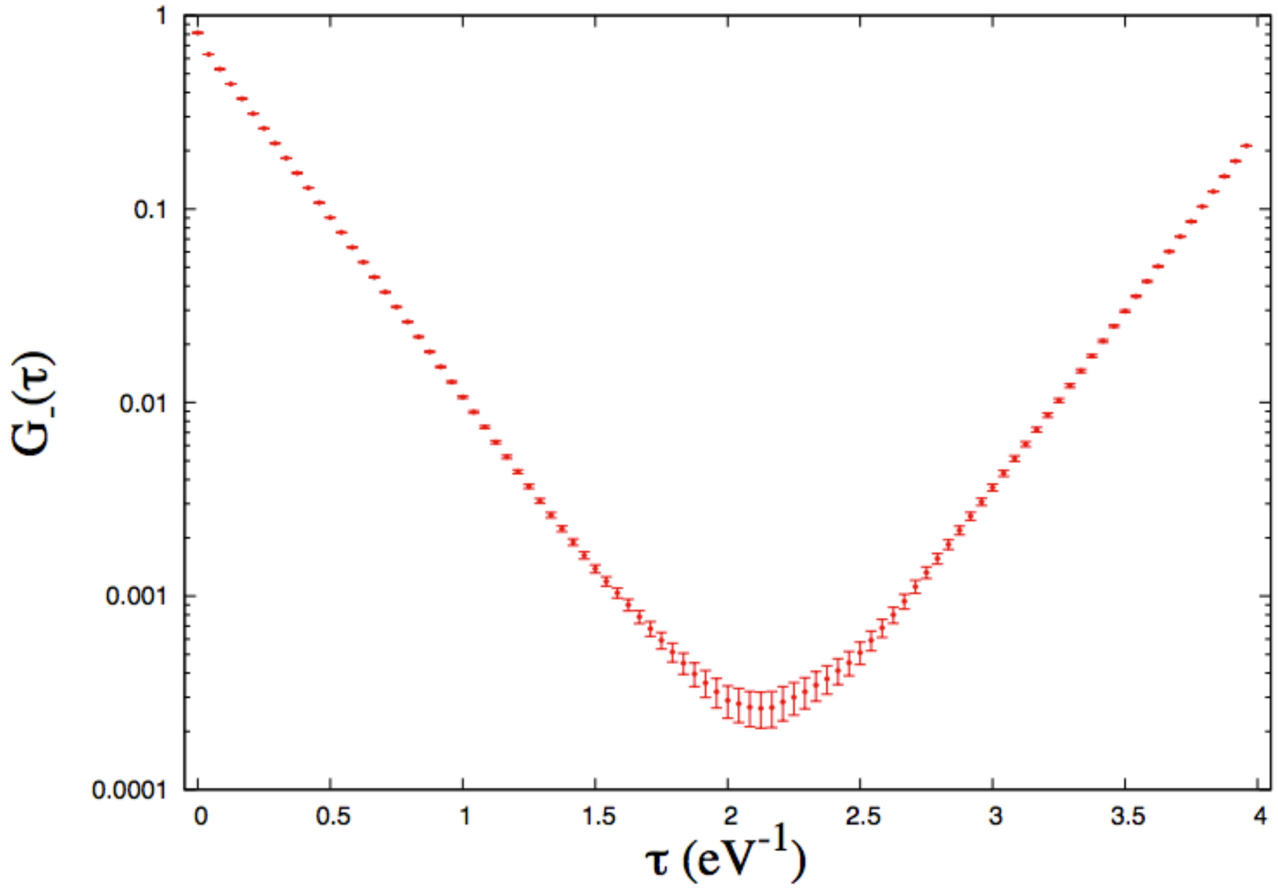
M. V. Ulybyshev et al.,
Phys. Rev. Lett. ***111****, (2013) 056801*

D. Smith and L. von Smekal,
Phys. Rev. B ***89****, (2014) 195429*

Ho-Kin Tang et al.,
Phys. Rev. Lett. ***115****, (2015) 186602*



Determination of the single-particle gap:
(3,3) armchair nanotube, actual Monte Carlo data …

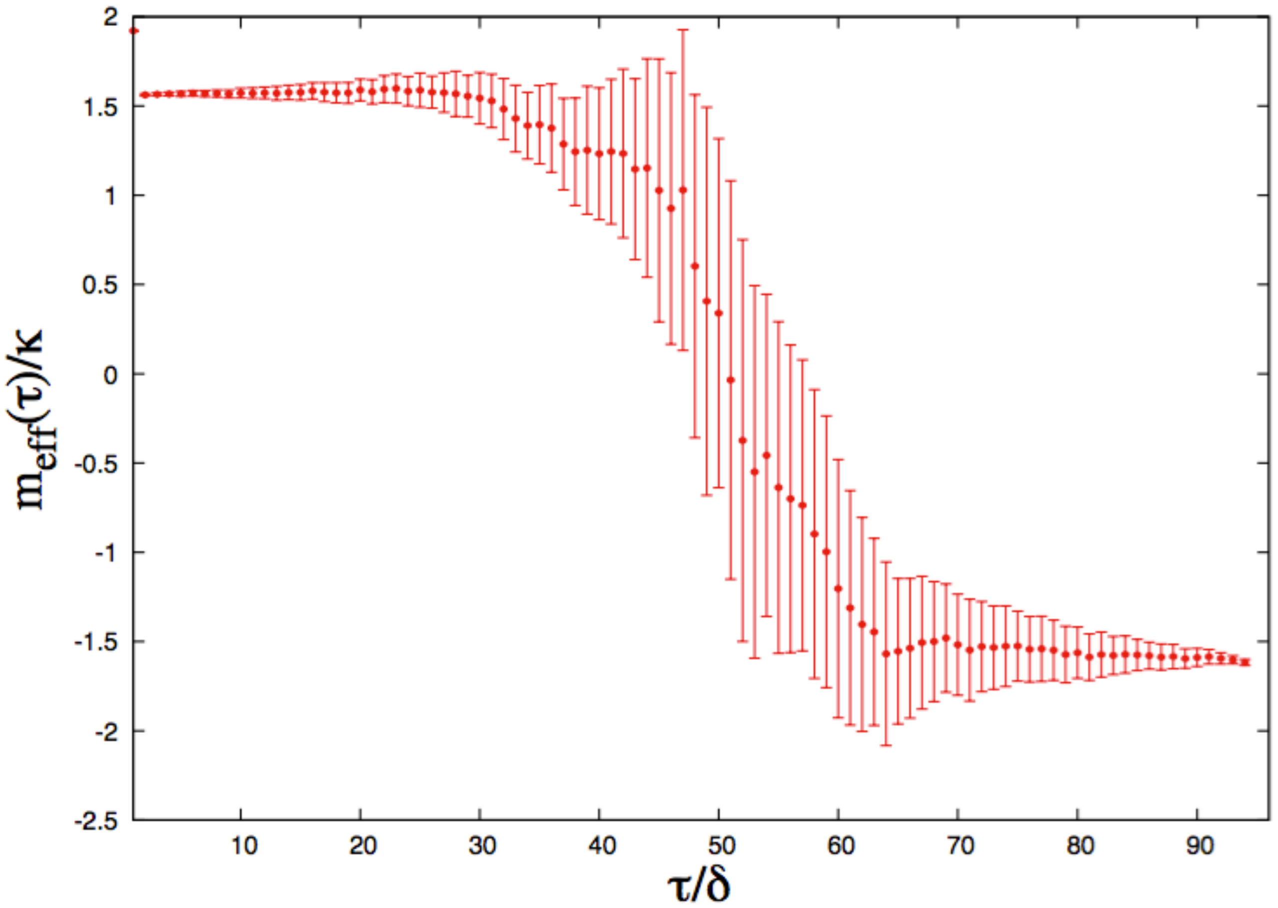


Projected onto a specific momentum mode in the spectrum

$$|\vec{T}|(|k_\perp|, |k_{||,i}|) = \left(\frac{2\pi}{3}, \frac{4\pi}{3\sqrt{3}}\right)$$

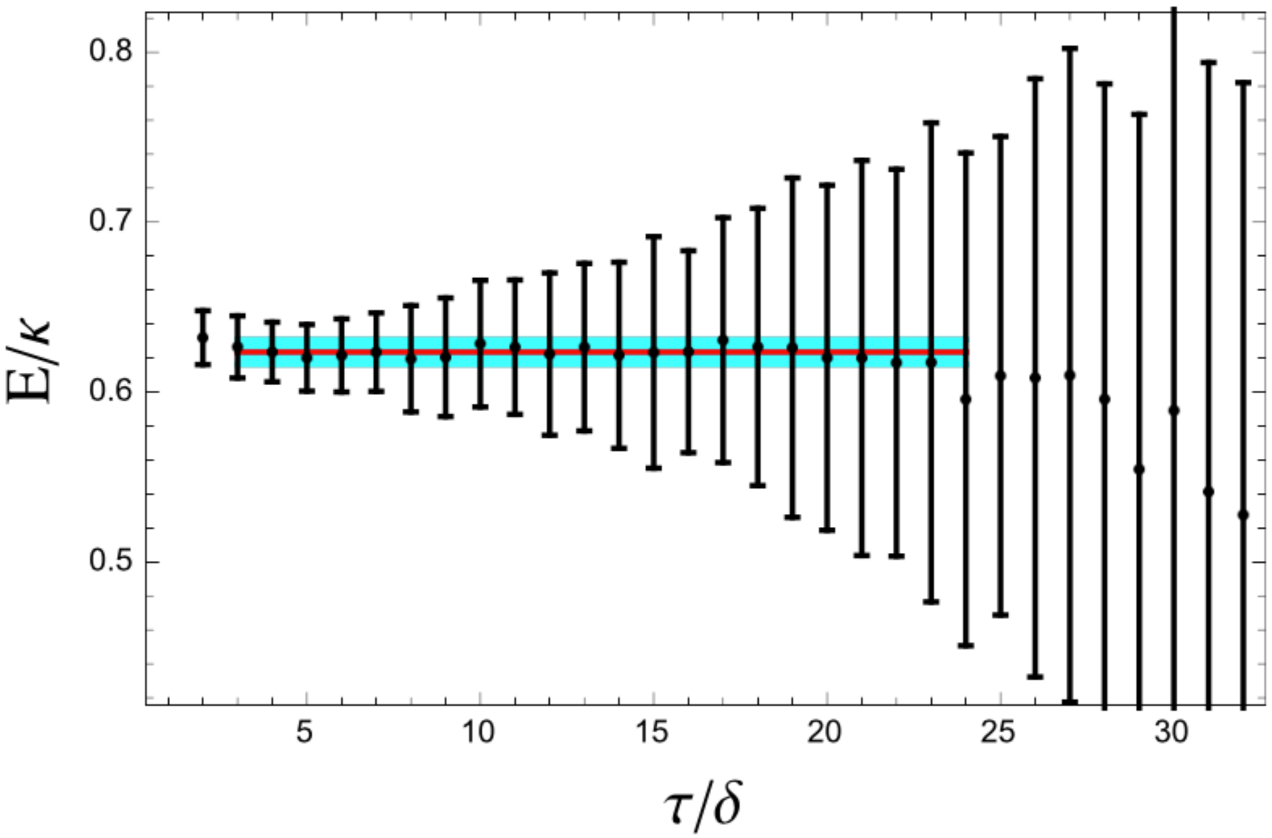
$$m_{\text{eff}}((\tau/\delta + \Delta)/2) = -\frac{1}{\Delta}\frac{\ln(G_-(\tau/\delta + \Delta))}{\ln(G_-(\tau/\delta))}$$

"Effective mass plot" à la Lattice QCD —>

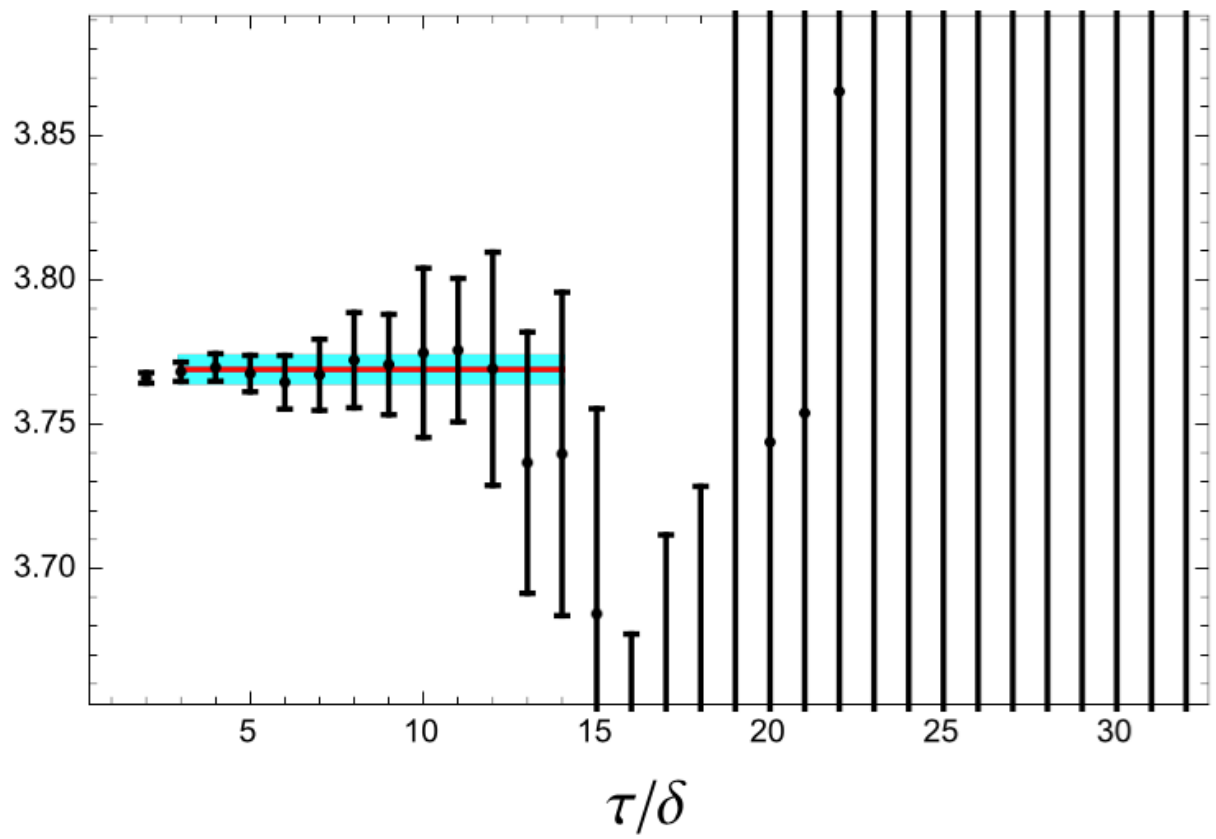


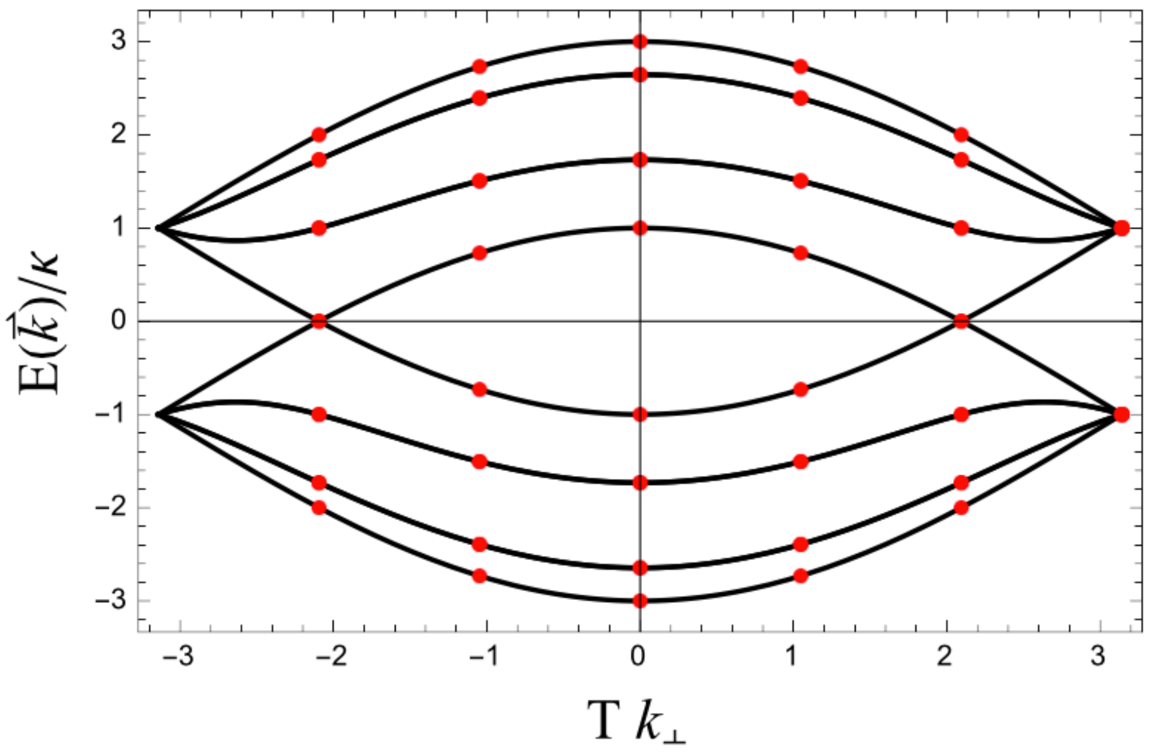
Colored bands —> fit to asymptotic correlator
Points with errorbars —> effective mass (illustration only)

K (Dirac) point



Gamma point (highest energy)



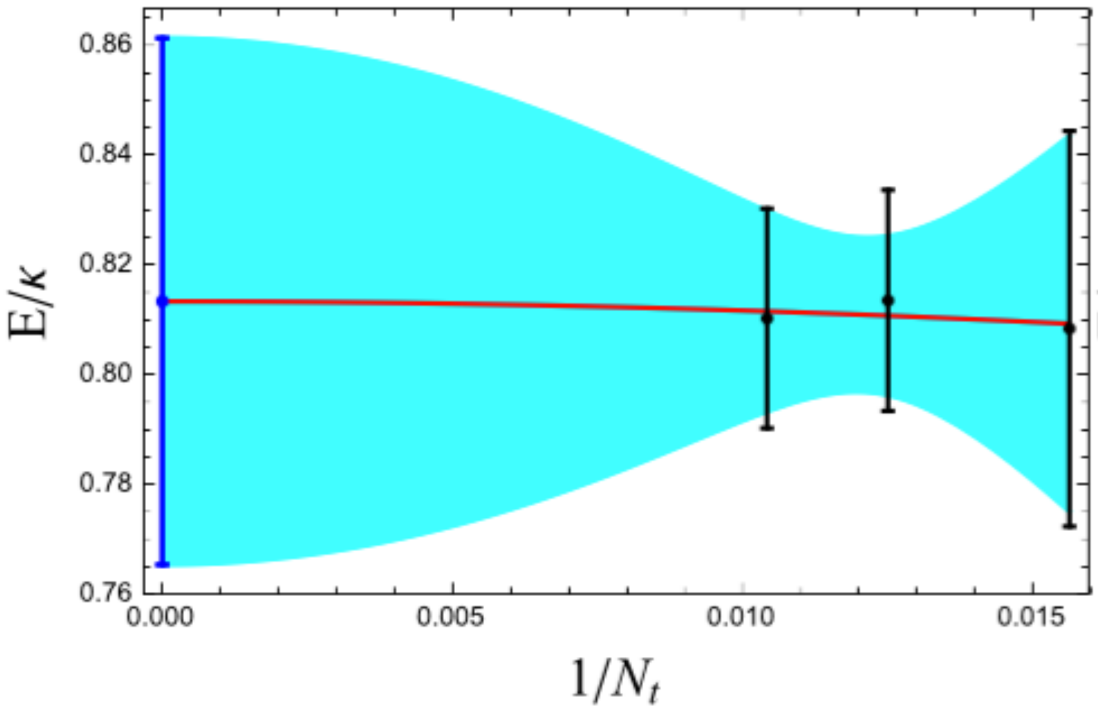


Extrapolation in Euclidean time, tube length ...

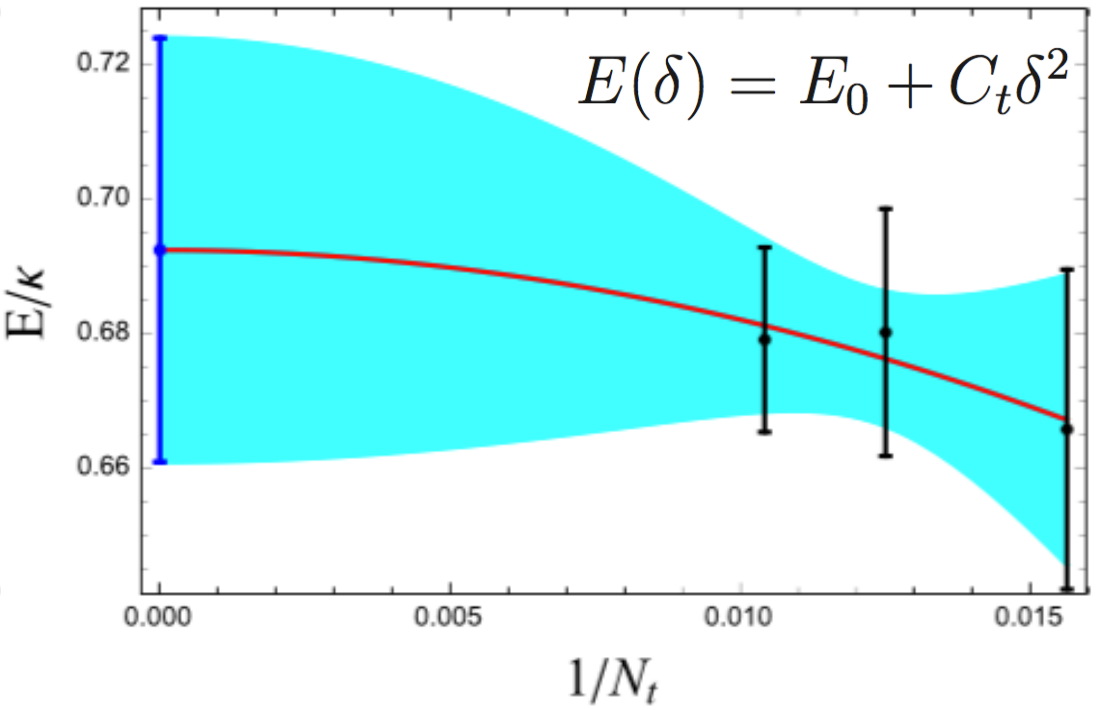
Dirac (K) point ...

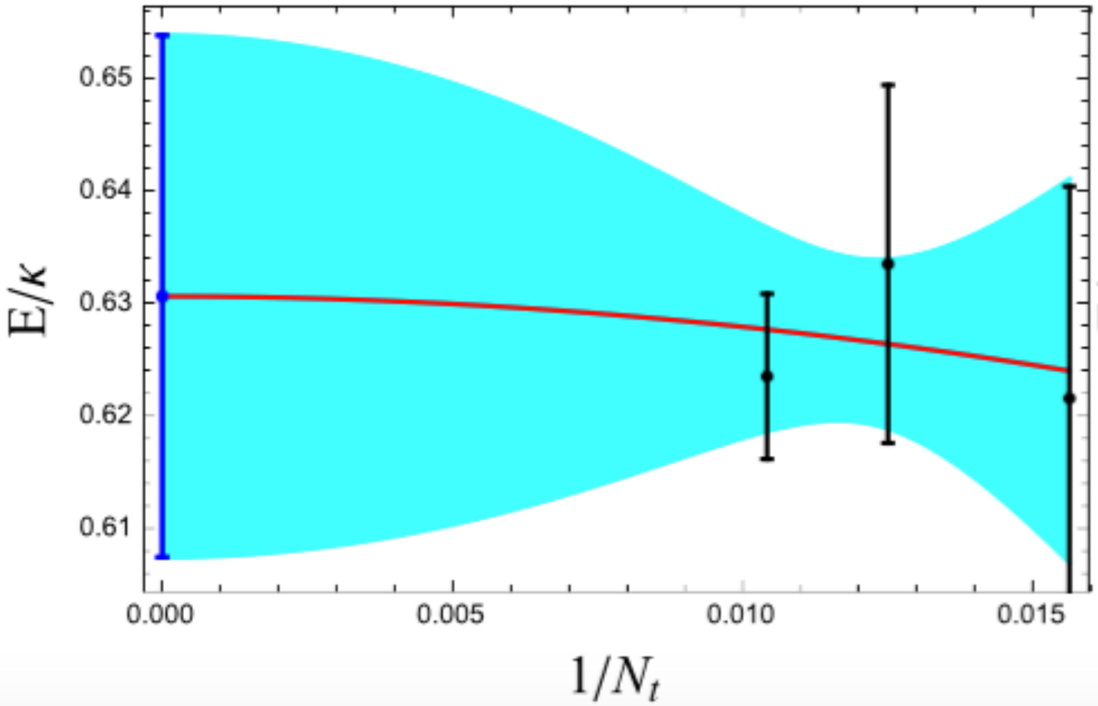
$$E_K/\kappa \;=\; .551(46)$$

3 unit cells

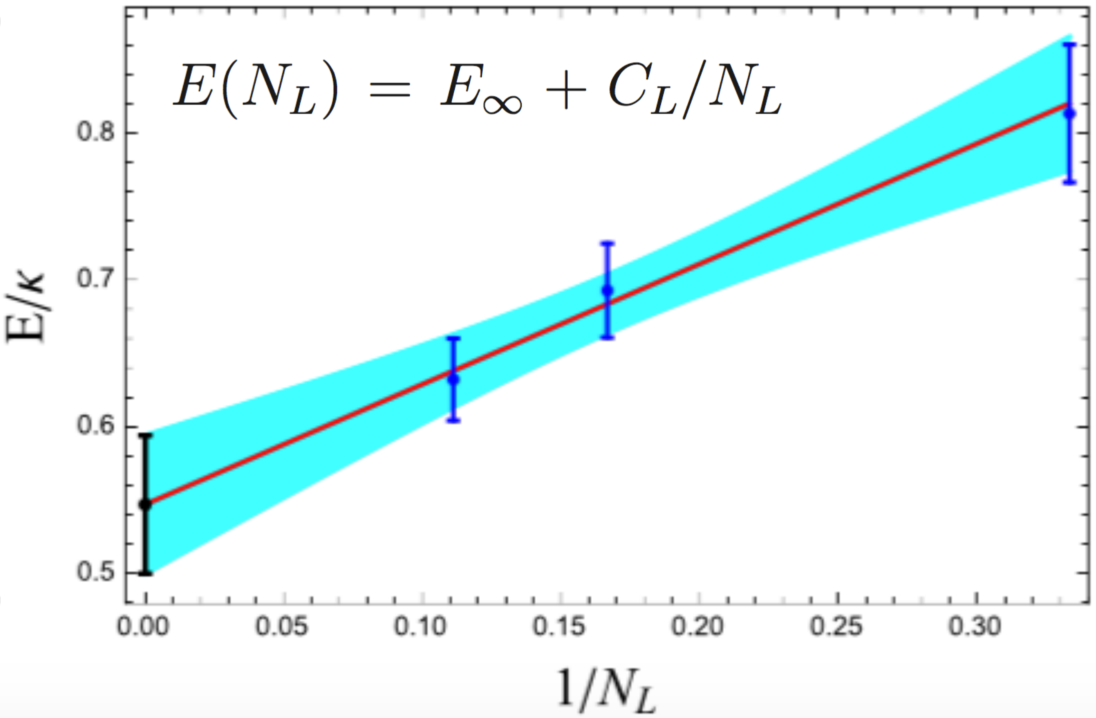


6 unit cells



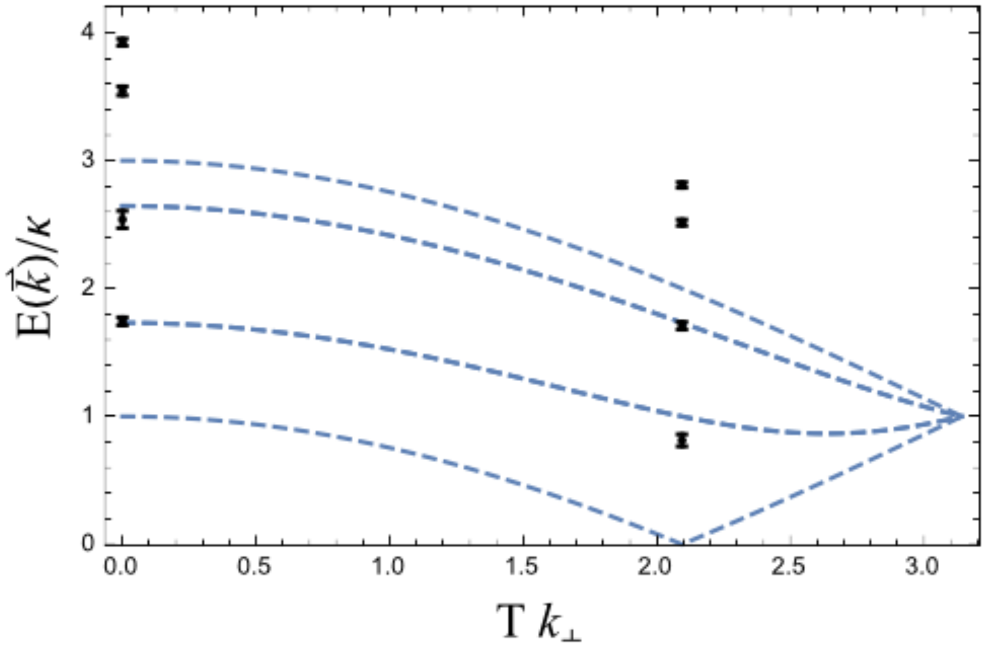


9 unit cells

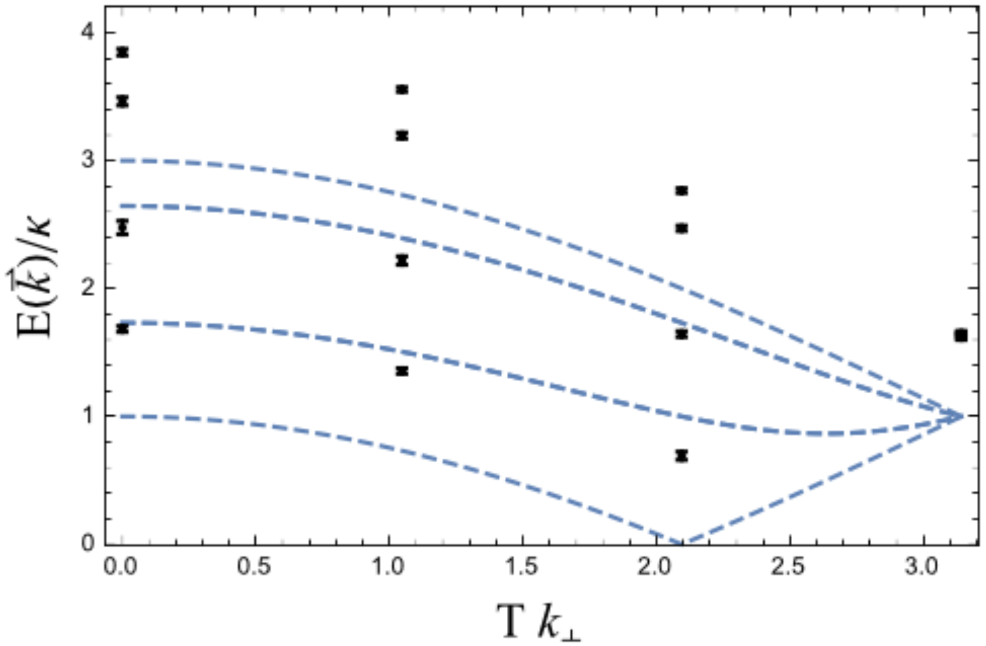


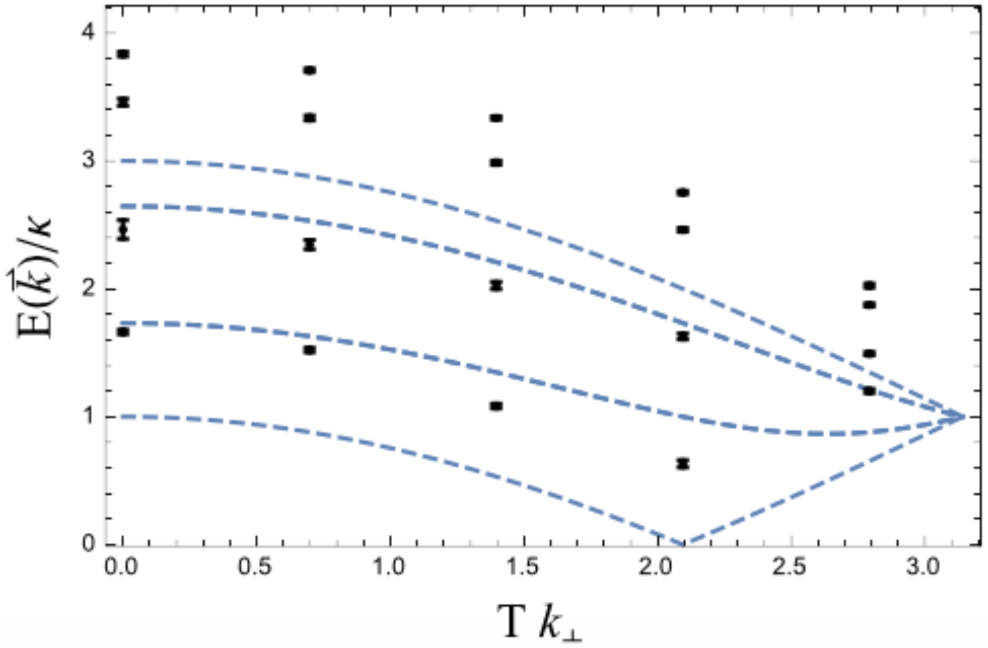
Extrapolated (but finite T) spectrum:
(3,3) nanotube ...

3 unit cells

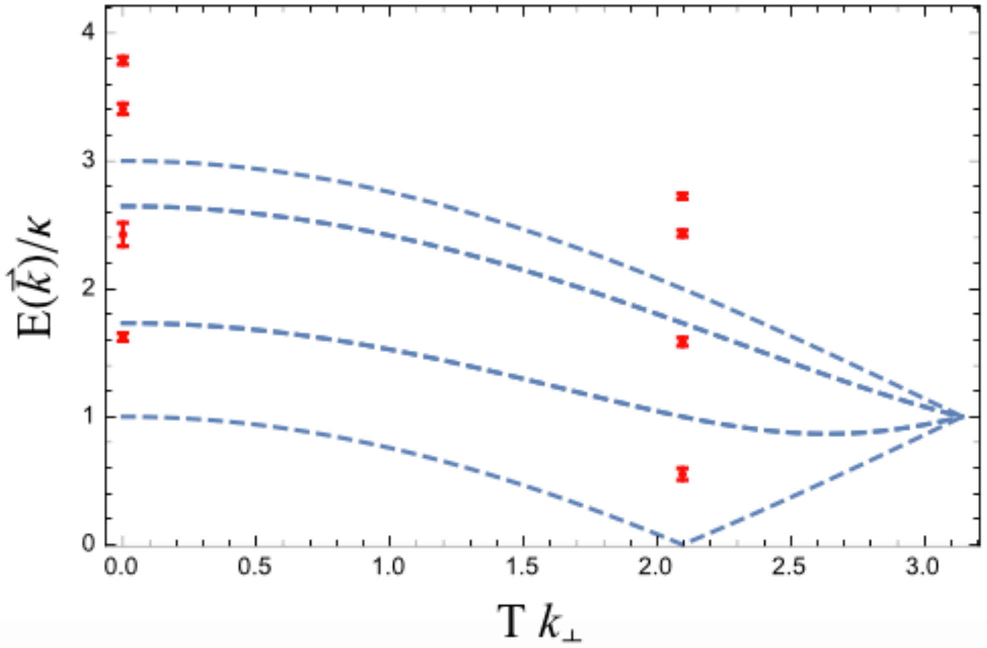


6 unit cells





9 unit cells

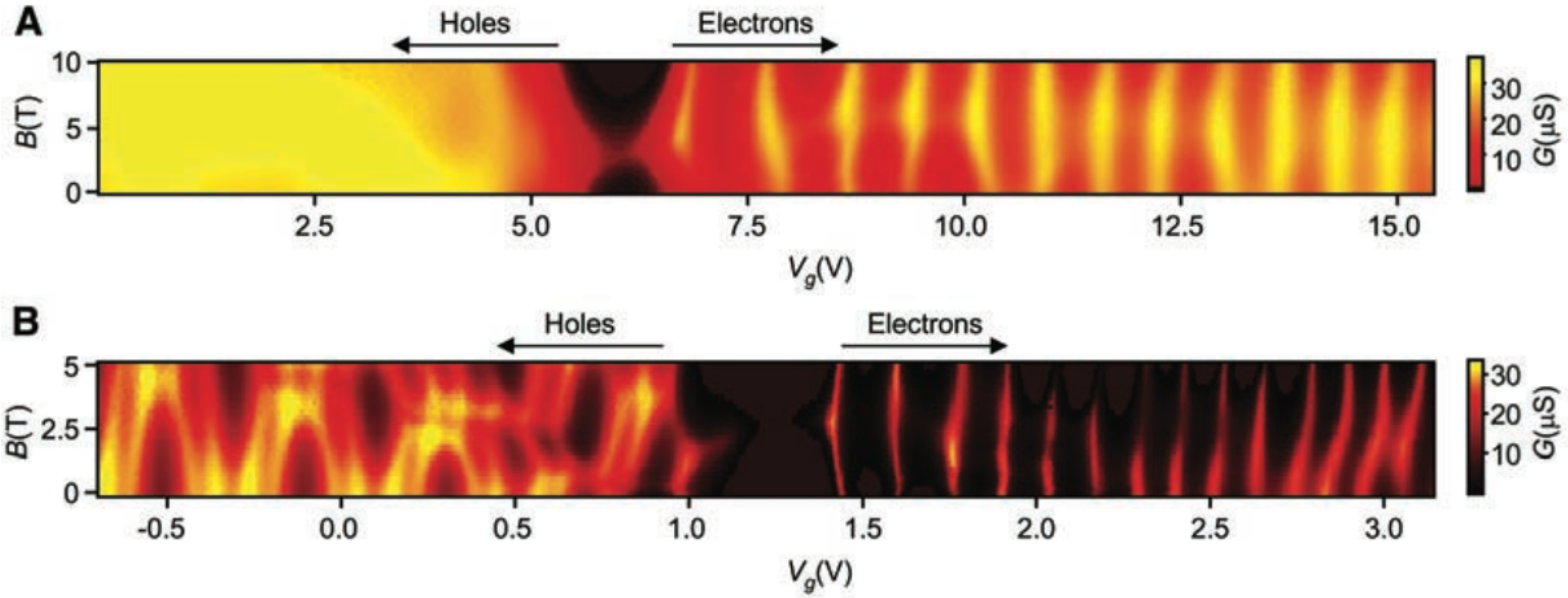


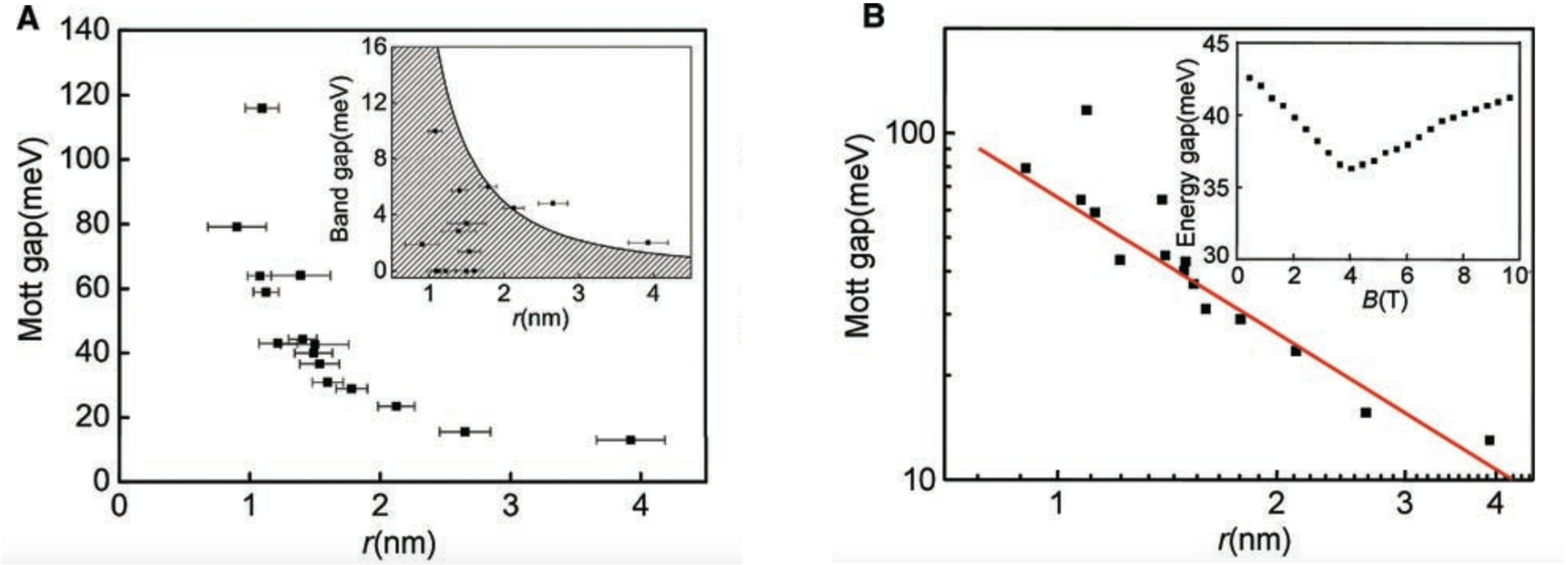
Spectrum is (too?) strongly modified by interactions

Experimental situation:

Tentative (?) observation of Mott insulating state …

V. V. Deshpande et al.,
*Science **323**, (2009) 106*





Preliminary results for (small-radius) nanotubes ...

(14,14), r = 0.95 nm
(18,18), r = 1.22 nm

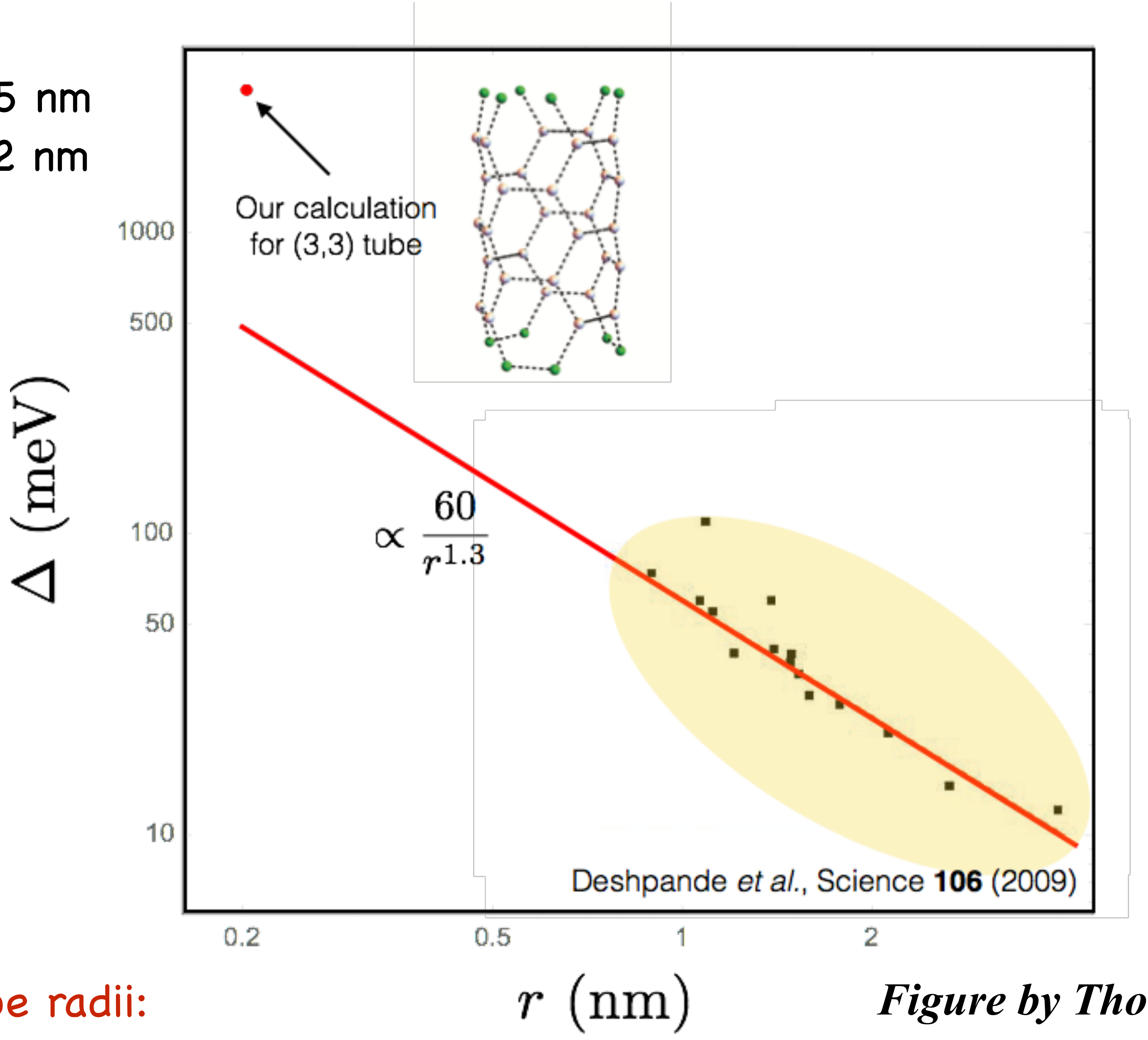


Figure by Thomas Luu

Need larger tube radii:
- Curvature effects on TB
- Comparison with experiment