Graphene Wrinkles

Antonio H. Castro Neto

What is unique about Graphene ?

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Is Diracness unique to Graphene ?

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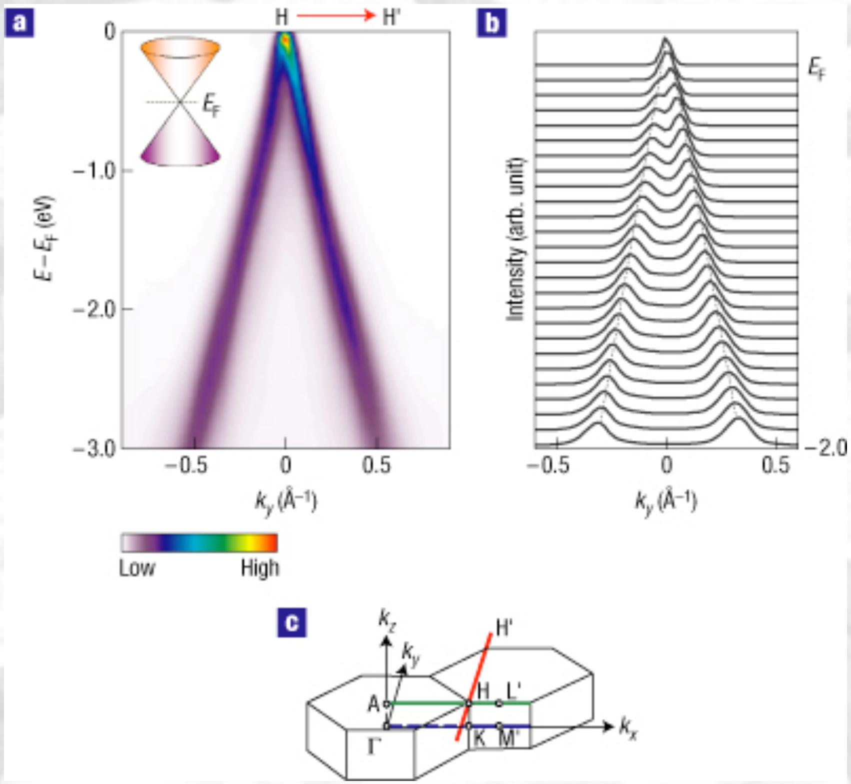

Zhou et al, Nature Physics 2, 595 - 599 (2006)

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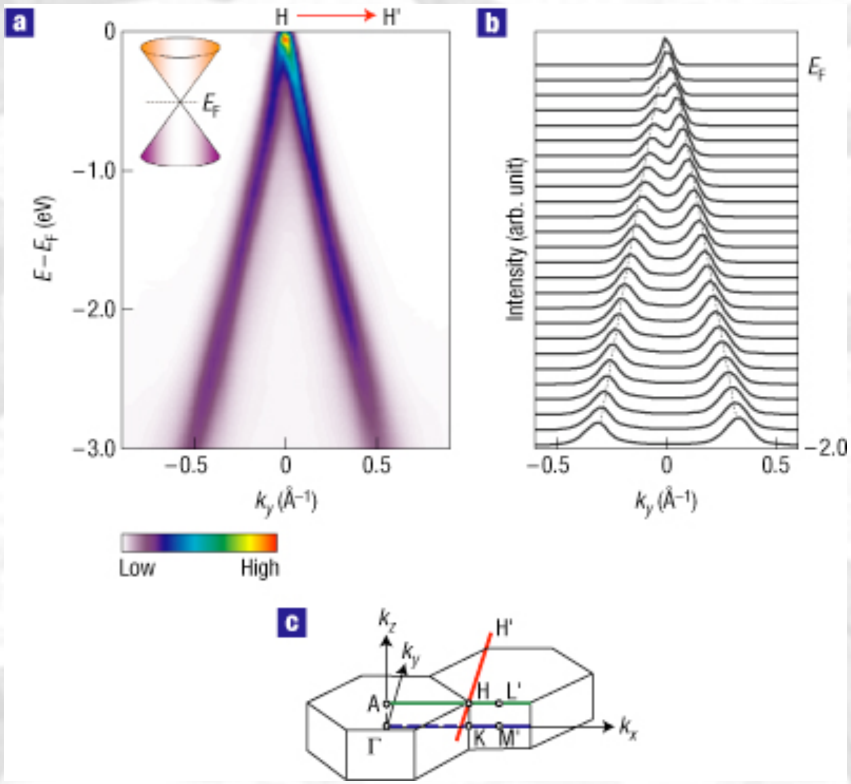
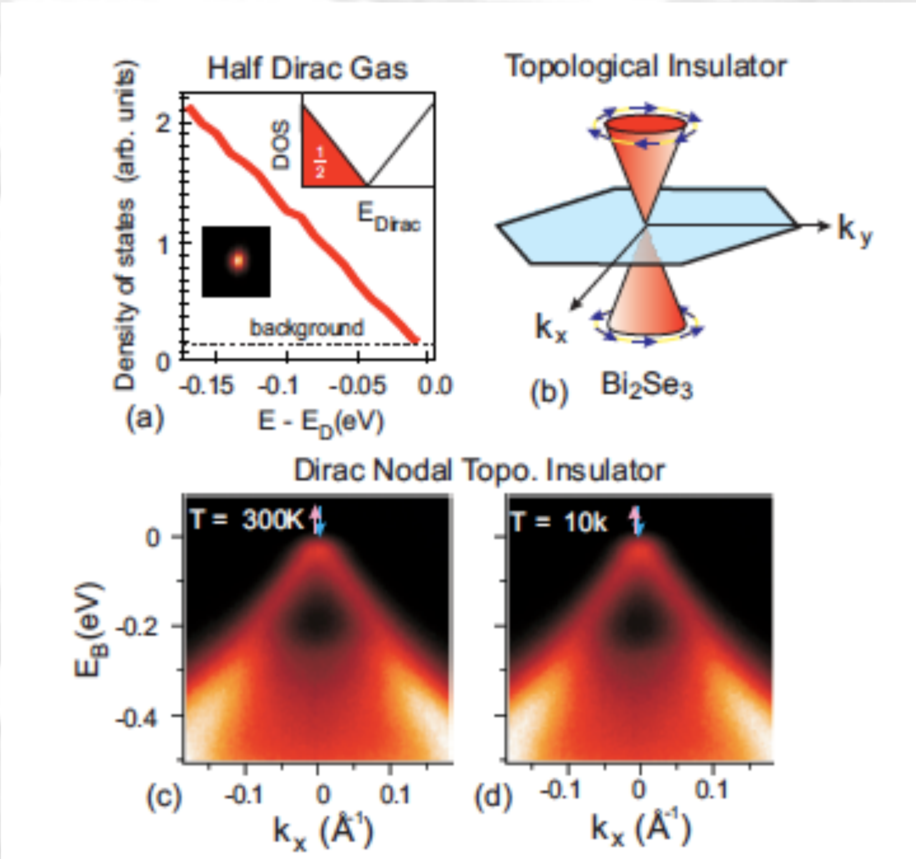


Zhou et al, Nature Physics 2, 595 - 599 (2006)



Hsieh et al, Nature 460, 1101 (2009).

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Lenoir et al. Transport properties of Bi-rich Bi–Sb alloys 91

J. Phys. Chem. Solids Vol. 57, No. 1, pp. 89–99, 1996

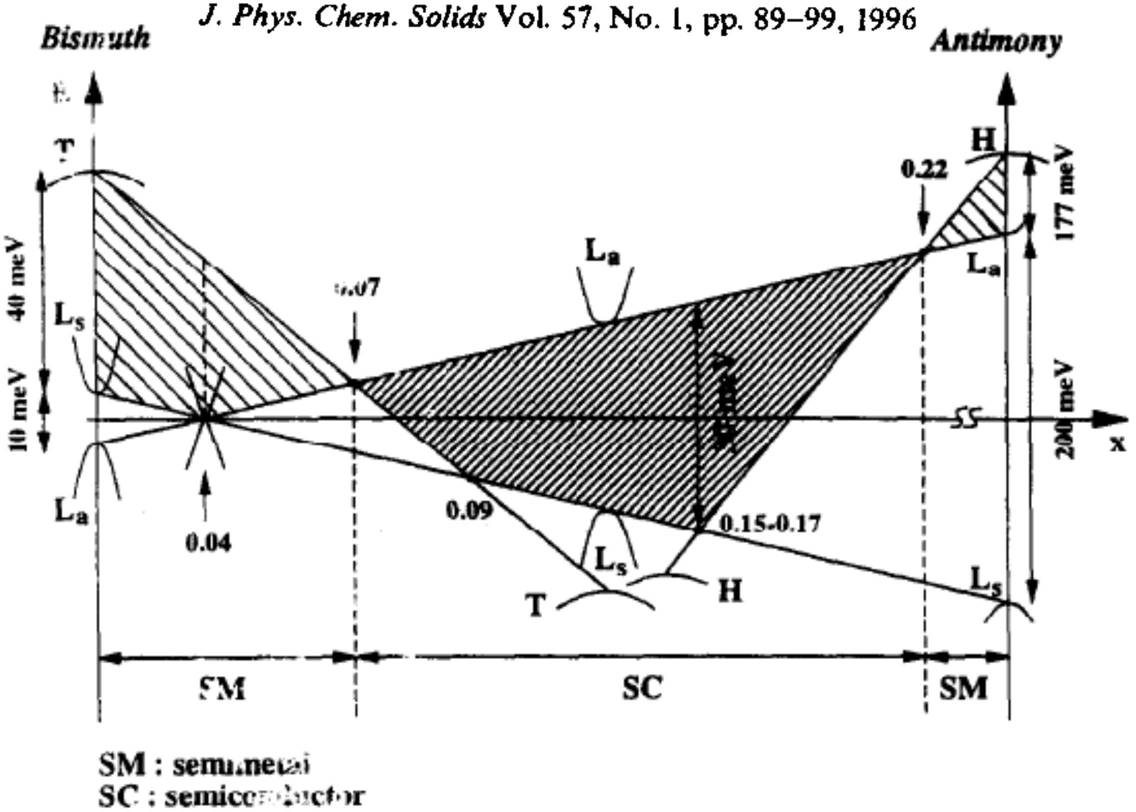
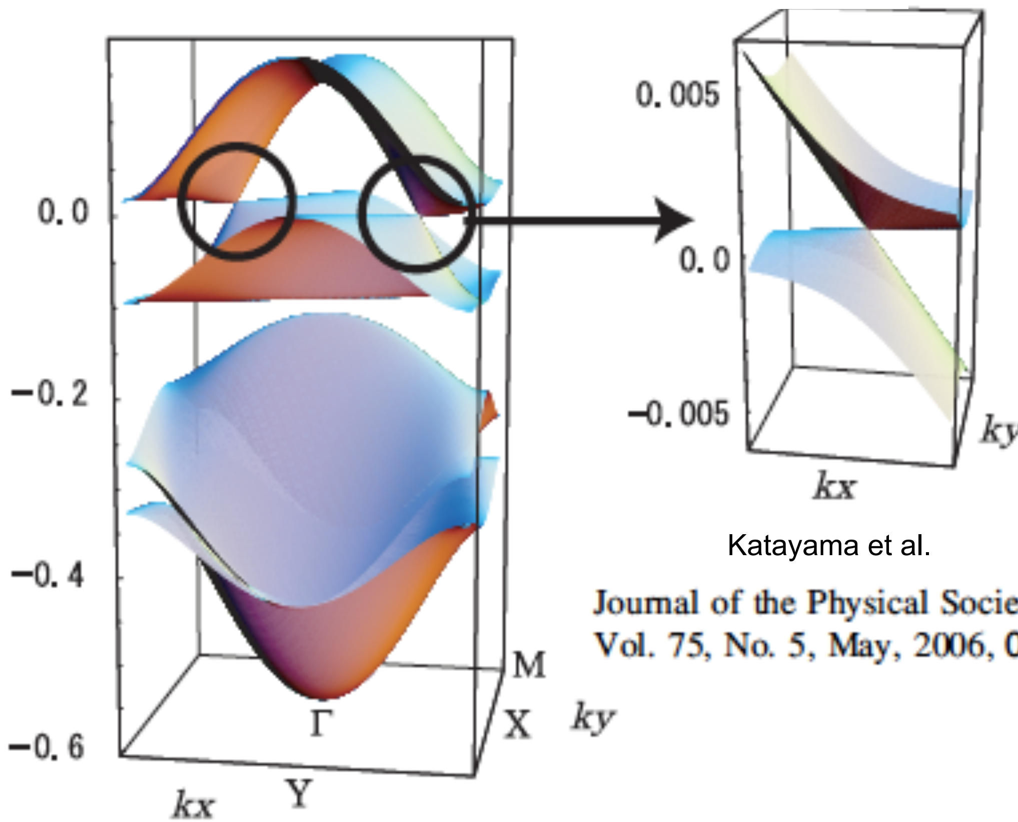


Fig. 1. Schematic representation of the energy bands near the Fermi level for $Bi_{1-x}Sb_x$ alloys as a function of x at 0 K. For simplicity, the L-, T- and H-point bands are drawn one on top of the others. Note that Mendez [4] has observed a strong temperature dependence of the L band parameters.

What is unique about Graphene ?

Is Diracness unique to Graphene ?

Pressure-Induced Zero-Gap Semiconducting State in Organic Conductor α-(BEDT-TTF)$_2$I$_3$ Salt



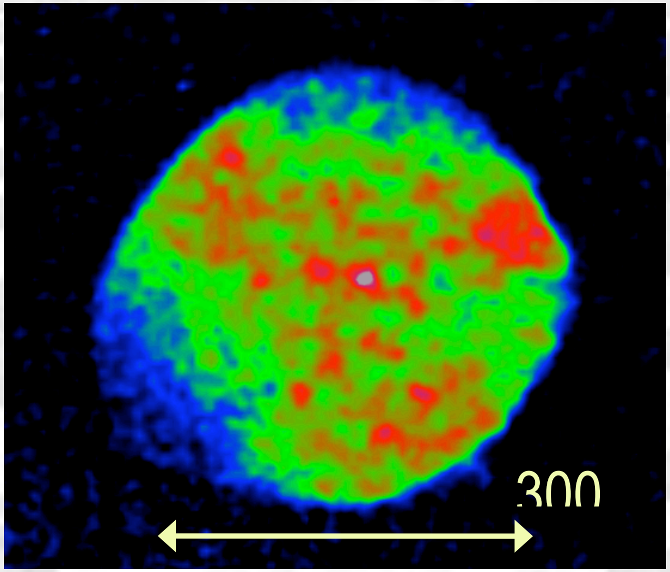
Katayama et al.

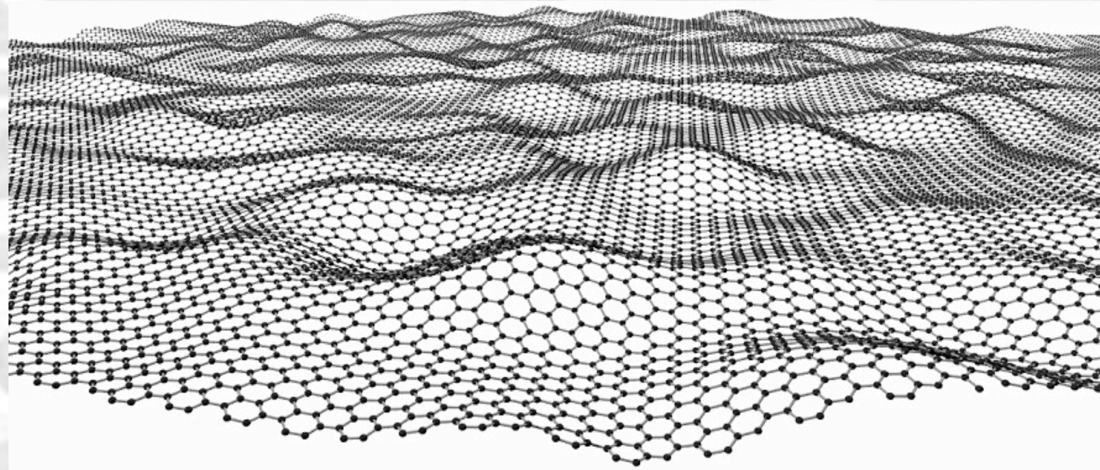
Journal of the Physical Society of Japan
Vol. 75, No. 5, May, 2006, 054705

Fig. 2. Band dispersion of α-(ET)$_2$I$_3$ at $P_a = 4$ kbar in first Brillouin zone (left panel) and enlarged one around contact point at $k^0 = (0.602\pi, -0.353\pi)$ (right panel), where Fermi energy is taken as origin.

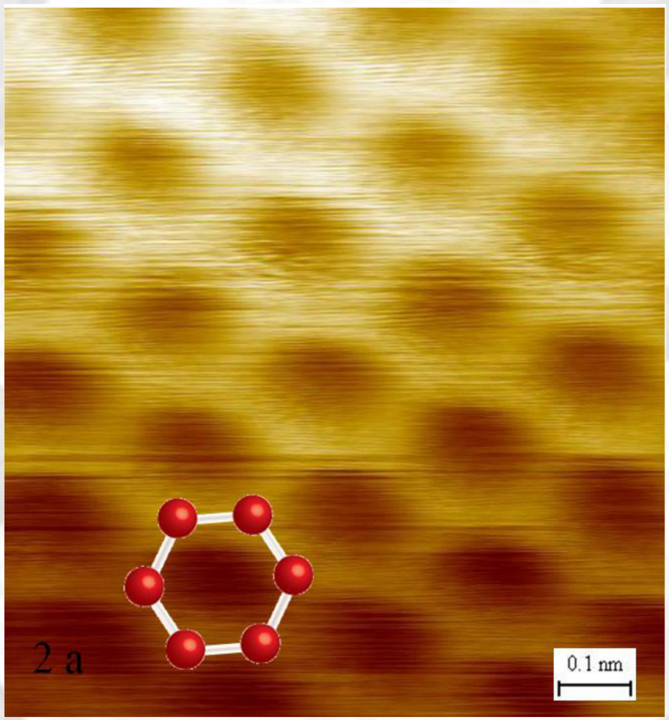
What is unique about Graphene ?

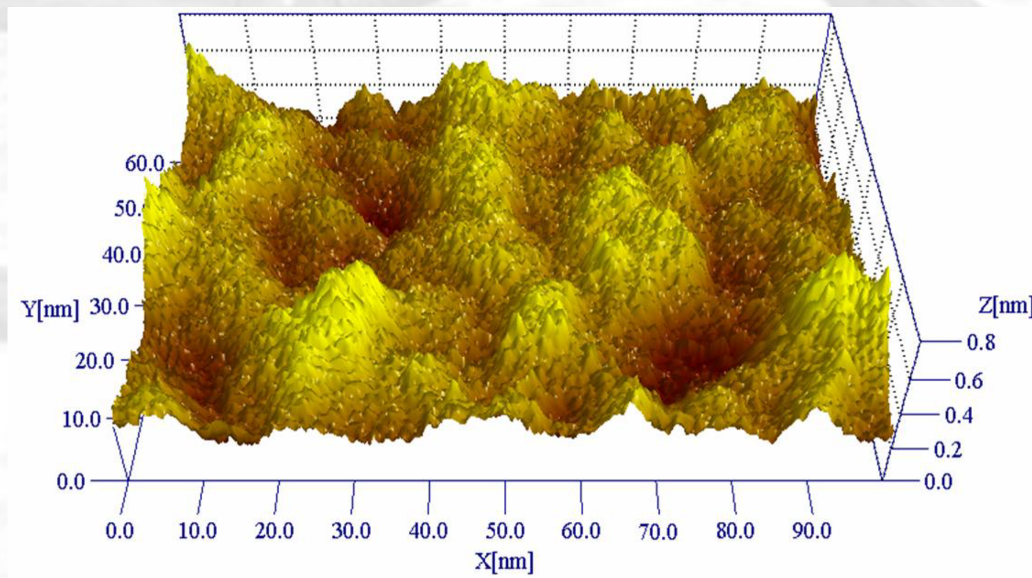
Lesson # 1: Graphene is soft !





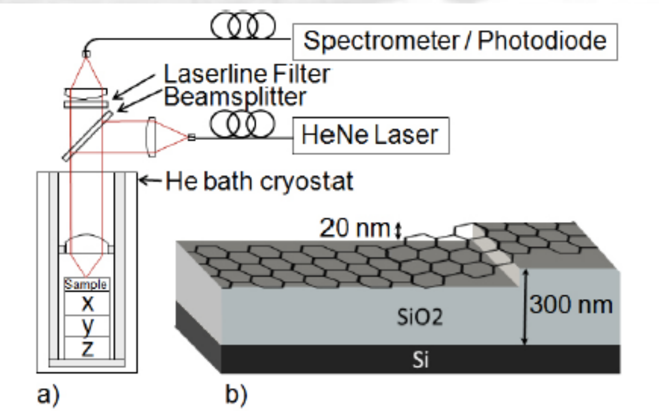
J. C. Meyer *et al.*, Nature **446**, 60 (07)

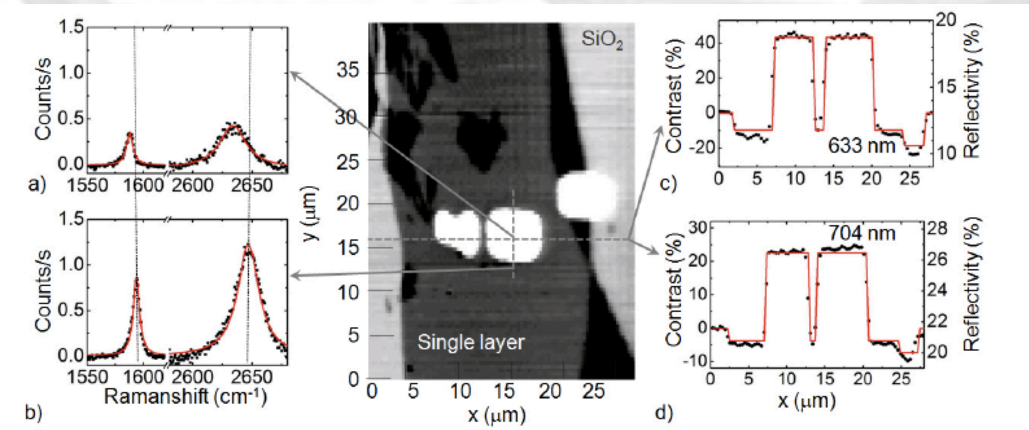


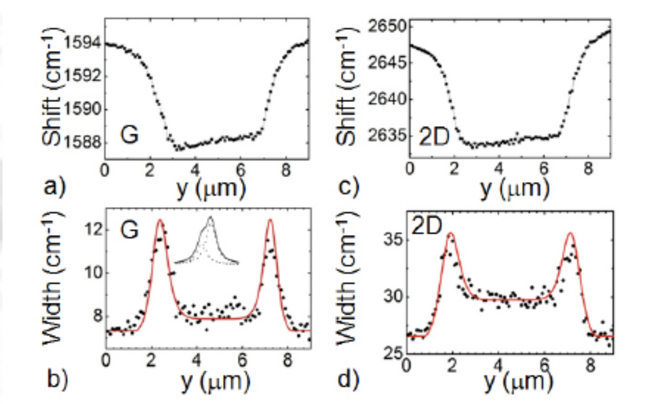


E. Stolyarova et al., PNAS 104, 9209 (2007)
Ishigami et al., Nano Letters (2007)

The physical properties are affected by its softness







[Biaxial Strain in Graphene Adhered to Shallow Depressions]

Constanze Metzger, Sebastian Rmi, Mengkun Liu, Silvia V. Kusminskiy, Antonio H. Castro Neto, Anna K. Swan and Bennett B. Goldberg
Nano Lett., **2010**, *10* (1), pp 6–10

Graphene Folds !

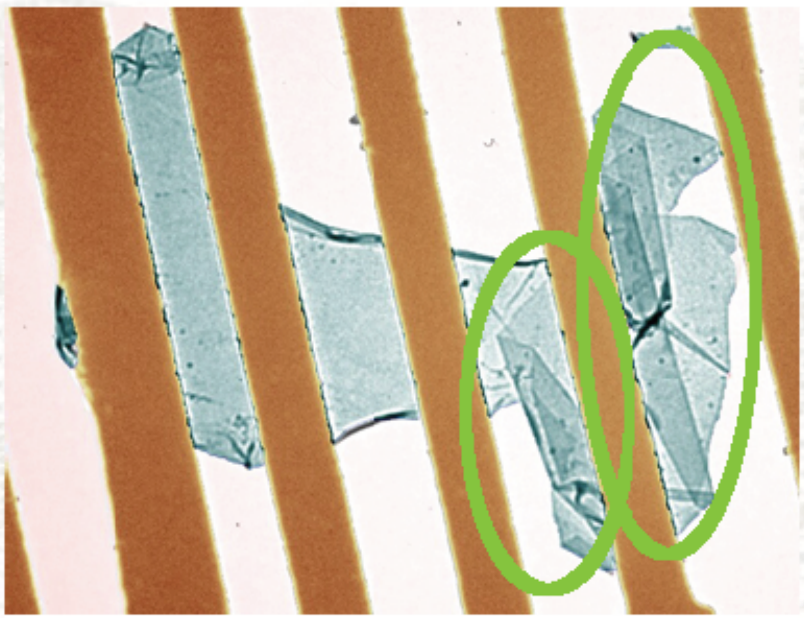


Fig. 3: Suspended graphene. Meyer *et al*., Nature.

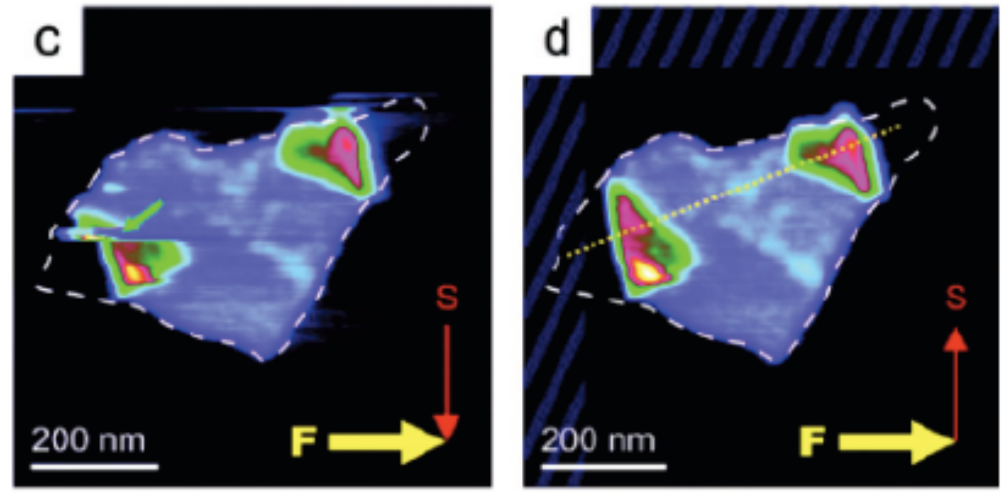


Fig. 4: AFM of graphene folds. Schniepp *et al*.Nano.

Softness is not a nuisance. It can be put to good use !

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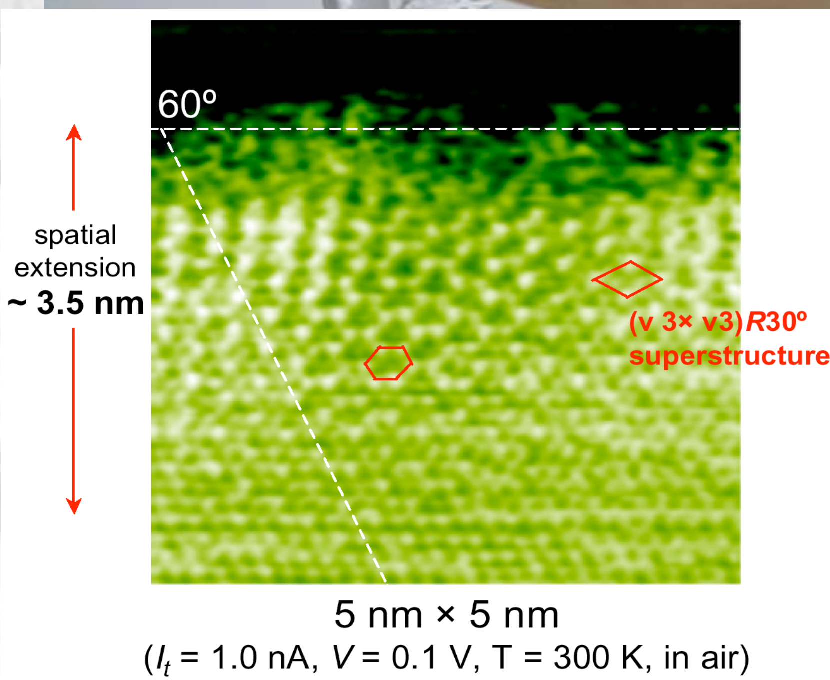
Paper-cutting

Origami

Softness is not a nuisance. It can be put to good use !



Rough edges are a “killer” for graphene nanotechnology !

Origami

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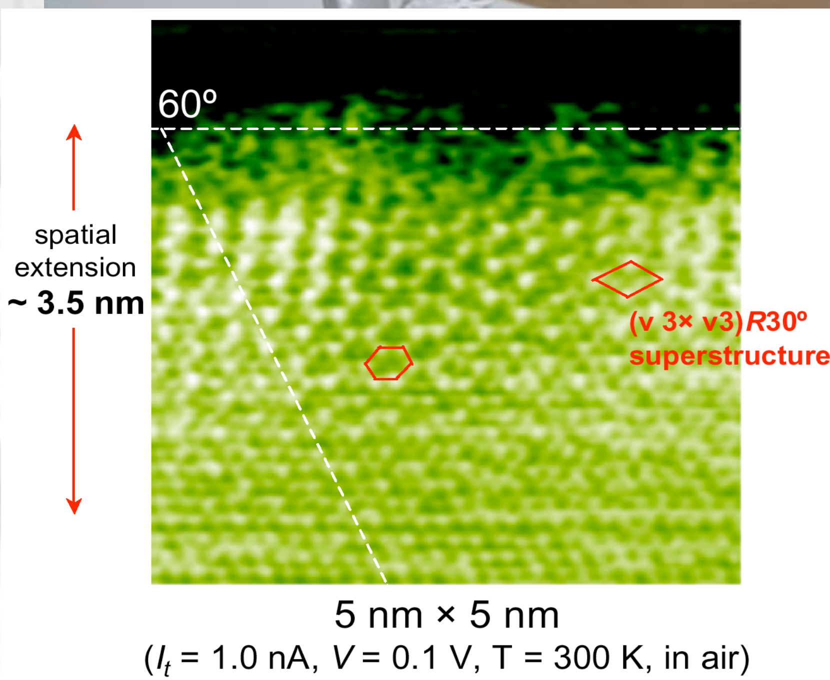
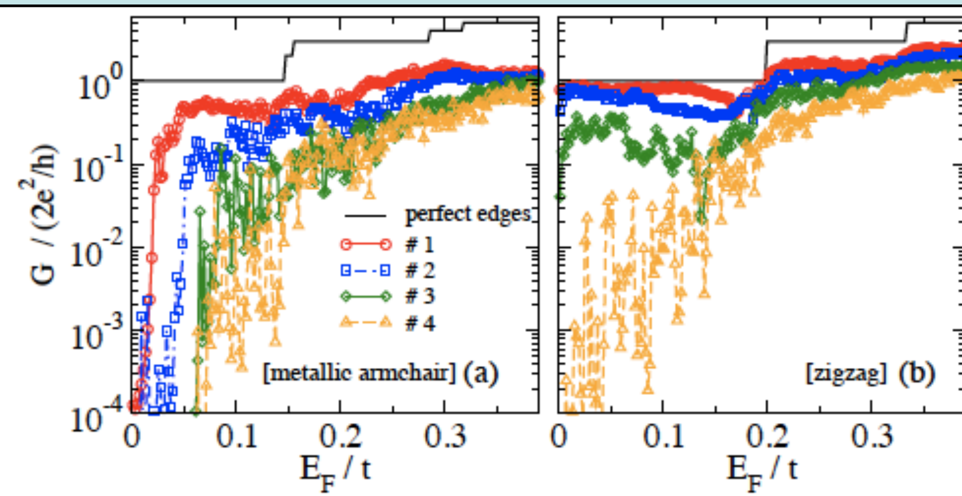


C. Lewenkopf, E.Mucciolo, AHCN, Phys. Rev. B 77, 081410R (2008)
E. Mucciolo, AHCN, C. Lewenkopf, Phys. Rev. B 79, 075407 (2009)



Rough edges are a “killer” for graphene nanotechnology !

Lesson #2: Electrons are affected by the strain, shear and bending deformations of graphene.

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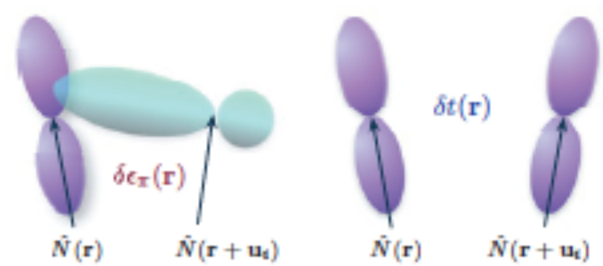
Graphene as an electronic membrane

EPL, 84 (2008) 57007
doi: 10.1209/0295-5075/84/57007

Eun-Ah Kim[1] and A. H. Castro Neto[2(a)]



(b) (c)

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Graphene as an electronic membrane

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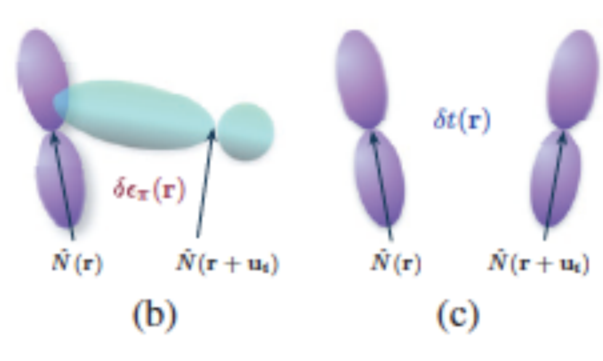
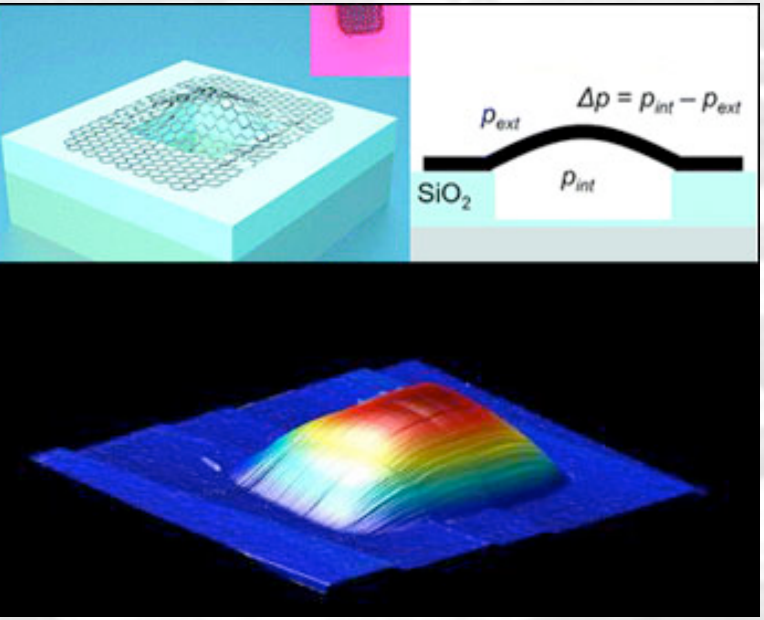
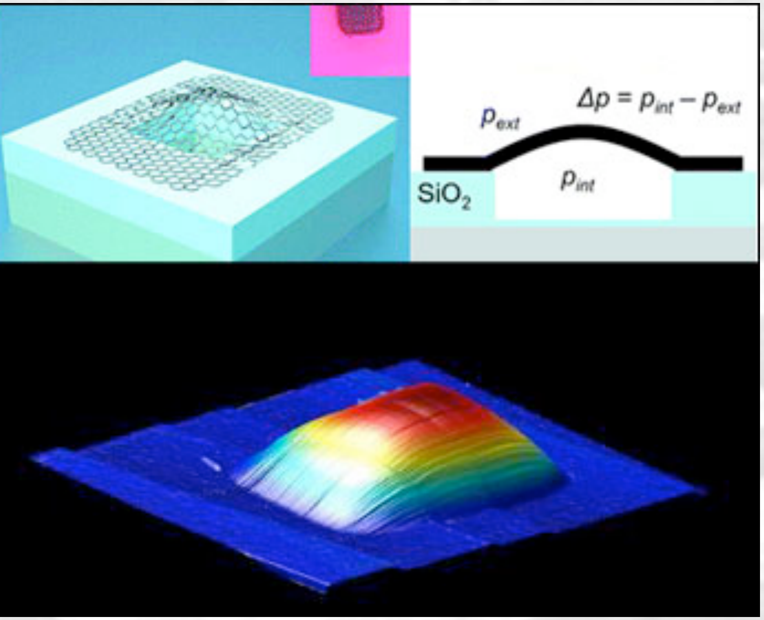
EUN-AH KIM[1] and A. H. CASTRO NETO[2(a)]



$$H=\int d\mathbf{r}\,\Psi^\dagger(\mathbf{r})\left\{\vec{\sigma}\cdot\left[iv_F\nabla+\vec{A}(\mathbf{r})\right]-\mu+\Phi(\mathbf{r})+U(\mathbf{r})\right\}\Psi(\mathbf{r})$$



J. S. Bunch et al., Nano Lett. 8, 2458 (2008).



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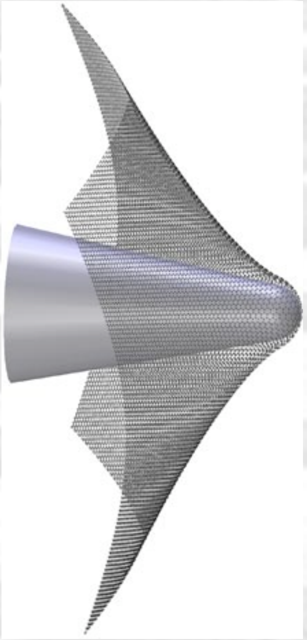
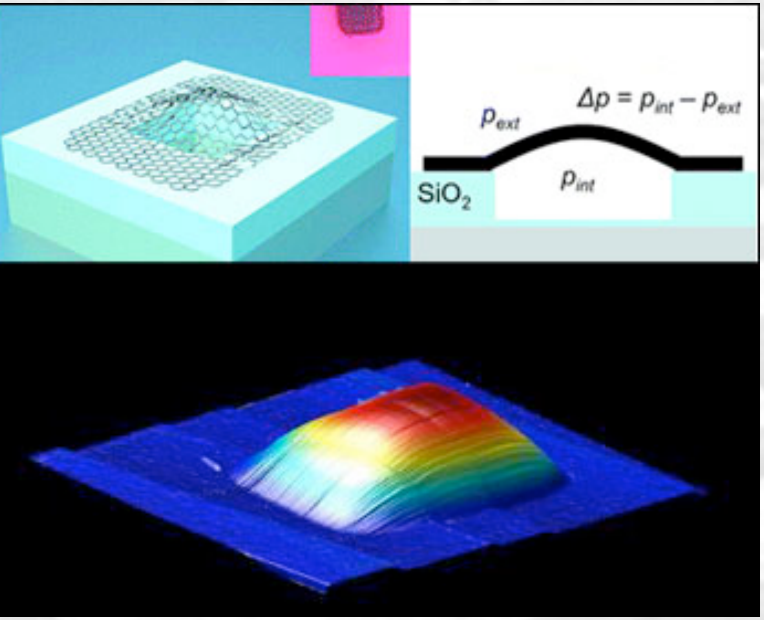


C. Lee et al. Science 321, 325 (2008)



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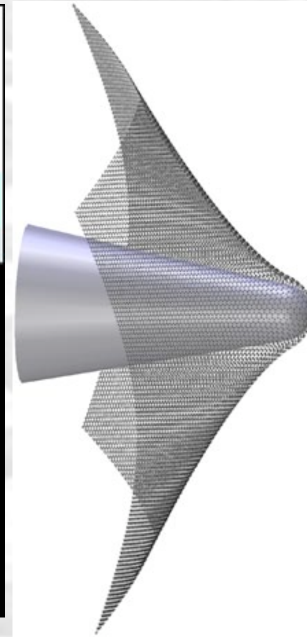

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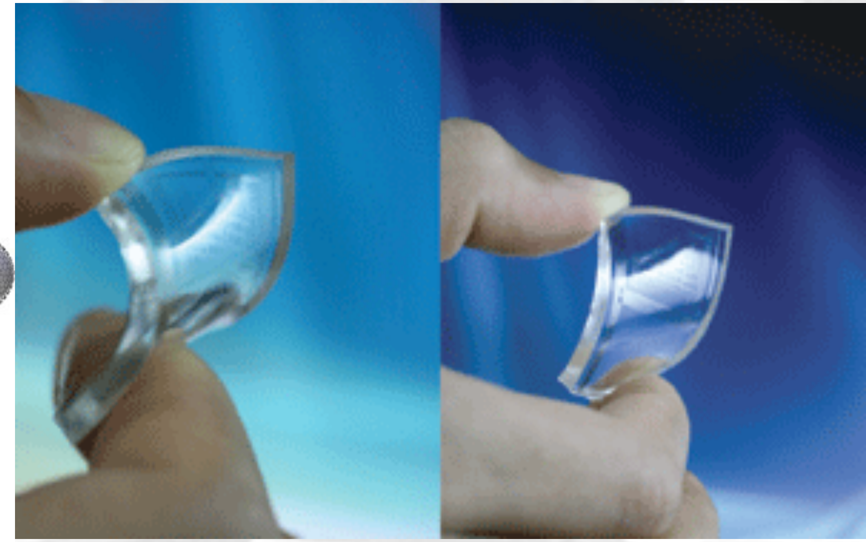
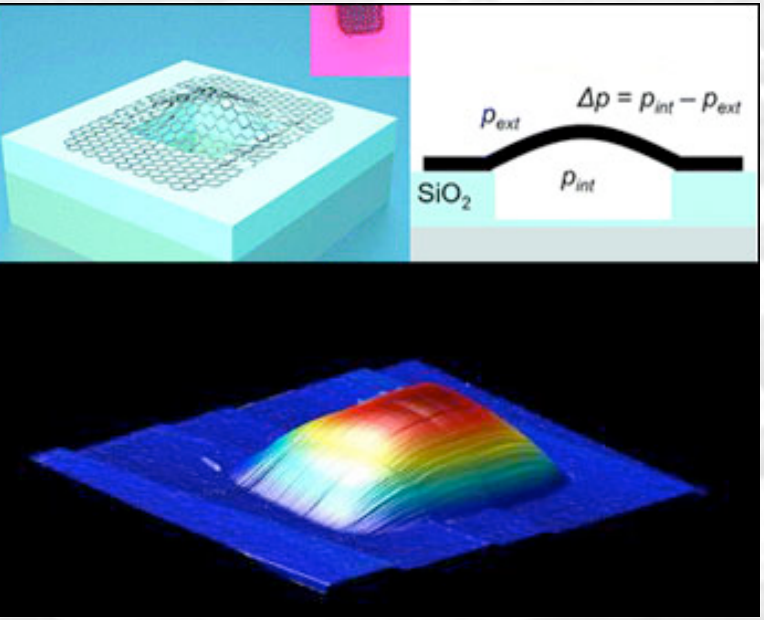


K.S.Kim et al., Nature 2009



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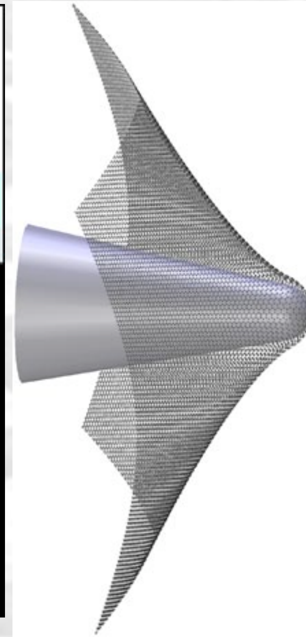

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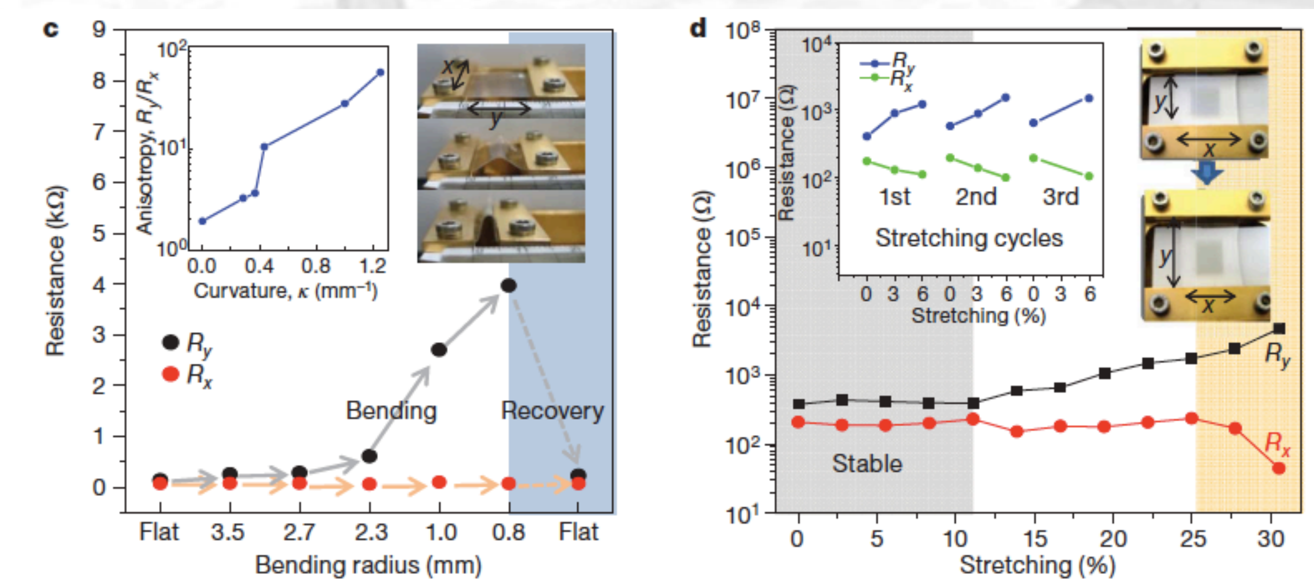

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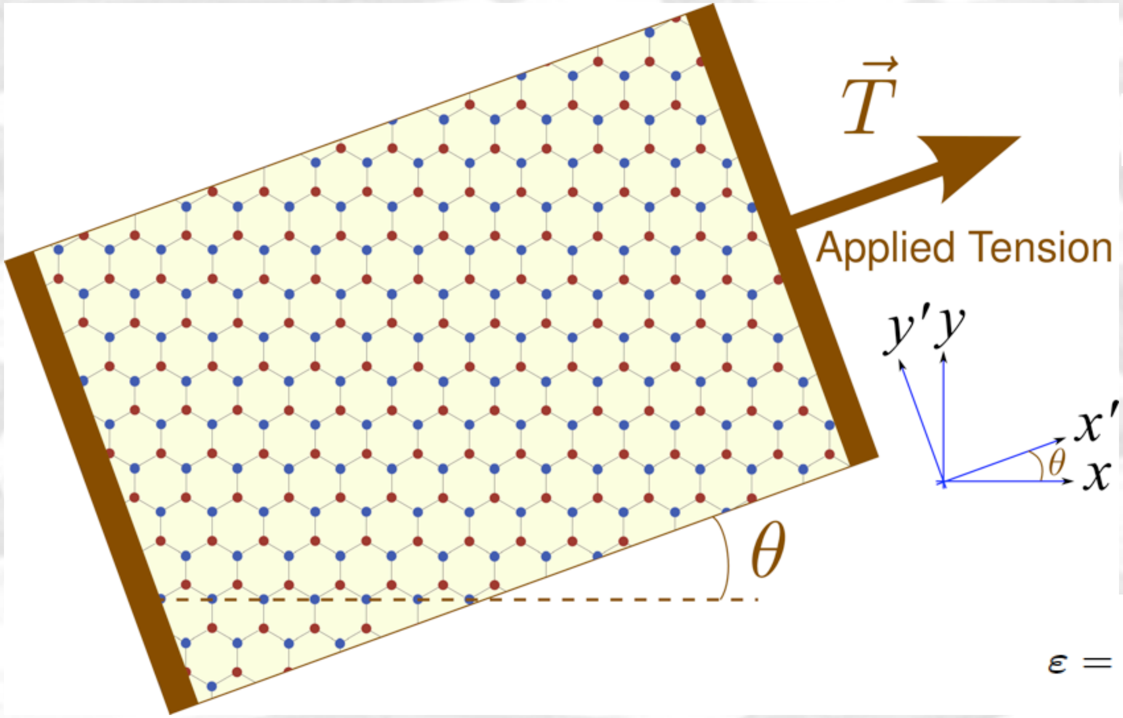
V. Pereira, AHCN, N. M.R. Peres, Phys. Rev. B. 80, 045401 (2009).



$$\boldsymbol{T} = T\boldsymbol{e}_{x'}$$

$$\varepsilon'_{ij} = S_{ijkl}\tau'_{kl} = T\,S_{ijkl}\delta_{kx}\delta_{lx} = T\,S_{ijxx}$$

$$\varepsilon'_{xx} = T\,S_{xxxx}\,,\ \varepsilon'_{yy} = T\,S_{xxyy}$$

$$\varepsilon = T\,S_{xxxx} \qquad \sigma = -S_{xxxy}/S_{xxxx}$$

$$\sigma = 0.165$$

$$\boldsymbol{\varepsilon}' = \varepsilon \begin{pmatrix} 1 & 0 \\ 0 & -\sigma \end{pmatrix}$$

$$\boldsymbol{T} = T\cos(\theta)\,\boldsymbol{e}_x + \sin(\theta)\,\boldsymbol{e}_y$$

$$\tau_{ij} = C_{ijkl}\,\varepsilon_{kl}\,, \quad \varepsilon_{ij} = S_{ijkl}\,\tau_{kl}$$

$$\boldsymbol{\varepsilon} = \varepsilon \begin{pmatrix} \cos^2\theta - \sigma\sin^2\theta & (1+\sigma)\cos\theta\sin\theta \\ (1+\sigma)\cos\theta\sin\theta & \sin^2\theta - \sigma\cos^2\theta \end{pmatrix}$$

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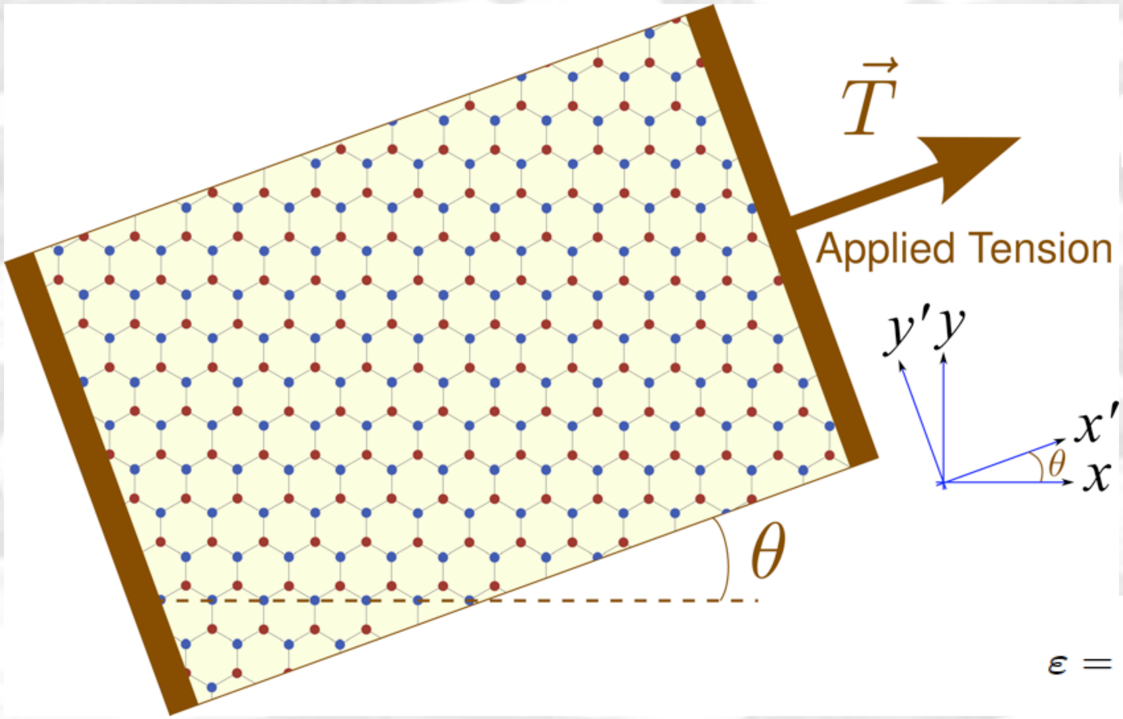

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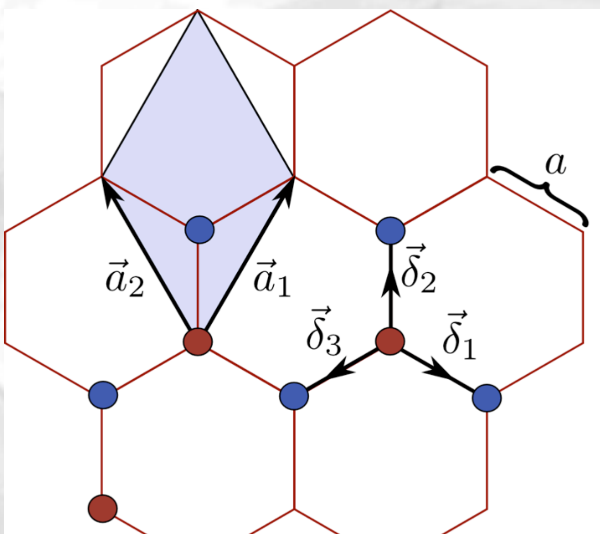


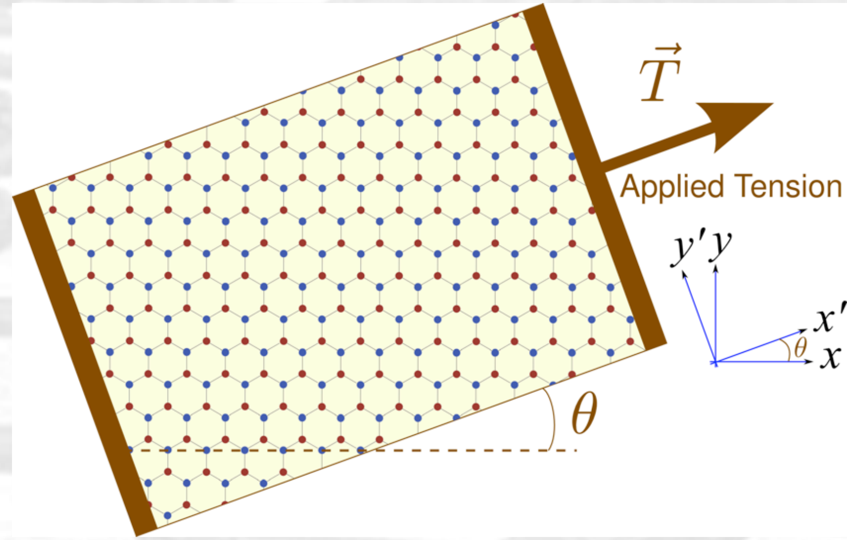
$$\boldsymbol{\delta}_\alpha = (\boldsymbol{1} + \boldsymbol{\varepsilon})\cdot\boldsymbol{\delta}^0_\alpha$$

$$V_{pp\pi}(l) = t_0 e^{-3.37(l-a_0)} \quad a_0 = 1.42\ \text{Å}$$

$$dV_{pp\pi}/dl\ = -6.4\ \text{eV/Å}$$

$$t_0 = t(\boldsymbol{\delta}^0_\alpha) \approx 2.7\text{eV}$$

$$E(k_x, k_y) = \pm \left| t_2 + t_3\, e^{-i\boldsymbol{k}.\boldsymbol{a}_1} + t_1\, e^{-i\boldsymbol{k}.\boldsymbol{a}_2} \right|$$



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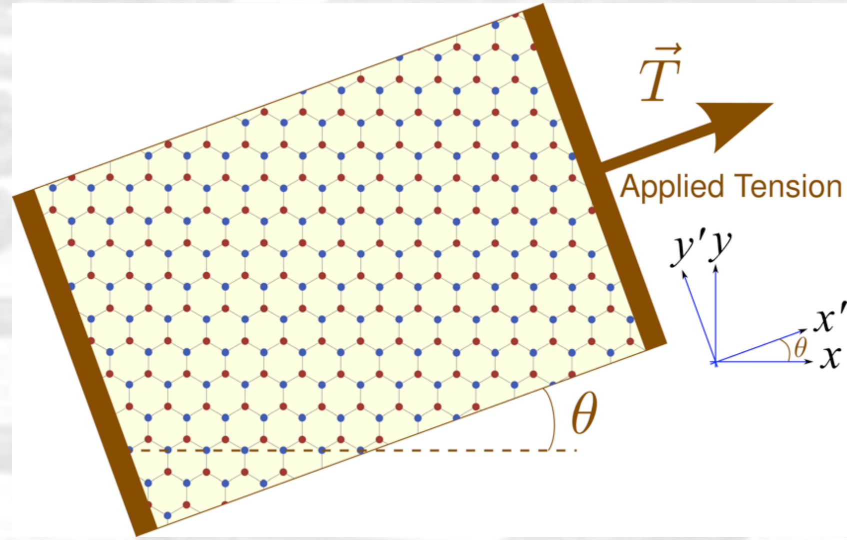
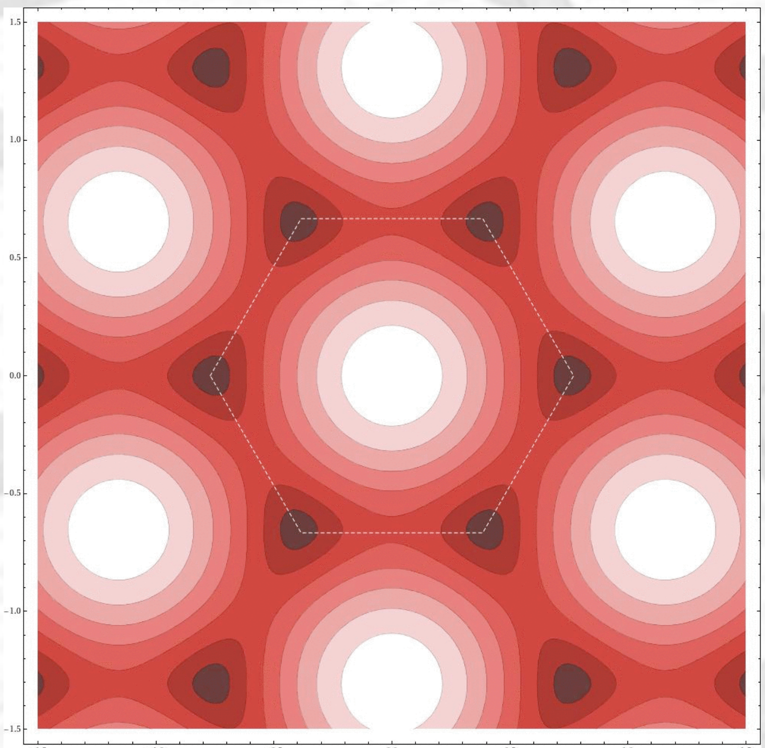
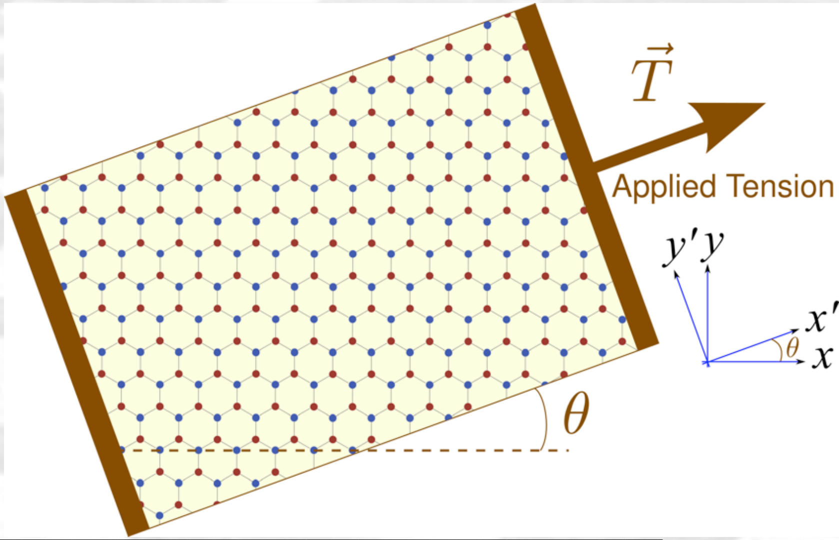
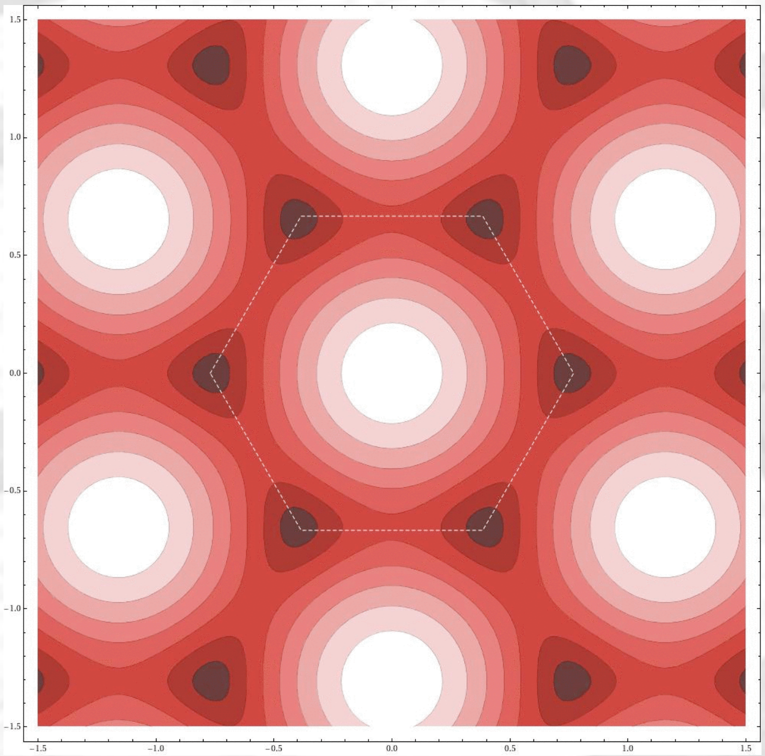




Armchair direction

$$E(k_x, k_y) = \pm \left| t_2 + t_3\, e^{-i\boldsymbol{k}.\boldsymbol{a}_1} + t_1\, e^{-i\boldsymbol{k}.\boldsymbol{a}_2} \right|$$





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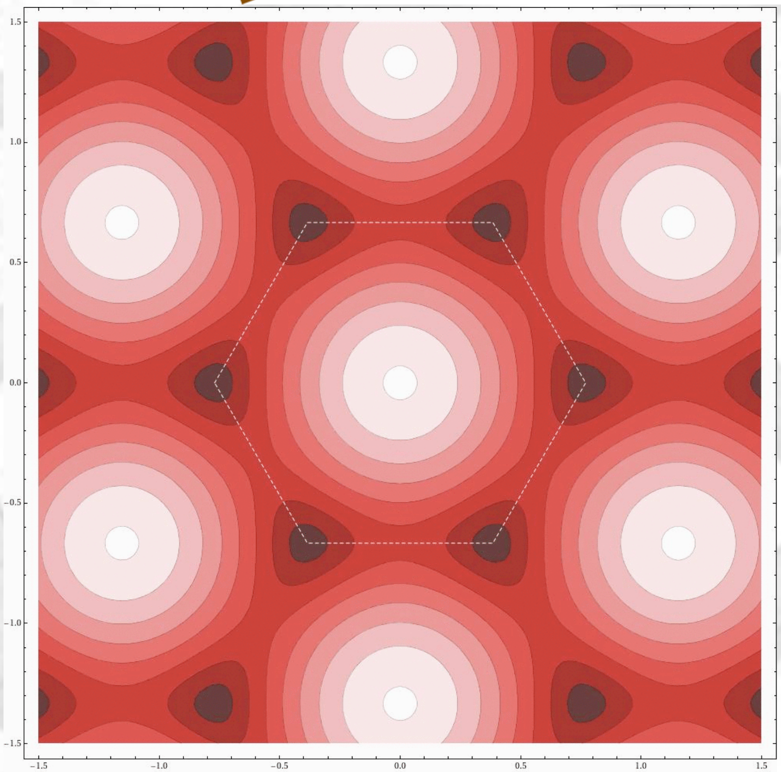
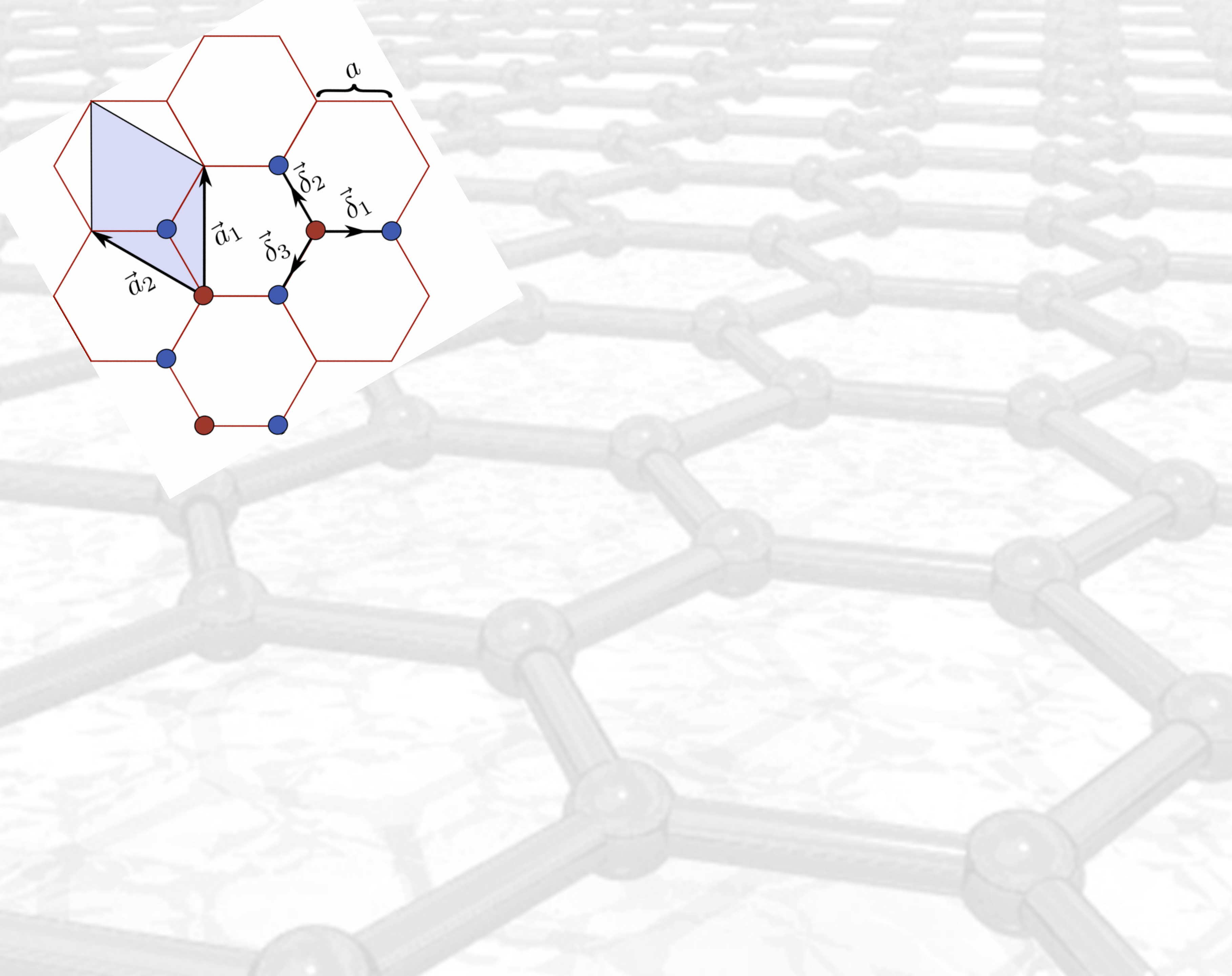


Zigzag direction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$\vec{a}_1$
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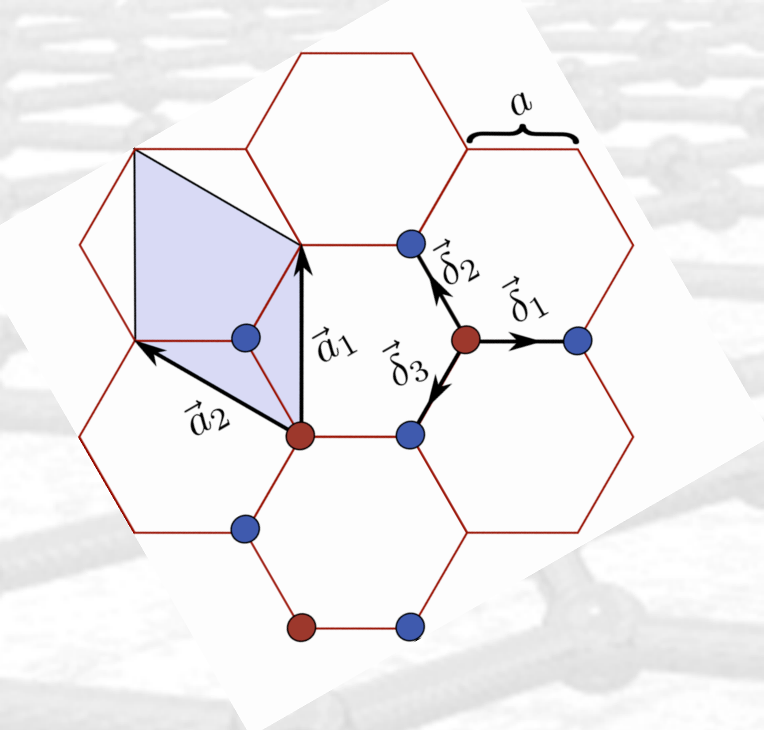
$\vec{a}_2$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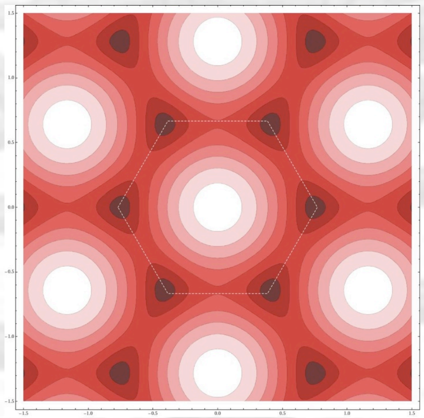

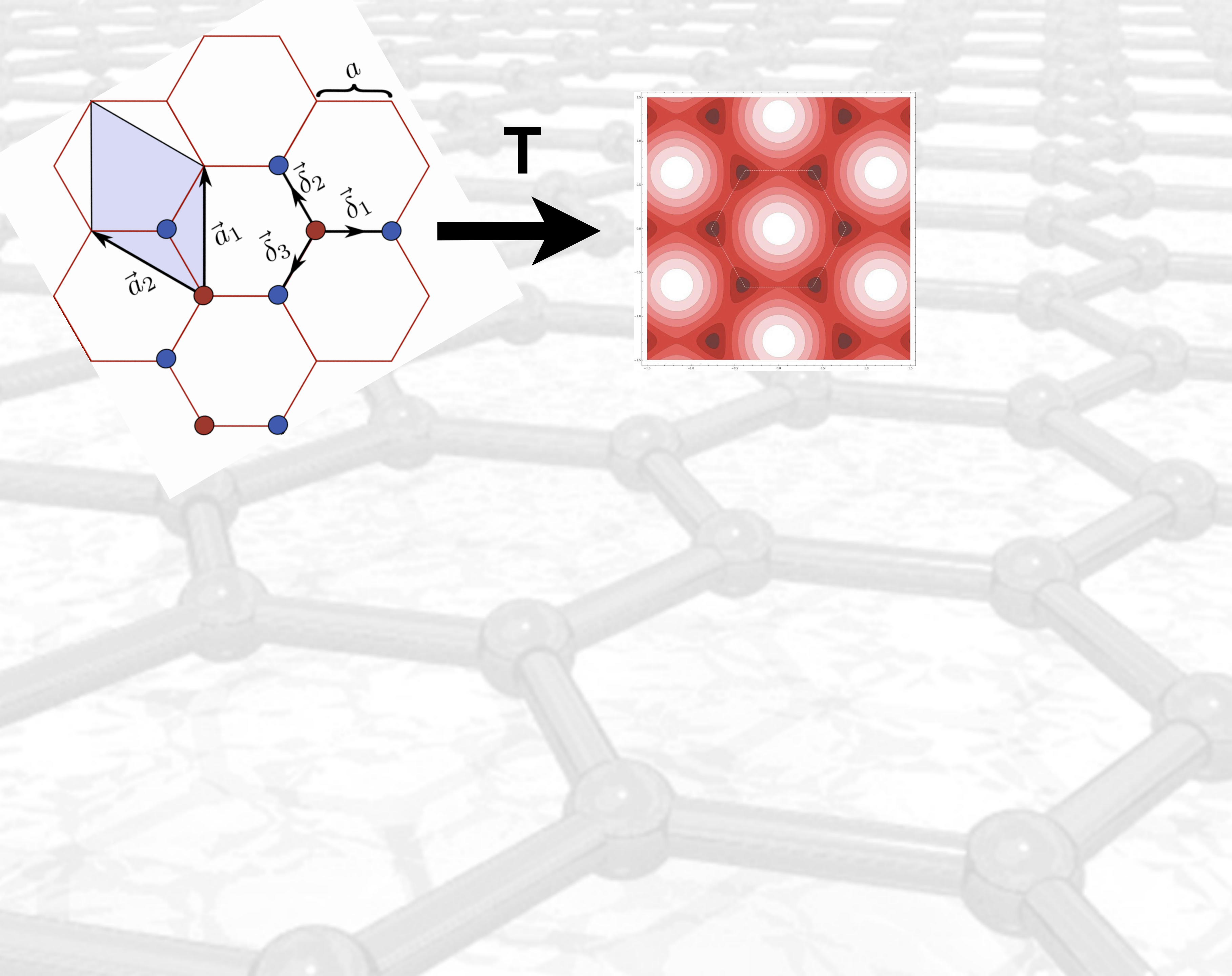

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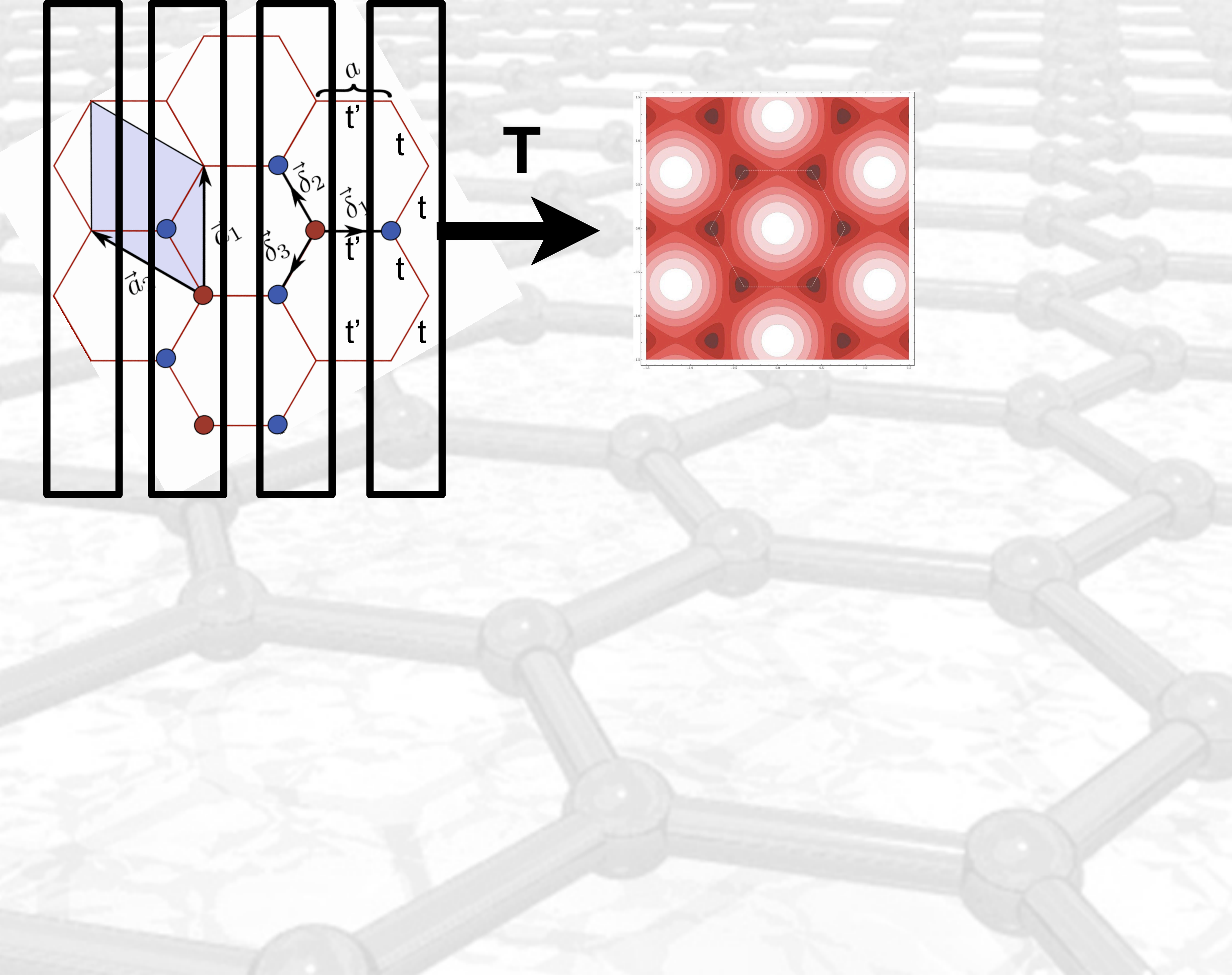

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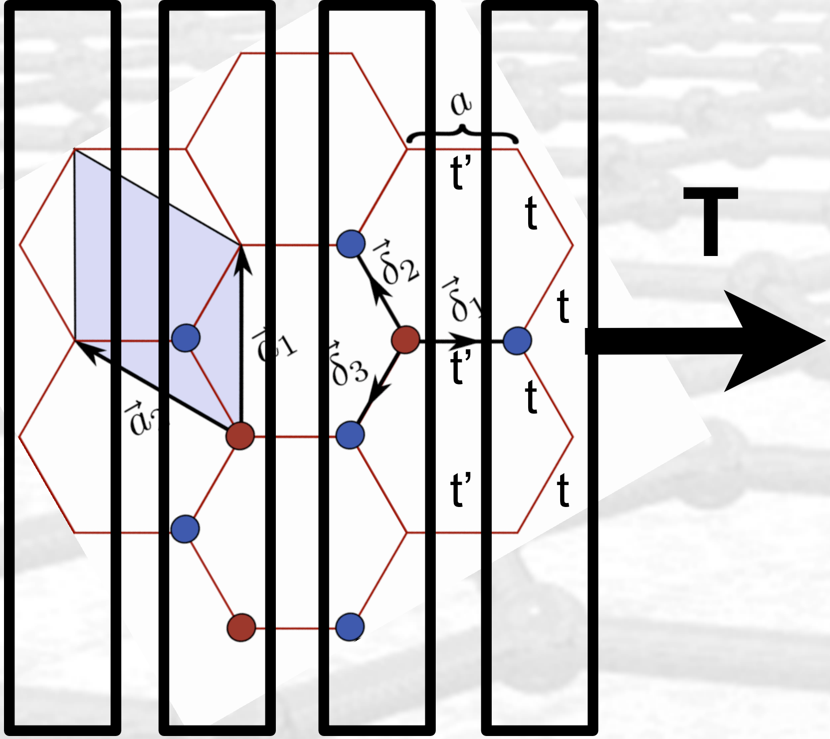
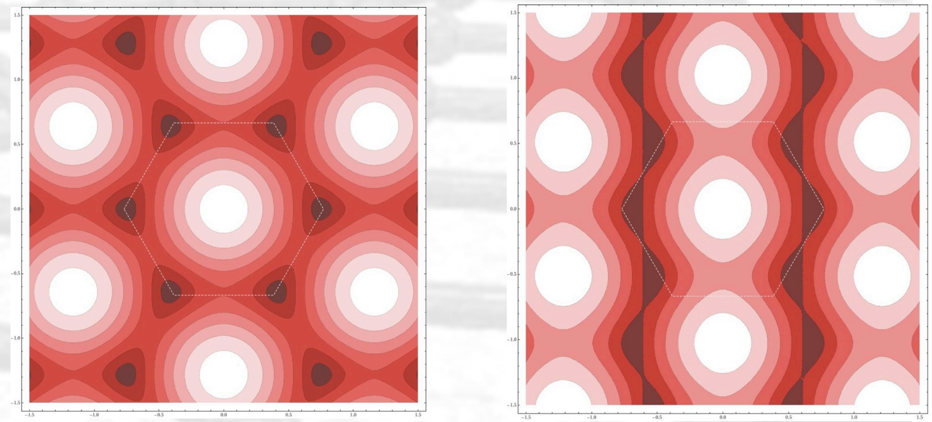


“one-dimensionalization”



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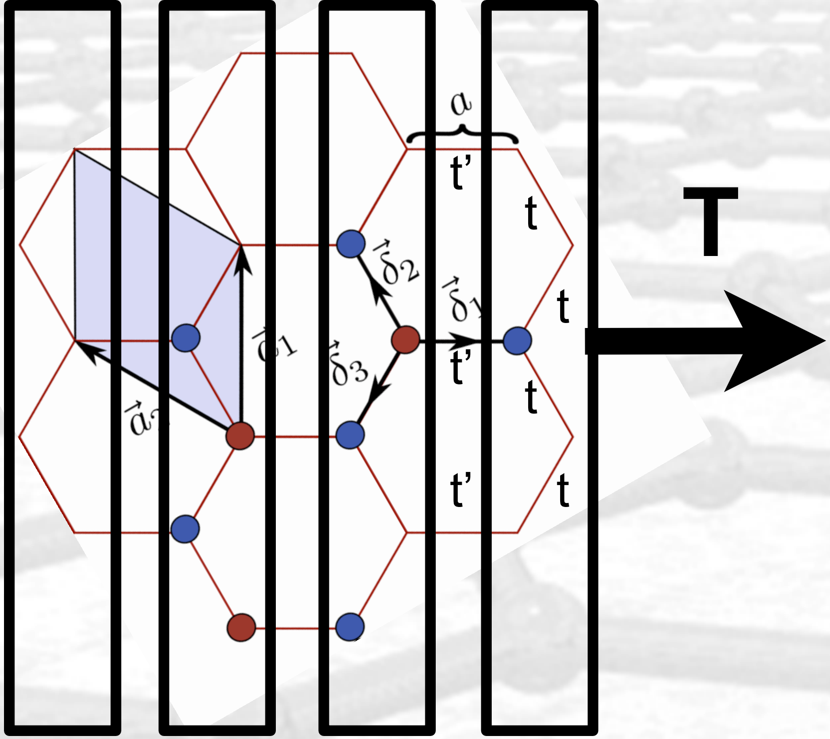
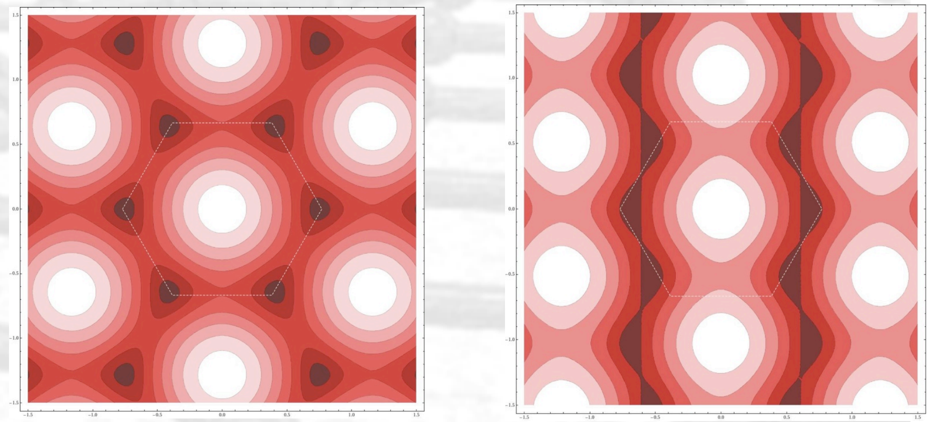
Weakly Coupled Luttinger Liquids

“one-dimensionalization”





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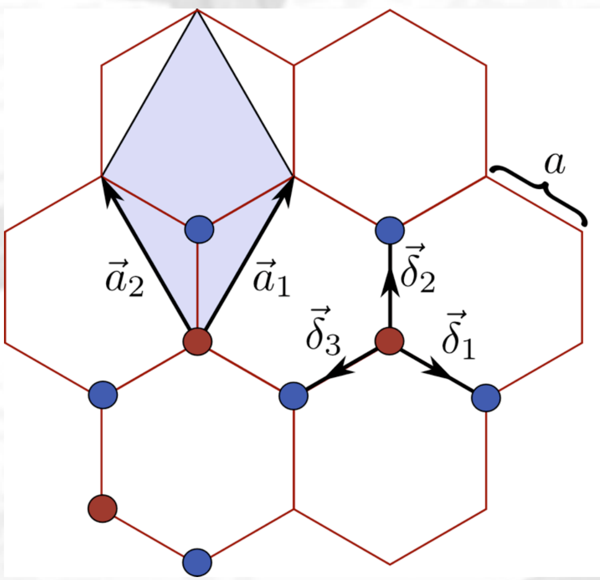

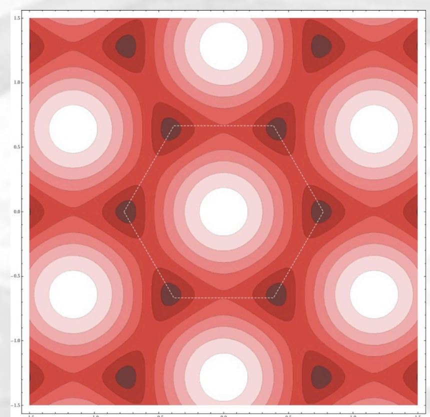

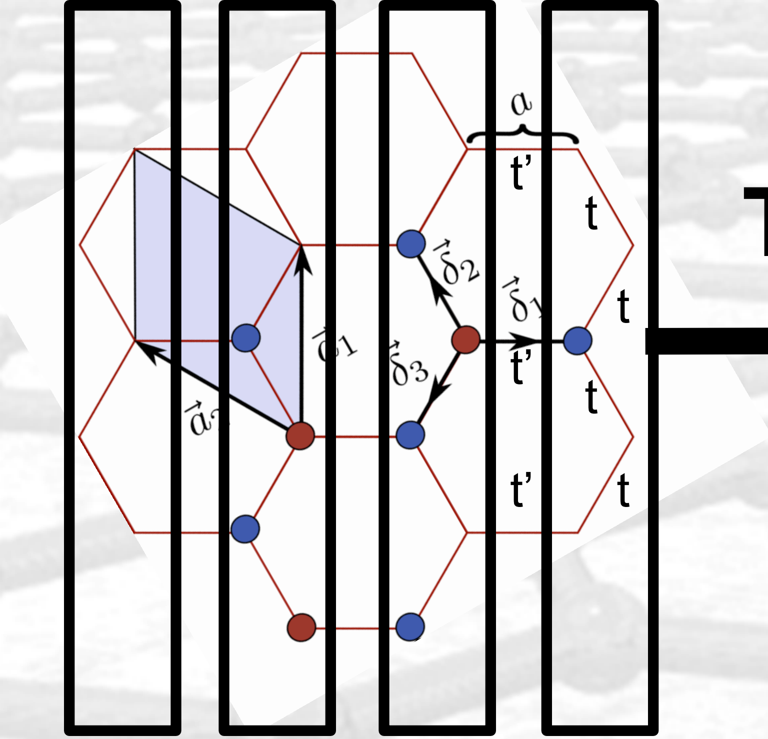

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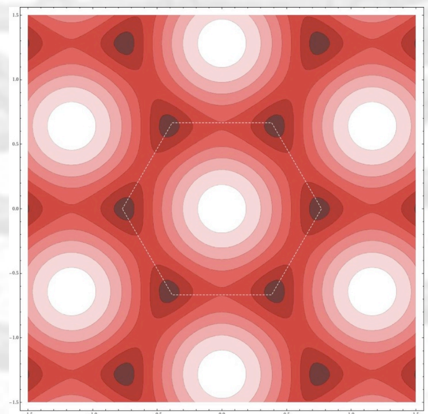

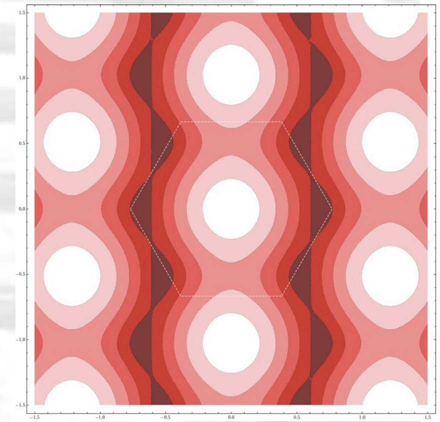

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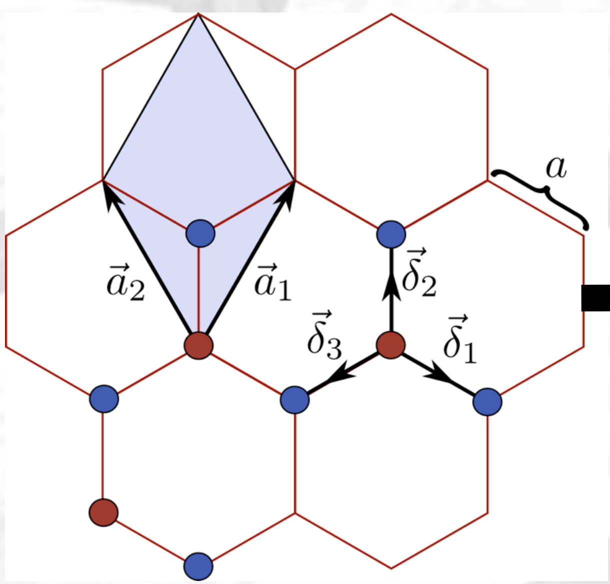

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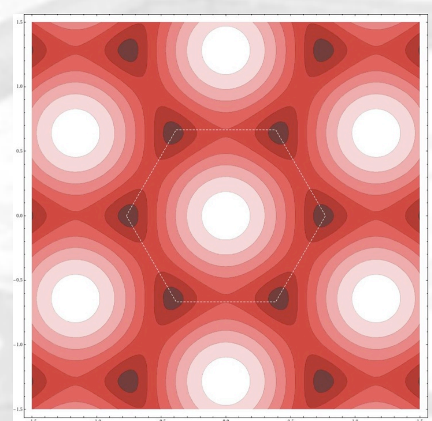
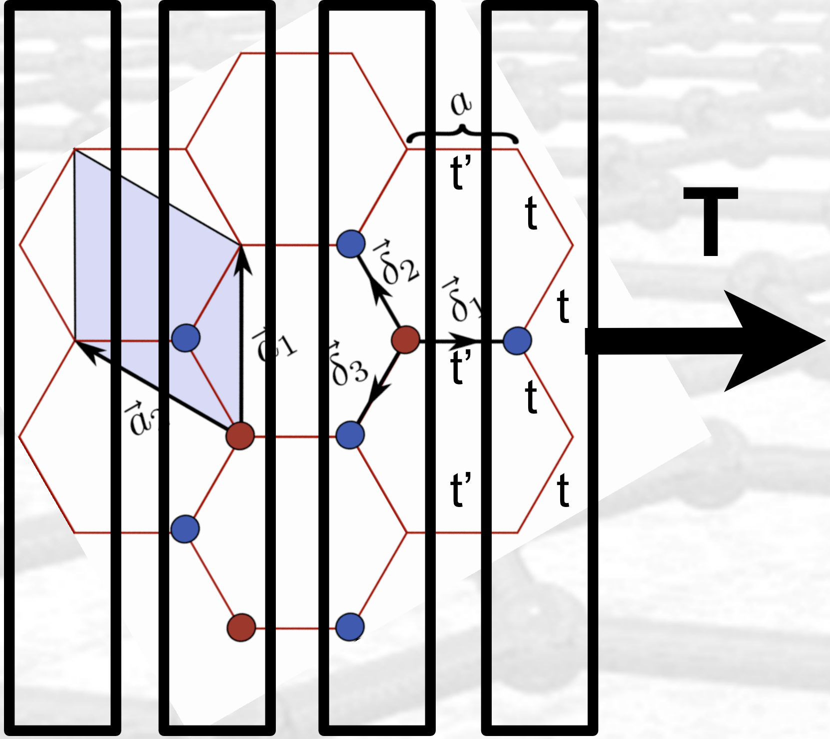
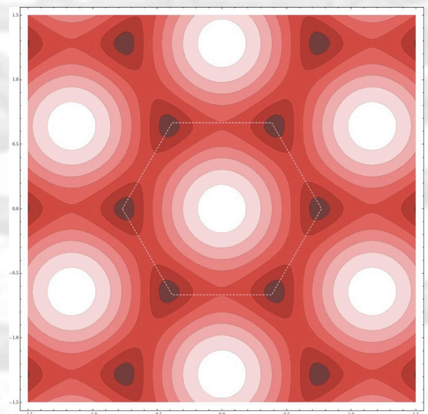
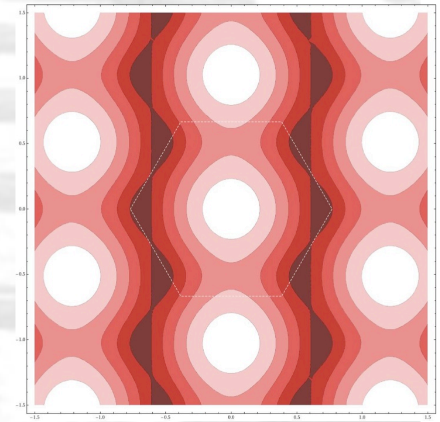


“one-dimensionalization”

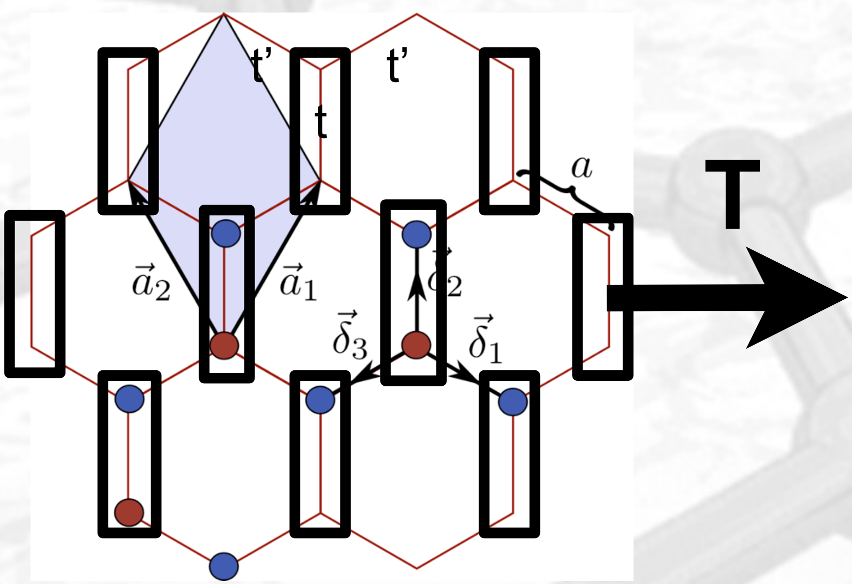


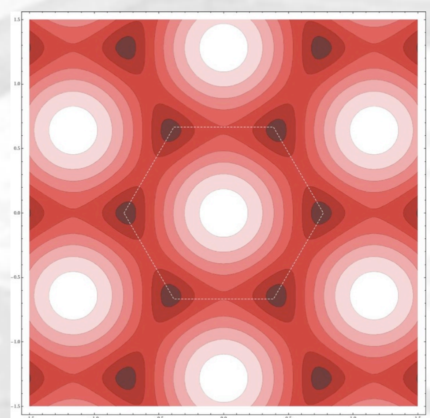


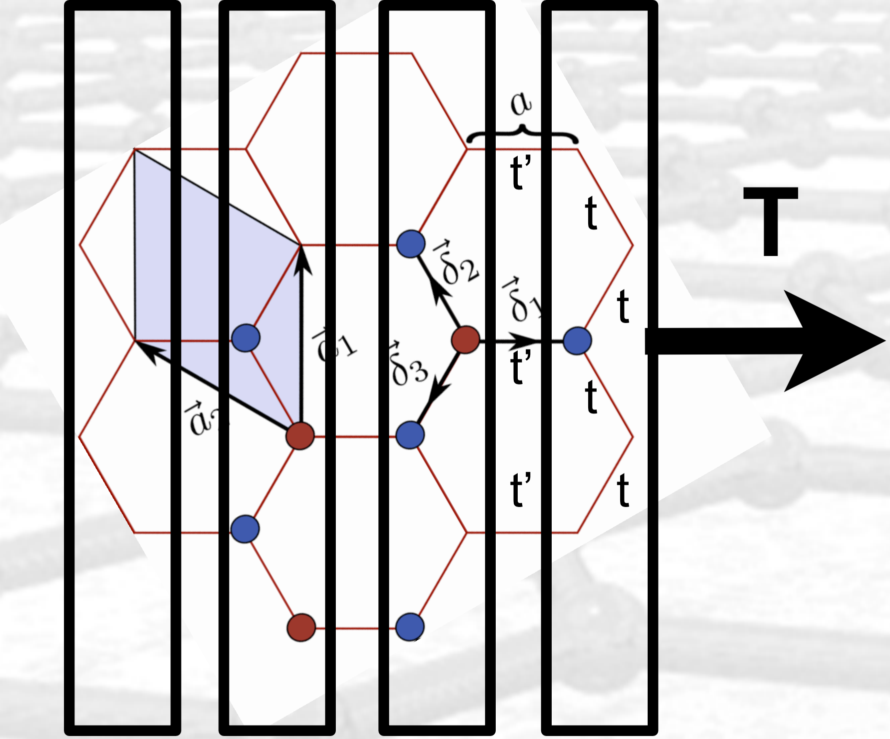


t’<<t

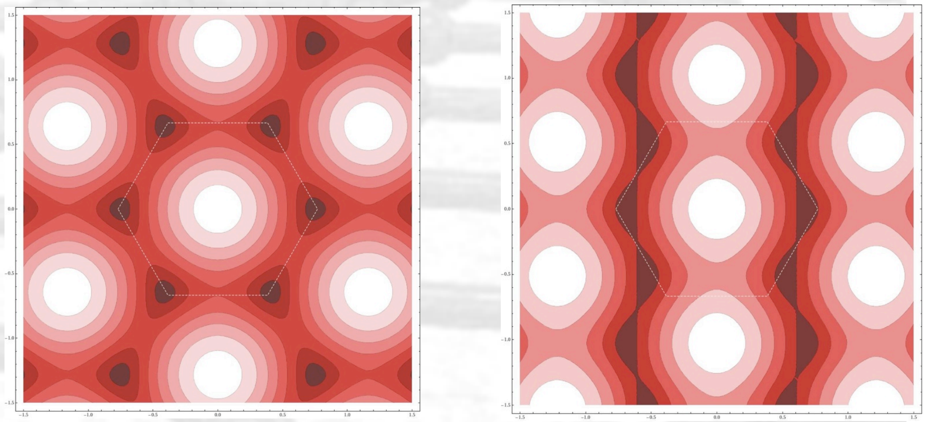
Weakly Coupled Luttinger Liquids





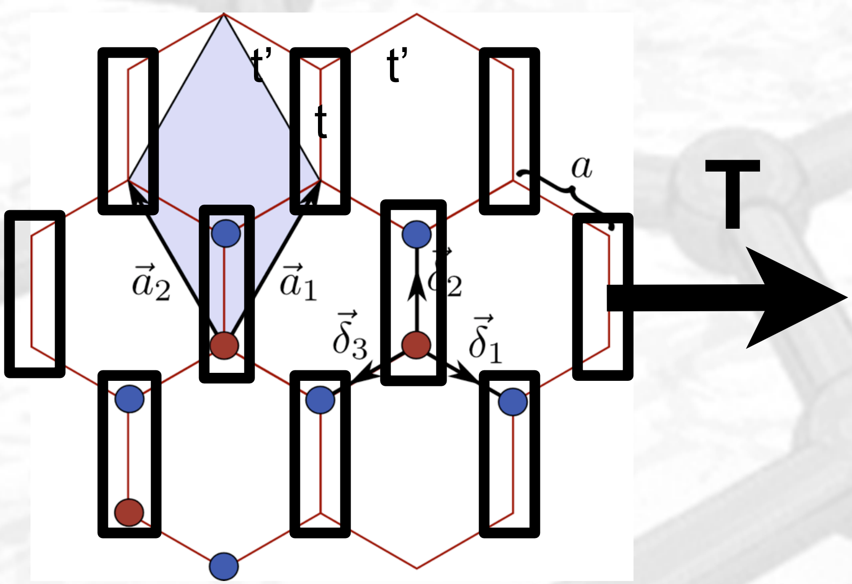


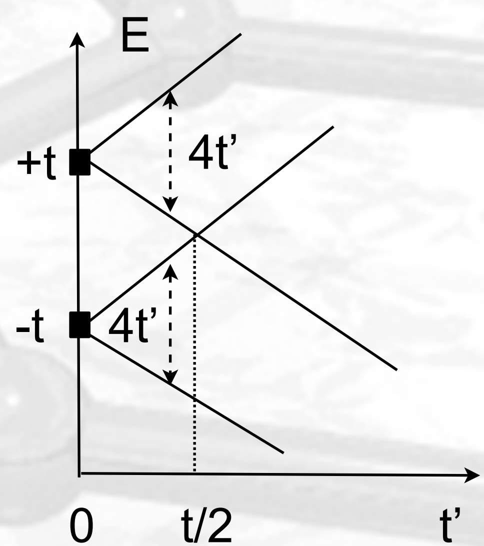
“one-dimensionalization”



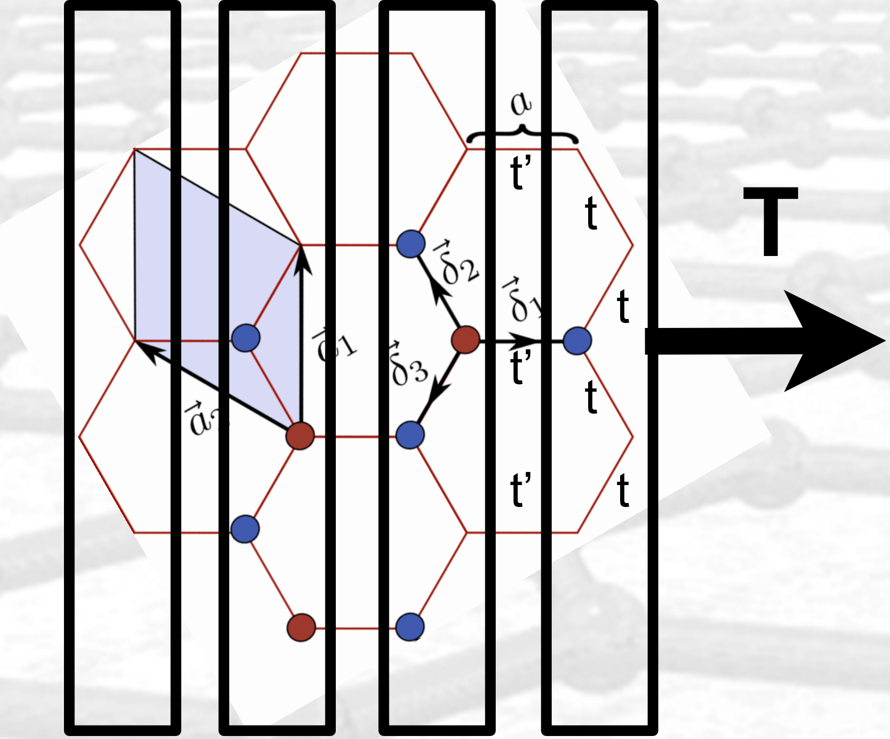
t’<<t

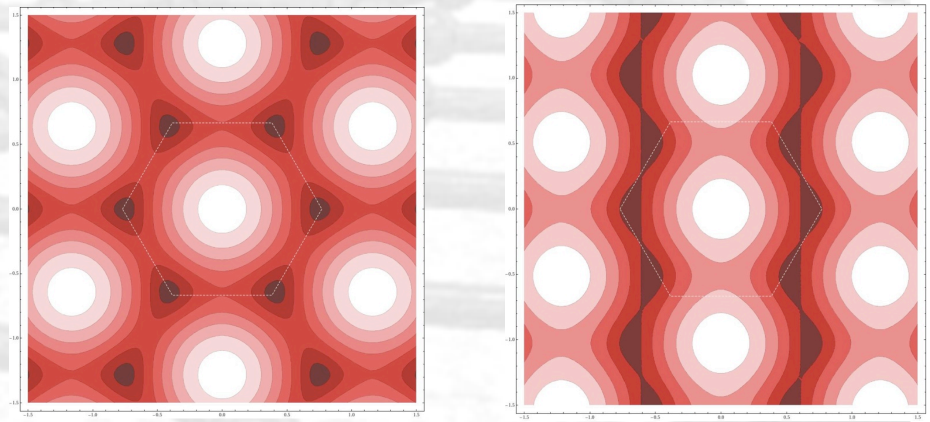
Weakly Coupled Luttinger Liquids





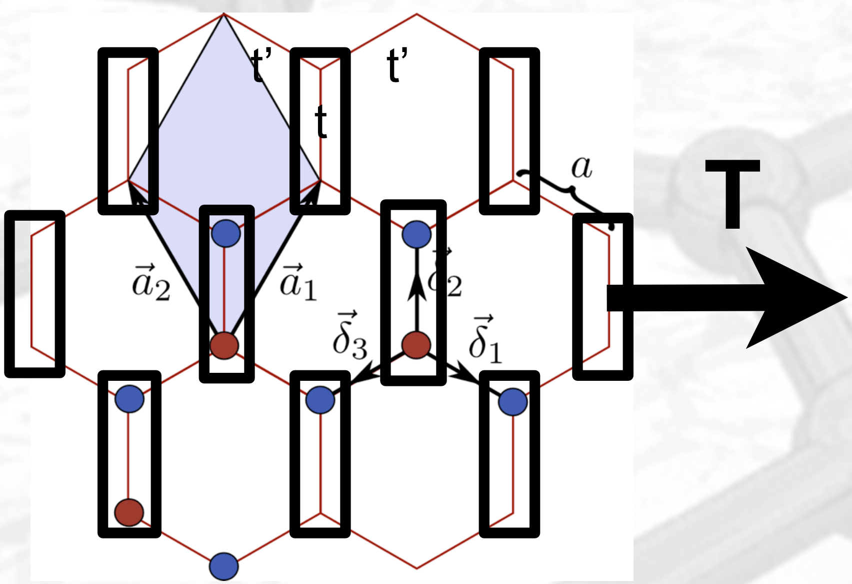
“one-dimensionalization”

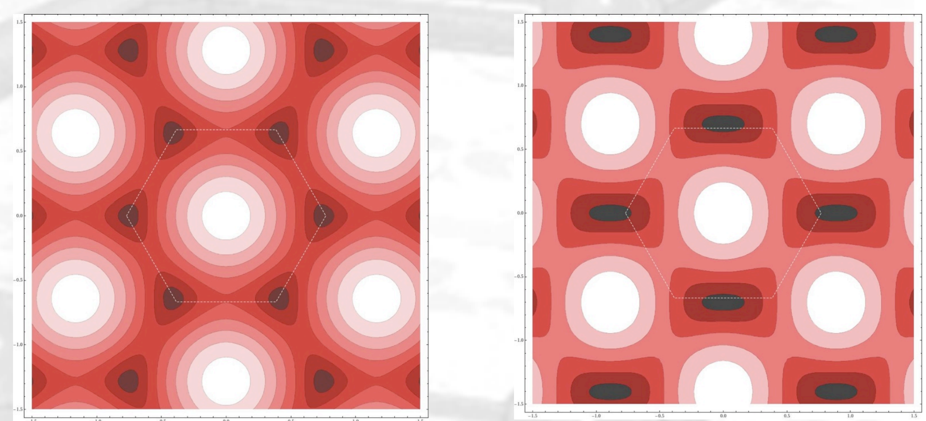




t’<<t

Weakly Coupled Luttinger Liquids



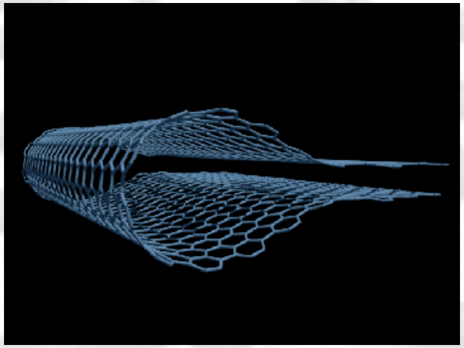


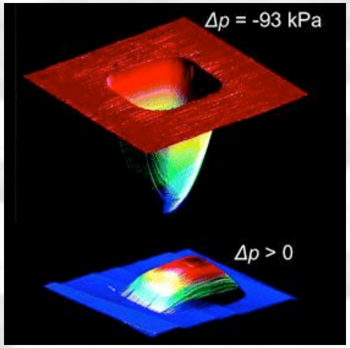
“zero-dimensionalization”

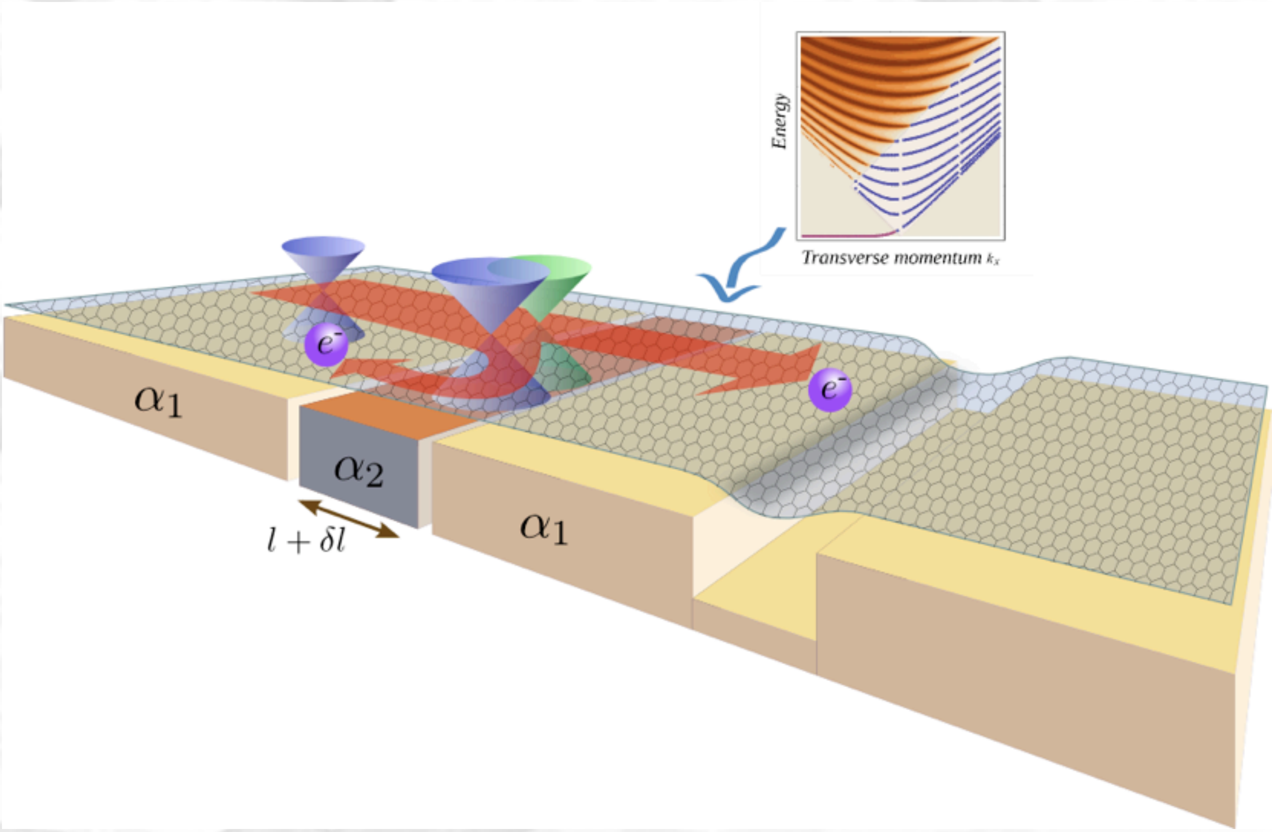
Dimerization

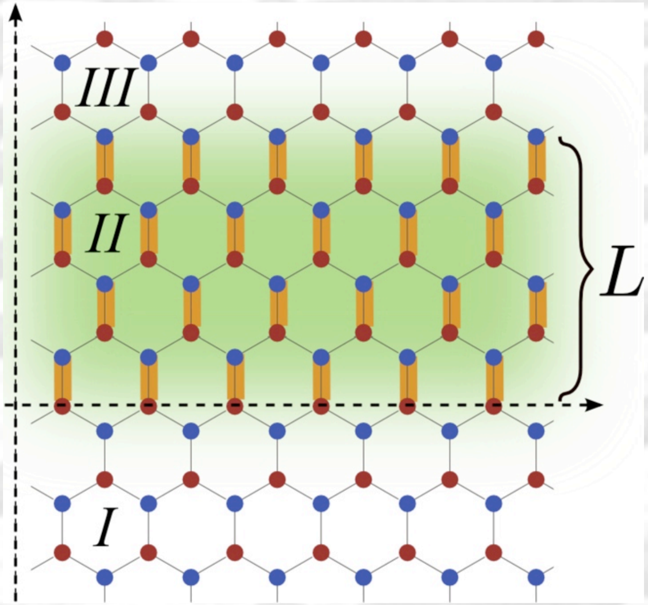
"ORIGAMI" ELECTRONICS

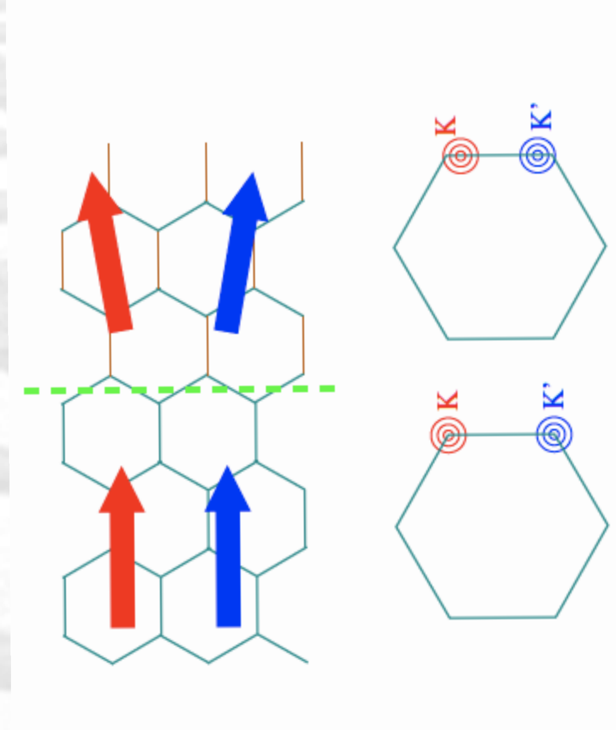
Vitor M. Pereira and A. H. Castro Neto, Phys. Rev. Lett. 103, 046801 (2009)

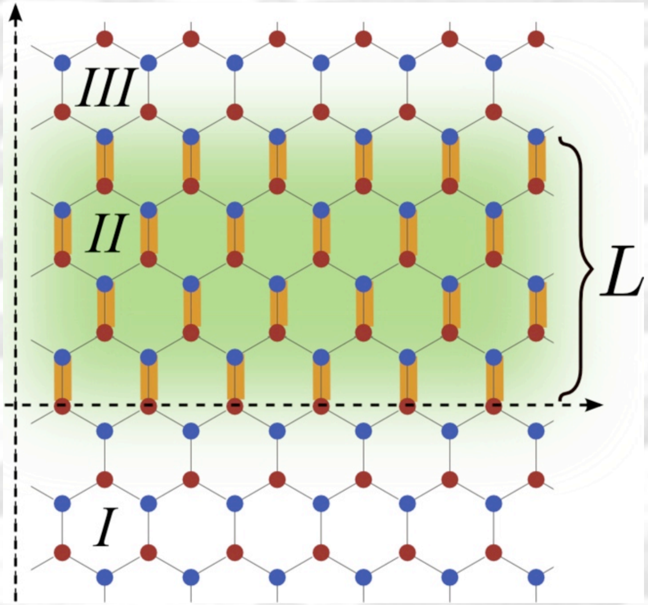


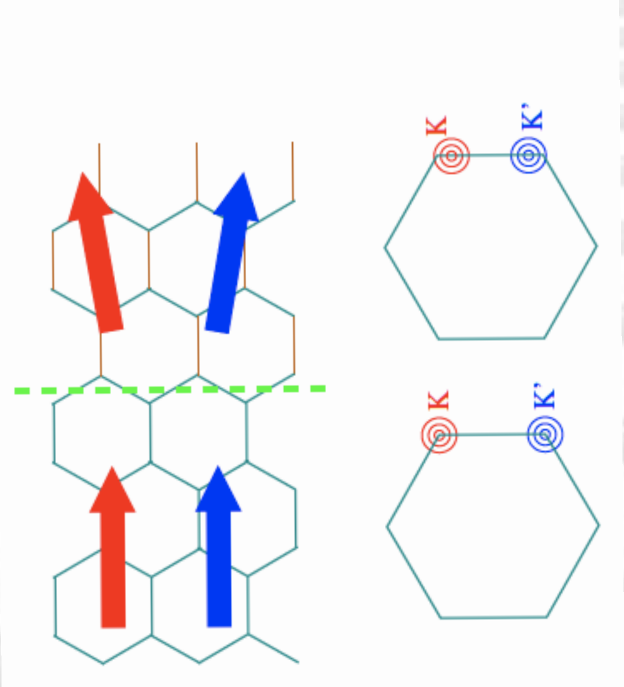





III
II
L
I


K
K'
K
K'

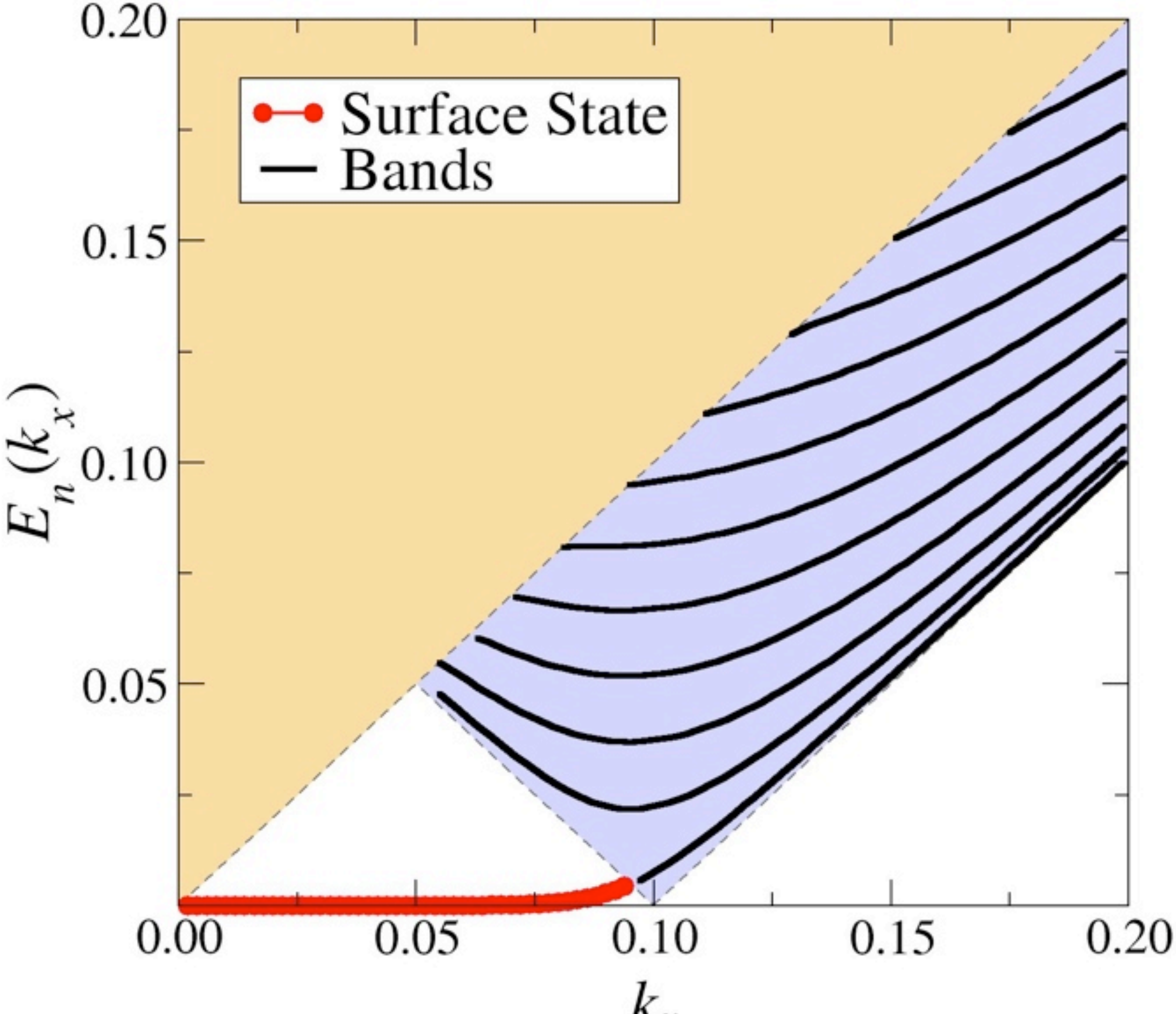


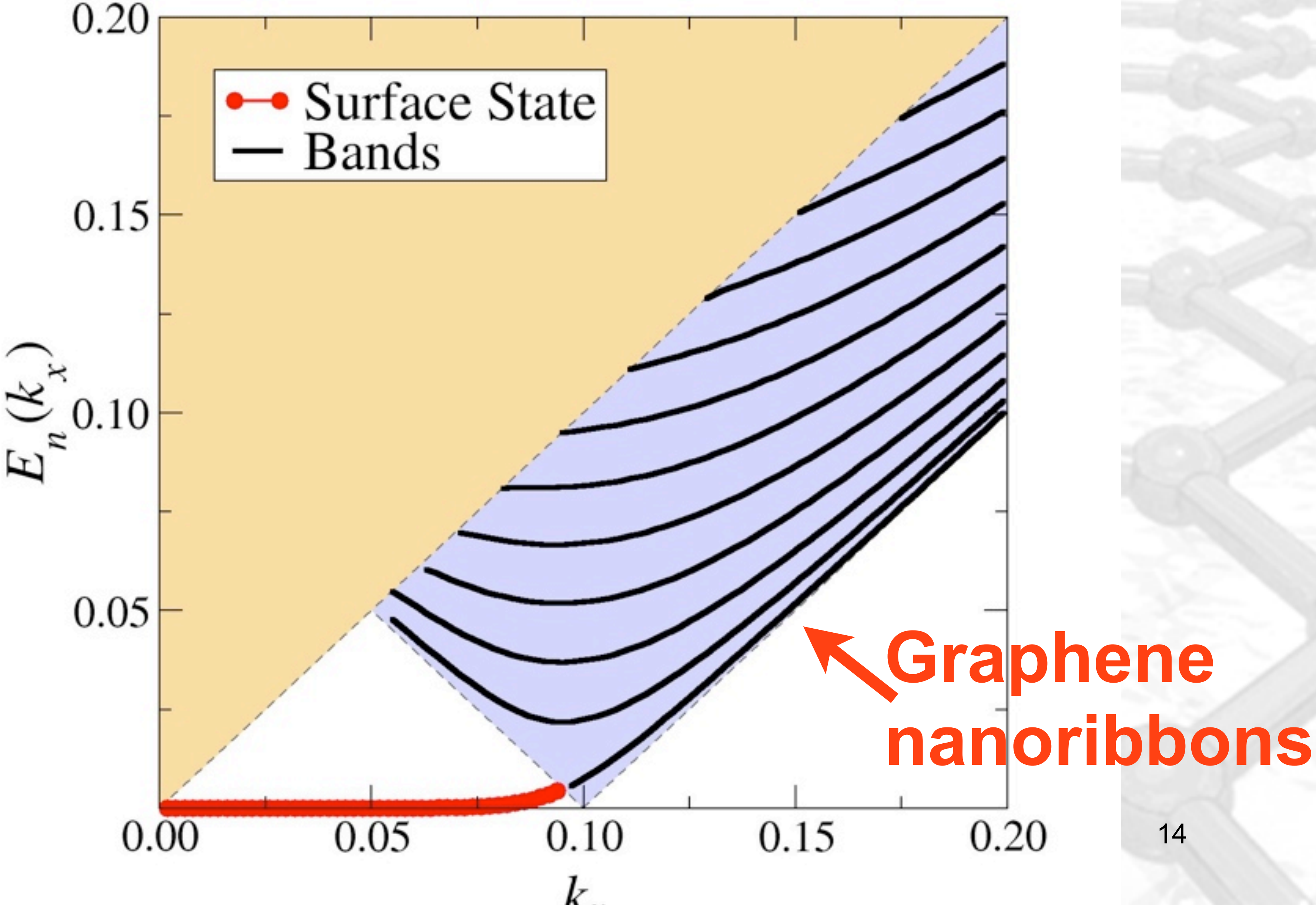


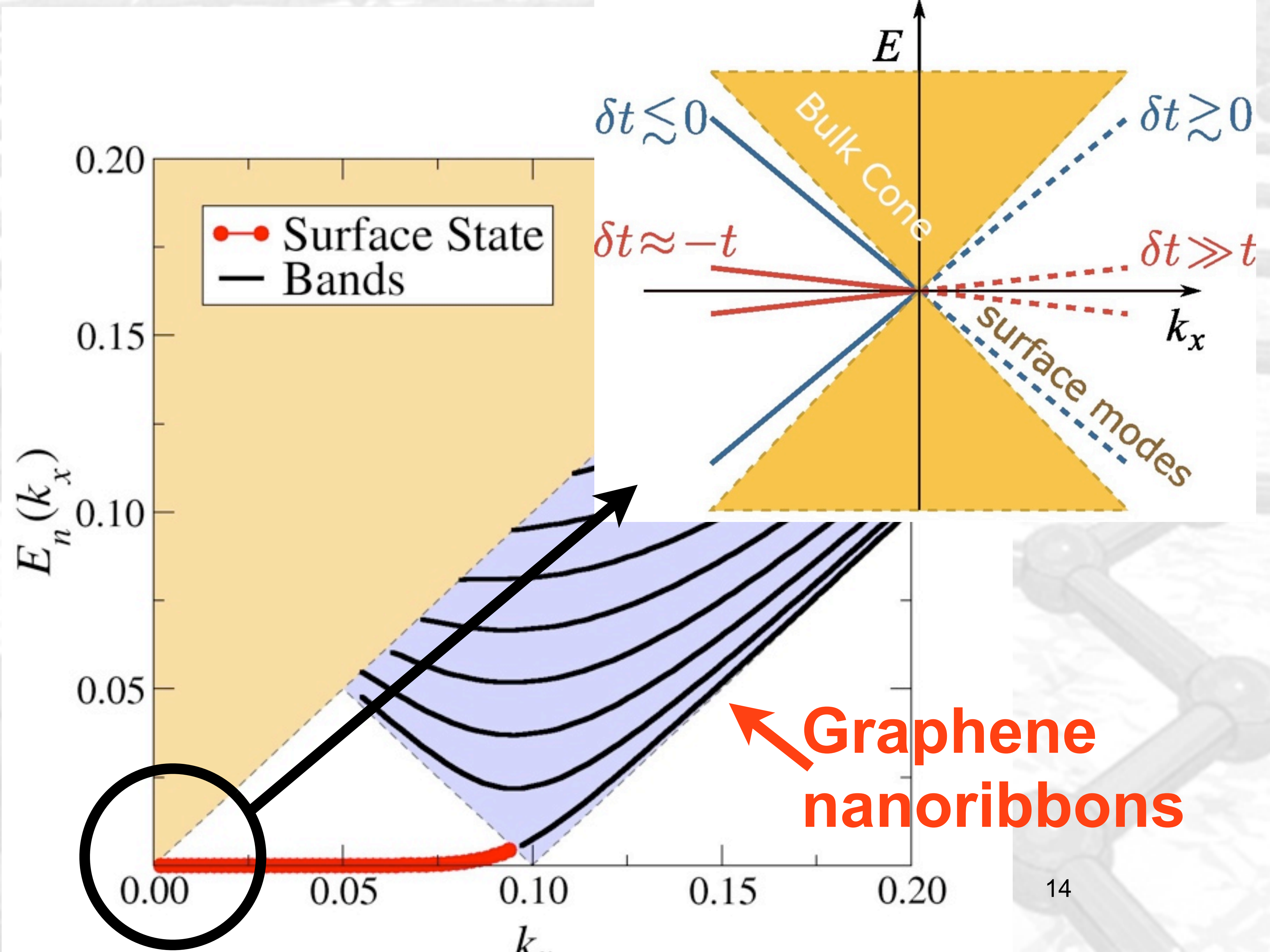
$$H = v_F \int d\boldsymbol{r}\, \Psi^\dagger \begin{bmatrix} \boldsymbol{\sigma}.(\boldsymbol{p} - \frac{1}{v_F}\boldsymbol{\mathcal{A}}) & 0 \\ 0 & -\boldsymbol{\sigma}.(\boldsymbol{p} + \frac{1}{v_F}\boldsymbol{\mathcal{A}}) \end{bmatrix} \Psi\,,$$

$$\mathcal{A}(\boldsymbol{r}) = \mathcal{A}_x(\boldsymbol{r}) - i\mathcal{A}_y(\boldsymbol{r})$$

$$\boldsymbol{\mathcal{A}}(\boldsymbol{r}) = \delta t\, \theta(y)\theta(L-y)\boldsymbol{u}_x\,.$$

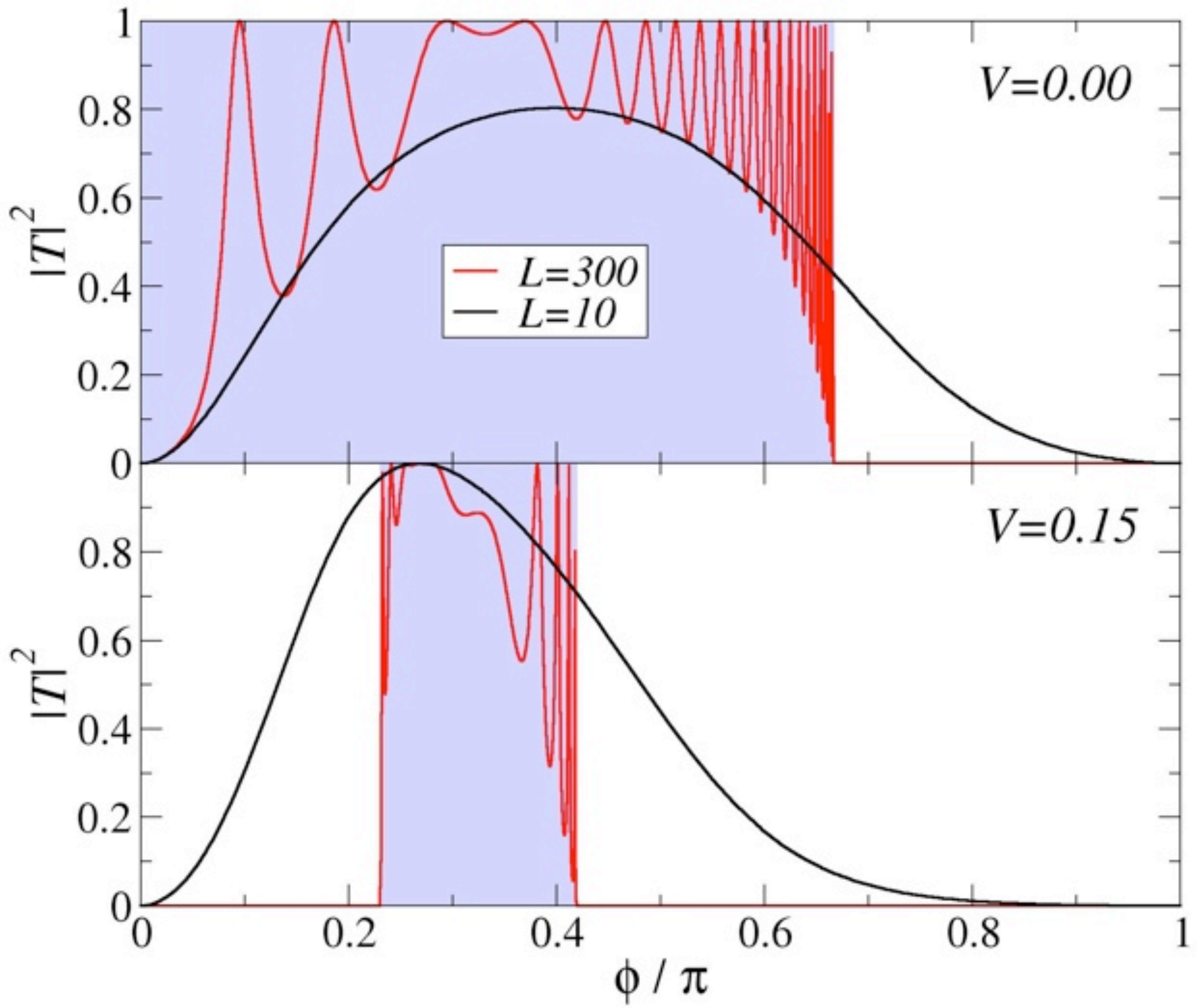

Surface State
Bands
0.20
0.15
0.10
0.05
0.00
0.05
0.10
0.15
0.20
$E_n(k_x)$

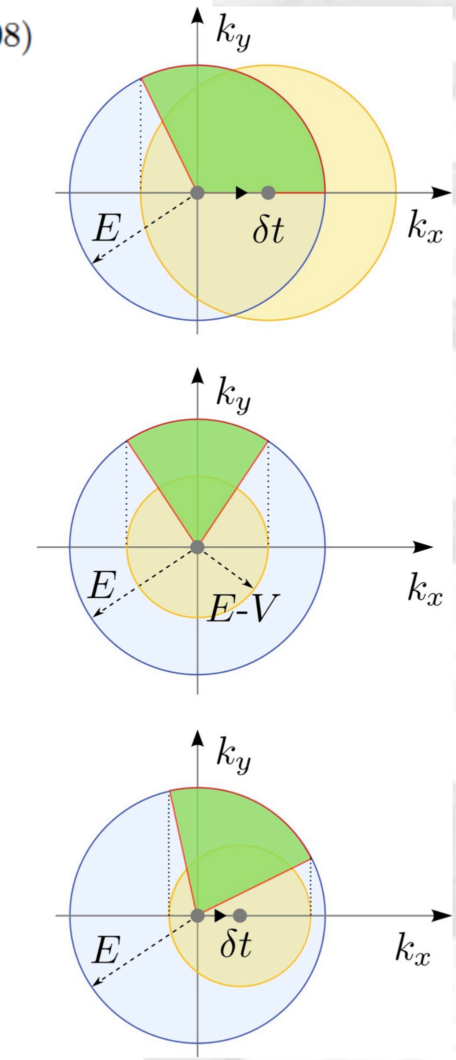

Surface State
Bands
$E_n(k_x)$
0.20
0.15
0.10
0.05
0.00
0.05
0.10
0.15
0.20
Graphene nanoribbons


Surface State
Bands
$E_n(k_x)$
0.20
0.15
0.10
0.05
0.00
0.05
0.10
0.15
0.20
k_x
E
$\delta t \lesssim 0$
$\delta t \gtrsim 0$
Bulk Cone
$\delta t \approx -t$
$\delta t \gg t$
k_x
surface modes
Graphene nanoribbons

$$T = \frac{e^{-ik_y L} \sin\phi \sin\varphi}{\cos(q_y L)\sin\phi\sin\varphi + i\sin(q_y L)(\cos\phi\cos\varphi - 1)}.$$

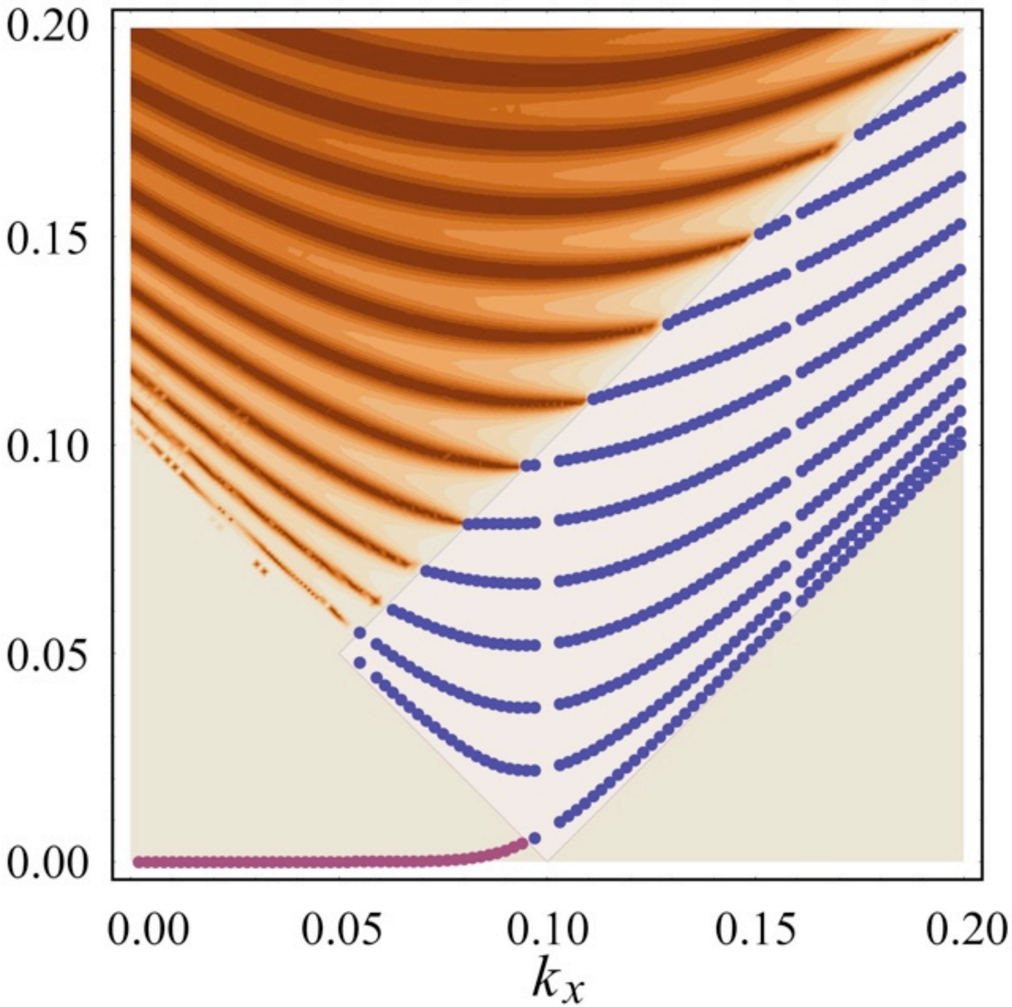
M. M. Fogler, F. Guinea, and M. I. Katsnelson, Phys. Rev. Lett. **101**, 226804 (2008)





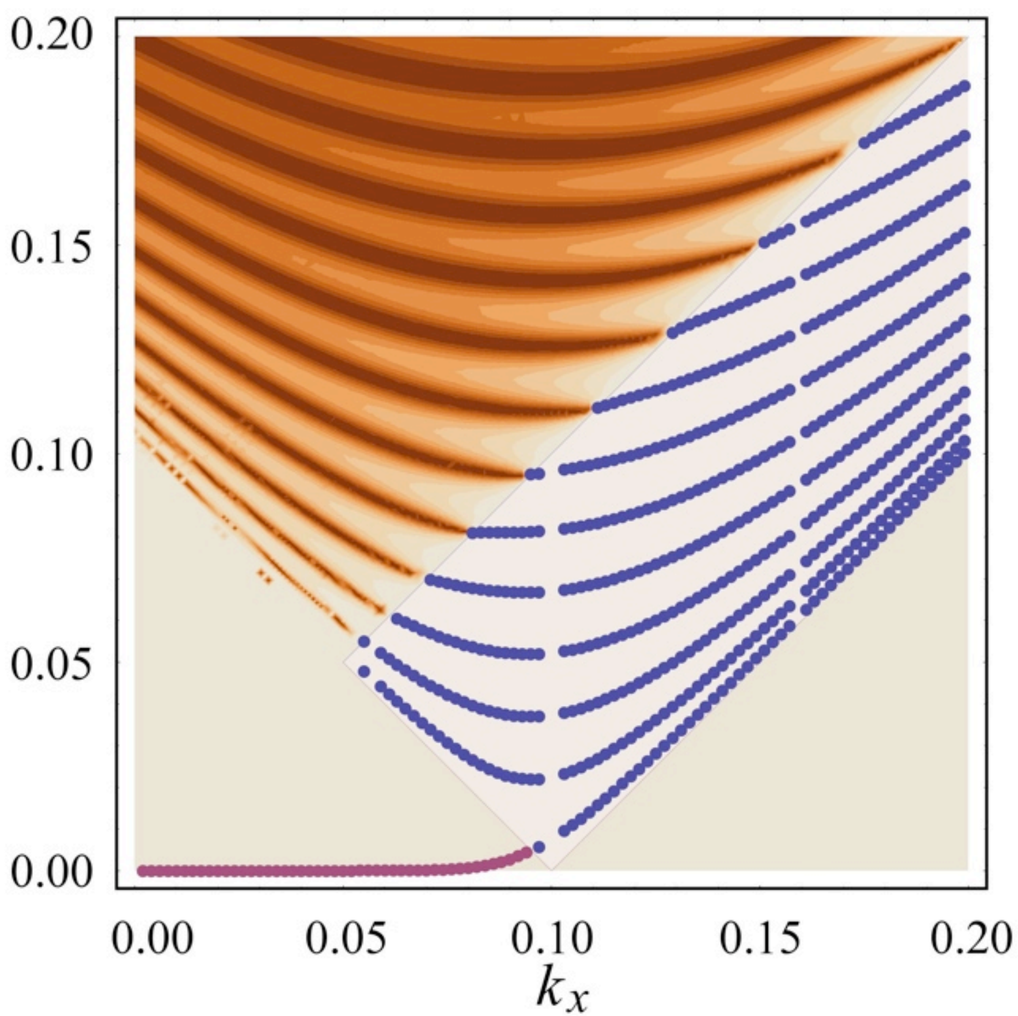
$$T=\frac{e^{-ik_yL}\sin\phi\sin\varphi}{\cos(q_yL)\sin\phi\sin\varphi+i\sin(q_yL)(\cos\phi\cos\varphi-1)}.$$

M. M. Fogler, F. Guinea, and M. I. Katsnelson, Phys. Rev. Lett. **101**, 226804 (2008).



$$T = \frac{e^{-ik_y L} \sin\phi \sin\varphi}{\cos(q_y L)\sin\phi\sin\varphi + i\sin(q_y L)(\cos\phi\cos\varphi - 1)}.$$

M. M. Fogler, F. Guinea, and M. I. Katsnelson, Phys. Rev. Lett. **101**, 226804 (2008).

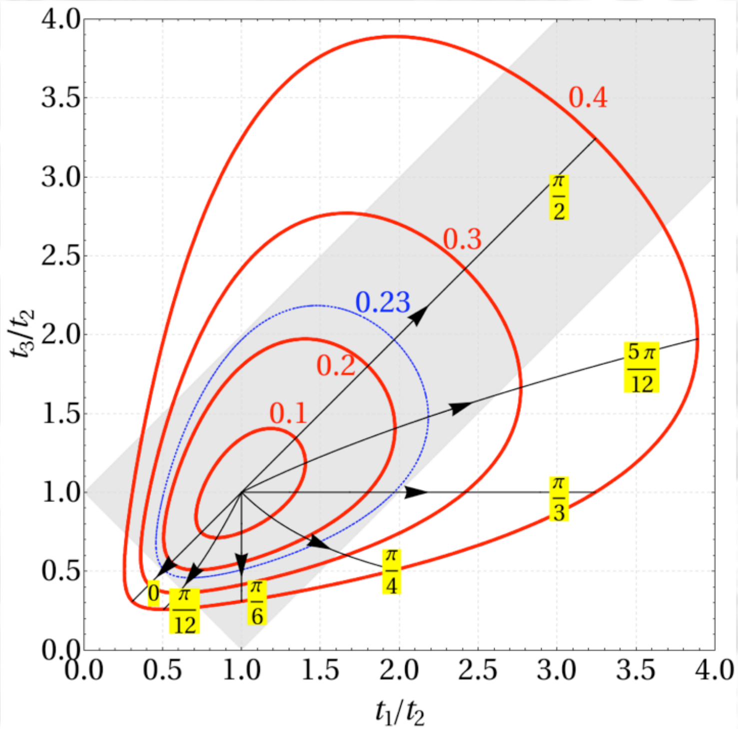


$L \approx 20\text{nm}$

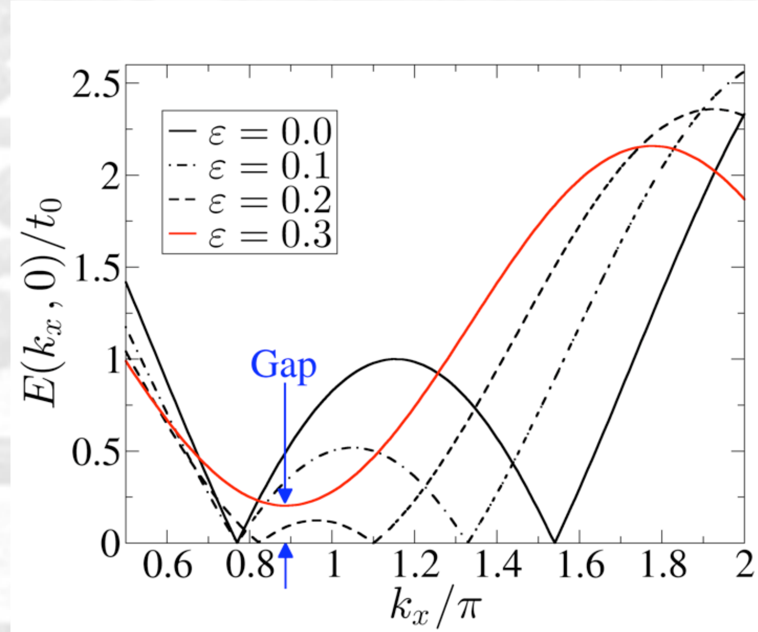
$E_g \approx 50$ meV

$\delta t/t \sim 10\%$

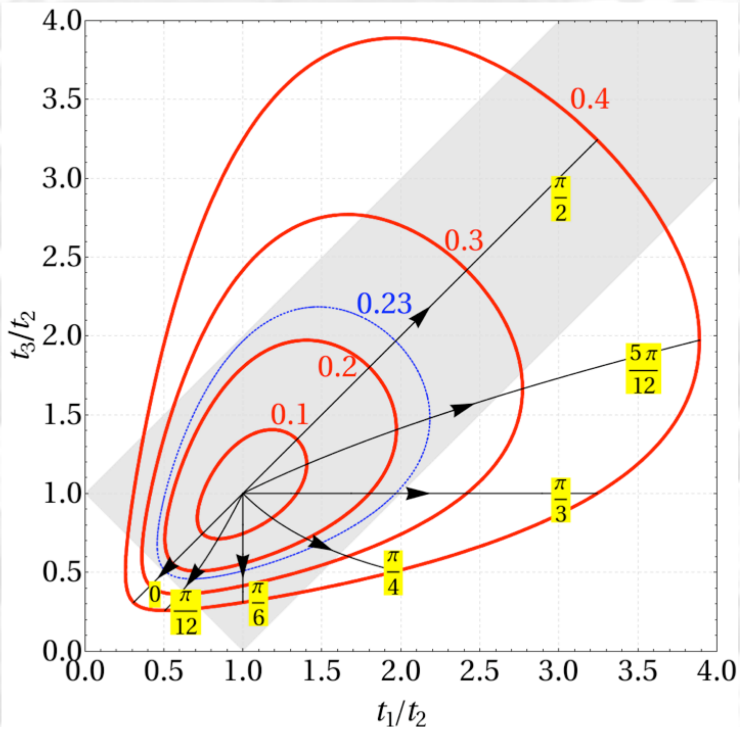
strain of $\sim 5\%$



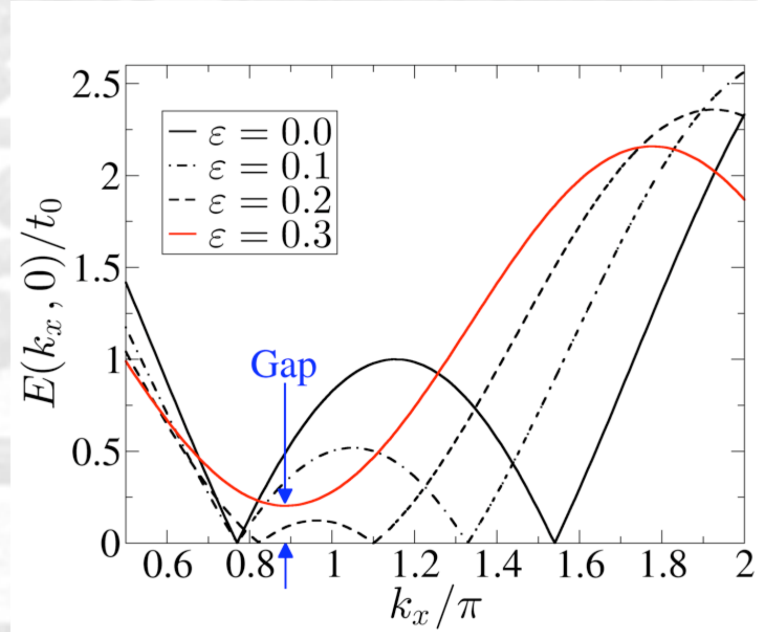
t_3/t_2
t_1/t_2
0.1
0.2
0.23
0.3
0.4
0
$\frac{\pi}{12}$
$\frac{\pi}{6}$
$\frac{\pi}{4}$
$\frac{\pi}{3}$
$\frac{5\pi}{12}$
$\frac{\pi}{2}$



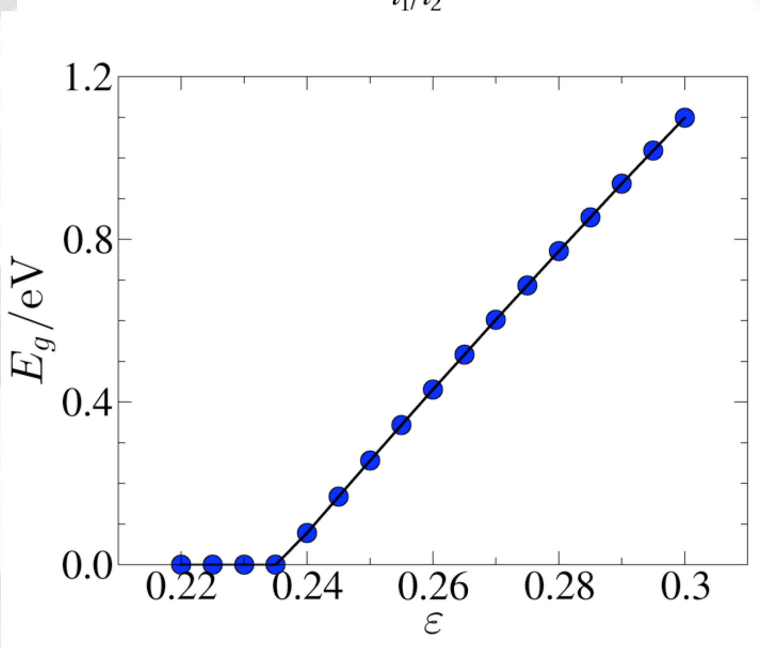
$\varepsilon = 0.0$
$\varepsilon = 0.1$
$\varepsilon = 0.2$
$\varepsilon = 0.3$
Gap
$E(k_x, 0)/t_0$
k_x/π



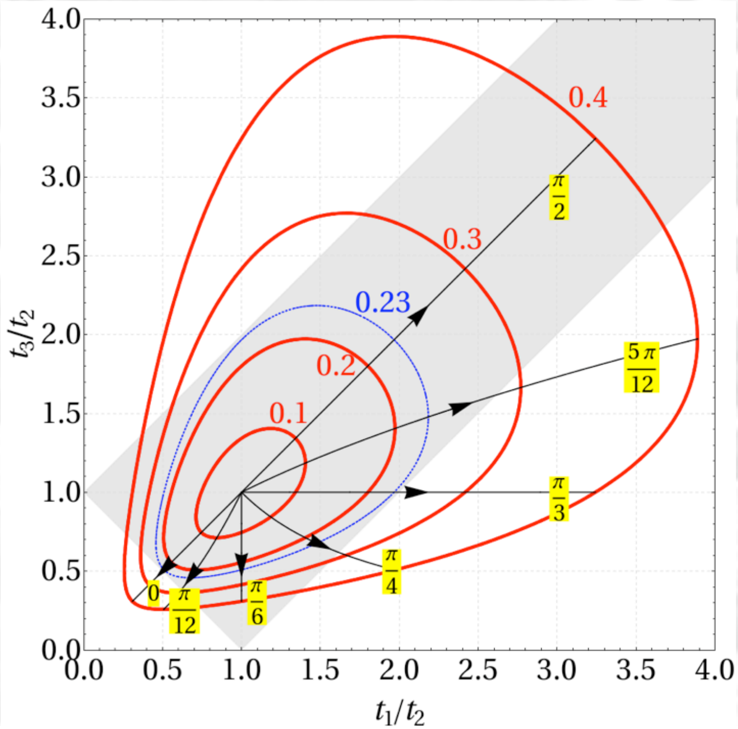
t_3/t_2
t_1/t_2
0.1
0.2
0.23
0.3
0.4
0
$\frac{\pi}{12}$
$\frac{\pi}{6}$
$\frac{\pi}{4}$
$\frac{\pi}{3}$
$\frac{5\pi}{12}$
$\frac{\pi}{2}$

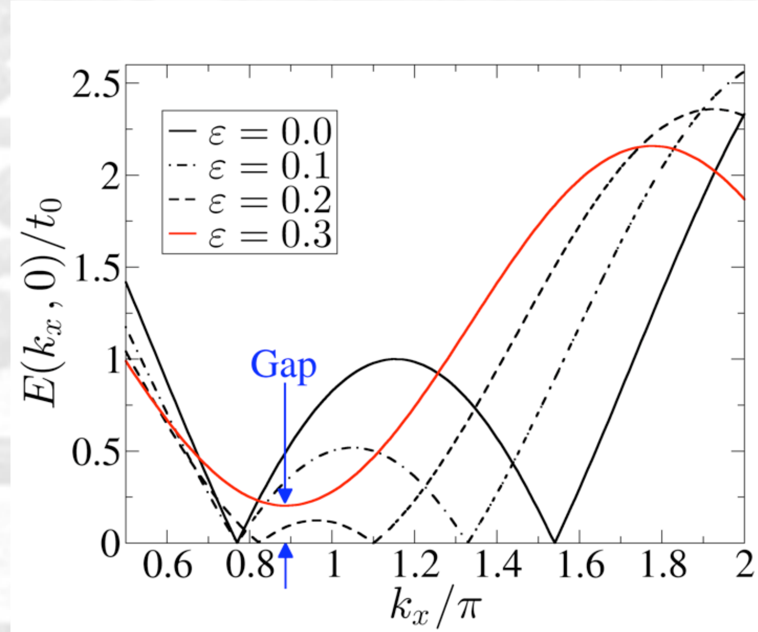


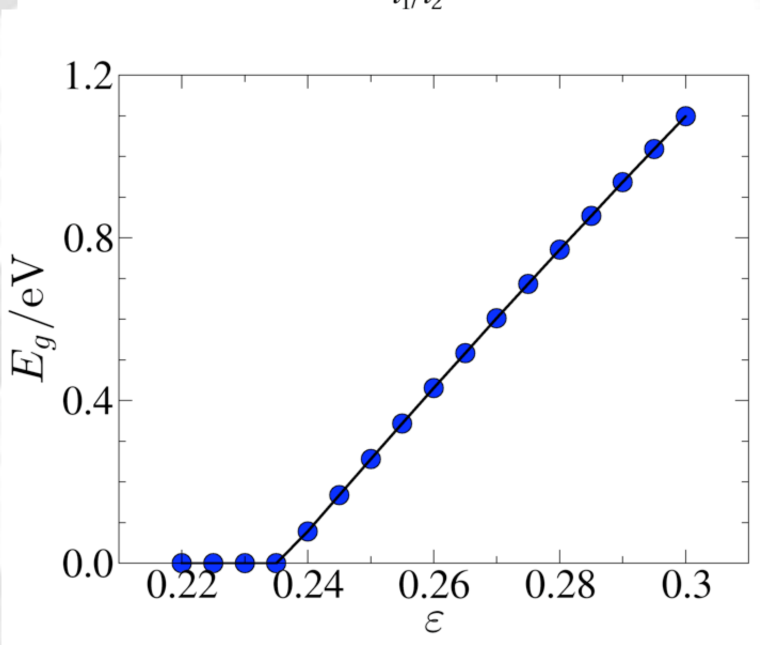
$E(k_x, 0)/t_0$
k_x/π
$\varepsilon = 0.0$
$\varepsilon = 0.1$
$\varepsilon = 0.2$
$\varepsilon = 0.3$
Gap

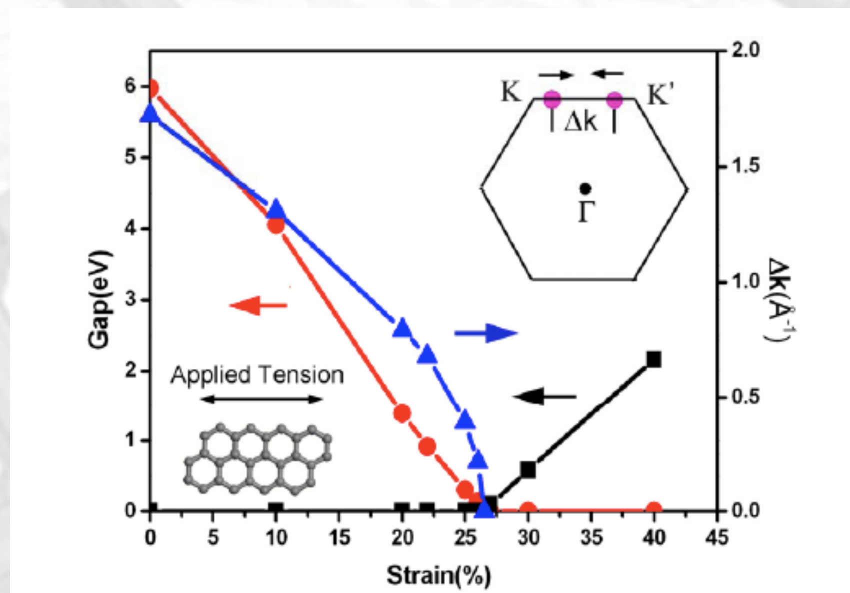


E_g/eV
ε









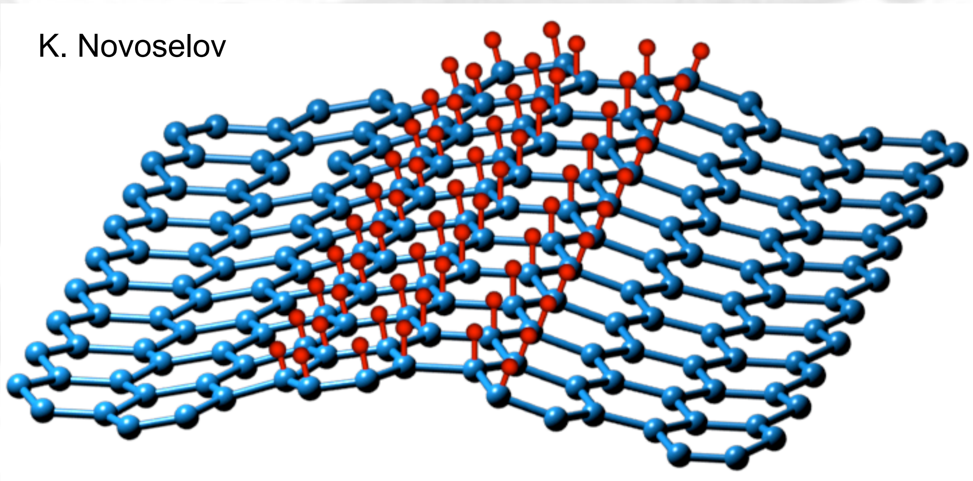
Z.H.Ni et al. Erratum: ACS Nano 2, 2301 (08)

Chemical reactivity change with folding

Origami chemistry

Chemical reactivity change with folding

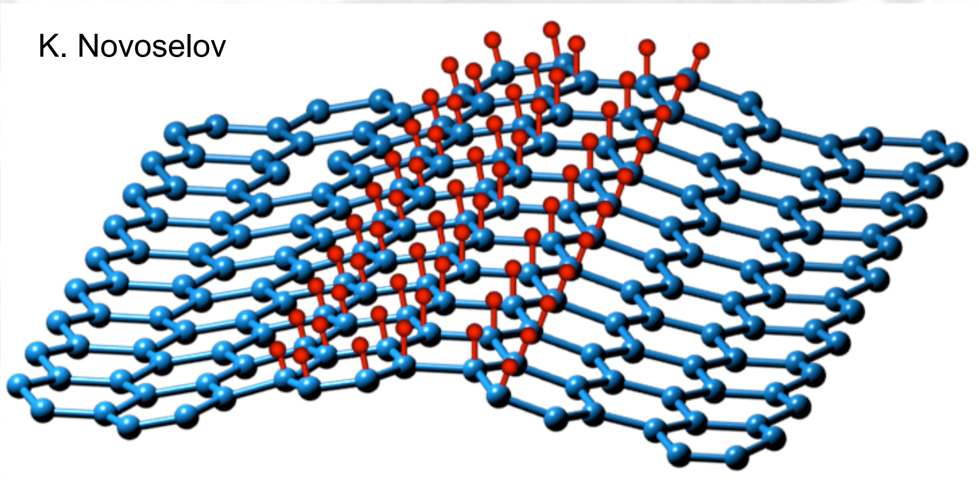
Origami chemistry



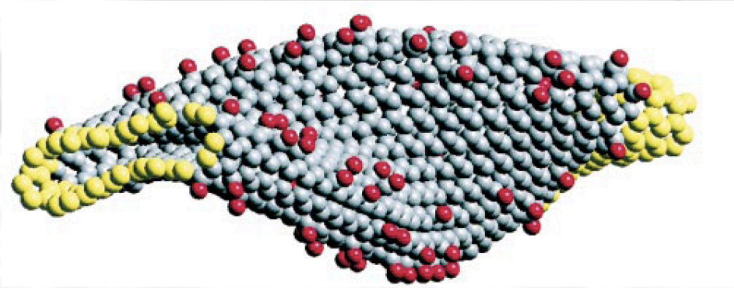
Graphane

Chemical reactivity change with folding

Origami chemistry



Graphane



Srivastava et al., J. Phys. Chem. B 103, 4330 (1999)

Graphene Mechanics: correct analogy not so much like this...

1 micrometer square of graphene = sheet of a plastic wrap as big as a living room

Graphene Mechanics: ...but more like so!

Buckling, Shear, Crumpling and Wrinkles

Due to its 2-dimensionality graphene

1. Cannot withstand compression
2. Cannot withstand shear

No Threshold for Buckling Instability

Inext. Memb. | Textiles | Paper | Metal

Universal aspects of Geometry and Physics of Wrinkling

1. Almost all area is locally flat;
2. Network of ridges linked/terminated by vertices;
3. Elastic energy strongly focused in vertices.

Witten, Rev. Mod. Phys. (2007), Vaziri & Mahadevan, PNAS (2008).

Vertices have universal shape ⇒ **Conical Singularities**

Are There Wrinkling Singularities in Graphene? Plenty!

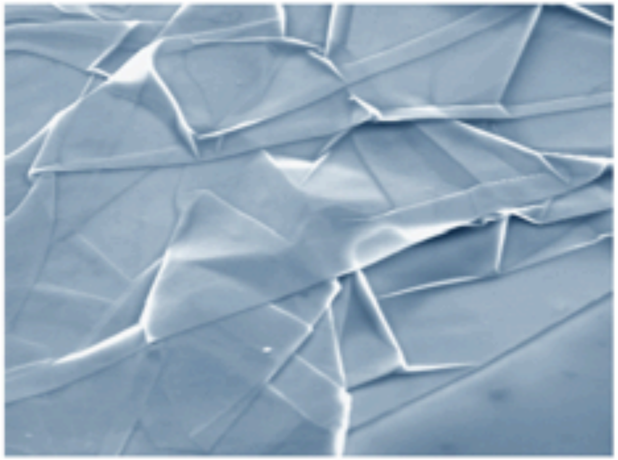


Fig. 5: Graphene on SiO_2 (A. Geim).

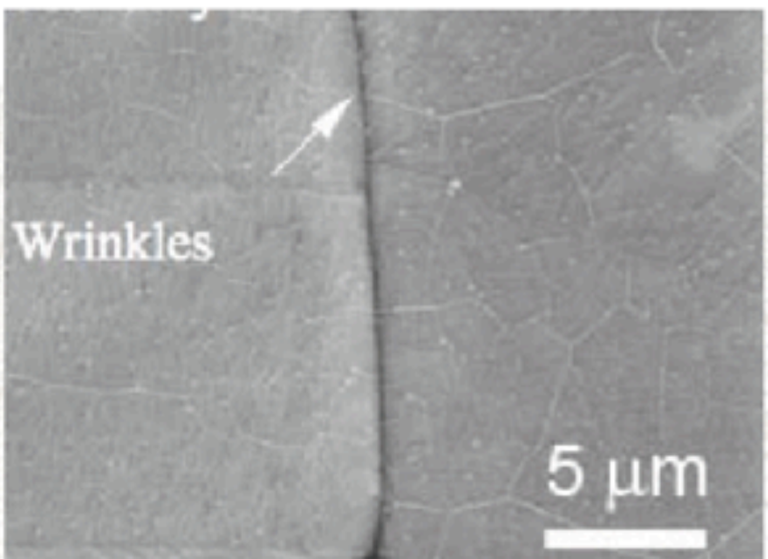


Fig. 6: CVD of graphene on Cu (R. Ruoff).

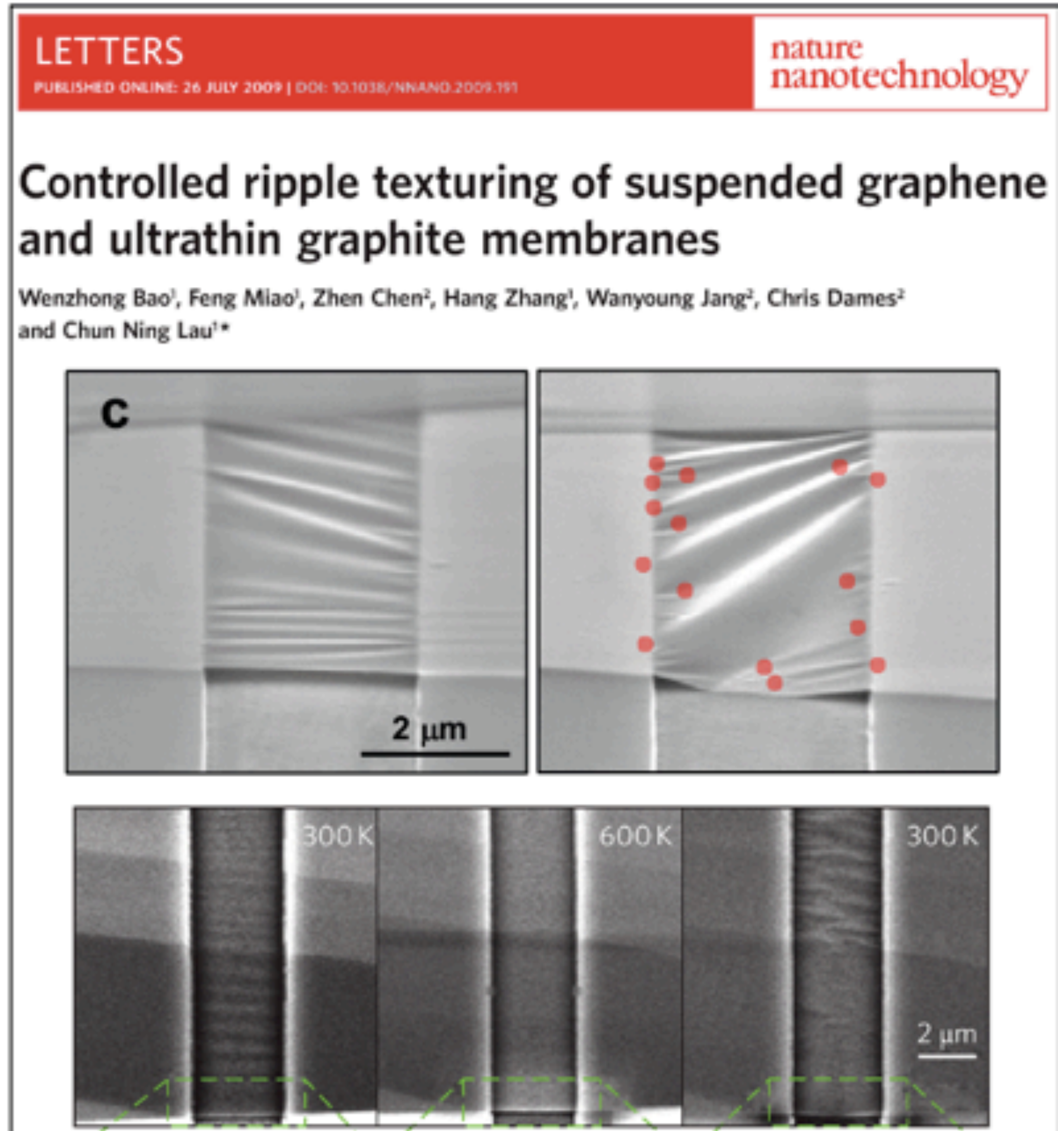


Fig. 7: Typical wrinkles in suspended graphene.

Simulation of Graphene Wrinkling and Singularities

- Unprecedented possibilities for mechanical simulation of ultra-thin sheets and plates.
- Accurate and efficient bond-order potentials available.

Atomistic simulation of graphene

- Biaxial Shear
- $T = 0$
- REBO + MD relaxation

Buckles just as a thin plate would.

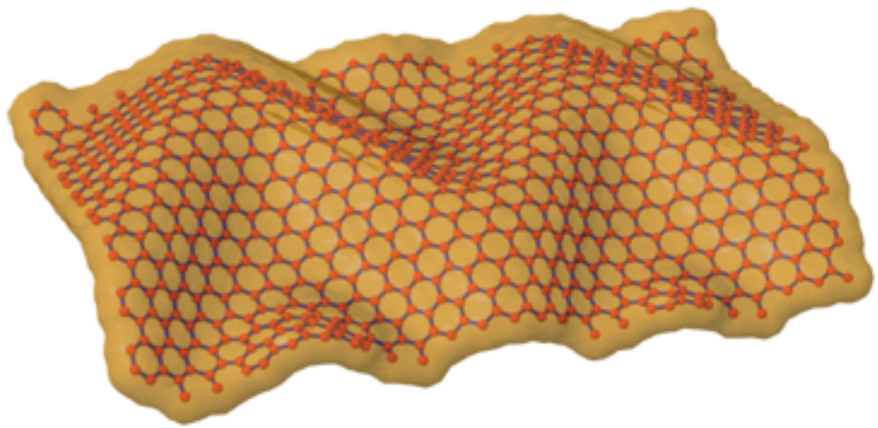


Fig. 8: Relaxed structure under biaxial shear.

Isolated Conical Singularities

- Same curvature profile;
- No strain except at the vertex.

Target for electron scattering

Fig. 9: Relaxed structure under biaxial shear.

Theory of Conical Singularities

Cerda & Mahadevan, PRL (1997)

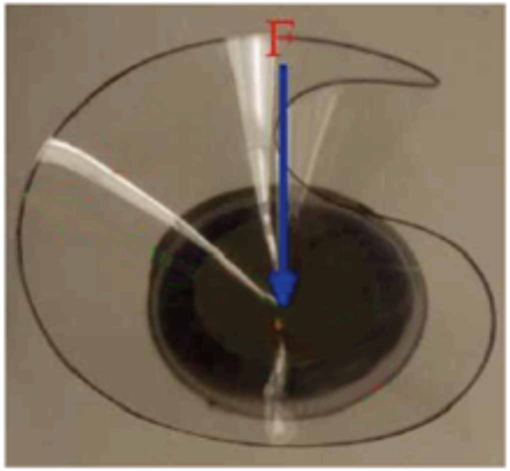


Fig. 10: Mellado *et al.*, 0912.3778

1. Indentation of a free, flat membrane;
2. Apply NL elasticity to obtain deformation field:

$$\boldsymbol{u}(\rho,\theta) = u_\rho(\rho,\theta)\boldsymbol{u}_\rho + u_\theta(\rho,\theta)\boldsymbol{u}_\theta + \zeta(\rho,\theta)\boldsymbol{z}$$

3. Vertical displacement is a generalized cone:

$$\zeta(\rho,\theta) = \rho\,\psi(\theta)$$

4. Extract exact solution for $\psi(\theta)$, $\boldsymbol{u}(\rho,\theta)$.

$\boldsymbol{u}(\rho,\theta)$ universal → independent of material parameters!

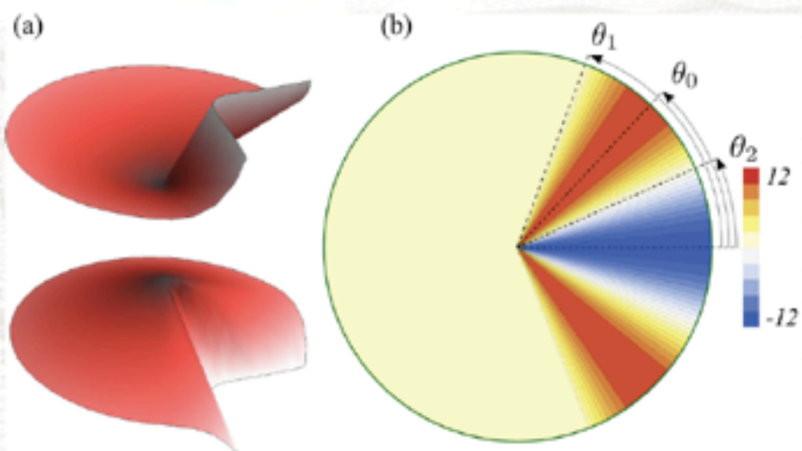
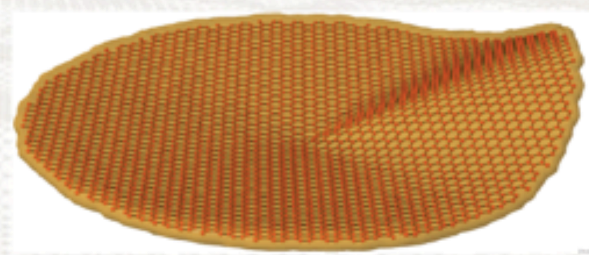


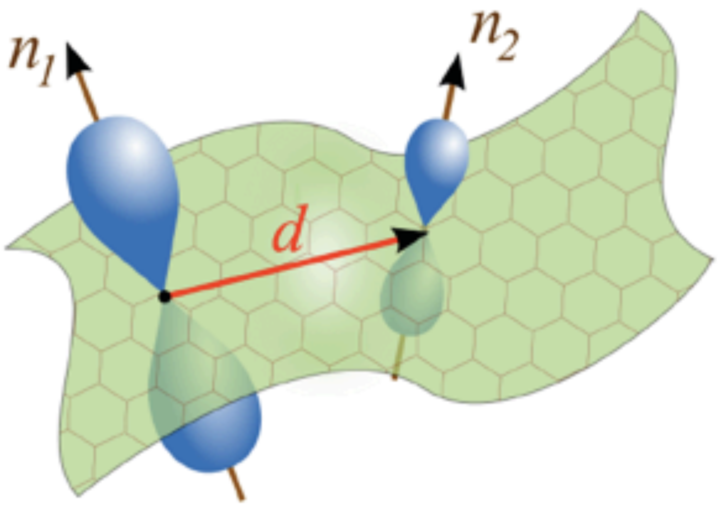
Fig. 11: Analitical Profile of $\boldsymbol{u}(\rho,\theta)$

Cauchy-Born Hypotesis



Carbon lattice follows $\boldsymbol{u}(\rho,\theta)$

Dirac Electrons on a Curved Surface



Arbitrary p_z orbitals in 3D space

$$t_{12} = V_{pp\pi}\, \boldsymbol{n}_1 \cdot \boldsymbol{n}_2 + (V_{pp\sigma} - V_{pp\pi})(\boldsymbol{n}_1 \cdot \hat{d})(\boldsymbol{n}_2 \cdot \hat{d})$$

See also Isacsson PRB (2008)

Remember: we know the exact geometry of the conical surface from the elasticity solution.

Differential geometry on the Conical Surface to obtain $\boldsymbol{n}_i$, $\boldsymbol{d}_i$:

$$t_{ij} = t + \delta t_{ij} \rightarrow \delta t_{ij} \approx -V^0_{pp\pi} \frac{1}{2} \left|(\delta_0 \cdot \nabla)\nabla\zeta\right|^2 + \frac{V^0_{pp\pi}/3 - V^0_{pp\sigma}/4}{d_0^2}\left[(\delta_0 \cdot \nabla)^2 \zeta\right]^2$$

But, at low energies, graphene electrons behave as Dirac fermions:

$$H \approx v_F \boldsymbol{\sigma}.\left[\boldsymbol{p} - \tfrac{1}{v_F}\mathcal{A}(\boldsymbol{r})\right] + \left[3t'_0 + \Phi(\boldsymbol{r})\right]\sigma^0 \qquad \left(\mathcal{A}(\boldsymbol{r}), \Phi(\boldsymbol{r}) \propto \delta t_{ij}\right)$$

$$\Phi(\boldsymbol{r}) = \alpha\, \mathrm{Tr}^2[K^i_j] - \beta \det[K^i_j] \qquad \left(K^i_j = \text{Curvature Tensor, Gauss II}\right)$$

$\alpha = 9a_0^2 V^0_{pp\pi}/8 + 27a_0^2 V_{pp\sigma}/32 \approx 1.5\text{eV}\text{Å}^2$, $\beta = 3a_0^2 V^0_{pp\pi} + 9a_0^2 V^0_{pp\sigma}/8 \approx 3\text{eV}\text{Å}^2$

Semiclassical Transport

Scattering rate

$$S(\mathbf{k},\mathbf{k}') = \frac{2\pi}{\hbar}|V_{\mathbf{k},\mathbf{k}'}|^2\delta(E_k - E_{k'}), \qquad V_{\mathbf{k},\mathbf{k}'} = \Phi_{\mathbf{k}-\mathbf{k}'}\,\frac{1+\exp(i\phi_k - i\phi_{k'})}{2}$$

Effective Potential

$$\Phi(\mathbf{r}) = \frac{\alpha}{r^2}\times\Big[\psi(\theta)+\psi''(\theta)\Big] \qquad \text{(supercritical!)} \qquad \Phi_{\mathbf{q}} \sim \log(qr_0) + \ldots$$

Conductivity

$$\sigma = \sigma_0\, G^{3,1}_{2,4}\left(4k_F^2 r_0^2 \,\middle|\, \begin{matrix} -\frac{1}{2}, \frac{1}{2} \\ 0, 0, 0, -2 \end{matrix}\right)^{-1}, \qquad \sigma_0 = \frac{v_F^2\hbar e^2}{2\pi^3 n_i v_0^2}$$

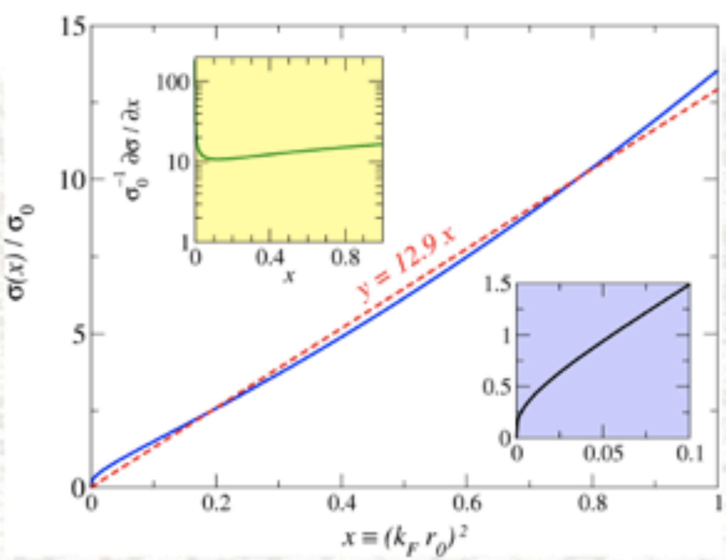


Fig. 12: DC conductivity vs density.

DC Conductivity

- Quasi-linear as function of n_e;
- Log-singularity at the origin;
- Same physics as mid-gap states.

Sanity check

Go back to the full tight-binding, and analyze local electronic structure.

$$H = \sum_{\langle i,j \rangle} t_{ij} c_i^\dagger c_j + \sum_{\langle\langle i,j \rangle\rangle} t'_{ij} c_i^\dagger c_j + \text{H. c.}$$

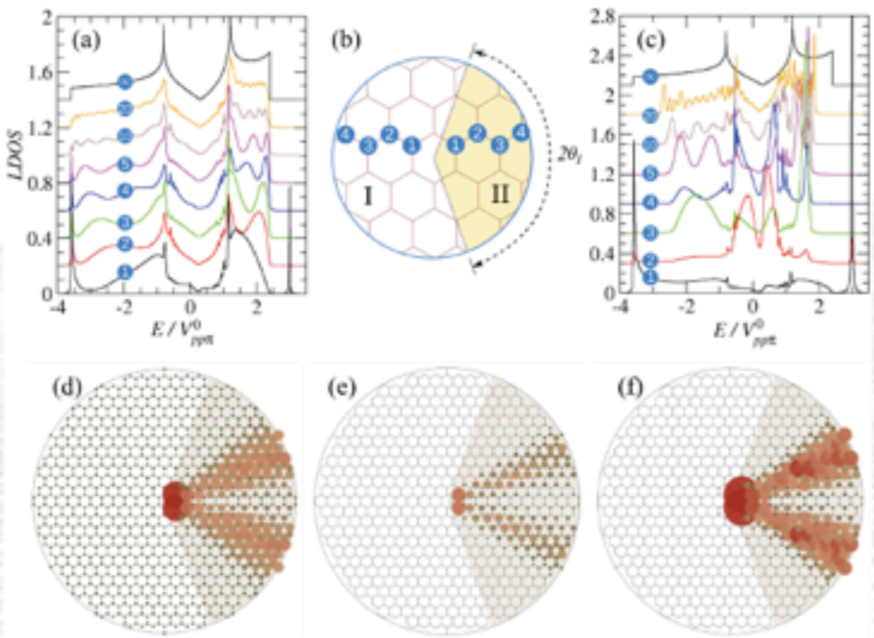


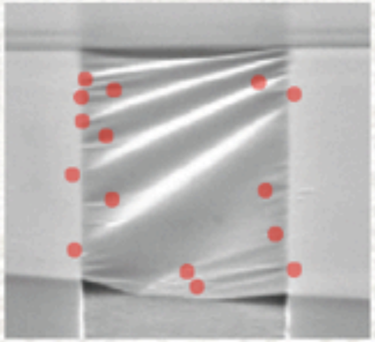
Fig. 13: LDOS nearby a CS.

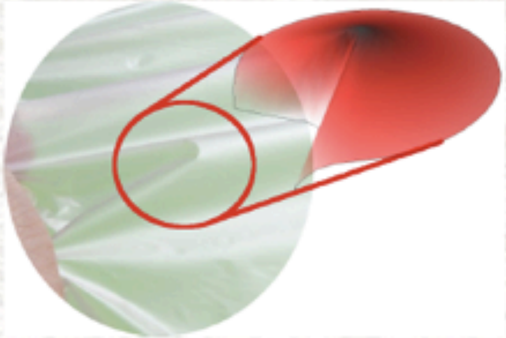
Local Electronic Structure

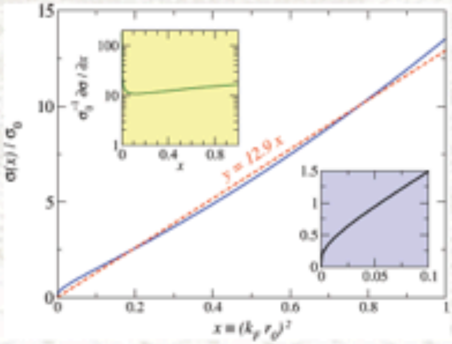
- Apex can be strongly confining;
- Charge density concentrated around apex;
- Extra leakage around rays of null curvature;
- Pseudo magnetic oscillations in LDOS vs E.

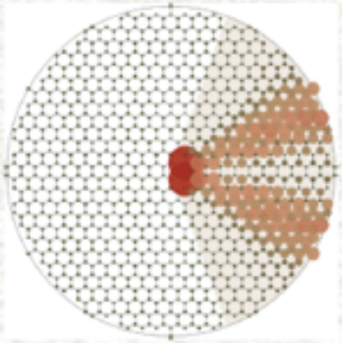
Conclusions

1. Graphene wrinkles. Often and easily.
2. Universal aspects of wrinkling in graphene $\Longrightarrow$ Conical singularities.
3. Graphene's electrons couple to structural deformations and curvature.
4. Conical singularities act as strong scatterers yielding
   - Linear-in-density σ;
   - Confining centers;
   - Variable pseudo magnetic fields.
5. Even in the absence of any impurities, mobility limited by wrinkling.
6. Pseudomagnetic fields interfere with Landau quantization.
7. Further possibilities in the realm of strain engineering.

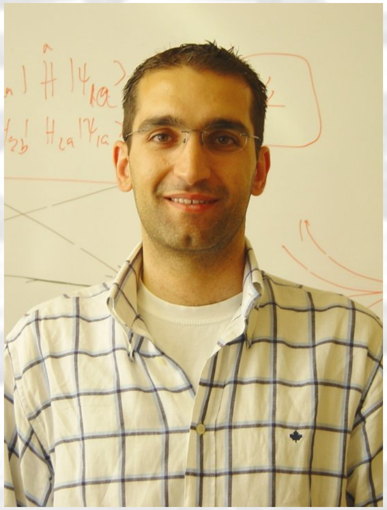




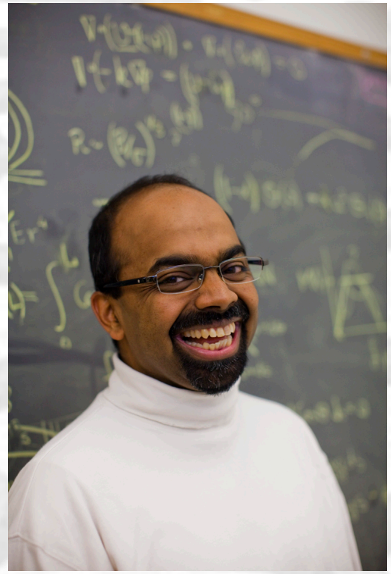




Collaborators on graphene's wrinkles



Vitor Pereira



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