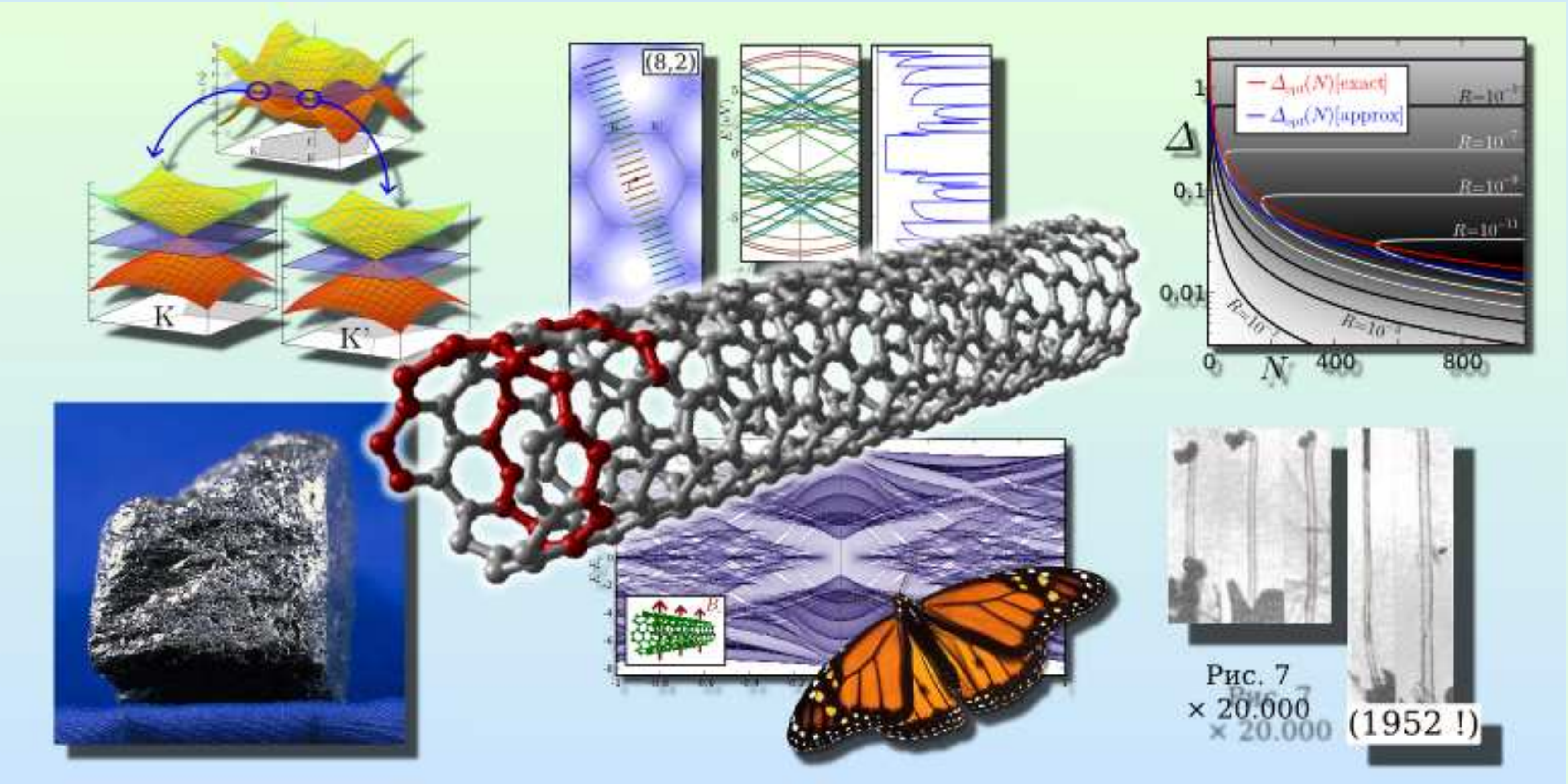
Quantum Transport in Carbon-based Nanostructures



Norbert Nemec

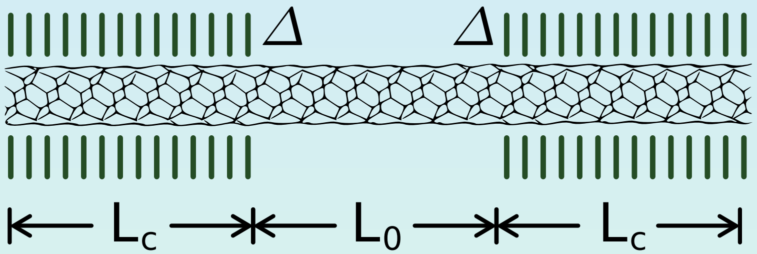
Promotionskolloquium

Regensburg, 27. Juli 2007

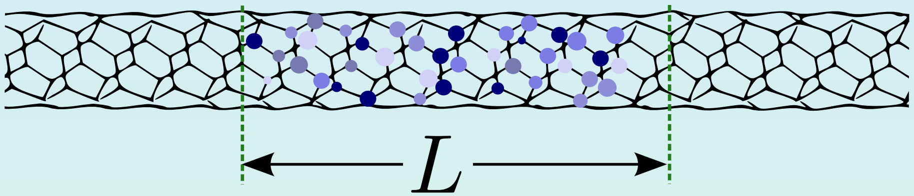
Outline

0) Background: Carbon hybridization and sp^2-carbon structures

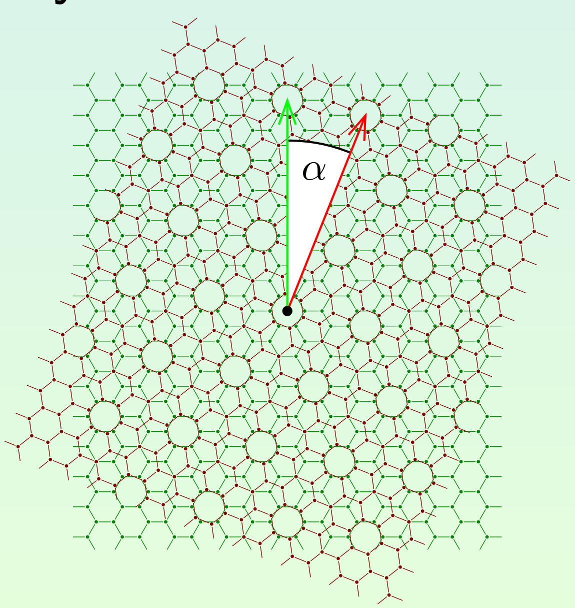
1) Electrical contacts to nanotubes



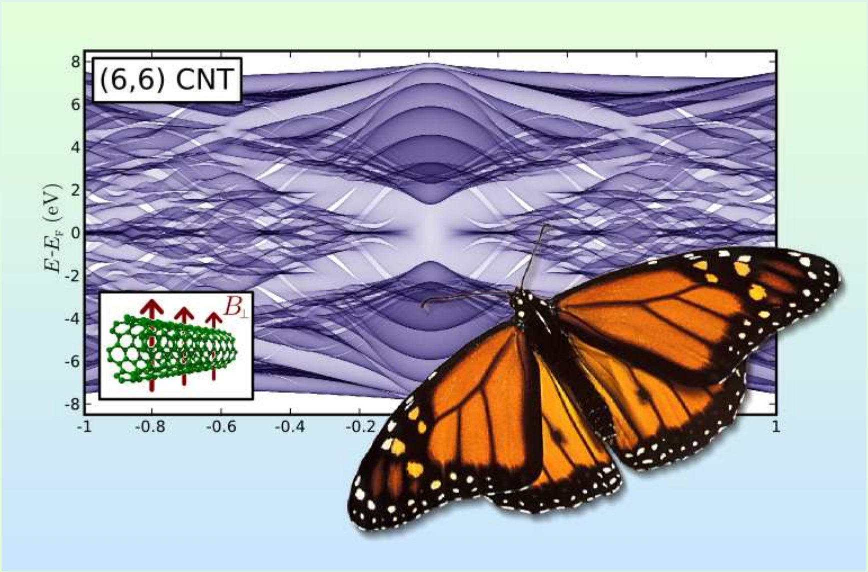
2) Length scales in disordered systems



3) Multilayer carbon structures

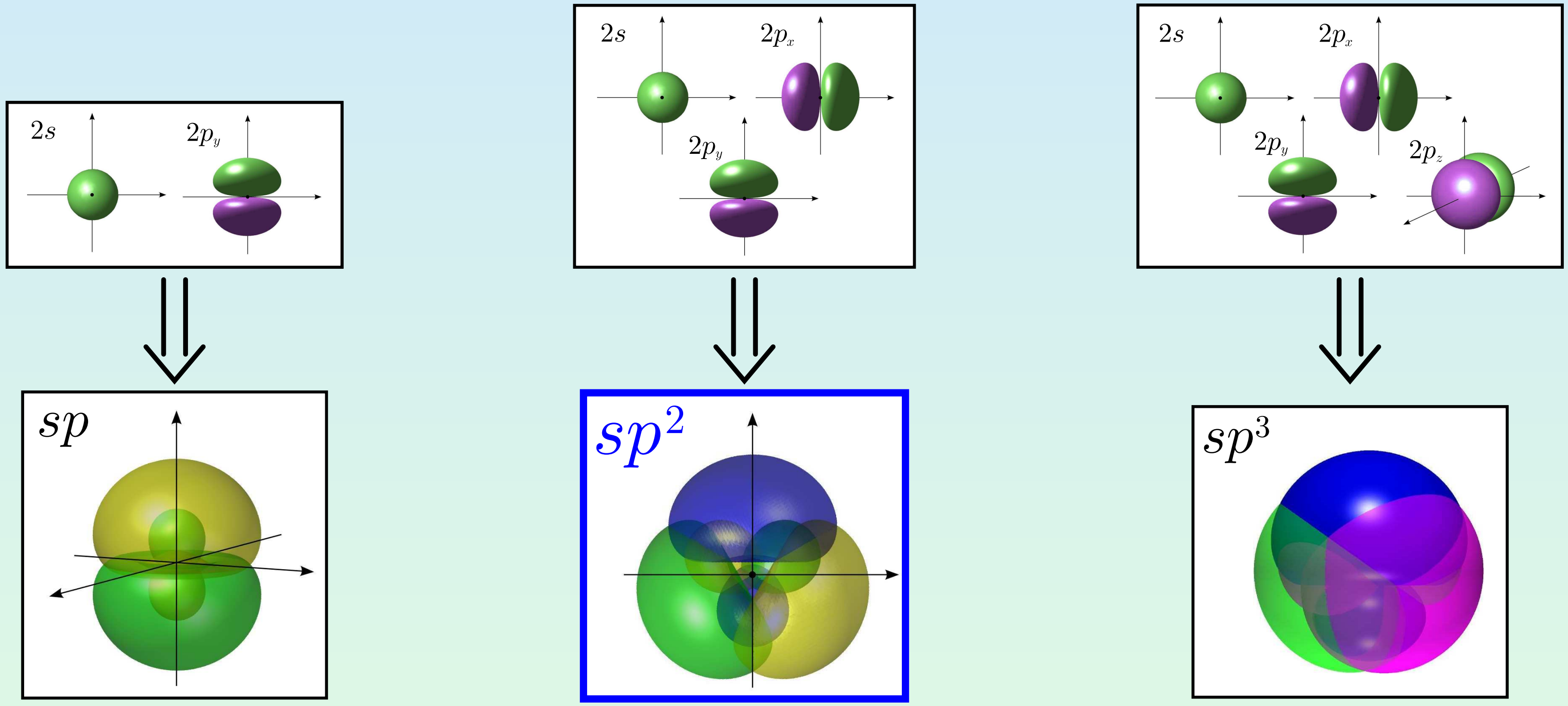


4) Magnetoelectronic structure

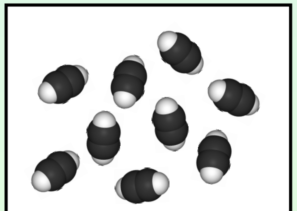


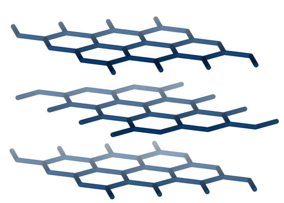
Orbital hybridization in carbon

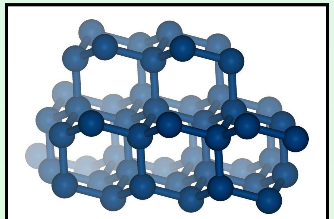
Carbon: atomic number 6, atomic ground state $1s^2 2s^2 2p^2$



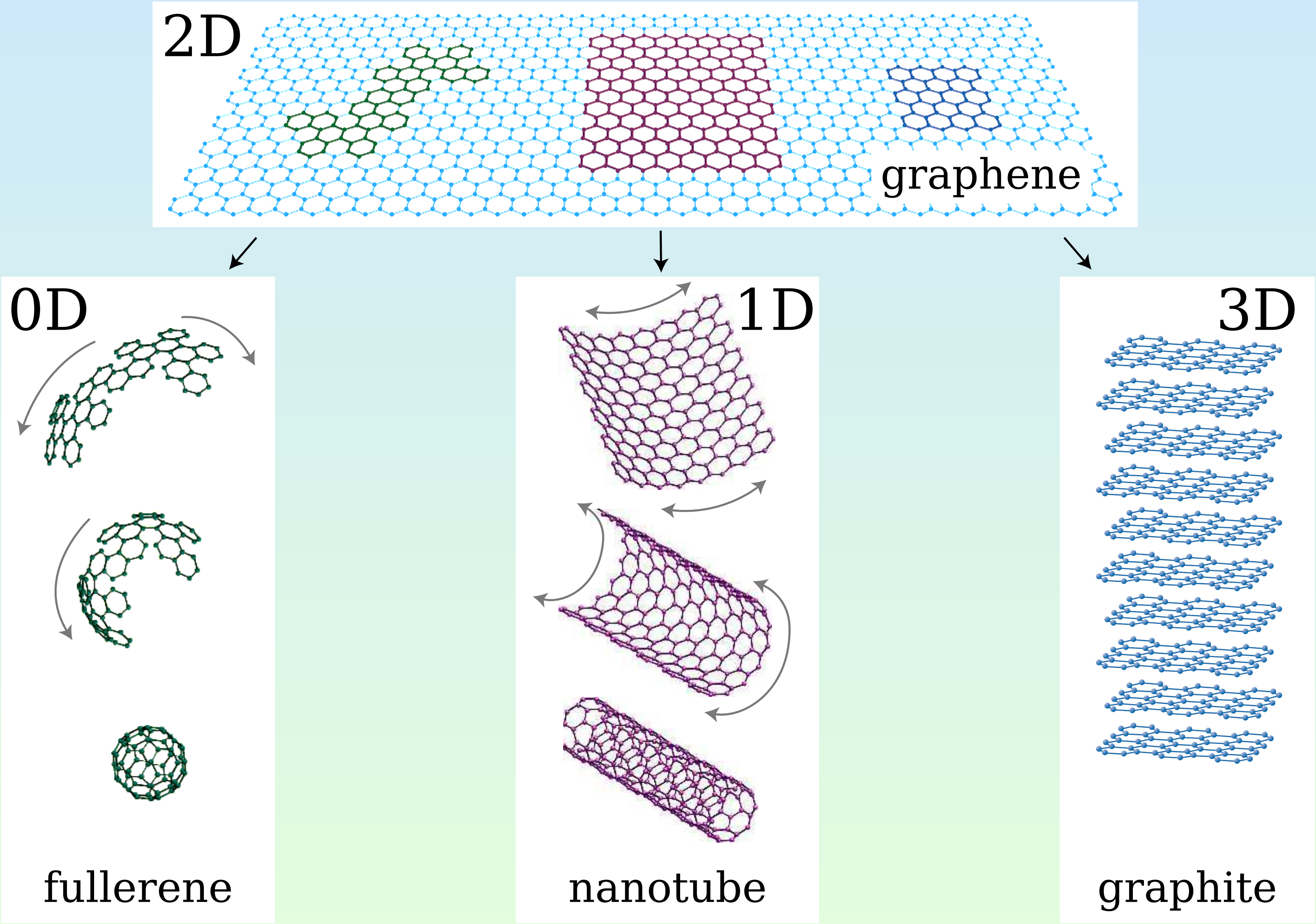
e.g.


acetylene


graphite


diamond

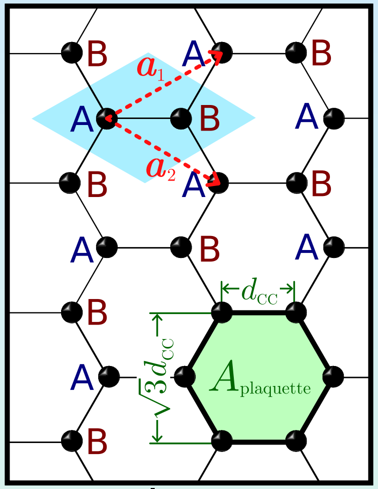
Nanostructures of sp^2-hybridized carbon



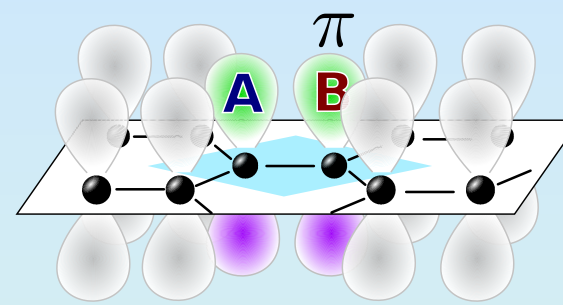
[figure from Geim and Novoselov, Nat. Mater. **6**, 183 (2007)]

Geometry and electronic structure of graphene

hexagonal lattice, 2-atom π-orbital basis:



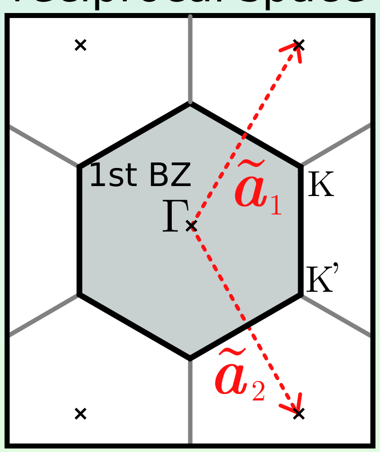
real space



$$\mathcal{H}(\boldsymbol{k}) = -\gamma_0 \begin{pmatrix} 0 & 1 + e^{i\boldsymbol{k}\boldsymbol{a}_1} + e^{i\boldsymbol{k}\boldsymbol{a}_2} \\ 1 + e^{-i\boldsymbol{k}\boldsymbol{a}_1} + e^{-i\boldsymbol{k}\boldsymbol{a}_2} & 0 \end{pmatrix}$$

$$E(\boldsymbol{k}) = \pm\gamma_0 \left| 1 + e^{i\boldsymbol{k}\boldsymbol{a}_1} + e^{i\boldsymbol{k}\boldsymbol{a}_2} \right|$$

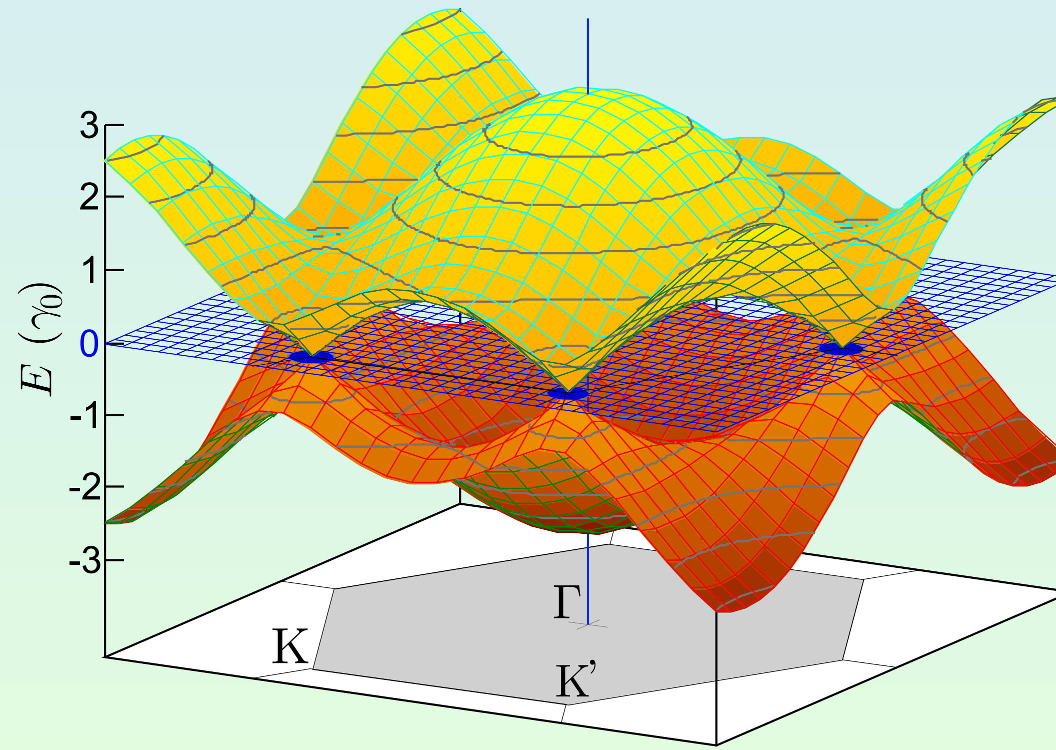
reciprocal space



$\Rightarrow$ pointlike Fermi-"surface" at K-points

$\Rightarrow$ semi-metallic character

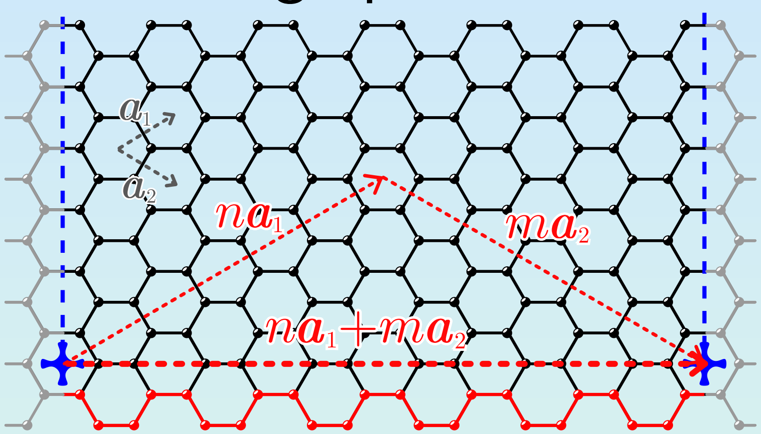
$\Rightarrow$ massless bands at E_F (Dirac-like theory)



[Wallace, Phys. Rev. **71**, 622 (1947)]

Carbon nanotubes

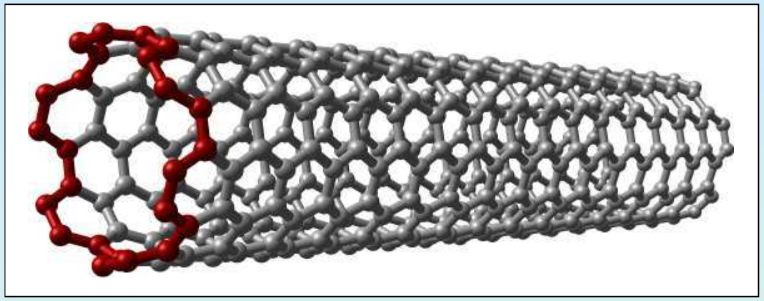
graphene



rollup

⟶

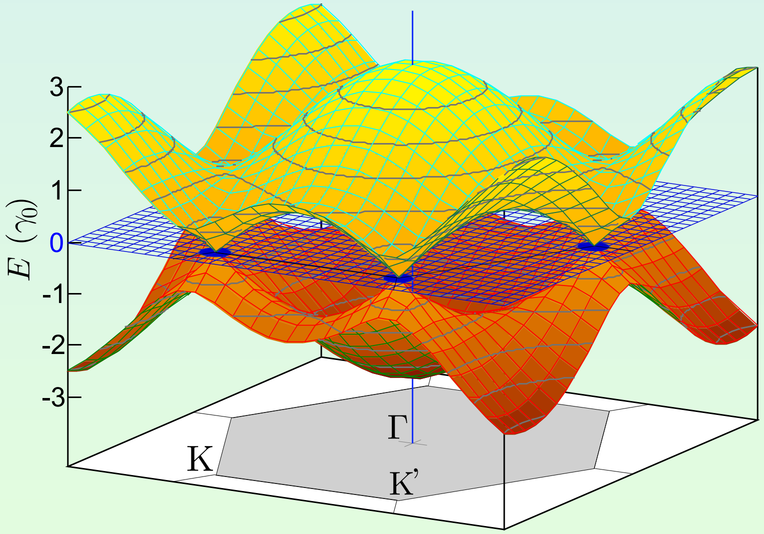
e.g.: (6,6) CNT



real space

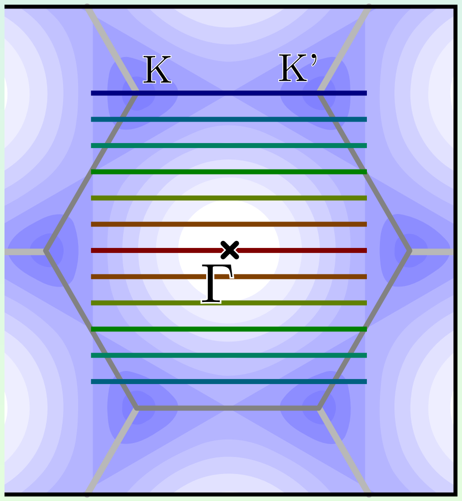
periodic boundaries

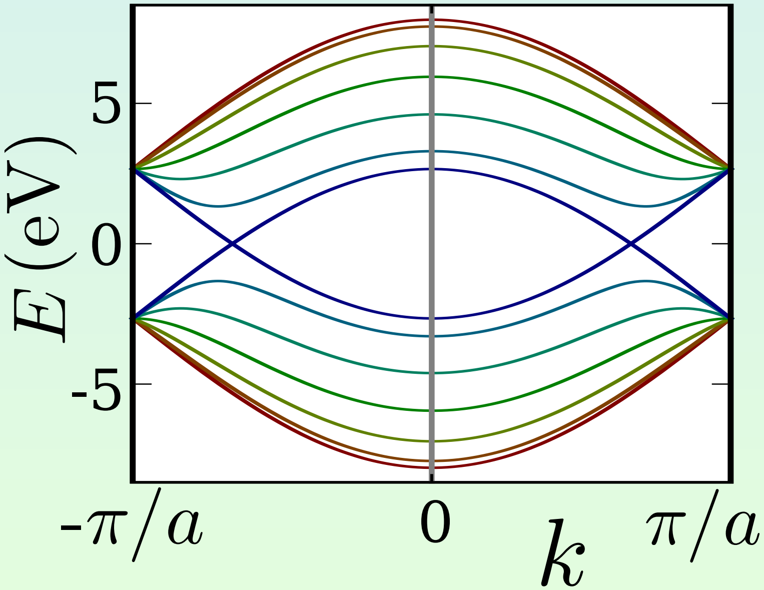
reciprocal space



zone-folding

⟶





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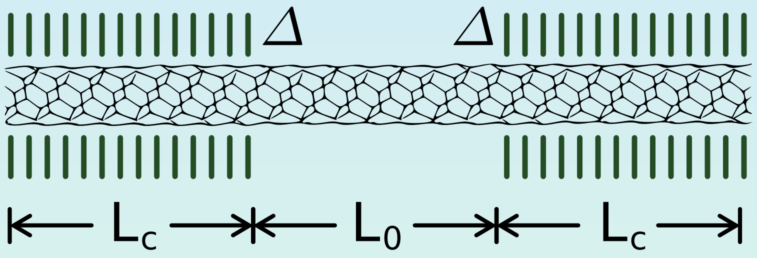
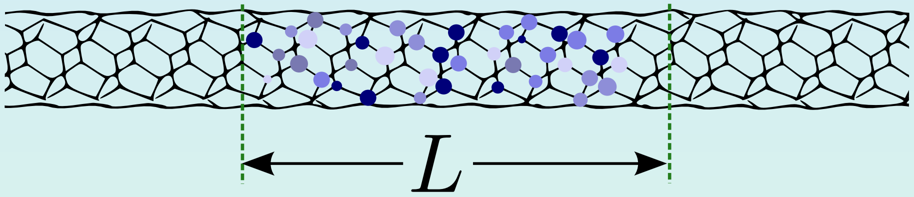


N. Nemec, D. Tománek and G. Cuniberti, Phys. Rev. Lett. **96**, 076802 (2006)

2) Length scales in disordered systems



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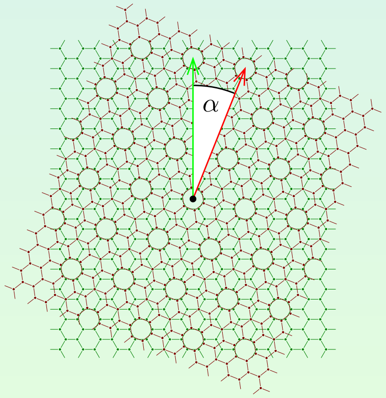

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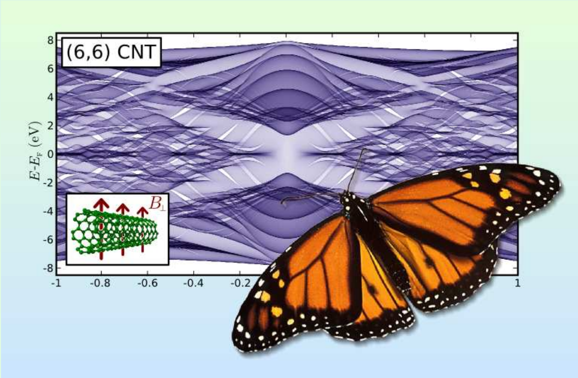
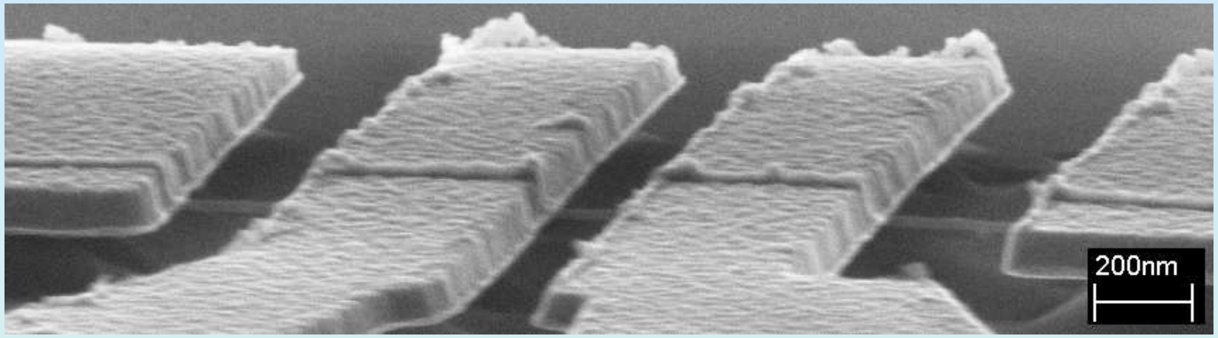


Extended contacts to carbon nanotubes

CNT-transport measurements – typical experimental setup:



courtesy of
C. Strunk
Regensburg, 2004

(contacts $\gtrsim$100nm by metal evaporation, e.g. Au, Cr, Pd etc.)

¿

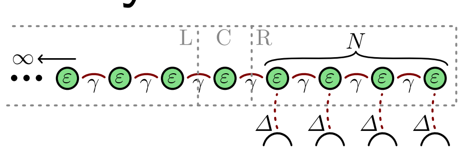
material dependent contact transparency: Au/Cr < Ti < Pd
varying results about the *effective contact length*

?

[e.g. Javey *et al.*, Nature **424**, 654 (2003)]

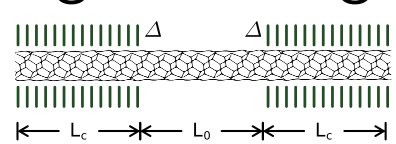
$\Rightarrow$ our study:

analytical model



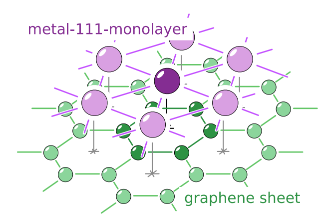
+

tight-binding



+

ab initio



[preprint]

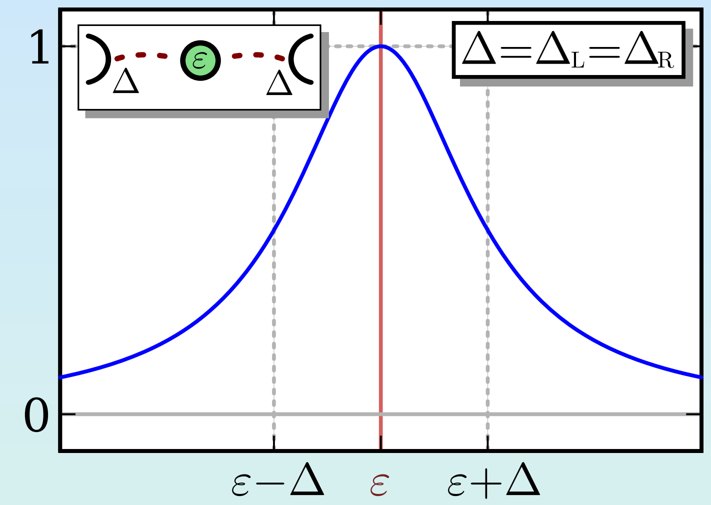
[Phys. Rev. Lett. **96**, 076802 (2006)]

Breit-Wigner resonance in molecular junction

Breit-Wigner resonance of single energy level:

$$T(E) = \frac{4\Delta_\mathrm{L}\Delta_\mathrm{R}}{4(E-\varepsilon)^2 + (\Delta_\mathrm{L}+\Delta_\mathrm{R})^2}$$

neutron capture: Breit and Wigner, Phys. Rev. **49**, 519 (1936)
resonant tunneling: Stone and Lee, Phys. Rev. Lett. **54**, 1196 (1985)



Breit-Wigner resonance in molecular junction

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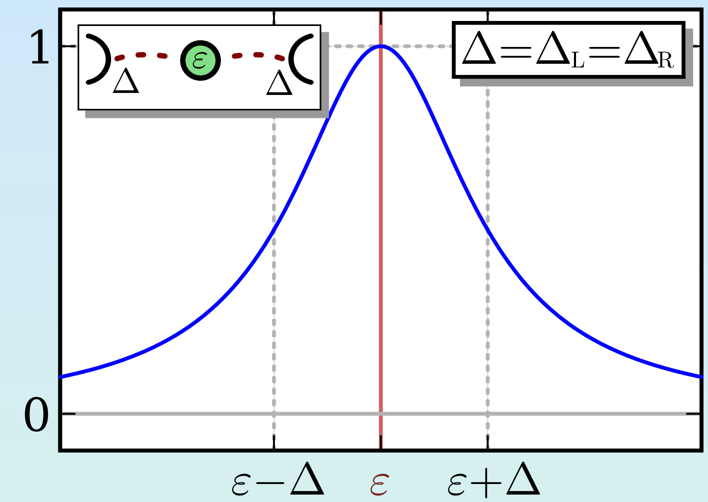
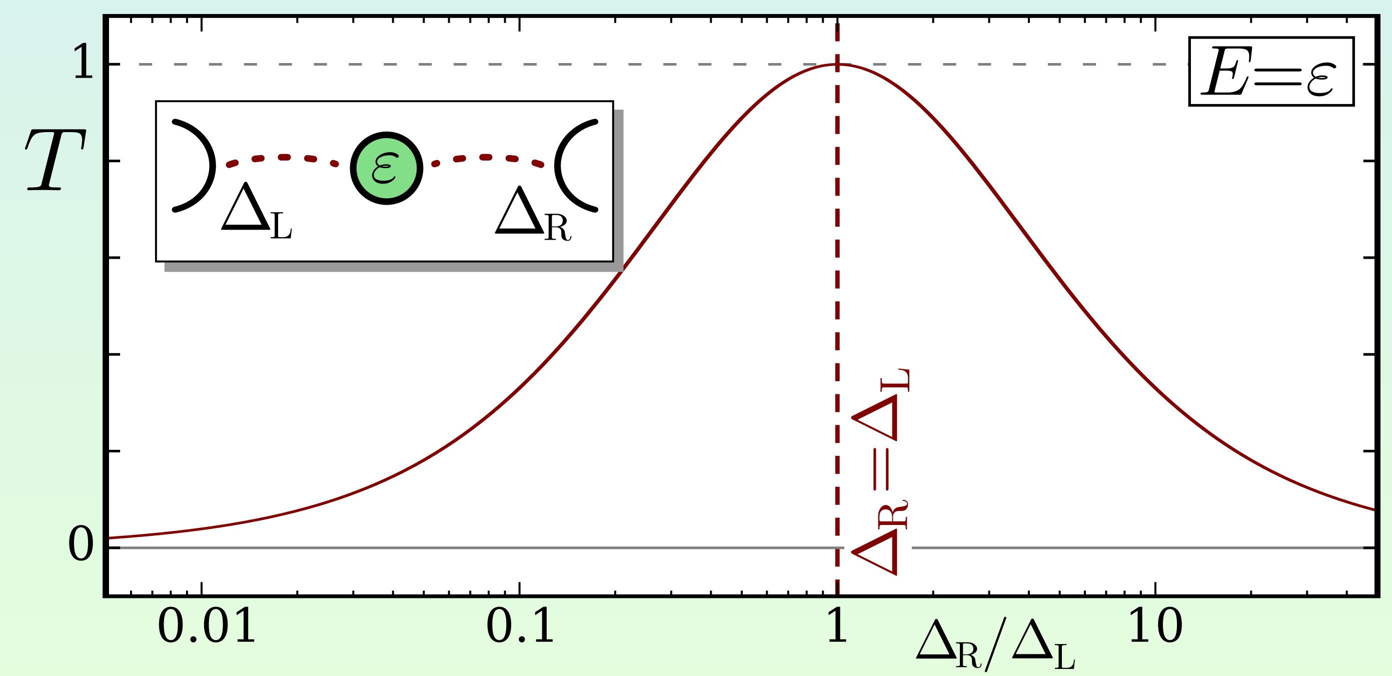
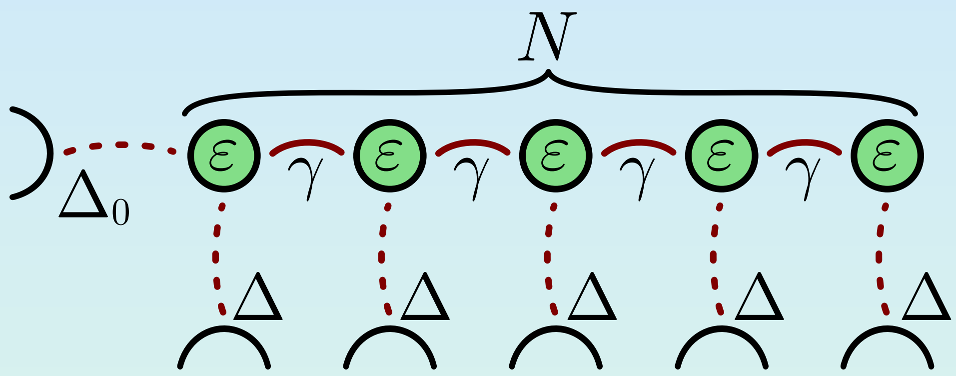


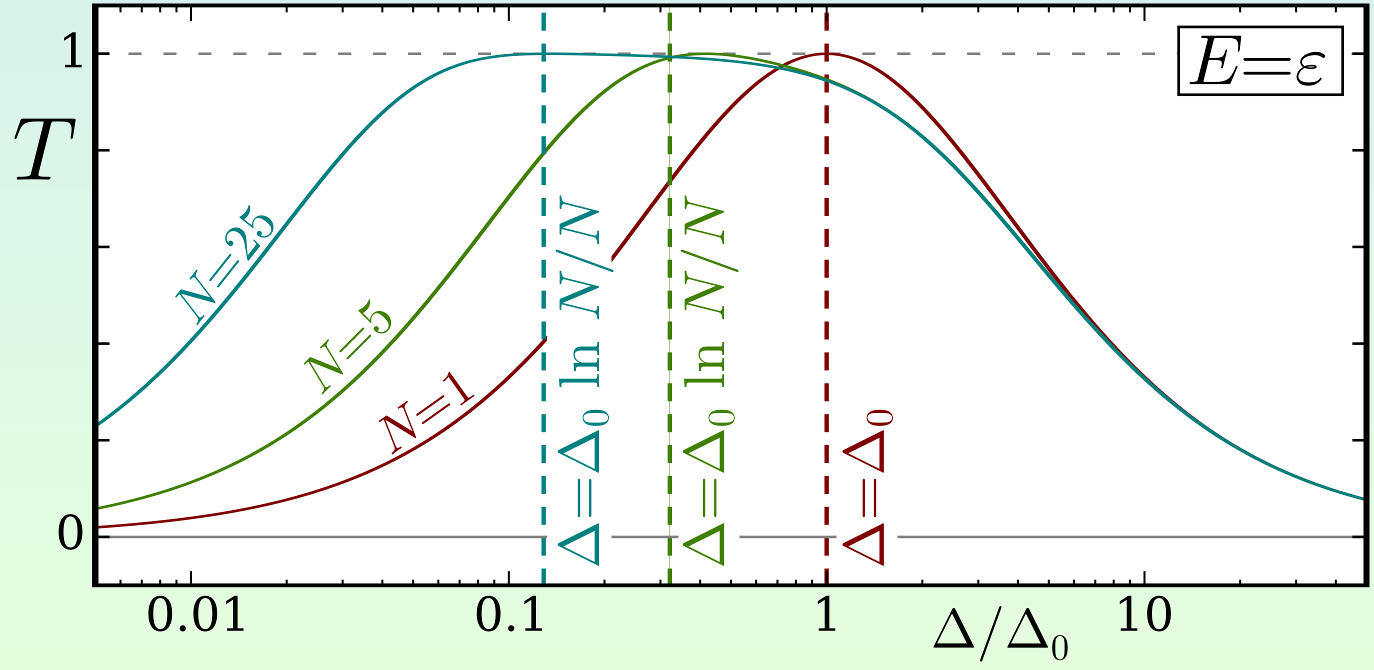
For $E = \varepsilon$:

$$T = 4\left(\frac{\Delta_L}{\Delta_R} + \frac{\Delta_R}{\Delta_L}\right)^{-2}$$

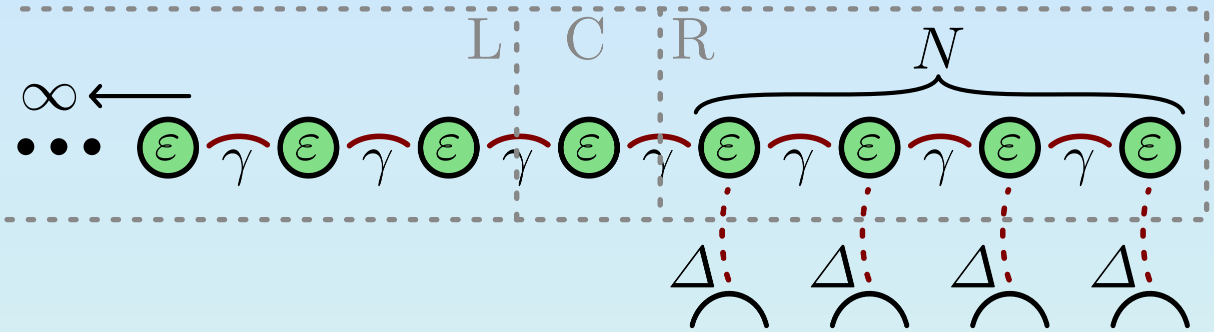


Breit-Wigner resonance of extended molecule





Minimal model for contact to CNT



→ semiinfinite linear chain

on-site energy $\varepsilon = 0$
hopping paramter $\gamma = 1$

→ contact length N

→ contact strength Δ

$$T(E) = \frac{8\sqrt{4-E^2}\,\mathrm{Im}(f_N(E/2-\mathrm{i}\Delta/4))}{\left|E-\mathrm{i}\sqrt{4-E^2}-2f_N(E/2-\mathrm{i}\Delta/4)\right|^2}$$

$f_N = U_{N-1}(x)/U_N(x)$ $\qquad$ $U_N(x)$: Chebyshev polynomials (2nd kind)

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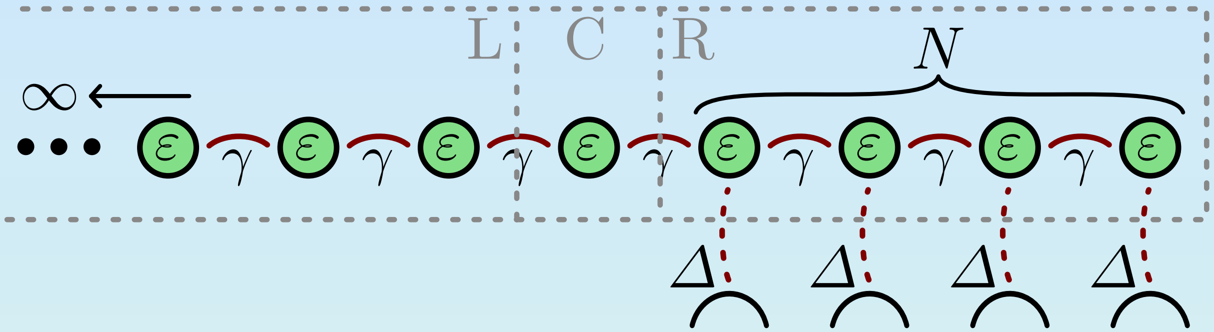

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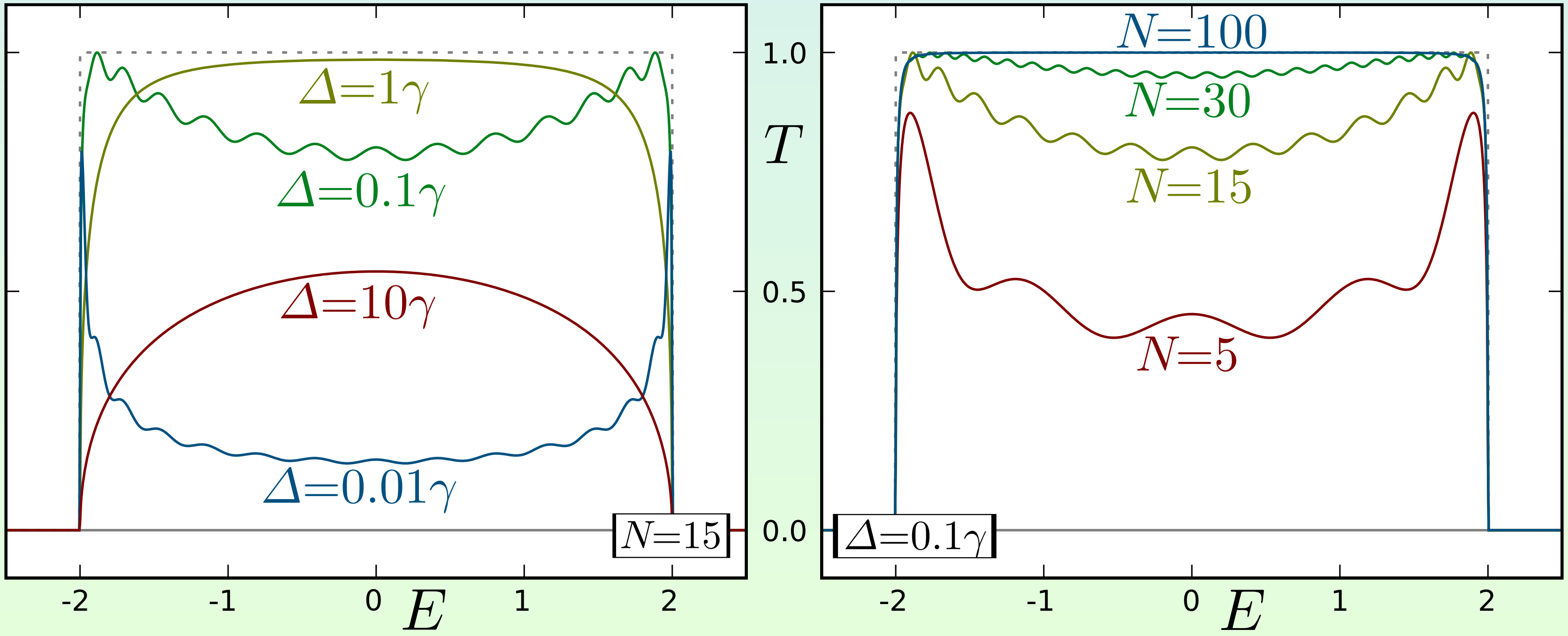
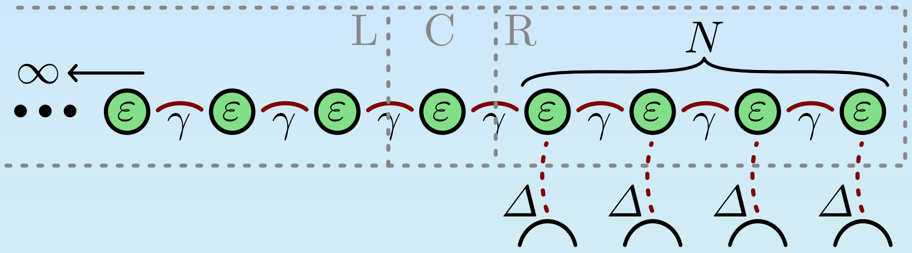
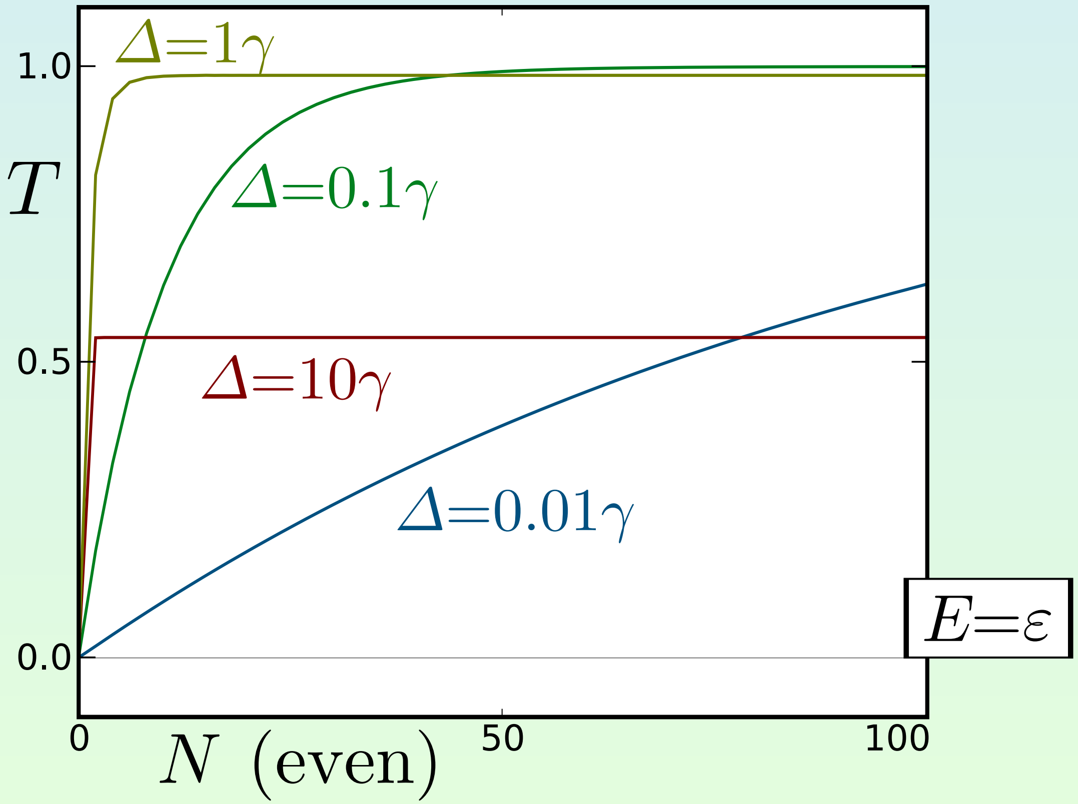


Length saturation of transmission

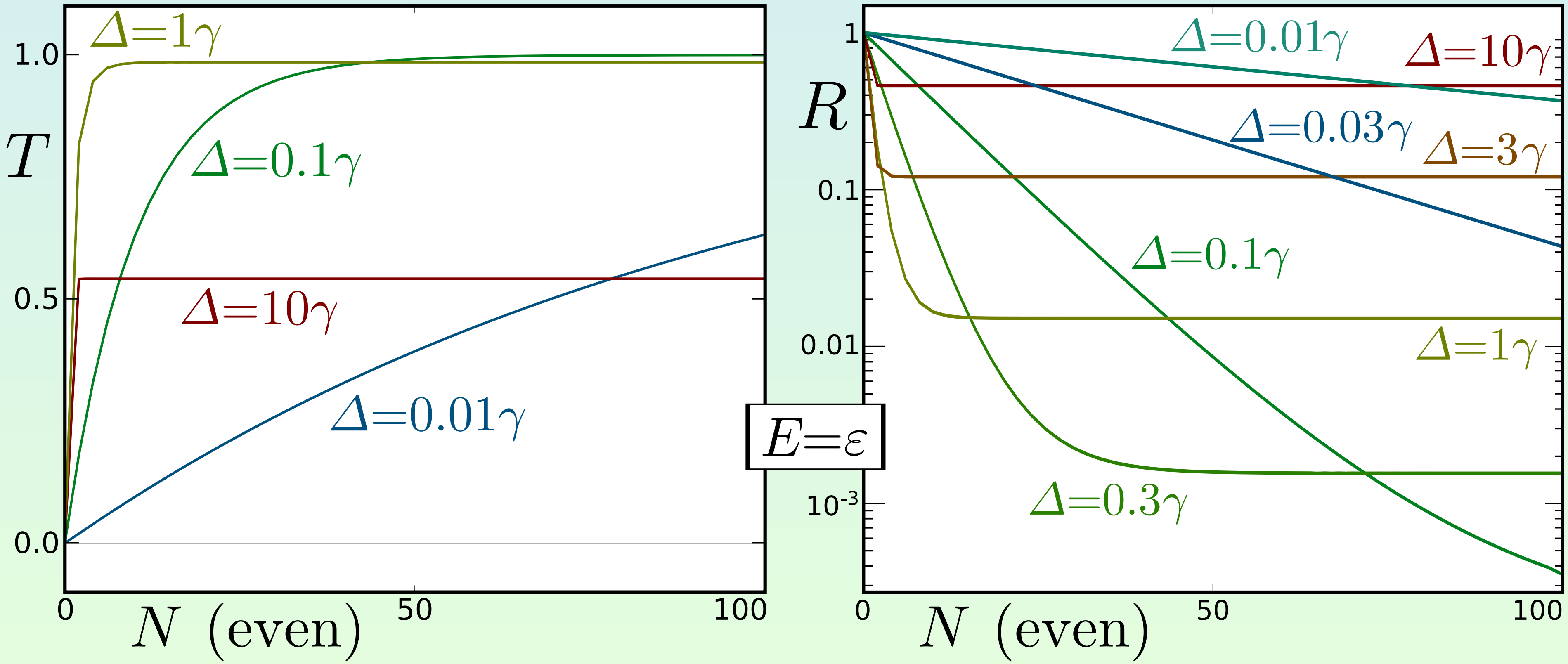




Length saturation of contact reflection

contact reflection: $R = 1 - T$

N-resonant regime: $R \approx \exp(-N\Delta)$ $\longleftrightarrow$ N-independent regime: $R \approx \Delta^2/64$



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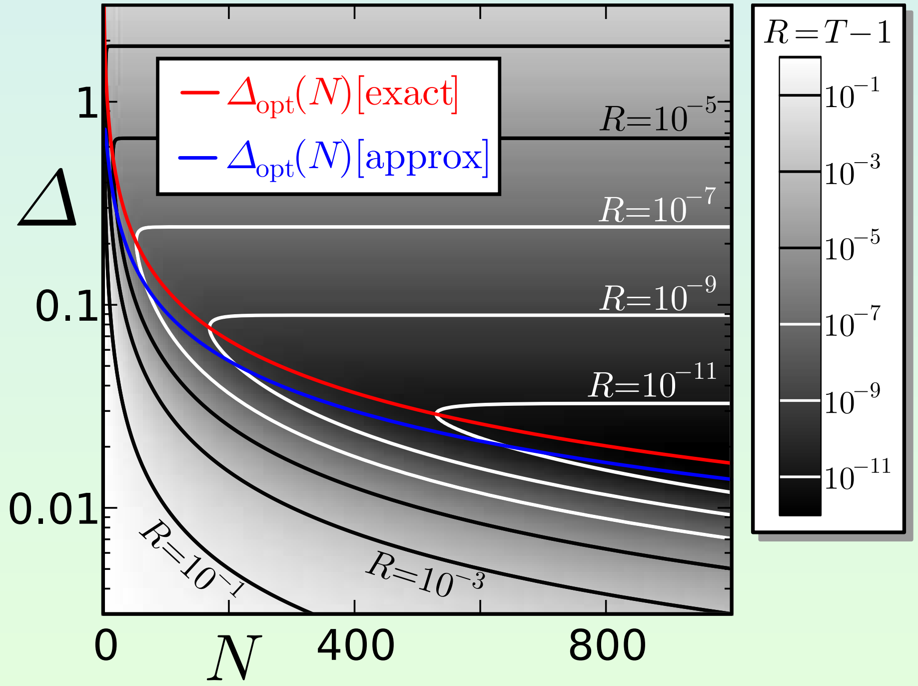
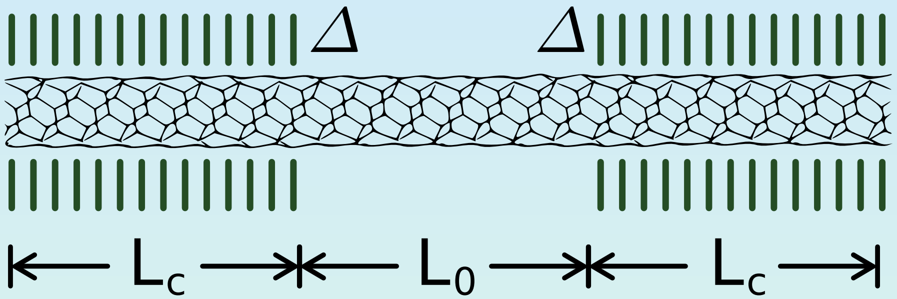


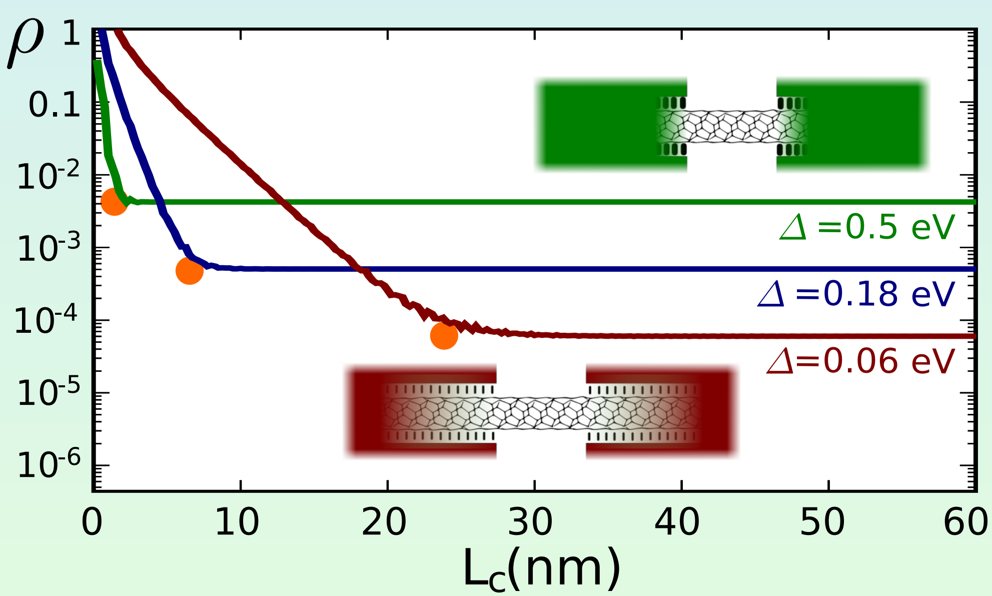
$$N_{\text{eff}}(\Delta) = \frac{2}{\Delta}\ln\left(\frac{8}{\Delta}\right)$$

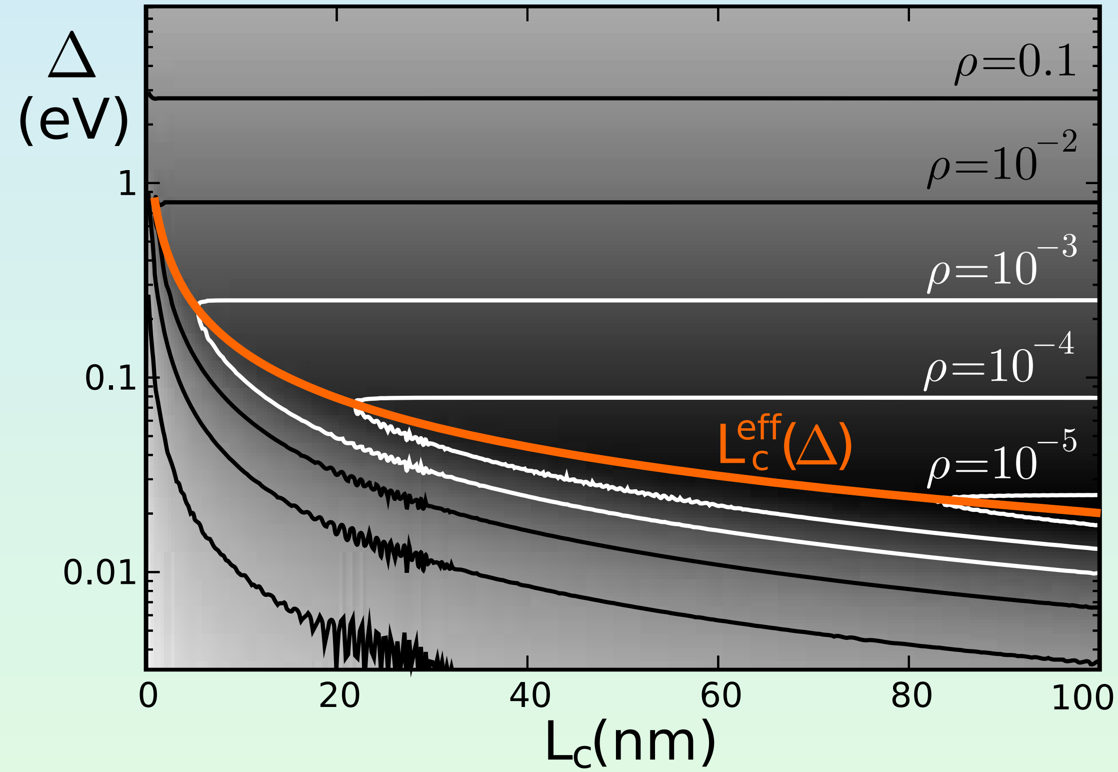
$$\Delta_{\text{opt}}(N) \approx \frac{2}{N}\ln N$$

Carbon nanotube in two-terminal setup

- finite length L_0 $\rightarrow$ Fabry-Perot oscillations (averaged out $\Rightarrow L_0$-independent results)





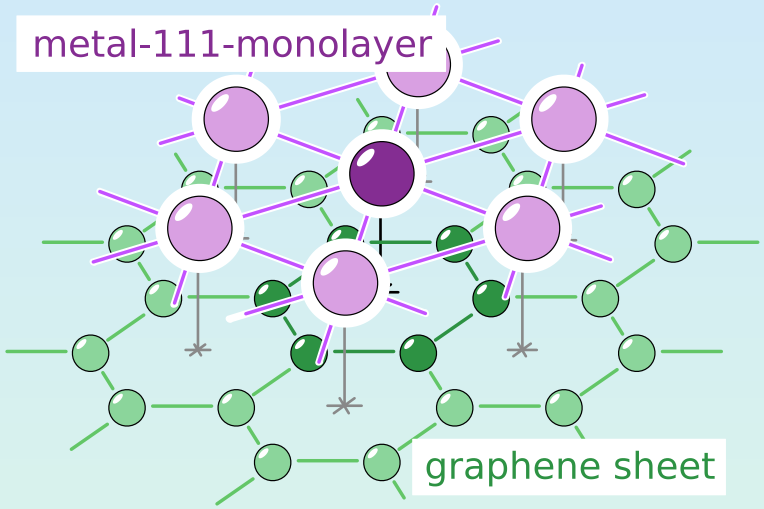


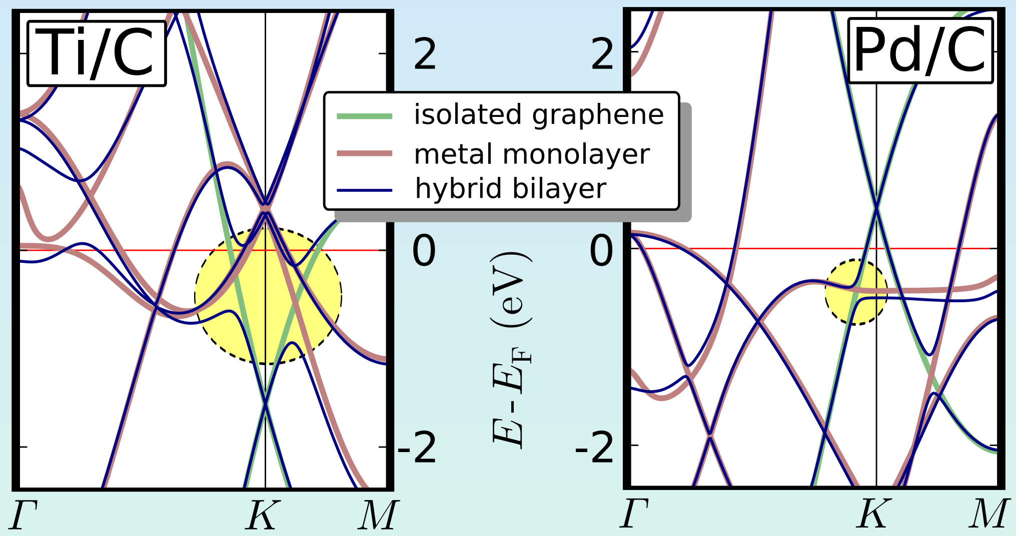
$$L_c^{\rm eff}(\Delta) = \ell_{\rm uc}\frac{\alpha_1}{\Delta}\ln\frac{\alpha_2}{\Delta} \qquad (\alpha_1 = 1.34\,{\rm eV},\ \alpha_2 = 9.14\,{\rm eV})$$

[N. Nemec, D. Tománek and G. Cuniberti, Phys. Rev. Lett. **96**, 076802 (2006)]

Ab initio results for contact metals Ti and Pd

microscopic DFT-study, using SIESTA *(LDA-PZ)*:





→ better matching of work function in Pd/C (less charge transfer)

→ lower binding energy in Pd/C

	Ti	Pd
$\Delta \quad = \gamma^2_{\mathrm{Me/C}} \times \mathrm{SDOS}_{\mathrm{metal}}$	0.1 eV	0.02 eV
$L^{\mathrm{c}}_{\mathrm{eff}} = \ell_{\mathrm{uc}} \frac{\alpha_1}{\Delta} \ln \frac{\alpha_2}{\Delta}$	$\sim 4\,\mathrm{nm}$	$\sim 30\,\mathrm{nm}$

⇒ transparent Pd contacts due to weak coupling

[N. Nemec, D. Tománek and G. Cuniberti, Phys. Rev. Lett. **96**, 076802 (2006)]

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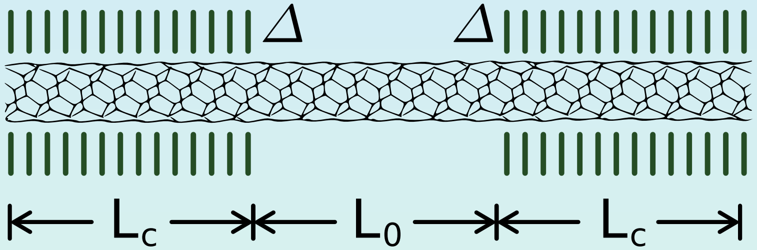

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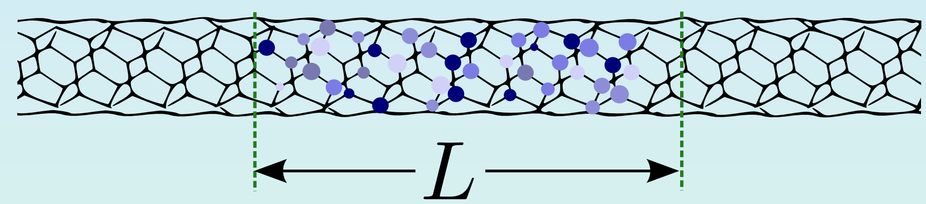

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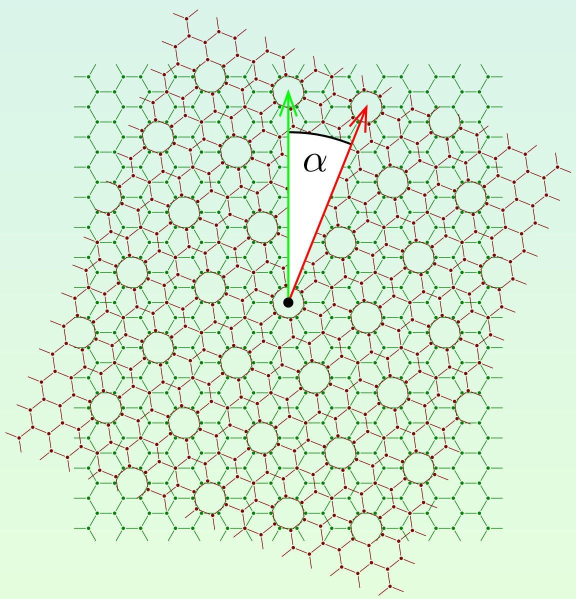

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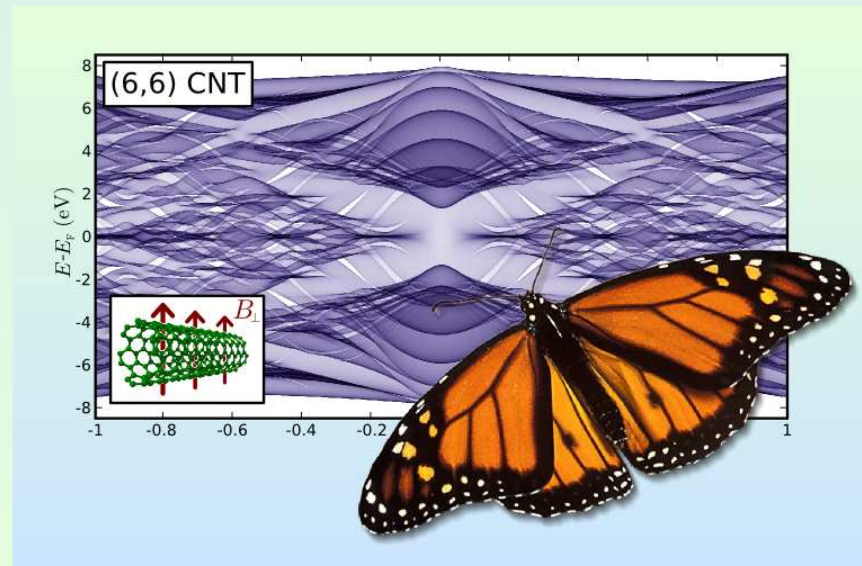
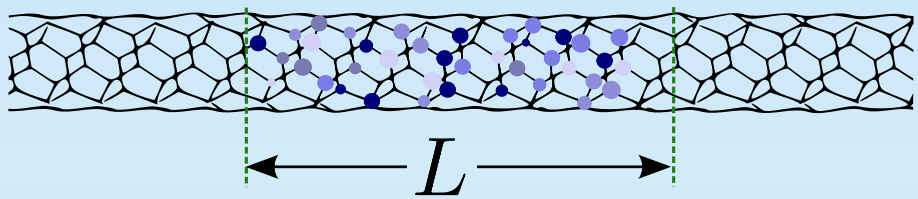


Length scales in disordered carbon systems

diffusive regime:

$$R = \frac{h}{2e^2 N_{\rm ch}}\left(1 + \frac{L}{\ell_{\rm el}}\right)$$



localized regime:

$$R \sim \exp\left(\frac{L}{\ell_{\rm loc}}\right)$$

elastic mean free path:

$$\ell_{\rm el} := -\frac{\rm d}{{\rm d}L} T/N_{\rm ch}\Big|_{L\to 0}$$

(obtained analytically)

crossover regime:

$$R = \frac{h}{2e^2 N_{\rm ch}}\left(\frac{\ell_{\rm loc}}{\ell_{\rm el}}\sinh\frac{L}{\ell_{\rm loc}} + \cosh\frac{L}{\ell_{\rm loc}}\right)$$

[Tartakovski, Phys. Rev. B **52**, 2704 (1995)]

localization length:

$$\ell_{\rm loc} := -\lim_{L\to\infty}\frac{L}{\langle \ln T\rangle}$$

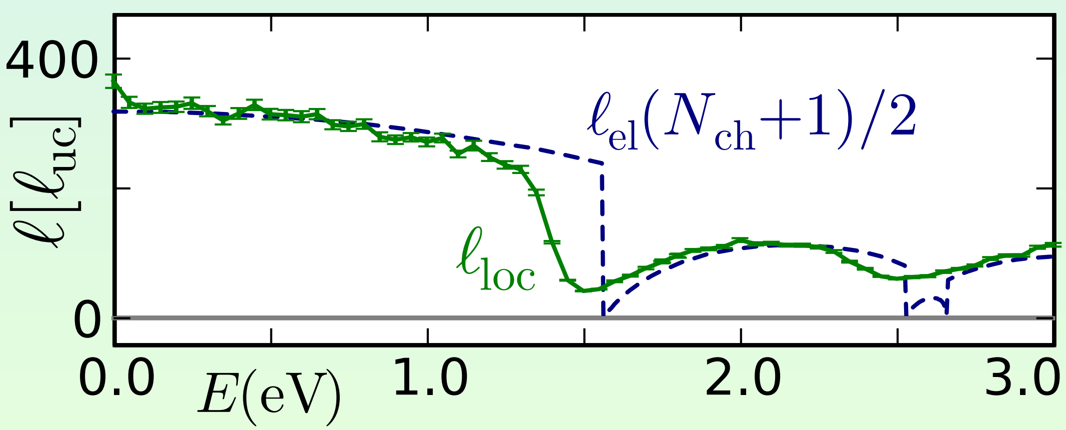
(obtained numerically)

$$\ell_{\rm loc} \approx \frac{\beta(N_{\rm ch}-1)+2}{2}\,\ell_{\rm el}$$

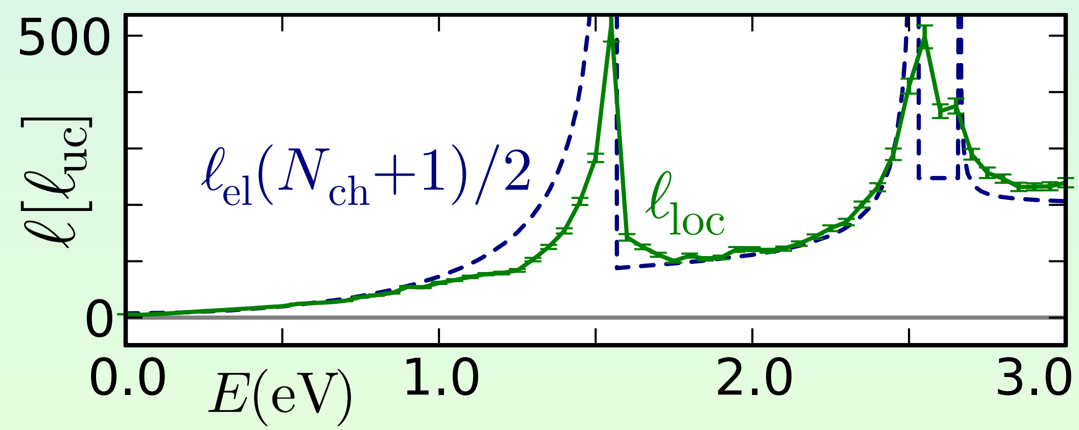
($\beta = 1$ for time-reversal-invariant systems)

[Beenakker, Rev. Mod. Phys. **69**, 731 (1997)]

Anderson disorder:



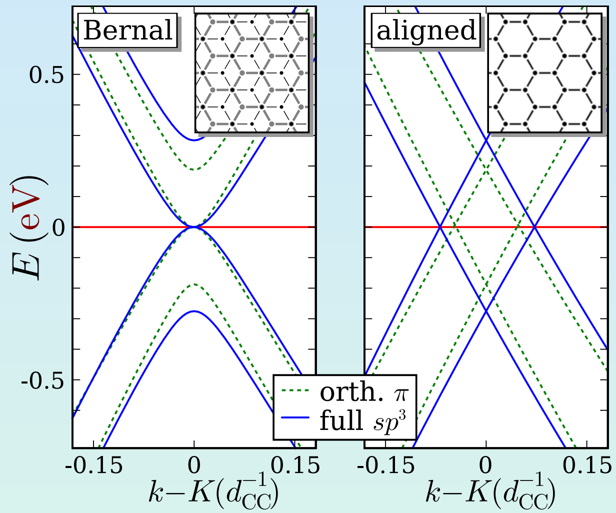
Point vacancies:



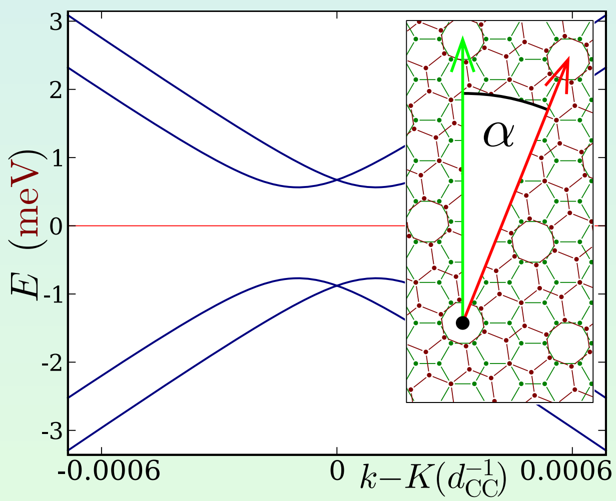
Multilayer graphene and carbon nanotubes

approximate momentum conservation

signficiant interlayer coupling for identically oriented bilayers ($\Delta E \sim 0.2\,\mathrm{eV}$)



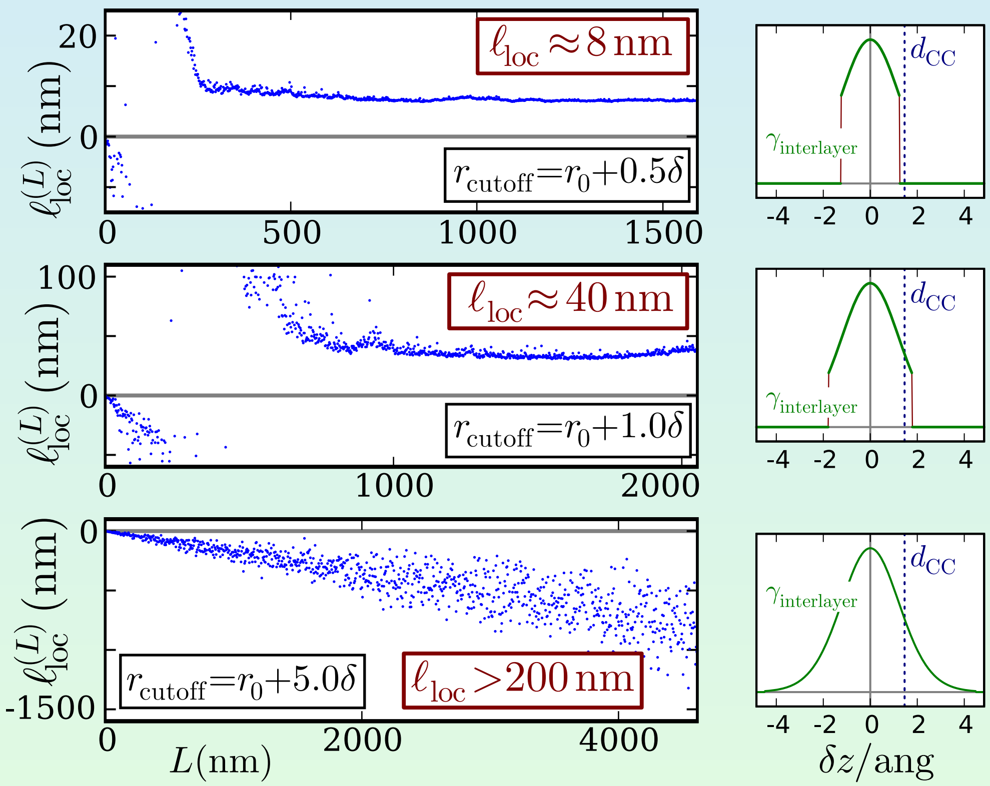
suppressed interlayer coupling for rotated bilayers ($\Delta E \sim 2\,\mathrm{meV}$)



→ significant interwall conductance at E_F only in *armchair* DWCNTs

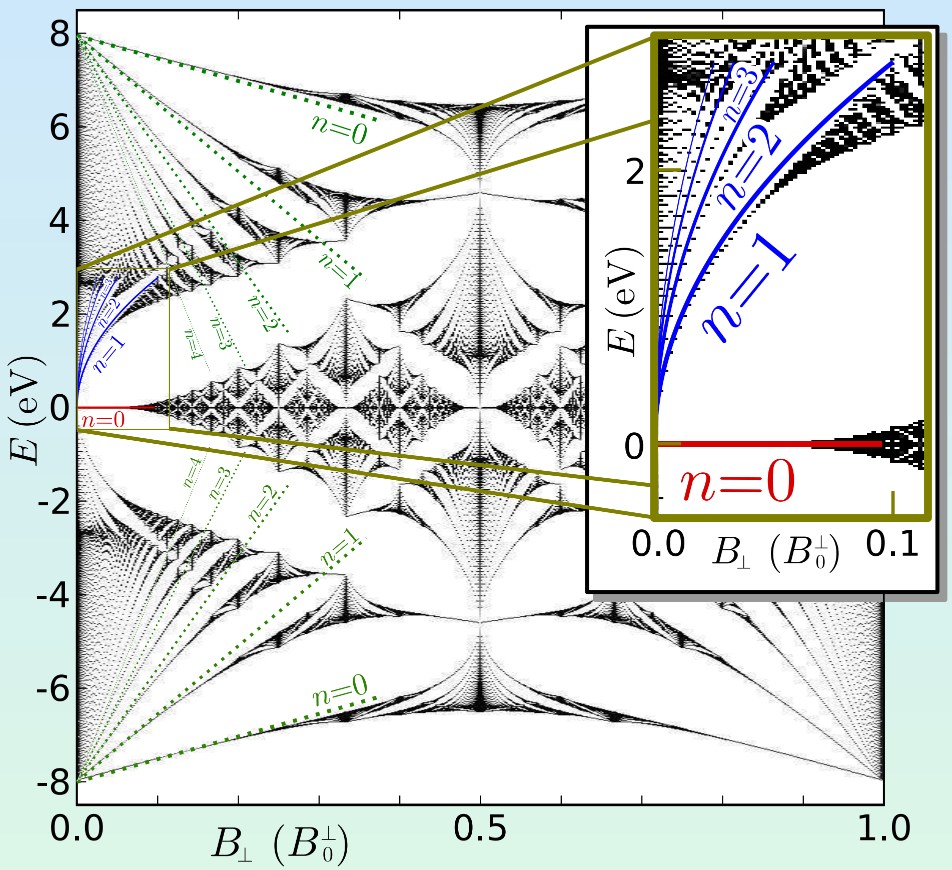
incommensurability-induced disorder

localization depends on modelling details of the interlayer coupling $\gamma_{\mathrm{interlayer}}$:



→ smooth (realistic?) interlayer coupling does not induce disorder effects

Hofstadter butterfly of graphene



Rammal, J. Phys. (Paris) **46**, 1345 (1985)

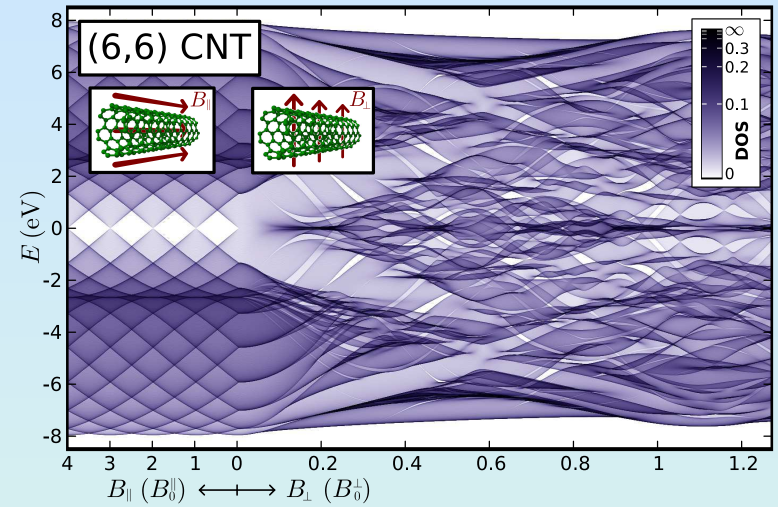
Landau levels (LL):

conventional: $E = \pm \frac{\hbar e}{2m^*} B\,(2n+1)$

relativistic: $E = \pm\, v_{\mathrm{F}} \sqrt{2e\hbar\, B\, n}$

supersymmetric: $E = 0$

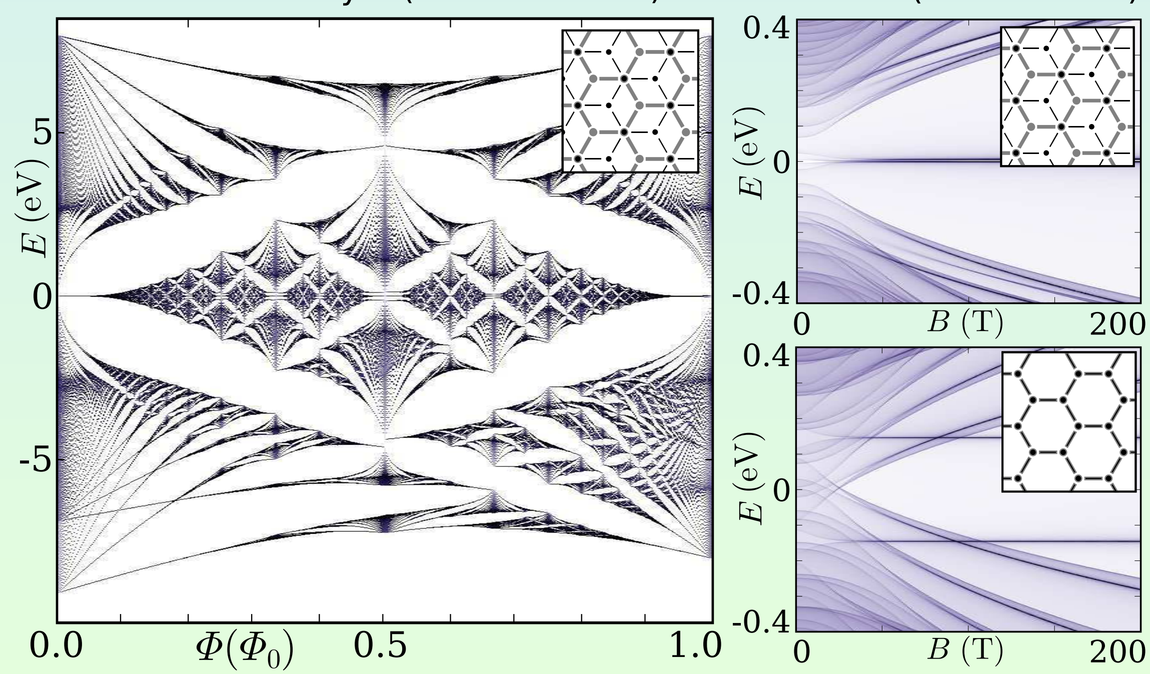
Hofstadter butterfly of carbon nanotubes



Nemec and Cuniberti, Phys. Rev. B **74**, 165411 (2006)

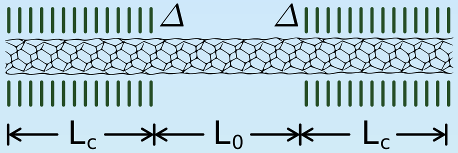
Hofstadter butterfly of bilayer graphene

2D infinite bilayer (Bernal-stacked) ribbons (width 20 nm)



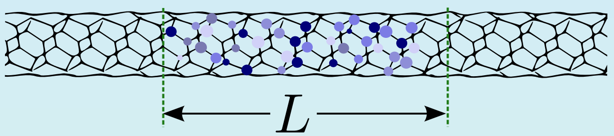
Nemec and Cuniberti, Phys. Rev. B **75**, 201404(R) (2007)

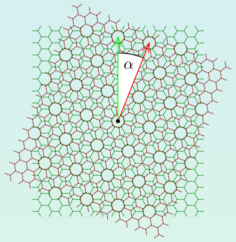
Summary



A) Extended contacts to carbon nanotubes:
weaker coupling ↔ higher transparency

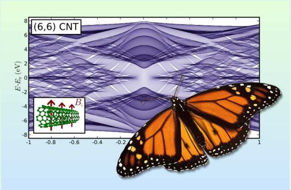
B) Effects of disorder:
elastic mean free path ↔ localization length





C) Multilayer systems:
approximate momentum conservation / incommensurability

D) Magnetoelectronic structure:
Hofstadter butterflies / anomalous Landau levels



– – Thanks – –

Giovanni Cuniberti ... and many, many others

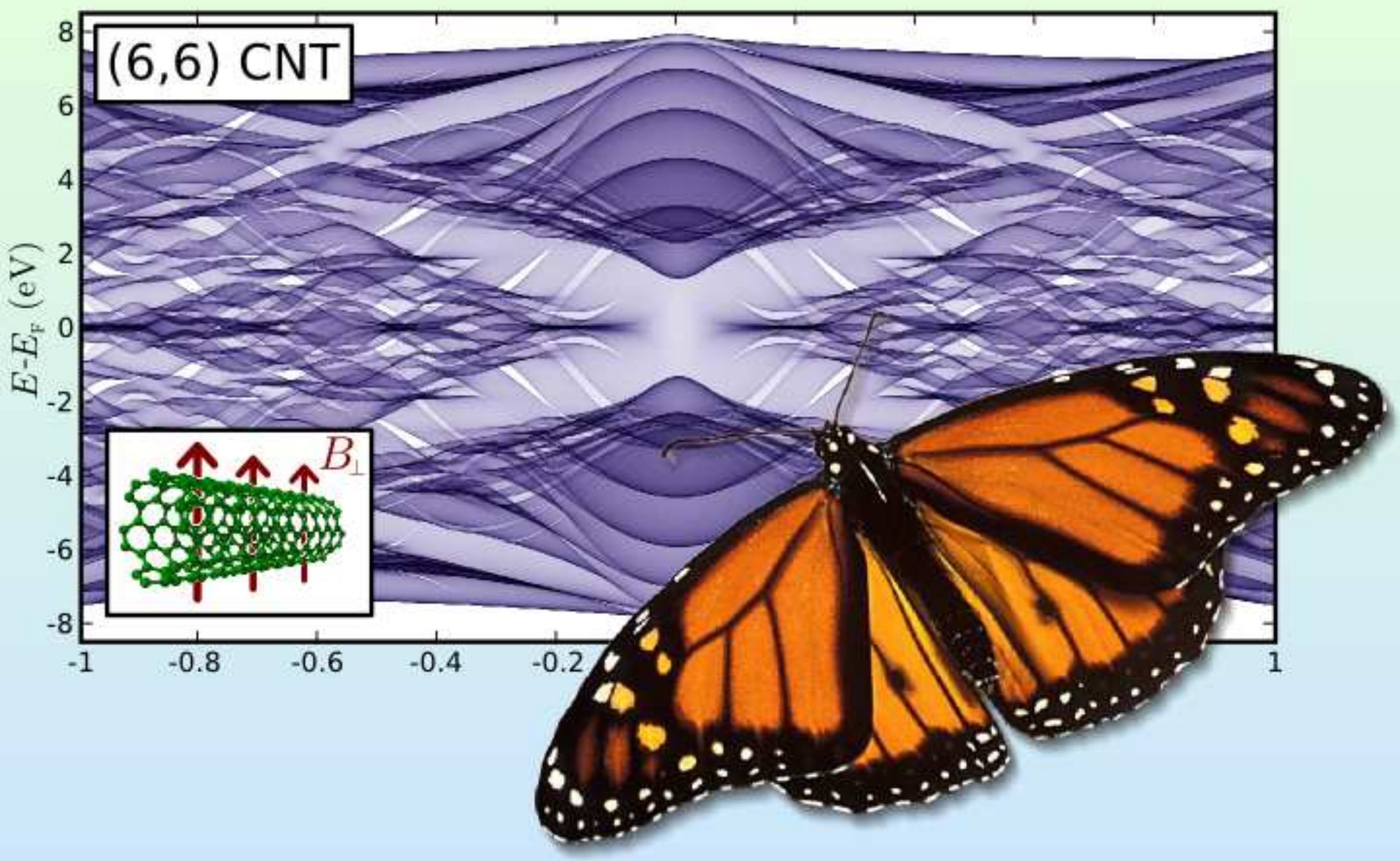
Additional slides

Supersymmetry in graphene

$$\mathcal{H}_D = \begin{pmatrix} 0 & \mathcal{Q} \\ \mathcal{Q} & 0 \end{pmatrix}; \; Q = v_\mathrm{F}(\sigma_x \mathcal{P}_x + \sigma_y \mathcal{P}_y)$$

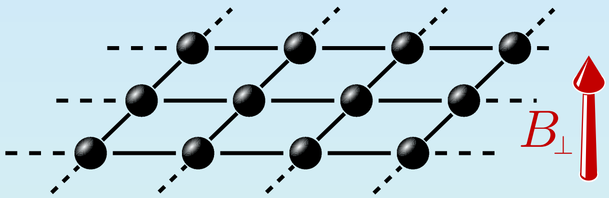
$$\mathcal{U}\mathcal{H}_D\mathcal{U}^\dagger = \begin{pmatrix} \mathcal{Q} & 0 \\ 0 & -\mathcal{Q} \end{pmatrix}$$

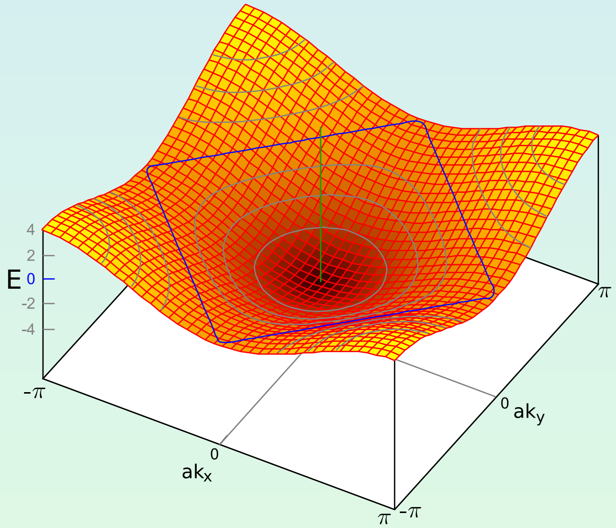
Hofstadter butterflies of graphitic nanostructures

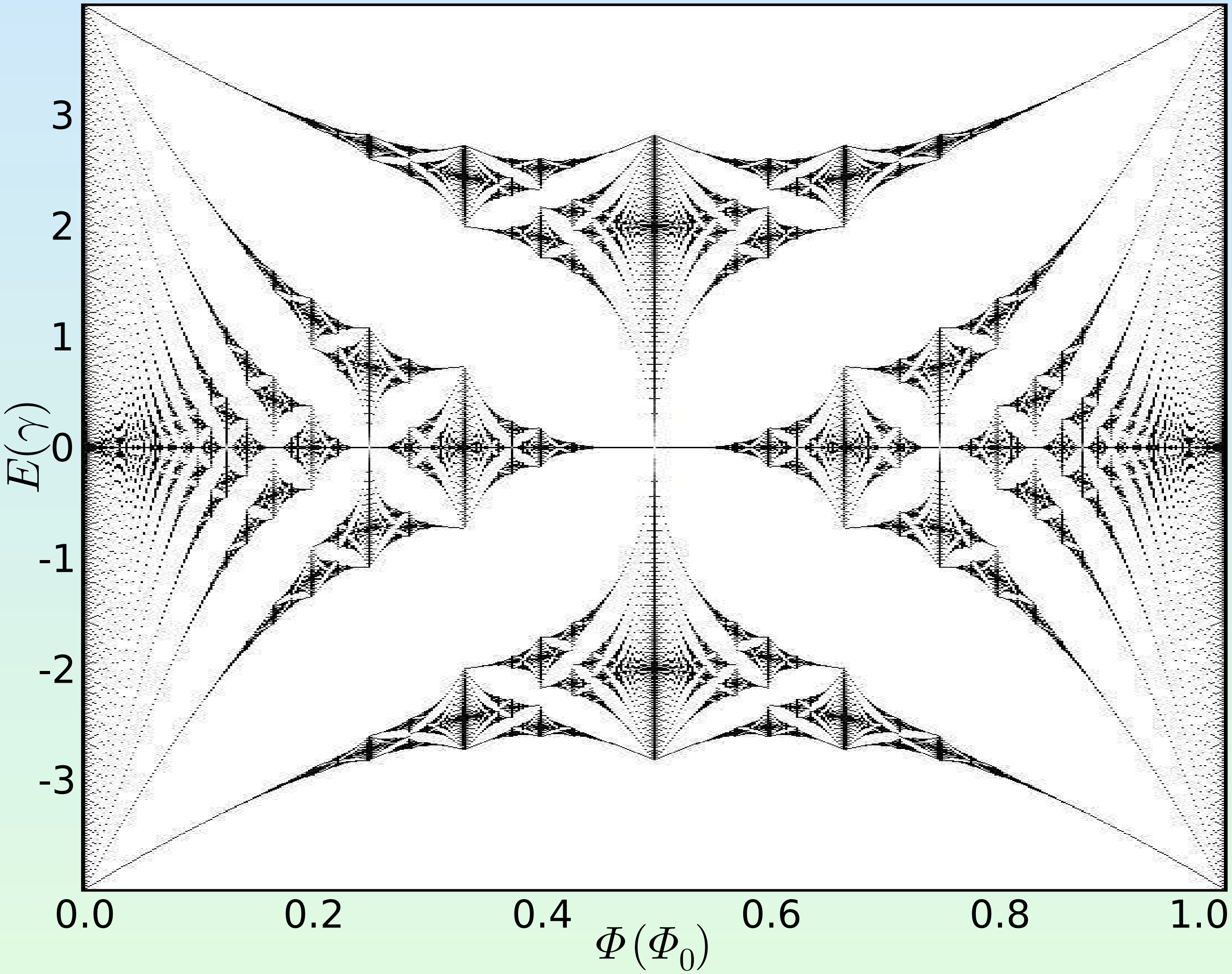


N. Nemec and G. Cuniberti, Phys. Rev. B **74**, 165411 (2006)
N. Nemec and G. Cuniberti, Phys. Rev. B **75** (Rapid Comm.), 201404 (2007)

The original Hofstadter butterfly

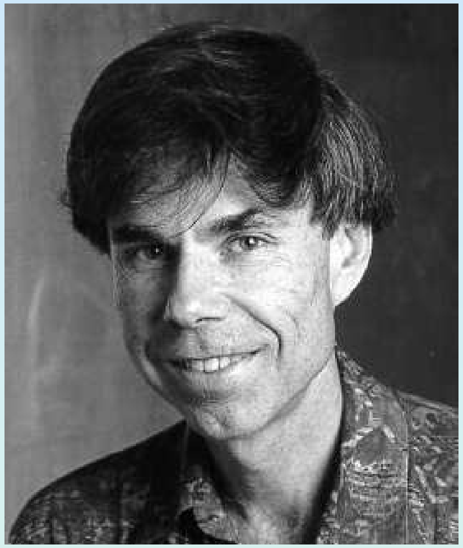






1975 in Regensburg...

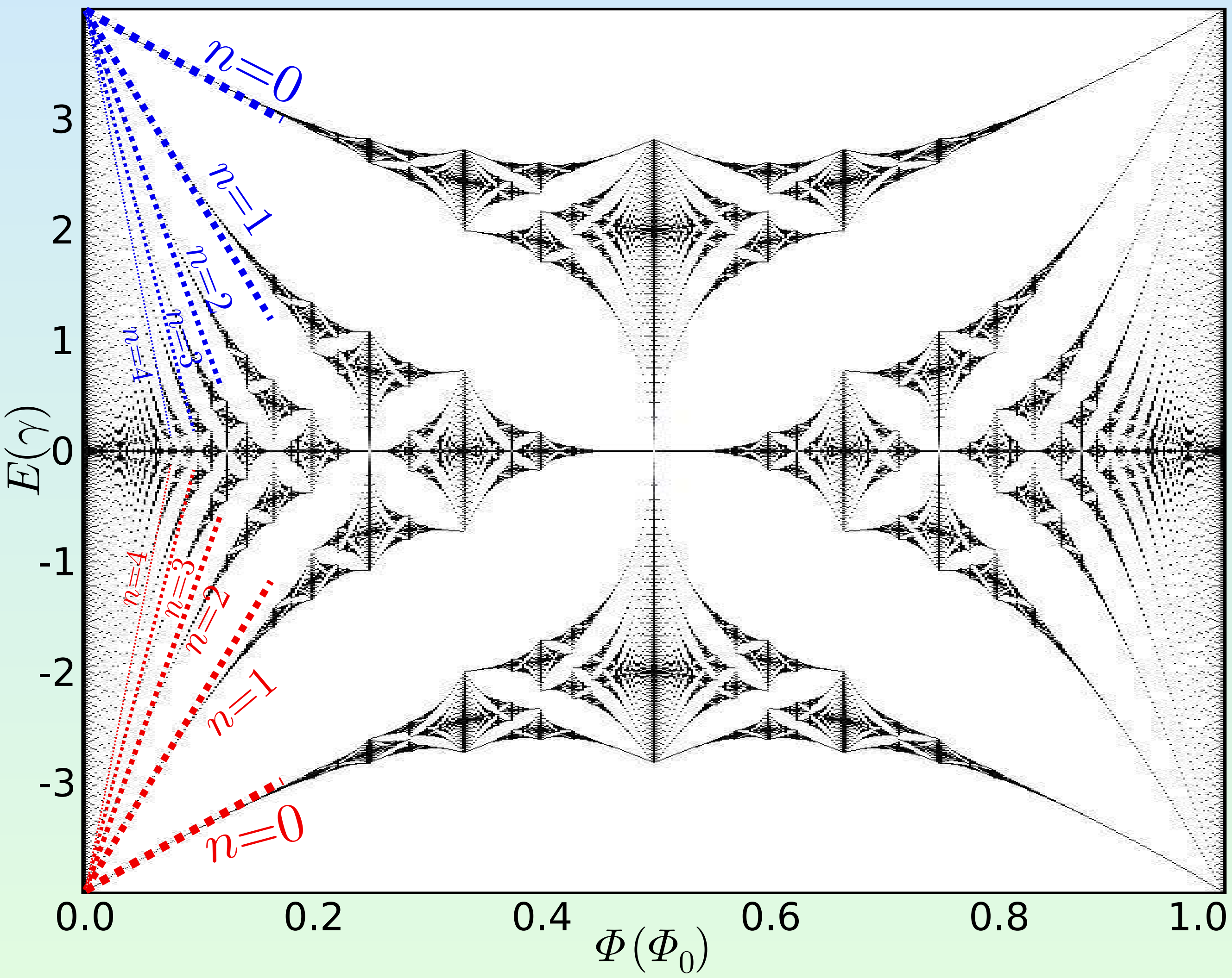
HP 9820A ("Rumpelstilzchen")
(8MHz/16bit CPU, 3432 byte RAM...)

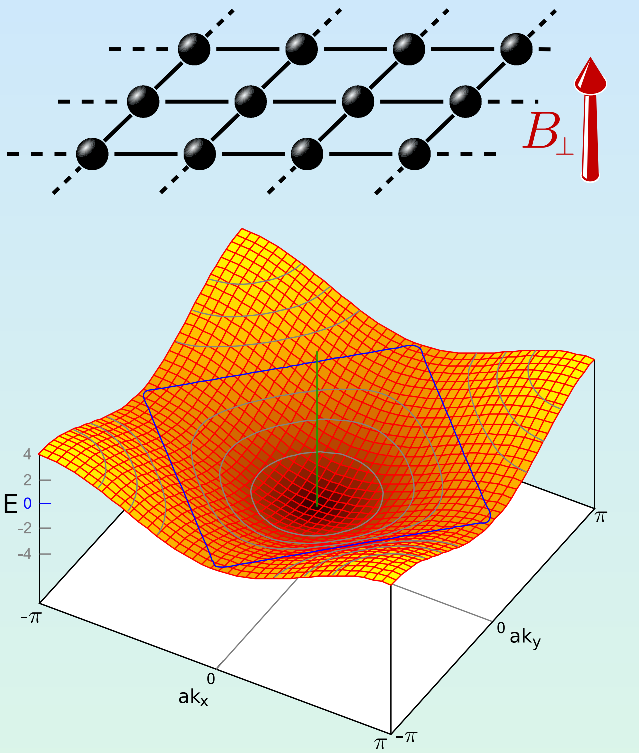


D. Hofstadter
G. Wannier
G. Obermair

Phys. Rev. B **14**, 2239 (1976)

Conventional Landau levels



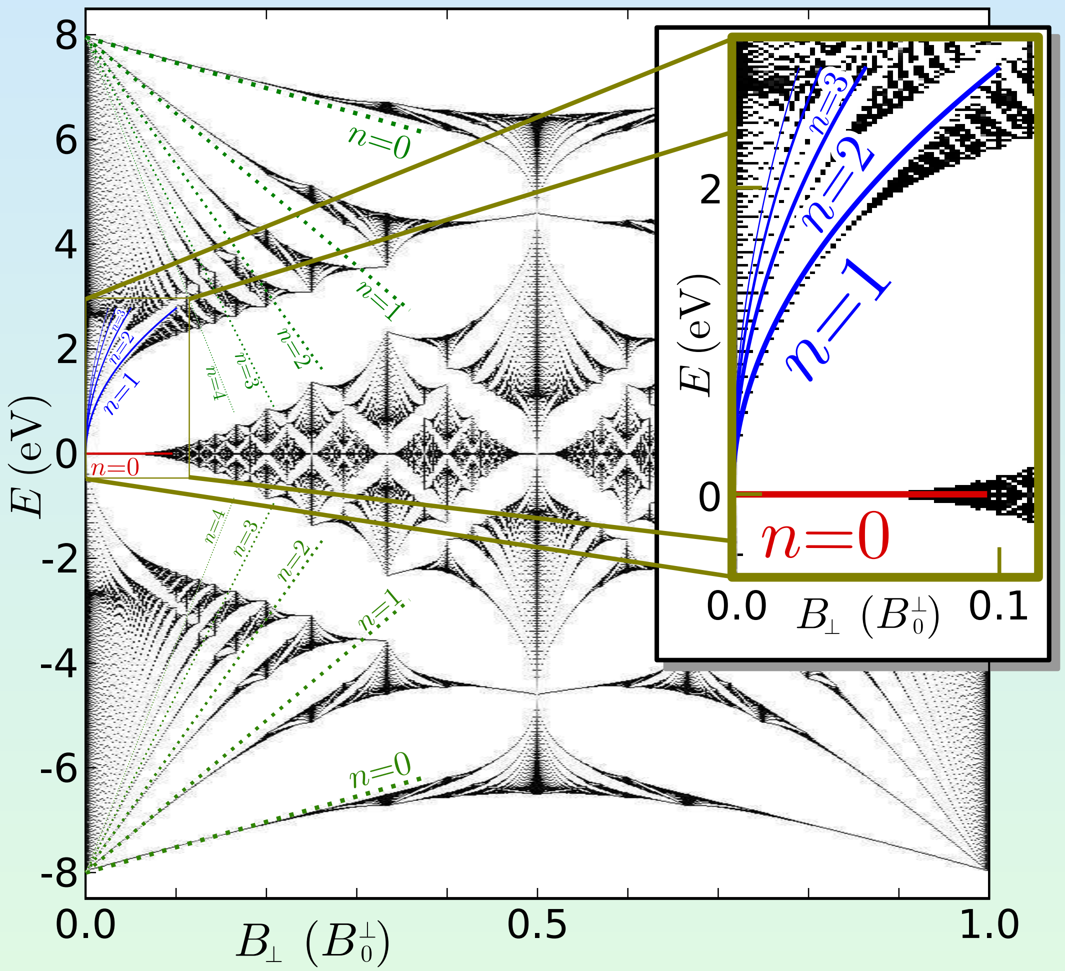


→ Landau levels at band band edges with effective mass $m^* = \frac{\hbar^2}{2\gamma a^2}$:

$$E = +4\gamma - \frac{\hbar e}{m^*} B_\perp \left(n + \frac{1}{2}\right)$$

$$E = -4\gamma + \frac{\hbar e}{m^*} B_\perp \left(n + \frac{1}{2}\right)$$

Hofstadter butterfly of graphene



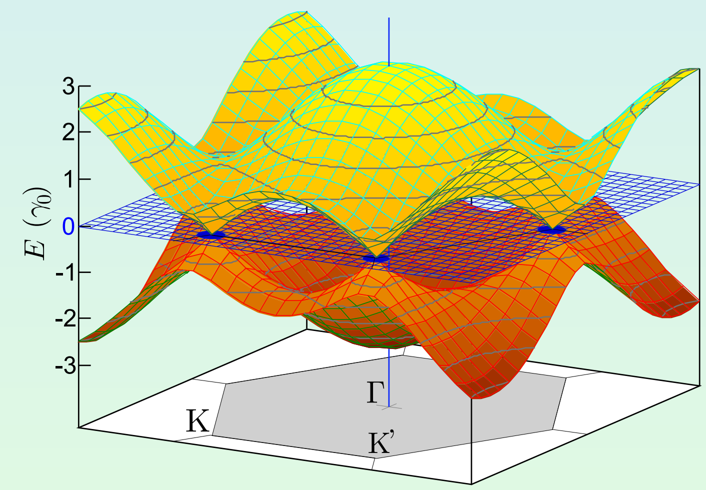
Rammal, J. Phys. (Paris) **46**, 1345 (1985)

(standard) Landau levels ("LL"):

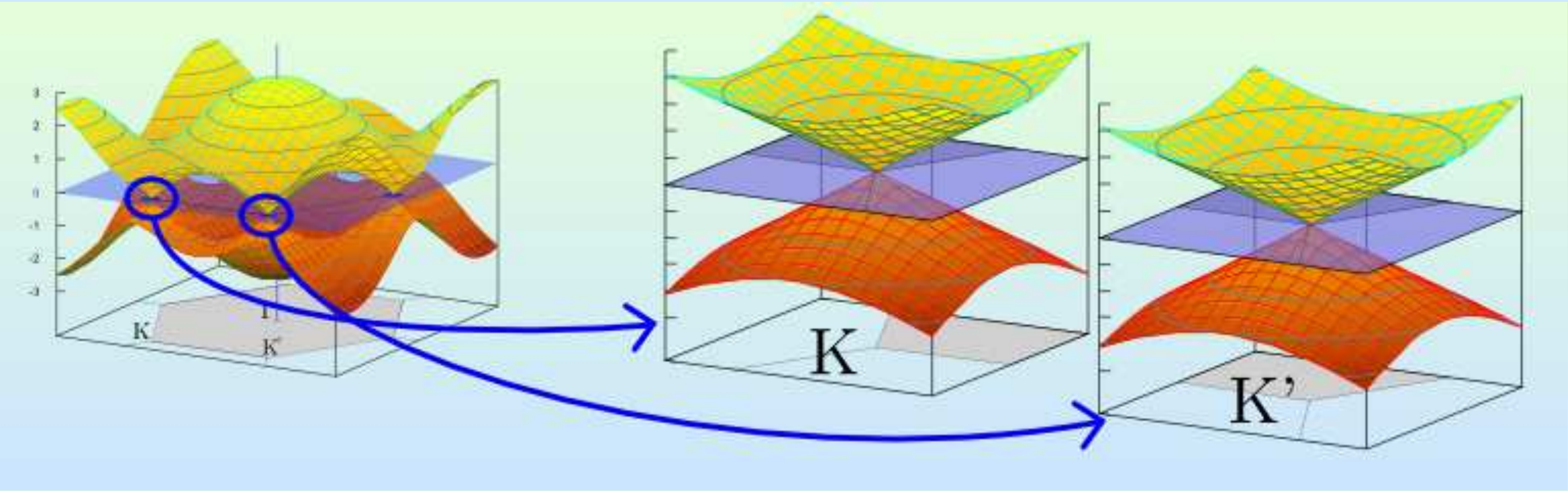
$$E - E_{\min} \propto \frac{\hbar e}{m^*} B \left(n + \frac{1}{2}\right)$$

$$E_{\max} - E \propto \frac{\hbar e}{m^*} B \left(n + \frac{1}{2}\right)$$

effective mass: $m^* = \frac{2\hbar^2}{\gamma a^2}$



Supersymmetric Dirac electrons



linearization around the K-points $\Rightarrow$ 2D-Dirac Hamiltonian:

$$H_D = \begin{pmatrix} 0 & Q \\ Q & 0 \end{pmatrix}, \text{ with } Q = v_\mathrm{F}(\sigma_x P_x + \sigma_y P_y), \boldsymbol{P} = -\mathrm{i}\hbar\boldsymbol{\partial} + e\boldsymbol{A}$$

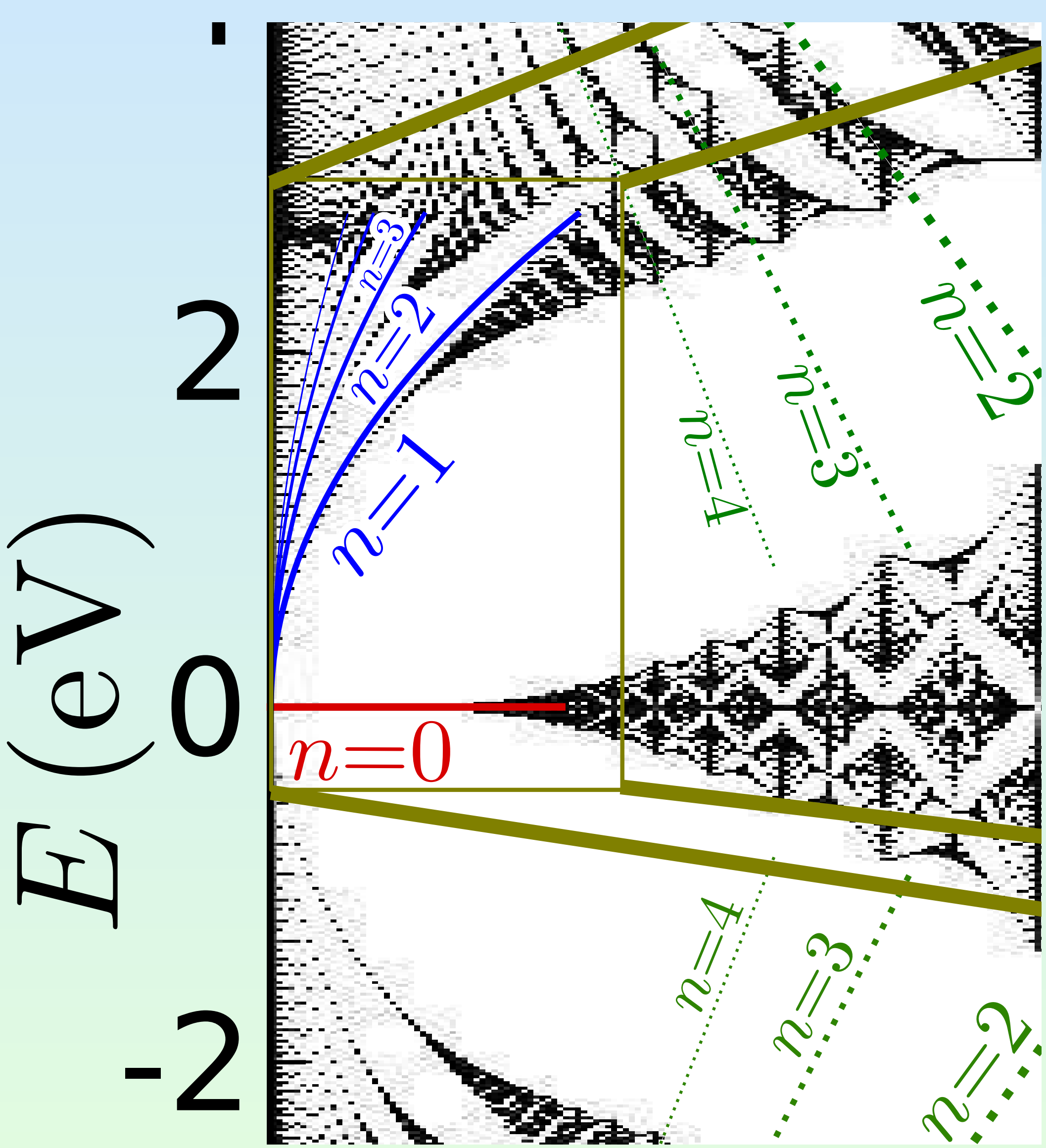
supersymmetric spectrum:

$E_0 = 0$ (4-fold degenerate, half-filled)

$E_n = \pm v_\mathrm{F}\sqrt{2e\hbar B n}$ (each 4-fold deg.)

see, e.g.: M. Ezawa, cond-mat/0606084

Hofstadter butterfly of graphene



(standard) Landau levels ("LL"):

$$E - E_{\mathrm{min}} \propto \frac{\hbar e}{m^*} B \left(n + \frac{1}{2}\right)$$

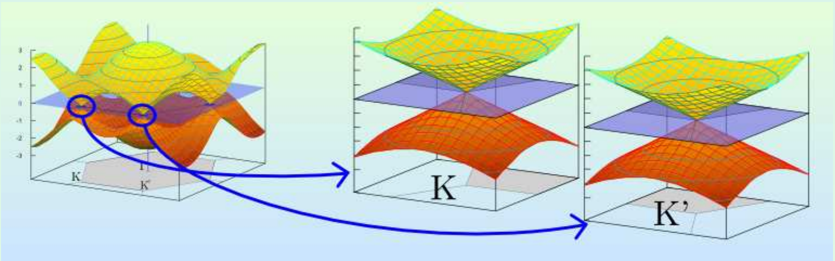
$$E_{\mathrm{max}} - E \propto \frac{\hbar e}{m^*} B \left(n + \frac{1}{2}\right)$$

relativistic LL: $(v_{\mathrm{F}} = \sqrt{3}\gamma a/2\hbar)$

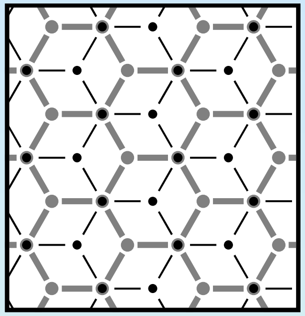
$$E - E_{\mathrm{F}} = \pm v_{\mathrm{F}} \sqrt{2e\hbar\, B\, n}$$

supersymmetric LL ("SuSyLL"):

$$E = E_{\mathrm{F}}$$



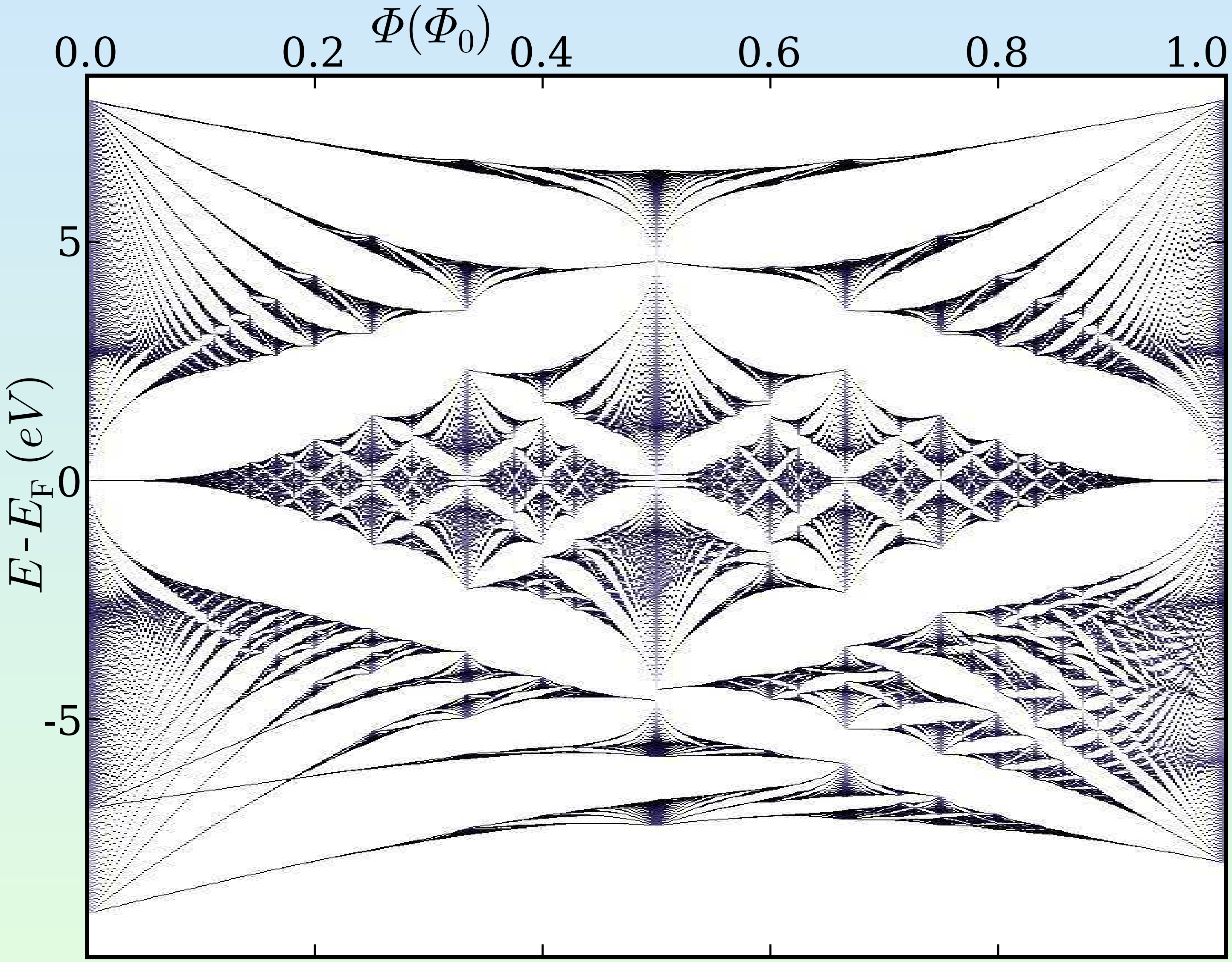
Hofstadter butterfly of bilayer graphene



Bernal-stacking

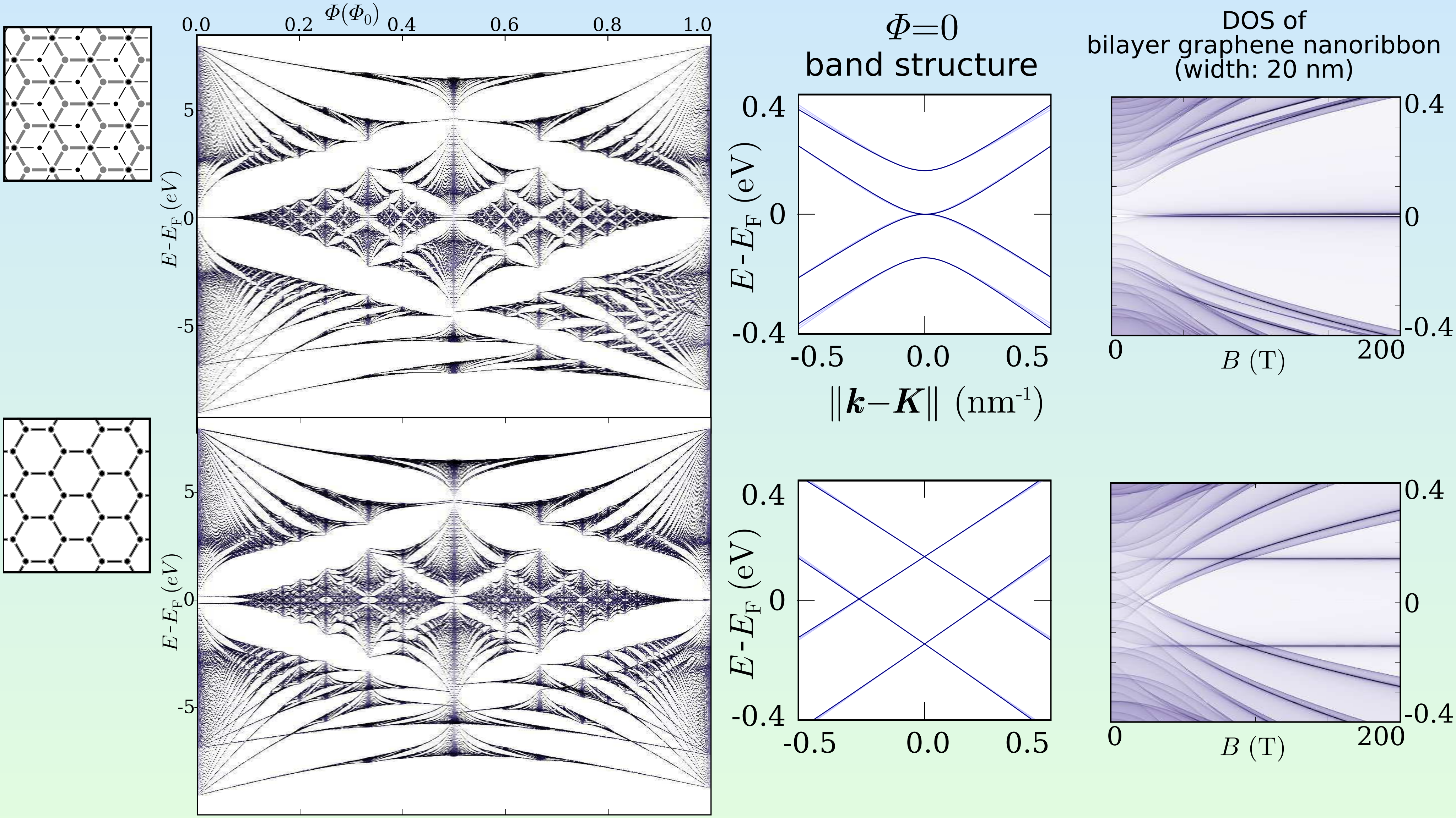
broken symmetries:
→ Φ_0-periodicity
→ electron-hole

SuSyLL protected by symmetry



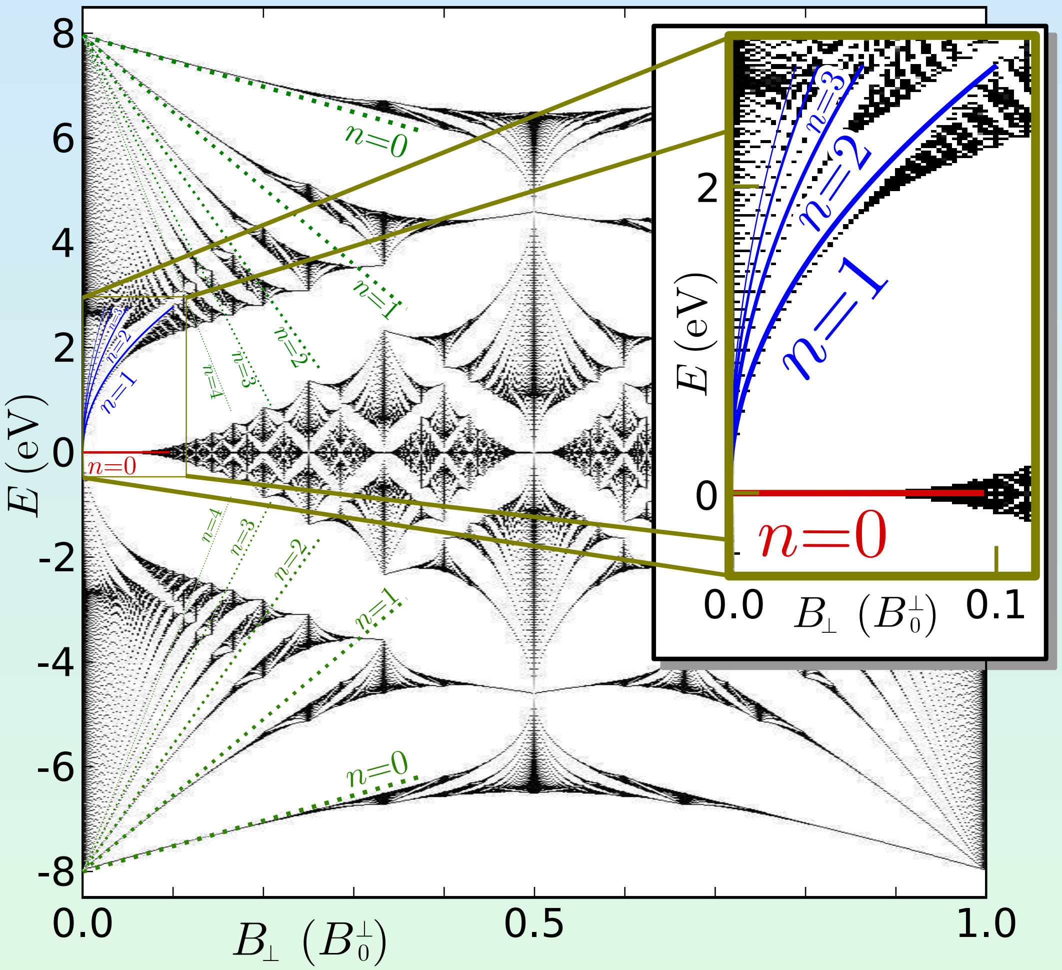
N. Nemec and G. Cuniberti, Phys. Rev. B **75** (Rapid Comm.), 201404 (2007)

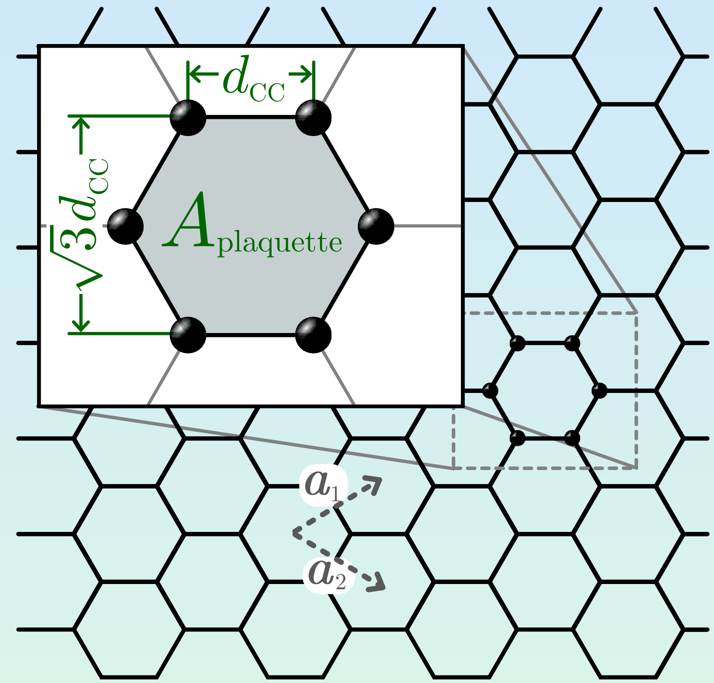
Shifted bilayer graphene



N. Nemec and G. Cuniberti, Phys. Rev. B **75** (Rapid Comm.), 201404 (2007)

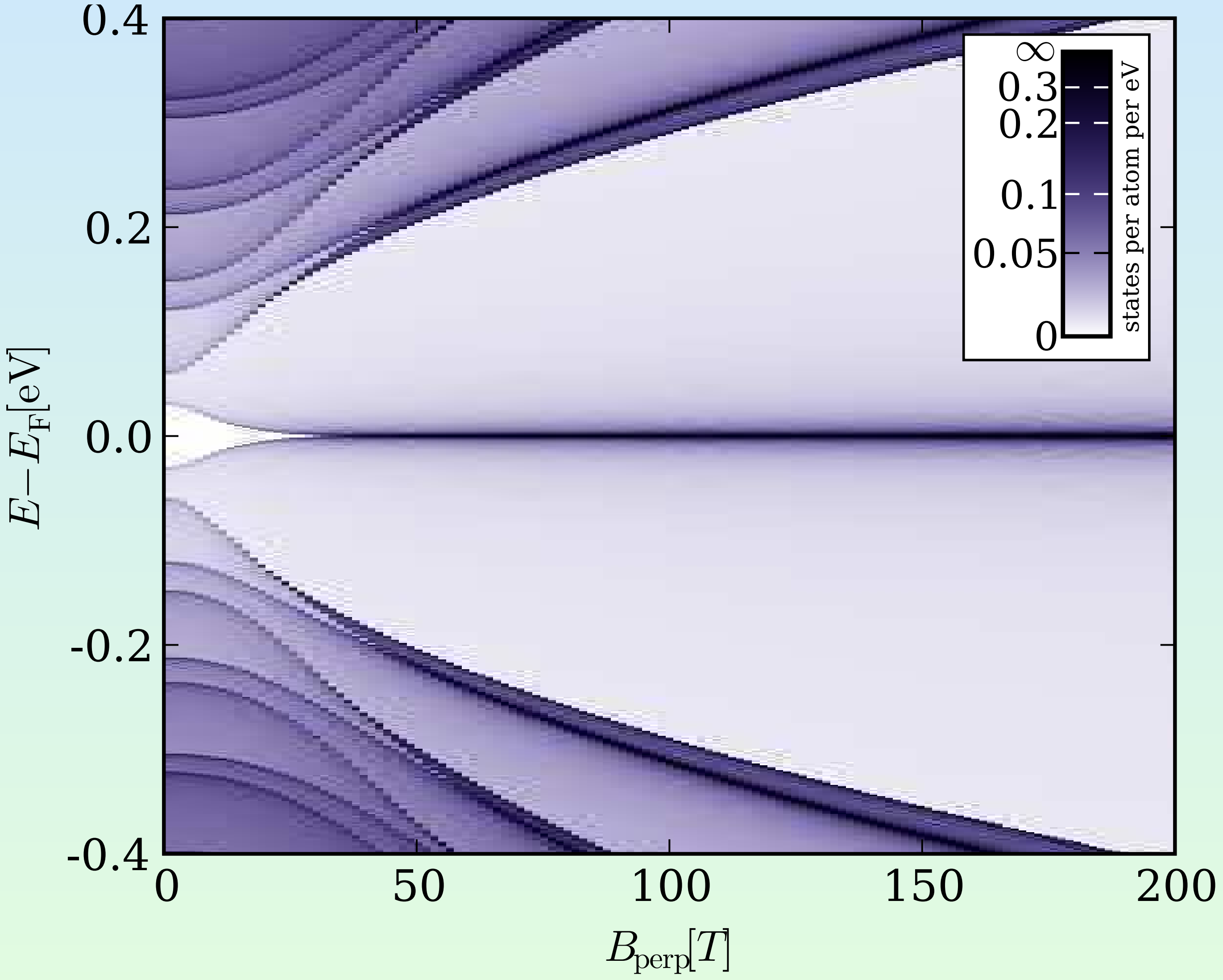
Hofstadter butterfly of graphene

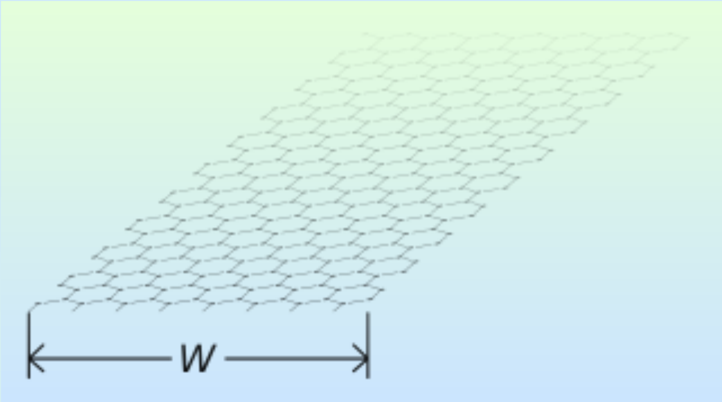




$$B_0^\perp = \Phi_0/A_{\text{plaquette}} \approx 79\,\text{kT (!!)}$$

Graphene nanoribbon

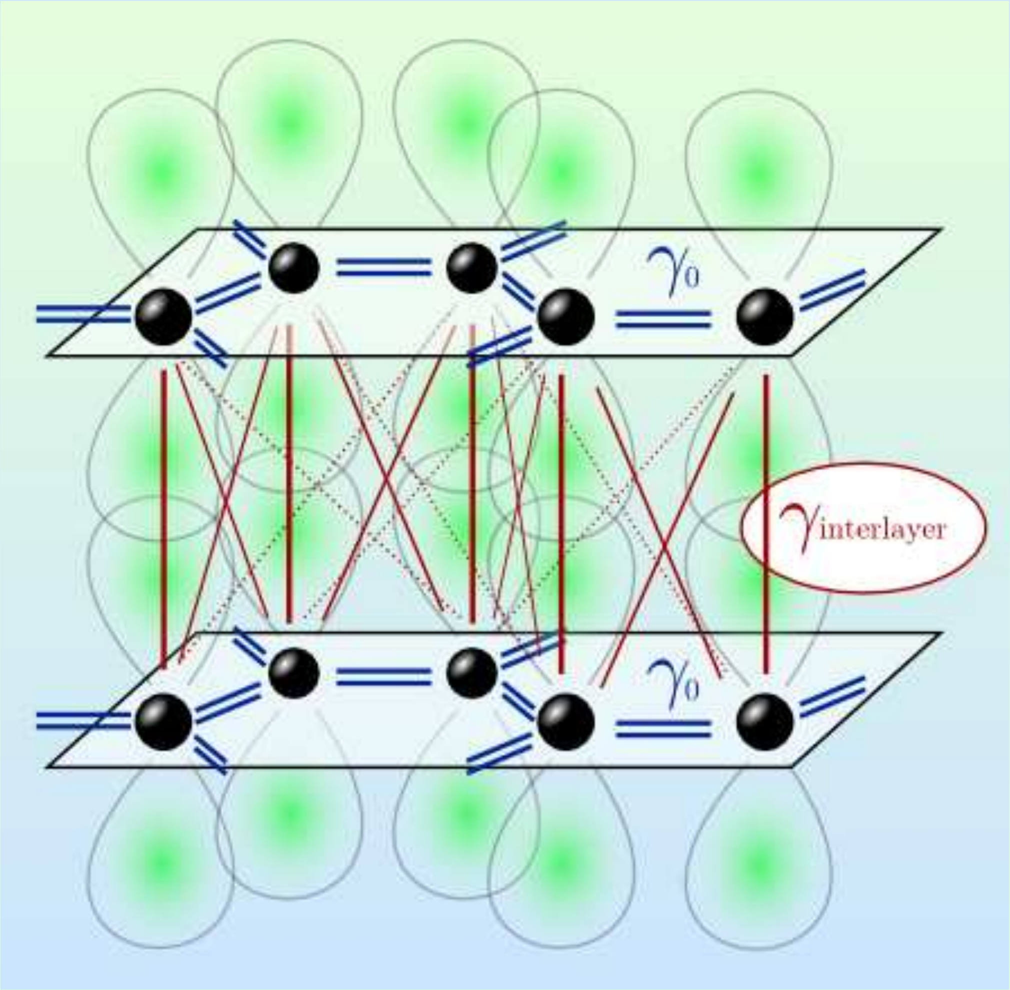




(80,0)-armchair ribbon
$W = 20\text{nm}$

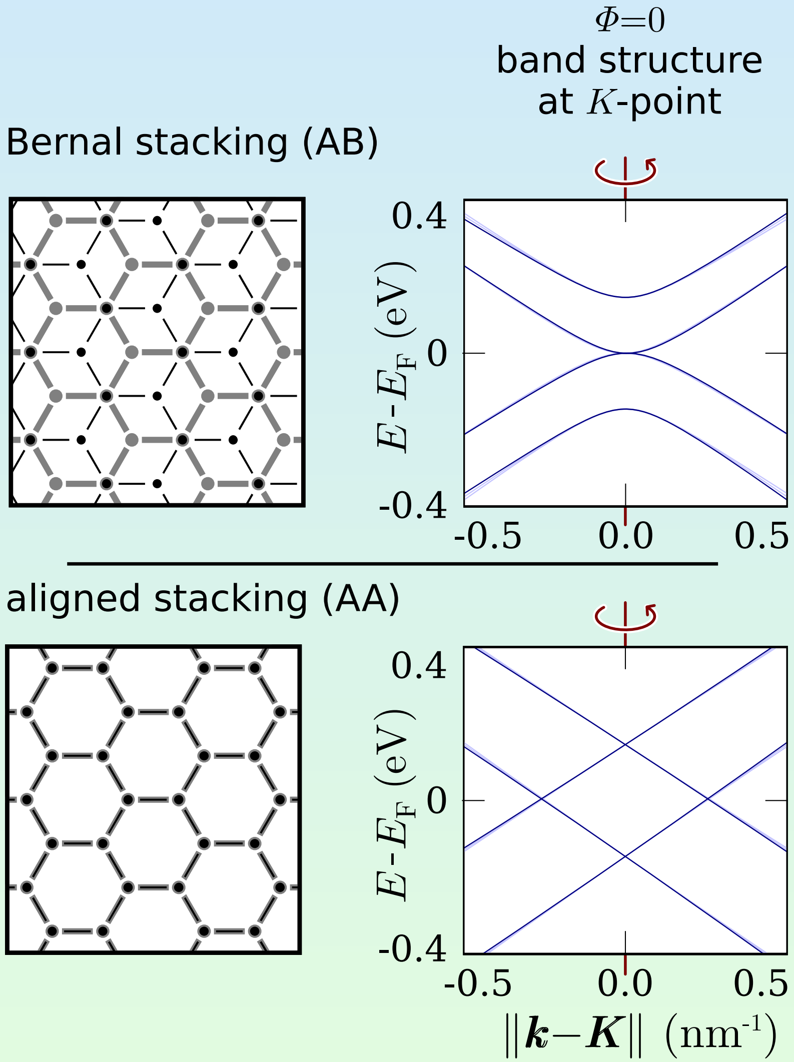
see also:
Peres *et al.*
PRB **73**, 241403 (2006)

Bilayer graphene



$$\gamma_{ij}^{\text{interlayer}} = \frac{\gamma_0}{8}\exp\left(\frac{d_{ij}-d_0}{\delta}\right)$$

$$\beta = \frac{\gamma_0}{8},\ d_0 = 3.34\,\text{Å},\ \delta = 0.45\,\text{Å}$$

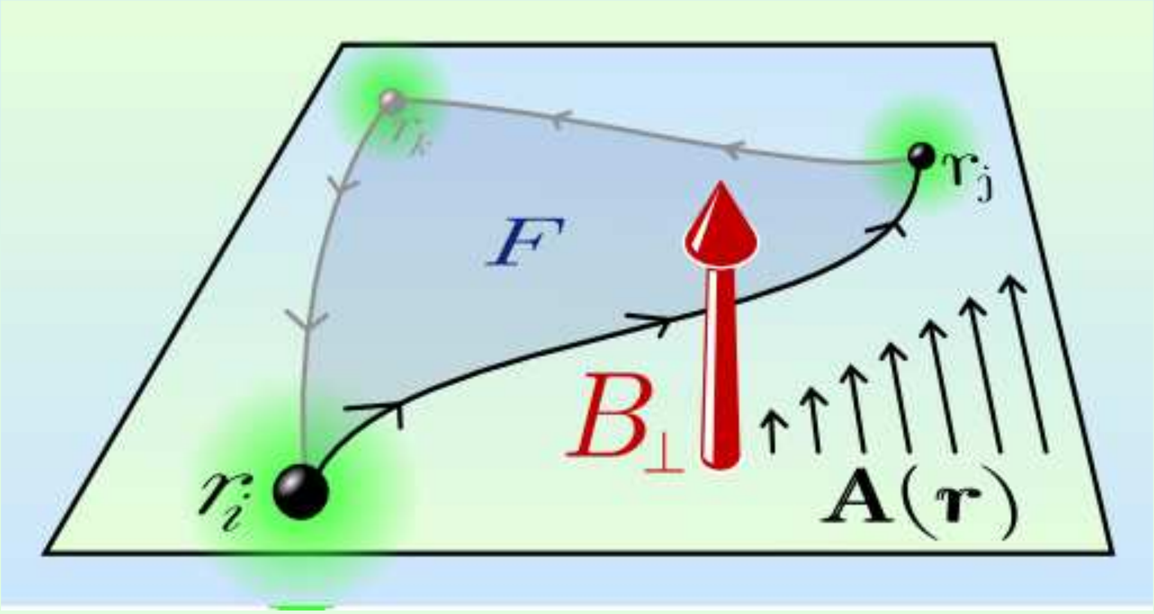


Theory: Peierls substitution

$$\gamma_{ij}(\boldsymbol{B}) \;=\; \gamma_{ij}^{0}\exp\left(\frac{2\pi\mathrm{i}}{\Phi_0}\int_{\boldsymbol{r}_i}^{\boldsymbol{r}_j}\mathrm{d}\boldsymbol{r}\cdot\boldsymbol{A}(\boldsymbol{r})\right)$$

magnetic field: $\boldsymbol{B}(\boldsymbol{r}) \;=\; \nabla\times\boldsymbol{A}(\boldsymbol{r})$
gauge field: $\boldsymbol{A}(\boldsymbol{r})$ (*"vector potential"*)
flux quantum: $\Phi_0 = h/e$

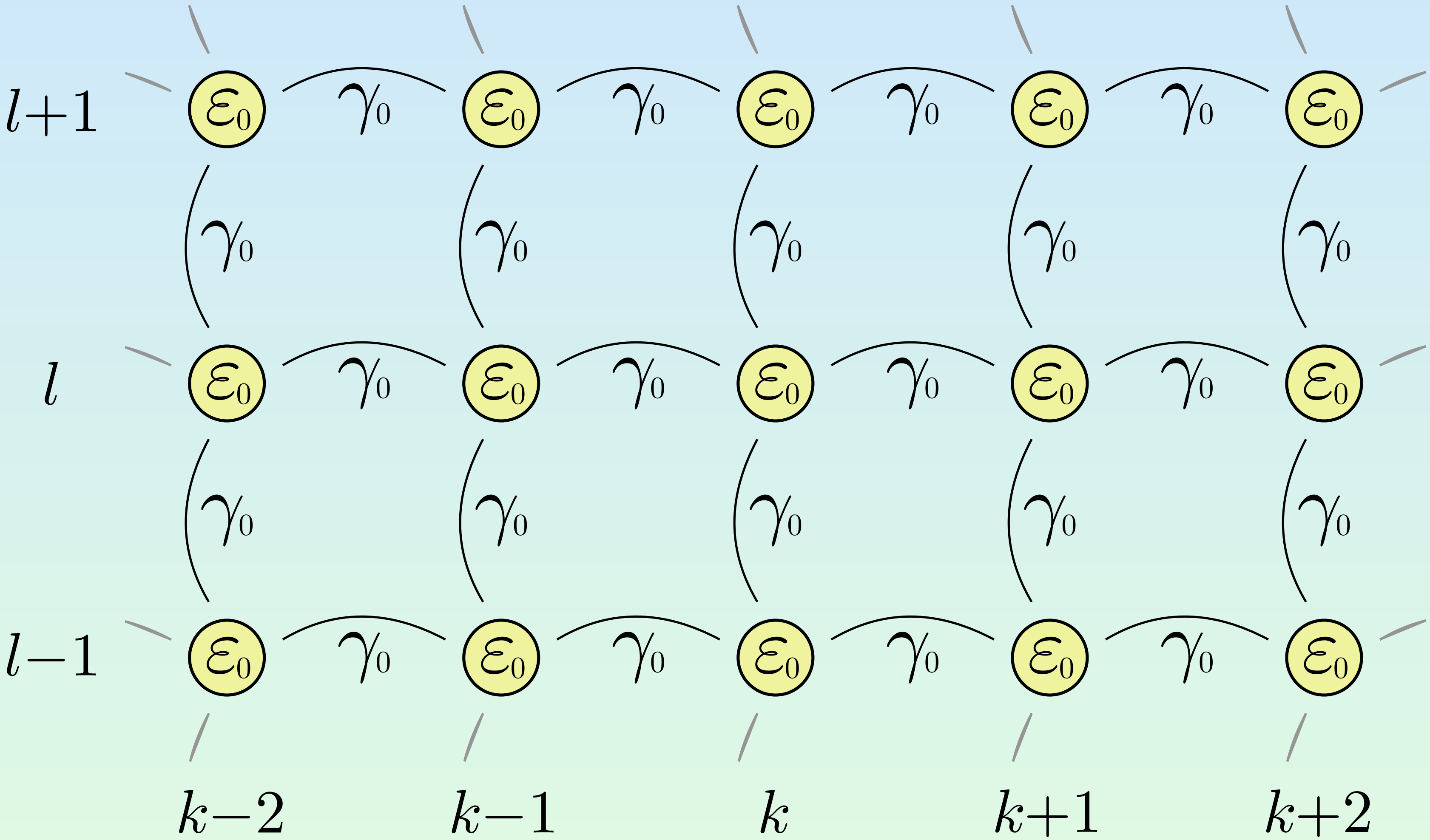
Peierls, Z. Phys. **80**, 763 (1933)



Phase of circular path given by enclosed area F:

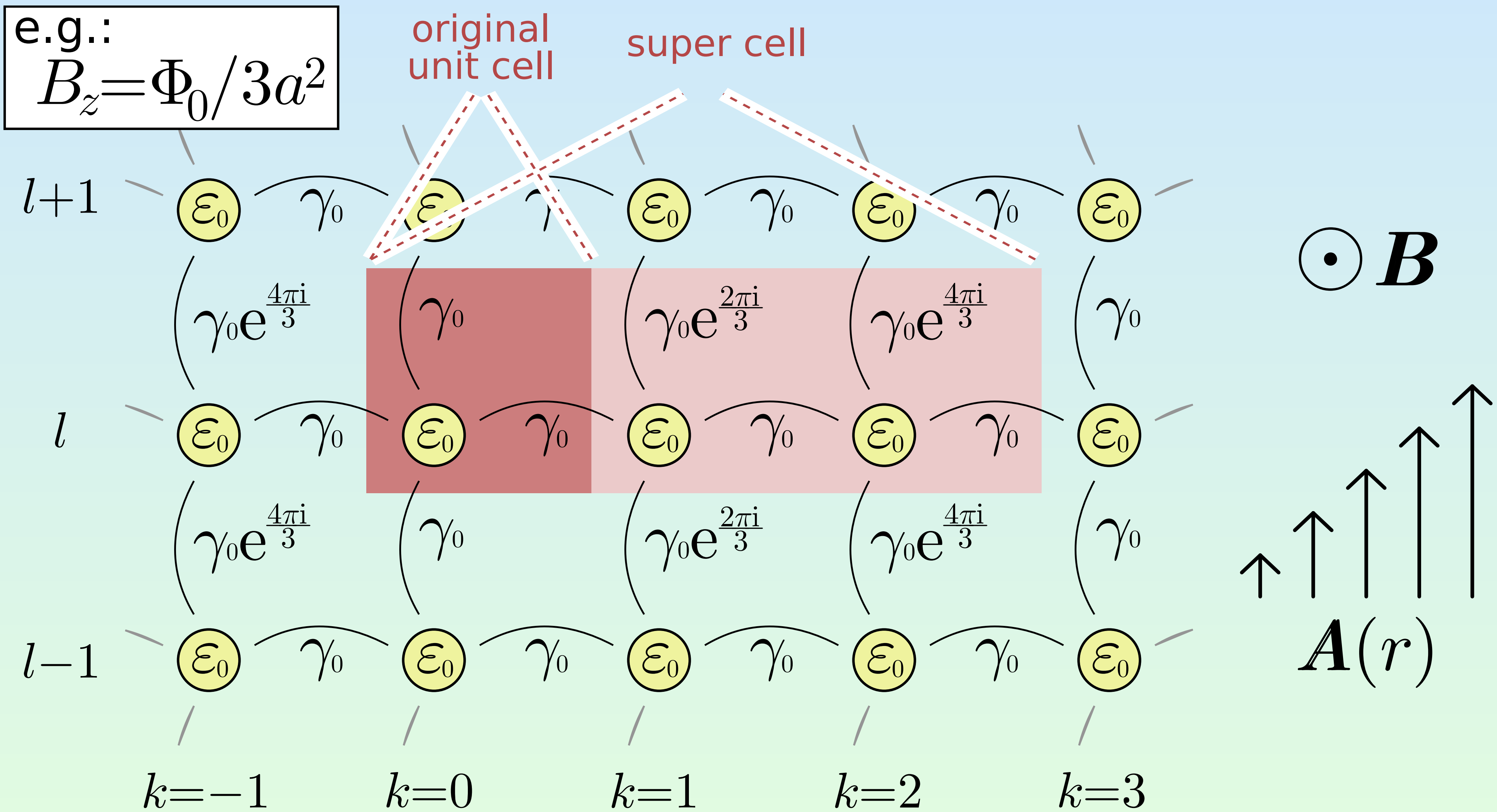
$$\varphi_{i\to j\to k\to i} = \exp\left(2\pi\mathrm{i}\frac{FB_\perp}{\Phi_0}\right)$$

The original Hofstadter butterfly (I)

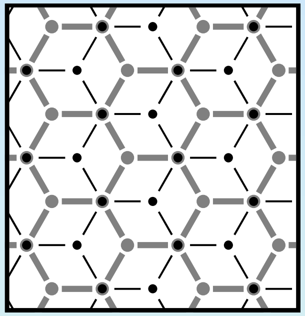


(lattice constant a)

The original Hofstadter butterfly (II)

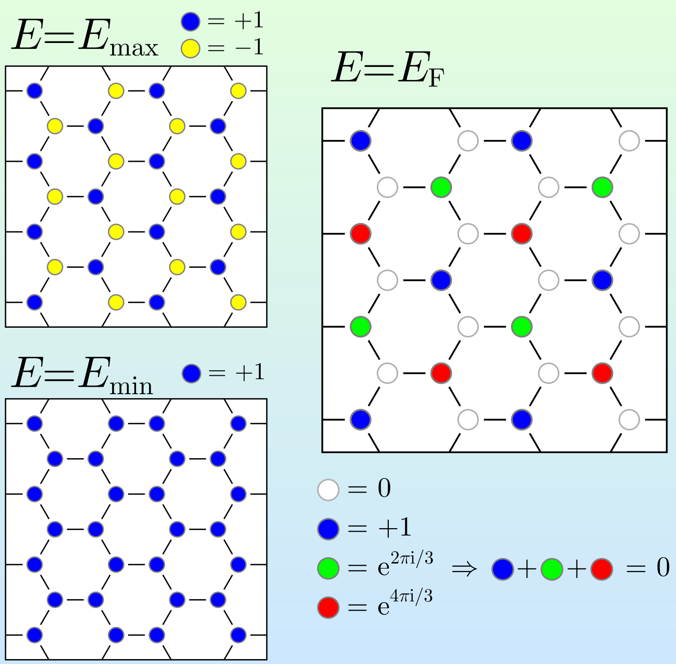


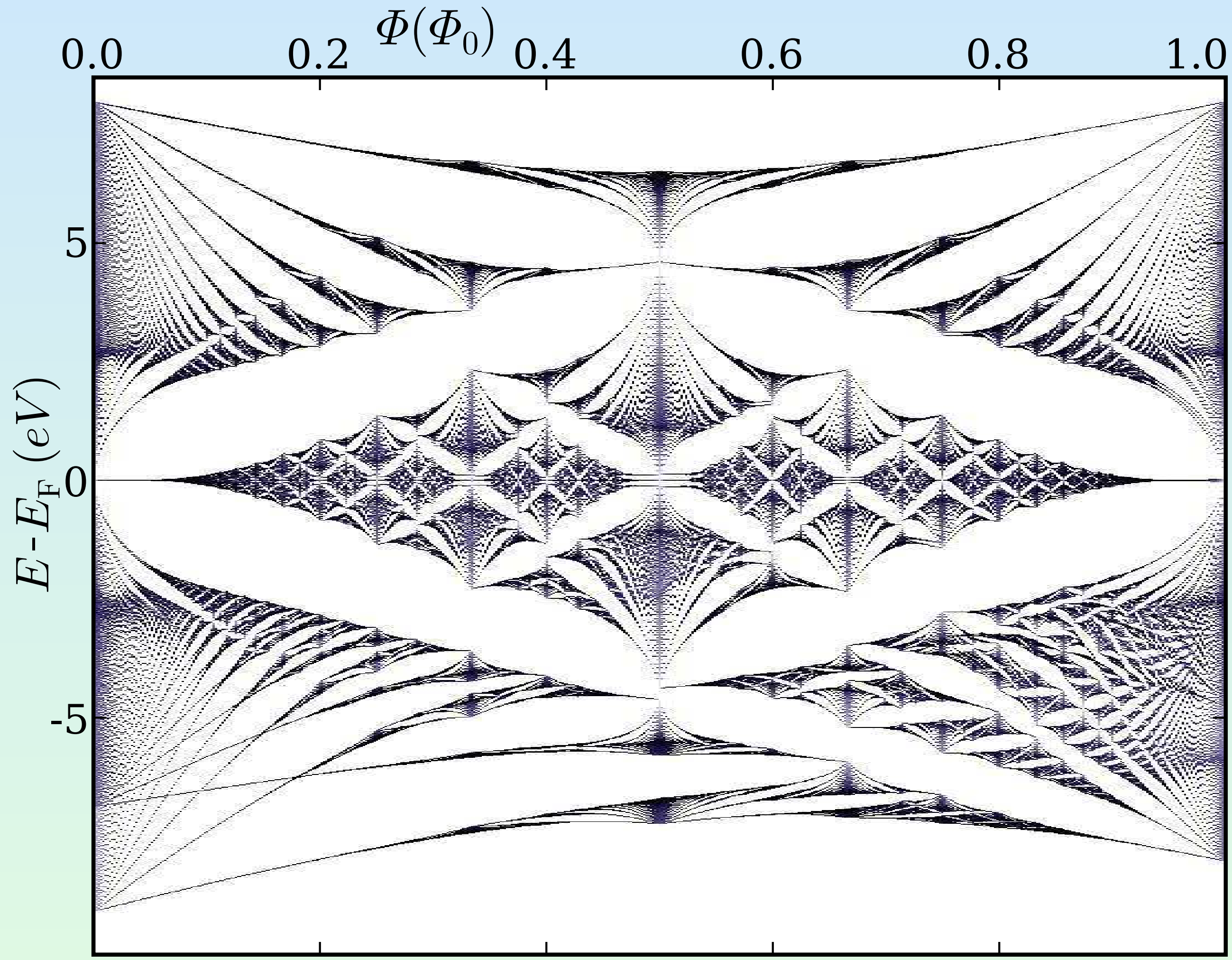
e-h asymmetry



Bernal-stacking

Wavefcns in graphene **monolayer**

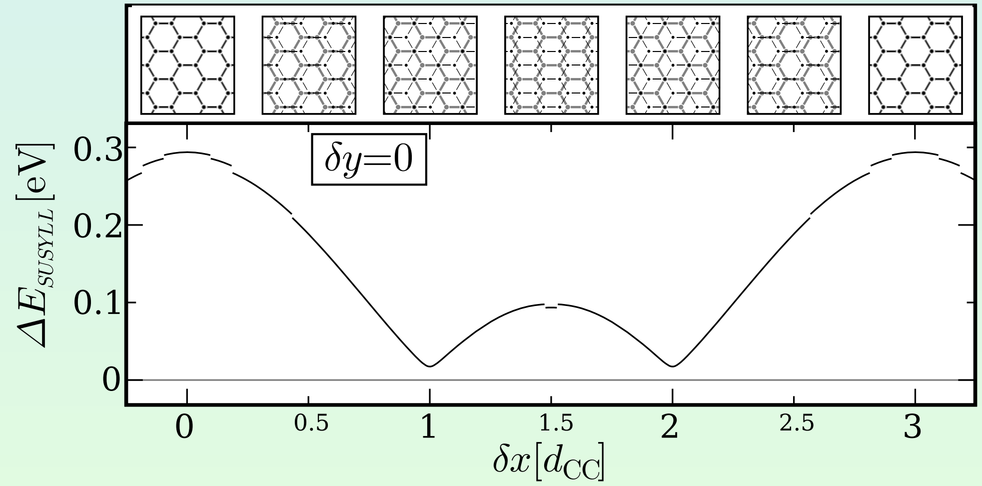




Split of the SUSYLL

Bernal stacking:
SuSyLL protected against split

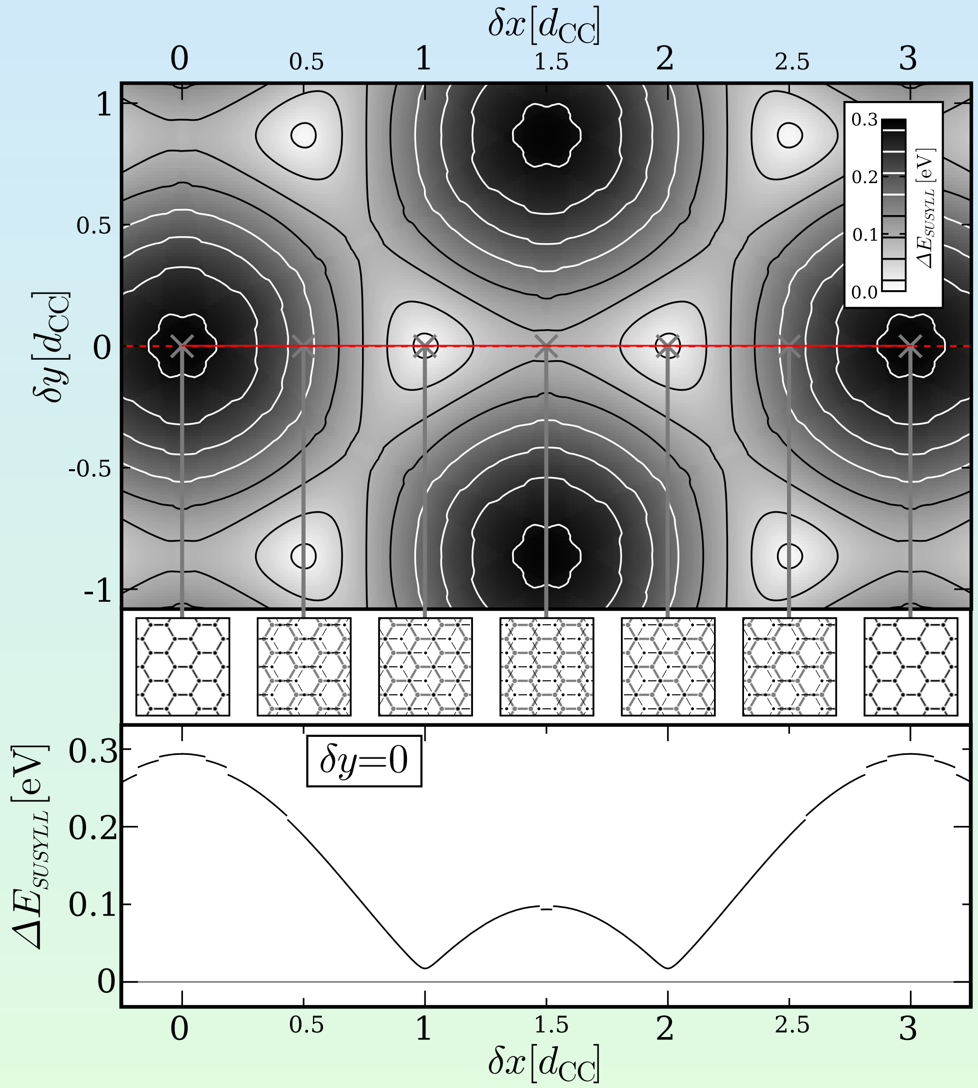
Shifted configurations:
SuSyLL split by varying amounts



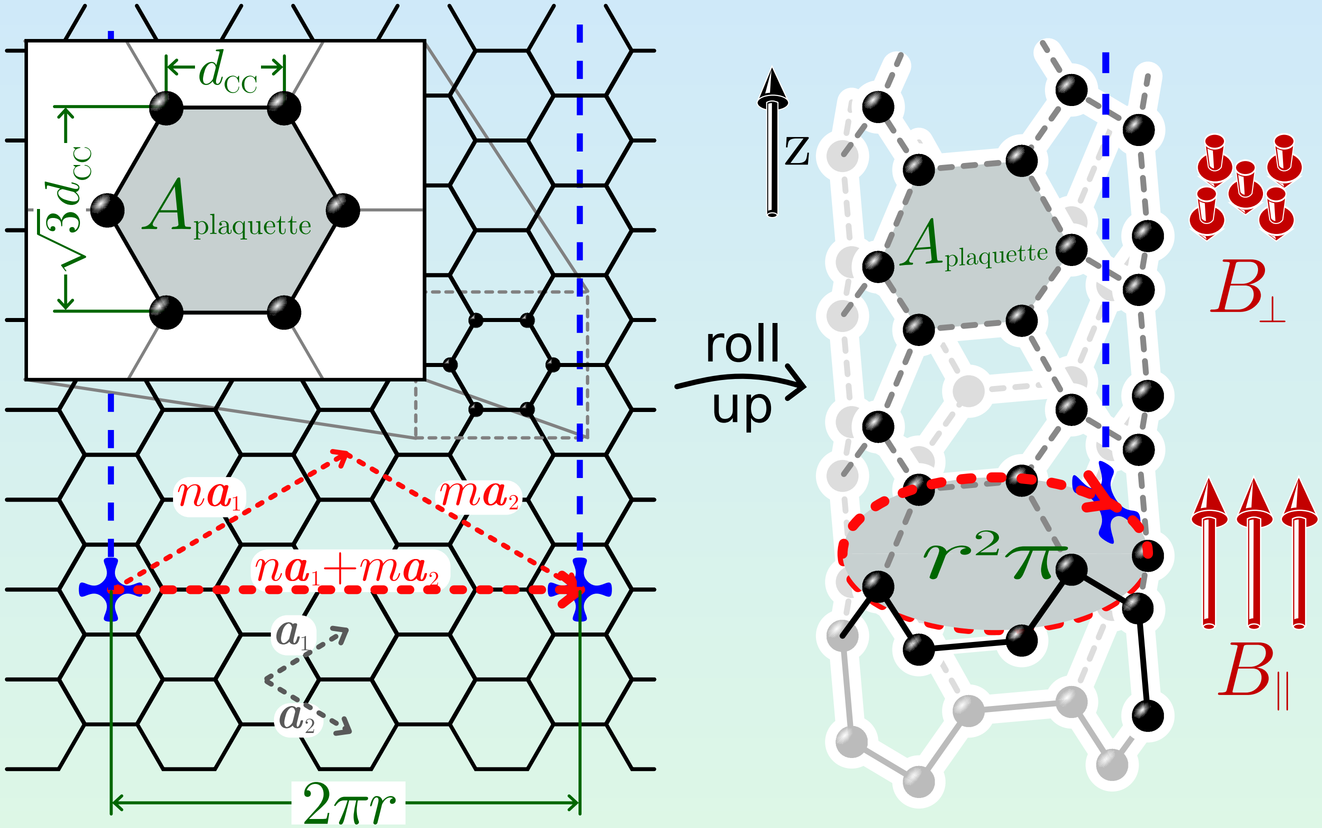
Split of the SUSYLL

Bernal stacking:
SuSyLL protected against split

Shifted configurations:
SuSyLL split by varying amounts



Magnetic field scales in CNT



$$B_0^\perp = \Phi_0/A_{\text{plaquette}} \approx 79\,\text{kT}$$

$$B_0^\parallel = \Phi_0/r^2\pi = \frac{2\sqrt{3}\pi}{m^2+n^2+m\,n}B_0^\perp$$

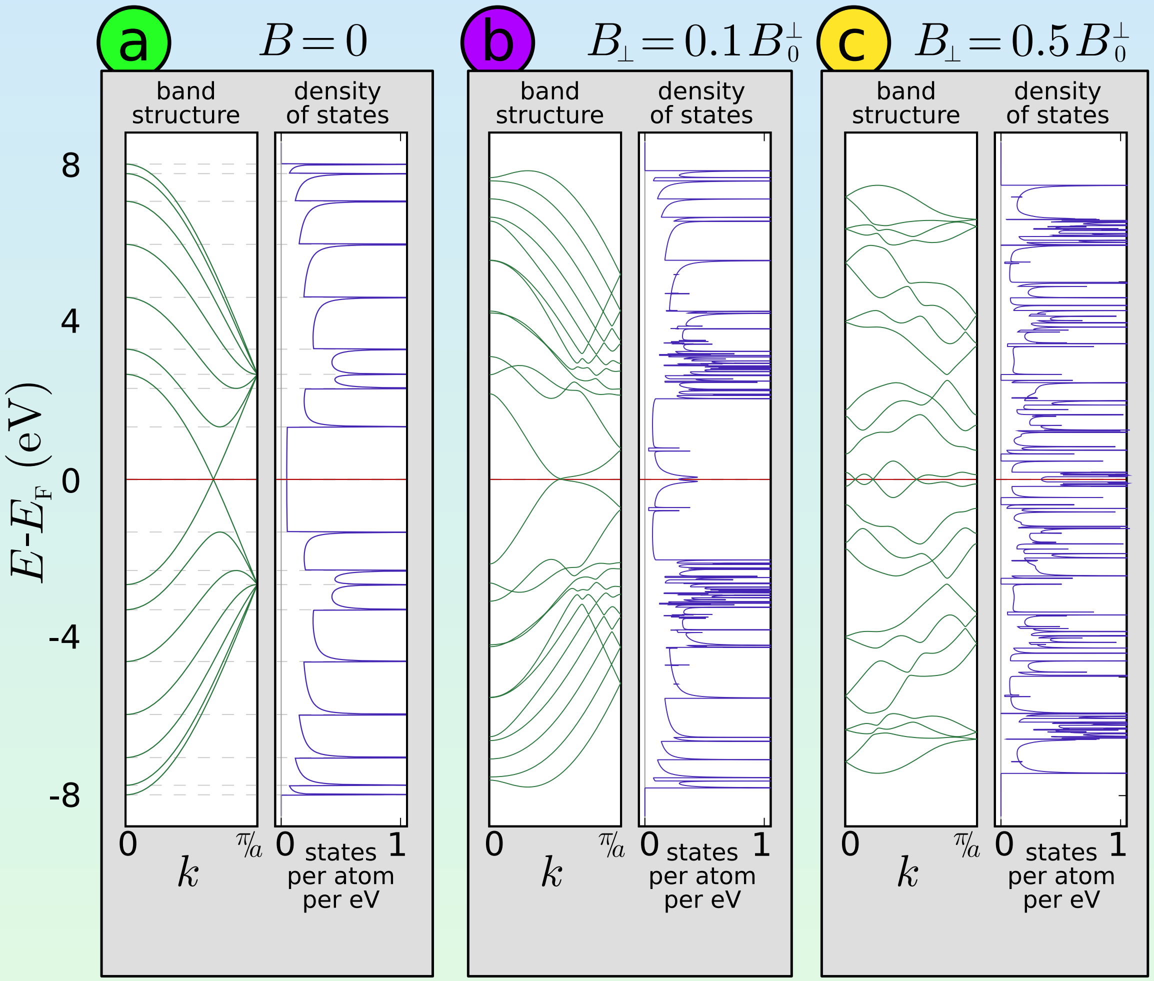
(6,6) CNT:

$$B_0^\parallel \approx 7.9\,\text{kT}$$

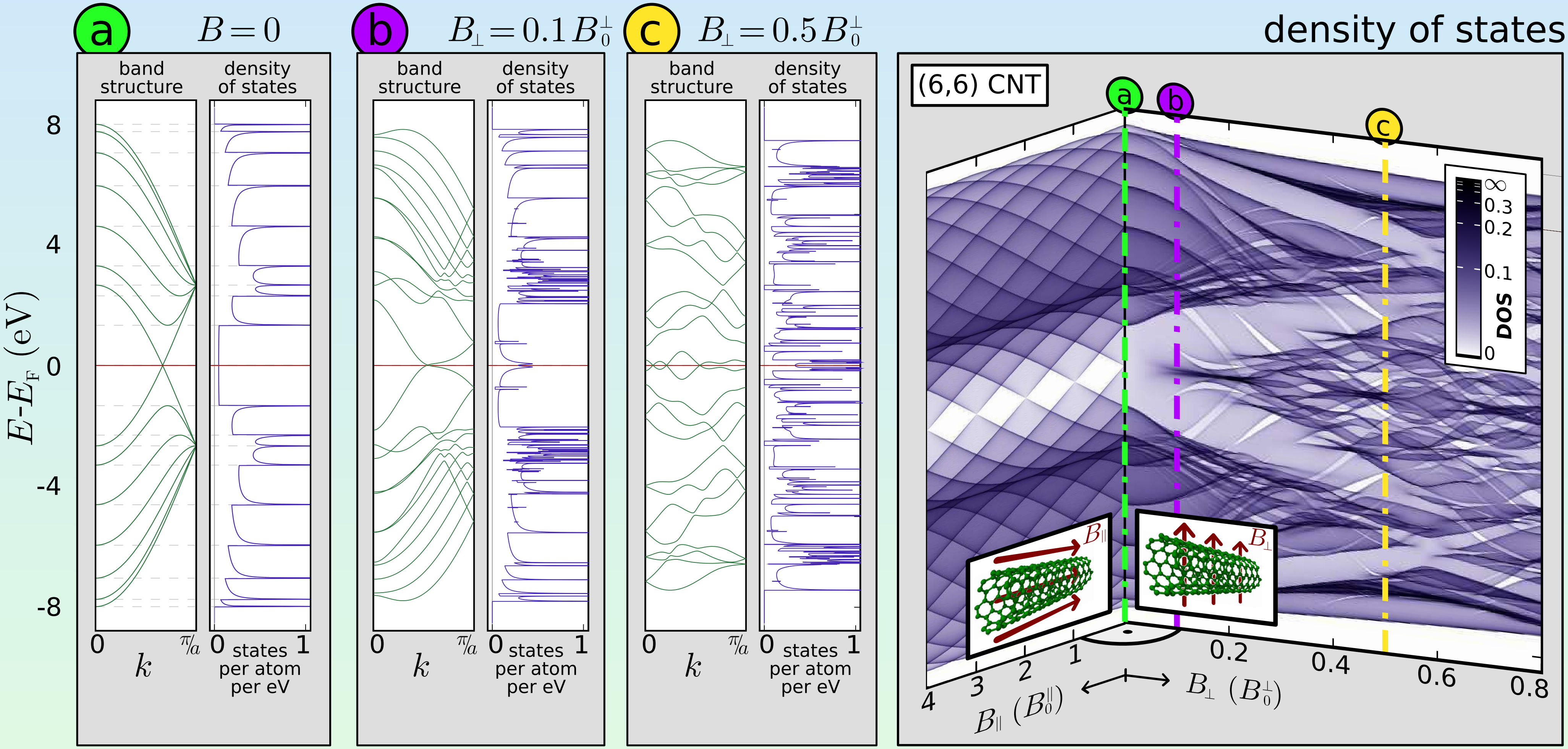
(100,100) CNT:

$$B_0^\parallel \approx 28\,T$$

Evolution of band structure and DOS

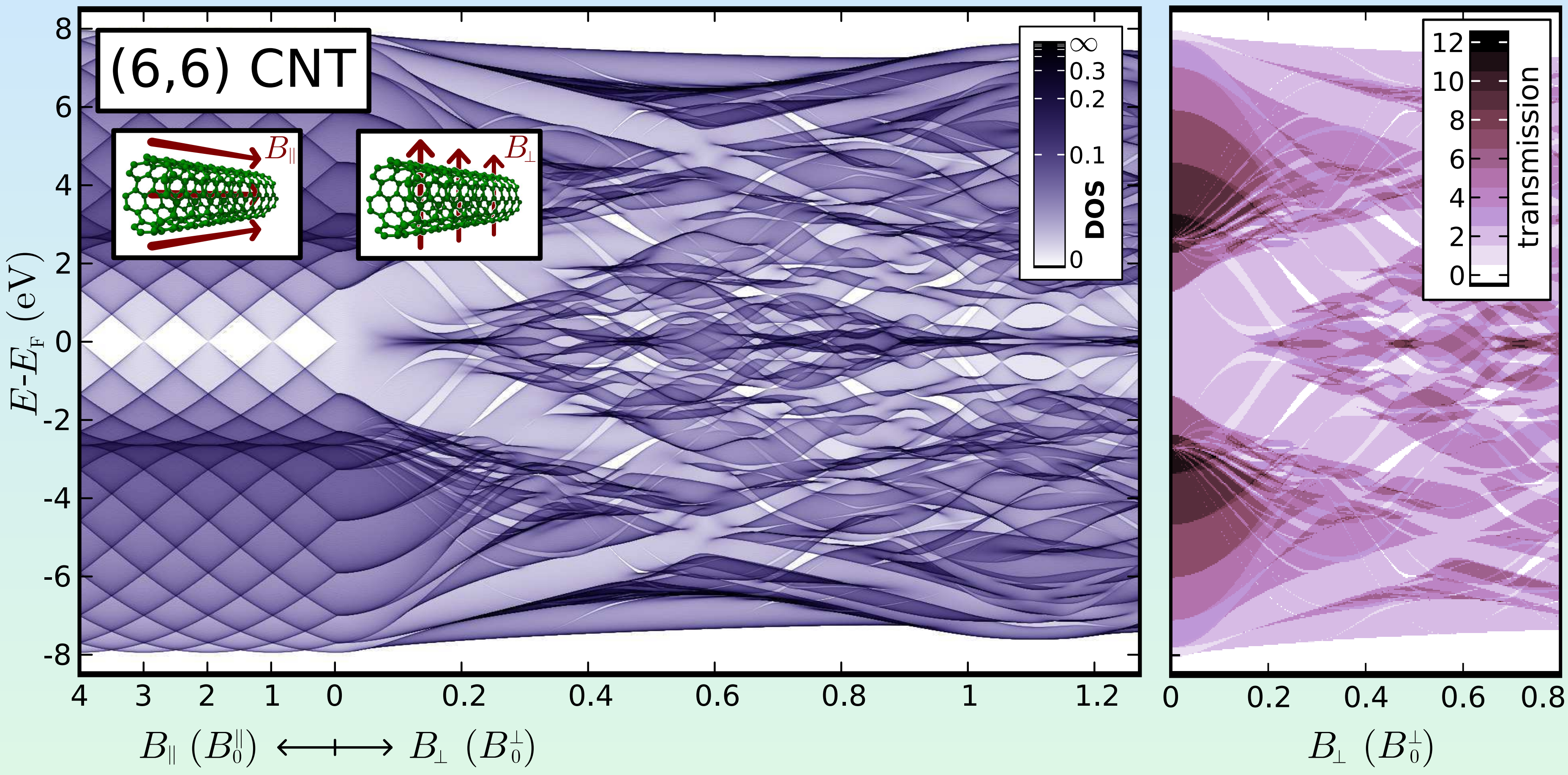


Visualization as butterfly plot



*for A-B-oscillations in parallel fields, see also: Bachtold et al., Nature **397**, 673 (1998)*

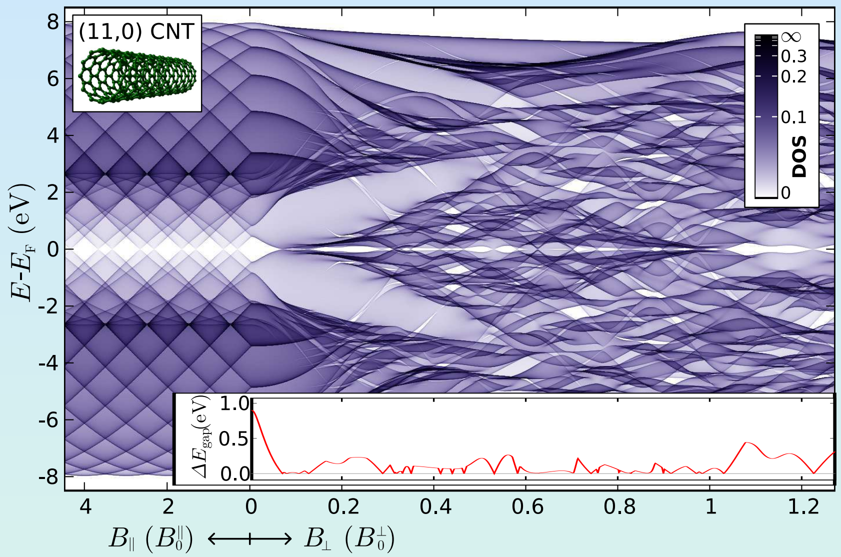
Armchair CNT



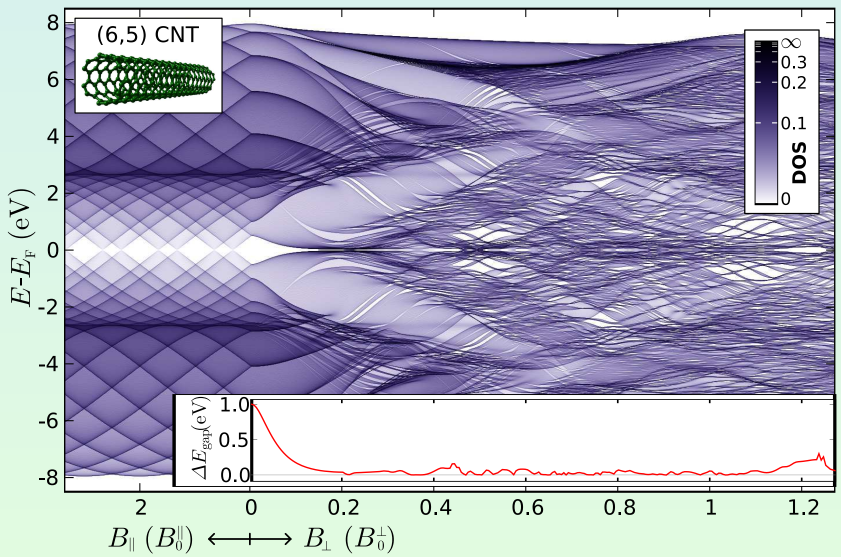
- $B_\perp$: states at Fermi level protected by supersymmetry

*see also: Lee and Novikov, Phys. Rev. B **68**, 155402 (2003)*

Semiconducting CNT



(11,0) CNT:
gap size oscillates nonperiodically

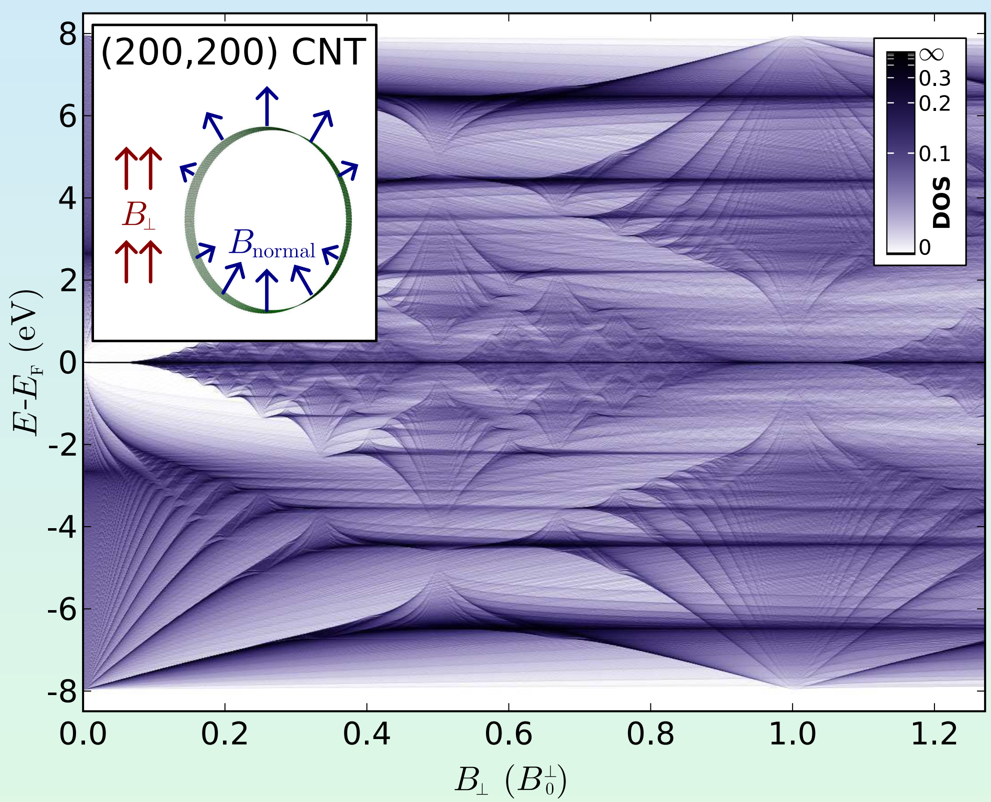


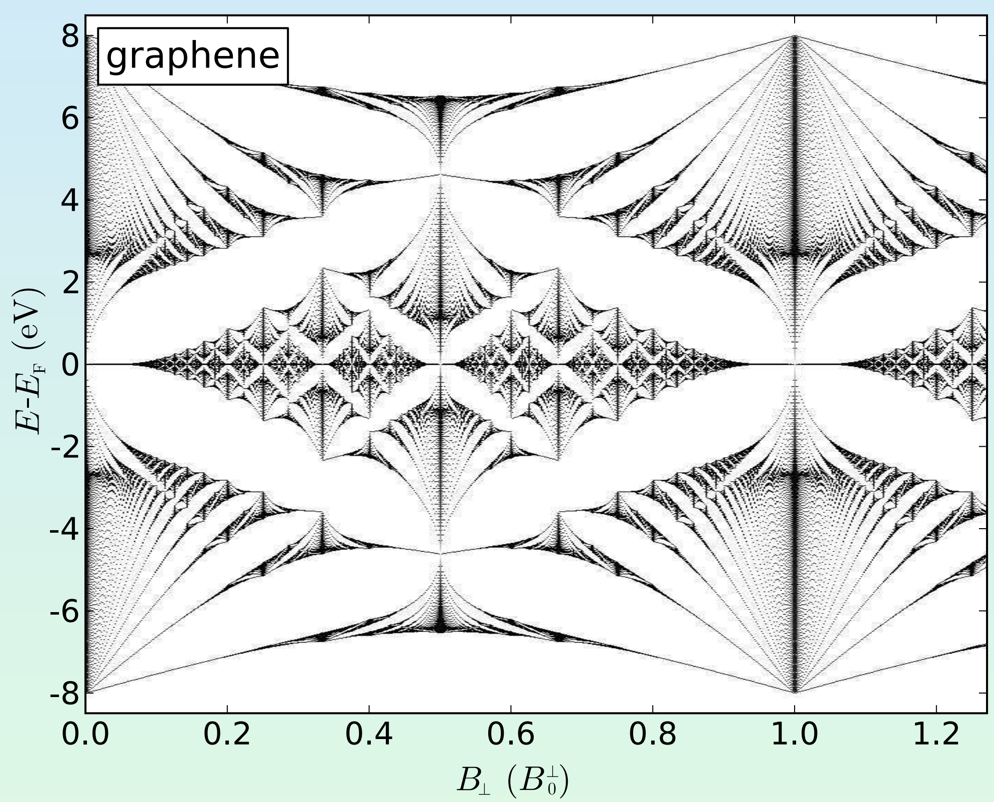
(6,5) CNT:
helical symmetry broken by B-field
$\Rightarrow$ large number of bands
low dispersion

(gap observed in optical experiments, J. Kono et al.)

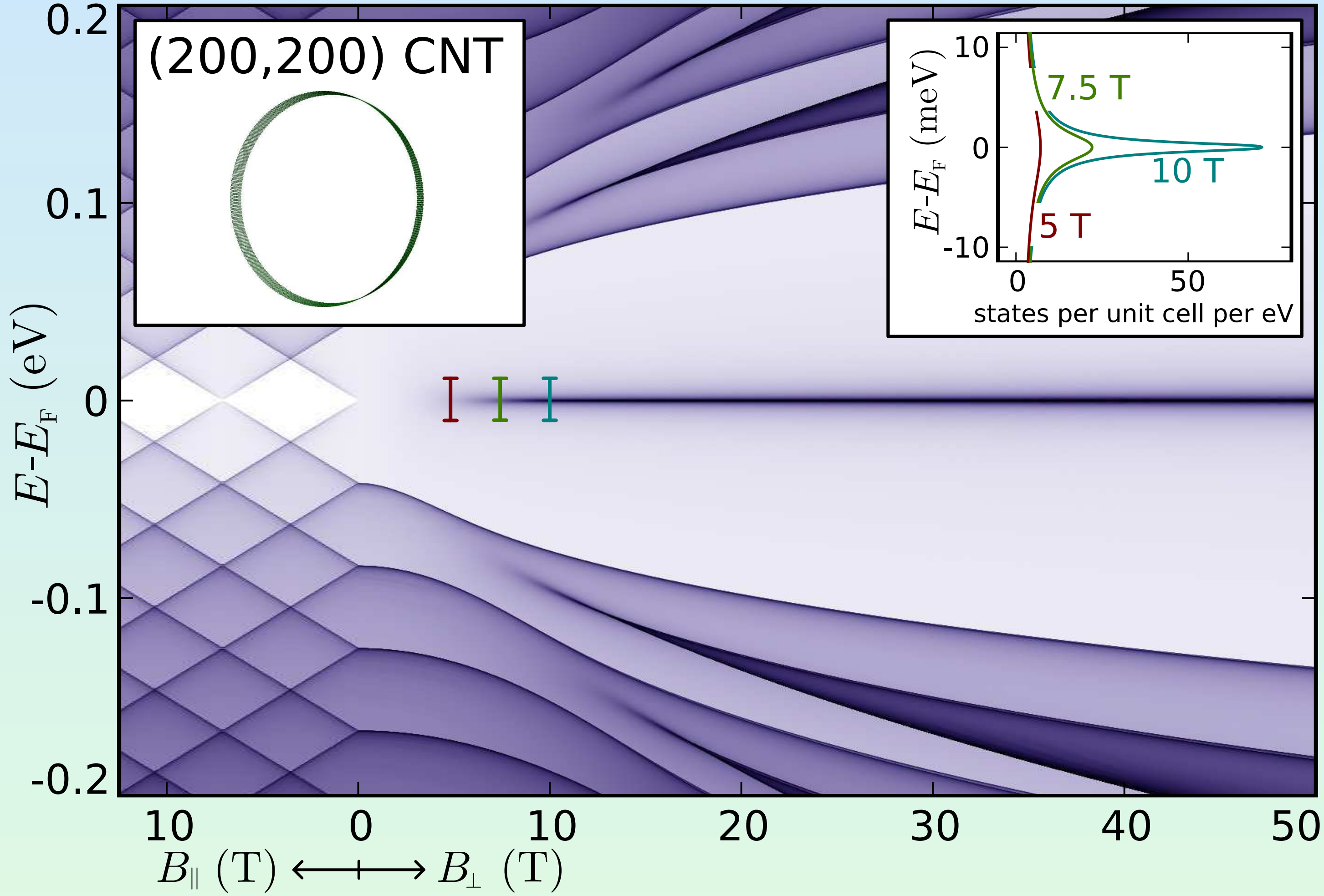
Huge SWCNT

(size comparable to external shell of MWCNT)



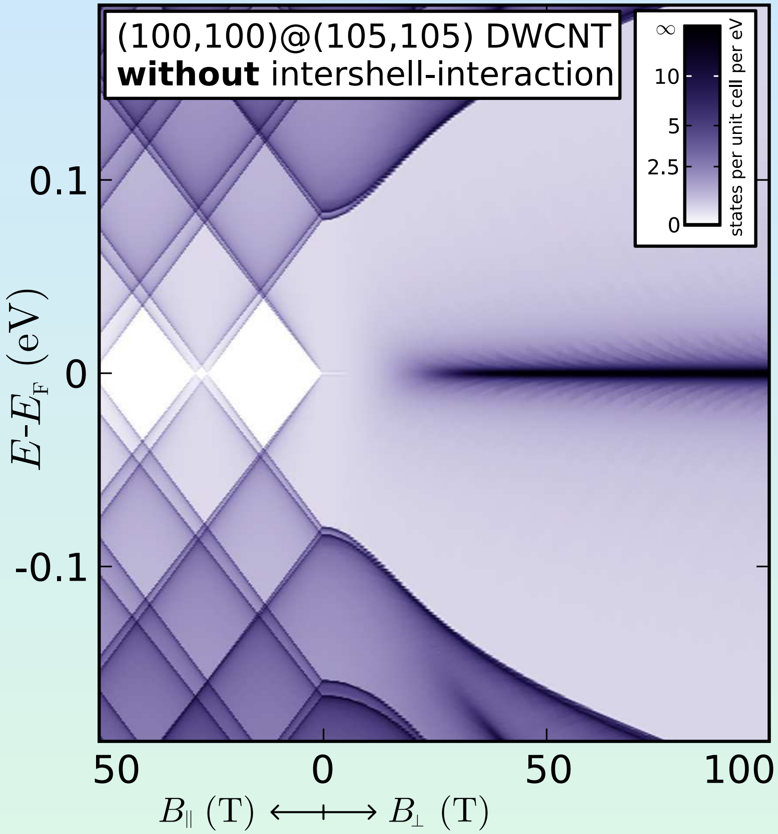


Experimentally accessible fields



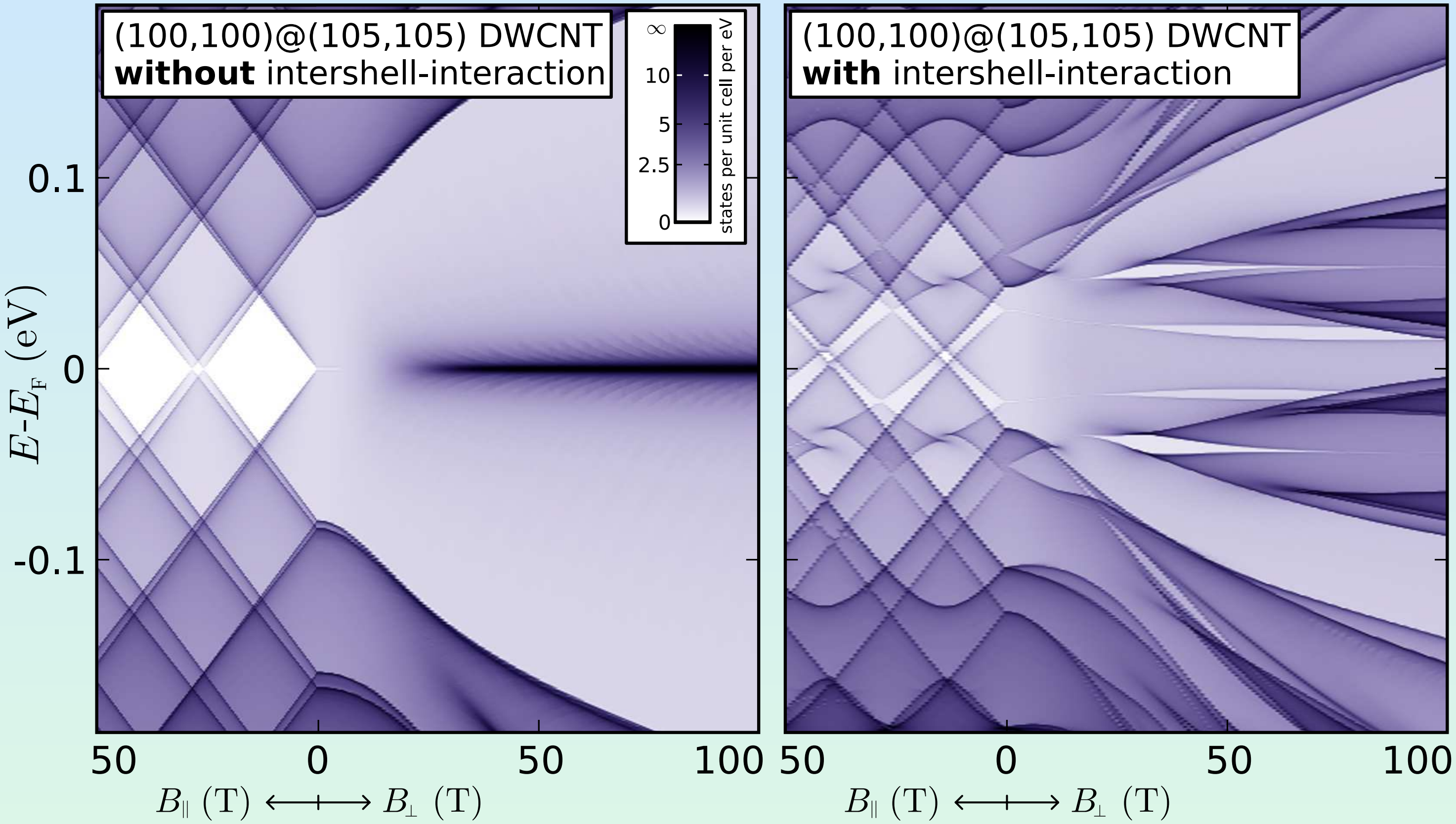
- scaling behavior: $\mathrm{DOS}_{(m,m)}(E,\boldsymbol{B}) = \mathrm{DOS}_{(m',m')}\left(\frac{m}{m'}E, \frac{m^2}{m'^2}\boldsymbol{B}\right)$

Inter-shell effects in DWCNT



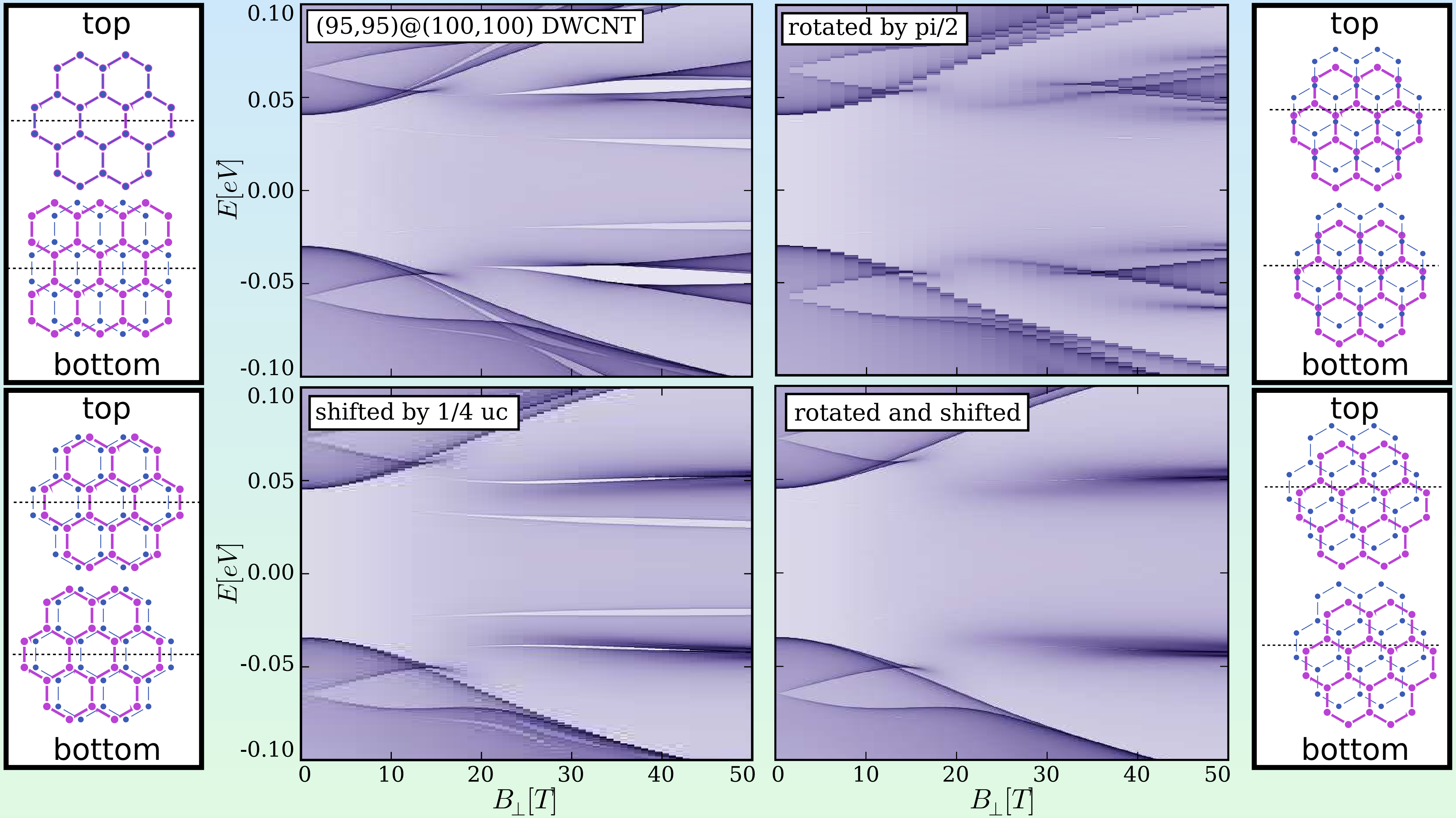
- no interaction $\Rightarrow$ $\mathrm{DOS} = \mathrm{DOS}_{\mathrm{inner}} + \mathrm{DOS}_{\mathrm{outer}}$

Inter-shell effects in DWCNT



- no interaction $\Rightarrow$ $\mathrm{DOS} = \mathrm{DOS}_{\mathrm{inner}} + \mathrm{DOS}_{\mathrm{outer}}$
- SuSyLL split up by intershell-interaction

Inter-shell effects in MWCNT



$\Delta E \approx 0.1\,\mathrm{eV}$ *independent of relative positions !!*