Graphene under uni-axial strain and 2D layered organic material α-(BEDT-TTF)$_2$I$_3$– the physics of tilted Dirac cones

CENTRE NATIONAL DE LA RECHERCHE SCIENTIFIQUE

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PRB **78**, 045415 (2008); EPL **85**, 57005 (2009);

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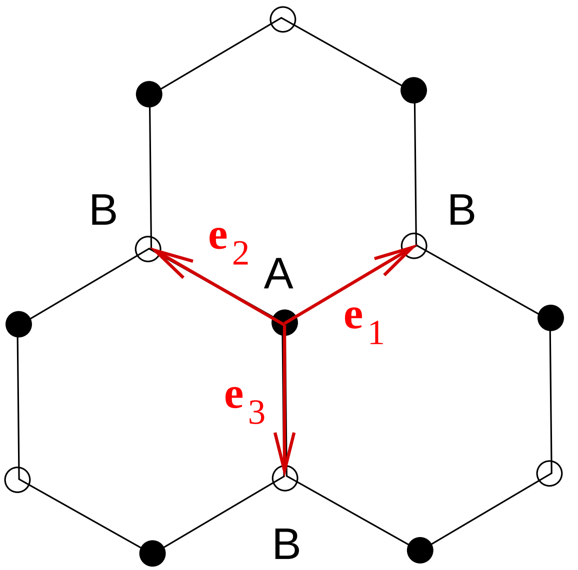
Outline of the Talk

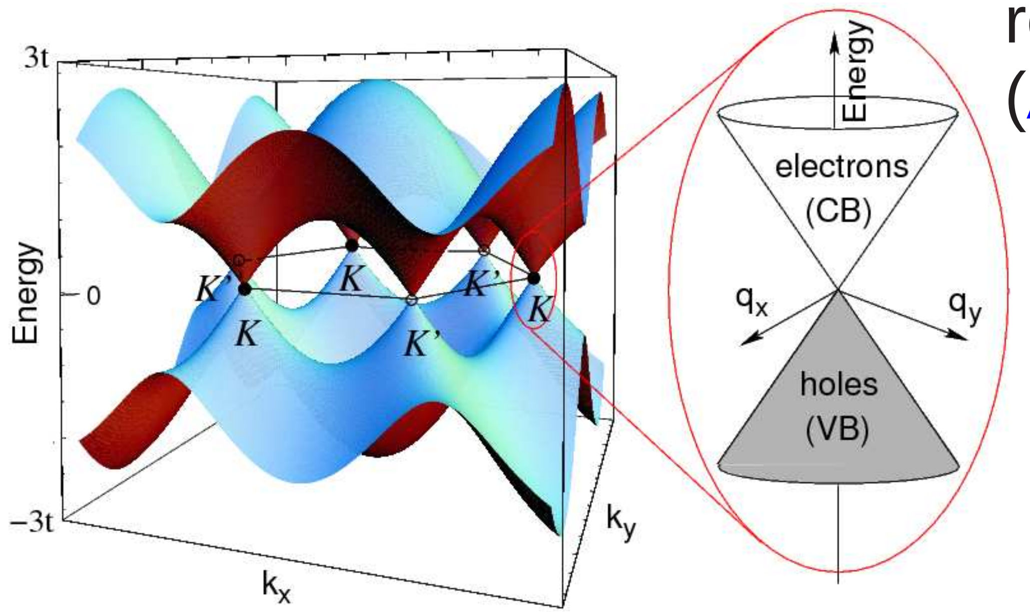
- Introduction to Dirac fermions in graphene and α-(BEDT-TTF)$_2$I$_3$
- Physical origin of tilted Dirac cones (example of distorted graphene)
- Motion of Dirac points and topological (semi-)metal-insulator transition
- (Tilted) Dirac cones in a strong magnetic field
  - reminder of graphene and effect of the tilt
  - crossed electric and magnetic fields in graphene and α-(BEDT-TTF)$_2$I$_3$
  - LL quantisation in the vicinity of the topological phase transition

Graphene bandstructure *papers from the '50ies*

- Tight-binding model (π-electron nn hopping)

$$H_0 = -t \sum_{i\in A} \sum_{j=1}^{3} \left(b^{\dagger}_{\mathbf{R}_i+\mathbf{e}_j} a_{\mathbf{R}_i} + \text{H.c.} \right)$$





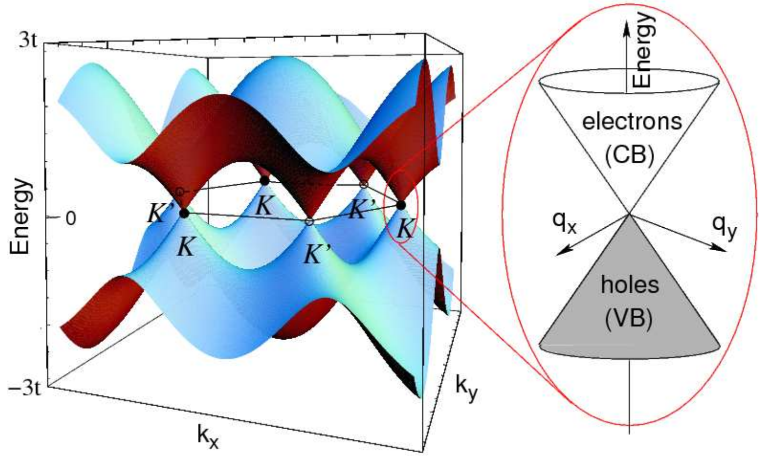
reciprocal-space Hamiltonian (A-B sublattice basis):

$$H_0 = t \begin{pmatrix} 0 & \gamma^*_{\mathbf{k}} \\ \gamma_{\mathbf{k}} & 0 \end{pmatrix}$$

$$\gamma_{\mathbf{k}} = \sum_j \exp(-i\mathbf{k}\cdot\mathbf{e}_j)$$

Dirac fermions

- Zero-energy states $(\varepsilon_{\mathbf{K}^{\pm}} = 0)$: at K and K' points of the 1st BZ



- Continuum limit $\mathbf{k} = \mathbf{K}^{\pm} + \mathbf{q}$ with $|\mathbf{q}| \ll 1/a$:

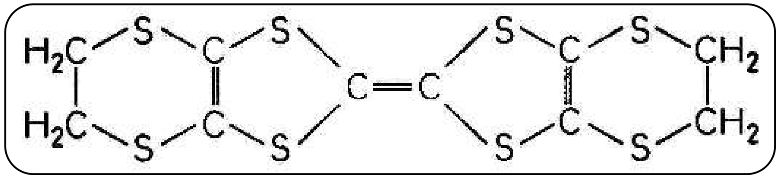
$$\mathcal{H}^{\xi=\pm}(\mathbf{q}) = \xi \frac{3}{2} t a \begin{pmatrix} 0 & q_x - i q_y \\ q_x + i q_y & 0 \end{pmatrix} = \xi \hbar v_F \boldsymbol{\sigma} \cdot \mathbf{q}$$

2D Dirac Hamiltonian for massless particles

- Energy dispersion (two-fold valley degeneracy $\xi = \pm$):

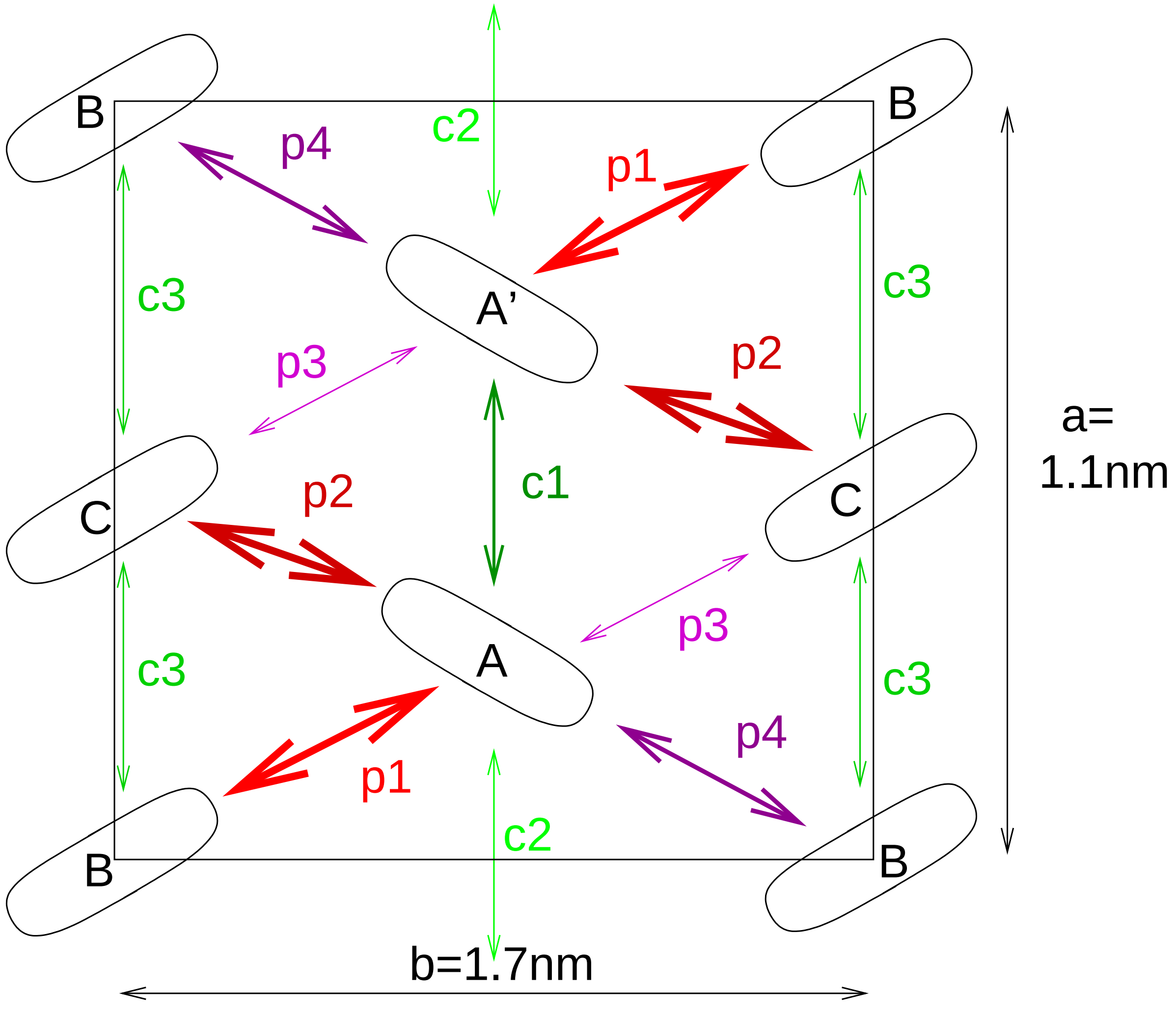
$$\varepsilon_{\lambda=\pm,\mathbf{q}}{}^{\xi=\pm} = \pm \hbar v_F |\mathbf{q}|$$

α-(BEDT-TTF)$_2$I$_3$: crystal structure



BEDT-TTF
=bis(ethylenedithio)tetrathiafulvalene

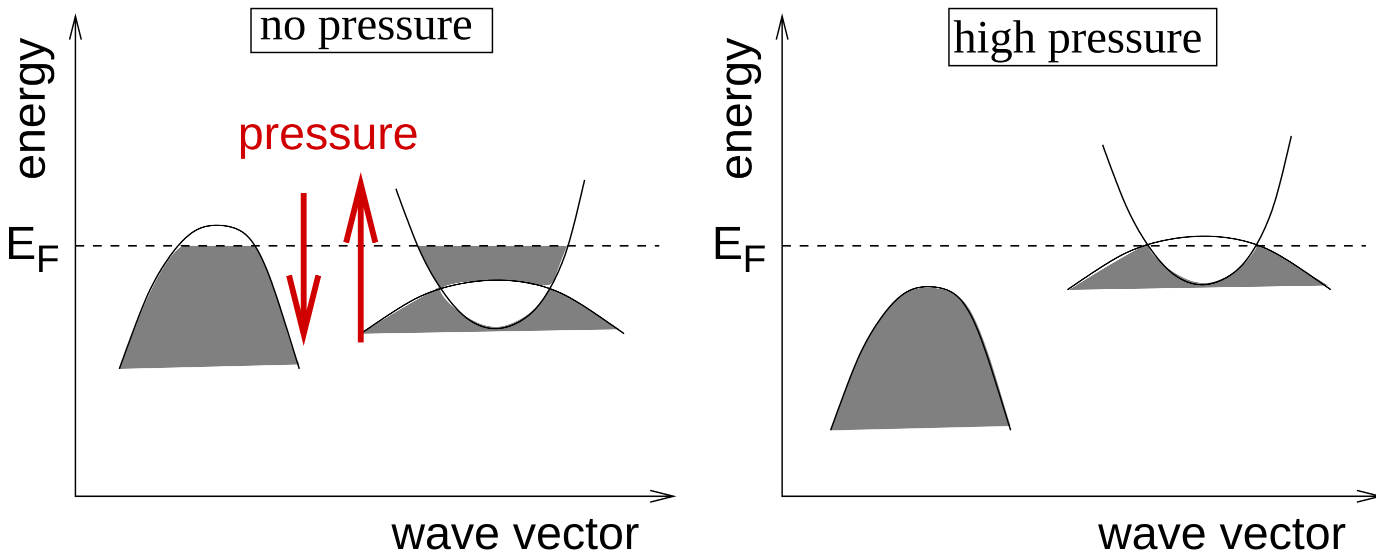
- quasi-2D (stacked layers)
- 4 molecules/unit cell $\rightarrow 4 \times 4$ Hamiltonian
- electronic filling: 3/4
- $t_i \sim 20 \ldots 140$ meV



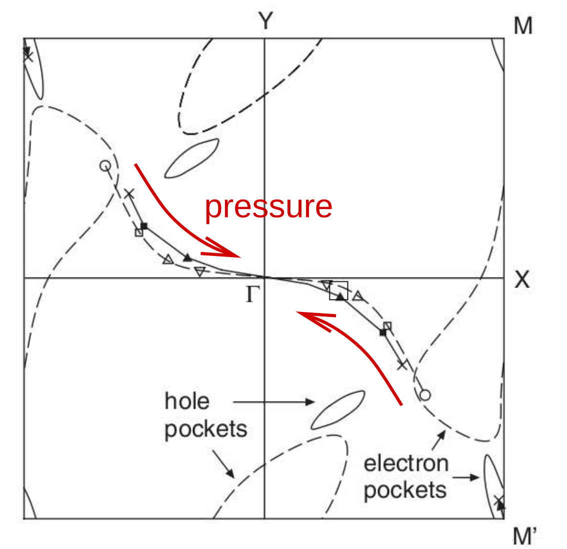
Review on 2D organics: Seo, Hotta, Fukuyama, Chem. Rev. **104**, 5005 (2004)

α-(BEDT-TTF)$_2$I$_3$: electronic band structure

schematic view on upper two bands



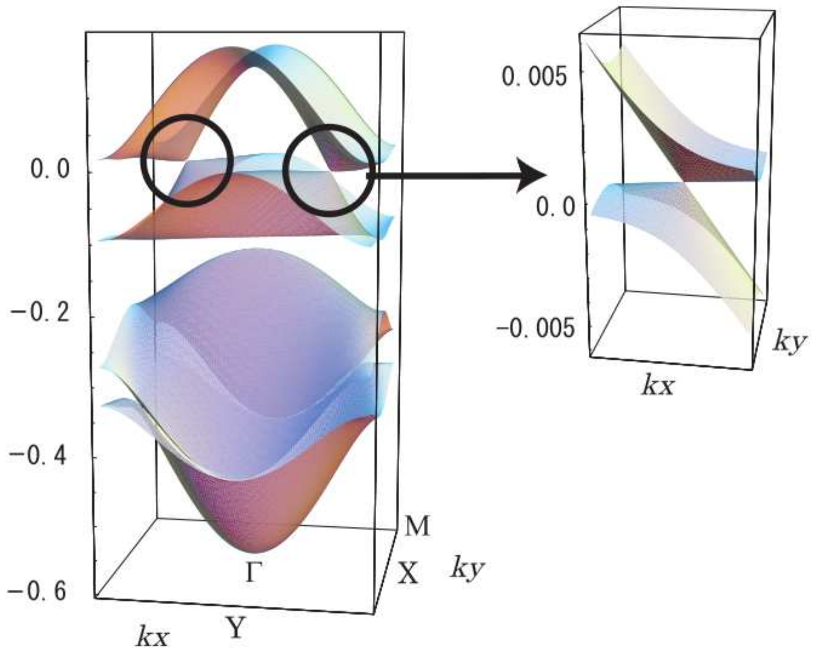
Brillouin zone



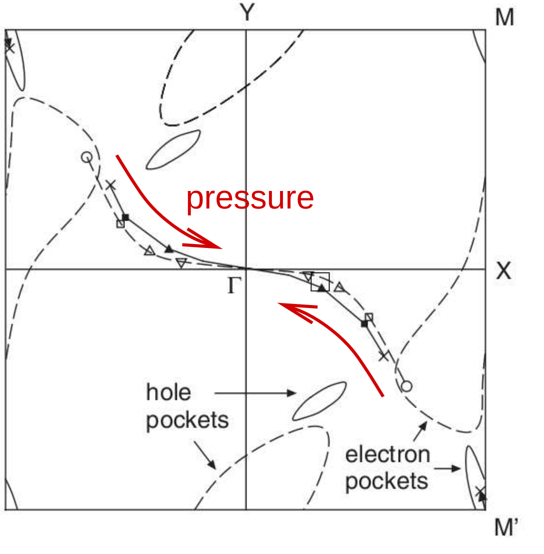
Katayama, Kobayashi, Suzumura, J. Phys. Soc. Japan **75**, 054705 (2006)

α-(BEDT-TTF)$_2$I$_3$: electronic band structure

energy bands under pressure



Brillouin zone



Katayama, Kobayashi, Suzumura, J. Phys. Soc. Japan **75**, 054705 (2006)

Complication due to strong correlations !

$\rightarrow$ charge ordering at $p = 0$ and $T = 0$

Correlation effects in α-(BEDT-TTF)$_2$I$_3$ and graphene

- extended Hubbard model:

$$U \simeq 0.4\,\mathrm{eV} \simeq 3t_{max}, \qquad V_c \simeq 0.17\,\mathrm{eV}, \qquad V_p \simeq 0.05\,\mathrm{eV}$$

U: onsite repulsion
V_p, V_c: nearest neighbour repulsion

- "fine-structure constant" in continuum model:

$$r_s = \frac{e^2}{\hbar \epsilon v_F} \simeq 20/\epsilon \simeq 10\, r_s^{\mathrm{graphene}}$$

$v_F \sim t \times a \simeq 0.1 v_F^{\mathrm{graphene}}$: average Fermi velocity

$\Rightarrow$ bare correlations in α-(BEDT-TTF)$_2$I$_3$ ~ 10 times larger than in graphene

Correlations and Dirac points – screening

- (static) RPA screening:

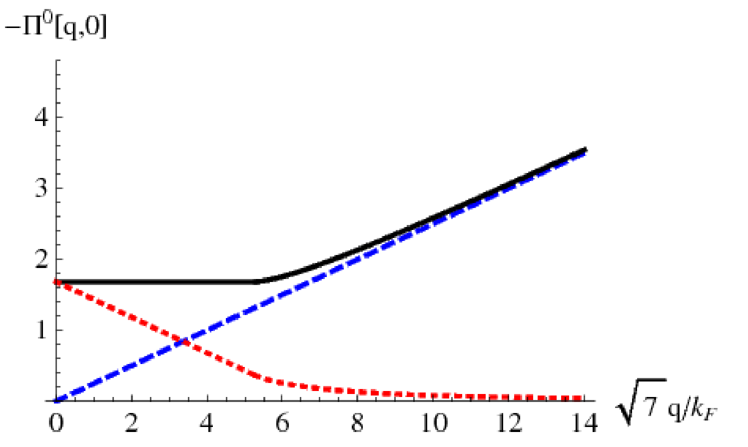
$$\epsilon^{\mathrm{RPA}}(\mathbf{q}) = 1 - \frac{2\pi e^2}{\epsilon q}\Pi^0(\mathbf{q})$$

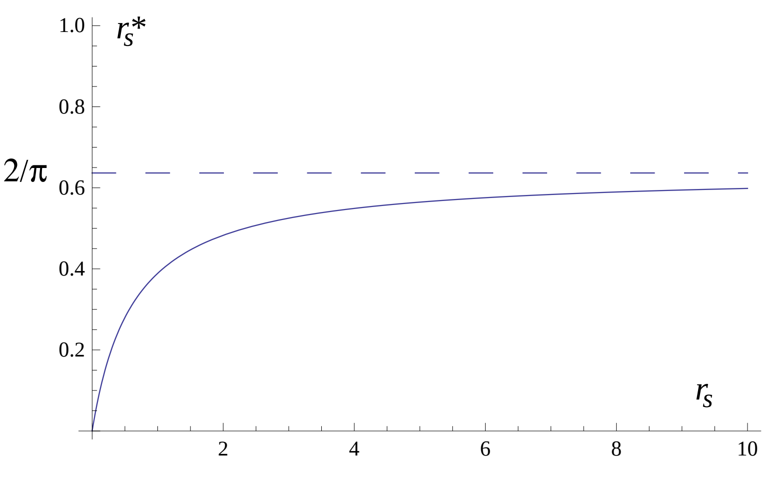
- bare polarisability:
  two contributions

$$\Pi^0(\mathbf{q}) = \Pi^{\mathrm{vac}}(\mathbf{q}) + \Pi^{\mathrm{dop}}(\mathbf{q})$$

- interband contribution absorbed in $\epsilon \longrightarrow \epsilon\epsilon_\infty$

$$r_s^* = \frac{r_s}{\epsilon_\infty} = \frac{r_s}{1+\pi r_s/2}$$





Thomas-Fermi type argument valid only for finite doping !

Generalised Weyl Hamiltonian

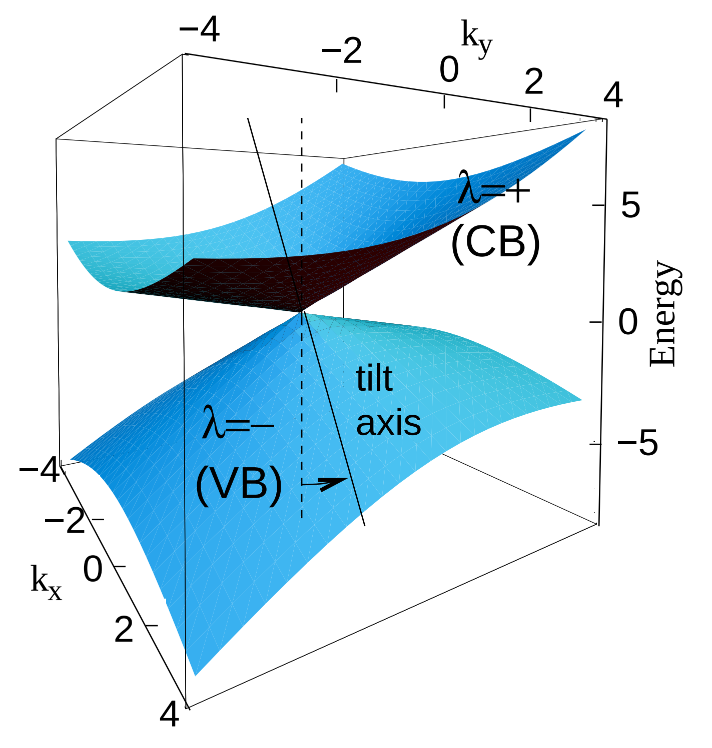
Most general 2D Hamiltonian (2×2 matrix) with linear dispersion (generalised Weyl Hamiltonian):

$$H = \sum_{\mu=0}^{3} \hbar \mathbf{v}_\mu \cdot \mathbf{q}\, \sigma^\mu \qquad (\sigma^0 = \mathbb{1},\ \sigma^1 = \sigma^x,\ \sigma^2 = \sigma^y,\ \sigma^3 = \sigma^z)$$

$$\hat{=}\ \hbar\,(\mathbf{w}_0 \cdot \mathbf{q}\,\mathbb{1} + w_x q_x \sigma^x + w_y q_y \sigma^y)$$

Energy dispersion ($\hbar \equiv 1,\ \lambda = \pm$):

$$\epsilon_\lambda(\mathbf{q}) = \mathbf{w}_0 \cdot \mathbf{q} + \lambda \sqrt{w_x^2 q_x^2 + w_y^2 q_y^2}$$



$\mathbf{w}_0$: “tilt velocity”

Graphene: $\mathbf{w}_0 = 0,\ w_x = w_y = v_F$

How to obtain tilted Dirac cones?

- In graphene: $\boldsymbol{\sigma}$ denotes $A - B$ sublattice isospin
- Term proportional to $\mathbb{1}$: nnn hopping $(A \leftrightarrow A, B \leftrightarrow B)$

$\Rightarrow$ In continuum limit:

$$H_{\mathrm{diag}} = \frac{9}{4} t_{nnn} |\mathbf{q}|^2 a^2 \, \mathbb{1}$$

i.e. not linear in $\mathbf{q}$, but quadratic

- Reason: Dirac points $[\epsilon(\mathbf{q}_D) = 0]$ coincide with K, K' (points of high crystallographic symmetry)

$\Rightarrow$ Drag Dirac points away from K, K' !

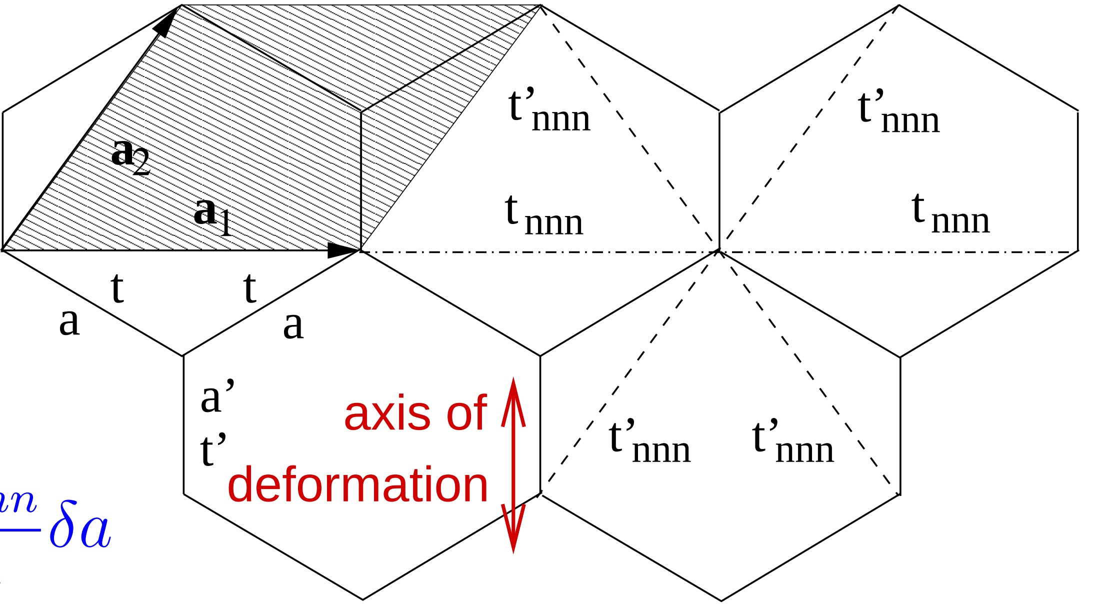
Graphene under strain (I)

Distortion:

$$a \longrightarrow a' = a + \delta a$$

$$t \longrightarrow t' = t + \frac{\partial t}{\partial a}\delta a$$

$$t_{nnn} \longrightarrow t'_{nnn} = t_{nnn} + \frac{\partial t_{nnn}}{\partial a}\delta a$$

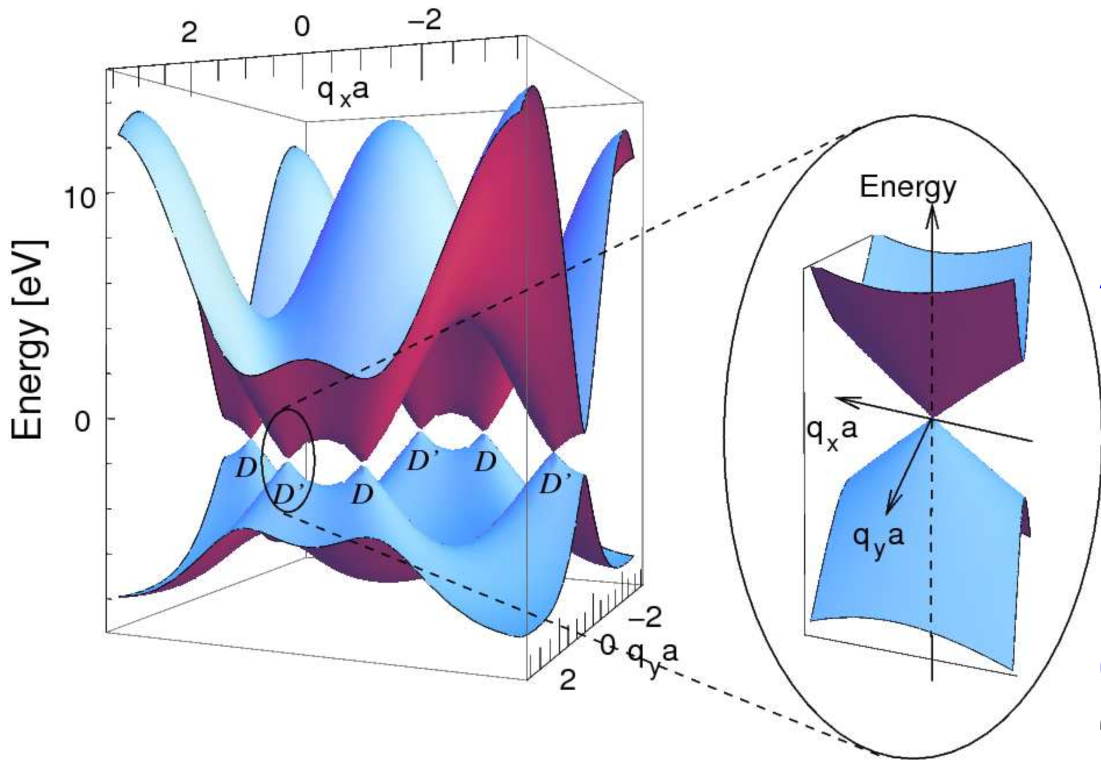


Dirac points move from K, K' to:

$$q_y^D = 0, \qquad q_x^D a = \xi \frac{2}{\sqrt{3}} \arccos\left(-\frac{t'}{2t}\right)$$

ξ: valley index

Graphene under strain (II)



Estimation of tilt:

$$\tilde{w}_0 \equiv \sqrt{\left(\frac{w_{0x}}{w_x}\right)^2 + \left(\frac{w_{0y}}{w_y}\right)^2} \simeq 0.6\,\frac{\delta a}{a}$$

$0 \leq \tilde{w}_0 < 1$:
"tilt parameter"

$\Rightarrow$ Effect linear in $\delta a/a$!

MOG, J.-N. Fuchs, F. Piéchon, G. Montambaux, PRB **78**, 045415 (2008)

Dirac-point motion and topological phase transition

Dirac points located at:

$$q_y^D = 0, \qquad q_x^D a = \xi \frac{2}{\sqrt{3}} \arccos\left(-\frac{t'}{2t}\right)$$

– topological PT at $t' = 2t$
(semi-metal/insulator)

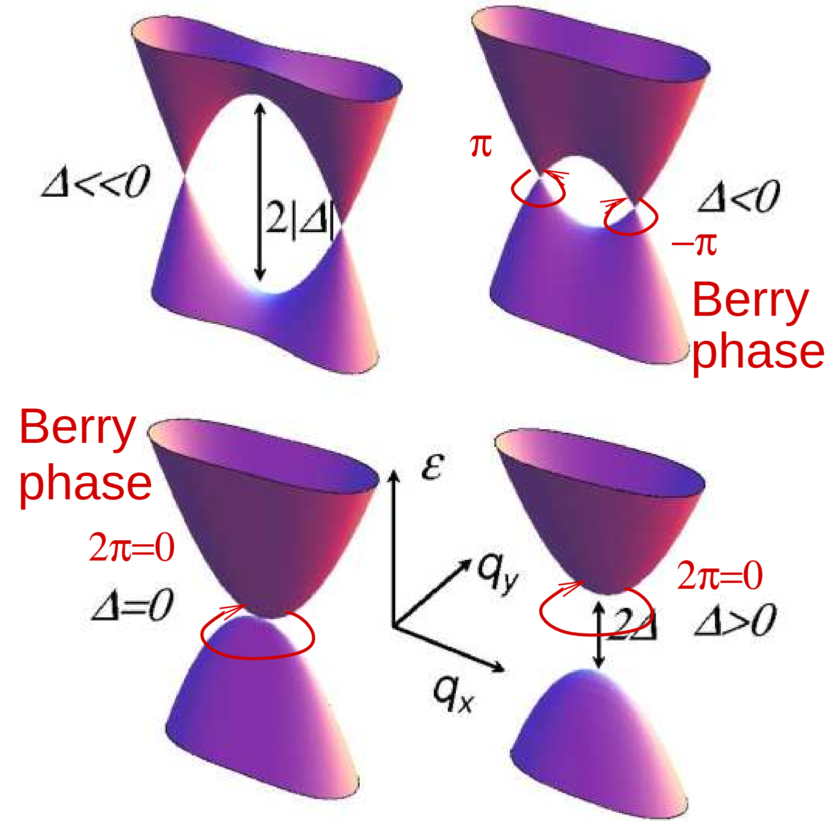
– effective Hamiltonian:

$$H' = \begin{pmatrix} 0 & \Delta + \frac{q_x^2}{2m^*} - icq_y \\ \Delta + \frac{q_x^2}{2m^*} + icq_y & 0 \end{pmatrix}$$

Montambaux, Piéchon, Fuchs, MOG, PRB **80**, 1534012 (2009)

– linear-quadratic dispersion at $t' = 2t$

Dietl, Piéchon, Montambaux, PRL **100**, 236405 (2008)



Dirac Cones in a Strong Magnetic Field

Relativistic Landau levels

Graphene Landau levels

- Magnetic field $(B\mathbf{e}_z = \nabla \times \mathbf{A})$ via Peierls substitution:

$$\mathbf{q} \rightarrow -i\nabla + e\mathbf{A}$$

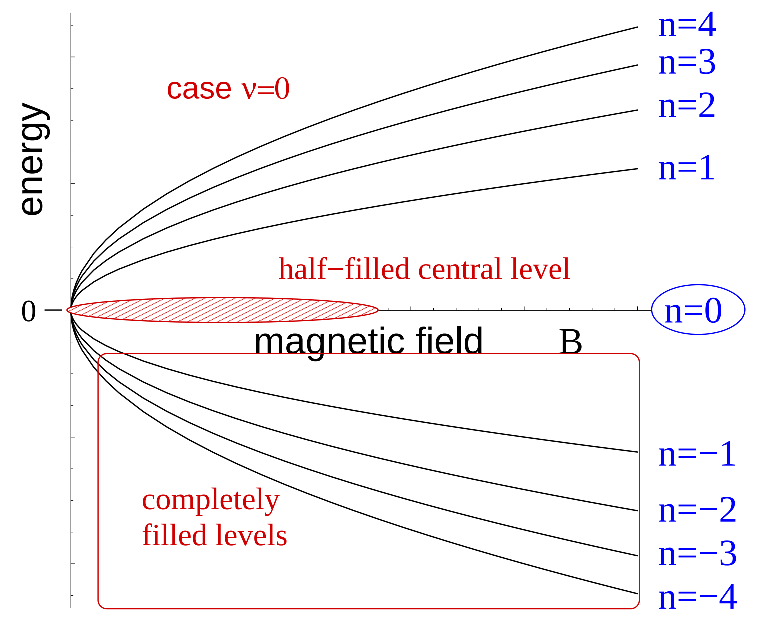
(semiclassical: $\varepsilon(\mathbf{q}) \rightarrow \varepsilon(\sqrt{2n}/l_B)$, $l_B = 1/\sqrt{eB}$)

- Energy dispersion with magnetic field (degenerate in valley isospin ξ):

$$\varepsilon_{\lambda,n} = \lambda \frac{v_F}{l_B} \sqrt{2|n|} \propto \sqrt{B|n|}$$

(Relativistic LLs)

- Quantum Hall effect at $\nu = \pm 2, \pm 6, \pm 10, \ldots$



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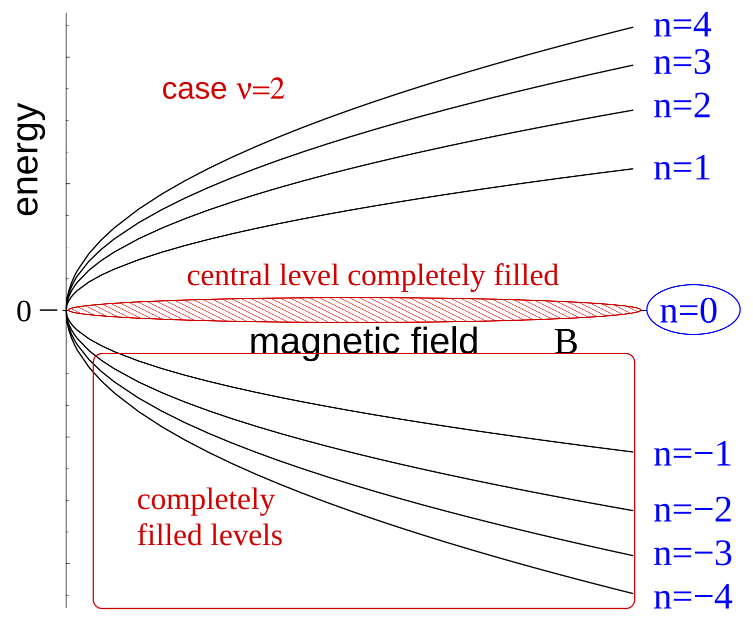
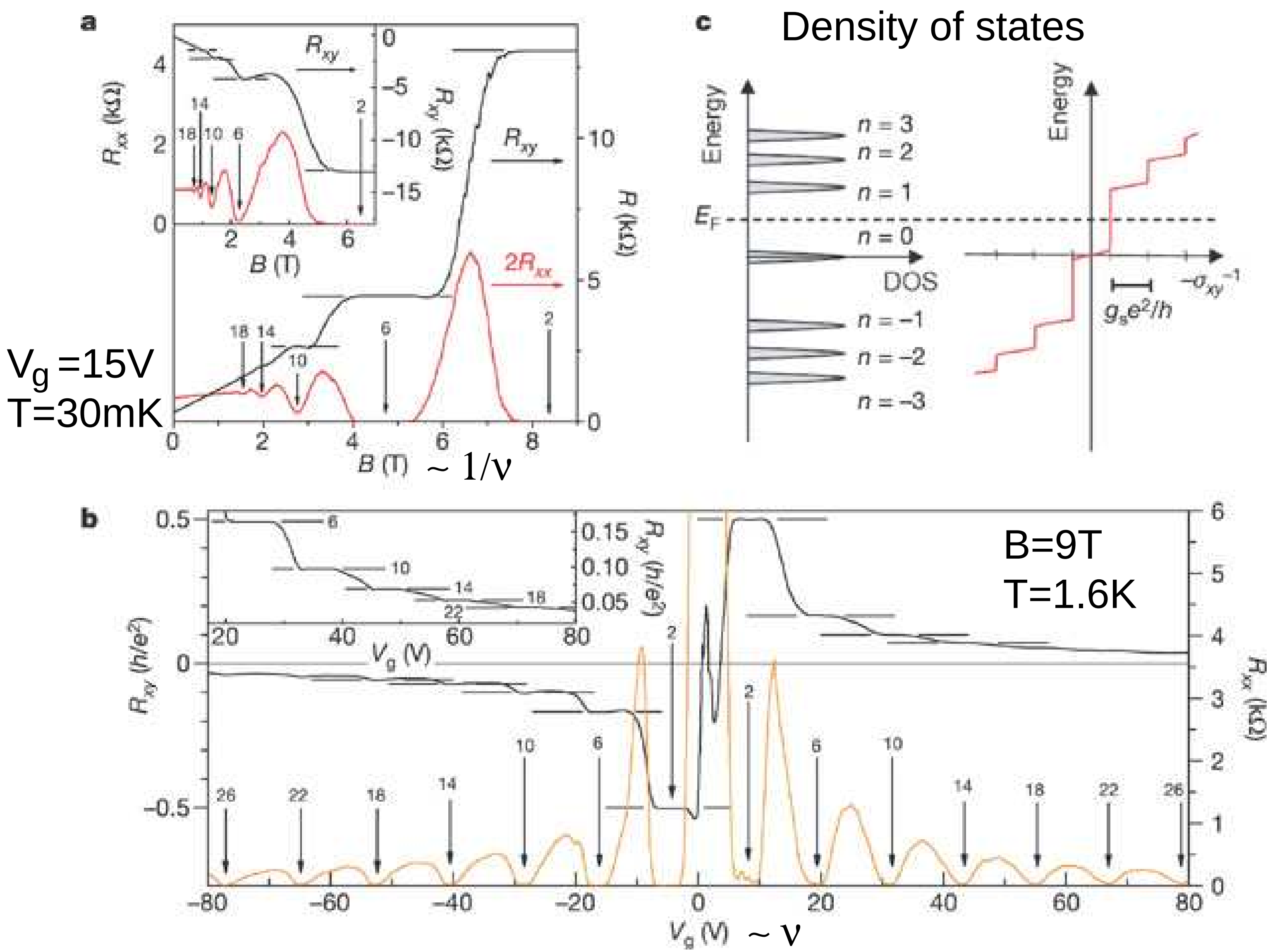


IQHE in graphene

Novoselov et al., Nature 438, 197 (2005)

Zhang et al., Nature 438, 201 (2005)



Graphene IQHE:

$R_H = h/e^2\nu$

at $\nu = 2(2n+1)$

Usual IQHE:

at $\nu = 2n$

(no Zeeman)

Tilted cones in a strong magnetic field

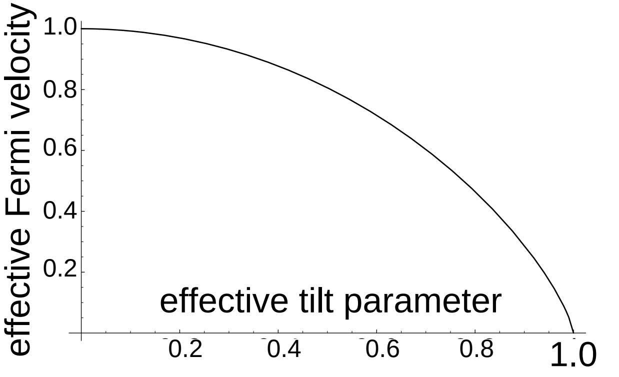
Motivation: How is the Landau level spectrum affected by the tilt?

$$H_\xi = \xi \left(\mathbf{w}_0 \cdot \mathbf{q}\, \mathbb{1} + w_x q_x \sigma^x + w_y q_y \sigma^y \right) \qquad \tilde{w}_0 = \sqrt{\left(\frac{w_{0x}}{w_x}\right)^2 + \left(\frac{w_{0y}}{w_y}\right)^2}$$

⊕ Peierls substitution: $\mathbf{q} \to \mathbf{q} + e\mathbf{A}(\mathbf{r})$ ⊕ semiclassics

⇒ energy spectrum in α-(BEDT-TTF)$_2$I$_3$ (as for graphene):

$$\epsilon_{\lambda,n} = \lambda \frac{v_F^*}{l_B}\sqrt{2n}$$



MOG, J.-N. Fuchs, F. Piéchon, G. Montambaux, PRB **78**, 045415 (2008)

- effect of the tilt: renormalisation $v_F^* = \sqrt{w_x w_y}\left(1 - \tilde{w}_0^2\right)^{3/4}$

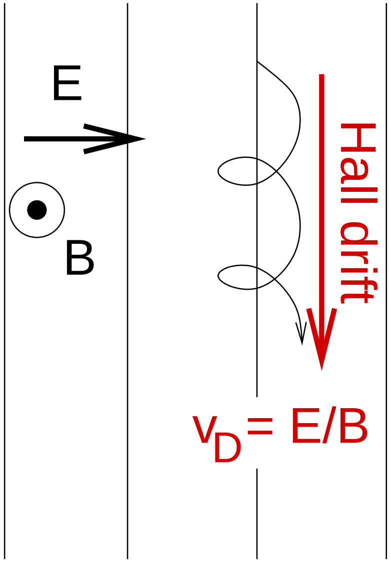
(!) Landau level $n = 0$ requires full quantum treatment !

Crossed magnetic and electric fields in graphene

graphene in crossed $B \perp E$ fields Lukose et al. PRL **98**, 116802 (2007)

- Lorentz boost into reference frame
  $v_D = E/B < v_F$:

$$B \to B' = B\sqrt{1 - (E/v_F B)^2}$$



- energy in rest frame:

$$\epsilon' \propto 1/l'_B \propto \sqrt{B'}$$

$\Rightarrow$ energy in lab frame:
$v_F \to v_F[1 - (E/v_F B)^2]^{3/4}$

- "tilt":

$$\tilde{w} = E/v_F B < 1$$

- magnetic regime
  $\sim$ closed orbits

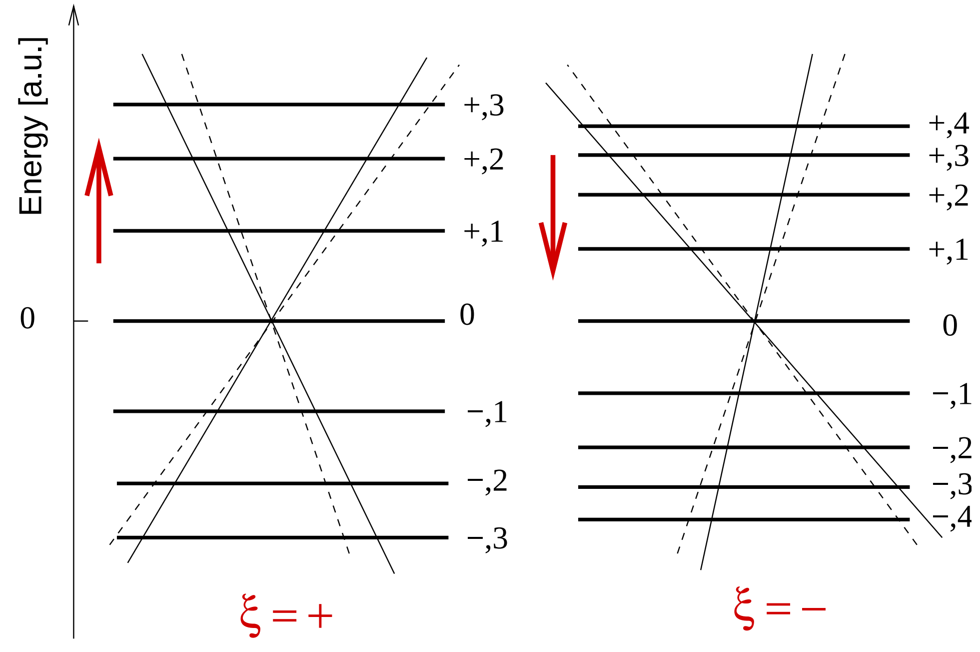
Crossed magnetic and electric fields (tilted cones)

- inplane E field affects tilt:

$$\mathbf{w}_0 \longrightarrow \mathbf{w}_\xi = \mathbf{w}_0 - \xi \frac{\mathbf{E} \times \mathbf{B}}{B^2}$$

$\Rightarrow$ new tilt parameter

$$\tilde{w}_\xi(E) = \sqrt{\frac{w_{\xi x}^2}{w_x^2} + \frac{w_{\xi y}^2}{w_y^2}}$$



Landau level spectrum depends on valley index ξ :

$$\epsilon_{\lambda,n;k}^{\xi}(E) = \lambda \frac{\sqrt{w_x w_y}}{l_B} \left[1 - \tilde{w}_\xi(E)^2\right]^{3/4} \sqrt{2n} + \frac{E}{B} k$$

MOG, J.-N. Fuchs, G. Montambaux, F. Piéchon, EPL **85**, 57005 (2009)

Possible experimental verification

1. Quantum Hall measurement: (problematic)

lifted valley degeneracy $\rightarrow$ additional plateaus

- no single-layer α-(BEDT-TTF)$_2$I$_3$!

$\Rightarrow$ field-effect doping may be inhomogenious over layers

- in graphene: strong strain ($10-20\%$) + large inplane electric field ($\sim 10^6$V/m)

$\Rightarrow$ maximal 1% effect

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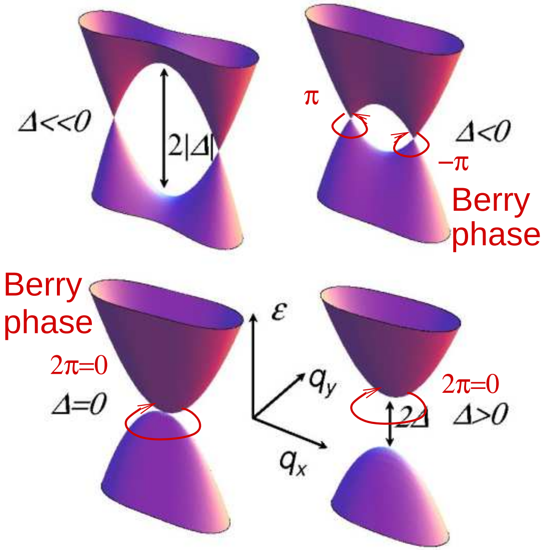
2. Infrared transmission spectroscopy in α-(BEDT-TTF)$_2$I$_3$

- $\sim 10\%$ effect in doubled absorption lines

$$\Delta_n^\xi(E) = \frac{\sqrt{2w_x w_y}}{l_B}\left[1-\tilde{w}_\xi(E)^2\right]^{3/4}\left(\sqrt{n}+\sqrt{n+1}\right)$$

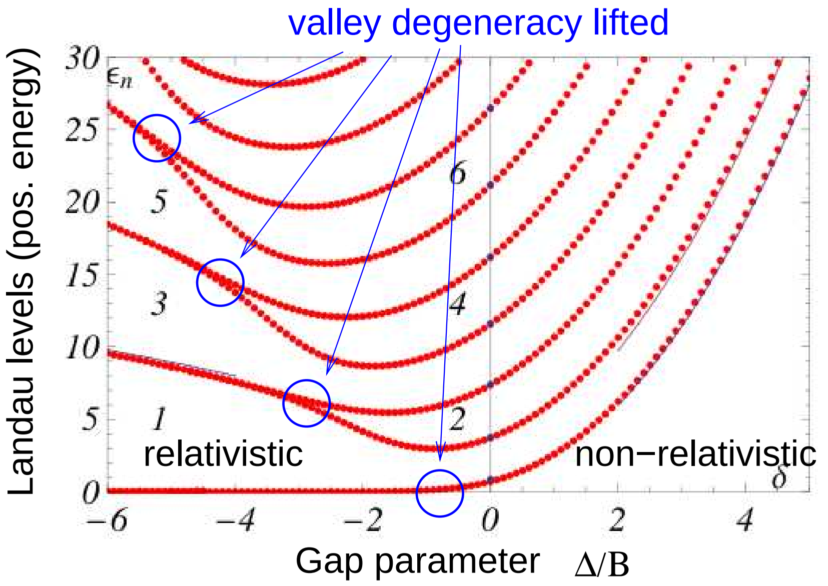
LL quantisation in the vicinity of the topological PT

$$H' = \begin{pmatrix} 0 & \Delta + \frac{q_x^2}{2m^*} - icq_y \\ \Delta + \frac{q_x^2}{2m^*} + icq_y & 0 \end{pmatrix}$$



– in a magnetic field:

- semi-metal ($m^*\Delta < 0$):
  $\epsilon_n \propto \pm\sqrt{nB}$
- transition ($\Delta = 0$):
  $\epsilon_n \propto \pm\left[\left(n+\frac{1}{2}\right)B\right]^{2/3}$
- insulator ($m^*\Delta > 0$):
  $\epsilon_n \propto \pm\left[\Delta + \#\left(n+\frac{1}{2}\right)B\right]$



Montambaux, Piéchon, Fuchs, MOG, PRB **80**, 153412 (2009); EPJB **73**, 509 (2009)

Conclusions (I)

Study of tilted Dirac cones in distorted graphene and α-(BEDT-TTF)$_2$I$_3$

- model: generalised Weyl Hamiltonian

$$H = \mathbf{w}_0 \cdot \mathbf{q}\, \mathbb{1} + w_x q_x \sigma^x + w_y q_y \sigma^y$$

- Landau level spectrum: decreased spacing due to tilt
- zero-energy Landau level survives the tilt
- possible valley degeneracy lifting in crossed magnetic and electric fields

$\Rightarrow$ possible experimental verification in infrared-transmission spectroscopy

Conclusions (II)

Dirac-point motion and topological (S)MIT

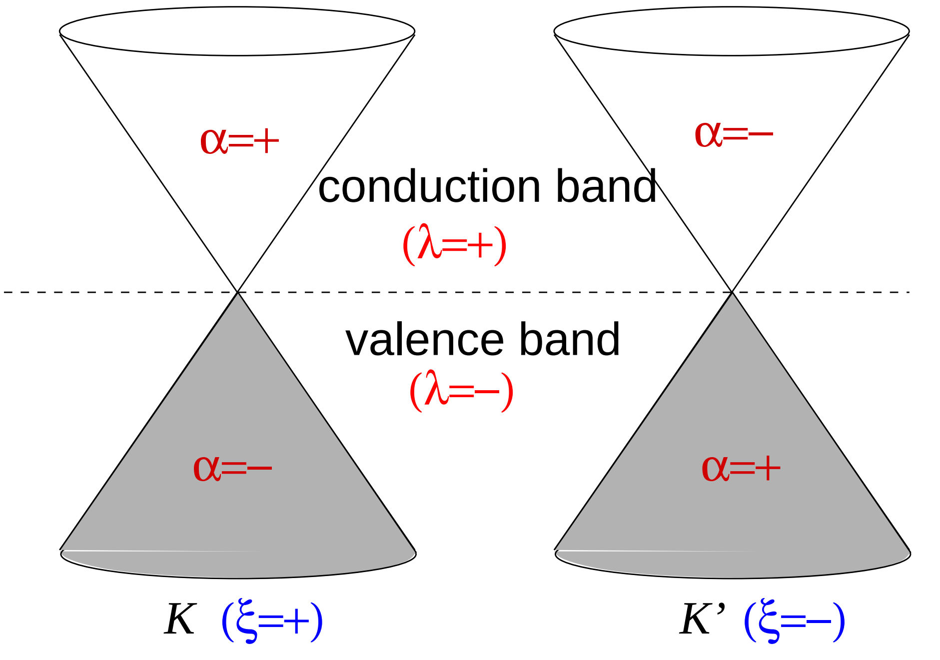
- model Hamiltonian (linear-quadratic)

$$H' = \begin{pmatrix} 0 & \Delta + \frac{q_x^2}{2m^*} - icq_y \\ \Delta + \frac{q_x^2}{2m^*} + icq_y & 0 \end{pmatrix}$$

- topological transition at $\Delta = 0$ (Berry phase): changefrom $\pm\pi$ around Dirac pts to 0 at merging pt
- particular LL quantisation
- possible experimental verification:
  - unphysical distortion in graphene
  - topological PT at Γ-point in α-(BEDT-TTF)$_2$I$_3$ (> 40 kbar)
  - cold atoms

Isospin quantum numbers

- valley isospin $\xi = \pm$: two-fold degeneracy
- band index $\lambda = \pm$: valence band (VB) or conduction band (CB)
- chirality $\alpha = \boldsymbol{\sigma} \cdot \boldsymbol{\kappa}/|\boldsymbol{\kappa}| = \pm$ (sublattice spin projected on wave vector)
- (true spin $s = \uparrow, \downarrow$)



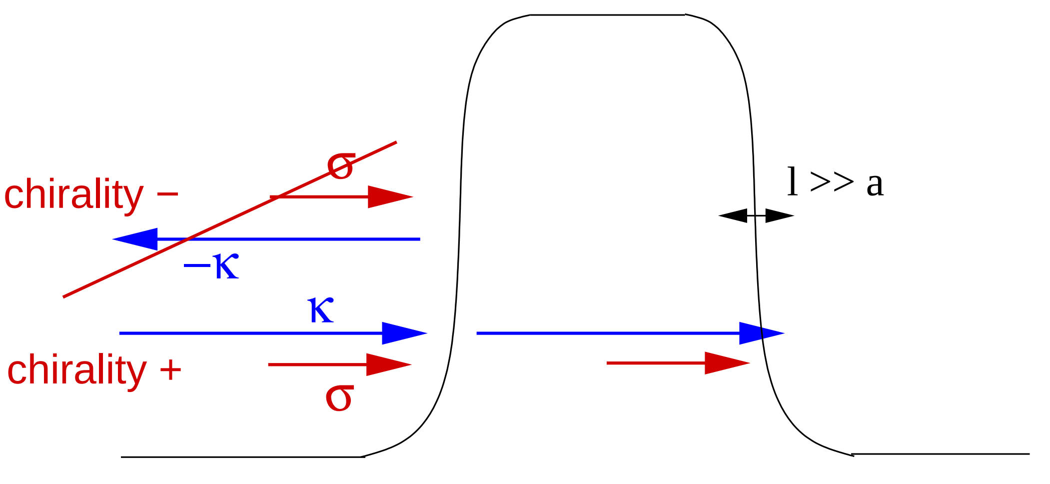
band index $=$ valley isospin $\times$ chirality

Absence of backscattering – Klein tunneling

Slowly varying obstacle (elasic scatterer): $H_{diff} = V(\mathbf{r})\mathbb{1}$

$\Rightarrow$ no sublattice-isospin coupling

$\Rightarrow$ no valley coupling + elastic $\Rightarrow$ chirality conservation



Chirality conservation $\Rightarrow$ absence of backscattering

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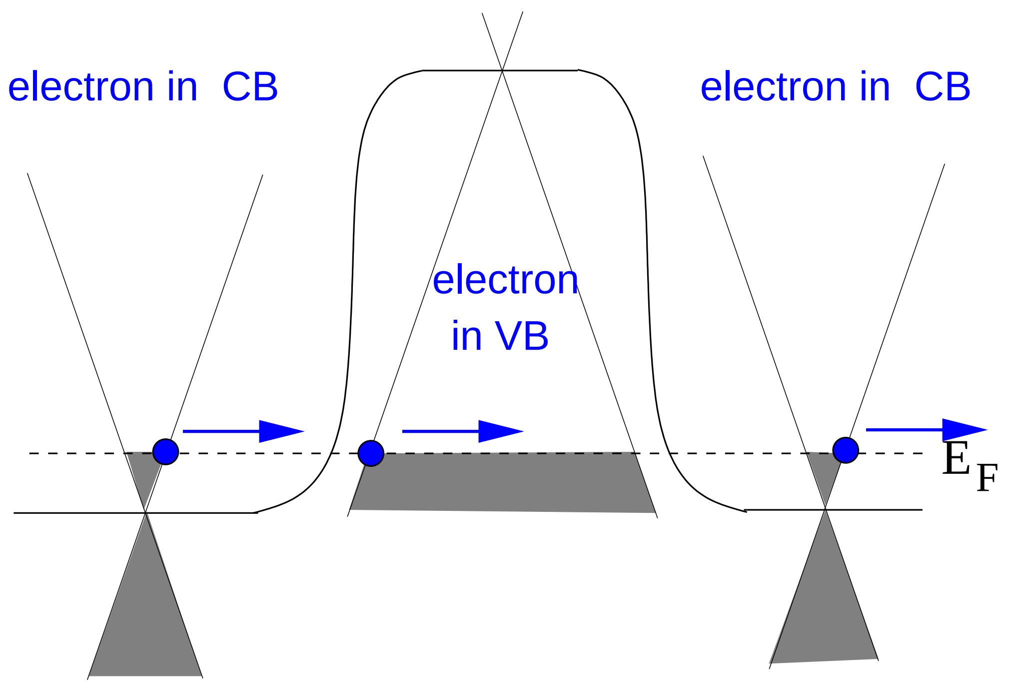

Klein tunneling at a potential barrier *vs.* qm tunneling