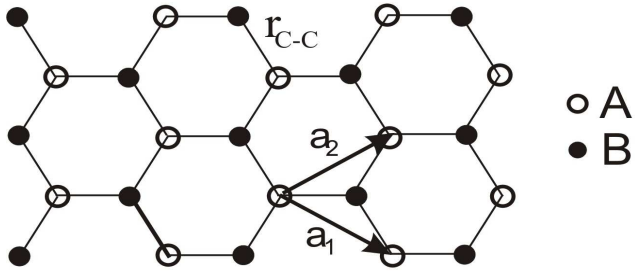
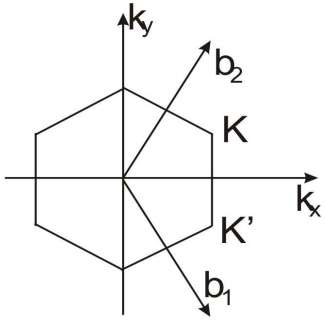
# Spin-pálya kölcsönhatás grafénben, fotoelektron-spektroszkópia

*Rakyta Péter*

EÖTVÖS LORÁND TUDOMÁNYEGYETEM, KOMPLEX RENDSZEREK FIZIKÁJA TANSZÉK

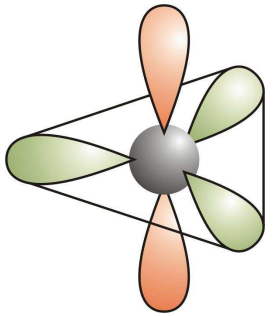
# Introduction to graphene



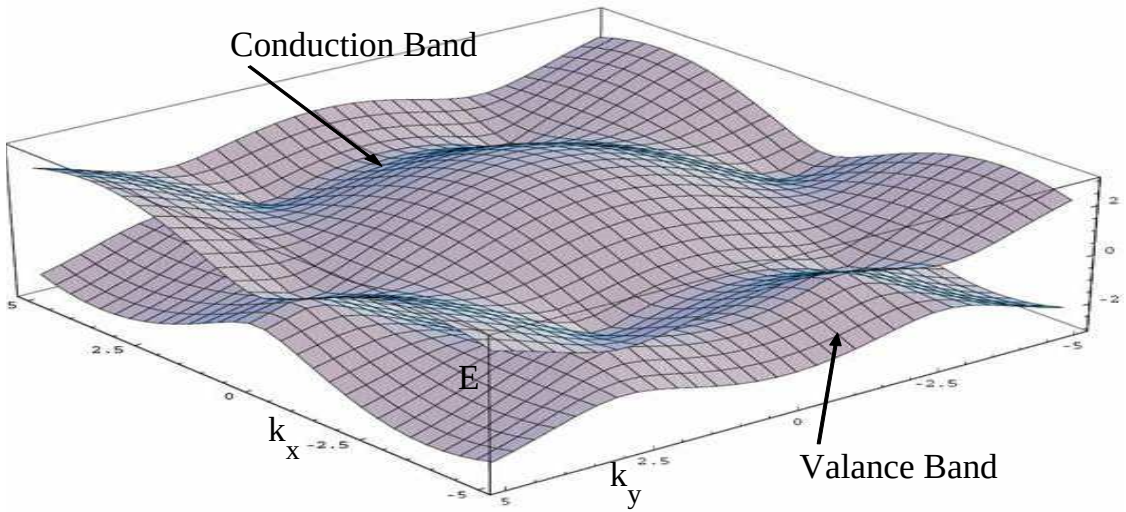

➠ $Sp^2$ hybridization

➠ $p^z$ orbitals

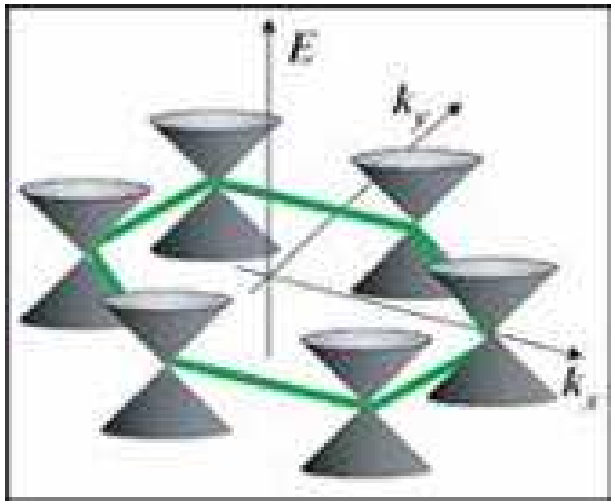


➠ Low energy excitation around $K$ points with linear dispersion.

Spin-degenerated energy bands ($p^z$ electrons):



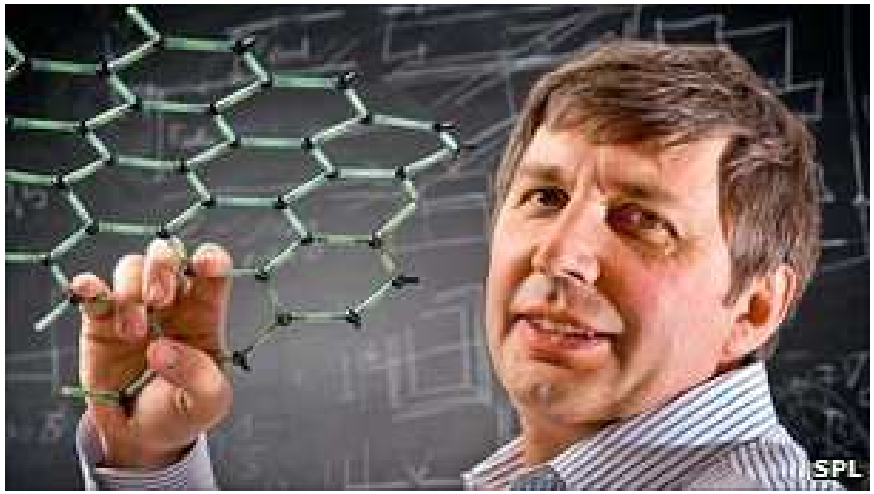

Dirac-cones:




Fermi level

# Nobel prize in 2010

Andre Geim (University of Manchester in the UK)




Konstantin Novoselov

➠ The Royal Swedish Academy of Sciences has decided to award the Nobel Prize in Physics for 2010

”for groundbreaking experiments regarding the two-dimensional material graphene”.

# Spin-orbit coupling in graphene

- Intrinsic spin-orbit (ISO) originates from $C$ atoms: $\Delta_{SO} \sim 12\mu eV$

  DFT calculations: D. Huertas-Hernando, F. Guinea, and A. Brataas, Phys. Rev. B **74**, 155426 (2006);

  M. Gmitra, S. Konschuh, C. Ertler, C. Ambrosch-Draxl, and J. Fabian, Phys. Rev. B **80**, 235431 (2009)...

- Rashba spin-orbit coupling from **external electric field** $\perp$ to the sheet.

  SARPES (Spin ans Angle Resolved Photoemission Spectroscopy) on Graphene/Au/Ni(111) structure: $\lambda_R \sim 4meV$

  Varykhalov at al., PRL **101**, 157601 (2008)

- For realistic systems: $\boxed{\Delta_{SO} \ll \lambda_R}$ $\Rightarrow$ We neglect ISO from the model.

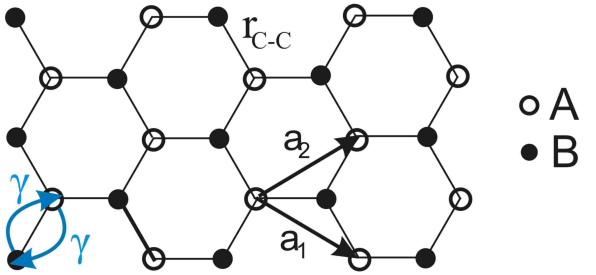
# Models of Rashba spin-orbit coupling (RSO) in graphene at the K point

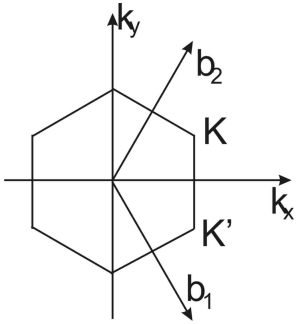
$$H_R^0 = \frac{3}{2}\lambda_R \left(\hat{\boldsymbol{\sigma}} \times \hat{\mathbf{s}}\right)_z$$

← **momentum independent** RSO based on symmetry considerations

Kane, C. L. and Mele, E. J., PRL **95**, 226801 (2005).

$H_R(\mathbf{k})$ from tight-binding model with longwave approximatioin:




$$H_R = \begin{pmatrix} 0 & 0 & 0 & -\mathbf{v}_\lambda \hat{\mathbf{p}}_+ \\ 0 & 0 & 3i\lambda_R & 0 \\ 0 & -3i\lambda_R & 0 & 0 \\ -\mathbf{v}_\lambda \hat{\mathbf{p}}_- & 0 & 0 & 0 \end{pmatrix} \qquad \Psi = \begin{pmatrix} A\uparrow \\ B\uparrow \\ A\downarrow \\ B\downarrow \end{pmatrix}$$

$$\hat{p}_\pm = \hat{p}_x \pm i\hat{p}_y \qquad \upsilon_\lambda = \frac{3\lambda_R r_{C-C}}{2\hbar}$$

- $\upsilon_\lambda \hat{p}_\pm \sim \lambda_R k$ are missing in Hamiltonian $H_R^0$ ($\lambda_R \ll \gamma$, $k \ll K$)
- Theese terms are responsible for trigonal warping (TW) effect (as in bilayer), and lead to $\mathbf{k}$-dependent bandsplitting.

# Mapping to Bilayer Graphene including trigonal warping (TW)

Bilayer with TW

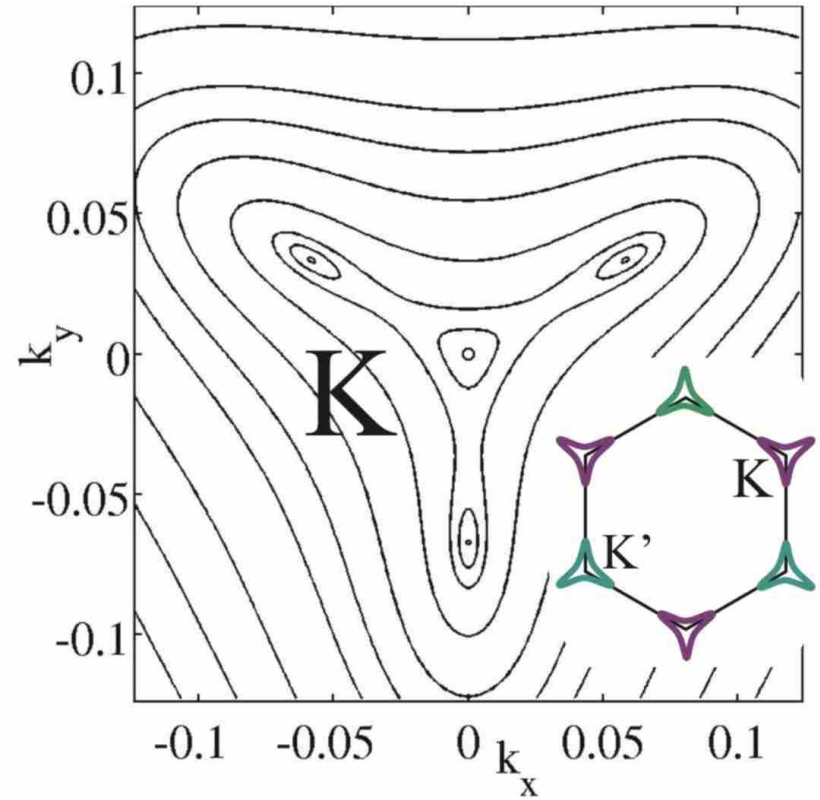
$$H_K^B = \begin{pmatrix} 0 & v_F\hat{p}_- & 0 & v_3\hat{p}_+ \\ v_F\hat{p}_+ & 0 & \gamma_1 & 0 \\ 0 & \gamma_1 & 0 & v_F\hat{p}_- \\ v_3\hat{p}_- & 0 & v_F\hat{p}_+ & 0 \end{pmatrix}$$

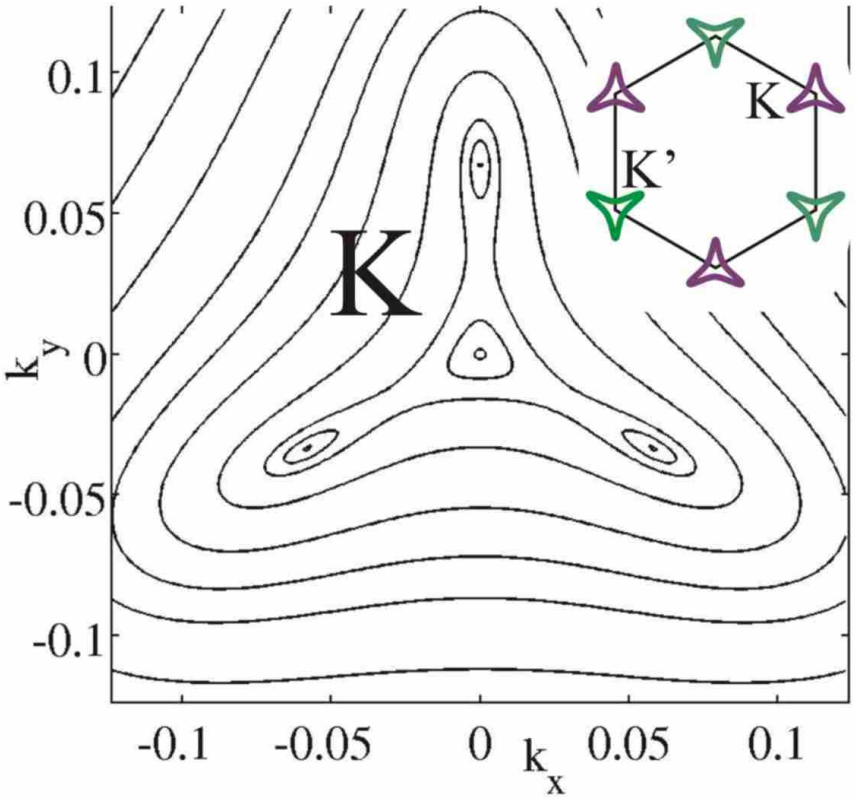
$$E^B(\gamma_1, k, \varphi)$$

Graphene with RSO

$$H_K = \begin{pmatrix} 0 & v_F\hat{p}_- & 0 & v_\lambda\hat{p}_+ \\ v_F\hat{p}_+ & 0 & -3\lambda_R i & 0 \\ 0 & 3\lambda_R i & 0 & v_F\hat{p}_- \\ v_\lambda\hat{p}_- & 0 & v_F\hat{p}_+ & 0 \end{pmatrix}$$

$$E(\lambda_R, k, \varphi) = E^B(3\lambda_R, k, \pi + \varphi)$$

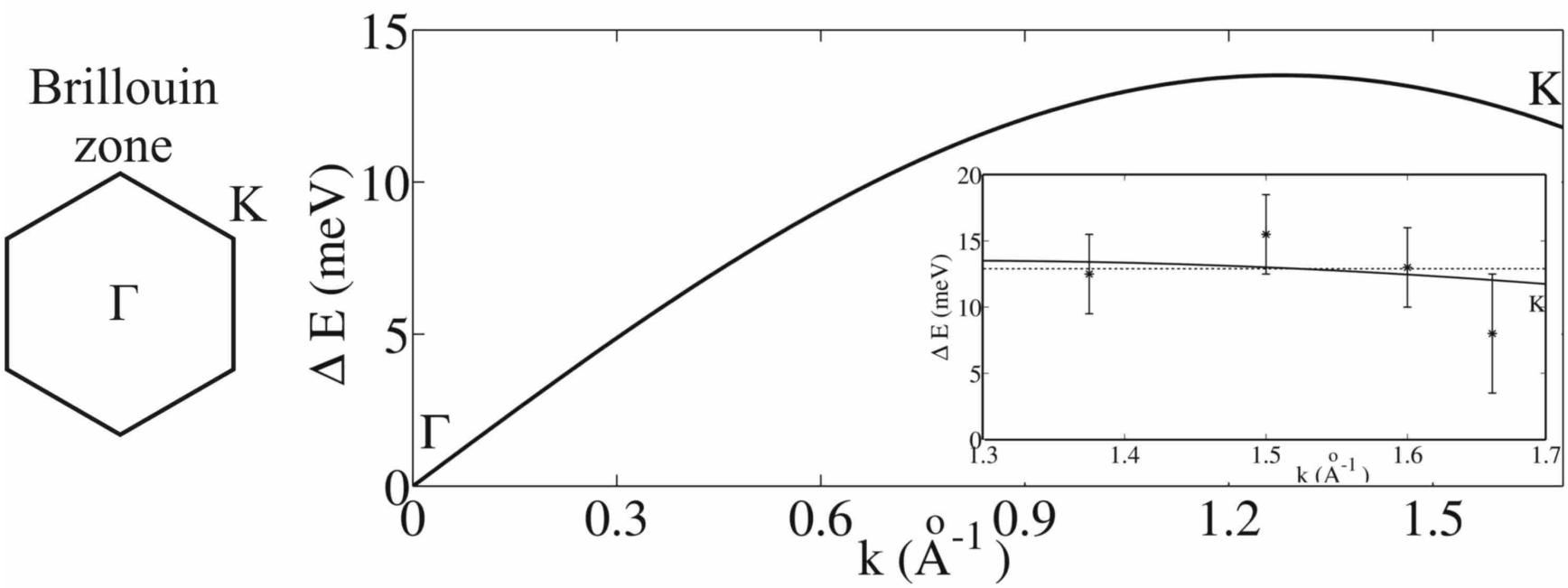


# k-dependent bandsplitting along $\Gamma K$

$\Delta E(\mathbf{k}) = E_2(\mathbf{k}) - E_1(\mathbf{k}),$ $E_{1,2}(\mathbf{k})$ are two valance bands

Solid lines: theory including TW Dashed line: theory without TW



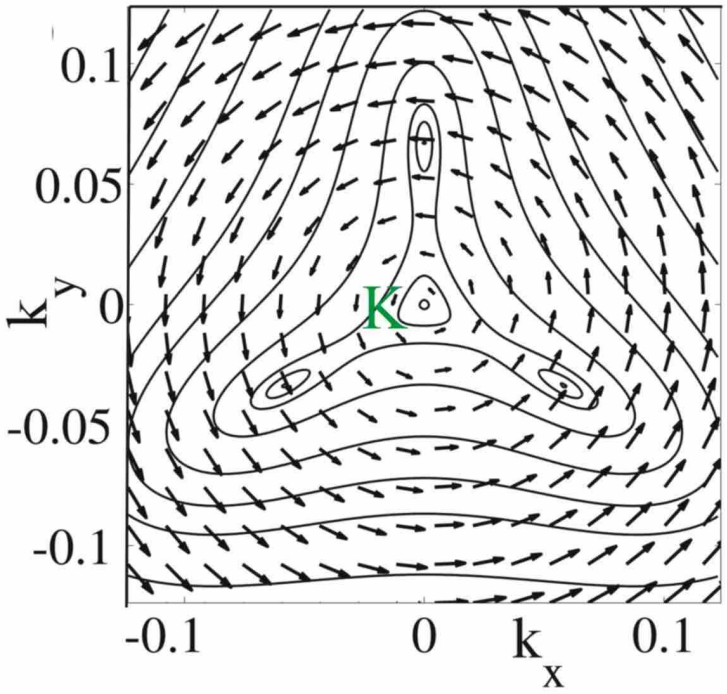

Errorbars:

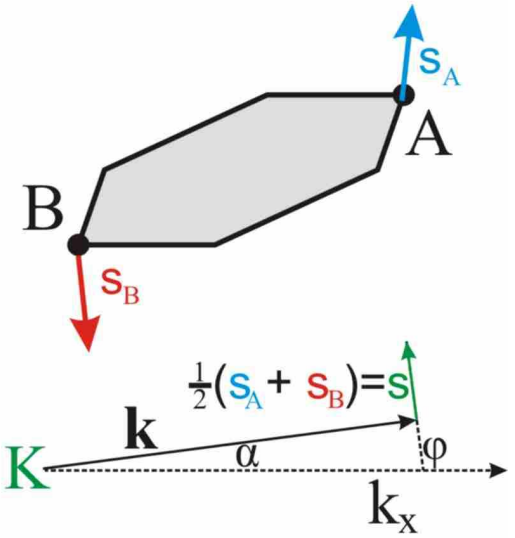
- Spin and Angle Resolved Photoemission Spectroscopy
- Graphene/Au/Ni(111) structure $\rightarrow$ quasifreestanding graphene (Varykhalov at al., PRL **101**, 157601 (2008).)

# Spin structure in the Brillouin zone

Energy contours in lower conduction band:

P. Rakyta, A. Kormányos, J. Cserti, PRB **82**, 113405 (2010).

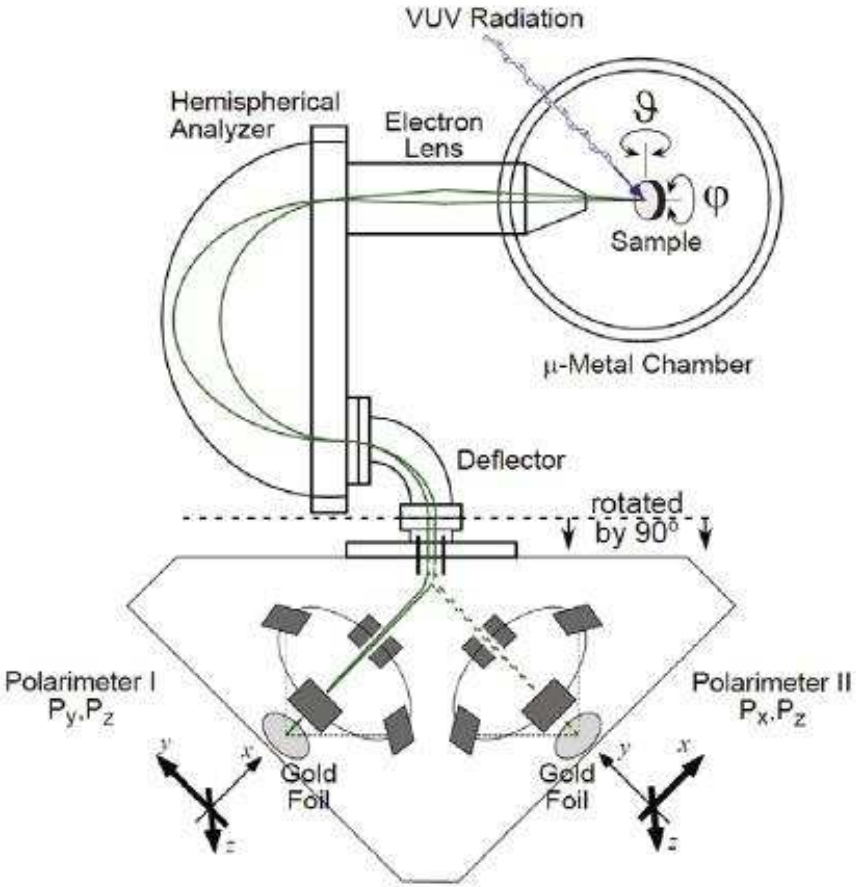



➠ The spin structure manifests rotational symmetry (TW leads to higher order correction):

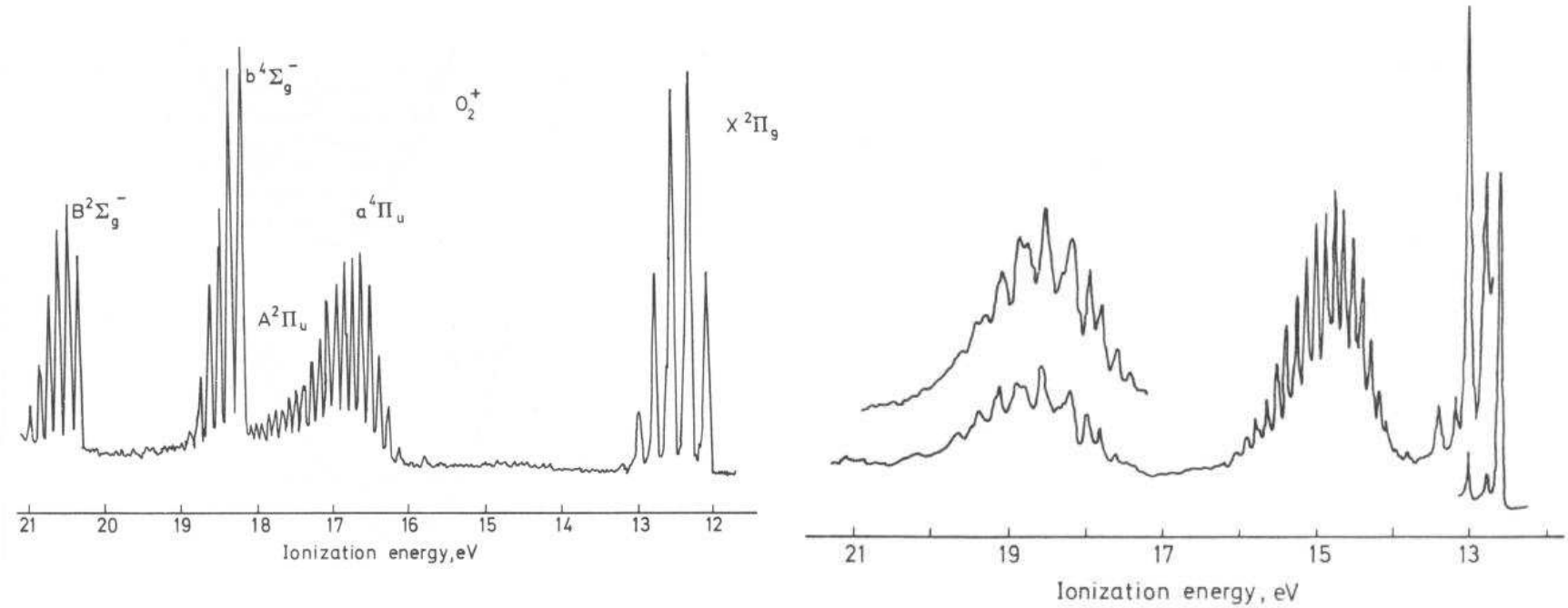
$$\varphi = \alpha + \frac{\pi}{2} - \frac{\cos 3\alpha}{2}|\mathbf{k}|r_{C-C}, \qquad \frac{|\mathbf{s}|}{\sqrt{(\hbar/2)^2 - \mathbf{s}^2}} = \frac{|\mathbf{k}|r_{C-C}}{\lambda_R/\gamma} - \frac{\sin 3\alpha}{2}\frac{\mathbf{k}^2 r_{C-C}^2}{\lambda_R/\gamma}$$

# Experimental equipment



# Ionization spectrum from Photoemission Spectroscopy

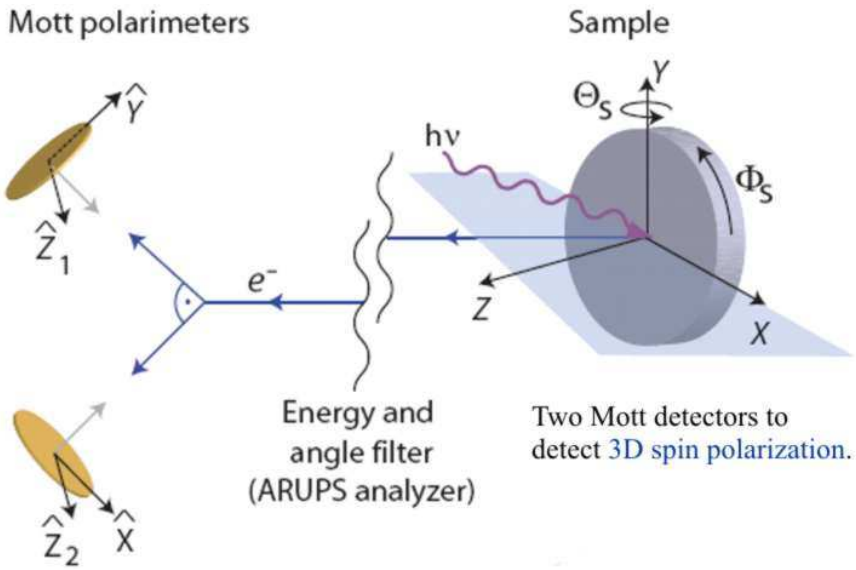
$O_2$ $H_2O$




- Finestructure: vibration modes of the ionizated molecules
- Photoemission Spectroscopy of solids: contributions from surface layers $\Rightarrow$ ideal for $2D$ systems.

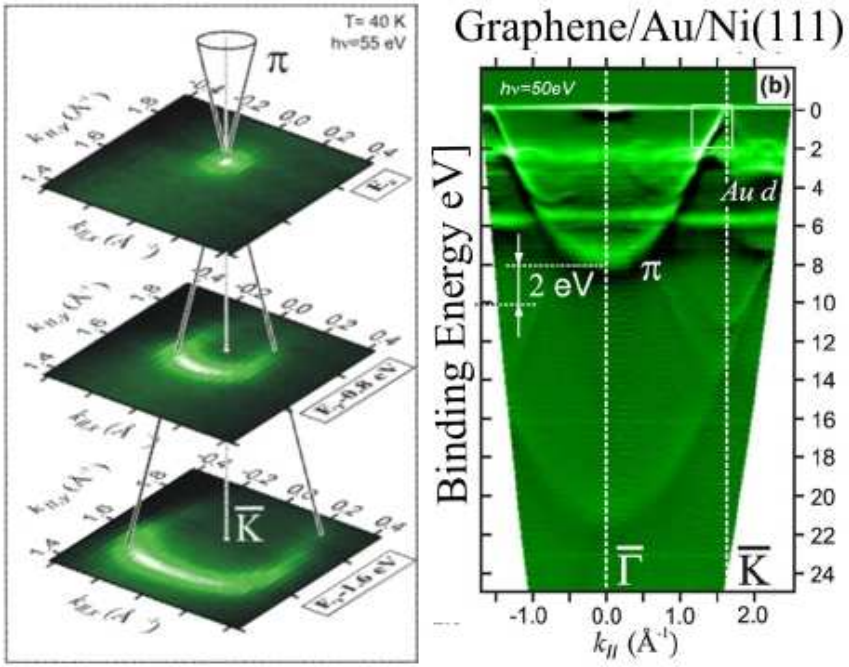
# Spin and Angle Resolved Photoemission Spectroscopy (SARPES)

F. Meier, J. H. Dill and J. Osterwalder, New Journal of Physics **11**, 125008 (2009)




- 2 detectors detect the spin-polarization in different planes.
- Restore 3D spin from the projections to the planes.

Varykhalov at al., PRL **101**, 157601 (2008)




- Anisotropic distribution of photoelectrons.
- Graphene/Au/Ni(111) - Quasifreestanding Graphene

# SARPES for graphene

➠ **Calculations based on Fermi's golden rule**:

Bloch-electron (band $\mu$) $|\mathbf{k},\mu\rangle$ in graphene + EM dipole interaction $H_{int} = -\frac{e}{m}\frac{\hbar}{\mathrm{i}}\boldsymbol{A}\nabla$

$\rightarrow$ Detected photoelectron with spin $\sigma$: $|\mathbf{p},\sigma\rangle = (H_{int})^{\mu\rightarrow\sigma}_{\mathbf{k}\rightarrow\mathbf{p}}\, e^{\mathrm{i}\mathbf{pr}/\hbar}|\sigma\rangle$

$$(H_{int})^{\mu\rightarrow\sigma}_{\mathbf{k}\rightarrow\mathbf{p}} \sim \mathbf{Ap}\left(\Psi^{\mu}_{A\sigma}(\mathbf{k}) + e^{\mathrm{i}\mathbf{G}\boldsymbol{\tau}}\Psi^{\mu}_{B\sigma}(\mathbf{k})\right)$$

with momentum and energy conservation.

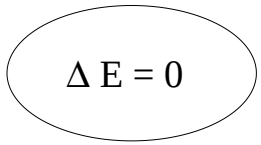
$\mathbf{G}$: reciprocal lattice vector, $\boldsymbol{\tau}$: $A \rightarrow B$ vector

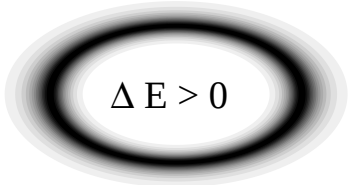
**Sublattice interference** in physical quantities:

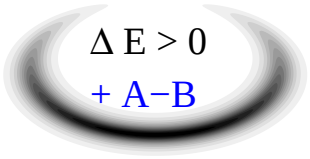
$$\langle\hat{O}\rangle_{\mathbf{p}} = \sum_{\sigma=\{\uparrow,\downarrow\}} \langle\mathbf{p},\sigma|\hat{O}|\mathbf{p},\sigma\rangle$$

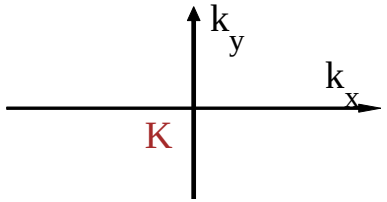
➠ Mucha-Kruczyński *et al.*, PRB **77**, 195403 (2008).

➠ Measurements on energy-contour with precision $\Delta E \Rightarrow$ bandstructure

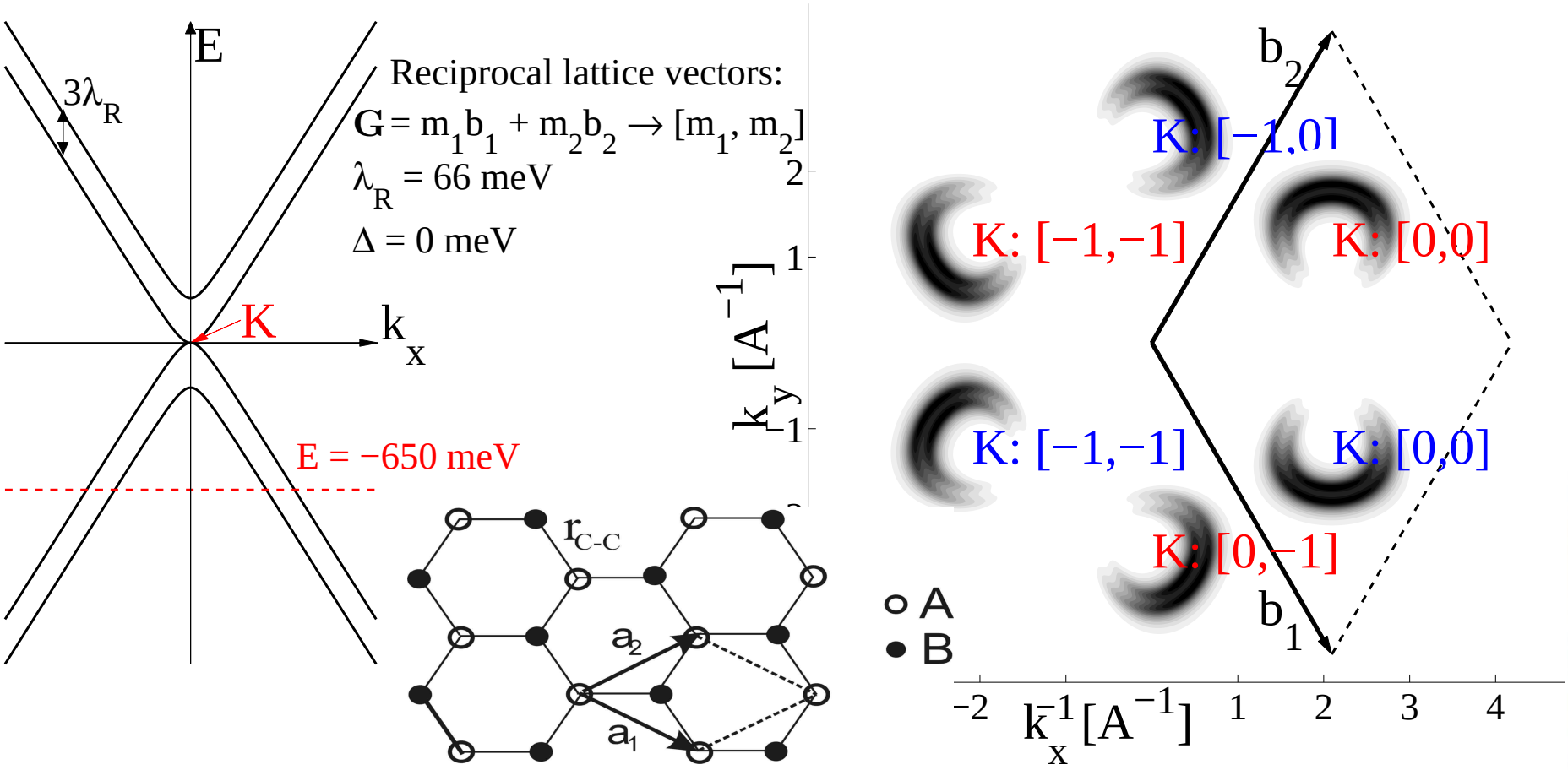


# Intensity of photoelectrons in the Brillouin Zone




➠ Inequivalent $K$ and $K$ points (like $A - B$ sublattice)

# Graphene/Y/Ni(111) structures with different Y atoms

$Y = Au$

band structure of graphene is unaffected within experimental precision: ideal graphene + RSO coupling („Quasifree-standing graphene", Rashba spin splitting $\approx 13$ meV)

$Y = (Cu, Ag)$

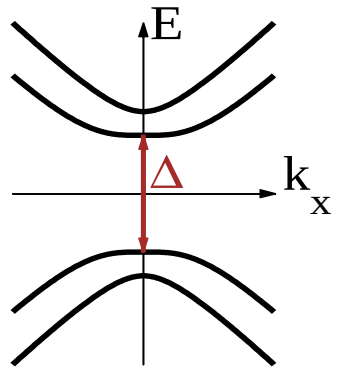
measured gap ($\sim 200$ meV) at the Dirac-point. The "ideal" model is not enough for description.

(Rashba spin splitting $\sim 100$ meV)

sublattice asymmetry:

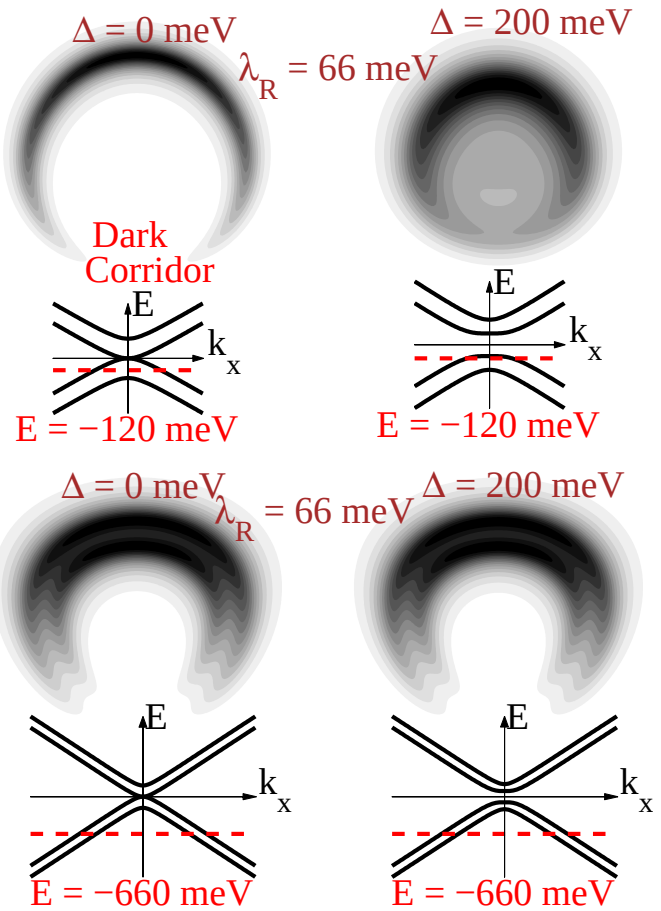
$$H_{AB} = \frac{\Delta}{2}(\sigma_z \otimes \hat{I}_2)$$

- different $A - B$ on-site energy
- not coupled to the spin
- gap opens at the Dirac-points



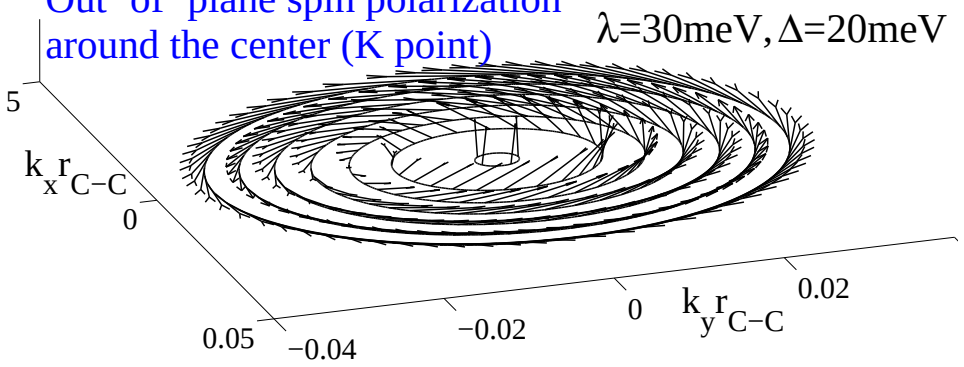
# Sublattice asymmetry in graphene with RSO

$$\langle s_z \rangle(\mathbf{k} \to 0) = \pm (1 - \delta_{0,\Delta}) \left(1 - \frac{\gamma^2}{2\lambda_R^2} k^2 r_{C-C}^2\right) \qquad \langle s_z \rangle(\mathbf{k} \to \infty) = \pm \frac{\Delta \lambda_R}{3\gamma^2 k^2 r_{C-C}^2}$$

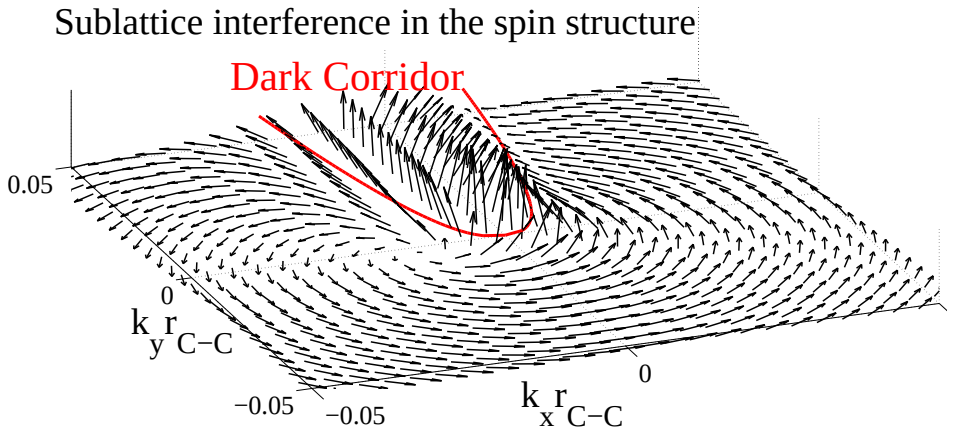



Spin structure around the K point:

Out−of−plane spin polarization around the center (K point)

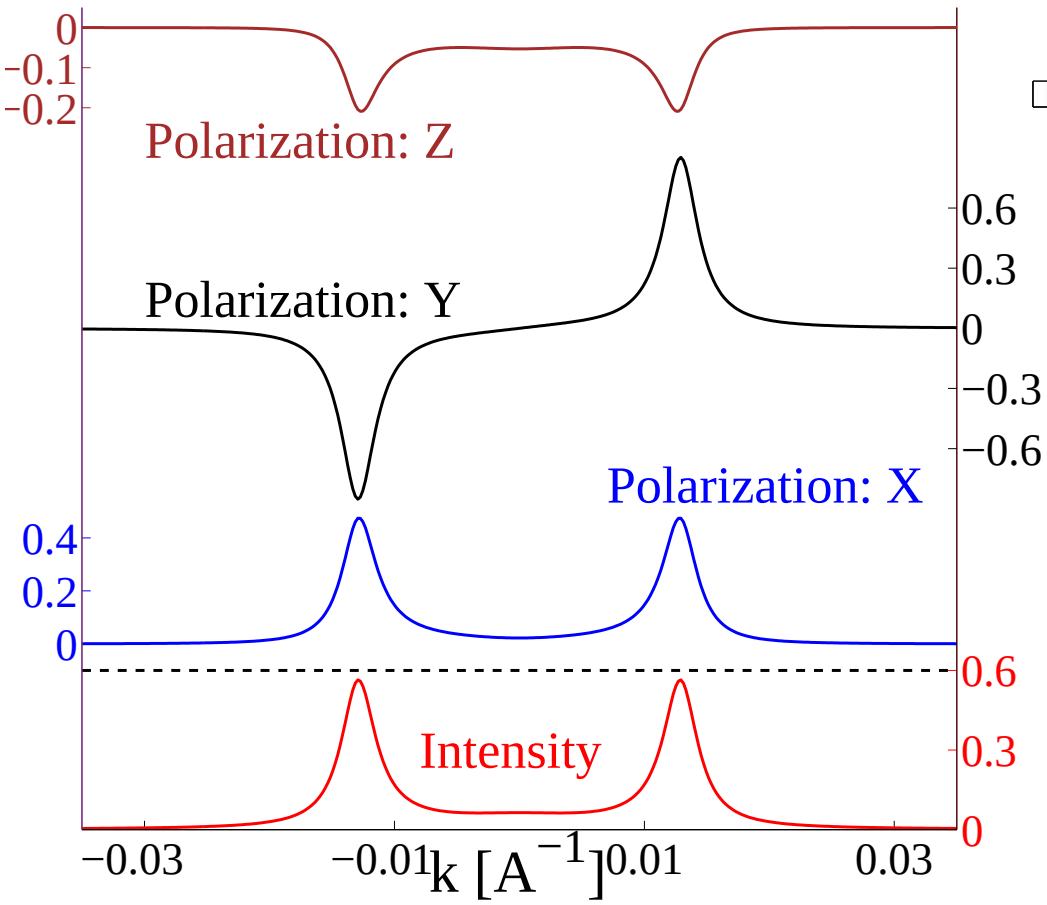


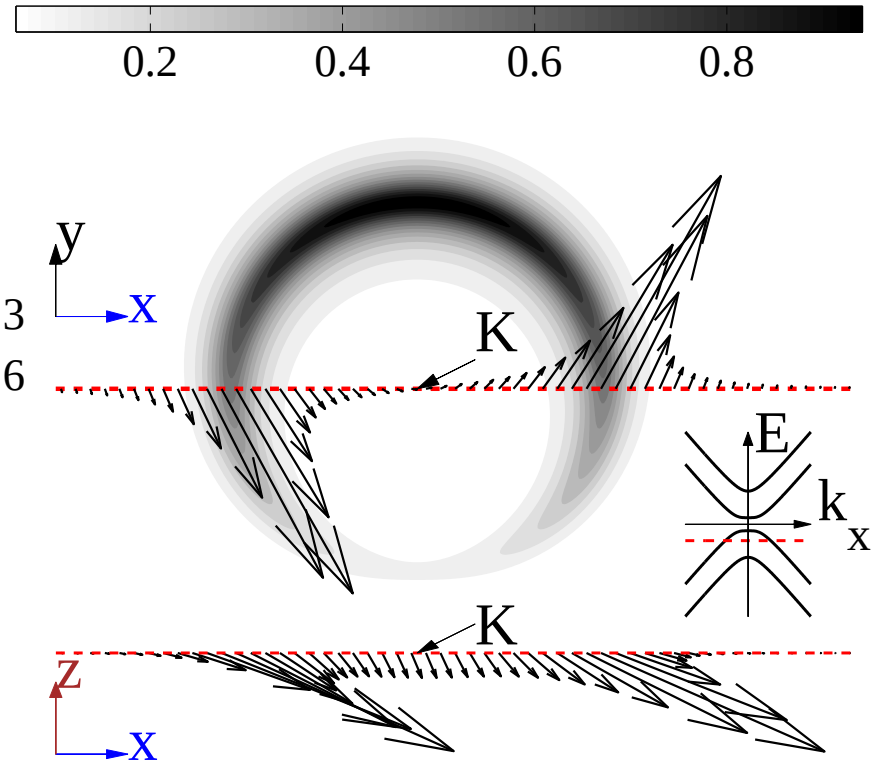

Sublattice interference in the spin structure




$\Delta = 0$ case: F. Kuemmeth, E. I. Rashba, PRB **80**, 241409(R), (2009).

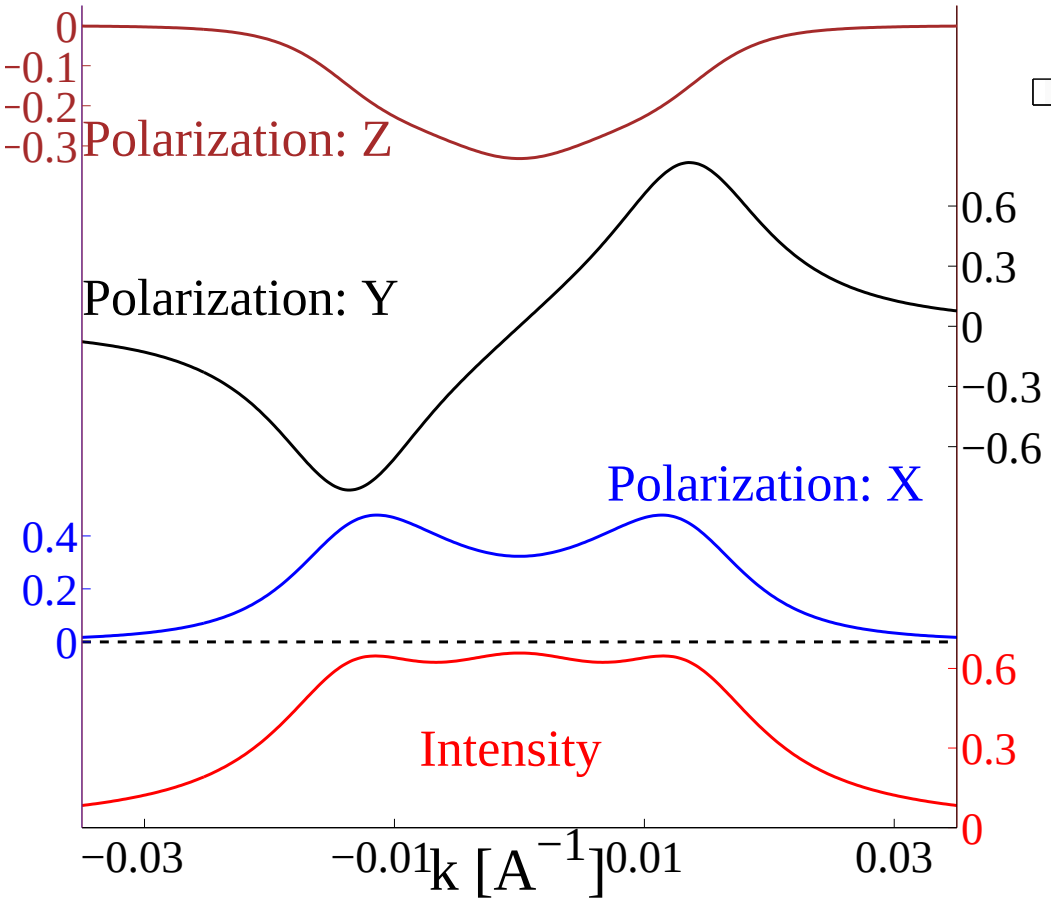
# Spin polarization in SARPES experiment

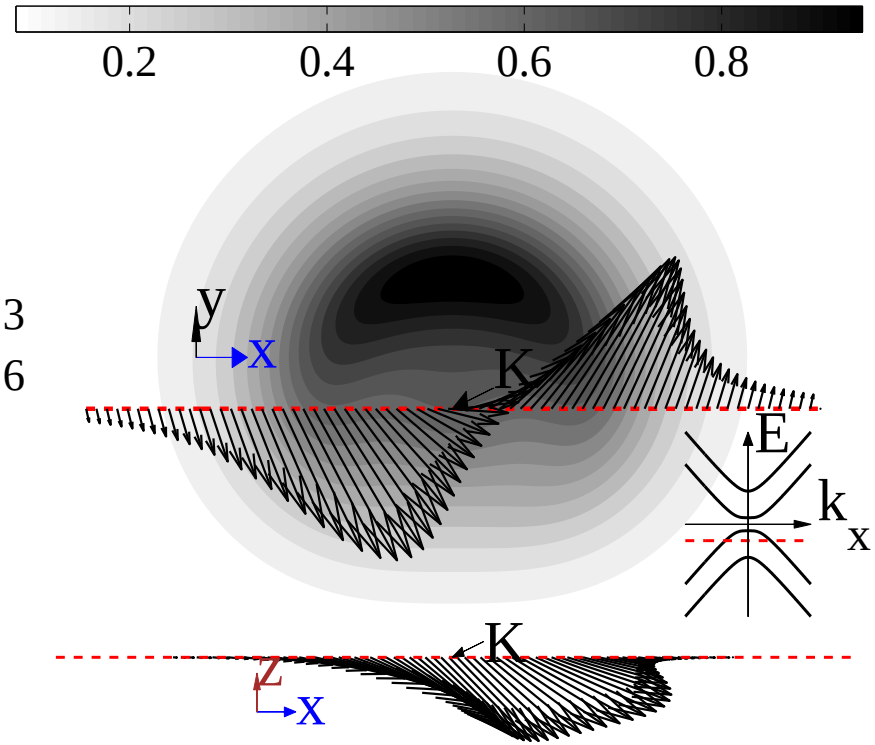



$$E = -50\text{meV}, \Delta = 40\text{meV}, \lambda_R = 33\text{meV}, \Delta E = 8\text{meV}$$

# Spin polarization in SARPES experiment




$$E = -50\text{meV}, \Delta = 40\text{meV}, \lambda_R = 33\text{meV}, \Delta E = 42\text{meV}$$

# Summary

- Anisotropic bandsplitting due to corrections of first order in Rashba spin-orbit coupling.
- SARPES experiments are heavily affected by sublatice interference:
- The intesity of the photoelectrons is minimal in the region called „the dark corridor”.

**Sublattice asymmetry:**

- Gap opens at the Dirac-point.
- Finite out-of-plane spin polarization can be measured in SARPES experiments over the dark corridor.

# Rashba spin-orbit coupling in tight-binding model (TB)

Lattice representation:

$$H_R^{TB} = i\lambda_R \sum_{\langle i,j\rangle,\mu,\nu} \left[ a_{i\mu}^{\dagger} \left( \hat{\boldsymbol{s}}_{\mu\nu} \times \frac{\mathbf{d}^{\langle i,j\rangle}}{d} \right)_z b_{j\nu} - h.c. \right]$$

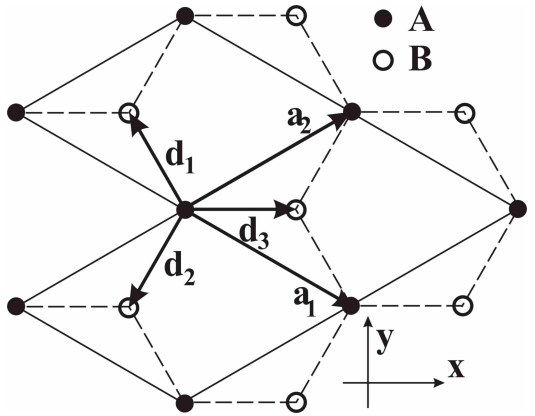
➠ $\lambda_R \sim E_z$; $\langle i,j\rangle$ nearest neighbours; $\mathbf{d}^{\langle i,j\rangle}$ points from site $j$ to $i$.

Momentum representation:

$$\mathbf{D}(\mathbf{q}) = -\sum_{j=1}^{3} e^{-i\mathbf{q}\mathbf{a}_j} \frac{\mathbf{d}_j}{d}$$

$$D_{\pm}(\mathbf{q}) = \pm D_x(\mathbf{q}) - iD_y(\mathbf{q})$$

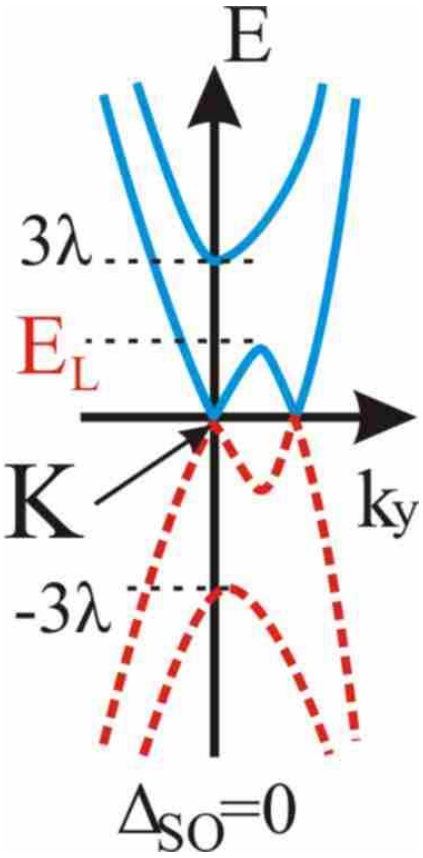
$$H_R^{TB}(\mathbf{q}) = \begin{pmatrix} 0 & 0 & 0 & -\lambda_R D_+(\mathbf{q}) \\ 0 & 0 & -\lambda_R D_-^*(\mathbf{q}) & 0 \\ 0 & -\lambda_R D_-(\mathbf{q}) & 0 & 0 \\ -\lambda_R D_+^*(\mathbf{q}) & 0 & 0 & 0 \end{pmatrix}$$




$$\Psi = \begin{pmatrix} A\uparrow \\ B\uparrow \\ A\downarrow \\ B\downarrow \end{pmatrix}$$

# The role of intrinsic SO: continous model

➠ $H = H_K + H_{SO}, \qquad H_{SO} = \Delta_{SO}\hat{\sigma}_z\hat{s}_z$




$k_x = 0$

Realistic parameters:

➠ $\boxed{\Delta_{SO} \sim 12\mu eV}$ (theory: first principles, DFT,

Gmitra, M. at al., Phys. Rev. B **80**, 235431 (2009).)

➠ $\boxed{\lambda_R \sim 4meV}$ (measurment: SARPES, Graphene/Au/Ni(111)

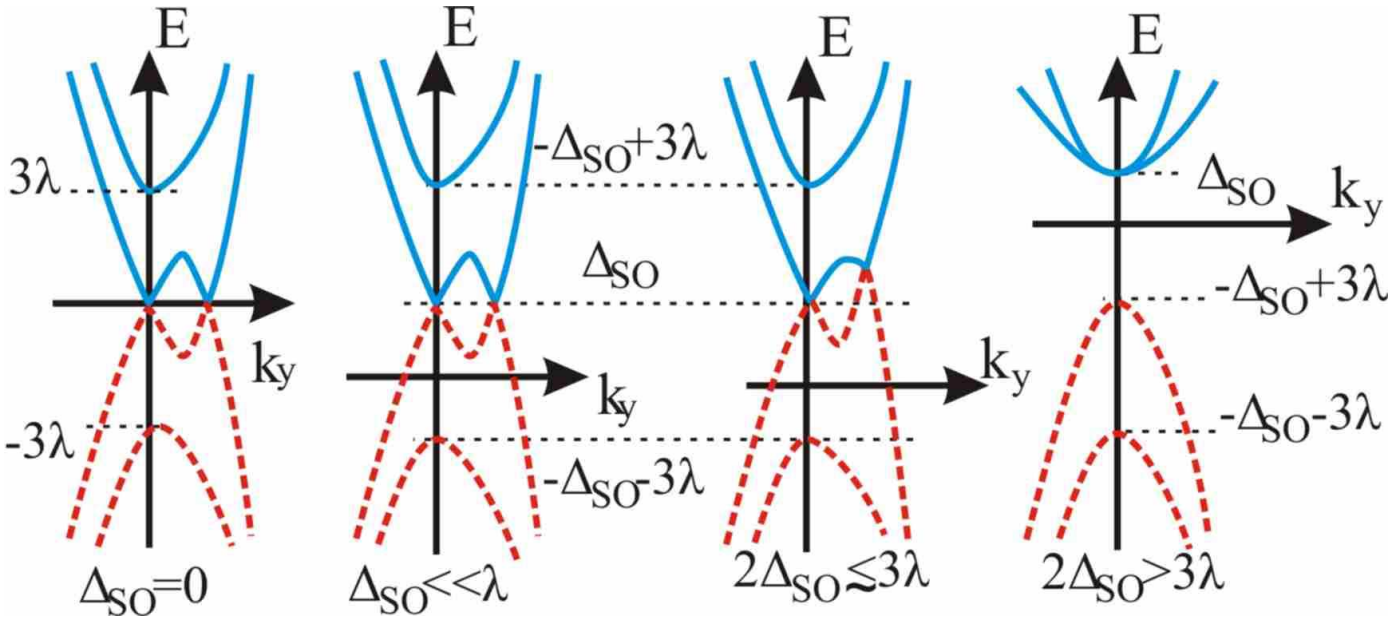
Varykhalov at al., PRL **101**, 157601 (2008). )

$$E_L \approx \frac{\lambda_R^2}{4\gamma}, \quad \boxed{E_L \approx 1.6\mu eV}$$

➠ The pockets are smaller than the ISO energy scale. ($E_L < \Delta_{SO}$)

# The role of intrinsic SO: continous model

$k_x = 0$



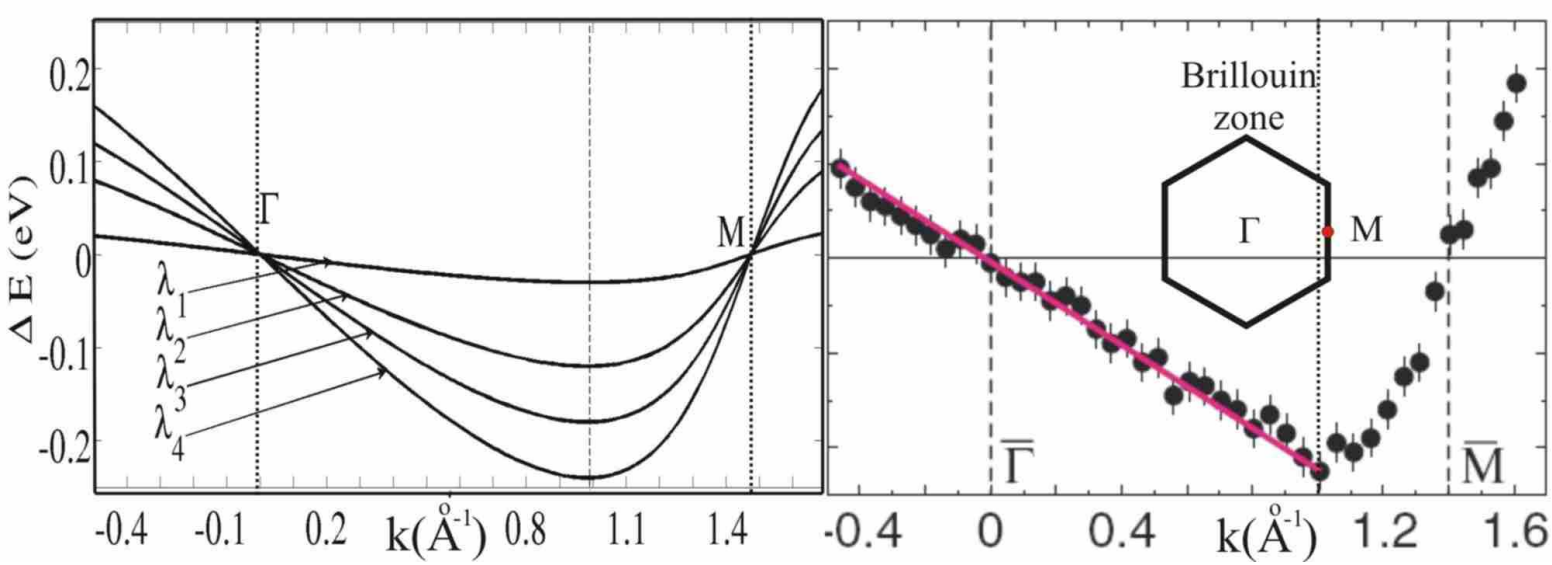

- Relevant case: $\Delta_{SO} \sim 12\mu eV \ll \lambda_R \sim 4meV$
- The pocket structure survives despite of finite ISO coupling.
- The dynamics of the electrons is affected by the TW: minimal conductivity: $\sigma_{\lambda_R} = 3\sigma_{\lambda_R=0}$ (as in bilayer: J. Cserti at al., PRL. **99**, 066802.)

# Spin splitting along $\Gamma M$

Solid lines: theory including TW

Experimental data

Dedkov at al., PRL. **100**, 107602 (2008).



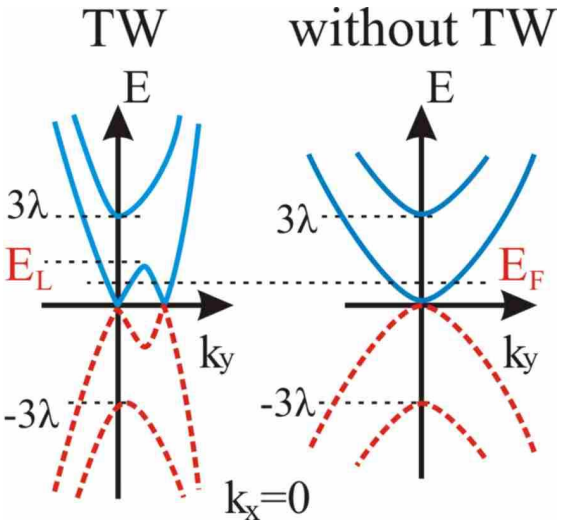

➠ Angle Resolved Photoemission Spectroscopy

Measurements:

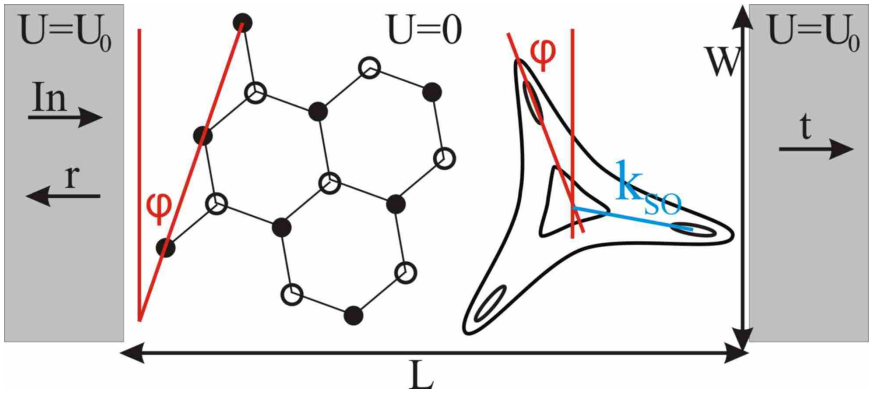
➠ Graphene/Ni(111) structure ($\lambda_R \approx 80\mathrm{meV}$)

➠ Rader at al.: The origin of the splitting is not clear PRL. **102**, 057602 (2009).

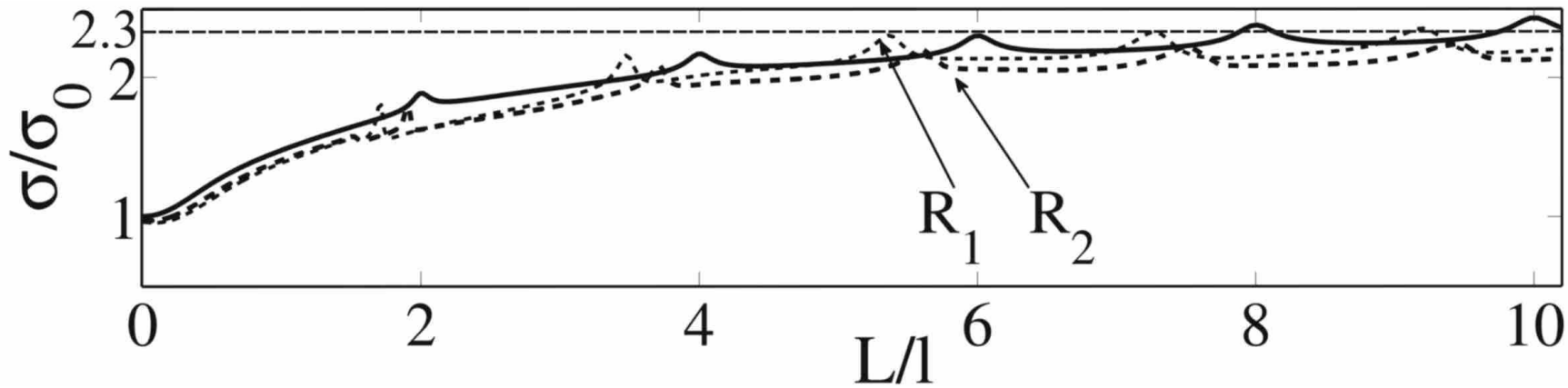
# Minimal Conductivity (MC)

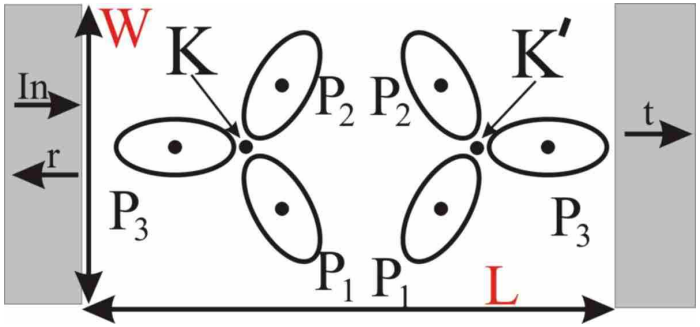



- MC: conductivity at $E_F = 0$.
- The MC is $3\times$ larger as the MC of the bulk without TW: $\boxed{\sigma = 3\sigma_0}$
- For realistic systems the interference between pockets leads to the anisotropy of the MC: $\boxed{\sigma = \kappa(\varphi)\sigma_0}$
  - $\sigma$: Landauer formalism in **Continous** ($W \to \infty$) and **tight-binding** (TB: finite $W$) model.
  - Characteristic length of RSO coupling: $l = \pi / k_{SO}$



# Minimal conductivity for Zig-Zag orientation ($\varphi = 30^0$)

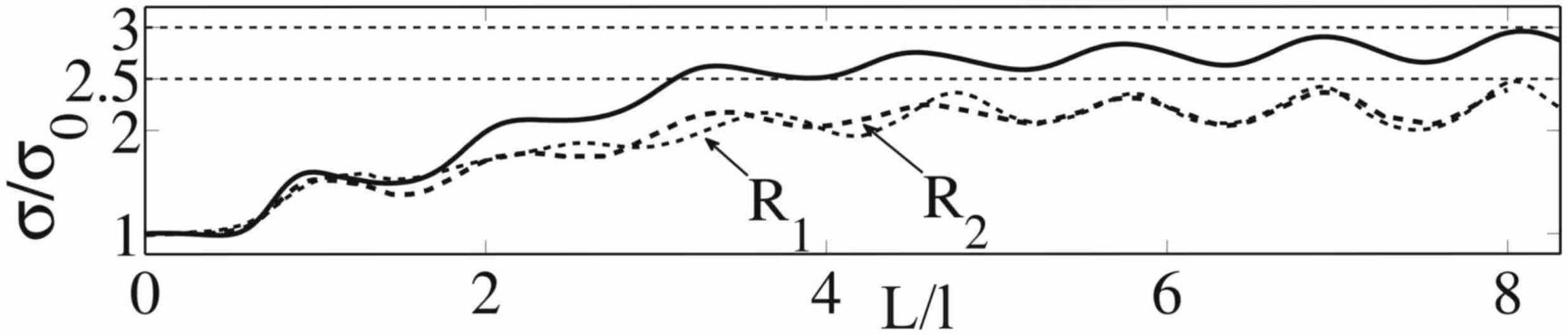


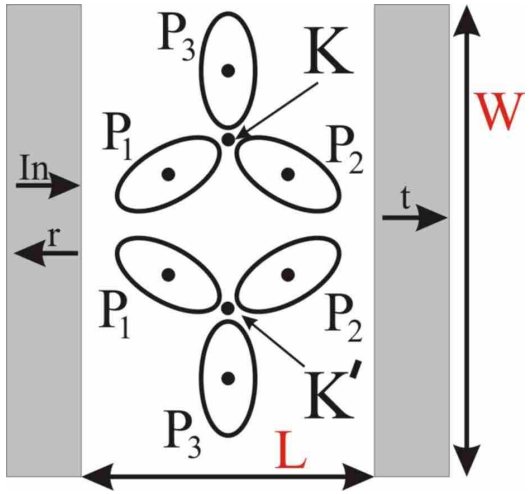




- Solid line: continous model ($W \to \infty$).
- Dashed lines: TB model for aspect ratio $R_1 = W/L = 4.71$ and $R_2 = 6.74$.
- Destructive interference between $P_3$ and $K \Rightarrow \sigma \approx 2(\sigma_{P_1} + \sigma_{P_2}) = 7/3\sigma_0$.

- Good agreement between Continous and TB results.

# Minimal conductivity for Armchair orientation ($\varphi = 0^0$)




- Solid line: continous model ($W \to \infty$)
- Dashed lines: TB model for aspect ratio $R_1 = W/L = 3.12$ and $R_2 = 5.80$
- Lower number of propagating modes in armchair nanoribbon due to boundary conditions.

$$\Rightarrow \sigma_{max}^{TB} = \sigma_{P_1} + \sigma_{P_2} + 2(\sigma_{P_3} + \sigma_K) = 2.5\sigma_0$$