A.M. Shikin
St.-Petersburg State University

Effects of anomalously large induced spin-orbit splitting in graphene under contact with heavy metals and their possible application in spintronics.

- Graphene. Electronic and spin structure
- Spin-orbit splitting of π-states of graphene induced by interaction with layer of metal (Au) with high atomic number intercalated underneath a graphene monolayer. Spin-dependent avoid-crossing effect
- Spin structure of graphene under contact with Cu, Bi, Pt.
- Possibility of topological phase formation
- Graphene and spintronics. Spin-orbit torque effect in magnetic nanostripes contacting with graphene.

Collaboration

St-Petersburg State University

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- Dr. A. Rybkin
- I. Klimovskikh
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- A. Rybkina
- E. Zhizhin
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- Prof. O. Tereshenko
- V. Golyahov

Tomsk and San Sebastian University

- Prof. E. Chulkov
- Dr. S.V. Eremeev
- Dr. S. Tsirkin

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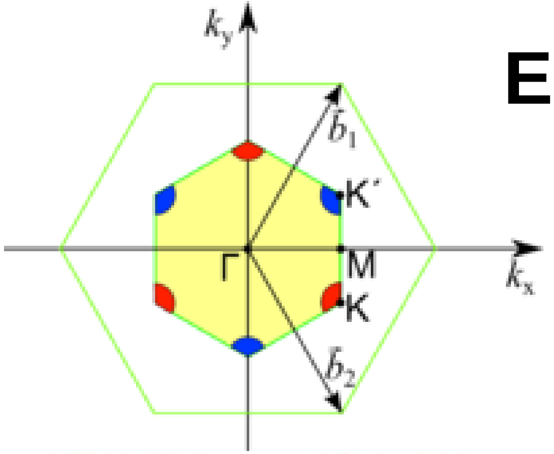
- Dr. T. Balusubramanian
- Dr. M. Leandersson
- C. Polley

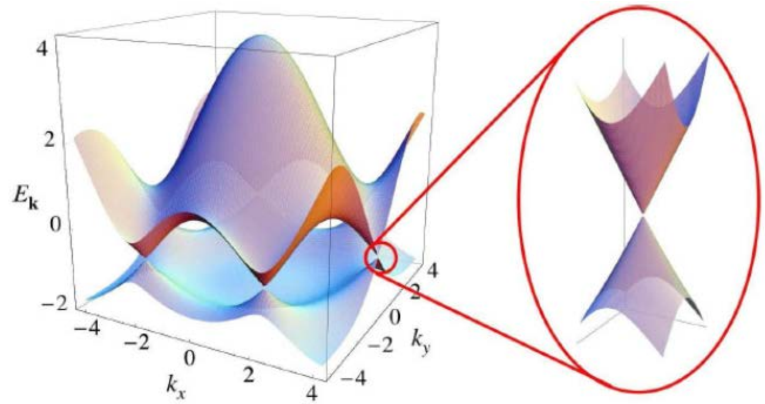
Free-standing graphene.
Electronic and spin structure.
Dirac cone states.

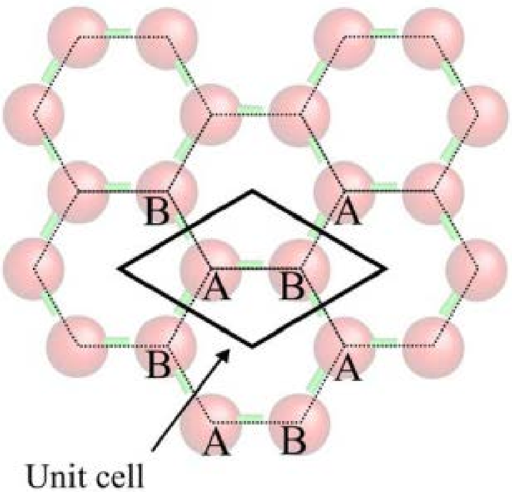
Electronic and crystalline structure

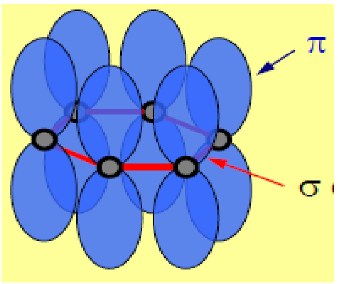


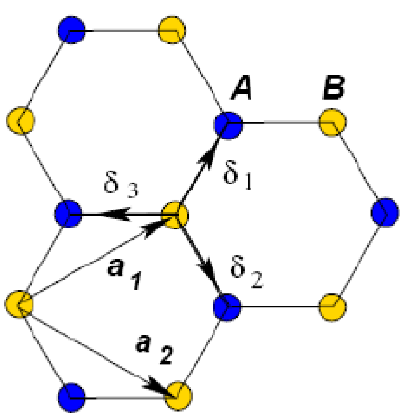
$$\psi = \phi_1 \pm \lambda \phi_2$$

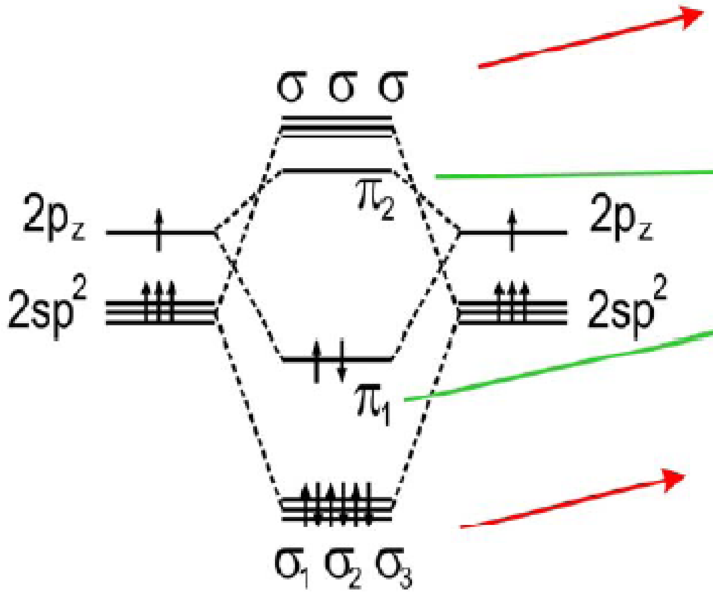
$$E = \pm \hbar \cdot V_F \cdot k$$

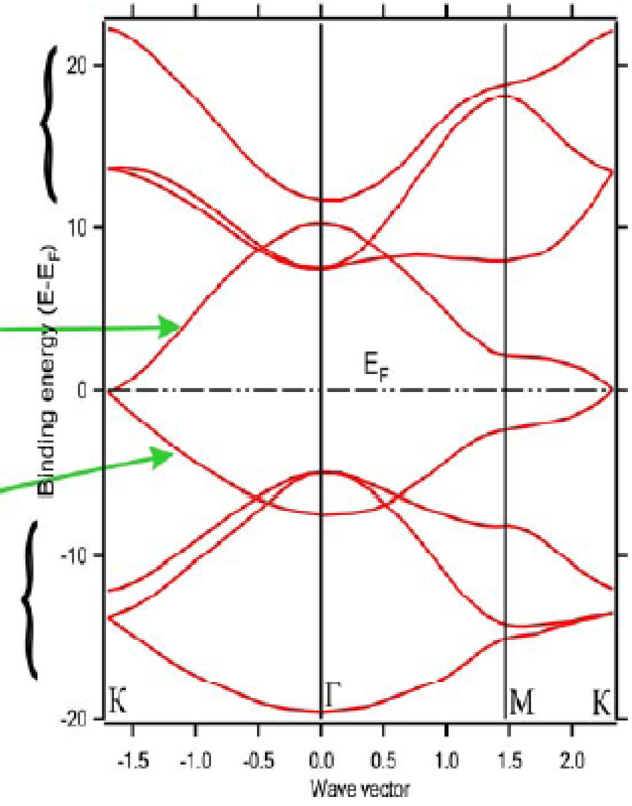




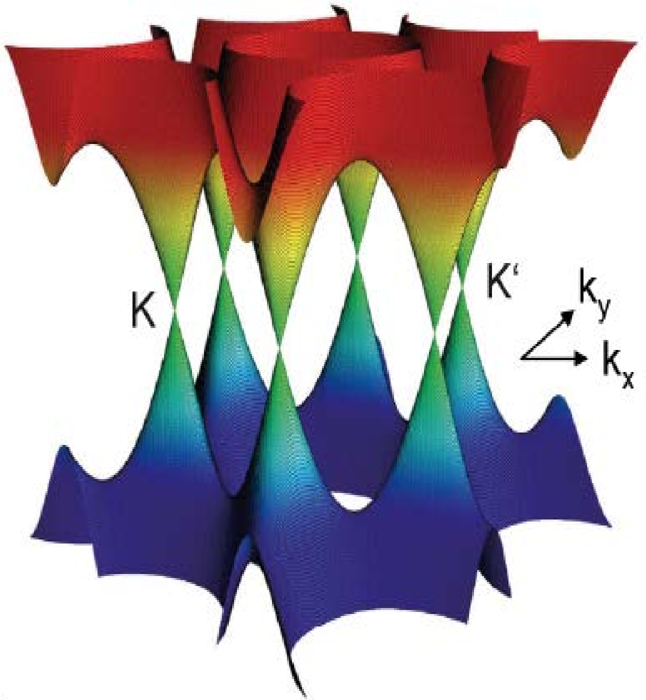




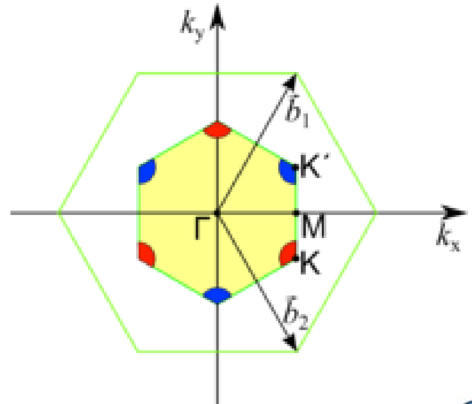


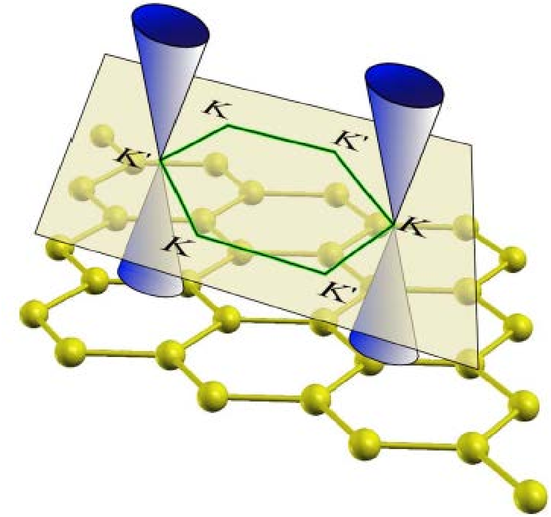


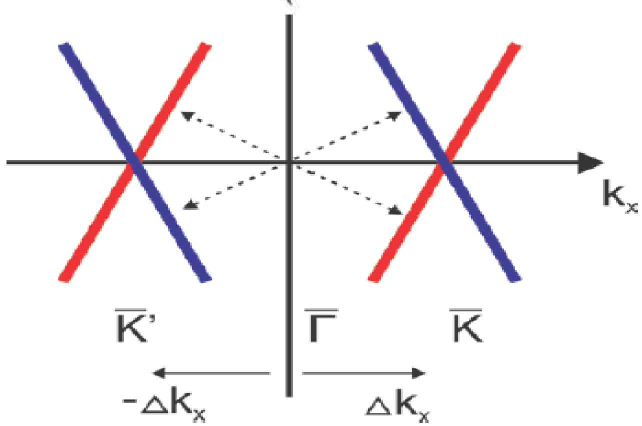
Graphene.
Dirac cone states.

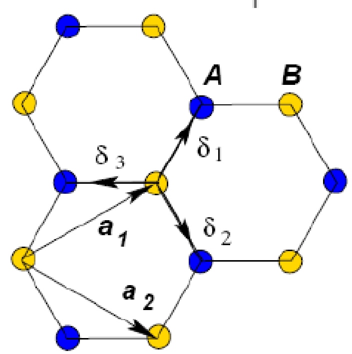


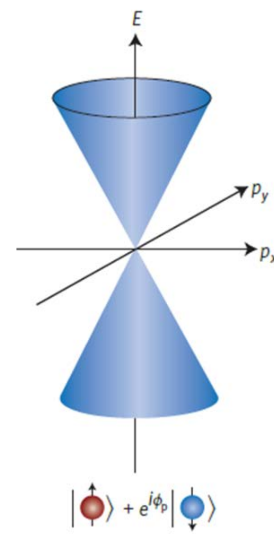
$$E = \pm \hbar \cdot V_F \cdot k$$

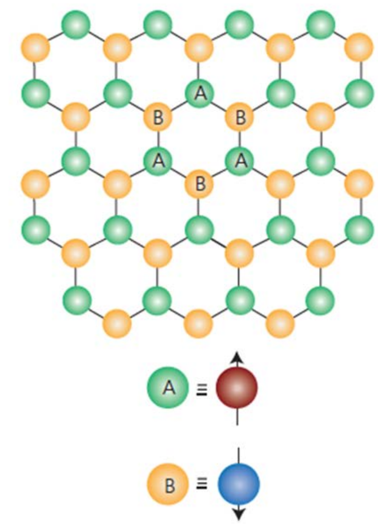












Interaction with neighbor atoms (at different sublattices) assumes the interaction between bonding π and antibonding π* states located at different K and K'

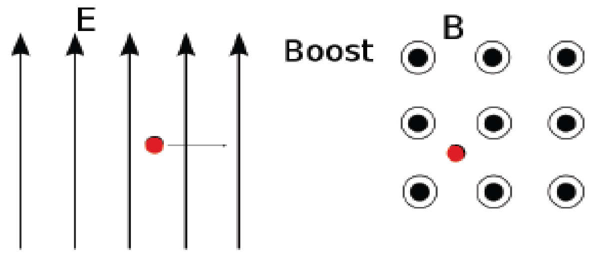
(It is important for hybridization and formation of topological phase)

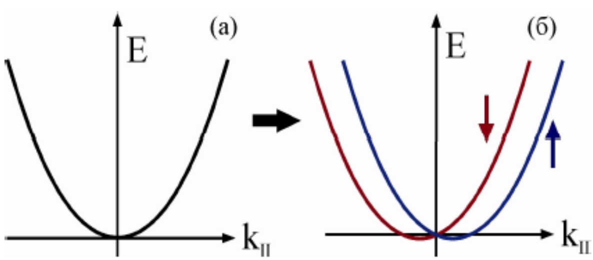
Transition from K to K' is followed by exchange between positive and negative k_{II}
Orientation of pseudospin is opposite for points K and K'.

Spin structure of graphene. Spins of individual electrons. 2D systems.

2D Rashba model for electron gas

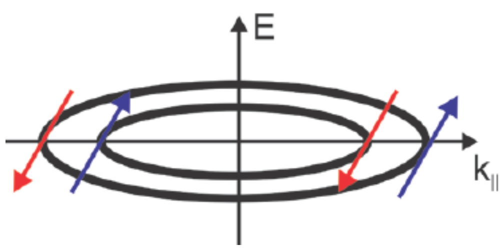
$$H_{SOC} = \alpha_R \left(\vec{\nabla} V \times \vec{k}_{\parallel} \right) \cdot \vec{\sigma}$$

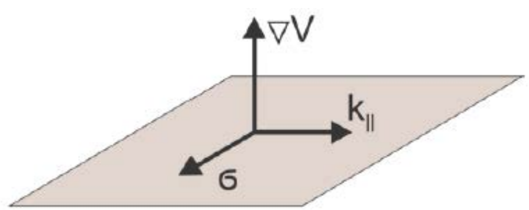




$$E = \frac{\hbar^2 k_{\parallel}^{\,2}}{2m} \pm \alpha_R \cdot k_{\parallel}$$

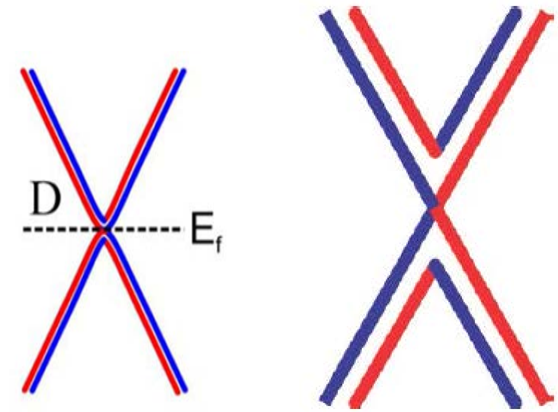
$\pm\Delta(k)_R$





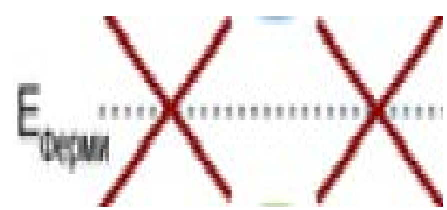
Spin is oriented in-plane and is locked perpendicular to momentum

Spin-orbit structure of graphene in accordance with Dirac equation



$$E = \pm\hbar \cdot V_F \cdot k \pm \Delta(k)_R$$

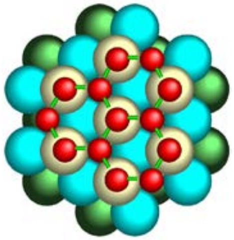
Spin-orbit coupling in graphene Is less than 100 μeV



K’ K

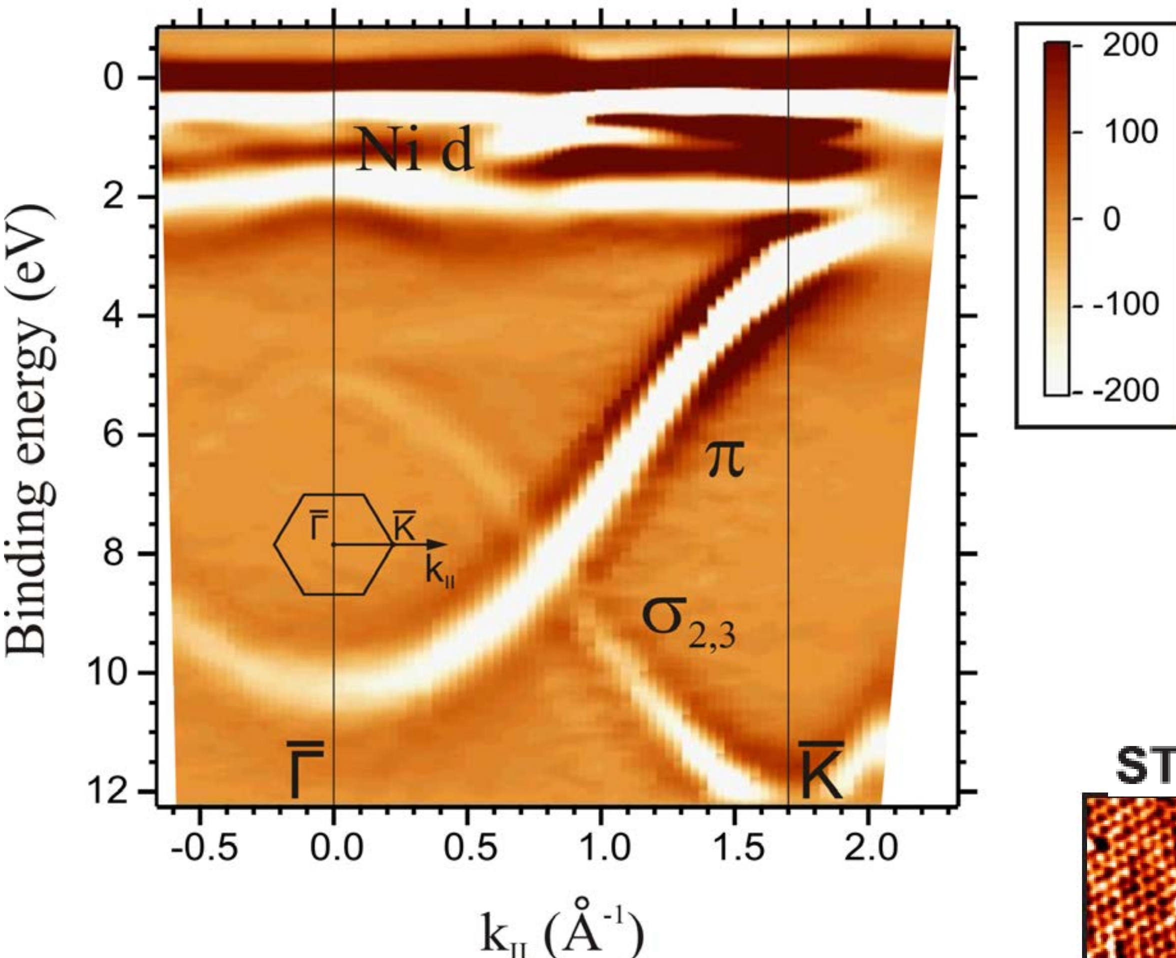
Formation of graphene.
Graphene intercalated by different metals.
Feature of electronic and spin structure.

Spin-orbit splitting of graphene pi-states induced under contact with heavy metals



Graphene on Ni(111). Electronic and atomic structure.

MG/Ni(111)/W(110)



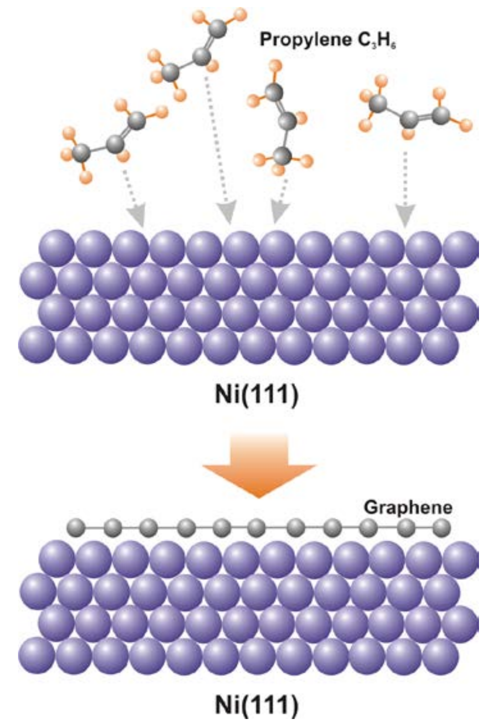
A. Varykhalov et al., Phys. Rev. Lett. 101, 157601 (2008)

O. Rader et al., Phys. Rev. Lett. 102, 057602 (2009)

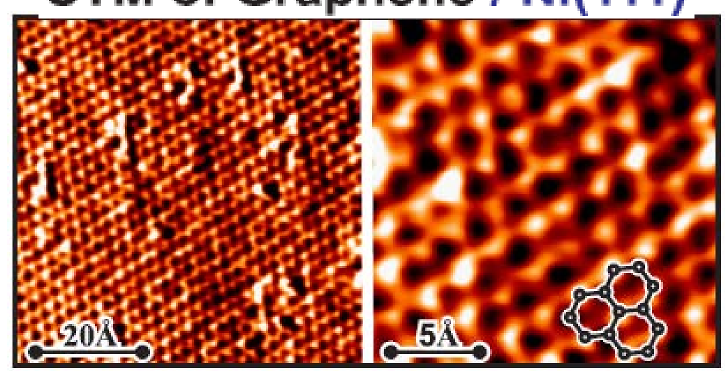
A. Popova et al., Phys. Solid State 153, 2539 (2011)

Synthesis of graphene by cracking of propylene (C3H6) at the Ni(111) surface

A.M. Shikin et al., Europhys. Lett. 44, 44 (1998)



STM of Graphene / Ni(111)

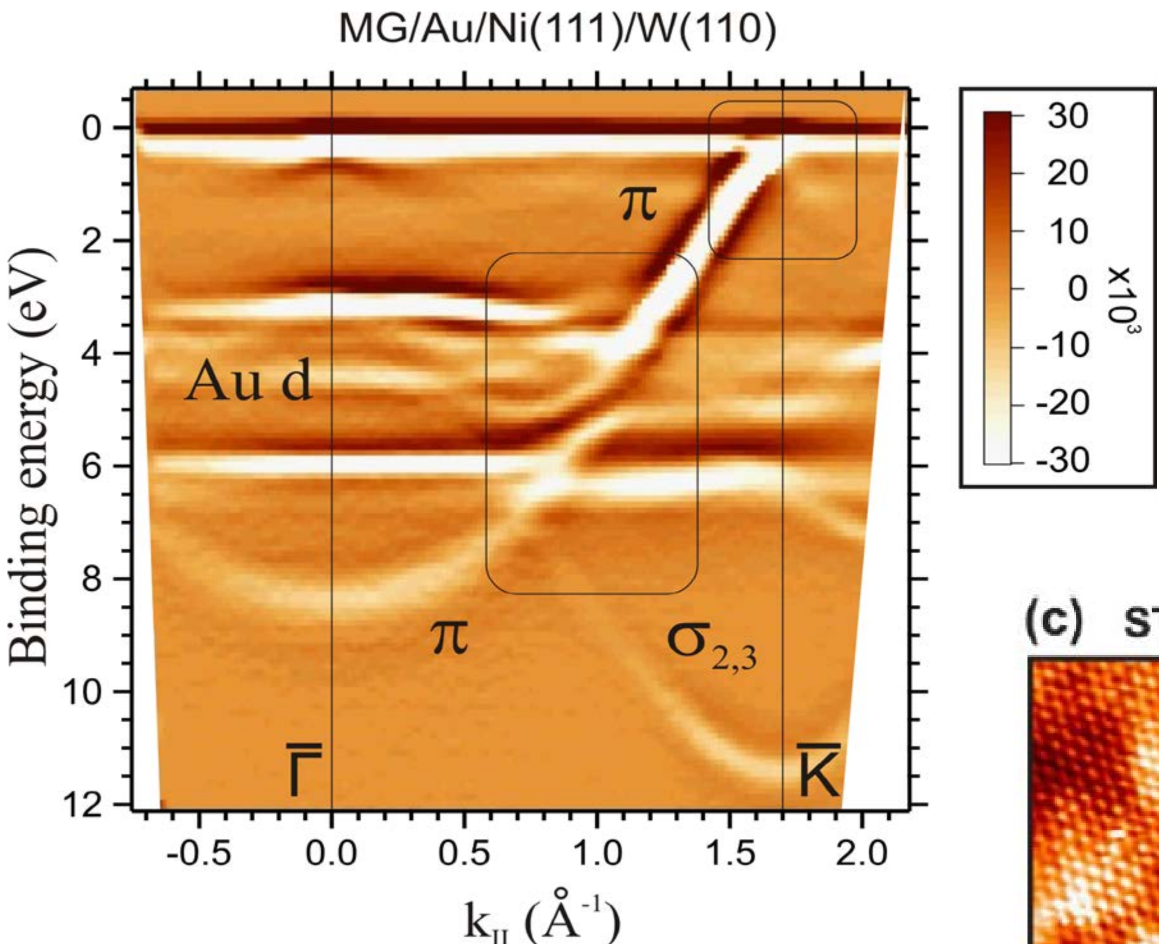


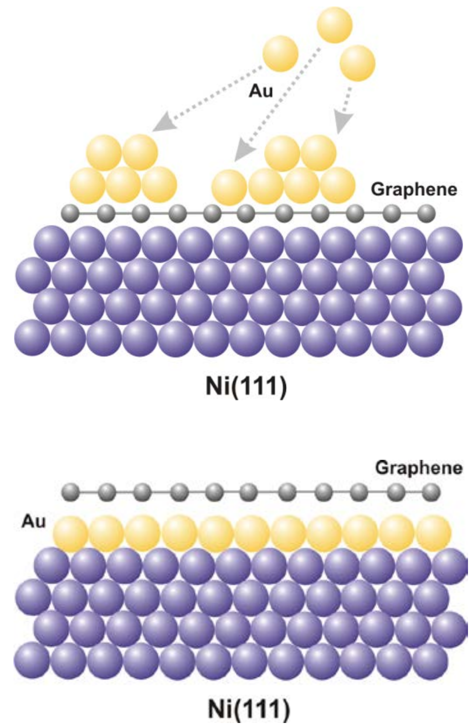
Graphene/Au

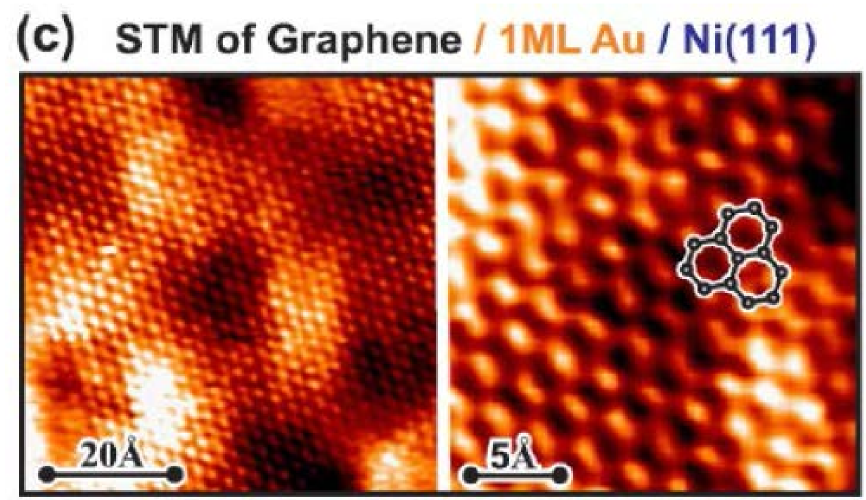
A.M. Shikin et al., Europhys. Lett. 44, 44 (1998)

A.M. Shikin et al., PRB 62, 13202 (2000)

Graphene on Ni(111) with intercalated monolayer of Au. Electronic and atomic structure.





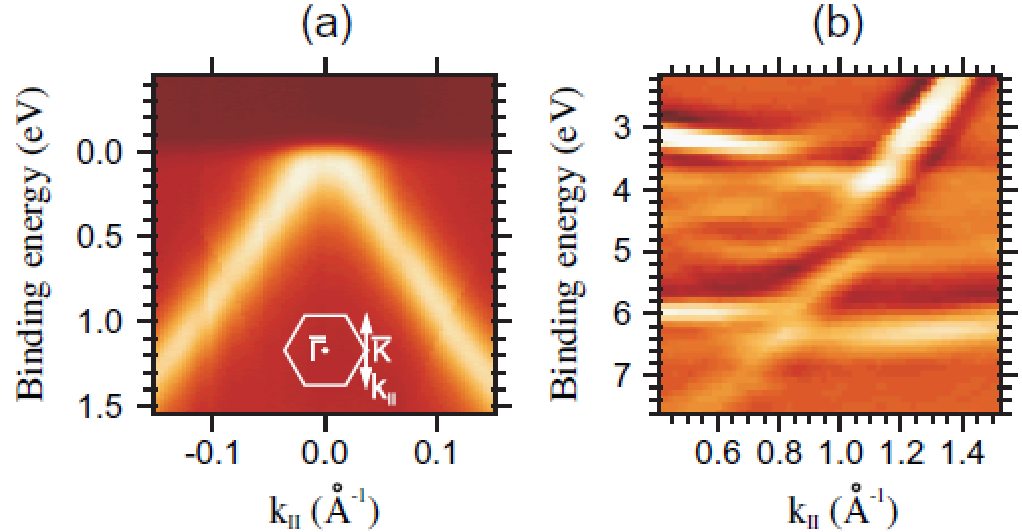


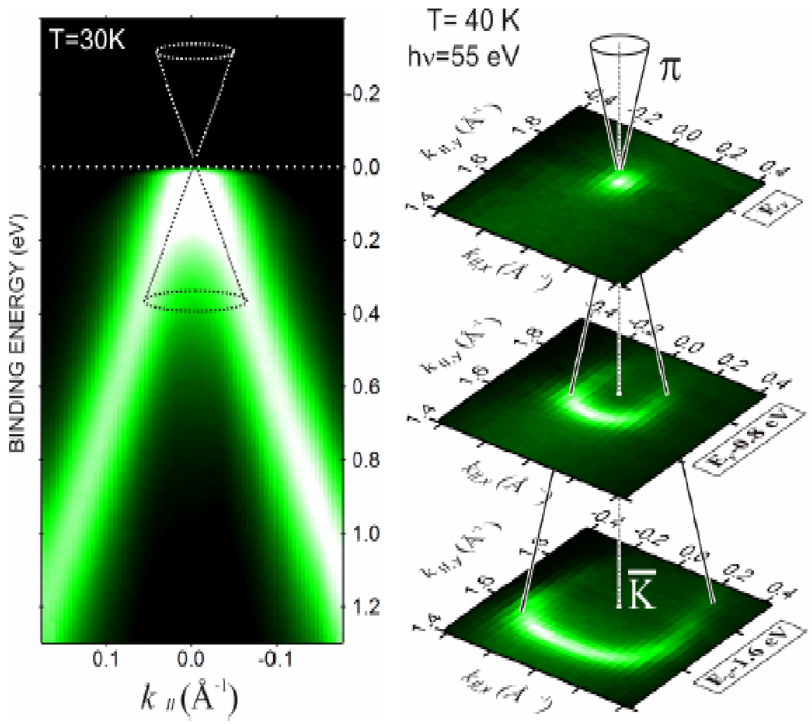
A.M.Shikin et al., New J. Phys. 15, 013016 (2013

A. Varykhalov et al., Phys. Rev. Lett. 101, 157601 (2008)

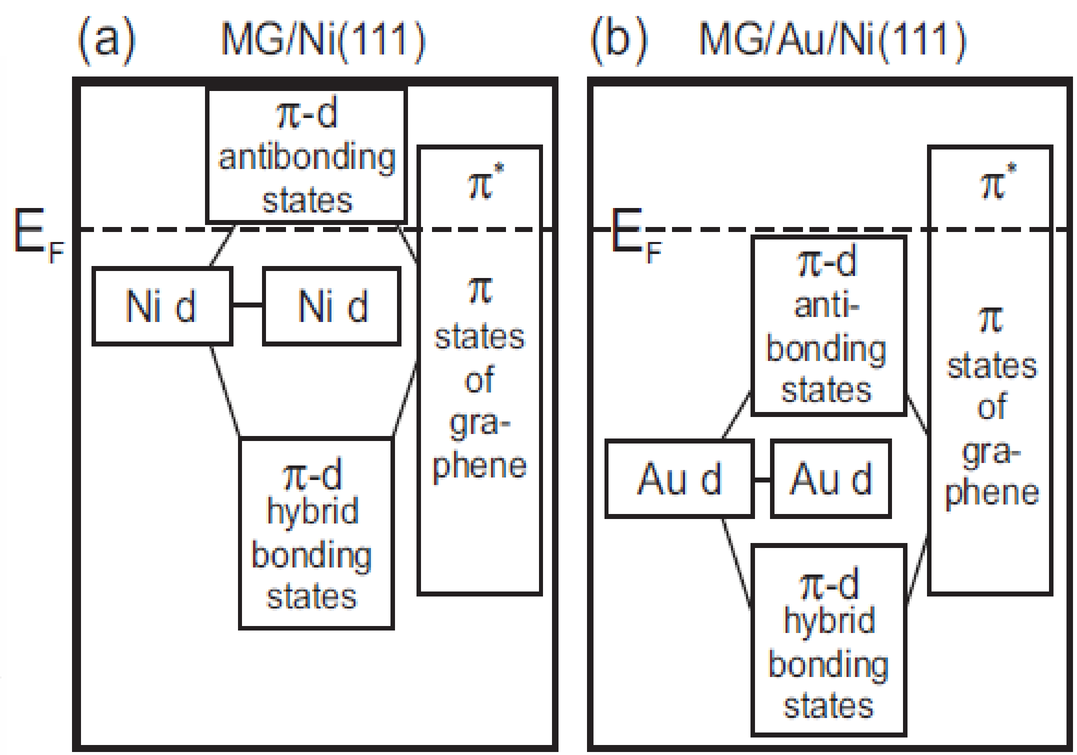
A. Popova et al., Phys. Solid State 153, 2539 (2011)

Schematic presentation of covalent interaction of graphene with underlying layers of Ni and Au. Formation and energetic localization of hybrid (π-d) states



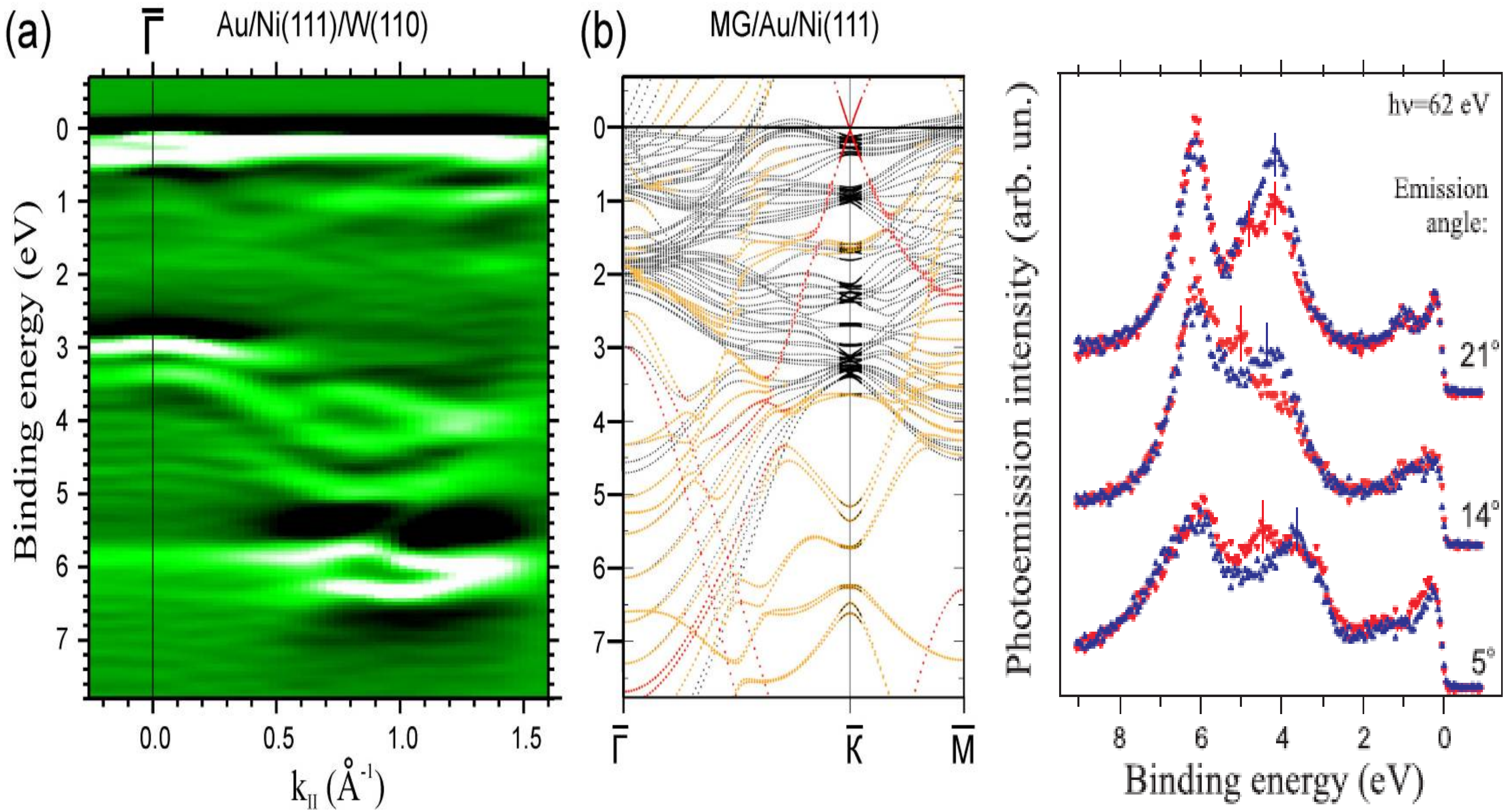


A. Varykhalov et al., Phys. Rev. Lett. 101, 157601 (2008)



A.M.Shikin et al., New J. Phys. 15, 013016 (2013)

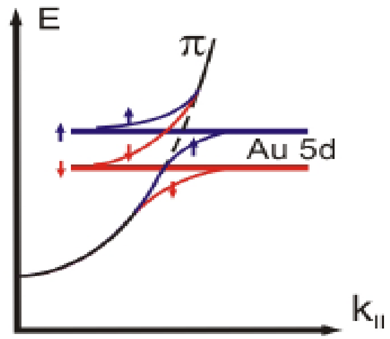
Spin splitting of Au d states.

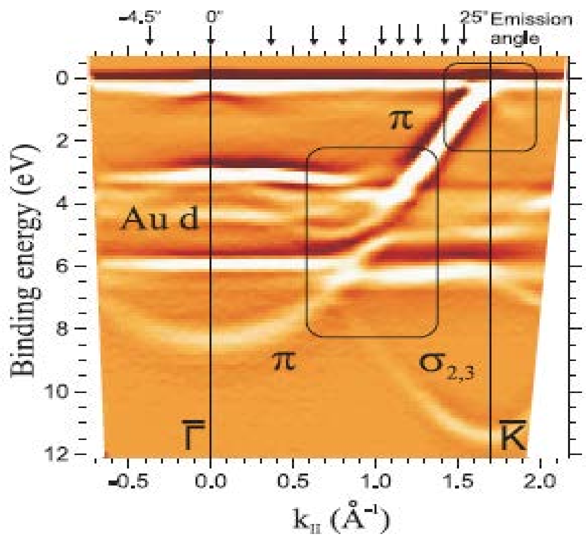


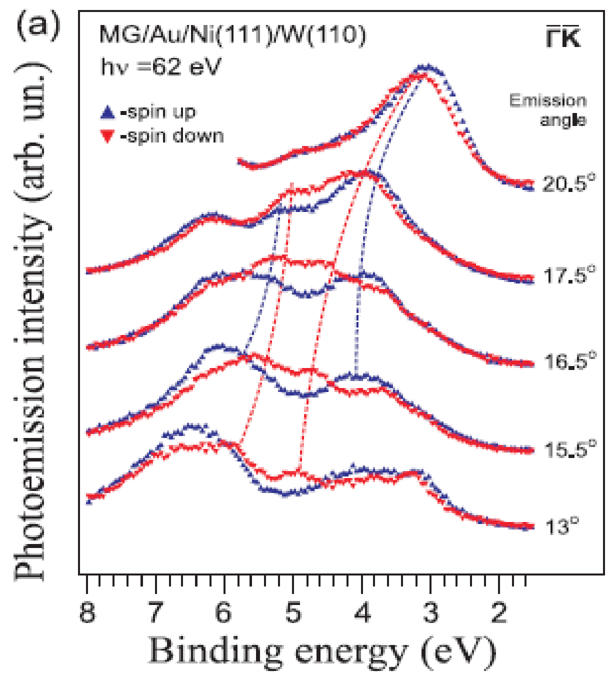
Hybridization between Au d and graphene states should be spin-polarized

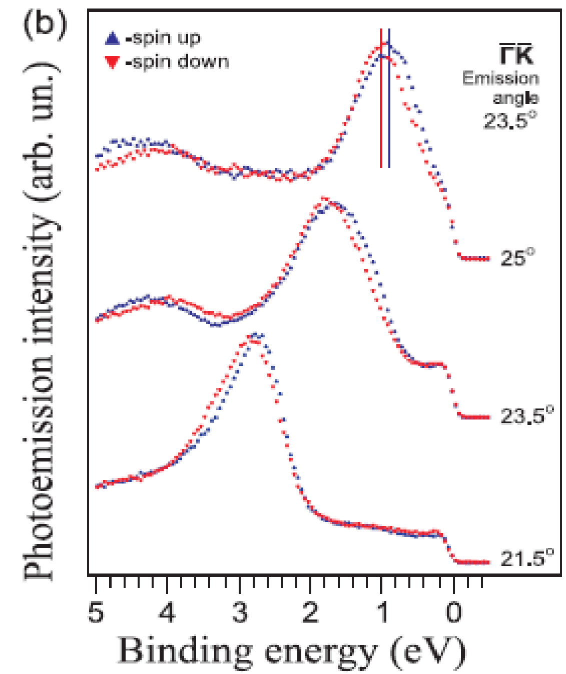
A.M.Shikin et al., New J. Phys. 15, 013016 (2013)

Modification of spin structure of π-states of graphene in the region of assumed intersection with Au d states and outside this region. Spin-dependent avoid-crossing effect





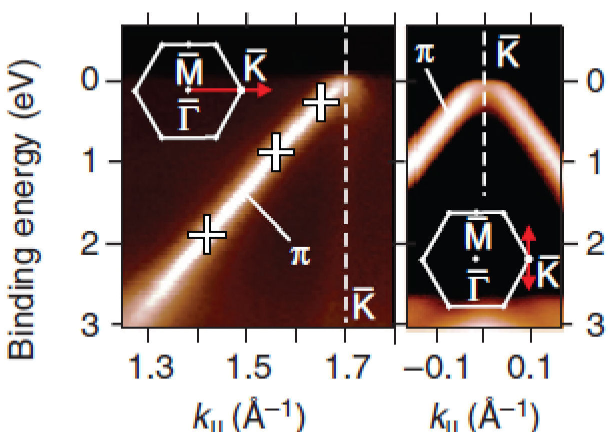


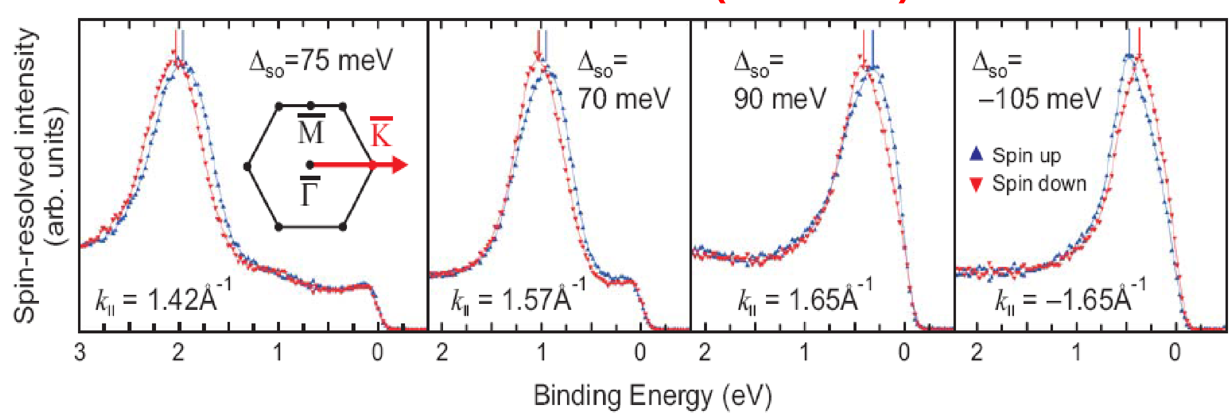


D.Marchenko et al., Nature Commun. 3, 1232 (2013)

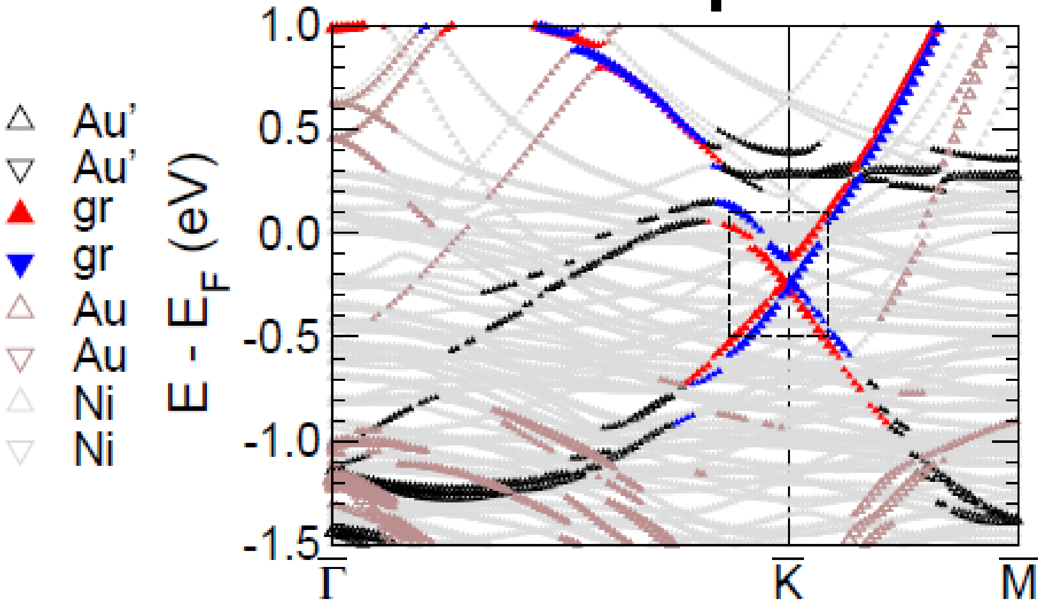
A.M.Shikin et al., New J. Phys.15, 013016 (2013)

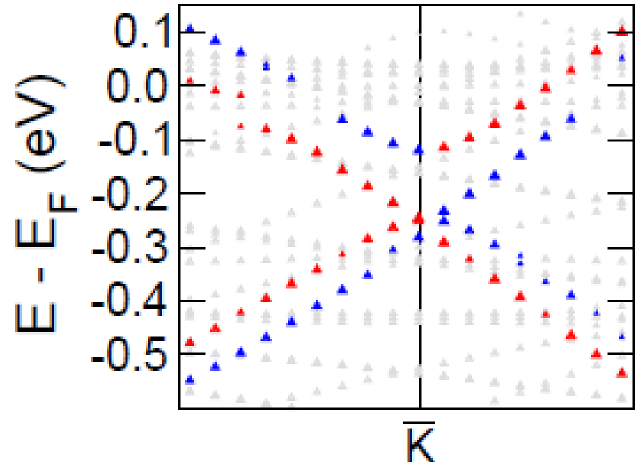
Enhanced spin-orbit splitting of the graphene π states near the Fermi level (~100 meV)

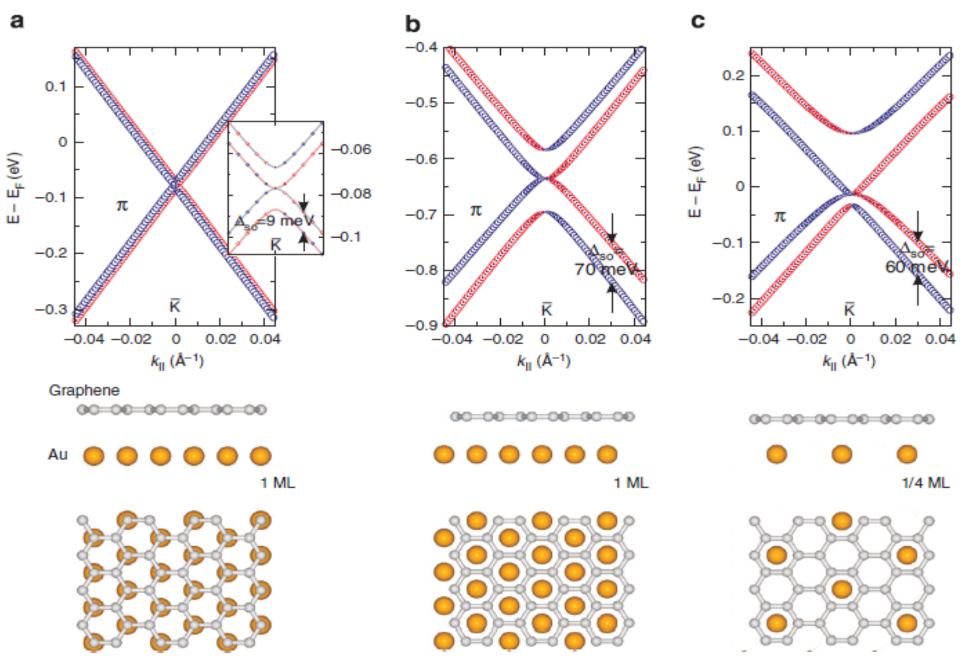




Graphene/Au/Ni

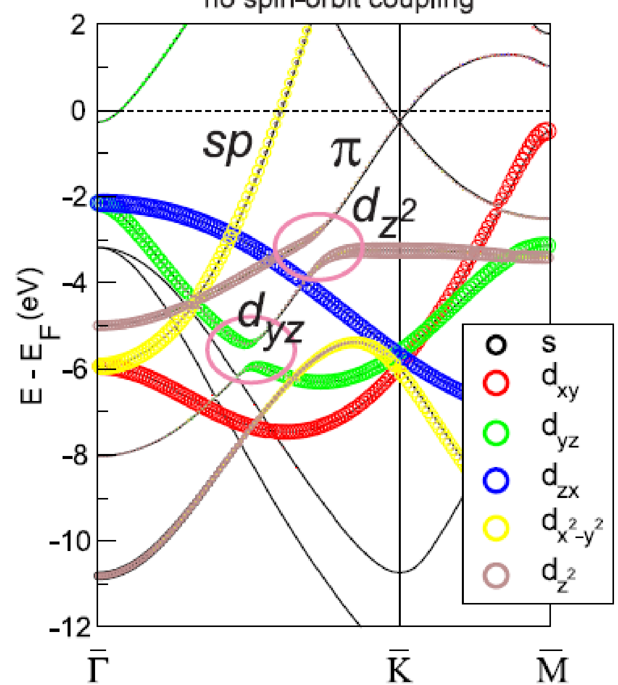






D.Marchenko et al., Nature Commun. 3, 1232 (2013)

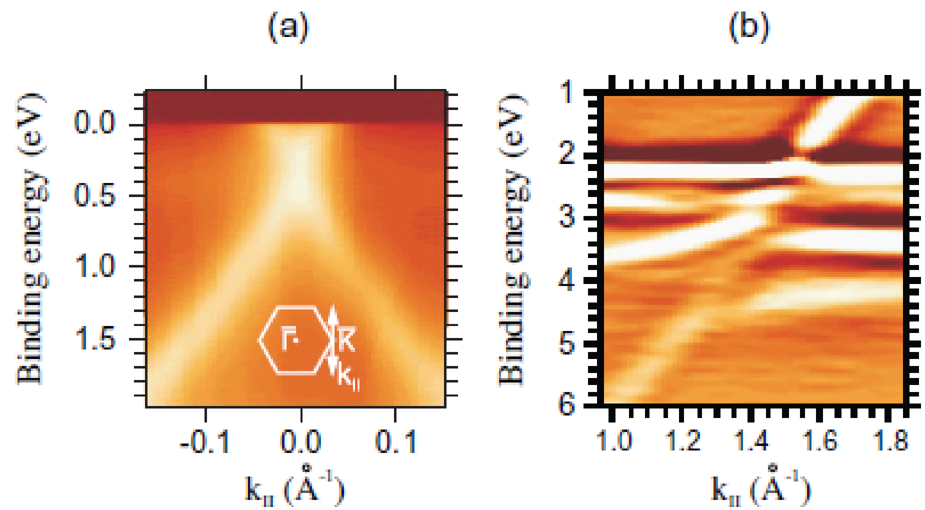
Graphene/1 ML Au
no spin-orbit coupling

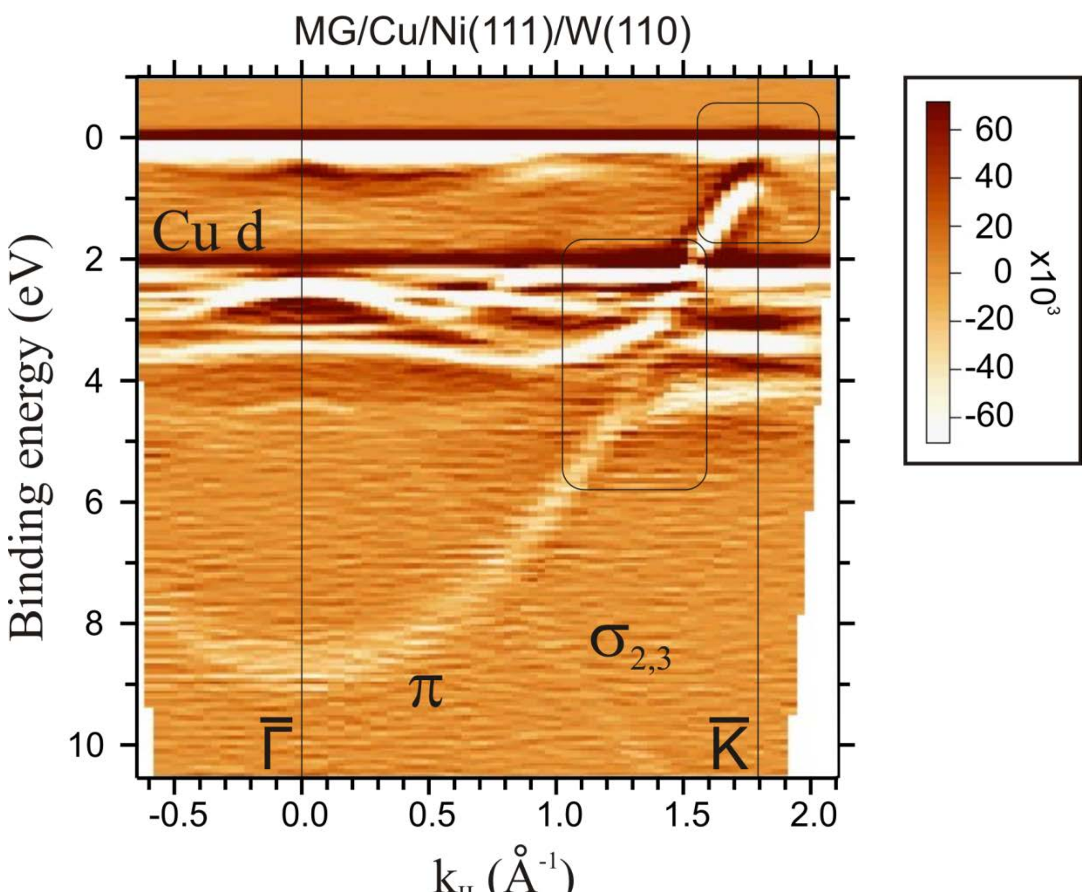


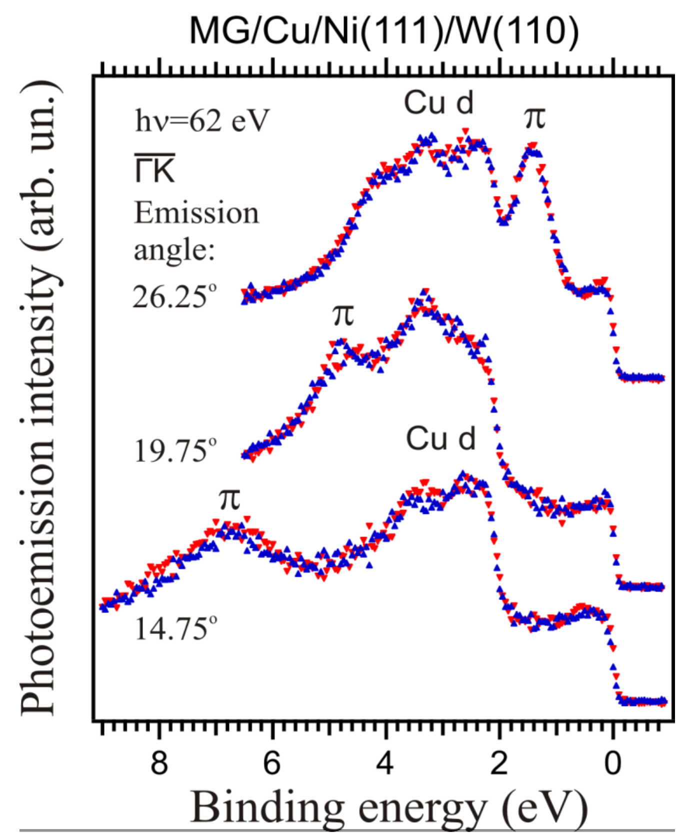
S. Blugel, G. Bihlmayer

Graphene/Cu

Spin structure of graphene synthesised at Ni(111) after intercalation of monolayer of Cu (Z=29).





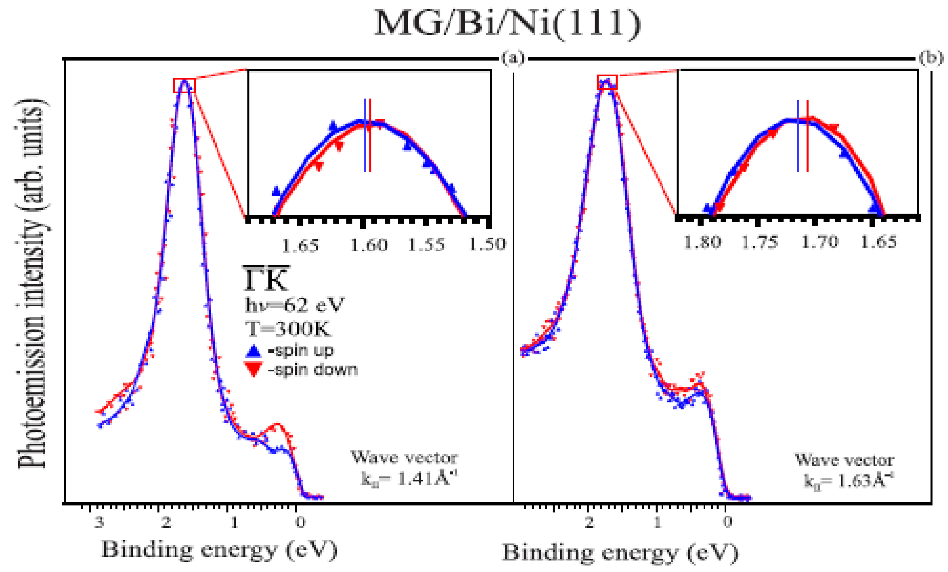


A.M.Shikin et al., New J. Phys. 15, 013016 (2013)

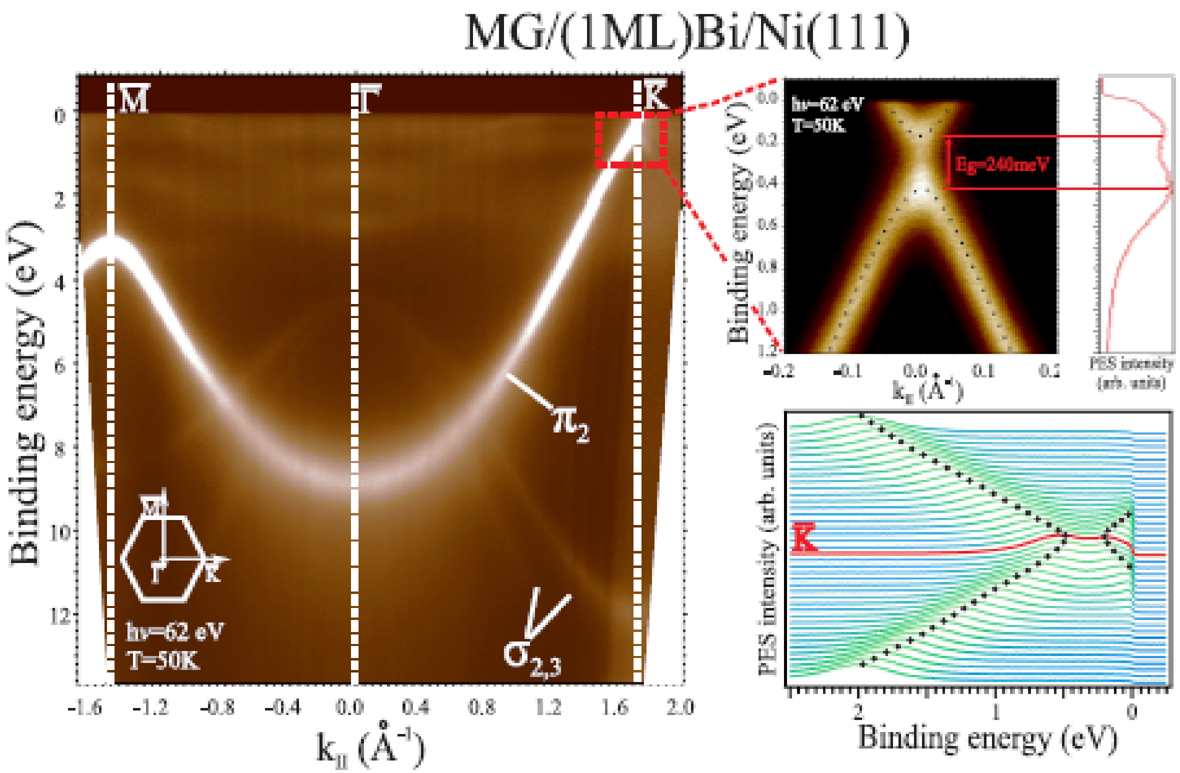
Graphene/Bi

Electronic and spin structure of graphene synthesized at Ni(111) after intercalation of monolayer of Bi (z=83).

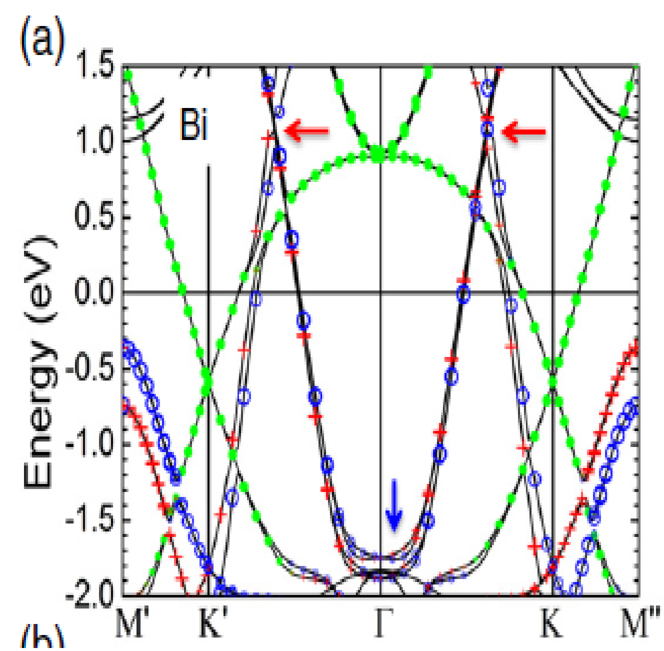
Induced spin-orbit splitting is less than 5-10 meV



E. Zhizhin et al., Carbone, accept for publication



E. Zhizhin et al., Carbone, accept for publication

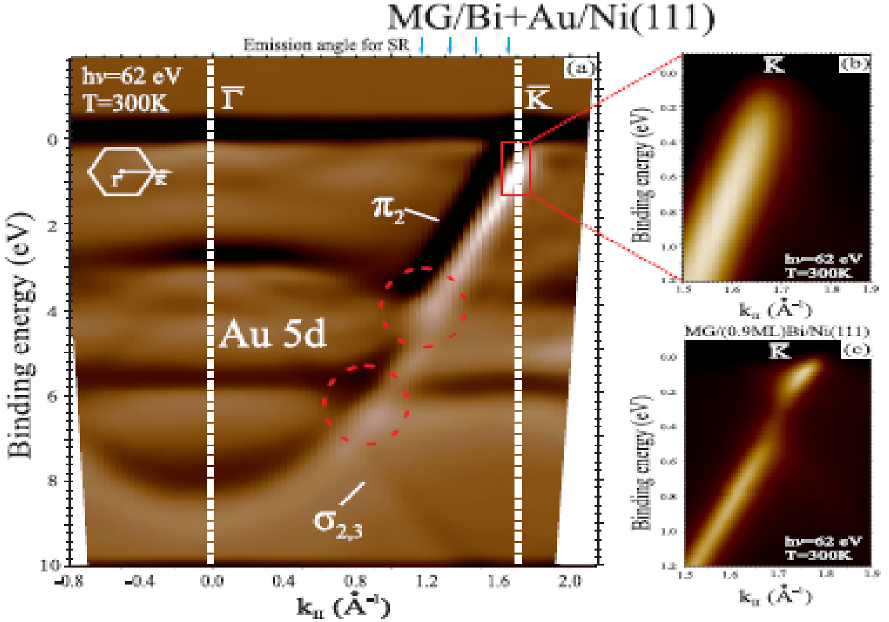


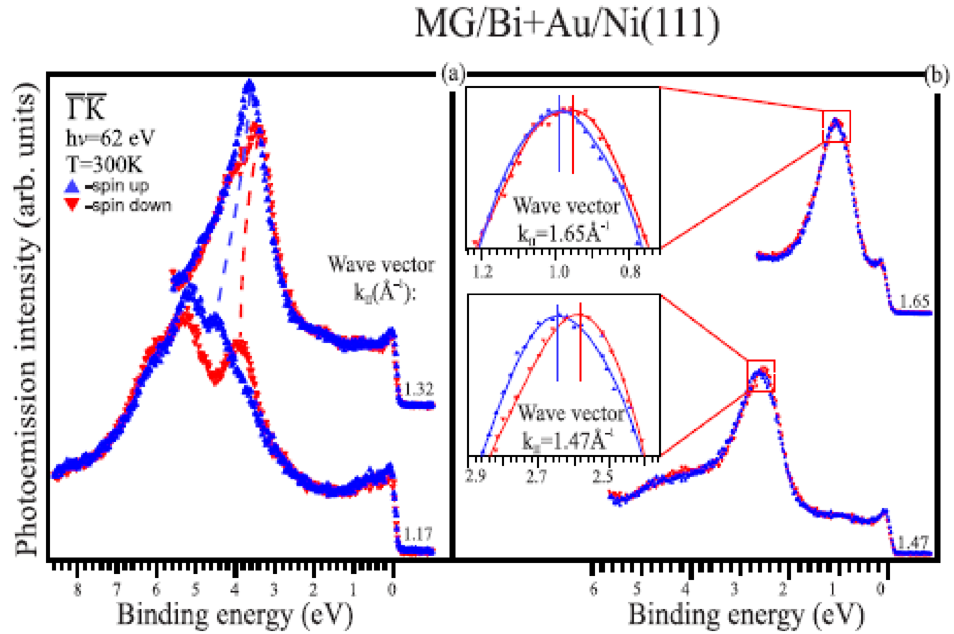
C-H. Hsu et al., Surf. Sci. 616, 148 (2013)

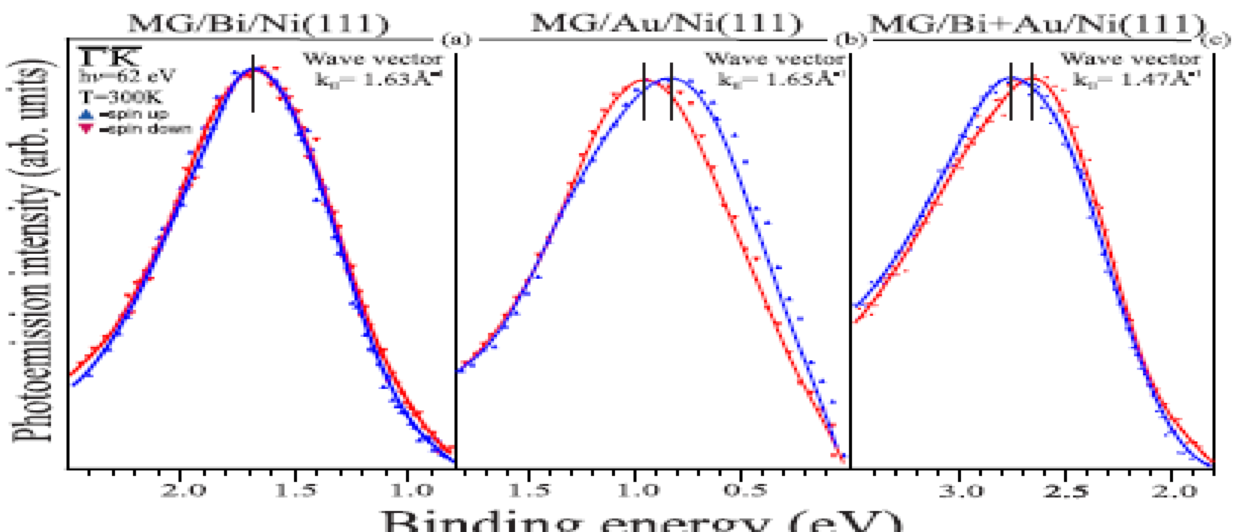
Induced spin-orbit splitting of the graphene π states takes place:

- Under contact with heavy metal with a high atomic number and high inner-atomic potential gradient (therefore, for contact with Cu and Ni it does not take place)
- When the hybridization between the graphene π states and the metal d-states takes place with corresponding spin-dependent avoided-crossing effects (therefore, for contact with heavy metal Bi it is not observed)

Joint intercalation of Bi and Au underneath graphene. Modification of spin-orbit splitting





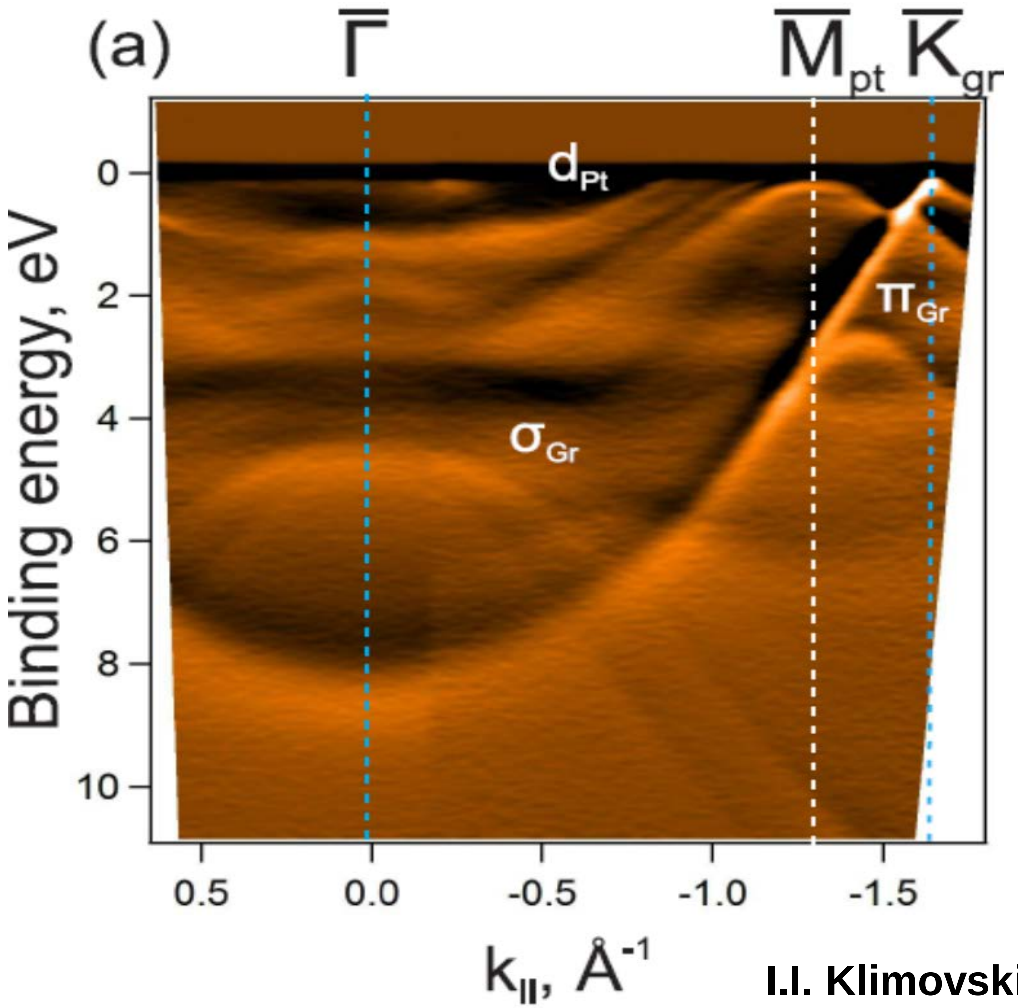


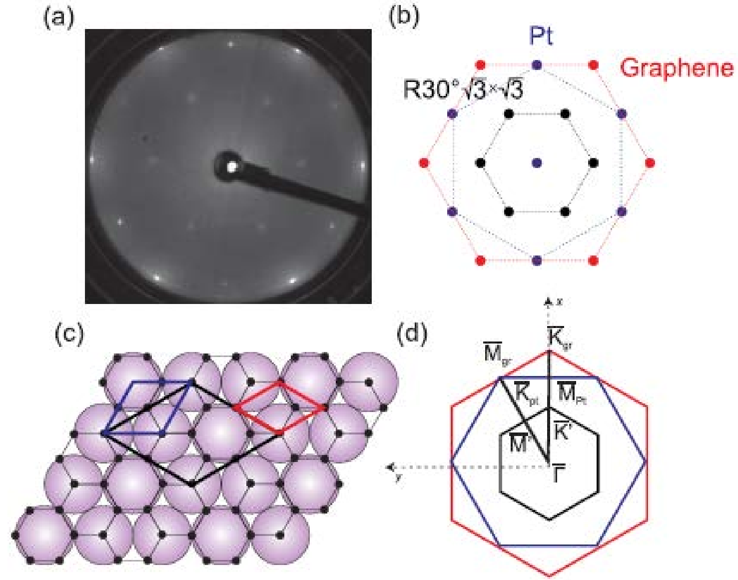
E. Zhizhin et al., Carbone, accept for publication

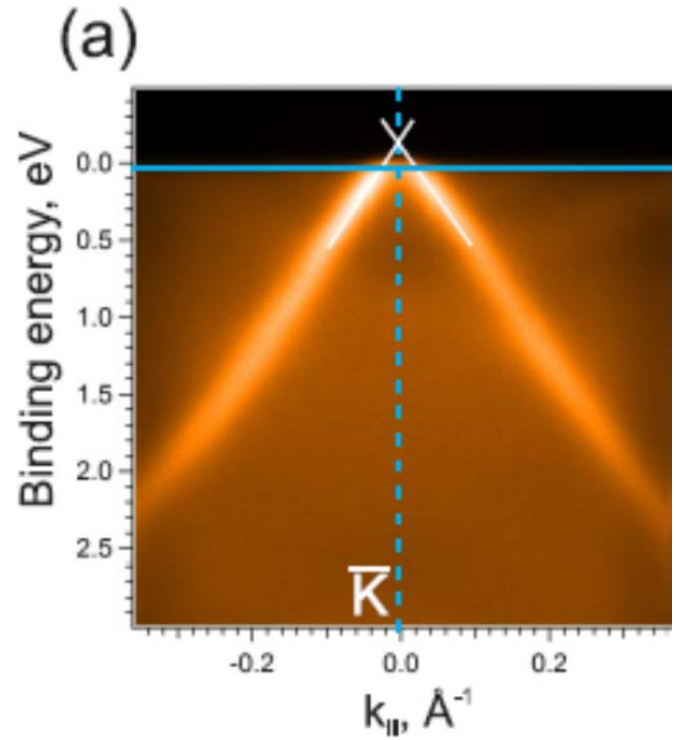
Graphene/Pt

Electronic structure of graphene synthesized at Pt(111)

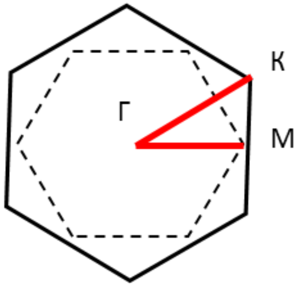
Influence of spin-dependent of avoided-crossing effects near the Fermi level





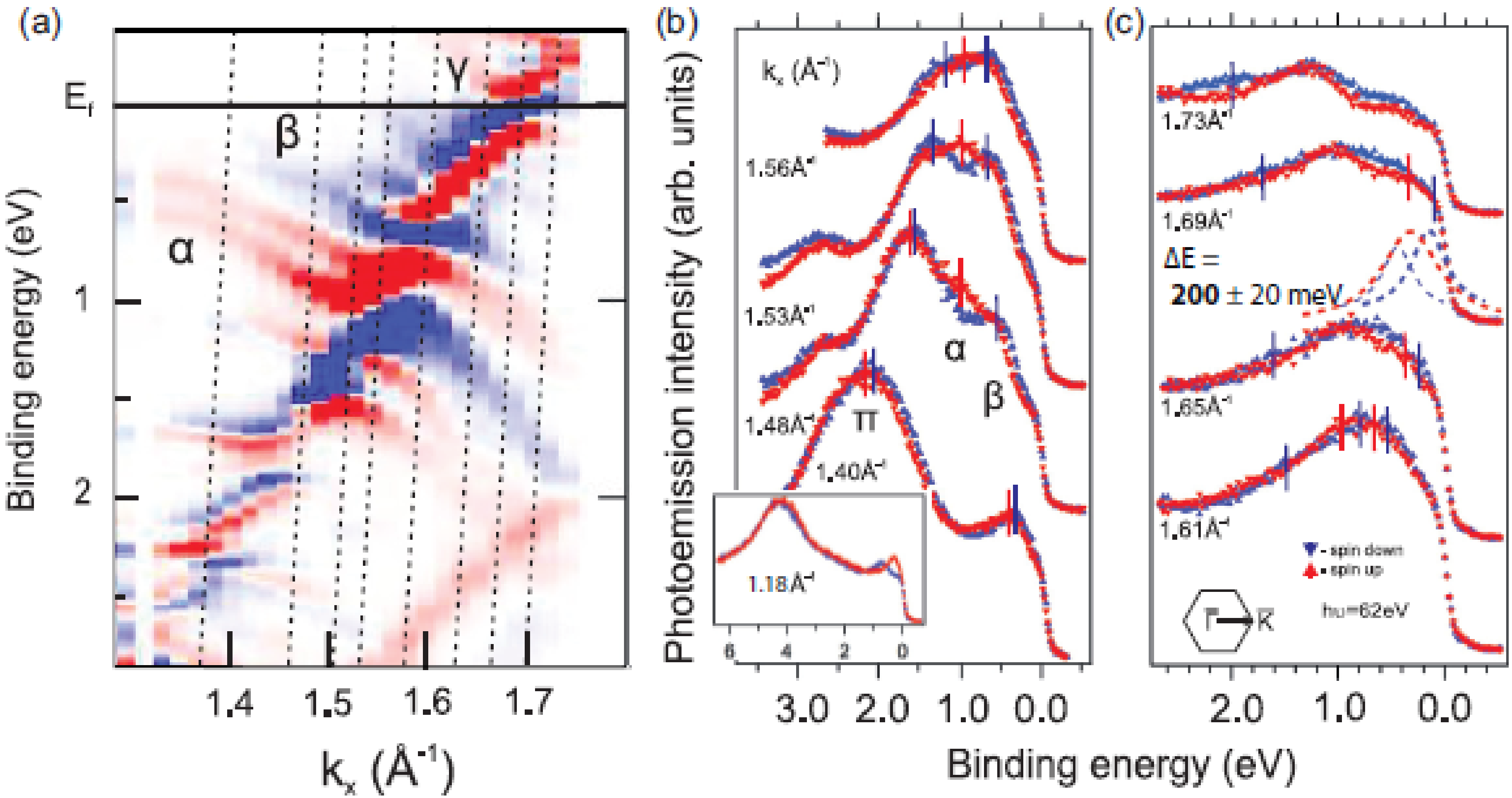


I.I. Klimovskikh et al., Phys. Rev. B 90, 235431 (2014)



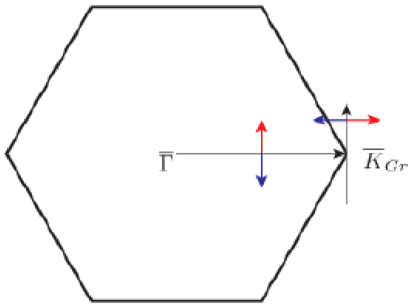
Spin structure of Graphene/Pt(111) near the K-point of the BZ along the ΓK direction.

Enhanced spin-orbit splitting at the Fermi level (up to 150 meV is possible)



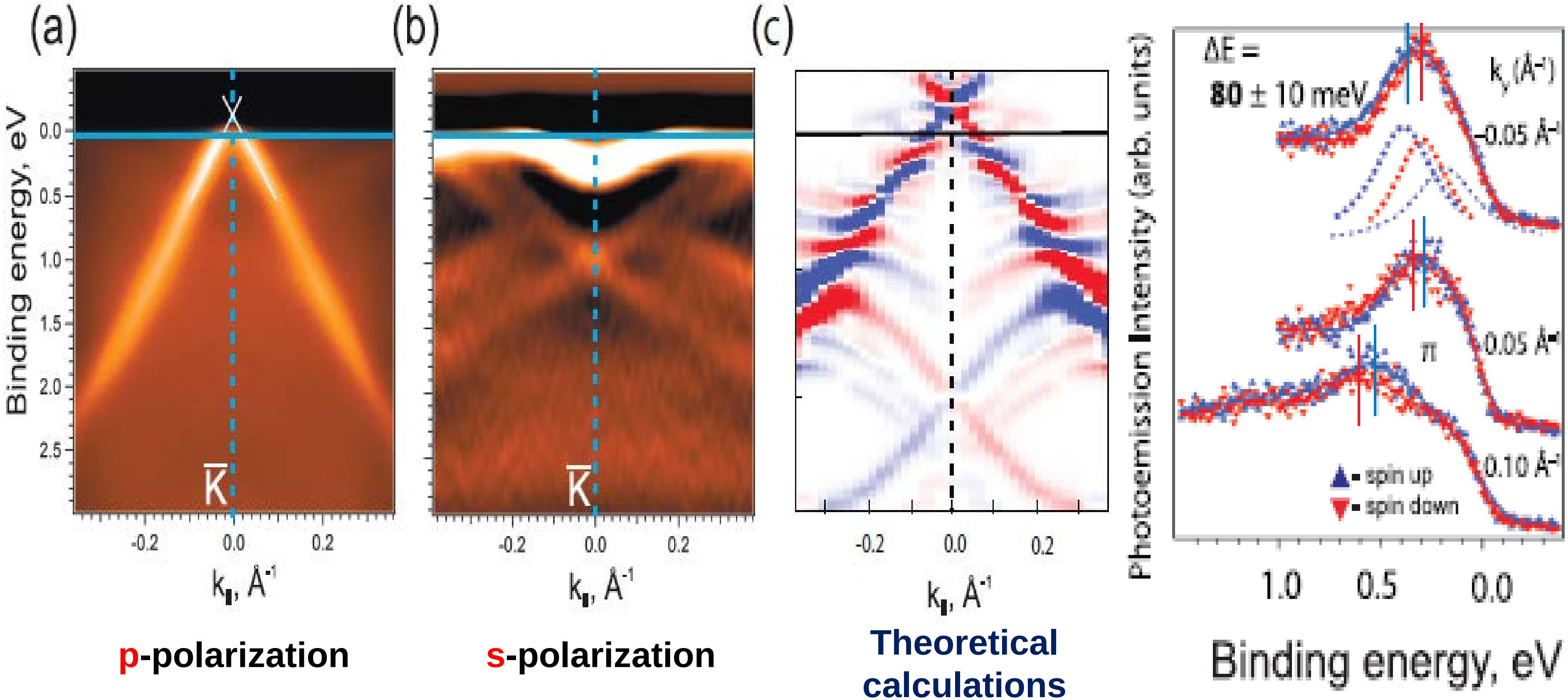
Tsirkin, Chulkov

I.I. Klimovskikh et al., Phys. Rev. B 90, 235431 (2014)



Spin structure of Graphene/Pt(111) near the K-point of the BZ perpendicular the ΓK direction

p-polarization



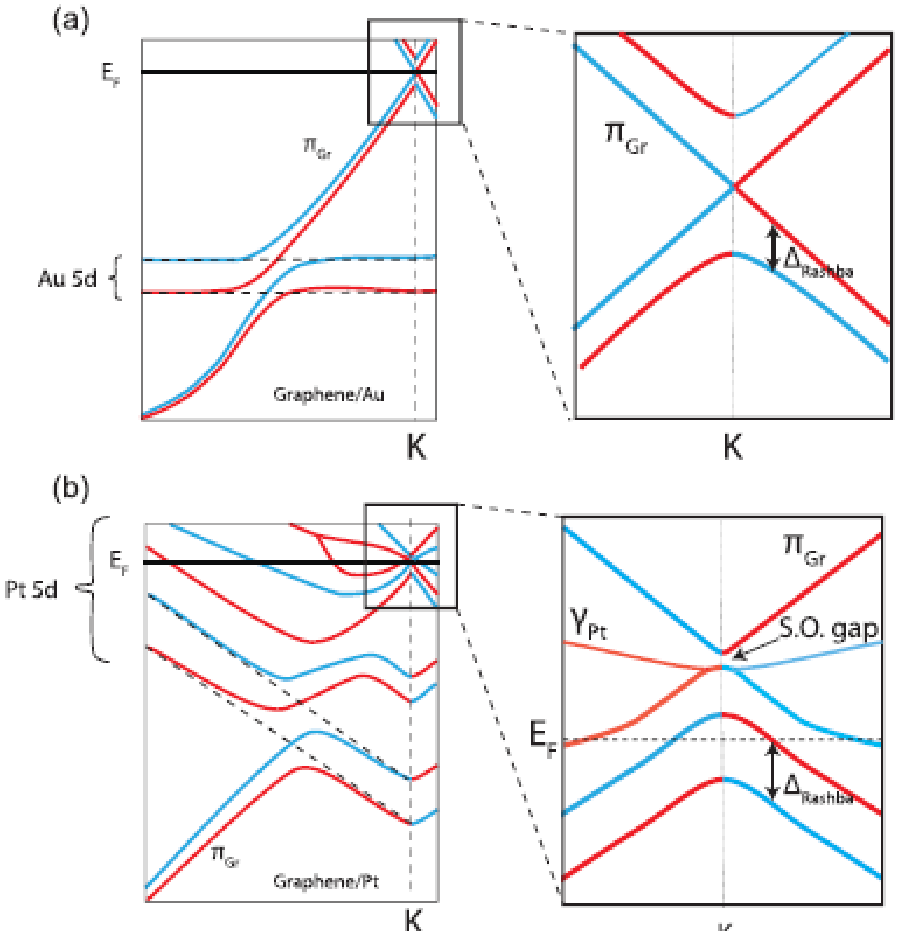
p-polarization

s-polarization

Theoretical calculations

Tsirkin, Chulkov

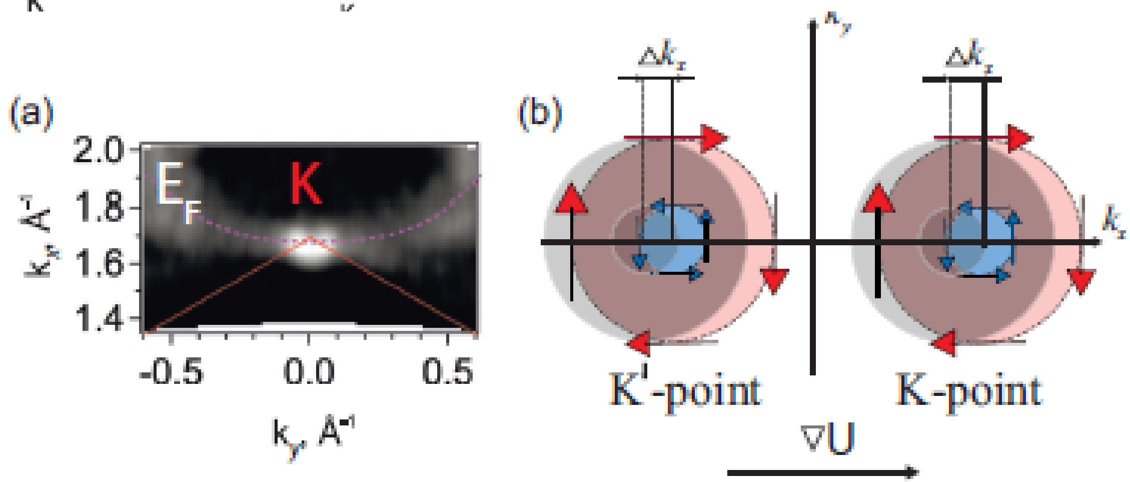
I.I. Klimovskikh et al., Phys. Rev. B 90, 235431 (2014)



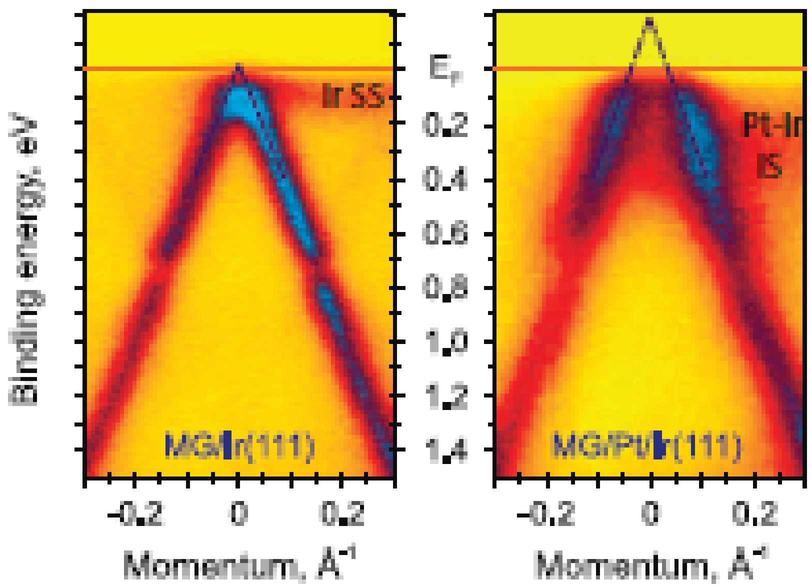
Comparison between spin electronic structure of Graphene/Au/Ni(111) and Graphene/Pt(111)

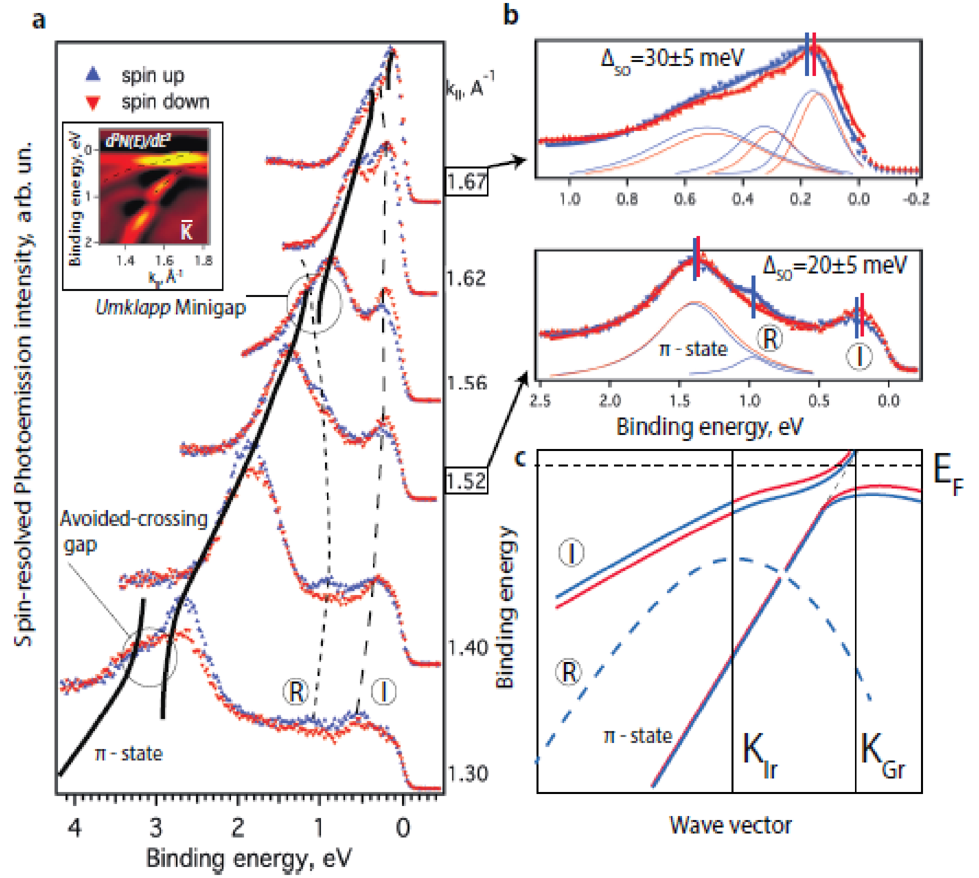
Spin-orbit splitting of graphene π-states at the Fermi level is higher for Graphene/Pt that that for Graphene/Au/Ni.
It means significantly higher efficiency of spin-polarized current generation.

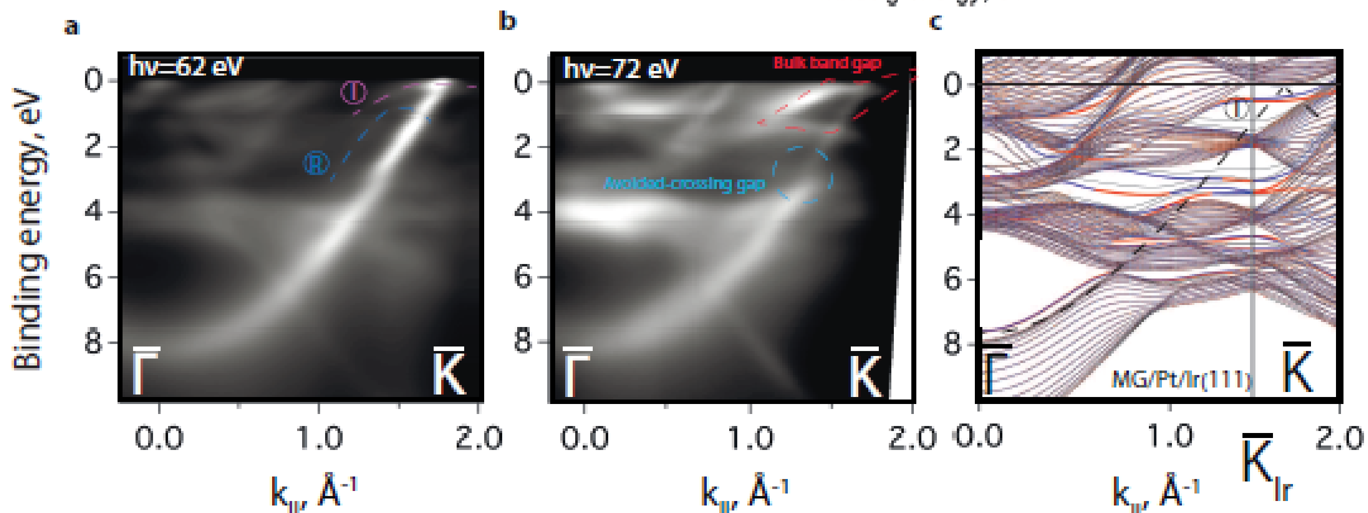
I.I. Klimovskikh et al., Phys. Rev. B 90, 235431 (2014)



Graphene/Pt/Ir(111). Modification of spin-dependent hybridization and Rashba interaction







I.I. Klimovskikh et al., sent for publication in PRB

Tsirkin, Chulkov

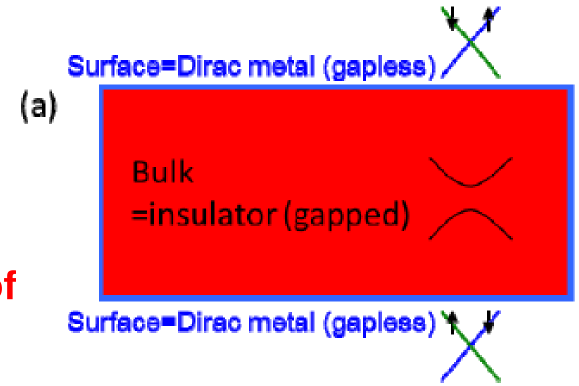
Possibility of formation of topological (Quantum spin Hall) 2D phase in graphene and Graphene/ Me systems

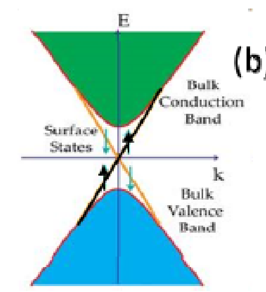
Potential conditions

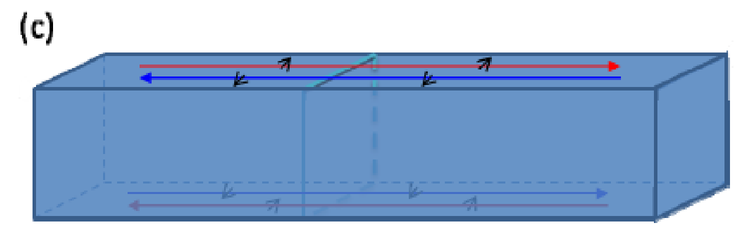
3D topological phase

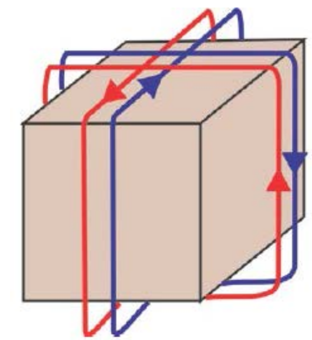
Conditions for formation of topological phase:

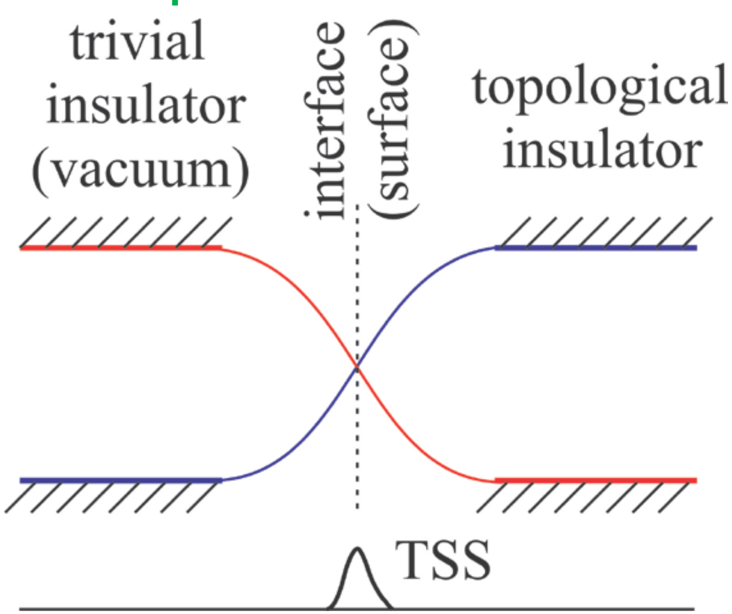
1. **Enhanced spin-orbit coupling and formation of spin-orbit gap at the Fermi level**
2. **Inversion of states with opposite parity relative to the spin-orbit gap edges**
3. **As a result at the interface between topological and trivial phases the metallic-like topological Dirac cone states with helical structure are formed**
4. **Spin current along these surface states, which are developed in opposite directions are dissipation-less**

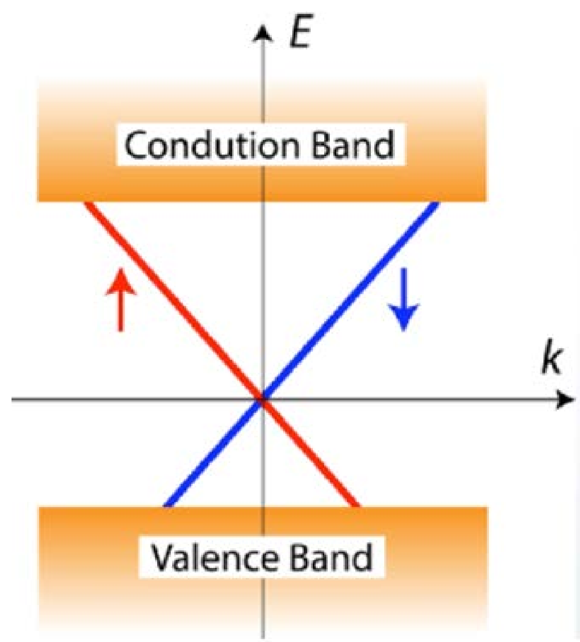


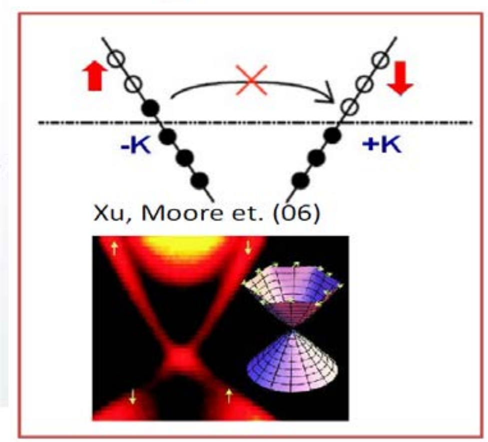




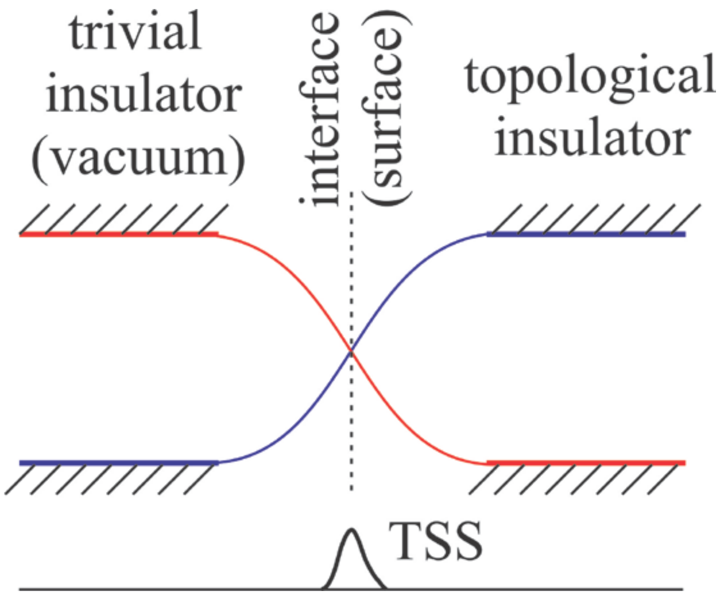


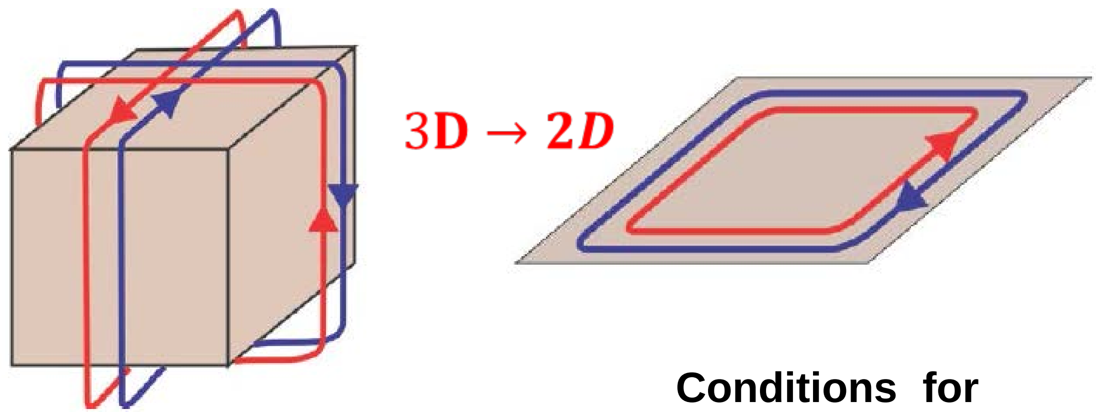




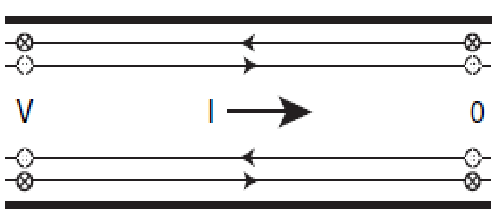


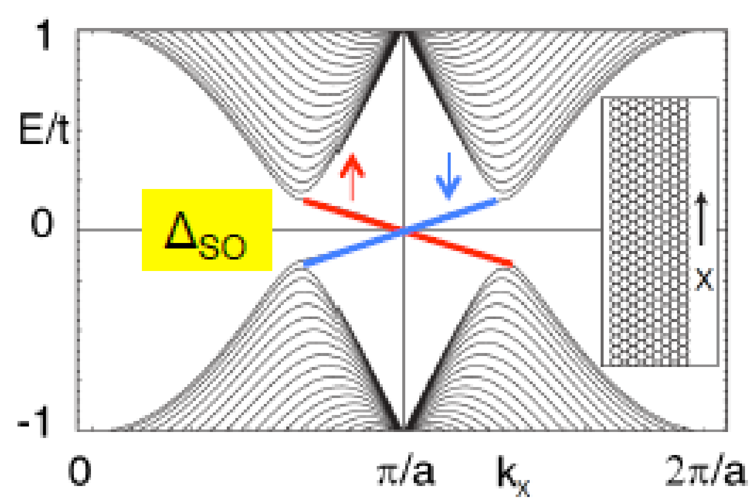
For 2D topological phase the role of topological surface states play the edges states (for 2D islands or nanoribbons)

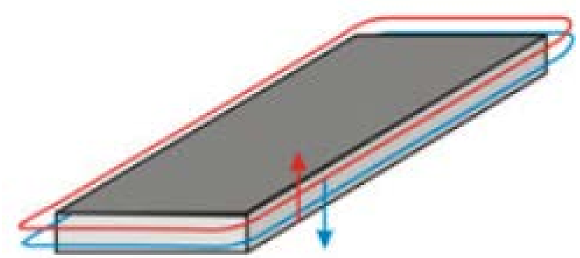


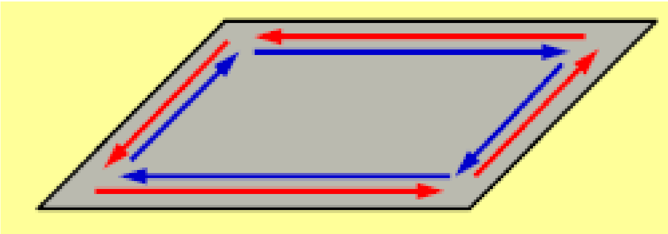


Conditions for formation are the same as for 3D topological phase



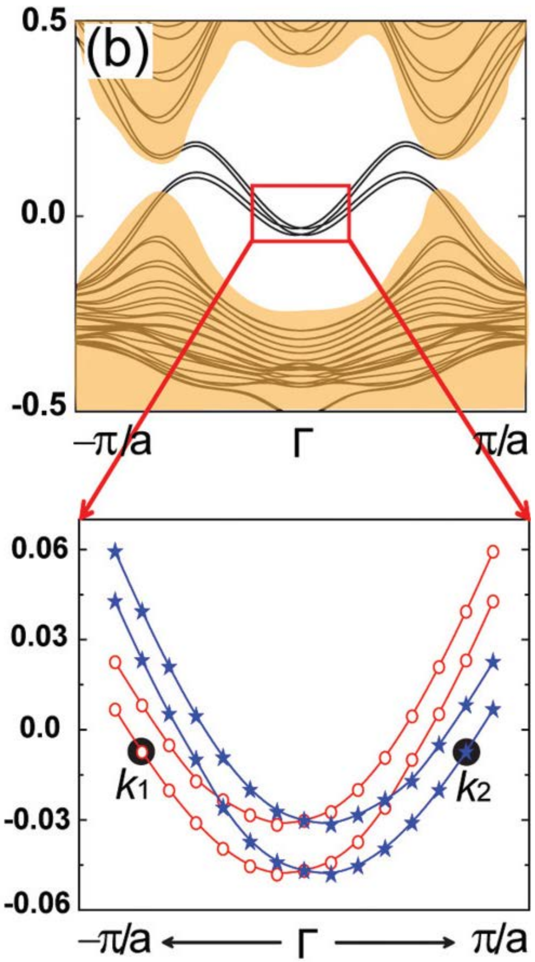


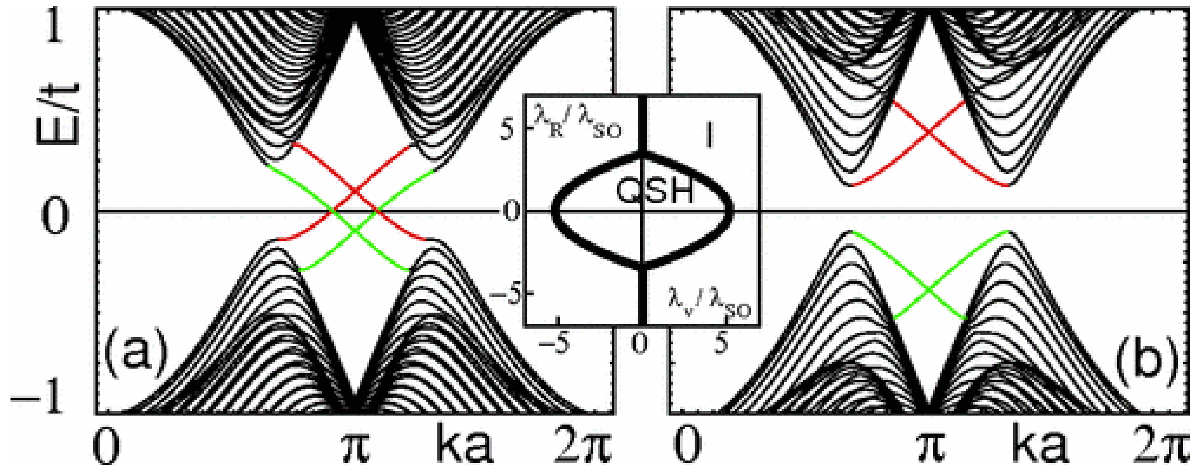




Topological (Quantum spin Hall) 2D phase in graphene

$\lambda_{SO\ int} > \lambda_R$ $\lambda_{SO\ int} < \lambda_R$





Topological insulator **Trivial insulator**

$$\lambda_{SO} = \frac{|s|}{18(sp\sigma)^2}\xi^2 \qquad \lambda_R = \frac{eEz_0}{3(sp\sigma)}\xi$$

ξ- atomic spin-orbit interaction strength

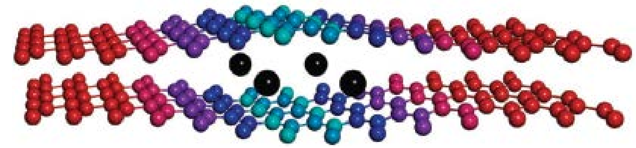
2D topological phase means formation of topological “surface” (edge) states around graphene islands and generation of opposite spin currents along the edge states

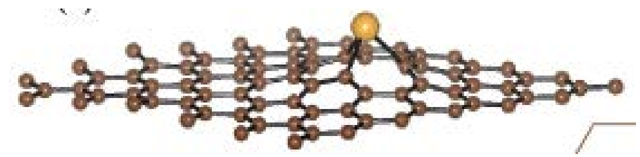
Y. Li et al., PRB 87, 245127 (2013) C.L. Kane, E.J. Mele, PRL 95, 226801 (2005) C.L. Kane, E.J. Mele, PRL 95, 146802 (2005)

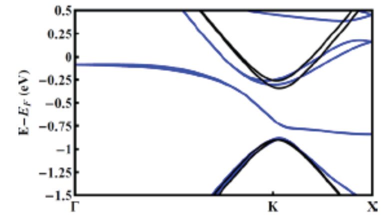
Conditions for formation of topological phase in graphene:

1. **Inversion of the states between K and K’ points of the BZ**
2. **$\lambda_{SO\ int} > \lambda_{SO\ R}$**

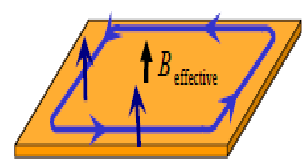
Strain of graphene or adatoms induce rehybridization between s, p, d states that leads to enhanced S-O coupling and formation of 2D topological phase and corresponding energy gap (Δ_{SO} in graphene 100 μeV) (Δ_{SO} of p states in carbon atom 6 meV)

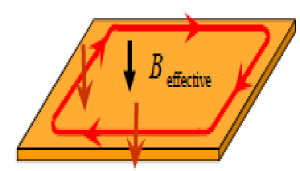




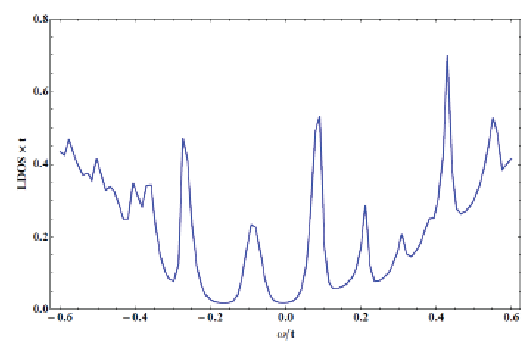


Formation of topological phase can be also observed for adatoms deposited on graphene including hybridization with adatom states (increase of contribution of d states and rehybridization due to spin-dependent avoided – crossing effects)

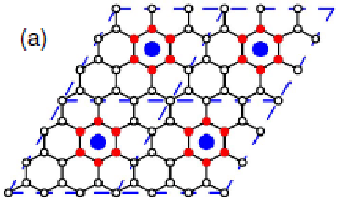




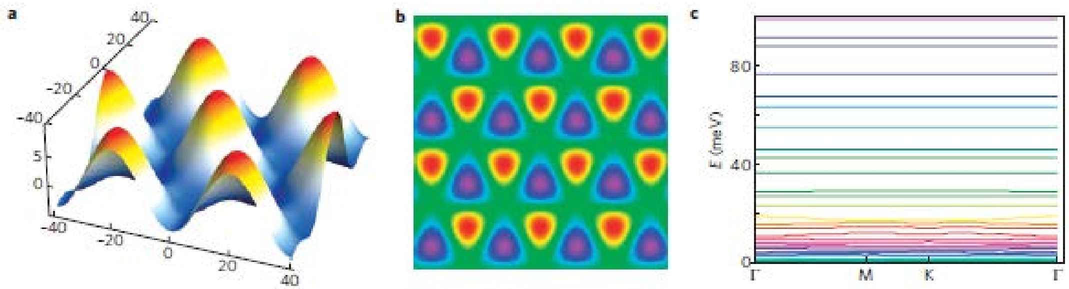
It is followed by spin quantum Hall effect. As a results a preudo-magnetic field is formed leading to formation of quantized Landau levels inside of the gap



Increase of internal spin-orbit coupling and formation of spin-orbit gap in strained graphene.
Formation of internal pseudomagnetic field and formation of quantized Landau levels without external magnetic field

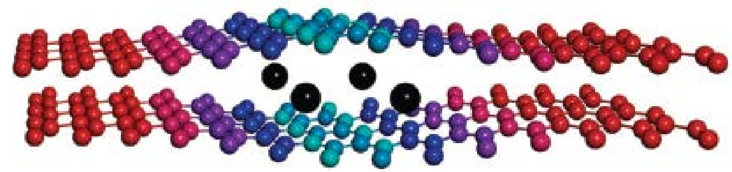


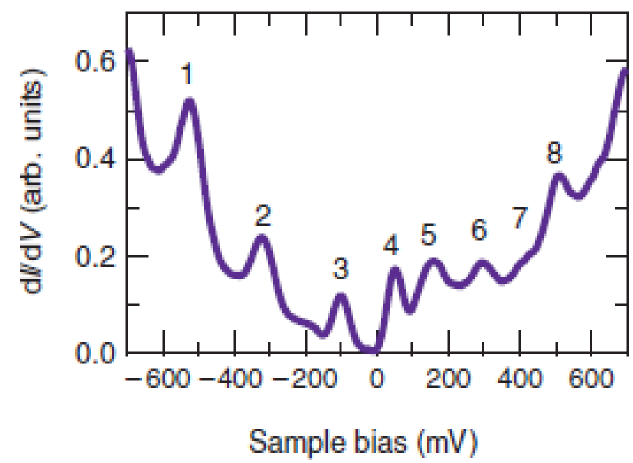
Opening spin-orbit energy gap in strained graphene superlattices (theoretical calculations)



F. Guinea et al., Nature Phys. 6, 30 (2010)

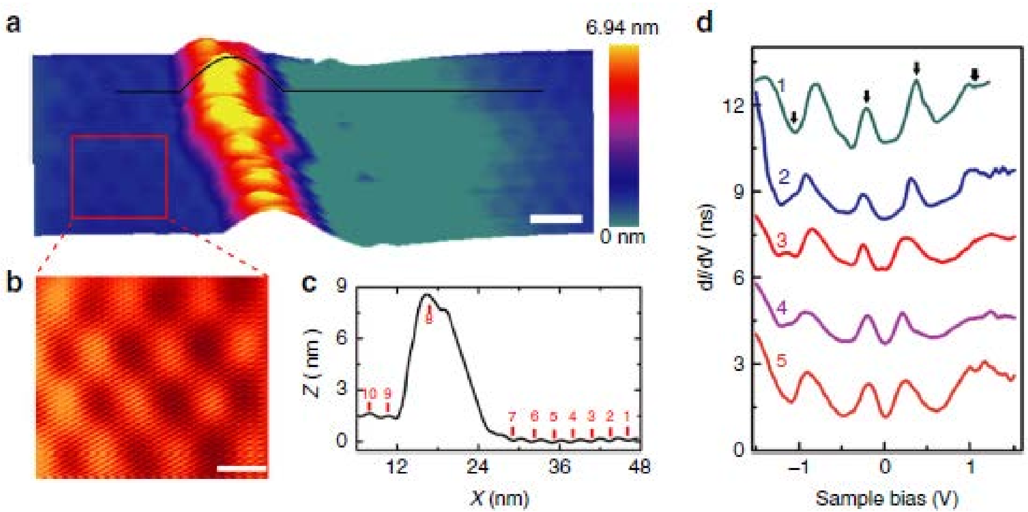
Formation of Landau levels in K-intercalated bilayer graphene (experimental results)





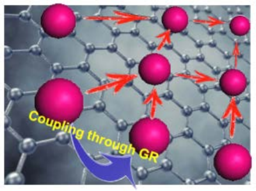
D. Guo et al., Nature Commun. 10, 1038 (2012)

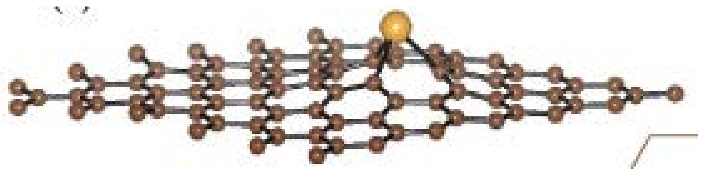
Twisted graphene bilayer on Rh-foil (experimental results)

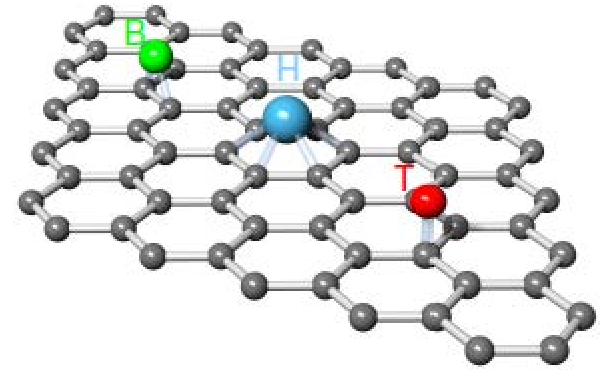


W. Yan et al., Nature Commun. 4, 2159 (2013)

Increase of internal spin-orbit coupling and formation of spin-orbit gap in graphene via metal (p,d) atom adsorption (Os,Tl, Re) .

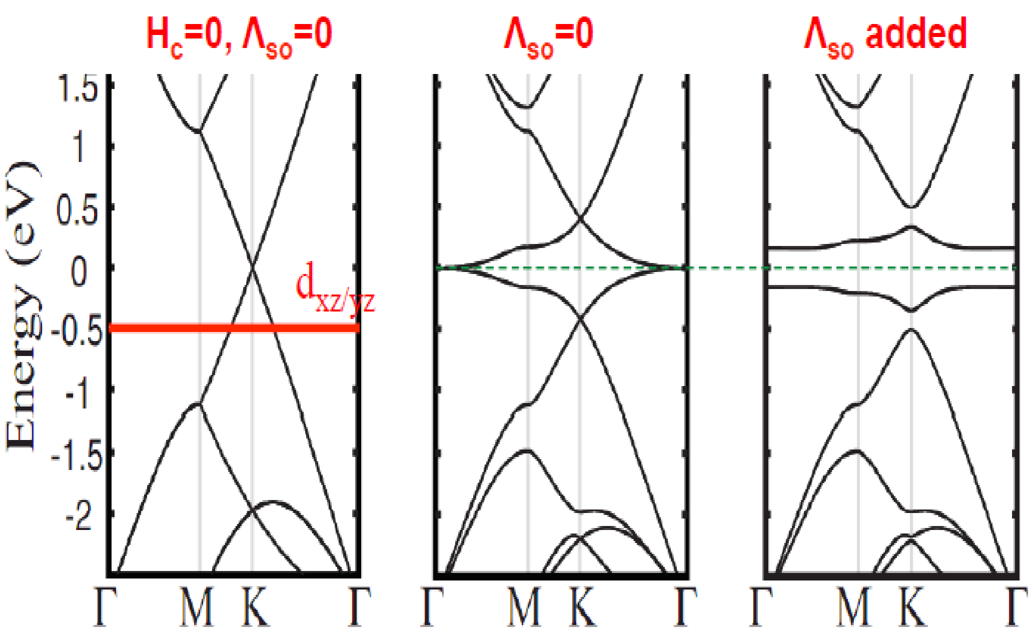


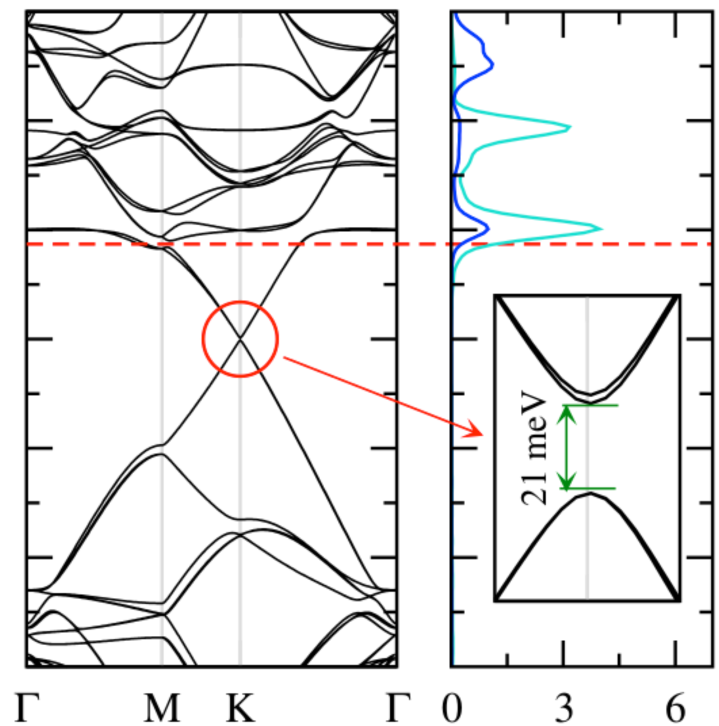




Tl on graphene

Os on graphene





J. Hu et al., Phys. Rev. Lett. 109, 266801 (2012)

C. Weeks et al., PRX 1, 021001 (2011)

Conditions for formation of topological phase in graphene:

1. **Formation of the SO gap due to enhanced SO coupling**
2. **Inversion of the states between K and K' points of the BZ**
3. $\lambda_{SO\ int} > \lambda_{SO\ R}$

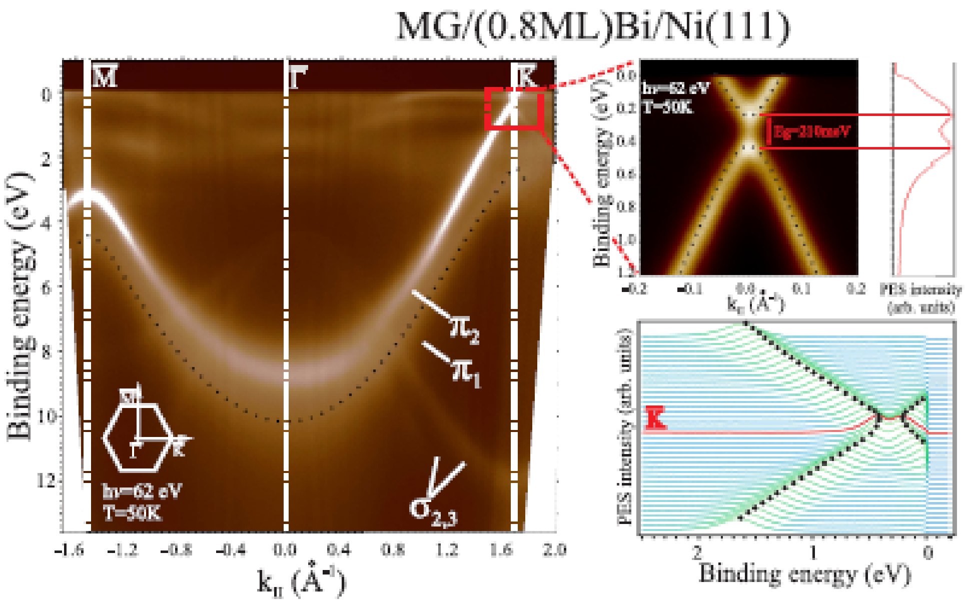
Can the topological phase be formed in the studied systems with graphene intercalated by heavy metals Au, Pt, Bi?

How the induced spin-orbit splitting and spin-dependent hybridization influence on formation of topological phase?

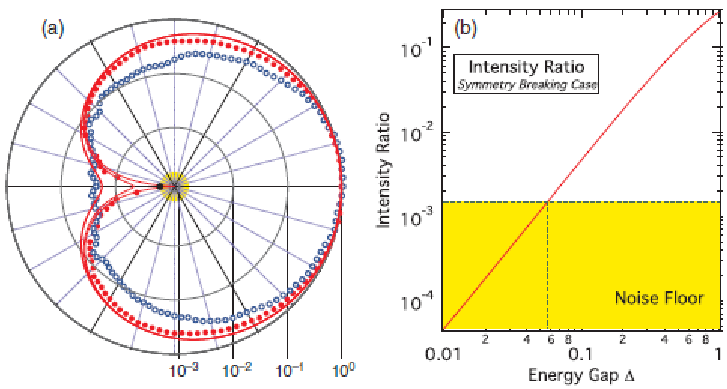
How can we distinguish the contributions of $\lambda_{SO\ int}$ and $\lambda_{SO\ ext}$ (i.e. λ_R) in the formed spin-dependent hybridization and spin-dependent avoided-crossing effects?
How do the relations between these terms influence on formation of topological phase?

Partially Bi-intercalated graphene

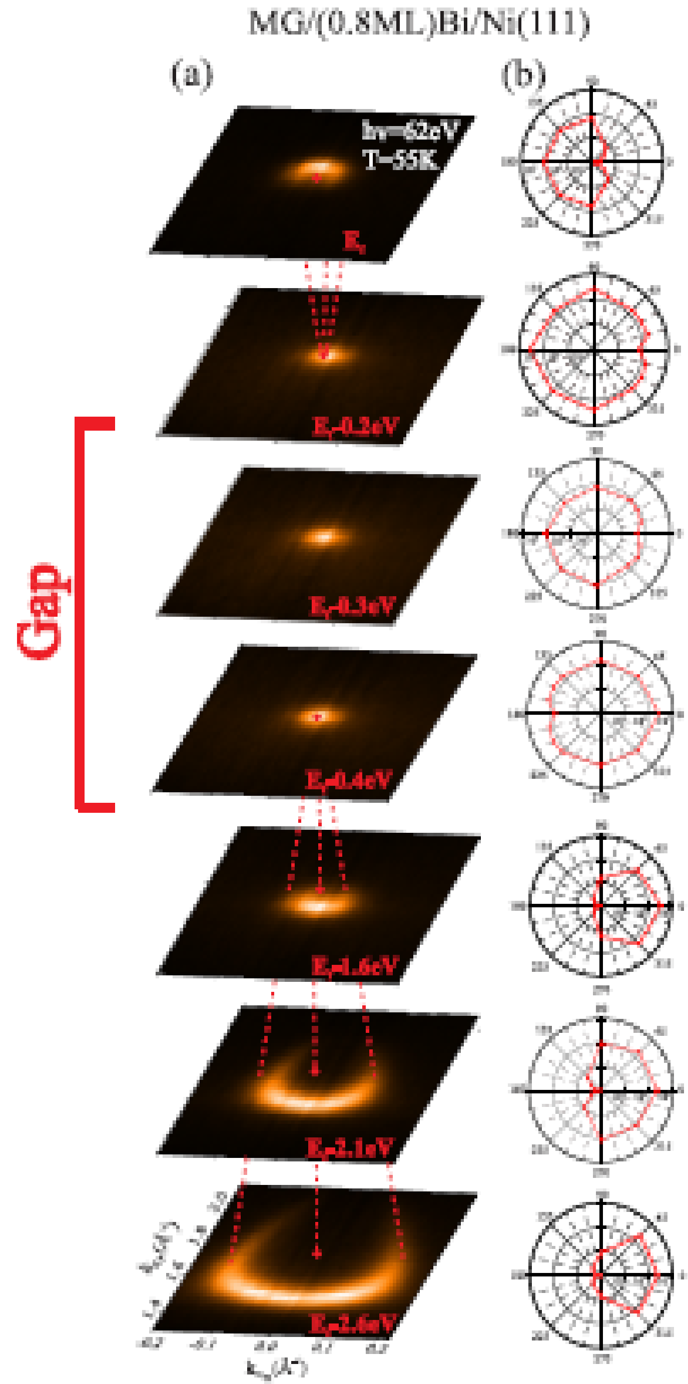
(no strain, no hybridization, 5-10 meV)

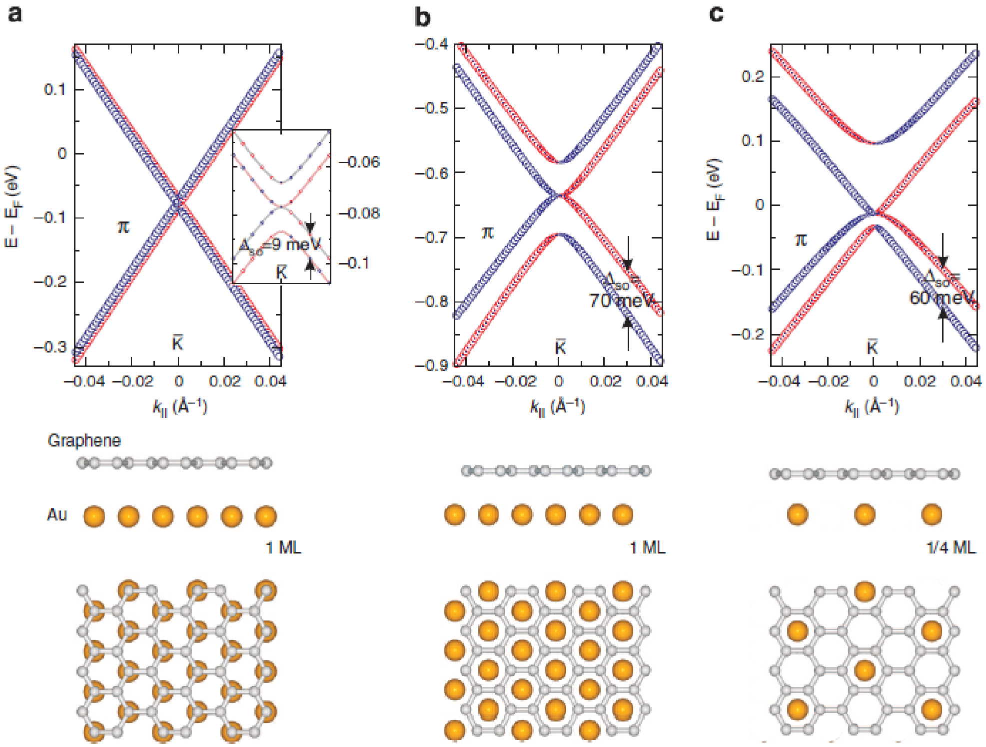


Relation of intensity in the first and the second BZ (~ 40) that is less than that appeared by the symmetry breaking

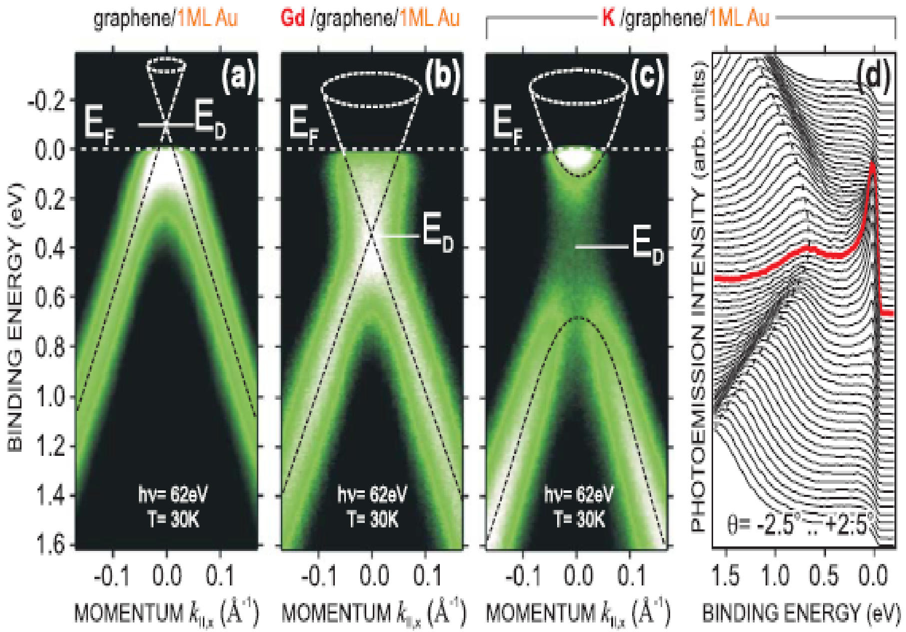


Constant induced ΔSO can be interpreted as λ_R. It means $\lambda_{SO\ int}$ is less than λ_R.

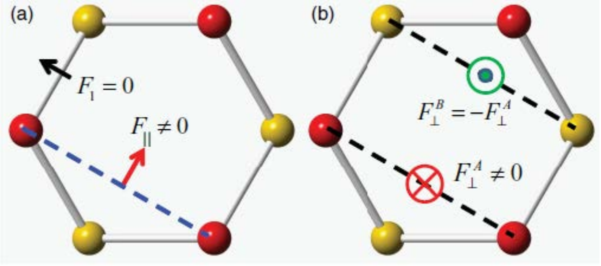




MG/Au/Ni(111)

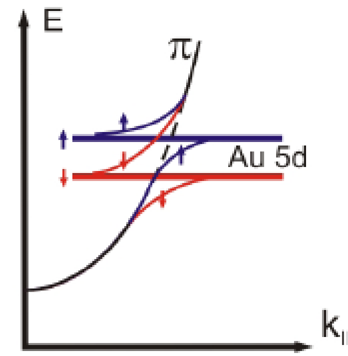


Δ_{SO} (Δ~100 meV) is approximately constant



However, enhanced spin-orbit (Δ~100 meV) can be induced only due to spin-dependent hybridization and (2x2) superstructure. It could correspond to $\lambda_{SO\ int}$ term. While Δ_{SO} (Δ~100 meV) is approximately constant

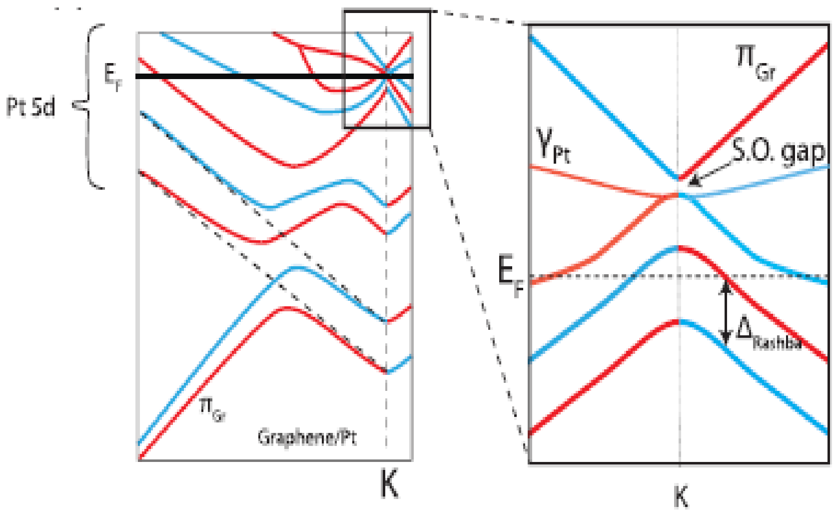
Without hybridization it can be Δ ~10meV, as in the case of Bi-intercalated graphene.
This contribution can correspond to Rashba term (λ_R).

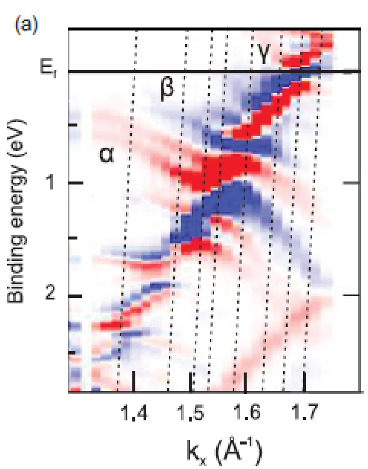


Formation of gap and topological phase is still open question

MG/Pt(111). Formation of topological phase.

Pt d states cross graphene π states at the Fermi level.
Spin-dependent avoided-crossing leads to Δ_{SO}~60-100 meV.
It can be considered as $\lambda_{SO\ int} > \lambda_R$



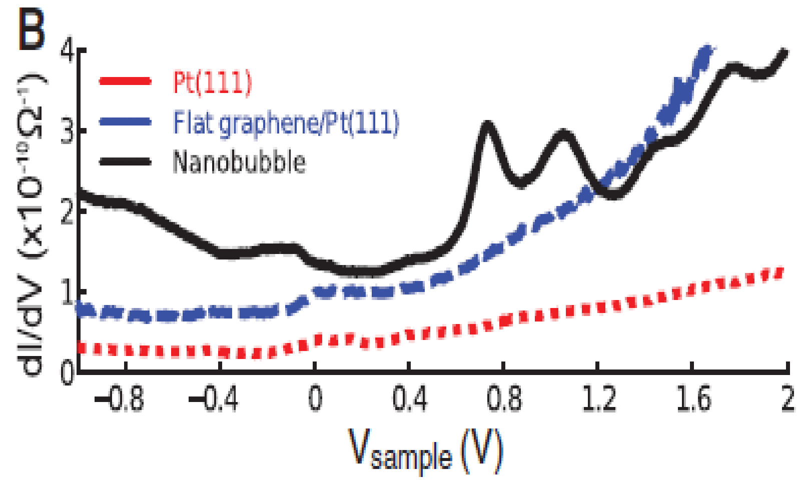


λ_R~30 meV

MG/Pt(111)
Topological gap is formed

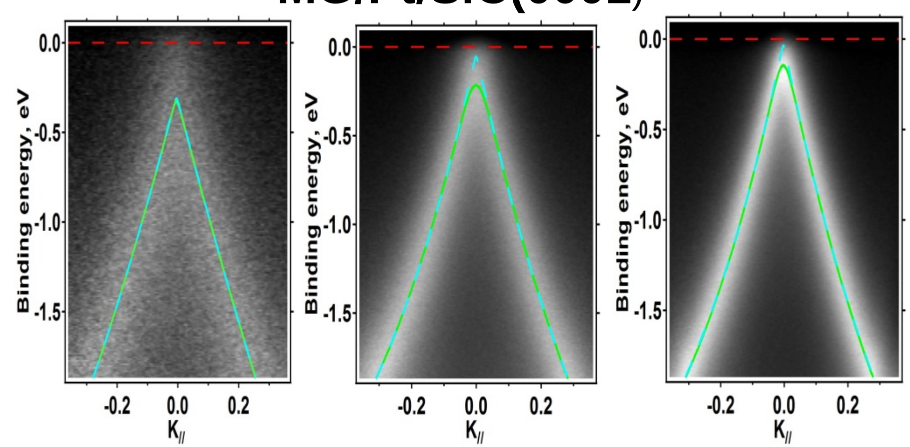
MG/Pt/SiC(0001) islands?

Observation of Landau quantization in Graphene Nanobubbles on Pt(111) due to strain-induced pseudo-magnetic field



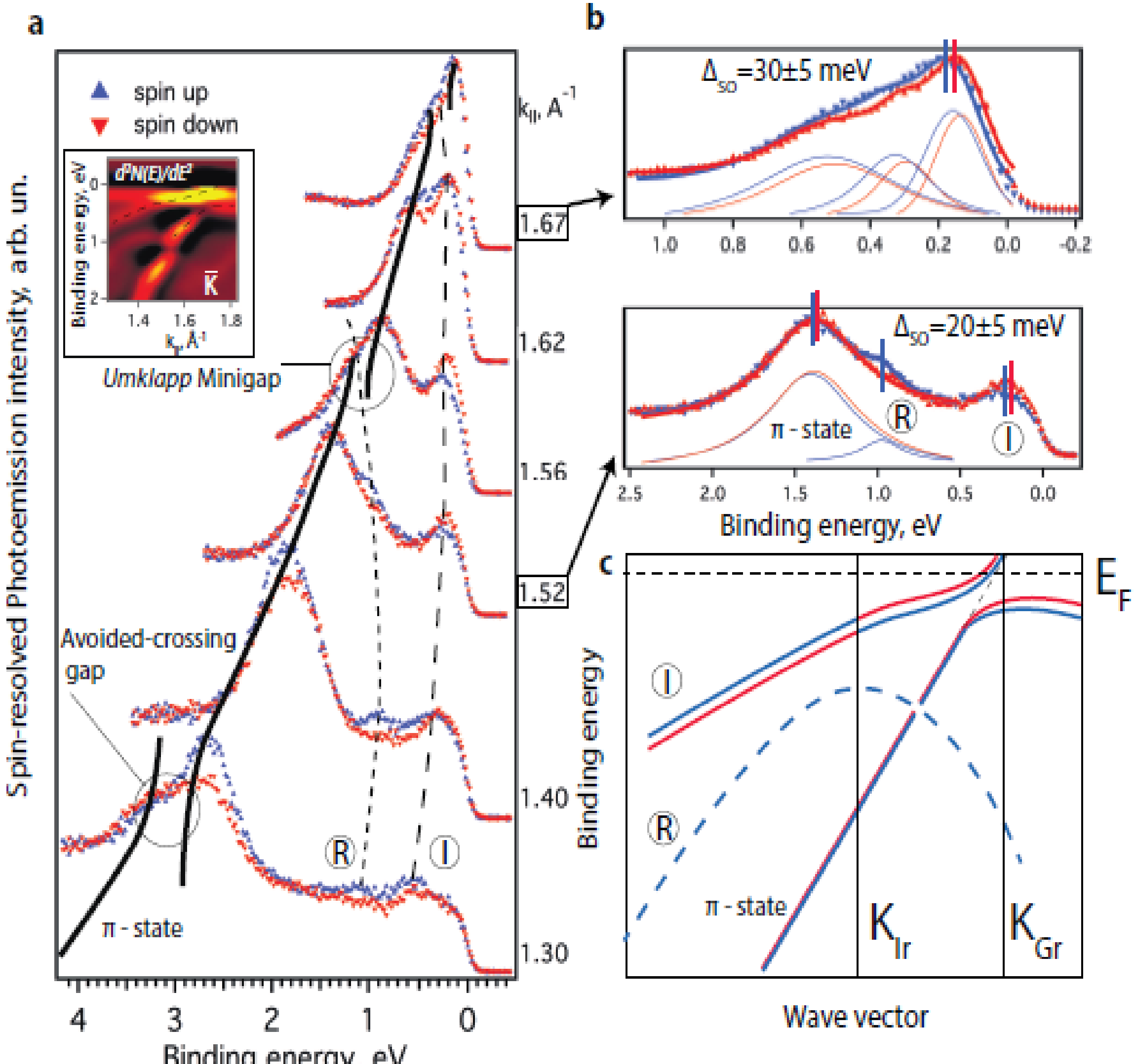
N. Levy et al., Science 329, 544 (2010)

MG/Pt/SiC(0001)



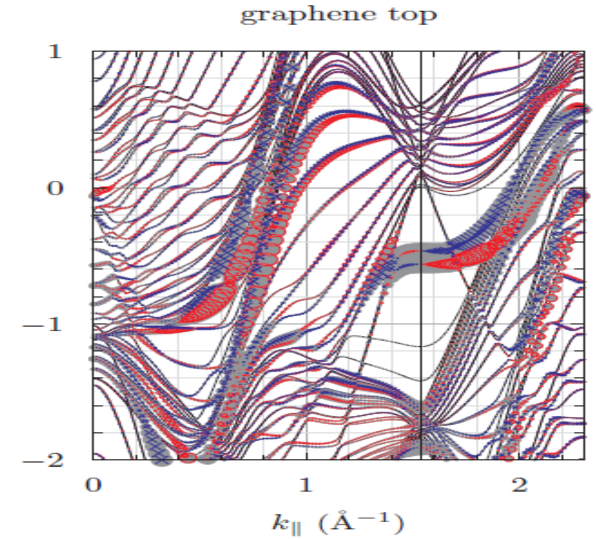
850°C **1000°C** **1200°C**

MG/Pt/Ir(111). Turning in formation of topological phase by reducing of the Rashba term in spin-orbit interaction



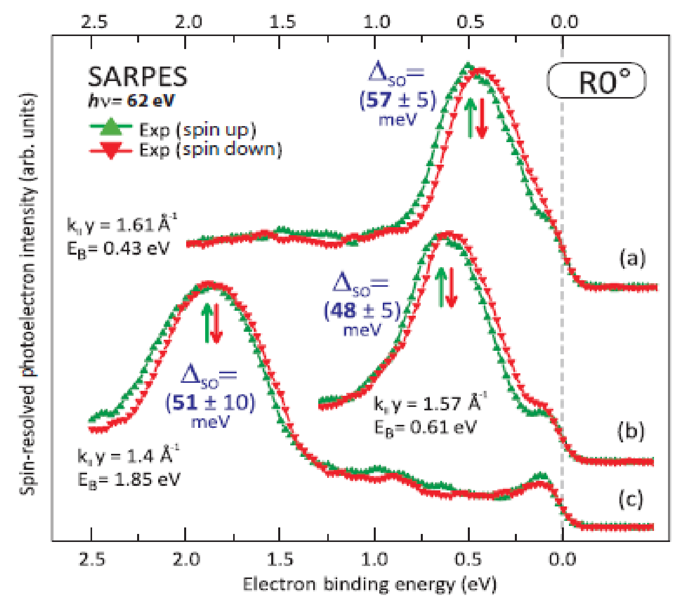
$\lambda_R < \lambda_{SO\ int}$

MG/Pt/Ir(111).



MG/Ir(111)

$\lambda_R \geq \lambda_{SO\ int}$



Conditions for formation of topological phase and its modification

Constant $\Delta_{R\ S0}$ splitting (~50meV)

D. Marchenko et al., PRB 87, 115426 (2013)

So, topological phase can be formed in Graphene/Me systems and modified

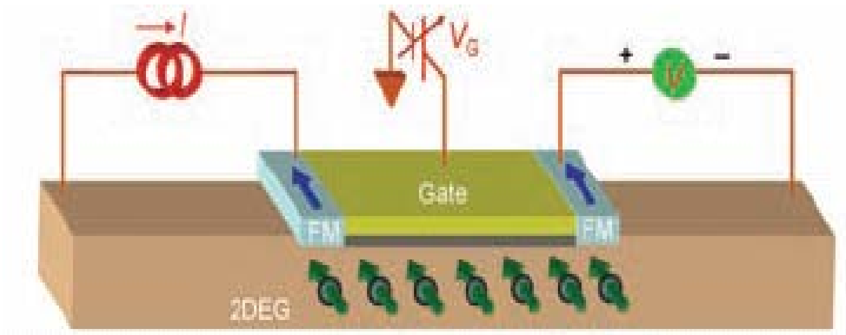
Graphene and spintronics

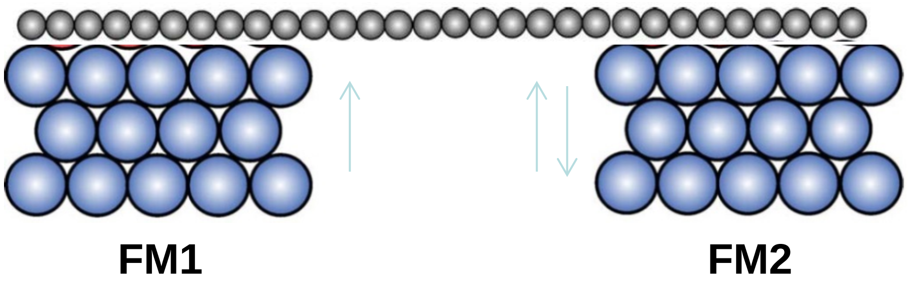
Graphene spin filter

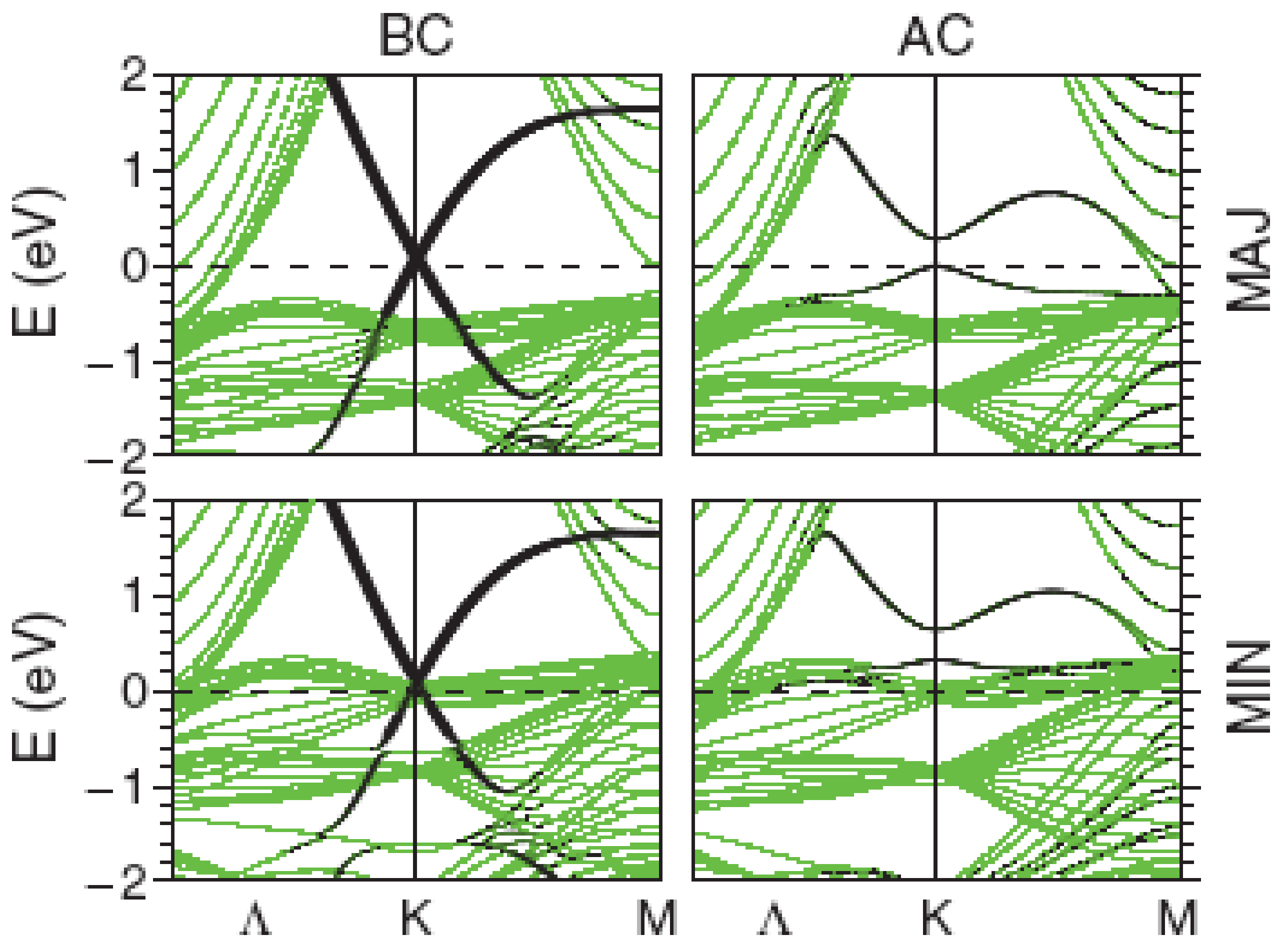
Spin-polarized current generation

Switch of reversal induced magnetization of contacting FM nano-obijects by developed spin current due to spin-torque effect (without external magnetic field)

Graphene spin filter





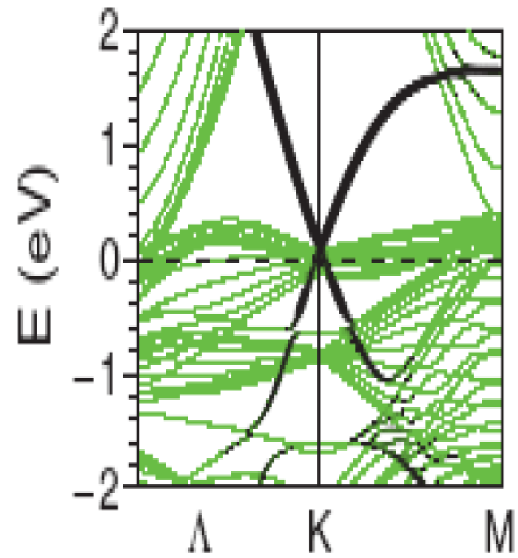


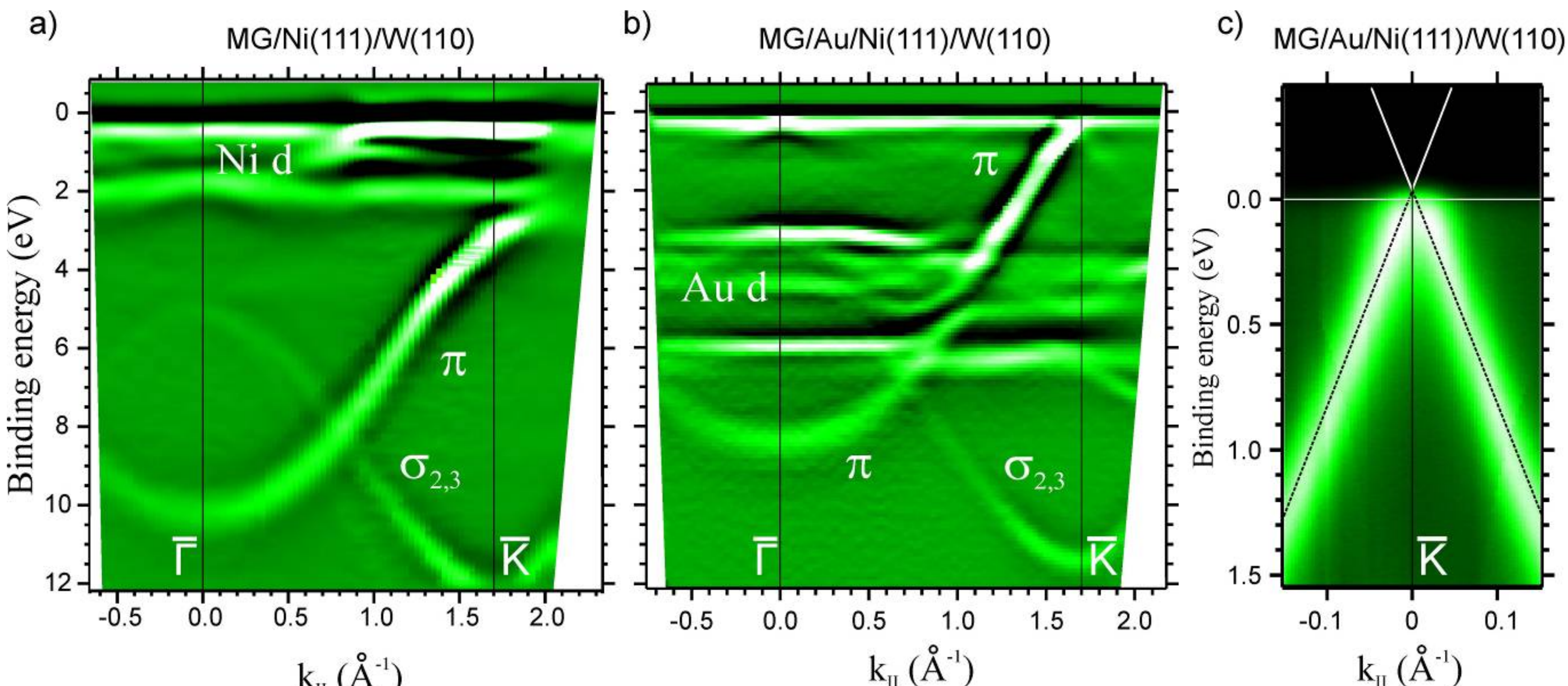
V.M. Karpan et al., PRB 78, 195419 (2007)

V.M. Karpan et al., PRL 99, 176602 (2007)

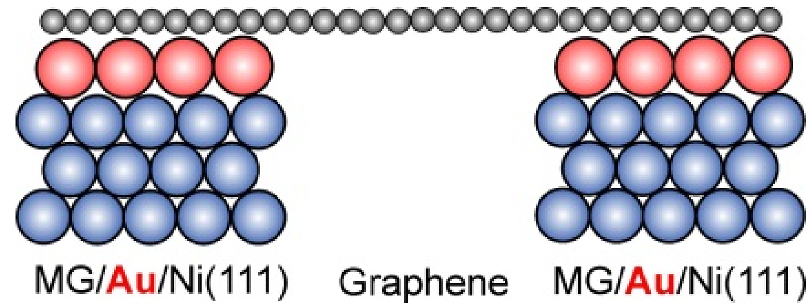
Necessary conditions for active using contact of graphene/Me in spintronics are:

1. Enhanced spin-orbit splitting of graphene π states at the Fermi level
2. Intersection between the graphene π states and spin polarized d states at the Fermi level In the region of the K-point of the BZ

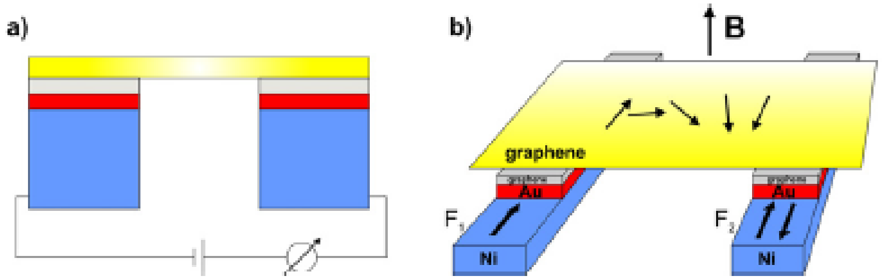


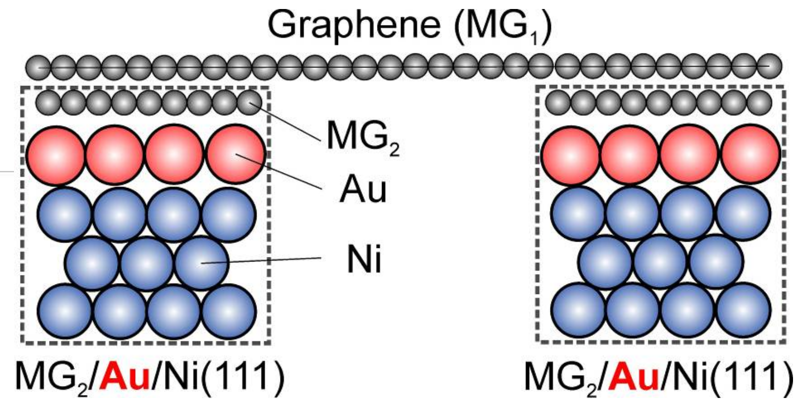


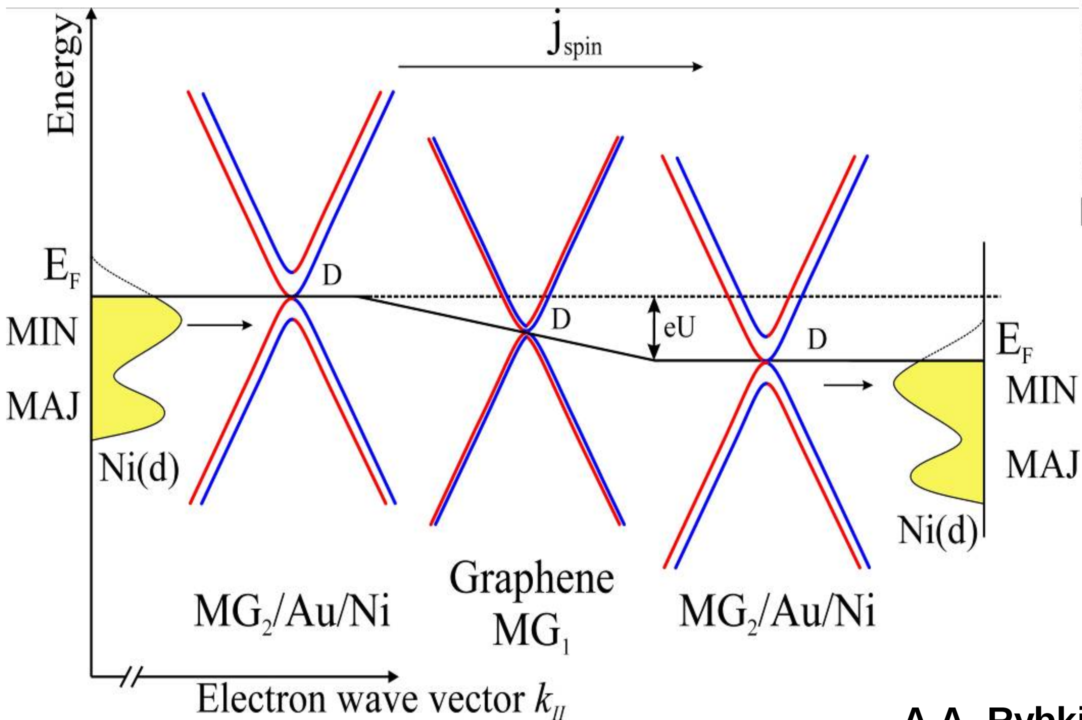
New scheme of graphene spin filter construction



Introduction of Au-monolayer prevents the Dirac cone destruction



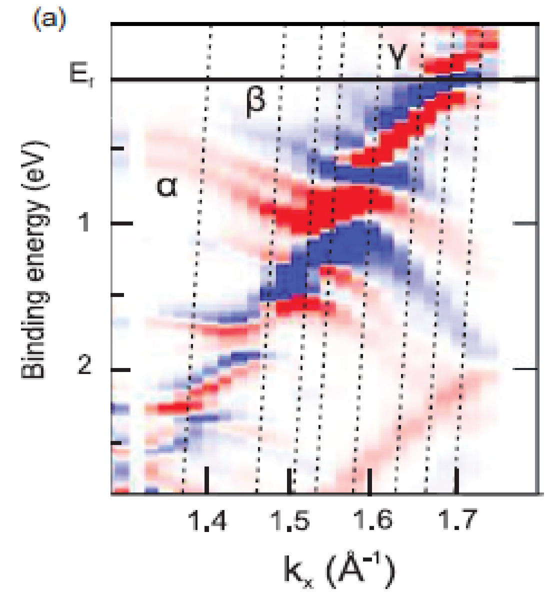




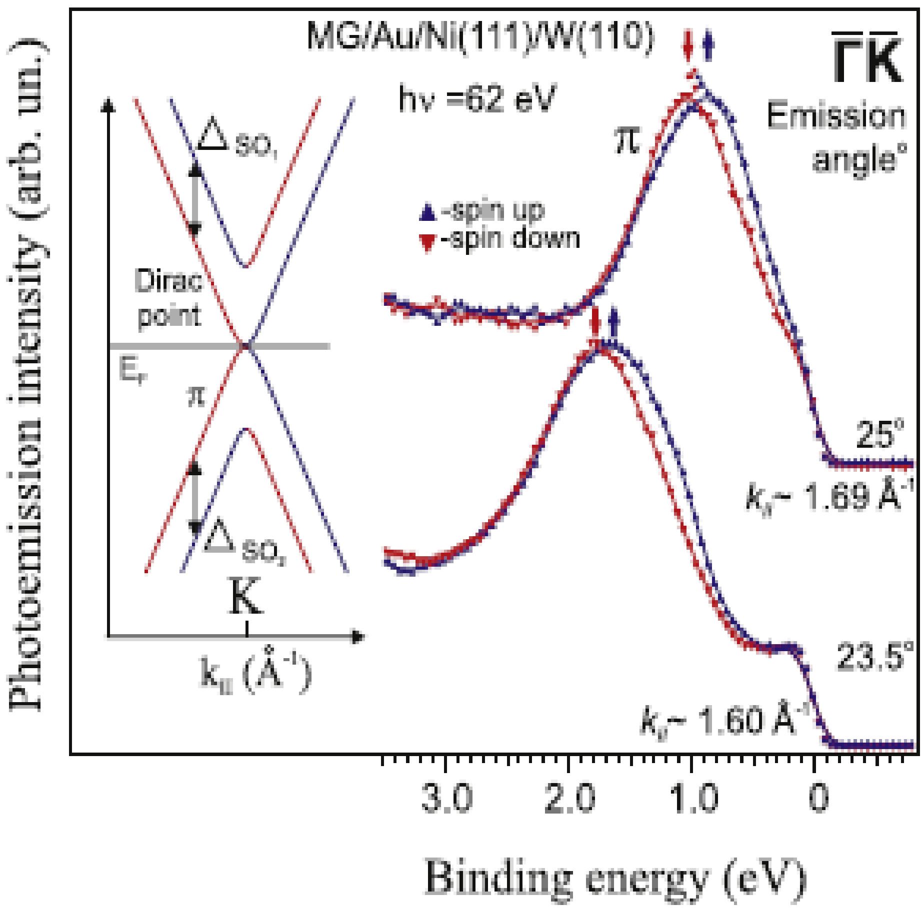
Introduction of additional graphene interlayer solves the problem of returned currents (like thin insulator layer, but with perfect structure (graphene can be considered as an insulator in the direction perpendicular to plane)

A.A. Rybkina et al., Nanotechnology, 24, 295201 (2013).

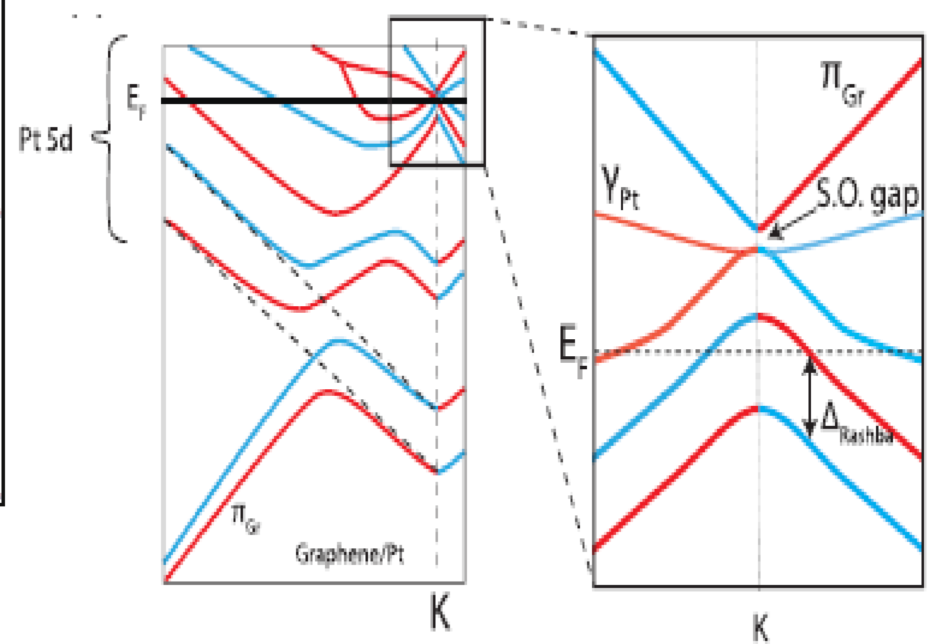
How can the unique spin structure of the Graphene/Au/Ni and Graphene/Pt interfaces be used in spintronics?



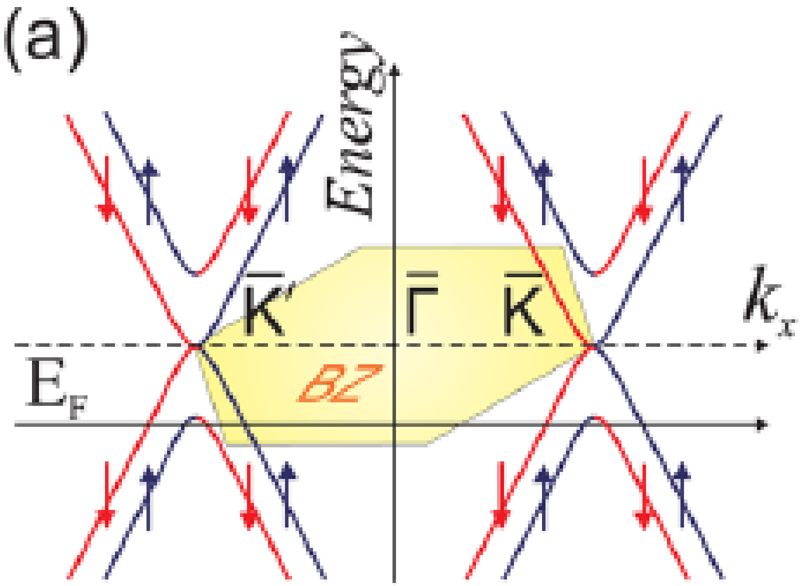
Graphene/Au/Ni



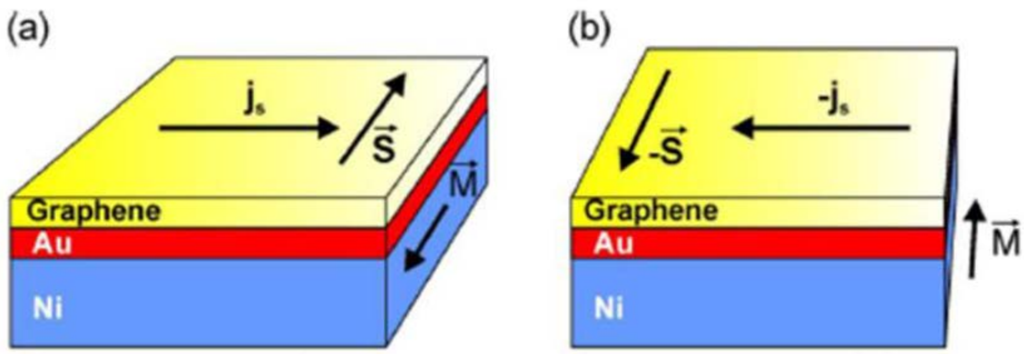
Graphene/Pt

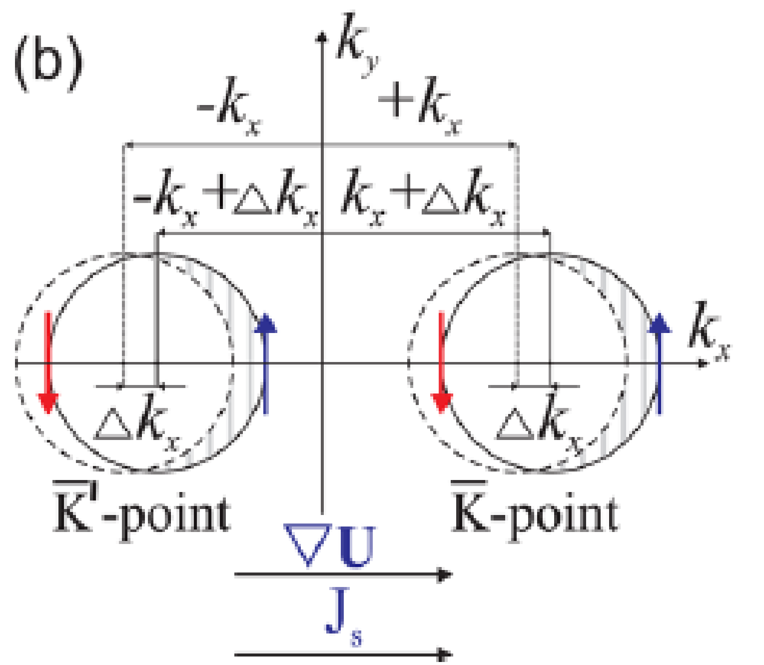


Possibility of spin current formation due applied electrical field.



Spin is strongly locked perpendicular to momentum of electron





Applied reversal electrical field leads to generation of reversal spin-polarized current

A.M. Shikin et al., Appl. Phys. Lett.. 105, 042407 (2014)

Spin current under application of electrical field along surface

Application of an electric field along the *x*-direction causes a spin current along the *x*-axis:

$$j_s^z = \frac{2\alpha}{\hbar\upsilon} j_e^x = \frac{2\alpha}{\hbar\upsilon}\sigma E_x$$

where σ is the graphene conductivity, α is the constant of spin-orbit interaction ($\alpha k_F = 80$ meV) with Hamiltonian $H_{so} = \varepsilon(k_x) - \frac{\alpha}{\hbar}\left(k_x\sigma_y - k_y\sigma_x\right)$ Here $\varepsilon(k_x) = \hbar\upsilon k_x$ is the dispersion law for the electrons of graphene

This leads to spin accumulation: appearance of uncompensated spin density with the spin polarization in the *y*-direction (the spin is locked perpendicular to the momentum):

$$\left\langle \delta\vec{\sigma} \right\rangle_y^{\nabla U} = -\alpha \frac{ne\tau}{\hbar\varepsilon_F} E_x$$

where *n* is the electron density in graphene, τ is the relaxation time. Taking into account that the velocity of electrons at the Dirac point equals to 10^6 we can estimate the dimensionless ratio of the spin and the electric currents in both cases: $\left|j_s\right| / \left|j_e\right| = 0.015$ At the same time in conventional generators of the spin current, based on the spin-Hall effect (SHE), this ratio is of the order of 0.07 or 0.03. Hence, we have comparable efficiency of the spin current excited at the graphene/Pt interface and, for instance, in Pt due to SHE.

We can estimate the value of an electric current as follows:

$$j_e^x \approx 2 \cdot 10^7 \left(\Delta U[mV] / \Delta X[nm]\right)$$

where ΔU is the electrical potential difference between the two contacts in millivolts

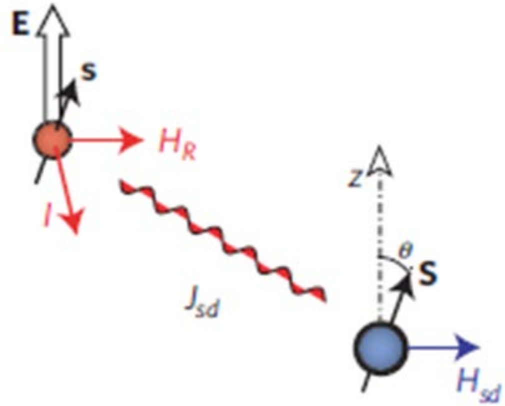
P. Skirdkov, A. Zvezdin

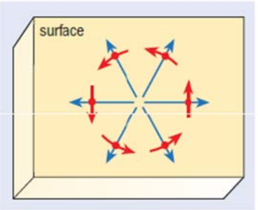
How to use the formed spin current?

It can be used for induced magnetization of ferromagnet nanostripes
due to

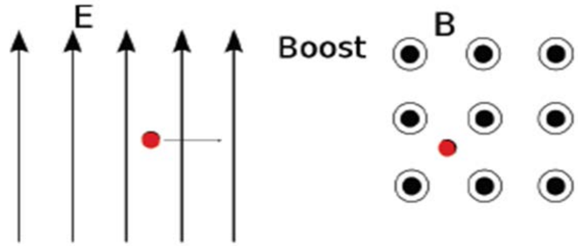
spit torque effect

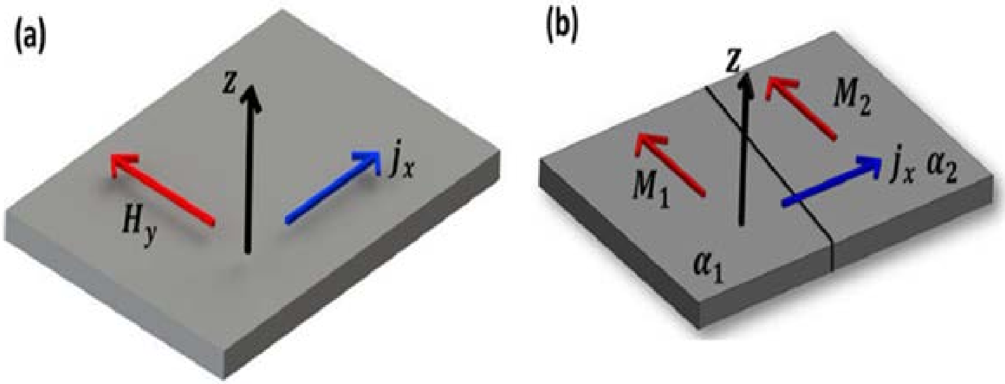
Spin transfer and spin-orbit torque





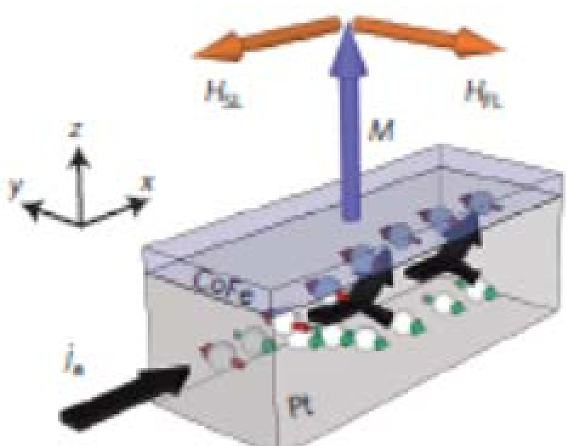
Spin-orbit torque

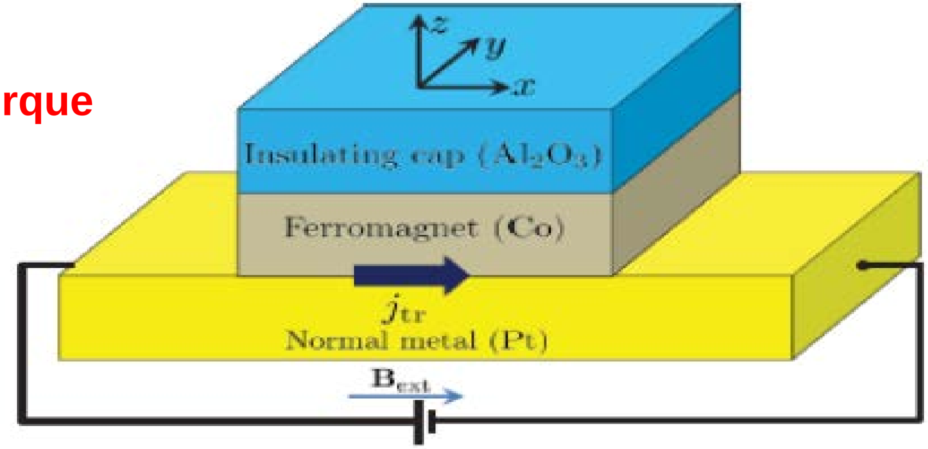




I.M. Miron et al., Nature Materials 9, 230 (2010)

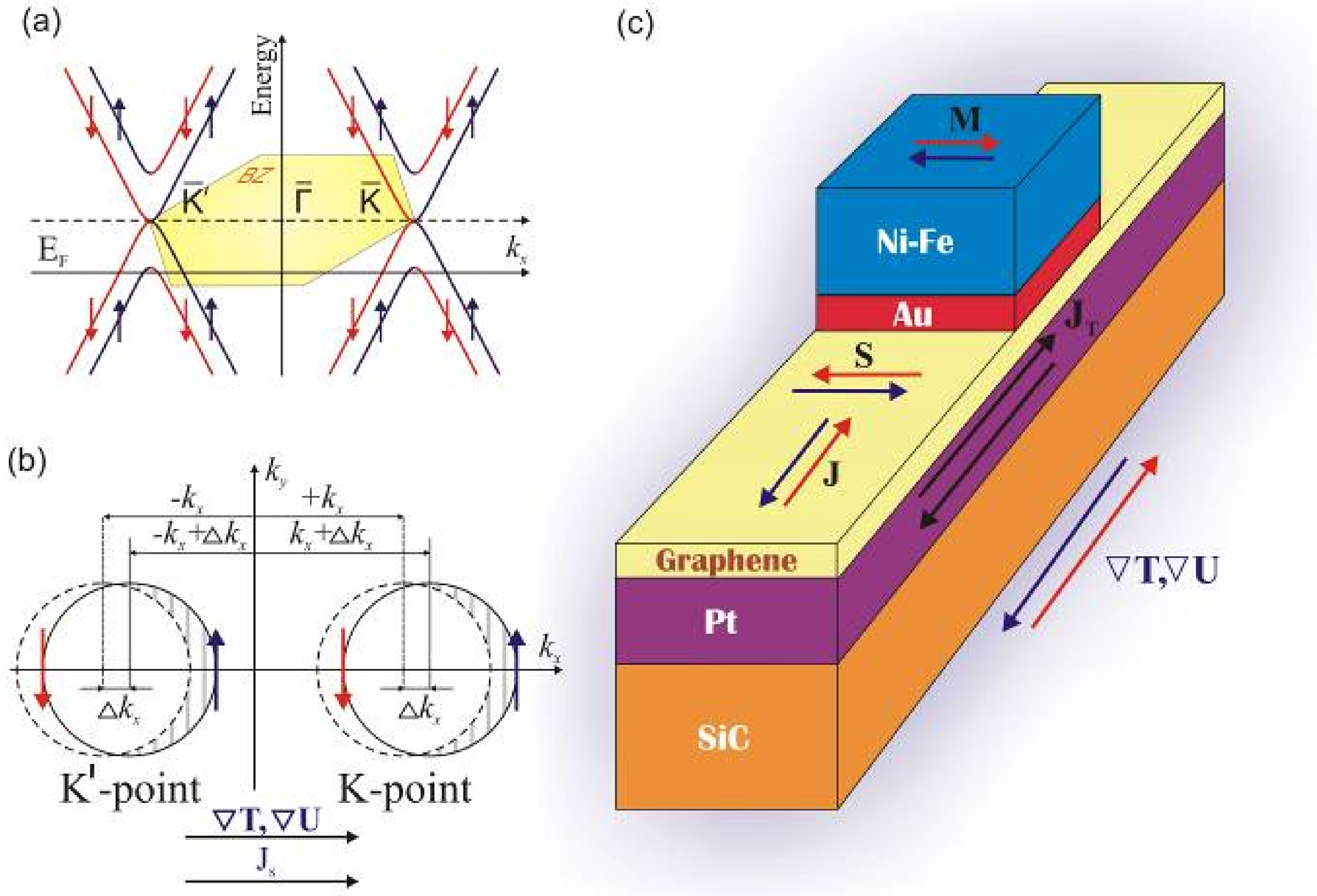
Spin transfer torque





Spin current can lead to the magnetization of FM due to spin-torque effect without external magnetic field

Use of spin current formed at the Graphene/Pt interface for the induced magnetization of Ni-nanostripes



A.M. Shikin et al., Appl. Phys. Lett. 105, 042407 (2014)

Magnetization induced by spin current. Spin-orbit torque effect.

The effective induced magnetic field $\vec{B}_{SO}$:

$$\vec{B}_{SO} = -\frac{\alpha k_F}{e\upsilon M_S} P \cdot j_e \left[\vec{z}^{\,0} \times \vec{j}^{\,0} \right], \quad \text{where } j^0 = 1, \; P = \frac{J}{\varepsilon_F}.$$

The estimations give

$$B_{SO} \sim \frac{\alpha k_F}{e\upsilon} \frac{J}{\varepsilon_F} \frac{1}{n\mu} \cdot j_e \sim 1{,}5 \cdot 10^3 P \, (Oe),$$

i.e. ~100-1000 (Oe)

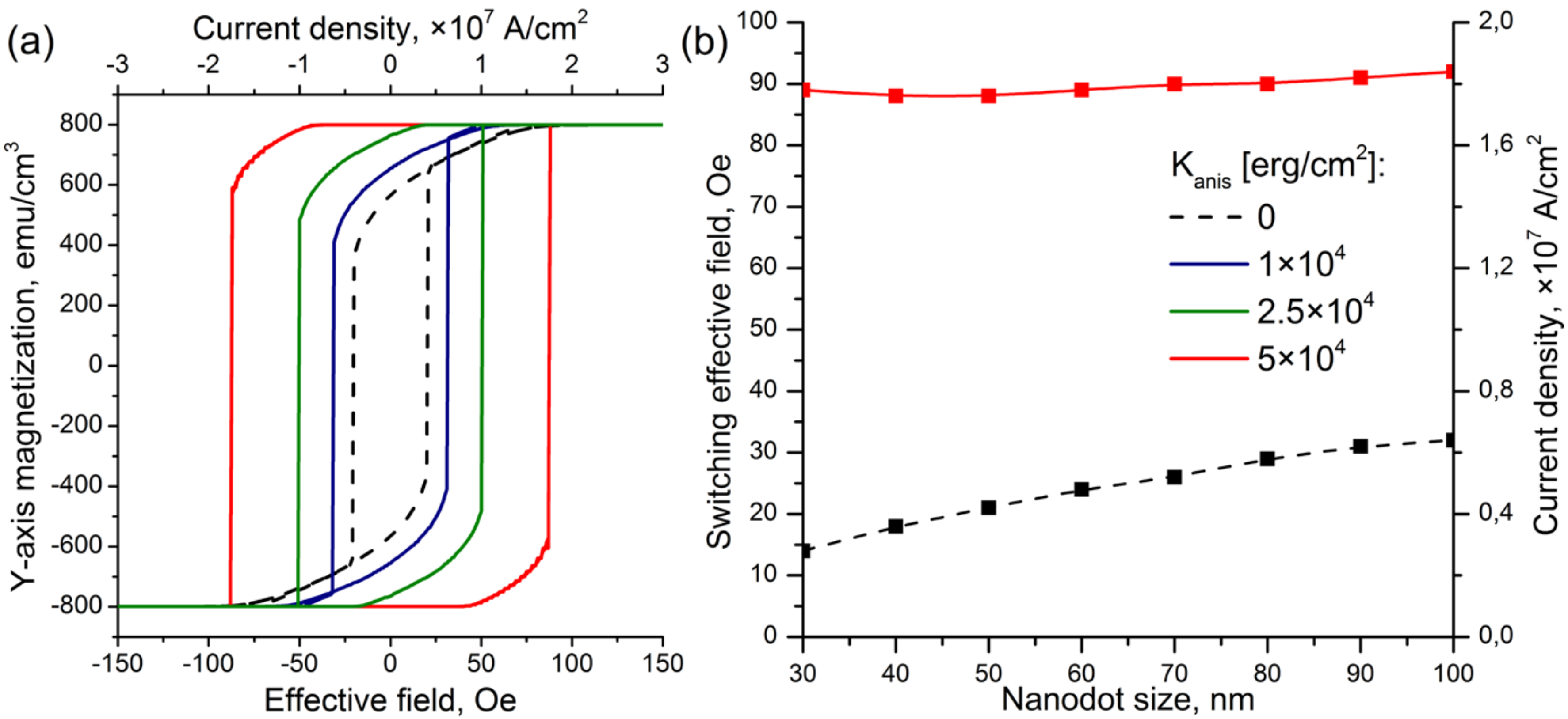
The spin torque, acting on magnetization in Ni layer:

$$\vec{T}_{SO} = \left[\vec{M}, \vec{B}_{SO} \right] = \frac{\alpha k_F}{e\upsilon M_S} P j_e \left[\vec{M}, \vec{j}^{\,0} \right] = \frac{\alpha k_F}{e\upsilon} P j_e \left[\vec{M}^0, \vec{j}^{\,0} \right]$$

P. Skirdkov, A. Zvezdin

A.M. Shikin et al., Appl. Phys. Lett. 105, 042407 (2014)

Hysteresis in magnetization of Ni-magnetic nanostripes due to spin-orbit torque effect



(a) - calculated hysteresis loops for the (Ni-Fe)-nanodot for different values of the anisotropy constant.

(b) - critical remagnetization effective field/current dependens on the nanodot size for hard and soft magnet cases. In the case of a soft magnet (dashed line) the critical switching field is determined by the shape anisotropy.

Thickness – 50 mkm
Width – 75 mkm
Length – 500-1500 mkm

A.M. Shikin et al., Appl. Phys. Lett. 105, 042407 (2014)

Contact of graphene with heavy metal characterized by d-states in the valence band is followed by enhanced induced spin-orbit splitting of the graphene π states.

It is caused by the hybridization between the graphene π states and the metal d-states takes with corresponding spin-dependent avoided-crossing effects

Under interaction with heavy metals with sp-structure of the valence band the induced spin-orbit splitting is reduced significantly

Topological phase can be formed in Graphene/Me systems and modified

Unique spin structure of the interface Graphene/ Pt (Graphene/Au/Ni) and FM can be effectively used in spintronics for induced magnetization of FM by generated spin current

Thank you very much for your attention