

Broken symmetry and spin-split bands at surfaces



Marco Grioni

On the positive side...



- Different ground states may be stable at the surface due to differences in the strength of interactions, dimensionality, symmetry
- surfaces as tuneable model systems

A case of symmetry breaking at surfaces

1. $E(k, \uparrow) = E(-k, \downarrow)$ time-reversal symmetry

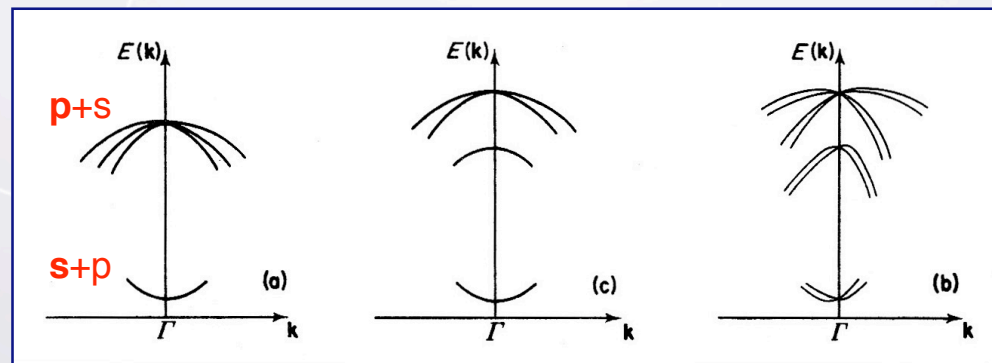
2. $E(k, \uparrow) = E(-k, \uparrow)$ inversion symmetry

1+2. $E(k, \uparrow) = E(k, \downarrow)$ Kramers' degeneracy

at an interface, or at the surface of a solid, inversion symmetry is broken. With SO interaction:

$$E(k, \uparrow) \neq E(k, \downarrow)$$

even without an external magnetic field



no SO

with SO

SO - no inversion

Heine: *Group Theory in Quantum Mechanics*

2D free-electrons - the Rashba effect

now also observed in 1D → talk by F.Himpsel

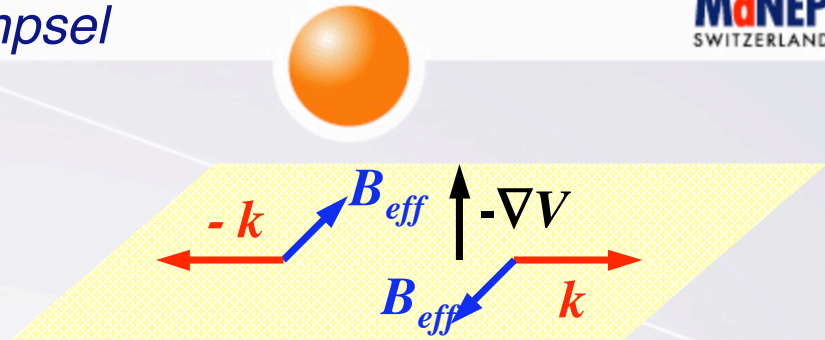


2D free electrons moving in an E field

$$\vec{E} = -\nabla V = -\frac{dV}{dz}\vec{e}_z$$

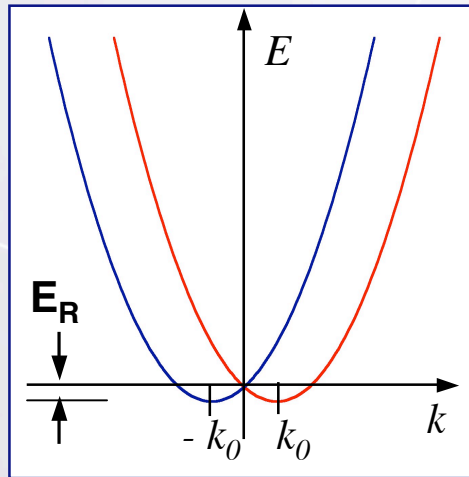
“see” a magnetic field

$$\vec{B}_{eff} = \frac{1}{c^2}\vec{v} \times \vec{E} = \frac{\hbar}{m^*c^2}\vec{k} \times \vec{E}$$



Zeeman coupling

$$H_{SOC} \approx -\vec{\mu}_S \cdot \vec{B} = \alpha_R(\vec{e}_z \times \vec{p}) \cdot \sigma$$

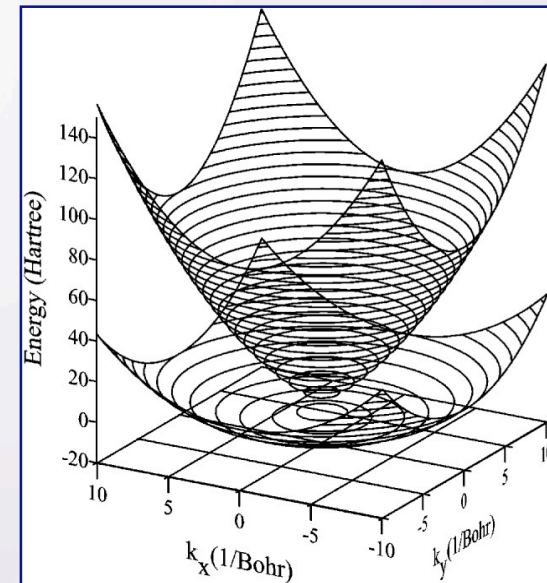


expected Rashba energy
 $E_R \sim 10^{-6}$ eV!

$$E = \frac{\hbar^2 k^2}{2m^*} \pm \alpha_R k$$

$$\alpha_R = \frac{\hbar^2 k_0}{m^*}$$

$$E_R = |E(\pm k_0)|$$



rotational invariance

Influence of atomic parameters: TB approach

The E field is largest near the nuclei

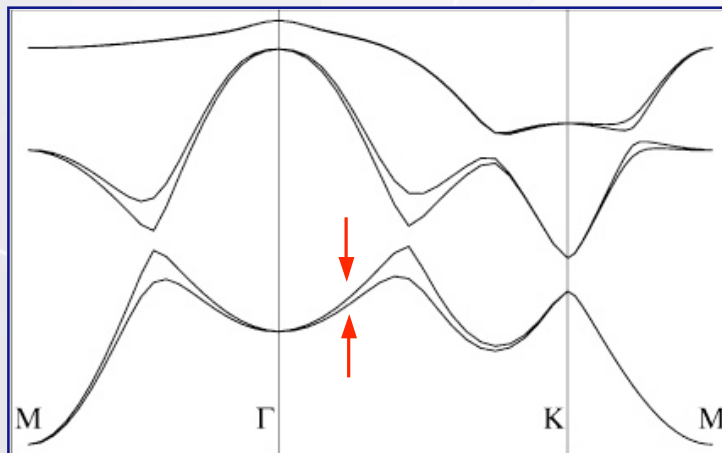
The intra-atomic SO interaction
MUST be important

$$\alpha_R \approx \alpha_{AT} \cdot \left(\frac{dV}{dz} \right)$$

intra-atomic SO
parameter

surface
term

SO split TB bands



Petersen and Hedegård (2000)

Much larger splittings are
then possible

Shockley states at the surface of noble metals

Au(111)

$$k_0 = 0.012 \text{ \AA}^{-1}; \alpha_R = 0.33 \text{ eV \AA}$$

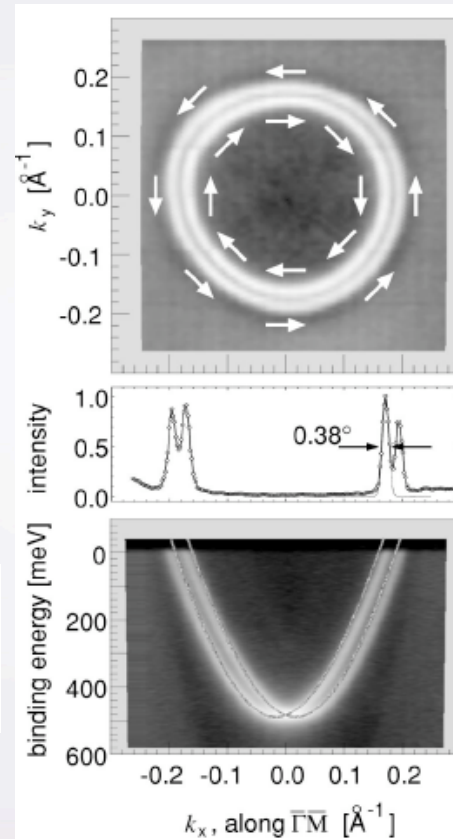
$$E_R \sim 2 \text{ meV}; \Delta E_{SO} = 470 \text{ meV}$$

Ag(111)

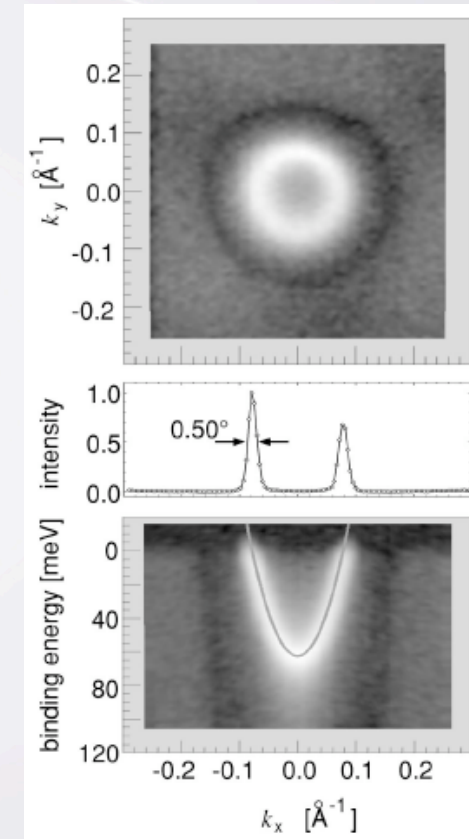
$$k_0 < 0.004 \text{ \AA}^{-1}; \alpha_R \sim 0.03 \text{ eV \AA}$$

$$E_R < 0.2 \text{ meV}; \Delta E_{SO} = 110 \text{ meV}$$

Au (111)

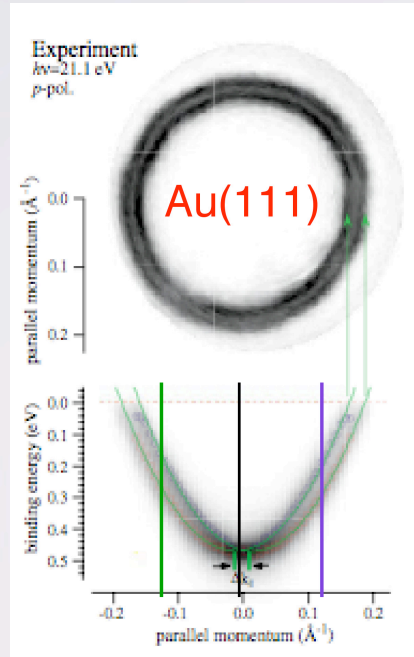


Ag (111)

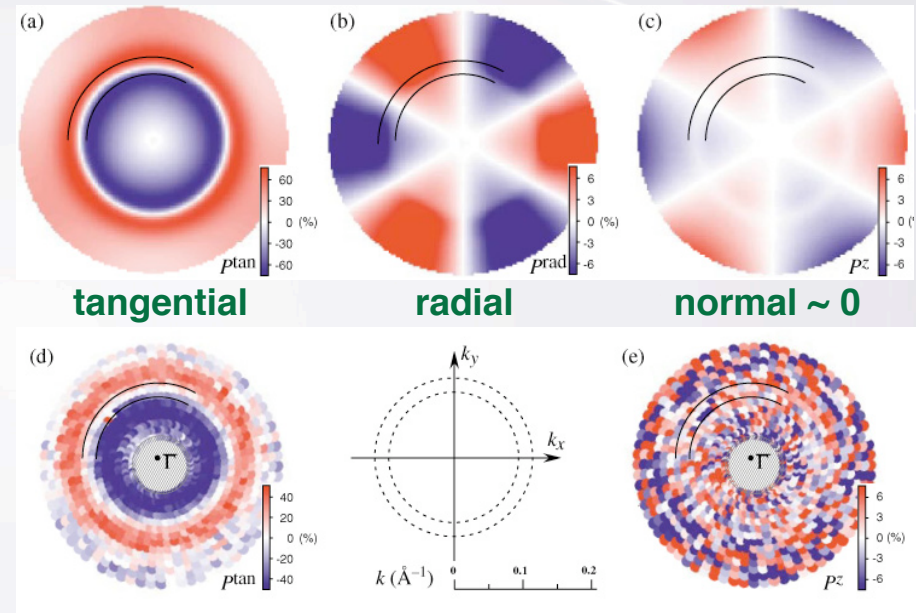


Reinert (2001)

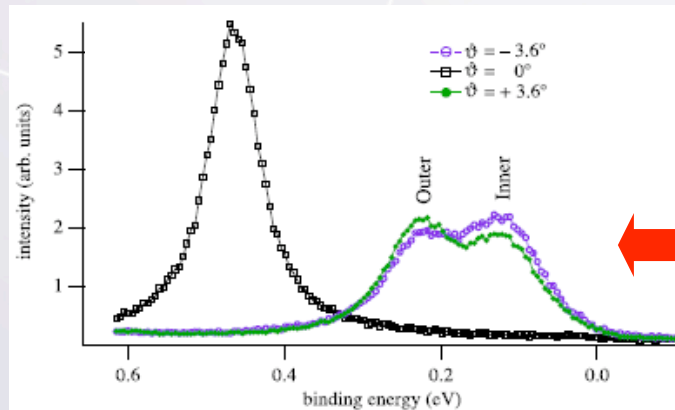
The spin configuration



Spin polarization



Spin-resolved PES : Hoesch, Osterwalder



“linear dichroism” in ARPES

Good agreement with theory confirms the predicted spin configuration

Henk (2004)

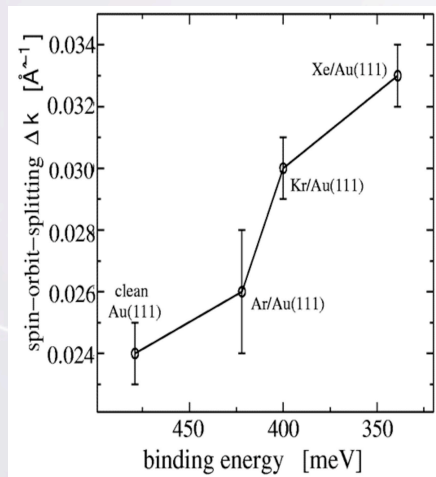
Strategies to enhance the SO splitting

Modifying the surface barrier

Alkali adsorption

Rotenberg (1999)

Rare-gas adsorption

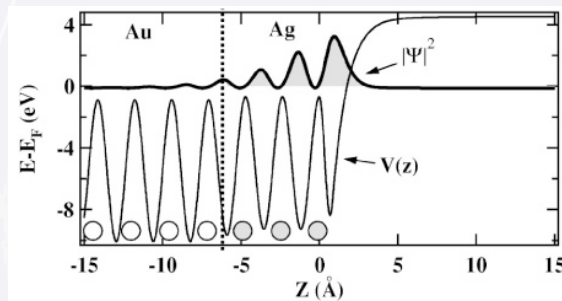


Forster (2004)

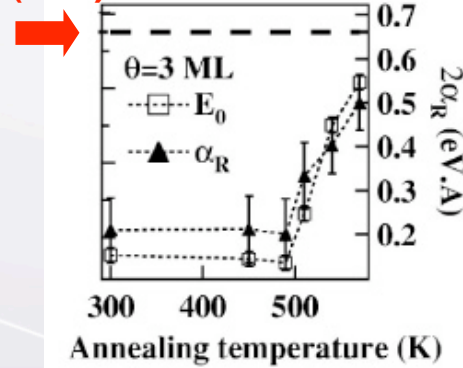
Moreschini et al., to be published (Lausanne-Würzburg)

Alloying

Au/Ag (disordered) alloy



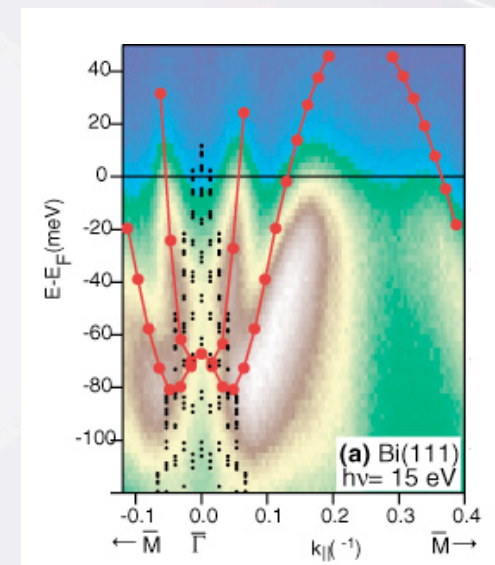
Au(111)



Cercellier (2004)

Other high-Z metals

Bi(111)

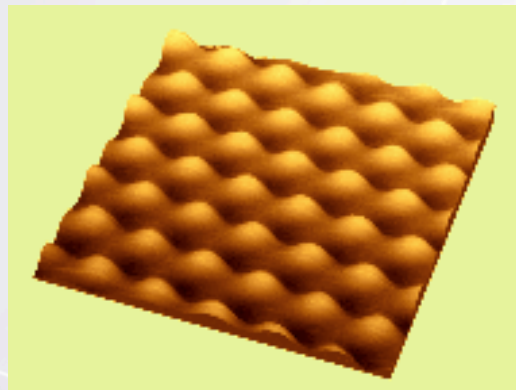


Koroteev (2004)

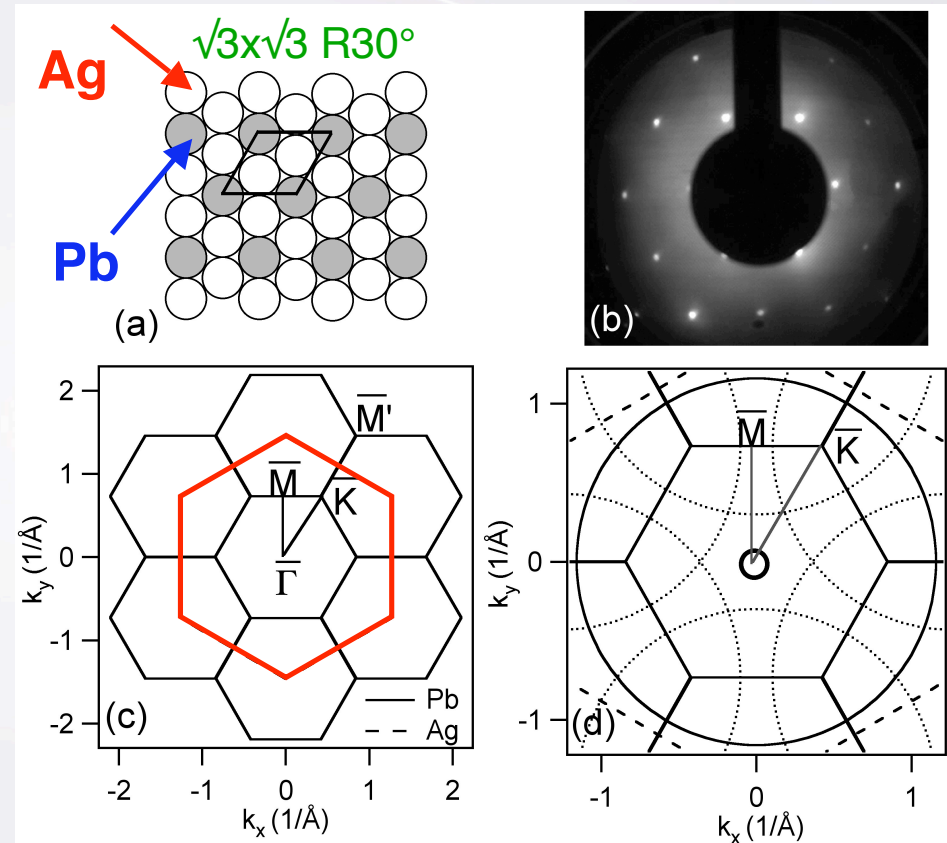
$k_0 \sim 0.05 \text{ \AA}^{-1}$
 $\Delta E_{SO} = 1.5 \text{ eV}$
(Z = 83)

An ordered surface alloy: Pb/Ag(111)

The deposition and reaction of 1/3 ML Pb yields an ordered PbAg_2 surface alloy

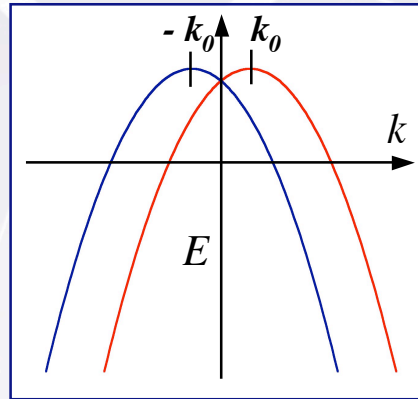
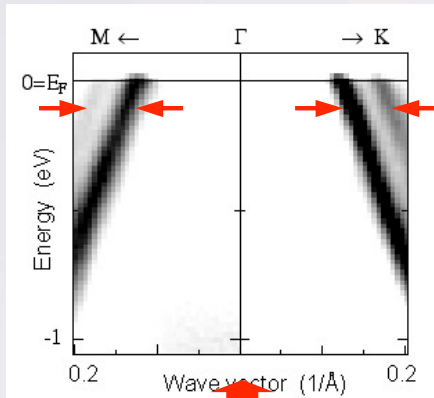


Dalmas (2005)



Split bands in the PbAg₂ surface alloy

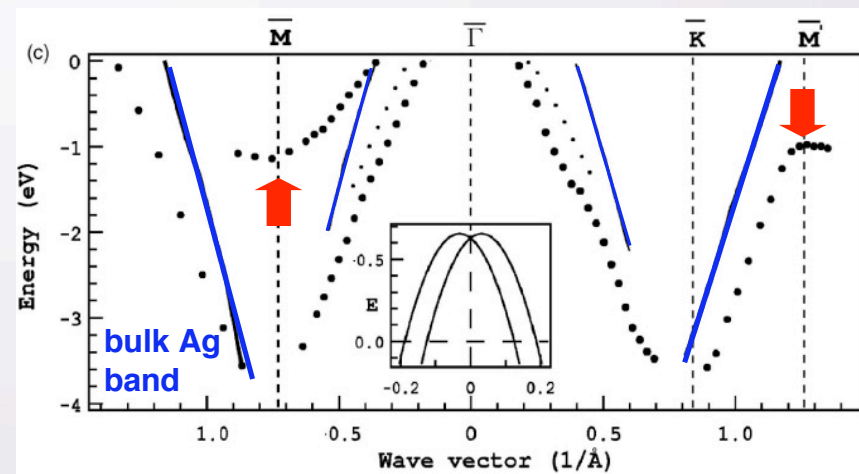
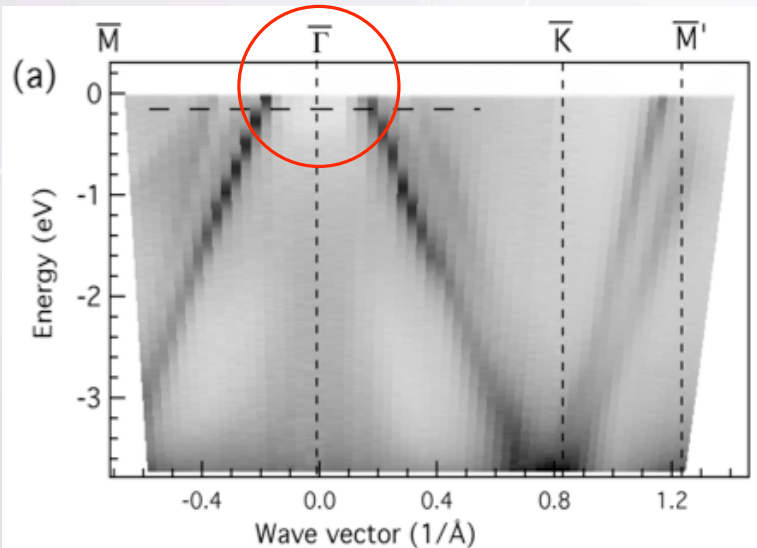
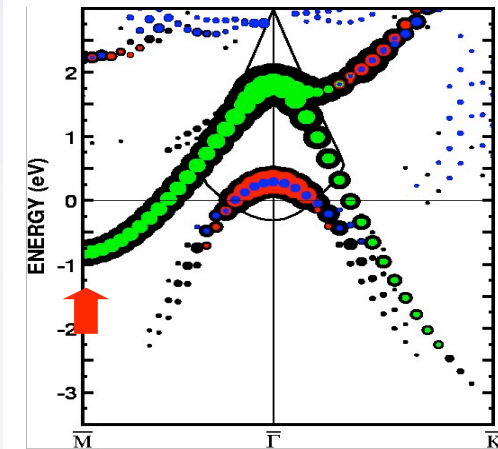
$k_0 = 0.03 \text{ \AA}^{-1}$;
[Bi(111): 0.05 \AA^{-1}]



Nearly-filled (mostly Pb 6p_z) hybrid bands

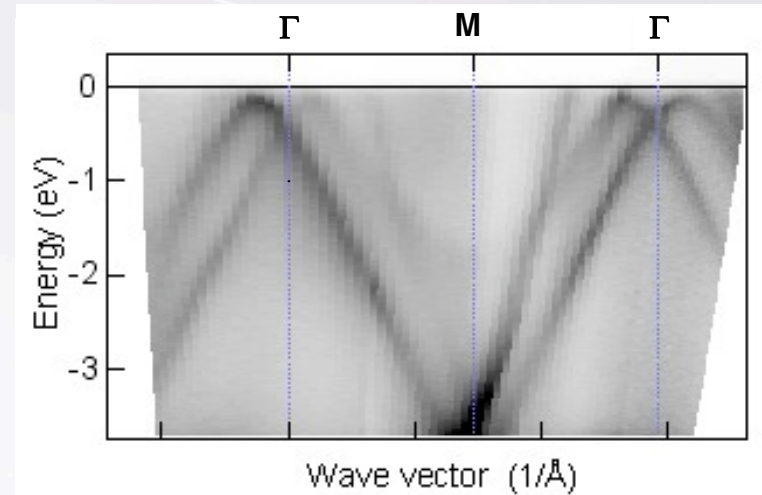
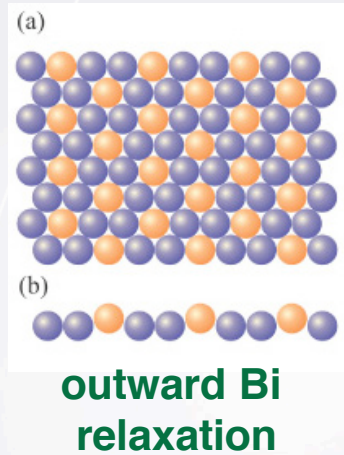
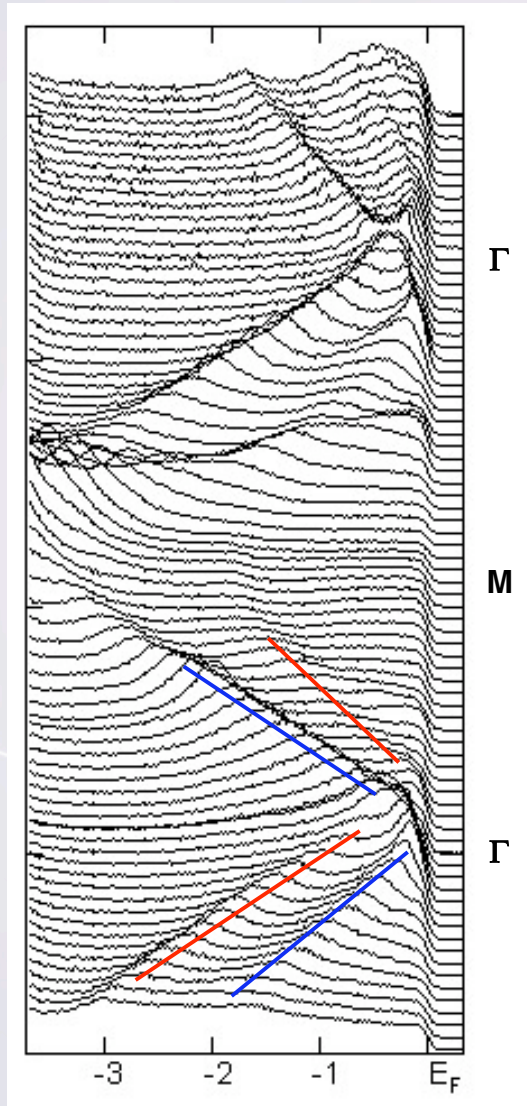


DFT-GGA: no SO
A. Seitsonen



The dispersion is well reproduced by the (slab/supercell) calculation

Pb + 1e/ = Bi; rigid-band model

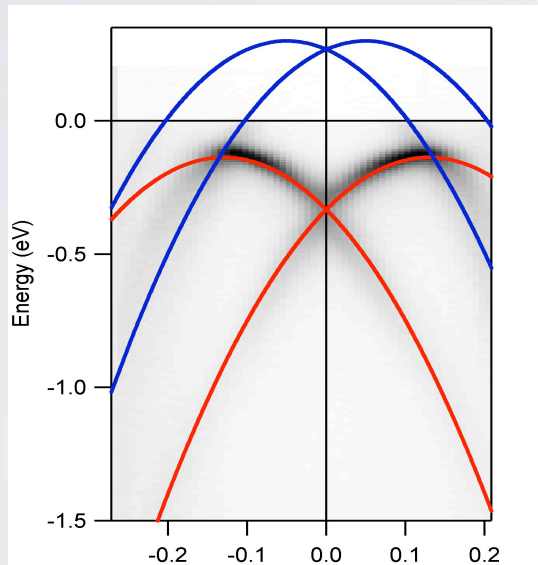


Same structure: BiAg₂ surface alloy

Rigid band shift to accommodate the extra electron

The splitting of the bands is further increased by a factor 4 (the atomic SO parameter is 40% larger)

Giant spin-orbit splitting



Ast *et al.*, to be published in PRL

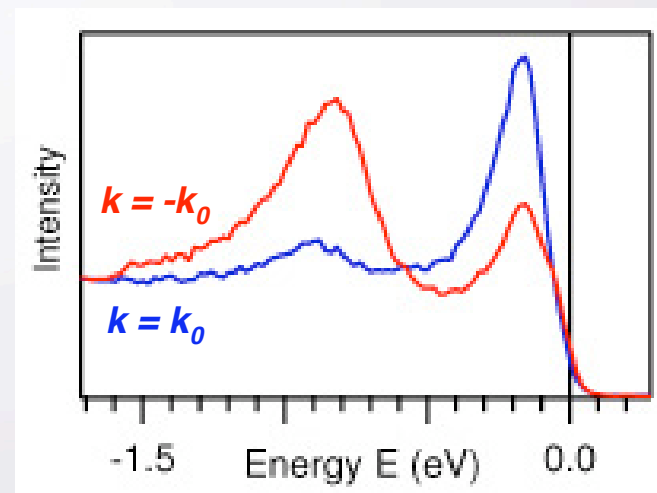
Is it SO splitting ?

What is the origin of the huge enhancement?

$$k_0 = 0.13 \text{ \AA}^{-1}; \alpha_R \sim 3 \text{ eV \AA}; \\ E_R = 200 \text{ meV}$$

$$\text{Au(111): } k_0 = 0.012 \text{ \AA}^{-1}; \alpha_R = 0.33 \text{ eV \AA}; \\ E_R \sim 2 \text{ meV}$$

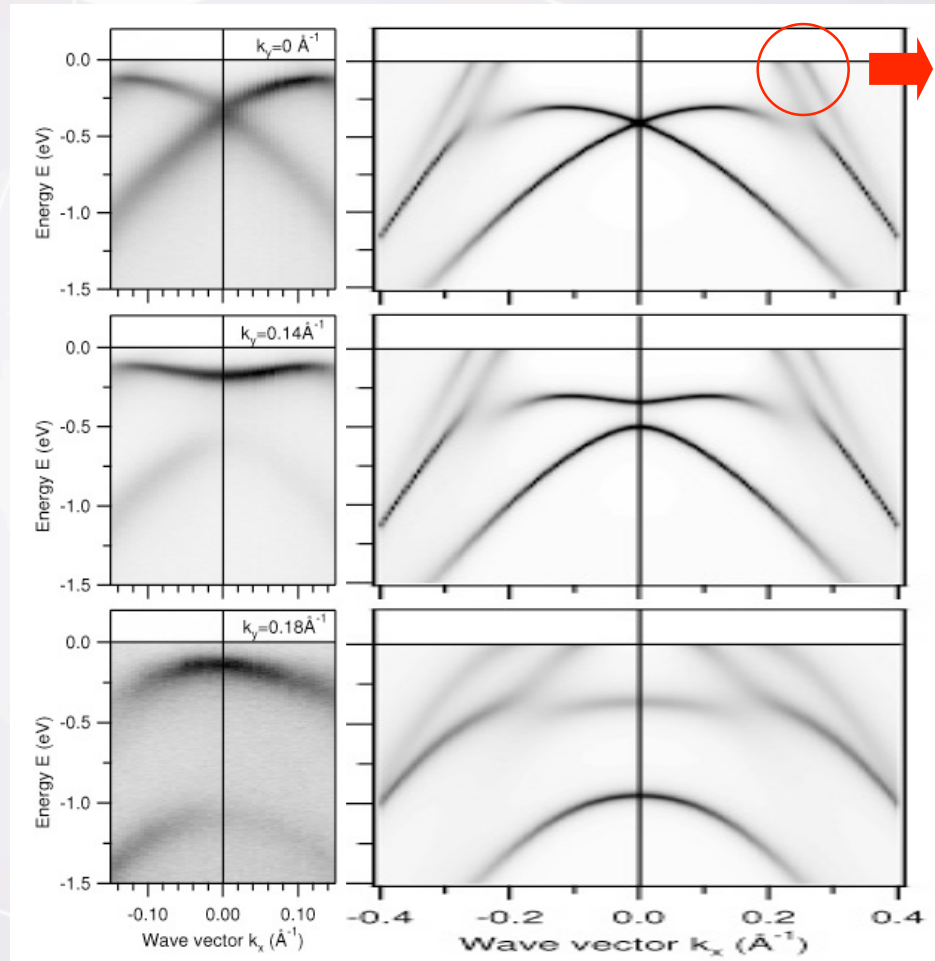
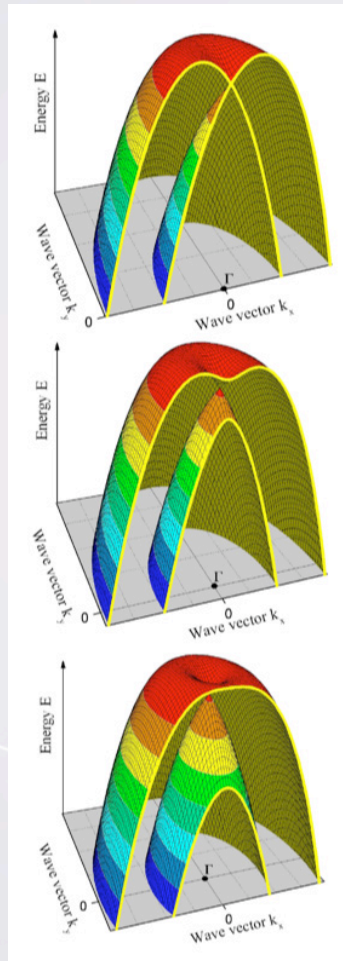
$$\text{Bi(111): } k_0 = 0.05 \text{ \AA}^{-1}; \alpha_R = 0.56 \text{ eV \AA}; \\ E_R = 14 \text{ meV}$$



dichroism in the angular distribution

Theory comes to the rescue

ARPES



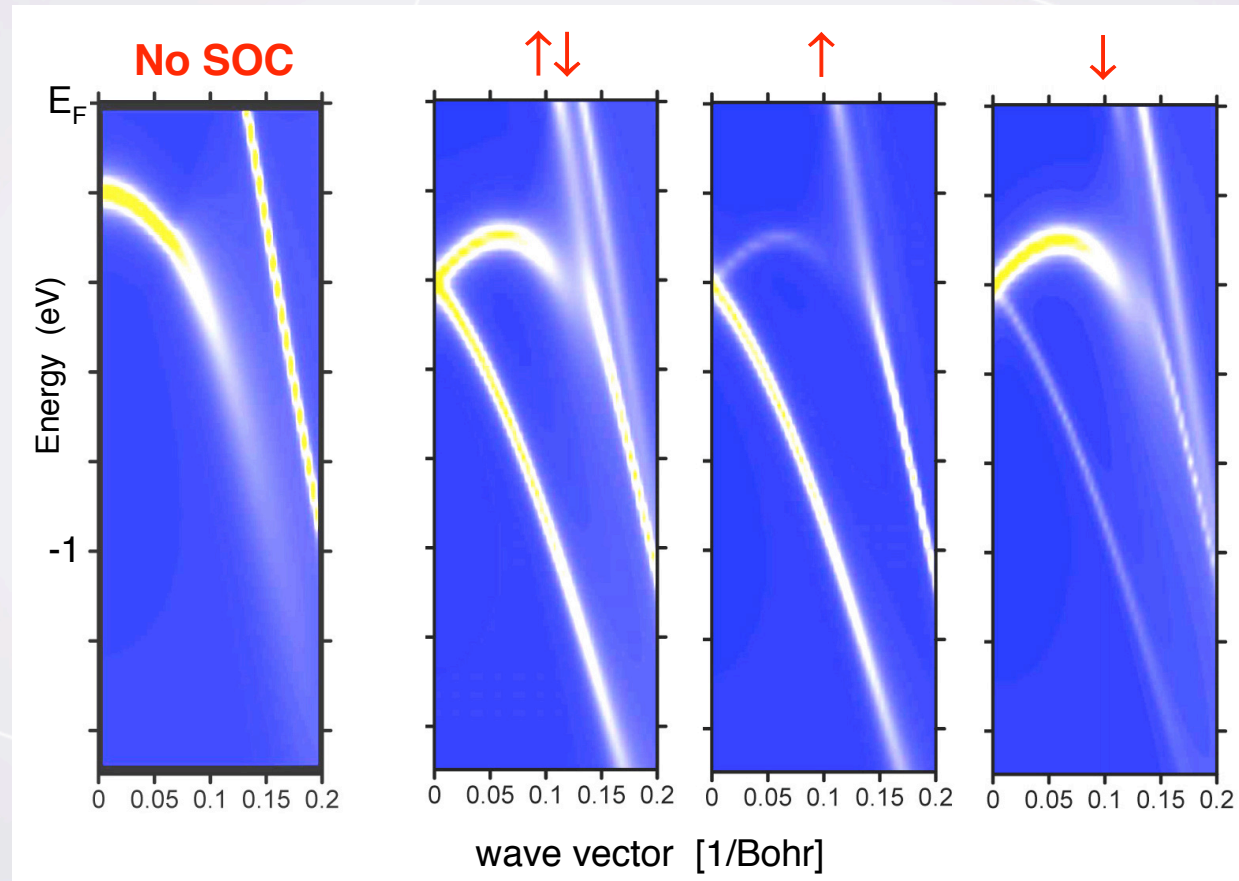
Bi p_{xy}

Fully relativistic layer KKR calculation - J. Henk
also G. Bihlmayer (PRB 2007)

The bands are spin-polarized



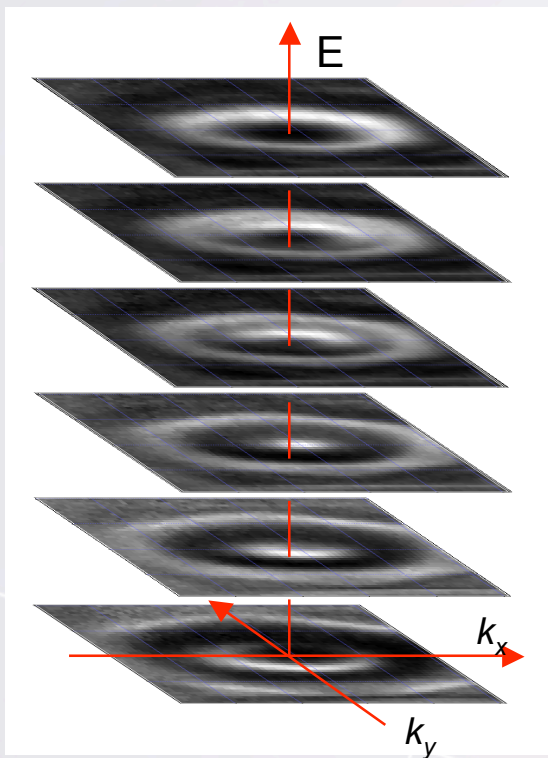
Fully relativistic



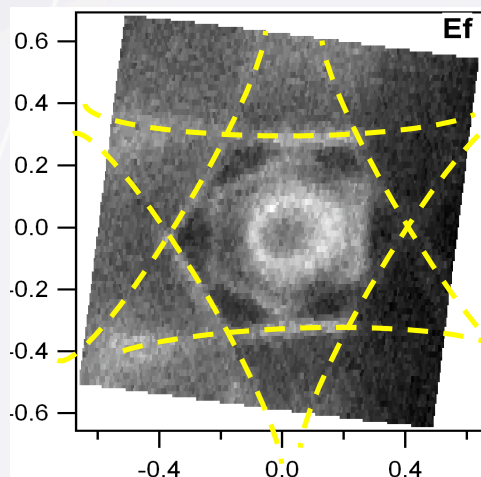
The hybrid states are tightly confined within the top layer

Beyond the Rashba model

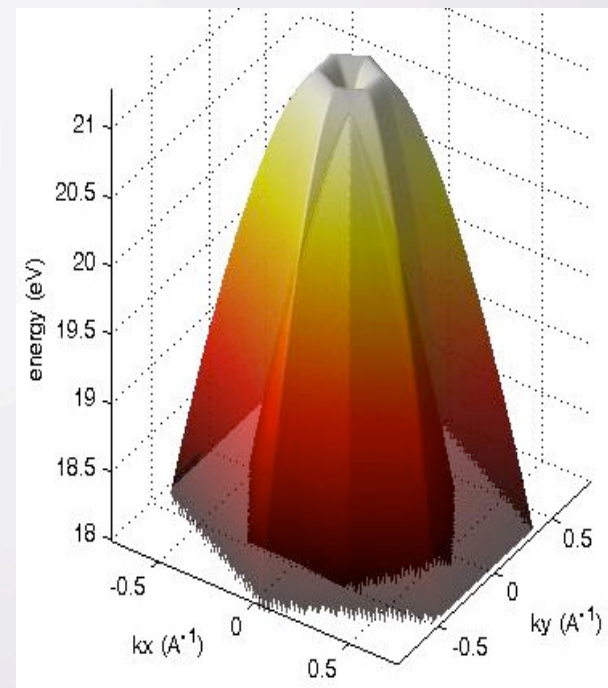
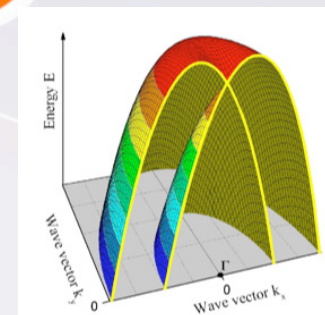
ARPES
constant energy cuts



Fermi surface



+ arcs of unklapped
bulk Ag Fermi surface



Unlike the case of Au(111), the spin-split FS is clearly influenced by the crystal potential

More realistic

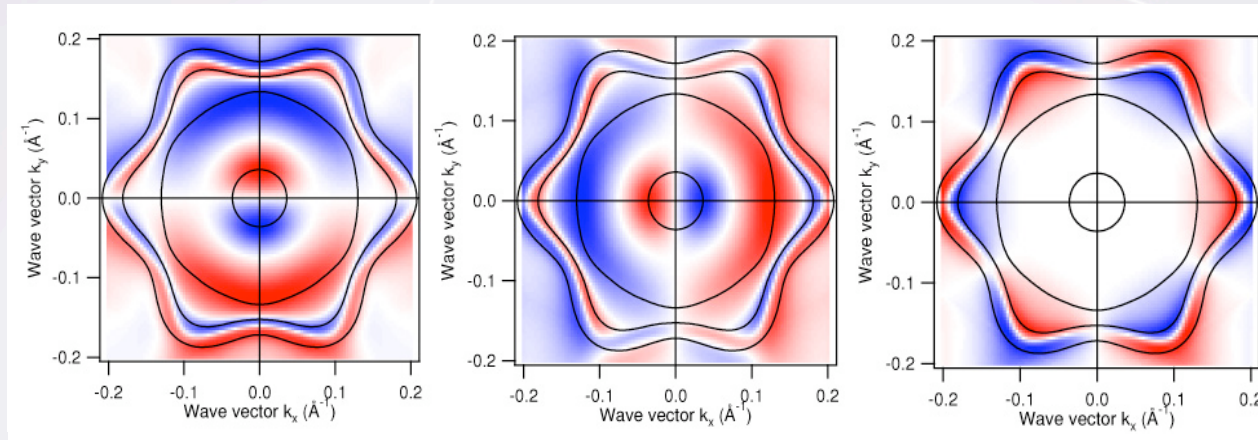
Beyond the Rashba model

**Spin
polarization**

P_x

P_y

P_z



Non-circular Fermi surface

**Out-of-normal component (10%) of the spin polarization
demonstrates an in-plane potential gradient**

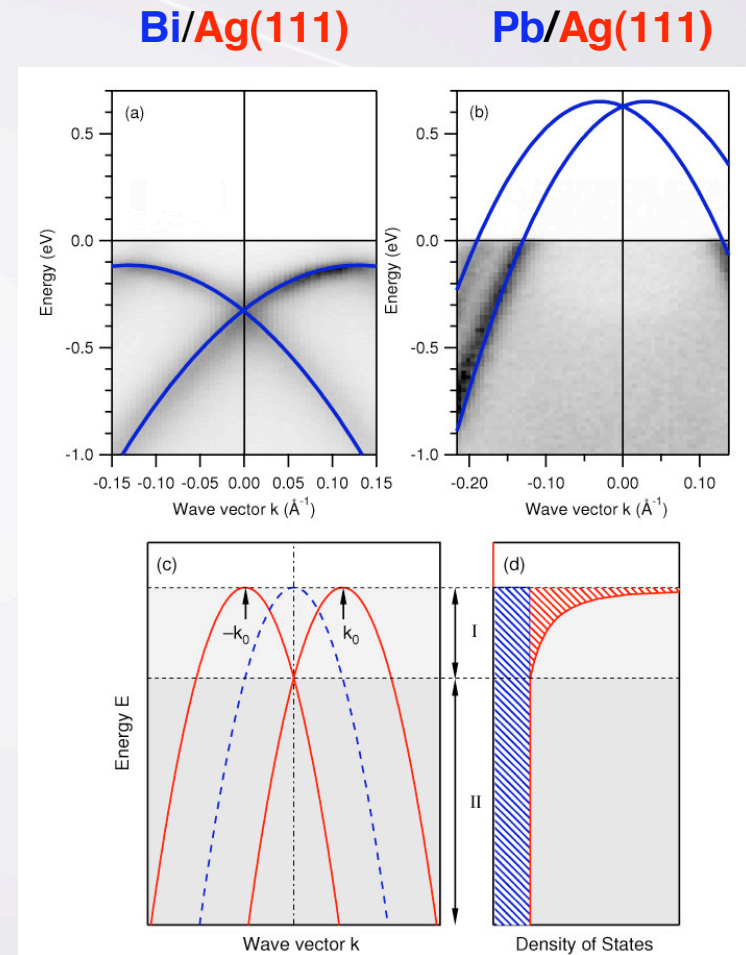
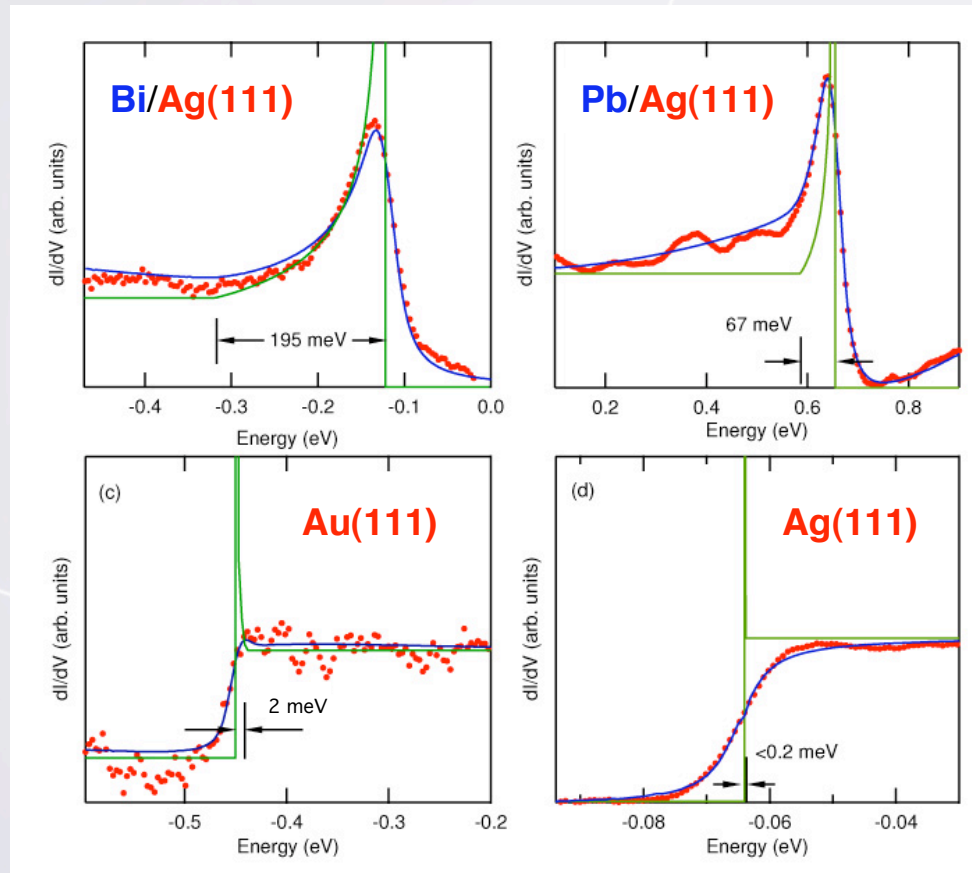
**The inhomogeneous charge distribution within
the surface alloy is the origin of the in-plane gradient**

Chemical bonding is a new “knob” to tune the spin splitting

Spin splitting and the density of states



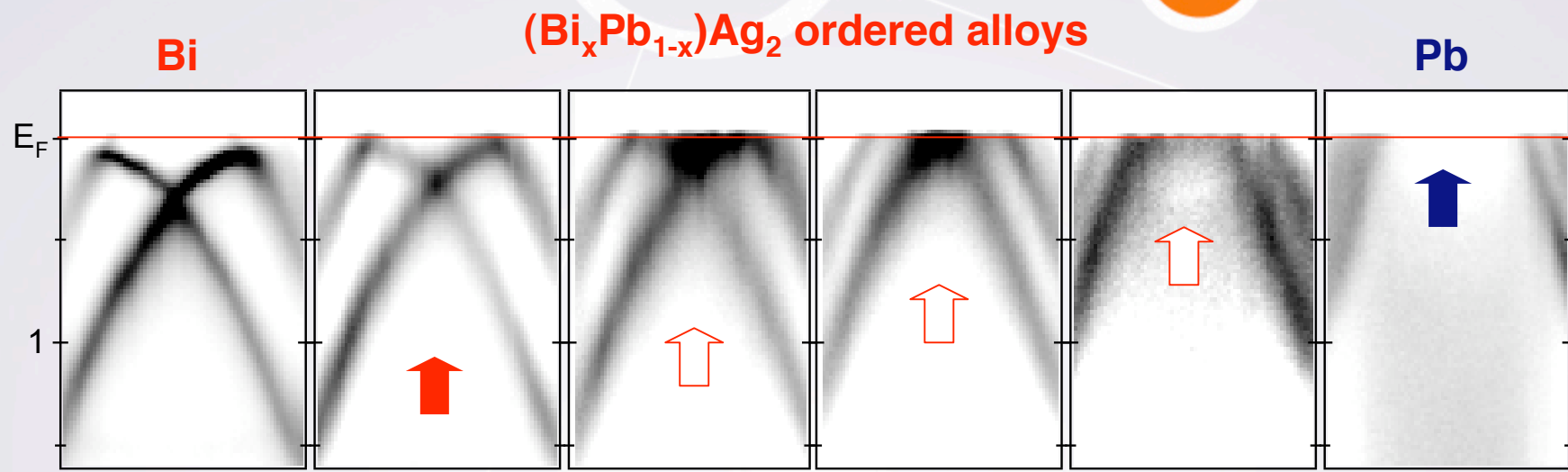
Signatures of spin-split states in tunneling spectra



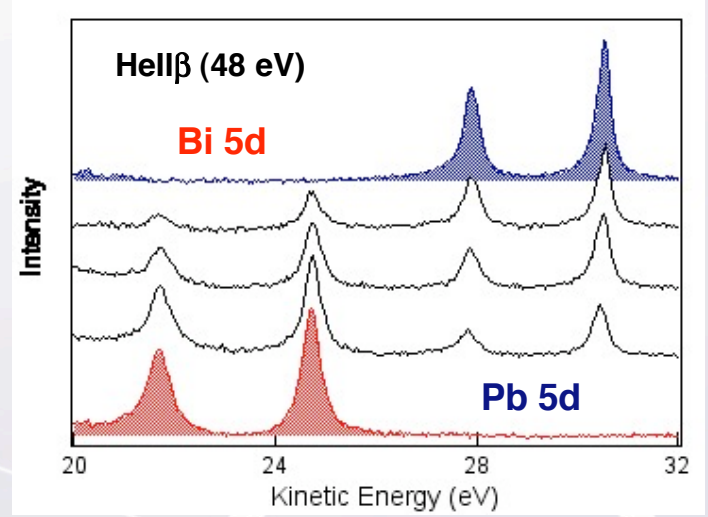
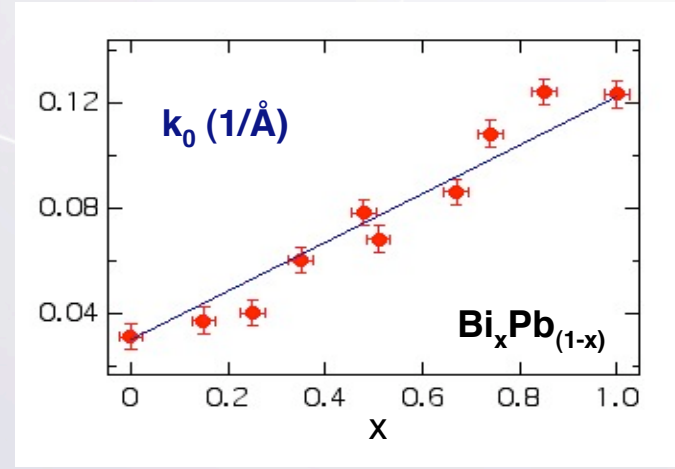
STS: MPI Stuttgart

“1D-like DOS”

Tuning the chemical potential and the SO splitting



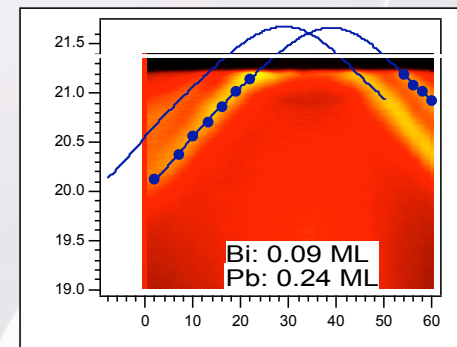
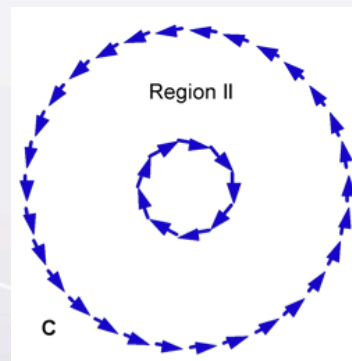
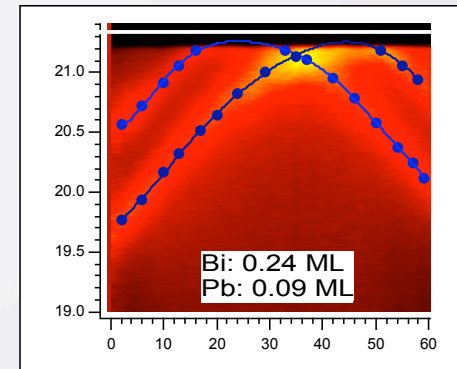
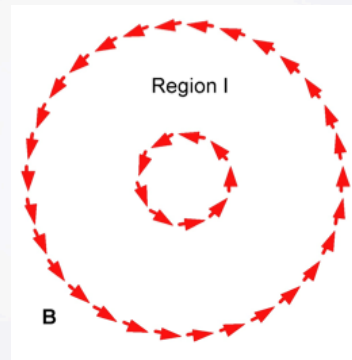
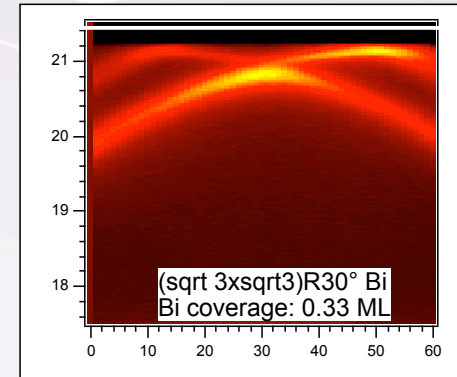
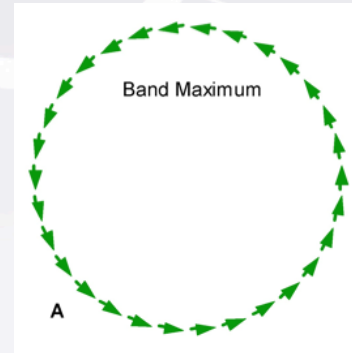
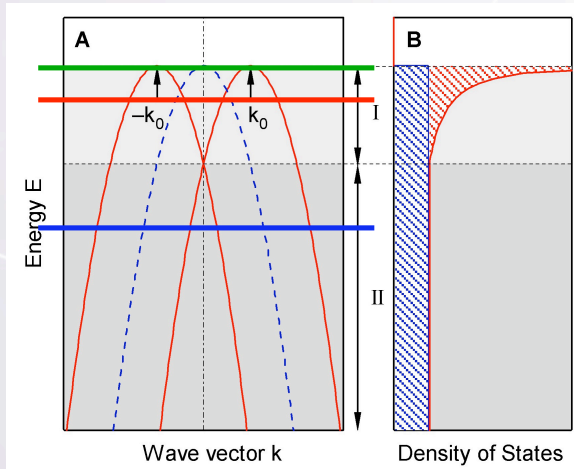
Internal calibration from core levels



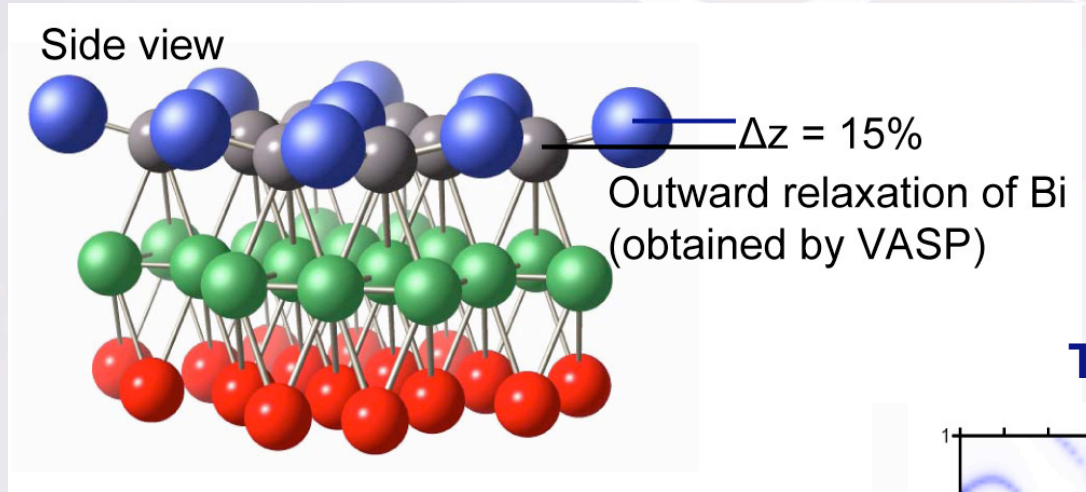
Adjusting the spin pattern by interface engineering

The spin pattern depends on the energy

Three different situations are realized by tuning E_F through the band as a function of stoichiometry



The true surface structure is crucial

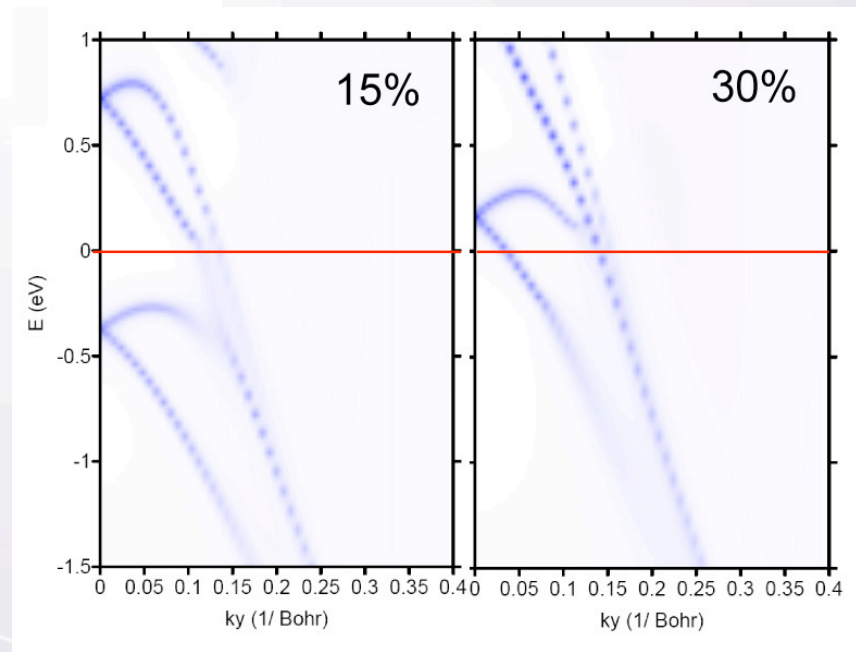


Outward relaxation

True

Larger

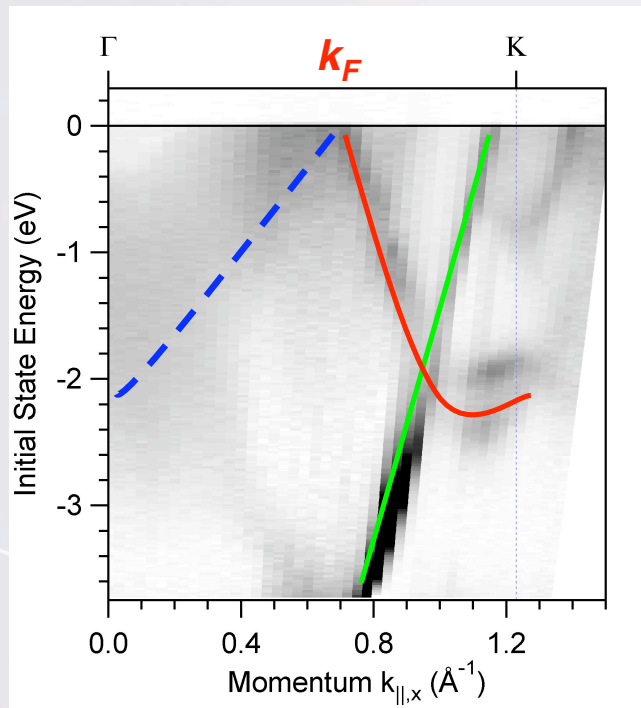
The band structure critically depends on the exact atomic position



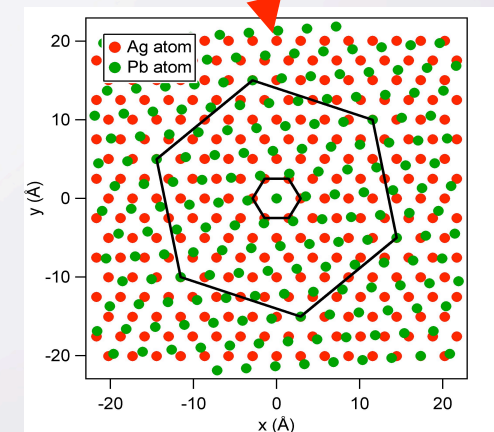
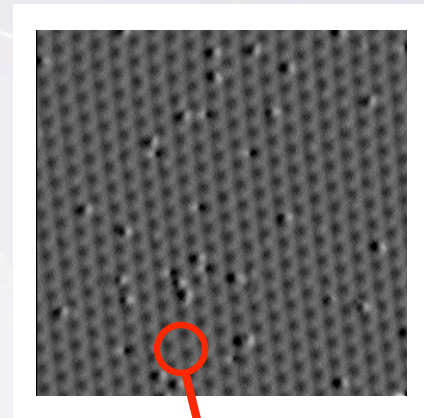
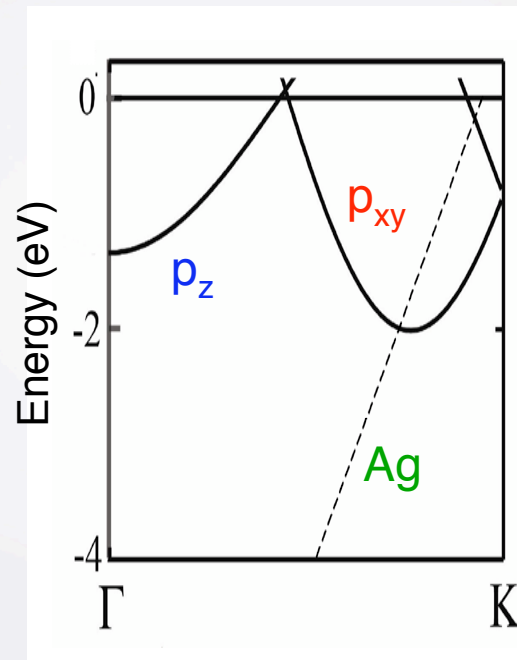
A radical change: 1ML Pb/Ag(111)

STM : G. Wittich
(MPI-Stuttgart)

No sign of SO splitting



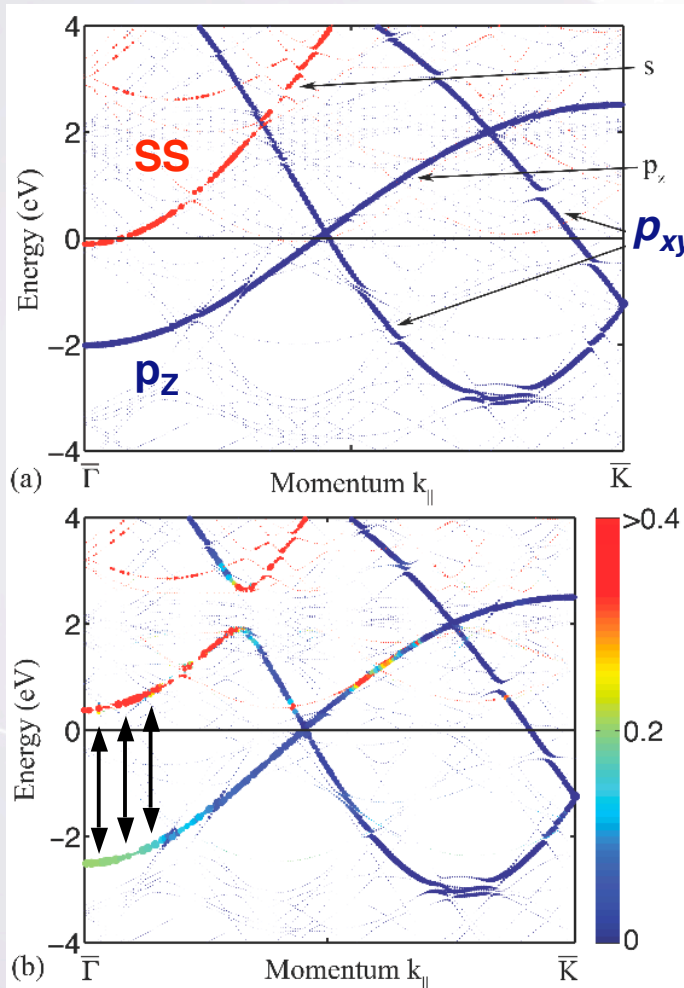
Tight-binding bands



The Pb p_z band is disrupted.
Spectral weight is spread in E and k

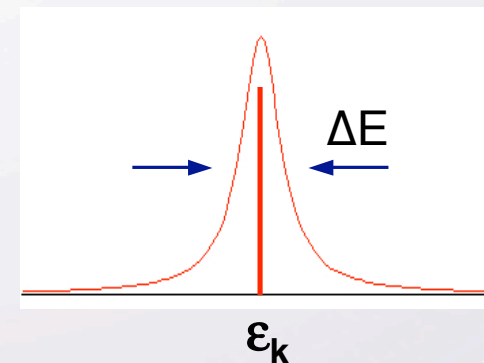
moiré structure
 $\approx \sqrt{28} \times \sqrt{28} R(\pm 19.1^\circ)$
19 Pb ATOMS, 28 Ag ATOMS

Selective hybridization with bulk via the Ag SS



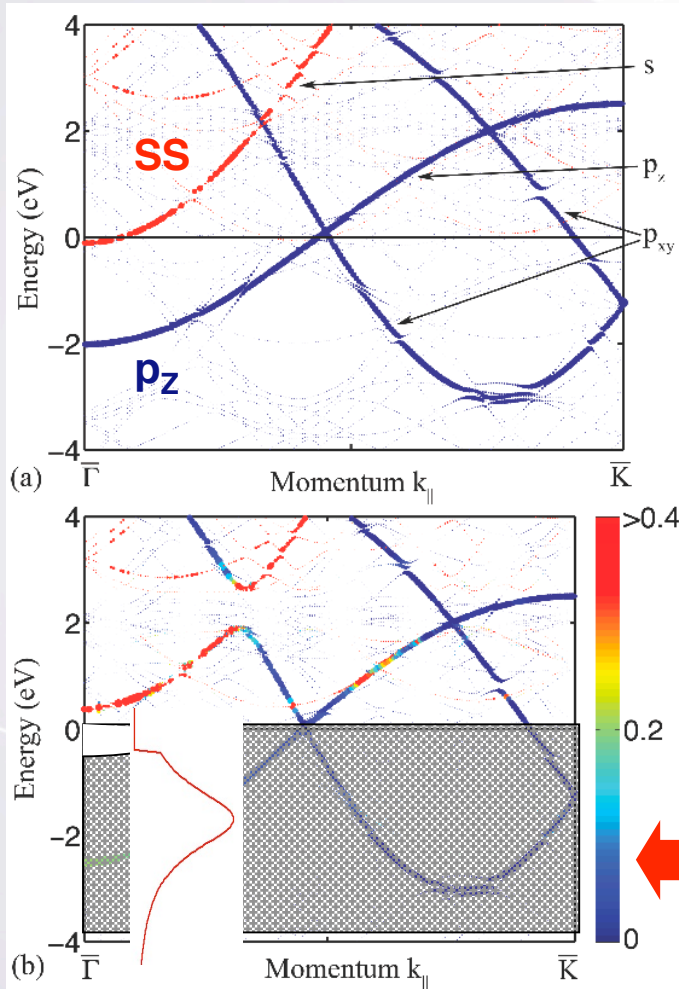
TB bands for the modulated structure
Gloor (2006)

- 1) Pb p_z states hybridize with the Ag surface band and, through this, with bulk states.
- 2) The mixed state is degenerate with the **continuum of bulk states**. For each k value, it is an **impurity problem**.

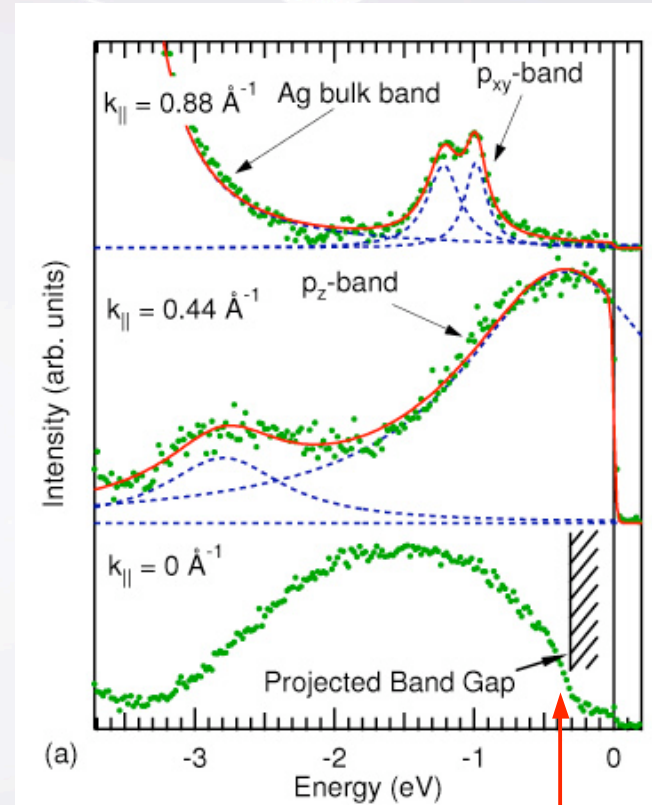


$\Delta E \sim$ hybridization

Selective hybridization with bulk via the Ag SS



projected bulk states



lorentzian limited by bulk gap

Summary



- *Enhanced correlations and lower symmetry lead to different ground states at surfaces. Ideal playground for ARPES practitioners*
- **Interface engineering to tune spin splitting and spin configuration**