

Surface state in Ag/Cu(111) and Ag/Au(111).  
Evolution of spin-orbit interaction, surface state energy  
and quasi-particle lifetime

**D. Malterre,**

*Laboratoire de Physique des Matériaux, Université H. Poincaré,  
F-54506, Vandoeuvre-les-Nancy (France)*

## Plan of the talk

I) some generalities about surface state

(energy, effective mass, QP lifetime)

II) how to use surface states of study atomic and electronic structure of surfaces and interfaces.

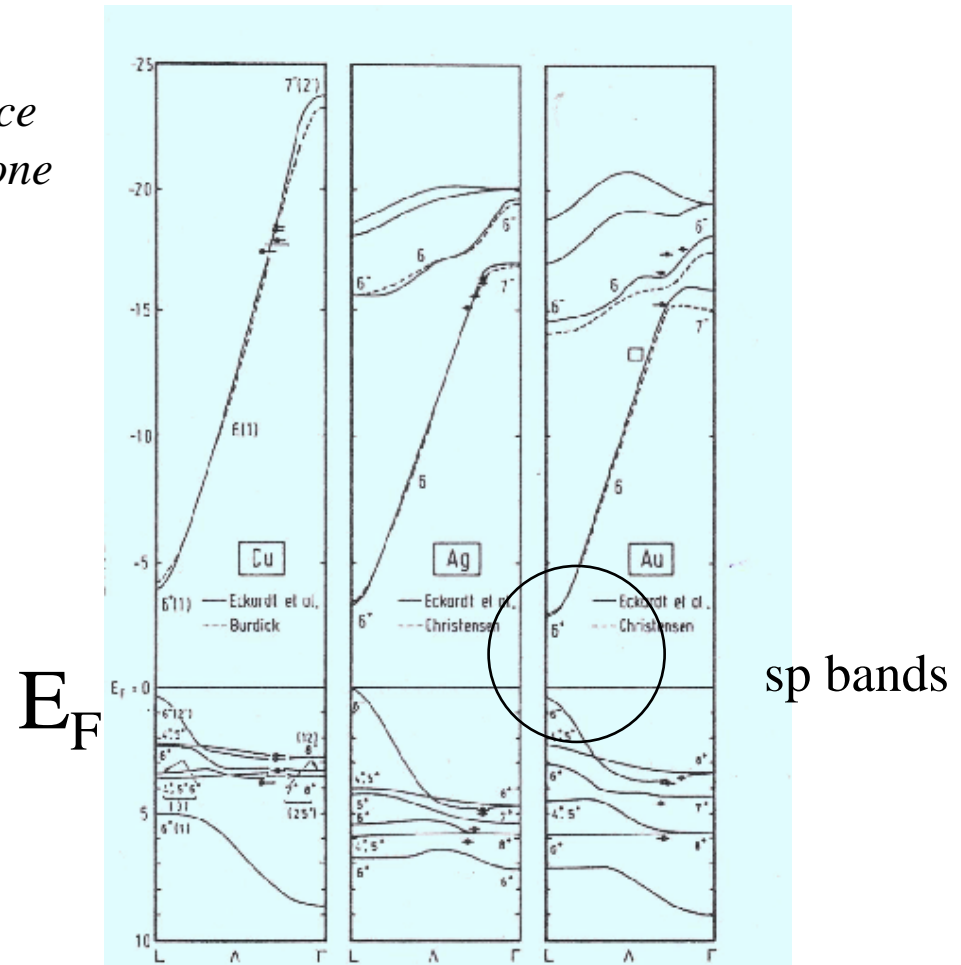
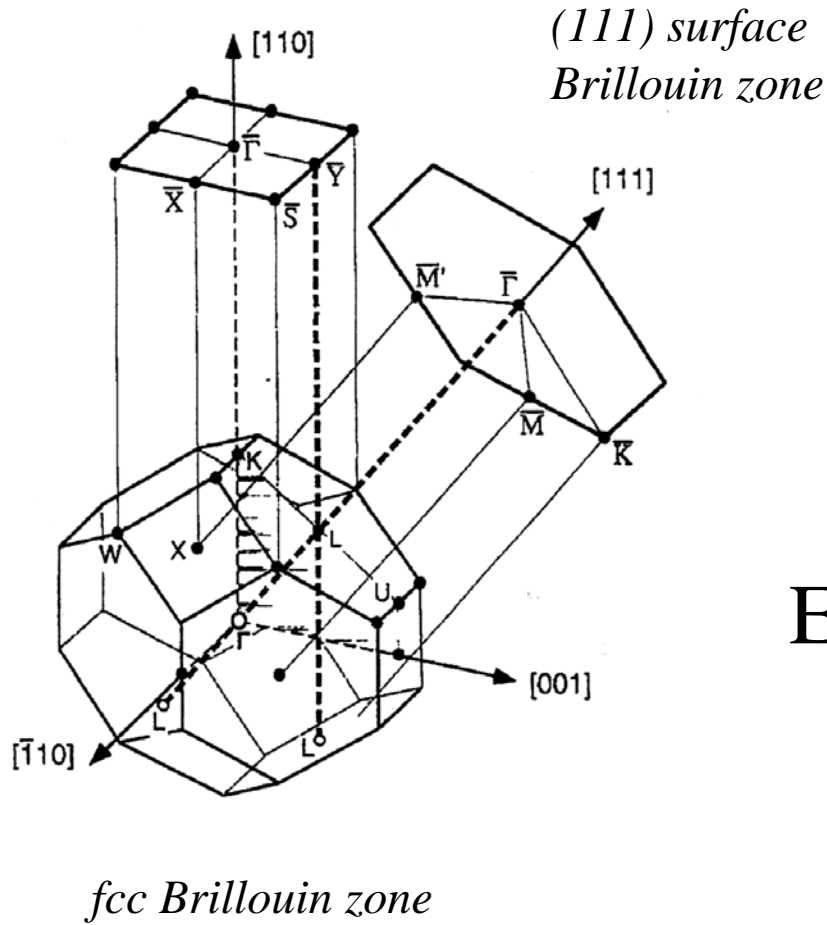
Ag/Cu(111) and Ag/Au(111) = model systems to study the progressive evolution of the electronic parameters

- the surface state energy at  $k_{\parallel}=0$ ,
- quasi-particle lifetime
- spin-orbit interaction

III) Conclusion and perspective

# I-Shockley surface state of noble metals

# Band structure of noble metals : Cu(111), Ag(111) et Au(111)



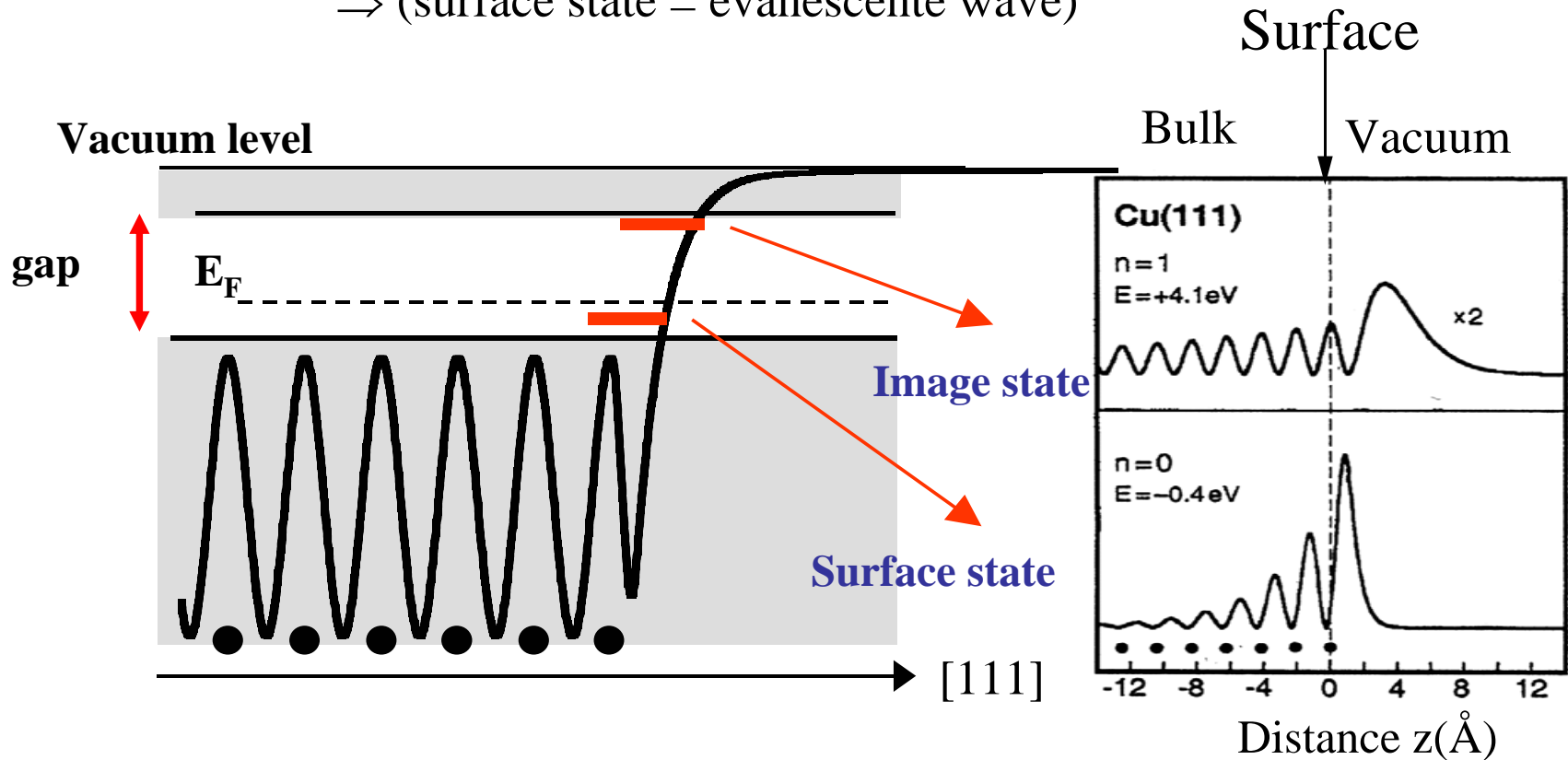
Gap at L point



- Surface = Breakdown of the translational symmetry

⇒ Peculiar solution in the gap with a complex wave vector  $k_{[111]}$

⇒ (surface state = evanescent wave)



→ localisation of the electronic density at the surface

Surf. State is very sensitive to any structural modification

Surface states = 2 D Bloch states

$$E(k_x, k_y) = E_0 + \frac{\hbar^2 (k_x + k_y)^2}{2m^*}$$

Parabolic dispersion  
(nearly free electron like)

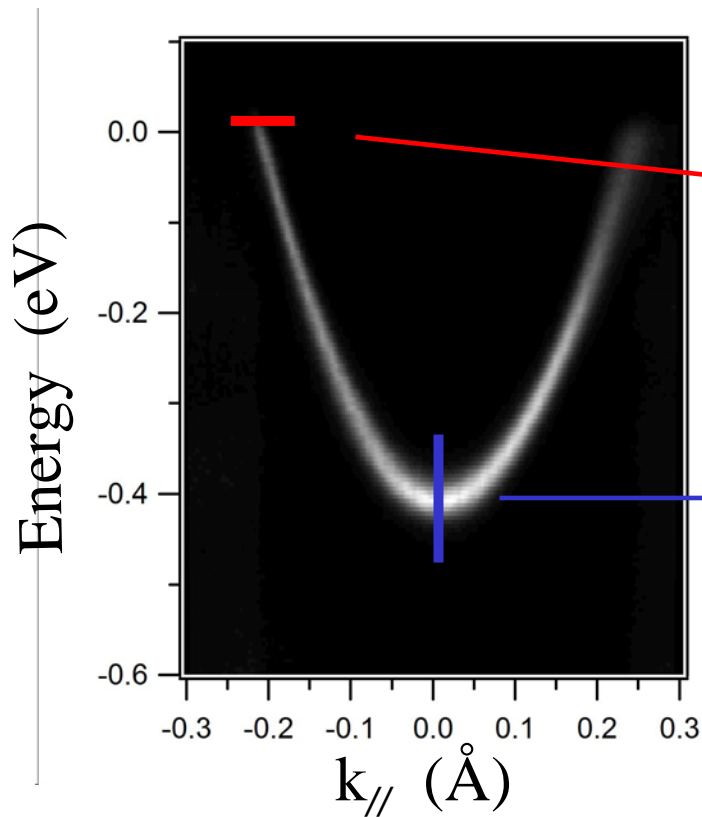
Electronic parameters ( $E_0$ ,  $m^*$ , QP lifetime) can be studied by Spectroscopic techniques :

-**ARPES** ( $E < E_F$ )

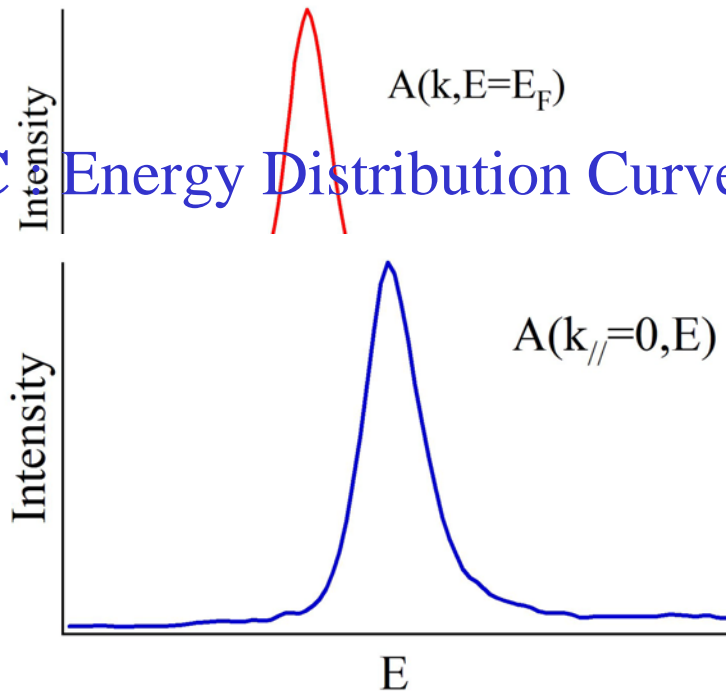
-**STS** ( $\forall E$  but with a  $k$  integration)

# ARPES Cu(111)

**MDC : Momentum Distribution Curve**



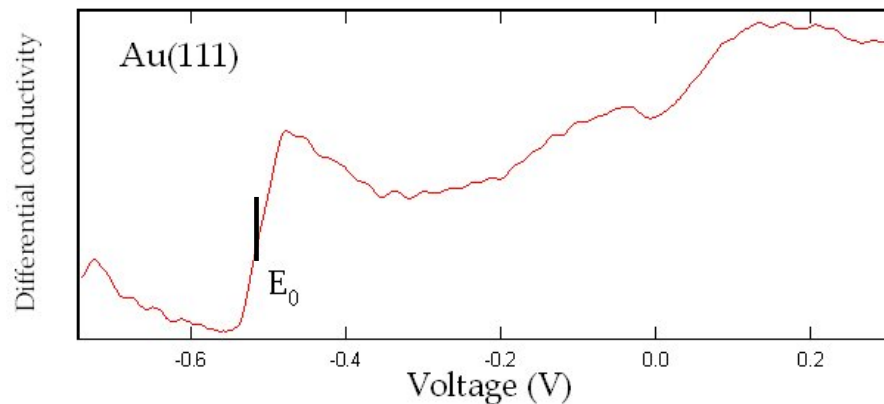
**EDC Energy Distribution Curve**



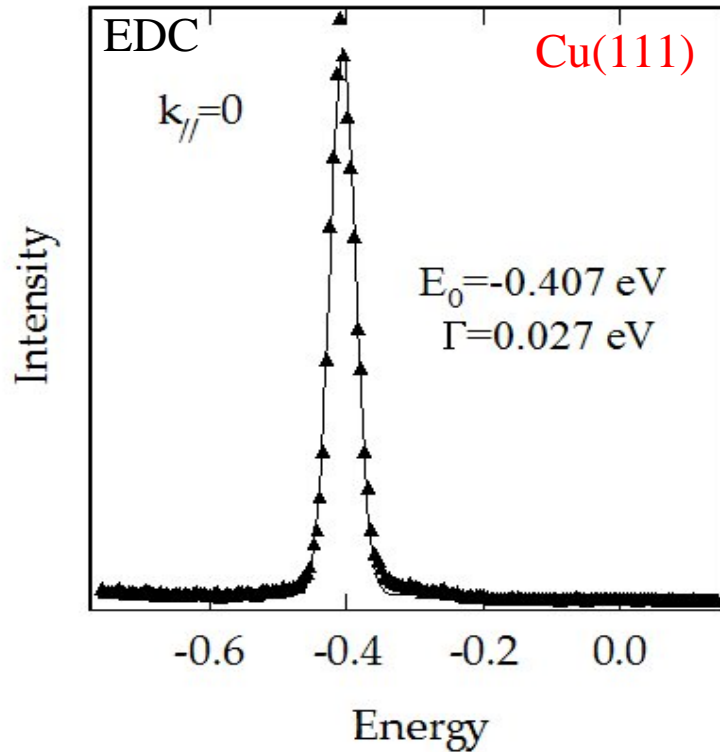
**STS**

$$\frac{dI}{dV} \propto N_{loc}(E)$$

Differential conductivity



# ARPES (T=80 K)



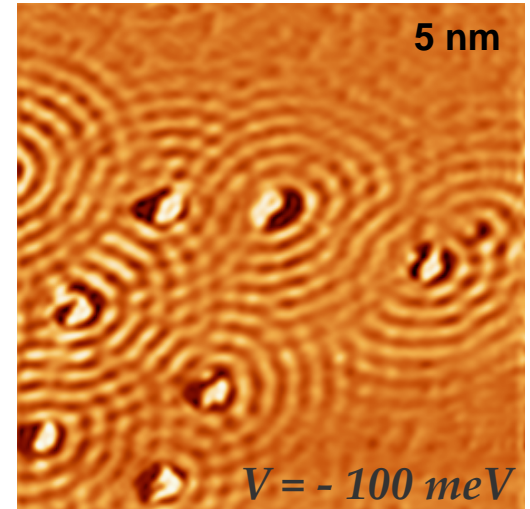
$-\Gamma$  corresponds to **the inverse lifetime** of the electronic excitations (quasi-particles)

3 different contributions can be distinguished in  $\Gamma$  :

- **electron-electron** interactions
- **electron-phonon** interactions
- **defects** (structural defects, disorder, impurities, etc.)

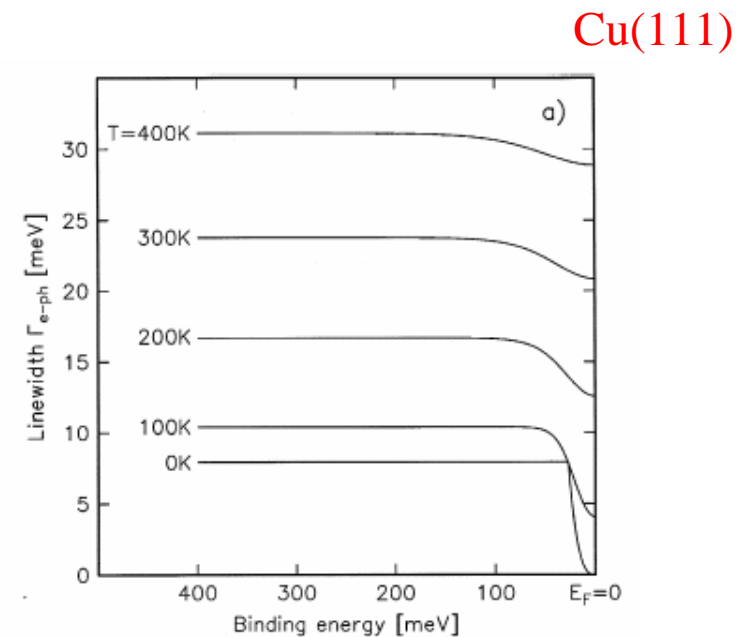
## defects

- scattering of the SS by impurities
- contribution to lifetime : constant but depends to the interface quality



## Electron-phonon interactions

- $\Gamma_{e-ph}$  depends on temperature and quasi-particle energy
- nearly constant for the energy for  $k_{//}=0$
- $\Gamma_{e-ph} \approx 5-10 \text{ meV}$  at low temperature



Matzdorf (1999)

## Electron-electron interactions

-creation of electron-hole pairs

Fermi liquid model :

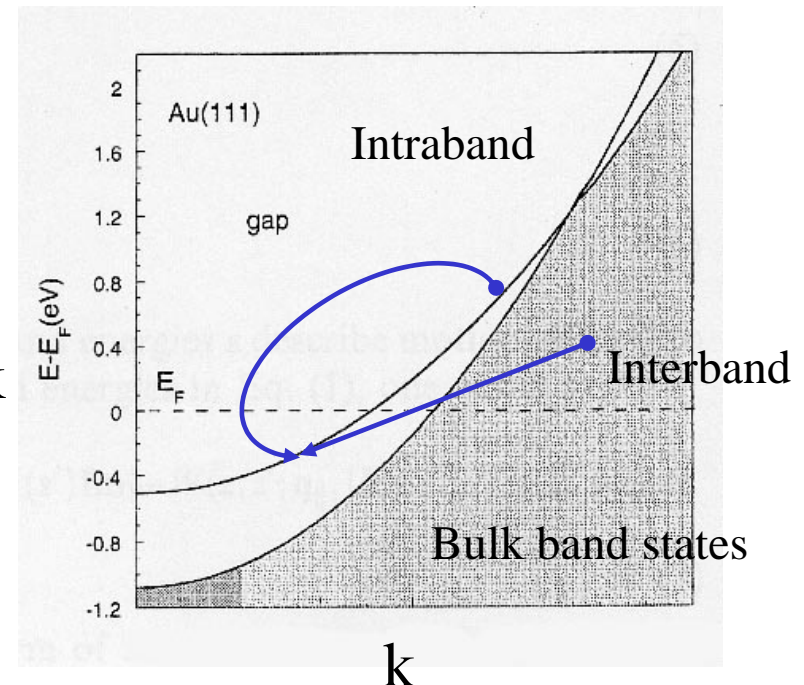
$$\Gamma_{e-e} \approx (E - E_F)^2$$

-identification of two mechanisms for S.S.

interband (bulk) and intraband decay mechanism

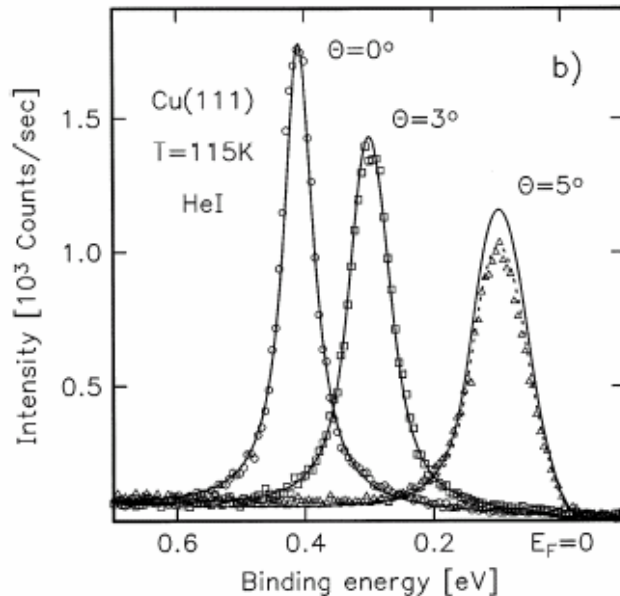
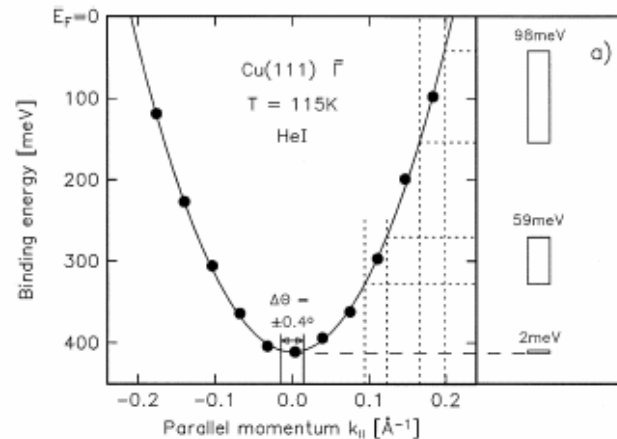
near  $\bar{\Gamma}$ , the linewidth is dominated by intraband decay mechanism

(2D decay screened by the 3D bulk states [Chulkov talk](#))



# It is not possible to study $\Gamma_{e-e}(E)$ with ARPES

R. Matzdorf / Chemical Physics 251 (2000) 151-166



Small  $m^*$  (0.4)



Large curvature of  $E(k)$

Broadening of the ARPES spectra with decreasing energy due to finite angle resolution

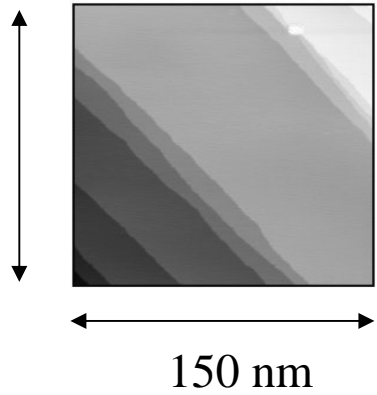
$\Gamma_{e-e}$  can only be measured at  $k_{||}=0$

# II-Ag/Cu(111) interface

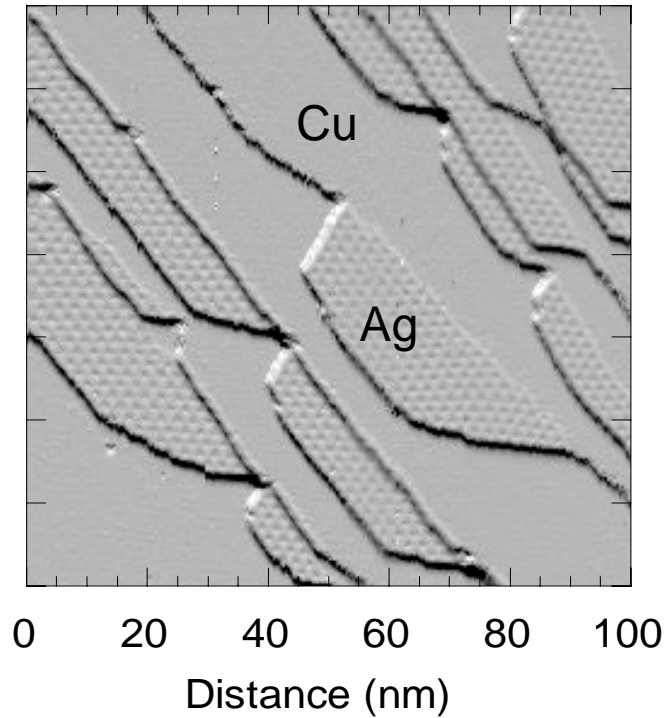
- growth and atomic structure
- surface state energy
- lifetime



Cu(111)

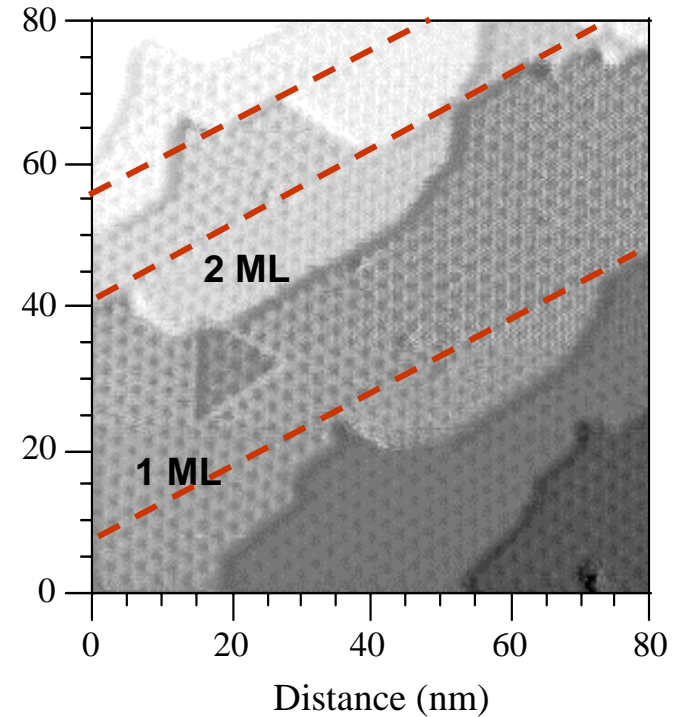


0.4 ML Ag/Cu(111)



Morphology ; Ag islands connected to step edges

1.2 ML Ag/Cu(111)

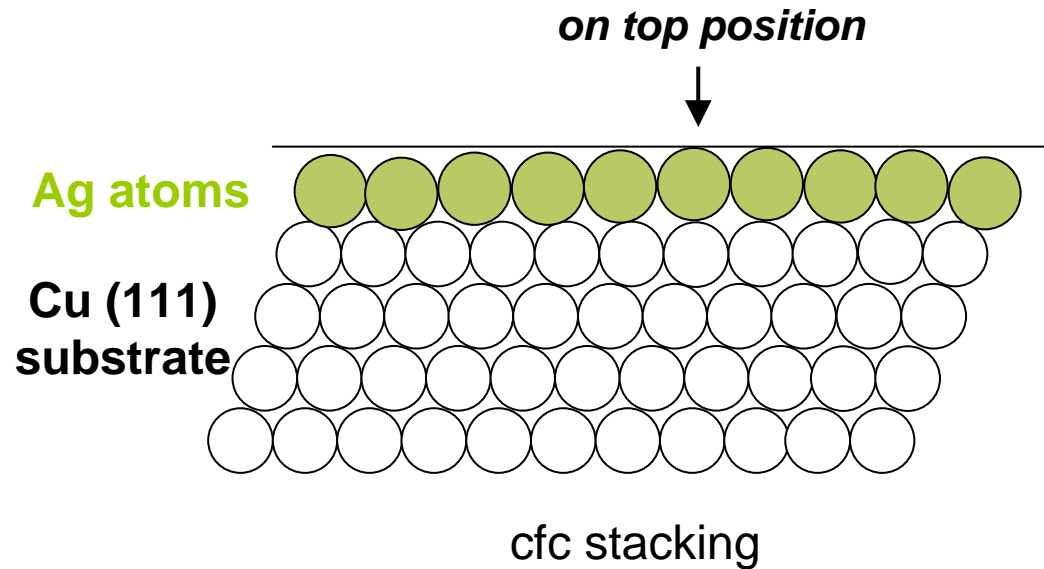
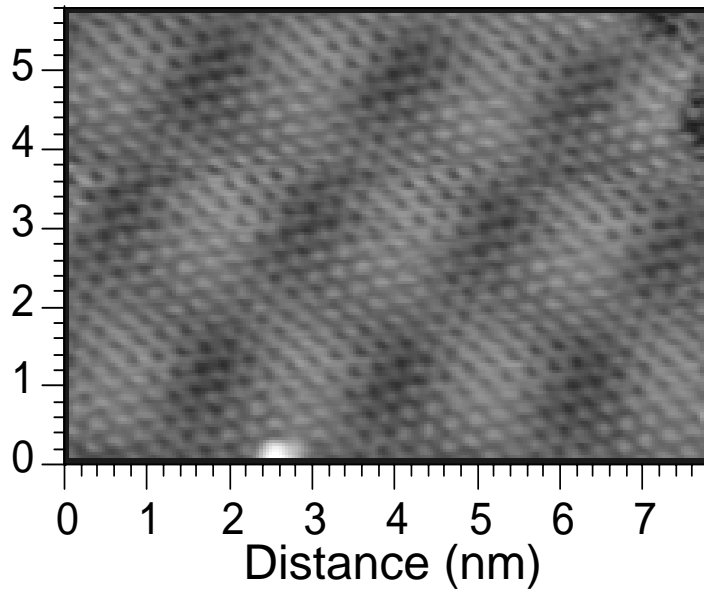


Layer by layer growth up to 3 ML

Rugosity of the films for higher thickness (3D growth)

# 1 MC d'Ag/Cu(111) - Films prepared at 150 K

## « moiré » structure



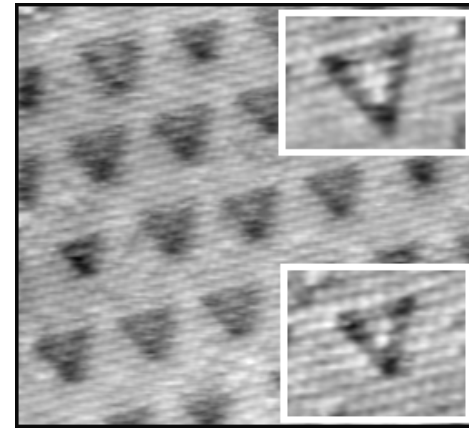
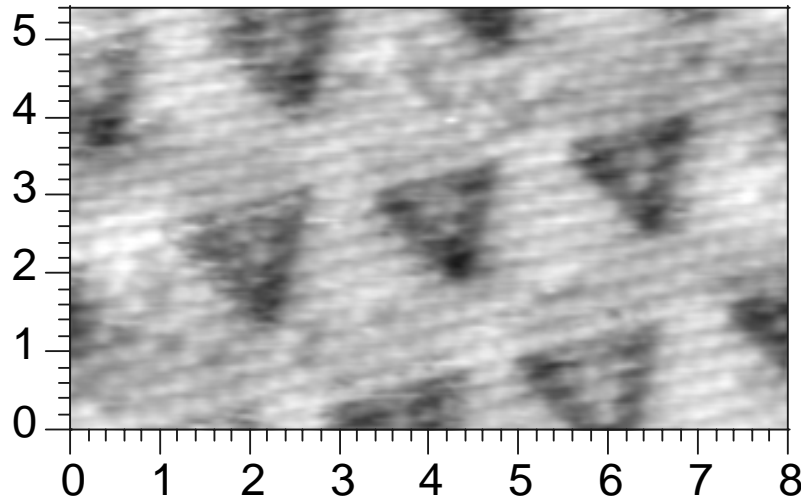
Superposition of Ag and Cu lattices

**9×9 reconstruction**  
(8 Ag atoms on 9 Cu atoms)

⇒ metastable phase

# 1 MC d'Ag/Cu(111) - Films prepared at 300 K

## «triangular » structure



Distance (nm)

9×9  
reconstruction

Ag atoms

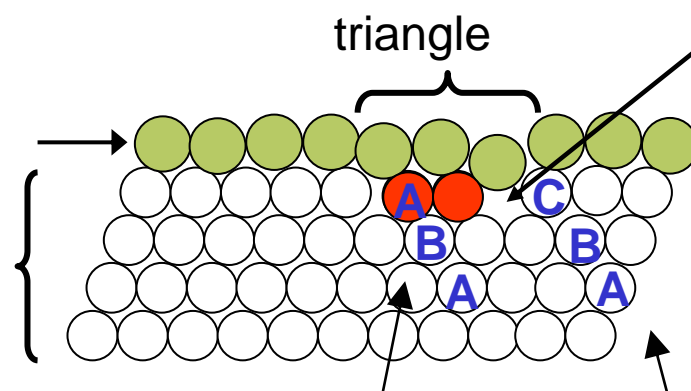
Cu(111)  
substrate

triangle

hcp stacking

fcc stacking

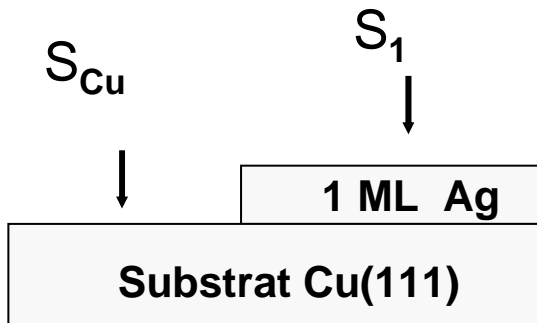
Some Cu  
atoms are  
ejected  
+ relaxation



# ARPES on the interface prepared at 300 K in the submonolayer range

Morphology of the surface :

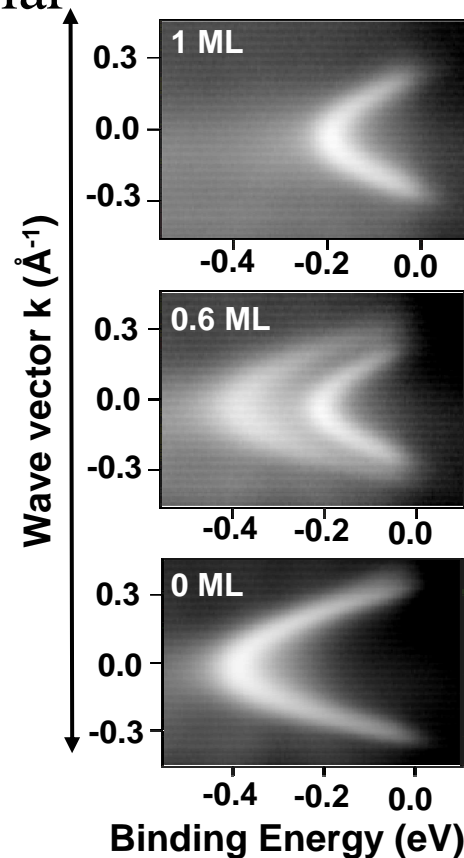
- Cu terraces
- Ag islands with the triangular structure



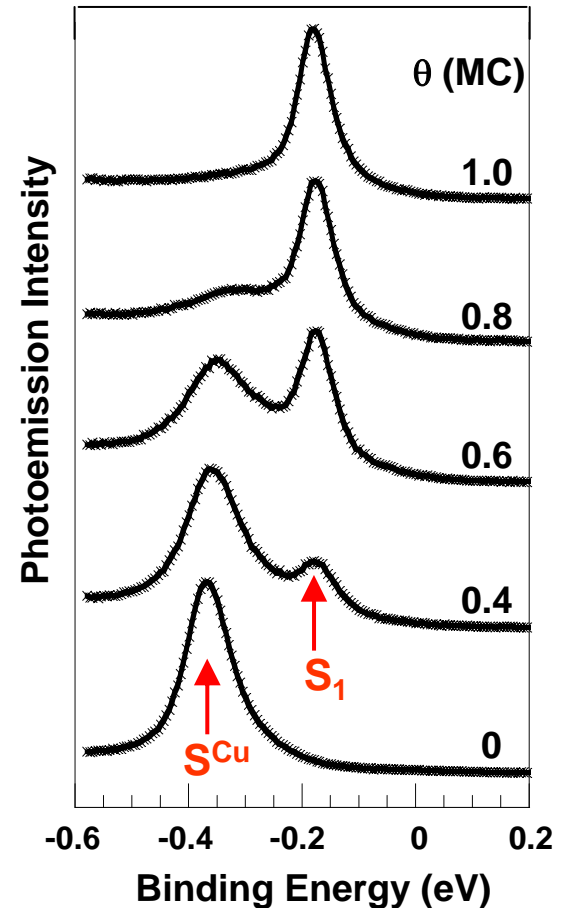
2 dispersive states  
(balance of intensity)

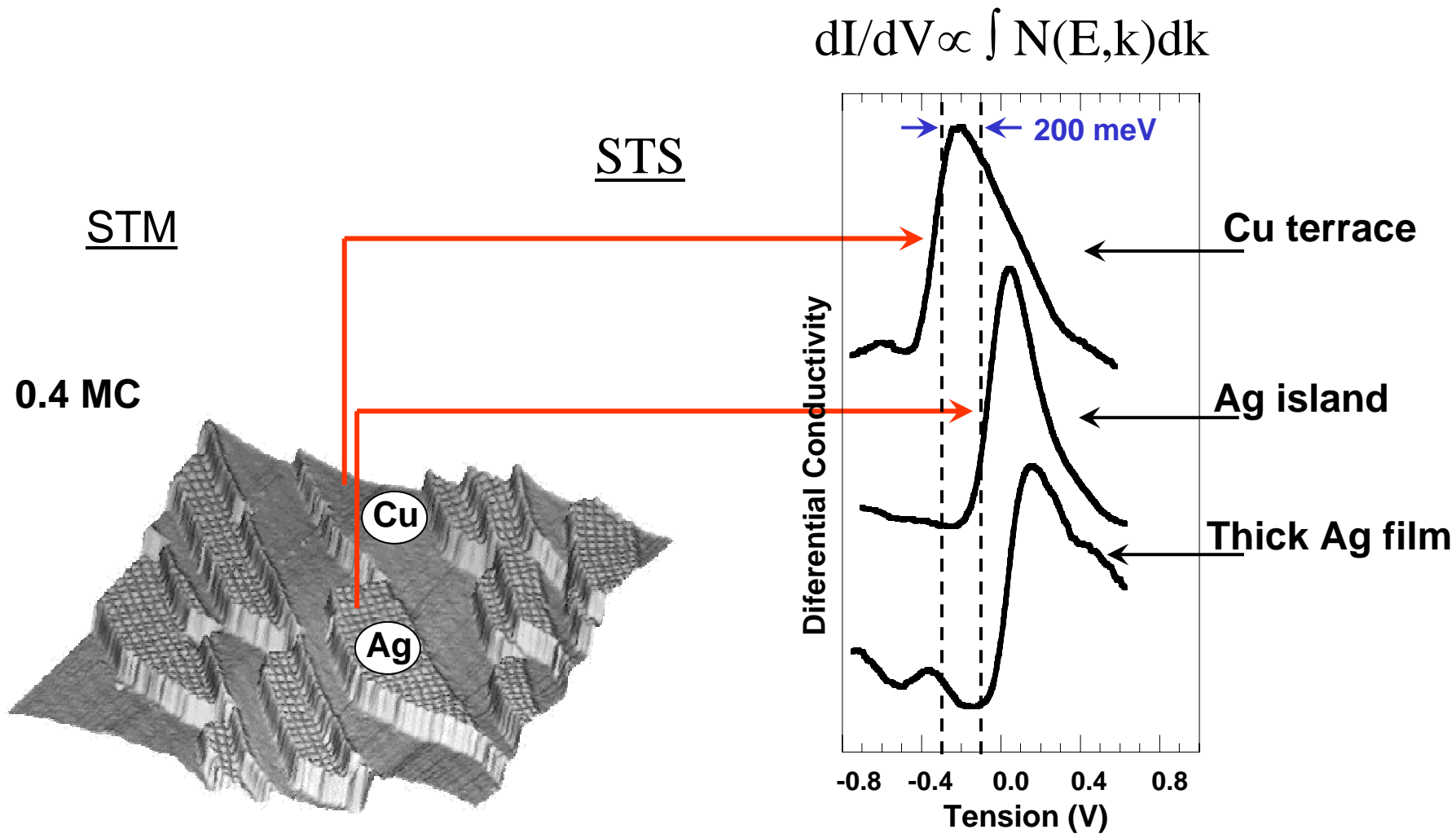
$$E_0^{Cu} - E_0^{S_1} = 200 \text{ meV}$$

$0 < \theta < 1 \text{ ML}$



EDC ( $k_{\parallel}=0$ )



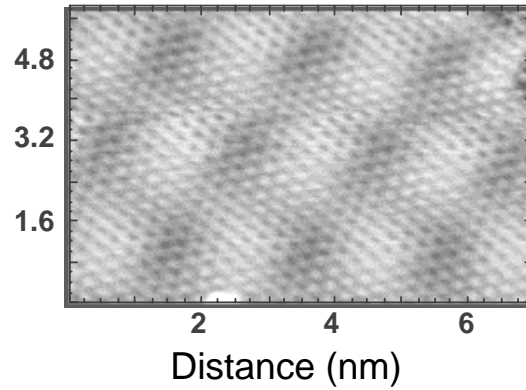


Surface states are **confined** :

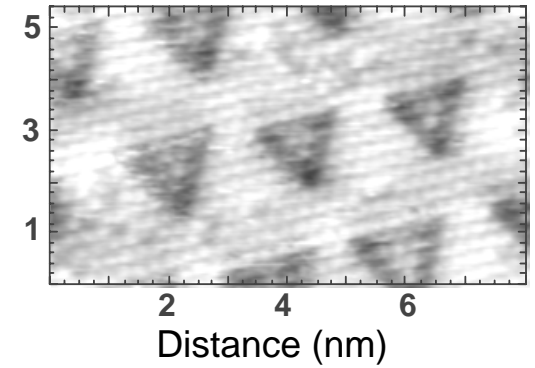
- in the Cu terraces
- in the Ag islands

# 0.4 ML Ag/Cu(111)

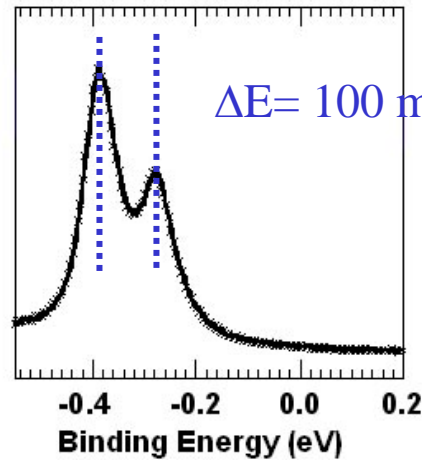
Prepared at 150 K



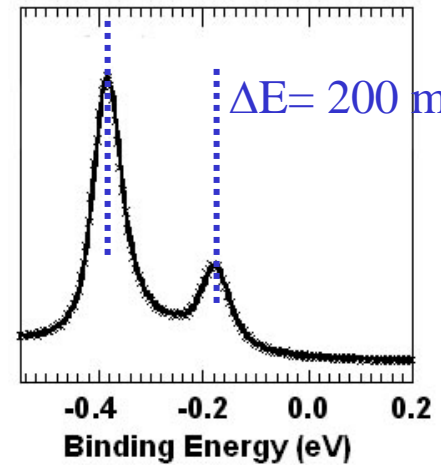
Prepared at 300 K



The S.S. energy reflects the atomic structure of the Ag film



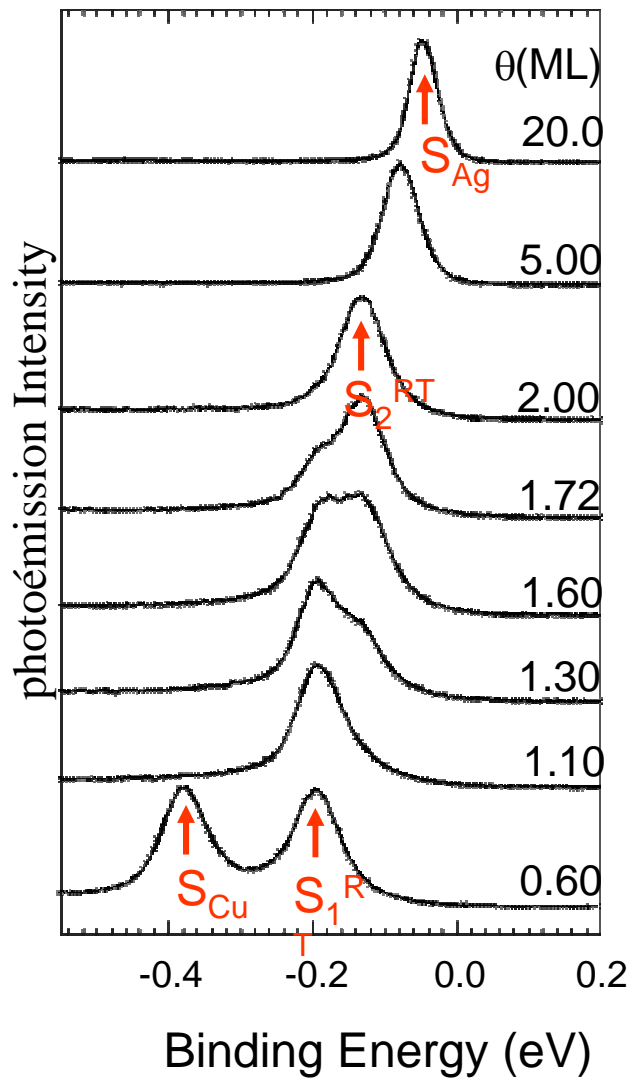
$k_{//} = 0$



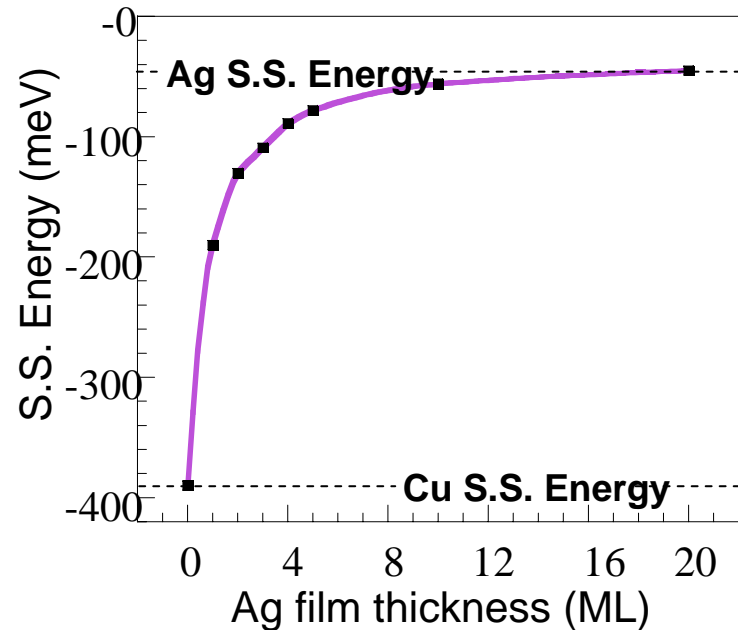
Moiré Structure

irreversible  
transition

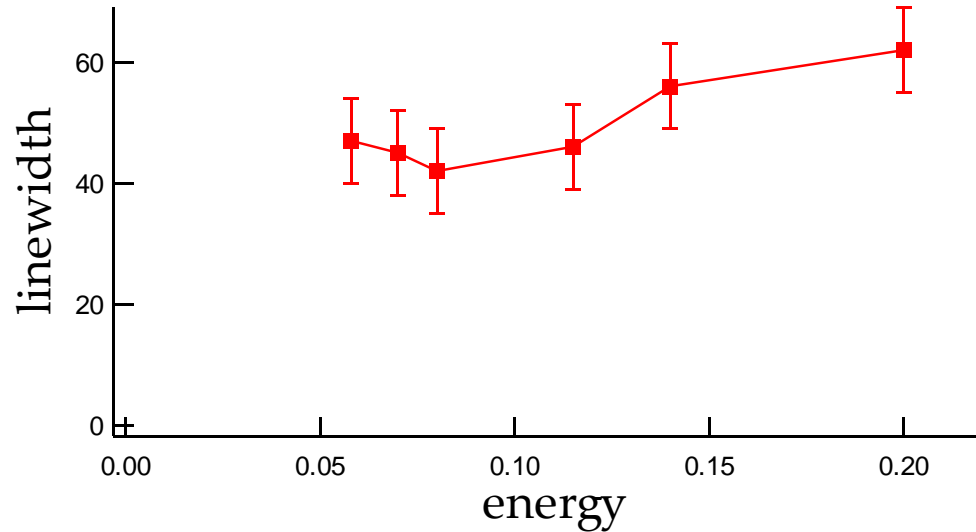
Triangular Structure



- The S.S. energy is a signature of the Ag film thickness
- Continuous evolution from the Cu S.S. energy to the Ag S.S. energy



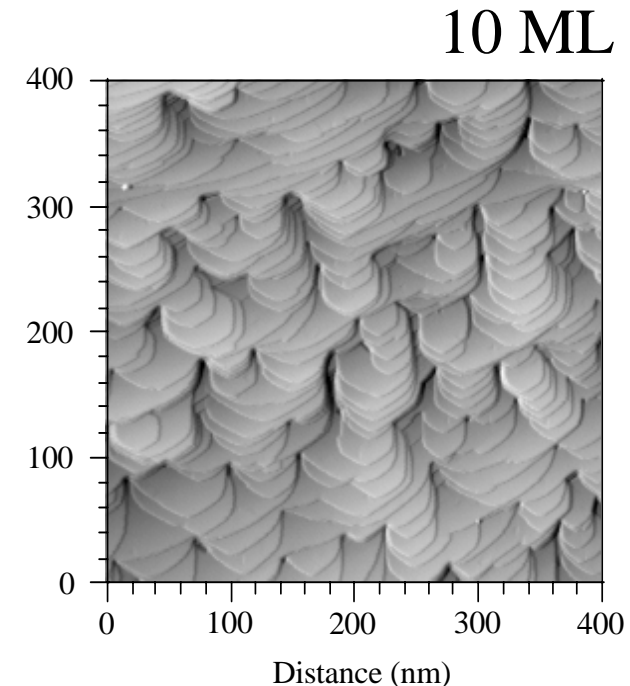
# Lifetime evolution at $\bar{\Gamma}$ as a function of energy



Linewidth remains large with decreasing energy  $\Gamma$  larger than for Cu(111)

-morphology of the 10 ML film shows a 3D growth  $\Gamma_{\text{def}}$  is the dominant contribution!

$$\Gamma_{\text{def}} > \Gamma_{\text{e-e}}$$

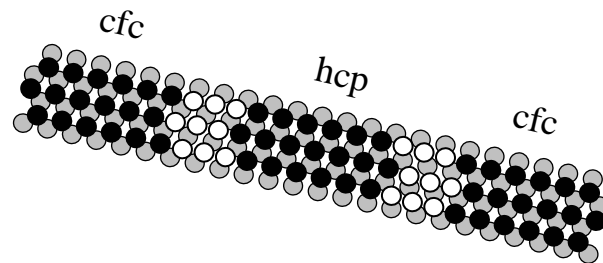
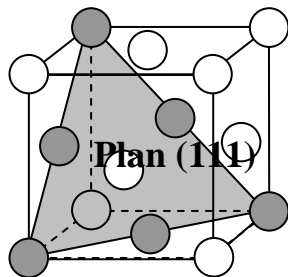




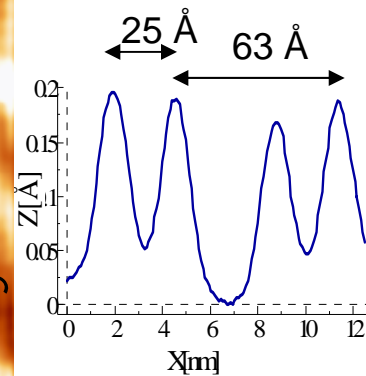
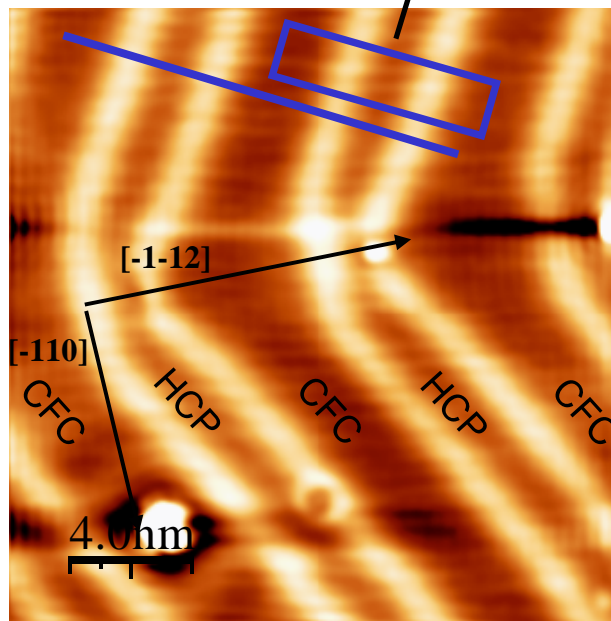
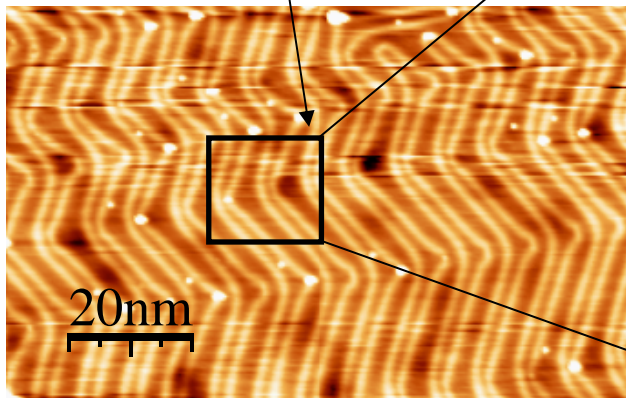
## III-Ag/Au(111) interface

- growth and atomic structure
- surface state energy
- spin-orbital
- lifetime

# Au (111)



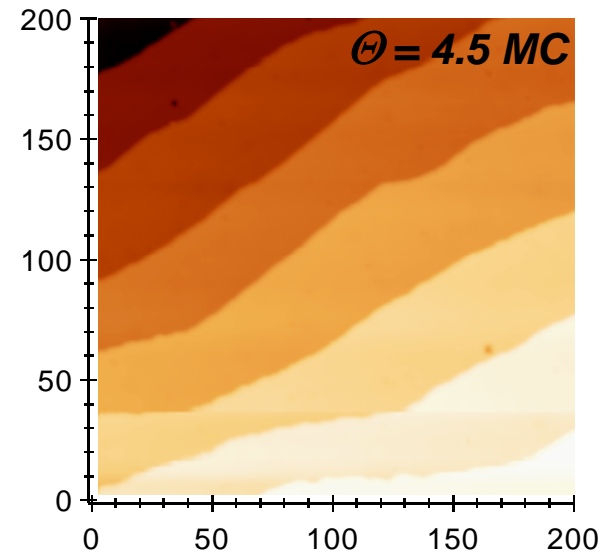
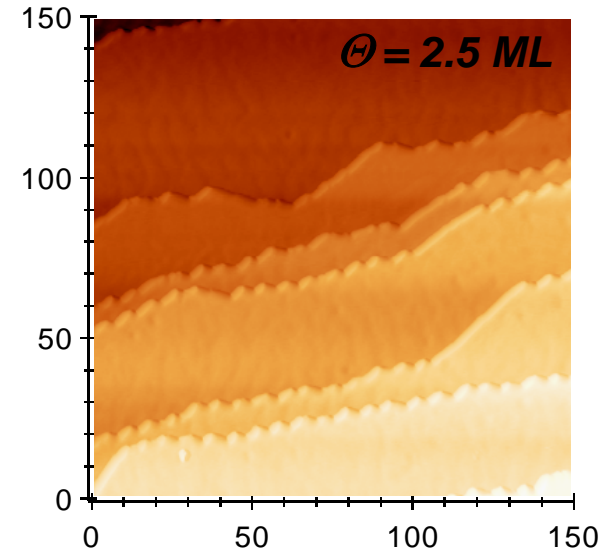
Herringbone surface  
Reconstruction



# The Ag/Au(111) interface

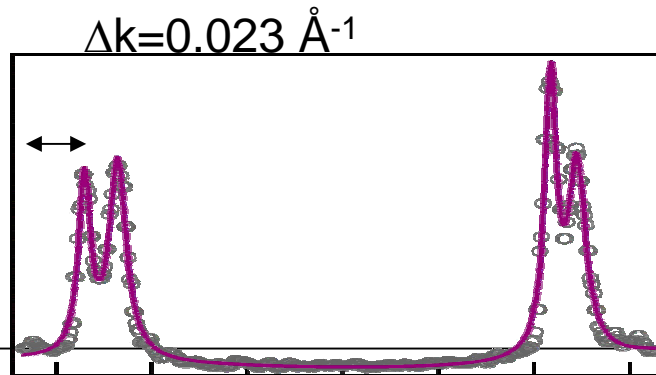
## Main characteristics

- perfect epitaxy  
lattice parameters of Ag and Au are quite identical
- layer by layer growth  
(step edge flow)
- formation of surface alloys  
by annealing



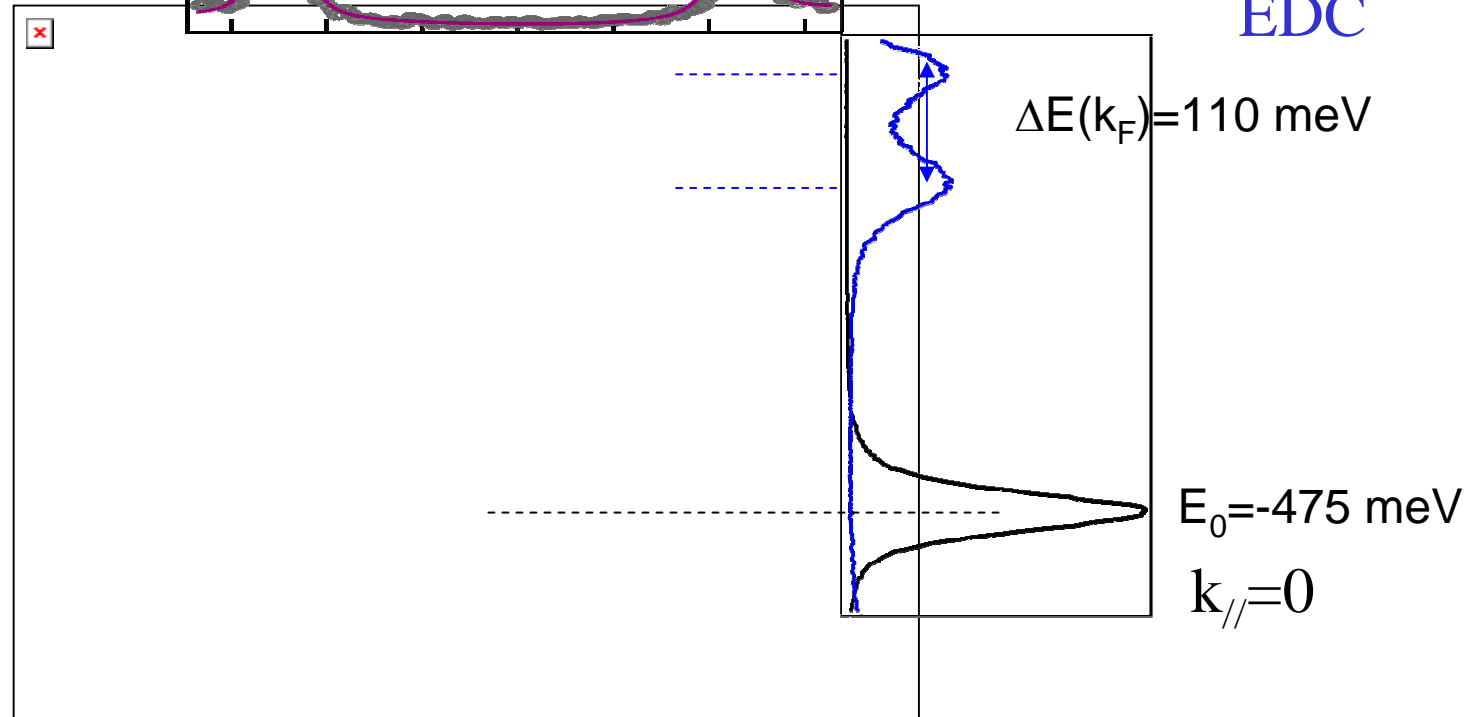
# Two dispersive Shockley surface states in Au(111)

MDC



$-E_0 = -475 \text{ meV @ } 90 \text{ K}$   
 $-m^* = 0.26 m_e$   
 $-\Delta k = 0.023 \text{ \AA}^{-1}$

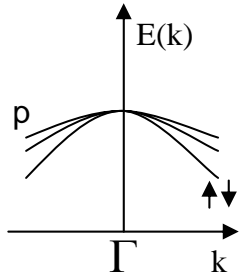
EDC



signature of  
the spin-orbit  
effect on the  
surface state  
*Lashell et al.,  
PRL (1996)*

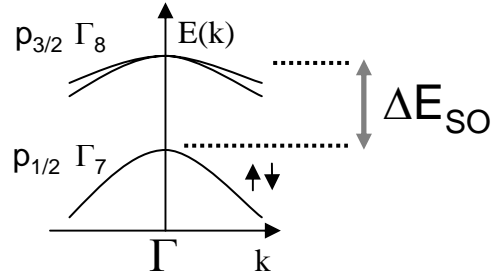
# Spin orbit effect on the p states at the top of the valence band of semiconductors

without spin-orbit



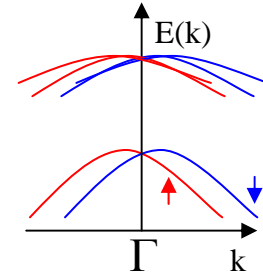
(Si)

with spin-orbit interaction  
centrosymmetric crystal



(Ge)

with spin-orbit interaction  
non-centrosymmetric crystal



(GaAs)

Time reversal symm.  $E(\vec{k}, \uparrow) = E(-\vec{k}, \downarrow)$   $E(\vec{k}, \uparrow) = E(-\vec{k}, \downarrow)$

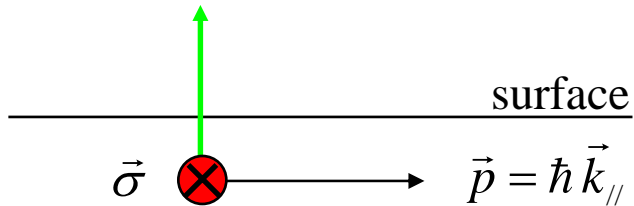
Inversion symm.  $E(\vec{k}, \uparrow) = E(-\vec{k}, \uparrow)$

$E(\vec{k}, \uparrow) = E(\vec{k}, \downarrow)$   $E(\vec{k}, \uparrow) \neq E(\vec{k}, \downarrow)$

# Au(111) : The inversion symmetry is broken at the surface

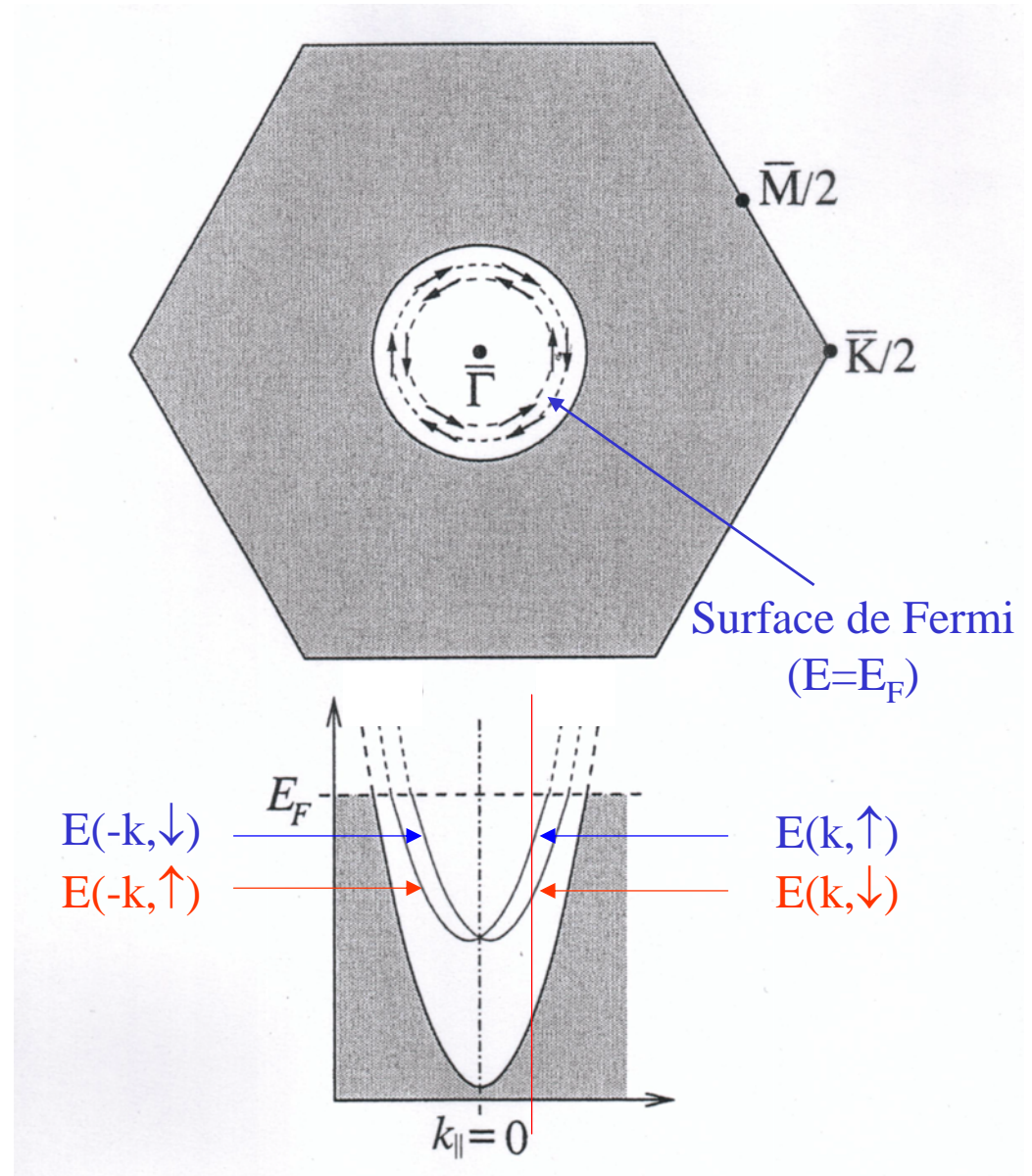
$$H_{so} = \frac{\hbar}{4m^2c^2} (\vec{\nabla}V \wedge \vec{p}) \cdot \vec{\sigma}$$

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



In a nearly free electron picture,  
 $\vec{\nabla}V$  is perpendicular to the surface

**→** two spin polarized SS bands



Qualitative explanation :  $\vec{\nabla}V$  is not large enough to explain the experimental splitting

$$H_{SO} = \alpha_R \mathbf{h} \cdot (\mathbf{e}_z \wedge \mathbf{k}_{//}) \cdot \boldsymbol{\sigma}$$

Rashba parameter

$$\alpha_R \propto \frac{dV_{surf}}{dz} \approx \frac{\Phi_S}{\lambda_F}$$

$$\Rightarrow \Delta E = 2\alpha_R k_F \approx 10^{-6} eV$$

$$\Delta E_{exp} = 0.1 eV !!$$

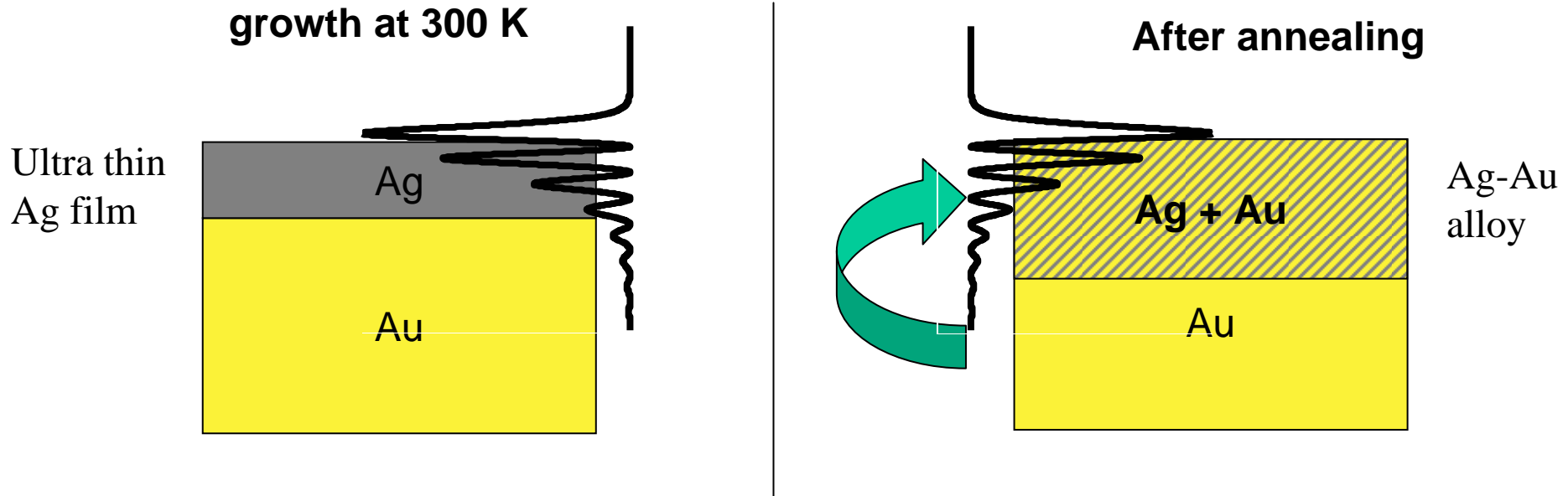
A simple tight-binding model shows that the Rashba coefficient is also proportional to the atomic spin orbit constant ( $\lambda_{at}$ )

$$\alpha_R \propto \lambda_{at} \frac{dV}{dz}$$

L. Petersen et al.,  
Surf. Sci. (2000)

Ag/Au(111) is an interesting **model system** to test this assumption

2 kinds of interface

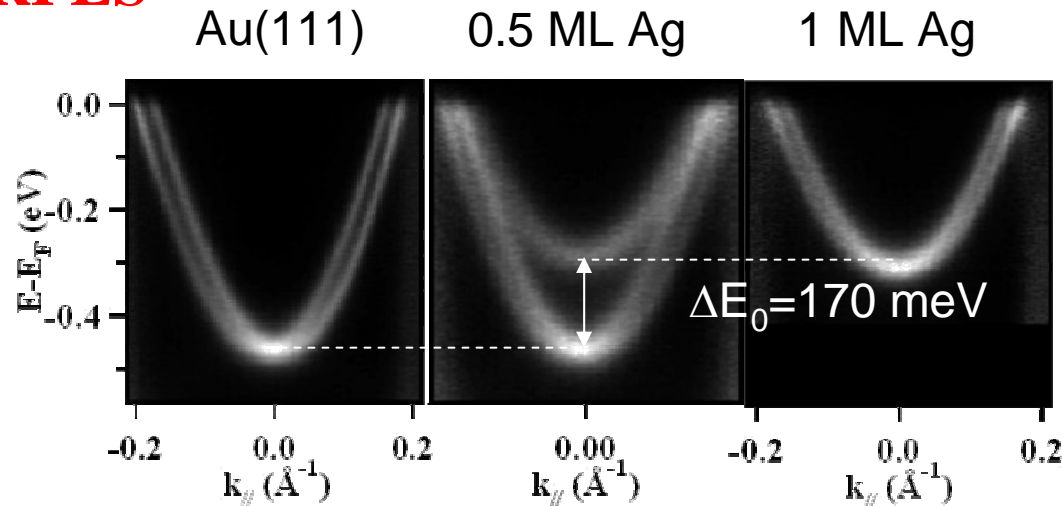


SO interaction in Au  $\gg$  SO interaction in Ag

The number of Au atoms probed by the surface state depends on Ag film thickness or alloy composition and should be reflected by the Spin-orbital splitting of the S.S.

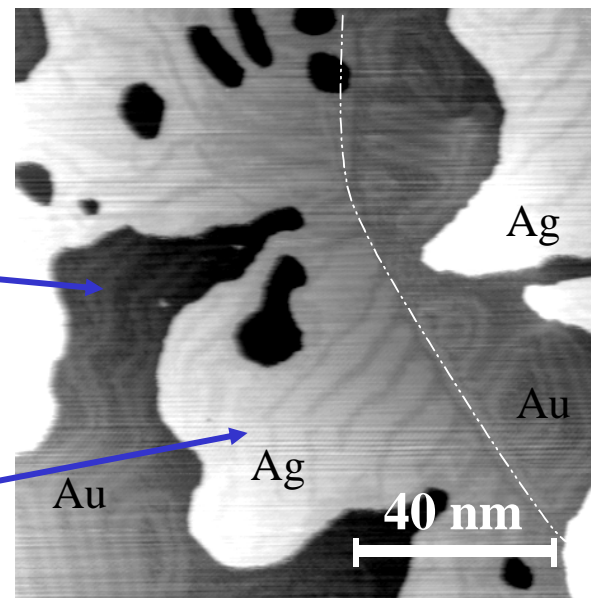


# ARPES

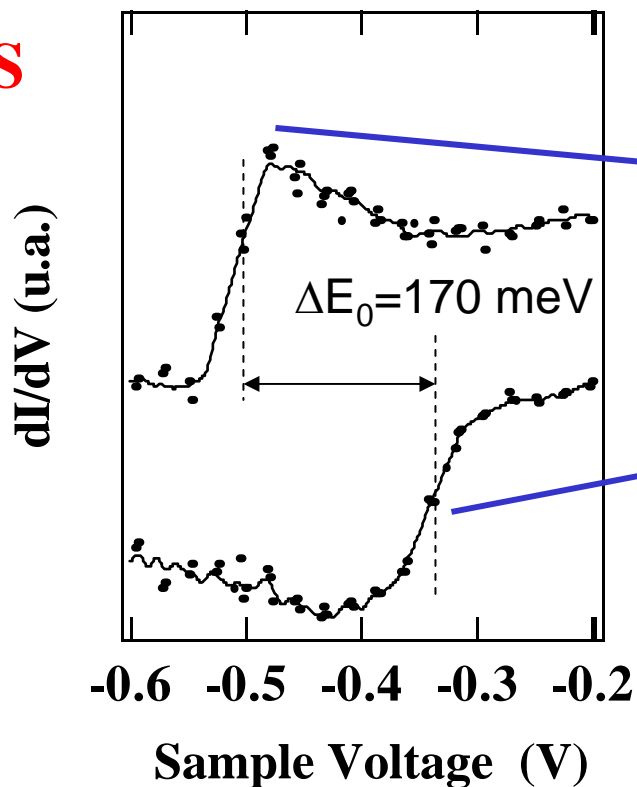


Evidence of two dispersive states

# STM

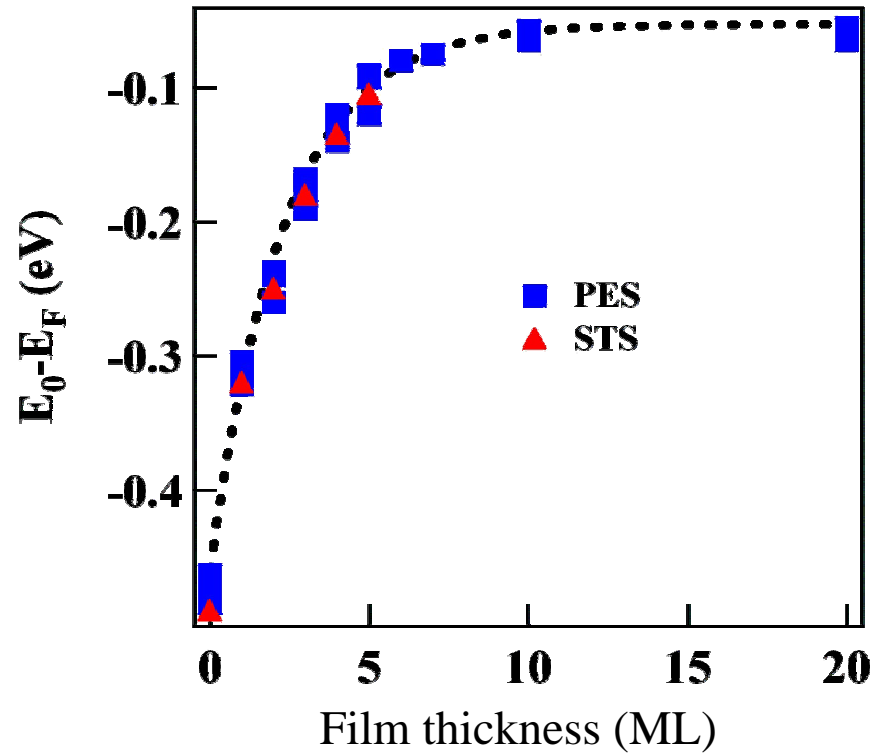
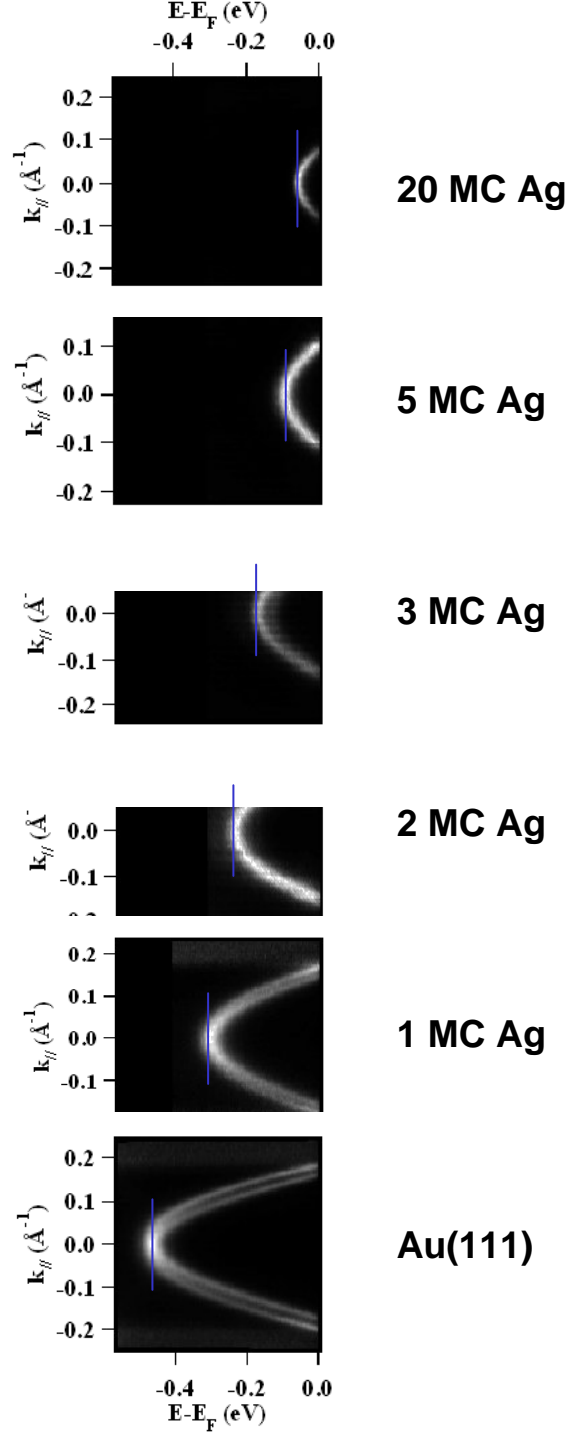


# STS

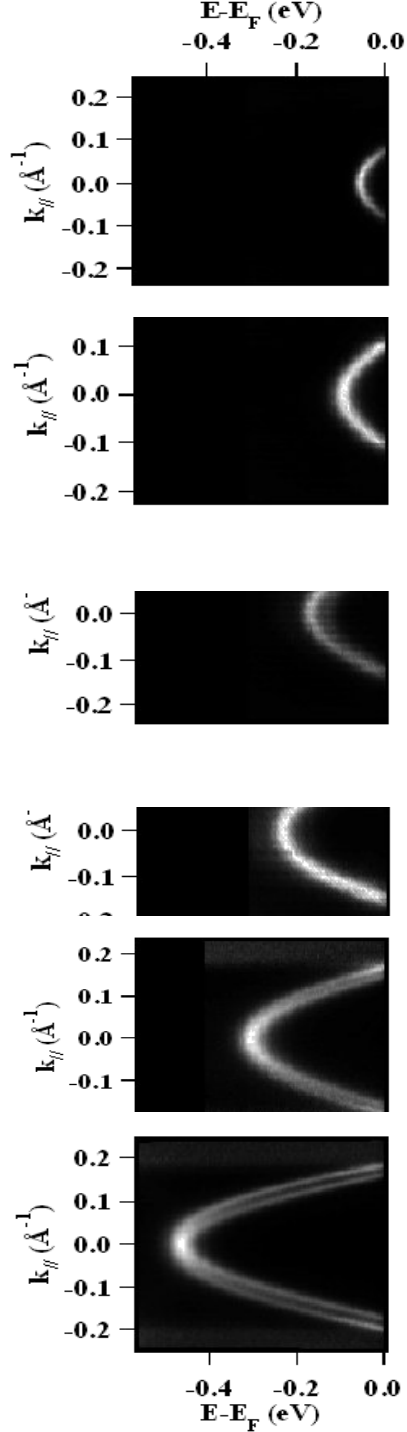


Confinement of the SS in Au terraces and Ag islands

## Evolution of the SS energy at $\bar{\Gamma}$



Very good agreement between PES and STS  
 Evolution from the Au S.S. to the Ag S.S. energy



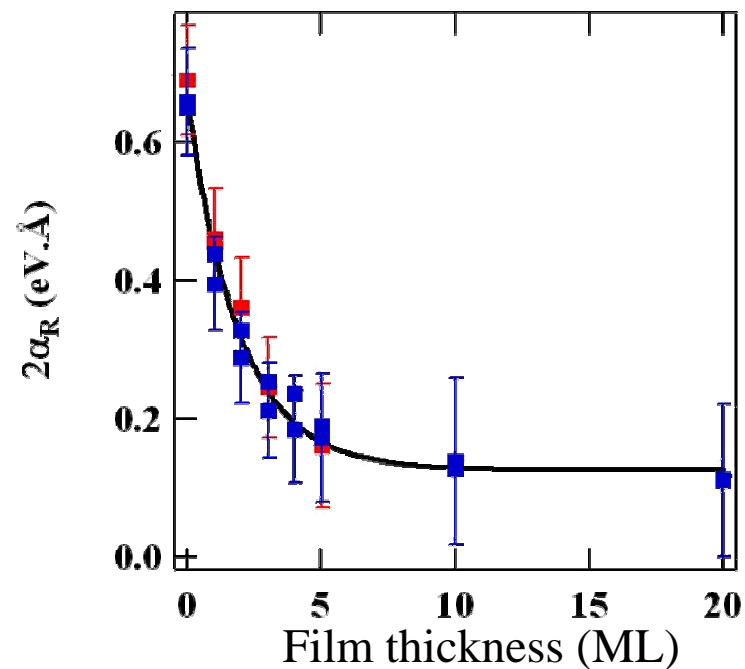
estimation of  
Rashba coeff.  
from MDC at  $E_F$

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

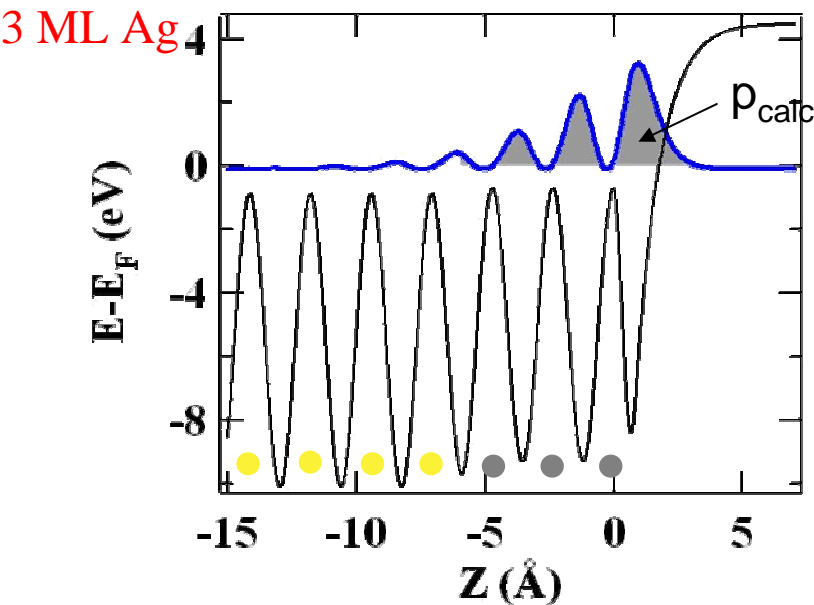
Or from EDC at  $k_F$

$$\alpha_R = \frac{\Delta E(k_F)}{2k_F}$$

$\alpha_R$  strongly  
decreases with  
increasing  
Ag thickness

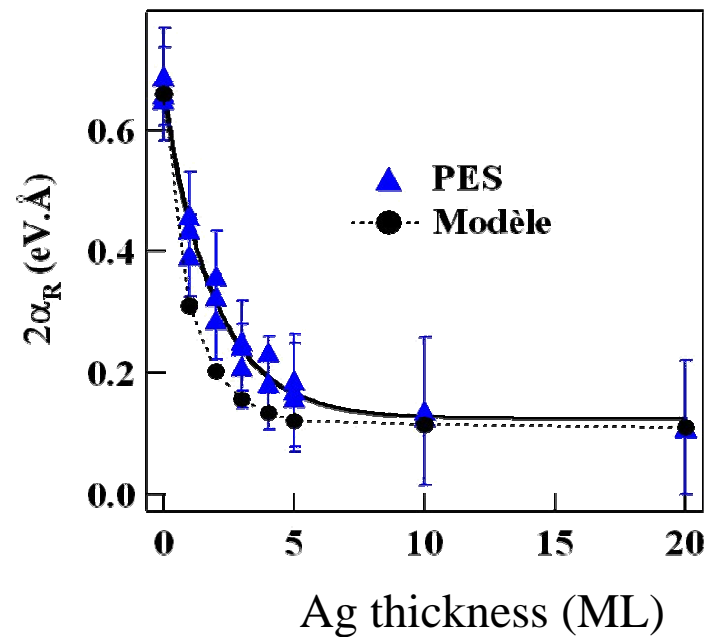
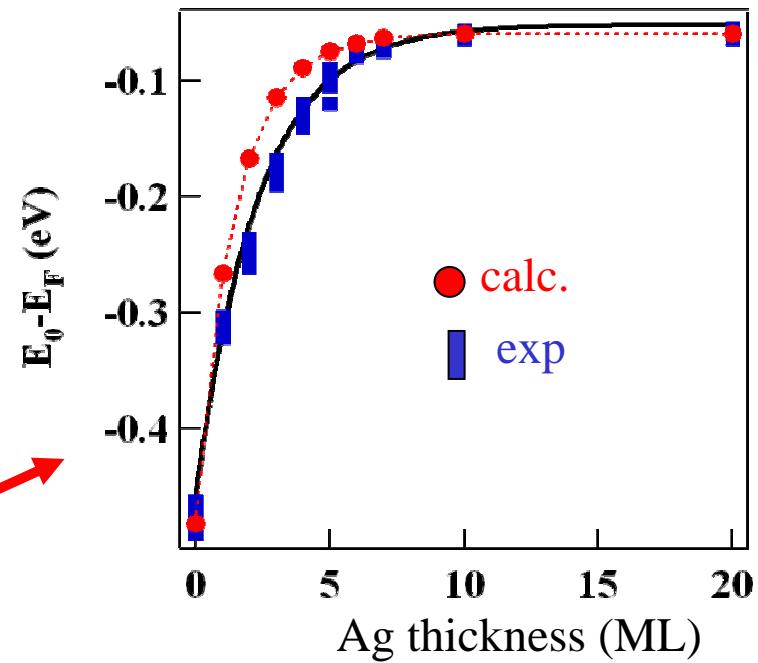


# Simulation of the data in a 1D pseudo potential approach

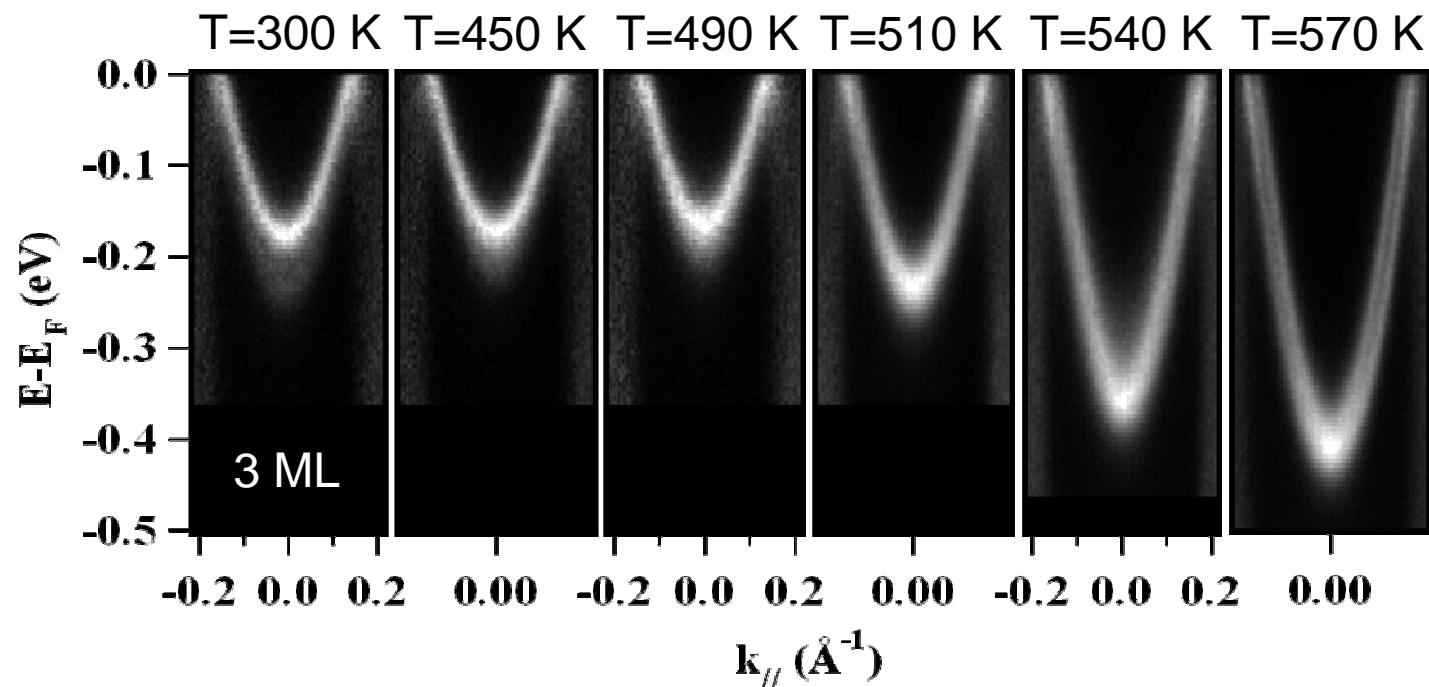


$P_{calc}$  : proba to be in the Ag film

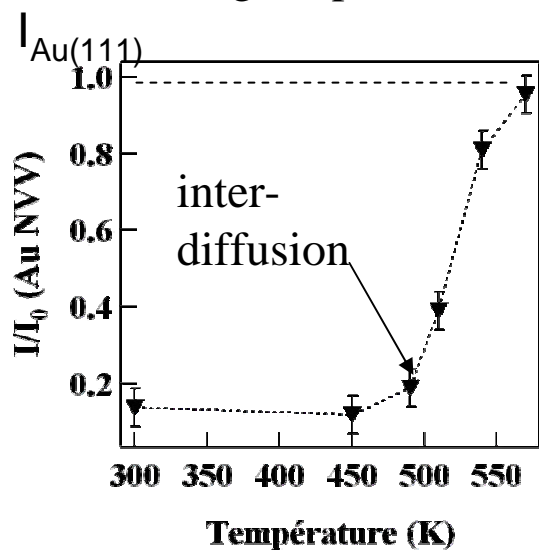
$$\alpha_R^{eff} = p_{calc} \alpha_R^{Ag} + (1 - p_{calc}) \alpha_R^{Au}$$



# 3 ML Ag /Au(111)

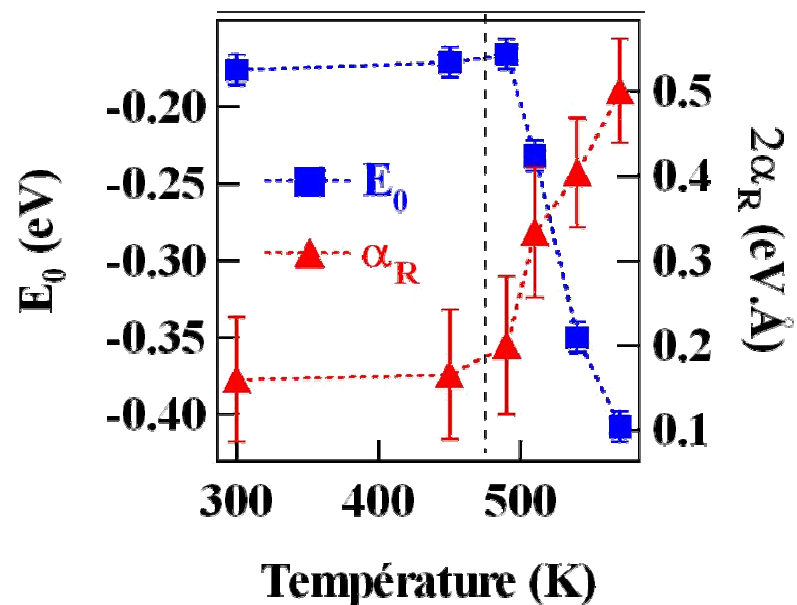


Auger Spectr.

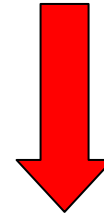
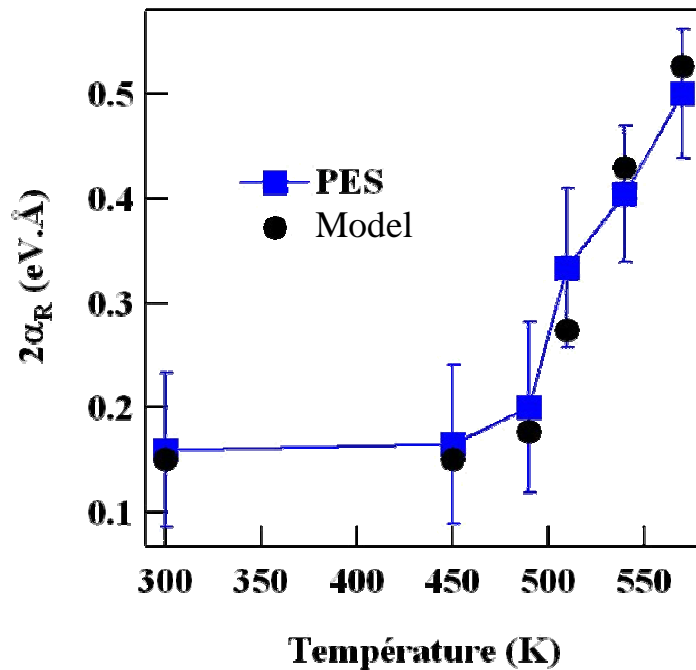


Formation of a surface alloy at 500 K  $\text{Au}_x\text{Cu}_{1-x}$

$x \nearrow$  with  $T \nearrow$



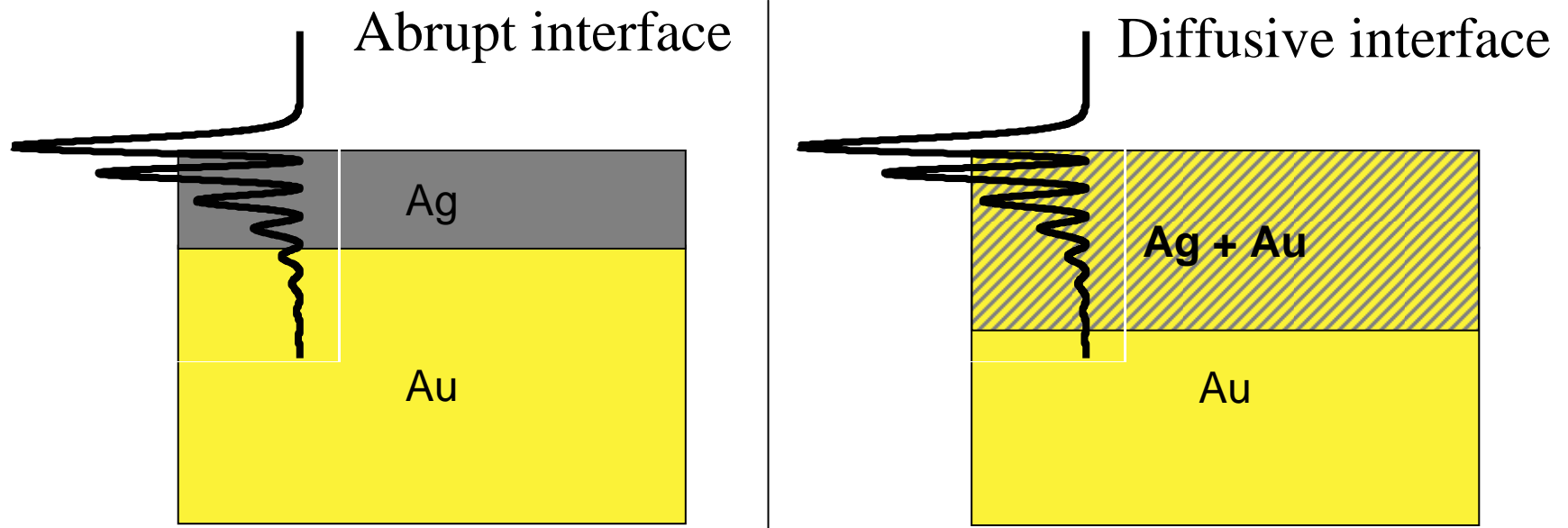
From Auger spectroscopy and a model of diffusion we can estimate the relative number of Au atoms probed by the S.S. as a function of temperature



We assume that the Rashba coefficient is simply an average of the pure Au and Ag ones weighed by  $p_{Au}(T)$  and  $p_{Ag}(T)$

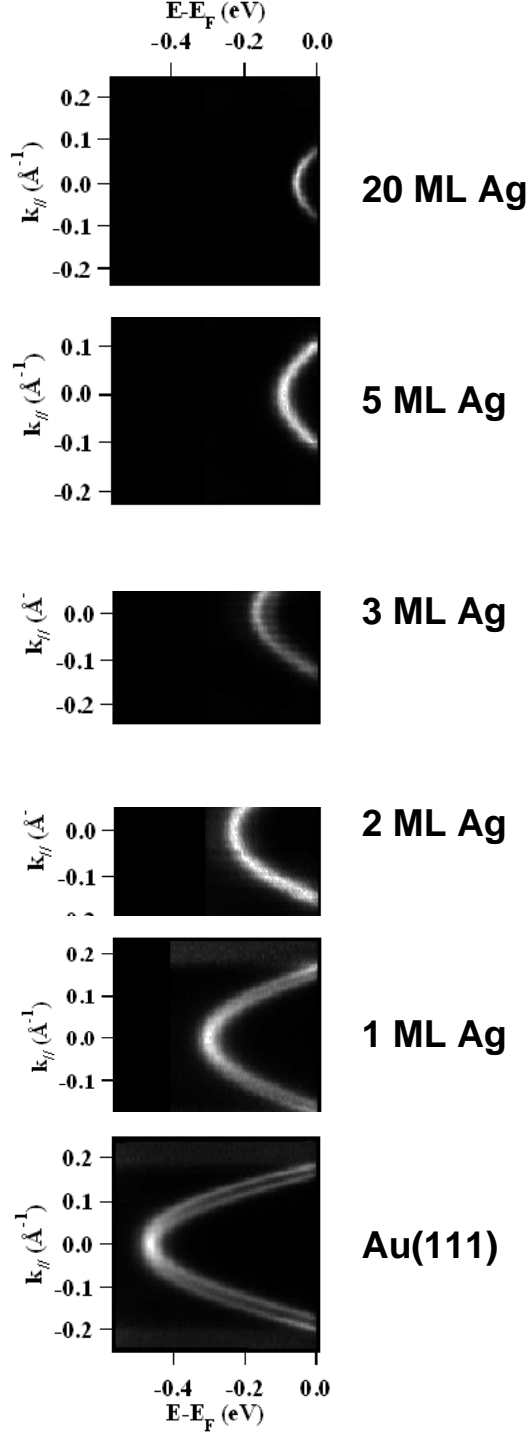
$$\alpha_R^{\text{eff}}(T) = p_{Ag}(T) \alpha_R^{\text{Ag}} + p_{Au}(T) \alpha_R^{\text{Au}}$$

## conclusion

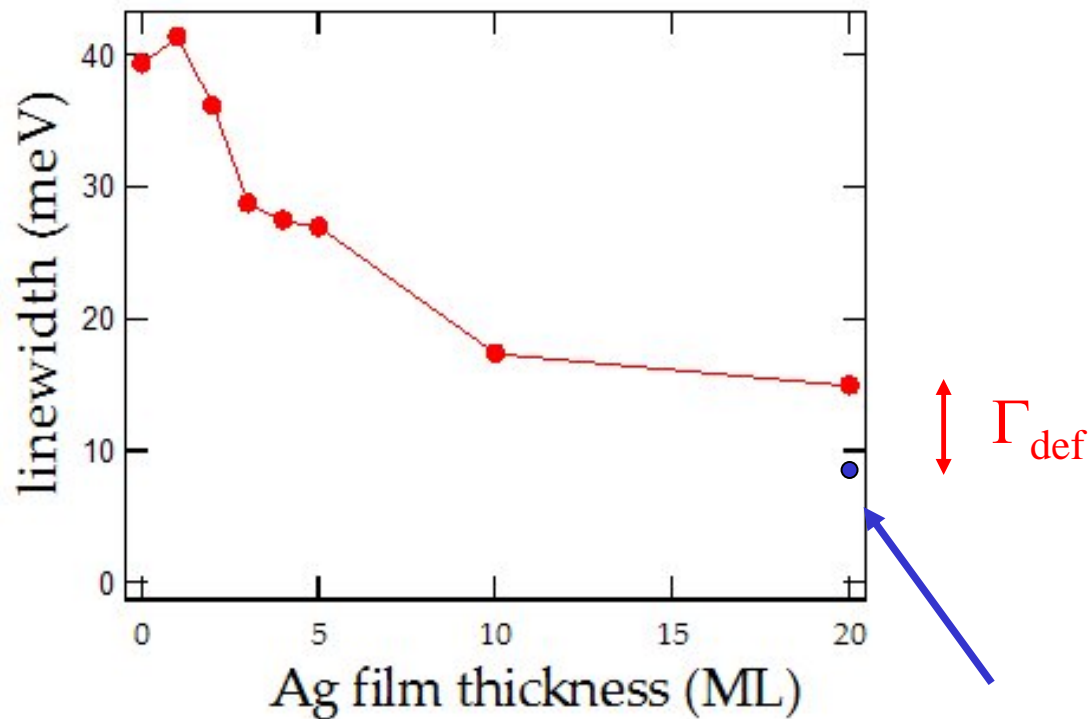


$\alpha_R$  is proportionnal to the Au atom number probed by the surface state

The spin-orbit splitting is dominated by atomic character



Lifetime evolution at  $\bar{\Gamma}$  as a function of energy

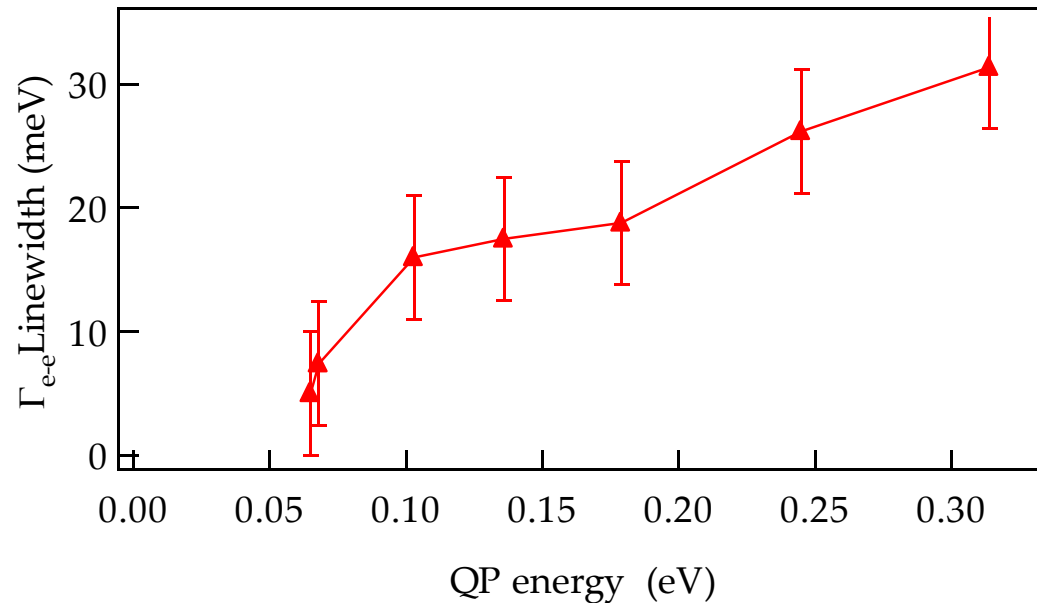


Best ARPES Linewidth  
 $\Delta E = 9$  meV in Ag crystal  
 Nicolay PRB (2000)



$$\Gamma_{e-e} = \Gamma_{\text{exp}} - (\Gamma_{e\text{-ph}} + \Gamma_{\text{def.}})$$

By assuming constant  
def and e-ph contributions  
 $\Gamma_{e\text{-ph}} + \Gamma_{\text{def.}} = 10 \text{ meV}$



it seems that  $\Gamma_{e-e}$  does not depend quadratically with QP energy!

## CONCLUSION

We have shown evolutions of the different electronic properties in the Ag/Cu(111) and Ag/Au(111) interfaces as a function of Ag film thickness (SS energy, spin-orbital, QP lifetime...)



- interfaces can be considered as model systems because the electronic parameters can be modified nearly continuously
- the quality of the interfaces is crucial for the study of intrinsic properties like electron-electron lifetime

**A. Bendounan, H. Cercellier, Y. Fagot-Revurat, B. Kierren, S. Pons**

*Laboratoire de Physique des Matériaux, Université H. Poincaré,  
F-54506, Vandoeuvre-les-Nancy (France)*

**V. Yu. Yurov** (Ag/Cu)

*Natural Science Center of General Physics Institute –  
RAS Moscow 117942 Vavilova Str 38 – Russia*

**D. Popovic, F. Reinert** (Ag/Au)

*Universität des Saarlandes, Fachrichtung 7.2 - Experimentalphysik,  
D-66041, Saarbrücken (Germany)*