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Spin-Resolved Angle-Resolved Photoemission

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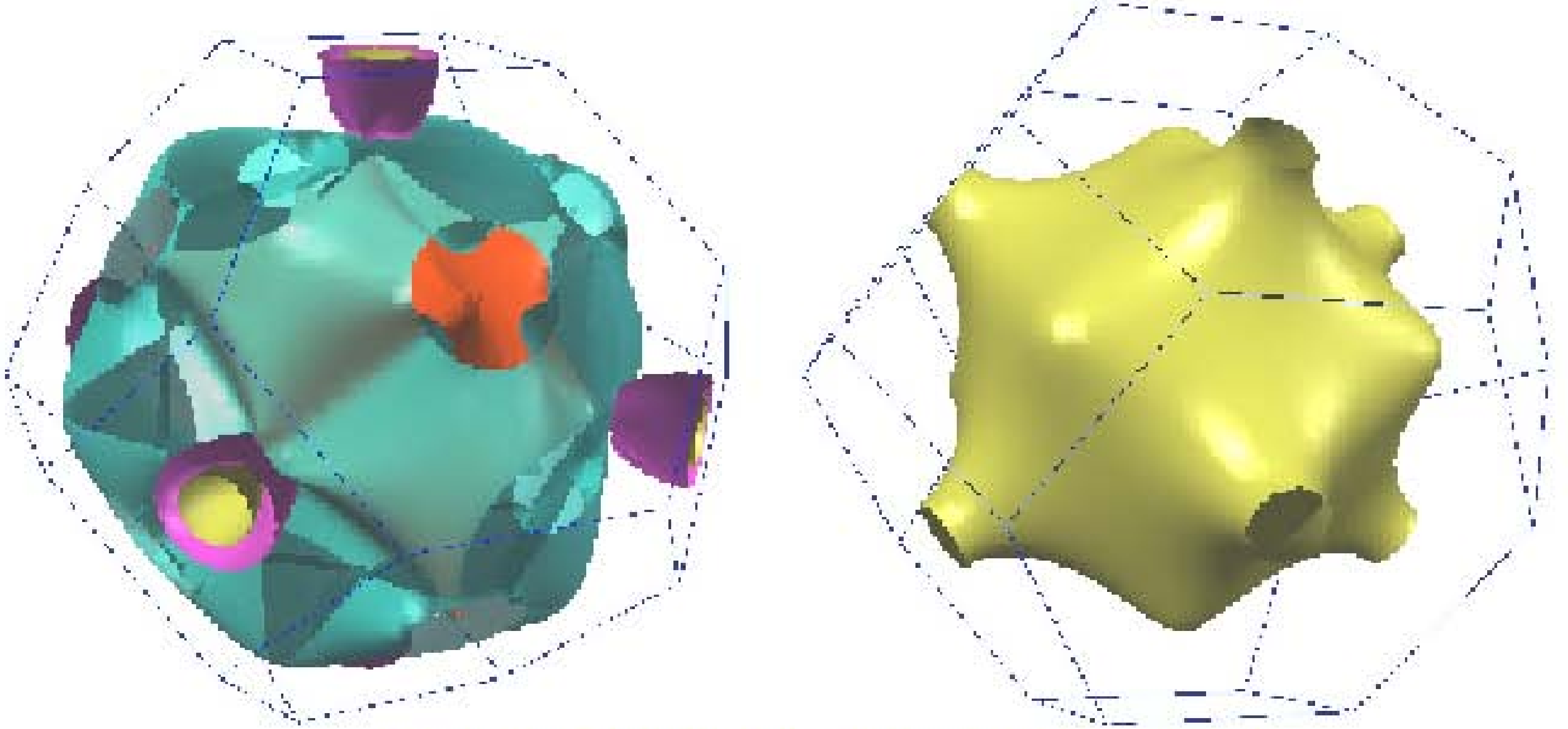
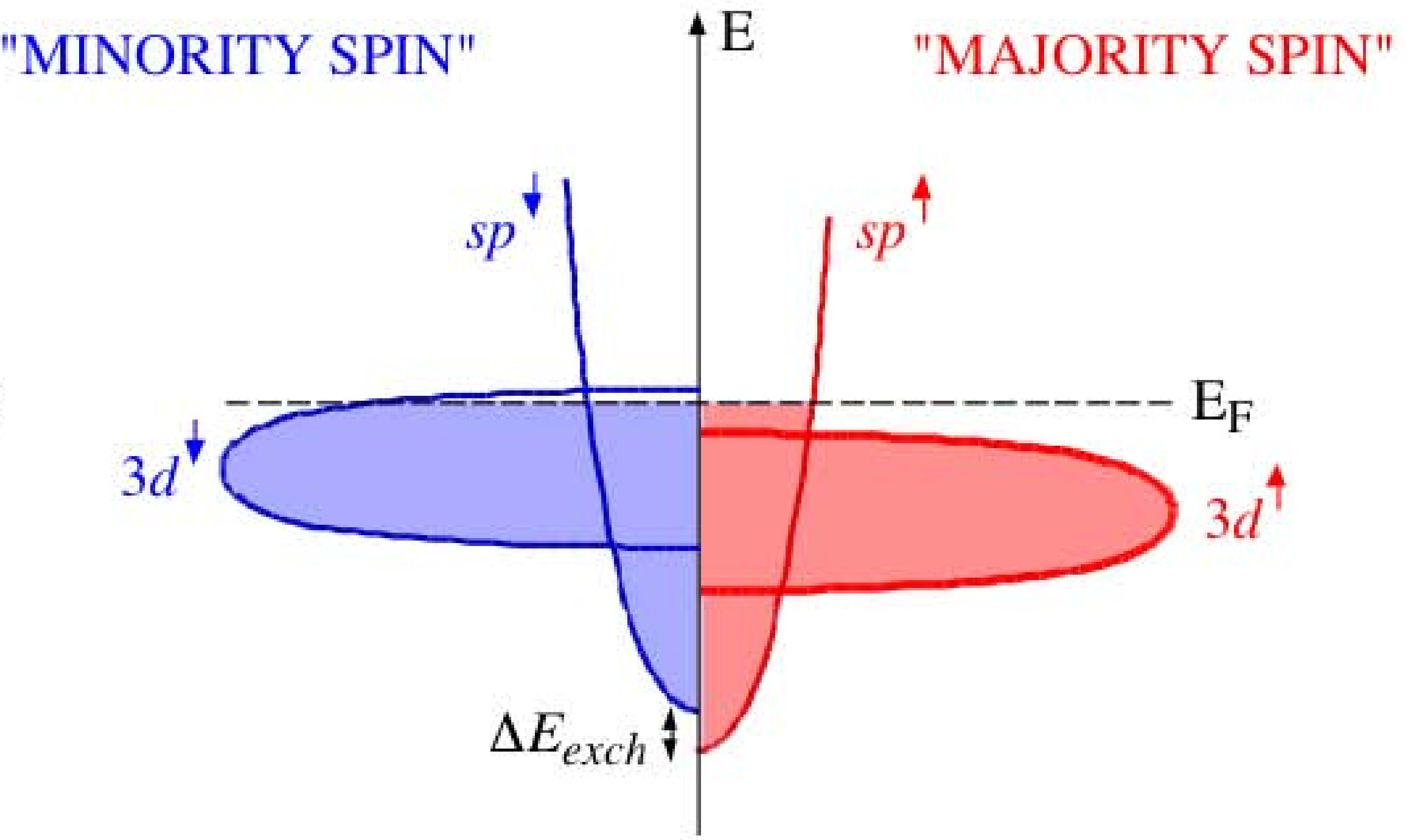
Lecture 4
- Band Structure of an Itinerant Ferromagnet
- Measuring the Electron Spin
- Quantitative Aspects of Spin Polarization: Ni(111)
- The Magnetic Phase Transition in Ni
- Ultrathin Films of Ni on Cu(001)
- The Rashba Effect in the Surface State on Au(111)

The Fermi Surface of Nickel Metal

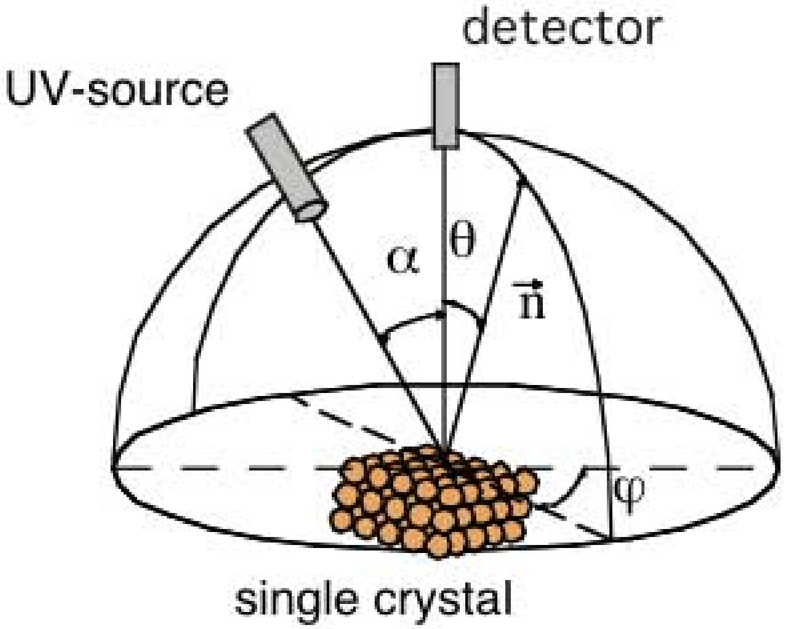


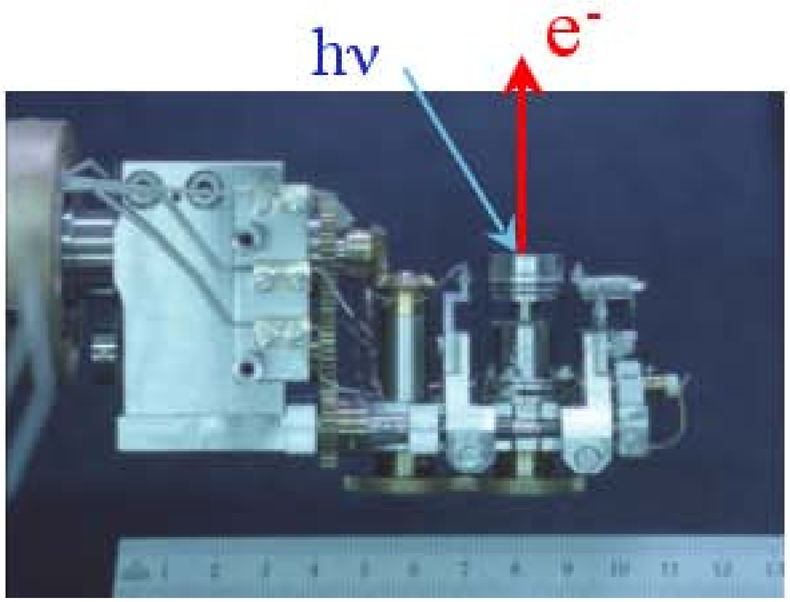
From http://www.phy.tu-dresden.de/~fermisur/

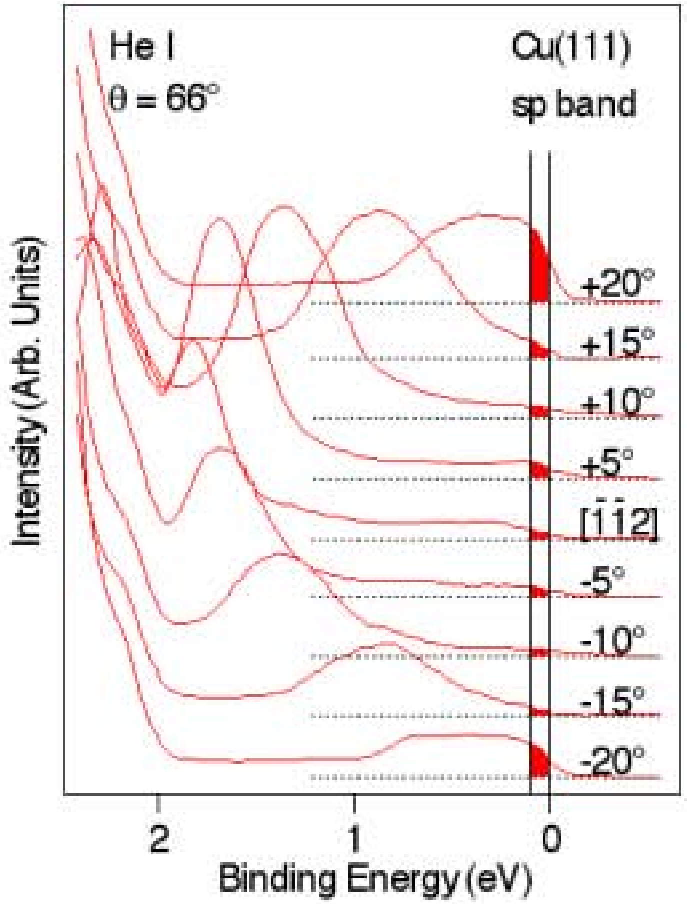
Schematic Density of States

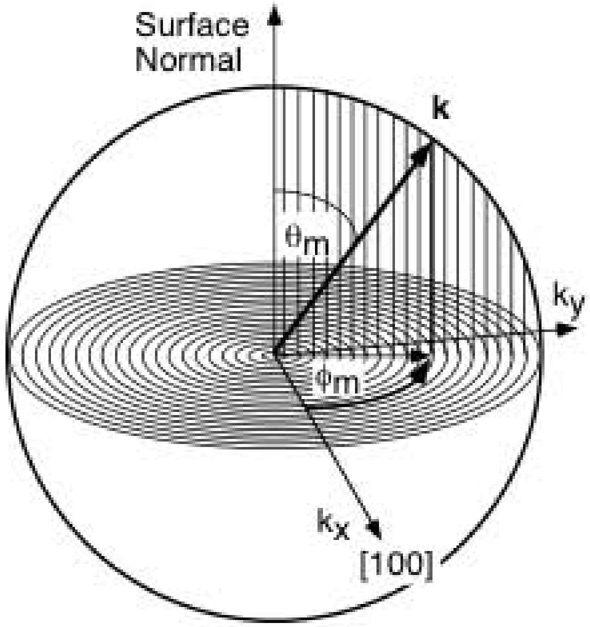


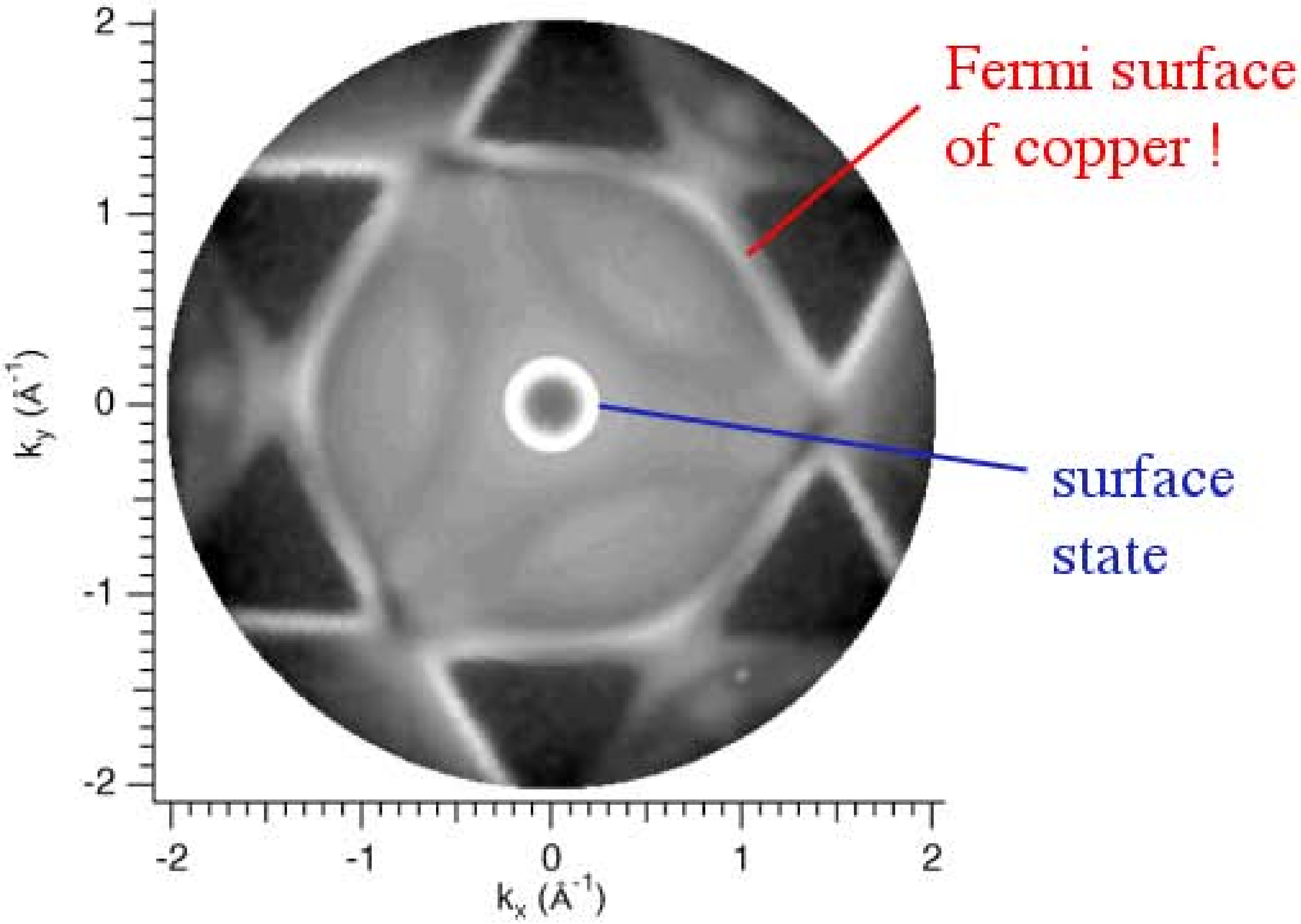
Fermi Surface Mapping by Photoemission







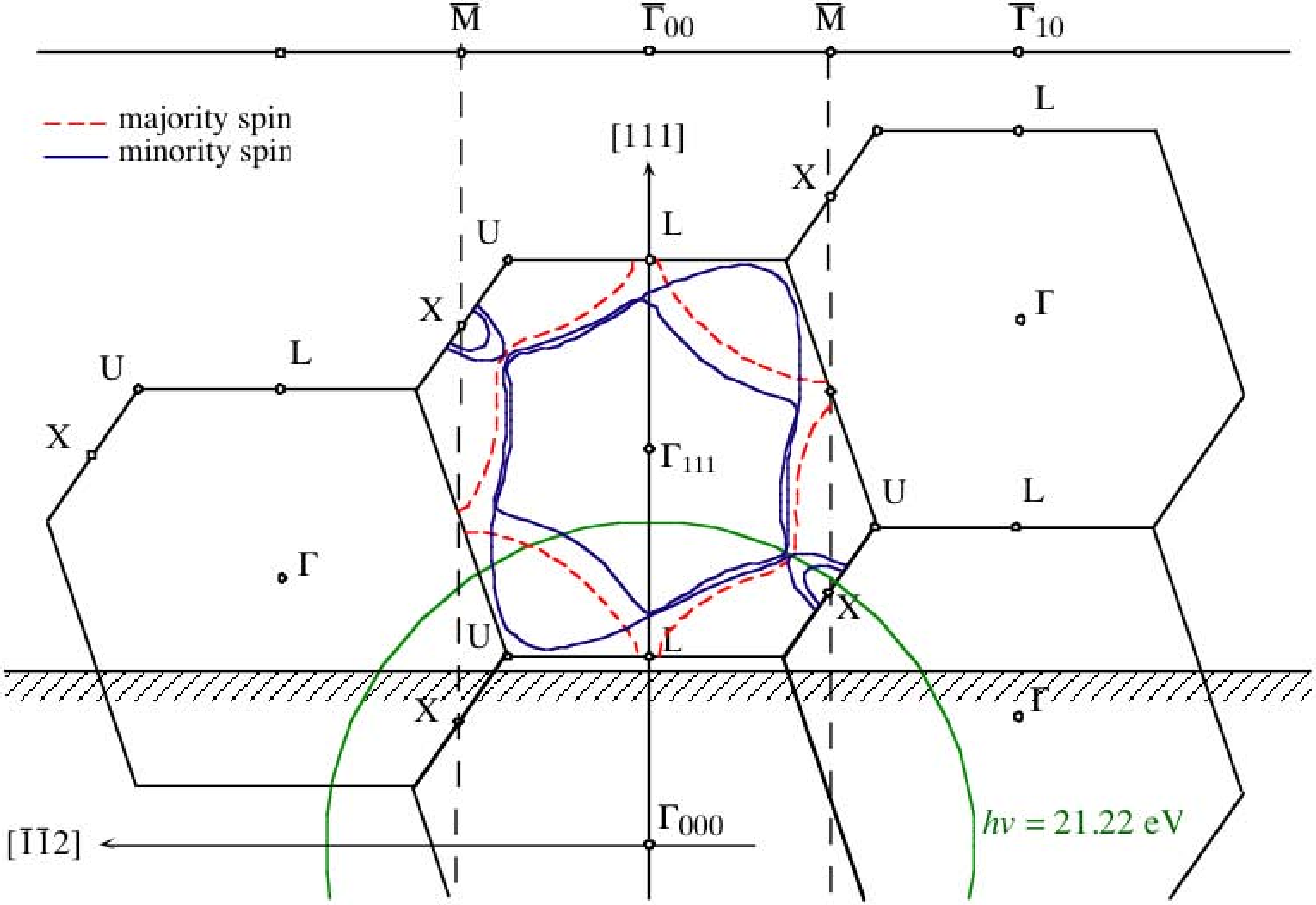




Fermi surface of copper !

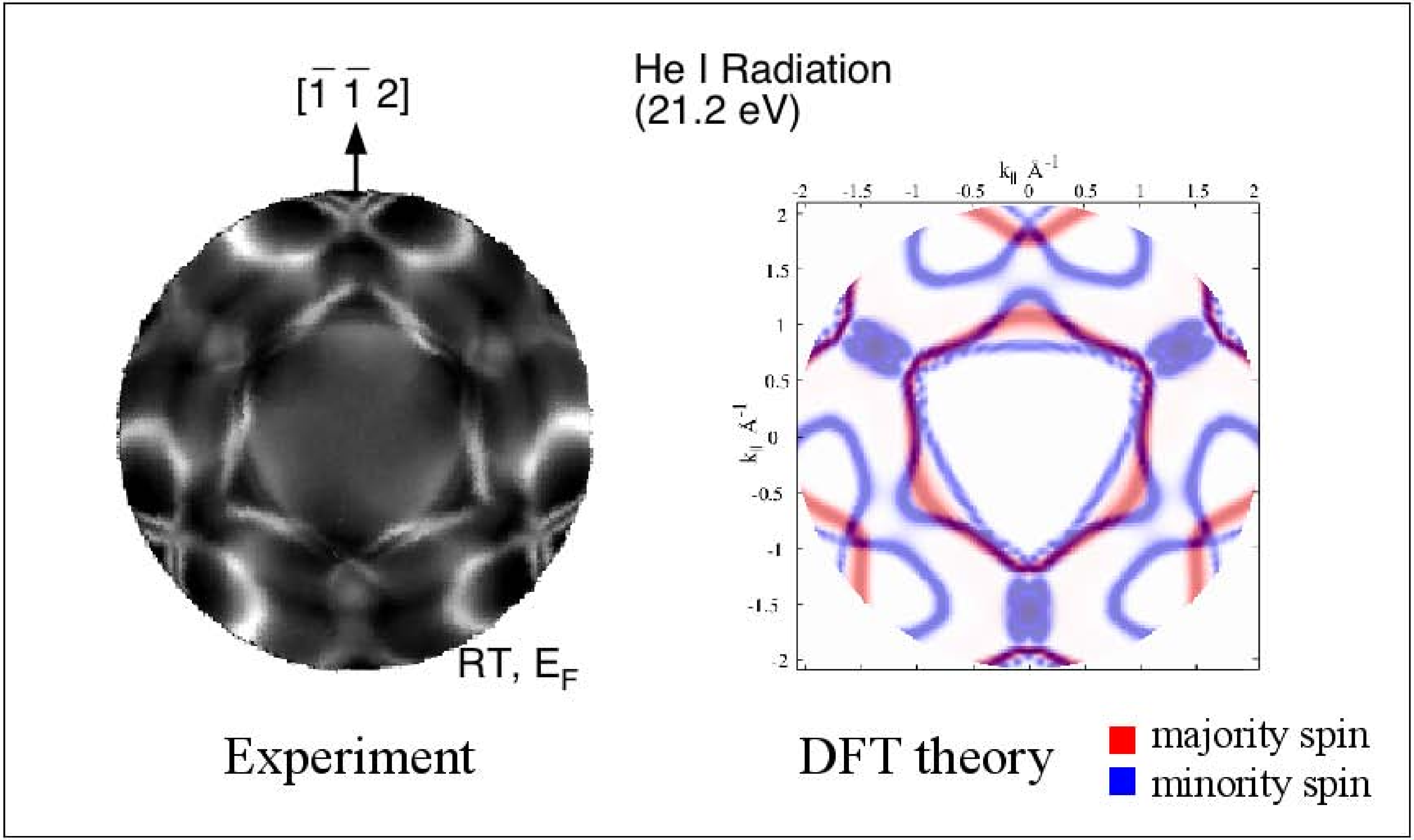
surface state

Reciprocal Space on Ni(111)



Fermi surface calculation (density functional theory, Wien 97)

Fermi Surface of Ni as Seen Through the Ni(111) Surface



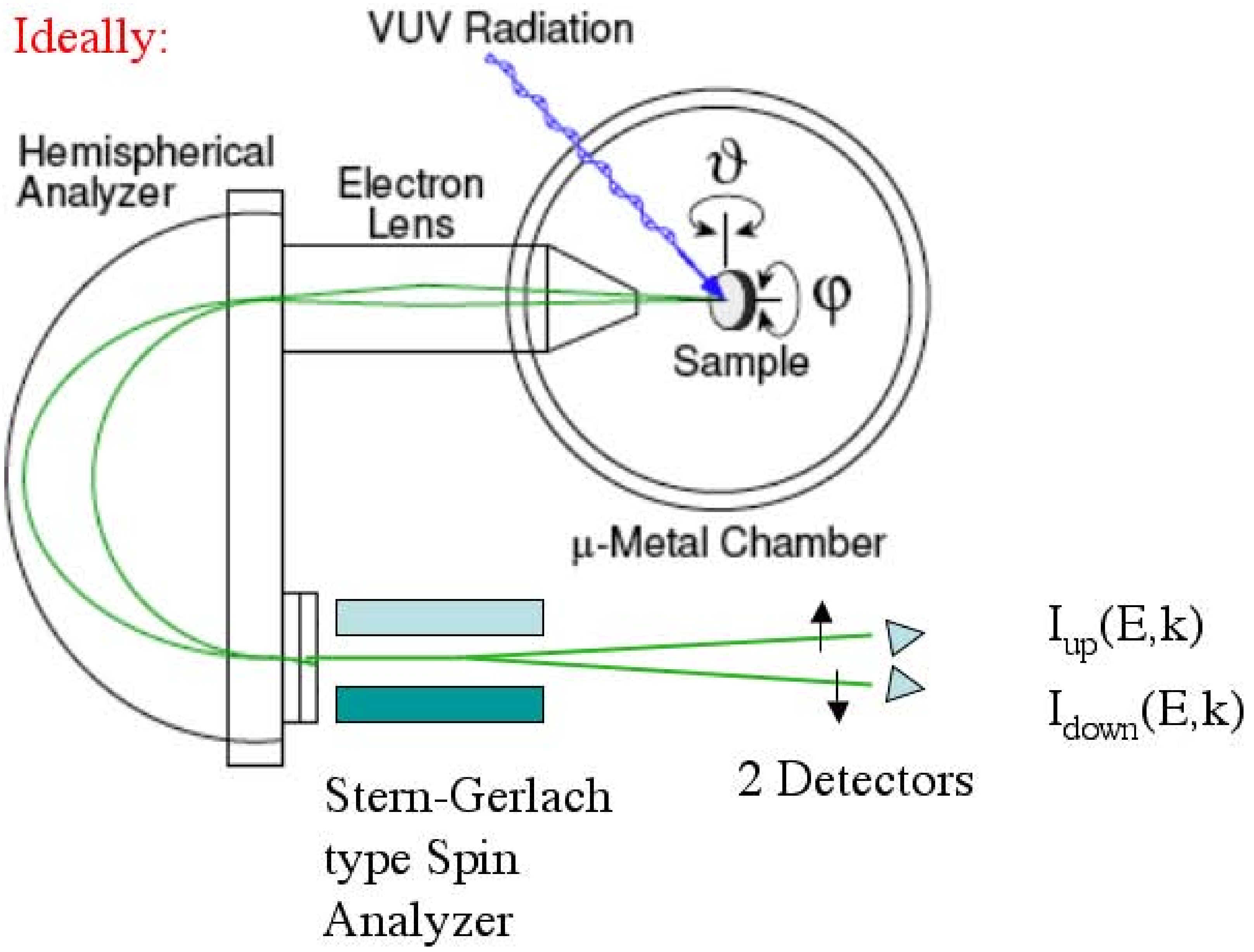
Experiment

DFT theory

M. Hoesch et al., JES 124, 263 (2002)

ARPES with Spin Resolution

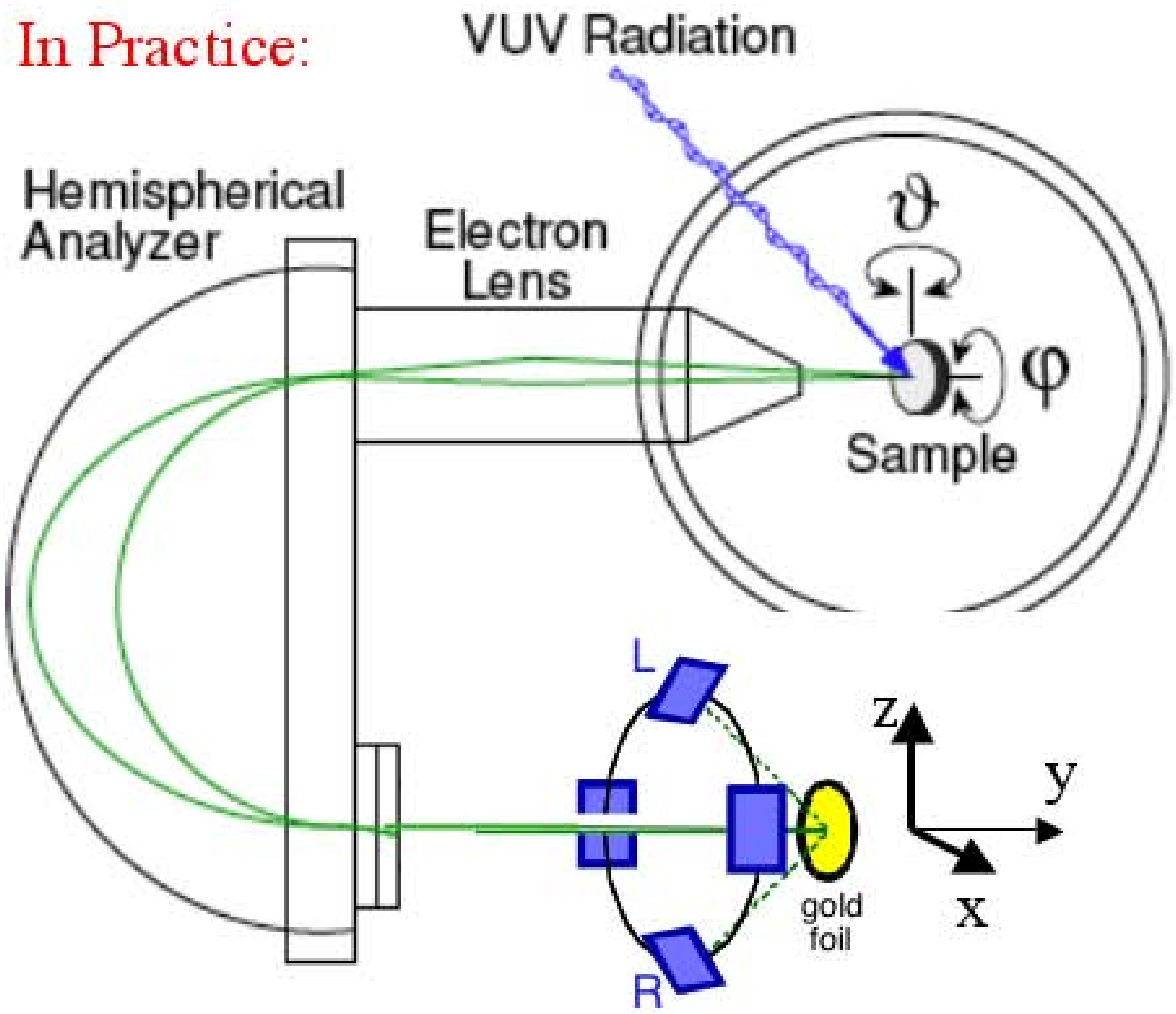
Ideally:



Does not work! (Lorentz forces & uncertainty principle)

ARPES with Spin Resolution

In Practice:



Mott Scattering
(~ 50 keV)

two orthogonal axes are spanned
by two pairs of detectors

4 Detectors

Scattering asymmetry

$$A_x = \frac{(I_L - I_R)}{(I_L + I_R)}$$

Spin Polarization

$$P_x = A_x/S.$$

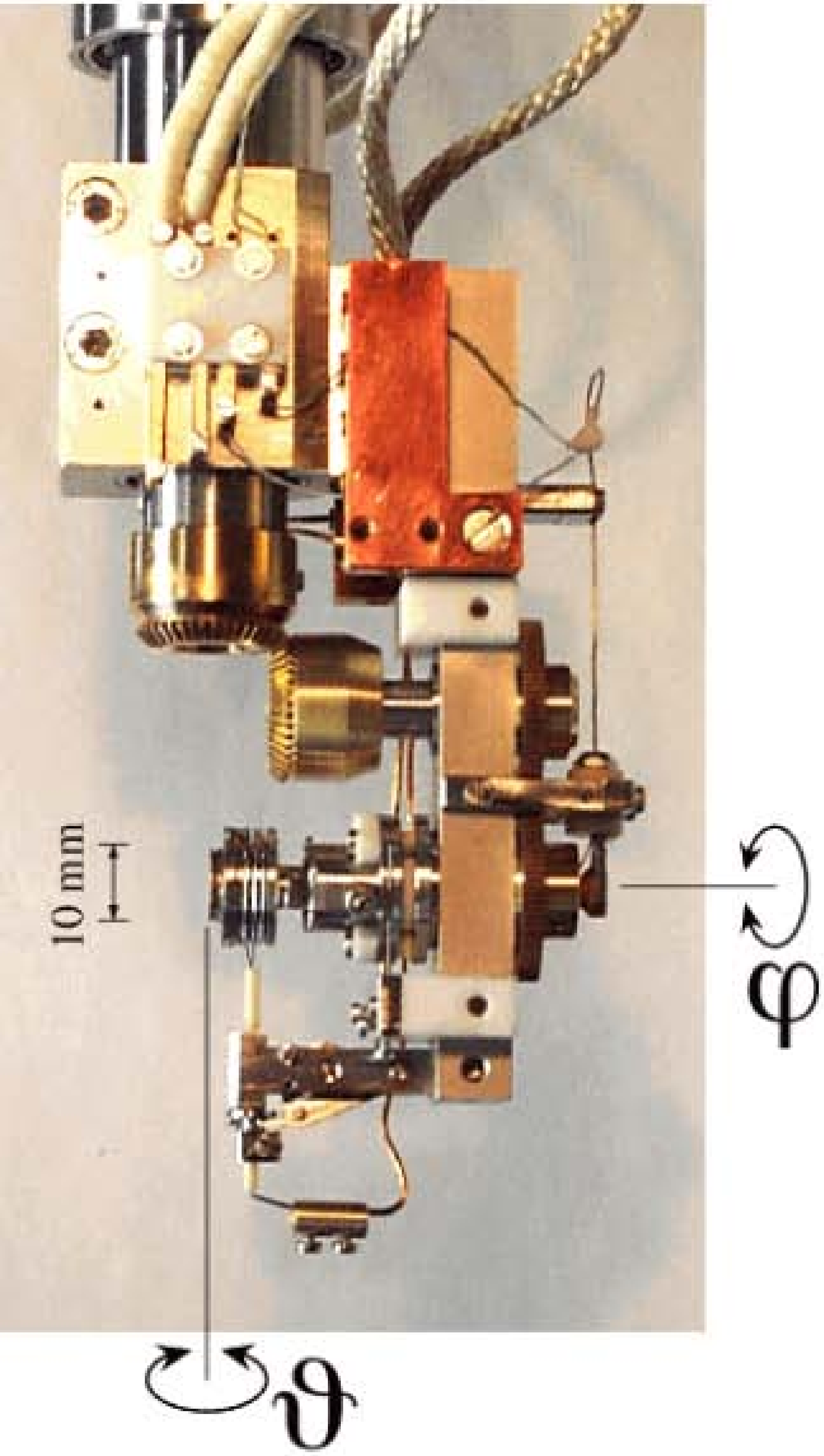
S: Sherman function

Inefficient !

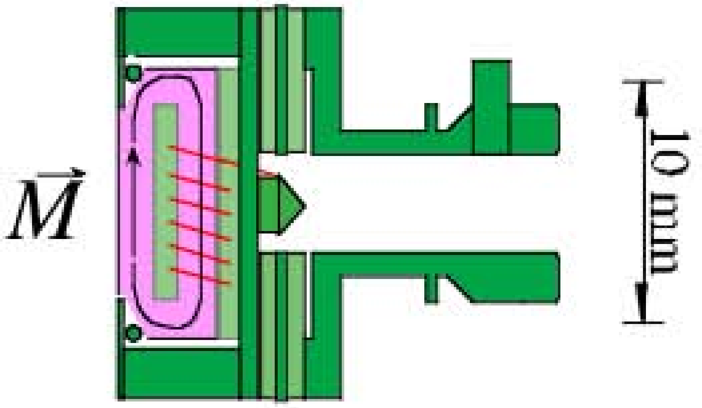
Spin Polarization: $$P_I(E,\boldsymbol{k}) = \frac{I_\uparrow(E,\boldsymbol{k}) - I_\downarrow(E,\boldsymbol{k})}{I_\uparrow(E,\boldsymbol{k}) + I_\downarrow(E,\boldsymbol{k})},$$

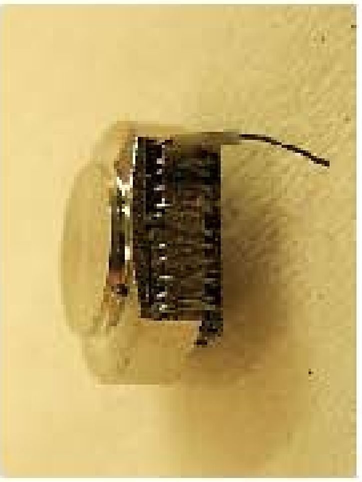
$$\boxed{I_{\uparrow,\downarrow}(E,\boldsymbol{k}) = I_M(E,\boldsymbol{k})(1 \pm P_I(E,\boldsymbol{k}))/2.}$$

Important: Control over the sample magnetization in ARPES !



We need a magnetized sample (ideally a single domain) to measure spin polarized bands !

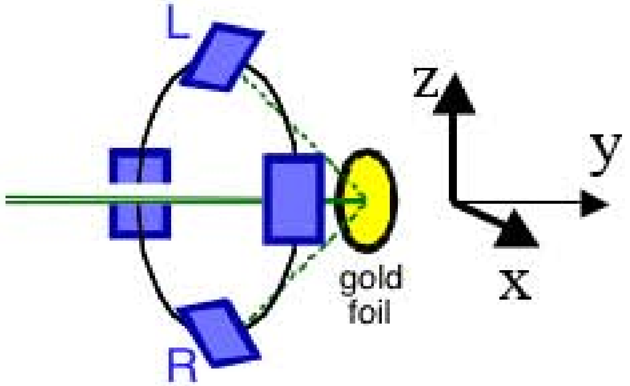




Switching magnetization direction (⊕ ⊖)

⇒ Forming cross asymmetries cancels instrumental asymmetries

$$A^{\otimes} = \frac{(I_L^{\oplus}+I_R^{\ominus})-(I_R^{\oplus}+I_L^{\ominus})}{(I_L^{\oplus}+I_R^{\ominus})+(I_R^{\oplus}+I_L^{\ominus})}.$$



two orthogonal axes are spanned by two pairs of detectors

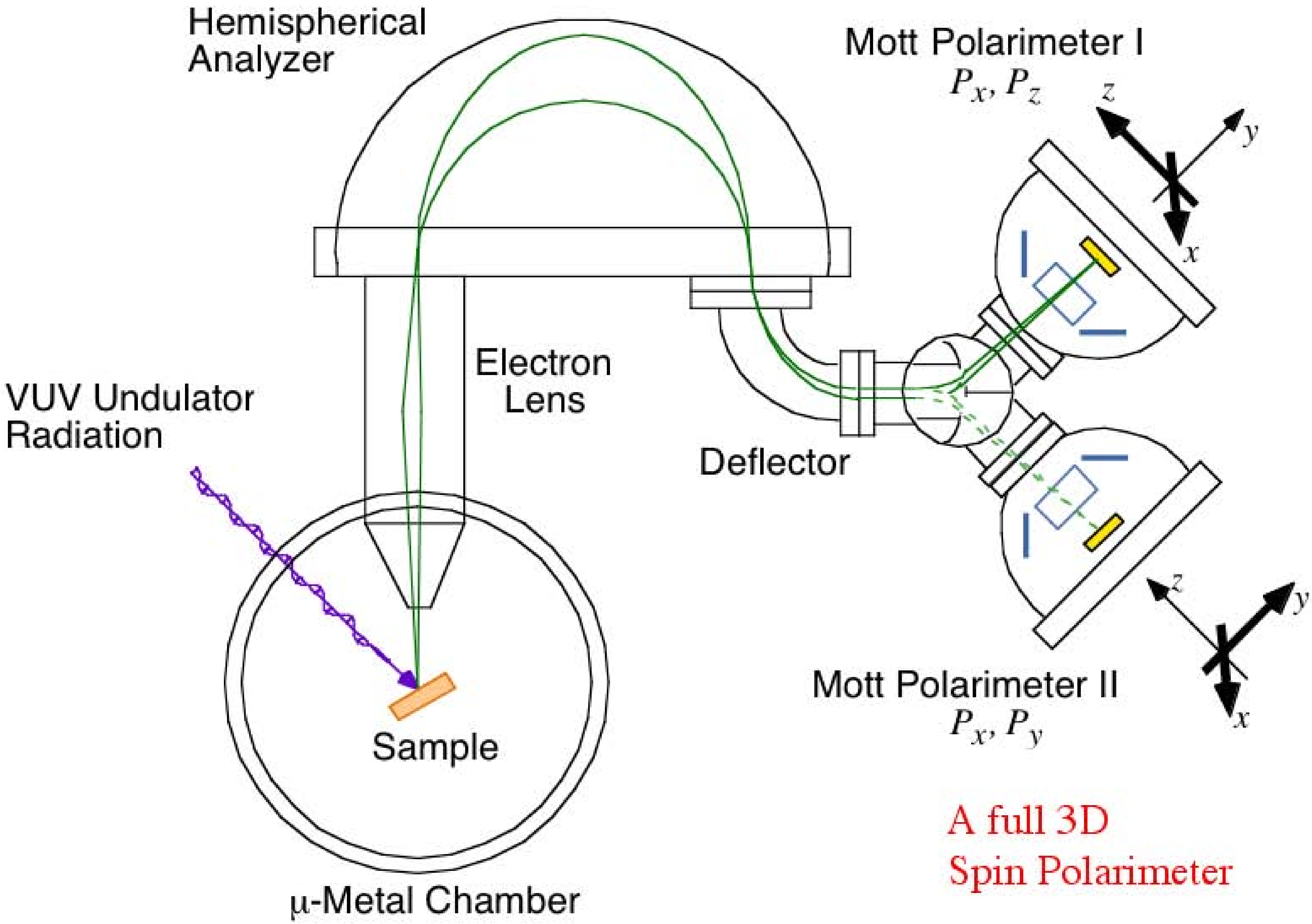
Polarization P = A / S

S ~ 0.15, but not precisely known (requires calibration)

⇒ P = 0 is measured exactly !

⇒ P scale is known roughly

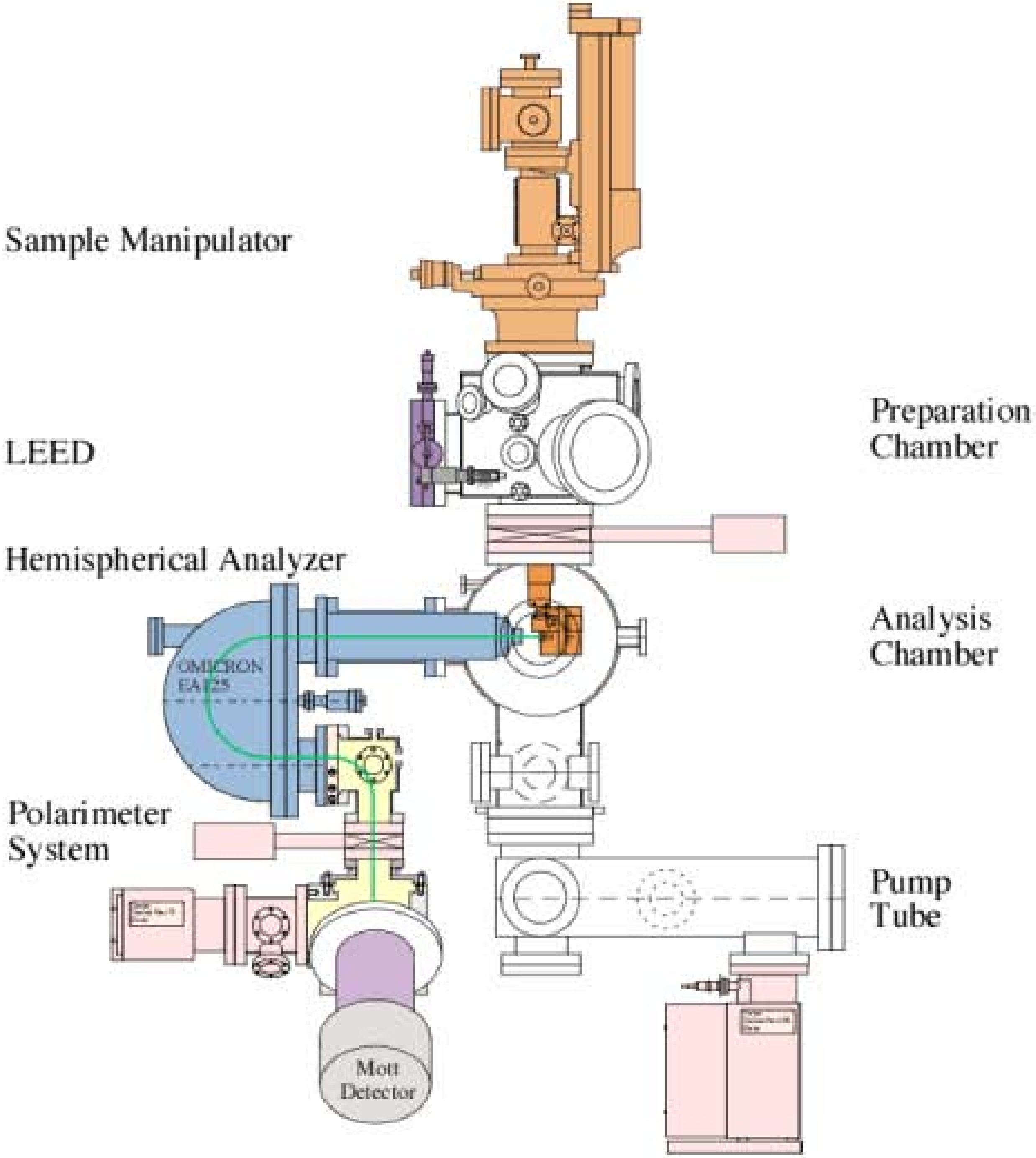
COPHEE - The Complete Photoemission Experiment

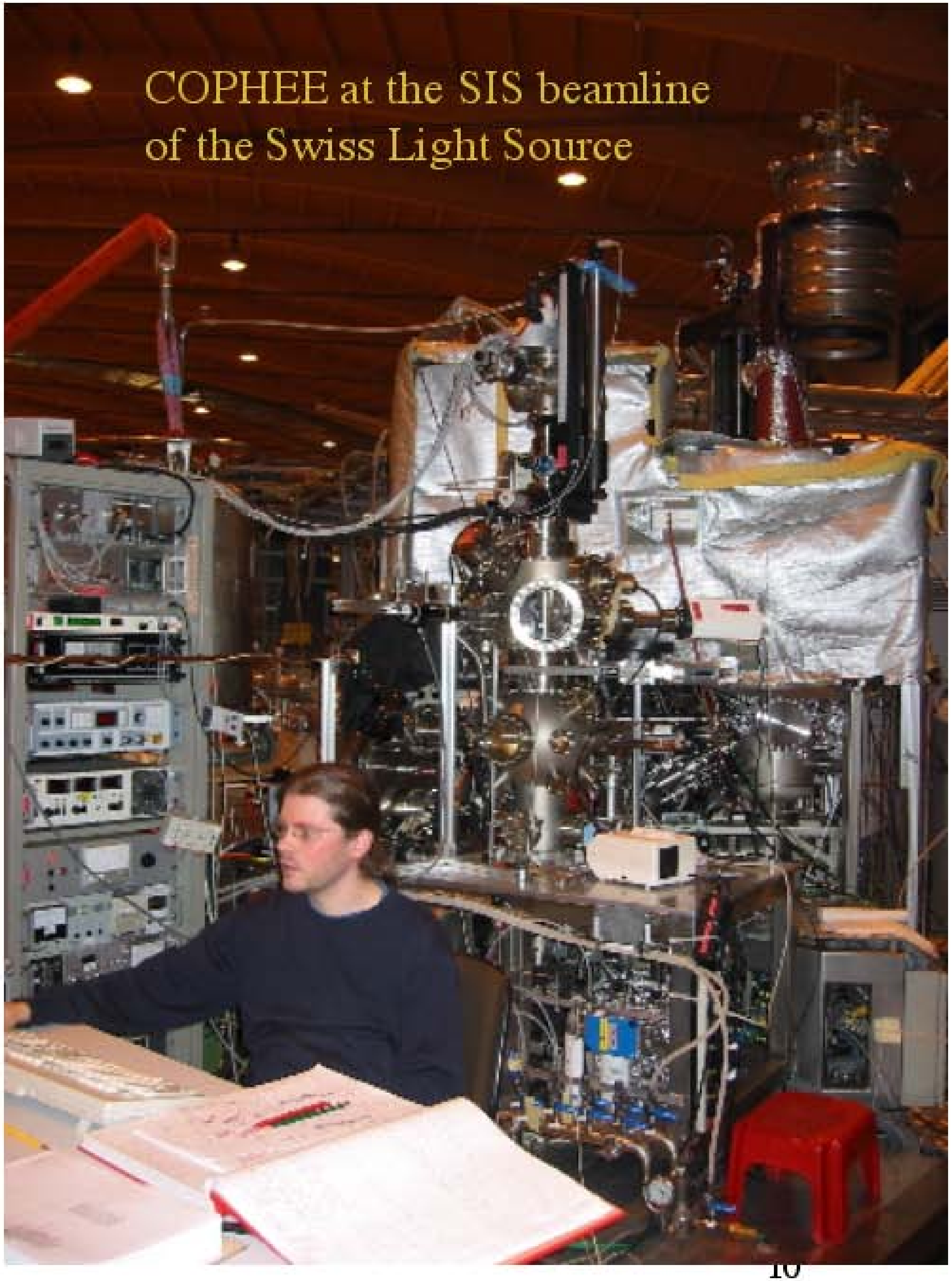


A full 3D Spin Polarimeter

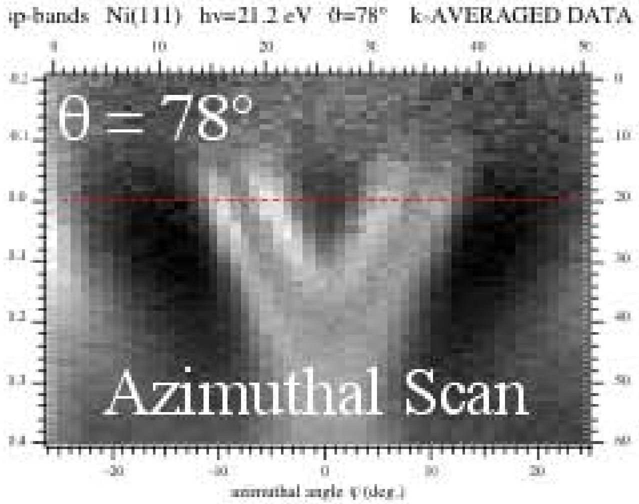
M. Hoesch, Ph. D. Thesis, 2002

Setup of COPHEE:



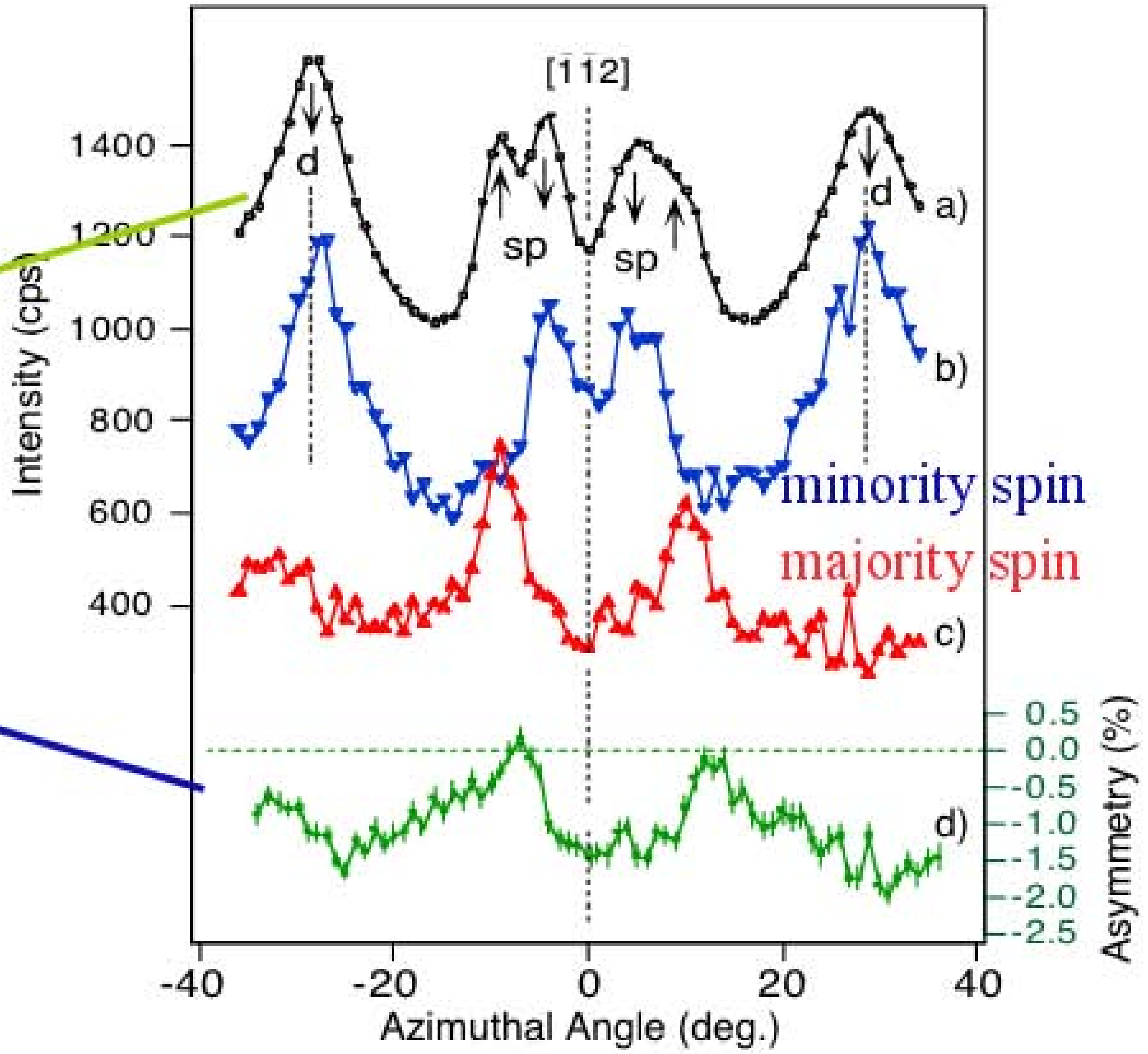

COPHEE at the SIS beamline of the Swiss Light Source

Spin-Resolved Momentum Mapping on Ni(111)



$$I_{\uparrow,\downarrow}(E,\boldsymbol{k}) = I_M(1 \pm P_I(E,\boldsymbol{k}))/2.$$

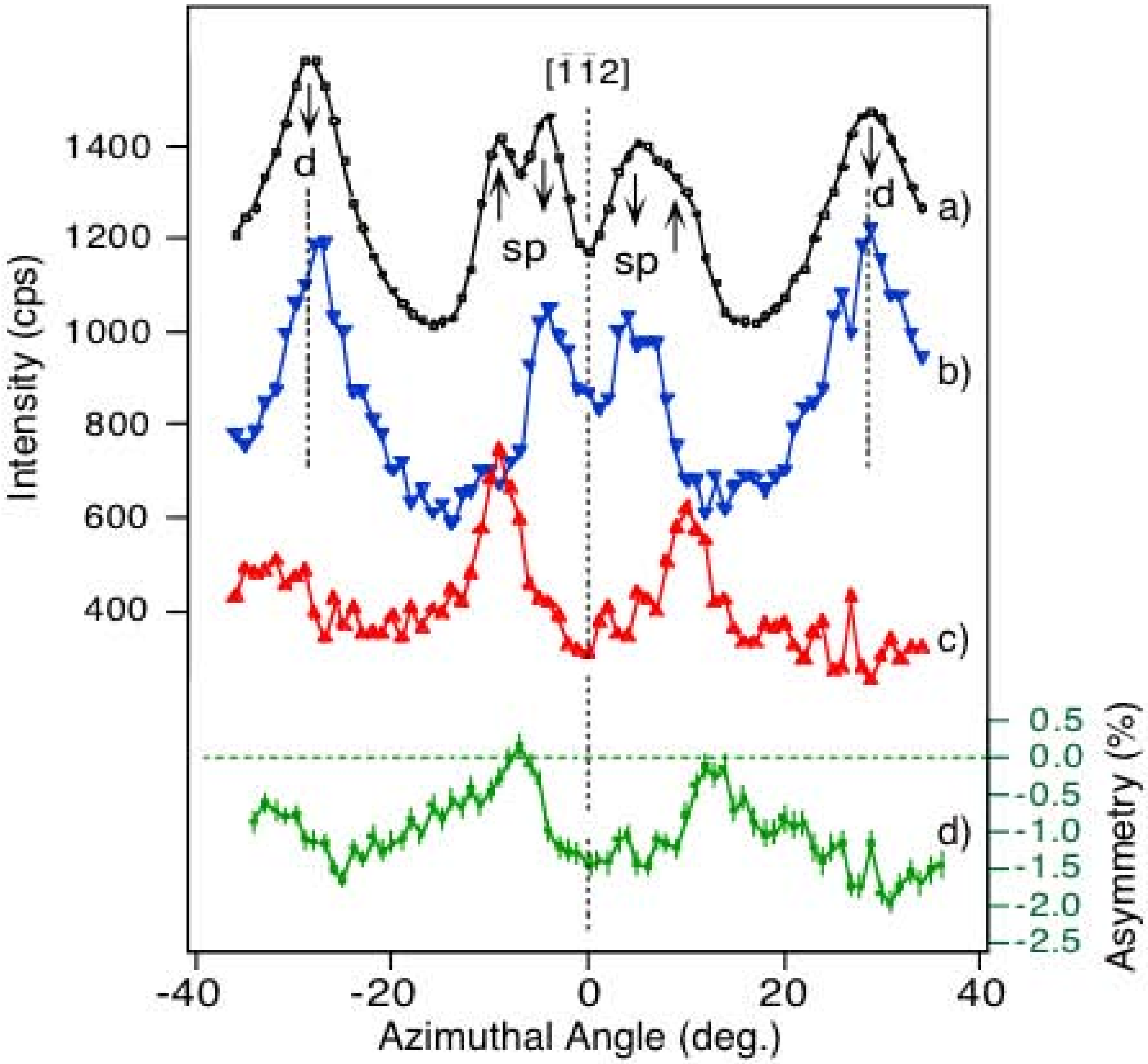
$$P_I(E,\boldsymbol{k}) = \frac{I_\uparrow(E,\boldsymbol{k}) - I_\downarrow(E,\boldsymbol{k})}{I_\uparrow(E,\boldsymbol{k}) + I_\downarrow(E,\boldsymbol{k})}.$$

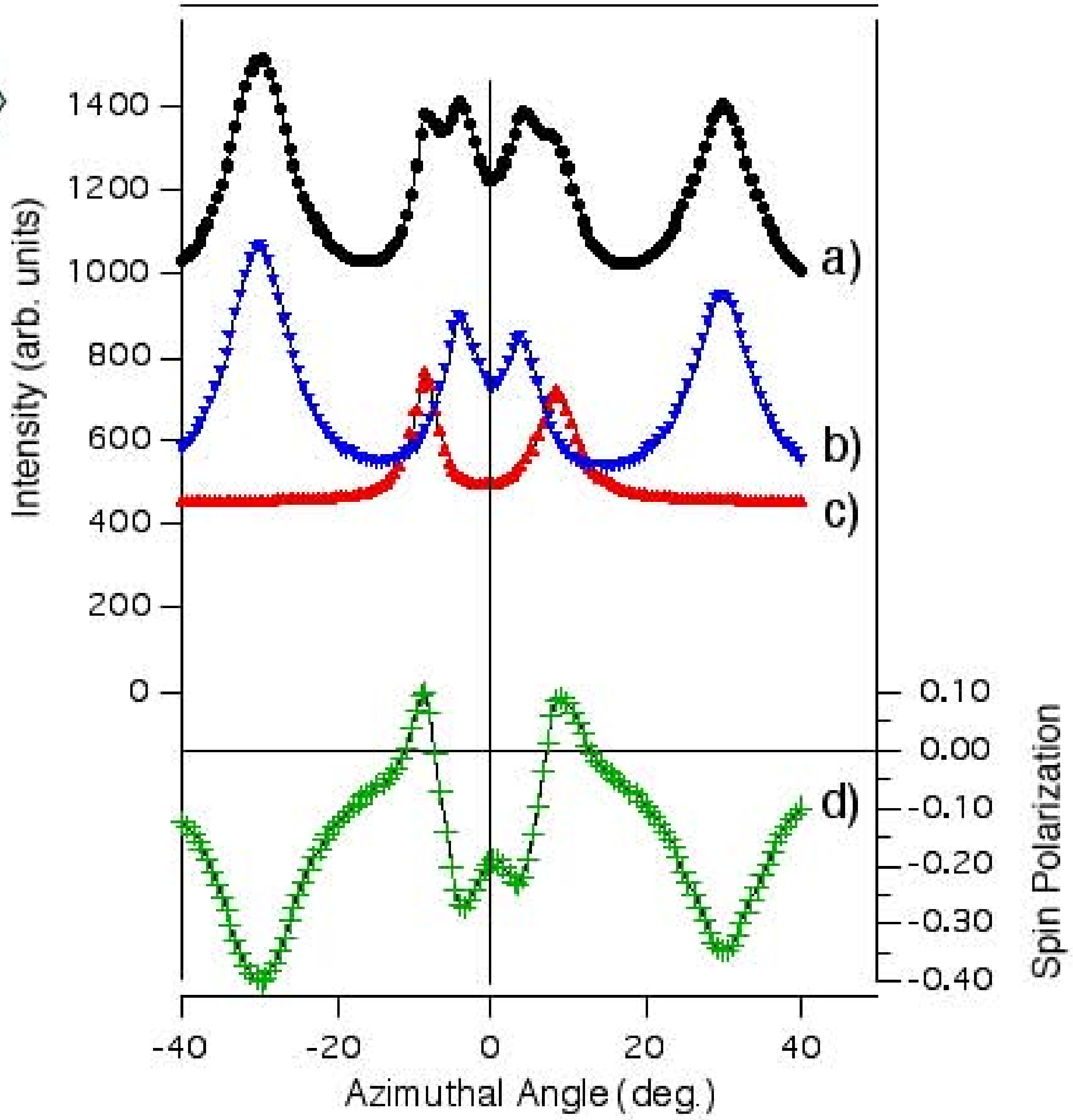


M. Hoesch et al., JES 124, 263 (2002)

Quantitative Aspects of Spin Polarization Measurements

Modelling Spin Polarization:
6 Lorentzians (2 up, 4 down)
+ constant background (S/B~0.6)

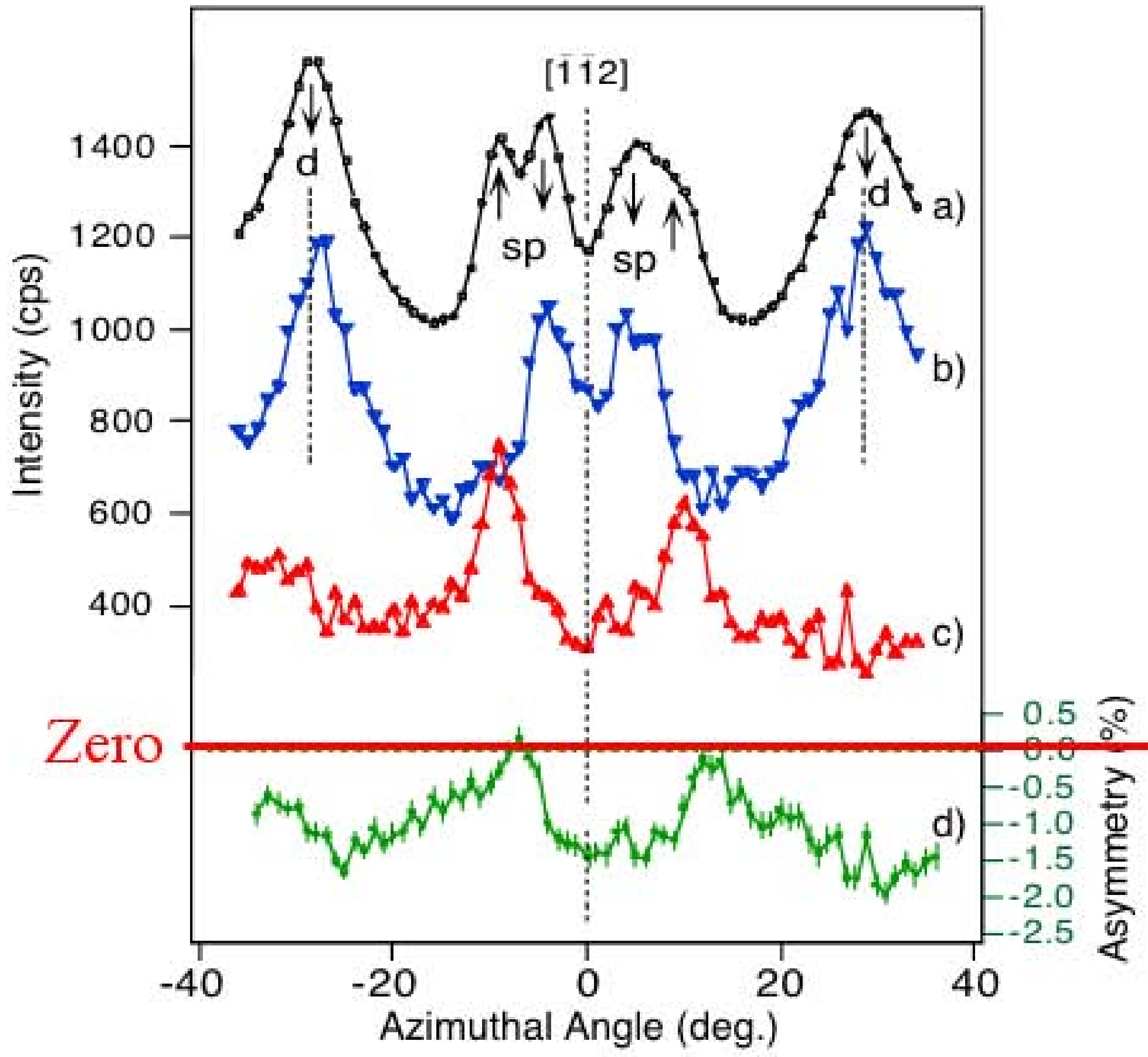


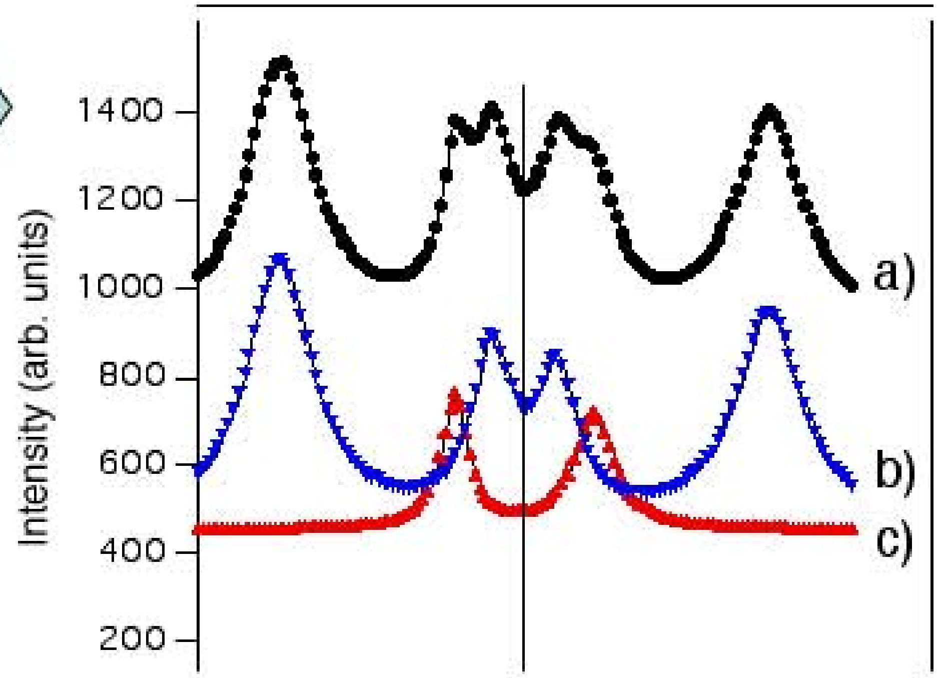


=> We can understand these asymmetry curves !

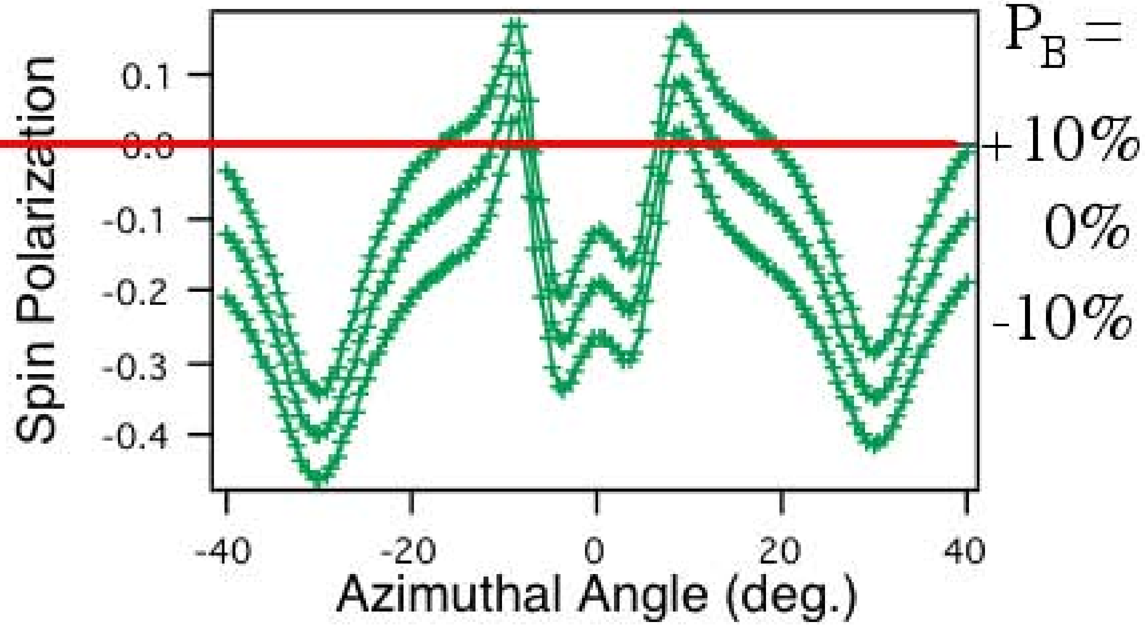
Quantitative Aspects of Spin Polarization Measurements

Modelling Spin Polarization:
6 Lorentzians (2 up, 4 down)
+ constant background (S/B~0.6)





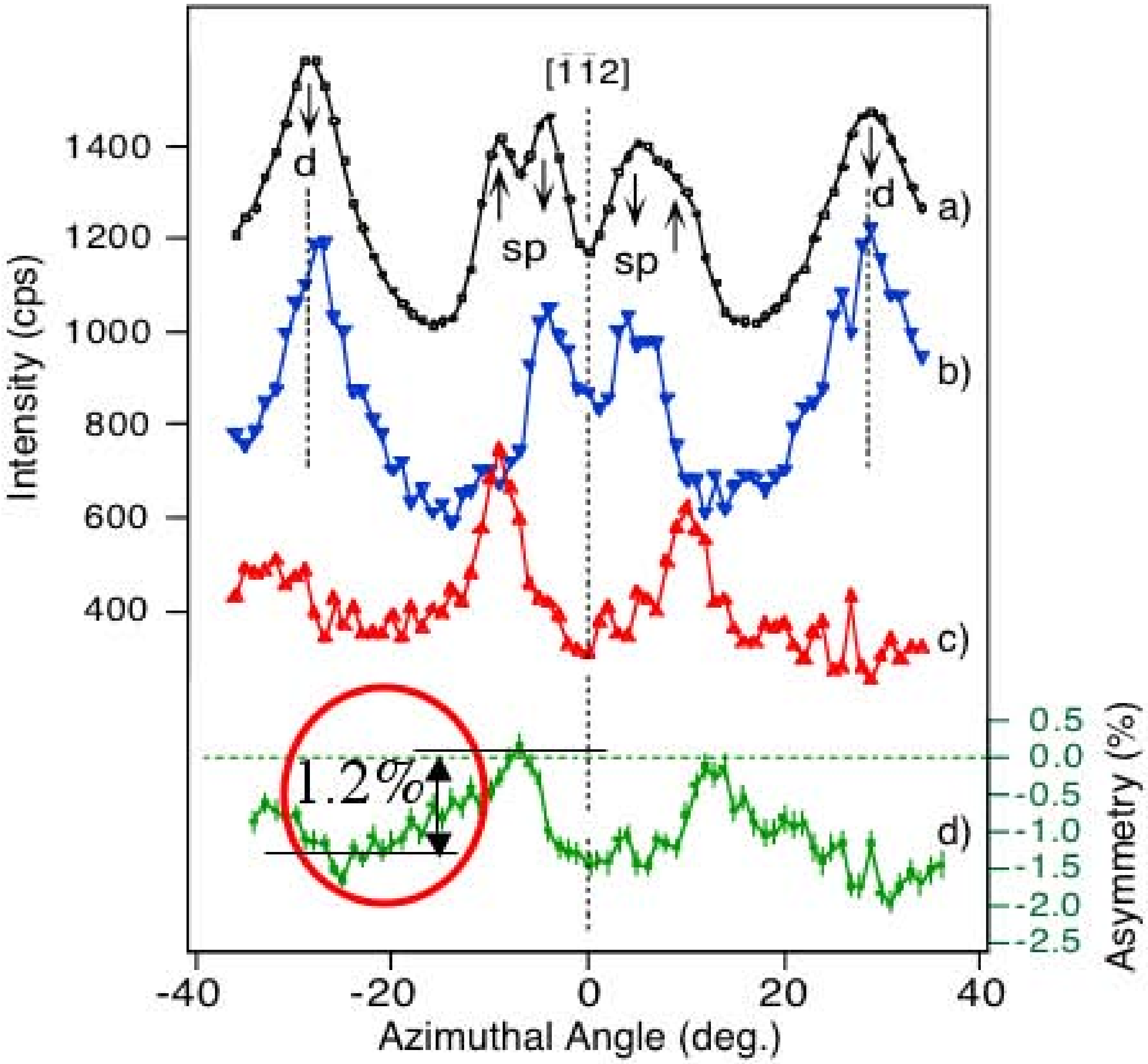
Vary polarization P_B of Background:

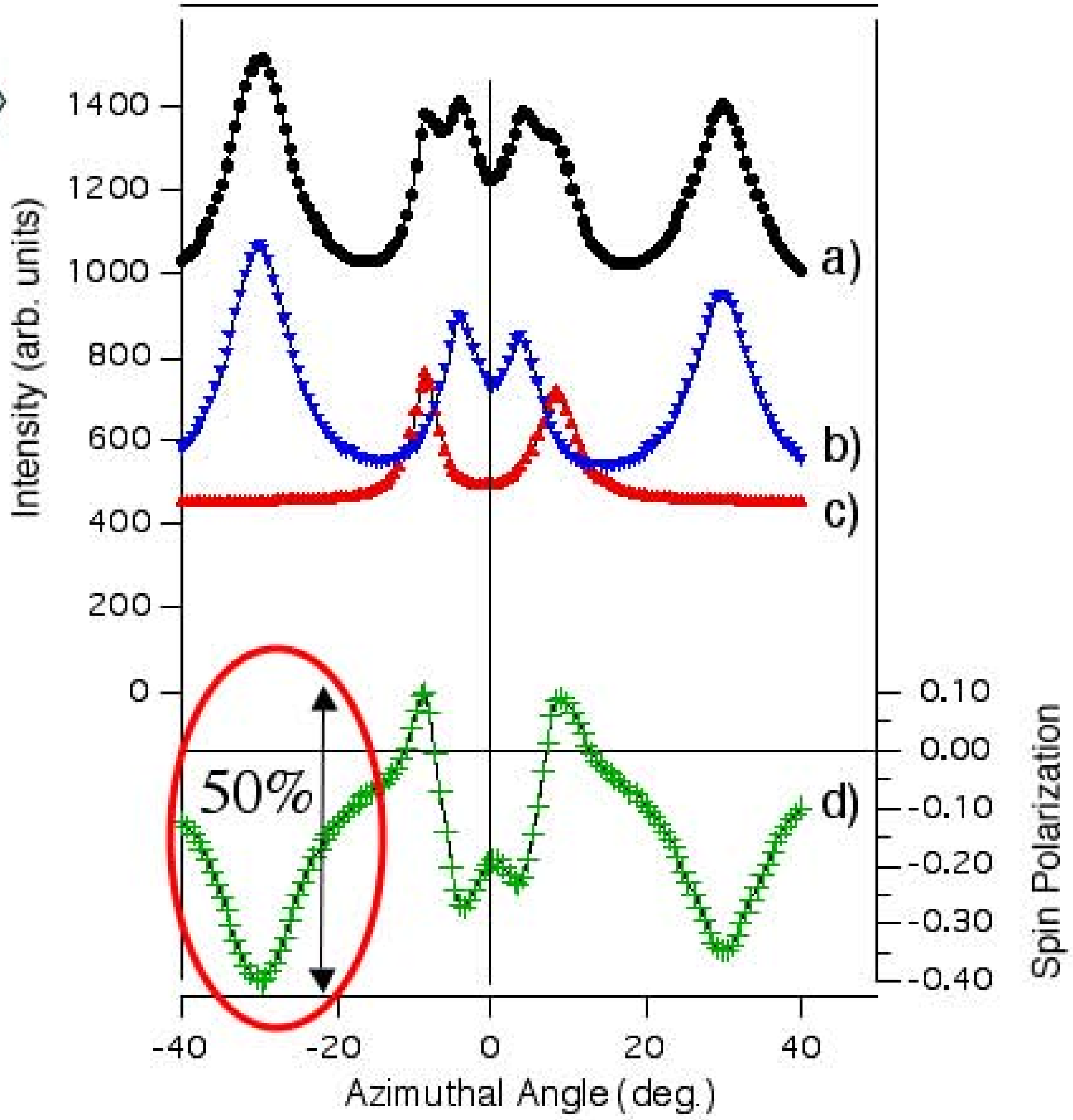


=> Background is ~ unpolarized !

Quantitative Aspects of Spin Polarization Measurements

Modelling Spin Polarization:
6 Lorentzians (2 up, 4 down)
+ constant background ($S/B \sim 0.6$)





Polarization $P = A / S$
$S \sim 0.15$, but not precisely

Here: $S_{eff} = A / P \sim 0.024$ $\Rightarrow$ Sample poorly magnetized

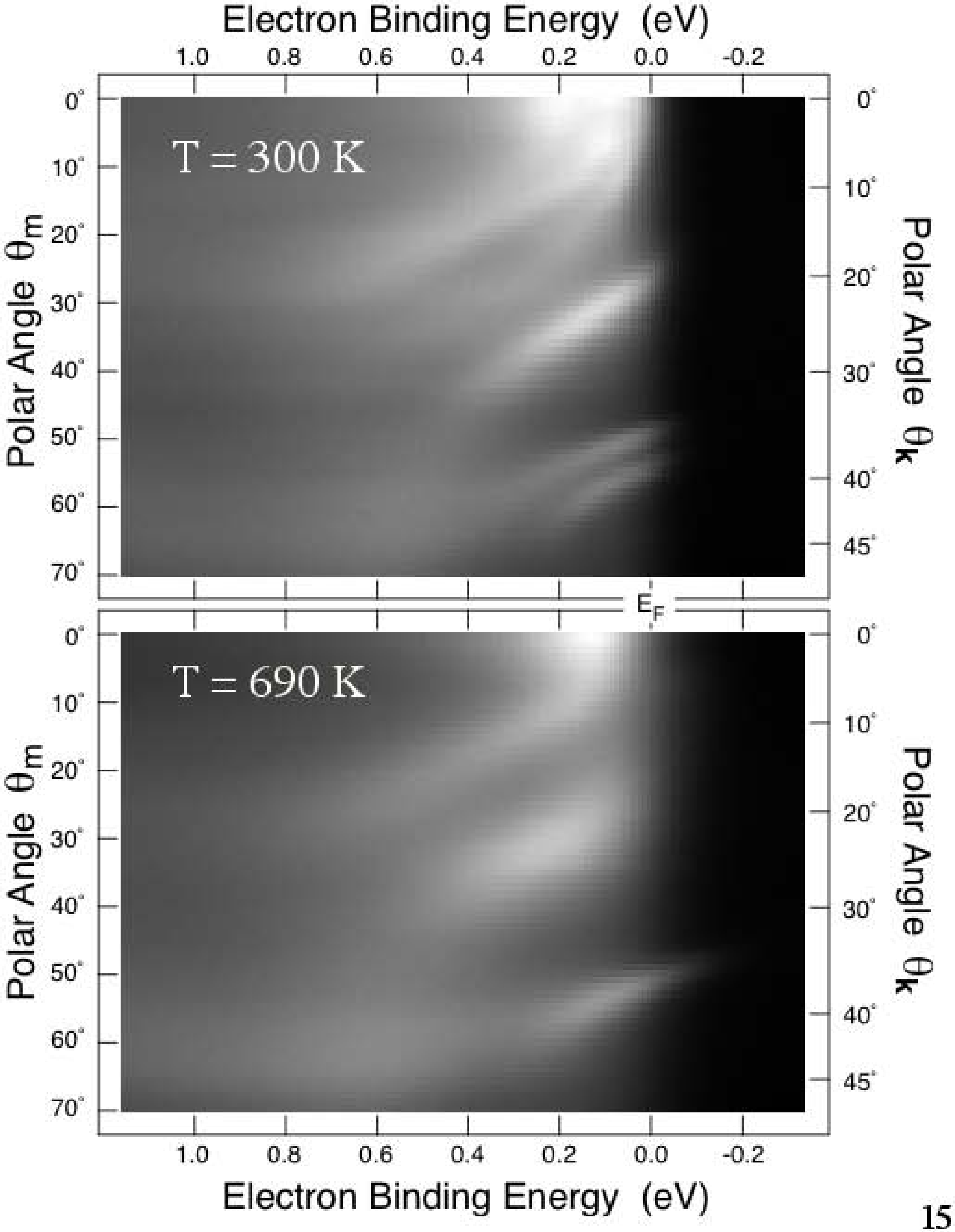
$M / M_S \sim 0.15$

Monitoring the Magnetic Phase Transition

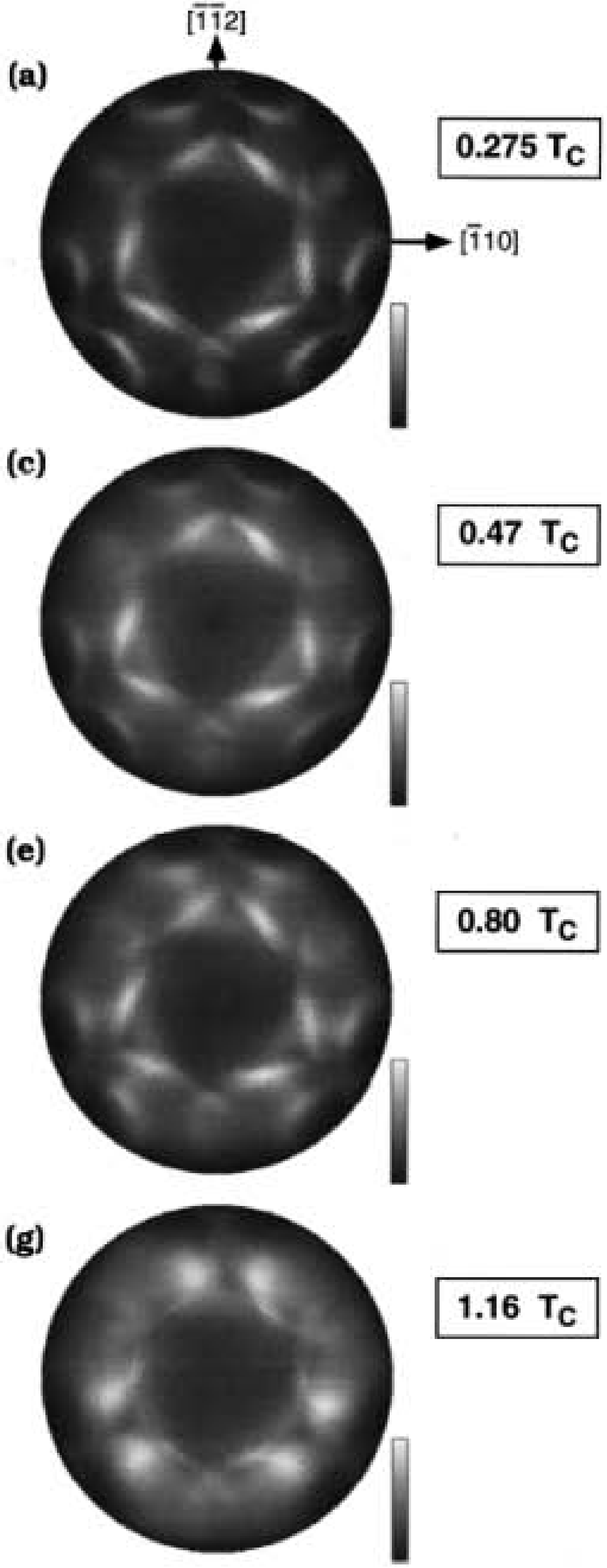
$T_c = 631$ K

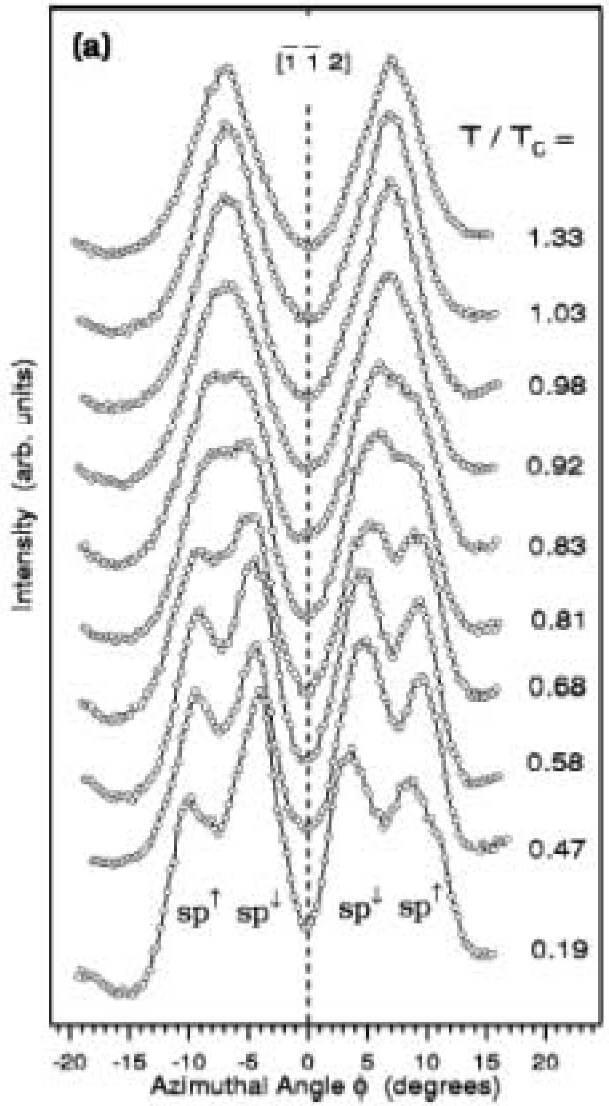
Question:
How does the band structure change when nickel goes from the ferromagnetic to the paramagnetic state ?

Answer:
Magnetic exchange splittings disappear

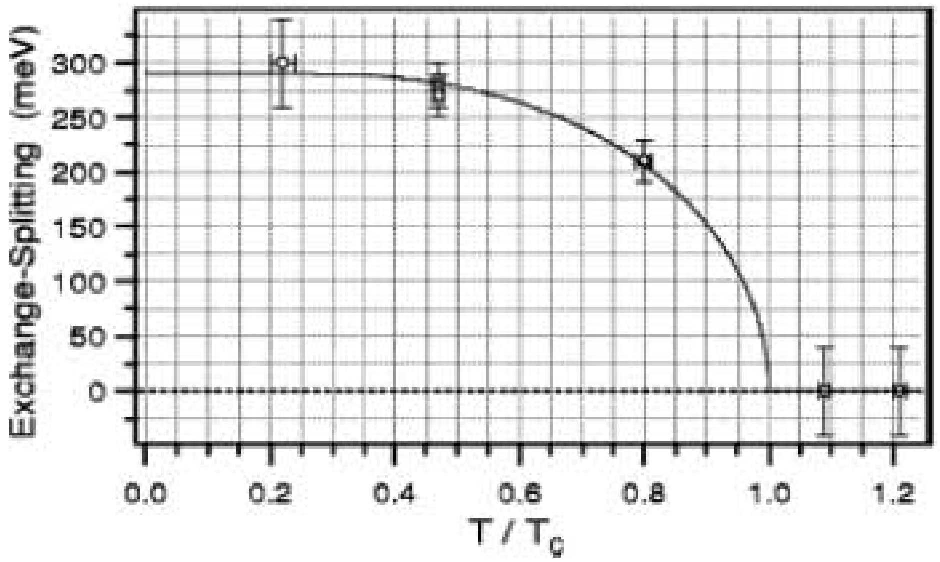


Evolution of the Fermi Surface During the Phase Transition

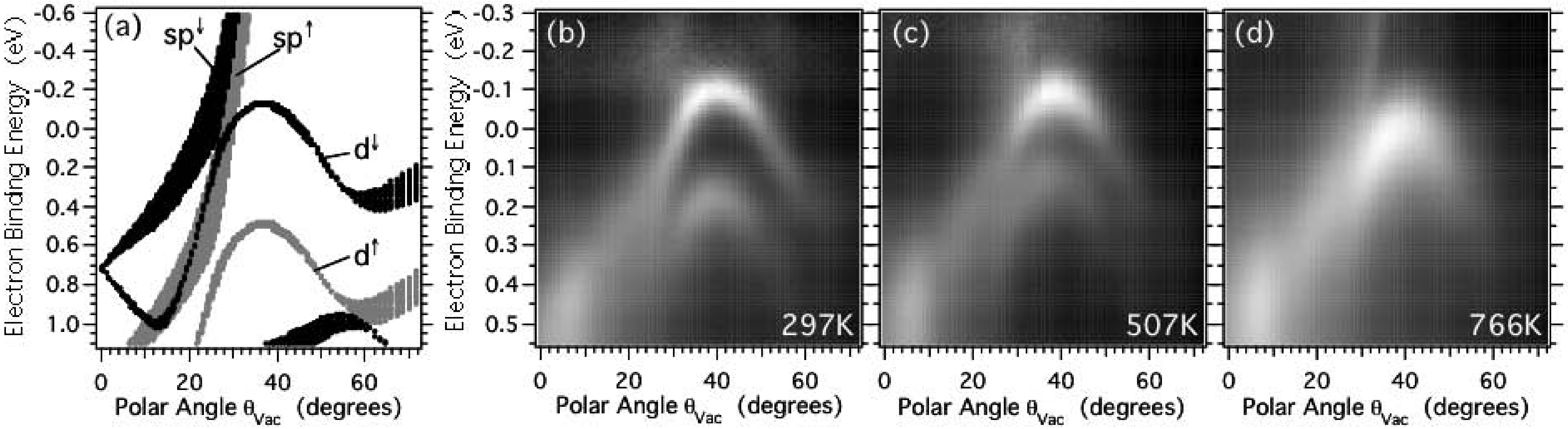




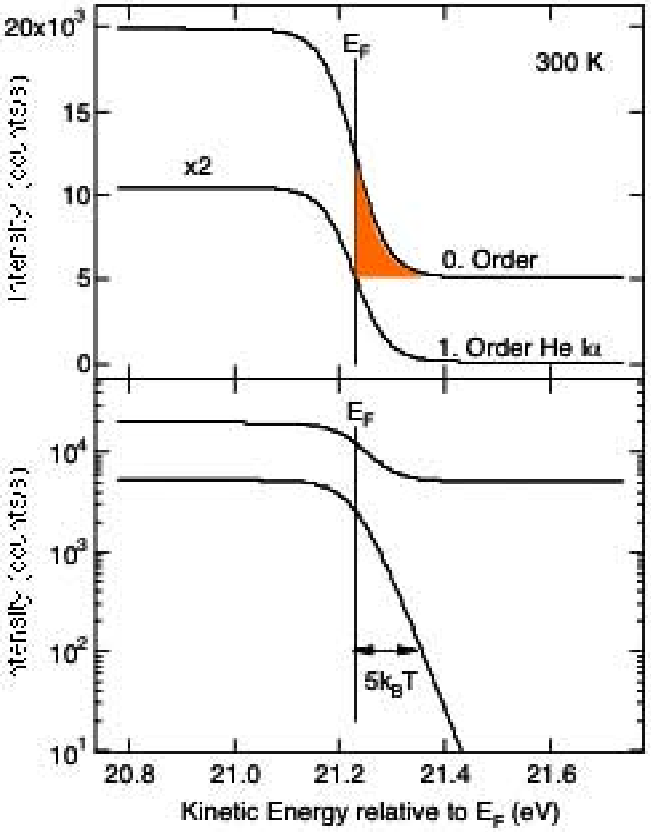
… and quantifying the exchange splitting:



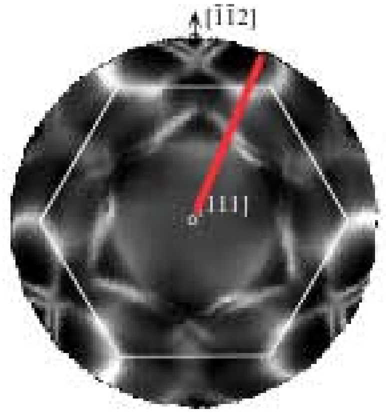
Observing the Magnetic Exchange Splitting in the d Band



Photoemission above the Fermi Level ?



Polar Scan

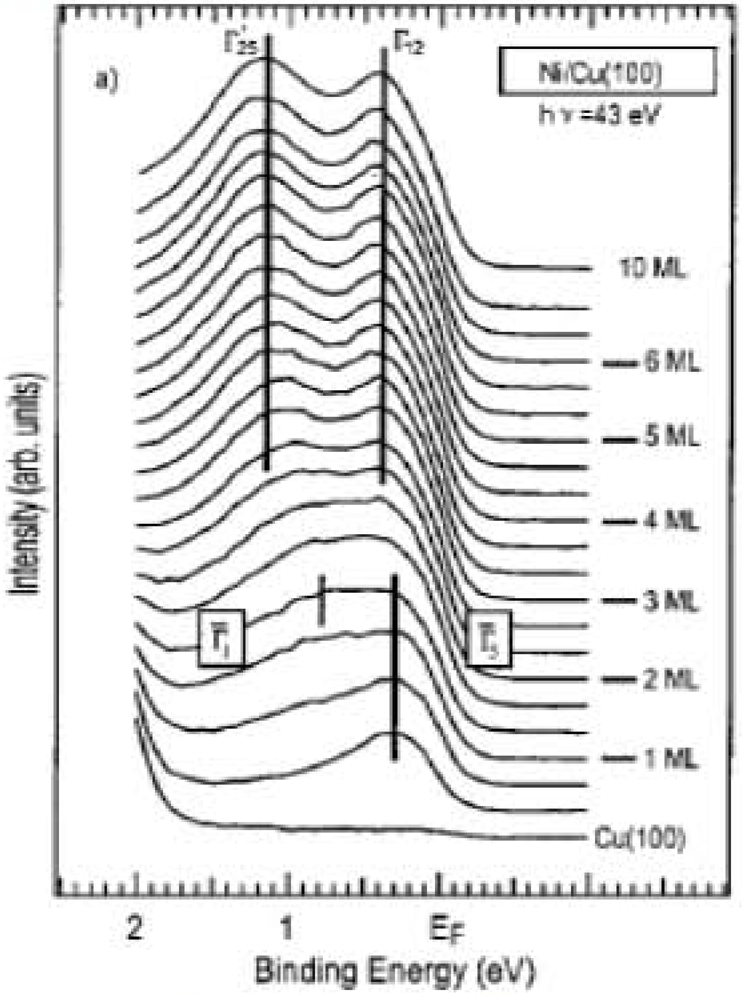


T. Greber et al., PRL 79, 4465 (1997)

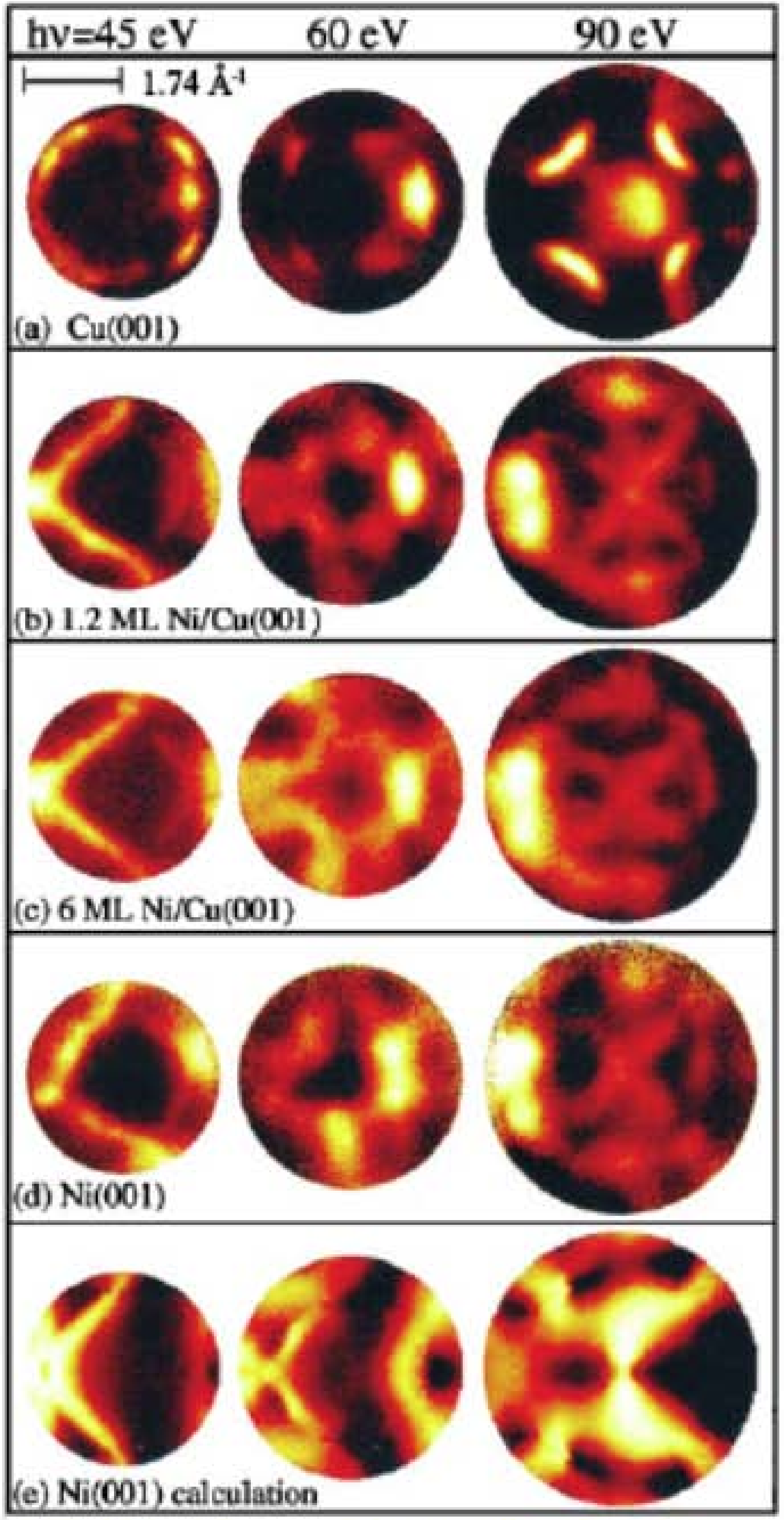
Ultrathin Films of Ni on Cu(001)

- grows epitaxially
- well ordered films, some roughness
- T_c depends on film thickness
- bulk-like Fermi surface already at 1 ML
- clear changes of the band width below 2.5 ML

?

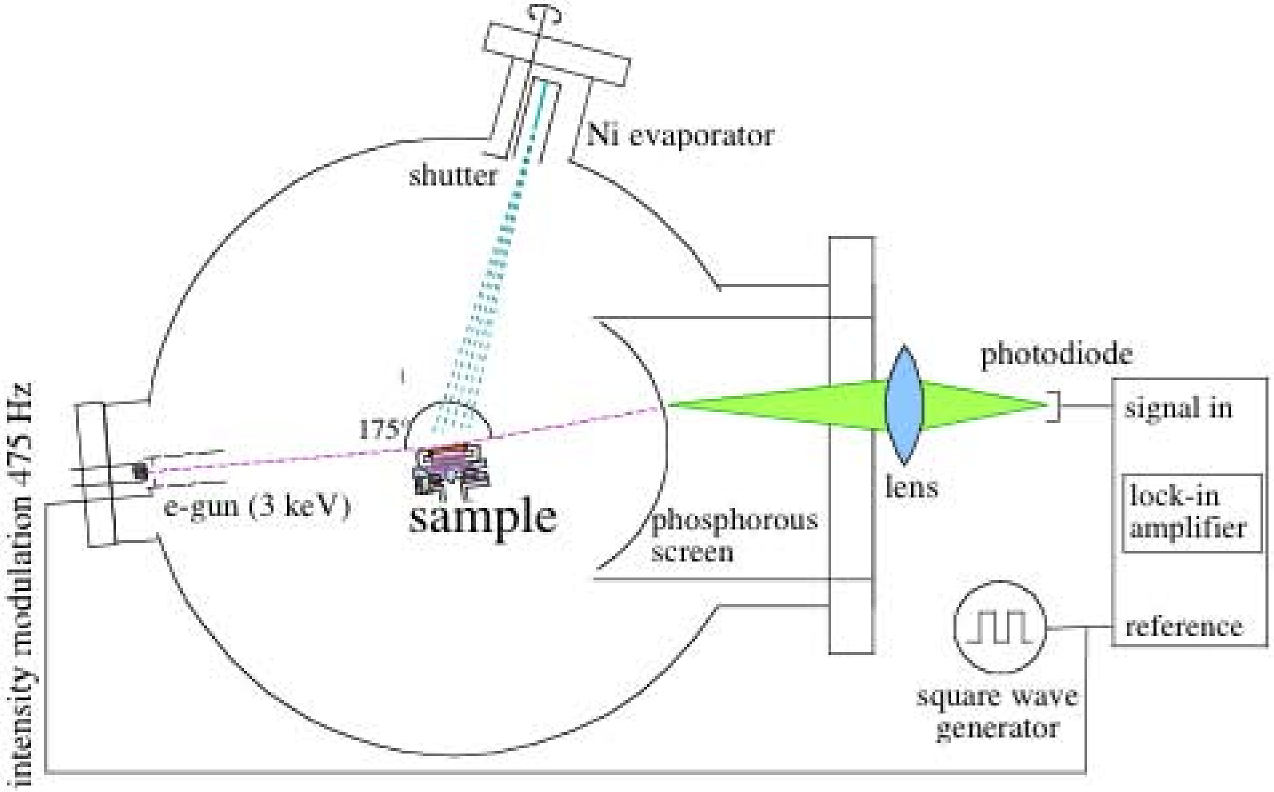


C. Pampuch et al.,
PRB 63, 153409 (2001).



G. J. Mankey et al., PRL 78, 1146 (1997).

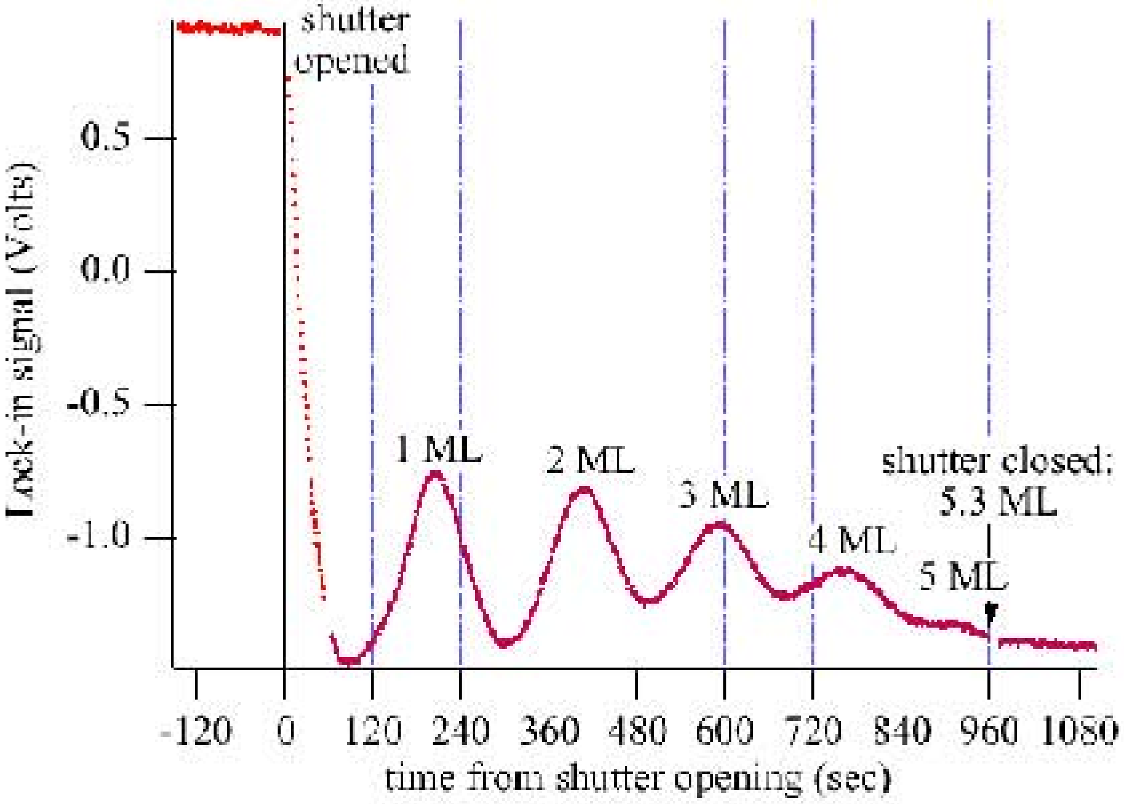
Preparation of Monolayer Films of Ni on Cu(001)



Ni deposited from e-beam heated rod at 0.33 ML/min.onto clean Cu(001)

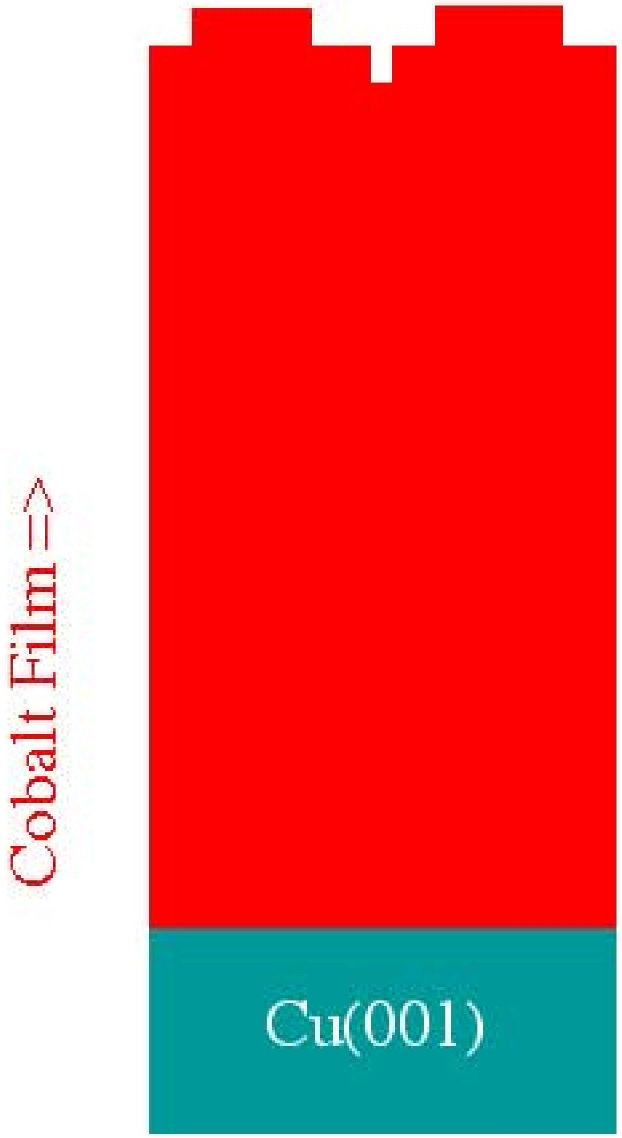
Annealing to 420 K.

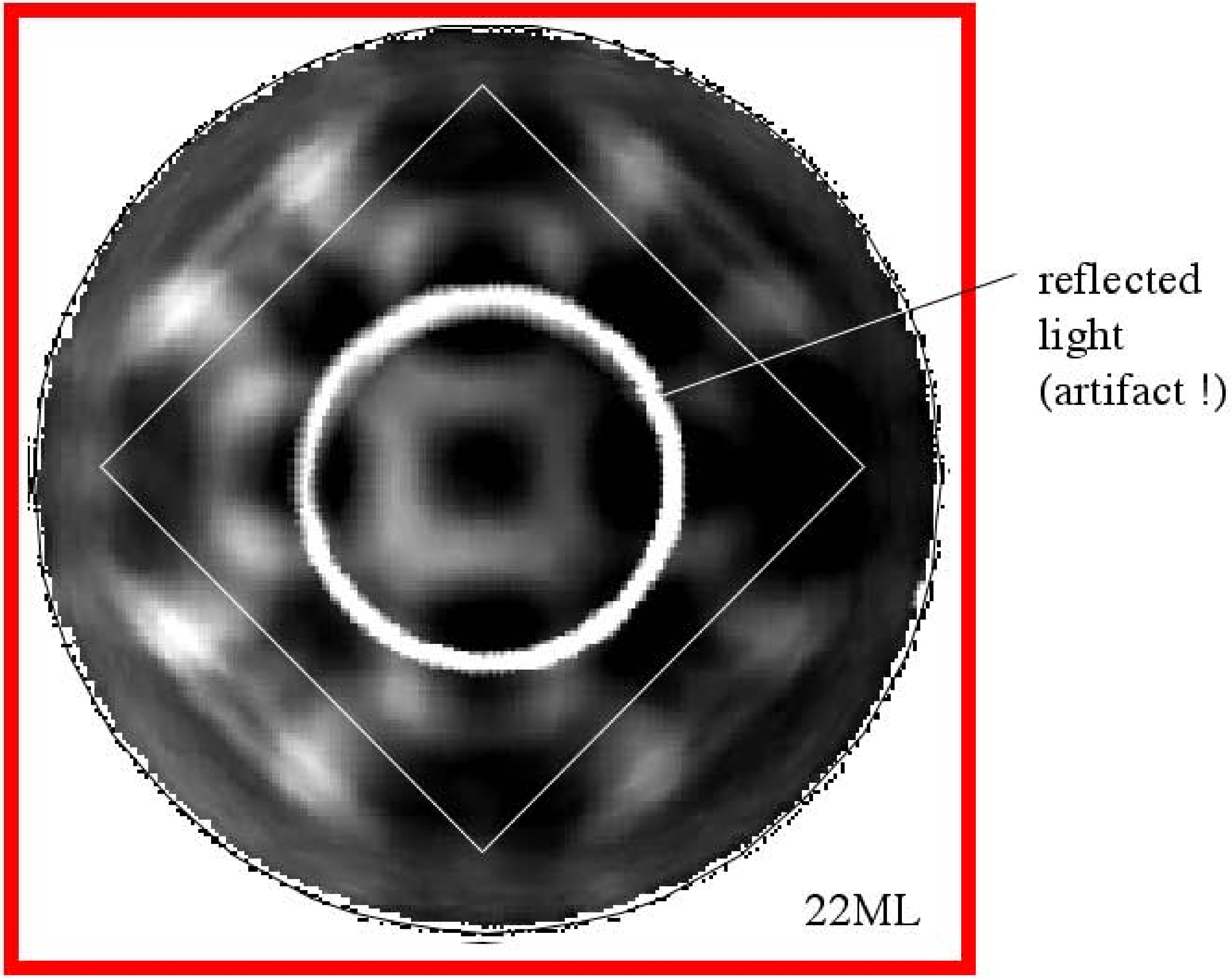
Cooling to 150 K (liquid N_2).



□ ARMCO magnet 🟧 Cu sample

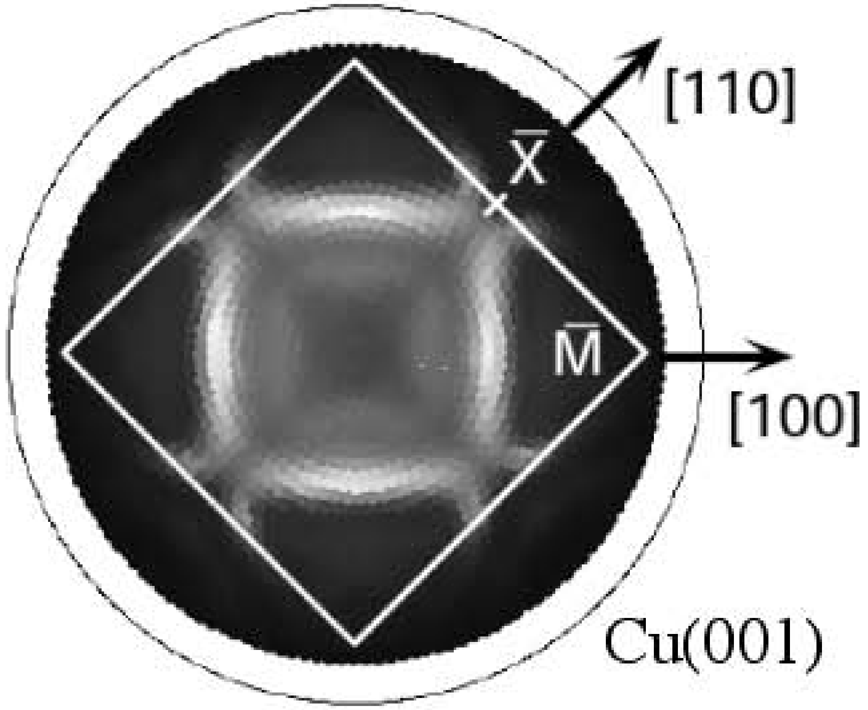
Evolution of the Fermi Surface with Film Thickness



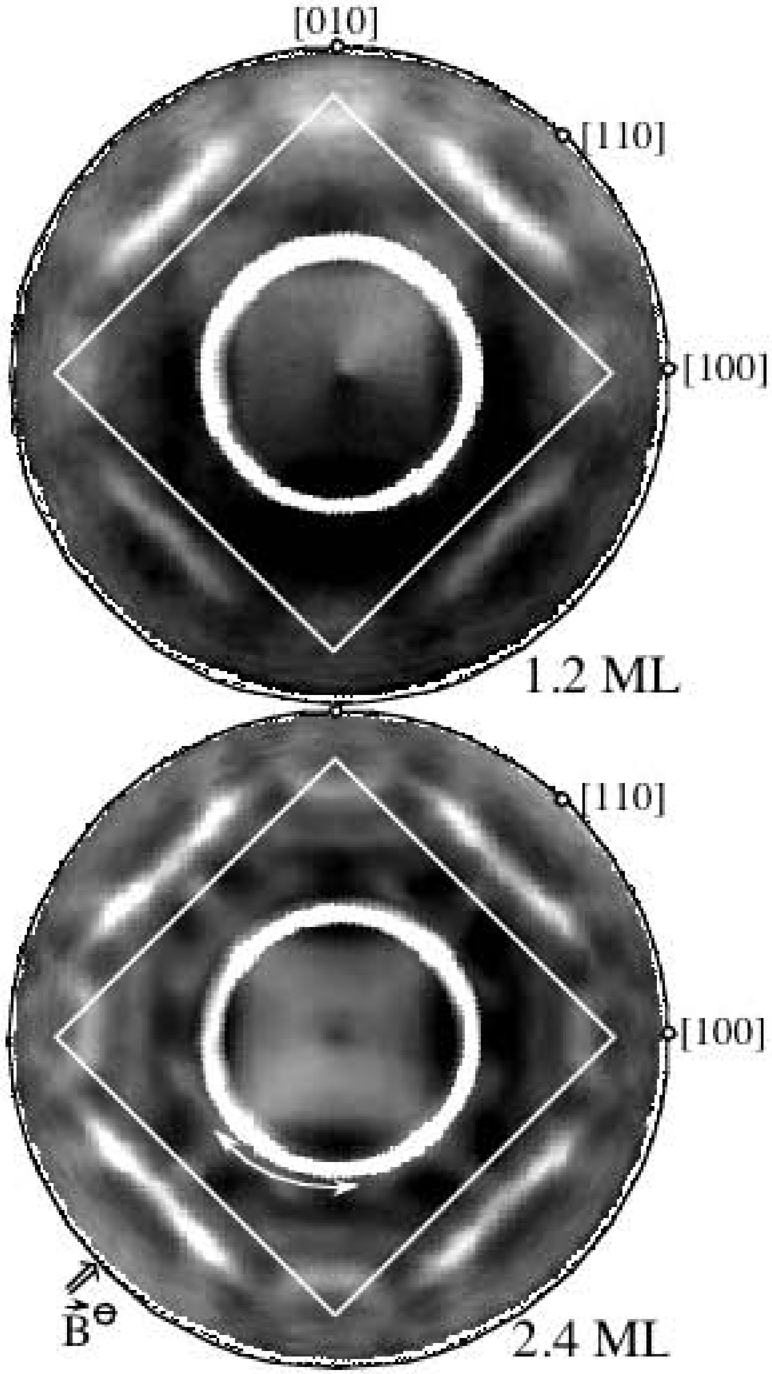


Contribution from the Cu(001) Fermi surface …

Substrate Fermi surface:



1.2 ML
Ni/Cu(001)
$T_C < 100$ K

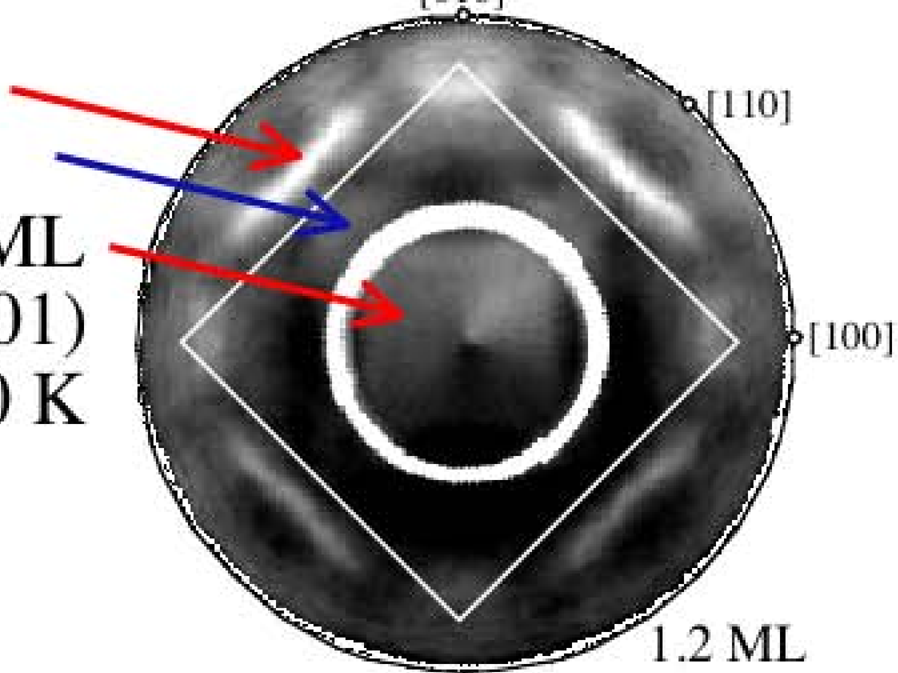


2.4 ML
Ni/Cu(001)
$T_C = 100$ K

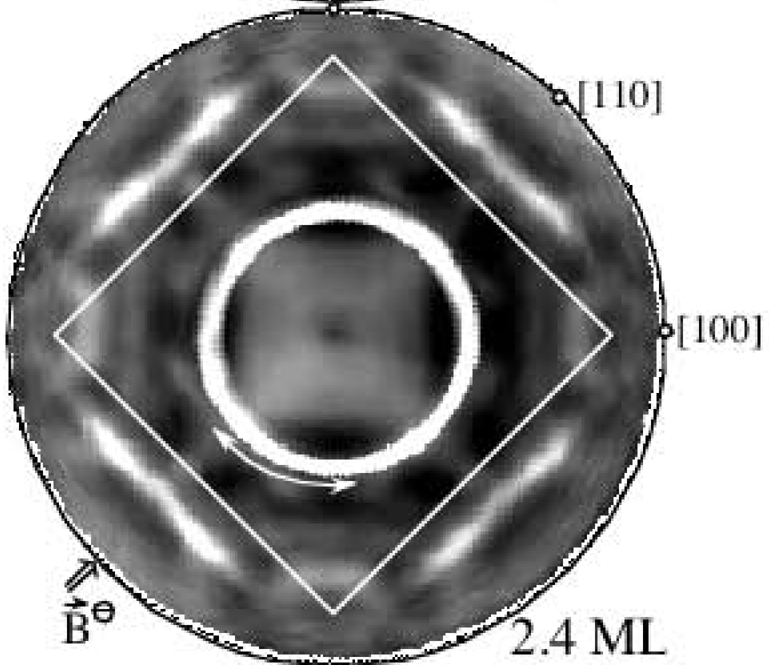
… is quenched from the first monolayer nickel !

(high cross section, strong inelastic scattering, …)

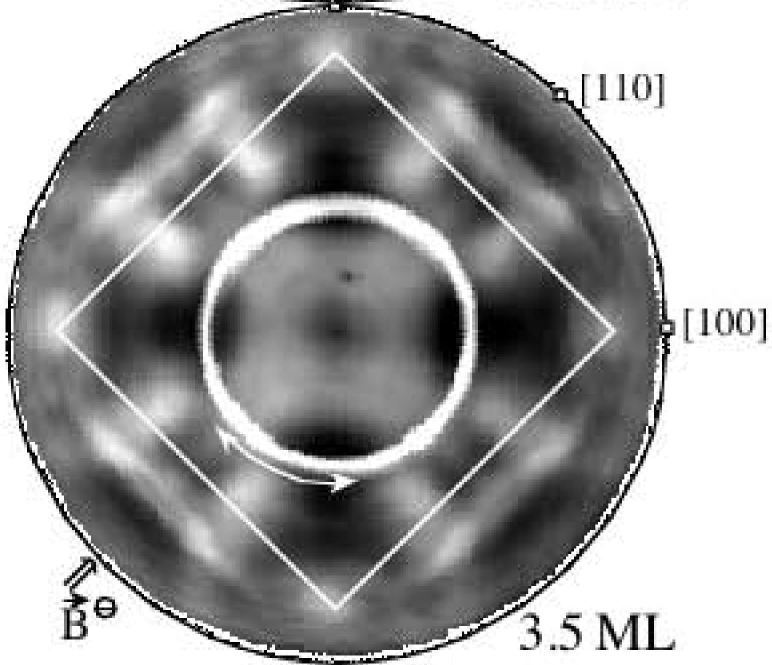
1.2 ML
Ni/Cu(001)
$T_C < 100$ K



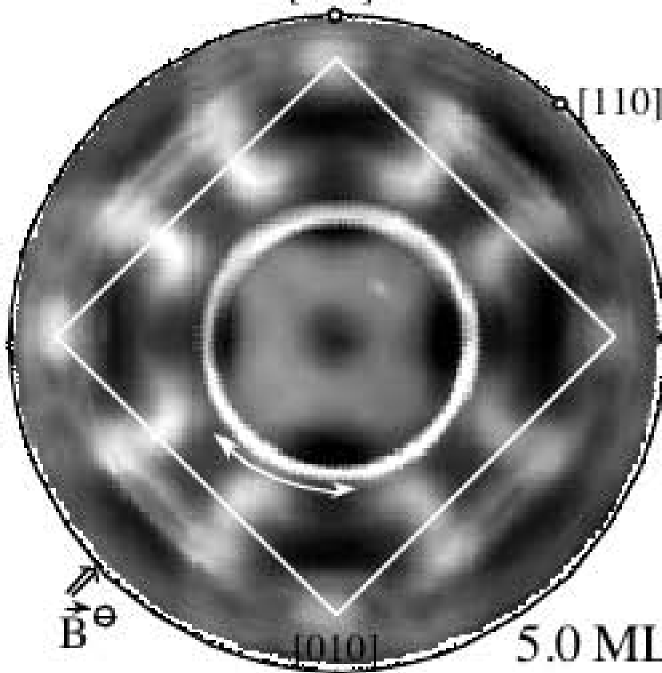
2.4 ML
Ni/Cu(001)
$T_C = 100$ K



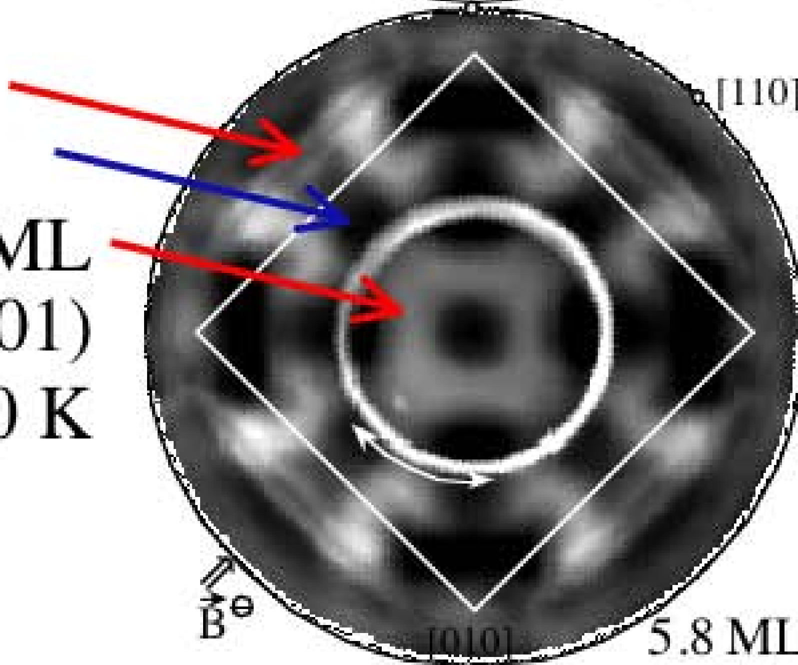
3.5 ML
Ni/Cu(001)
$T_C = 180$ K



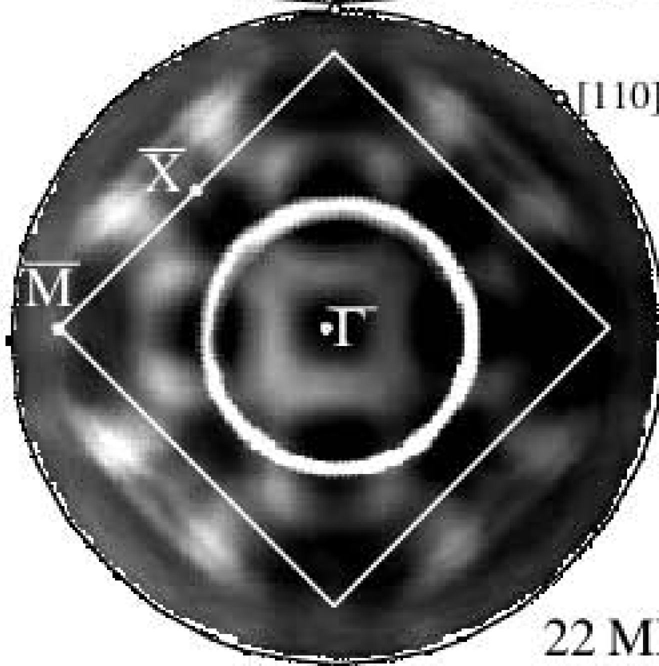
5.0 ML
Ni/Cu(001)
$T_C = 260$ K



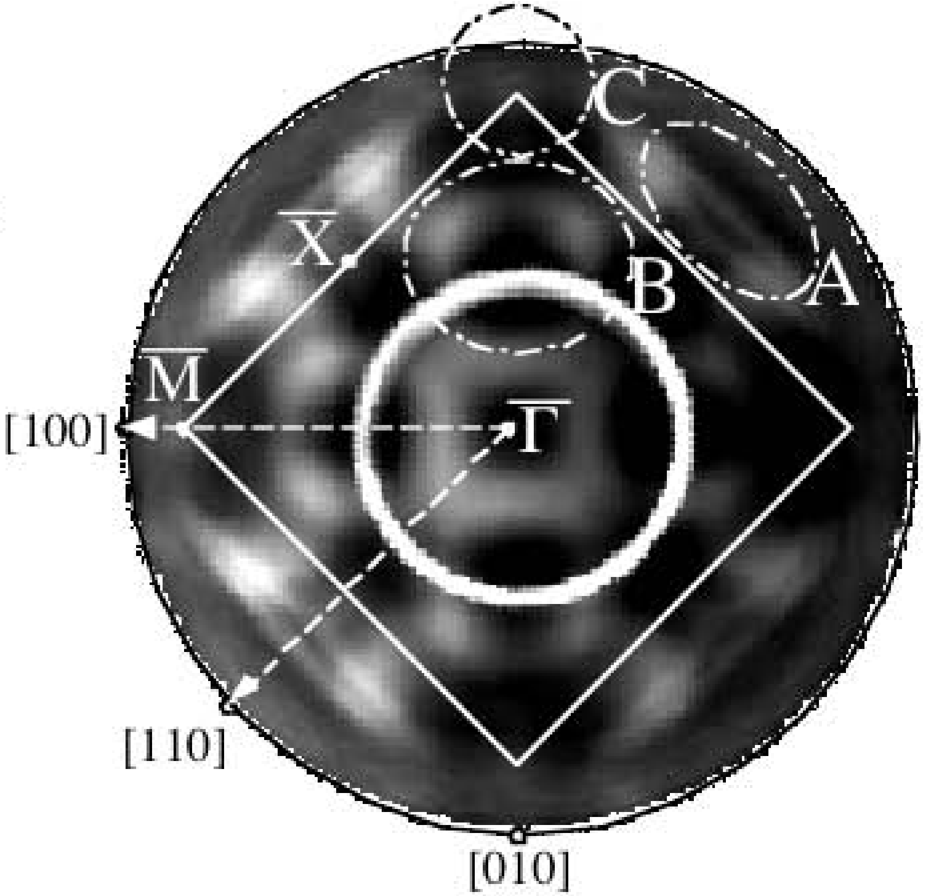
5.8 ML
Ni/Cu(001)
$T_C = 330$ K



22 ML
Ni/Cu(001)
$T_C = 627$ K
equivalent
bulk Ni(001)



Band Character of Fermi Surface Contours (thick film ~ bulk-like)



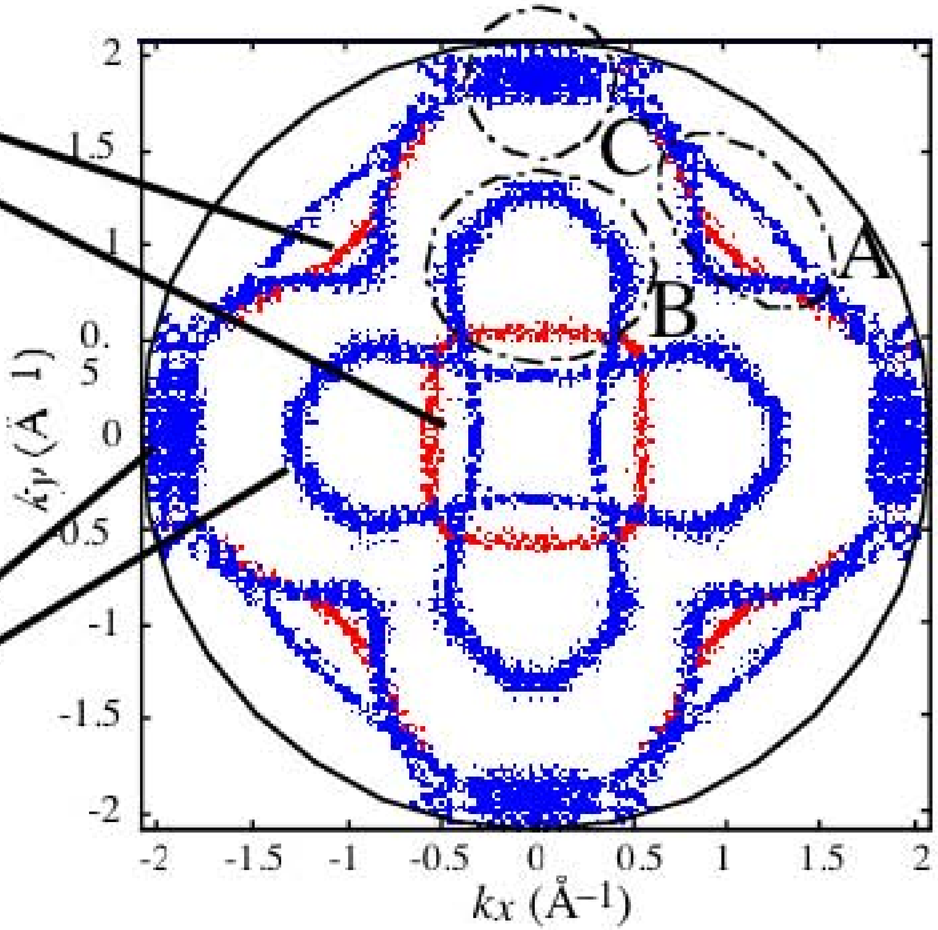
data from
22 ML
Ni/Cu(001)

$h\nu$ =21.22 eV

equivalent
bulk Ni(001)

sp band

d band

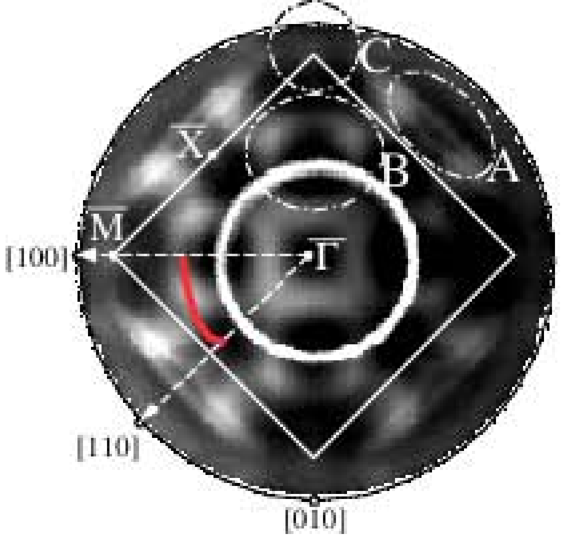


Fermi surface
calculation

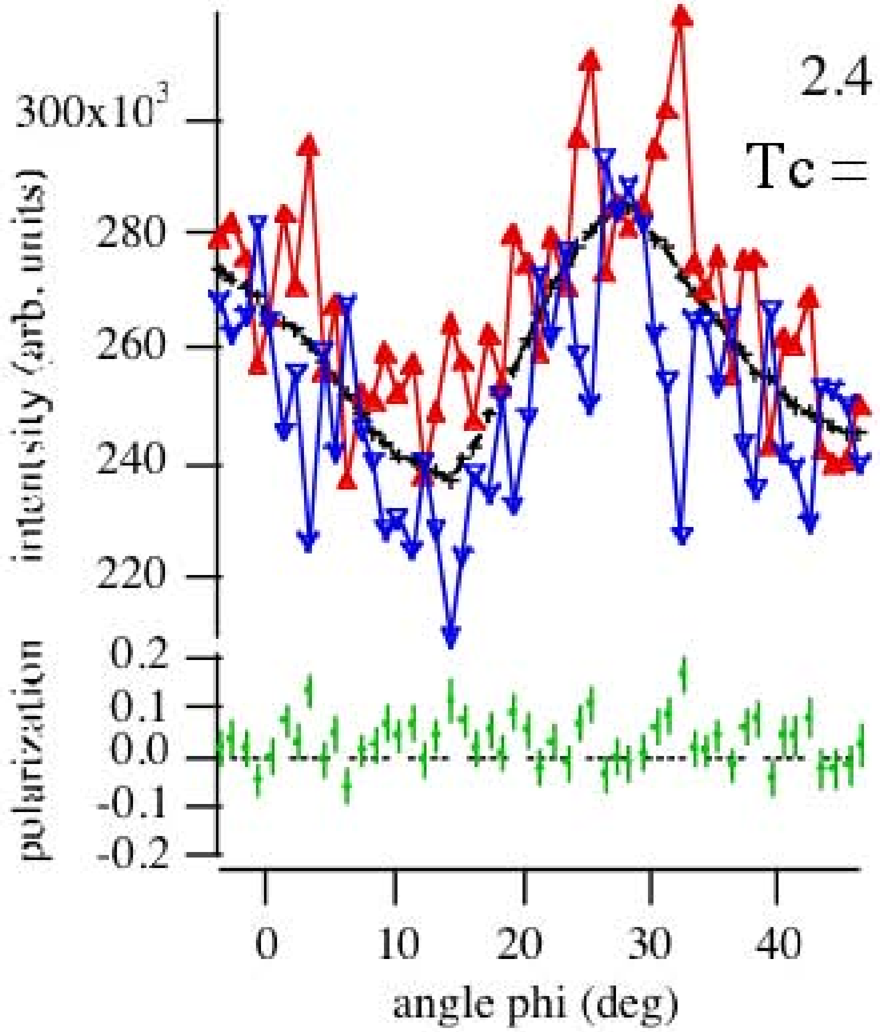
free electron
final state
approximation

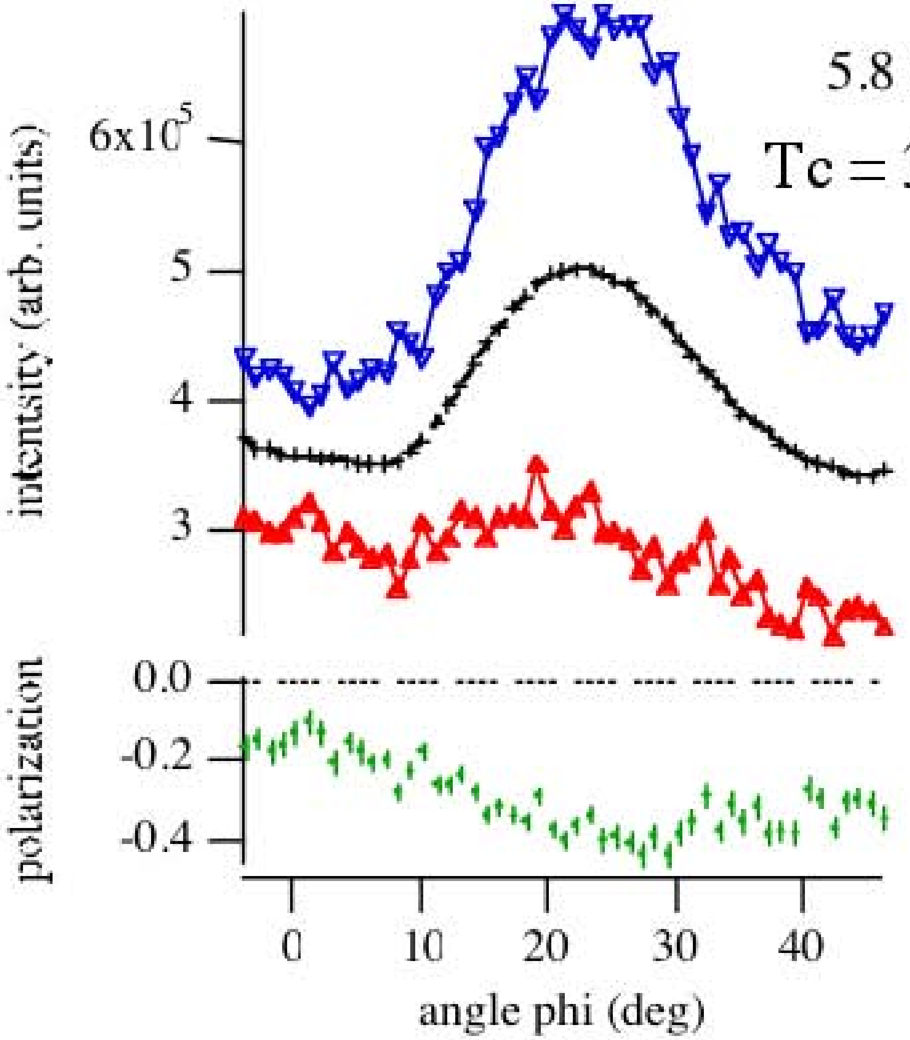
$h\nu$ = 21.22 eV
Φ = 5.2 eV
V_0 = 10.2 eV

Use Spin Polarization for Band Identification



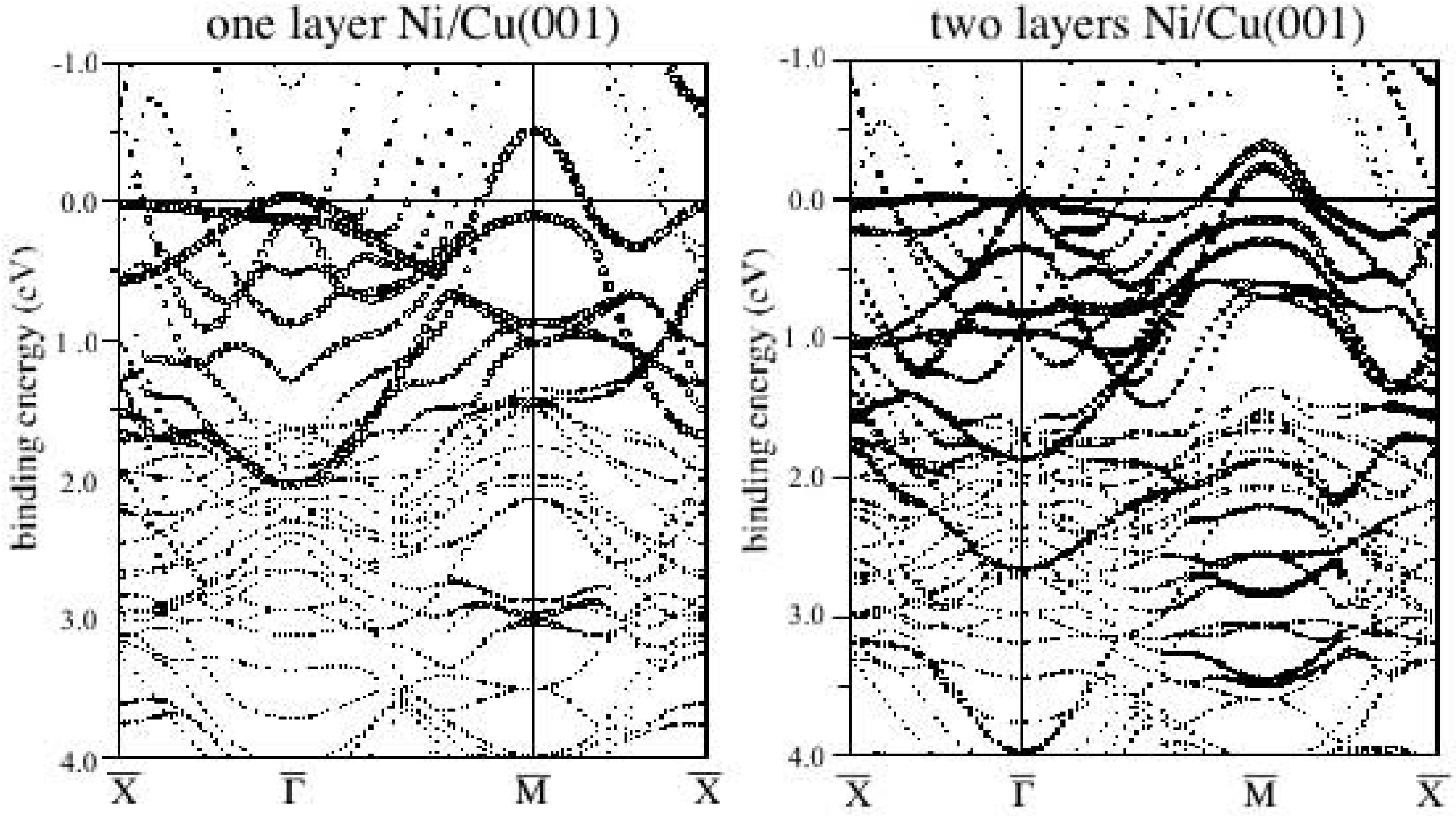
Azimuthal Momentum Scans at θ = 28°
(hν = 21.22 eV)
T = 150 K

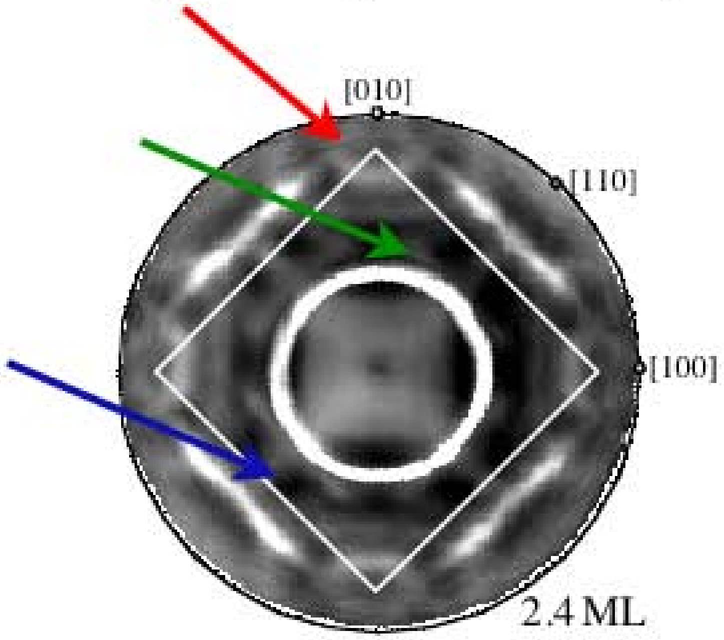




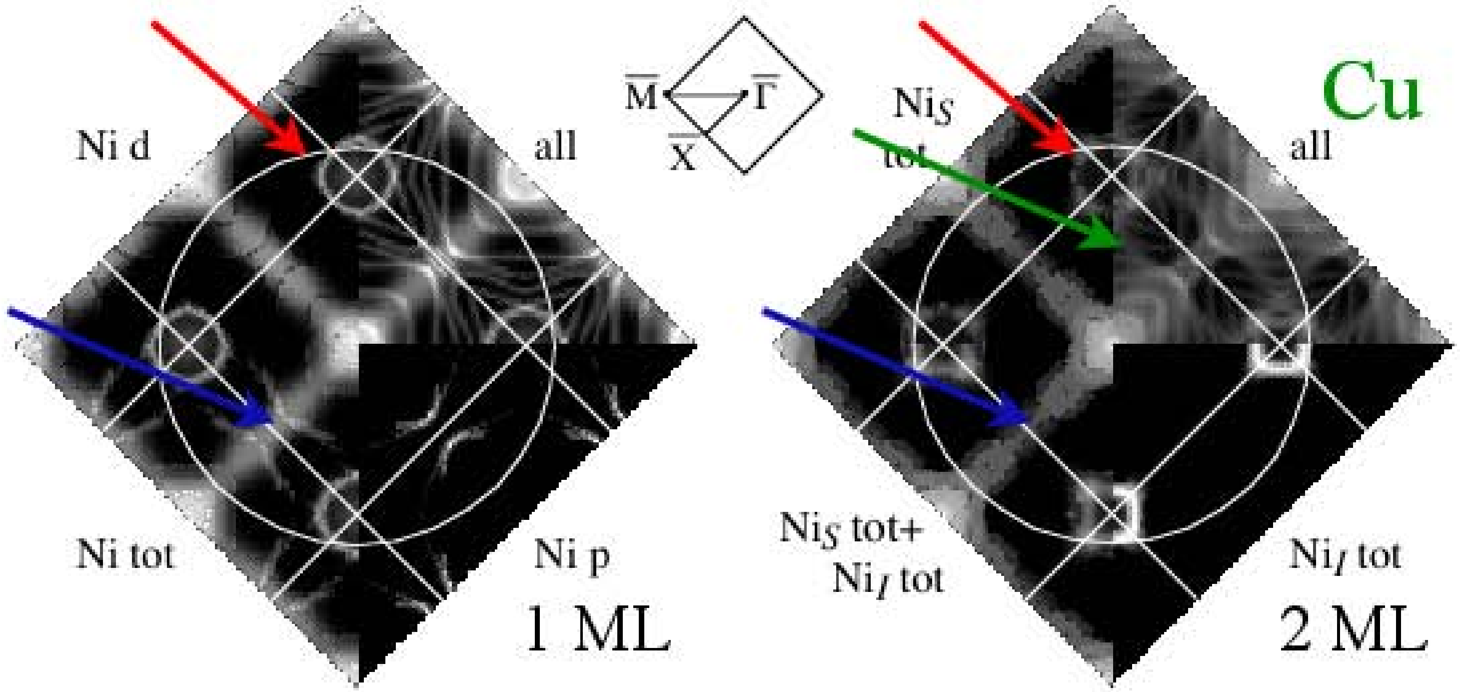
The ferromagnetic film (5.8 ML) shows strong minority spin polarization => minority d band

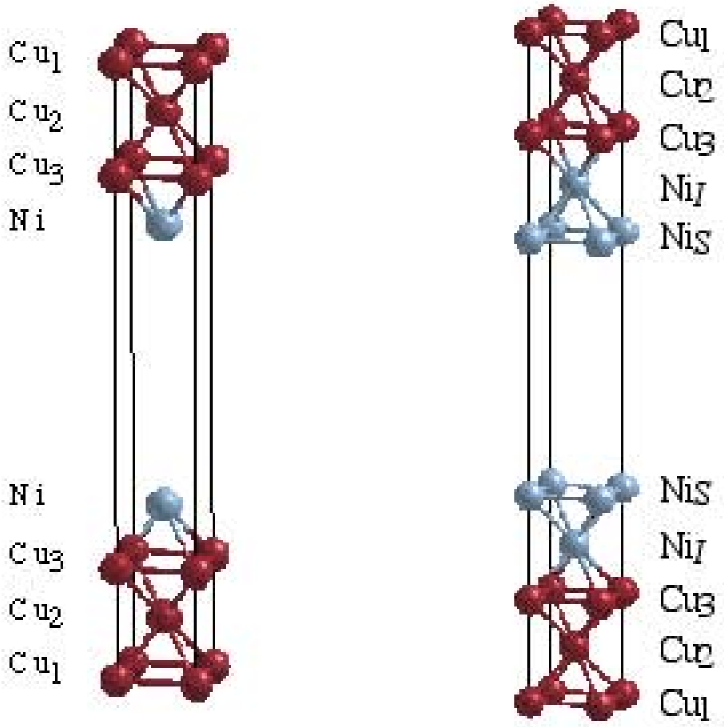
Ni/Cu(001) Slab Calculations for Monolayer Regime (Wien97)





data from
2.4 ML Ni/Cu(001)
$h\nu = 21.22$ eV

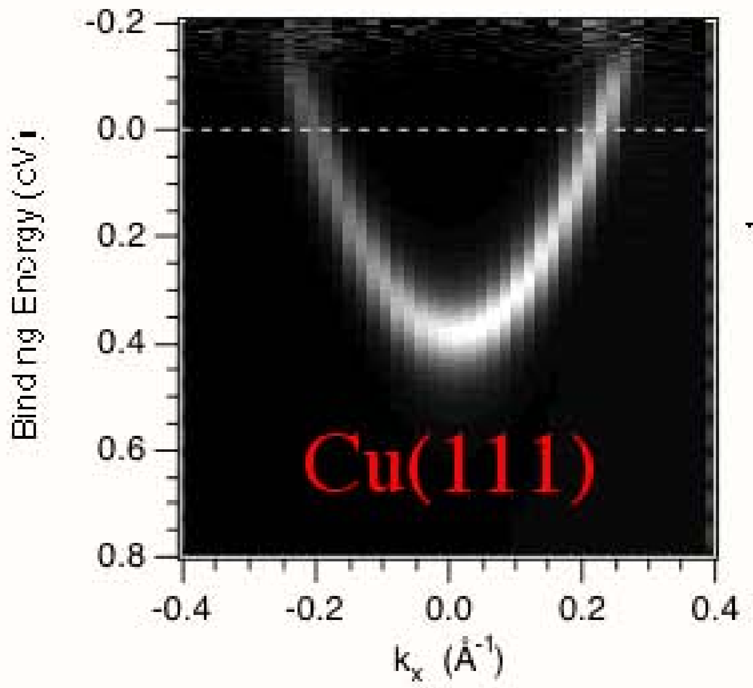




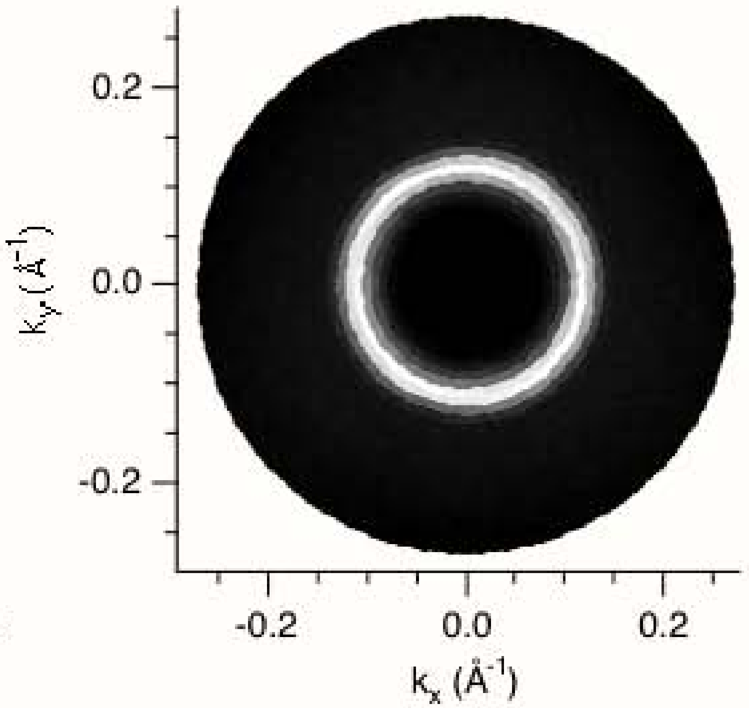
Ultrathin films of Ni on Cu(001)

- < 3 ML: paramagnetic interface band structure
- Around 3 ML two things happen:
  - $T_c > T_{meas}$ => Exchange splitting appears
  - Band structure becomes bulk-like (3D)
- The sp bands are bulk-like already at ~1 ML
- The d bands form interface states for < 3 ML

The Shockley Surface State on Noble Metal (111) Surfaces

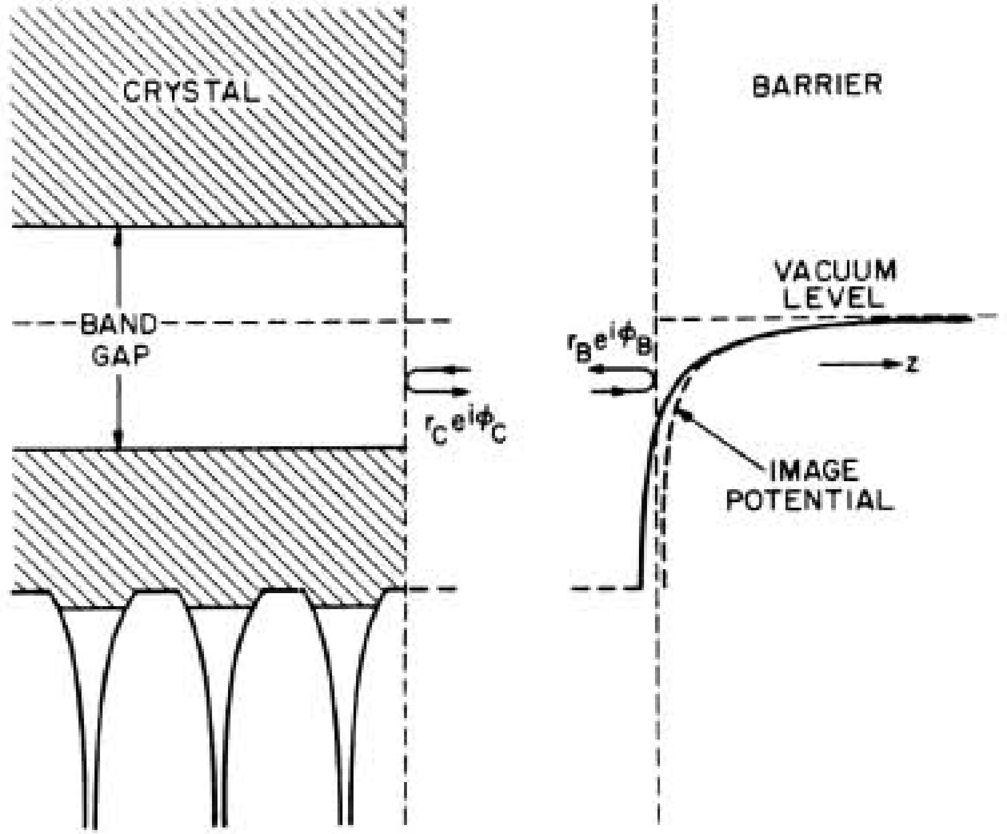


Energy dispersion

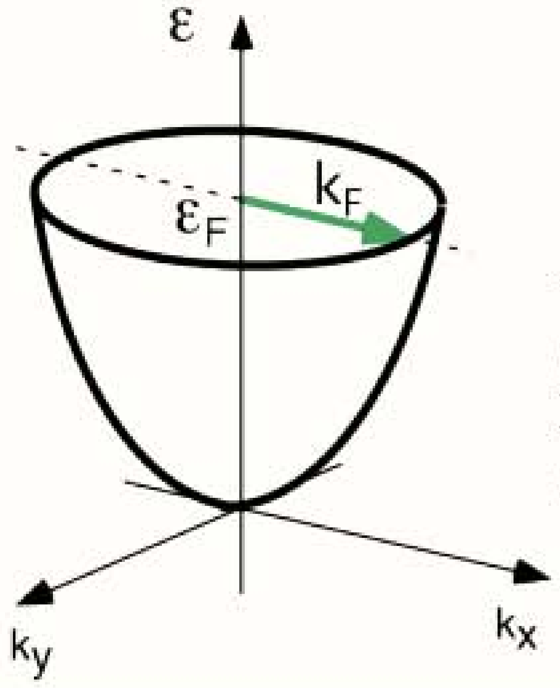


Fermi surface

Multiple reflection model for surface states:



(N.V. Smith, PRB **32**, 3549 (1985))



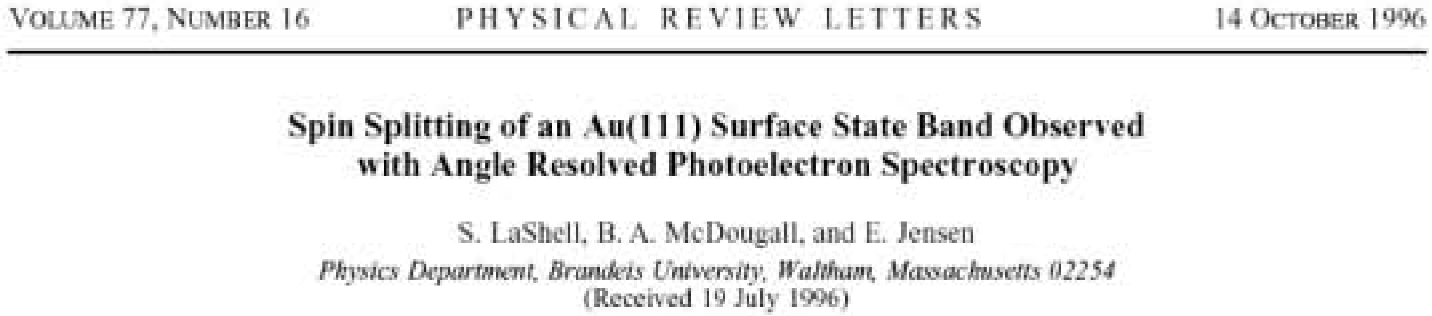
2D free electron gas

binding energy:	390 meV
effective mass:	0.42 m_e
Fermi wave length:	2.9 nm
coherence length:	~ 10 nm (300K)

Courtesy F. Baumberger

What does this have to do with spin and magnetism ?

The Spin-Orbit Split Surface State on Au(111)

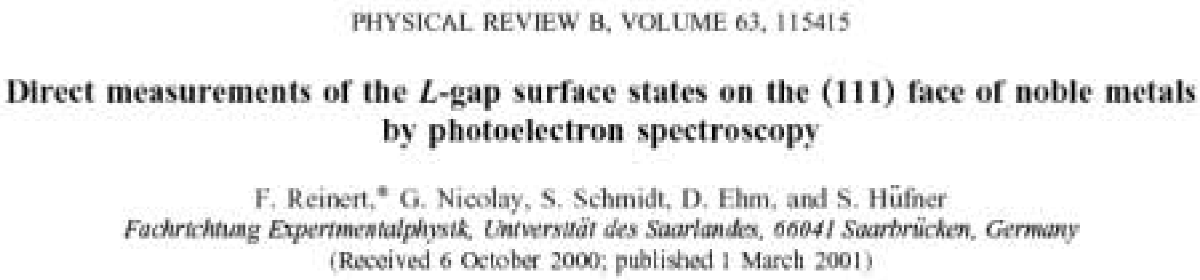


VOLUME 77, NUMBER 16 PHYSICAL REVIEW LETTERS 14 OCTOBER 1996

Spin Splitting of an Au(111) Surface State Band Observed with Angle Resolved Photoelectron Spectroscopy

S. LaShell, B. A. McDougall, and E. Jensen
Physics Department, Brandeis University, Waltham, Massachusetts 02254

(Received 19 July 1996)

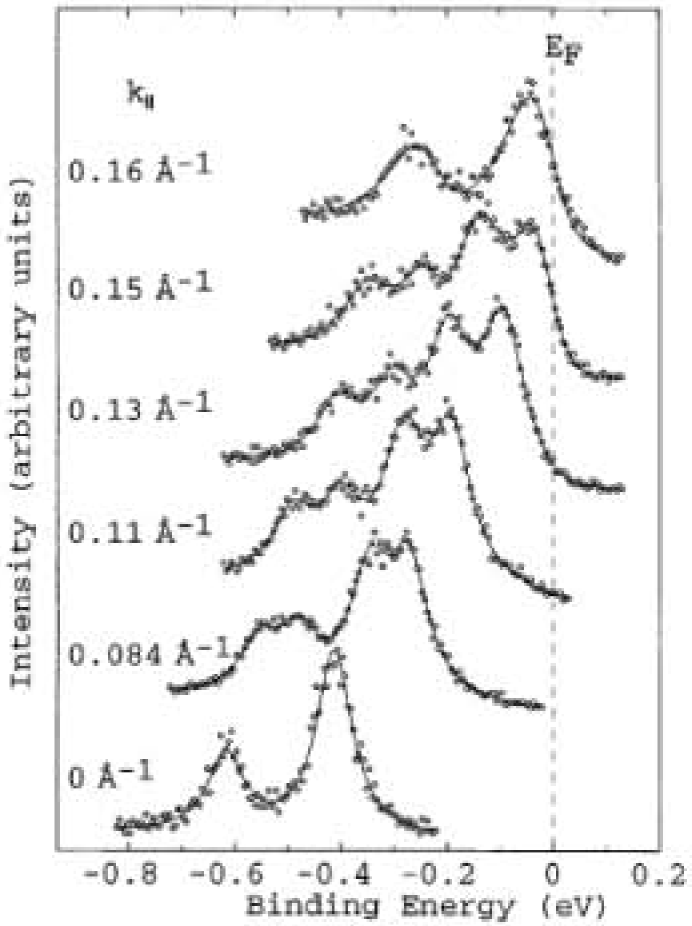


PHYSICAL REVIEW B, VOLUME 63, 115415

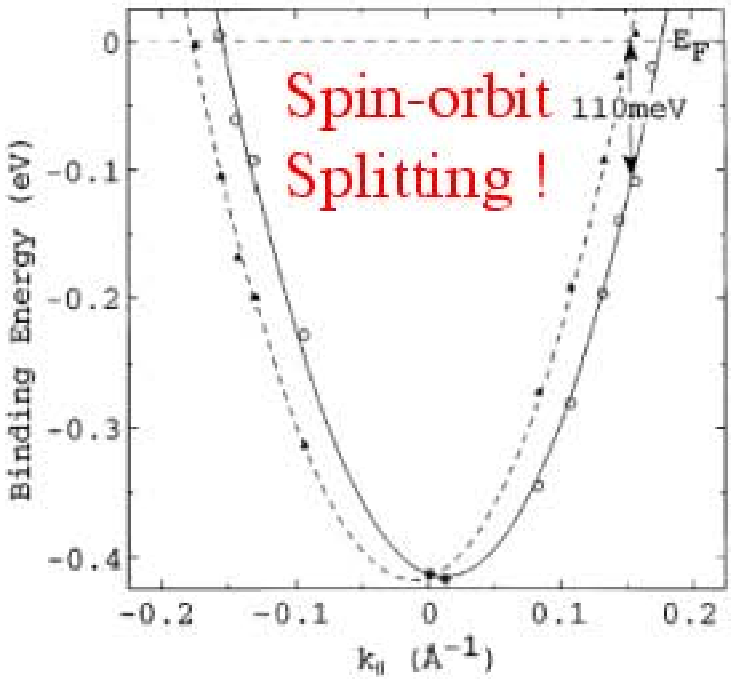
Direct measurements of the *L*-gap surface states on the (111) face of noble metals by photoelectron spectroscopy

F. Reinert,* G. Nicolay, S. Schmidt, D. Ehm, and S. Hüfner
Fachrichtung Experimentalphysik, Universität des Saarlandes, 66041 Saarbrücken, Germany

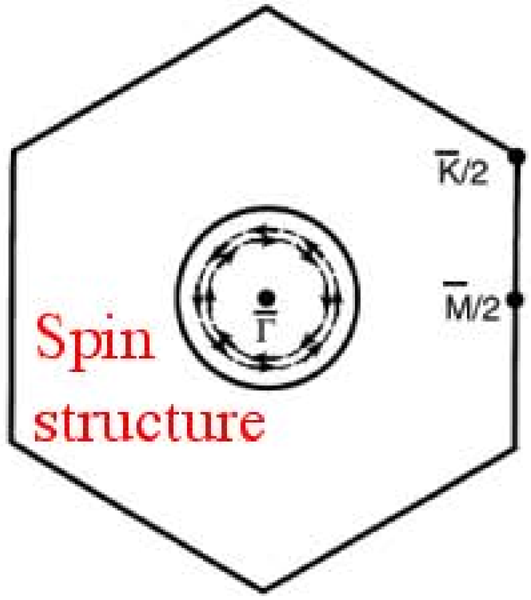
(Received 6 October 2000; published 1 March 2001)



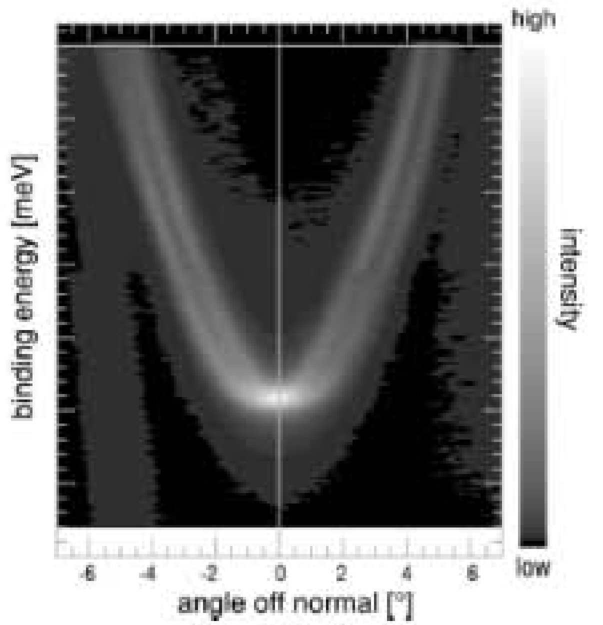
hν=11.62eV

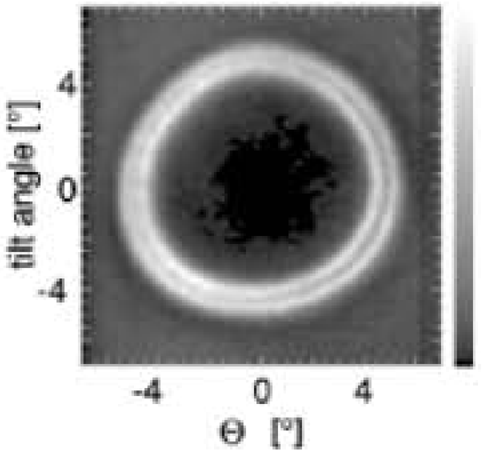


Spin-orbit Splitting !



Spin structure





Scienta 2002 spectrometer

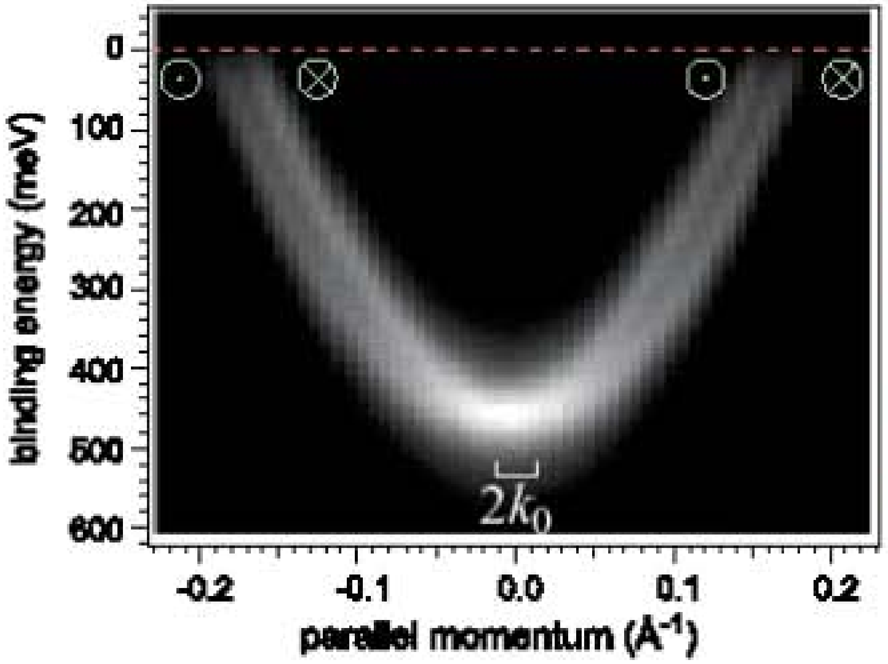
hν=21.21eV

=> spin-resolution is a challenging task !

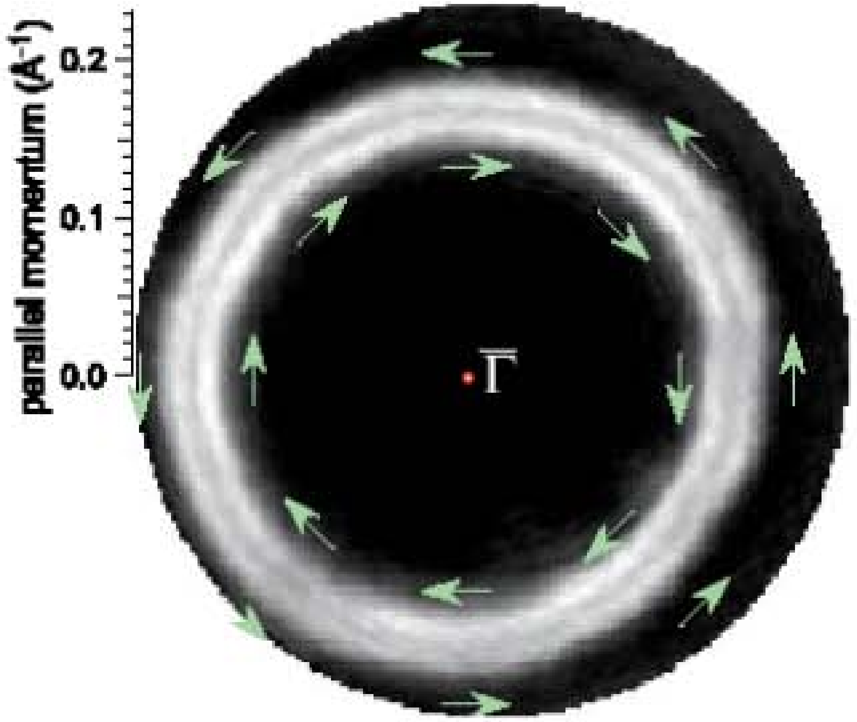
The Shockley surface state on Au(111)

spin-integrated photoemission at $h\nu$ = 21.1 eV, T = 160 K

instrumental resolution
ΔE = 25 meV, $\Delta\vartheta$ = 0.5° (FWHM)



dispersion map



Fermi surface map

$2k_0$ = 0.026 Å^{-1} E_B = 470 meV k_F = 0.173 Å^{-1} ± k_0 m^* = 0.24 m_e

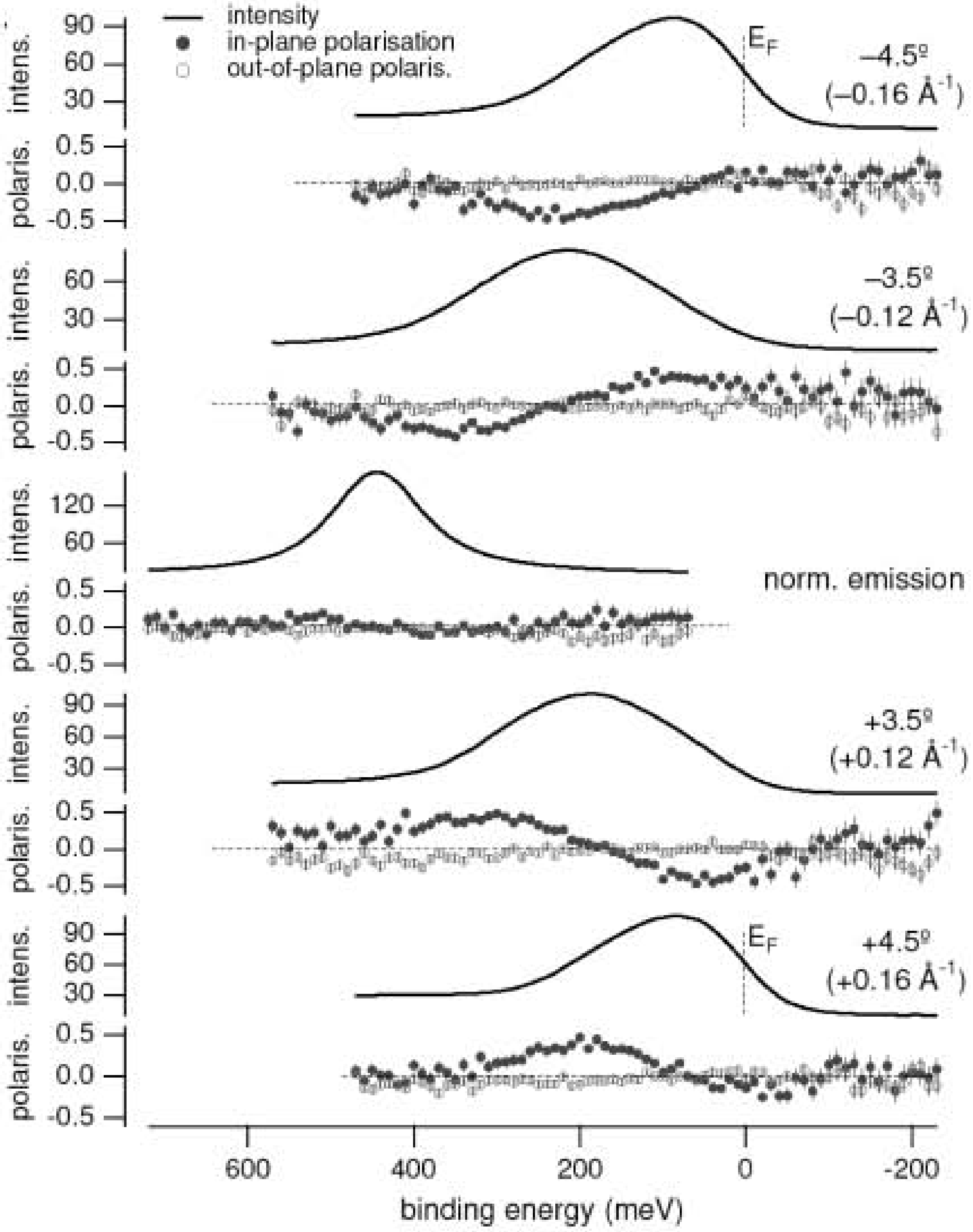
Theory: spin-orbit coupling

$$H_{SOC} = \frac{\mu_B}{2c^2}(\vec{v} \times \vec{E}) \cdot \vec{\sigma} \qquad E^{\uparrow\downarrow}(k) = E_0 + \frac{(k \pm k_0)^2}{2m^*}$$

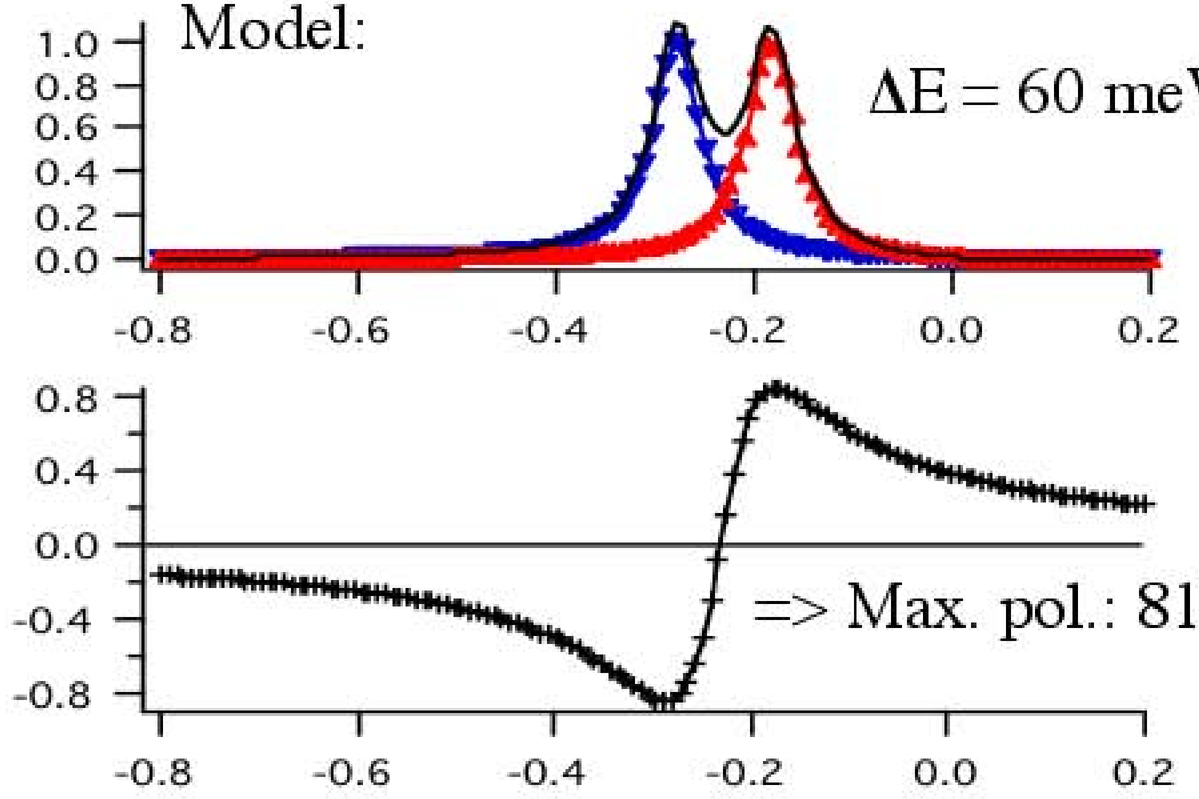
⇒ COPHEE can resolve the spin-orbit splitting in spin-integraded mode of operation.

Quantitative Spin Polarimetry on the Au(111) Surface State

Experiment:



Model:



ΔE = 60 meV

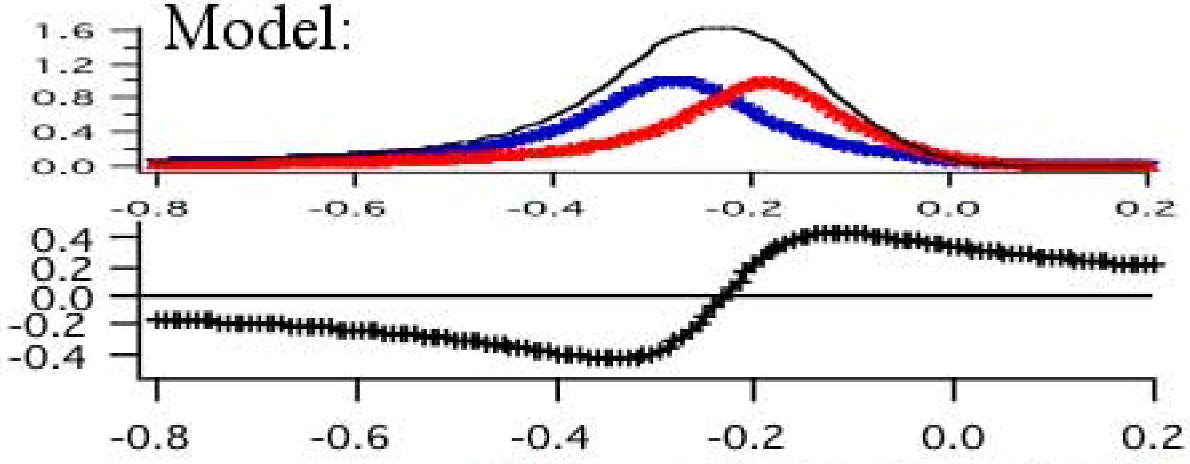
=> Max. pol.: 81 %

Spin-resolved experiment:

Energy resolution = 120 meV

Angular resolution = 1.2°

Model:



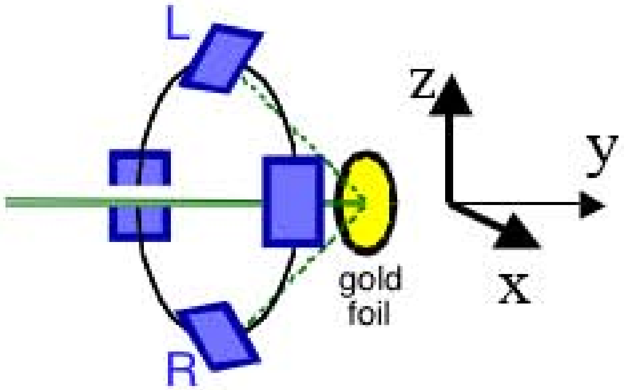
=> Max. polarization: 40 %

… and Recovering the Spin-Resolved Spectra

Here, sample is not magnetized !
=> Cross asymmetries cannot me obtained !

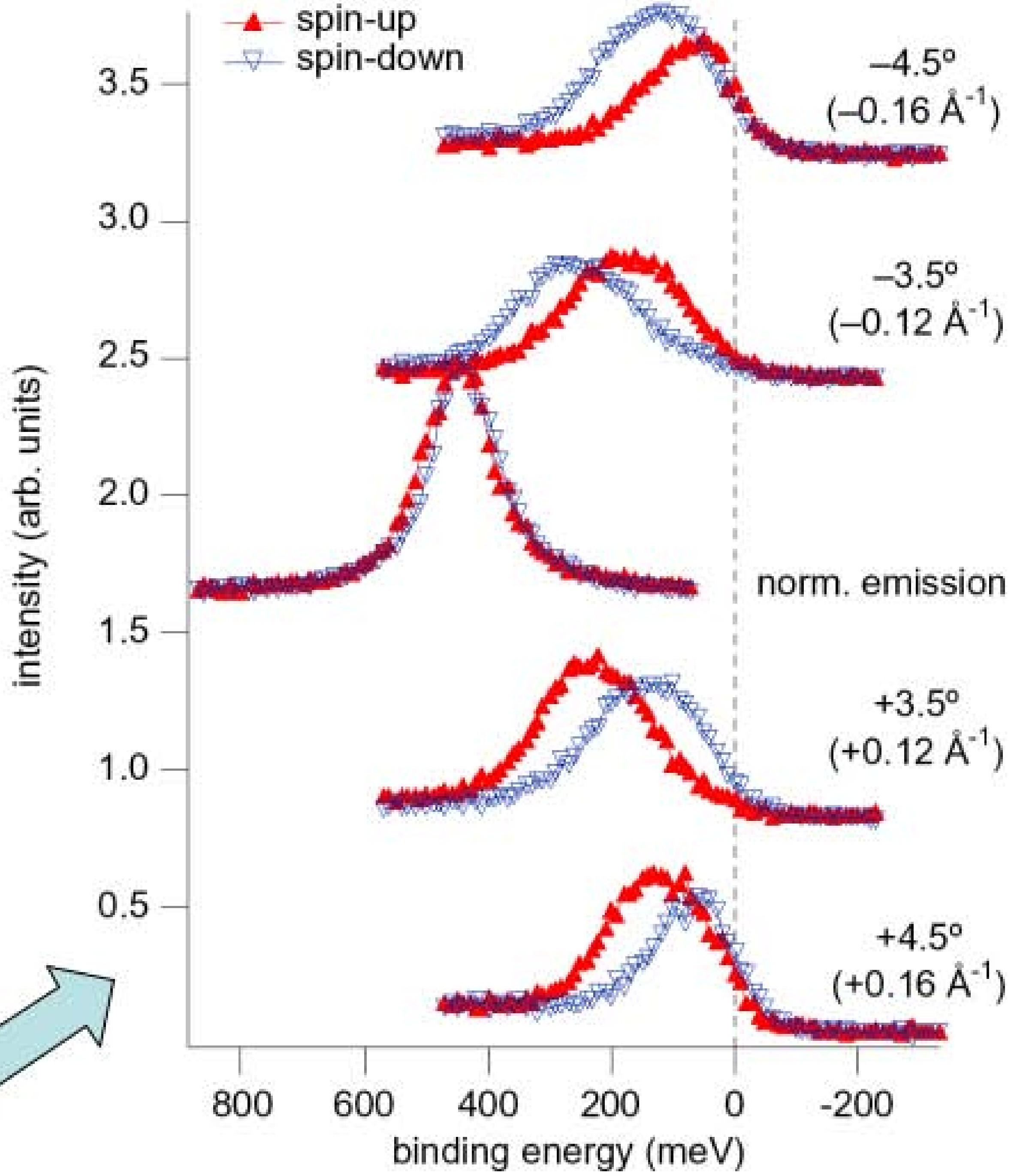
$$P = \frac{1}{S_{eff}} \frac{(I_L - \eta I_R)}{(I_L + \eta I_R)}$$

Use empirical sensitivity factors η
to remove instrumental asymmetries

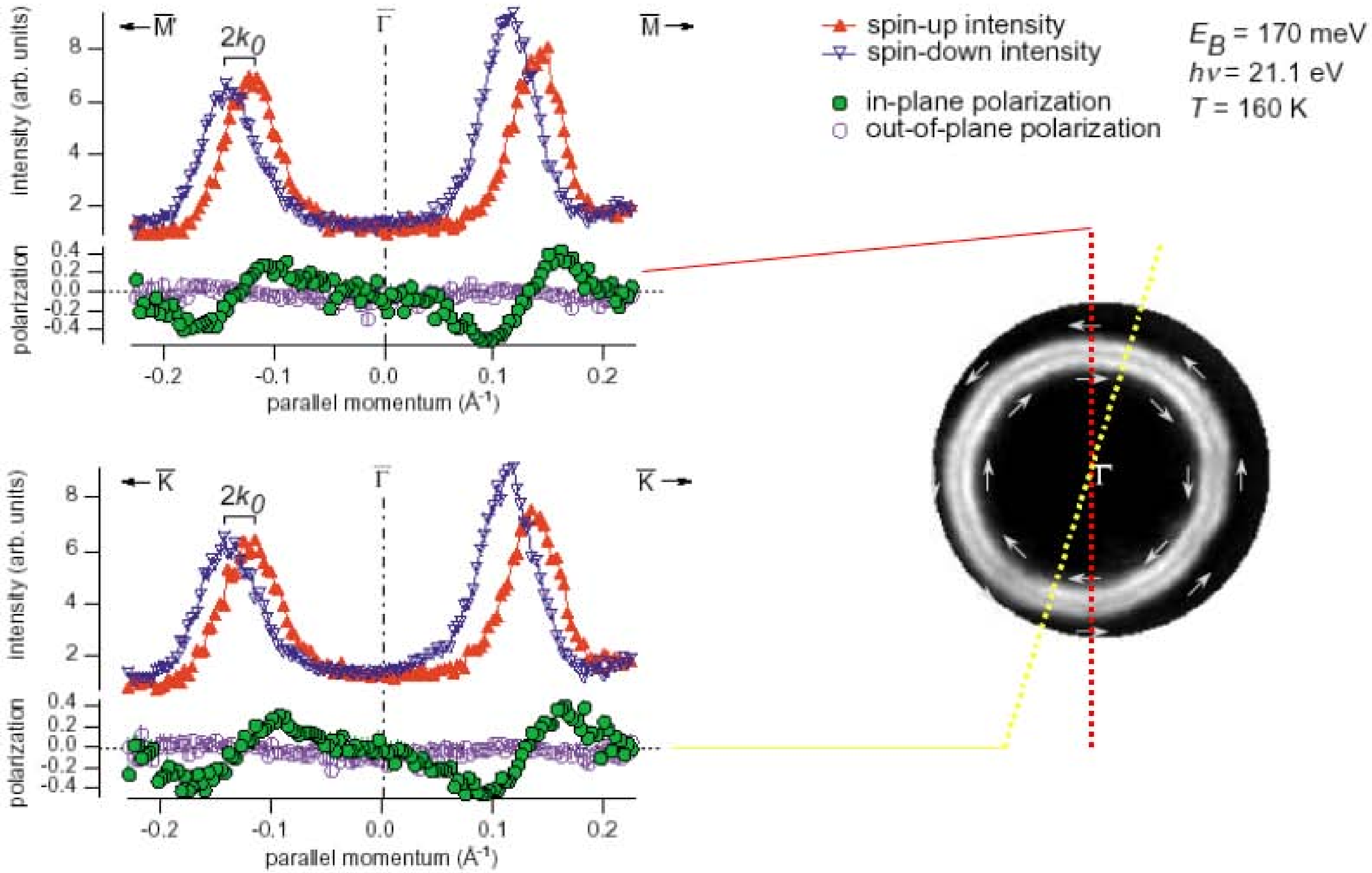


two orthogonal axes are spanned
by two pairs of detectors

$$I_{\uparrow,\downarrow}(E,\boldsymbol{k}) = I_M(1 \pm P_I(E,\boldsymbol{k}))/2.$$



Spin-resolved momentum distribution curves of the surface state on Au(111)

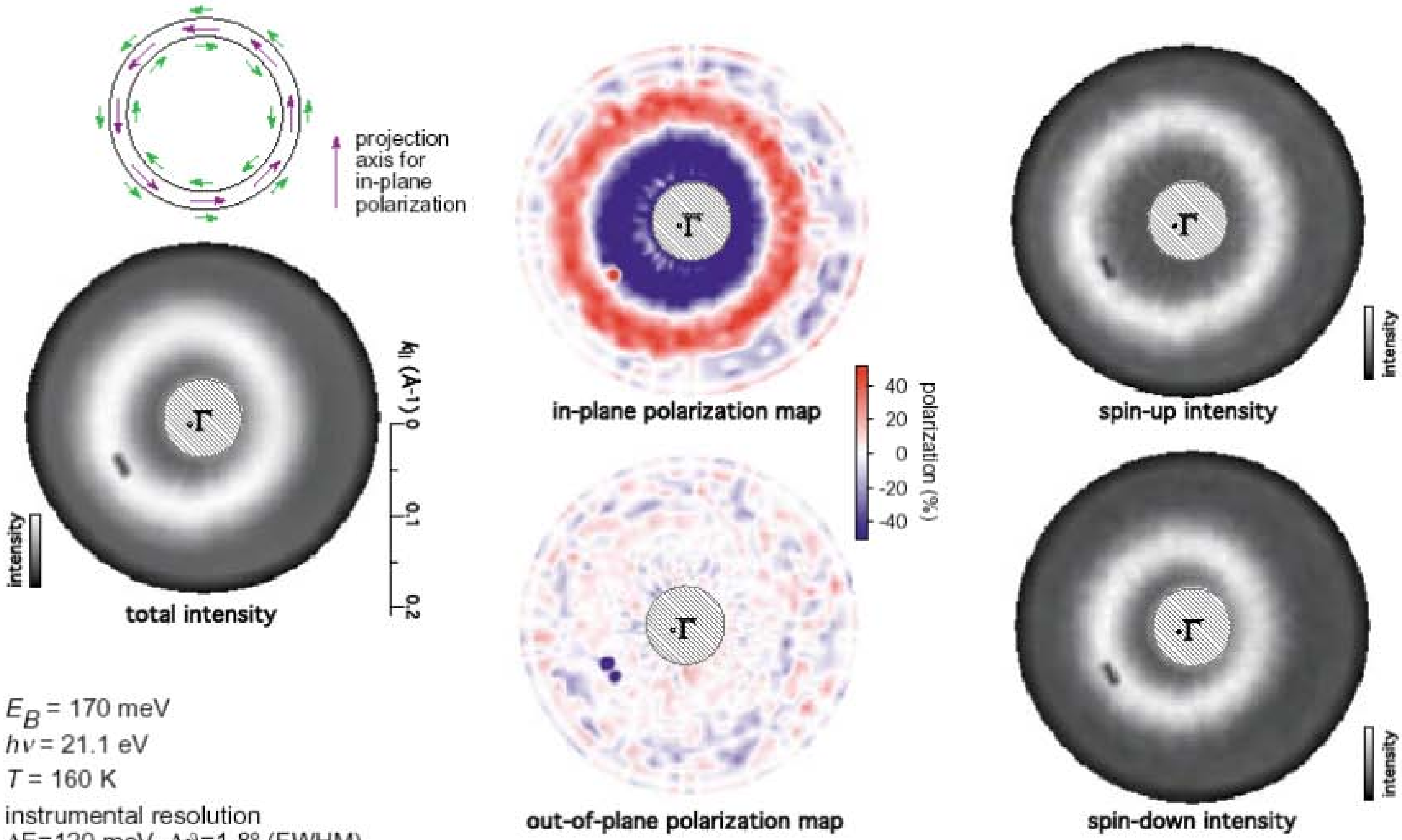


E_B = 170 meV
$h\nu$ = 21.1 eV
T = 160 K

=> The momentum shift is well resolved

instrumental resolution
ΔE=120 meV, Δϑ=1.8° (FWHM)

Spin-resolved momentum distribution map of the surface state on Au(111)



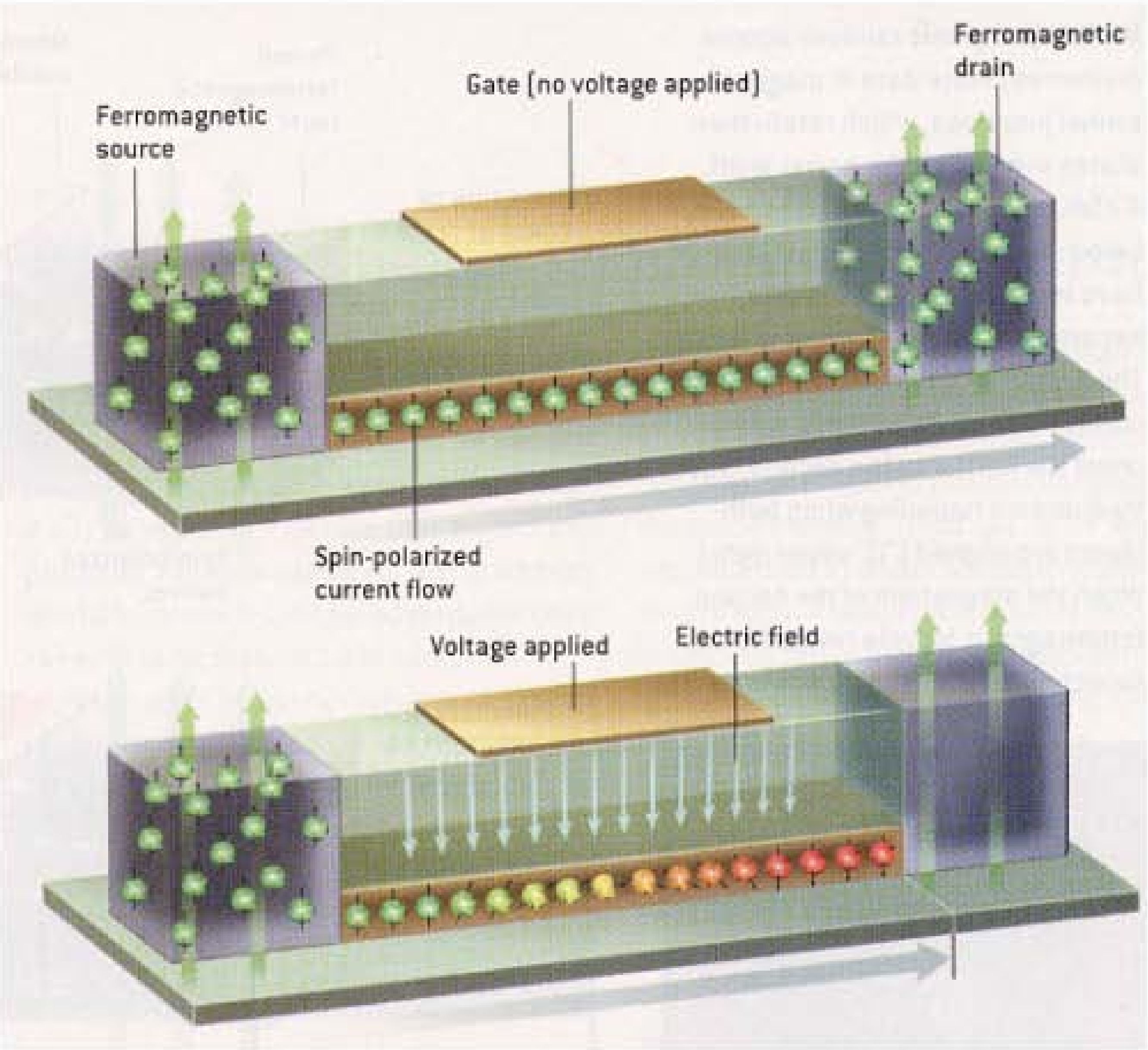
E_B = 170 meV
$h\nu$ = 21.1 eV
T = 160 K
instrumental resolution
ΔE=120 meV, Δϑ=1.8° (FWHM)

⇒ First spin-resolved "Fermi surface" map

⇒ Rotating spin structure is confirmed

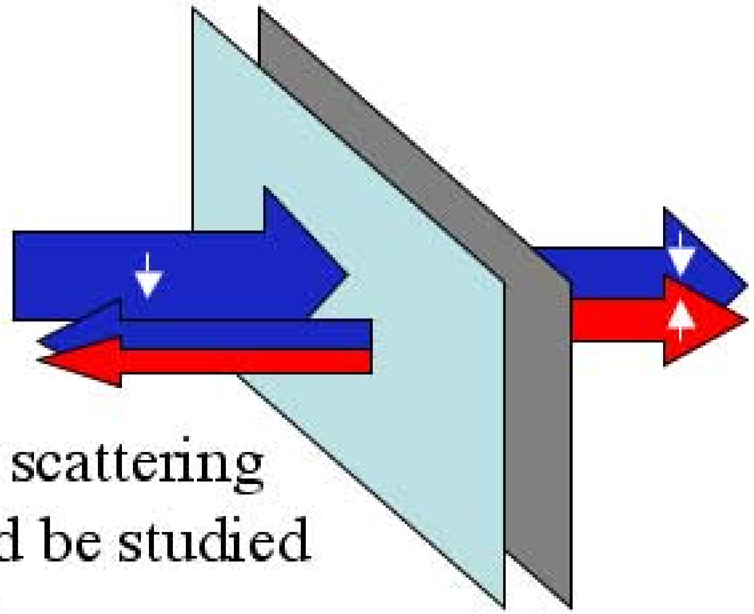
Spintronics: The Importance of Interfaces and Defects

e.g. the spin field effect transistor
… largely bases on such spin-orbit effects.

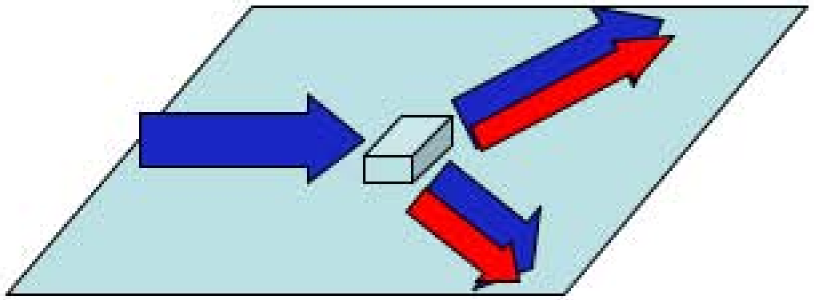


S. Datta & B. Das,
Appl. Phys. Lett. 56, 665 (1990)

But to make such a device work:



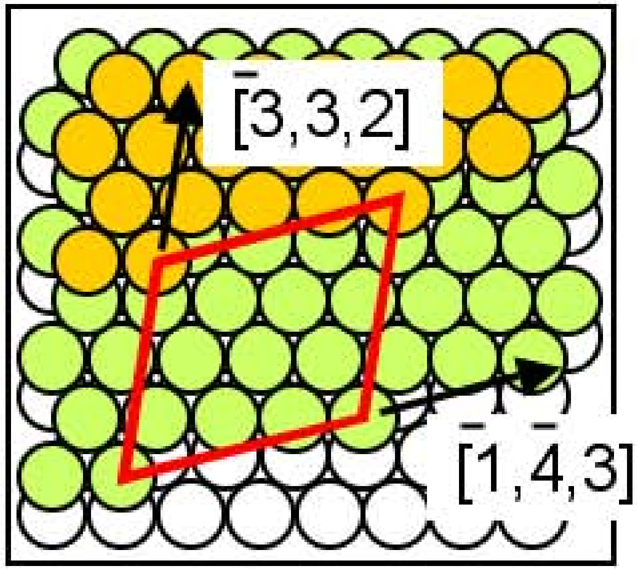
Spin-dependent scattering
processes should be studied
and controlled !



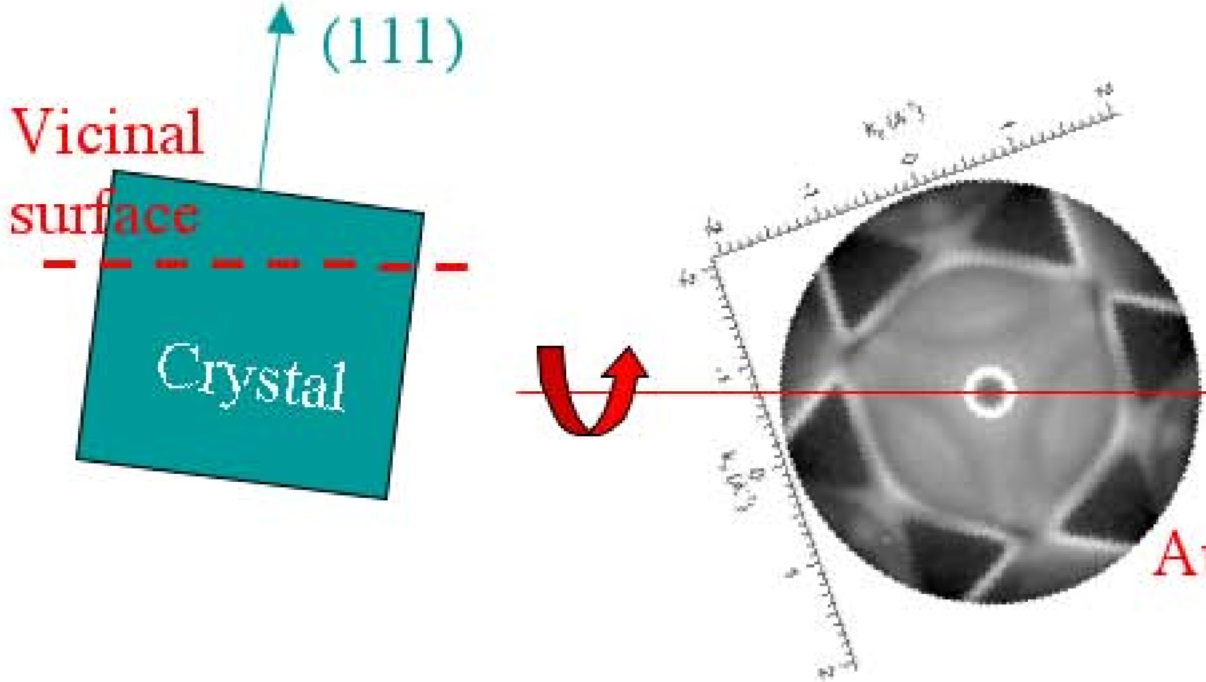
⇒ We currently study the effect of steps and kinks on the spin structure of Au(111).

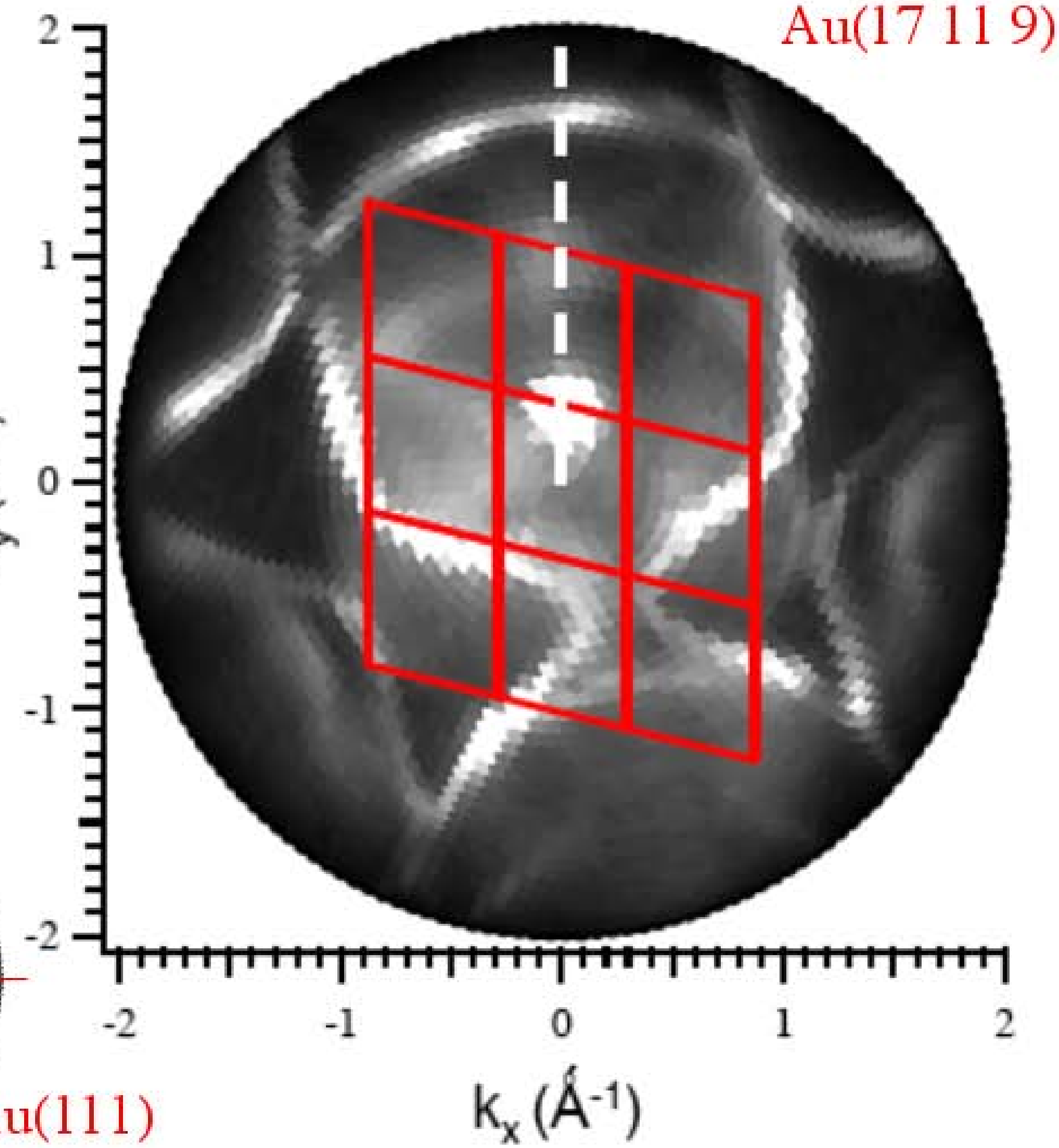
Au(111) Surface State - Interaction with Steps and Kinks

Vicinal Au(17 11 9) surface:



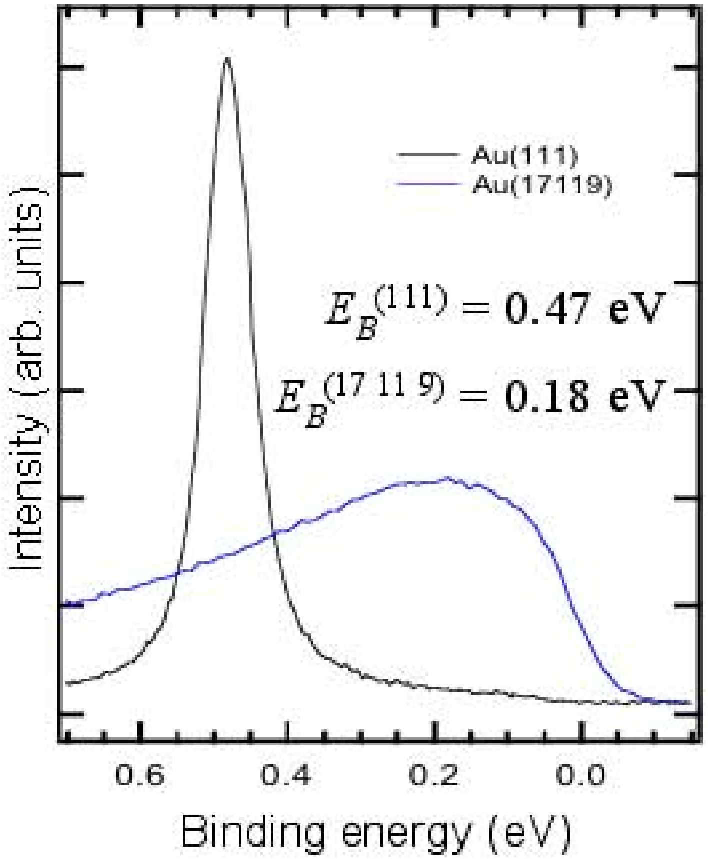
Steps and Kinks !



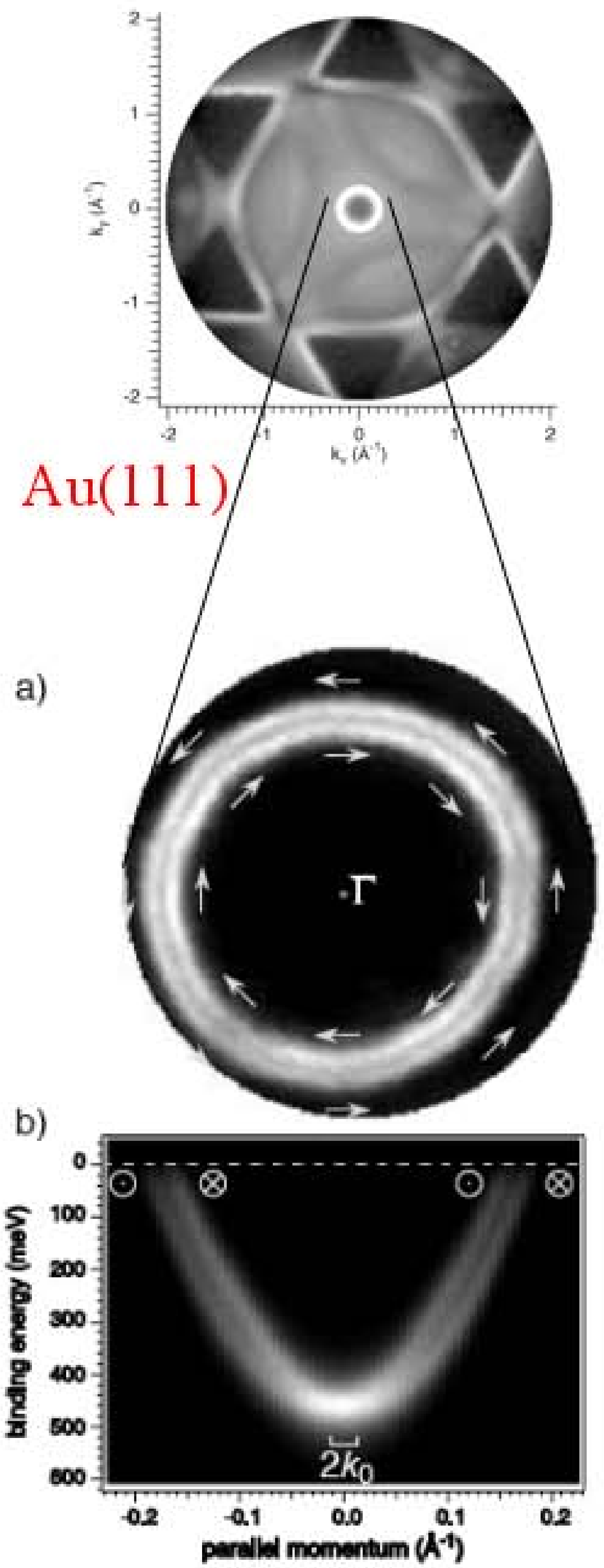


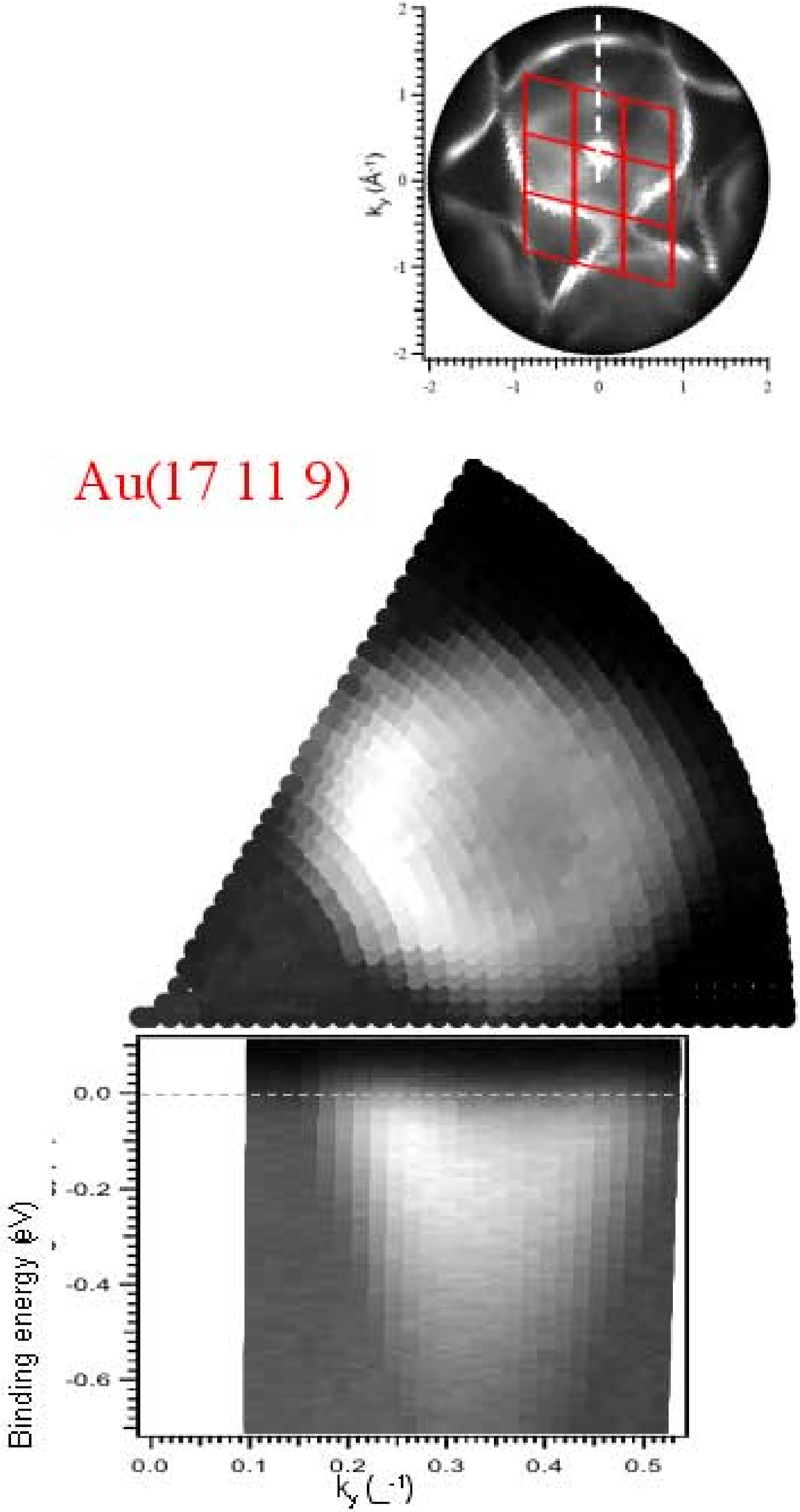
Surface state centered at zone boundary
=> propagates along the macroscopic surface

Effects on the Surface State



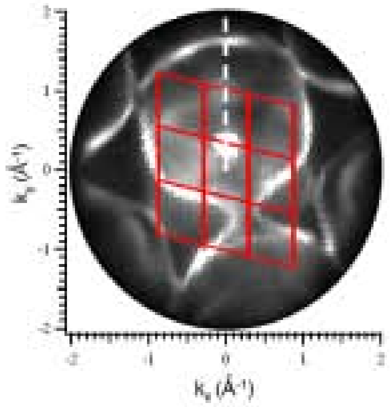
- Band bottom shifts towards E_F
- Effective mass increases (from $m^* = 0.24\ m_e$ to $m^* = 0.38\ m_e$)
- Peak width large due to terrace width distribution … and spin splitting ?



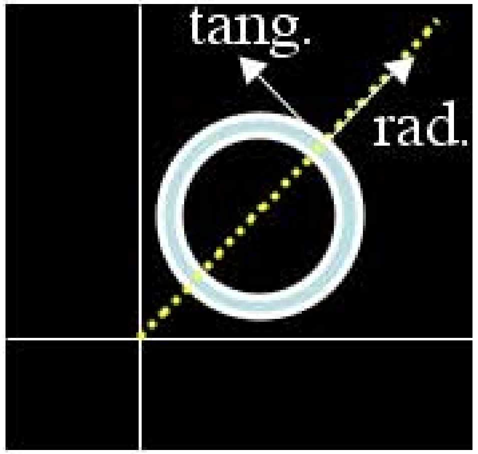


Spin-Polarized ARPES on Au(17 11 9)

MDC through surface state:
(preliminary data)



Peaks are much broader than Rashba splitting

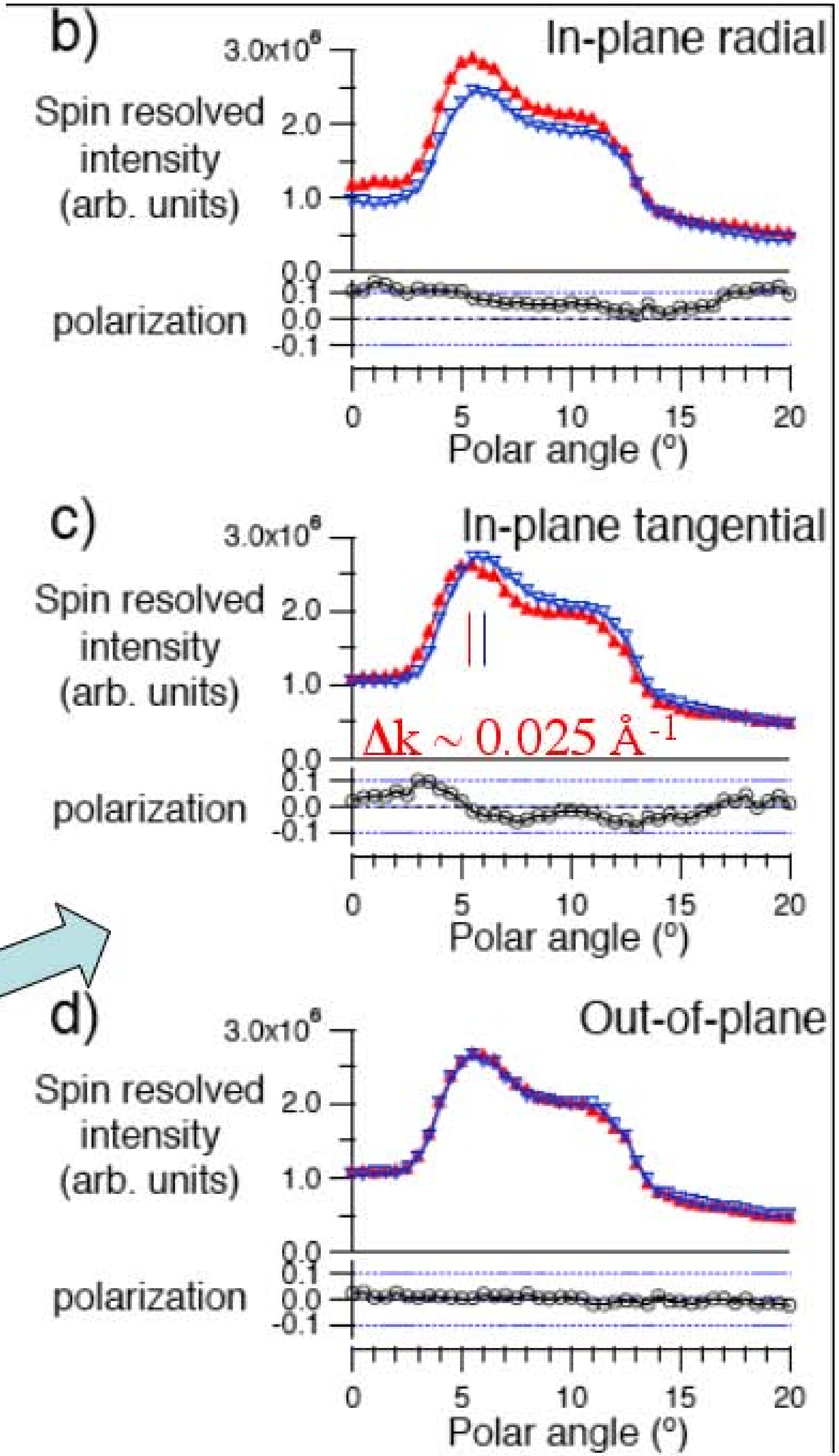


Spin polarized ARPES:

… sees polarization in the radial in-plane component ?

… can resolve a spin splitting in the tangential in-plane component !

… sees no out-of-plane polarization



… We are currently exploring the detailed spin structure …

Conclusions

- Spin-polarized photoemission is still a tedious experiment !
  (ca. 10^3 times slower than ARPES)
  But it is worth the effort !
- Spin-resolution can identify specific bands as minority or minority bands.
- Spin-polarized photoemission can measure spin structures in reciprocal space: e.g. Au(111) and Au(17 11 9)
- Spin resolution can markedly enhance the effective energy resolution for spin-split peaks, even if there individual peaks are broad.