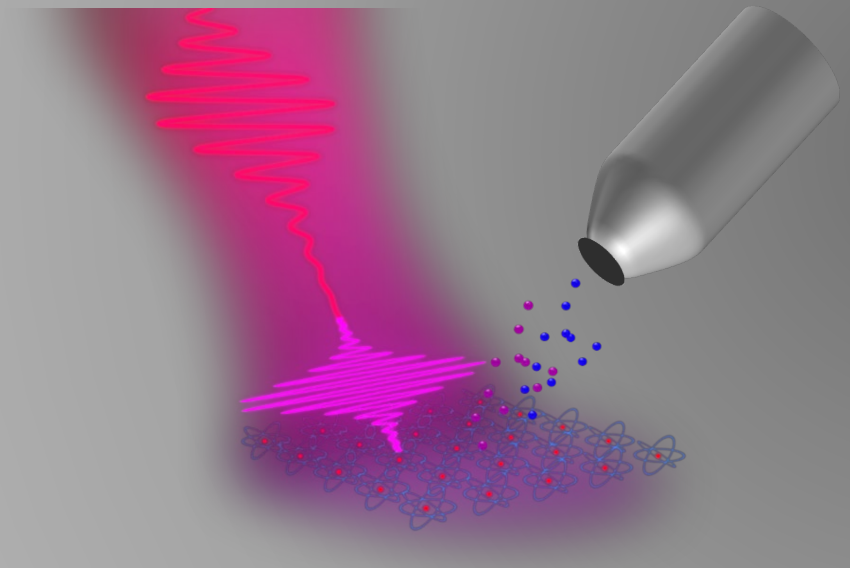
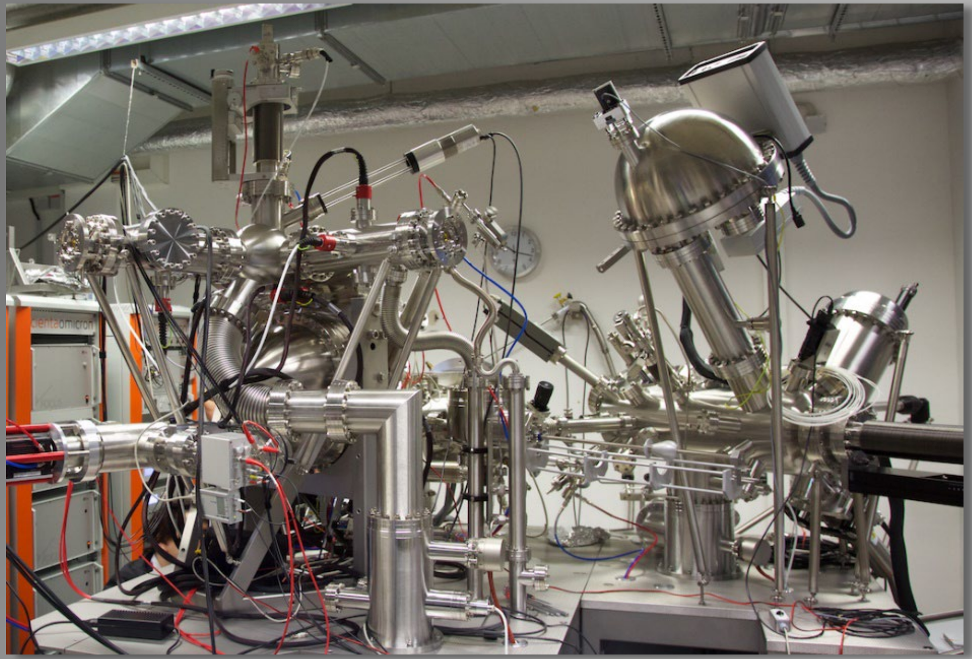
ELI ALPS NanoEsca surface science end station

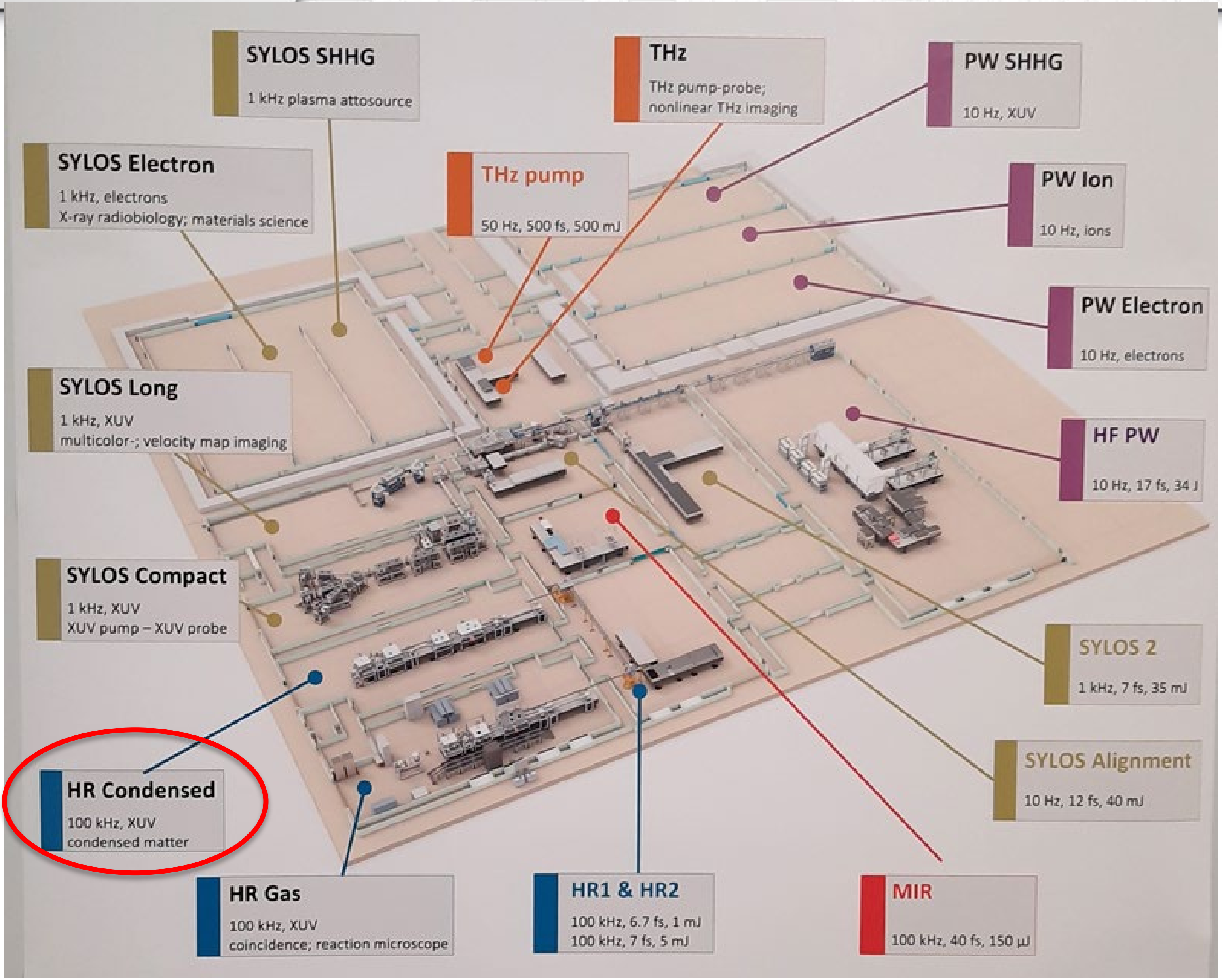
Spin and time-resolved momentum microscopy on solid surfaces





Martin Aeschlimann University of Kaiserslautern

NanoESCA surface science end station



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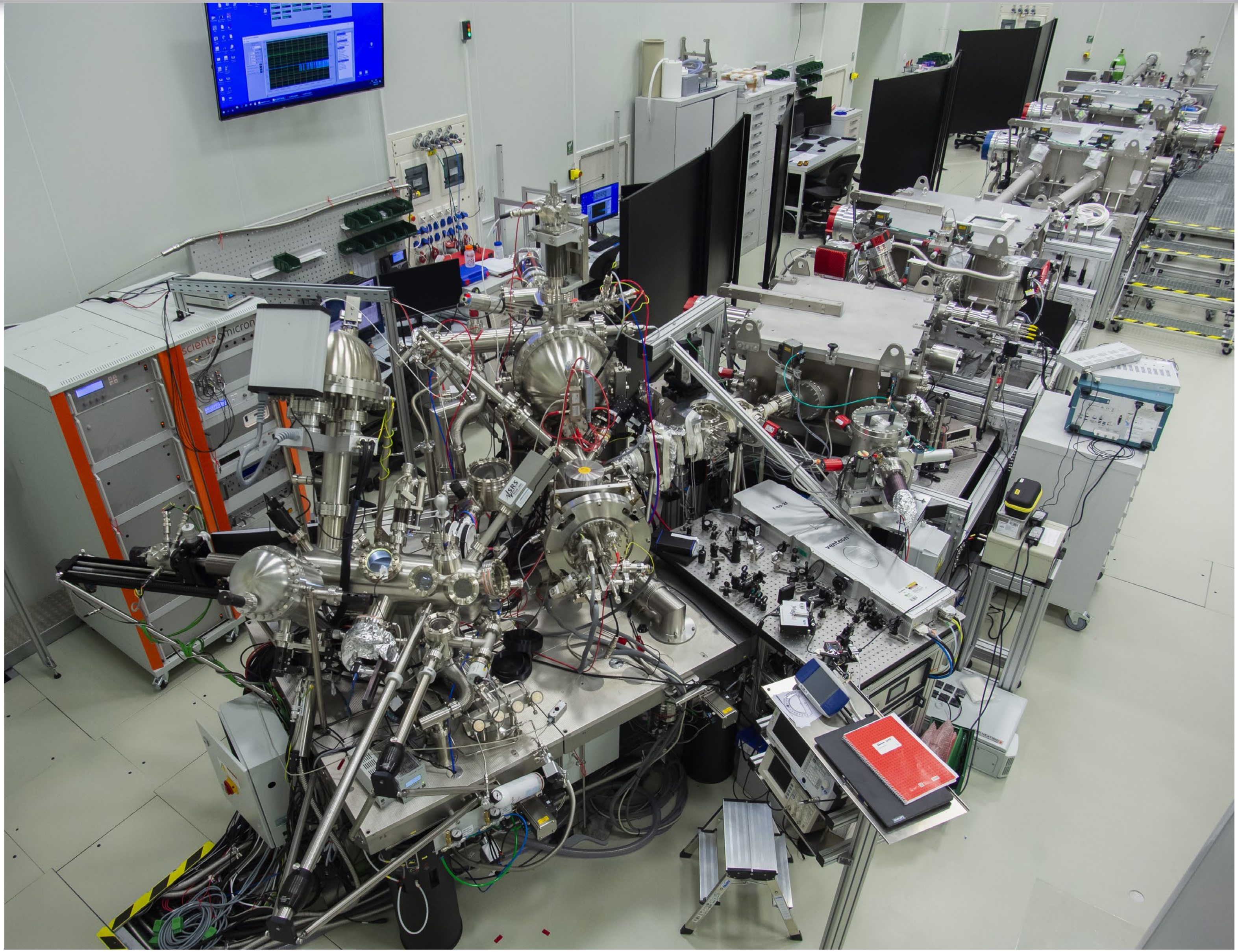


NanoESCA surface science end station

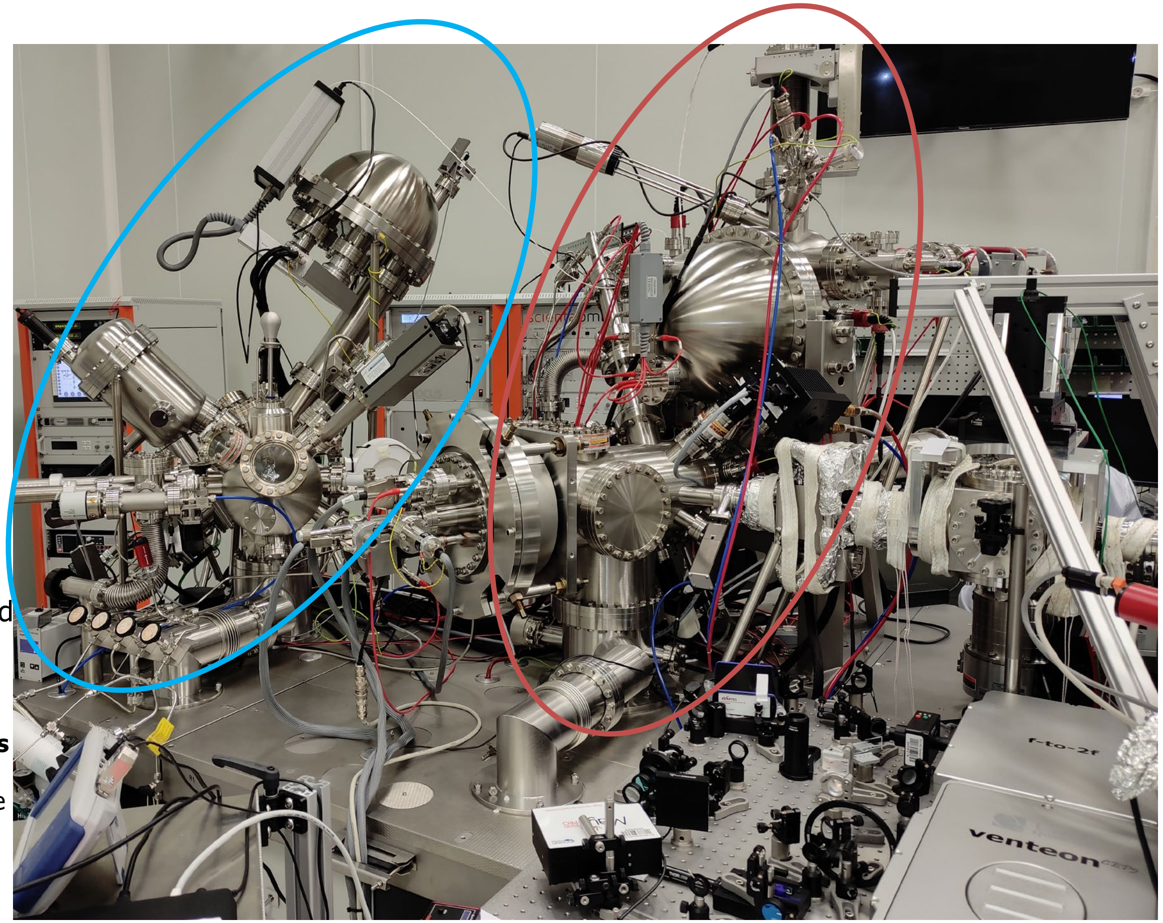
Preparation chamber
sample cleaning, preparation, and characterization

Analysis chamber
NanoESCA – nano- Electron Spectroscopy for Chemical Analysis

Cleaning:
- Ar^+ ion sputtering
- Annealing (heat- and coolable manipulator)

Preparation:
- **e-beam evaporator** for metal deposition
- **gas dozer** based on a capillary array
- **magnetizer**
    - Helmholtz coil
    - B field: up to 43 mT

Characterization (laterally averaged):
- **LEED** (Low Energy Electron Diffraction)
- **AES** (Auger Electron Spectroscopy): determination of surface structure and composition
- **XPS** (X-ray photoelectron spectroscopy):
    - **quantitative chemical analysis of the surface** (top few nm)
    - monochromatic Al K_{α} X-ray source
    - 128 detection channels
- **RGA** (Residual Gas Analyzer) by quadrupole mass spectrometer



Modes (outputs):

PEEM: Photoemission electron microscope

ToF-PEEM: Time of flight + DLD analyzer (delay line detector)

Spectroscopy for selected area: Channeltron detector after first hemisphere

IDEA: Imaging with dispersive energy analyzer

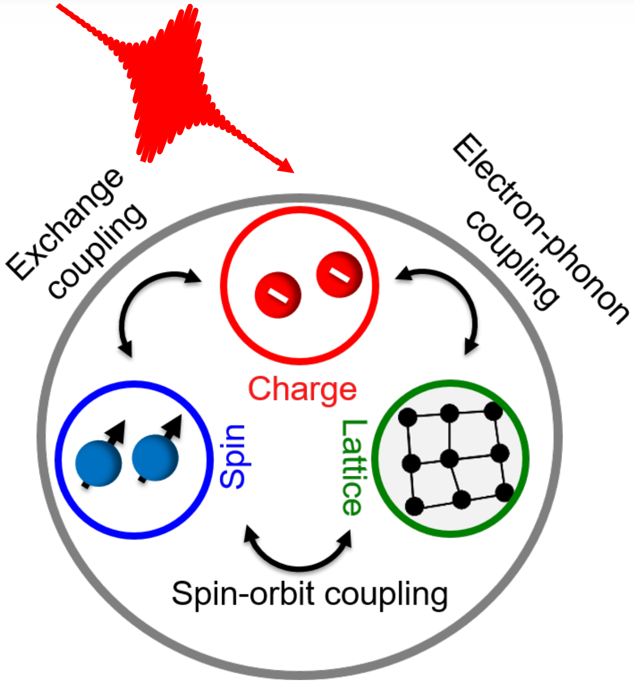
Imaging spin filter: IDEA + spin selective mirror: gold coated Ir(100) crystal

Light sources:

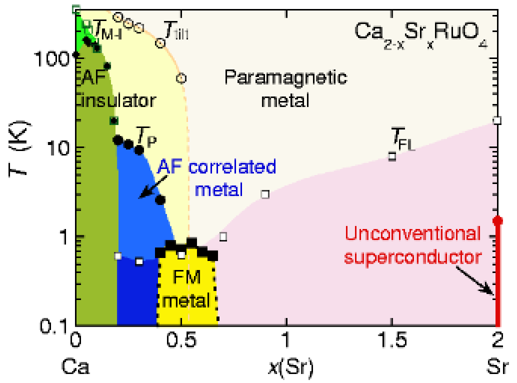
Internal (CW) sources:
- Hg arc lamp (5.2 eV – 238 nm)
- He discharge lamp
    - **HeI: 21.22 eV – 58 nm**
    - HeII: 40.81 eV – 30 nm

Short pulsed sources:
- pump: HR laser fundamental (1.2 eV – 1030 nm) 100kHz
- probe: HHG generated XUV (20-90 eV – 62-13.8 nm) 100 kHz – „BeamLine condensed"
- Venteon CEP5 oscillator (1.5 eV – 830 nm) 80MHz

Competing interactions of spin with charge and lattice



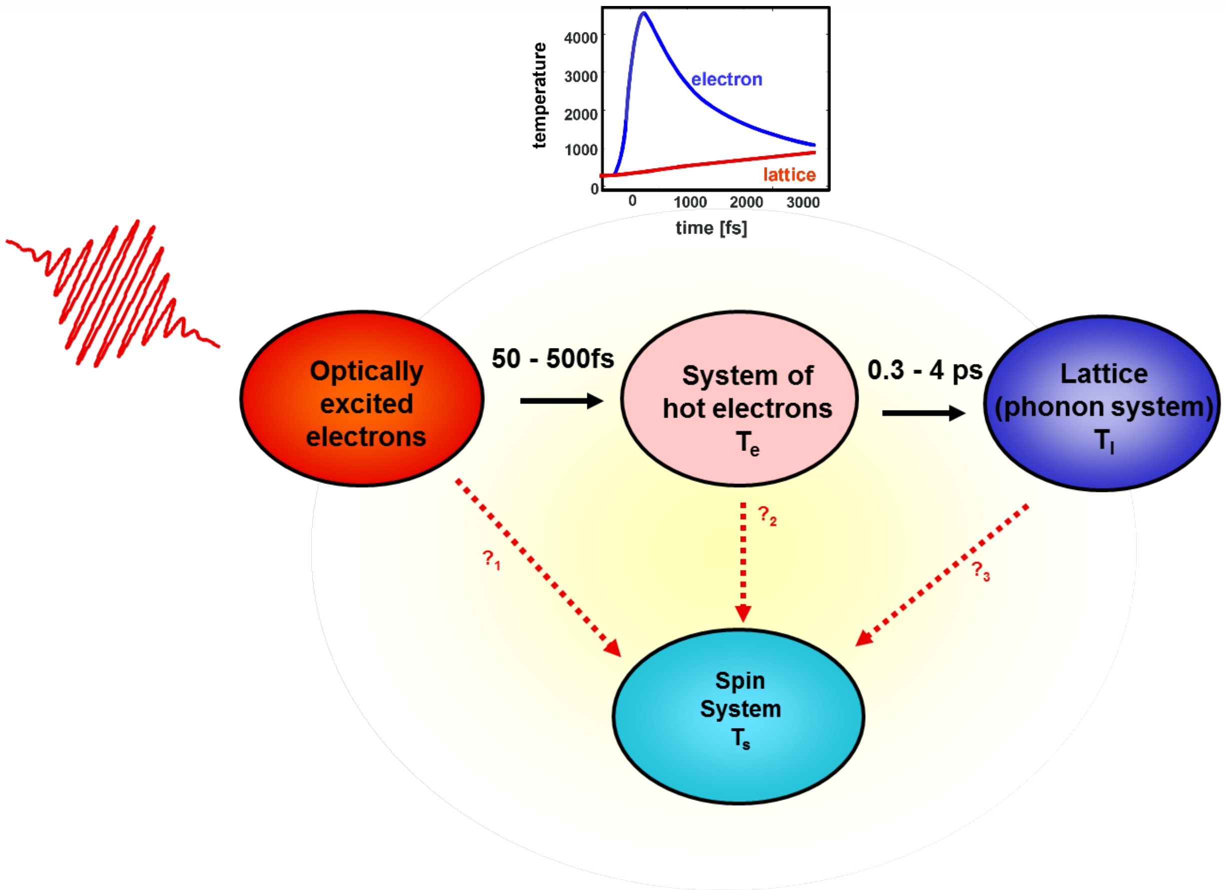
- the complex interplay between the fundamental sub-systems spin, charge, and lattice are responsible for the appearance of novel phases of matter.
- yield rich phase diagrams of states in novel correlated-electron materials



S. Nakatsuji et al., Phys. Rev. Lett. 93, 146401 (2004).

- the dominant interaction that is responsible for the formation of a specific phase is most often hard to determine in thermal equilibrium,
- by driving the material system temporally out of equilibrium and using time-resolved spectroscopy techniques (e.g. various photoemission techniques) paves the way to overcome these limitations.

Competing interactions of spin with charge and lattice



microscopic mechanism ?

⇒ **Utrafast thermal nonequilibrium allows to study the competing interactions of spin with charge and lattice**

First HHG light in the NanoESCA end station :

23. September 2022

⇒ Static measurements

Internal CW Light sources:

- Hg arc lamp (5.2 eV – 238 nm)
- He discharge lamp
  - **HeI: 21.22 eV – 58 nm**
  - HeII: 40.81 eV – 30 nm

ELI-ALPS Research Institute
TOWARDS THE SHARP END OF ATTOSCIENCE

The Electronic Structure of Two-Dimensional Hexagonal Boron Nitride on Au coated Rh(111) surface studied by NanoESCA

Csaba Vass[1], Gy. Halasi[1,2], G. Vári[2], A. P. Farkas[1,2], A. Berkó[2], K. Palotás[2,3], J. Kiss[2], K.M. Yu[4], B. Stadtmüller[4], M. Aeschlimann[4], P. Dombi[1,3], Z. Kónya[2], L. Óvári[1,2]

[1]ELI-ALPS (Szeged, Hungary)
[2]ELKH-University of Szeged
[3]Wigner Research Centre for Physics (Hungary);
[4]Technical University of Kaiserslautern

SZÉCHENYI 2020
Hungarian Government
European Union
European Regional Development Fund
INVESTING IN YOUR FUTURE

Hexagonal boron nitride (h-BN) on metals

Why hexagonal boron nitride (h-BN) ?

- structurally **similar to the graphene**
- very **good insulating support for graphene** nanoelectronics - only weakly influencing the properties of graphene.
- **easy to synthetize** and outstanding **stability**

hBN on metals: The surface structure is determined by

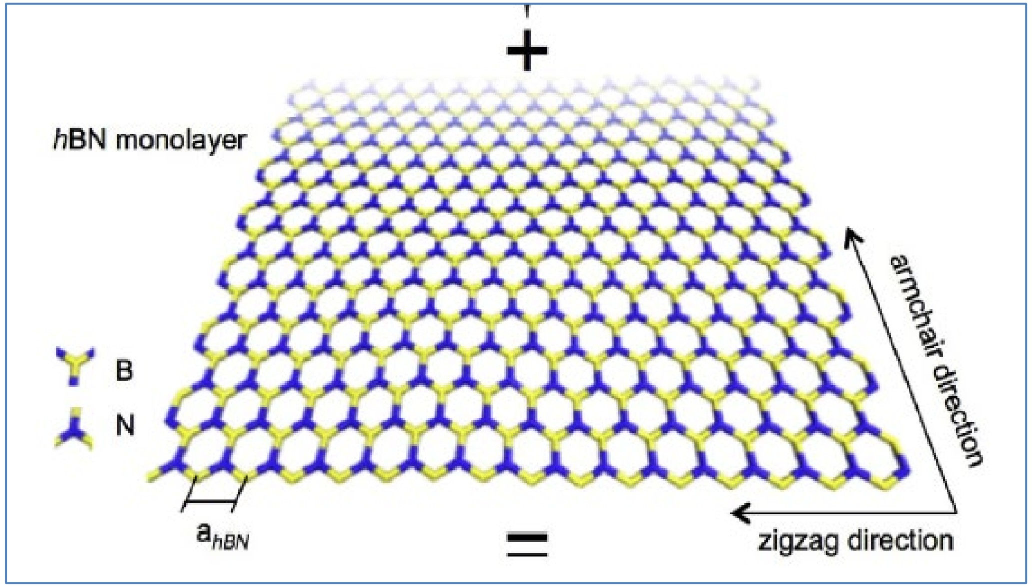
- the strength of interaction with the metal
- lattice mismatch

The interaction of h-BN with noble metals (Cu, Ag, **Au**) is weak.   => **(nearly) flat layers of h-BN**

hBN on Rh(111) surface:

- strong interaction
- lattice mismatch 7%
- A „13 h-BN on 12 Rh” moiré structure is formed.
- The interaction is stronger with N atoms located on top of Rh atoms. These „pore” regions are closer to the surface.

=> periodic corrugation, called **nanomesh**; template function



Auwaerter Surface Science Reports 74 (2019)

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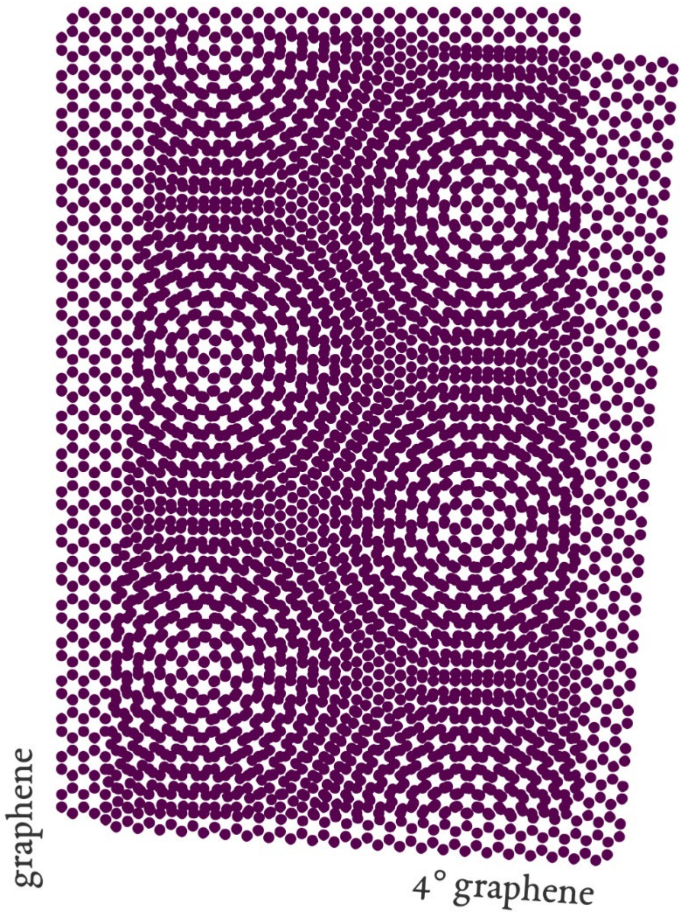

Moiré pattern arising from the superposition of two graphene lattices twisted by 4°

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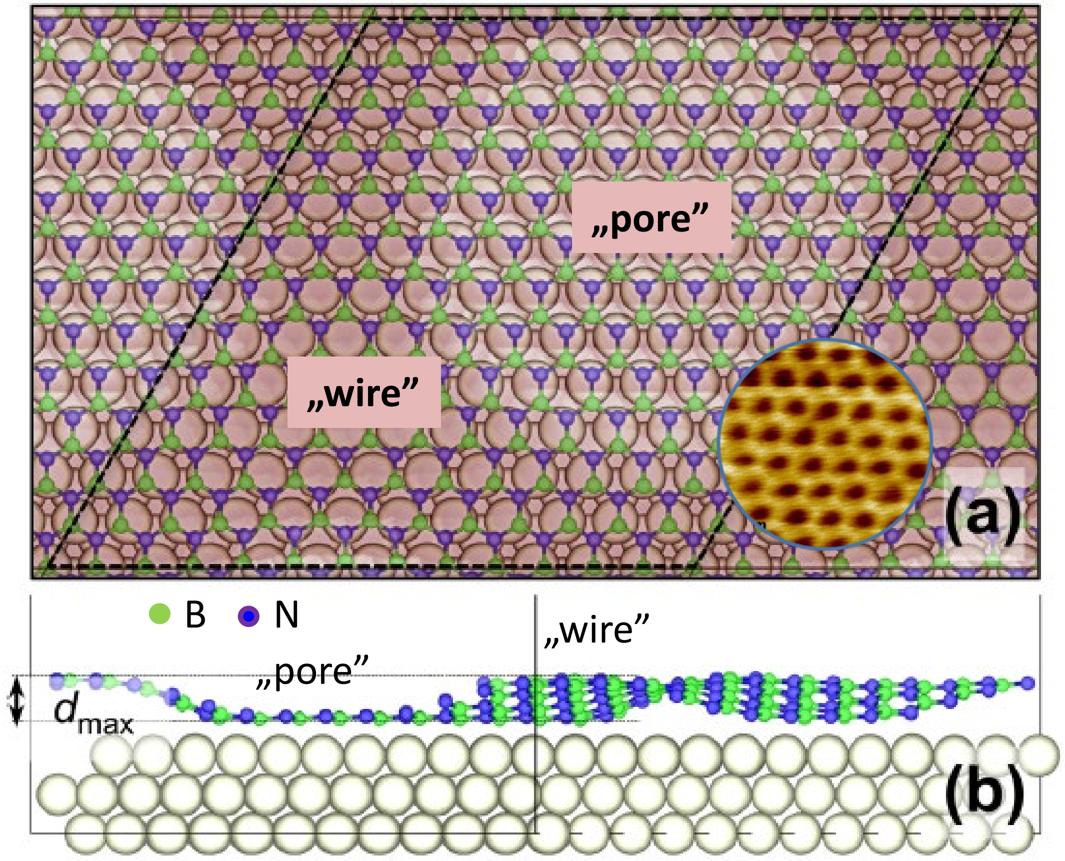


M. C. Patterson et al. Phys. Rev. B. 89 (2014) 205423.; S. Berner, et al. Angew. Chemie Int. Ed. 2007, 46, 5115

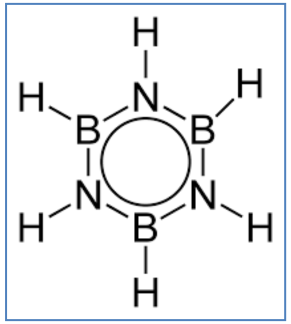
Auwaerter Surface Science Reports 74 (2019)

Sample preparation & STM measurements

What happen if we cover the Rh(111) surface with epitaxial grown Au film?

Sample preparation:

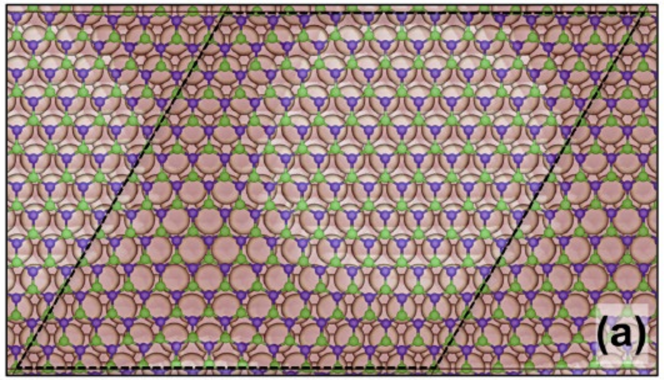
- Rh(111) cleaning with Ar^+ sputtering & annealing cycles
- Au deposition @500K & annealing @1000K, afterwards
- Borazine exposure @1000-1050 K
  - decomposition of borazine ($B_3N_3H_6$) at ~1050 K
  - h-BN nanomesh, self limited to 1 ML

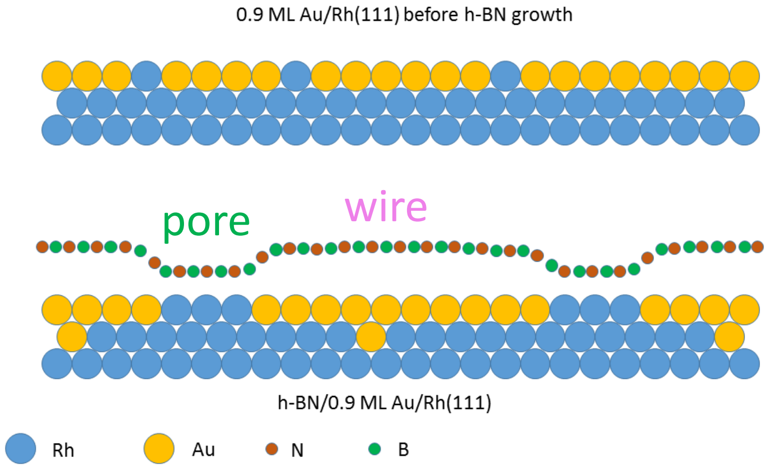


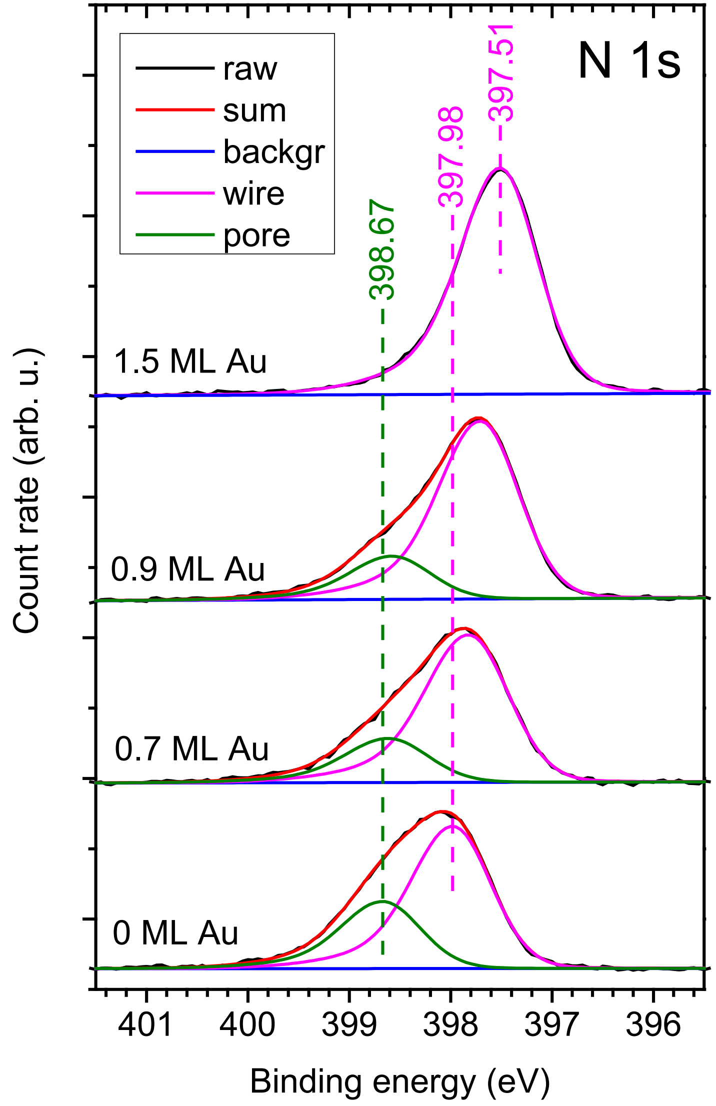
borazine

XPS analysis of h-BN on Au covered Rh(111)

- The N 1s region of the h-BN nanomesh is composed of two components due to the pore-wire duality.
- The position of the wire component in the N 1s region is sensitively influenced by Au.
- The pore component shifts much less in the presence of Au.
- **During h-BN growth, Au is removed from below the pore region, and is accumulated below the wire region with strong preference.**
- In accordance with STM, the pore /wire XPS area ratio decreases as an effect of Au.







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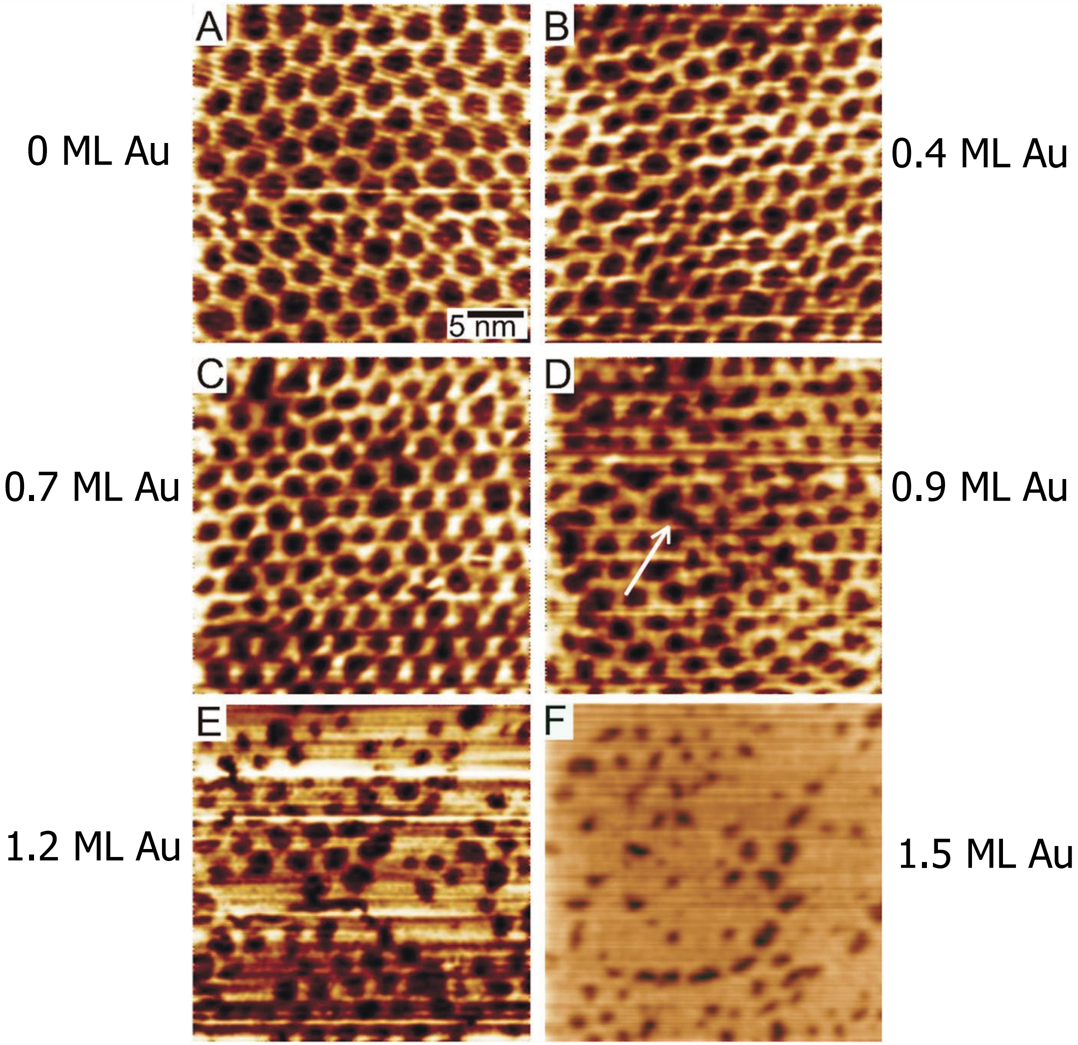
STM:
- The nanomesh morphology is present up to 0.9 ML of Au
- The pore diameter decreases with increasing amounts of Au
- At higher Au doses the surface stepwise flattens out



R. Gubó, G. Vári, J. Kiss, A. P. Farkas, K. Palotás, L. Óvári, A. Berkó, Z. Kónya, *Phys.Chem.Chem.Phys.* 2018, **20**, 15473.

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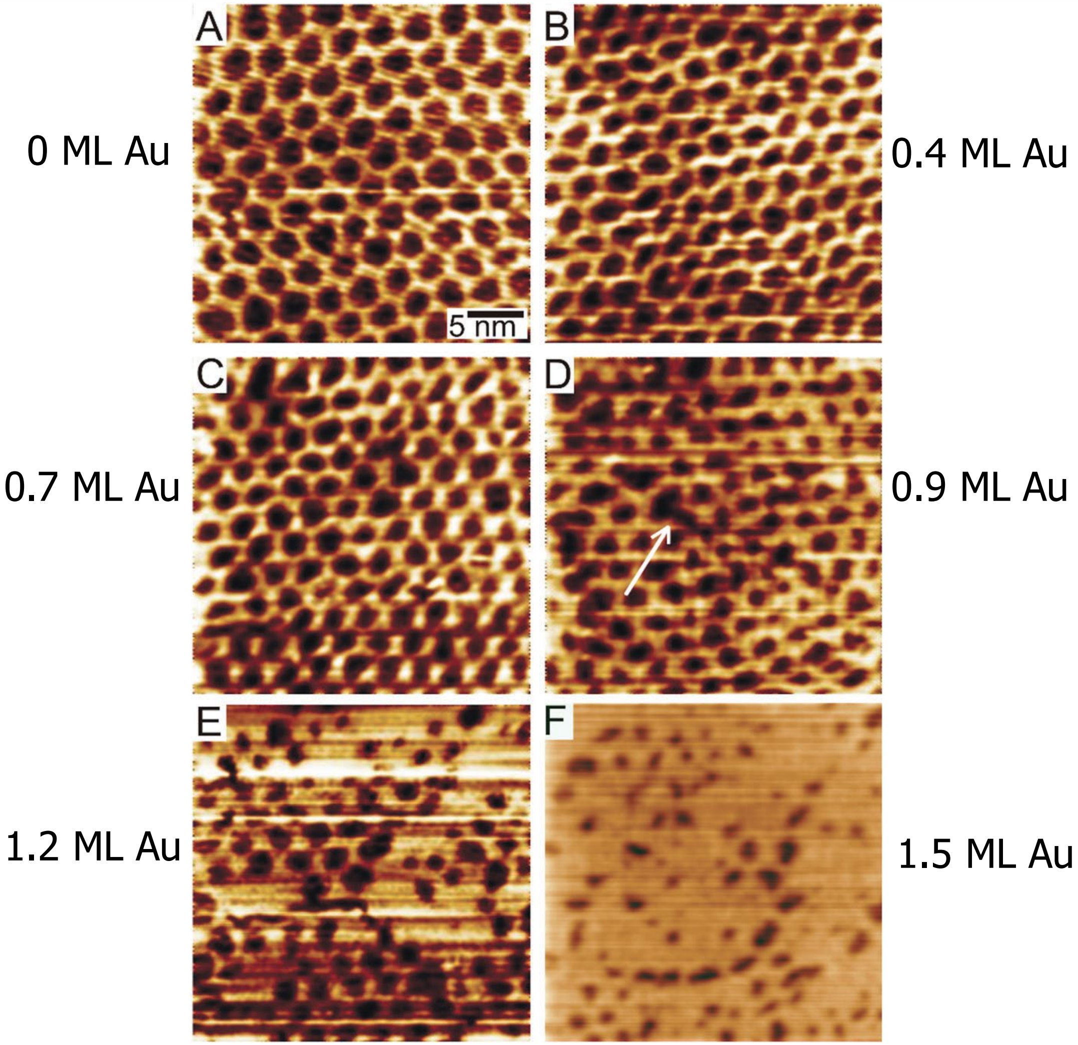
How does the electronic structure change as a result of increasing Au coverage?



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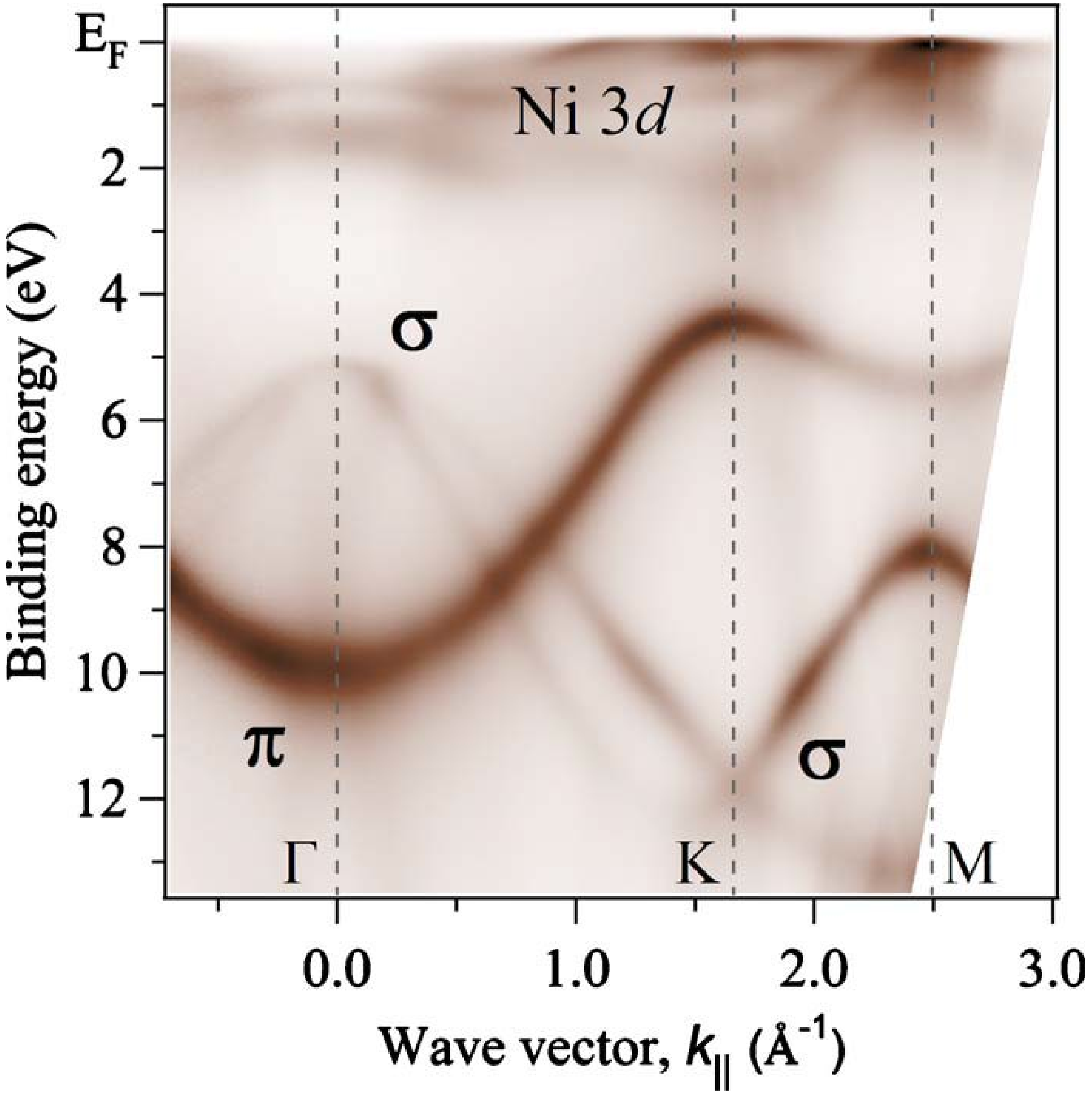
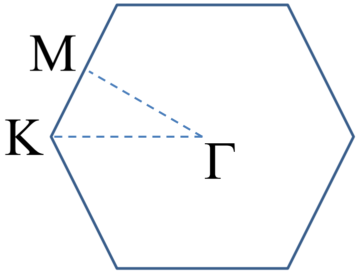
ARPES of h-BN/Ni(111)



- h-BN on h-BN/Ni(111) has a ***flat morphology*** because of the lack of lattice mismatch.
- Flat h-BN is characterized by one π band and two σ bands.

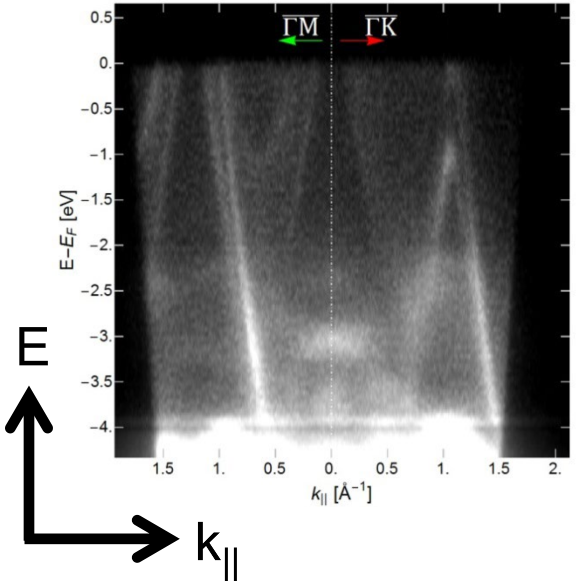


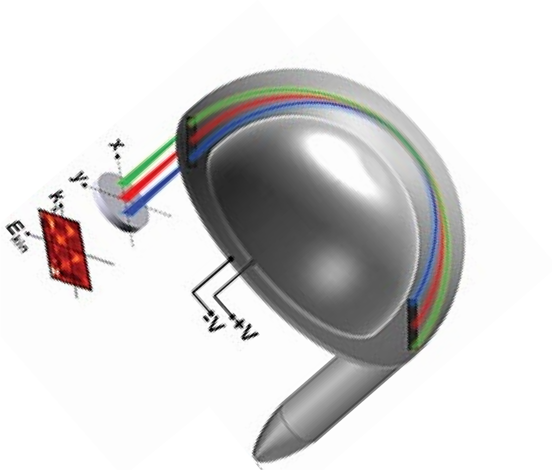
surface Brillouin zone

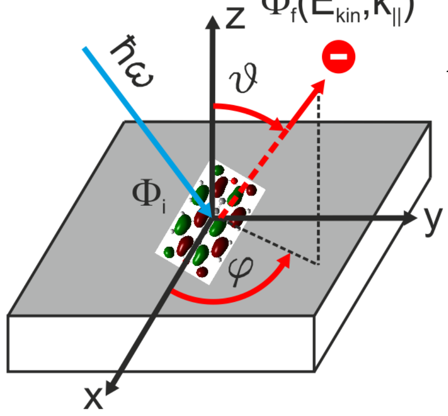
Usachov et al PHYSICAL REVIEW B 82, 075415 2010

Typical ARPES laboratory setup

2D datasets $E(k_x)$







$$E_{kin} = \frac{\hbar^2}{2\, m_e} k^2$$

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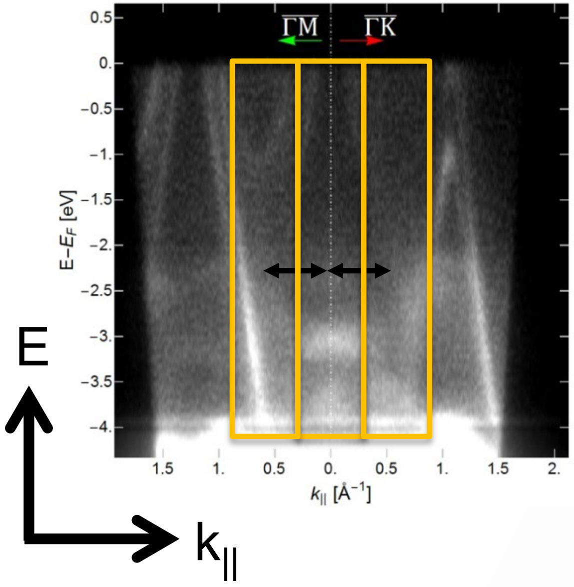

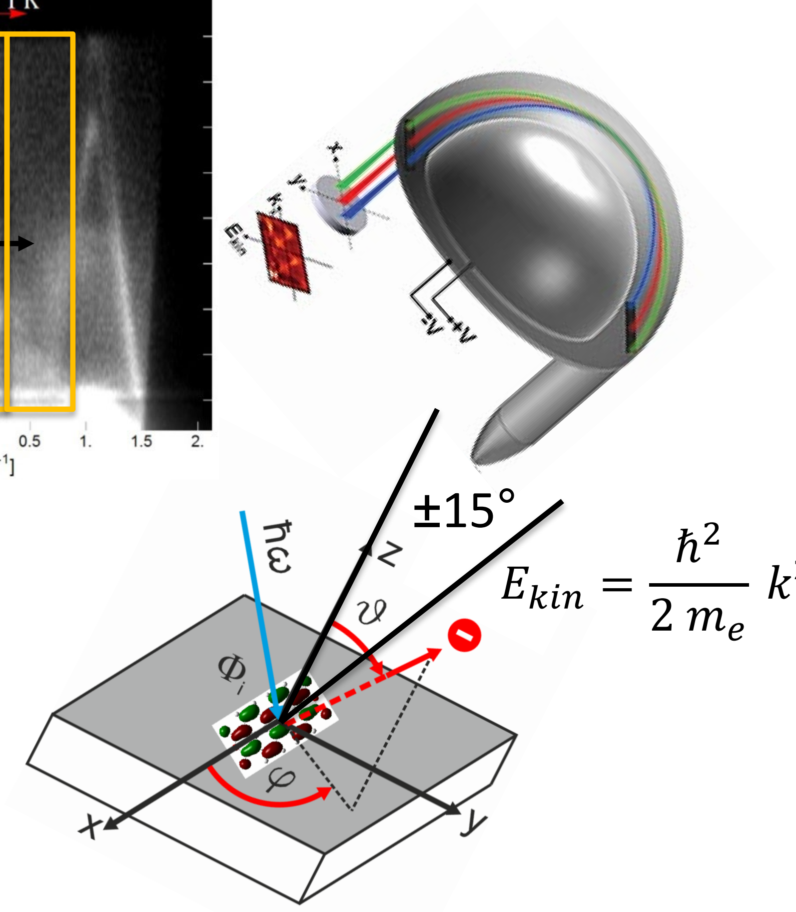

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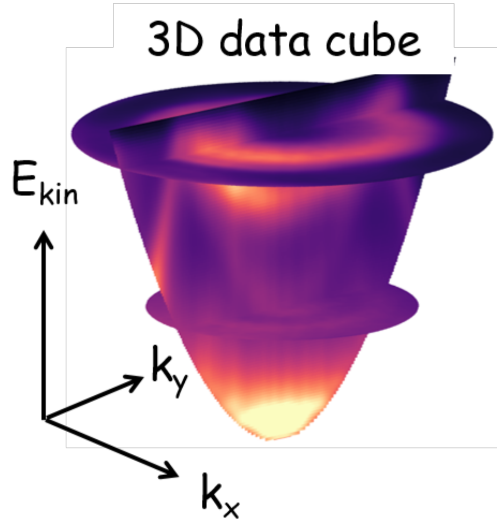
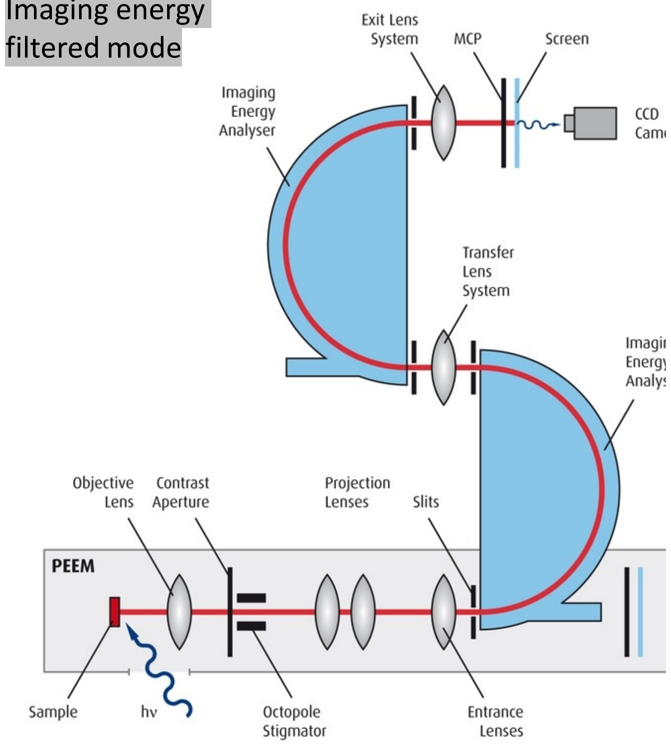
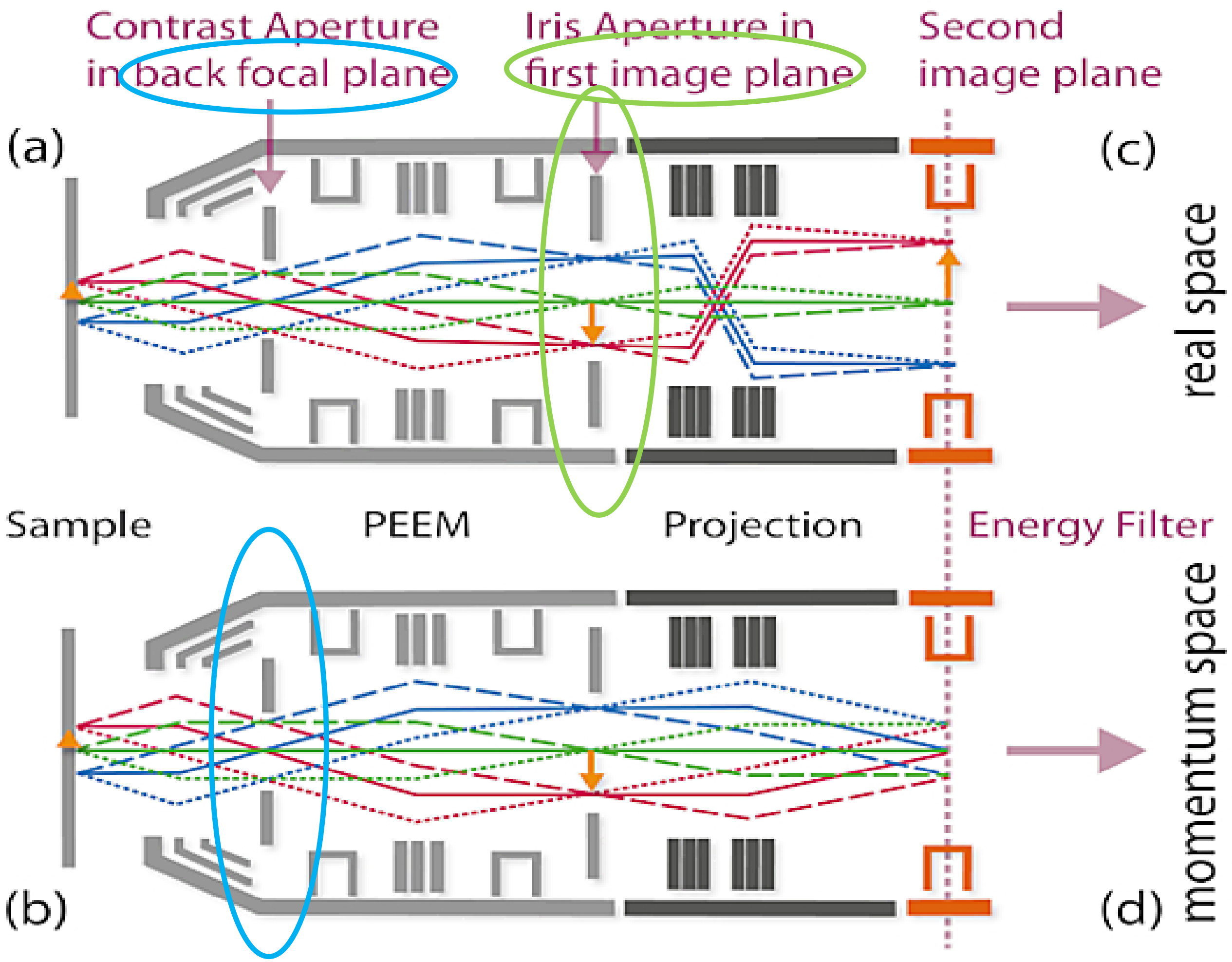


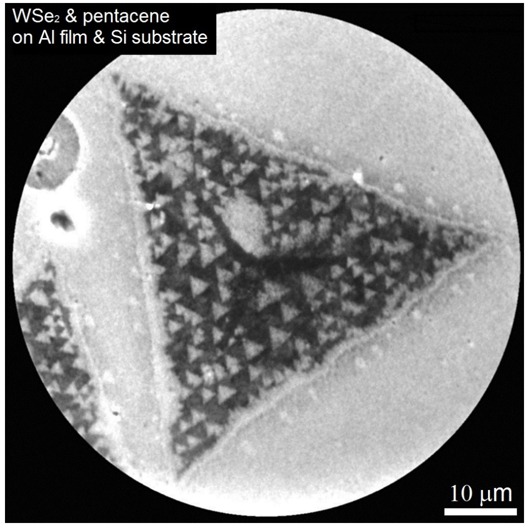
Is there another way for ARPES?

Operational modes: real space vs. momentum space

Imaging energy filtered mode

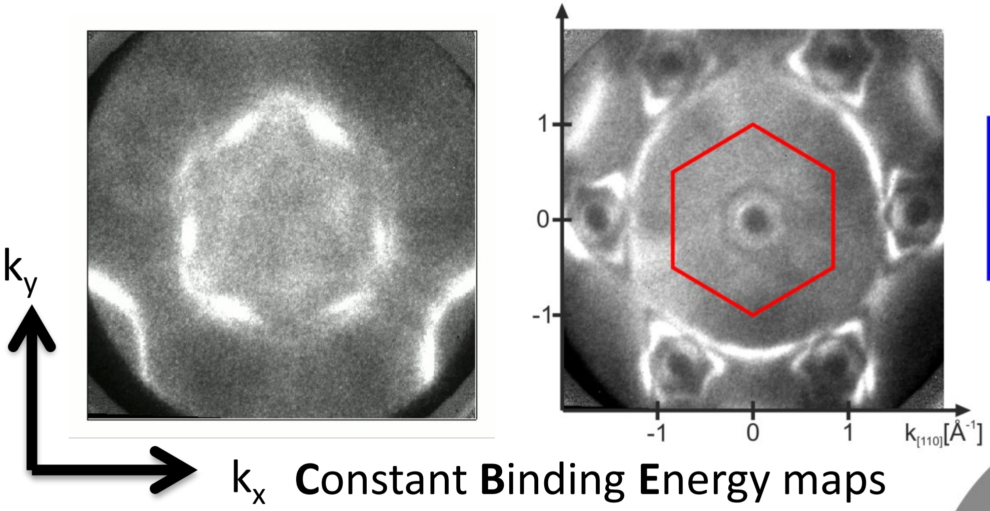




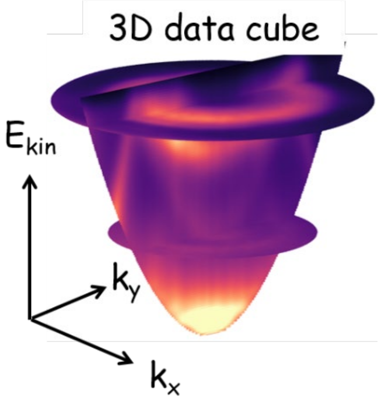


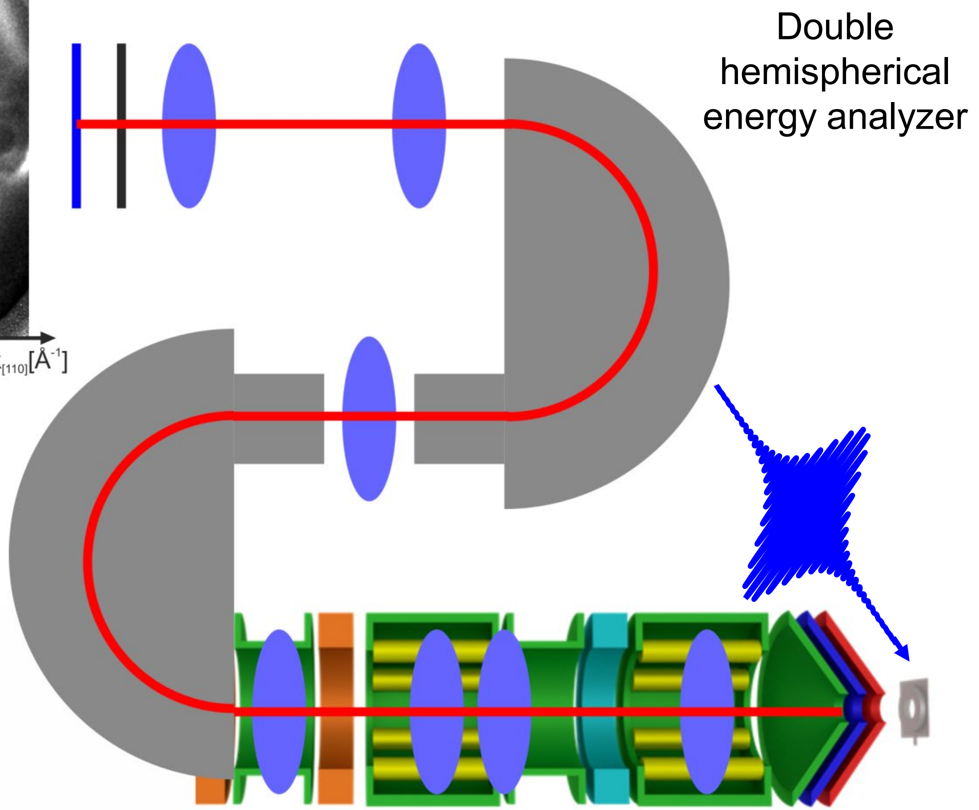
Au(111) Fermi surface @117 K

Momentum space microscopy



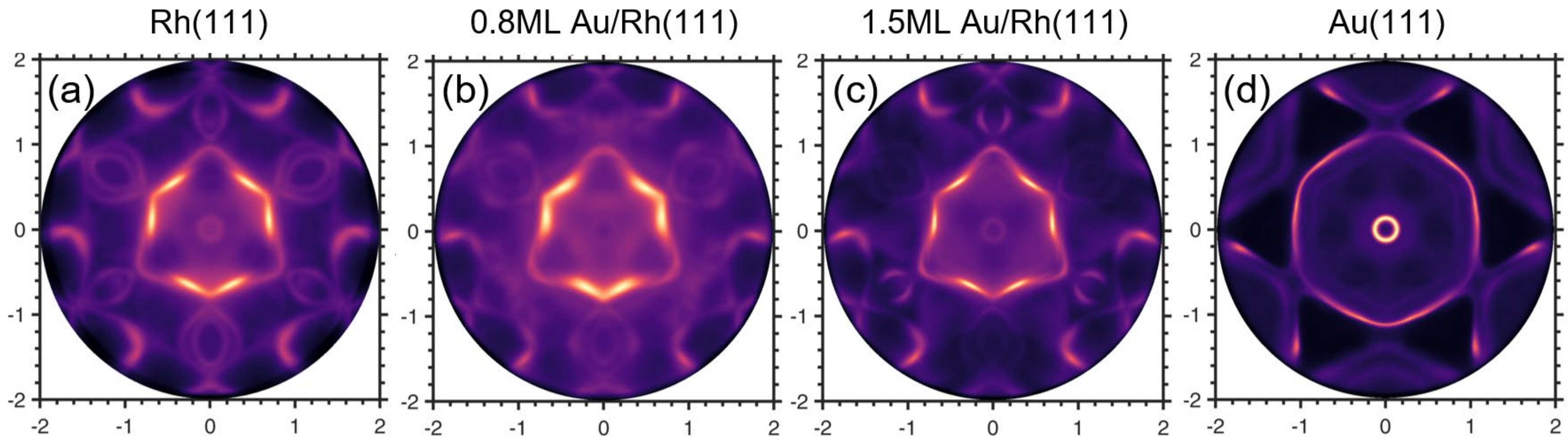
Constant Binding Energy maps



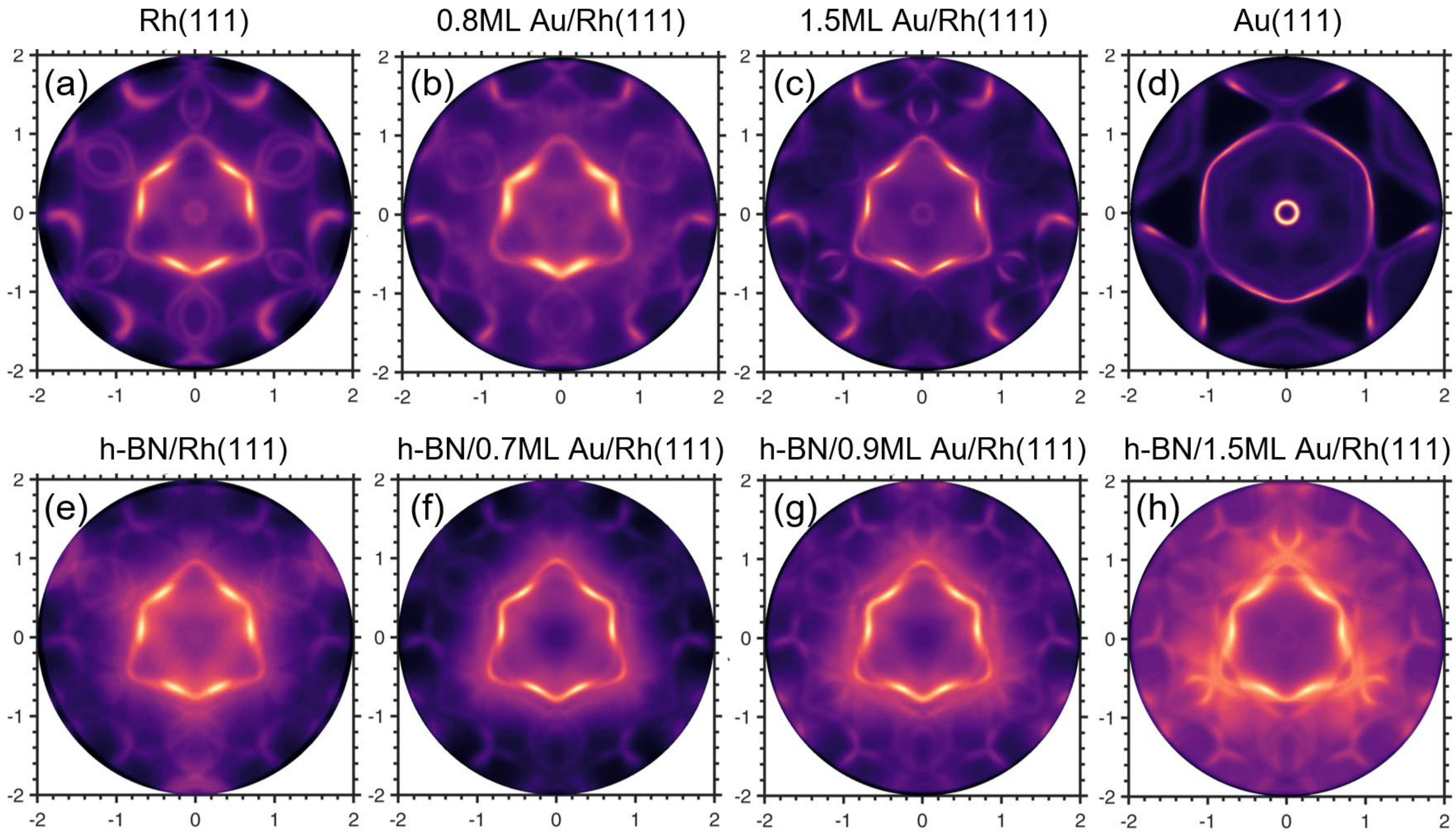


Photoemission electron microscope optics

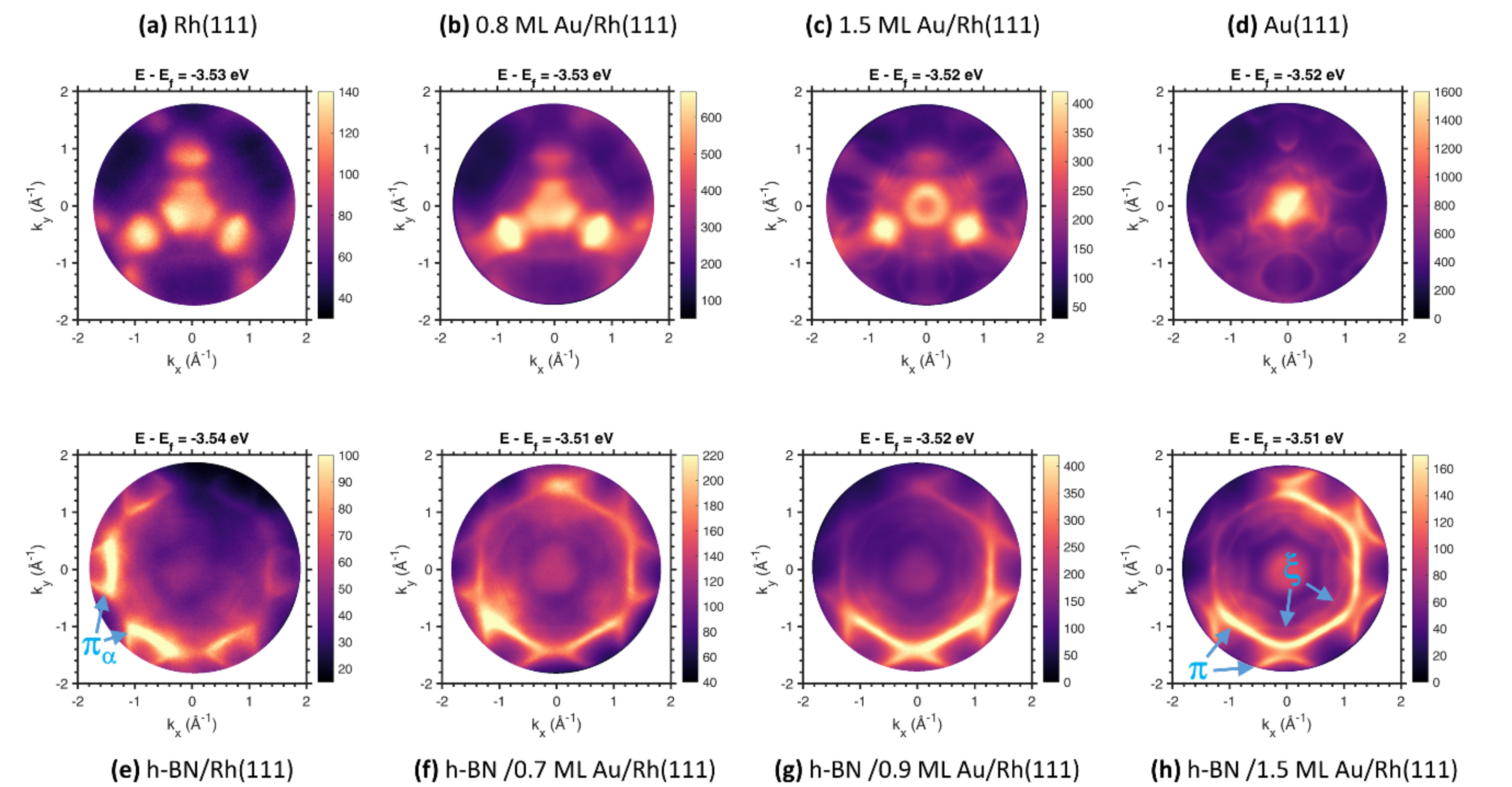
Energy slices at the Fermi level



* Energy slices at the Fermi level

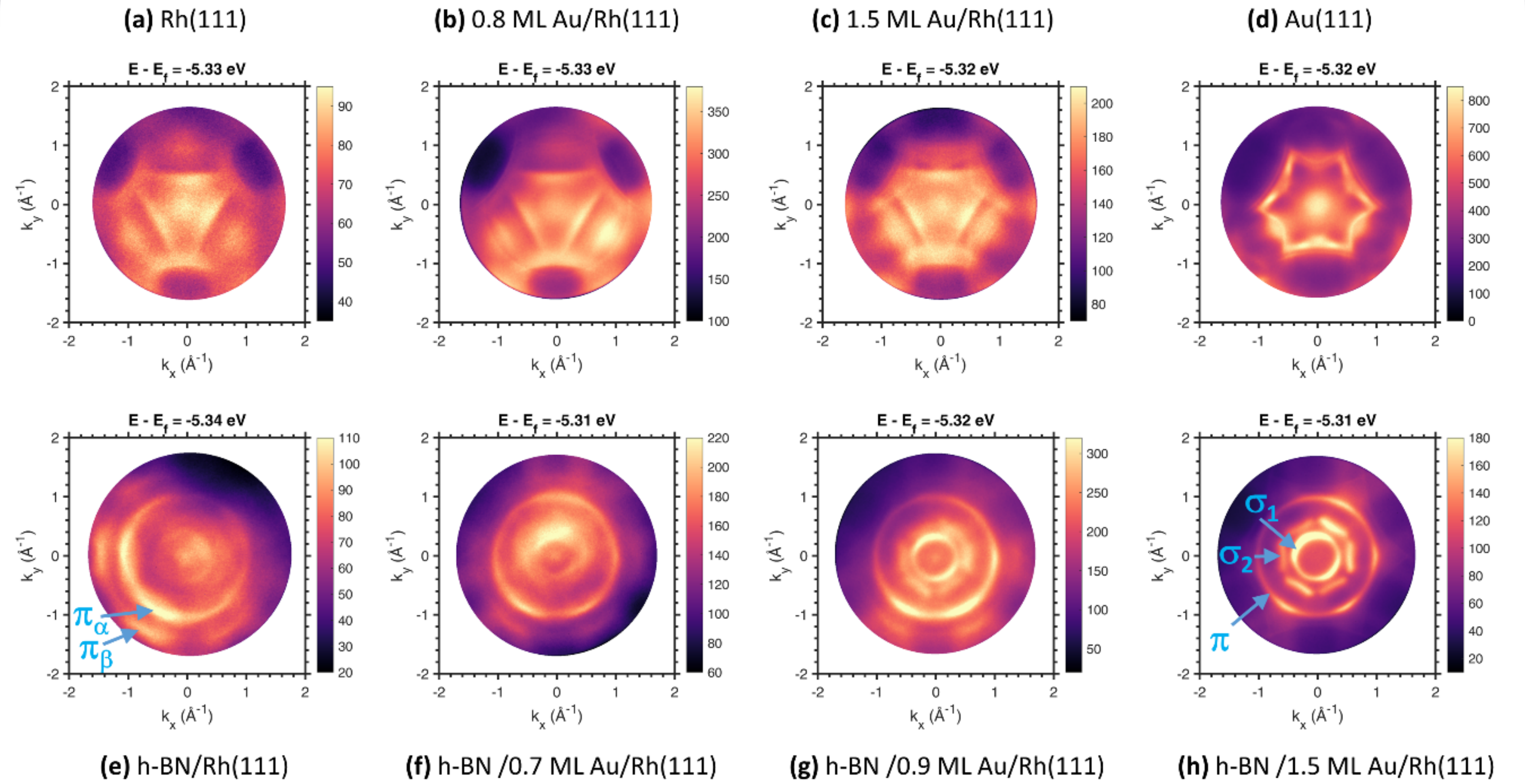


Constant energy slices I.



- The π band shifted toward the lower binding energies with increasing Au coverage (the triangles corresponds to π bands open and became larger)
- The appearance of the new ξ band also visible on the relevant constant energy slice (h-BN/1.5MLAu/Rh(111))

Constant energy slices II.

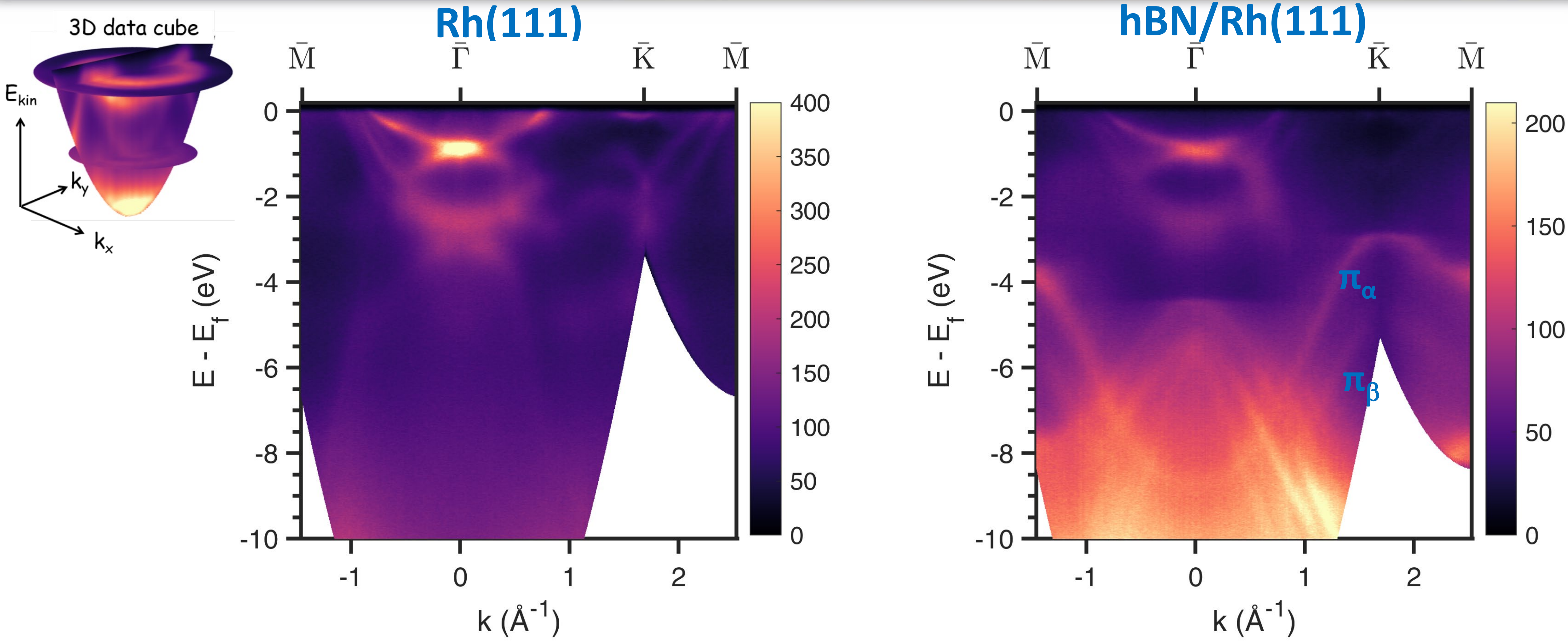


The strong interaction of the Rh and h-BN influences the appearance of bands:

- π_β (pore): strong interaction – dashed rings
- π_α (wire): weak interaction – continuous rings

The stronger hybridization resulted in stronger changes in the appearance of the bands.

h-BN on Rh(111): consequences of nanomesh morphology



The twofold character of the **nanomesh surface (pore and wire regions)** results in the doubling of h-BN bands: **two vertically shifted π bands** are observed. This is in accordance with Greber et al. Surf. Sci. 672-673(2018) 33.

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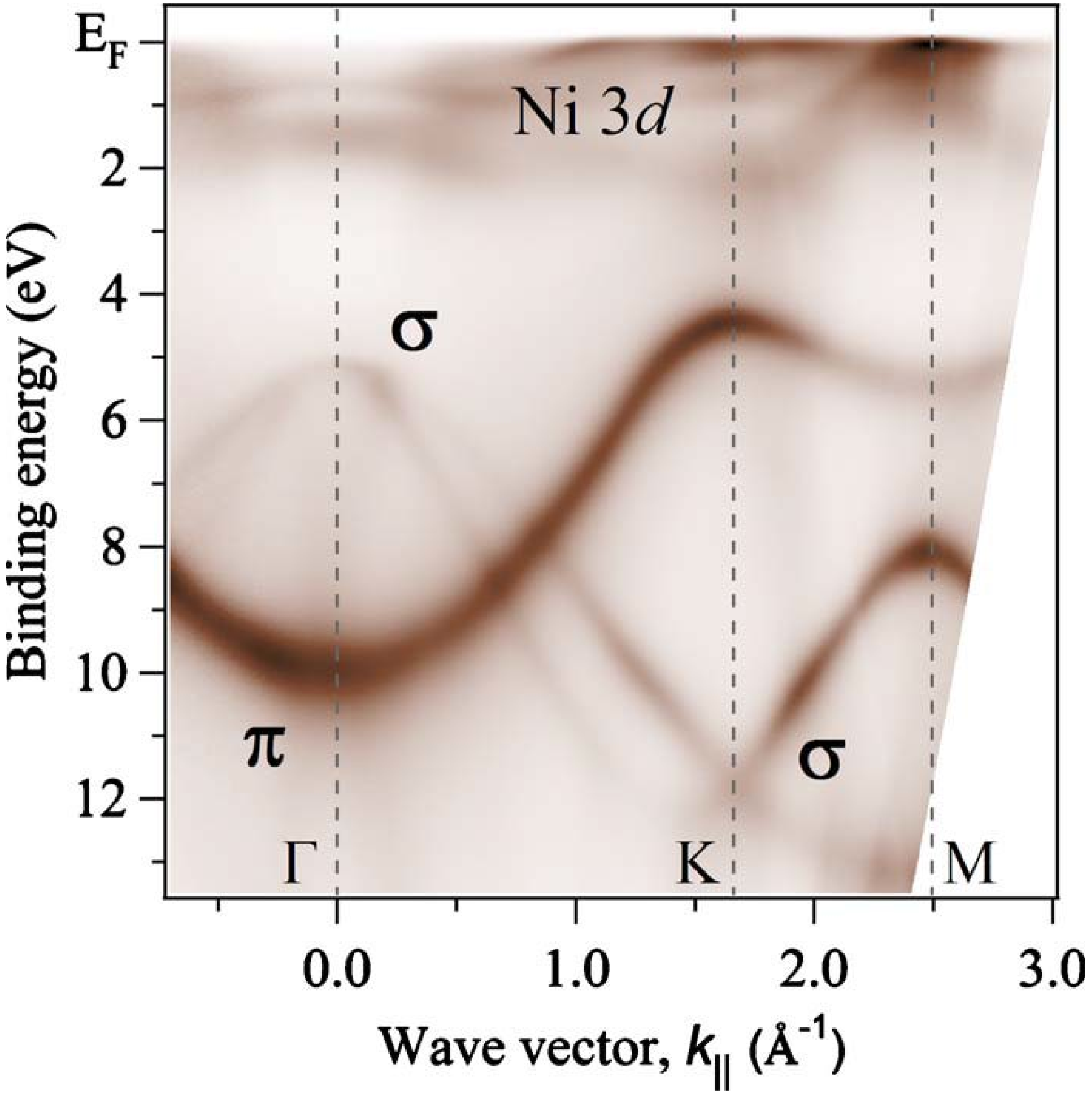

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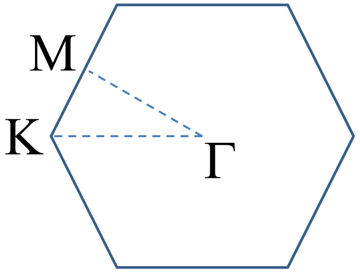
- Flat h-BN is characterized by one π band and two σ bands.



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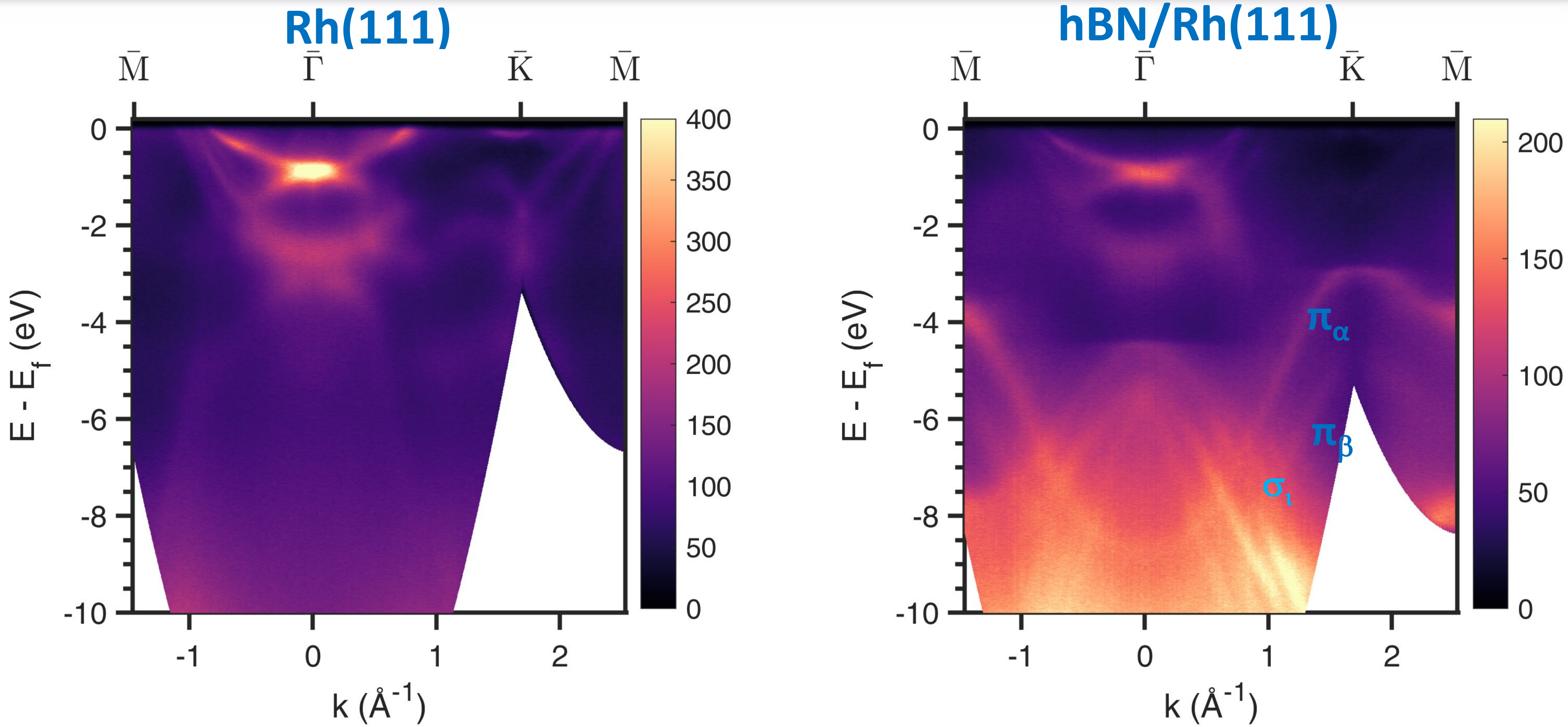

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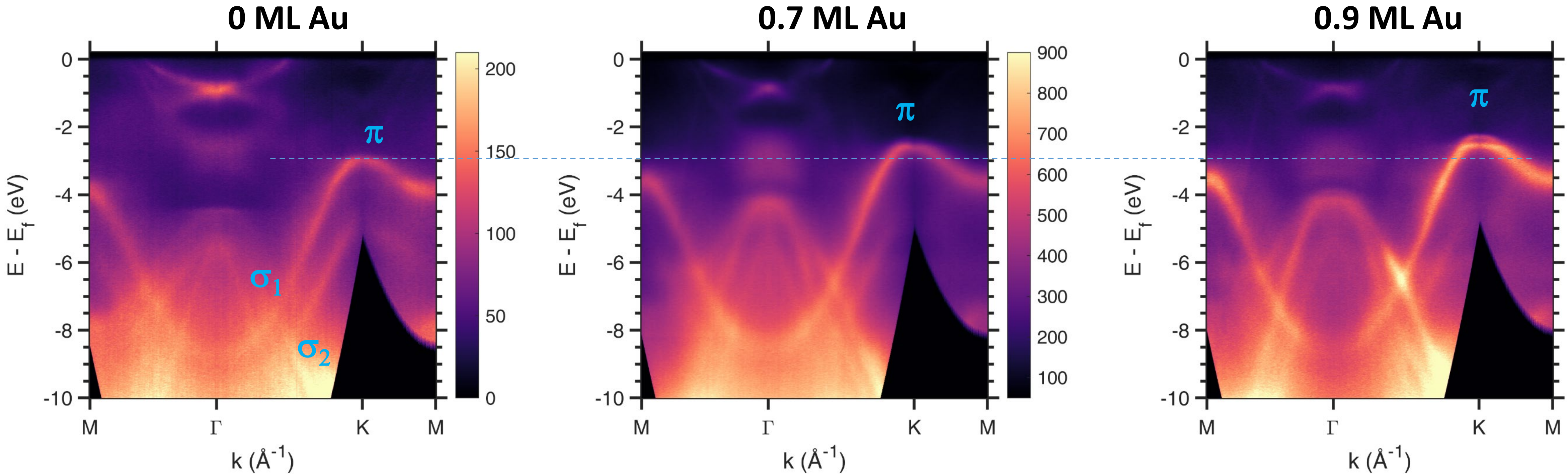
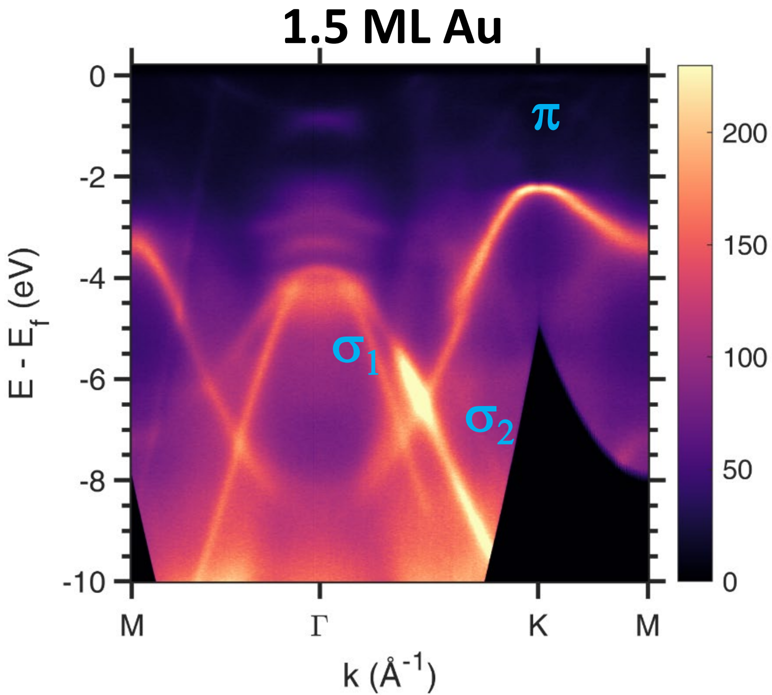
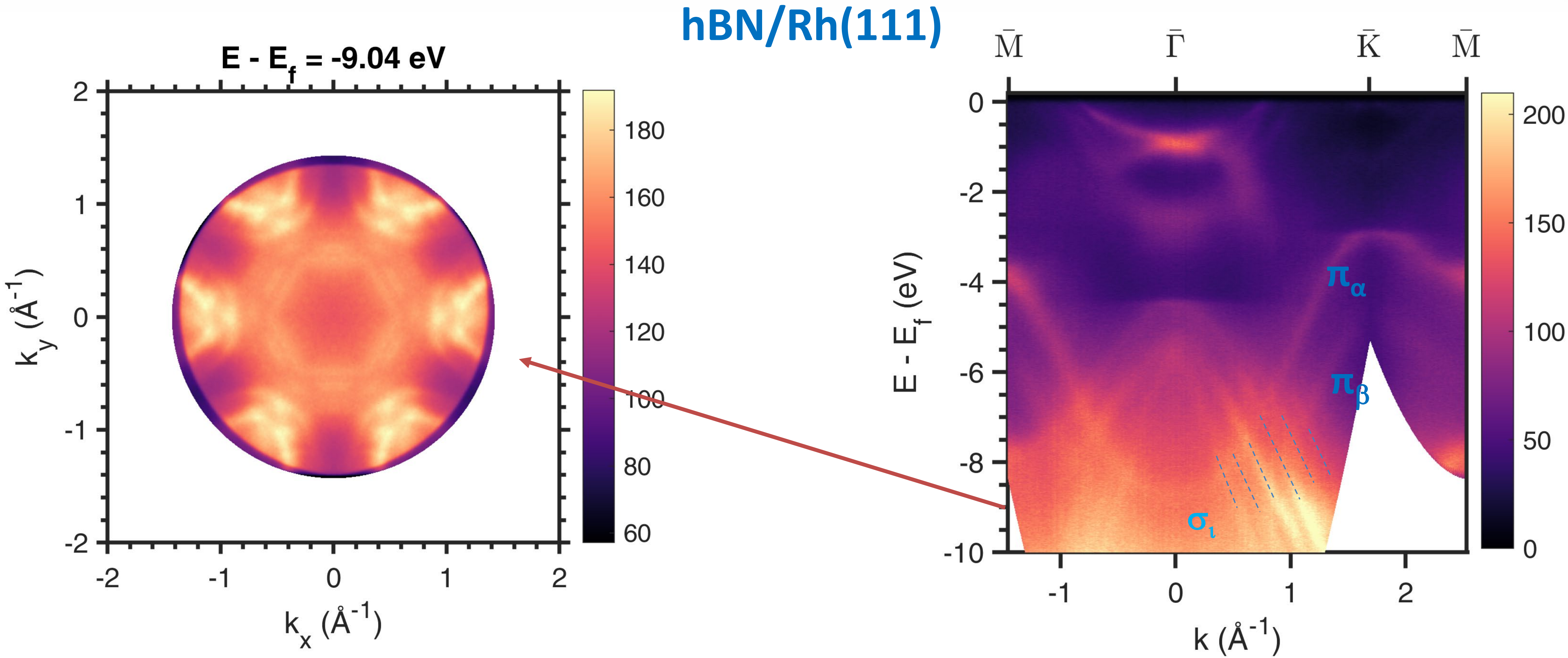


h-BN/xAu/Rh(111) surfaces

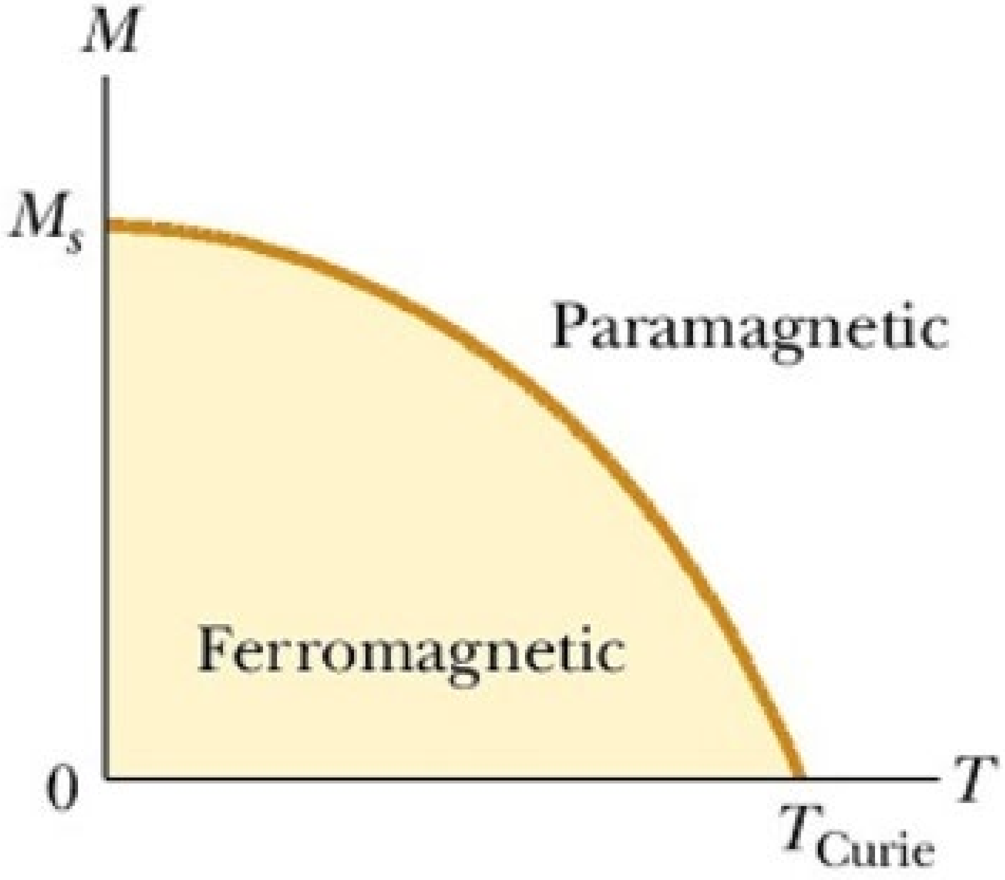


- The pore-wire duality, is present in the band structure also at Θ_{Au}=0.7 ML, and at Θ_{Au}=0.9 ML, but vanishes at Θ_{Au}=1.5 ML Au, when the h-BN monolayer is completely flat according to STM.
- The π and σ band-splitting also observable at intermediate Au coverages: 0.7 & 0.9 ML, respectively. The π band shifted toward the lower binding energies with increasing Au coverage.

h-BN on Rh(111): consequences of nanomesh morphology



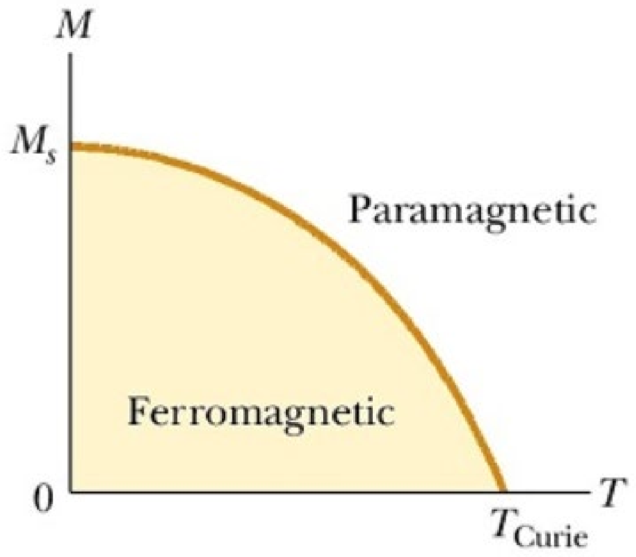
Pore-wire duality was observed also for the **σ bands**, but those are split into even more **branches (~6):** replica bands as observed on h-BN/Ir(111), e.g due to photoelectron diffraction on the superlattice (Usachov et al. Phys. Rev. B 86, 155151 (2012)).

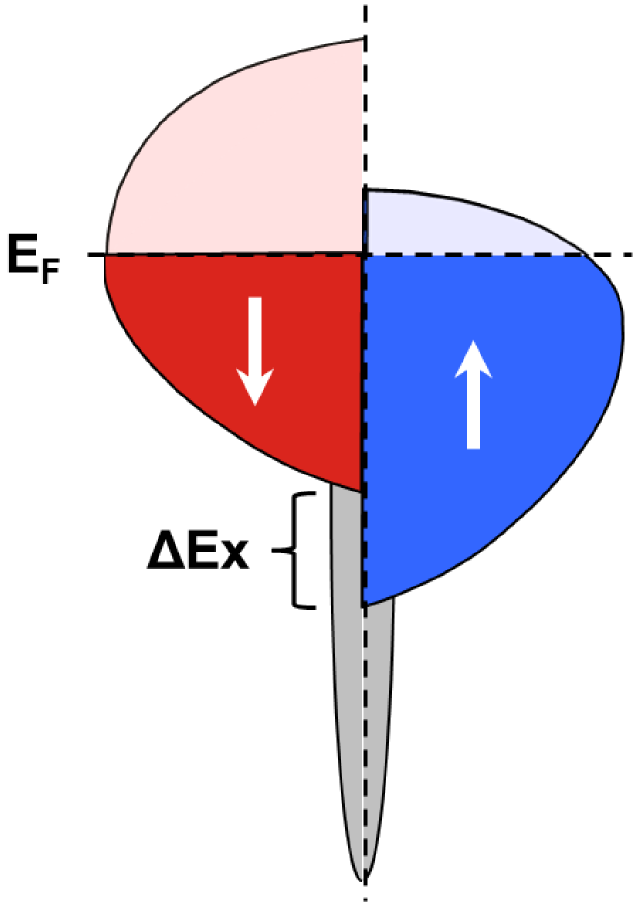


$$\mathbf{M} \propto \frac{\mathbf{N}^{\uparrow} - \mathbf{N}^{\downarrow}}{\mathbf{N}^{\uparrow} + \mathbf{N}^{\downarrow}}$$

Stoner versus Heisenberg

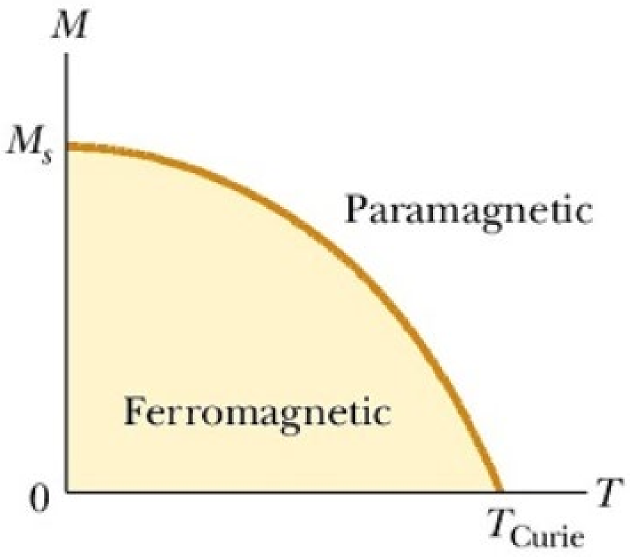
Conventional demagnetization: spin-dependent band structure



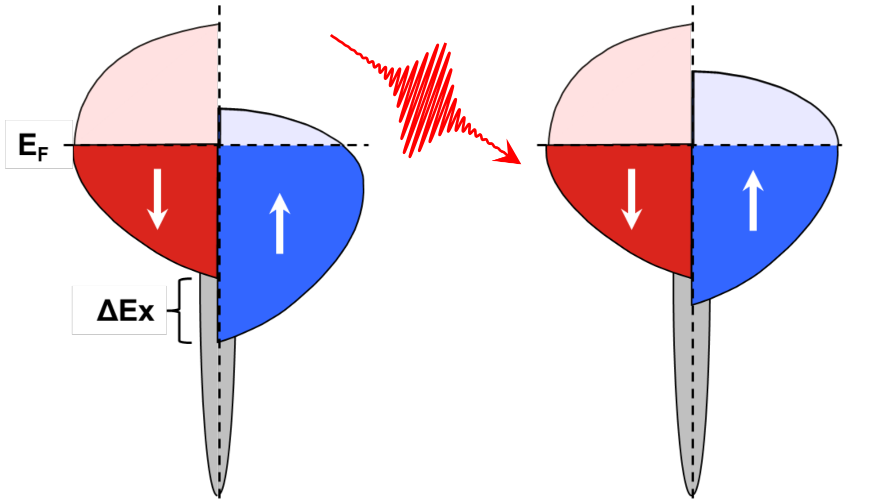


$$\mathbf{M} \propto \frac{\mathbf{N}^{\uparrow} - \mathbf{N}^{\downarrow}}{\mathbf{N}^{\uparrow} + \mathbf{N}^{\downarrow}}$$

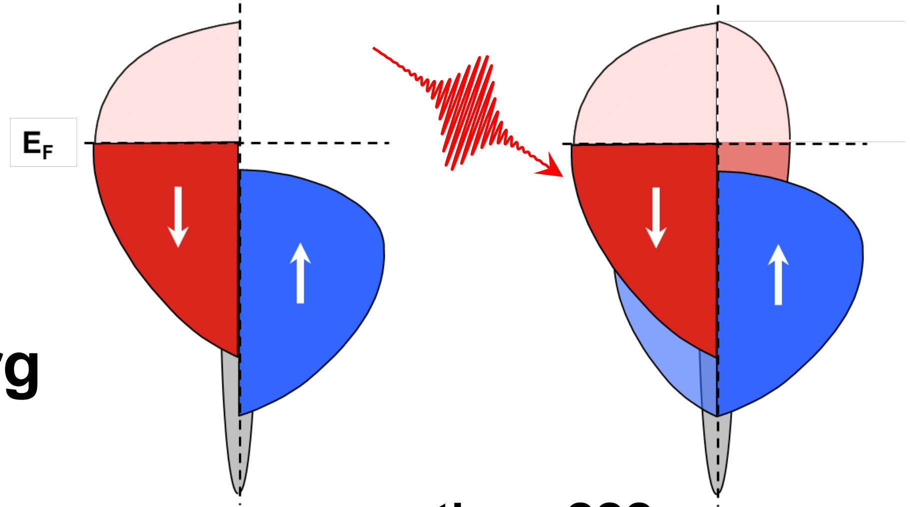
Conventional demagnetization: spin-dependent band structure



Reduction of ΔEx



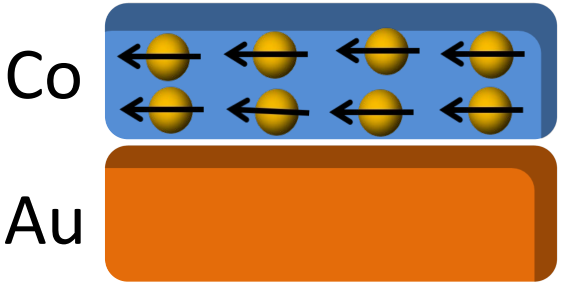
Mirroring of the band structure

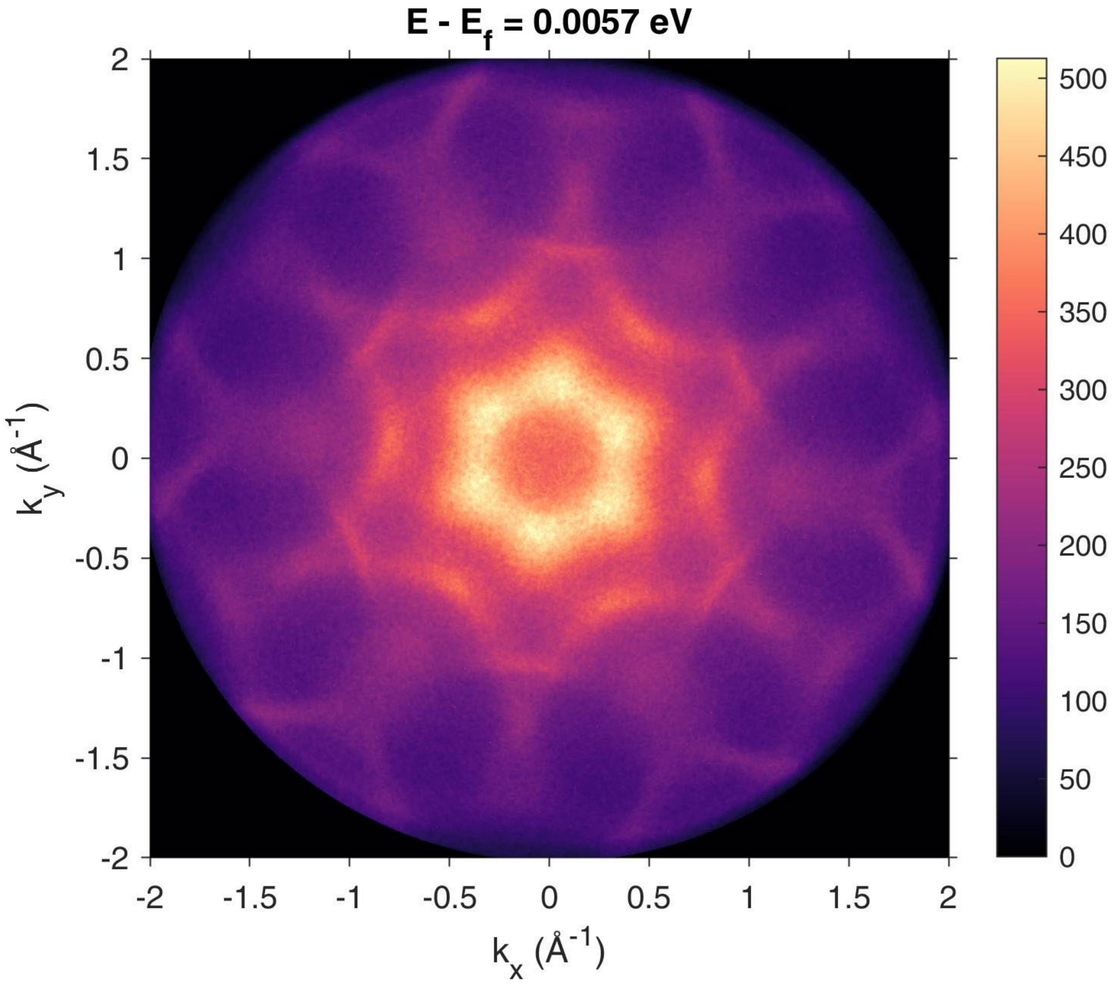


Stoner versus Heisenberg

or others ???

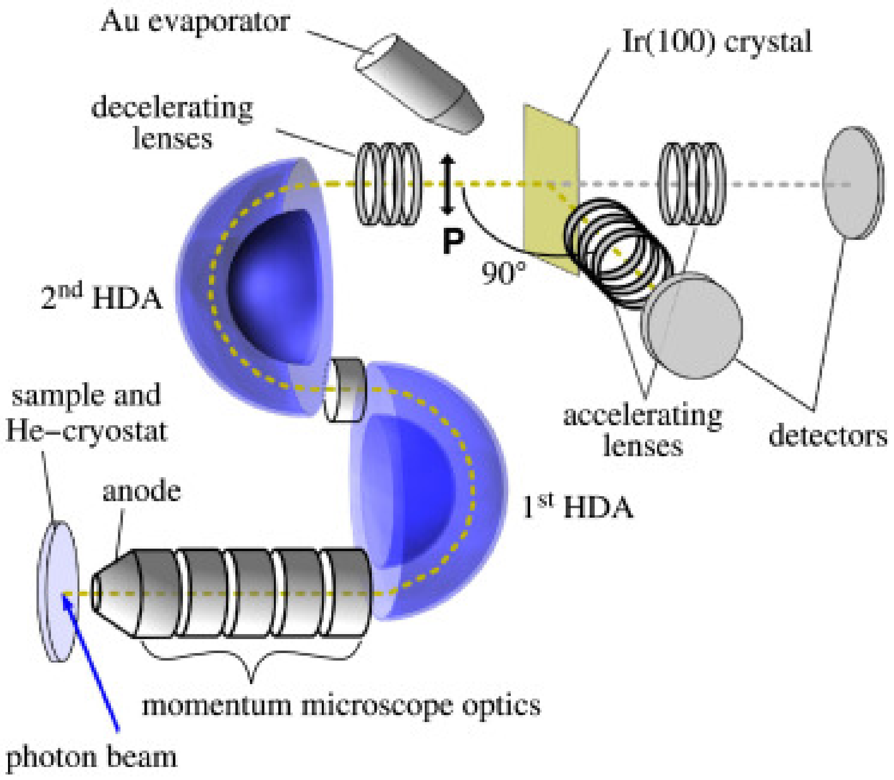
Momentum Microscopy Mode





T ~ 120K, VUV He I Symmetrized (3 fold) CBE near E_f.

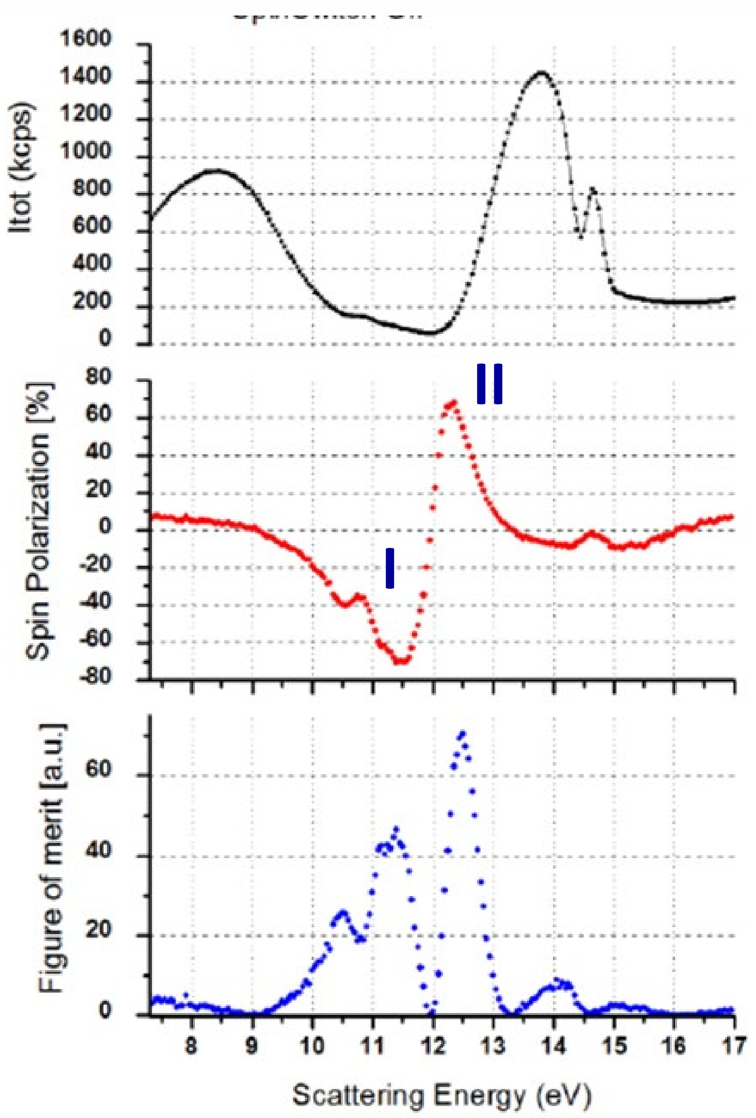
2D Spin Filter



- A monolayer gold on a Ir(100) single crystal.
- Spin polarizing mirror to detect the lateral spin-polarization of either an real or k-space image. (Spin quantization axis **P**)
- Opposite sign of Sherman-functions at two working points for imaging.

C. Tusche et al. Ultramicroscopy, ***159****, 520-529 (2015)*

Spinfiltereffect: Two working points I and II



Reflection (scattering yield)

Spin Asymmetry in Elastic Electron Scattering

Figure of Merit

Technical Reference Manual of 2D Imaging Spin Filter, FOCUS

Spin-resolved PEEM in *Real Space*

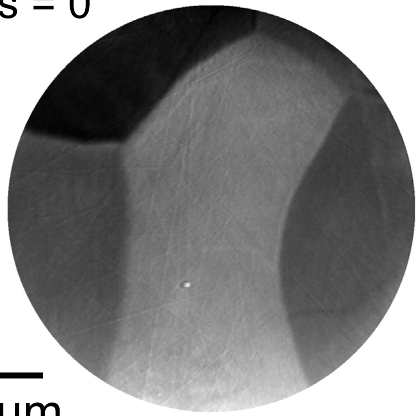
Experiment

I_1, $s_1 < 0$

I_2, $s_2 > 0$

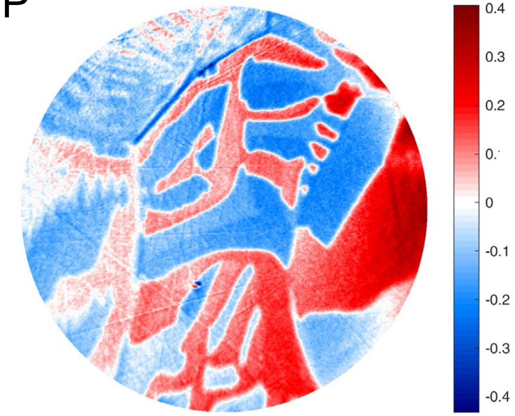
Computation

I_0, s = 0



30 µm

P



Work flow:

I_1 I_2

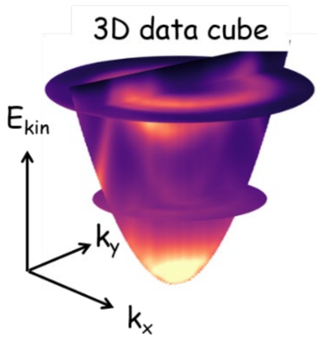
$$I_i = I_0 R_i (1 + s_i * P)$$

I_0 P

$$I_{\uparrow,\downarrow} = \frac{I_0}{2}(1 \pm P)$$

$I_\uparrow$ $I_\downarrow$

Spin-resolved PEEM in *Momentum Space*



I_1 I_2

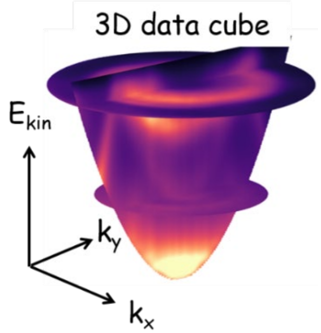
$$I_i = I_0 R_i (1 + s_i * P)$$

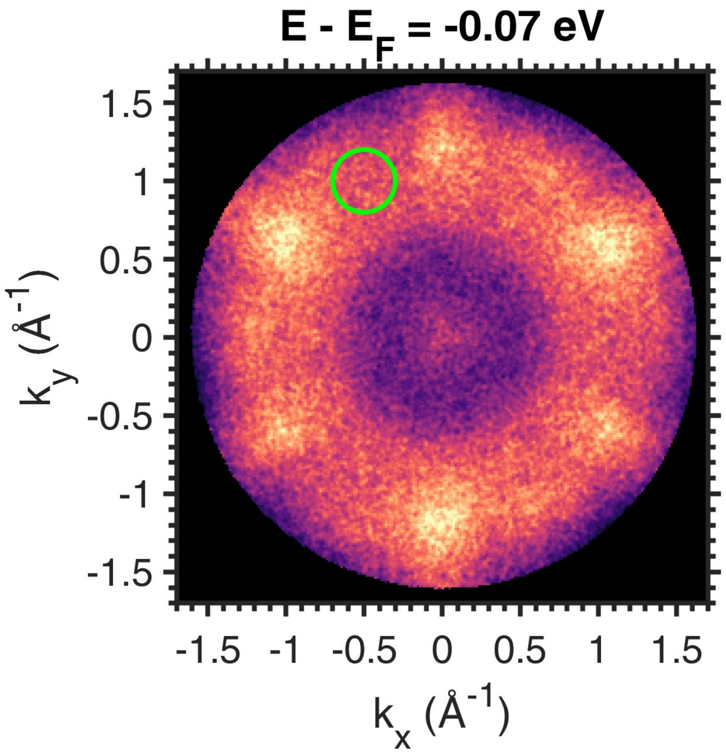
$\mathbf{I_0}$ **P**

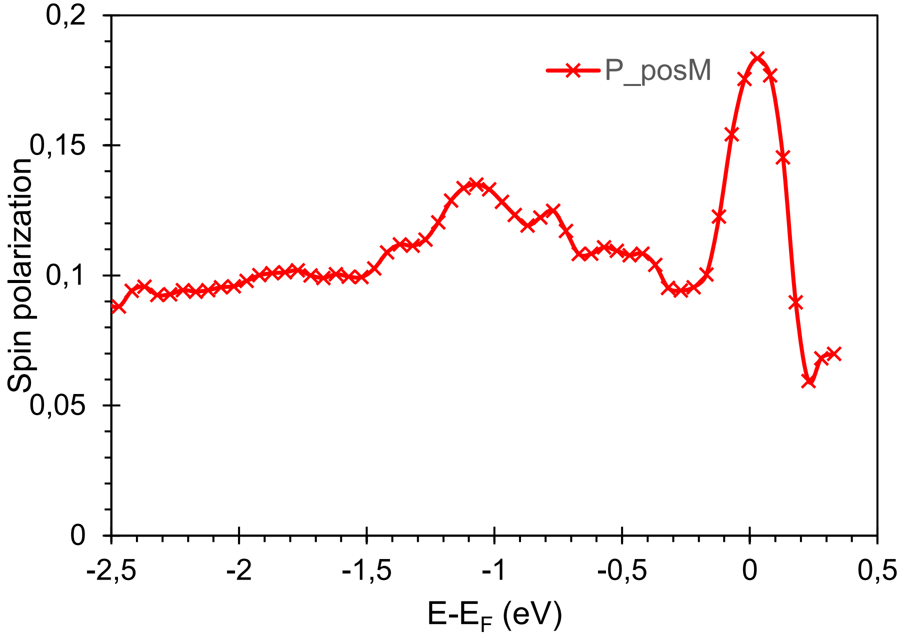
$$I_{\uparrow,\downarrow} = \frac{I_0}{2}(1 \pm P)$$

$\mathbf{I_\uparrow}$ $\mathbf{I_\downarrow}$

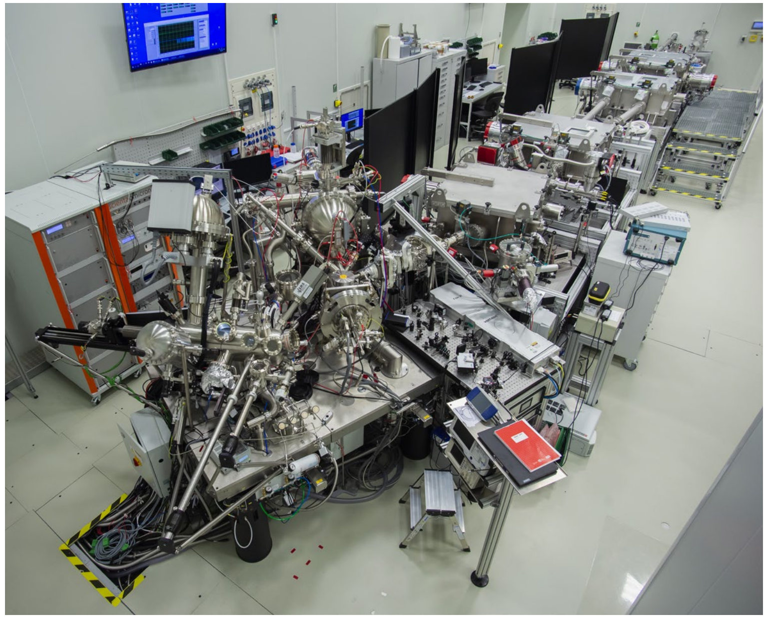
Spin-resolved PEEM in *Momentum Space*







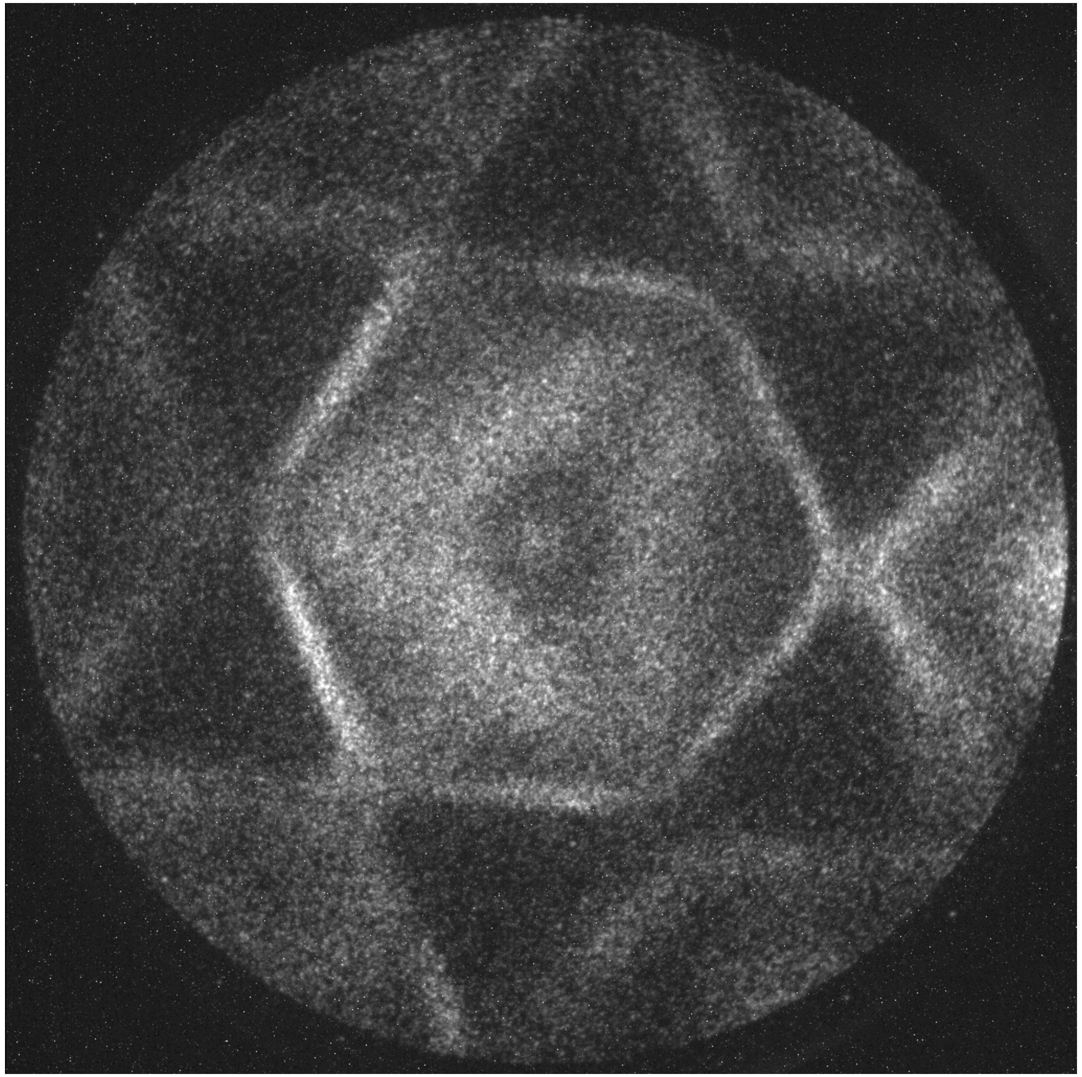
23. September 2022



Finally, there is HHG light!!!

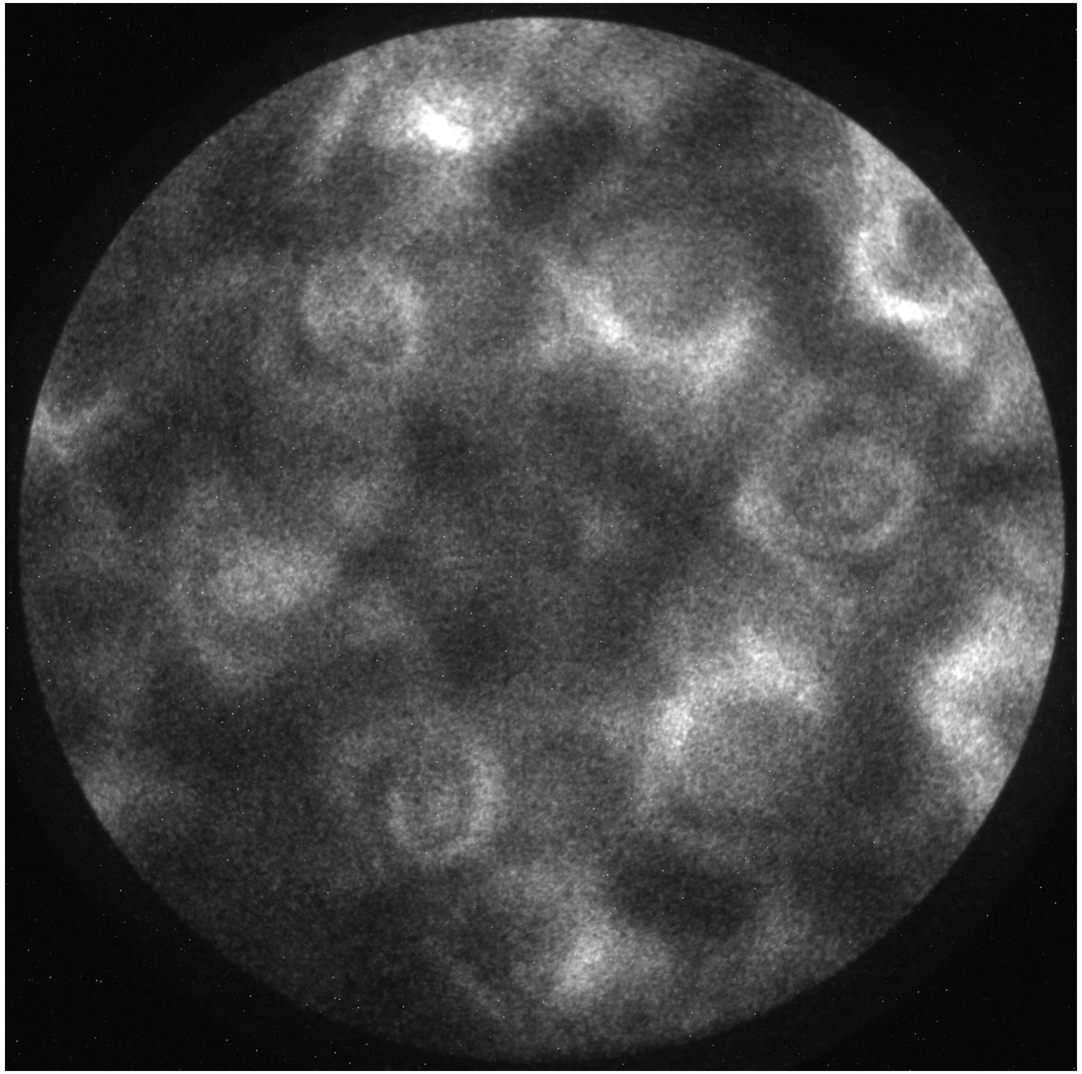
K-space images by NanoESCA using XUV source (33th harmonic) of the GHHG beamline

Au(111) near Fermi surface



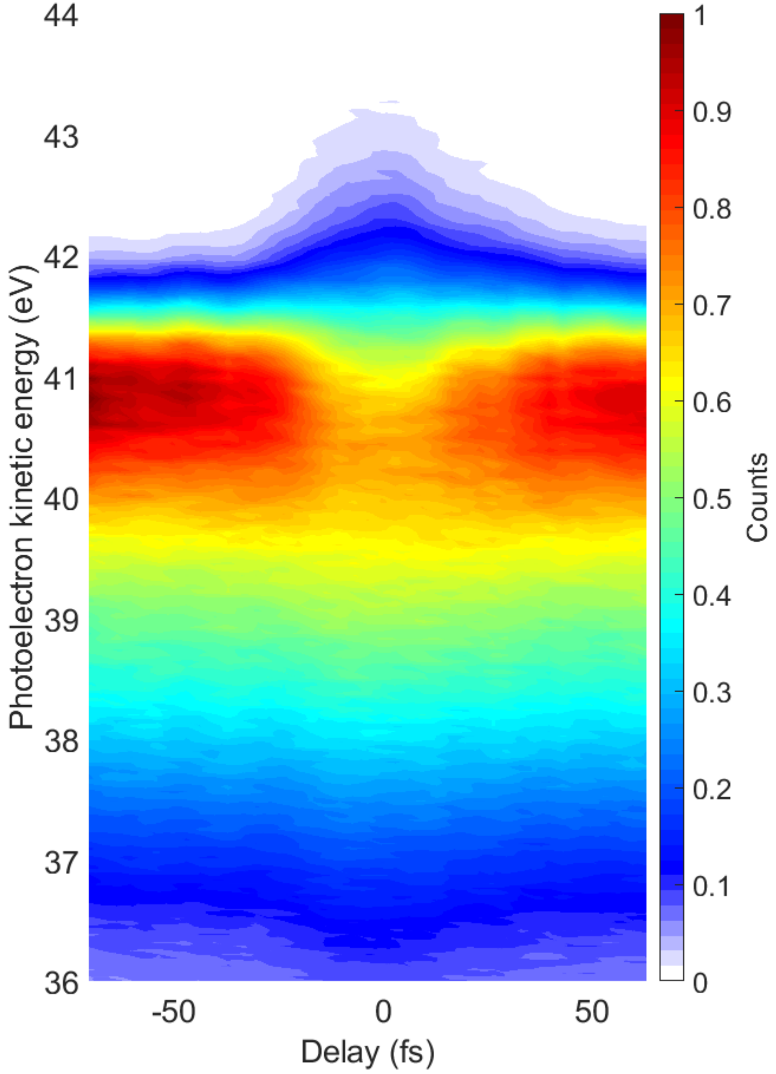
Exposure time: 10 min

Rh(111) near Fermi surface



Exposure time: 6.7 min

Time-zero on Rh(111)



- NIR pump (1.2 eV, 12-15 fs) and XUV probe (~42 eV, ~50-60 fs) were applied.
- The XUV monochromator was used without pulse front tilt compensation to achieve higher photon flux.
- Time zero was found using the TOF analyzer of NanoESCA in spectroscopy mode.

Area normalized TOF spectra as a function of delay. Kinetic energy is referred to the Fermi level.

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