UNIVERSITY OF GRAZ

PEEM: a tool for accessing electronic structure, spin texture and ultrafast dynamics of the interface.

Giovanni Zamborlini

July 10th, 2025

Young Research Leaders Group Workshop - WASEM winery, Ingelheim.

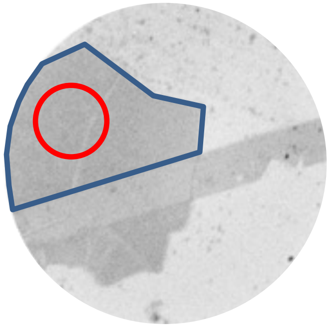
Why do we need PEEM for 2D materials?

PEEM: PhotoEmission Electron Microscopy
It combines **SPECTROSCOPY** and **MICROSCOPY**

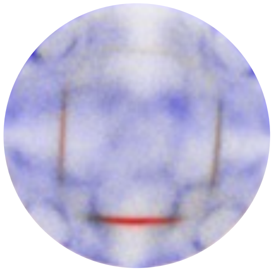
We can access chemical, electronic and magnetic properties of 2D materials with the same instrument.

<u>(some of the possible) Applications:</u>

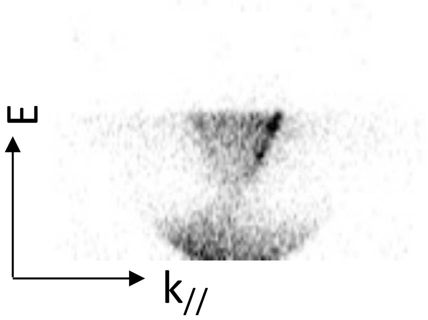
μ-probing



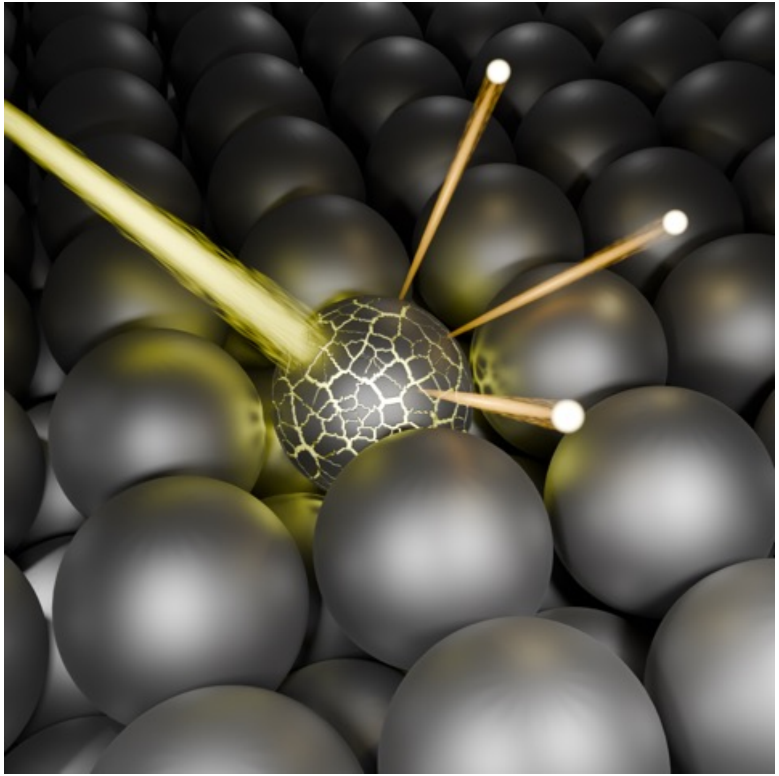
Spin-resolved band structure mapping



Time-resolved Photoemission



The photoelectric effect



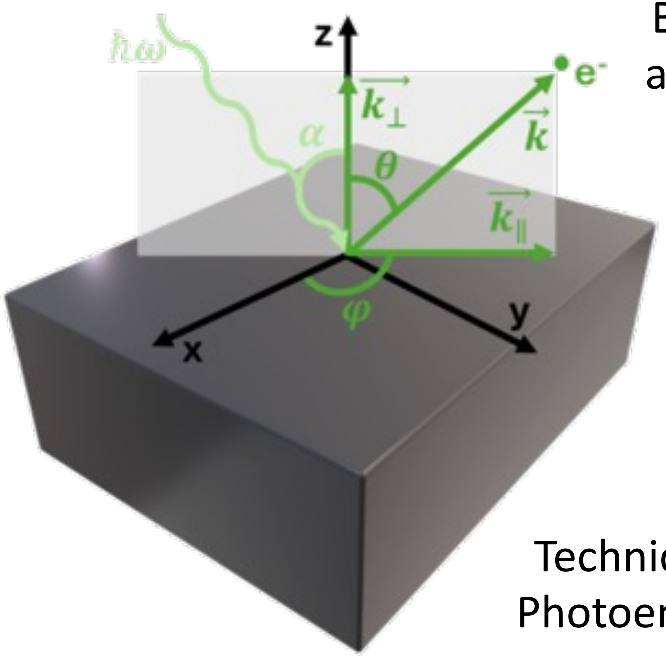
Photon energy

e⁻ Binding Energy

$$E_{kin} = h\nu - \phi - E_B$$

Work function

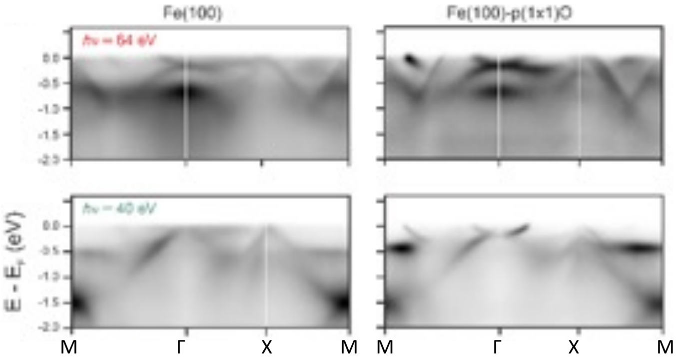
Photoemission Spectroscopy - ARPES

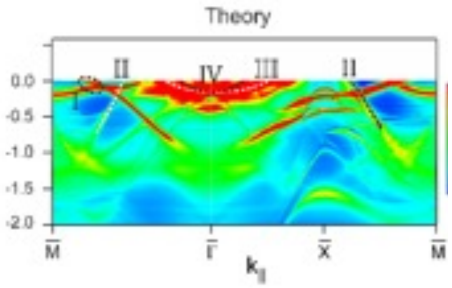


By measuring the take-off angle of the photoemitted electron…

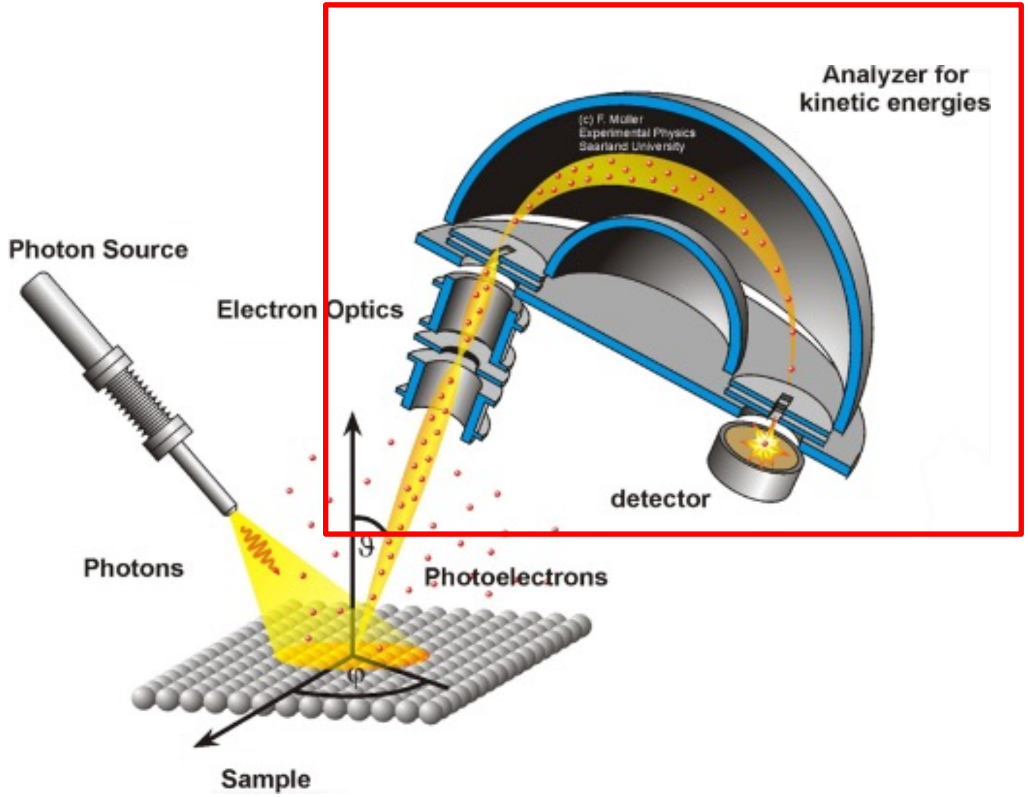
… We can probe the material's band structure

Technique: Angle-Resolved Photoemission Spectroscopy (**<u>ARPES</u>**)

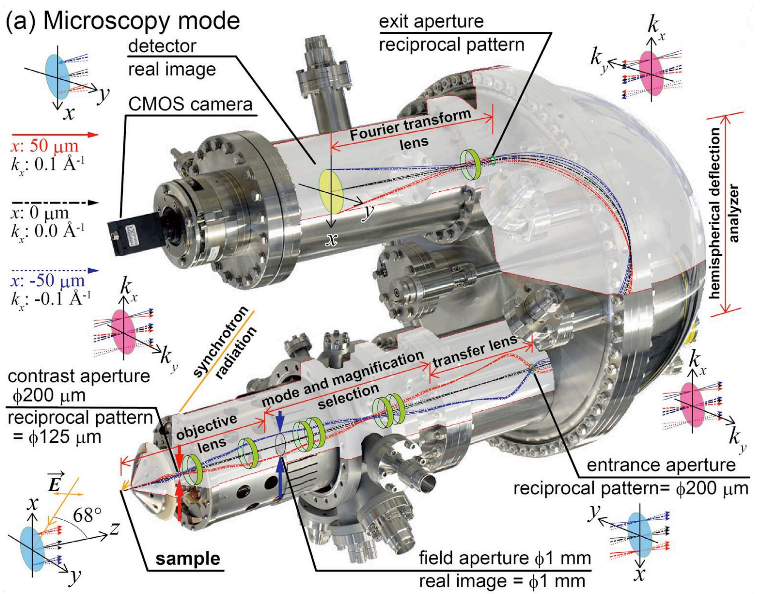




Photoemission Spectroscopy

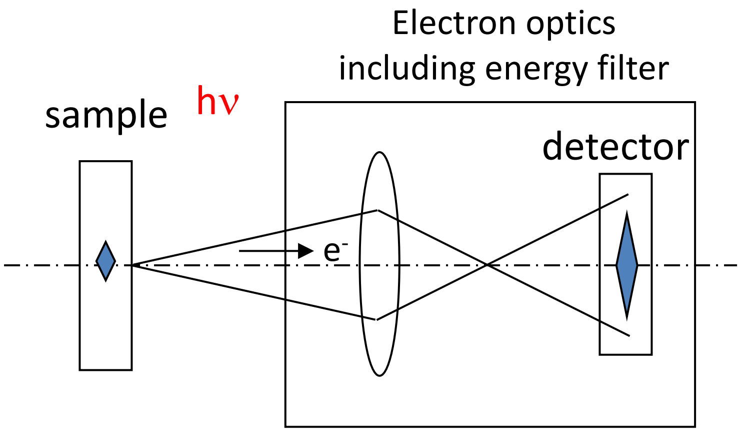


PEEM



F. Matsui et al. *Japanese J. of Appl. Phys.* **59**, 067001 (2020).

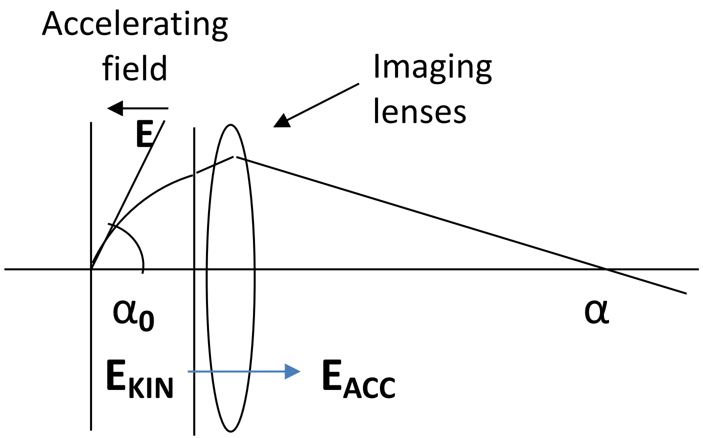
PEEM - Principle



Direct imaging → **Parallel detection**

Lateral resolution → 10-50 nm
(depending on the electron optics)

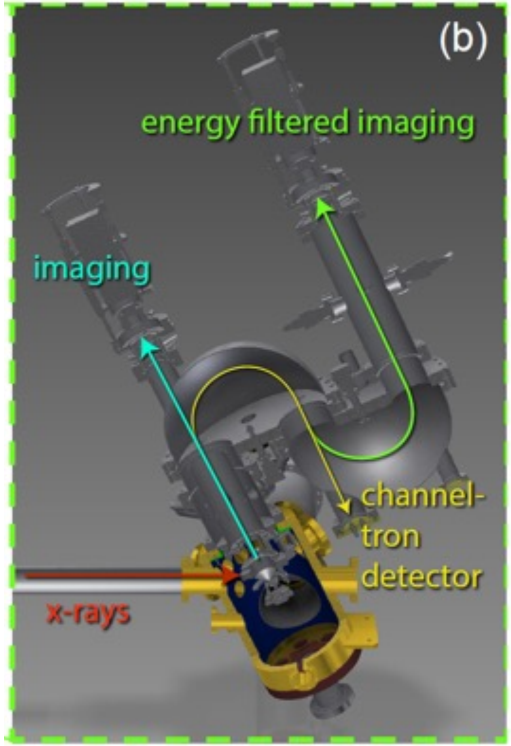
Immersion objective lens
or cathode lens



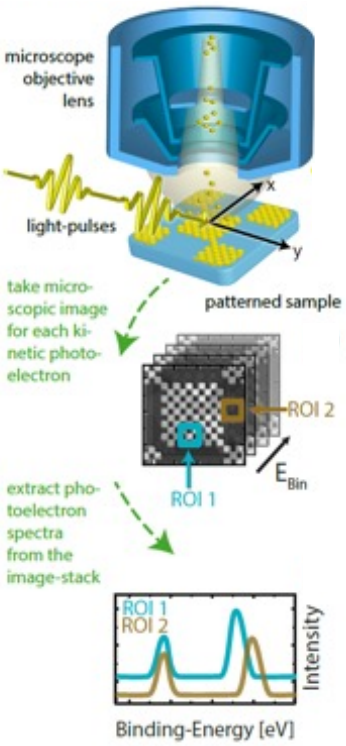
$\sin \alpha / \sin \alpha_0 = \sqrt{E_0/E}$
Example for E_{ACC} = 20000 eV:

E_{KIN}	2 eV	200 eV
α for α_0 45°	0.4°	4.5°

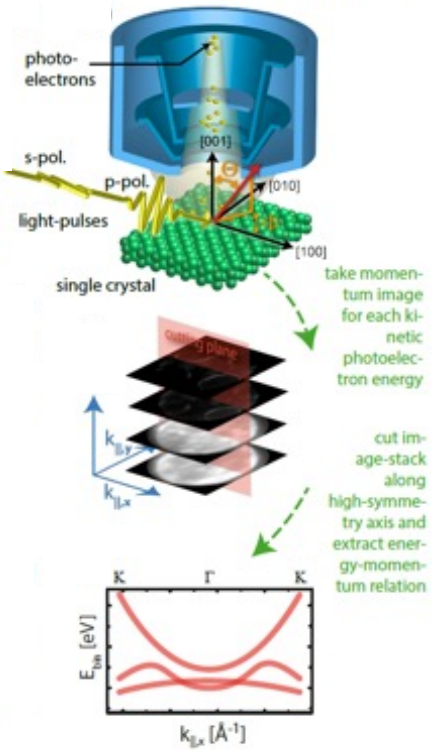
Operation modes



Imaging mode



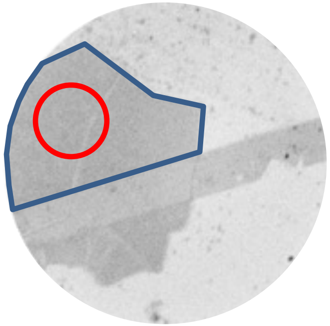
Momentum mode



What can we do with it?

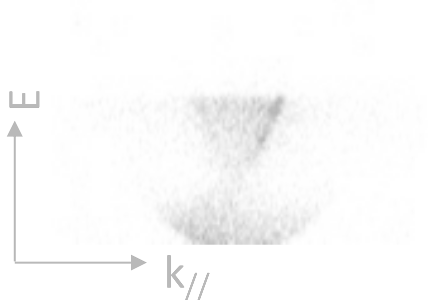
(some of the possible) Applications:

μ-probing



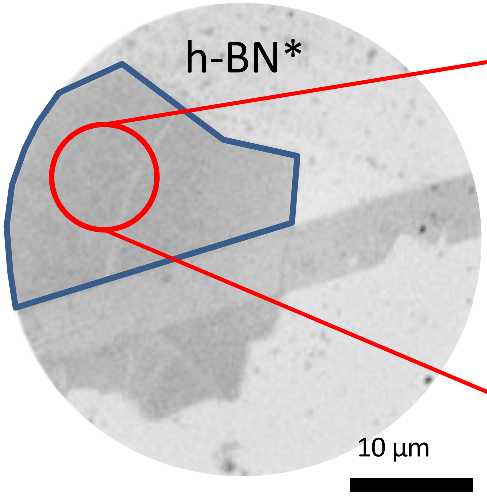
Spin-resolved band structure mapping

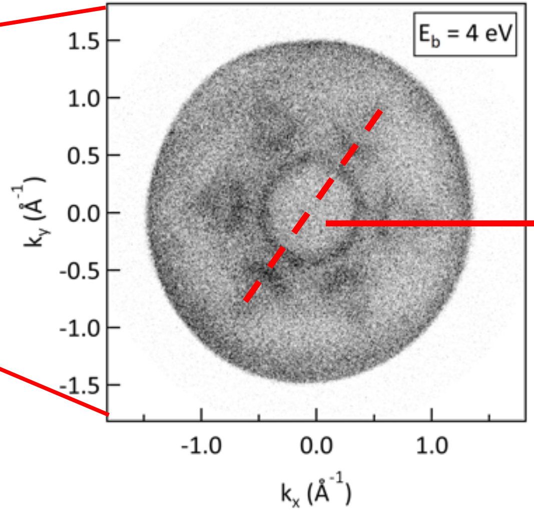
Time-resolved Photoemission



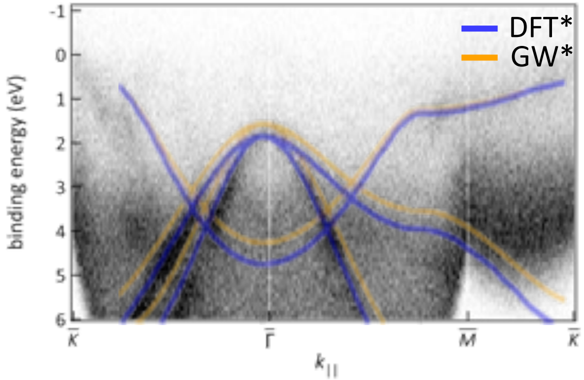
Combining imaging and momentum

Imaging mode → Momentum mode





ARPES



Real space → I(x;y)

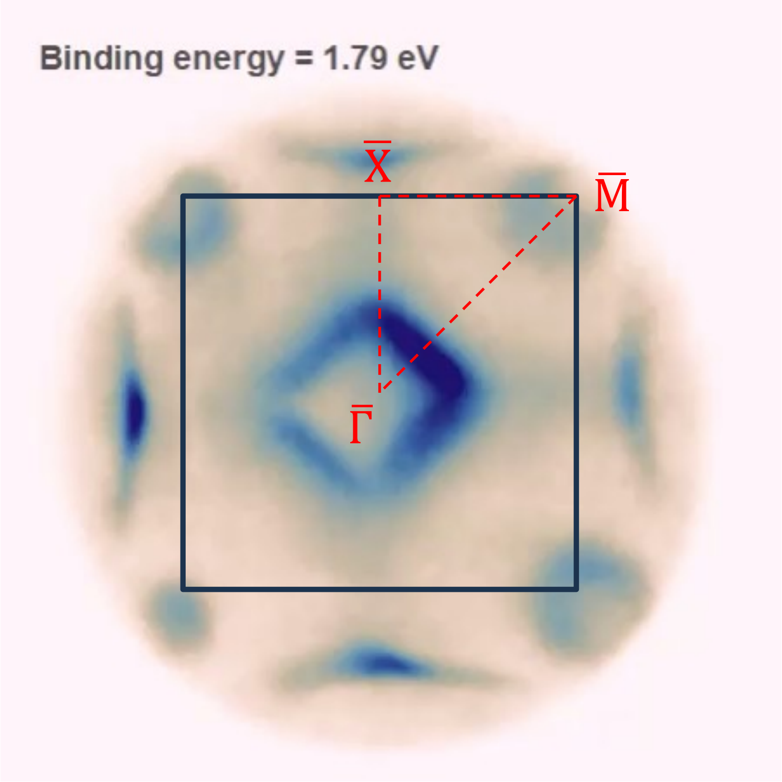
Reciprocal space → I(k_x;k_y)

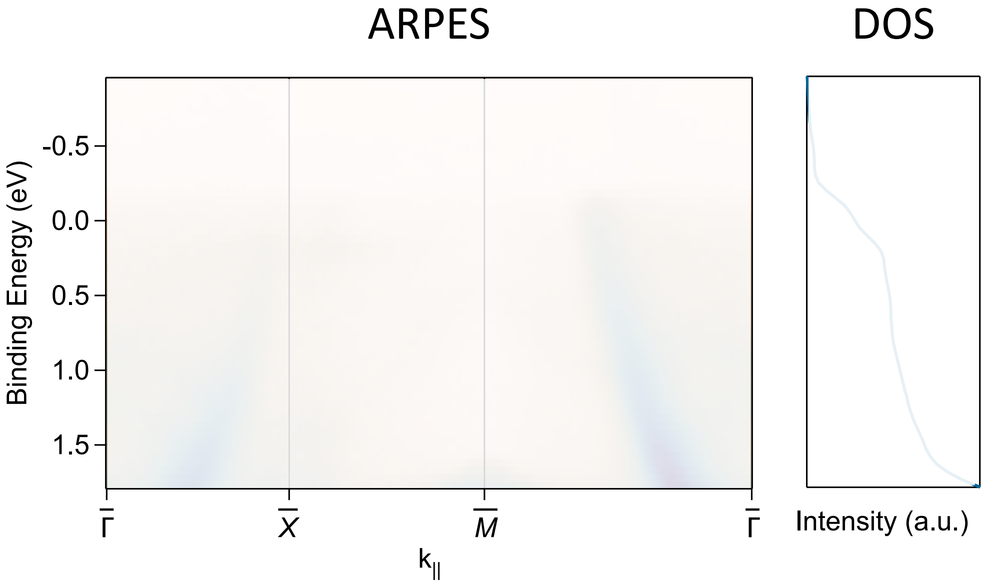
*h-BN courtesy of S. Mañas-Valero, E. Coronado

*F. Ferreira et al. *J. Opt. Soc. Am. B* **36**, 3 674-683 (2019)

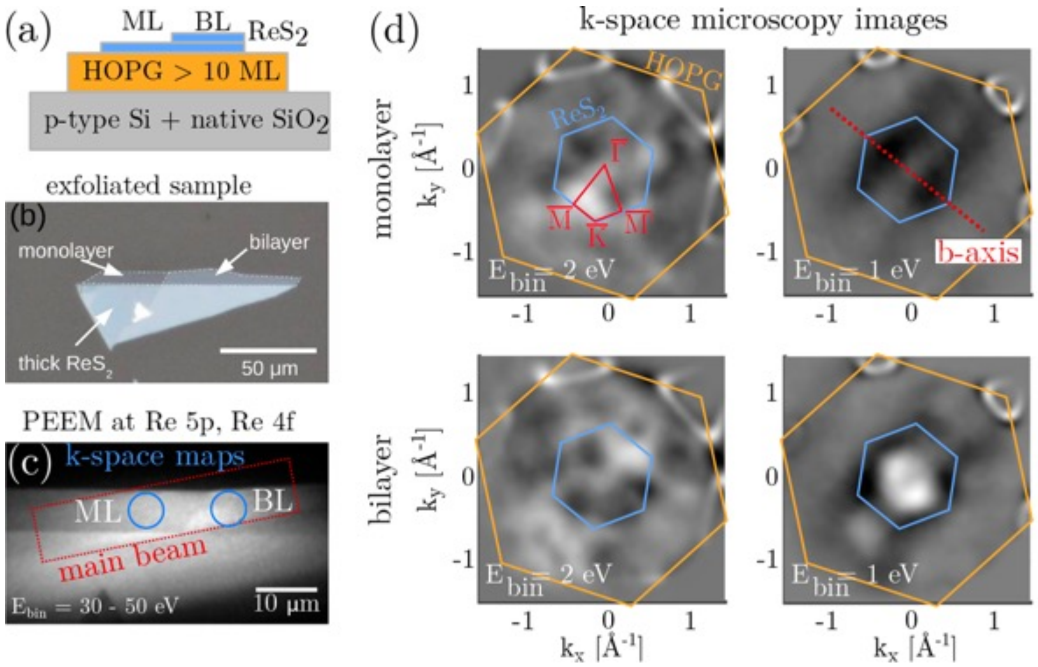
Practical example: Cu(001)

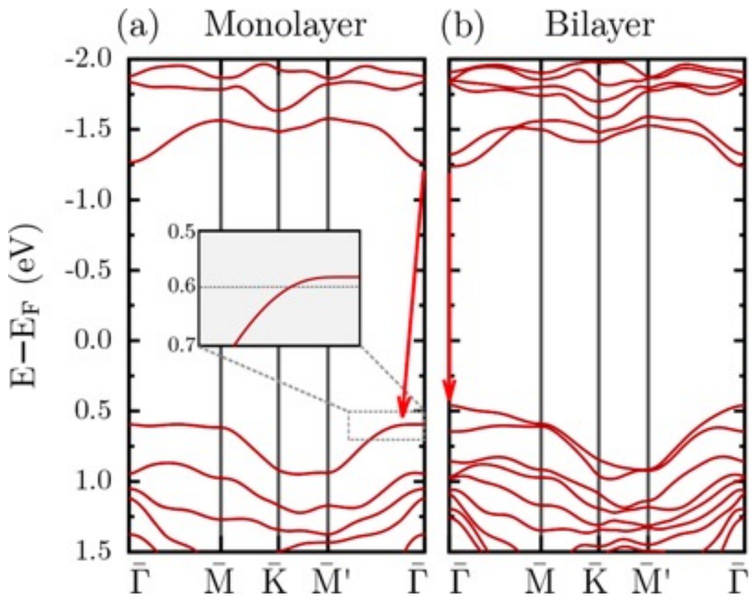
2D momentum map ($I(E,k_x;k_y)$)





ReS_2 – bandgap vs. thickness





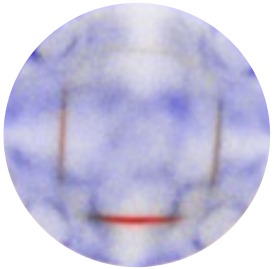
M. Gehlmann, .. GZ, … C. M. Schneider. *Nano Lett.* **17**, 5187–5192 (2017).

What can we do with it?

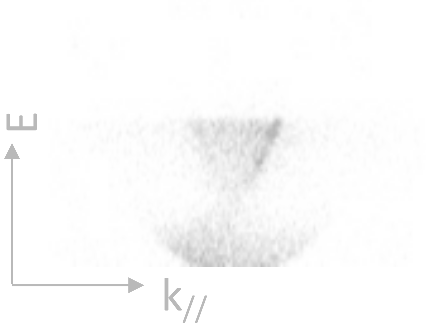
(some of the possible) Applications:

μ-probing

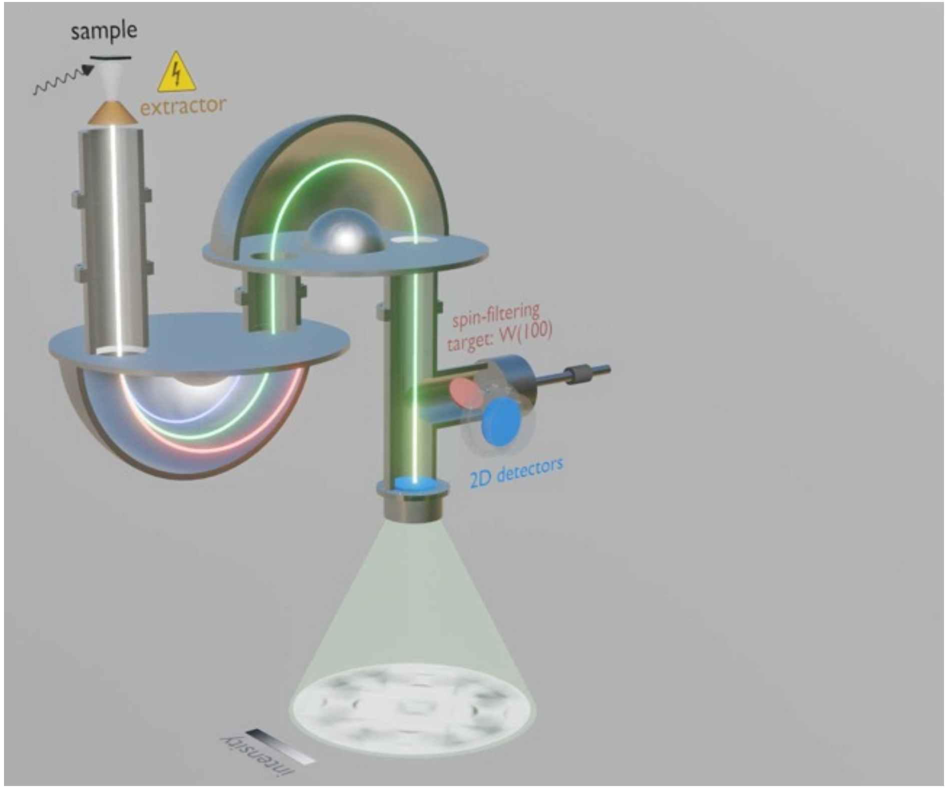
Spin-resolved Photoemission



Time-resolved Photoemission



Adding a spin polarimeter



Working Principle:

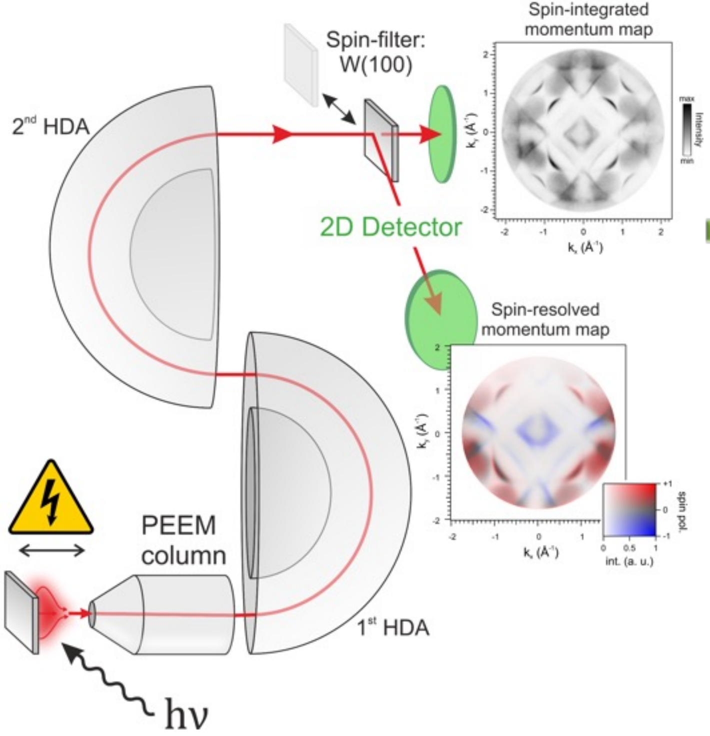
- Electron mirror:
  Scattering from from W(100)
- Scattering amplitude in the 0-order diffracted spot is spin dependent (spin-orbit coupling)

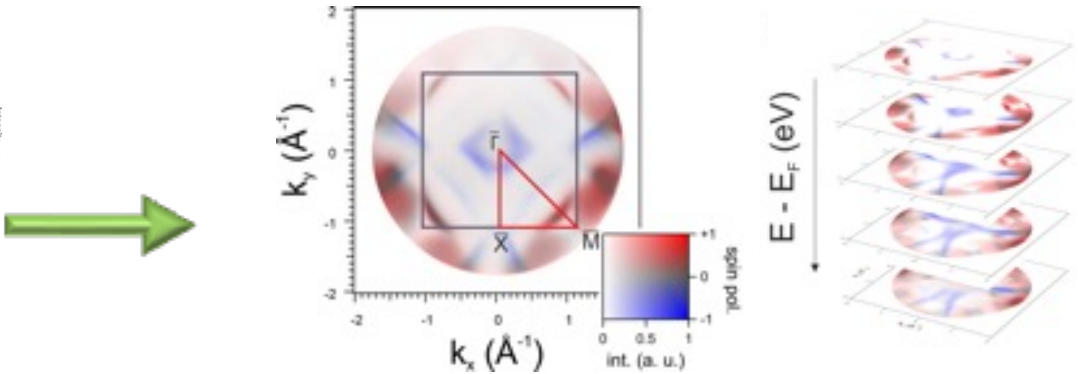
What we measure?

- The spin projection onto the vertical quantization axis

G. Zamborlini et al. *Synchrotron Radiation News* **37**, 24-29 (2024)

Spin-resolved momentum microscopy





4D data acquisition: $I_{PE}(E_{kin}, k_x, k_y, \sigma)$

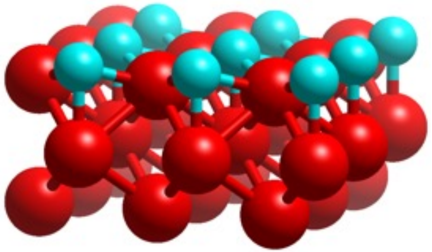
Access to

- **spin-resolved** surface **band structure**!

G. Zamborlini et al. *Synchrotron Radiation News* **37**, 24-29 (2024)

Model system: Oxygen-passivated Fe(100)

➔ long-range ordered superstructure Fe(100)-*p*(1x1)O

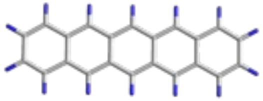


Ideal platform for
molecular/FM interfaces

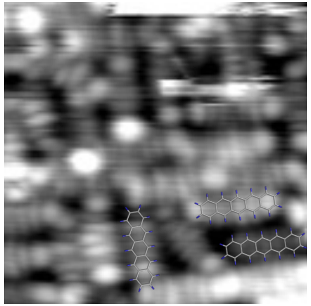
WHY?

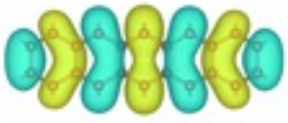
On bare FM substrates:
Strong molecule-surface interaction

Pentacene (5A)



5A/Fe(100)

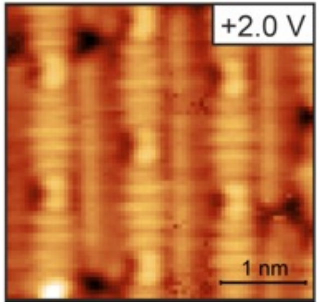




LUMO

→ **No self assembly**

5A/O-Fe(100)

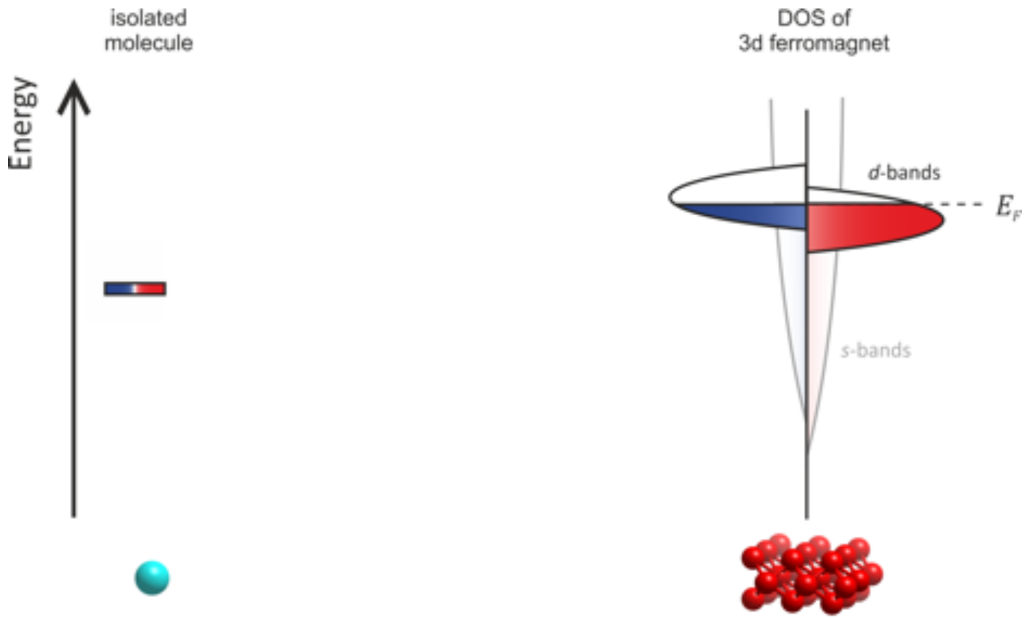


<u>Solution:</u>
Oxygen decoupling

How oxygen modifies the Fe properties?

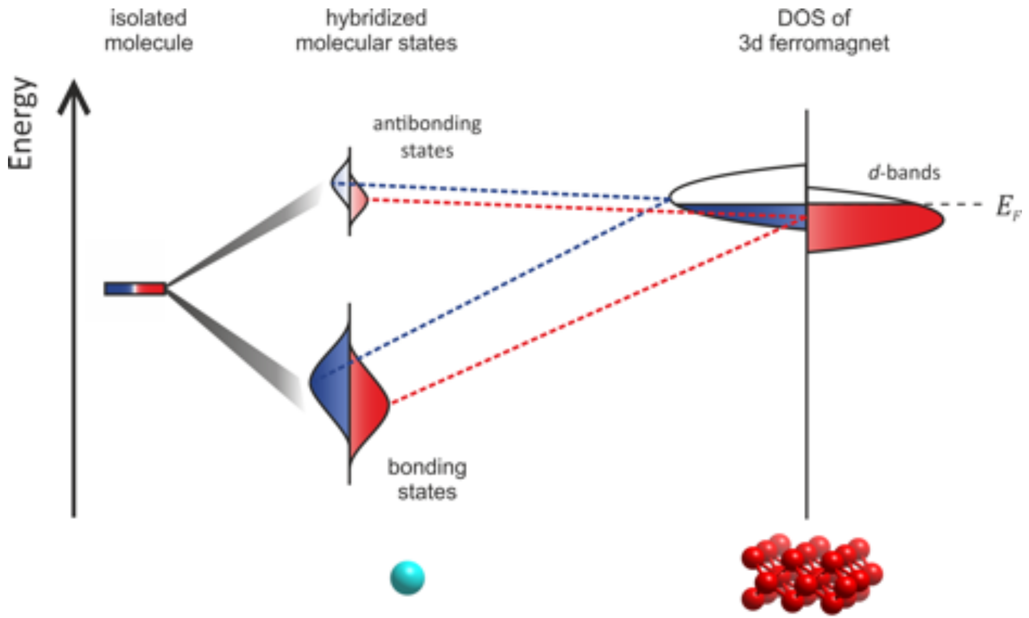
Oxygen on Fe

single particle model



Oxygen on Fe

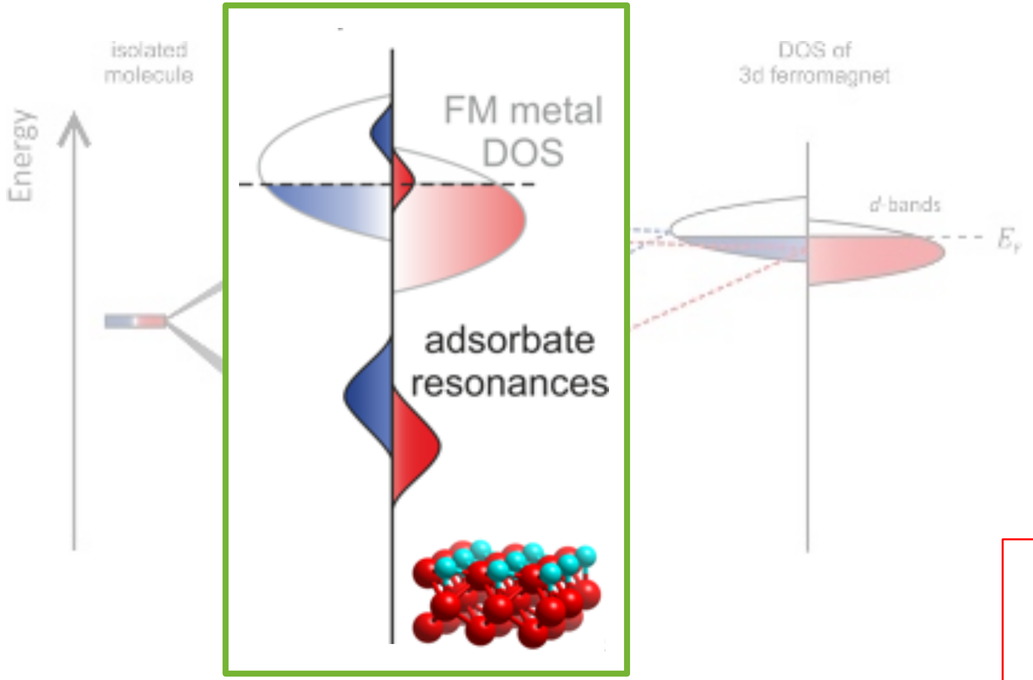
single particle model



B. Hammer and J. Norskov, *Nature* **376**, 238 (1995)
S. Bhattacharjee et al. *Sci Rep* **6**, 35916 (2016)

Oxygen on Fe

Hybrid Interface



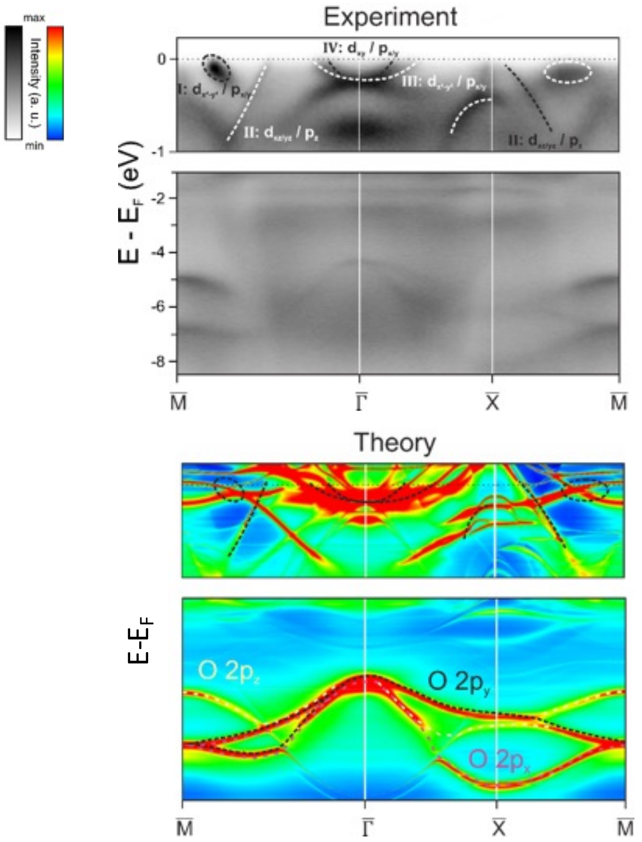
Single particle model (DFT):

- spin-split adsorbate states
- minor influence on the metal DOS

Is this really the case?

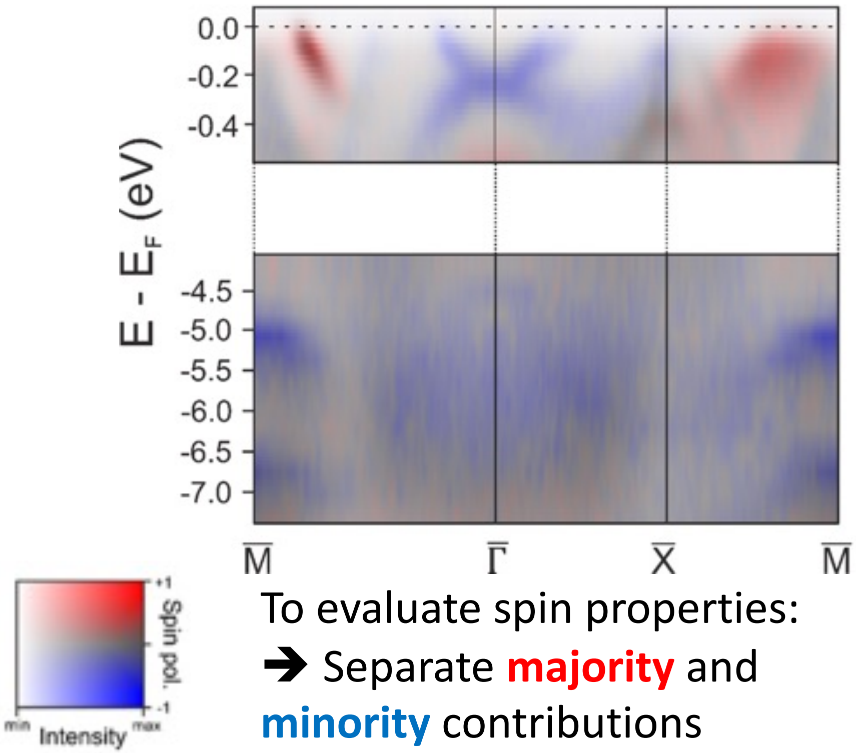
We need access to the spin properties of the interface

Spin-resolved band structure



Fe *d* bands + antibonding O states

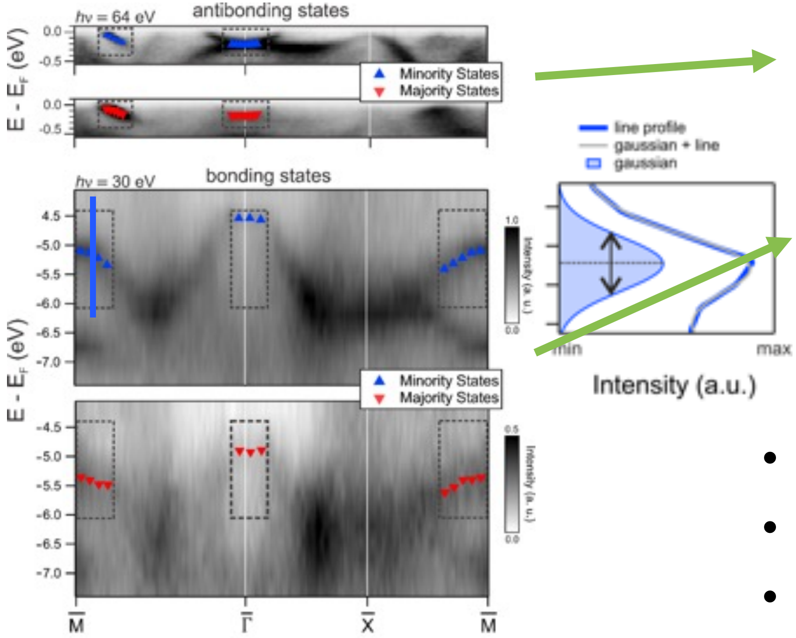
bonding O states

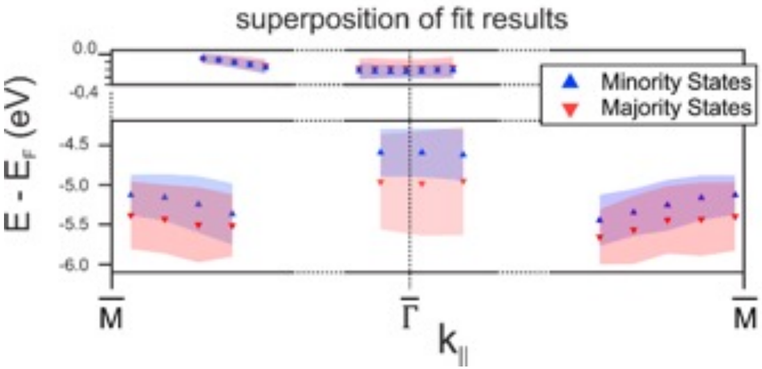


To evaluate spin properties:
➔ Separate **majority** and **minority** contributions

D. Janas et al. *Adv. Mater.* **35**, 2205698 (2023)

Spin-resolved band structure





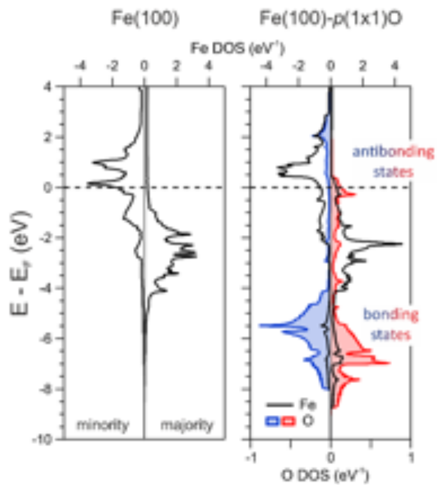
- Exchange splitting at E_F drastically reduced
- Spin splitting of bonding states: $\Delta \approx 0.3$eV
- Broadening of majority states

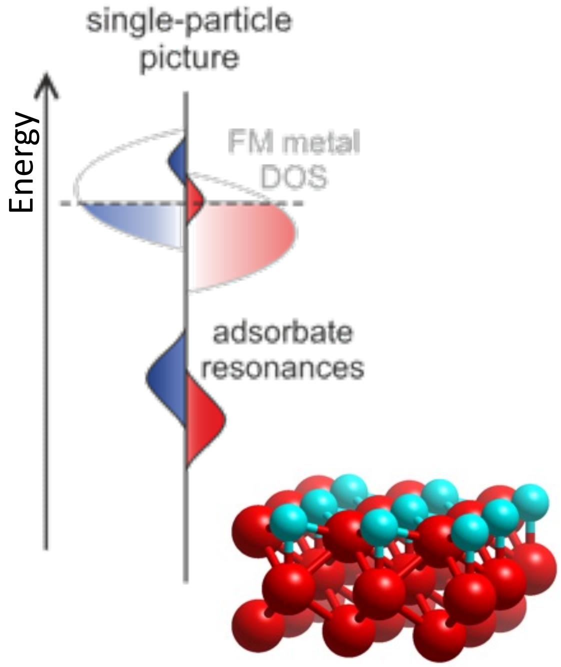
➔ Does theory describe these traits?

D. Janas et al. *Adv. Mater.* **35**, 2205698 (2023)

Impact of electron correlation

DFT





Enhancement of **many body interactions**:

- Strongly reduced exchange splitting at E_F
- Narrowing of the Fe d-bands

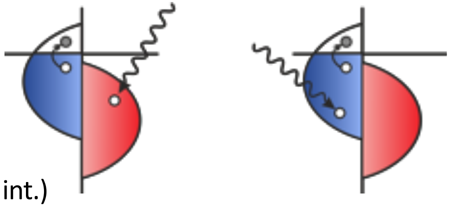
D. Janas et al. *Adv. Mater.* **35**, 2205698 (2023)

DFT calculations: Fe vs Fe-O

Fe(100): no unoccupied majority states

majority excitation $\propto U$

(U = eff. **Coulomb** int.)



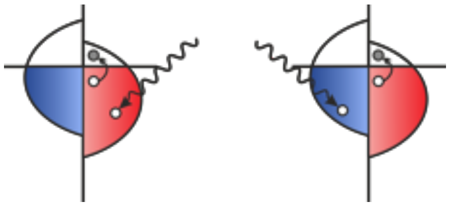
minority excitation $\propto U - J$

(J = eff. **exchange** int.)

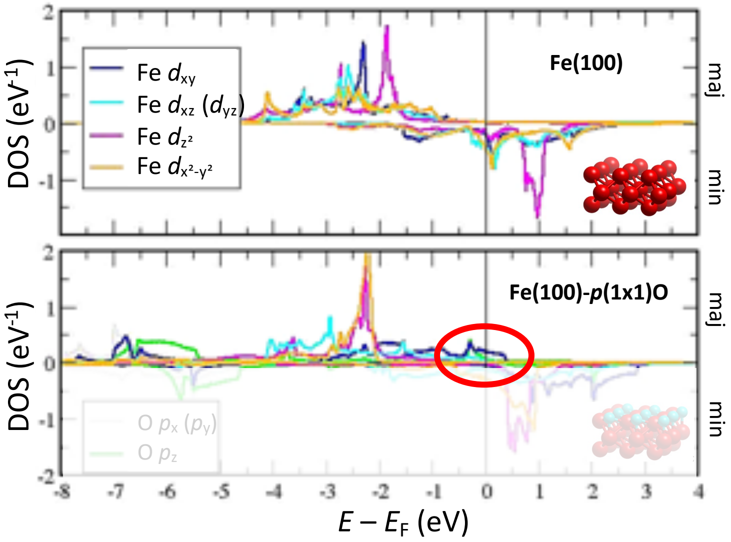
$U > U - J$ ➔ Majority spins stronger correlated[10]

[10] F. Manghi et al., *Phys. Rev. B* **59**, R10409 (1999)

Fe(100)-*p*(1x1)O: band filling reduced



DFT: Orbital-resolved surface DOS



New scattering channels ➔ **Correlation increases,** and **correlation enabled for minority electrons**!

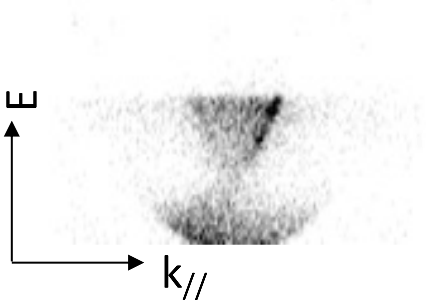
What can we do with it?

(some of the possible) Applications:

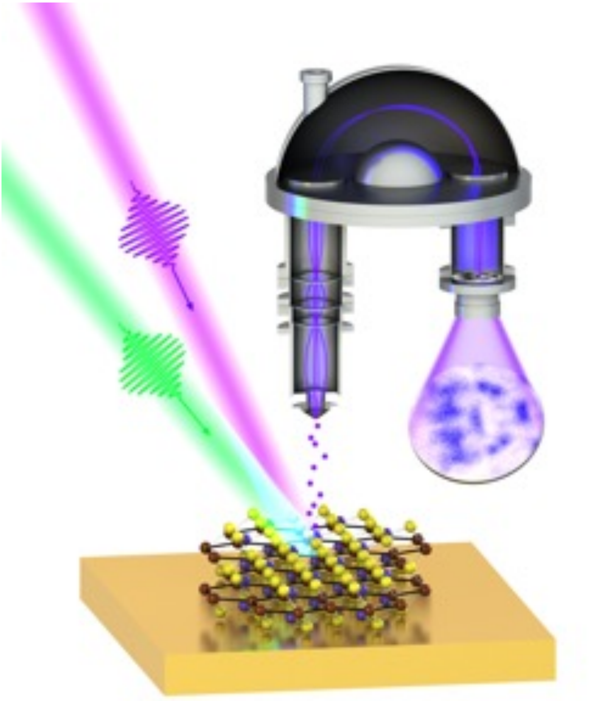
μ-probing

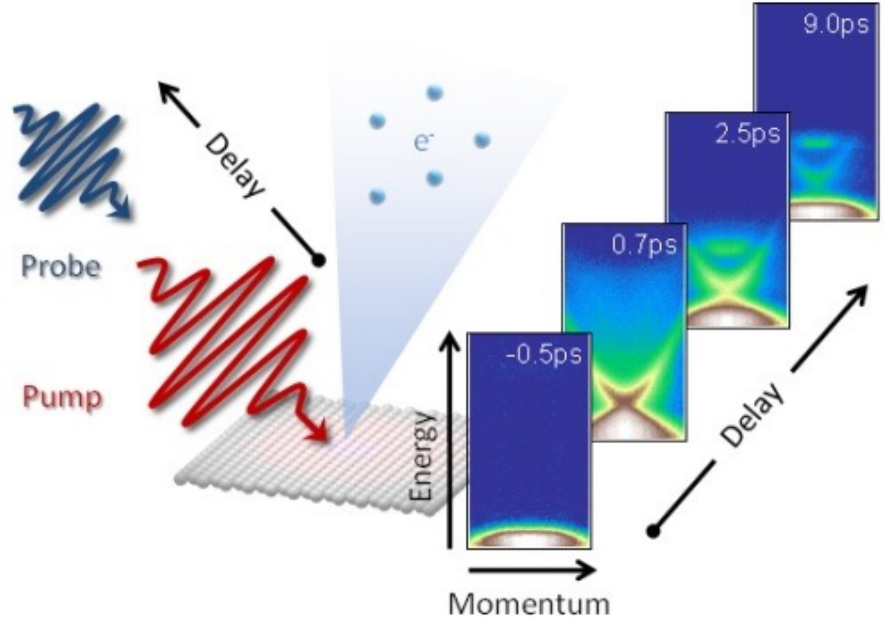
Spin-resolved band structure mapping

Time-resolved Photoemission

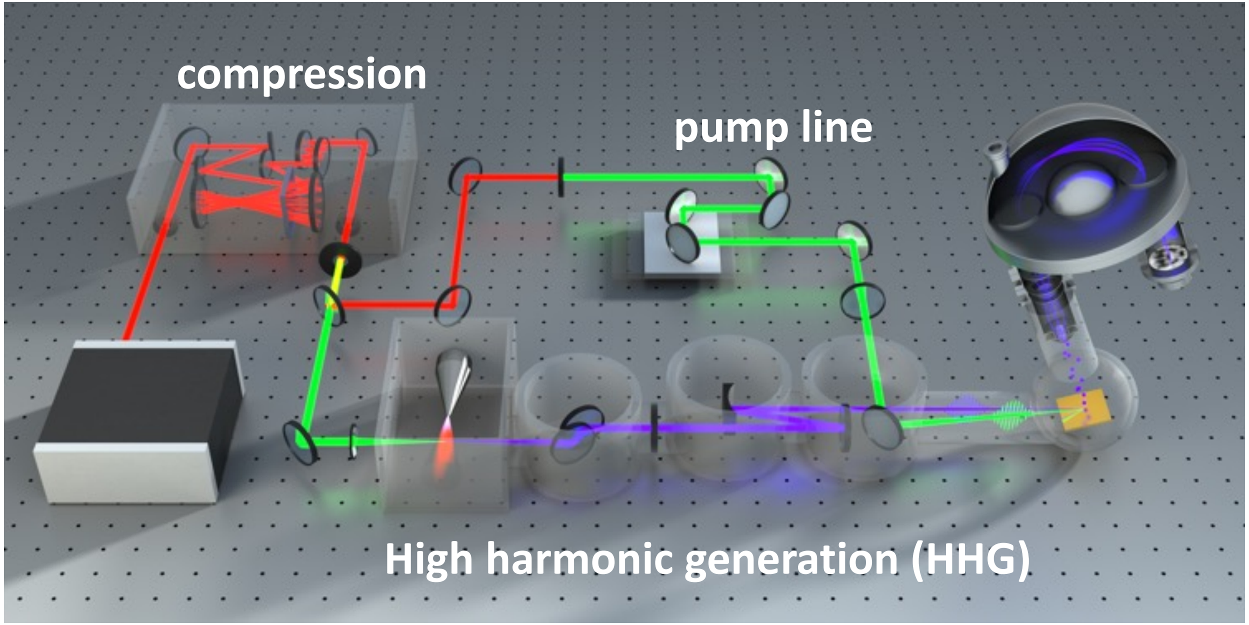


Time-resolved photoemission

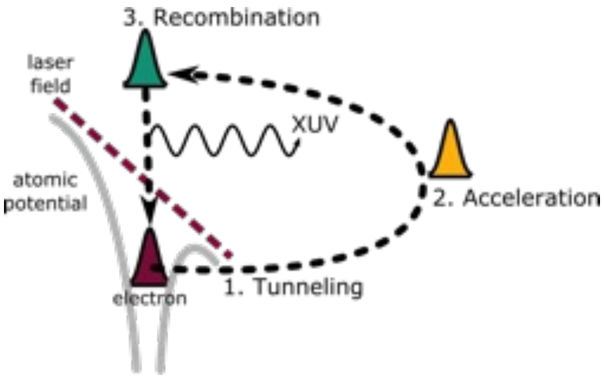




Pump-probe set-up



K. Schiller, <u>GZ</u>, … M. Cinchetti *Sci. Rep.* **15**, 3611 (2025)

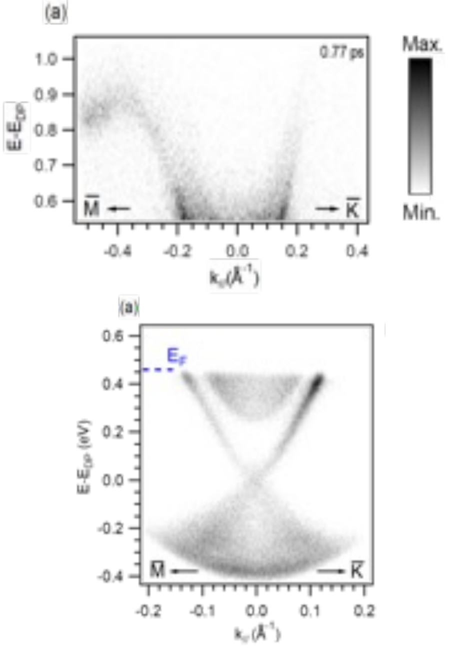


HHG line:

- hν: [20-40 eV]
- Monochromatization: Multilayer mirrors
- Temporal res. 40 fs
- Spot size 20x80 μm^2
- Low energy res. 200 mev <

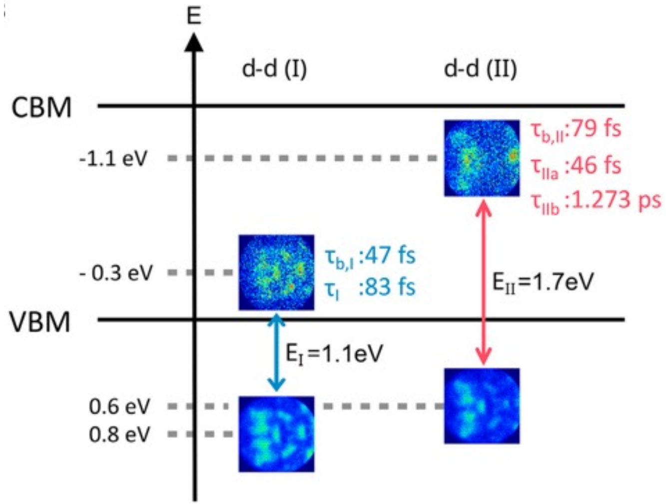
Looking at different systems

Bi_2Se_3



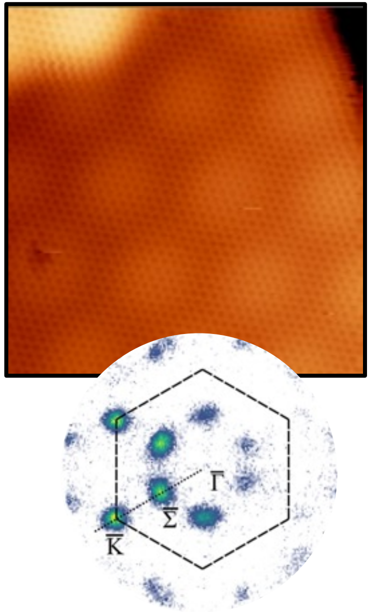
S. Ponzoni, .. <u>GZ</u> and M. Cinchetti
Adv. Phys. Res. **2**, 2200016 (2023).

$FePS_3$



J. E. Nitschke, .. <u>GZ</u>, … M. Cinchetti *Newton* **1**, 100019 (2025).

WS_2

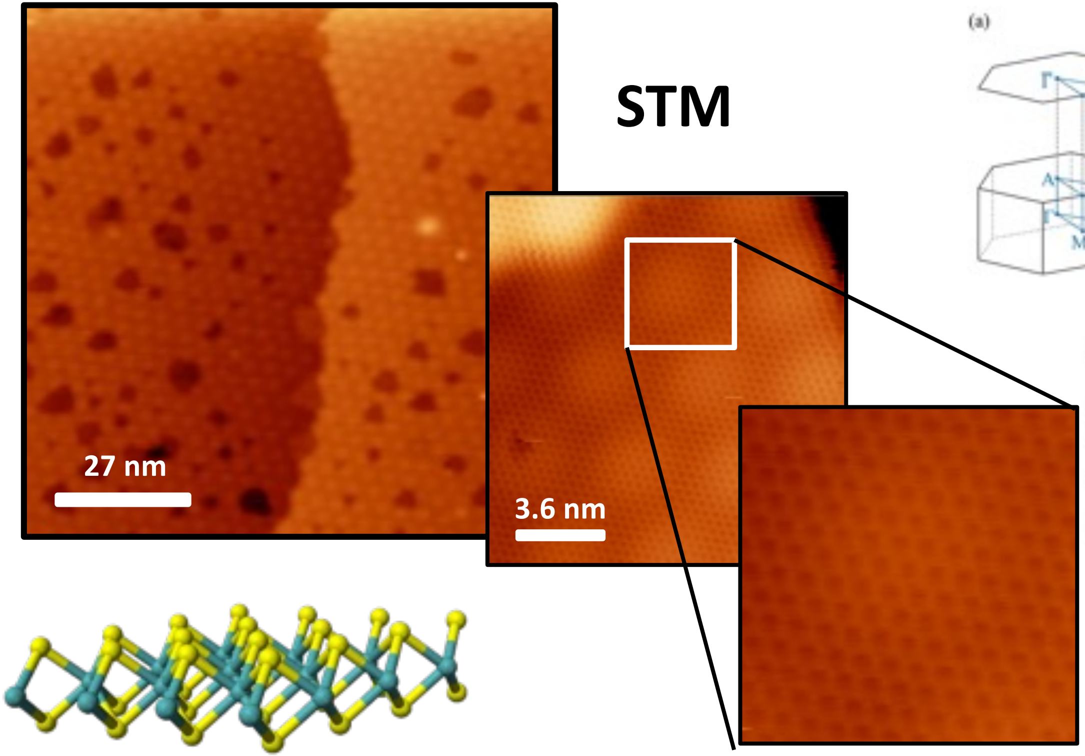


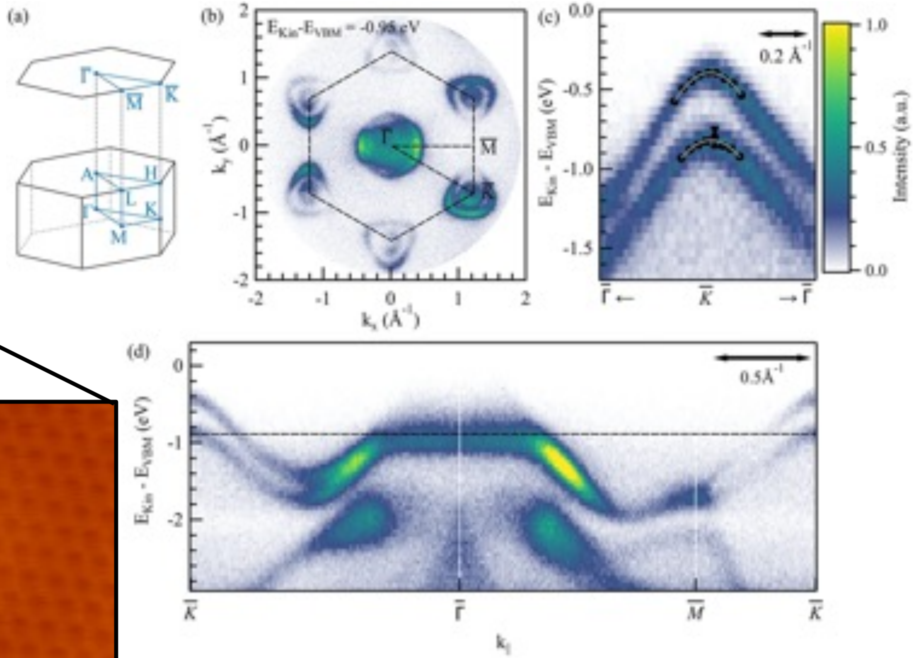
K. Schiller, <u>GZ</u>, … M. Cinchetti *Sci. Rep.* **15**, 3611 (2025)
M. Capra,… <u>GZ</u> in preparation (2025)

WS_2 – synthesis/characterization

Single layer WS_2/Au(111) (in-house grown)

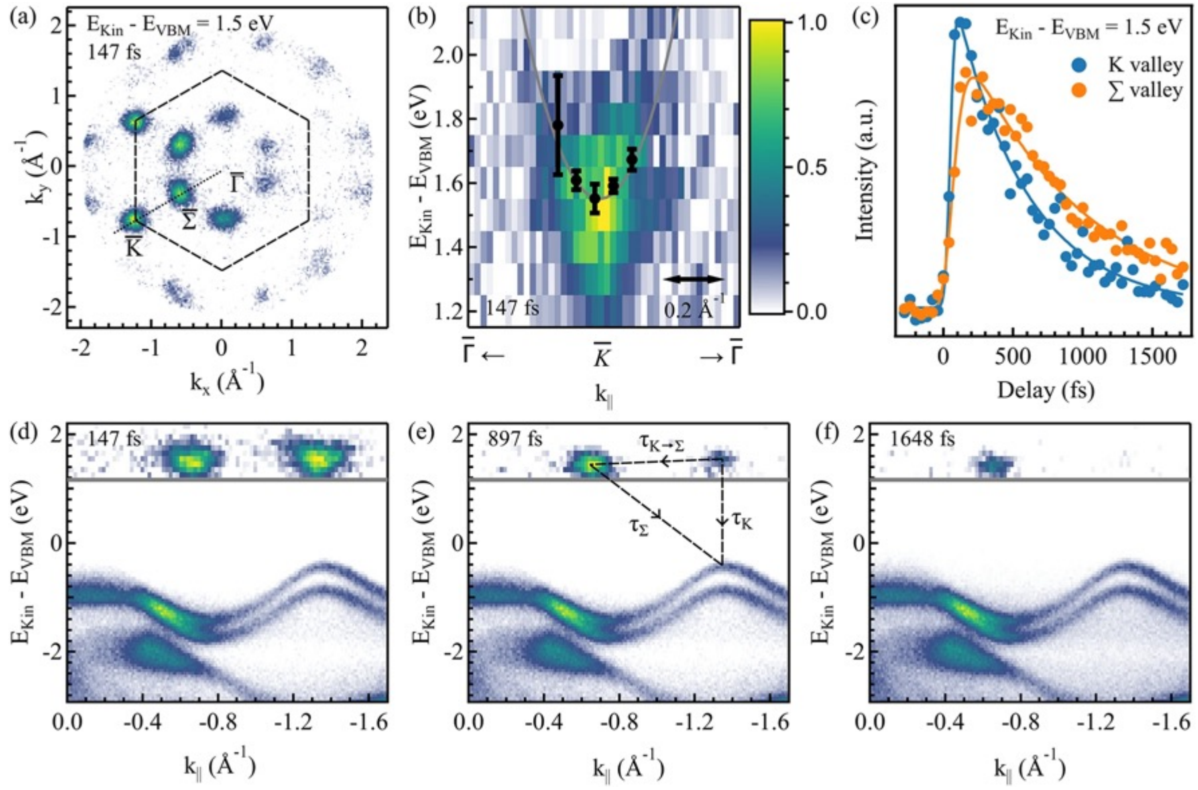
Bulk WS_2



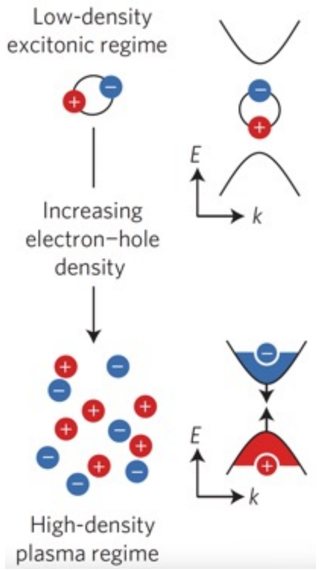


K. Schiller et al. *Scientific Reports* **15**, 3611 (2025)
M. Capra et al. in preparation (2025)

WS_2 – Dynamics



K. Schiller et al. *Scientific Reports* **15**, 3611 (2025)



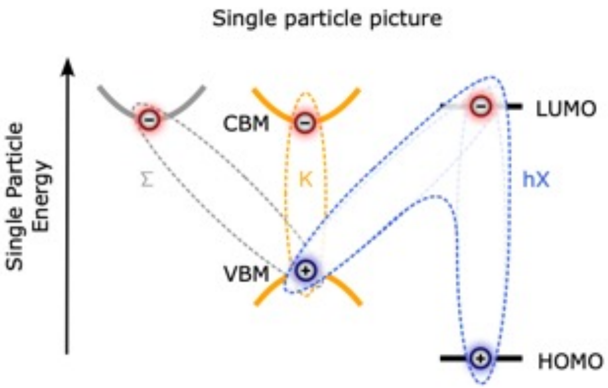
Mott Transition

Adapted from: A. Chernikov et al. *Nat. Photonics* **9**, 466 (2015)

What’s next?

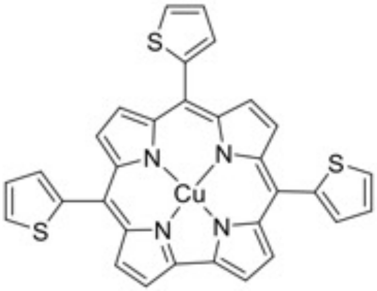
Engineering Organic/TMDs heterostructures

New excitonic channels:
Interlayer excitons



Adapted from: W. Bennecke et al. arXiv:2411.14993

On-surface tuning of TMD’s electronic properties *via* molecular doping

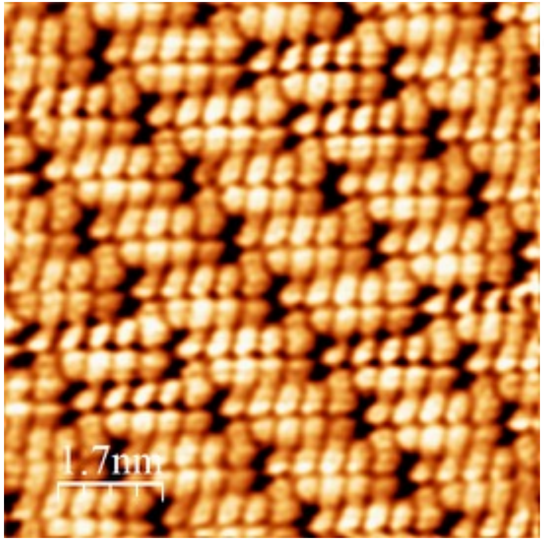


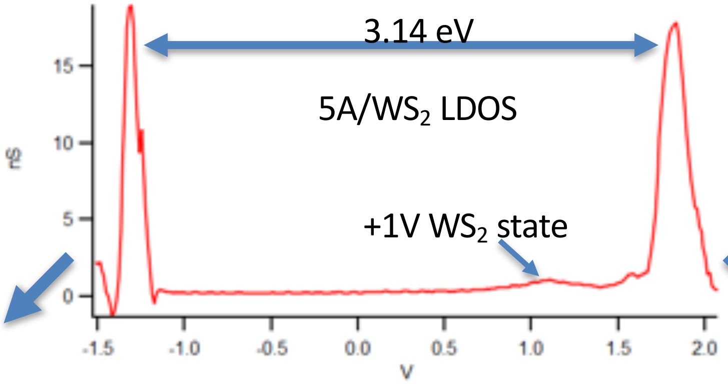
In collaboration with
Abhik. Ghosh, Uni Tromsø.

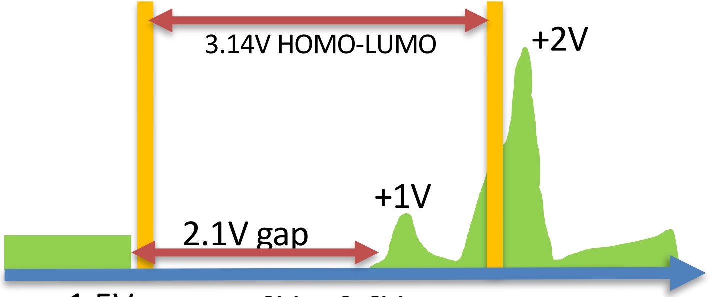
WS_2 + Pentacene (5A)

STM + STS

-1.4V, 5A HOMO

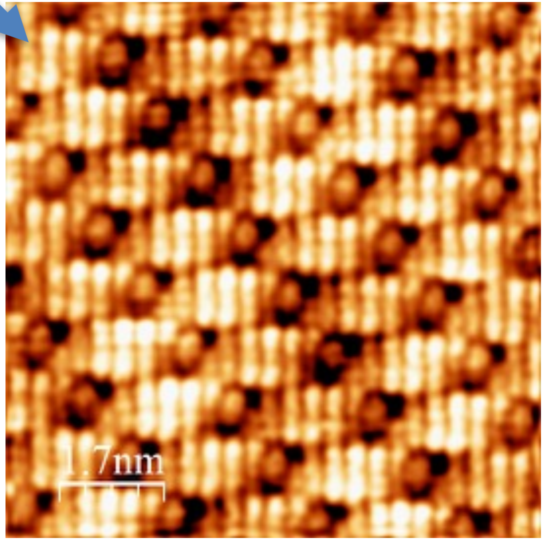






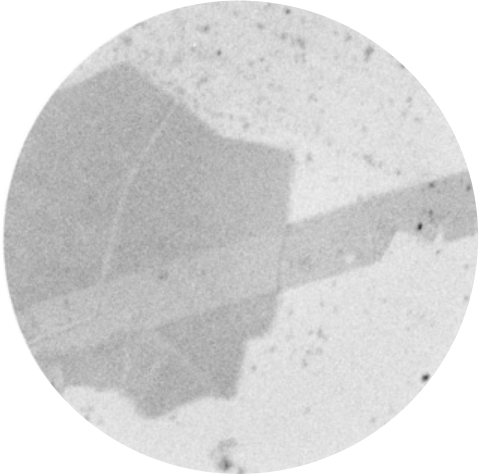
Type II alignment

+1.8V, 5A LUMO

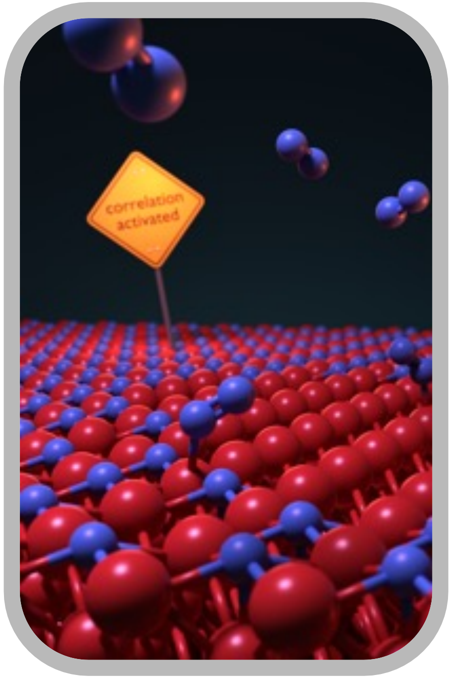


With PEEM we can access…

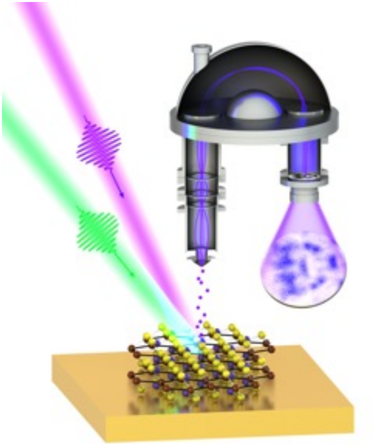
µ-probing of the local electronic structure



Electronic and spin-properties of surface



Excited states (out-of-equilibrium dynamics)



Acknowledgements

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Thank you for your attention!!