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Dolní Břežany, Czech Republic

Probing Excitons in Time, Energy, Momentum, and Space by Photoelectron Momentum Microscopy

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University of Göttingen

1 September 2023

Dolní Břežany, Czech Republic

eli

IMPULSE

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SFB 1073

CRC 1456
MATHEMATICS OF EXPERIMENT

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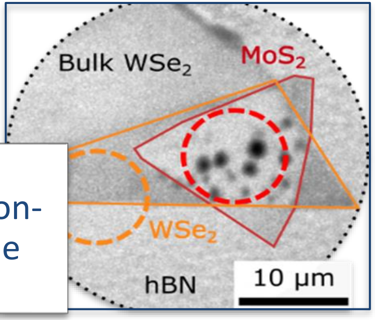
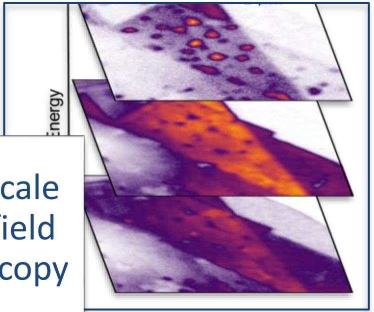
DFG Deutsche Forschungsgemeinschaft
German Research Foundation

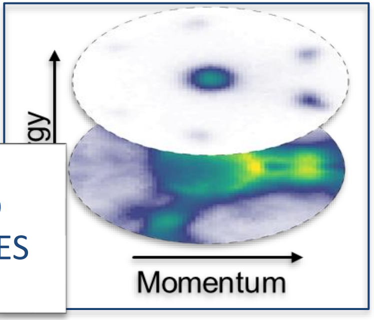
Photoelectron Momentum Microscopy



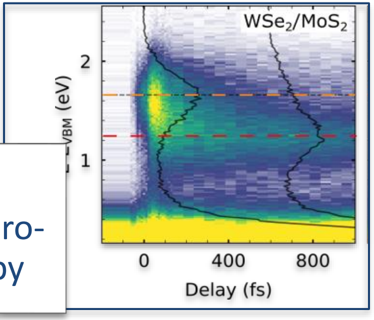
Micron-scale



Nanoscale Dark-field Microscopy



3D ARPES

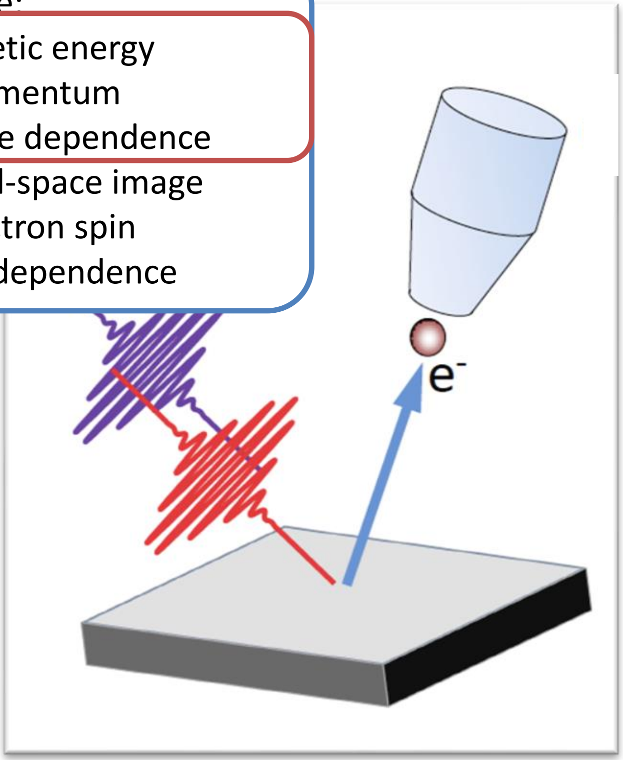


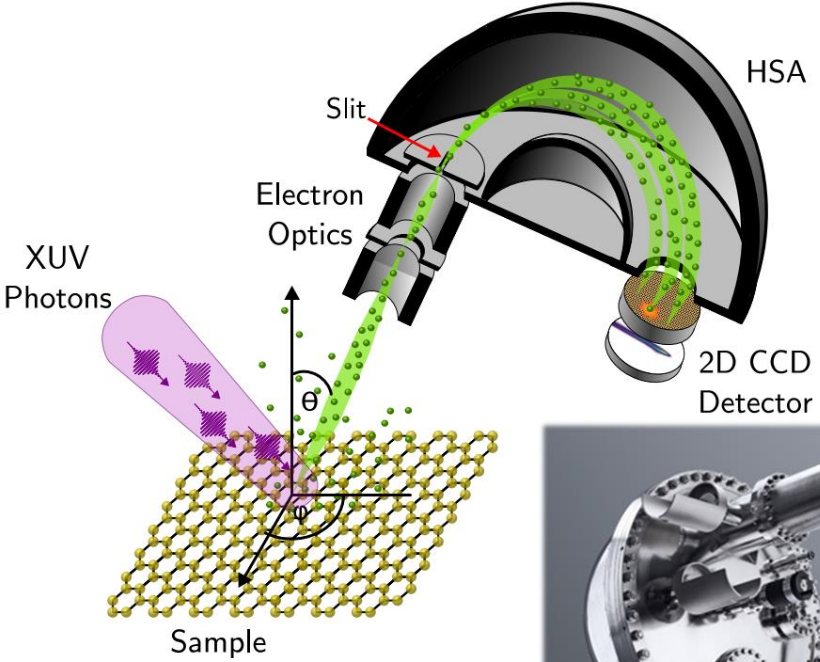
fs-Spectro-scopy

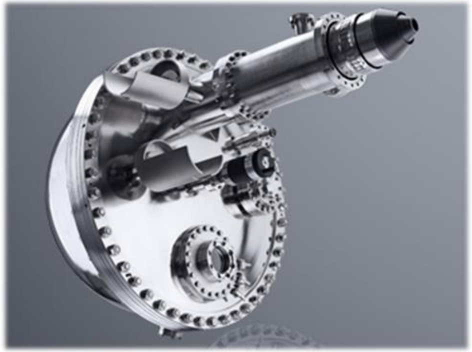
Photoelectron Spectroscopy

Measure:

- Kinetic energy
- Momentum
- Time dependence
- Real-space image
- Electron spin
- $h\nu$-dependence







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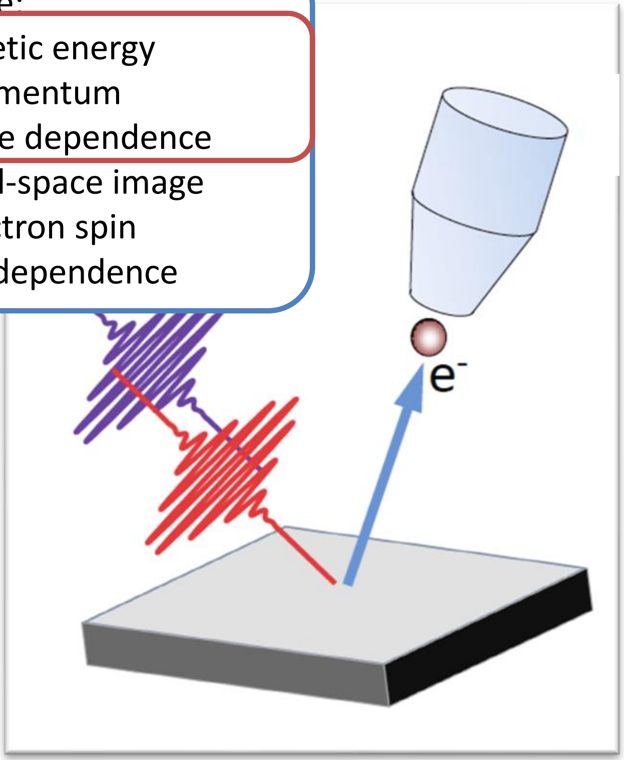
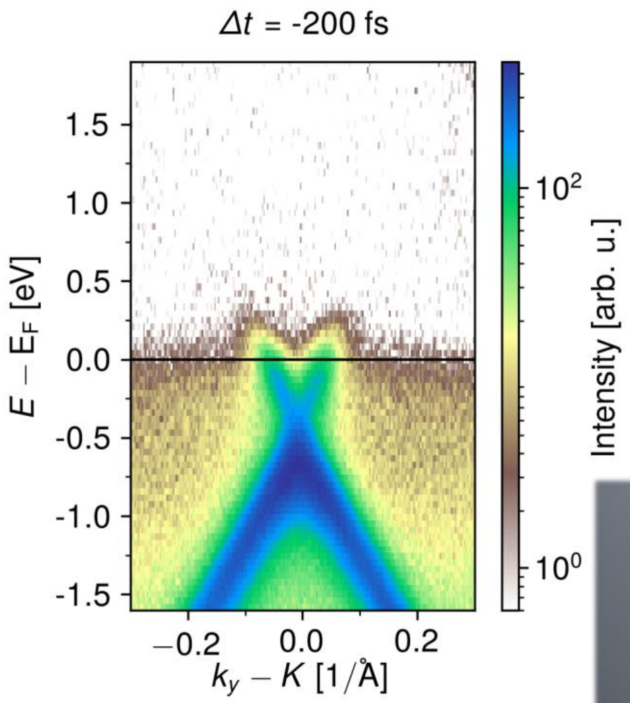
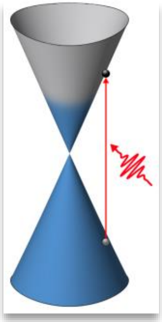
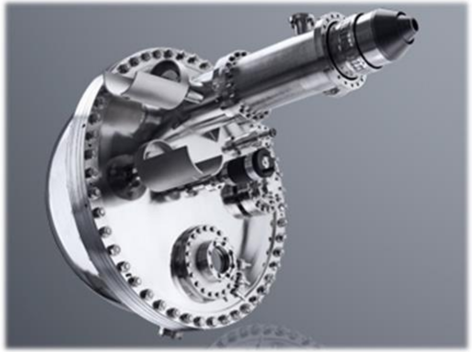


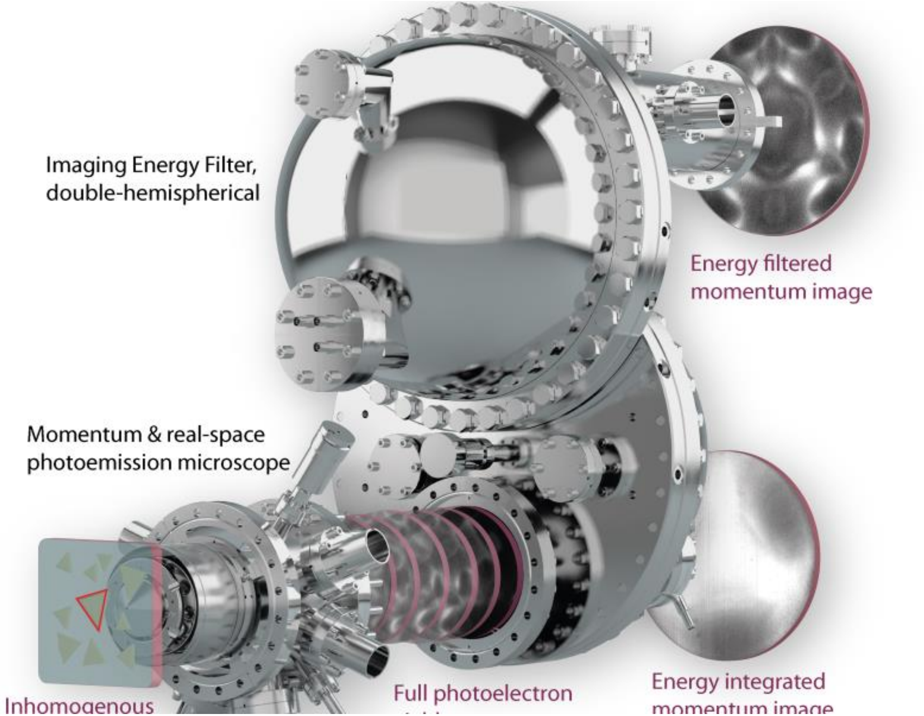






The Future

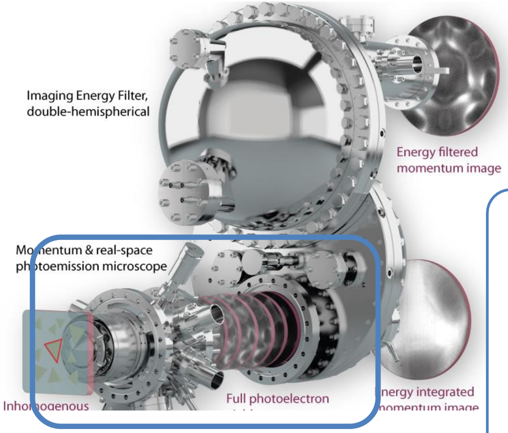
- Momentum Microscopy



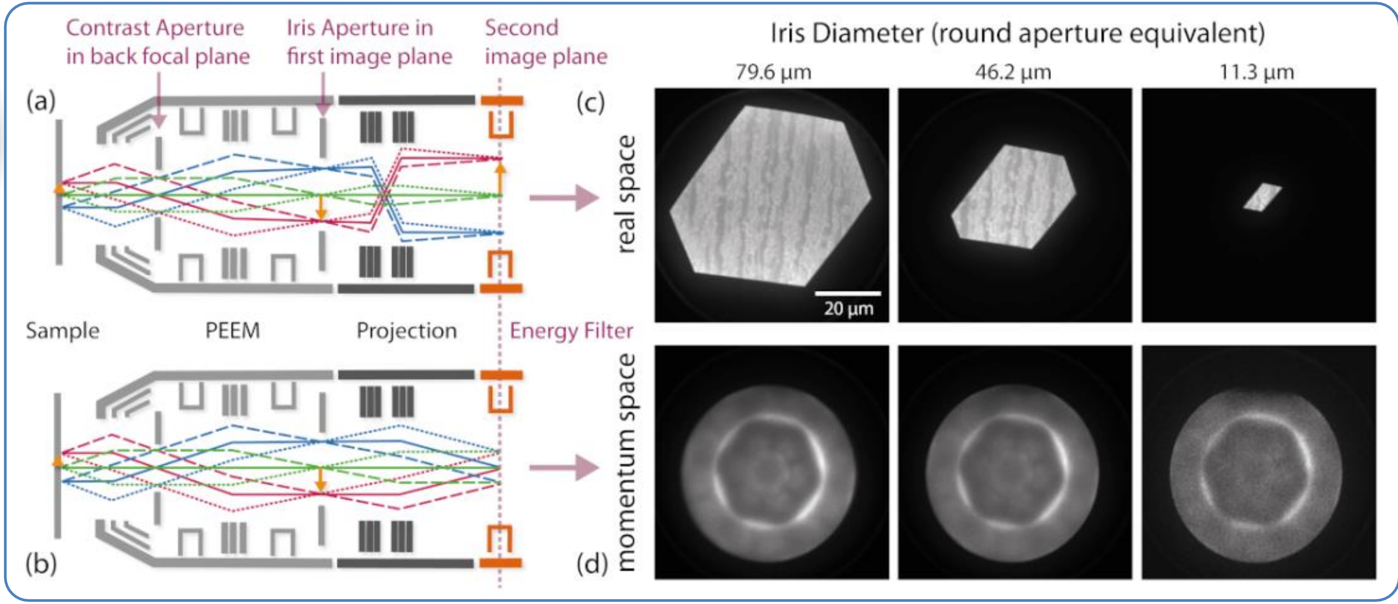
https://scientaomicron.com

https://www.specs-group.com/

Momentum Microscope



- Electron microscope: electrostatic lenses image the electron beam



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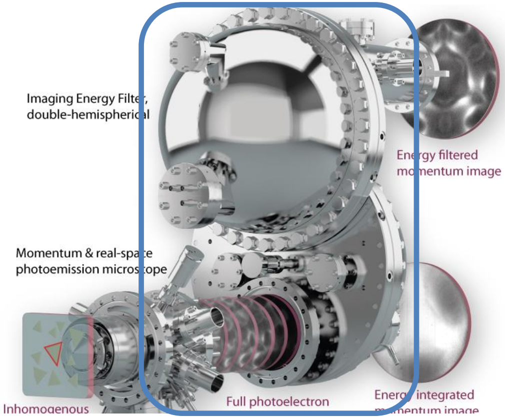
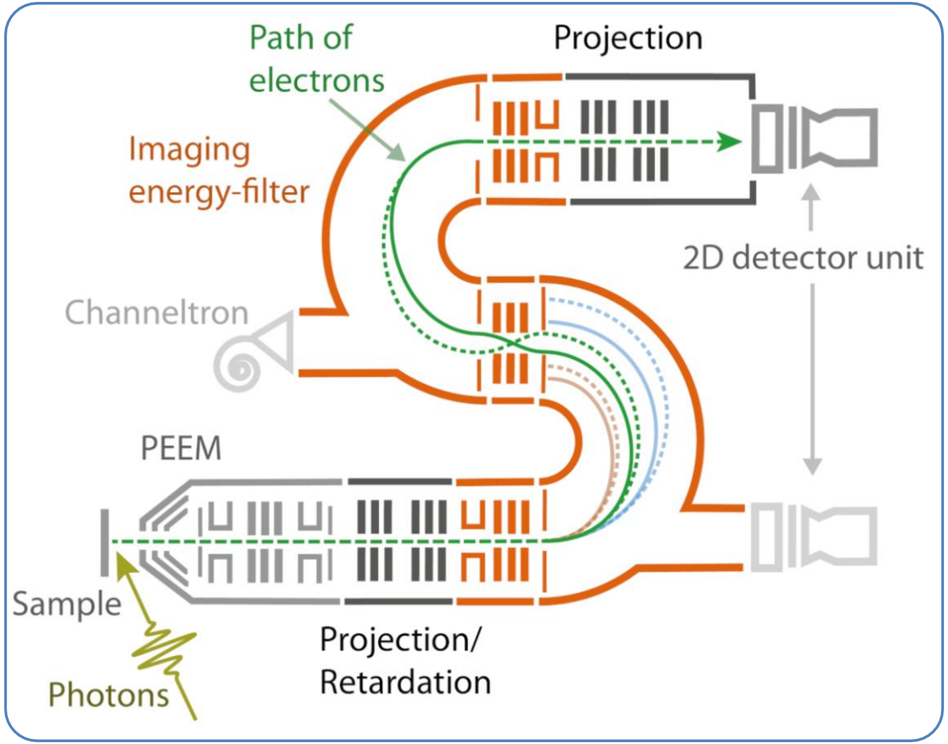
Momentum Microscope: Detection



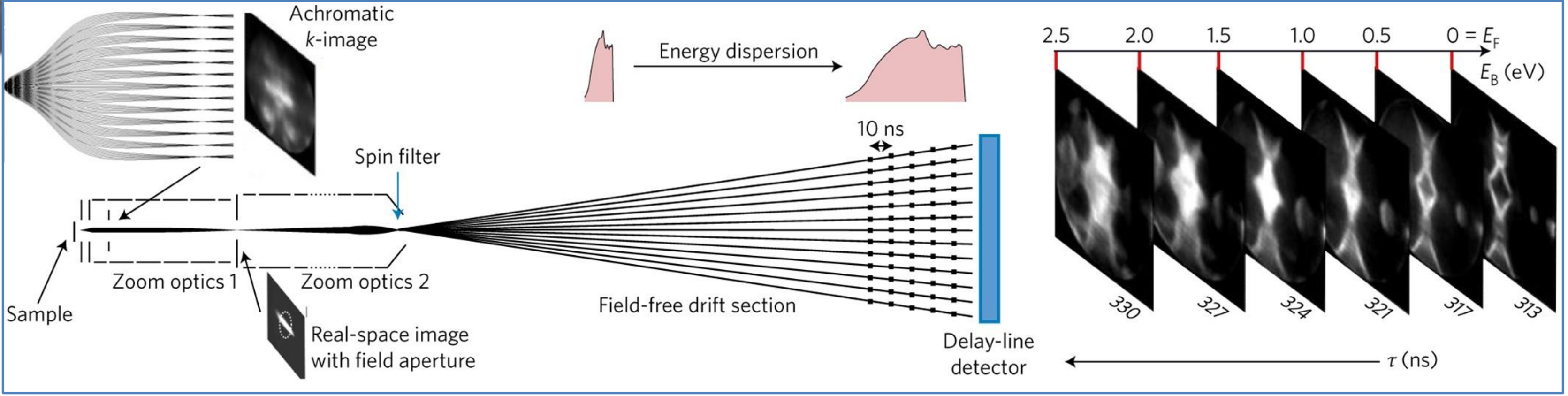
- Energy dispersion:
  select photon energy range



https://scientaomicron.com

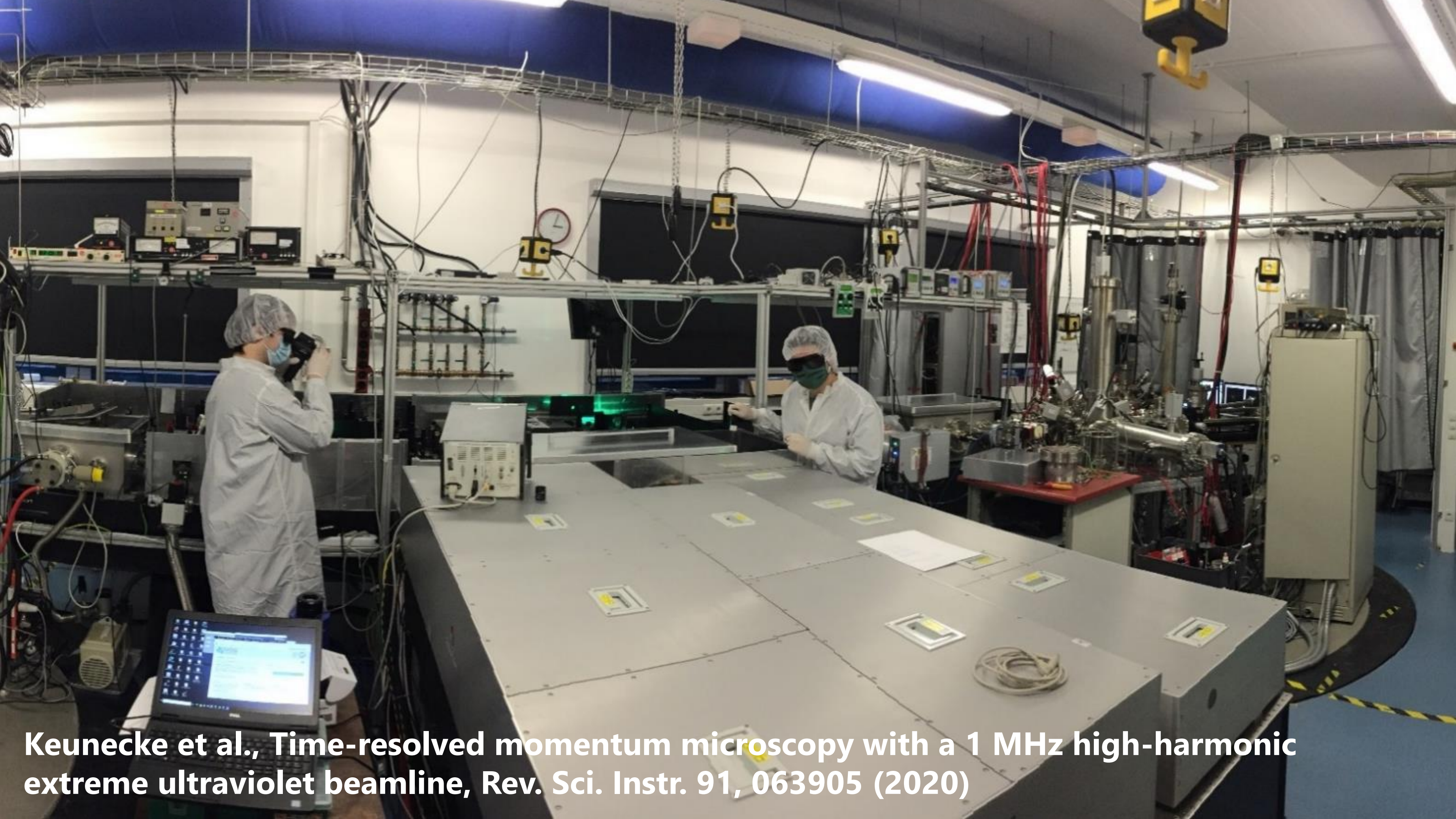
Momentum Microscope: Detection

- Time-of-flight detection: energy and 2D momentum for each electron



- Event-based detection: one electron per pulse

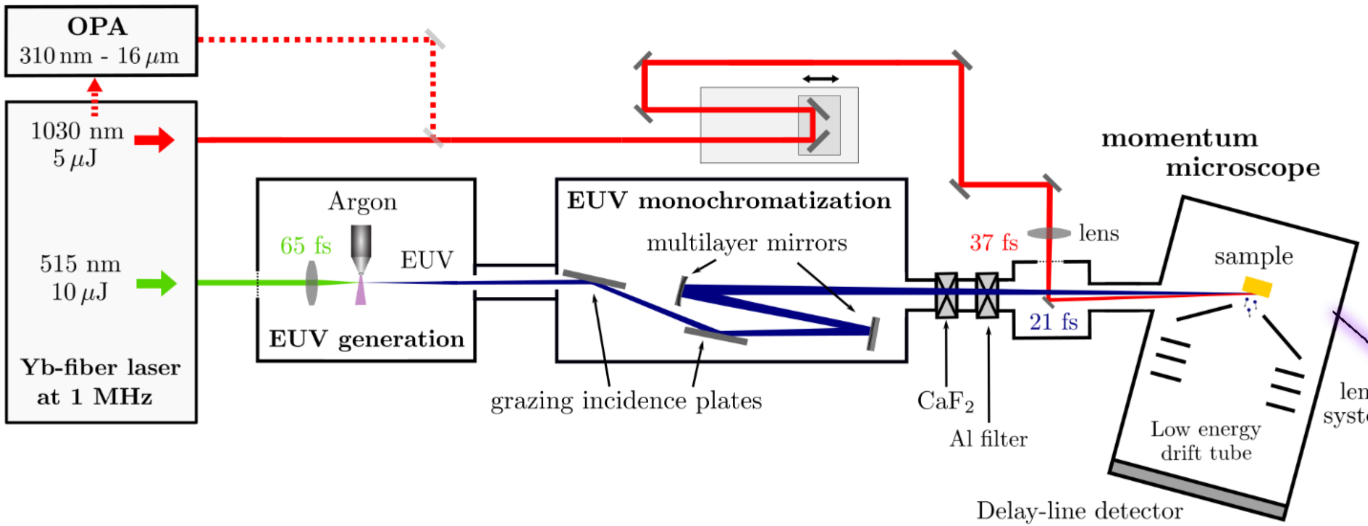
K. Medjanik *et al.*, Nat. Materials **16**, 615–621 (2017)

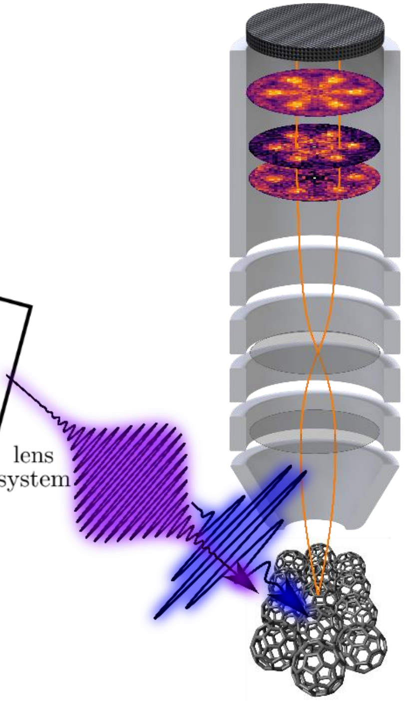


Keunecke et al., Time-resolved momentum microscopy with a 1 MHz high-harmonic extreme ultraviolet beamline, Rev. Sci. Instr. 91, 063905 (2020)

The photoelectron momentum microscope

- Light source: **≥500 kHz** HHG, 26 eV (20 – 70 eV)
- Pump pulses: OPA, 300 nm – 16 µm, 30 – 50 fs





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Time-resolved momentum microscopy with a 1 MHz high-harmonic extreme ultraviolet beamline,
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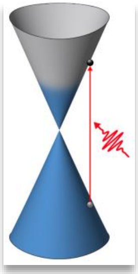
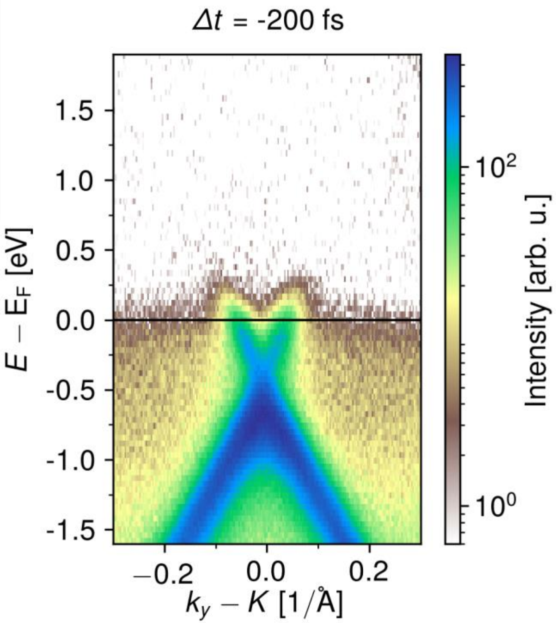
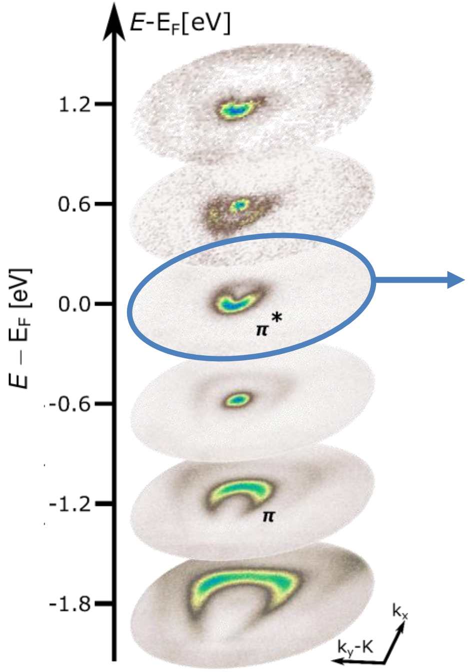
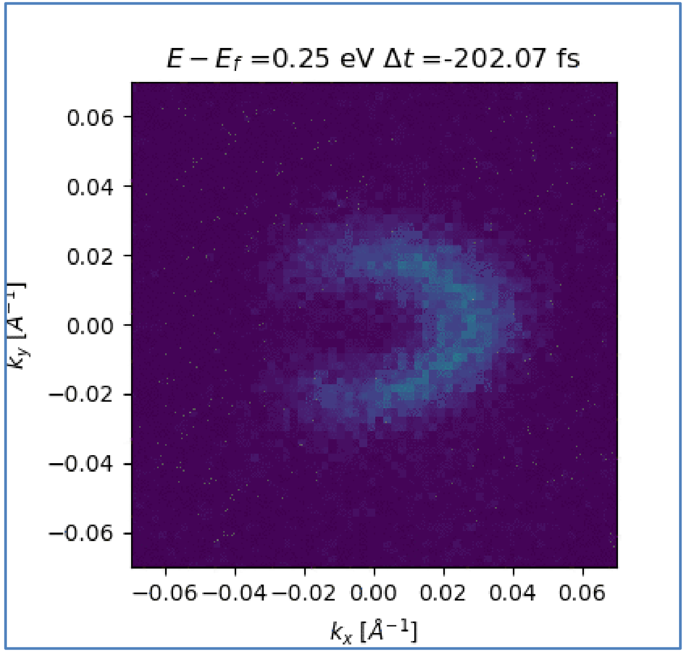
Into the third dimension

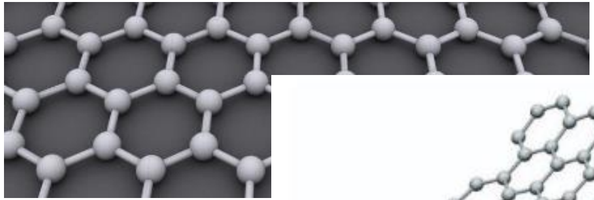


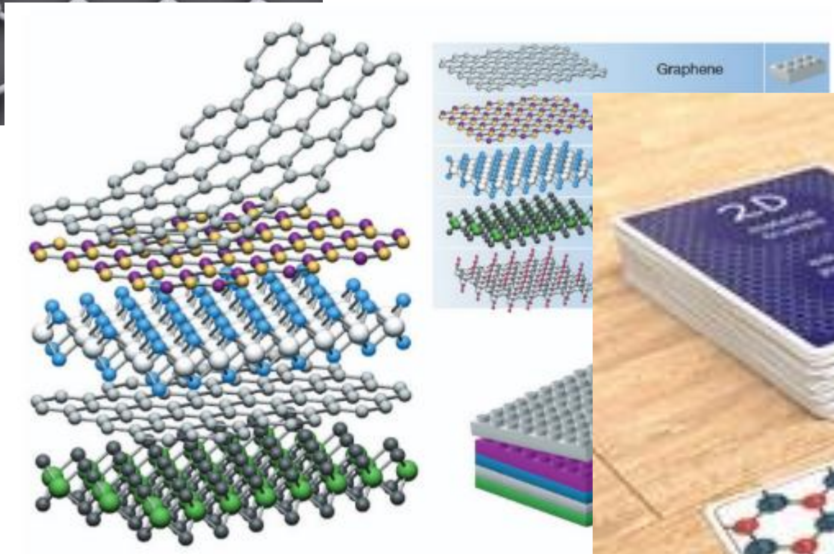




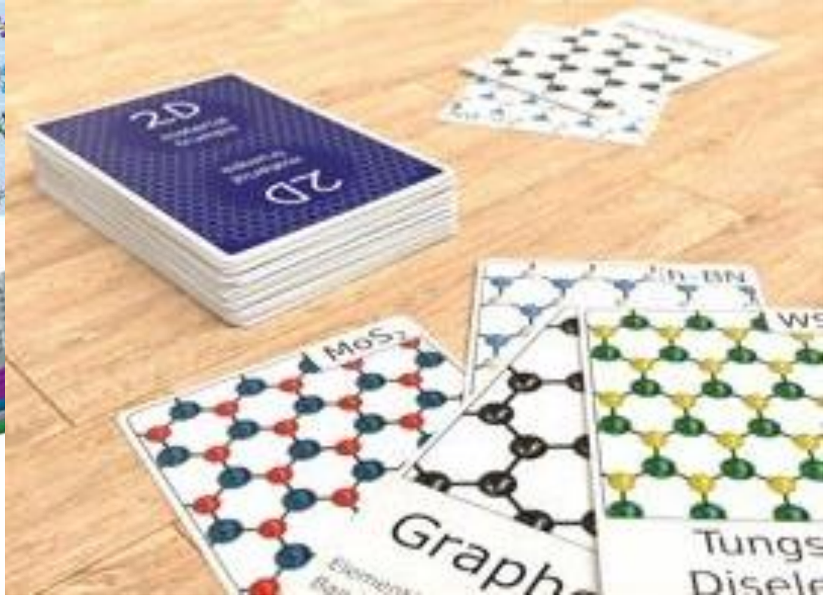


TMD Nanostructures





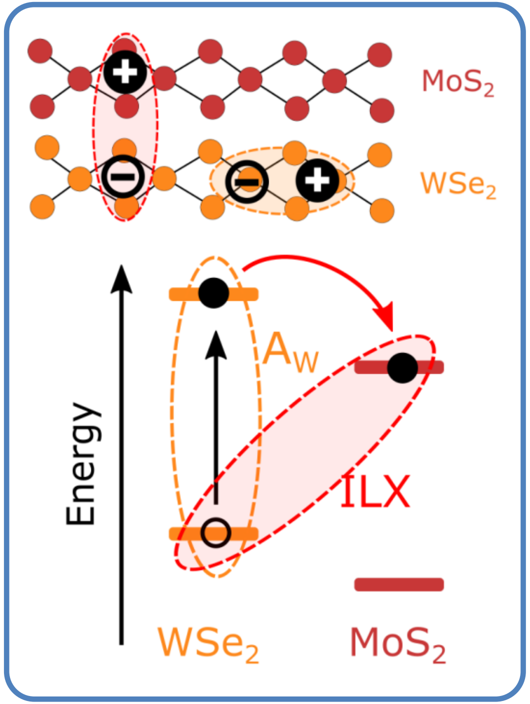
Nature **499**, 419 (2013)



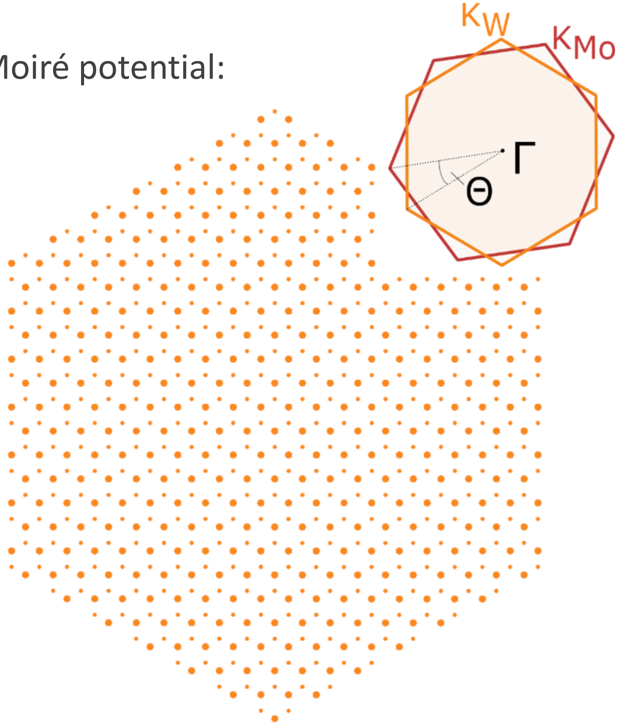
Physik Journal **18**, 29 (2019).

Engineering light-matter interaction

- Interlayer excitons: electron-hole separation at atomic scales



Moiré potential:



Control of opto-electronic properties through:

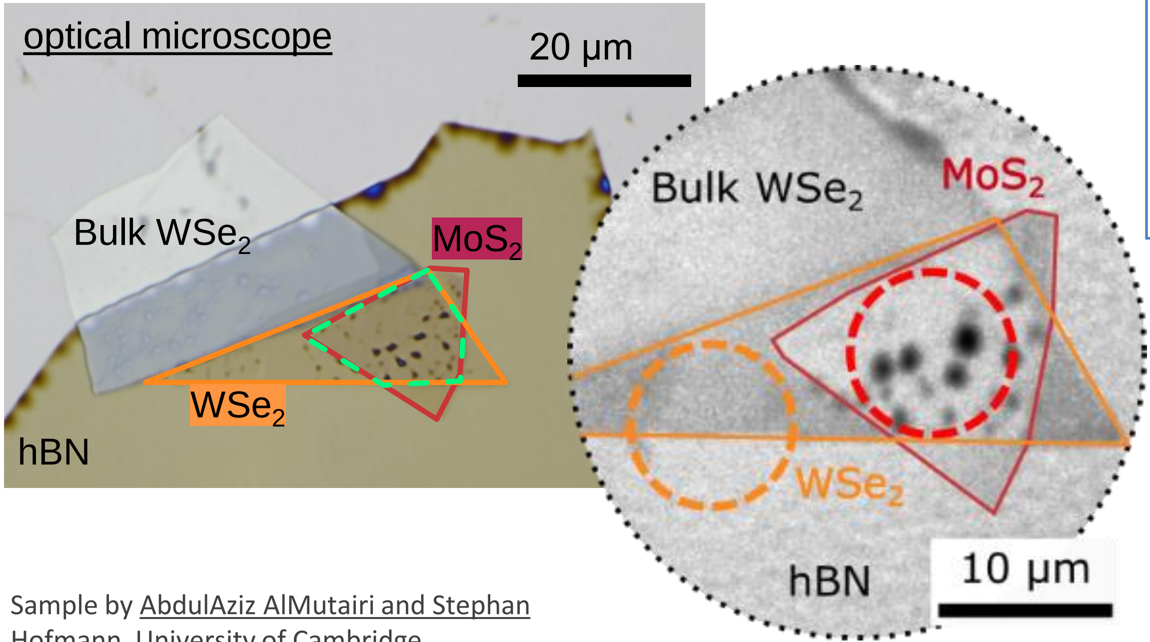
- Band alignment (W/M, S/Se)
- Interlayer twist
  - Moiré potential

Interlayer exciton promises:

- Enhanced lifetime
- Confined states
- Strongly-correlated phases

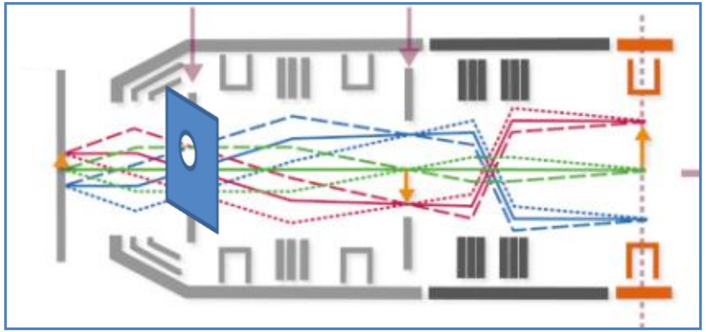
ARPES at the microscale?

- 2D materials are often exfoliated, and have <100 µm size
- Layered structures are smaller, typically 10 µm



Sample by AbdulAziz AlMutairi and Stephan Hofmann, University of Cambridge

Real-space mode (PEEM)



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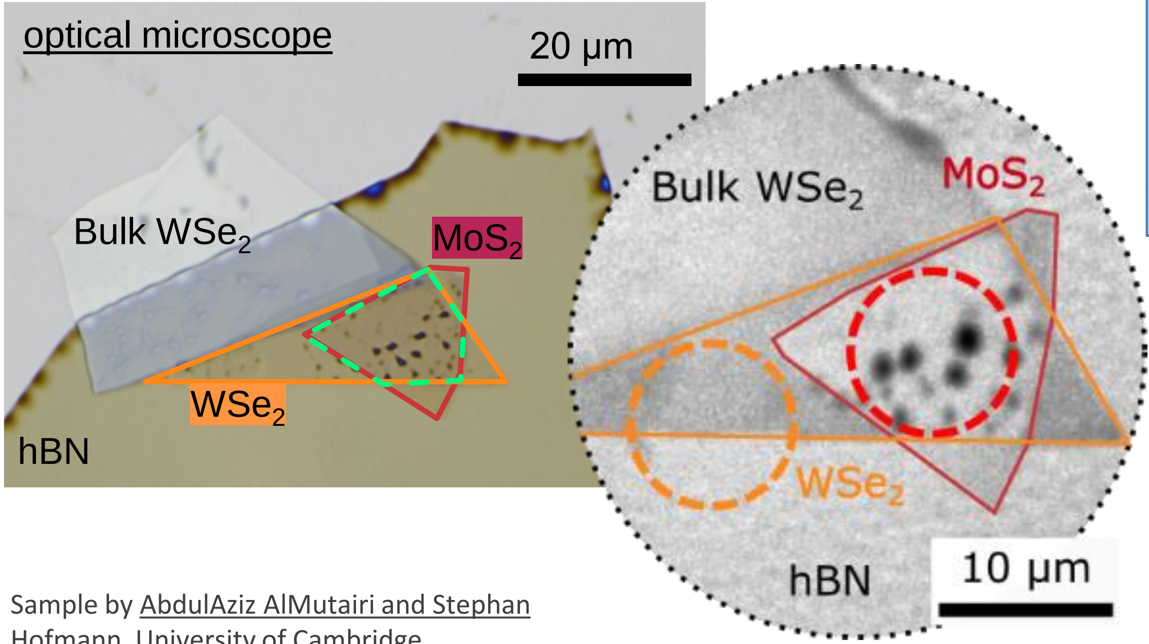
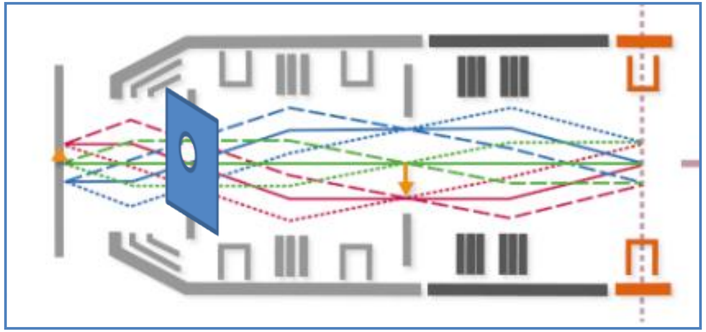
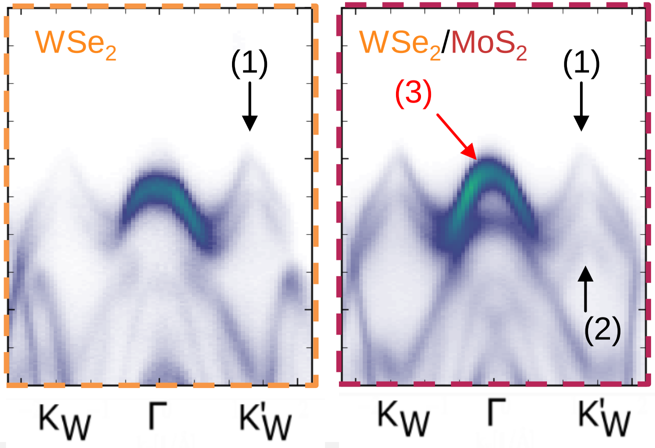


Sample by AbdulAziz AlMutairi and Stephan Hofmann, University of Cambridge

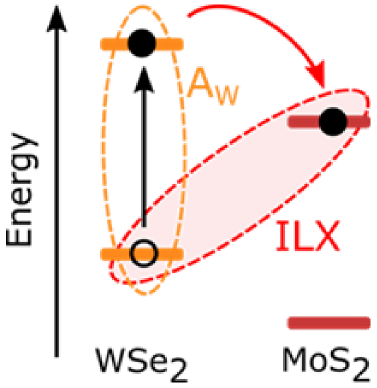
k-space mode (ARPES) – with aperture

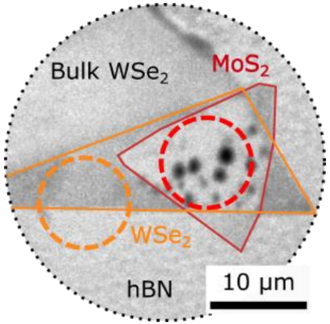


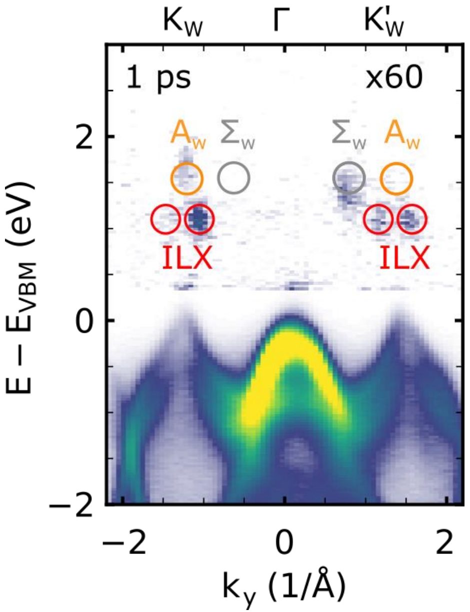


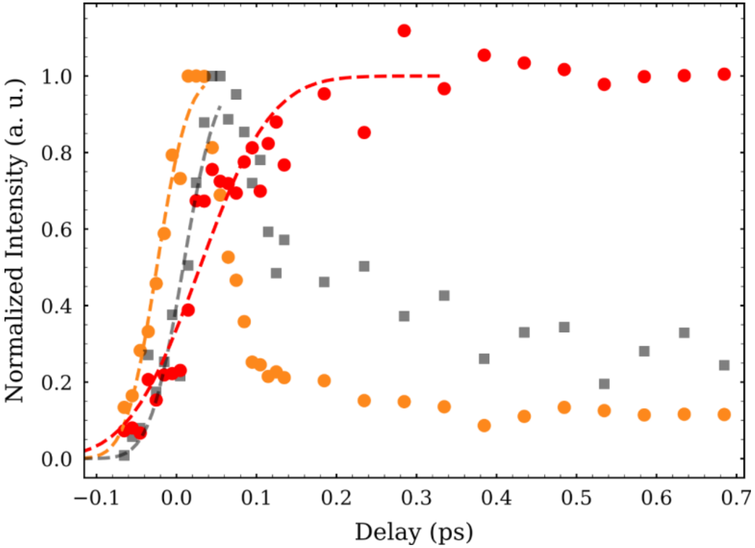
Time-resolved μ-ARPES

- ARPES separates different excitons by energy & momentum
- Direct access to dynamics



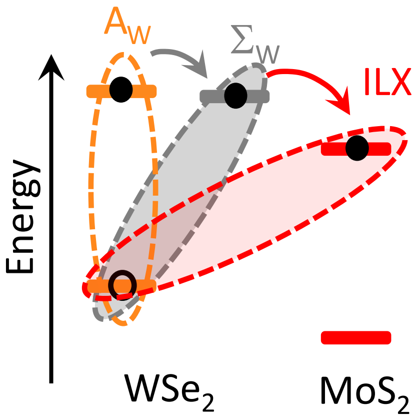


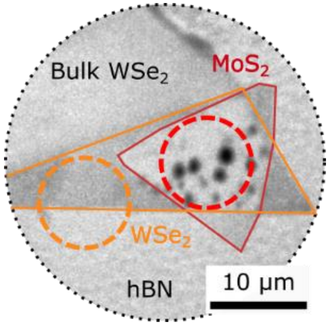


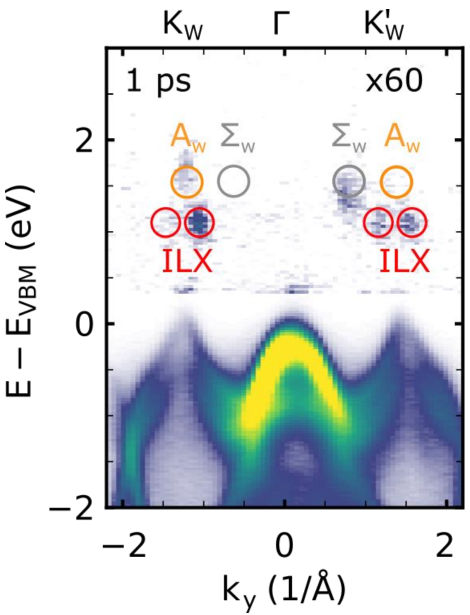


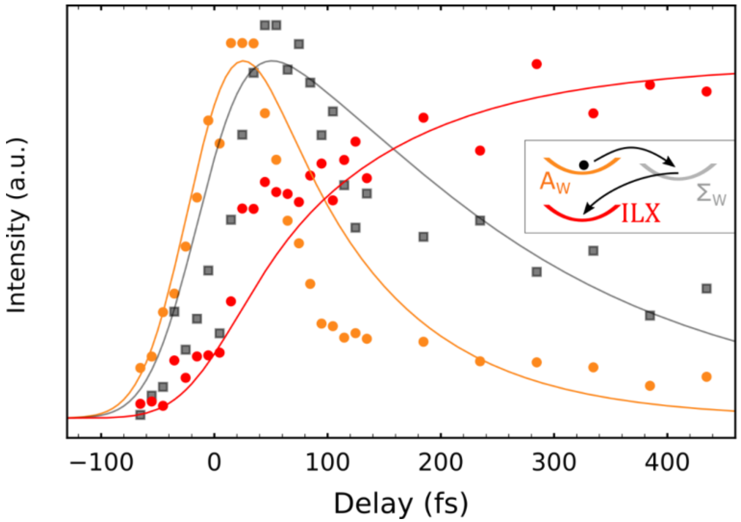
Time-resolved μ-ARPES

- ARPES separates different excitons by energy & momentum
- Direct access to dynamics
- And excellent comparison with theory



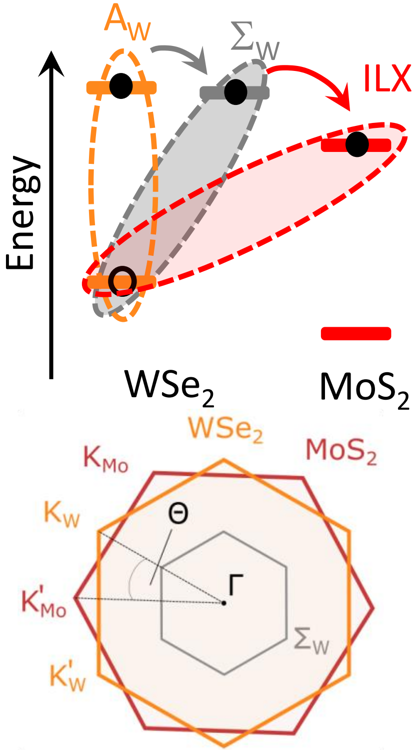


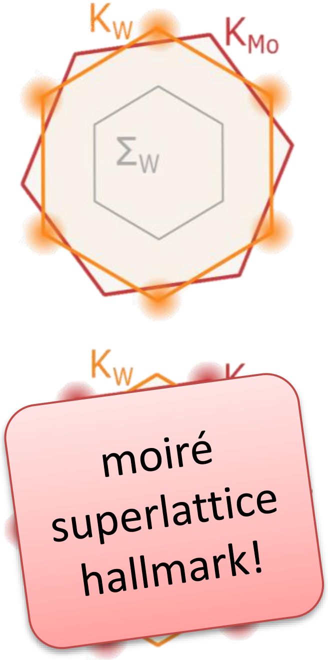


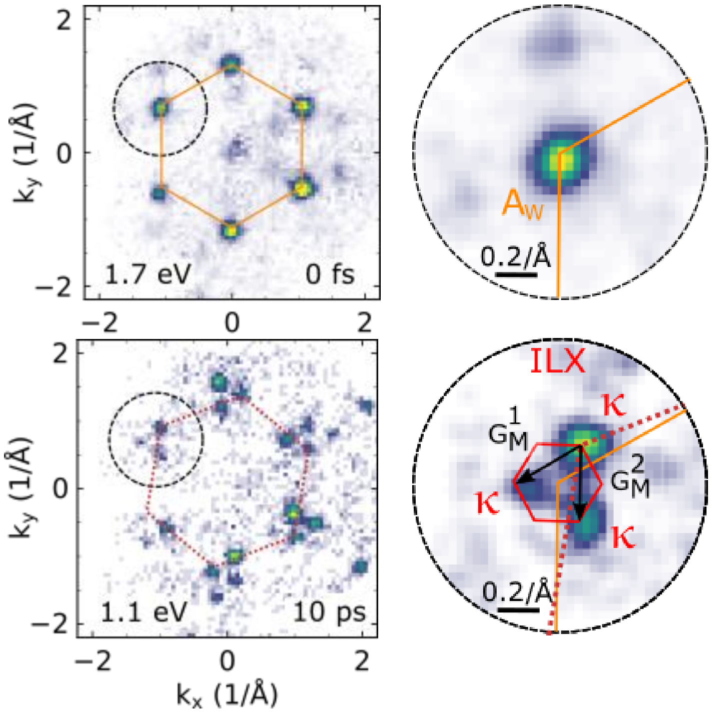


microscopic modelling by Giuseppe Meneghini, Samuel Brem, and Ermin Malic, University of Marburg

A unique momentum fingerprint





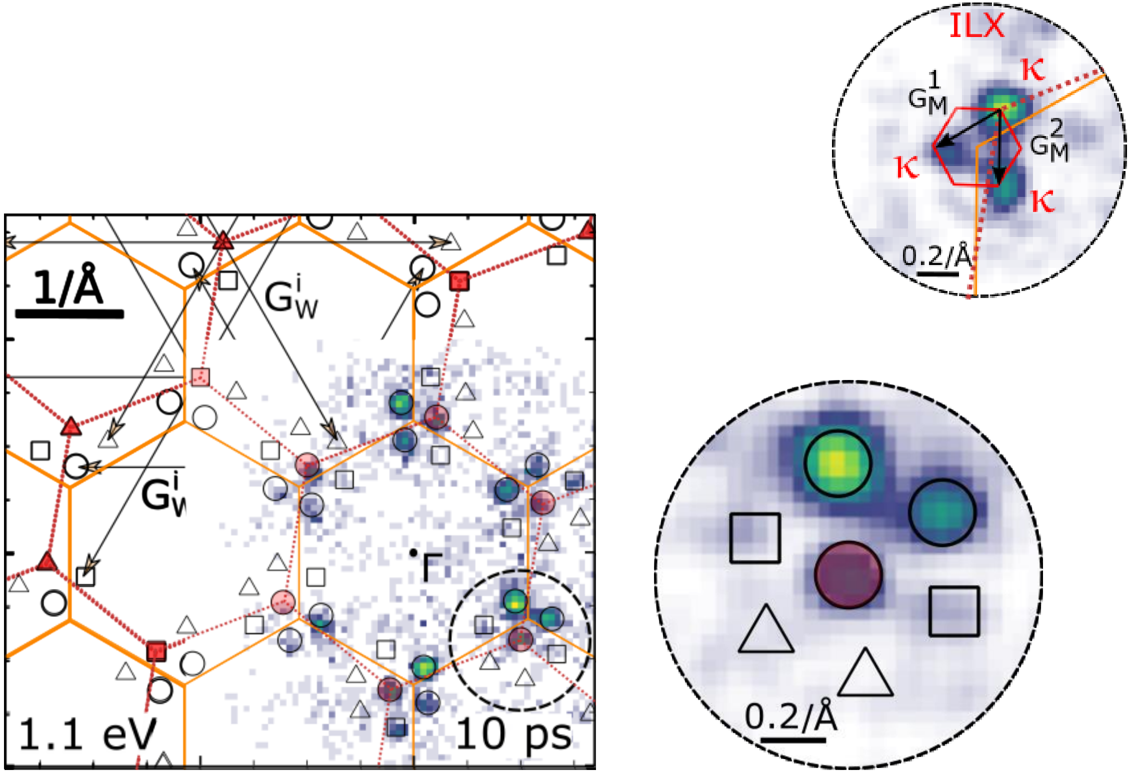


Electron-hole interaction

Threefold momentum signature:

- ✖ Umklapp scattering
  - Only for *interlayer exciton*
- ✖ Exciton confinement?
- ✖ Orbital hybridization?
  - Not expected for 10° twist

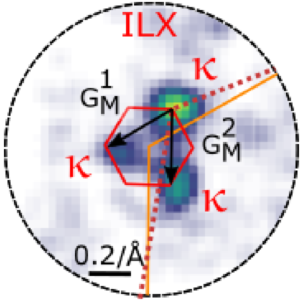
- Generalized interaction between lattices:
  - Coulomb (e-h) interaction

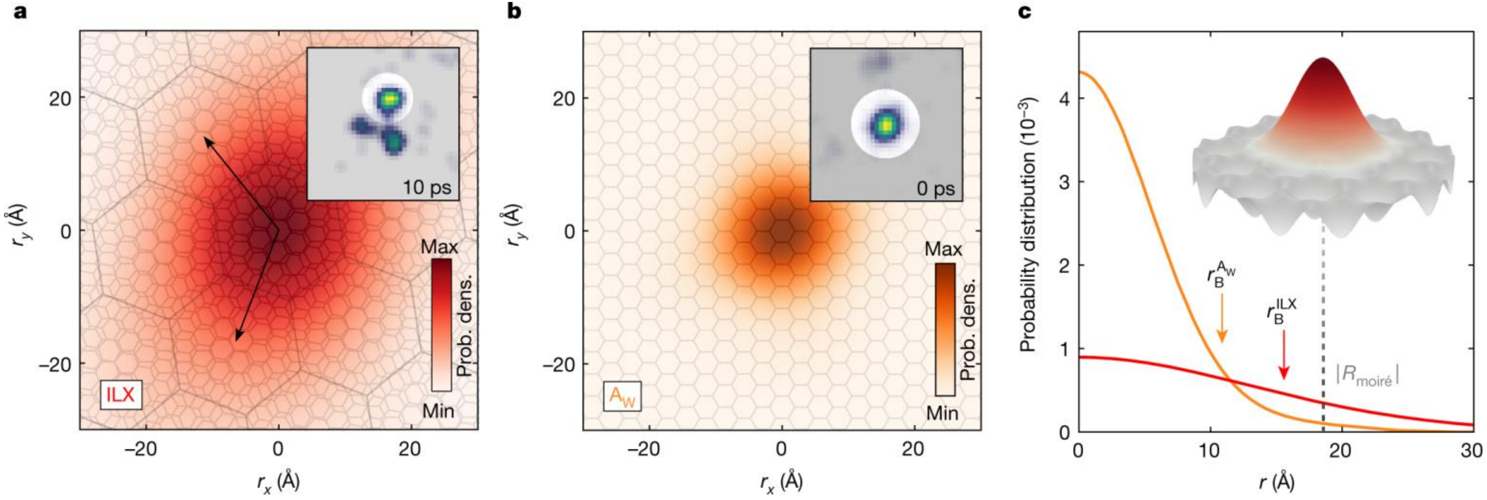


Exciton delocalization

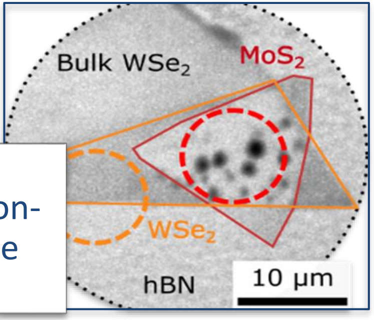
- Plane wave model of photoemission:
  - $I(k_{\|}) \propto \left|\boldsymbol{A} \cdot \boldsymbol{k}_f\right|^2 |\mathcal{F}(\psi)|^2 \times \delta(h\nu - E_B - \Phi - E_{kin})$

- Bohr radii:
- Interlayer exciton:
  - 1.6 ± 0.2 nm
- WSe_2 A-exciton:
  - 1.1 ± 0.1 nm





Photoelectron Momentum Microscopy



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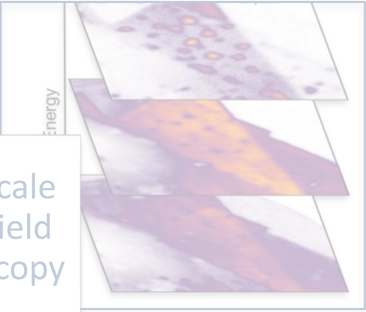
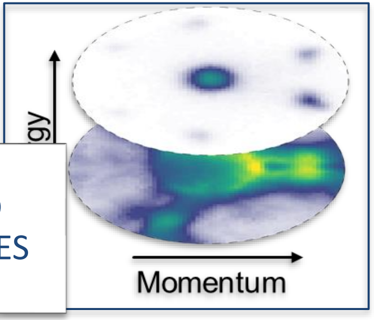
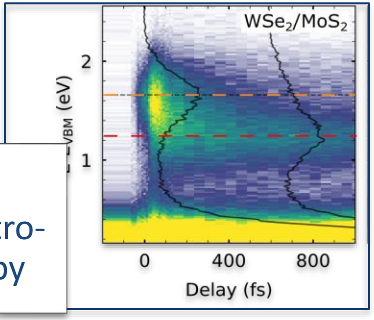


Nanoscale Dark-field Microscopy



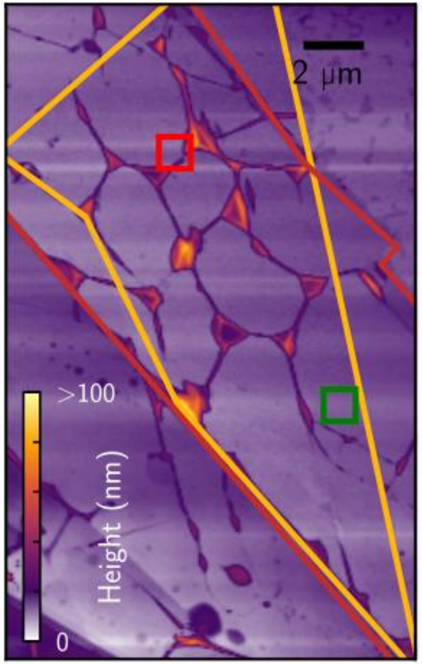
3D ARPES



fs-Spectro-scopy

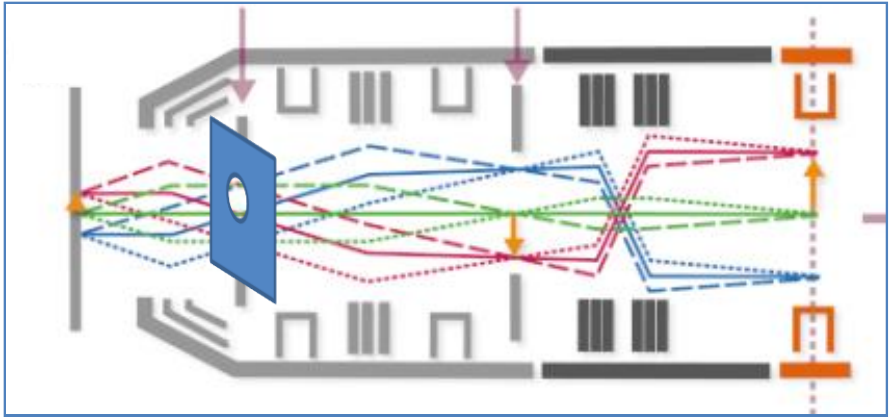
Ultrafast nano-imaging of dark excitons

AFM image

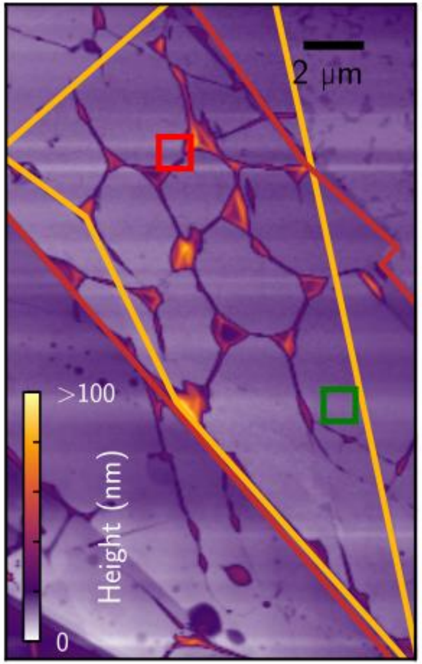


WSe_2/MoS_2 (with 30° twist)

- Heterogeneity in 2D quantum materials:
  - A major research challenge
- Objective: Ultrafast nanoimaging of dark excitons
- Solution: Dark field momentum microscopy

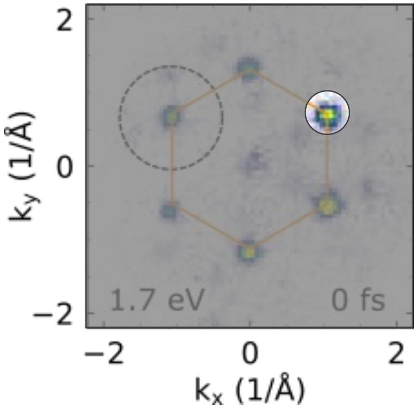


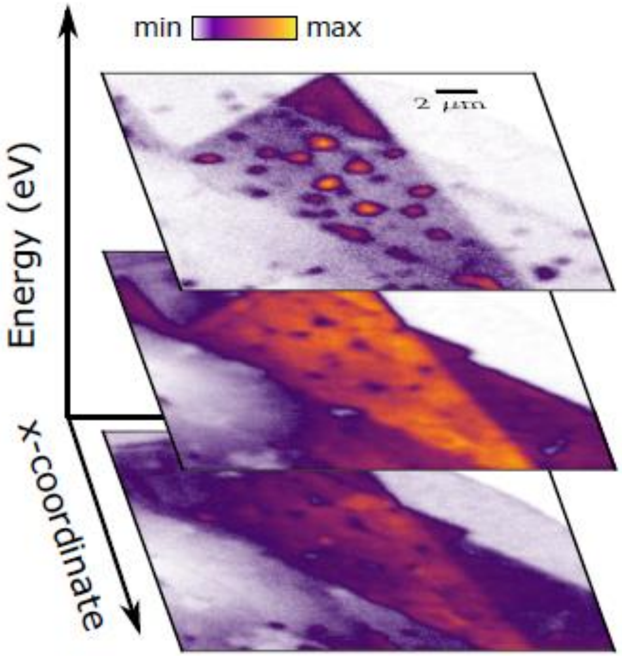
Dark-field imaging



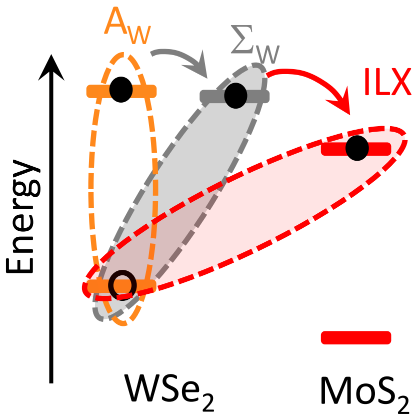
WSe_2/MoS_2 (with 30° twist)

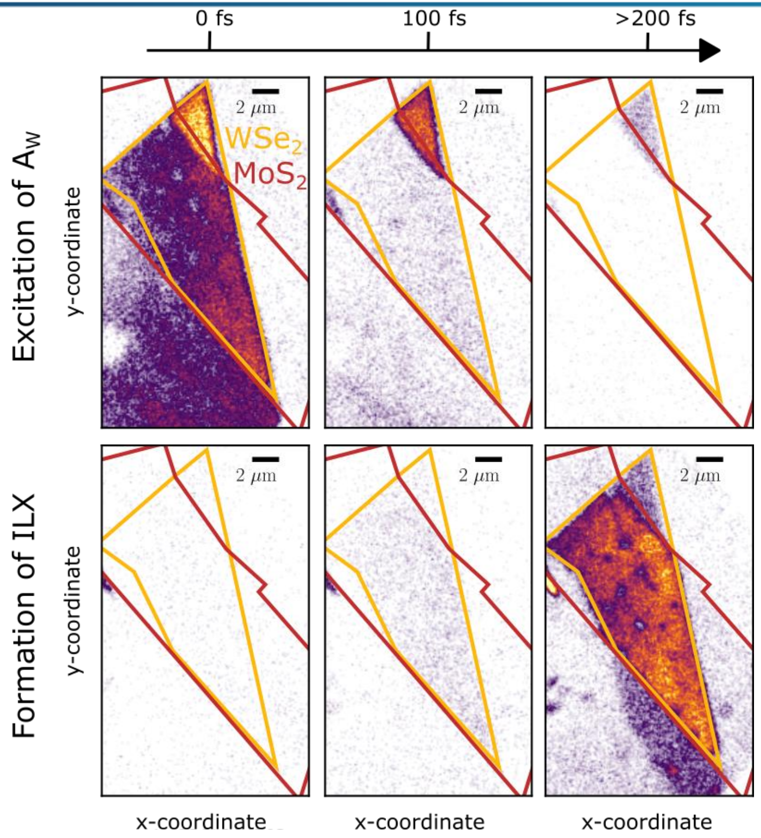
- Suitable apertures in the momentum microscope select specific states:
  - Exciton-specific imaging

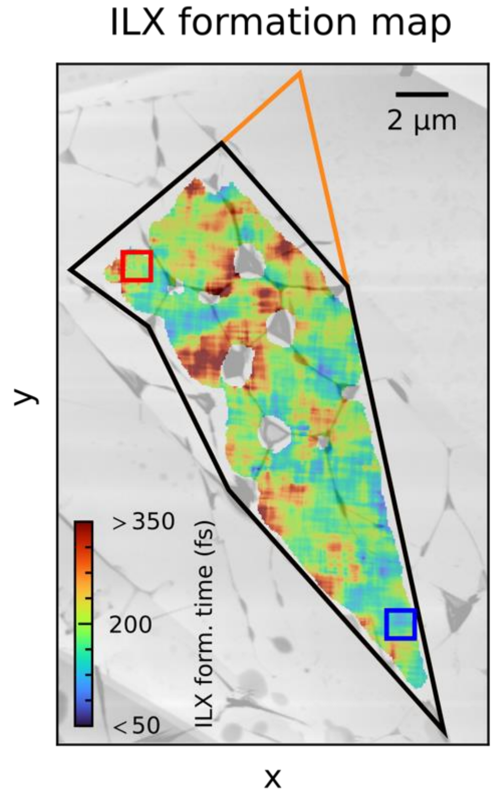




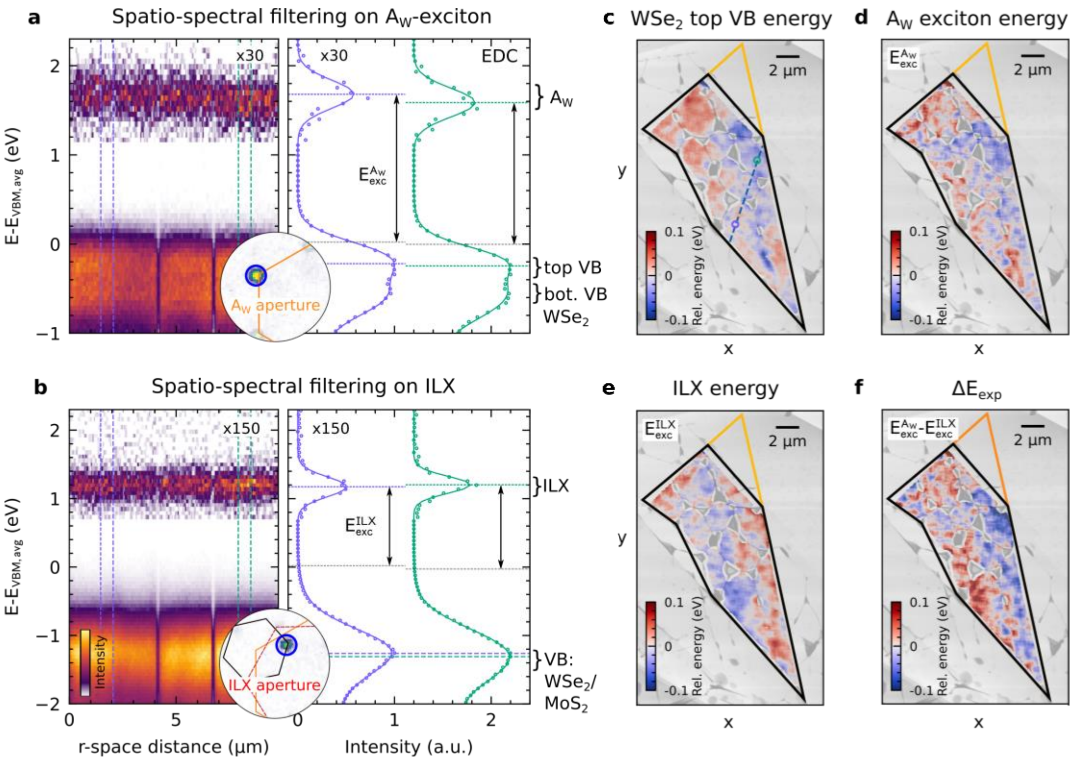
Resolving nanoscale dynamics

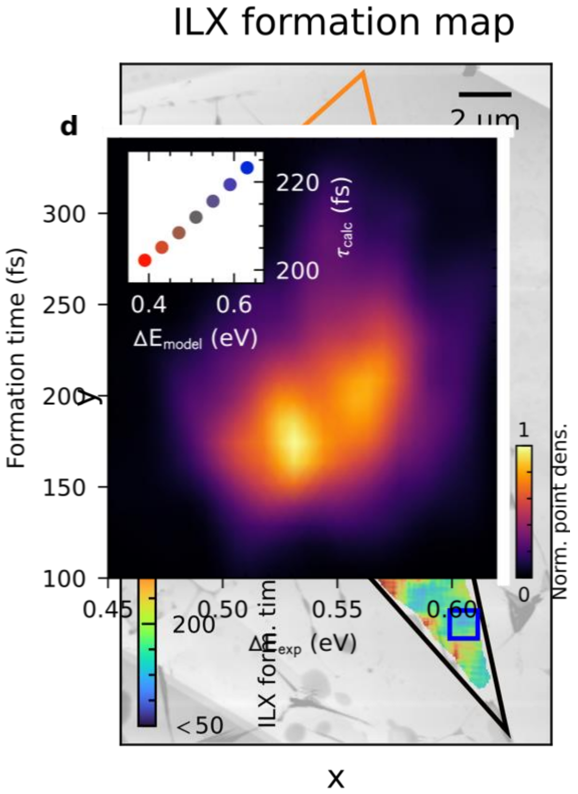




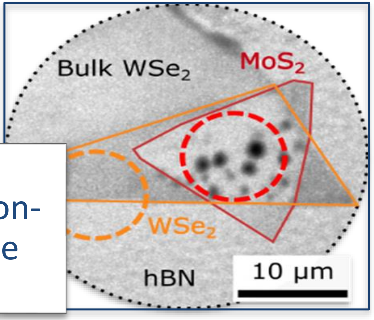


Correlating signatures





Conclusions



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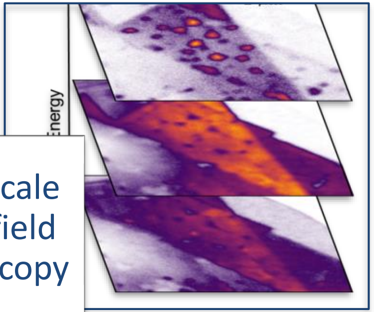
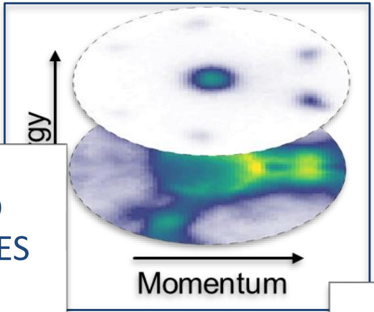
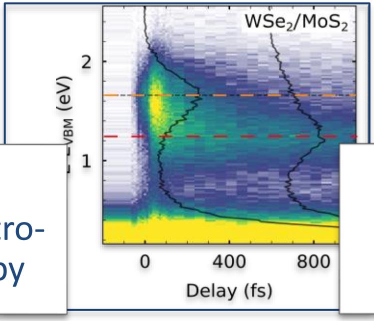


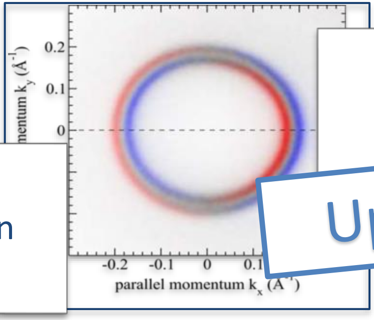
Nanoscale Dark-field Microscopy



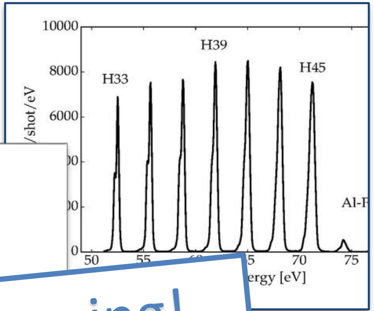
3D ARPES



fs-Spectro-scopy



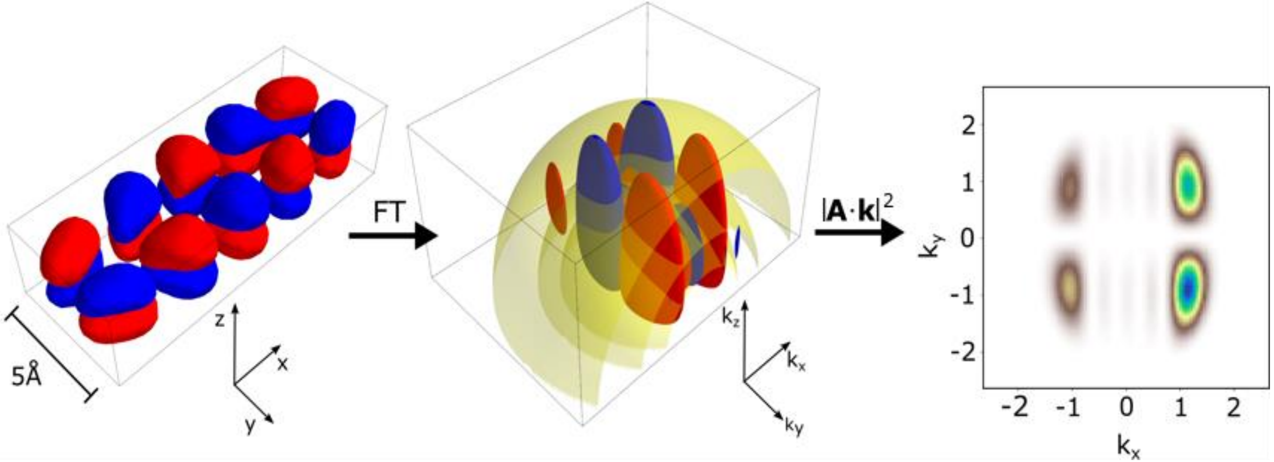
Spin



hν

Upcoming!

3D orbital images



$$I \propto \left|\boldsymbol{A}\cdot\boldsymbol{k}_f\right|^2 |\mathcal{F}(\psi)|^2 \times \delta(h\nu - E_B - \Phi - E_{kin})$$

Momentum distribution

Energy conservation

- Single photon energy: 2D information
- Many photon energies: 3D!
- HHG: ideal for 3D **and** fs time by time-compensating monochromation