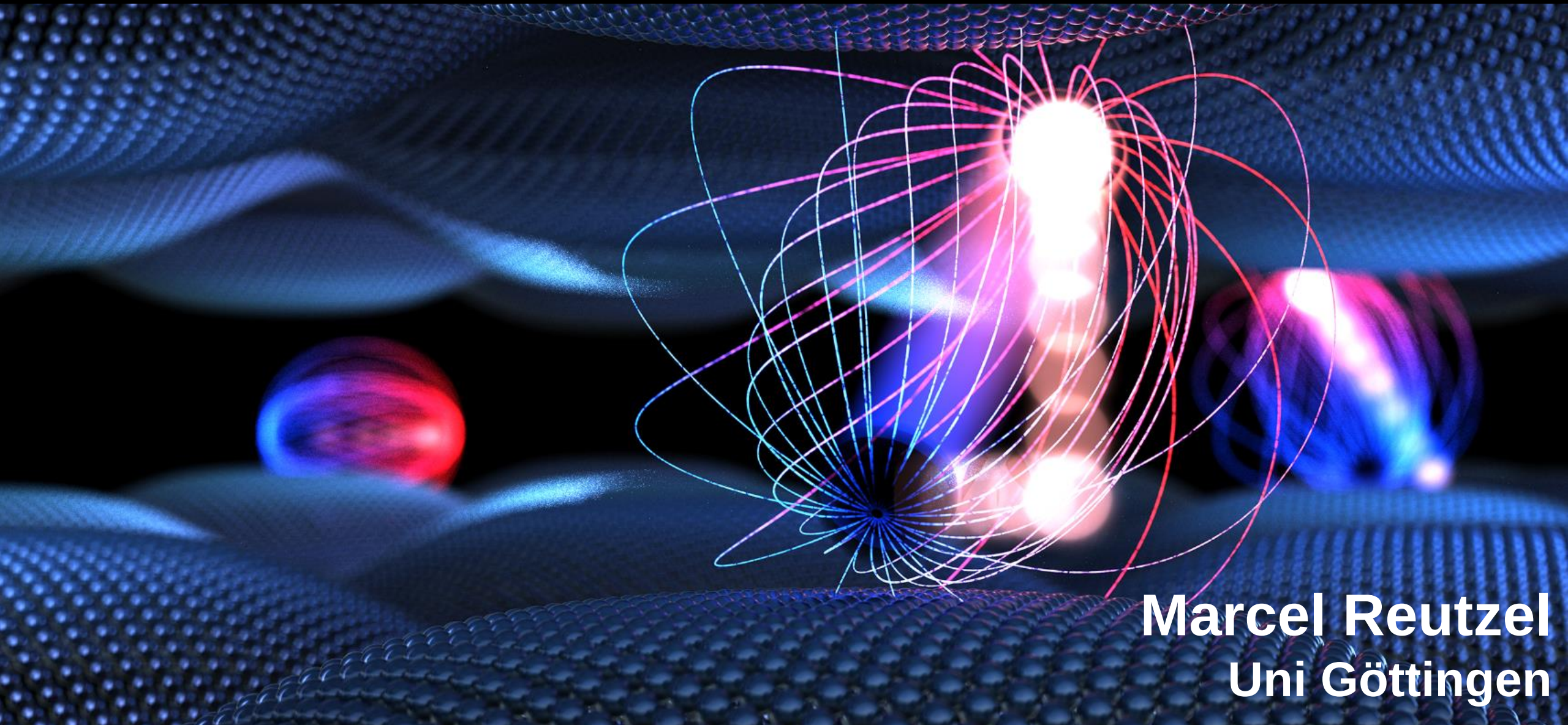
Ultrafast exciton dynamics in moiré heterostructures: A time-resolved momentum microscopy study



Marcel Reutzel
Uni Göttingen

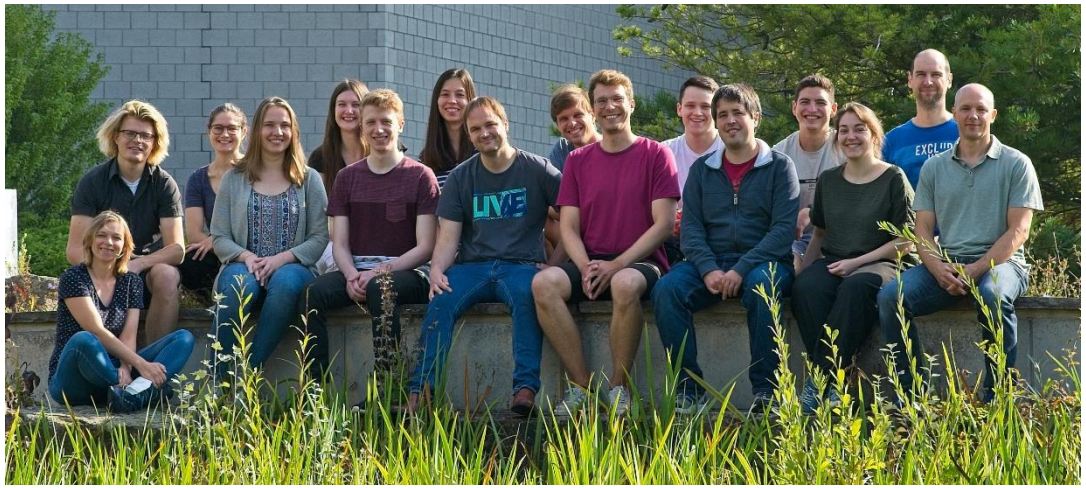
The team

GEORG-AUGUST-UNIVERSITÄT GÖTTINGEN IN PUBLICA COMMODA SEIT 1737

SFB 1073

CRC 1456 MATHEMATICS OF EXPERIMENT

AG Stefan Mathias, Göttingen



sample fabrication, characterization

- AbdulAziz AlMutairi
- Stephan Hofmann
- Thomas Weitz

organic semiconductors

- Peter Puschnig

UNI GRAZ

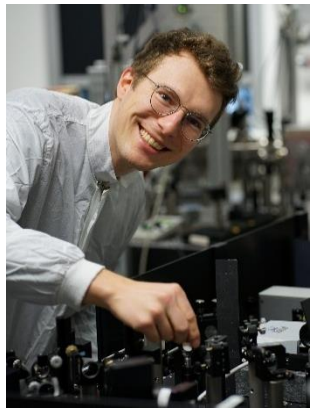
exciton dynamics

- Giuseppe Meneghini
- Samuel Brem
- Ermin Malic

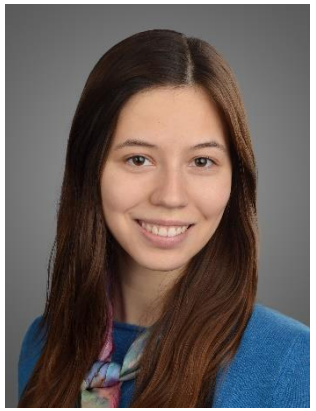
NEQ interactions

- Dino Novko

David Schmitt



Jan Philipp Bange

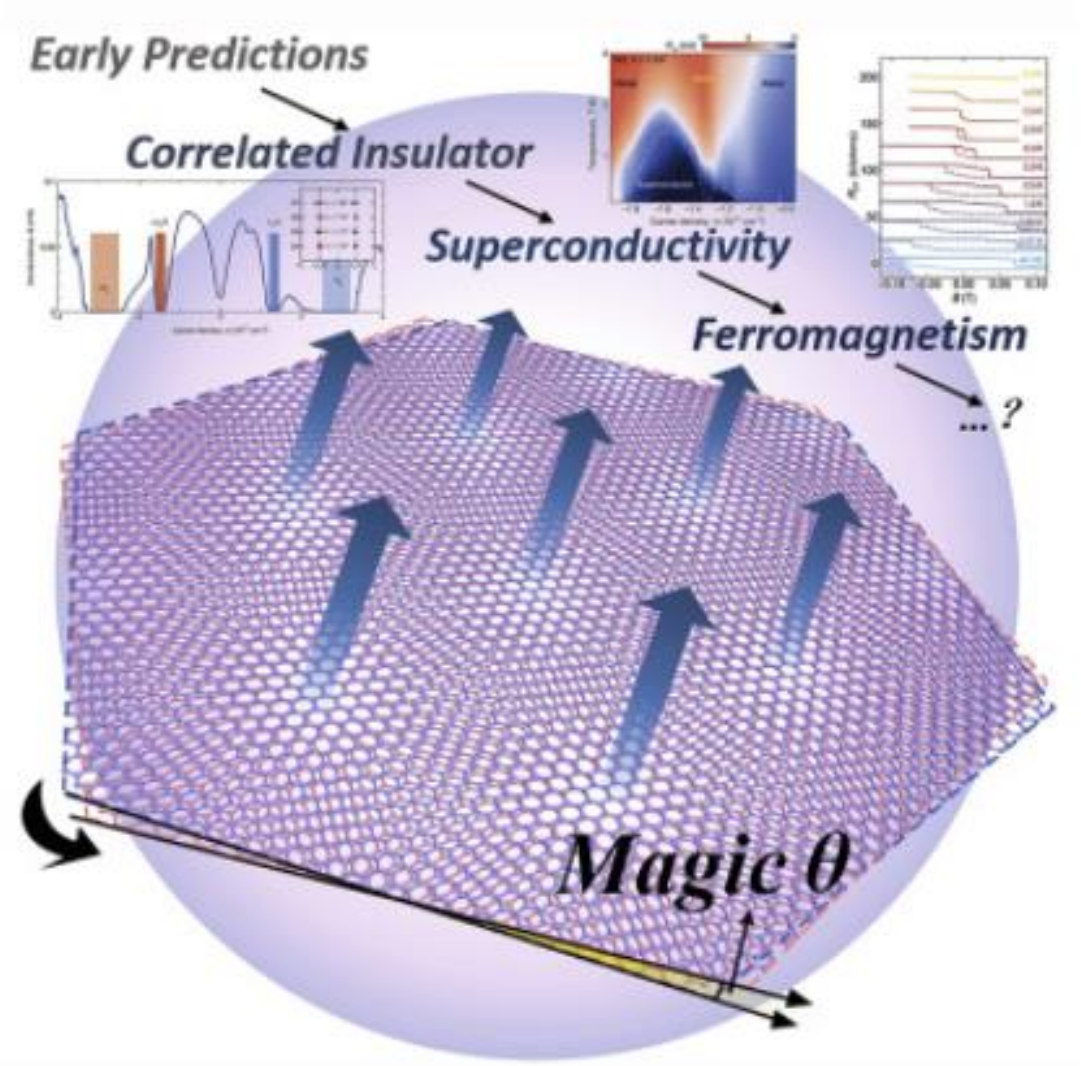


Wiebke Bennecke

Marco Merboldt

Matthijs Jansen

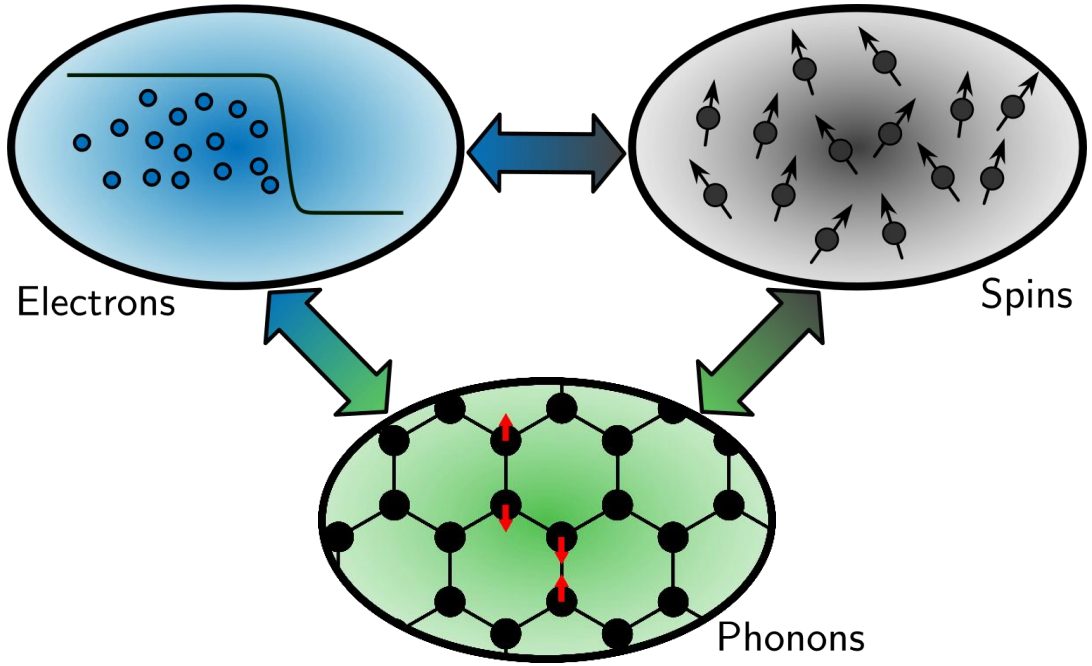
Microscopic interactions in solids



Matter, 2, 5, 6 May 2020, 1106-1114.

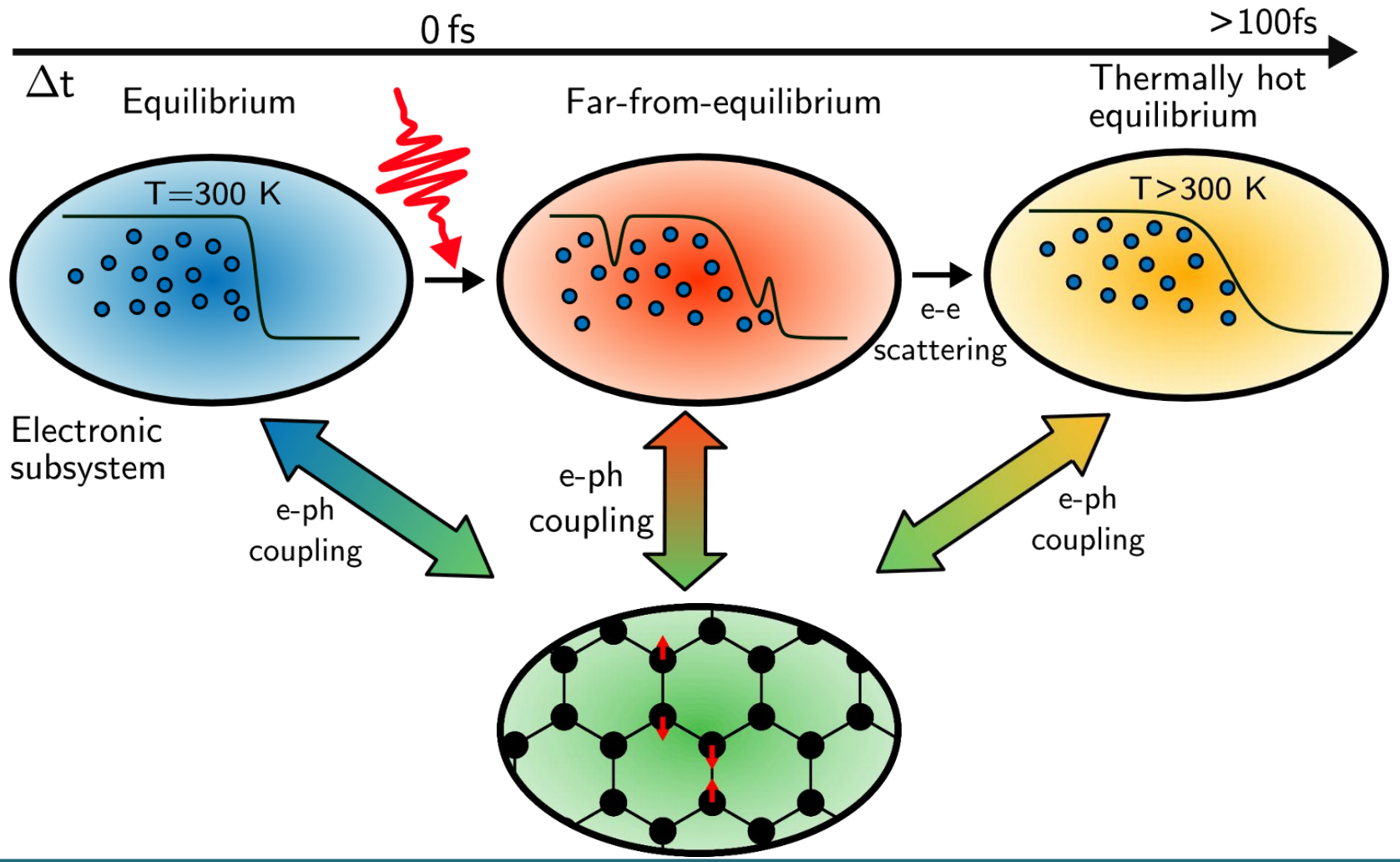
Quantum Materials

$$\hat{H} = H_{kin} + H_0 + H_{e-e} + H_{e-ph} + H_{SOC}$$



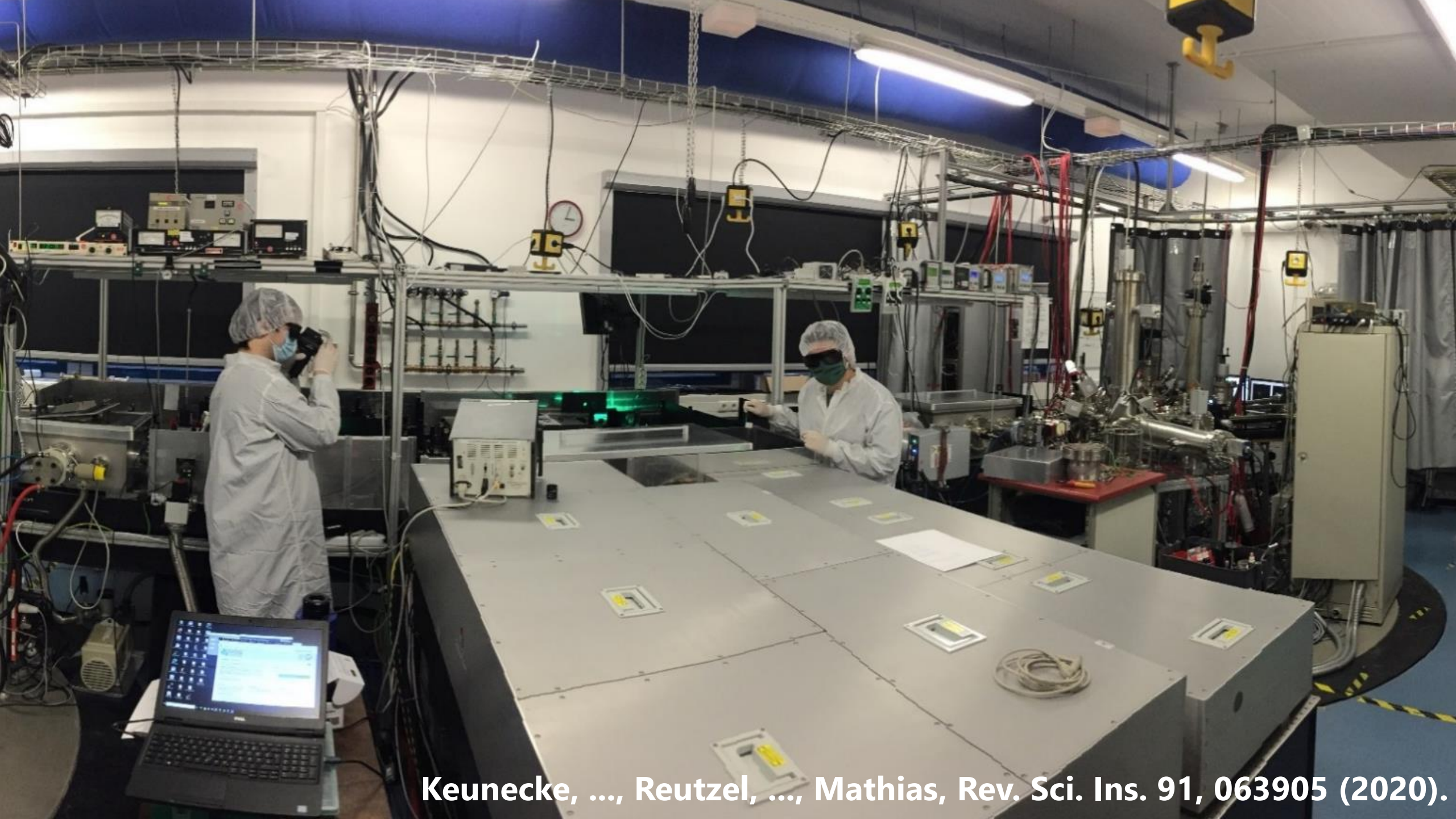
https://www.semiconductors.org/semiconductors-101/industry-impact/ 18.11.22

Far-from-equilibrium many-body interactions

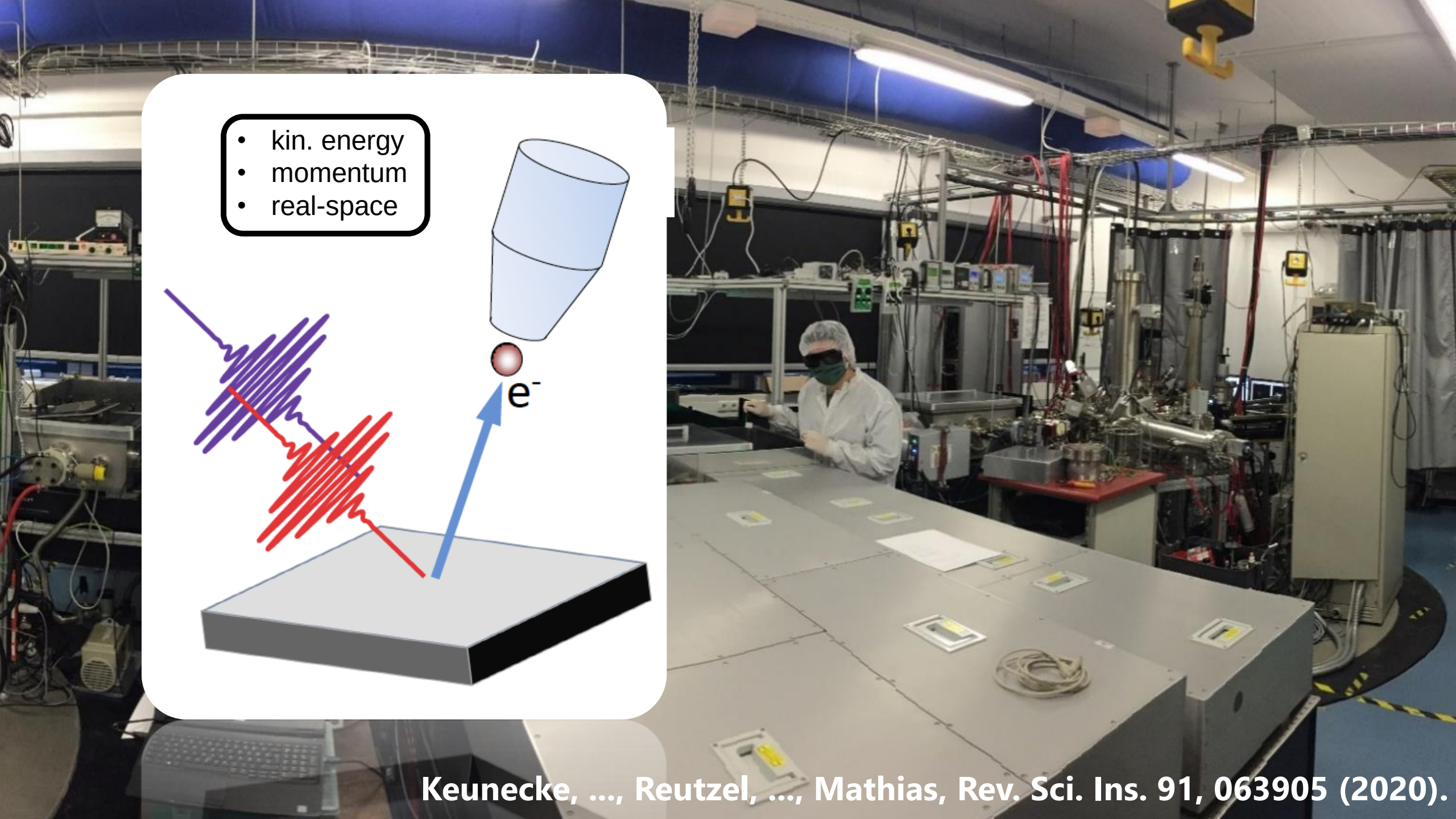


Typical Questions:

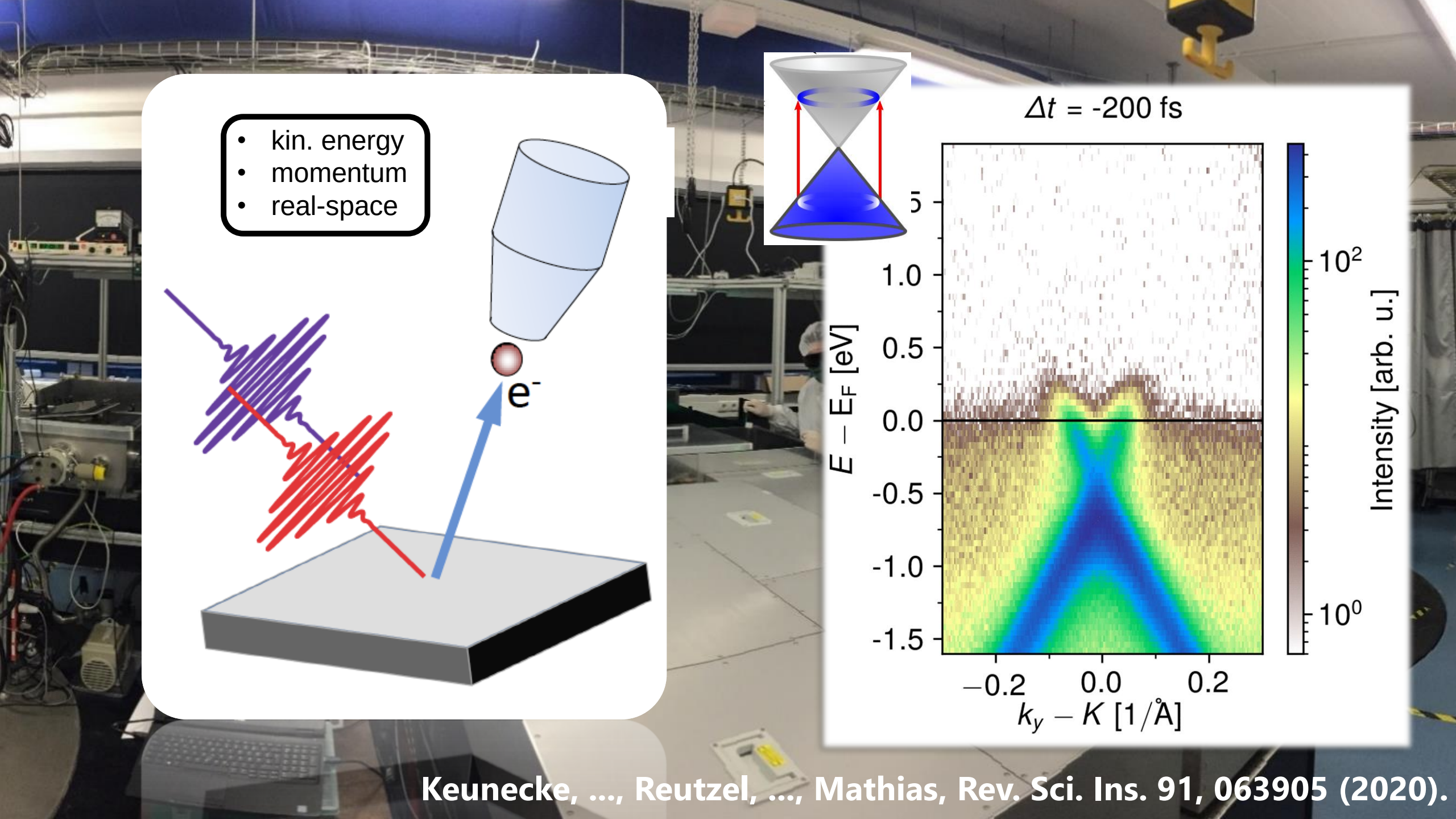
- How do we characterize matter far away from equilibrium?
- How do we control materials on-demand?



Keunecke, ..., Reutzel, ..., Mathias, Rev. Sci. Ins. 91, 063905 (2020).

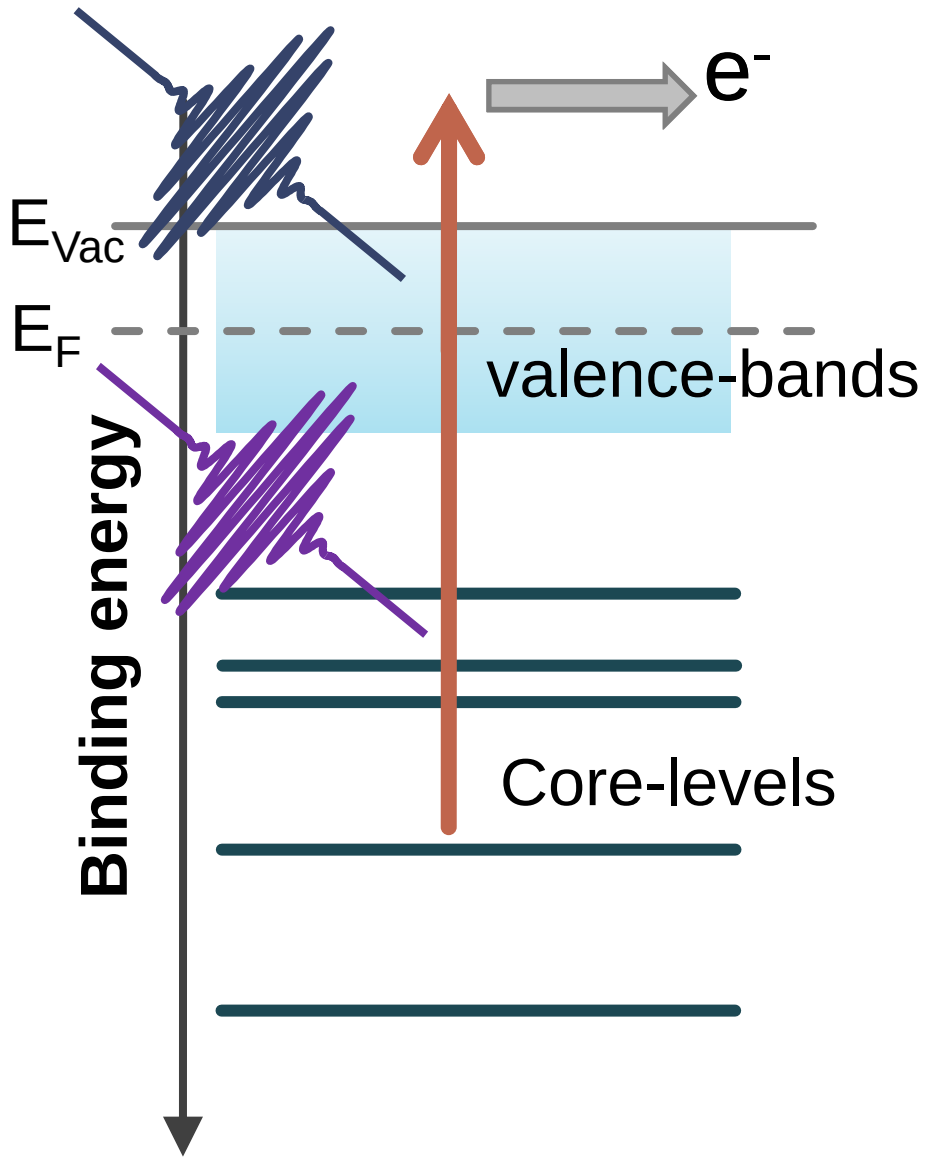


Keunecke, ..., Reutzel, ..., Mathias, Rev. Sci. Ins. 91, 063905 (2020).

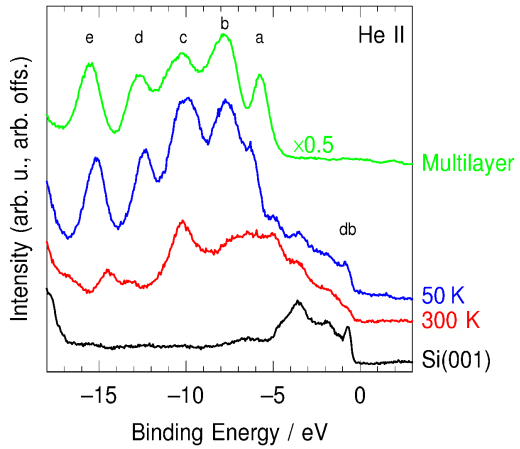


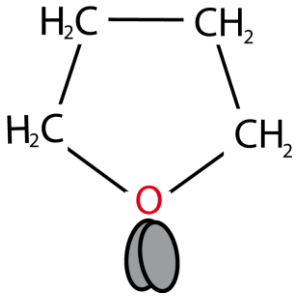
Keunecke, ..., Reutzel, ..., Mathias, Rev. Sci. Ins. 91, 063905 (2020).

Static photoelectron spectroscopy

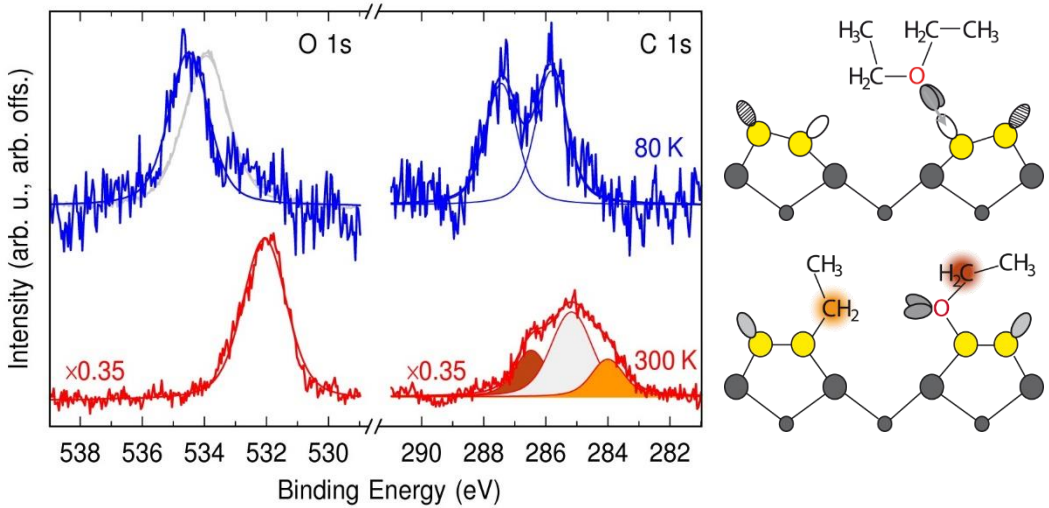


ultraviolet photoelectron spectroscopy





X-ray photoelectron spectroscopy:



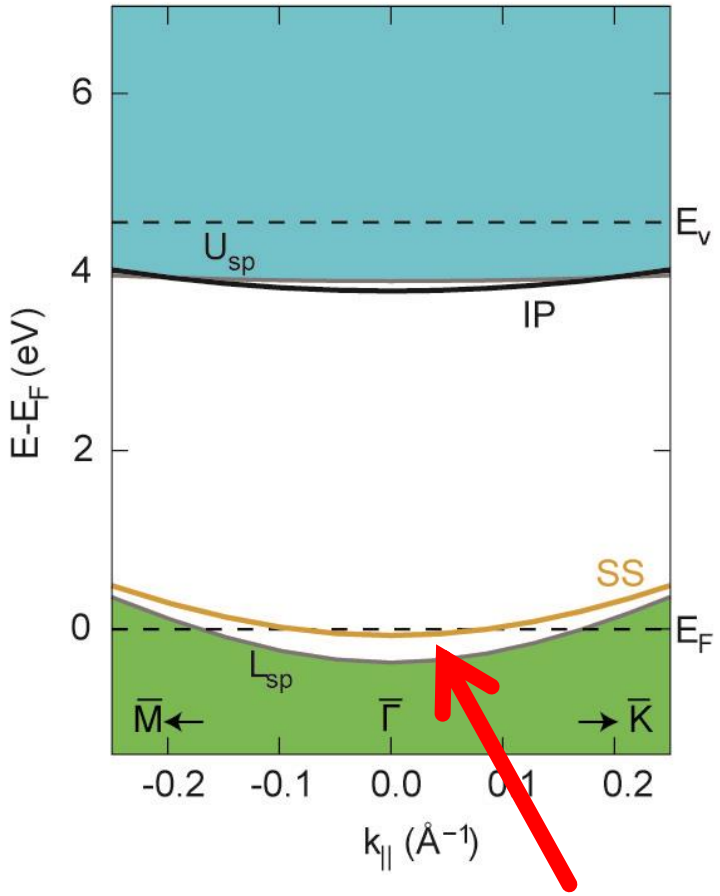
Reutzel, …, Höfer, *J. Phys. Chem. C* **119**, 6018 (2015).

Angle-resolved photoelectron spectroscopy

- **ARPES: powerful tool for new materials, many-body interactions, correlated phases…**

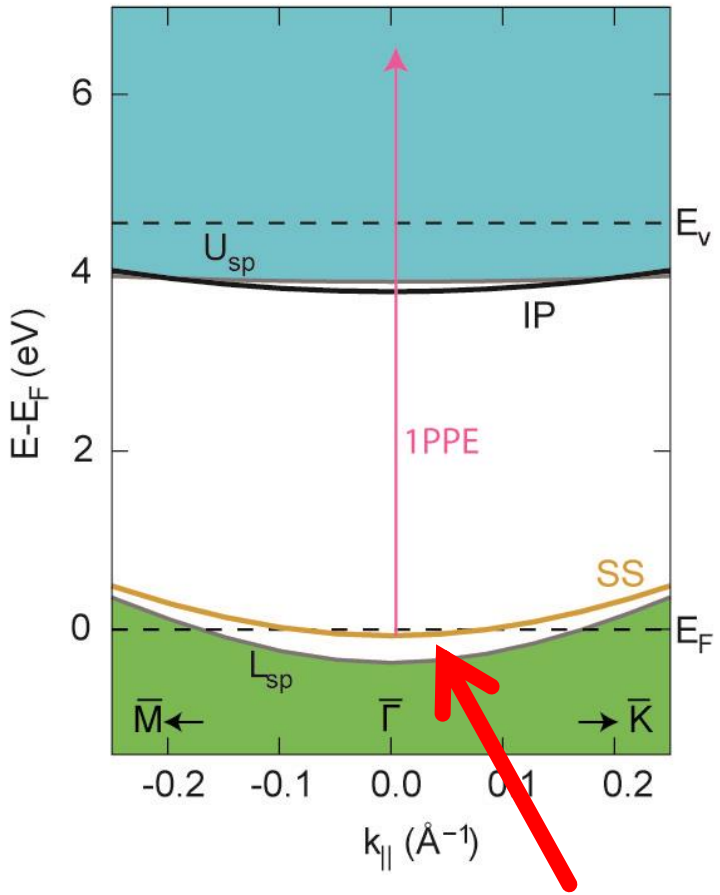
Angle-resolved photoelectron spectroscopy

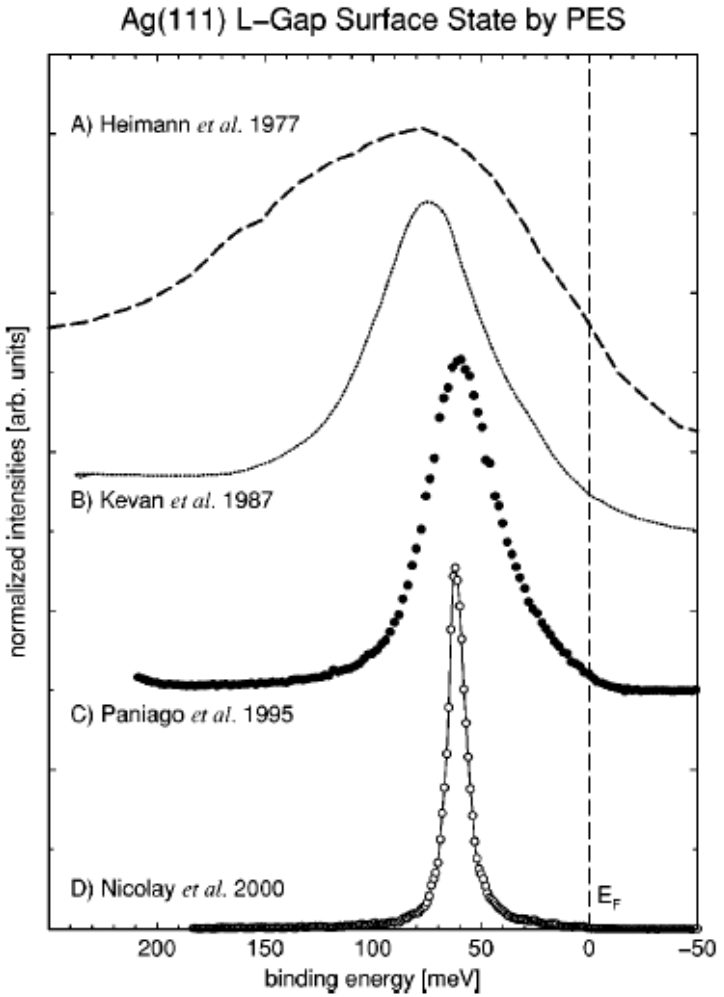
- **ARPES: powerful tool for new materials, many-body interactions, correlated phases…**



Angle-resolved photoelectron spectroscopy

- **ARPES: powerful tool for new materials, many-body interactions, correlated phases…**

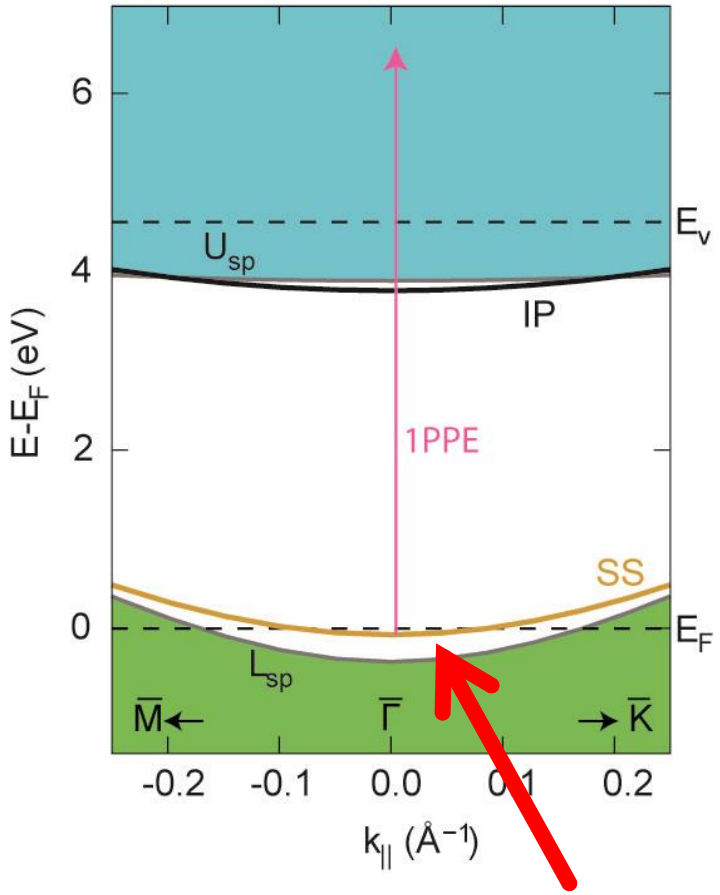


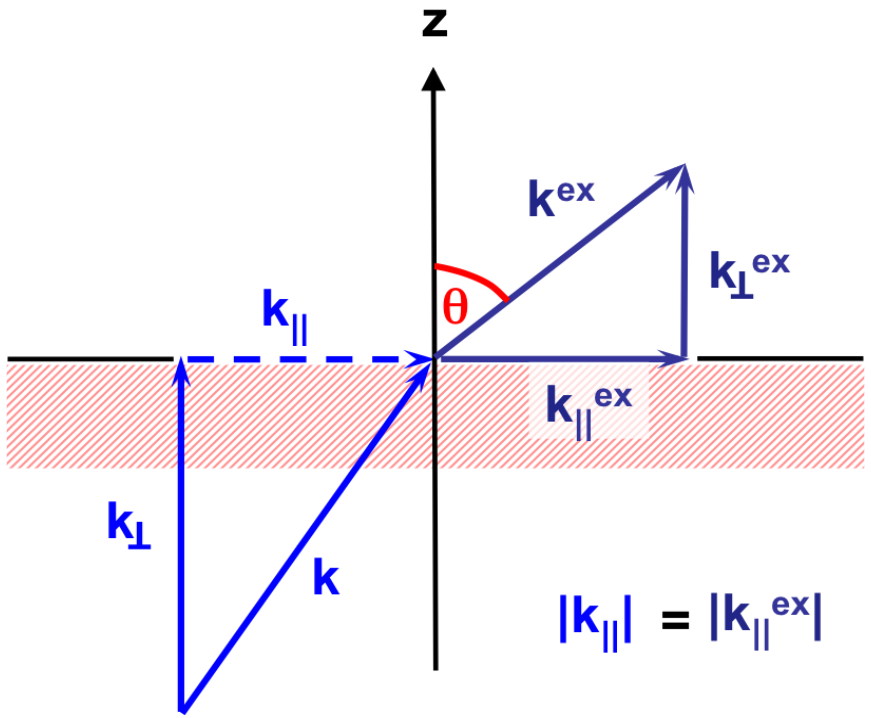


Reinert *et al.*, PRB **63**, 115415 (2001).

Angle-resolved photoelectron spectroscopy

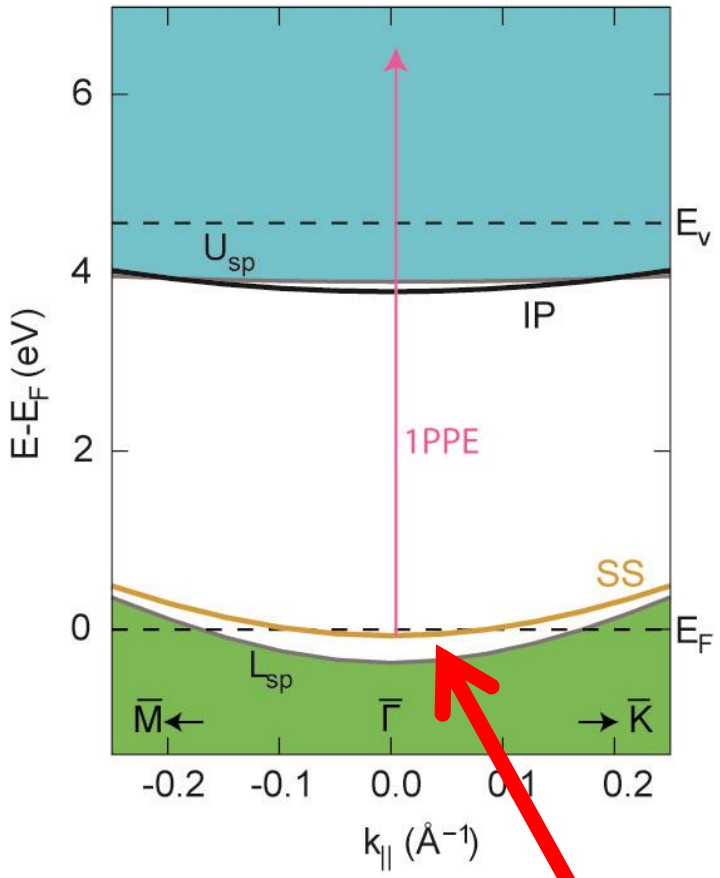
- **ARPES: powerful tool for new materials, many-body interactions, correlated phases…**

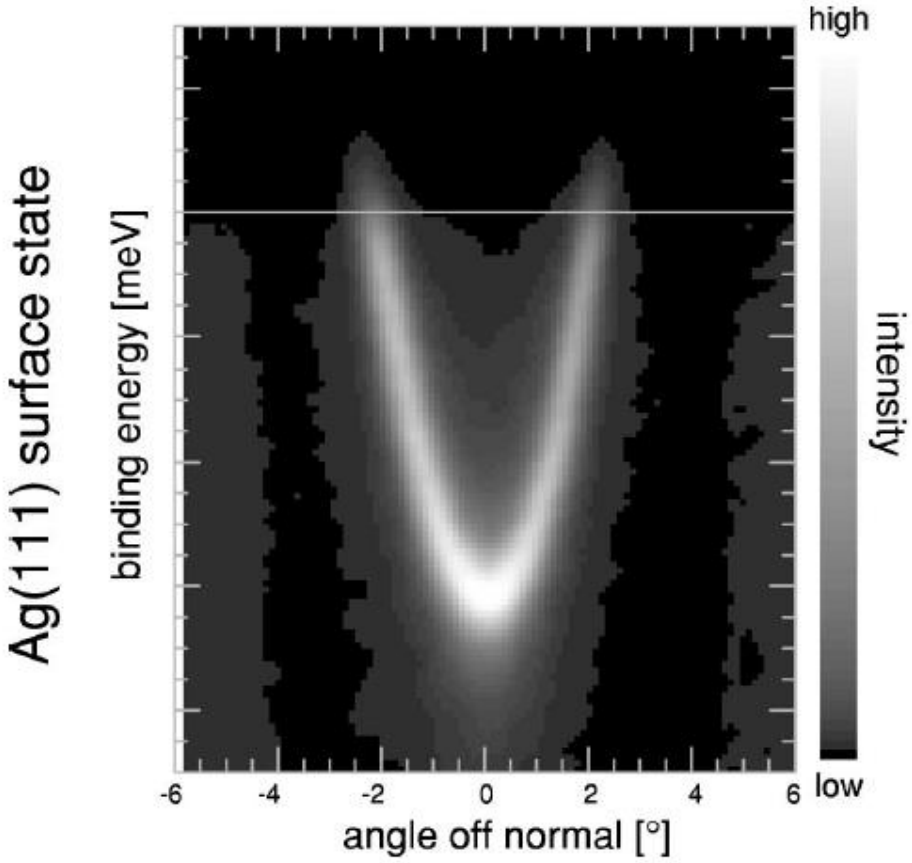




$$\vec{k}_{\parallel}^{ex} = \sqrt{\frac{2m_e}{\hbar^2}}\sqrt{h\nu - |E_b| - W}\,\sin(\Theta) = \sqrt{\frac{2m_e}{\hbar^2}E_{kin}}\,\sin(\Theta)$$

Angle-resolved photoelectron spectroscopy

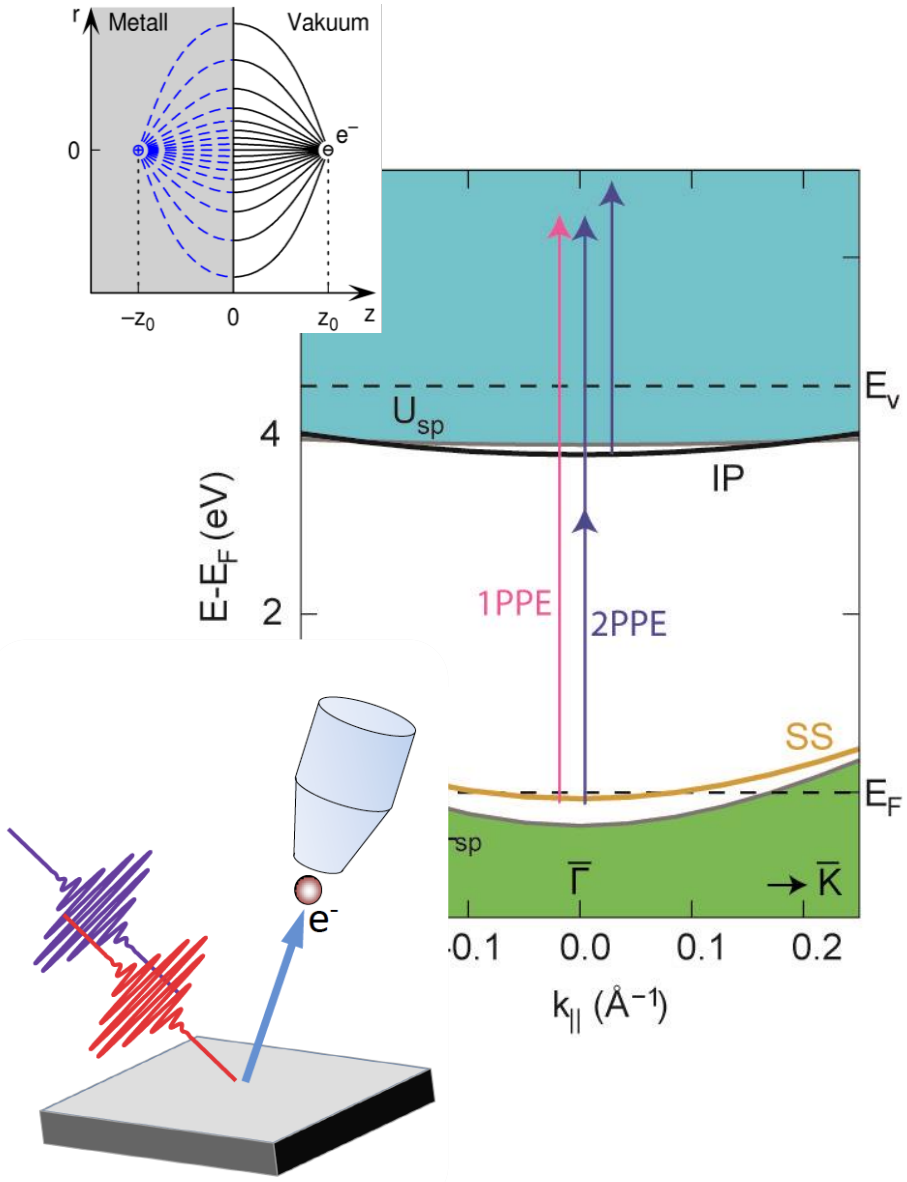


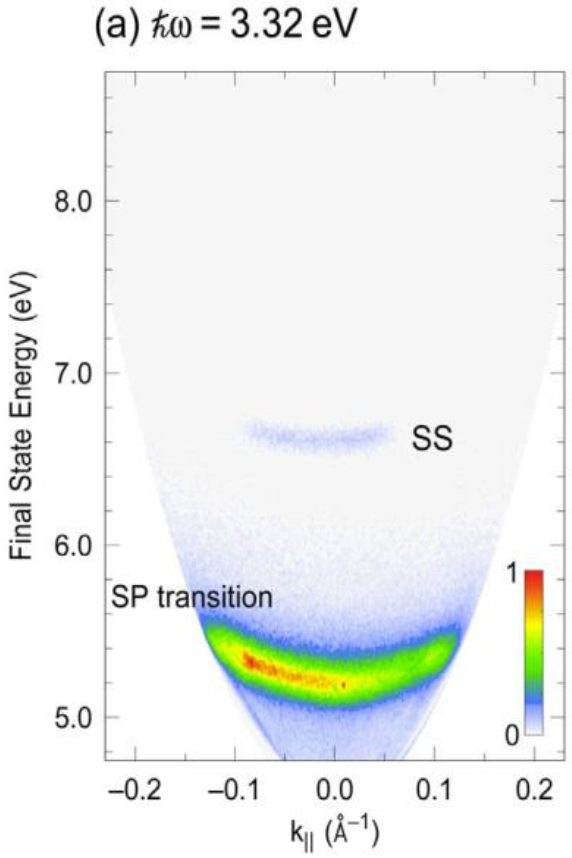


$$E_{SS}(k_{||}) = \frac{\hbar^2}{2m_e m_{\text{eff},SS}} k_{||}^2$$

Reinert *et al.*, PRB **63**, 115415 (2001).

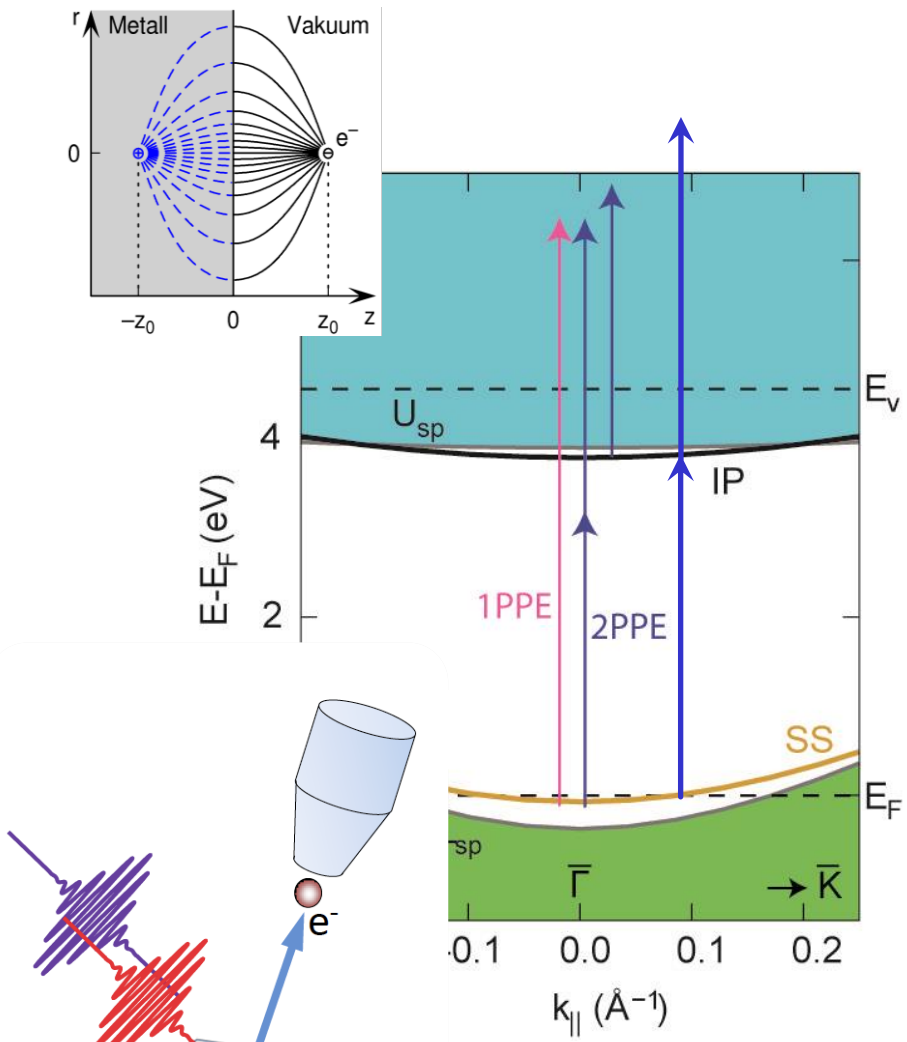
Time-resolved Two-photon Photoemission (TR-2PPE)

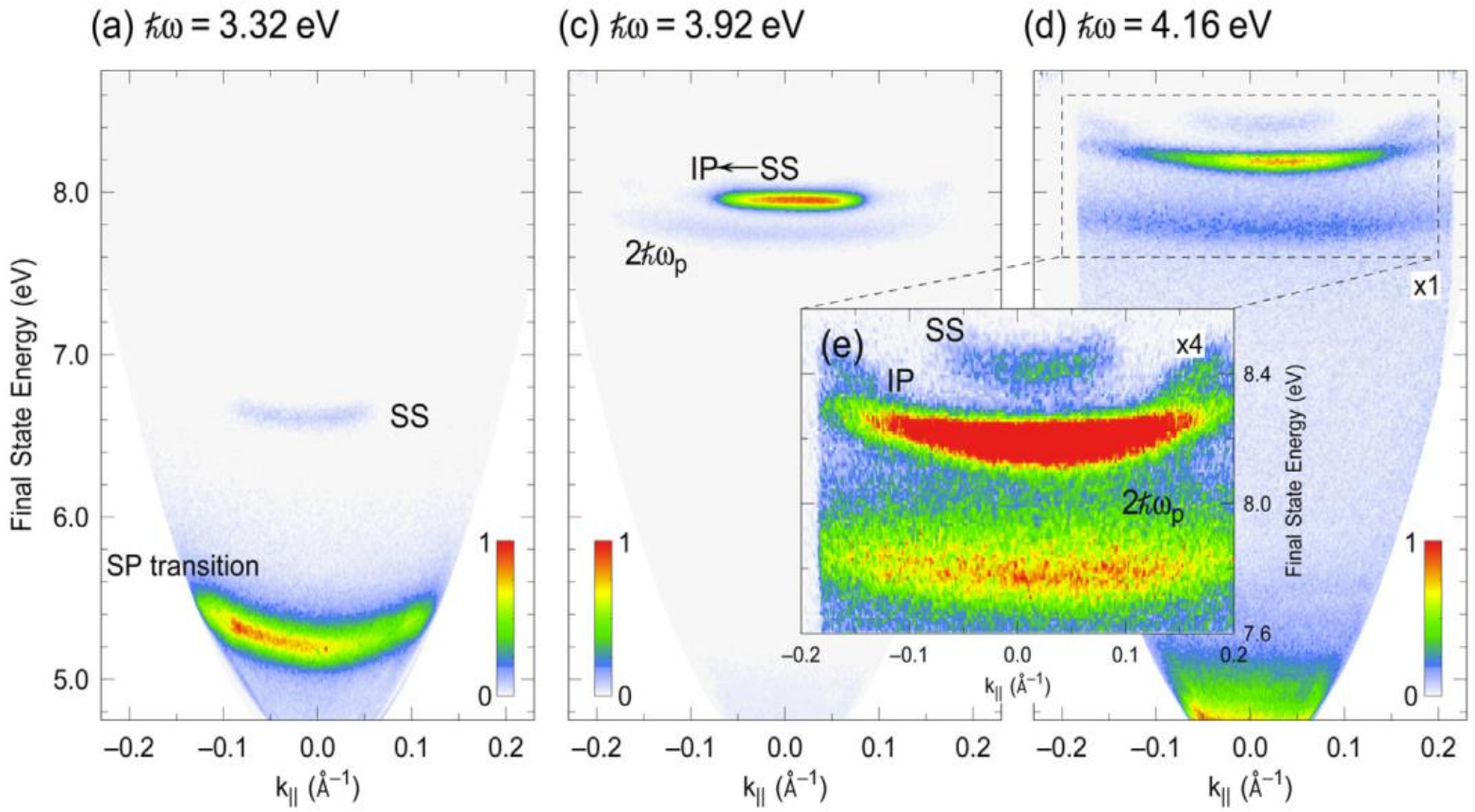




Reutzel, …, Petek, PRL **123**, 017404 (2019).

Time-resolved Two-photon Photoemission (TR-2PPE)

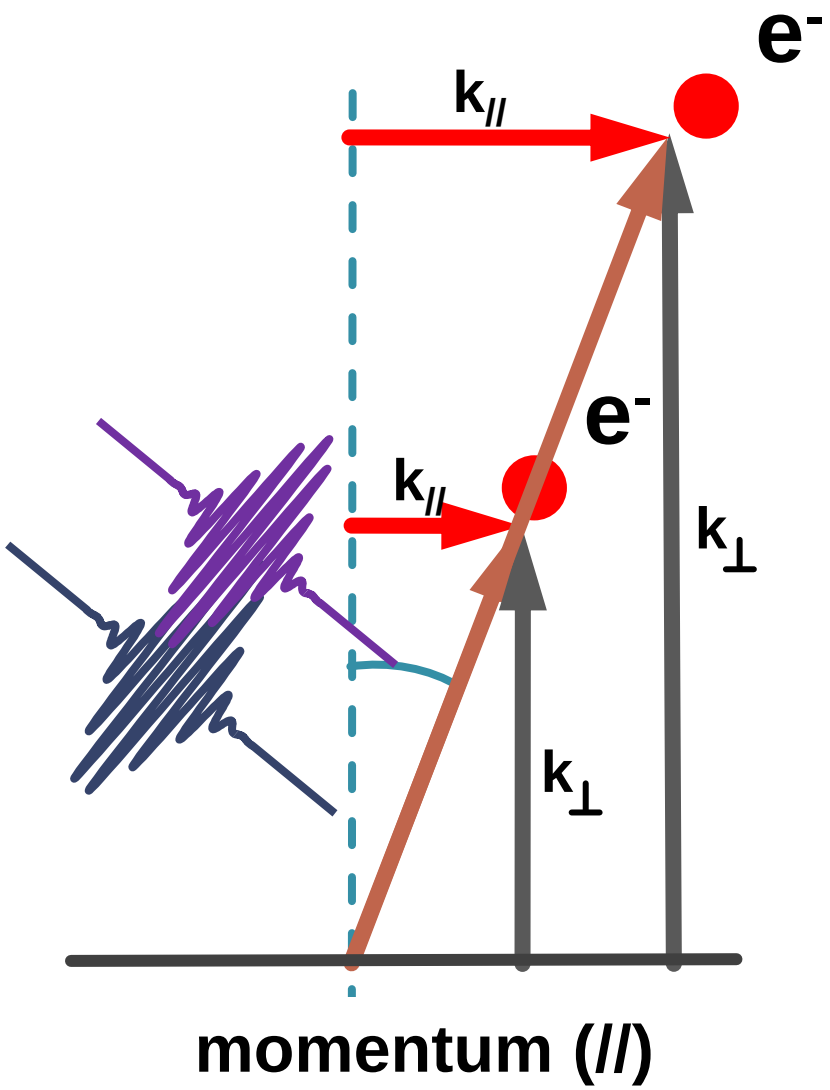


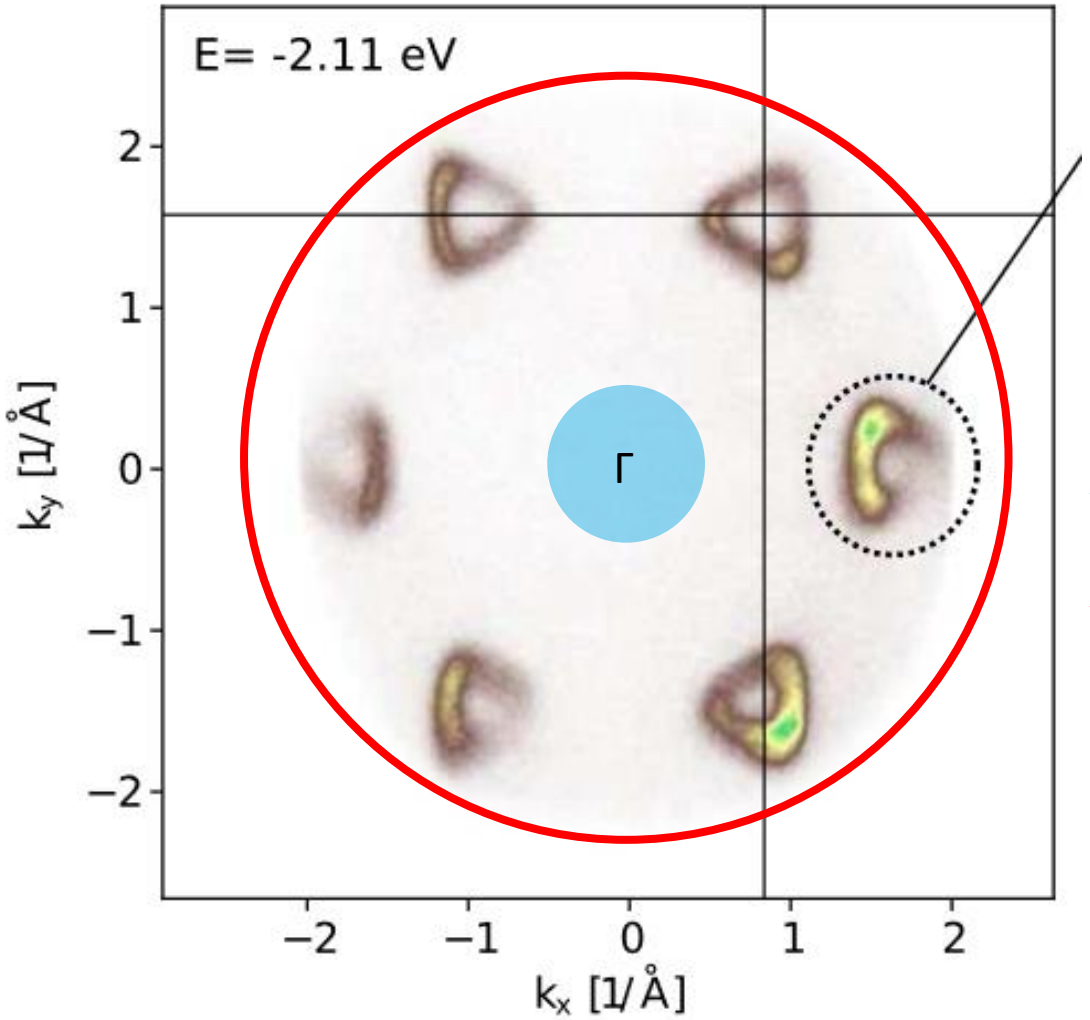


Reutzel, …, Petek, PRL **123**, 017404 (2019).

The experimental challenge…

so far: focus on femtosecond dynamics around the Γ point → Why?

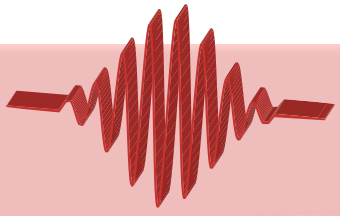
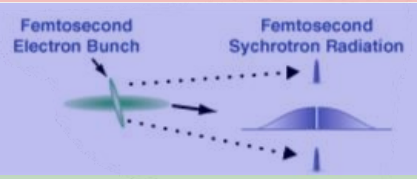
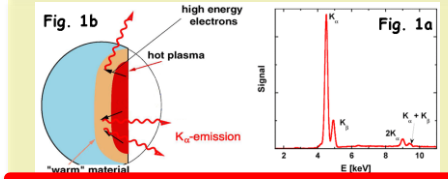


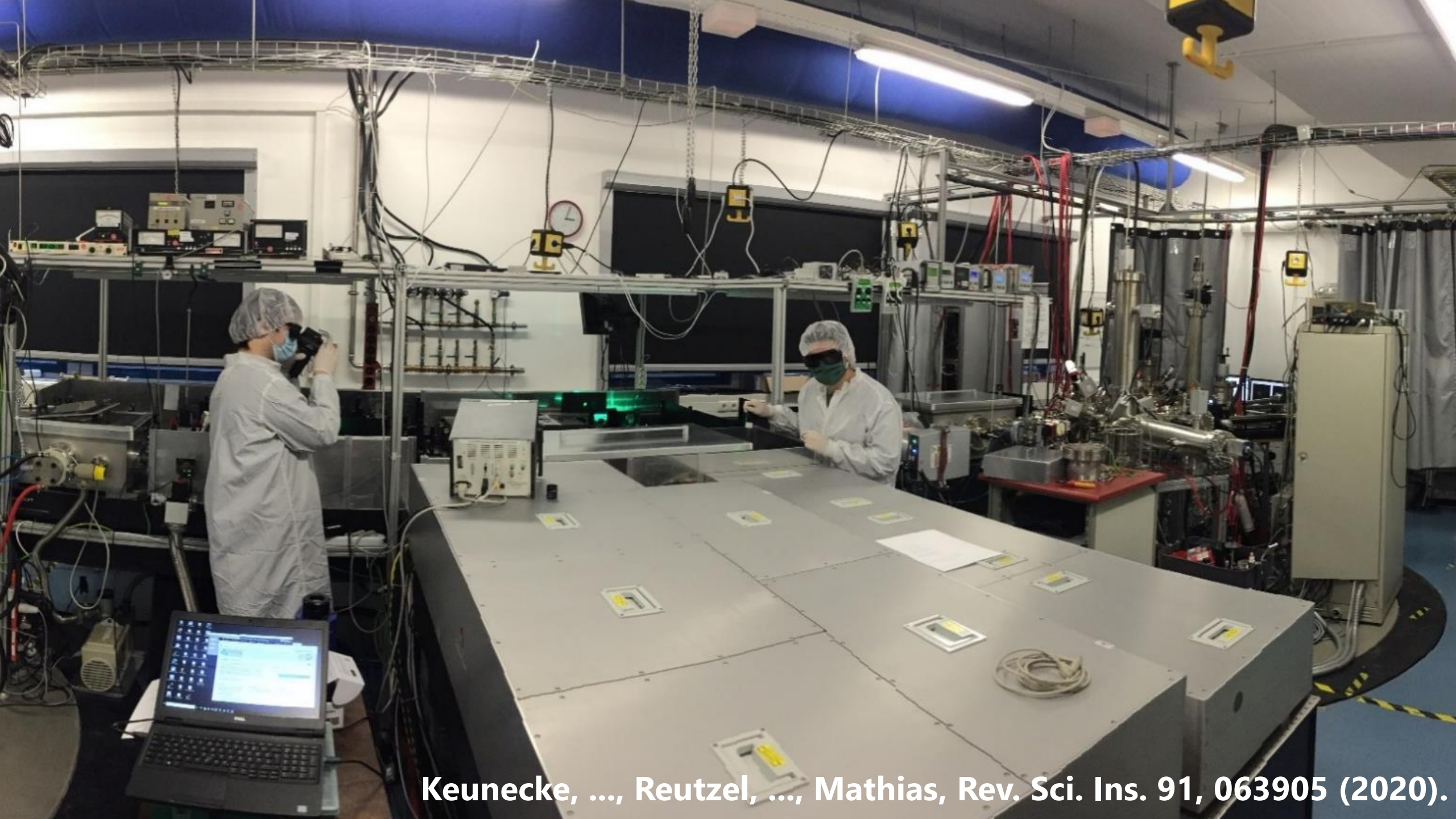


$\hbar\omega_{Probe}$ = 7 eV

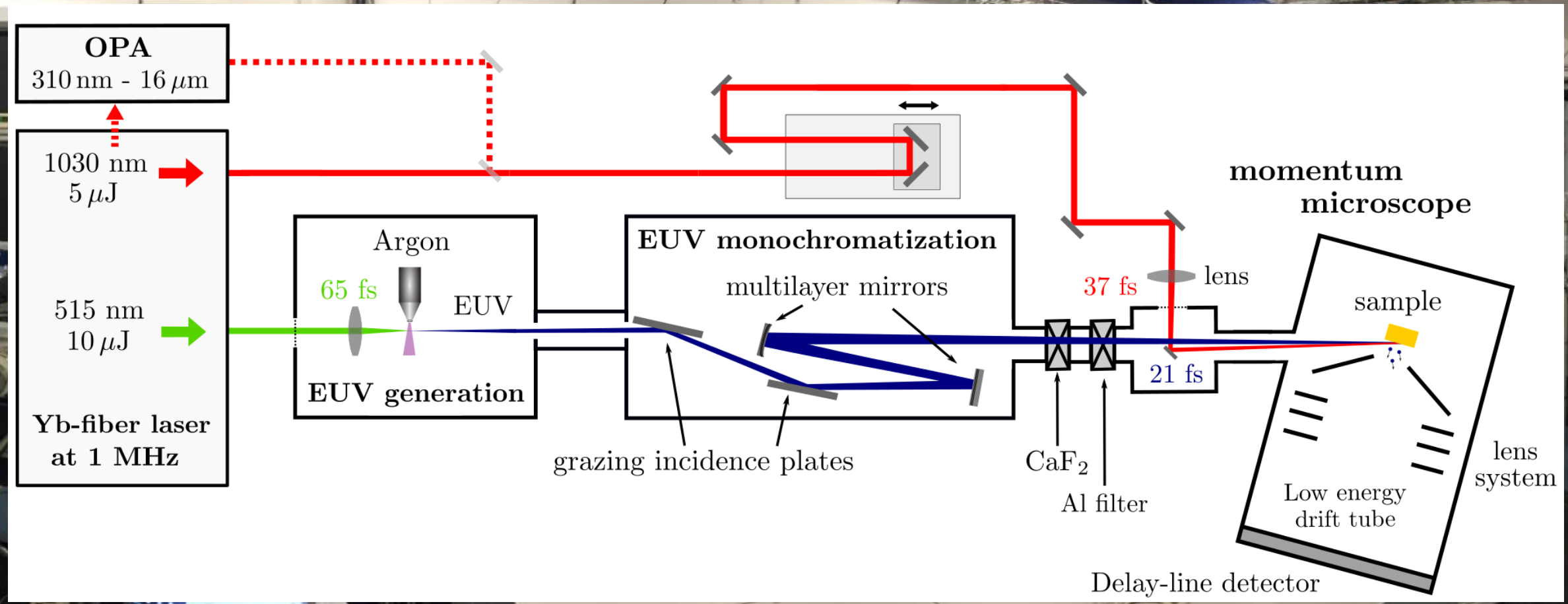
$\hbar\omega_{Probe}$ > 20 eV

Creating femtosecond XUV pulses

		fs pulses	XUV or X-ray	Tabletop
	Femtosecond laser	✓	✗	✓
	Femto-sliced Synchrotron	✓ (100 fs)	✓	✗
	Free-electron Laser	✓ (10-100 fs)	✓	✗
	Laser-plasma source	✓ (100 fs)	✓	✓
	High harmonics	✓ (<10 fs)	✓	✓

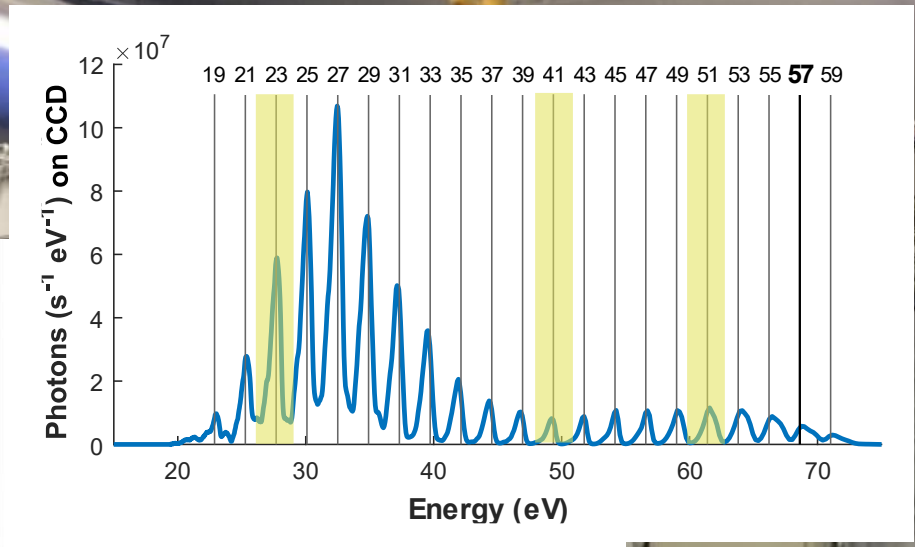


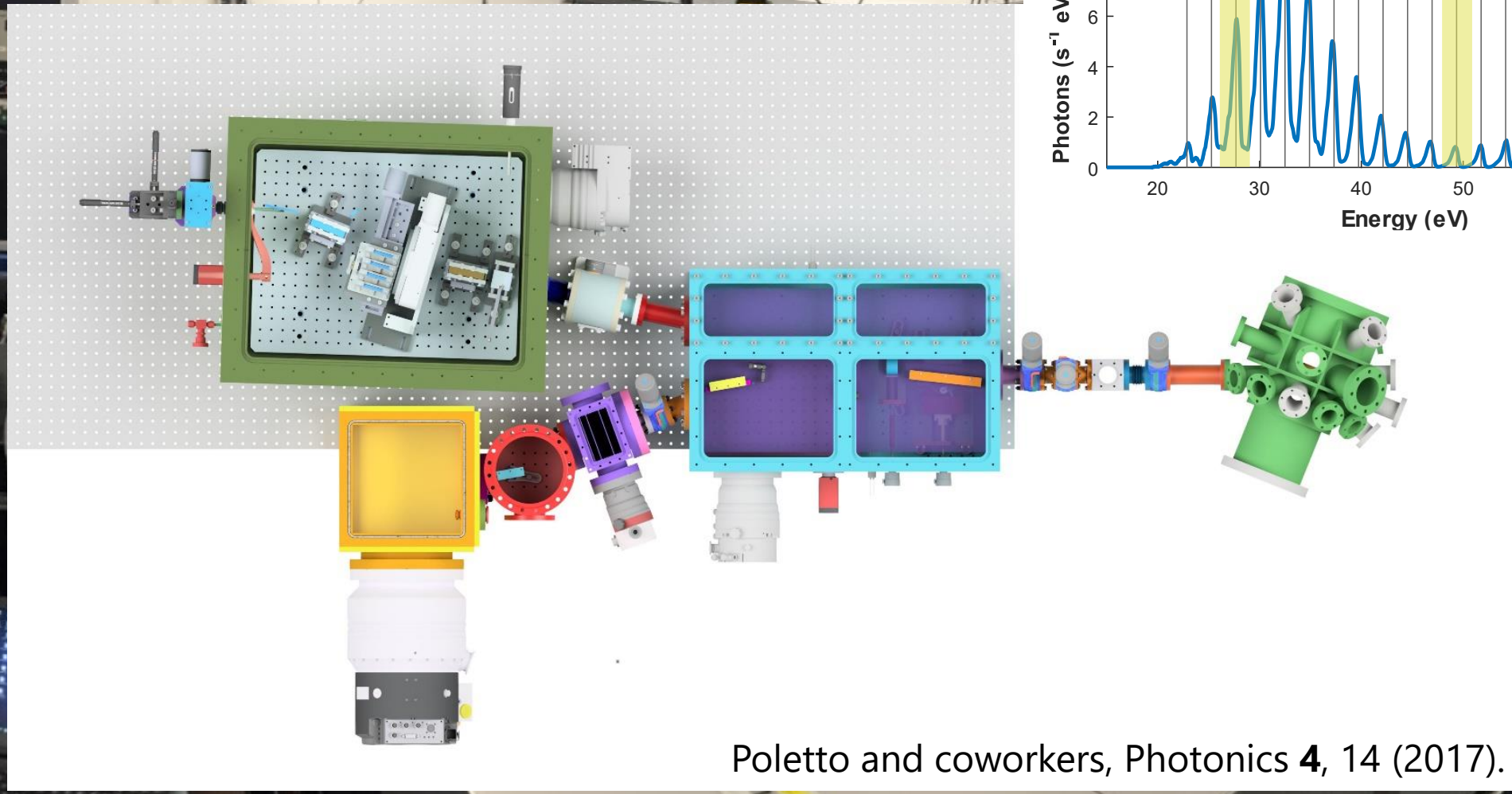
Keunecke, ..., Reutzel, ..., Mathias, Rev. Sci. Ins. 91, 063905 (2020).



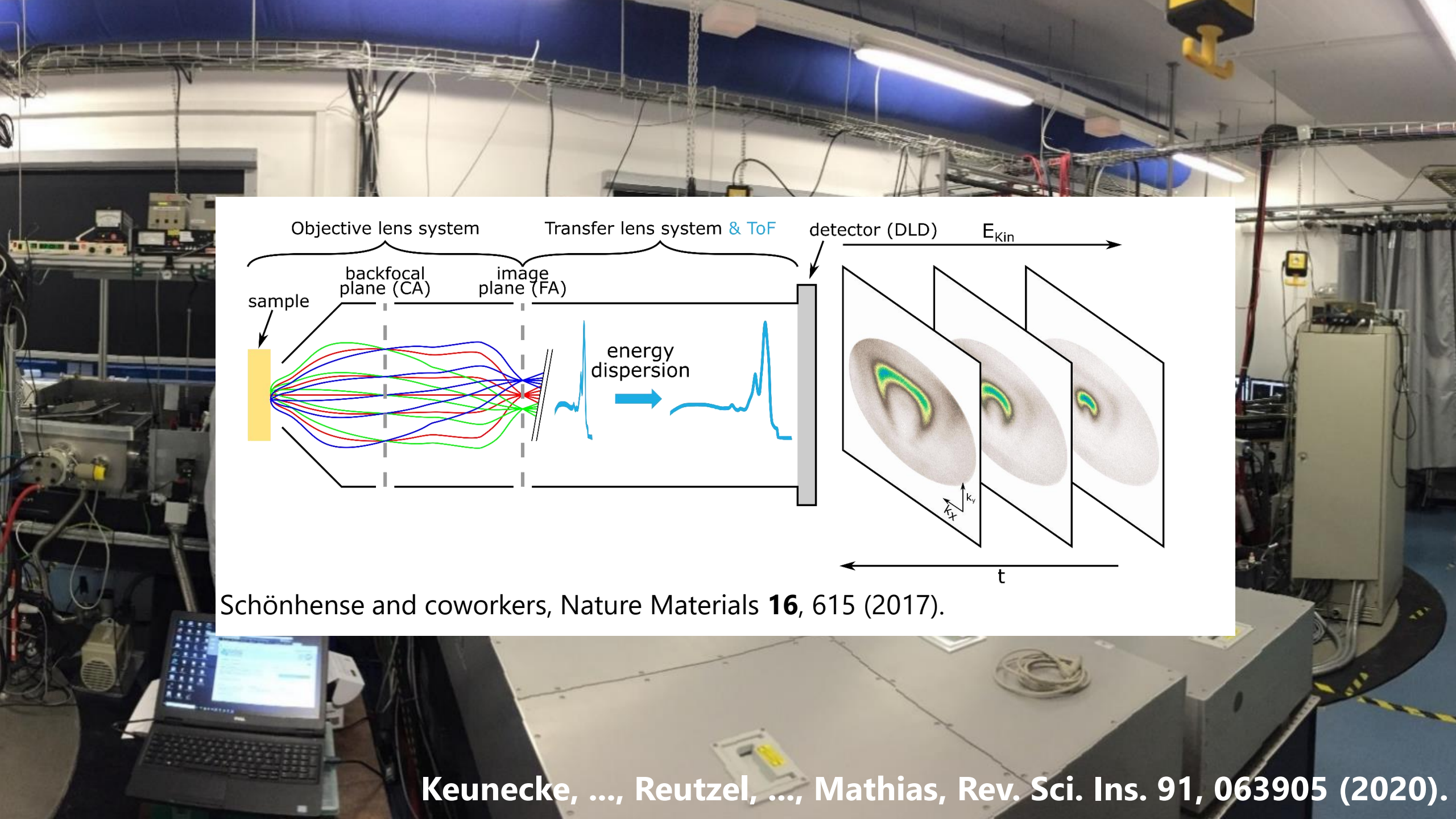
Keunecke, ..., Reutzel, ..., Mathias, Rev. Sci. Ins. 91, 063905 (2020).

soon...



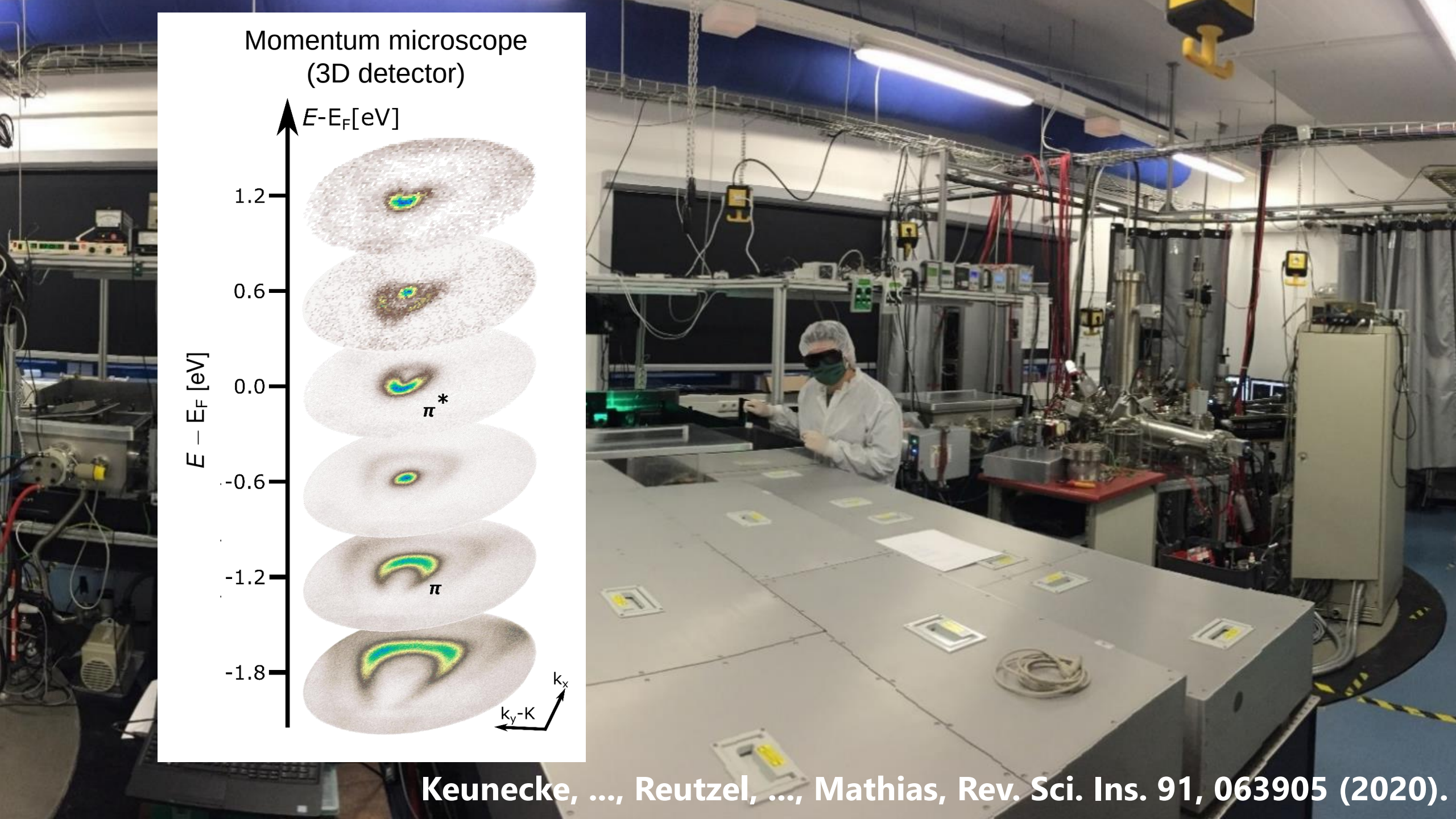


Poletto and coworkers, Photonics **4**, 14 (2017).



Schönhense and coworkers, Nature Materials **16**, 615 (2017).

Keunecke, ..., Reutzel, ..., Mathias, Rev. Sci. Ins. 91, 063905 (2020).



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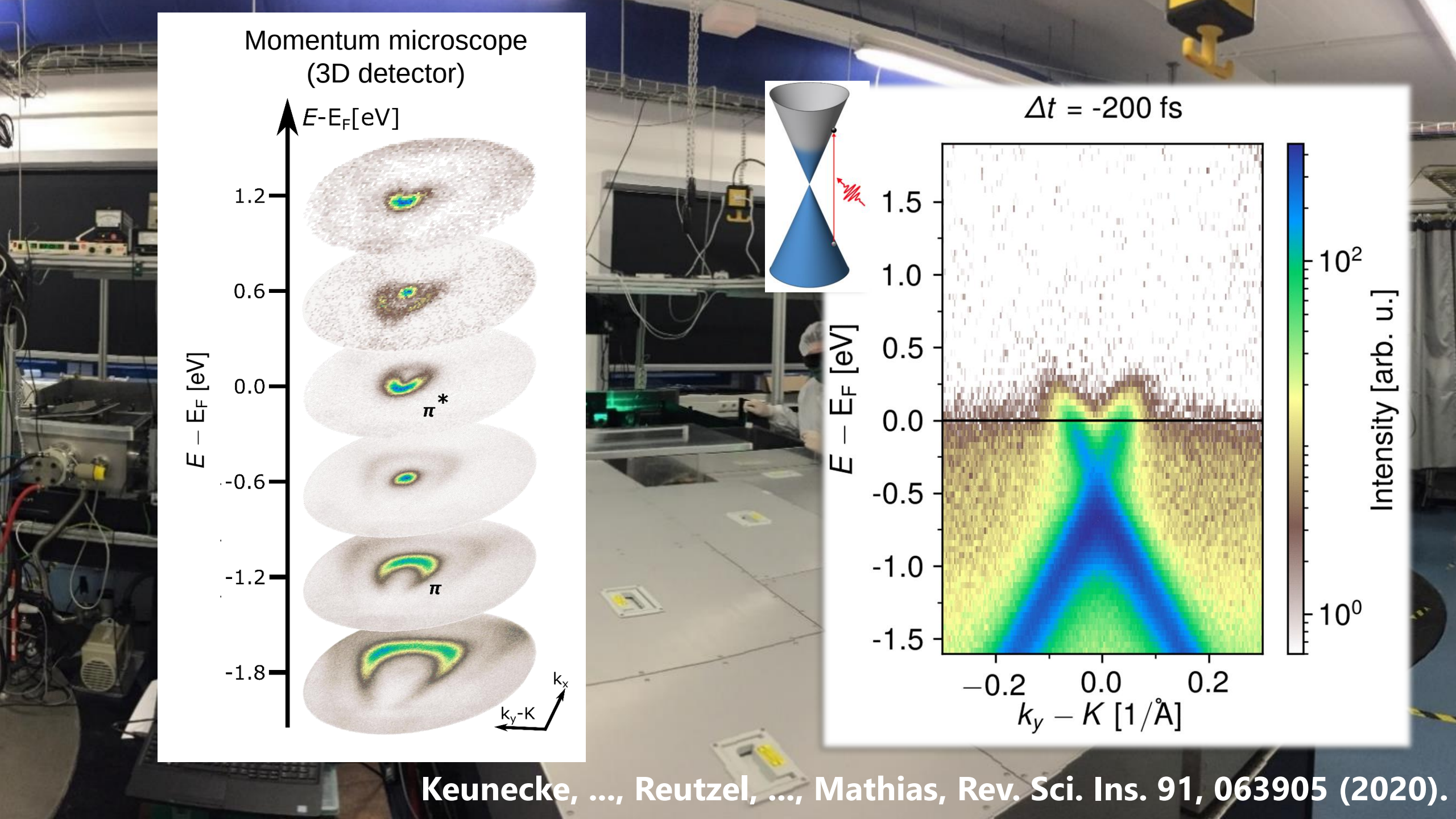

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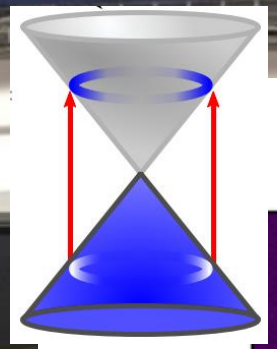

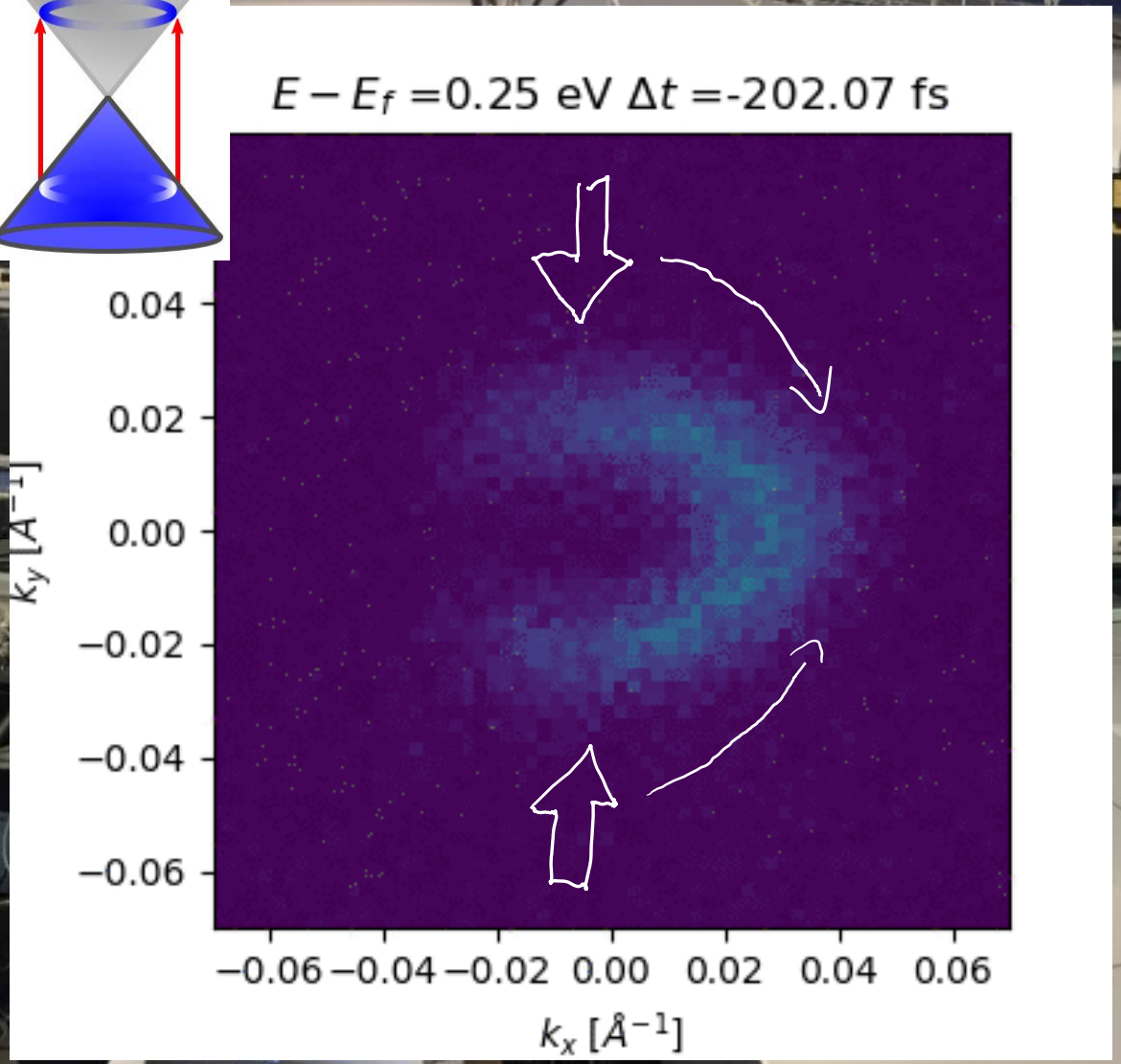

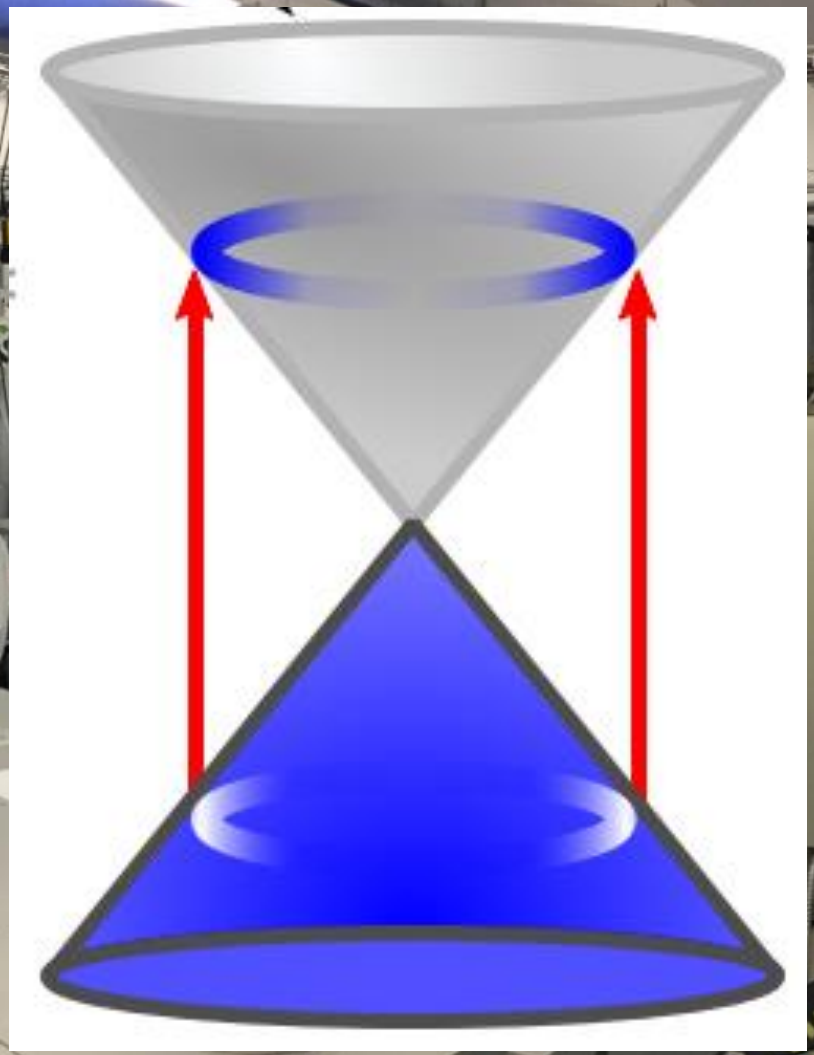

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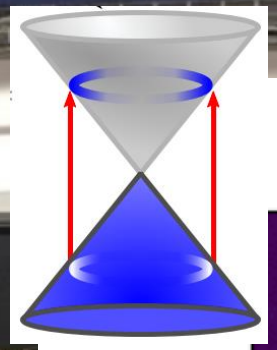

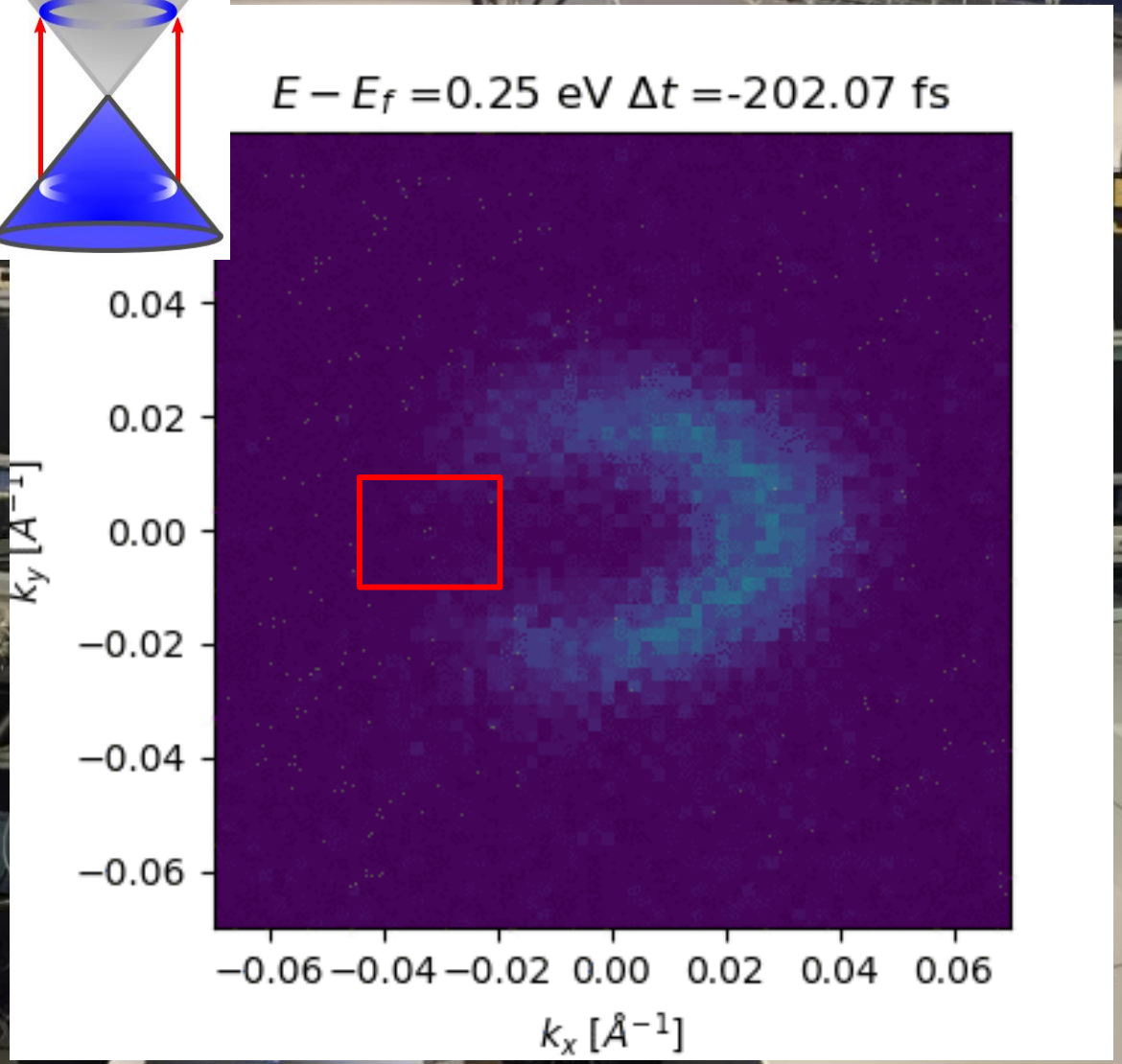

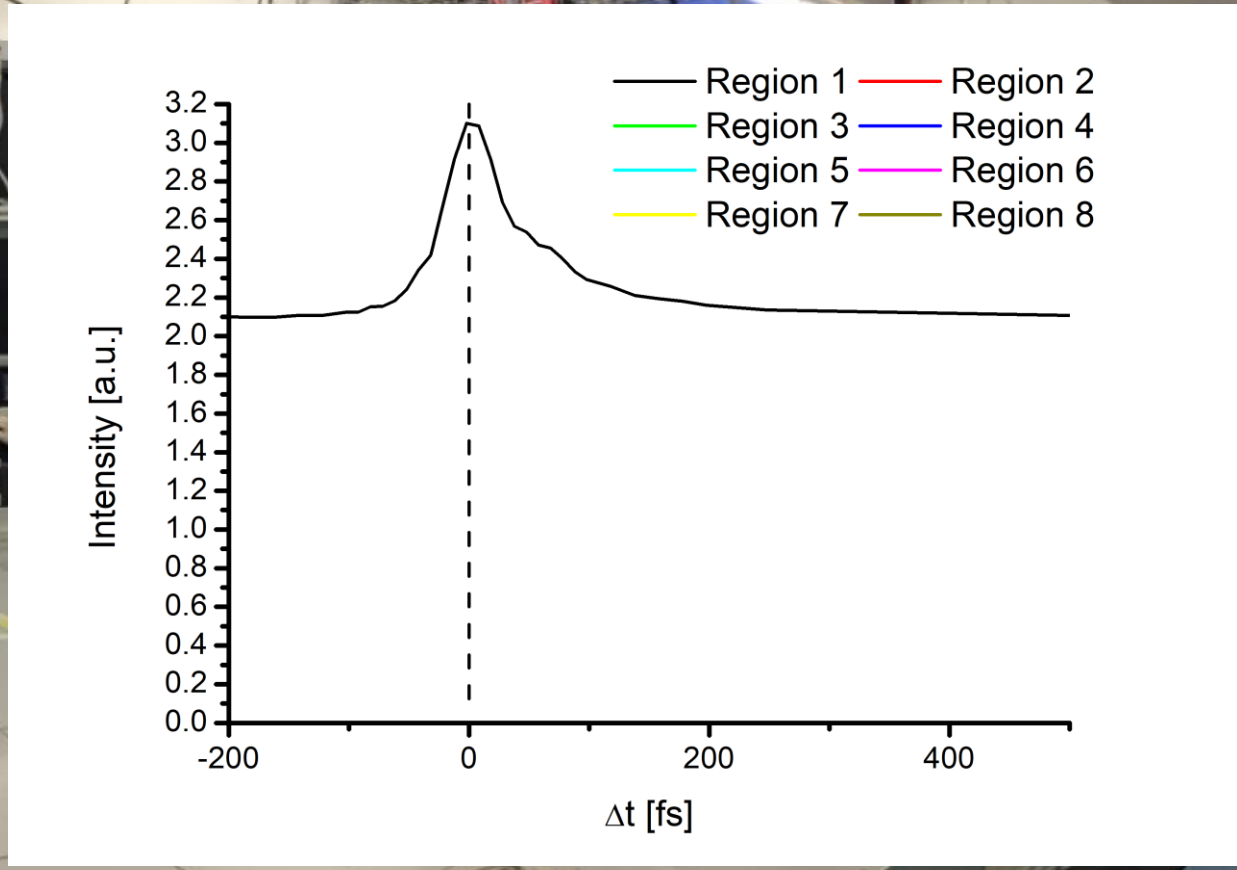

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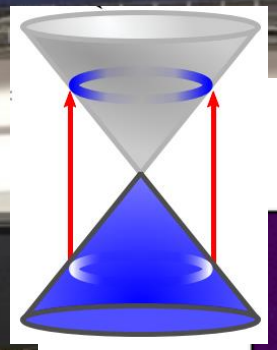

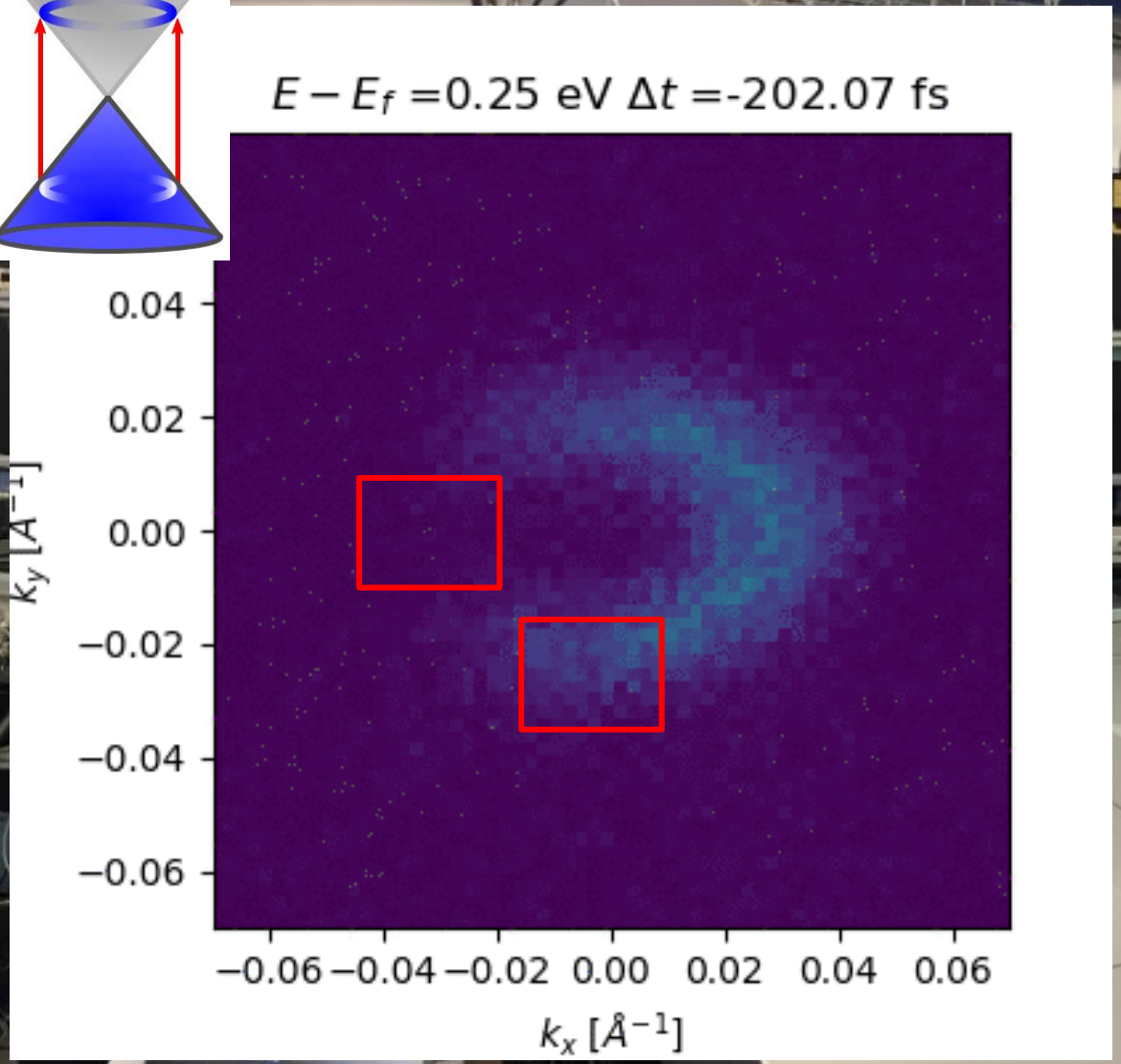

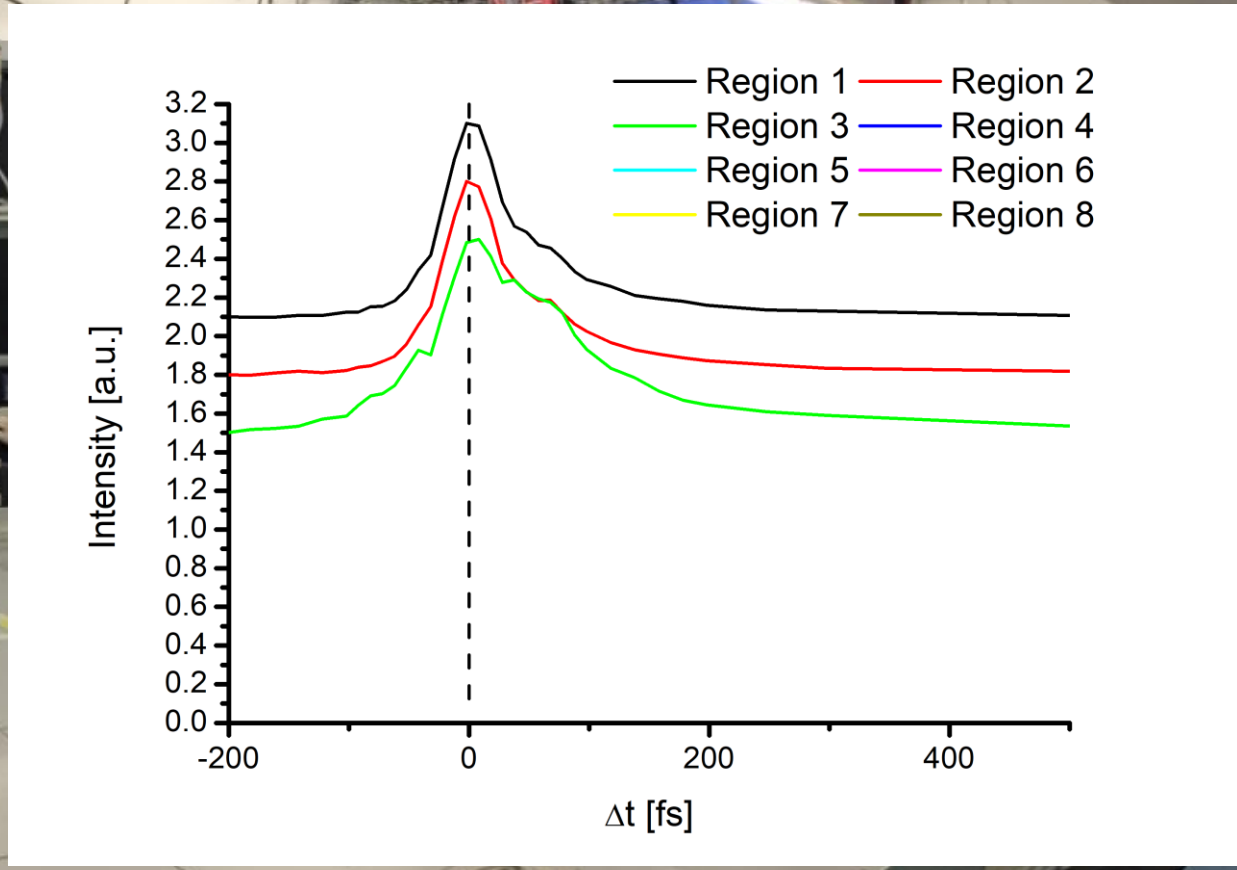

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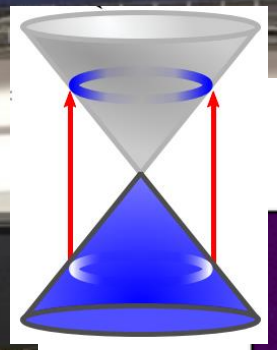
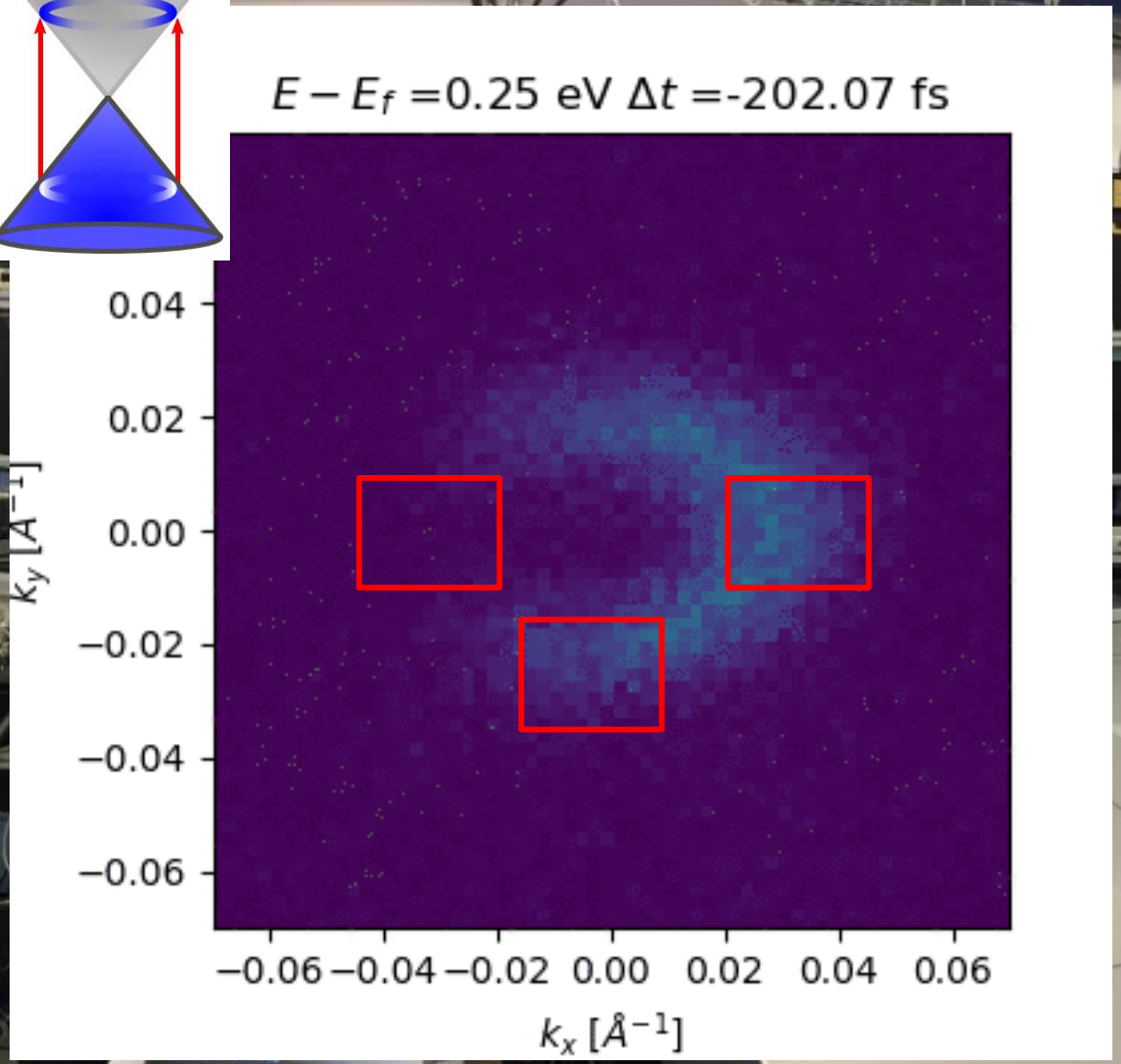
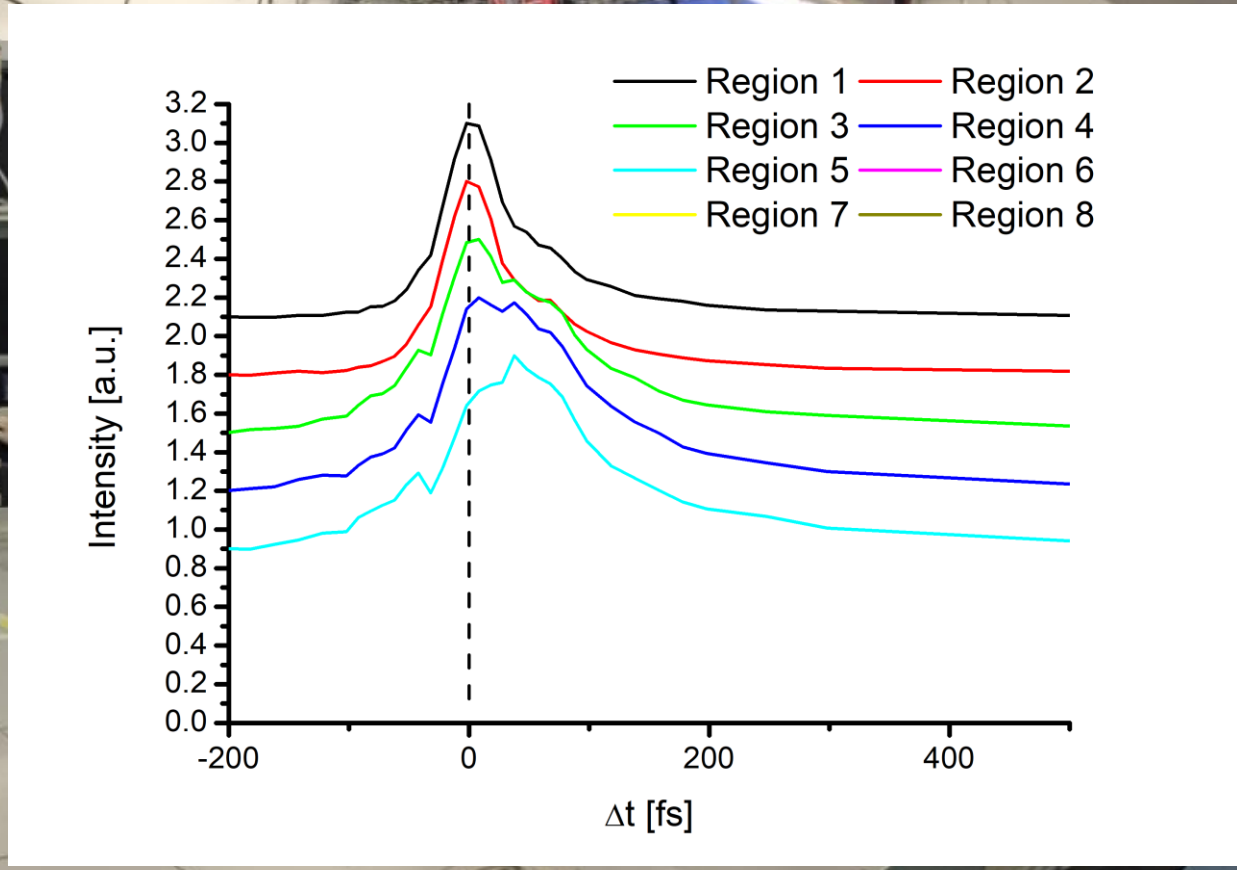
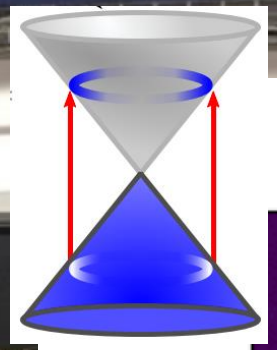
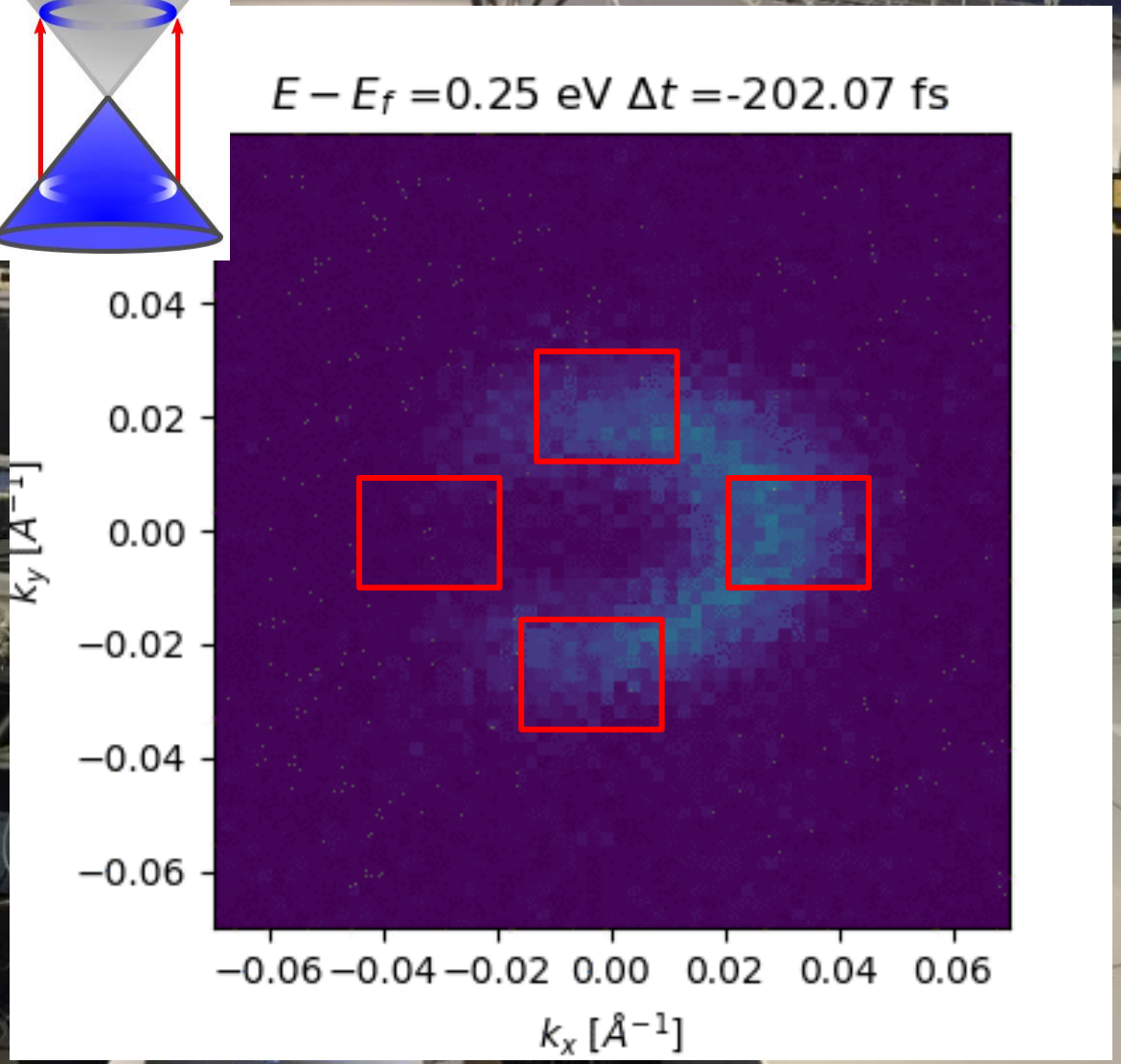


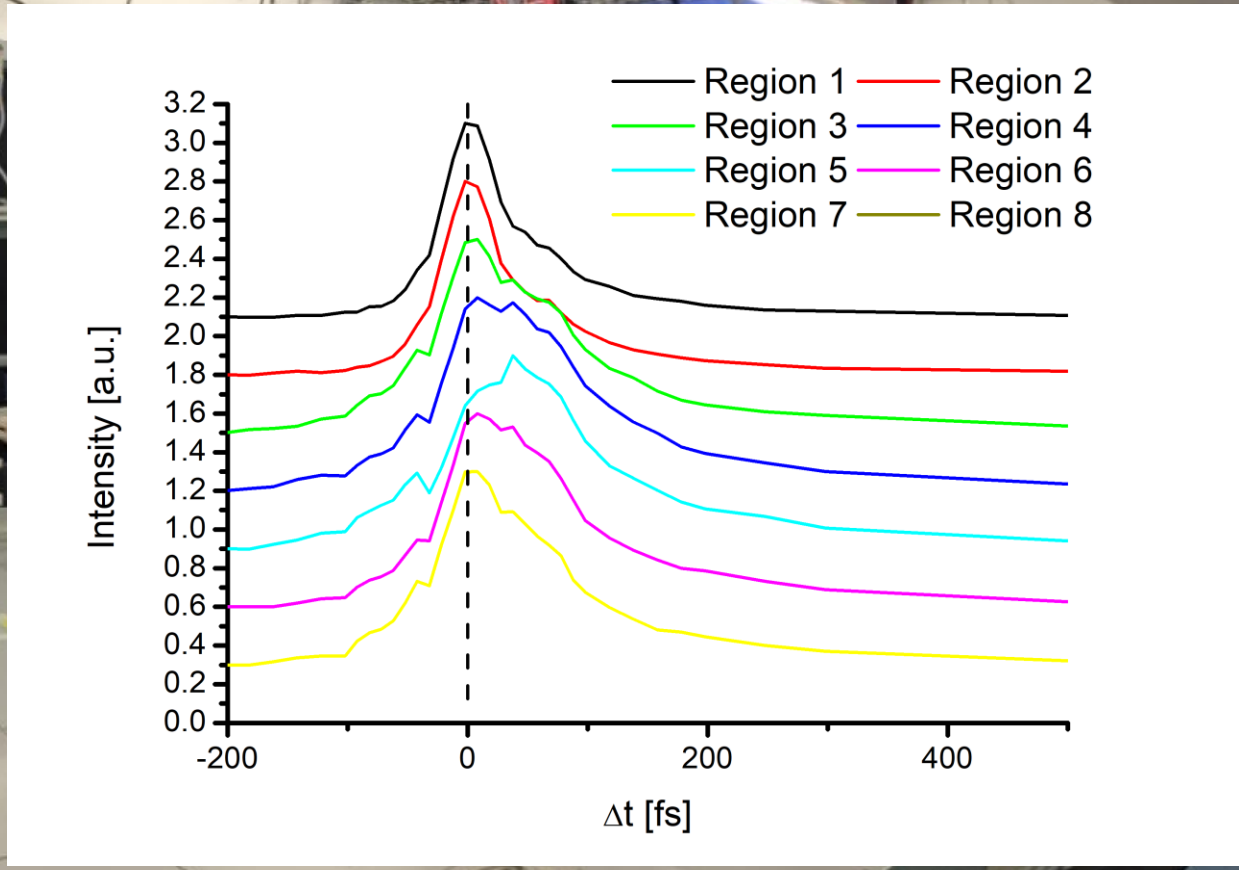




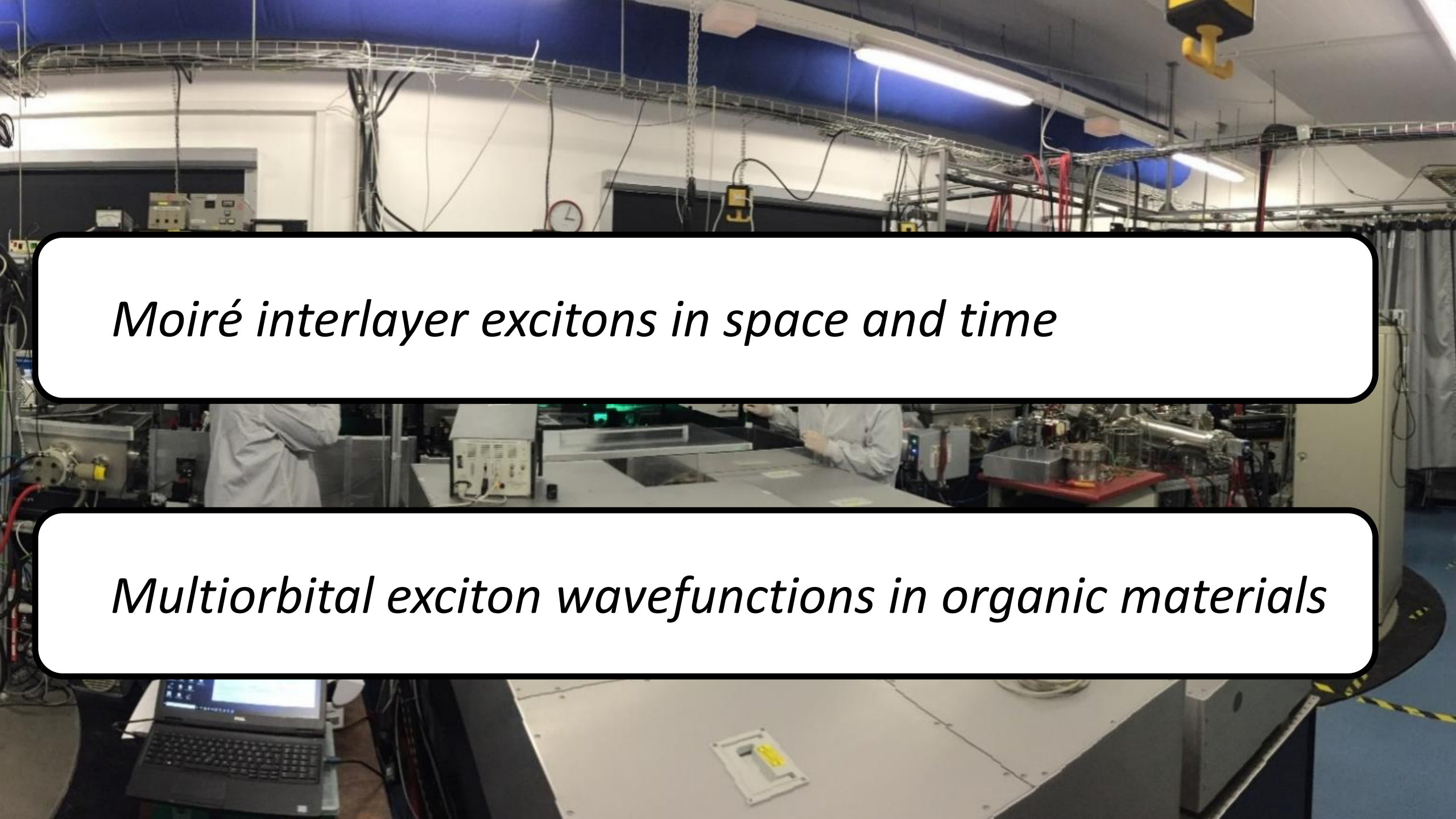
Keunecke, ..., Reutzel, ..., Mathias, Rev. Sci. Ins. 91, 063905 (2020).



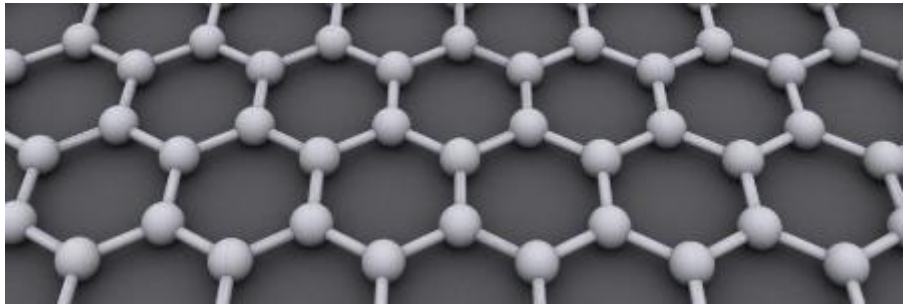




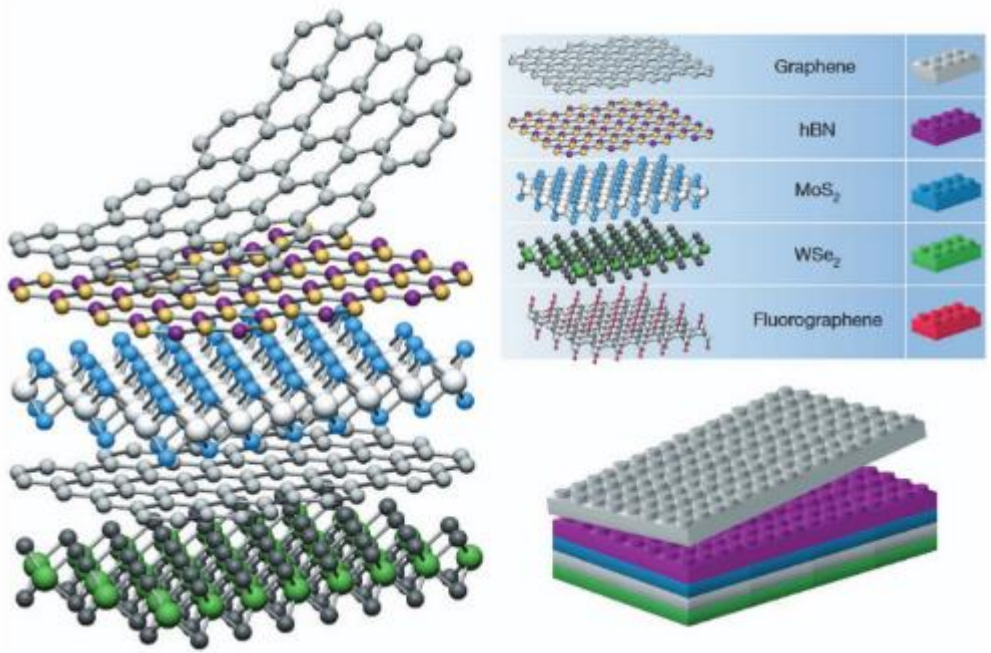
Keunecke, ..., Reutzel, ..., Mathias, Rev. Sci. Ins. 91, 063905 (2020).


Moiré interlayer excitons in space and time
Multiorbital exciton wavefunctions in organic materials

Emergent properties in moiré materials

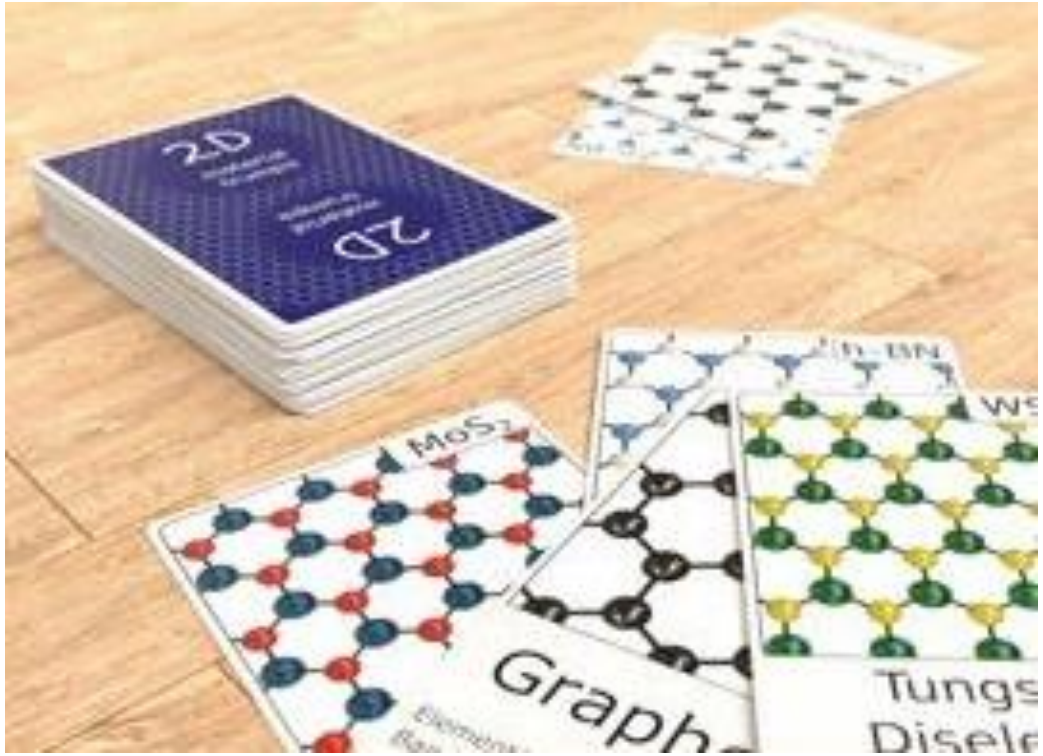


Emergent properties in moiré materials



Nature **499**, 419 (2013)

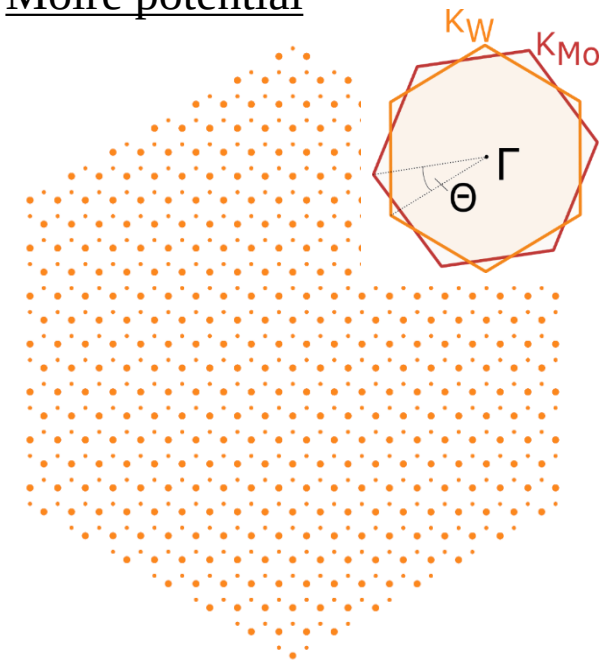
Emergent properties in moiré materials



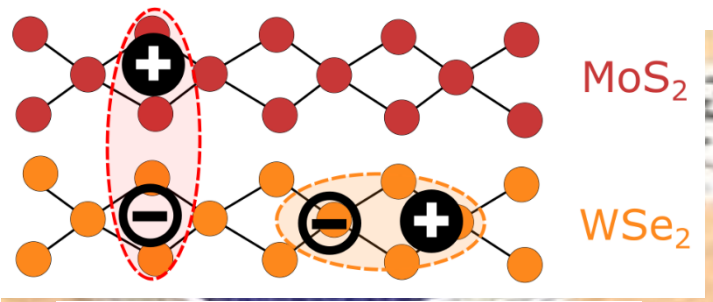
Physik Journal **18**, 29 (2019).

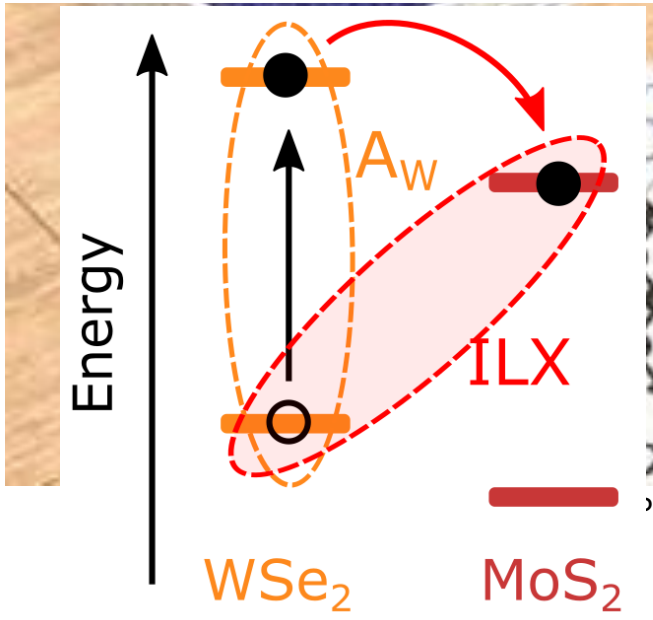
Emergent properties in moiré materials

Moiré potential



moiré interlayer excitons



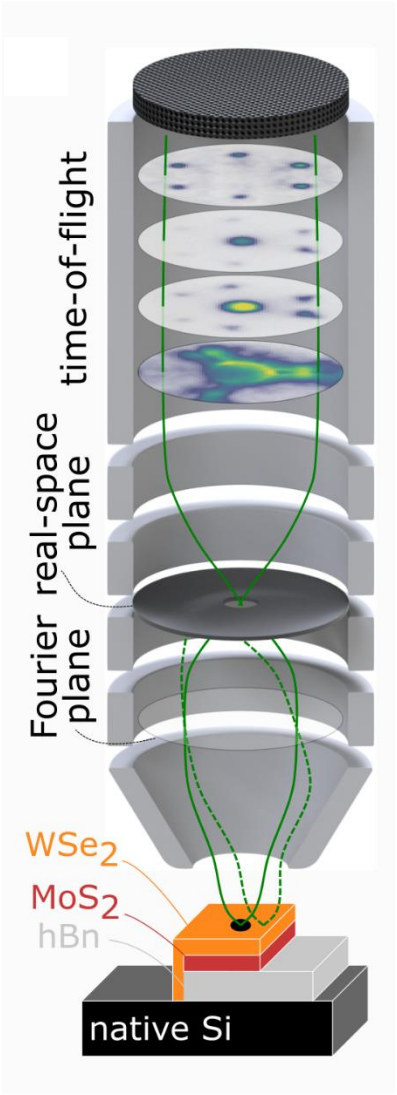


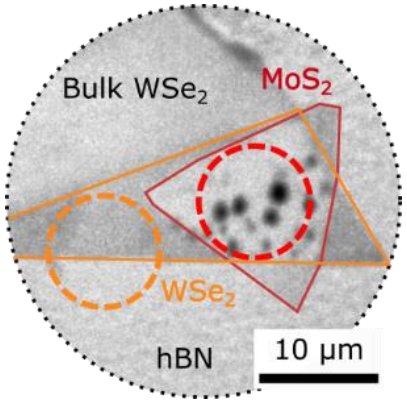
Two major research questions:
1) **How are ILX formed?**
2) **Impact of the moiré potential?**

What do we need?
- **femtosecond time-resolution**
- **100 meV energy resolution**
- ➢ **momentum resolution**

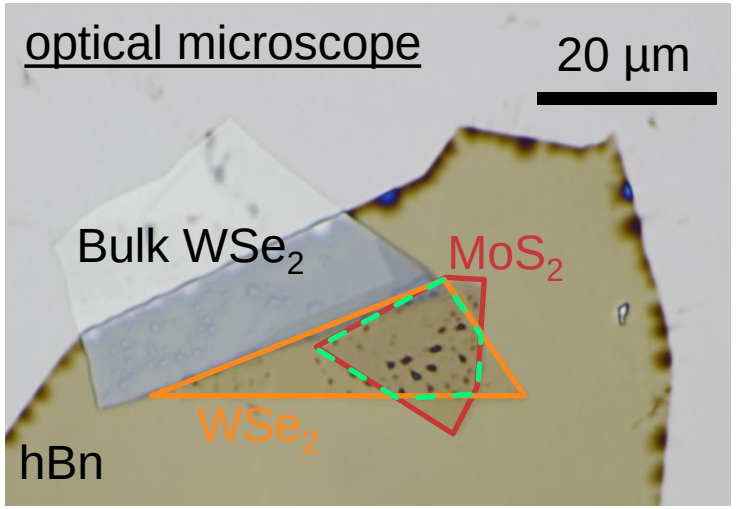
hysik Journal **18**, 29 (2019).

10° twisted WSe_2/MoS_2: sample characterization

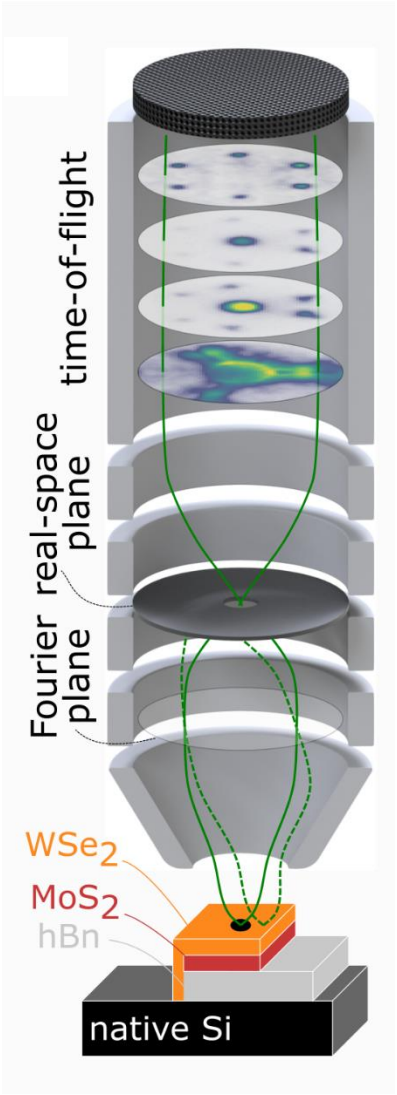




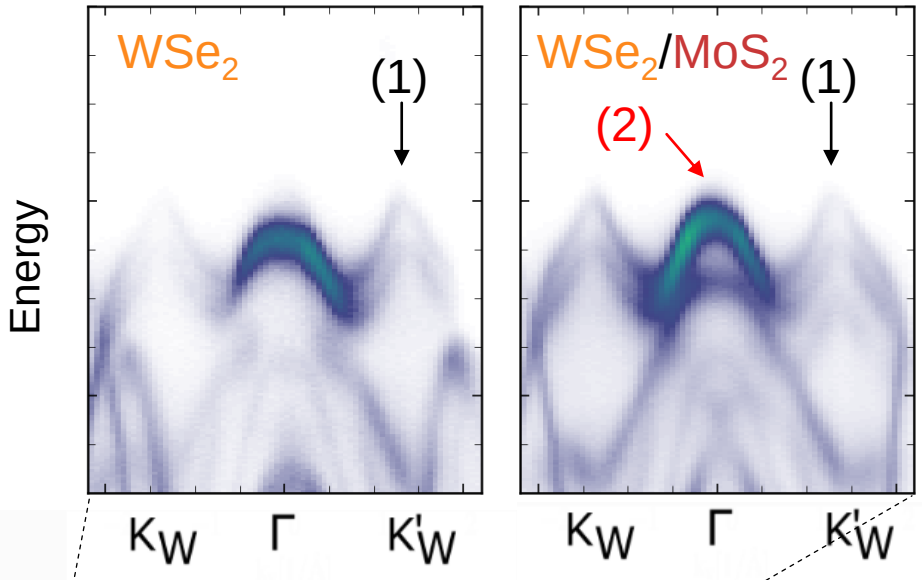
sample fabrication: AbdulAziz AlMutairi and Stephan Hofmann, University of Cambridge

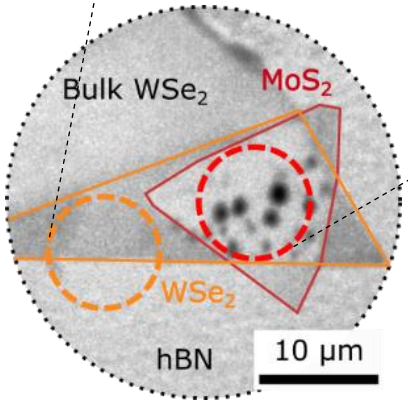


10° twisted WSe_2/MoS_2: static band mapping



static band mapping

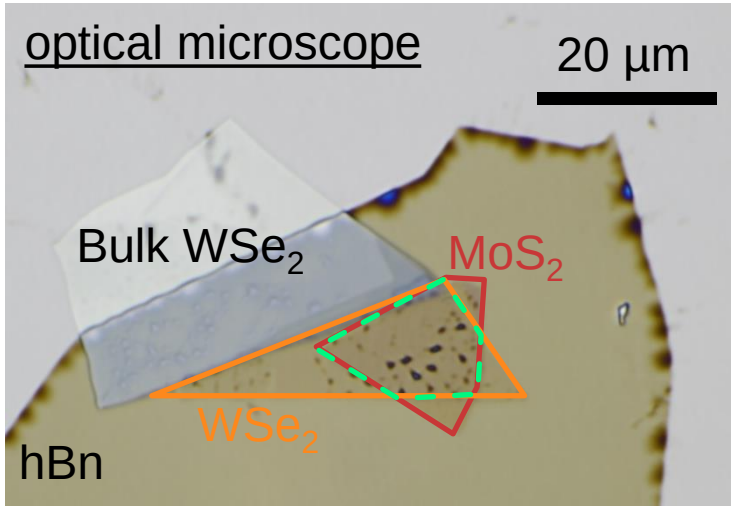




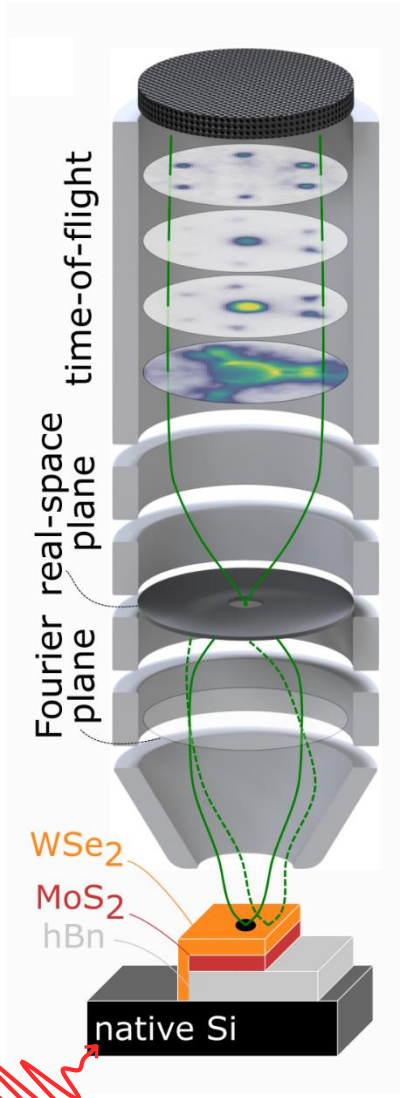
(1) spin-split WSe_2 valence band
(2) hybridized bands at Γ valley

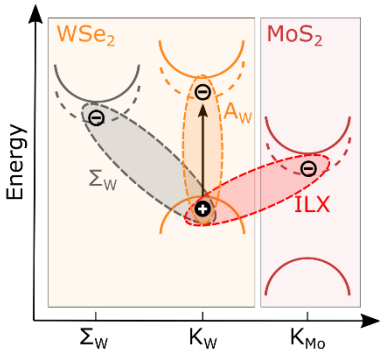
Wilson *et al.*, Sci. Adv. **3**, e1601832 (2017).

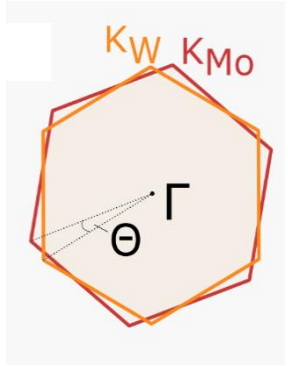
optical microscope

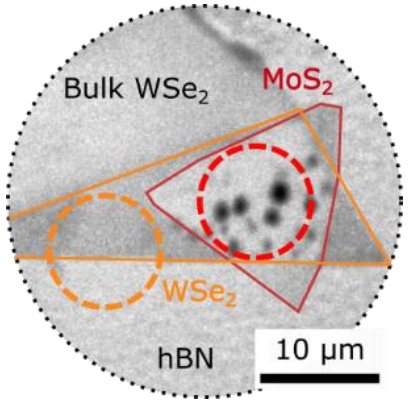


Open tasks: exciton dynamics and moiré hallmarks

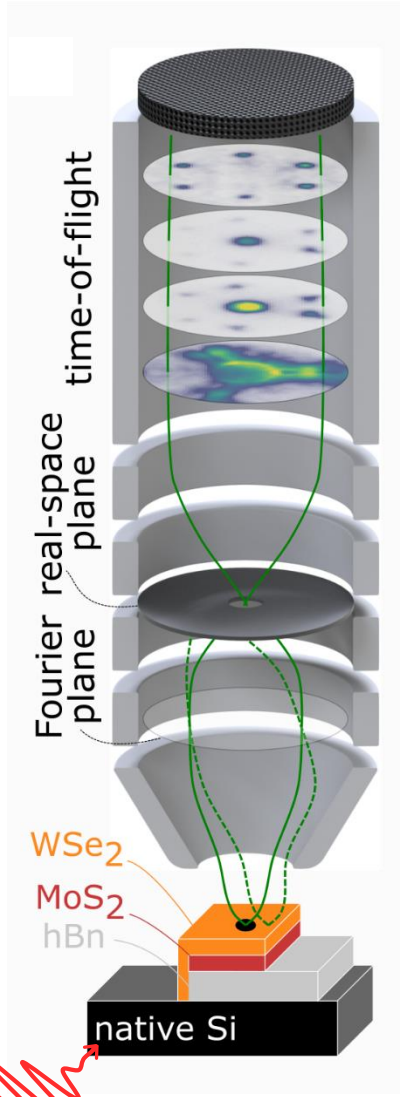


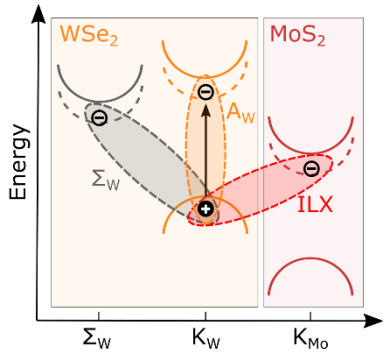


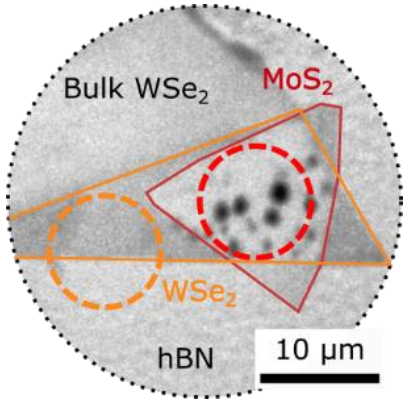




Interlayer excitons probed in trARPES

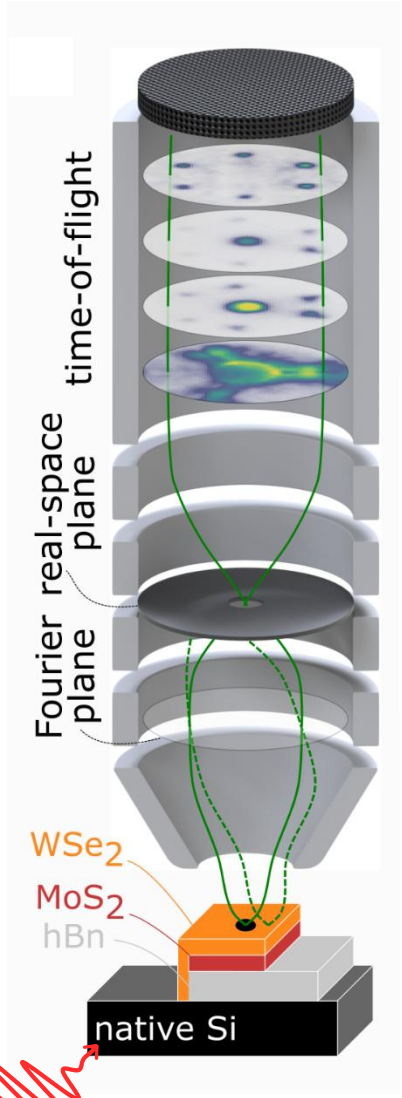


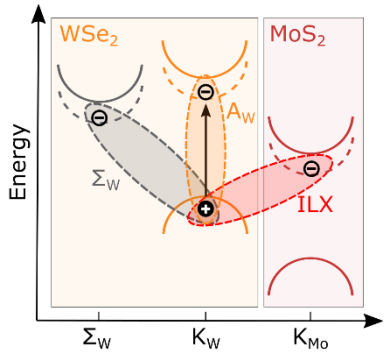


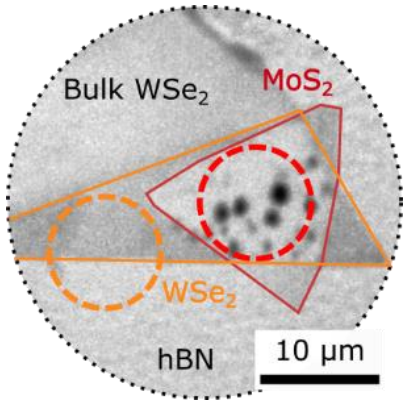


identification of ILX in momentum integrated energy-distribution-curves

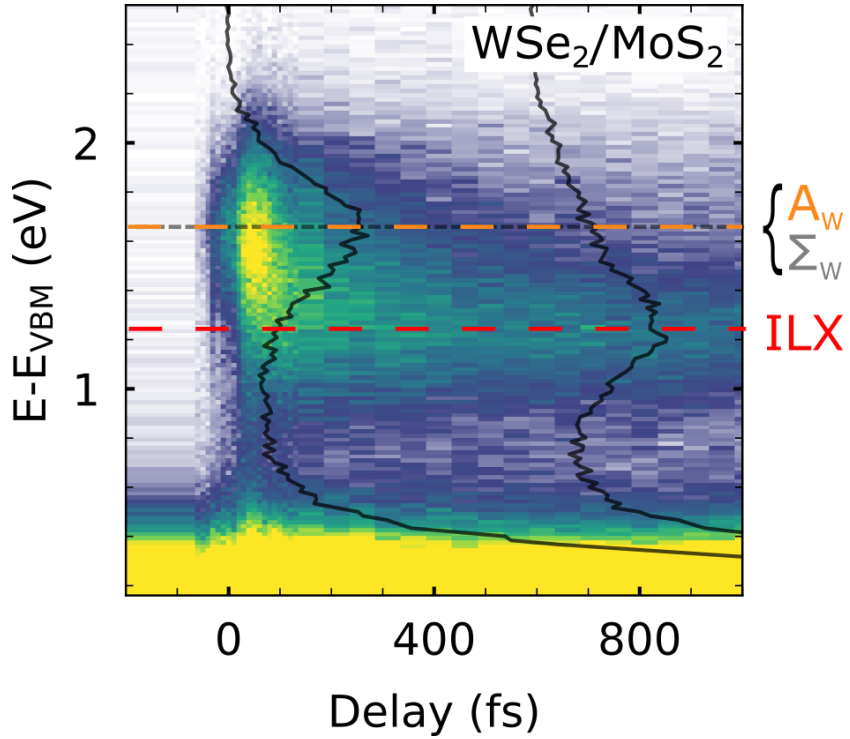
Interlayer excitons probed in trARPES





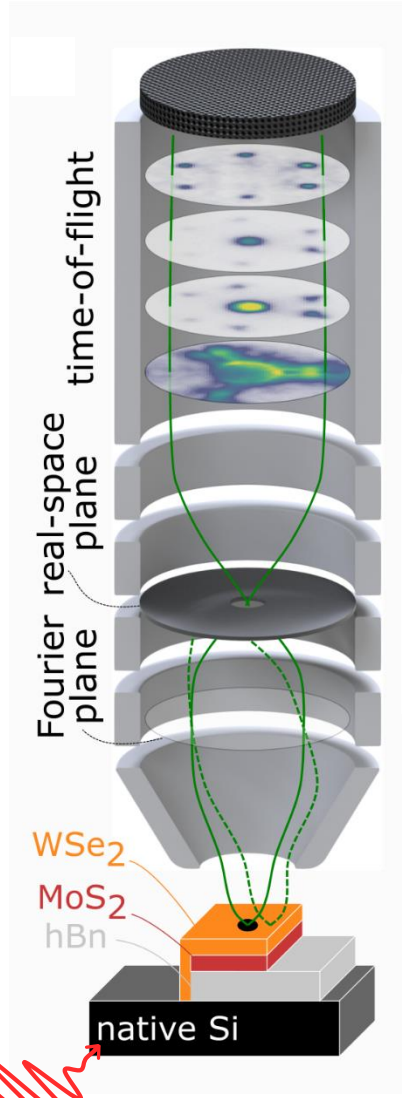


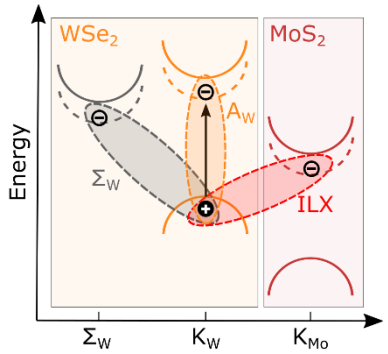
identification of ILX in momentum integrated energy-distribution-curves

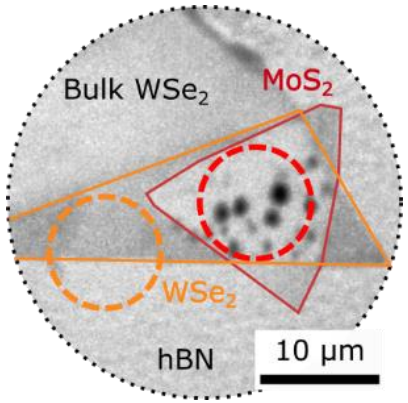


- ILX formation on sub-100 fs timescale

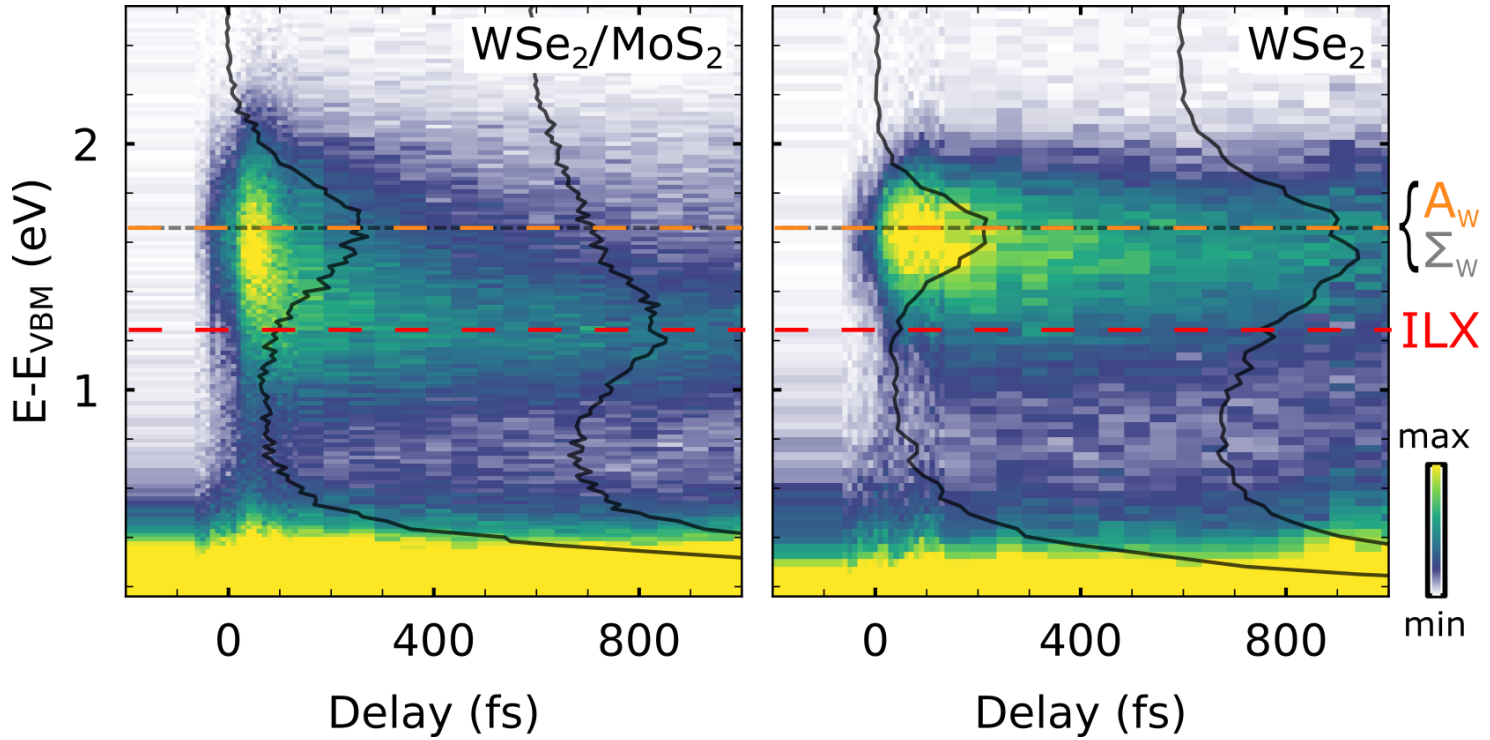
Interlayer excitons probed in trARPES





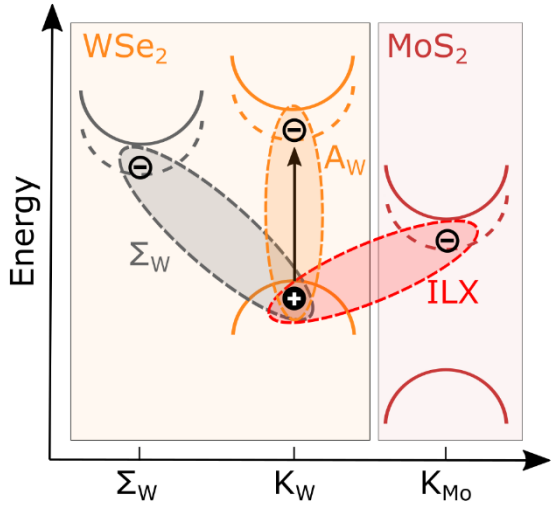


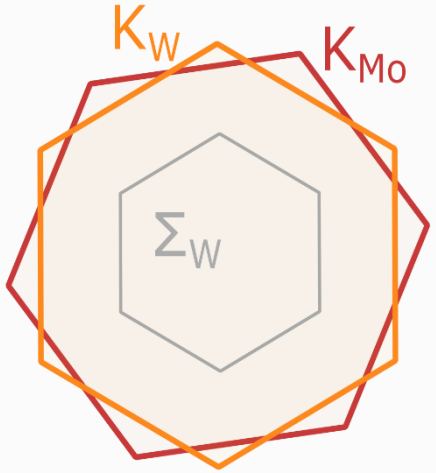
identification of ILX in momentum integrated energy-distribution-curves

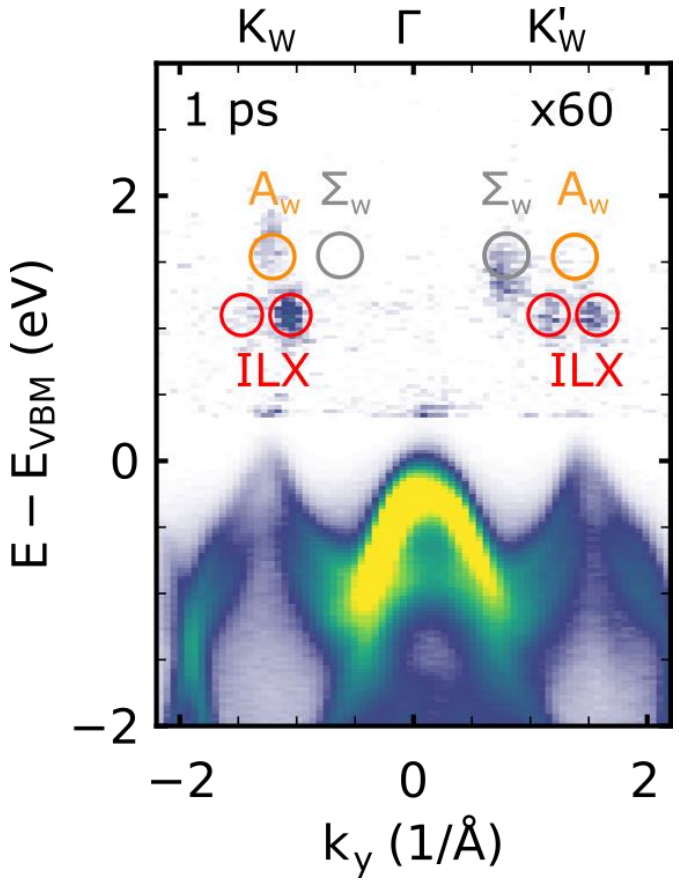


- ILX formation on sub-100 fs timescale
- no ILX in WSe_2 monolayer!

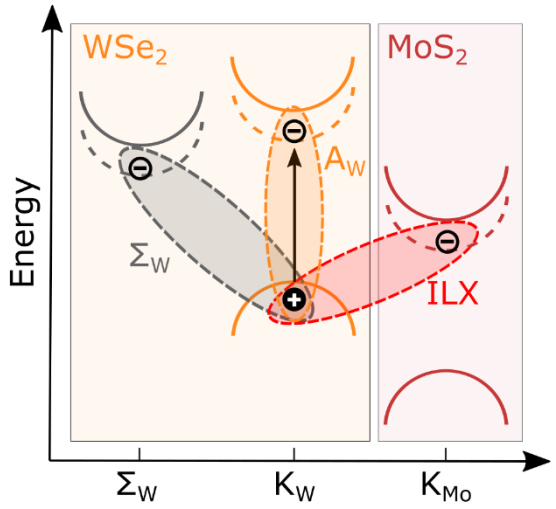
Momentum-resolved identification of excitonic fingerprints

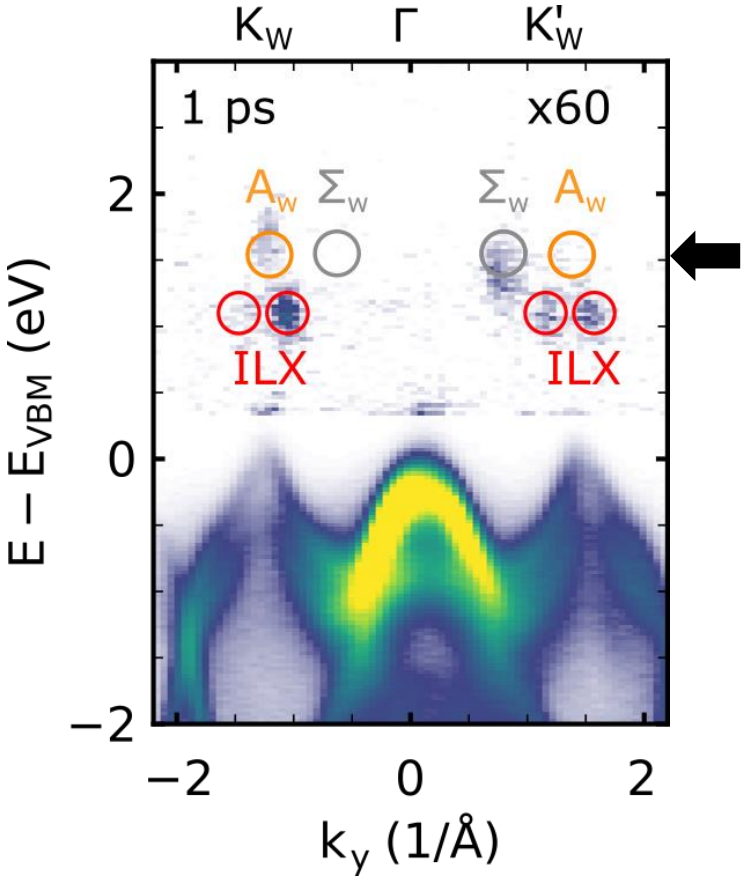




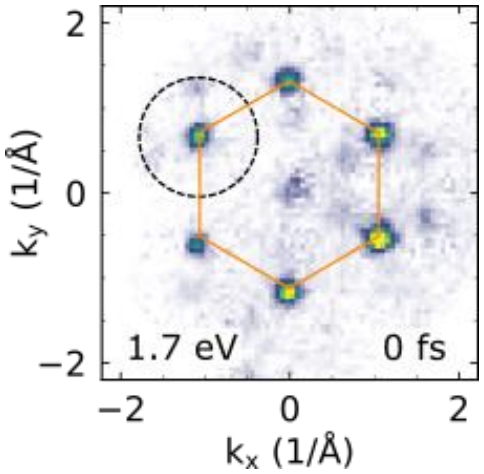


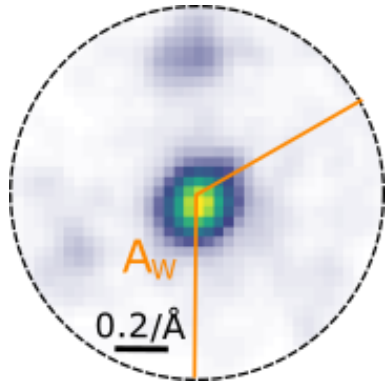
Excitonic momentum fingerprints





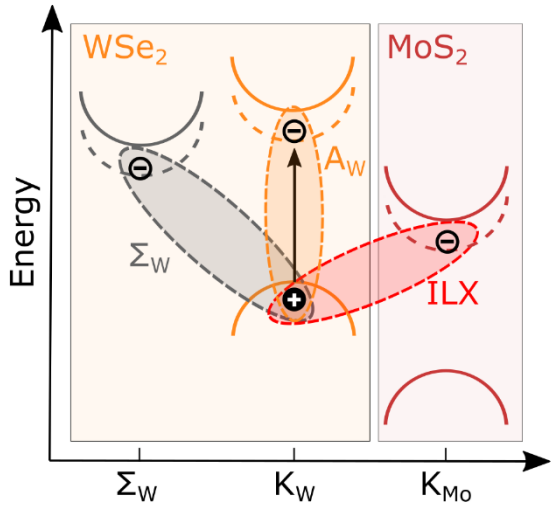
bright intralayer A_W-exciton

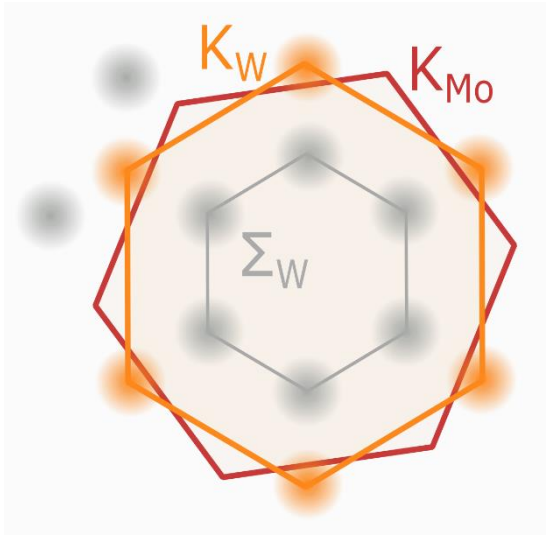


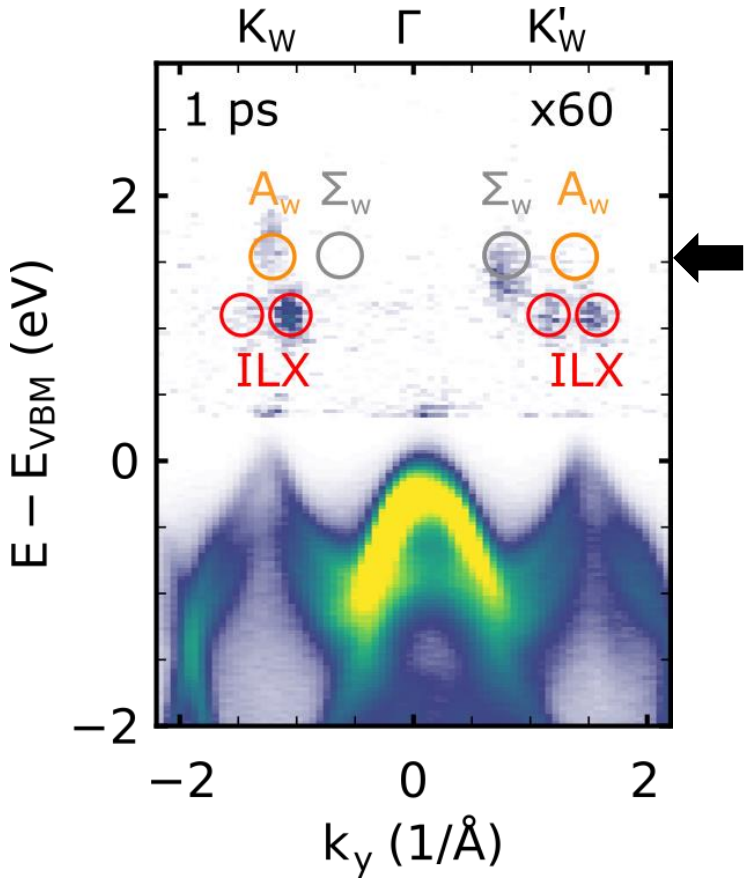


Madéo et al., Science 370, 1199 (2020); Wallauer *et al.*, Nano Lett. 21, 5867 (2021); Dong et al., Natural Sciences 1, e10010 (2021).

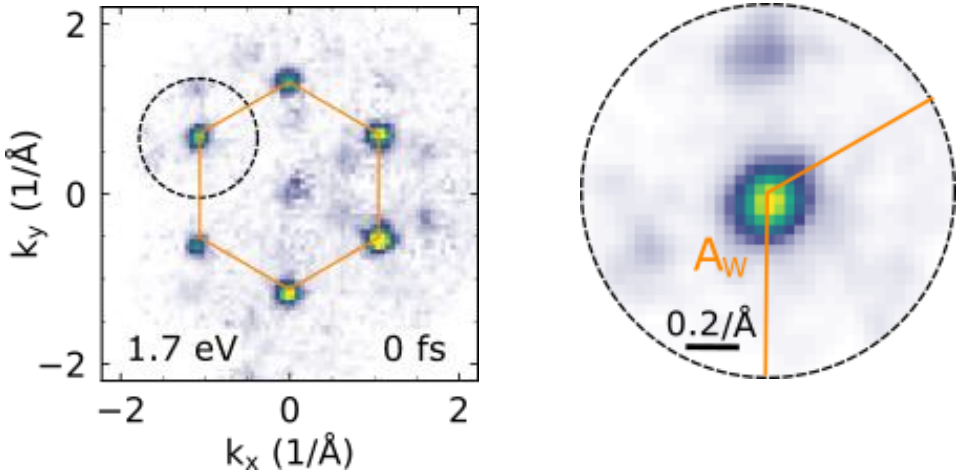
Excitonic momentum fingerprints



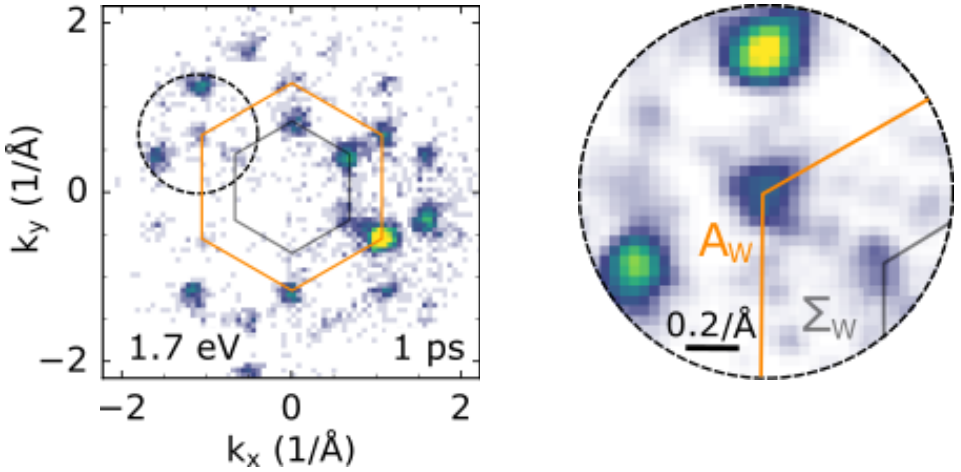




bright intralayer A_W-exciton



dark intralayer Σ_W-exciton



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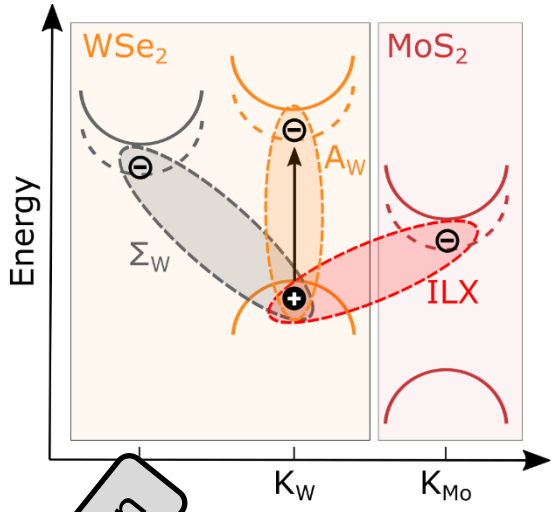
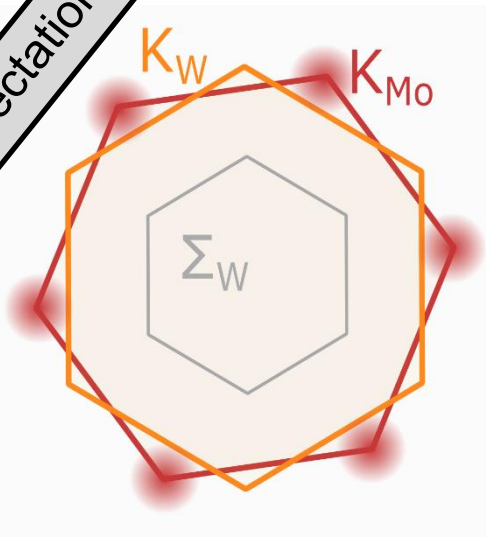
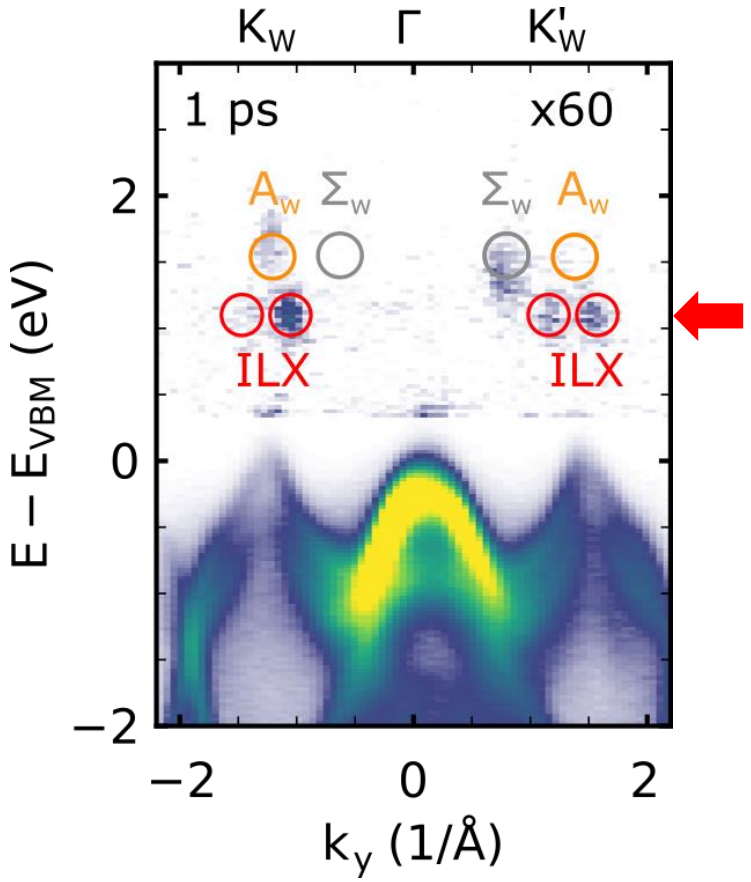
Excitonic momentum fingerprints



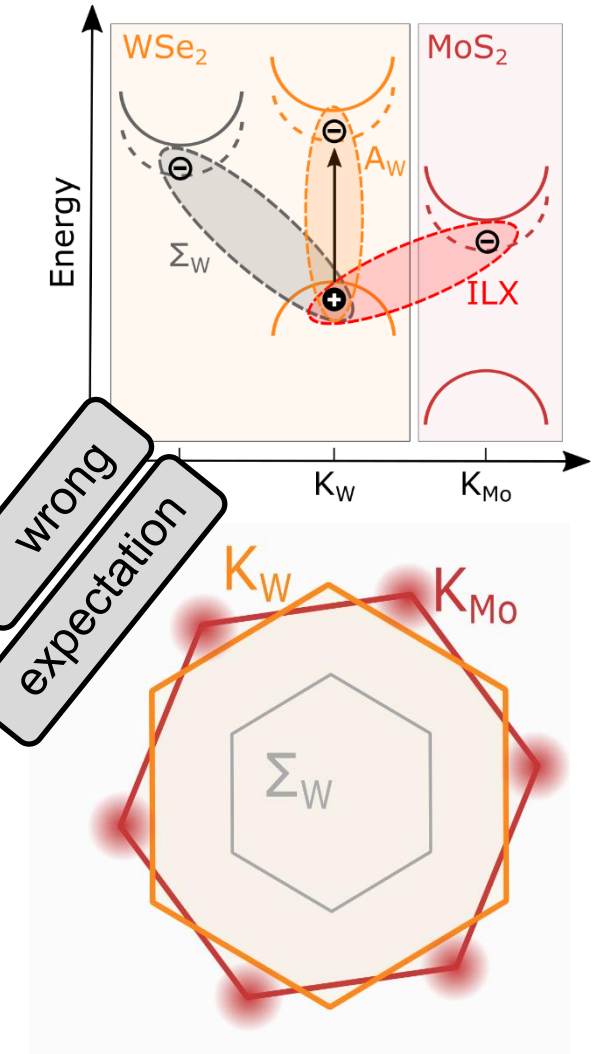
expectation

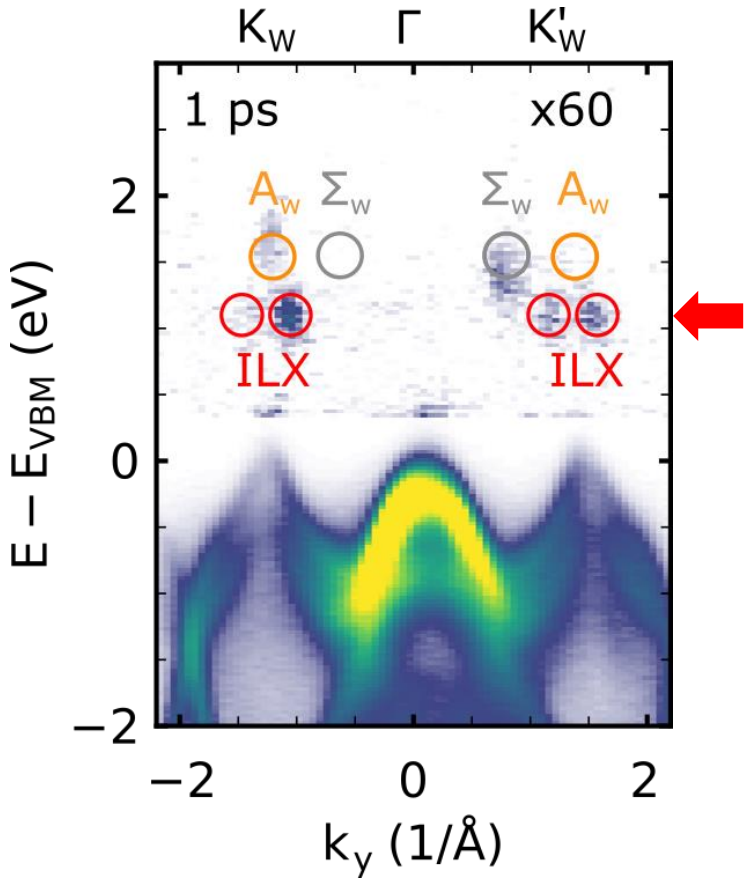




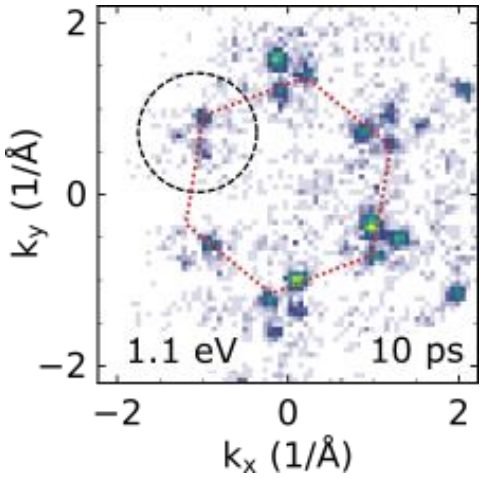
interlayer exciton

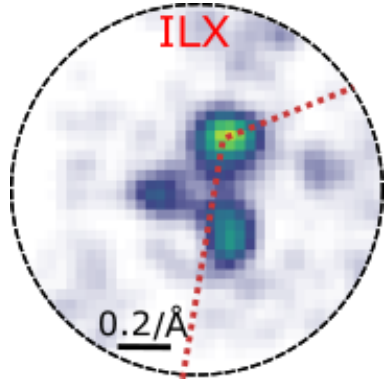
Excitonic momentum fingerprints





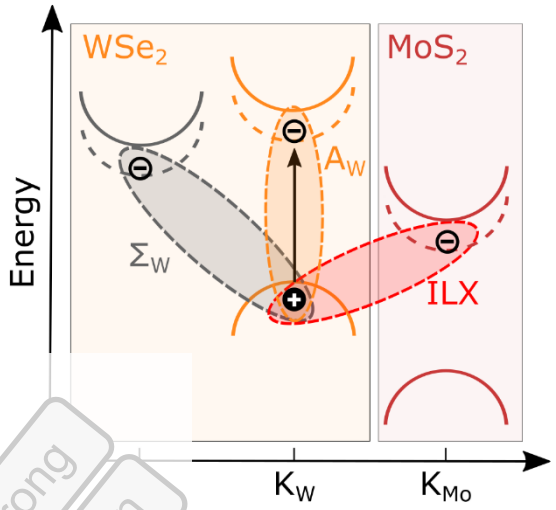
interlayer exciton



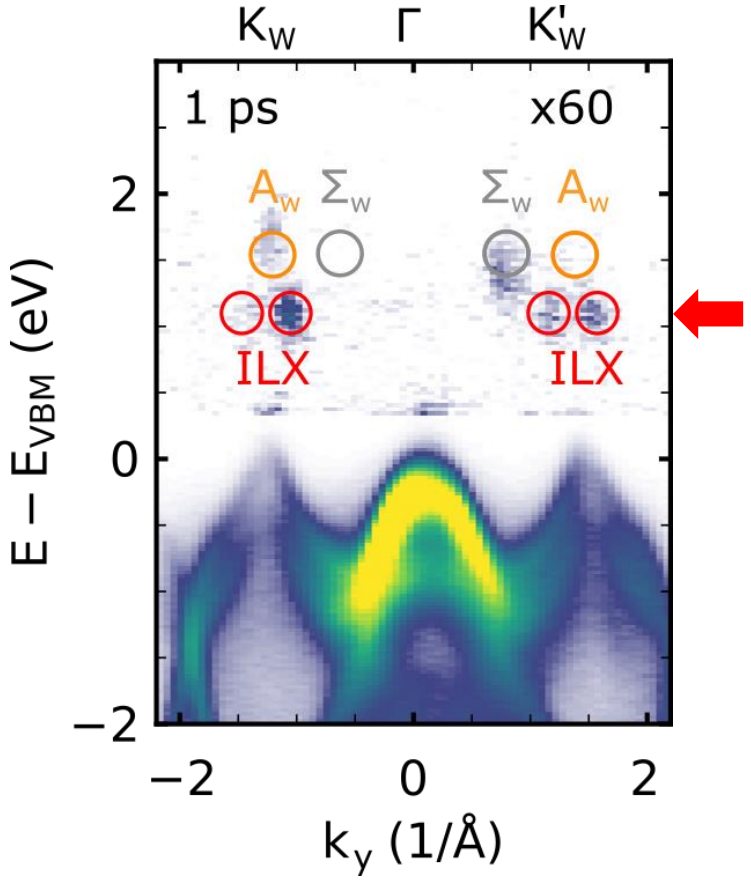


- unexpected threefold signature

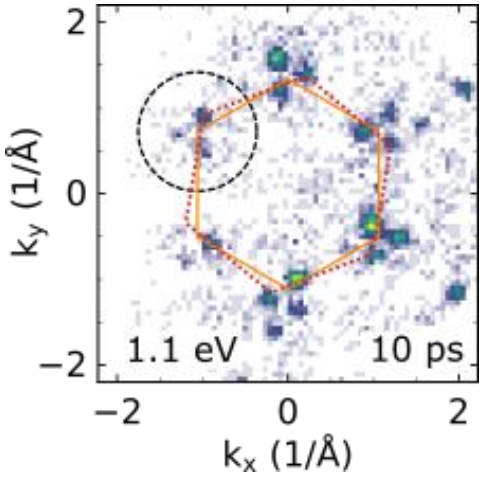
Excitonic momentum fingerprints

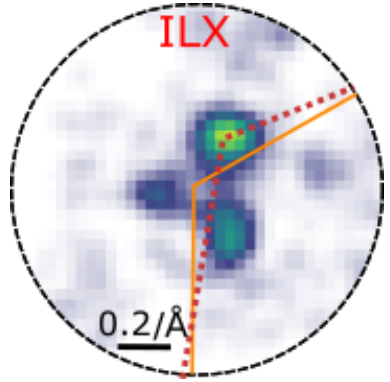


wrong expectation



interlayer exciton





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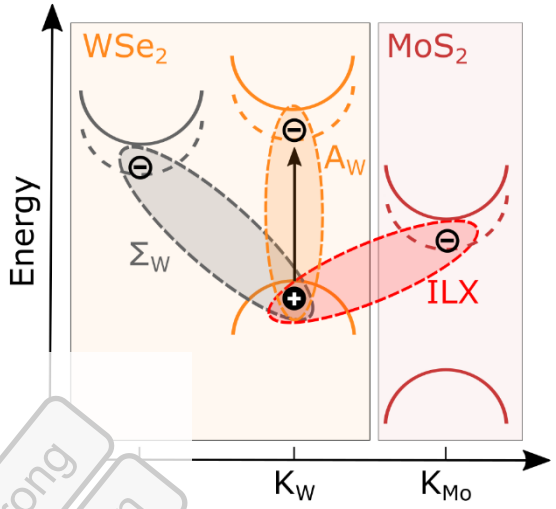

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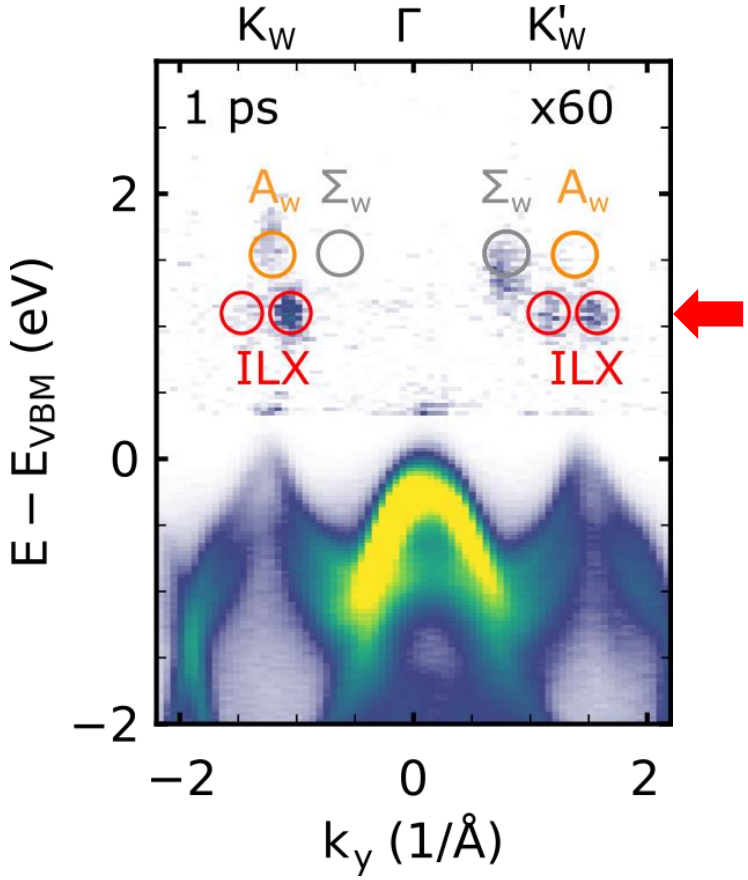
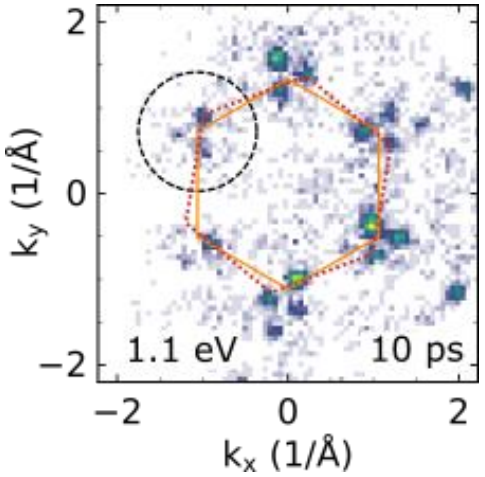
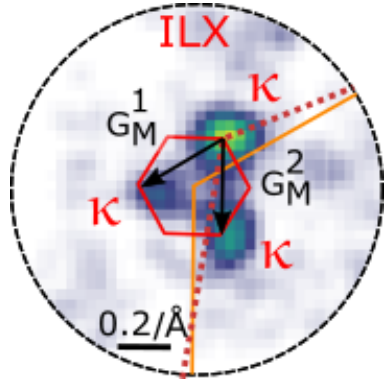


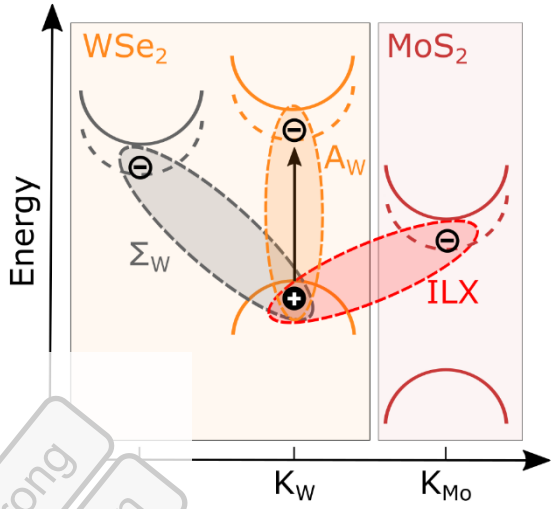
moiré interlayer exciton





- unexpected threefold signature
- **moiré superlattice hallmark!**

Excitonic momentum fingerprints



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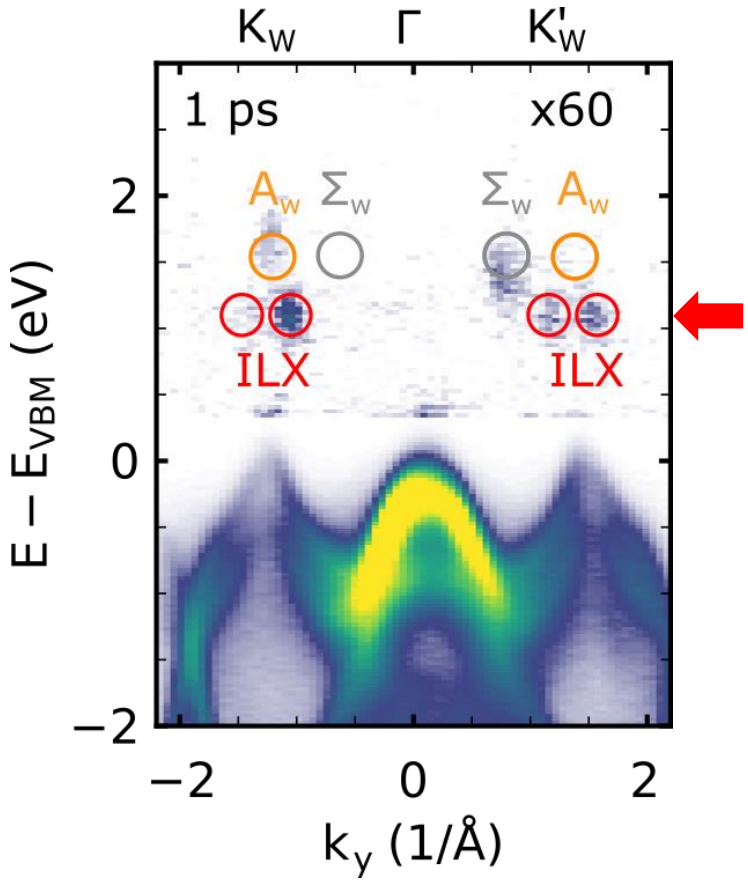
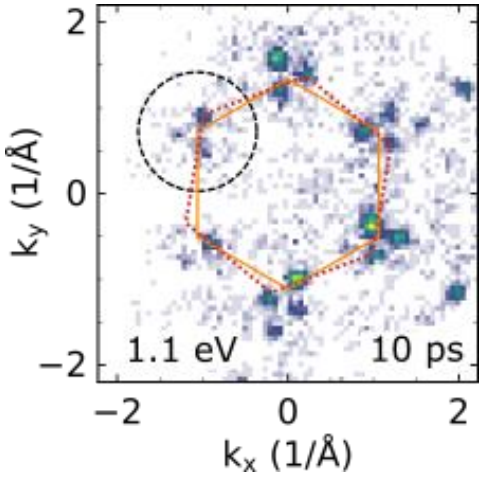
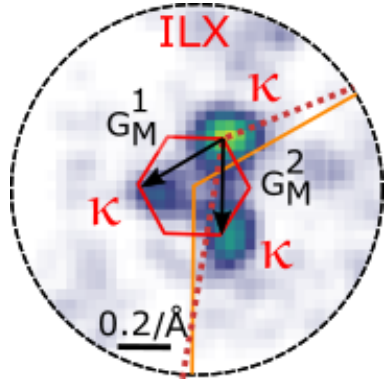


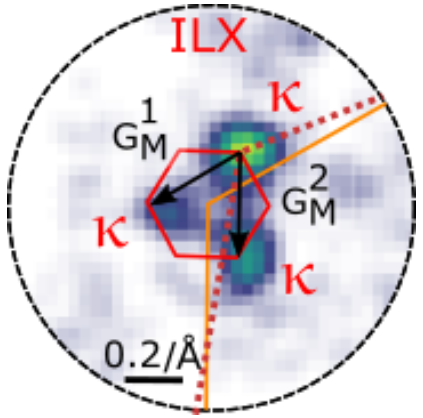
moiré interlayer exciton

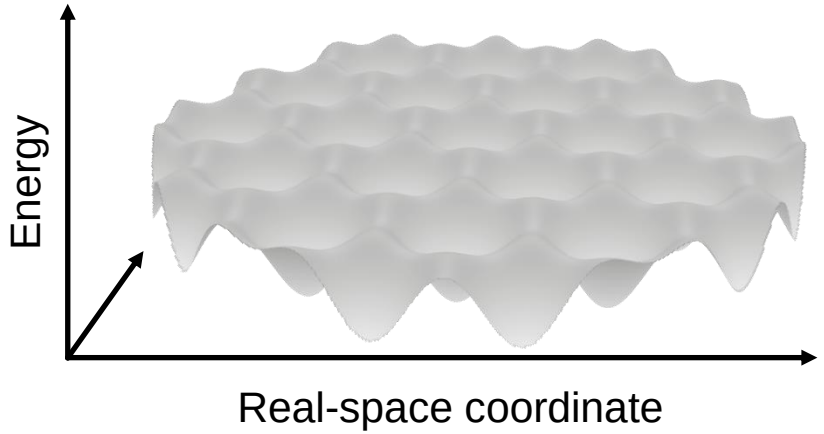




- unexpected threefold signature
- **moiré superlattice hallmark!**
- **only observed for ILX!**

ILX wavefunction in the moiré superlattice





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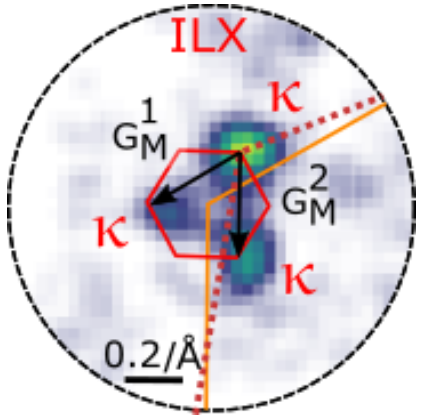

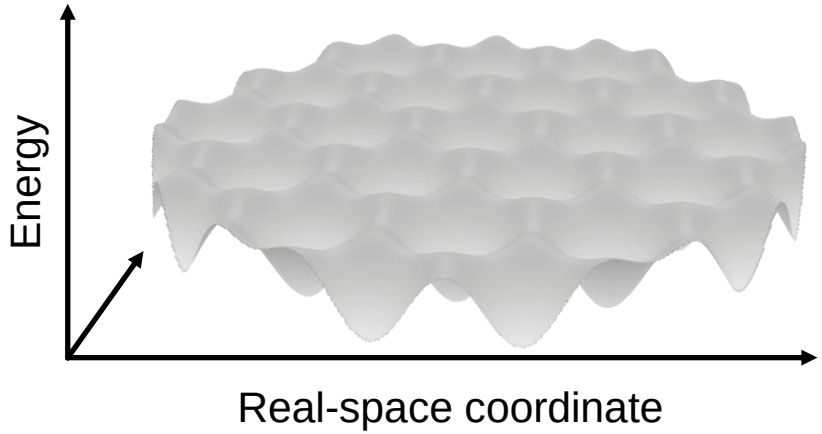

orbital tomography

$$I(k_x, k_y) \propto \left|\mathrm{FT}\{\Psi(r_x, r_y)\}\right|^2$$

Puschnig *et al.*, Science 326, 702 (2009).
Jansen *et al.*, NJP **22**, 063012 (2020).
Dong *et al.*, Natural Sciences **1**, e10010 (2021).
Man *et al.*, Science Advances **7**, eabg0192 (2021).

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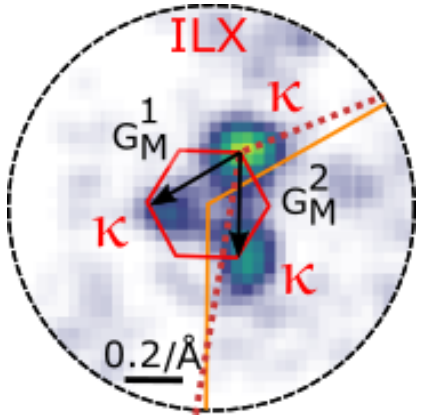

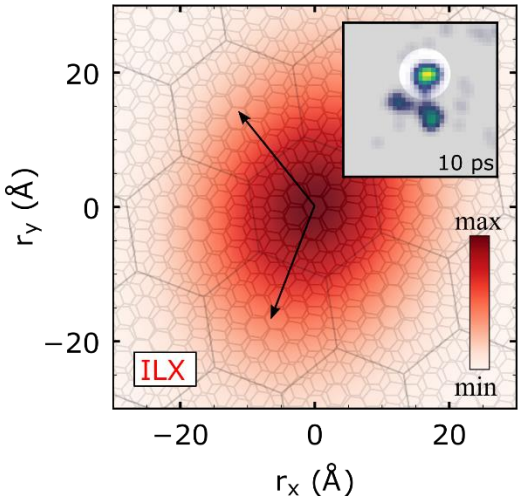

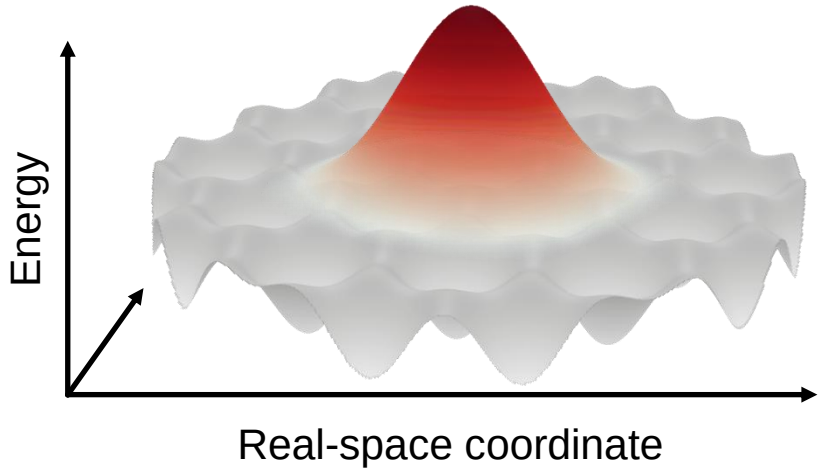

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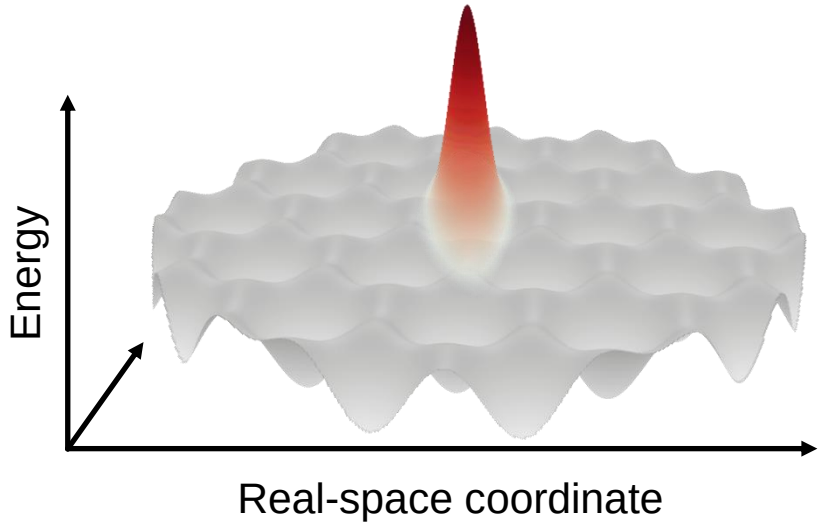
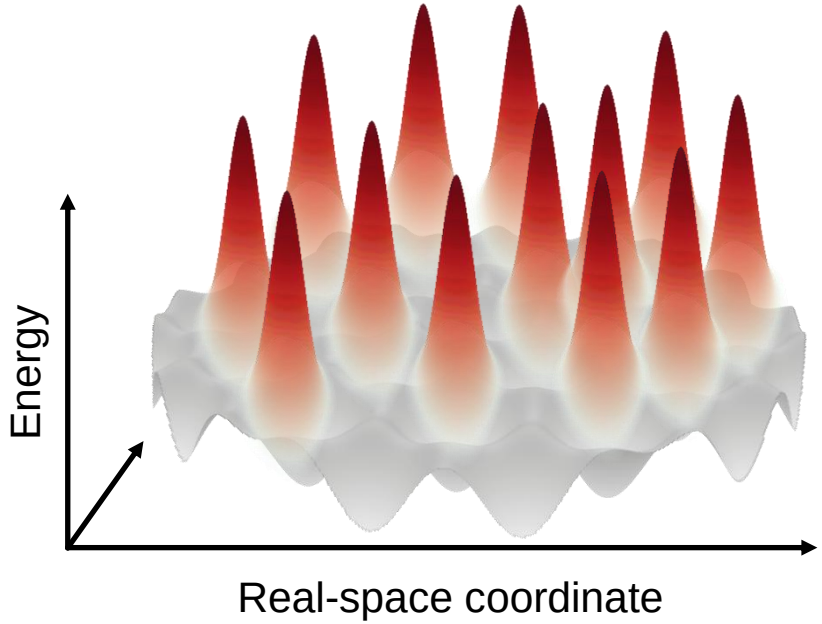
ILX wavefunction in the moiré superlattice

1) manipulate twist angle



ILX wavefunction in the moiré superlattice

1) manipulate twist angle
2) change exciton density



ILX wavefunction in the moiré superlattice

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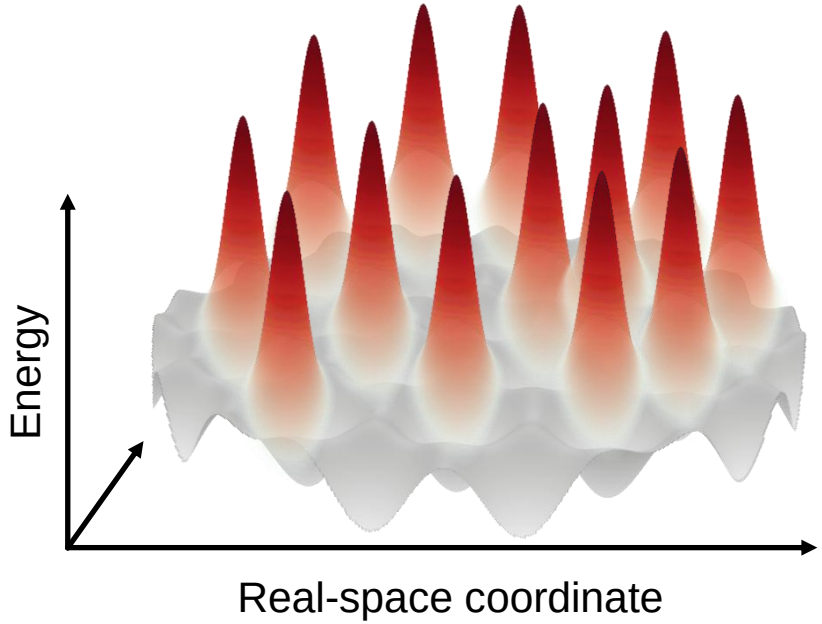
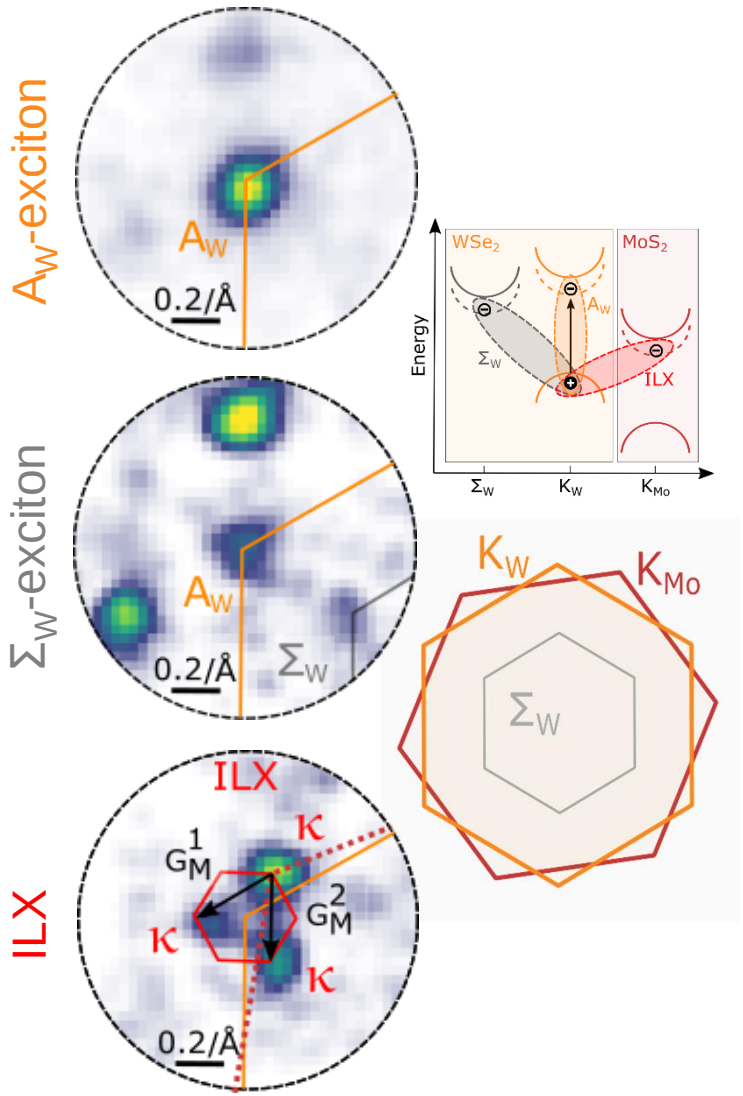


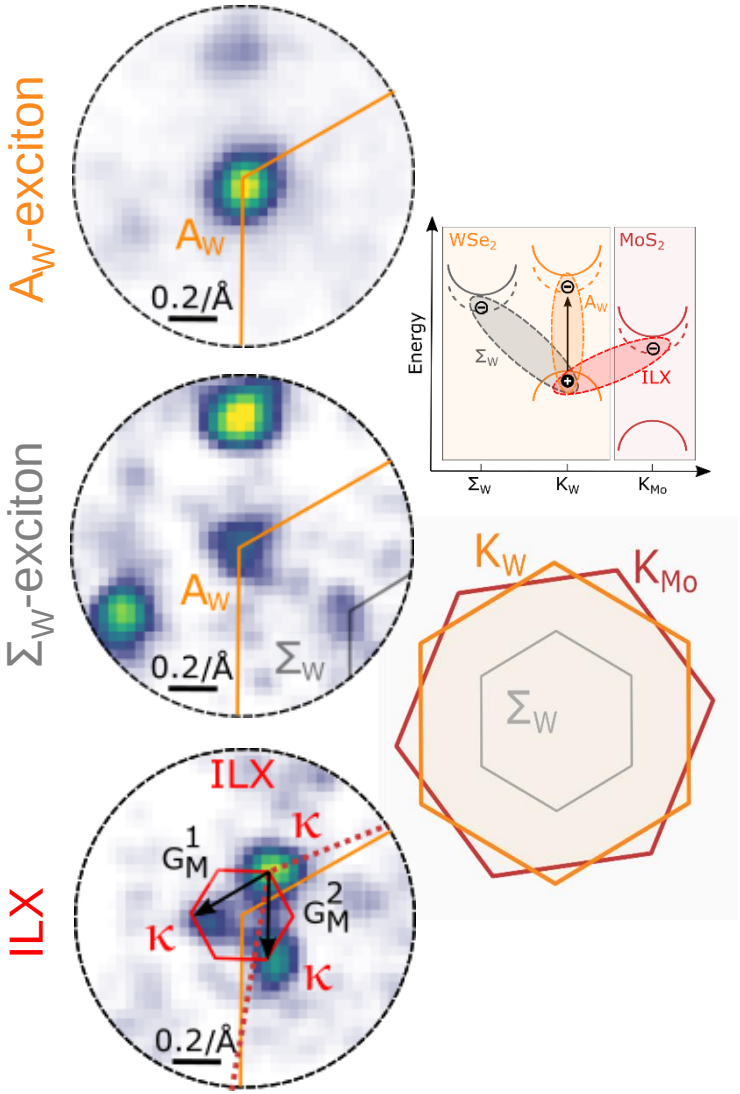
➢ **trARPES: access to correlated states**

ILX formation dynamics



- What is the dominant charge transfer channel?

ILX formation dynamics



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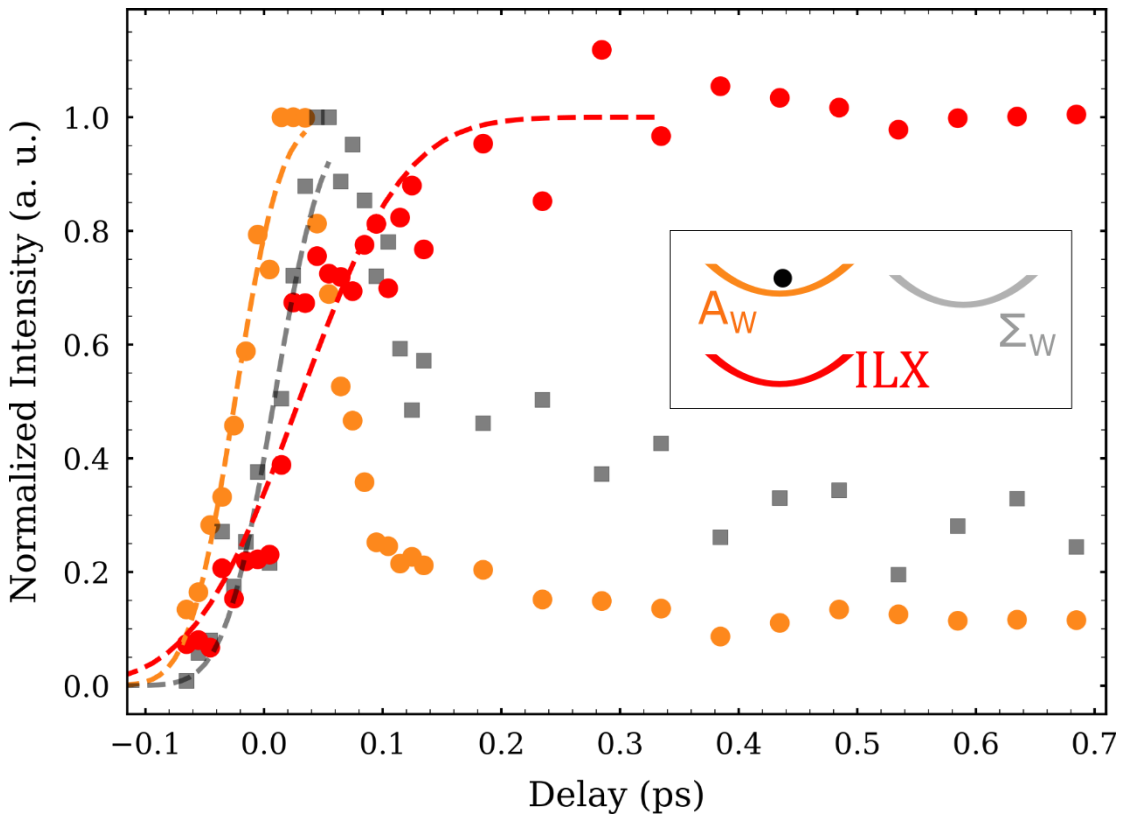
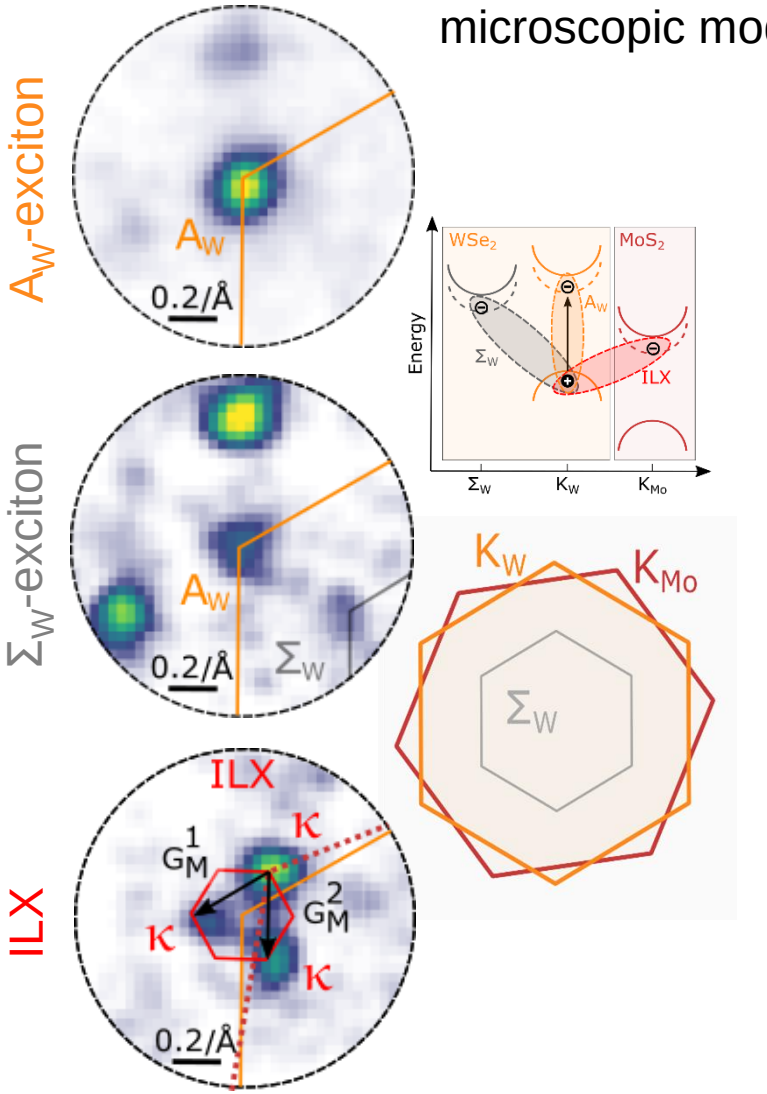


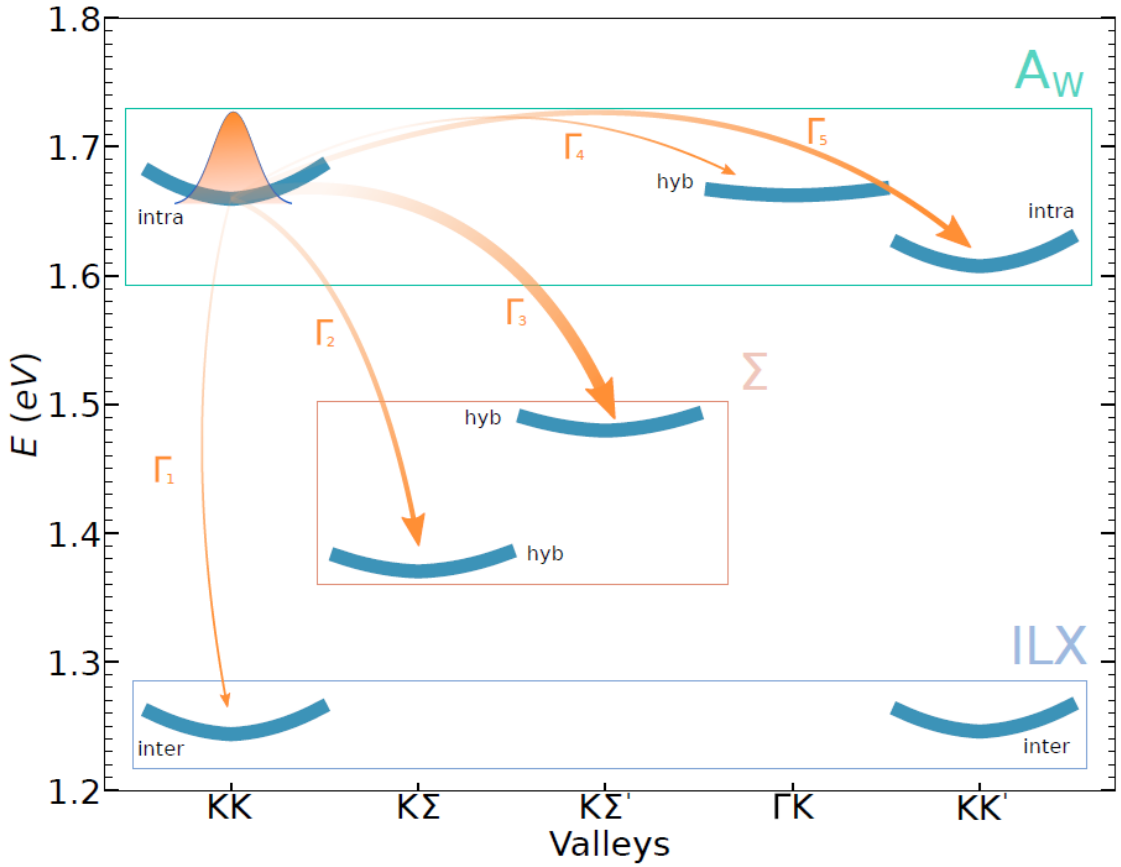
<u>delayed onsets:</u>

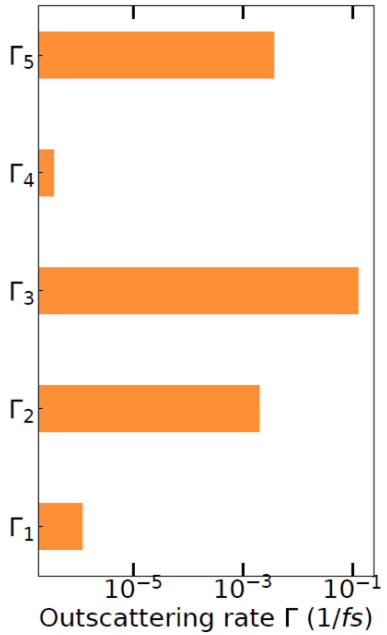
- t_{ILX}-t_A = 54±7 fs
- t_Σ-t_A = 33±6 fs

ILX formation dynamics

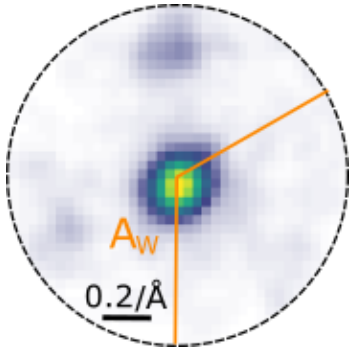
microscopic modelling: Giuseppe Meneghini, Samuel Brem, and Ermin Malic, University of Marburg

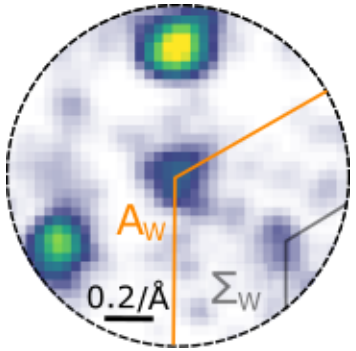


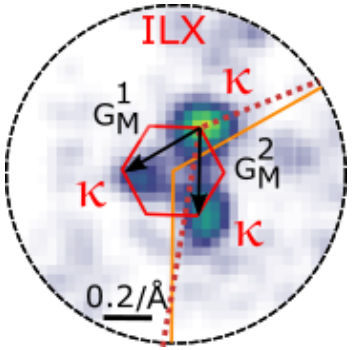




ILX formation dynamics

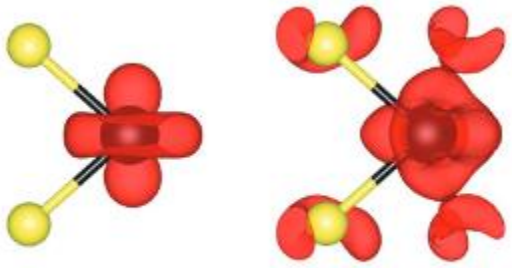




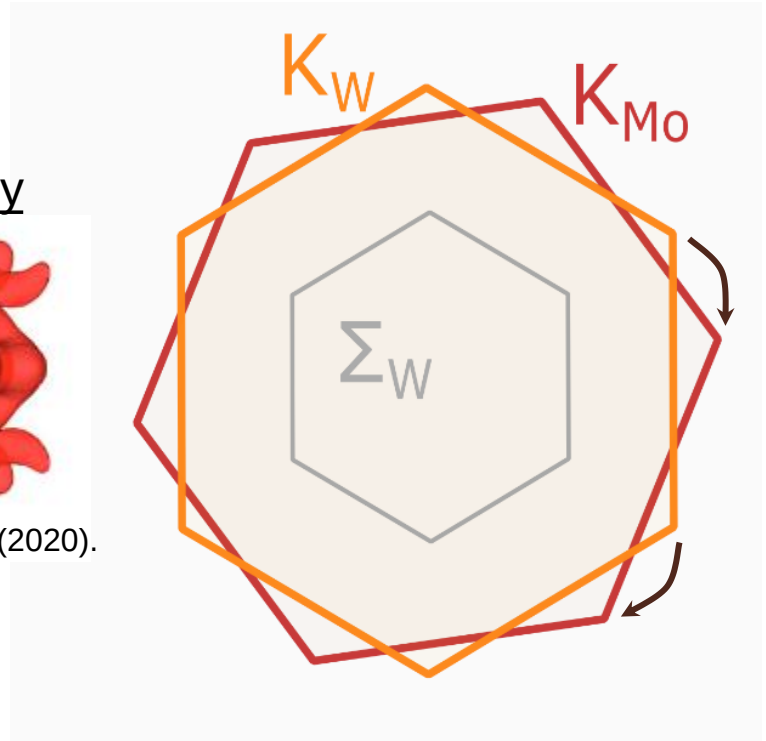


microscopic modelling: Giuseppe Meneghini, Samuel Brem, and Ermin Malic, University of Marburg

K-valley Σ-valley



Brem *et al.*, Nanoscale **12**, 11088 (2020).

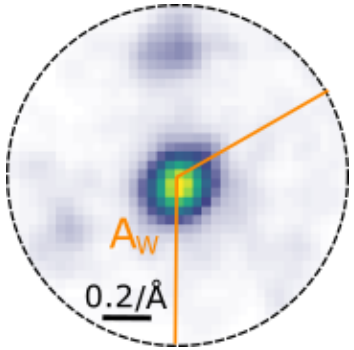


- direct tunneling at the K-valleys is strongly suppressed!

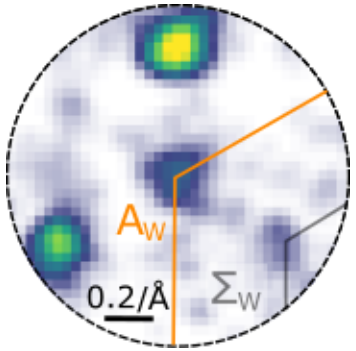
ILX formation dynamics

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

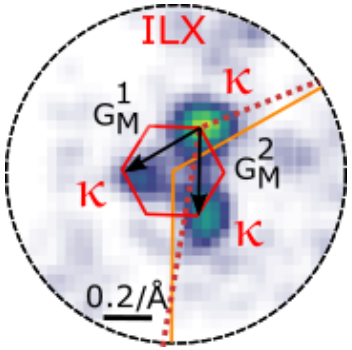
A_W-exciton



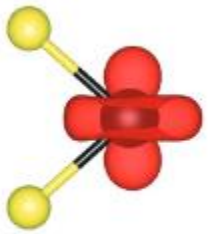
Σ_W-exciton



ILX



K-valley



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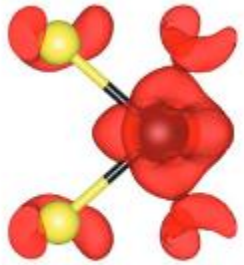

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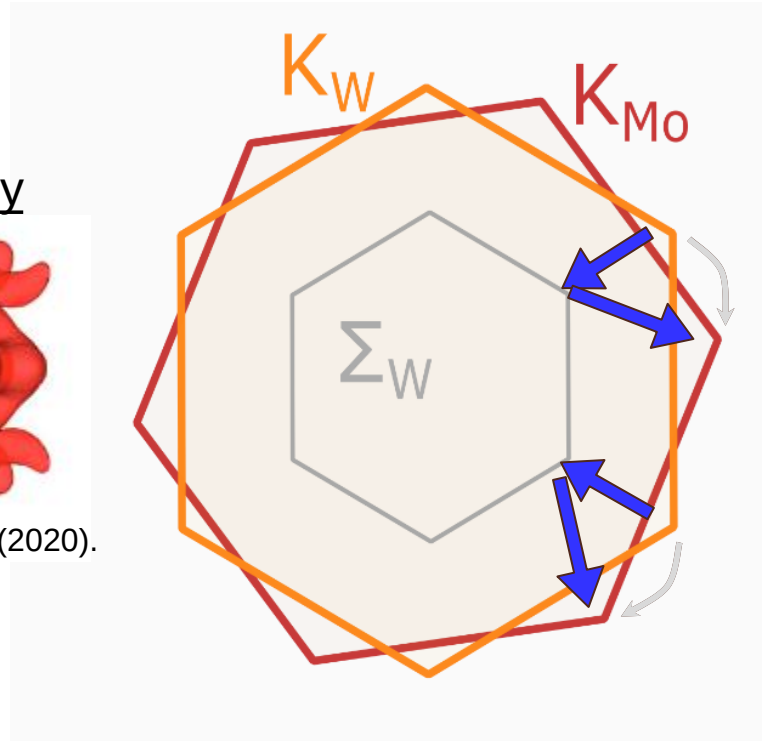
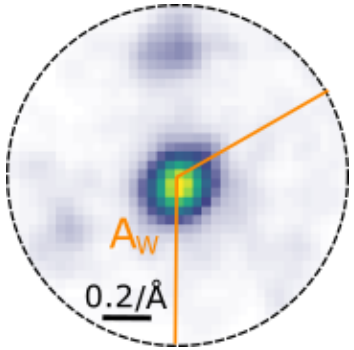


- direct tunneling at the K-valleys is strongly suppressed!
- **factor 100 to 1000 higher scattering rate over Σ valley!**

ILX formation dynamics

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

A_W-exciton



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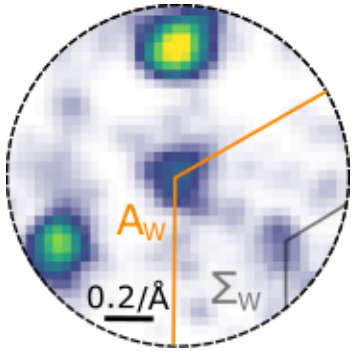
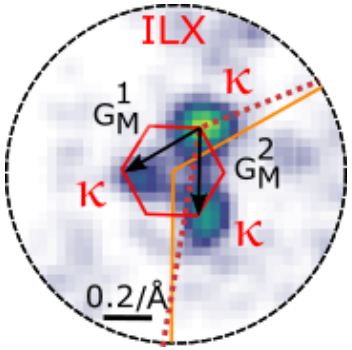
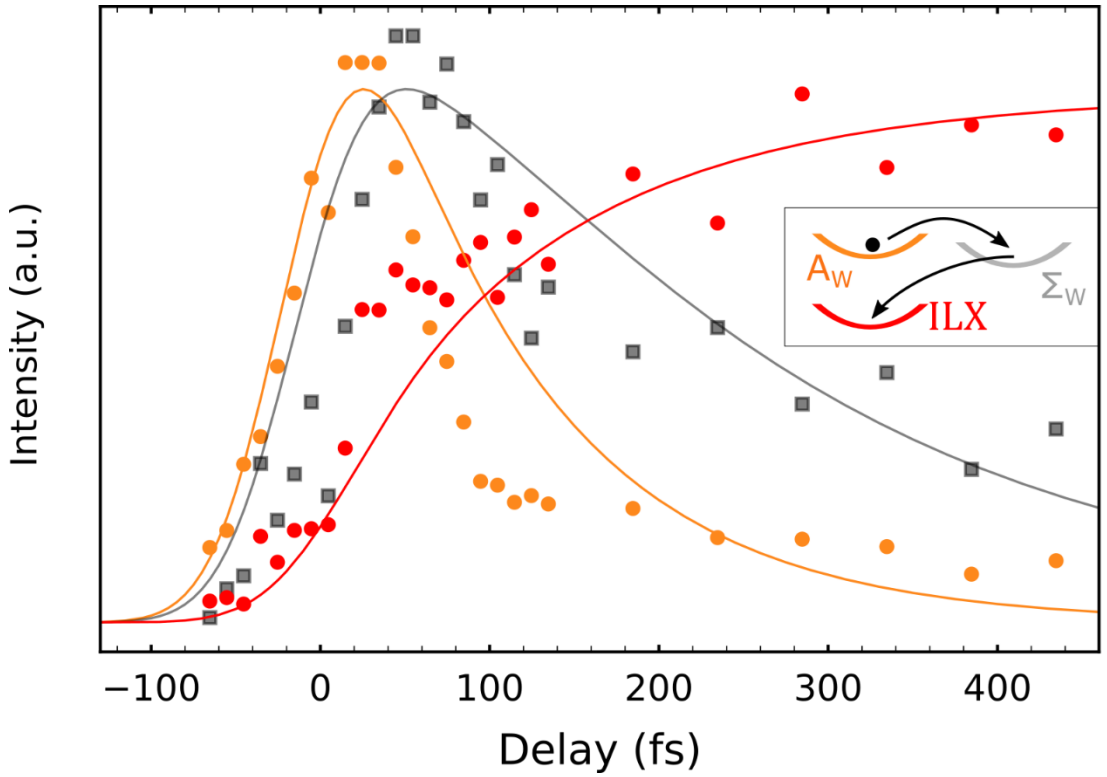


ILX





symbols: experiment
lines: microscopic model

1) exciton-phonon scattering to Σ valley
2) charge transfer at layer hybridized Σ valley

The perfect model system?

take-home messages:

(1) ILX formation: K_W → Σ → K_{Mo}

(2) moiré hallmark on ILX momentum map