

# SYNERGIES IN ULTRAFAST K-MICROSCOPY.



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**HELMHOLTZ**

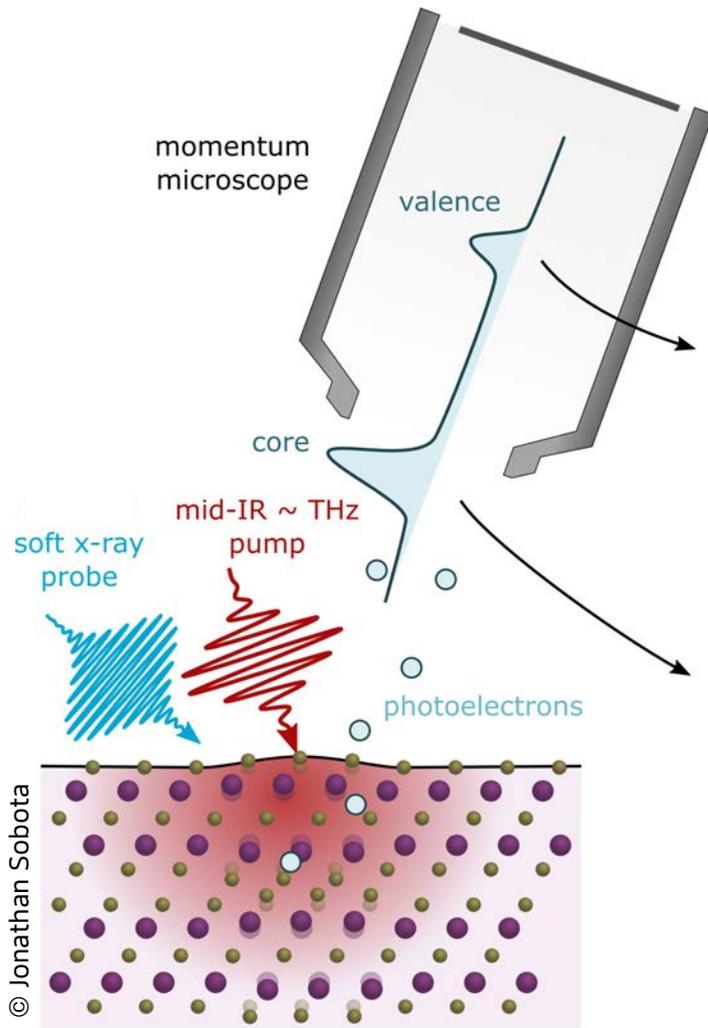


Kiel University  
Christian-Albrechts-Universität zu Kiel

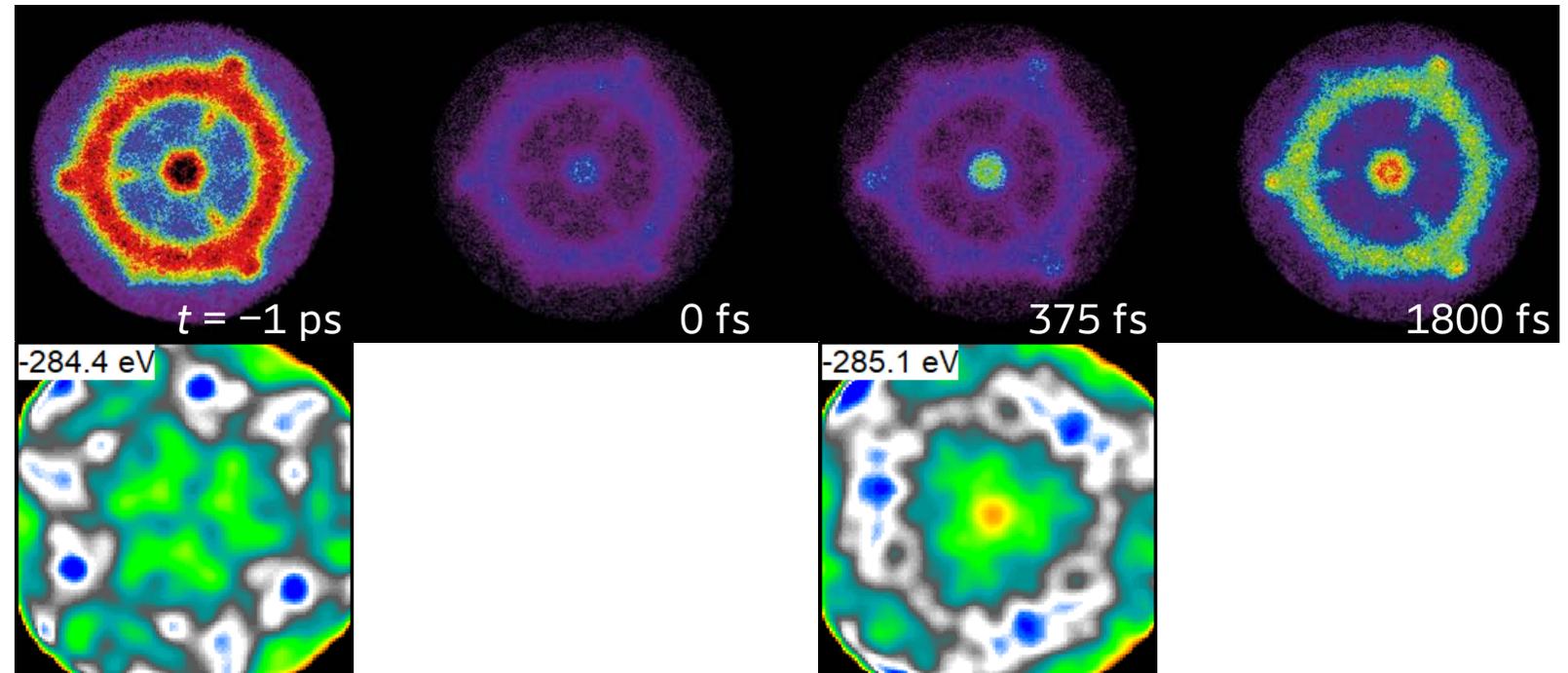


# Soft x-ray flashes make photoemission multimodal & ultrafast

Direct photoelectric tracking of electronic–structural dynamics on their natural timescales—in a single experiment



Quantum valence-*cum*-core electron movie

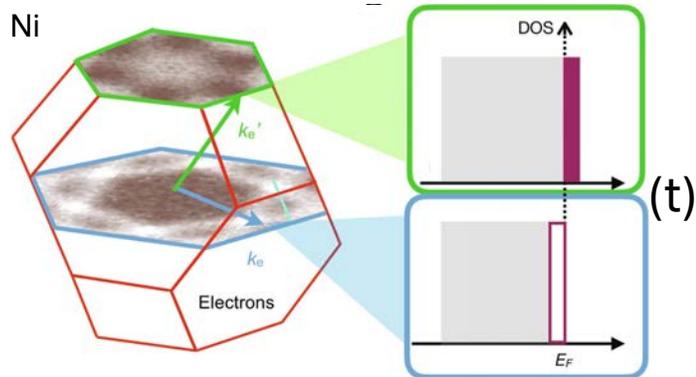


Baumgärtner *et al.*, arXiv:2305.07773

# FLASH has pioneered ultrafast multimodal k-microscopy

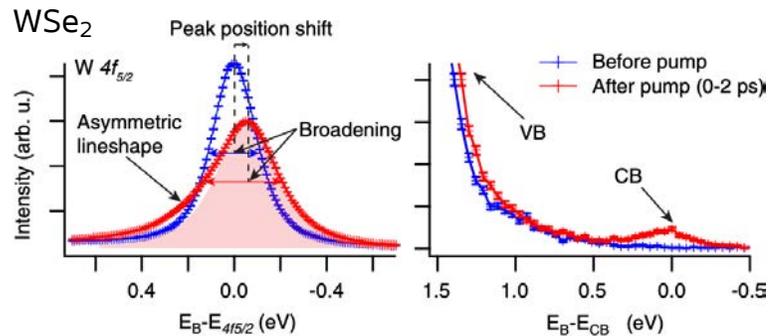
Direct photoelectric imaging of femtosecond electronic, orbital, chemical, and structural dynamics

## Electronic structure dynamics (tr-ARPES)

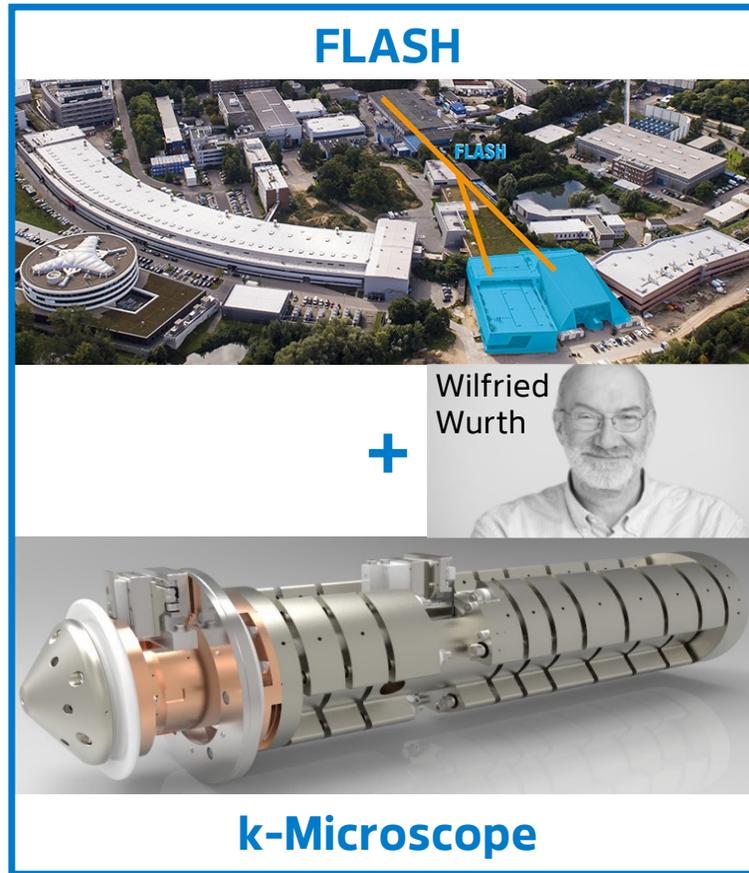


Shokeen *et al.*, *Sci. Adv.* **10**, eadj2407 (2024)

## Local chemical dynamics (tr-XPS)

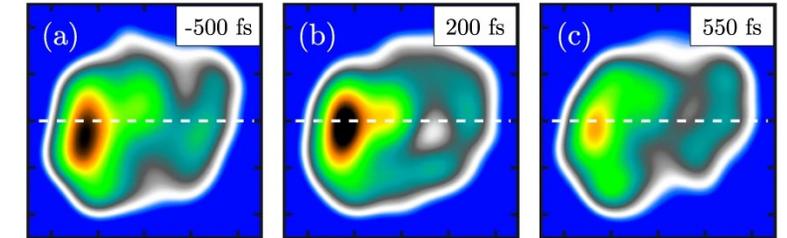


Denzik *et al.*, *Phys. Rev. Lett.* **125**, 096401 (2020)



## Orbital dynamics (tr-ARPES)

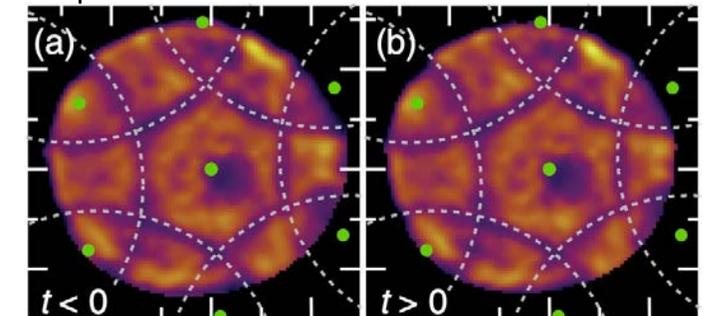
Pentacene / Ag(110)



Baumgärtner *et al.*, *Nat. Commun.* **13**, 2741 (2022)

## Local structural dynamics (tr-XPD)

Graphene



Curcio *et al.*, *Phys. Rev. B* **104**, L161104 (2021)

# The ToF k-microscope is the key enabling technology

Slitless 3D photoelectron energy-momentum imaging



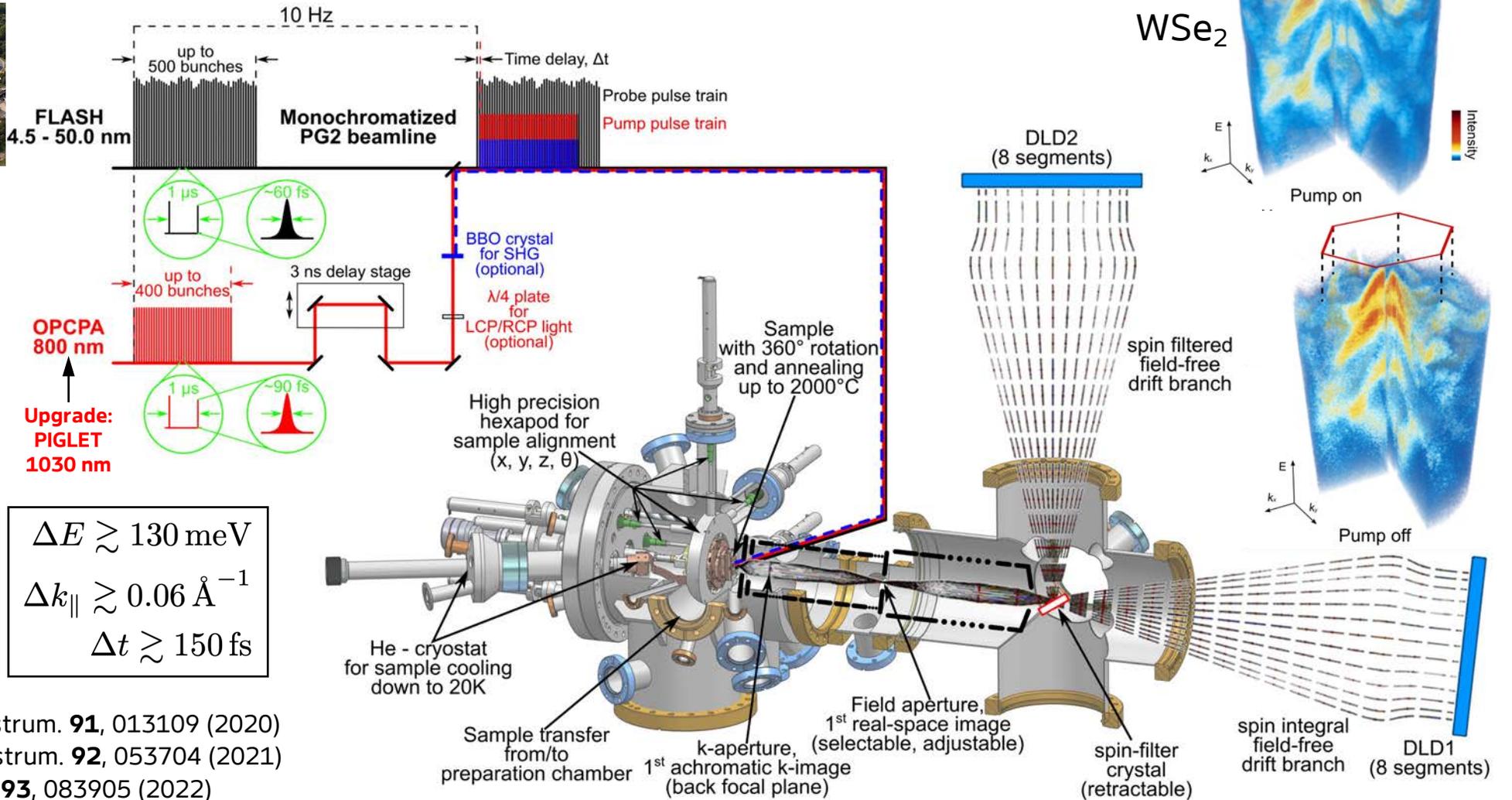
Gerd Schönhense



JOHANNES GUTENBERG  
UNIVERSITÄT MAINZ



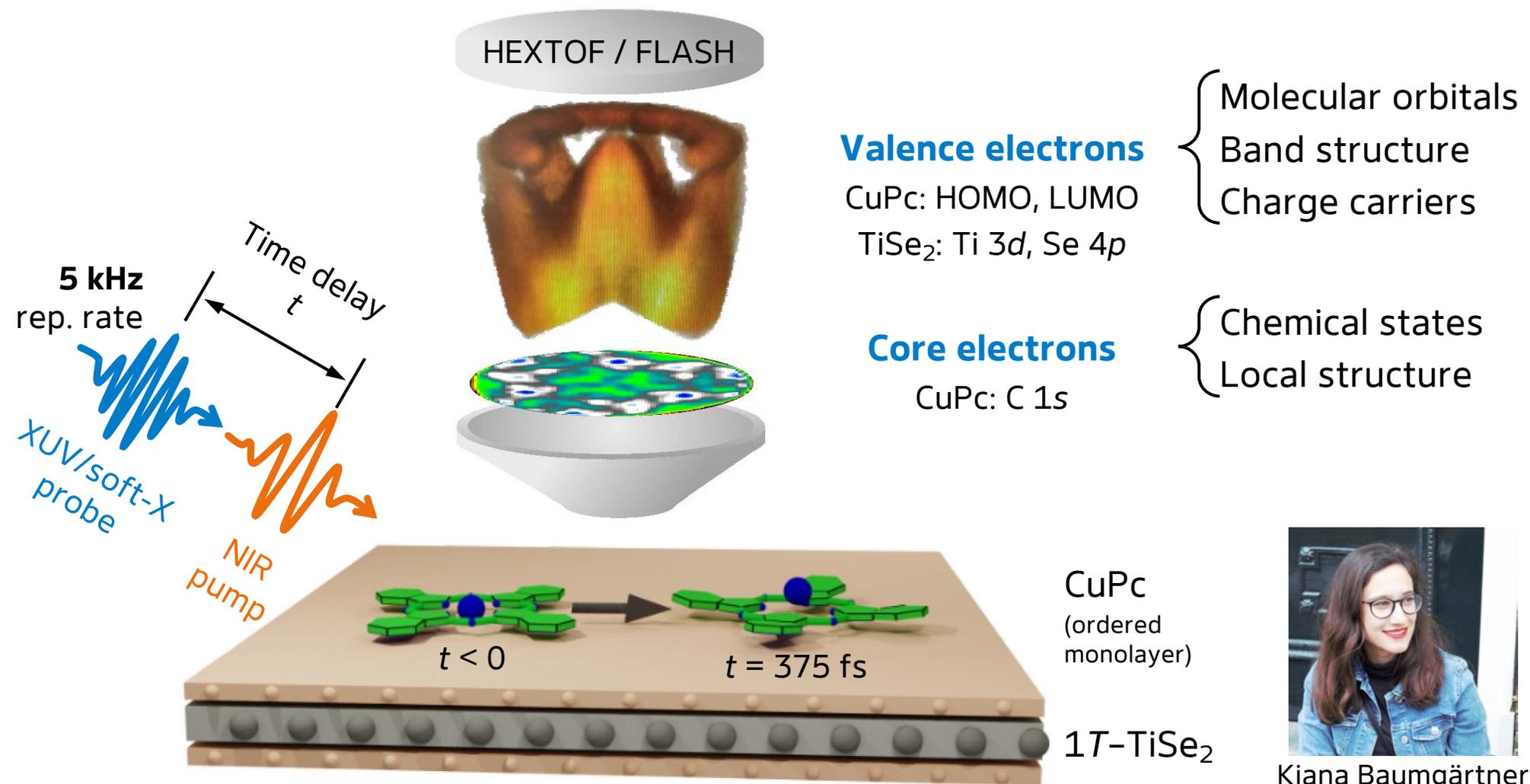
Dima Kutnyakhov



Kutnyakhov *et al.*, Rev. Sci. Instrum. **91**, 013109 (2020)  
 Schönhense *et al.*, Rev. Sci. Instrum. **92**, 053704 (2021)  
 Heber *et al.*, Rev. Sci. Instrum. **93**, 083905 (2022)

# FLASH k-microscopy creates quantum duplex electron movies

Femtostroboscopic imaging of valence-*cum*-core electron dynamics at a molecule-material interface



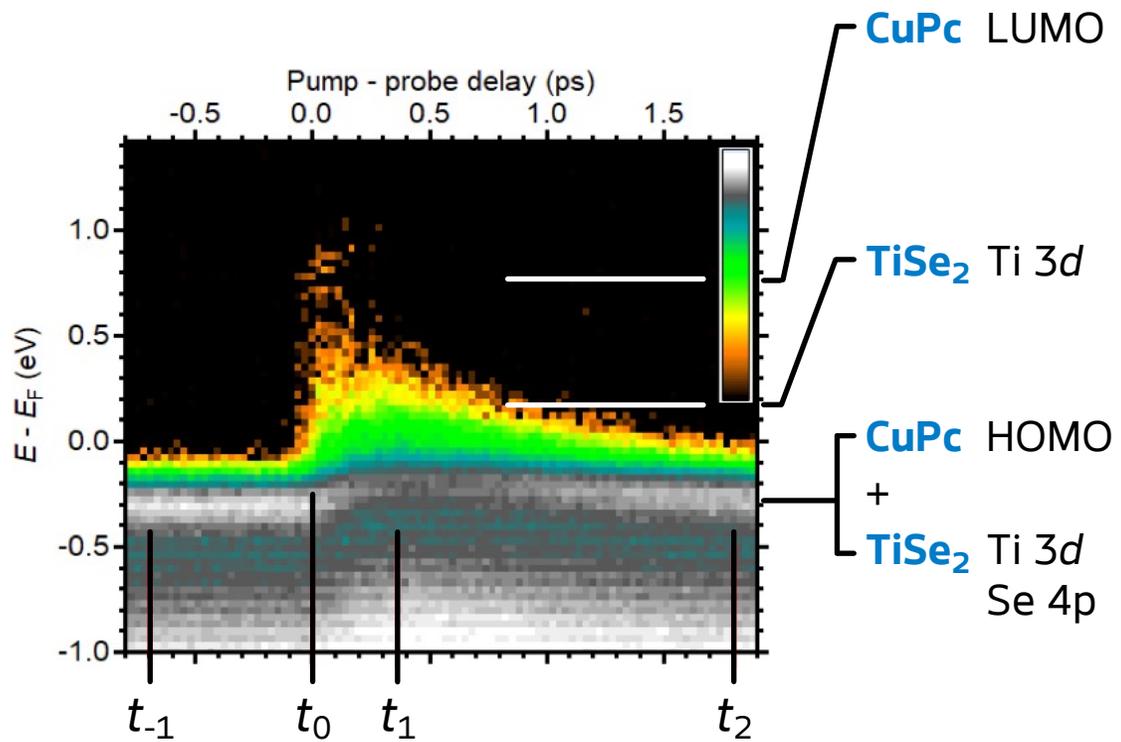
Kiana Baumgärtner



Markus Scholz

# FLASH k-microscopy reveals electronic structure dynamics

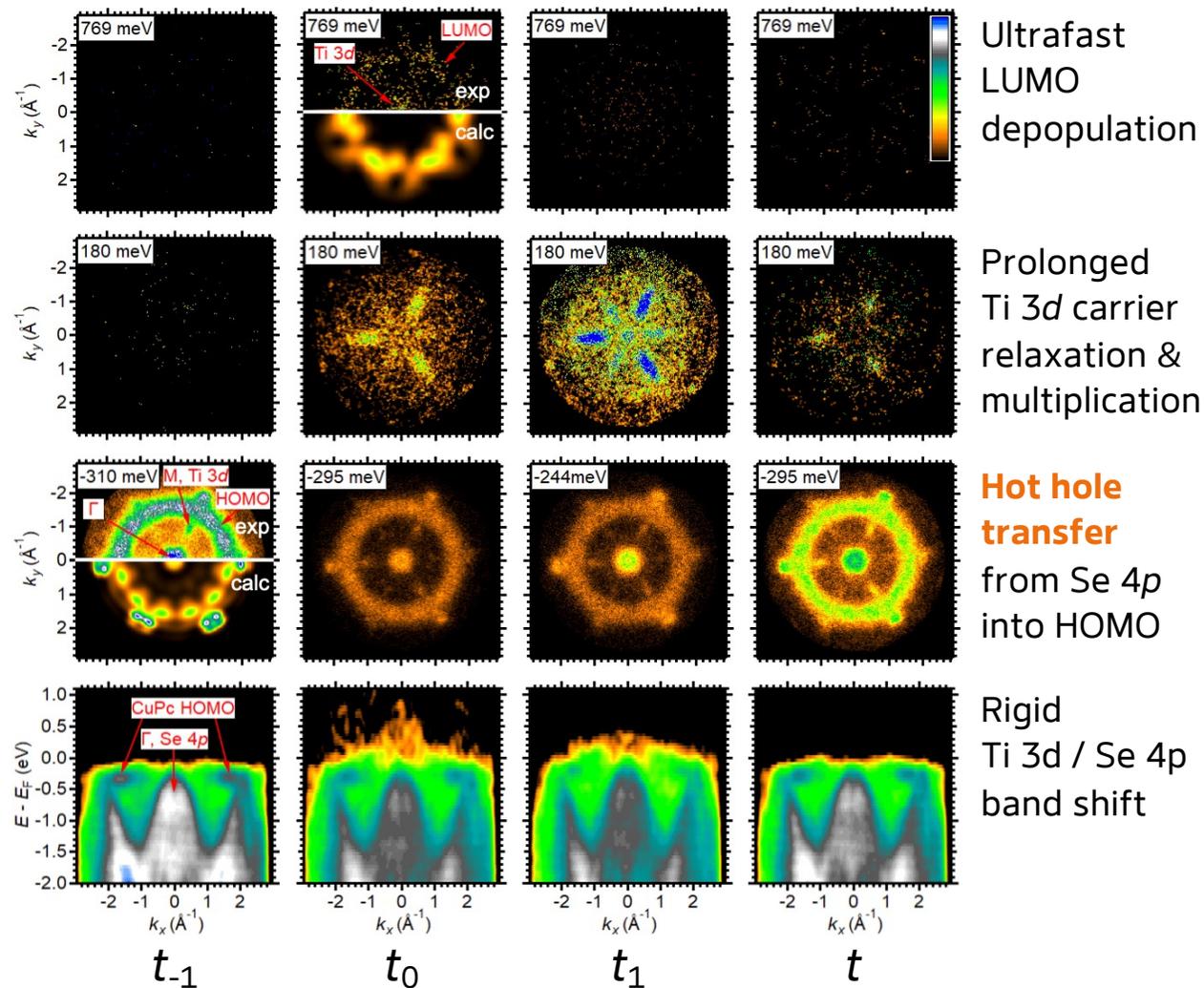
Femtostroboscopic imaging of transient k-dependent bands and orbitals near the Fermi level



$h\nu_{\text{probe}} = 36.3 \text{ eV}$   
 $h\nu_{\text{pump}} = 1.6 \text{ eV}$   
 $F \approx 3.5 \text{ mJ/cm}^2$

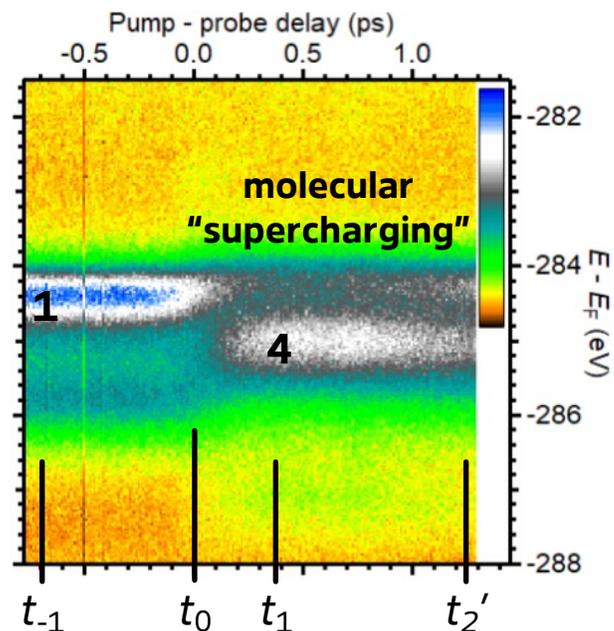
Baumgärtner *et al.*,  
 arXiv:2305.07773

DESY



# FLASH k-microscopy reveals chemical-structural dynamics

Femtostroboscopic imaging of transient site-specific core-level emissions



**Theory**

XPS: Marvin Reuner, Daria Gorelova

XPD: Kaori Niki, Masato Nozakami,

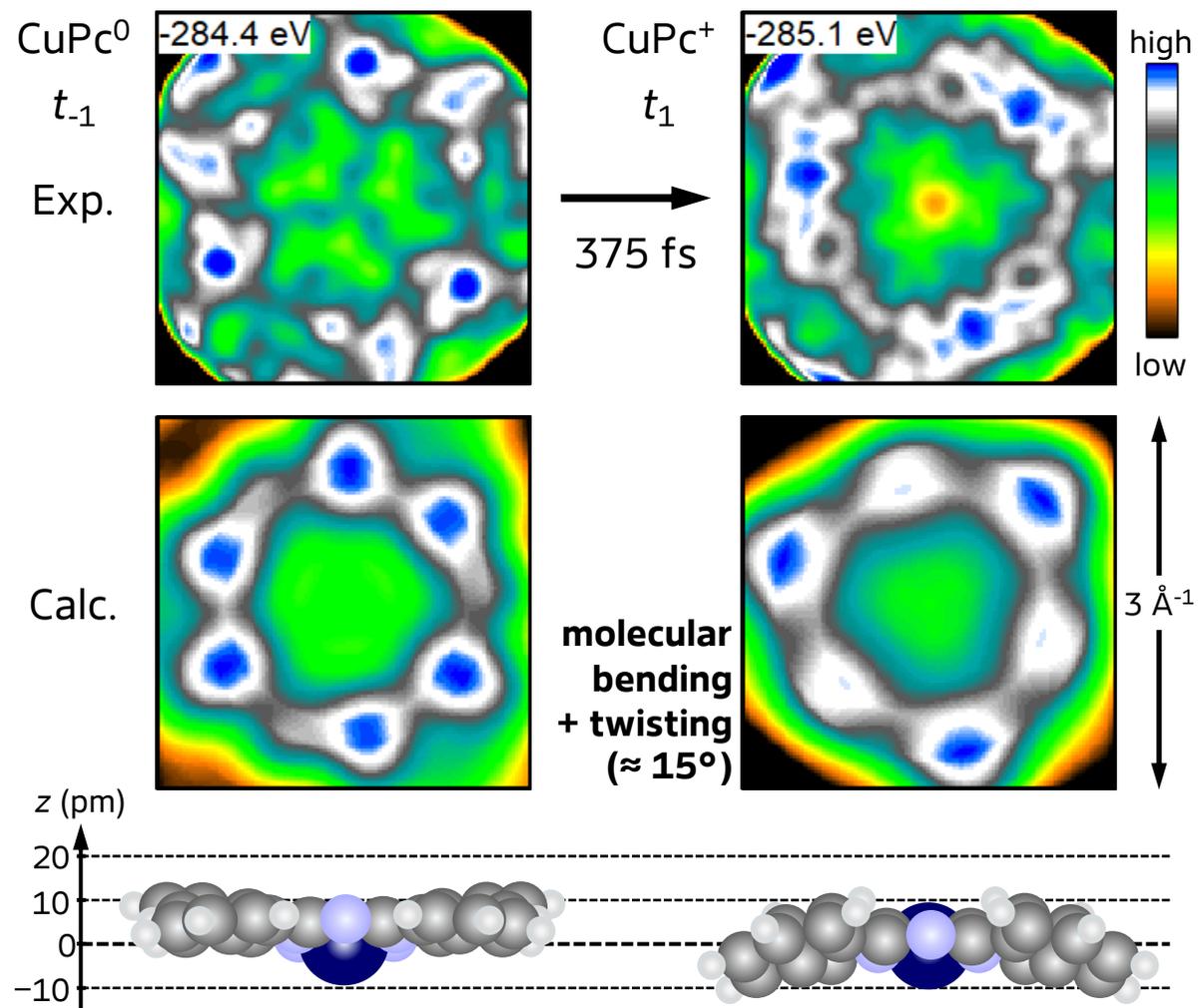
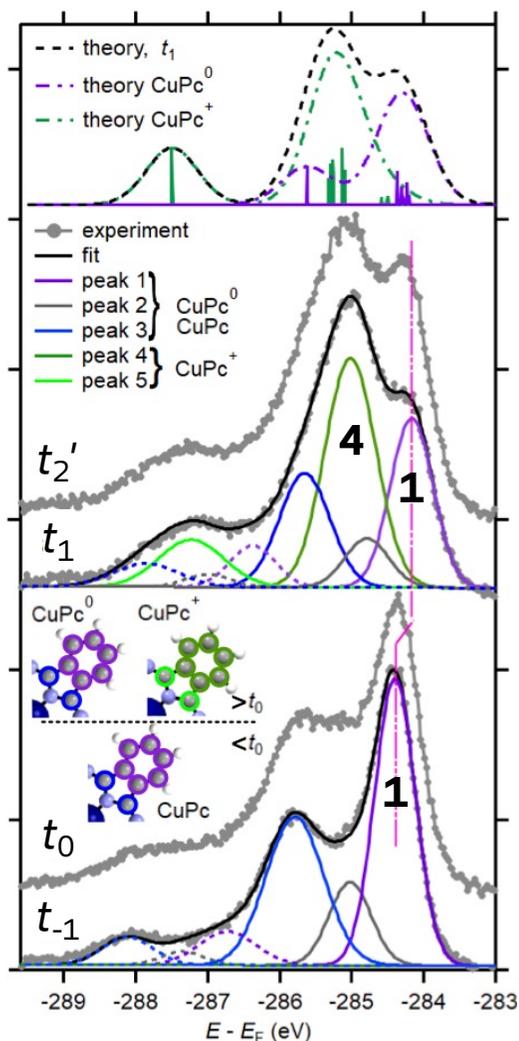
Misa Nozaki

$$h\nu_{\text{probe}} = 371 \text{ eV}$$

$$h\nu_{\text{pump}} = 1.6 \text{ eV}$$

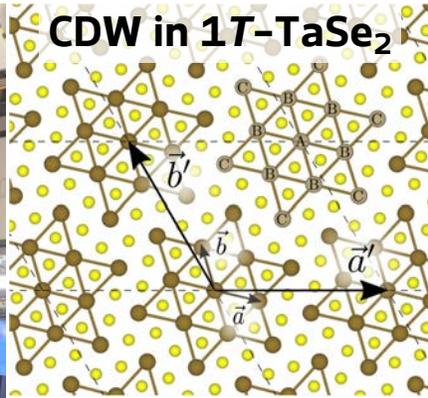
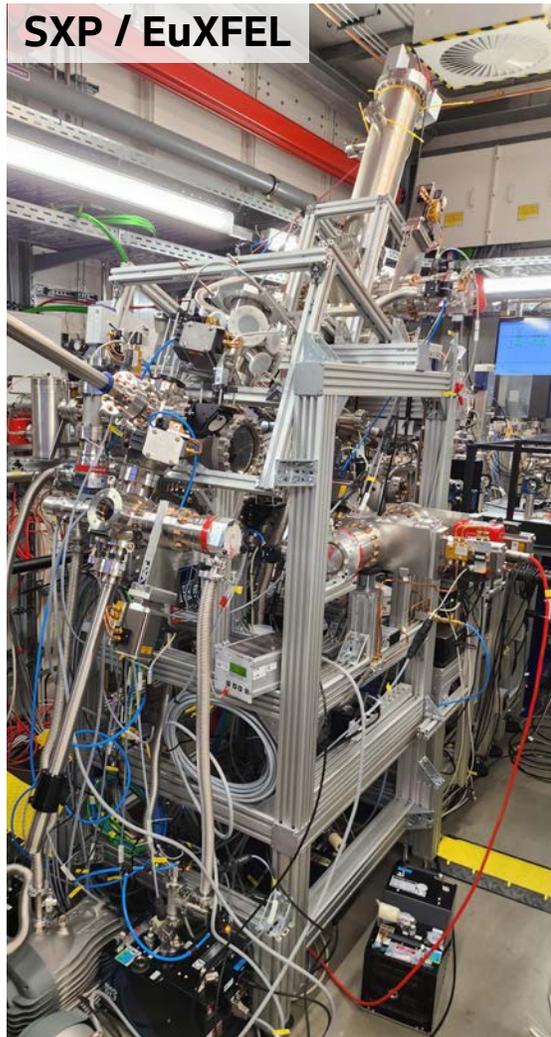
$$F \approx 3.5 \text{ mJ/cm}^2$$

Baumgärtner *et al.*,  
arXiv:2305.07773



# EuXFEL k-microscopy has pioneered ultrafast "real" XPS

Femtostroboscopic tracking of bulk Ta 4f emissions reveals the nature of the charge-density wave in TaSe<sub>2</sub>.

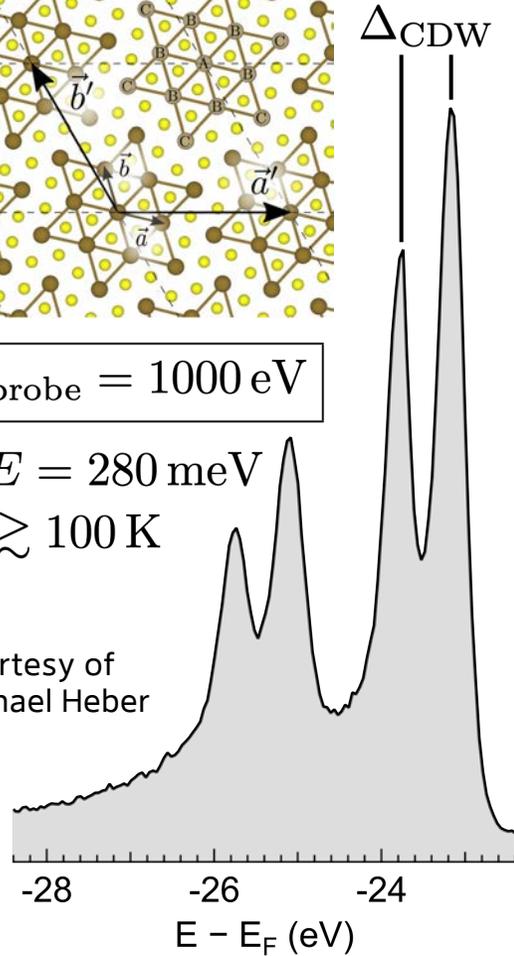


$$h\nu_{\text{probe}} = 1000 \text{ eV}$$

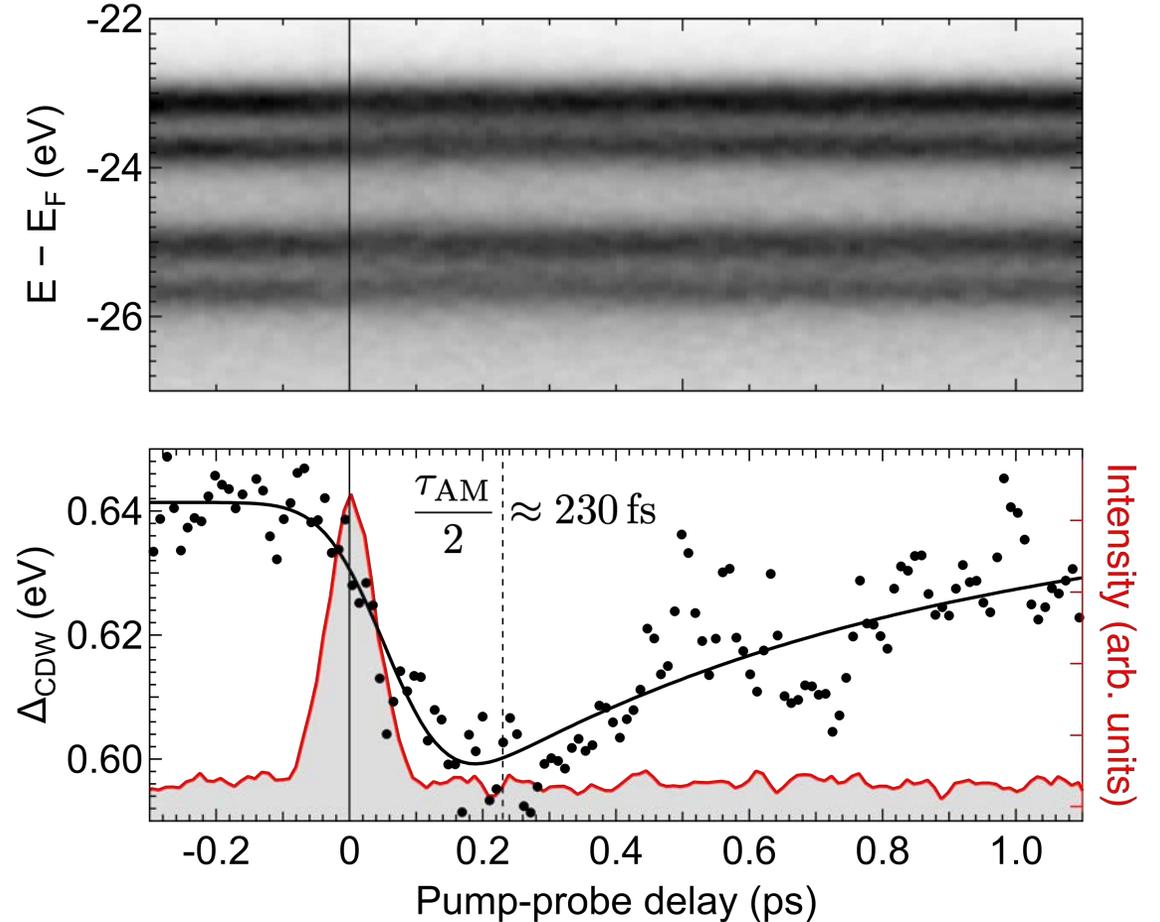
$$\Delta E = 280 \text{ meV}$$

$$T \gtrsim 100 \text{ K}$$

Courtesy of Michael Heber

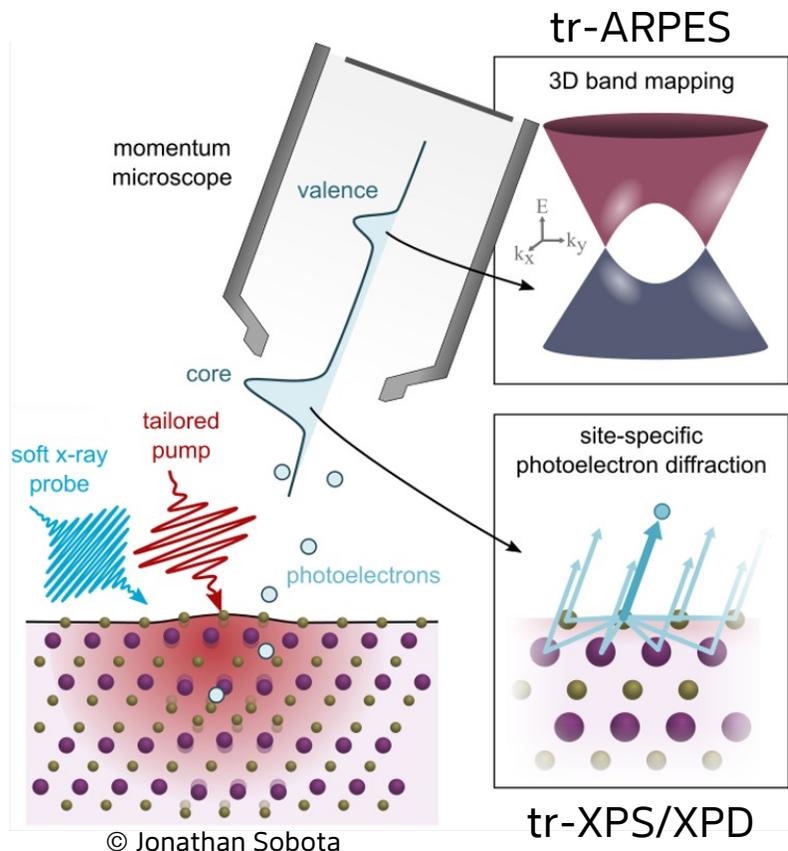


$$h\nu_{\text{pump}} = 1.55 \text{ eV}. \quad F \approx 6.3 \text{ mJ/cm}^2. \quad \Delta t \approx 90 \text{ fs}$$



# Ultrafast k-microscopy fosters source synergism

We combine complementary photon sources to create the all-in-one valence-*cum*-core electron movie.



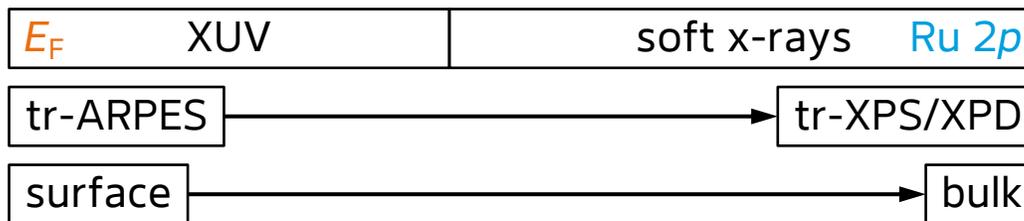
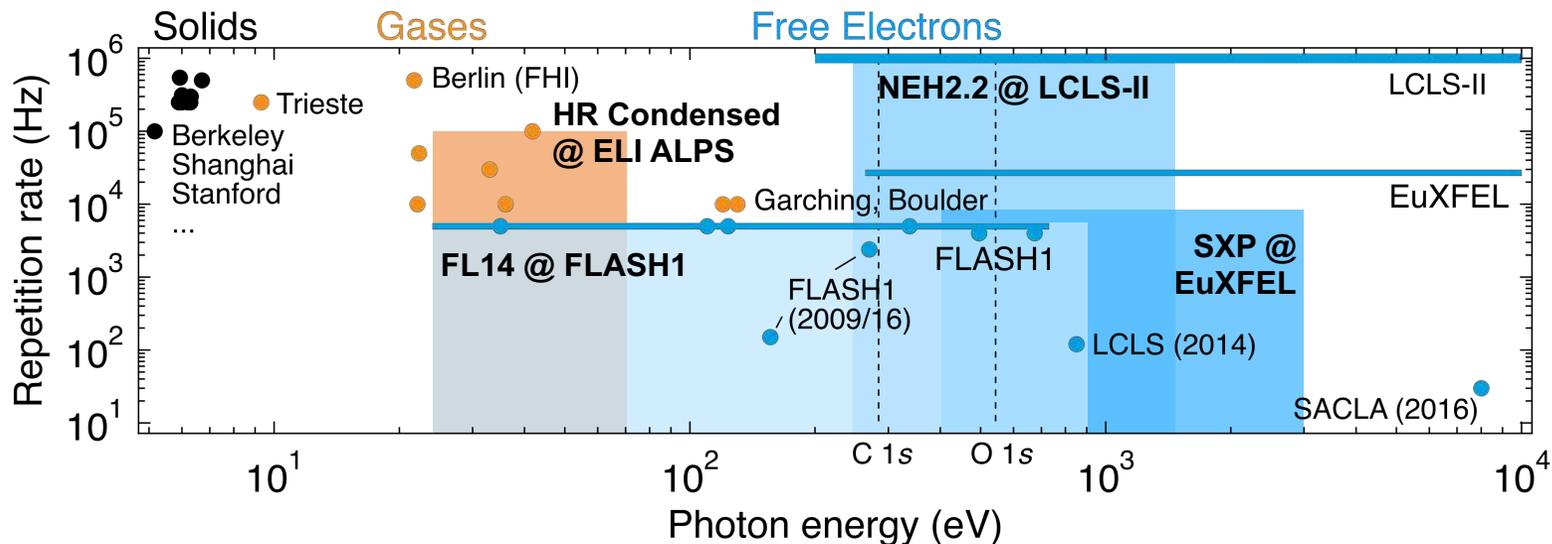
**HHG**  
(CFEL)



**FLASH**  
(FL14, FL23)



**EuXFEL**  
(SXP)



# Seeded FLASH advances ultrafast k-microscopy

Higher precision. Higher resolution. Higher sensitivity. Circular dichroism.

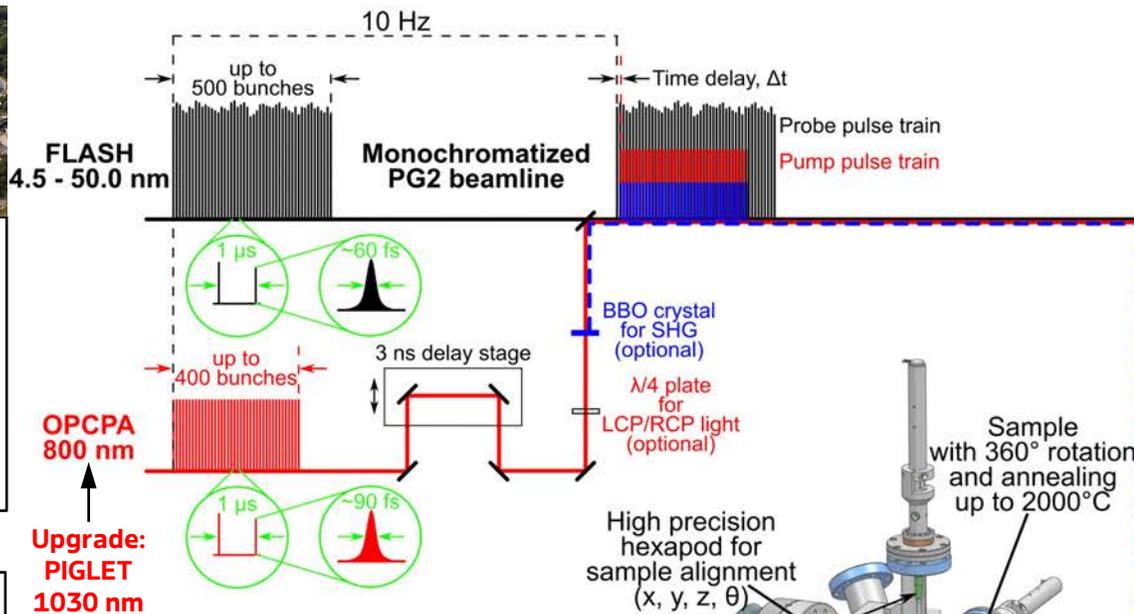


## Seeded FLASH1

- shorter pulses
- narrower bandwidth
- shot-to-shot stability
- **polarization control**

## tr-k-microscopy

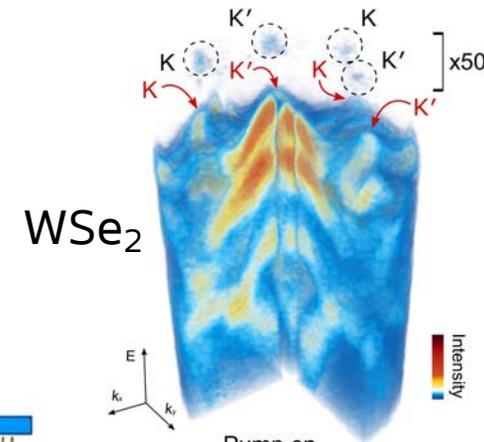
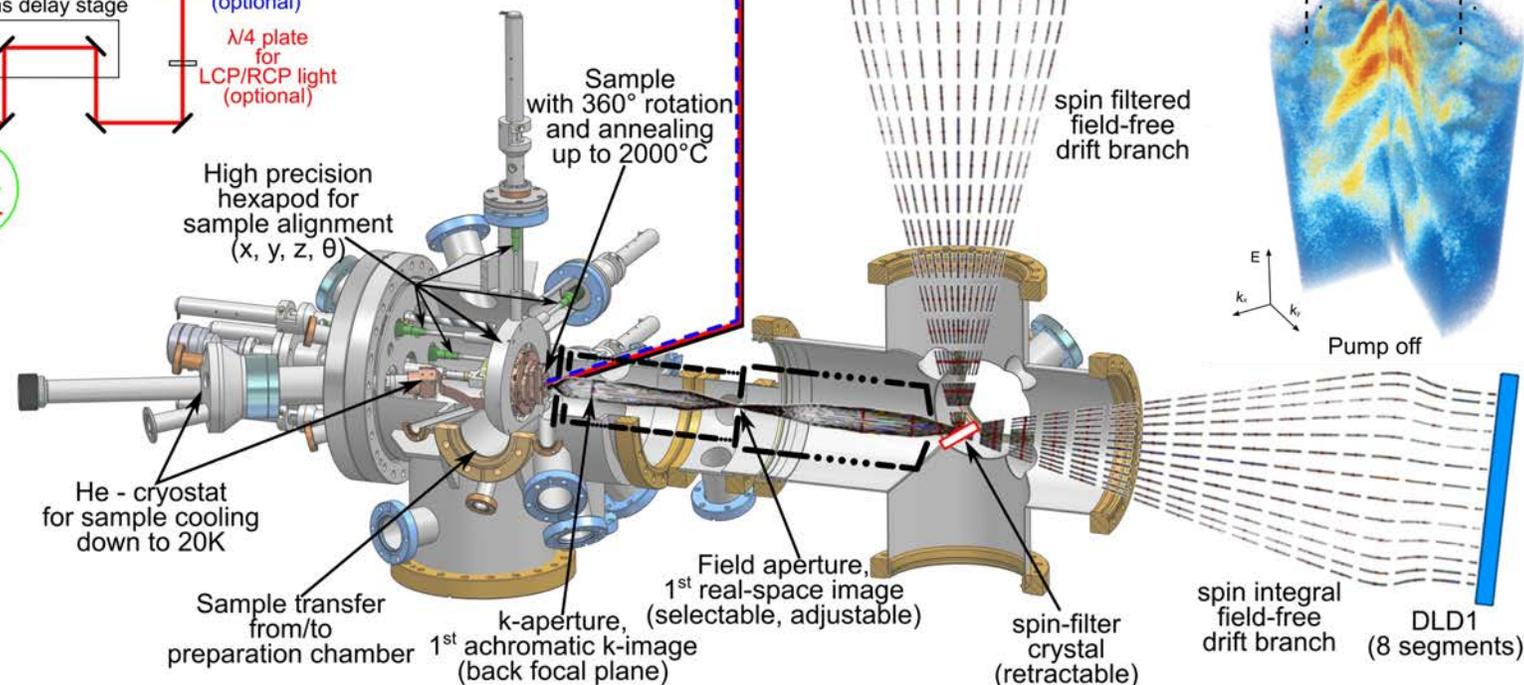
- higher resolution (time & energy)
- higher precision
- higher sensitivity



$$\Delta E < 100 \text{ meV}$$

$$\Delta k_{\parallel} \leq 0.05 \text{ \AA}^{-1}$$

$$\Delta t < 100 \text{ fs}$$

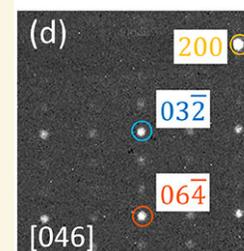
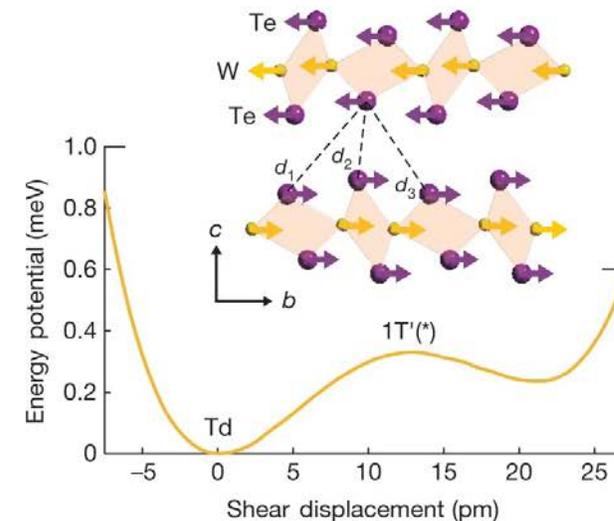
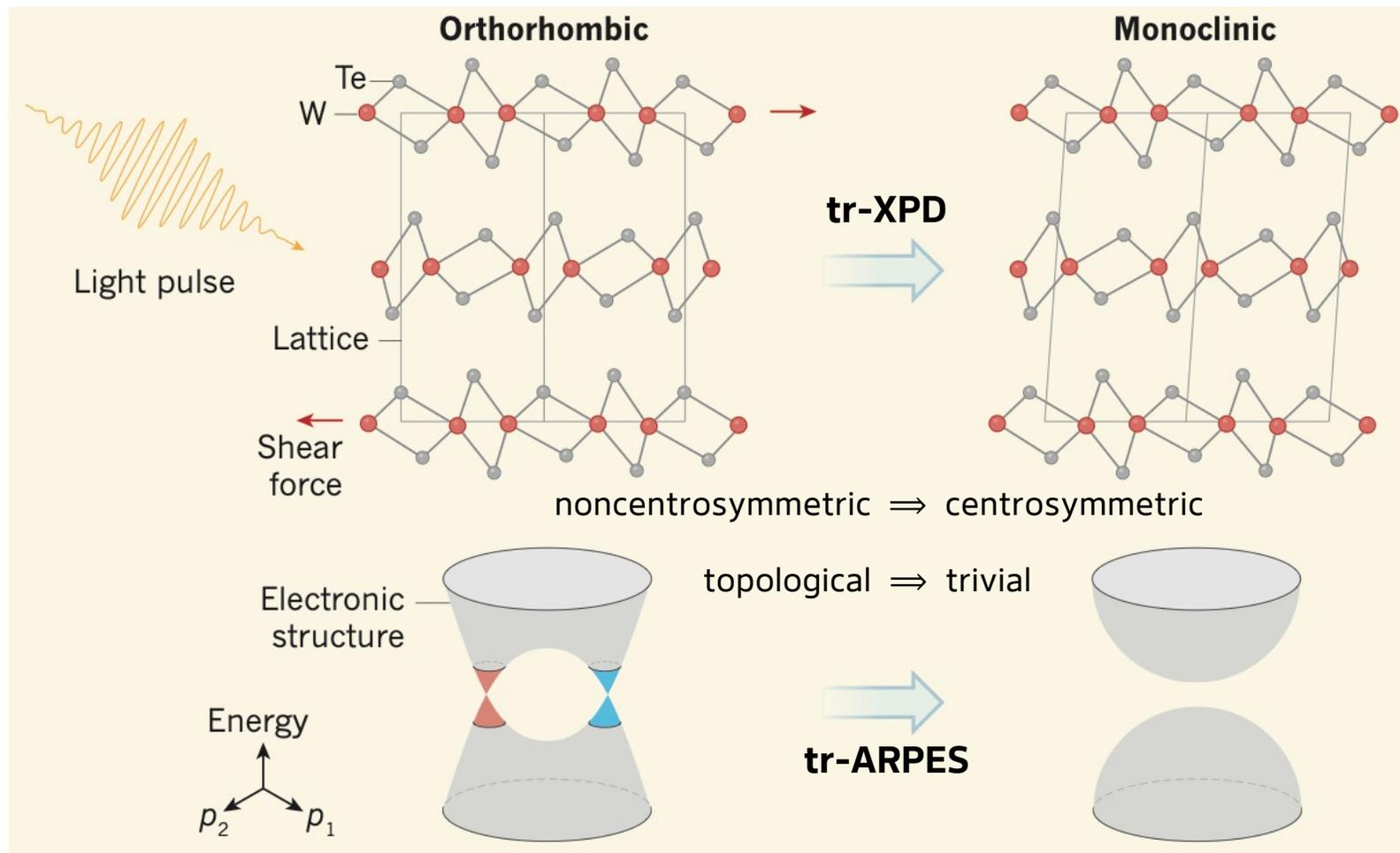


Kutnyakhov *et al.*, Rev. Sci. Instrum. **91**, 013109 (2020)  
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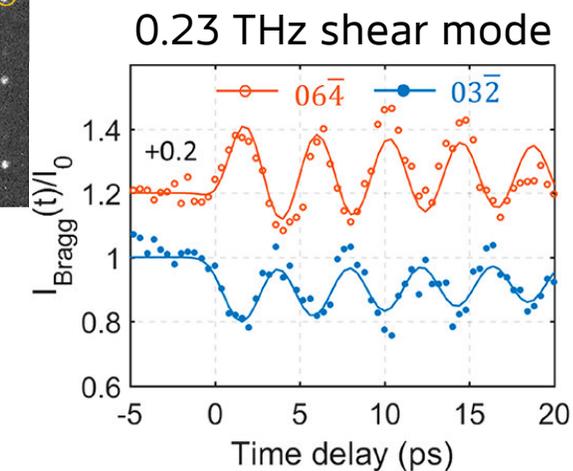


# Light pulse switches structure switches topology

sFLASH Community Proposal: Ultrafast duplex k-microscopy of Td-WTe<sub>2</sub>



515 nm  
(2.4 eV)  
pump

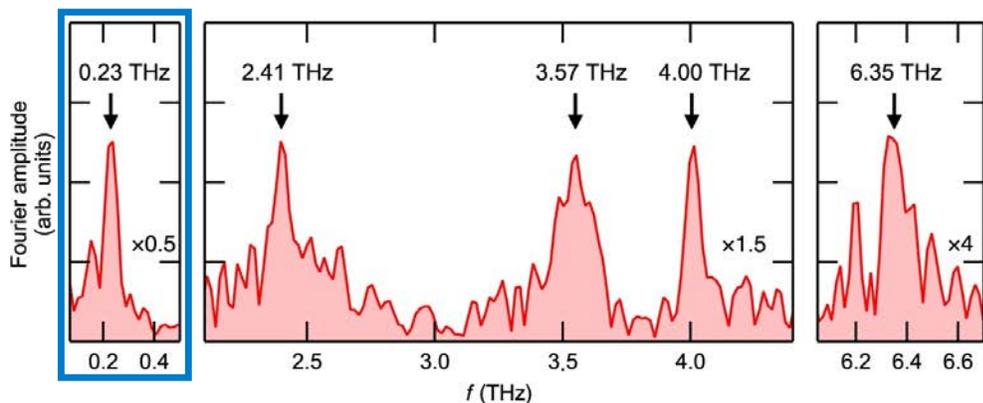
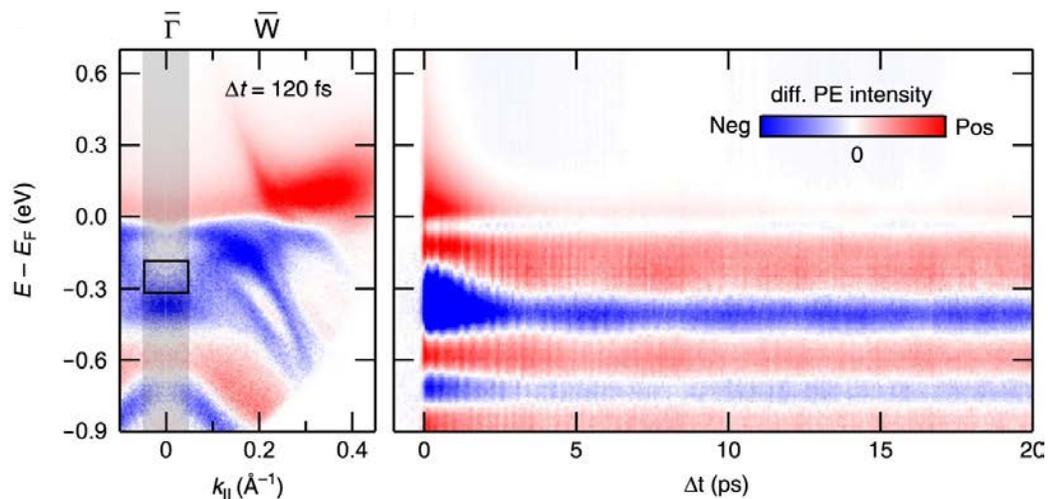


Son, Nature **565**, 32 (2019); Sie *et al.*, Nature **565**, 61 (2019); Ji *et al.*, ACS Nano **15**, 8826 (2021)

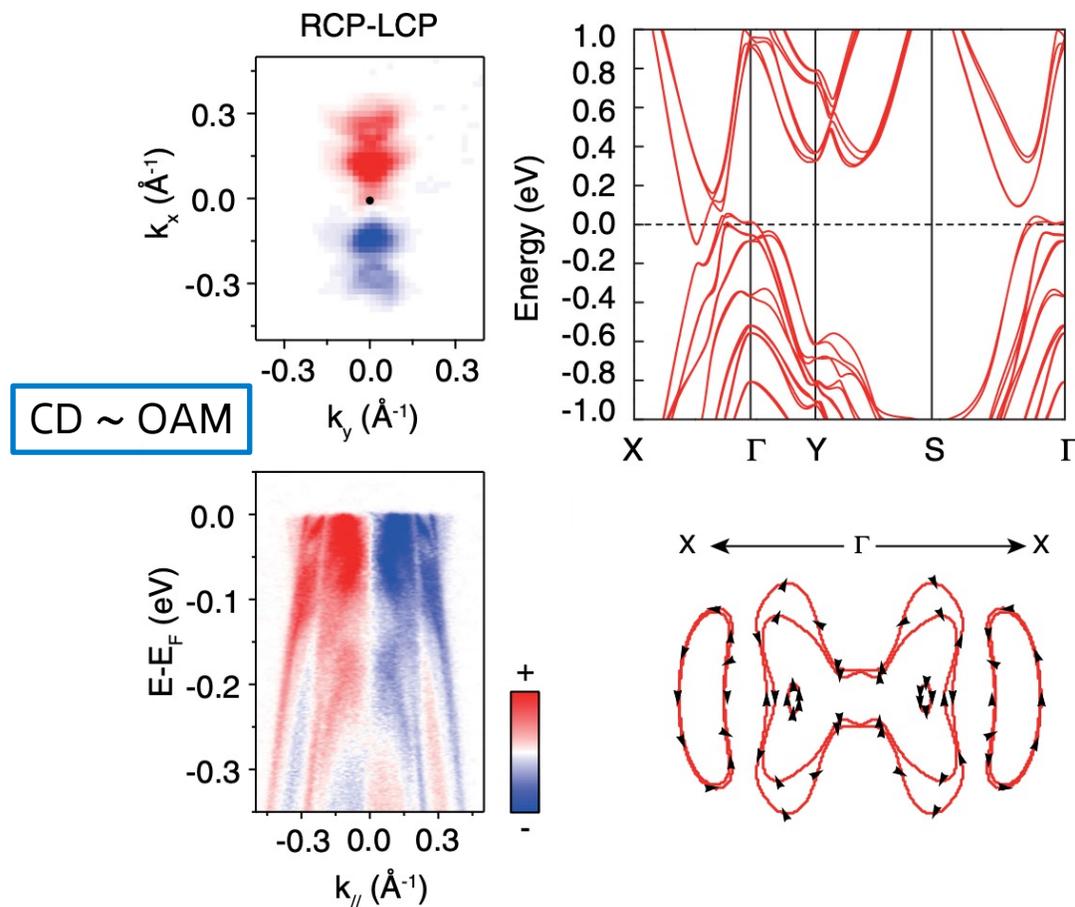
# Chiral soft x-ray flashes can trace e-ph-OAM dynamics

sFLASH Community Proposal: Ultrafast circular dichroism k-microscopy of Td-WTe<sub>2</sub>

Coherent phonons



Orbital angular momentum & spin texture

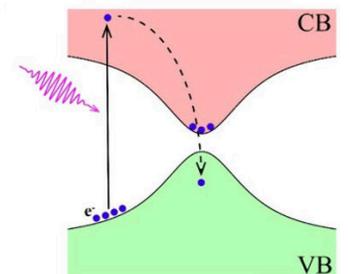
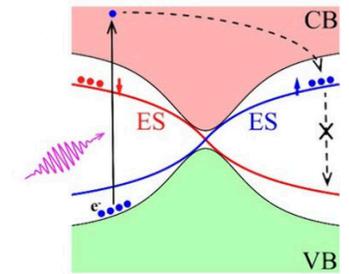
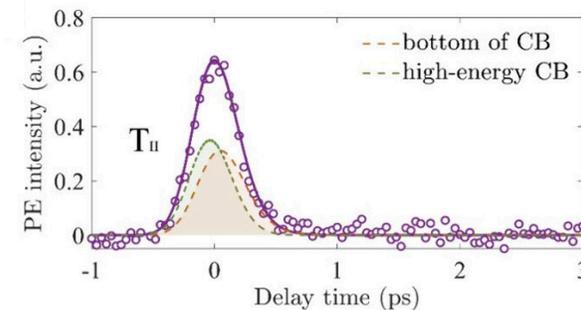
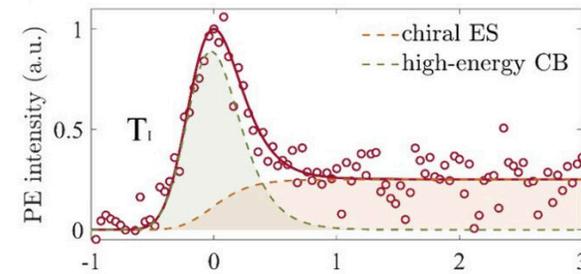
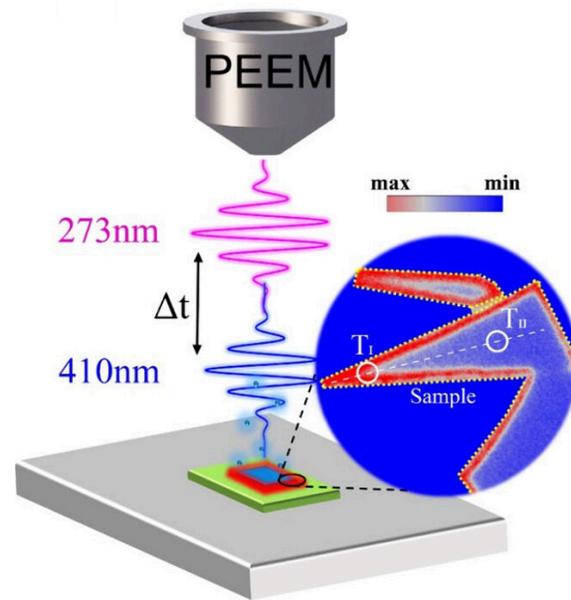
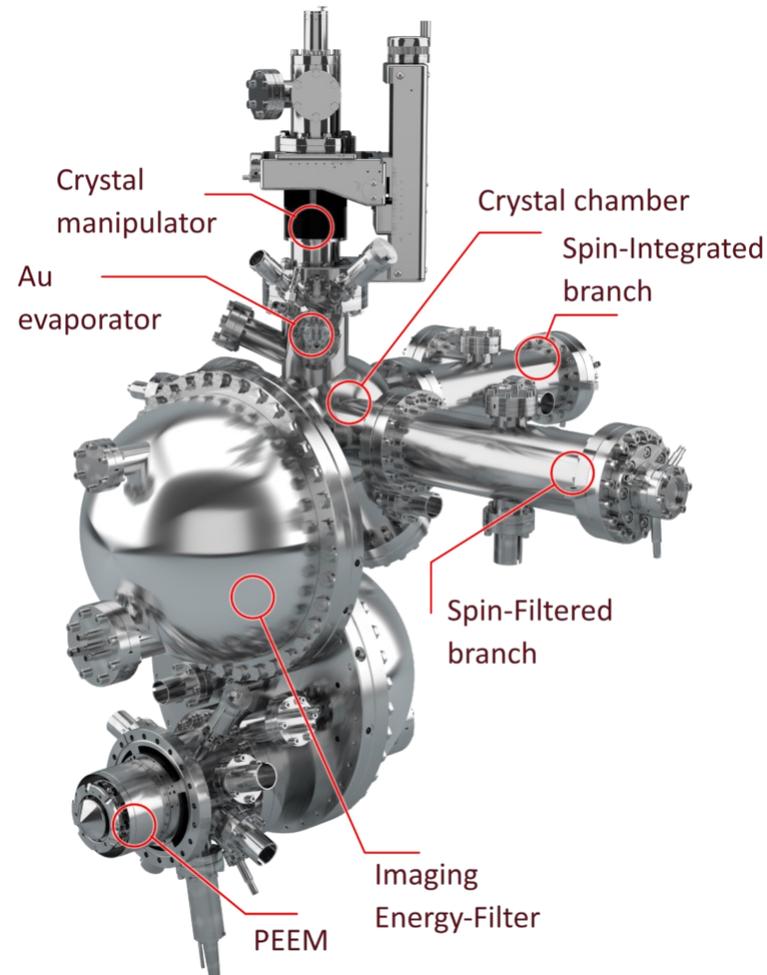


Hein *et al.*, Nat. Commun. **11**, 2613 (2020)

Jiang *et al.*, Phys. Rev. Lett. **115**, 166601 (2015)

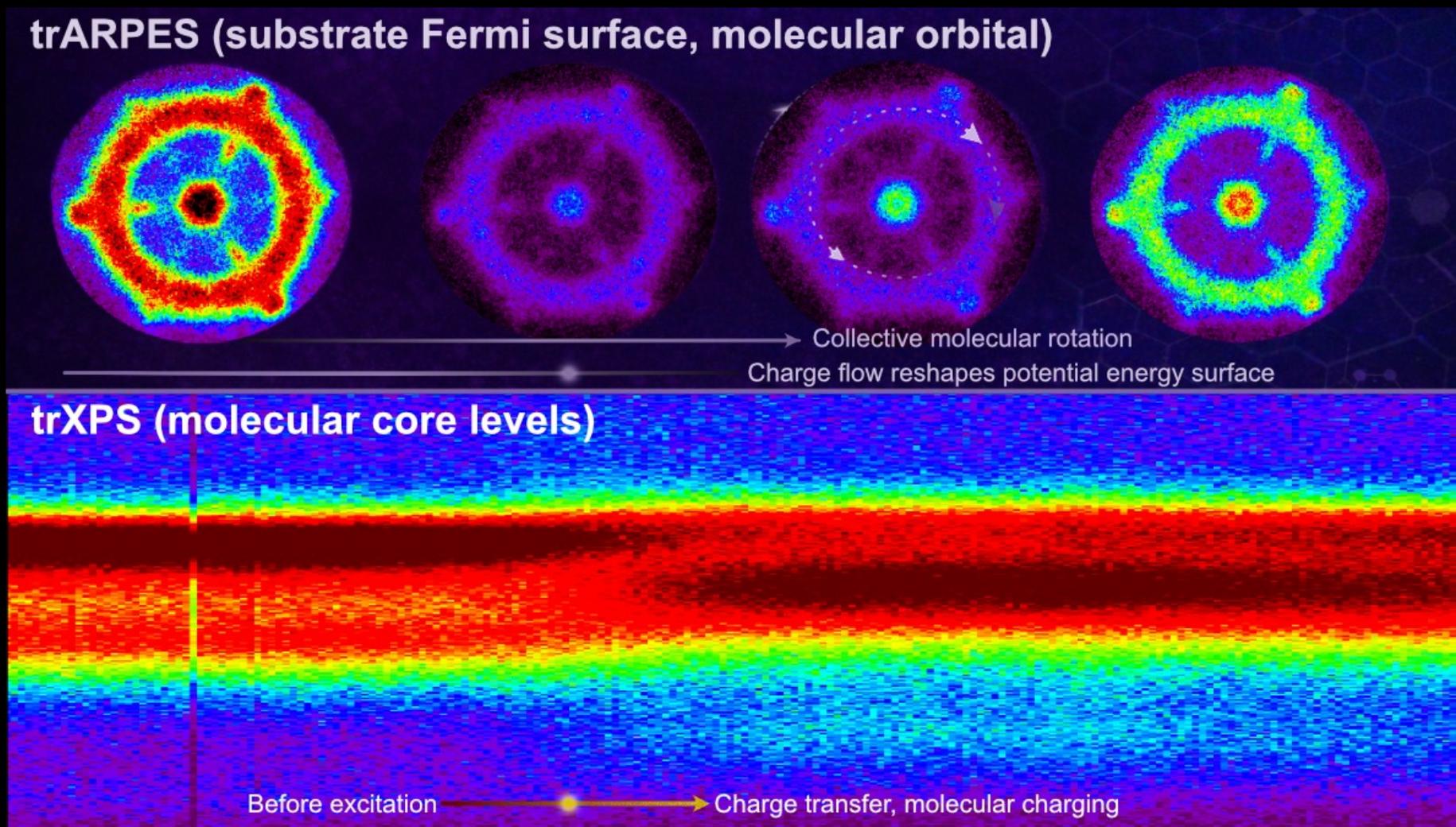
# Ultrafast spin-filter NanoESCA is the perfect complement

Ultrafast nanoscale imaging of topological edge-state dynamics in  $1T'$ -MoTe<sub>2</sub>



# Ultrafast k-microscopy advances correlative dynamic analytics

Direct photoelectric recording of quantum valence-*cum*-core electron movies

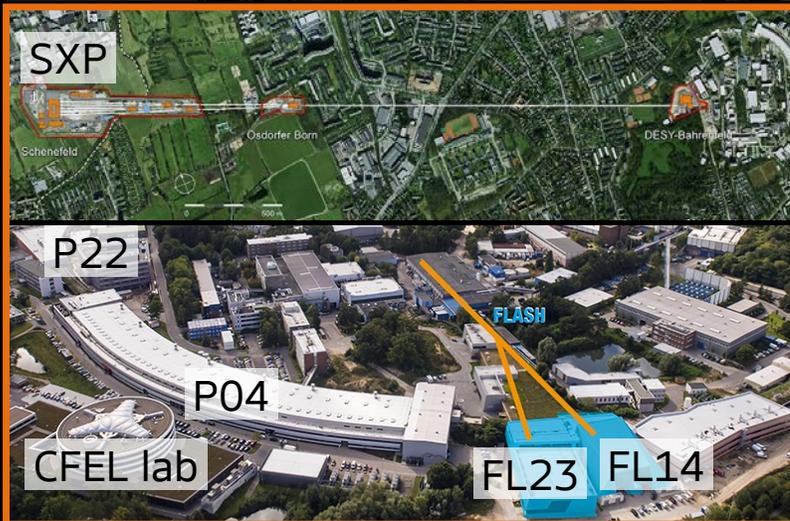
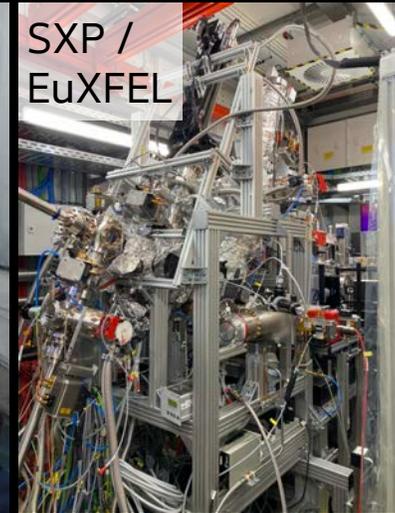
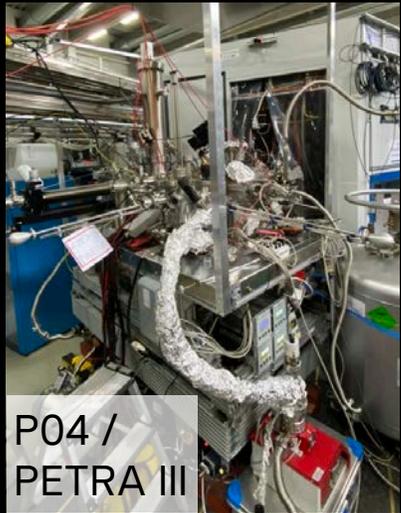


Courtesy of  
Markus Scholz

Measured at  
FL14/FLASH1

# (Ultrafast) k-microscopy is an integrative synergy engine

We are the premier k-microscopy hub, bringing together sources, instruments, and the global community.



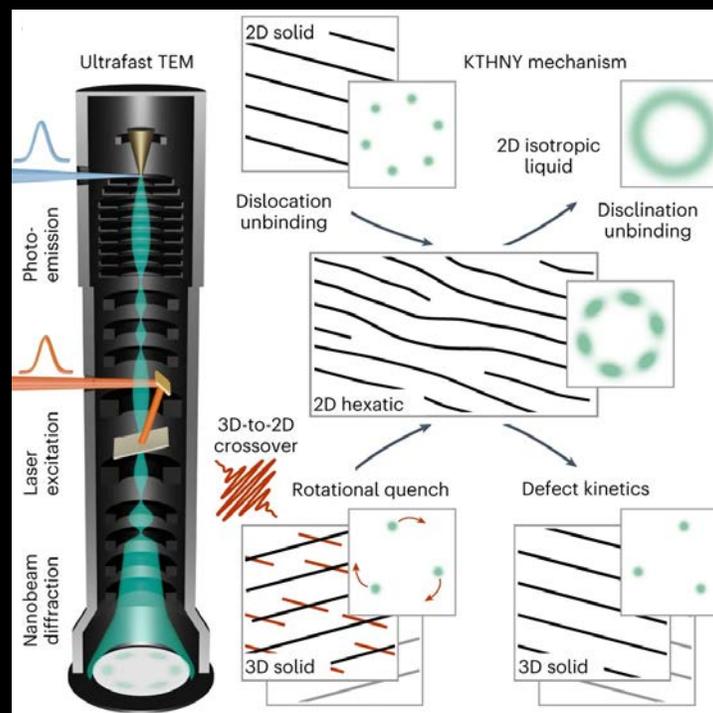
# We love to collaborate on TMDCs

Want one? Send an email to [kai.rossnagel@desy.de](mailto:kai.rossnagel@desy.de)



## Recent novel state

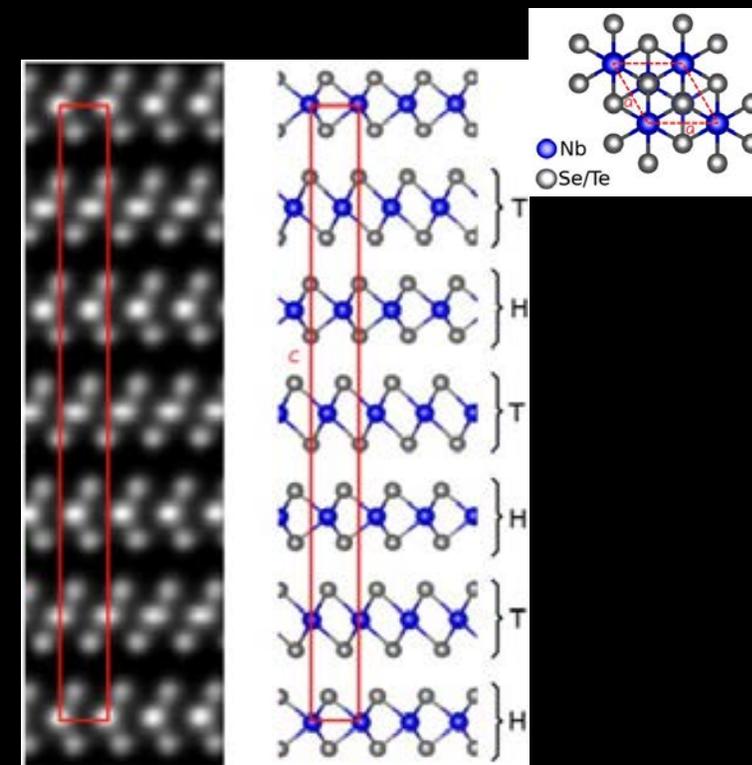
Transient 2D hexatic state in 1T-TaS<sub>2</sub>  
(with Ropers group, Göttingen)



Domröse *et al.*, Nat. Mater. **22**, 1345 (2023)

## Recent novel material

Self-stacked 6R-NbSeTe heterostructure  
(with Blanco-Canosa group, San Sebastian)



Mahatha *et al.*, ACS Nano **18**, 21052 (2024)