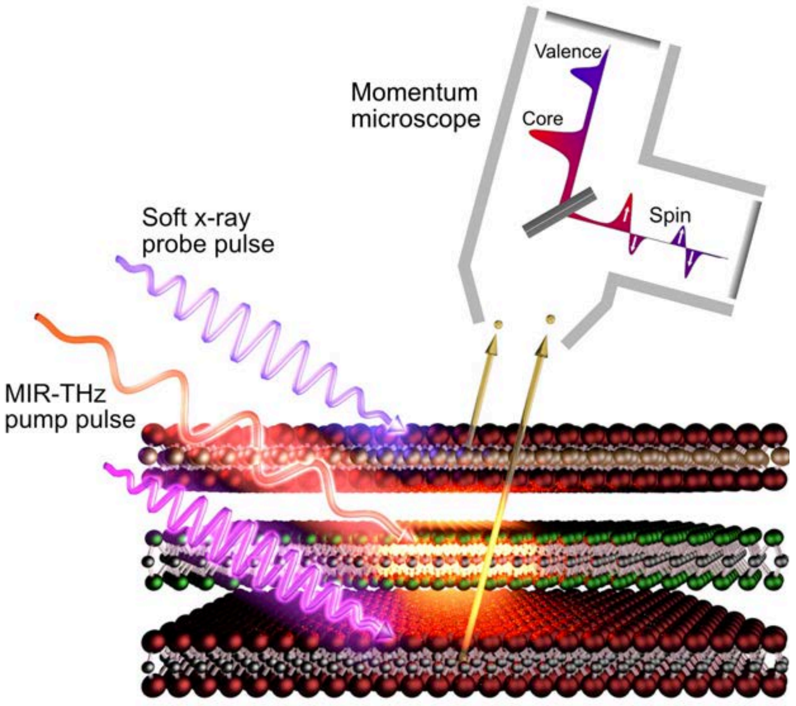
Complete ultrafast PES @ SXP/EuXFEL



- Science cases
- Instrumentation
- First experiments
- Future direction

Kai Rossnagel (on behalf of the **TR-XPES User Consortium** and the **XFEL-k-Spin-multi-D Project Team**)
Ruprecht Haensel Laboratory
Kiel Nano, Surface and Interface Science KiNSIS
Kiel University
Deutsches Elektronen-Synchrotron DESY

HELMHOLTZ RESEARCH FOR GRAND CHALLENGES

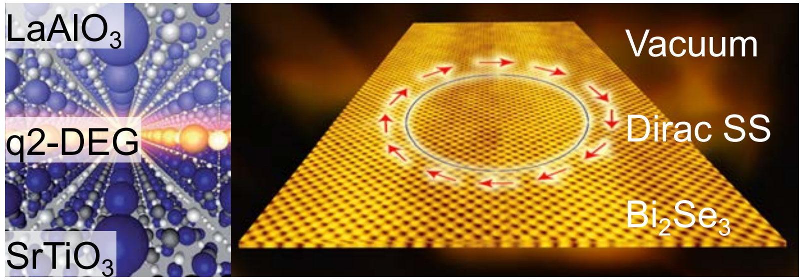
KIEL NANO SURFACE & INTERFACE SCIENCE

CAU Kiel University Christian-Albrechts-Universität zu Kiel

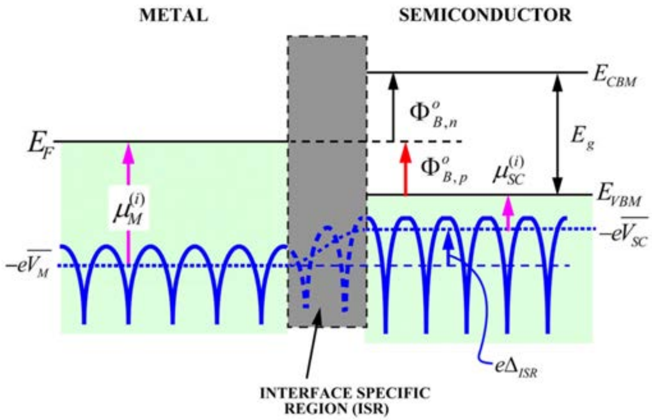
DESY.

Function by interfaces ("1 + 1 > 2")

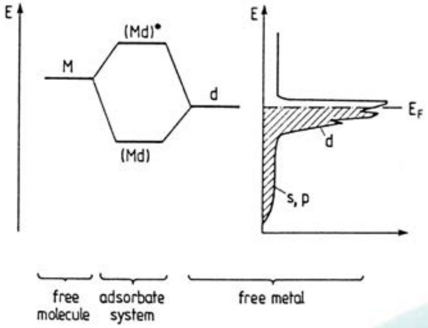
"The interface is (still) a new material, the device, and the catalyst."



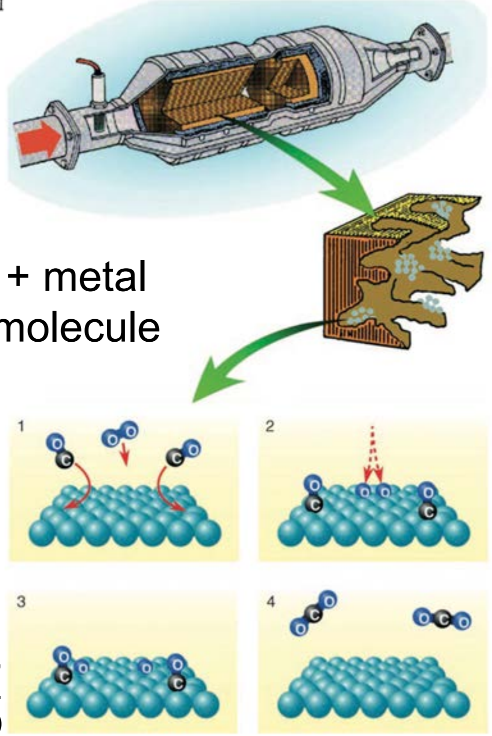
Insulator + insulator
= interface metal (superconductor)



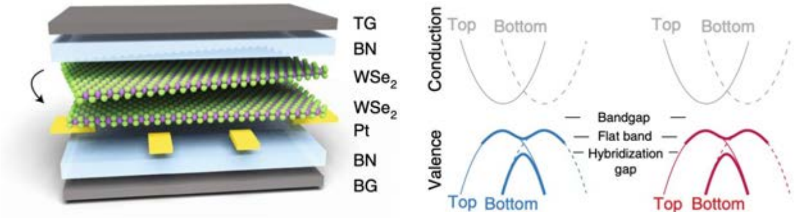
Metal + semiconductor = diode



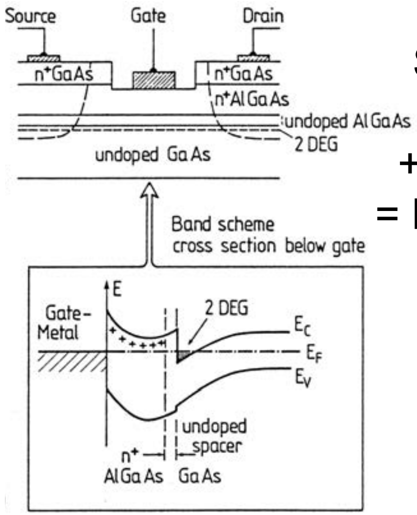
Molecule + metal
= hybrid system



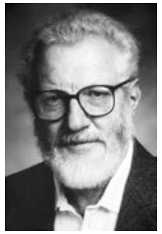
Wang *et al.*, Nat. Mater. **19**, 861 (2020)



2D semiconductor + 2D semiconductor
= correlated insulator (superconductor?)



Semiconductor
+ spacer
+ semiconductor
= high-μ_e transistor



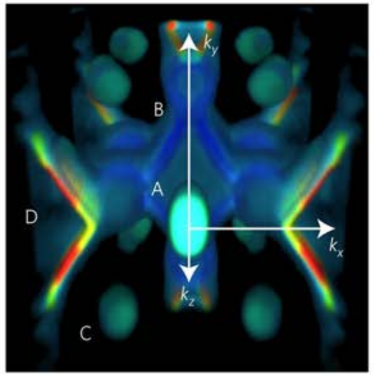
Molecules + metal
= different molecule

Ertl,
Angew. Chem. Int. Ed.
47, 3524 (2008)

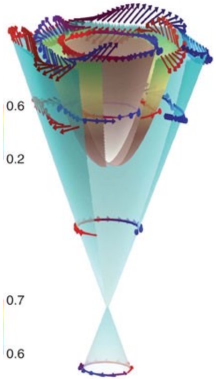
Function via dynamics

“If you want to understand function, study structure” … and dynamics!

Band structure



Spin structure



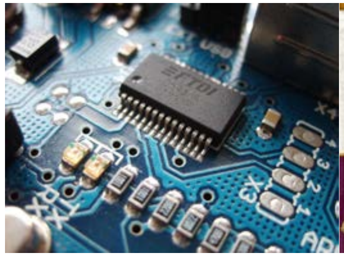
Electron hopping

$$\tau_e = \frac{h}{W} = \mathcal{O}\left(\frac{h}{1\,\mathrm{eV}}\right) = \mathcal{O}(4\,\mathrm{fs})$$

Exchange interaction

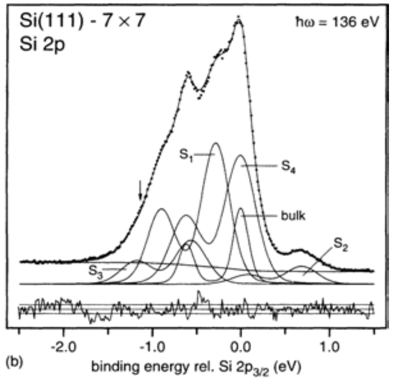
$$\tau_{\mathrm{spin}} = \frac{h}{J_{\mathrm{ex}}} = \mathcal{O}\left(\frac{h}{100\,\mathrm{meV}}\right) = \mathcal{O}(40\,\mathrm{fs})$$

Electronics

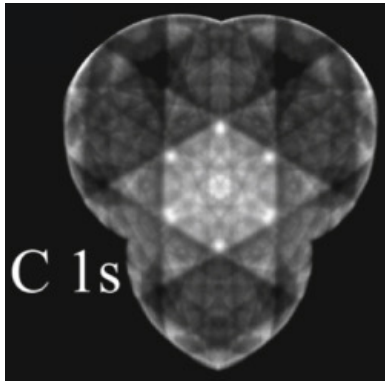


Spintronics

STRUCTURE — DYNAMICS (Electrons at interfaces!) — FUNCTION



Chemical structure



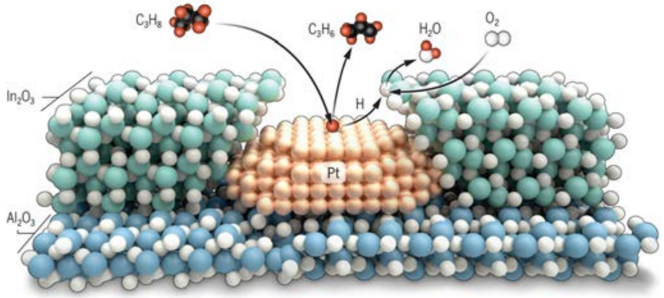
Lattice structure

Charge transfer

$$\tau_{\mathrm{CT}} = \mathcal{O}(\tau_e) = \mathcal{O}(\tau_{\mathrm{core}}) = \mathcal{O}(4\,\mathrm{fs})$$

Lattice vibration

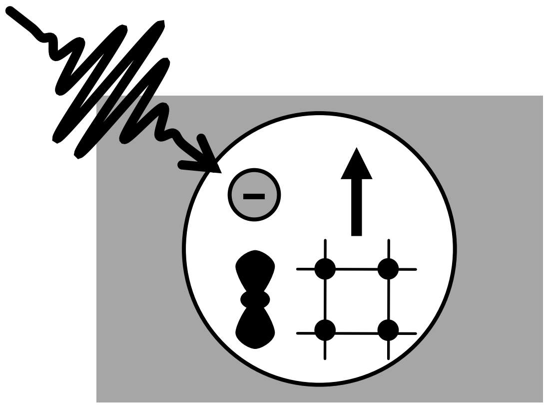
$$\tau_{\mathrm{ph}} = \frac{h}{E_{\mathrm{ph}}} = \mathcal{O}\left(\frac{h}{10\,\mathrm{meV}}\right) = \mathcal{O}(400\,\mathrm{fs})$$



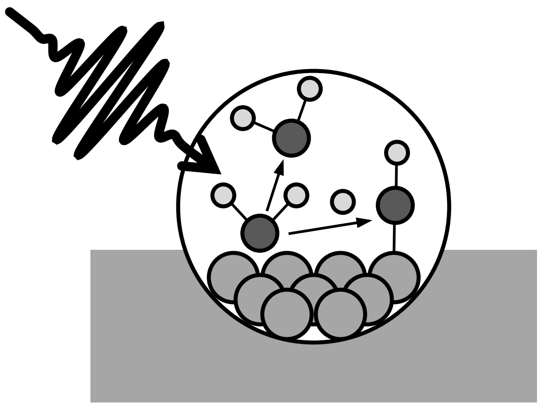
Catalysis

Science cases

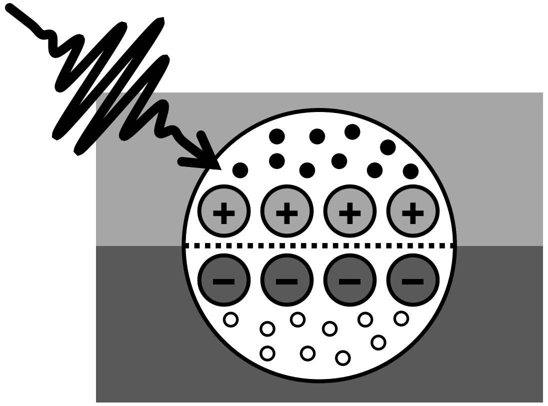
Dynamics of electronic, magnetic, chemical, and geometric structure in materials and at interfaces



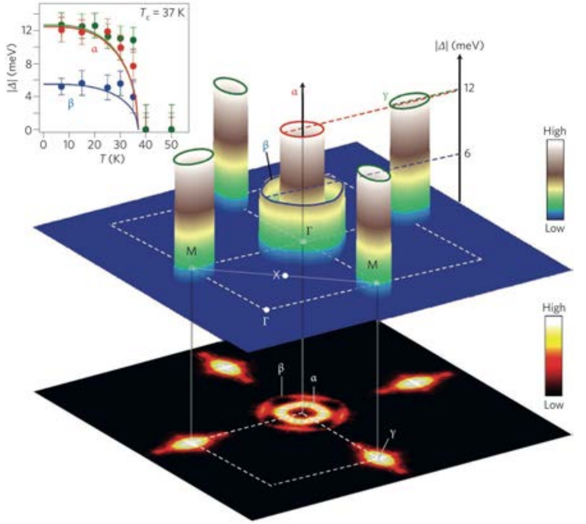
Quantum materials



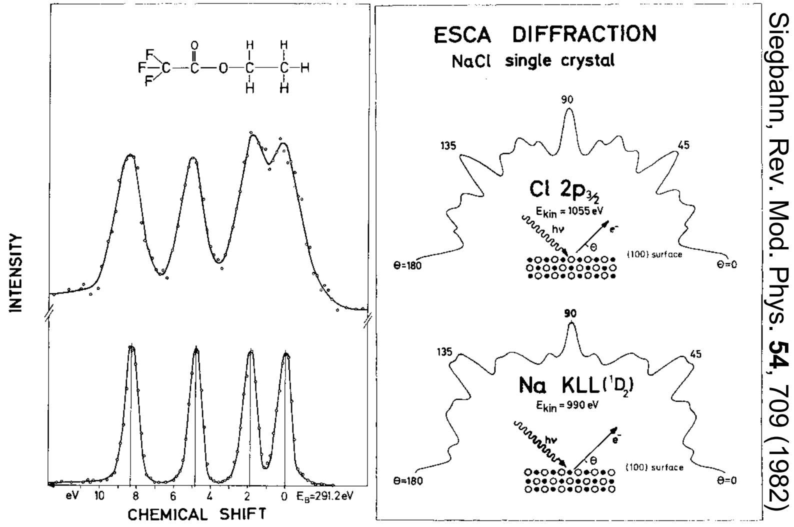
Surface chemistry



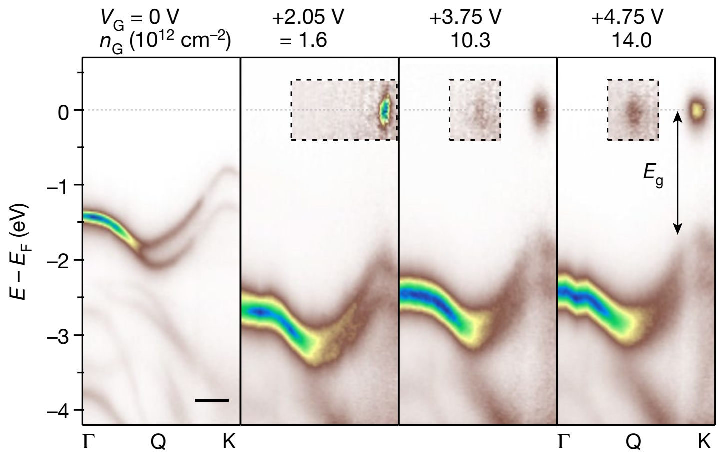
Electronic devices



Gedik & Vishik, Nat. Phys. **13**, 1029 (2017)



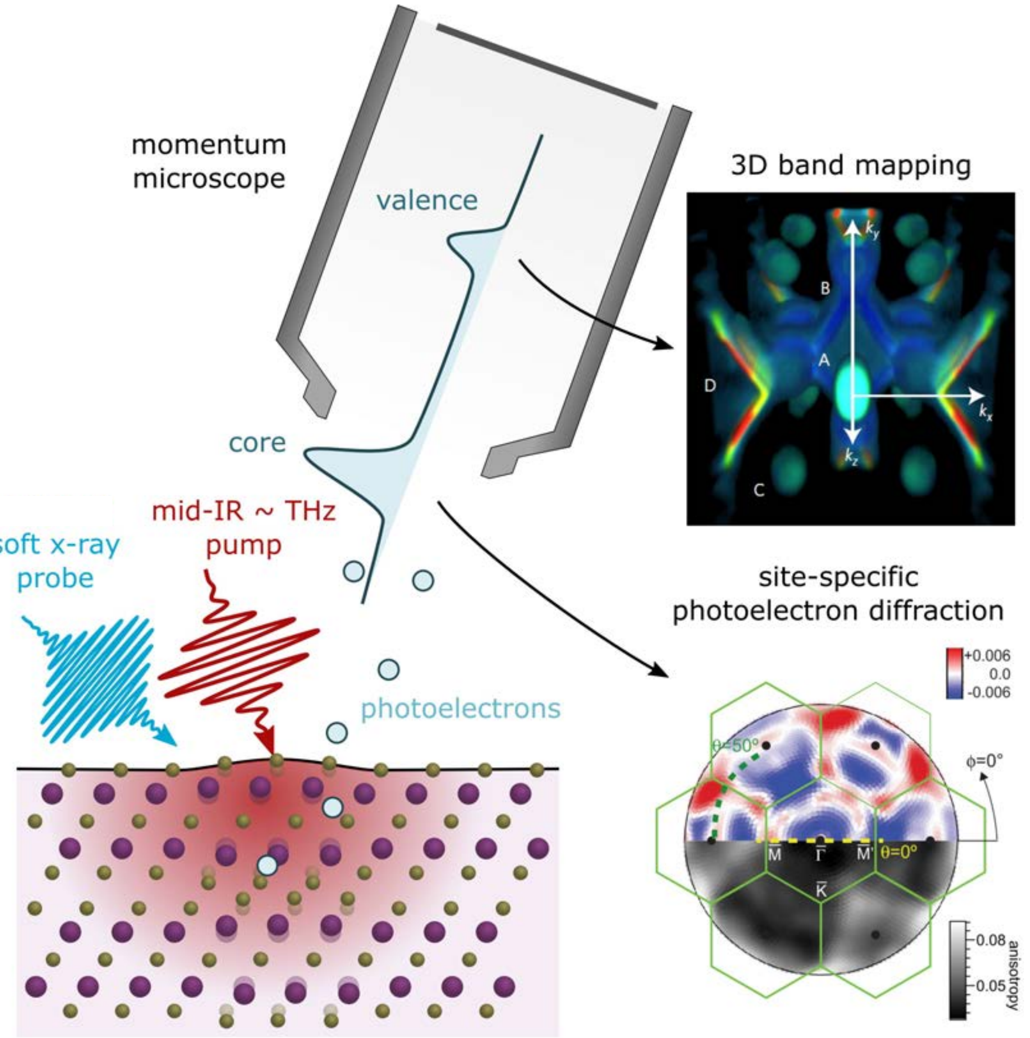
Siegbahn, Rev. Mod. Phys. **54**, 709 (1982)



Nguyen *et al.*, Nature **572**, 220 (2019)

The technique

Complete time-resolved soft x-ray photoelectron spectroscopy



Courtesy of J. Sobota

femtosecond time resolution (via pump–probe)

+

tunable MIR–THz pump

- low-energy resonances

+

tunable soft x-ray probe

- **interface sensitivity**
- 3D momentum selectivity
- core resonances
- forward scattering

+

ultra-efficient 3D energy-momentum detection

+

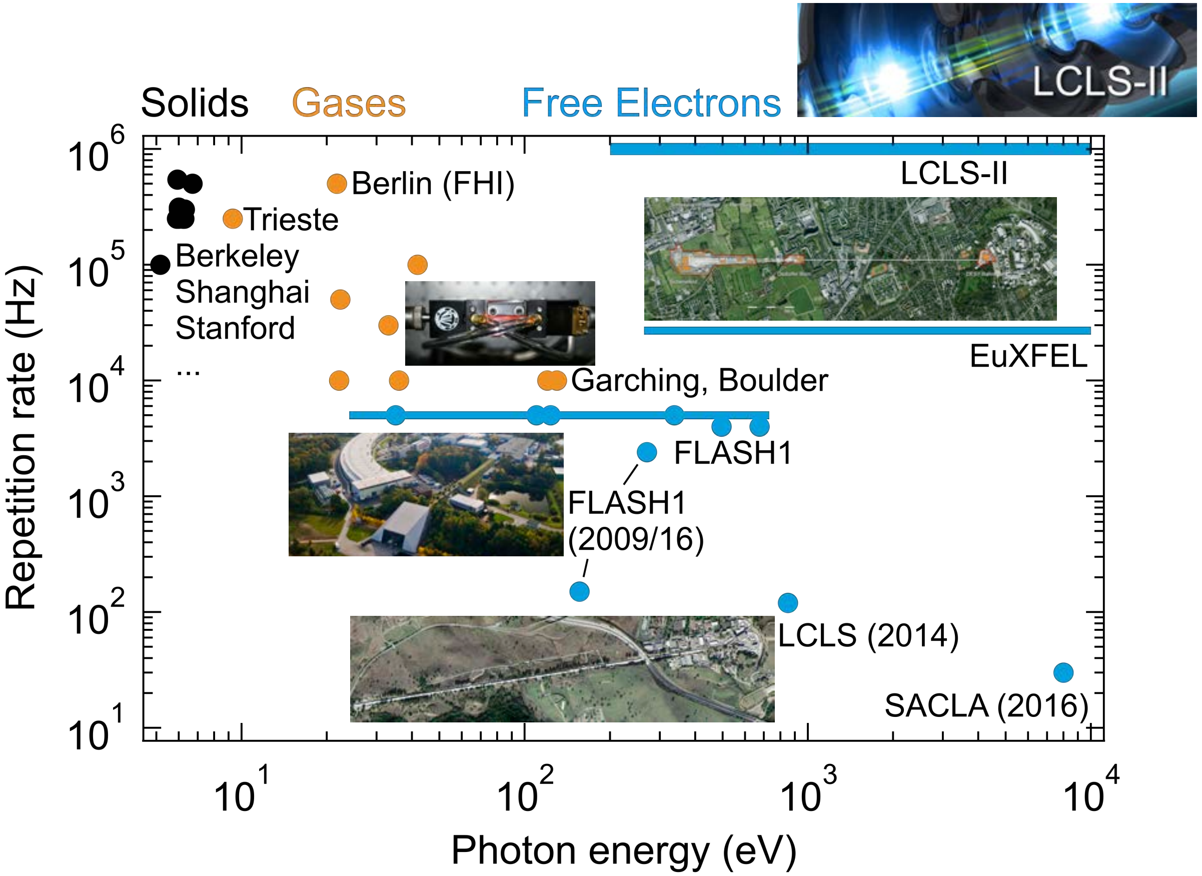
(ultra-efficient 2D spin detection)

=

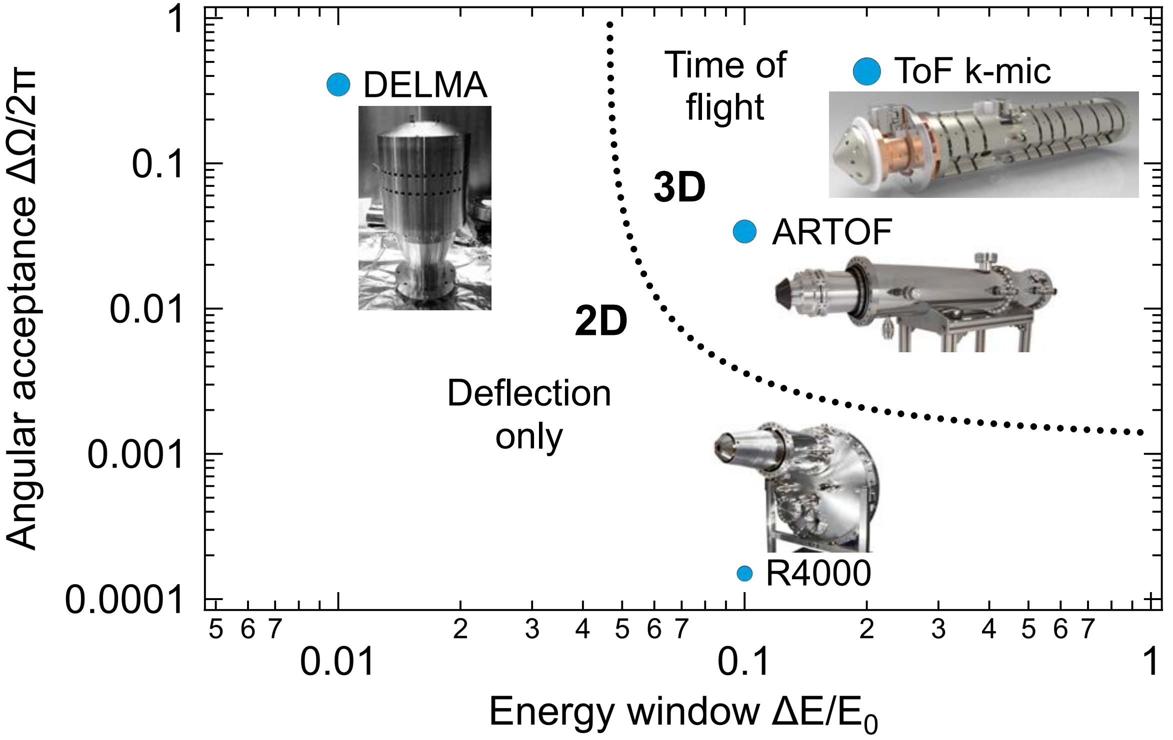
complete ultrafast
"core-*cum*-conduction(-*cum*-spin)"
photoelectron spectroscopy

The source & instrument: kHz···MHz-XFEL + ToF k-mic

Highest repetition rate of soft x-ray pulses + highest efficiency in photoelectron detection

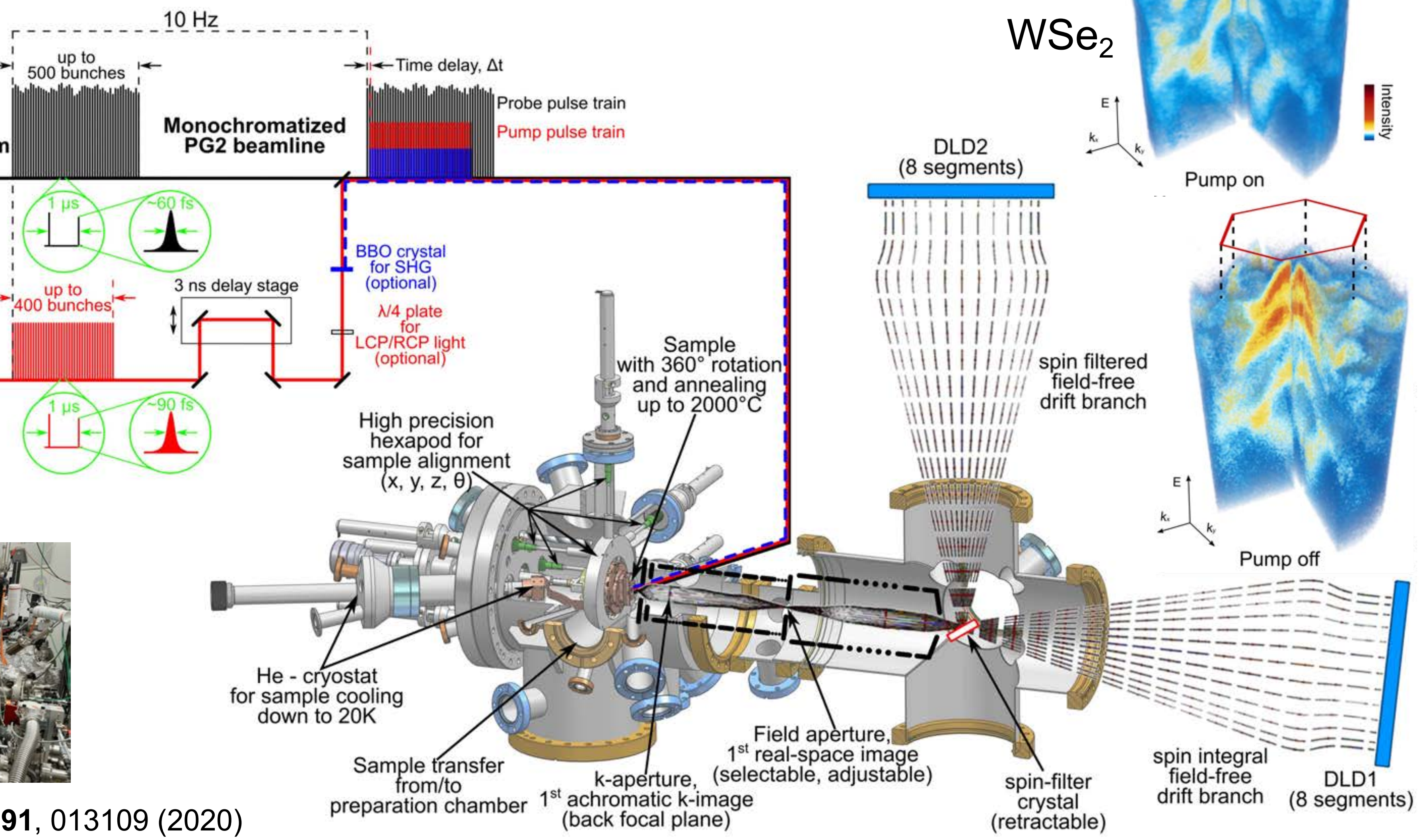


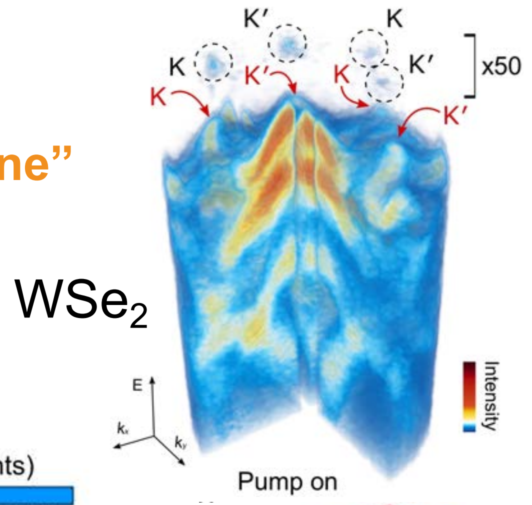
E_F 4*f* C 1*s*
O 1*s*
2*p* resonances



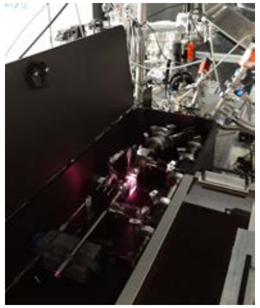
HEXTOF @ PG2 / FLASH

“The momentum microscopy (ARPES) and photoelectron diffraction (XPD) machine”





HHG lab @ CFEL / DESY

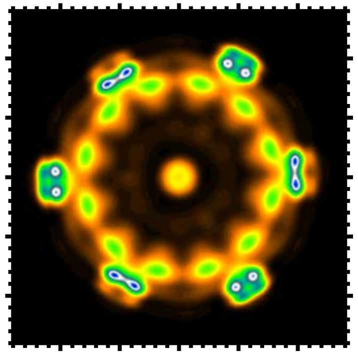


Kutnyakhov *et al.*, Rev. Sci. Instrum. **91**, 013109 (2020)

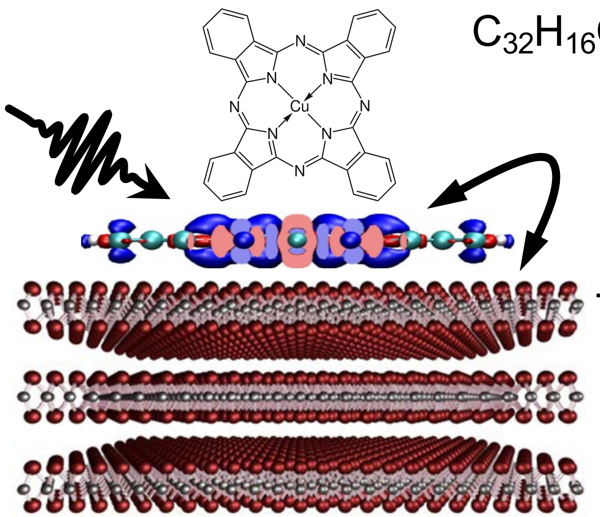
CuPc / $TiSe_2$: “E_F” + C 1*s*

Spectral, angular, and temporal dissection of intertwined charge, orbital, and structure dynamics

HOMO momentum map

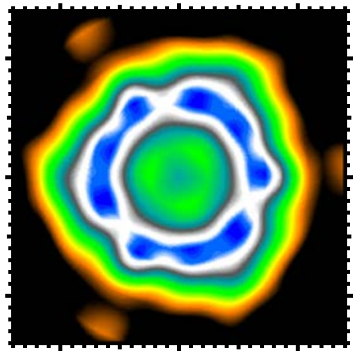


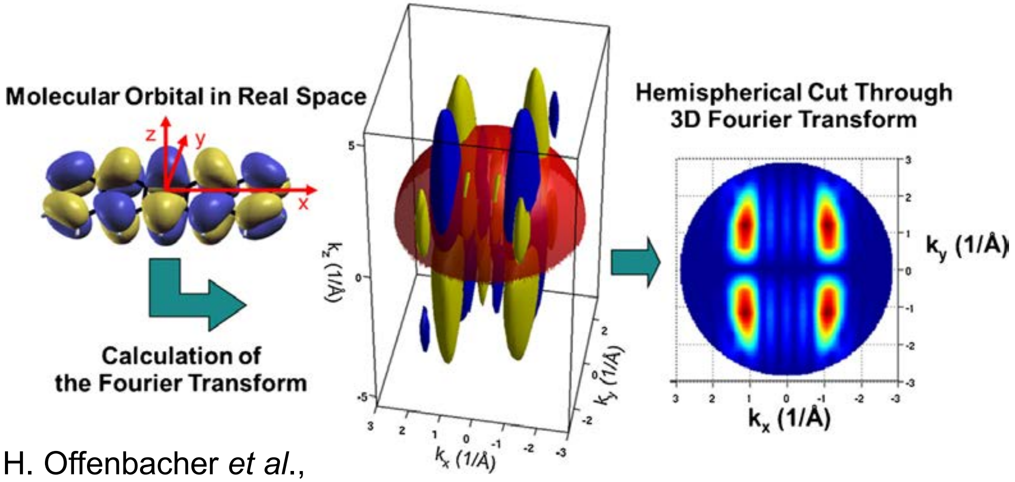
$C_{32}H_{16}CuN_8$



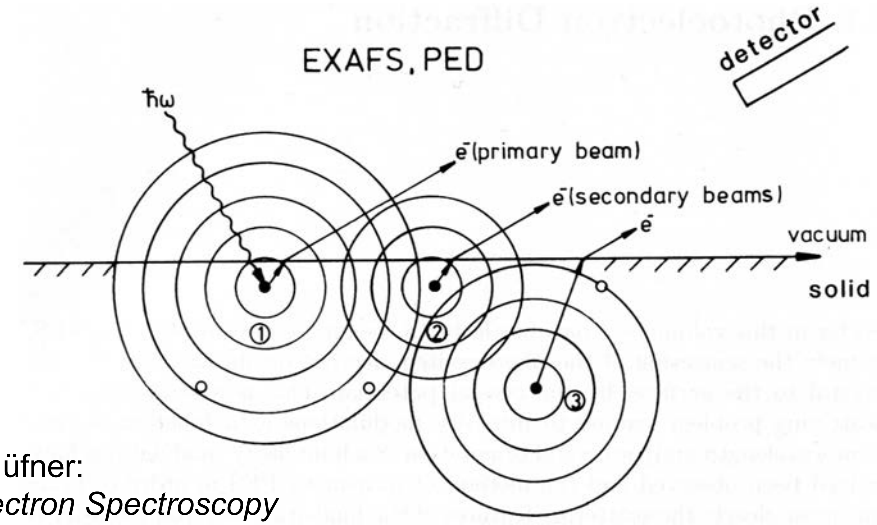
$TiSe_2$

C 1*s* momentum map





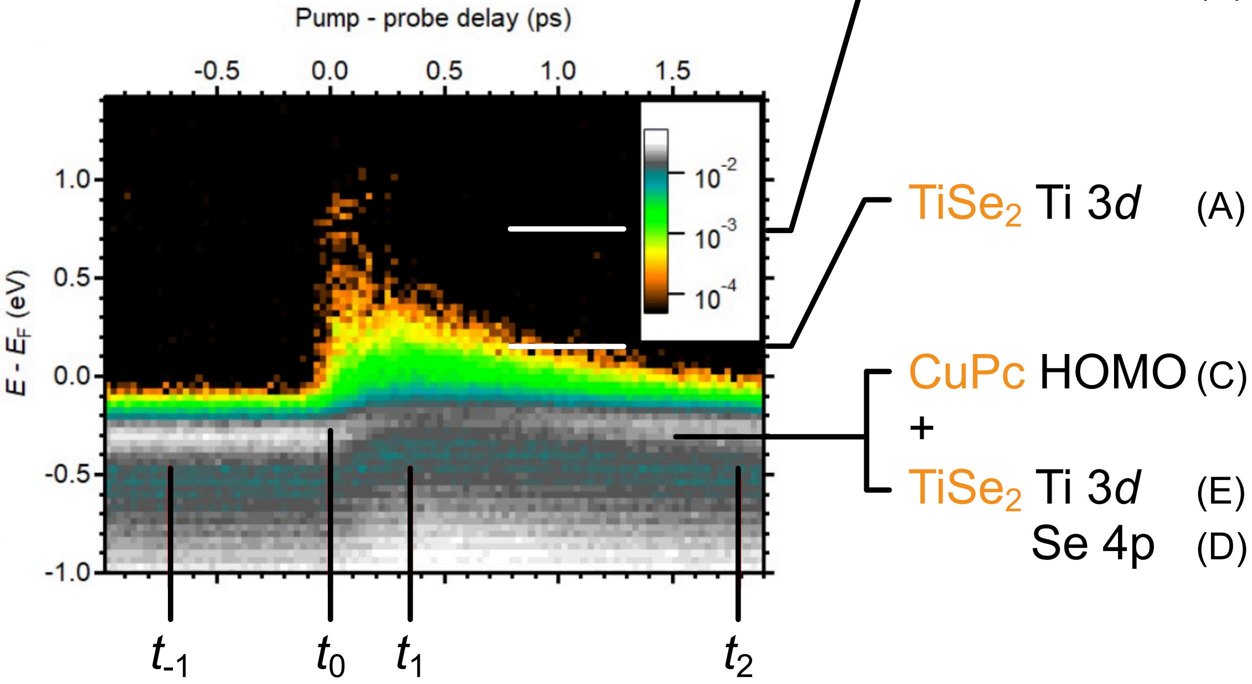
H. Offenbacher *et al.*,
J. Electron Spectrosc. Relat. Phenom. **204**, 92 (2015)



Stefan Hüfner:
Photoelectron Spectroscopy

CuPc / $TiSe_2$: "E_F"

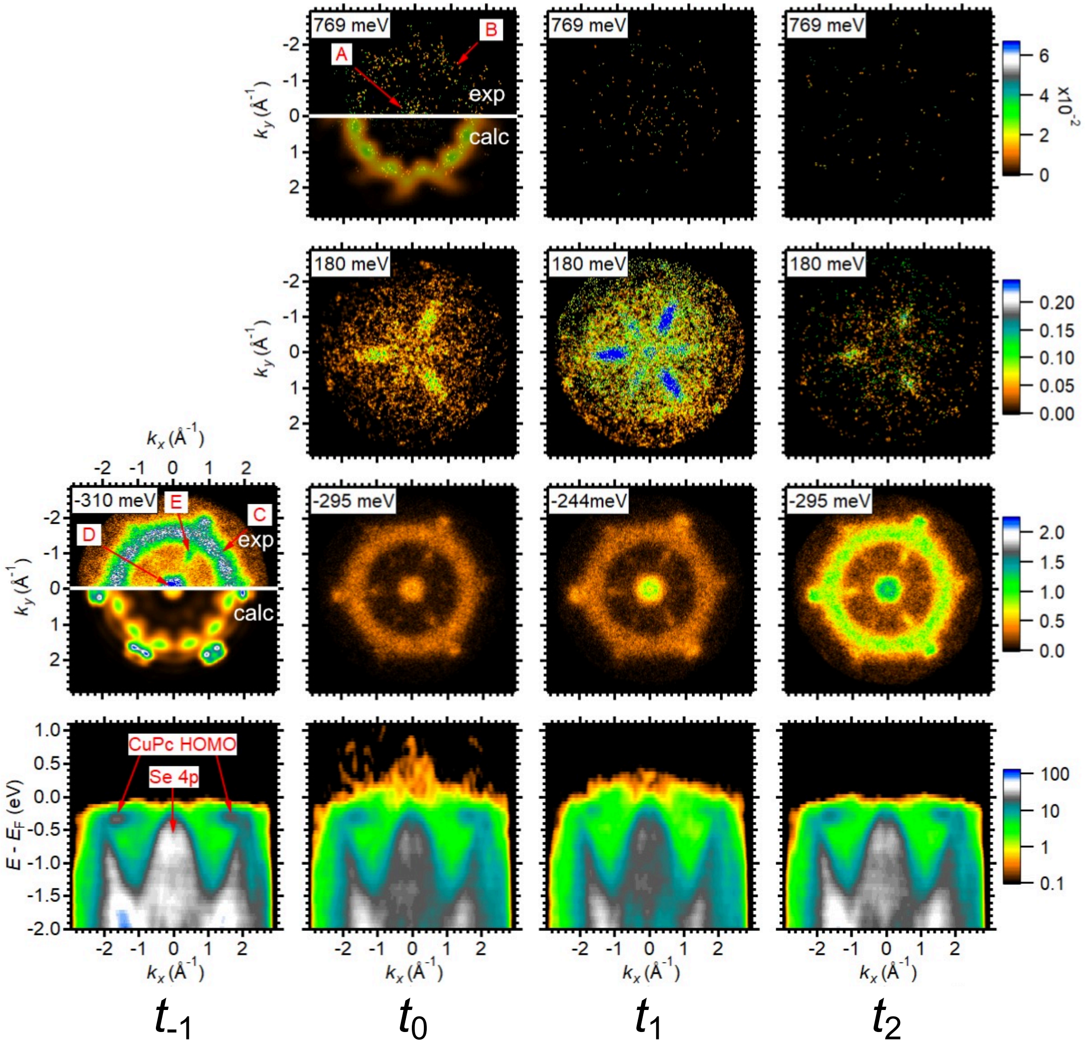
Time-resolved ARPES (k-microscopy)



$$h\nu_{\mathrm{probe}} = 36.3\,\mathrm{eV}$$

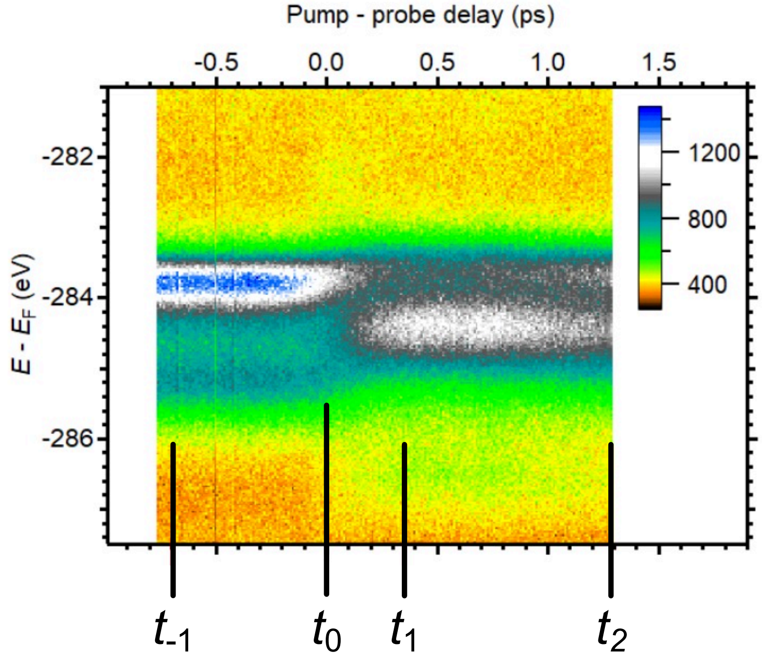
$$h\nu_{\mathrm{pump}} = 1.6\,\mathrm{eV}$$

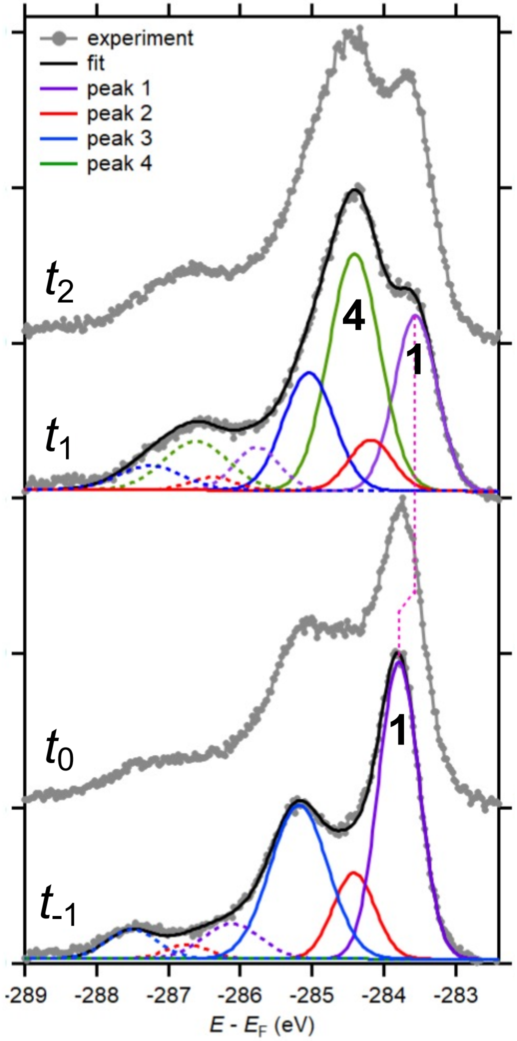
$$F \approx 3.5\,\mathrm{mJ/cm^2}$$

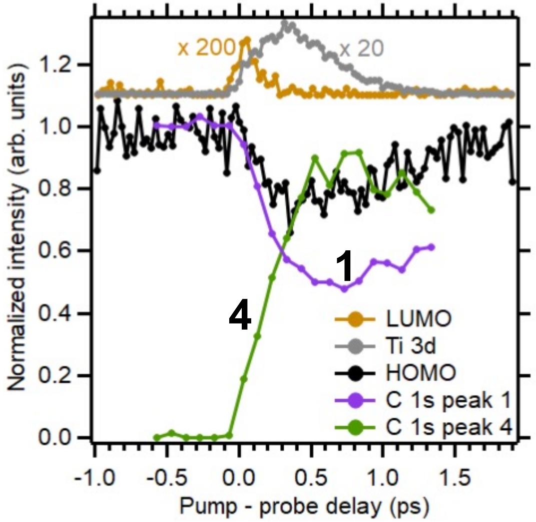


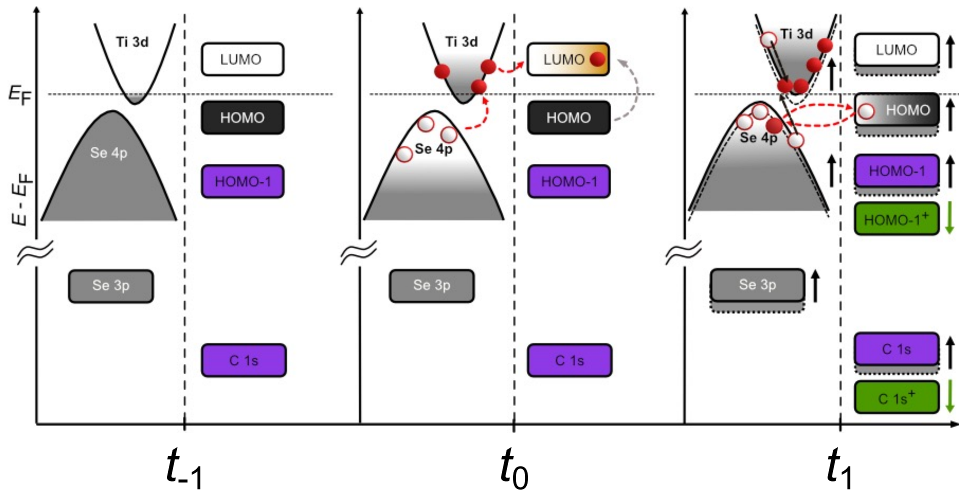
CuPc / $TiSe_2$: C 1*s*

Time-resolved XPS





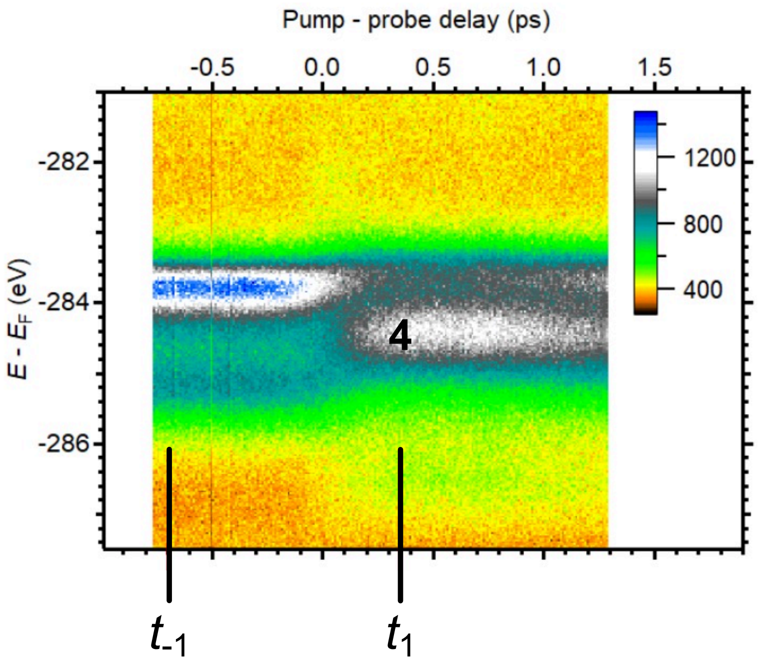


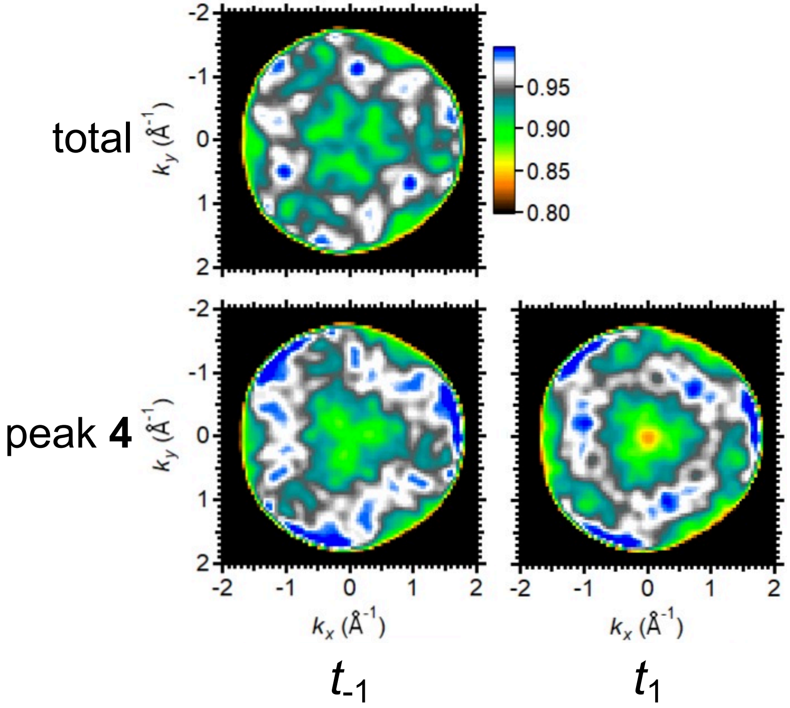


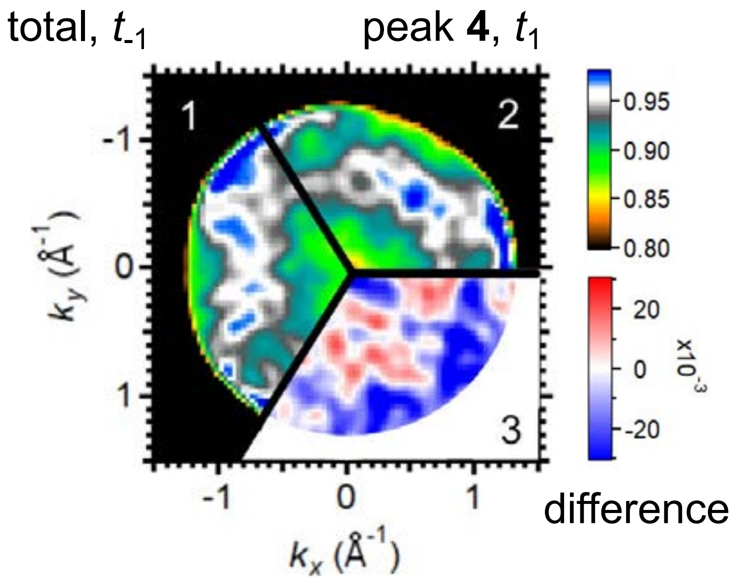
$$h\nu_{\mathrm{probe}} = 371\,\mathrm{eV}$$
$$h\nu_{\mathrm{pump}} = 1.6\,\mathrm{eV}$$
$$F \approx 3.5\,\mathrm{mJ/cm^2}$$

CuPc / $TiSe_2$: C 1*s*

Time-resolved XPS *cum* XPD



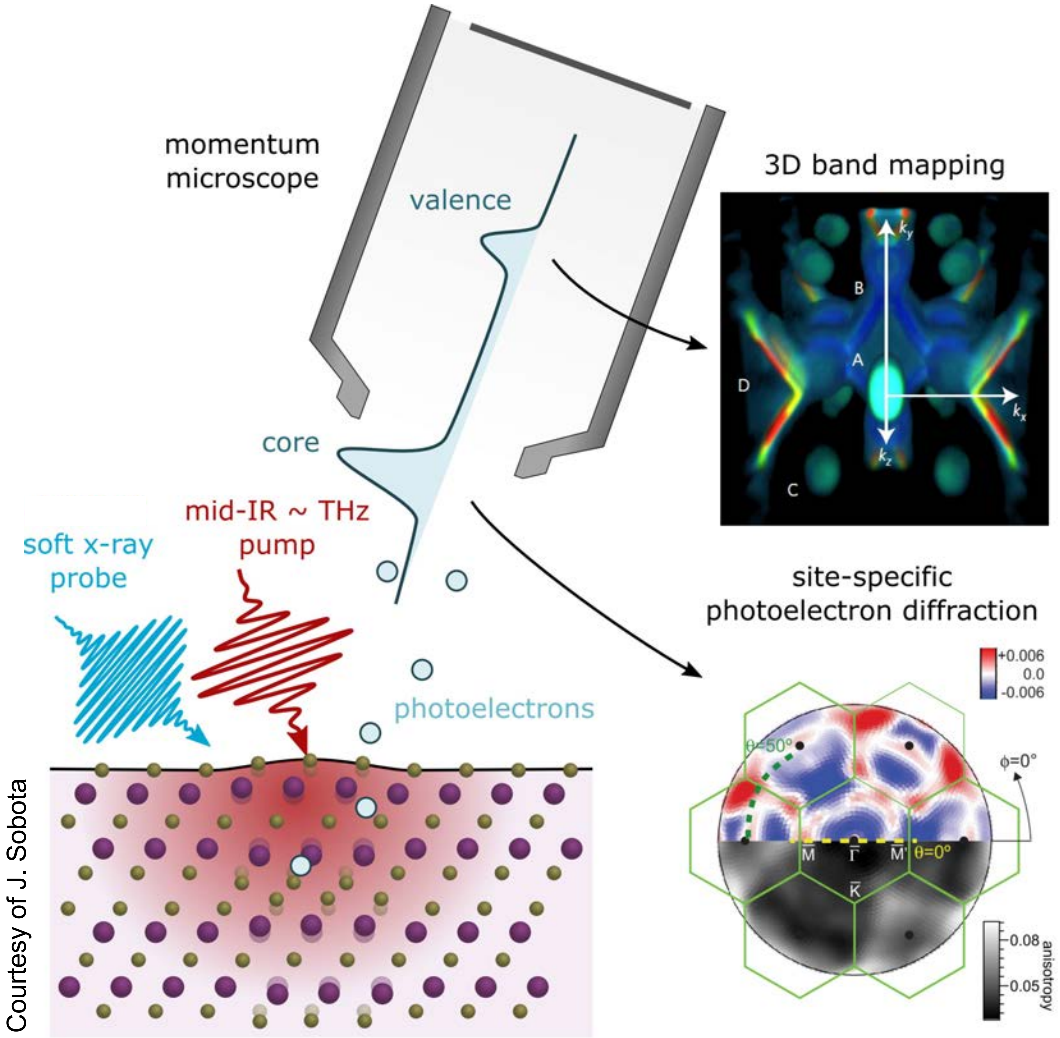




$$h\nu_{\text{probe}} = 371\,\text{eV}$$
$$h\nu_{\text{pump}} = 1.6\,\text{eV}$$
$$F \approx 3.5\,\text{mJ/cm}^2$$

ToF k-mic @ SXP/EuXFEL

Complete time-resolved soft x-ray core-*cum*-conduction(-*cum*-spin) photoelectron spectroscopy



HHG	PG2/FLASH
$\leq 500\,\mathrm{kHz}$	$5\,\mathrm{kHz}$
$\lesssim 100\,\mathrm{eV}$	$24\cdots730\,\mathrm{eV}$
trARPES	**trXPS** trARPES trXPD

SXP/EuXFEL	NEH 2.2/LCLS-II
$27/3\,\mathrm{kHz}$	$1\,\mathrm{MHz}$
$\gtrsim 700\,\mathrm{eV}$	$250\cdots1600\,\mathrm{eV}$
trXPD trXPS trARPES	**trARPES** trXPD trXPS

Contributions to SXP ("Open Port")

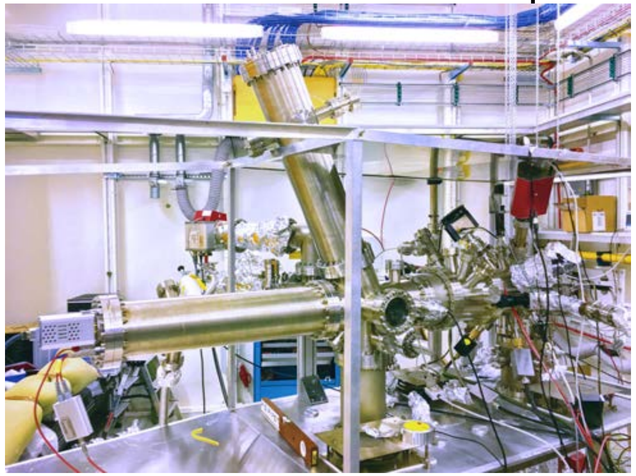
Total BMBF funding (Hamburg, Mainz, Kiel, Duisburg, 2013–2022): 5.1 MEUR

Universität Hamburg
DER FORSCHUNG | DER LEHRE | DER BILDUNG

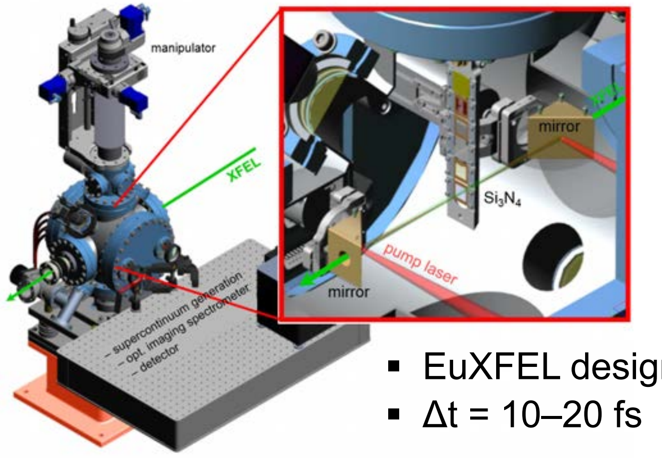
Kiel University
Christian-Albrechts-Universität zu Kiel

JOHANNES GUTENBERG UNIVERSITÄT MAINZ

ToF momentum microscope

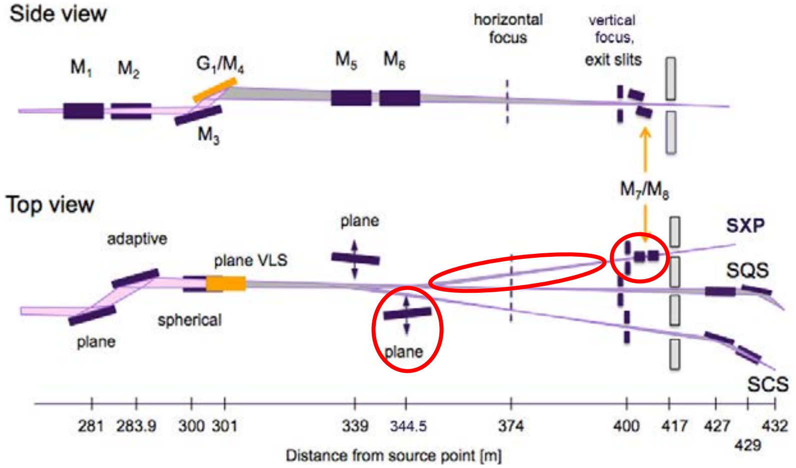


FEL pulse arrival time monitor

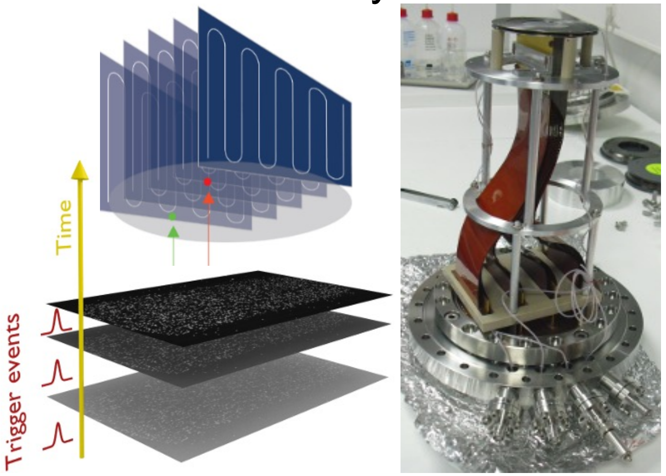


- EuXFEL design
- Δt = 10–20 fs

X-ray optics

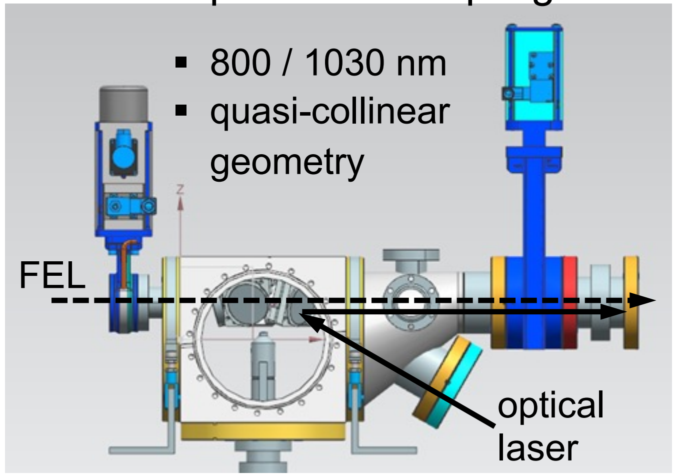


Multi-channel delay-line detector

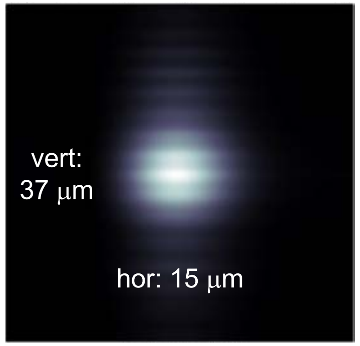


Pump laser in-coupling

- 800 / 1030 nm
- quasi-collinear geometry



Wavefront propagation simulations

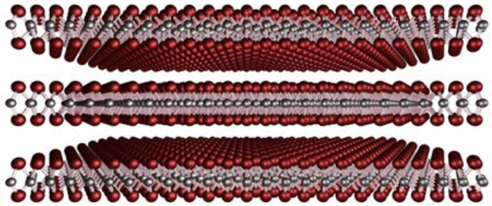


Roughness: 8.0 nm (RMS)

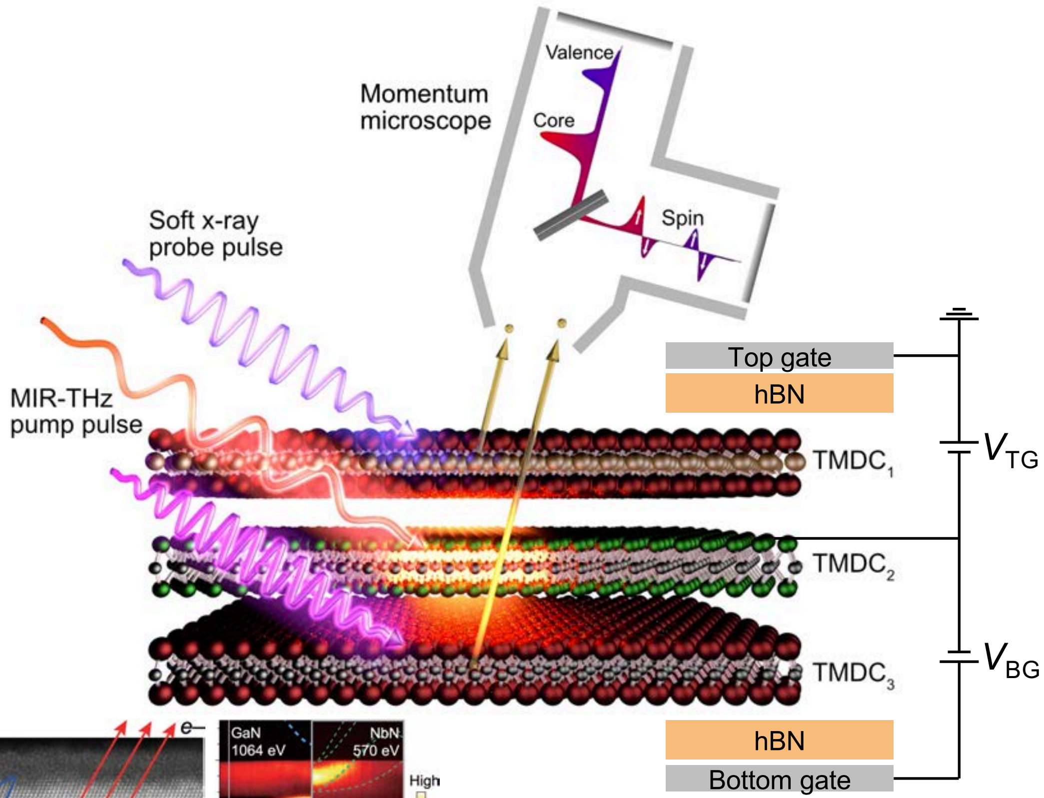
Real-time FEL pulse diagnostics

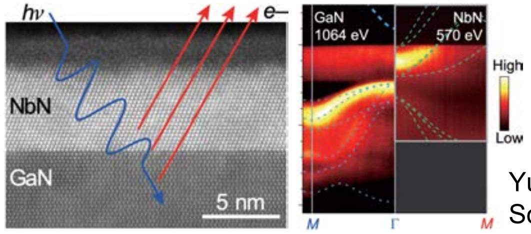
First experiments: 2D materials (+ molecules)

CAU Kiel / DESY Hamburg



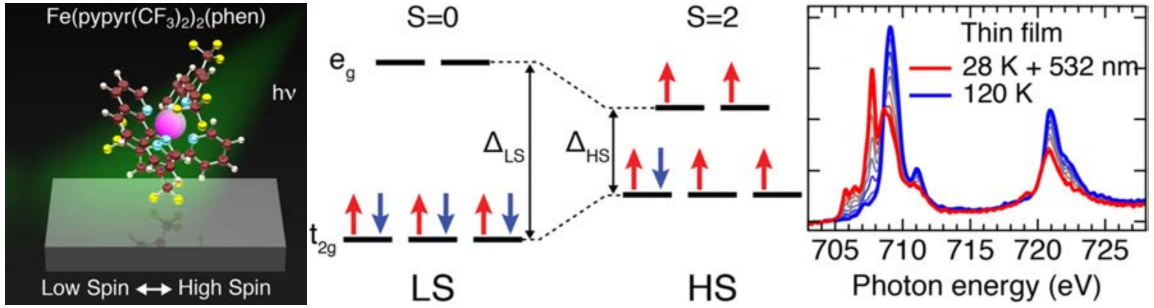
Layer-selective pump–probe in TMDC heterostructures



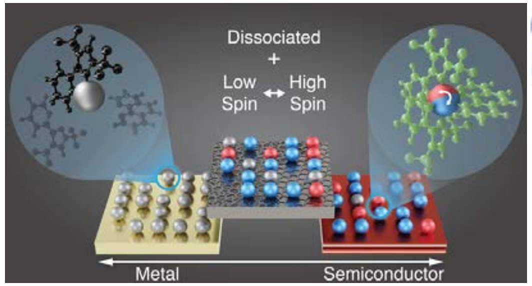


Yu *et al.*,
Sci. Adv. **7**, eabi5833 (2021)

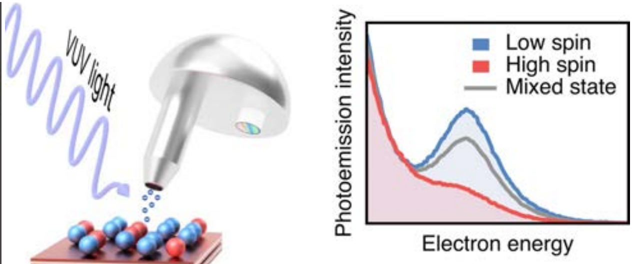
Ultrafast spincrossover on TMDC surfaces



Rohlf *et al.*,
J. Phys. Chem. Lett. **9**, 1491 (2018)



Rohlf *et al.*,
J. Phys. Chem. C **123**, 17774 (2019)

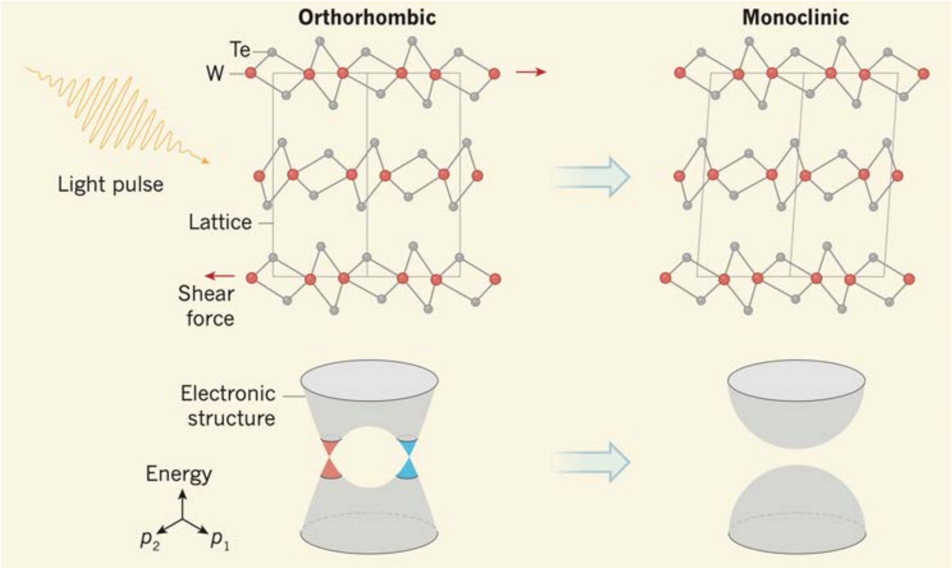


Rohlf *et al.*,
J. Phys. Chem. C **125**, 14105 (2021)

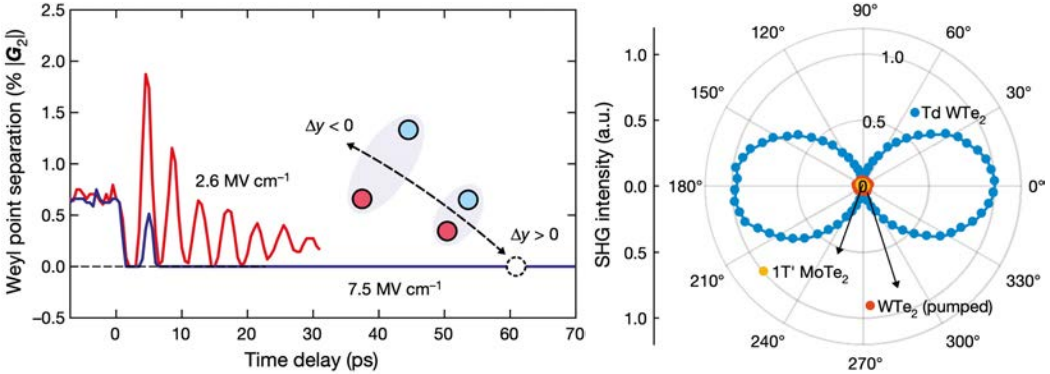
First experiments: Topological & correlated materials

KTH Stockholm / JGU Mainz

Ultrafast strain-induced topological phase transition

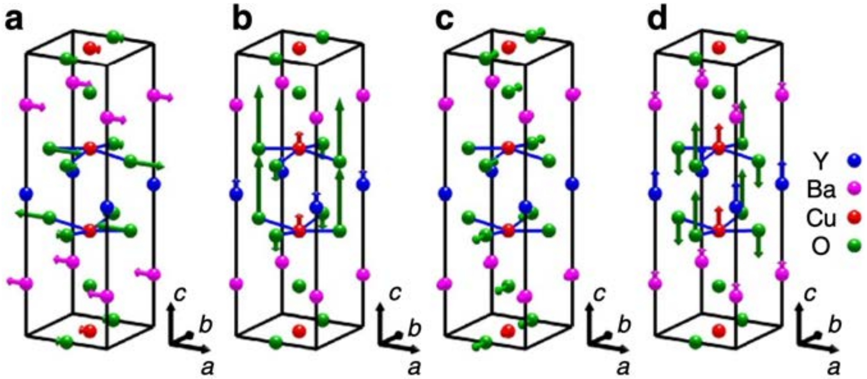


Son *et al.*, Nature **565**, 32 (2019) [News & views]

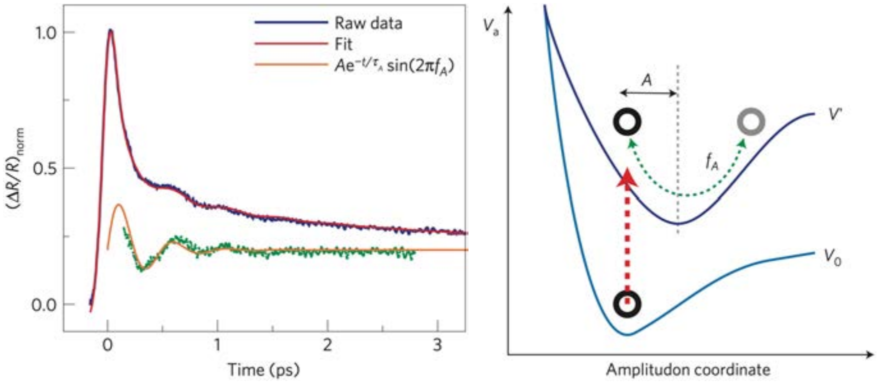


Sie *et al.*, Nature **565**, 61 (2019)

Ultrafast dynamics of coexisting electronic orders



Forgan *et al.*, Nat. Commun. **6**, 10064 (2015)



Torchinsky *et al.*, Nat. Mater. **12**, 387 (2013)

Future direction

Tracking of coherent electron dynamics in quantum materials with 5-fs soft x-ray pulses

few-100 as spectroscopy	$\tau_{\mathrm{e-e}} \approx 10\,\mathrm{fs}$	few-10 fs spectroscopy
1 fs	10 fs	100 fs
ultrahigh time resolution	$5\,\mathrm{fs} \longleftrightarrow 360\,\mathrm{meV}$	high energy resolution
Electron motion	**Coherence** Incoherence	Relaxation & thermalization

- laser-induced coherences
- Coulomb-induced correlations
- resonantly coupled excitations
- quantum-kinetic retardations

cf. Axt & Kuhn,
Femtosecond spectroscopy in semiconductors:
a key to coherences, correlations and quantum kinetics,
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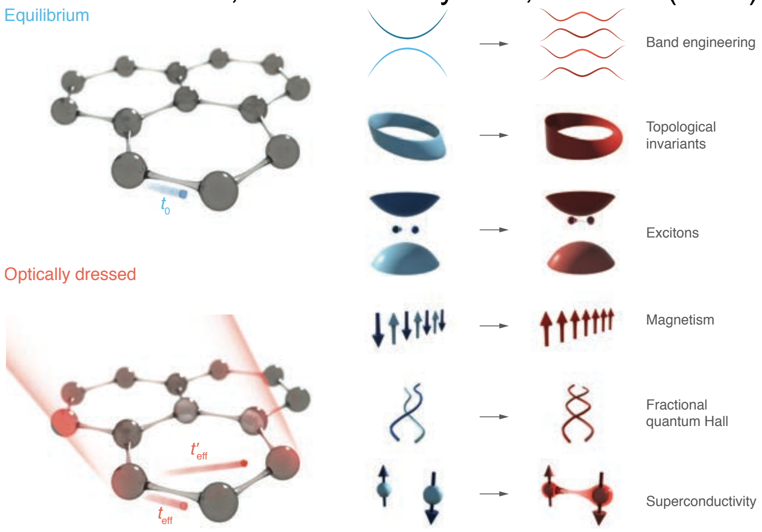

Quantum materials dynamics in real time

- observe on timescales shorter than typical interaction times
- control the outcome of interaction processes

Quantum engineering of correlated many-particle states