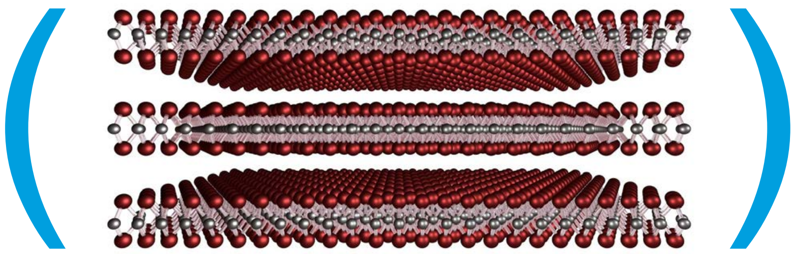
Experiments at Synchrotron & FEL Sources for QTech



Kai Rossnagel
Ruprecht Haensel Laboratory
Kiel Nano, Surface and Interface Science KiNSIS
Kiel University
Deutsches Elektronen-Synchrotron DESY

DESY. QUANTUM

RUPRECHT HAENSEL LABOR

KIEL NANO SURFACE & INTERFACE SCIENCE

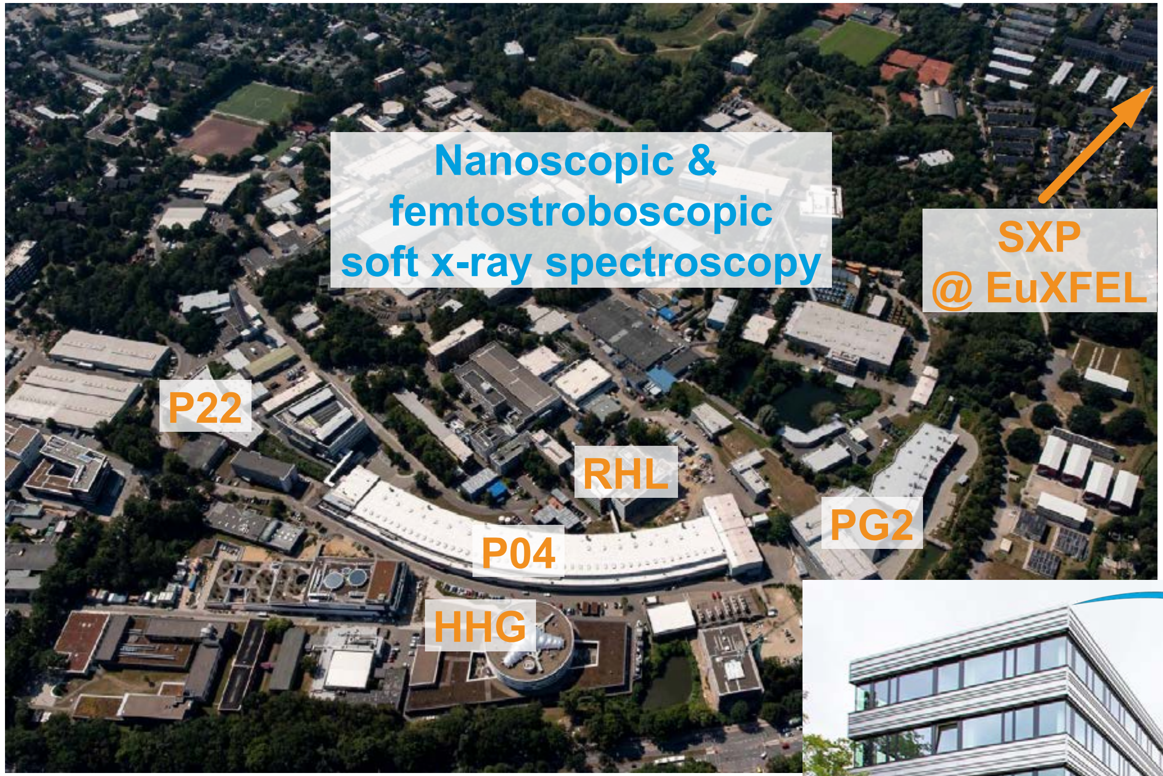
C A U Christian-Albrechts-Universität zu Kiel

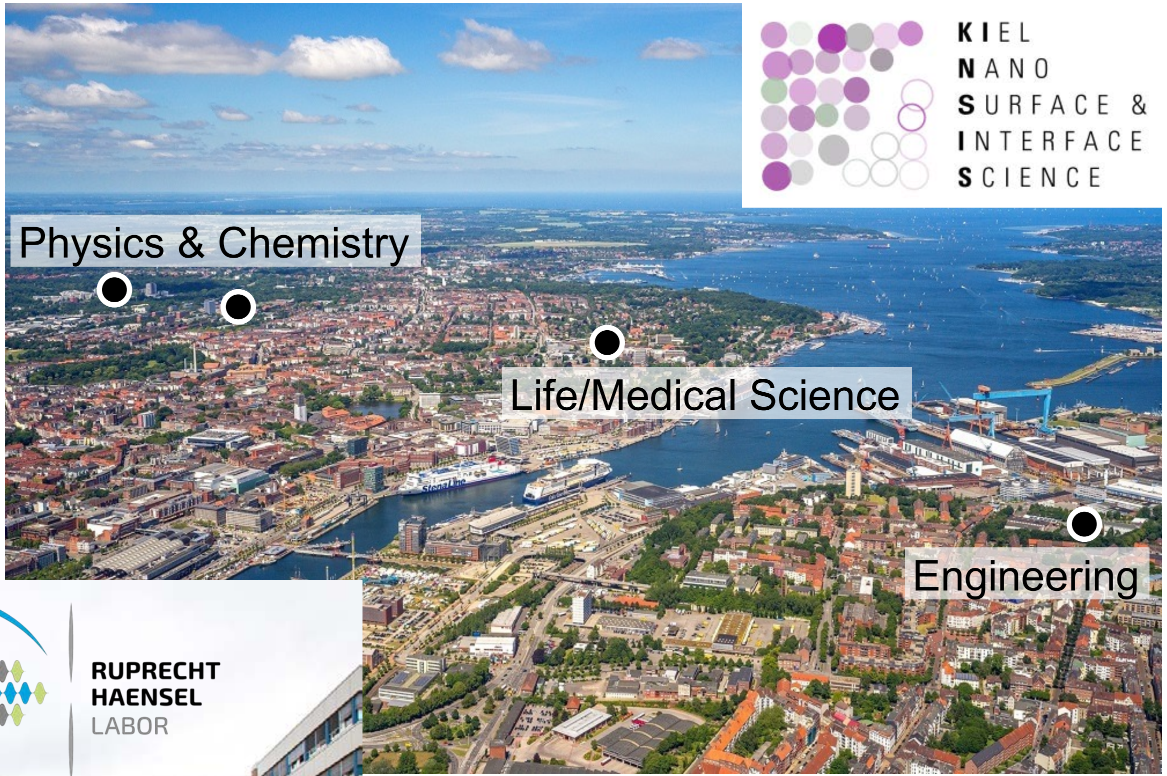
DESY.

HELMHOLTZ RESEARCH FOR GRAND CHALLENGES

Bridging DESY & CAU Kiel

Ruprecht Haensel Laboratory (RHL) / Soft X-ray Spectroscopy of Quantum Materials (SXQM)





KIEL NANO SURFACE & INTERFACE SCIENCE

DESY.

C A U
Christian-Albrechts-Universität zu Kiel

Bridging X & Q

Versatile high-precision x-ray analytics for quantum technology hardware

X

High-precision (low-throughput) x-ray methods

Cutting-edge science of novel, “exotic” quantum materials

keV photons (meV resolution)

Q

Promising (solid-state) qubit platforms

“Profane” (high-throughput) characterization of technologically relevant, conventional materials

μeV energy splittings

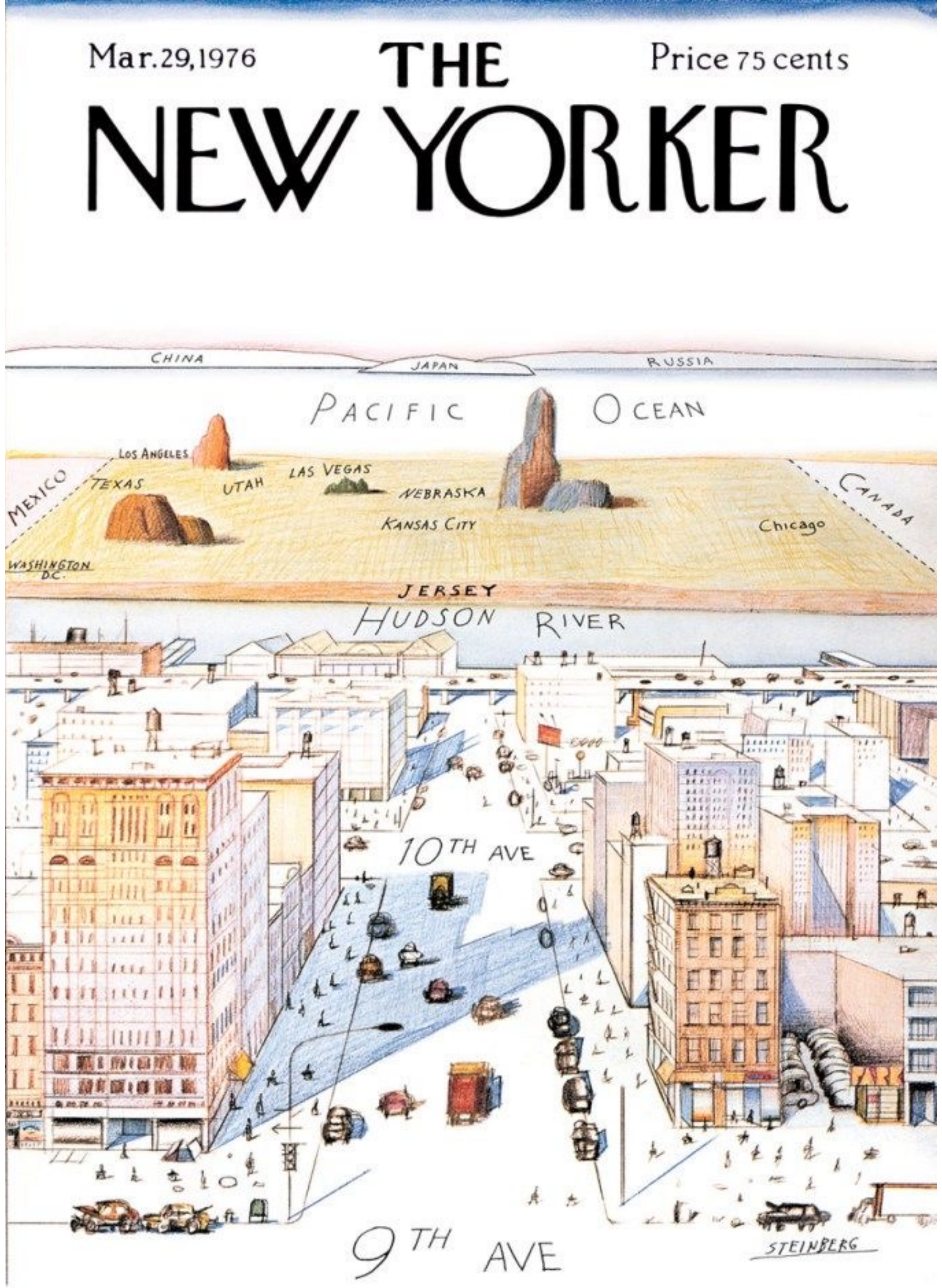
I. Synchrotron & free-electron-laser experiments — general concepts

II. Possible applications — needs of quantum technology community?

III. 2D materials — viable quantum technology platform?

Disclaimer

It's a wide field …



DESY

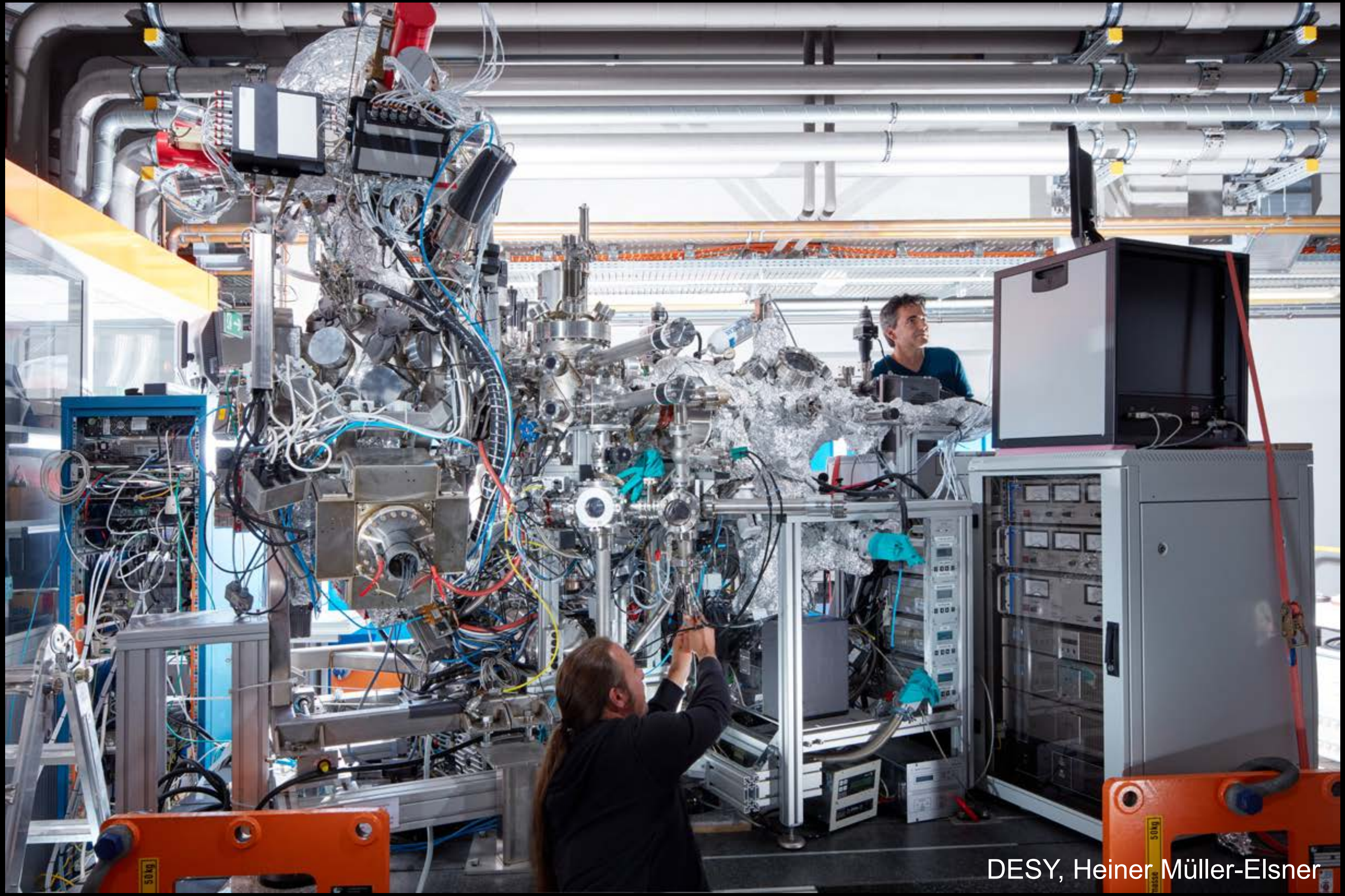
Soft x-rays

Photoelectron Spectroscopy

2D materials

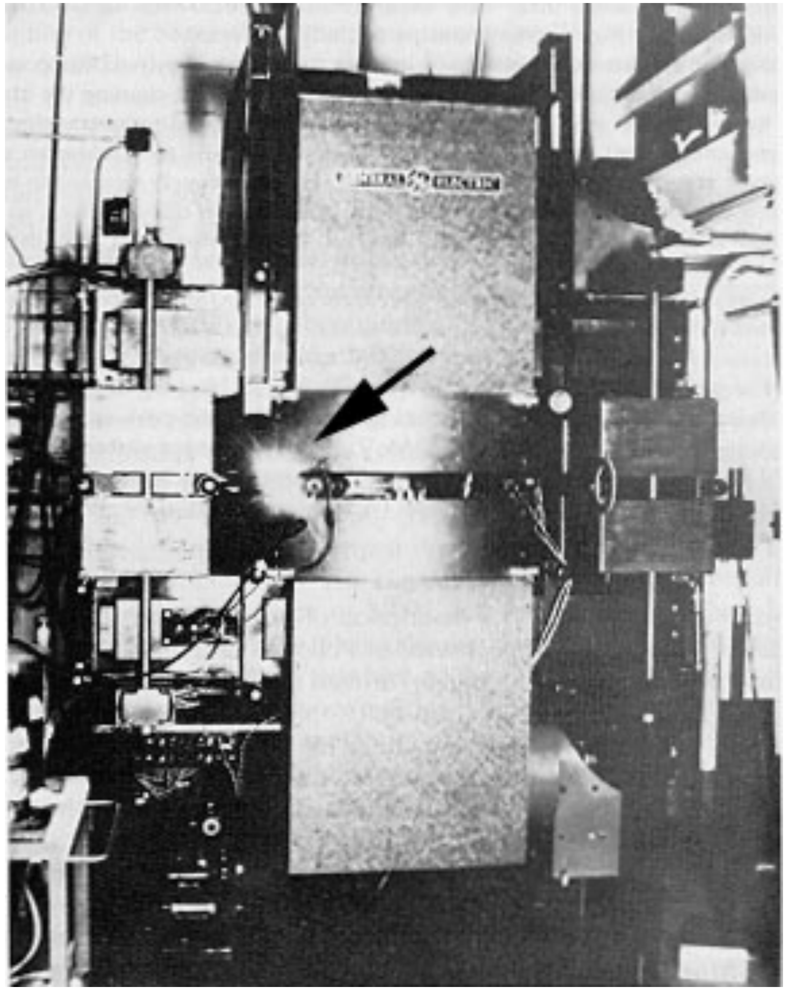
Part I: Synchrotron & FEL experiments

General concepts

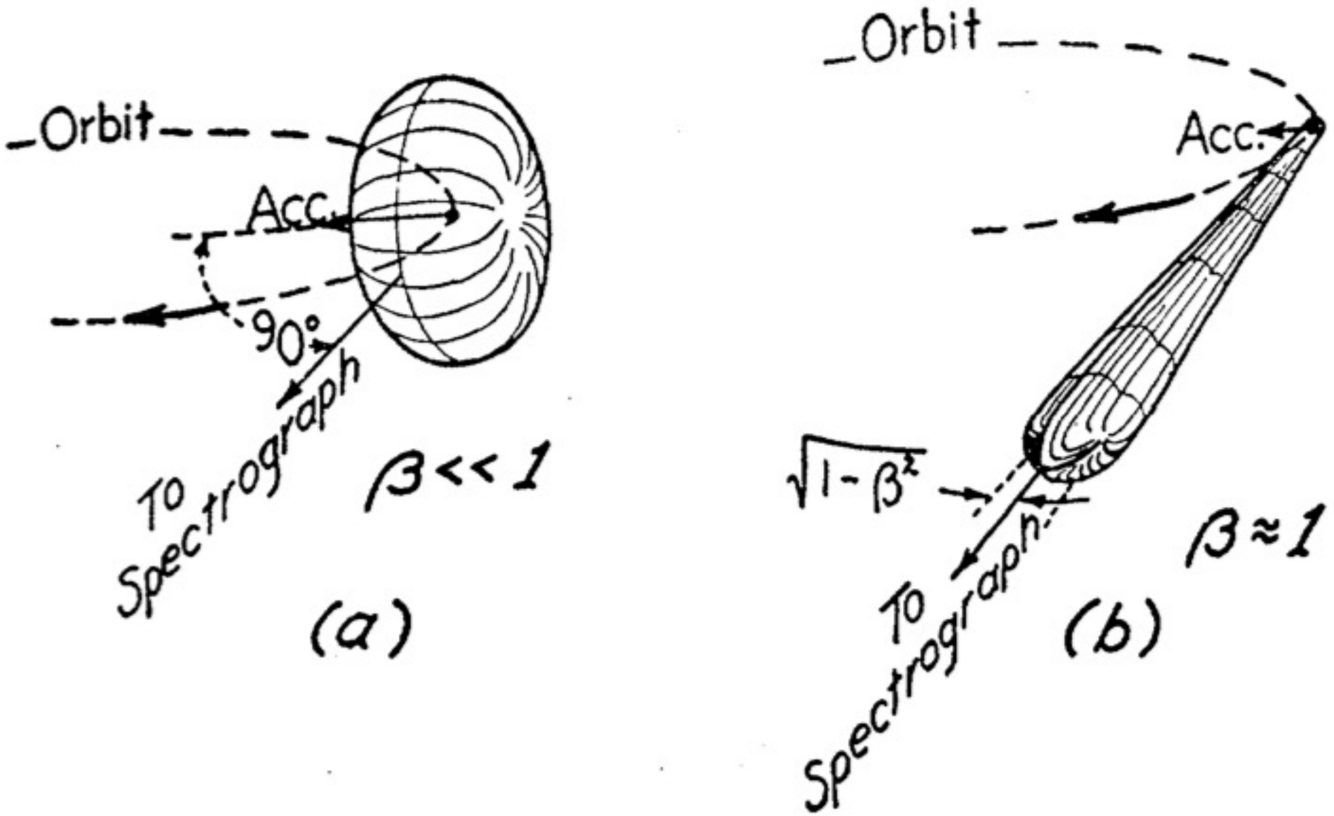

DESY, Heiner Müller-Elsner

First observation of synchrotron light

April 24, 1947 — General Electric Research Laboratory, Schenectady, New York



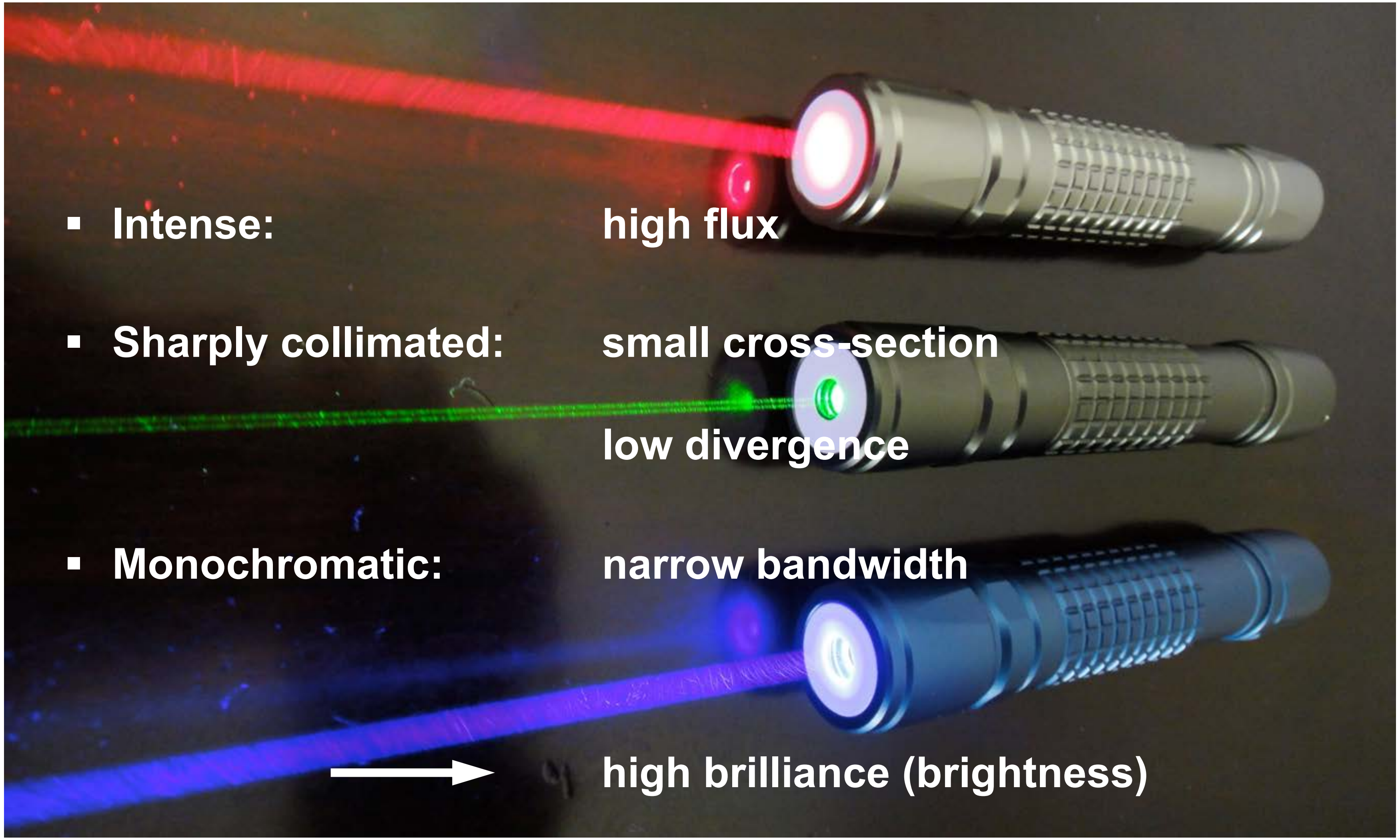
xdb.lbl.gov/Section2/Sec_2-2.html



Tomboulian & Hartman, Phys. Rev. **102**, 1423 (1956)

Laser radiation

Laser-like x-rays?



- **Intense:** high flux — $$10^{15}\,\frac{\text{photons}}{\text{s}} \simeq 1\,\text{mW}$$
- **Sharply collimated:** small cross-section — $$1\,\text{mm}^2$$
  low divergence — $$1\,\text{mrad}^2 = (0.057^\circ)^2$$
- **Monochromatic:** narrow bandwidth — $$10^{-3} = \frac{\Delta\nu}{\nu}$$

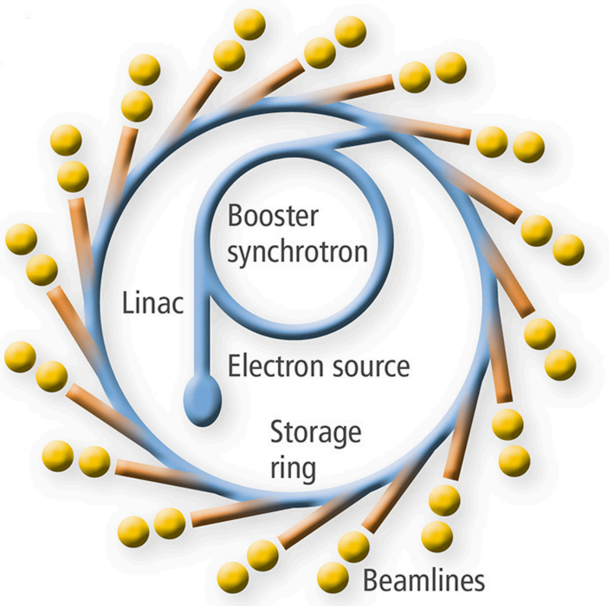
→ high brilliance (brightness)

$$10^{15}\,\frac{\text{photons}}{\text{s}\,\text{mm}^2\,\text{mrad}^2\,0.1\%\text{BW}}$$

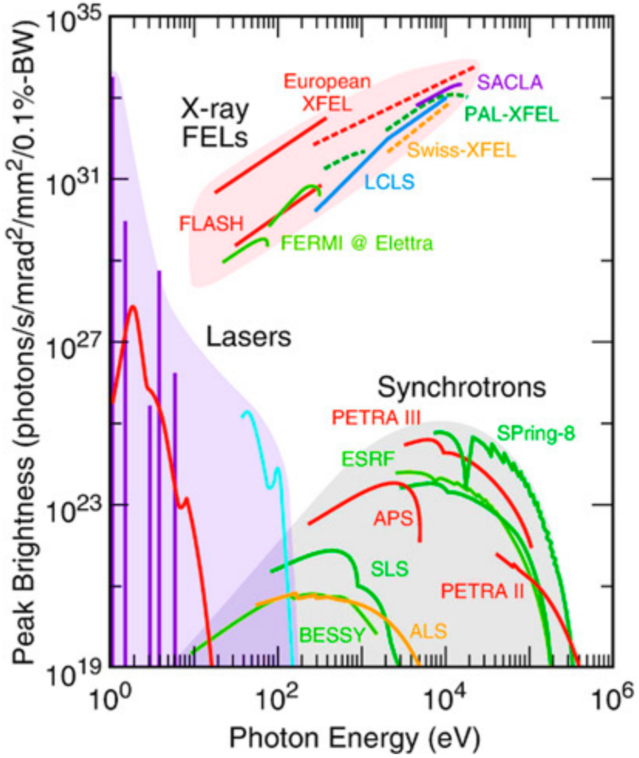
Storage rings & free-electron lasers

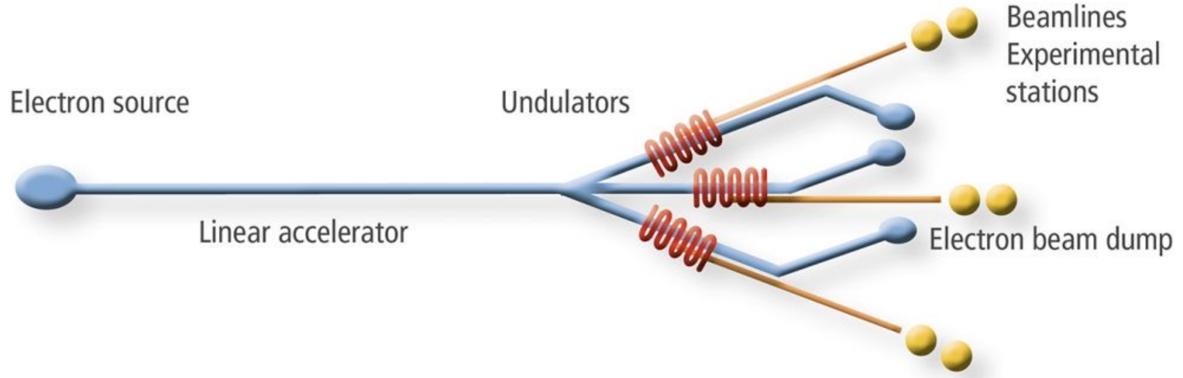
Not “big science”, but big collection of “small science”

leaps-initiative.eu



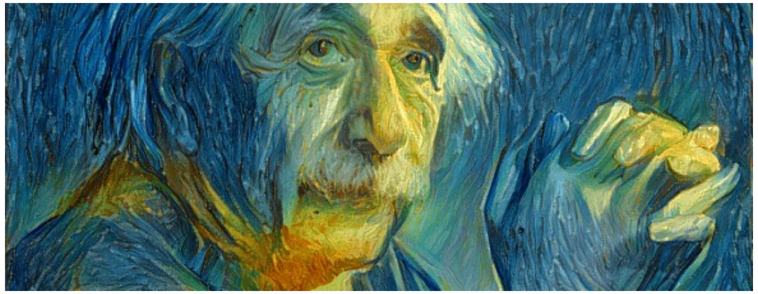
doi.org/10.1007/978-3-030-00551-1_1





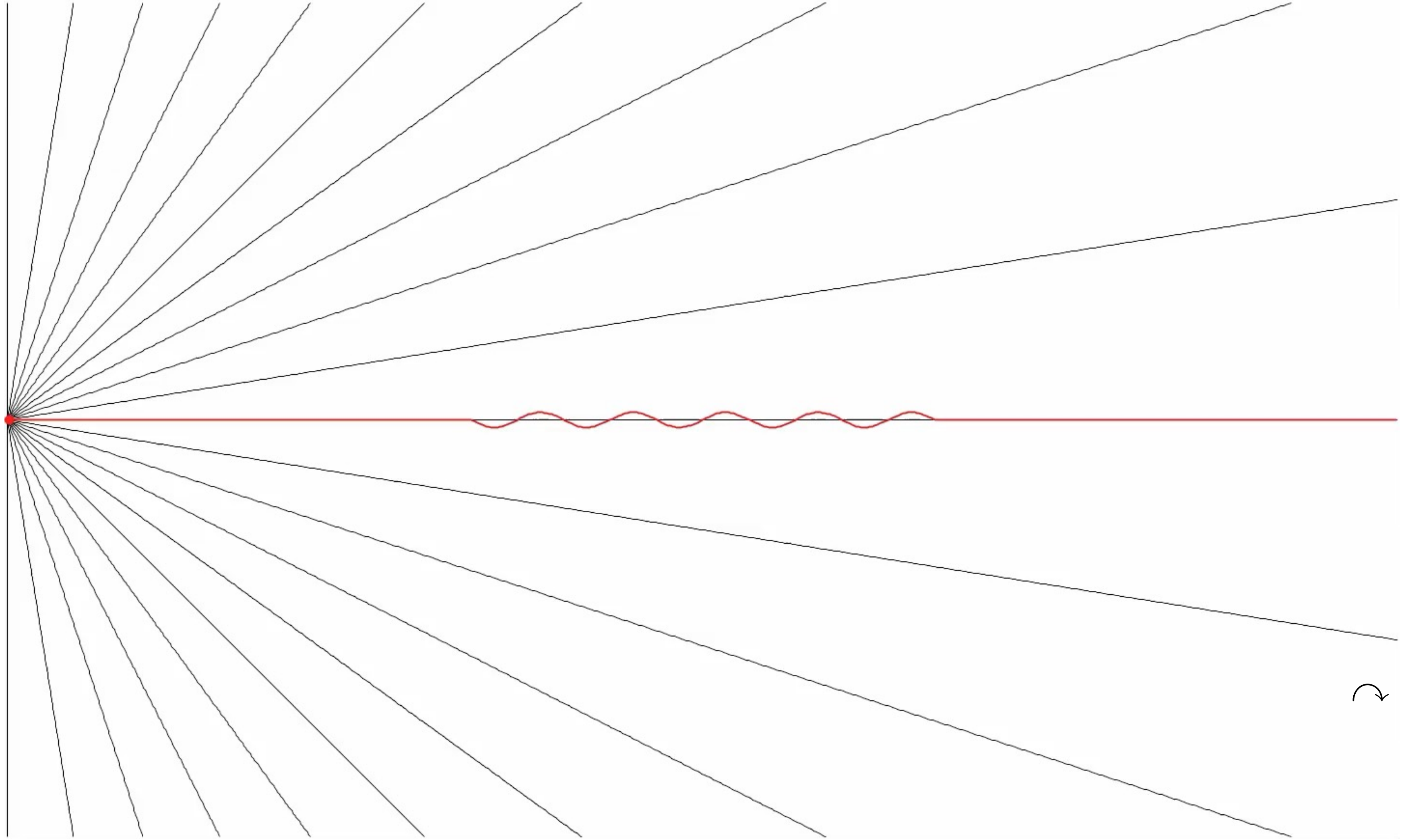
Undulator radiation

Laser-like x-rays by the power of γ



$$\gamma = 1957\, E(\mathrm{GeV})$$

$$1\,\mathrm{W} \iff 5 \cdot 10^{15}\, \lambda(\mathrm{nm})\, \frac{\mathrm{photons}}{\mathrm{s}}$$



$$\lambda \approx \frac{\lambda_\mathrm{u}}{2\gamma^2} = \mathcal{O}(1\,\mathrm{nm})$$

$$\left(\frac{\Delta\lambda}{\lambda}\right)_\mathrm{cen} = \frac{1}{N} = \mathcal{O}(1\%)$$

$$\vartheta_\mathrm{cen} \approx \frac{1}{\gamma\sqrt{N}} = \mathcal{O}(10\,\mu\mathrm{rad})$$

$$\overline{P}_\mathrm{cen} \propto \frac{\gamma^2 N_\mathrm{e}}{\lambda_\mathrm{u}} = \mathcal{O}(1\,\mathrm{W})$$

$$\sigma_{y,x} = \mathcal{O}(10 \cdots 100\,\mu\mathrm{m})$$

$$\curvearrowright \quad \approx 10^{20}\, \frac{\mathrm{photons}}{\mathrm{s\,mm^2\,mrad^2\,0.1\%BW}}$$

+ coherence + polarization

Beamlines

From source to sample: Monochromatization plus focusing

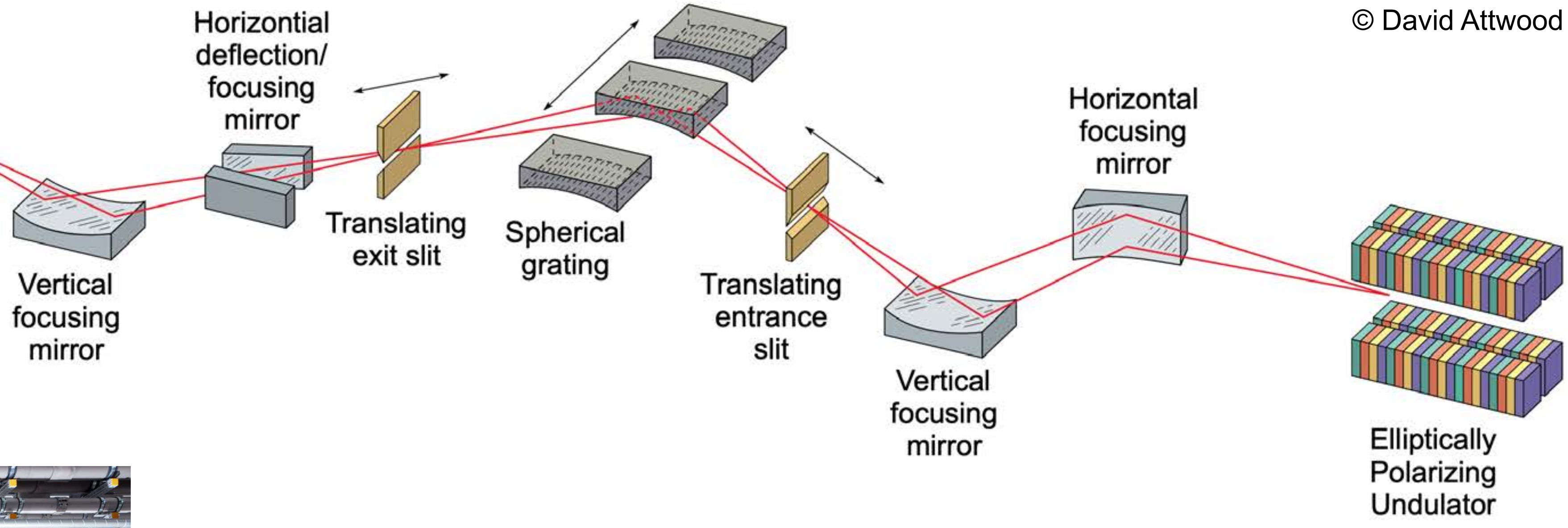
$\hbar\omega', \mathbf{q}', \mathbf{x}, \tau$

$E_{\mathrm{kin}}, \mathbf{k}, \mathbf{x}, \tau$

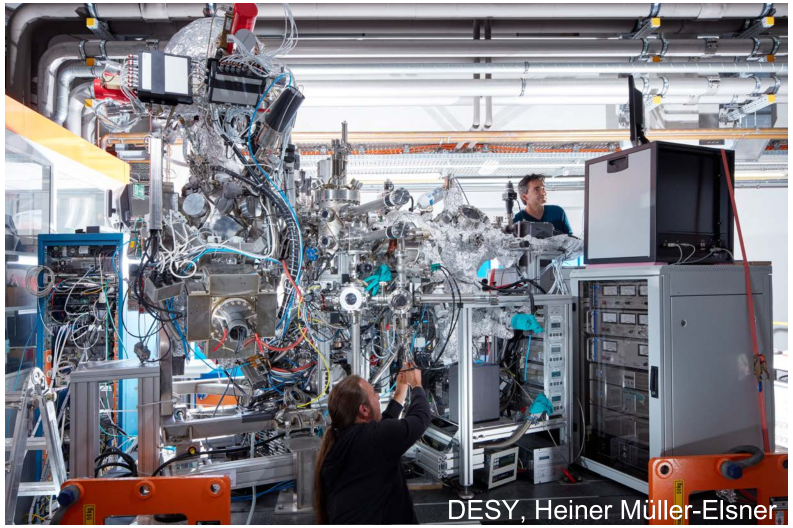
$$\frac{\Delta\hbar\omega}{\hbar\omega} \gtrsim 10^{-6}$$

$$\Delta x = 10\,\mathrm{nm} \cdots 100\,\mu\mathrm{m}$$

$$I \approx 10^{12}\,\frac{\mathrm{photons}}{\mathrm{s}}$$



© David Attwood



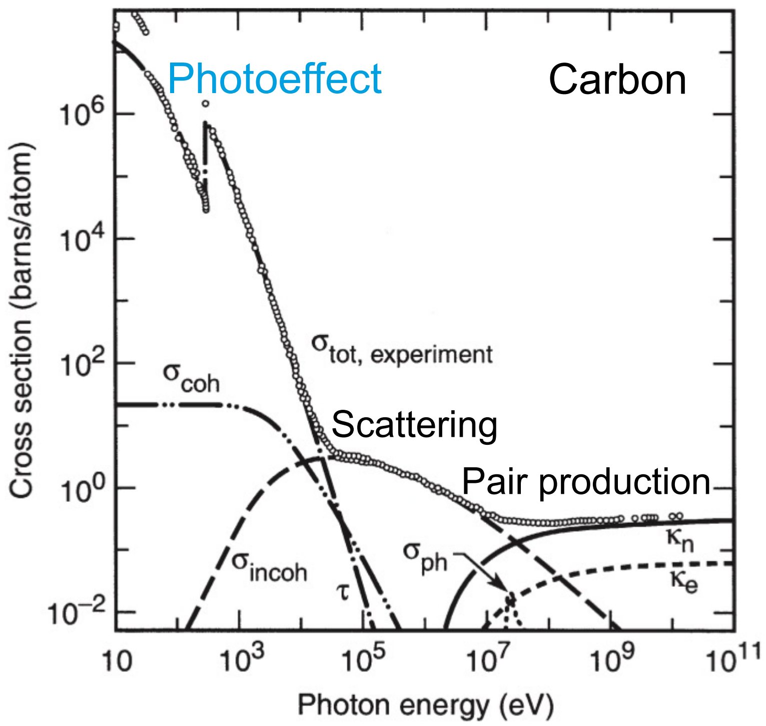
DESY, Heiner Müller-Elsner

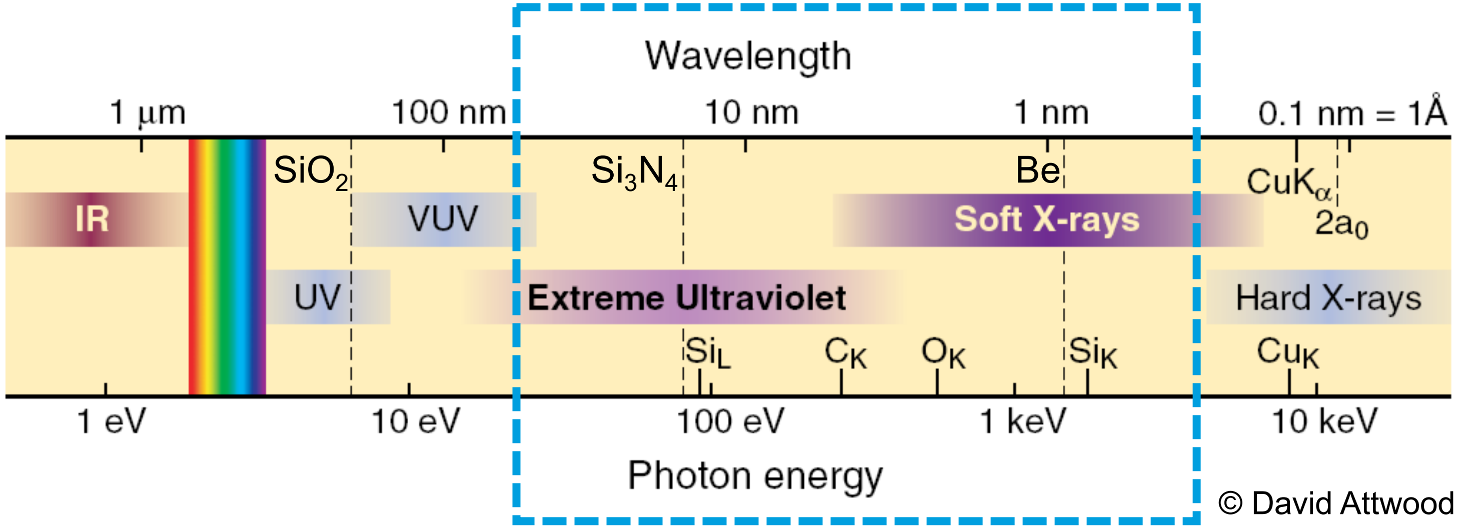
Detect at sample: Photons & electrons

Measure:	Energy	Spectroscopy
	Momentum	Scattering & diffraction
	Position	Imaging & microscopy
	Time	Dynamics

Why (soft) x-rays?

“Right” cross-section and useful spectral range





$$\hbar\omega\,\lambda = 1240\,\mathrm{eV\,nm}$$

$$\hbar\omega \approx 30 \cdots 3000\,\mathrm{eV}$$

$$\lambda \approx 0.4 \cdots 40\,\mathrm{nm}$$

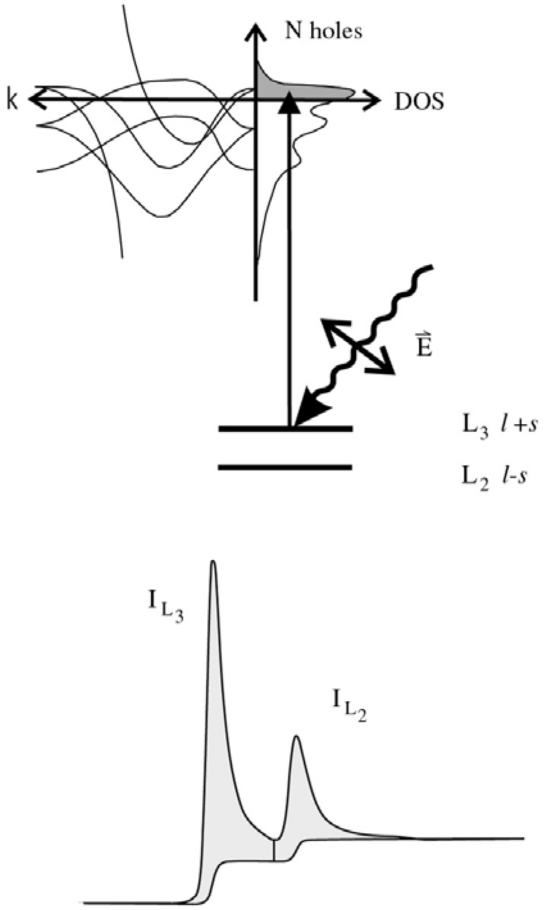
Absorption lengths in h-^{10}BN:

- 1 keV electrons: 2 nm
- 1 keV photons: 2 μm
- Thermal neutrons: 50 μm

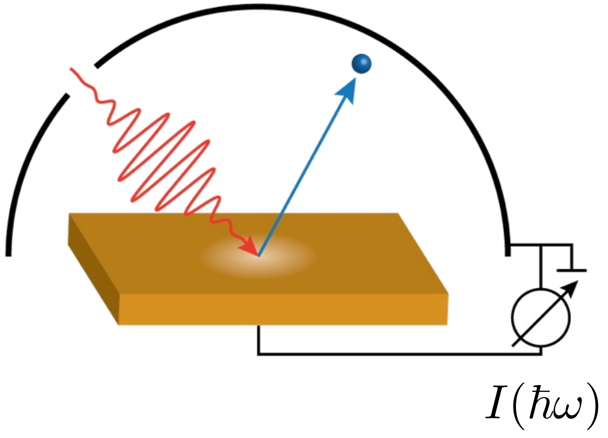
- Many atomic resonances: element & chemical sensitivity, cross-section enhancements
- Relatively short photon wavelengths: nanometer spatial resolution
- Intermediate electron kinetic energies: surface & interface sensitivity

X-ray absorption spectroscopy (XAS)

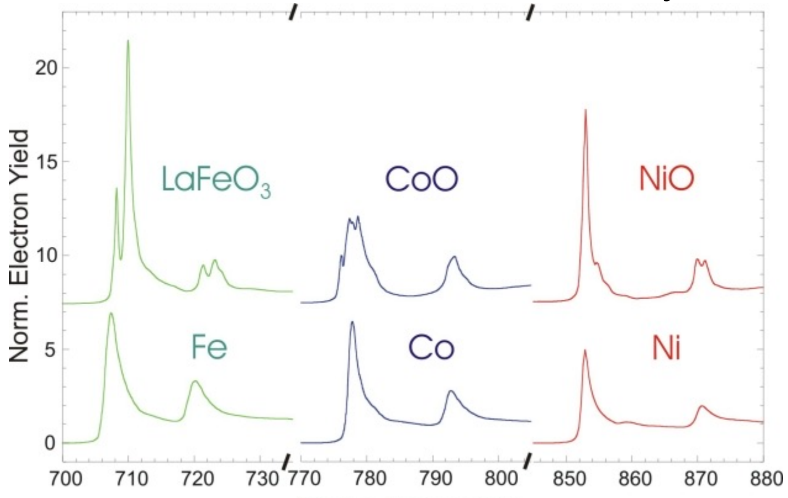
Tunable x-ray energies and a picoammeter make a thin-film spectroscopy



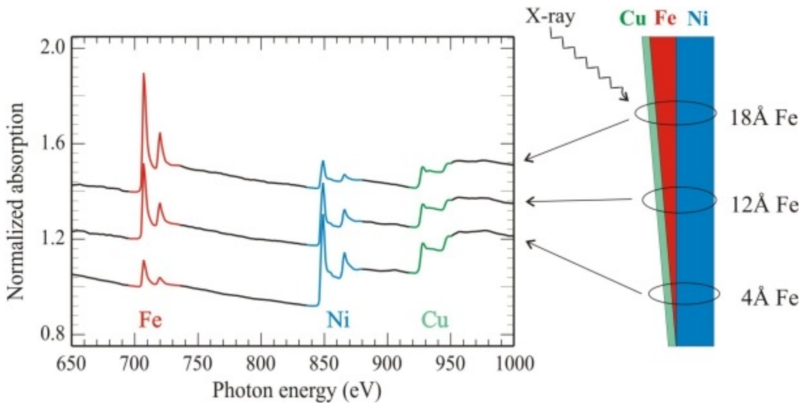
www-ssrl.slac.stanford.edu/stohr/xmcd.htm
Stöhr, J. Magn. Magn. Mater. **200**, 470 (1999)



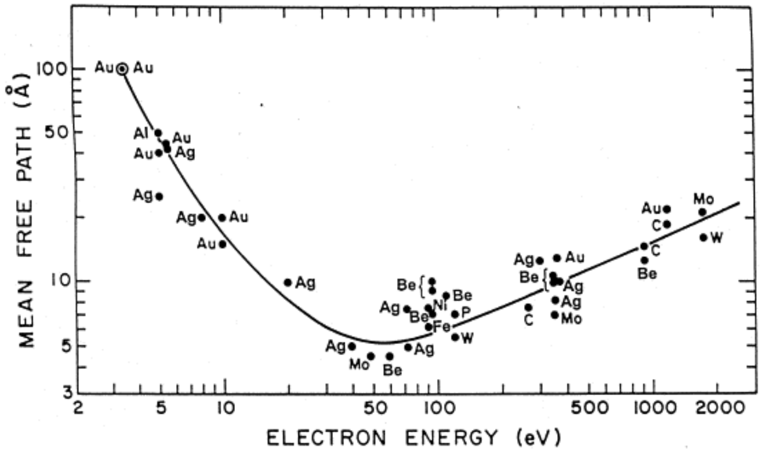
Element & chemical sensitivity



Layer specificity

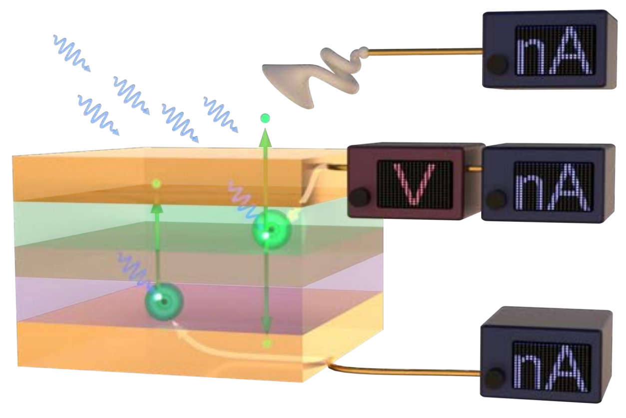


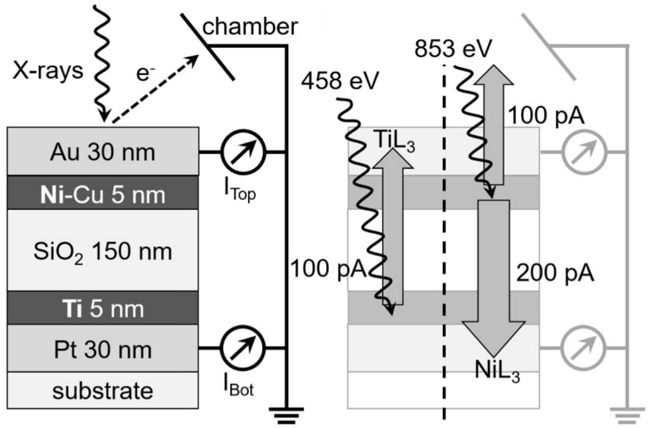
Surface & interface sensitivity

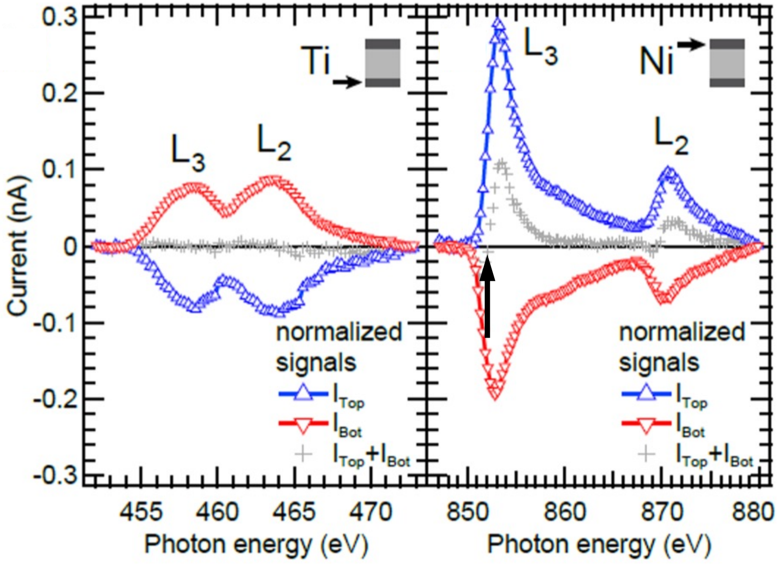


In operando XAS

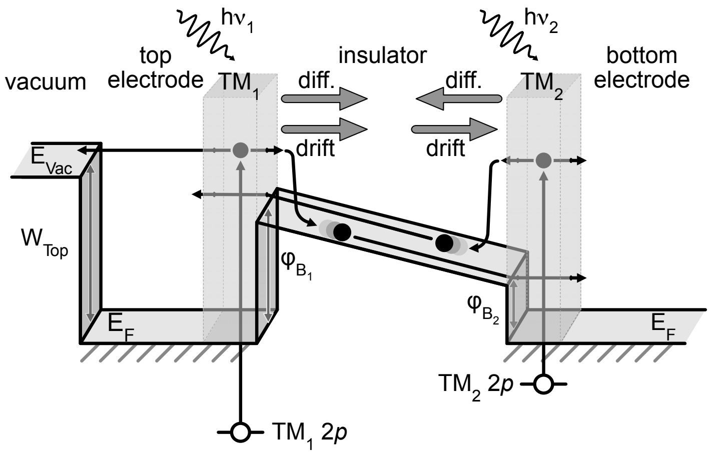
Tunable x-ray energies and two picoammeters make a device spectroscopy







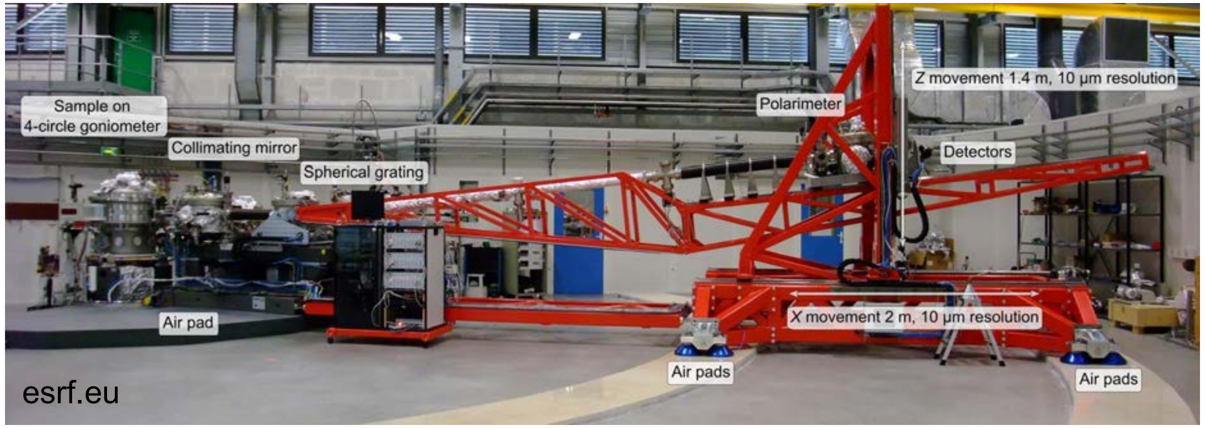
- Interface specificity
- Chemical sensitivity
- Drift & diffusion currents
- Barrier heights
- Built-in potentials
- …



Kröger *et al.*, Appl. Phys. Lett. **120**, 181601 (2022)

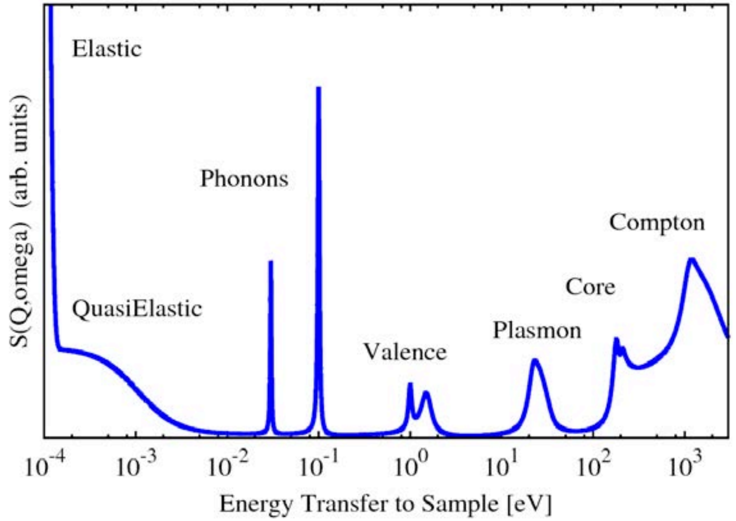
Energy

Resolution down to 1 meV

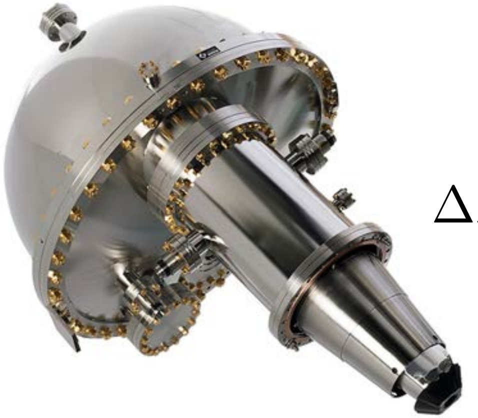


$$\Delta\hbar\Omega \gtrsim 1\,\mathrm{meV}$$

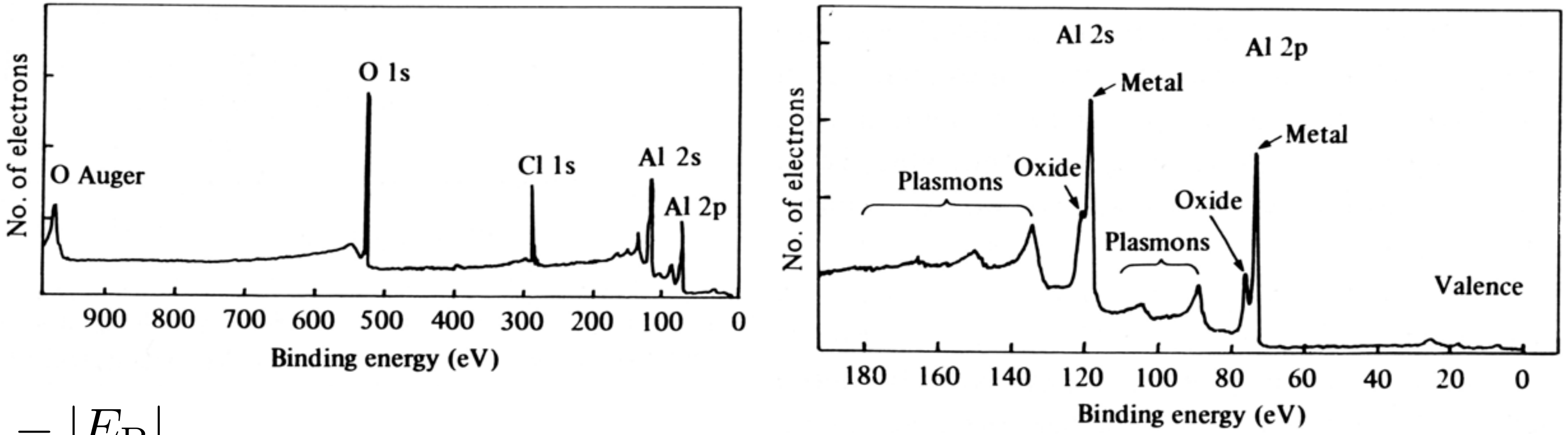
Baron, arXiv:1504.01098



Photon energy loss: $\hbar\Omega = \hbar\omega - \hbar\omega'$



$$\Delta E_{\mathrm{kin}} \gtrsim 1\,\mathrm{meV}$$

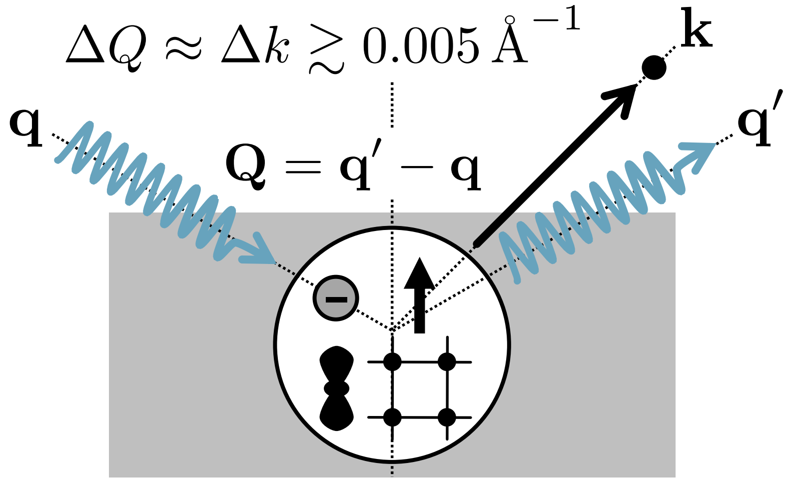


Electron energy gain: $E_{\mathrm{kin}} = \hbar\omega - \Phi - |E_{\mathrm{B}}|$

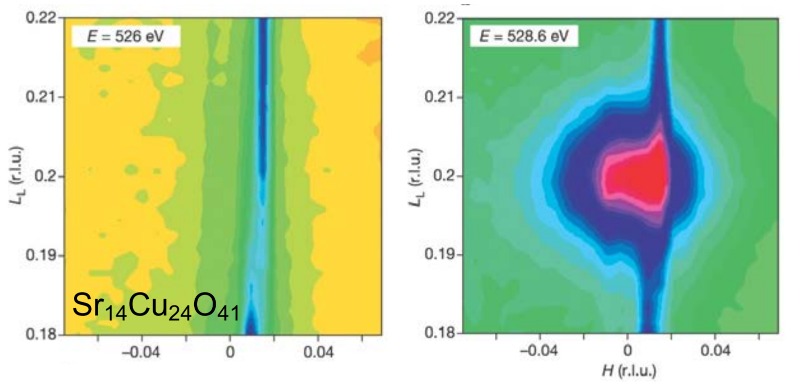
Woodruff & Delchar, *Modern Techniques of Surface Science*

Momentum

Resolution down to 0.005 Å^{-1}

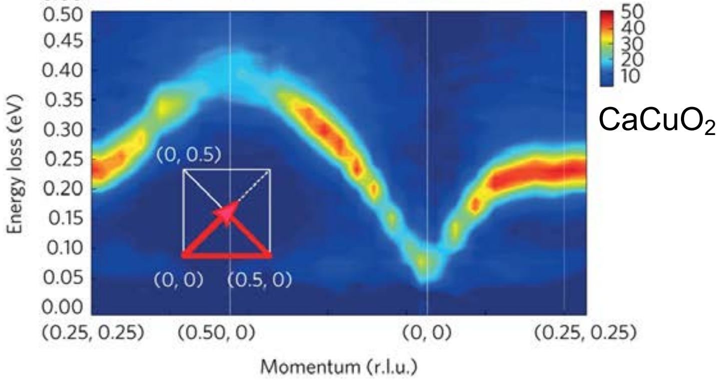


Ordered (super)structures



Abbamonte *et al.*, Nature **431**, 1078 (2004)

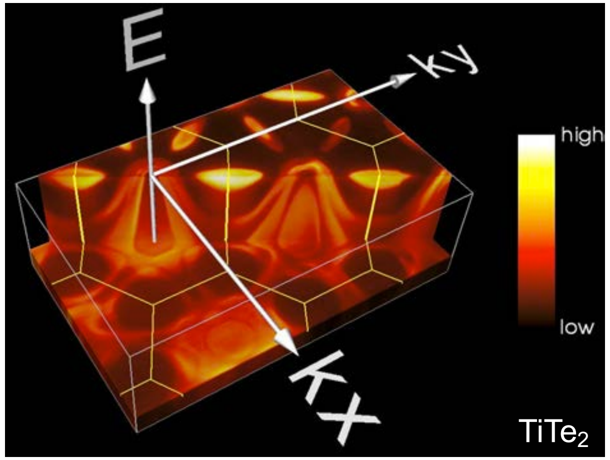
Collective excitations



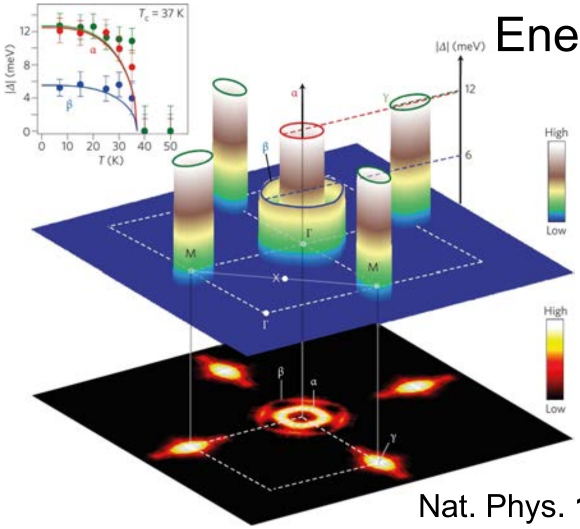
$CaCuO_2$

Peng *et al.*, Nat. Phys. **13**, 1201 (2017)

Band structures & Fermi surfaces



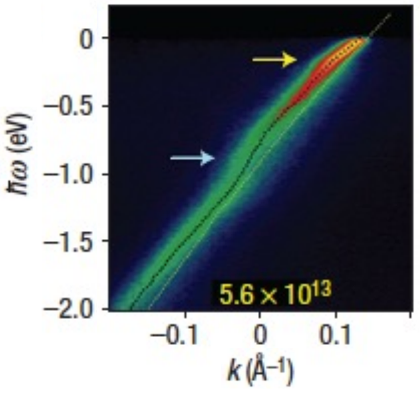
Energy gaps



$Ba_{0.6}K_{0.4}Fe_2As_2$

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

Quasiparticle interactions



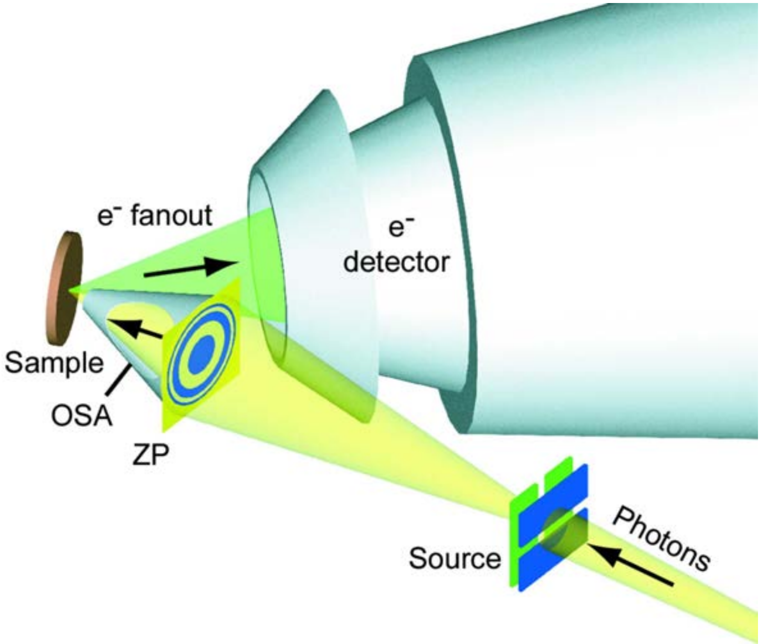
Graphene

Bostwick *et al.*, Nat. Phys. **3**, 36 (2007)

Position

Resolution down to 10 nm

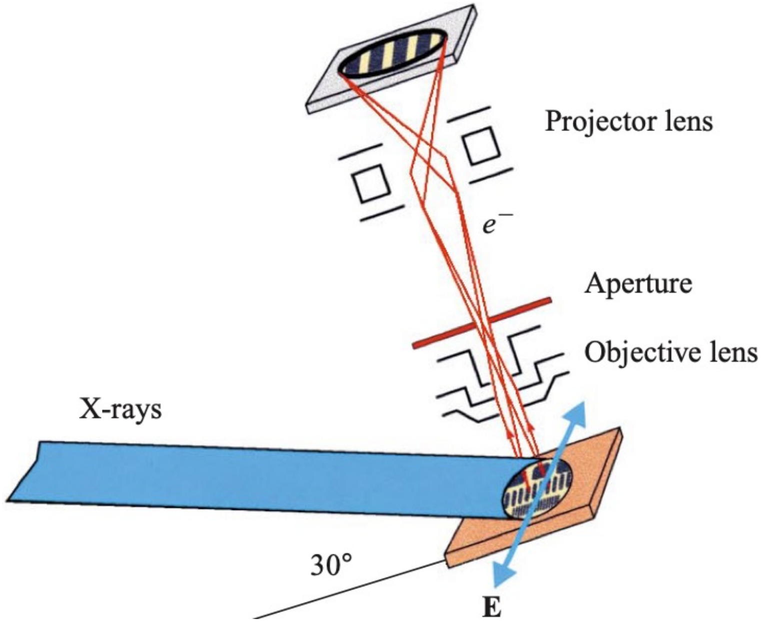
Nanofocusing



Rotenberg & Bostwick,
J. Synchrotron Rad. **21**, 1048 (2014)

$$\Delta x = \Delta y \geq 20\,\mathrm{nm}$$

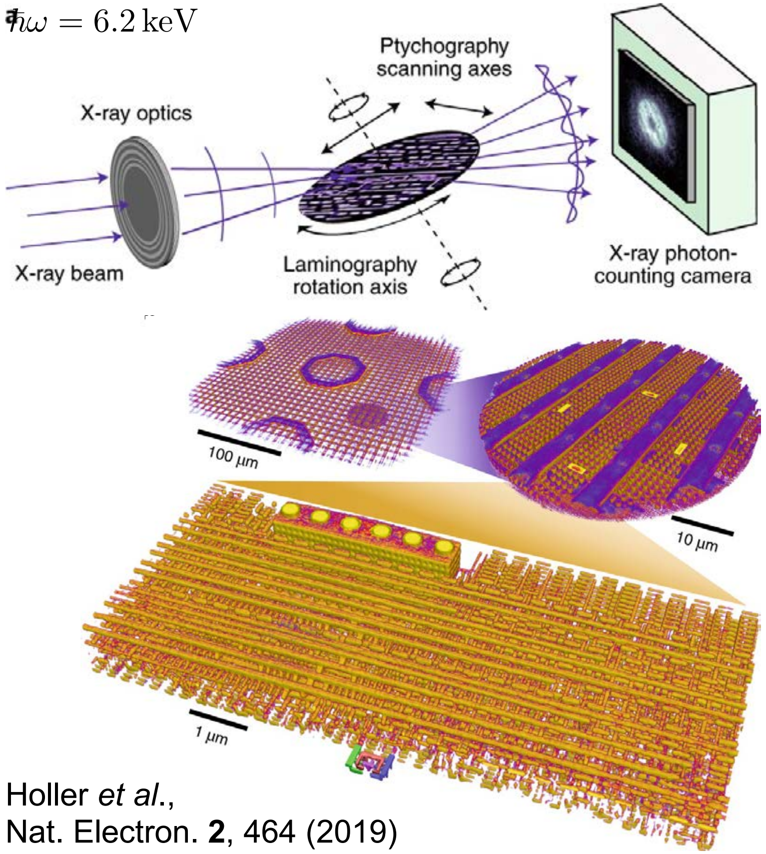
Microscopy



Stöhr & Anders,
IBM J. Res. Develop. **44**, 535 (2000)

$$\Delta x = \Delta y \gtrsim 10\,\mathrm{nm}$$

Ptychography



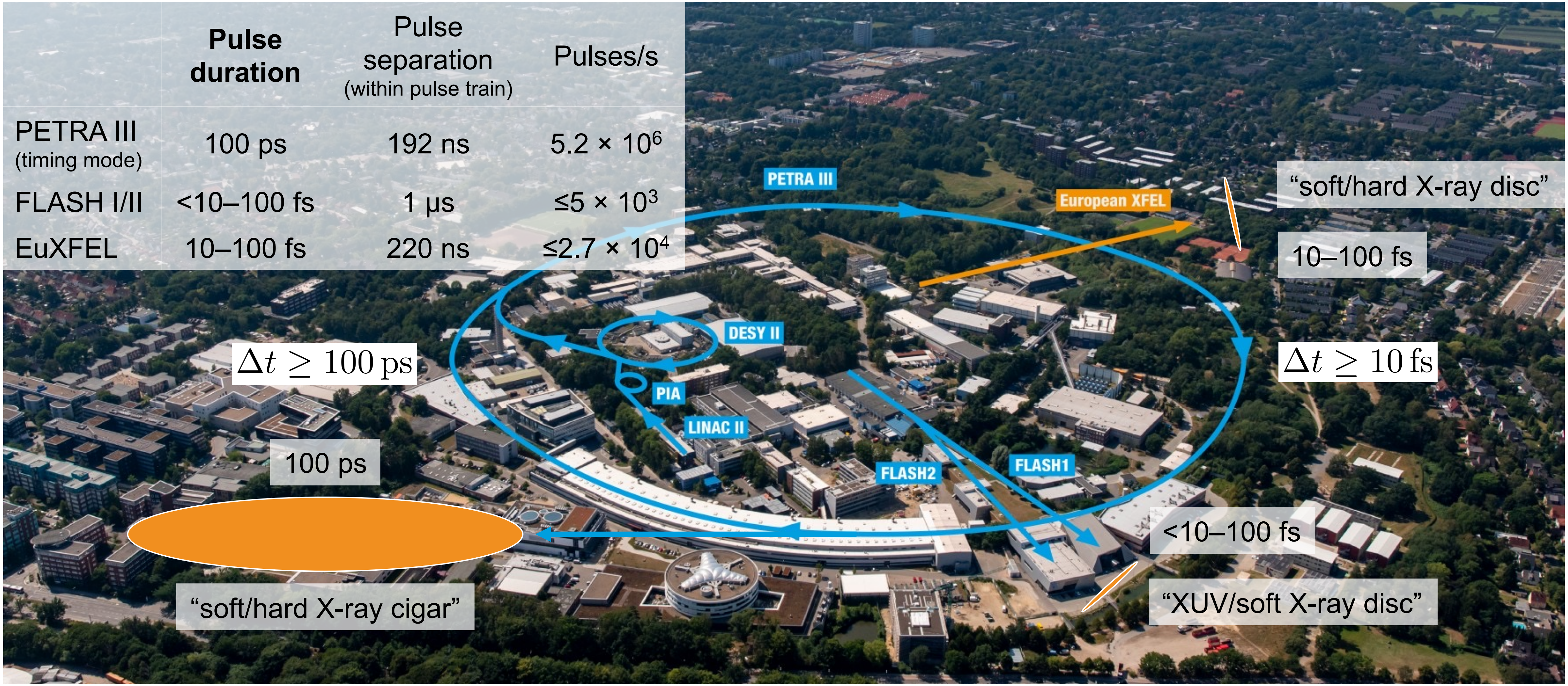
Holler *et al.*,
Nat. Electron. **2**, 464 (2019)

$$\Delta x = \Delta y = \Delta z \approx 10\,\mathrm{nm}$$

Time

Resolution down to 10 fs

	Pulse duration	Pulse separation (within pulse train)	Pulses/s
PETRA III (timing mode)	100 ps	192 ns	5.2×10^6
FLASH I/II	<10–100 fs	1 μs	$\leq 5 \times 10^3$
EuXFEL	10–100 fs	220 ns	$\leq 2.7 \times 10^4$



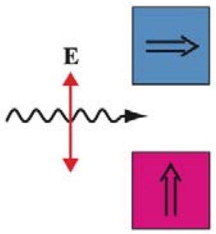
Pump-probe technique

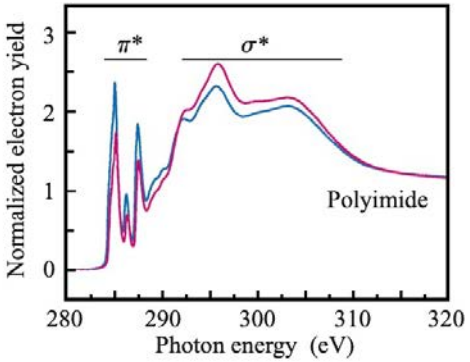
toutestquantique.fr

Polarization

Probing charge and spin anisotropies of empty valence states in core excitations

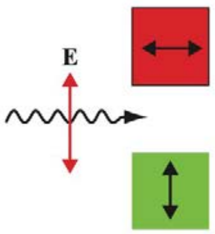
X-ray
linear dichroism

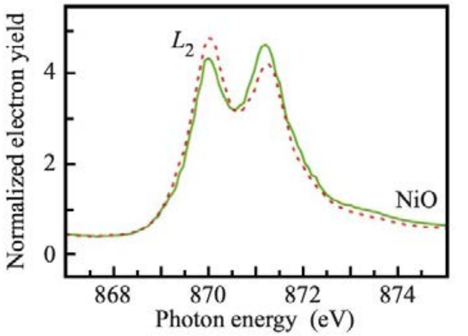




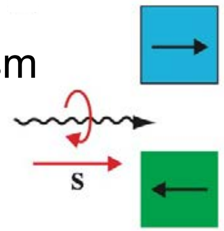
$\mathbf{E} =$ "search light"

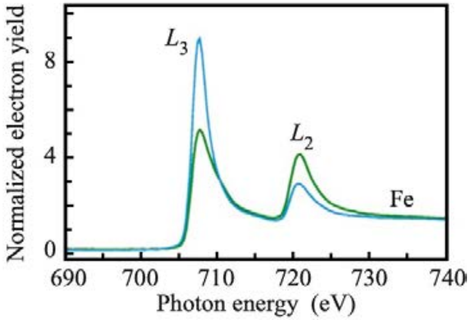
X-ray magnetic
linear dichroism



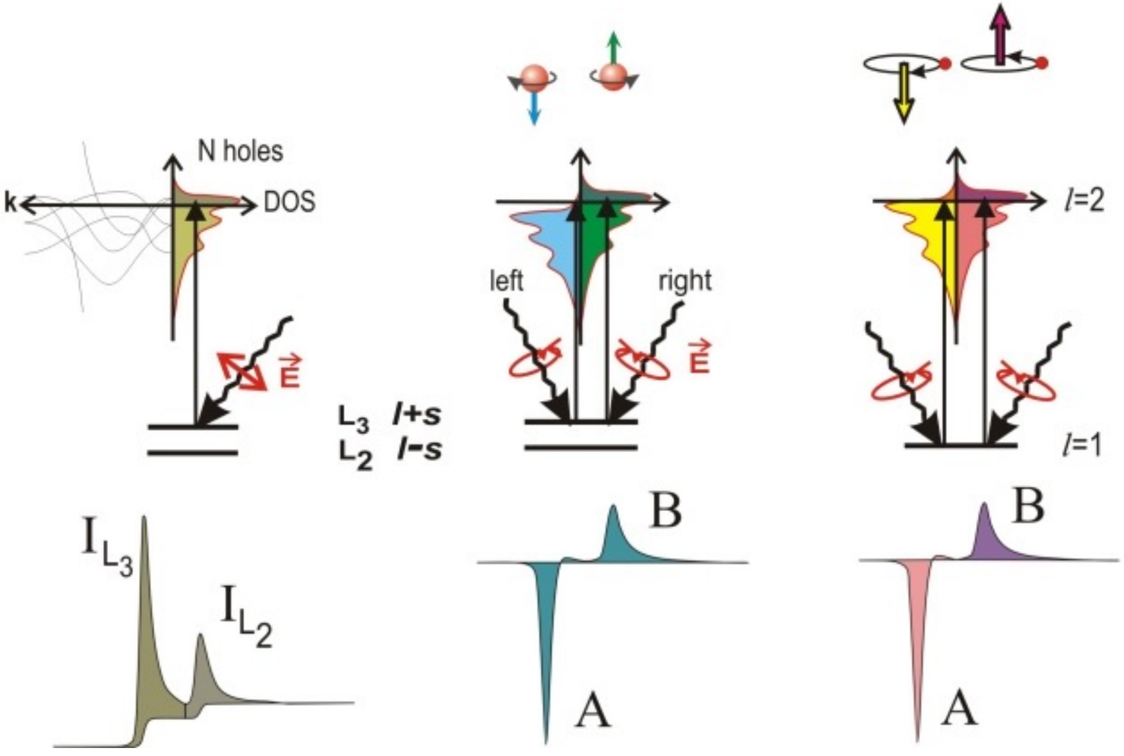


X-ray magnetic
circular dichroism





Stöhr & Anders,
IBM J. Res. Develop.



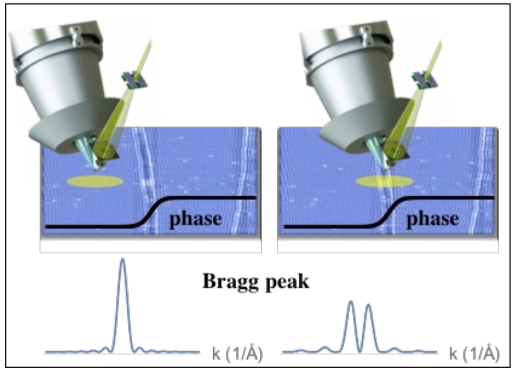
$(I_{\mathrm{L}_3} + I_{\mathrm{L}_2}) \sim N$ $\quad$ $(\mathrm{A} - 2\mathrm{B}) \sim m_{\mathrm{s}}$ $\quad$ $(\mathrm{A} + \mathrm{B}) \sim m_{\mathrm{o}}$

www-ssrl.slac.stanford.edu/stohr/xmcd.htm
Stöhr, J. Magn. Magn. Mater. **200**, 470 (1999)

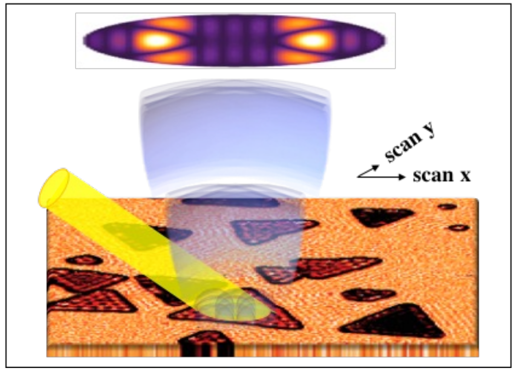
Coherence

Toward phase-sensitive spectroscopy using nanofocused & structured soft x-ray beams

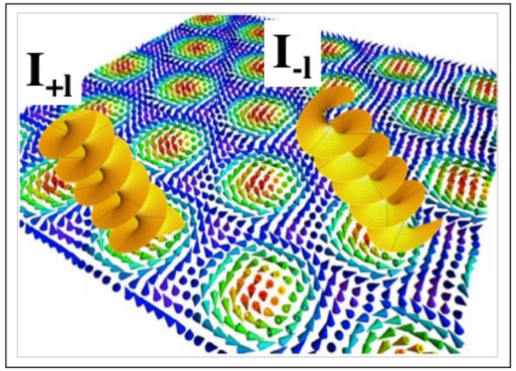
Differential interference Contrast



Photoelectron Ptychography



Twisted Dichroism



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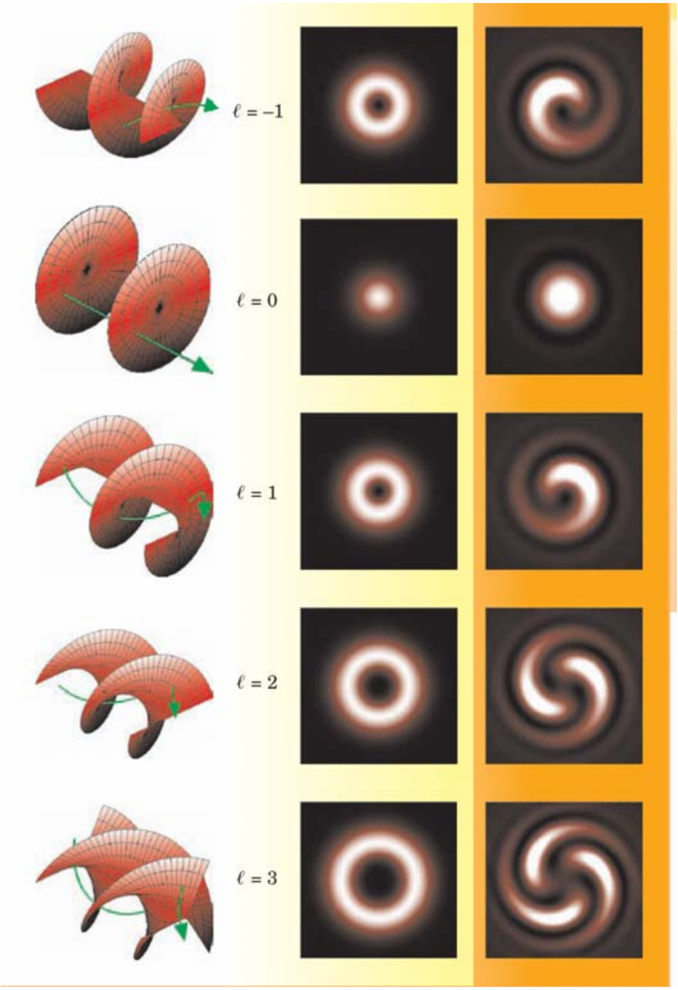
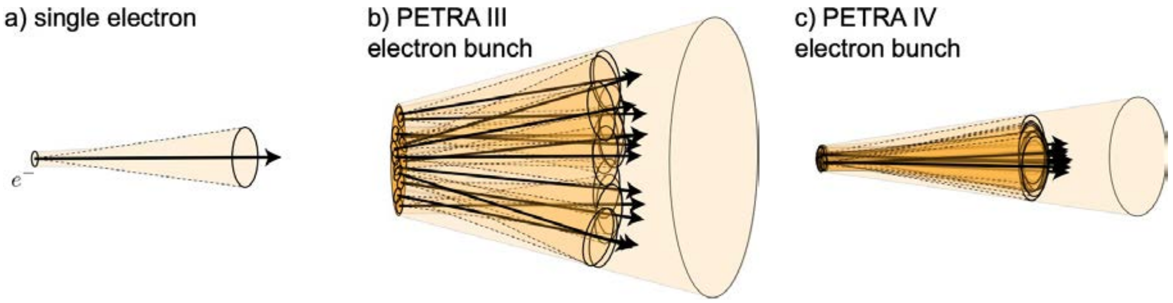
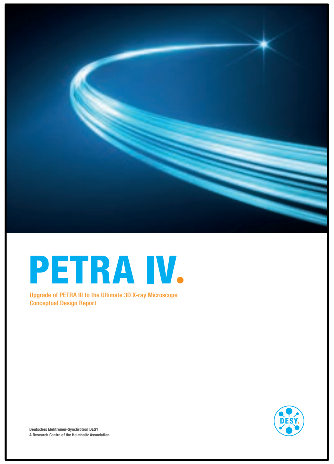


Padgett *et al.*, Physics Today **57**, 5, 35 (2004)

"The ultimate 3D X-ray microscope"



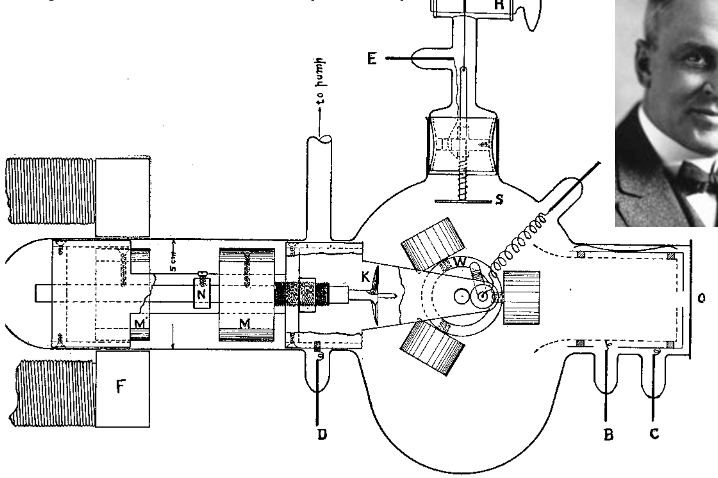


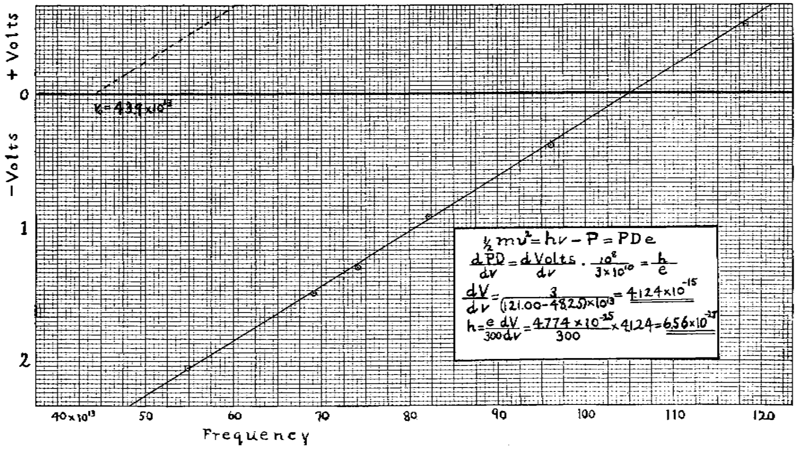
GEFÖRDERT VOM
Bundesministerium für Bildung und Forschung

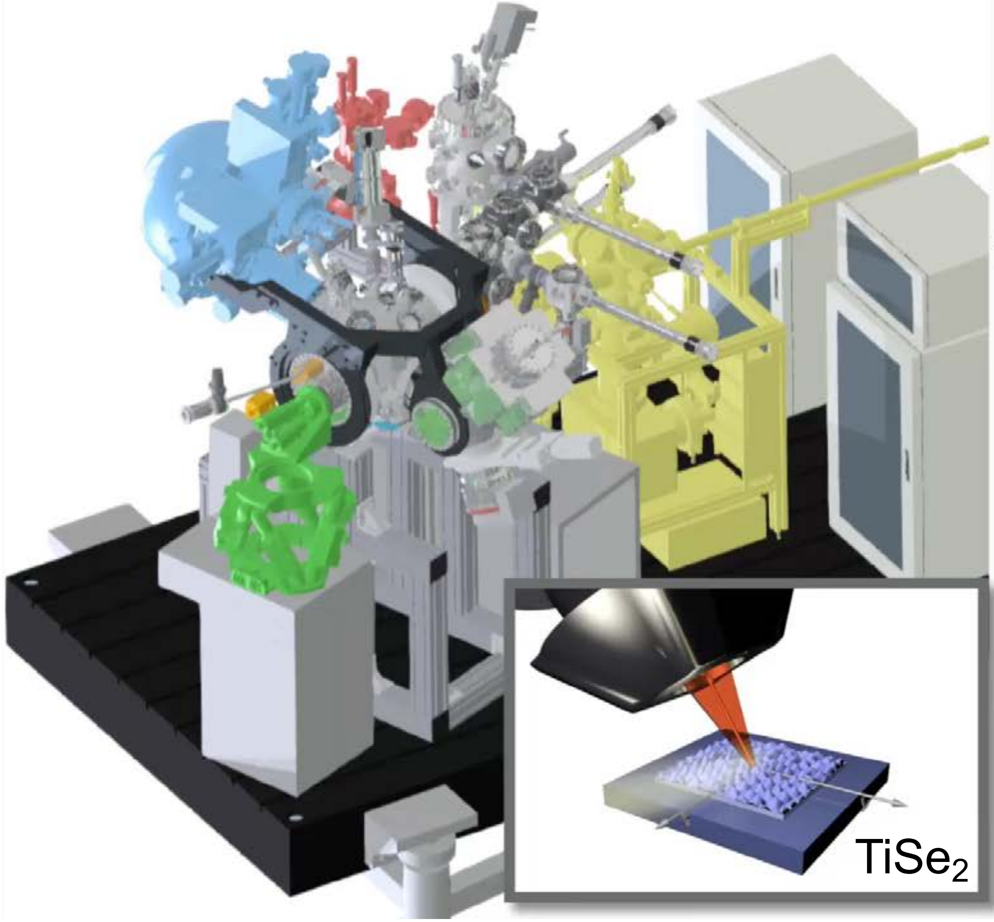
Photoelectron spectroscopy (ARPES)

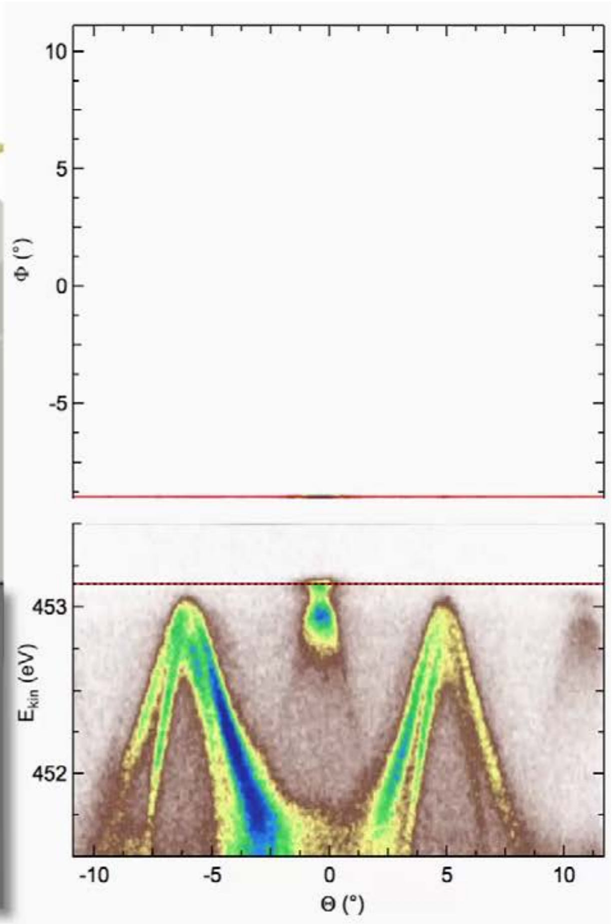
Modern “machine shop *in vacuo*”: ASPHERE III @ PETRA III / DESY

Phys. Rev. **7**, 355 (1916)









$T = 32\,\mathrm{K}$

$h\nu = 458\,\mathrm{eV}$

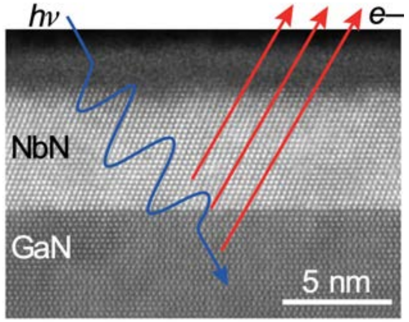
Soft x-ray ARPES

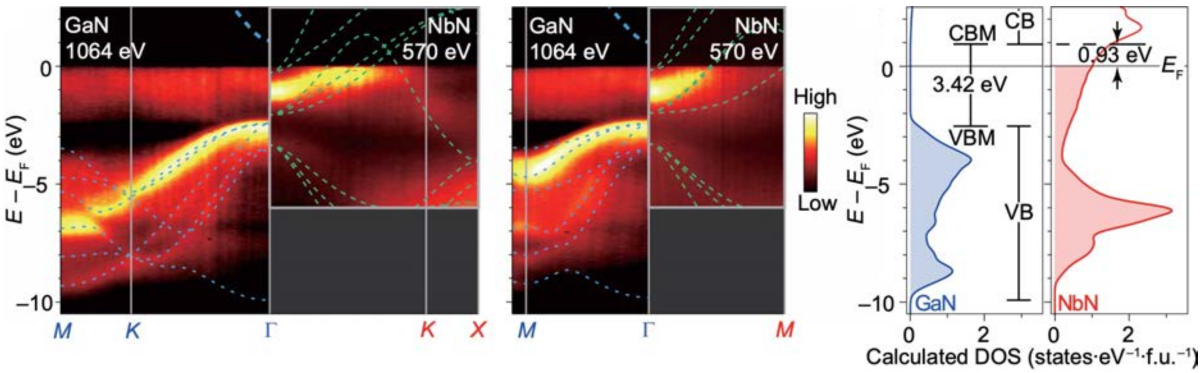
Electronic structure "in the bulk" & at interfaces

"Bulk" & interface sensitivity
(via soft x-ray energies)

Superconductor

Semiconductor

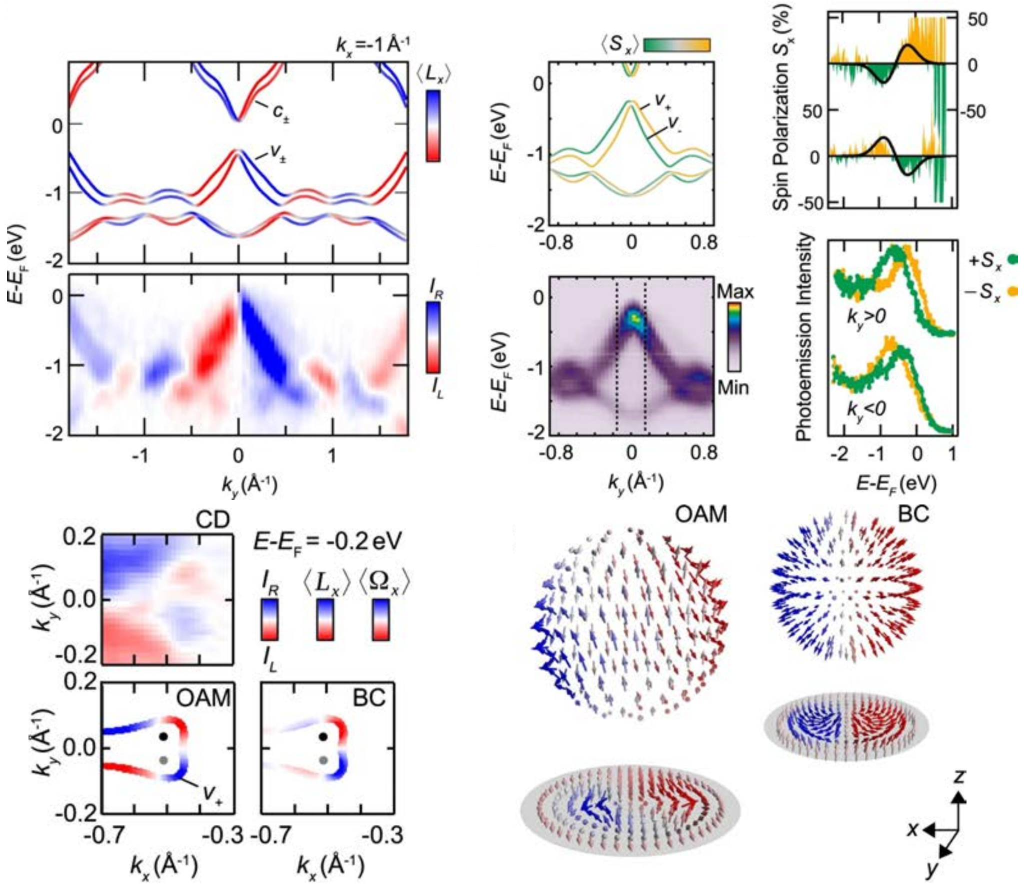




Yu *et al.*, Sci. Adv. **7**, eabi5833 (2021)

Orbital angular momentum & spin polarization
(via circular dichroism & spin detection)

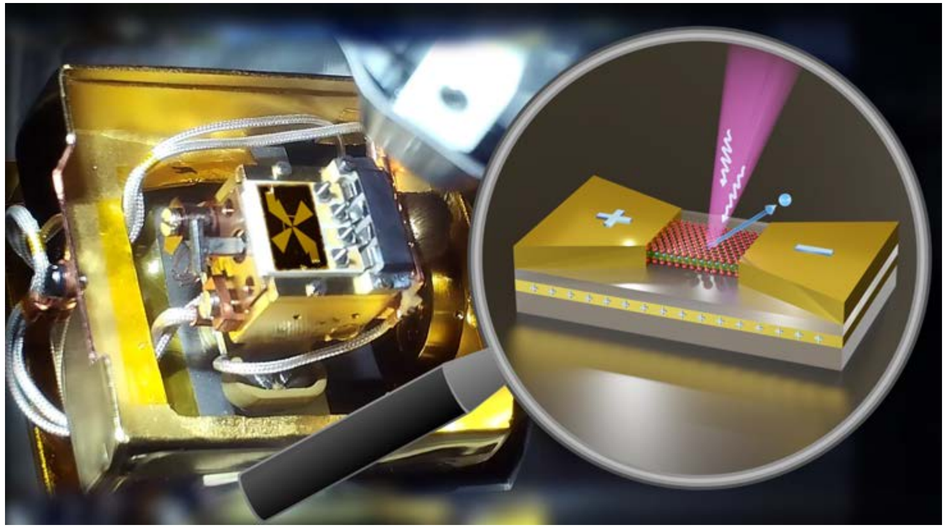
Weyl semimetal TaAs

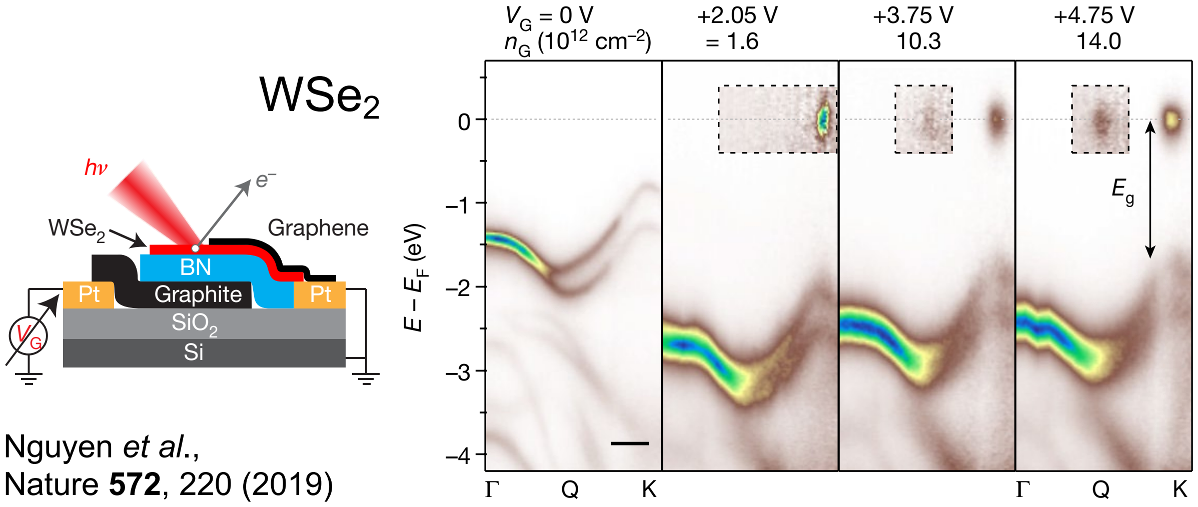


Ünzelmann *et al.*, Nat. Commun. **12**, 3650 (2021)

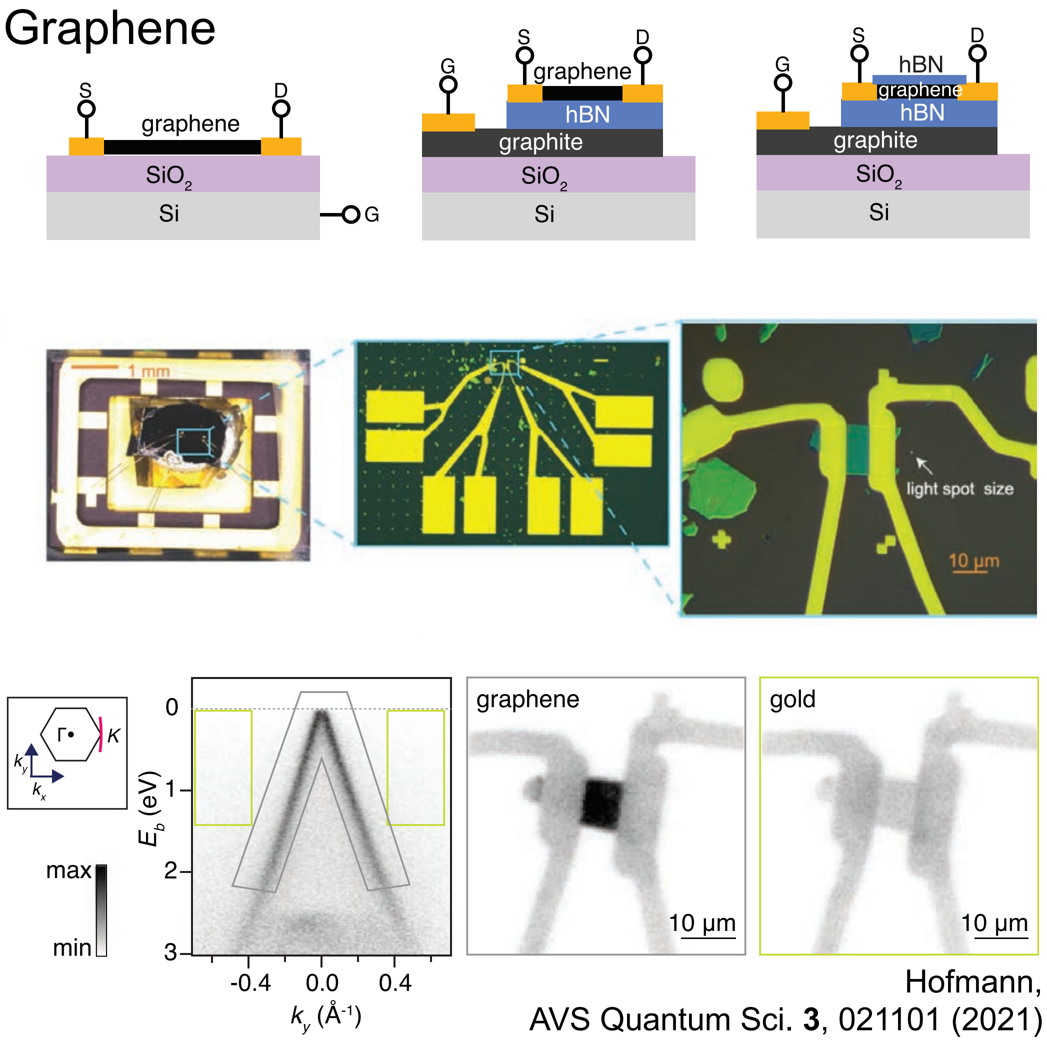
In operando micro-/nano-ARPES

Electronic structure in devices under bias





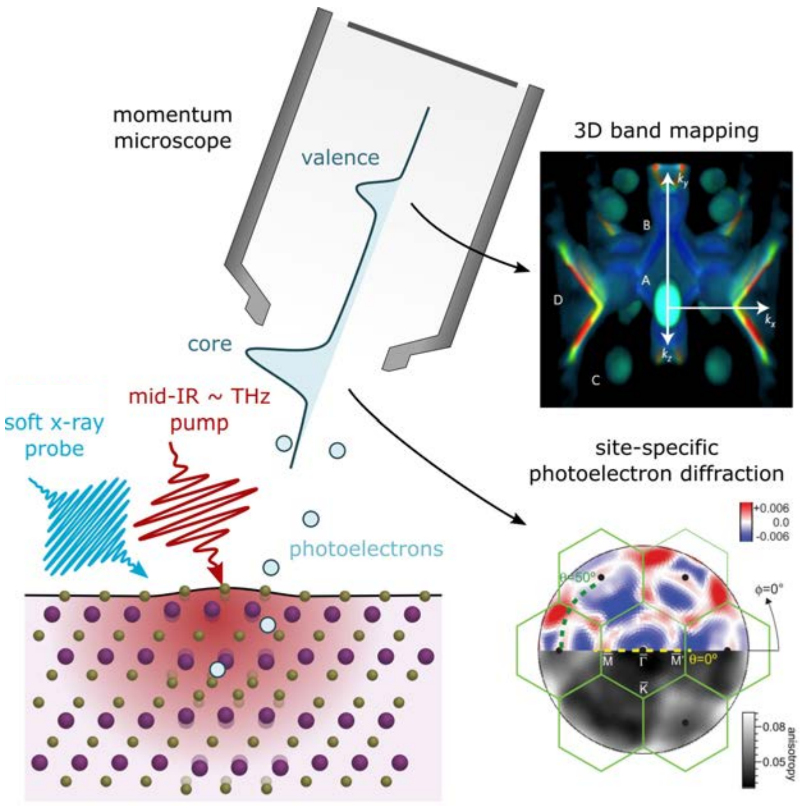
Nguyen *et al.*,
Nature **572**, 220 (2019)



Hofmann,
AVS Quantum Sci. **3**, 021101 (2021)

Ultrafast soft x-ray ARPES

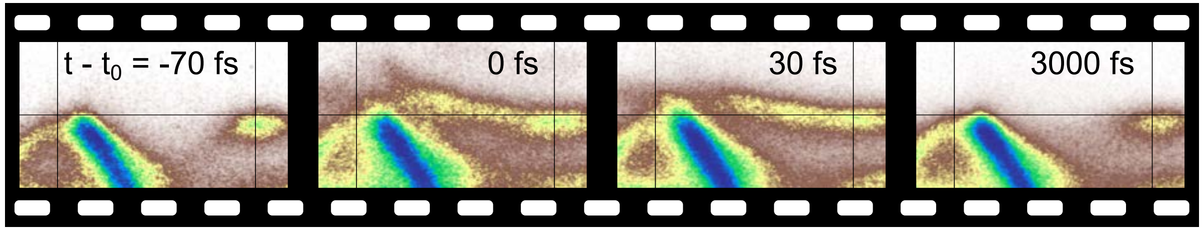
Electronic structural dynamics on fundamental time scales



© Jonathan Sobota (Stanford)

Electron hopping

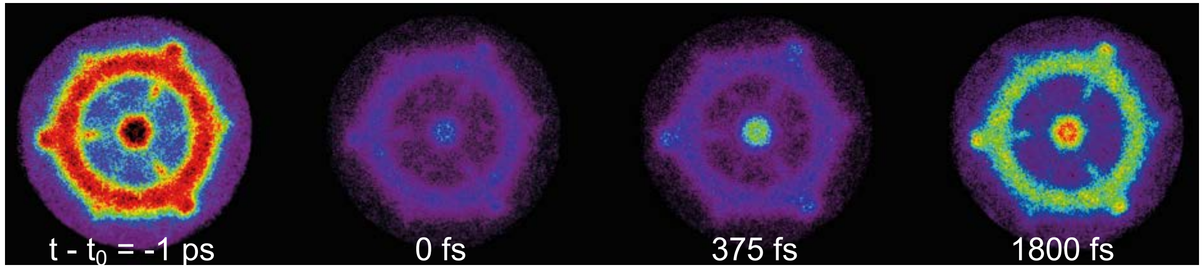
$$\tau_e = \frac{h}{W} = \mathcal{O}\left(\frac{h}{1\,\mathrm{eV}}\right) = \mathcal{O}(4\,\mathrm{fs})$$



1*T*-$TiSe_2$

Charge transfer

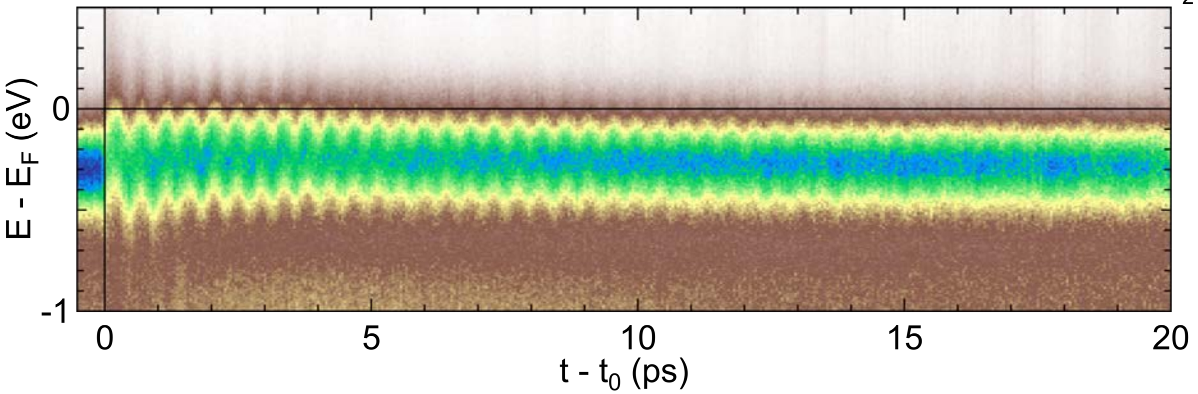
CuPc/$TiSe_2$



Lattice vibration

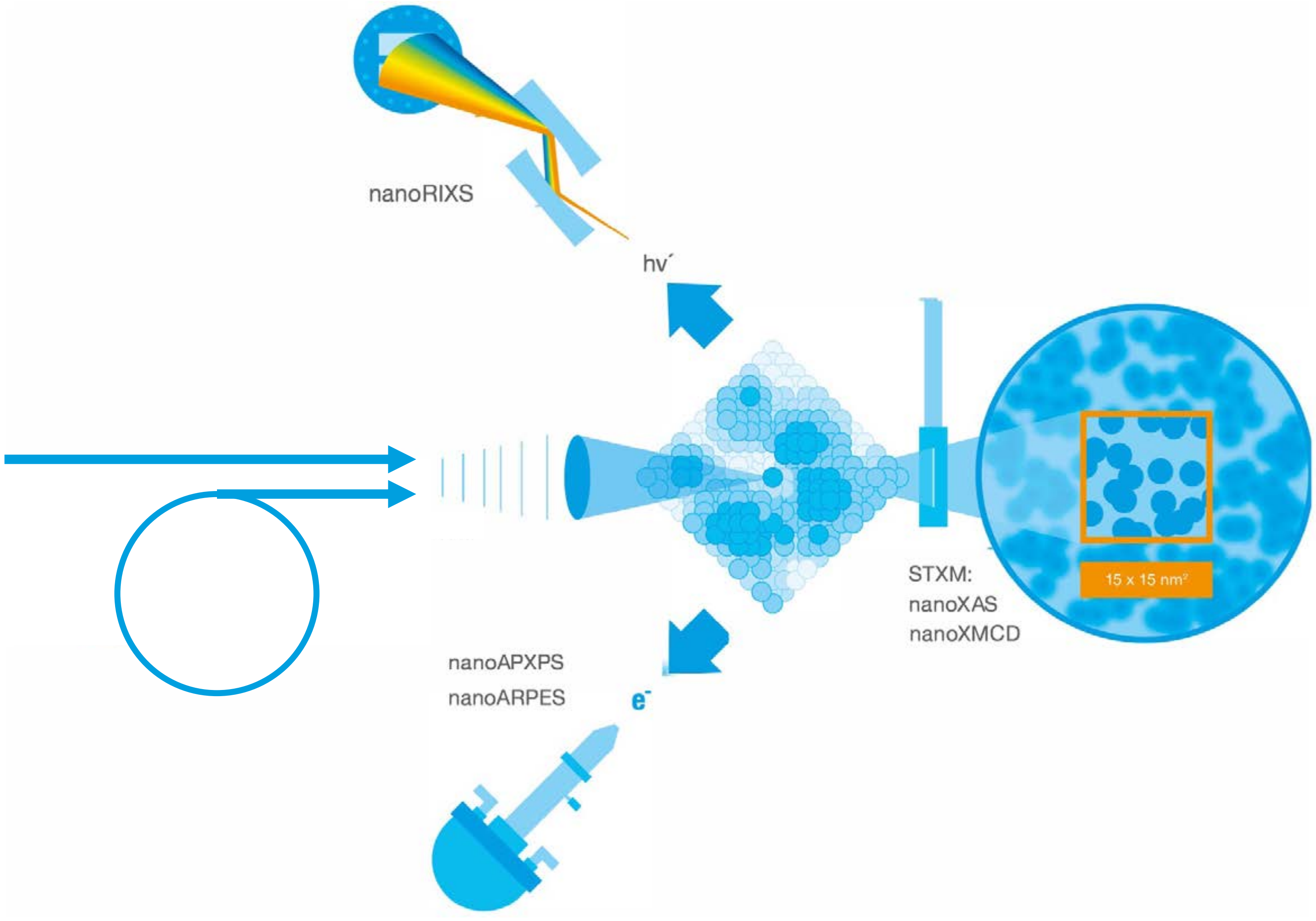
$$\tau_{ph} = \frac{h}{E_{ph}} = \mathcal{O}\left(\frac{h}{10\,\mathrm{meV}}\right) = \mathcal{O}(400\,\mathrm{fs})$$

1*T*-$TaSe_2$



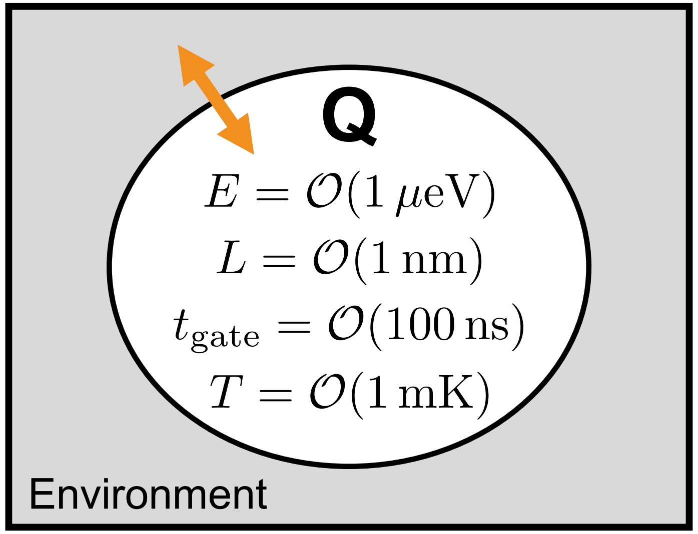
High-precision multimodal x-ray nano- and femtoanalytics

A good match to solid-state qubits?



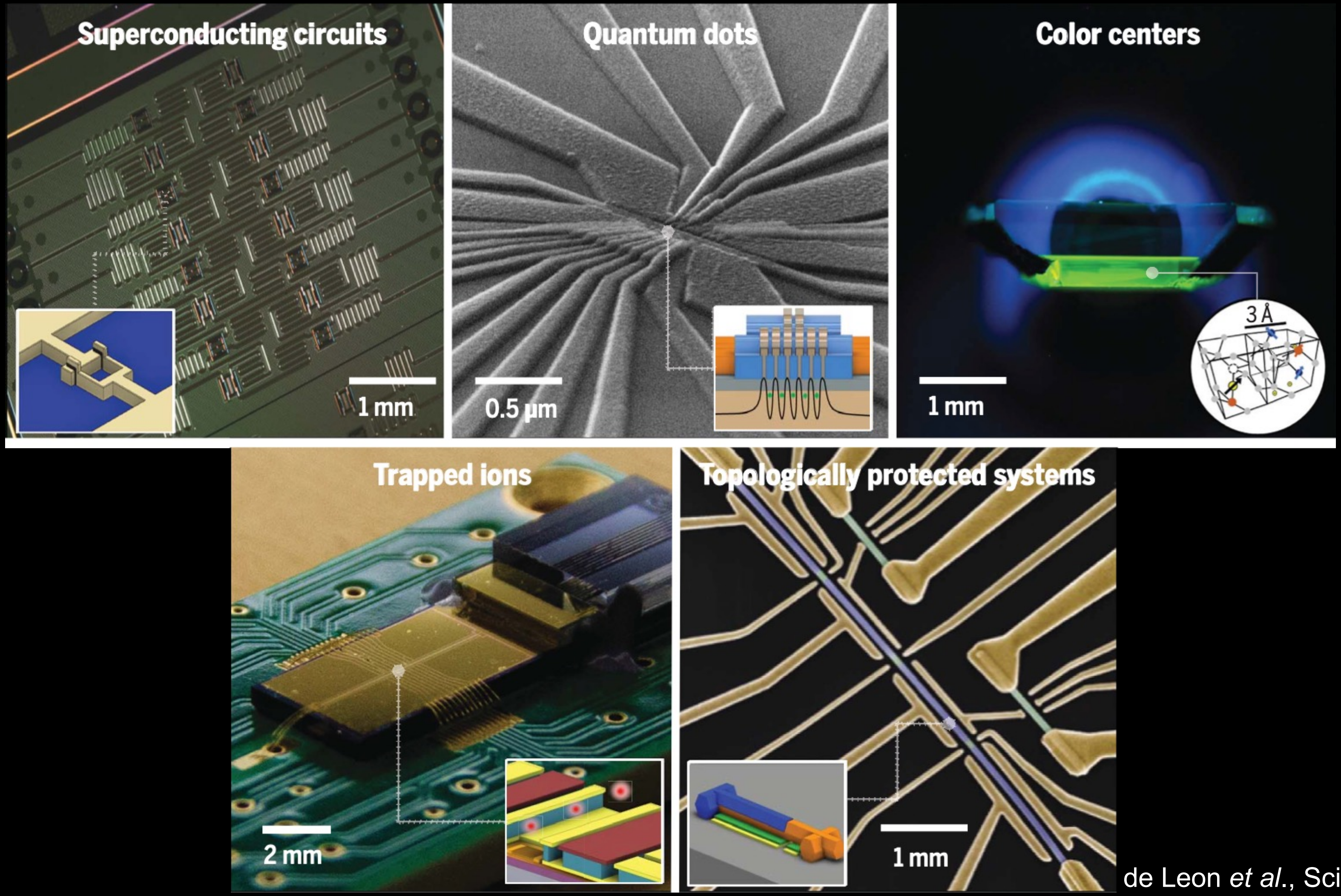
X

- Energy: ΔE ≳ 1 meV
- Momentum: Δk ≳ 0.005 Å^{-1}
- Position: Δx = Δy = Δz ≳ 10 nm
- Time: Δt ≳ 10 fs, 100 ps
- Angular momentum: spin & orbital



Part II: Possible applications

Needs of the quantum technology community?



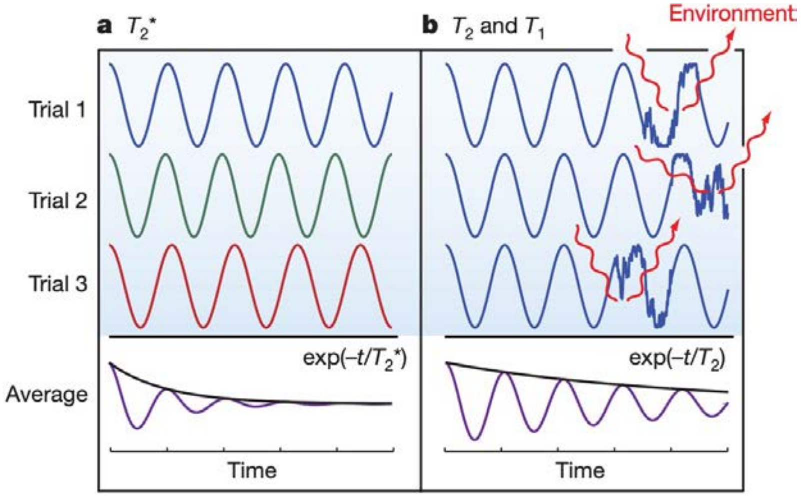
de Leon *et al.*, Science **327**, eabb2823 (2021)

Key challenge

Understand microscopic mechanisms for noise, loss & decoherence

Dephasing

Decoherence
Dissipation



- Coherence time T_2
- Dephasing time T_2*
- Relaxation time T_1

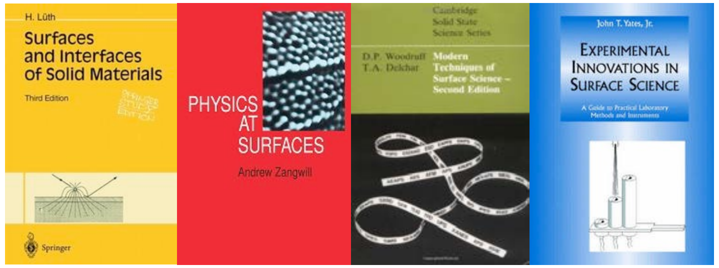
Ladd *et al.*, Nature **464**, 45(2010)

Correlate qubit measurements with direct materials characterization

Device operation & environment

- mK temperatures
- µeV energies
- nm length scales
- µs-to-s coherence times
- ultrahigh-vacuum conditions

Revival of Surface Science?
(Si ⟶ “semiconductor vacuum”)

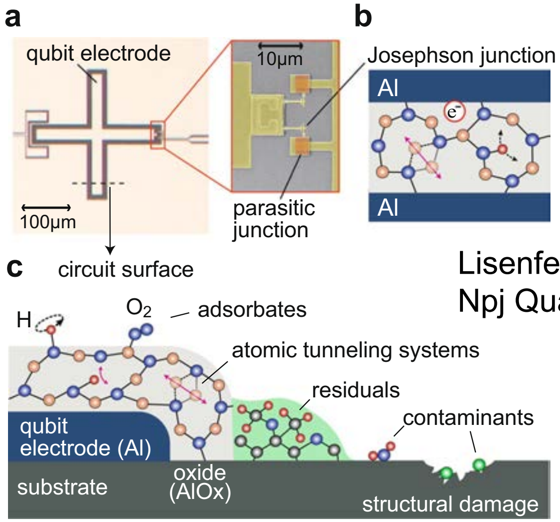


Sources of noise

- Inhomogeneity
- Defects & impurities
- Surfaces & interfaces
- Substrates
- Strain
- Oxides
- Charges
- Spins (electronic & nuclear)
- Quasiparticles
- Phonons

Superconducting qubits

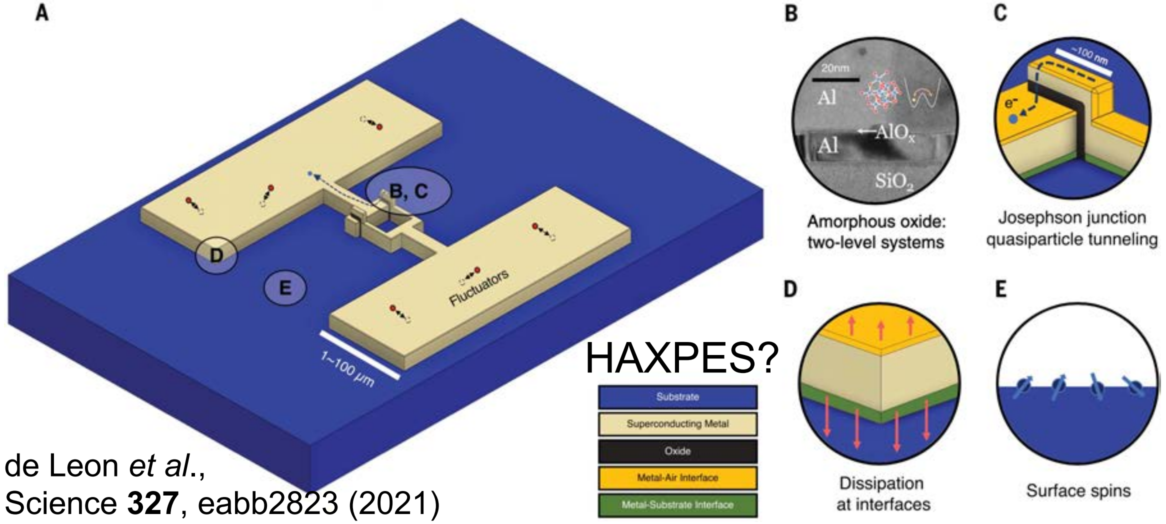
Quantum information stored in energy eigenstates of Josephson junction-based circuits



Lisenfeld *et al.*,
Npj Quantum Inf. **5**, 105 (2019)

XPS
SX-ARPES
?

femto-
ARPES
?

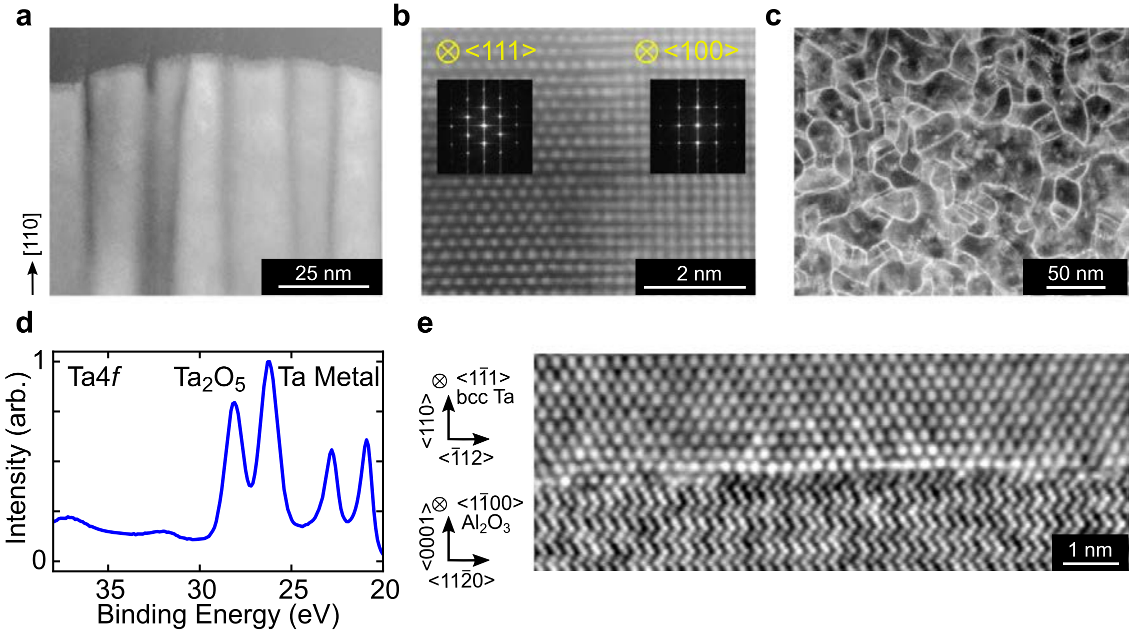


HAXPES?

XMCD, spin-ARPES?

de Leon *et al.*,
Science **327**, eabb2823 (2021)

STEM + XPS



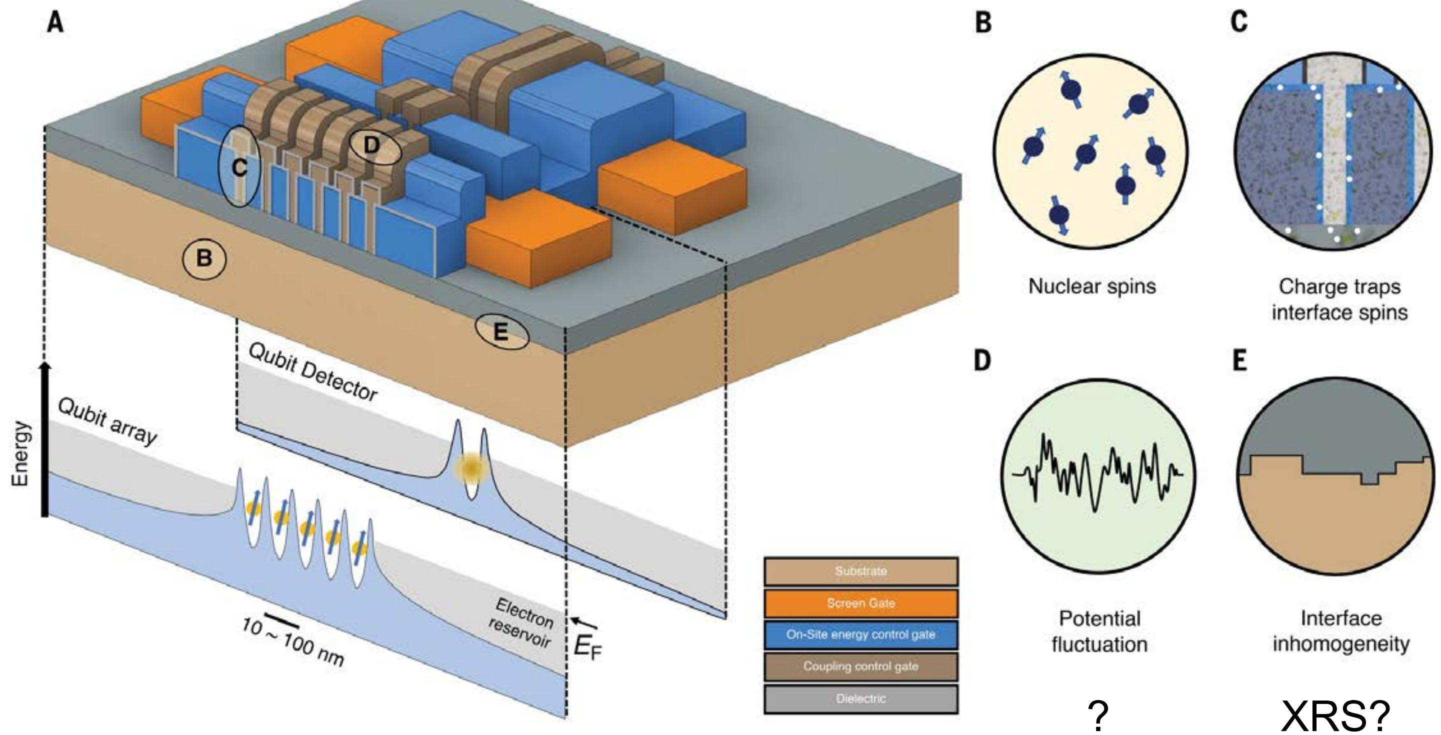
Place *et al.*, Nat. Commun. **12**, 1779 (2021)

Semiconductor quantum dot qubits

Quantum information stored in spin states of confined carriers

Nuclear resonant scattering?

SX-ARPES XMCD ?



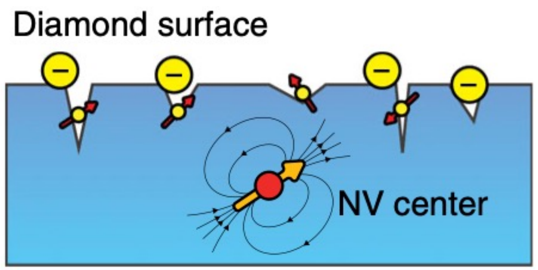
? XRS?

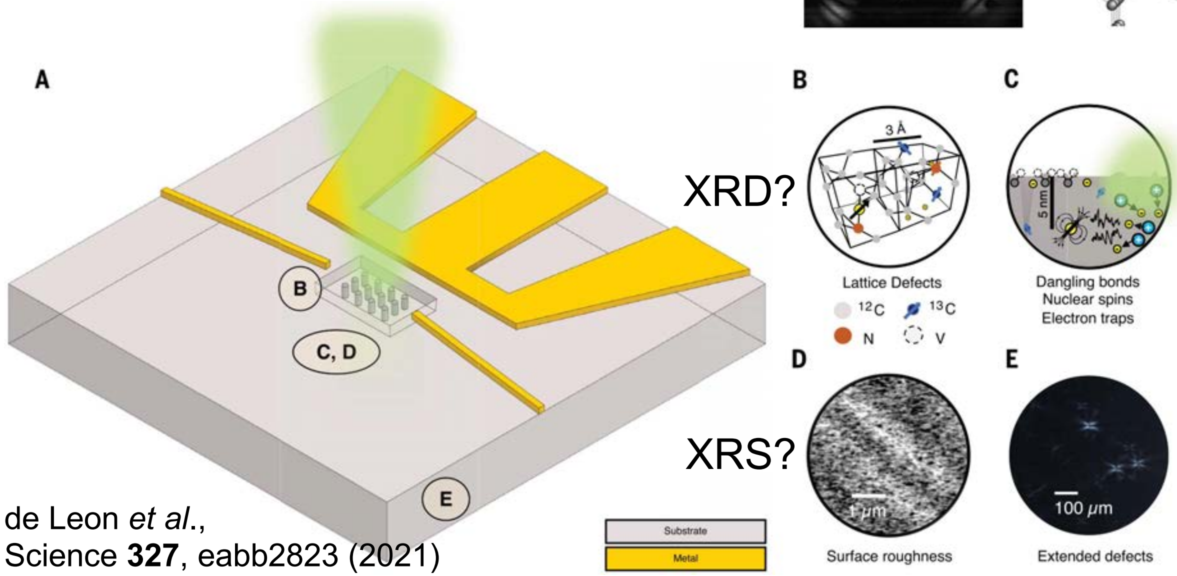
HAXPES?

de Leon *et al.*,
Science **327**, eabb2823 (2021)

Color center qubits

Quantum information stored in electronic orbital & spin states

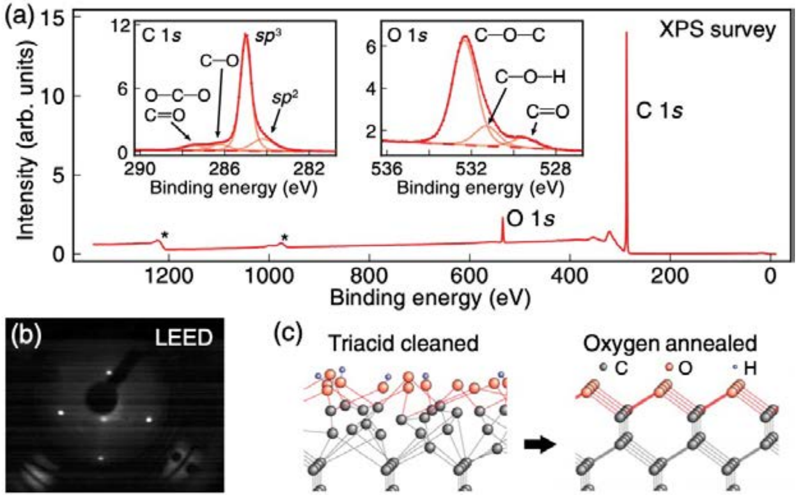




XRD?

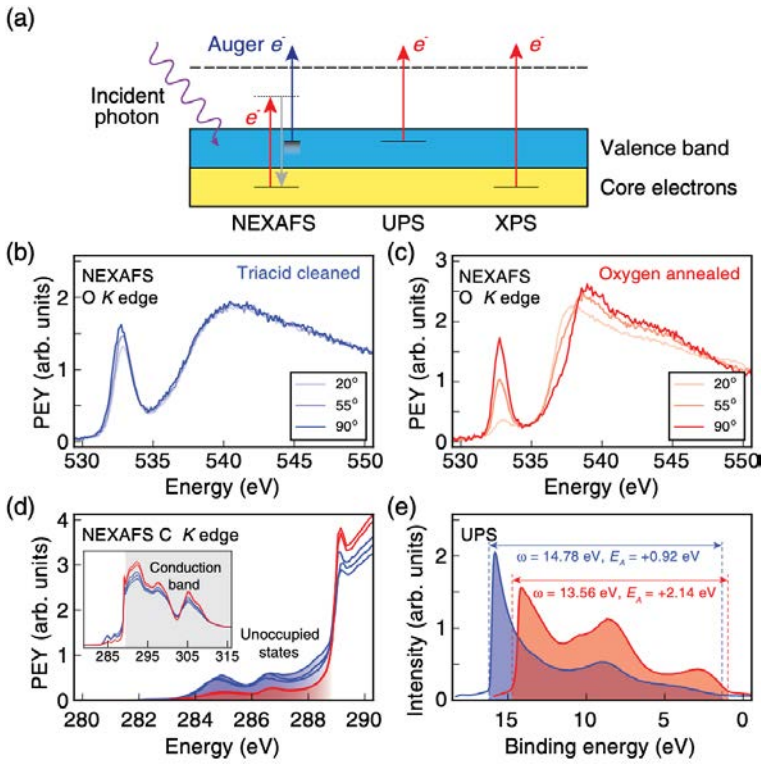
XRS?

de Leon *et al.*,
Science **327**, eabb2823 (2021)



NEXAFS
XPS
ARPES
Nuclear resonant scattering?

CDI?
Ptychography?

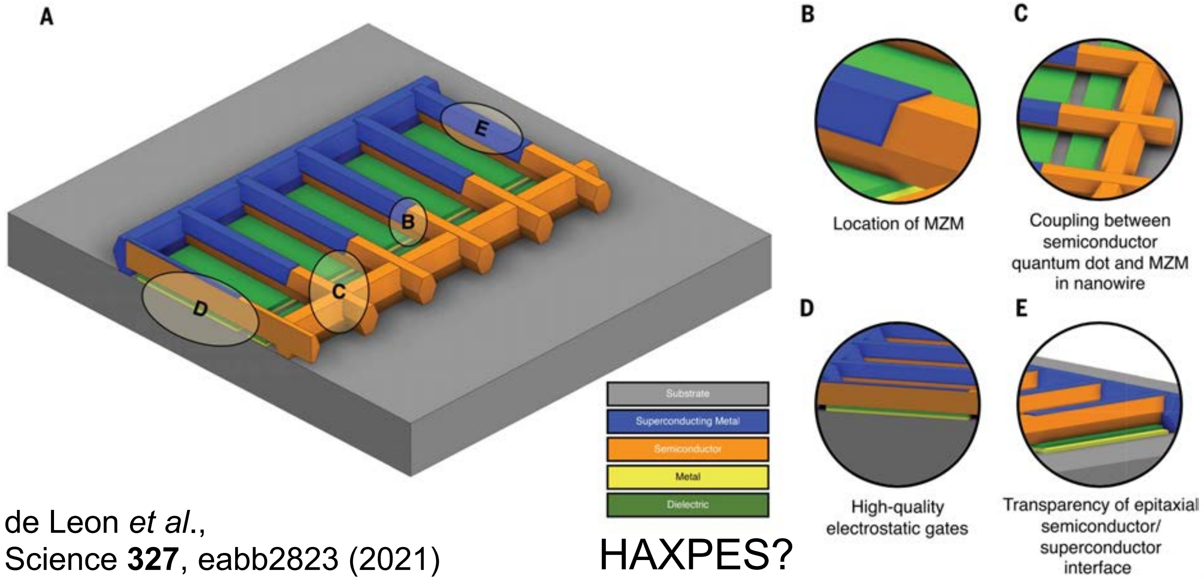


Sangtawesin *et al.*, Phys. Rev. X **9**, 031052 (2019)

Topological qubits

Quantum information stored in non-Abelian topological phase of Majorana zero modes

Where is the Majorana zero mode?
Does it obey non-Abelian statistics?

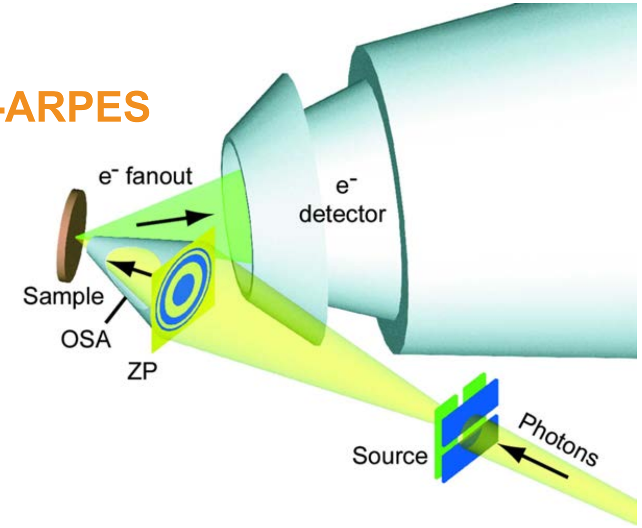


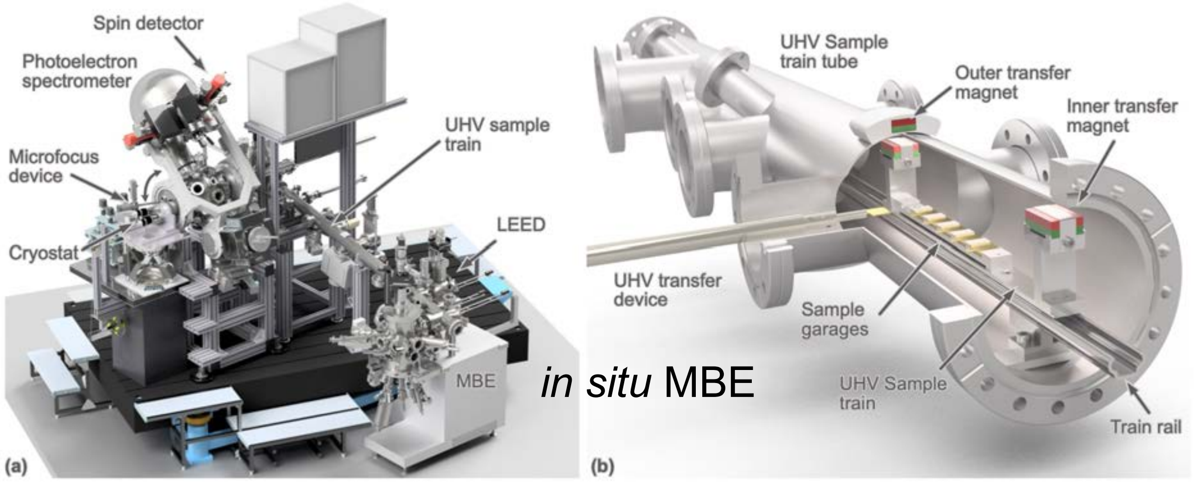
de Leon *et al.*,
Science **327**, eabb2823 (2021)

HAXPES?

Exploratory platform: no physical qubit yet

nano-ARPES

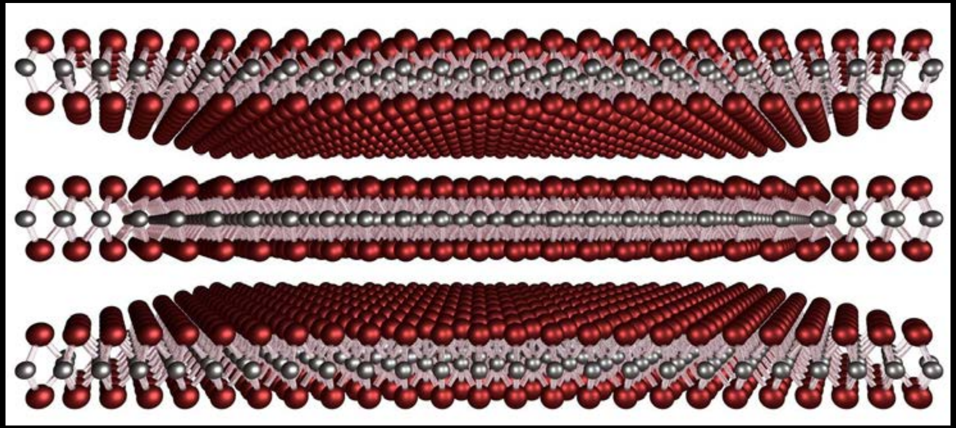


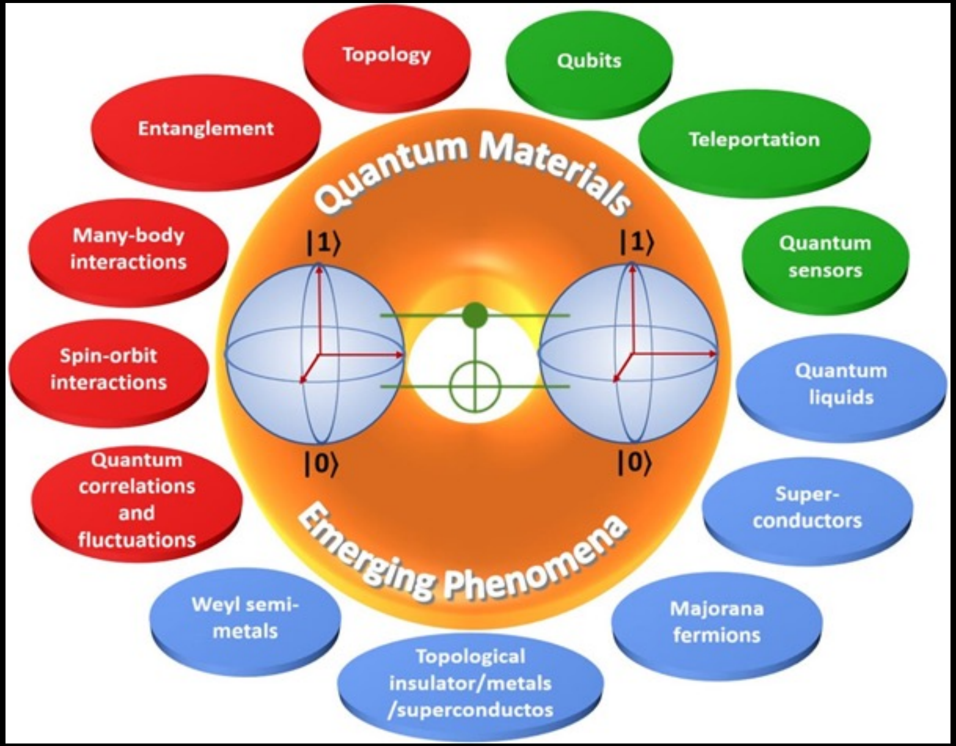


in situ MBE

Part III: 2D quantum materials

Viable quantum technology platform?

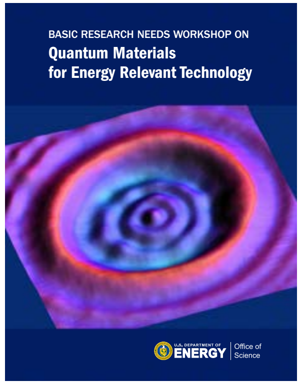




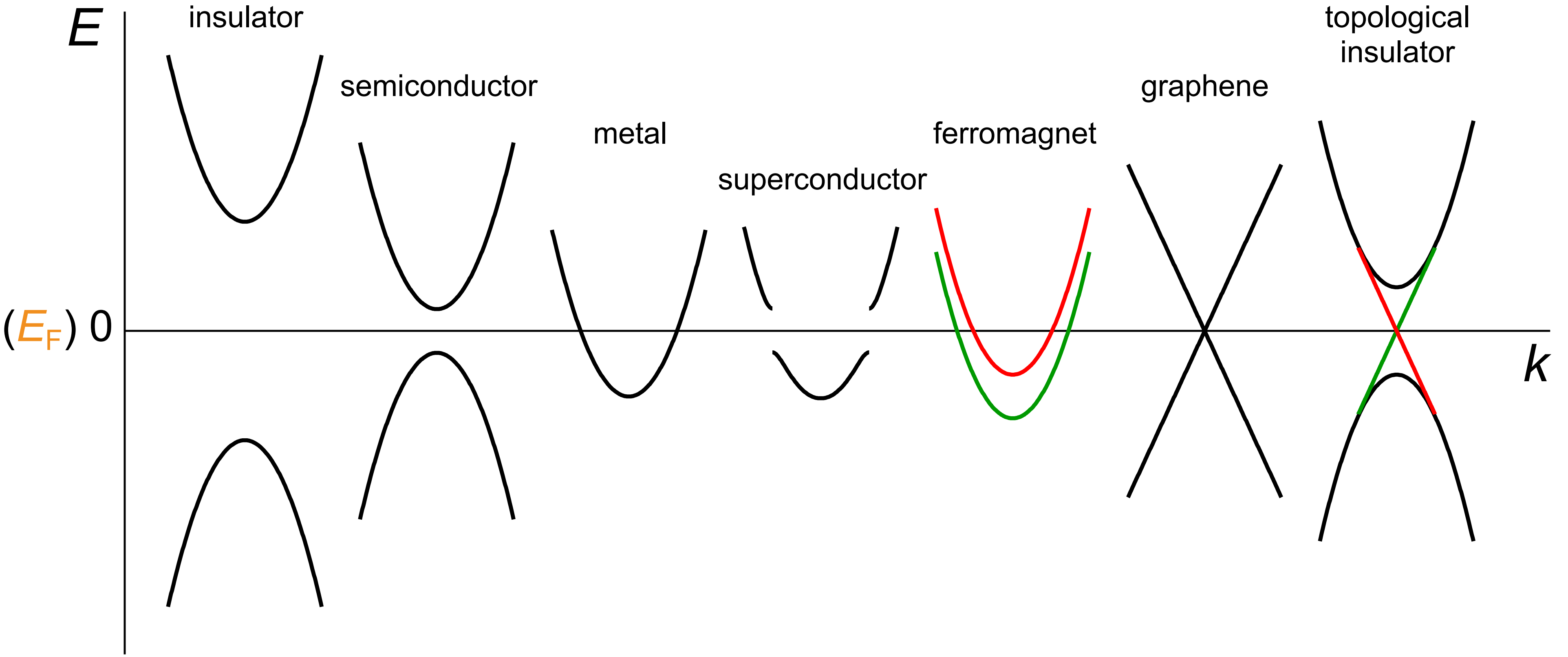
iopscience.iop.org/journal/0022-3727/page/
special-issue-on-quantum-materials-and-emerging-phenomena

Quantum materials

Quantum materials? All materials are quantum.



… solids with **exotic** physical properties, arising from the **quantum mechanical** properties of their constituent **electrons**; such materials have great scientific and/or technological potential.



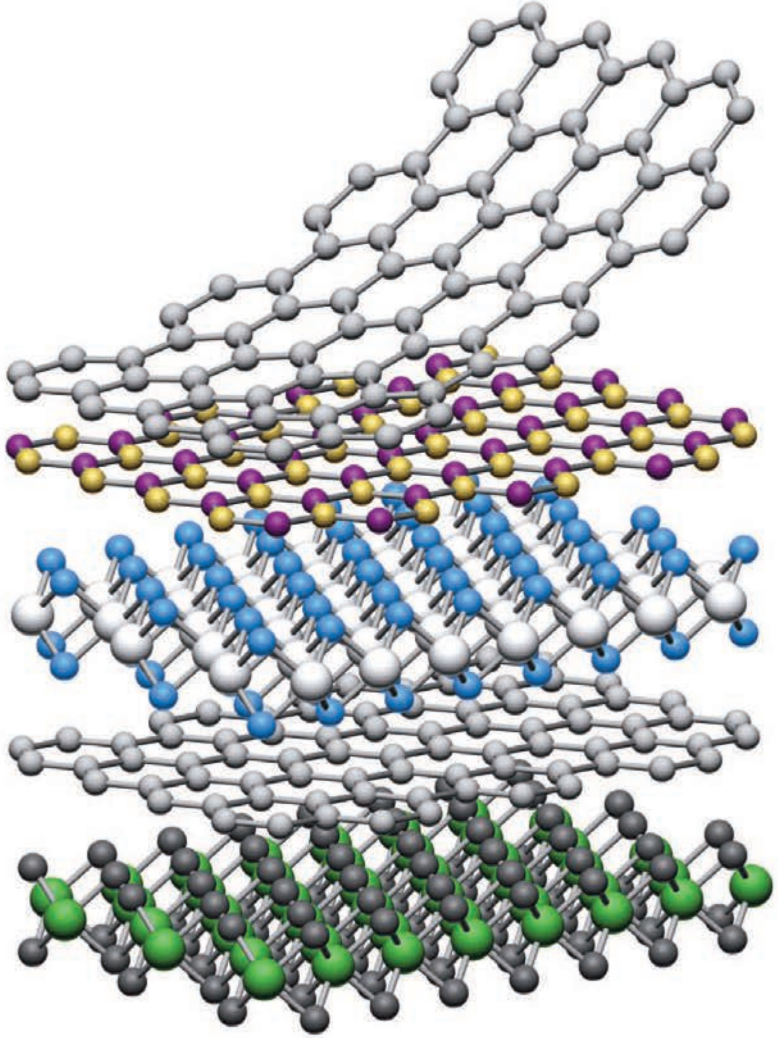
Van der Waals heterostructures

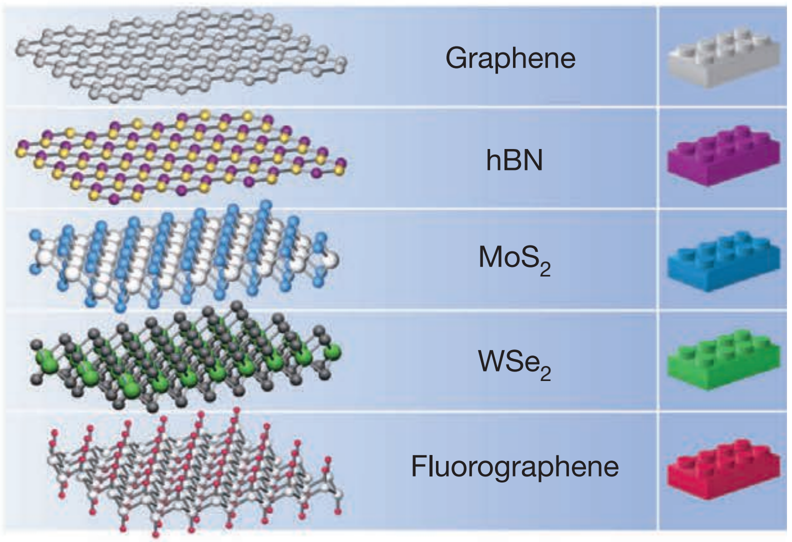
“What if we mimic layered superconductors by atomic-scale LEGO?”

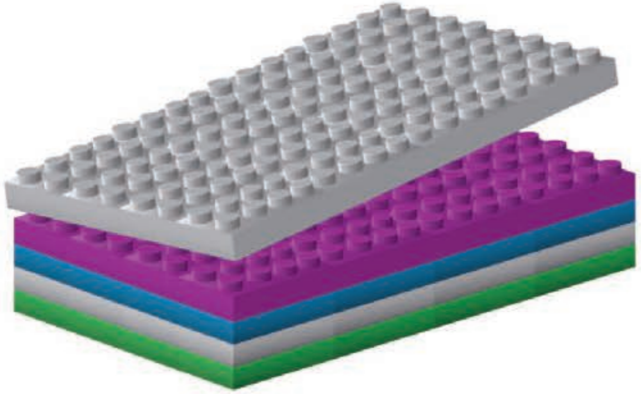
clean, strain-free
atomically sharp interfaces

(gate) tunable, emergent
electronic properties

new kinds of
devices & **electronics**



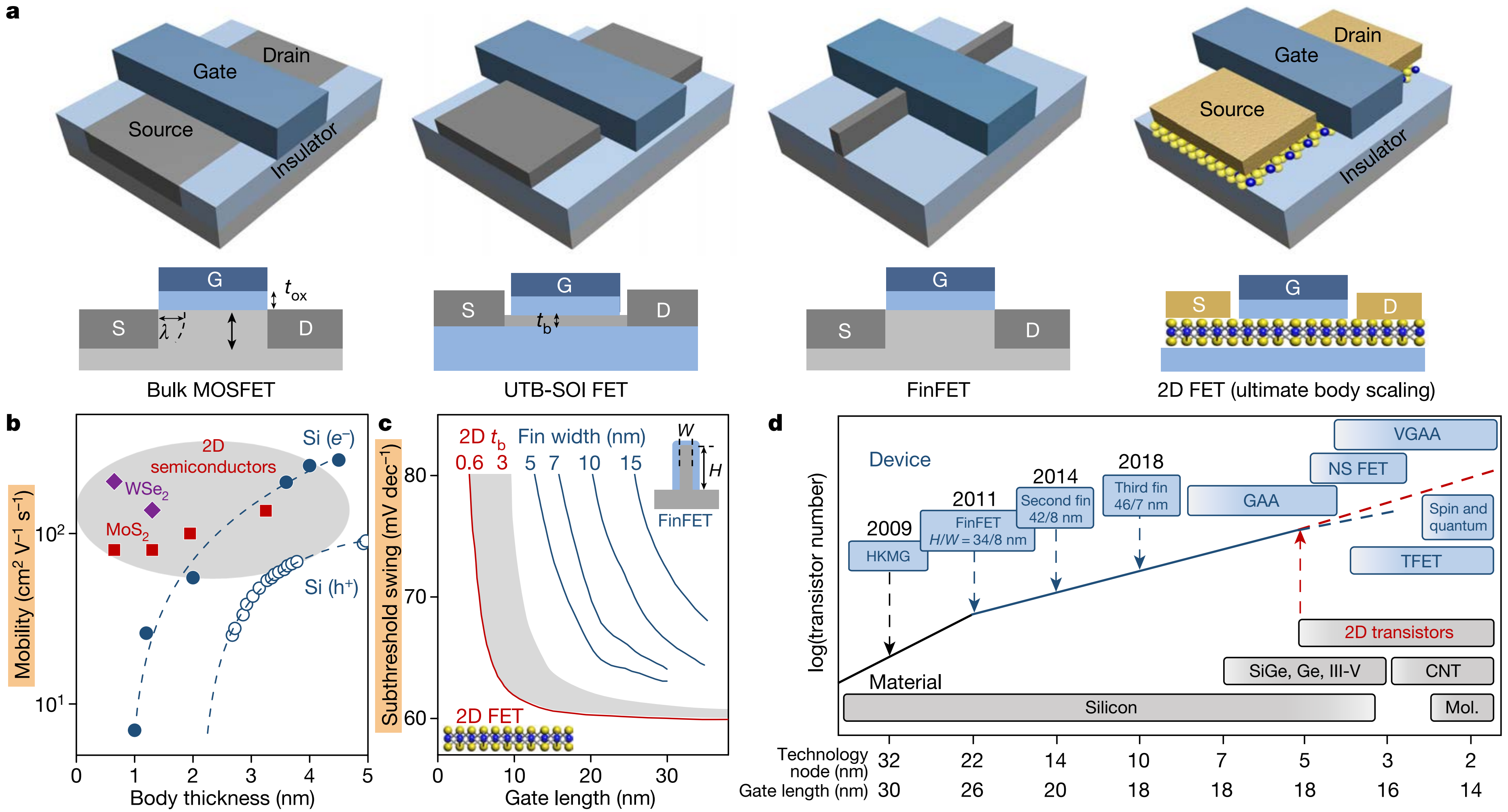




Geim & Grigorieva, Nature **499**, 419 (2013)

2D transistors

Toward ultimate transistor scaling



Liu *et al.*,
Nature **591**, 43 (2021)

Twistronics

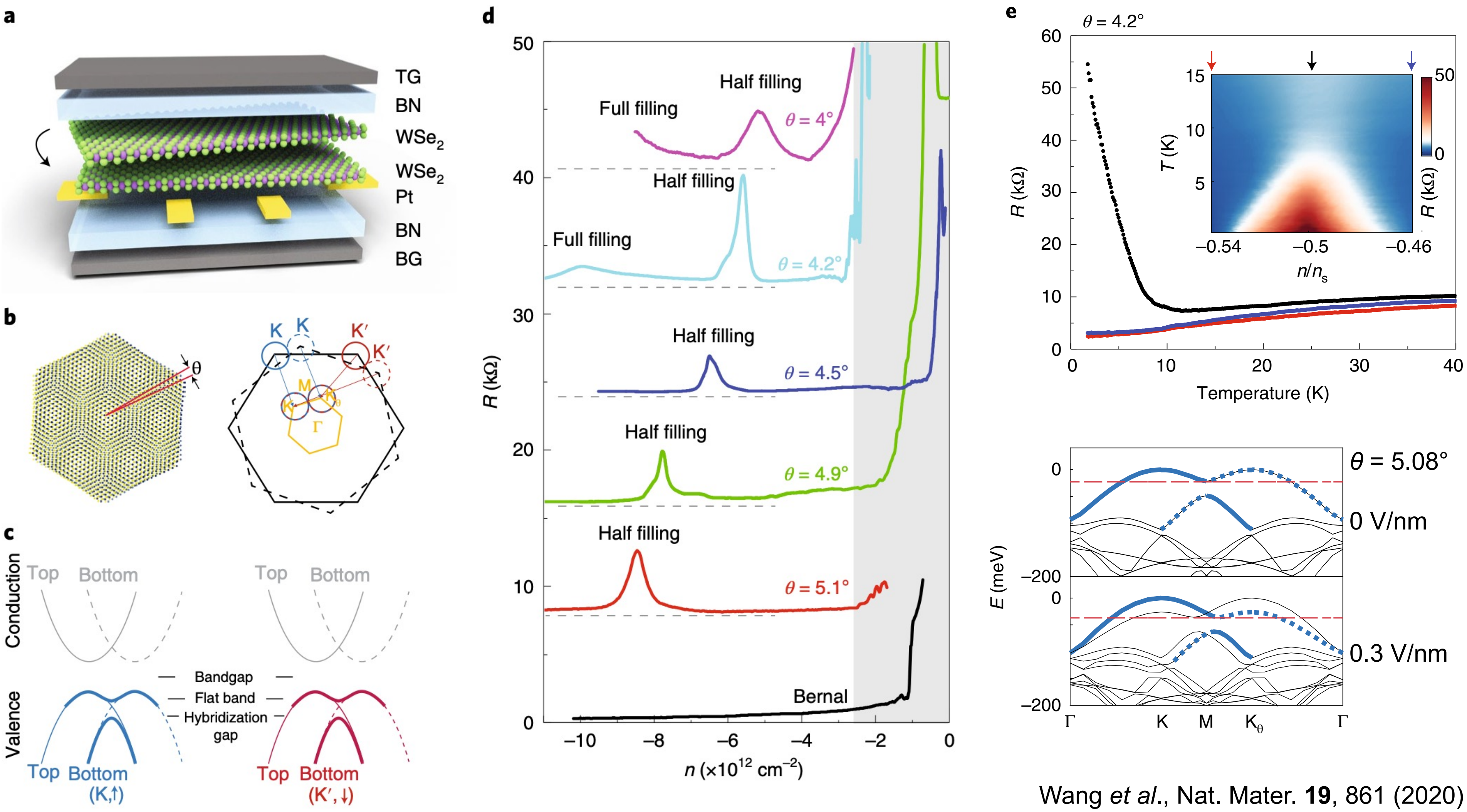
Twisted bilayer WSe_2: Flat band via Moiré superlattice

Bandwidth control
(via **twist** angle
& gate voltages)

Carrier density control
(via gate voltages)

Moiré superlattice

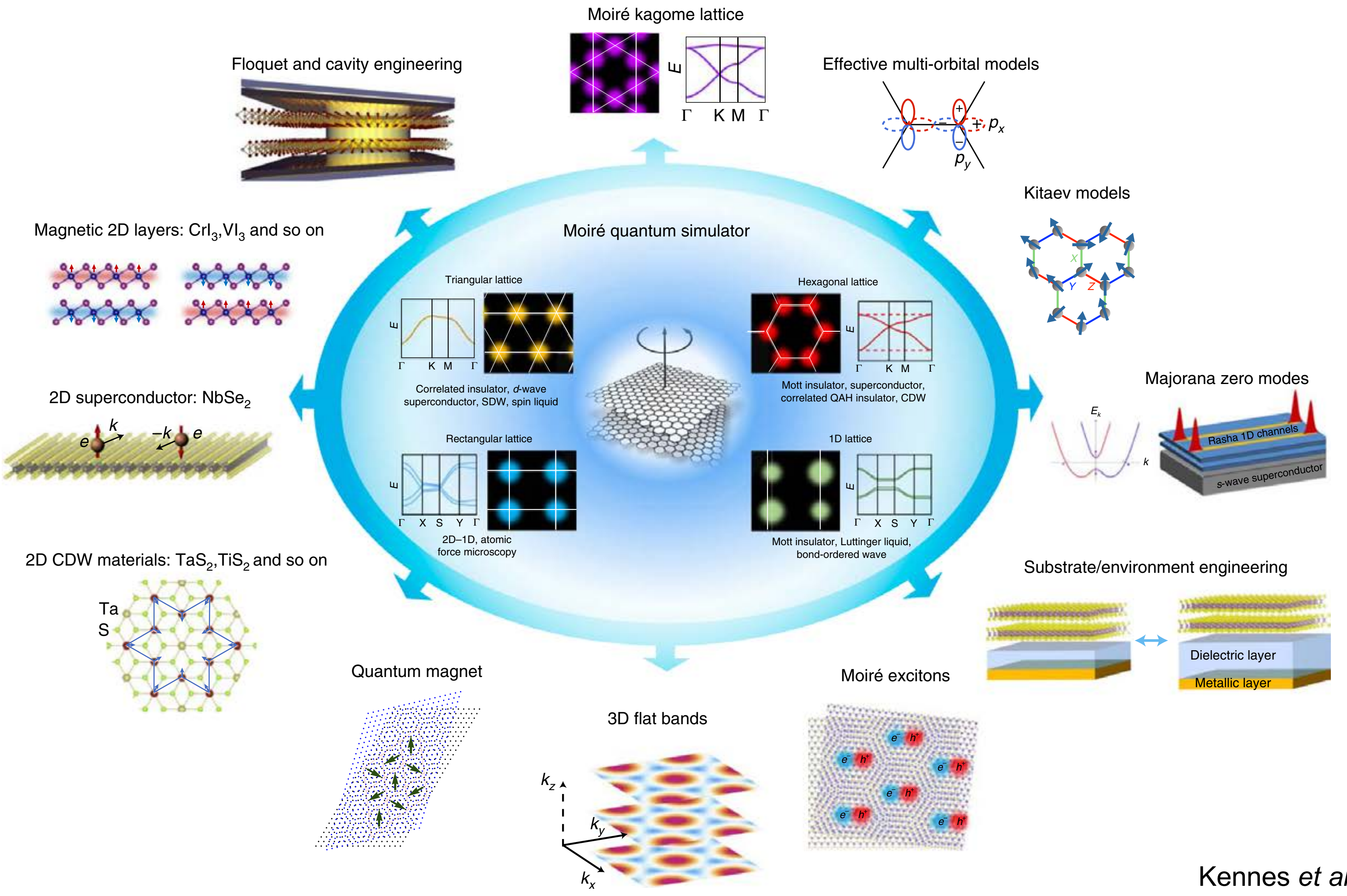
Flat band
(pseudo-spin degenerate,
$2e^-$ per 126···205 W atoms)



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

Quantum simulation: Moiré quantum matter

Controllable quantum Hamiltonians realized by twisted van der Waals heterostructures

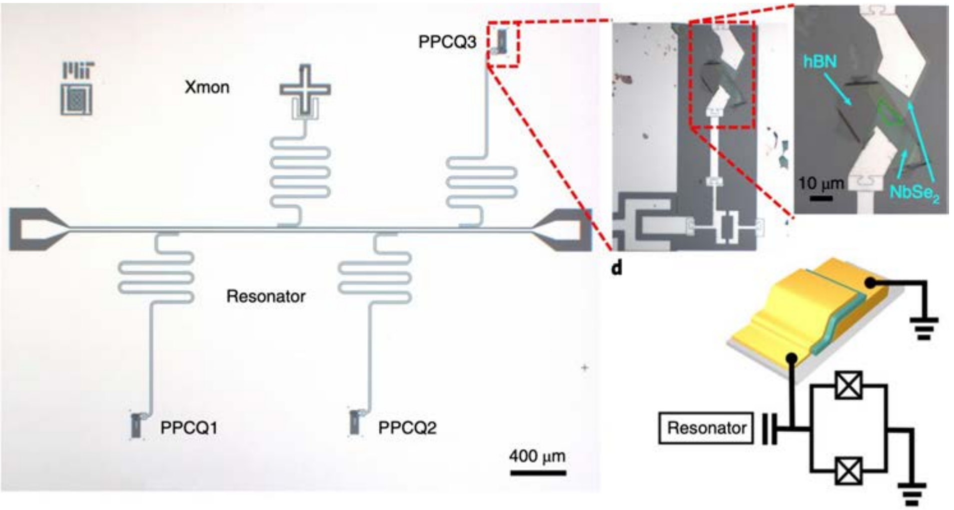


Kennes *et al.*, Nat. Phys. **17**, 155 (2021)

2D materials for quantum technology hardware

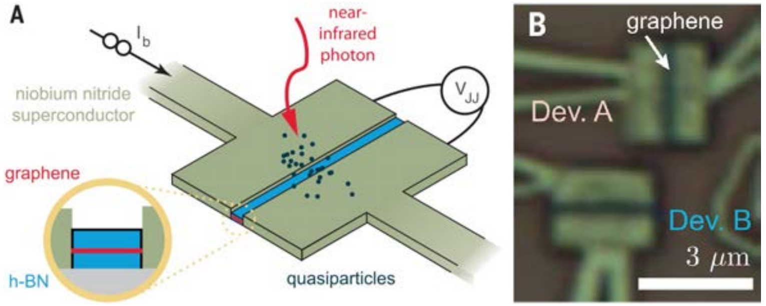
Quantum computing, communication & sensing

Ultrasmall capacitor
for superconducting qubits

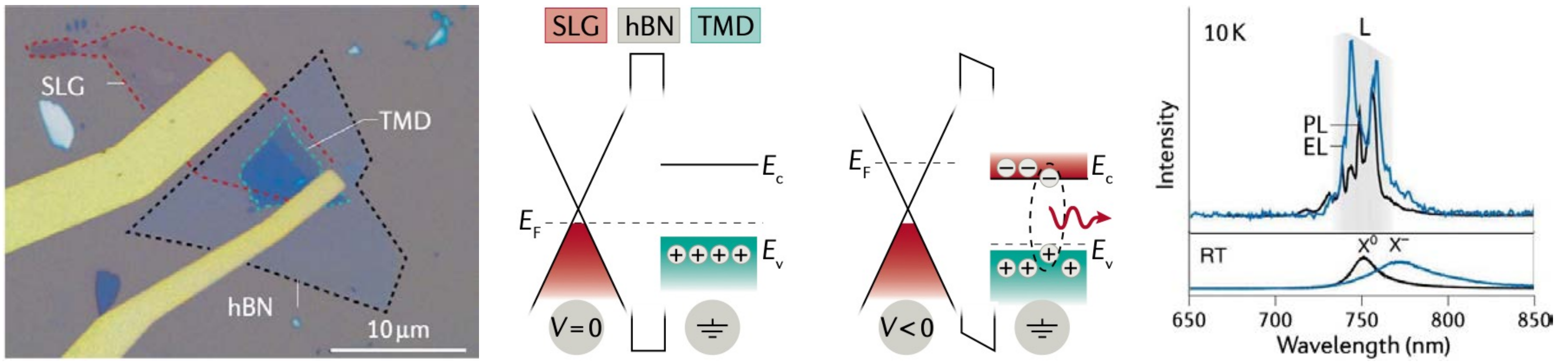


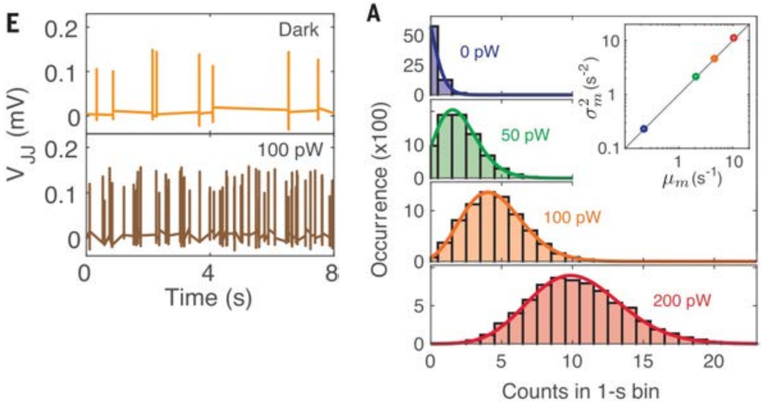
Wang *et al.*,
Nat. Mater. **21**, 398 (2022)

Single-photon detector



Single-photon LED



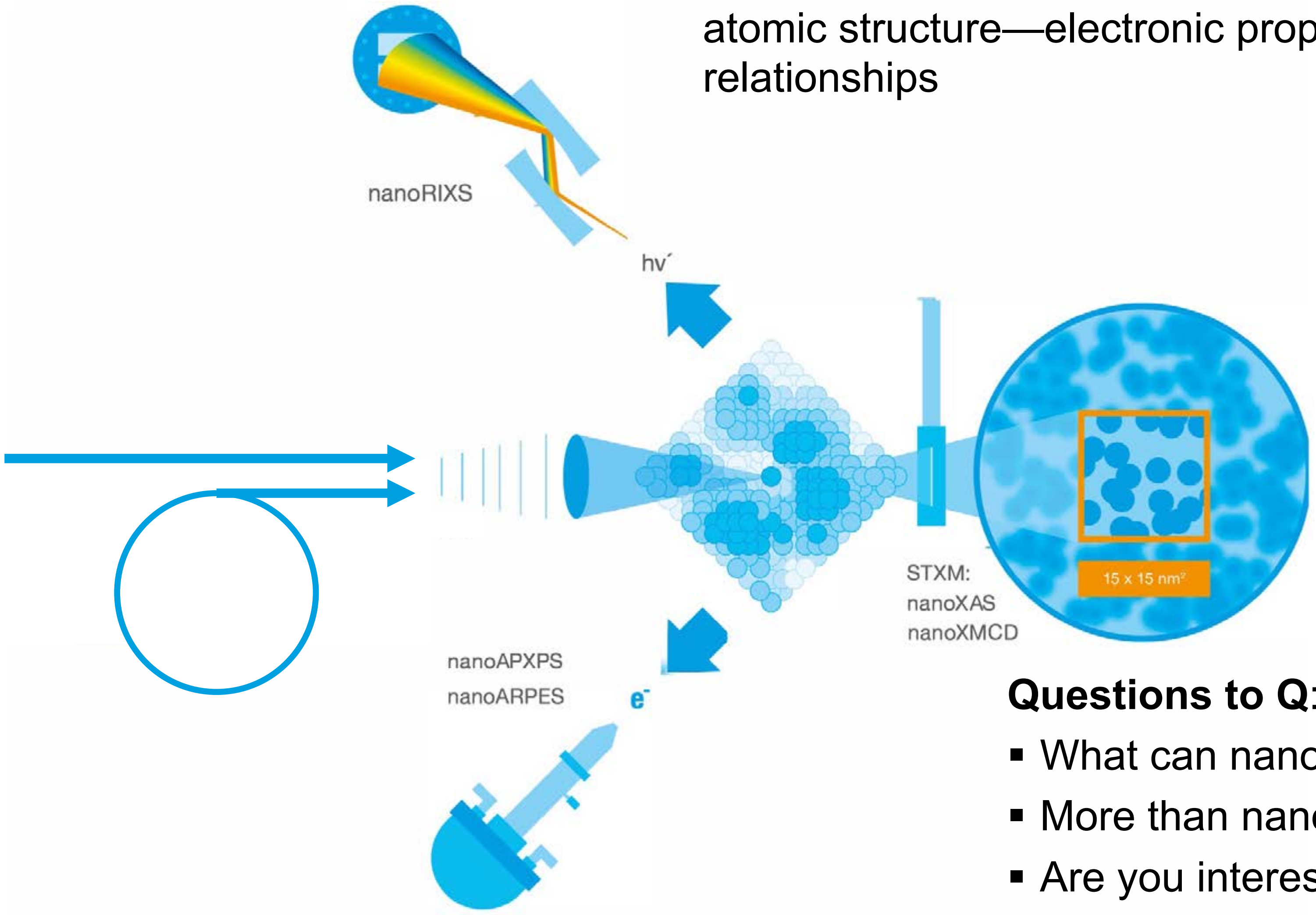


Turunen *et al.*,
Nat. Rev. Phys. **4**, 219 (2022)

Walsh *et al.*, Science **372**, 409 (2021)

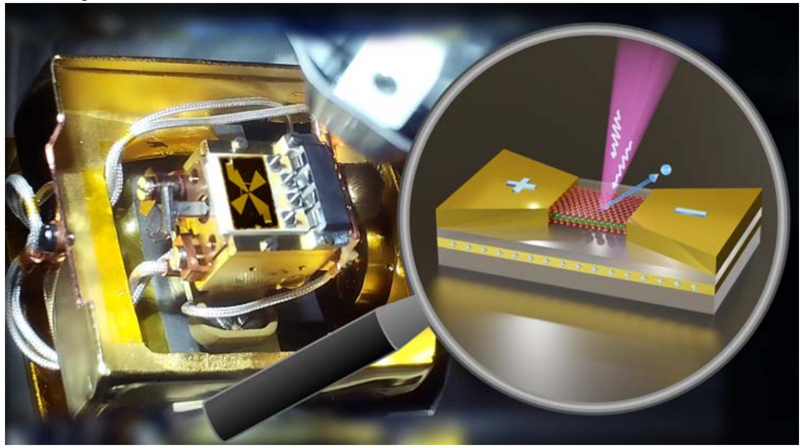
X meets Q

High-precision multimodal x-ray nano- & femtoanalytics for …



atomic structure—electronic properties (charge, orbital, spin)—function relationships

Quantum materials & devices
in operando

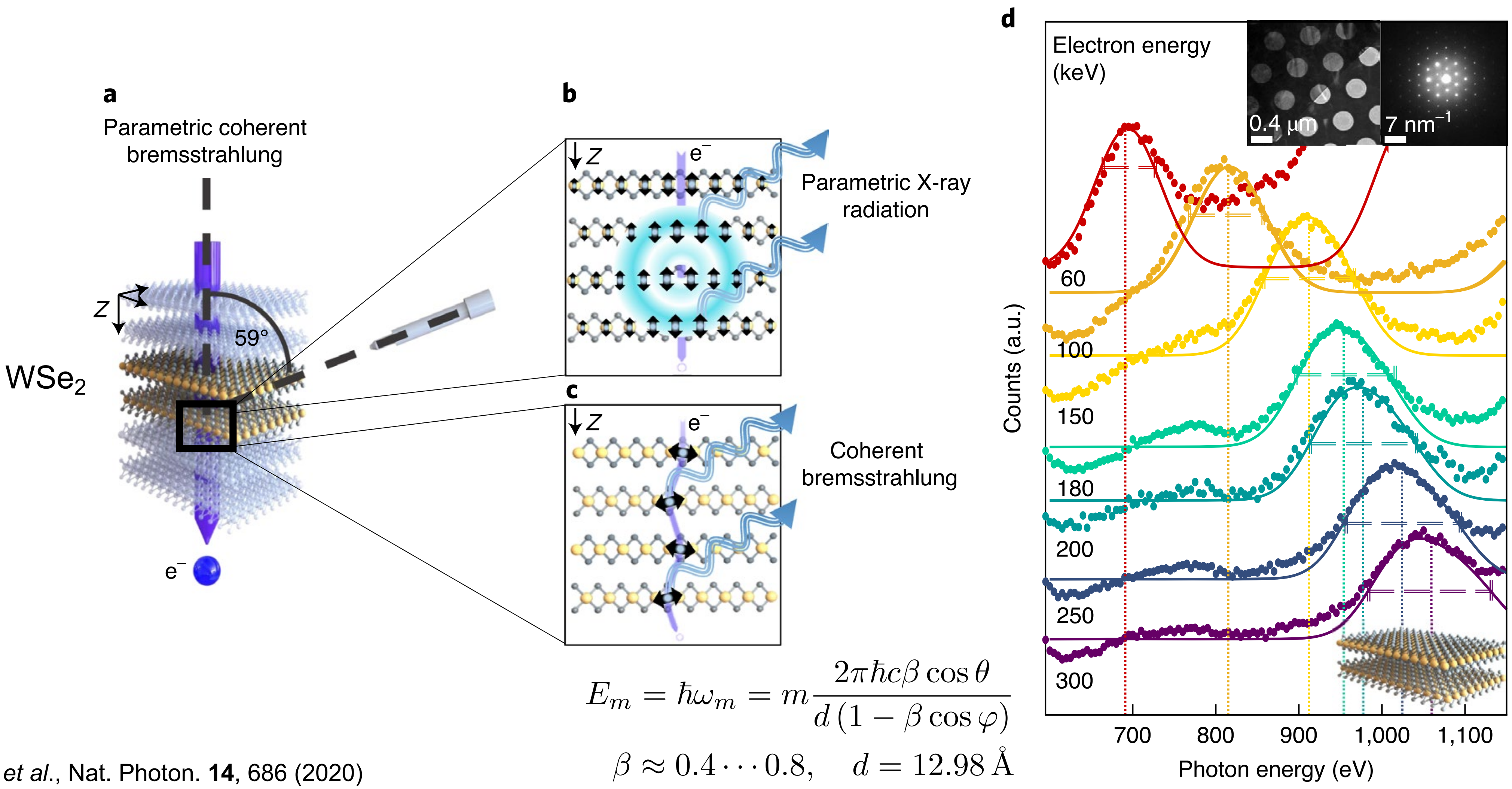


Questions to Q:

- What can nano-X & femto-X do for you?
- More than nanochemical analysis?
- Are you interested in new materials?

Free-electron x-ray radiation from WSe_2

Electrons in — x-rays out



$$E_m = \hbar\omega_m = m\frac{2\pi\hbar c\beta\cos\theta}{d\,(1-\beta\cos\varphi)}$$

$$\beta \approx 0.4\cdots 0.8, \quad d = 12.98\ \text{Å}$$

Shentcis *et al.*, Nat. Photon. **14**, 686 (2020)

We welcome collaborations

Soft x-rays @ DESY/EuXFEL + TMDC crystals

kai.rossnagel@desy.de