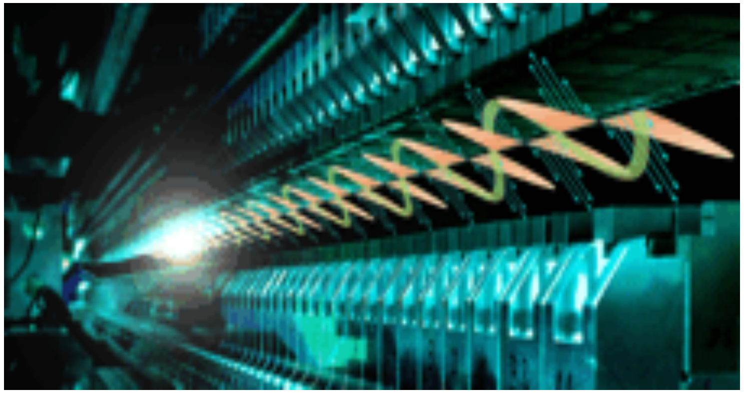
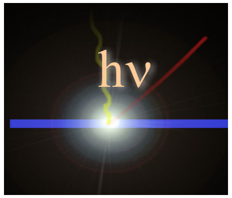
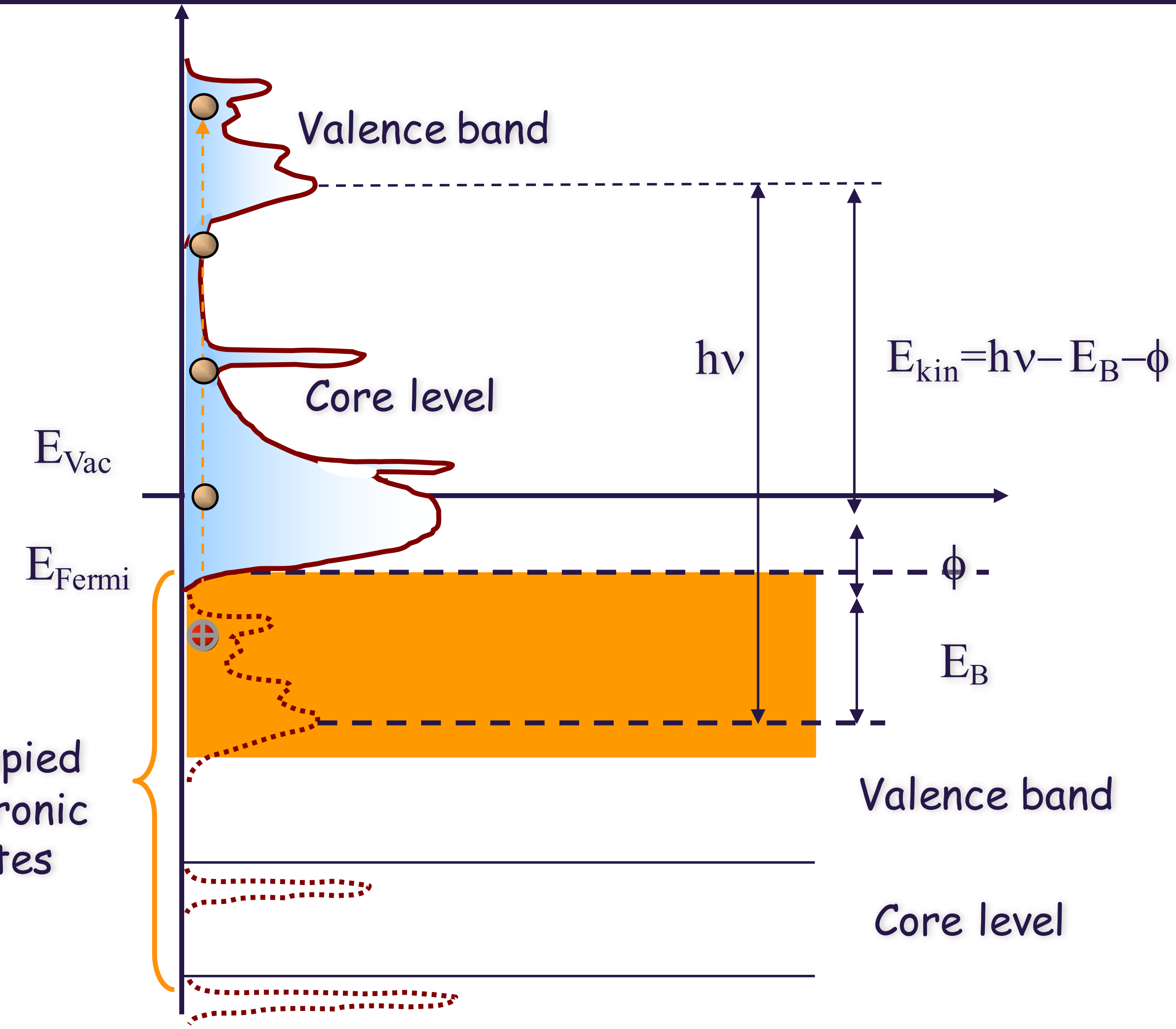
New Opportunities for Spectroscopy (PES) Opened by High-Repetition Rate XFELs

S.L. Molodtsov
European XFEL GmbH

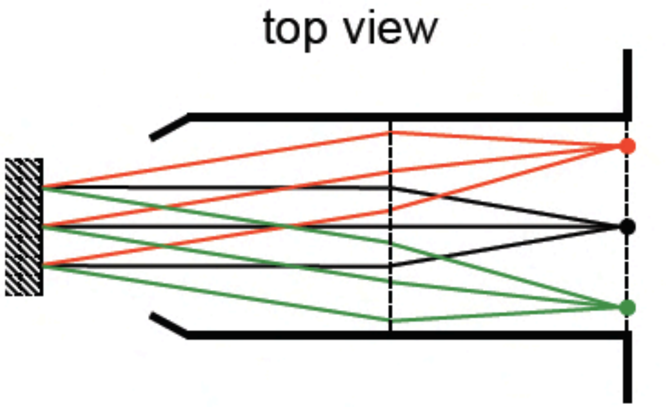


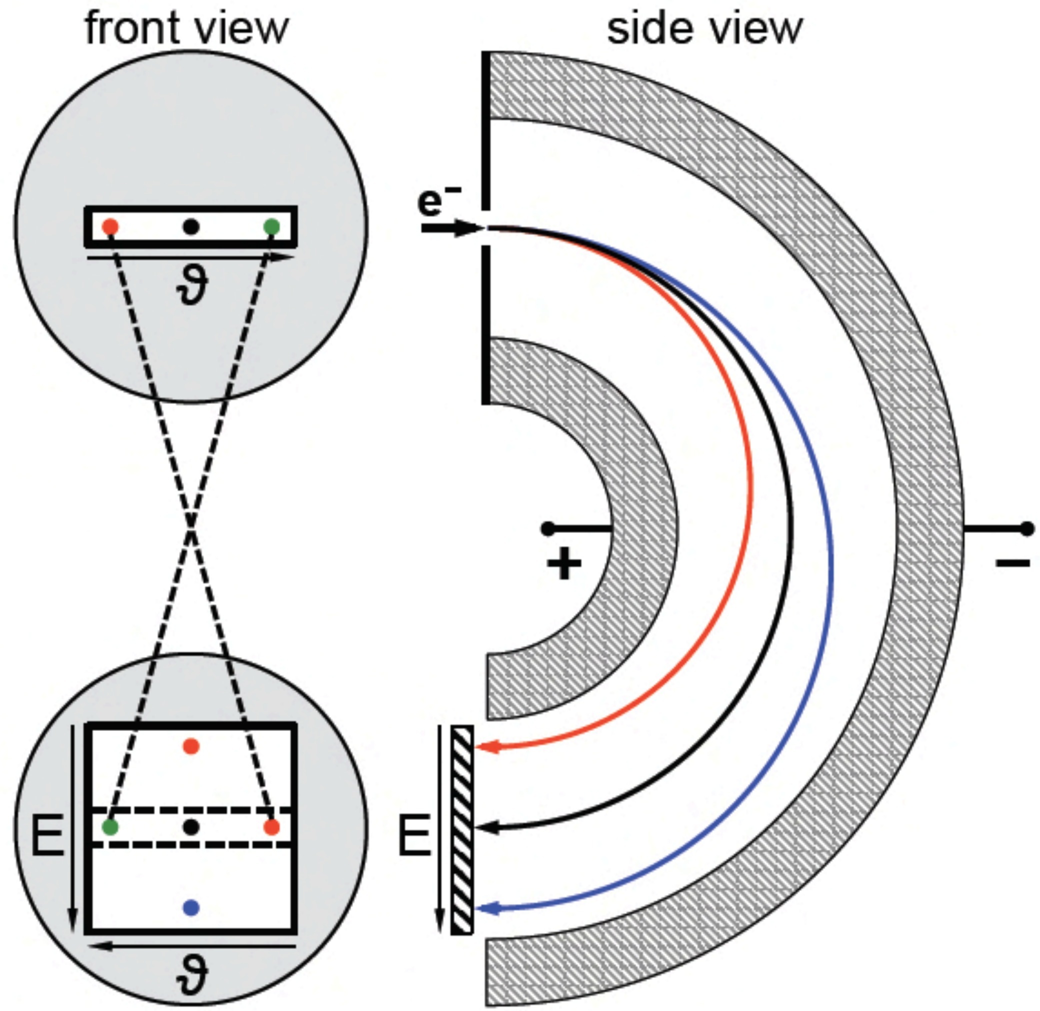
Photoemission spectroscopy: Basics

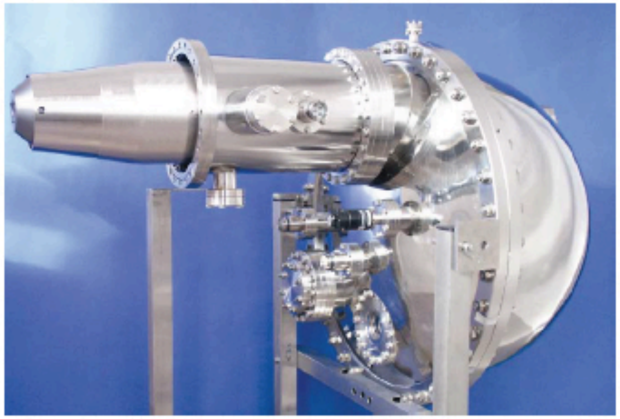




Photoelectron analyzer

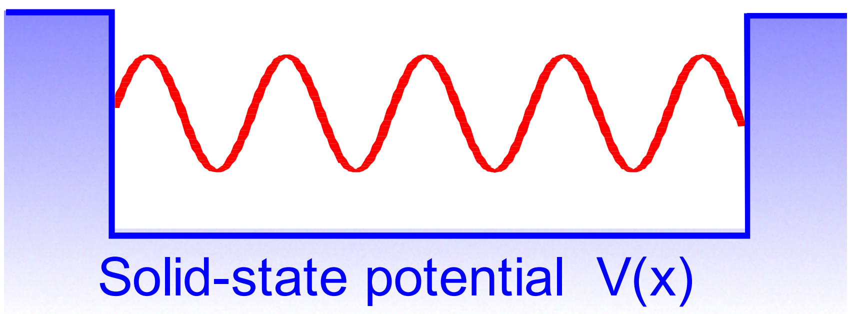




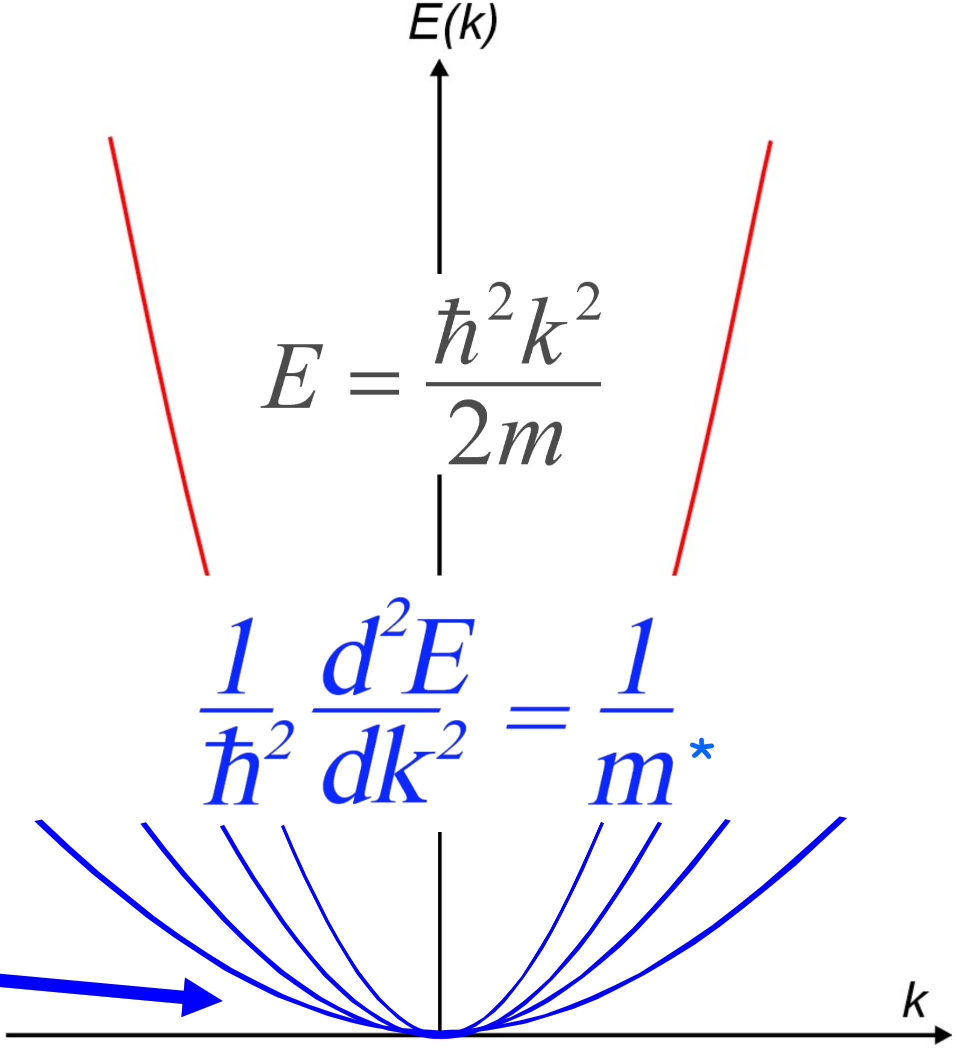


Single-particle system

1 - dim. solid state:



Heavy-fermion system

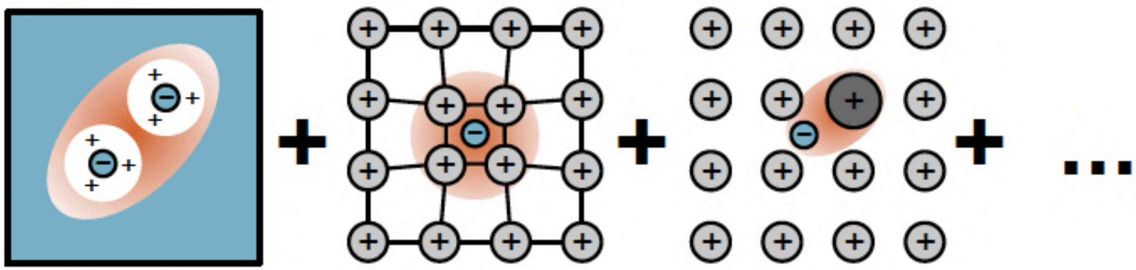


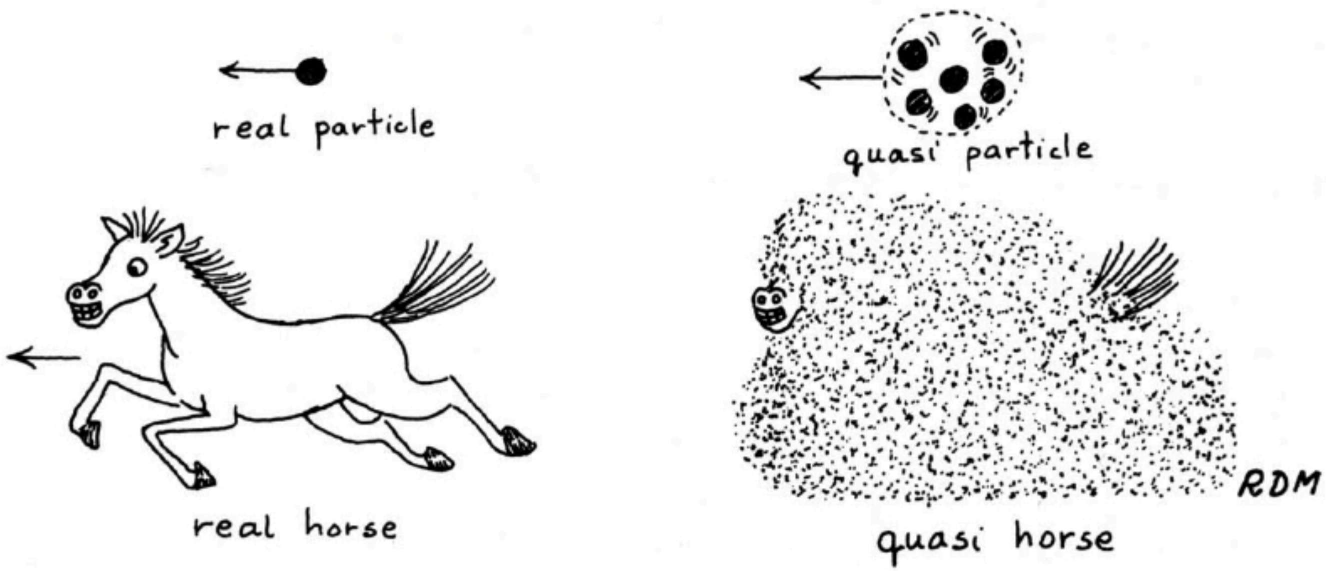
$$E = \frac{\hbar^2 k^2}{2m}$$

$$\frac{1}{\hbar^2}\frac{d^2E}{dk^2} = \frac{1}{m^*}$$

Quasiparticles properties (Kai Rossnagel)

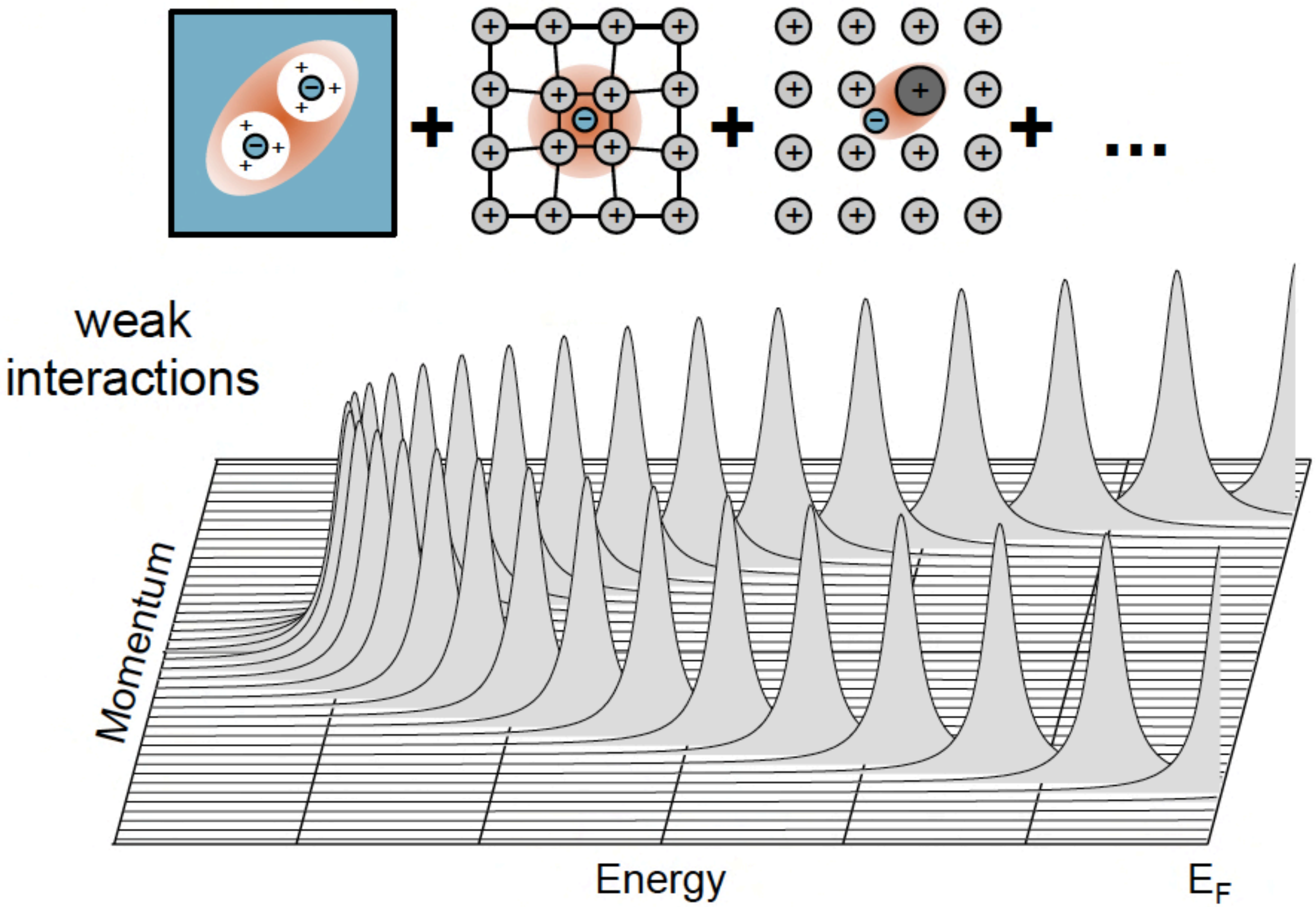
Lead actor: The quasi-electron





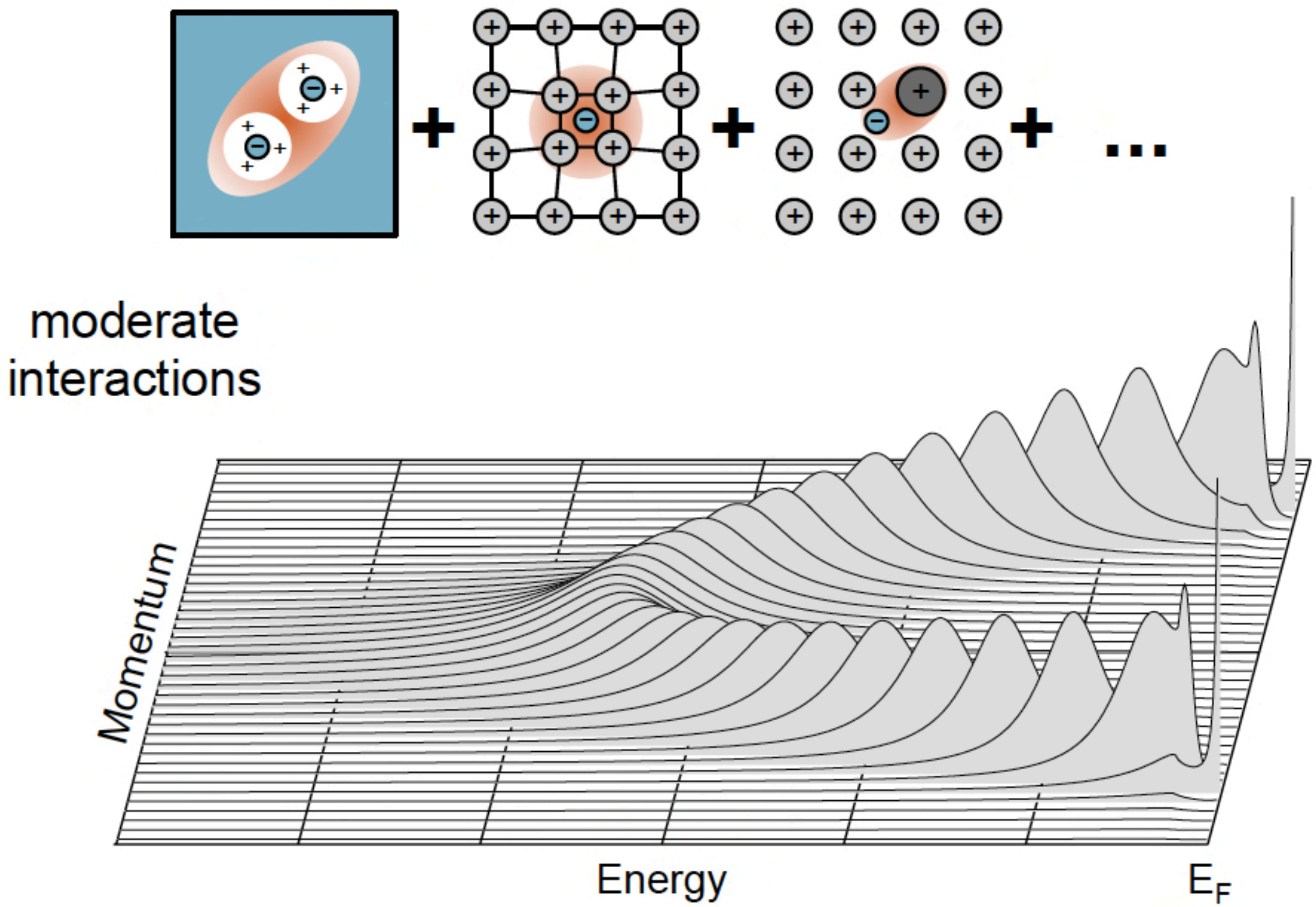
Weak correlations

Lead actor: The quasi-electron



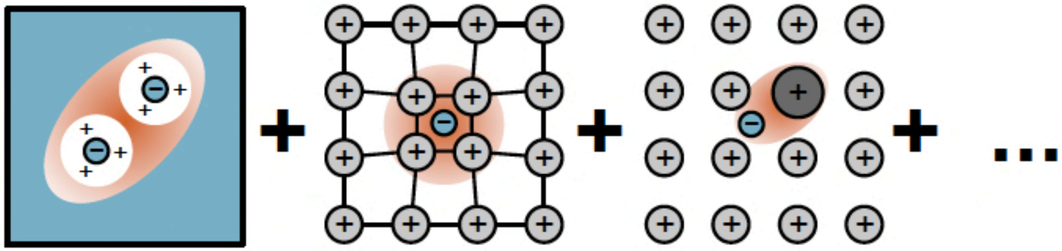
Moderate correlations

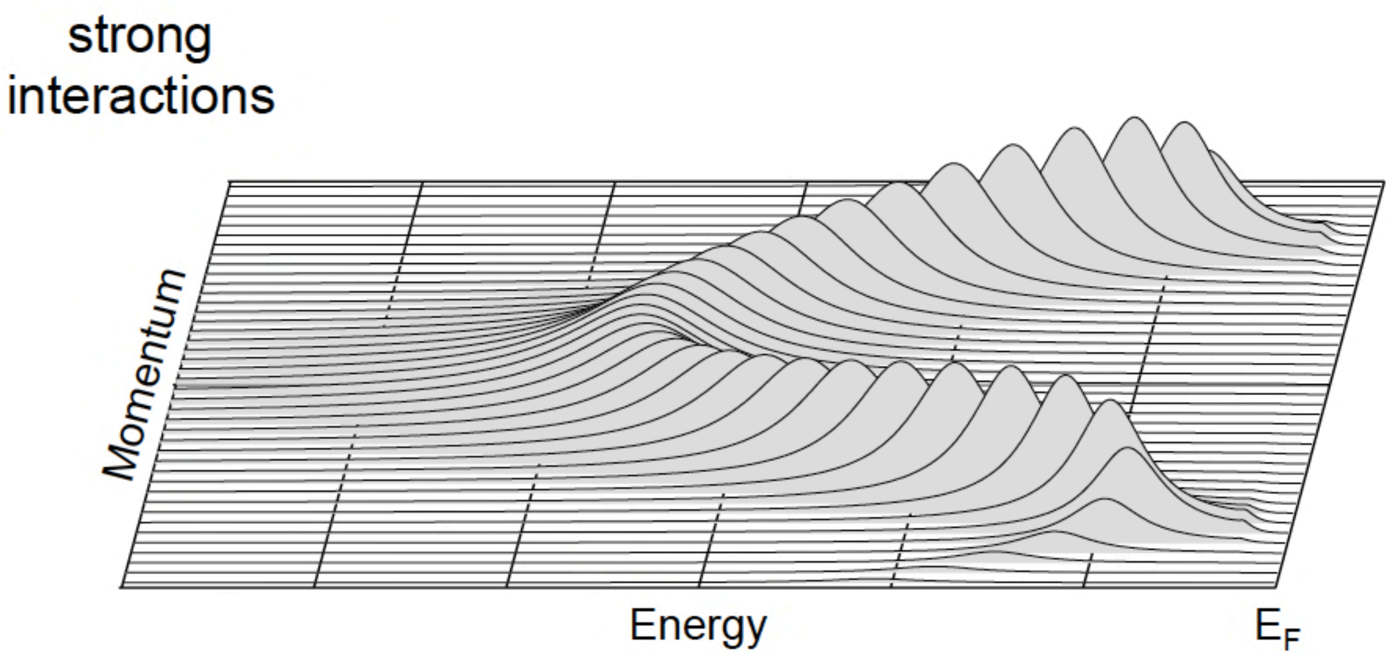
Lead actor: The quasi-electron



Strong correlations

Lead actor: The quasi-electron

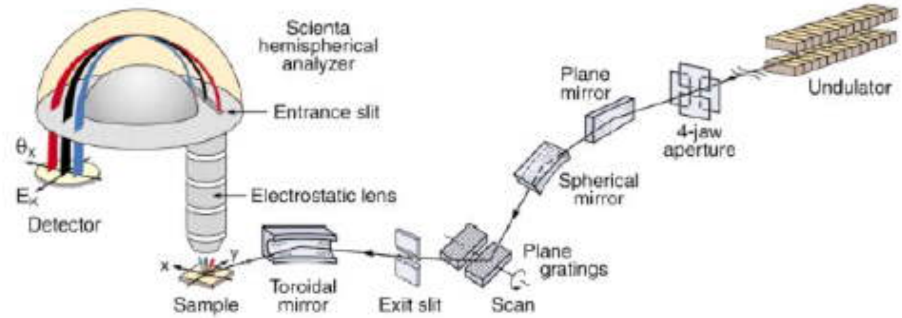




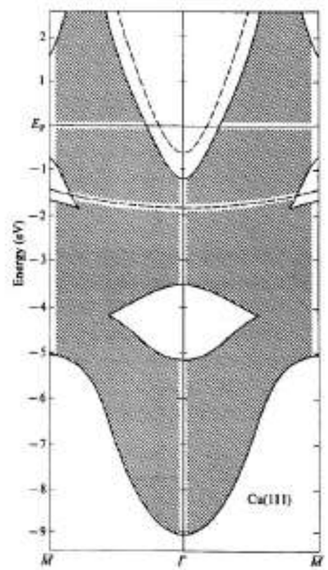
Nobel prize to X-ray spectroscopy work

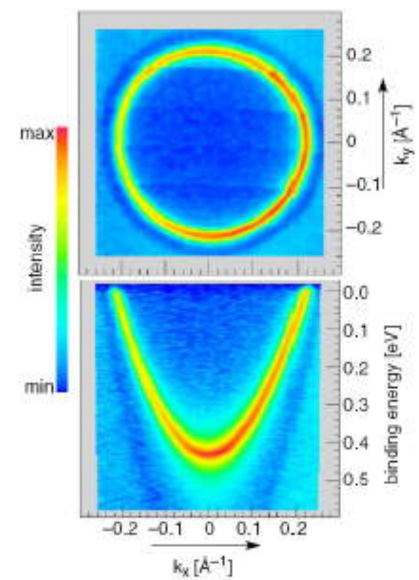
Angle-resolved photoemission - ARPES

K. Siegbahn
Nobelprize 1981



e.g. Cu-sp Shockley surface state





Taken from F. Reinert und S. Hüfner, New Journal of Physics 7, 97 (2005)

Non-correlated materials

solid state

↓

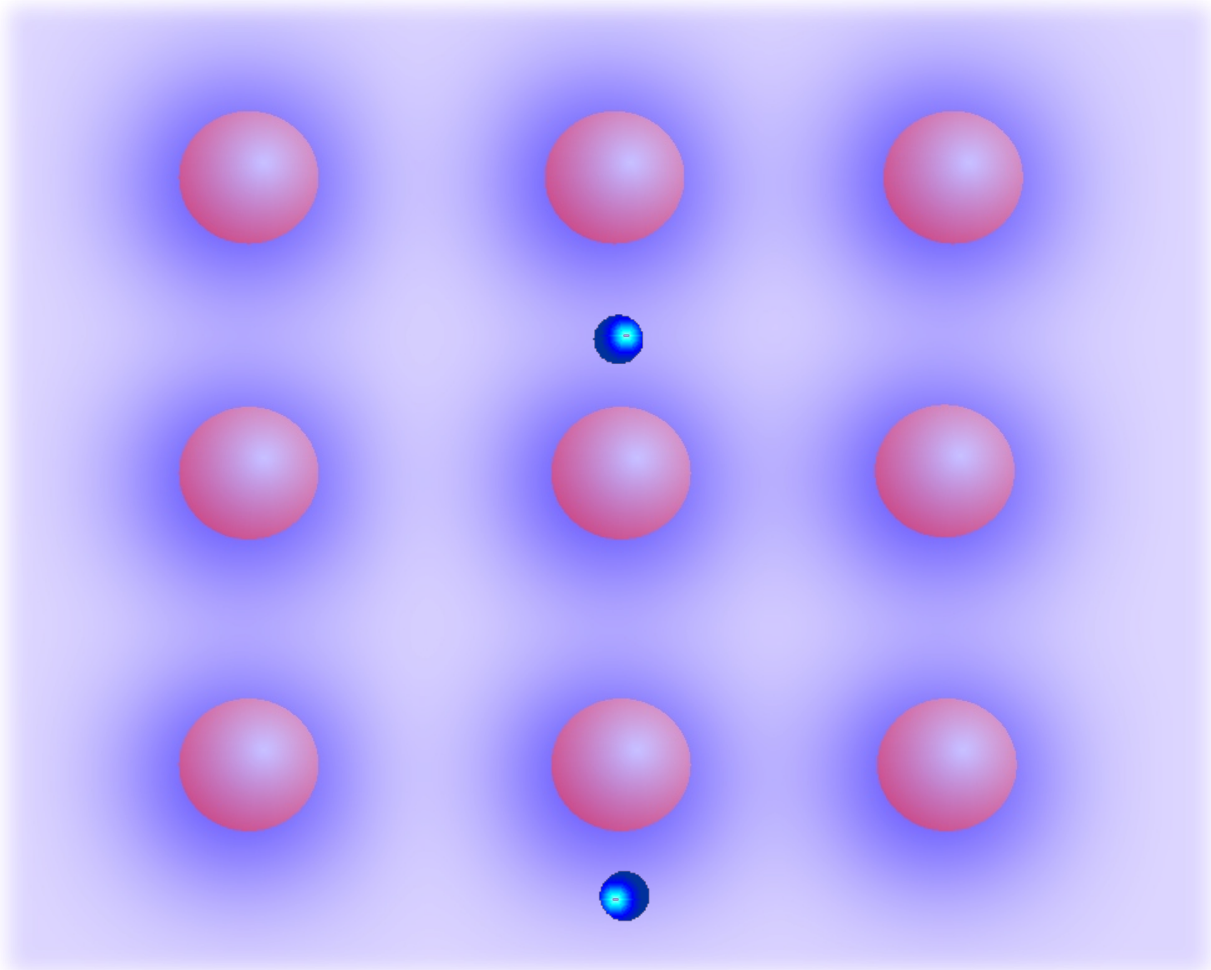
potential averaged over all electrons

↓

single-particle calculations, LDA approach

Problem: ↓

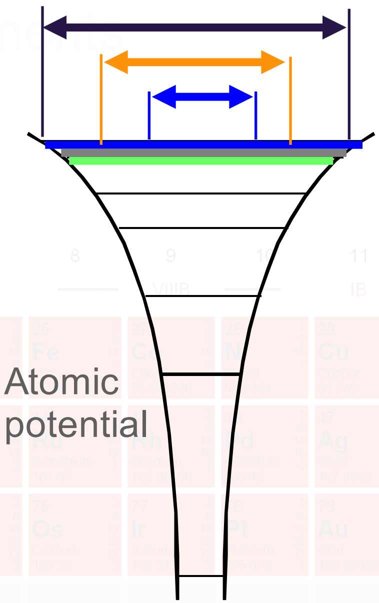
negligence of electron correlations

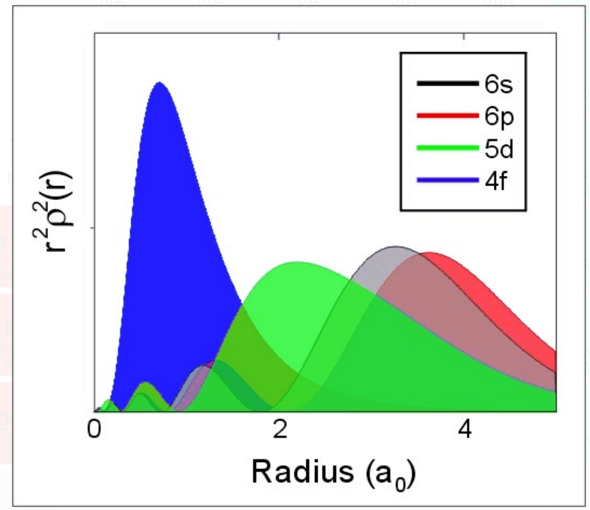


Correlated *f*-materials

Electron configuration:

[Xe] $4f^{n}\ 5d^{m}\ 6s^{2}$





Lanthanoide

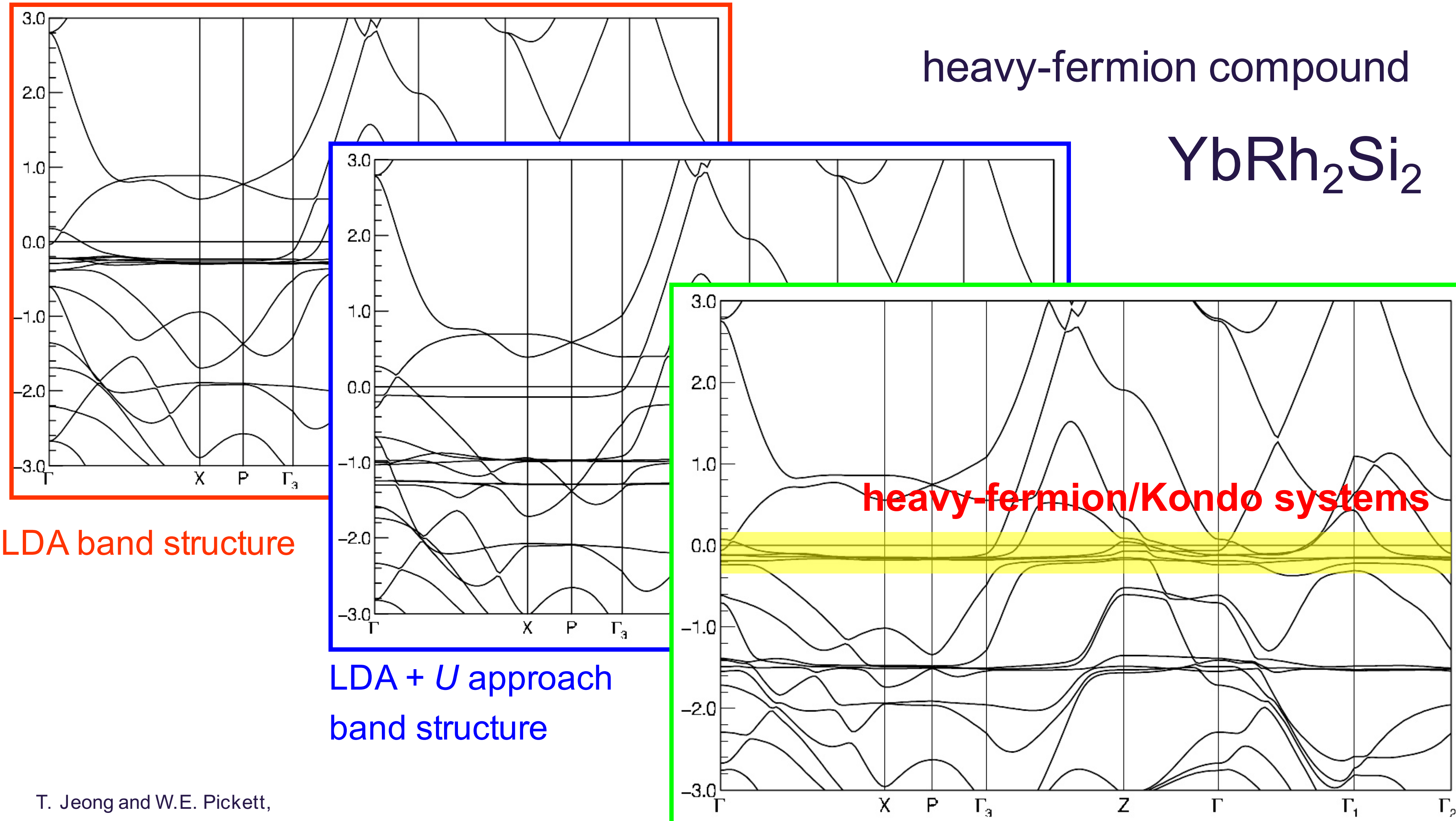
57	58	59	60	61	62	63	64	65	66	67	68	69	70	71
La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
Lanthanum	Cerium	Praseodymium	Neodymium	Promethium	Samarium	Europium	Gadolinium	Terbium	Dysprosium	Holmium	Erbium	Thulium	Ytterbium	Lutetium
138.9055	140.116	140.90765	144.24	(145)	150.36	151.964	157.25	158.92534	162.50	164.93032	167.26	168.93421	173.04	174.967

f - electrons

Correlated *f*-materials

heavy-fermion compound

$YbRh_2Si_2$



LDA band structure

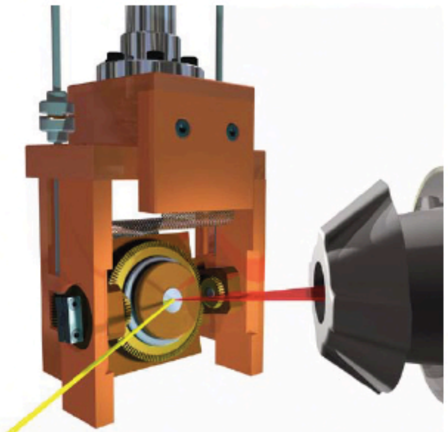
LDA + *U* approach band structure

fully relativistic band structure

T. Jeong and W.E. Pickett, J. Phys.: Condens. Matter **18** (2006) 6289

ARPES, basics (Kai Rossnagel)

Angle-Resolved PhotoElectron Spectroscopy



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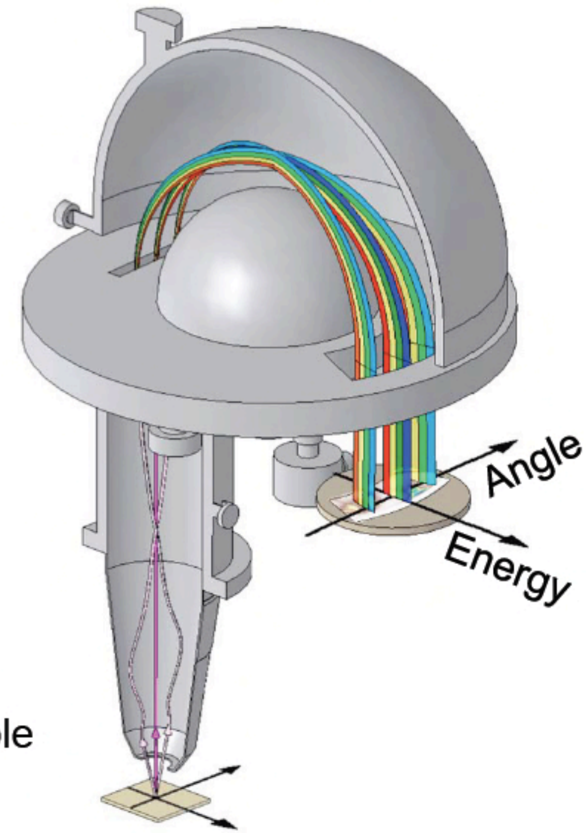

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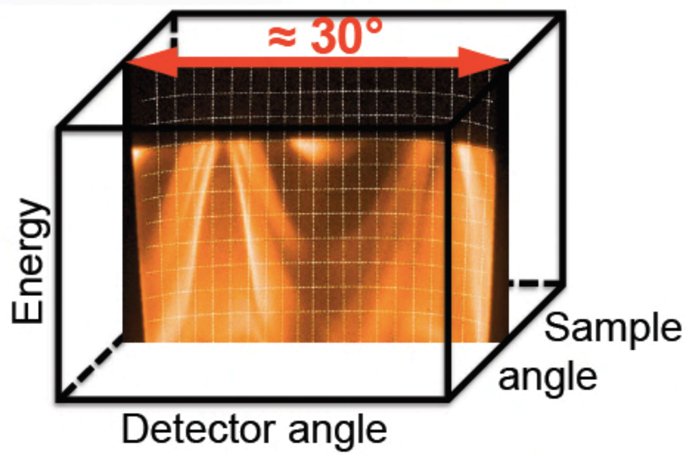
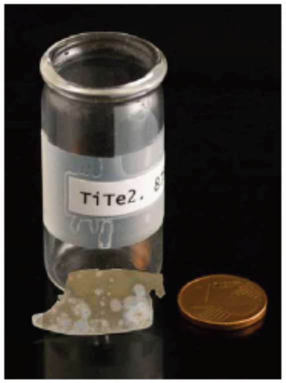
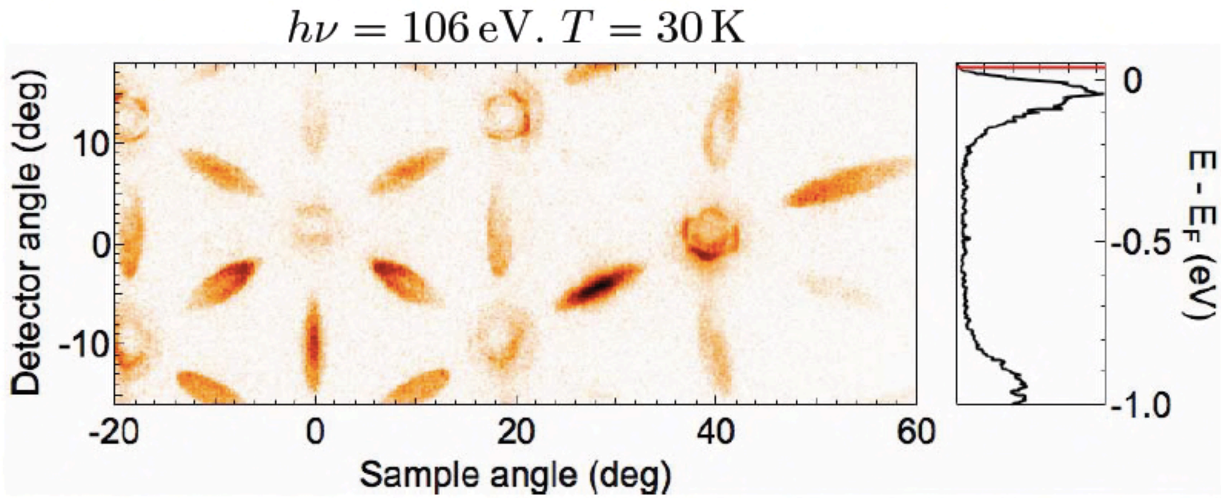
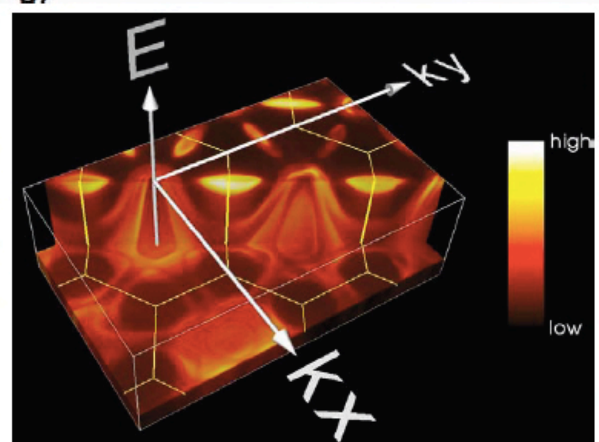


Band mapping (seeing is believing)





K. Rossnagel, et al. Uni Kiel

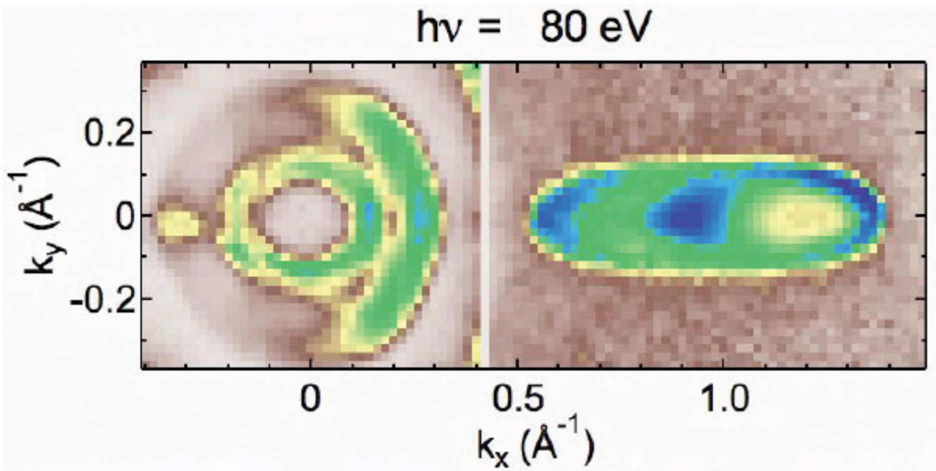


$$E - E_F = E_{\mathrm{kin}} + W - h\nu$$

$$\begin{pmatrix} k_{\parallel x} \\ k_{\parallel y} \end{pmatrix} = \sqrt{\frac{2m}{\hbar^2} E_{\mathrm{kin}}} \begin{pmatrix} \sin\Theta_{\mathrm{D}} \\ \cos\Theta_{\mathrm{D}} \sin\Phi_{\mathrm{S}} \end{pmatrix}$$

Beamline 7, ALS, Berkeley

Fermi surface tomography

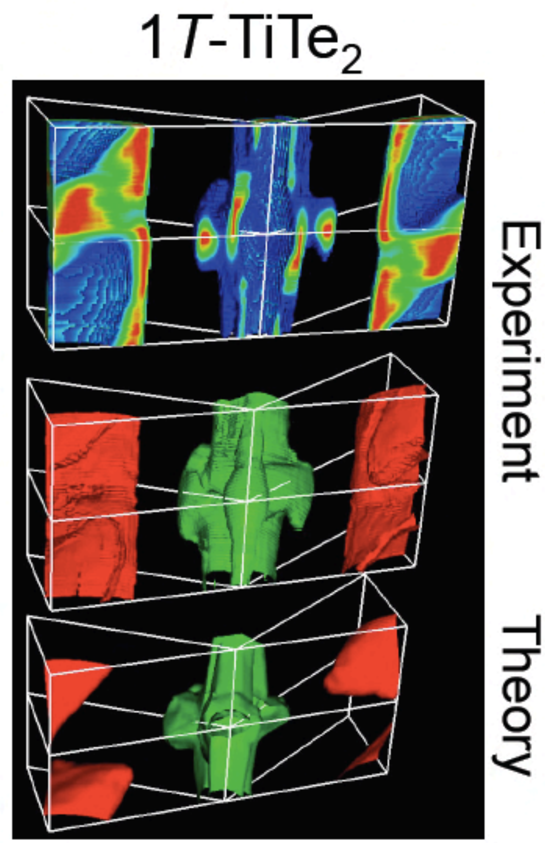


K. Rossnagel, et al. Uni Kiel

$$\mathbf{k}_{\parallel}^2 + k_{\perp}^2 = \frac{2m}{\hbar^2}(E_{\mathrm{kin},F} + V_0)$$

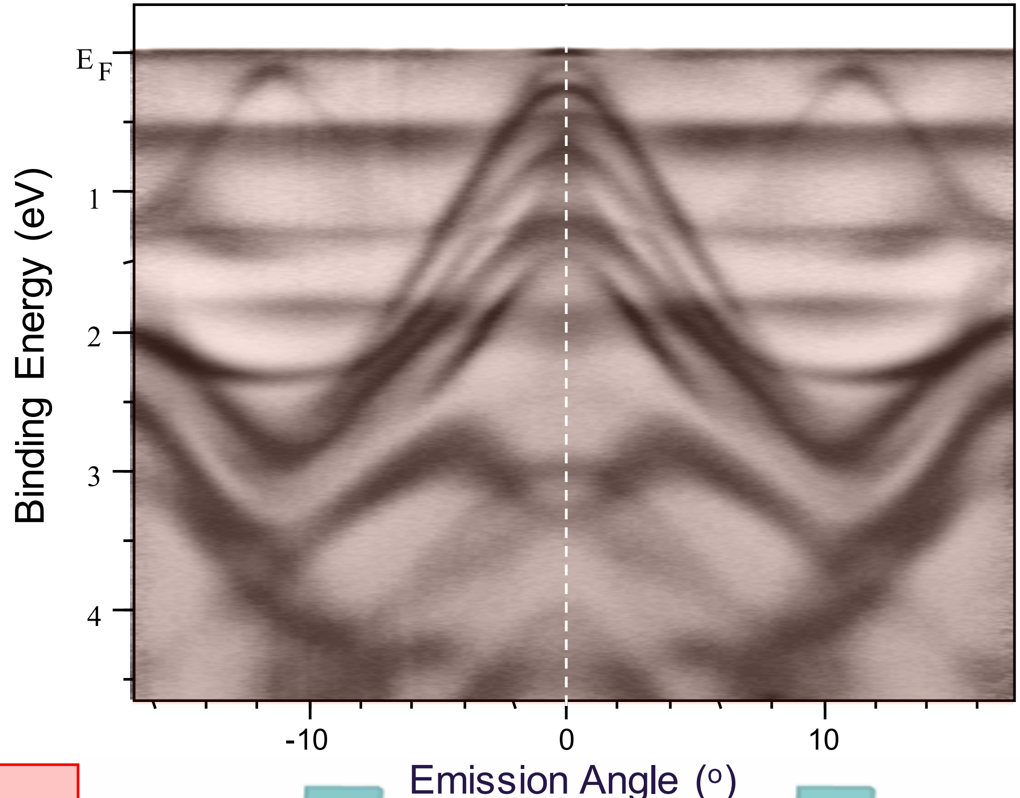
$$\mathbf{k}_{\parallel} = \sqrt{\frac{2m}{\hbar^2}E_{\mathrm{kin},F}}\begin{pmatrix}\sin\Theta_\mathrm{D} \\ \cos\Theta_\mathrm{D}\sin\Phi_\mathrm{S}\end{pmatrix}$$

$$E_{\mathrm{kin},F} = h\nu - W$$

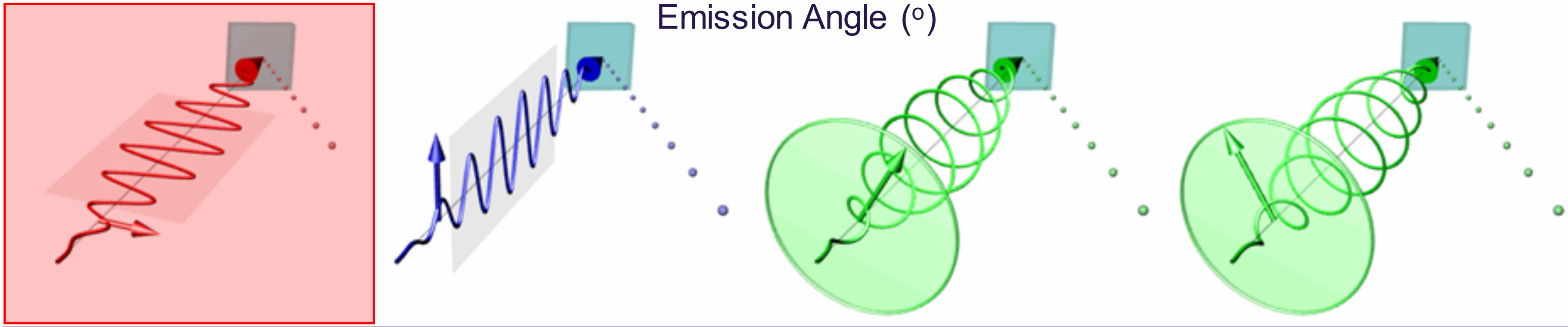


Beamline 7, ALS, Berkeley

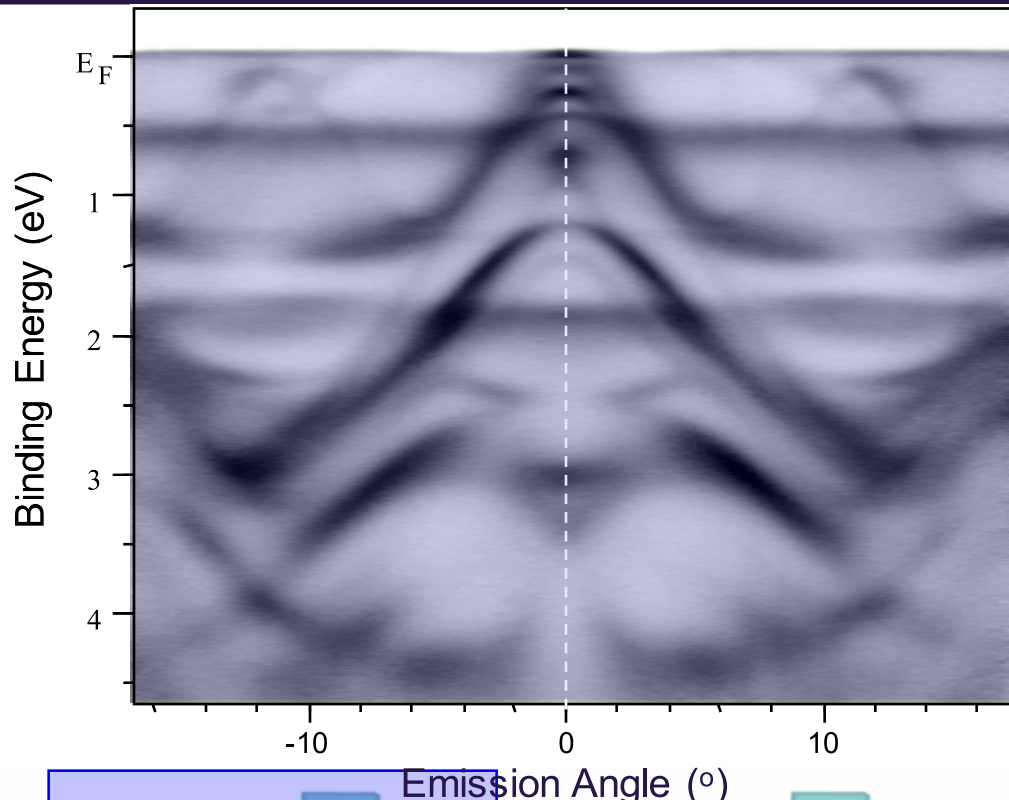
Light-polarized ARPES on heavy-fermion $YbRh_2Si_2$ (S. Molodtsov, et al.)



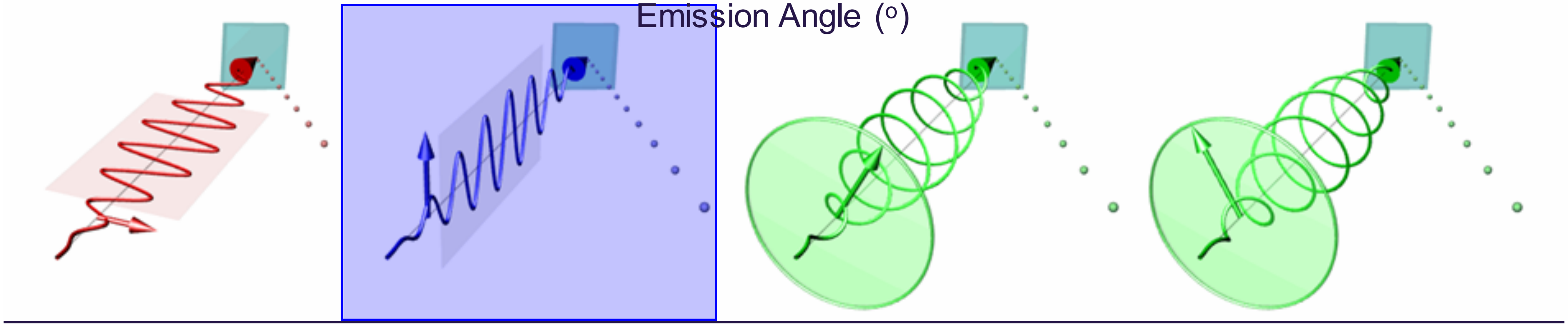
$YbIr_2Si_2$



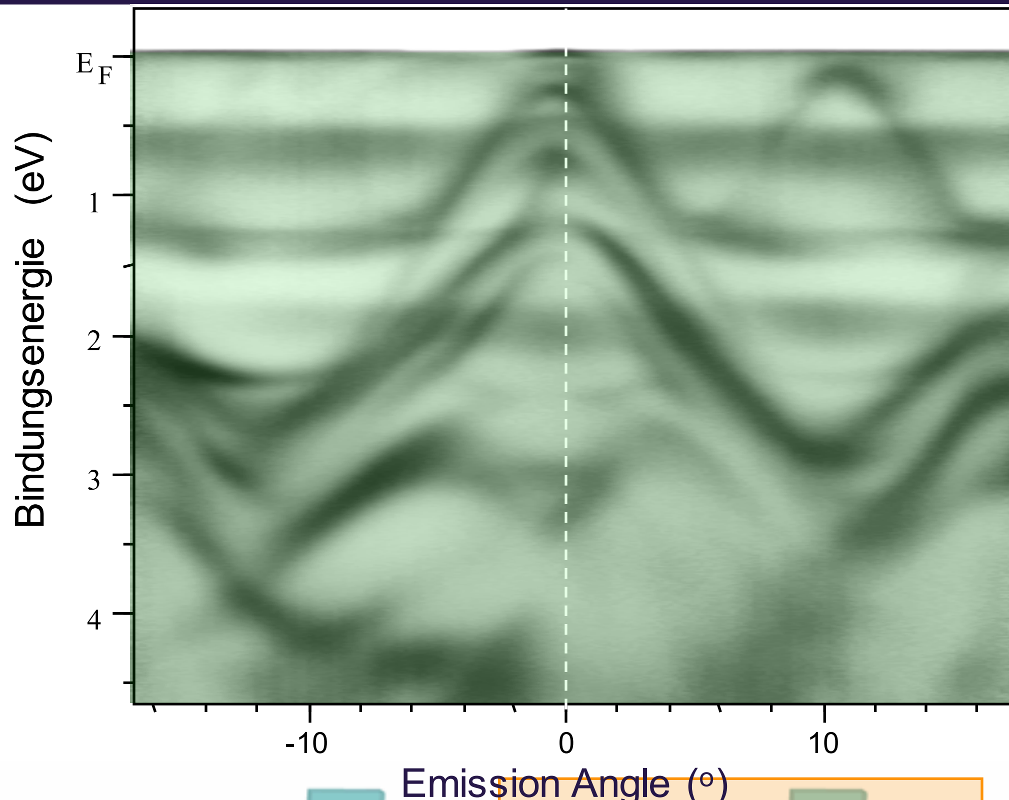
Light-polarized ARPES ($YbRh_2Si_2$)



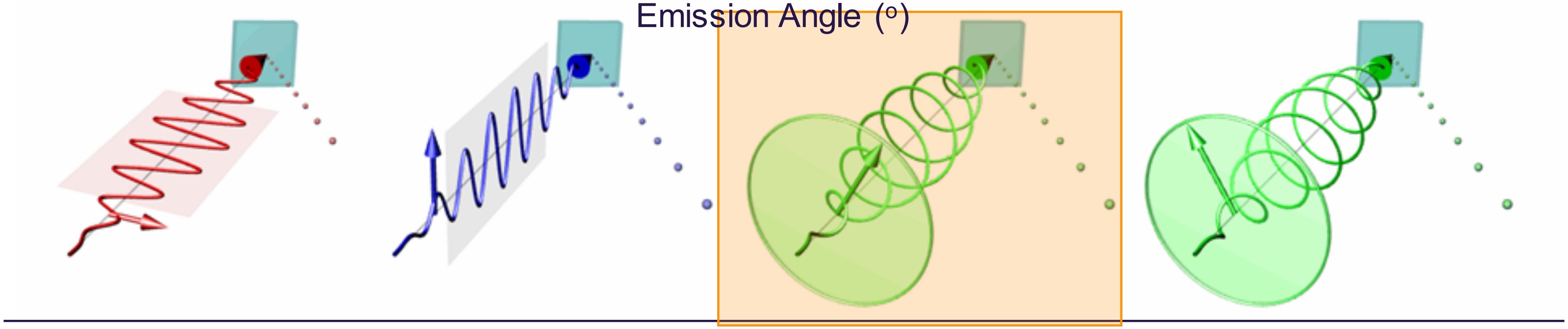
$YbIr_2Si_2$



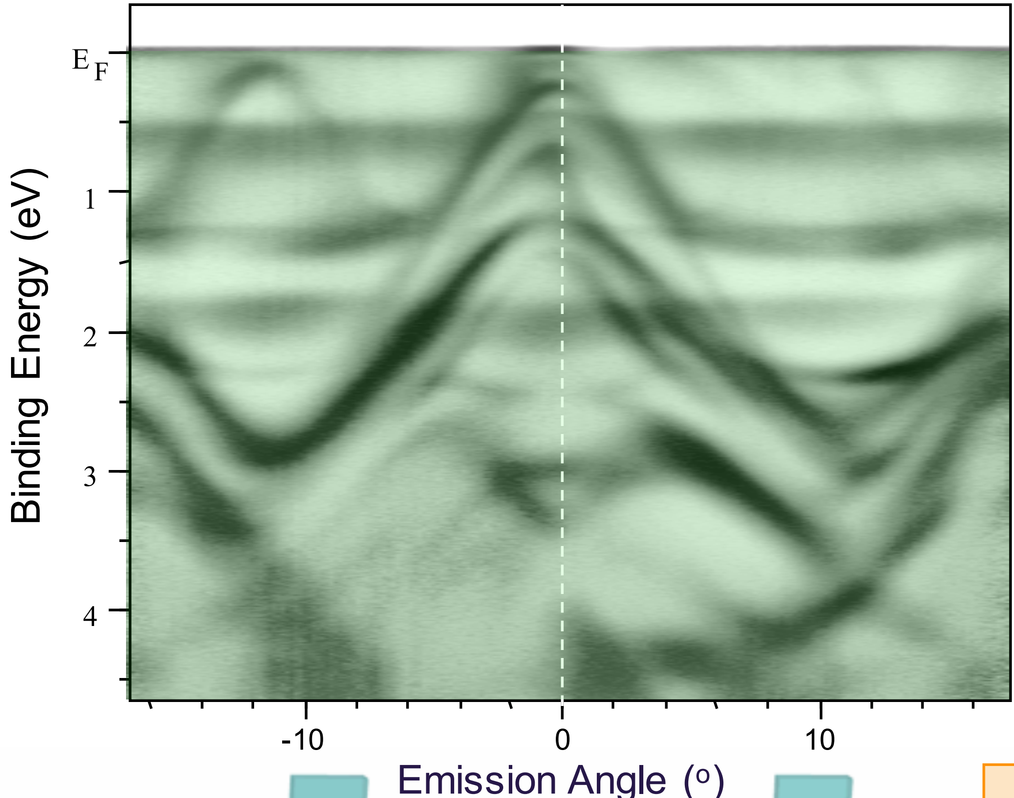
Light-polarized ARPES ($YbRh_2Si_2$)



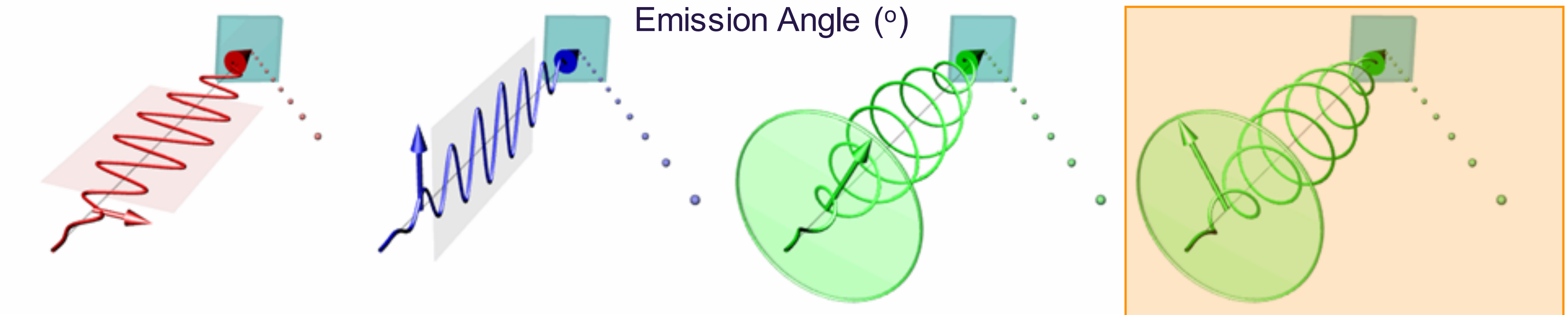
$YbIr_2Si_2$



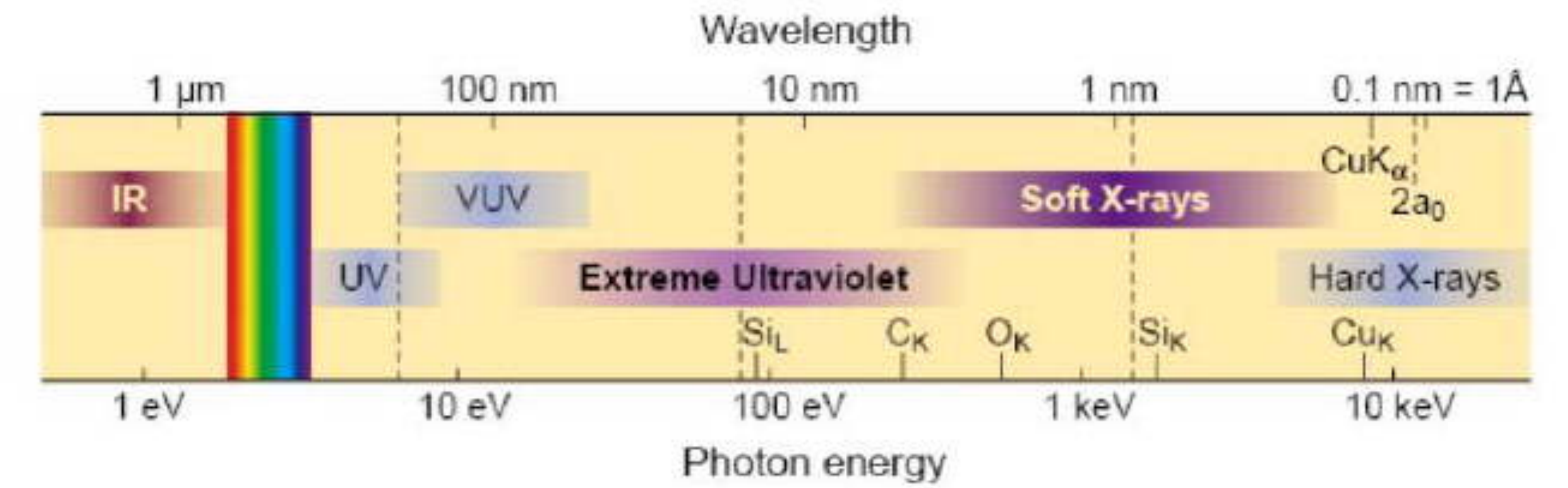
Light-polarized ARPES ($YbRh_2Si_2$)



$YbIr_2Si_2$



Spectral range and radiation sources



Excharge (He) lamps – VUV/Extreme Ultraviolet
Röntgen (Cu) tubes – Soft/Hard X-rays

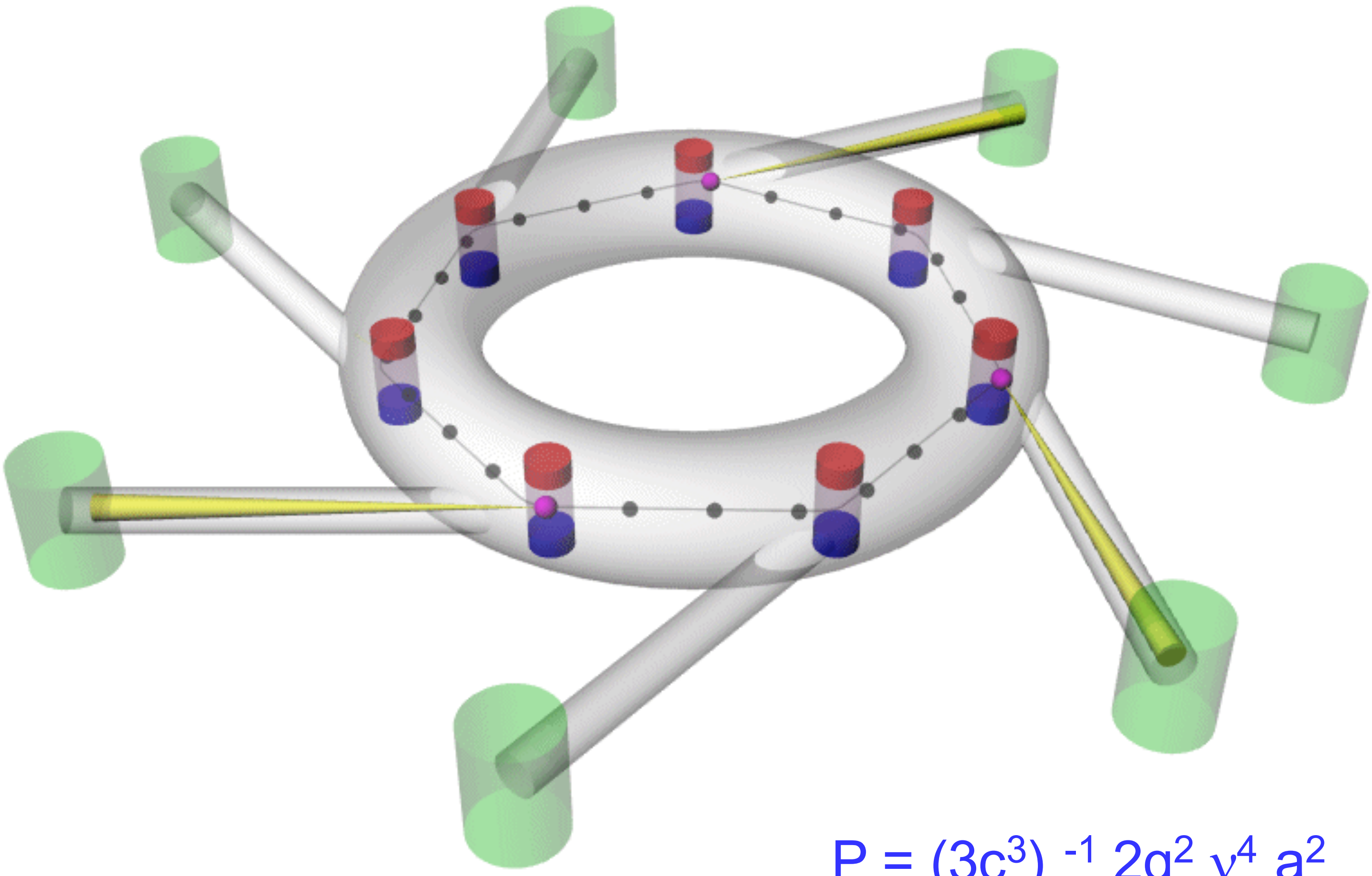
Disadvantages: (i) low intensity; (ii) discrete spectrum; (iii) no time structure

Revolution with synchrotron radiation !!!

Synchrotron Radiation

Synchrotrons/Storage Rings

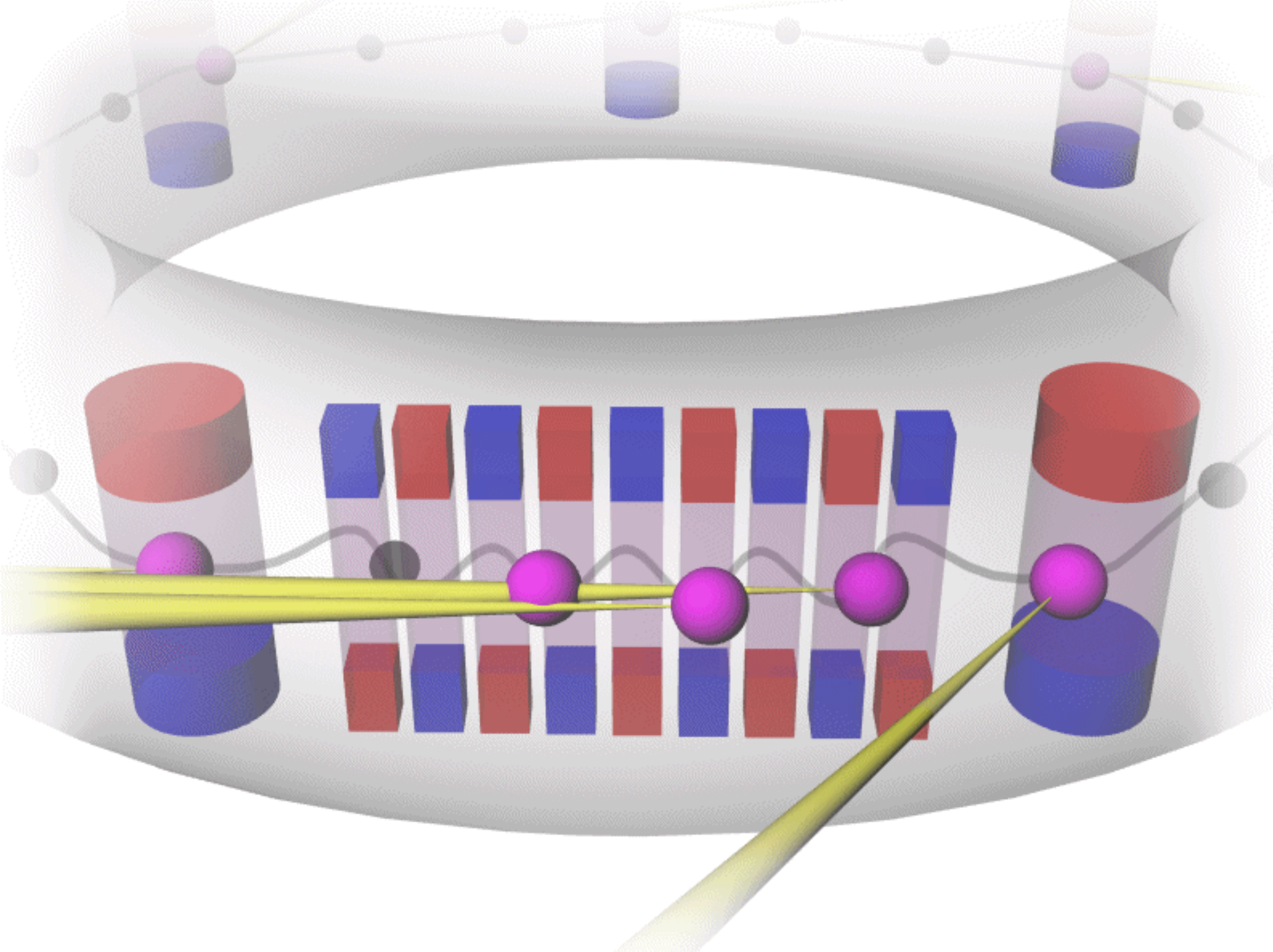
Synchrotron Radiation (dipoles)



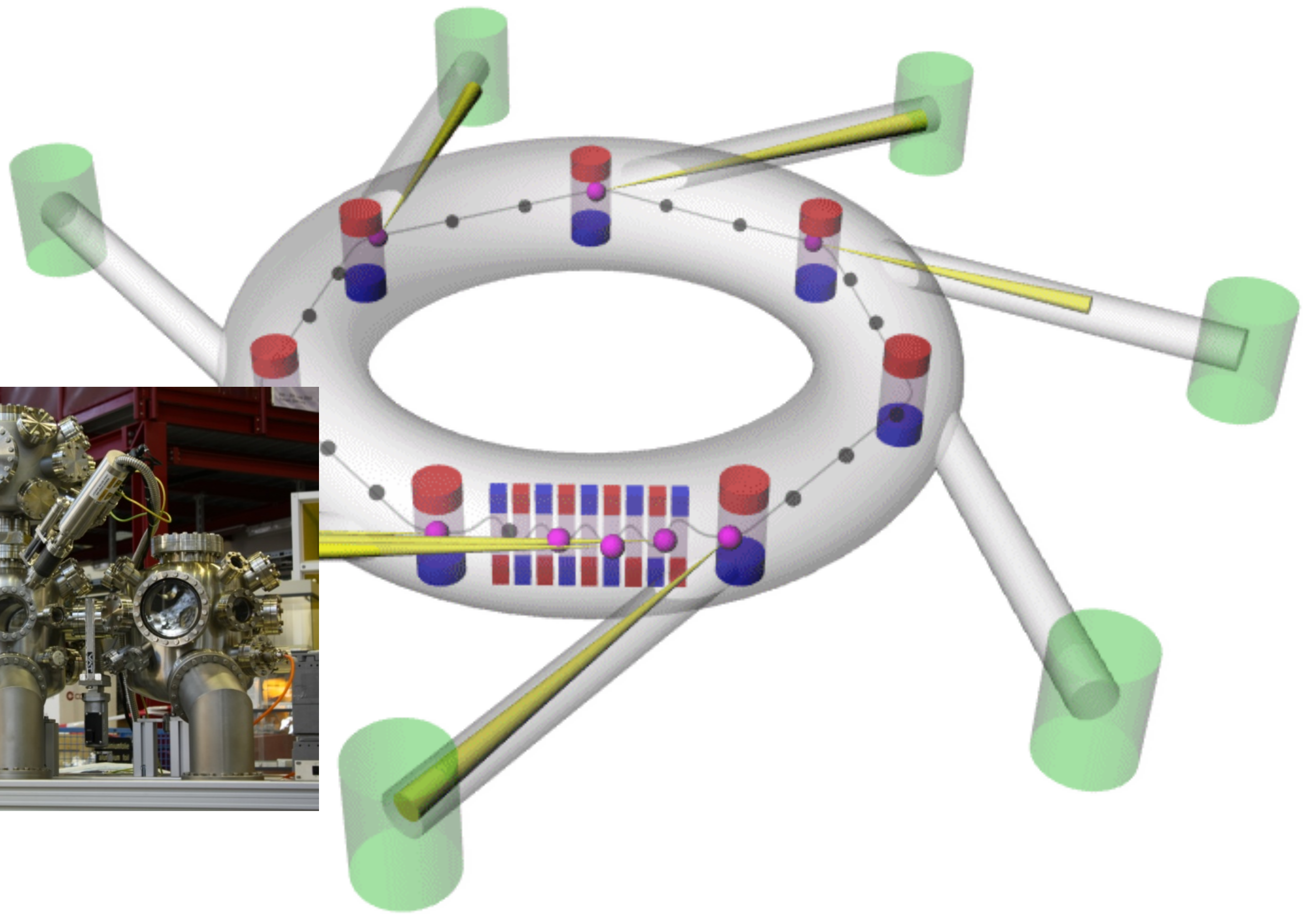
$$P = (3c^3)^{-1}\, 2q^2\, \nu^4\, a^2$$

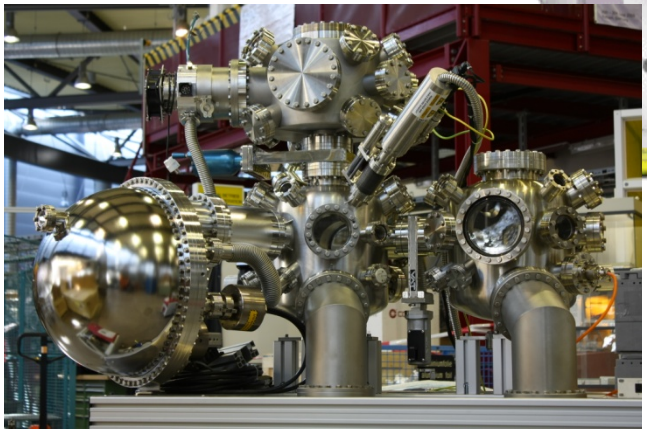
P – radiated power; c – light velocity; q – particle charge; a – acceleration; ν - normalized energy

Synchrotron radiation (undulators)

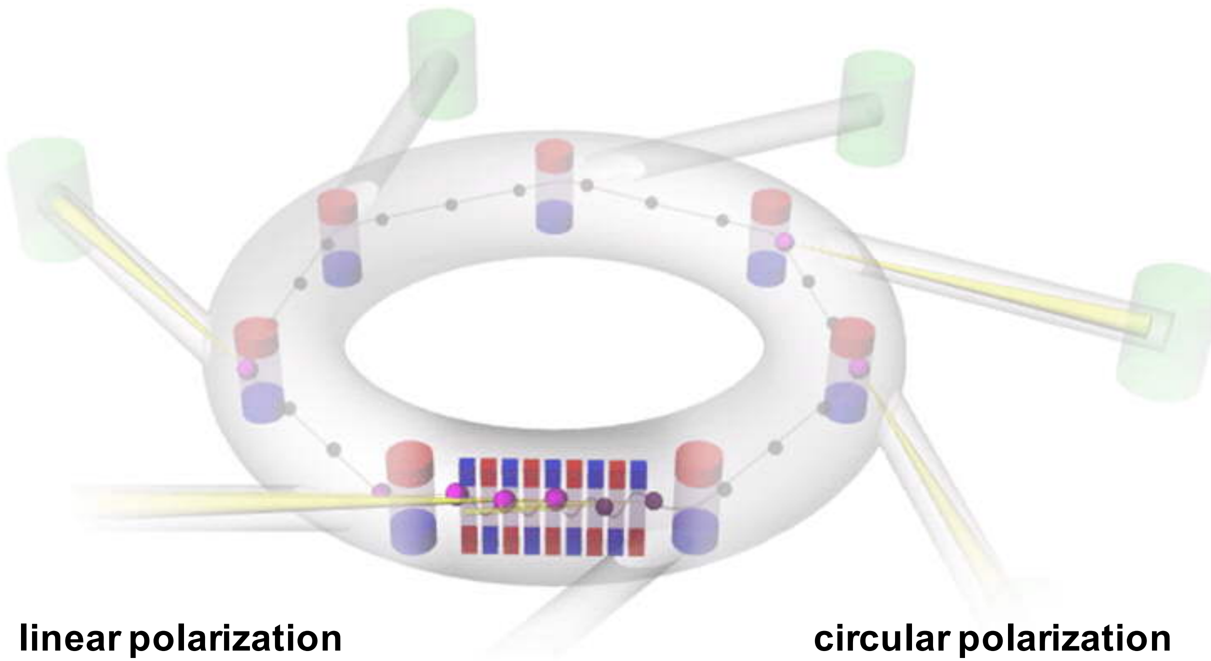


Synchrotron radiation (sources + exp. stations)

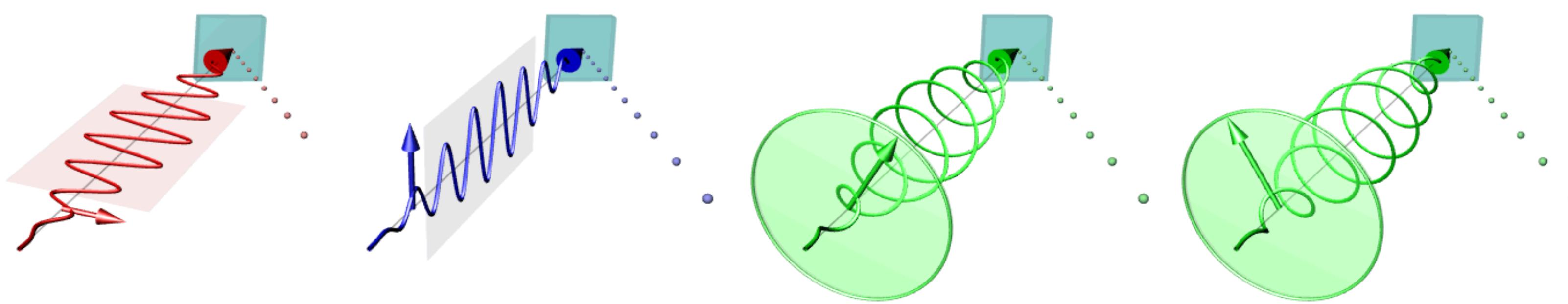




Synchrotron Radiation (light polarization)

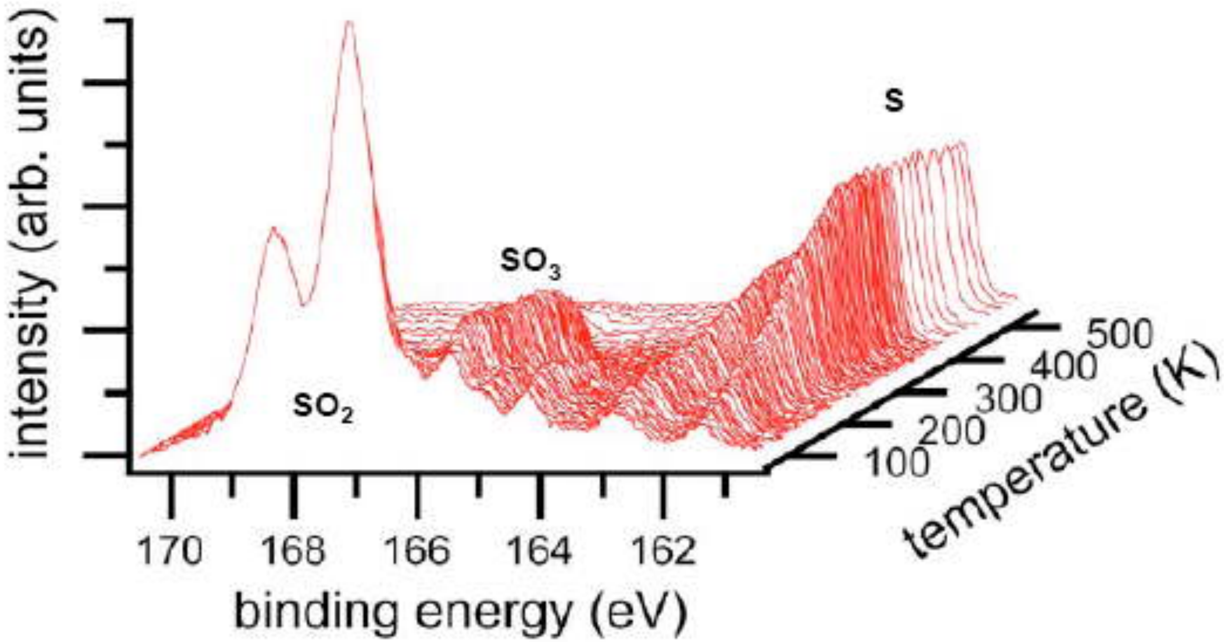


linear polarization **circular polarization**



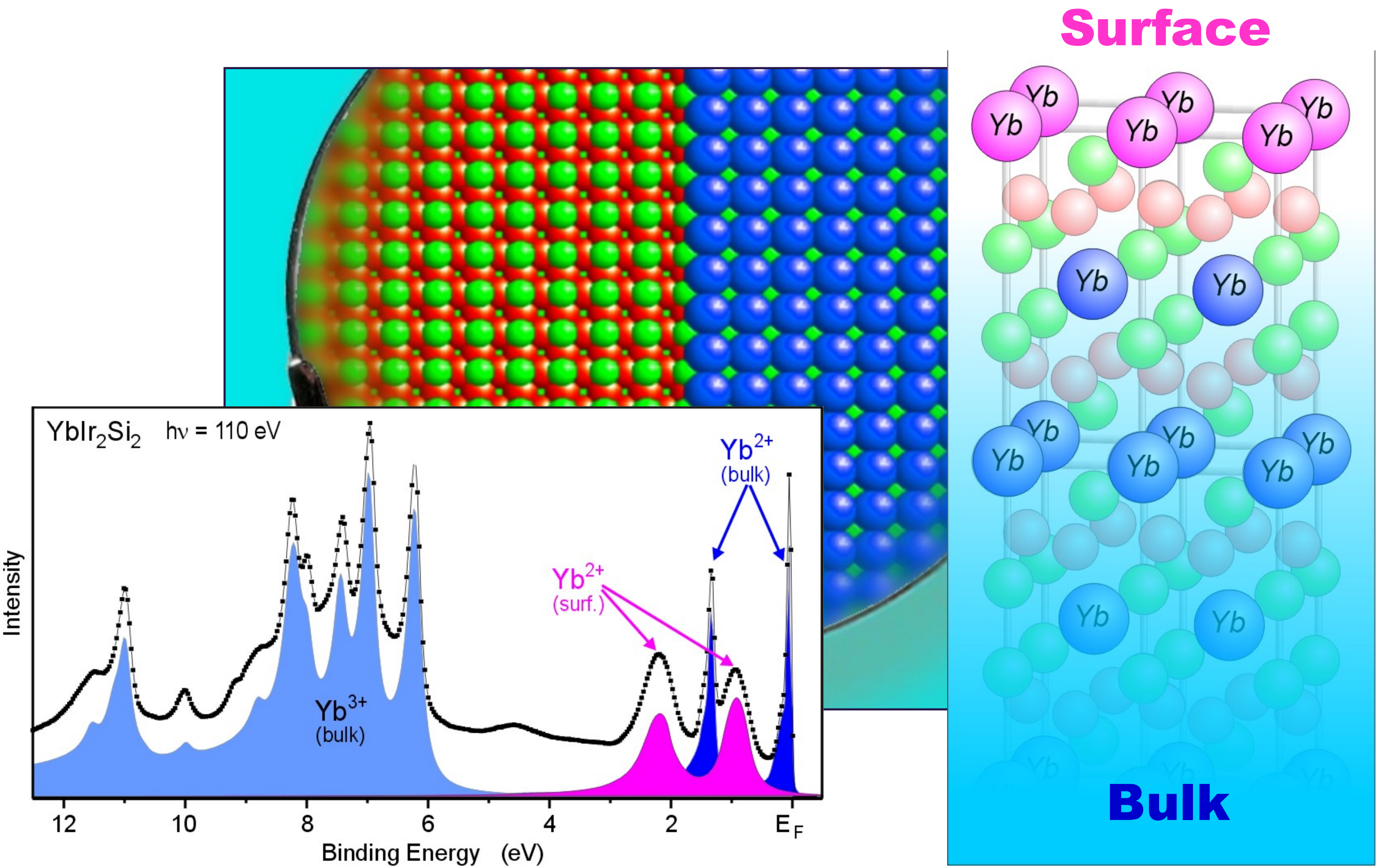
Spectroscopic toolbox: X-ray photoelectron spectroscopy, ESCA

SO_2 dissociation on Ru(001)

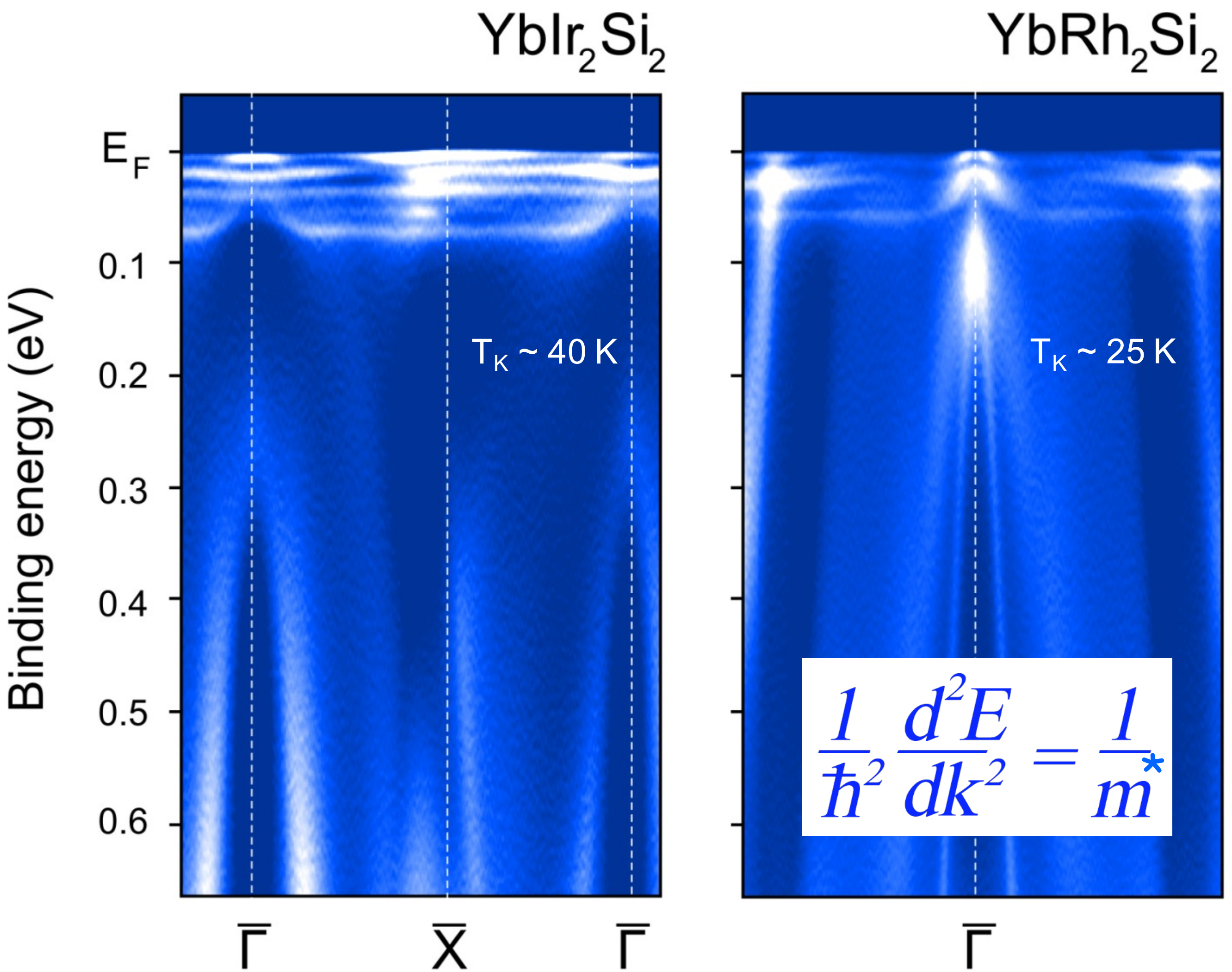


F. Hennies et al., J.Chem.Phys. 127, 154709 (2007)

Characterization of cleaved samples: $YbRh_2Si_2$ (S. Molodtsov, et al.)



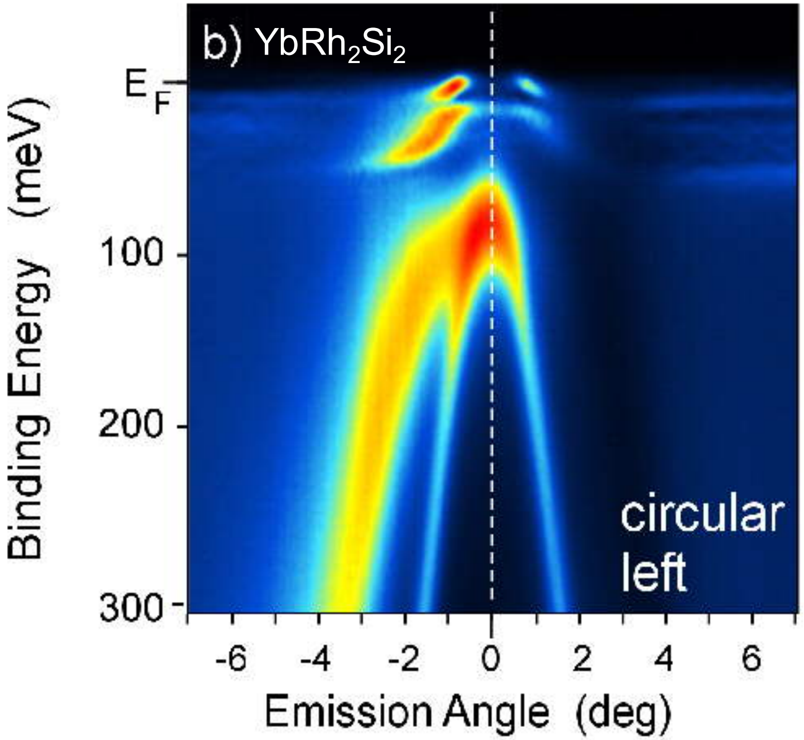
Crystal-field split f-states in Kondo systems

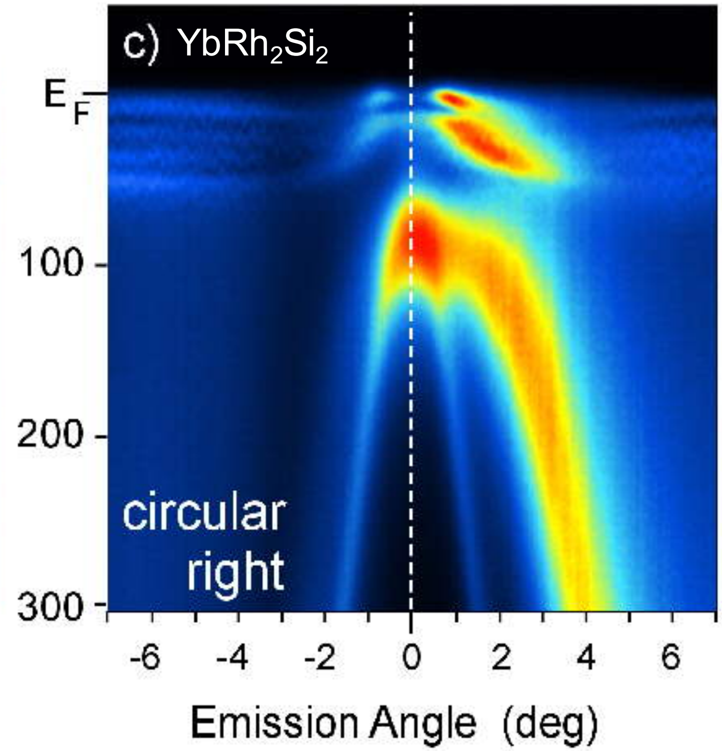


$$\frac{1}{\hbar^2}\frac{d^2E}{dk^2} = \frac{1}{m^*}$$

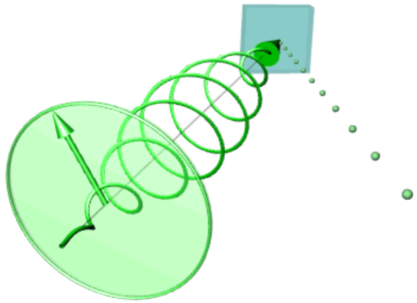
- effective mass mapping (transport phenomena)
- crystal field-split *4f* states probing (magnetic properties)
- strength of electron states correlation (Kondo behavior)

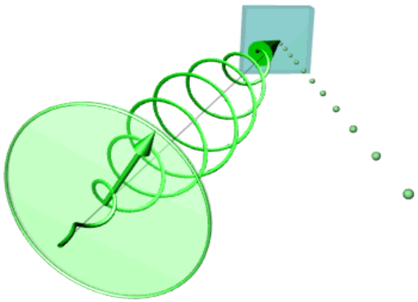
Symmetry of f-states in $YbRh_2Si_2$





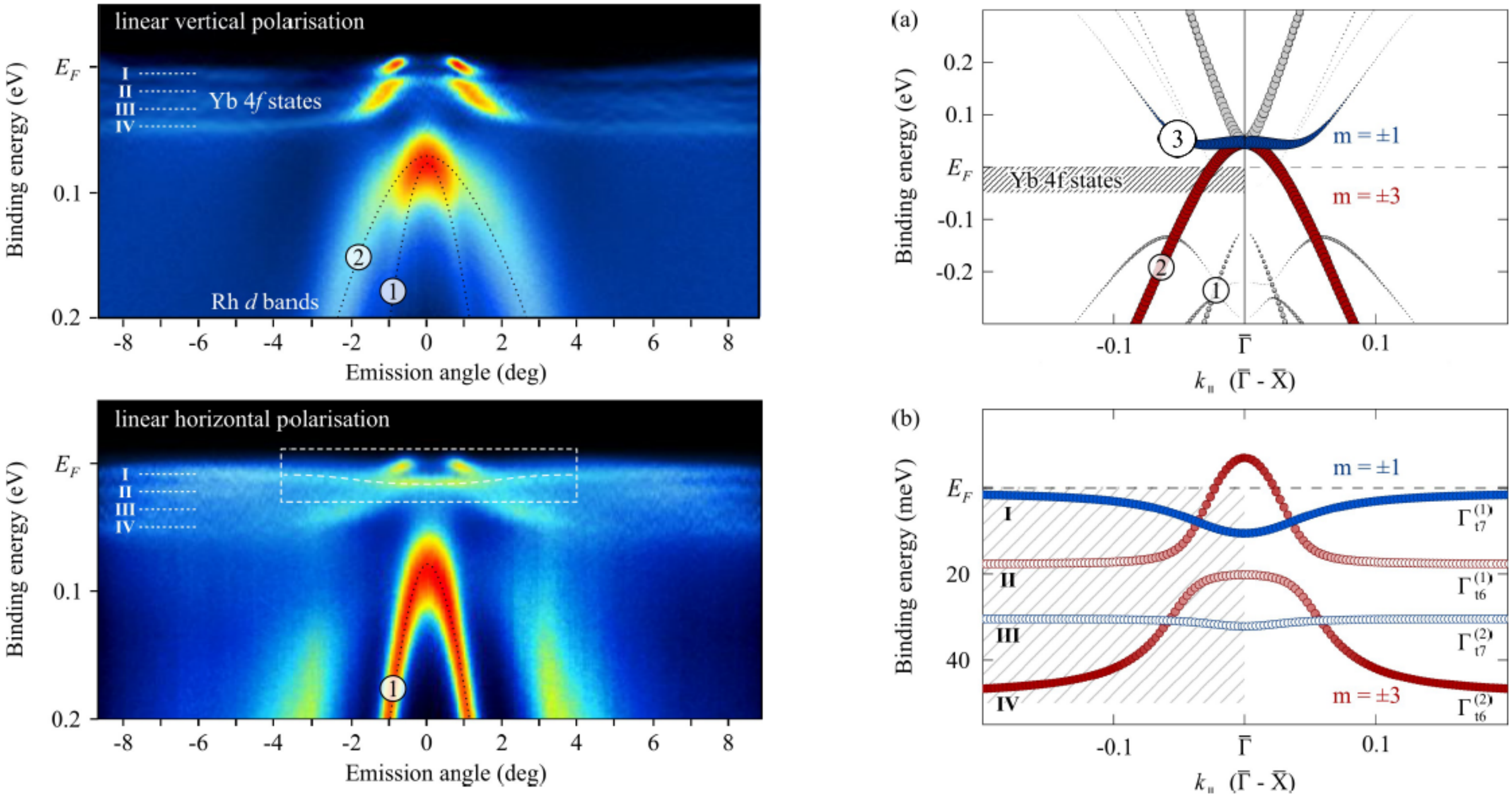
circular left





circular right

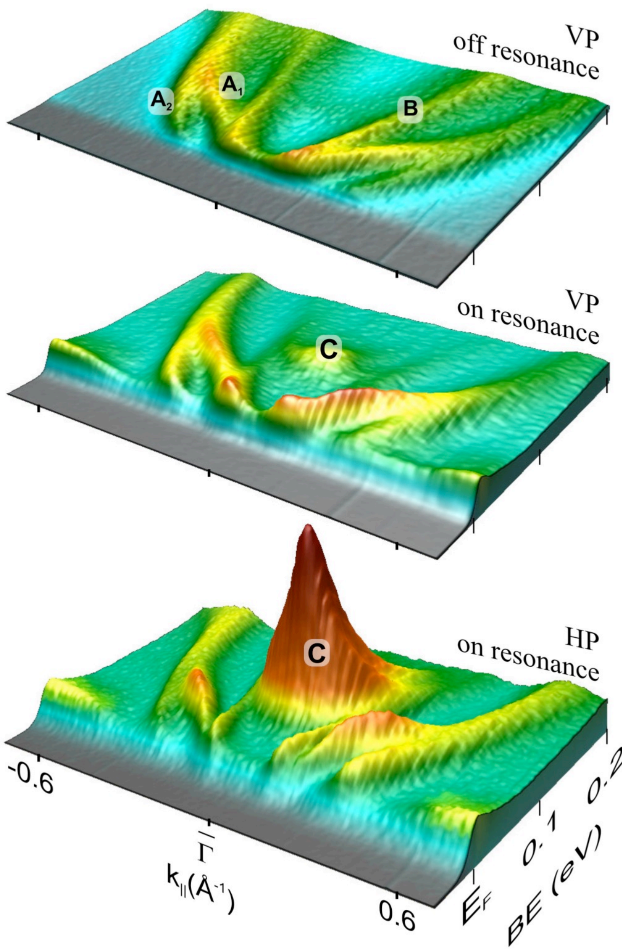
Heavy-fermion behavior in $YbRh_2Si_2$



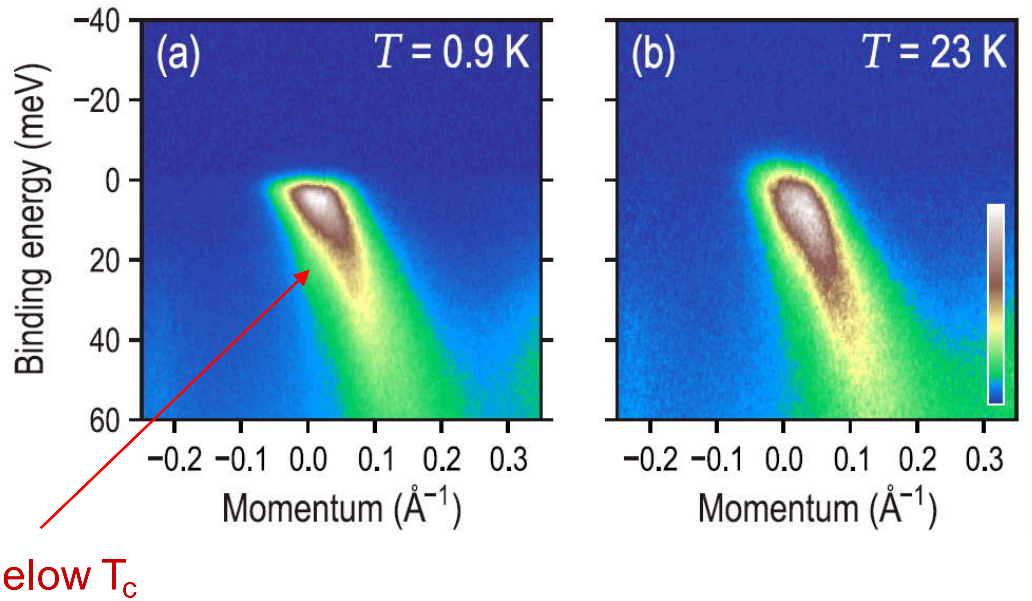
Dispersion of the 4f states around Γ where they hybridize to Rh d bands: Experiment & theory

High-temperature superconductors (S. Borisenko, et al.)

CeFePO



LiFeAs



- which band is responsible for superconductivity?
- how large is superconducting gap?
- how strong is electron pairing (kink energy)?

What is missing?

Electron system dynamics that is of the time scale order < 0.1 ps

Probing dynamics one can decide, e.g. in favor of spin or phonon mediated mechanism of electron pairing both in superconducting and Kondo systems

Time scales for dynamics

Molecules

Phase relaxation
electronic transitions
vibrations

Intramolecular vibrational redistribution

Intermolecular energy transport

Electronic radiation lifetimes

10^{-9} s

10 fs — 100 fs — 1 ps — 10 ps — 100 ps — 1 ns — Time

Quantum kinetics (carriers & phonons)

Energy transfer to lattice
Hot phonon effects

Phase relaxation interband continuum/excitons

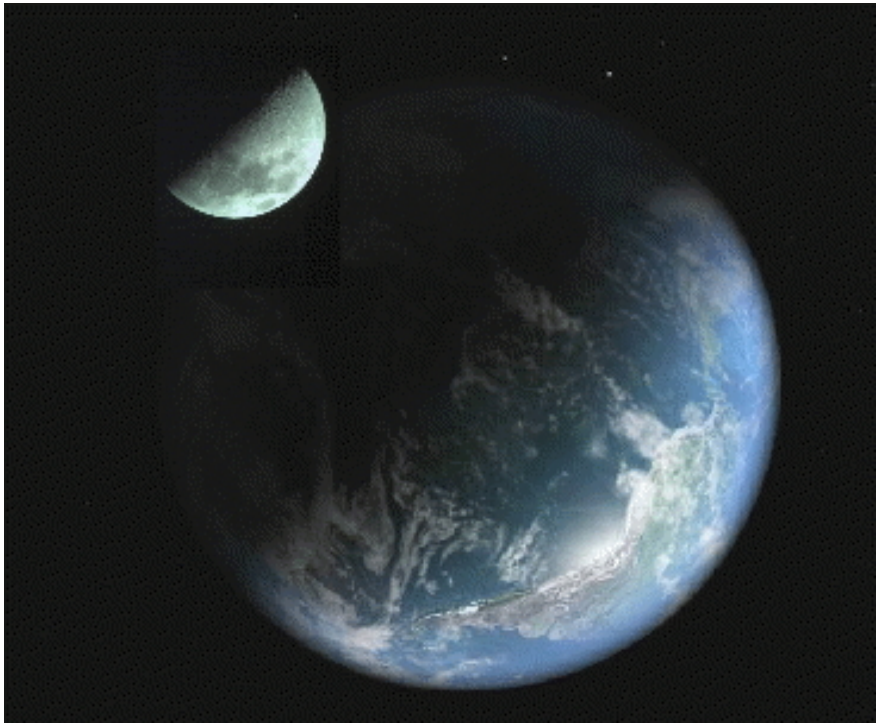
Phonon propagation

Intraband and intersubband redistribution of carriers

Solids

What is a picosecond?

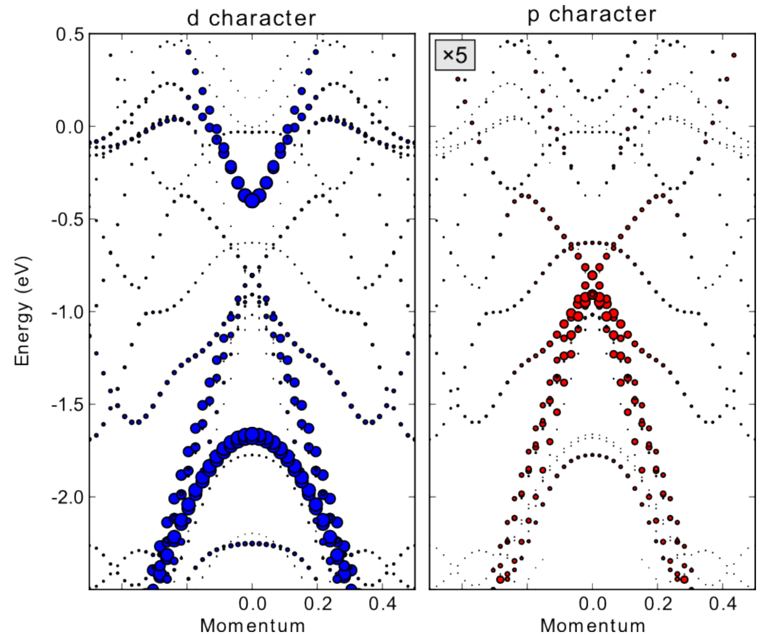
In 1 s light travels 300 000 km

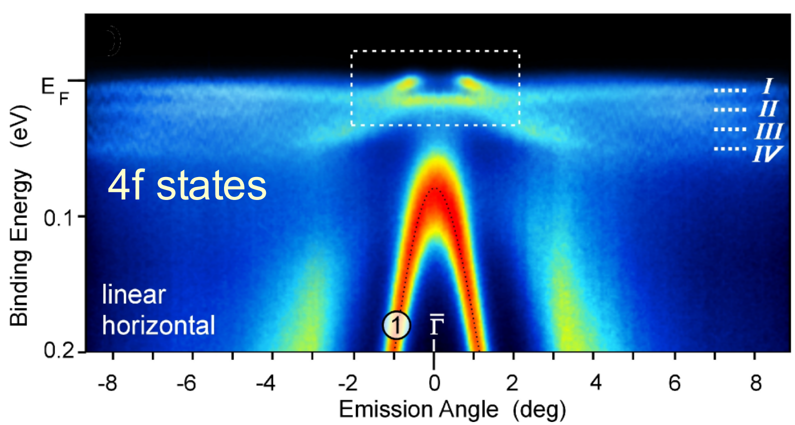


Distance between earth and moon is 384 000 km

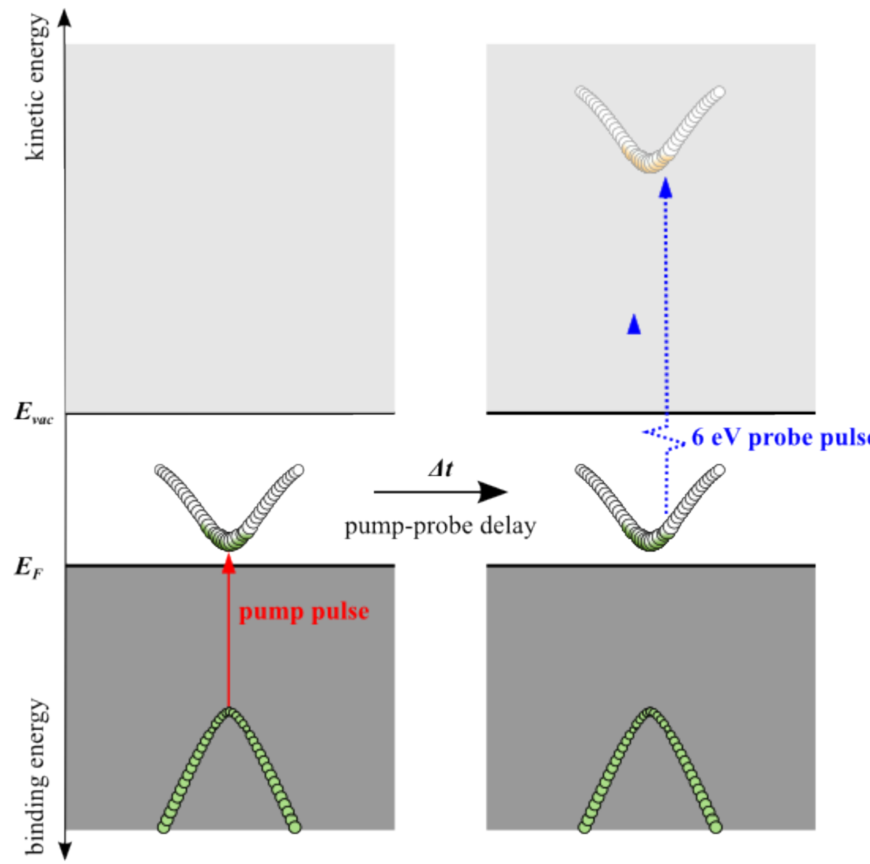
In 1 ps light travels 0,3 mm

Pump-probe experiment (K. Kummer, et al.)





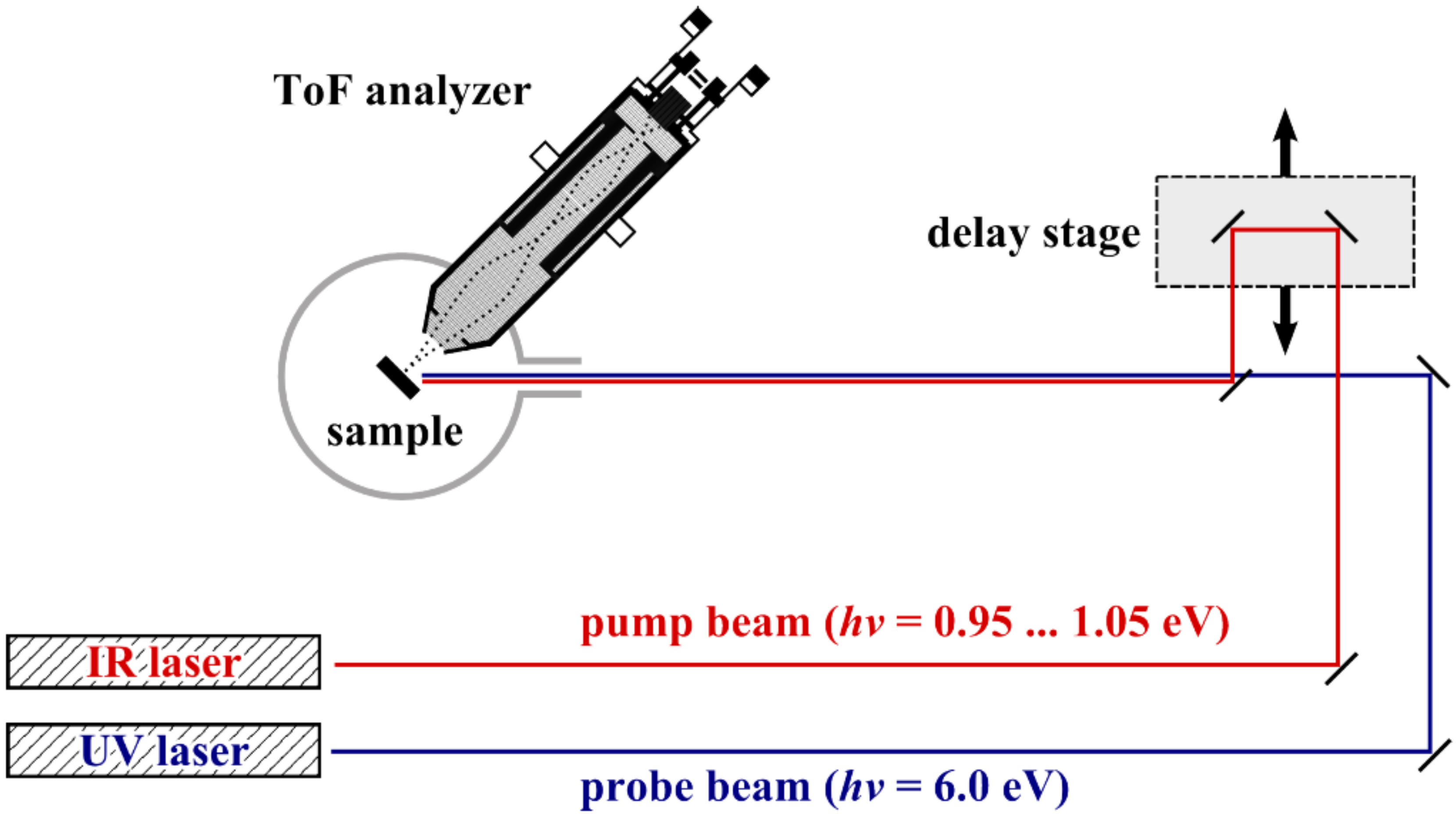
■ $YbRh_2Si_2$



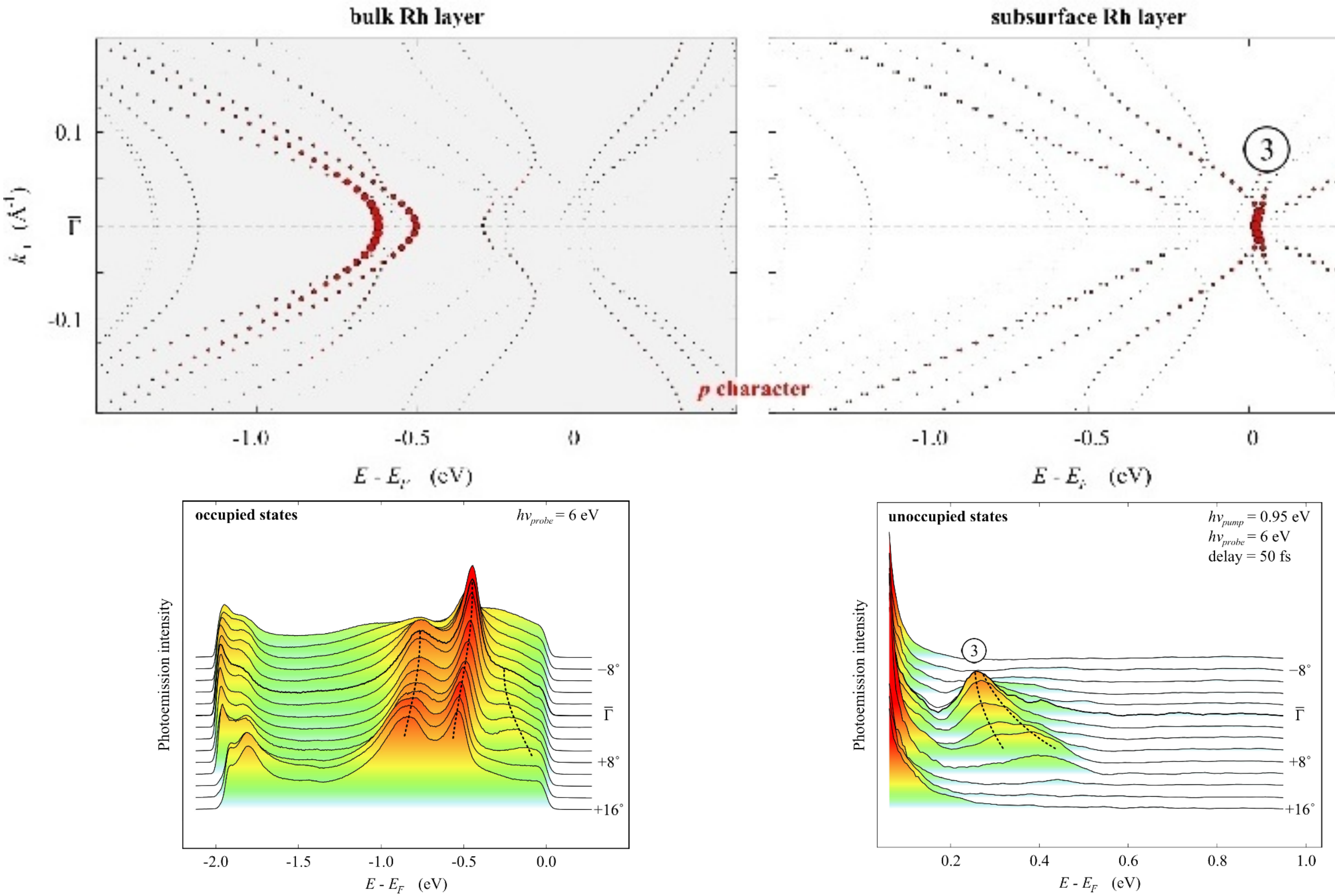
ARPES with MHz optical lasers

European XFEL

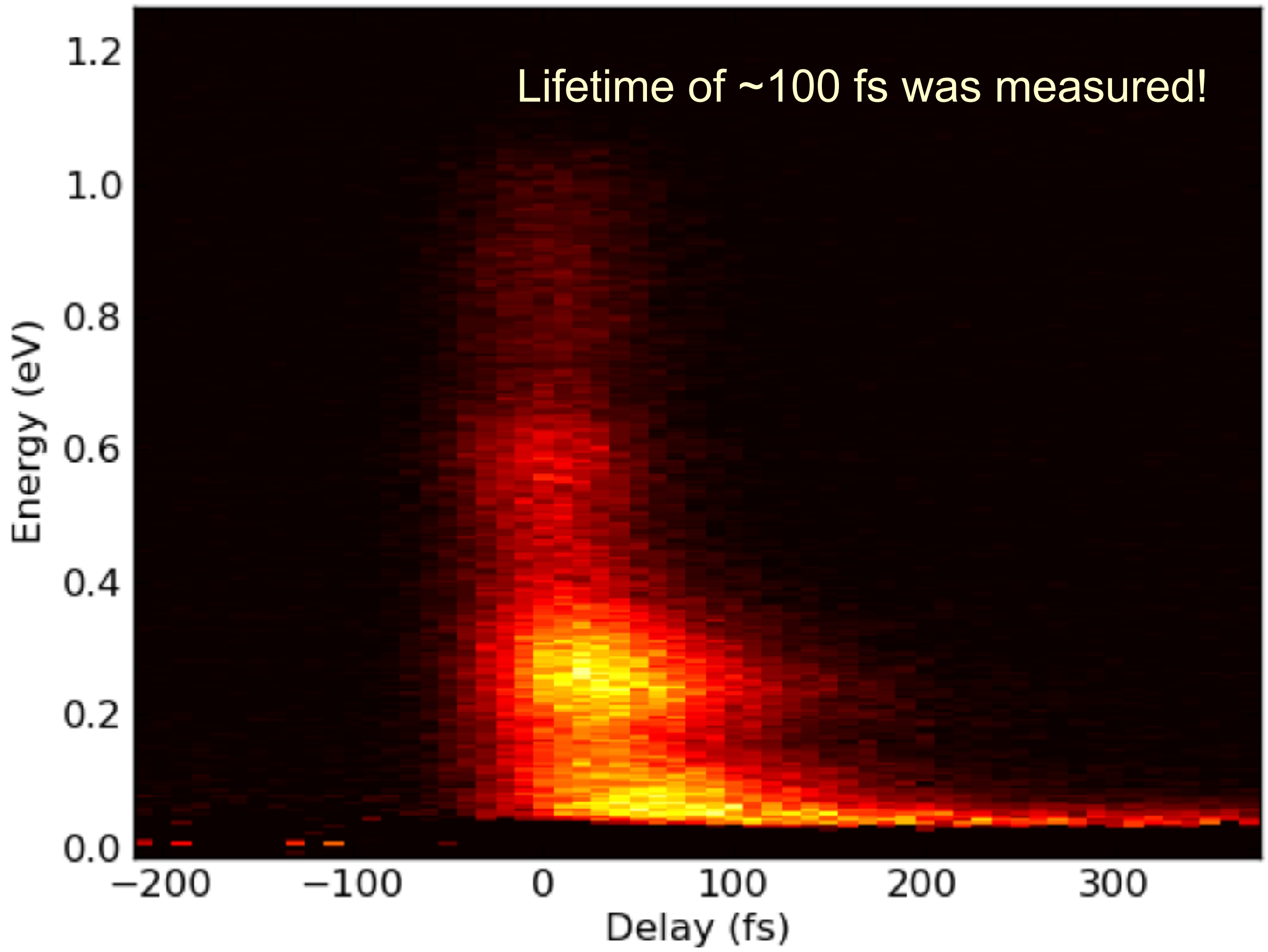
Experimental setup



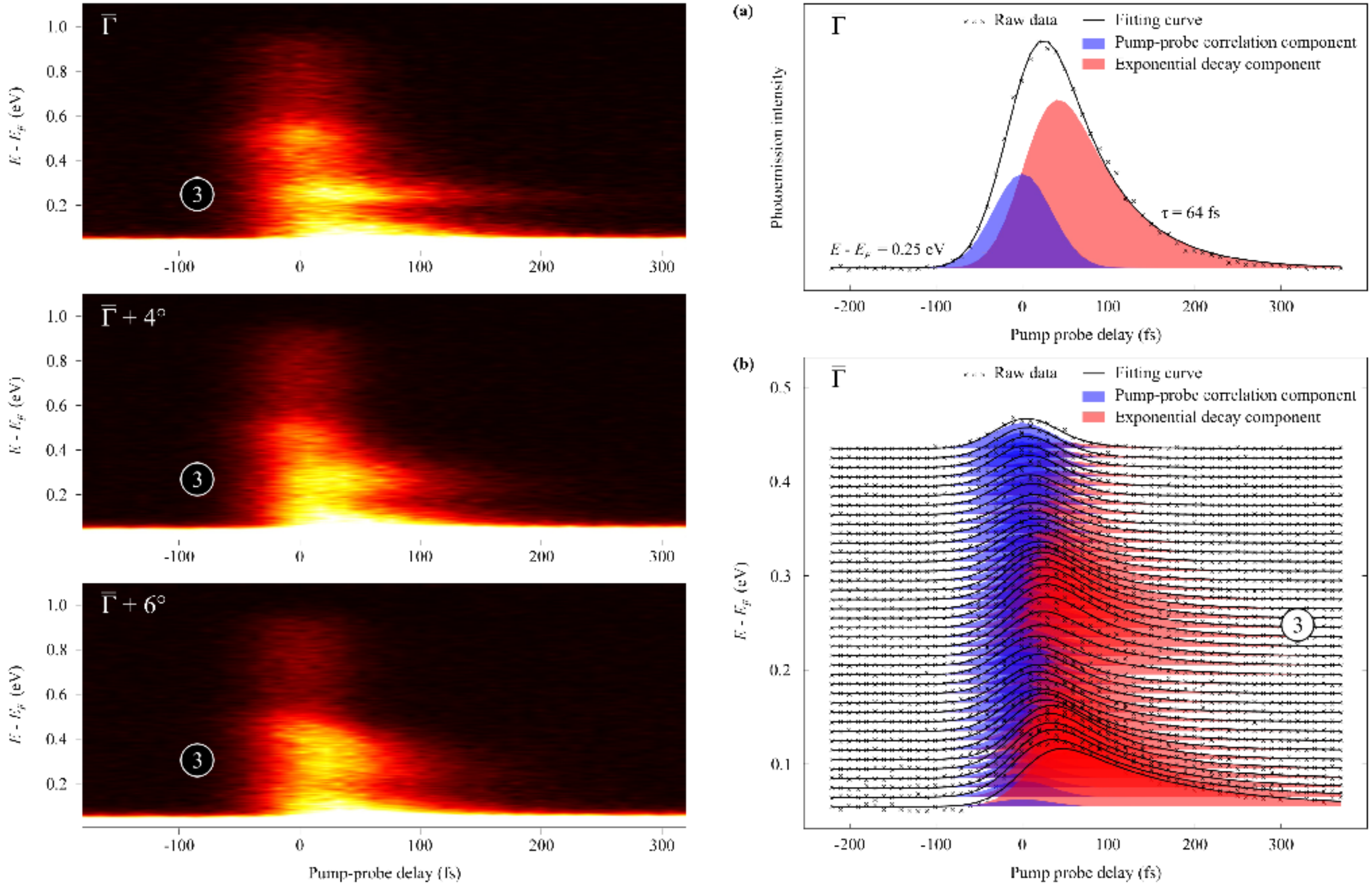
$YbRh_2Si_2$: Projected *p*-character band structure



Proof of principle pump-probe experiment

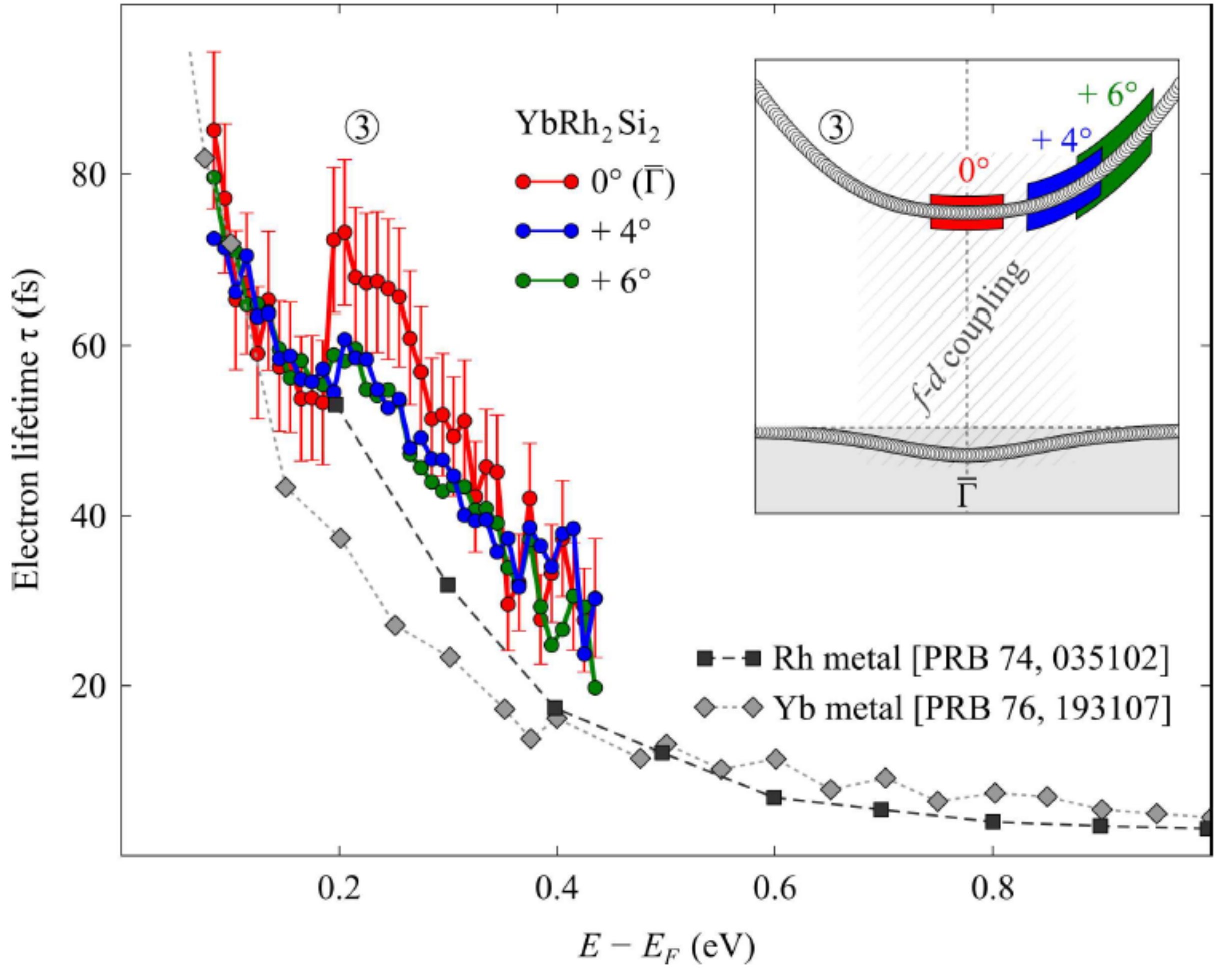


Angle-resolved pump-probe experiments



Pump-probe delay maps of the photoemission intensity above E_F and results of fit analysis

Lifetime of electrons above Fermi energy



Jump in electron lifetime around Γ points at deviation from Fermi liquid theory and can be related to strength of correlation between *d* and *f* electrons. Effect depends on energy gap between *d* and *f* states.

But is it really time scale of Kondo (*f*-*d*) interaction?

Cross sections of *d* and *f* electron excitations are extremely low at optical laser energies

Go to hν close (high harmonic generation, HHG) or above (XFEL) 100 eV!

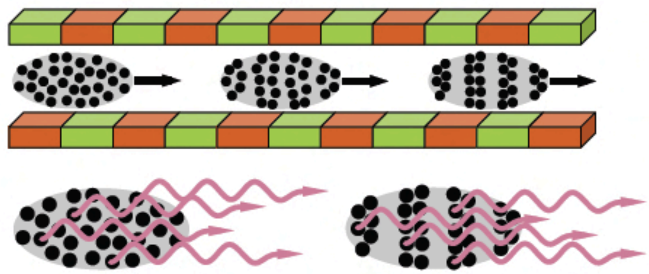
Photon sources for trARPES (Kai Rossnagel)

Sync. Rad. News **25**:5, 12 (2012)

Strobe lights: FEL versus HHG

FEL (FLASH)

$L = 27\,\mathrm{m}$



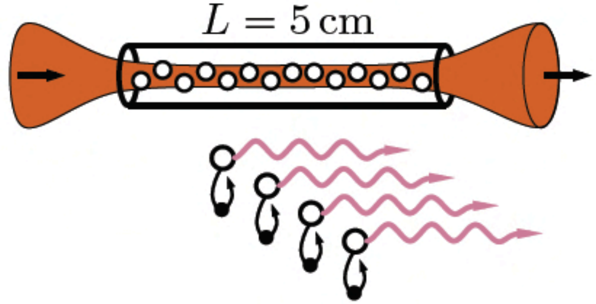
$h\nu \approx 25 - 300\,\mathrm{eV}$
(fundamental)

$I \approx 10^{10}\,\mathrm{photons/s}$
(space-charge & rep-rate limited)

$\tau > 10\,\mathrm{fs}$

HHG

$L = 5\,\mathrm{cm}$



$h\nu < 100\,\mathrm{eV}$
(practically)

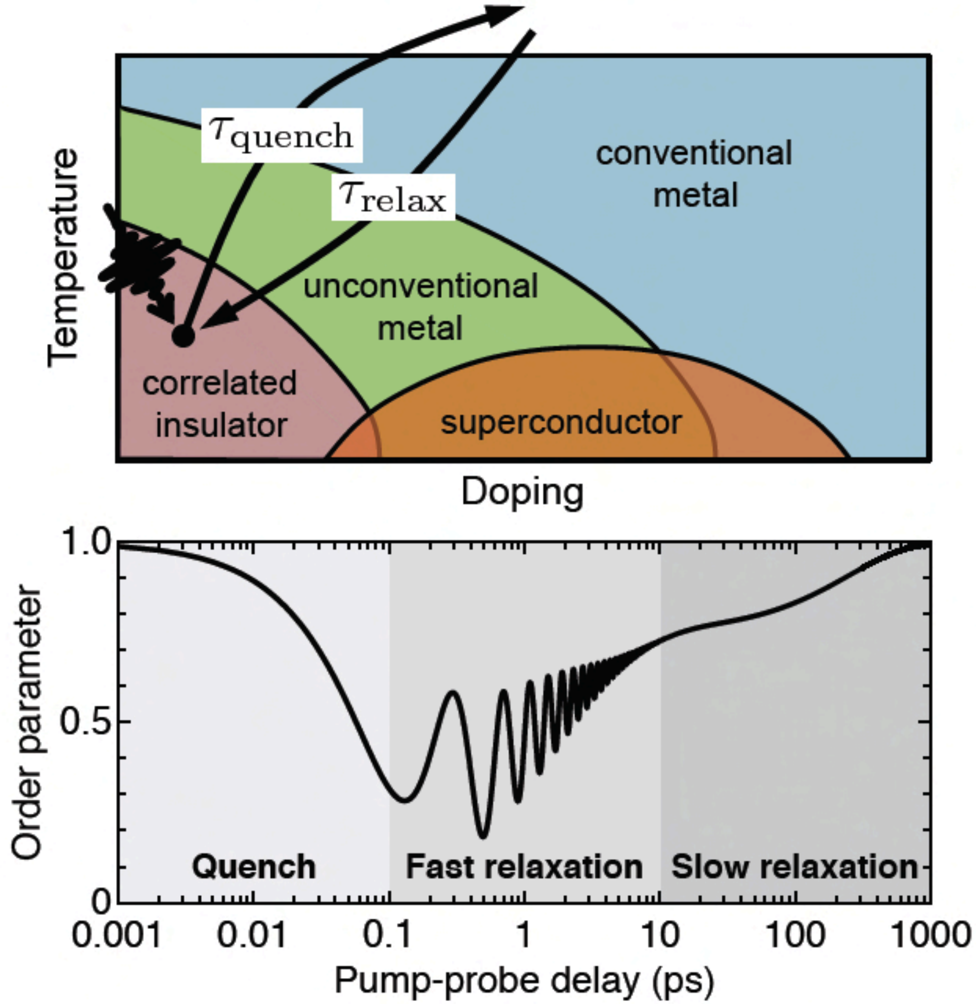
$I \approx 10^{10}\,\mathrm{photons/s}$

$\tau \lesssim 10\,\mathrm{fs}$

European XFEL

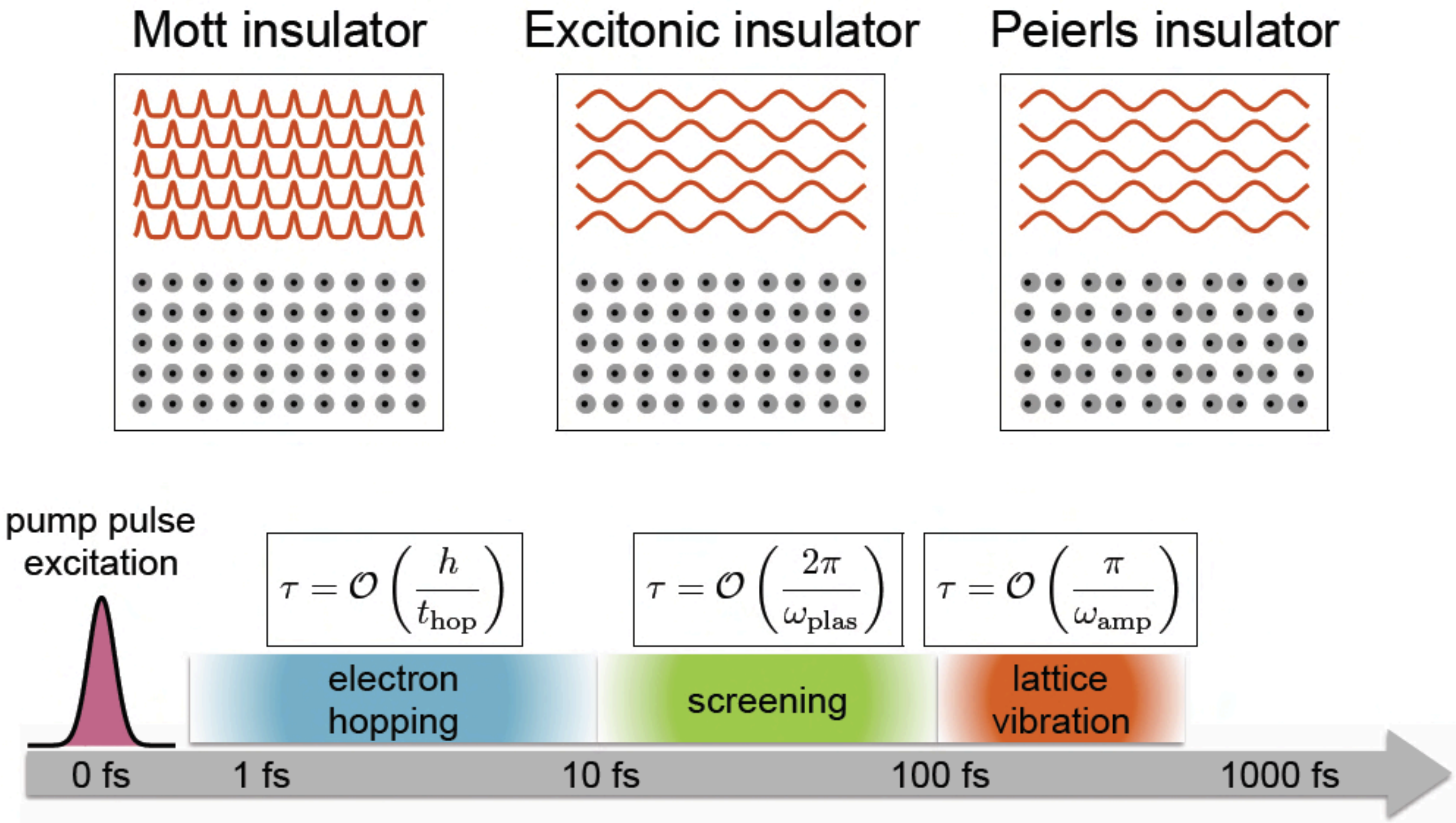
Study by destruction

Nature of condensed matter phases



Time scale of different interactions

Time-domain classification (learning by destroying)



What can be done with HHG sources?

Nature **471**, 490 (2011)

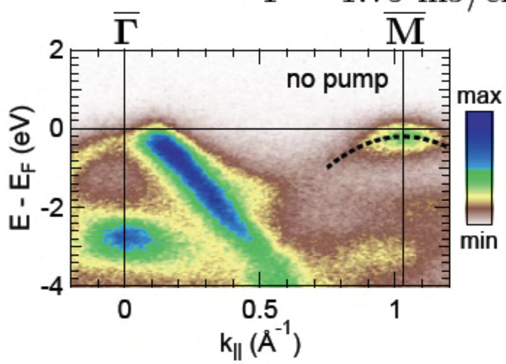
trARPES using HHG

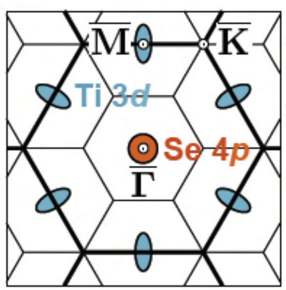
$T = 125\,\text{K}$

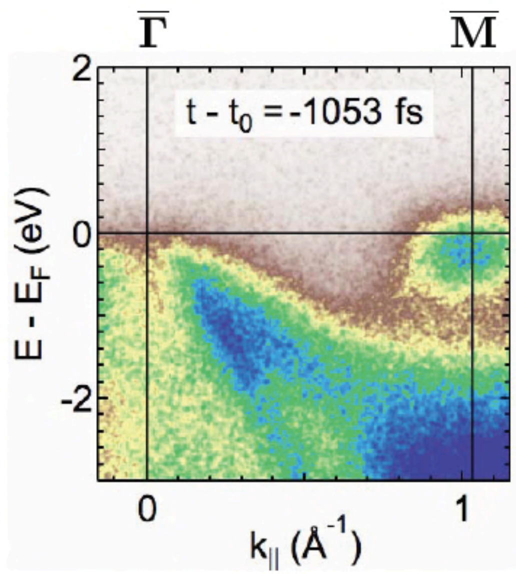
$h\nu_{\text{pump}} = 1.57\,\text{eV}.$ $\boxed{h\nu_{\text{probe}} = 43\,\text{eV}}$

$\Delta E \approx 400\,\text{meV}.$ $\Delta t \approx 30\,\text{fs}$

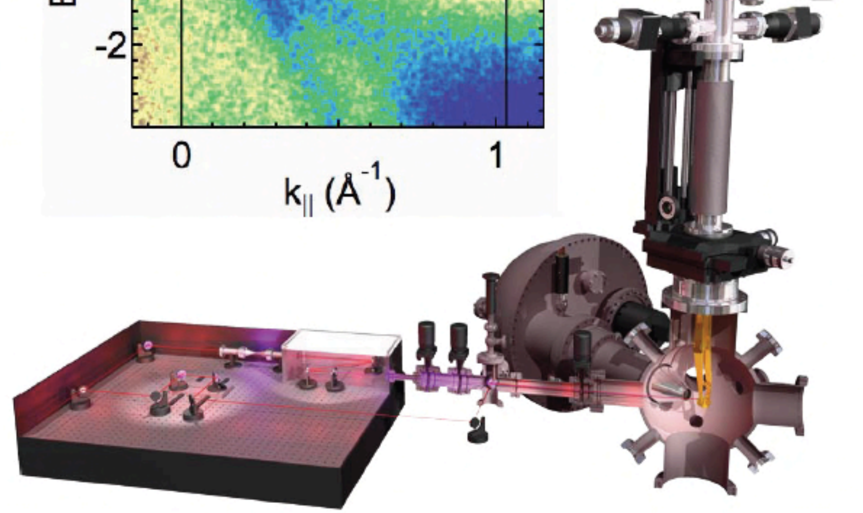
$F = 1.75\ \text{mJ/cm}^2$







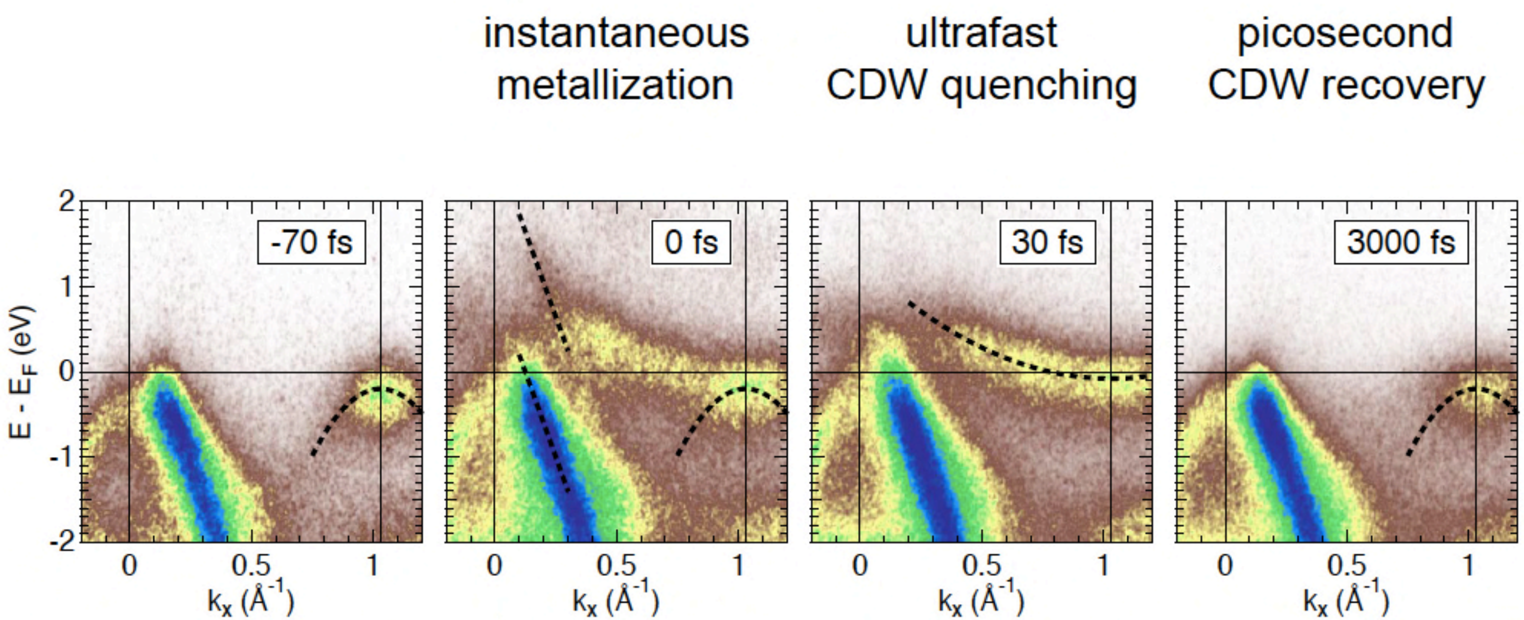
HHG setup
Bauer group, Kiel



What can be done with HHG sources?

Nature **471**, 490 (2011)

Snapshots



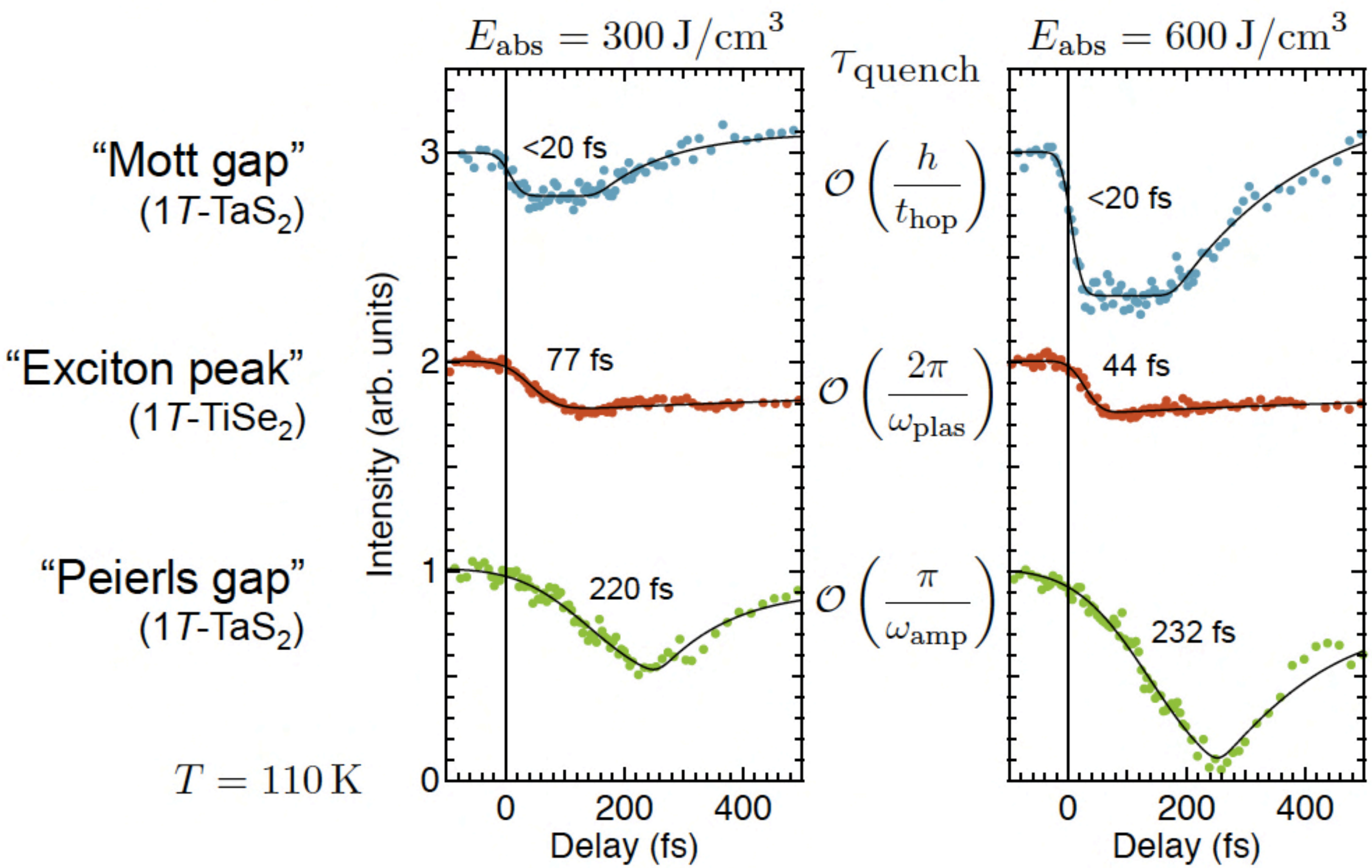
s-polarized probe

$F_{\mathrm{abs}} = 5\,\mathrm{mJ/cm^2}$

Examples of pump-probe research

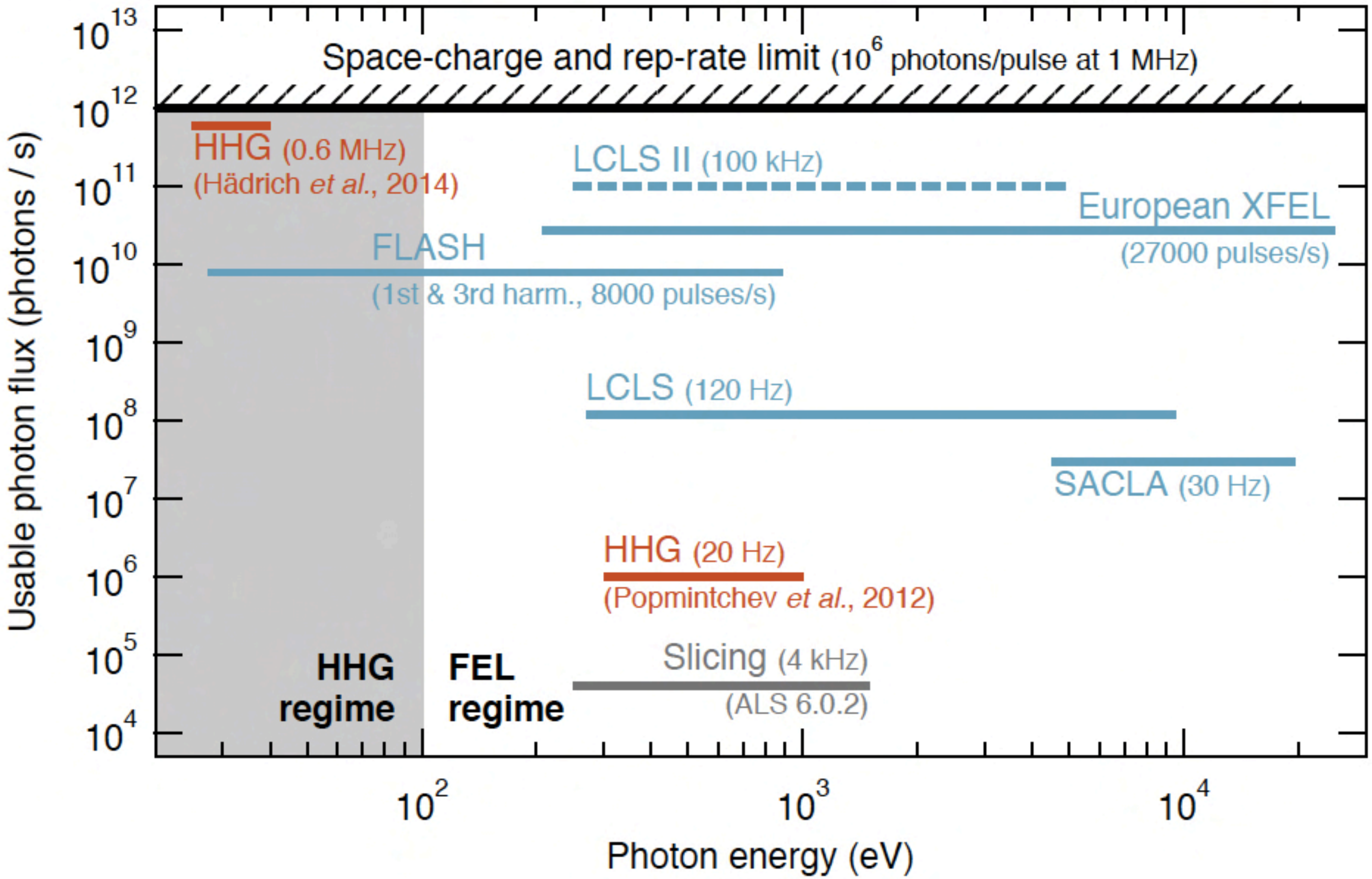
Nature Commun. **3**, 1069 (2012)

Hierarchy of quenching times



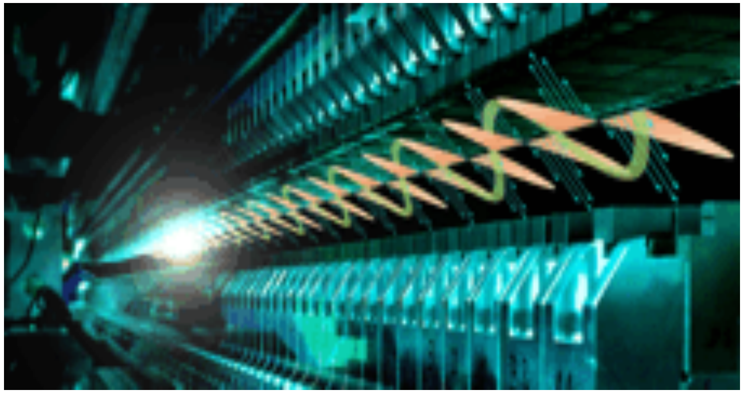
HHG and FELs: Complimentary tools

FEL photoemission

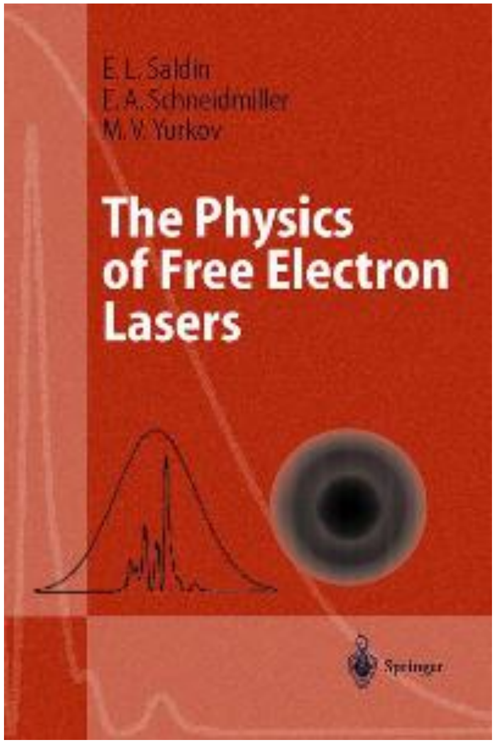


X-Rays

New Generation Sources Free Electron Lasers (FELs)

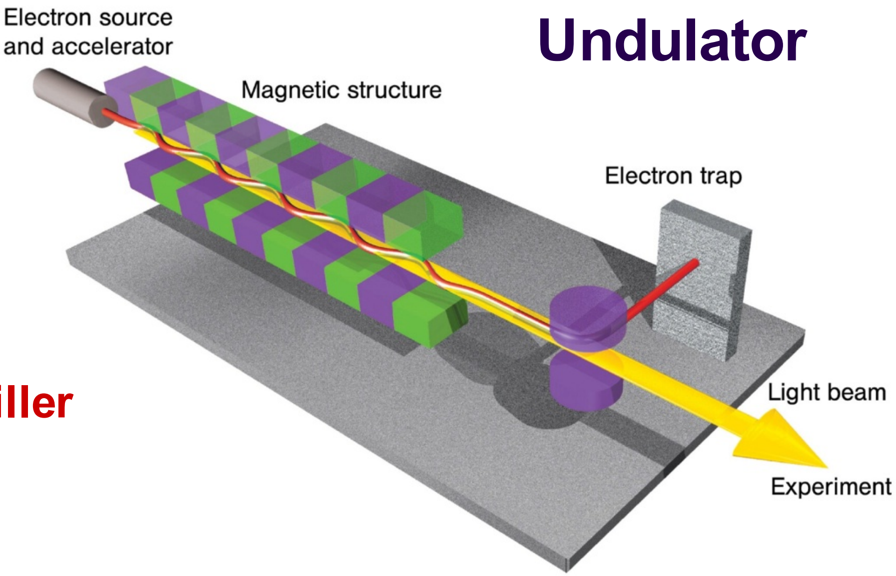


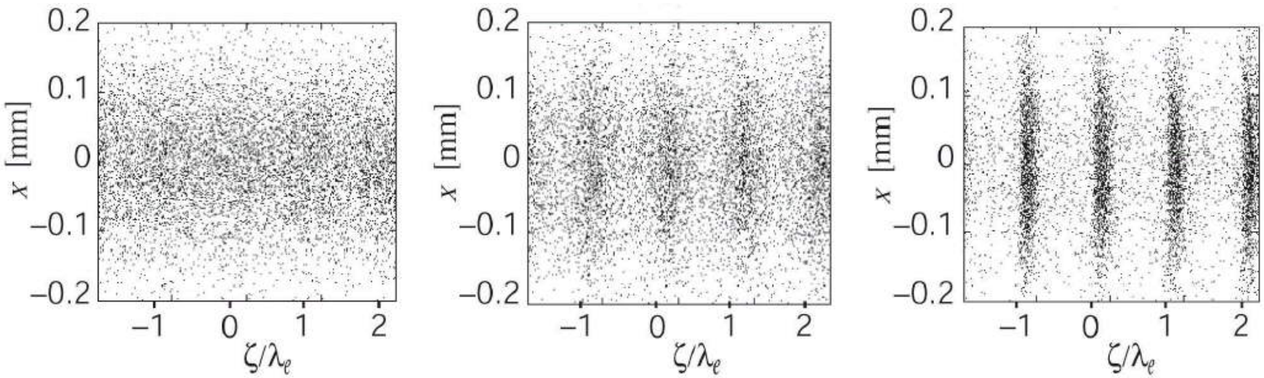
Basics of SASE FEL process



E.L. Saldin
E.A. Schneidmiller
M.V. Yurkov

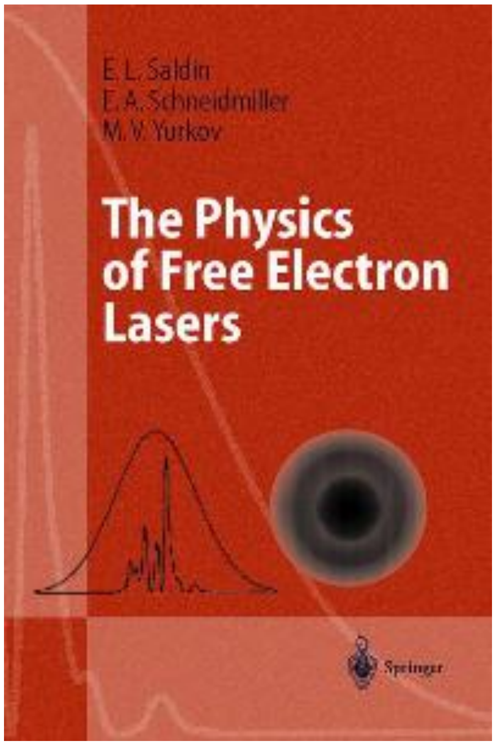
Undulator





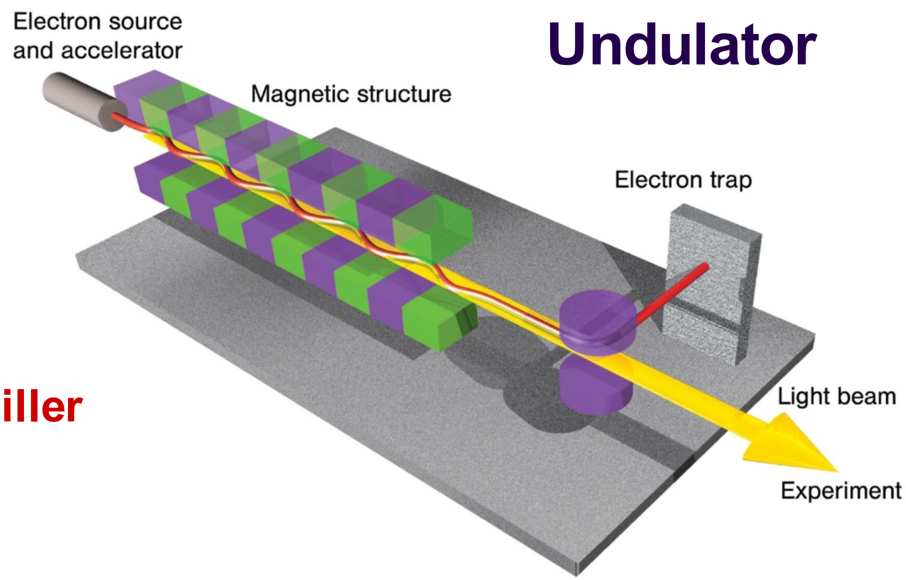
simulations at the radiation wavelength (λ_e), ζ – distance inside the undulator

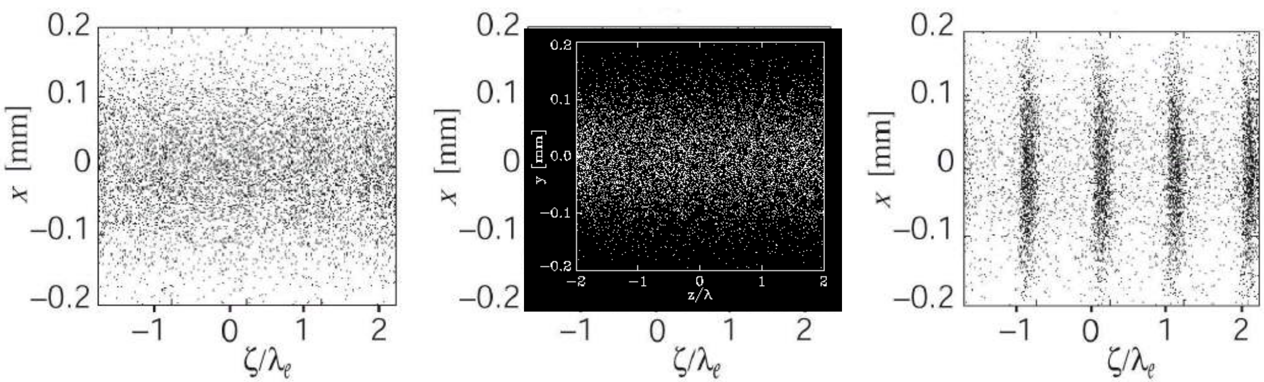
Basics of SASE FEL process



E.L. Saldin
E.A. Schneidmiller
M.V. Yurkov

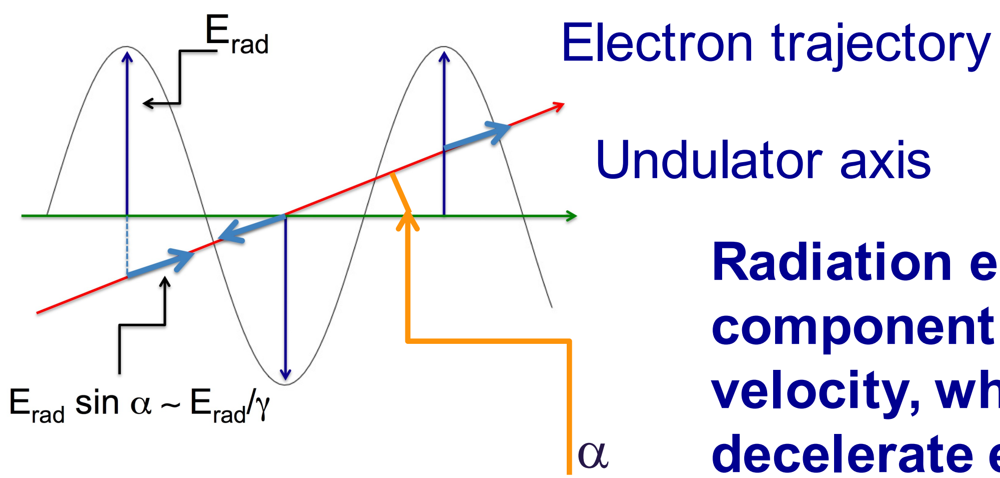
Undulator



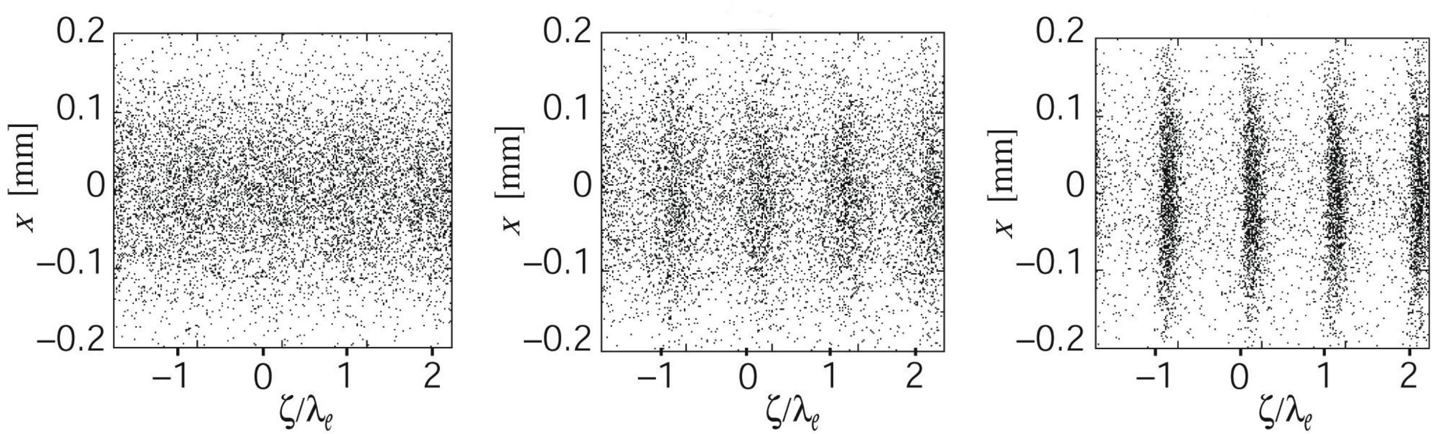


simulations at the radiation wavelength (λ_e), ζ – distance inside the undulator

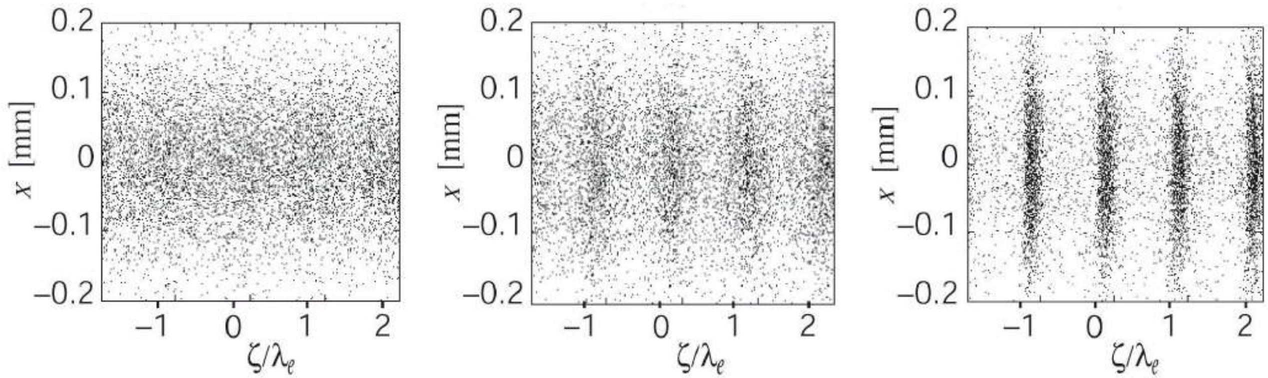
Origin of microbunching

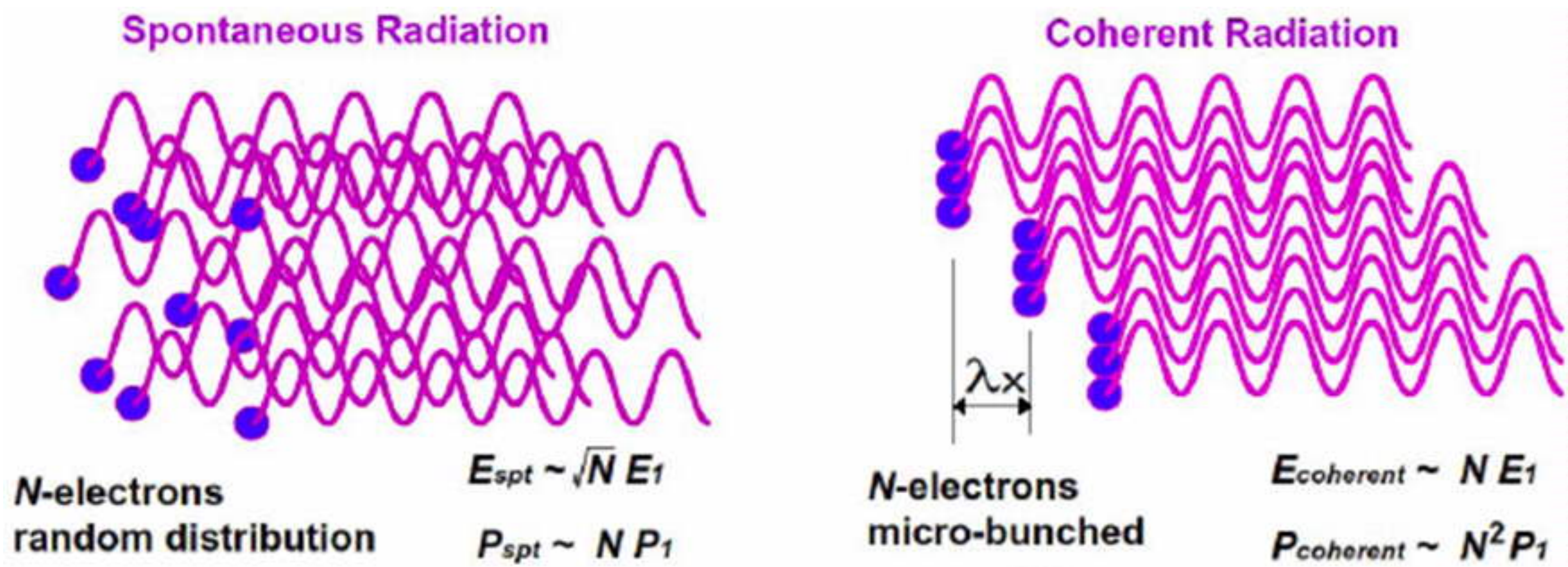


Radiation electric field has a small component parallel to electron velocity, which can accelerate or decelerate electrons

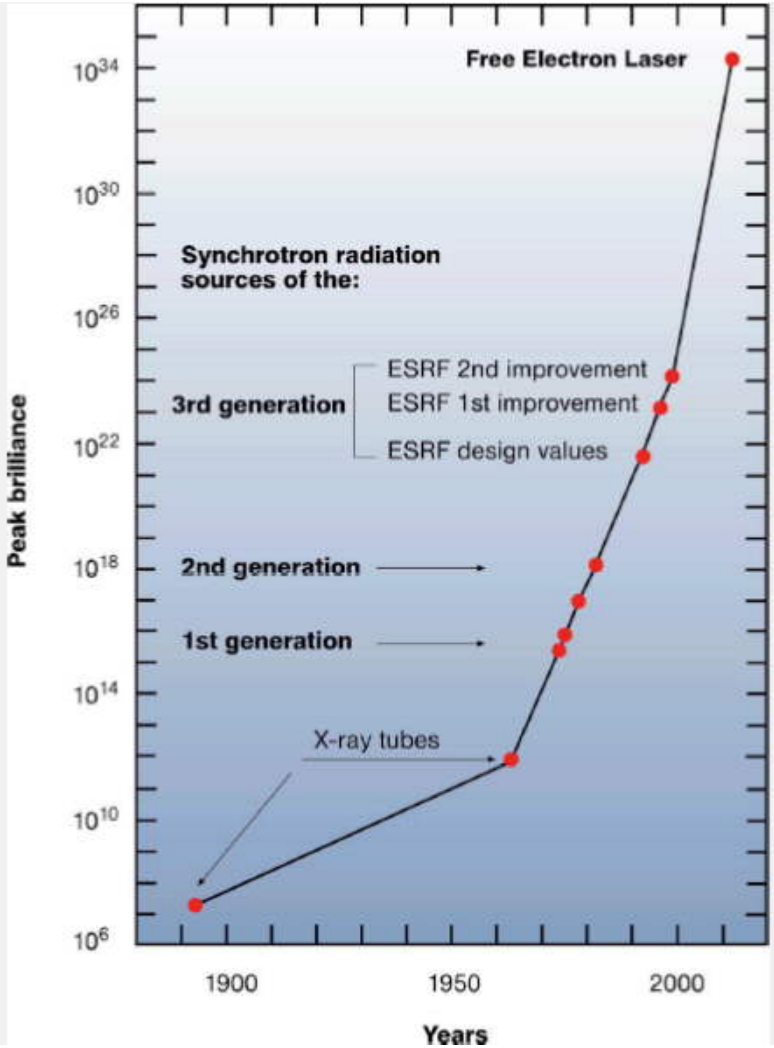


Spontaneous vs. coherent radiation in undulators





Peak brilliance of X-Ray sources vs. time



Free Electron Lasers:

-Based on Linear Accelerator

-Delivers ultrashort pulses

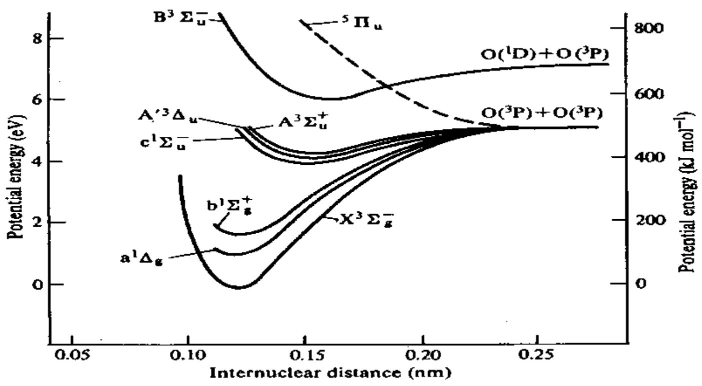
(100 fs = 0.1 ps= 10^{-13} s or less)

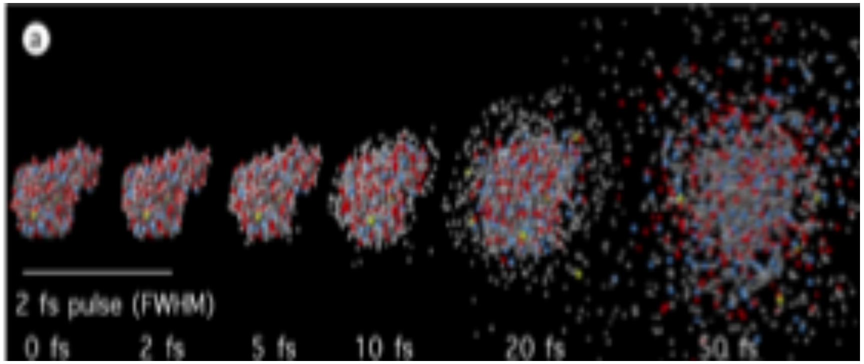
- (Transversely) Spatially coherent (laser-like) radiation

Wanted … More brilliant X-ray sources, with:

wavelength down to < 0.1 nm ==> *atomic-scale resolution*

ultrashort (<1 ps) pulses
==> "molecular movies"





ultra-high peak brightness,
transverse spatial coherence

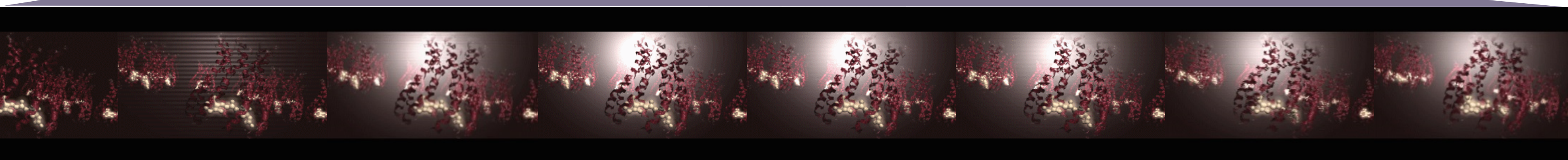
==> imaging of single nanoscale objects, possibly down to individual macromolecules (no crystals)

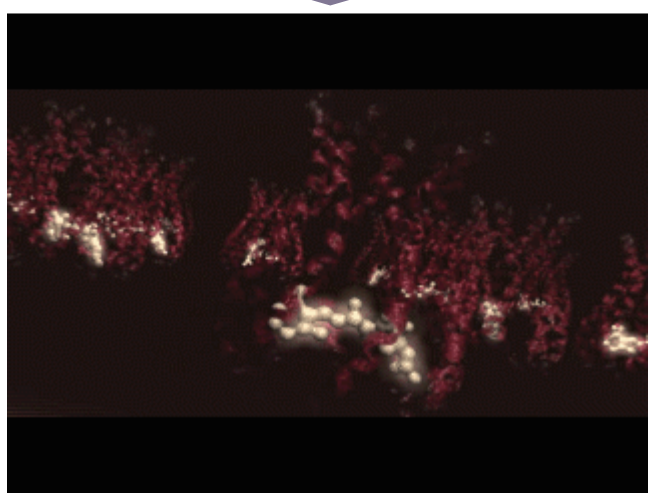
==> investigation of matter under extreme conditions…

Making molecular movies

Eadward Muybridge
1892

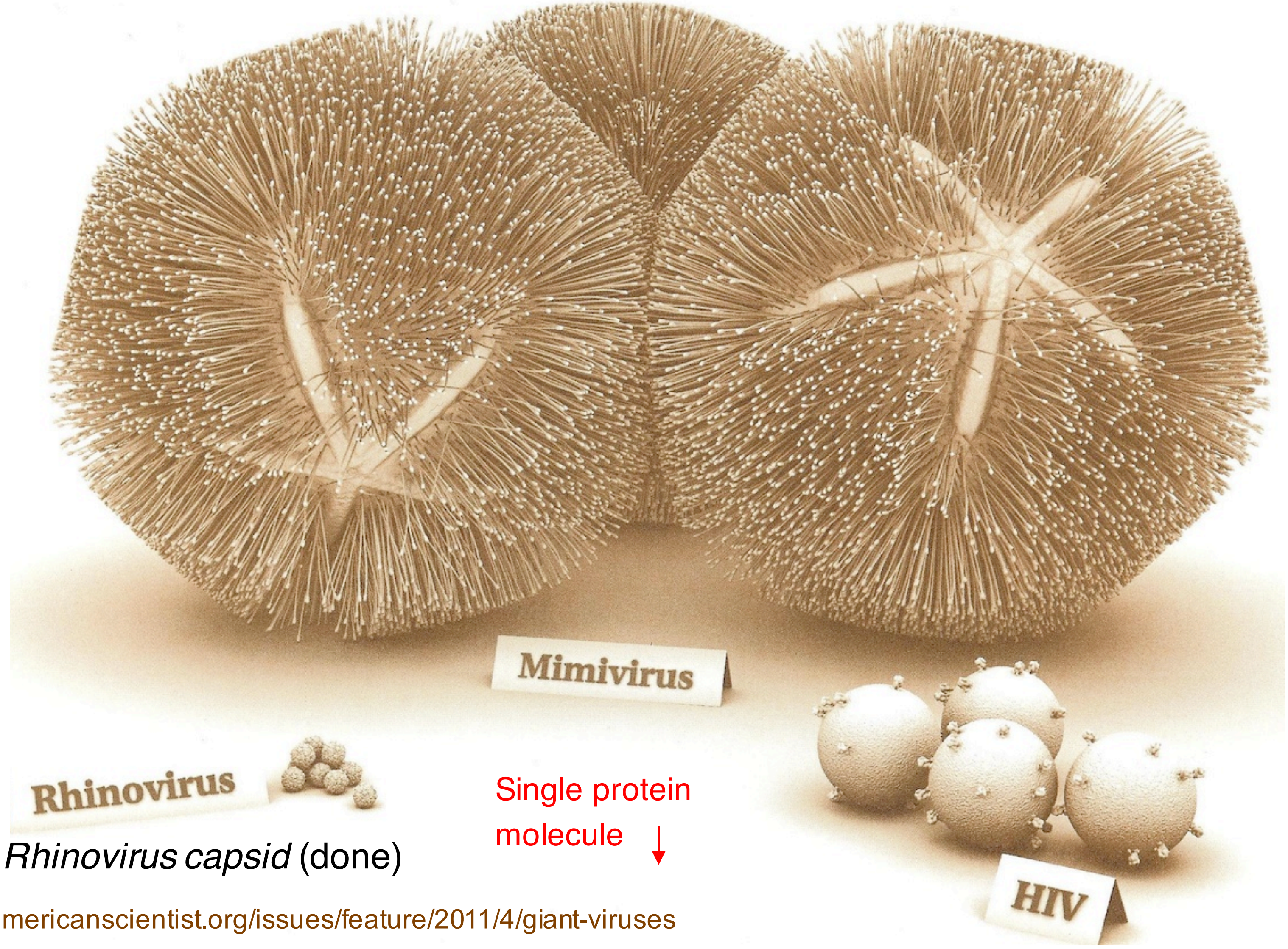
European XFEL
2017





European XFEL

Tremendous variety of bio-objects to be studied



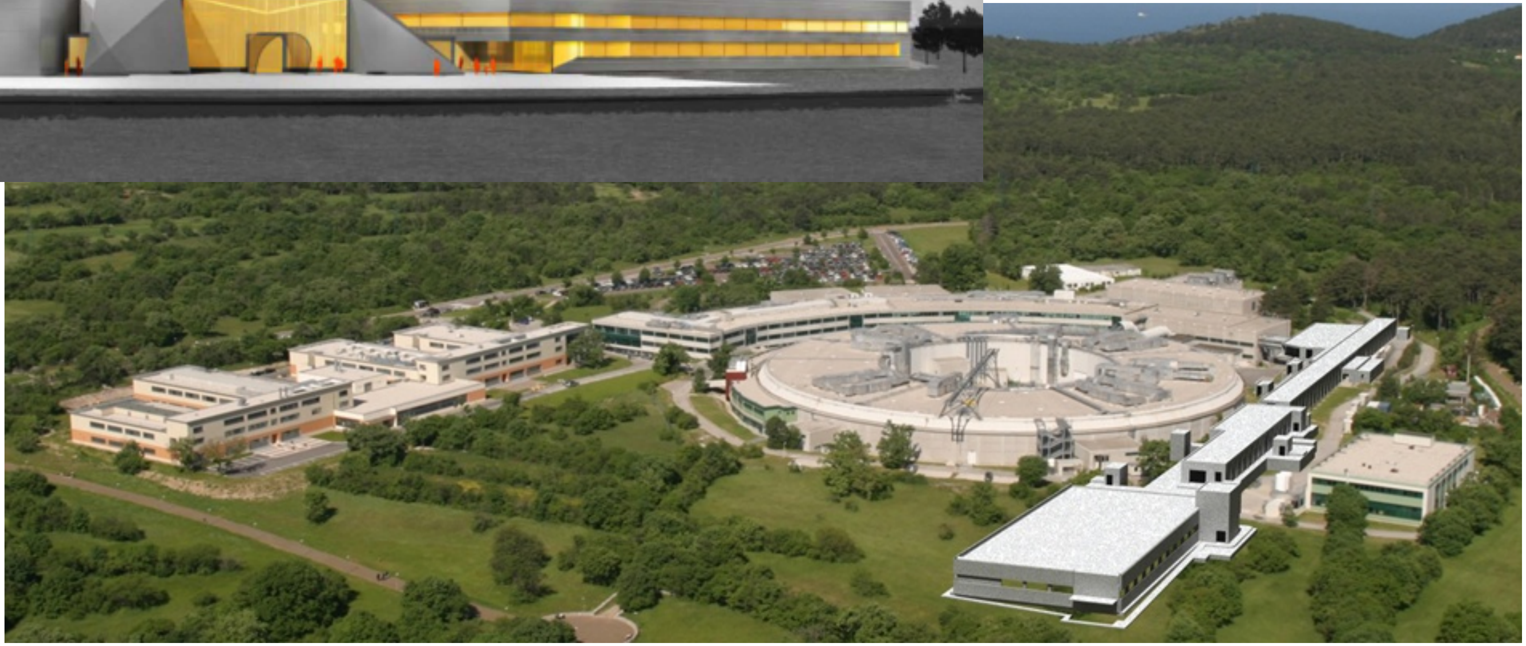
Single protein molecule ↓

Rhinovirus capsid (done)

http://www.americanscientist.org/issues/feature/2011/4/giant-viruses

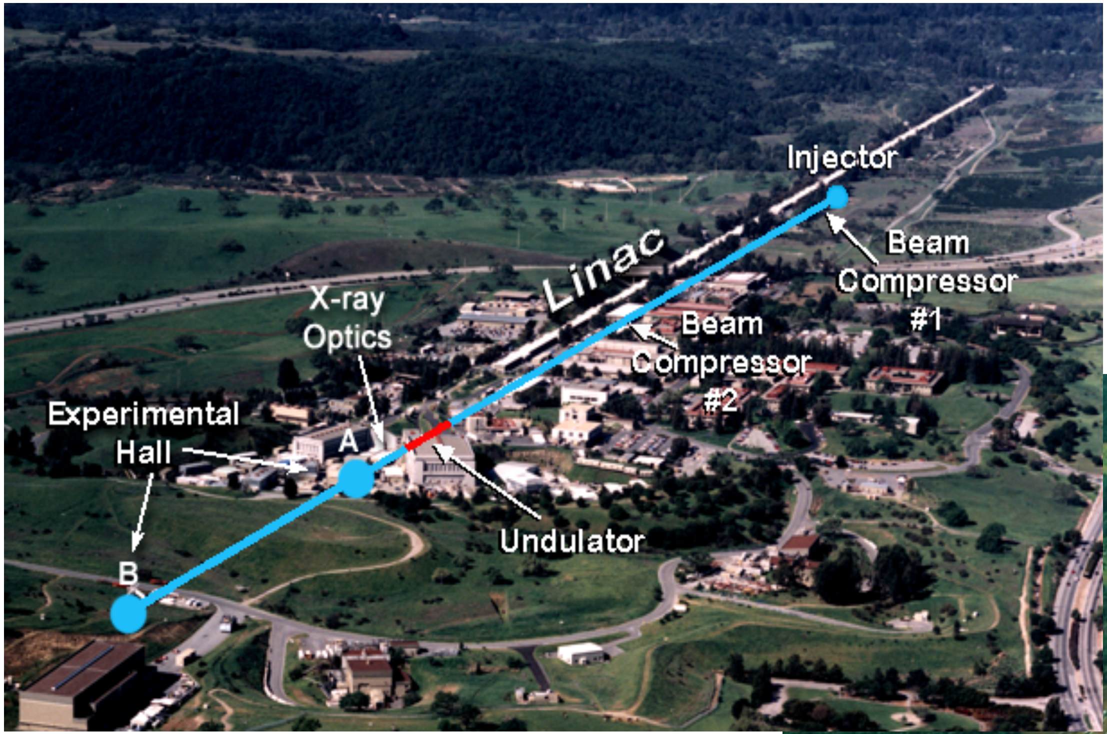
Soft X-ray projects

FLASH / DESY



FERMI / ELETTRA

Hard X-Ray FEL facilities



2011 - 60 p/s

SCSS

SPring-8 Compact

SASE Source

2009 -120 p/s

LCLS

LINAC COHERENT

LIGHT SOURCE

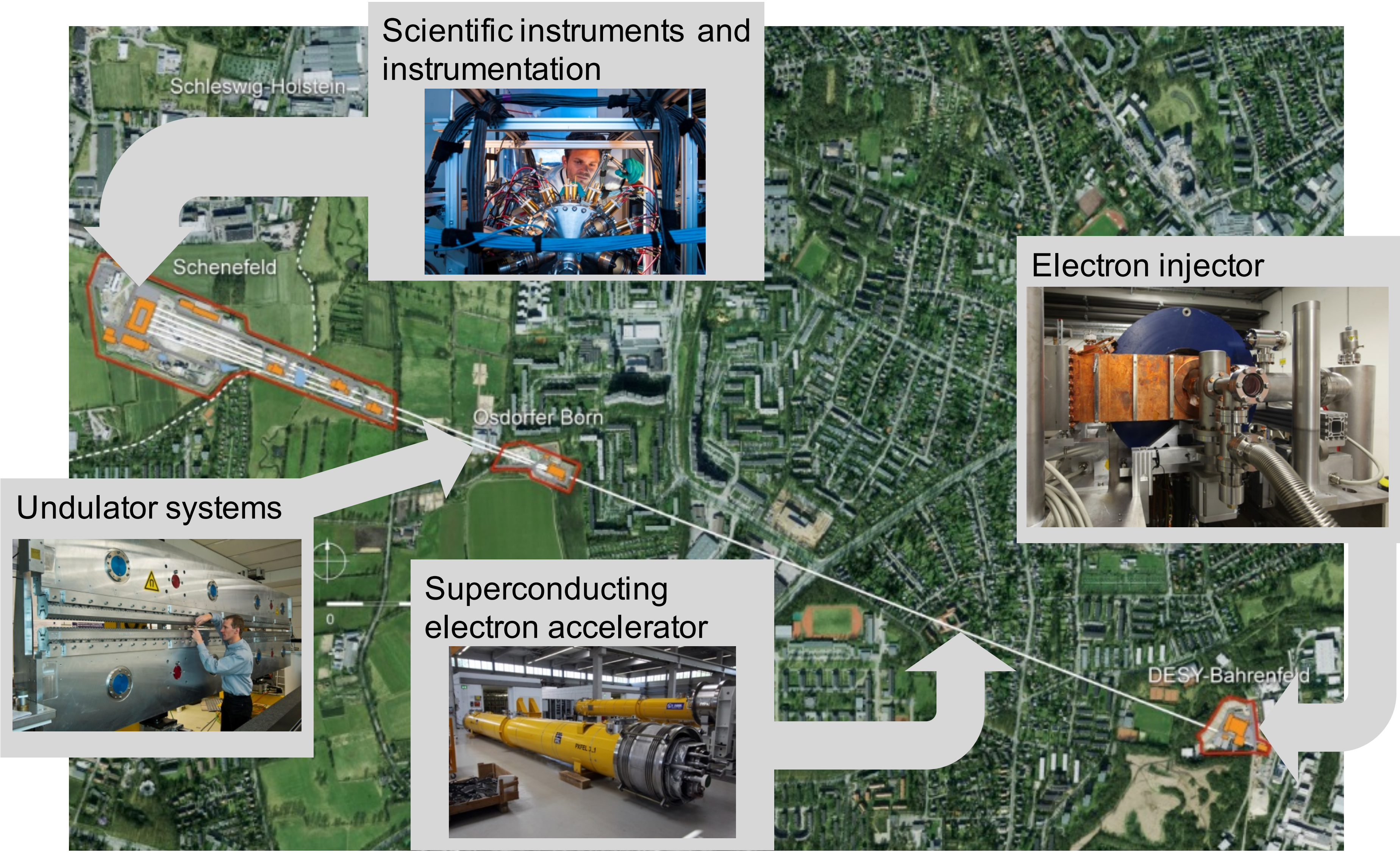
Hamburg, 30.11.2009: the European XFEL Convention Signing Ceremony

Total costs ≈1.500 MEUR

European XFEL

European XFEL - a leading new research facility

How it works – a closer look at the facility



European XFEL – a leading new research facility

The European XFEL is a research facility, now under operation, which is using high-energy X-ray light to help scientists better understand the nature of matter.

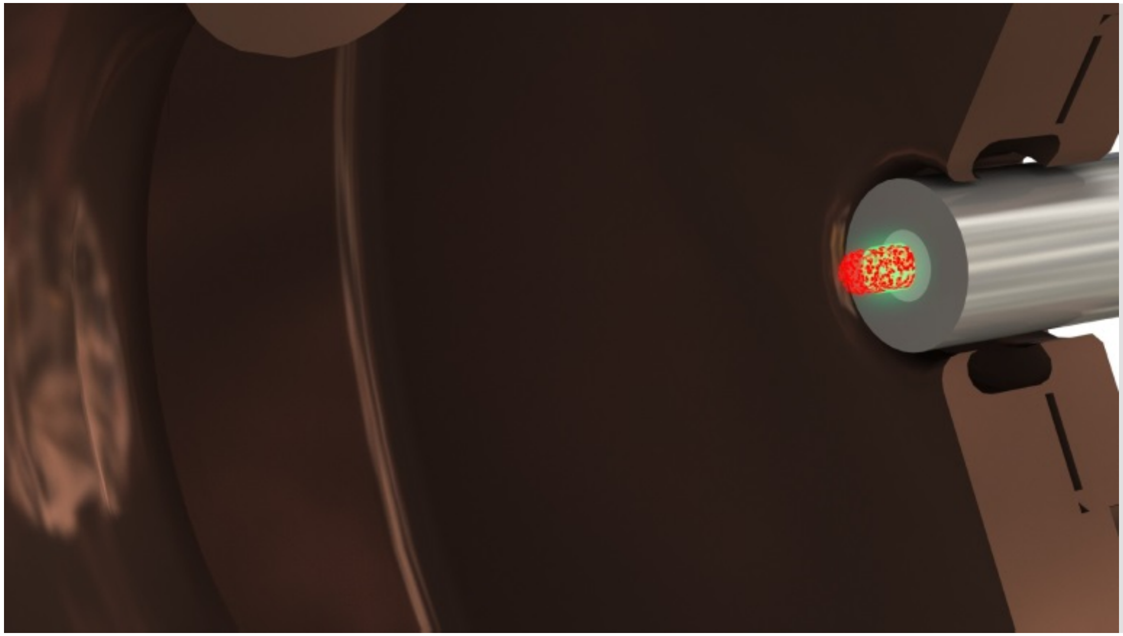
Schenefeld & Hamburg, Germany

User facility with 260 staff (+ 230 from DESY)

2017: Start of user operation

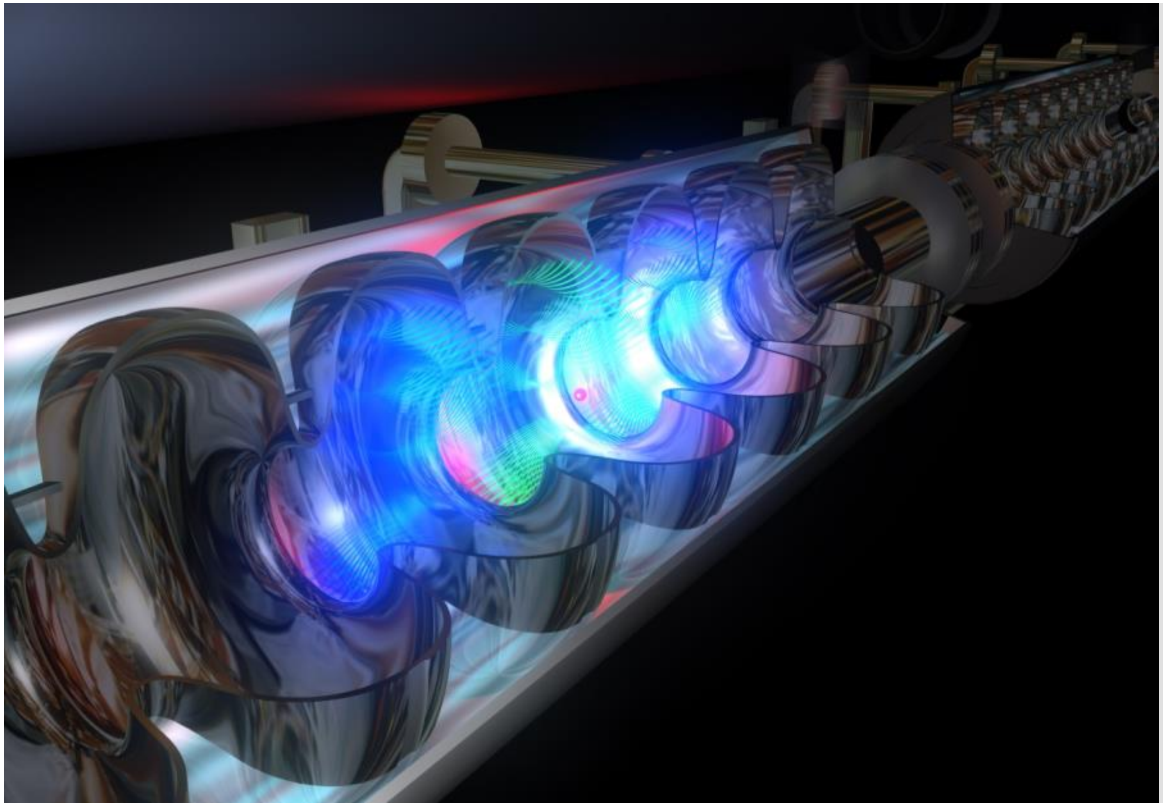
Site photo taken on 9 August 2013
at the start of user operation

Injector: creating bunches of electrons



- Optical laser strikes Cs_2Te surface, releasing a cloud of electrons
- Electrons move into a magnetic field, shaping into a bunch
- Small accelerator module "fires" bunch into the main electron accelerator

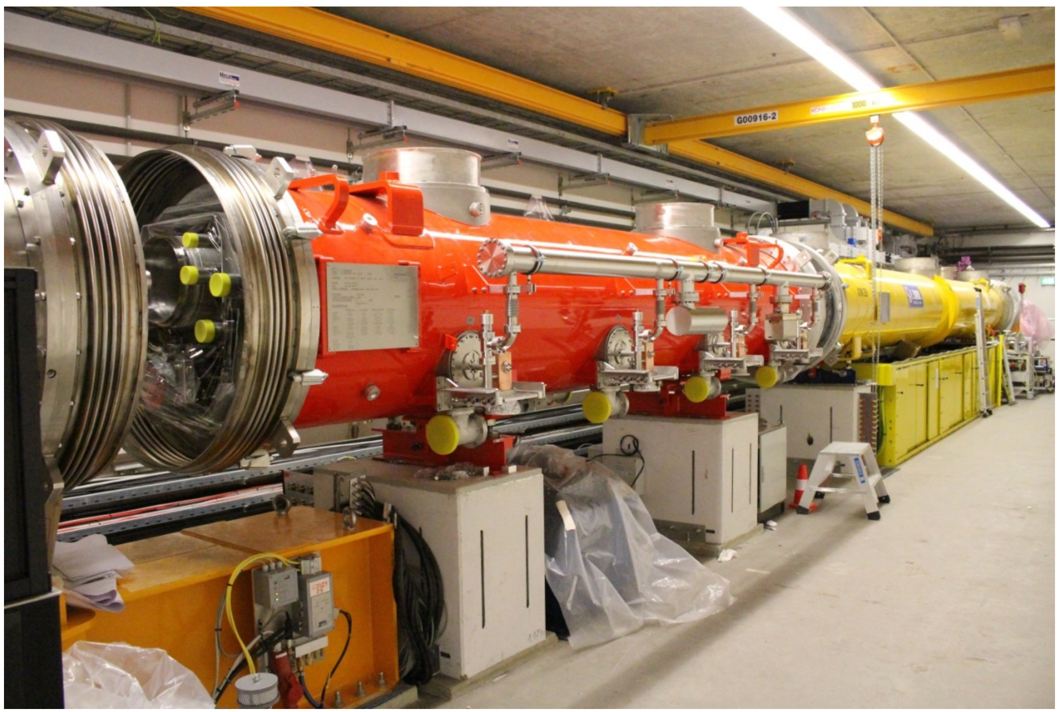
Accelerator: electrons at close to light speed

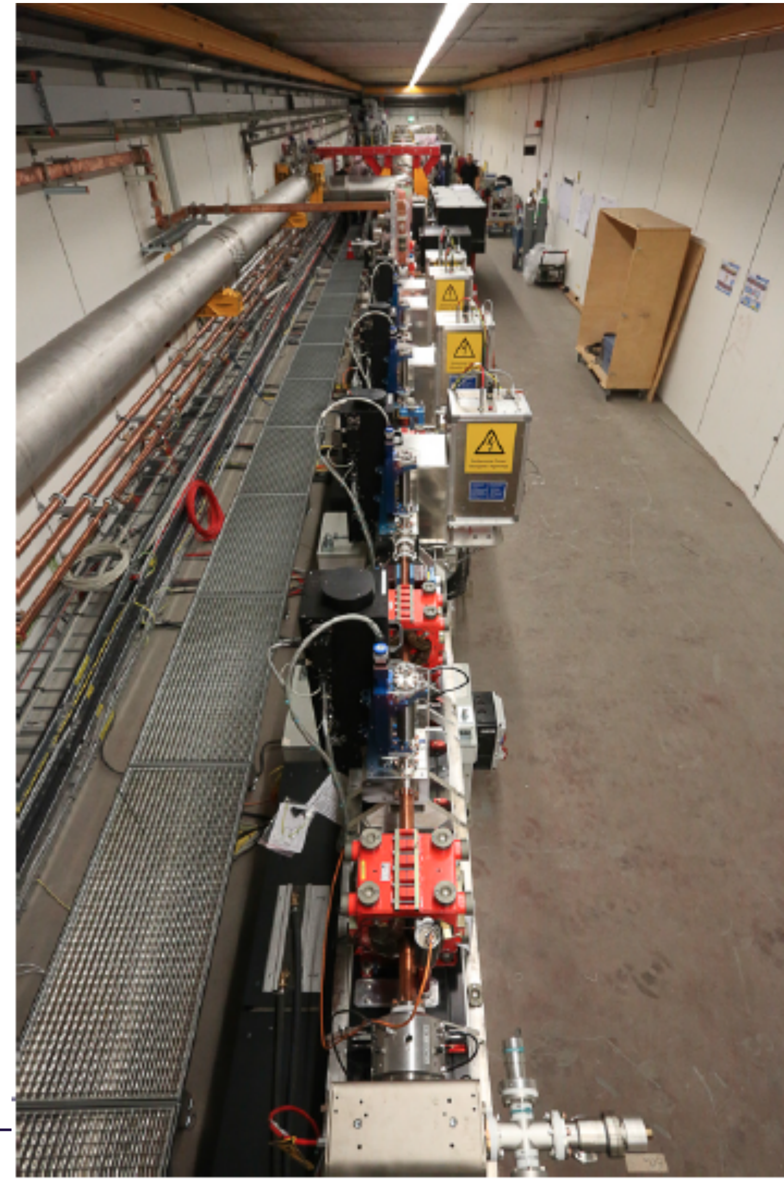


- 100 accelerator modules over 2 km bring the electron bunch to near light speed and high energies
- Superconducting niobium cavities powered by intense radio frequency accelerate electrons

First accelerated electron beam in the injector

- **Injector commissioning started**, injector tunnel closed, cool down to 2 K successful.
- First 130 MeV Electron beam on 18.12.2015!





European XFEL

Testing accelerator modules prior to installation

28 March 2014

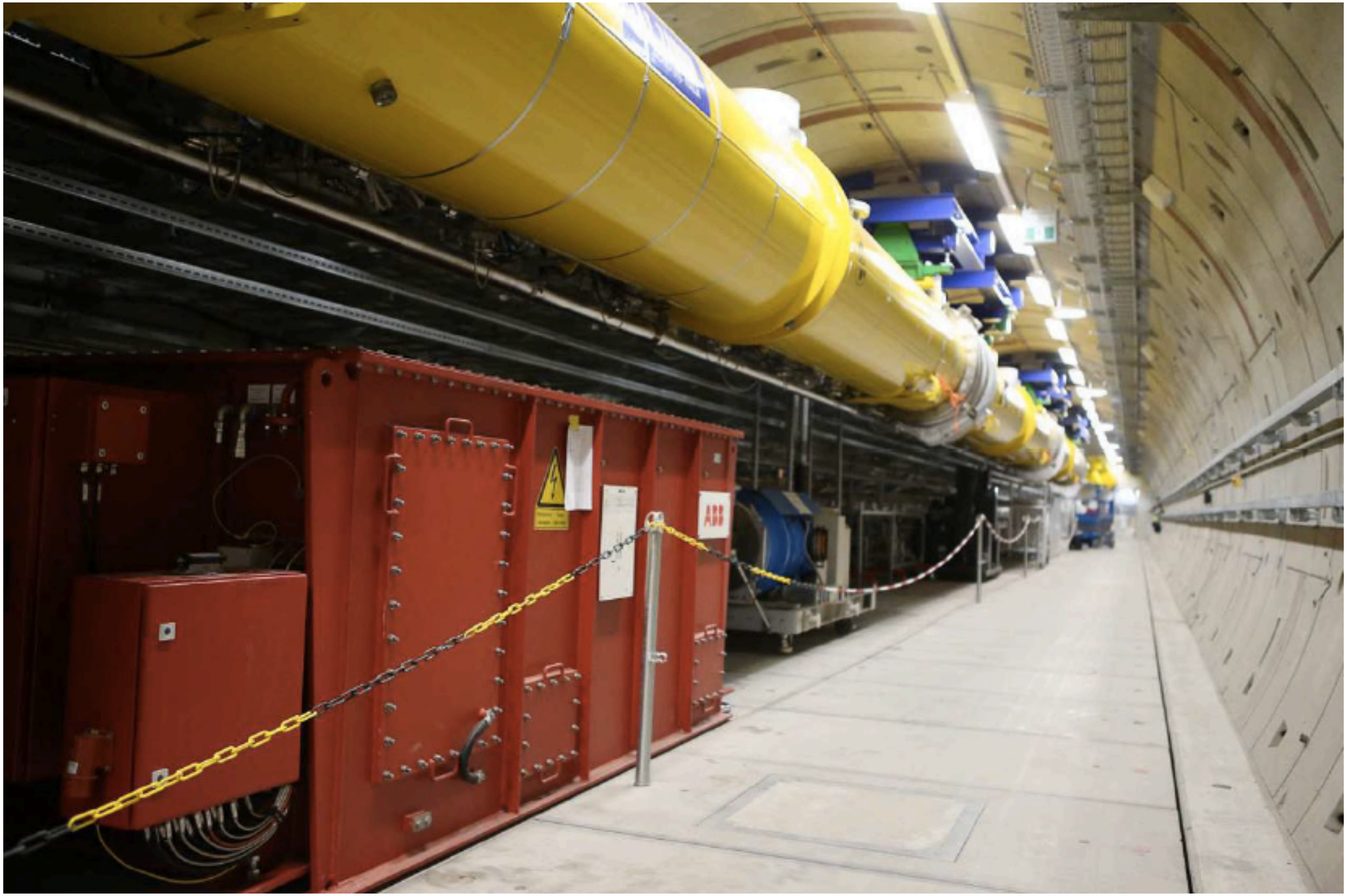
Accelerator module test facility

3 June 2014

Injector complex DESY-Bahrenfeld

European XFEL

Power RF – Installation check



Tunnel branch Osdorfer Born (2017)

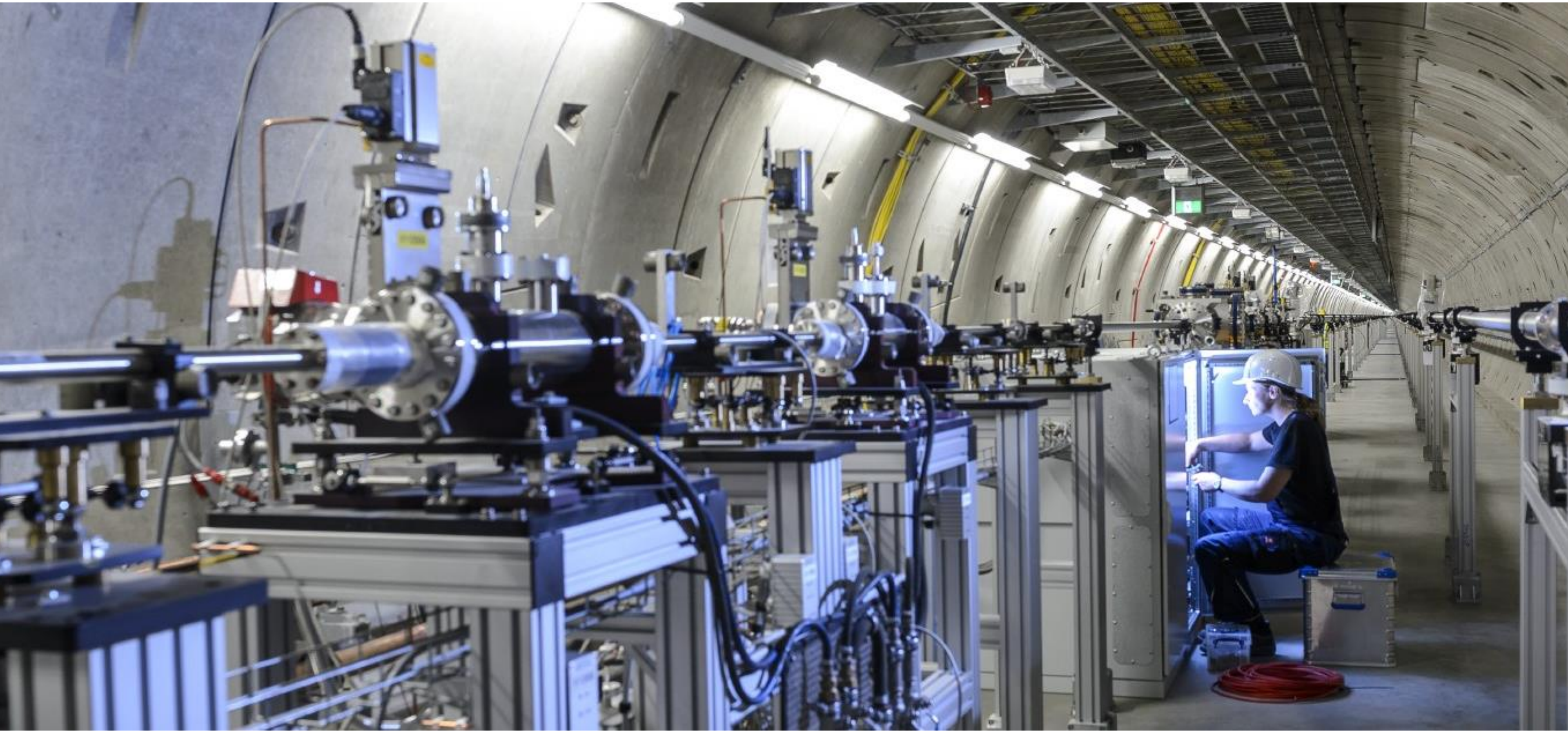
European XFEL

Aligning the undulators

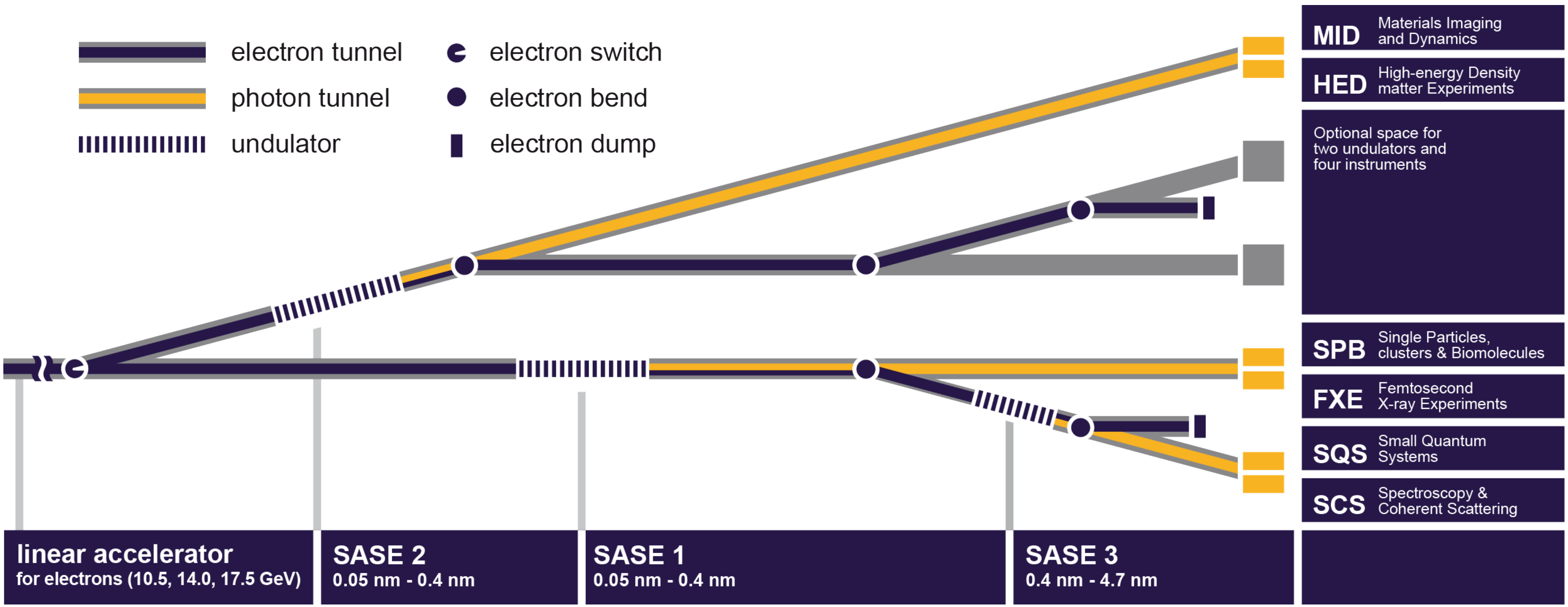
Undulators in tunnel

Optical elements of the SASE1 beamline

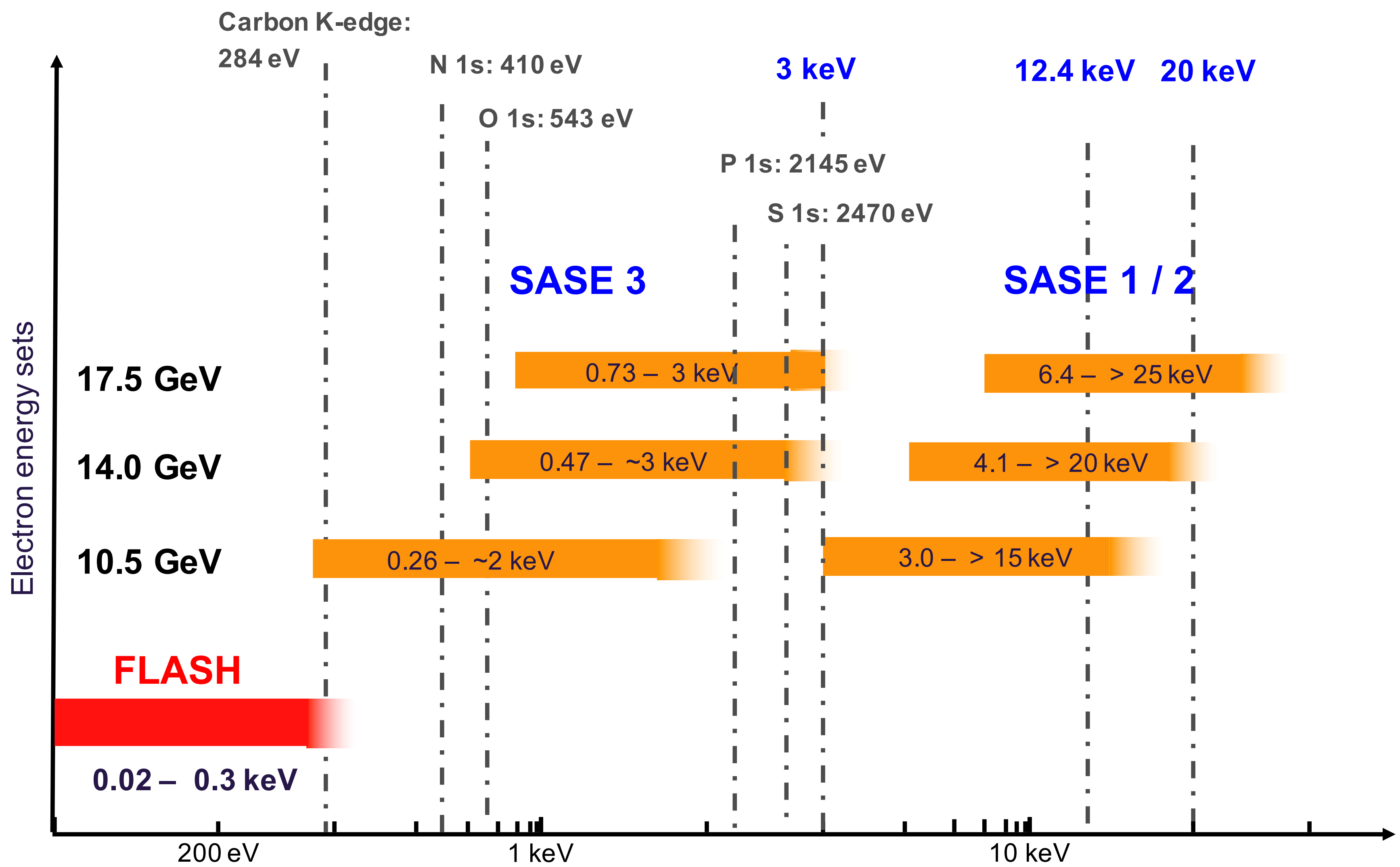
Photon beamlines



Beamline layout & experiment stations



Photon energy ranges



Scientific instruments

Hard X-rays

SPB: Single Particles, Clusters, and Biomolecules

- Will determine the structure of single particles, such as atomic clusters, viruses, and biomolecules

MID: Materials Imaging and Dynamics

- Will be able to image and analyze nano-sized devices and materials used in engineering

FXE: Femtosecond X-Ray Experiments

- Will investigate chemical reactions at the atomic scale in short time scales—molecular movies

HED: High Energy Density Physics

- Will look into some of the most extreme states of matter in the universe, such as the conditions at the center of planets

Soft X-rays

SQS: Small Quantum Systems

- Will examine the quantum mechanical properties of atoms and molecules.

SCS: Spectroscopy and Coherent Scattering

- Will determine the structure and properties of large, complex molecules and nano-sized structures.

SASE3 Undulator

General Soft X-Ray radiation parameters

Pulse widths	**2 – 100 fs**	**Coherence time**	0.3 – 1.8 fs
Pulse energy	**0.2 – 11.0 mJ**	**Bandwidth**	0.25 – 0.7 %
Peak power	50 – 120 GW	**Number of photons**	**$0.1 – 2 \times 10^{14}$**
Average power	3 – 300 W	**Average flux of photons**	$0.3 – 5.4 \times 10^{18}$
Beam size	40 – 80 µm	**Average brilliance**	$0.03 – 2.6 \times 10^{24}$

Rep. rate **10 Hz (2700 pulses in bunch train) = 27.000 pulses/s**

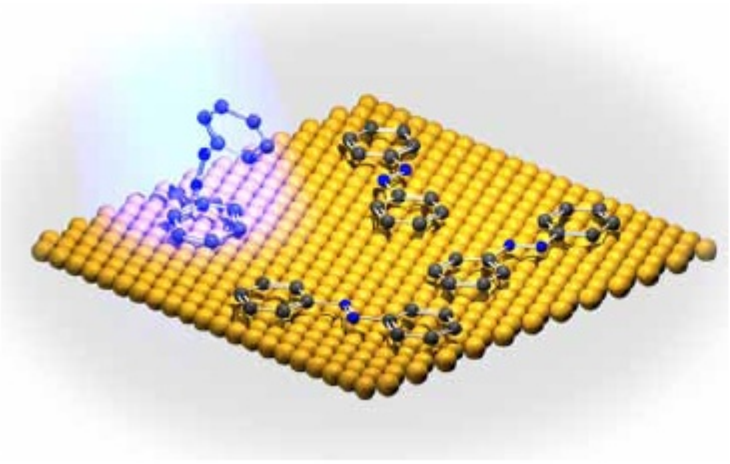
Parameter	Unit					
Bunch charge	pC	20	100	250	500	1000
Pulse duration (FWHM)	fs	2	9	23	43	107

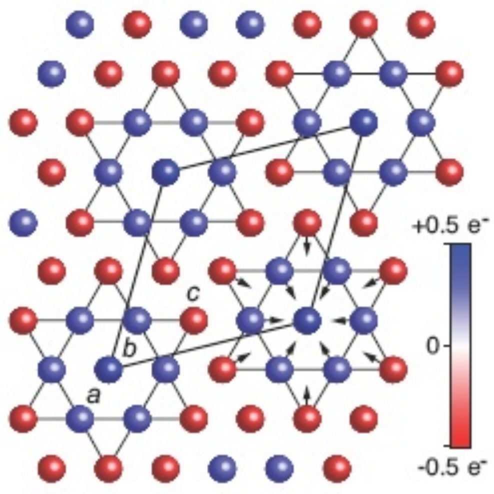
Science
@
SCS Scientific Instrument
(trXPS and trARPES)
Spin-resolved photoemission (?)

Photoemission at FELs: Motivation

Surface chemical reactions

Charge order dynamics



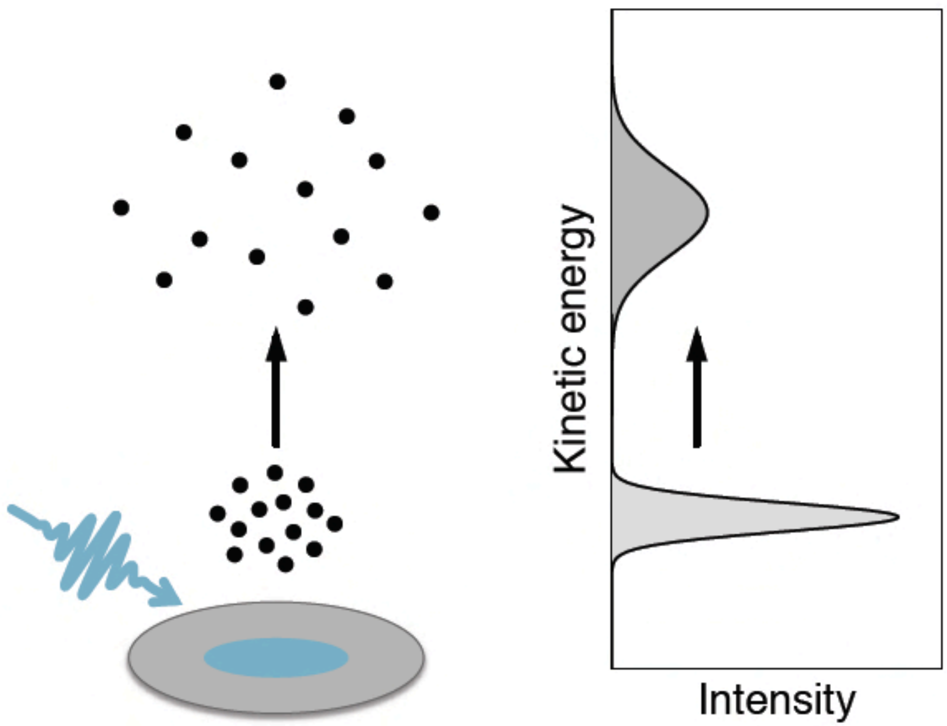


Time scale: 10 fs - 1 μs, reversible processes are preferable

Pump-probe experiments at XFELs

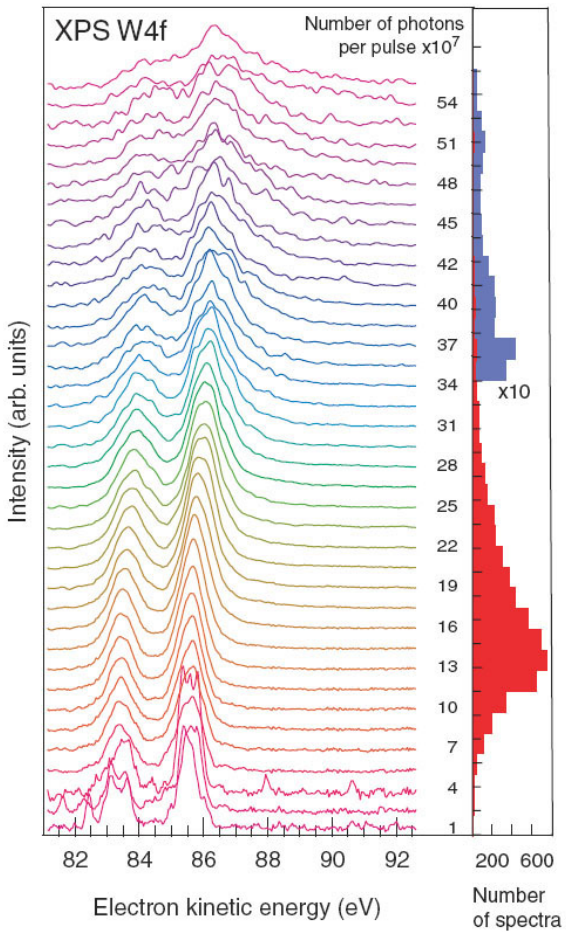
Problems to be solved (Kai Rossnagel)

Challenge I:
Vacuum space-charge effects

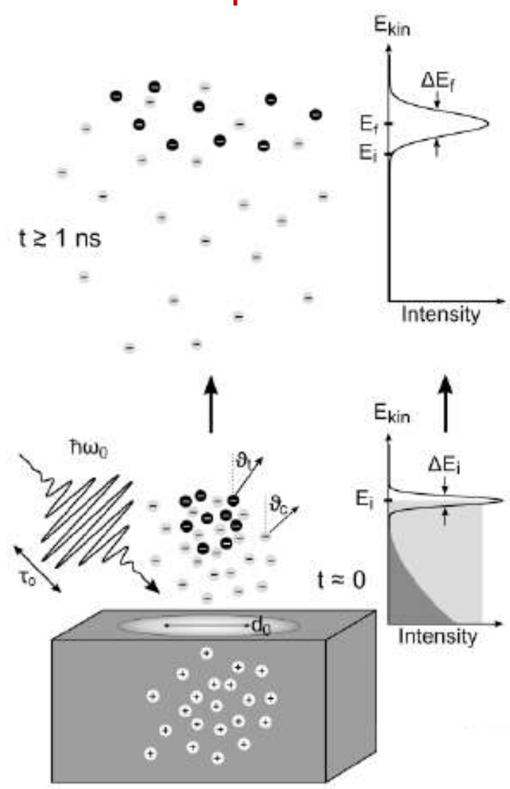


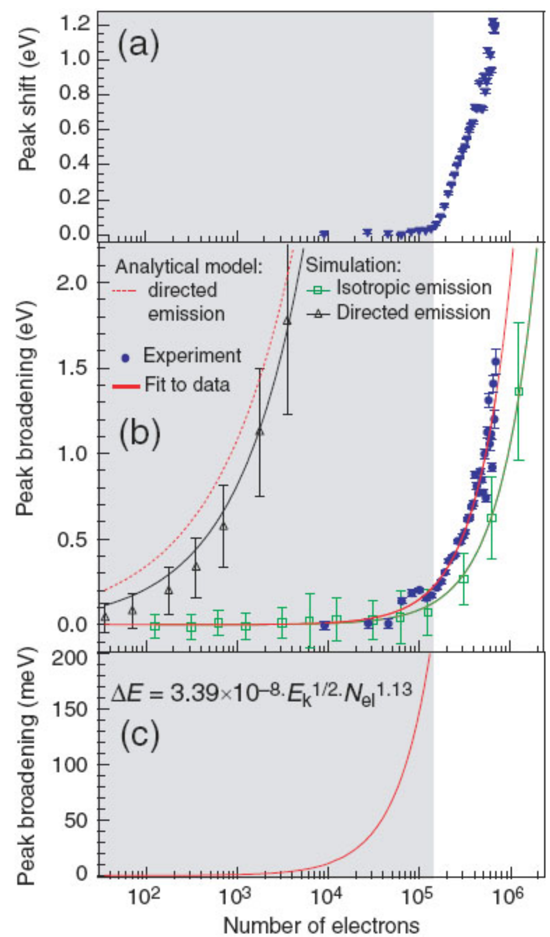
Core-level X-ray photoemission spectroscopy (XPS)

Core-level PE was proven to be extremely useful tool for time-resolved studies of, e.g. chemical interactions at FLASH and LCLS (W. Wurth, L. Kipp, A. Nilsson).



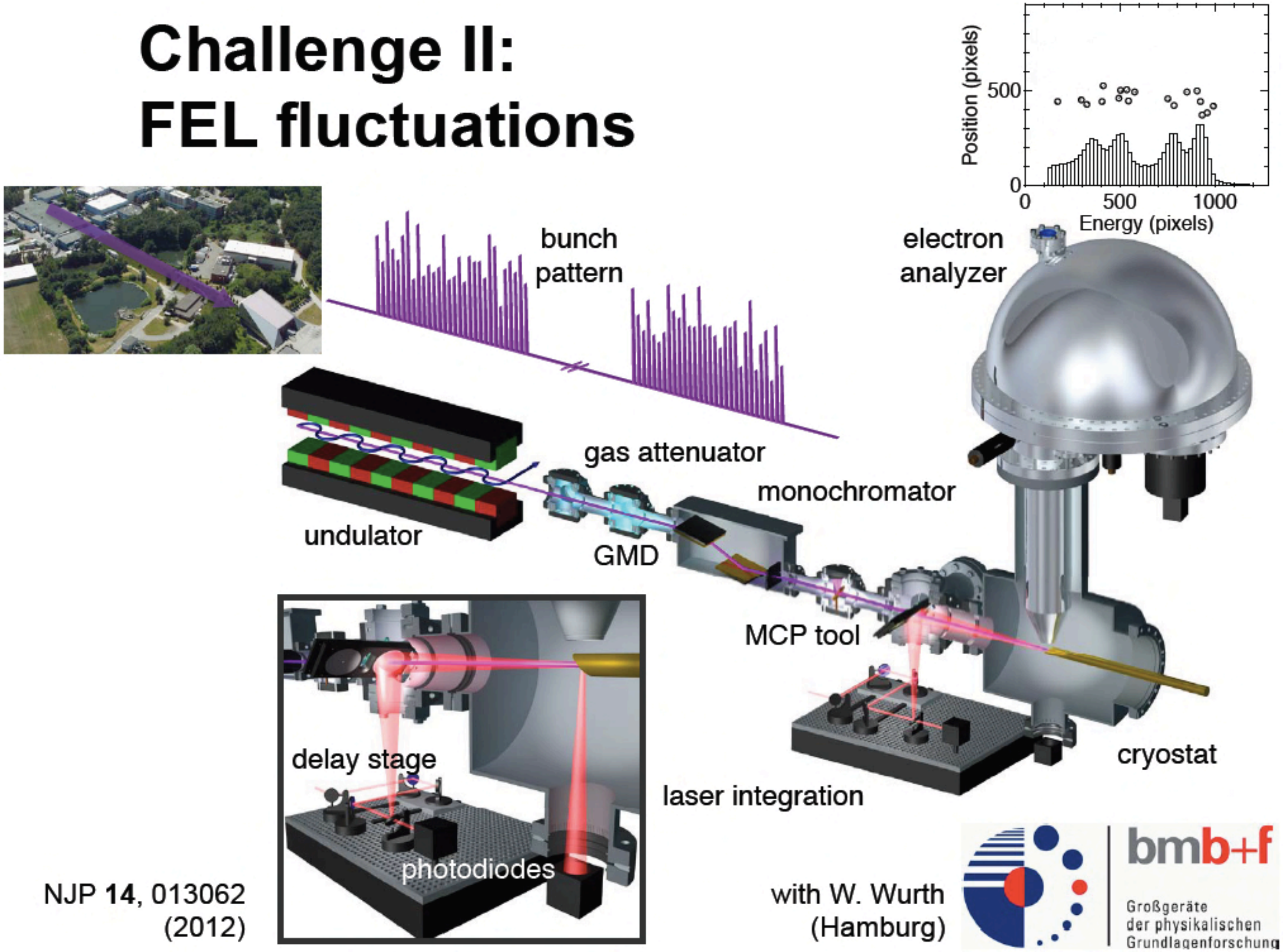
space charge (1mm spot)
$> 10^8$ phot/pulse
$> 10^4$ el/pulse





Problems to be solved

Challenge II: FEL fluctuations



NJP **14**, 013062 (2012)

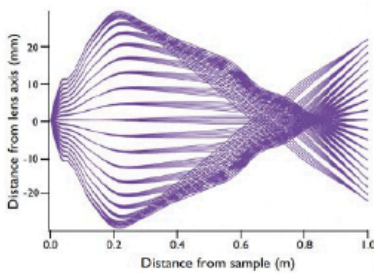
with W. Wurth (Hamburg)

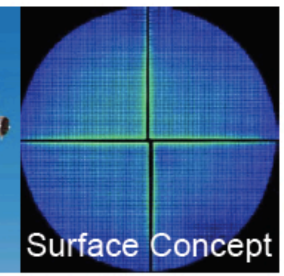
bmb+f
Großgeräte der physikalischen Grundlagenforschung

Problems to be solved

Challenge III:
Low FEL repetition rates

Angle imaging + TOF spectroscopy + multi-hit detection





Space-charge limit: $I_0 \approx 10^4 \, \frac{\mathrm{e}^-}{\mathrm{pulse}}$

Energy window: $\frac{\Delta E}{E} \approx 0.01$

Angular acceptance: $\frac{\Delta \Omega}{2\pi} \approx 0.034$

Electron counts per photon pulse:

$$\boxed{I_0 \times \frac{\Delta E}{E} \times \frac{\Delta \Omega}{2\pi} \approx 3.4 \, \frac{\mathrm{e}^-}{\mathrm{pulse}}}$$

1 *T*-TaSe2: trXPS using FLASH

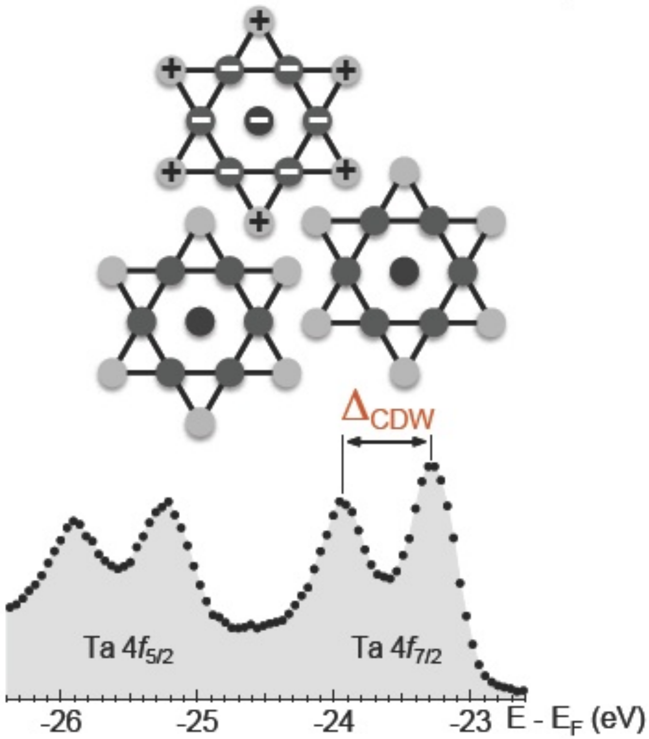
PRL **105**, 187401 (2010)

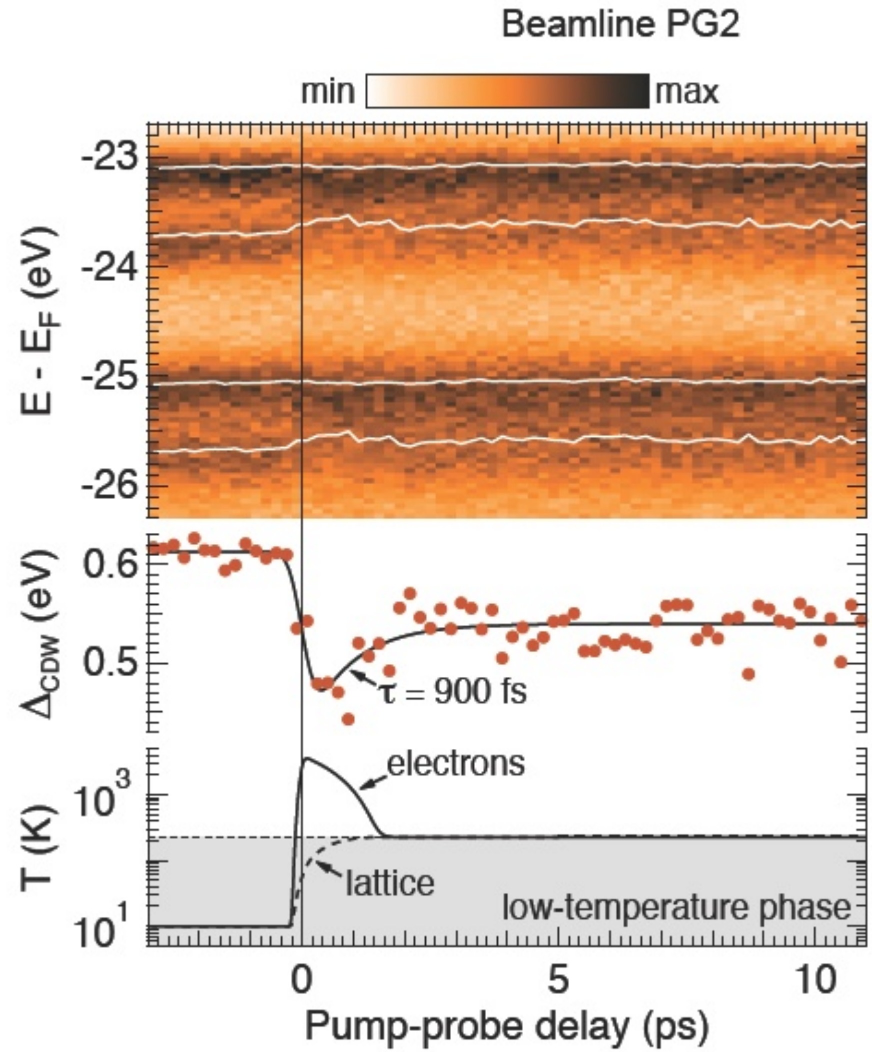
$T = 10$ K

$h\nu_{pump} = 1.55$ eV. $h\nu_{probe} = 156$ eV

$\Delta E \approx 300$ meV. $\Delta t \approx 700$ fs

$F = 1.8$ mJ/cm^2





European XFEL

Angle-resolved photoemission (ARPES)

In contrast, angle(spin)-resolved photoemission (ARPES) that for crystalline species is the only tool providing direct information on

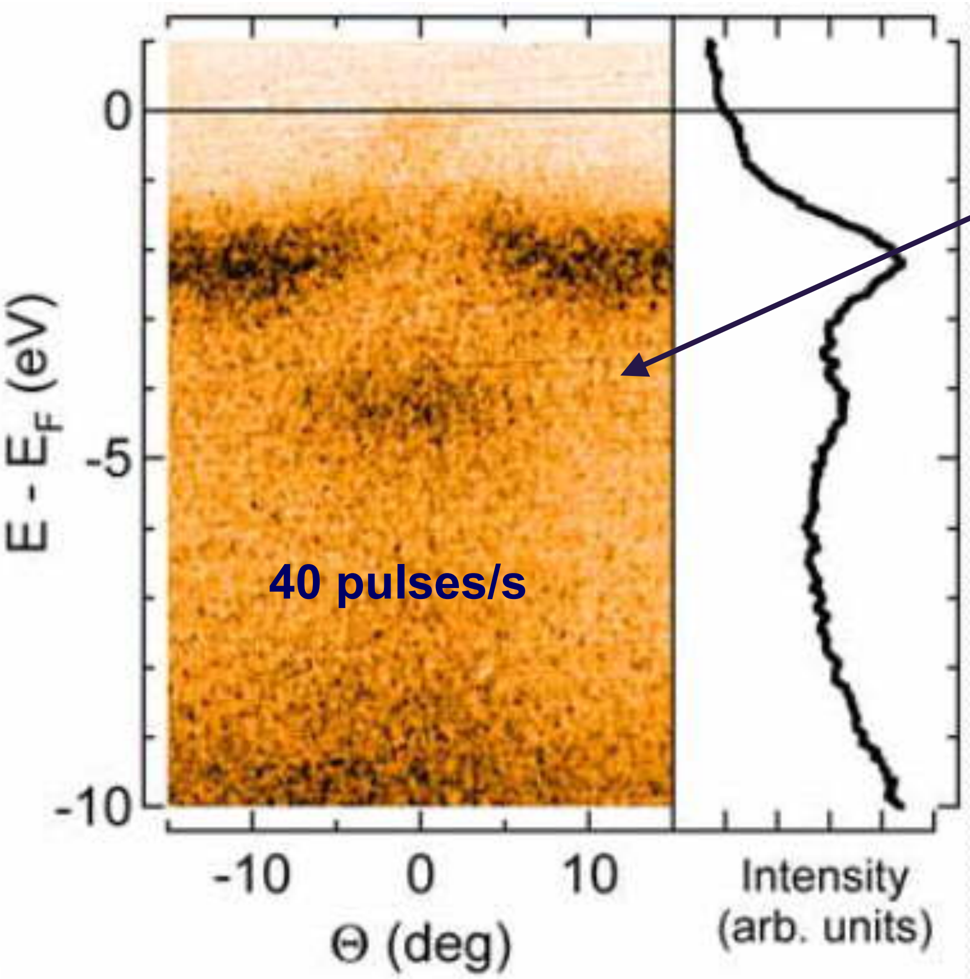
- single-particle excitations (simple *s*- and *p*-like systems) and
- electron interactions (correlated *d*- and *f*-systems)

is not straightforward at the existing low repetition rate FELs!

Reason: too less "allowed" excited electrons per second to acquire reasonable statistics

*Note: ARPES signal is 100-1000 lower than XPS one

ARPES at XFELs with low repetition rate



What you get at non-superconducting XFEL facilities (60 - 100 Hz rep. rate)

European XFEL

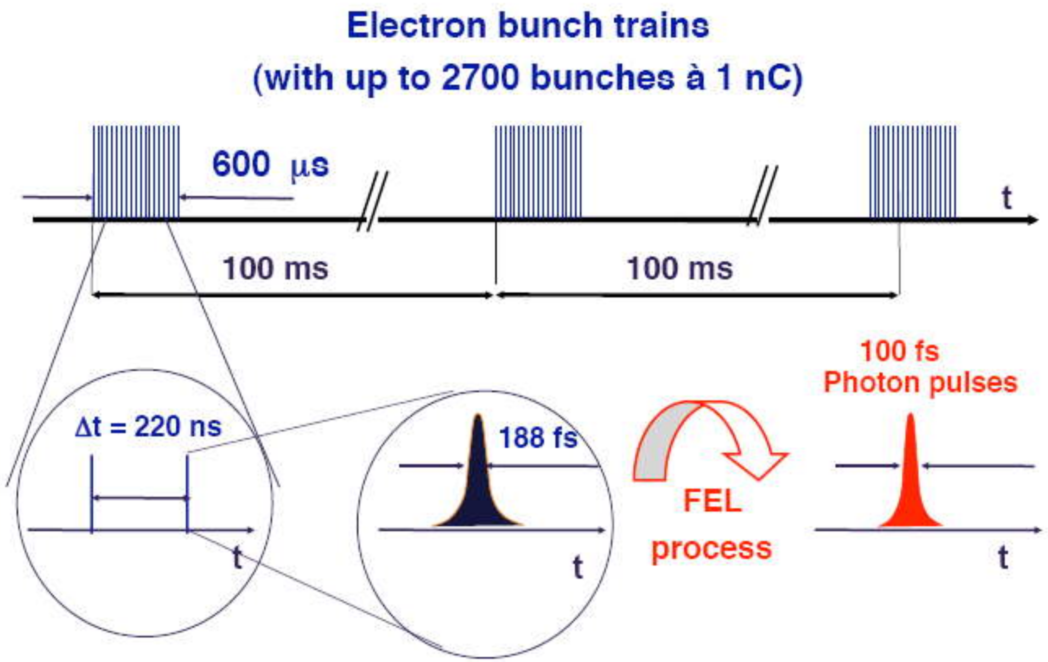
Why angle-resolved photoemission?

On the other hand, particularly temperature dependent electron dynamics that causes transitions between

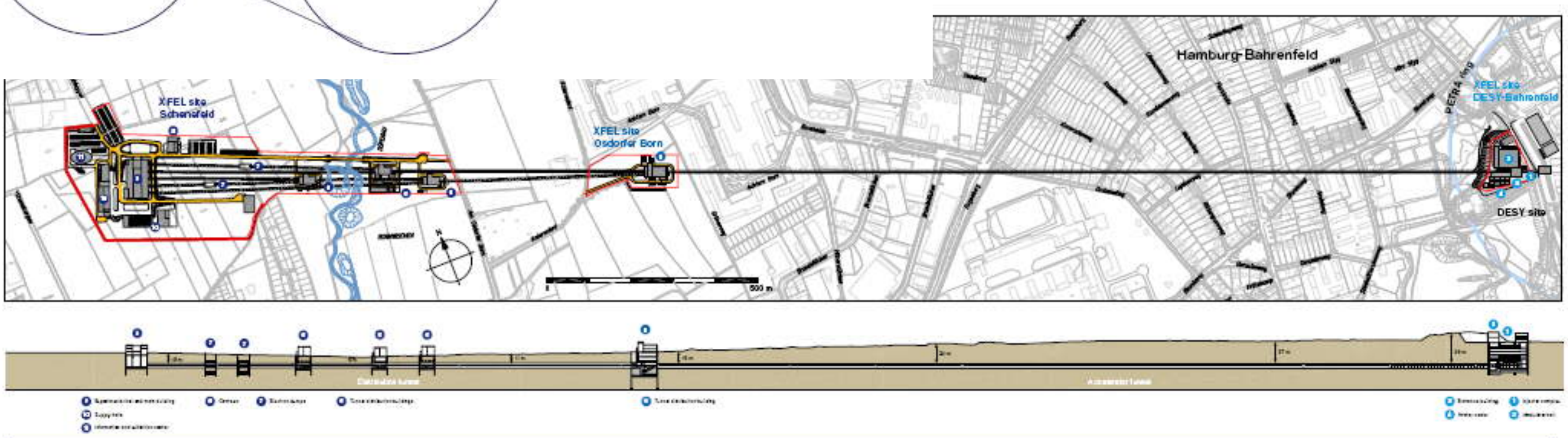
superconcting, magnetic and
Kondo (heavy fermion) properties

in correlated systems is dreamed to be studied, since it allows
- understanding of underlying mechanisms of the phenomena
- switching from one behavior to another
- engineering of novel materials with well-defined properties

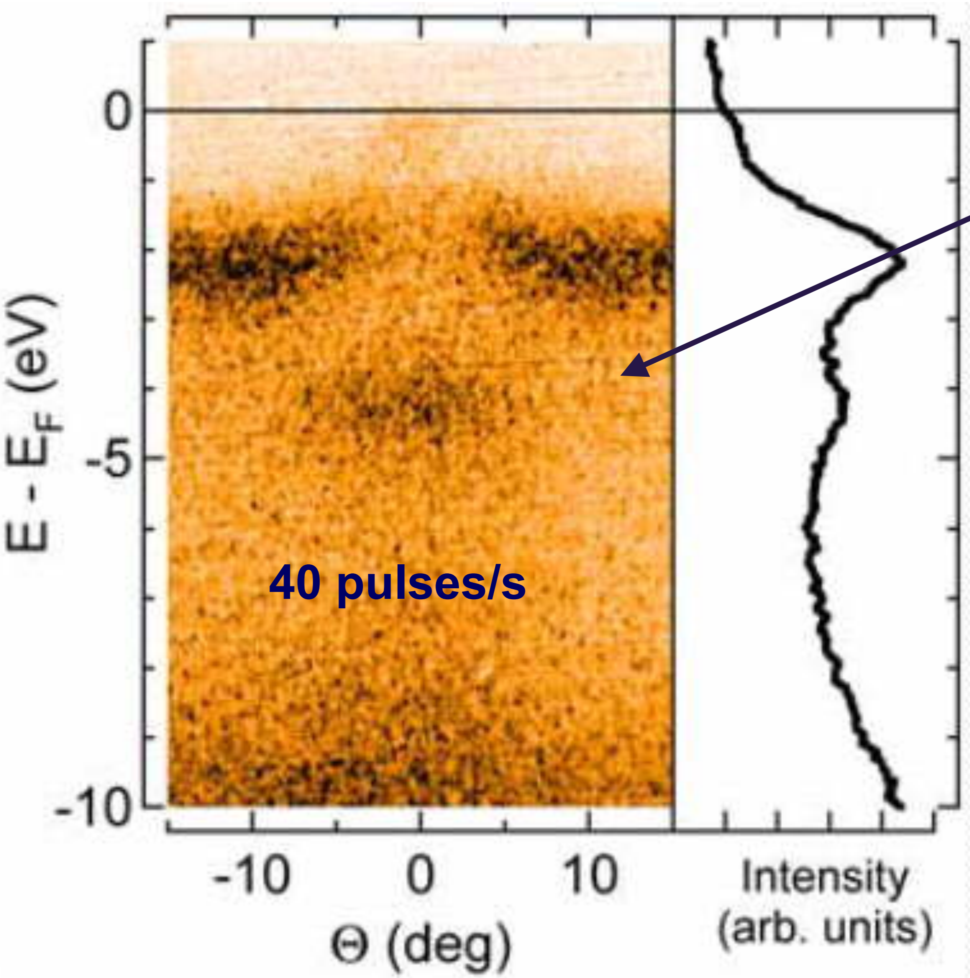
European XFEL: Time structure



Superconducting LINAC technology provides 27.000 light pulses/s in burst-like structure. It makes XFEL.EU attractive for photon-hungry experiments.



ARPES at European XFEL



What you get at non-superconducting XFEL facilities (60 - 100 Hz rep. rate)

Due to unique rep. rate photoemission response at the European XFEL is about 10^3 higher (statistics) → strong case for ARPES:

- **two-color exp. (unfilled states)**
- **pump-probe (electron dynamics)**

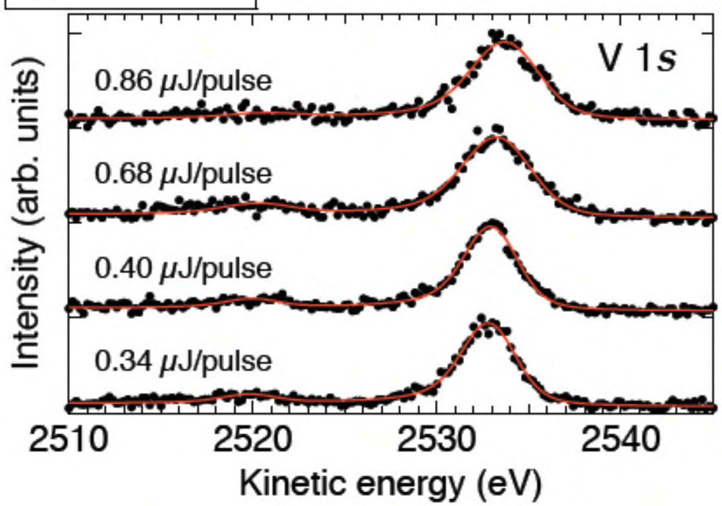
V 1s in VO2, SACLA (Japan)

NJP 16, 123045 (2014)

$h\nu = 7937\,\mathrm{eV}$

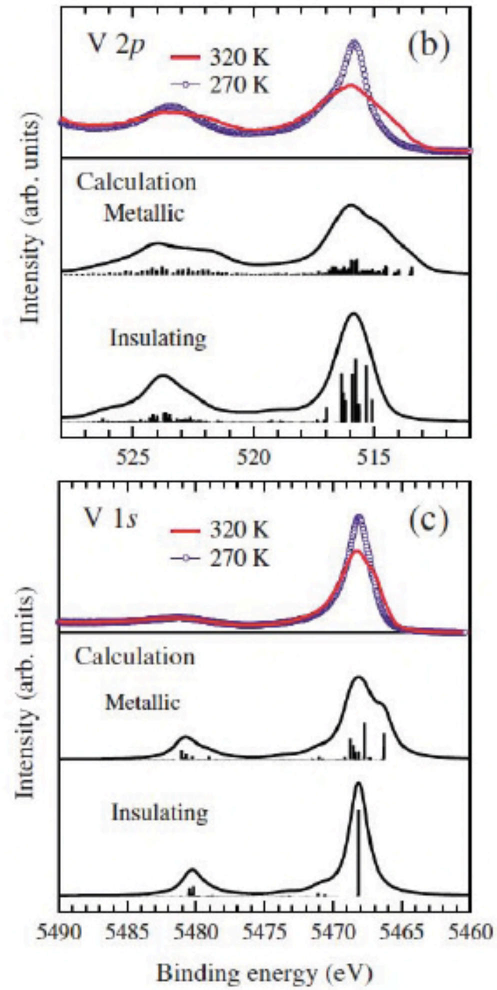
$\Delta E \approx 250\,\mathrm{meV}$

$h\nu = 8\,\mathrm{keV}$. $\Delta E \approx 1\,\mathrm{eV}$



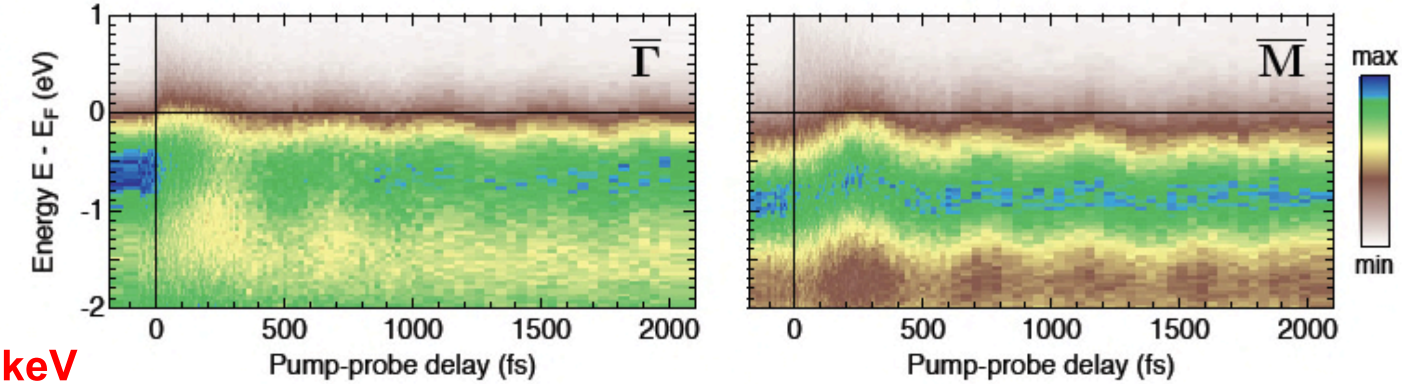
20 min at 20 Hz

Eguchi *et al.*, PRB **78**, 075115 (2008)



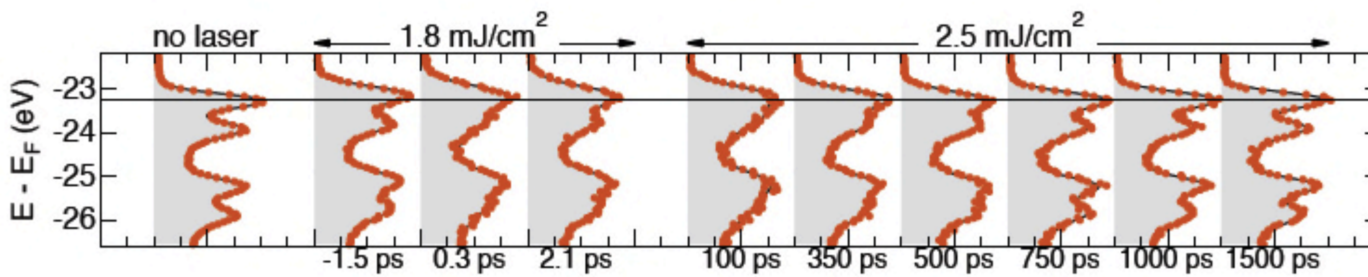
Combined probing of electronic & lattice order (Kai Rossnagel)

Spectral gaps
trARPES

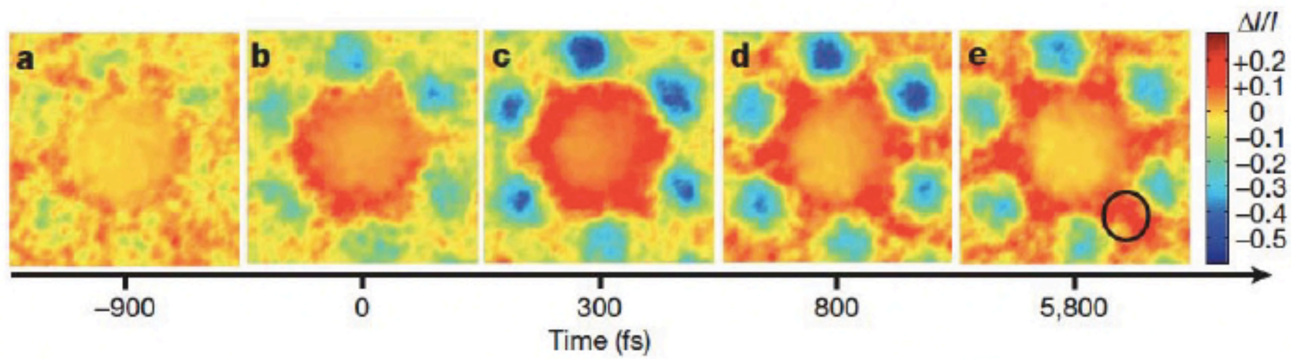


$h\nu_{probe}$ **≈ 0.1–2(> 5) keV**

Charge density wave
trXPS



Periodic lattice distortion
UED
trXPD



Participants of the TR-XPES User Consortium (K. Rossnagel)

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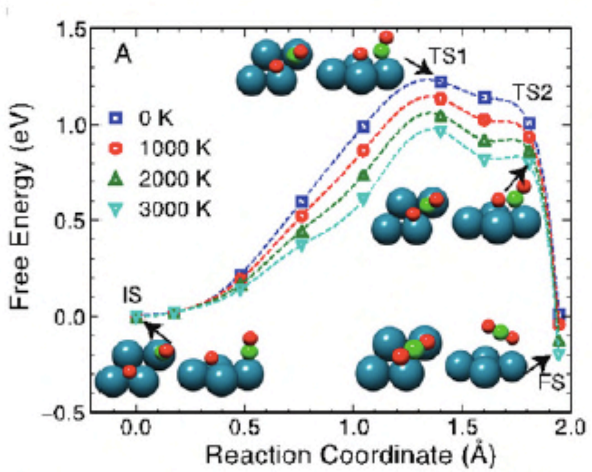
Alexander Föhlisch	HZB	Gerd Ganteför	Universität Kons
Mats Göthelid	KTH	Nils Martensson	Uppsala Universi
Anders Nilsson	Stockholm Univ./	Henrik Öström	Stockholm Unive
Hirohito Ogasawara	SLAC	Giancarlo Pannacione	IOM-CNR
Eric Pellegrin	ALBA	Giorgio Rossi	UMilano/IOM-CN
Alexander Soldatov	So. Fed. Univ. Ros	Gerd Schönhense	Universität Mainz
Giovanni Stefani	Universita Roma	Svante Svensson	Uppsala Universi
Oscar Tjernberg	KTH	Geoff Thornton	UCL
Martin Weinelt	FU Berlin	Jonas Weissenberger	KTH
Martin Wolf	FHI Berlin		
XFEL contacts			
Serguei Molodtsov	Harald Sinn	Andreas Scherz	

Science with TR-XPES

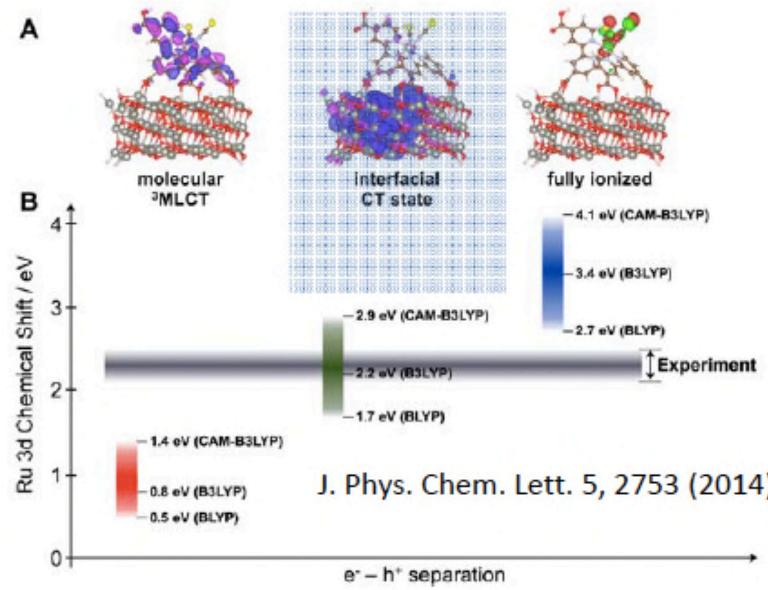
Science case for TR-XPES at EuXFEL

Surface and Interface Chemistry and Catalysis – observe transition states

Photovoltaics – follow charge transfer at interfaces



H. Öström et al. Science 347,978 (2015)



J. Phys. Chem. Lett. 5, 2753 (2014)

Methods

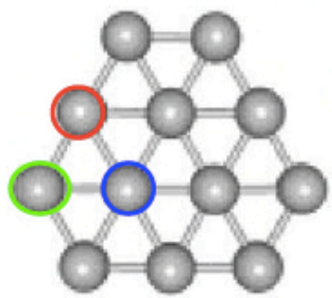
TR-XPS (ESCA) → Element specific, chemical state selective,
→ Local charge state

TR-XPED → element specific, structural information

Science with TR-XPES

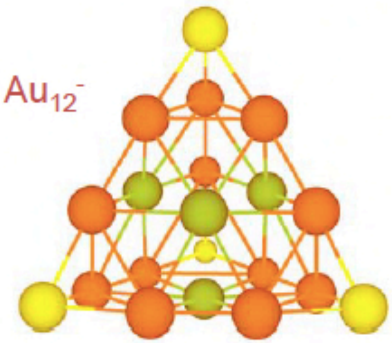
Science case for TR-XPES at EuXFEL

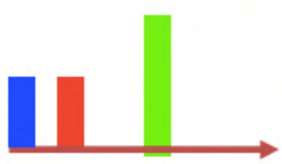
Cluster physics
– structure and dynamics (e.g. dissociation, non-thermal melting) as function of size



planar clusters up to at least Au_{12}^{-}

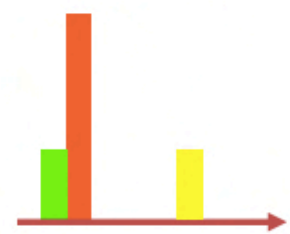
vs. tetrahedral Au_{20}^{-}





Methods

TR-XPS (ESCA)

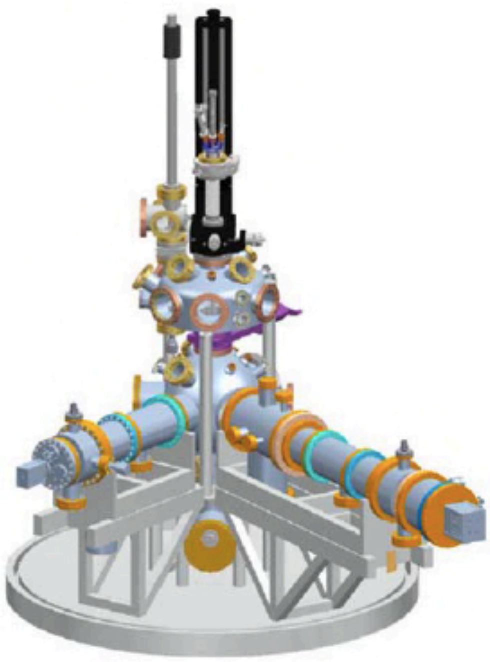


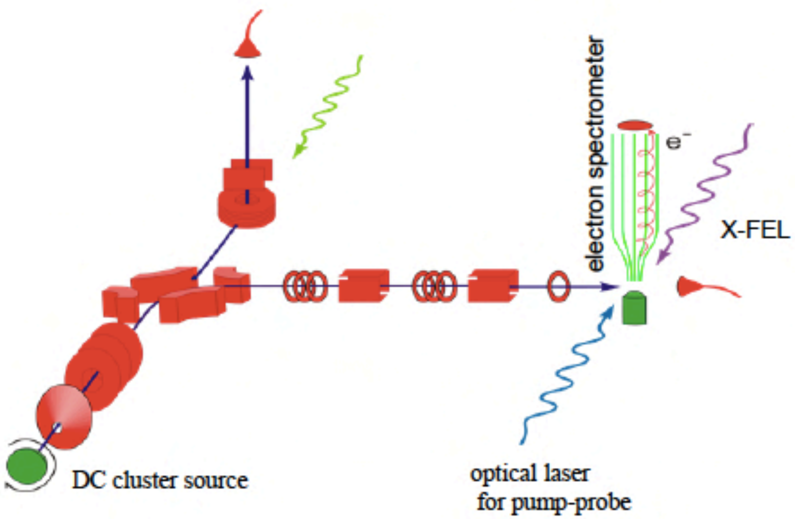
→ **Element specific, chemical state selective,**
→ **Local coordination**

TR-XPES: Endstations

Technical concept - Endstations

Stage 1 (2016/2017) – use existing endstations





TR-PES instrument (Wurth)

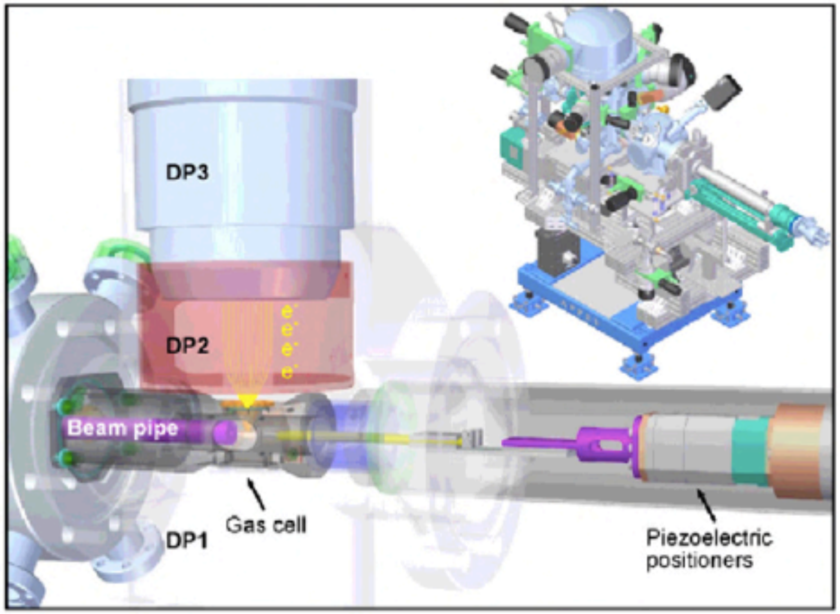
Cluster instrument (Eberhardt, Ganteför)

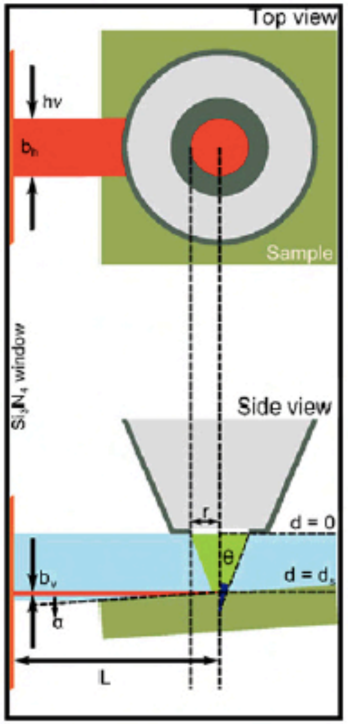
TR-XPES: Endstations

Technical concept - Endstations

Stage 2 – dedicated instruments under development

e.g. Nilsson group - Ambient Pressure XPS





S. Kaya, et al, *Catal. Today* **205**, 101 (2012).

You are very welcome

to plan your experiments

at the European XFEL