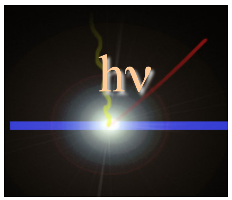
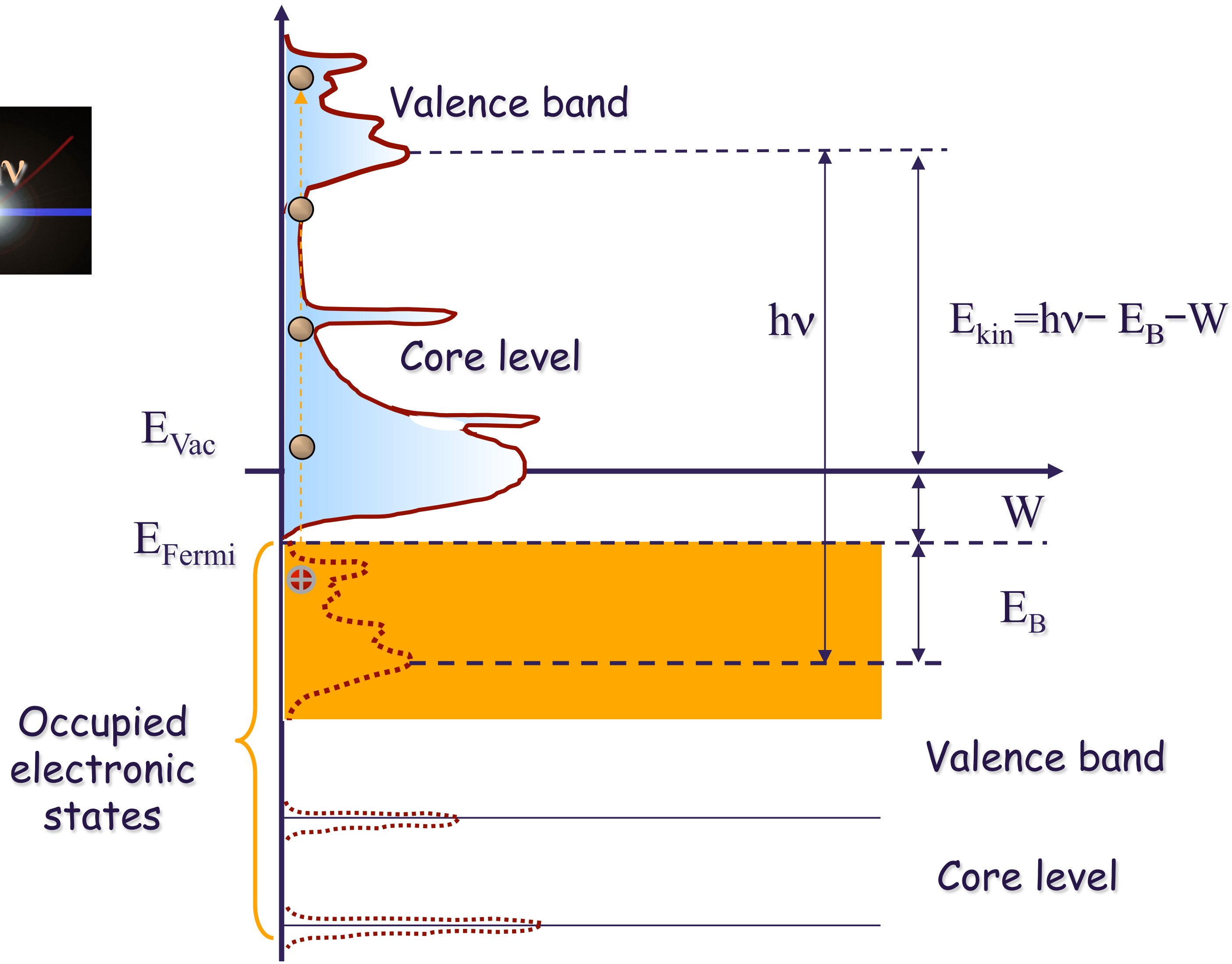
New Opportunities for PES of Condensed Matter Using FELs

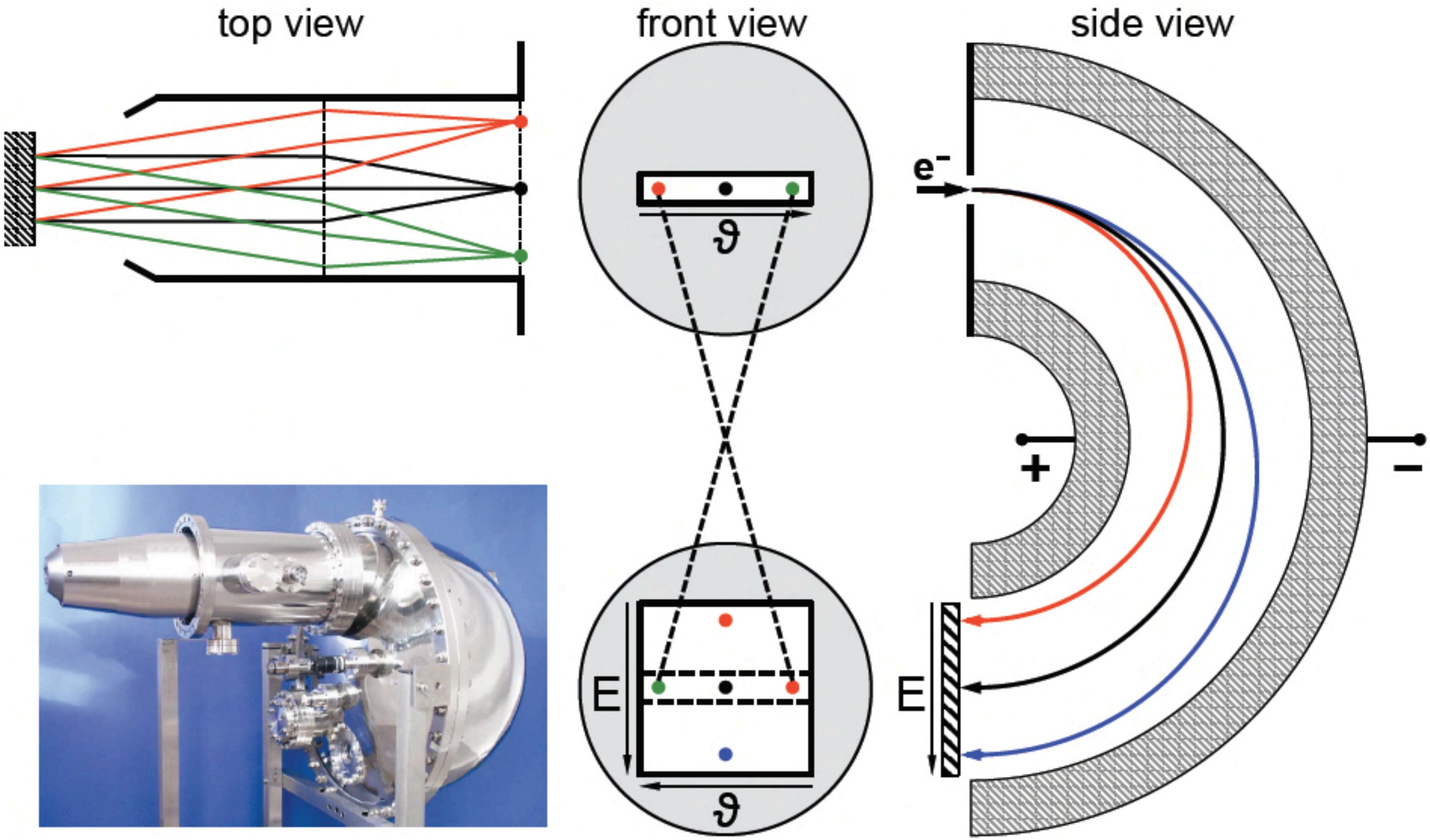
S.L. Molodtsov
European XFEL GmbH

Photoemission spectroscopy: Basics





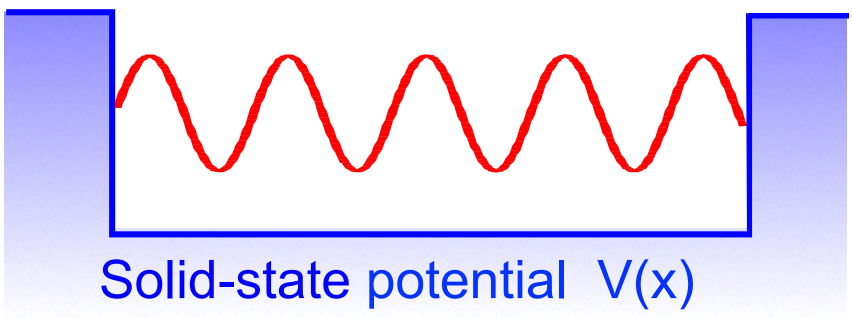
Photoelectron analyzer



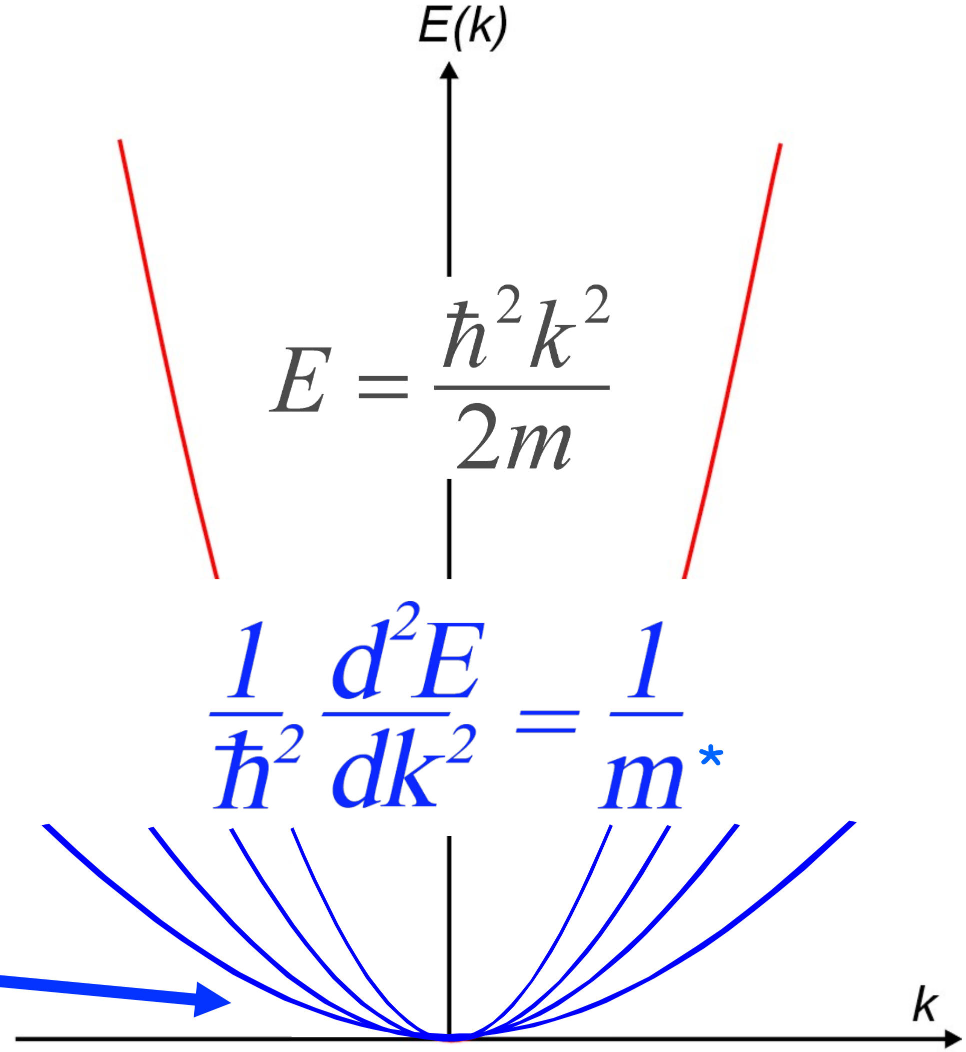
Single-particle system

freie Elektronen :

1 - dim. solid state:



Heavy-fermion system

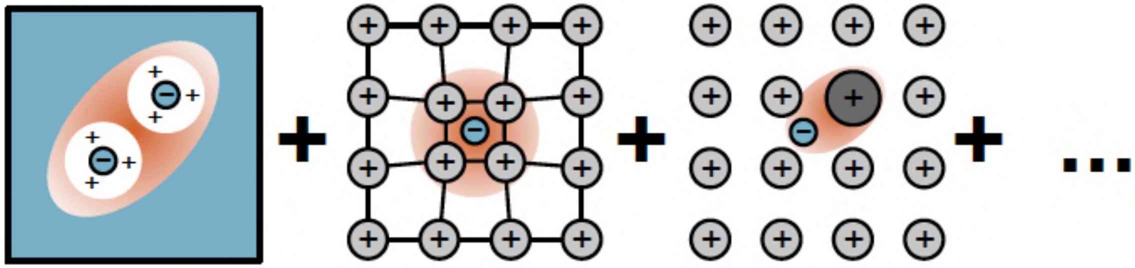


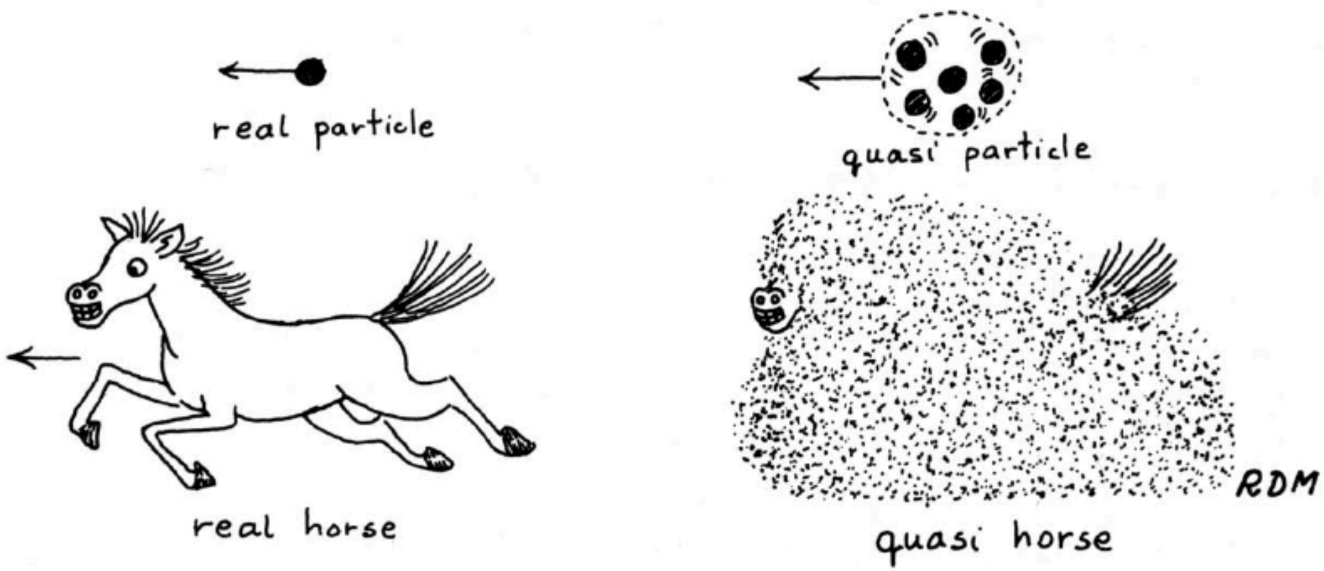
$$E = \frac{\hbar^2 k^2}{2m}$$

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

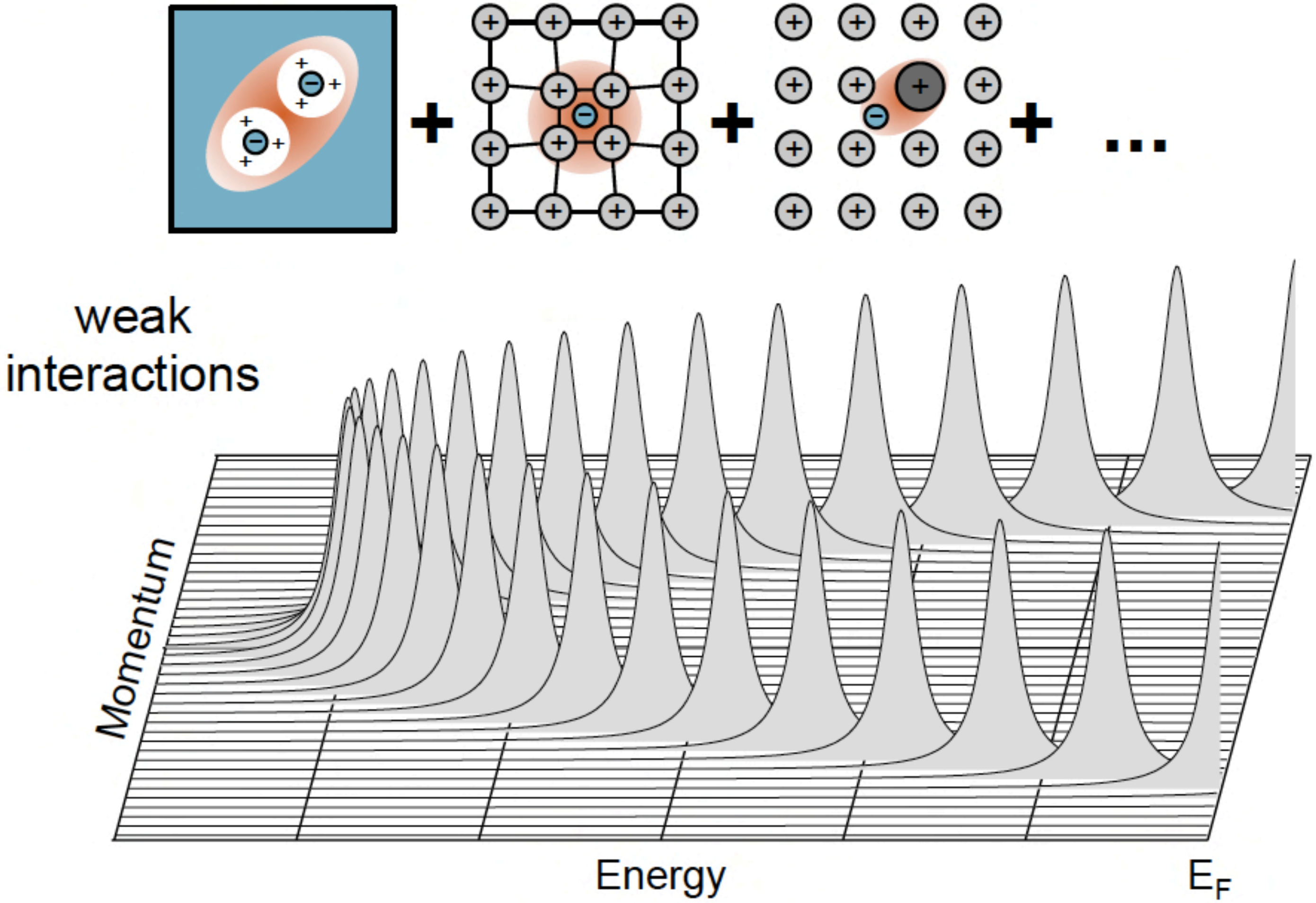
Quasiparticles properties (Kai Rossnagel)

Lead actor: The quasi-electron



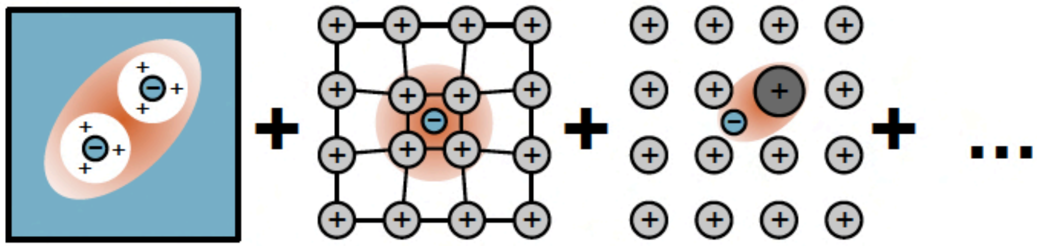


Lead actor: The quasi-electron

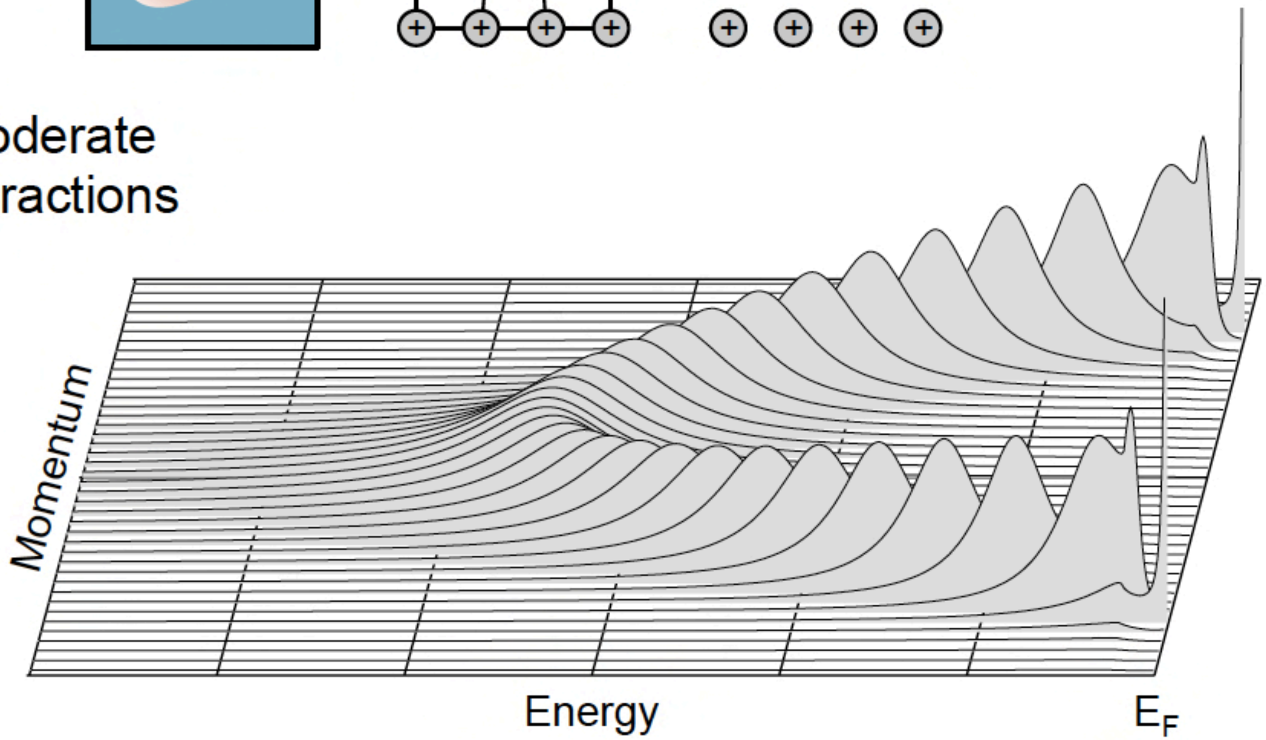


Moderate correlations

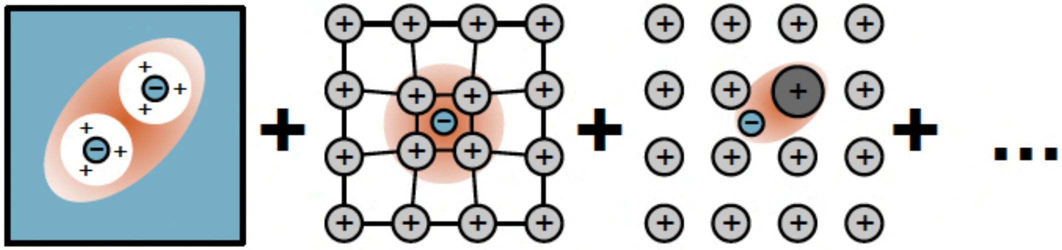
Lead actor: The quasi-electron

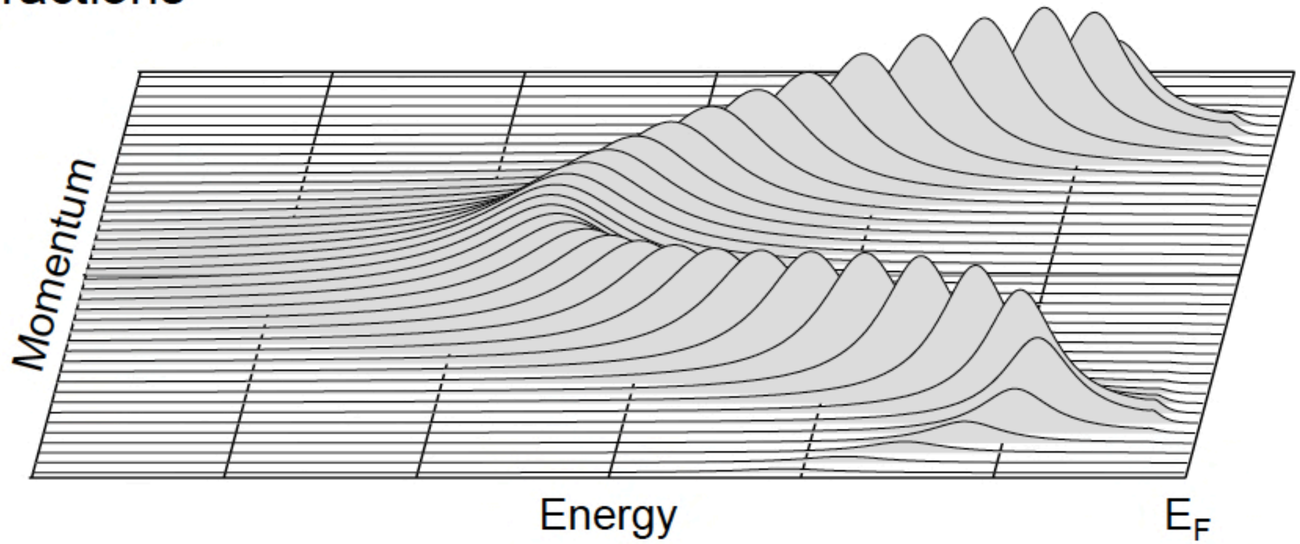


moderate interactions



Lead actor: The quasi-electron

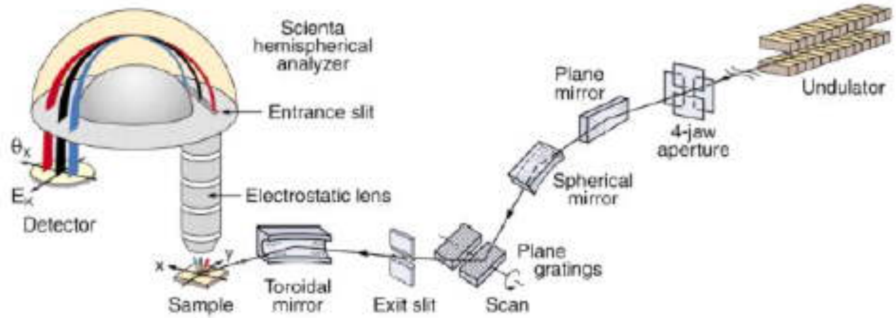




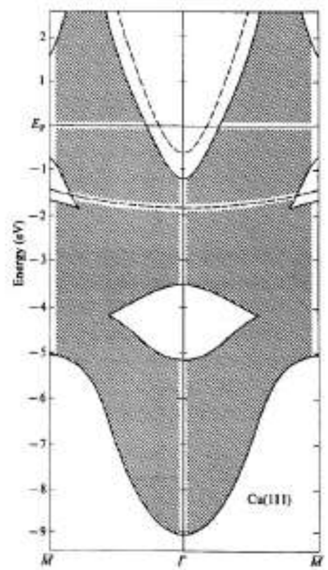
Nobel prize to X-ray spectroscopy work

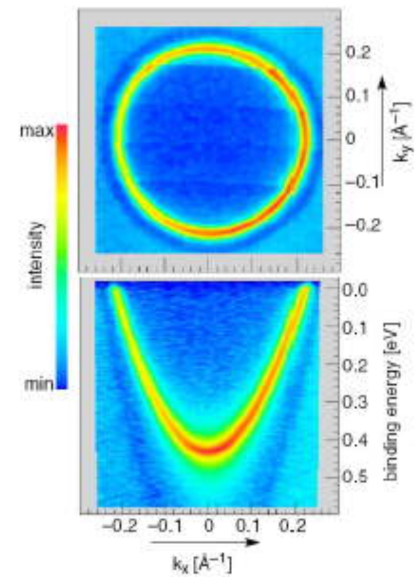
Angle-resolved photoemission - ARPES

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)

European XFEL

Non-correlated materials

solid state

↓

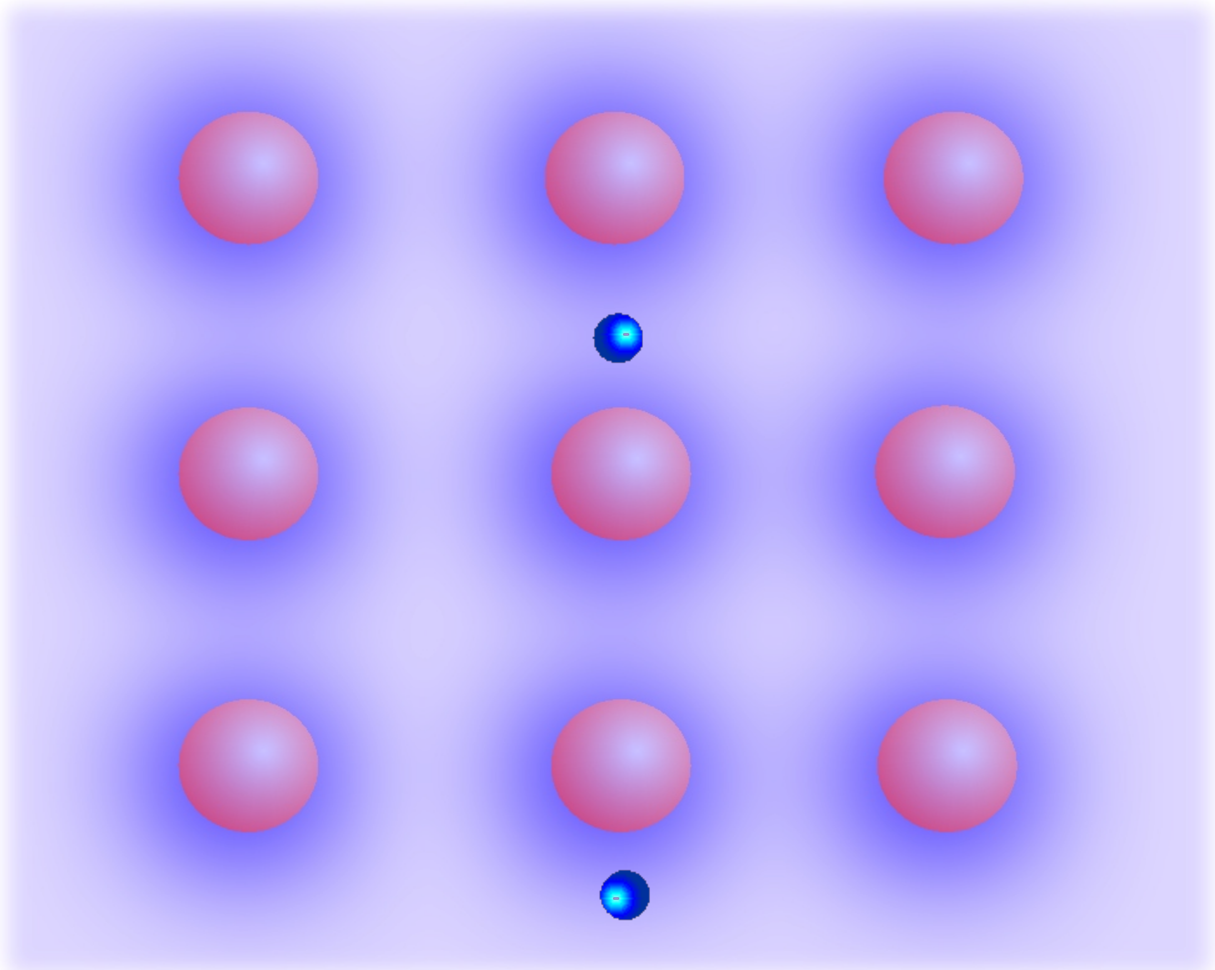
potential averaged
over all electrons

↓

single-particle
calculations,
LDA approach

Problem: ↓

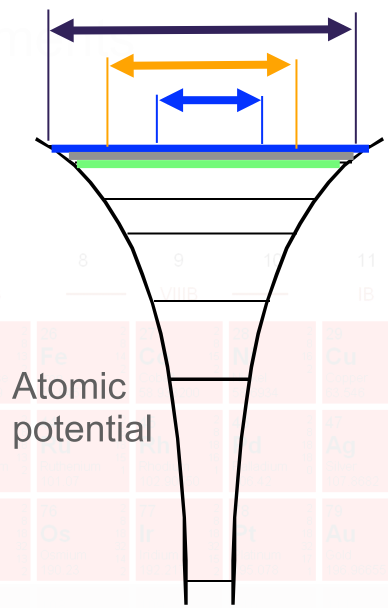
negligence
of electron correlations

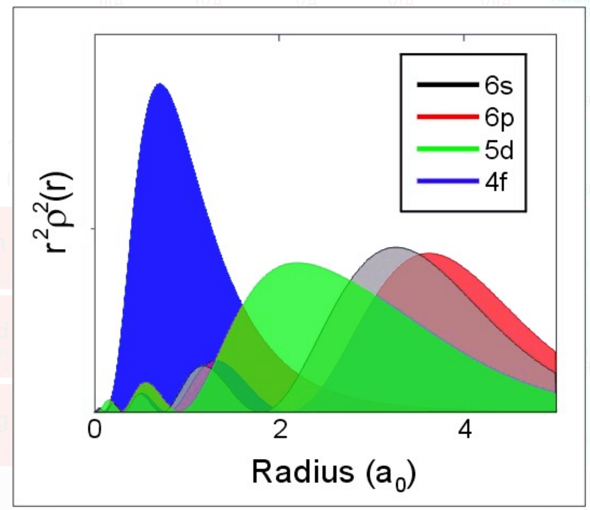


Correlated *f*-materials

Electron configuration:

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





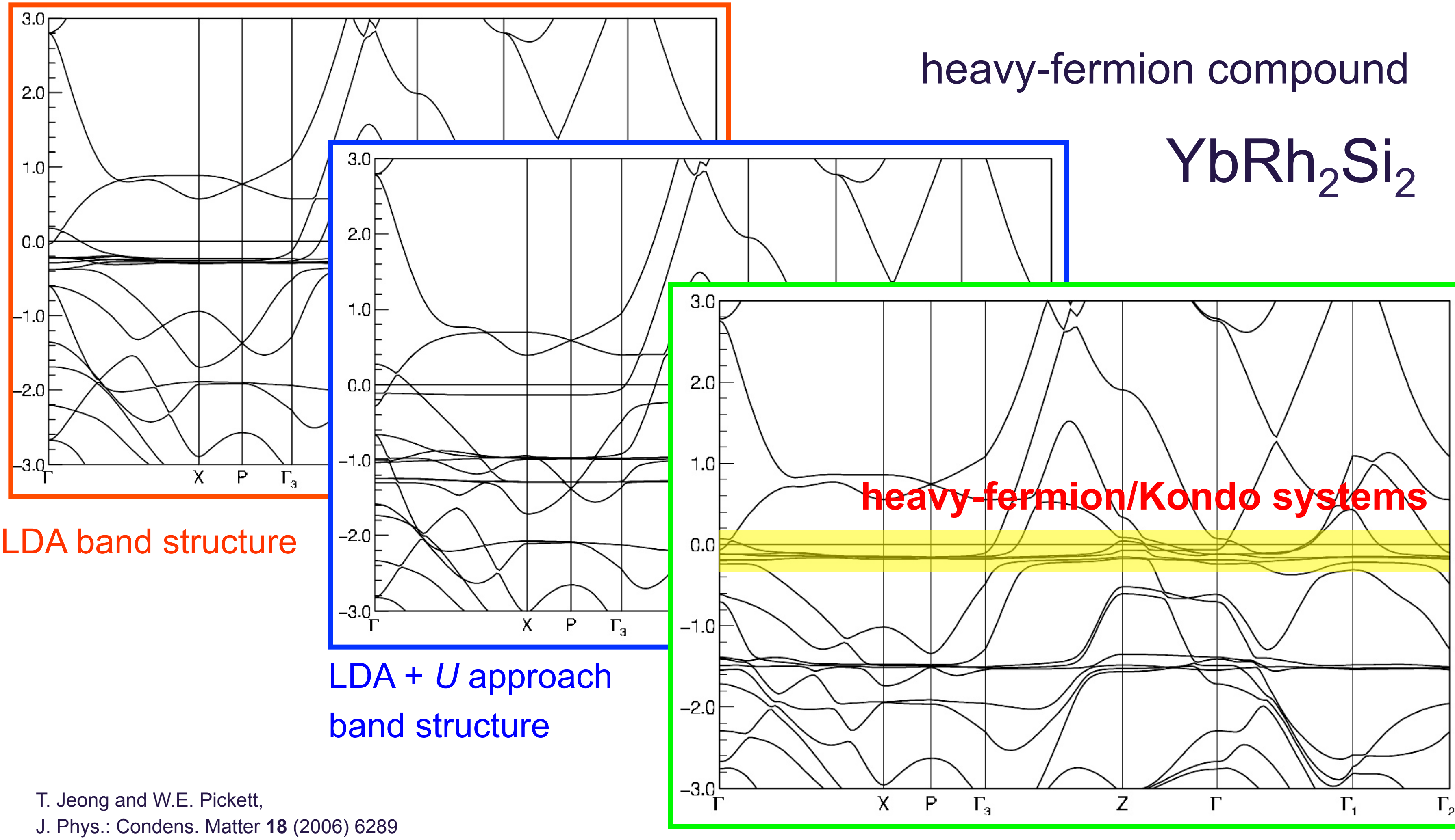
Lanthanoide

f - electrons

Correlated *f*-materials

heavy-fermion compound

$YbRh_2Si_2$



LDA band structure

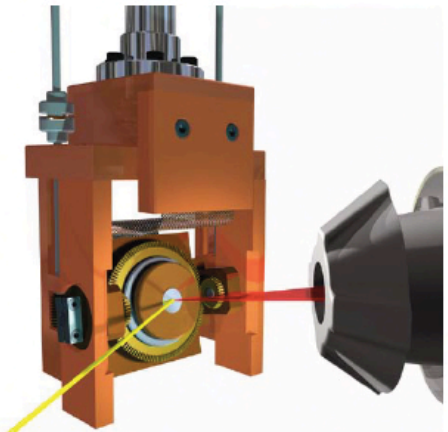
LDA + *U* approach band structure

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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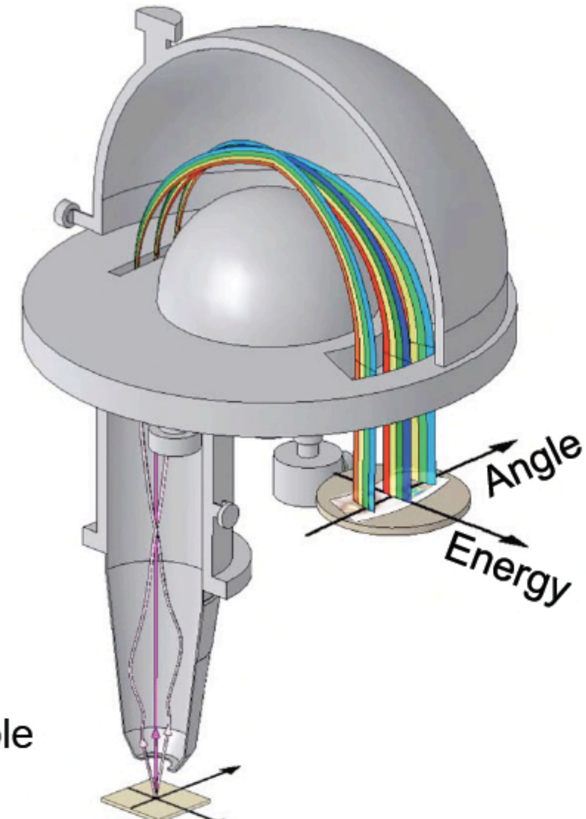

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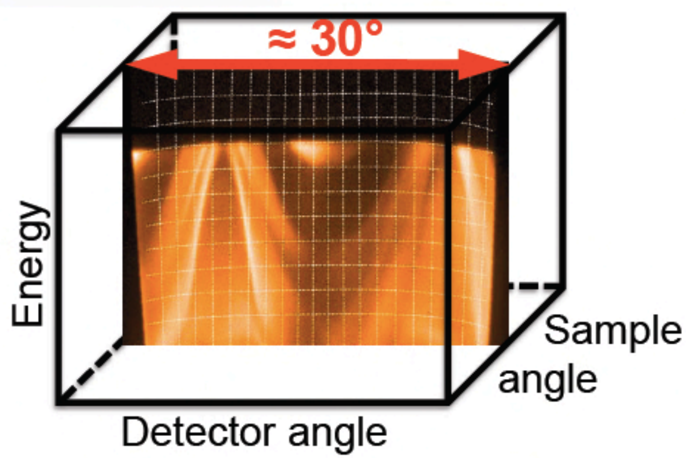
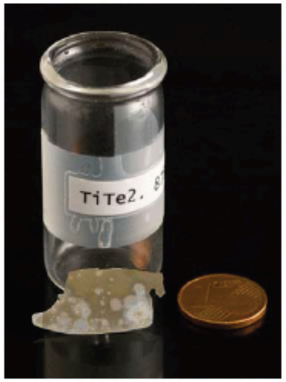
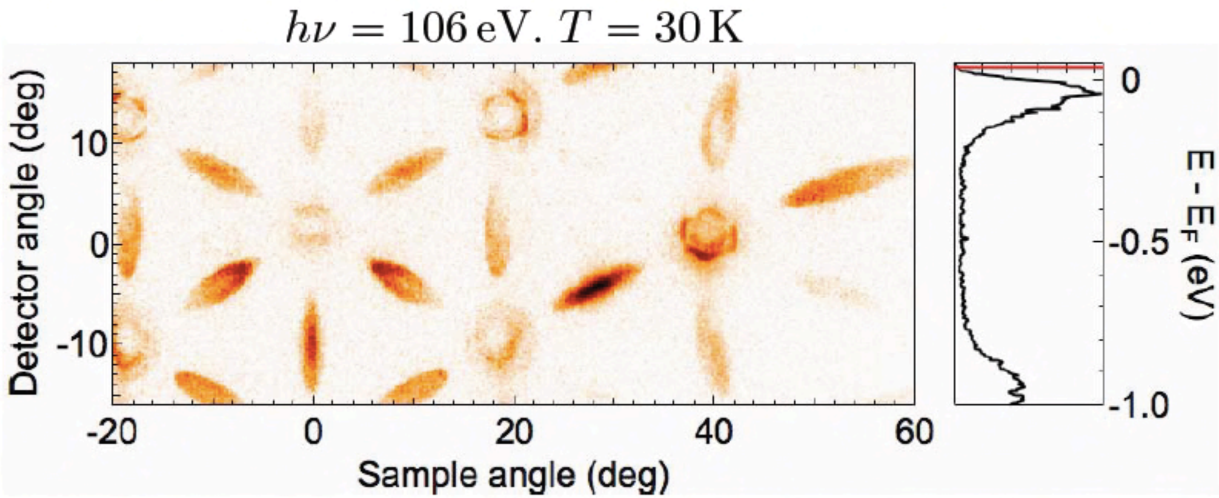


Band mapping (seeing is believing)

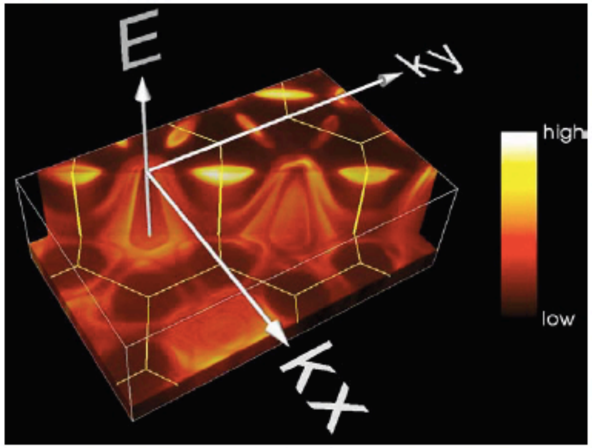




K. Rossnagel, et al. Uni Kiel

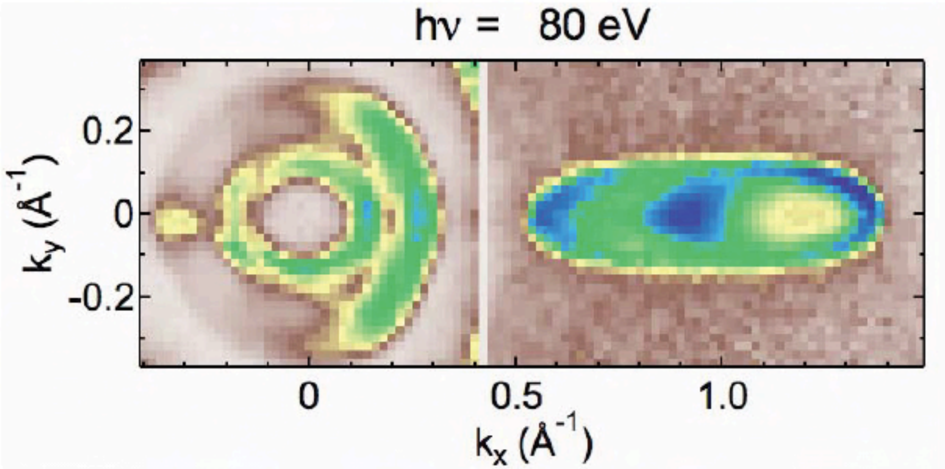
$$E - E_F = E_{\text{kin}} + W - h\nu$$

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

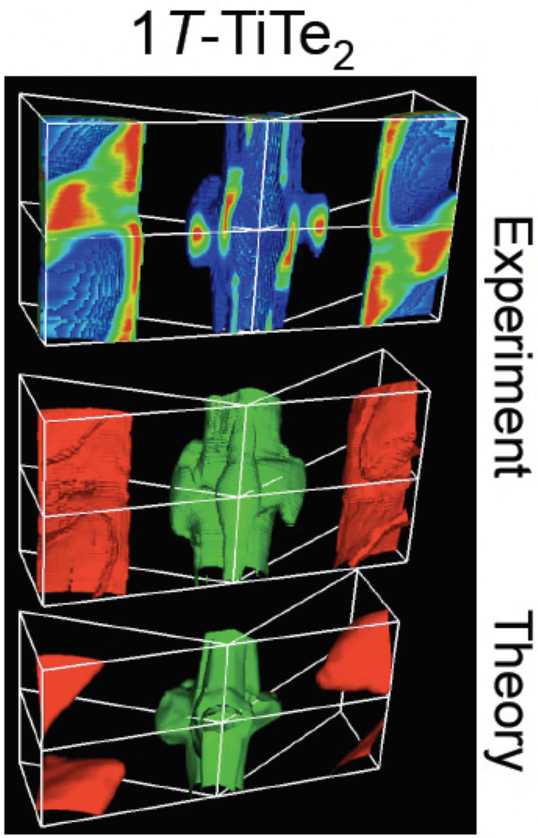


Beamline 7, ALS, Berkeley

Fermi surface tomography



K. Rossnagel, et al. Uni Kiel



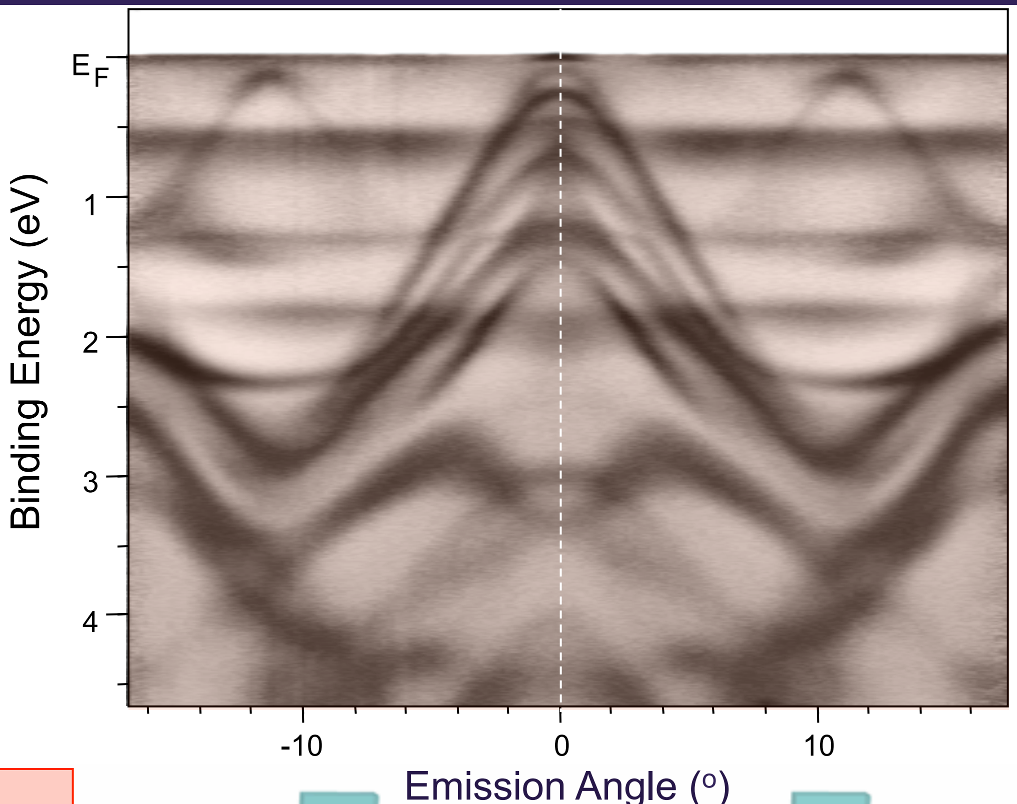
$$\mathbf{k}_{\|}^2 + k_{\perp}^2 = \frac{2m}{\hbar^2}\left(E_{\mathrm{kin},F} + V_0\right)$$

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

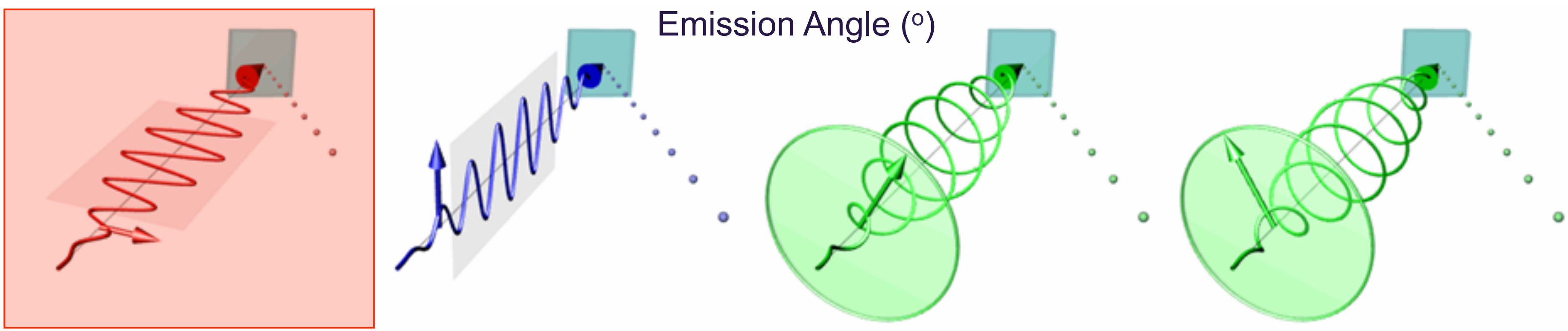
$$E_{\mathrm{kin},F} = h\nu - W$$

Beamline 7, ALS, Berkeley

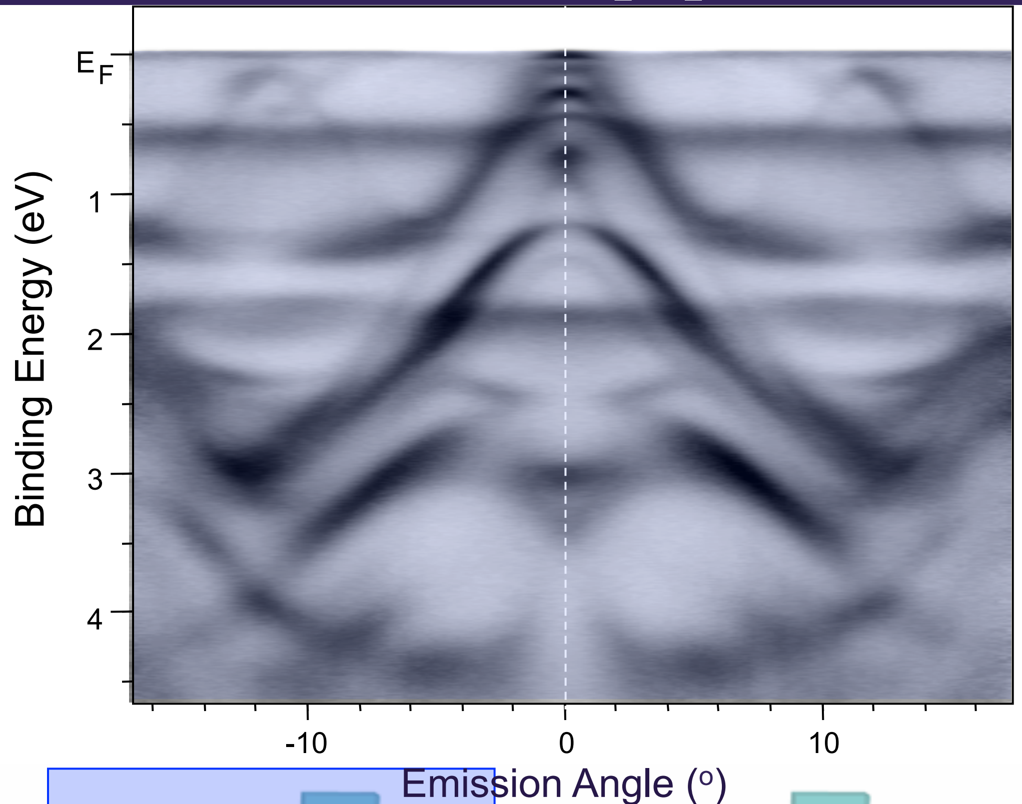
Light-polarized ARPES on heavy-fermion $YbRh_2Si_2$ (S. Molodtsov, et al.)



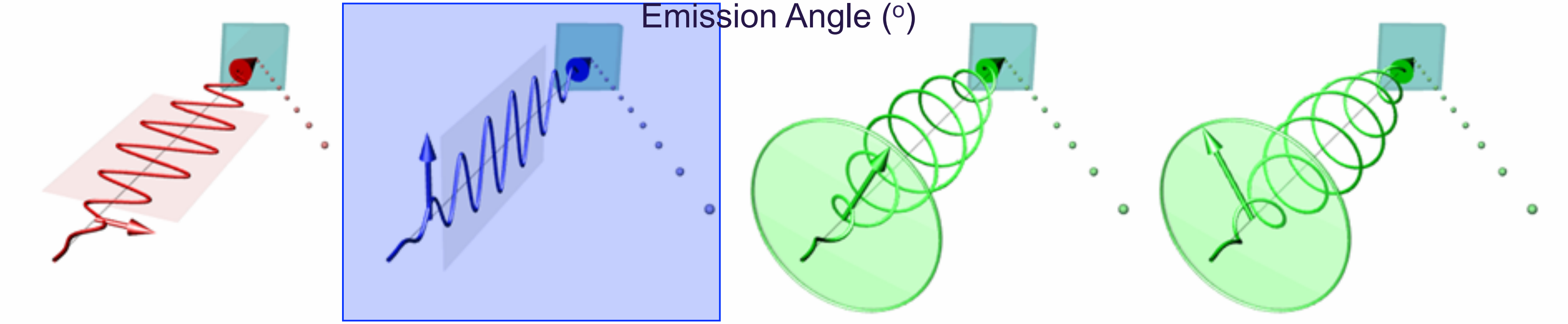
$YbIr_2Si_2$



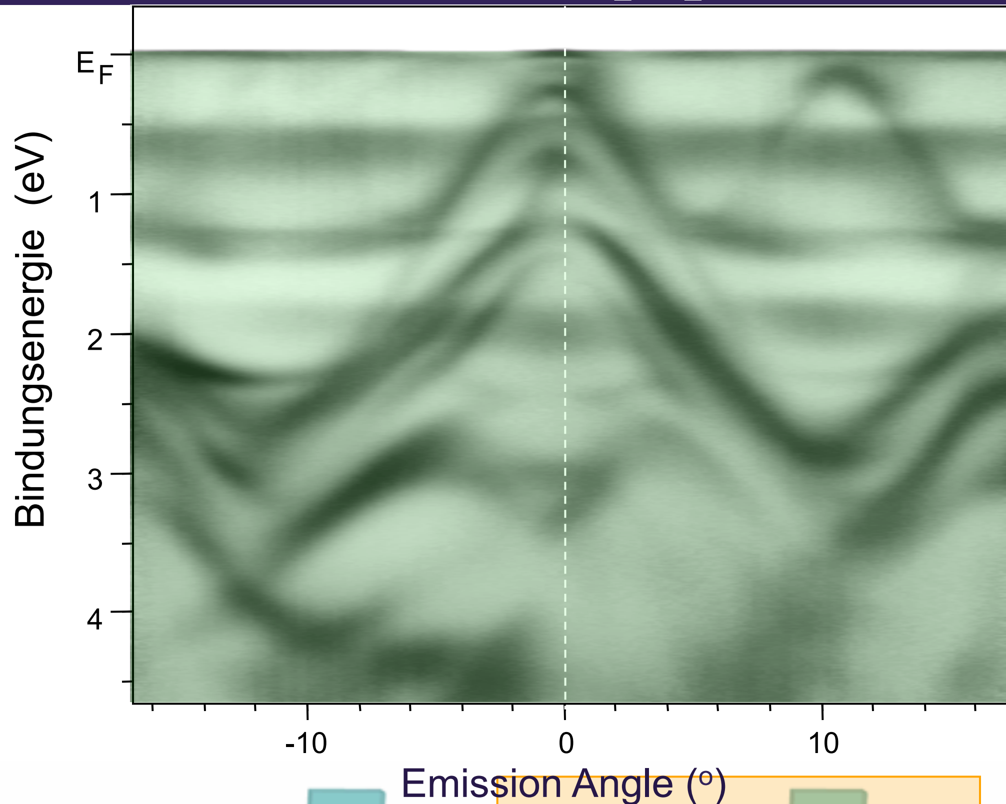
Light-polarized ARPES ($YbRh_2Si_2$)



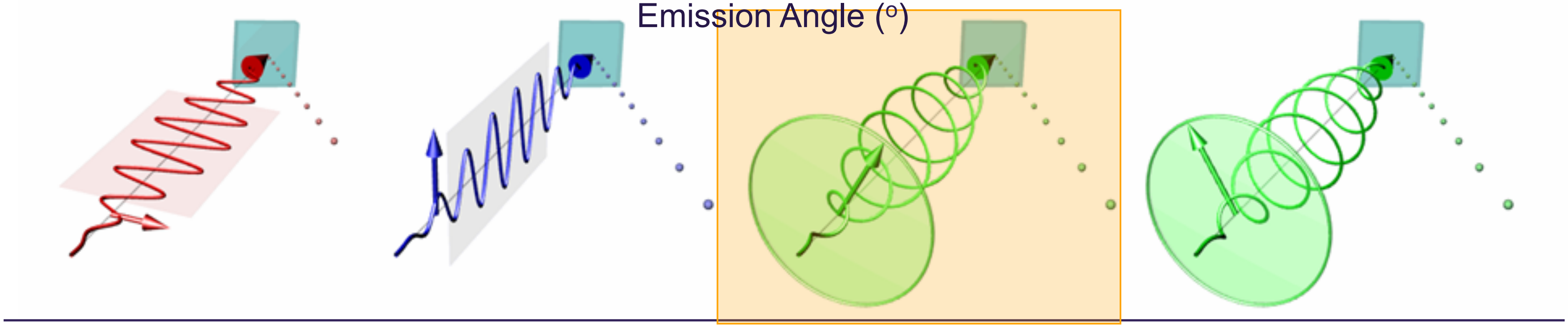
$YbIr_2Si_2$



Light-polarized ARPES ($YbRh_2Si_2$)



$YbIr_2Si_2$



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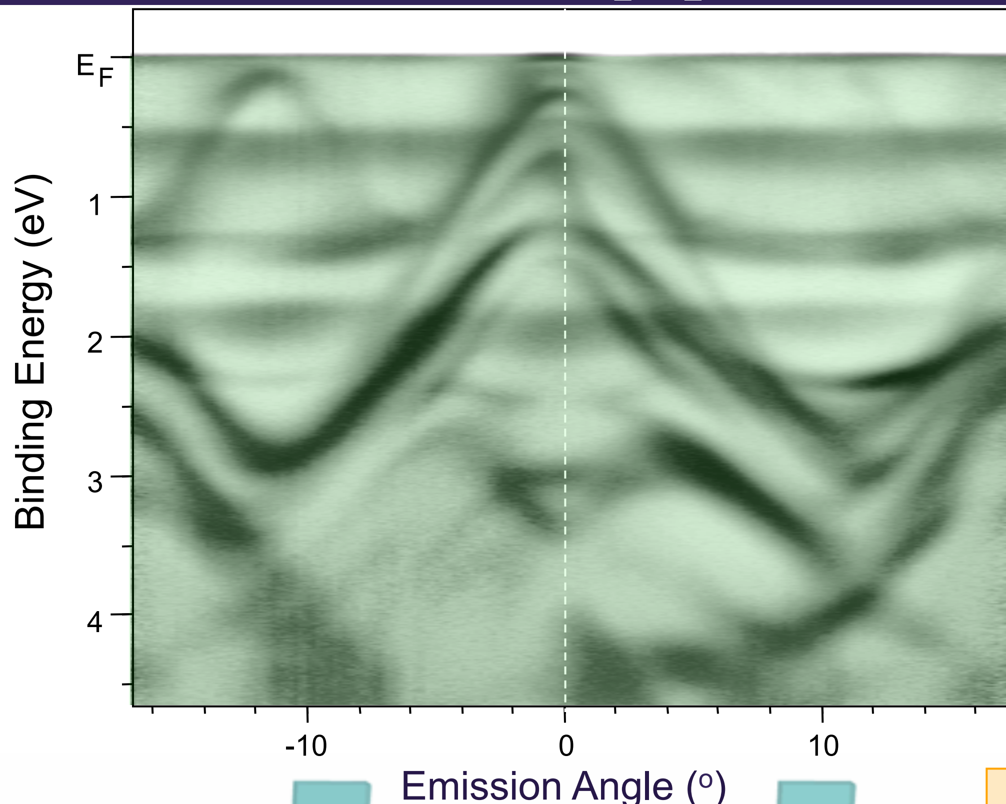
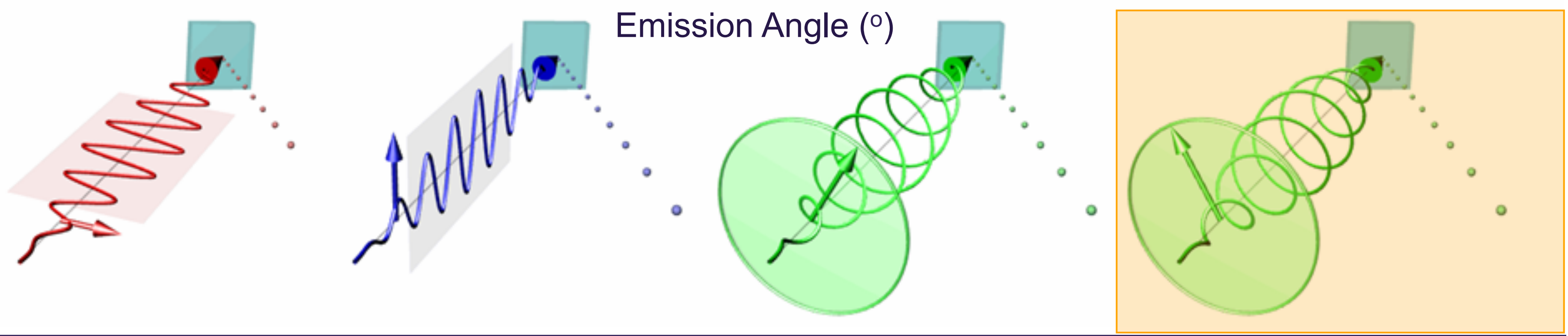
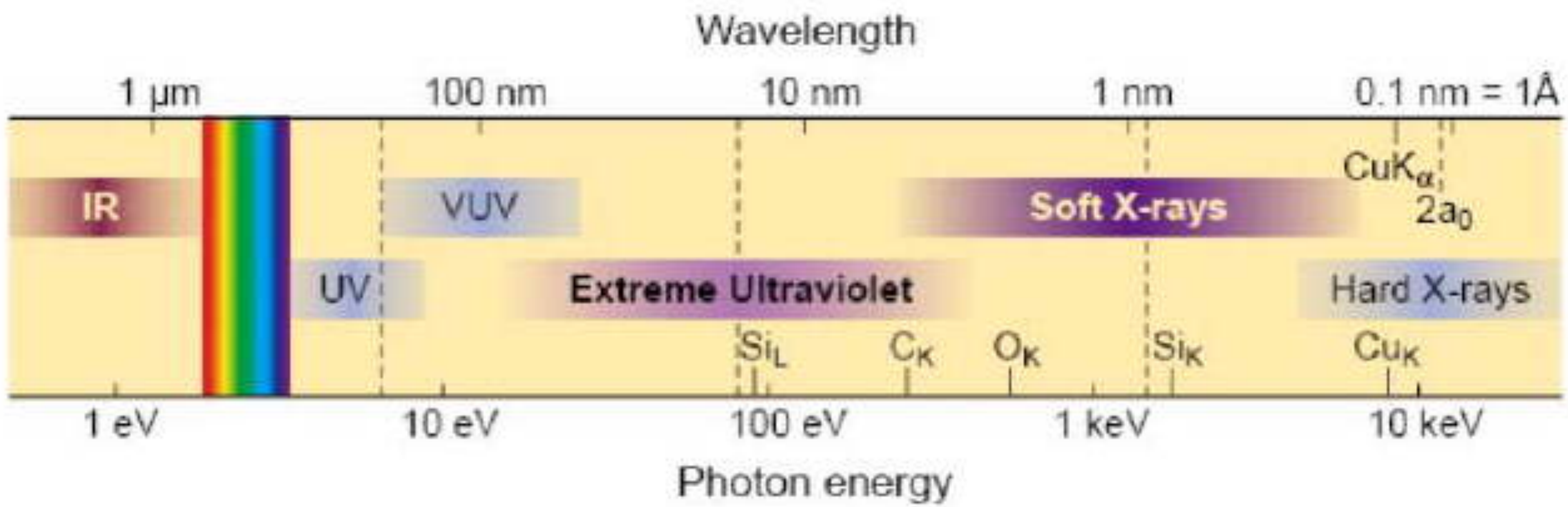
Light-polarized ARPES ($YbRh_2Si_2$)



$YbIr_2Si_2$



Spectral range and radiation sources



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

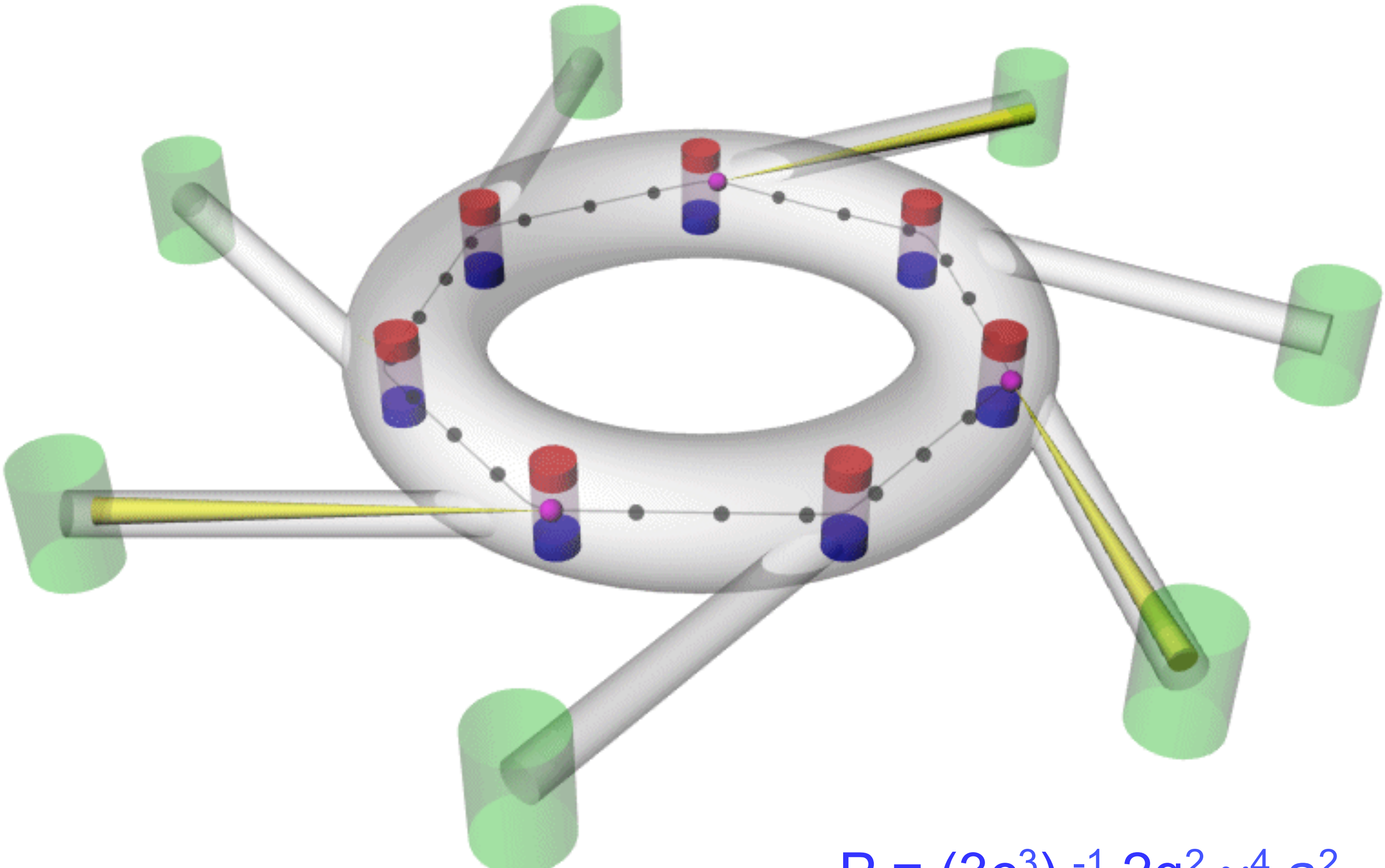
Disadvantages: (i) low intensity; (ii) discrete spectrum; (iii) no time structure

Revolution with synchrotron radiation !!!

Synchrotron Radiation

Synchrotrons/Storage Rings

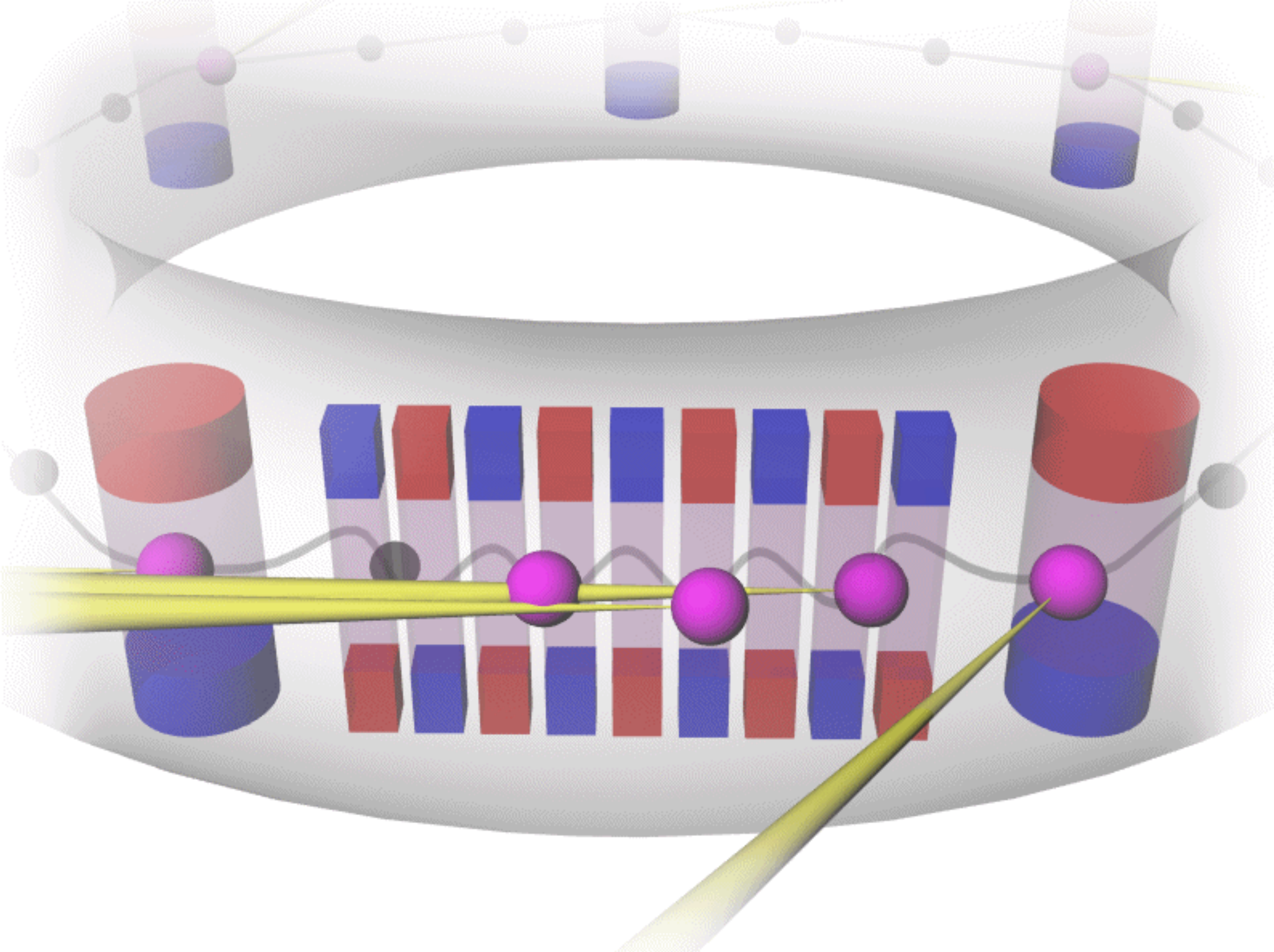
Synchrotron Radiation (dipoles)



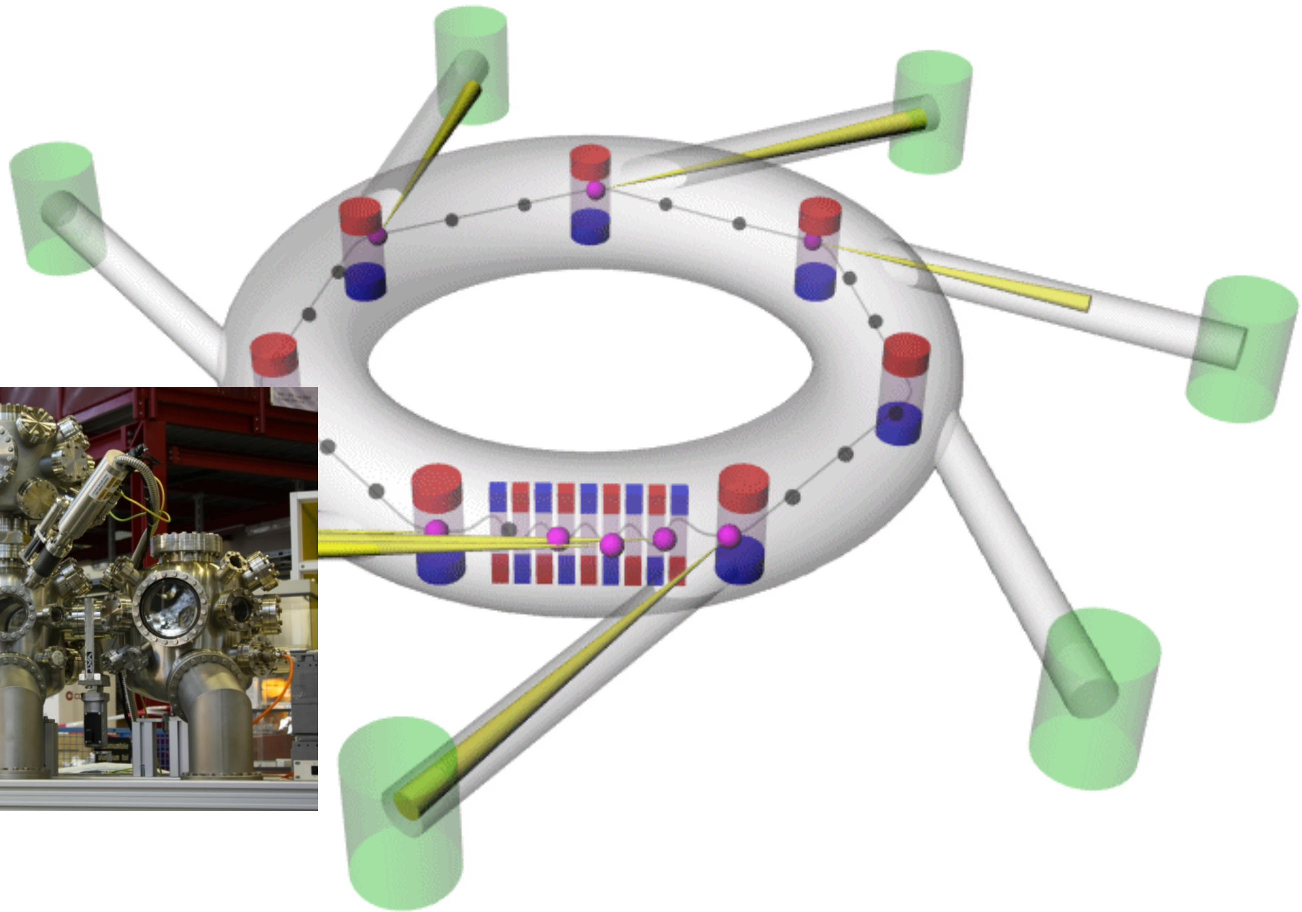
$$P = (3c^3)^{-1}\, 2q^2\, \nu^4\, a^2$$

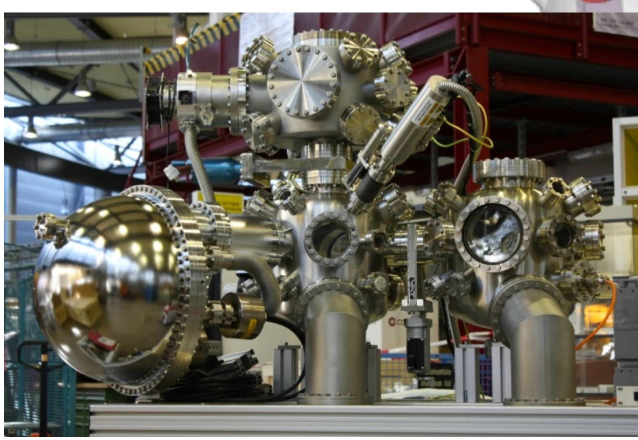
P – radiated power; c – light velocity; q – particle charge; a – acceleration; ν - normalized energy

Synchrotron radiation (undulators)



Synchrotron radiation (sources + exp. stations)





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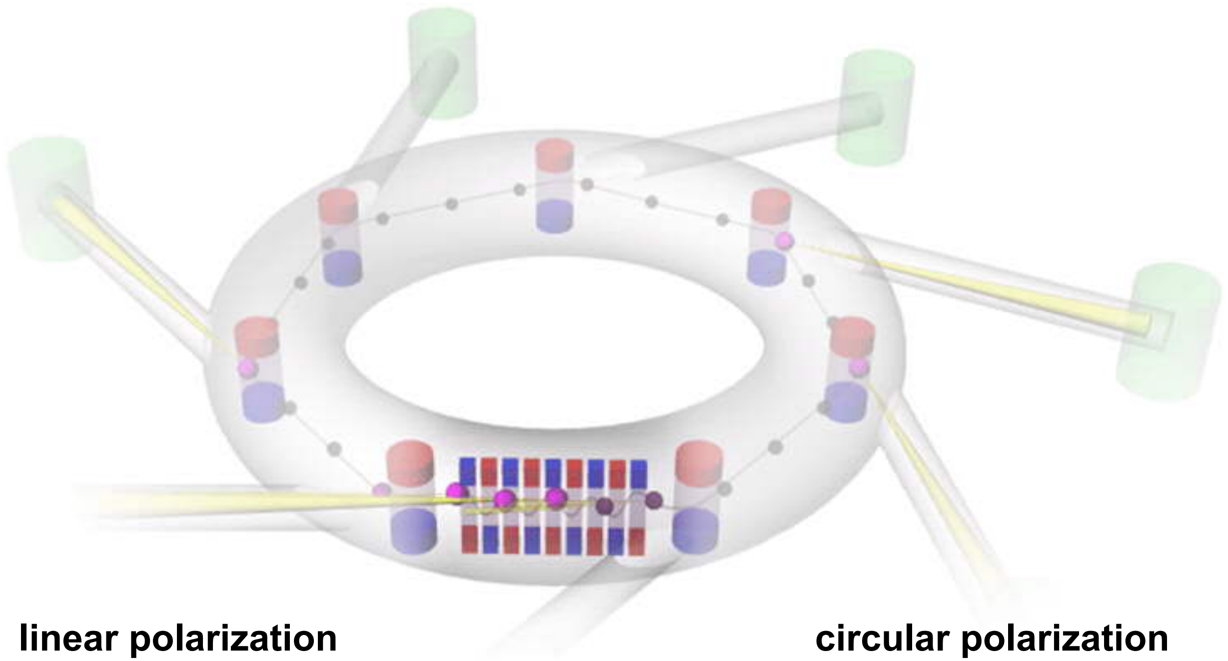
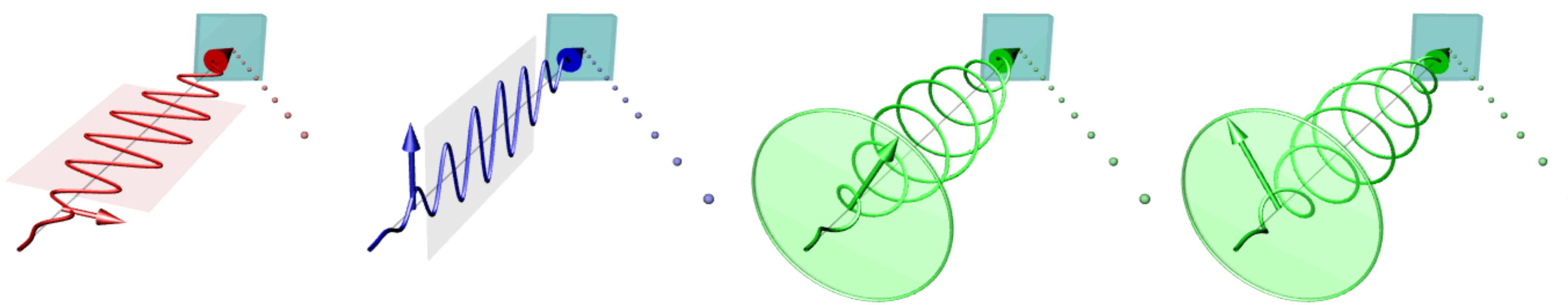
Synchrotron Radiation (light polarization)

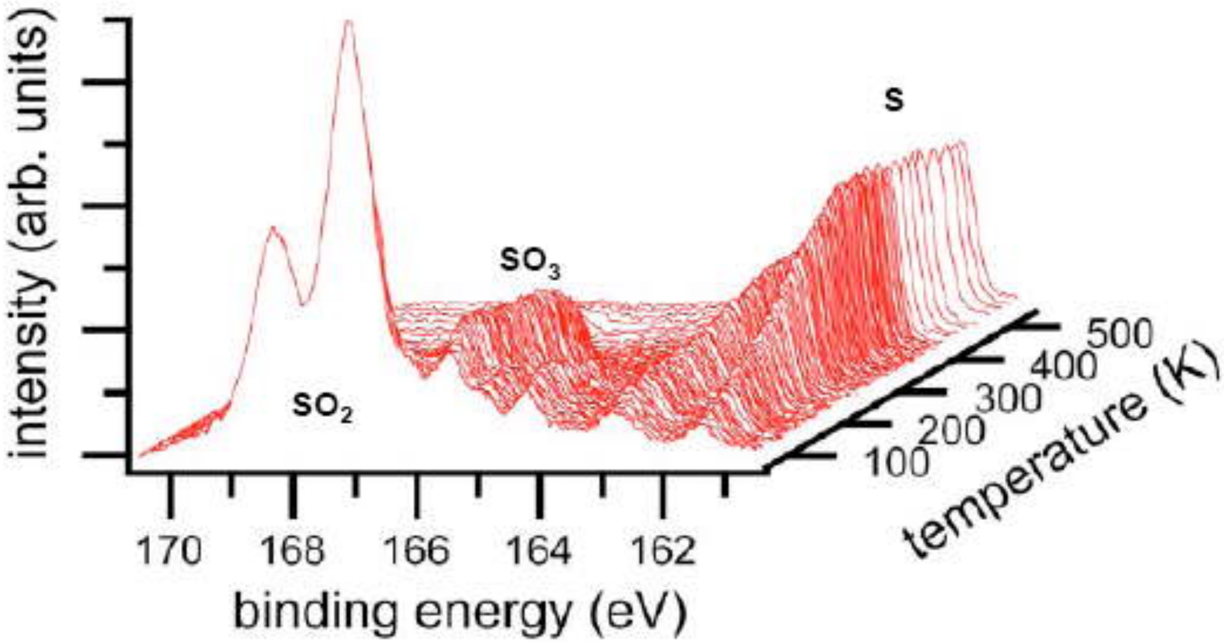


linear polarization **circular polarization**



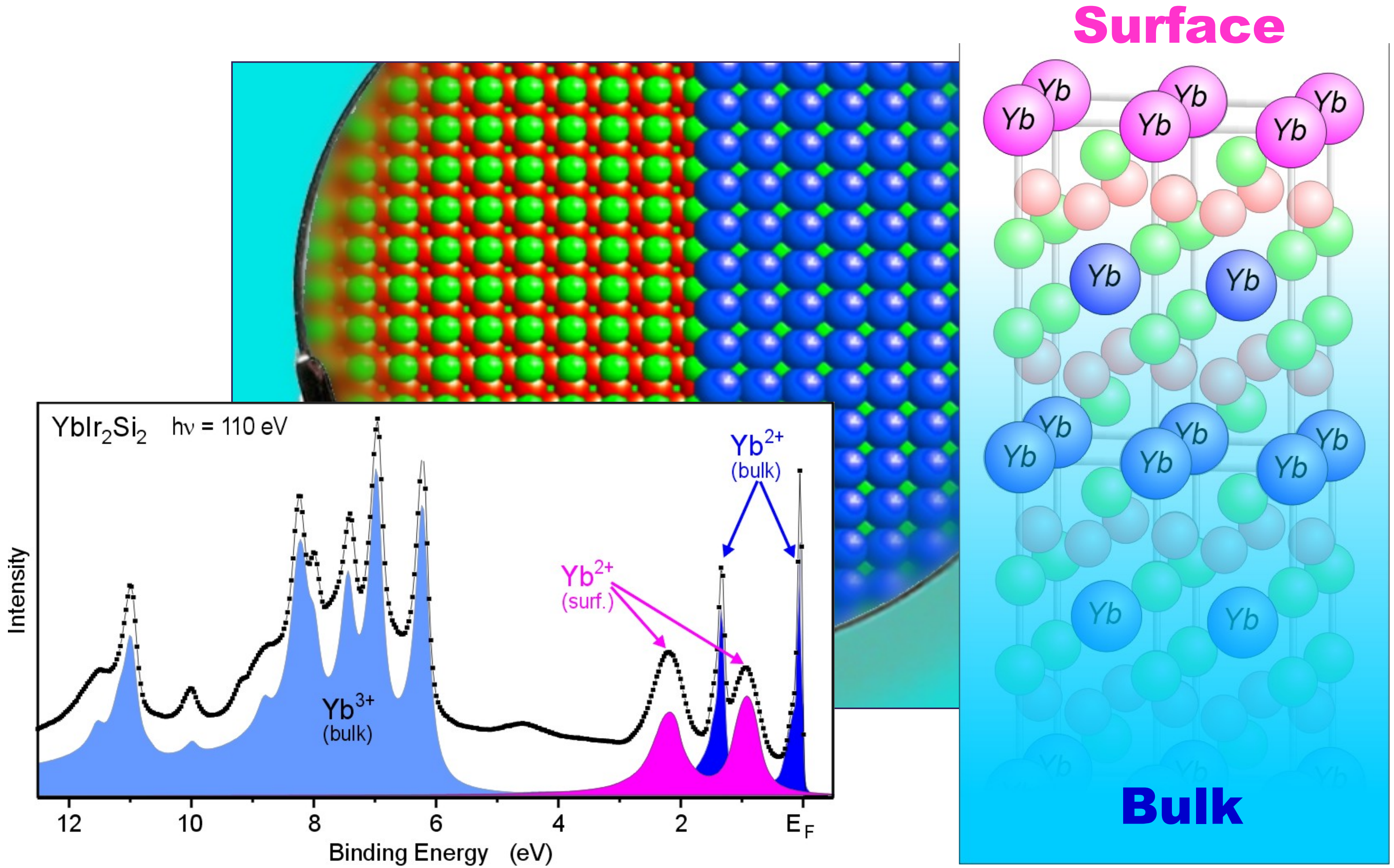
Spectroscopic toolbox: X-ray photoelectron spectroscopy, ESCA

SO_2 dissociation on Ru(001)

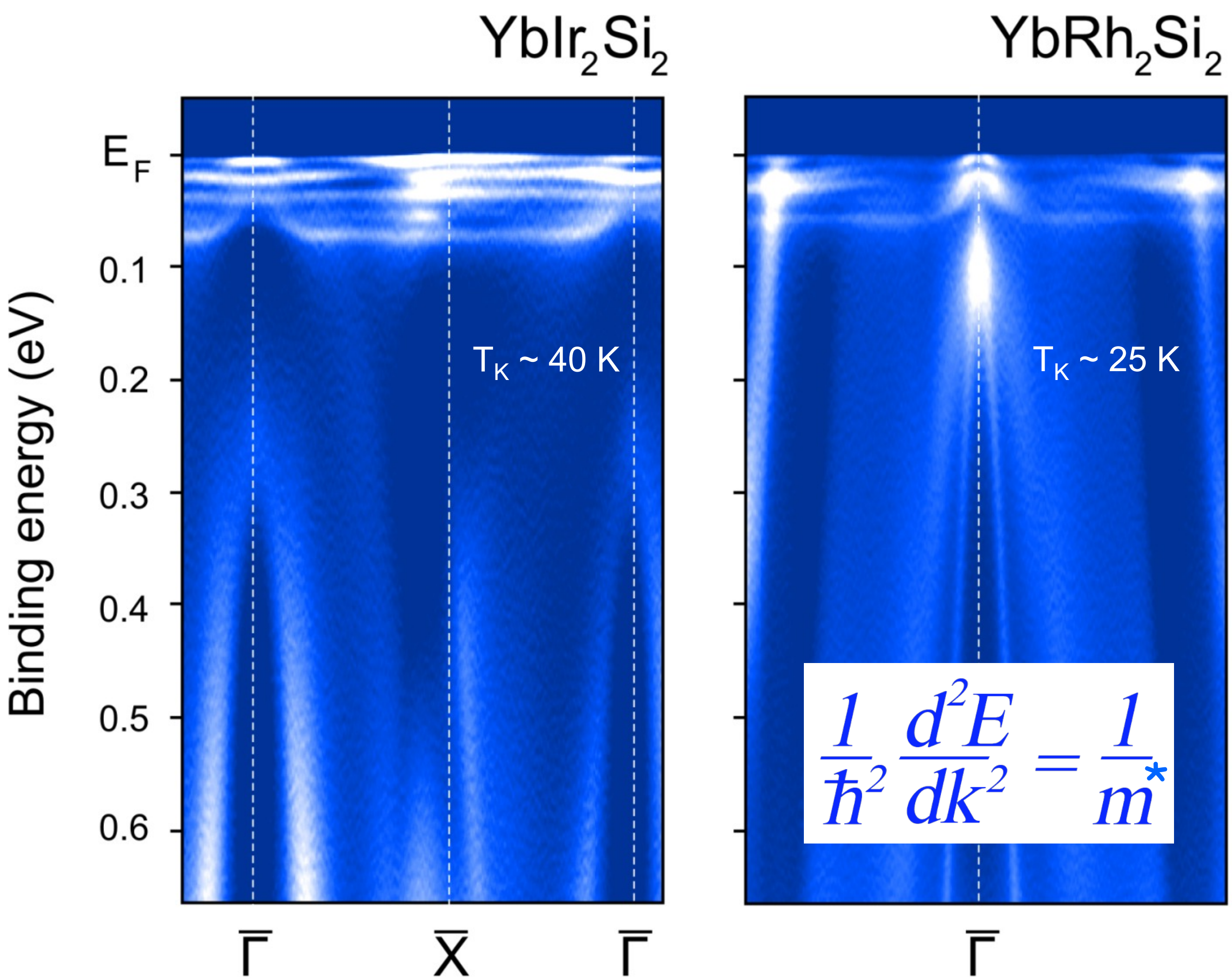


F. Hennies et al., J.Chem.Phys. 127, 154709 (2007)

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)

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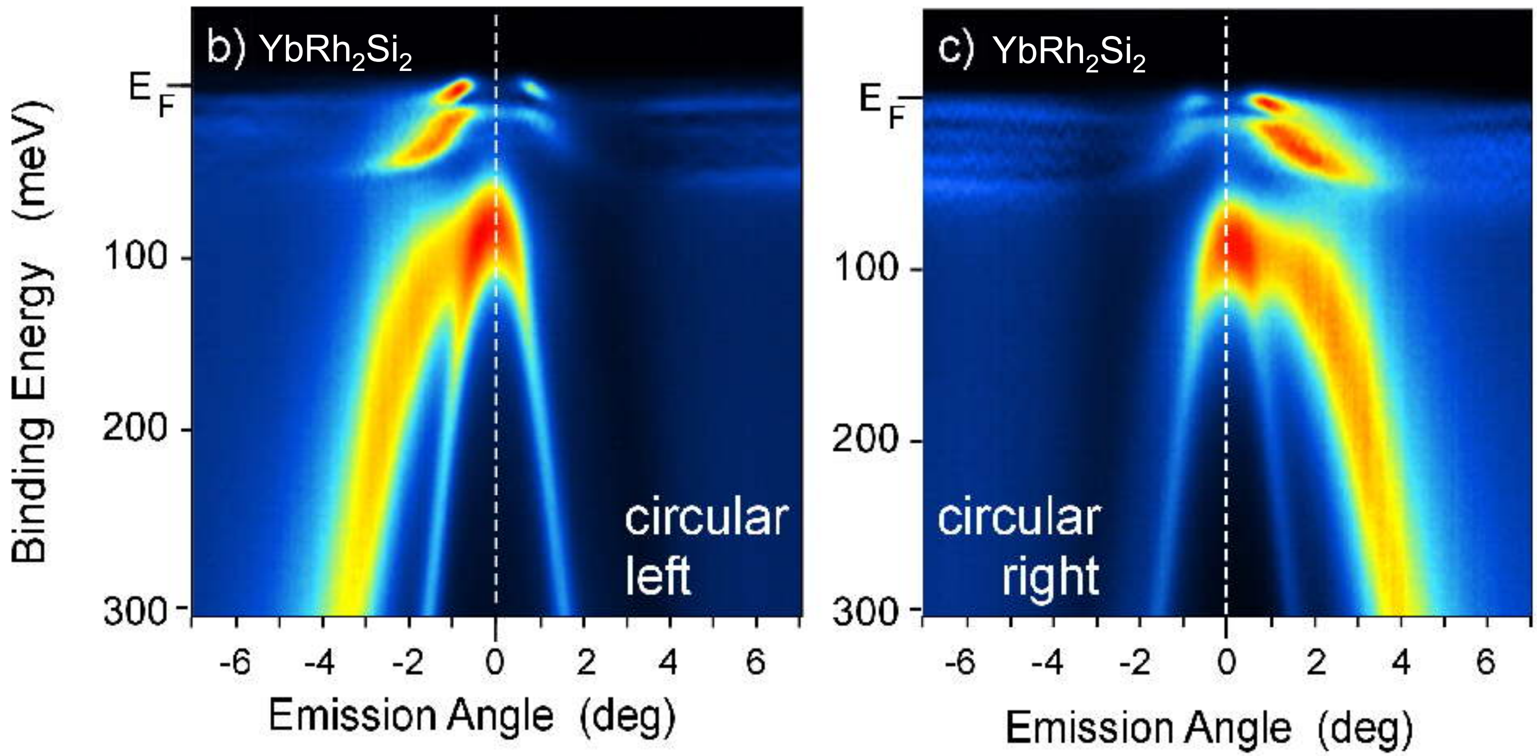
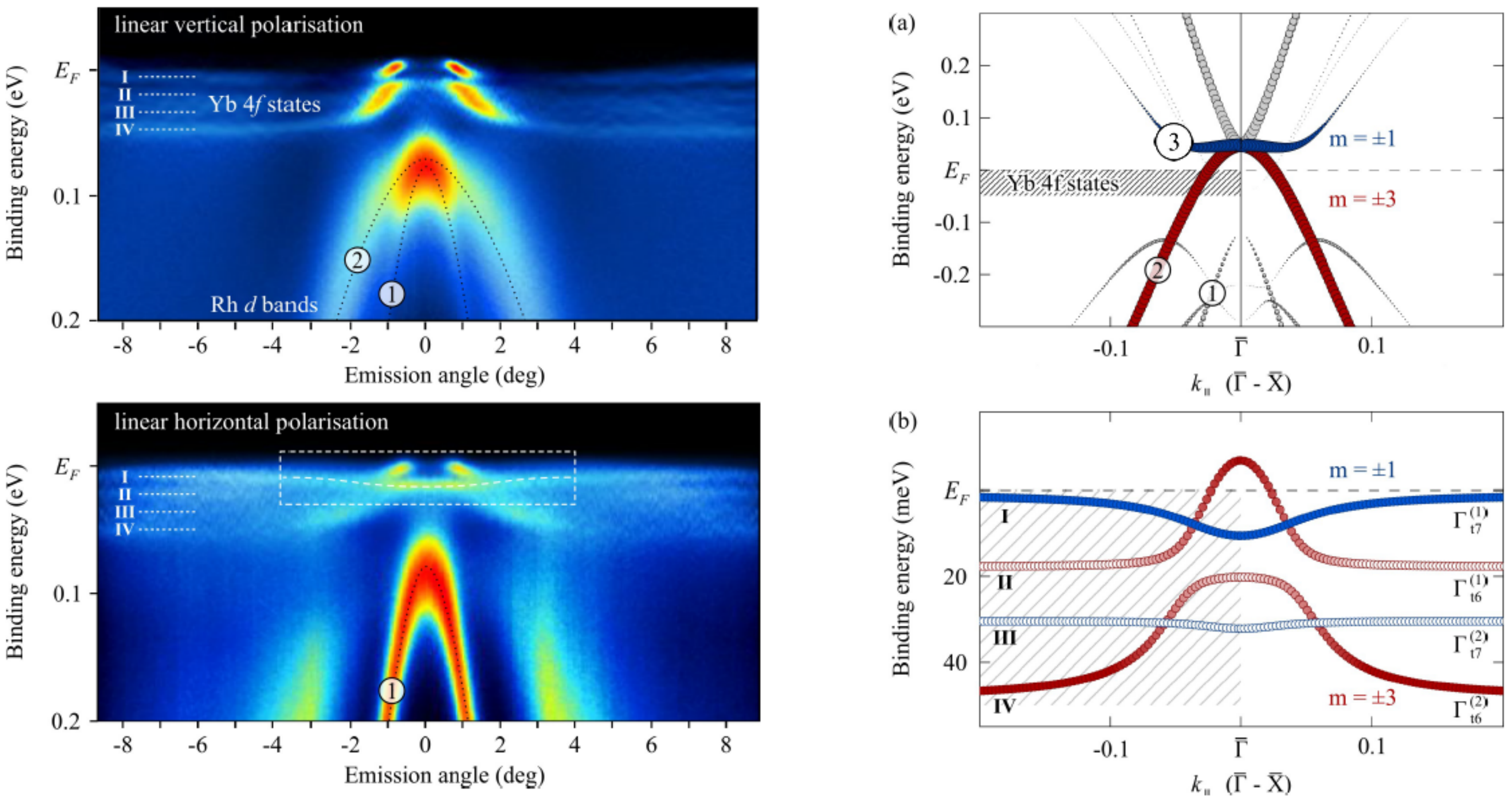
Symmetry of f-states in $YbRh_2Si_2$



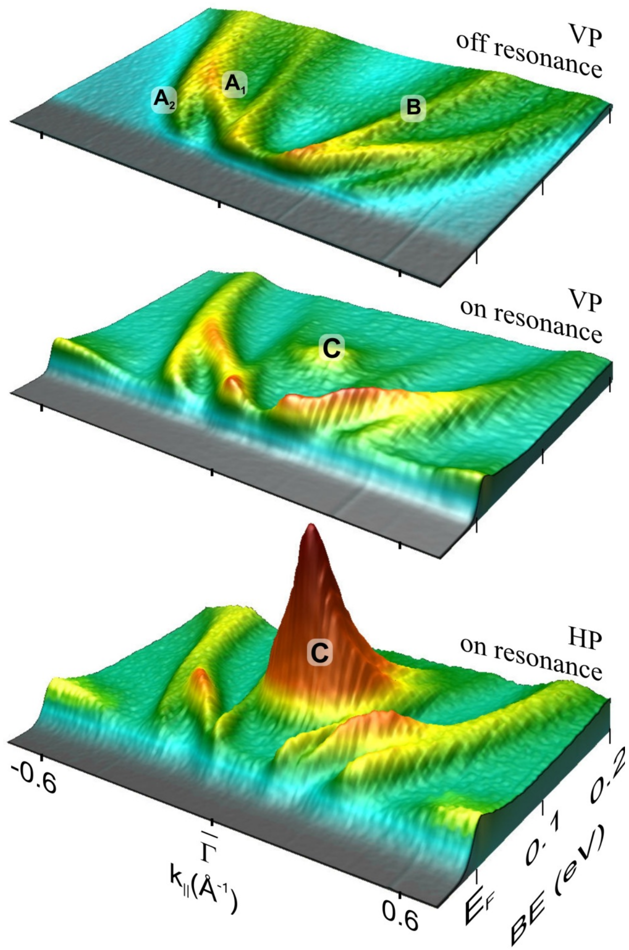
Heavy-fermion behavior in $YbRh_2Si_2$



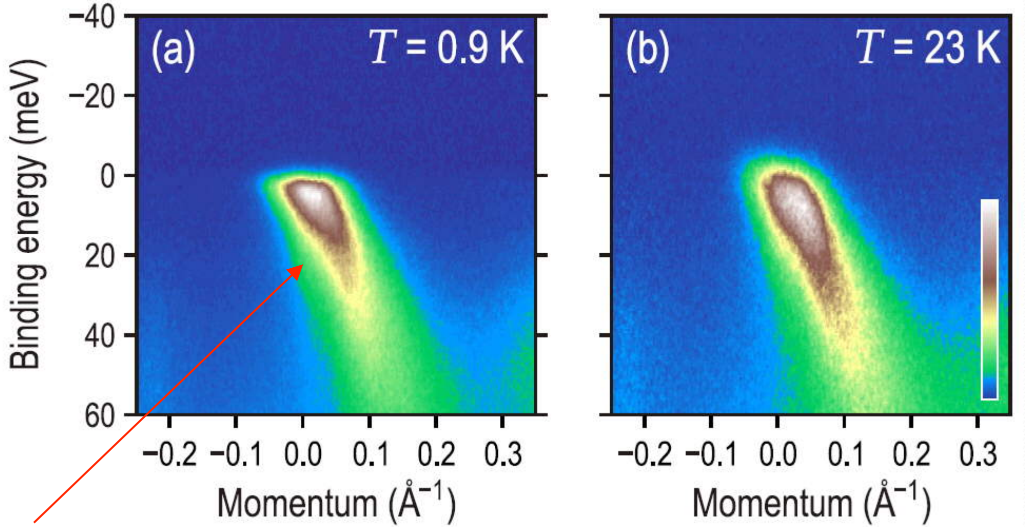
Dispersion of the 4f states around Γ where they hybridize to Rh d bands: Experiment & theory

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

CeFePO



LiFeAs



kink below T_c

- 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

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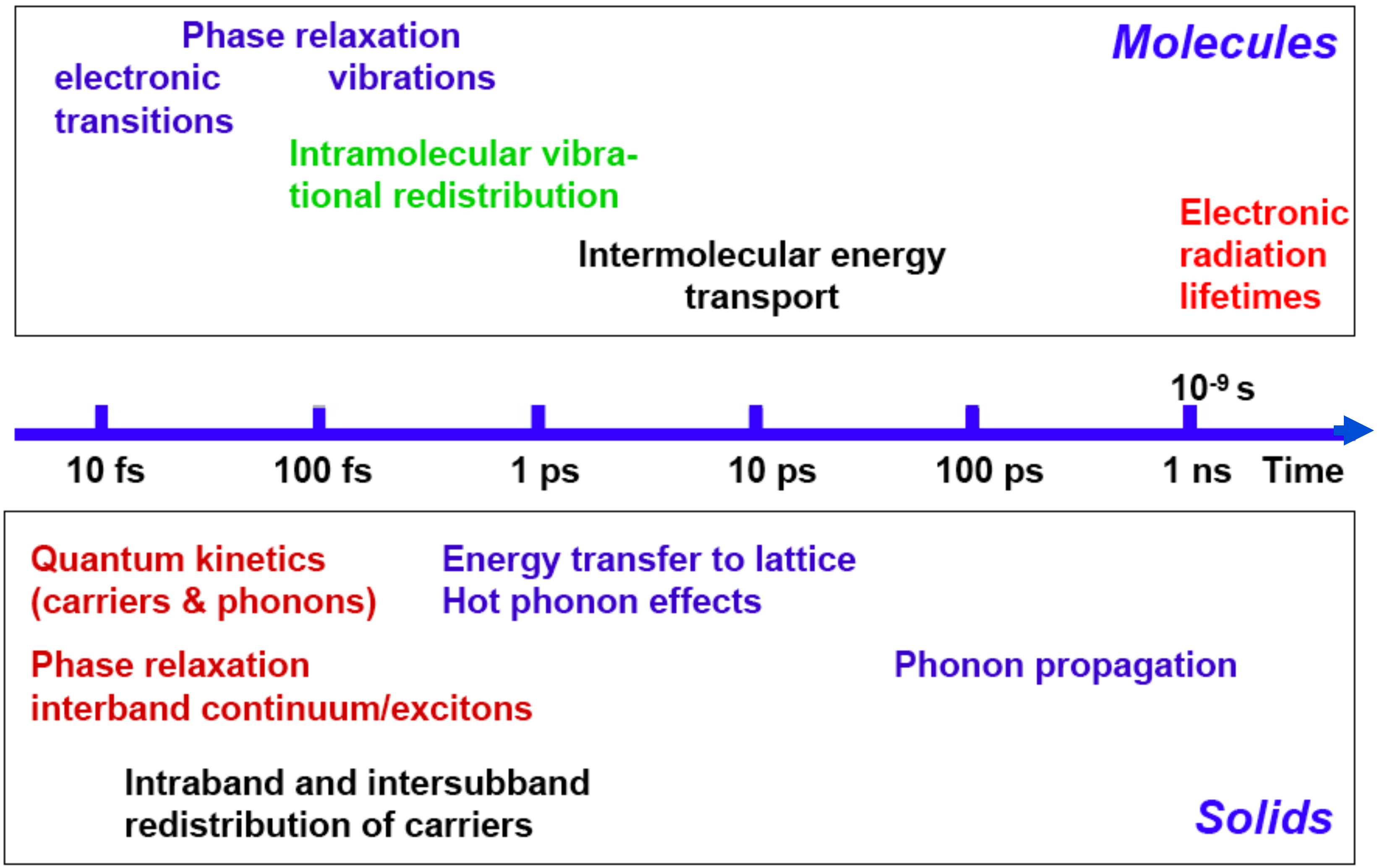
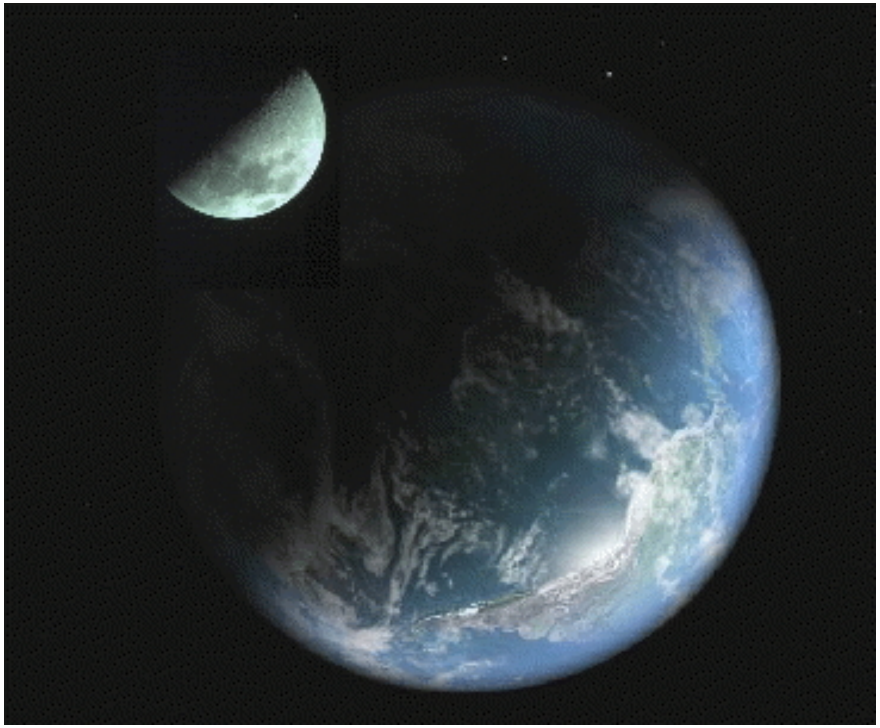
Time scales for dynamics



What is a picosecond?

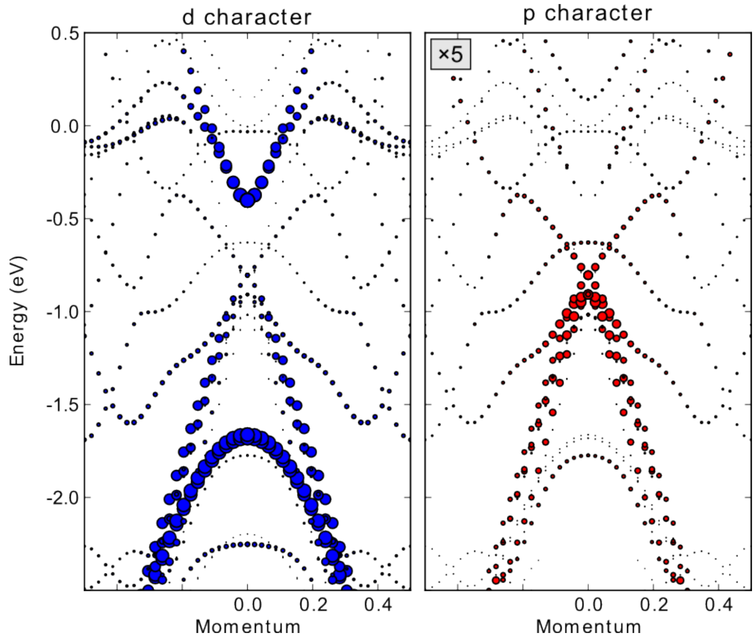
In 1 s light travels 300 000 km

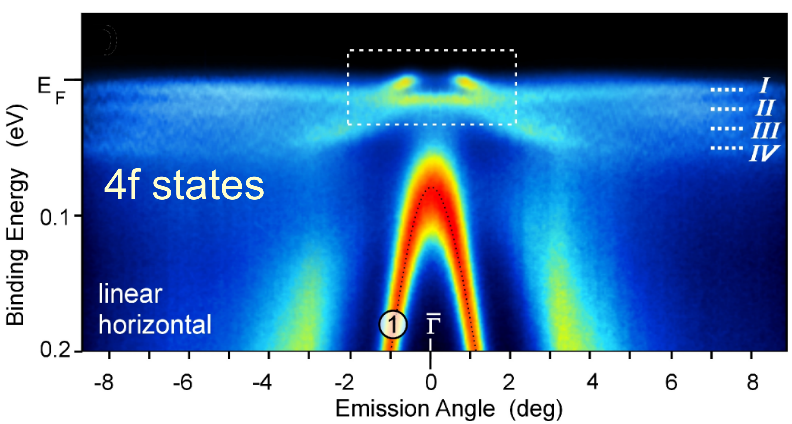


Distance between earth and moon is 384 000 km

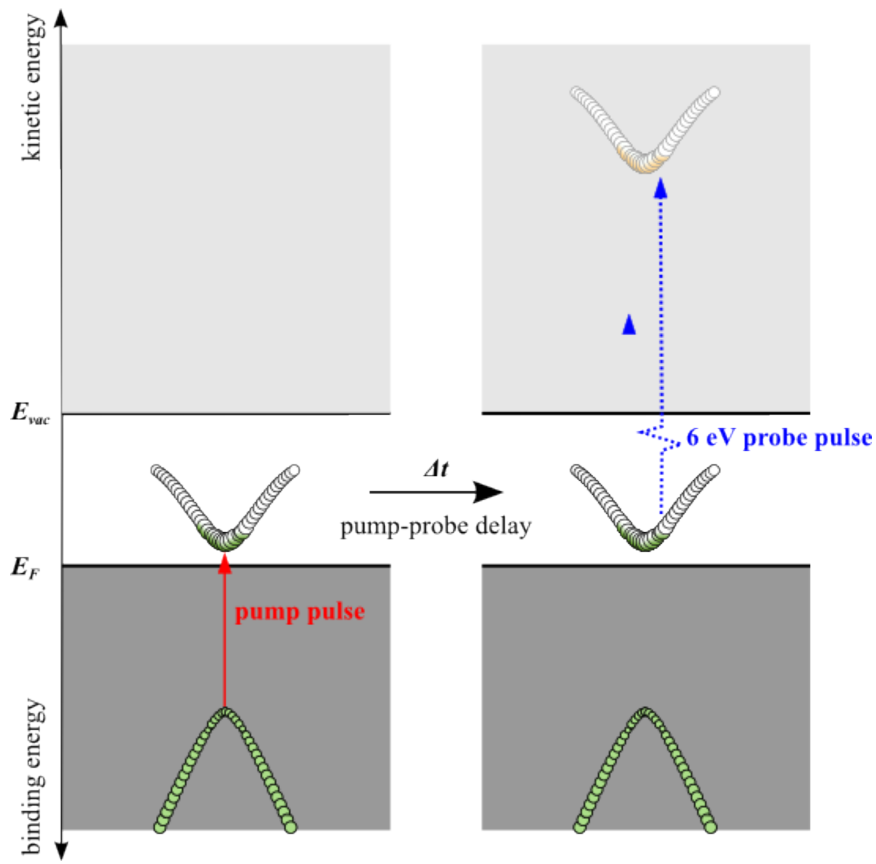
In 1 ps light travels 0,3 mm

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



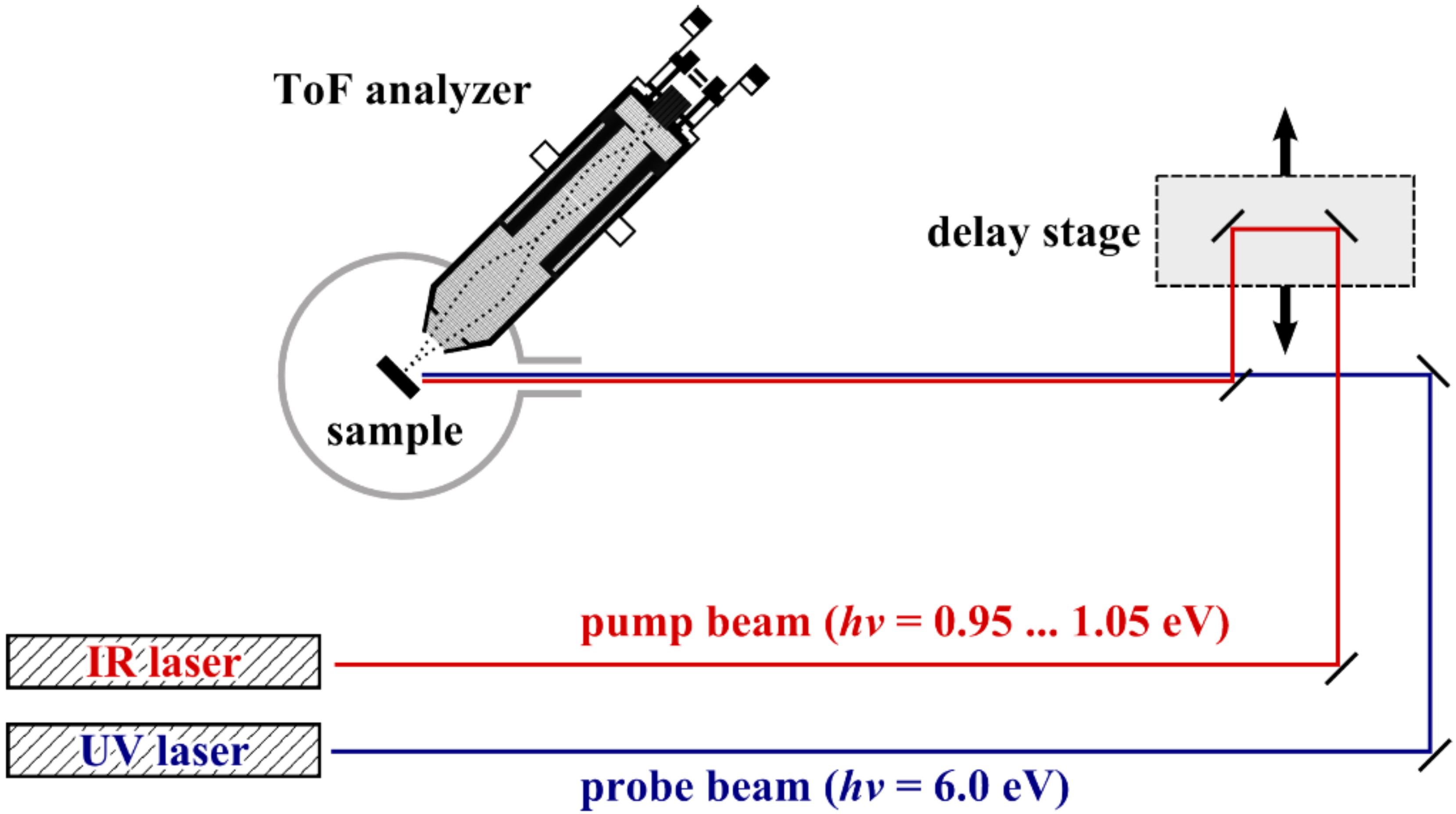


- $YbRh_2Si_2$

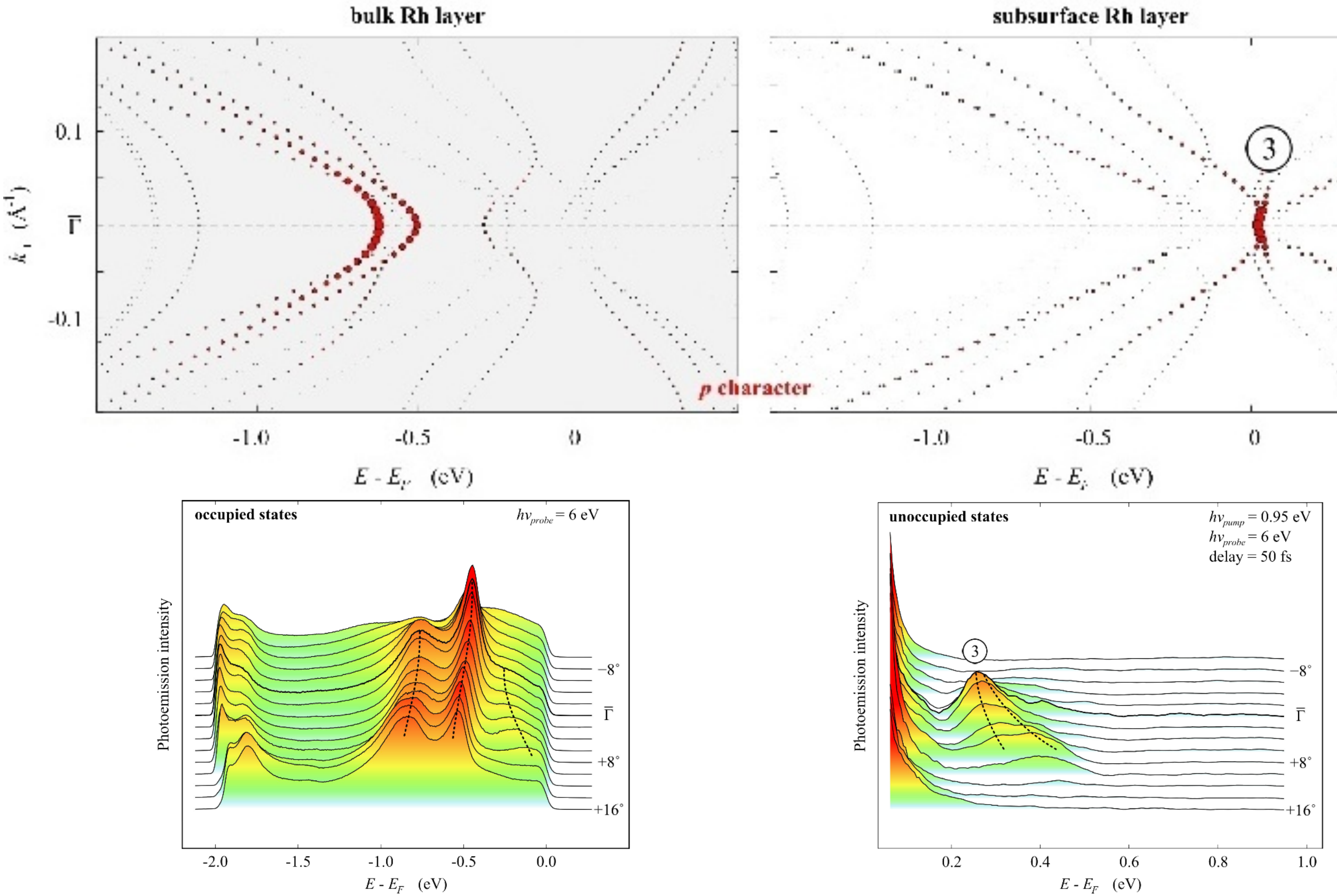


ARPES with MHz optical lasers

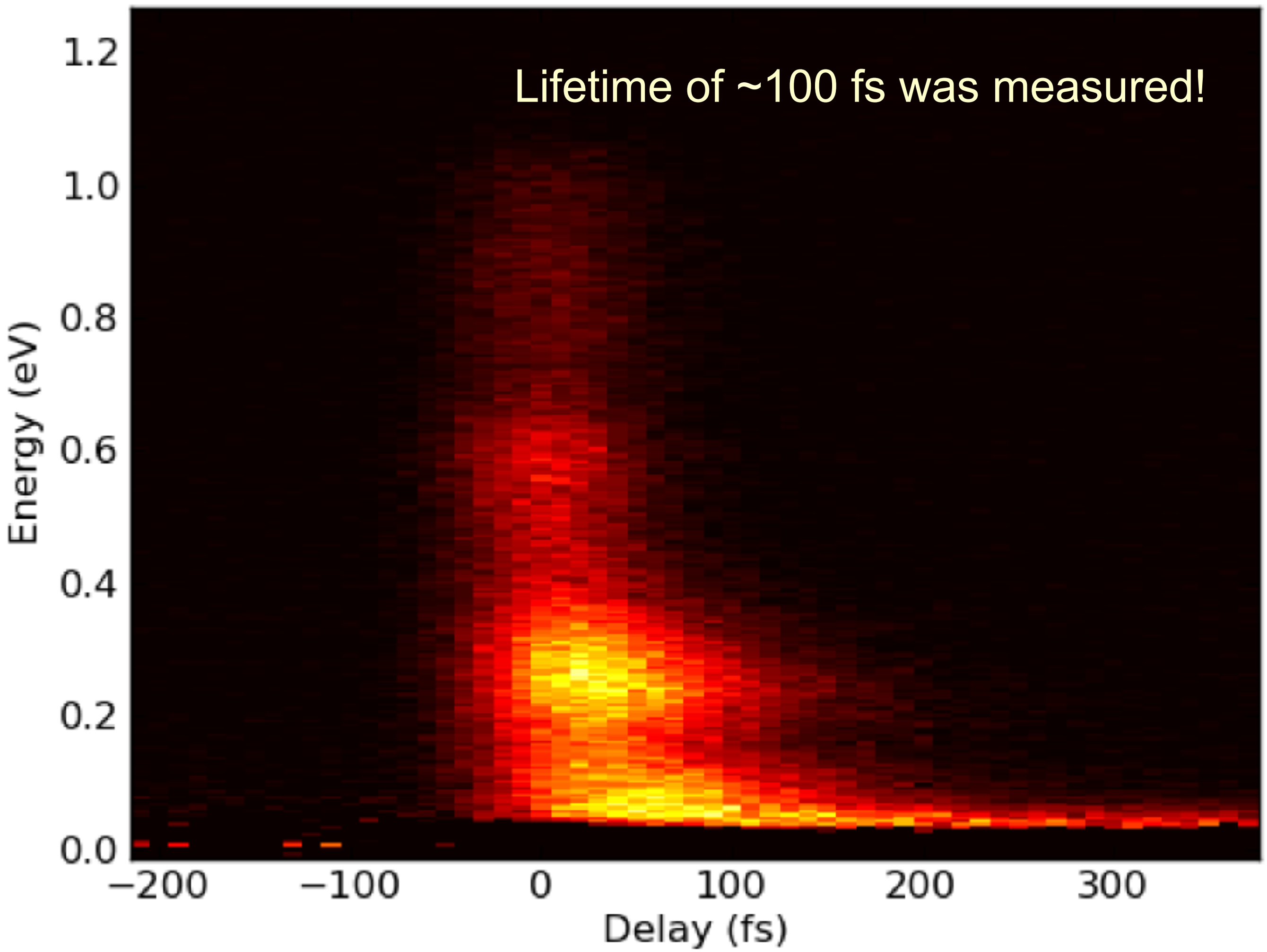
Experimental setup



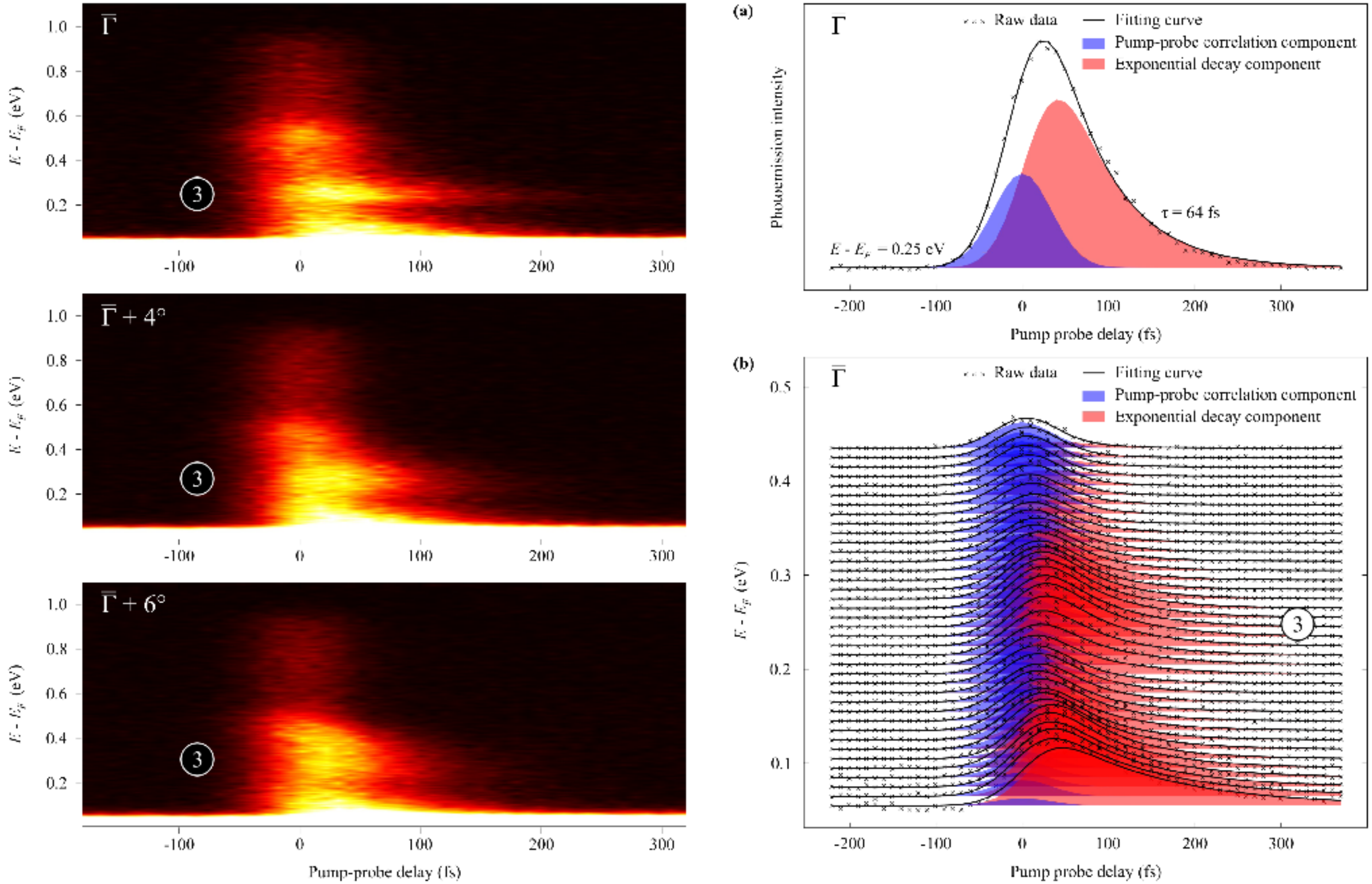
$YbRh_2Si_2$: Projected *p*-character band structure



Proof of principle pump-probe experiment

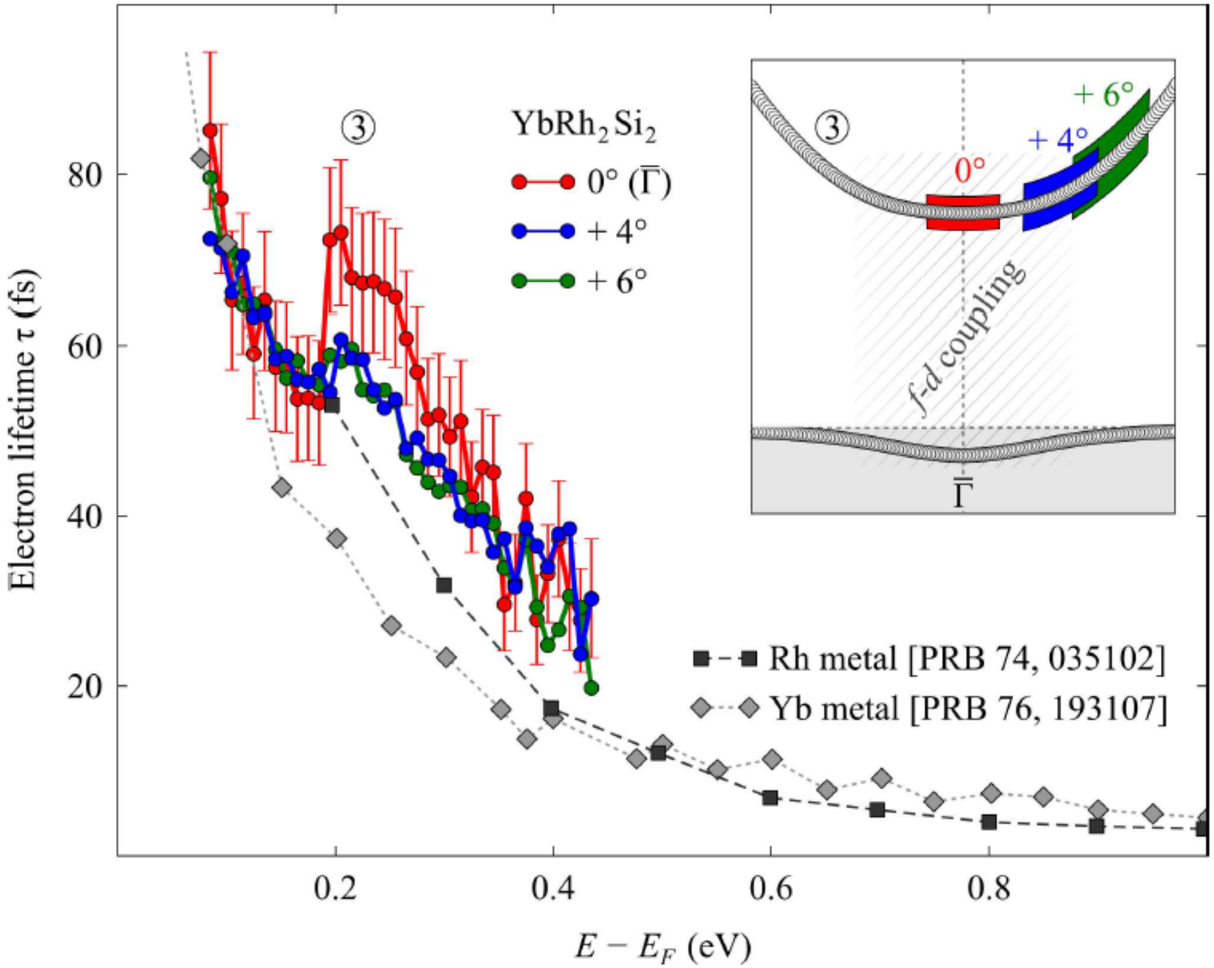


Angle-resolved pump-probe experiments



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

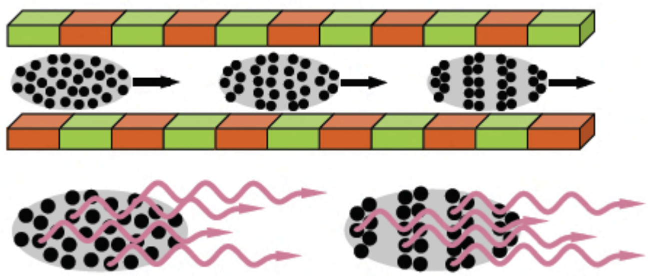
Go to hν close (high harmonic generation, HHG) or above (XFEL) 100 eV!

Photon sources for trARPES (Kai Rossnagel)

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

Strobe lights: FEL versus HHG

FEL (FLASH)

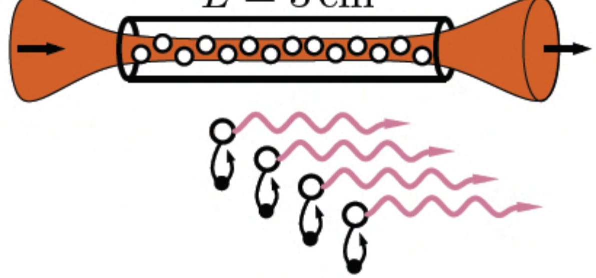


$h\nu \approx 25 - 300\,\mathrm{eV}$
(fundamental)

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

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

HHG



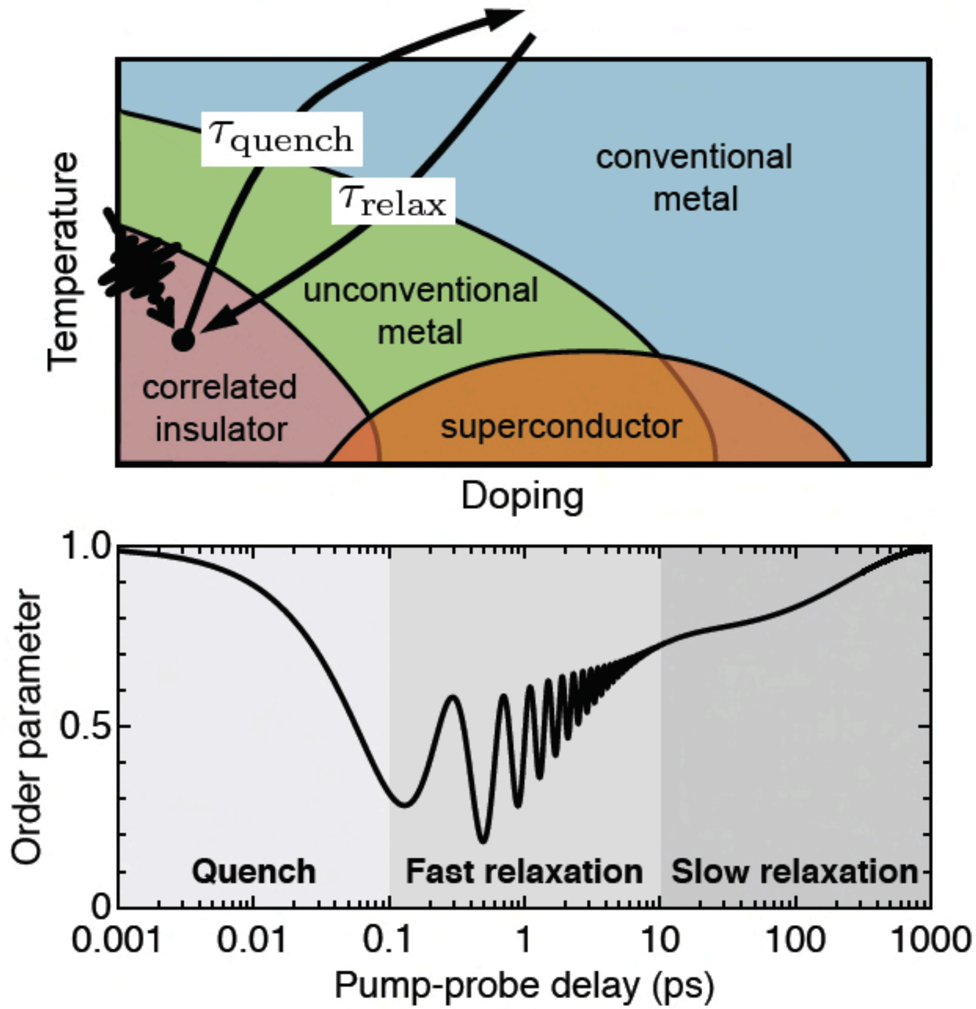
$h\nu < 100\,\mathrm{eV}$
(practically)

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

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

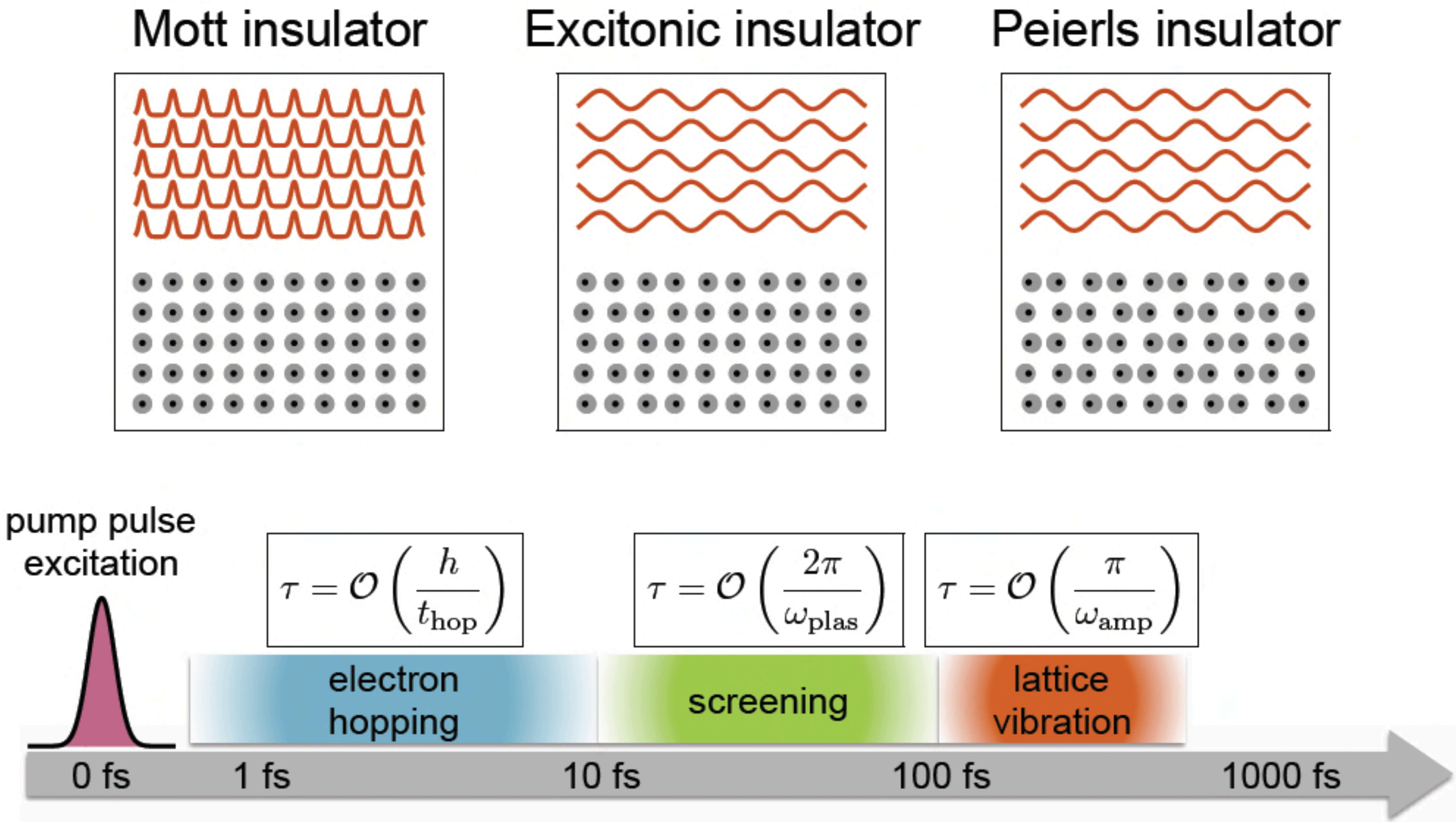
Study by destruction

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)

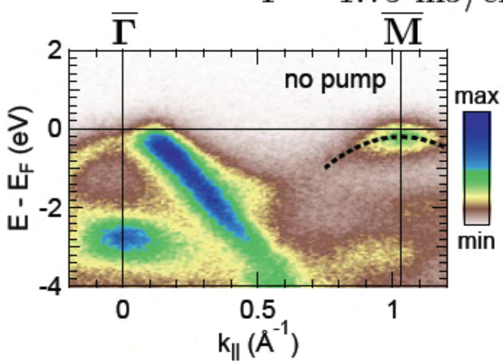
trARPES using HHG

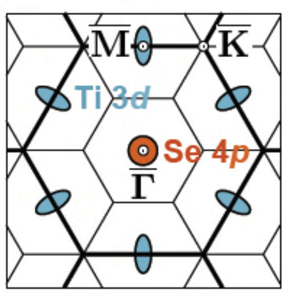
$T = 125\,\mathrm{K}$

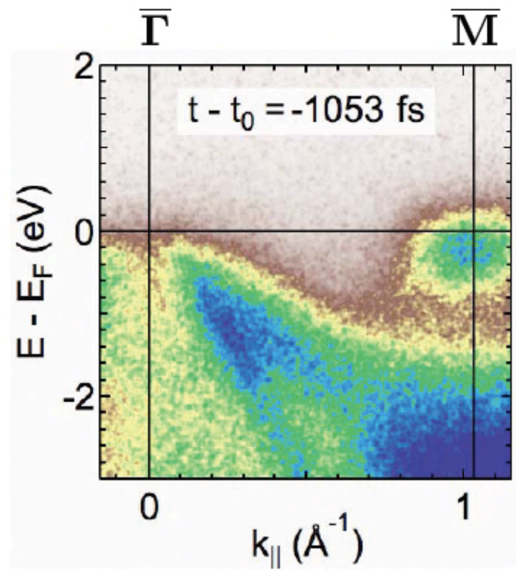
$h\nu_{\mathrm{pump}} = 1.57\,\mathrm{eV}$. $h\nu_{\mathrm{probe}} = 43\,\mathrm{eV}$

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

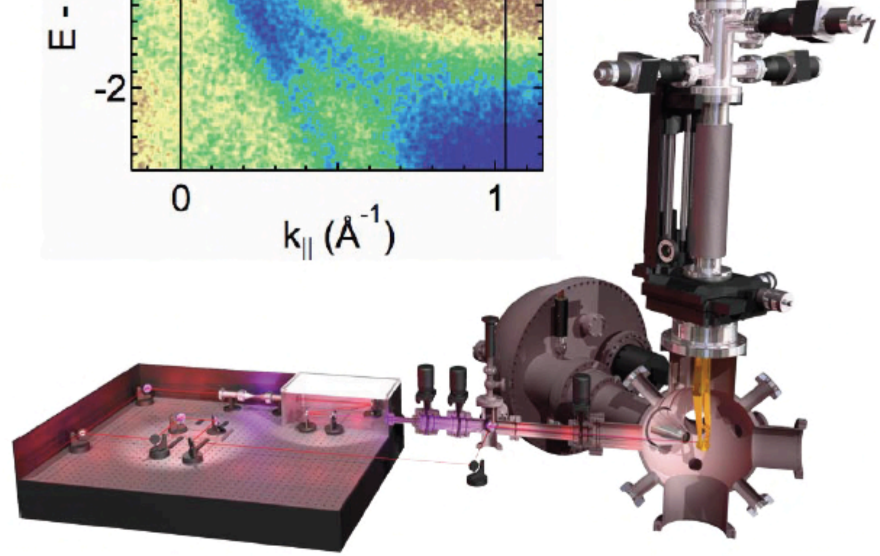
$F = 1.75\ \mathrm{mJ/cm}^2$







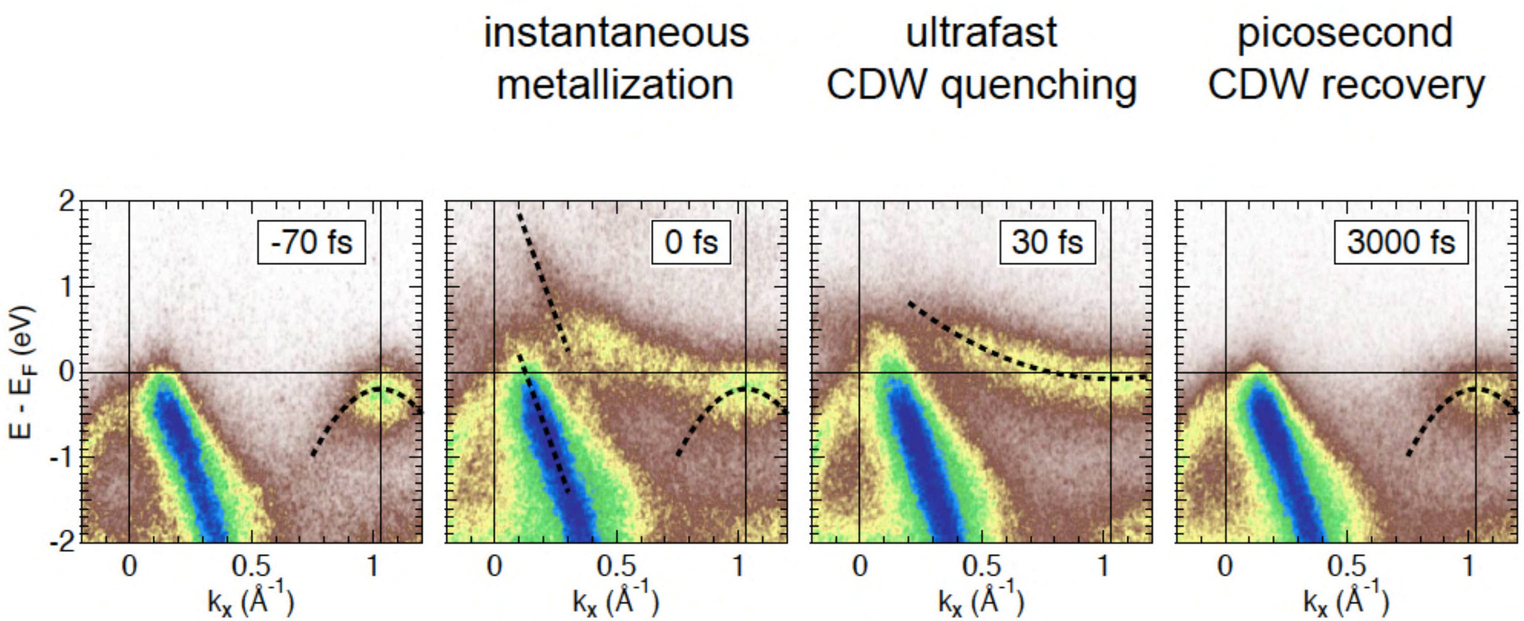
HHG setup
Bauer group, Kiel



What can be done with HHG sources?

Nature **471**, 490 (2011)

Snapshots



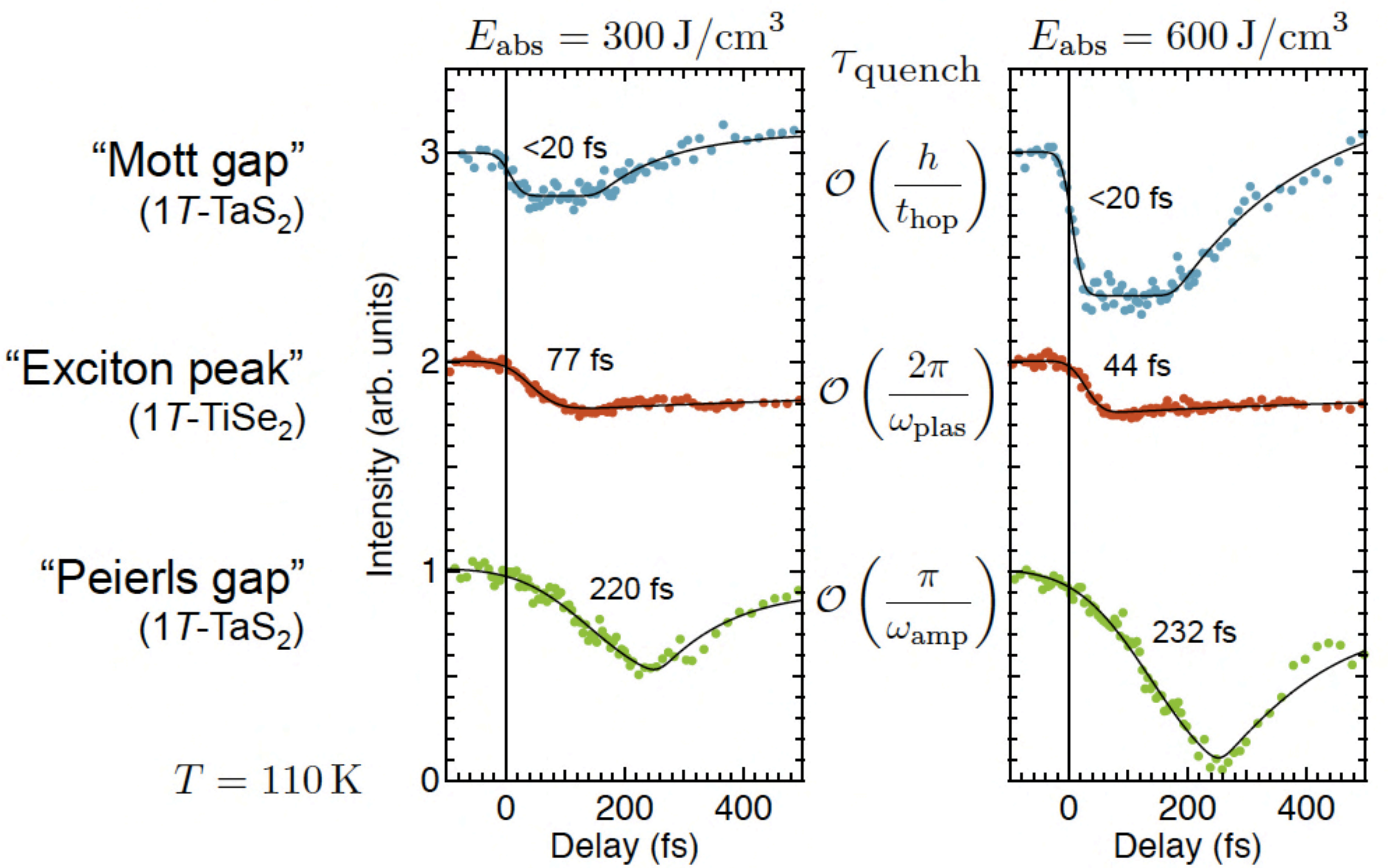
s-polarized probe

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

Examples of pump-probe research

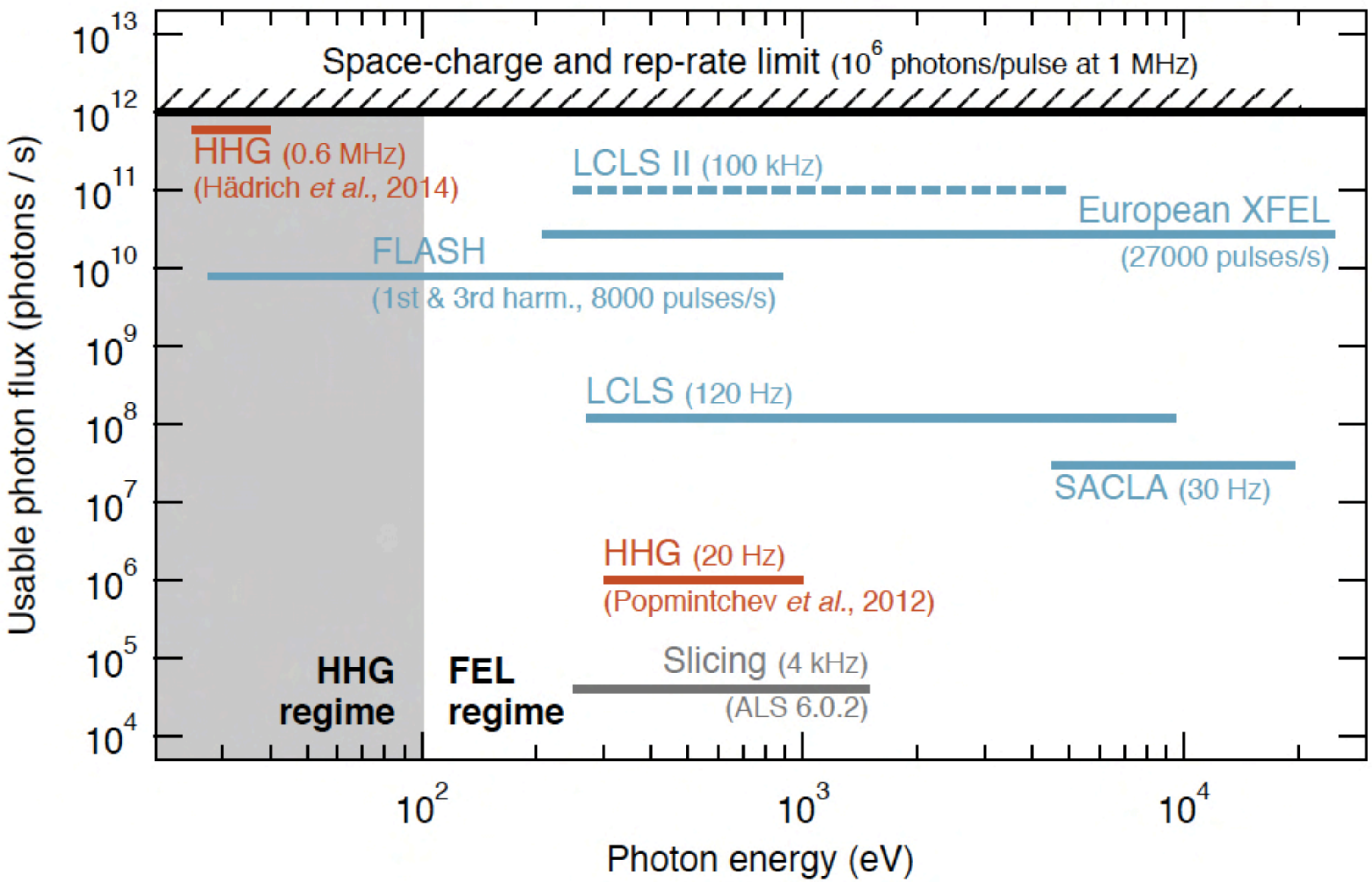
Nature Commun. **3**, 1069 (2012)

Hierarchy of quenching times



HHG and FELs: Complimentary tools

FEL photoemission



Problem

Problem to be solved:

Please outline core-level PES spectra (intensity versus kinetic energy of electrons relative to the Fermi level) of an elemental sample that is characterized by electron-energy levels:
3p – 18.3 eV (binding energy); 3s – 34.8 eV; 2p – 297.3 eV and 294.6 eV; 2s – 378.6 eV that are excited by radiation with wavelength 8.26 and 3.13 nm. Which element builds this sample?

Thank you for your attention
and
see you again today