The dual nature of 4*f* electrons in rare-earth intermetallics: ARPES view

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Collaboration:

PES and ARPES experiments:

University of Technology (Dresden, Germany)

- C. Laubschat
- D.V. Vyalikh
- S. Danzenbächer.
- K. Kummer (now: ESRF, Grenoble, France)
- M. Höppner (now: Max-Plank-Institute for Solid State Research, Stuttgart, Germany)

High-quality single-crystal samples:

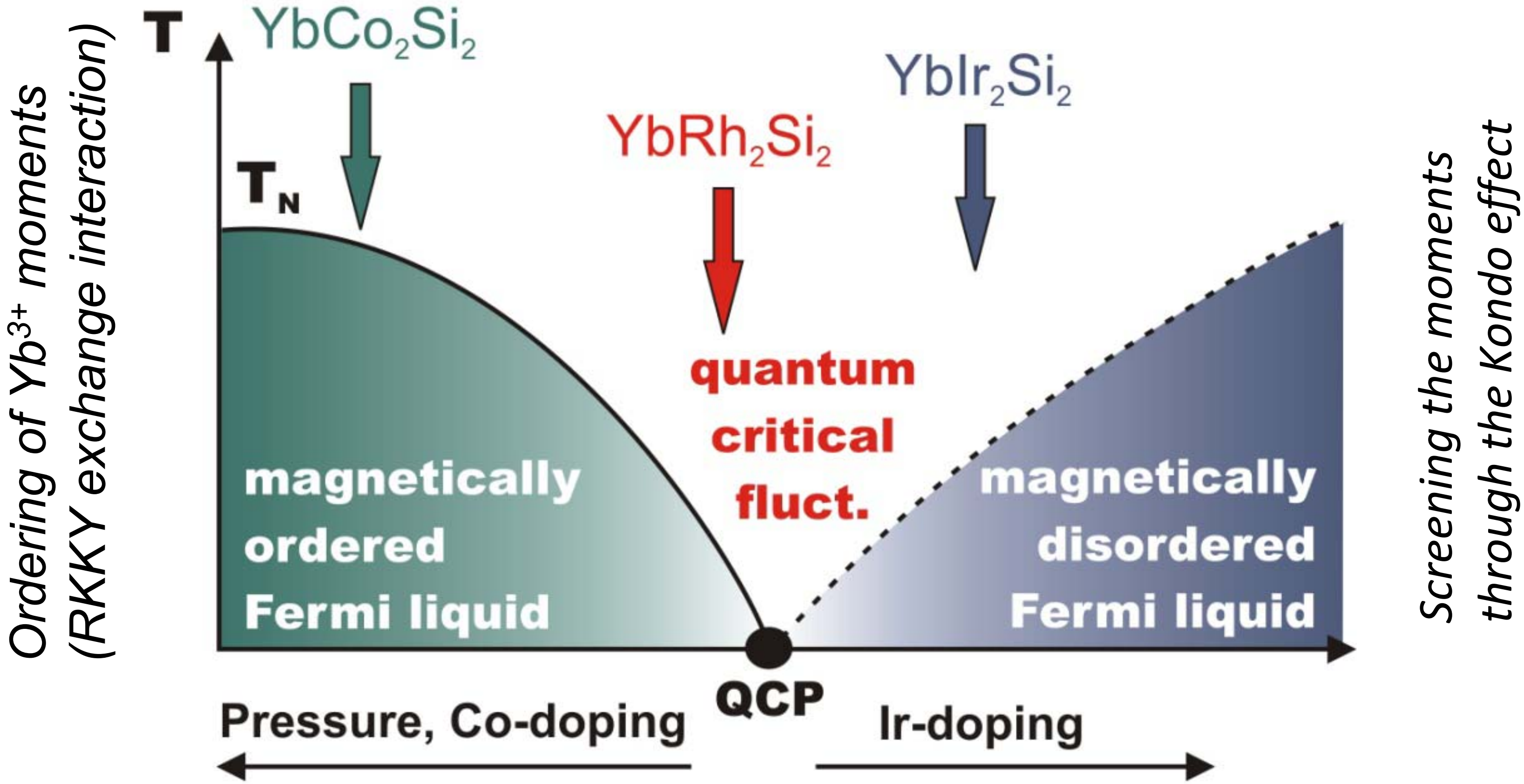
Max-Plank-Institute for Chemical Physics of Solids (Dresden, Germany)

- C. Geibel
- C. Crellner

Equipment:

- Swiss light source, Paul Scherrer Institute (Villigen, Switzerland)
- BESSY GmbH (Berlin, Germany)

Magnetic order vs. Kondo lattice



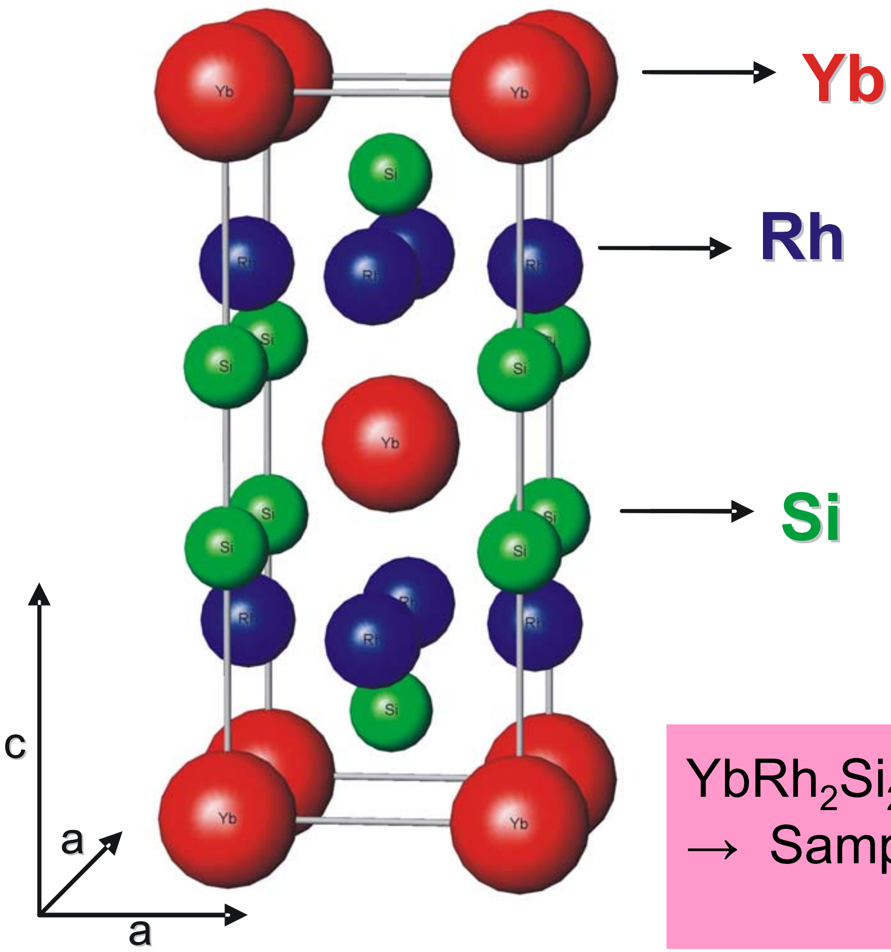
Magnetic order disappears usually continuously → Quantum Critical Point (QCP)
Formation of heavy quasiparticles (heavy fermions)

- $YbRh_2Si_2$: Prominent QCP system → Very close to a QCP (T_N = 70 mK)

Questions to the electronic structure of $YbRh_2Si_2$

- What is the role of Yb 4*f* states in the electronic structure at the Fermi level (formation of the Fermi surface) ?
- What 4*f* hole state in the $4f^{13}$ configuration of Yb^{3+} forms the ground state in $YbRh_2Si_2$?
- How can the observed experimental results be described in terms of hybridization between the 4*f* states and valence bands ?

Crystal structure



$ThCr_2Si_2$ structure type:
The body-centered tetragonal lattice
(space group *I4/mmm*, No.139),
so called 122 phase

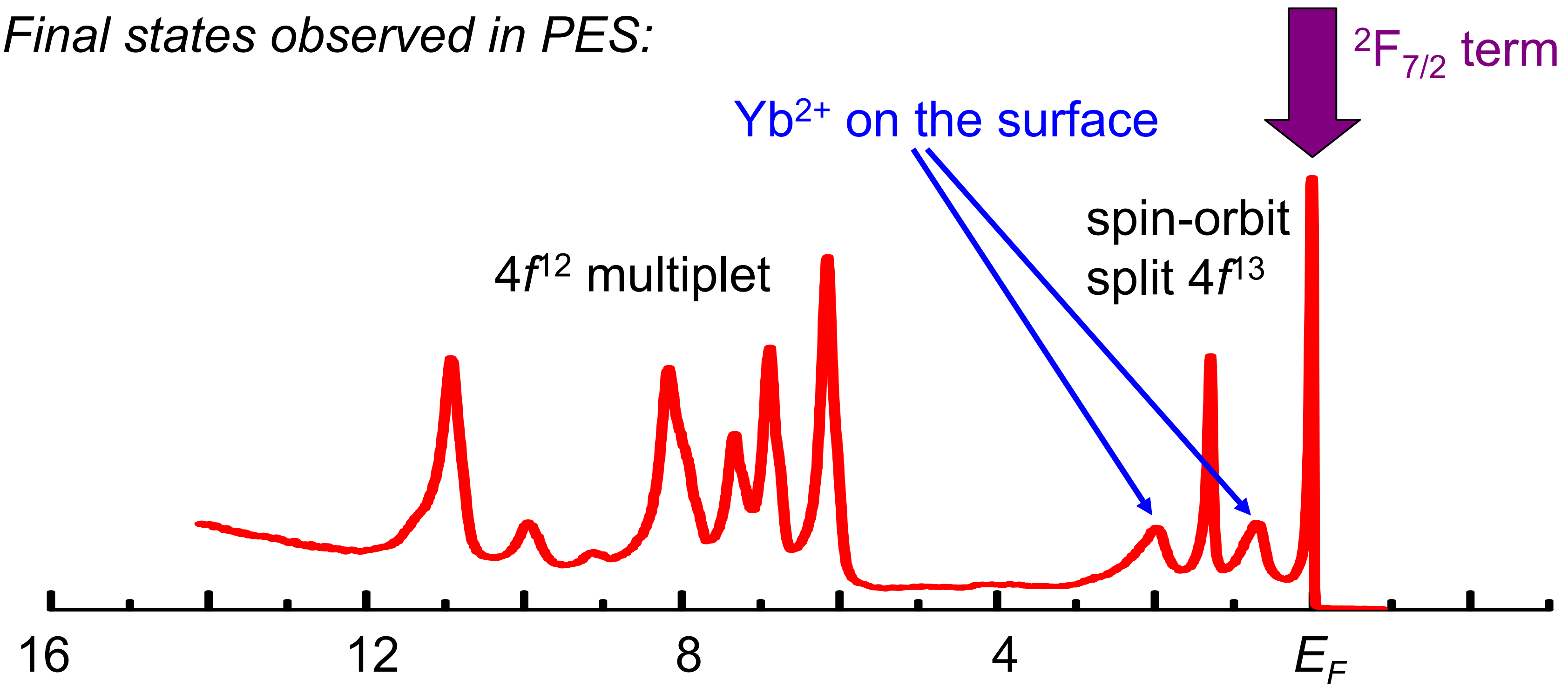
Stacks of alternating elemental layers consisting of only one type of atoms:
– Rh – Si – Yb – Si – Rh –

$YbRh_2Si_2$ usually cleaves along the Yb layers
→ Sample surfaces terminated either by Si or Yb atoms

Photoemission spectra

Bulk Yb ground state – mainly $4f^{13}$ configuration, small admixture of $4f^{14}$

Final states observed in PES:



$4f^{13}$ final-state configuration (directly observed in PES) could reflect some features of the **Yb ground state**

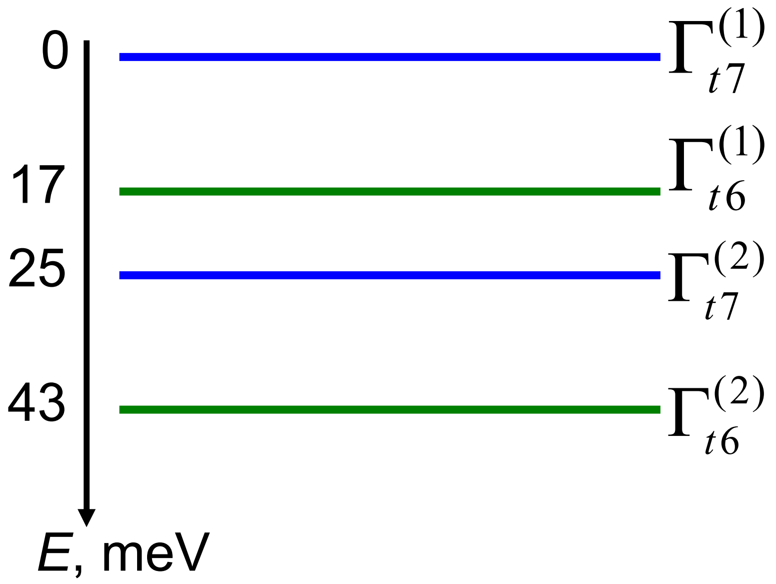
Crystal electric field (CEF) splitting

The $^2F_{7/2}$ multiplet term of the Yb $4f^{13}$ final-state configuration (with $4f$ hole in the j =7/2 state) – close to the Fermi level

Tetragonal crystal field splits it into 4 Kramers doublets related to the irreducible representations:

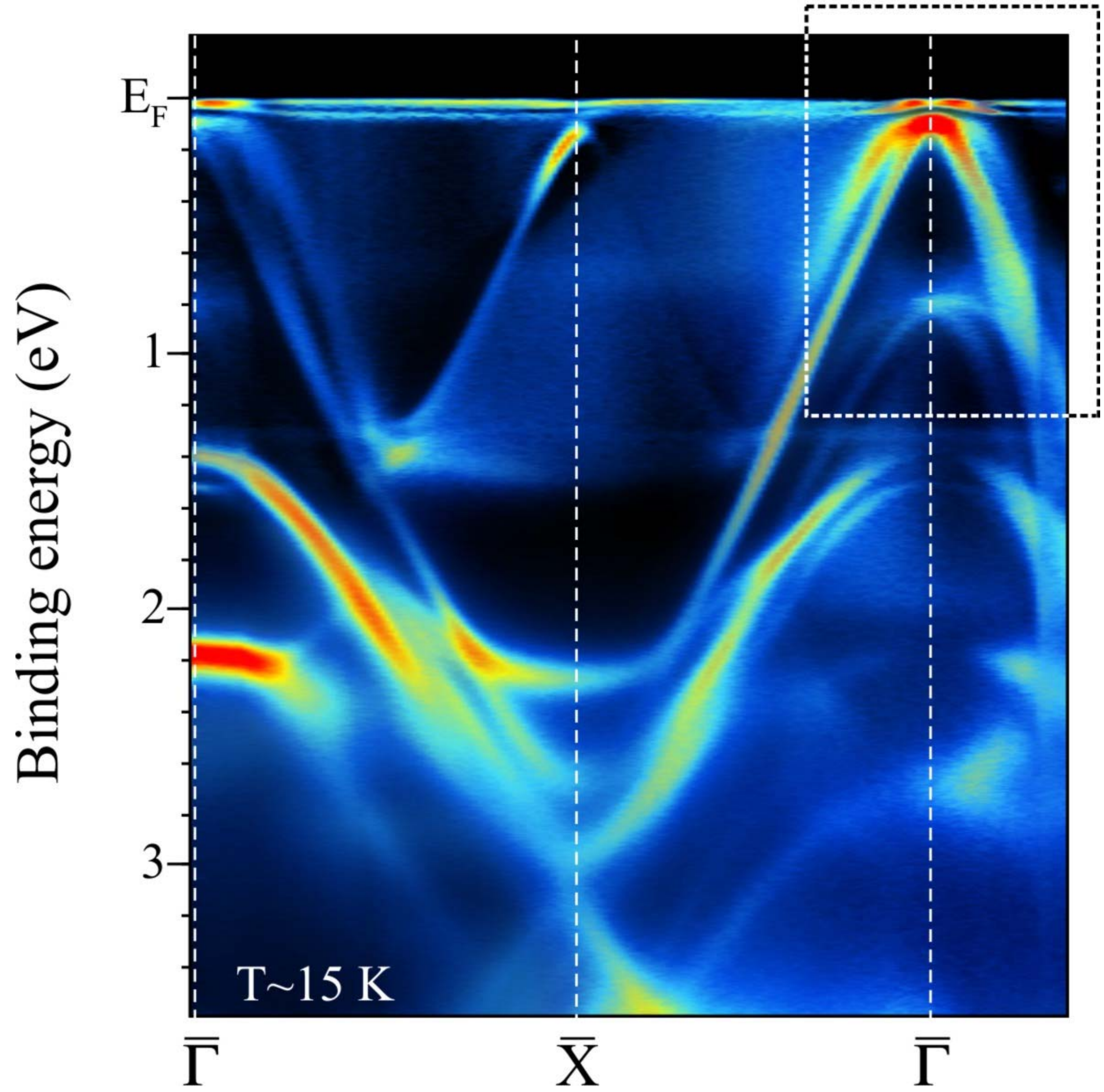
Γ_{t6} (m_j = ±1/2, ±7/2),

Γ_{t7} (m_j = ±3/2, ±5/2)



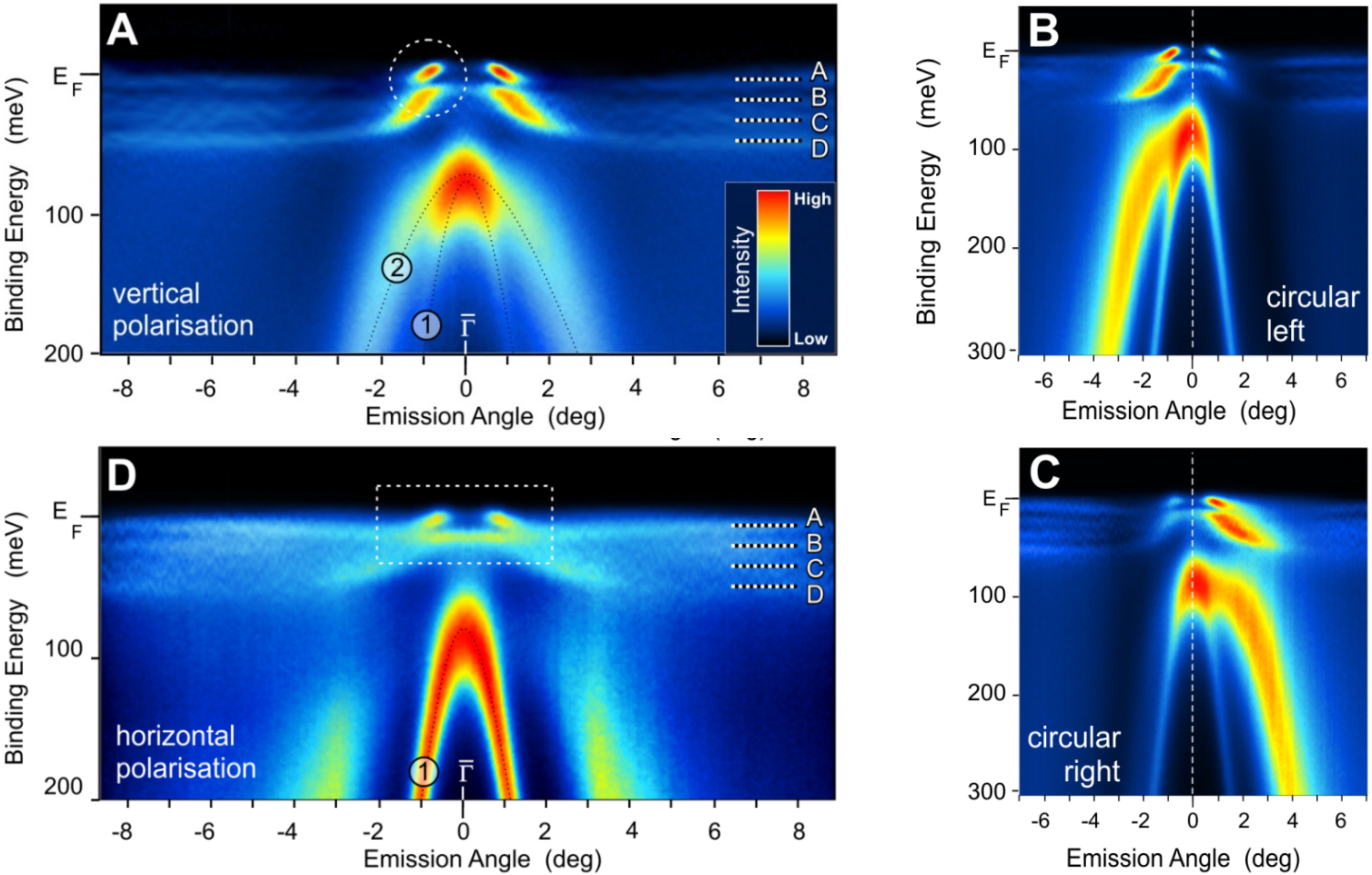
These states are seen in ARPES as 4 non-dispersing bands (where they are not perturbed by valence bands)

ARPES experiment



CEF resolved hybridization

D.V.Vyalikh, *et al*. Phys.Rev.Lett. **105**, 237601 (2010)



Hybridization model

Rh 4*d* states create at Yb site the combinations with *f* character

Non-zero hybridization parameters Δ
only for states with the same symmetry on the Yb site

$$
\left[\begin{array}{ccc|cccc}
E_1(k) & 0 & 0 & \Delta_7^{11} & \Delta_7^{12} & \Delta_6^{11} & \Delta_6^{12} \\
0 & E_2(k) & 0 & \Delta_7^{21} & \Delta_7^{22} & \Delta_6^{21} & \Delta_6^{22} \\
0 & 0 & E_3(k) & \Delta_7^{31} & \Delta_7^{32} & \Delta_6^{31} & \Delta_6^{32} \\
\hline
\Delta_7^{11} & \Delta_7^{21} & \Delta_7^{31} & \varepsilon(\Gamma_{t7}^{(1)}) & S_7 & 0 & 0 \\
\Delta_7^{12} & \Delta_7^{22} & \Delta_7^{32} & S_7 & \varepsilon(\Gamma_{t7}^{(2)}) & 0 & 0 \\
\Delta_6^{11} & \Delta_6^{21} & \Delta_6^{31} & 0 & 0 & \varepsilon(\Gamma_{t6}^{(1)}) & S_6 \\
\Delta_6^{12} & \Delta_6^{22} & \Delta_6^{32} & 0 & 0 & S_6 & \varepsilon(\Gamma_{t6}^{(2)})
\end{array}\right]
$$

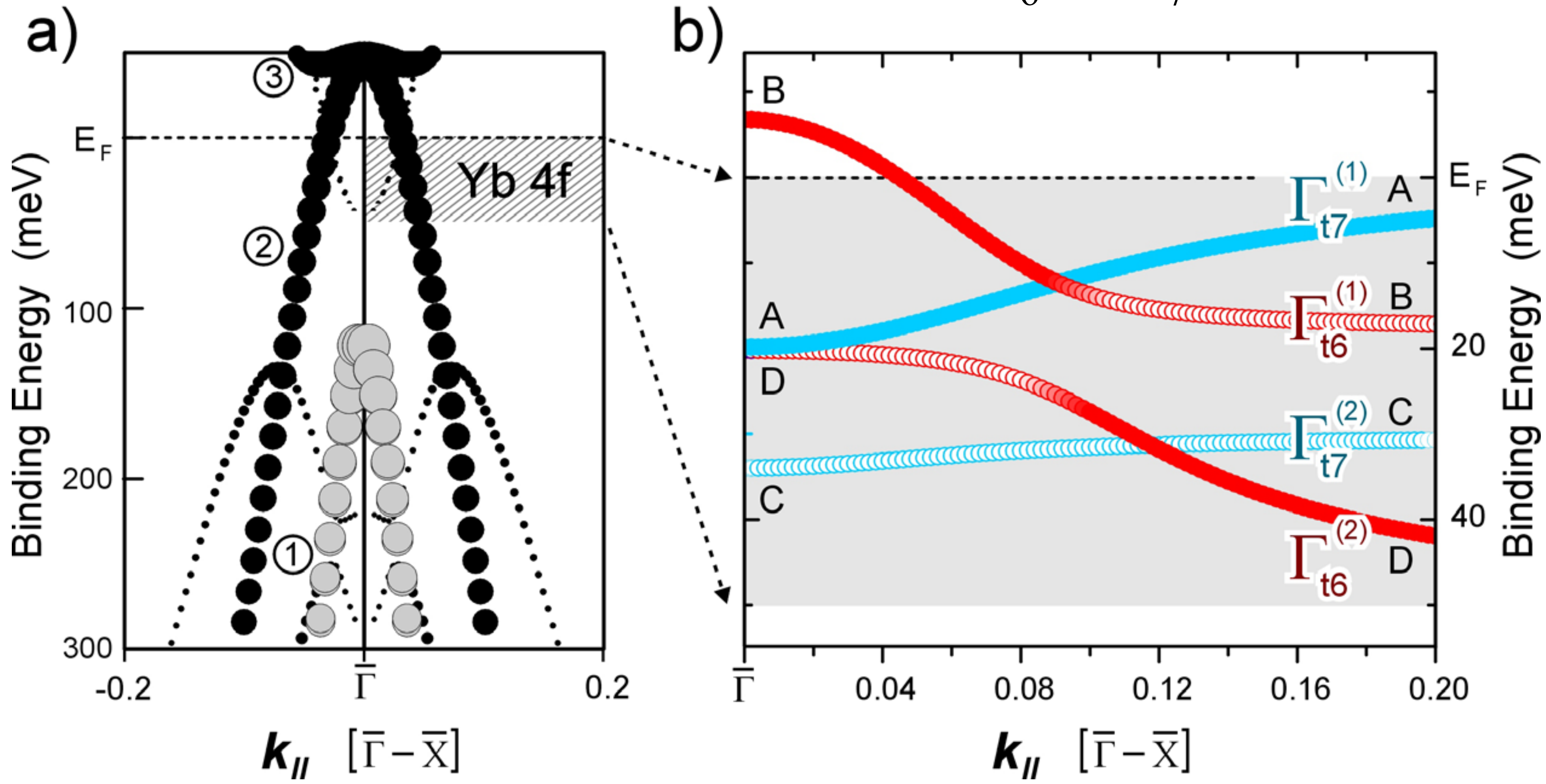
Computer simulation

Slab calculations for Si-terminated surface:

Subsurface Rh 4*d* bands at Γ point

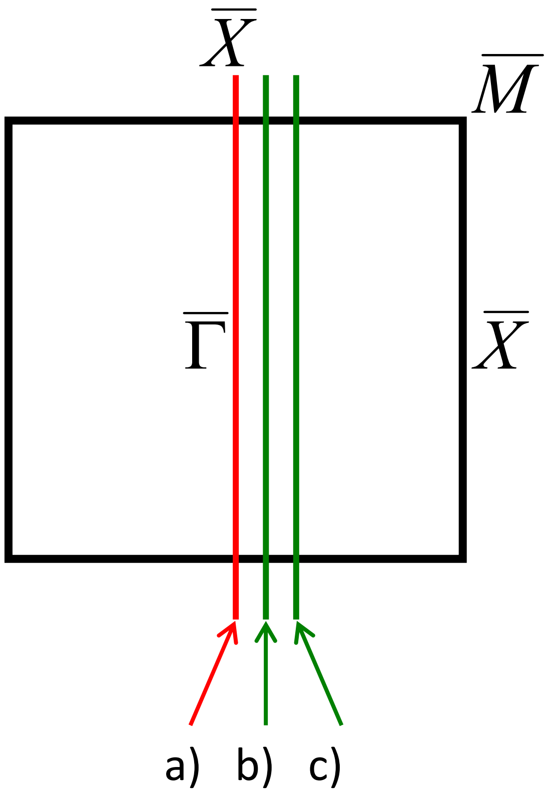
$$\Delta_6^{22} = \Delta_7^{31} = 40 meV$$

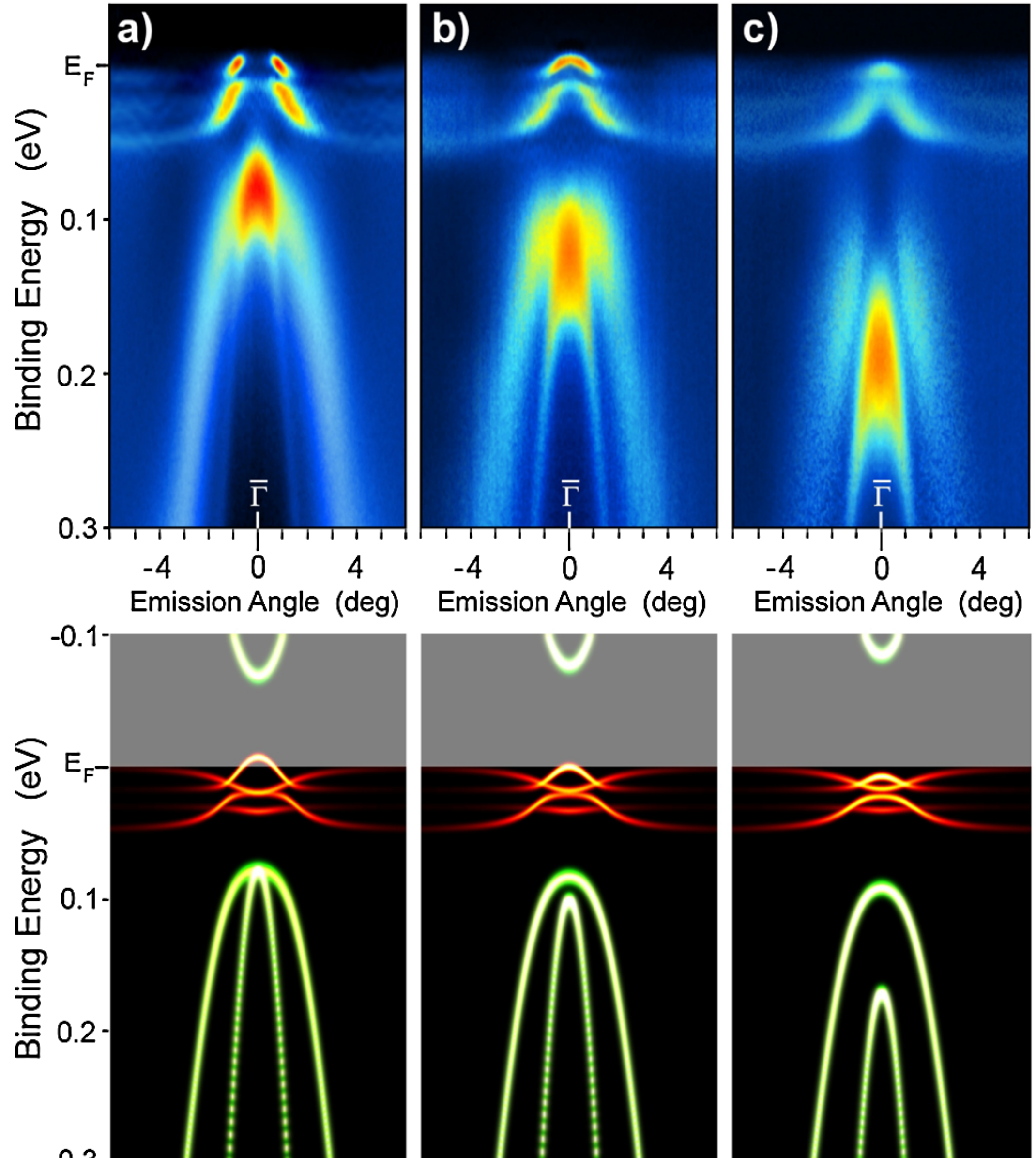
$$S_6 = S_7 = 8 meV$$



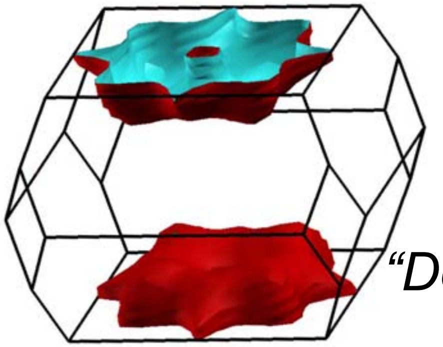
Comparison with experiment

Surface BZ:

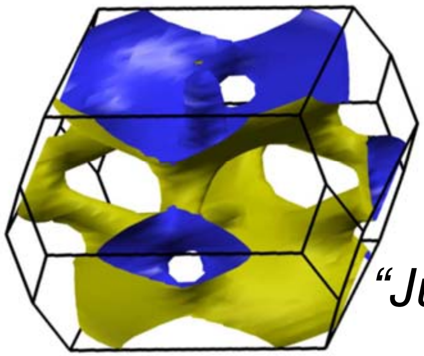




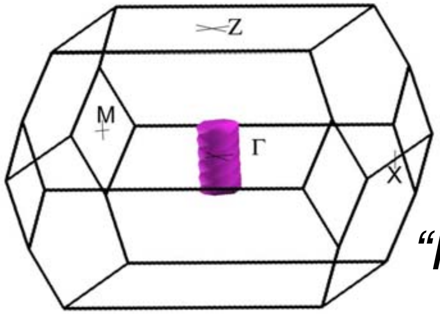
Fermi surface from LDA



"Donut"



"Jungle-gym"



"Pillbox"

Band calculations with

1) LDA+*U* approach (Yb 4*f* are shifted downwards from the Fermi level),
2) Treating Yb 4*f* states as quasi-core (no hybridization with the valence states),
3) Yb atoms substituted by Lu (4*f* states are filled and lie far from the Fermi level)

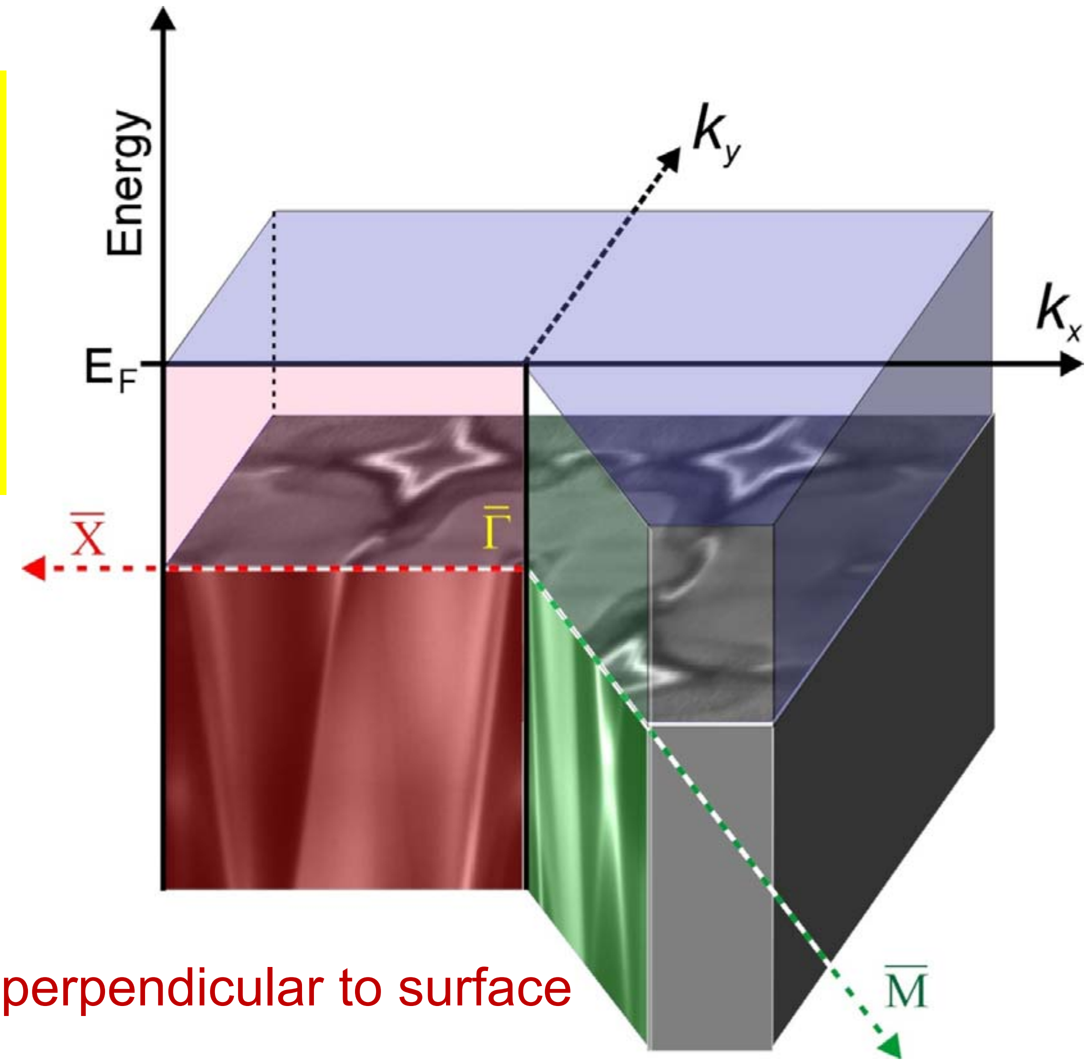
lead to the similar results for the Fermi surface.

"Donut" is the most interesting
- seems to be very sensitive to the hybridization with 4f states;
- can be well observed in ARPES.

ARPES: Problems to be overcome

- Small probing depth
+ surface states

- k_z not well defined
$\rightarrow$ some projection along k_z



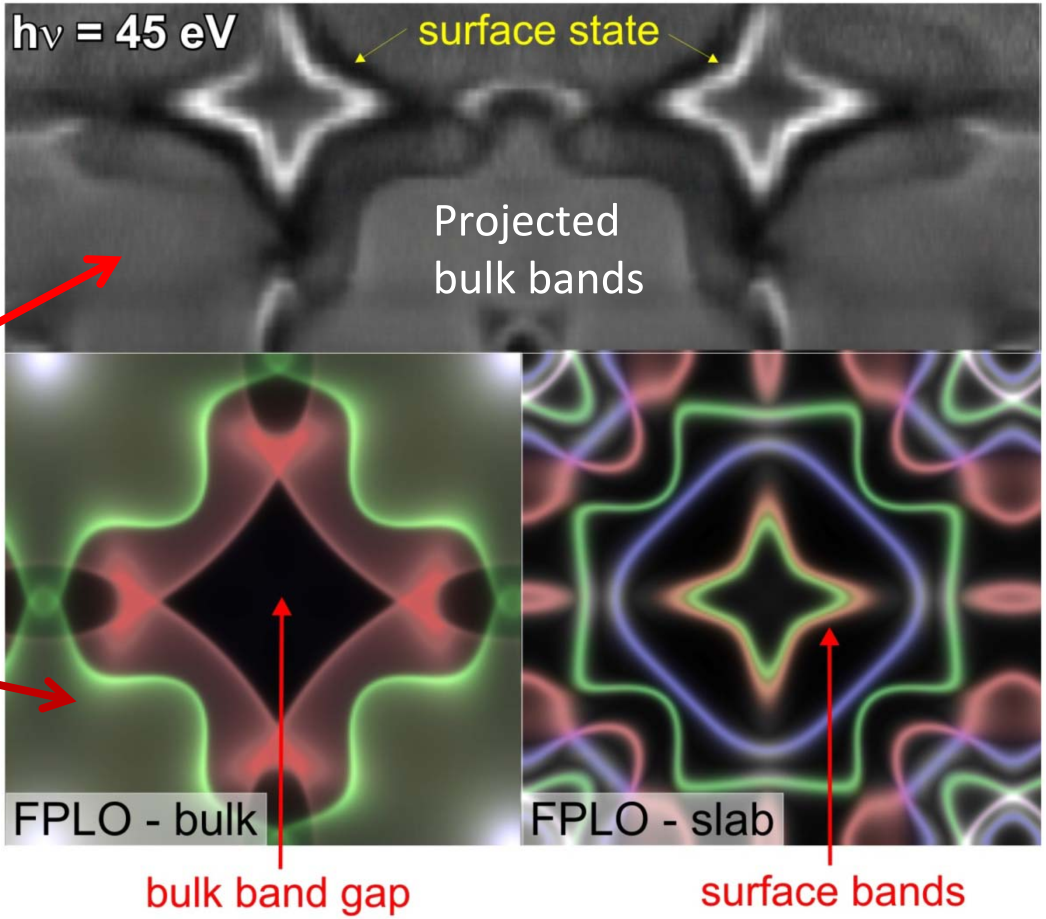
Dispersion along directions perpendicular to surface

Discrimination of surface bands

• No contribution of *f* states expected at E = 300 meV
→ Test of ARPES by means of LDA

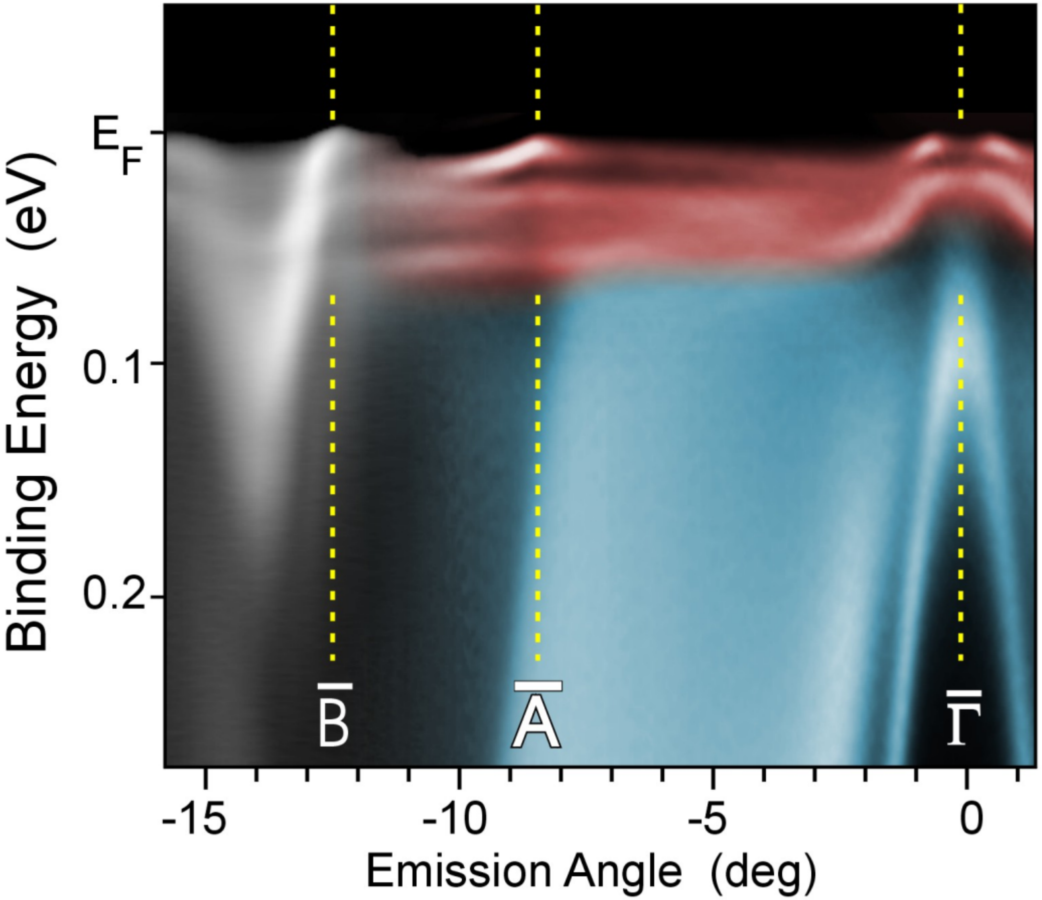
Experimental ARPES results

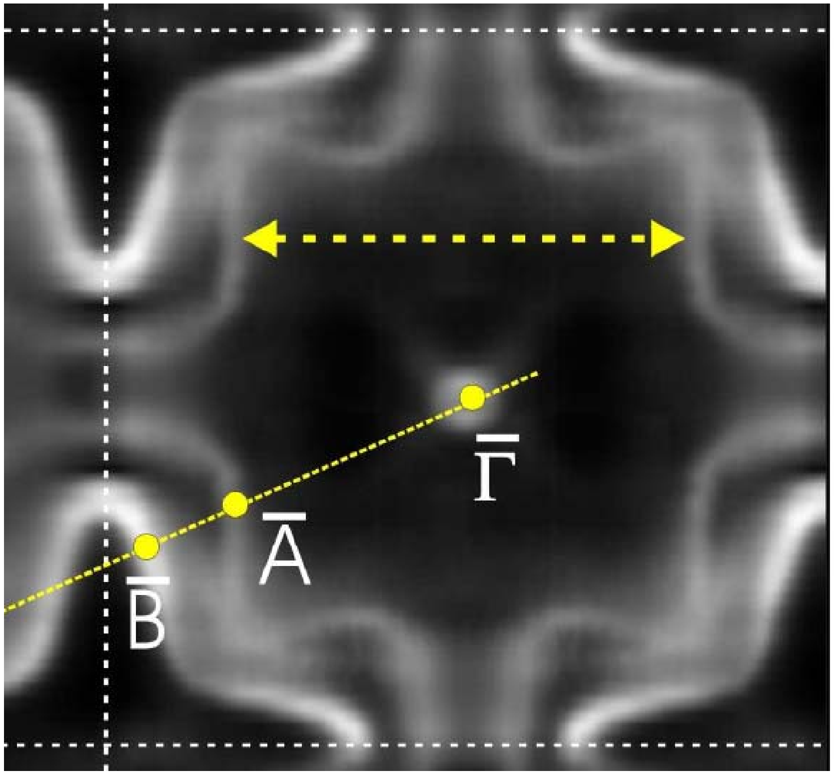
LDA calculations (4*f* in core):
- Projected bulk bands
- Slab geometry



- Bright stars around M: only seen in the slab calculations → surface bands
- Shaded area with a sharp boundary reproduced by the bulk calculations
  → projected bulk bands

Projected bands seen in the ARPES cut



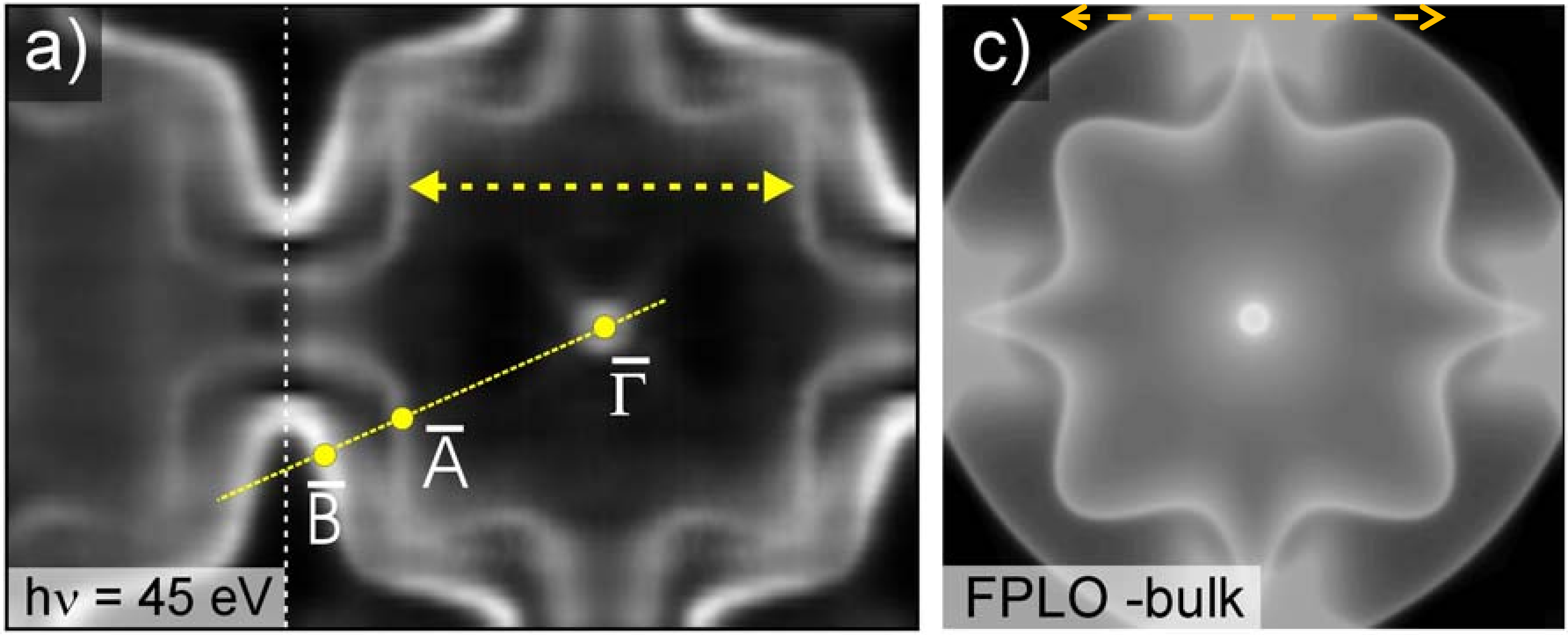


Projected bands that create the “Donut” show a sharp boundary at the A point corresponding to the edge of the “Donut” at the top of the bulk BZ

ARPES view of $YbRh_2Si_2$ Fermi surface

S.Danzenbächer, *et al*. Phys.Rev.Lett. **107**, 267601 (2011)

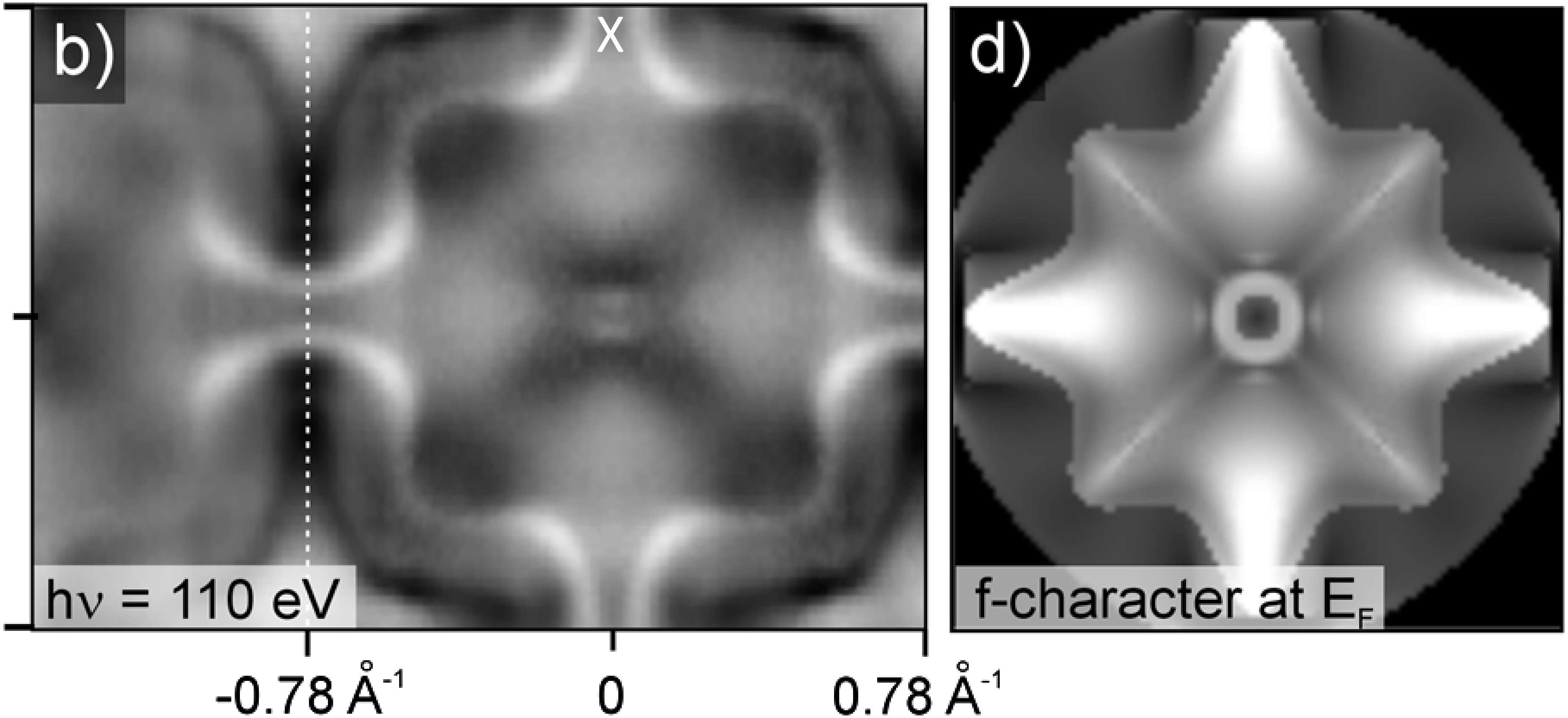
- At T ≅ 10 K, below T_K ≅ 25 K
- ARPES : hν = 45 eV → strong Rh 4*d* contributions → valence states



- ARPES : Necks near X in contradiction to LDA calculations (sharp edges)

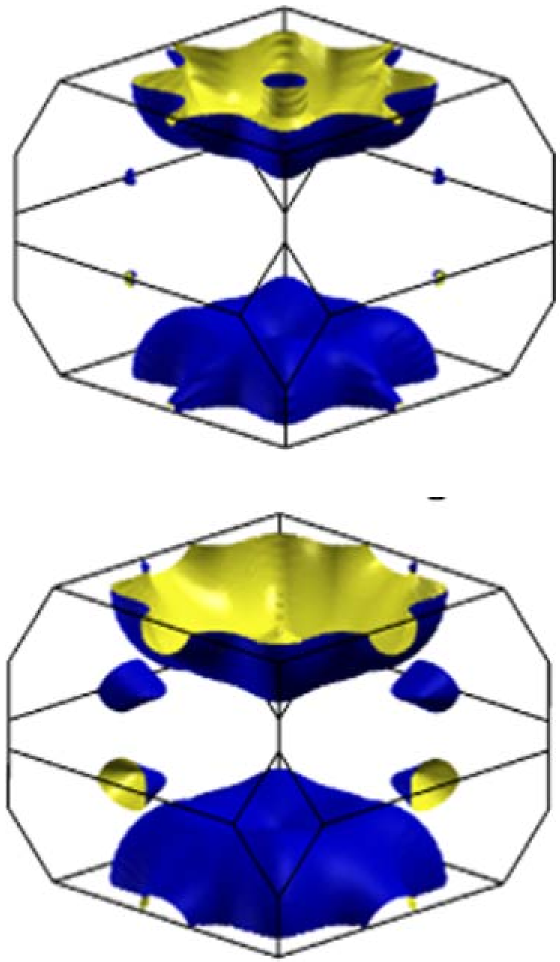
ARPES view of $YbRh_2Si_2$ Fermi surface

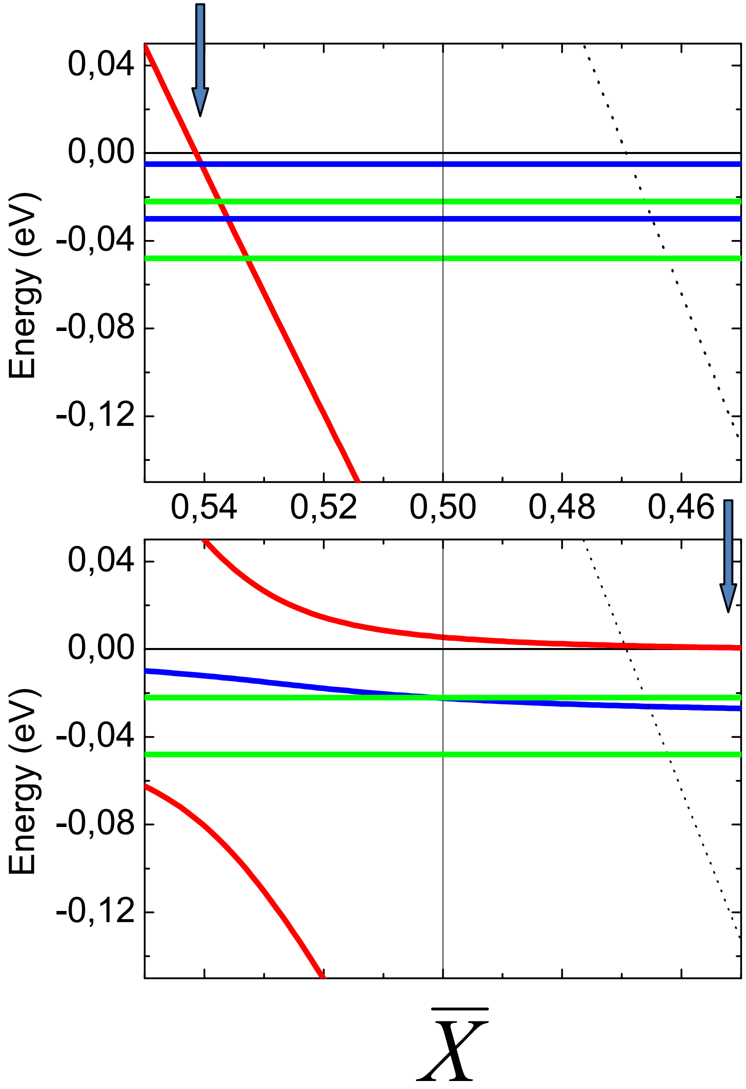
- ARPES: hν = 110 eV
  → strong 4*f* cross-sections
- LDA calculations:
  → *f*-character of valence states at Yb-site



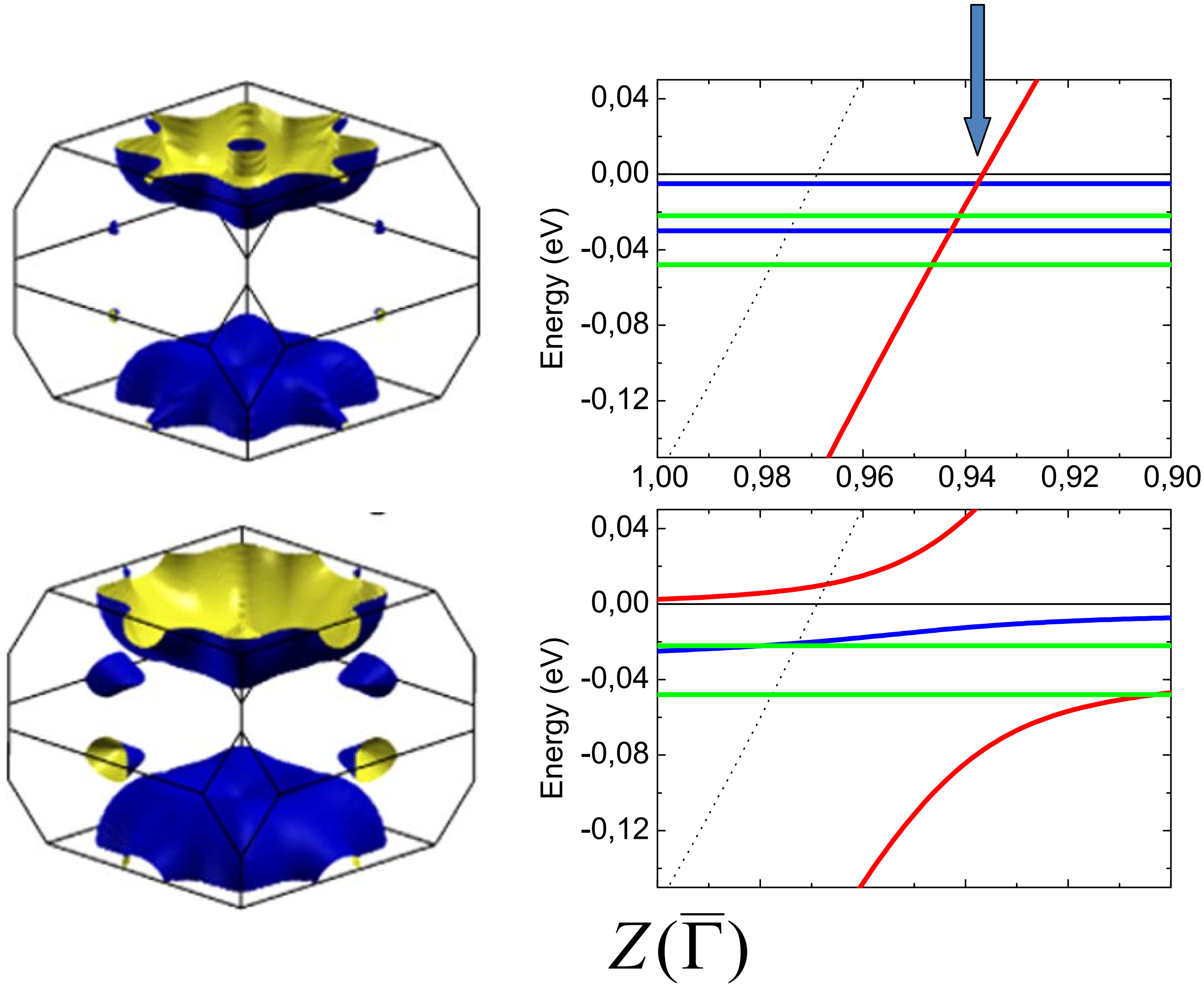
- At Necks: strong 4*f* contributions → Heavy 4*f* hybrid bands dominate Fermi surface

Hybridization effects: Opening the necks





Hybridization effects: Closing the hole at Z



Summary: ARPES on $YbRh_2Si_2$

• High-quality single crystals, excellent cleave, high-resolution spectrometer $\rightarrow$ Nice experimental data

$\Rightarrow$ Information on the Fermi surface of the quantum critical heavy-fermion system $YbRh_2Si_2$

• One sheet of Fermi surface ("Donut") can be nicely observed

• Computer simulation (LDA + hybridization model) agree well with the experimental results as well as with the model of renormalized band structure (*G. Zwicknagl; J. Phys.: Cond. Matt. 2011*)

• Electronic states on the Fermi surface have significant contribution of Yb 4*f* states (of Γ_{t7} symmetry)

• 4*f* states of Γ_{t6} symmetry appear at E_F only at some points of the BZ close to the sample surface

$\Rightarrow$ Next steps:

• Study of the Fermi surface evolution upon chemical doping (over QCP) and temperature variations (over T_K)

Linear bands in $EuRh_2Si_2$

M.Höppner, *et al*. Nature Commun. 4:1646 doi: 10.1038/ncomms2654 (2013)