Correlated electrons and new temperature scales at the surfaces of 4f materials

by Denis Vyalikh

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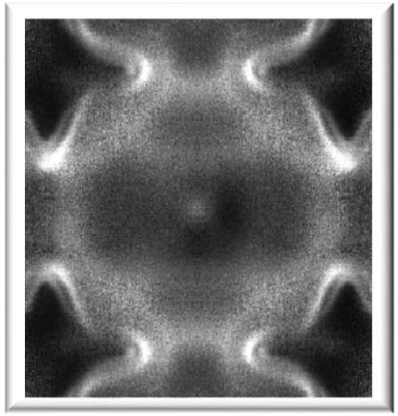
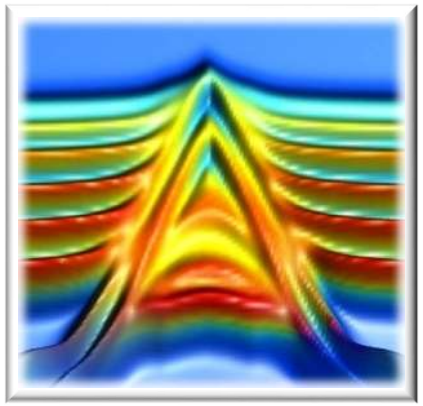
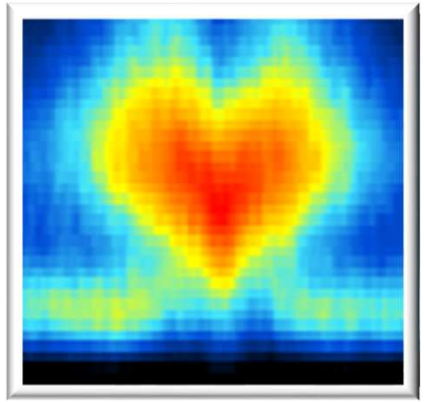
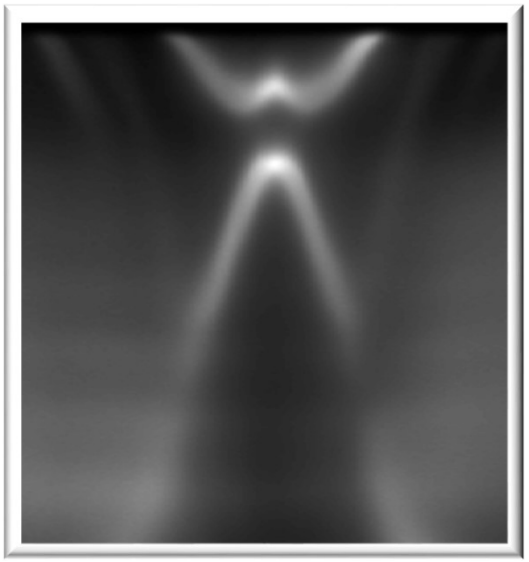
San Sebastian, Spain

*"**Unveiling** the **diversity** and **beauty** of **4f physics** with **photoemission** at the **surface** and in the **bulk**..."*

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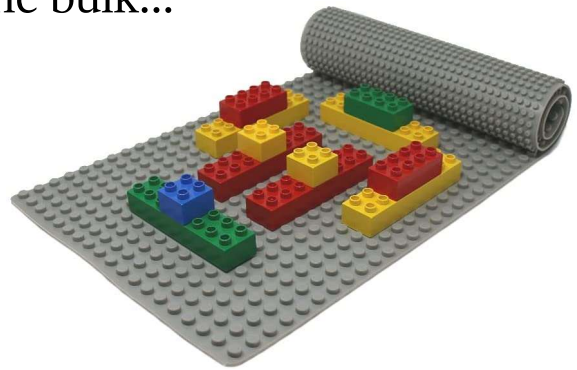
Extraordinary properties of 4f materials

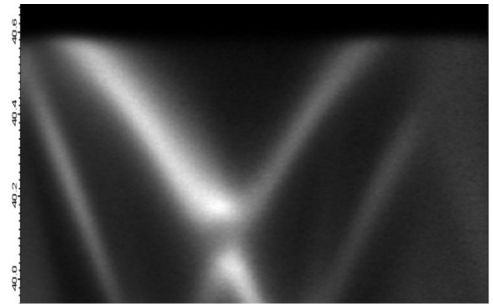
...in the **bulk** including complex magnetic phases, unconventional superconductivity, heavy-fermion properties, Kondo physics, quantum criticality, valence fluctuations, and others...

Lanthanide Series	57 La Lanthanum 138.9055	58 Ce Cerium 140.115	59 Pr Praseodymium 140.90765	60 Nd Neodymium 144.24	61 Pm Promethium 144.9127	62 Sm Samarium 150.36	63 Eu Europium 151.9655	64 Gd Gadolinium 157.25	65 Tb Terbium 158.92534	66 Dy Dysprosium 162.50	67 Ho Holmium 164.93032	68 Er Erbium 167.26	69 Tm Thulium 168.93421	70 Yb Ytterbium 173.04	71 Lu Lutetium 174.967

...reasonable to anticipate that the **4f**-driven **physics** at the **surface** can be even much **richer** and more **compelling** than in the bulk.

Lack of inversion symmetry and spin-orbit coupling (SOC), appearance of surface-electron states and resonances, relaxation and reconstruction, as well as strong changes of the crystal-electric field near and at the surface are the driving forces for novel 4f-driven phenomena, phases and temperature scales that are in remarkable difference to those in the bulk...



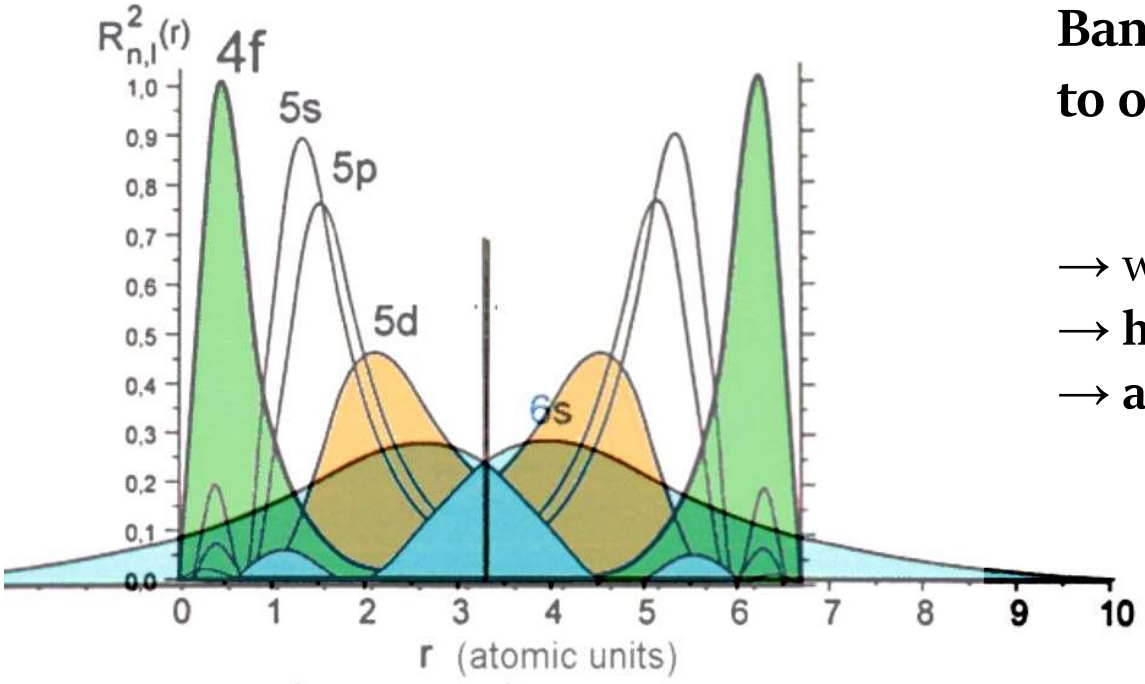


Extraordinary properties of 4f materials

...in the **bulk** including complex magnetic phases, unconventional superconductivity, heavy-fermion properties, Kondo physics, quantum criticality, valence fluctuations, and others...

Lanthanide Series	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 138.9055	Cerium 140.115	Praseodymium 140.90765	Neodymium 144.24	Promethium 144.9127	Samarium 150.36	Europium 151.9655	Gadolinium 157.25	Terbium 158.92534	Dysprosium 162.50	Holmium 164.93032	Erbium 167.26	Thulium 168.93421	Ytterbium 173.04	Lutetium 174.967

* filling of the 4*f* shell: [Xe] $4f^{n}$ $(5d6s)^{m}$



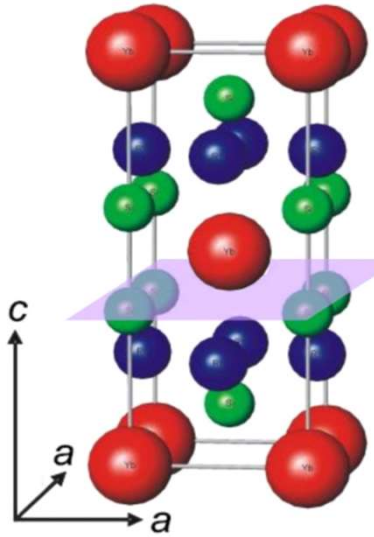
Band-width proportional to overlapp of orbitals:

→ width of ***4f*-bands** ~ **10^{th} of meV;**
→ **highly localized;**
→ **atomic-like magnetic moments.**

RT_2Si_2 compounds (R = rare-earth, U; T = Co, Rh, Ir):

Playground for studying electron correlation phenomena

Tetragonal body-centered $ThCr_2Si_2$ structure



Kondo lattices

$YbRh_2Si_2$, $YbIr_2Si_2$, $CeRh_2Si_2$,…

Unconventional superconductivity

$CeCu_2Si_2$, URu_2Si_2,…

Mixed-valent behaviour

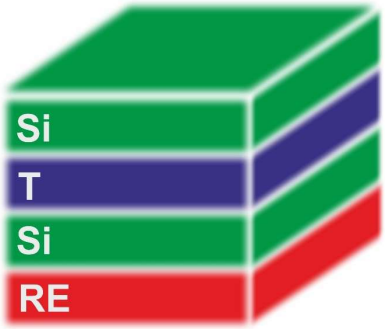
$SmRh_2Si_2$, $EuIr_2Si_2$, $EuNi_2P_2$ …

Different types of magnetic order

$HoRh_2Si_2$, $EuRh_2Si_2$, $GdRh_2Si_2$, …

Quantum criticality

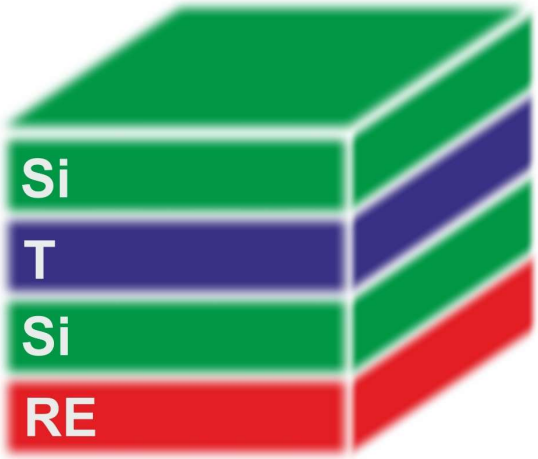
($YbRh_2Si_2$ phase transitions at T=0 K)

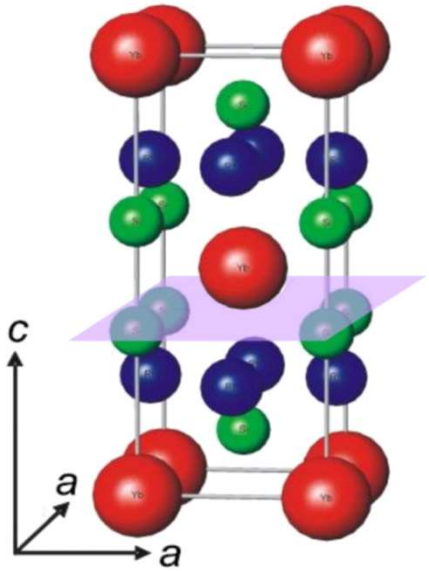


Electronic properties and magnetic phenomena in magnetically active *4f*-based nanostructural objects. A kind offer from RET_2Si_2

- The silicide **Si-T-Si-RE** surface allows to **design** a system for different scenarios **combining** fundamental interactions like **spin-orbit**, **Kondo**, **crystal-electric field**, and **exchange magnetic** interactions and to study the diversity of physics at reduced dimensionality;
- It becomes useful for systematic studies of the **interplay** between fundamental ***f*-driven** properties and the **emergent Rashba effect**.

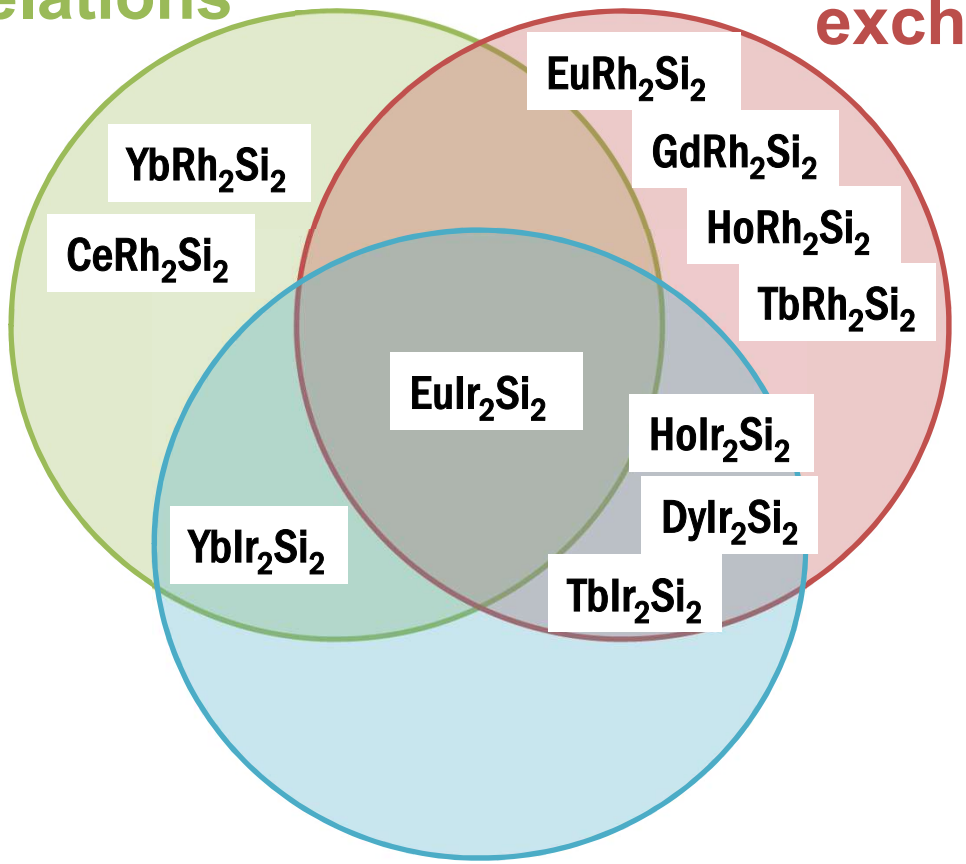
- **Magnetically**-active *f*-based **nanostructures** with **Si**, **Ge**, **P** are attractive for **novel** electronic and magnetic **applications**;



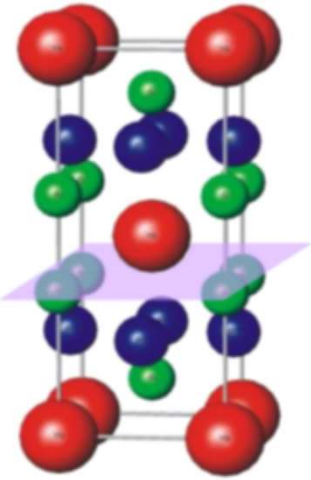


RET_2Si_2 materials for systematic studies of *f*-driven physics at the surface

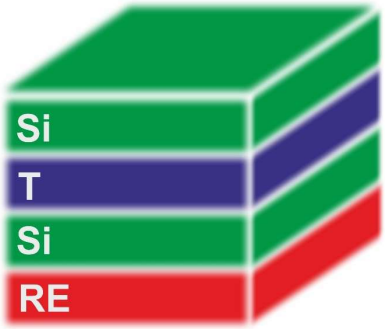
strong correlations
Ce, Yb



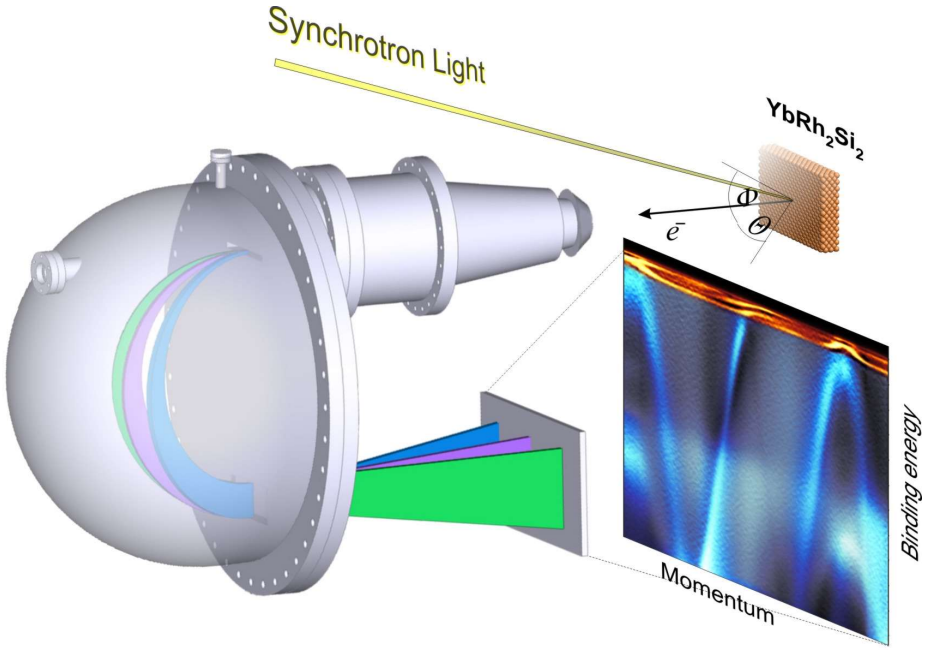
exchange magnetic interaction

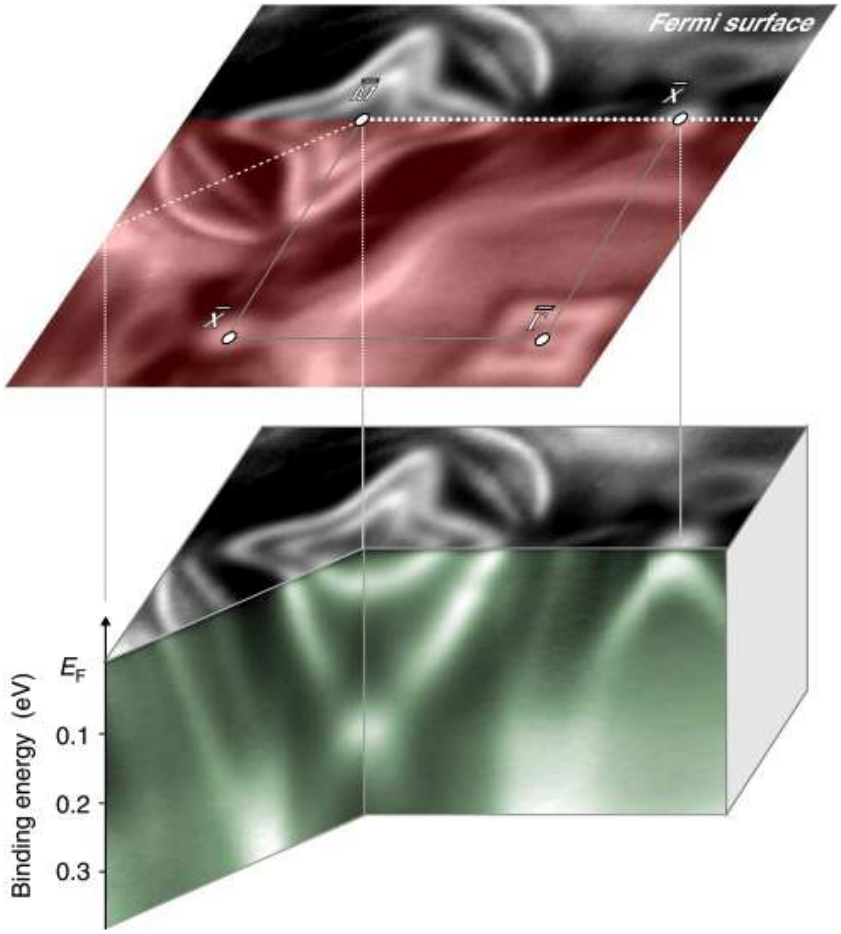


spin-orbit coupling
Co 3*d* → Rh 4*d* → Ir 5*d*



Angle-Resolved Photoelectron Spectroscopy ARPES





Energy Conservation

$$E_{kin} = h\nu - \phi - |E_B|$$

Momentum Conservation

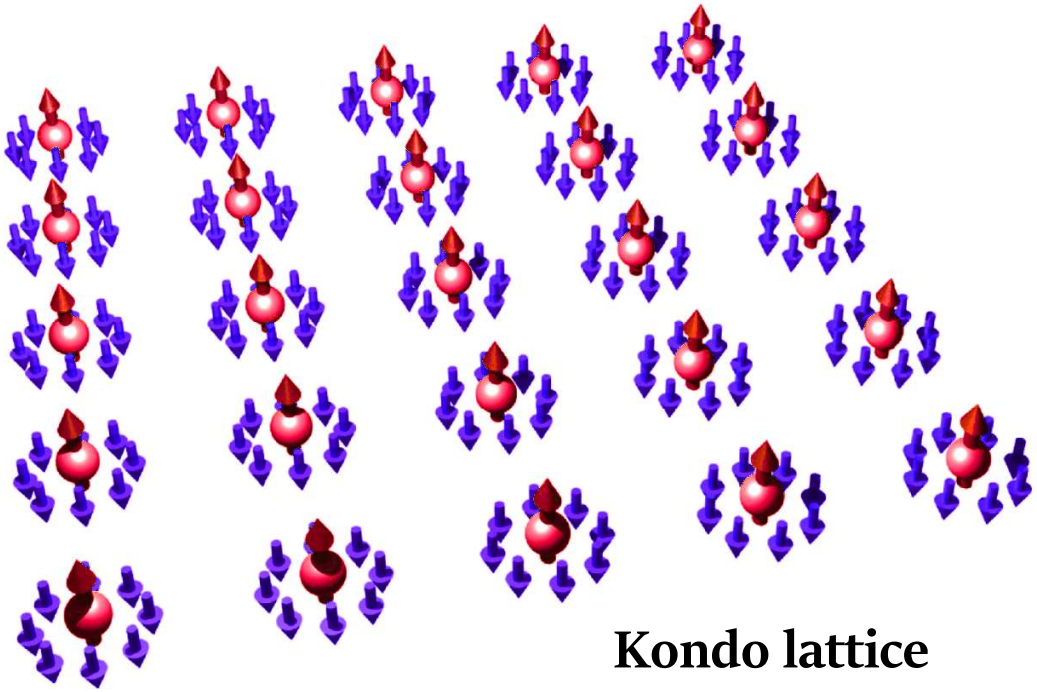
$$\mathbf{p}_{\parallel} = \hbar\mathbf{k}_{\parallel} = \sqrt{2mE_{kin}} \cdot \sin\vartheta$$

- It measures **band structure** of crystalline solids …
- It also measures **interactions** …

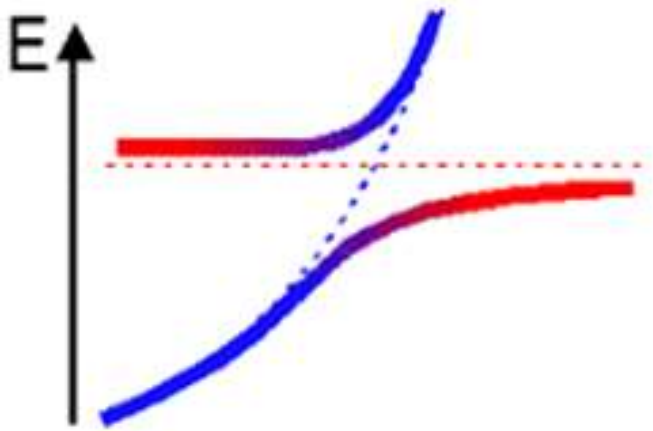
Heavy-Fermion systems: $CeRh_2Si_2$ and $YbRh_2Si_2$

- Screening of the local magnetic moments in the strong coupling limit → **Kondo effect**

- ***f-d*** hybrid formation in Kondo-lattice compounds → ***4f*** admixture to conduction band leads to **"heavy fermions"**

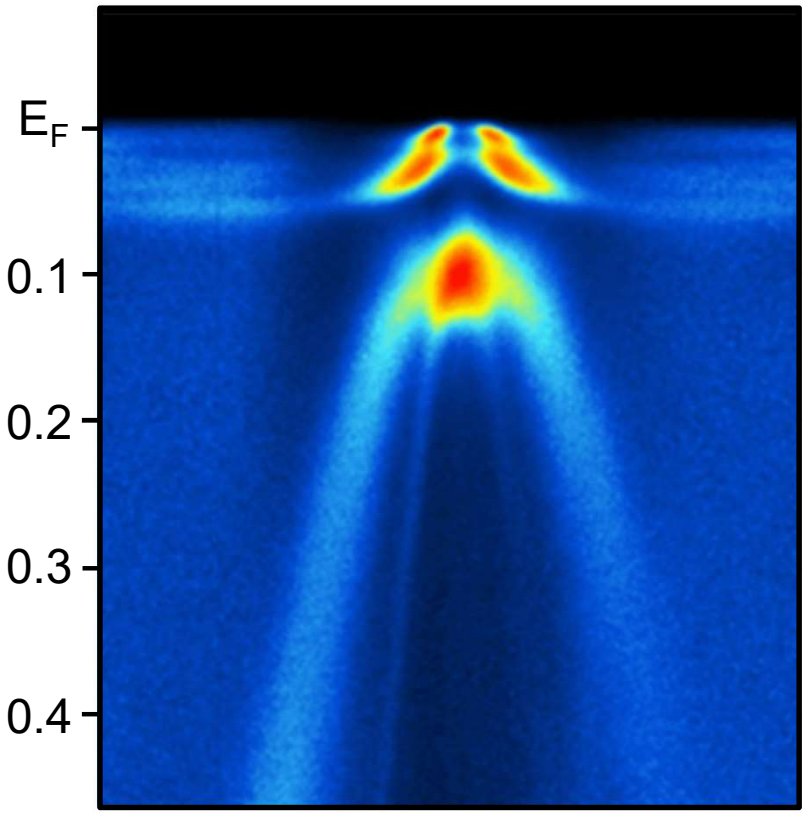


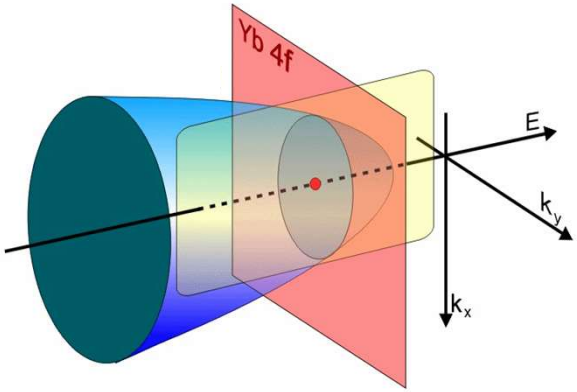
Kondo lattice

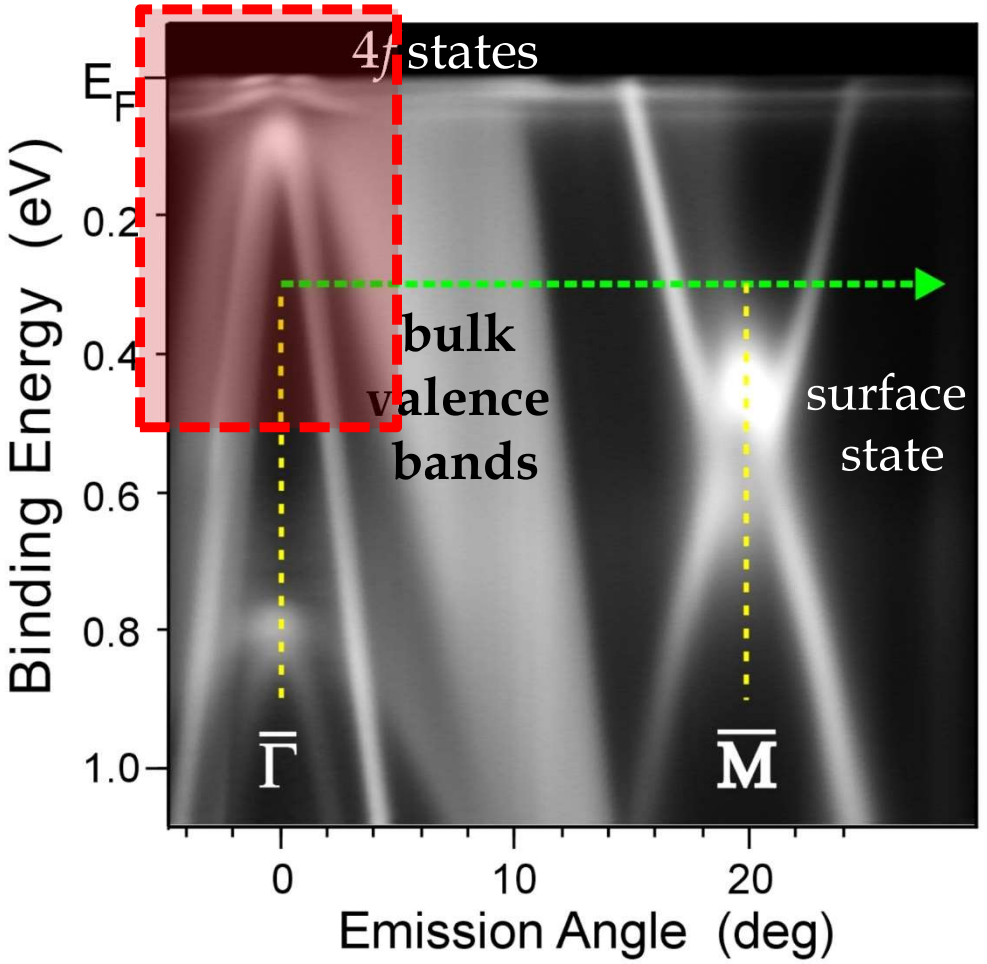


f-d hybridization leads to non-crossing behavior and ***4f***- admixture to the ***d***- band

k-dependence of the crystal-field splittings of 4f's seen in ARPES for $YbRh_2Si_2$

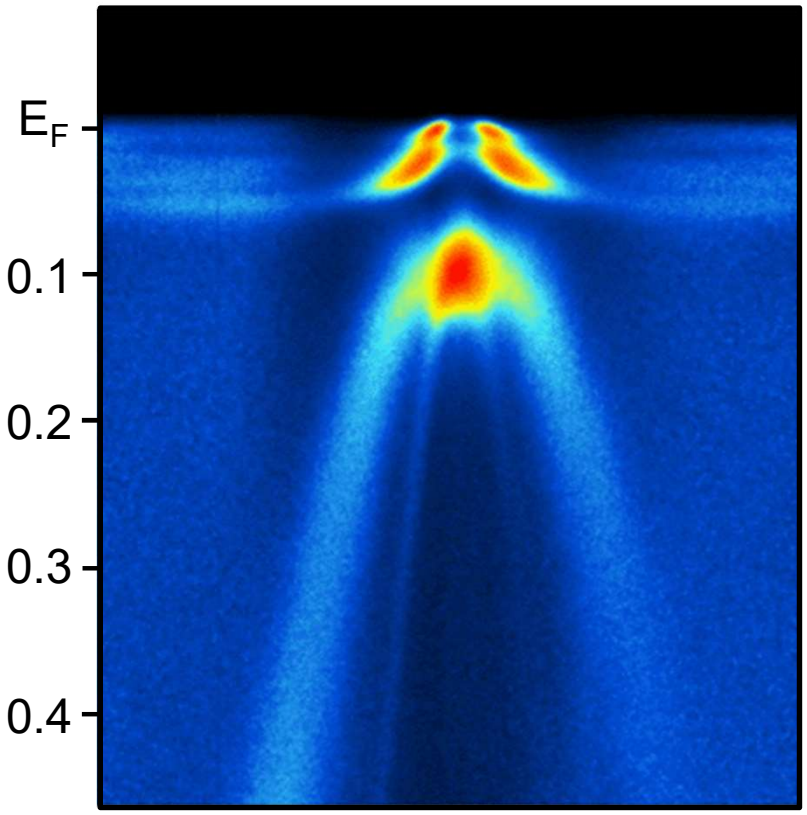


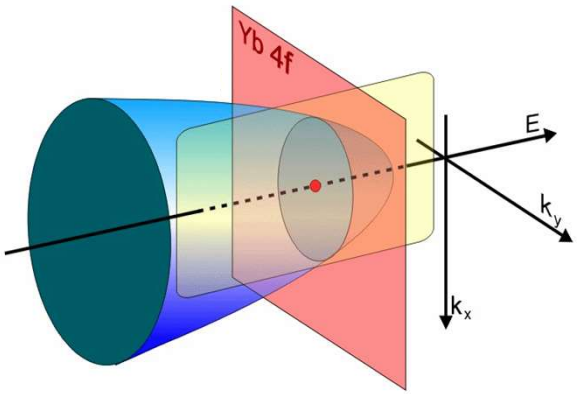




PRL (2008), PRL (2010), PRL (2011).
PRB (2018), PRB (2020), PRB (2021).
PRL (2020).

k-dependence of the crystal-field splittings of 4f's seen in ARPES for $YbRh_2Si_2$





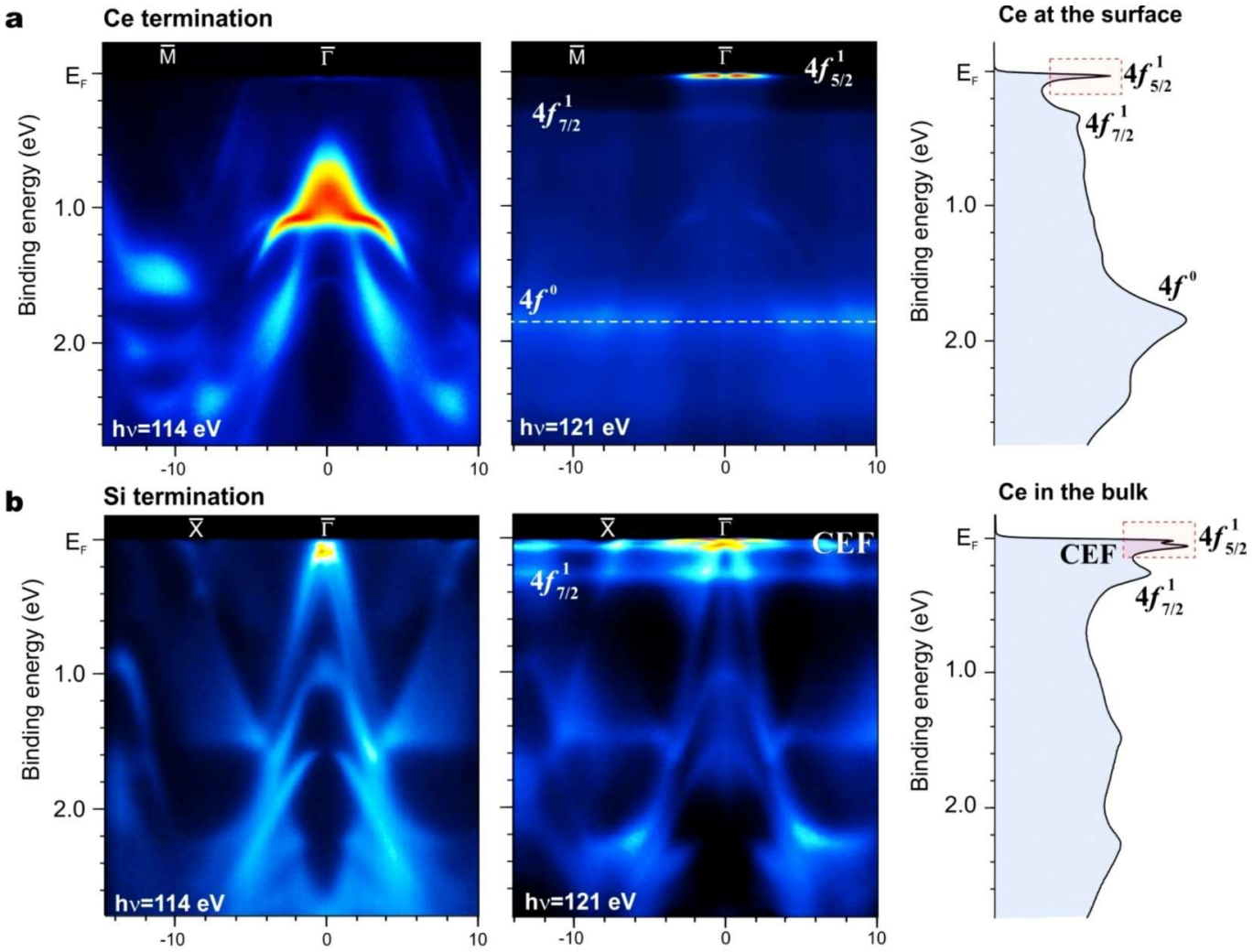
Application of ARPES allows to comprehensively explore the Kondo-lattice materials and :

- To find the hot regions in ***k***-space where *f-d* interaction occurs,
- To disclose the fine spectral pattern of this interplay,
- To investigate the CEF scheme and dispersion of the *f*-states,
- To analyze the symmetry properties of the band-like *f*-states,
- To explore the *f*-derived Fermi surface and its properties.

Heavy-fermion compound $CeRh_2Si_2$: Surface and bulk electronic structure

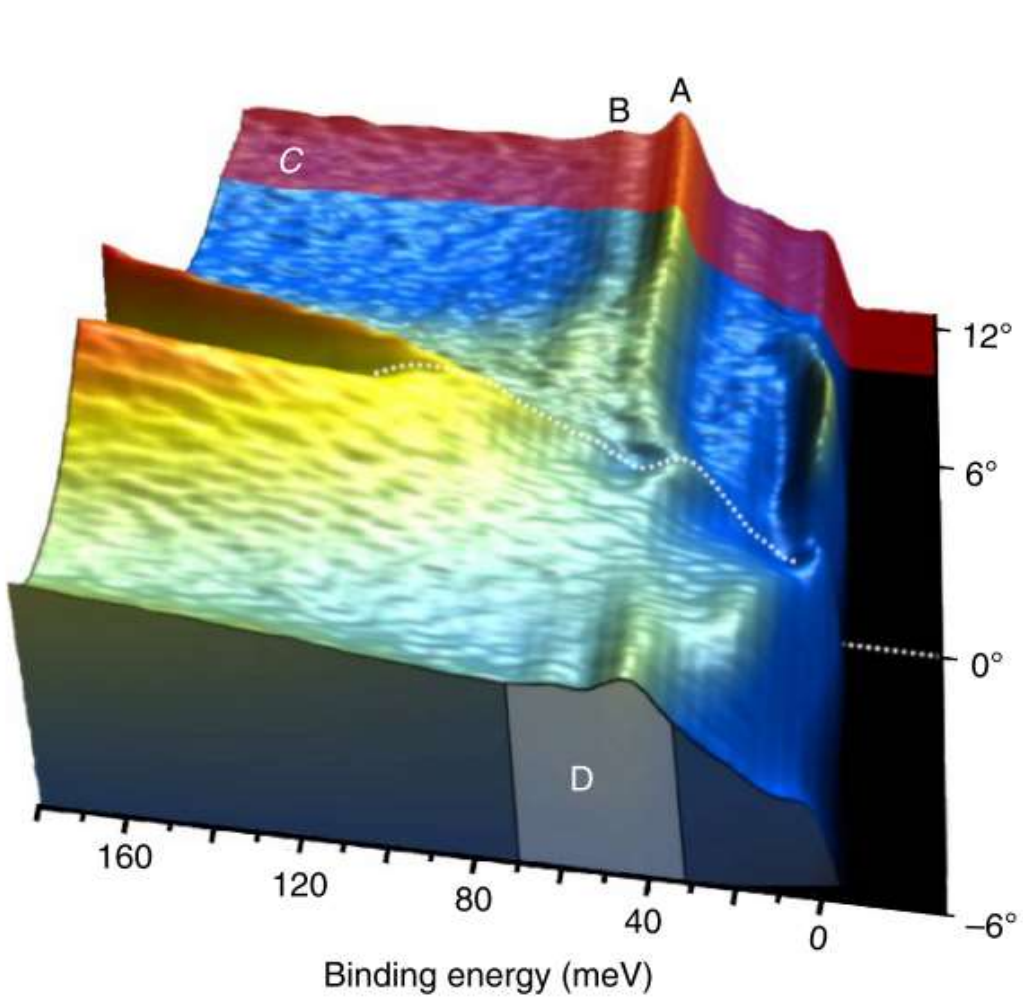
- **Ce at surface sites:**
  Weak Kondo-peak at E_F ("$4f^1$")
  strong ionization peak ("$4f^0$"
  $\Rightarrow$ **weakly hybridized**

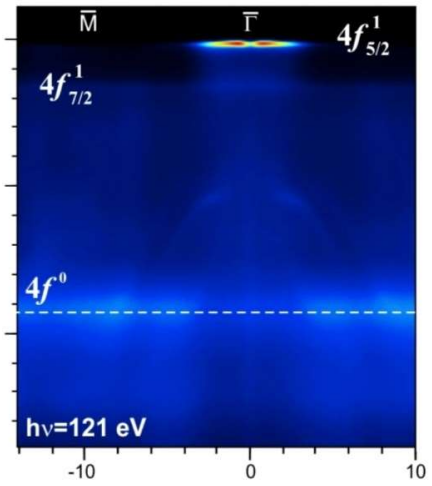
- **Ce in the bulk:**
  Strong Kondo-peak at E_F ("$4f^1$"),
  weak ionization peak ("$4f^0$")
  $\Rightarrow$ **strongly hybridized**

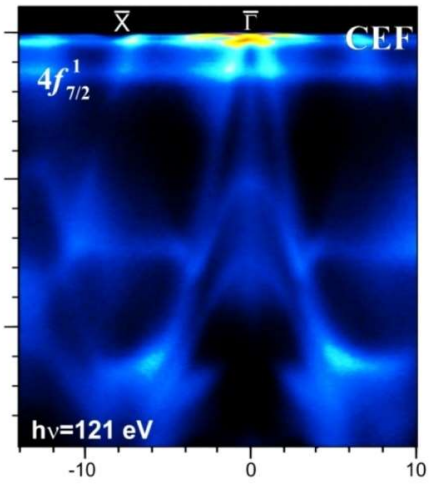


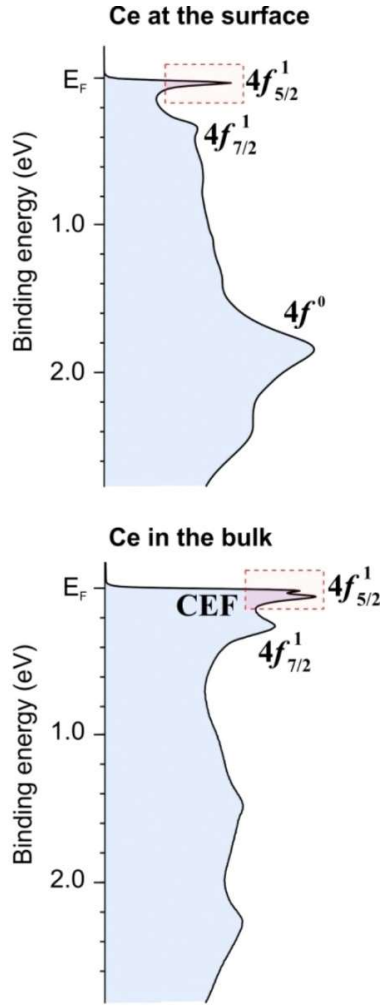
Nature Comm. **7** 11029 (2016)

Heavy-fermion compound $CeRh_2Si_2$: Surface and bulk electronic structure



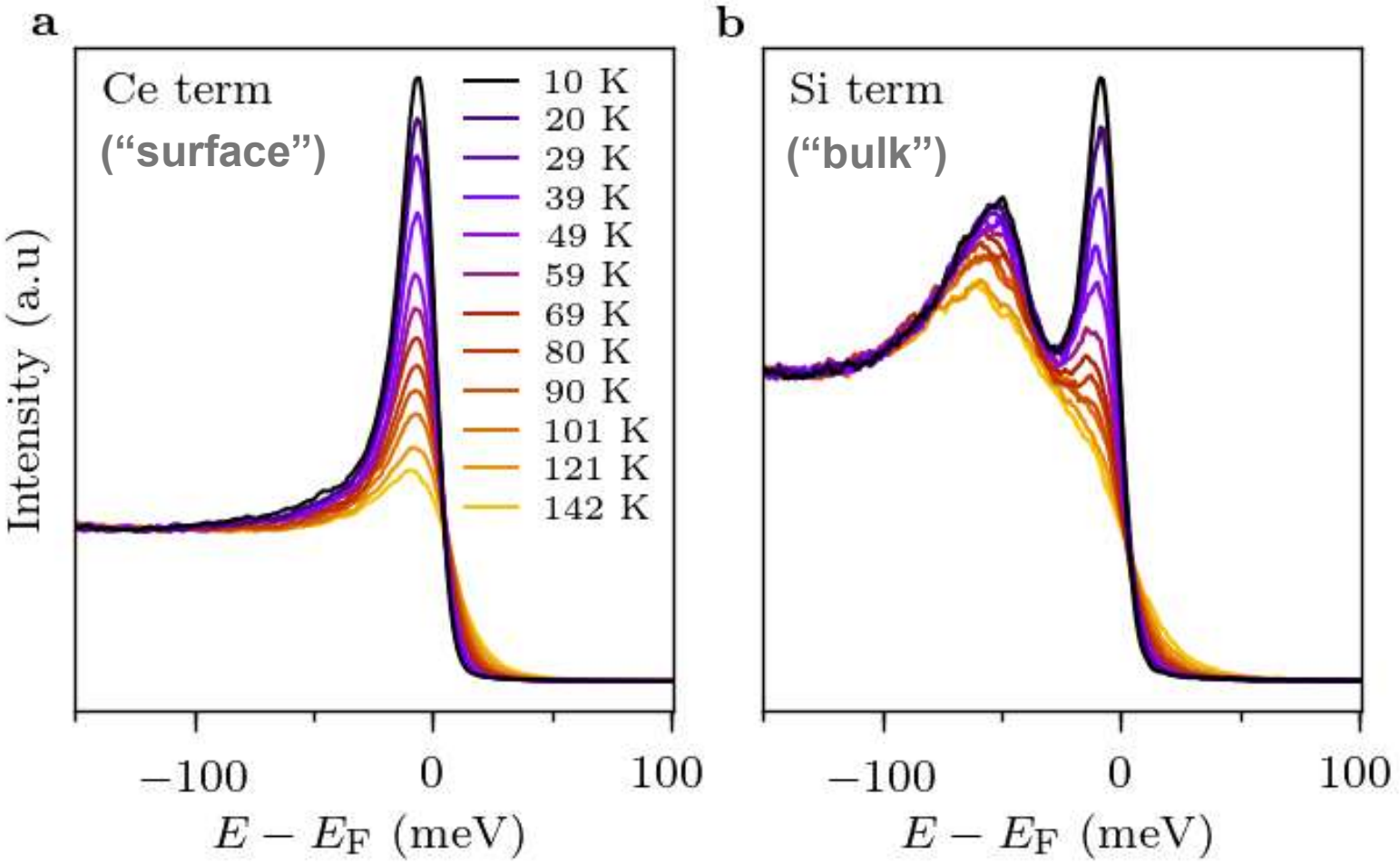






Nature Comm. **7** 11029 (2016)

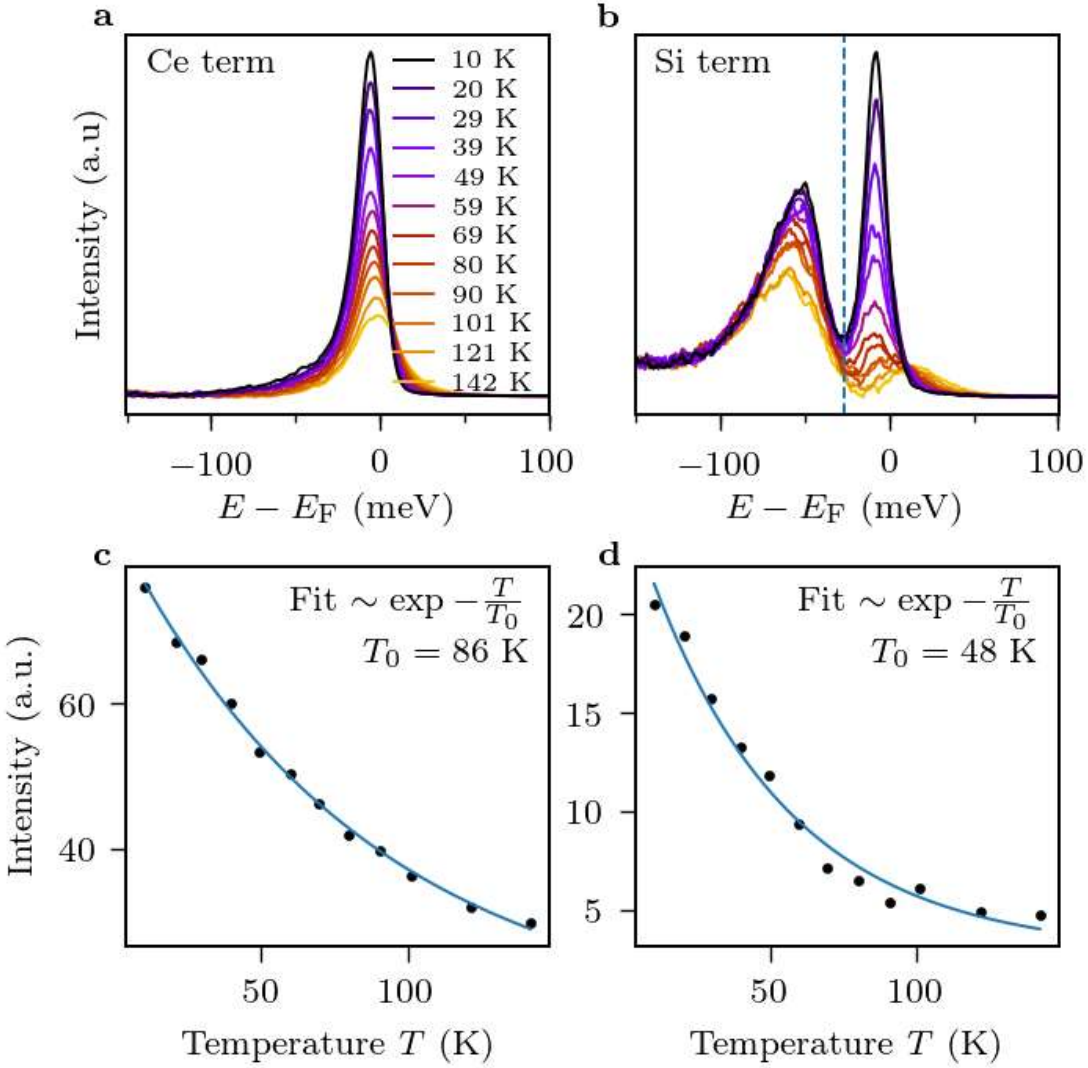
$CeRh_2Si_2$: Temperature dependence of the Kondo peak for surface and bulk



- crystal electric field (CEF) splitting of $4f^1_{5/2}$ suppressed at surface
- strong **temperature dependence** of the Kondo peak

npj Quantum Materials **5** 70 (2020).

$CeRh_2Si_2$: Temperature dependence of the Kondo peak for surface and bulk

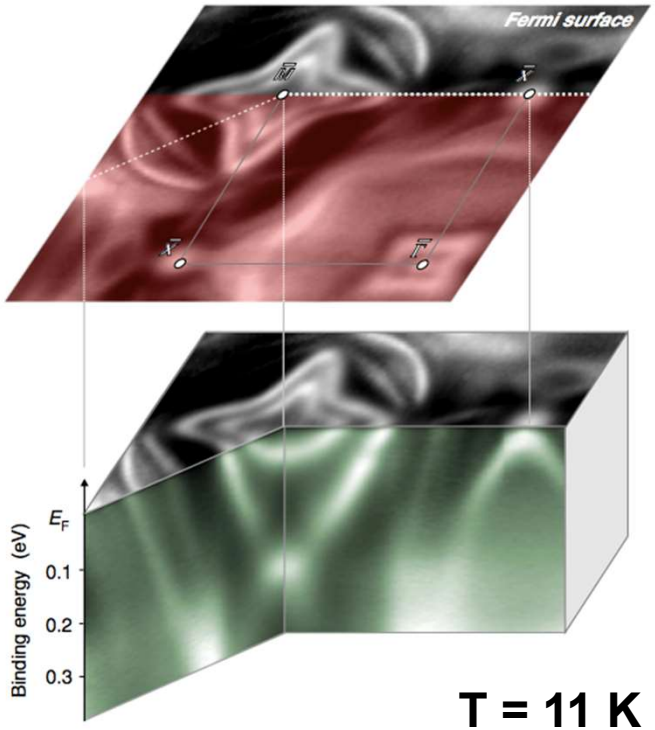


- **Stronger damping of the Kondo-peak in bulk as compared to surface**,
- **Opposite behavior expected** from hybridization strength!

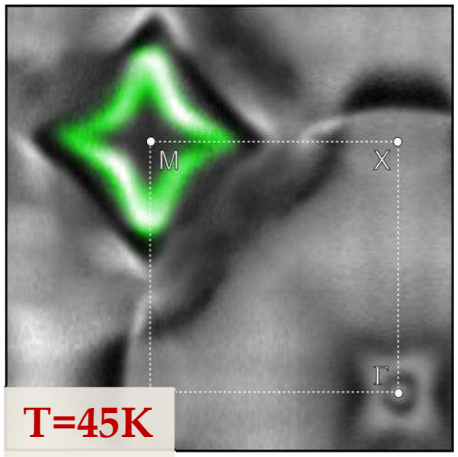
npj Quantum Materials **5** 70 (2020).

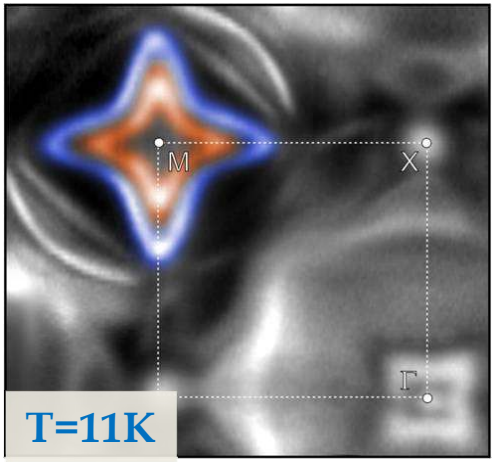
Observation of ferromagnetic silicide surface $EuRh_2Si_2$

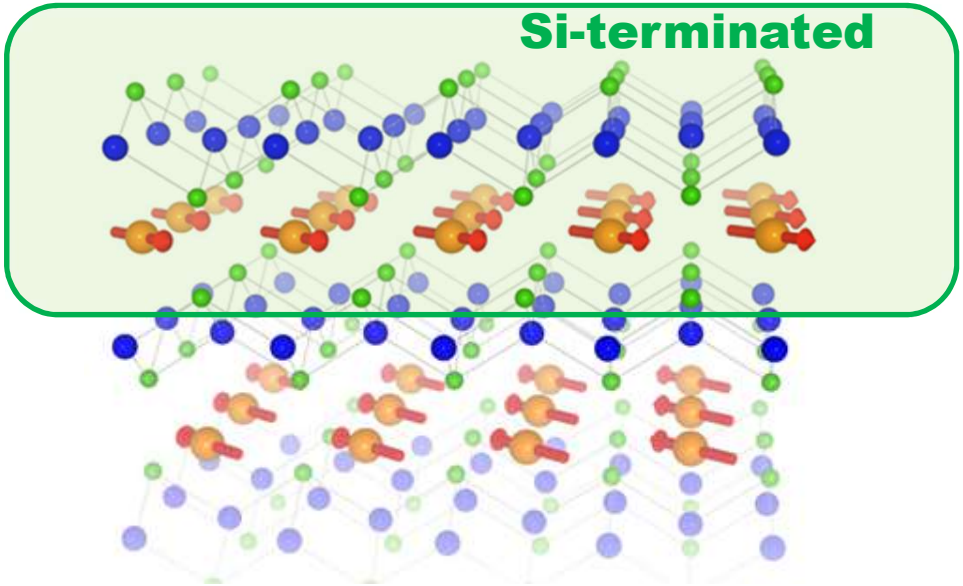
AFM below T_N = 24.5 K



T = 11 K





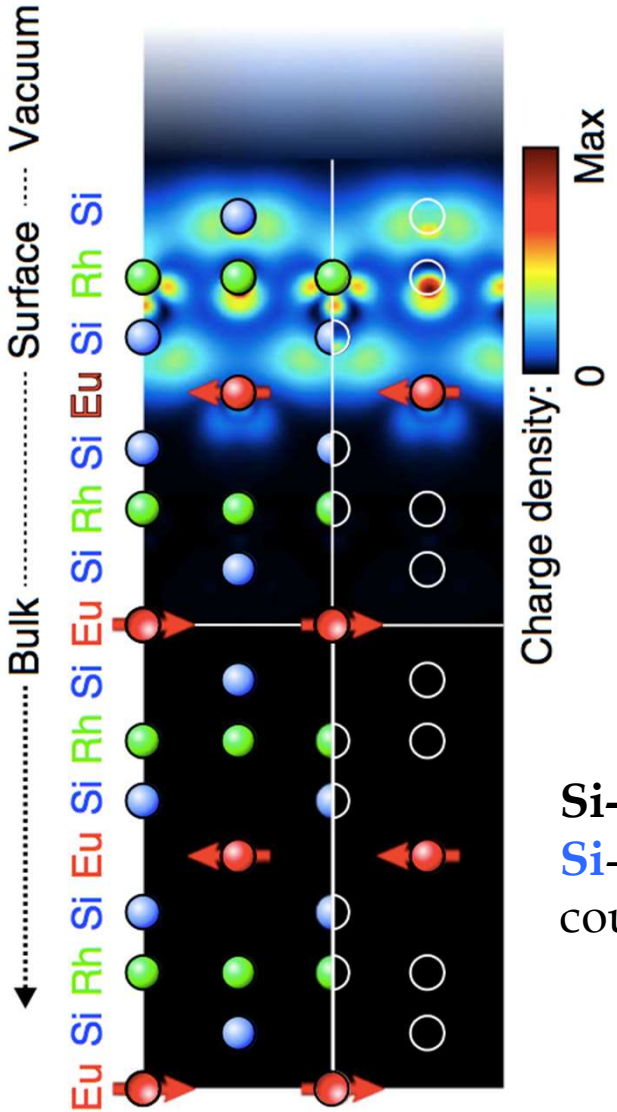


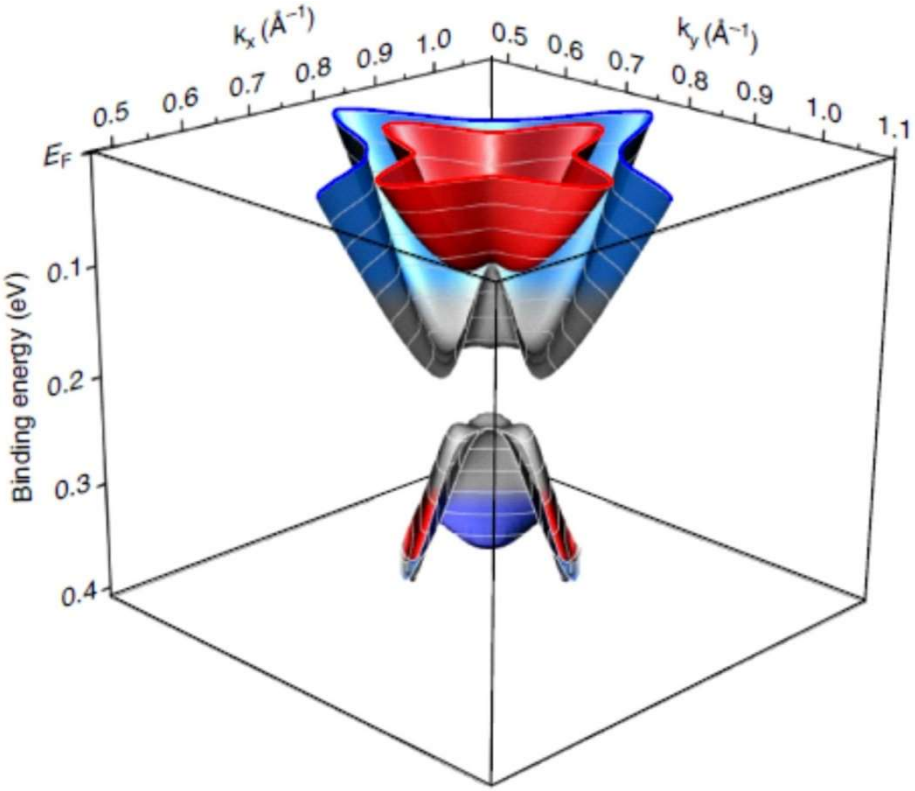
AFM order in the **bulk** starts at T_N ~ **24.5 K**,

Nature Comm. **5** 3171 (2014).
Nature Comm. **10** 796 (2019).

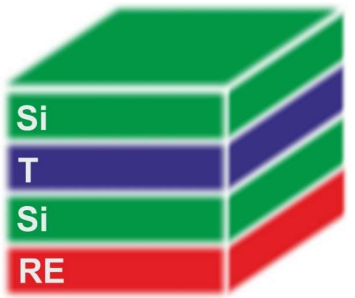
Surface state in the Si-Rh-Si-Eu surface block

$EuRh_2Si_2$



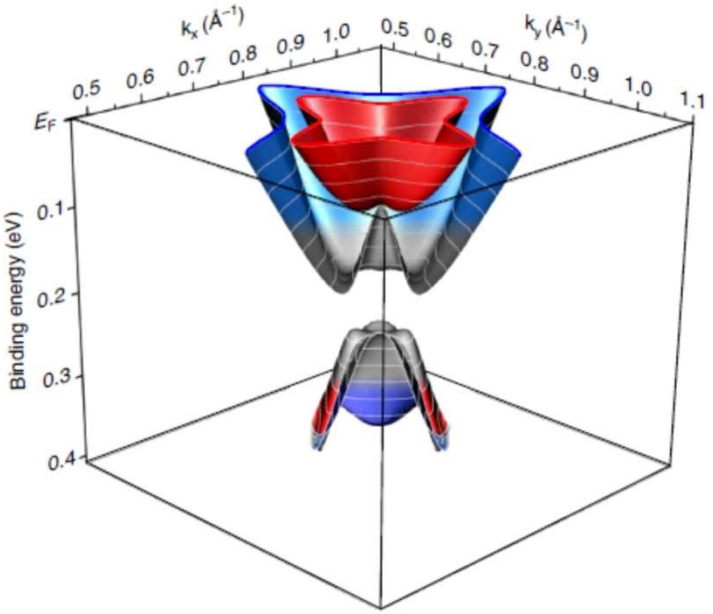


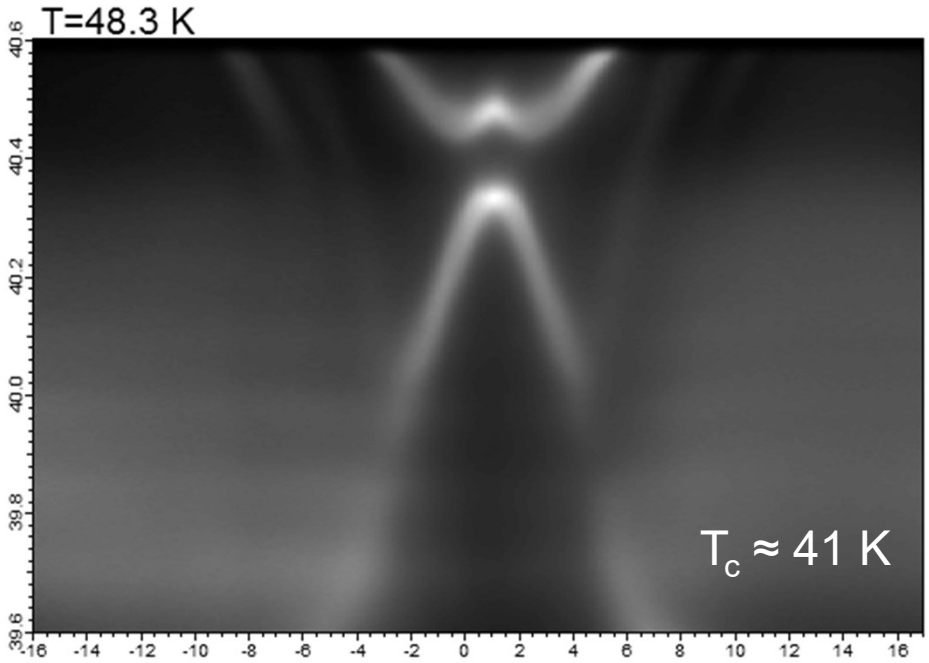
Si-termination: ferromagnetic **Eu** plane below topmost **Si**-**Rh**-**Si** trilayer splits Shockley state via exchange coupling and polarizes 2DES.



The ARPES-derived temperature evolution of the magnitude of the spin splitting

$EuRh_2Si_2$

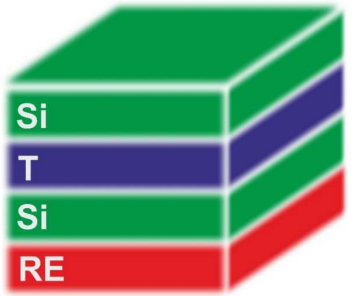




AFM order in the **bulk** starts at $T_N \sim 24.5$ **K**,

while the **Si-Rh-Si-Eu surface** becomes **ferromagnetic** already at **41 K** !!!

The spin splitting reaches a valueof 150 meV

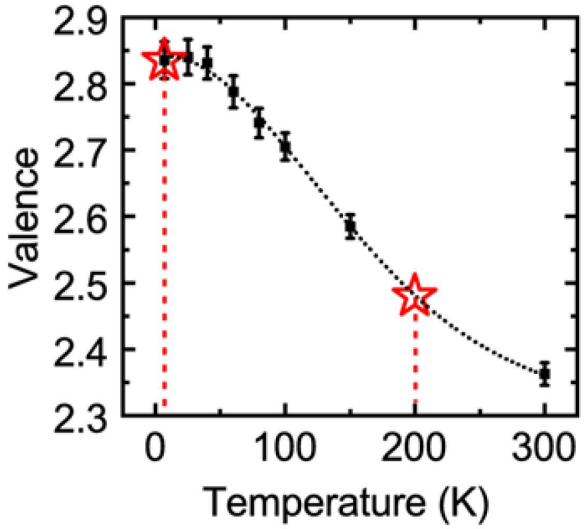


Nano Letters **17** 811 (2017).

$EuIr_2Si_2$: Valence fluctuating material discovered in 1986

- continuous valence change from **2.8** (4.2 K) to **2.2** (300 K);

- no magnetic ordering down to 4.2 K in the bulk;



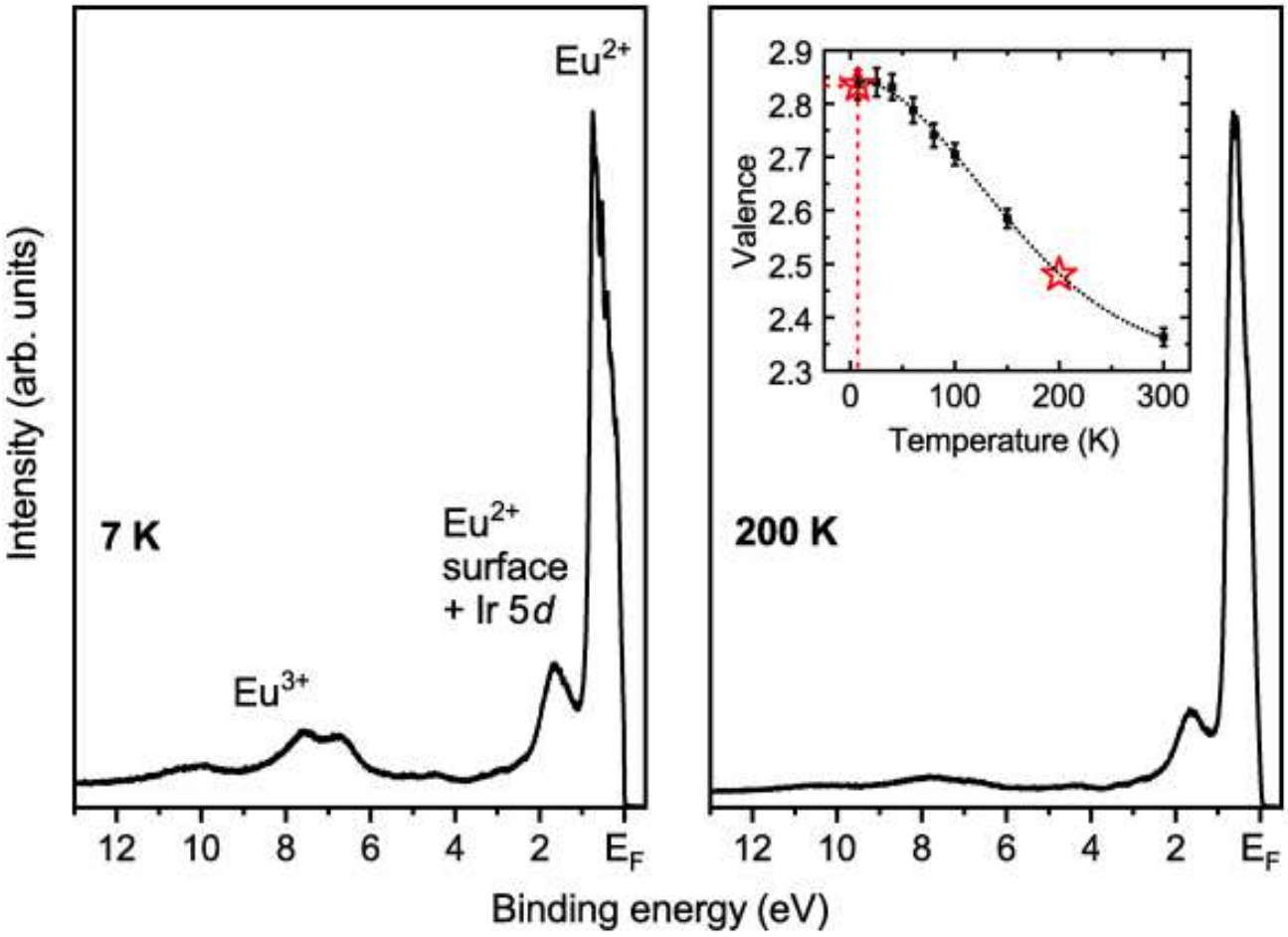
Our interest in this system:

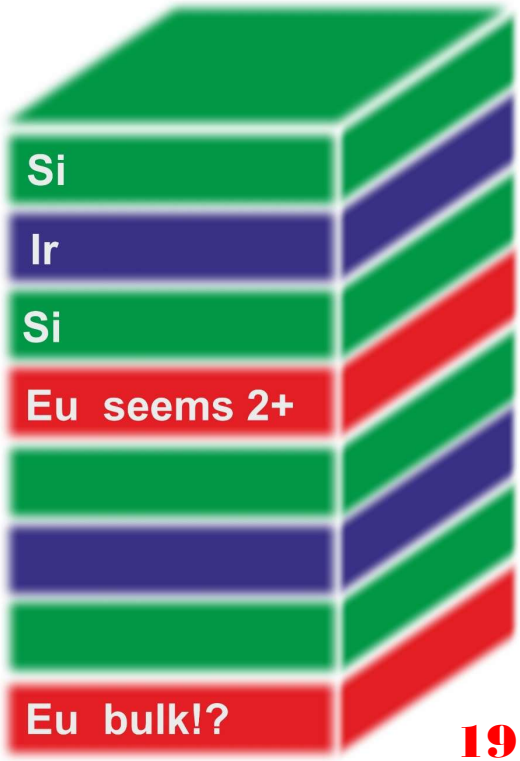
i) **Spin-orbit coupling** (Rashba effect) is **highly anticipated** at the **Si-Ir-Si-Eu** surface;

ii) **Valence-fluctuating properties** have **not** been **well explored** by **ARPES** so far.

npj Quantum Materials **4** 26 (2019).

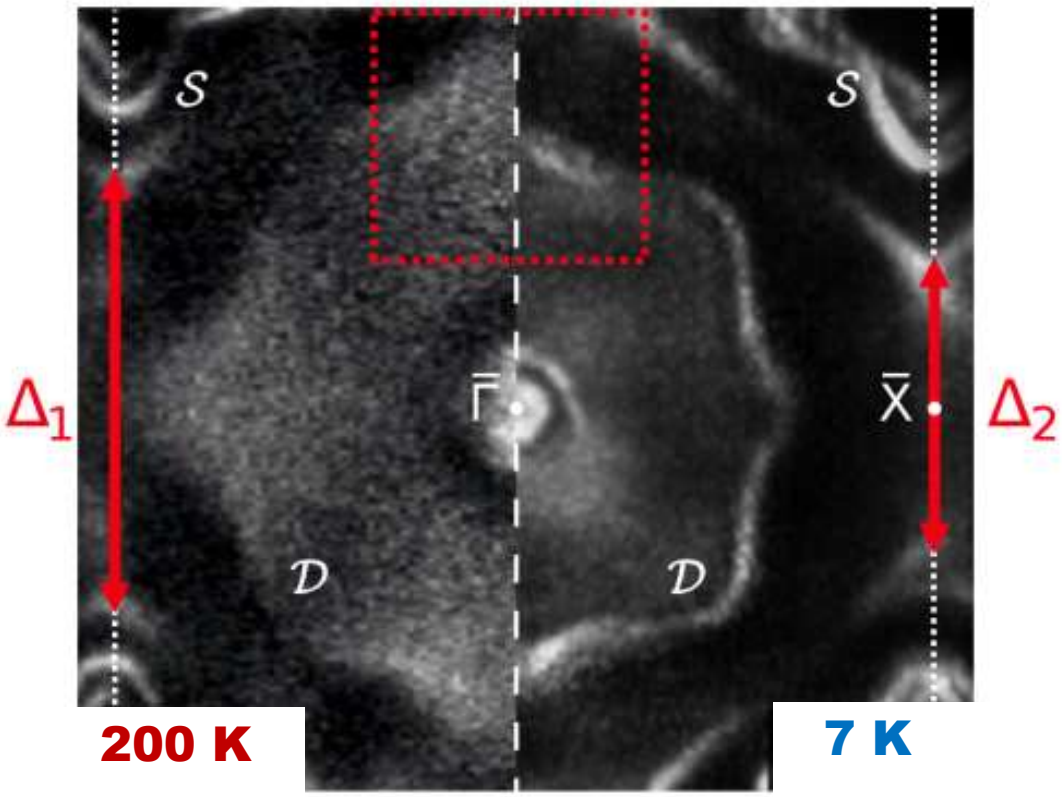
A huge Eu 2+ PE signal below Si-Ir-Si is a surprise ☺

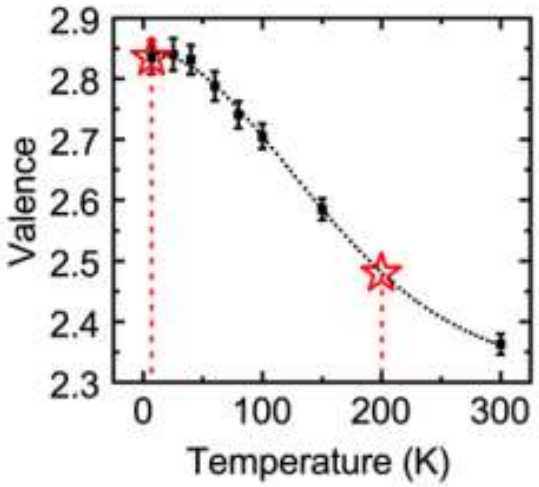


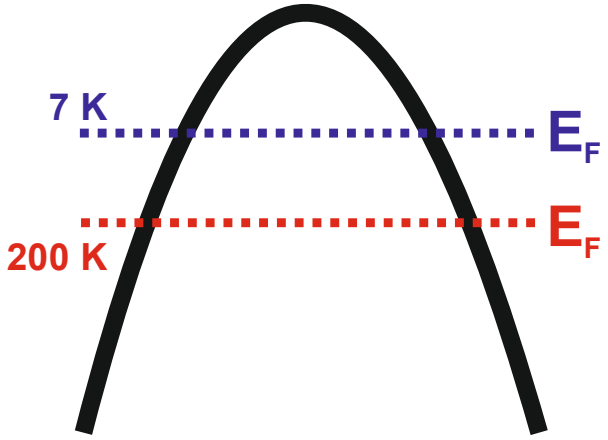


npj Quantum Materials **4** 26 (2019).

$EuIr_2Si_2$: ARPES insight into the valence fluctuation



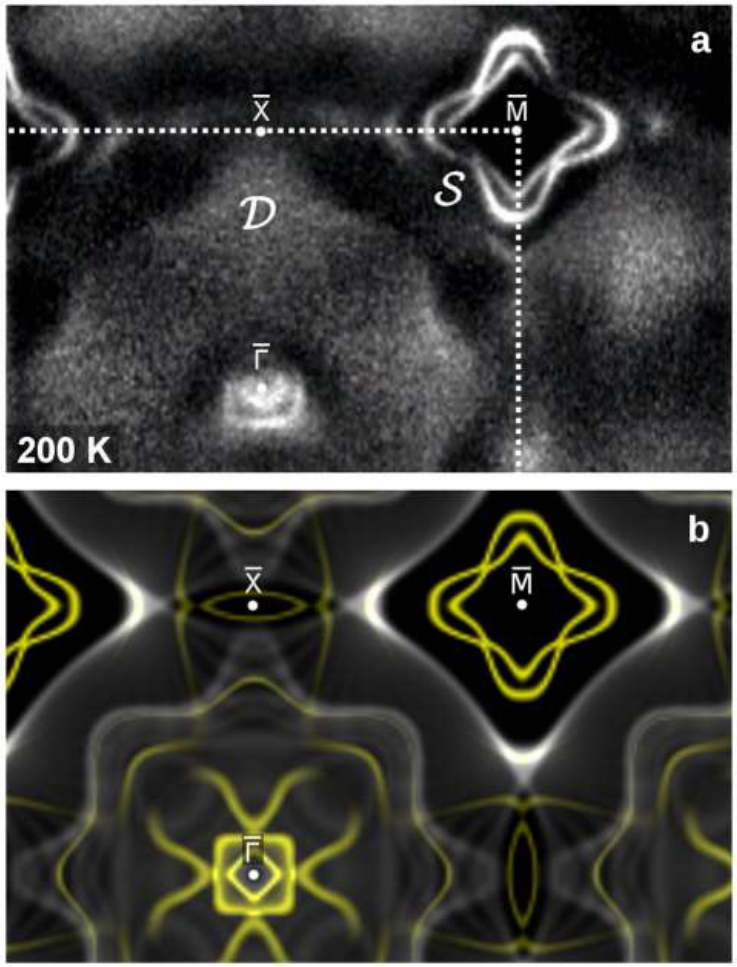




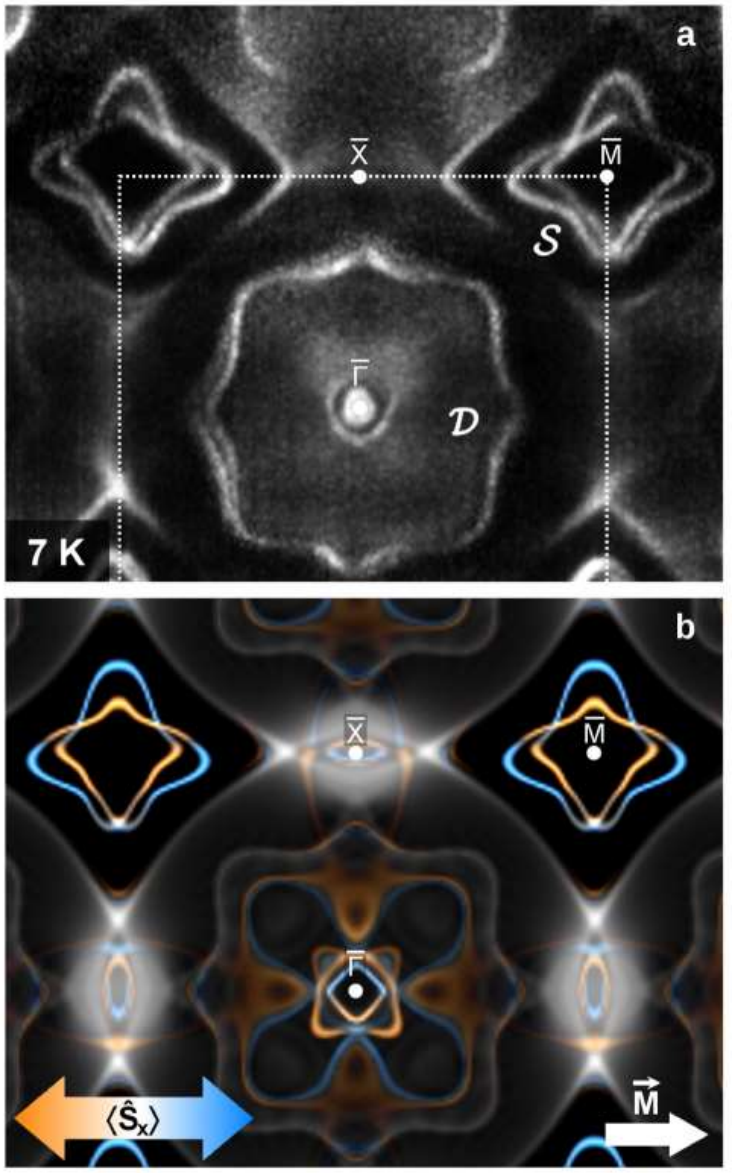
npj Quantum Materials **4** 26 (2019).

$EuIr_2Si_2$: Strong spin-orbit coupling and magnetism! ☺

200 K

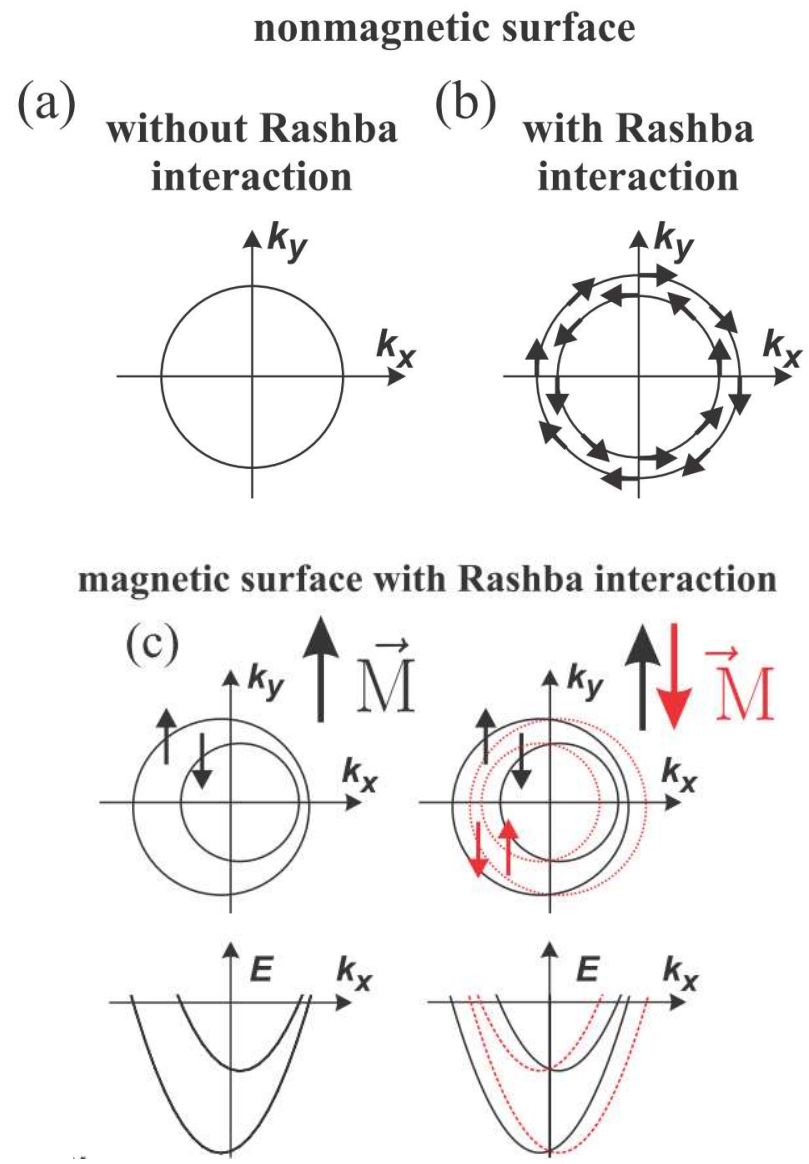


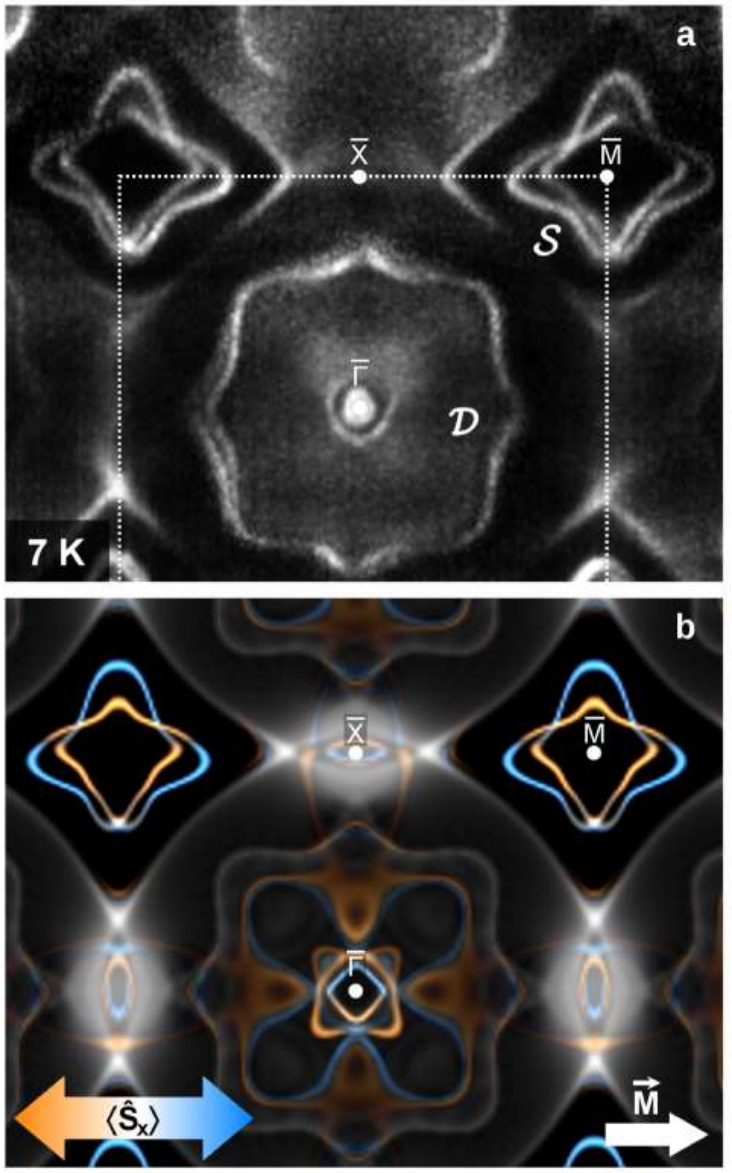
7 K



npj Quantum Materials **4** 26 (2019).

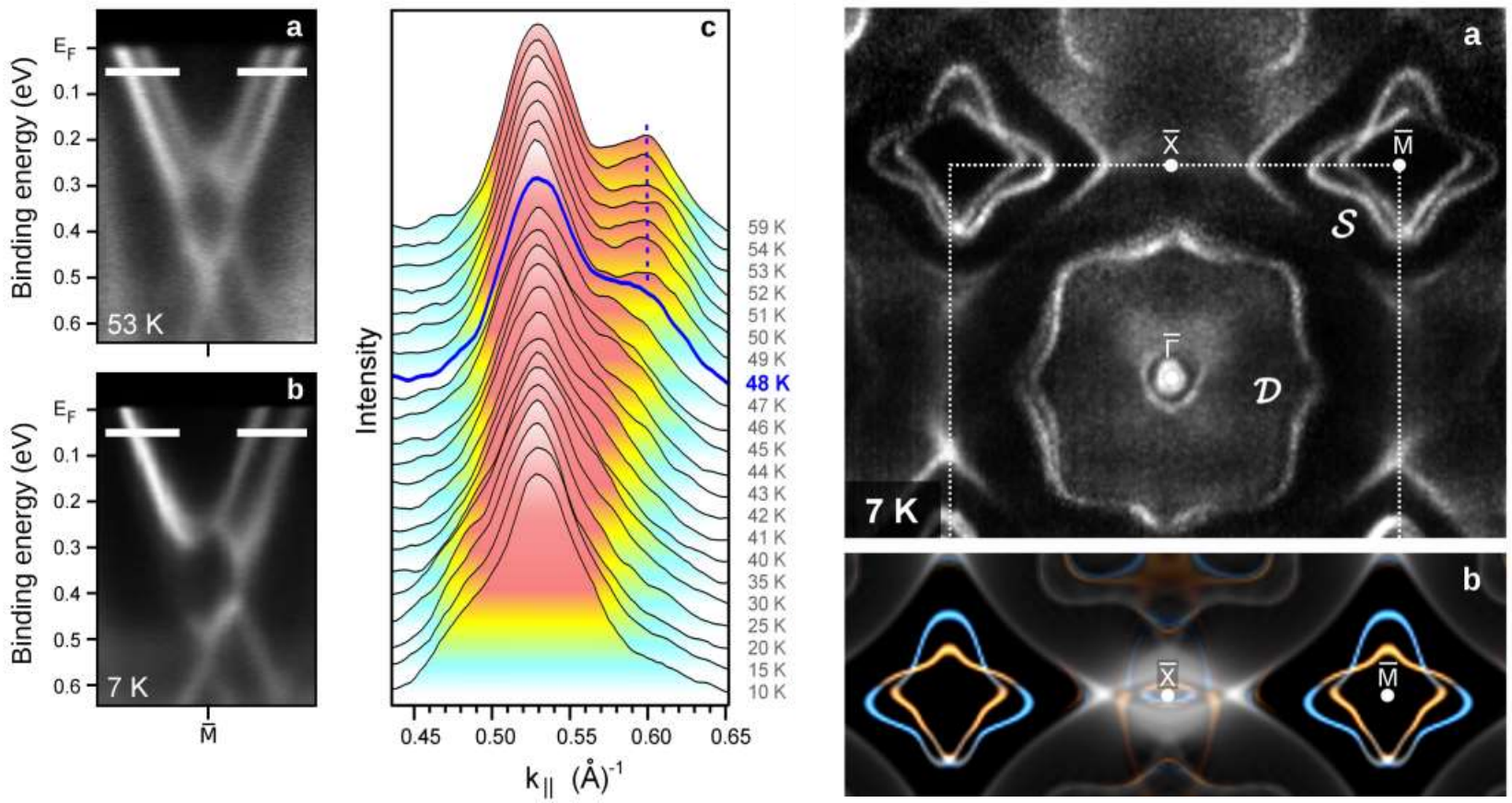
$EuIr_2Si_2$: Strong spin-orbit coupling and magnetism! ☺





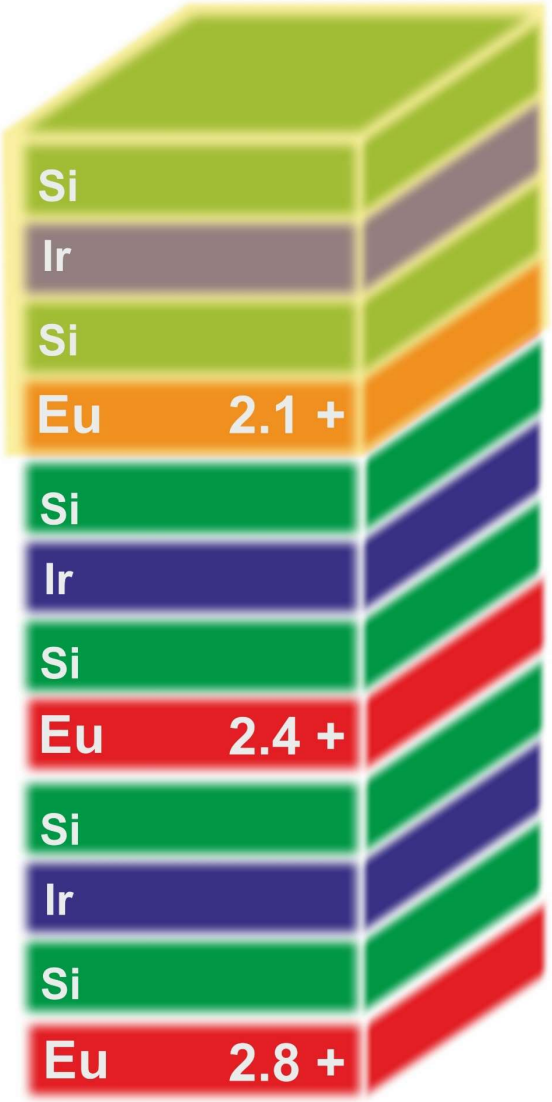
Gustav Bihlmayer NJP (2005).

$EuIr_2Si_2$: New temperature scale at the surface



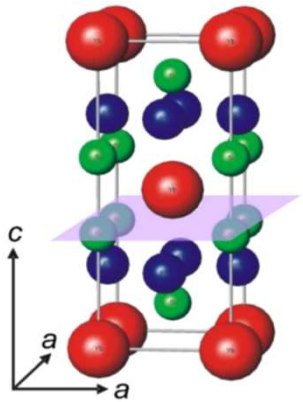
Eu 4*f* moments order below **48** K. The emerging exchange interaction modifies the spin polarization of the 2D surface electrons originally induced by the strong Rashba spin-orbit coupling effect.

$EuIr_2Si_2$: Non-magnetic in the *bulk*, strongly ferromagnetic at the *surface*



$T_C \sim 48K$

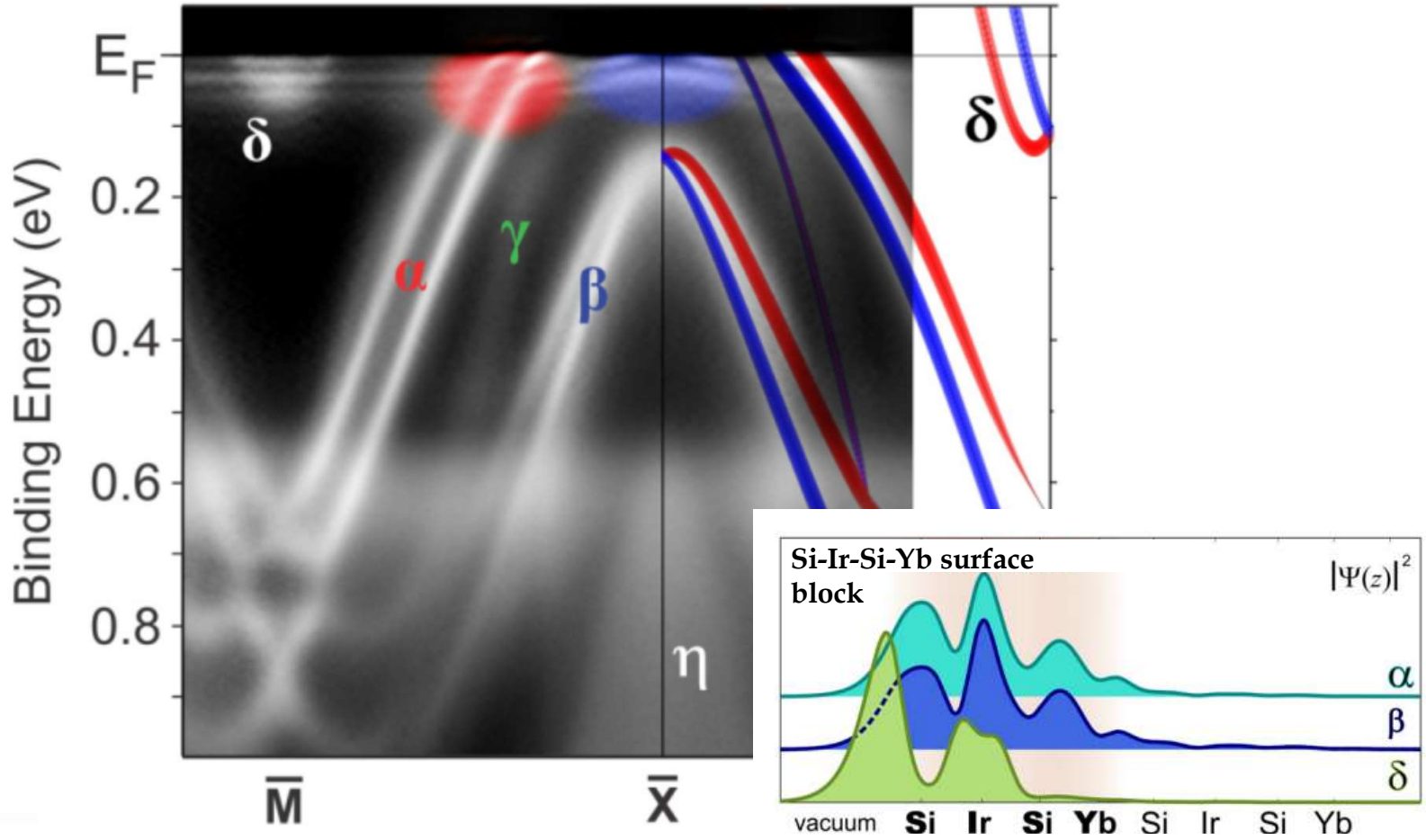
$T_C \sim 10K$



npj Quant. Mat. **4** 26 (2019); Phys. Rev. B **102** 205102 (2020) Phys. Rev. B **101** 245140 (2020)

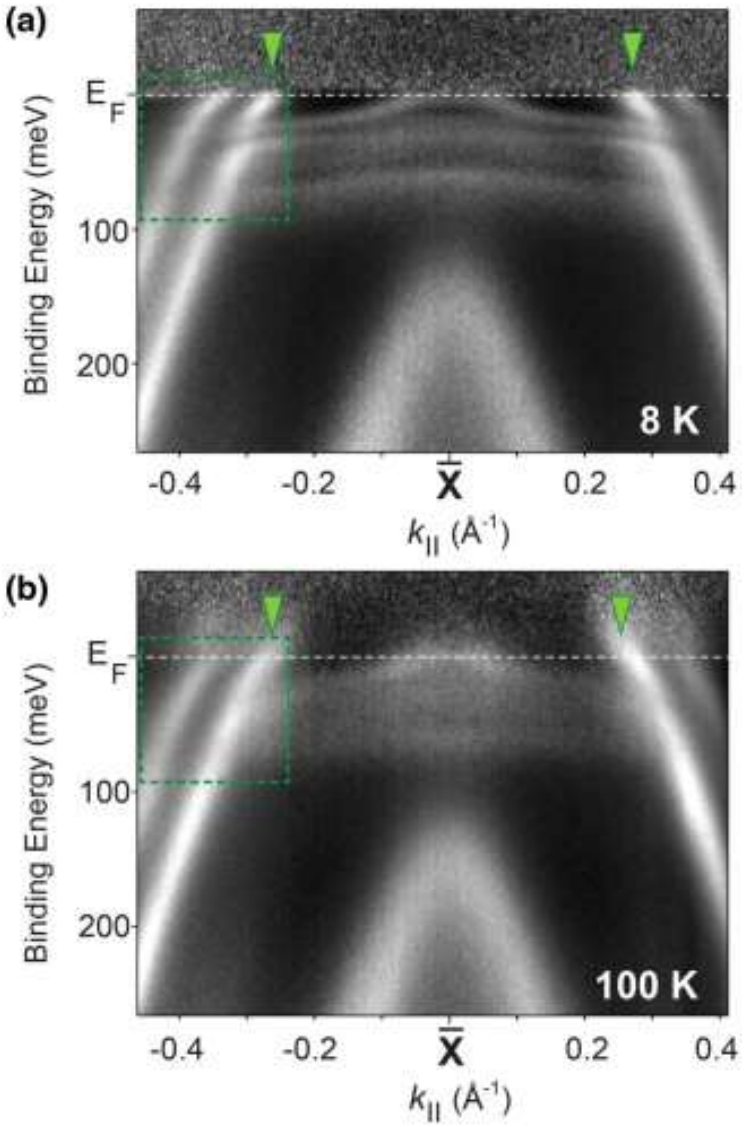
Surface of $YbIr_2Si_2$: 2D Kondo lattice with strong spin-orbit coupling

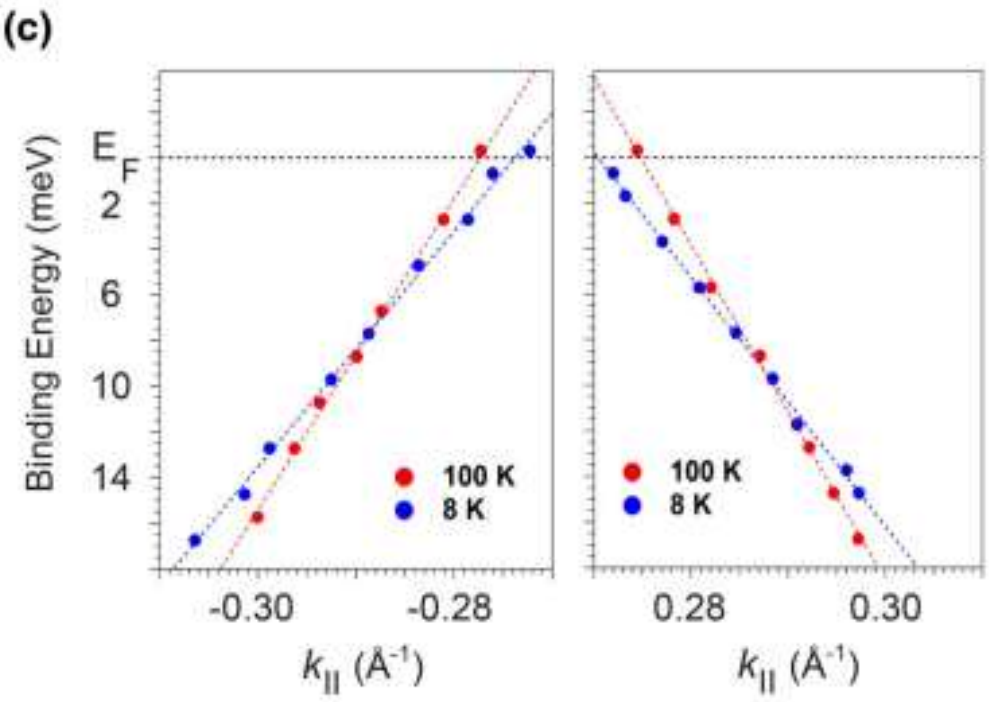
Si-Ir-Si-Yb surface block



Phys. Rev. B **98** 195438 (2018) (Editors' Suggestion).

Temperature-dependent *f-d* hybridization



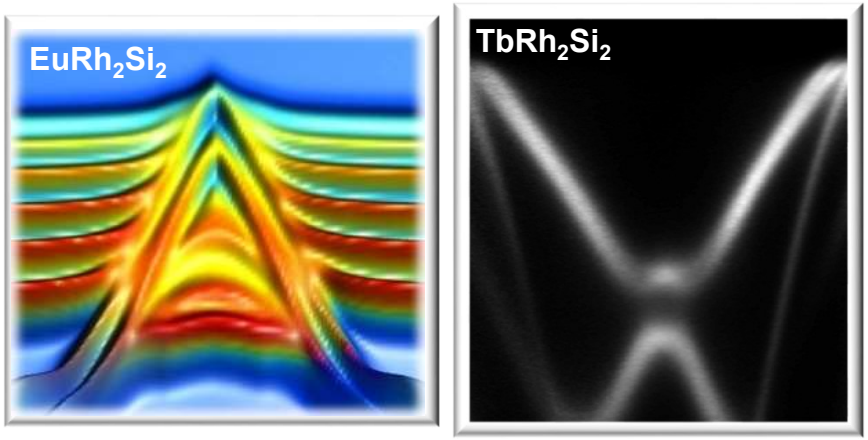


The 2D spin-polarized 4*f* electrons interact in a temperature-dependent fashion with highly mobile spin-polarized 2D itinerant states, gently guiding their velocities and spin texture.

Phys. Rev. B **98** 195438 (2018) (Editors' Suggestion); Phys. Rev. Lett. **124** 237202 (2020).

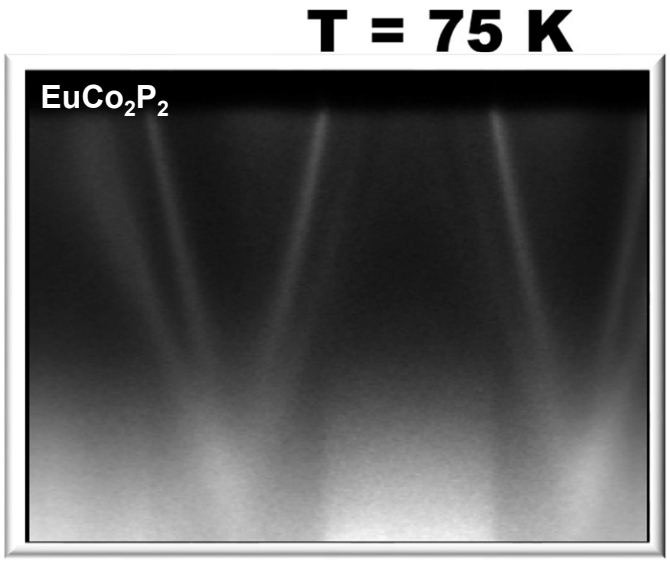
Outline of research activities: 2D-4f based systems

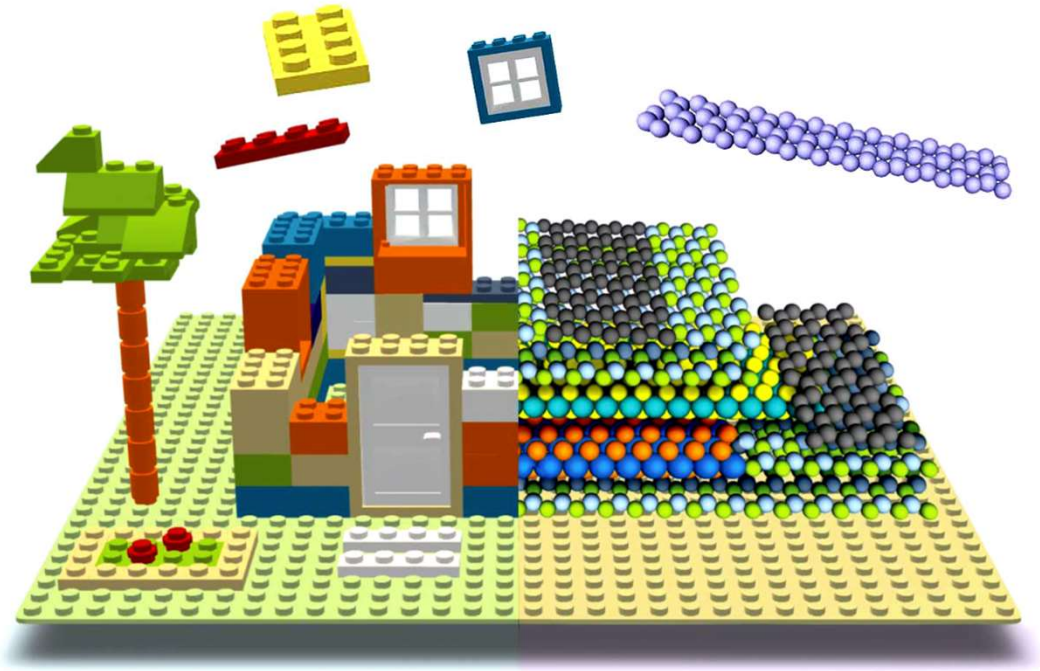
4f-electron systems;



… a veritable construction kit with **spin-orbit**, **Kondo, crystal-electric field**, and **exchange magnetic interactions** as building blocks…

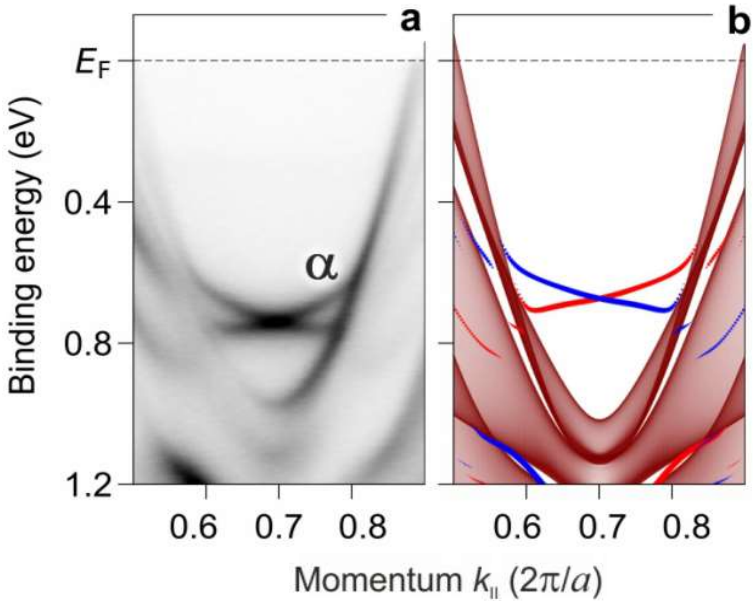
… to **design systems** for different scenarios and to study the **diversity** of **physics** driven by ***f-d* interactions**…





Outline of research activities: 2D-4f based systems

$CeIrIn_5$ Advanced Electronic Materials 2100768 (2021).



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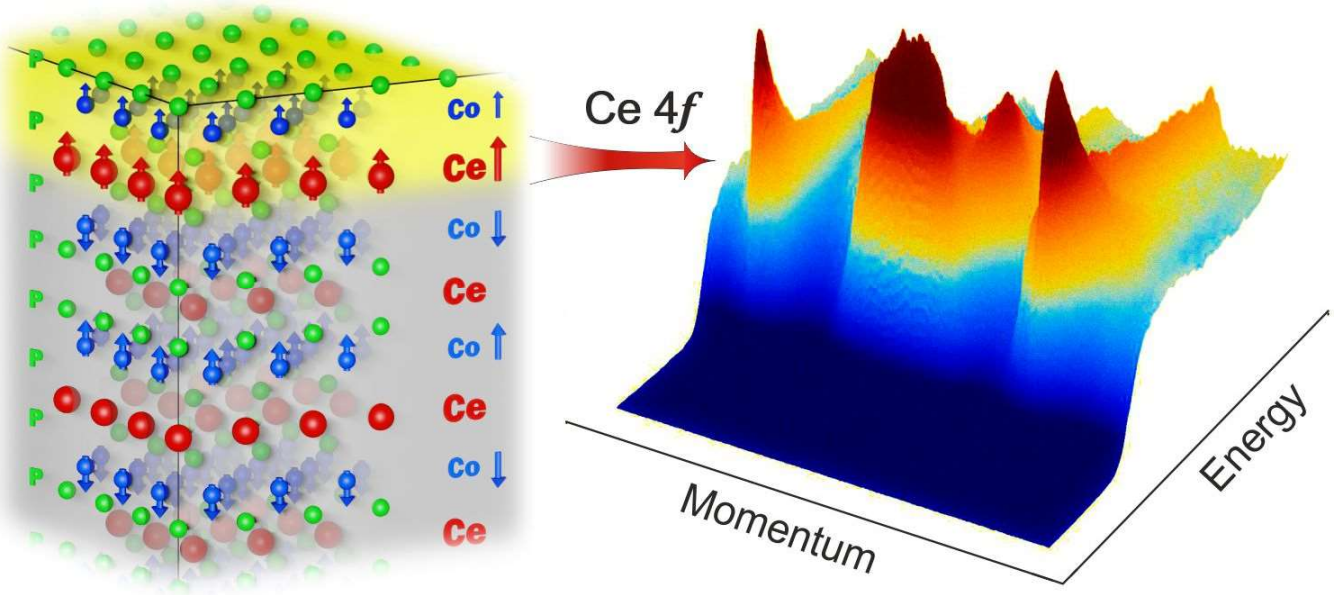
$CeCo_2P_2$



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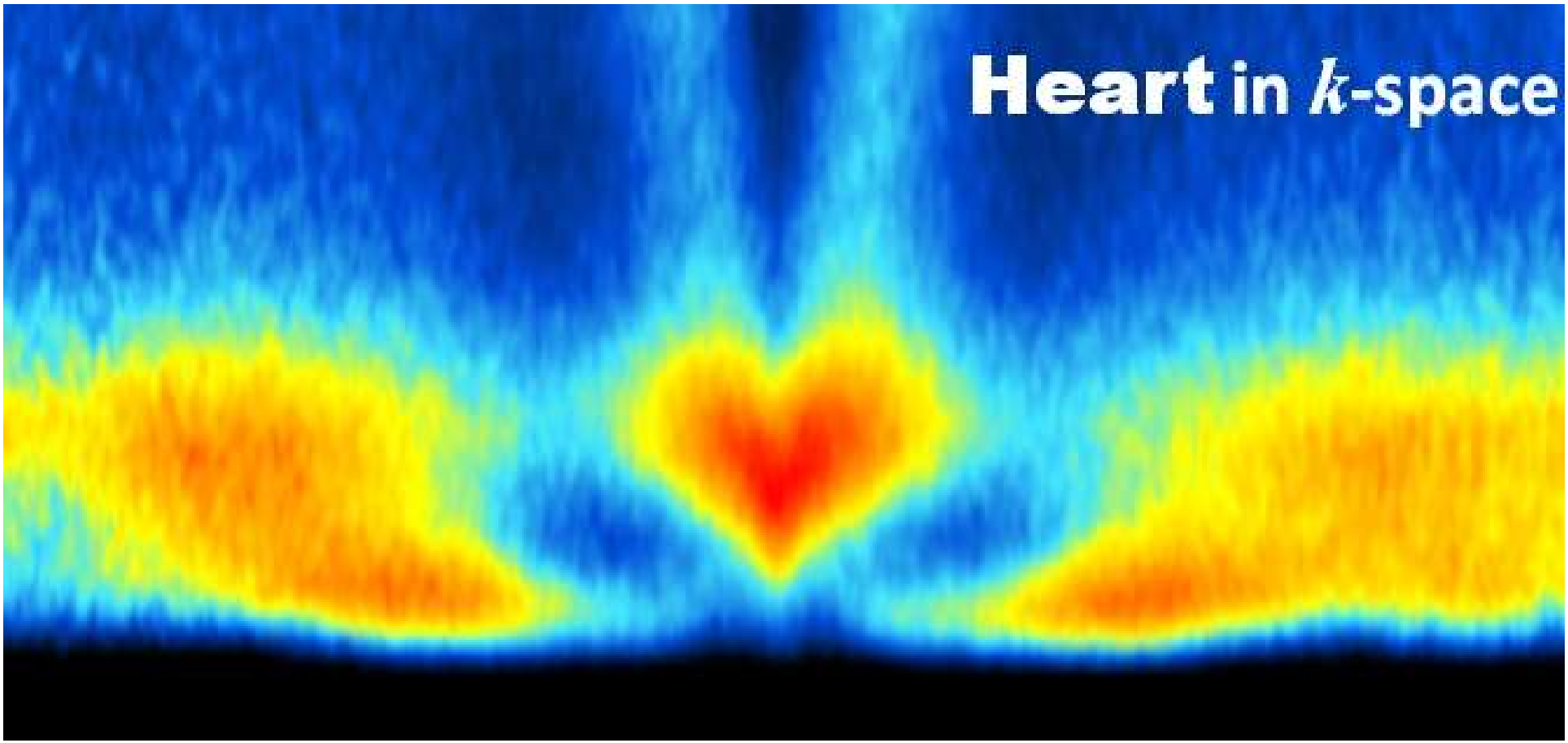
ikerbasque
Basque Foundation for Science


Heart in k-space