HAXPES on 4f and 5f
Strongly Correlated Systems

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Volumes, Localization & Total Energy

f-electrons on the threshold of instability. Small changes in energy or temperature can change the valence or localization.
Localization, Itinerancy and Narrow bands

4f and 5f states span the range of localized, itinerant and the interesting middle ground of narrow bands.
HAXPES can deliver on the promise of PES from air-prepared samples (in some cases). But we still need single crystals for intrinsic properties.

Sharp peak at the Fermi level is a characteristic feature of strongly correlated materials (magnetic or enhanced mass). Left: 5 strongly correlated uranium compounds with a sharp peak at the Fermi level.
USb$_2$ ARPES – Linewidths & Dispersion

Left: RESPES and ARPES which has better energy and angular resolution (also probably lifetime effects). Above: ARPES at the gamma point for 34 eV.

HAXPES adds core levels for screening, MFP, higher RESPES
USb$_2$ HAXPES and VUV

USb$_2$ data at 22 eV and 7600 eV. The valence bands are consistent with one another. One can get reliable electronic properties information from the most surface sensitive photon energies, but HAXPES widens the range of acceptable practices.
Surfaces Can Still Matter in HAXPES

The U 4f levels for USb$_2$, UPd$_3$ and UGe$_2$

$h\nu \sim$7.6 KeV  4f BE $\sim$377 eV

USb$_2$ cleaved in air and cleaved in vacuum differences
Not a deal breaker, but need to know about it
Surfaces Can Still Matter II

The Sb 3d levels for USb$_2$, and the U 4f with O 1s for UGe$_2$

$\nu \sim 7.6$ KeV

Note - resolution matters for the cores as well as the VB
The U 3d and 4d core levels for USb$_2$, UPd$_3$ and UGe$_2$

\( h\nu \sim 7.6 \text{ KeV} \) – 4d BE \( \sim 750 \text{ eV} \), - 3d BE \( \sim 3500 \text{ eV} \)

2 components to each SOS level except UPd$_3$
The Pu 4f core-levels for PuSb$_2$ binding energy ~ 430 eV at photon energies of 1500 and 3000 eV, and PuCoGa$_5$ at 1500 eV.
YbInCu$_4$ Divalent and Trivalent $f$-Electrons

The valence band and shallow cores for YbInCu$_4$. The In 4d levels are typical shallow cores with a filled shell initial state and a well defined SOS pair in the final state. The Cu 3d levels are band states that show dispersion in reciprocal space. The trivalent Yb 4f levels arise from an atomic-like $4f^{13}$ initial state going to a $4f^{12}$ final state.
Yb Occupancy, Valence Cross-sections

The relative and absolute cross sections for Yb 4f levels change as well as the MFP.

Valence for YbInCu₄ is constant from 90 to 5950 eV discounting a subsurface region.
For the heavy fermion YbAl₃ we see a consistent picture of the valence between 100 and 6000 eV. High quality single crystal are important for consistent results. Non 4f contributions are minimized in YbAl₃.
Yb Temperature, VB and 3d HAXPES

Valence band T-dep for 3 Yb materials with Yb 3d levels and reduction approach.
The temperature dependence and valence of the 3 Yb heavy fermions. The measured valence changes systematically depending on the binding energy of the probed energy level. The overall trends with T are consistent between different binding energies.
Summary – 4f & 5f Materials

- HAXPES deliver on the versatility for samples with varied preparation but one needs to be on guard. Contamination can be observable at 7.6 KeV.

- Good agreement for U and Yb materials between low and high energy with good samples and consistent experimental technique.

- Early work on Pu materials indicates a single screening channel for PuSb$_2$ and PuCoGa$_5$, USb$_2$ shows 2 U 4f screening channels.

- Determining the true valence in 4f and 5f systems continues to be a challenging issues for strongly correlated f-electron systems.

- The dependence on the f-electron occupancy with the binding energy of the probe level remains enigmatic.
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VUV/XPS

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