f- and *d*-derived Electronic Structure Studies of Rare-earth Systems

Jason N. Hancock
University of Connecticut

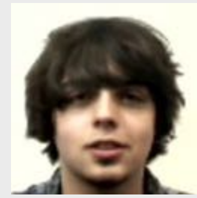
Outline

- UConn in a nutshell
- RIXS and heavy fermion/mixed-valent physics
- *L* edge RIXS in $YbInCu_4$
- *M* edge RIXS in $YbInCu_4$
- *L* edge RIXS in rare-earth hexaborides

Collaborators and support

Sahan Handunkanda (PhD '18)
Erin Curry
Vincent Flynn
Donal Sheets






Maxim Dzero

Mary Upton
Diego Casa
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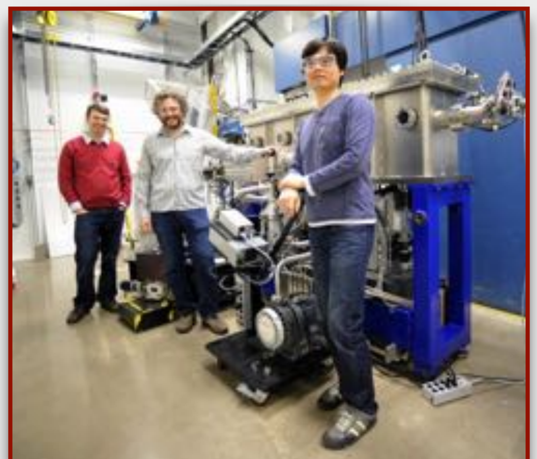
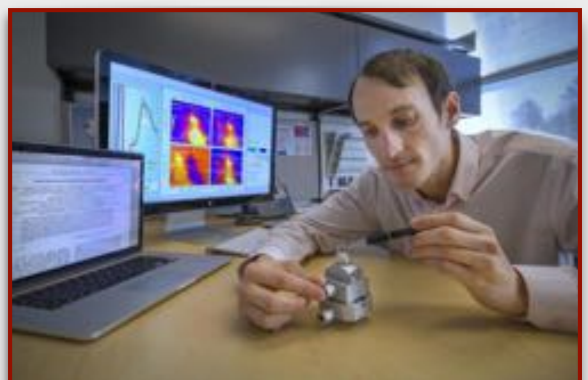


Vladimir Strocov
Thorsten Schmitt

Marco Grioni
Marco Guarise



Ignace Jarrige
Daniel Mazzone

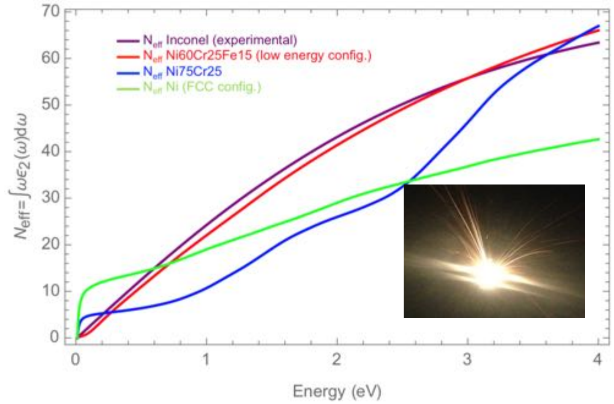
Akio Kotani

Hiro Yamaoka
Naohito Tsujii
Kenji Ishii

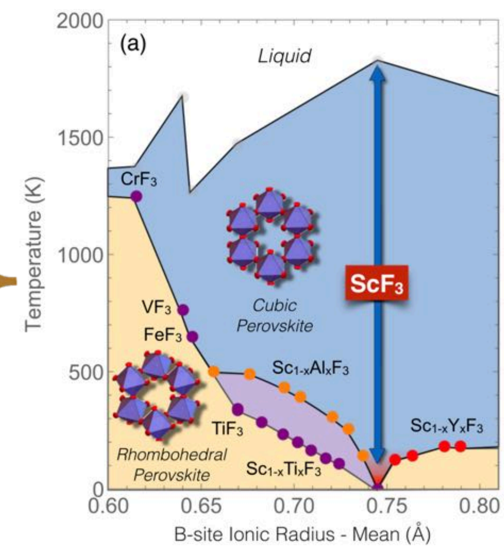
Work supported by:

Photon Science at UConn

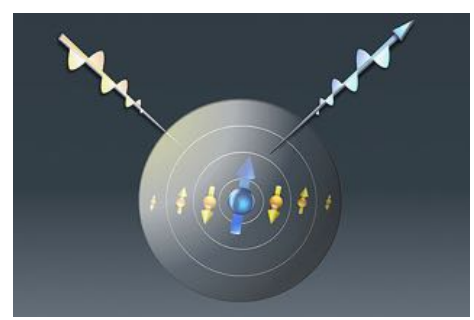
- Additive manufacturing



- Negative thermal expansion near structural quantum phase transitions



- *f*-electron physics via RIXS



UConn Condensed Matter Physics

Alexander Balatsky
Theory

Gayanath Fernando
Theory

Boris Sinkovic
Thin films synthesis
Electron spectroscopy

Niloy Dutta
Photonics
Applied physics

Jason Hancock
THz, Infrared, X-ray
Applied physics

Ilya Sochnikov
Transport
Scanning SQUID

Elena Dormidontova
Soft Matter Theory

Menka Jain
Thin film synthesis

Barrett Wells
PLD films, Muons,
Neutrons, ARPES

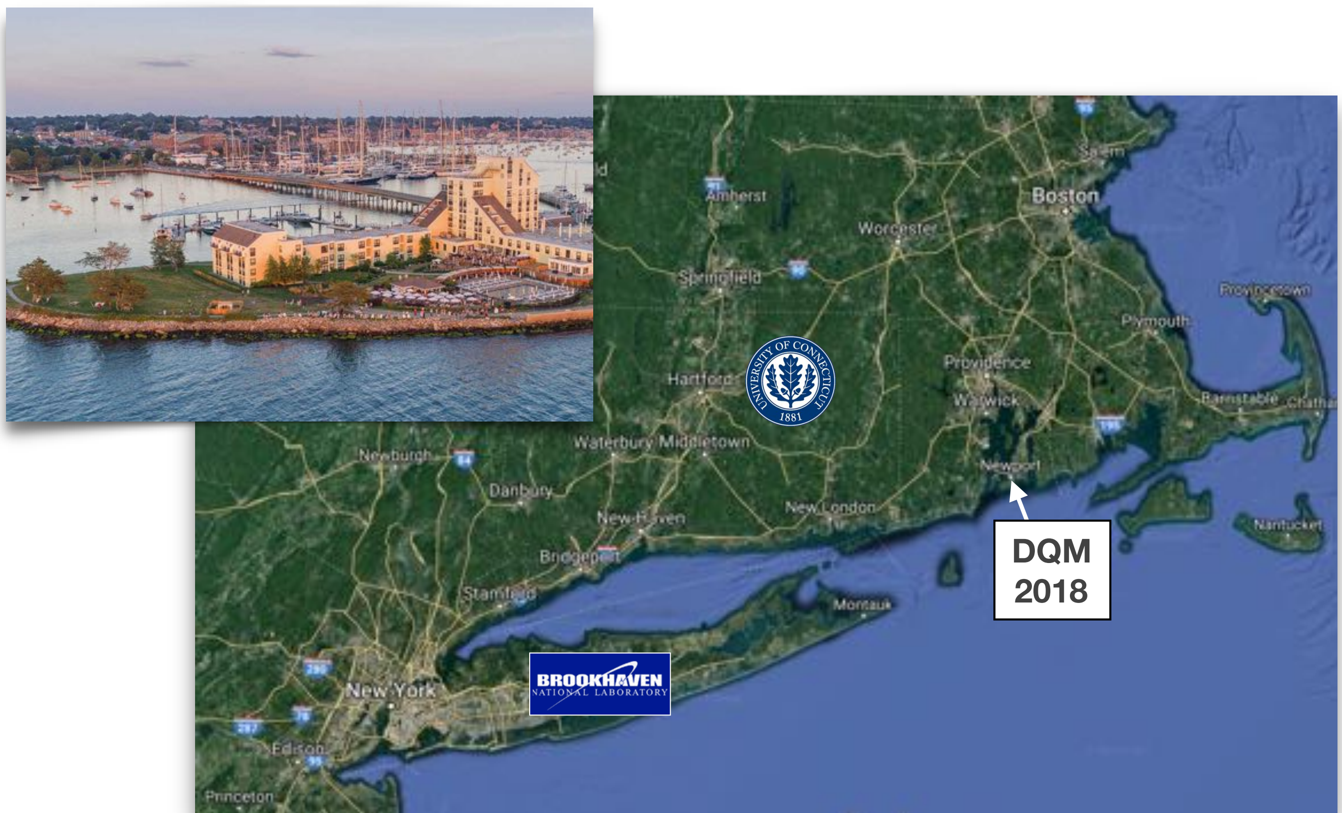
Plus strong connections with: AMO group, UConn Institute for Materials Science, New UConn Tech Park

Dynamic Quantum Matter, Entangled Order and Quantum Criticality Workshop

http://quantum-matter.uconn.edu/

Newport, Rhode Island June 18-June 19, 2018

Organizers: A. Balatsky, I. Sochnikov, G. Fernando
P. Chandra, J. N. Hancock, C. Trallero



Confirmed Invited Speakers

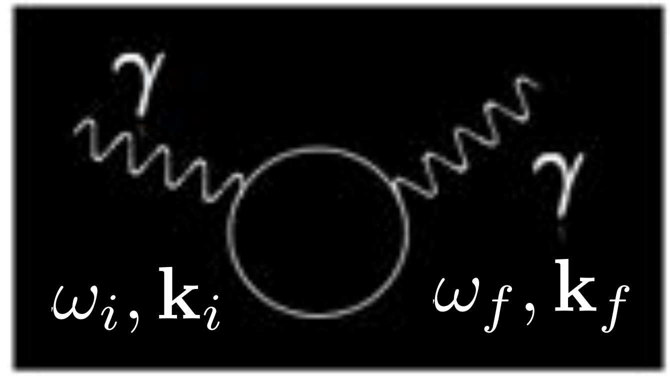
Premi Chandra (Rutgers)
Jason Haraldsen (UNF)
Beena Kalisky (Bar-Ilan University)
Jeremy Levy (PITT)
Peter Littlewood (U. Chicago)
Rohit Prasankumar (LANL)
Kamran Behnia (ESPCI-Paris)
Kazushi Kanoda (U-Tokyo)
Peter Johnson (Brookhaven)
Jagadeesh Moodera (MIT)
Susanne Stemmer (UCSB)
Ignace Jarrige (Brookhaven)
Matthias Geilhufe (Nordita)
Charles Ahn (Yale)
Keith Nelson (MIT)
Alexander Balatsky (UConn)
Barrett Wells (UConn)
Gayanath Fernando (UConn)
Ilya Sochnikov (UConn)
Daniel Mazzone (Brookhaven)
Kirsty Dunnett (Nordita)
Vladimir Juricic (Nordita)

Sponsors

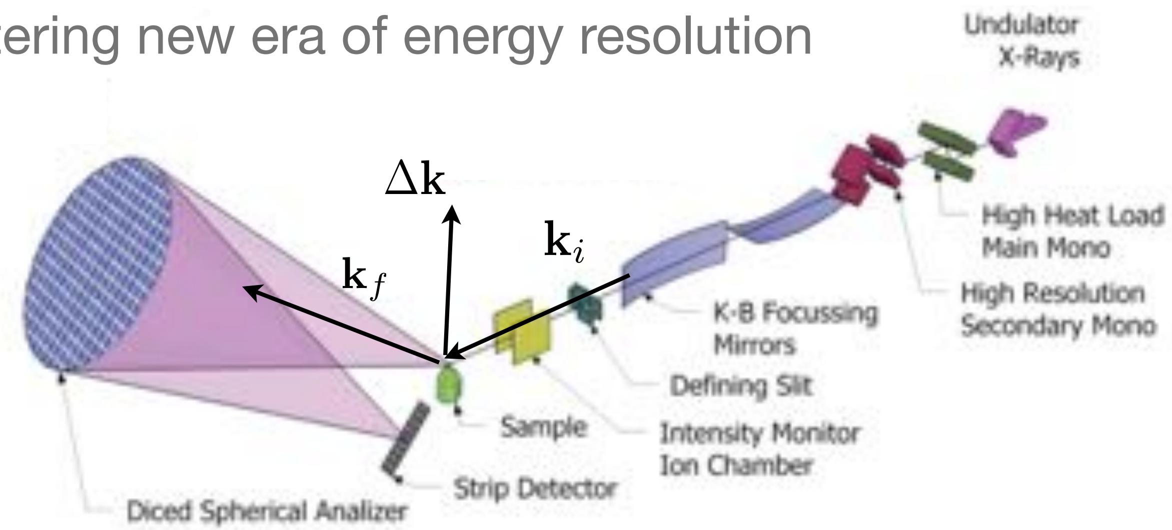
University of Connecticut 1881 · Department of Energy, United States of America · NSF · VILLUM FONDEN · WILEY · NORDITA · Los Alamos National Laboratory EST. 1943

Resonant Inelastic X-ray Scattering (RIXS)

- Resonant Raman spectroscopy, with resonance at an X-ray edge
- Bulk sensitive - can probe surface-sensitive samples
- Large momentum transfer, 3D control
- Atomic-species and valence-specific information
- Lots of control (polarization, angle, *Q*, E_i)
- Entering new era of energy resolution



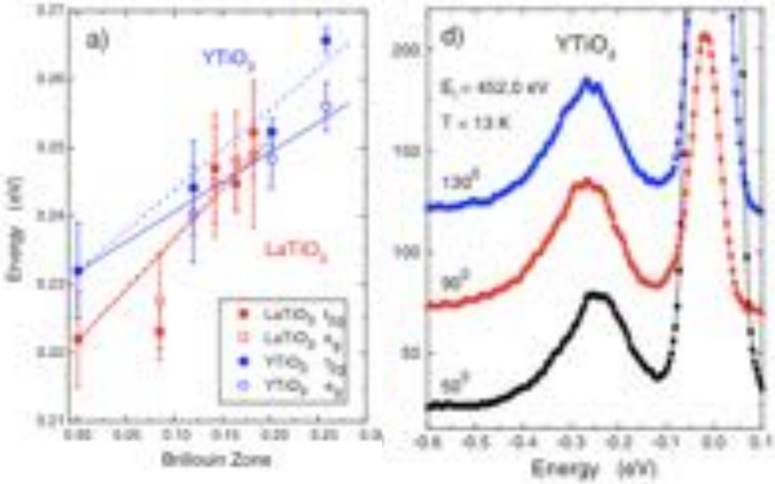
$$\Delta\omega = \omega_i - \omega_f$$
$$\Delta\mathbf{k} = \mathbf{k}_i - \mathbf{k}_f$$



Kramers-Heisenberg:
$$\frac{d^2\sigma}{d\Omega_{k'} d(\hbar\omega_k')} = \frac{\omega_k'}{\omega_k} \sum_{|f\rangle} \left| \sum_{|n\rangle} \frac{\langle f|T^\dagger|n\rangle\langle n|T|i\rangle}{E_i - E_n + \hbar\omega_k + i\frac{\Gamma_n}{2}} \right|^2 \delta(E_i - E_f + \hbar\omega_k - \hbar\omega_k')$$

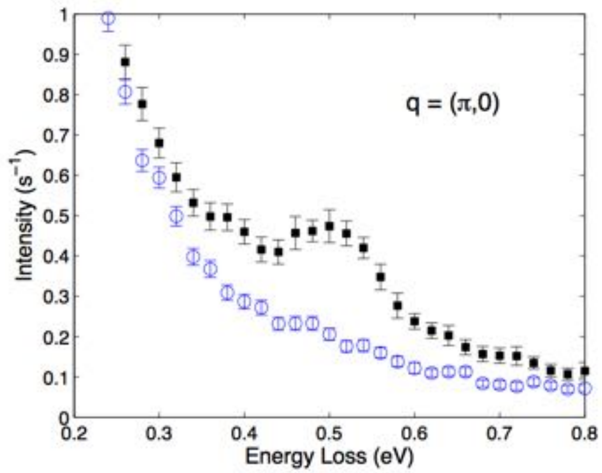
Some excitations observed with RIXS

Orbitons



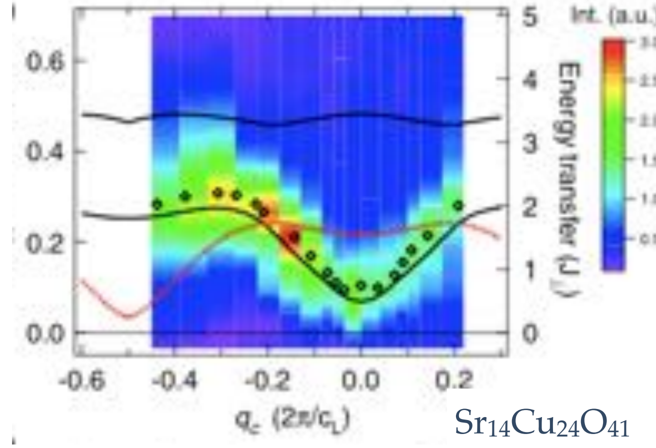
Ulrich, et al PRL **103**, 107205 (2009)

2-magnon



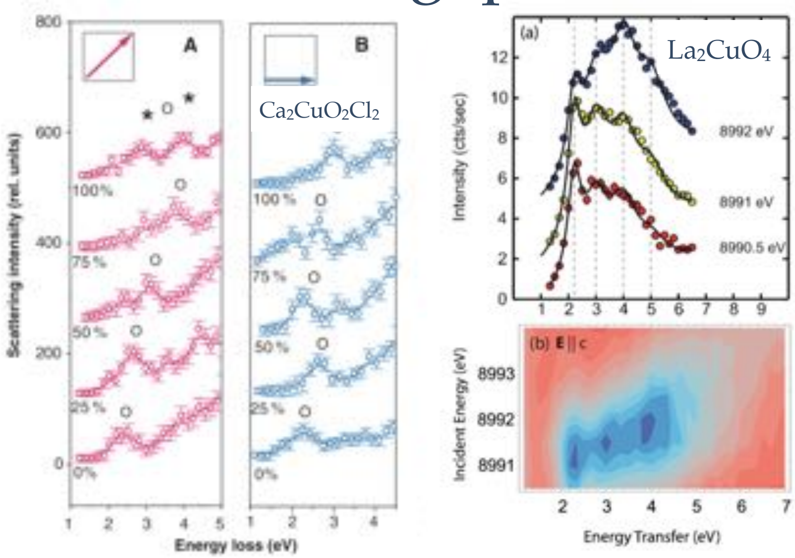
Hill, et al PRL **100**, 097001 (2008)

Triplon



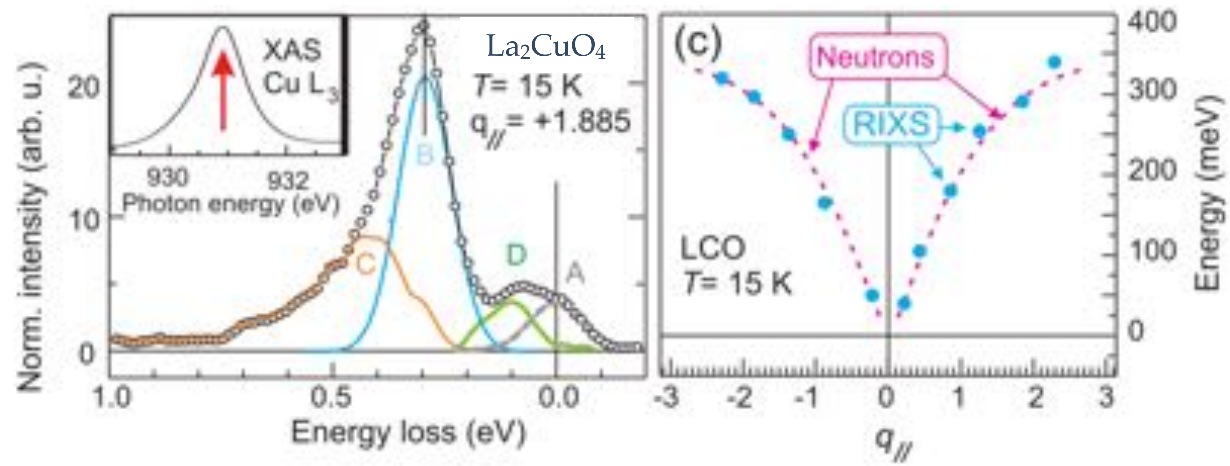
Schlappa, et al PRL **103**, 047401 (2009)

Mott gap



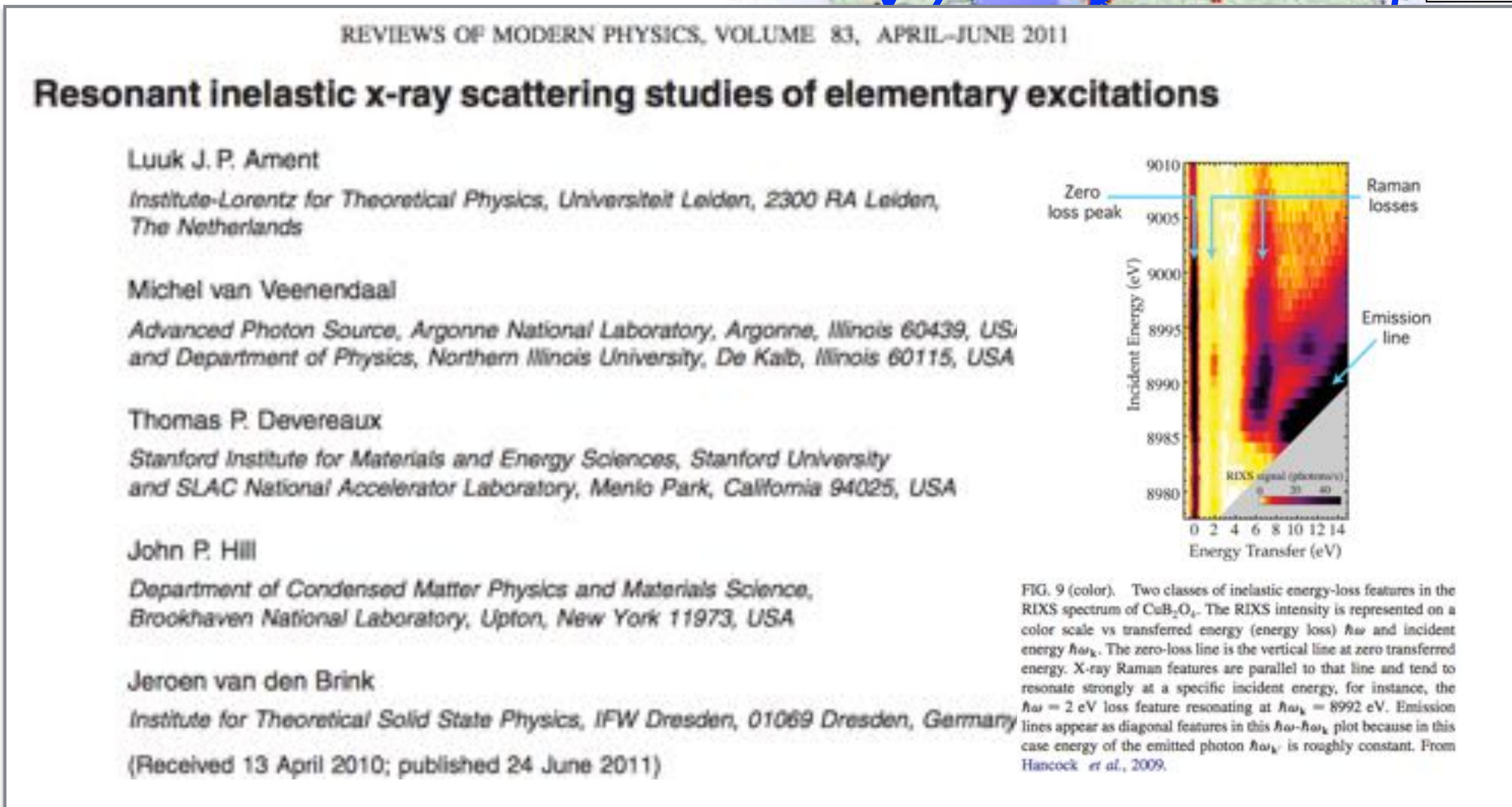
Hasan, et al Science, **288**, 1811 (2000)
Li, JNH, et al, PRB **74**, 224509 (2006)

Single-magnon



Braichovich, et al PRL **102**, 167401 (2009)
Braichovich, et al PRB **81**, 174533 (2010)

RIXS today



REVIEWS OF MODERN PHYSICS, VOLUME 83, APRIL–JUNE 2011

Resonant inelastic x-ray scattering studies of elementary excitations

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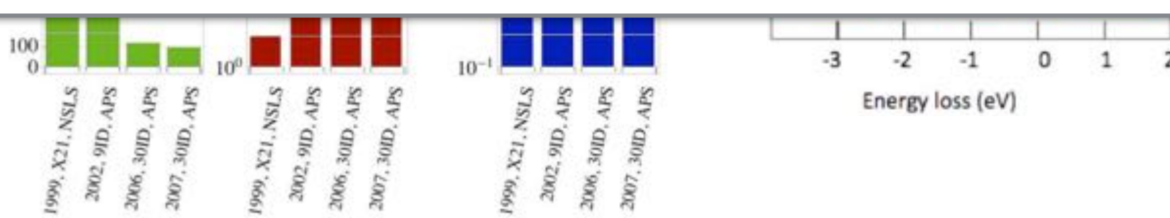
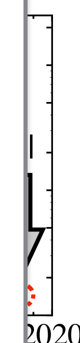
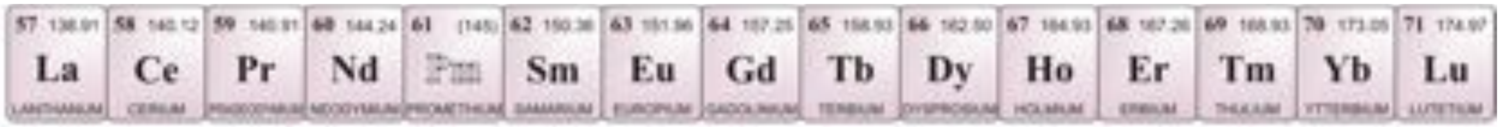
(Received 13 April 2010; published 24 June 2011)

FIG. 9 (color). Two classes of inelastic energy-loss features in the RIXS spectrum of CuB_2O_4. The RIXS intensity is represented on a color scale vs transferred energy (energy loss) $\hbar\omega$ and incident energy $\hbar\omega_k$. The zero-loss line is the vertical line at zero transferred energy. X-ray Raman features are parallel to that line and tend to resonate strongly at a specific incident energy, for instance, the $\hbar\omega = 2$ eV loss feature resonating at $\hbar\omega_k = 8992$ eV. Emission lines appear as diagonal features in this $\hbar\omega$-$\hbar\omega_k$ plot because in this case energy of the emitted photon $\hbar\omega_{k'}$ is roughly constant. From Hancock *et al.*, 2009.

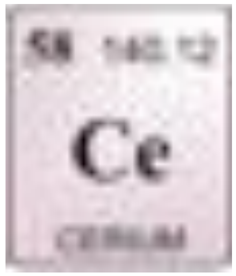




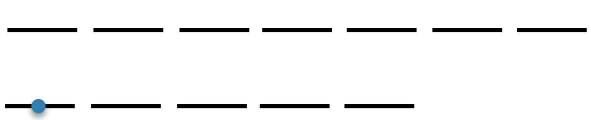
Yb versus Ce compounds for RIXS experiments



f^1

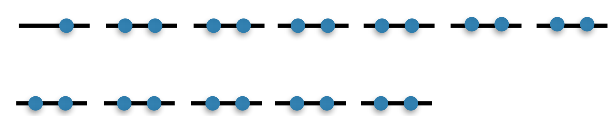


f^{13}



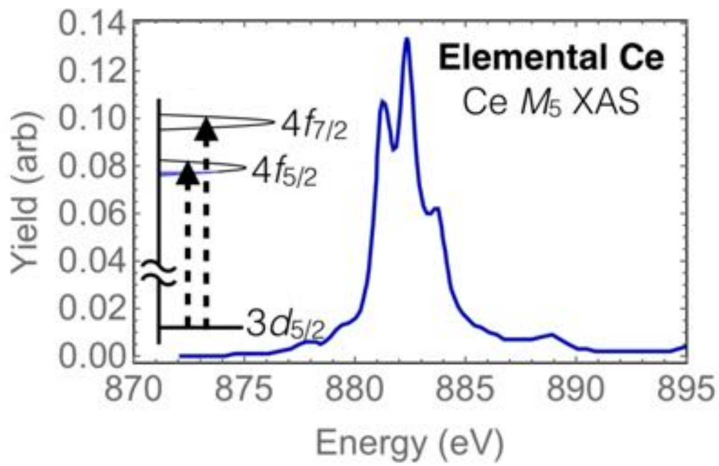
J=7/2

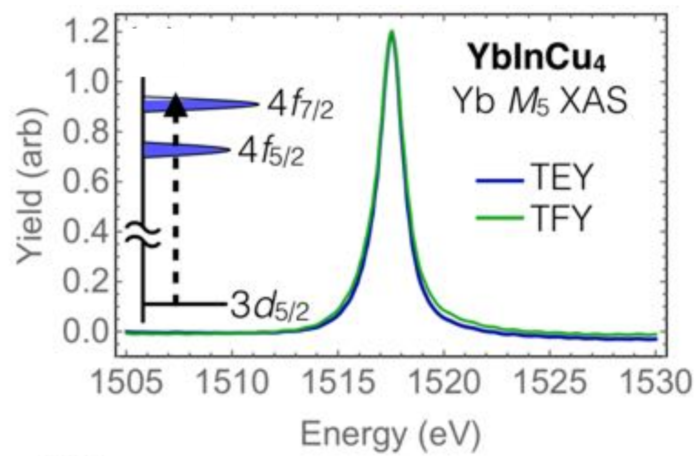
J=5/2



Complex *M* edge absorption,
high multiplet intermediates

Simple *M* edge absorption
Full shell intermediate state



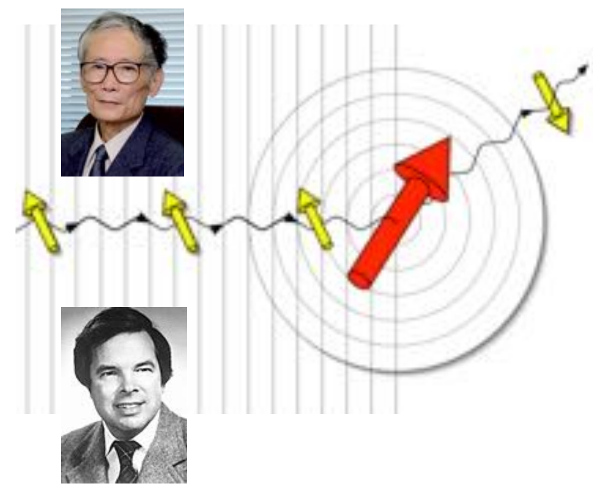


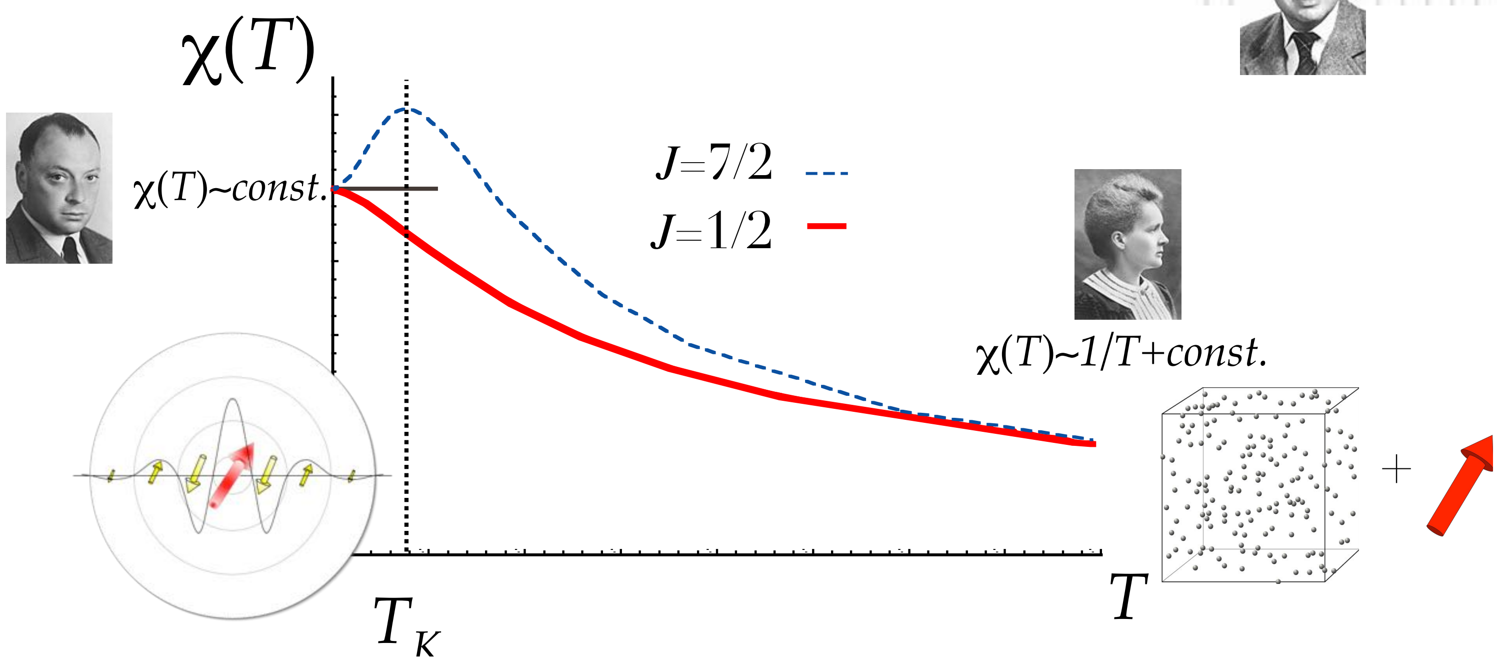
Heavy fermions ($CeAl_3$), m*~1000 me
Small energy scale ~10s of meV for hybridization

Physically larger orbitals
Smaller effective mass
Larger hybridization scale ~100s of meV

The Kondo problem - new area for RIXS

- Kondo effect - metallic electron states exchange-coupled to a localized magnetic impurity
- Theoretically challenging problem- solved by Ken Wilson and others (1982 Nobel in Physics)

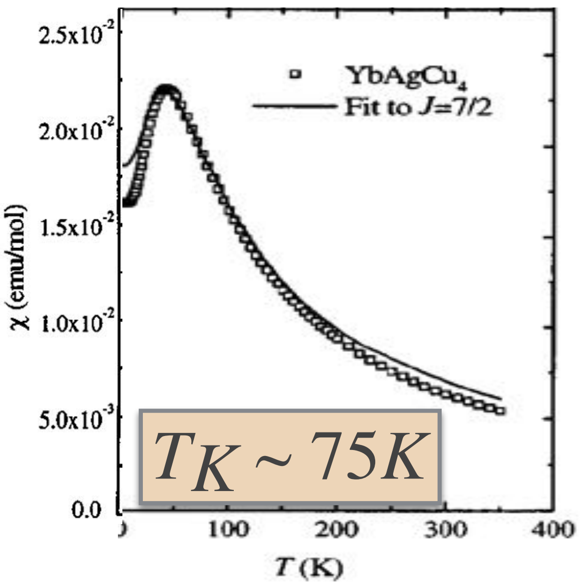




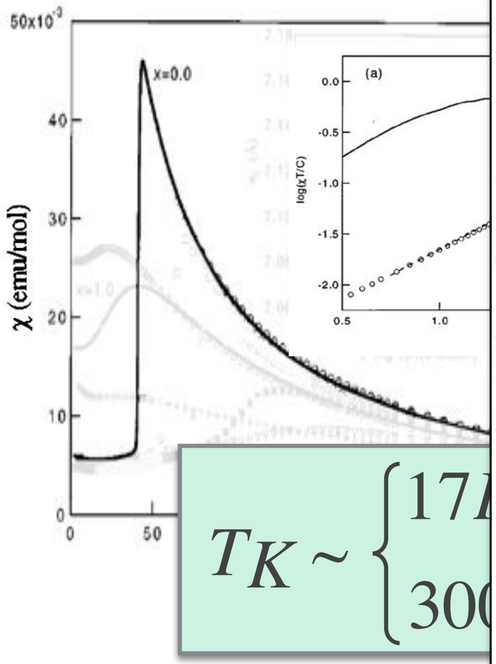
Difficulties arise from mixture of localized and metallic electronic states

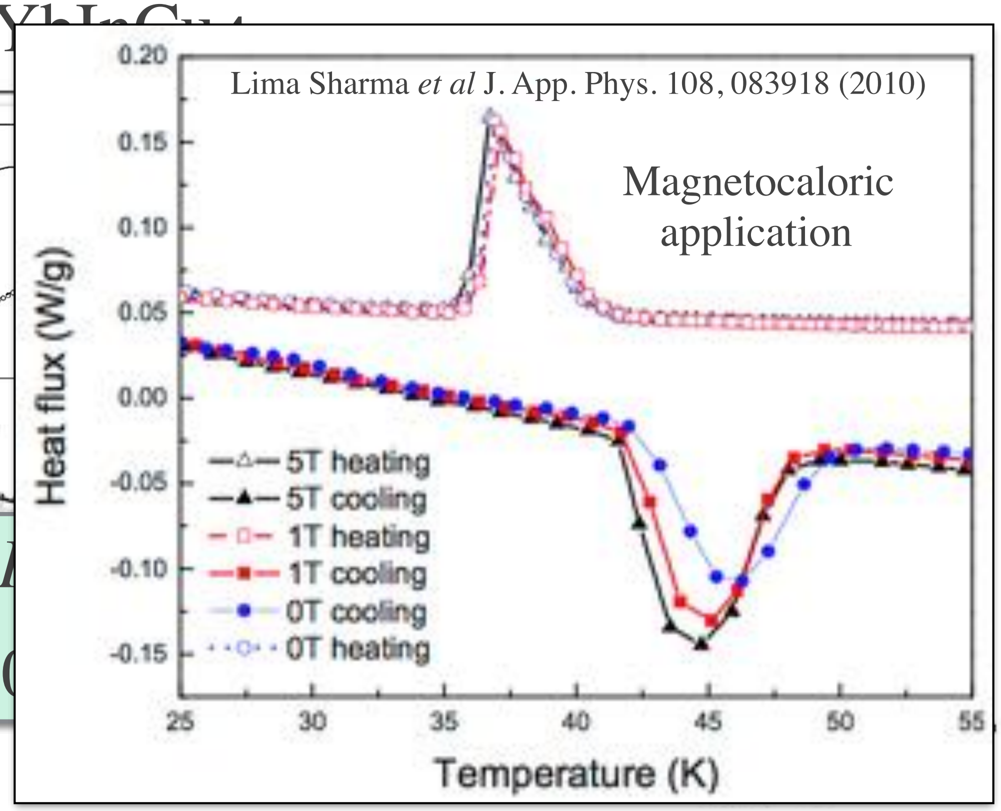
Valence transition in $YbInCu_4$

Heavy fermion $YbAgCu_4$



Kondo-switching $YbInCu_4$





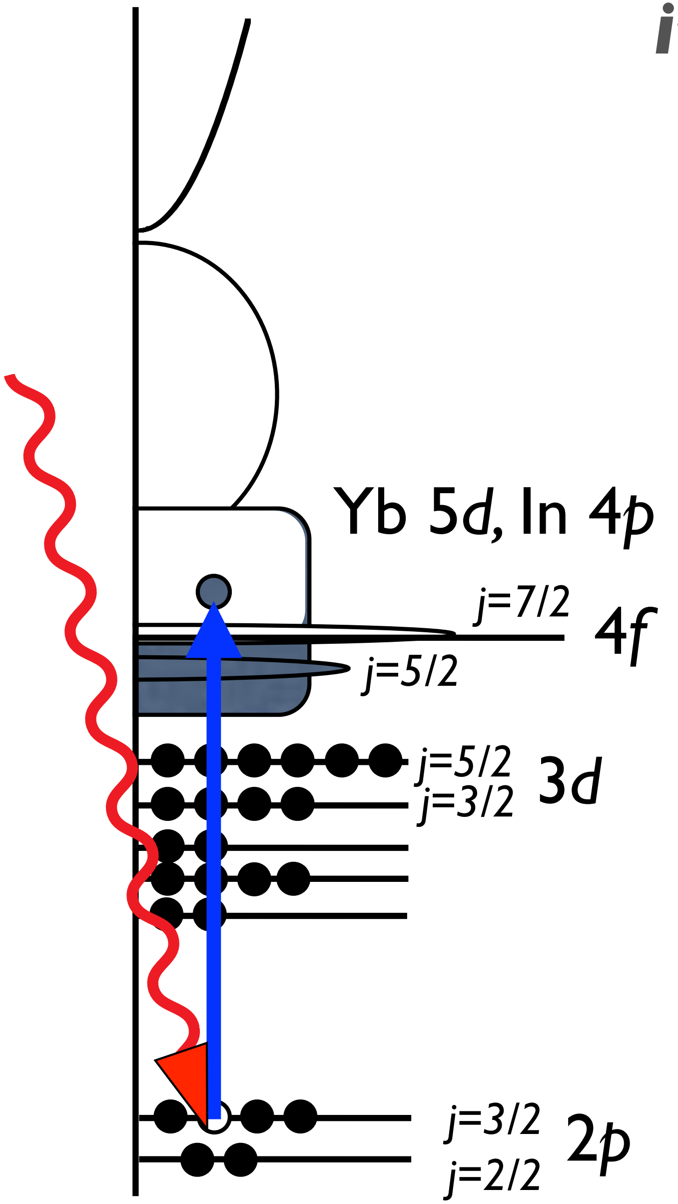
Archetype heavy fermion
Single Kondo temperature
describes magnetic,
valence behavior

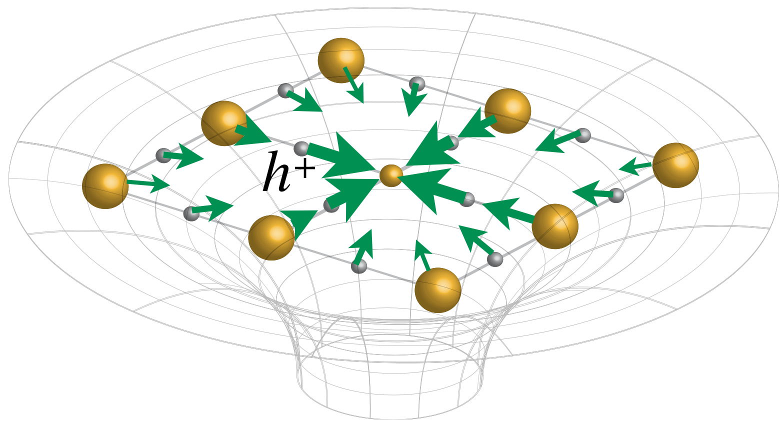
Isostructural transition, *C15b* (cubic) structure
1st order valence transition, T_v=42 K
Changes in n_f , χ support T_K jump scenario
Small 0.15% volume change at transition
Entropy liberated ~R ln(8)
Kondo Volume Collapse scenario insufficient

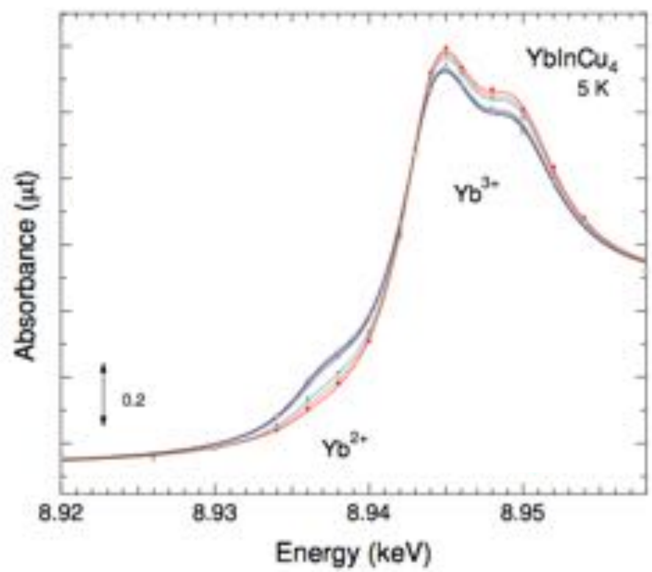
Sarrao *et al*, *PRB*, **59** 6855 (1999)   Cornelius *et al*, *PRB*, **56** 7993 (1997)   Severing *et al*, *Physica B*, **163** 409 (1990)

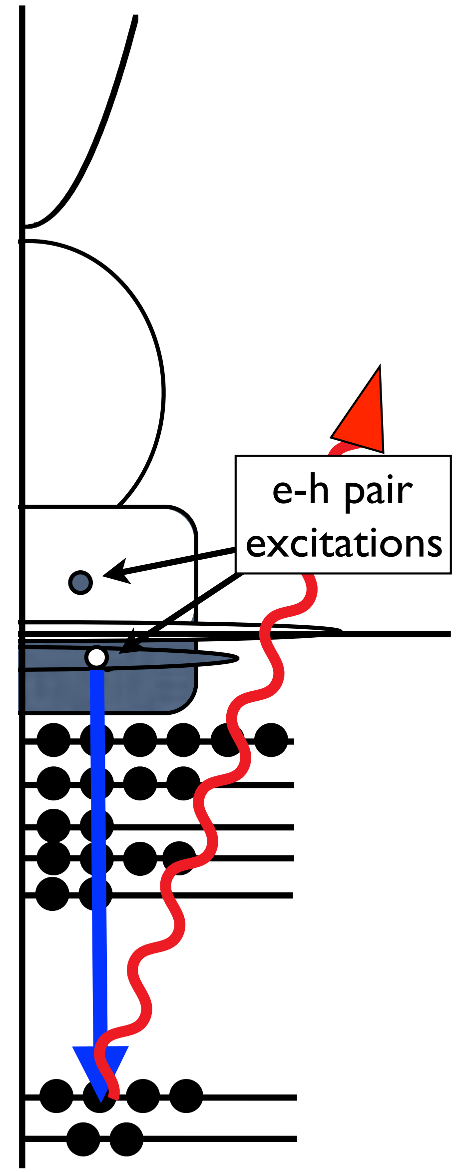
Resonant **I**nelastic **X**-ray **S**cattering

Rare earth ***L edge****: most sensitive to*
itinerant *5d electronic states*

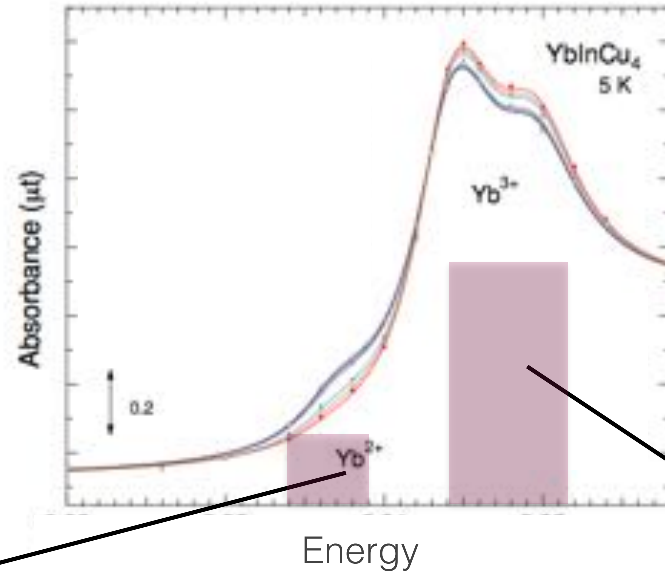


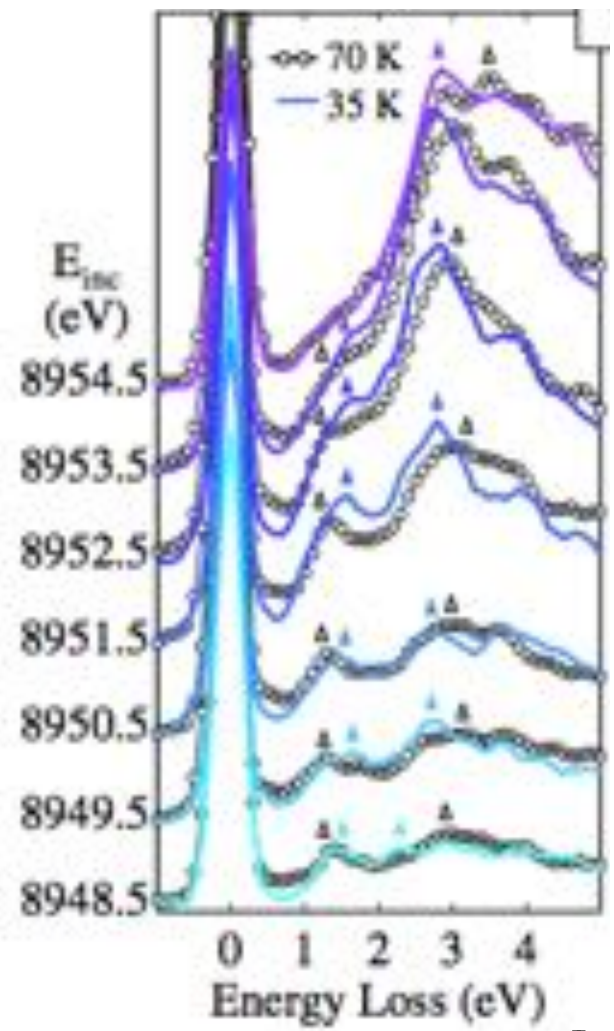




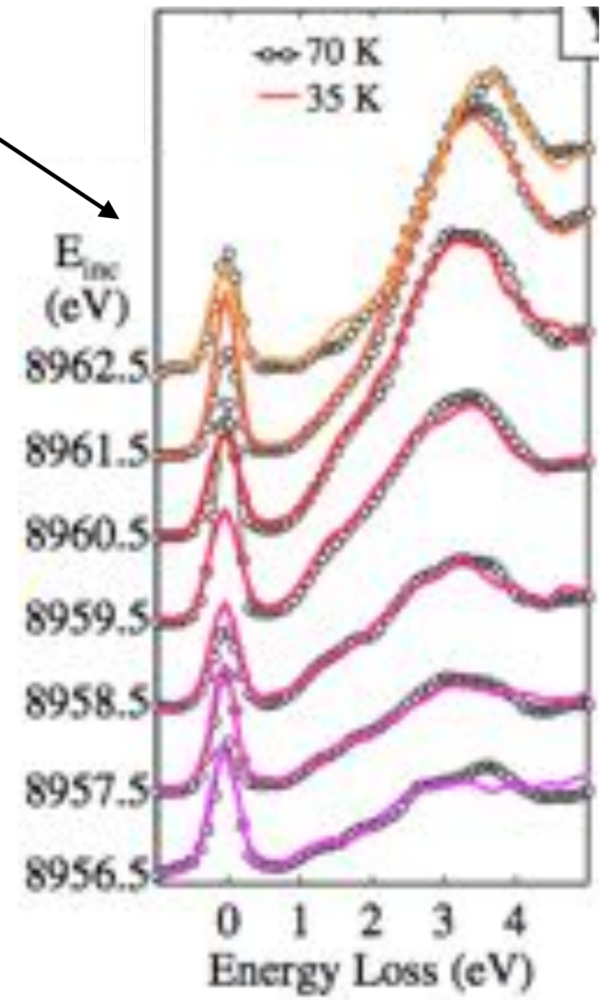


RIXS study of valence transition in $YbInCu_4$



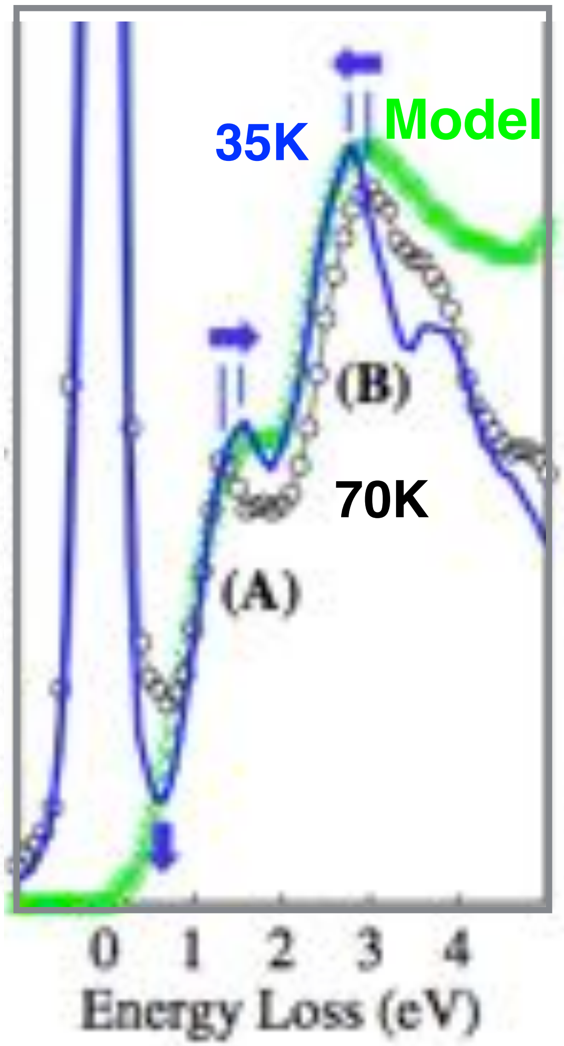


Strong changes with temperature from divalent-excited RIXS



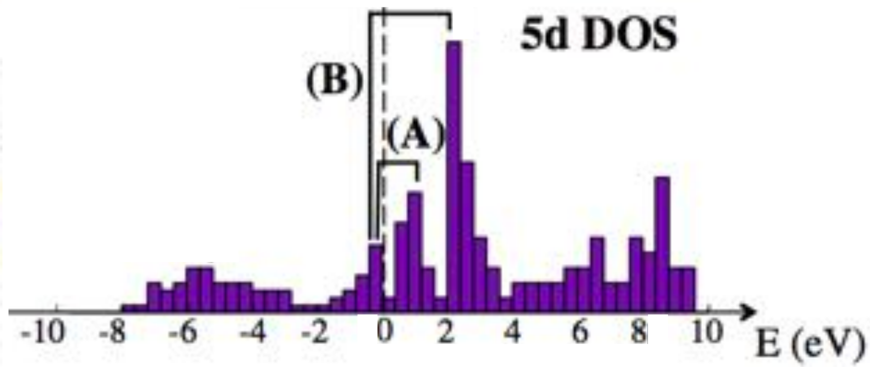
Jarrige, JNH, et al Phys. Rev. Lett. **114**, 126401 (2015)

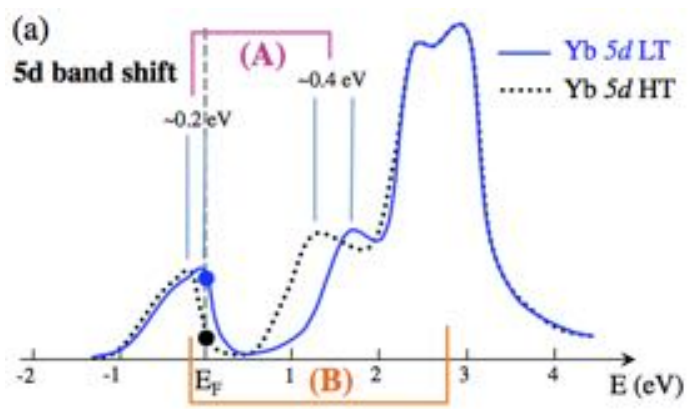
RIXS study of valence transition in $YbInCu_4$

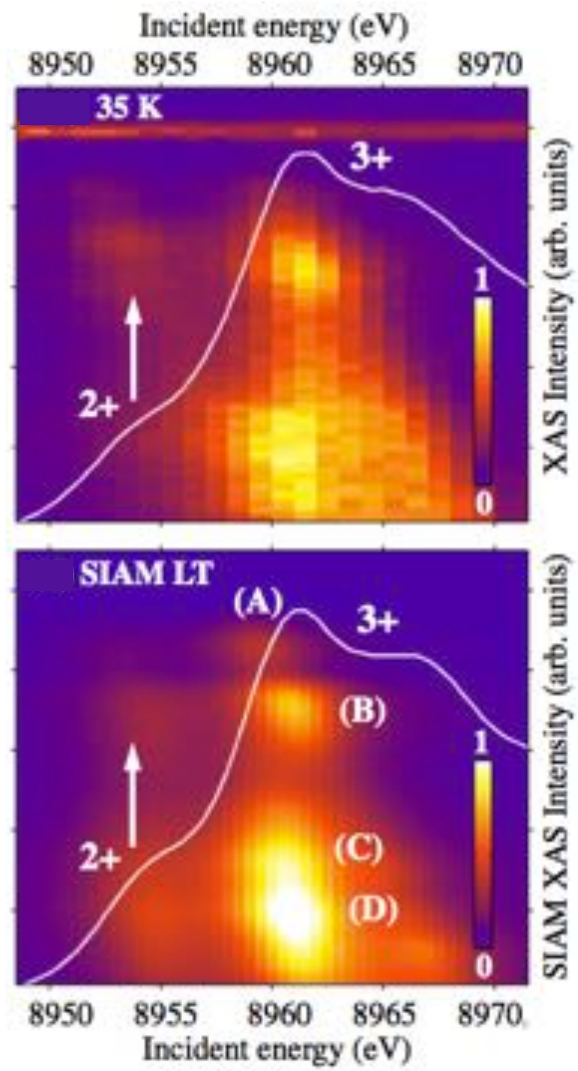


Sum over +2 state to simplify spectra

Compare to electronic calculation LSDA+U







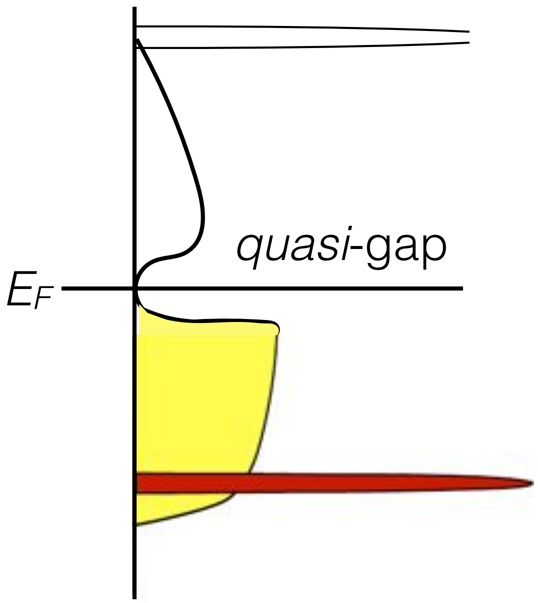
Shift of spectral features imply E_F change

Quasi-gap revealed by +2 RIXS spectra

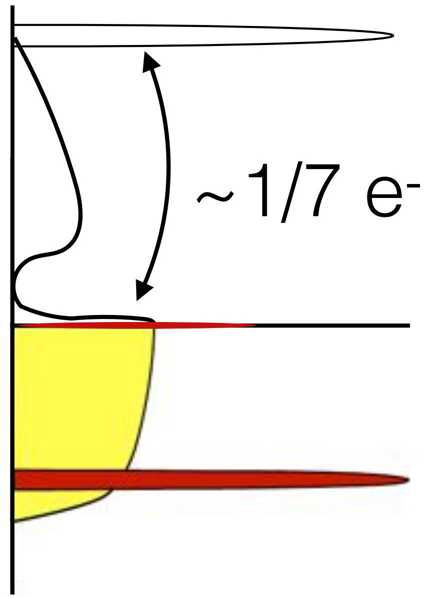
Jarrige, JNH, et al PRL **114**, 126401 (2015)

Calculations by Akio Kotani

High-contrast DOS behind valence transition

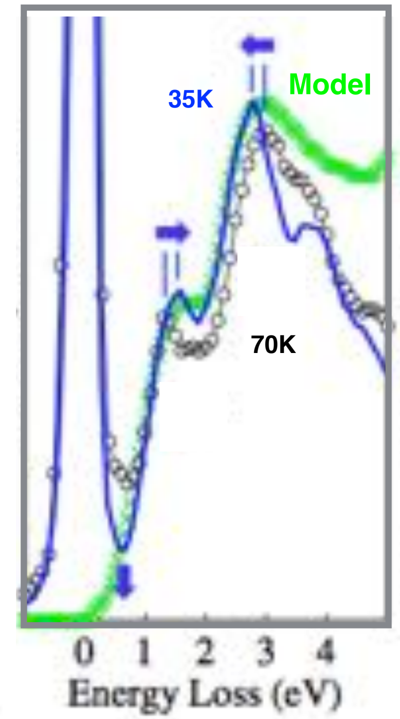


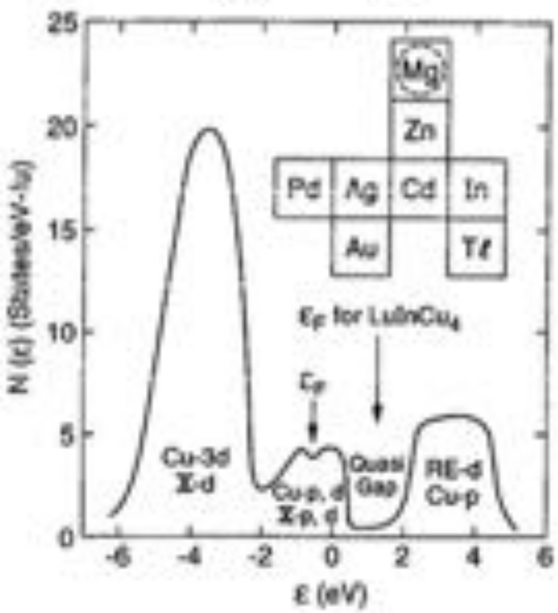
High-T,
poorly conducting,
paramagnetic,
integer valent state



Low-T,
highly conducting,
moment-screened,
mixed valent state

Jarrige, JNH, et al PRL **114**, 126401 (2015)





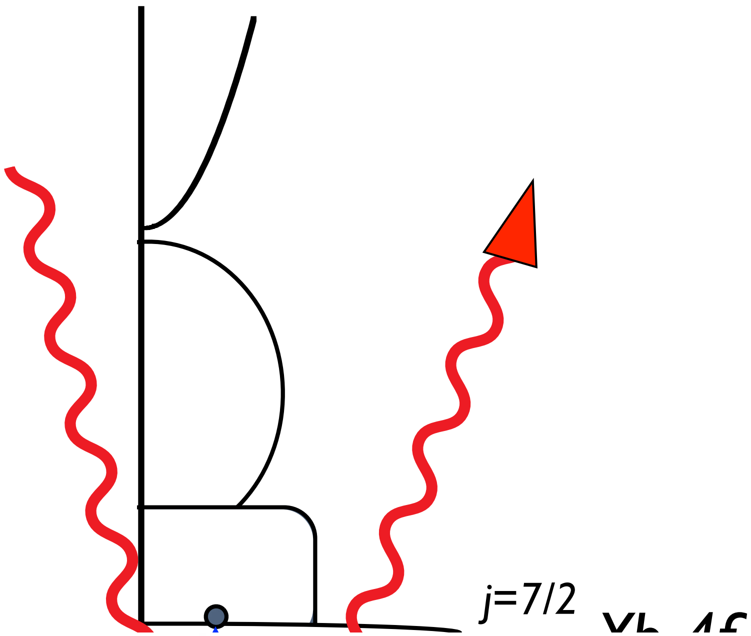
Interacting Falicov/Kimball approach

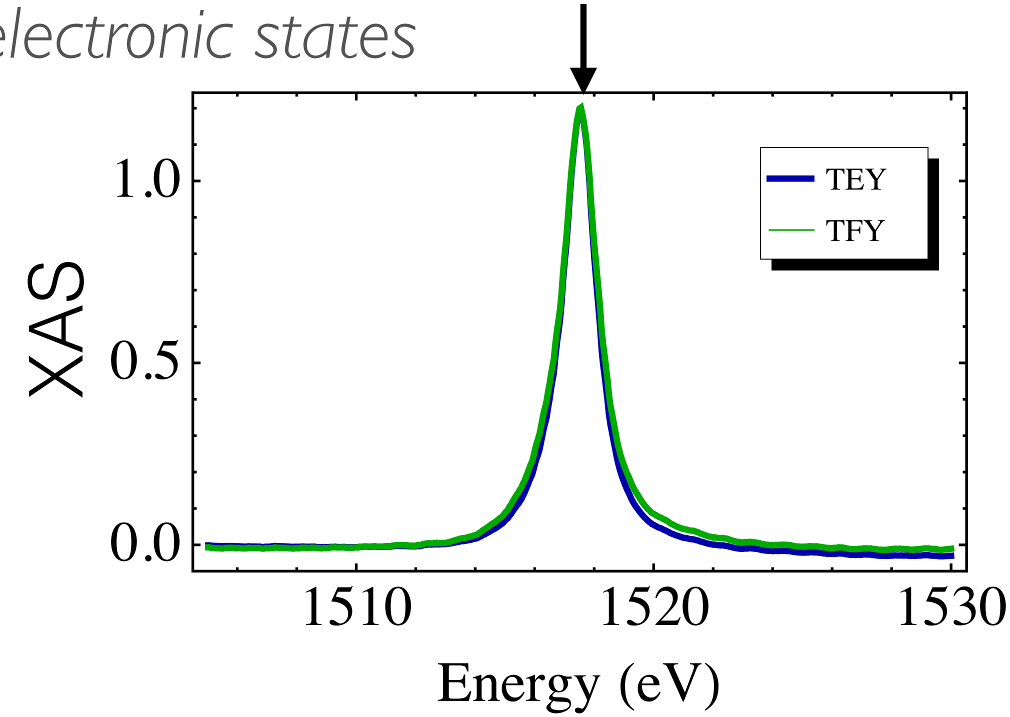
J. K. Freericks and V. Zlatic´, Rev. Mod. Phys. 75, 1333 (2003)

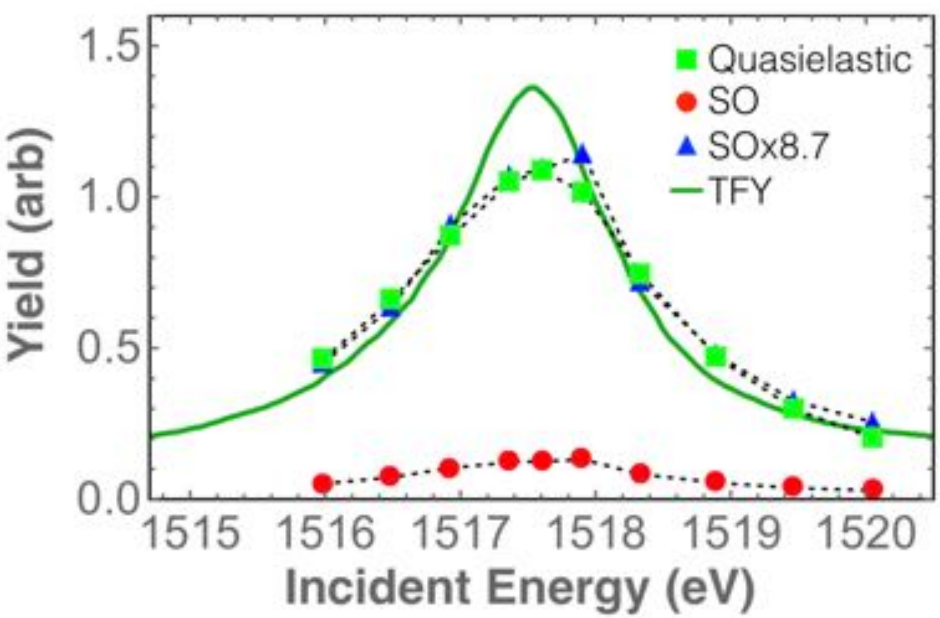
Figueroa, et al, Solid State Comm. 106, 347 (1998)

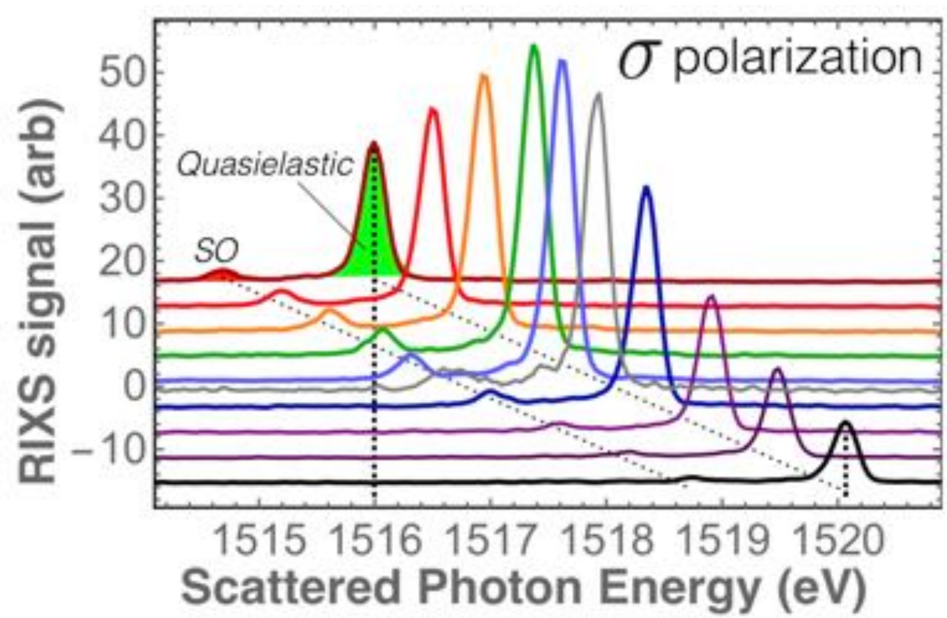
RIXS at a *different* edge

Rare earth ***M edge****: most sensitive to* ***localized*** *4f electronic states*



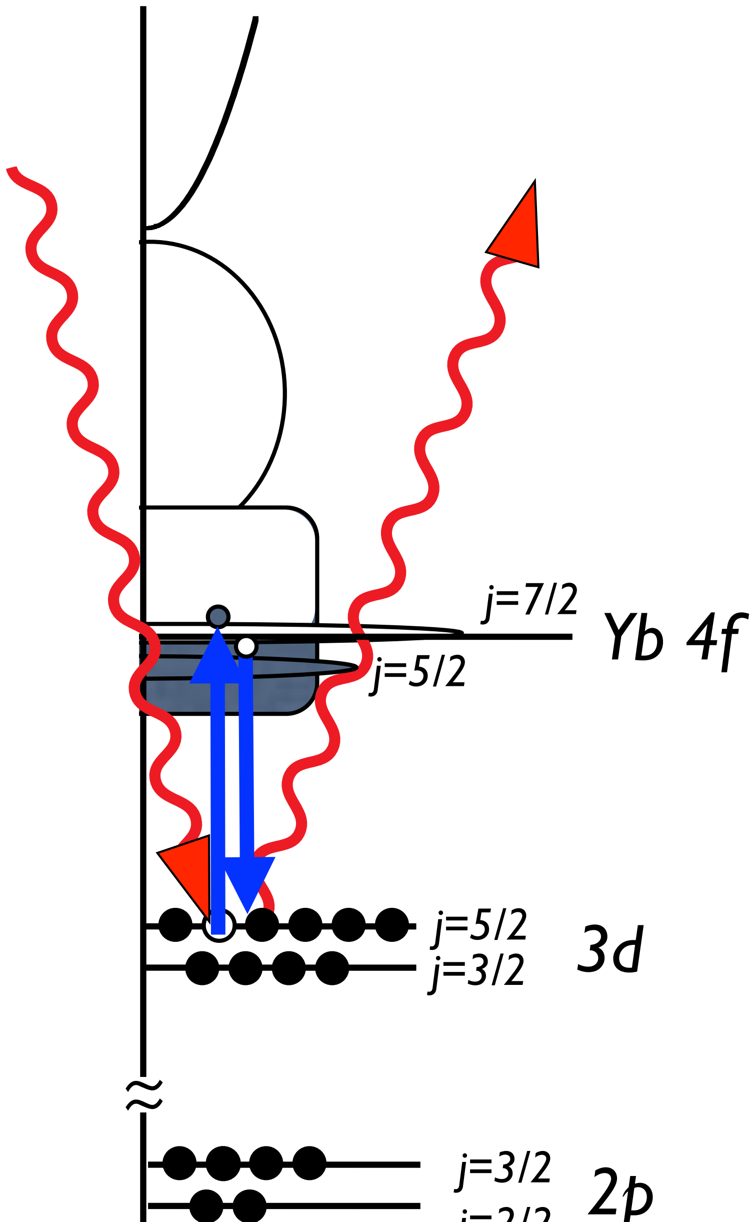


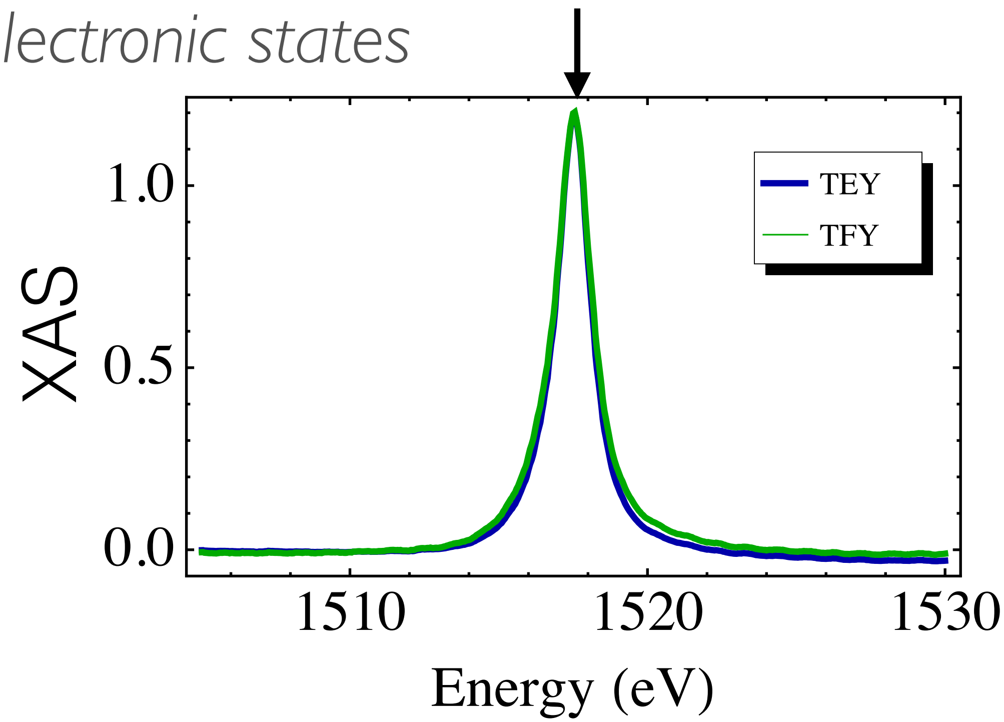


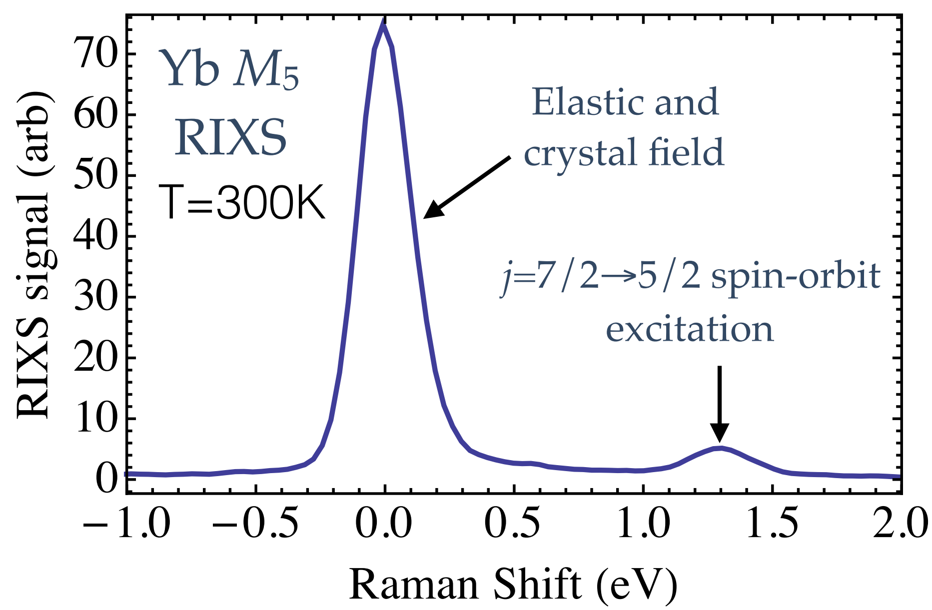


RIXS at a *different* edge

Rare earth ***M edge****: most sensitive to*
localized *4f electronic states*

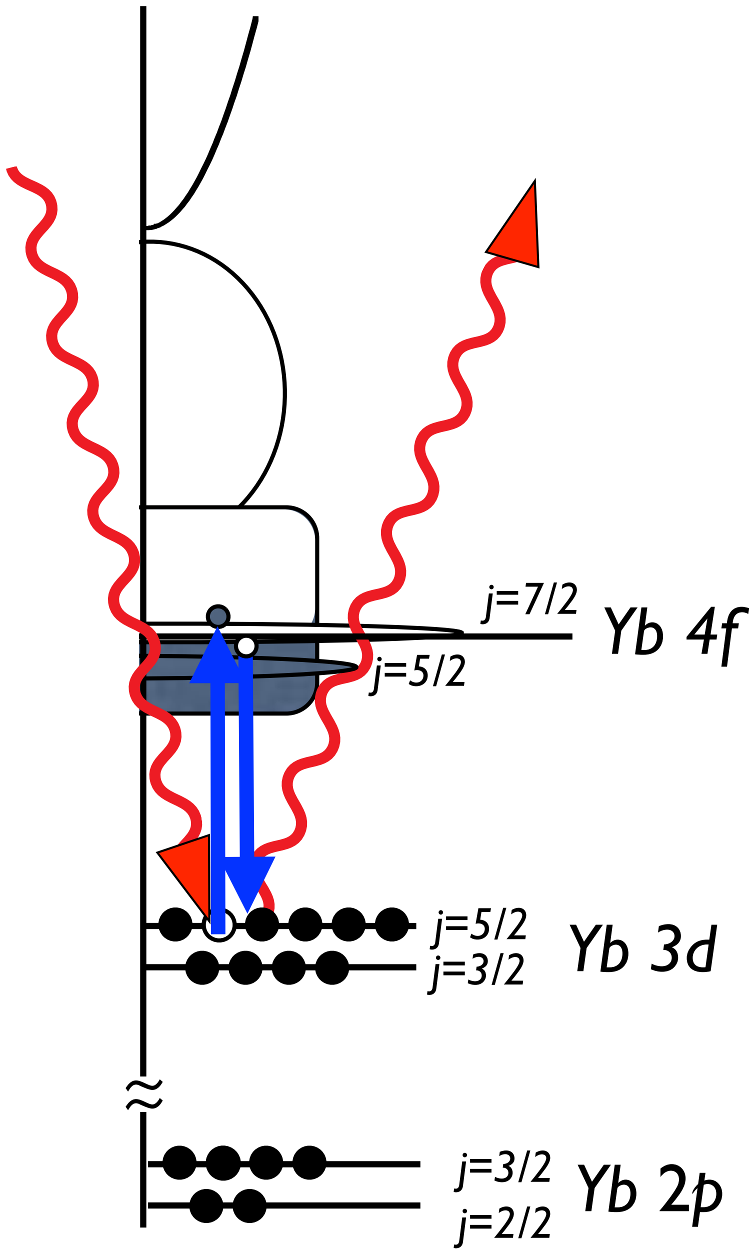


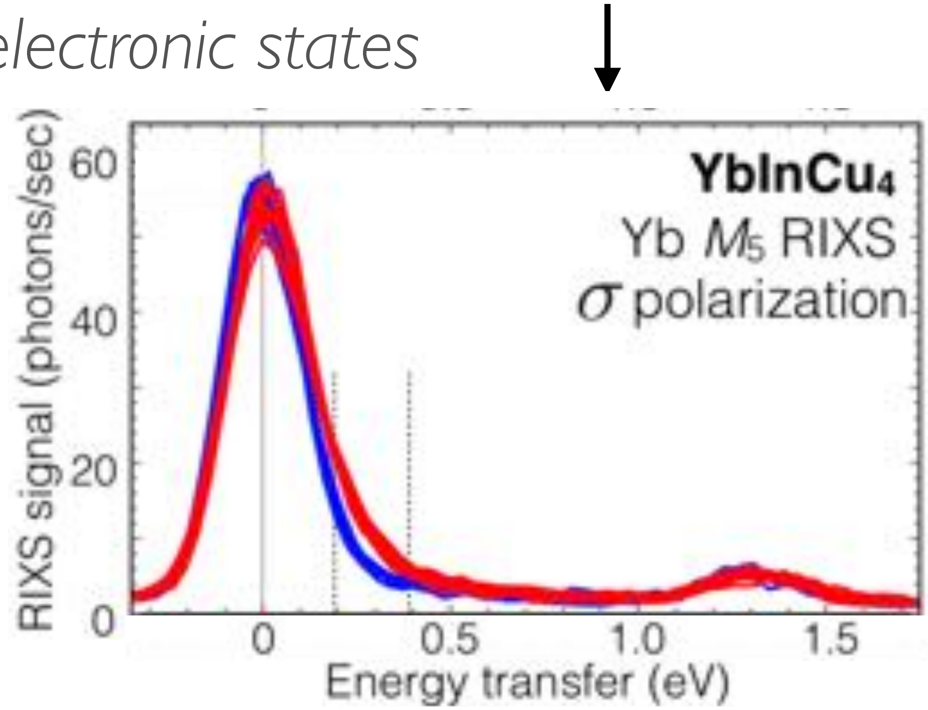


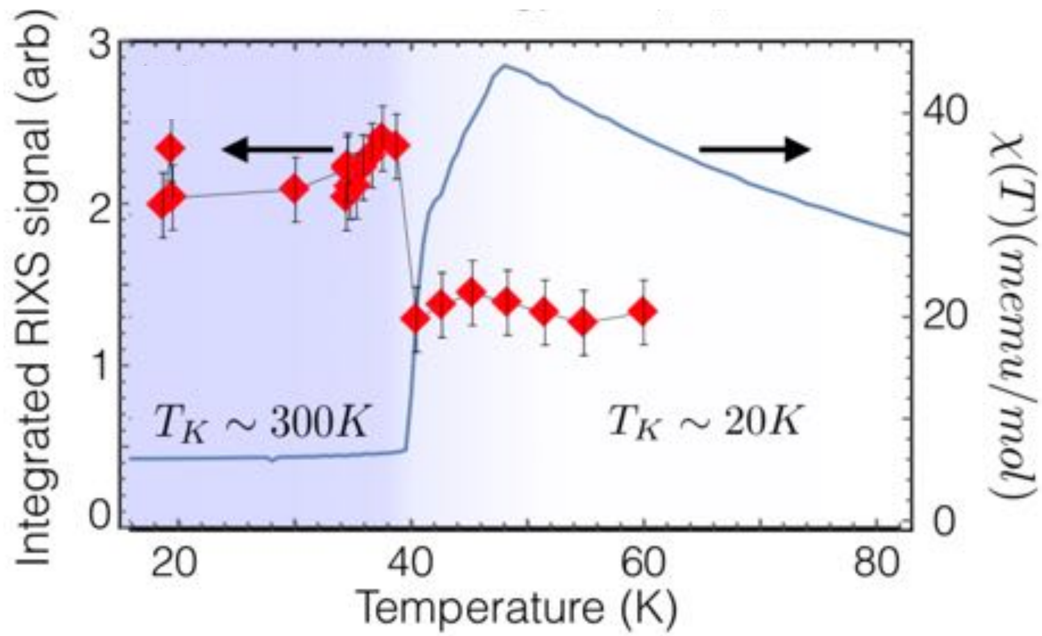


RIXS at a *different* edge

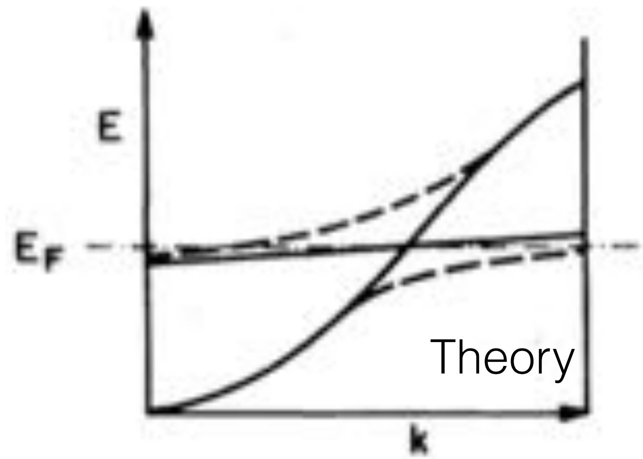
*Rare earth **M edge**: most sensitive to*
***localized** 4f electronic states*

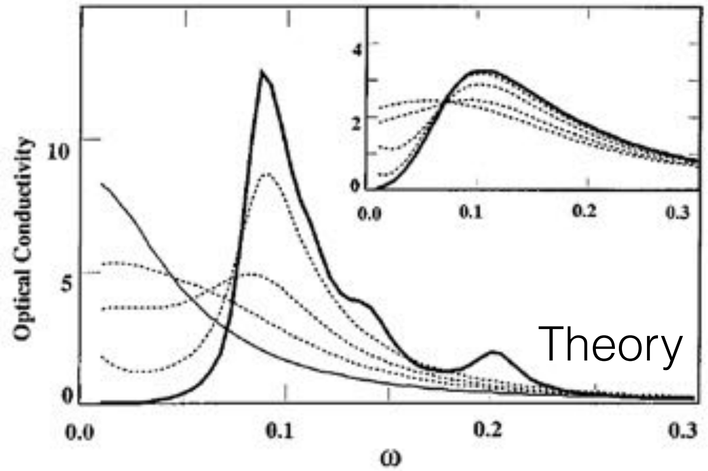


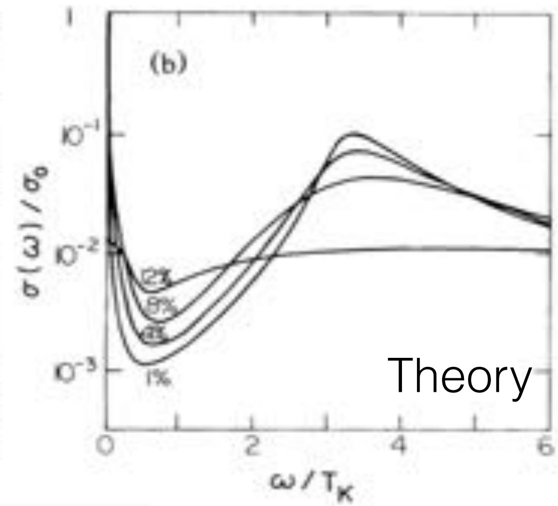


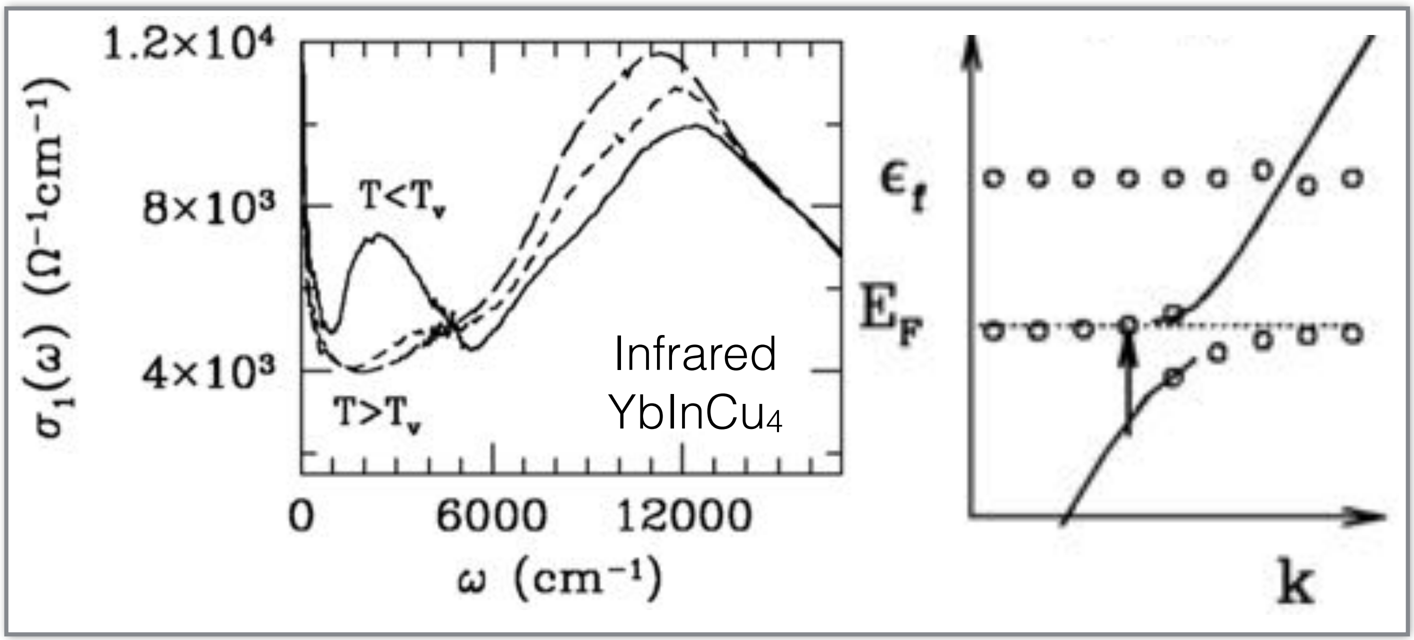


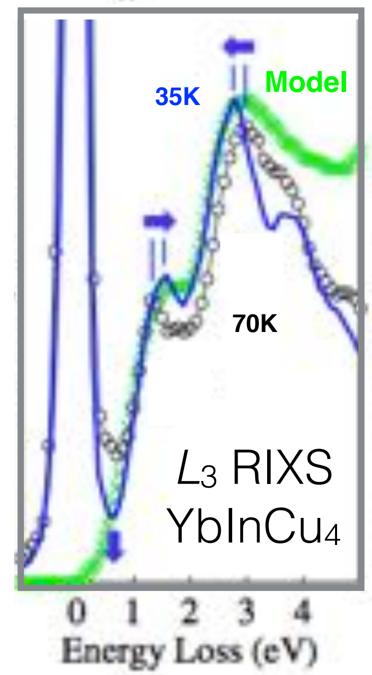
RIXS for studies of the hybridization gap











Garner, JNH, et al PRB **62**, 4778 (2000)
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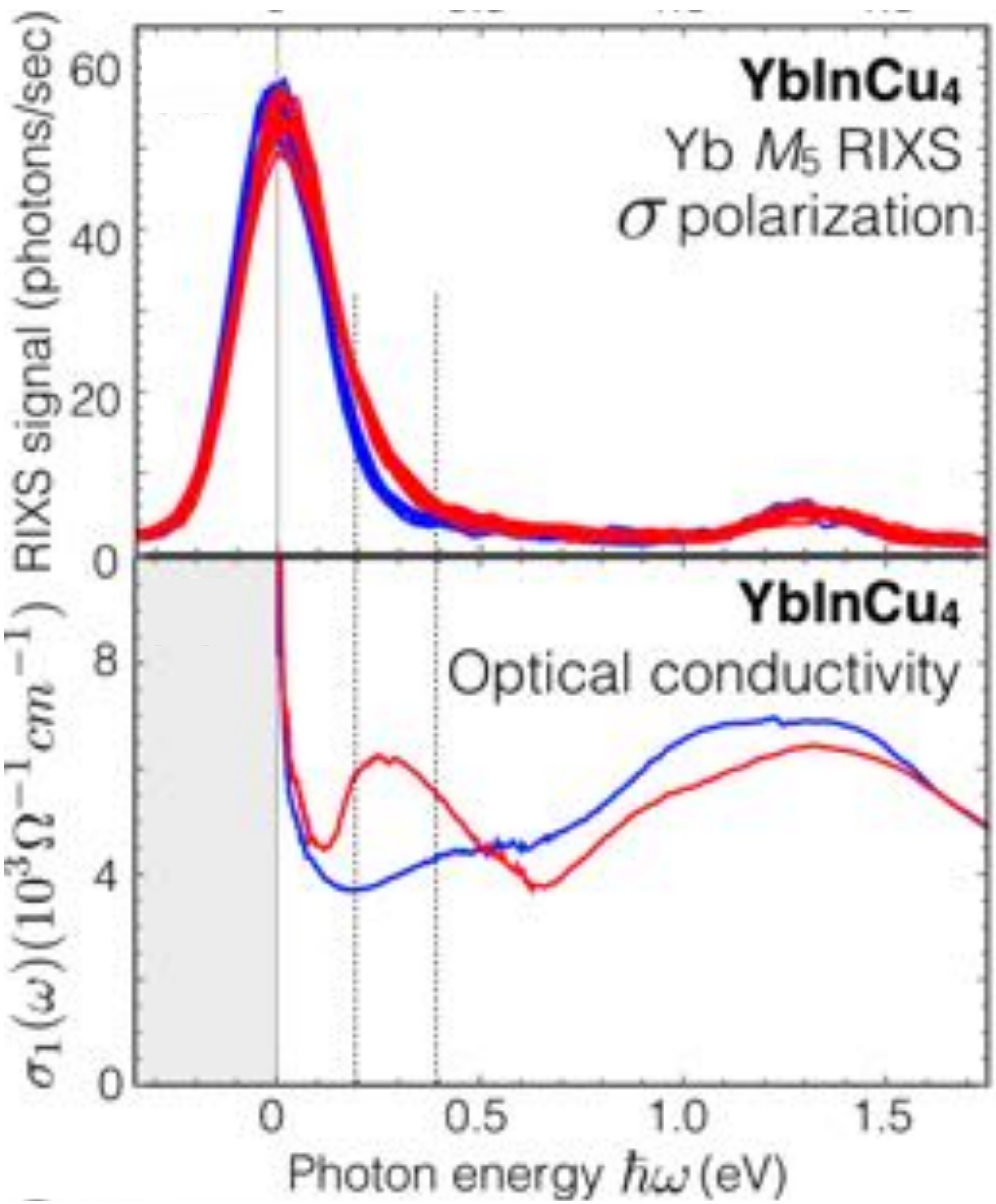
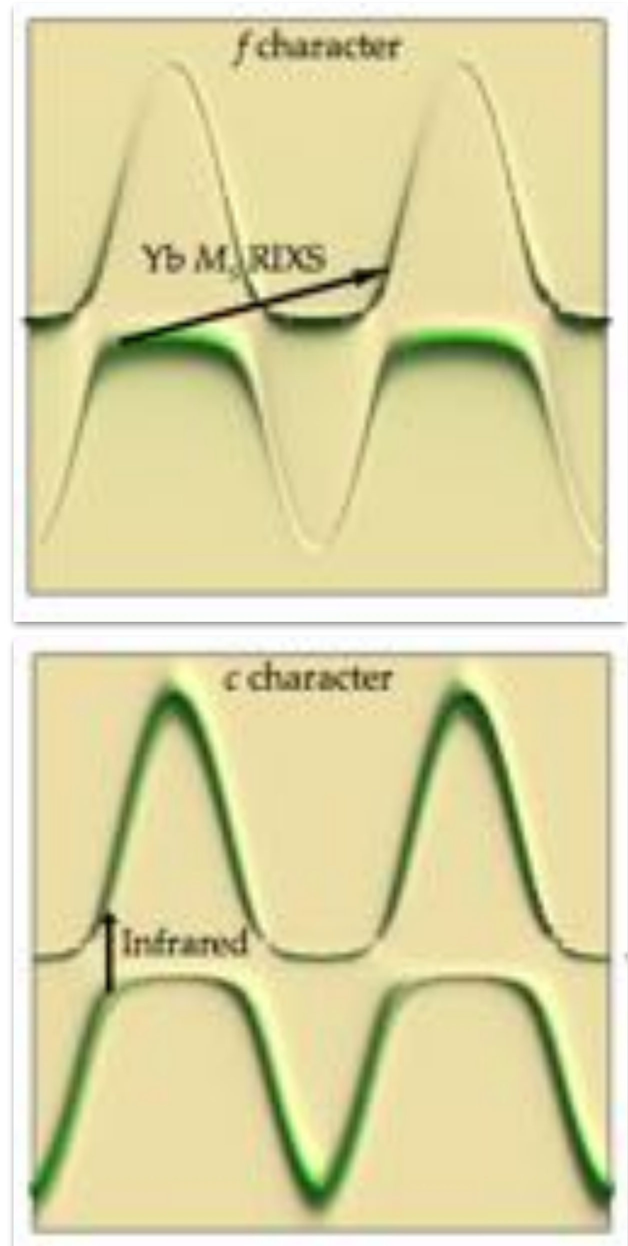
H. Okamura, *et al*. PRB 75, 041101(R) (2007)
Matsunami, *et al*. Phys. Rev. B 87, 165141 (2013)

Georges *et al*. RMP **68**, 13 (1996)
P. Coleman, PRL 59, 1026 (1987)
C. M. Varma, RMP 48, 219 (1976)

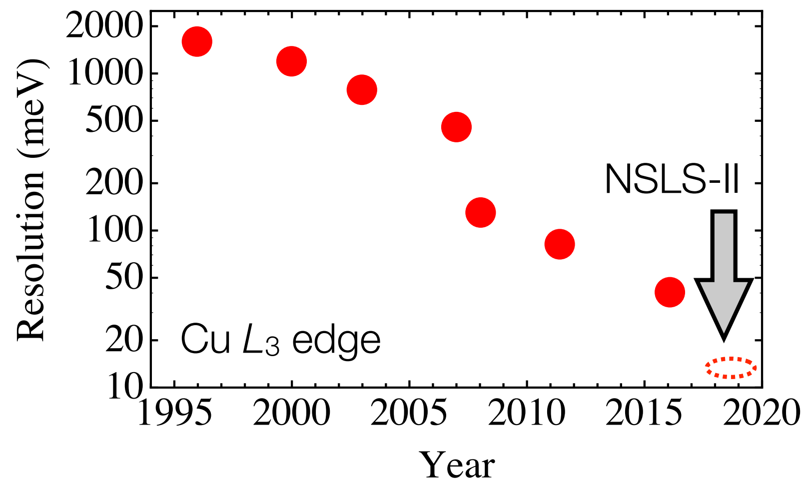
Jarrige, JNH, et al PRL **114**, 126401 (2015)

Infrared optics and *M* edge RIXS





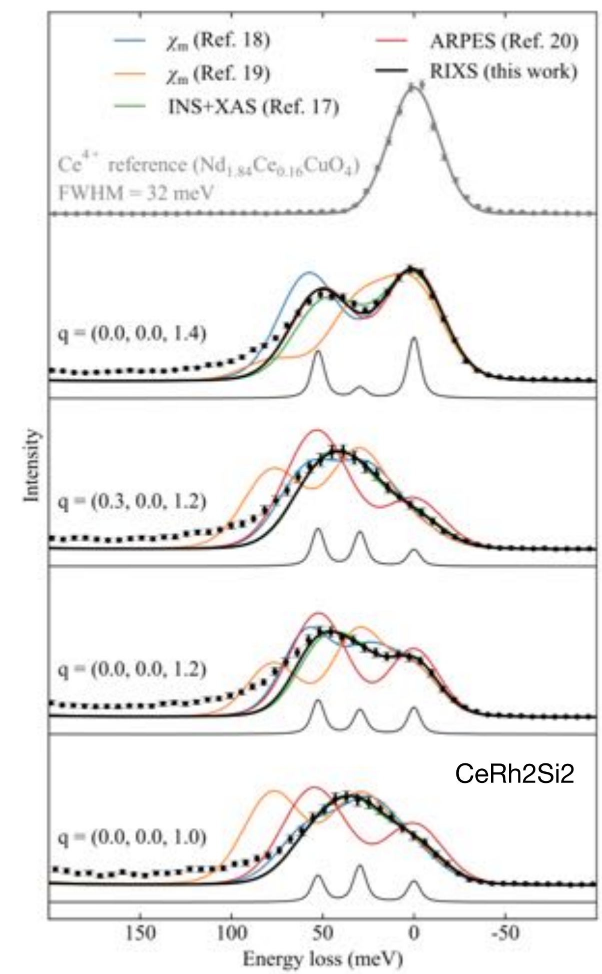
Recent RIXS in *f*-electron materials



- ESRF RIXS gives competitive/ superior determination of crystal field levels

- Requires comparison with detailed *atomic* calculations

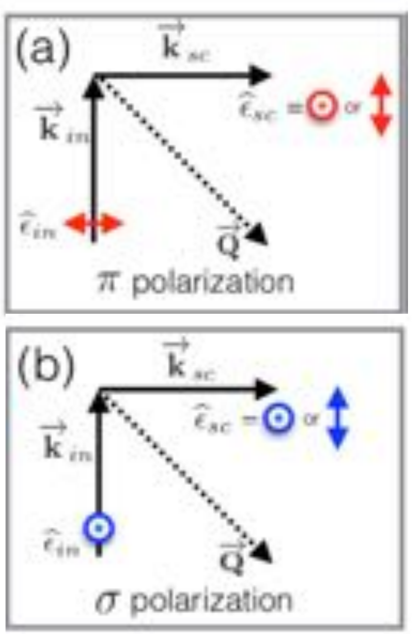
- New resolution promises new dawn of insight into atomic parameters *and* collective behavior

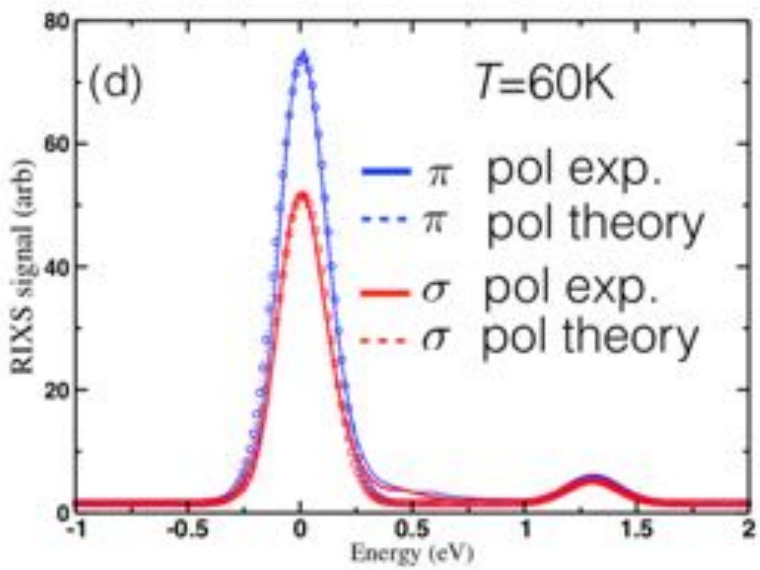


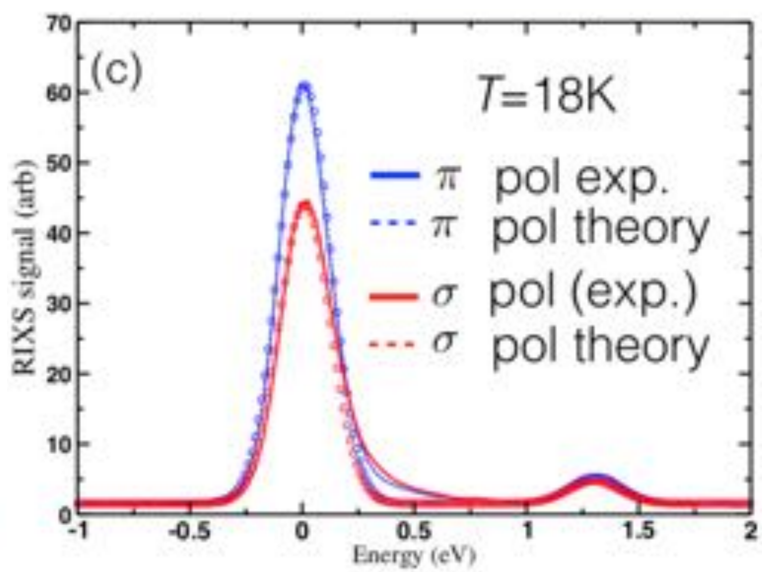
Amorese, et al, PRB 93, 165134 (2016)

Polarization dependent *M*5 RIXS of YbInCu4

- EPU polarization switching permits tests of single-impurity Anderson modeling

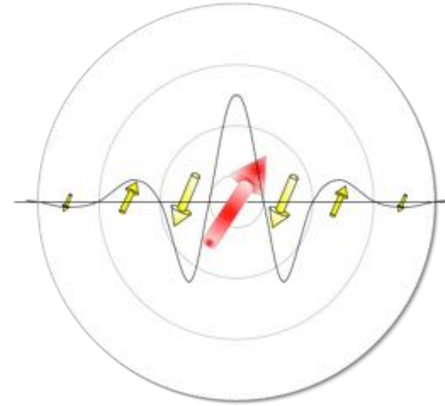




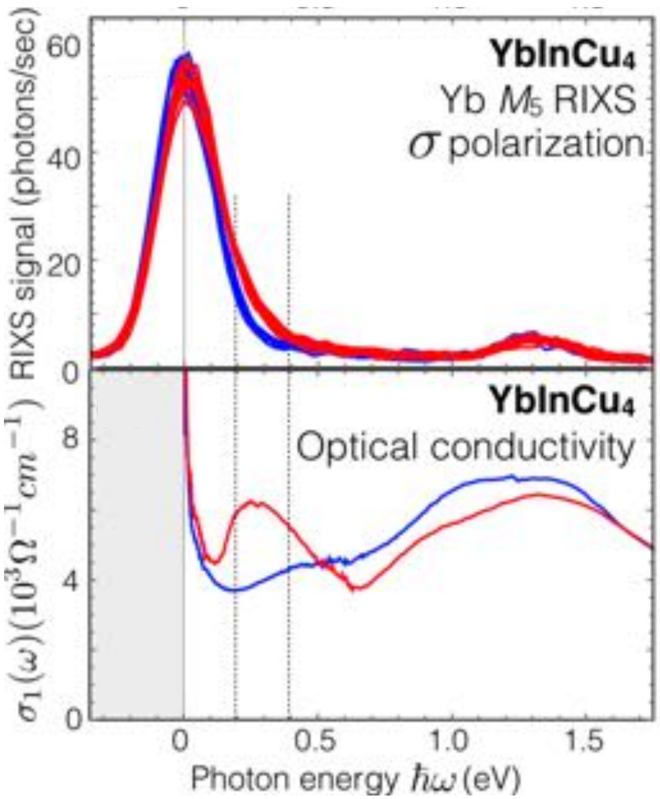


Calculations by Maxim Dzero

Heavy fermion physics in three spectra



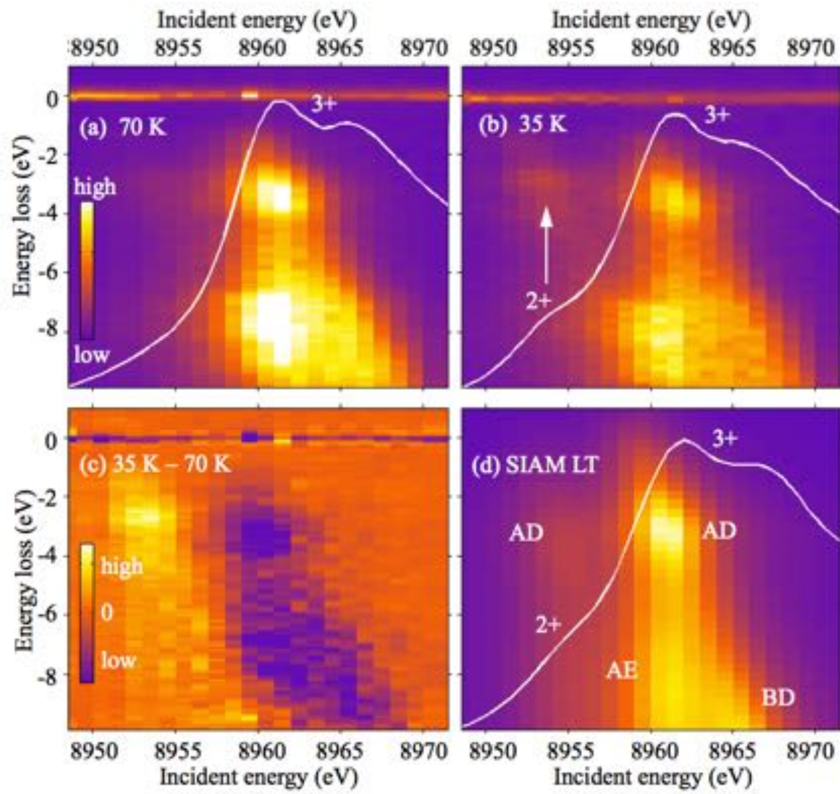
Yb *M* edge RIXS
couples to *f*-electrons

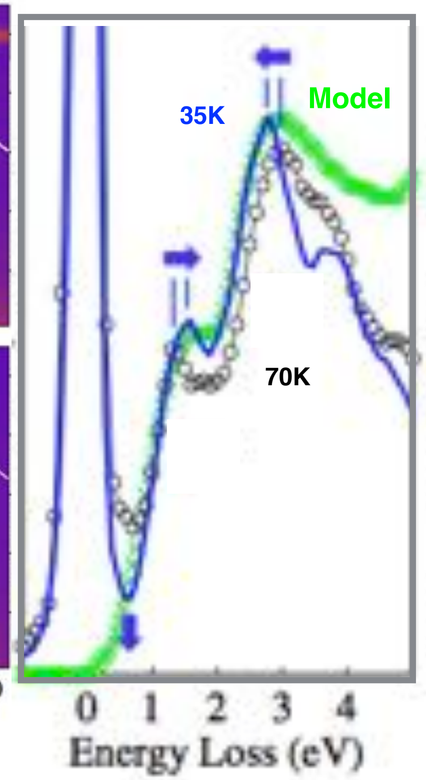


Optical conductivity
couples to charge

Hancock, *et al*. (in review 2018)

Yb *L* edge RIXS
couples to 5*d* itinerant states





Jarrige, JNH, *et al*. (2015)

Optics+RIXS at two different edges shows composite nature of quasiparticles