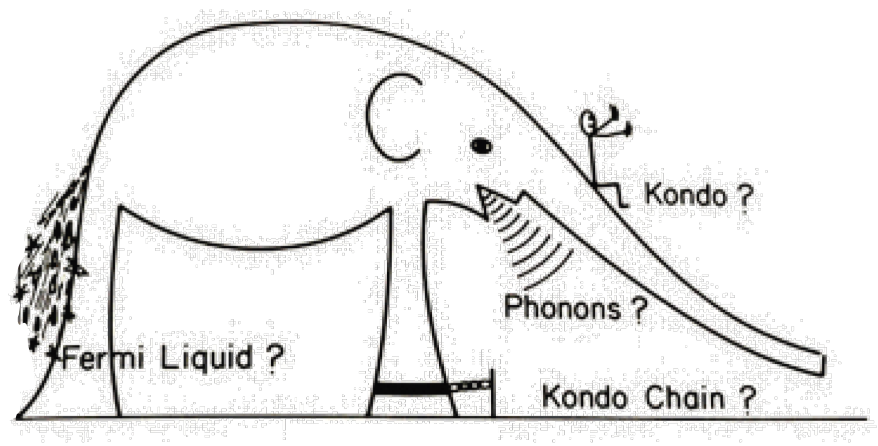
Cooperative valence dynamics in Anderson Lattices observed by resonant inelastic x-ray scattering

Marein Rahn
Los Alamos National Laboratory



LANL
Eric Bauer
Darrin Byler
Jianxin Zhu
Jon Lawrence
Filip Ronning
Marc Janoschek (→PSI)

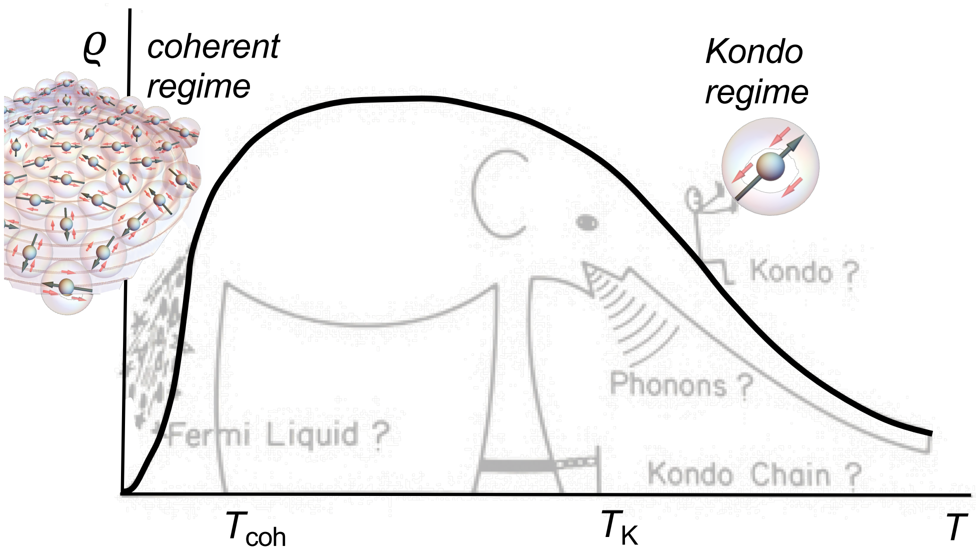
ARPES
Jonathan Denlinger (ALS)
Donghui Lu (SSRL)
Makoto Hashimoto (SSRL)
Fanny Rodolakis Simoes (APS)
Jessica McChesney (APS)
Emile Rienks (HZB)

RIXS
Kurt Kummer (ESRF)
Andrea Amorese (U Cologne)

REXS
Daniel Mazzone (BNL)

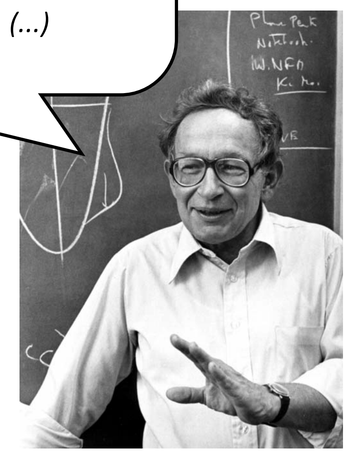
n Lattices
scattering
Marein Rahn
l Laboratory

see P. Coleman in “PWA90” (World Scientific)



*“What you really have here is a **lattice full of these objects that fluctuate back and forth from one valence to another**. There are the phonons, there is the fact that the electrons fluctuate by tossing electrons into the* d *level on the next site which can then go down into the* f *levels on yet another site. So the **things which toss the valence back and forth are definitely coupled between one site and another**. (...) This is an extremely hard problem. ”*

- P. W. Anderson
Rochester Conference on Mixed Valence (1976)



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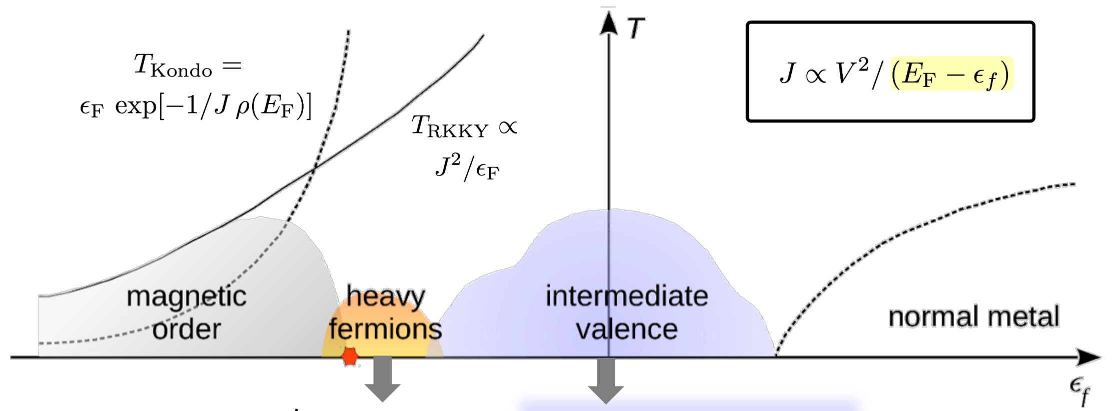
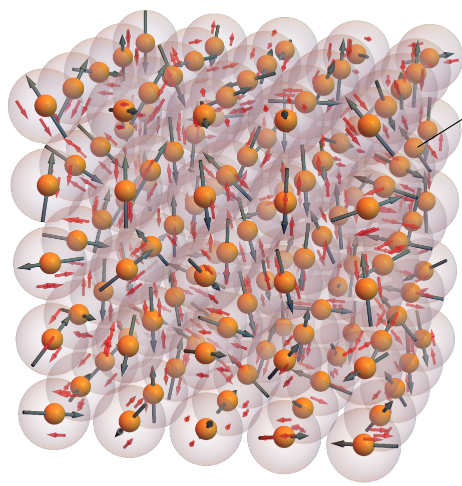
D.I. Khomskii, Cambridge University Press (2010)



- low symmetry
- CEF splitting
- AFM correlations
- heavy FL

- high symmetry
- $T_K \gg T_{CEF}$
- no AFM correlations
- *moderately* heavy FL

$CePd_3$ typical intermediate valence compound



$$|\psi\rangle = a_1\,|f^1\rangle + a_0\,|f^0\rangle$$

	Ce^{3+}	Ce^{4+}
S	1/2	0
L	3	0
J	5/2	0
μ_{eff}	2.5 μ_B	0 μ_B

- large Kondo energy scale $T_K \approx$ 50 meV / 600 K
  (*quenching of magnetic moments*)
- coherence energy scale $T_{coh} \approx$ 12 meV / 150 K
  (*crossover to Anderson Lattice*)
- effective valence: **$Ce^{3.25+}$ $4f^{0.75}$** ,
  not strongly T-dependent below RT

Questions

What is the microscopic mechanism of the quantum coherent state?
(relevance of inter-site fluctuations, phonons, ... ?)

How do the relevant energy scales arise?
(T_K T_{coh} T_{RKKY})

How are elementary excitations (charge, spin) being renormalized?

How does this break down when magnetism is approached?

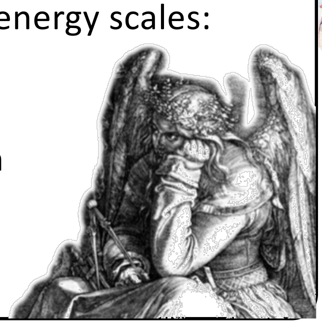
(How) are the resulting quantum fluctuations driving emergent phases?

Challenges

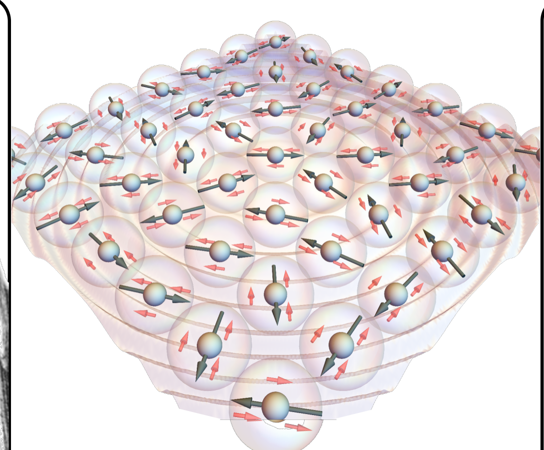
theory

need to take into account electronic and magnetic, local and itinerant degrees of freedom, on disparate energy scales:

- on-site correlation
- inter-site correlation
- lattice coupling (?)



Dürer/Grover (CNLS 2019)



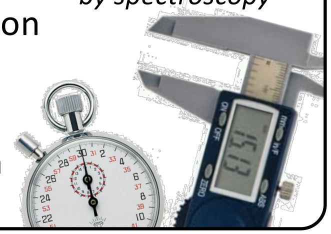
experiment

Interest:

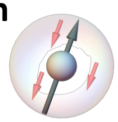
- *f* level energies
- widths and splitting
- occupation numbers
- character and dispersion of quasiparticle states

often...
not directly measured by bulk methods,
not well resolved by spectroscopy

time- and length scale of the probe must match



Anderson Impurity Model



Universality

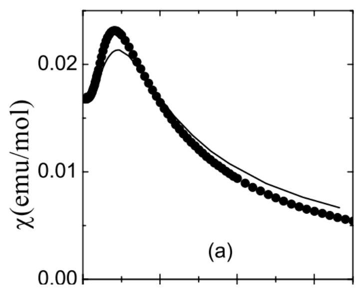
$\chi(T)$
Pauli ↔ Curie-Weiss

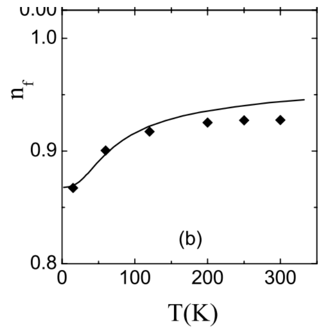
$n_f(T)$ [XAS]
non-integral ↔ integral

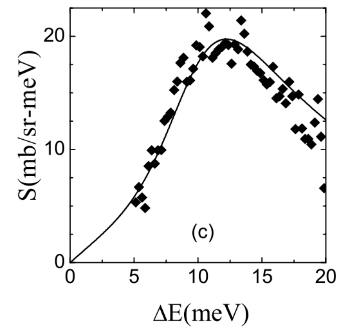
$\chi''(\omega)$ [INS]
gapped ↔ broadened quasiel.

$YbAgCu_4$

W = 0.865
E_f = -0.4485
V = 0.148
T_K = 95

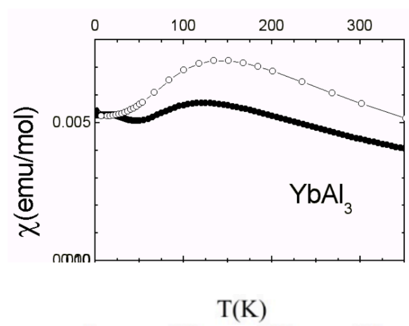


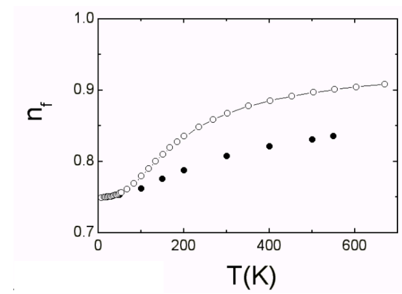


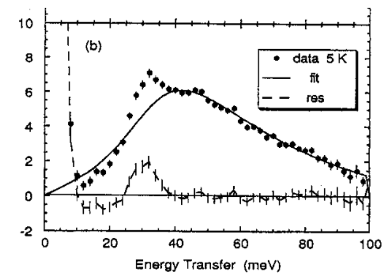


$YbAl_3$

W = 4.33eV
E_f = -0.58264eV
V = 0.3425eV
T_K = 670K

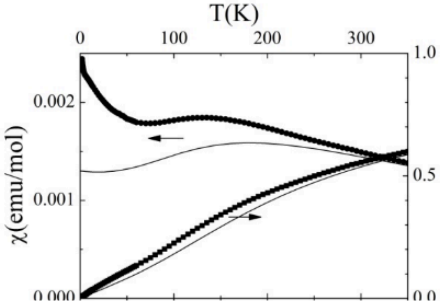


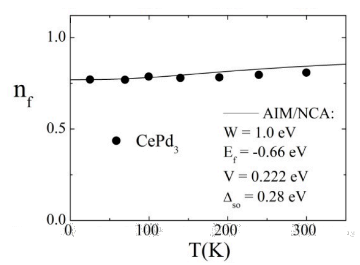


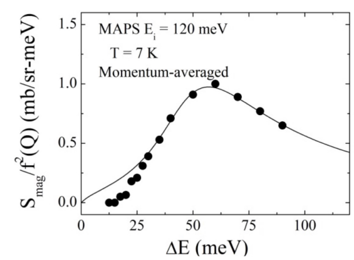


$CePd_3$

W = 1.0 eV
E_f = -0.66 eV
V = 0.222 eV
Δ_{so} = 0.28 eV





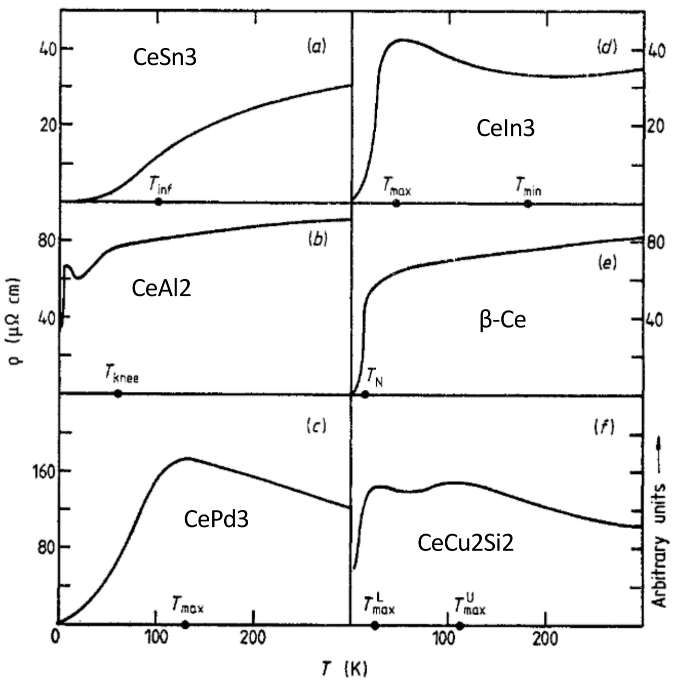


Fanelli *et al.*, J. Phys.: Cond. Mat. 26 (2014)
Murani PRB 50, 9882 (1994)
J. Lawrence, Mod. Phys. Lett. 22, 1273 (2008)
Cornelius *et al.*, PRL 88, 117201 (2002)

Kondo ***lattice*** *properties*

resistivity $\varrho(T)$

observe Fermi-liquid $\varrho(T) \propto T$
↯ AIM predicts finite $\varrho(0)$

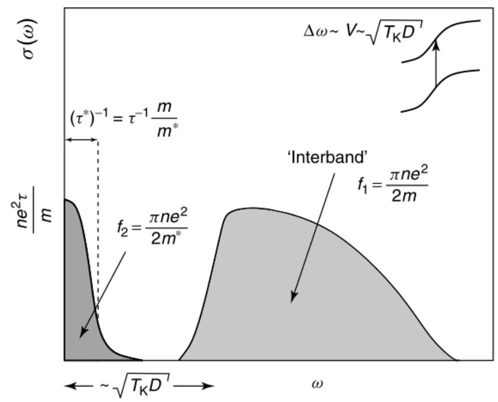


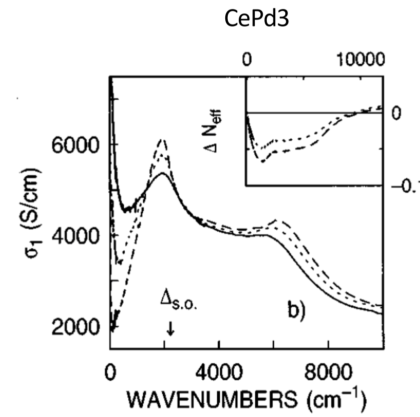
- low temperature resistivity: depends on periodicity of scattering potential → strong effect of lattice coherence
- identify coherence temperature T_{coh}

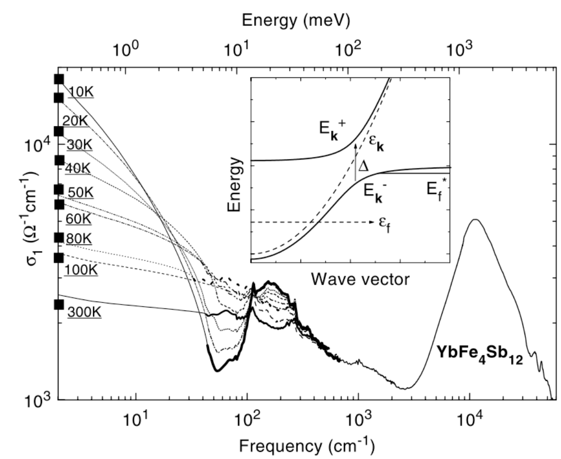
J. Lawrence, Rep. Prog. Phys. 44 (1981)

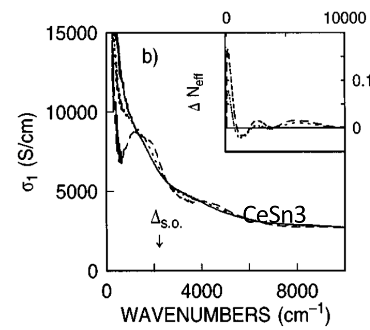
optical conductivity σ(ω)

- Drude peak narrows
- direct interband transitions emerge





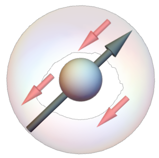




P. Coleman, *Heavy Fermions* (2007)
Dordevič *et al.*, PRL 86,684 (2001)

Burch *et al.*, PRB 75, 054523 (2007)

Experimental probes *local* vs. *lattice properties*

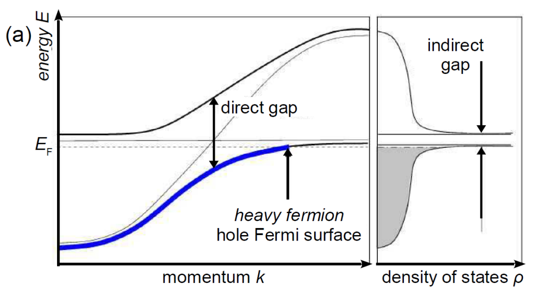


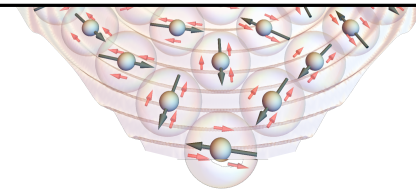
local probes

dominated by localized spin-fluctuations

universally modeled Anderson Impurity Model

- magnetic susceptibility $\chi(T)$
- effective *f*-occupation $n_f(T)$
- specific heat $C(T)$
- momentum-averaged dynamic magnetic susceptibility $\chi''(\omega)$



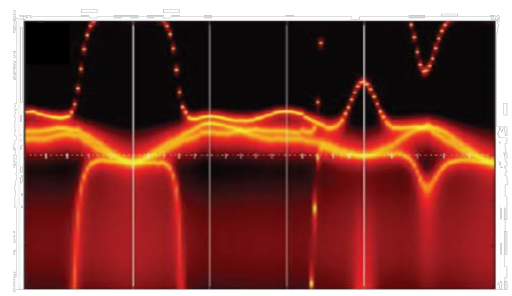


lattice probes

sensitive to lattice periodicity and low-*E dynamics*

impurity models fail

- quantum oscillations
- optical conductivity $\sigma(\omega)$
- resistivity $\varrho(T)$
- ***momentum-resolved spectroscopies***
  - **ARPES**
  - **INS**
  - ***RIXS ?***



Universality of AIM: Lawrence, PRB **63**, 054427 (2001)

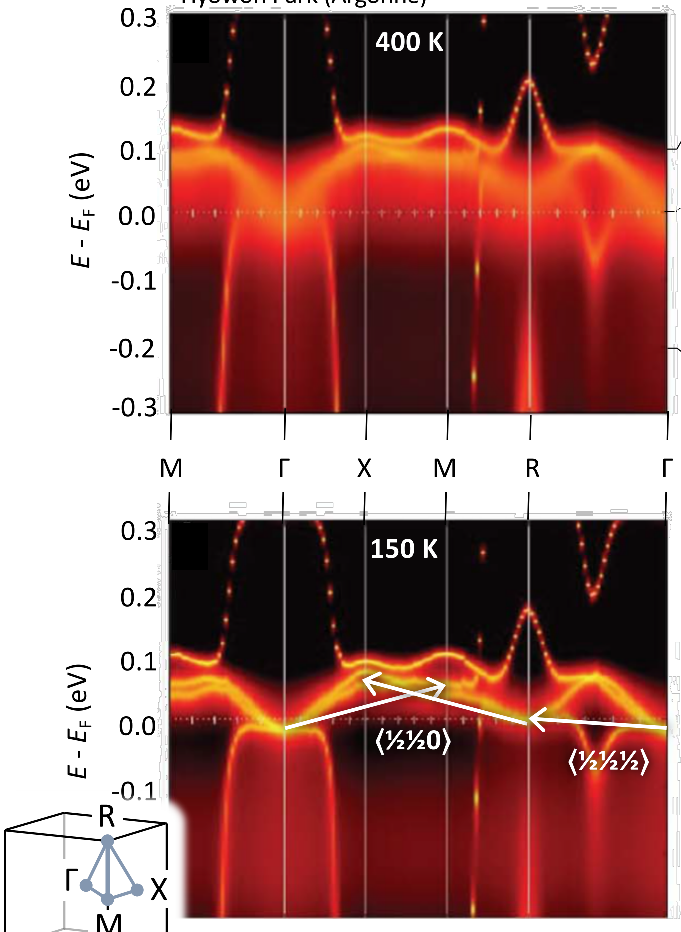
IV DMFT: Goremychkin, Science **359**, 186 (2018)

Coherent quasiparticle excitations in $CePd_3$

Fanelli, J. Phys.: Cond. Mat. 26 (2014)

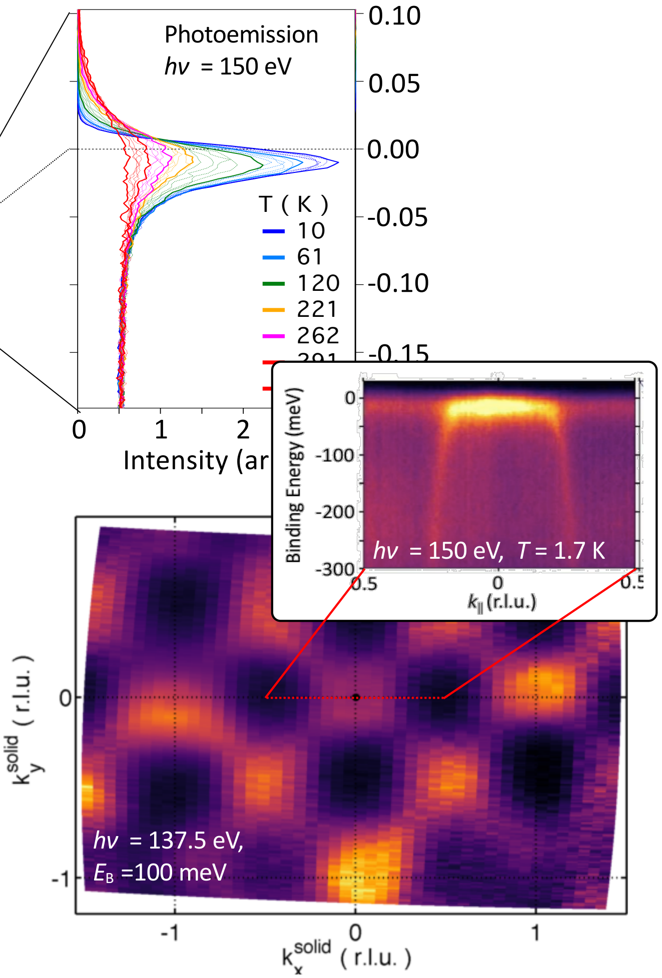
DFT+DMFT

Hyowon Park (Argonne)

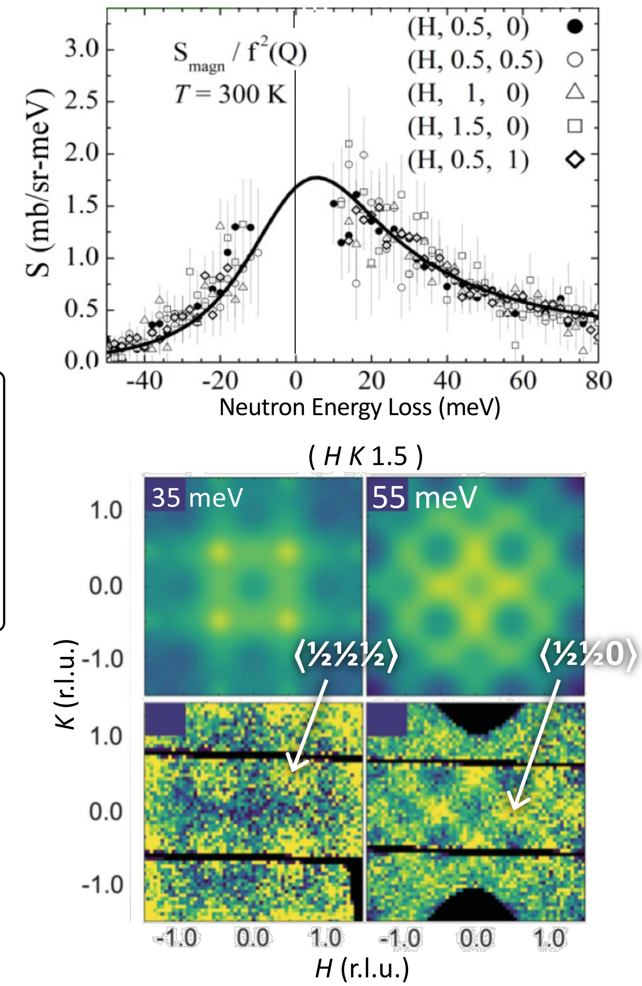


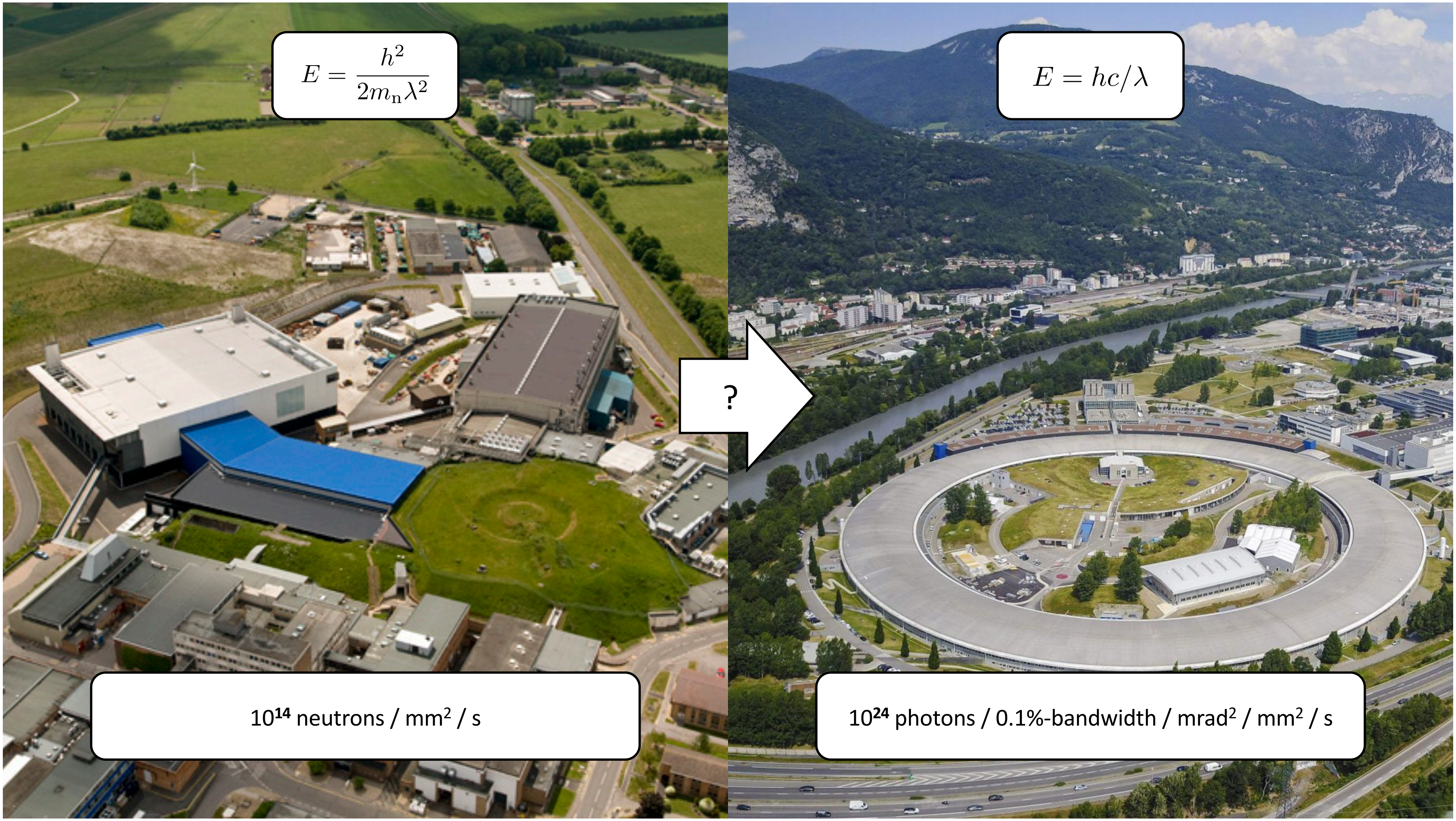
Goremychkin, Science 359, 186 (2018)

Photoemission



Neutron scattering




$E = \frac{h^2}{2m_\mathrm{n}\lambda^2}$
$E = hc/\lambda$
?
10^{14} neutrons / mm^2 / s
10^{24} photons / 0.1%-bandwidth / mrad^2 / mm^2 / s

Resonant X-ray Scattering (RIXS/REXS)

interaction Hamiltonian: *movement of an electron in an electromagnetic field*

$$\mathcal{H}_{\text{int}} = \sum_{i=1}^{N} \underbrace{\frac{e^2}{2mc^2}\boldsymbol{A}^2}_{\mathcal{H}_1} \underbrace{-\frac{e}{mc}\boldsymbol{p}_i\cdot\boldsymbol{A}}_{\mathcal{H}_2} \underbrace{-\frac{e\hbar}{mc}\boldsymbol{s}_i\cdot(\nabla\times\boldsymbol{A})}_{\mathcal{H}_3} + \underbrace{\frac{e\hbar}{2m^2c^3}\boldsymbol{s}_i\cdot\left[\left(\frac{\partial}{\partial t}\boldsymbol{A}\right)\times\left(\boldsymbol{p}_i-\frac{e}{c}\boldsymbol{A}\right)\right]}_{\mathcal{H}_4}$$

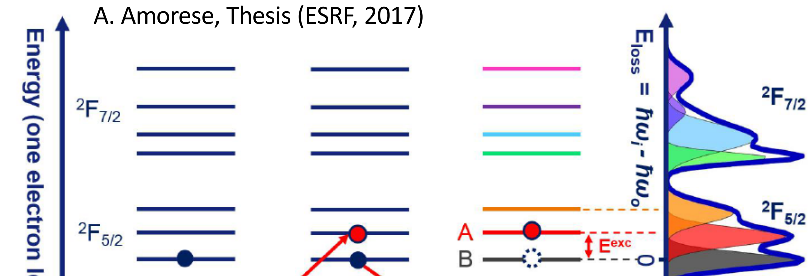
scattering amplitude in 2nd order perturbation theory:
Kramers-Heisenberg relation

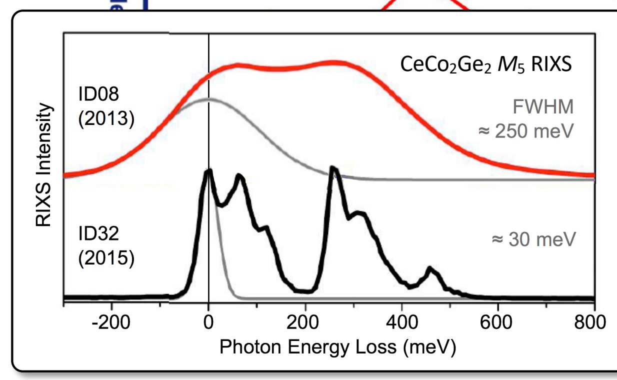
$$\mathcal{W}_{\text{res}} = \frac{2\pi}{\hbar}\sum_f \left|\sum_n \frac{\langle f|\mathcal{H}_2|n\rangle\langle n|\mathcal{H}_2|i\rangle}{E_i - E_n + \hbar\omega_{k_i} + i\Gamma_n/2}\right|^2 \delta(\hbar\omega_Q - (E_f - E_i))$$

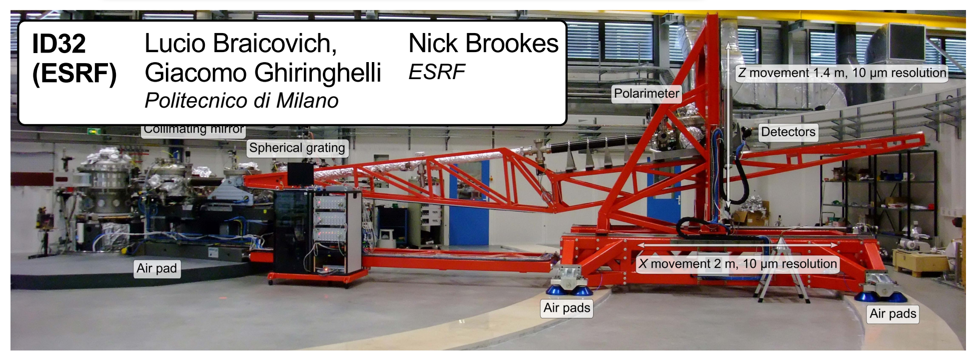
implies strong resonant enhancement at x-ray absorption edges

access to a wide range inelastic processes:

- magnons
- phonons
- charge-density wave excitations
- **inter-band transitions**
- ...







M-edge RIXS for localized Ce:

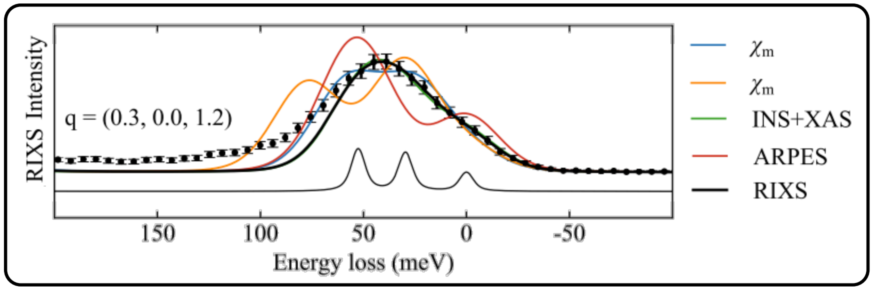
$CeRh_2Si_2$ $T_K = 2.6$ K $\ll E_{CEF} = 350\sim600$ K

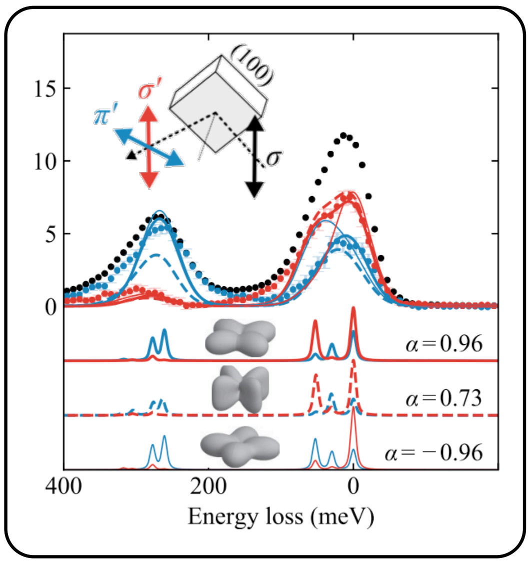
Amorese, PRB 97, 245130 (2018)

$$G^3(\omega_1,\omega_2) = \left\langle \psi_i \middle| T_1^\dagger \frac{1}{\omega_1 - H_1 - \imath\Gamma/2} T_2^\dagger \frac{1}{\omega_2 - H_2 + \imath\Gamma/2} T_2 \frac{1}{\omega_1 - H_1 + \imath\Gamma/2} T_1 \middle| \psi_i \right\rangle$$

multiplet (i.e. single ion) calculations using 3rd order Green's-function

(*Quanty* algorithm, M. Haverkort)

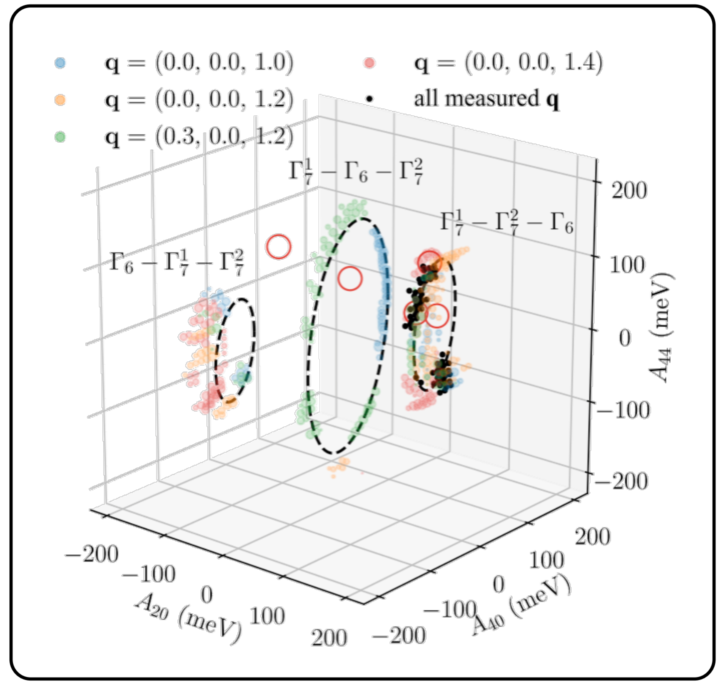




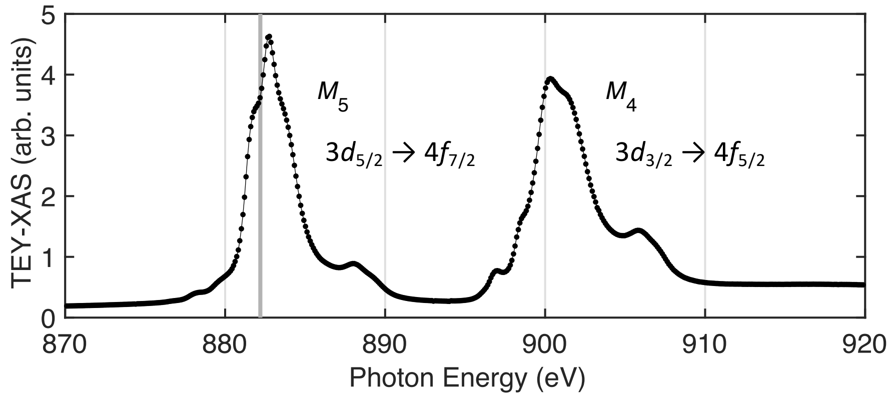
complete CEF determination, using

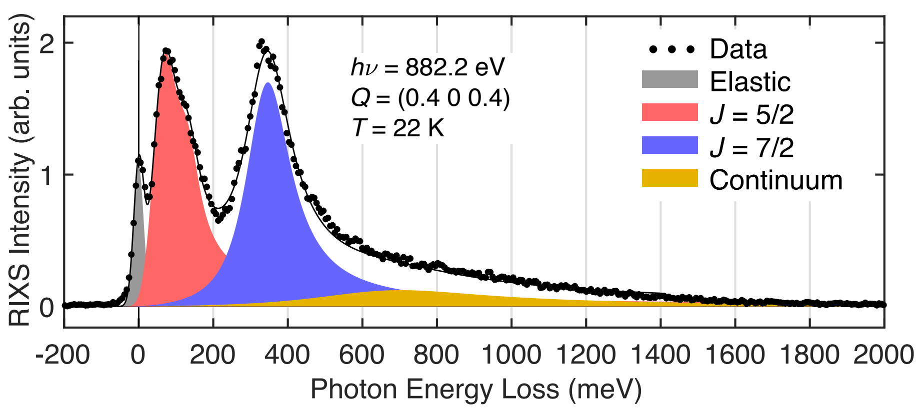
- 30 meV *energy resolution*
- *momentum resolution*
- *full polarization analysis*

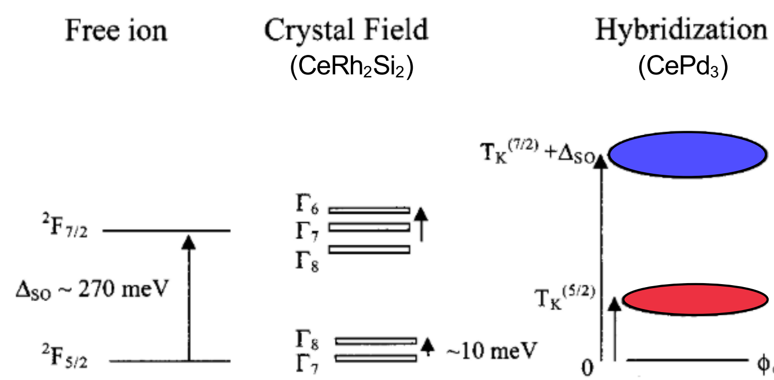
	A_{20}	A_{40}	A_{44}	Δ_1	Δ_2	α
RIXS	95	35	45	30	53	0.96
INS + XAS	64	101	88	30	52	±0.73
Magn. suscept.	106	65	33	27	59	±0.97
Magn. suscept.	48	−174	177	32	80	±0.90
ARPES	85	−83	120	48	62	±0.93



CePd$_3$ *M*-edge XAS & RIXS





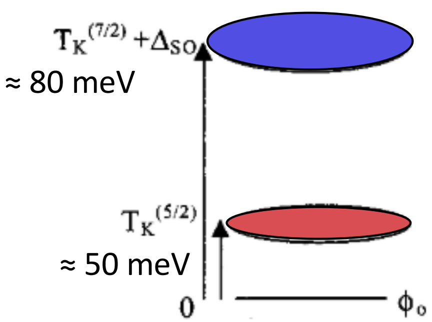


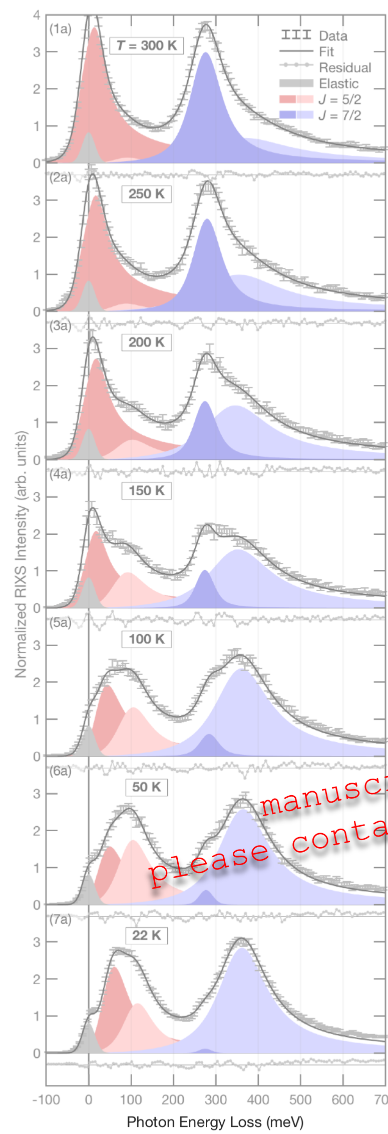
Murani *et al.*, PRB **65**, 095616 (2002)

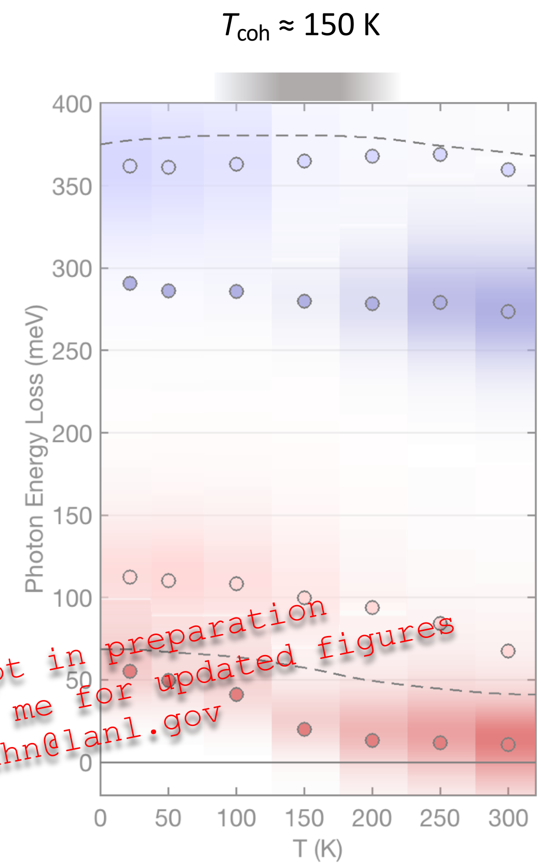
$CePd_3$ *M*-edge RIXS

thermal variation

strong renormalization of
quasiparticle interband excitations

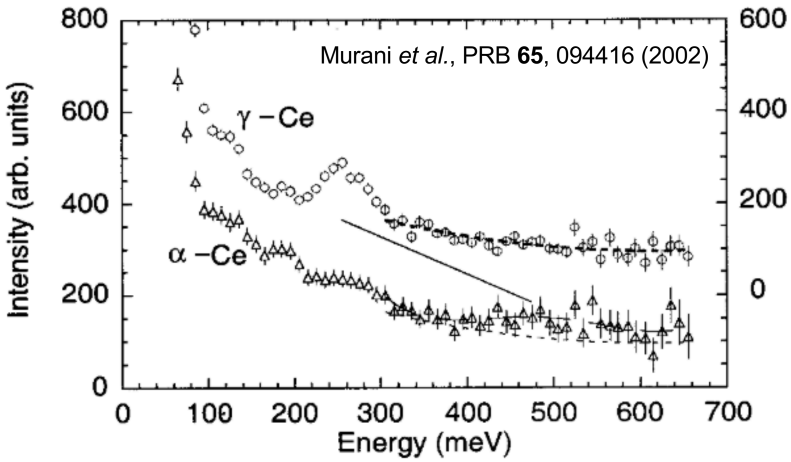




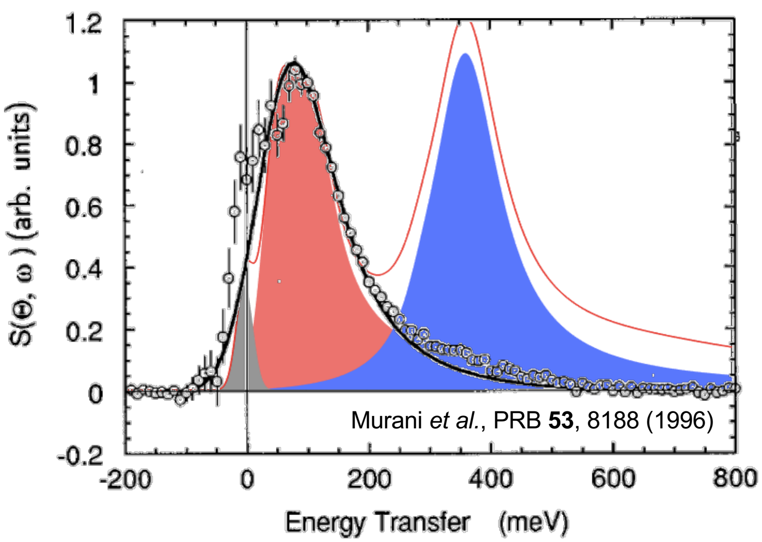


high-energy neutron scattering in Ce IV systems

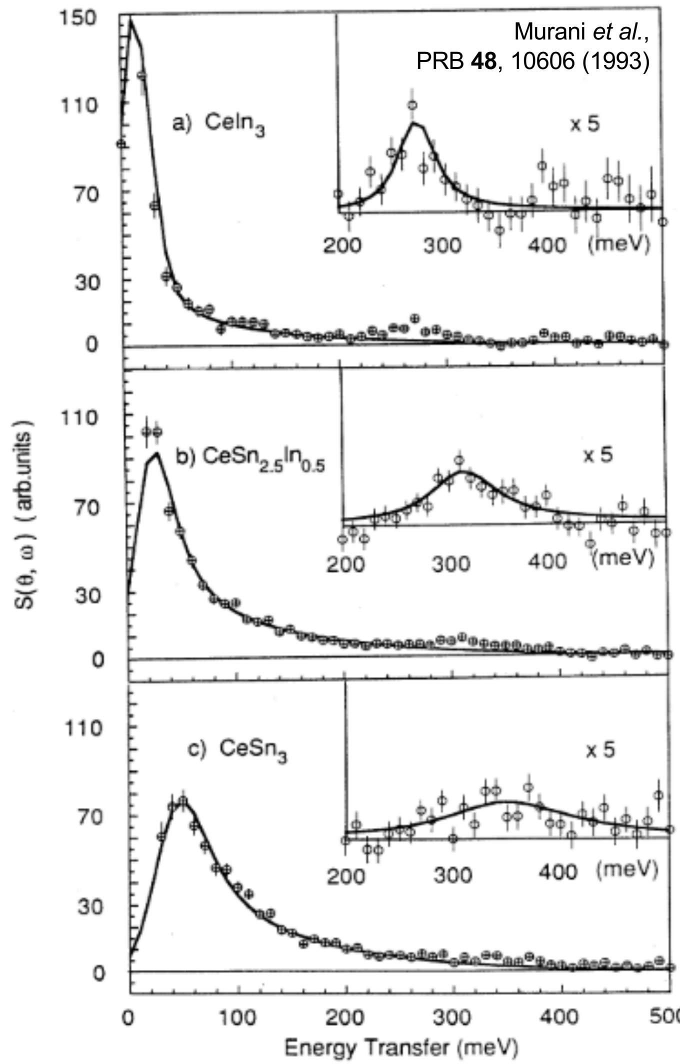
Ce metal
collapse at 9 kbar



$CePd_3$

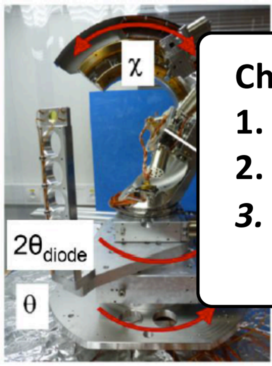


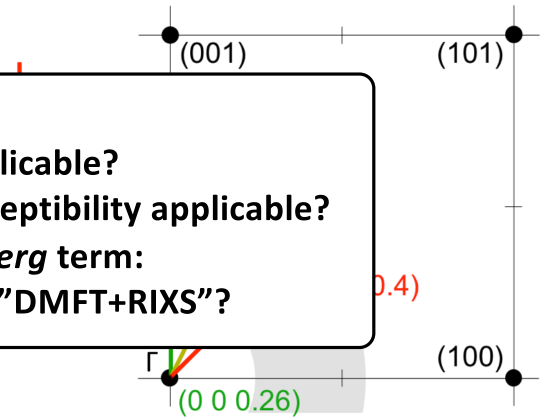
$CeIn_{3-x}Sn_x$



increasing Kondo energy

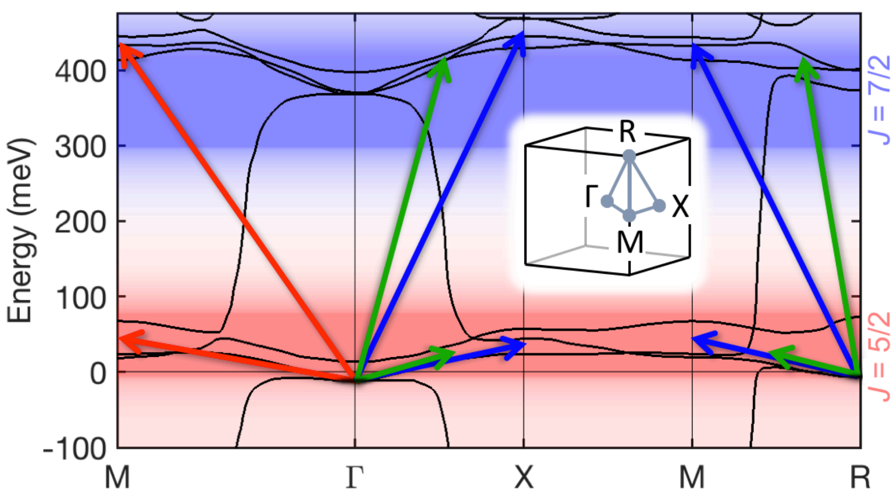
$CePd_3$ RIXS momentum dependence



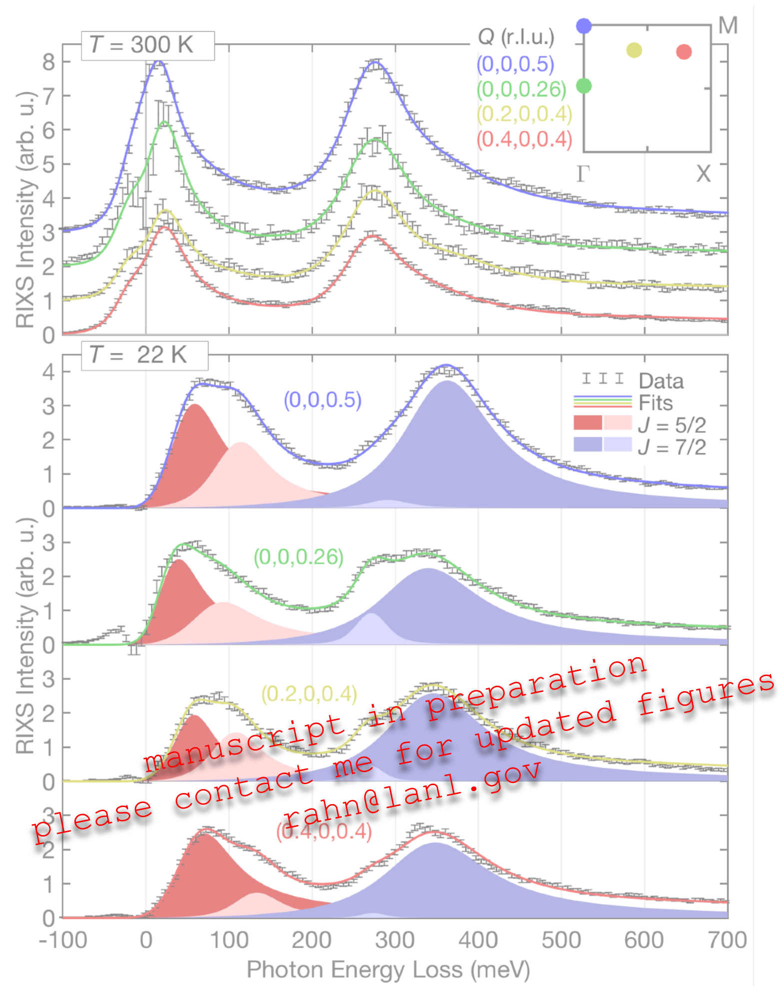


Challenges

1. **is DFT+DMFT applicable?**
2. **is a Lindhard susceptibility applicable?**
3. ***Kramers-Heisenberg* term:**
   How to calculate "DMFT+RIXS"?



Sakai, JPSJ 79 (11), 114701 (2010)



Thank You!
Marein Rahn, *rahn@lanl.gov*

Los Alamos NATIONAL LABORATORY EST. 1943

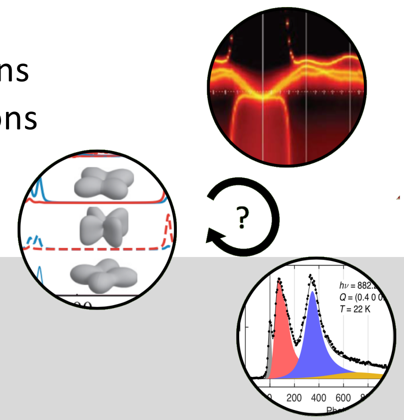
Alexander von Humboldt Stiftung / Foundation

- high-resolution soft-x-ray RIXS is an emerging technique which is well suited to probe excitations across hybridization gaps in valence fluctuating materials, as a function of energy- and momentum transfer as well as polarization

- RIXS is complementary to other spectroscopies (XAS, INS, ARPES) and offers a number of experimental advantages

  - tiny samples
  - no phonons
  - ionic specificity
  - fast
  - wide *E*-range

- the data provides a benchmark for computational methods – in terms of ab-initio calculations of correlated *f*-electronic bands, as well as calculations of the RIXS cross section for excitations between interacting itinerant states



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