Kurt Kummer, ESRF

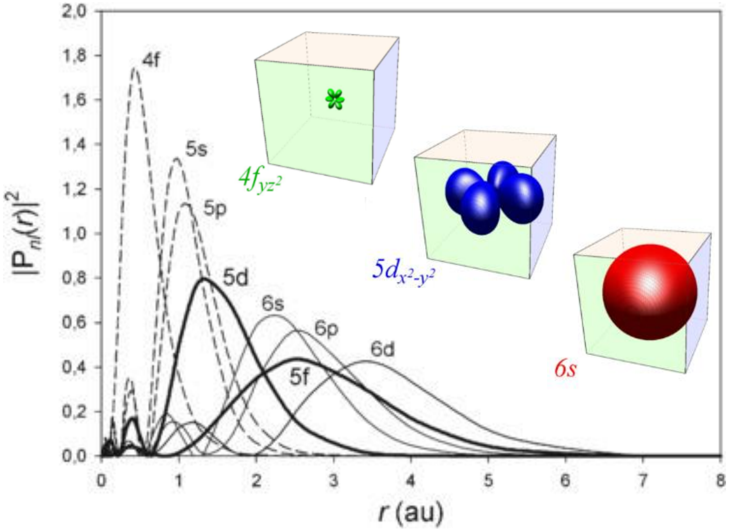
Low-energy excitations in the lanthanides studied with RIXS

Current status and opportunities

IXS 2019, BNL, 24 June 2019

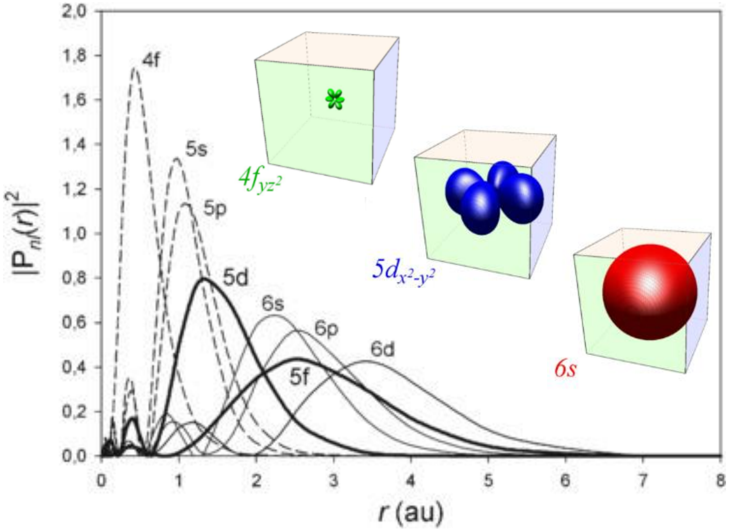
Dual nature of 4f electronic states

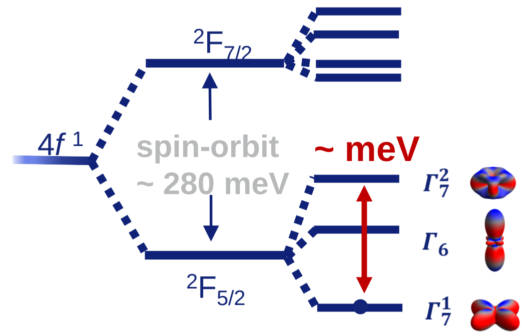
58	59	60	61	62	63	64	65	66	67	68	69	70	71
Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu

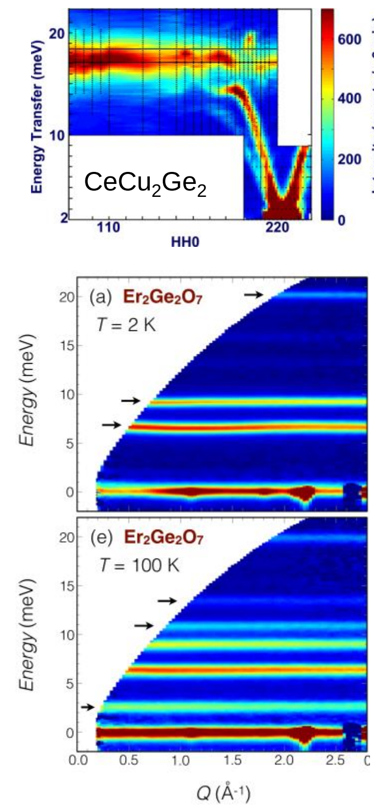


Dual nature of 4f electronic states

58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu



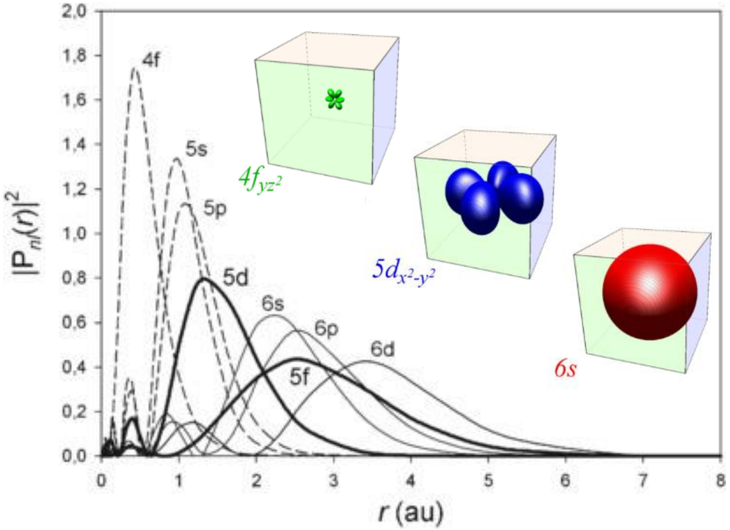


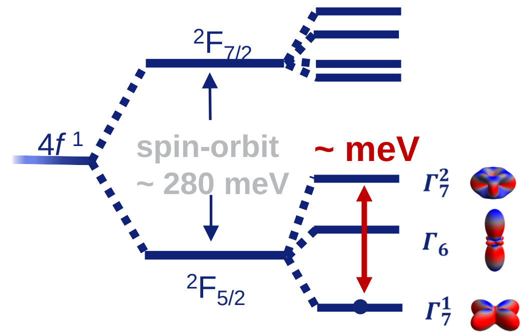


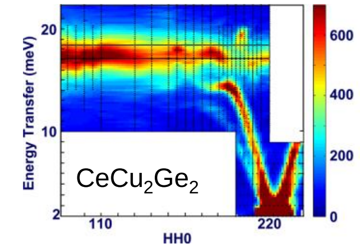
Loewenhaupt *et al.* J. Appl. Phys. **111**, 07E124 (2012)
Gaudet *et al.* PRB **97**, 024415 (2018)

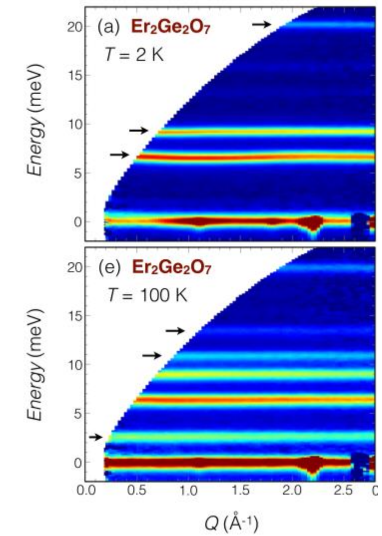
Dual nature of 4f electronic states

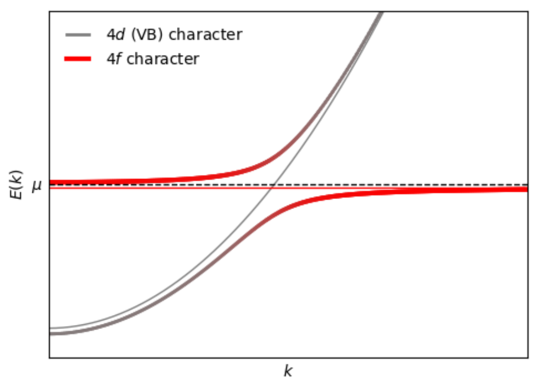
58	59	60	61	62	63	64	65	66	67	68	69	70	71
Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu

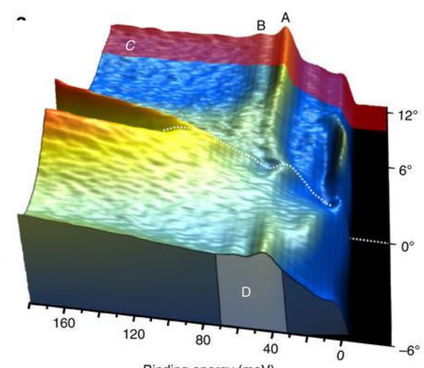










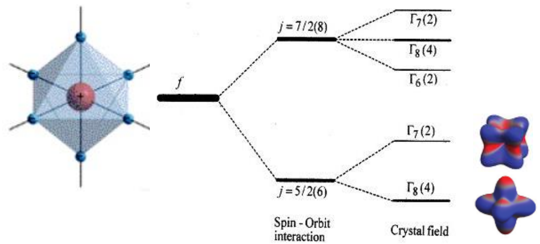


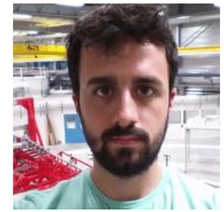
Patil *et al.* Nature Comm. **7**, 11029 (2016)

Loewenhaupt *et al.* J. Appl. Phys. **111**, 07E124 (2012)
Gaudet *et al.* PRB **97**, 024415 (2018)

In this talk

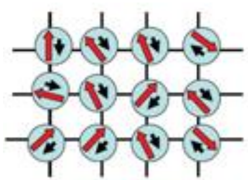
1. Local excitations

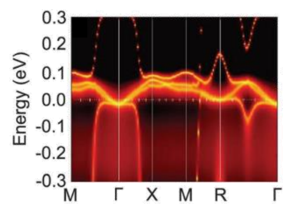




Andrea Amorese

2. Lattice excitations



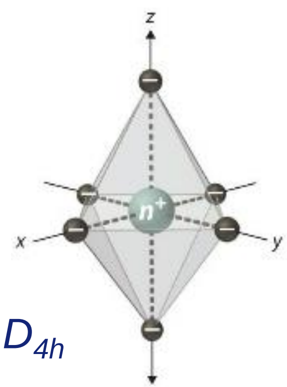


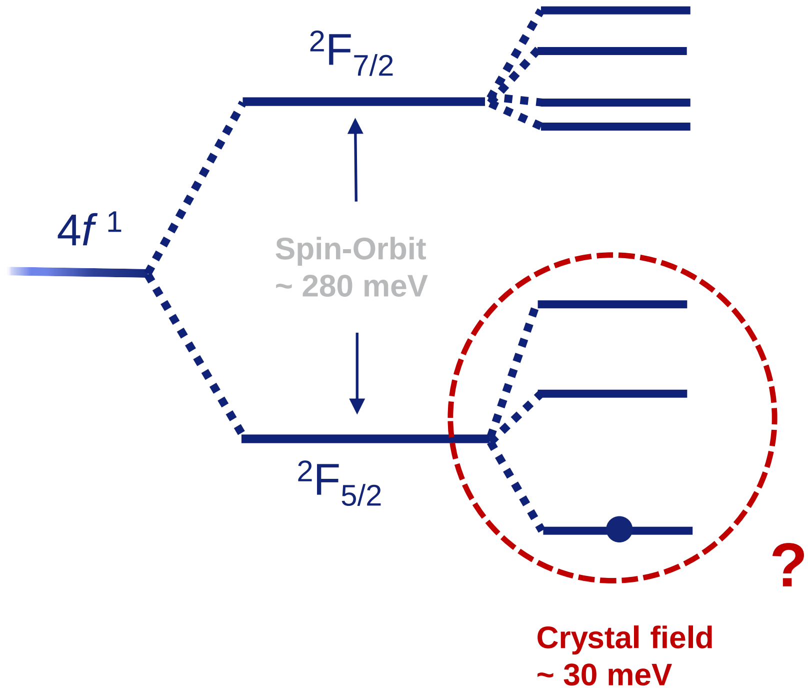
Marein Rahn

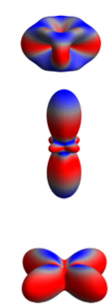
3. Perspectives

Crystal electric field in Ce^{3+}

Influence of crystal lattice reduced to an effective electrostatic field at the 4*f* site.







Γ_7^2 $\sqrt{(1-\alpha^2)}|\pm 5/2\rangle \pm \alpha|\mp 3/2\rangle$

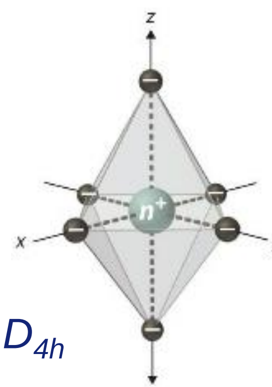
Γ_6 $|\pm 1/2\rangle$

Γ_7^1 $\alpha|\pm 5/2\rangle \mp \sqrt{(1-\alpha^2)}\,|\mp 3/2\rangle$

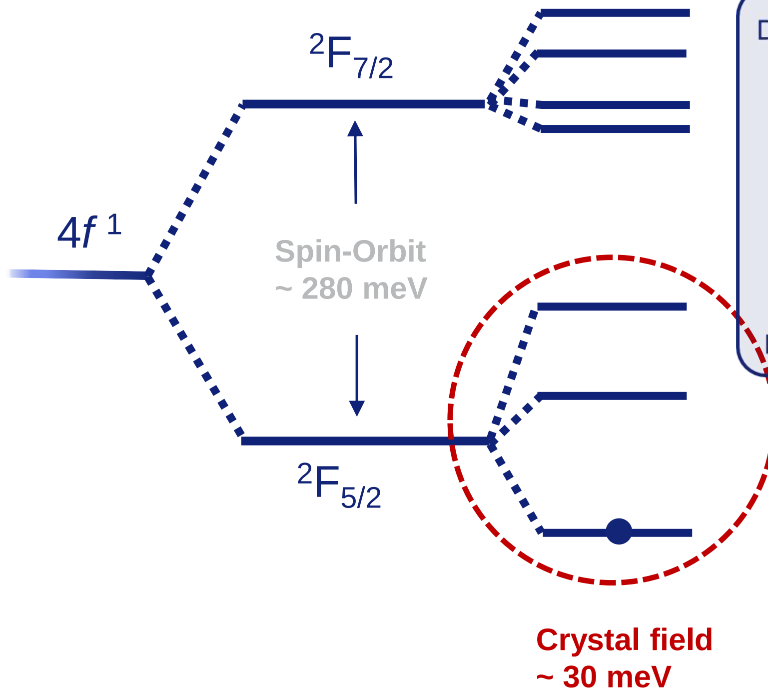
$$H = H_0 + H_{SO} + H_{CEF}$$

Crystal electric field in Ce^{3+}

Influence of crystal lattice reduced to an effective electrostatic field at the 4*f* site.



D_{4h}



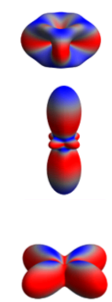
Description with local (single site) model Hamiltonian

$V^{CEF} = \sum_{k,q} A_k^q r^k C_k^q$ acting on $\varphi_i = R_{n_i}^{l_i} Y_{l_i}^{m_i}$

$$H_{i,j}^{CEF} = e \sum_{k,q} \underbrace{A_k^q \left\langle R_{n_i}^{l_i} \middle| r^k \middle| R_{n_j}^{l_j} \right\rangle}_{\text{crystal field parameters } \tilde{\mathbf{A}}_{\mathbf{k}}^{\mathbf{q}}} \left\langle Y_{l_i}^{m_i} \middle| C_k^q \middle| Y_{l_j}^{m_j} \right\rangle$$

D_{4h} symmetry, J = 5/2 → fully described by $\tilde{\mathbf{A}}_{\mathbf{2}}^{\mathbf{0}}$, $\tilde{\mathbf{A}}_{\mathbf{4}}^{\mathbf{0}}$ and $\tilde{\mathbf{A}}_{\mathbf{4}}^{\mathbf{4}}$.

?



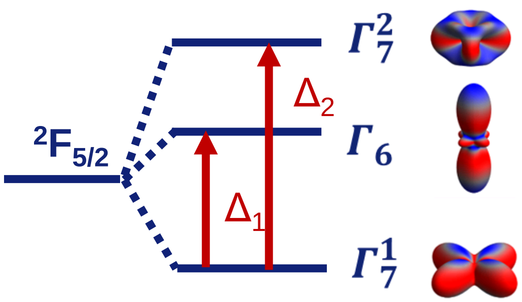
$\boldsymbol{\Gamma_7^2}$ $\sqrt{(1-\boldsymbol{\alpha}^2)}|\pm 5/2\rangle \pm \boldsymbol{\alpha}|\mp 3/2\rangle$

$\boldsymbol{\Gamma_6}$ $|\pm 1/2\rangle$

$\boldsymbol{\Gamma_7^1}$ $\boldsymbol{\alpha}|\pm 5/2\rangle \mp \sqrt{(1-\boldsymbol{\alpha}^2)}\,|\mp 3/2\rangle$

$$H = H_0 + H_{SO} + H_{CEF}$$

Crystal electric field in Ce^{3+}

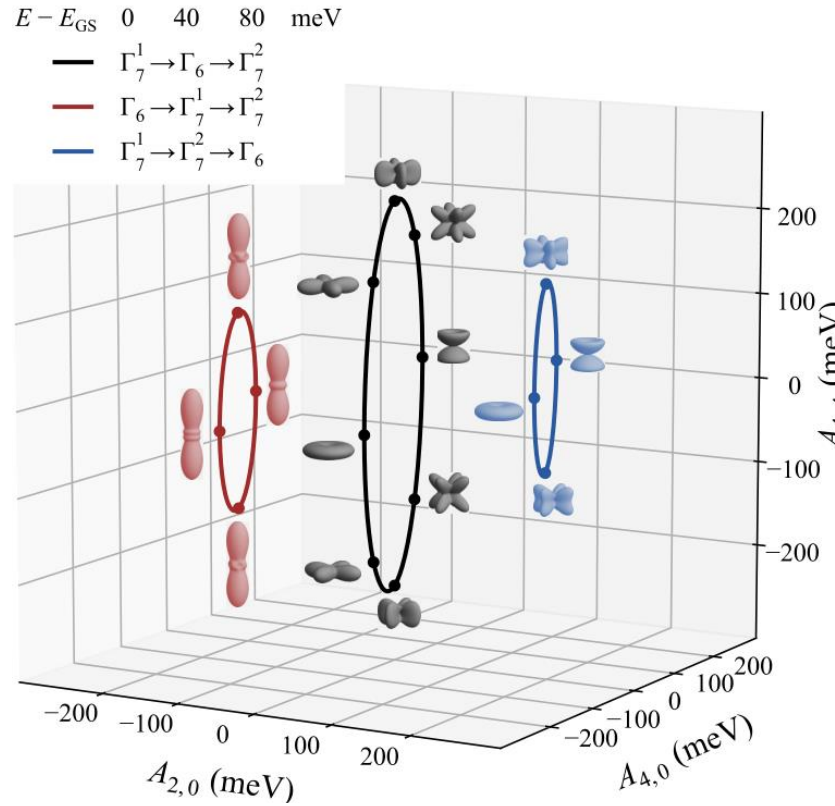


$\sqrt{(1-\alpha^2)}|\pm 5/2\rangle \pm \alpha|\mp 3/2\rangle$

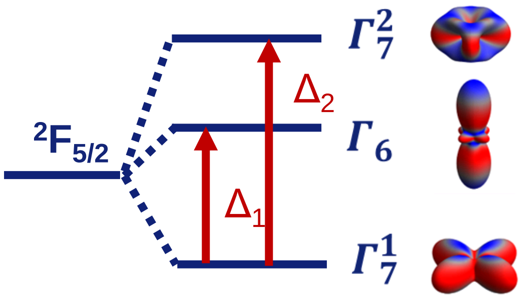
$|\pm 1/2\rangle$

$\alpha|\pm 5/2\rangle \mp \sqrt{(1-\alpha^2)}\,|\mp 3/2\rangle$

$$H_{i,j}^{CEF} = e\sum_{k,q} \underbrace{A_k^q \left\langle R_{n_i}^{l_i}\middle|r^k\middle|R_{n_j}^{l_j}\right\rangle}_{\text{crystal field parameters } \breve{\mathbf{A}}_{\mathbf{k}}^{\mathbf{q}}} \left\langle Y_{l_i}^{m_i}\middle|C_k^q\middle|Y_{l_j}^{m_j}\right\rangle$$



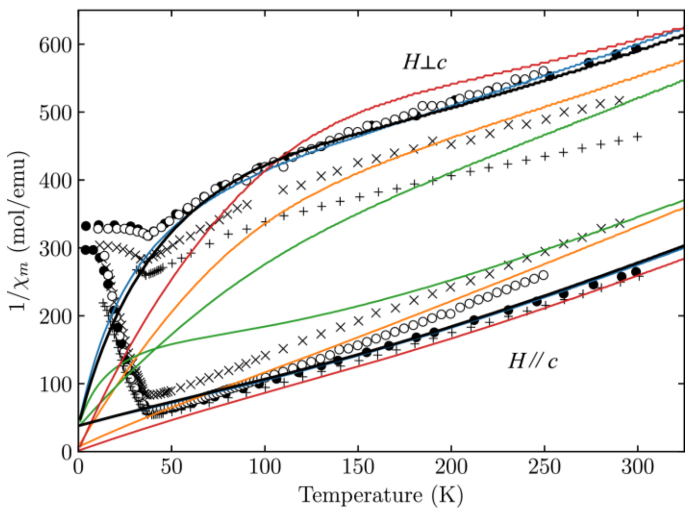
Crystal electric field in Ce^{3+}



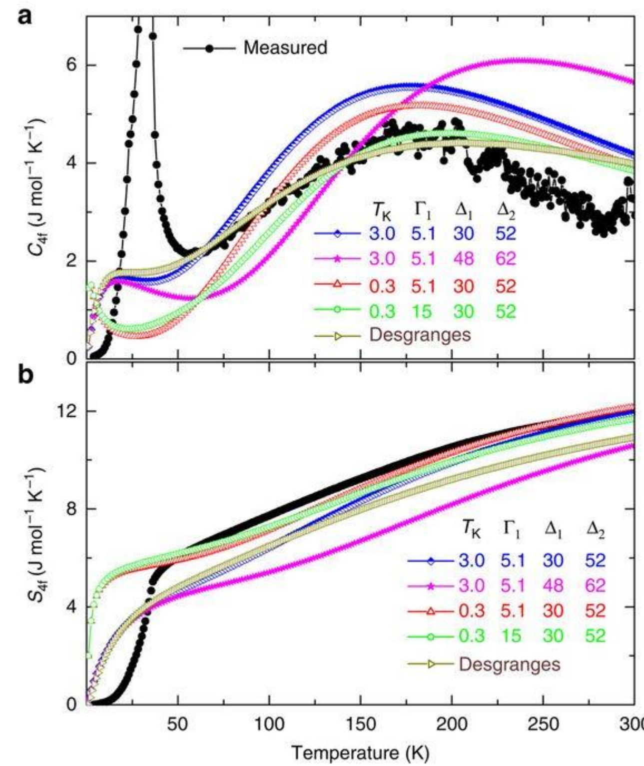
$$\Gamma_7^2: \sqrt{(1-\alpha^2)}|\pm 5/2\rangle \pm \alpha|\mp 3/2\rangle$$

$$\Gamma_6: |\pm 1/2\rangle$$

$$\Gamma_7^1: \alpha|\pm 5/2\rangle \mp \sqrt{(1-\alpha^2)}\,|\mp 3/2\rangle$$

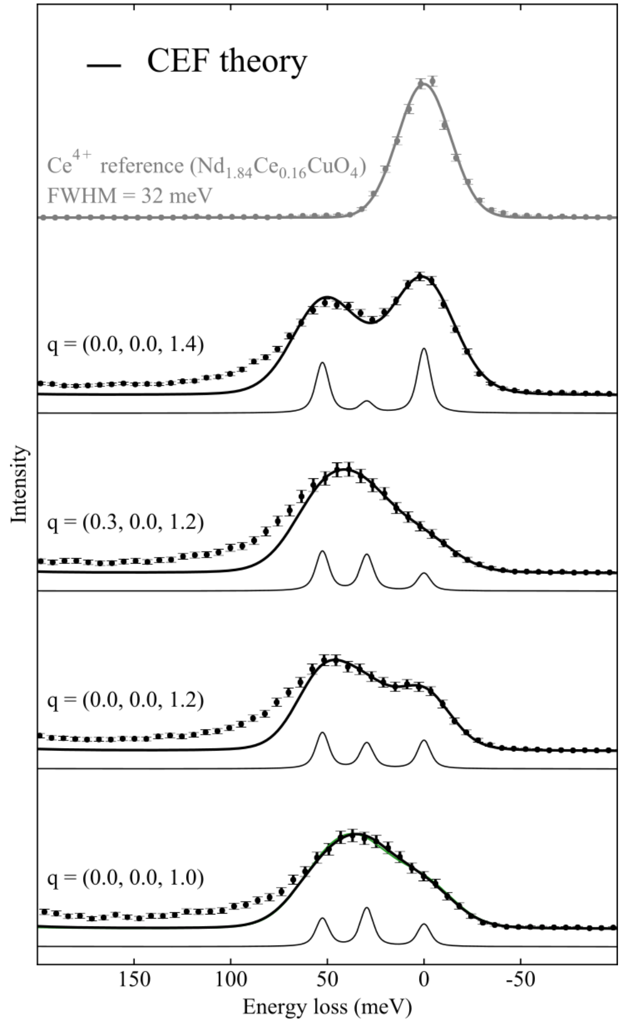


Amorese *et al.* PRB **97**, 24513 (2018)



S. Patil *et al.* Nature Comm. **7**, 11029 (2016)

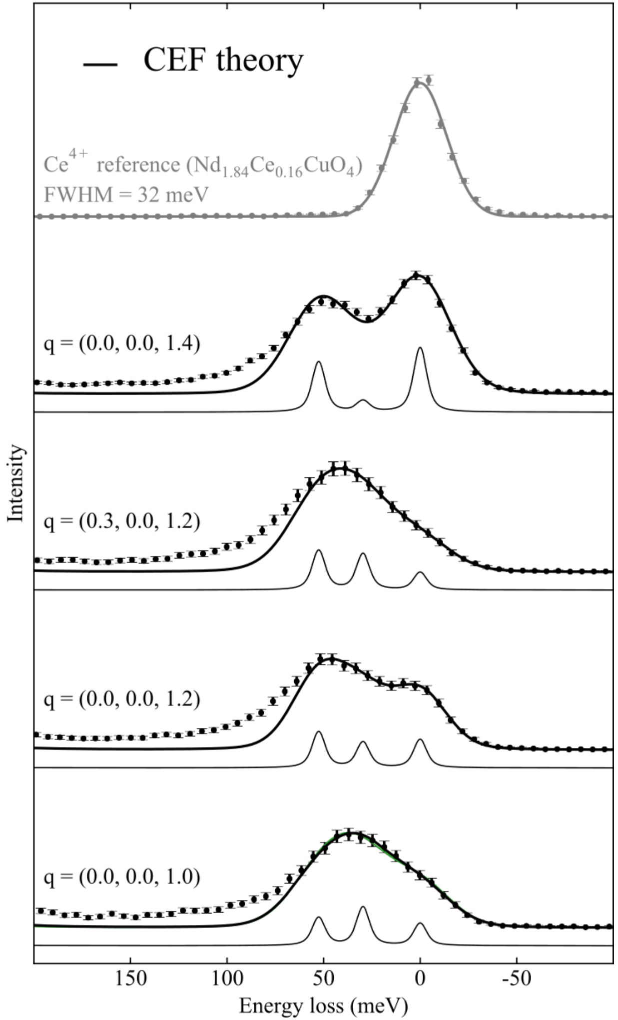
Example: $CeRh_2Si_2$



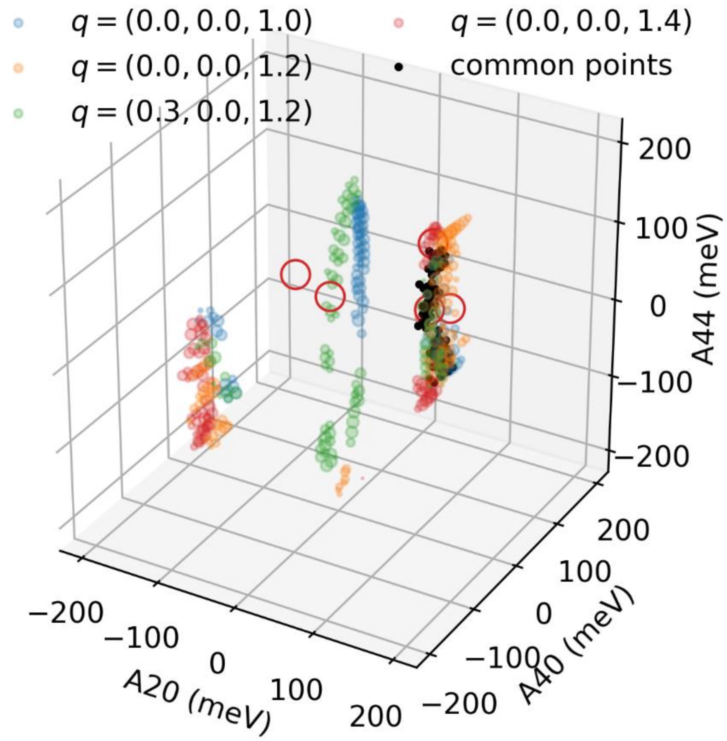
$$\frac{d^2\sigma}{d\Omega\ d(\hbar\omega_k')} \propto \sum_{|f\rangle} \left| \sum_{|n\rangle} \frac{\langle f|T^\dagger|n\rangle\langle n|T|i\rangle}{E_i - E_n + \hbar\omega_k + i\frac{\Gamma_n}{2}} \right|^2 \times \delta\left(E_i - E_f + \hbar\omega_k - \hbar\omega_k'\right)$$

Amorese *et al.* PRB **97**, 24513 (2018)

Example: $CeRh_2Si_2$



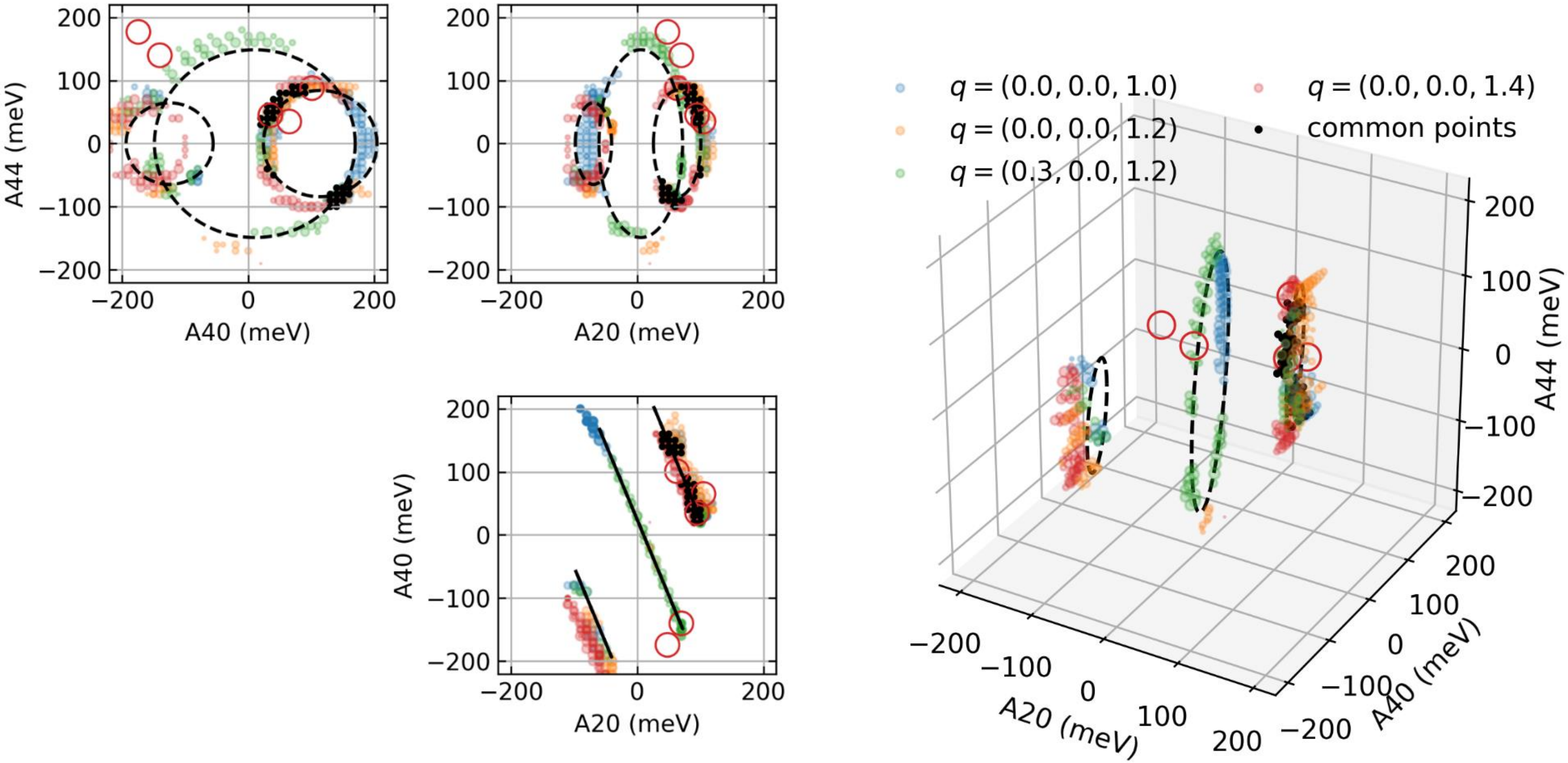
$$\frac{d^2\sigma}{d\Omega\, d(\hbar\omega_k')} \propto \sum_{|f\rangle} \left| \sum_{|n\rangle} \frac{\langle f|T^\dagger|n\rangle\langle n|T|i\rangle}{E_i - E_n + \hbar\omega_k + i\frac{\Gamma_n}{2}} \right|^2 \times \delta\left(E_i - E_f + \hbar\omega_k - \hbar\omega_k'\right)$$



Amorese *et al.* PRB **97**, 24513 (2018)

Example: $CeRh_2Si_2$

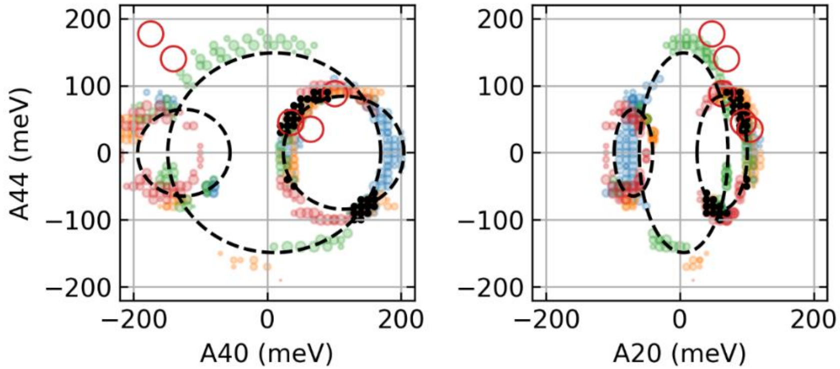
$$\frac{d^2\sigma}{d\Omega\ d(\hbar\omega_k')} \propto \sum_{|f\rangle} \left| \sum_{|n\rangle} \frac{\langle f|T^\dagger|n\rangle\langle n|T|i\rangle}{E_i - E_n + \hbar\omega_k + i\frac{\Gamma_n}{2}} \right|^2 \times \delta\left(E_i - E_f + \hbar\omega_k - \hbar\omega_k'\right)$$

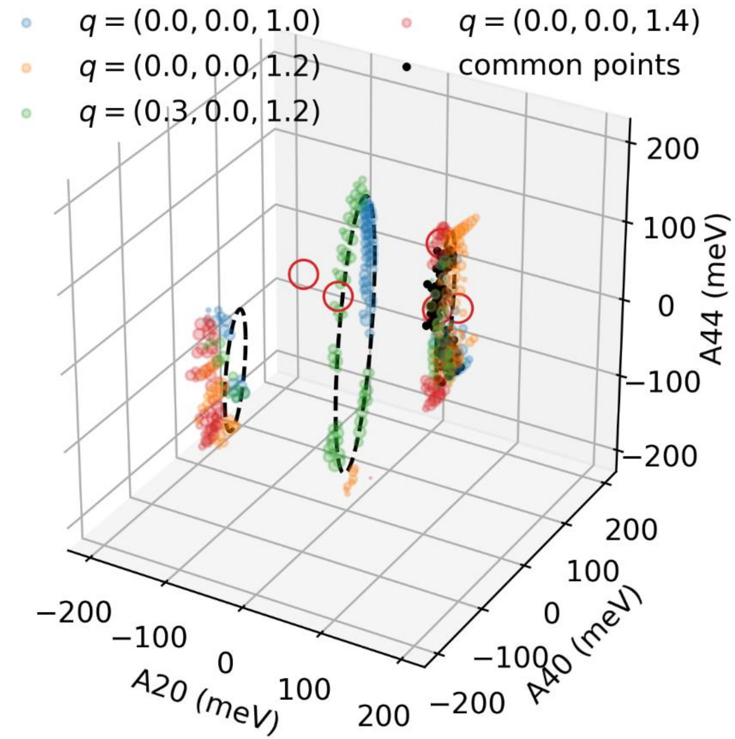


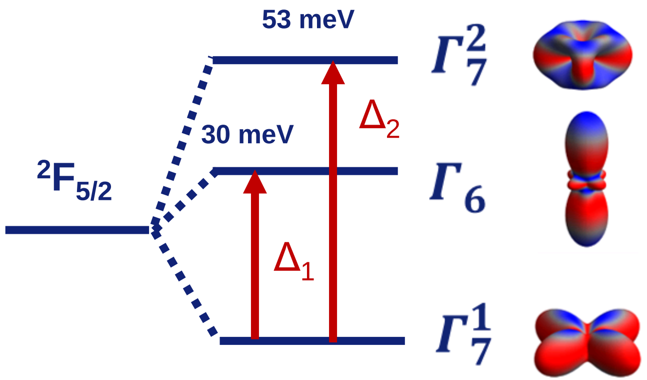
Amorese *et al.* PRB **97**, 24513 (2018)

Example: $CeRh_2Si_2$

$$\frac{d^2\sigma}{d\Omega\, d(\hbar\omega_k')} \propto \sum_{|f\rangle} \left| \sum_{|n\rangle} \frac{\langle f|T^\dagger|n\rangle\langle n|T|i\rangle}{E_i - E_n + \hbar\omega_k + i\frac{\Gamma_n}{2}} \right|^2 \times \delta\left(E_i - E_f + \hbar\omega_k - \hbar\omega_k'\right)$$



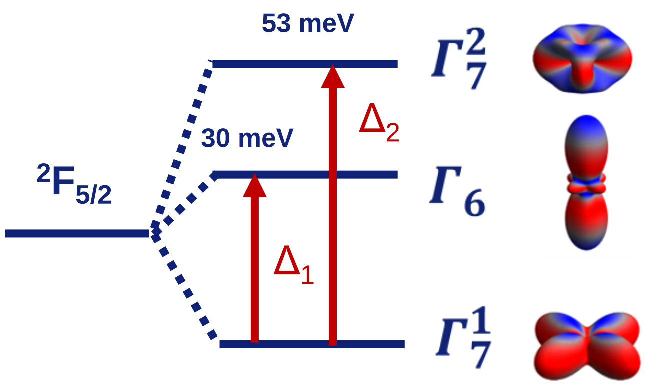


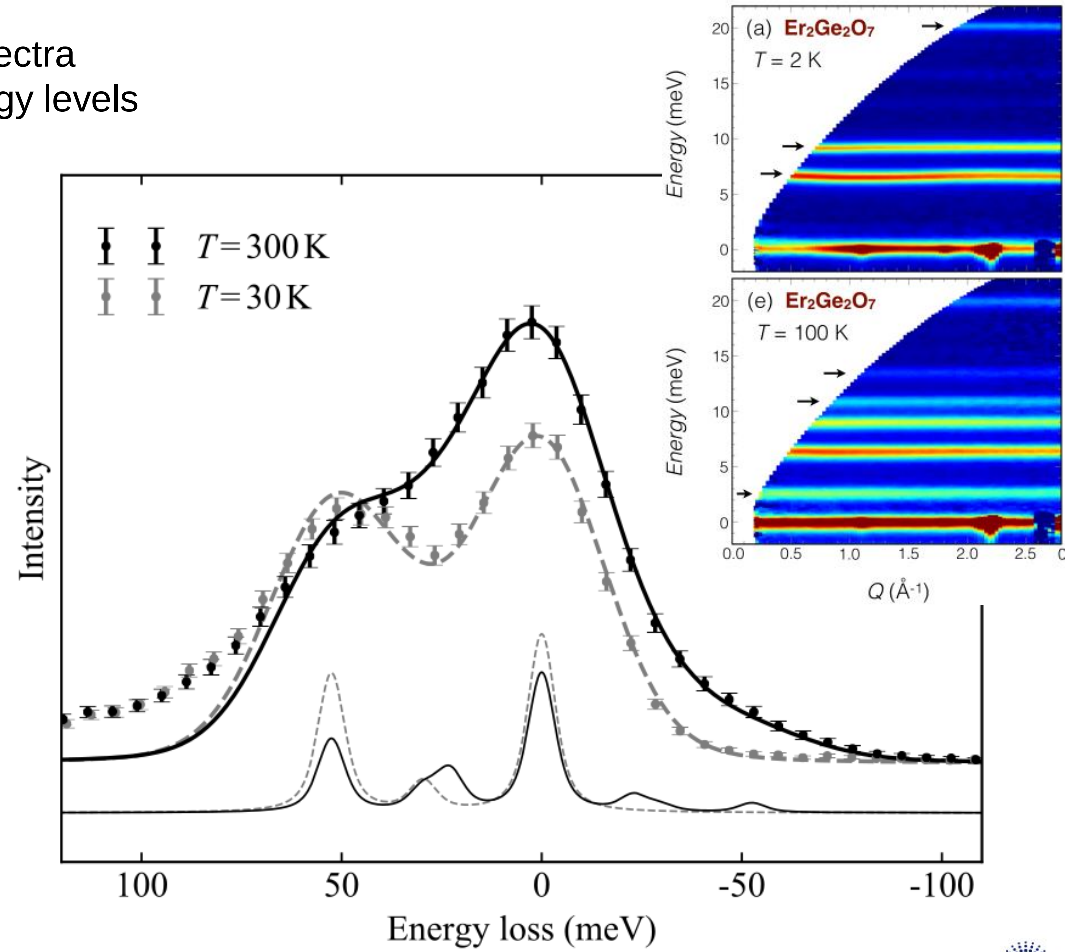


Amorese *et al.* PRB **97**, 24513 (2018)

Example: $CeRh_2Si_2$

Temperature dependence of excitation spectra
→ Boltzmann statistics over discrete energy levels

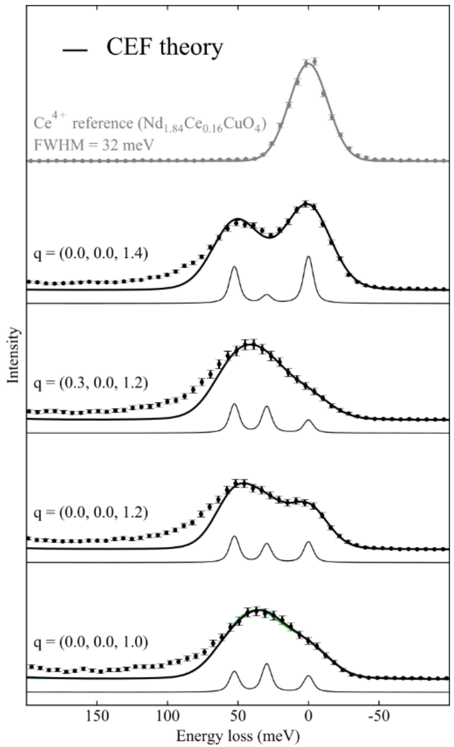


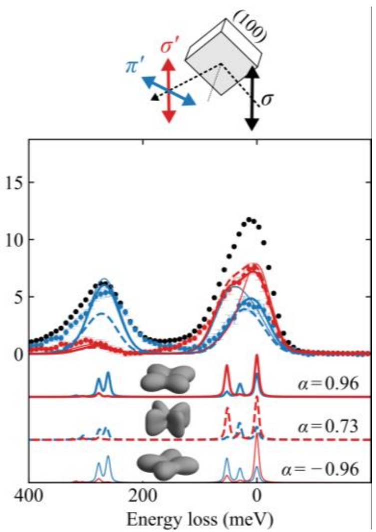


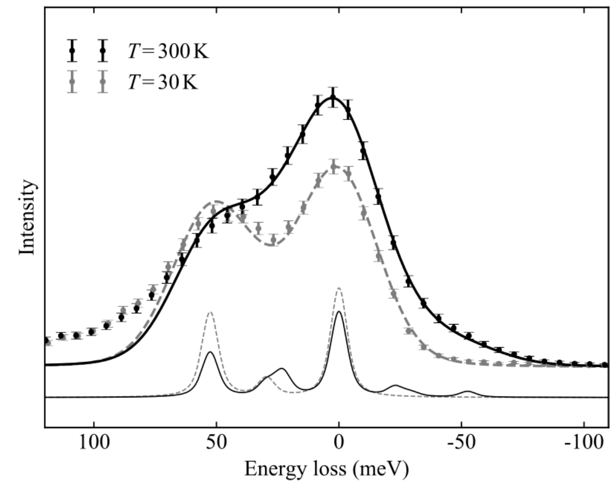
Amorese *et al.* PRB **97**, 24513 (2018)

Local excitations: summary

- Geometry dependent modulations of cross section
- Temperature dependence = Boltzmann statistics over discrete levels
- CEF theory describes local excitation spectra well

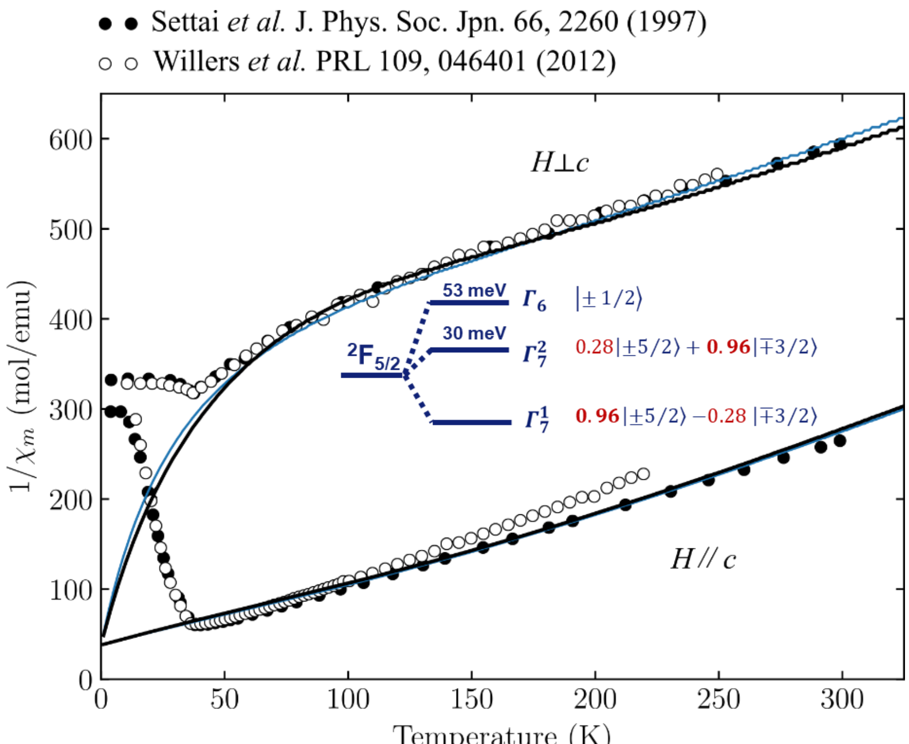


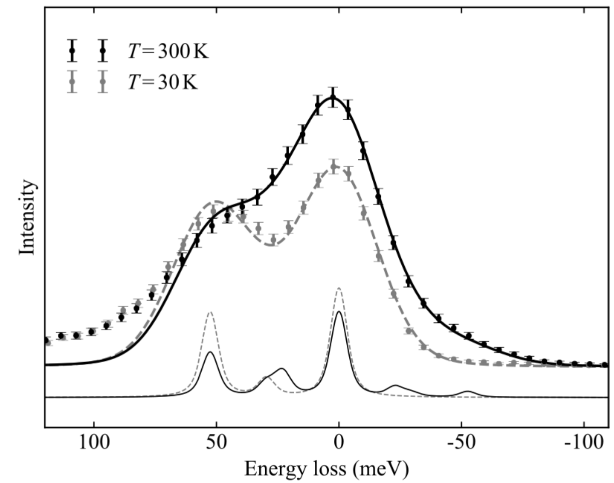




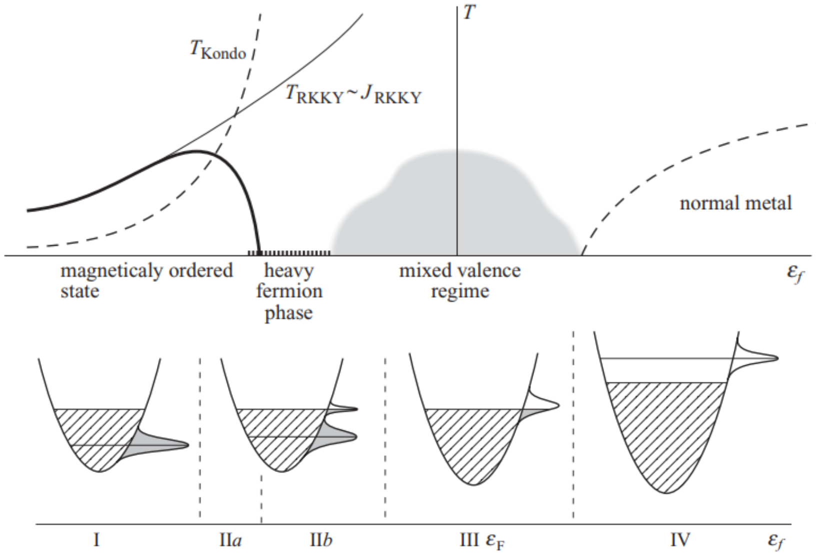
Local excitations: summary

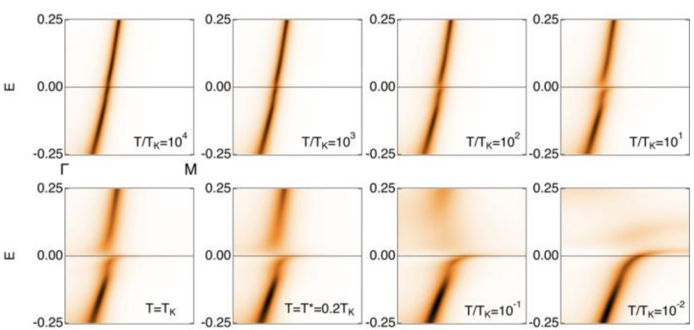
- Geometry dependent modulations of cross section
- Temperature dependence = Boltzmann statistics over discrete levels
- CEF theory describes local excitation spectra well





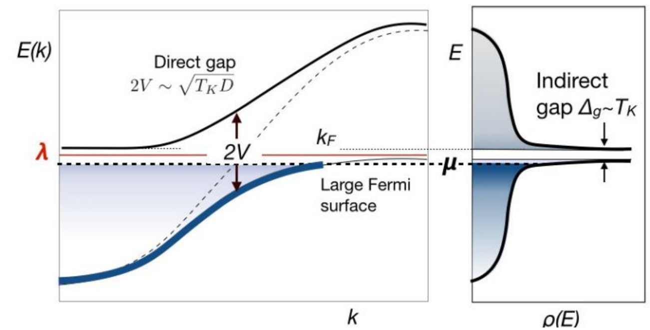
Break down of the local model

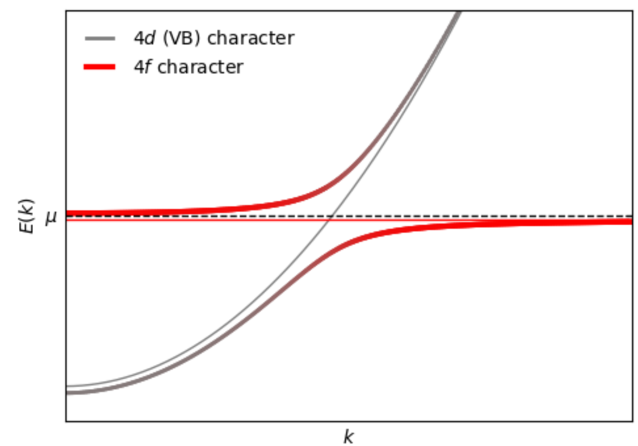




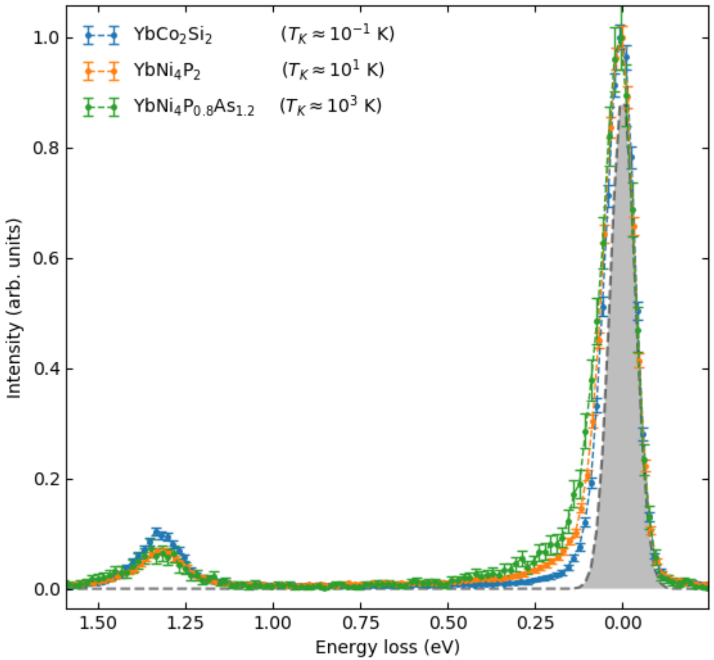
Kang *et al.* PRB **99**, 165115 (2019)

T dependence and **q** dependence of low energy excitations in Anderson lattice?

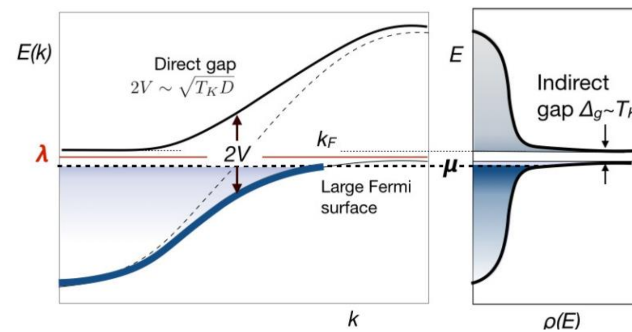


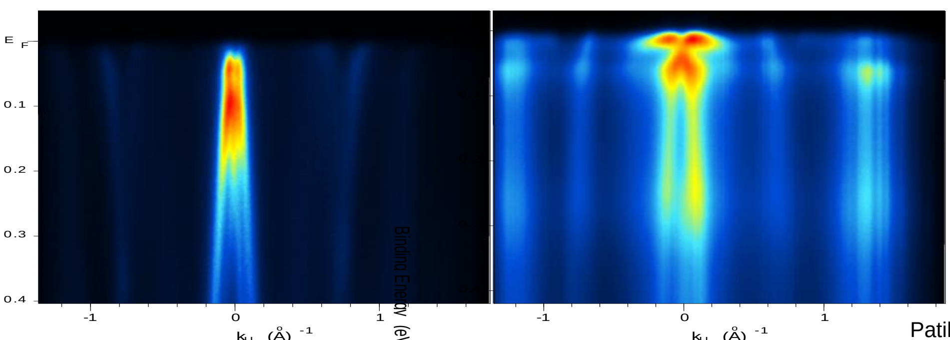


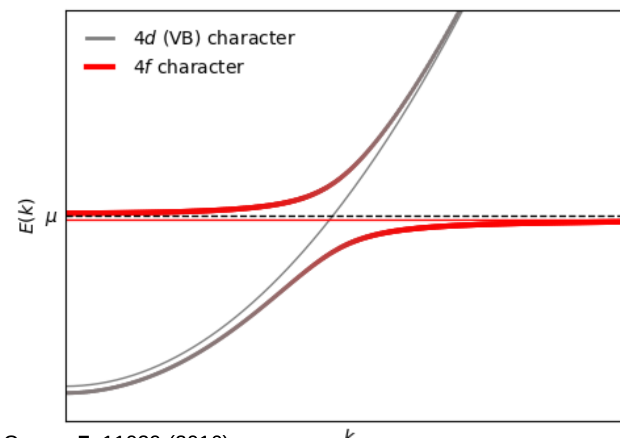
Break down of the local model



T dependence and **q** dependence of low energy excitations in Anderson lattice?



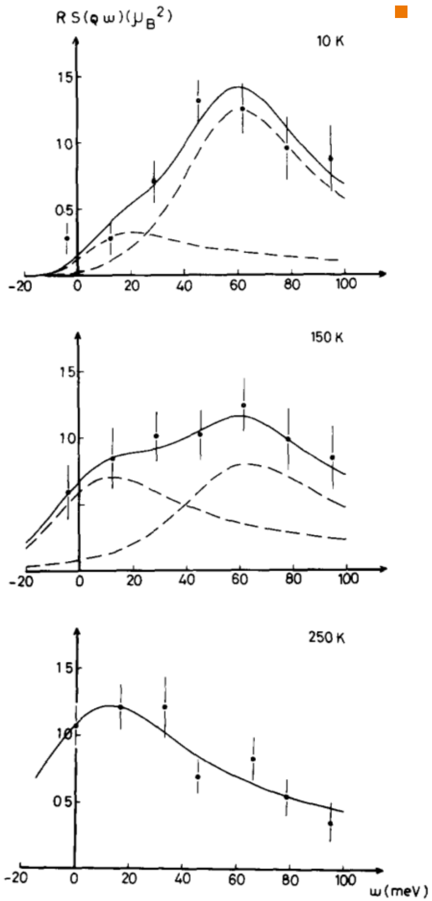




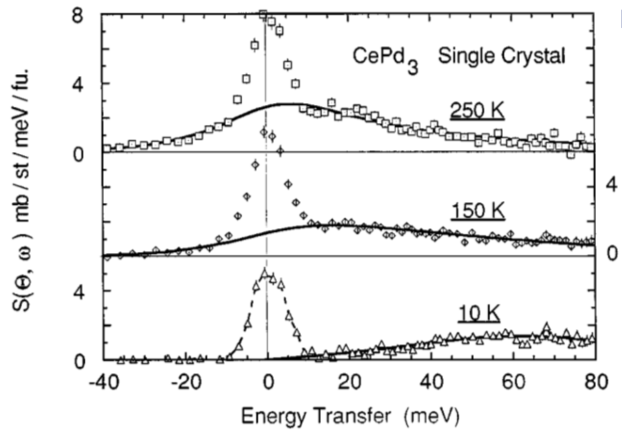
Patil *et al.* Nature Comm. **7**, 11029 (2016)

Characteristics of the excitation spectra in $CePd_3$

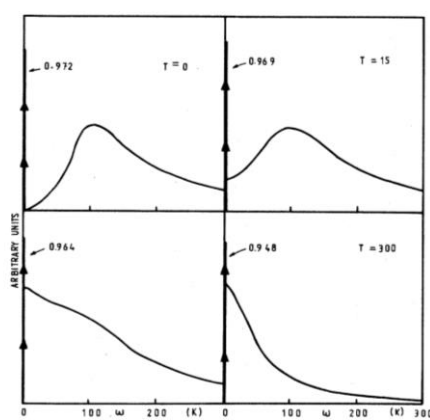
- Strong *T* dependence with broad inelastic response at low *T* and quasi-elastic excitations at room temperature



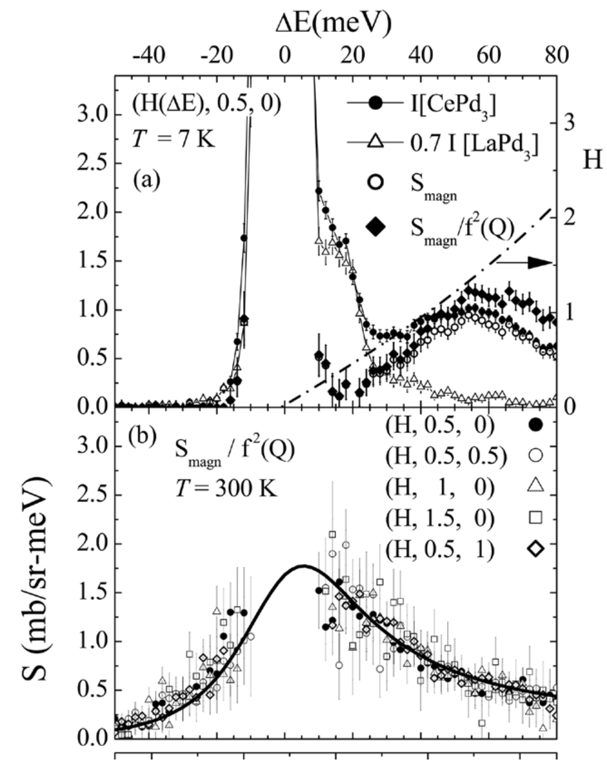
Galera *et al.* JMMM **63-64**, 594 (1987)



Murani *et al.* PRB **53**, 2641 (1996)



Balseiro *et al.* PRB **26**, 2615 (1982)

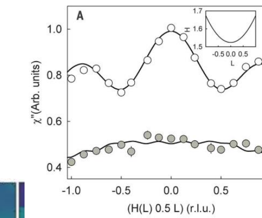


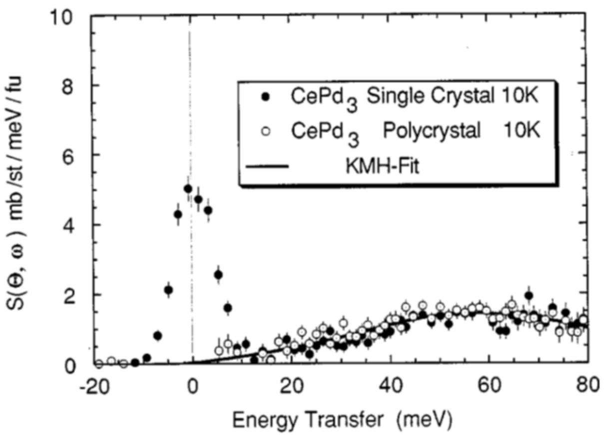
Fanelli *et al.* J. Phys. Cond. Mat. **26**, 25602 (2014)

Characteristics of the excitation spectra in $CePd_3$

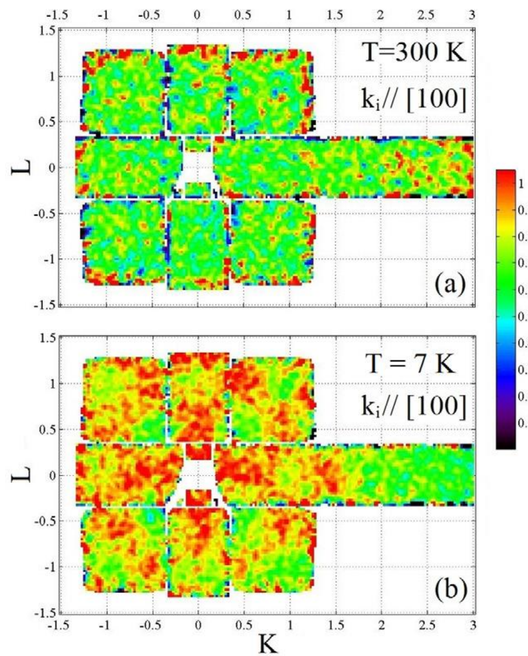
- Strong T dependence with broad inelastic response at low T and quasi-elastic excitations at room temperature

- **q** dependence at low T, **q** independent response at high T

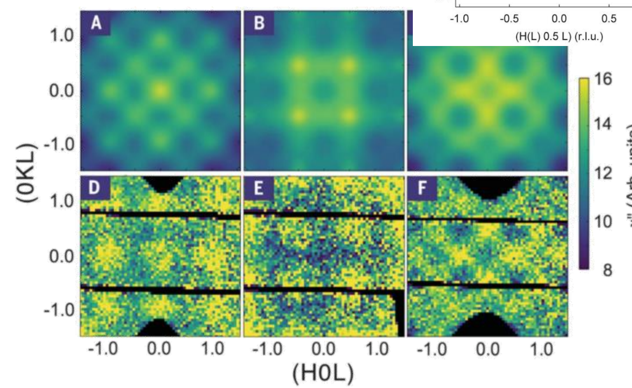




Murani *et al.* PRB **53**, 2641 (1996)



Fanelli *et al.* J. Phys. Cond. Mat. **26**, 25602 (2014)



Coherent band excitations in $CePd_3$: A comparison of neutron scattering and ab initio theory

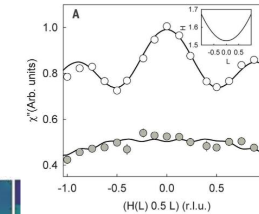
Eugene A. Goremychkin,[1] Hyowon Park,[2,3] Raymond Osborn,[2*] Stephan Rosenkranz,[2] John-Paul Castellan,[2,4] Victor R. Fanelli,[5] Andrew D. Christianson,[6] Matthew B. Stone,[6] Eric D. Bauer,[7] Kenneth J. McClellan,[7] Darrin D. Byler,[7] Jon M. Lawrence[7,8]

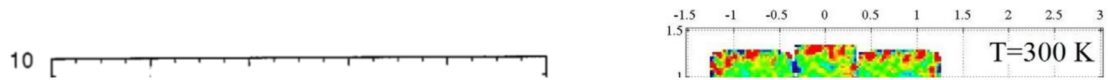
Goremychkin *et al.* Science **359**, 186 (2018)

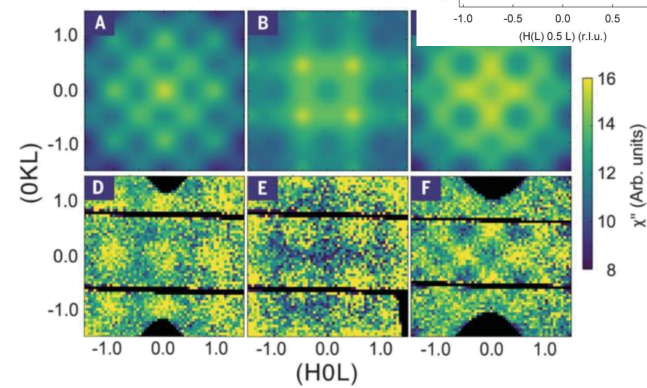
Characteristics of the excitation spectra in $CePd_3$

- Strong *T* dependence with broad inelastic response at low *T* and quasi-elastic excitations at room temperature

- **q** dependence at low *T*, **q** independent response at high *T*



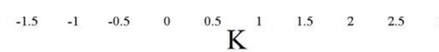




- ➢ *T* dependence already reproduced by Anderson impurity models
- ➢ **q** dependence requires a lattice model
- ➢ Measuring low energy excitations (*T*, **q)** dependently allows to test theories of Kondo/Anderson lattices
- ➢ Strong experimental input from RIXS

Coherent band excitations in $CePd_3$: A comparison of neutron scattering and ab initio theory

Eugene A. Goremychkin,[1] Hyowon Park,[2,3] Raymond Osborn,[2*] Stephan Rosenkranz,[2] John-Paul Castellan,[2,4] Victor R. Fanelli,[5] Andrew D. Christianson,[6] Matthew B. Stone,[6] Eric D. Bauer,[7] Kenneth J. McClellan,[7] Darrin D. Byler,[7] Jon M. Lawrence[7,8]

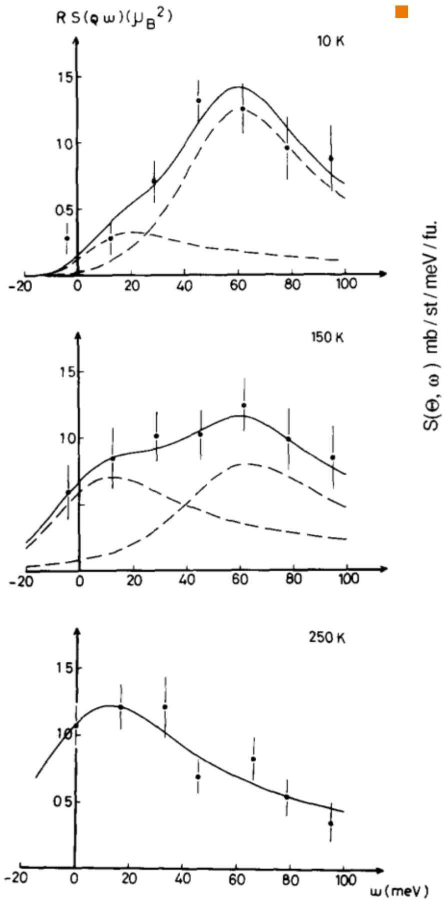


Fanelli *et al.* J. Phys. Cond. Mat. **26**, 25602 (2014)

Goremychkin *et al.* Science **359**, 186 (2018)

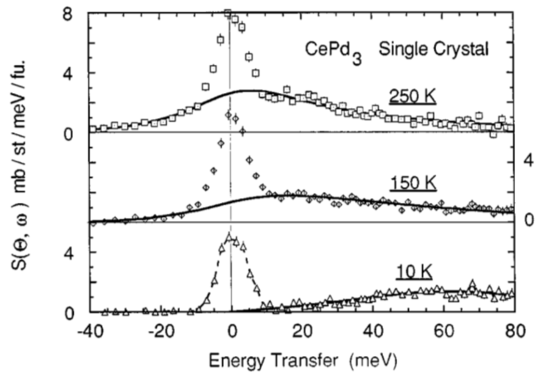
Temperature dependence

- Strong *T* dependence with broad inelastic response at low *T* and quasi-elastic excitations at room temperature



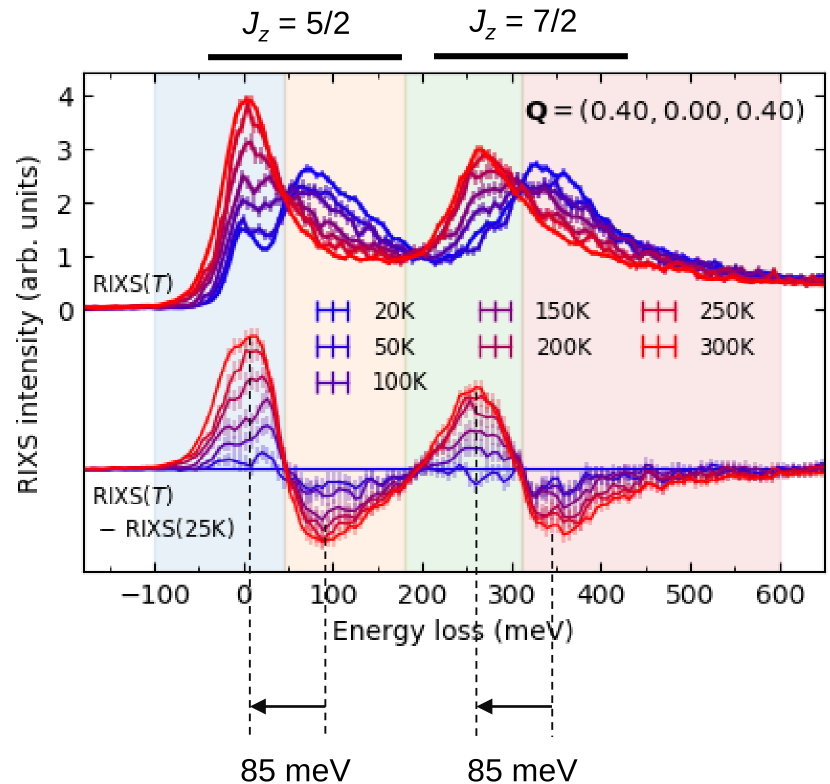
Galera *et al.* J. Magn. Magn. Mat. **63-64**, 594 (1987)

Murani *et al.* PRB **53**, 2641 (1996)

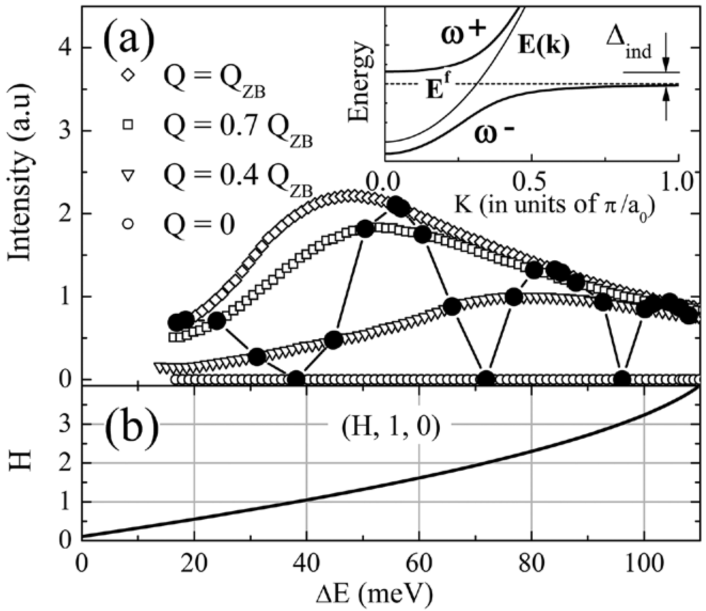


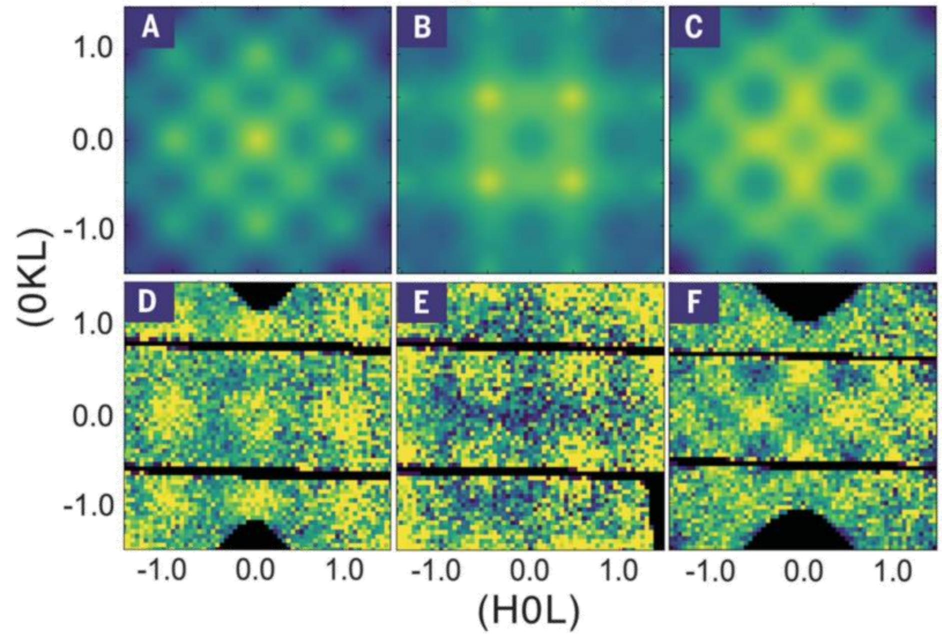
M. Rahn
LANL

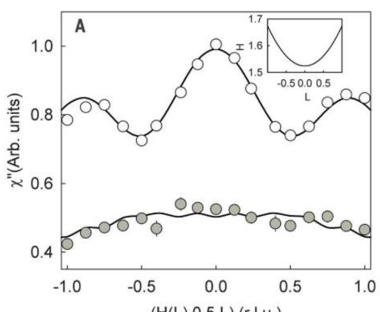
M. Janoscheck
LANL

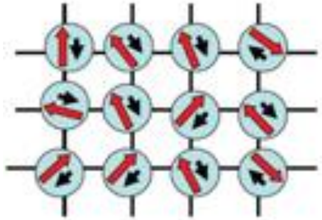


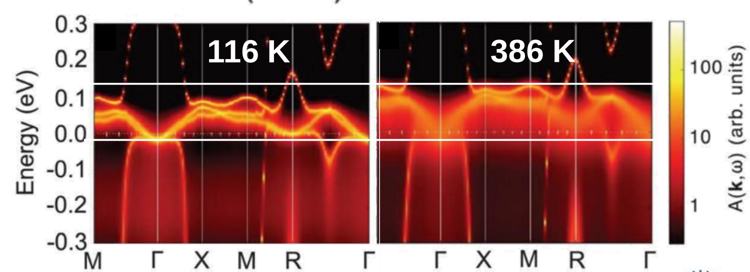
q dependence





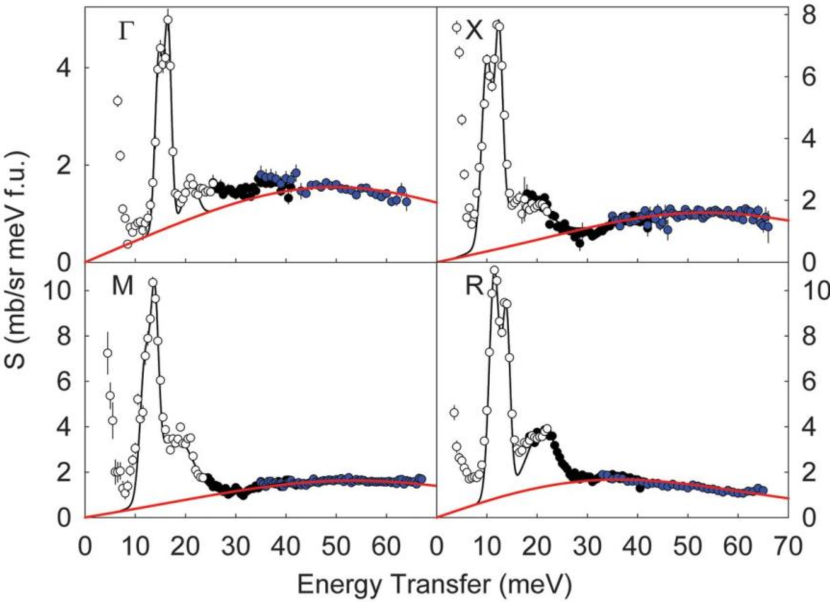


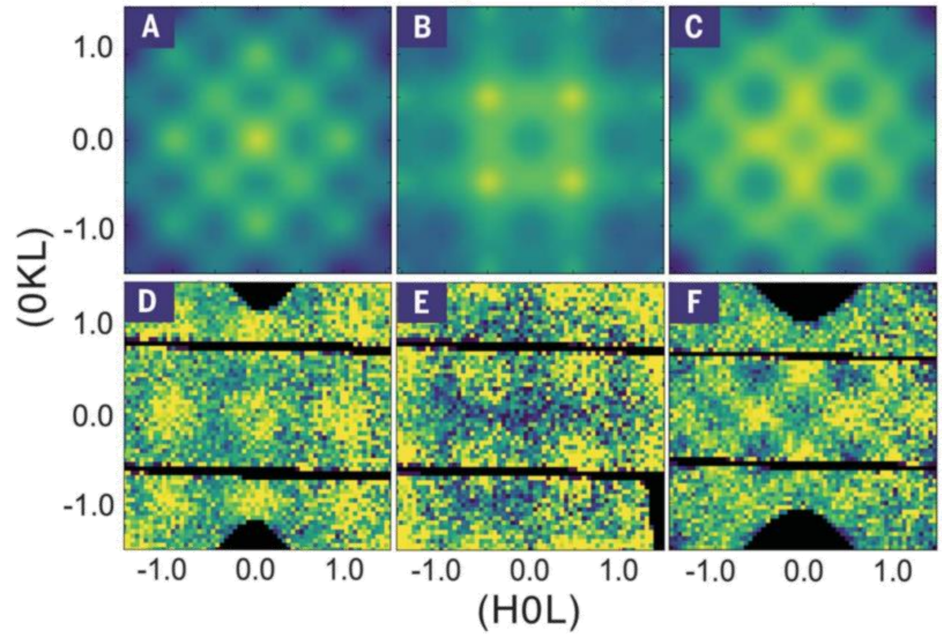


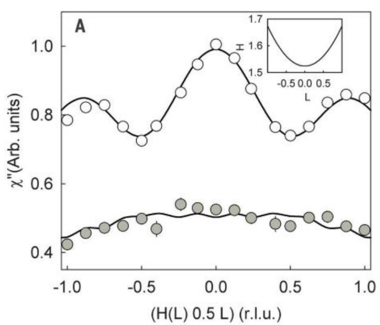


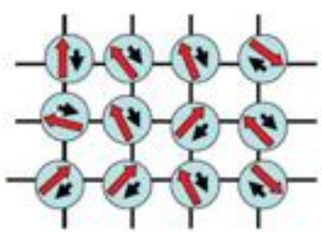
Fanelli *et al.* J. Phys. Cond. Mat. **26**, 25602 (2014)
Goremychkin *et al.* Science **359**, 186 (2018)

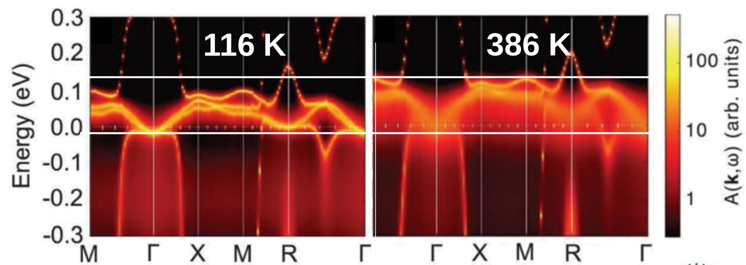
q dependence





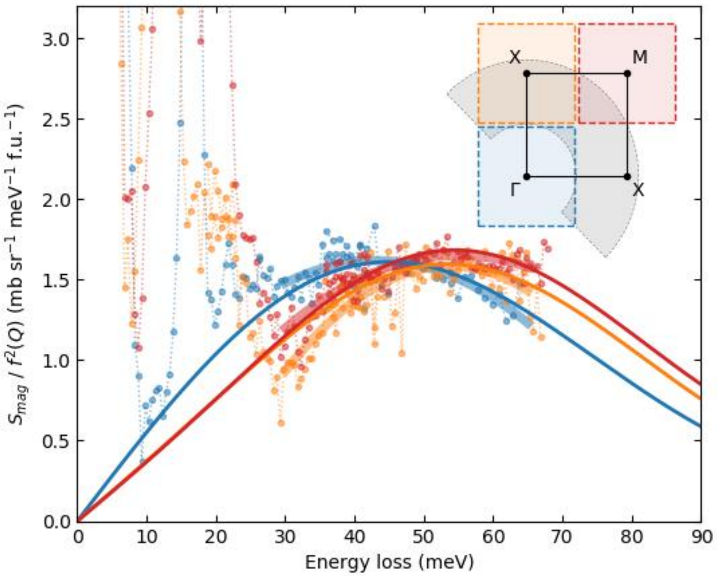


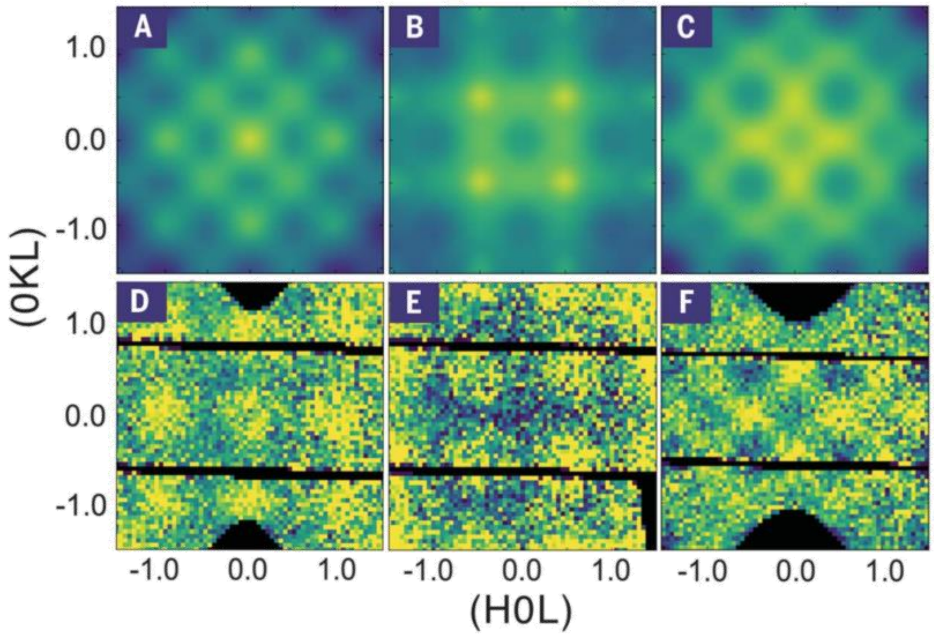


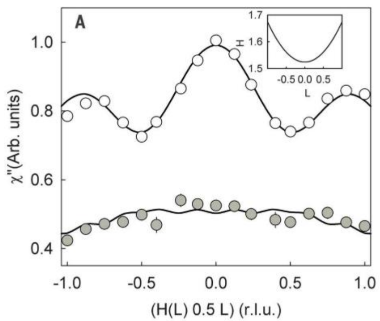


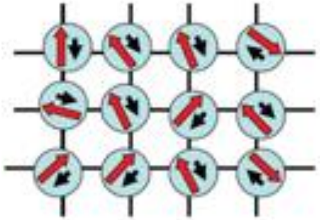
Goremychkin *et al.* Science **359**, 186 (2018)

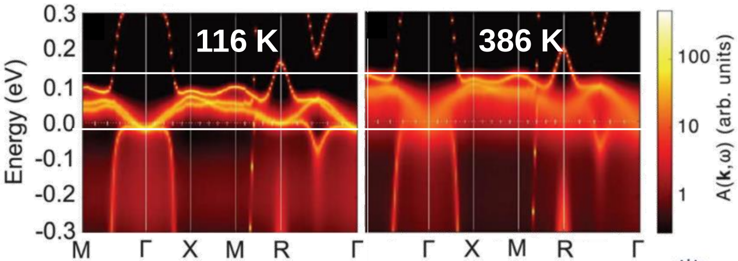
q dependence





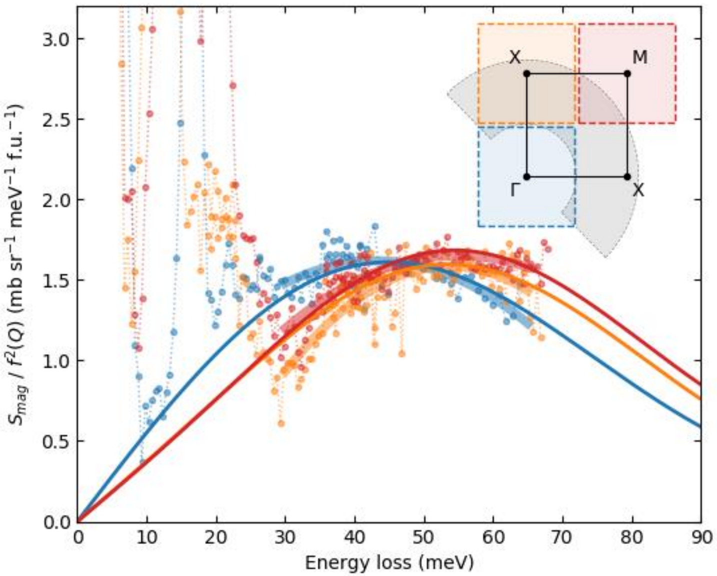


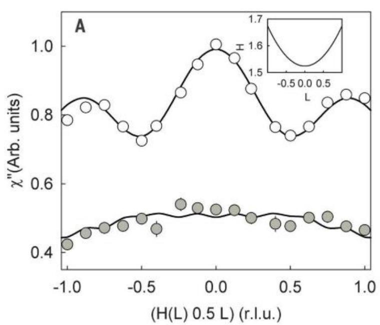


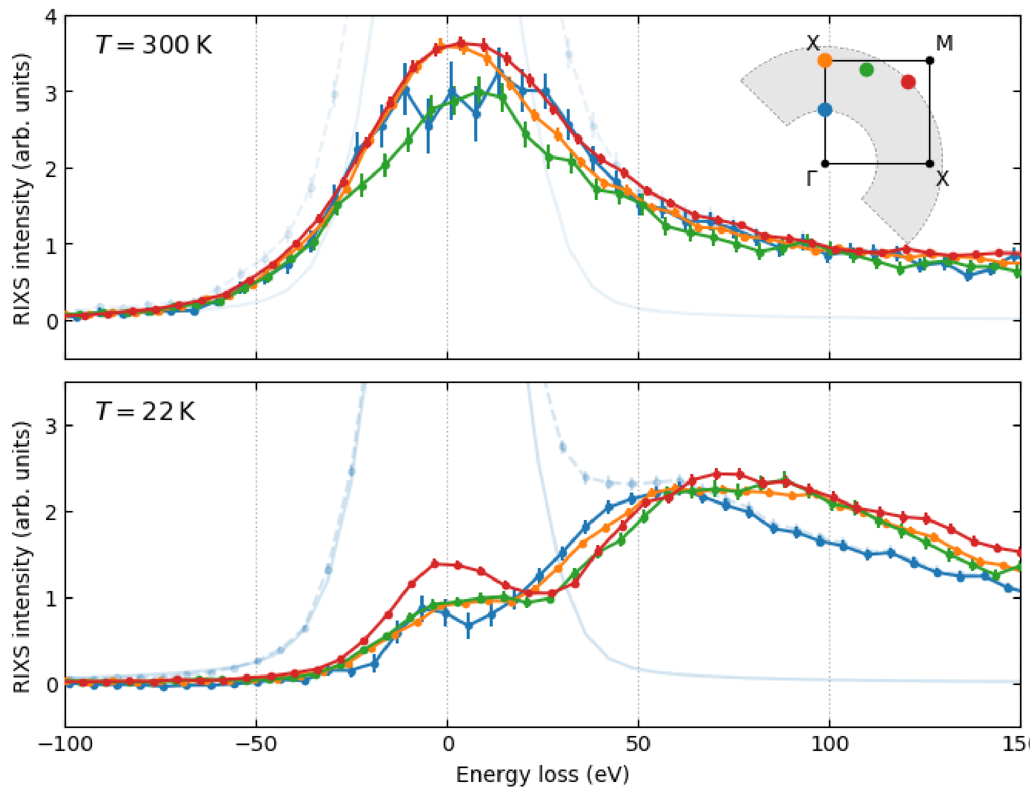


Goremychkin *et al.* Science **359**, 186 (2018)

q dependence

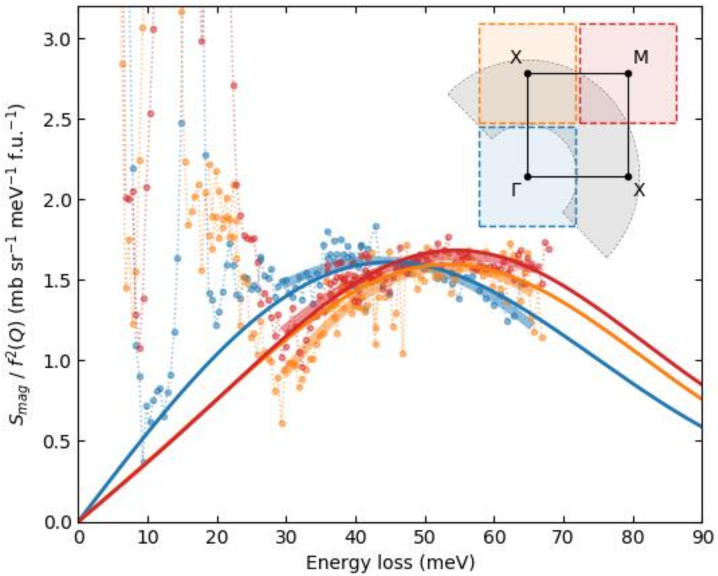


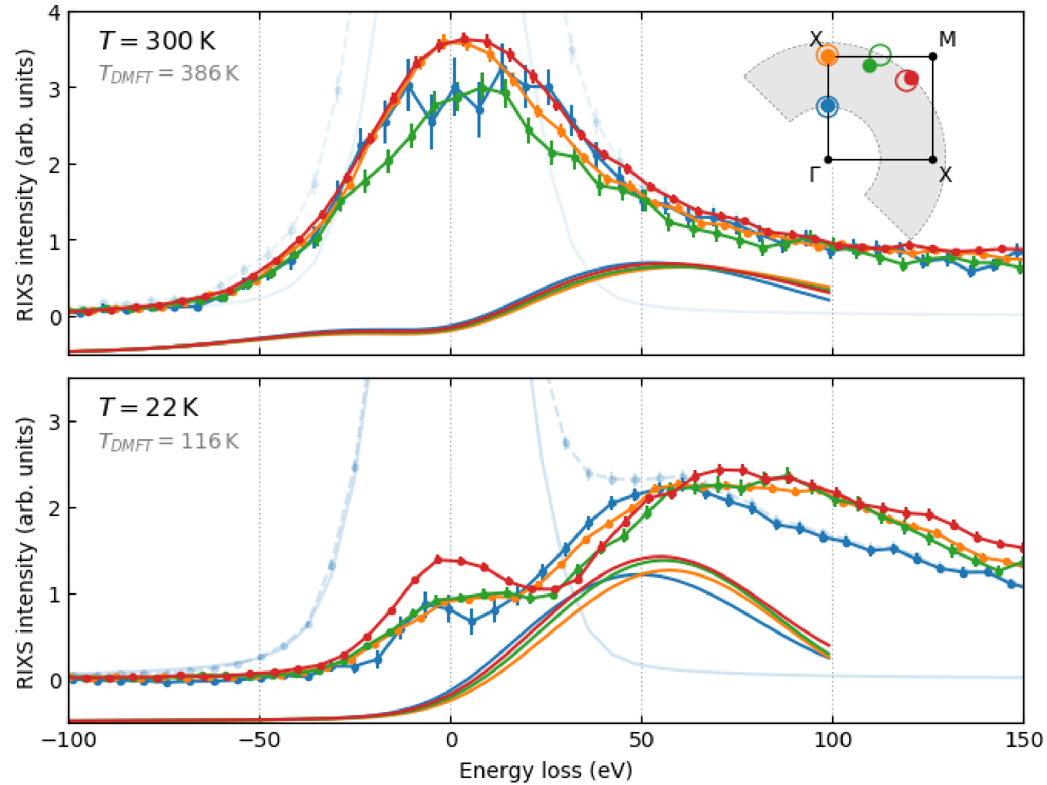


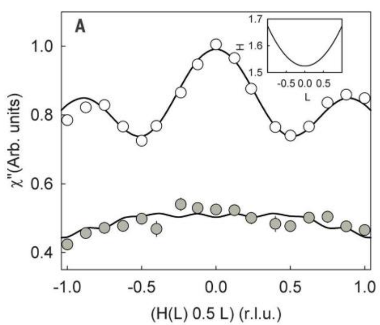


Goremychkin *et al.* Science **359**, 186 (2018)

q dependence

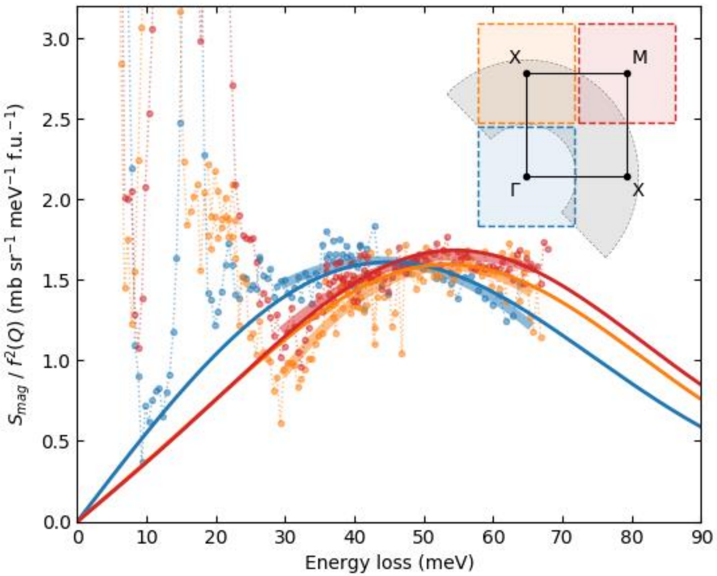


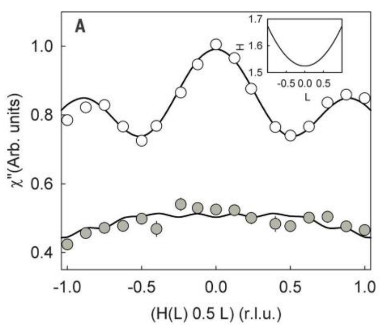


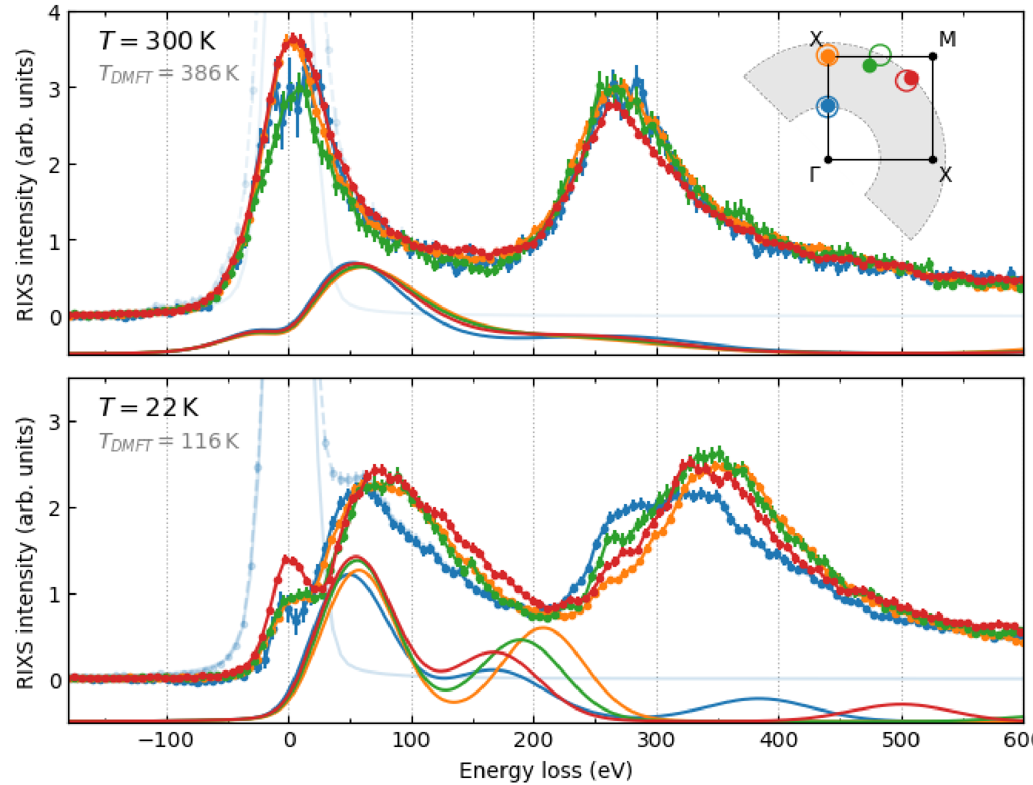


Goremychkin *et al.* Science **359**, 186 (2018)

q dependence

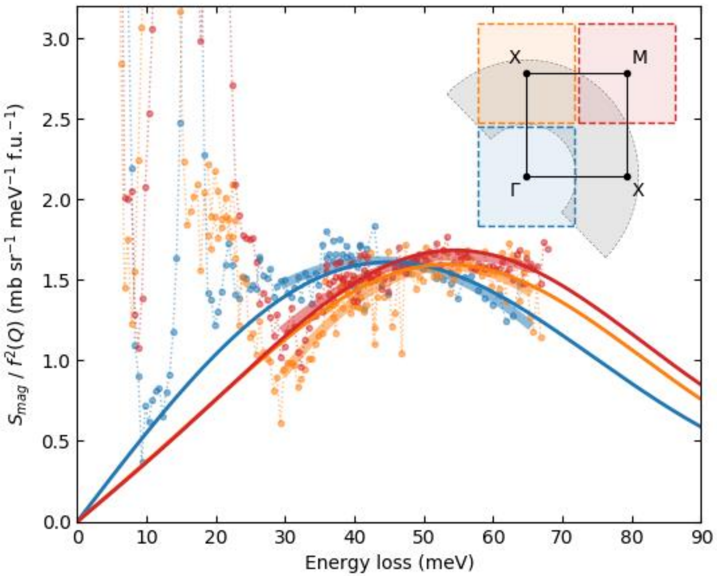


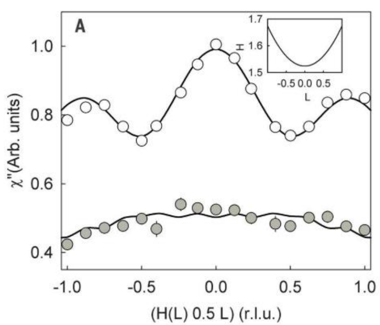




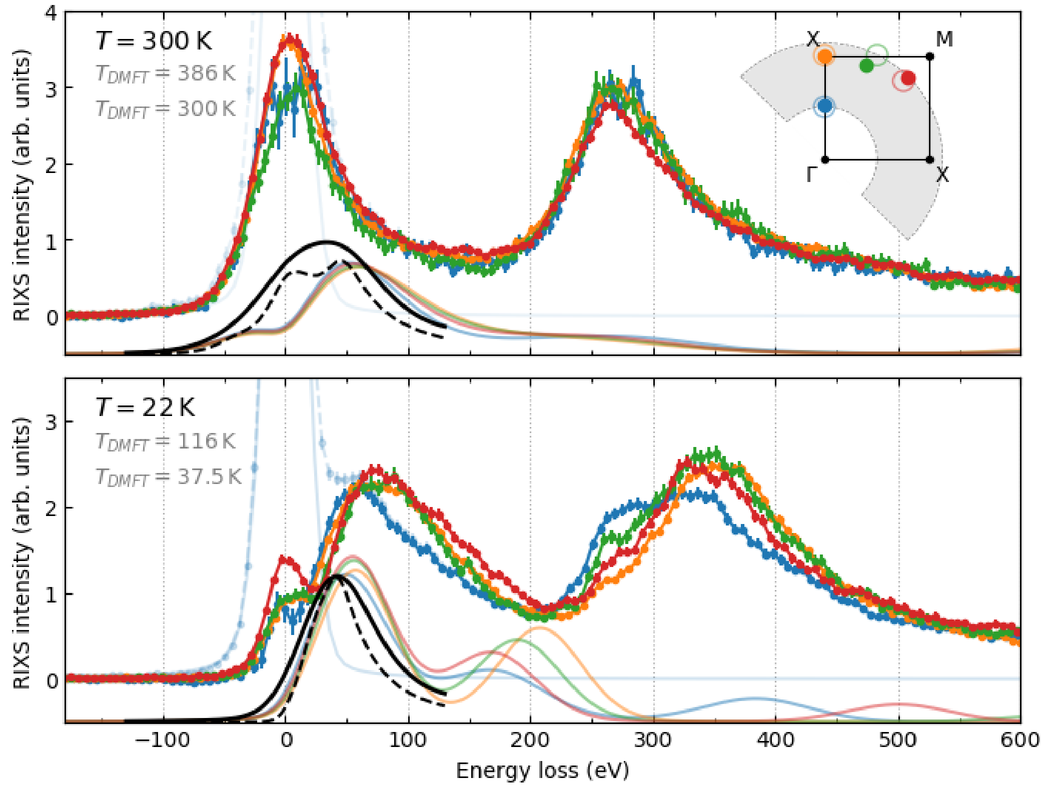
Goremychkin *et al.* Science **359**, 186 (2018)

q dependence





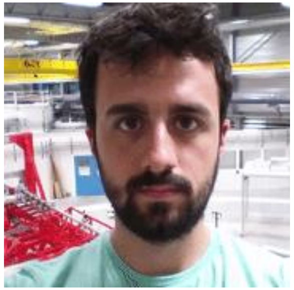
Goremychkin *et al.* Science **359**, 186 (2018)



Sakai *et al.* J. Phys. Soc. Jpn. **79**, 114701 (2010)

Conclusions and outlook

Andrea Amorese



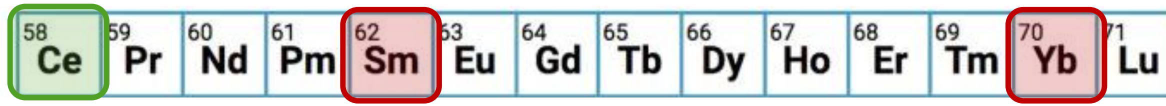
- **Local systems**
  - CEF theory describes RIXS spectra well
  - A more predictive theory for local excitations?
  - Direct information on local vs itinerant character of the 4*f*
- **Lattice systems**
  - Kondo lattices, intermediate-valent systems, systems with sharp valence transitions, …
- **Rare-earth magnets**
- **Dimensionally confined systems, artificial structures, …**
- **Spin-waves in 4*f* metals**

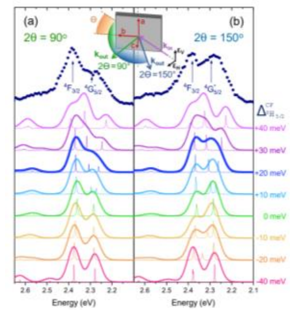
58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu

Conclusions and outlook

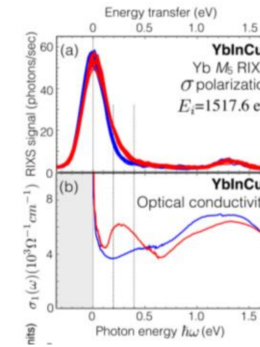
➢ Instrumentation

- Resolution !!!





Amorese *et al.* arXiv:1901.10808

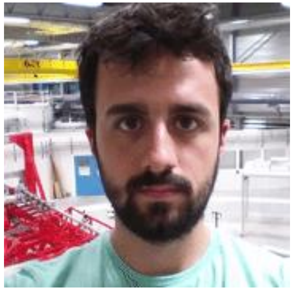


Hancock *et al.* PRB **98**, 075158 (2018)

➢ Theory

- Faster codes for CEF theory.
- Advanced *ab initio* theories for 4*f* systems.
- RIXS experiments can give strong input not accessible to other techniques.

Thank you for your attention!



Andrea Amorese

ESRF
The European Synchrotron

University of Cologne

Marein Rahn

Los Alamos
NATIONAL LABORATORY
EST. 1943

Giacomo Ghiringhelli
Lucio Braicovich

POLITECNICO
MILANO 1863

Denis V. Vyalikh

ikerbasque
Basque Foundation for Science

DIPC
Donostia International Physics Center

Christoph Geibel
Silvia Seiro

MAX-PLANCK-INSTITUT
FÜR CHEMISCHE PHYSIK FESTER STOFFE

Andrea Severing

University of Cologne

Cornelius Krellner
Kristin Kliemt

GOETHE
UNIVERSITÄT
FRANKFURT AM MAIN

Mark Janoschek

Los Alamos
NATIONAL LABORATORY
EST. 1943