Magnetic Scattering in the 4f-Intermediate Valence Compound $CePd_3$

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Introduction

Intermediate valence metals:

- ***Fermi Liquid*** *ground state that evolves towards a high-T* ***local moment*** *regime.*

The ***Anderson lattice model*** *applicable to these compounds, capturing:*

- *band-like* ***coherent character*** *in the low-T limit, and*
- ***local moment*** *character at high-T*

Physical properties reveal the effects of the lattice coherence.

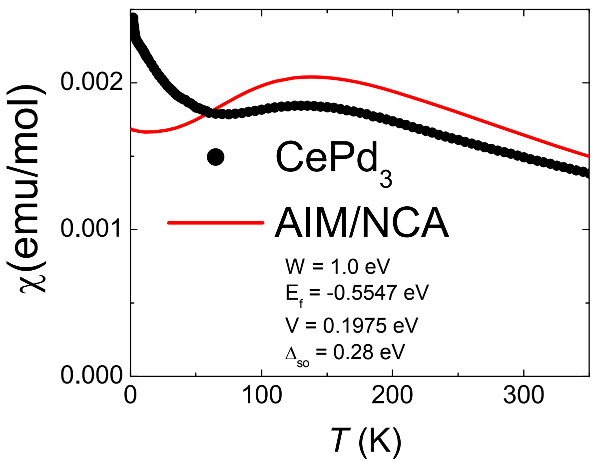
However……..

Other properties, depending on spin fluctuations, are better described by the ***Anderson impurity model****, despite the magnetic moments sit on a lattice.*

Anderson Impurity Model (AIM)

*Good semi-quantitative agreement with **AIM***

High –T **LOCAL MOMENT** → *crossover* → *Low-T* **FERMI LIQUID**



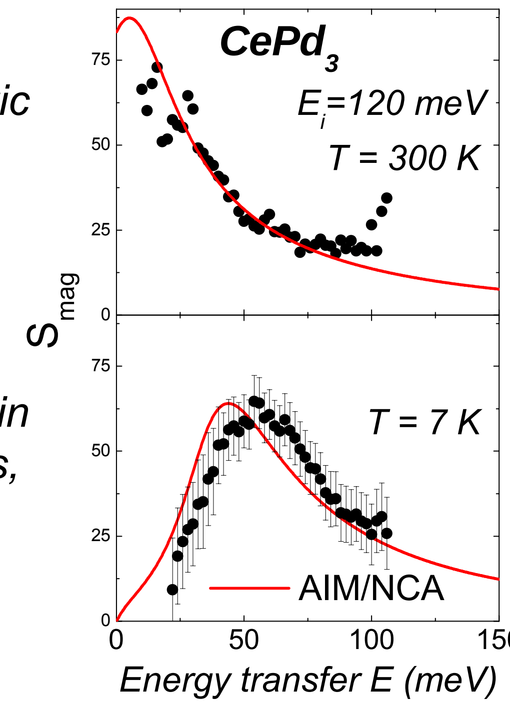
Pauli Paramagnet : finite $\chi(T\rightarrow 0)$ ← *Curie-Weiss: $\chi(T) = C_J / (T + \theta)$*

Non-crossing approx. (NCA)
C. Booth, private communication

Quasi-elastic spectrum
$2\Gamma \sim k_B T_K$

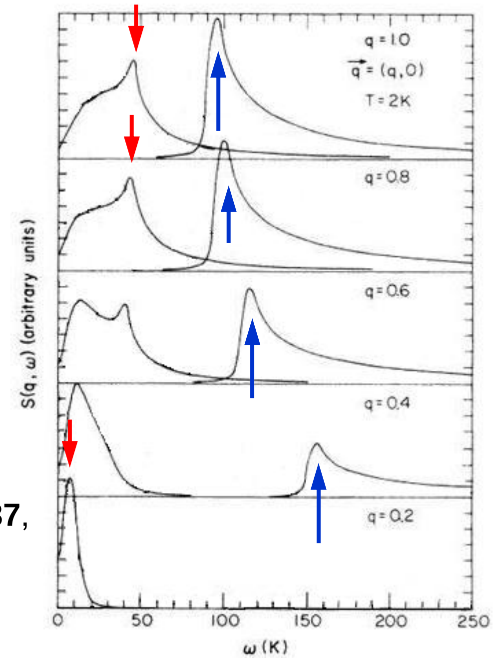
↓

Inelastic spin fluctuations,
$E_0 \sim k_B T_K$

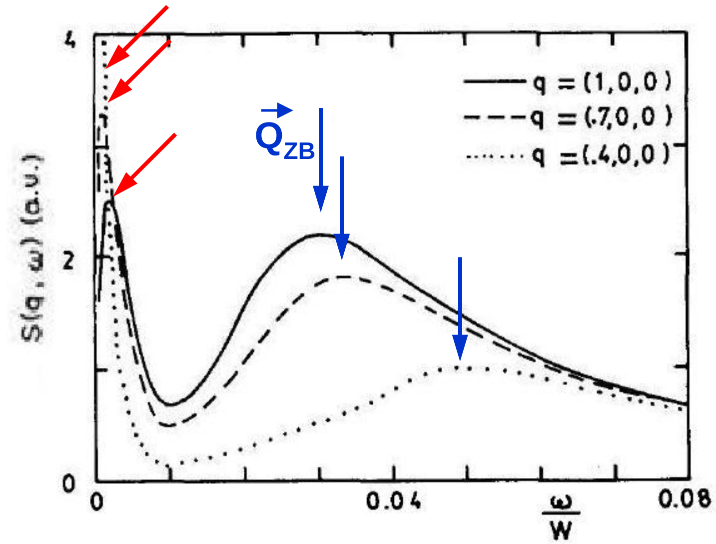


Anderson Lattice Model

Spectrum of S(Q, E) for different values of momentum transfer q



Brandow, Phys. Rev.B **37**, 250 (1988).



Aligia, et al., J. Mag. Mag. Mat. **46**, 321 (1985).

- ***Inter-band** transitions*: *The high energy peak is maximum for q at zone boundary. It shifts to high E with decreasing q.*
- ***Intra-band** transitions*: *The low energy peak moves to high E as q increases towards zone boundary.*

Motivation

magnetic scattering

- *spectral response*
- *dependence over the reciprocal space*

elucidate →

character of the spin fluctuations:

Local / coherent ?

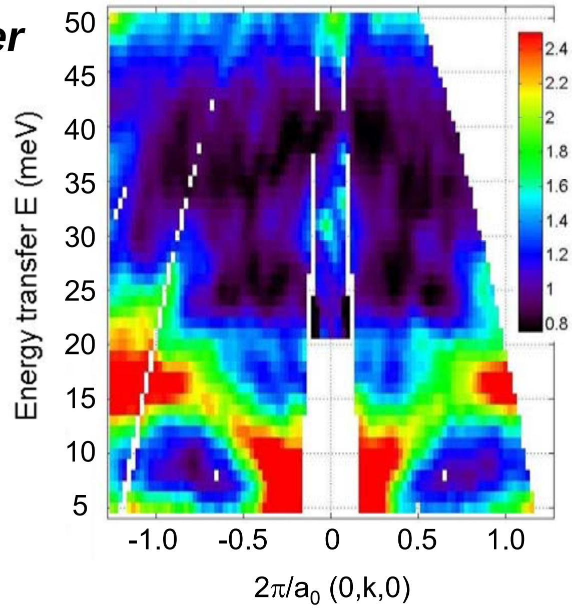
This Q-dependence has remained an open question

- *Most of INS performed on polycrystalline samples.*
- *Only four compounds studied by INS on single crystals*

Magnetic and non-magnetic Scattering

MAPS spectrometer

7 K, E_i=60 meV



Phonon Scattering *dominant*

Multiple Scattering

(+ Multi-phonons)

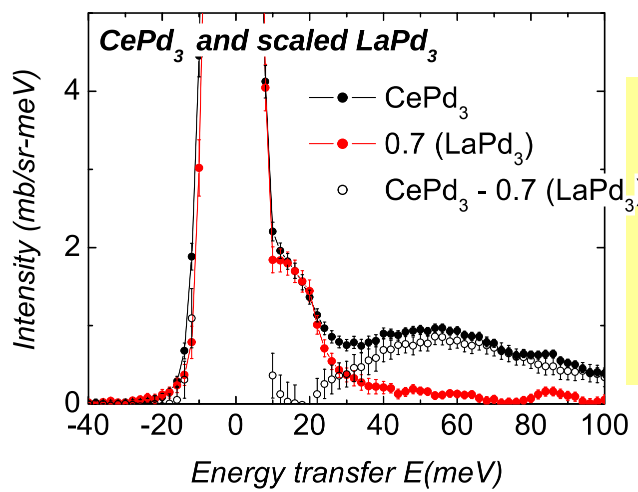
and

Magnetic scattering

Non-magnetic Scattering

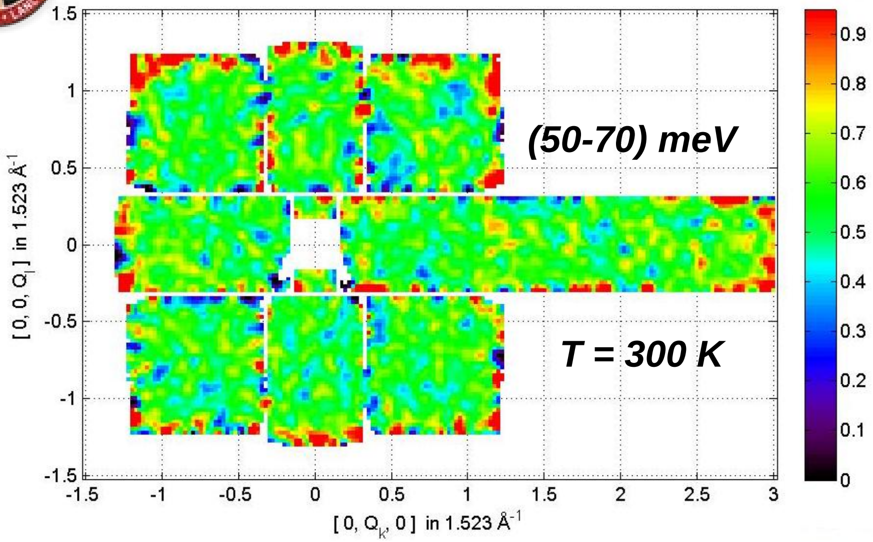
Represented by

"scaled $LaPd_3$"



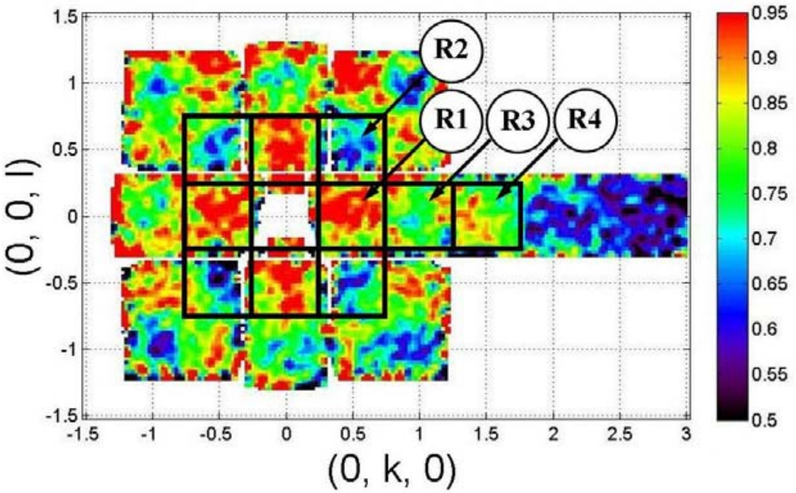
Difference should account for *magnetic contribution* to the scattering

Inelastic Neutron Scattering on $CePd_3$



Distribution of the intensity over momentum transfer space:

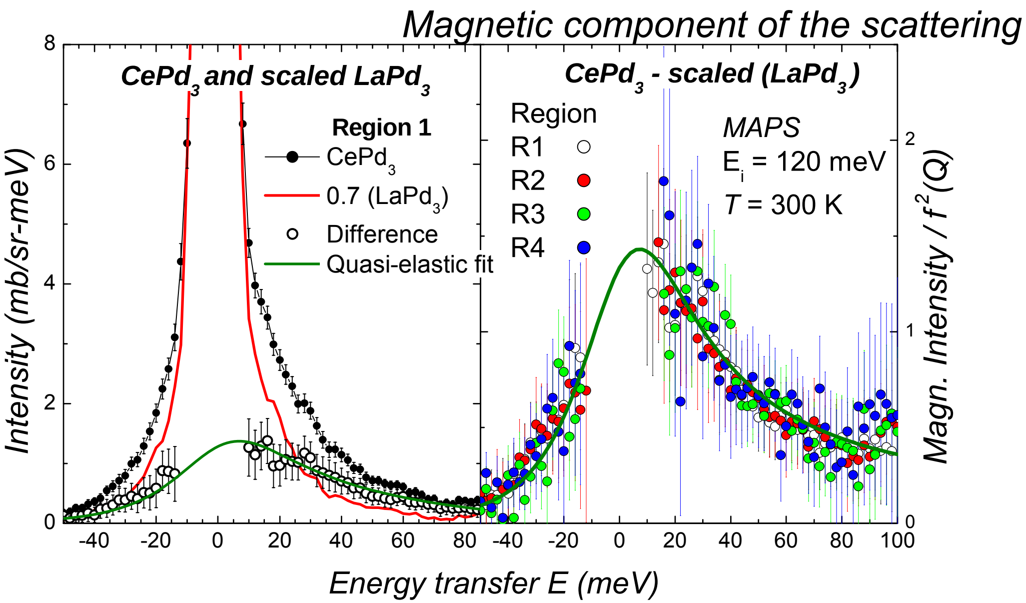
- *Relatively uniform at room temperature*

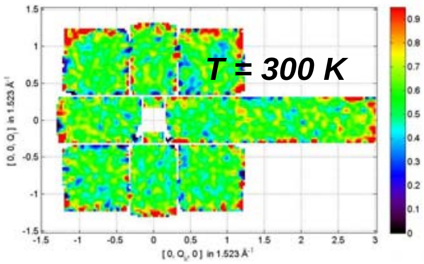


To explore the Q-dependence

Look at different regions in Q-space

$CePd_3$ at 300 K (~ ½ T_K)





- Quasi-elastic Lorentzian
- Q-independent

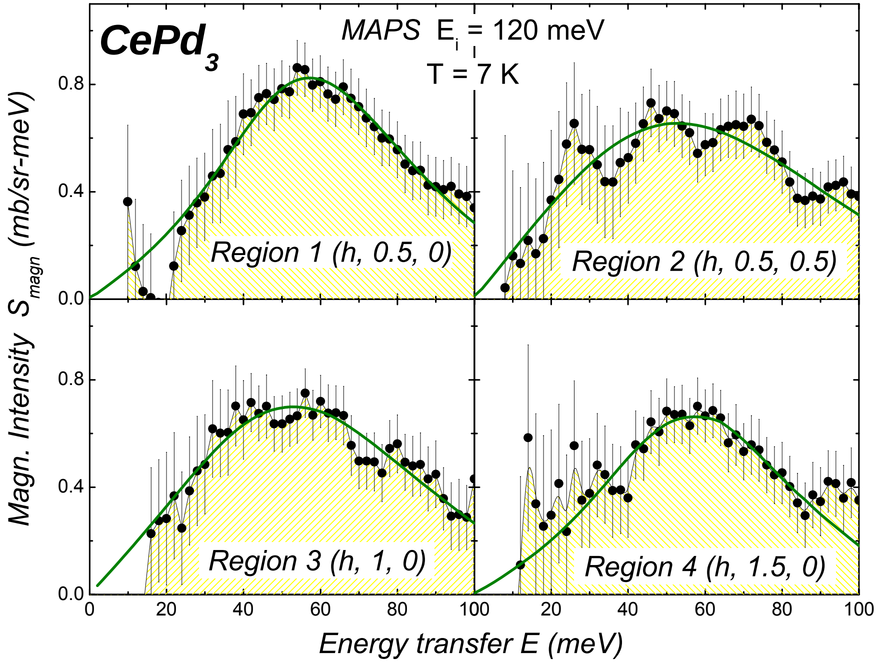
$$S_{magn}(Q,E) = \frac{1}{(1-e^{-E/k_B T})} f^2(Q) \frac{E A_L}{\pi} \frac{\Gamma}{E^2+\Gamma^2}$$

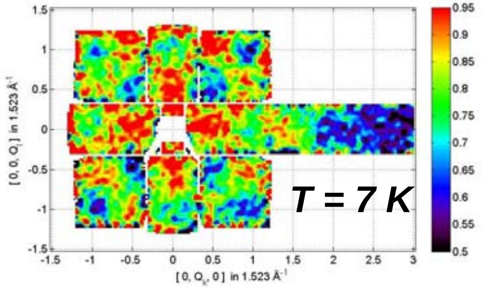
with Γ = 27 meV

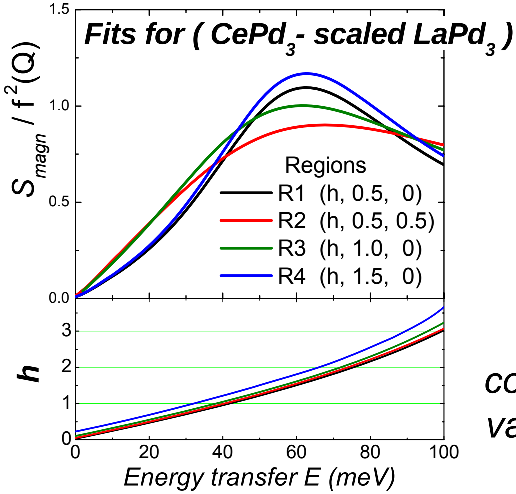
*Q-independence implies a **local character** for spin fluctuations as expected for high-T **uncorrelated local moment** limit*

CePd$_3$ at 7 K (Low-T regime)

Magnetic scattering of CePd$_3$ at four regions in the (k,l) plane







In TOF: component h varies with E

- *Variations of intensity ~ 25%*
- *Similar Inelastic Lorentzians*

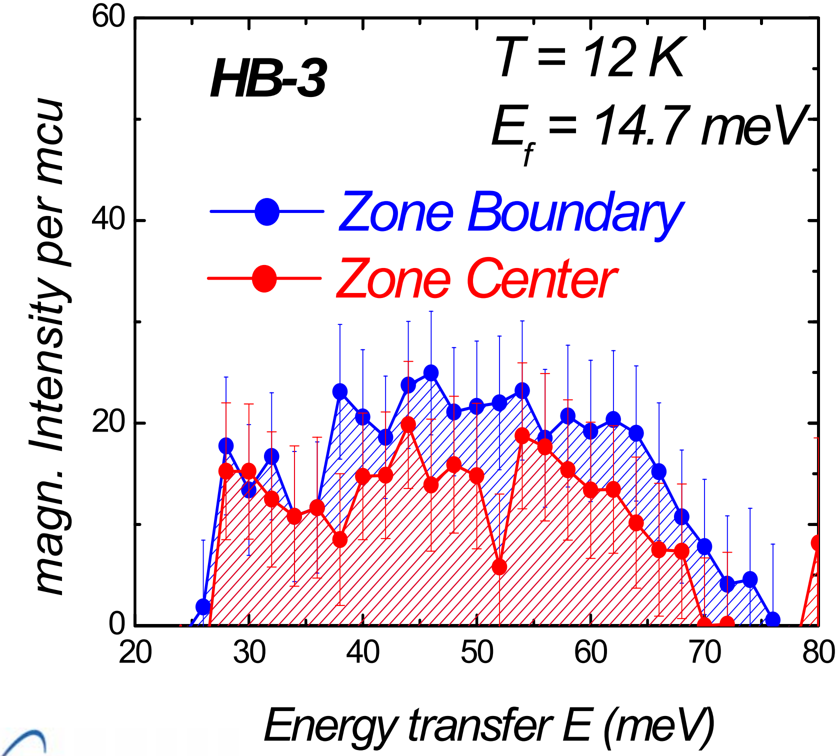
Average: Γ = 43.5 meV
E_0=45 meV

Q-dependence not as drastic as expected by Anderson Lattice model (NO shift of spectral weight)

$CePd_3$ (Low-T)
Triple-axis spectrometer

Constant-Q scans:

- at the Brillouin **zone boundary** (2.5, 1.5, 0)
- at the Brillouin **zone center** (2, 2, 0)



Again:

Difference between ZB and ZC is **NOT** at all as the pronounced variation predicted by the ALM

Discussion / Conclusions

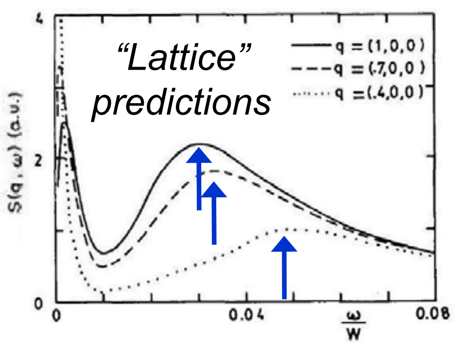
Conclusions (i)

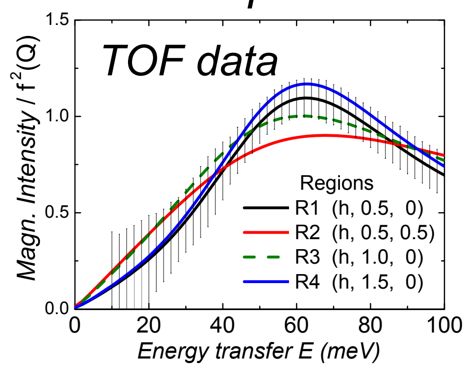
1) High-temperature magnetic scattering:

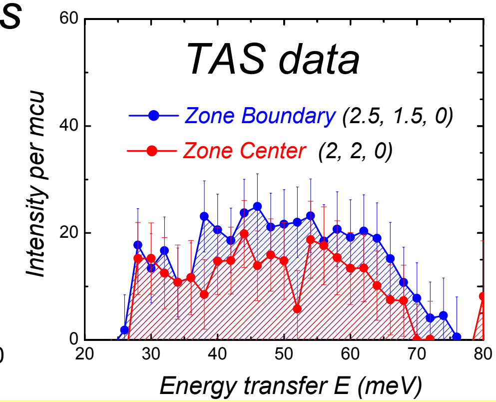
Q-independent, *quasi-elastic spectrum proving*
Local *character of* ***uncorrelated magnetic moments***

2) Low-temperature magnetic scattering:

- *Broad Inelastic Lorentzian + weak Q-dependence*
- ***Qualitatively different*** *from ALM predictions*







Much more like the ***predictions of the AIM***

Similar "Kondo-like" spectral response: { ***$YbAl_3$*** *,(Christianson, PRL,2006)* ***$YbInCu_4$*** *(Lawrence, PRB 1997),* ***$CeInSn_2$*** *, (Murani, PRL 2008)*

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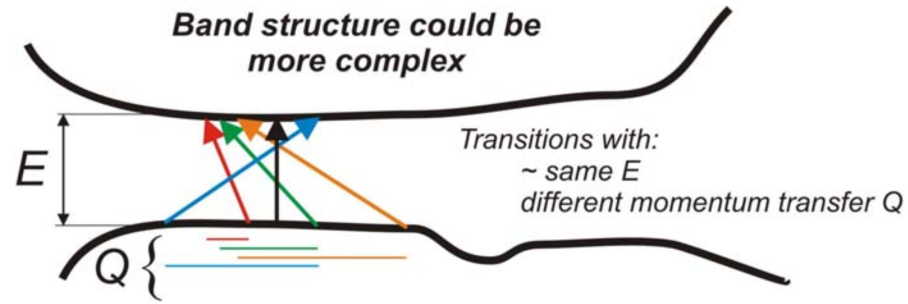
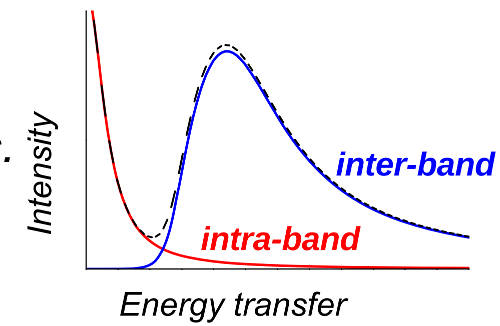
Conclusions (ii)

Possible reasons for the failure of ALM predictions for spin dynamics of IV comp.

a) **Band structure more complex** than just two hybridized bands.
These bands remain to be calculated.



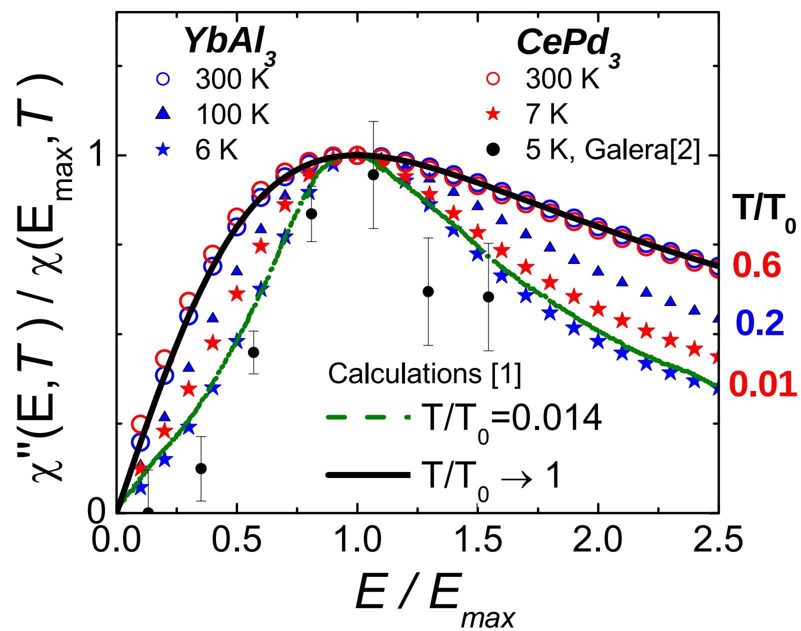
b) There may be significant **overlap** of the **intra-band** Fermi Liquid excitations with the **inter-band** transitions.
We are uncertain about the energy scale of the former (above 10 meV?)

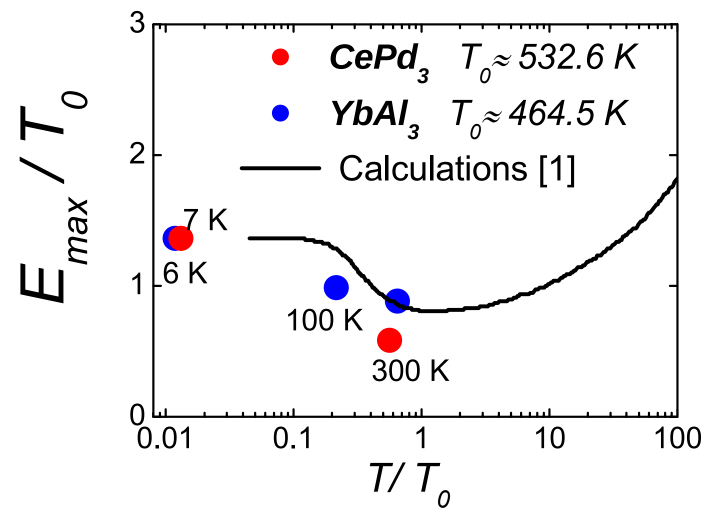


c) ALM approximations may require improvements. Some are versions of mean field (MF) approx. which do not necessarily get the **excitation energies away from the** ε_F correctly.

Conclusions (iii)

3) Evolution with temperature: *Agreement with AIM calculations*





[1] Cox, Bickers, Wilkins, J. Appl. Phys. **57**, 1 (1985)

[2] Galera, *et al.*, J. Magn. Magn. Mat. **47-48**, 139 (1985).

Excitations in the Anderson lattice are much more like those of an Anderson ***impurity*** *than has been previously recognized*

Thank you!

Supporting Slides

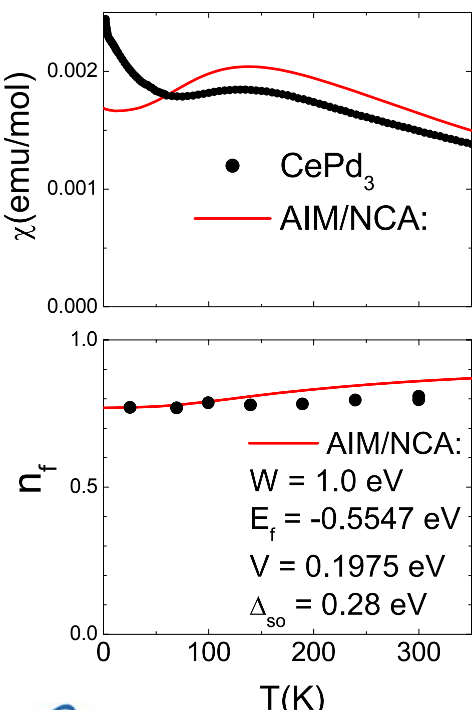
Anderson Impurity Model (AIM)

*Even though RE's ions sit on a **lattice**, good semi-quantitative agreement with **AIM***

High –T
LOCAL MOMENT

crossover →

Low-T
FERMI LIQUID



Curie-Weiss:
$\chi = C_J / (T + \theta)$

↓

Pauli Paramagnet : finite $\chi(0)$

Integral valence ($n_f \rightarrow 1$)

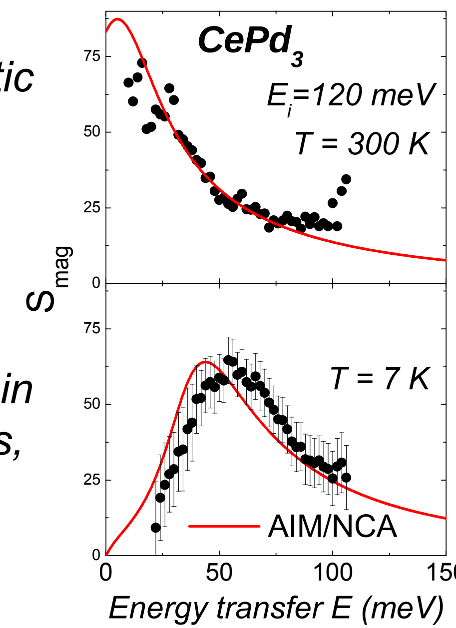
↓

Non-integral valence

Quasi-elastic spectrum
$2\Gamma \sim k_B T_K$

↓

Inelastic spin fluctuations,
$E_0 \sim k_B T_K$



Non-crossing approx. (NCA)
C. Booth, private communication

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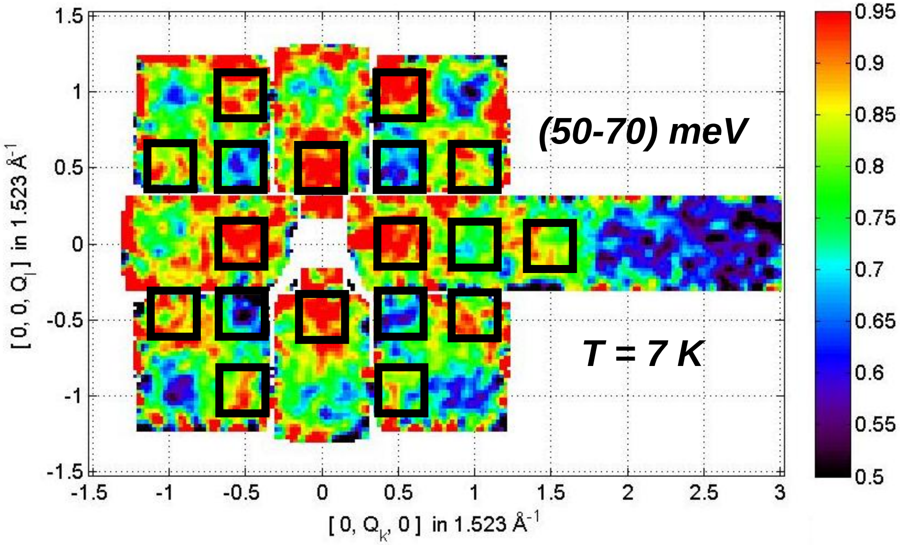
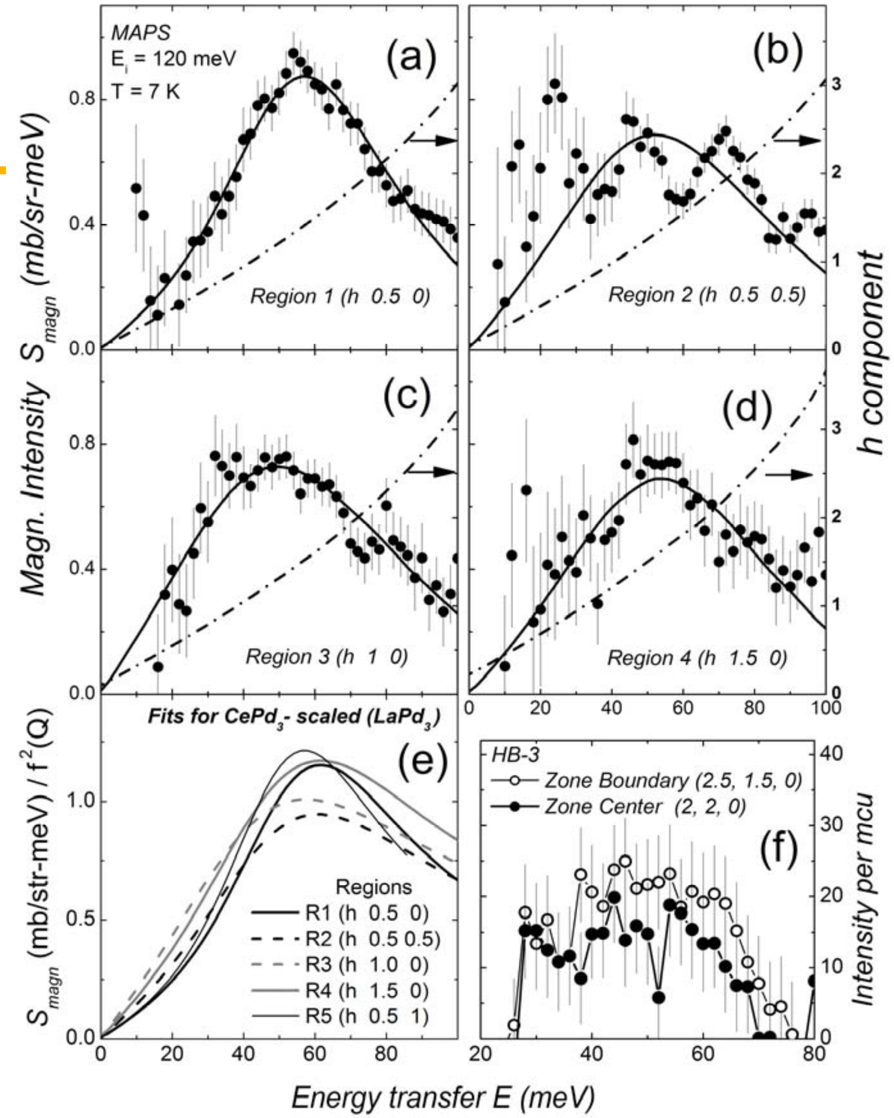
Smaller Q-Regions





Q-region	T (K)	E_0 (meV)	Γ (meV)
Region 1	7	52.6 ± 0.8	32 ± 1
Region 2	7	42 ± 5	41 ± 5
Region 3	7	37 ± 3	44 ± 4
Region 4	7	46 ± 2	41 ± 4
average	7	46 ± 3	39.5 ± 4
all regions	300	0	23.3 ± 0.8

Conclusions (i)

1) High-temperature magnetic scattering:

Q-independent *quasi-elastic Lorentzian spectrum*

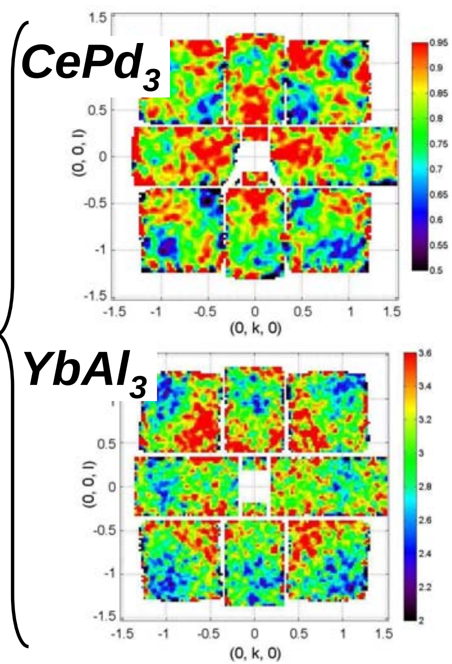
proving

Local *character of spin fluctuations for* ***uncorrelated magnetic moments***

2) Low-temperature magnetic scattering:

(inelastic Lorentzian + weak Q-dependence)

Much more like the ***predictions of the AIM*** *than the ALM.*



Similar "Kondo-like" spectral response:

$YbAl_3$, Christianson, et al, Phys. Lett.96,117206 (2006)

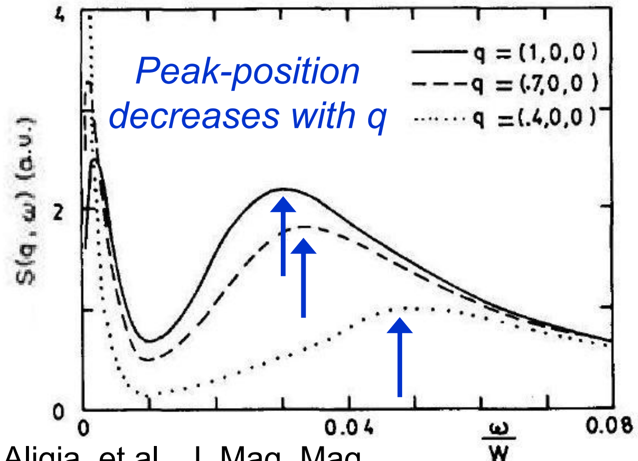
*$YbInCu_4$, Lawrence, et al, Phys. Rev. B **55**, 14467 (1997)*

*$CeInSn_2$, Murani, et al, Phys. Rev. Lett. **101**, 206405 (2008)*

Conclusions (iii)

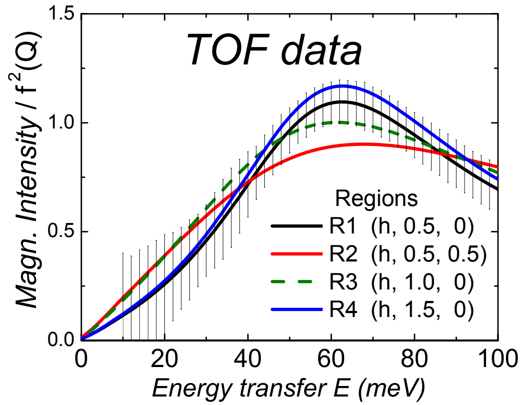
3) Low-T magnetic scattering qualitatively different from ALM predictions

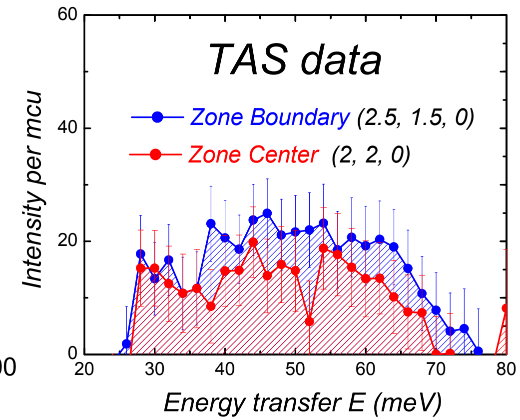
"Lattice" predictions



Aligia, et al., J. Mag. Mag. Mat. **46**, 321 (1985).

$CePd_3$ at low-T, for different Q's



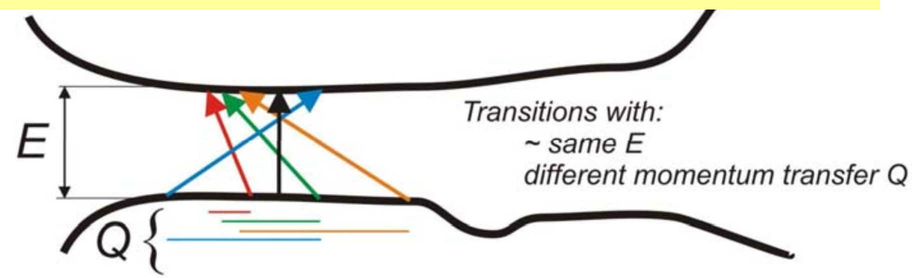


- ***Q-dependence*** *not at all as severe as ALM prediction*
- ***No gap*** *in the scattering*

Band structure more complex

than just two hybridized bands.

These bands remain to be calculated.



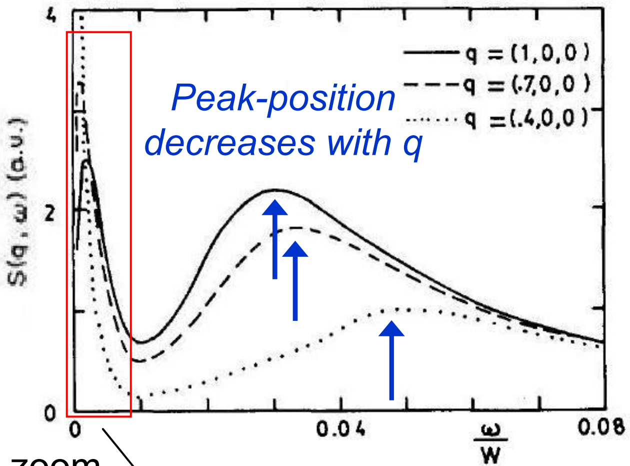
Values for $CePd_3$ using big Q-regions

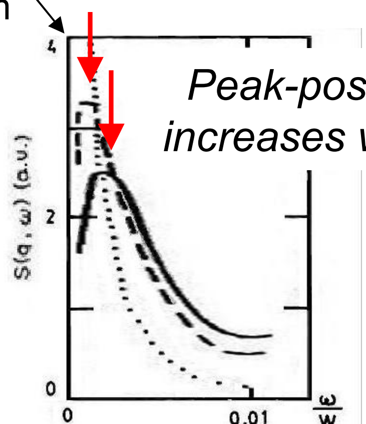
Q–region	T (K)	E_0 (meV)	Γ (meV)	χ_{DC} (10^{-3} emu/mol)
Average value	8	45 ± 2	43.5 ± 4	1.93 ± 0.2
All zones	300	0	26.6 ± 0.7	1.98 ± 0.03

Conclusions (iii)

Low-T magnetic scattering differs from the predictions of the ALM.

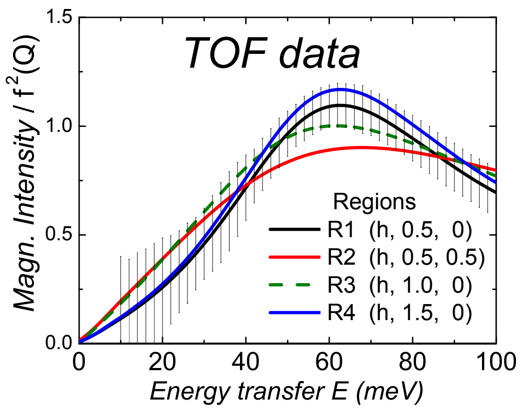
"Lattice" predictions
S(Q,E) for different Q's

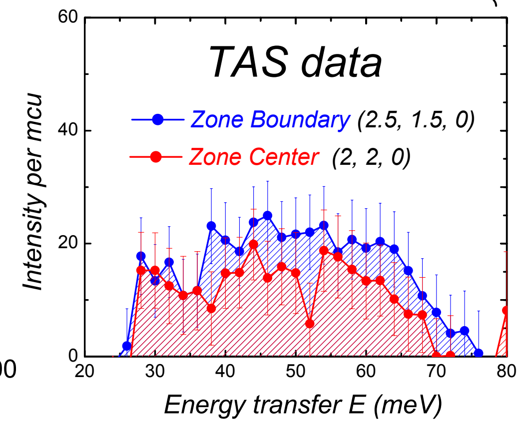




Aligia, et al., J. Mag. Mag. Mat. **46**, 321 (1985).

Magnetic Intensity spectra:
***$CePd_3$** at low-T, for different Q's*





- ***Q-dependence*** *not at all as severe as ALM prediction*
- ***No gap*** *in the scattering*
  (Overlap of intra- and inter-band transitions?)

Anderson Lattice Model (ALM)

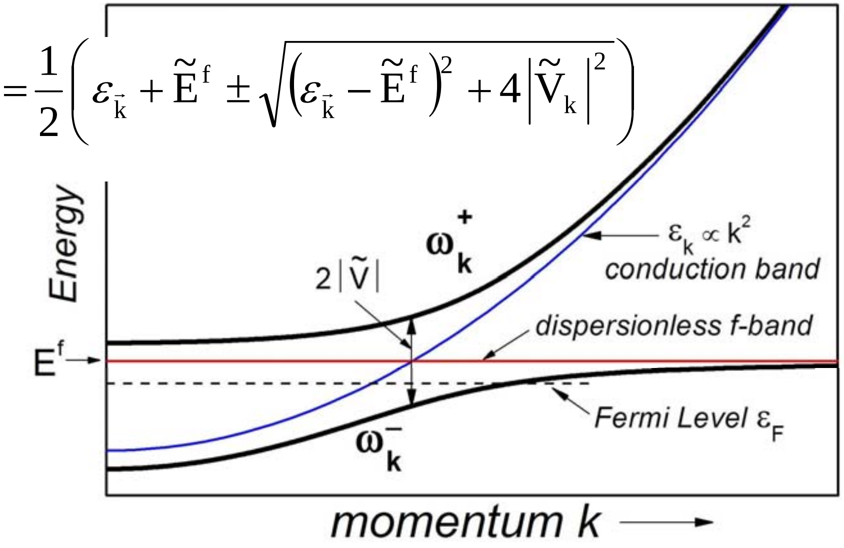
$$\mathbf{H} = \sum_{\vec{k},\sigma} \varepsilon_{\vec{k}} C^{+}_{\vec{k},\sigma} C_{\vec{k},\sigma} + \sum_{i} \left[\sum_{\sigma} E^{f} f^{+}_{i,\sigma} f_{i,\sigma} + \sum_{\vec{k},\sigma} \left(V_k f^{+}_{i,\sigma} C_{\vec{k},\sigma} + V^{*}_{k} C^{+}_{\vec{k},\sigma} f_{i,\sigma} \right) + U\, f^{+}_{i,\uparrow} f_{i,\uparrow} f^{+}_{i,\downarrow} f_{i,\downarrow} \right]$$

Dispersion of the hybridized bands.

$$\omega^{\pm}_{\vec{k}} = \frac{1}{2}\left(\varepsilon_{\vec{k}} + \tilde{E}^{f} \pm \sqrt{\left(\varepsilon_{\vec{k}} - \tilde{E}^{f}\right)^{2} + 4\left|\tilde{V}_{k}\right|^{2}} \right)$$

The direct gap has a value $2\tilde{V}$

Coulomb interaction U renormalizes the parameters V and E^f.

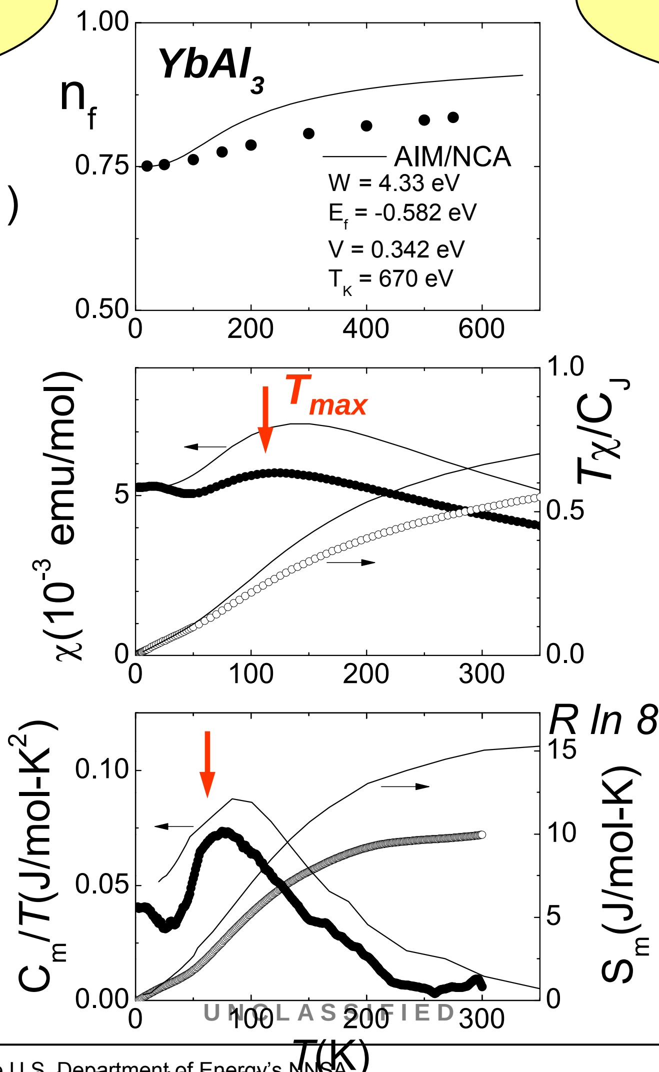


Basic Phenomenological Description

Low-T
FERMI LIQUID

crossover to

High –T
LOCAL MOMENT PARAMAGN



- Non-integral valence (n_f <1)
- Pauli Paramagnet

  finite $\chi(0) \sim \mu_B^2 N(\varepsilon_F)$
- Linear Specific Heat

  $C(T) \sim \gamma T$

  $\gamma = 1/3\ \pi^2 N(\varepsilon_F)\ k_B^2$

- Integral valence ($n_f \rightarrow 1$)
- Curie-Weiss: $\chi = C_J / (T + \theta)$
- Entropy: $S_m \rightarrow R\ \ln(2J+1)$

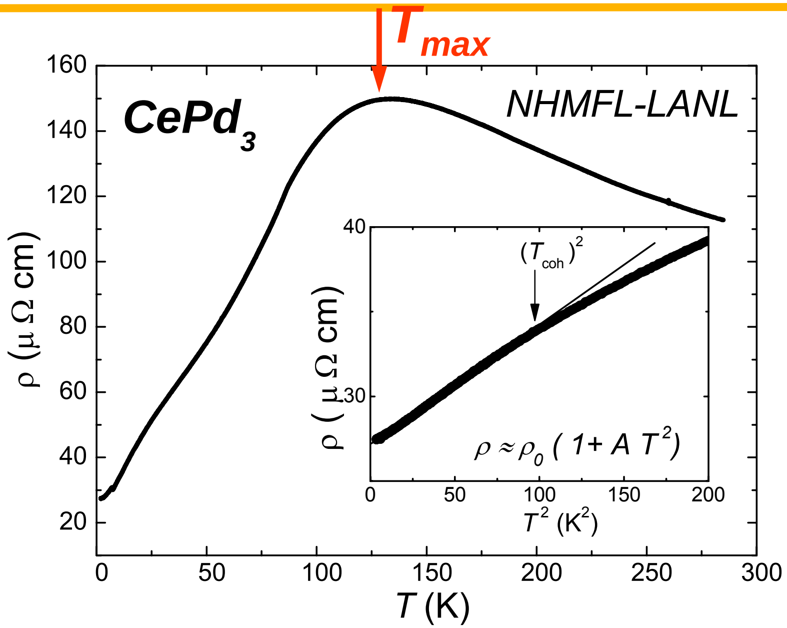
Cornelius, et al. Phys. Rev. Lett. **88**, 117201 (2002)

Basic Phenomenological Description

- *Onset of* ***coherence***

  i) Bloch's law

  ii) $\rho \propto T^2$



- ***Scaling***

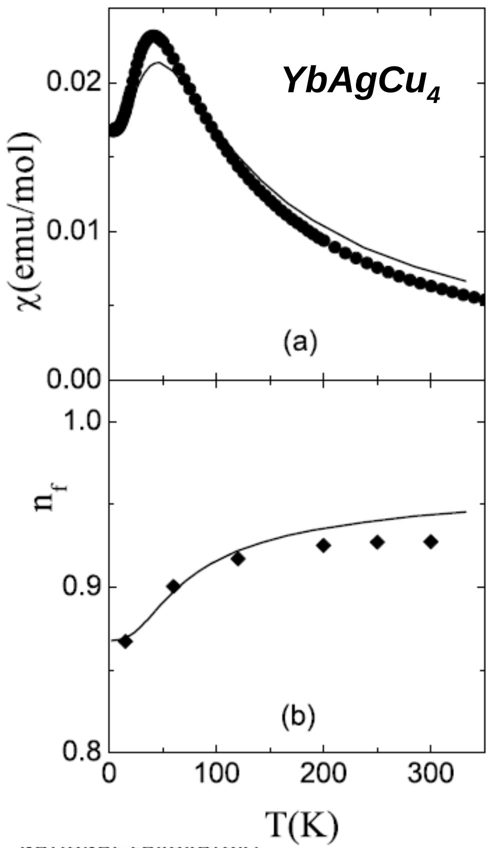
  $1/\chi_0,\ 1/\gamma,\ \Gamma,\ E_0 \propto k_B T_{max}$

  Thermodynamic properties are universal function of a scaled temperature T_{max}

Anderson Impurity Model (AIM)

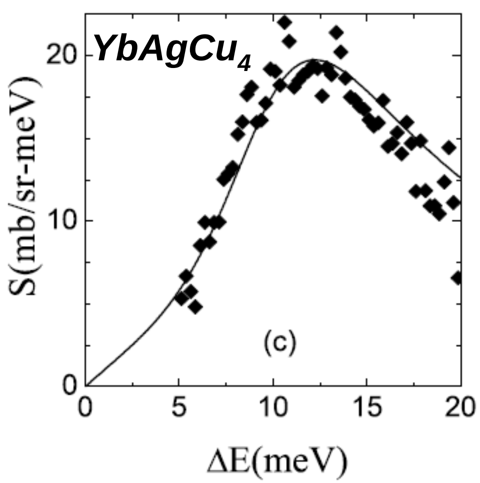
*Even though RE's sit on a lattice, and are not "impurities", good agreement with **AIM***

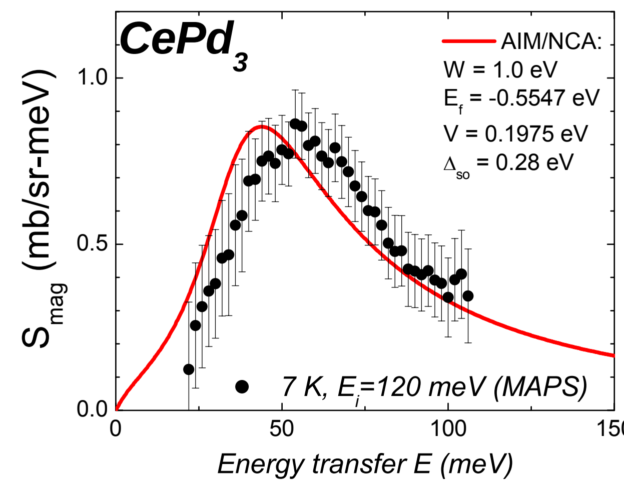
- ***Qualitative agreement***
- *Some quantitative agreement*
- *Slower crossover than AIM prediction*



Non-crossing approx. (NCA)

Lawrence et al, Phys. Rev. B **63,** 054427 (2001).





Calculation: C. Booth, private communication

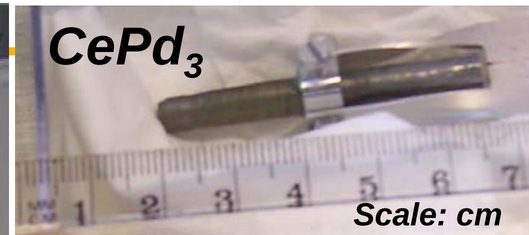
Single-crystalline samples

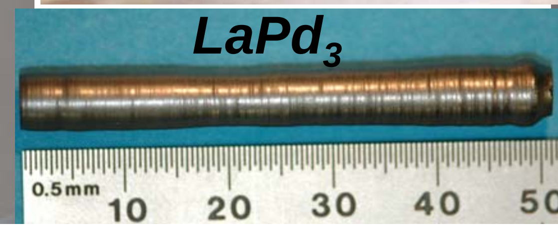
$CePd_3$ (18 g) $LaPd_3$ (11 g)

Grown by Czochralski method

Annealed at 950°C (6 days)

E. D. Bauer,
C. H. Wang,
K. J. McClellan



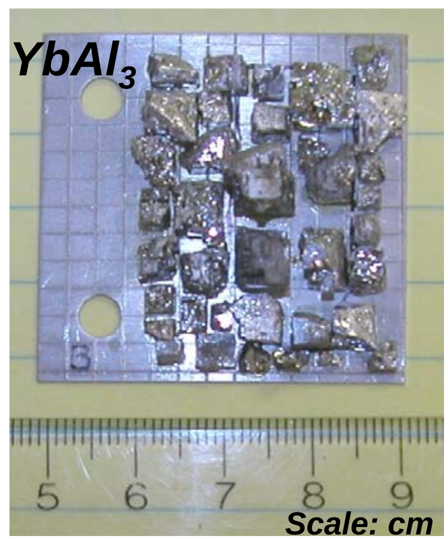


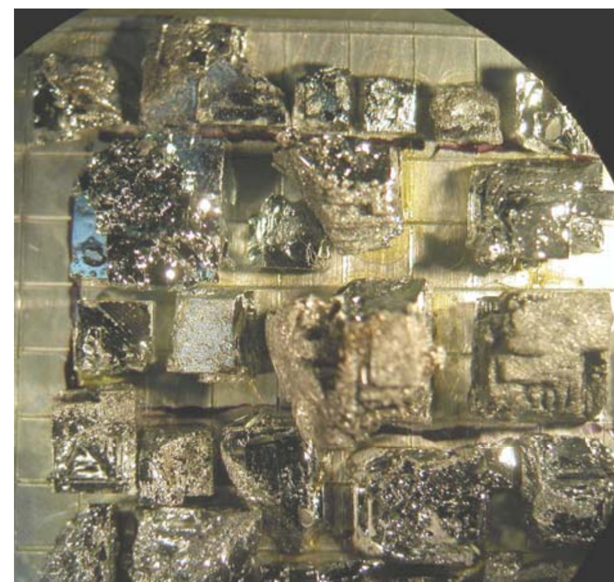
$YbAl_3$ ($\approx$ 5 g)*

Grown by self-flux method

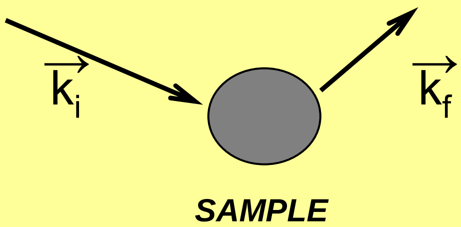
A. D. Christianson,
E. D. Bauer

* *In the pictures:* ***not*** *the actual set-up used in the experiments reported in this work*

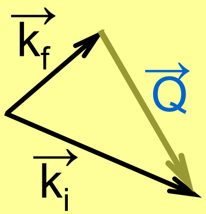




Inelastic Neutron Scattering



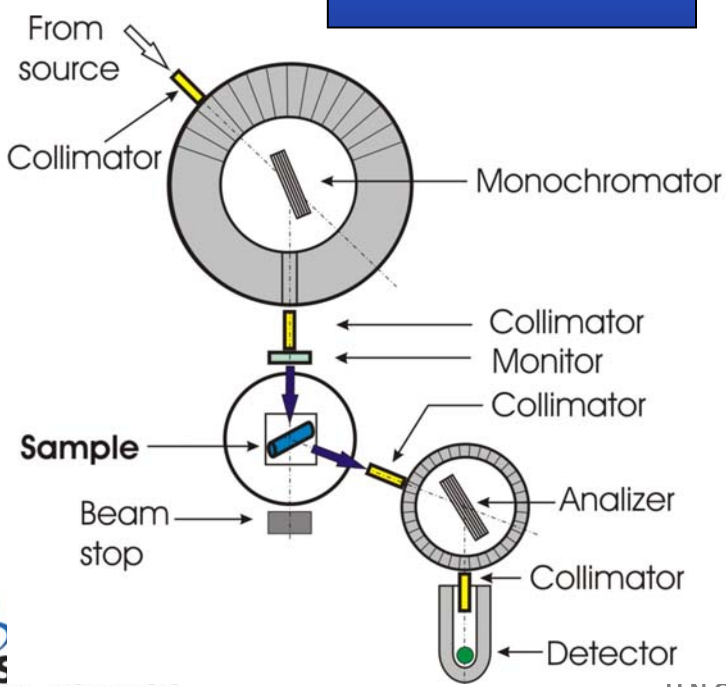
Momentum and energy transfer



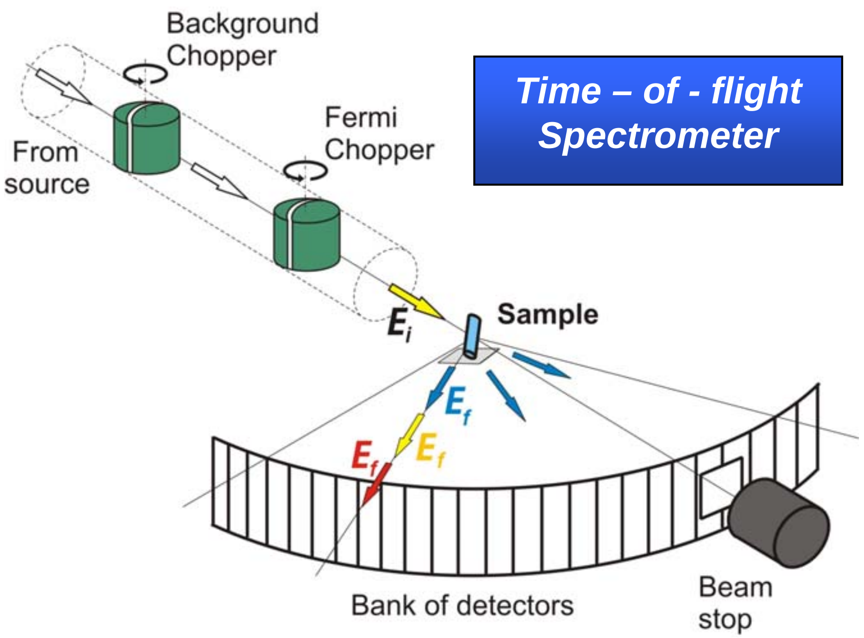
$$\vec{Q} = \vec{k}_i - \vec{k}_f$$

$$E = \frac{\hbar^2}{2m_N}\left(k_f^2 - k_i^2\right)$$

Triple - axis Spectrometer



Time – of - flight Spectrometer



Inelastic Neutron Scattering

Measuring spin dynamics

Scattering cross section σ

$$\frac{d^2\sigma}{d\Omega\, dE} = \frac{k_f}{k_i} \underbrace{S(\vec{Q}, E)}_{\text{Scattering Function}}$$

***Case of** Magnetic Scattering* $S_{mag}(\vec{Q}, E)$

$S_{mag:}$ $\approx$ ***Fourier Transform of the moment-moment correlation function.***

S_{mag} ***Related to the imaginary part of the*** *dynamic susceptibility* χ

$$S_{mag}(\vec{Q}, E) = \frac{1}{\pi}\left[\frac{1}{1 - e^{-E/k_B T}}\right]\chi''(\vec{Q}, E)$$

$$\chi''(Q, E) \propto \underbrace{f^2(Q)}_{\text{Magnetic form factor}} \frac{E\,\chi_{DC}\,\Gamma_Q}{2\pi} \underbrace{\left(\frac{1}{(E - E_{0,Q})^2 + \Gamma_Q^2} + \frac{1}{(E + E_{0,Q})^2 + \Gamma_Q^2}\right)}_{\text{Power-spectrum}}$$

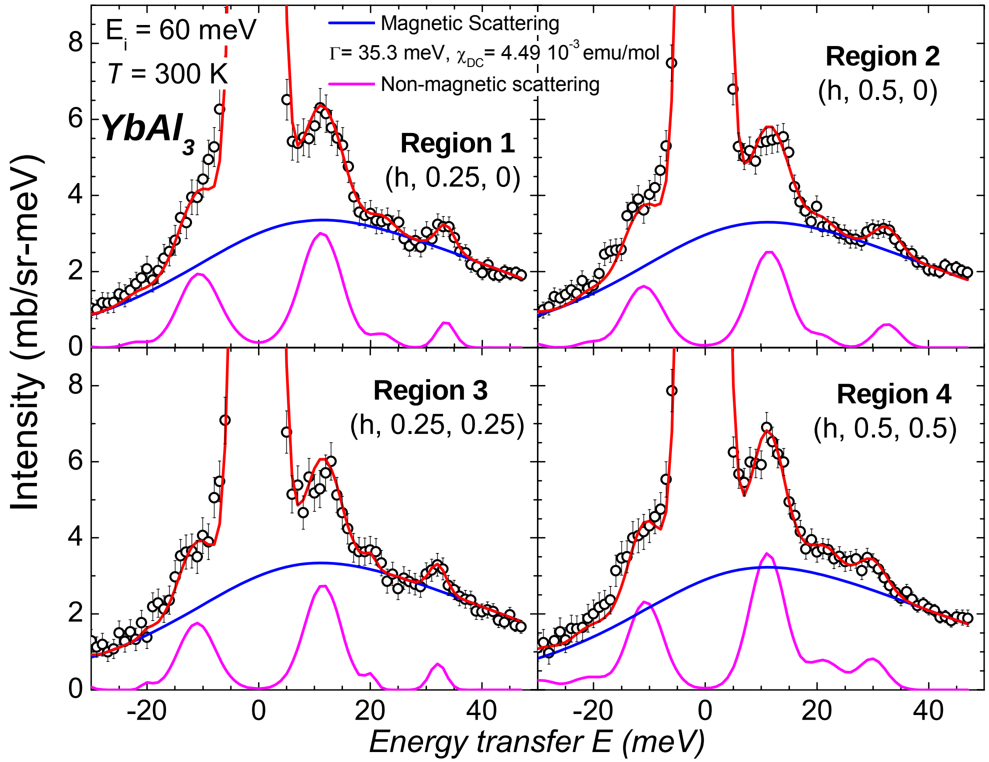
Inelastic Neutron Scattering on $YbAl_3$

Measurements at 300 K, 100 K and 8 K

on time – of – flight spectrometer

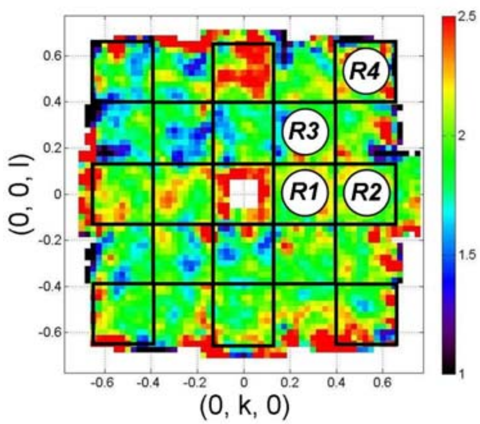
(no non-magnetic analog compound)

$YbAl_3$ Magnetic scattering at 300 K



We have not measured INS on a non-magnetic counterpart compound

Regions in Q- space



Intensity = (Magn. Comp.) + (Non-magn. Comp)

Assuming: (Non-magn) = 3 Gaussian peaks + elastic peak

we obtain: Q-indep, Quasi-elastic Lorentzian Γ = 35 meV

$YbAl_3$ magnetic scattering

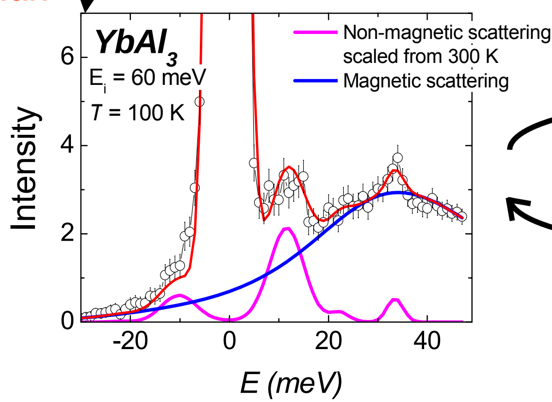
300 K

E_i = 60 meV

E_i = 120 meV

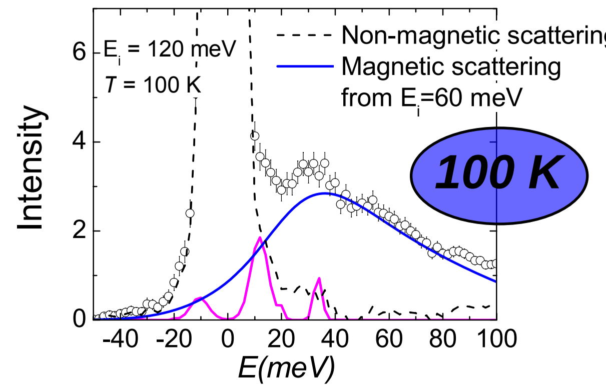
Temp-scale
3-Gaussian

100 K



Check
use Lorentzian

If good agreement



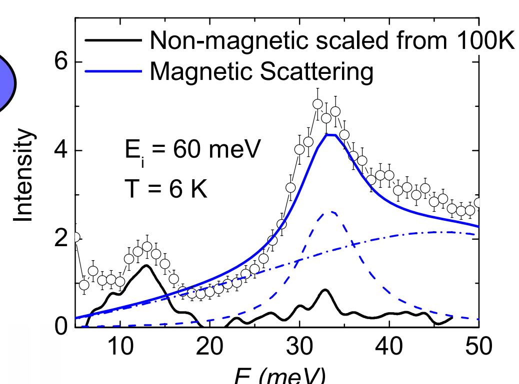
100 K

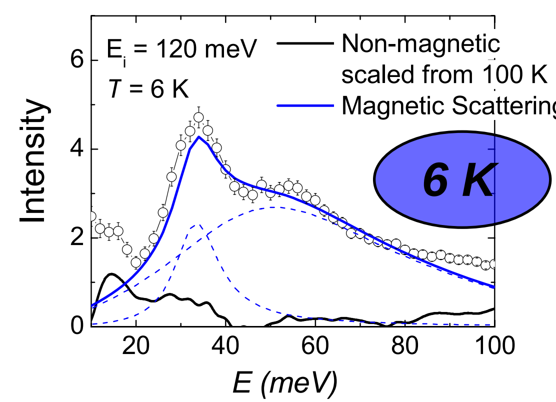
(Magn.) = I – (Non-magn)

I = (Magn) + (Non-magn)

Temp-scale
Non-magnetic

6 K



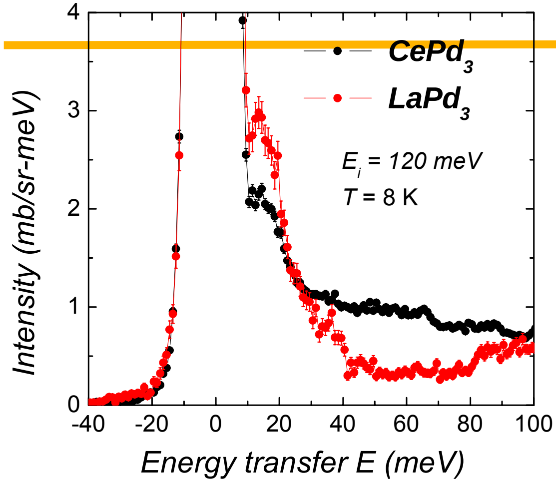


6 K

(Magn.) = I – (Non-magn)

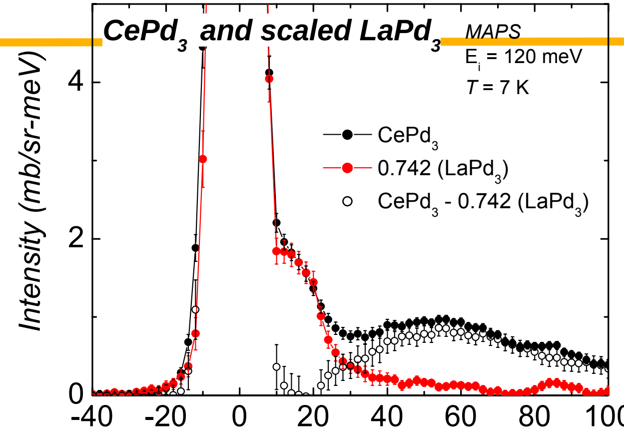
(Magn.) = I – (Non-magn)

Magnetic and non-magnetic Scattering

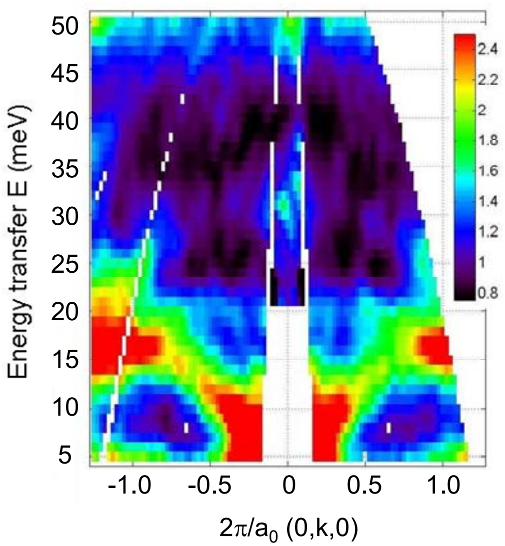


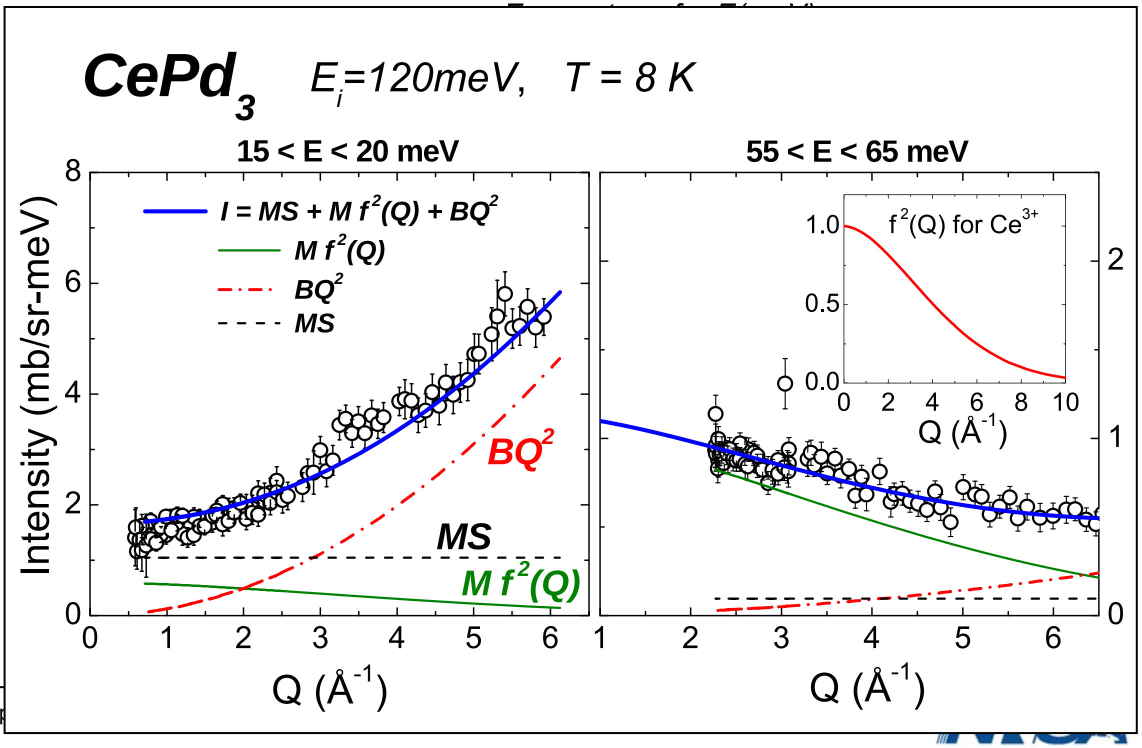
Non-magnetic scattering

Represented as a "scaled $LaPd_3$"



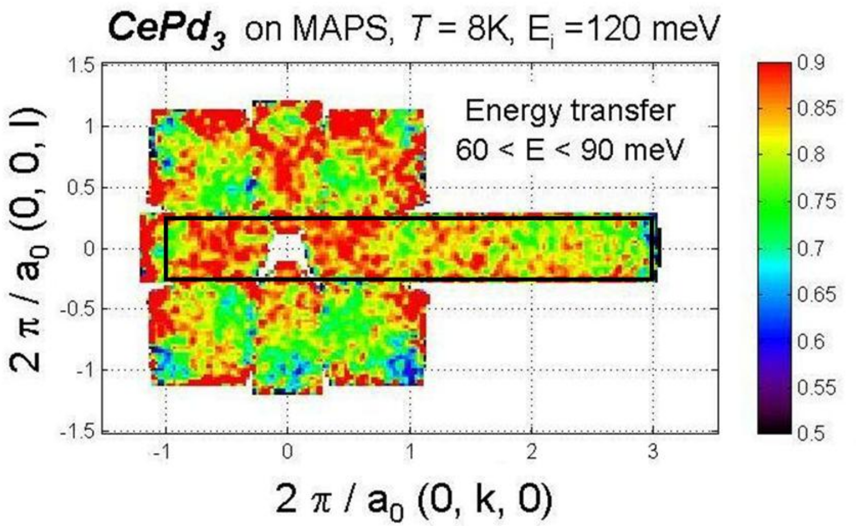
Intensity color map E vs k

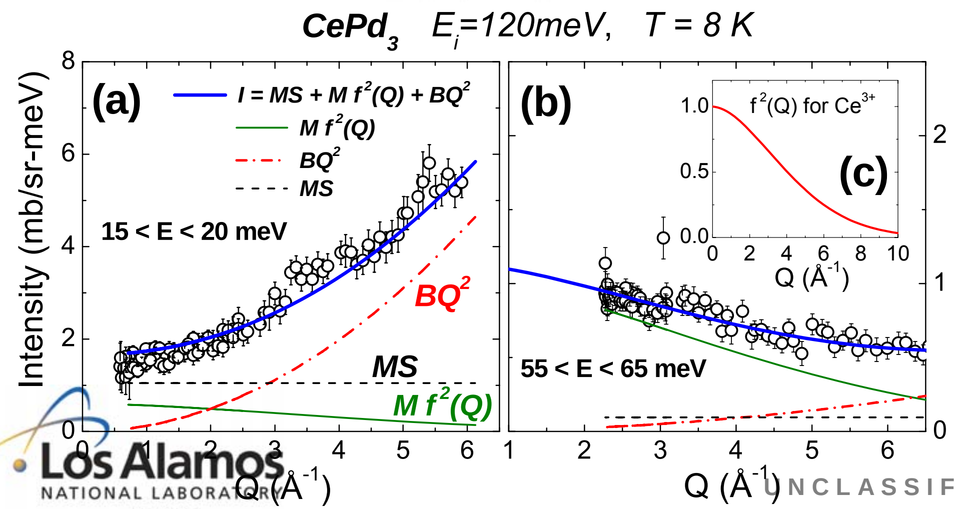




Polycrystalline average (i)

- *Integrate on a large portion of the reciprocal space*





- *Plot intensity I vs.Q for different ranges of energy transfer*
- *Analyze Intensity as composed by 3 contributions for the scattering:*

$$I = \underbrace{MS}_{\text{Multiple scattering}} + \underbrace{BQ^2}_{\text{Single phonon scattering}} + \underbrace{M f^2(Q)}_{\text{Magnetic scattering}}$$

$f^2(Q)$: magnetic form factor for Ce 4f orbital

***Assumption**:*

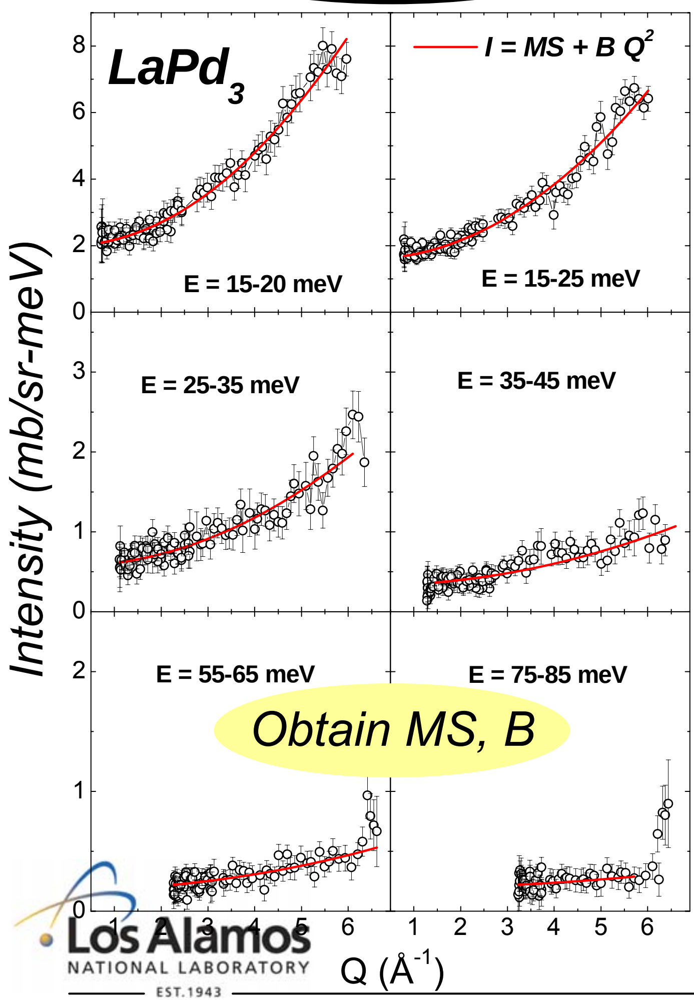
MS and M are Q-independent

Polycrystalline average (ii)

Non-magnetic scattering *from* $LaPd_3$
$MS+BQ^2$

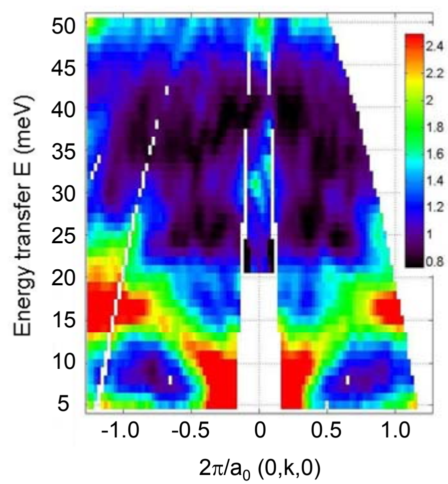
scaling $MS(LaPd_3)$, $B(LaPd_3)$

Magnetic scattering
$I=MS + BQ^2 + M f^2(Q)$



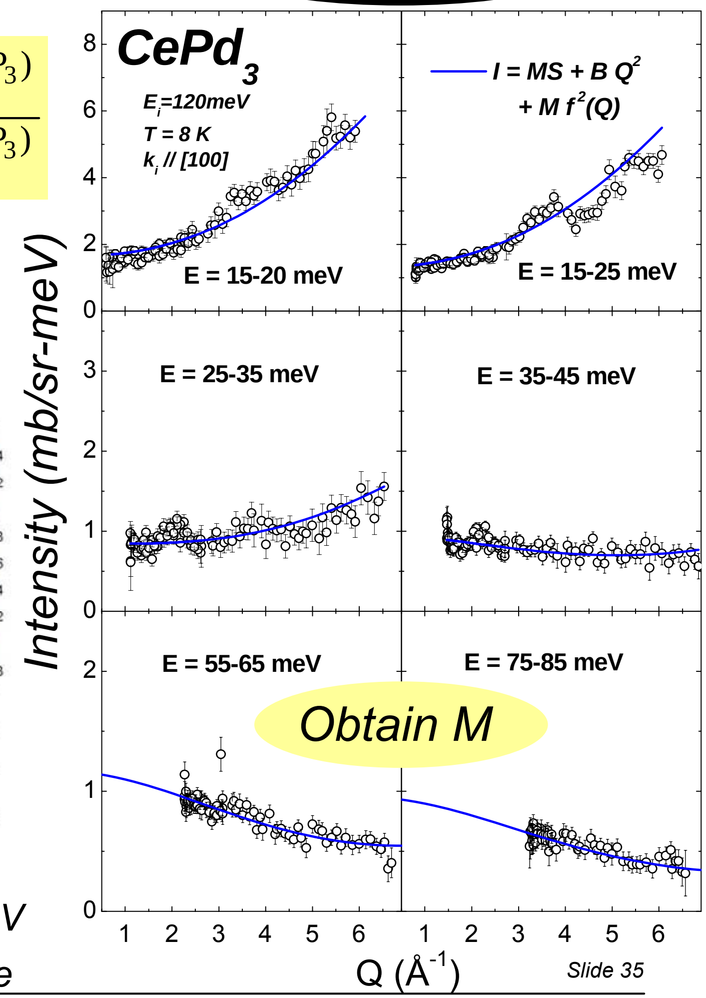
$$\text{scale factor} = \frac{\sigma_{\text{coh}}^{(\text{CeP}_3)}}{\sigma_{\text{coh}}^{(\text{LaP}_3)}}$$

ratio of ***coherent scattering*** *cross sections*



BQ² *dominant < 30 meV*

Mf²(Q) *dominant above phonon cutoff*



Polycrystalline average (iii)

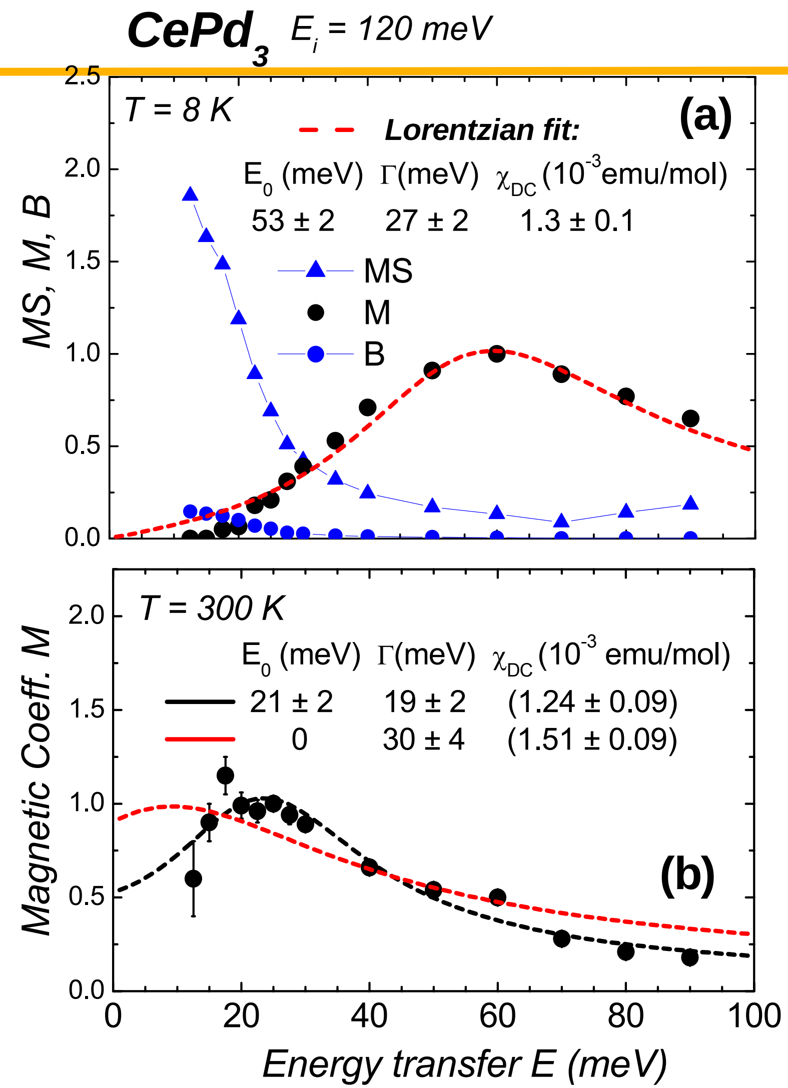
T = 8 K: Inelastic Kondo-like magnetic scattering:

maximum between 50 and 60 meV, (same scale of T_K~(500-600) K.

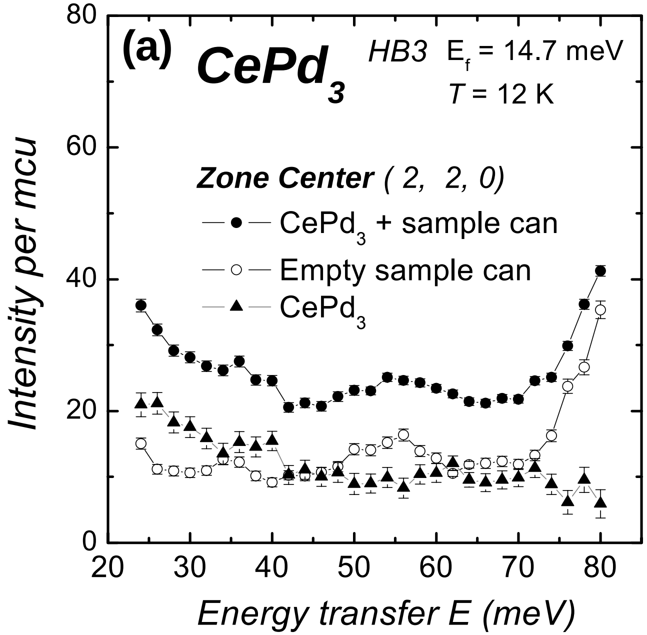
As temperature increases, it evolves towards a quasielastic Lorentzian

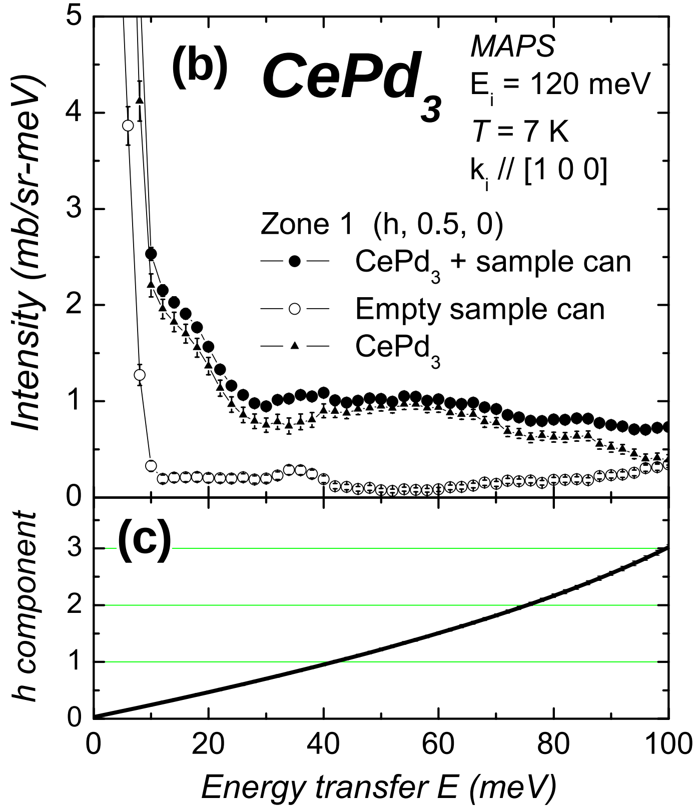
Scattering cross sections

Cross sections	σ_{coh} (barns)	σ_{inc} (barns)	σ^{total}_{scatt} (barns)	σ_{abs} (barns)	b_{coh} (fm)
La	8.53	1.13	9.66	8.94	8.24
Ce	2.94	0	2.94	0.63	4.84
Pd	4.39	0.093	4.48	6.9	5.91
$LaPd_3$	21.70	1.409	23.10	29.64	25.97
$CePd_3$	16.11	0.279	16.38	21.33	22.57
Ratio ($CePd_3$/$LaPd_3$)	0.742	0.198	0.709	0.720	0.869



Comparison Triple-axis vs Time-of-flight spectrometers



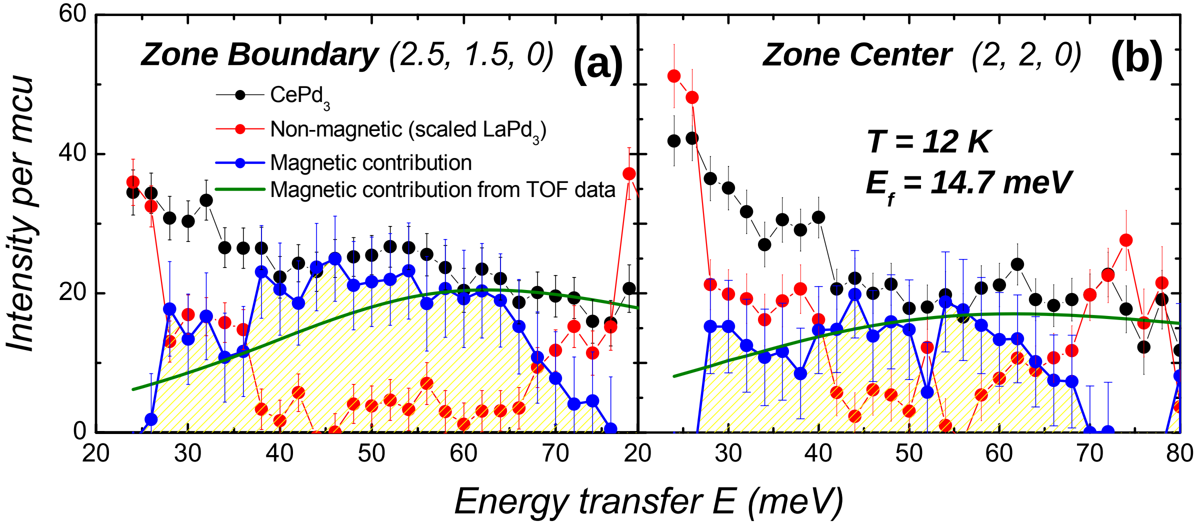


9 hs. of beam

12 hs. of beam

Measuring on Triple-axis spectrometer

HB3 Triple axis spectrometer *T = 12 K, E_f=14.7 meV*



TAS has **higher background**, and lesser statistics:

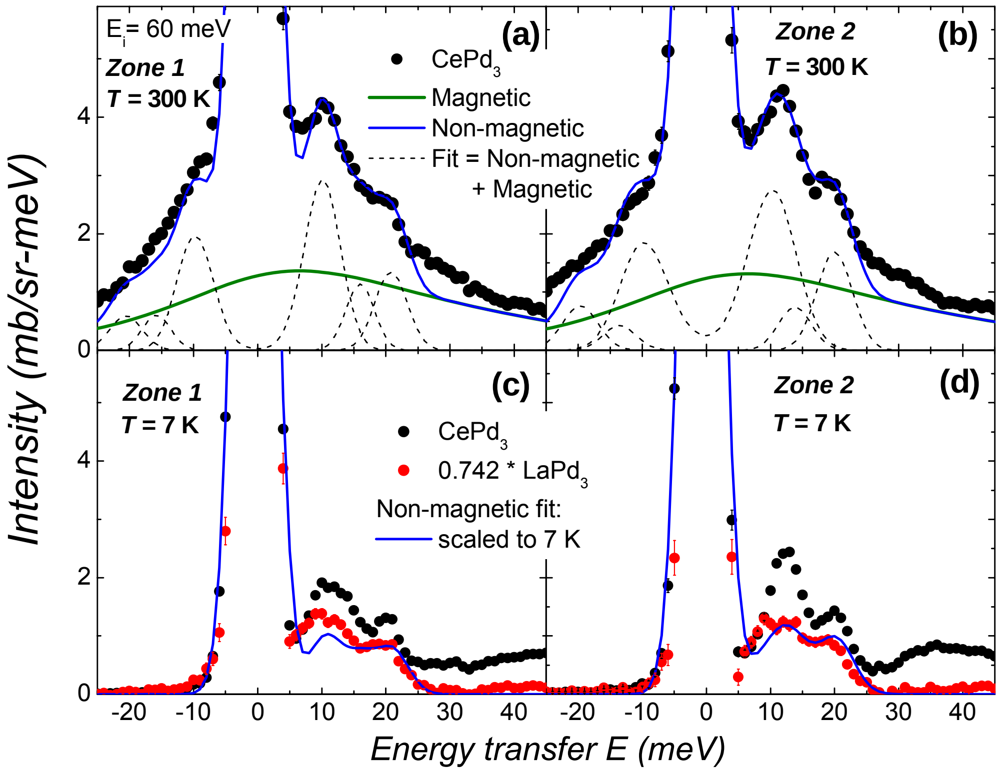
- Maxwell **profile of the reactor** (~ 70 meV)
- Larger amount of **aluminum "in the beam"**

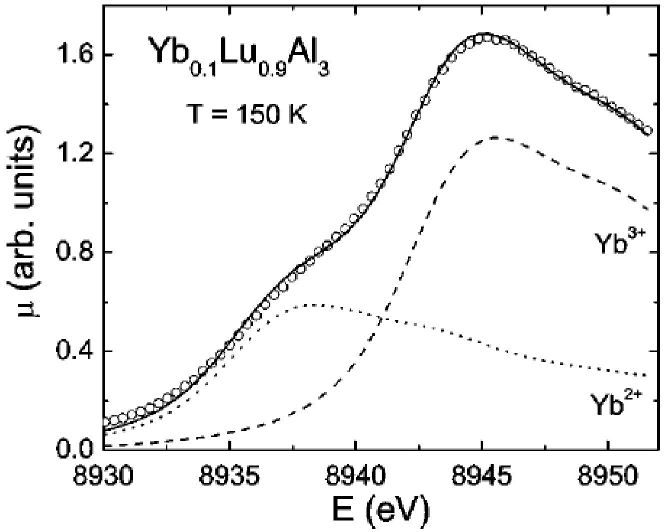
Constant Q- scans:

(a) Energy-scan at the Brillouin zone boundary , and (b) at the Brillouin zone center

The magnetic intensity from TOF measurements (green line) is also included for comparison.

Alternative Method to Obtain the Non-magnetic Scattering





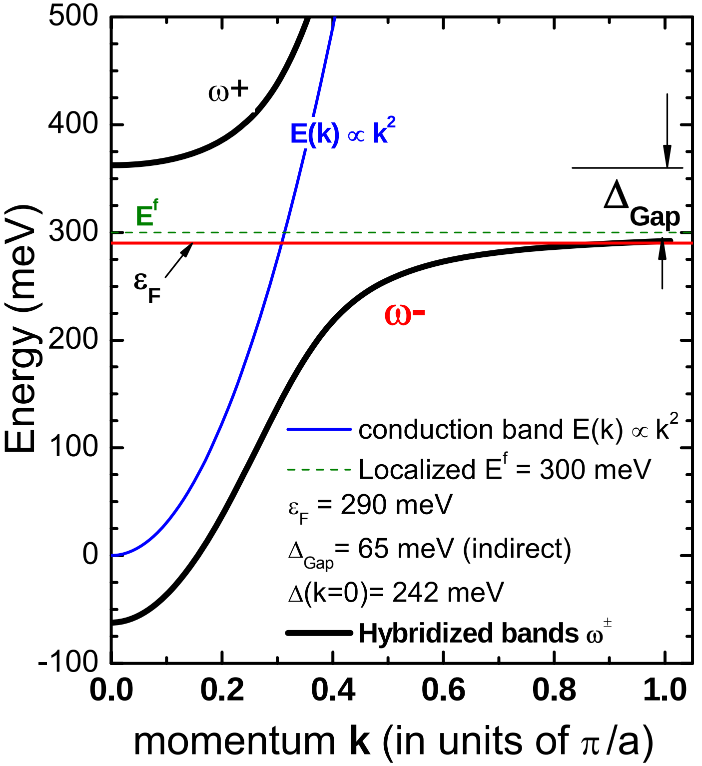
Bauer, *et al*., Phys. Rev. B **69**, 125102 (2004)

L_{III} X-ray absorption spectrum

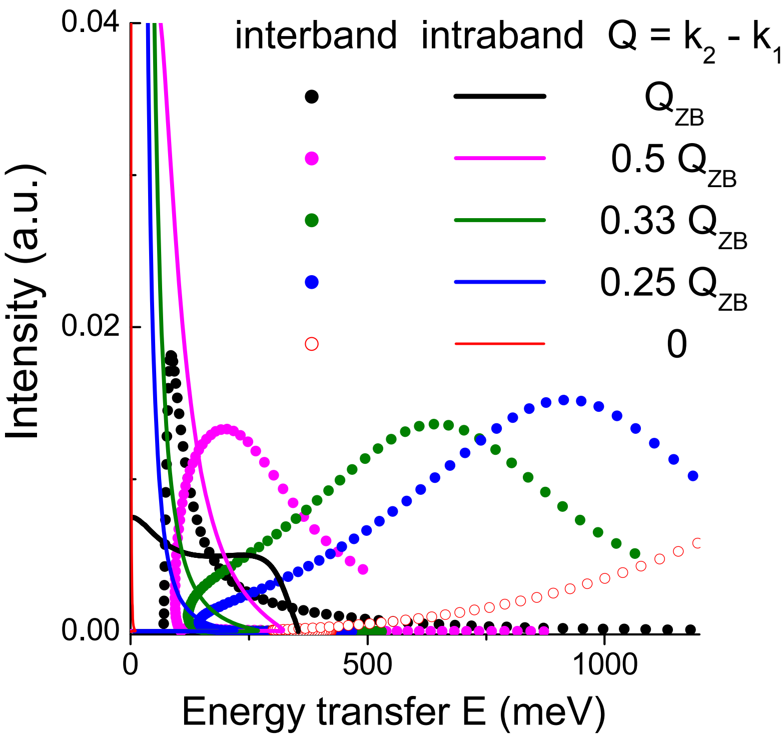
Spectra correspondent to divalent and trivalent Yb absorptions

Estimation for the spectrum of particle-hole excitations for a basic scheme of hybridized bands

$$S(\vec{Q},E) \propto \int dE' f(E')\left(1-f(E')\right) D(E',E'+E;\vec{Q})$$



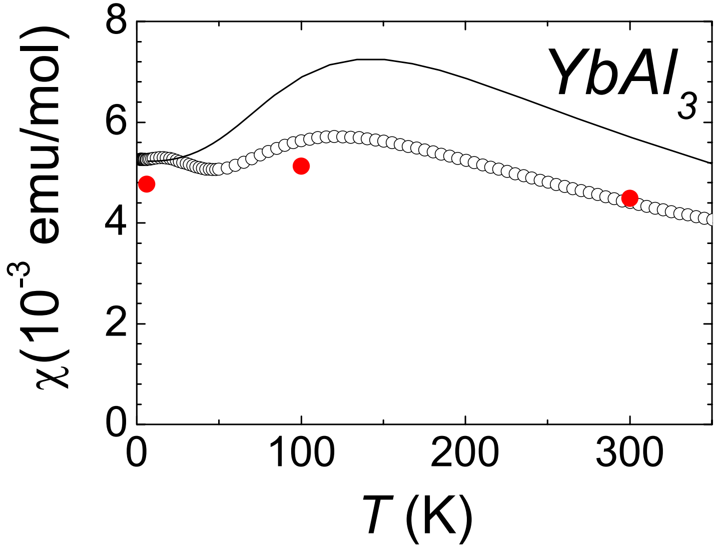
Hybridized bands ω+ and ω-

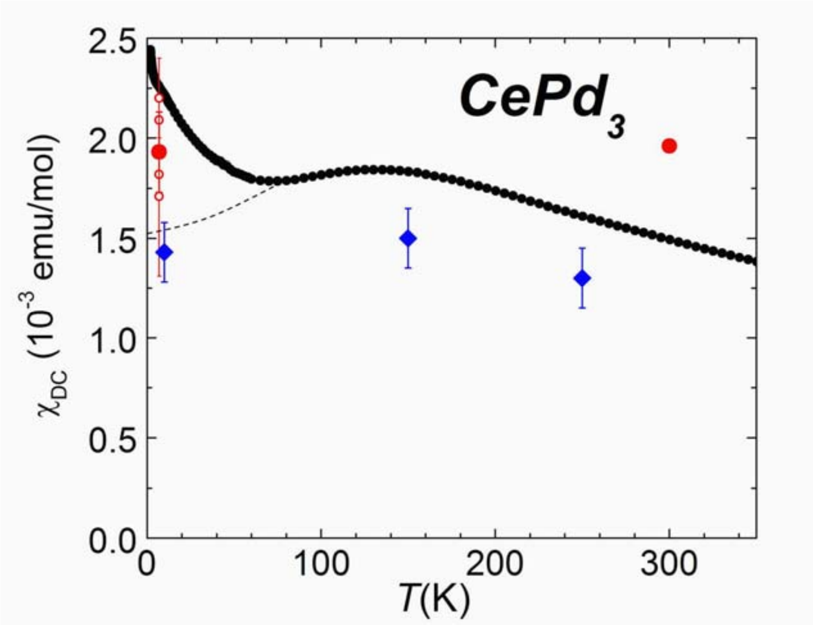


Spectra for intraband (lines) and interband (circles) excitations for momentum transfer in the range between 0 and $Q=Q_{ZB}$.

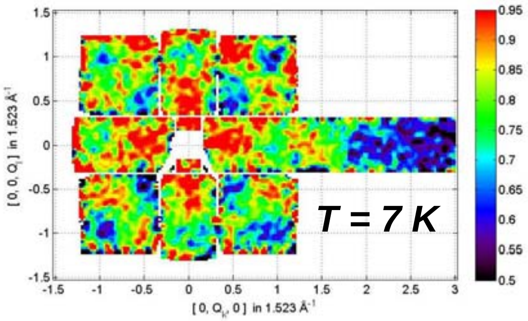
Magnetic Susceptibility

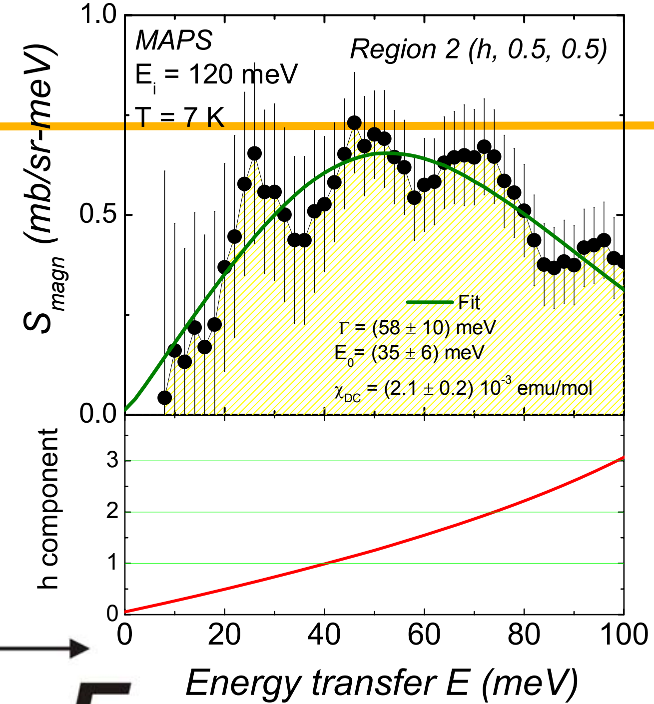
Bulk measurements and χ obtained from INS





Oscillation of Magnetic Intensity





Variations of order 20%