

Wir schaffen Wissen – heute für morgen

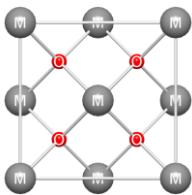
Paul Scherrer Institut

Romain Sibille (Laboratory for Developments and Methods)

**Spin Liquids in Novel Pyrochlore Materials
Investigated using Large Facilities**

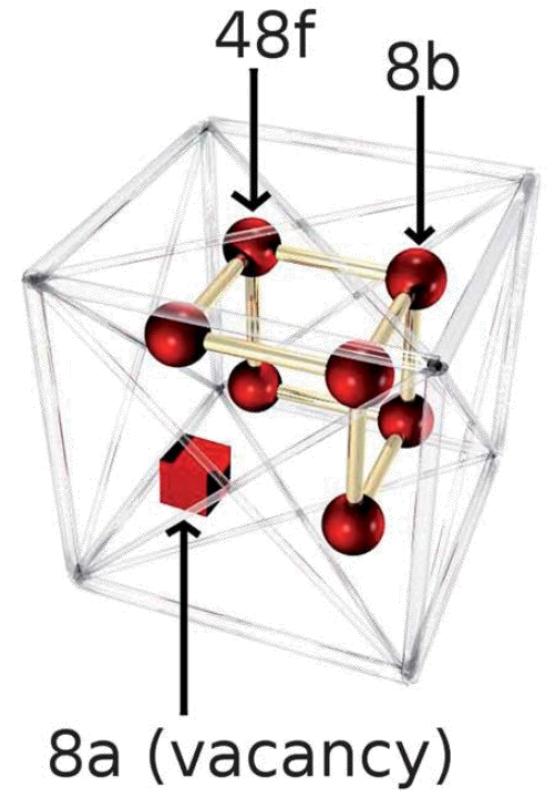
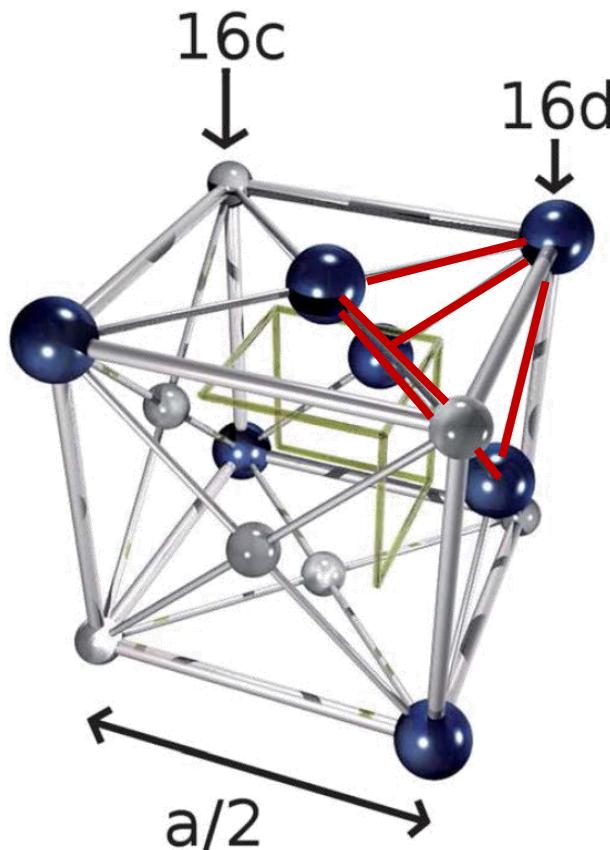
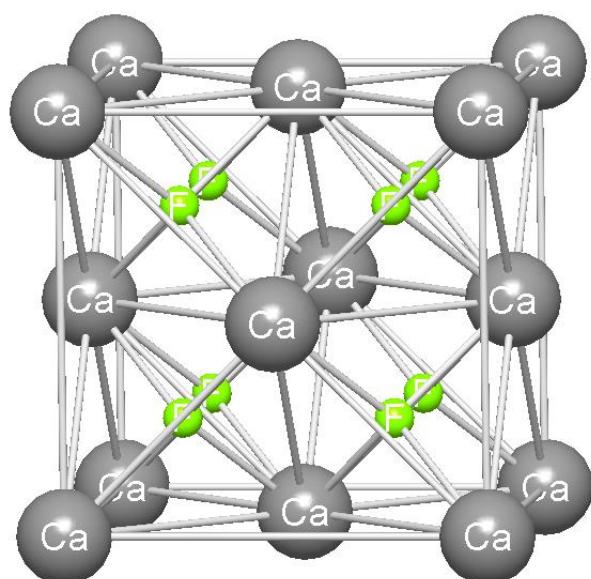
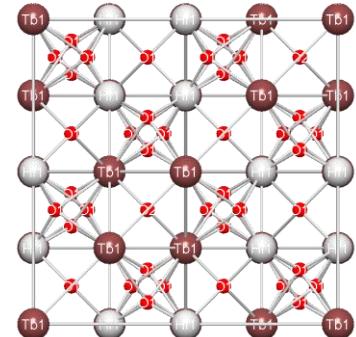
Introduction

CaF_2 (fluorite)
Fm-3m



$\times 8$

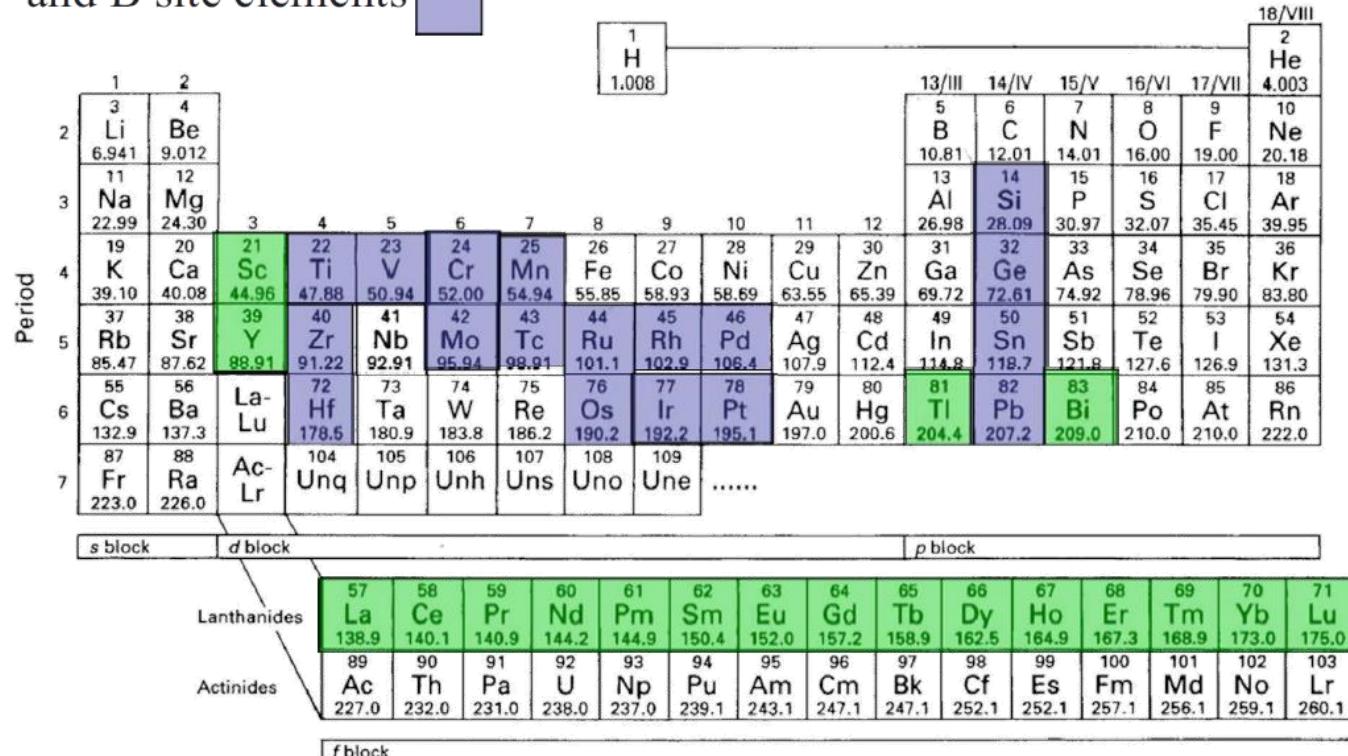
$\text{Ca}_2\text{Nb}_2\text{O}_7$ (pyrochlore)
Fd-3m



Introduction

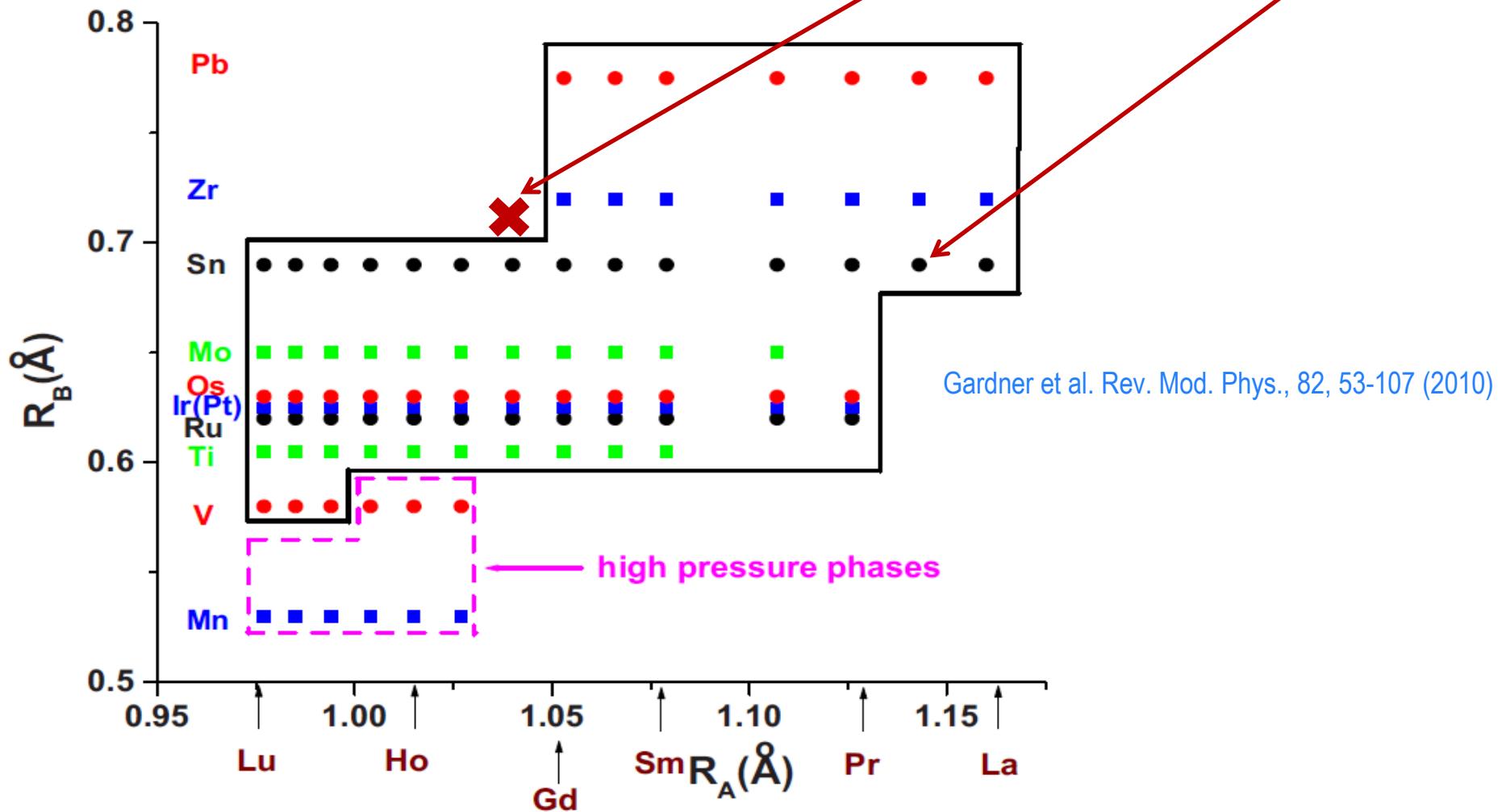


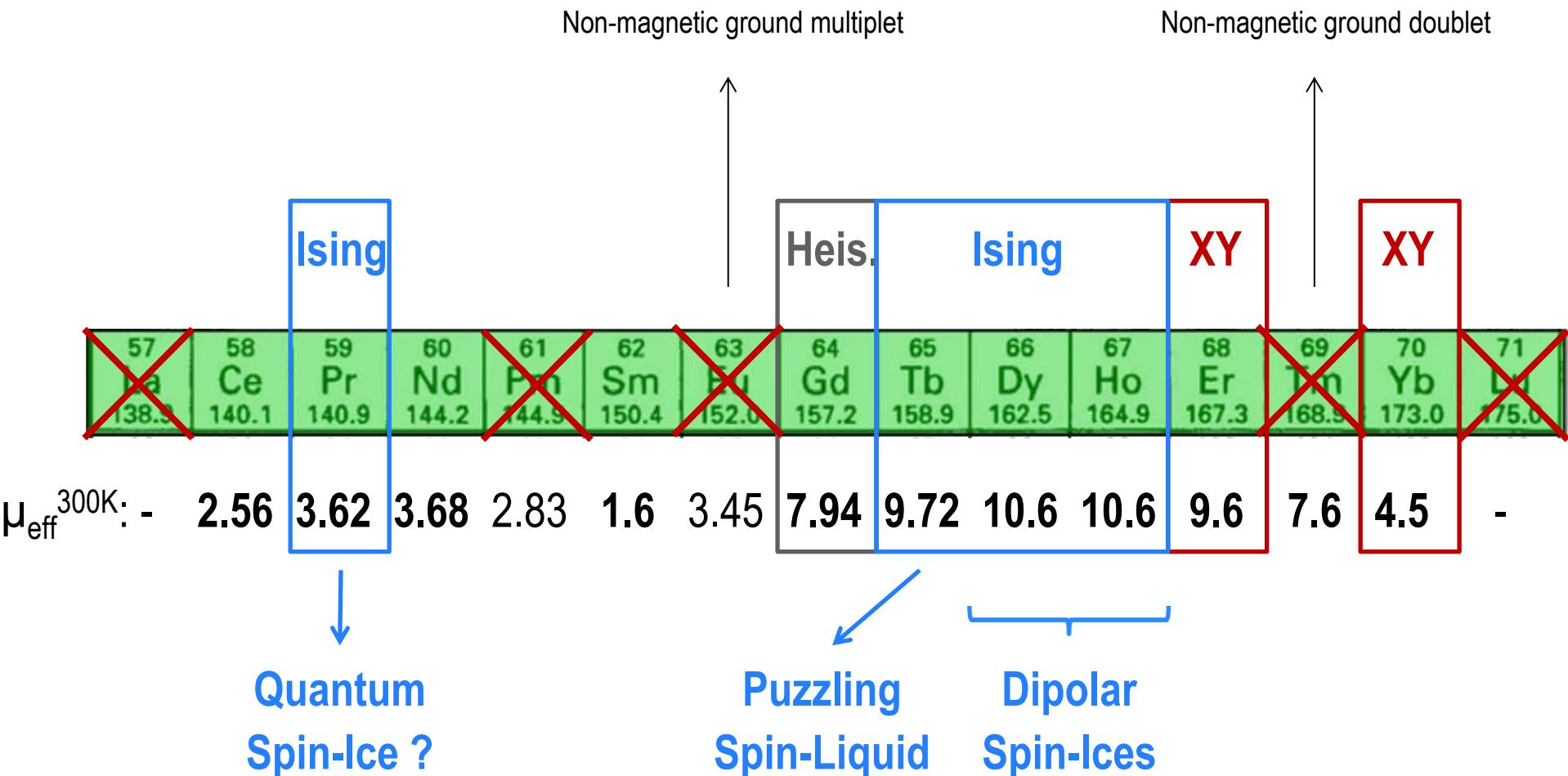
Possible A-site elements and B site elements



Gardner et al. Rev. Mod. Phys., 82, 53-107 (2010)

The insulating pyrochlore magnets $\text{Tb}_2\text{Hf}_2\text{O}_7$ and $\text{Ce}_2\text{Sn}_2\text{O}_7$





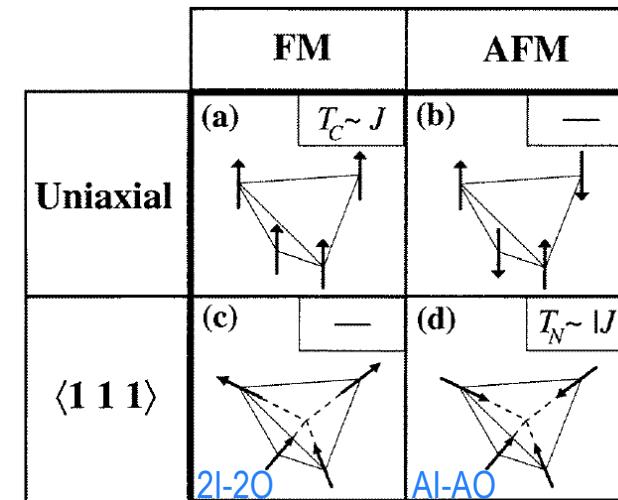
Ising pyrochlore magnets

Only two-states uniaxial spin symmetry compatible with the crystal lattice

= four local $\langle 111 \rangle$ anisotropy axes pointing in or out of a tetrahedron

→ reverses the role of ferro- / antiferro-magnetic exchanges with regard to frustration:

Bramwell et al. J. Phys. Cond. Mat., 10, L215-220 (1998)



Interests: FM → spin ices ($\text{Ho}_2\text{Ti}_2\text{O}_7$, $\text{Dy}_2\text{Ti}_2\text{O}_7$, ...)

AFM → puzzling cases complicated by other effects (e.g. $\text{Tb}_2\text{Ti}_2\text{O}_7$)

$\text{Tb}_2\text{Hf}_2\text{O}_7$

A rapid tour of $\text{Tb}_2\text{Ti}_2\text{O}_7$

$\theta_p \approx -13 \text{ K}$ (no order down to 0.4 K)

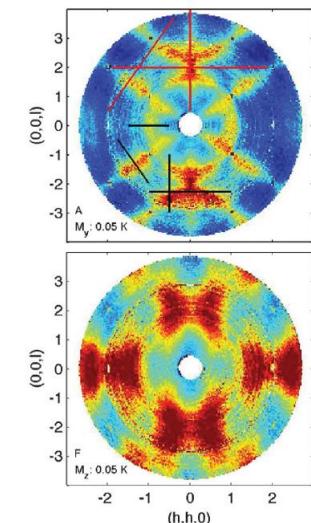
?

H. A. Craig, Explorations, page 23 (2009)

Ising antiferromagnet: $\theta_p \approx -19 \text{ K}$

Gardner et al. Phys. Rev. Lett., 82, 1012-1015 (1999)

Spin liquid state (cooperative paramagnet) below $\approx 20 \text{ K}$



→ power-law spin correlations at very low temperature

Fennell et al. Phys. Rev. Lett., 109, 017201 (2012)

→ magnetic order induced by pressure, magnetic field, $\text{Tb}_{2-x}\text{Ti}_{2+x}\text{O}_7$

Mirebeau et al. Nature, 112, 017203 (2014)

Taniguchi et al. Phys. Rev. B, 87,

Mirebeau et al. Phys. Rev. Lett., 93, 187204 (2004)

060408(R) (2013)

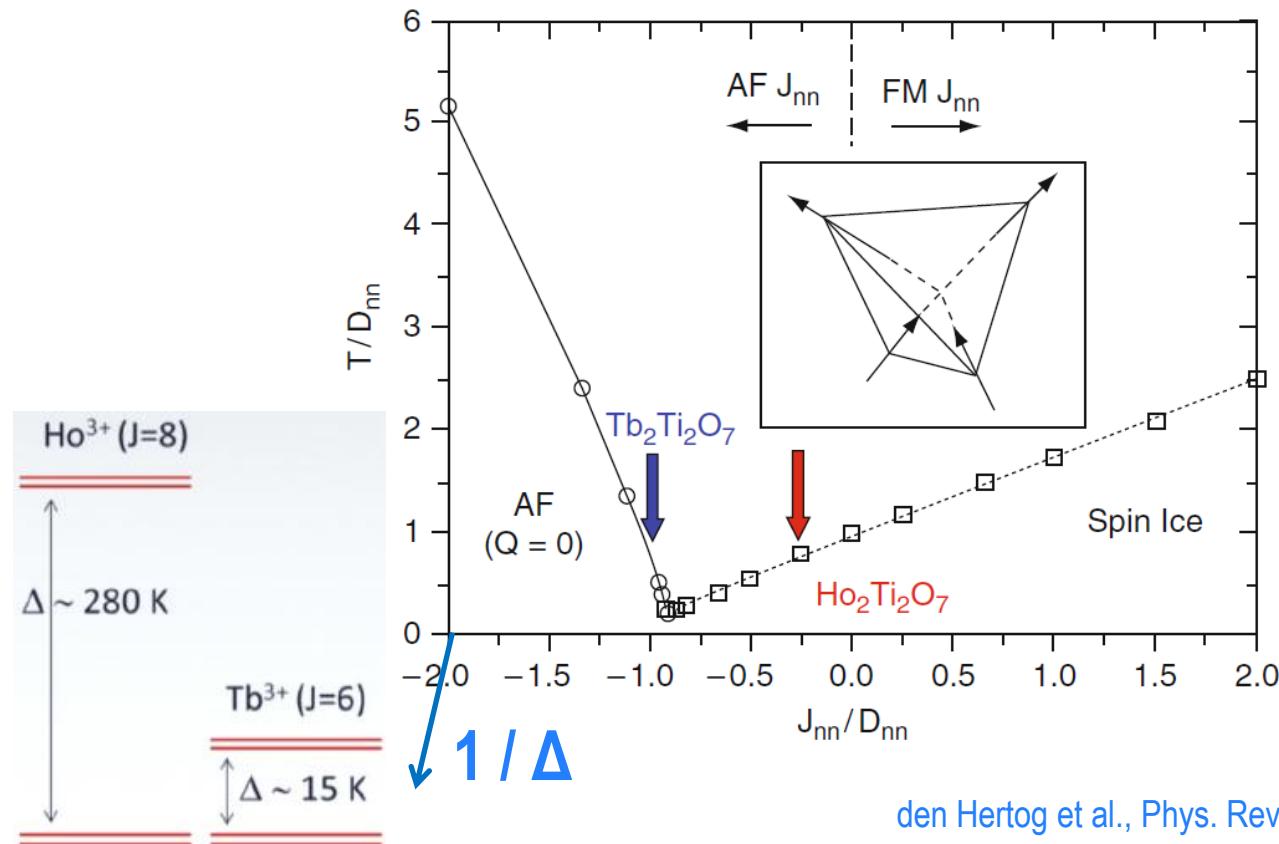
→ magneto-elastic excitations, anisotropic propagating excitations

Fennell et al. Phys. Rev. Lett., 112, 017203 (2014)

Guittet et al. Phys. Rev. Lett., 111, 087201 (2013)

A rapid tour of $Tb_2Ti_2O_7$

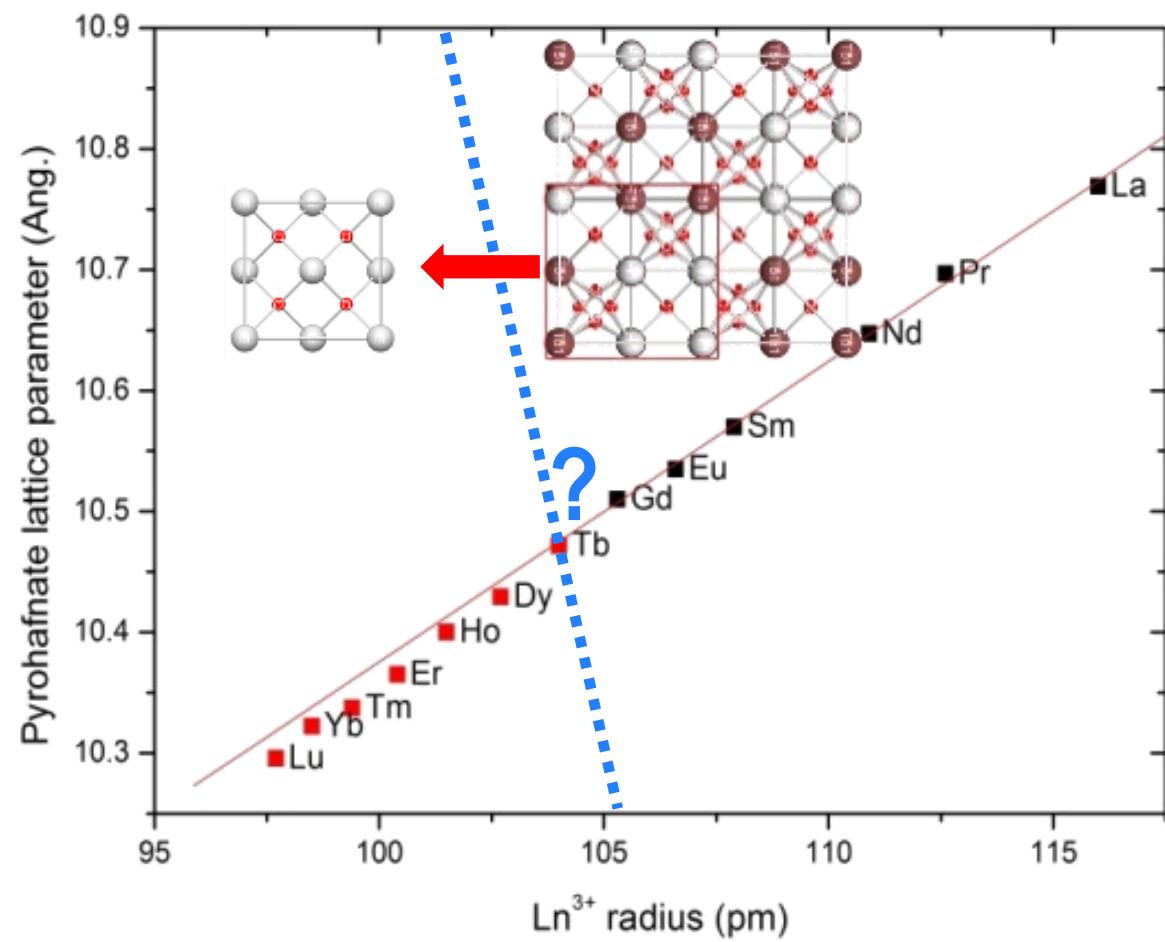
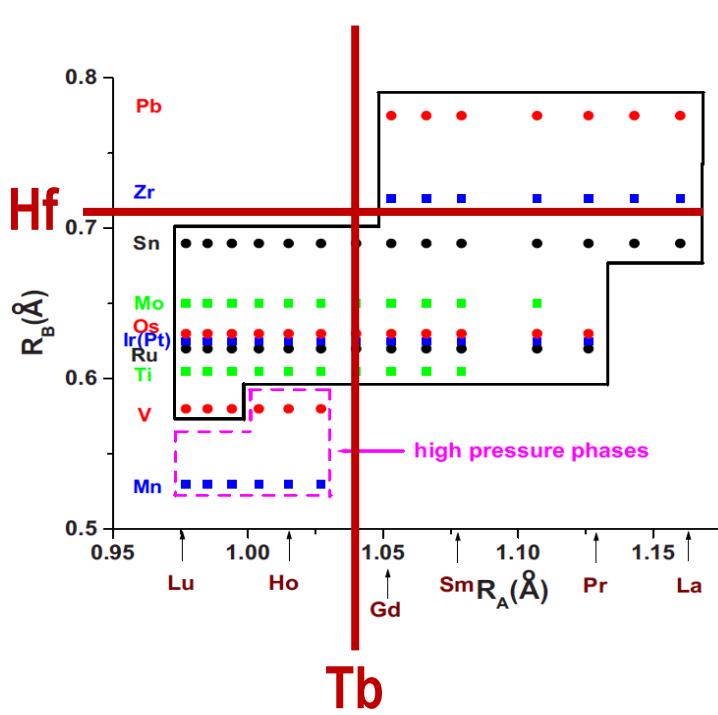
The ‘quantum spin-ice scenario’



Molavian et al. Phys. Rev. Lett., 98, 157204 (2007)

Hafnium pyrochlores

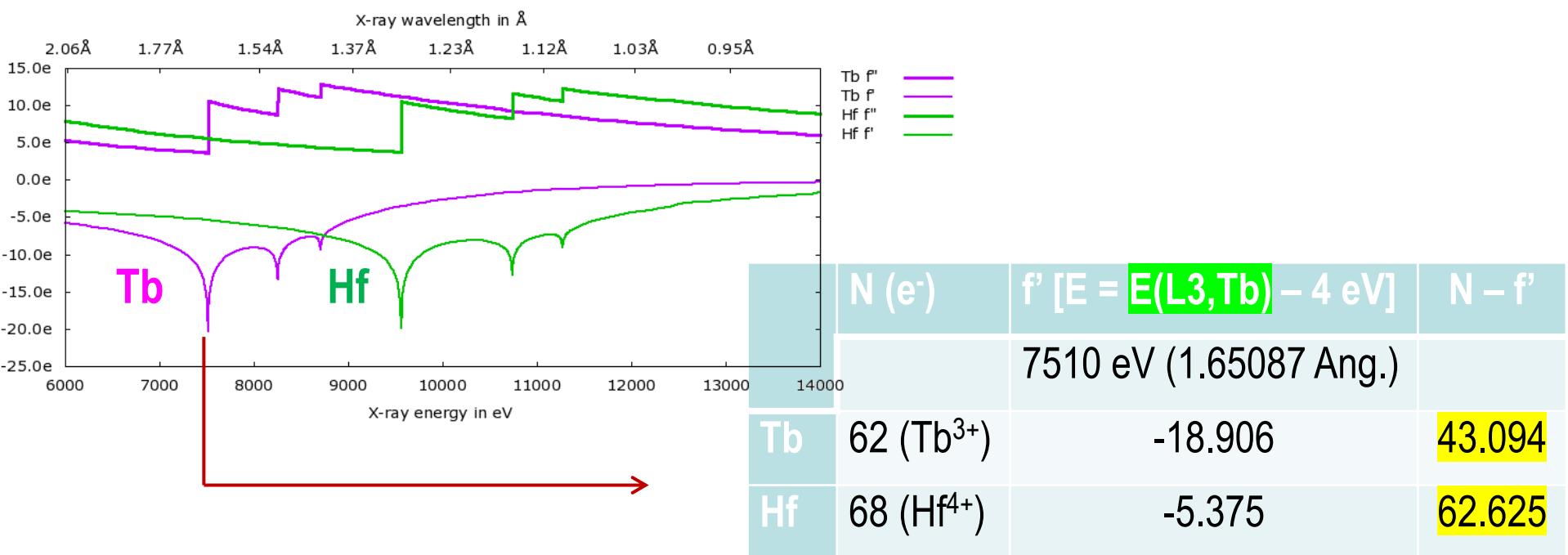
Place of $Tb_2Hf_2O_7$



Crystal structure

Low Tb / Hf contrast for X-rays Tb ($Z = 65$) vs. Hf ($Z = 72$)
 neutrons $b_{Tb} = 7.38$ fm vs. $b_{Hf} = 7.7$ fm

→ Resonant Contrast X-ray Diffraction: $f = f^0 + f' + i f''$



Crystal structure

joint refinement:

Resonant Contrast Diffraction

(MSX04SA at SLS)

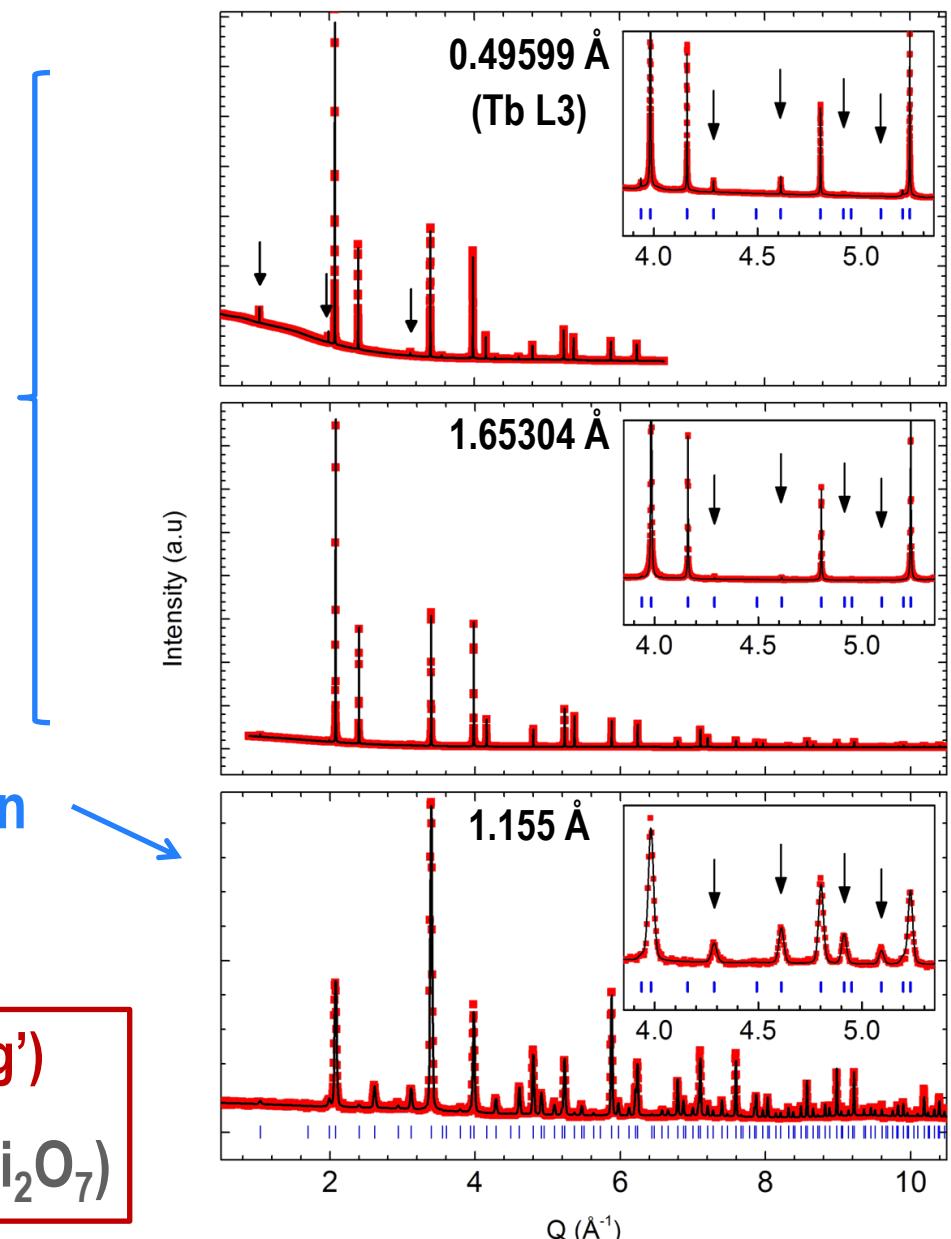
+

High Resolution Neutron Diffraction

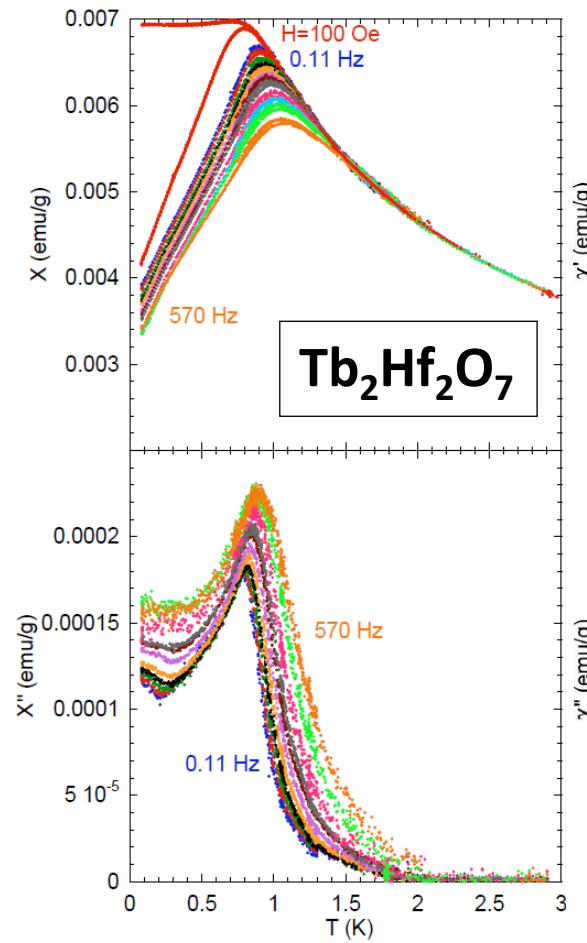
(HRPT at SINQ)

→ No cation antisite disorder ('stuffing')

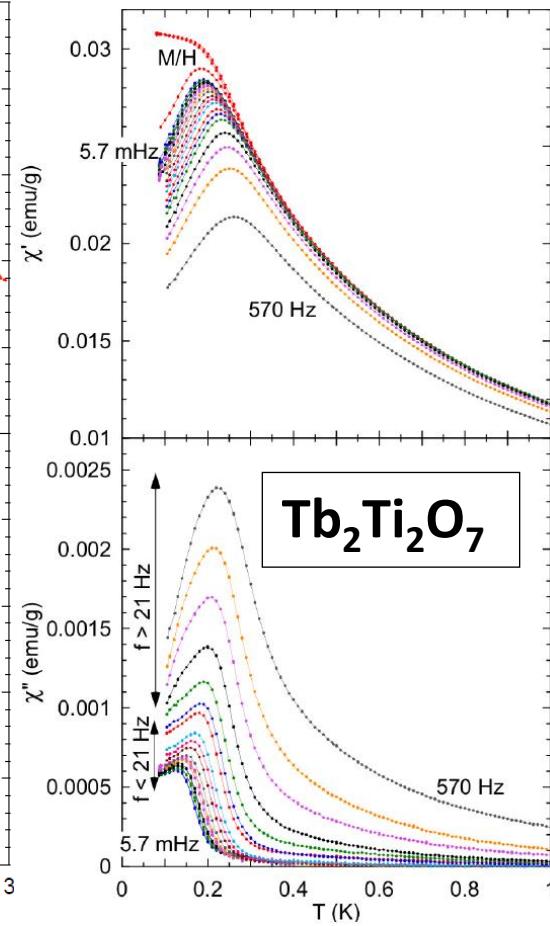
with an upper limit of 2.5 % ($\approx \text{Tb}_2\text{Ti}_2\text{O}_7$)



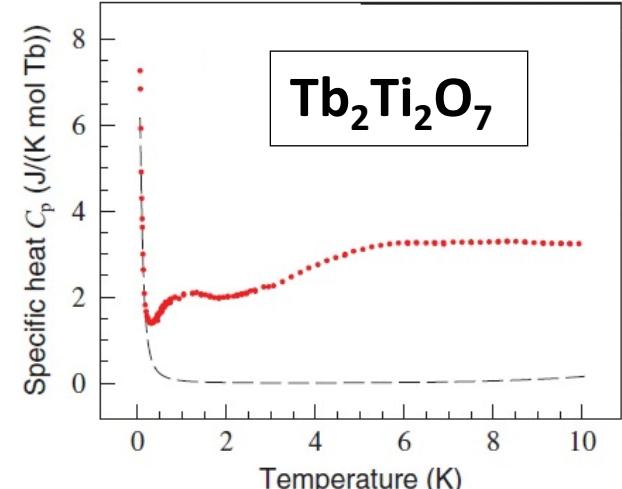
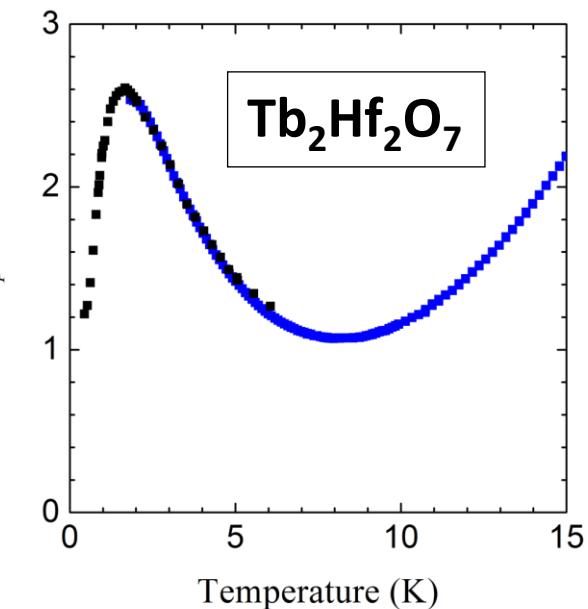
Macroscopic behavior



Lhotel et al. Phys. Rev. B, 86, 020410(R) (2012)

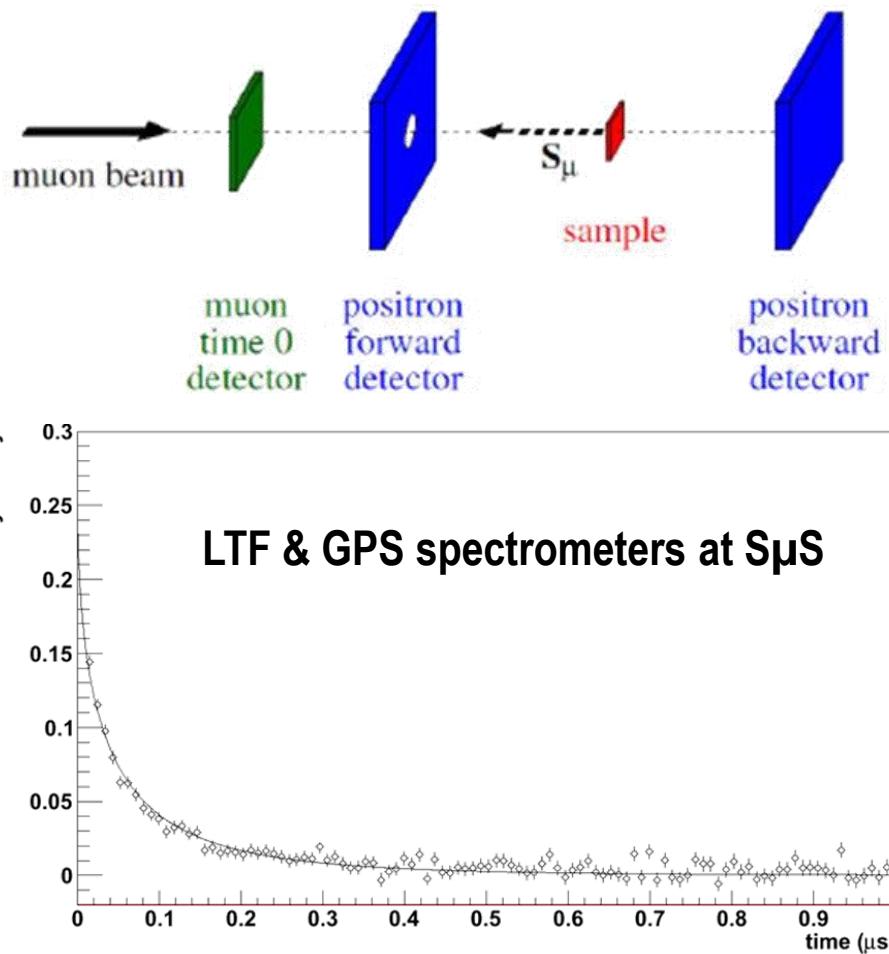


Yaouanc et al. Phys. Rev. B, 84, 184403 (2011)

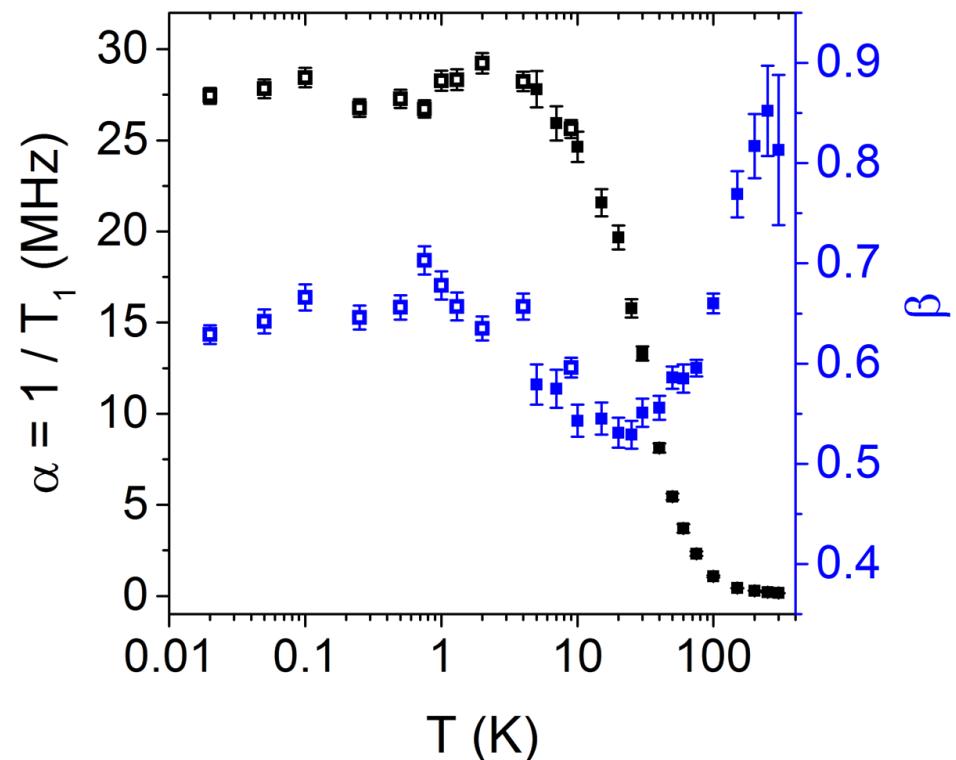


Muon spin relaxation

dynamic system down to 0.02 K, temperature dependence of fluctuations:

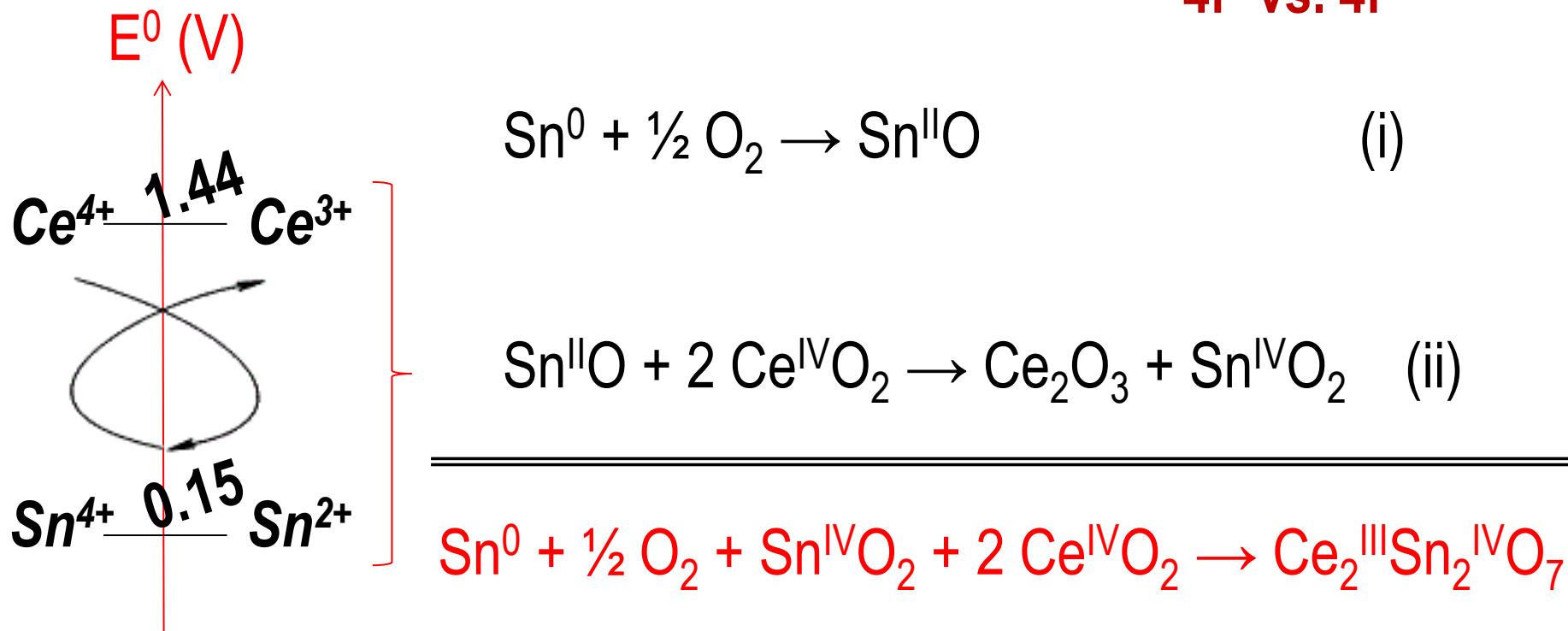


$$A(t) = A_0 \exp[-(t/T_1)^\beta] + A_{bg}$$



Ce³⁺ pyrochlores

Preparation inherently difficult (stabilization of Ce³⁺ vs. Ce⁴⁺)

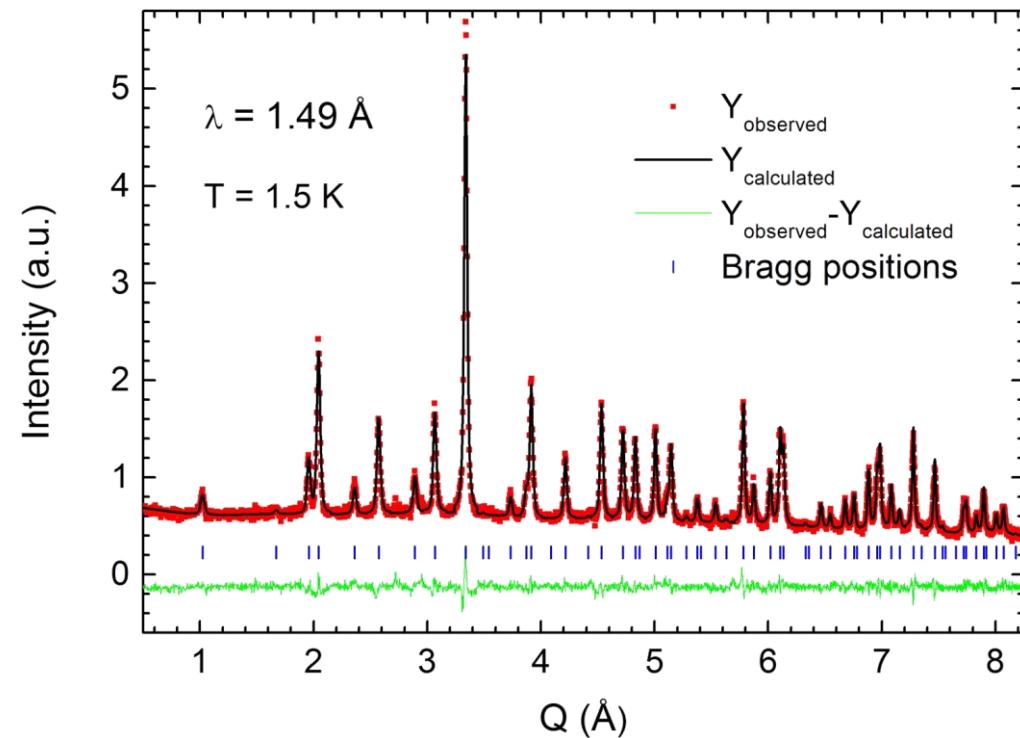
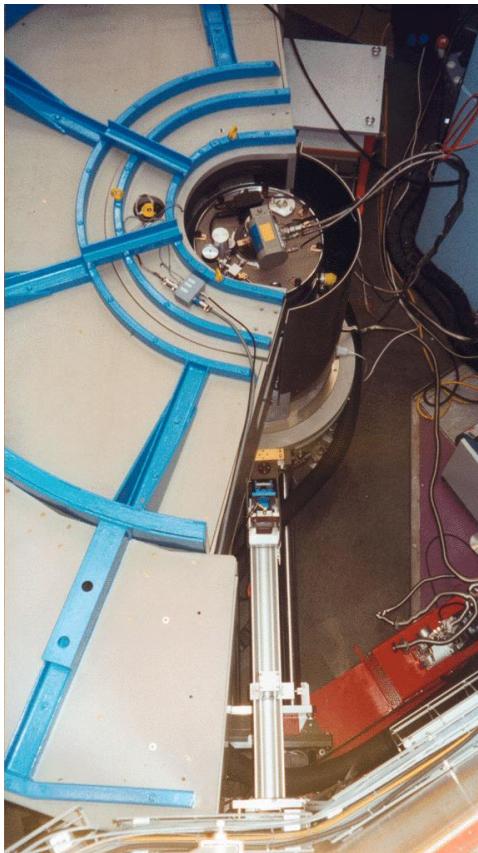


Tolla et al. C. R. Acad. Sci. Paris, IIc, 139-146 (1999)

Crystal structure

Well ordered pyrochlore ($b_{\text{Sn}} = 6.23 \text{ fm}$, $b_{\text{Ce}} = 4.84 \text{ fm}$, $b_0 = 5.81 \text{ fm}$)

HRPT at SINQ

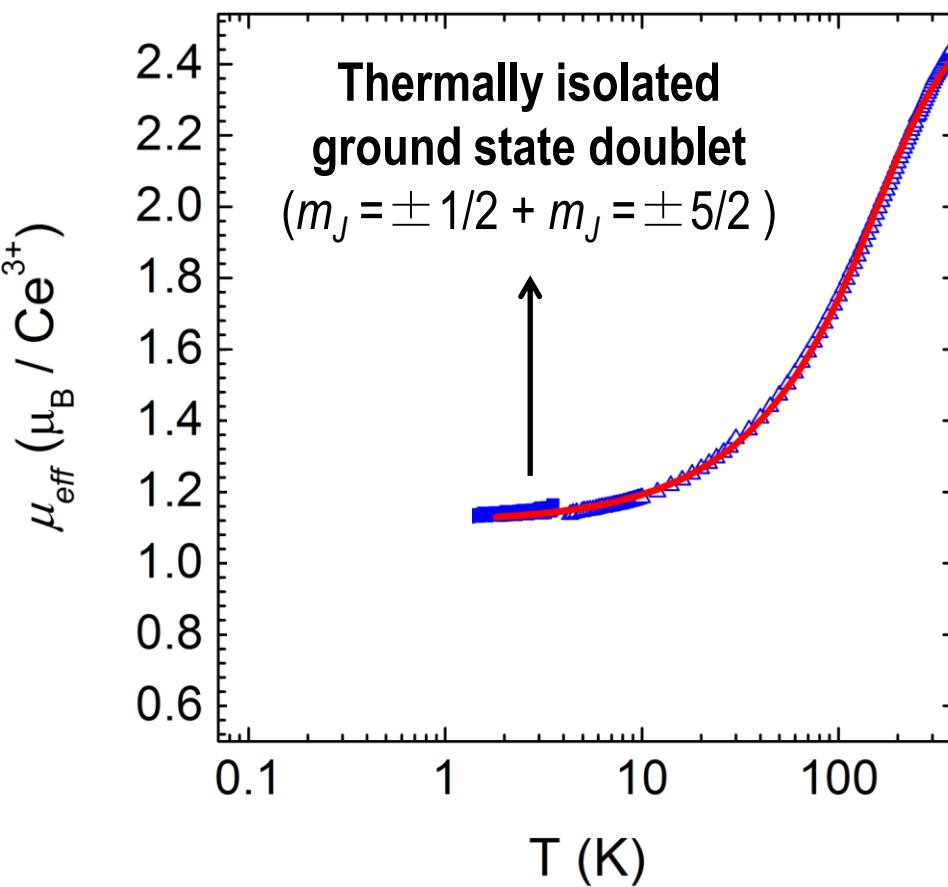


→ No evidence for structural defects
 (antisite $0.5 \pm 2.5 \%$, Frenkel $0.36 \pm 0.16 \%$)

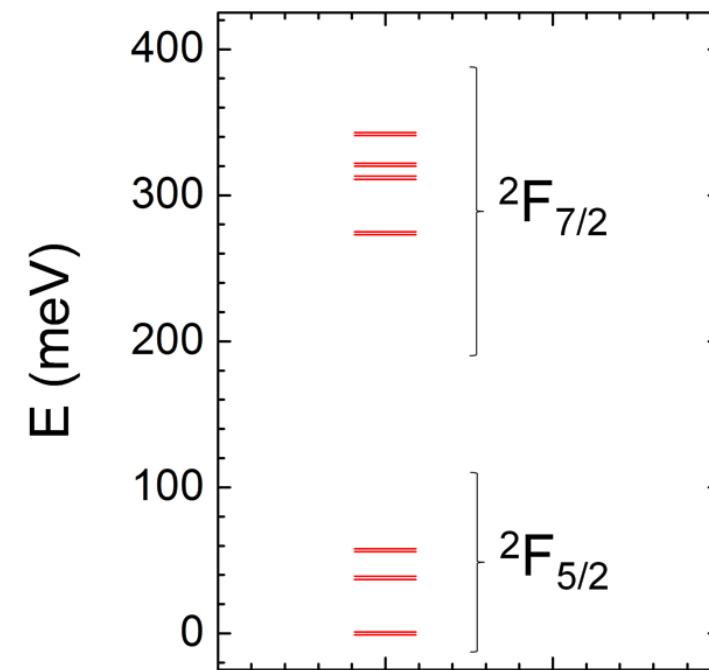
Sibille et al., arXiv 1502.00662v1 (2015)

High temperature magnetic susceptibility

→ crystal-field effects



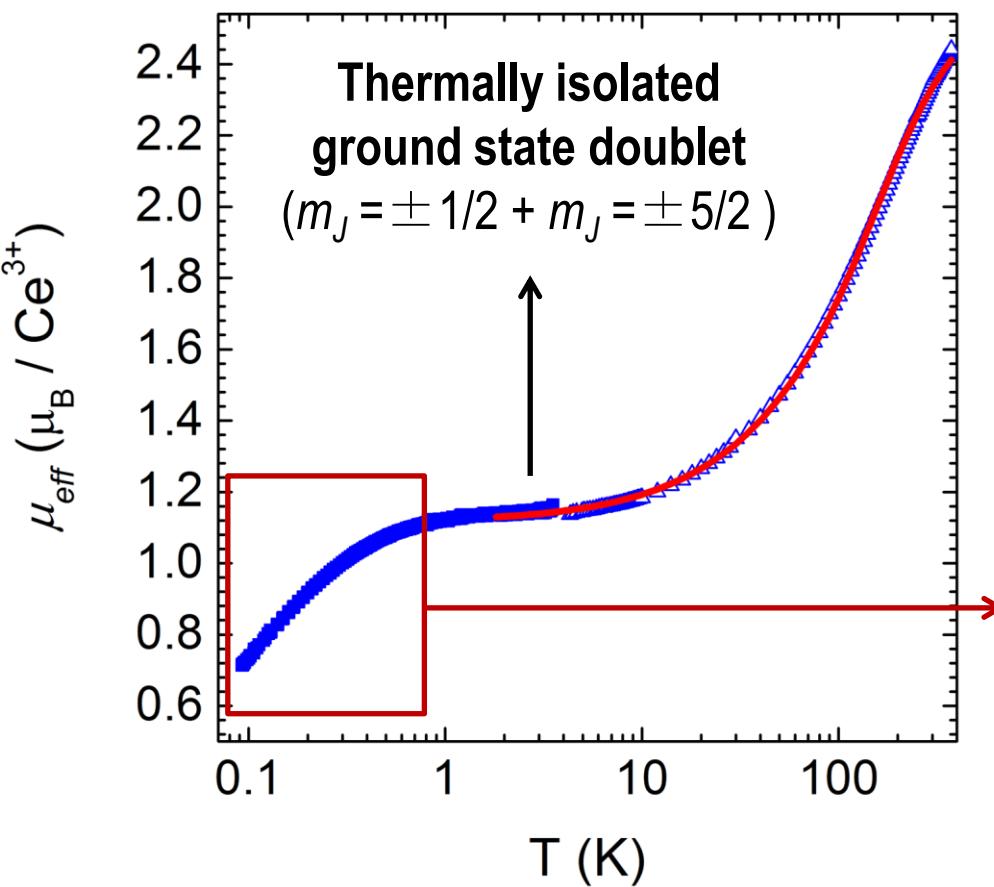
Susceptibility fit
(Crystal Field Hamiltonian)



Sibille et al., arXiv 1502.00662v1 (2015)

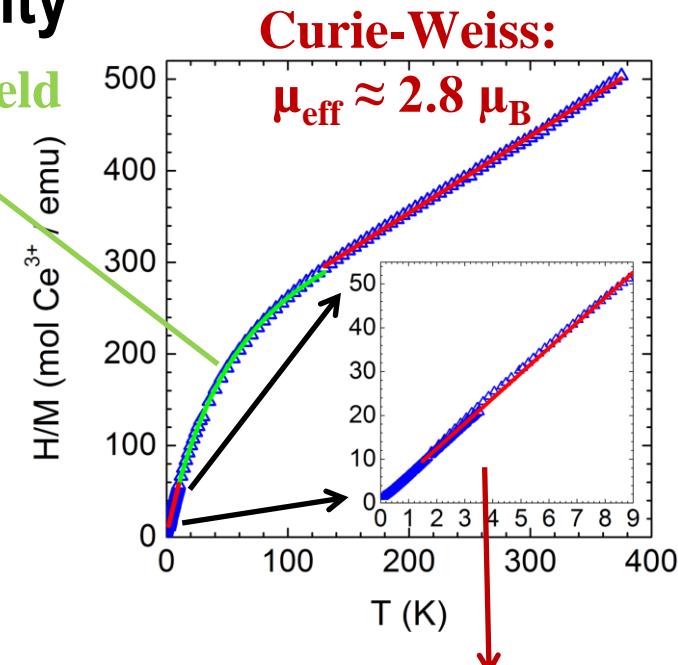
Low temperature magnetic susceptibility

→ sub-Kelvin correlations



Crystal-field effects

AF correlations



Curie-Weiss:
 $\mu_{\text{eff}} \approx 1.15 \mu_{\text{B}}$
 $\theta_p = -0.30(8)$

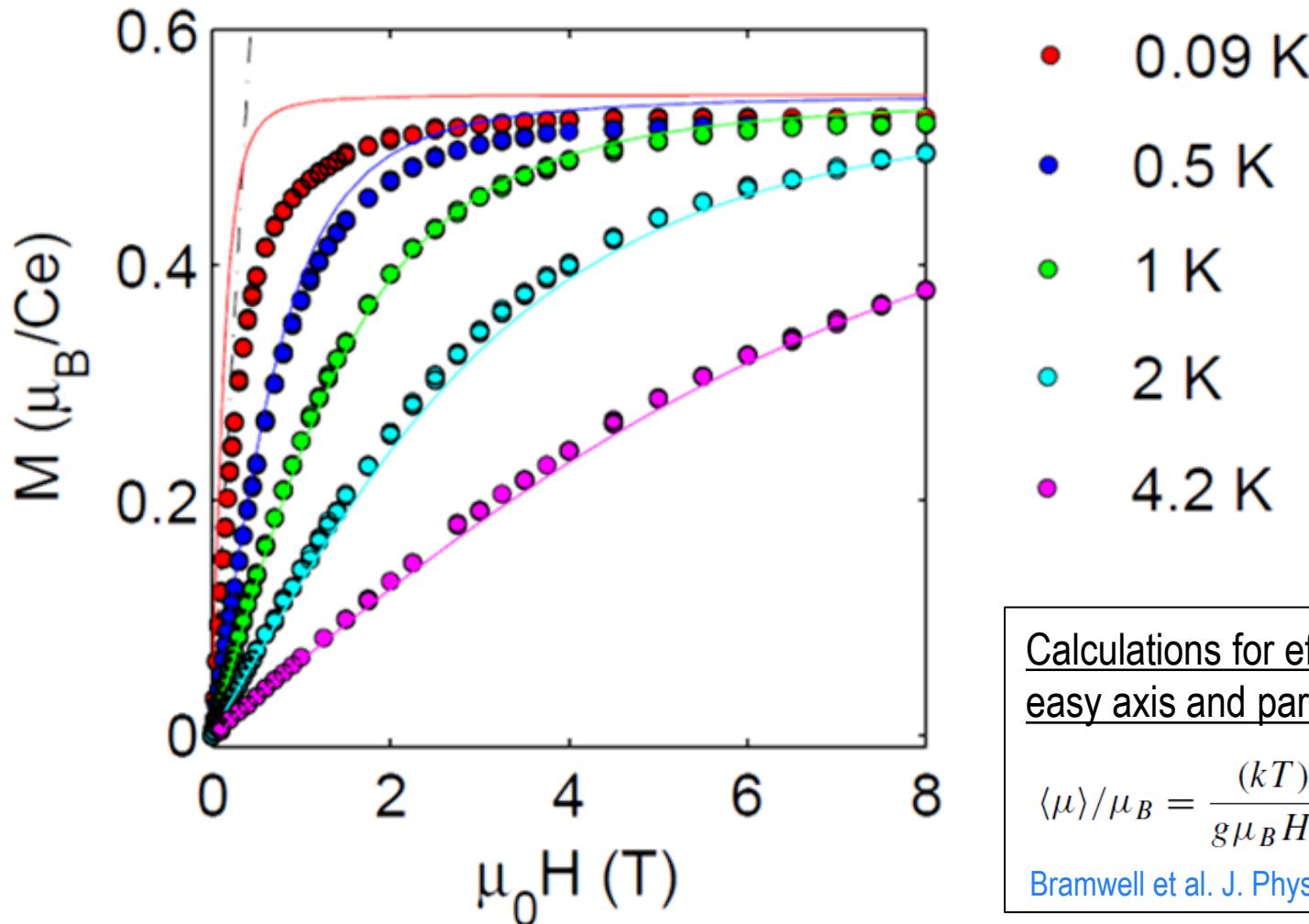
vs. $D \approx +0.025 \text{ K}$

Sibille et al., arXiv 1502.00662v1 (2015)

Low temperature magnetization curves

Sibille et al., arXiv 1502.00662v1 (2015)

→ local Ising anisotropy



Calculations for effective spins with $<111>$ easy axis and parameterized g-factor:

$$\langle \mu \rangle / \mu_B = \frac{(kT)^2}{g\mu_B H^2 S} \int_0^{g\mu_B HS/kT} x \tanh(x) dx$$

Bramwell et al. J. Phys. Cond. Mat., 12, 483-495 (2000)

Summary

Sibille et al., arXiv 1502.00662v1 (2015)

- well-ordered pyrochlore, small effective spins
- small Ising spins ($\approx 1.15 \mu_B$)
- but no order down to $0.02 \text{ K} \approx \theta_p/20$ (μSR)
- dynamical (μSR) correlated (AF) state below $\approx 0.5 \text{ K}$



origin of these fluctuations ?...

Interests

- J is an order of magnitude weaker than D (exchange-based physics)
- a model system for quantum effects on the pyrochlore lattice
(quantum spin liquid, quantum melting of classically predicted order)
- exotic excitations (photons) using neutrons (c.f. talk by T. Fennell)

Mein Dank geht an

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