Macroscopic and microscopic properties of magnetically frustrated $Tb_2Ti_2O_7$

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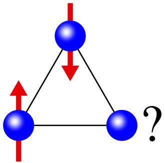
Groupement de Recherche
Matériaux et Interaction en COmpétition
Aspet, 12-15 octobre 2009

Outline

Geometrical magnetic frustration

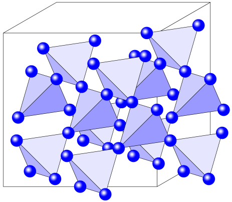
It arises when all pairwise interactions in a system cannot be satisfied simultaneously due to the geometry of the system



Three spins with antiferromagnetic interactions.

- Antiferromagnetically coupled Heisenberg spins on a pyrochlore lattice do not order down to $T = 0$
- Importance of other interactions:
  - exchange interactions with further neighbors
  - dipolar interaction
  - single ion anisotropy
  - etc

Example: spin ice state

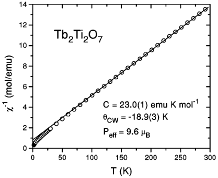


Pyrochlore crystal structure.

The puzzle of $Tb_2Ti_2O_7$

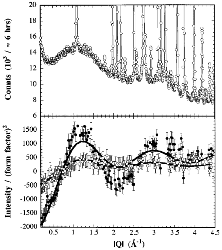
- Pyrochlore crystal structure
- Ising type anisotropy, but $Tb_2Ti_2O_7$ is a spin liquid, not a spin ice!
- Theory including Ising anisotropy, nearest neighbor exchange and dipolar coupling predicts order below $\simeq 1$ K

  (B.C. den Hertog and M.J.P. Gingras Phys. Rev. Lett. **84** 3430 (2000))
- Even more sophisticated theories, e.g. including ground and first excited CEF states, fail to catch the essential features of $Tb_2Ti_2O_7$



No magnetic order down to 50 mK

M.J.P. Gingras *et al*, Phys. Rev. B **62** 6496 (2000)



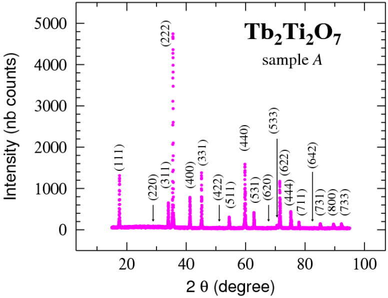
Spin liquid correlations

J.S. Gardner *et al*, Phys. Rev. Lett. **82** 1012 (1999)

Crystal growth of $Tb_2Ti_2O_7$

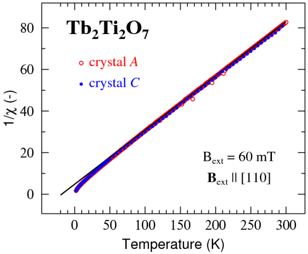
Growth conditions of different crystals

crystals	initial powders	growth rate, flow
A	$TiO_2 + Tb_4O_7$	8 mm/h, argon
B	$TiO_2 + Tb_4O_7$	8 mm/h, argon
C	$TiO_2 + Tb_2O_3$	7 mm/h, oxygen
D	$TiO_2 + Tb_4O_7$	3 & 8 mm/h, argon

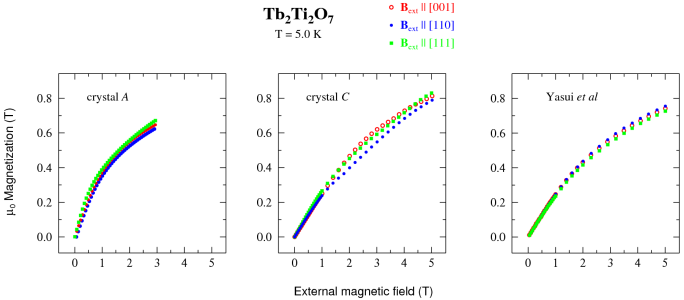


Typical x-ray powder diffraction pattern.
No foreign phase detected.

Susceptibility and magnetization of $Tb_2Ti_2O_7$

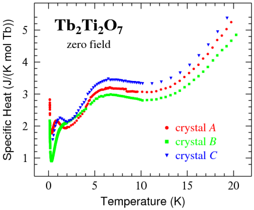


- Above 2 K, susceptibility independent of sample
- Slight differences in low temperature magnetization



Specific heat of $Tb_2Ti_2O_7$

Our data



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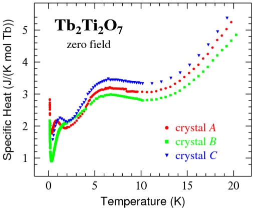

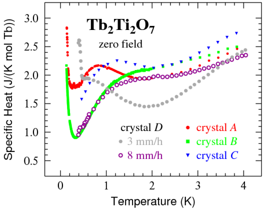

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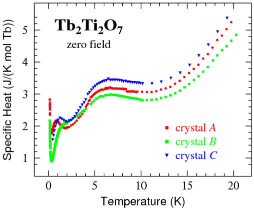
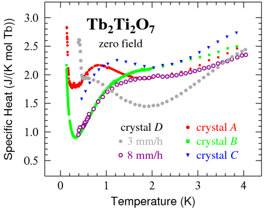
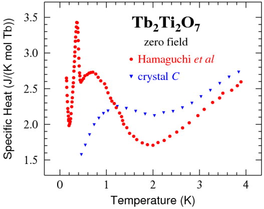




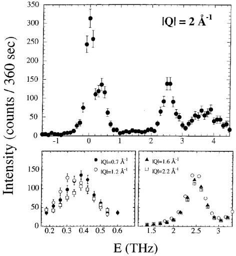
- Low temperature specific heat very sensitive to crystal growth conditions
- Transition at $\simeq 0.4$ K seems related to crystal growth velocity

Comparison with published data



N. Hamaguchi *et al*, Phys. Rev. B **69** 132413 (2004)

Crystal field levels of Tb^{3+} in $Tb_2Ti_2O_7$



J.S. Gardner *et al*, Phys. Rev. B **64** 224416 (2001)

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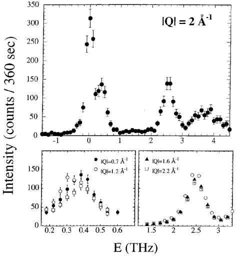

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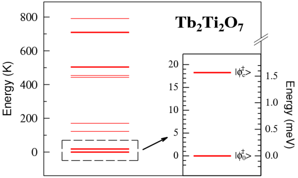

Two low-lying doublets at 0 and 18 K.

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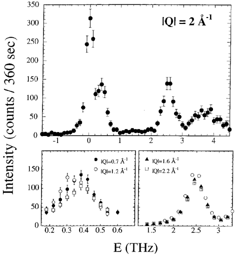

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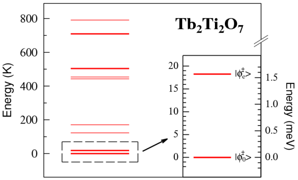

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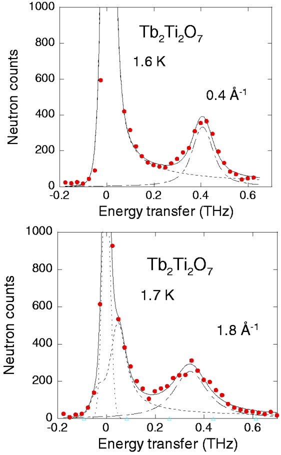

I. Mirebeau *et al*, Phys. Rev. B **76** 184436 (2007)

Indication for an extra CEF level around 2 K.

Entropy variation and CEF levels

- First hypothesis: two doublets separated by an energy Δ.
  - For $T \gg \Delta$: $S = R \log 4$
  - For $T \ll \Delta$: $S = R \log 2$
  - Entropy variation:
    $\Delta S = R(\log 4 - \log 2) = R \log 2$

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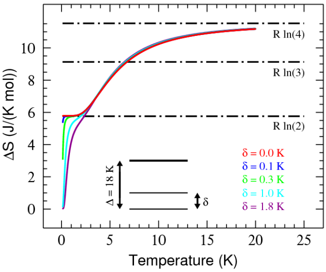
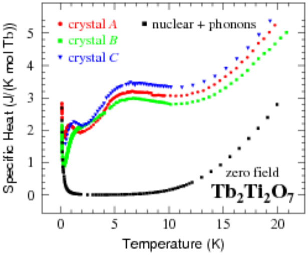


Entropy variation from $T = 0.13$ K to $+\infty$

Entropy variation in $Tb_2Ti_2O_7$



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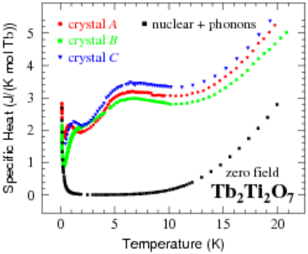

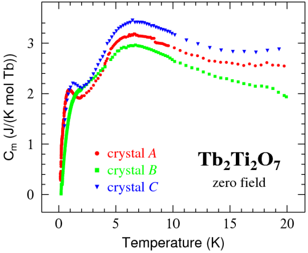

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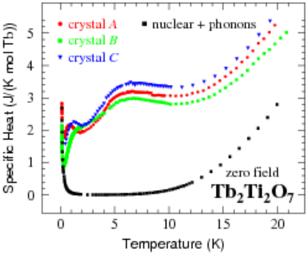
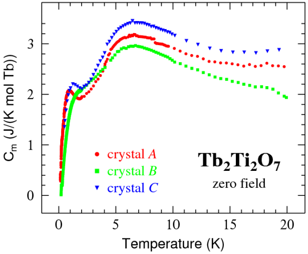
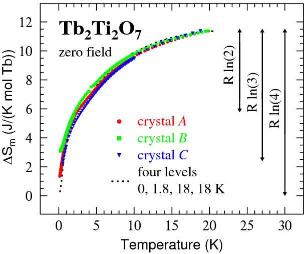




$$\Delta S(T_1 \rightarrow T_2) = \int_{T_1}^{T_2} \frac{C_m}{T} \, \mathrm{d}T$$

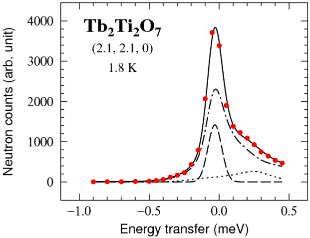


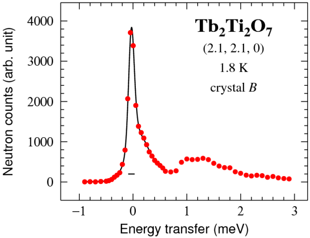
Entropy variation consistent with levels at 0, 1.8, 18 and 18 K

$\rightarrow$ the lowest levels are two singlets

Spin dynamics in $Tb_2Ti_2O_7$

Inelastic neutron scattering experiments (IN12 at Institut Laue Langevin, Grenoble)





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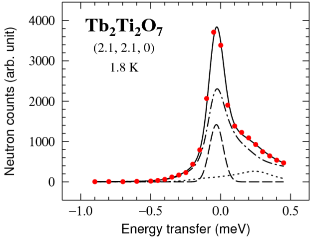

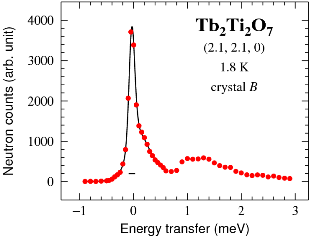

Three contributions to the low energy intensity:

- quasi-elastic scattering (Lorentzian of width Γ)
- weakly inelastic signal (CEF transition)
- incoherent scattering

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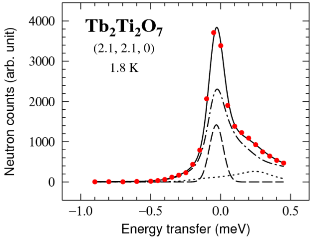

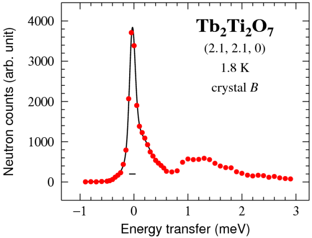

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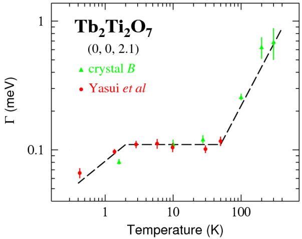
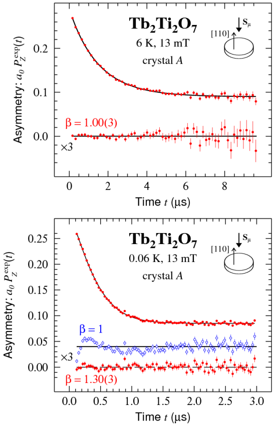


Three temperature regimes for Γ

Spin dynamics in $Tb_2Ti_2O_7$

Muon spin relaxation measurements (ISIS, UK and SμS at PSI, Switzerland)



Exponential-power relaxation:

$$P_Z(t) = \exp[-(\lambda_Z t)^\beta]$$

Below $T \simeq 2$ K, $\beta > 1$

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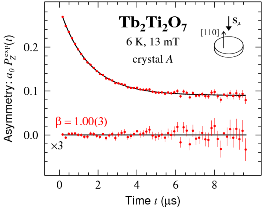

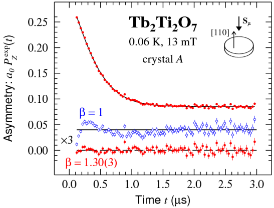

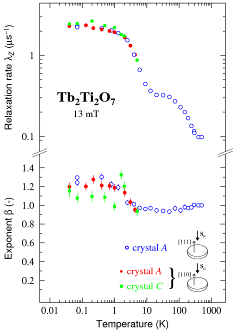

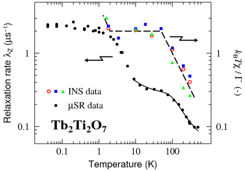

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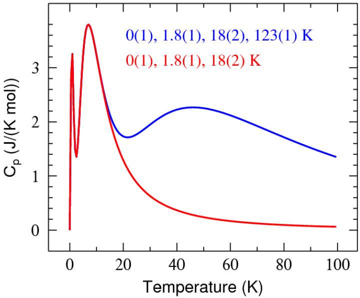
Conclusion and Outlook

- Issues in sample preparation
- Specific heat is a sensitive probe
- Entropy variation: two singlets as lower energy CEF levels
- Two characteristic temperatures in the spin dynamics
  - above 50 K: relaxation through high energy CEF levels
  - below 2 K: slowing down of Tb^{3+} fluctuations

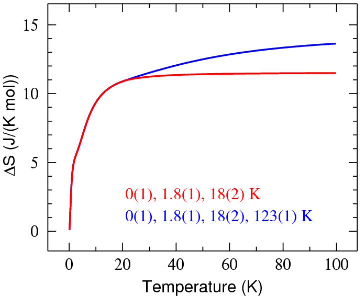
Further work

- Insight into the difference in samples
- Checking that microscopic probe results are robust
- Influence of the presence of two low-lying singlets on current models
- Why is $Tb_2Ti_2O_7$ so different from sister compound $Tb_2Sn_2O_7$?

Influence of higher energy CEF levels on entropy variation



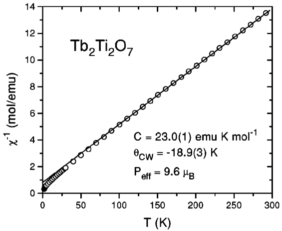
Simulated specific heat



Simulated entropy variation

Susceptibility of $Tb_2M_2O_7$

$Tb_2Ti_2O_7$



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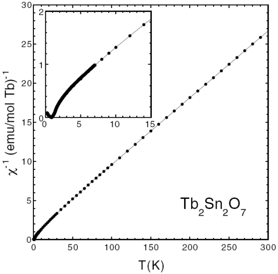
M.J.P. Gingras *et al*, Phys. Rev. B **62** 6496 (2000)

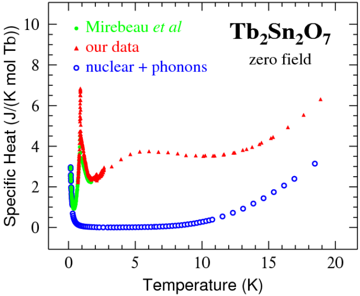
$Tb_2Sn_2O_7$

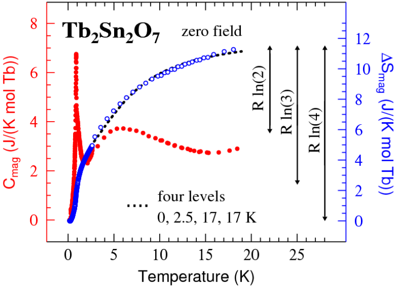


$\theta_{\mathrm{CW}} = -13$ K, $T_{\mathrm{N}} = 0.88$ K

K. Matsuhira *et al*, J. Phys. Soc. Japan **71** 1576 (2002)

Specific heat and entropy variation in $Tb_2Sn_2O_7$





Entropy variation consistent consistent with levels at 0, 2.5, 17 and 17 K.