# Muon spin relaxation studies of geometrically frustrated magnets

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# Outline


Introduction

- Geometrical magnetic frustration
- Muon spin rotation and relaxation

Exotic spin dynamics in the ordered phase of geometrically frustrated $Tb_2Sn_2O_7$

- Bulk magnetic properties
- $\mu$SR response
- Neutron scattering confirmation for spin dynamics

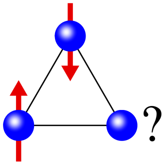
Conclusions and Outlook

# Geometrical magnetic frustration

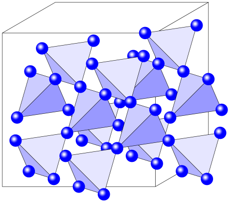
It arises when all pairwise interactions in a system cannot be satisfied simultaneously due to the geometry of the system

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

  Example: spin ice state (with magnetic excitations formally equivalent to magnetic monopoles).



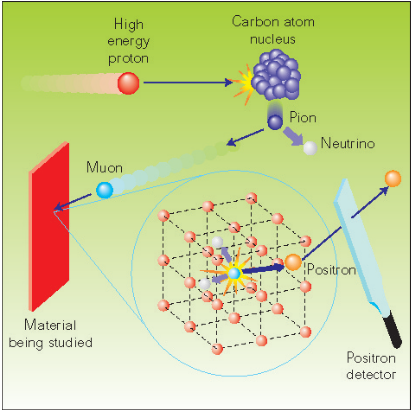

*Three spins with antiferromagnetic interactions.*

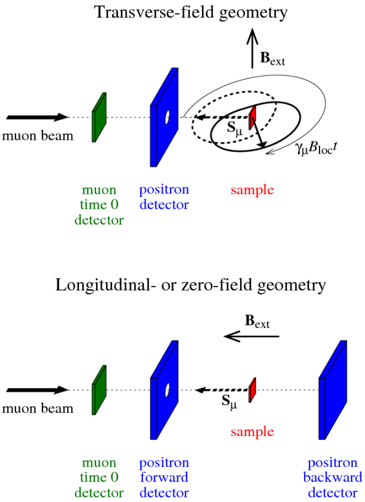


*Pyrochlore crystal structure.*

# Muon spin rotation and relaxation

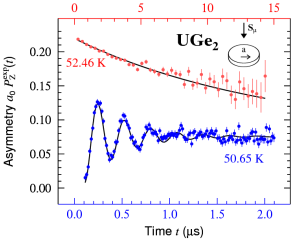
An experimental tool, technically close to TDPAC, which uses fully polarised implanted muons.

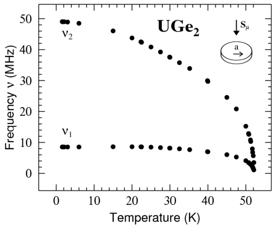


# Muon spin rotation and relaxation

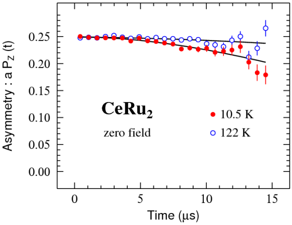
## The ultimate tool to detect the presence of a magnetic transition

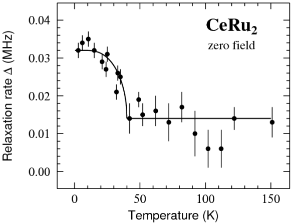



*Spontaneous muon spin precession below $T_C \simeq 52$ K. $\mu_U = 1.4\ \mu_B$.*

*Sakarya* et al *Phys. Rev.* **B81**, *024429 (2010).*



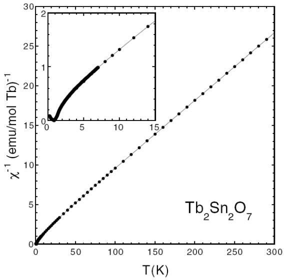




*Magnetic transition with $\mu_{Ce} \simeq 10^{-4} \mu_B$ below 40 K.*

*Huxley* et al *Phys. Rev.* **B54**, *R9666 (1996).*

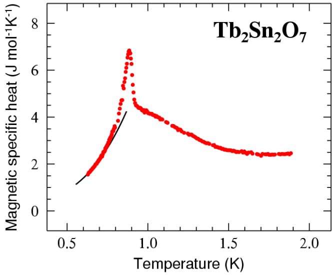
# $Tb_2Sn_2O_7$: a magnet on a pyrochlore lattice

## Bulk magnetic properties




*Inverse magnetic susceptibility.*

Matsuhira *et al*, JPSJ **71**, 1576 (2002).




*Specific heat vs temperature.*

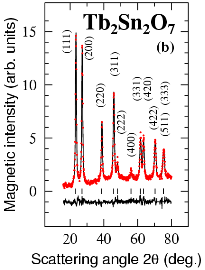
Mirebeau *et al*, PRL. **94** (2005) 246402,

Dalmas de Réotier *et al*, PRL **96** (2006) 127202.

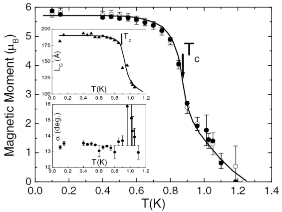
- Antiferromagnetic interactions: $\Theta_{\rm CW} = -12.5$ K.
- Transition at $T_{\rm lr} = 0.88$ K.

$|\Theta_{\rm CW}| \gg T_{\rm lr}$ : $Tb_2Sn_2O_7$ is a geometrically frustrated system.

# Powder neutron diffraction




*Magnetic powder diffraction pattern, (subtraction of 0.11 and 1.23 K data).*

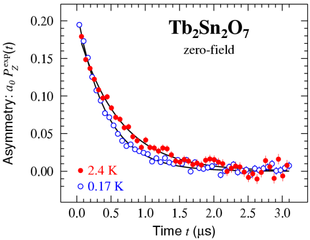


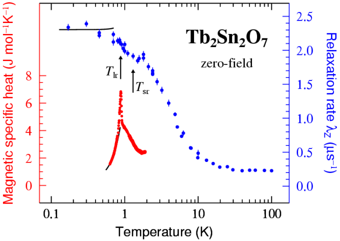

*Thermal variation of ordered $Tb^{3+}$ magnetic moment.*

- Abrupt transition at $T_{\rm lr} = 0.88$ K to an ordered spin-ice structure ($\mathbf{k} = 0$): 2 spins in/2 spins out + ferromagnetic component (2.2 $\mu_B$).
- Smeared transition at $T_{\rm sr} = 1.3\,(1)$ K.
- Magnetic moment at low temperature: $\mu_{\rm Tb} = 5.4$ (1) $\mu_{\rm B}$.

# Zero-field muon spin relaxation spectra

Dalmas de Réotier *et al*, PRL **96**, 127202 (2006).




*Spectra with exponential shape in the whole temperature range,*

$$P_Z(t) = \exp(-\lambda_Z t).$$

*Spin-lattice relaxation rate $\lambda_Z(T)$ and specific heat.*

Consistent with another $\mu SR$ study (Bert *et al*, PRL **97**, 117203 (2006)).

# $Tb_2Sn_2O_7$: a magnetic phase transition with no $\mu$SR signature !

Spontaneous field at muon site is the $Tb^{3+}$ dipolar field.
No signature of it because, either

- it cancels for symmetry reason.
  Unlikely because of
  - low symmetry expected for the muon site
  - relatively complicated magnetic structure with ferromagnetic and antiferromagnetic components
- or it is dynamical.

# The polarisation function $P_Z(t)$ for a stochastic field

Account of field dynamics by a Gaussian Markovian process:

$$P_Z(s) = \frac{P_Z^{\mathrm{stat}}(s+\nu_c)}{1-\nu_c P_Z^{\mathrm{stat}}(s+\nu_c)}$$

Assumption: $\mathbf{B}_{\mathrm{loc}}$ takes two values $\pm\mathbf{B}_{\mathrm{fl}}$: $P_Z^{\mathrm{stat}}(t) = \cos(\gamma_\mu B_{\mathrm{fl}} t)$.

$$P_Z^{\mathrm{stat}}(s) = \frac{s}{s^2+\gamma_\mu^2 B_{\mathrm{fl}}^2}; \qquad P_Z(s) = \frac{s+\nu_c}{s^2+\nu_c s+\gamma_\mu^2 B_{\mathrm{fl}}^2}.$$

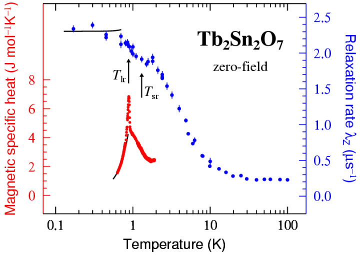
Two cases:

- $\nu_c < 2\gamma_\mu B_{\mathrm{fl}}$ : $\quad P_Z(t) = \exp(-\nu_c t)\dfrac{\cos(\omega_{\mathrm{eff}} t-\varphi)}{\cos\varphi}$
- $\nu_c > 2\gamma_\mu B_{\mathrm{fl}}$ : $\quad P_Z(t) = \exp(-\nu_c t)\dfrac{\cosh(\nu_{\mathrm{eff}} t-\varphi)}{\cosh\varphi}$

In the extreme motional narrowing limit: $\nu_c \gg 2\gamma_\mu B_{\mathrm{fl}}$

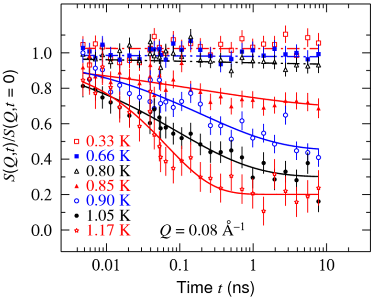
$$P_Z(t) = \exp(-\lambda_Z t) \quad \text{with} \quad \lambda_Z = \frac{\gamma_\mu^2 B_{\mathrm{fl}}^2}{\nu_c}$$

# Estimate of $\nu_c$ in $Tb_2Sn_2O_7$

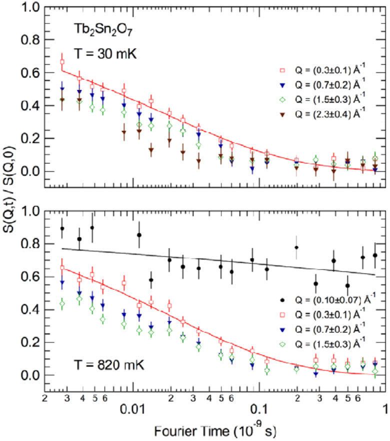



- $\lambda_Z = \gamma_\mu^2 B_{\text{fl}}^2 / \nu_c$
- $B_{\text{fl}} = 0.2$ T
  $\Rightarrow \nu_c \simeq 10^{10}$ s$^{-1}$.

# Neutron scattering evidence for spin dynamics when $T \to 0$ in $Tb_2Sn_2O_7$




Chapuis *et al*, JPCM **19** 446206 (2007).




Rule *et al*, JPCM **21** 486005 (2009).

Neutron spin echo data: spin correlations are static for $Q \to 0$ and dynamical ($5 \times 10^{10}$ s$^{-1}$) for $Q \gtrsim 0.3$ Å$^{-1}$.

# Conclusions and Outlook

- Spin dynamics for $T \to 0$, *i.e.* in the ordered phase.
- Broad range of time scales,
  static ferromagnetic component,
  antiferromagnetic fluctuations with $\nu_c \simeq 10^{10}$ s$^{-1}$.
- Origin of these dynamics ?
  Relation with other ordered frustrated magnets ($Gd_2Ti_2O_7$, $Gd_2Sn_2O_7$, $Er_2Ti_2O_7$, $Cu_2Cl(OH)_3$...).
- Muons are insensitive to ferromagnetic component.
  Need for a detailed investigation of muon-system coupling.

Overall moral of the story: persistent spin dynamics in the ordered phase of an exotic magnet may hinder the detection of a phase transition, even when using a very sensitive probe.

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# Muon Spin Rotation, Relaxation, and Resonance

## Applications to Condensed Matter

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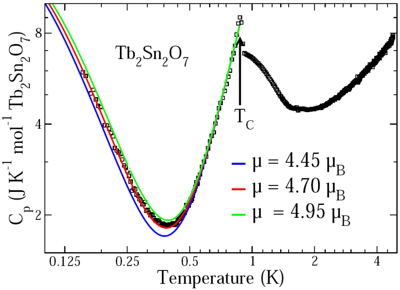
# Neutron diffraction vs nuclear specific heat




*Specific heat vs temperature.*

Bonville, JPCS **217**, 012119 (2010).

$T \lesssim$ 0.4 K: Schottky anomaly arising from nuclear specific heat.

Analysis gives $\mu_{\mathrm{Tb}} =$ 4.7 (5) $\mu_{\mathrm{B}}$ $\neq$ 5.4 (1) $\mu_B$ obtained from neutron diffraction.

Hint to explain this possible discrepancy:
Persistent spin fluctuations in the ordered phase and $\tau_c \simeq T_1$, with $\tau_c$, the characteristic electronic fluctuation time, and $T_1$, the nuclear relaxation time.

# The polarisation function $P_Z(t)$ for a stochastic field

Account of field dynamics by a Gaussian Markovian process.

$$P_Z(t) = \sum_{\ell=0}^{+\infty} R_\ell(t)$$

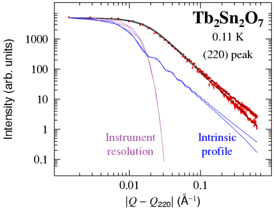
$R_\ell(t)$ is the average contribution of the muons which have experienced $\ell$ changes of field between time 0 and $t$.

Example: $R_0(t) = P_Z^{\mathrm{stat}}(t) \exp(-\nu_c t)$.

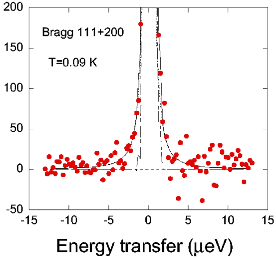
The following relation between the static and dynamical Laplace transforms can be shown:

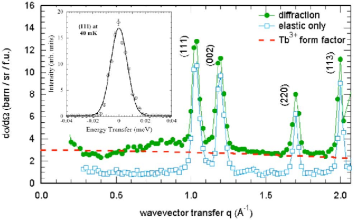
$$P_Z(s) = \frac{P_Z^{\mathrm{stat}}(s + \nu_c)}{1 - \nu_c P_Z^{\mathrm{stat}}(s + \nu_c)}$$

# Neutron scattering evidence for spin dynamics when $T \to 0$ in $Tb_2Sn_2O_7$

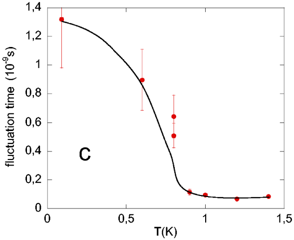



Chapuis *et al*, JPCM **19** 446206 (2007).




Rule *et al*, PRB **76** 212405 (2007).




Mirebeau *et al*, PRB **78** 174416 (2008).