#### Title: Quantum fluctuations in spin ice

#### Date: Apr 26, 2011 04:00 PM

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Abstract: Geometrical frustration in magnetic systems provides a rich playground to study the emergence of novel ground states. In systems where not all magnetic couplings can be simultaneously satisfied, conventional long range magnetic order is often precluded, or pushed to much lower emperature scales than would be expected from the strength of the magnetic interactions. Dy2Ti2O7 has a pyrochlore lattice, where the magnetic Dy ions lie on the vertices of corner sharing tetrahedra. The Dy magnetic moments are constrained by crystal fields to lie along local <111&gt; axes, pointing towards or away from the tetrahedral centres. With a ferromagnetic interaction between nearest neighbours, the ground state for each tetrahedron has two spins pointing in and two out. Due to the number of possible ways of satisfying this constraint, the overall ground state is highly degenerate; in fact this system can be mapped onto the problem of water ice where each oxygen atom has two strongly bound and two weakly bound protons, leading the magnetic problem to be referred to as spin ice. Recent theoretical work in understanding the magnetic excitation in spin ice, where the spin flip excitations can be described in terms of magnetic monopoles. Bramwell et al., reported muon spin rotation measurements of spin ice which they interpreted in terms of this monopole picture. In contrast to the work of Bramwell et al., our muon spin relaxation measurements do not exhibit highly temperature dependent Arrhenius processes expected for monopoles. Instead we report temperature independent spin fluctuations well into the spin ice state and that the previous interpretations are incorrect.

# Low-Temperature Spin Dynamics in Spin Ice

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Canadian Institute for Advanced Research

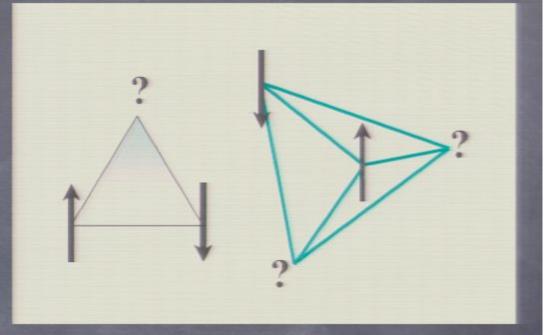
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### Collaborators

- S.R. Dunsiger, A.A. Aczel, T. Goko, F. Ning, Y.J. Uemura, T.J. Williams
- H. Dabkowska, A. Dabkowski, H. Noad
- B. Javanparast, T. Lin, M.J.P. Gingras

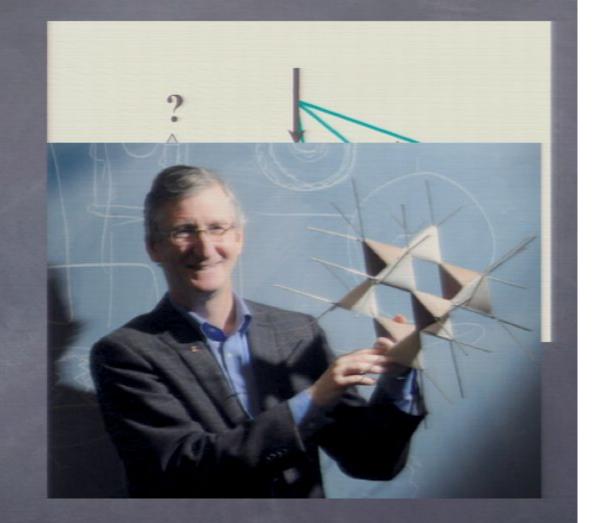
### Geometrical Frustration

- No periodic arrangements of moments which can simultaneously minimize energy.
- Can have large number of classically degenerate ground states.
- Leads to competing low temperature phases and enhanced quantum mechanical spin dynamics.



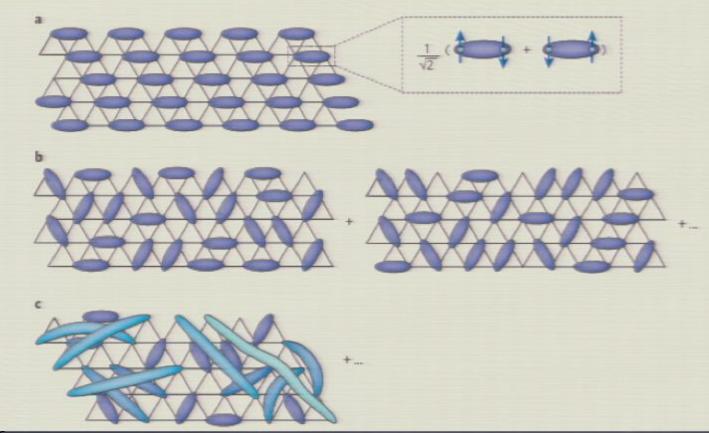
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### Spin Liquids

- Magnetic system that don't develop long range magnetic order down to lowest temperatures. Proposed by Anderson (RVB – cuprates).
- Only a few candidate materials have been positively identified.



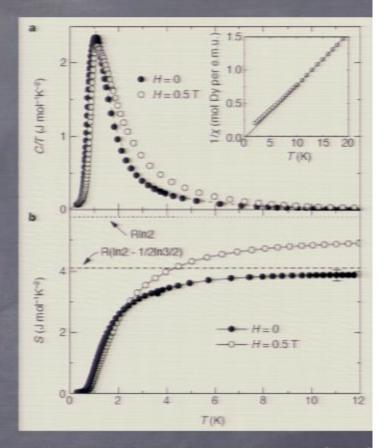
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Balents, Nature 464, 199 (2010).

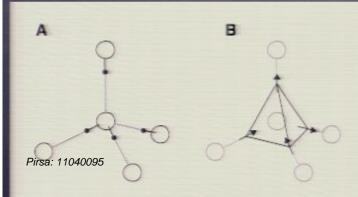
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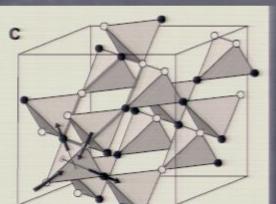
#### Spin Ice

- Ho<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>, Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>, Ho<sup>3+</sup>/Dy<sup>3+</sup> occupy vertices of corner-sharing tetrahedra in pyrochlore lattice.
- Strong crystal field anisotropy results in "Ising"-spins, either in or out along local <111>.
- Ferromagnetic interactions give "icerules" - 2 in / 2 out. 6-fold degeneracy for each tetrahedron.
- macroscopic degeneracy, corresponding to water ice.



Ramirez et al., Nature 399, 333 (1999)

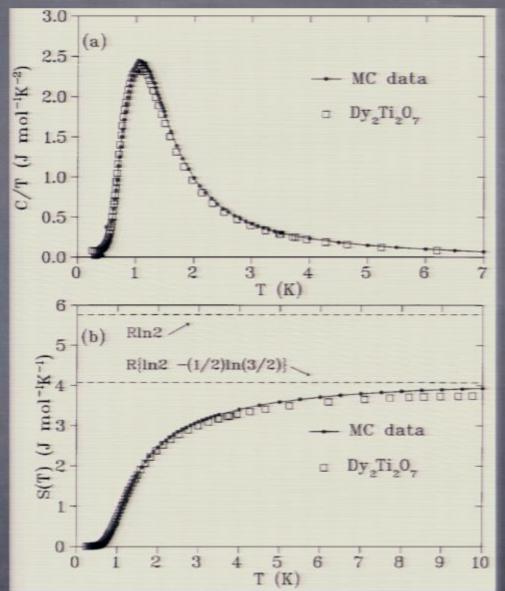




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### Spin Ice

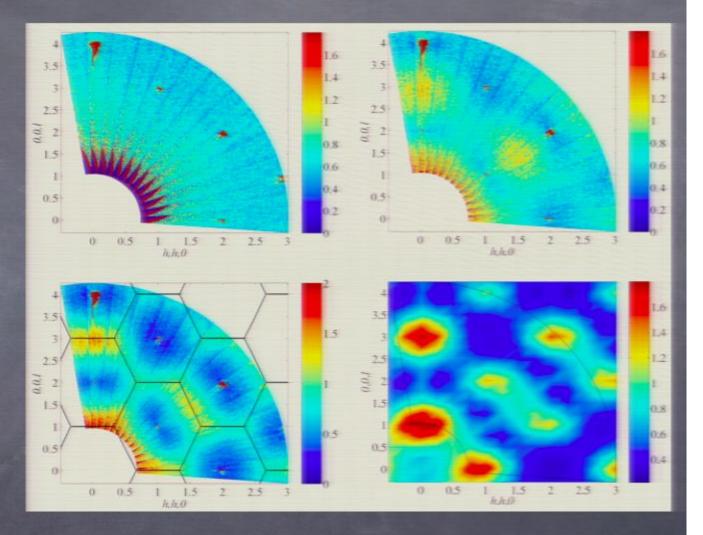
- Ho<sup>3+</sup> and Dy<sup>3+</sup> have large magnetic moment ~10µ<sub>B</sub>.
- Dipole interactions large D<sub>nn</sub>~1K.
- Exchange J<sub>nn</sub>: Need J<sub>eff</sub>=J<sub>nn</sub>+D<sub>nn</sub>>0.
- Upon cooling, spin configurations which don't obey ice rules freeze out.
  - gives a crossover in C<sub>p</sub>.
- Spin ice state should have extremely slow dynamics due to Ising nature precluding classical flips or tunneling.



den Hertog & Gingras, Phys. Rev. Lett. 84, 3430 (2000).

### Spin Ice - No Long Range Order

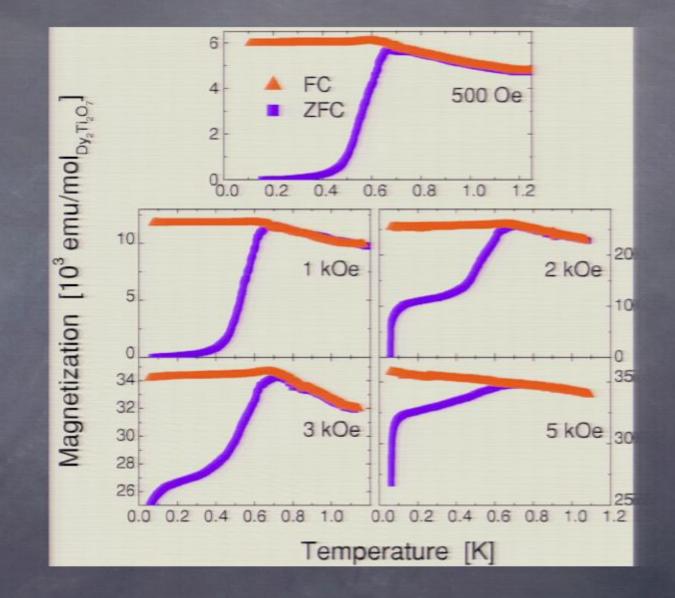
- Only diffuse magnetic peaks seen.
- Short range FM correlations.



Fennell et al., Phys. Rev. B 70, 134408 (2004).

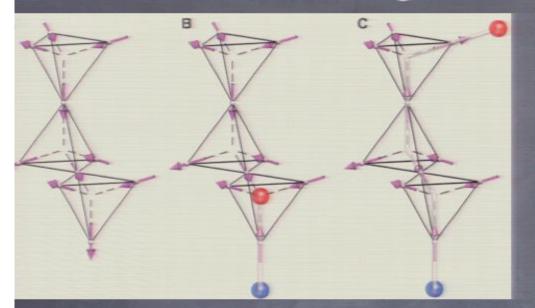
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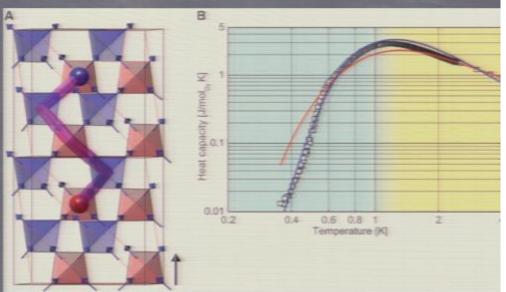
### Dy2Ti2O7 Low Temperature Irreversibility



Snyder et al., Phys. Rev. B 69, 064414 (2004). Page 10/44

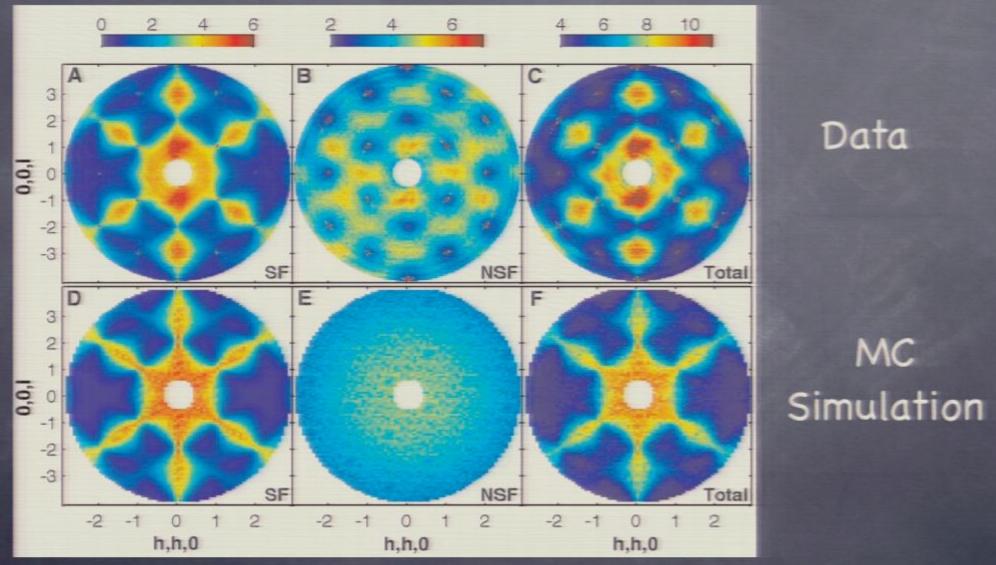
### Magnetic Monopoles?





- Spin flip excitations from ground state create magnetic dipoles which can separate with minimal energy cost.
- Separated excitations correspond to magnetic monopoles.
  - Interact via Coulomb potential

### Ho2Ti2O7 Magnetic Neutron Scattering

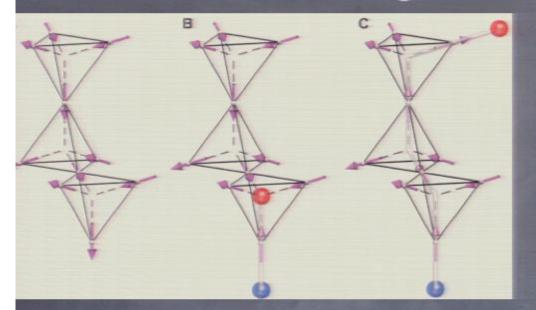


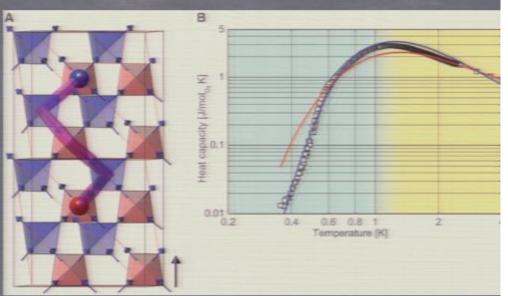
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Fennell et al., Science 415, 1177582 (2009).

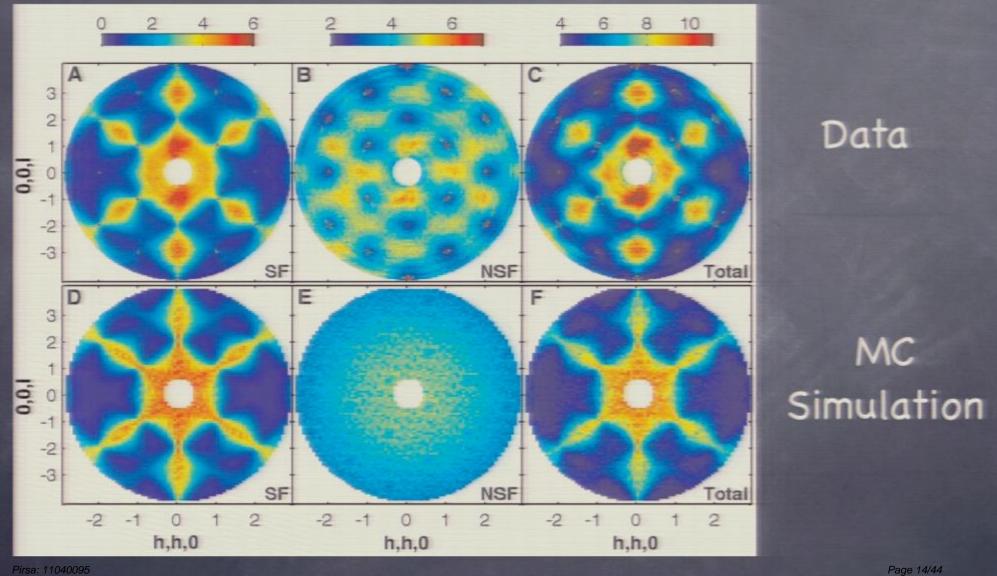
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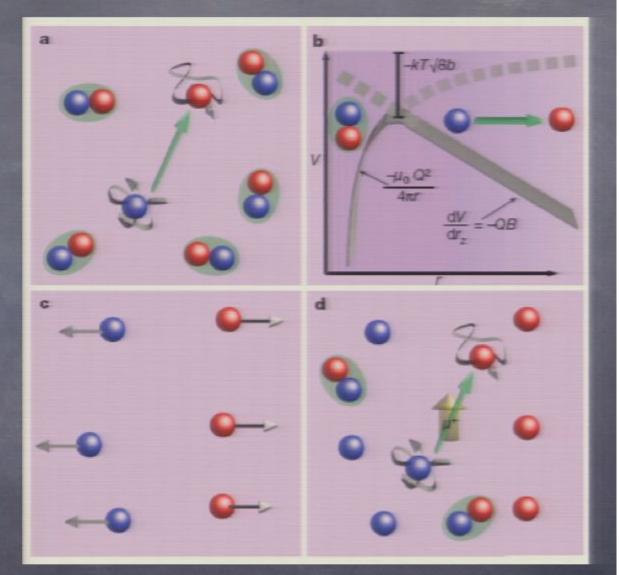
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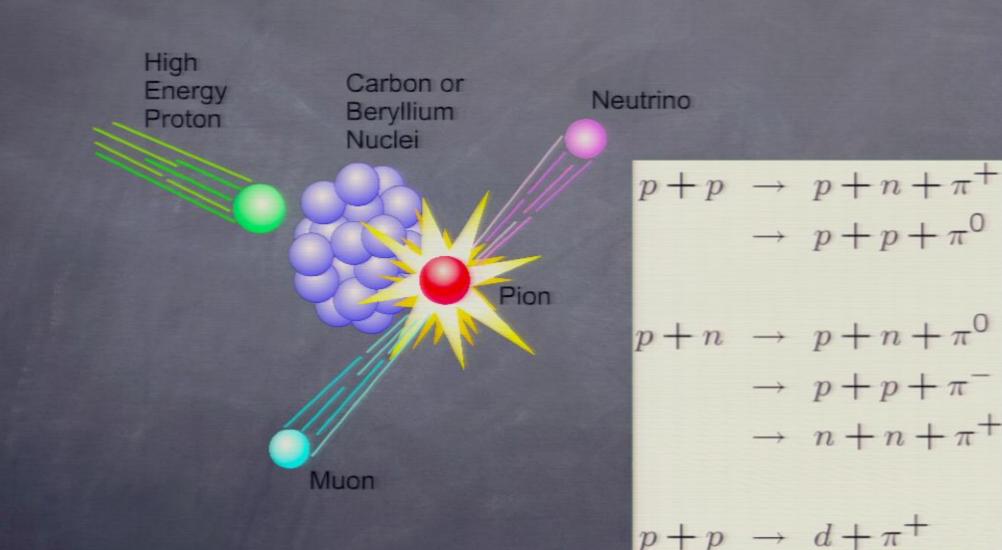
### Monopoles in a Magnetic Field

- Excitations correspond to .monopoles which are thermally excited.
- Application of dc magnetic field causes separation of monopoles (Coulomb force).
- Separated monopoles gives field fluctuation rate (like hyperfine field).



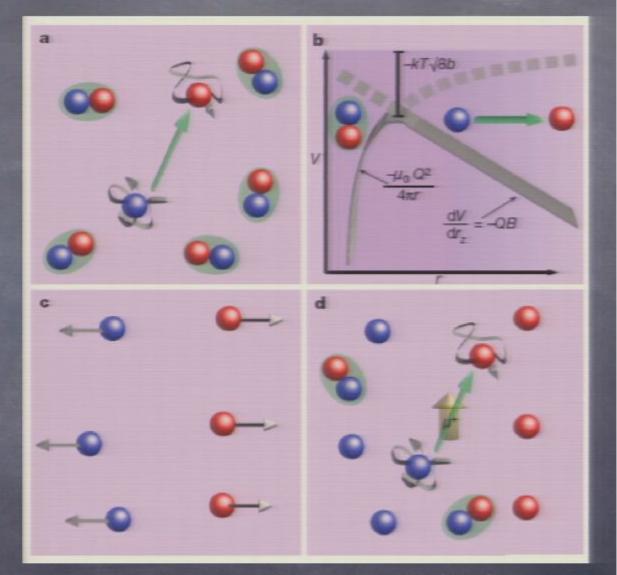
#### Bramwell et al., Nature 461 956 (2009).

### **Pion Production**

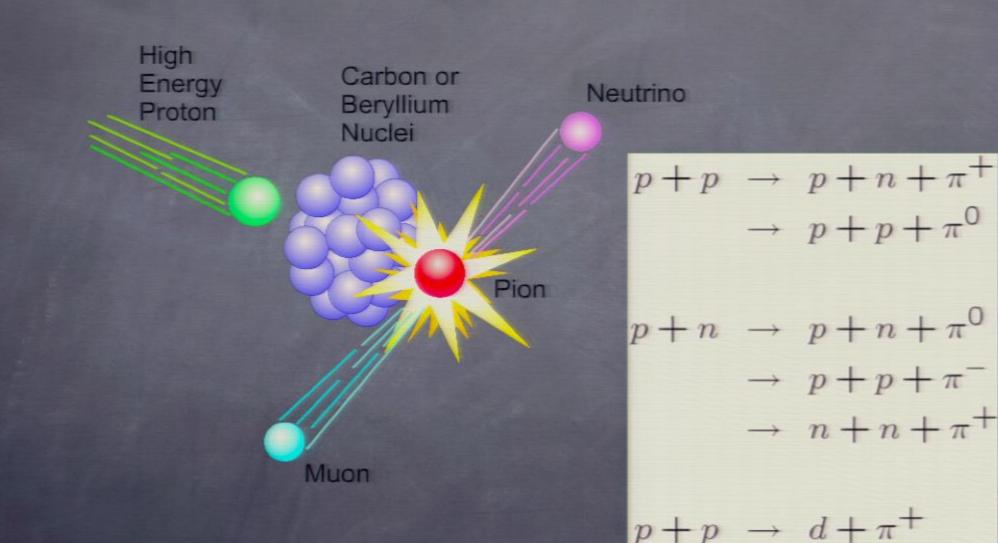


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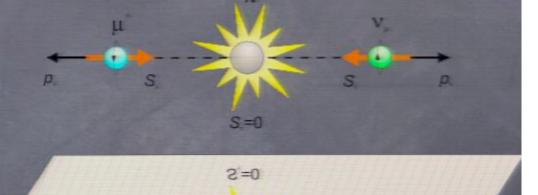


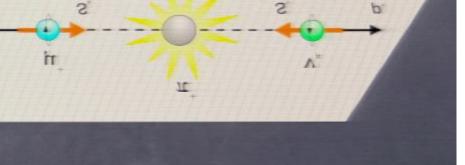
### Pion Decay

b

- π⁺→μ⁺+ν<sub>µ</sub>
  - mono-energetic E=4.1MeV
- Conservation of linear momentum, parity violation
  - Muon neutrinos have spin ½ and momentum antiparallel (ie. lefthanded spin, -1 helicity).
  - muons ~100% spin polarized

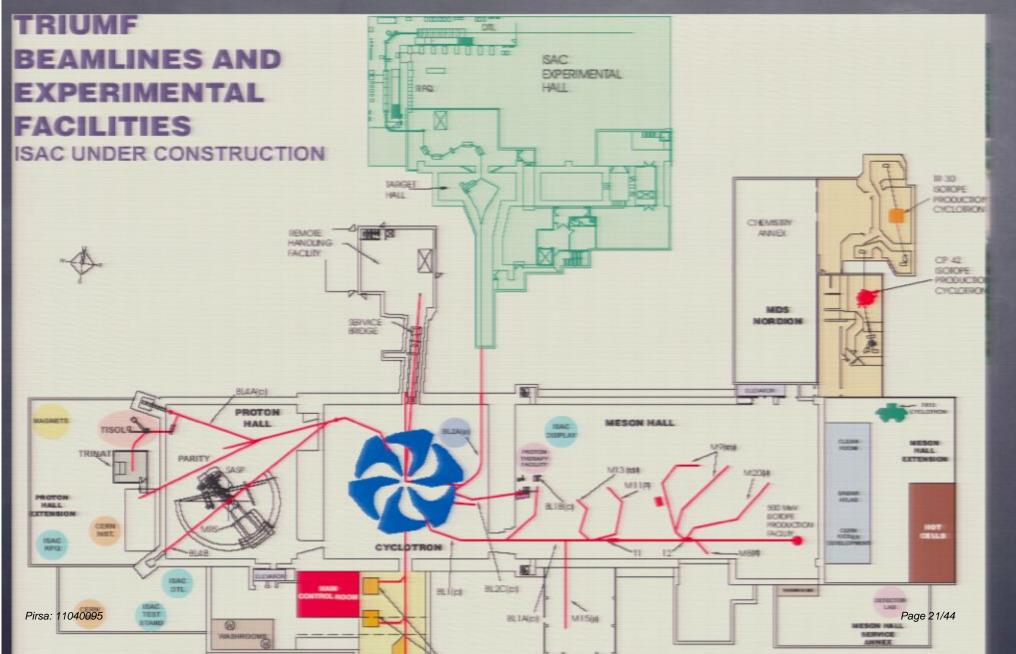








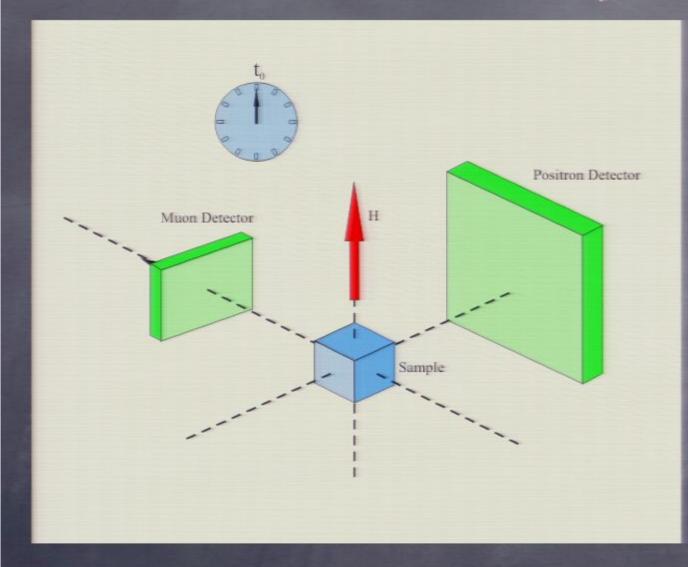
#### TRIUMF



## TRIUMF Experimental Hall

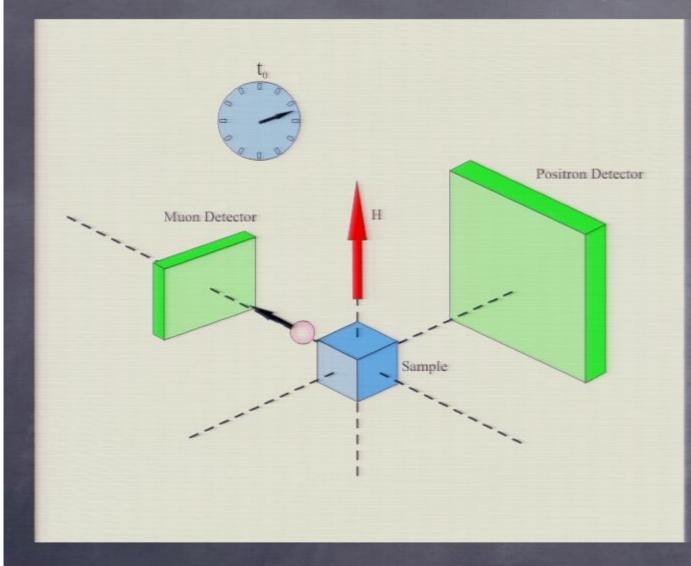




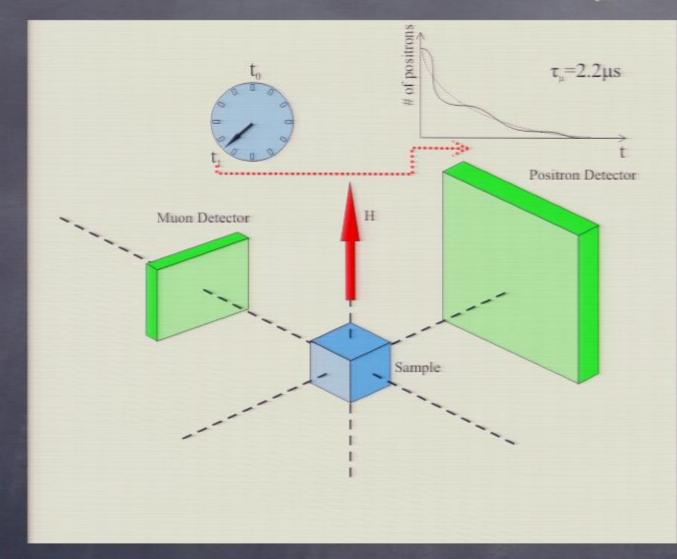


 $\mu^+ \to e^+ + \nu_\mu + \bar{\nu}_e$  $\tau_\mu = 2.2 \mu s$ 

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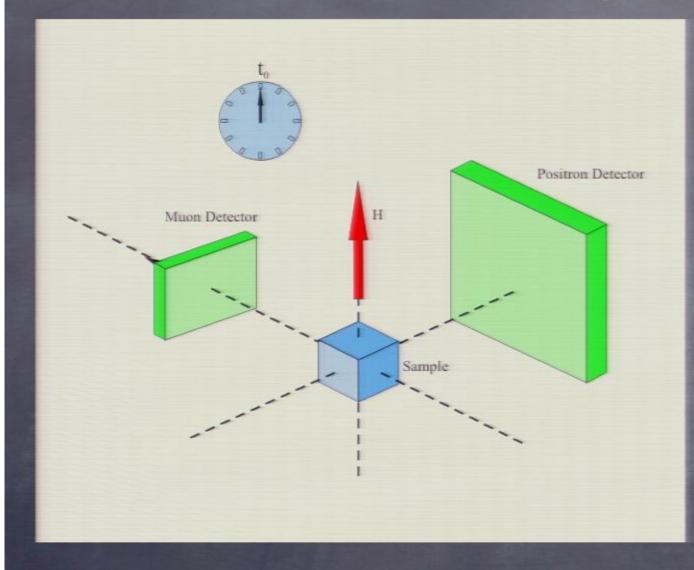


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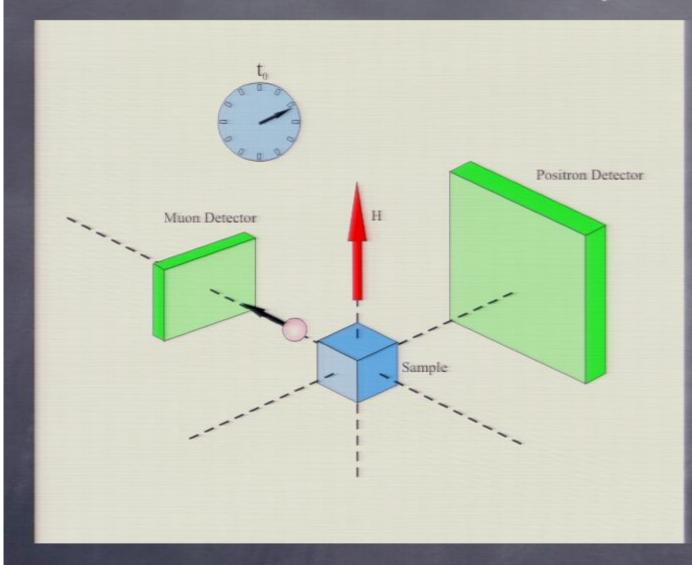
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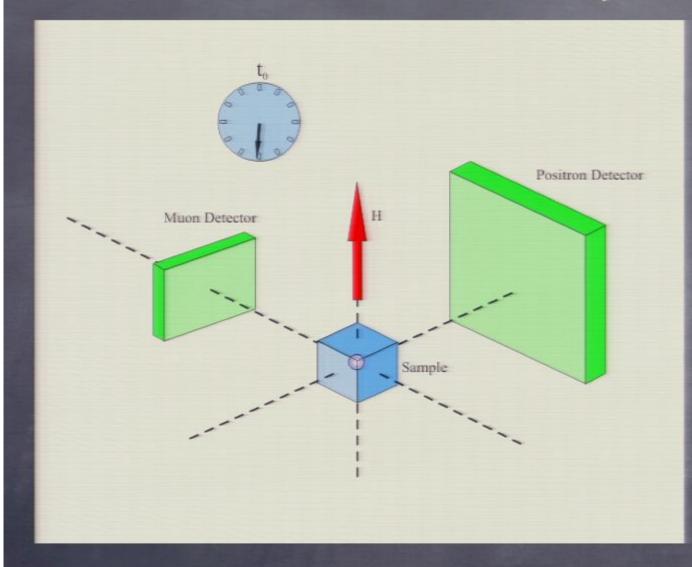




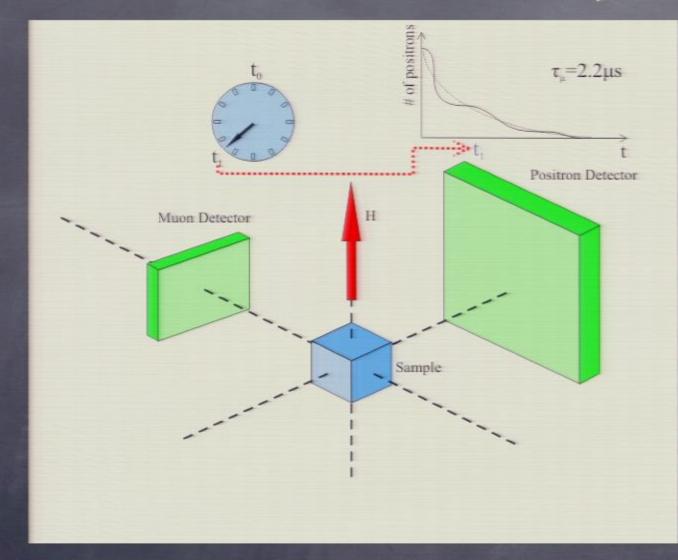
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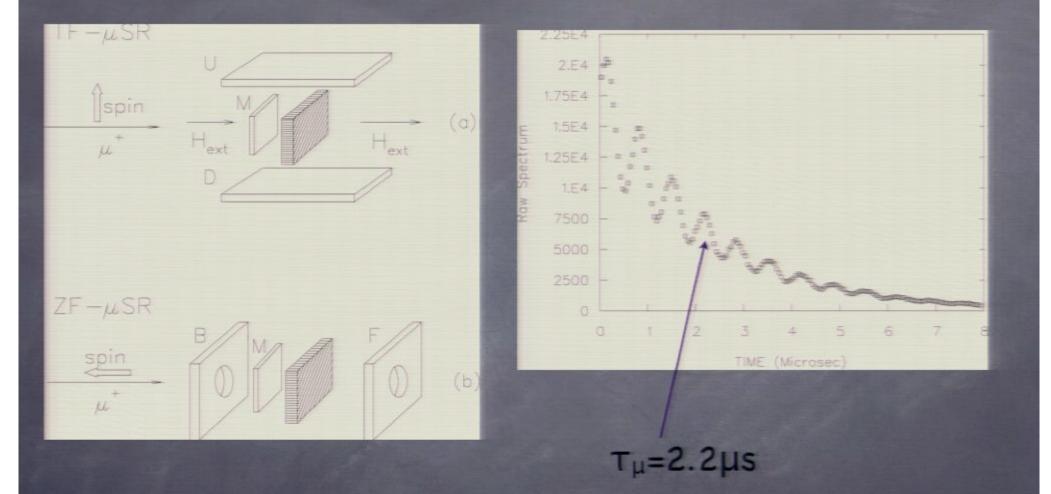


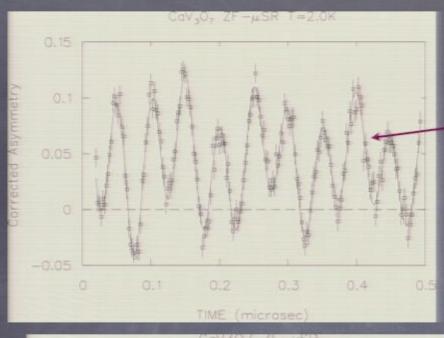
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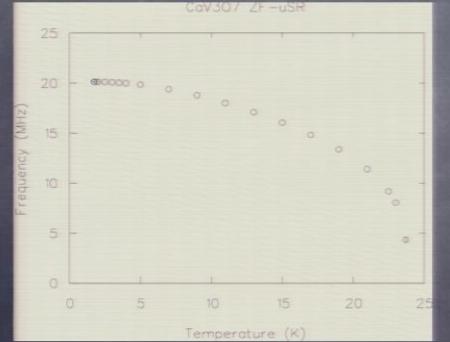


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### Experimental Geometry

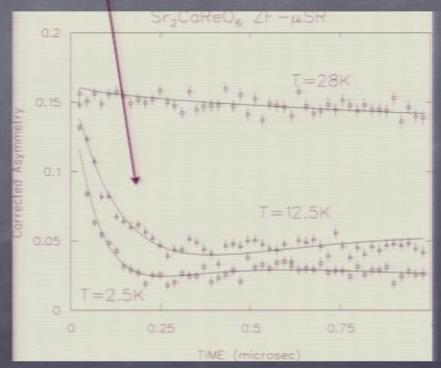






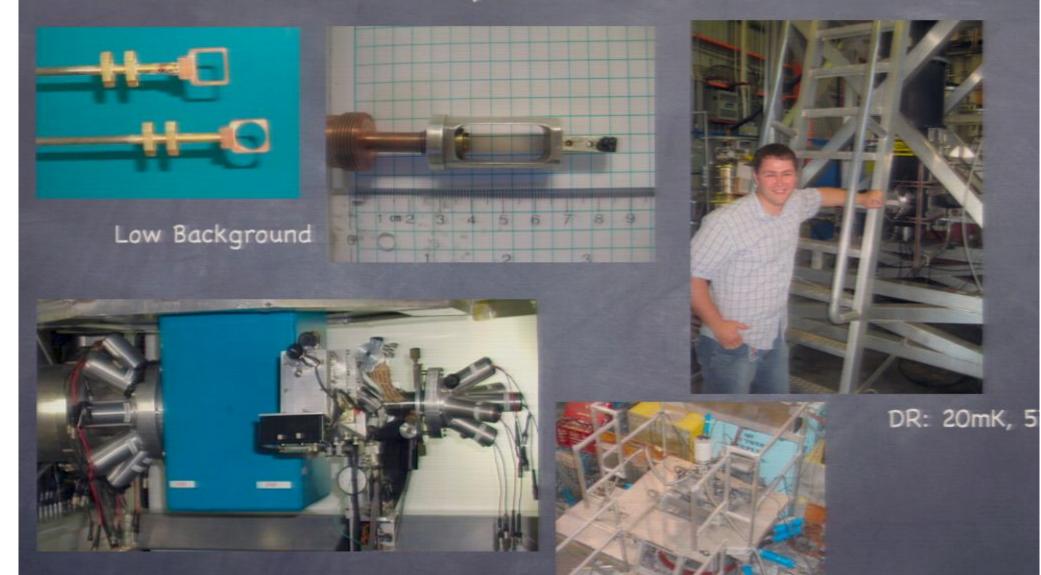
### Zero Field ZF-µSR

Ordered state, v∝ Moment ∝√I<sub>B</sub>
 Random frozen spins give characteristic relaxation.



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### Capabilities

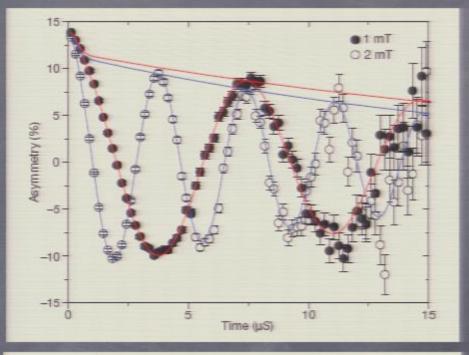


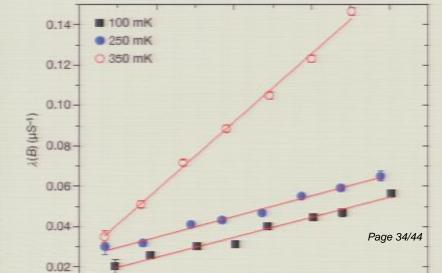
HiTime: 7.5T, 1GHz, 2K Pirsa: 11040095 High Pressure: 22kbar, 2.5K,

### Muon Spin Rotation Experiment

- Transverse field relaxation rate experiment.
- in Dilution refrigerator.
- measured relaxation rate of long-lived precession signal, fit the field dependence of relaxation rate to monopole model, extracted microscopic parameters.
- Second Wien effect

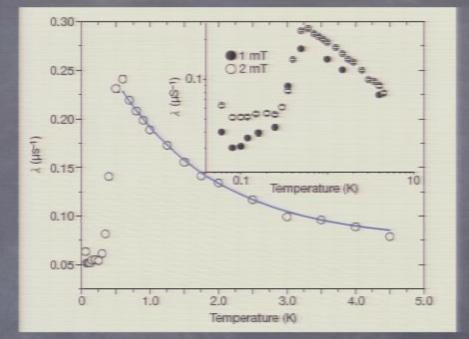
   (Onsager) for increase in
   dissociation of electrolyte in
   applied field applied to
   magnetic system.

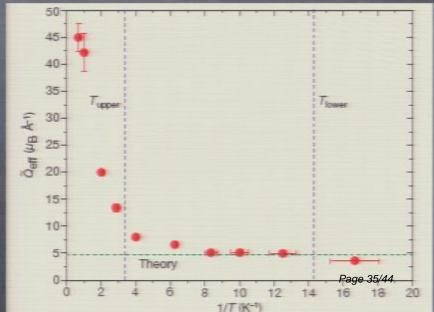




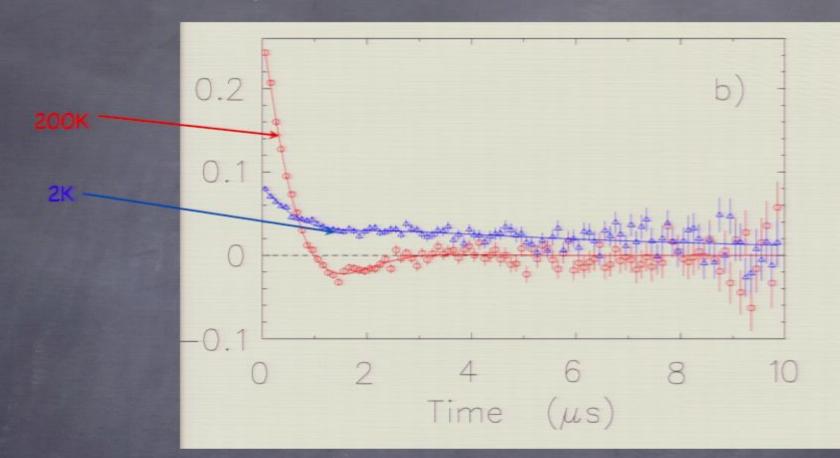
#### Muon Spin Rotation Experiment

- Low temperature data between 75mK and 300mK fit to find monopole "charge".
- Problem with analysis: 1/T<sub>2</sub> relaxation rate taken to be proportional to field fluctuation rate v. In fact, in transverse field, the inverse is the case.
- T<sub>2</sub> has additional sources than fluctuations (T<sub>1</sub>), such as field inhomogeneity (static broadening).



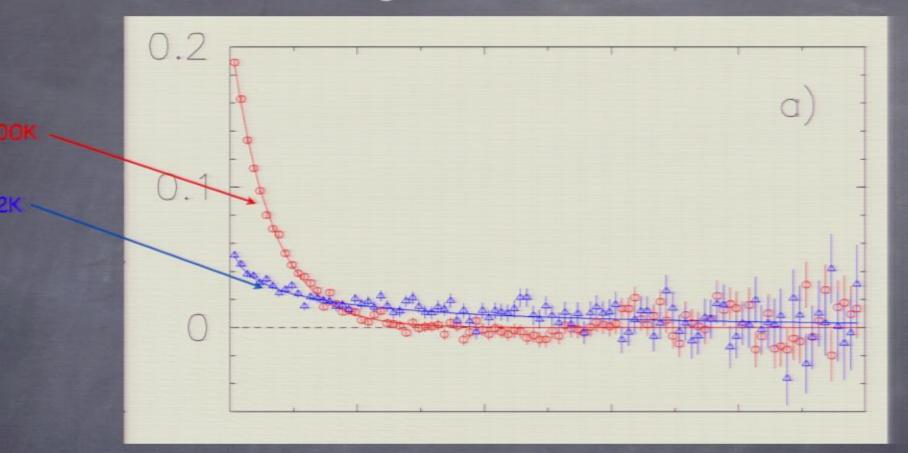


### Dy2Ti2O7 Transverse Field µSR



- High (200K) temperature, precession signal rapidly damped by fluctuating Dy<sup>3+</sup> moments.
- Low (2K) temperature, no precession in applied field as internal
   quasi-static fields much greater in magnitude. Some T<sub>1</sub> visible in t<sup>Page 36/44</sup>

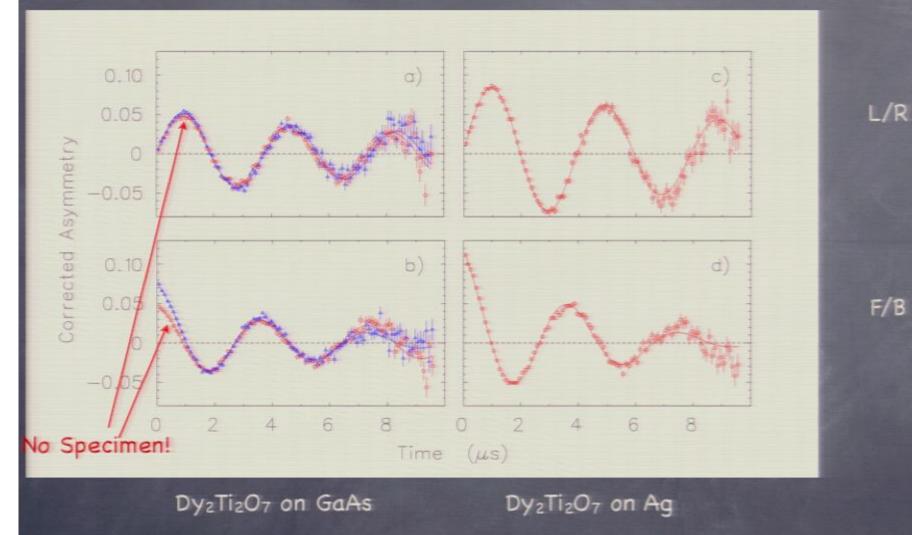
### Dy2Ti2O7 Longitudinal Field µSR



High (200K) temperature: full signal relaxes due to T<sub>1</sub>.

Low (2K) temperature: 1/3 tail relaxes slowly from T<sub>1</sub>.

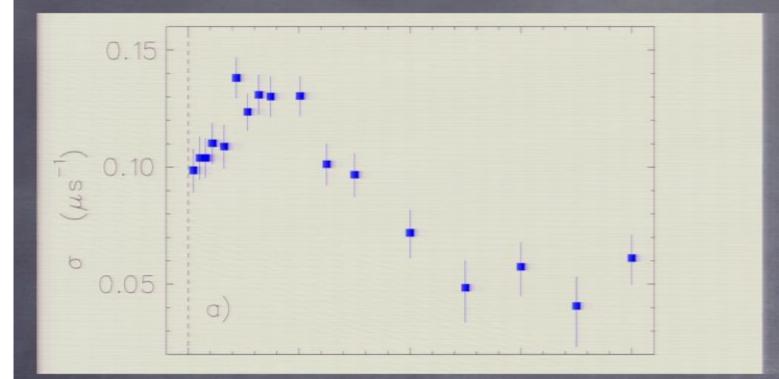
### Origin of TF-µSR Signal



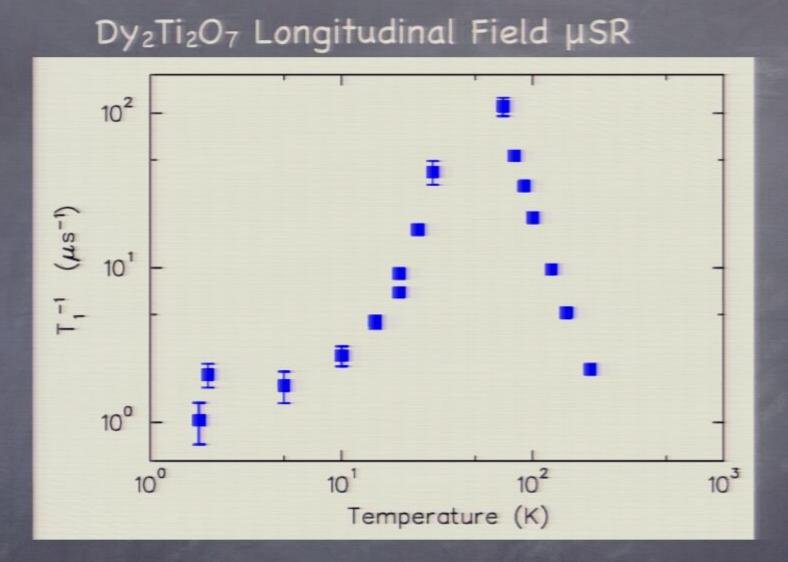
Precession signal from cryostat tails, silver mounting plate, NOT Dy2Ti2O7

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### Origin of TF-µSR Signal



- TF relaxation rate of Ag/cryostat tail signal qualitatively similar to results of Bramwell et al.
- Origin likely stray field from Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> sample magnetization.

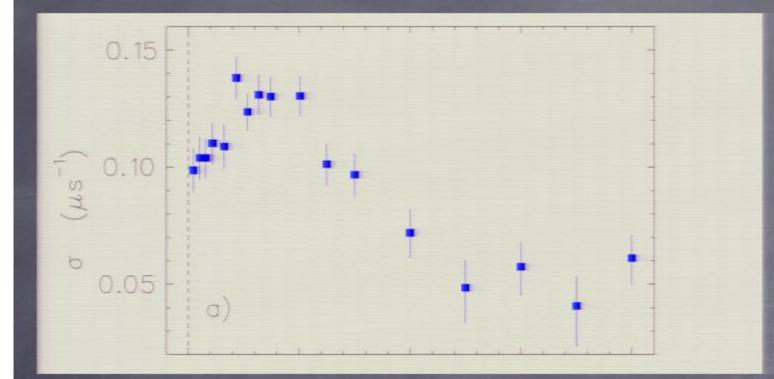


Above 70K full amplitude, below 70K, 1/3 only.

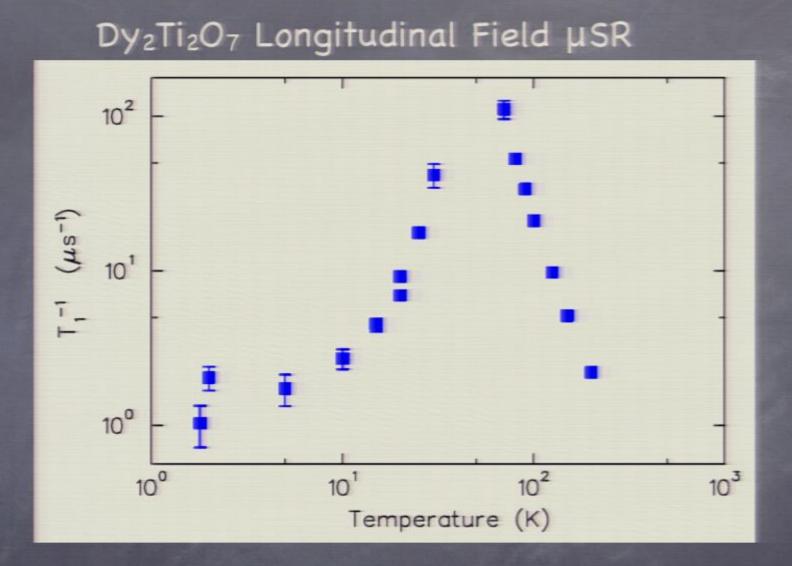
Maximum 1/T<sub>1</sub> where moments slow, crystal field levels depopulated. Page 40/44

a Tominimum - slow dynamics at low temperature

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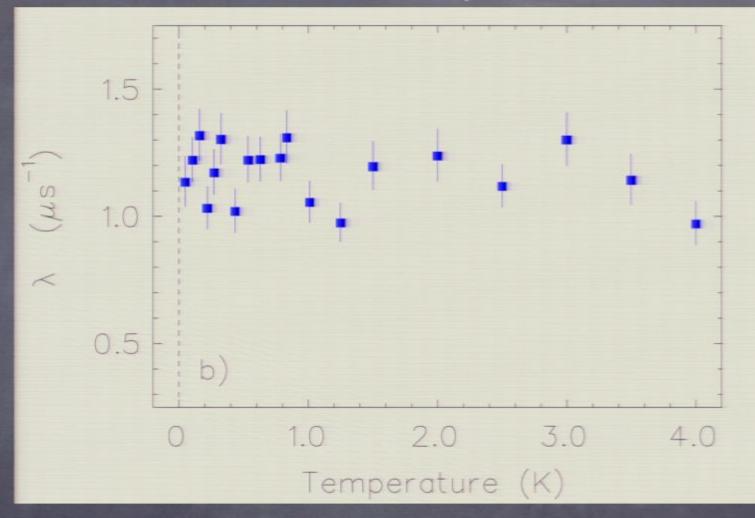


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### Dy2Ti2O7 LF-µSR in Spin Ice State



- Temperature-independent T<sub>1</sub> relaxation at low temperatures.
- Not activated as anticipated for monopoles.

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Large compared to expected for large energy barrier for single spin

### Conclusions

Report of monopole observation via TF-µSR highly flawed.

- Signal originated in sample holder.
- Incorrect analysis of relaxation rate in terms of fluctuation rate (should be 1/v)
- True T<sub>1</sub> relaxation rate is temperature-independent at low temperatures, origin unclear but requires additional dynamics beyond spin ice model.
- Thanks for your attention!