Title: New Neutron Scattering Results on the Enigmatic Ground State of the Pyrochlore Magnet Tb2Ti2O7

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Abstract: The ground state of the candidate spin liquid pyrochlore magnet Tb2Ti2O7 (TTO) has been long debated. Despite theoretical expectations of magnetic order below ~1K based on classical Ising-like Tb3+ spins, earlier muSR and neutron scattering experiments showed no long range order down to 50mK. This motivated two theoretical scenarios to account for the apparently disordered ground state: a quantum spin ice scenario and a non-magnetic singlet ground state. I will discuss new neutron scattering measurements on TTO that show short range spin correlations developing below ~ 0.5 K with a (Â½, Â½, Â½) ordering wavevector, and a concomitant opening of a spin gap across most of the Brillouin zone. Our measurements also refine the crystal field ground state for Tb3+ in TTO and in its sister, â€œsoftâ€• spin ice compound Tb2Sn2O7.

Outline

- $Tb_2Ti_2O_7$ – intro
- Experimental findings
- Two possible scenarios for ground state: quantum spin ice and non-magnetic singlet
- Neutron scattering measurements
- Sample variability
- Conclusion

$Tb_2Ti_2O_7$ - the most puzzling rare earth titanate?

$R_2Ti_2O_7$

R^{3+}

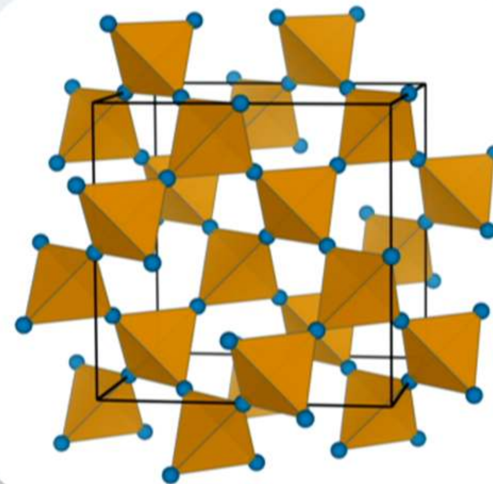
Ho/Dy
spin ice/magnetic monopoles/coulomb phase

Yb
quantum spin ice/coulomb phase

Er
long-range order/order-by-disorder/quantum critical point

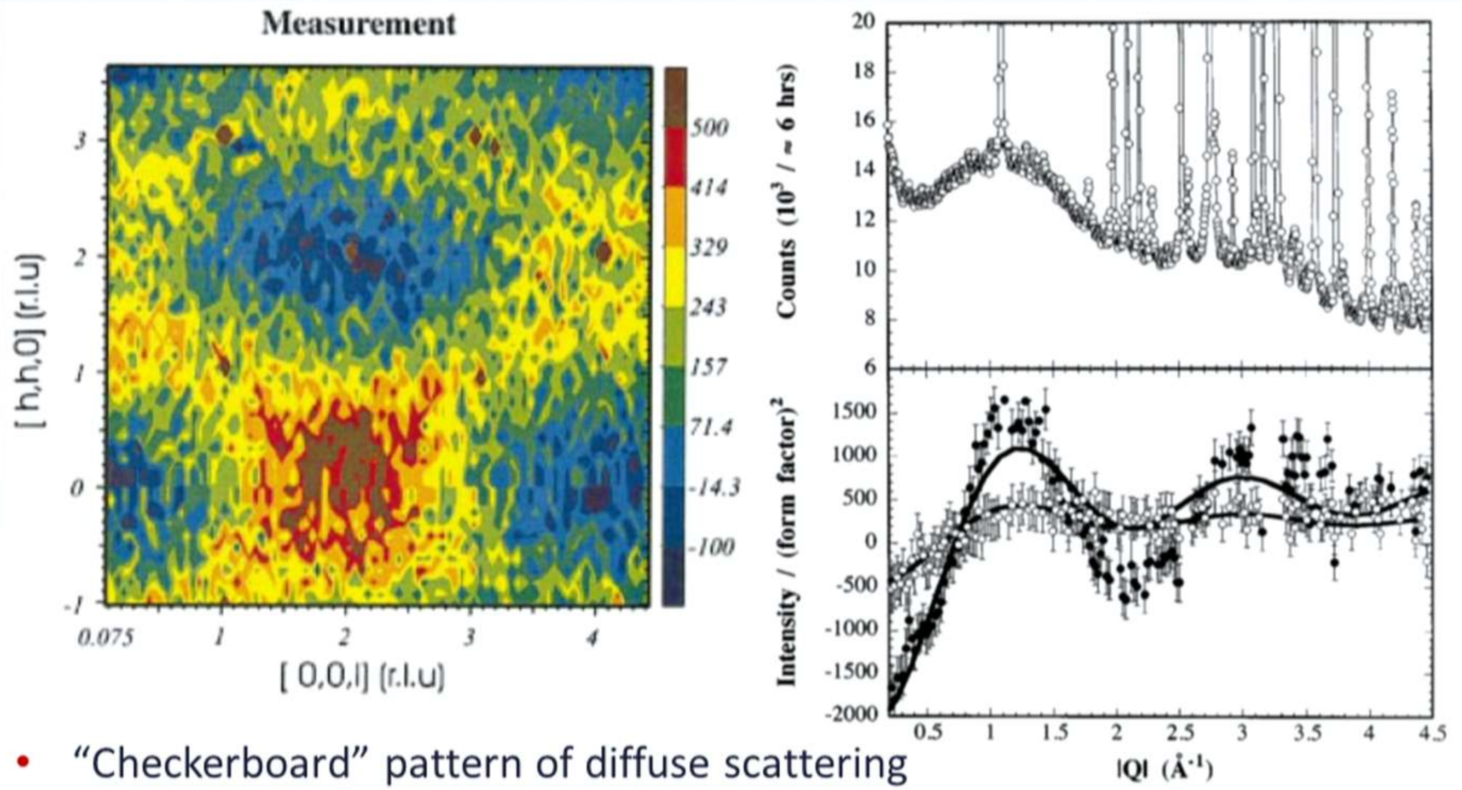
Gd
partial ordering

Tb
spin liquid/quantum spin ice/ non-magnetic singlet?



- Ising anisotropy constraining moments to point along <111>
- Antiferromagnetic interactions Θ_{cw}~-14K
- No magnetic long-range order down to 50mK

Experimental findings

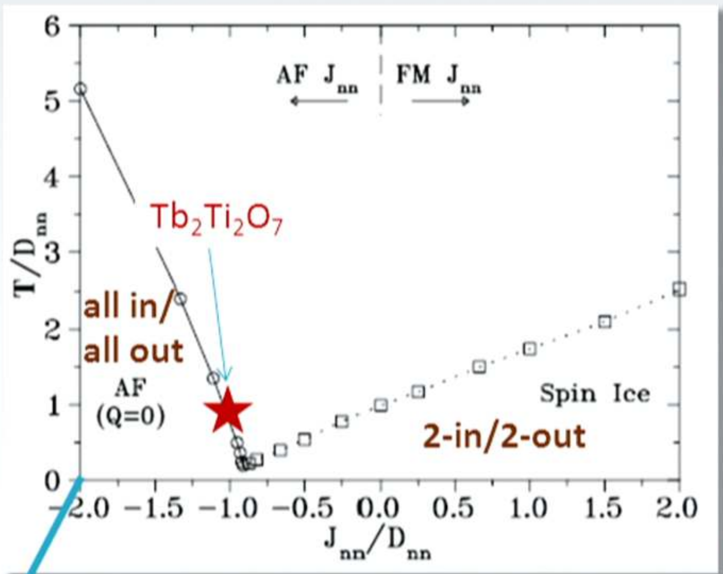


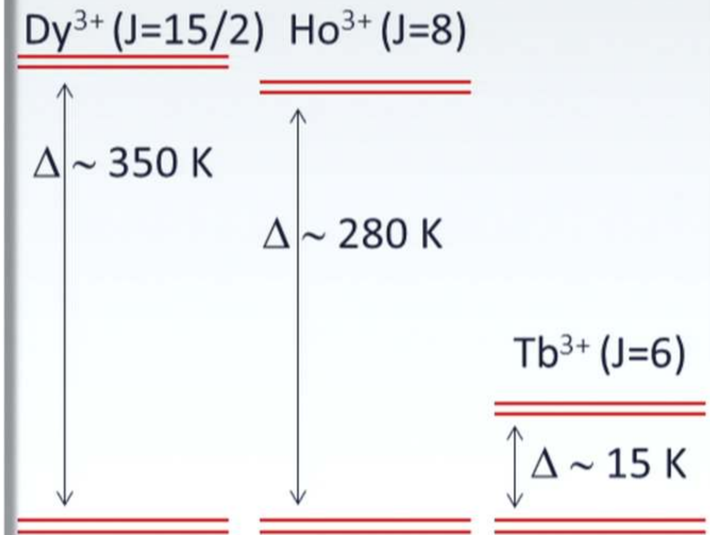
- “Checkerboard” pattern of diffuse scattering implies correlations over single tetrahedron

Gardner et al., Phys. Rev. Lett 82, 1012 (1999); Phys. Rev. B 64, 224416 (2001)

Den Hertog and Gingras, Phys. Rev. Lett. 84, 3430 (2000); Molavian et al., Phys. Rev. Lett. 98, 157204 (2007)

Two scenarios: 1 – quantum spin ice





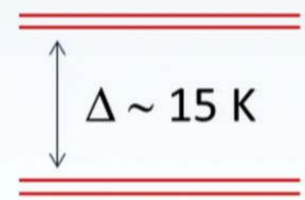
- Dipolar spin ice model works well if GS doublet is well isolated from 1st excited doublet

$1/\Delta$
- Introduce quantum fluctuations as 3rd axis (scaling as $1/\Delta$)
- Virtual quantum fluctuations of the ions between GS and excited doublet states create more complex interactions

Two scenarios: 2 - non-magnetic singlet

- A possible cubic-tetragonal distortion of Tb^{3+} CF environment splits the GS doublet into two non-magnetic singlets
- X-ray measurements show evidence of structural fluctuations as a possible precursor to a lower T transition

Tb^{3+} (non-Kramers)



PHYSICAL REVIEW B **84**, 184409 (2011)

Tetragonal distortion yielding a two-singlet spin liquid in pyrochlore $Tb_2Ti_2O_7$

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(Received 17 May 2011; revised manuscript received 7 October 2011; published 10 November 2011)

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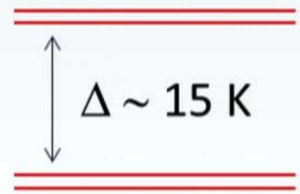

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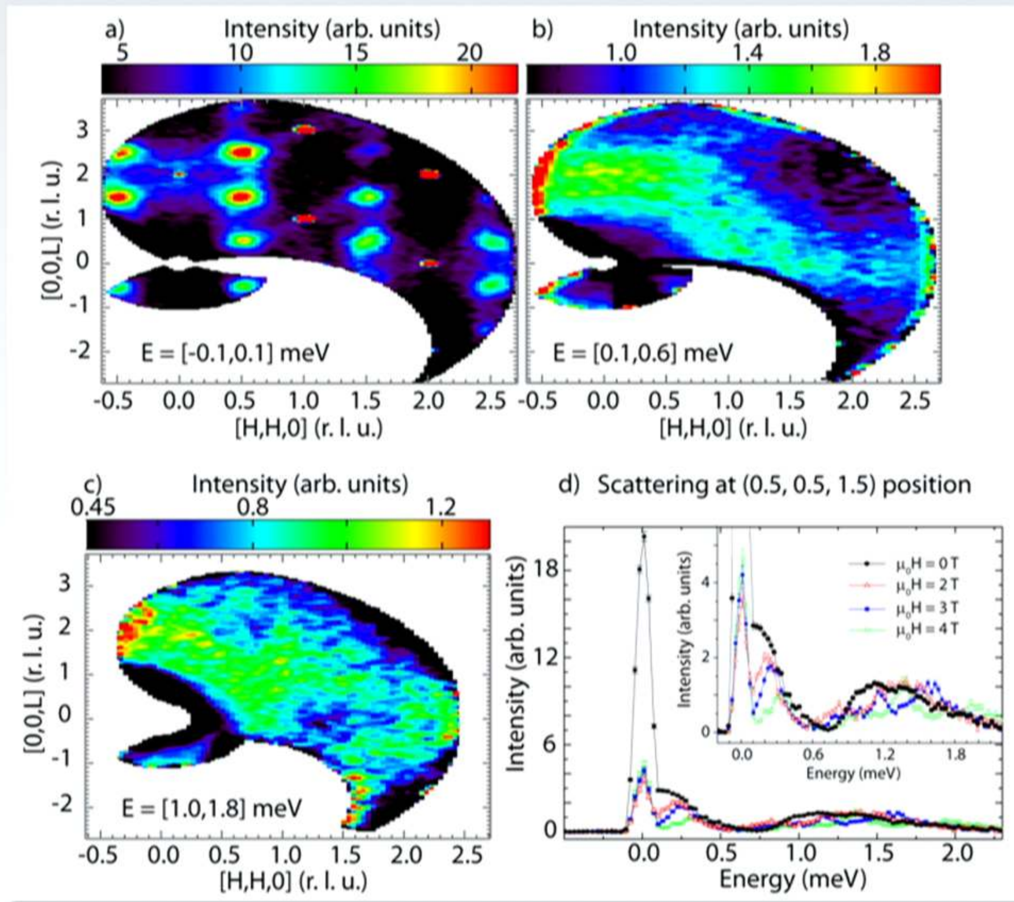
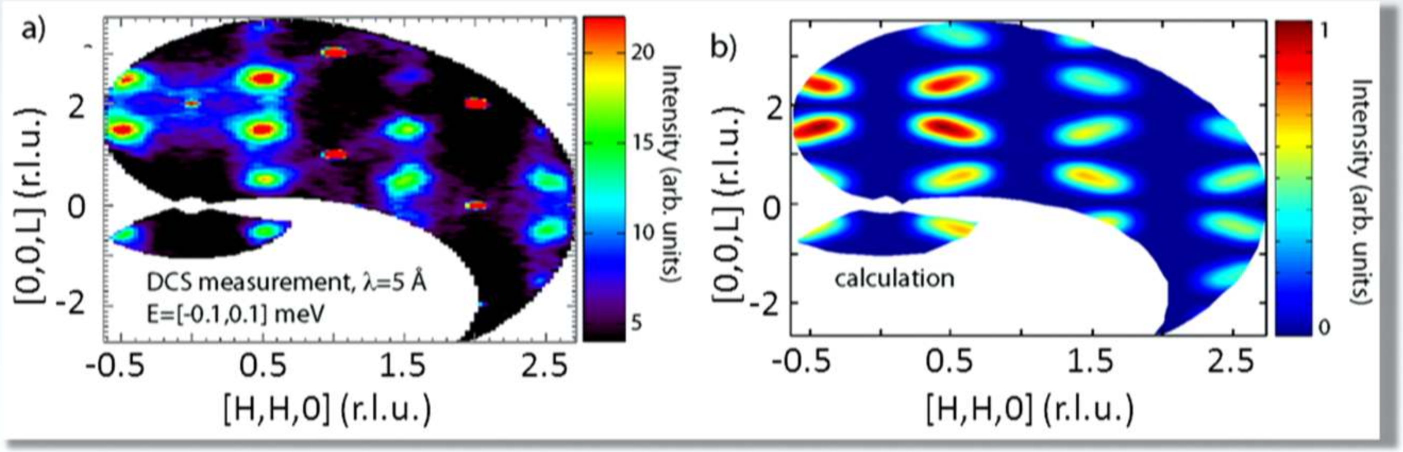
Scattering regimes at 70 mK (DCS)

- Elastic: (±0.1 meV) diffuse scattering at $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ positions
- Inelastic: low E (0.06 - 0.6 meV) diffuse "checkerboard"
- Inelastic: higher E (1.0 - 1.8 meV) transitions to lowest excited CF doublet



Model of the SRO elastic scattering at $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$

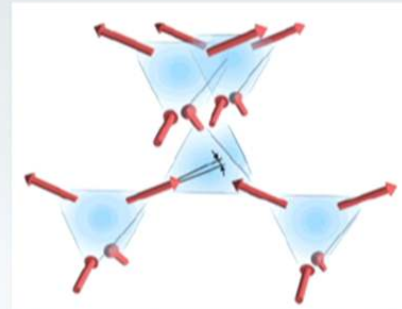


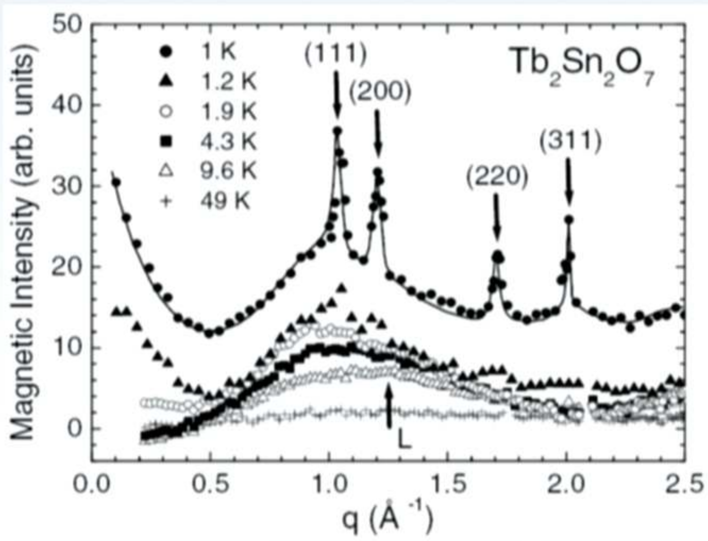
- Calculation of S(Q) based on
  a) observed phase shift of π between adjacent unit cells
  b) width of scattering (implying isotropic correlations over roughly two conventional unit cells)

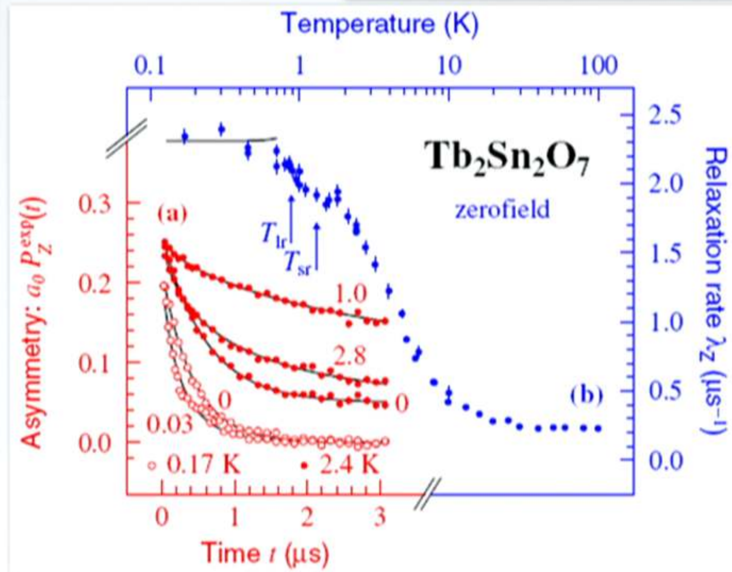
- Best fit result for intensities of 9 $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$-like Bragg positions in a 2x2x2 supercell containing 128 spins

Short aside – $Tb_2Sn_2O_7$

- "soft" ordered spin ice state below Tc = 0.87K

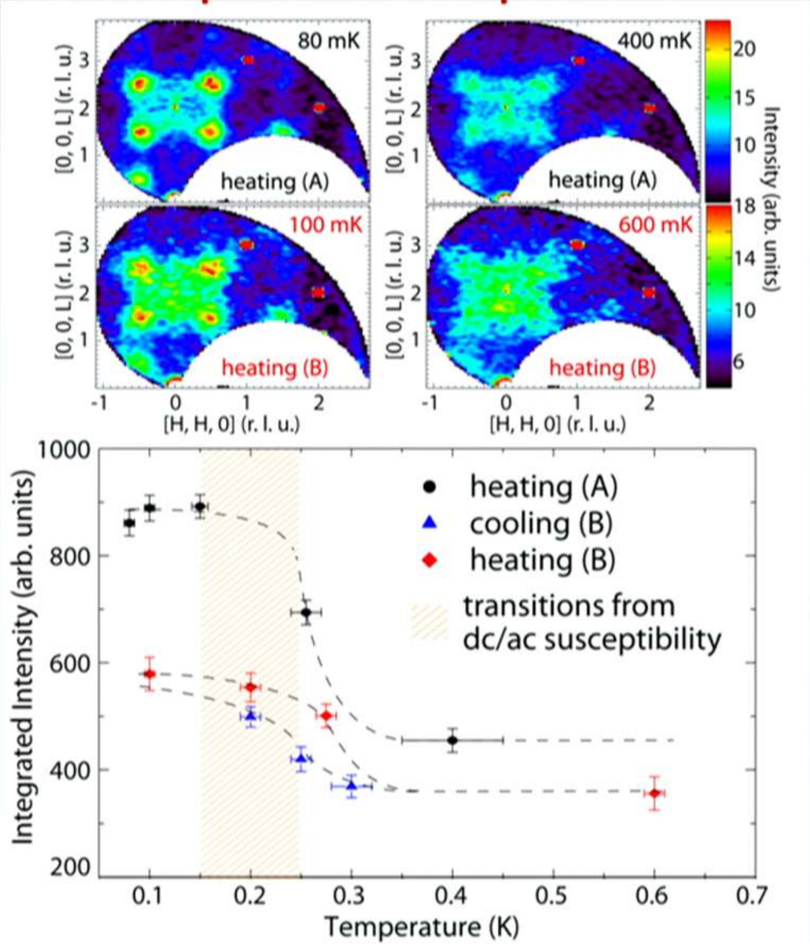






Mirebeau et al., Phys. Rev. Lett. 94, 246402 (2005); Petit et al., Phys. Rev. B 85, 054428 (2012); Dalmas de Reotier et a., Phys. Rev. Lett. 96, 127202 (2006)

Temperature dependence of $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$



- Integrated elastic scattering intensity disappears as T increases above ~275 mK
- History dependence (glassy state)

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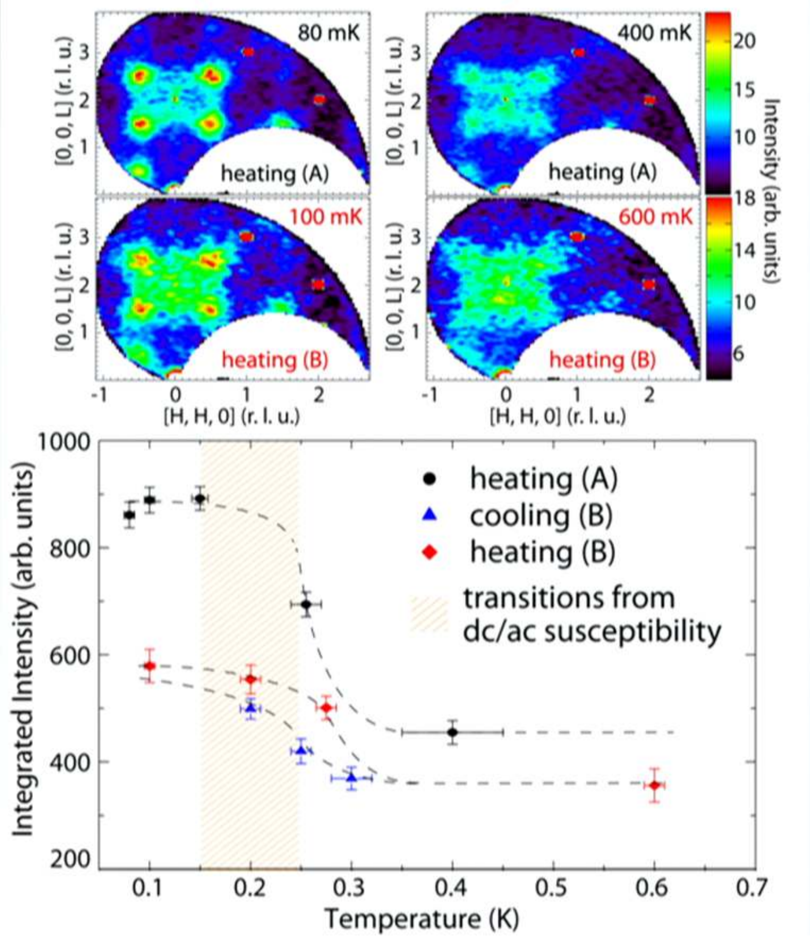

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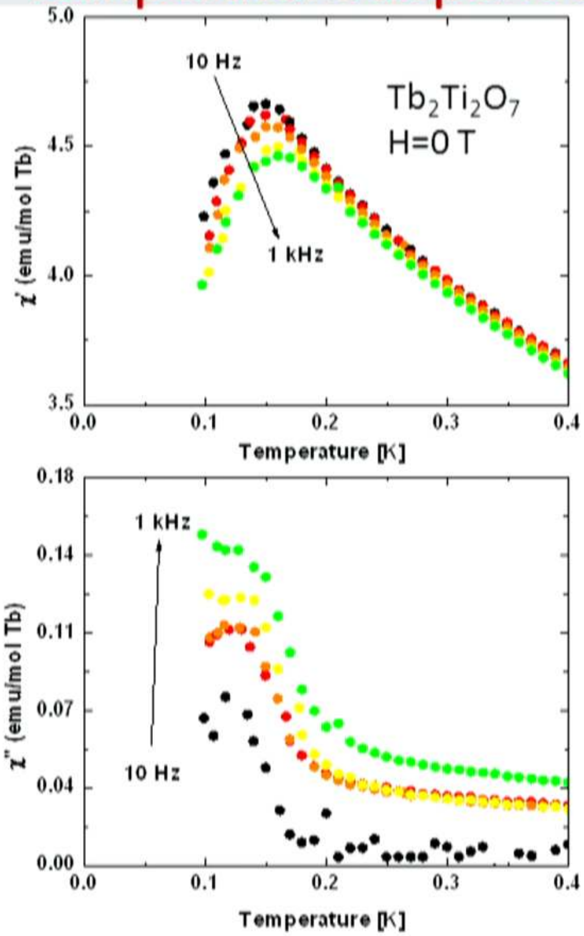
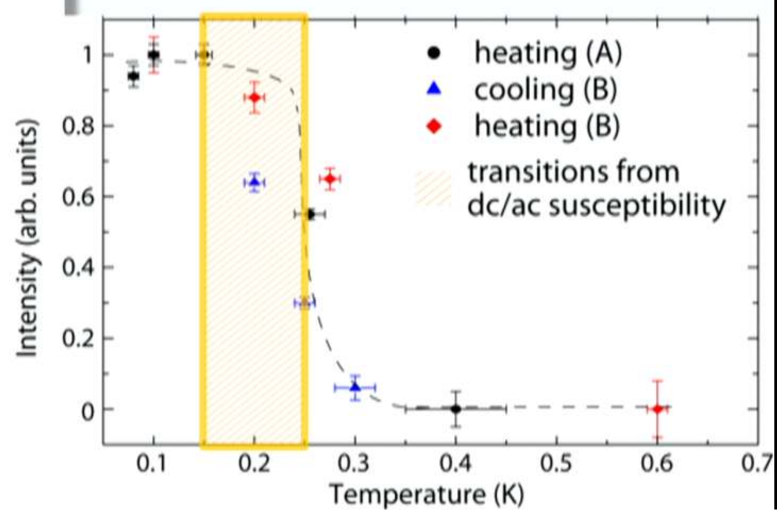
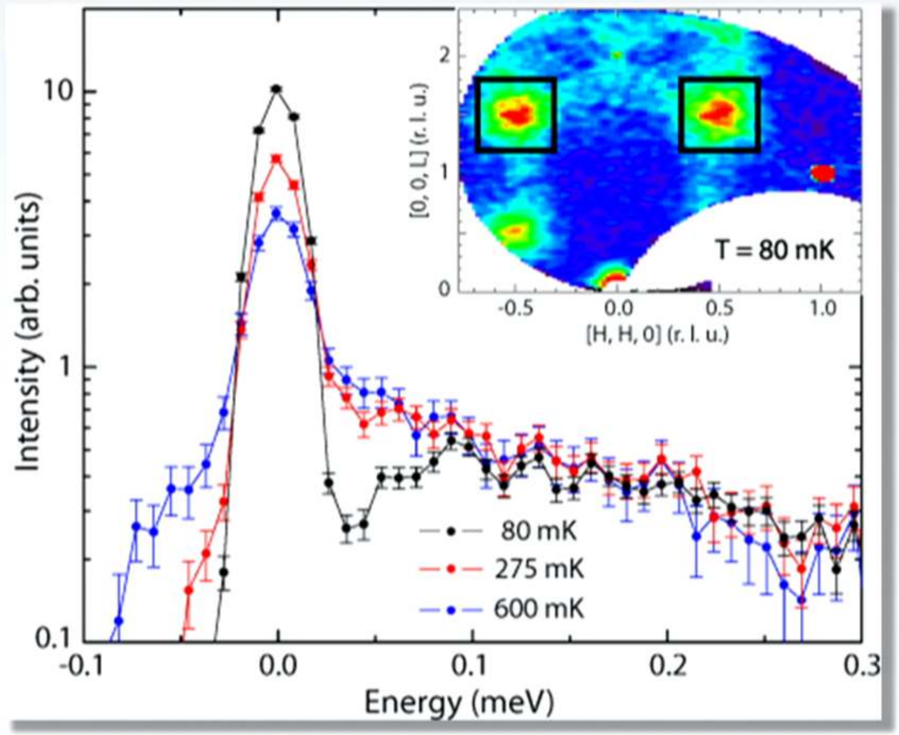


- Integrated elastic scattering intensity disappears as T increases above ~275 mK
- History and frequency dependence (glassy state)



Spin dynamics – low energy inelastic scattering

- Existence of peaks correlates with spin gap of ~ 0.06 - 0.08 meV in excitation spectrum
- Gap opens up below ~ 300 mK, where $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ elastic peaks appear and spectral weight is pushed into elastic line



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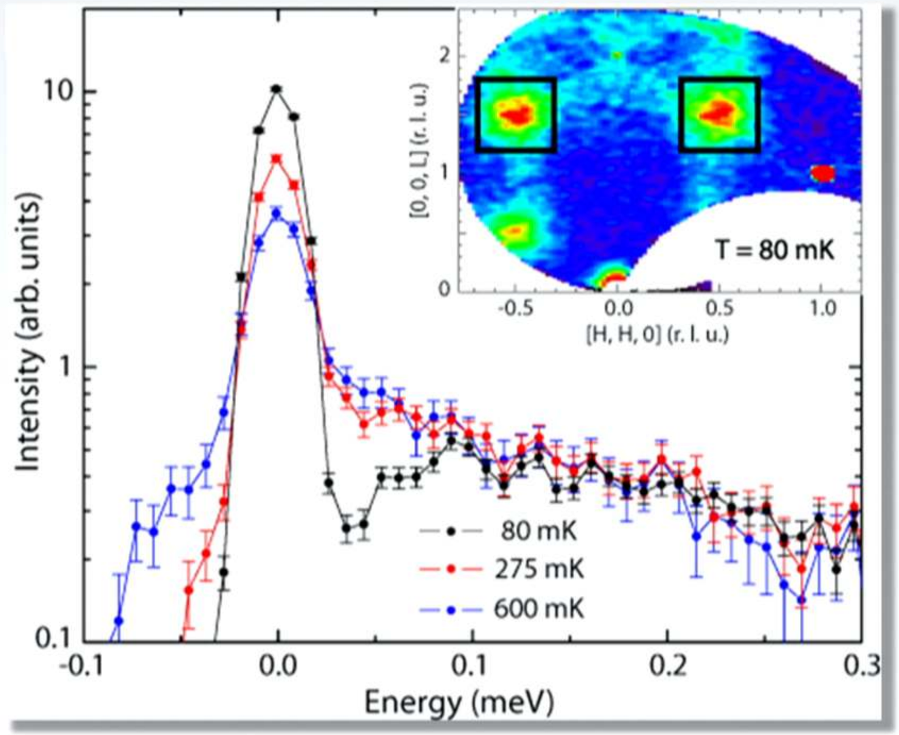
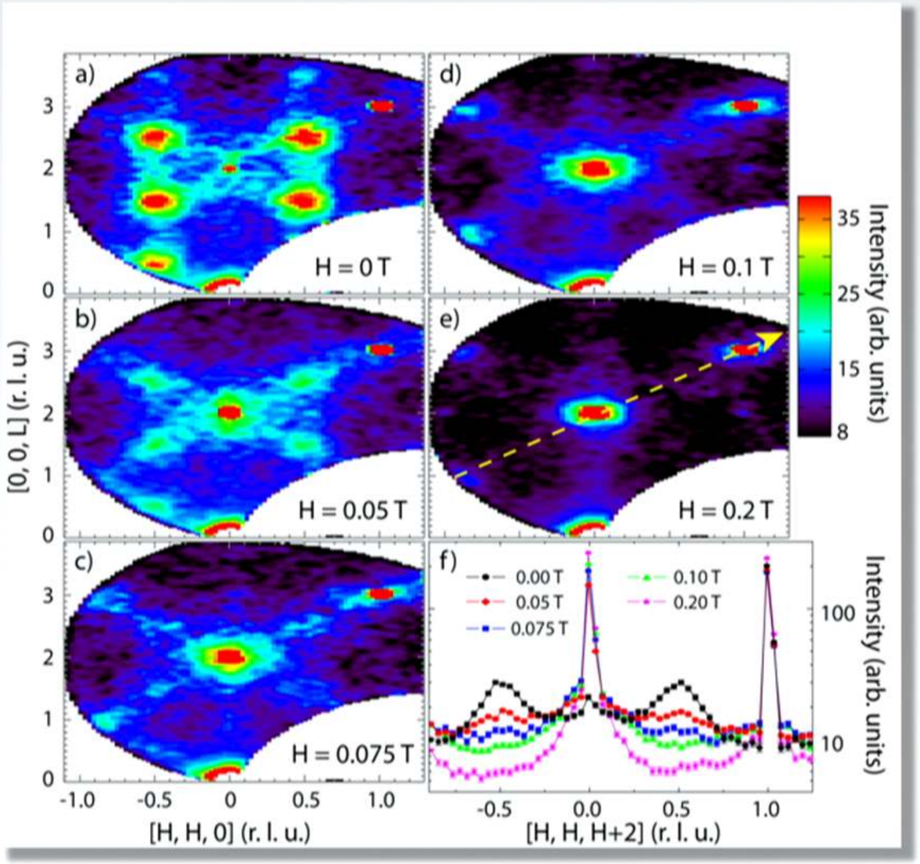


Field dependence at 80 mK (H | | [1-10])



- Small field of ~ 0.075 T destroys SRO $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ state
- Signs of anisotropic exchange: additional scattering features

Conclusions

- At 70mK, SR AF spin ice correlations extending over roughly two conventional unit cells
- Elastic scattering described based on an ordered local 2-in 2-out spin ice structure with spin canting angle of ~12°
- SRO correlations disappear upon heating to ~300mK
- Correlations appear to be glassy in nature
- Elastic scattering is separated by a gap of ~0.06-0.08 meV from low-lying inelastic magnetic scattering over most of BZ
- Gap appears to go soft at the $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ positions
- Magnetic scattering is destroyed by small magnetic fields H ~ 0.075 T (H || [110])
- Sample variability/non-stoichiometry an issue to investigate