

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 ($\hat{A}^{1/2}, \hat{A}^{1/2}, \hat{A}^{1/2}$) 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, "soft" spin ice compound Tb₂Sn₂O₇.

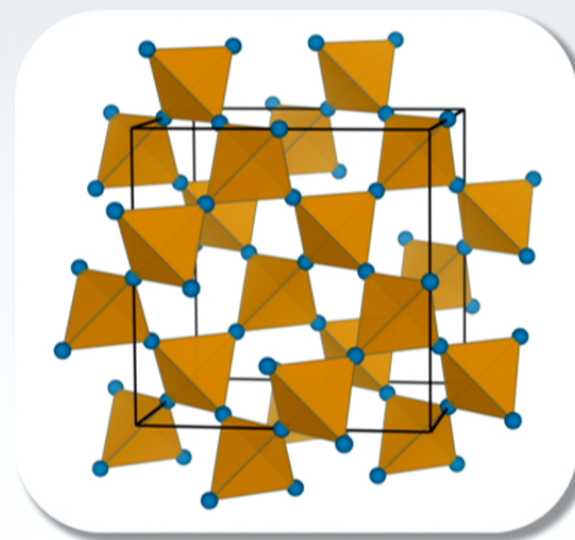
Outline

- $\text{Tb}_2\text{Ti}_2\text{O}_7$ – intro
- Experimental findings
- Two possible scenarios for ground state:
quantum spin ice and non-magnetic singlet
- Neutron scattering measurements
- Sample variability
- Conclusion

Tb₂Ti₂O₇ - the most puzzling rare earth titanate?

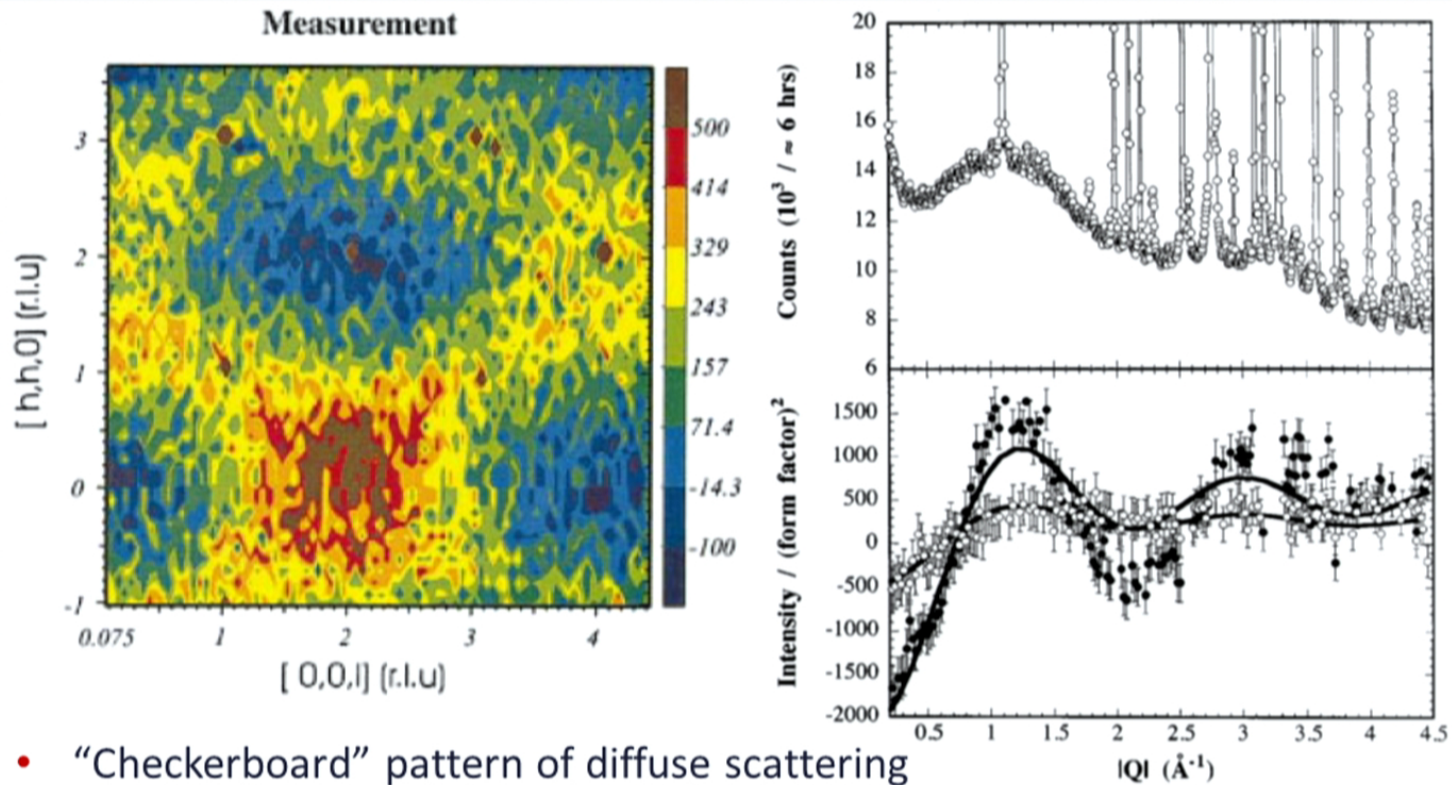


R^{3+}	Ho/Dy spin ice/magnetic monopoles/coulomb phase
	Yb quantum spin ice/coulomb phase
	Er long-range order/order-by- disorder/quantum critical point
	Gd partial ordering
	Tb spin liquid/quantum spin ice/ non-magnetic singlet?



- Ising anisotropy constraining moments to point along $\langle 111 \rangle$
- Antiferromagnetic interactions
 $\Theta_{CW} \sim -14K$
- No magnetic long-range order down to 50mK

Experimental findings

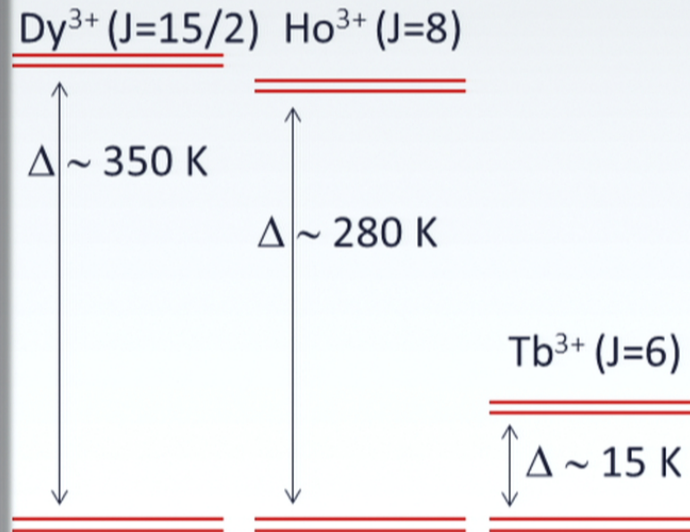
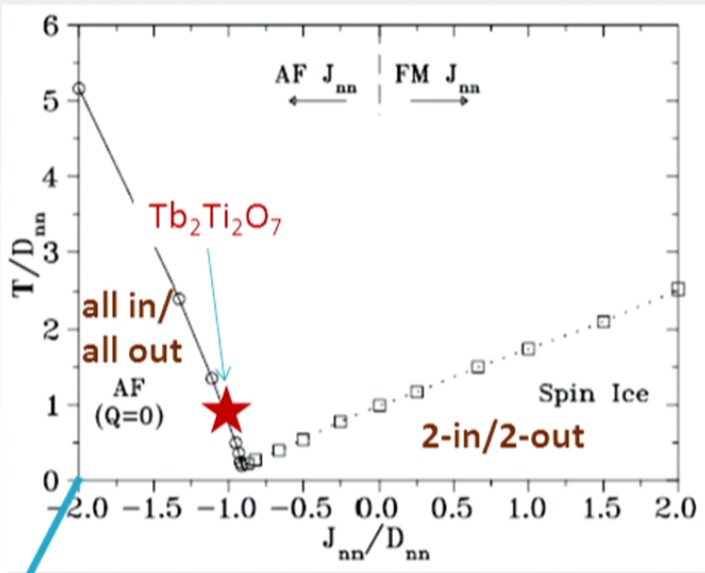


- “Checkerboard” pattern of diffuse scattering implies correlations over single tetrahedron

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

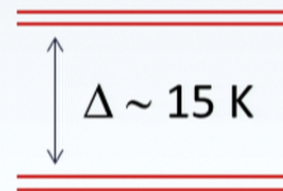


- Dipolar spin ice model works well if GS doublet is well isolated from 1st excited doublet
- 1/Δ • Introduce quantum fluctuations as 3rd axis (scaling as 1/Δ)
- Virtual quantum fluctuations of the ions between GS and excited doublet states create more complex interactions

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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Isabelle Mirebeau,* Arsène Gukasov, Sylvain Petit, and Julien Robert

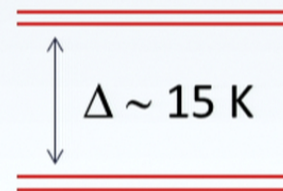
CEA, Centre de Saclay, DSM/IRAMIS/Laboratoire Léon Brillouin, F-91191 Gif-sur-Yvette, France

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

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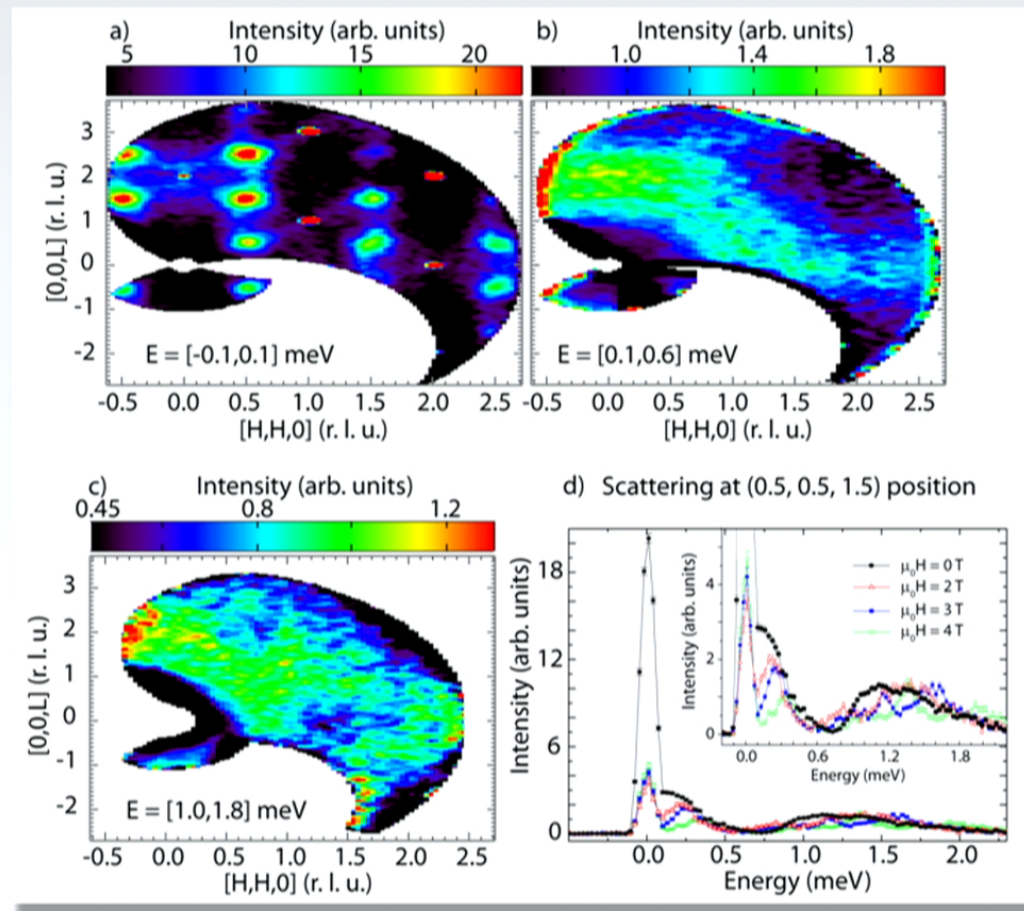
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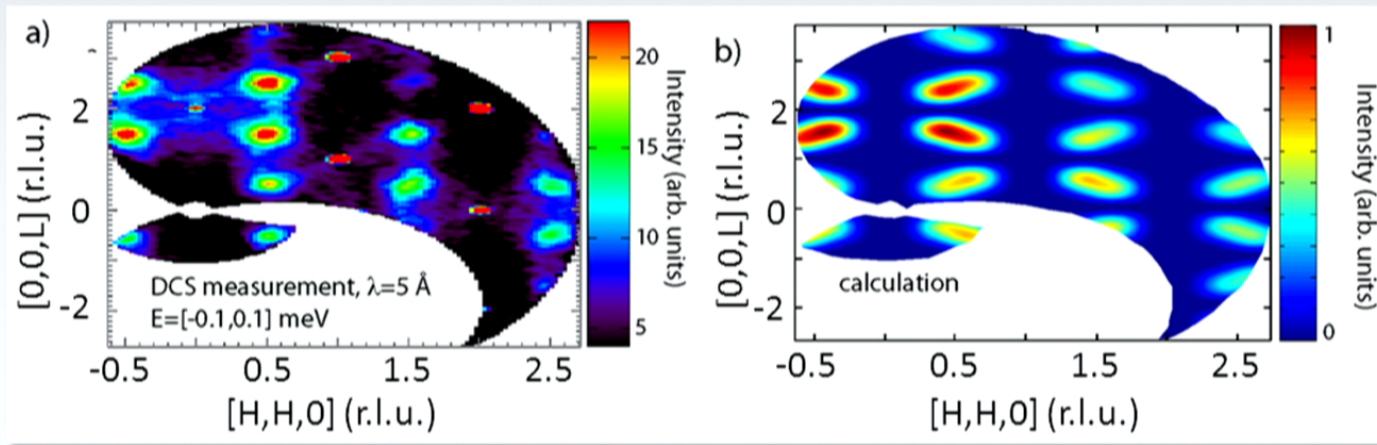
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Scattering regimes at 70 mK (DCS)

- 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



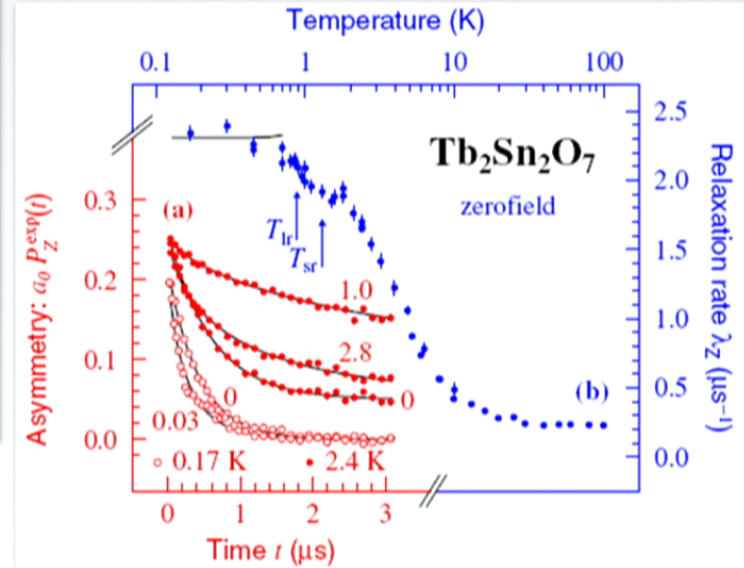
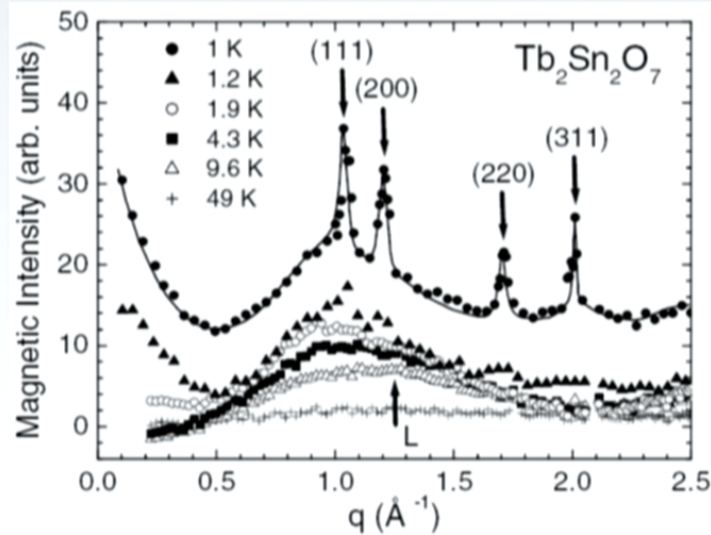
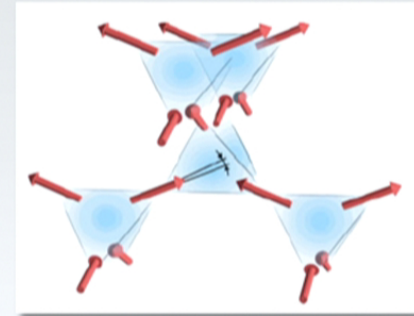
Model of the SRO elastic scattering at $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$



- 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 $2 \times 2 \times 2$ supercell containing 128 spins

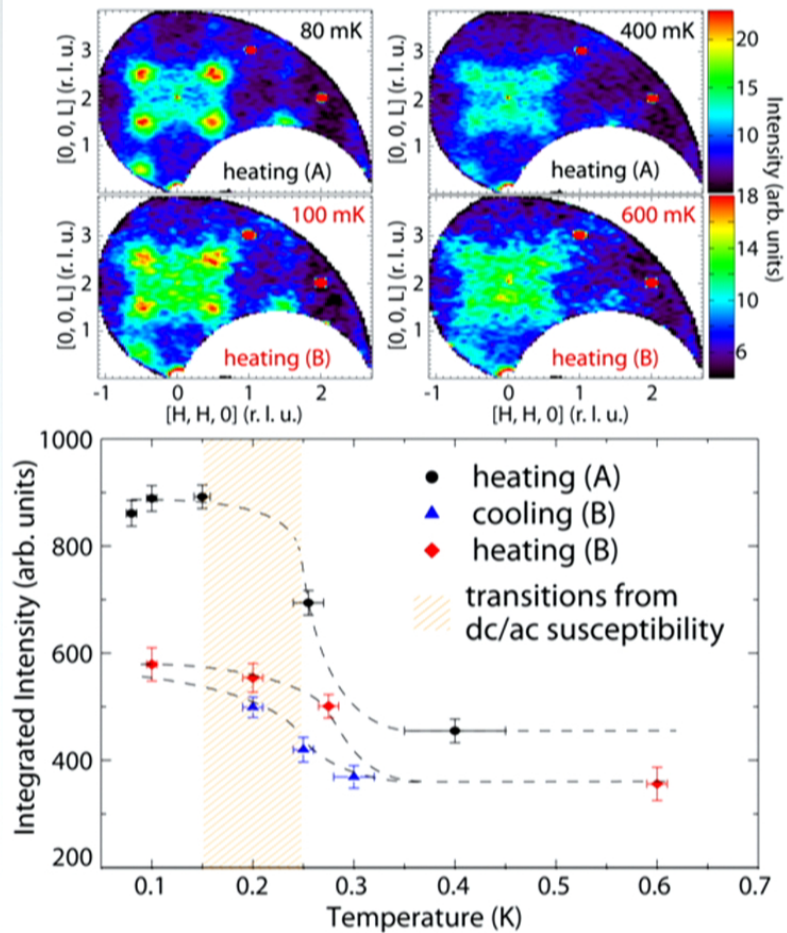
Short aside – $\text{Tb}_2\text{Sn}_2\text{O}_7$

- “soft” ordered spin ice state below $T_c = 0.87\text{K}$



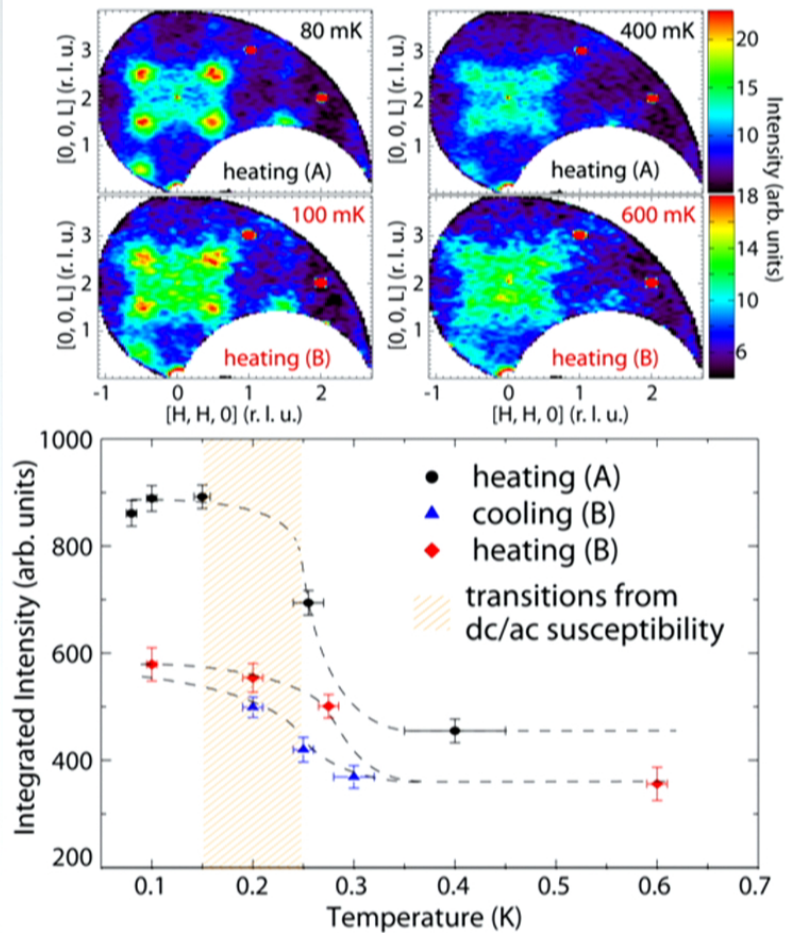
Mirebeau et al., Phys. Rev. Lett. 94, 246402 (2005); Petit et al., Phys. Rev. B 85, 054428 (2012); Dalmas de Reotier et al., Phys. Rev. Lett. 96, 127202 (2006)

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



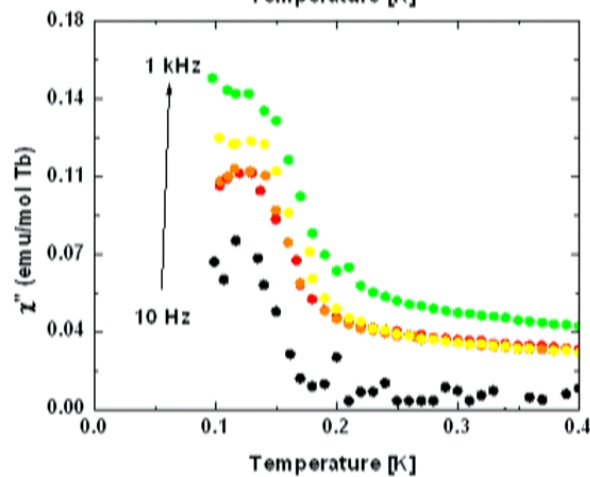
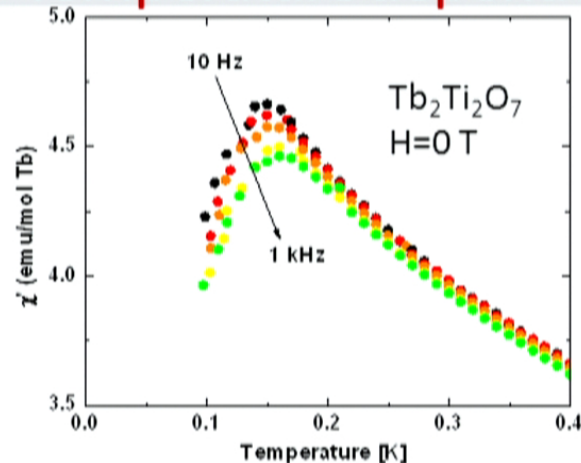
- Integrated elastic scattering intensity disappears as T increases above ~ 275 mK
- History dependence (glassy state)

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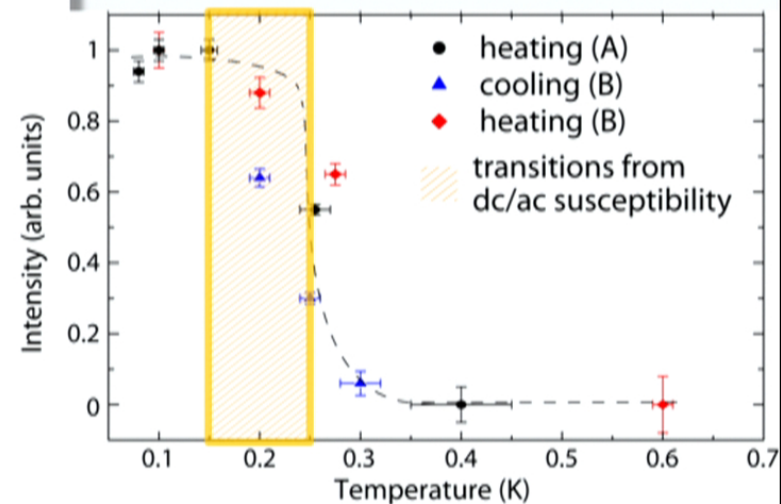


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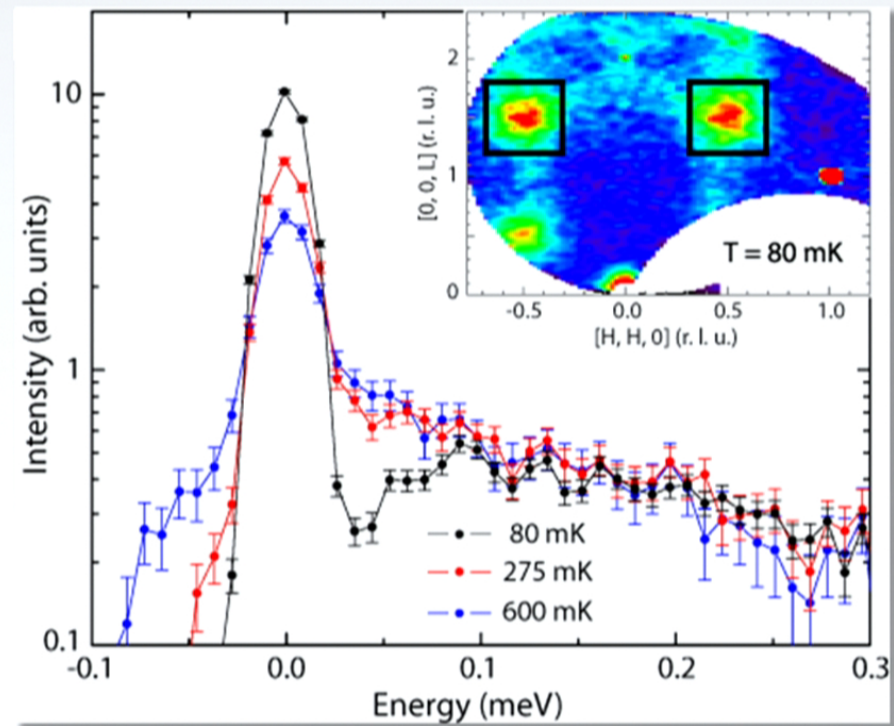


- Integrated elastic scattering intensity disappears as T increases above ~ 275 mK
- History and frequency dependence (glassy state)



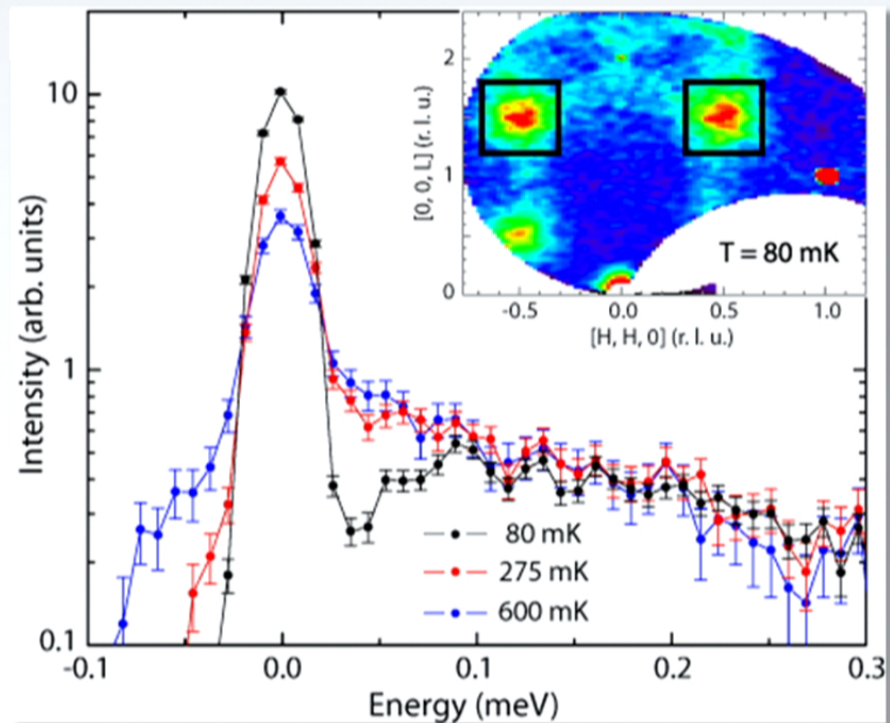
Spin dynamics – low energy inelastic scattering

- Existence of peaks correlates with spin gap of $\sim 0.06 - 0.08$ 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

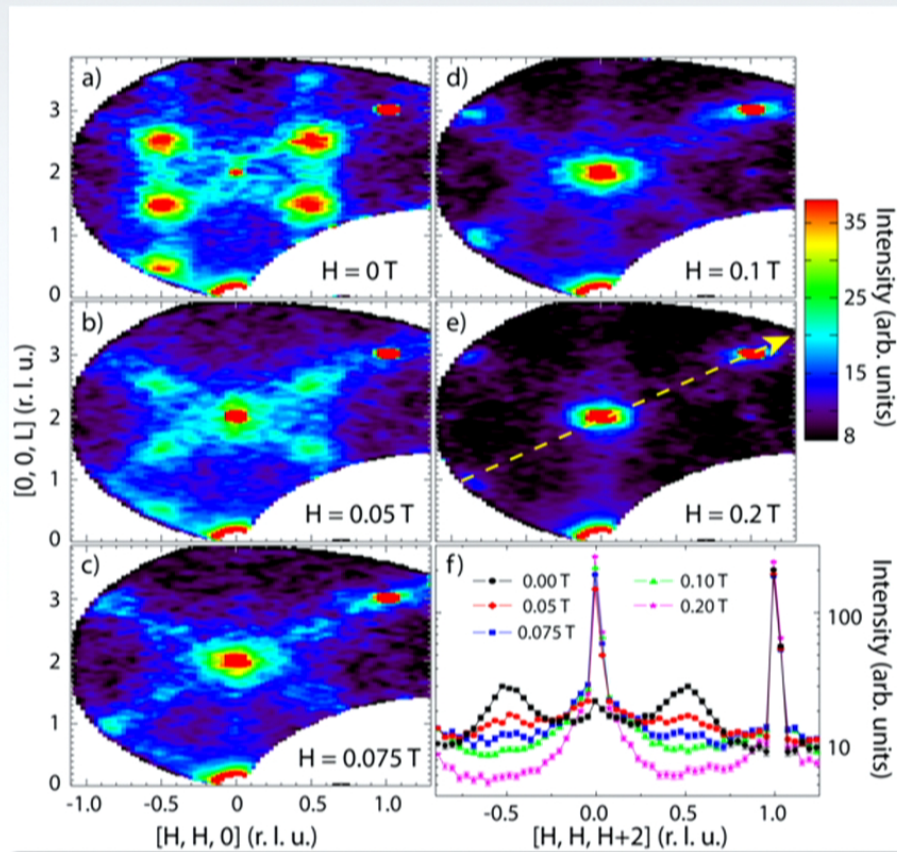


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Field dependence at 80 mK ($H \parallel [1-10]$)



- 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 $\sim 12^\circ$
- SRO correlations disappear upon heating to $\sim 300\text{mK}$
- Correlations appear to be glassy in nature
- Elastic scattering is separated by a gap of $\sim 0.06\text{-}0.08\text{ meV}$ 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 $H \sim 0.075\text{ T}$ ($H \parallel [110]$)
- Sample variability/non-stoichiometry an issue to investigate