Title: Specific heat and ac susceptibility measurements on the Spin Ice, $Dy_2Ti_2O_7$

Date: Feb 11, 2014 09:00 AM

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Abstract: Some time ago (1999), $Dy_2Ti_2O_7$, was shown to be a magnetic analog of water ice, and thus dubbed "spin ice". Recently, theories and experiments have developed the perspective of viewing excitations within the low temperature phase of this spin ice as monopoles. I will present early results of specific heat, ac susceptibility and magnetization measurements as well as my group's recent results on this system

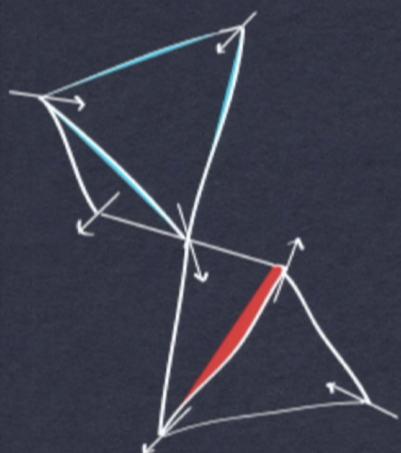
Specific heat and magnetization studies of the spin ice material, $Dy_2Ti_2O_7$

Jan Kycia

Halle Revell, Luke Yaraskavitch, David Pomaranski, Shuchao Meng, Jeff Mason, Jeff Quilliam
Department of Physics and Astronomy, University of Waterloo

Patrik Henelius
Department of Theoretical Physics,
Royal Institute of Technology, Stockholm, Sweden

Kate Ross, Hilary Noad, Hanna Dabkowska, Bruce Gaulin
McMaster University, Brockhouse Institute



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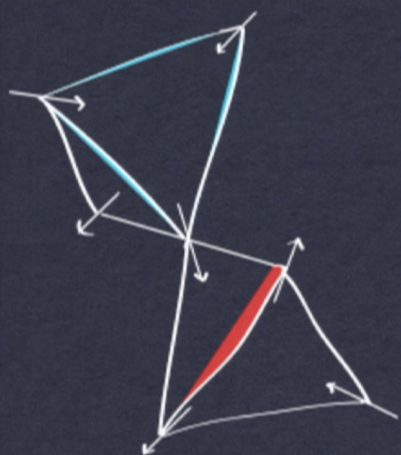

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M.P.A. Fisher (Colloquium)
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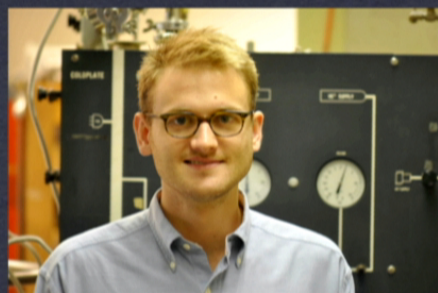
The Group at Waterloo

Halle Revell



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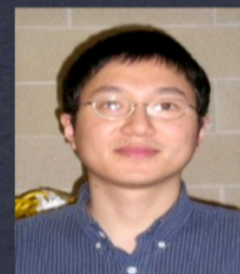

Shuchao Meng

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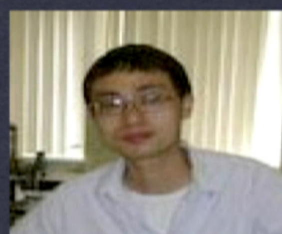


Shaoxiong Li

Chris Michelitis

Jeff Quilliam

Conventional Susceptometer

Advantage: Easy to put together and use.

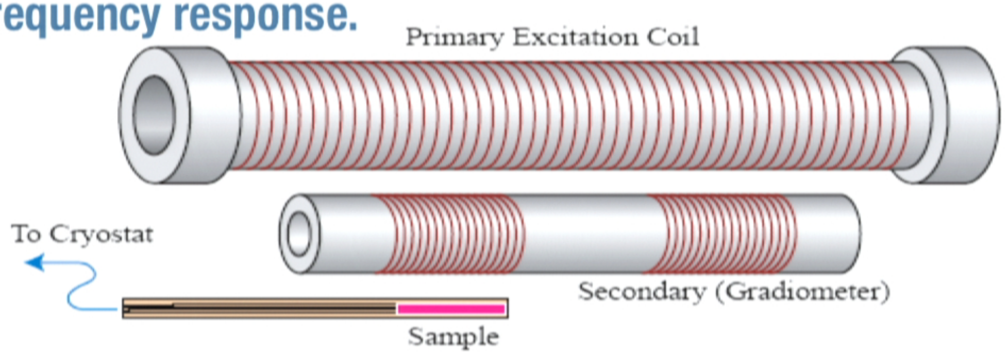
Disadvantages:

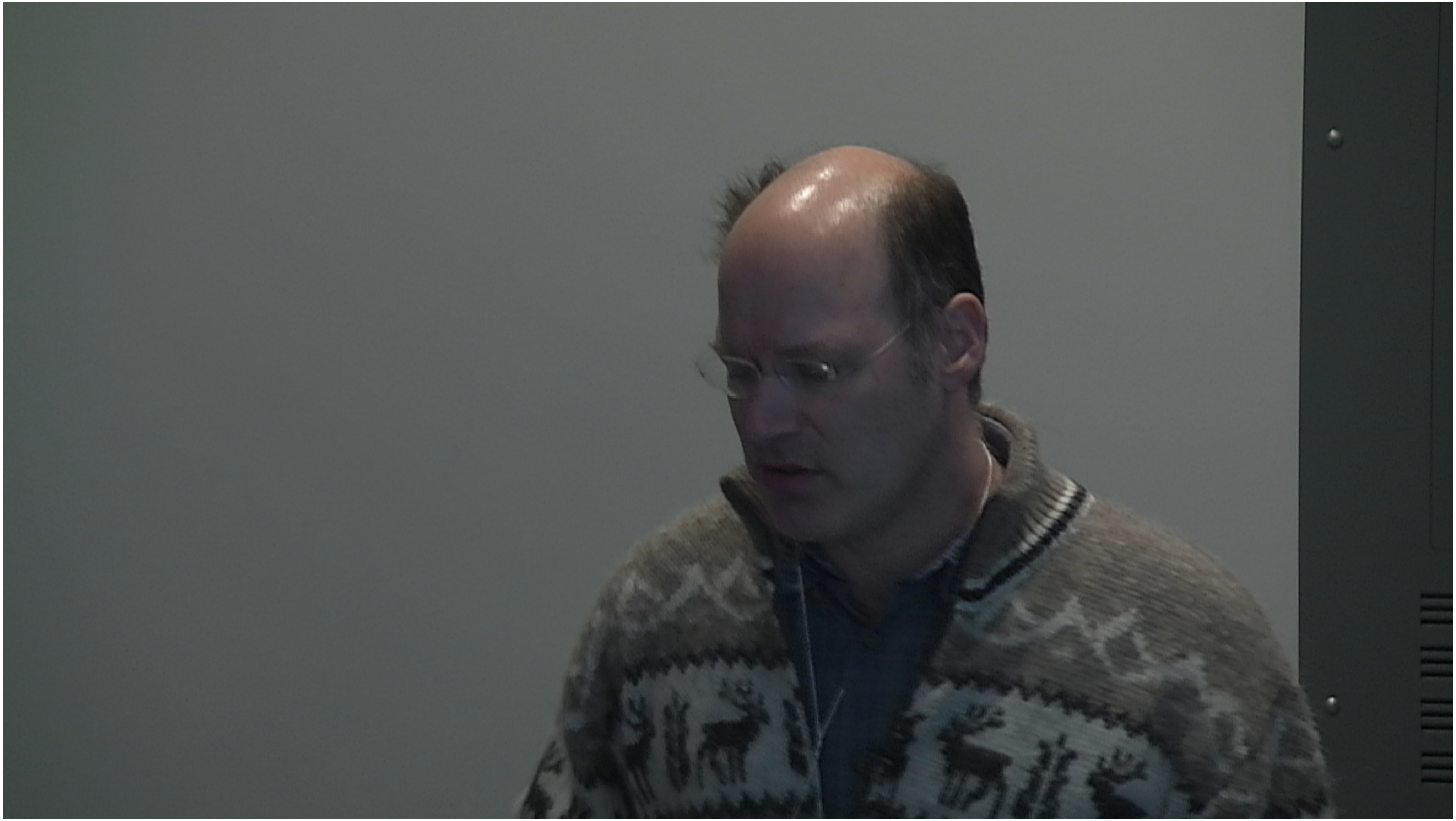
Loses sensitivity at low frequencies since signal is due to induced EMF.

$$V_{EMF} \propto \frac{d\phi}{dt} \propto \omega$$

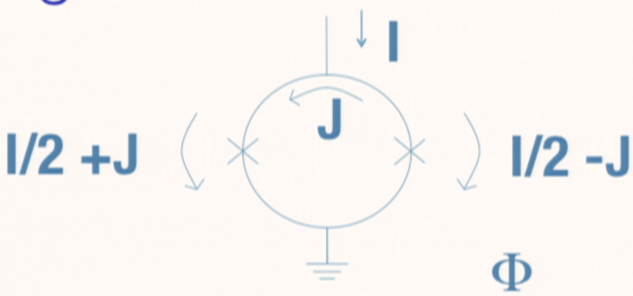
Too many turns reduces highest useable frequency due to intercoil resonance.

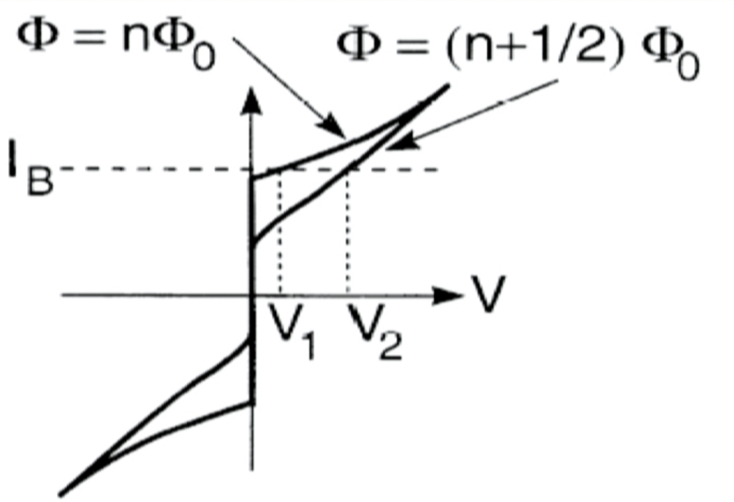
---Phase shifts and non-flat frequency response.



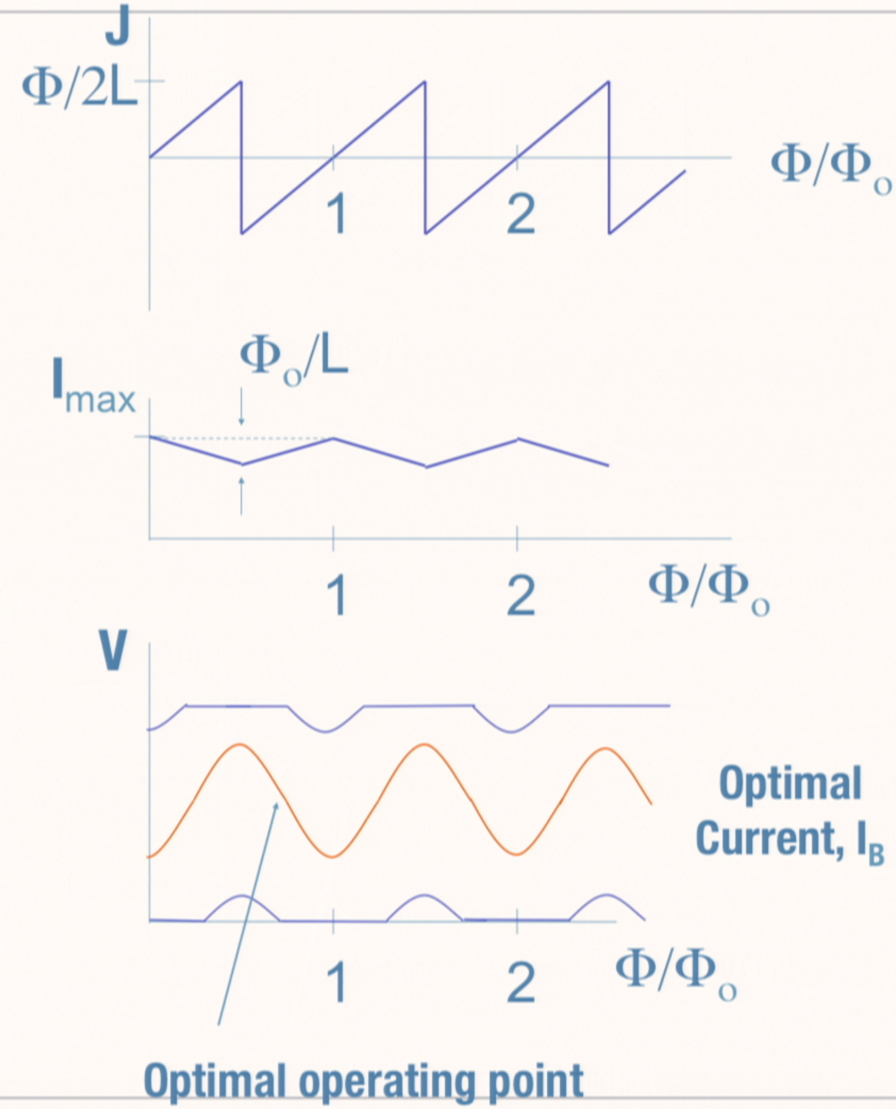


The DC SQUID is the most sensitive detector of magnetic flux





Sensitivity $\sim 1\mu\phi_0/\sqrt{Hz}$

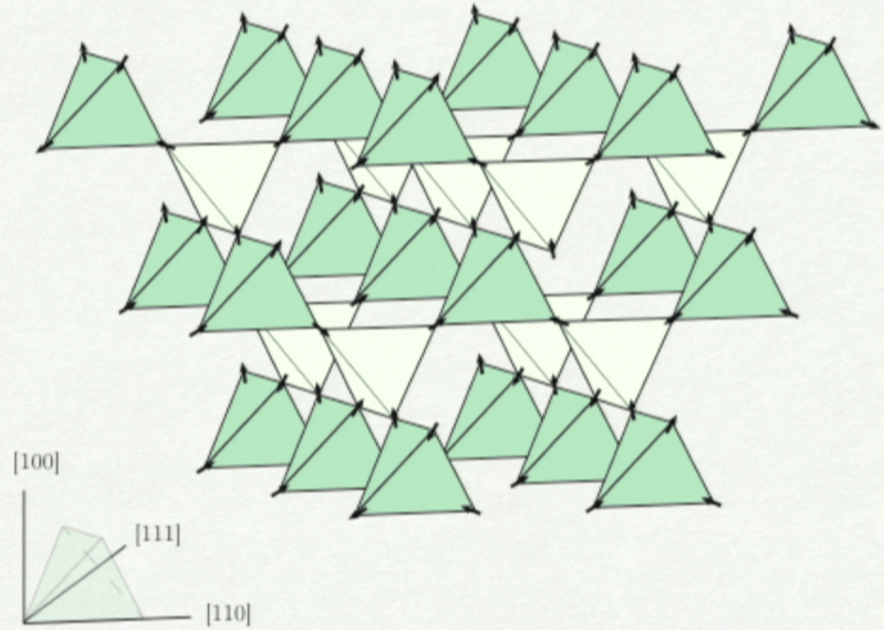


The material, $Dy_2Ti_2O_7$

- ***Interesting because***: magnetic analogue of water ice, magnetic monopole excitations!
- Pyrochlore lattice → Dy (magnetic) ions on corners
- Below 10 K spins are Ising along local <111> axes
- Can be modelled with FM NN exchange interaction

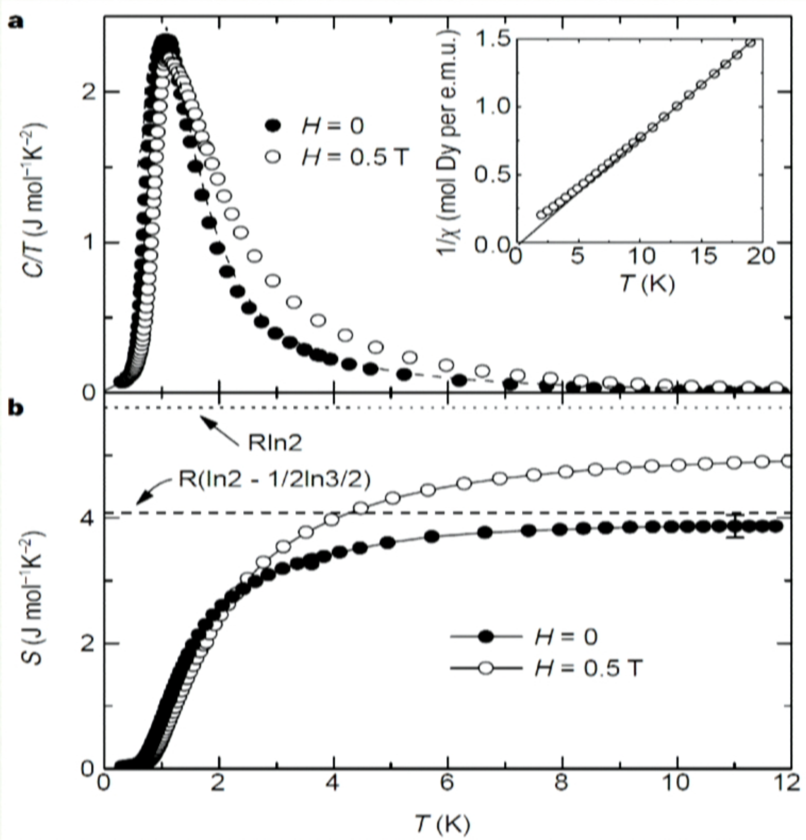
$$H = -J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

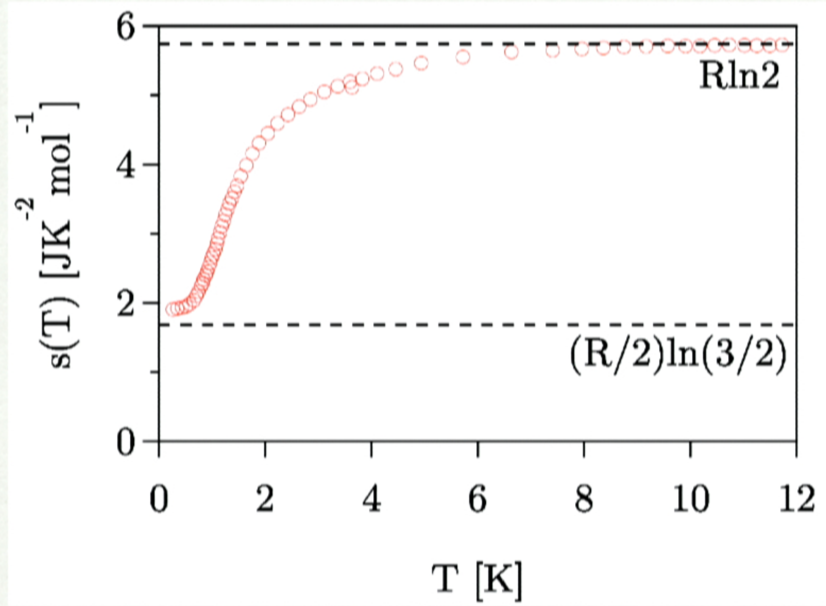
- 2-in-2-out ground state → Bernal & Fowler ice rules → residual entropy



References: Harris et al. PRL 1997, Ramirez et al. Nature 1999

Ramirez et al Measure Specific Heat at Low Temperatures



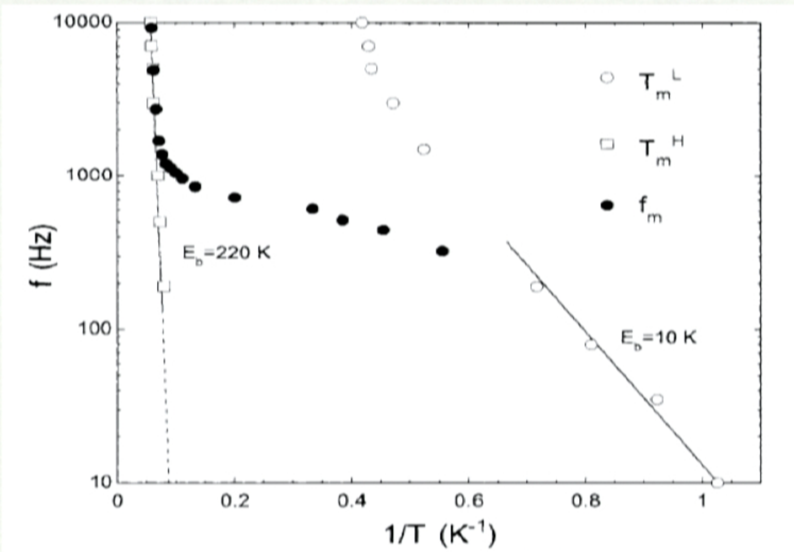


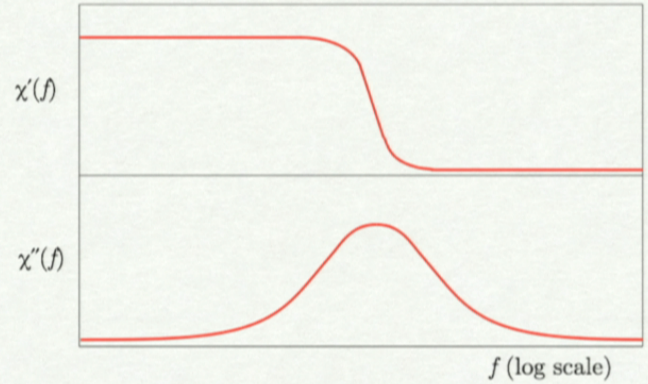
-Find residual entropy just as seen in water ice.

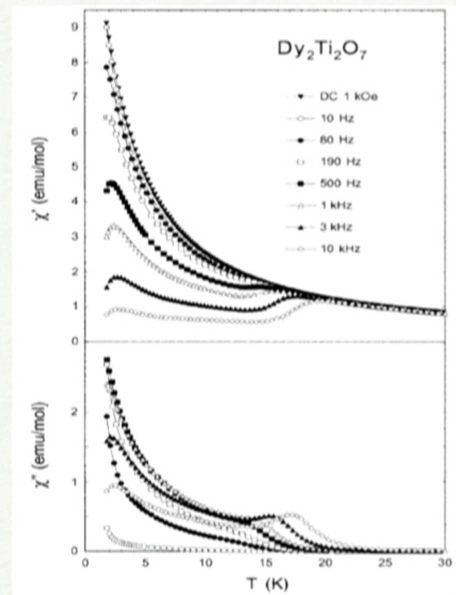
Ramirez, Hayashi, Cava, Siddharthan, Shastry Nature (1999)

Early Susceptibility Measurements

Matsuhira, Hinatsu, Sakakibara
Journal of Physics, Condensed Matter (2001)







Somewhat similar parallel result:

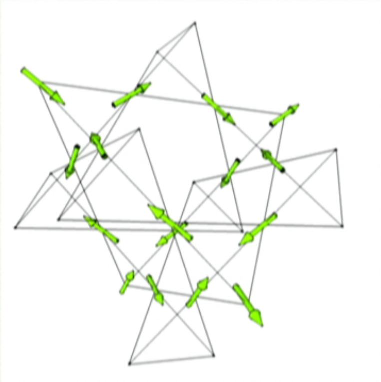
J. Snyder, J. S. Slusky, R. J. Cava, and

P. Schiffer, Nature (2001)

Dipolar spin ice model

$$H = -J \sum_{<ij>} \mathbf{S}_i^{z_i} \cdot \mathbf{S}_j^{z_j} + D r_{nn}^3 \sum_{j>i} \frac{\mathbf{S}_i^{z_i} \cdot \mathbf{S}_j^{z_j}}{|\mathbf{r}_{ij}|^3} - \frac{3(\mathbf{S}_i^{z_i} \cdot \mathbf{r}_{ij})(\mathbf{S}_j^{z_j} \cdot \mathbf{r}_{ij})}{|\mathbf{r}_{ij}|^5}$$

exchange term   dipolar term



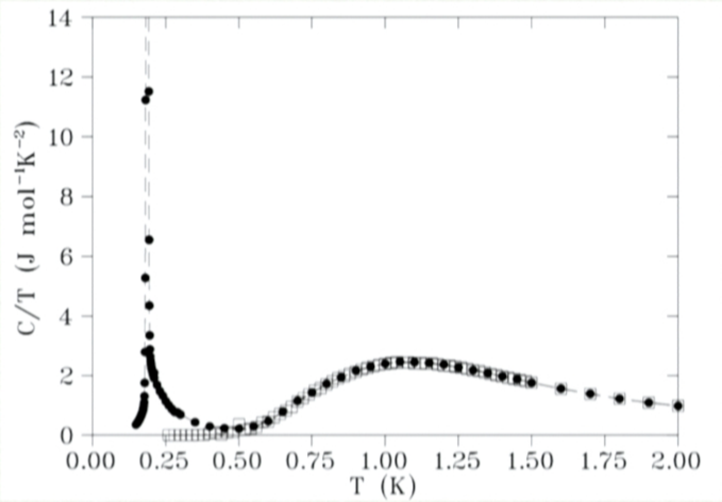
NN effective exchange model:

$$H = -J_{\text{eff}} \sum_{\langle i,j \rangle} \mathbf{S}_i^{z_i} \cdot \mathbf{S}_j^{z_j}$$

$$J_{\text{eff}} = J_{nn} + D_{nn}$$

FM +1.11 K   AFM −1.24 K   FM +2.35 K

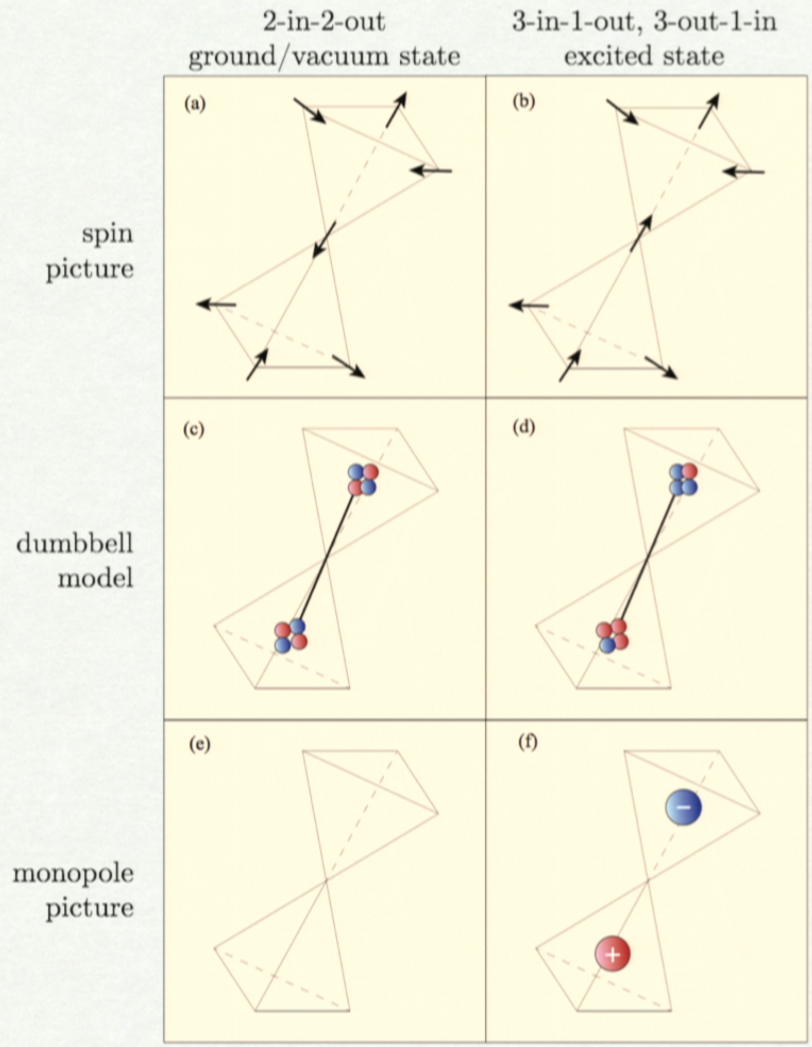
den Hertog and Gingras PRL 2000,
Melko, den Hertog, Gingras PRL 2001
Bramwell and Gingras Science 2001
Yavors'kii et al. PRL 2008



Using loop algorithm, Melko *et al* predict a low temperature ordered state

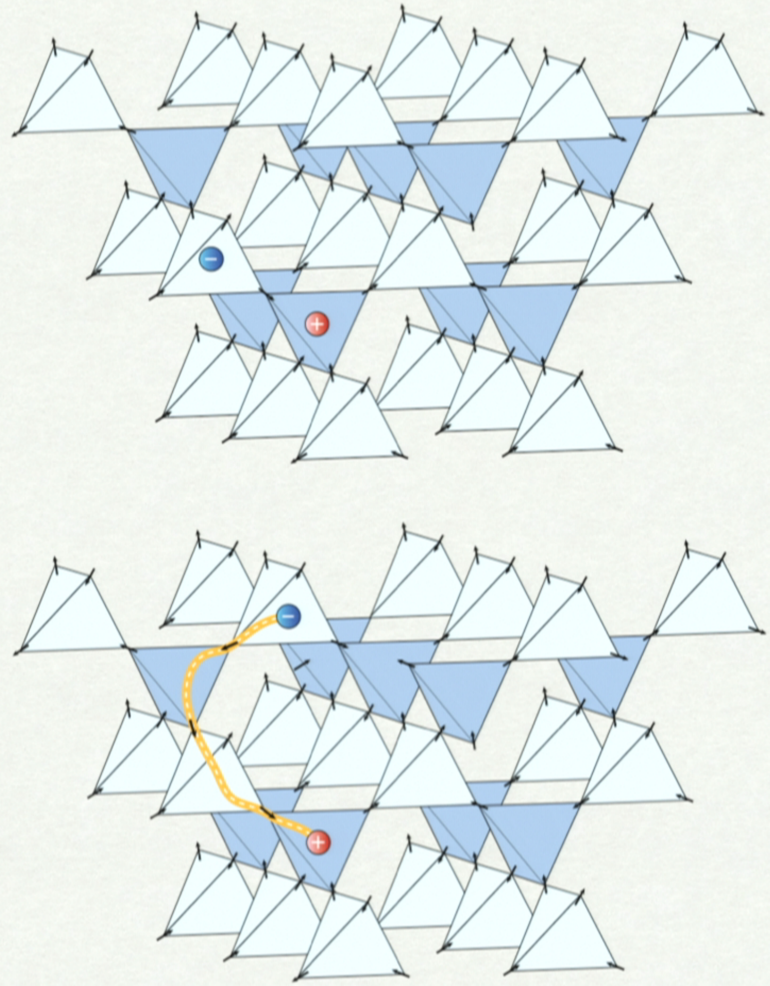
Monopole picture

- Excitations out of ground state viewed as "magnetic charges"
- Motion of monopoles governs dynamics below 2 K



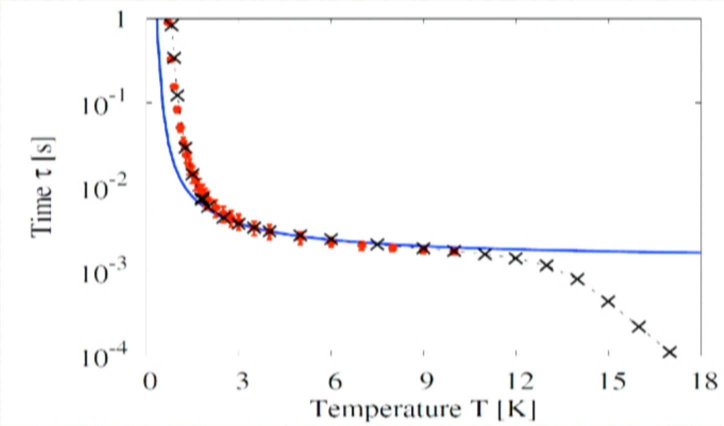
Ryzhkin JETP 2005, Castelnovo et al. Nature 2008, Morris et al. Science 2009, Jaubert and Holdsworth Nature Phys. 2009, Jaubert and Holdsworth, JPCM 2011

- Spin flipped out of the 2-in-2-out ground state
- Single spin flip creates two excitations of equal and opposite charge
- Can be viewed as magnetic monopoles
- The monopole pair can travel apart through subsequent spin flips

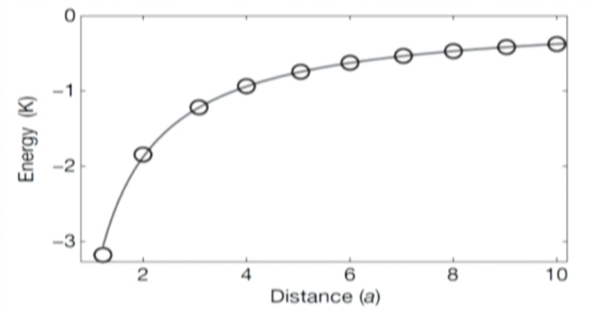


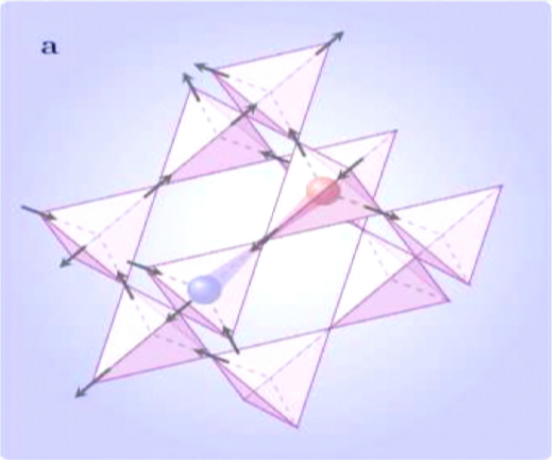
Drawing based on a figure from: Gingras, Science Vol. 326 (375), 2009

Monopole Picture



- Work by Jaubert and Holdsworth interpret features in measurements by Snyder et al. as signatures of monopole-like excitations in spin ices.
- Two-in two-out can be seen as a vacuum
- Three-in one-out (three-out one-in) can be seen as a quasi-particle (monopole).
- Magnetic charges behave as a Coulomb gas of monopoles.





Castelnovo, Moessner, Sondhi, Nature (2008).
Jaubert and Holdsworth, Nature Physics, (2009).

Actually first predicted by Ryzhkin in JETP 101, 481 (2005).

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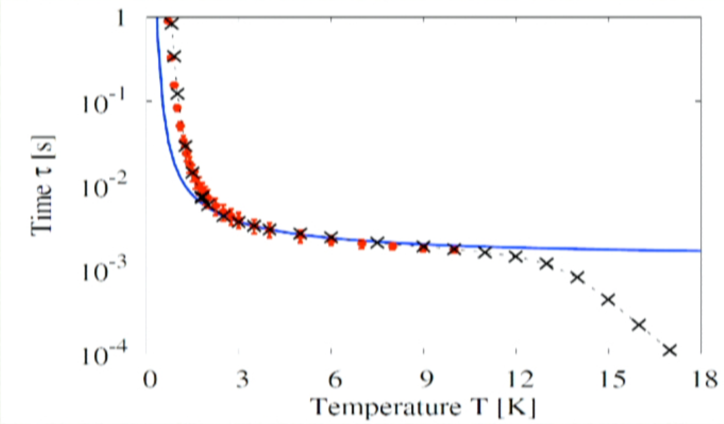

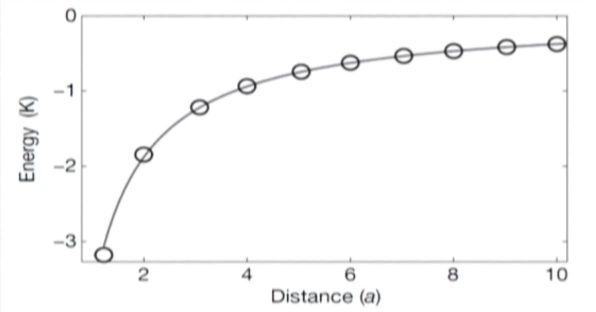

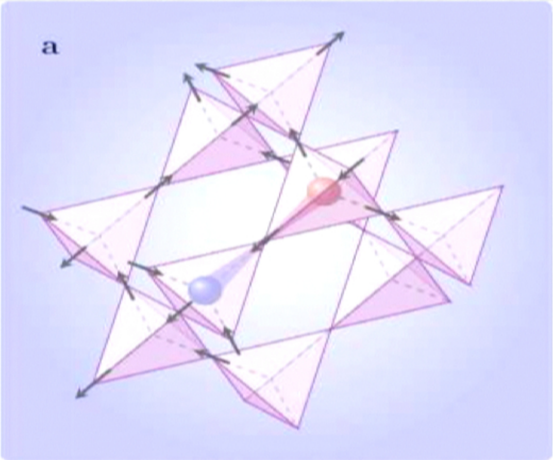

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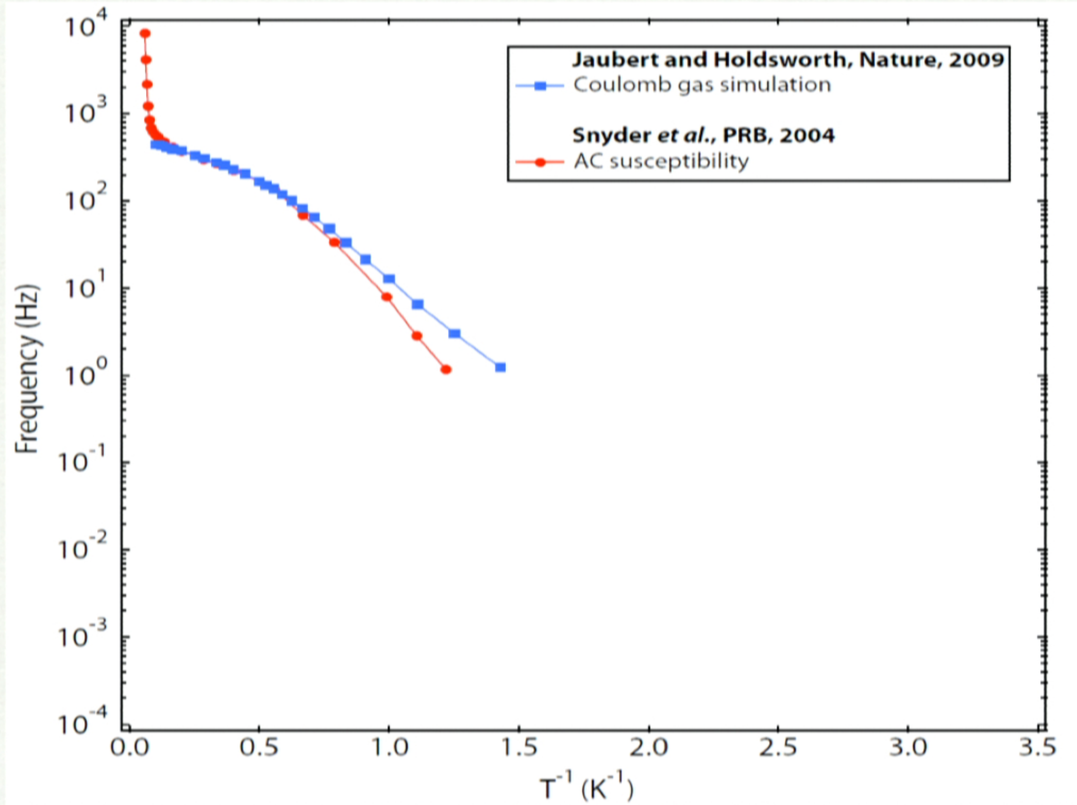
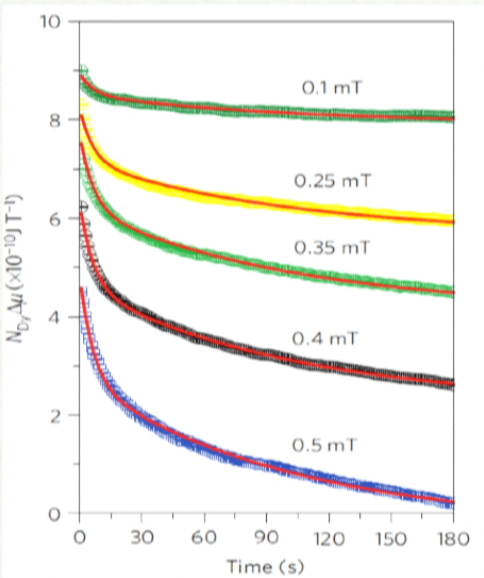

Jaubert and Holdsworth, Nature, 2009
Coulomb gas simulation
Snyder et al., PRB, 2004
AC susceptibility
Frequency (Hz)
T^-1 (K^-1)
10^4
10^3
10^2
10^1
10^0
10^-1
10^-2
10^-3
10^-4
0.0
0.5
1.0
1.5
2.0
2.5
3.0
3.5

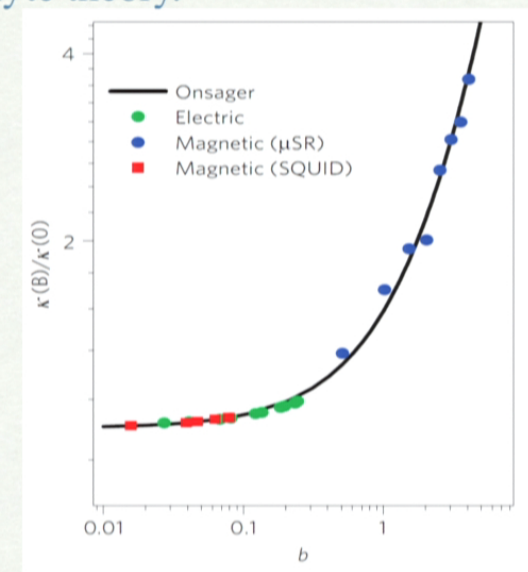
Other Experiments:

Bramwell et al. measured uSR. Found unusual low frequency field fluctuations in a small applied field consistent with theory of monopoles moving through material. “magnetricity” Bramwell et al. Nature (2009)

Giblin et al. (Bramwell group) measure magnetization at 360 mK with SQUID. Find relaxation rate supports monopole magnetolyte theory.

Giblin et al., Nature (2011)





Dunsiger et al. measured uSR , Found fluctuations consistent with silver sample holder
Dunsiger et al Phys. Rev. Lett. (2011)

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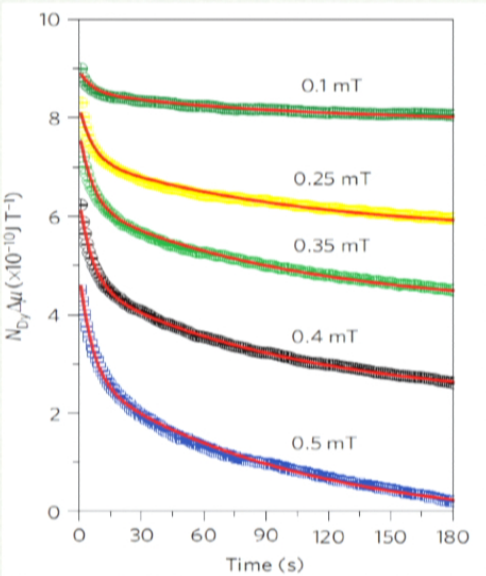

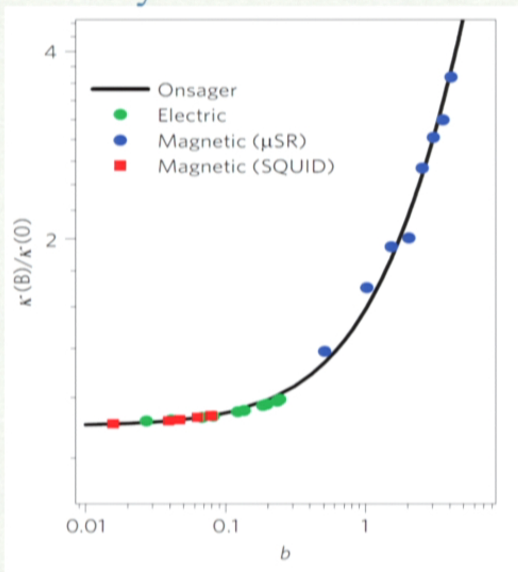

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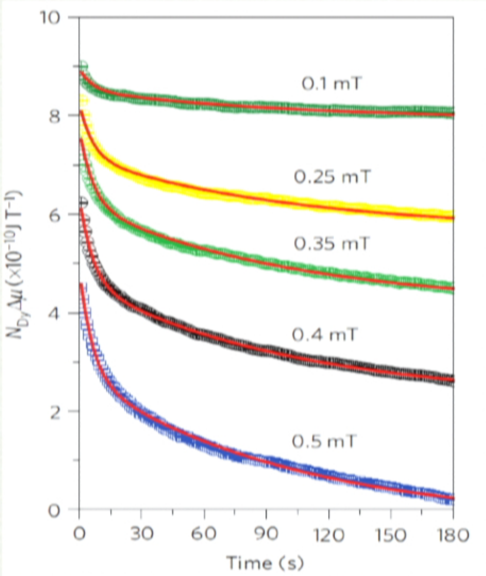

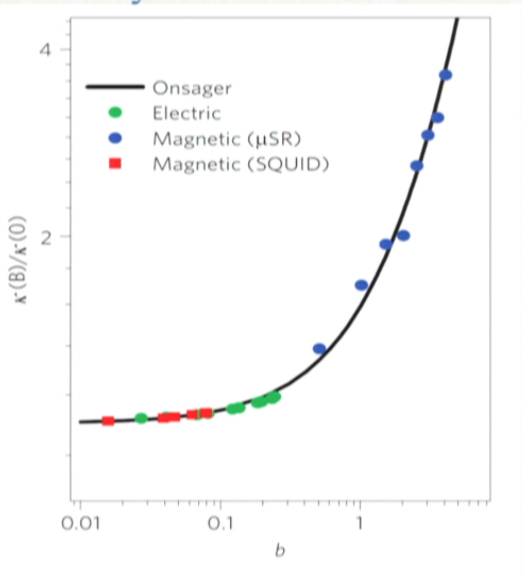

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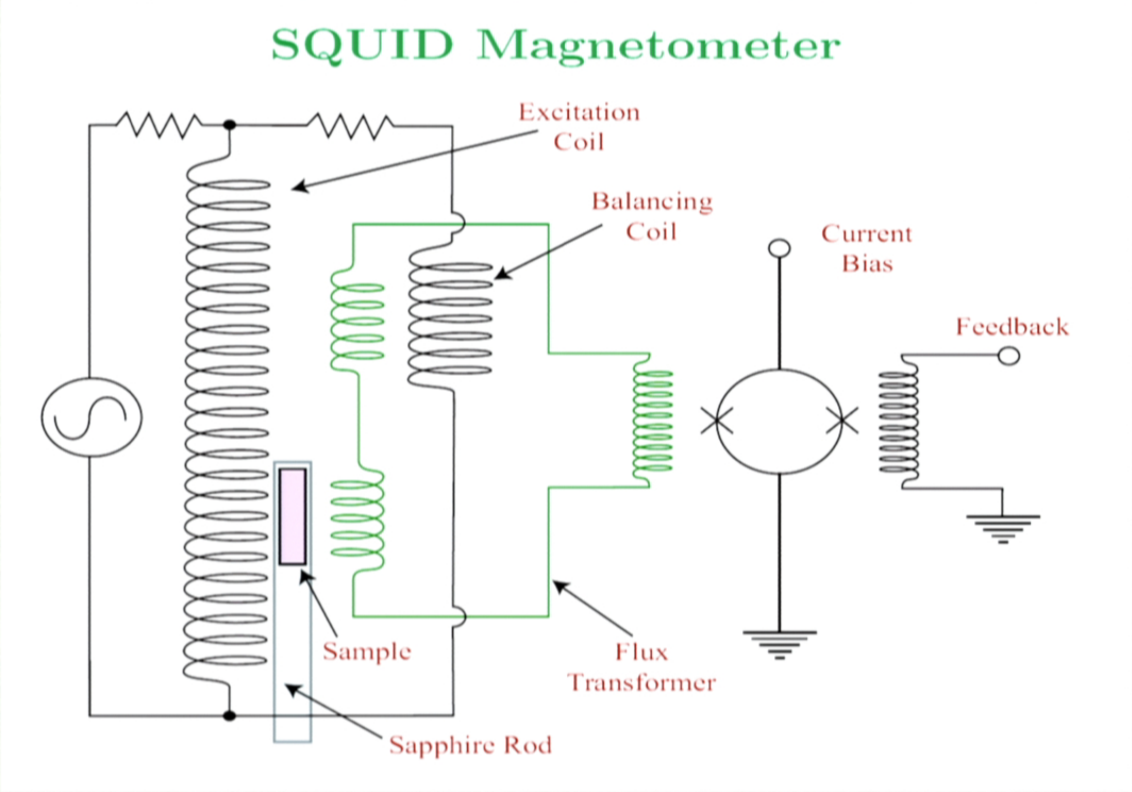
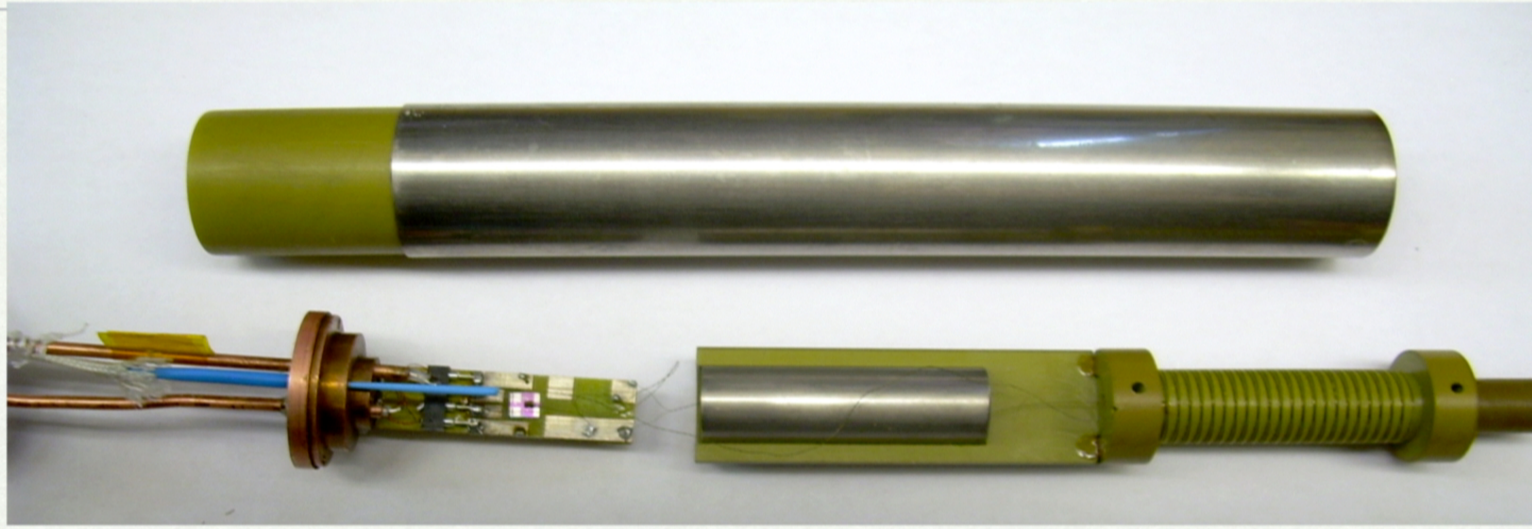
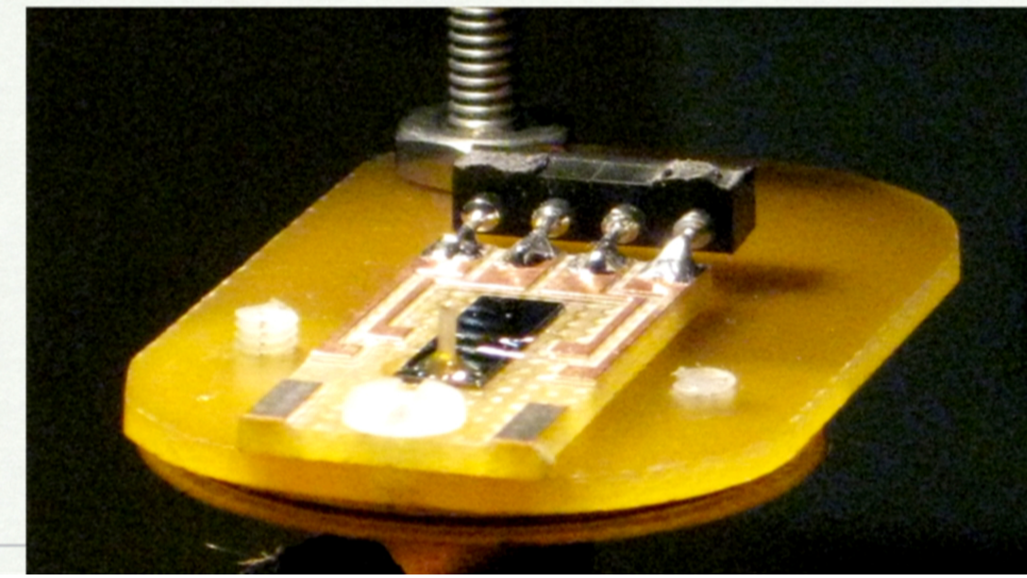
SQUID Magnetometer Measurement

- Use a SQUID with a superconducting flux transformer to make a magnetometer.
- The current sent to the feedback coil produces an equal and opposite field to that provided by the flux transformer.
- This device directly measures flux, as opposed to induced EMF. **Flat Frequency response. No problems with phase shifts.**

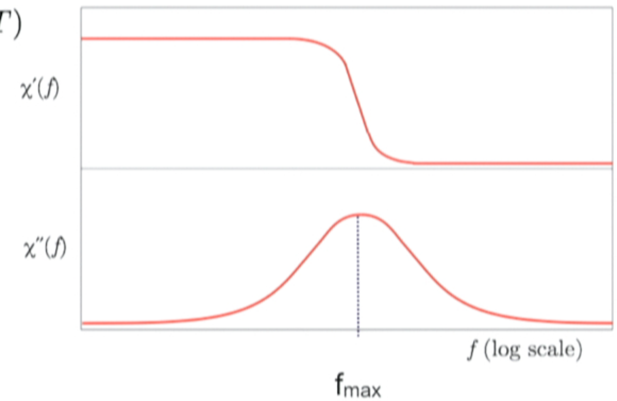




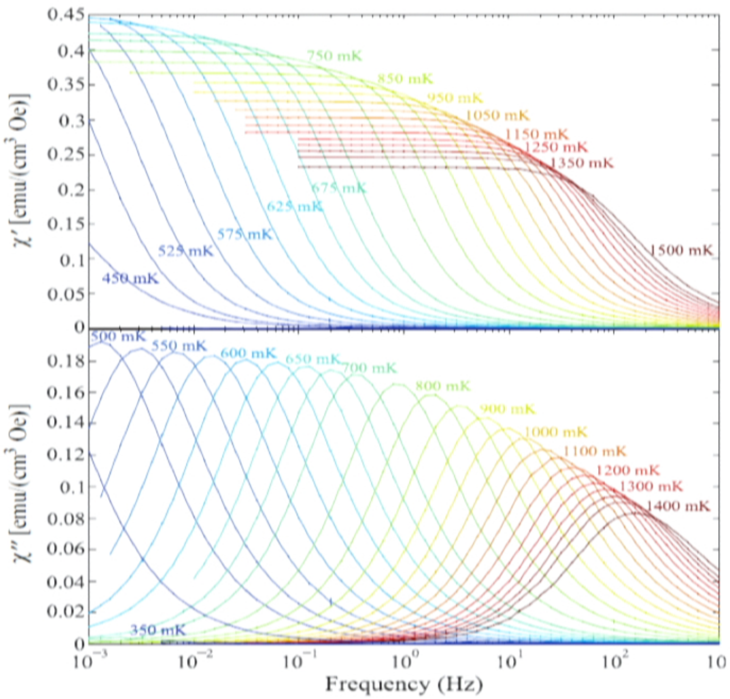


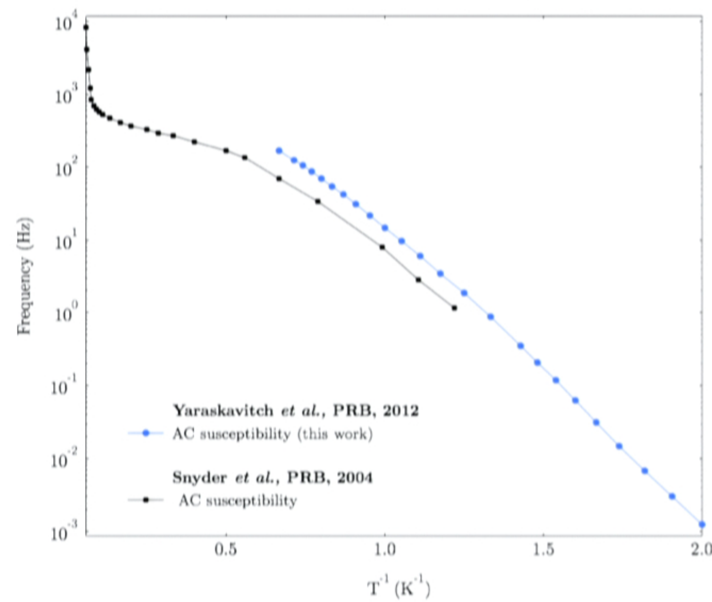
ac susceptibility measurement results for DTO

$$f(T) = f_o \exp(-E_B/kT)$$
$$\tau(T) = \tau_o \exp(E_B/kT)$$
$$\tau_o = \frac{1}{2\pi f_o}$$



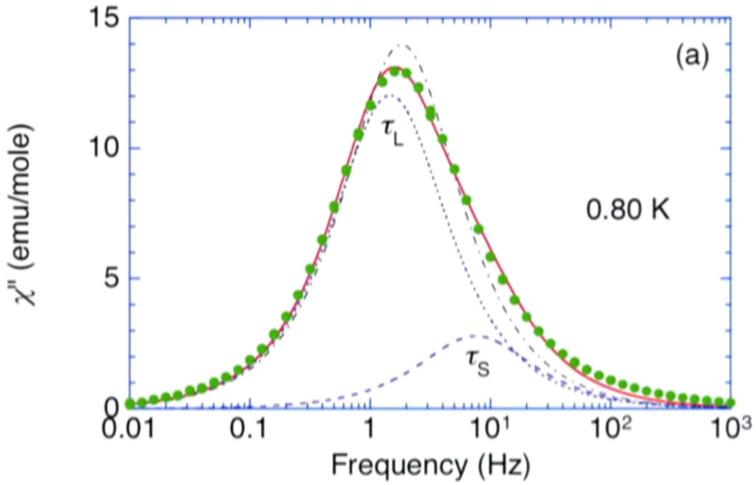
- Measures the relaxation time of the magnetic spins

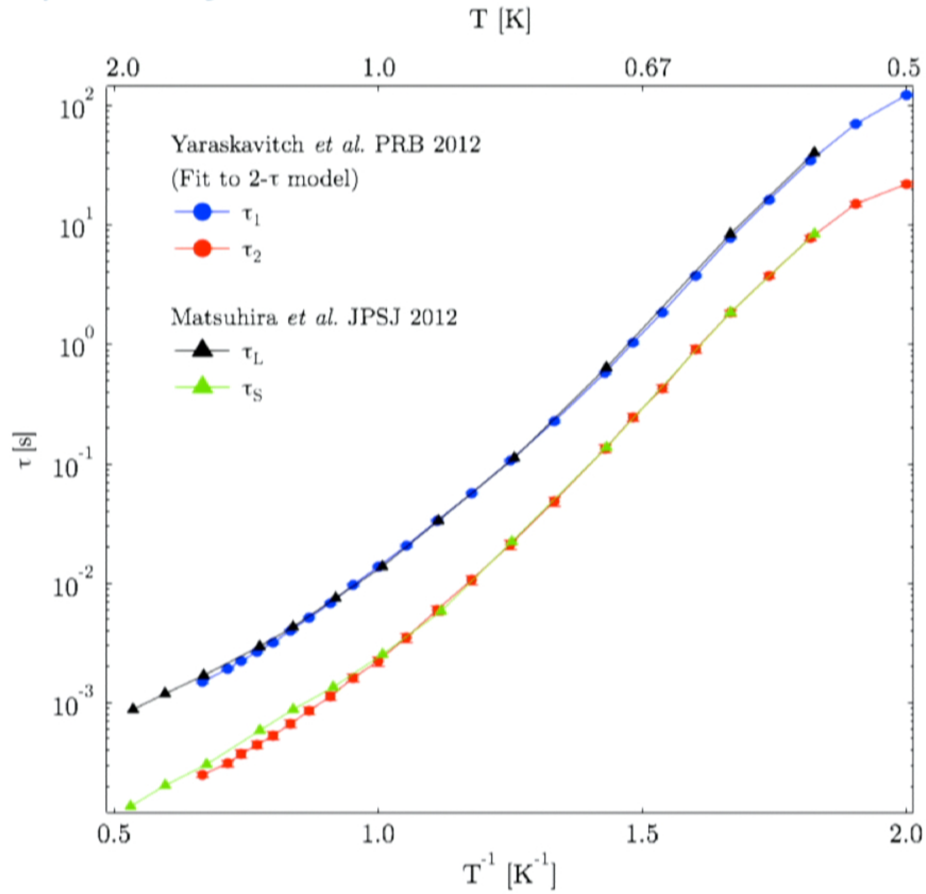




Comparison of our ac susceptibility measurements with Matsuhira *et al*

- The ac susceptibility was analyzed with a 2-τ Debye model to compare with the data from Matsuhira *et al.*
- The results agree very well



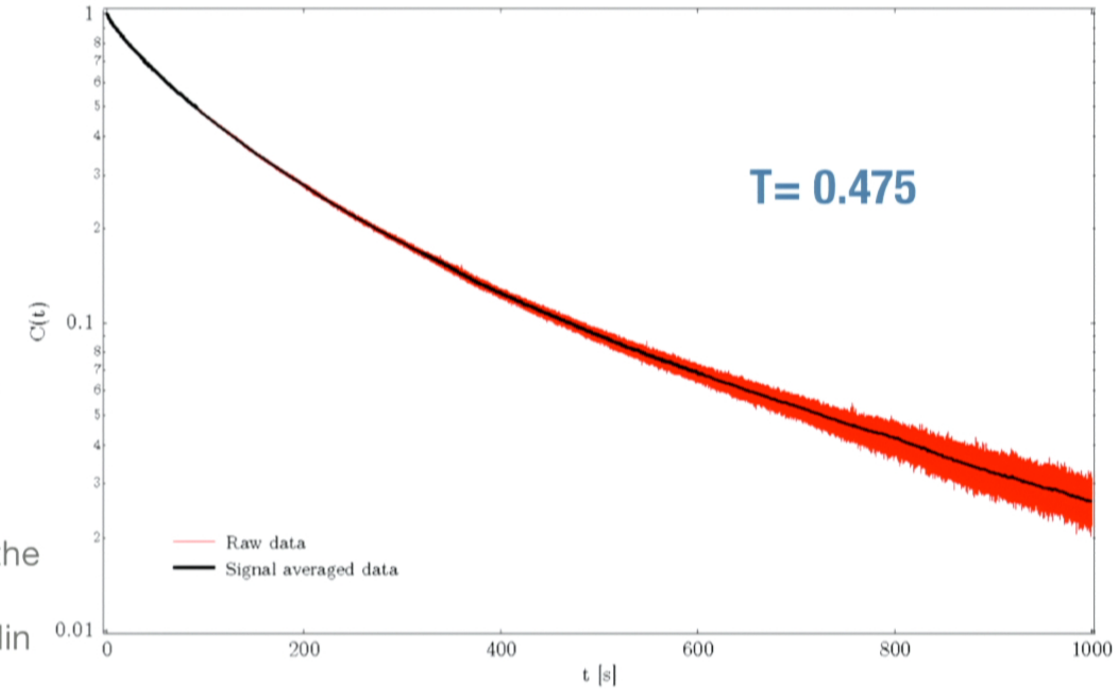


References: Matsuhira et al., JPSJ, 2011, Yaraskavitch et al. PRB, 2012

Magnetization Measurement Results

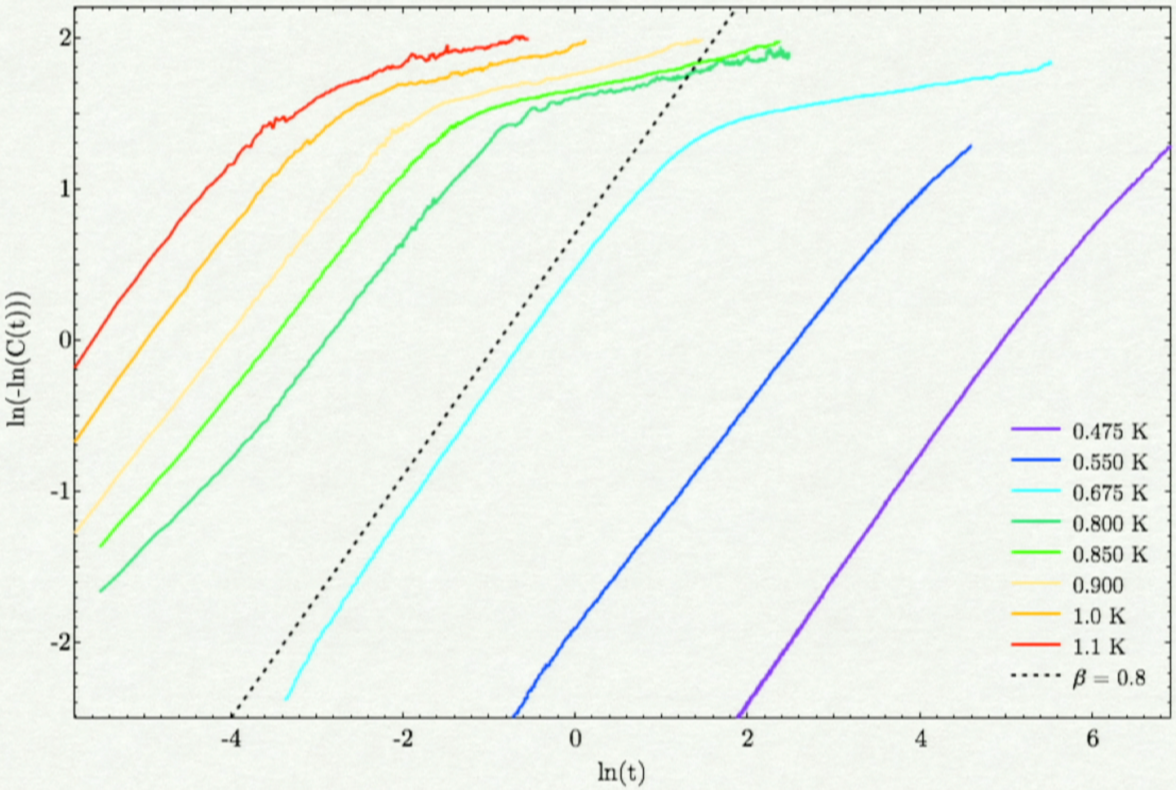
$C(t) = \langle M(t)M(0) \rangle$

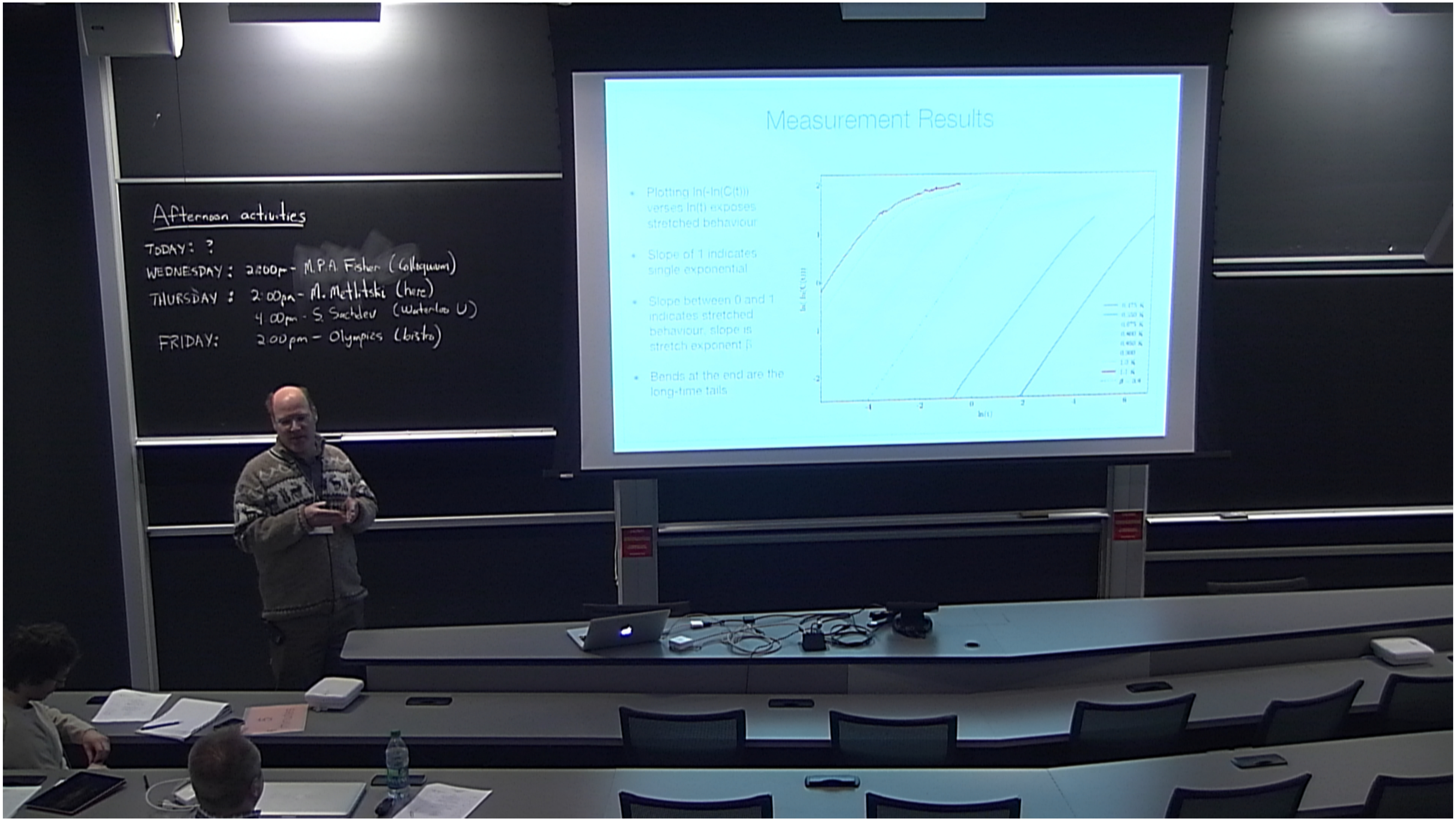
- Apply a small 5 mOe field to magnetize the sample
- Turn off the field to observe the relaxation with the SQUID
- Temperatures measured: 0.475, 0.55, 0.675, 0.8, 0.85, 0.9, 1.0, 1.1 K
- Our results are consistent with the time constants found is the AC susceptbility. Disagree with Giblin *et al.*



Measurement Results

- Plotting ln(-ln(C(t))) verses ln(t) exposes stretched behaviour
- Slope of 1 indicates single exponential
- Slope between 0 and 1 indicates stretched behaviour, slope is stretch exponent β
- Bends at the end are the long-time tails




Afternoon activities
TODAY: ?
WEDNESDAY: 2:00pm - M.P.A. Fisher (Colloquium)
THURSDAY: 2:00pm - M. Metlitski (here)
4:00pm - S. Sachdev (Waterloo U)
FRIDAY: 2:00pm - Olympics (bistro)
Measurement Results
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Slope of 1 indicates single exponential
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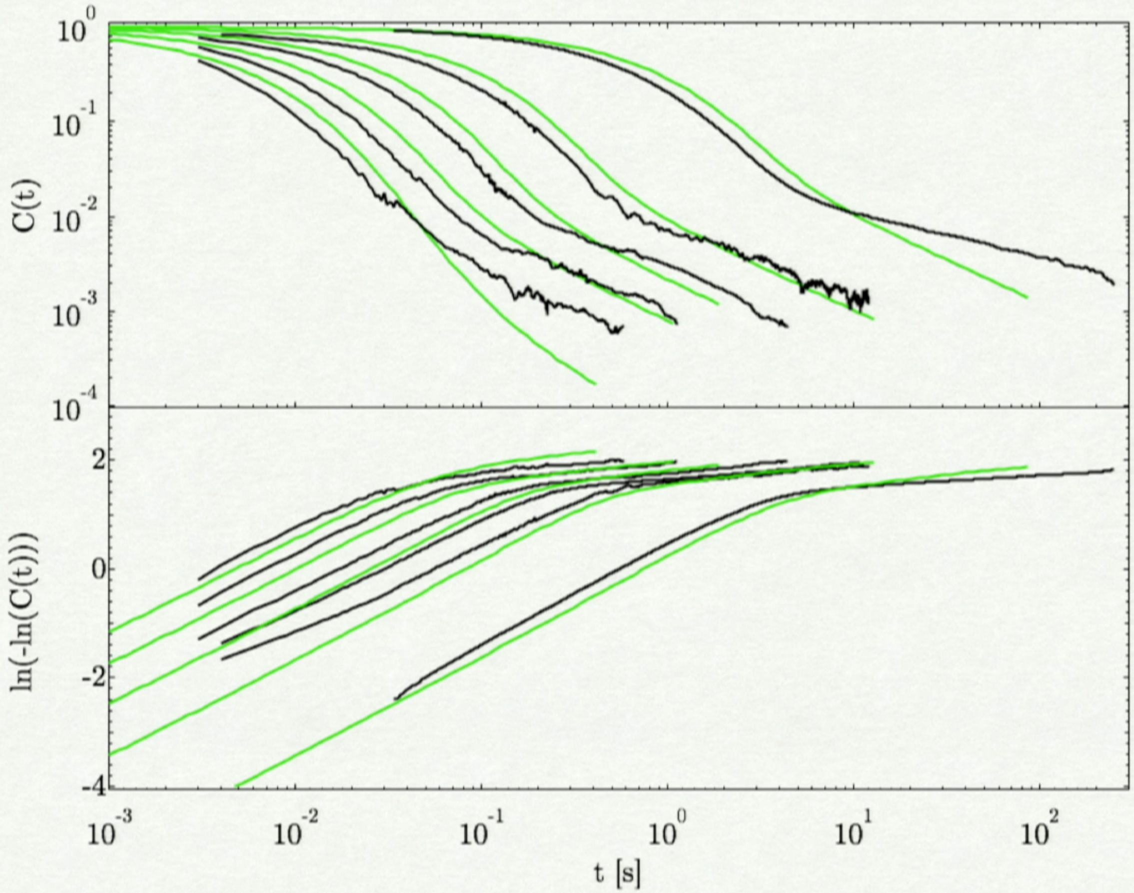
Bends at the end are the long-time tails
ln(t)

Comparison of trasnsformed AC susceptibility to DC magnetization.

- AC susceptibility was transformed into time domain
- Long-time tail
- Stretched decay, exponent ~0.7-0.8
- Arrhenius relaxation with an energy barrier ~9 K

Black - dc magnetization
Green - transformed ac susceptibility



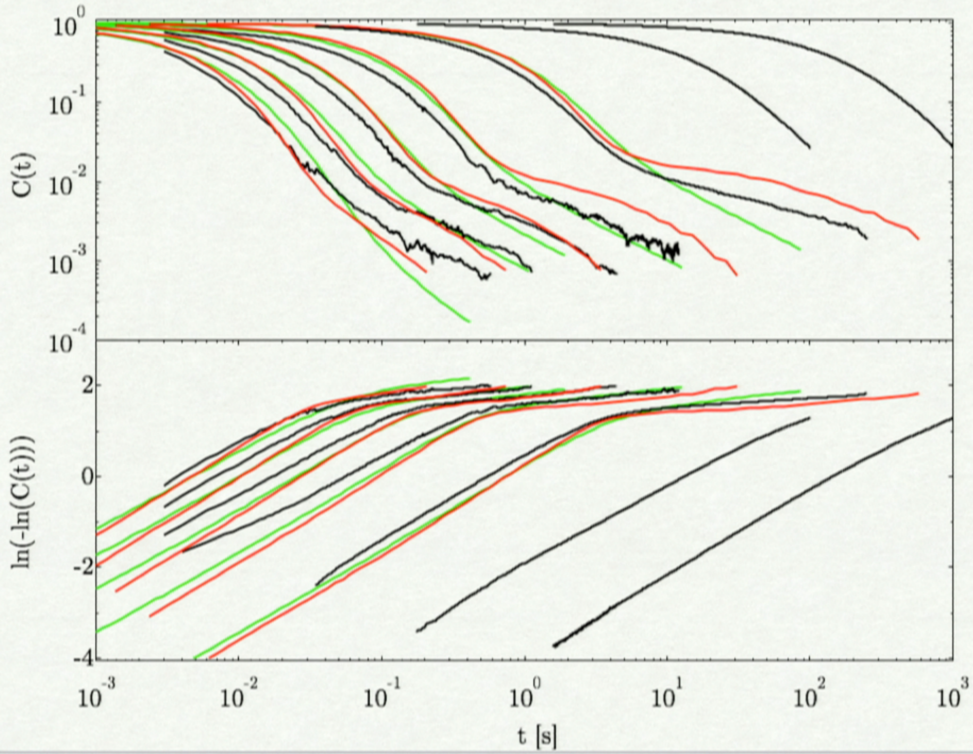
Modified Monte Carlo Simulation

Dipolar Spin Ice Model: no tail, single exponential decay, energy barrier 5 K

Modified MC simulation in red (Patrik Henelius) :

- **Tail: 0.3 % stuffed spins**
- **Stretched decay: open boundary**
- **Barrier 9K: attempt rate scales with monopole density**

Black - dc magnetization
Green - transformed ac susceptibility
Red - Monte Carlo simulation



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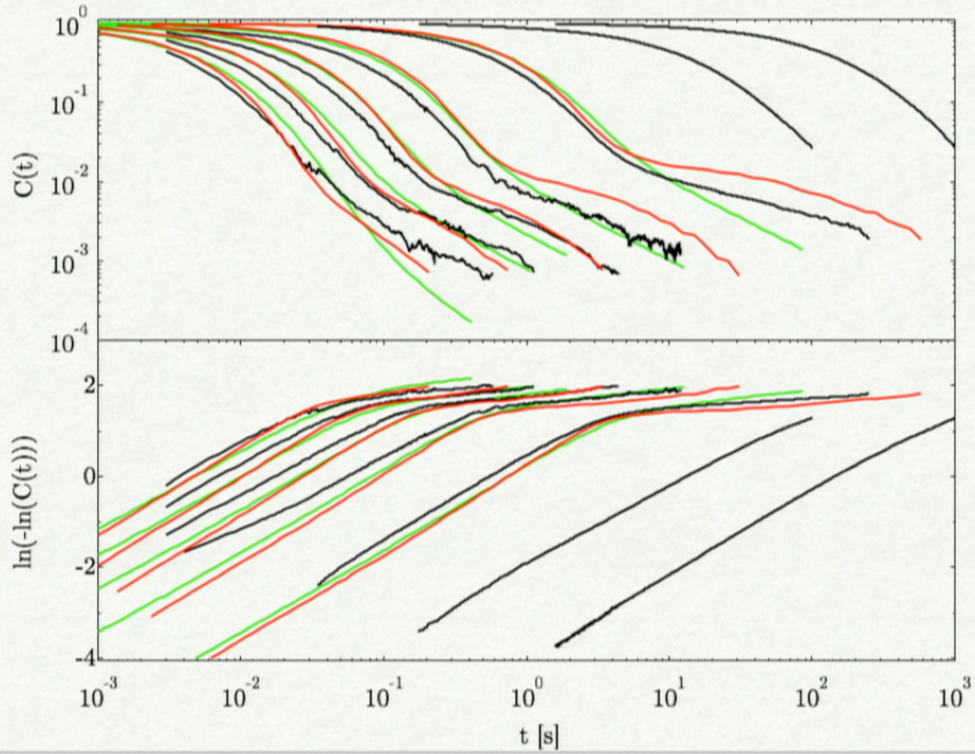
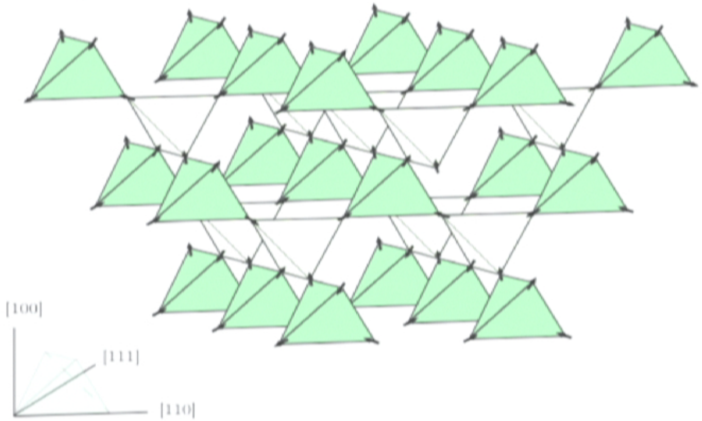
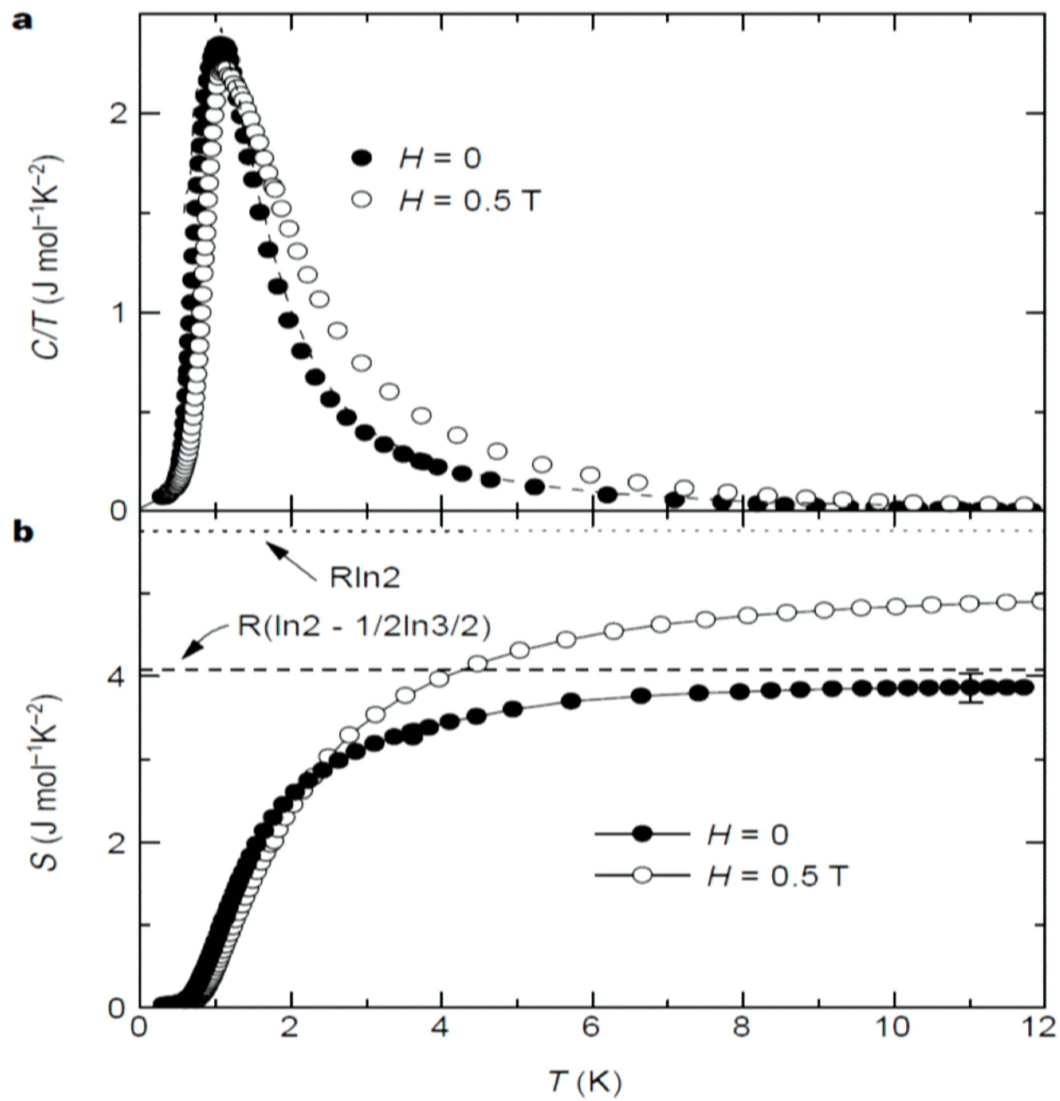


Residual entropy in spin ice

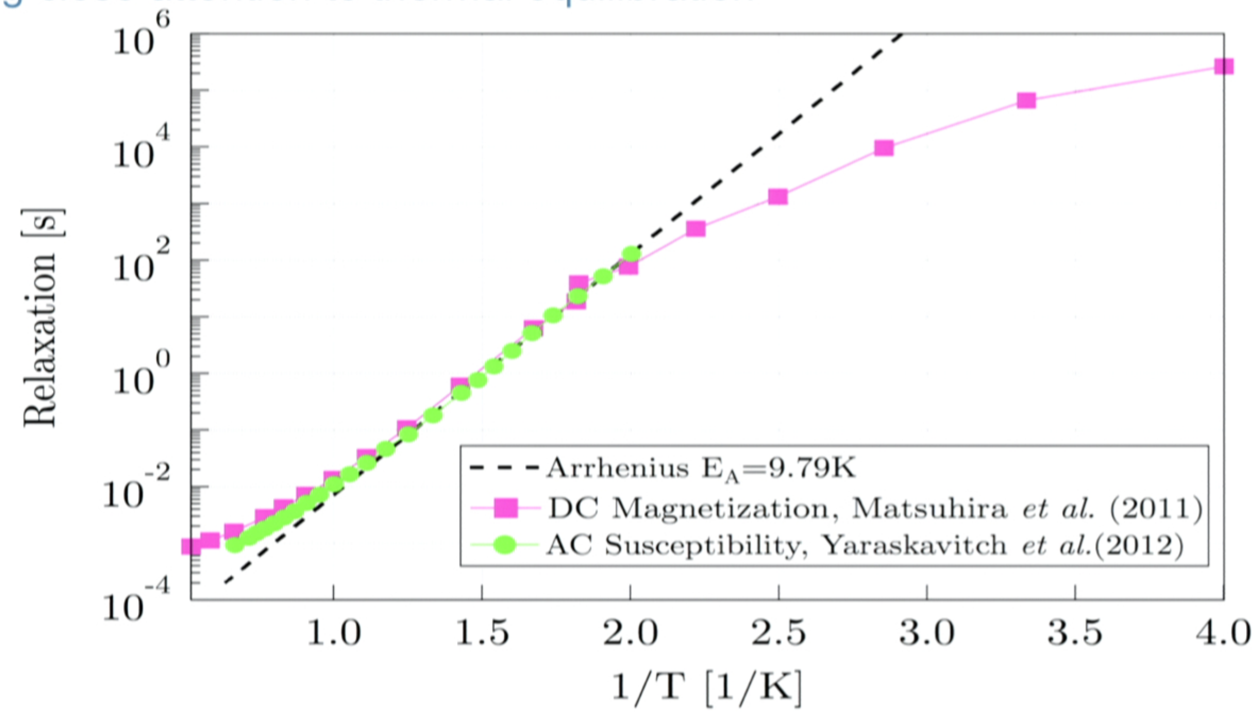


- Geometrical frustration in water ice:
  L. Pauling, J. A. C. M., Vol. 57, p.2680 (1935)
- Ising antiferromagnetism on pyrochlore lattice:
  P.W. Anderson, Phys. Rev. 102, p.1008 (1956)
- "Spin ice" is discovered:
  M. J. Harris, S. T. Bramwell, D. F. McMorrow, T. Zeiske, and K. W. Godfrey, Phys. Rev. Lett. 79, p.2554 (1997)
- Pauling's residual entropy measured in spin ice: A. P. Ramirez, A. Hayashi, R. J. Cava, R. Siddharthan & B. S. Shastry, Nature 399, p.333 (1999)
- Refined model of spin ice with dipolar interactions:
  B. C. den Hertog and M. J. P. Gingras, Phys. Rev. Lett., Vol. 84, p.3430 (2000)



Magnetic relaxation in $Dy_2Ti_2O_7$

- AC susceptibility and DC magnetization indicate that the spin relaxation time markedly increases as the temperature is lowered
- The *extremely slow spin dynamics* in this material motivated us to re-measure its specific heat, while paying close attention to thermal equilibration



[K. Matsuhira, C. Paulsen, E. Lhotel, C. Sekine, Z. Hiroi, S. Takagi, J. Phys. Soc. Jpn. 80 (2011) 123711]
[Yaraskavitch, L. R. and Revell, H. M. and Meng, S. and Ross, K. A. and Noad, H. M. L. and Dabkowska, H. A. and Gaulin, B. D. and Kycia, J. B.Phys. Rev. B 85, 020410(R) (2012)]

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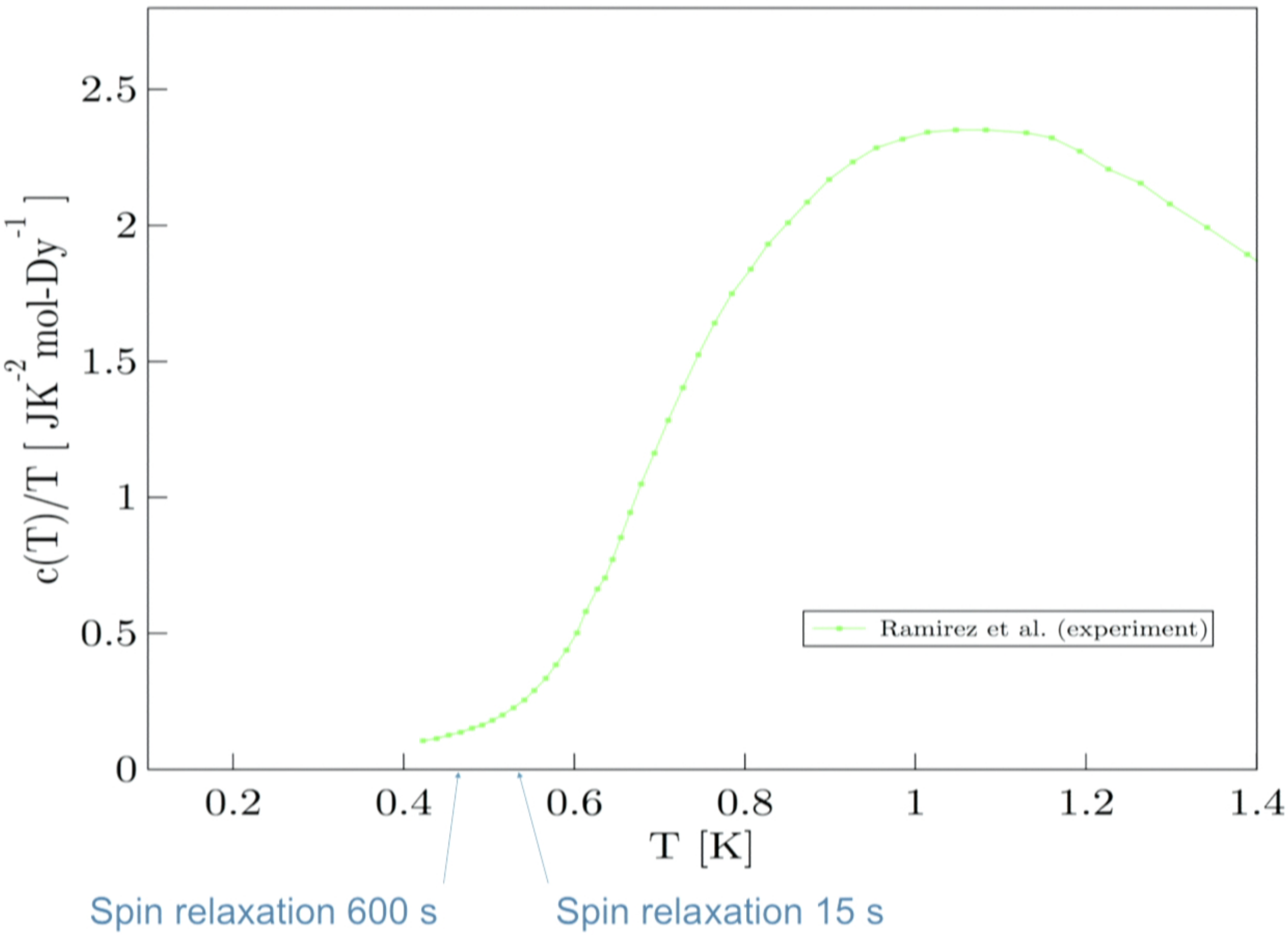
Previous specific heat measurements

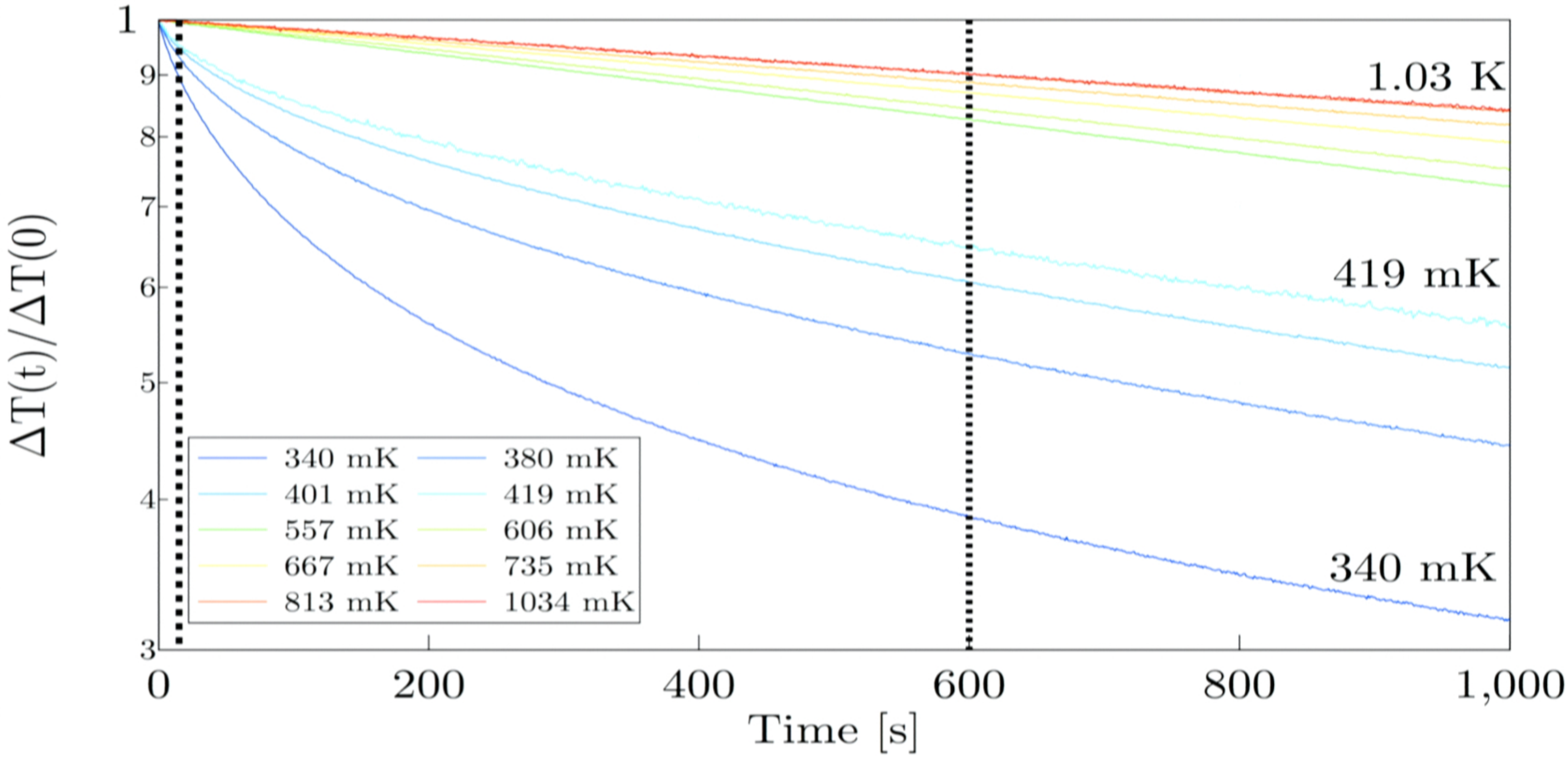


Thermal equilibration time:

- Ramirez *et al.* ~15 s
- Klemke *et al.* ~ 600 s
- Others did not specify

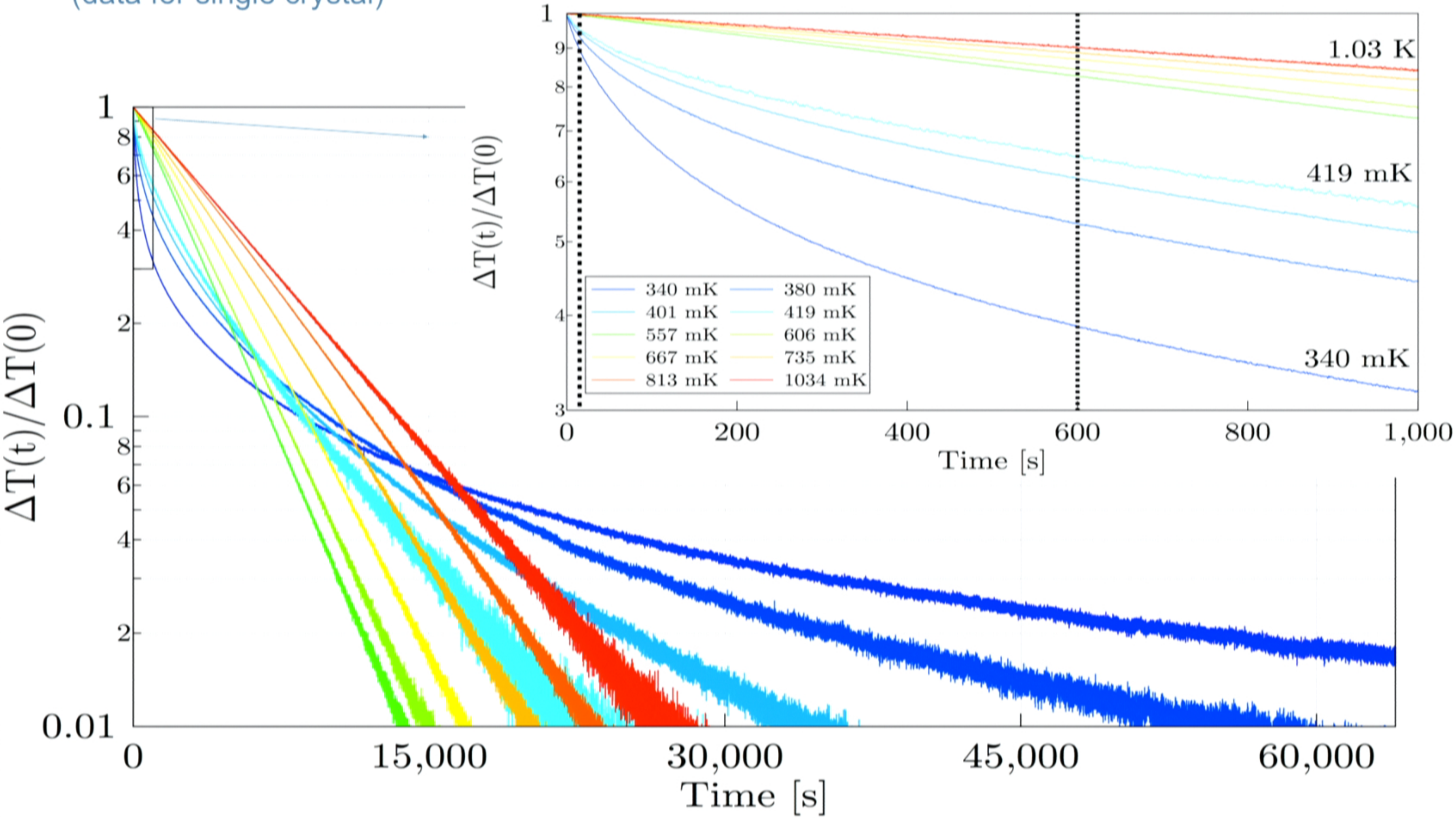
Thermal Relaxation in $Dy_2Ti_2O_7$

(data for single crystal)

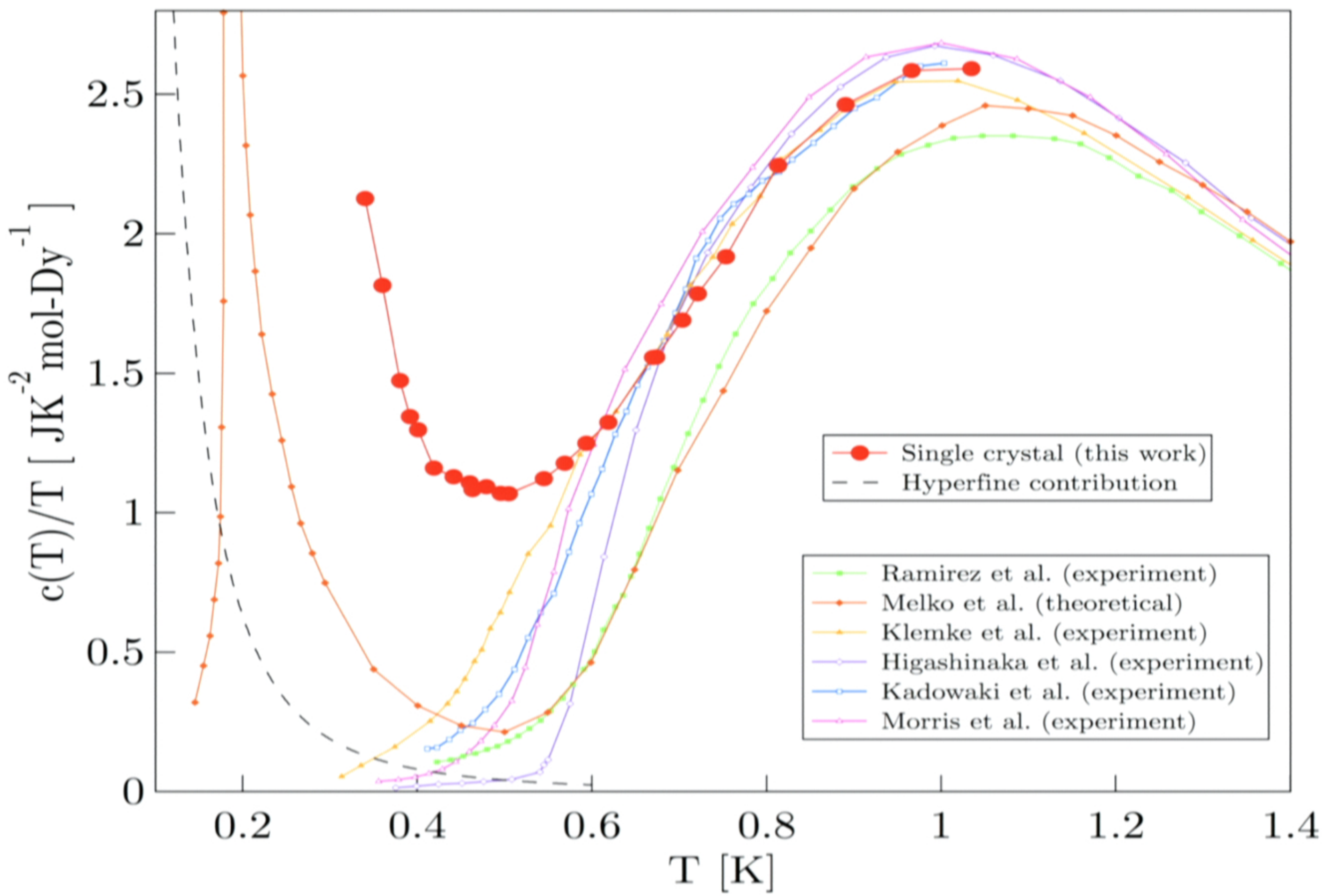


Thermal Relaxation in $Dy_2Ti_2O_7$

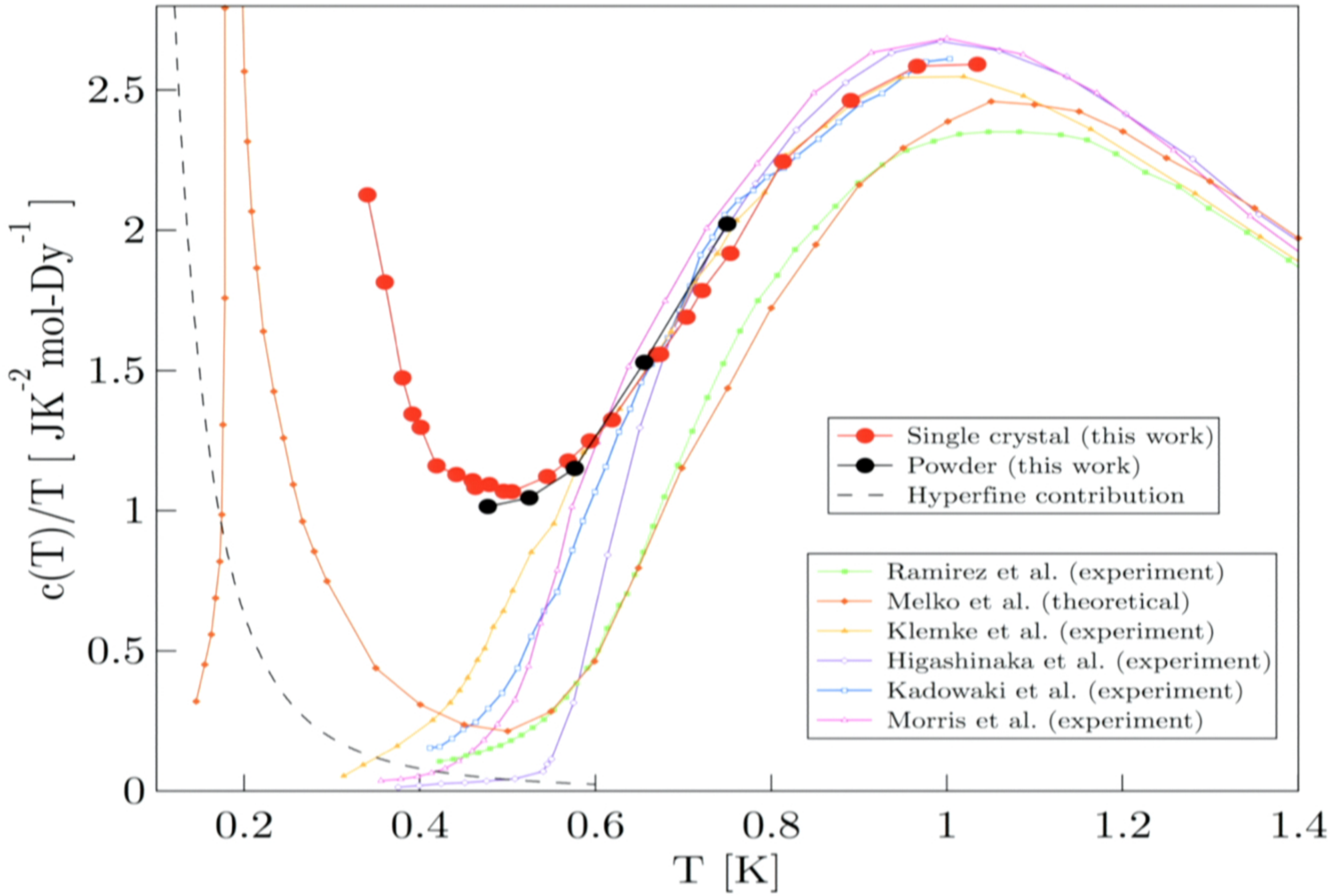
(data for single crystal)



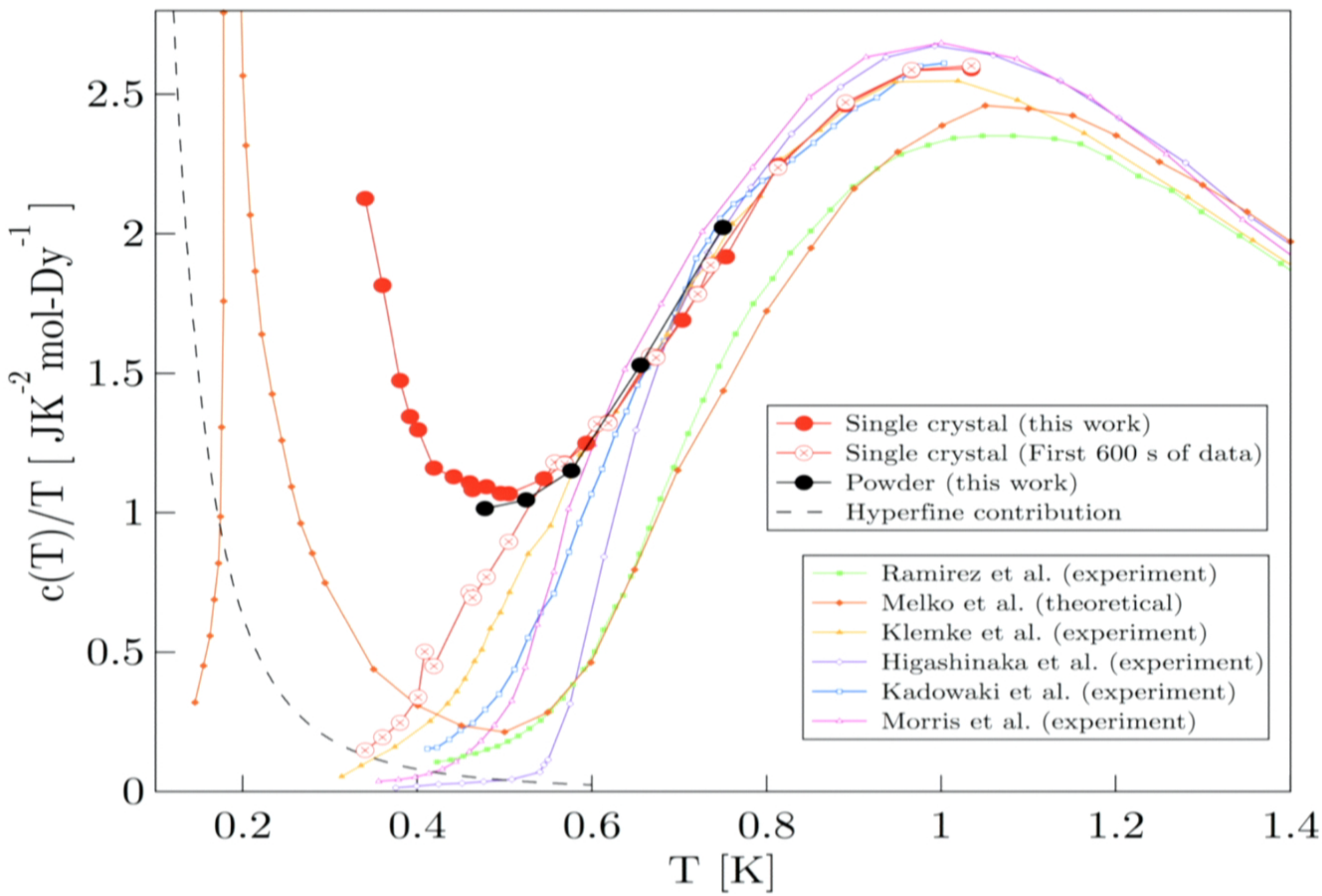
Specific heat of $Dy_2Ti_2O_7$



Specific heat of $Dy_2Ti_2O_7$



Specific heat of $Dy_2Ti_2O_7$



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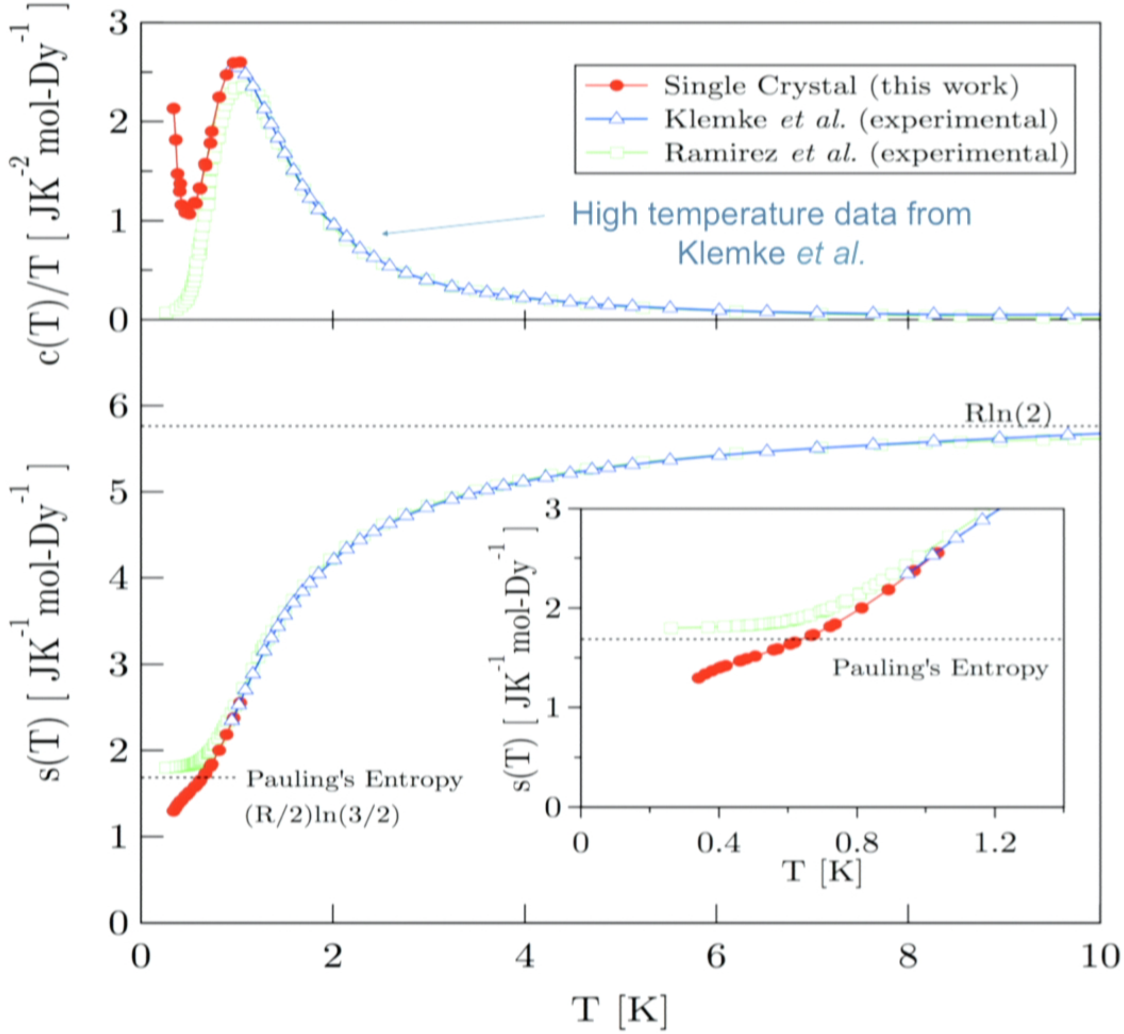
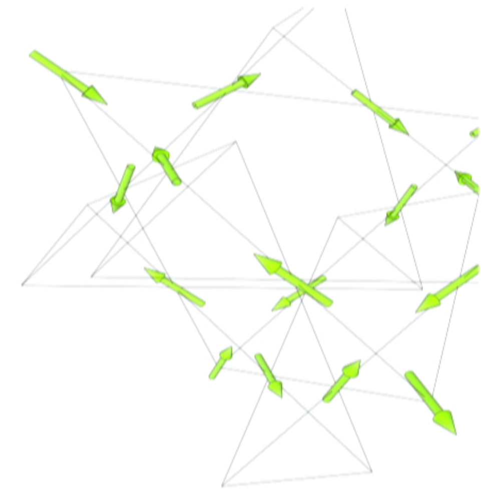
Absence of Pauling's residual entropy in thermally equilibrated $Dy_2Ti_2O_7$

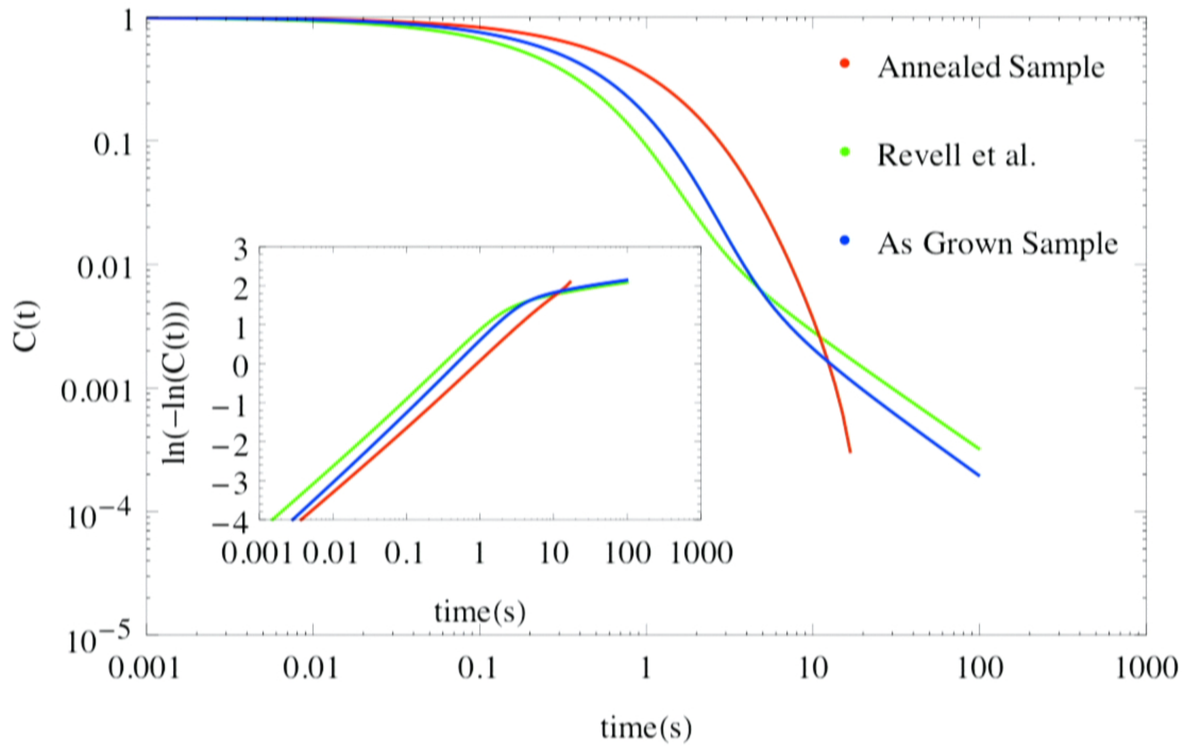




MDG phase?

Detailed nature of the ordered state cannot be concluded from heat capacity measurements alone

Annealing Dy2Ti2O7, removes long time tail



"Vacancy defects and monopole dynamics in oxygen-deficient pyrochlores"
G. Sala, M. J. Gutmann, D. Prabhakaran, D. Pomeranski, C. Michelitis J. B. Kycia, D. G. Porter, C. Castelnovo and J. P. Goff, Nature Materials (just accepted).

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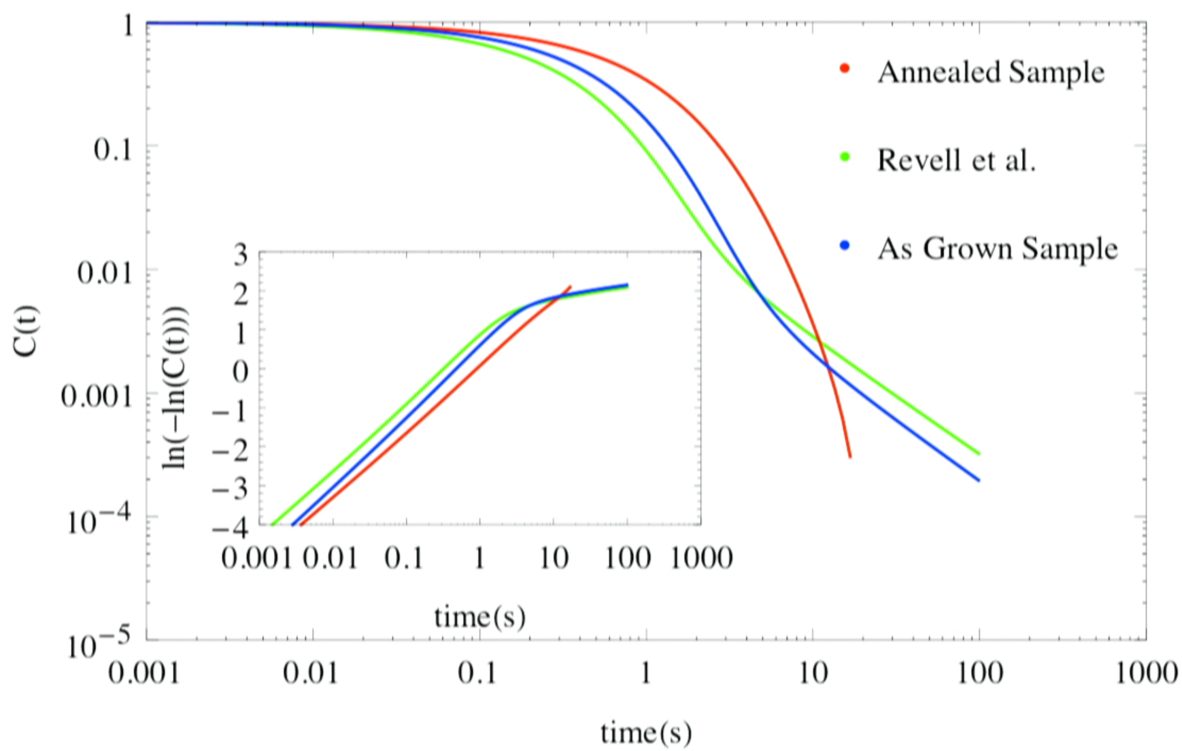

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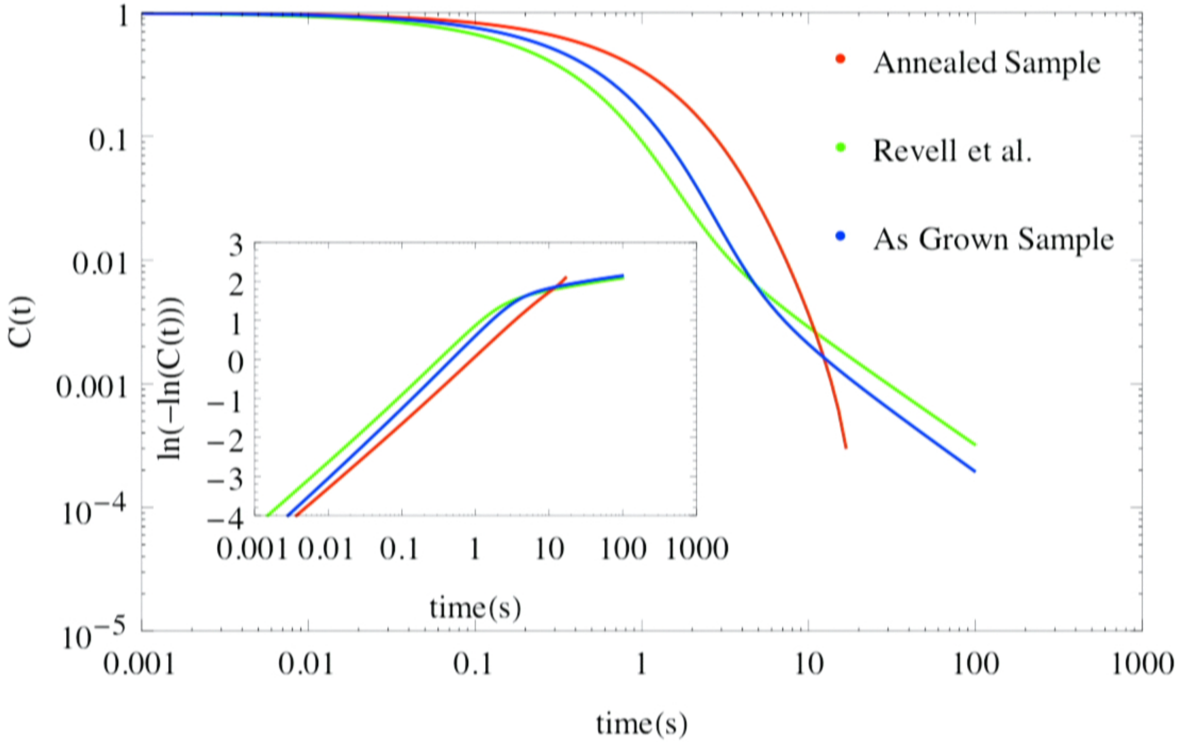

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Conclusion

- We have shown the first example of the effect of a small amount of defects on monopole dynamics.
- The next step will be to quantitatively relate characteristics in the relaxation to the levels of impurities. Maybe relaxation measurement will be the equivalent to residual resistance measurement for electrical conductors.
- We have identified an upturn is c/T. DTO does not exhibit a Pauling entropy when in equilibrium. What is the nature of the ordering at low temperature?
- We will measure the specific heat at lower temperatures (longer times) to determine the peak is c/T. Our dewar will only need to be filled every 20 days (compared to 1 day before).
- We will study the dependence of specific heat characteristic on stuffing.

D. Pomaranski, L. R. Yaraskavitch, S. Meng, K. A. Ross, H. M. L. Noad, H. A. Dabkowska, B. D. Gaulin, and J. B. Kycia, Nature Physics (2013).

H. M. Revell, L. R. Yaraskavitch, J. D. Mason, K. A. Ross, H. M. L. Noad, H. A. Dabkowska, B. D. Gaulin, P. Henelius and J. B. Kycia, Nature Physics, **9** 34 (2013).