Title: Characterization of the Dilute, Dipolar-Coupled, Ising Magent LiHo_xY_{1-x}F4 through Specific Heat and AC Susceptibility Measurements

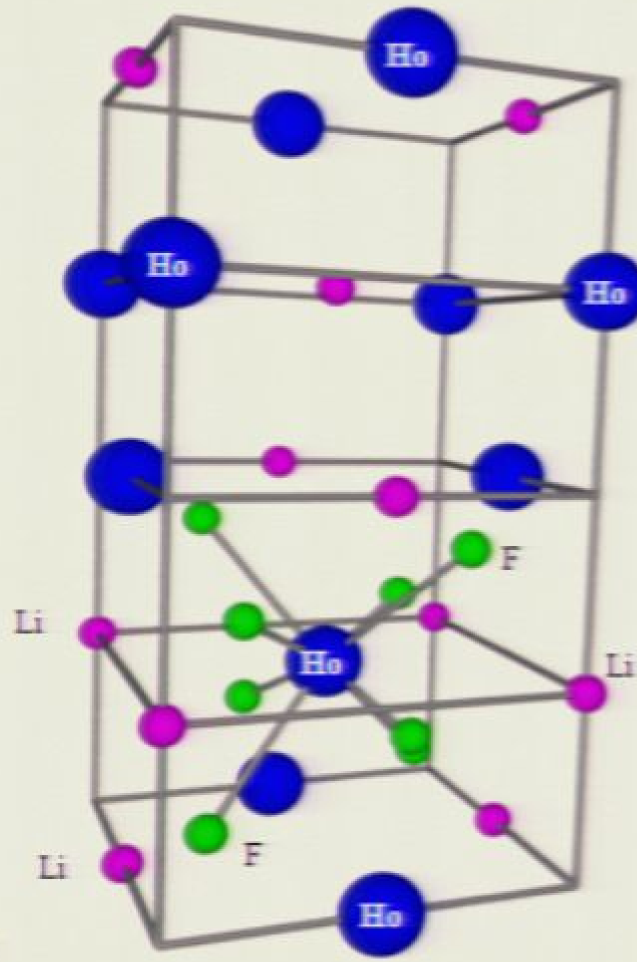
Date: Apr 23, 2009 10:00 AM

URL: http://pirsa.org/09040022

Abstract: <div id="Cleaner">Ising Magent LiHo_xY_{1-x}F4 through Specific Heat and AC Susceptibility Measurements

$LiHo_xY_{1-x}F_4$

- Tetragonal CaWO4 structure
- F^- ions create strong crystal field, makes the Ho^{3+} ions nearly perfect Ising moments along *c*-axis
- Next excited state at ~11 K
- Can replace Ho with non-magnetic Y (dilution)
- Small NN exchange interaction
- Primarily dipolar coupled – angle dependent interaction which leads to frustration in the system.



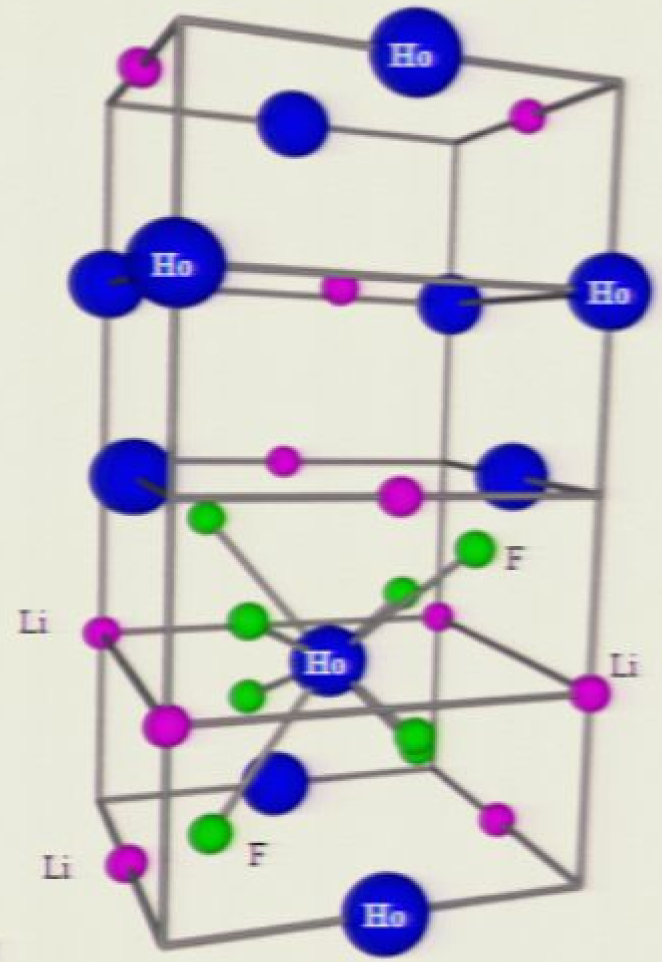
$$\mathcal{H}_{\text{Dipolar}} = \frac{1}{2}\frac{\mu_0}{4\pi}\mu_B^2 g_J^2 \sum_{ij}\left[\frac{\mathbf{J}_i \cdot \mathbf{J}_j}{r_{ij}^3} - \frac{3(\mathbf{J}_i \cdot \mathbf{r}_{ij})(\mathbf{J}_j \cdot \mathbf{r}_{ij})}{r_{ij}^5}\right]$$

$a = b = 5.176$ Å

$c = 10.75$ Å

$LiHo_xY_{1-x}F_4$

- Tetragonal CaWO4 structure
- F^- ions create strong crystal field, makes the Ho^{3+} ions nearly perfect Ising moments along c-axis
- Next excited state at ~11 K
- Can replace Ho with non-magnetic Y (dilution)
- Small NN exchange interaction
- Primarily dipolar coupled – angle dependent interaction which leads to frustration in the system.



$$\mathcal{H}_{\text{Dipolar}} = \frac{1}{2}\frac{\mu_0}{4\pi}\mu_B^2 g_J^2 \sum_{ij}\left[\frac{\mathbf{J}_i \cdot \mathbf{J}_j}{r_{ij}^3} - \frac{3(\mathbf{J}_i \cdot \mathbf{r}_{ij})(\mathbf{J}_j \cdot \mathbf{r}_{ij})}{r_{ij}^5}\right]$$

$a = b = 5.176$ Å

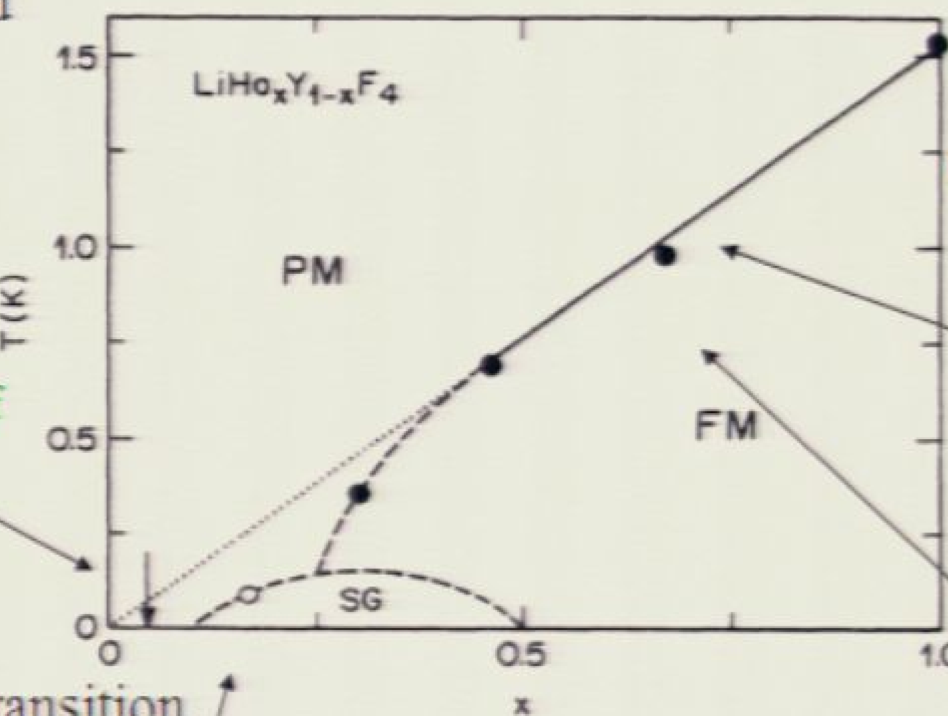
$c = 10.75$ Å

Phase Diagram of $LiHo_xY_{1-x}F_4$

Reich *et al* PRB (1990) (general overview, initial phase diagram)

At x=4.5% unusual "anti-glass", spin liquid state was observed.

Ghosh *et al* Science (2002)
Coherent Spin Oscillations (hole burning)
Ghosh *et al* Nature (2003)
Entangled Quantum State of Magnetic Dipoles



Pure material orders ferromagnetically

Mennenga *et al* JMMM (1984

Lowering x lowers transition temperature (xT_C at first)

Reich *et al* PRB (1990)

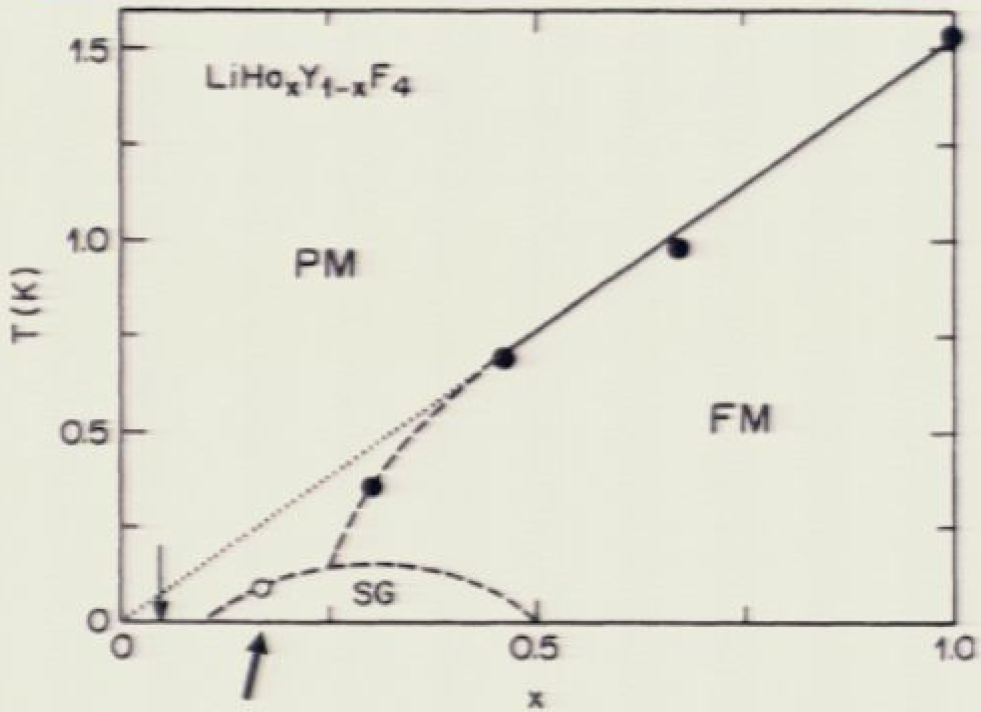
Silevitch et al Nature (2007)
Transverse field provides continuously tunable random field

At x=16% Spin glass transition observed Reich *et al* PRB (1990)

Wu *et al* PRL 1991
(transverse field Tglass = 0)
Wu et al PRL 1993
(transverse field classical to quantum glass transition)

Ancona-Torres *et al* PRL (2008).
Transverse field, quantum and classical glass transitions

Phase Diagram of $LiHo_xY_{1-x}F_4$



At x = 16% debate on whether spin glass state really exists.

Reich et al PRB (1990), Wu *et al* PRL 1991, Wu et al PRL (1993) Yes …Spin Glass

Snider and Yu PRB (2005) (Theory) No ….No Finite temperature Spin Glass

Jonsson *et al* PRL (2007) No … No Finite temperature Spin Glass

Ancona-Torres *et al* PRL (2008) Yes …Spin Glass

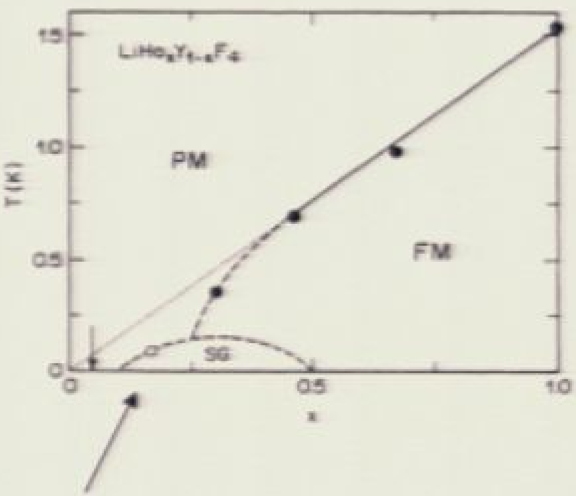
Schechter and Stamp PRB (2008) (Theory) Yes …Spin Glass

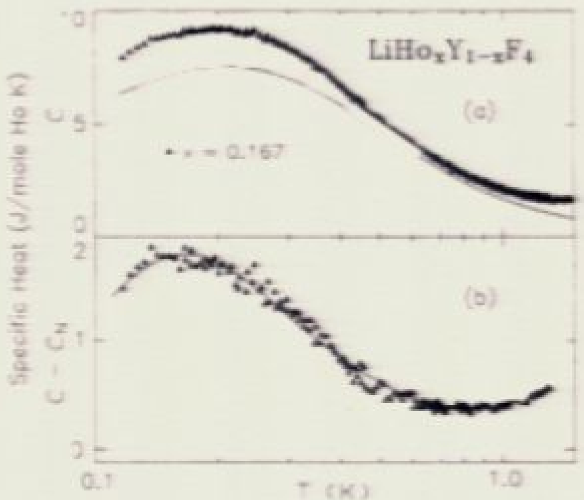
Tam and Gingras arxiv: 0810.085 (2008) (Theory) Yes …Spin Glass

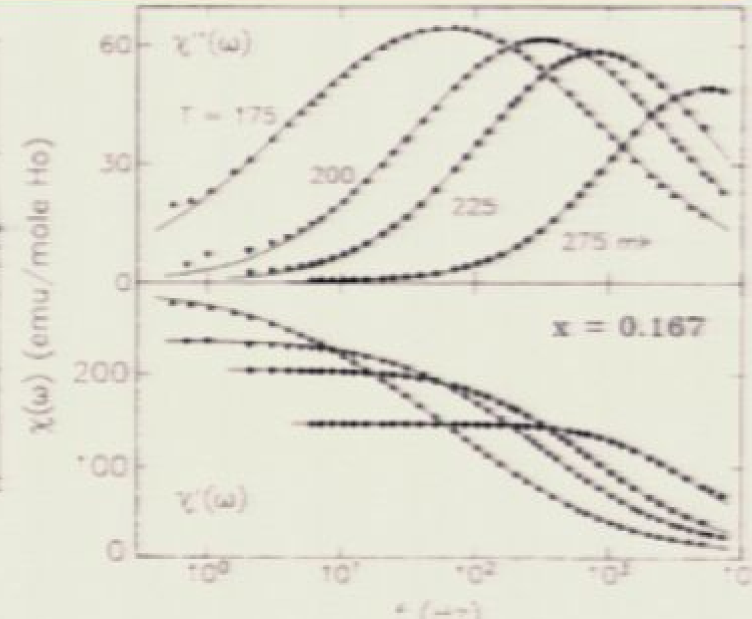
With a debate existing over x = 16% being a spin glass, the state of the lower concentrations maybe even more controversial.

Spin Glass Phase at x = 0.16

Reich *et al* PRB (1990)

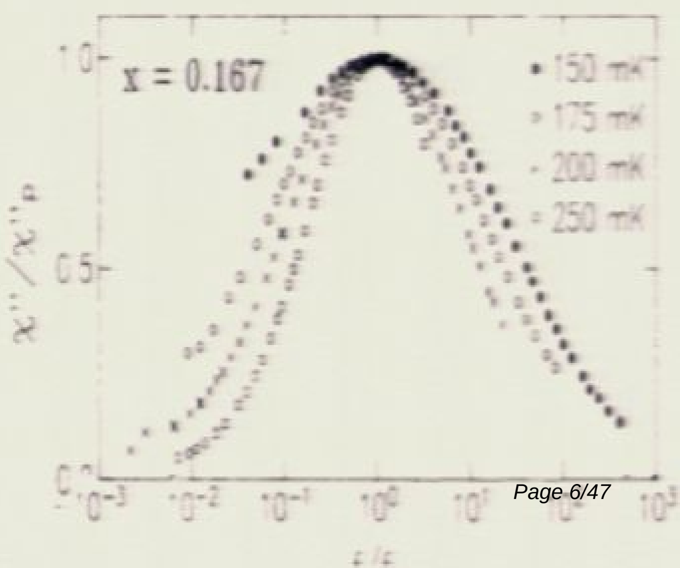






At x=16% spin glass transition observed

•Broadening of χ" as temperature decreases is consistent with approaching glass transition.
•Specific heat characteristics consistent with what is expecte above glass transition



Scaling analyses determines:

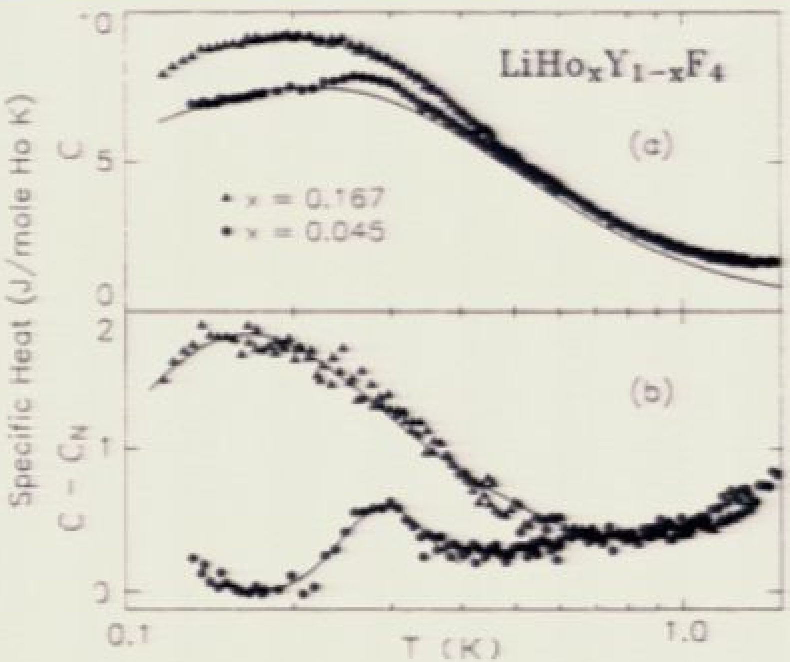
$$\tau_{avg}(T)=\tau_o\left(T/T_g-1\right)^{-z\nu}$$

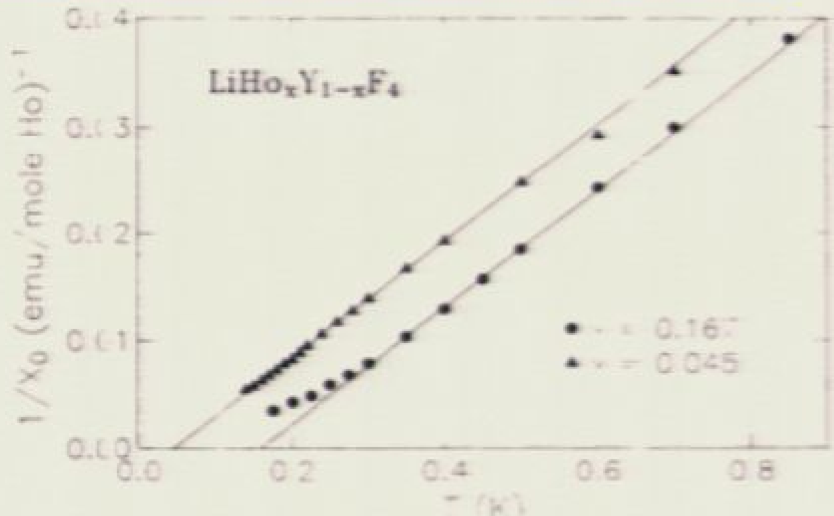
$$T_g=100mK\pm10mK$$

$$z\nu=7.0\pm1.0$$

$$\tau=2.2\times10^{-3}s\pm1.1\times10^{-3}s$$

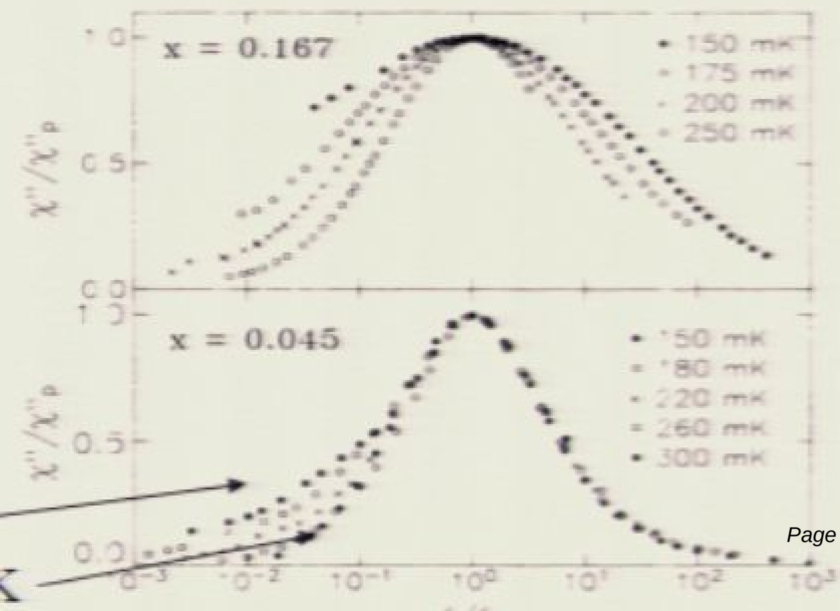
The "Anti-Glass" Phase at x = 0.045   Reich *et al* PRB (1990)





DC Susceptibility has
1/T temperature dependence.

Unusually sharp features
in the specific heat.



Narrowing of absorption
spectrum χ''(f) with lower *T*
(opposite of a spin glass).

150 mK

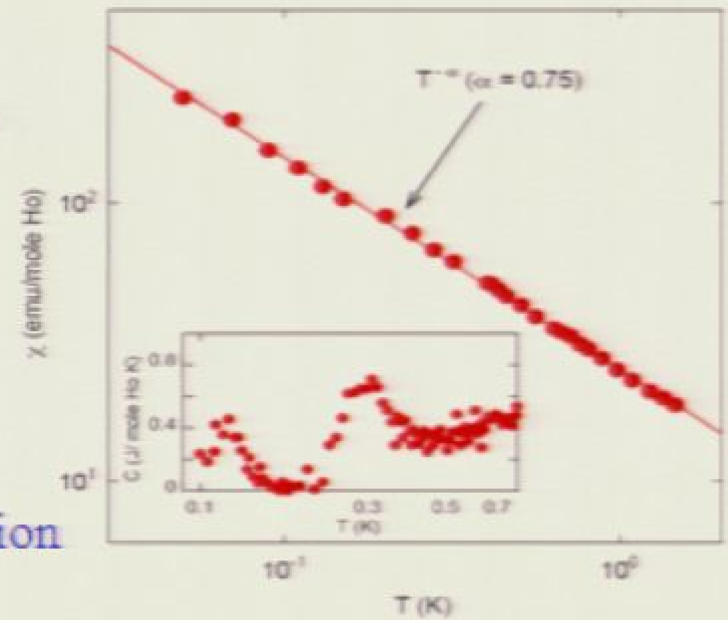
300 mK

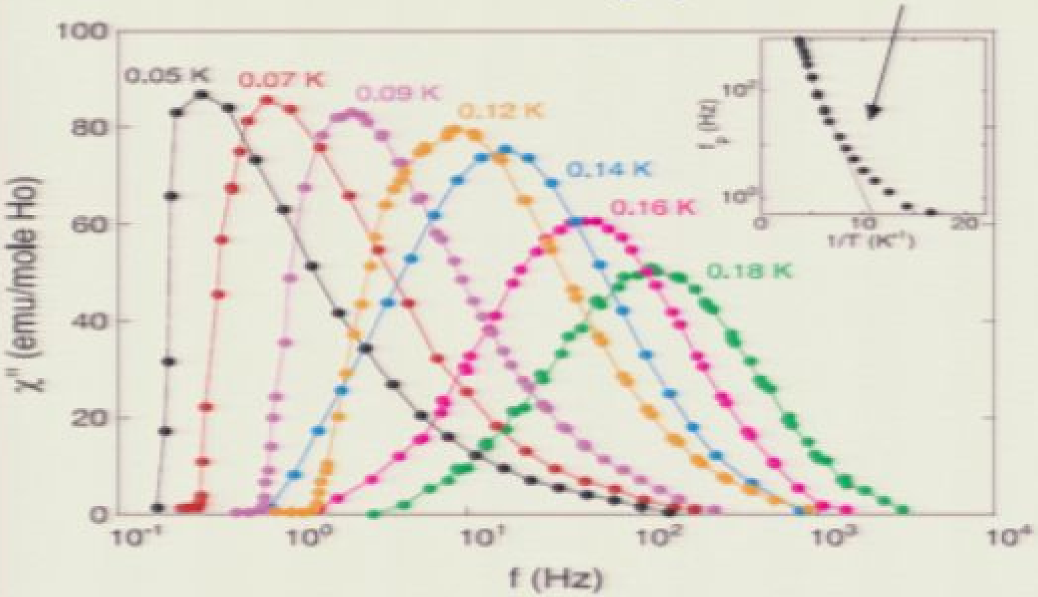
The "Anti-Glass" Phase at x = 0.045

Ghosh *et al* Science (2002)
Ghosh *et al* Nature (2003)

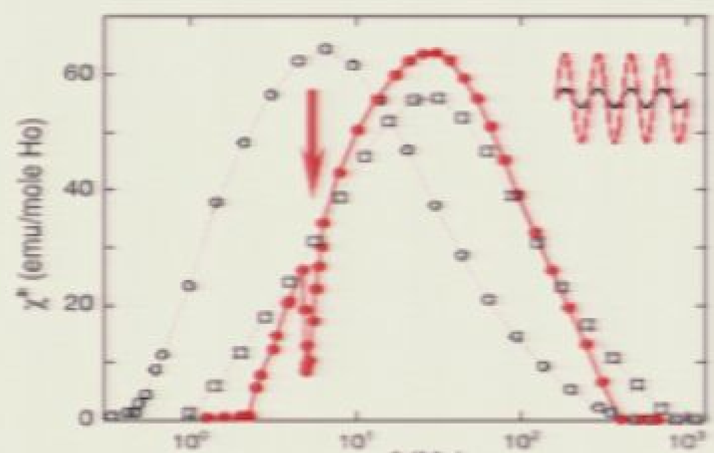
- Remeasured DC susceptibility, found $\chi \propto T^{-0.75}$ instead of $\chi \propto T^{-1}$.
- Point out how unusual it is that the specific heat has peaks while susceptibility is smooth.
-- explain this with entanglement
- Measure χ'' to be even more asymmetric at low temperatures.

Peak frequency temperature dependence deviates from Arrhenius behaviour indicating Quantum –Classical transition





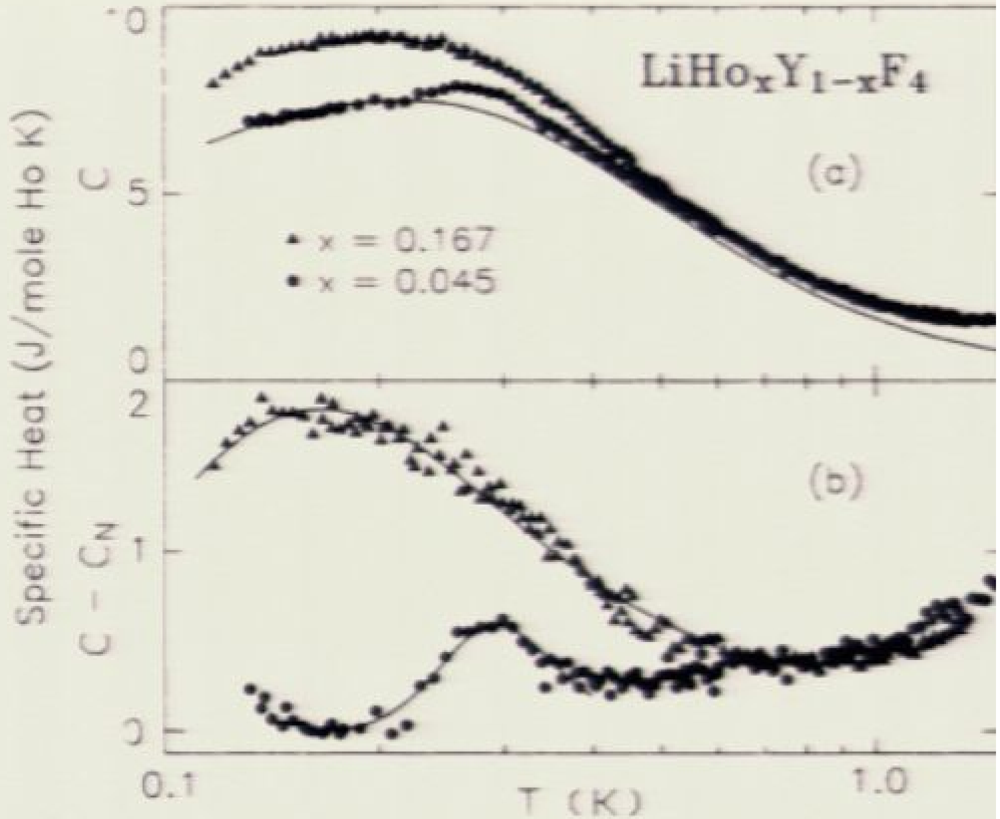
T = 110 mK, transverse 5 Hz ac field



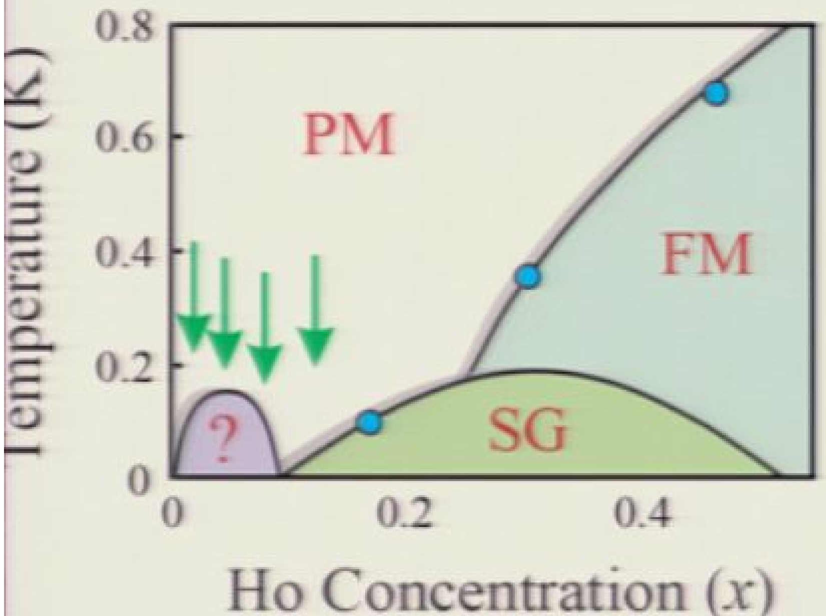
See coherent spin oscillations with lifetime of 10 seconds.
Demonstrate hole burning and calculate that this is due to coherent spin oscillations of clusters of 260 spins

Our first goal was to measure the low temperature specific heat of $LiHo_xY_{1-x}F_4$

- More accurately
- Lower temperatures
- Different Ho concentrations



Reich *et al.* PRB 42, 4631 (1990).
Mennenga *et al.* JMMM 44, 59 (1984).

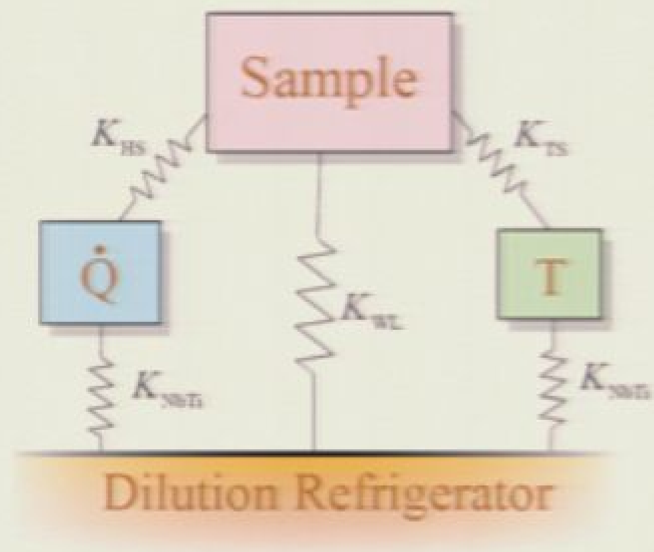


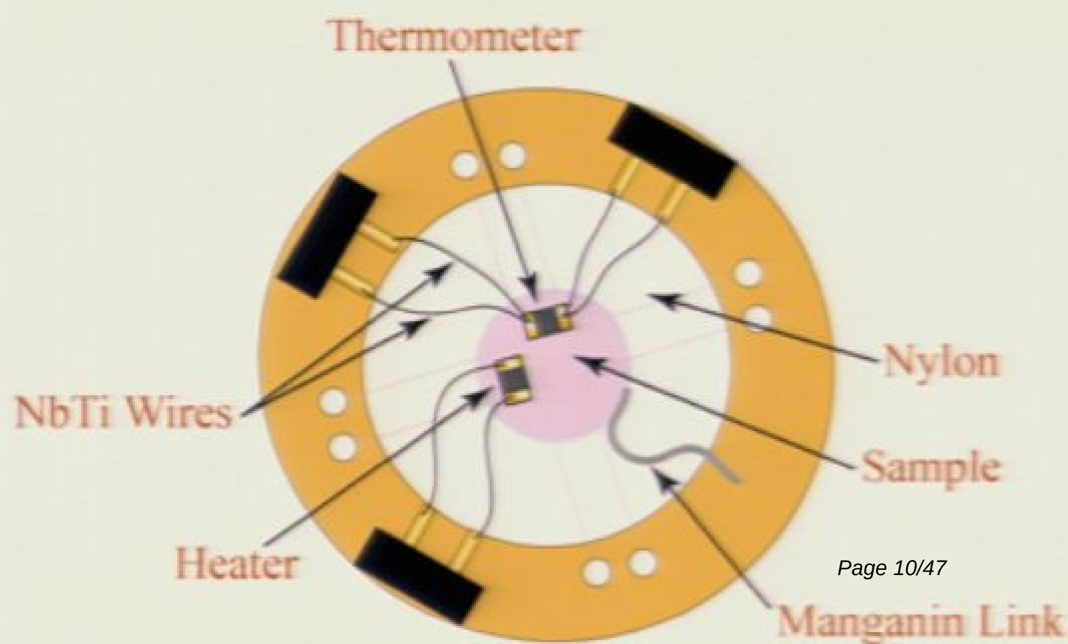
- Subtraction of Ho Nuclear term is tricky
- 16.7% Ho sample looks like spin glass
- 4.5% Ho sample looks like "anti-glass"

Arrows indicate samples that we have, purchased from TYDEX J.S. Co., St. Petersburg, Russia

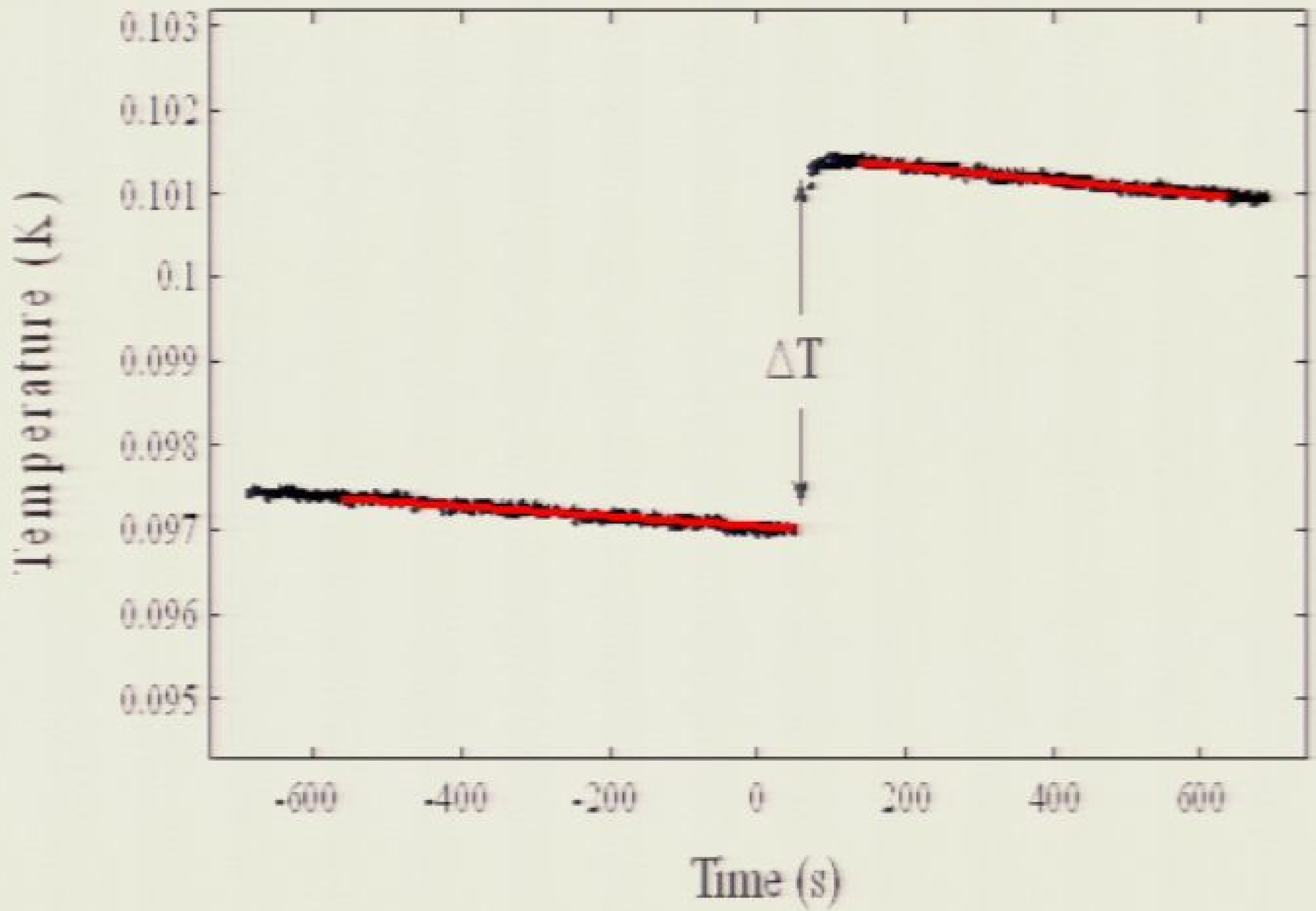
Heat Capacity Measurement

- Dilution Refrigerator with 13 mK base temperature
- Used quasi-adiabatic method – heat pulse Q is applied and ΔT is measured
- Careful attention was paid to thermal leaks, decoupling of thermometers, etc.
- Leads are 6 μm diameter, 1cm long superconducting wires (conduct very little heat).
- No substrate used (components fastened directly to sample)
- RuO_2 resistance thermometer calibrated to a GRT and CMN thermometer.

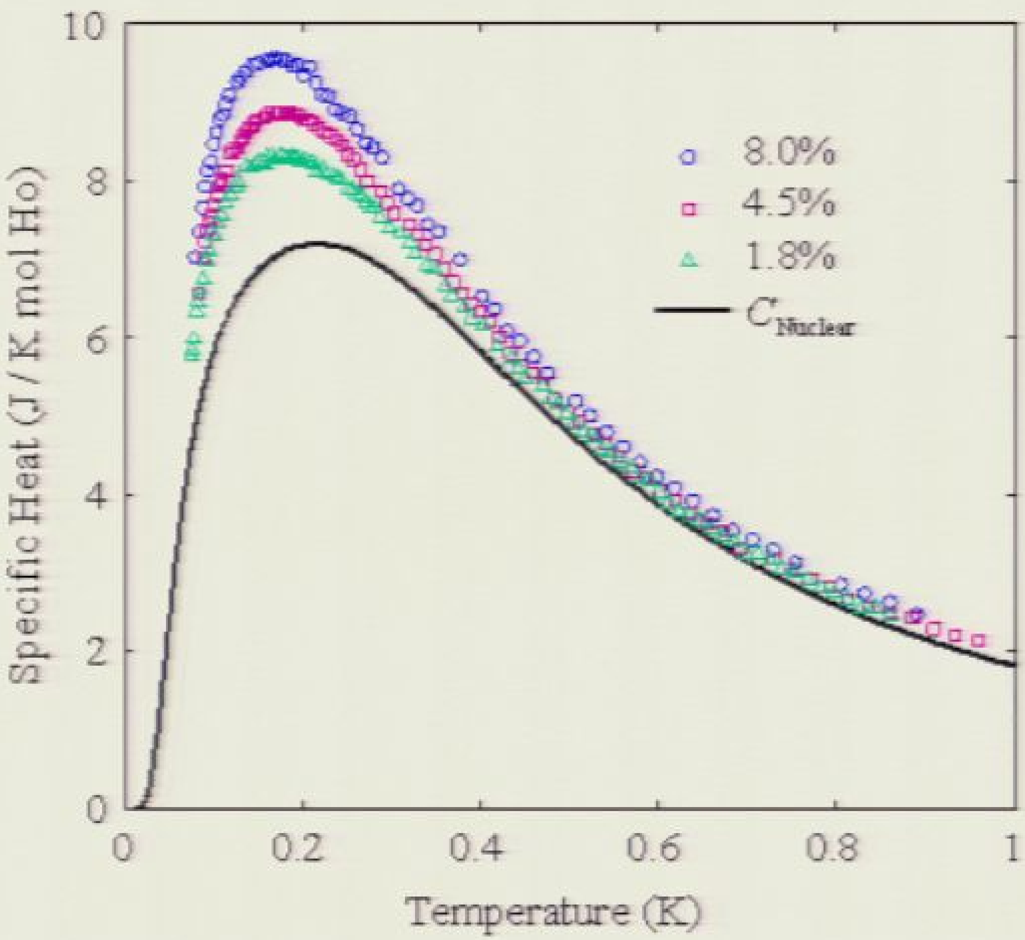




Typical data for a single heat pulse



Total Specific Heat



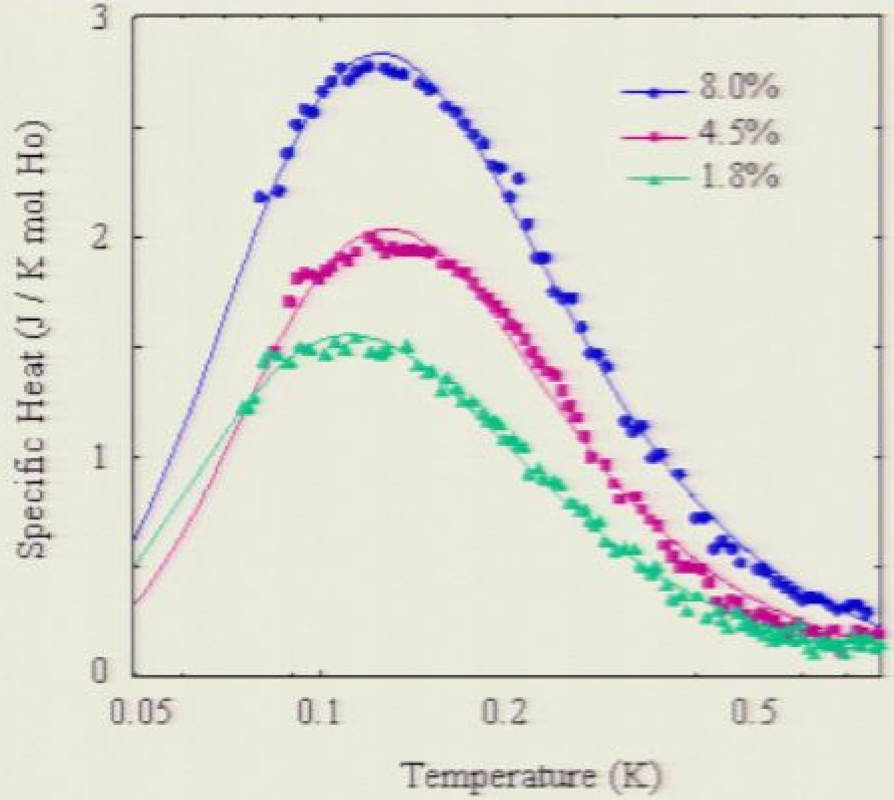
- Total specific heat is dominated by nuclear term
- Ho nuclei have 7/2 spin, strong hyperfine interaction with tightly bound 4f electrons
- Non-interacting C_N calculated from crystal field, hyperfine interaction and nuclear quadrupole interaction.
- Very small phonon term ($\sim T^3$) present as well.

After Subtraction of Nuclear Specific Heat

- Non-interacting C_N subtracted to give electronic part ΔC
- Broad feature remains which is consistent with a spin glass for all 3 samples
- Spin glass C does not have a sharp feature at T_0
- Indicative of excitations above the transition
- Simplest model: 1 excited energy level with degeneracy n w.r.t. ground state (fits)

$$\Delta C \propto \frac{(E_1/kT)^2 e^{-E_1/kT}}{(1+ne^{-E_1/kT})^2}$$

- More low-temperature data required to look for linear temperature dependence



Parameter	1.8% sample	4.5% sample	8.0% sample	16.7% sample [1]
C_0 (J/K mol Ho)	4.06	3.45	7.31	2.85
E_1/k_B (K)	0.26	0.32	0.29	0.46
n	0.85	1.43	0.86	1.89
$T_{\rm peak}$ (K)	0.11	0.13	0.12	0.17
FWHM/$T_{\rm peak}$	1.7	1.6	1.7	1.5
S_0/R	0.31	0.21	0.00	0.18

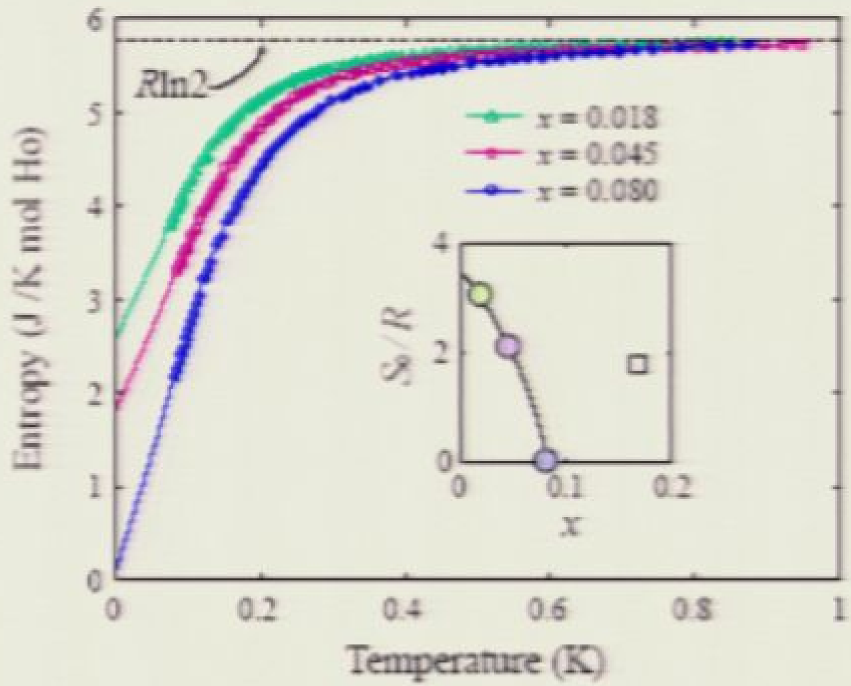
[1] Reich *et al.* PRB 42, 4631 (1990).

Residual Entropy?

- Entropy can be determined with numerical integral

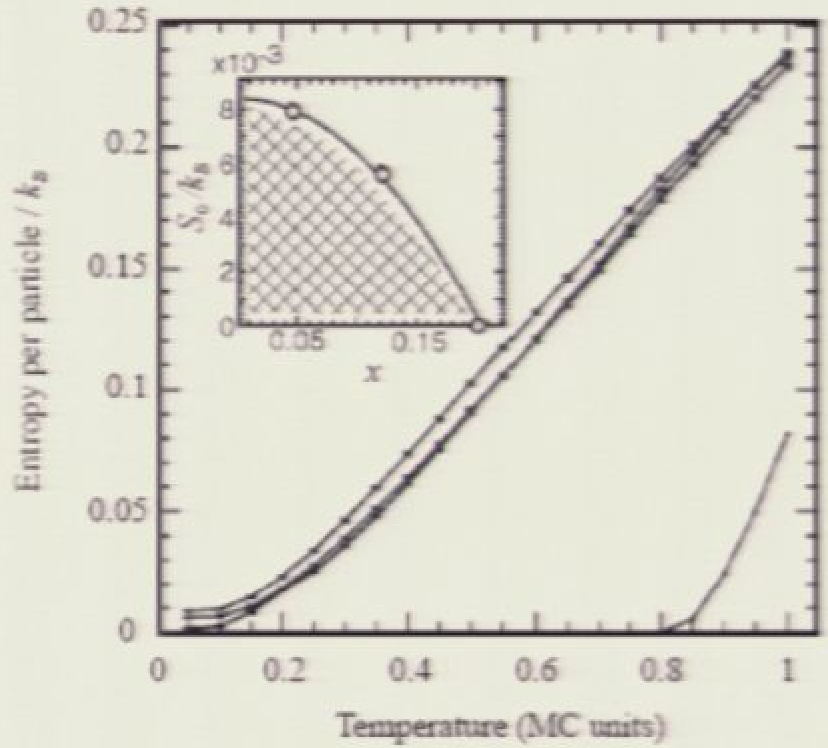
$$S(T) = \int_0^T dT \frac{\Delta C(T)}{T}$$

- Total entropy should be $R \ln 2$
- Possibility of residual entropy S_0
- Integral done here with linear extrapolation to $T = 0$
- S_0 is increasing with decreasing x



Residual entropy agrees qualitatively with Snider and Yu, PRB 72, 214203 (2005)

- Increase in residual entropy with lower x is consistent with Monte Carlo simulations of Snider and Yu :
  - Ising spins on a simple-cubic lattice
  - No spin glass order below 20% filling
  - $S_0 = 0$ at $x_c = 0.20$ and rises below
  - Smooth $S(T) \Rightarrow$ broad bump in C
- Different lattice, no nuclear moments

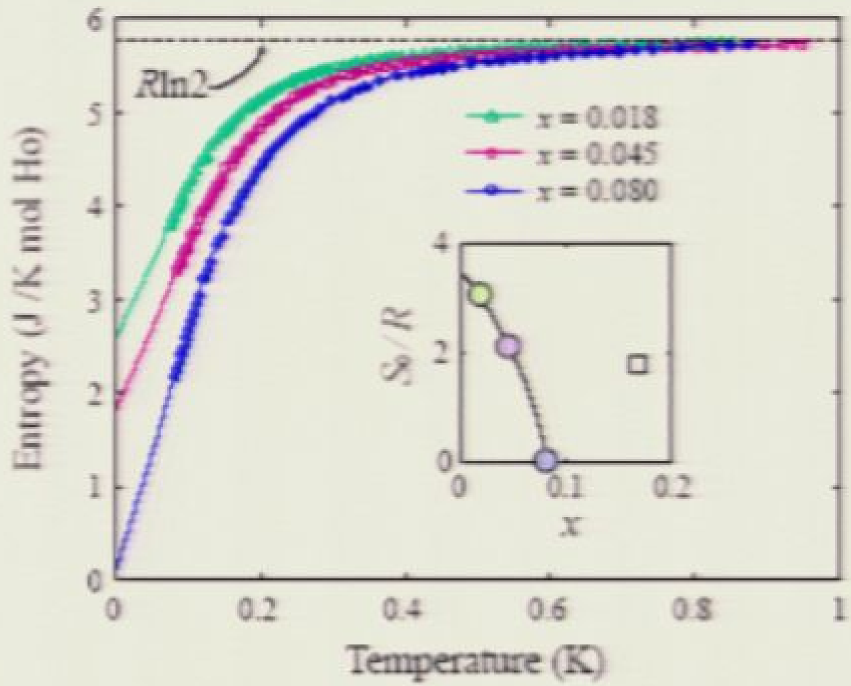


Residual Entropy?

- Entropy can be determined with numerical integral

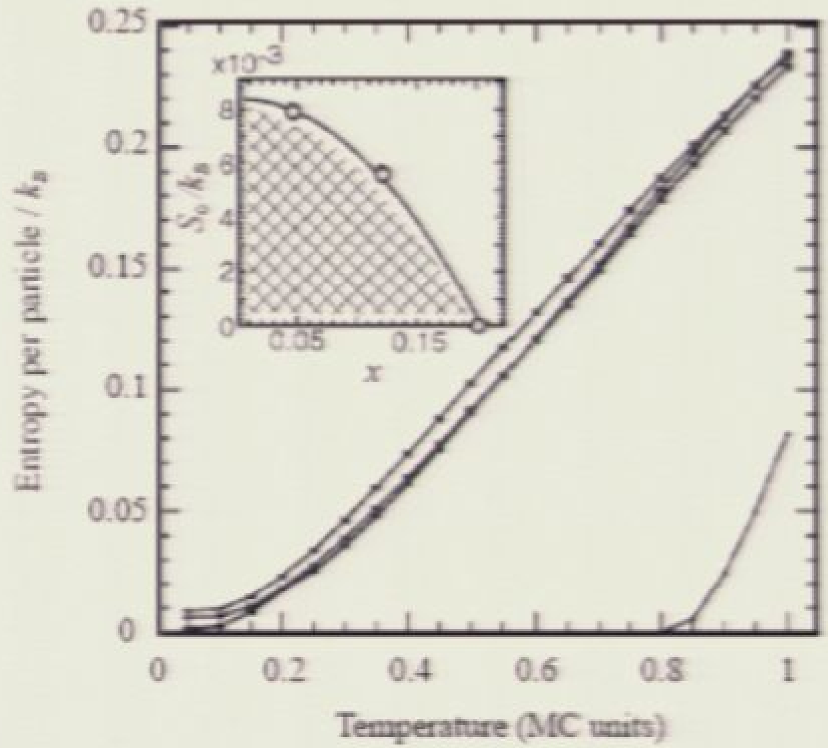
$$S(T) = \int_0^T dT \frac{\Delta C(T)}{T}$$

- Total entropy should be $R \ln 2$
- Possibility of residual entropy S_0
- Integral done here with linear extrapolation to $T = 0$
- S_0 is increasing with decreasing x



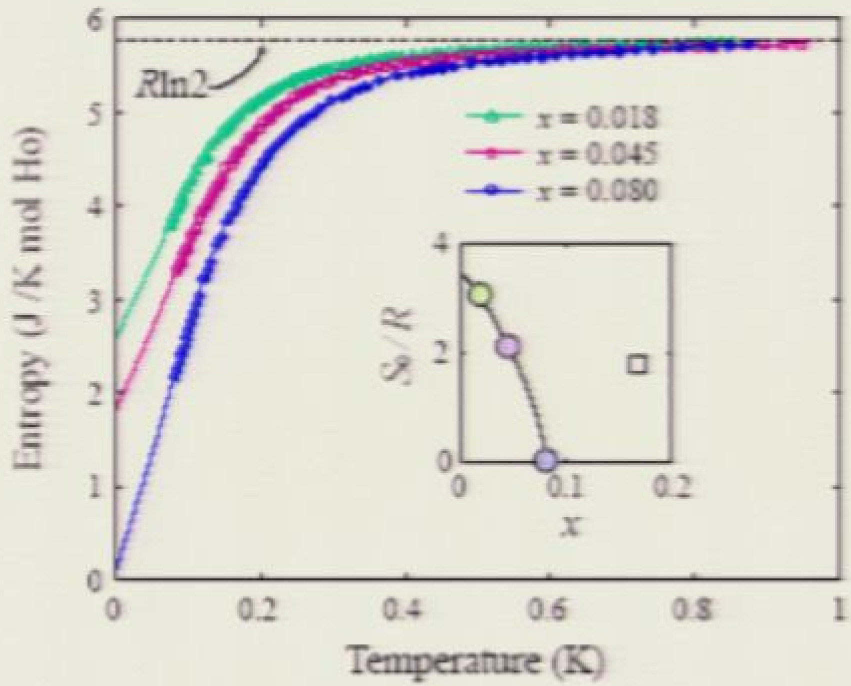
Residual entropy agrees qualitatively with Snider and Yu, PRB 72, 214203 (2005)

- Increase in residual entropy with lower x is consistent with Monte Carlo simulations of Snider and Yu :
  - Ising spins on a simple-cubic lattice
  - No spin glass order below 20% filling
  - $S_0 = 0$ at $x_c = 0.20$ and rises below
  - Smooth $S(T) \Rightarrow$ broad bump in C
- Different lattice, no nuclear moments



Residual Entropy?

- Entropy can be determined with numerical integral
$$S(T) = \int_0^T dT \frac{\Delta C(T)}{T}$$
- Total entropy should be $R \ln 2$
- Possibility of residual entropy S_0
- Integral done here with linear extrapolation to $T = 0$
- S_0 is increasing with decreasing x

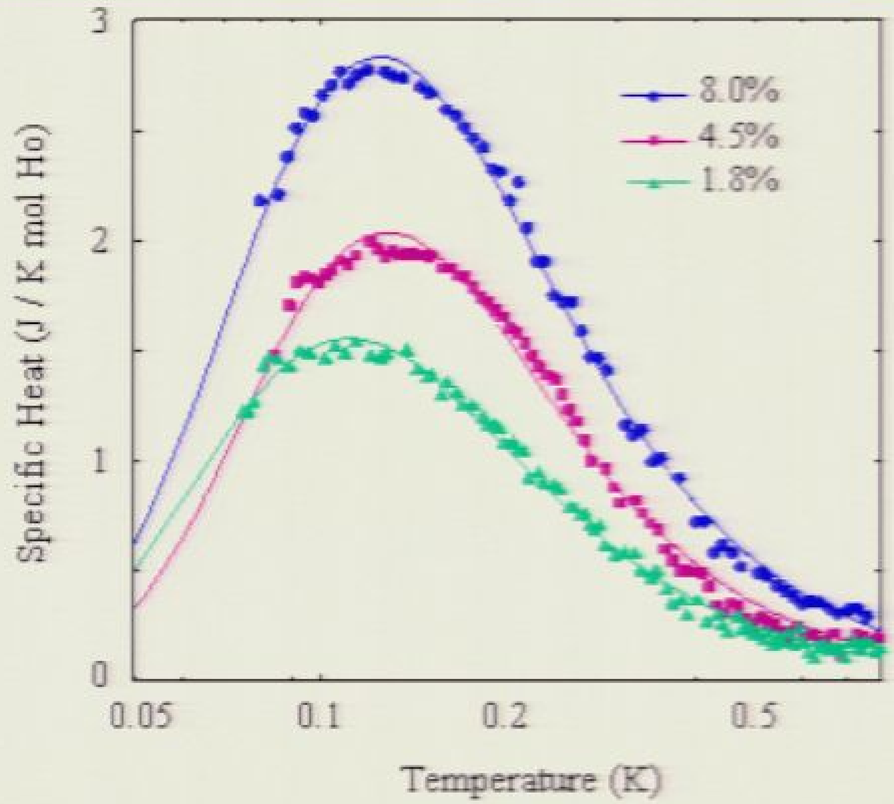


After Subtraction of Nuclear Specific Heat

- Non-interacting C_N subtracted to give electronic part ΔC
- Broad feature remains which is consistent with a spin glass for all 3 samples
- Spin glass C does not have a sharp feature at T_0
- Indicative of excitations above the transition
- Simplest model: 1 excited energy level with degeneracy n w.r.t. ground state (fits)

$$\Delta C \propto \frac{(E_1/kT)^2 e^{-E_1/kT}}{(1+ne^{-E_1/kT})^2}$$

- More low-temperature data required to look for linear temperature dependence



Parameter	1.8% sample	4.5% sample	8.0% sample	16.7% sample [1]
C_0 (J/K mol Ho)	4.06	3.45	7.31	2.85
E_1/k_B (K)	0.26	0.32	0.29	0.46
n	0.85	1.43	0.86	1.89
T_{peak} (K)	0.11	0.13	0.12	0.17
FWHM/T_{peak}	1.7	1.6	1.7	1.5
S_0/R	0.31	0.21	0.00	0.18

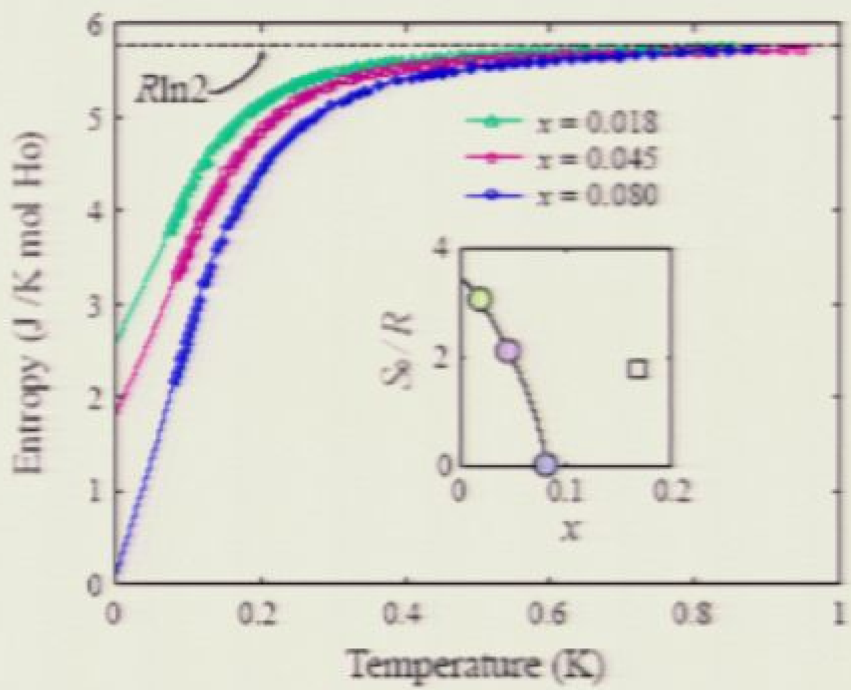
[1] Reich *et al.* PRB 42, 4631 (1990).

Residual Entropy?

- Entropy can be determined with numerical integral

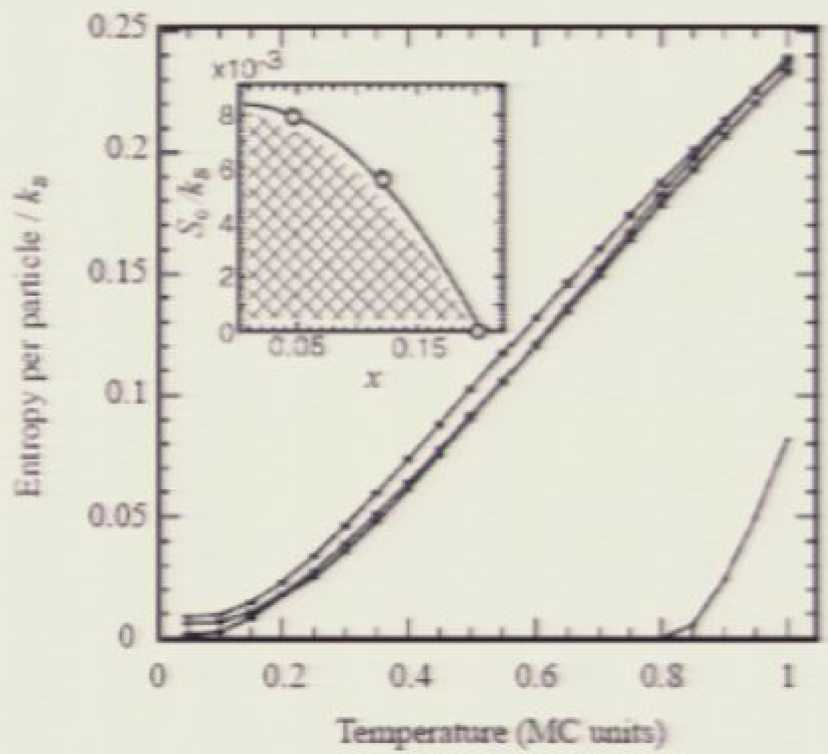
$$S(T) = \int_0^T dT \frac{\Delta C(T)}{T}$$

- Total entropy should be $R \ln 2$
- Possibility of residual entropy S_0
- Integral done here with linear extrapolation to $T = 0$
- S_0 is increasing with decreasing x



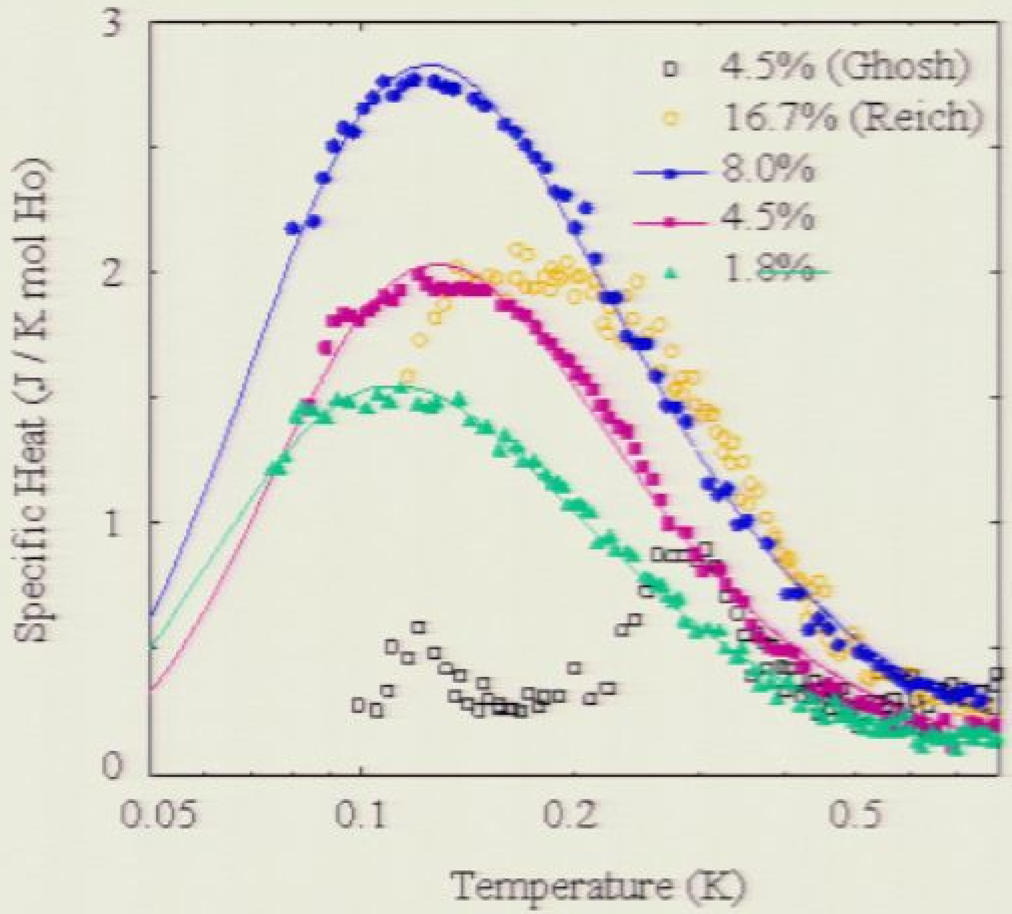
Residual entropy agrees qualitatively with Snider and Yu, PRB 72, 214203 (2005)

- Increase in residual entropy with lower x is consistent with Monte Carlo simulations of Snider and Yu :
  - Ising spins on a simple-cubic lattice
  - No spin glass order below 20% filling
  - $S_0 = 0$ at $x_c = 0.20$ and rises below
  - Smooth $S(T) \Rightarrow$ broad bump in C
- Different lattice, no nuclear moments



Comparison with Previous Results

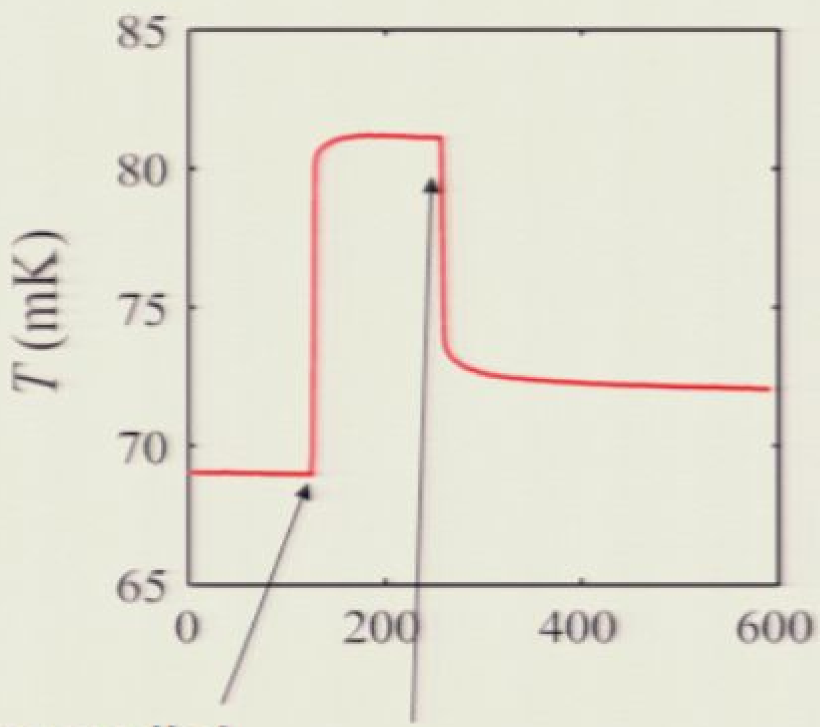
- Our results do not reproduce the unusually sharp features observed by Ghosh *et al*. in 4.5% Ho:YLF
- Thermal conductivity of 4% sample also saw no sharp features (Nikkel & Ellman CondMat 0504269)
- Data is qualitatively consistent with the 16.7% sample measured by Reich *et al*.
- We account for much more of the expected entropy in the system (*R*ln2)
- Heat capacity does not give us a measure of the dynamics of the system so cannot say whether "anti-glass" or not.



Reich *et al*. PRB 42, 4631 (1990)
Ghosh *et al*. Science 296, 2195 (2002)

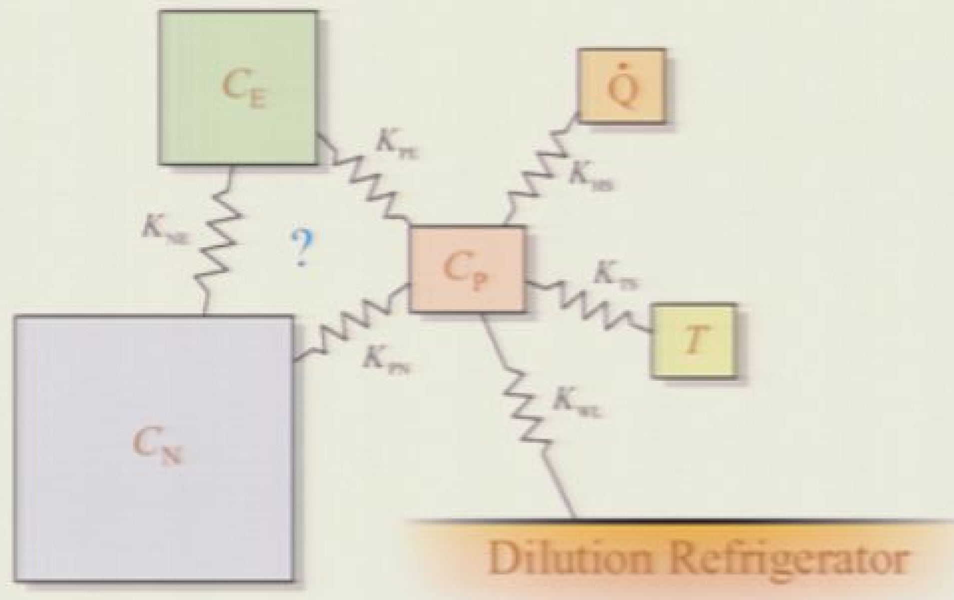
Specific heat at temperatures below 100 mK

- We find a decoupling of the lattice and phonons from the main source of specific heat.
- At low temperature it is as if we were using the substrate configuration.

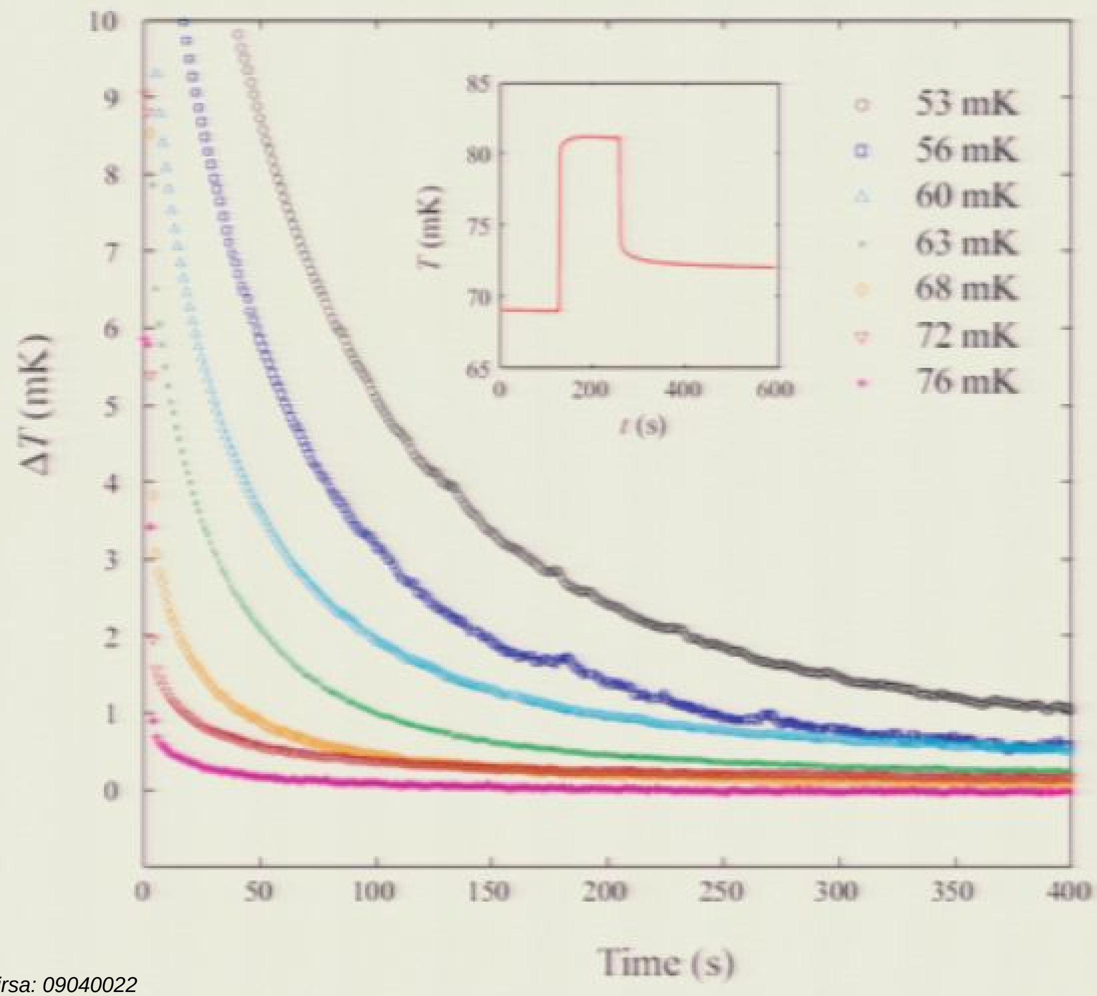


Heat applied

Heat turned off



Preliminary temperature dependence of the decoupling at low temperatures



1.8% Ho

The relaxation time maybe goes as:
$\tau = C_{lattice} / K_{lattice\text{-}nuclei}$

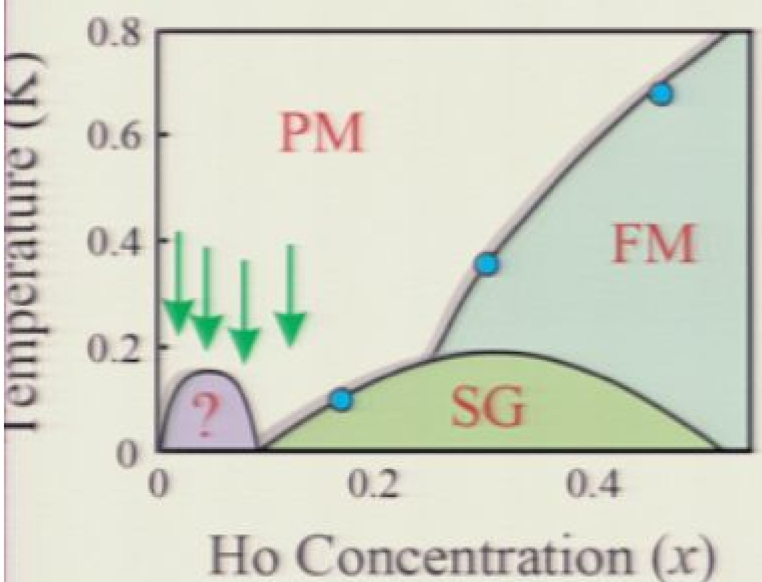
Conclusions for Specific Heat Measurement

- Measured specific heat of x = 0.018, 0.045 and 0.080 Ho samples
- Do not reproduce sharp features in specific heat seen by Ghosh et al. in the x =0.045 sample.
- All have qualitative behavior of the x = 0.0167 sample measured by Reich *et al.*
- A residual entropy may exist for the x=0.018 and 0.045 concentrations, that or the temperature dependence of the low temperature specific heat is sub-linear in temperature.
- Unusual that peak in specific heat does not move to lower temperatures as concentration is reduced (problem with estimation and subtraction of the nuclear term?)
- Observe significant decoupling of the lattice specific heat from the electrons and/or nuclear components below 100 mK.
- Our specific heat work has been published:

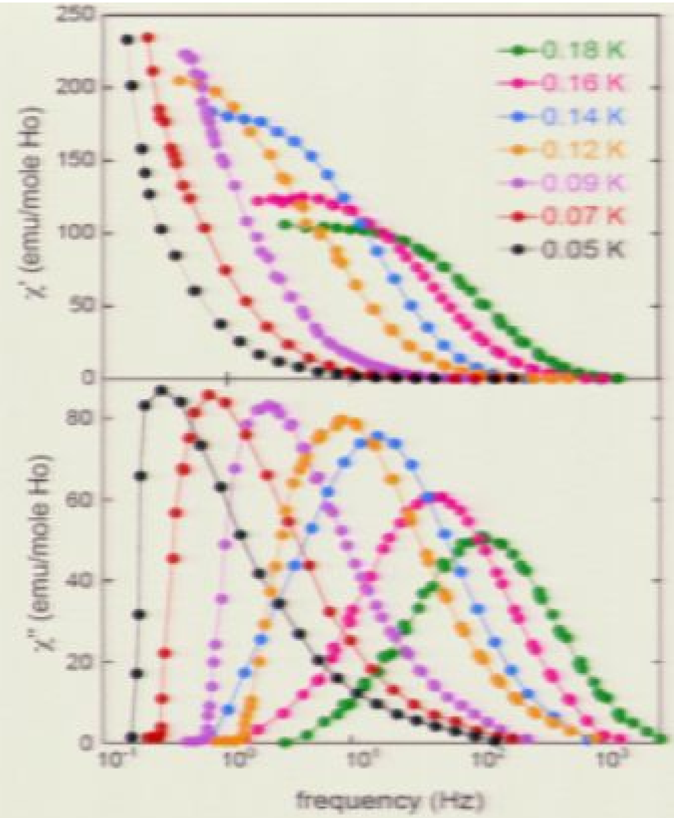
"Specific Heat of the Dilute Ising Magnet $LiHo_xY_{1-x}F_4$"
J.A. Quilliam, C.G.A. Mugford, A. Gomez, S.W. Kycia, and J.B. Kycia
Phys Rev Lett. **98,** 037203 (2007).

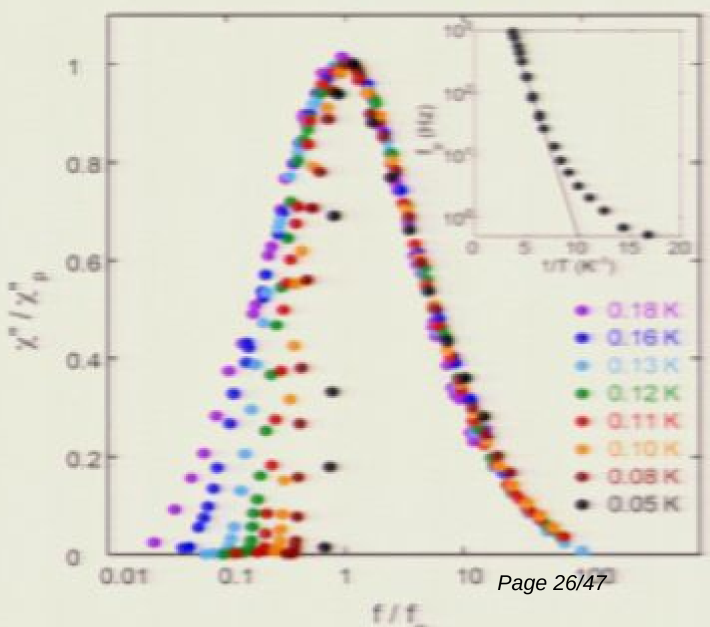
Motivation for More Susceptibility Measurements on $LiHo_xY_{1-x}F_4$

- Use SQUID for improved performance at low frequencies.
- Confirm that x=0.045 sample has anti-glass characteristics. Check connection of specific heat characteristics with susceptibility characteristics for antiglass.
- Study different Ho concentrations x = 0.018, x = 0.08



Arrows indicate samples that we have, purchased from TYDEX J.S. Co., St. Petersburg, Russia





Conventional Susceptometer

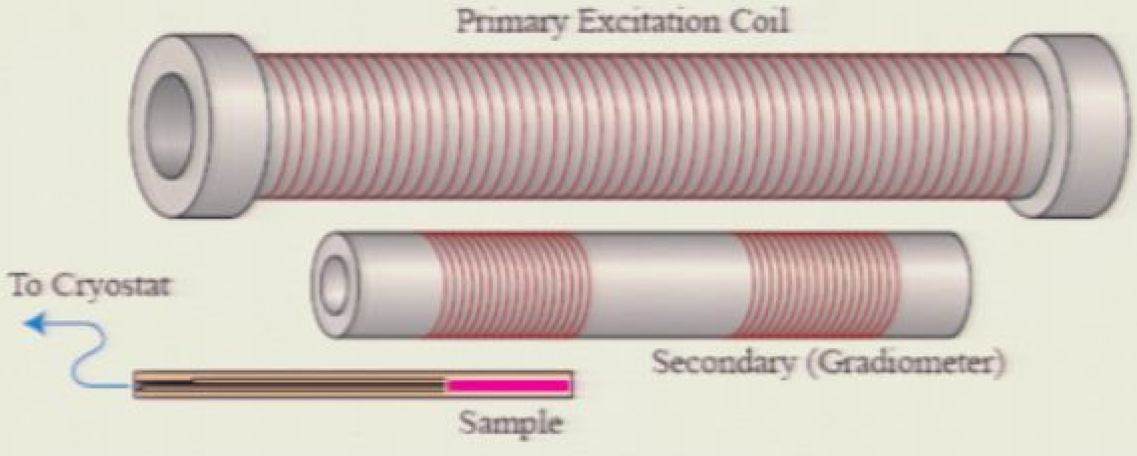
Advantage: Easy to put together and use.

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

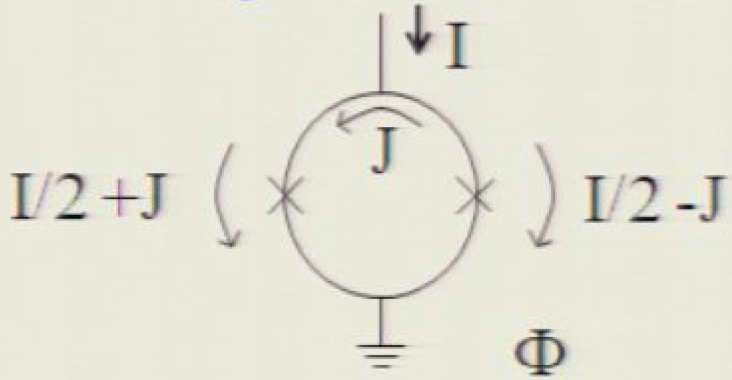
$$V_{EMF} \propto \frac{d\phi}{dt} \propto \omega$$

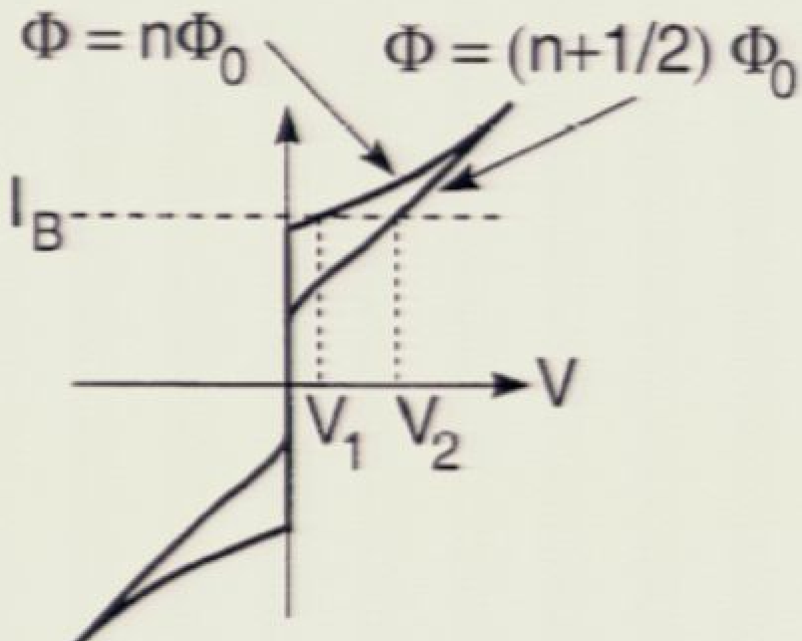
Too many turns reduces highest useable frequency due to intercoil resonance.

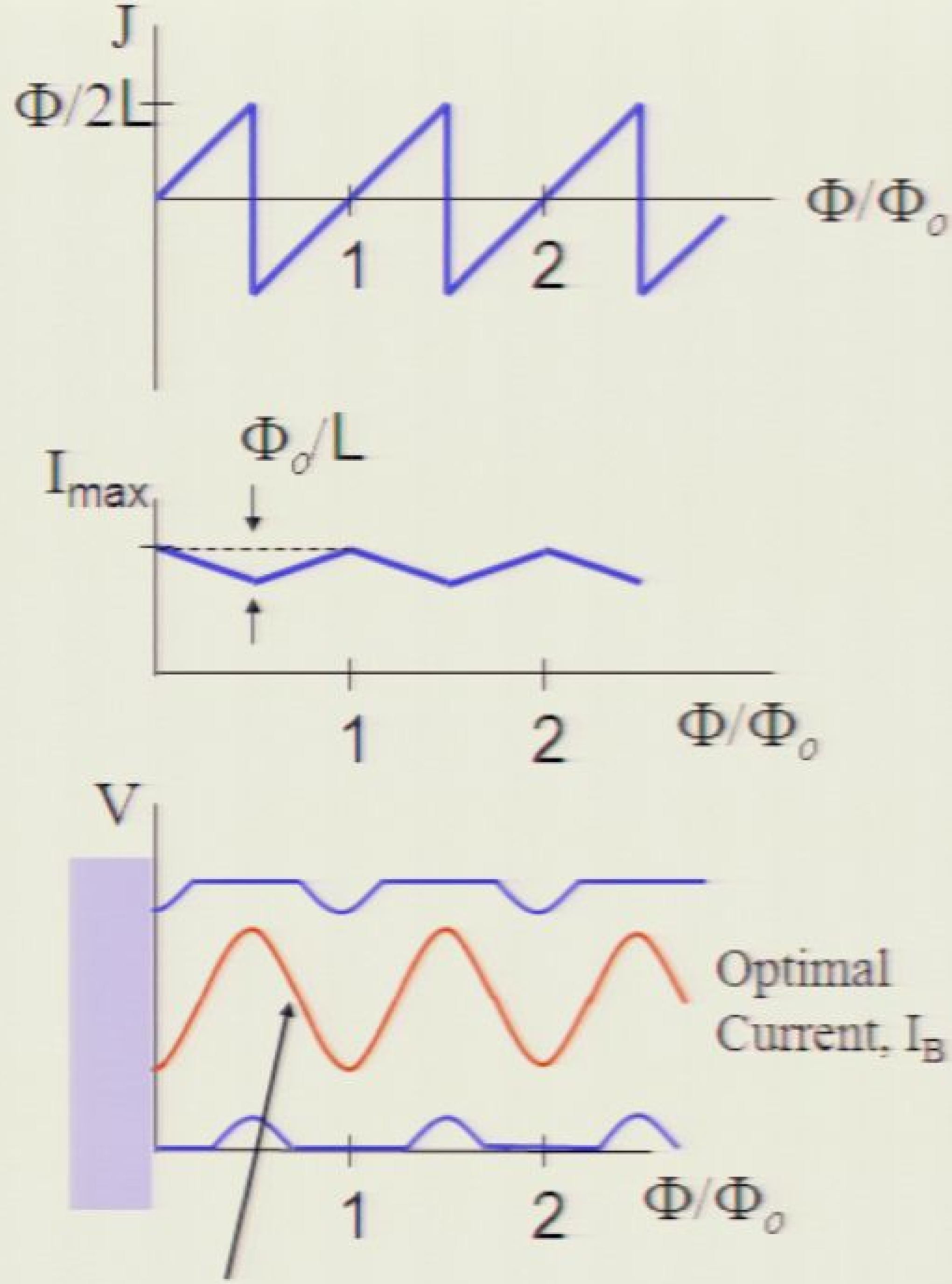
---Phase shifts and non-flat frequency response.



The DC SQUID is the most sensitive detector of magnetic flux







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

Optimal operating point

While our experiment was in progress, Jonnson *et al* remeasured χ_1 and χ_3 for x = 0.045 and x = 0.165 using a micro-SQUID. They swept the field at rates from 1 to 50 Oe/s.

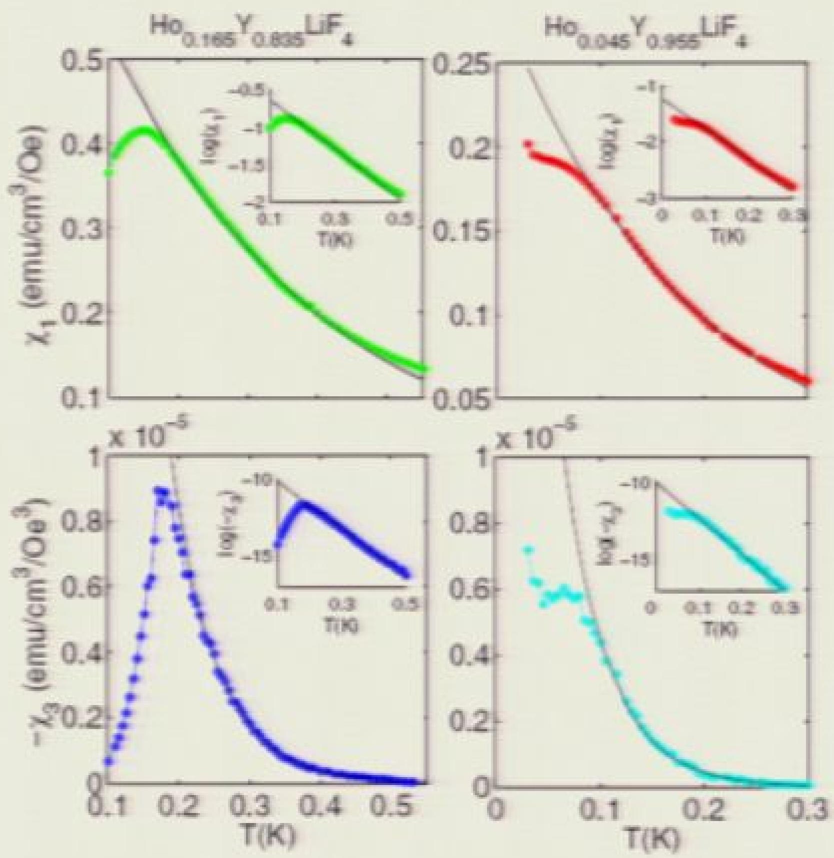
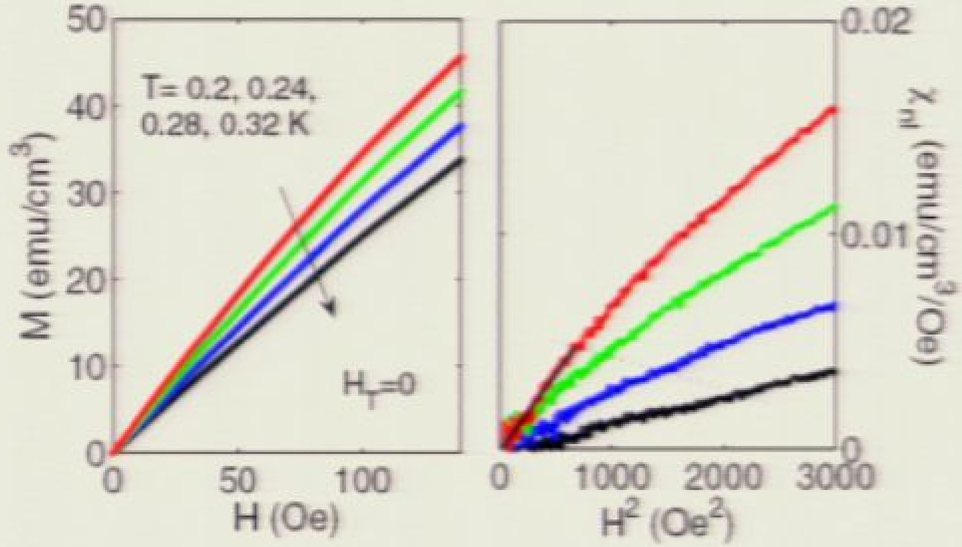


FIG. 4 (color online). Main frames: Temperature dependence of the linear and nonlinear susceptibilities χ_1 and χ_3 for (left) $LiHo_{0.165}Y_{0.835}F_4$ and (right) $LiHo_{0.165}Y_{0.835}F_4$ without transverse field. The continuous lines represent $\exp(-T/T_0)$ fits (see main text), obtained from the T-linear fits of $\log(\chi_1)$) and $\log(\chi_3)$) shown in the insets of each panel.



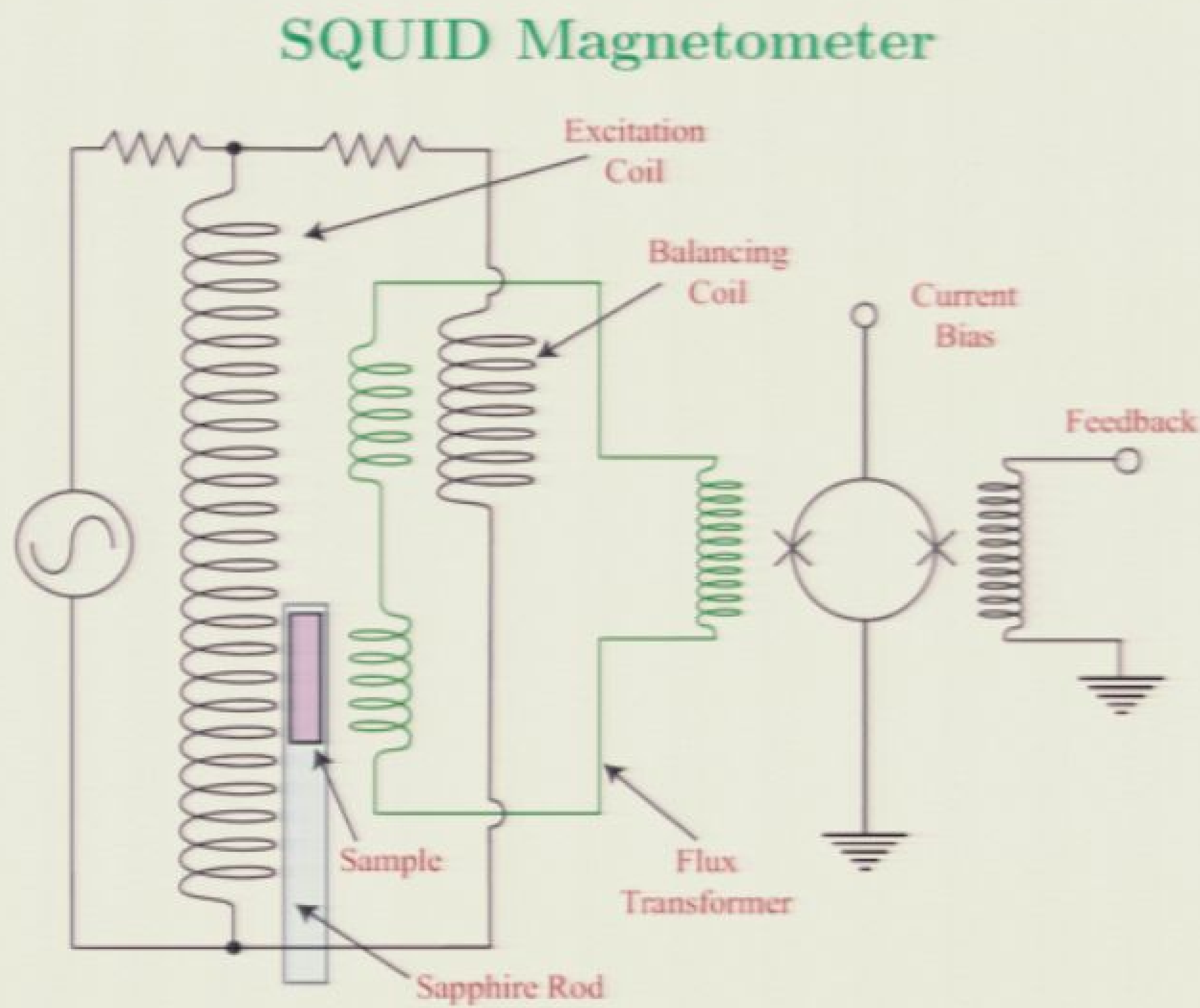
Conclude absence of spin glass transition for both x=0.045 and x = 0.165.
Since both compositions are qualitatively similar, they **question the existence of an antiglass phase for x = 0.045.**

Ancona Torres ***et al*** **disagree** and claim Jonnsen *et al* swept to their field to quickly at low temperatures.

Jonnson *et al* PRL (2007)
Ancona Torres *et al* PRL (2008)

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.**



SQUID and controller from ez-SQUID

While our experiment was in progress, Jonnson *et al* remeasured χ_1 and χ_3 for x = 0.045 and x = 0.165 using a micro-SQUID. They swept the field at rates from 1 to 50 Oe/s.

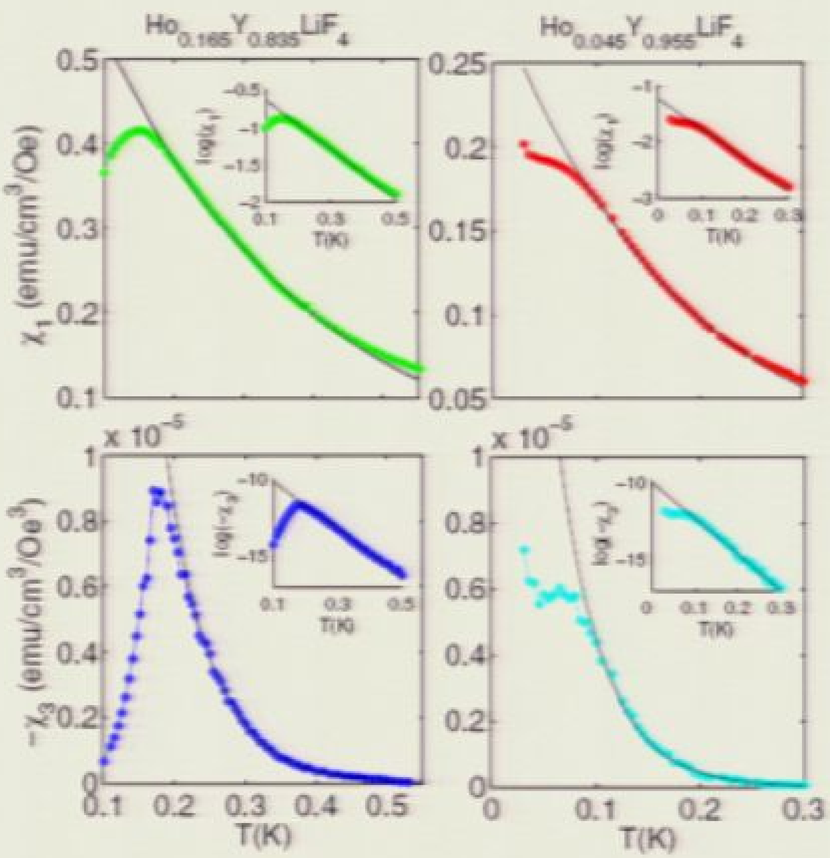
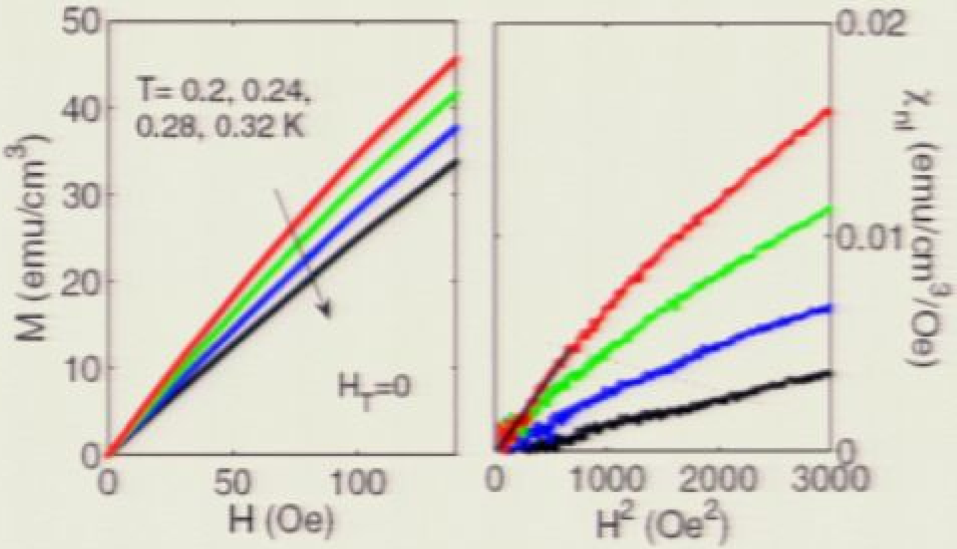


FIG. 4 (color online). Main frames: Temperature dependence of the linear and nonlinear susceptibilities χ_1 and χ_3 for (left) $LiHo_{0.165}Y_{0.835}F_4$ and (right) $LiHo_{0.165}Y_{0.835}F_4$ without transverse field. The continuous lines represent $\exp(-T/T_0)$ fits (see main text), obtained from the T-linear fits of $\log(\chi_1)$) and $\log(\chi_3)$) shown in the insets of each panel.



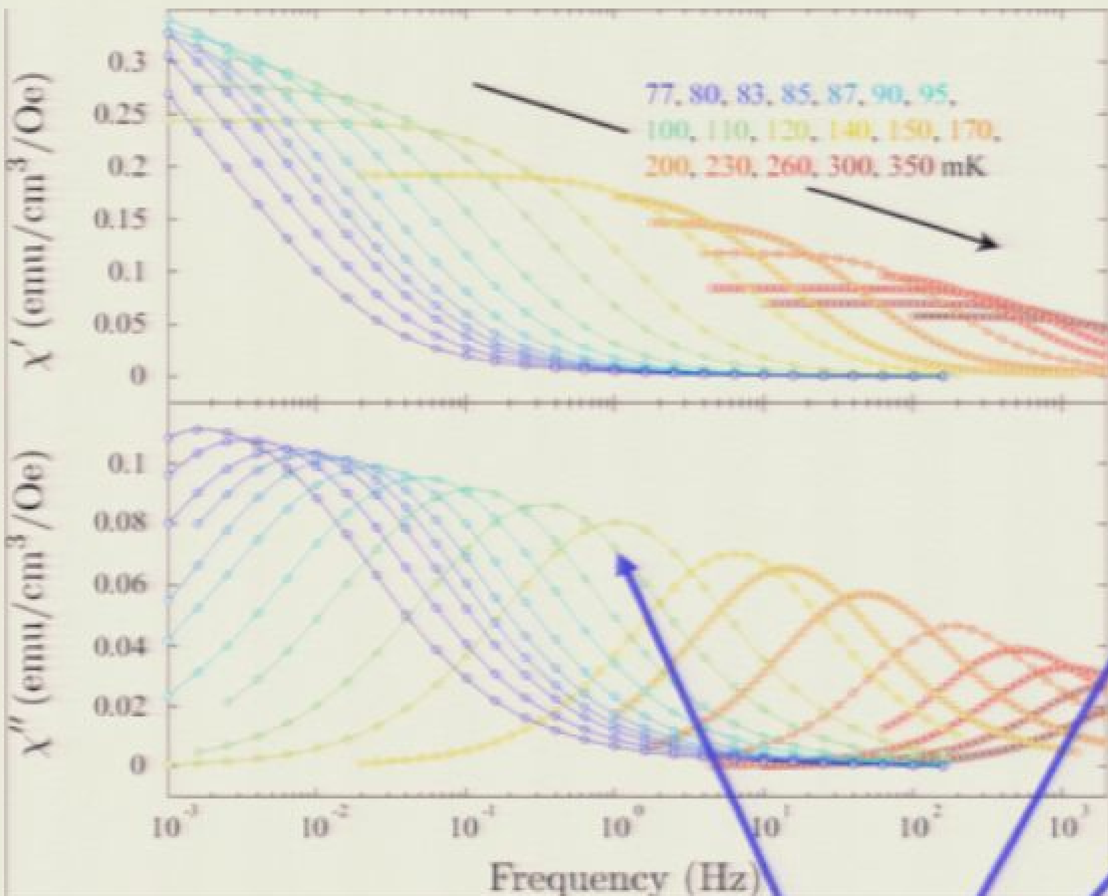
Conclude absence of spin glass transition for both x=0.045 and x = 0.165.
Since both compositions are qualitatively similar, they **question the existence of an antiglass phase for x = 0.045.**

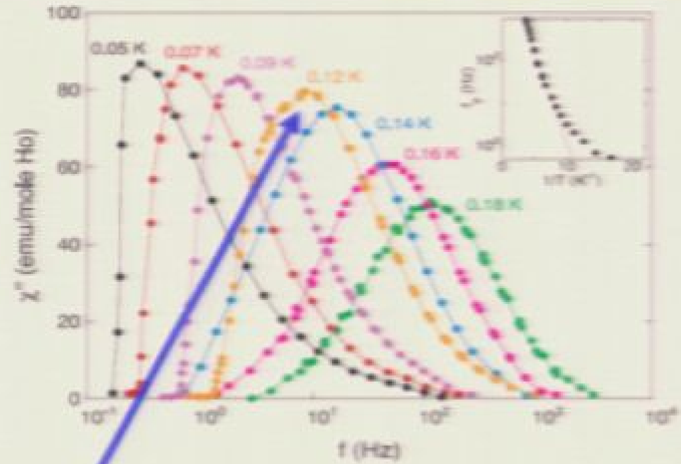
Ancona Torres *et al* disagree and claim Jonnsen *et al* swept to their field to quickly at low temperatures.

Jonnson *et al* PRL (2007)

ac Susceptibility for Various Temperatures, x = 0.045

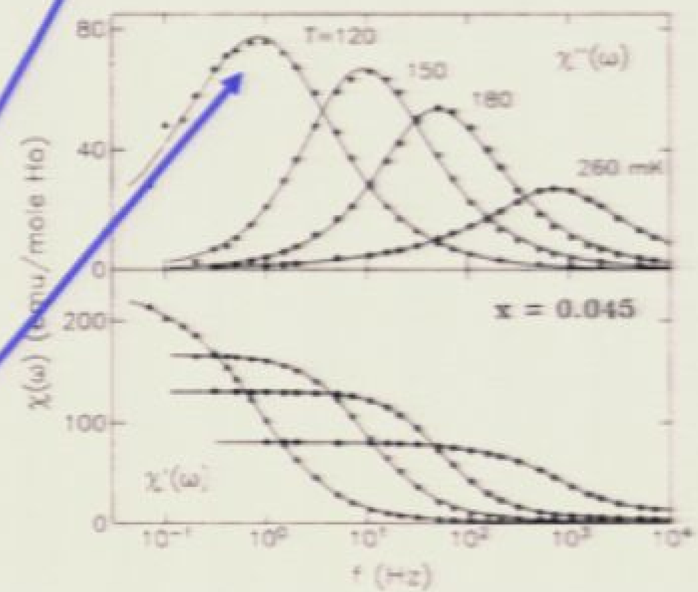
Our Result:





Ghosh *et al*. Science (2002)

T =120 mK

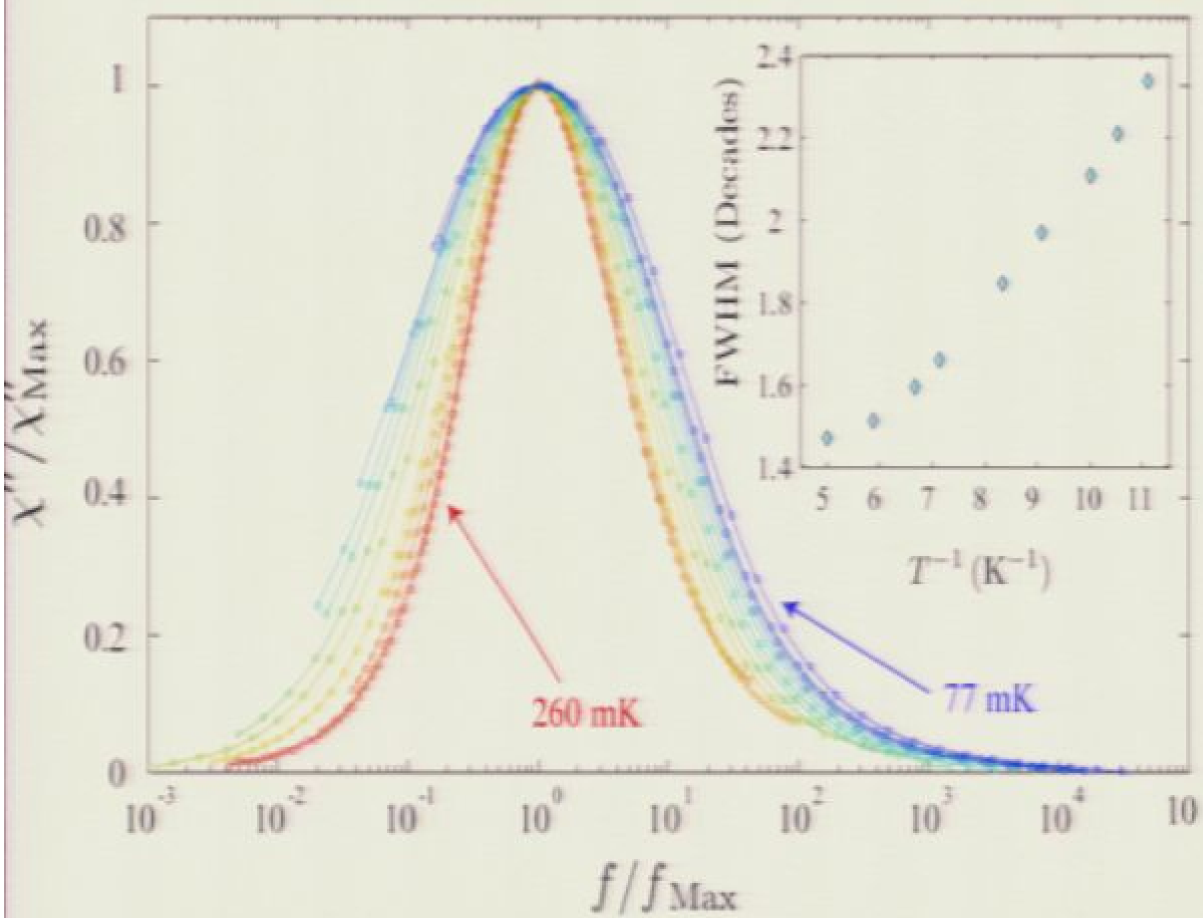


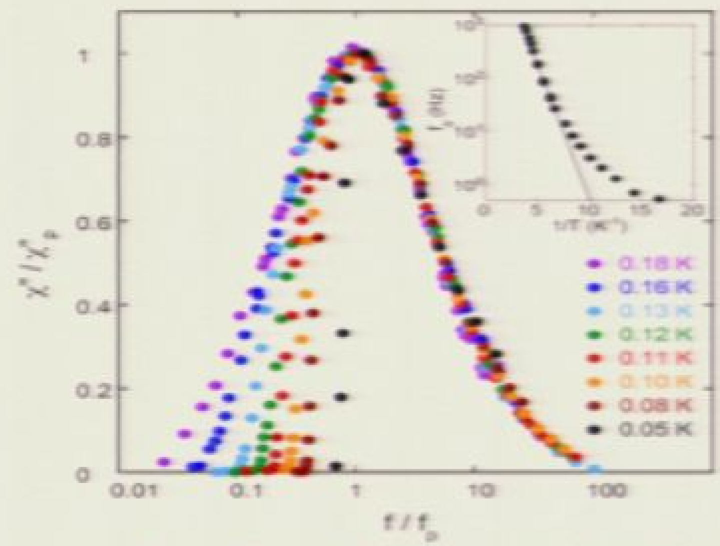
Reich et al. PRB (1990)

Our data shows slower response than Ghosh *et al* for a given temperature. Agrees better with Reich *et al*.

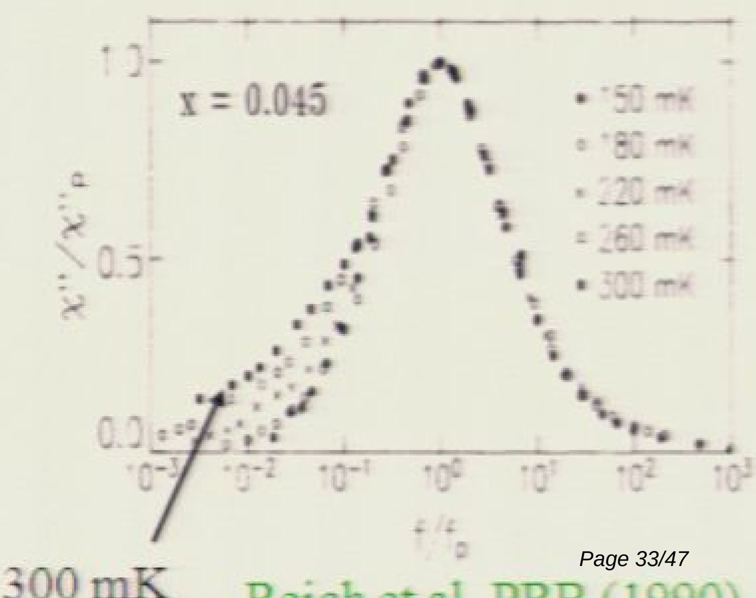
Width of χ" for Various Temperatures

Our Result:





Ghosh *et al*. Science (2002)



Reich et al. PRB (1990)

Our χ" broadens as temperature decreases.
Consistent with a spin glass.
Not consistent with antiglass.

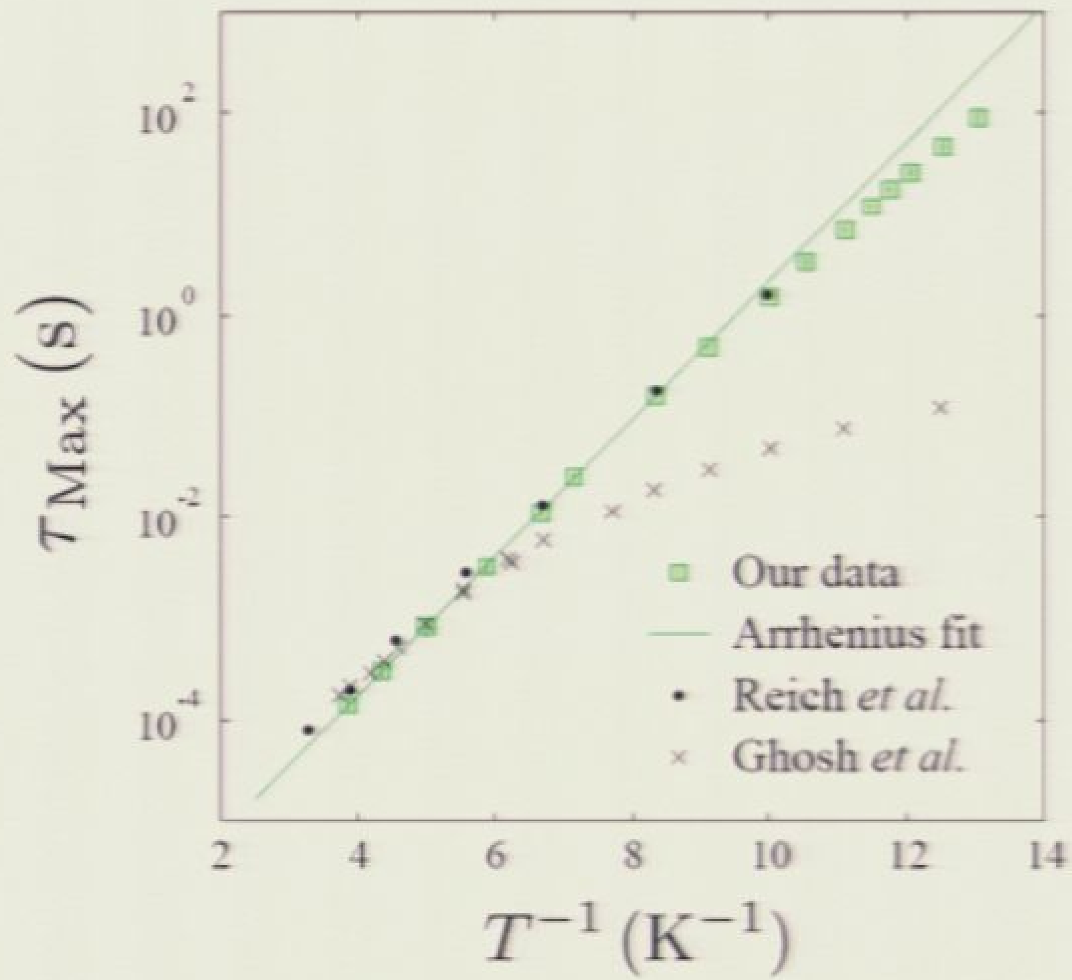
Fit to Arrhenius Law

τ_{Max} is the determined from the frequency of the peak in χ''(f) for a given temperature, T.

$$\tau_{Max}(T) = \tau_{oA} e^{-E_a/k_B T}$$

$$E_A = 1.57K$$

$$\tau_{oA} = 0.32\mu s$$

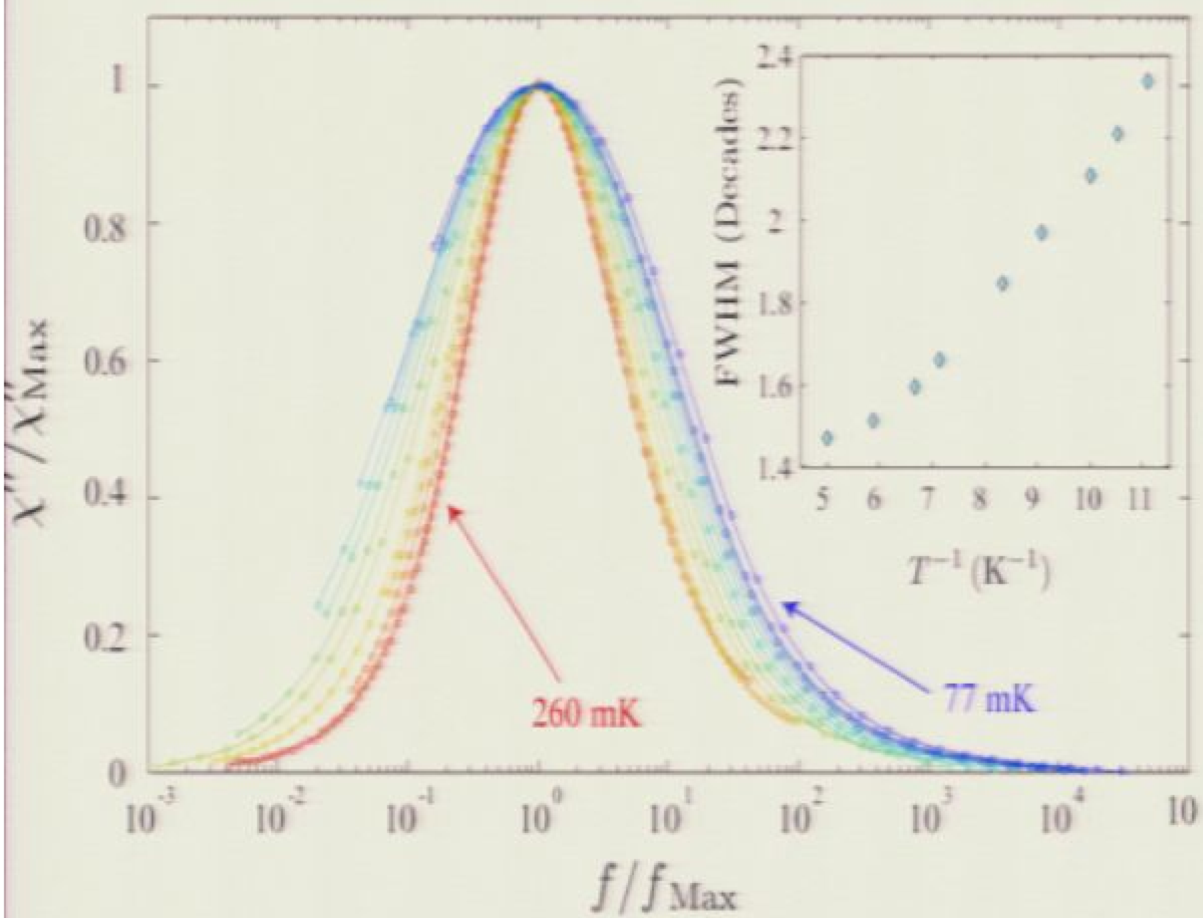


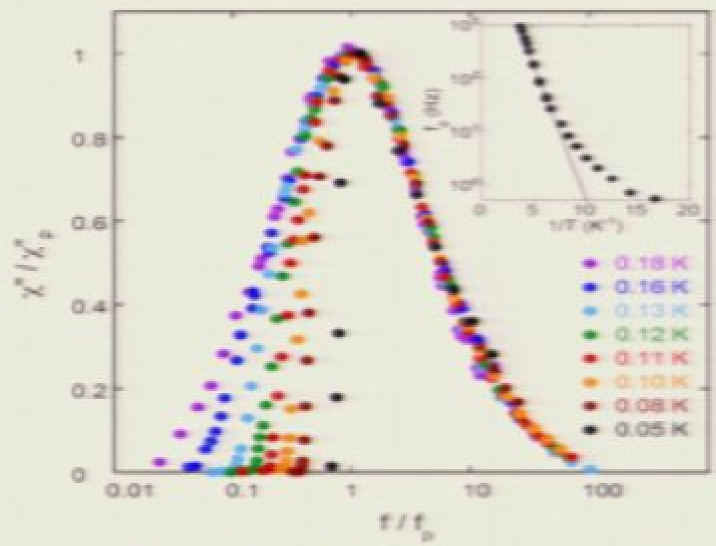
Arrhenius behavior can be attributed to a superparamagnet.

Deviation from Arrhenius behavior at lower temperature may indicate that this is a spin glass with $T > T_g$.

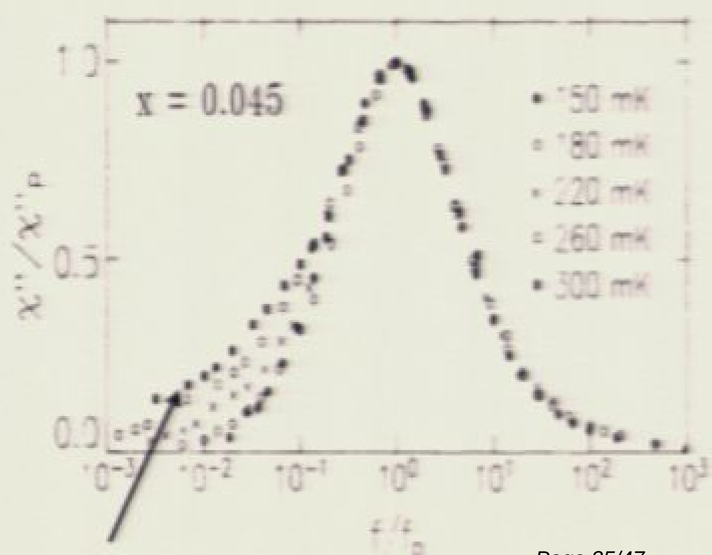
Width of χ'' for Various Temperatures

Our Result:





Ghosh *et al*. Science (2002)



300 mK

Reich et al. PRB (1990)

Our χ'' broadens as temperature decreases.

Consistent with a spin glass.

Not consistent with antiglass.

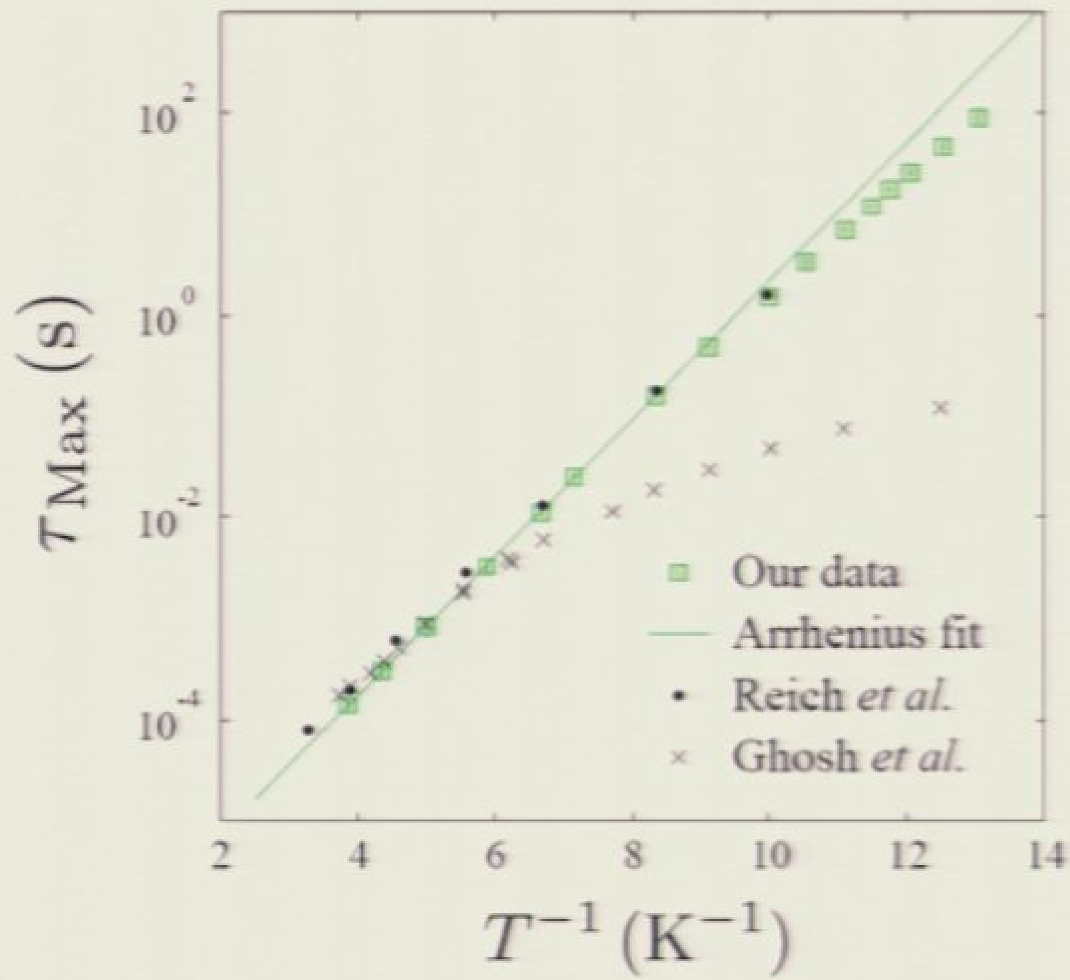
Fit to Arrhenius Law

τ_{Max} is the determined from the frequency of the peak in $\chi''(f)$ for a given temperature, T.

$$\tau_{Max}(T) = \tau_{oA} e^{-E_a/k_B T}$$

$$E_A = 1.57K$$

$$\tau_{oA} = 0.32\mu s$$



Arrhenius behavior can be attributed to a superparamagnet.

Deviation from Arrhenius behavior at lower temperature may indicate that this is a spin glass with $T > T_g$.

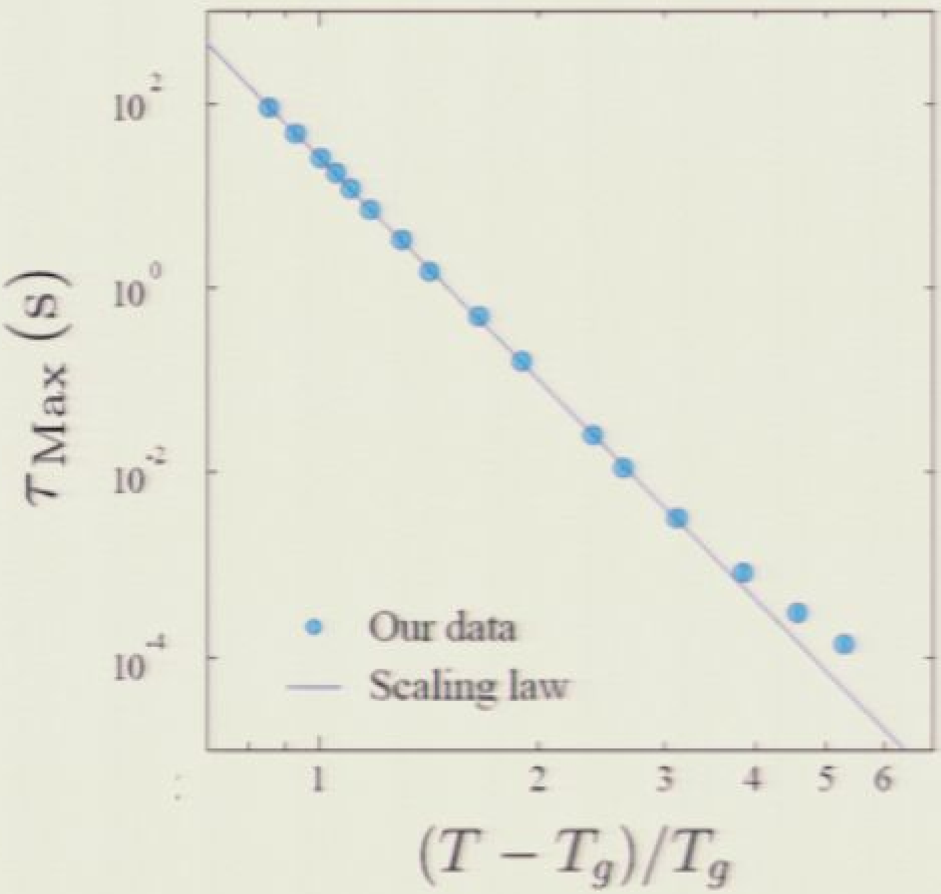
Dynamical Scaling Law for Spin Glass

$$\tau_{Max}(T) = \tau_o \left(T / T_g - 1\right)^{-z\nu}$$

$$T_g = 43 \pm 2mK$$

$$z\nu = 7.8 \pm 0.2$$

$$\tau_o = 16 \pm 7s$$



Dynamic scaling analysis points to the x = 0.045 system being a spin glass with a transition temperature of 43 mK and an intrinsic time constant of 16 seconds.

Six orders of magnitude slower than for example $Eu_{0.4}Sr_{0.6}S$.

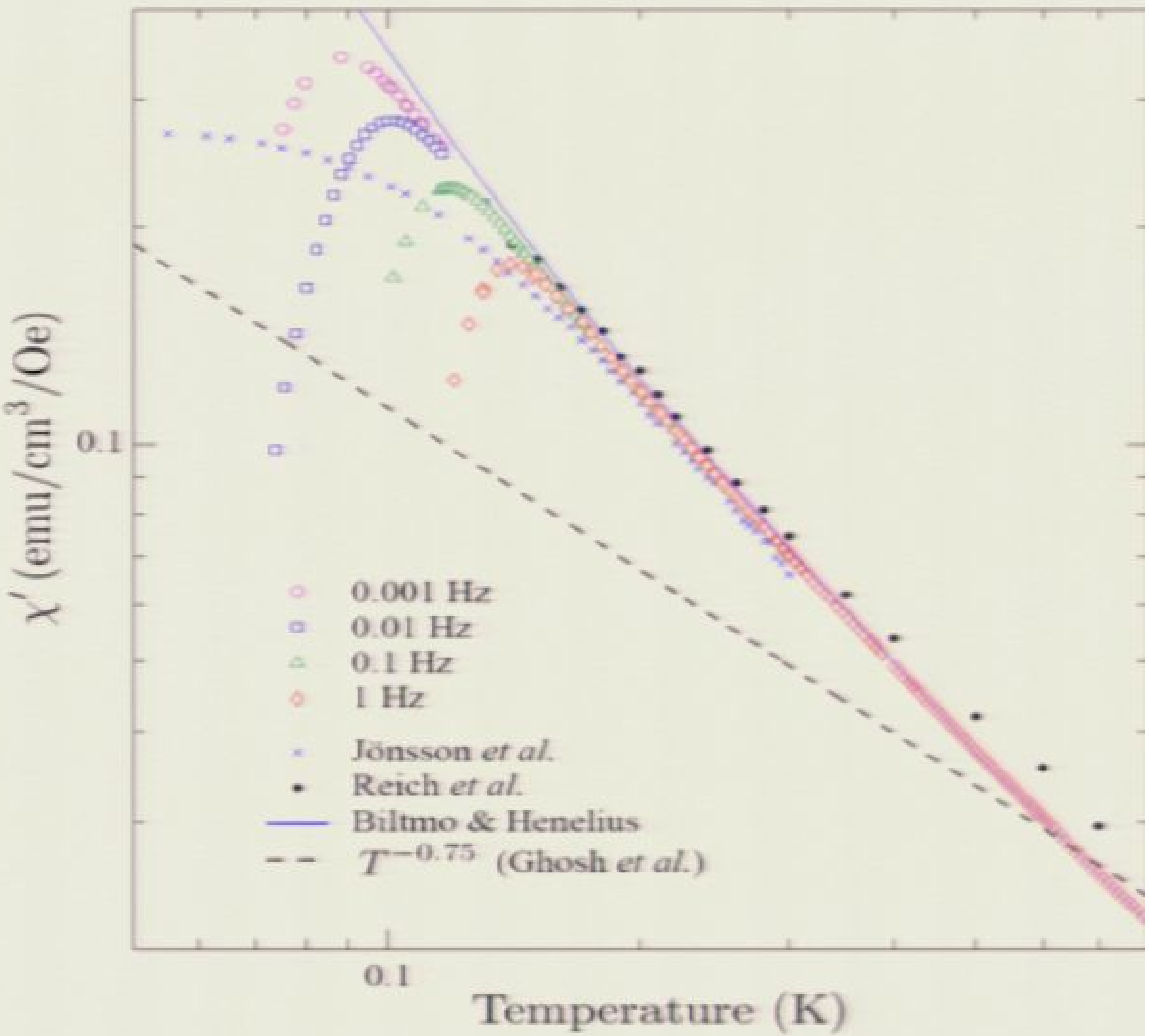
Temperature Dependence of χ'

•At higher temperatures, our χ' vs. T agrees with Jonsson *et al*, Reich *et al* and Biltmo and Henelius.

$$\chi' \propto T^{-1}$$

•At low temperature, even χ' measured with f = 0.001 Hz is not in the static limit.

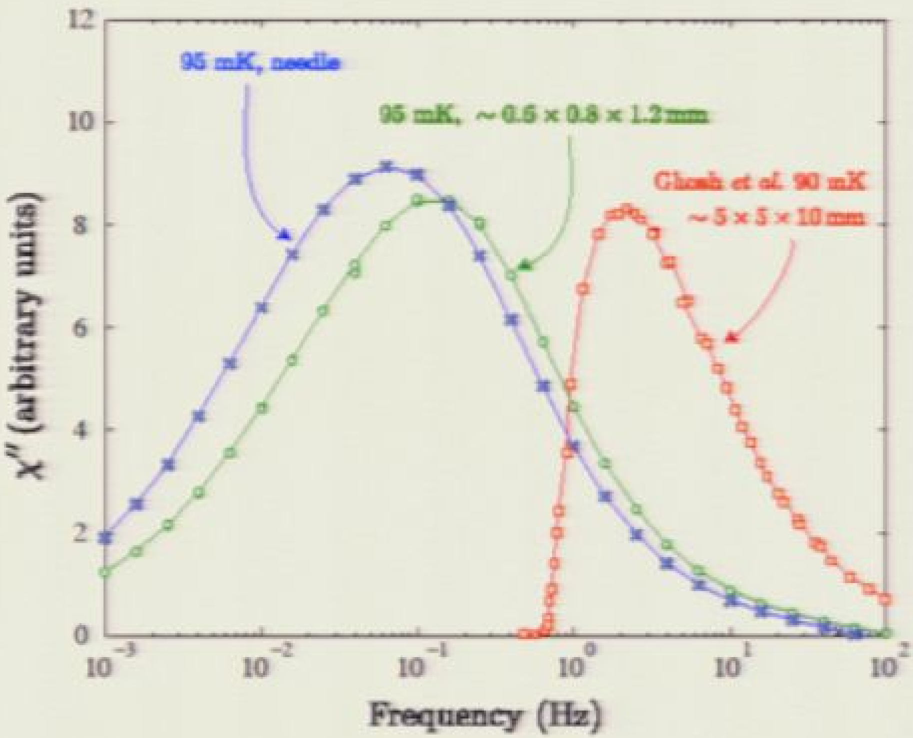
•It appears that Jonsson *et al* swept too quickly below 200 mK.
They swept the field at a rates between 1 to 50 Oe/s from H = 0 to H = 150 Oe.
---Sweep time was between 3 and 150 seconds.

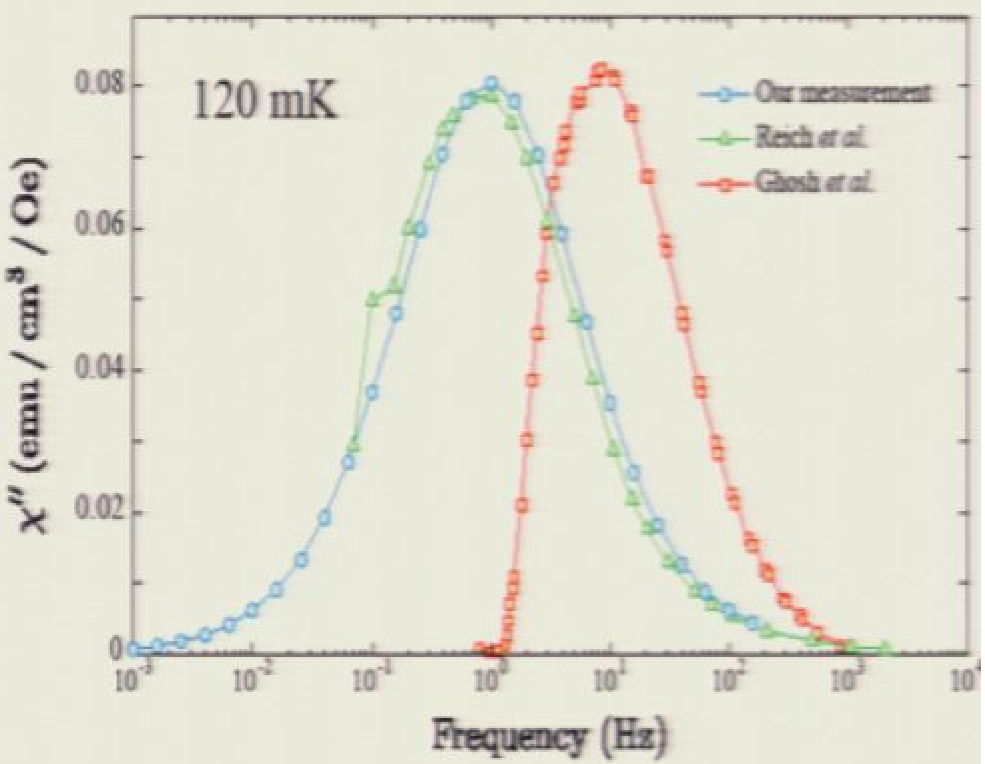


• Disagrees with Ghosh et al.

In a recent meeting in Toronto, Aeppli proposed that the difference between his group's results and ours and Jonnson et al's could be due to the geometry.
We have thin slivers and they have a fat chunk.

We just recently measured a sample with a similar aspect ratio to Ghosh et al and and found no difference





Temperature Dependence of χ'

- At higher temperatures, our χ' vs. T agrees with Jonsson *et al*, Reich *et al* and Biltmo and Henelius.

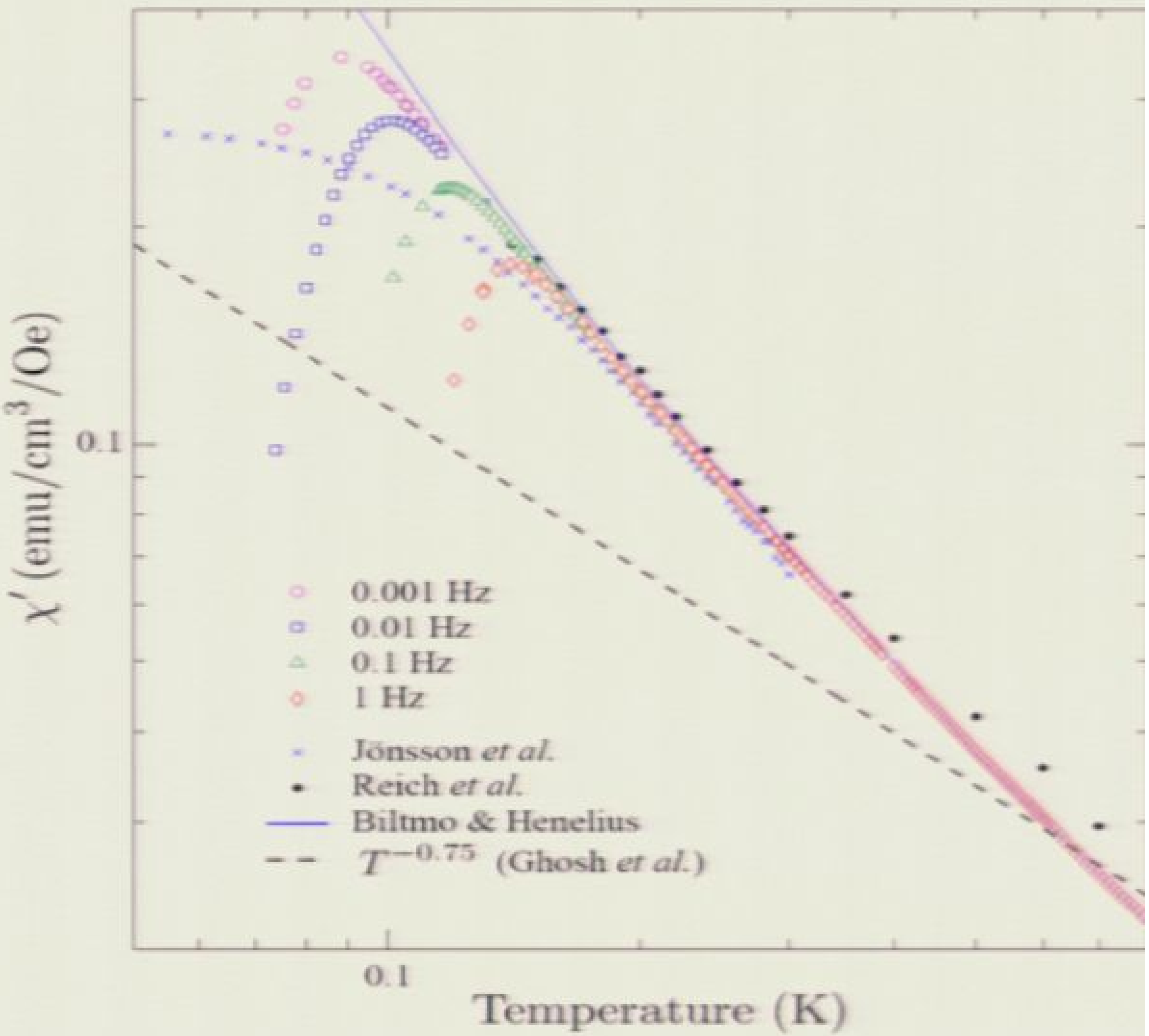
$$\chi' \propto T^{-1}$$

- At low temperature, even χ' measured with f = 0.001 Hz is not in the static limit.

- It appears that Jonsson *et al* swept too quickly below 200 mK.

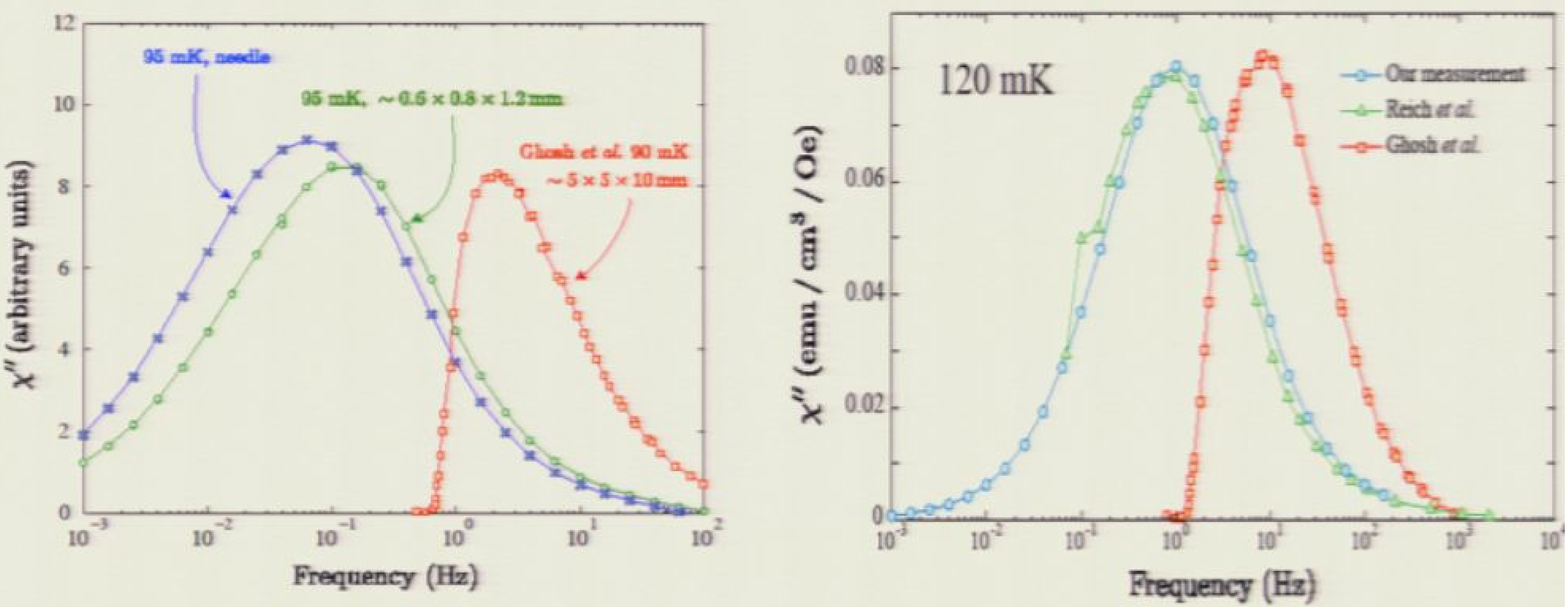
They swept the field at a rates between 1 to 50 Oe/s from H = 0 to H = 150 Oe.
---Sweep time was between 3 and 150 seconds.

- Disagrees with Ghosh et al.



In a recent meeting in Toronto, Aeppli proposed that the difference between his group's results and ours and Jonnson et al's could be due to the geometry.
We have thin slivers and they have a fat chunk.

We just recently measured a sample with a similar aspect ratio to Ghosh et al and and found no difference



Conclusions for ac Susceptibility Measurement

- Measured ac susceptibility of x = 0.045 sample. No exotic anti-glass behavior seen.
- Measured $\chi'_{DC} \propto T^{-1}$ in agreement with Jonsson *et al* and Reich *et al*, disagreement with Ghosh *et al*.
- The broadening of the absorption spectrum as temperature is lowered is consistent with behavior of a spin glass.
- The temperature dependence of χ'' follows a near Arrhenius behavior indicating that the system is either a spin glass or superparamagnet.
- Dynamic scaling analysis points to a spin glass transition temperature of 43 mK+-2mK.
- This is close to prediction made by simulations of Tam and Gingras which predict a spin glass transition for x = 0.065 to occur at a temperature of 43 mK and 95 mK for x= 0.125. (arXiv:0810.0854).

Our work has been published:
"Evidence of Spin Glass Dynamics in Dilute $LiHo_xY_{1-x}F_4$"
J.A. Quilliam, S. Meng, C.G.A. Mugford, and J.B. Kycia, Phys Rev Lett. (2008).

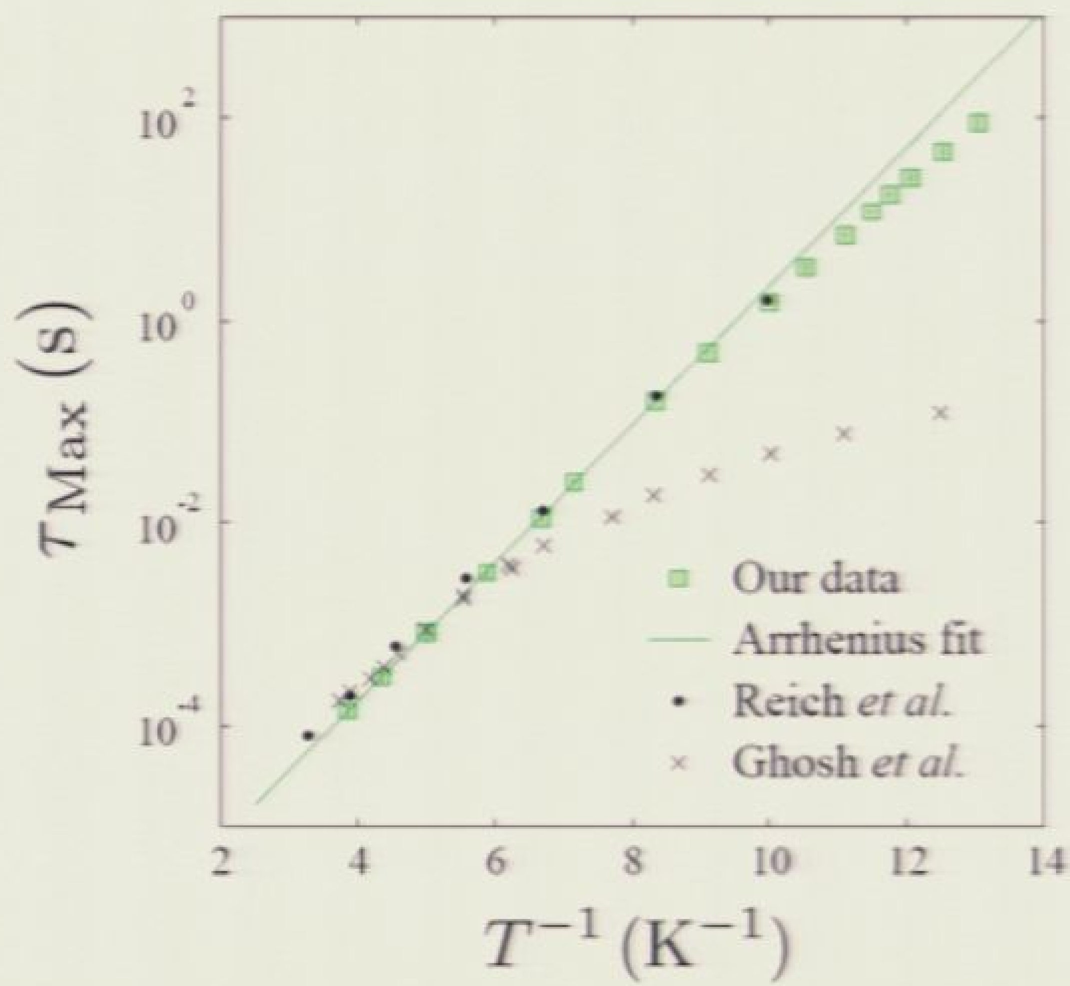
Fit to Arrhenius Law

τ_{Max} is the determined from the frequency of the peak in $\chi''(f)$ for a given temperature, T.

$$\tau_{Max}(T) = \tau_{oA} e^{-E_a/k_B T}$$

$$E_A = 1.57K$$

$$\tau_{oA} = 0.32\mu s$$

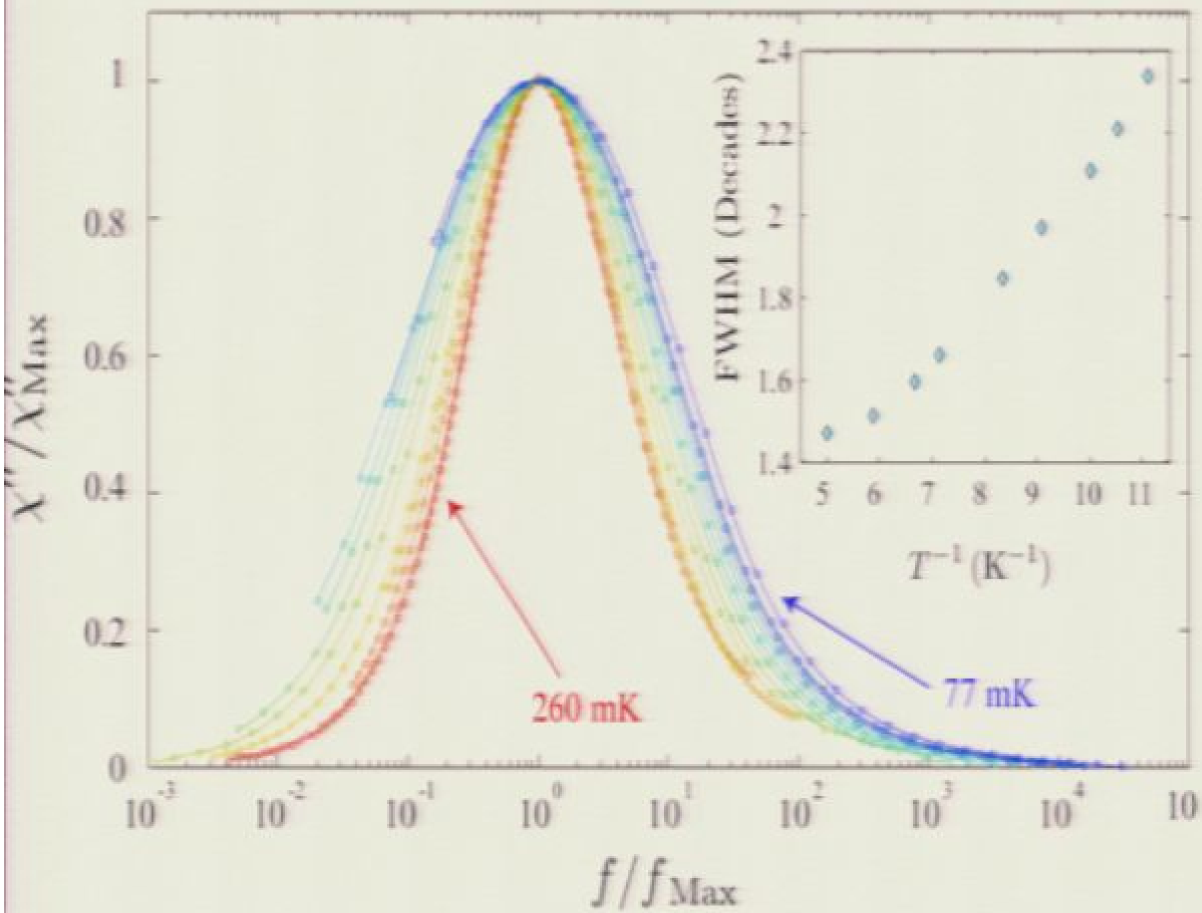


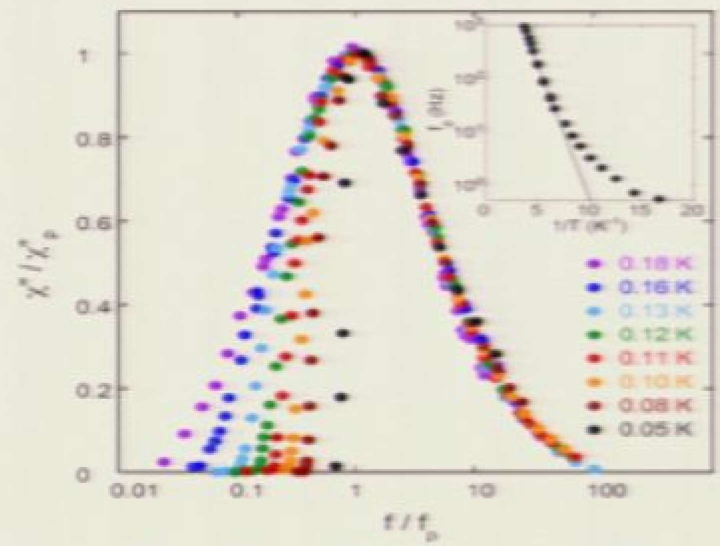
Arrhenius behavior can be attributed to a superparamagnet.

Deviation from Arrhenius behavior at lower temperature may indicate that this is a spin glass with $T > T_g$.

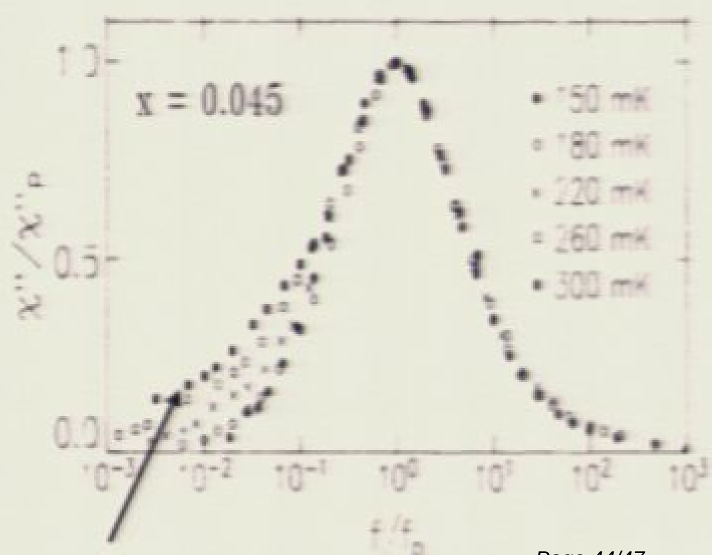
Width of χ” for Various Temperatures

Our Result:





Ghosh *et al*. Science (2002)



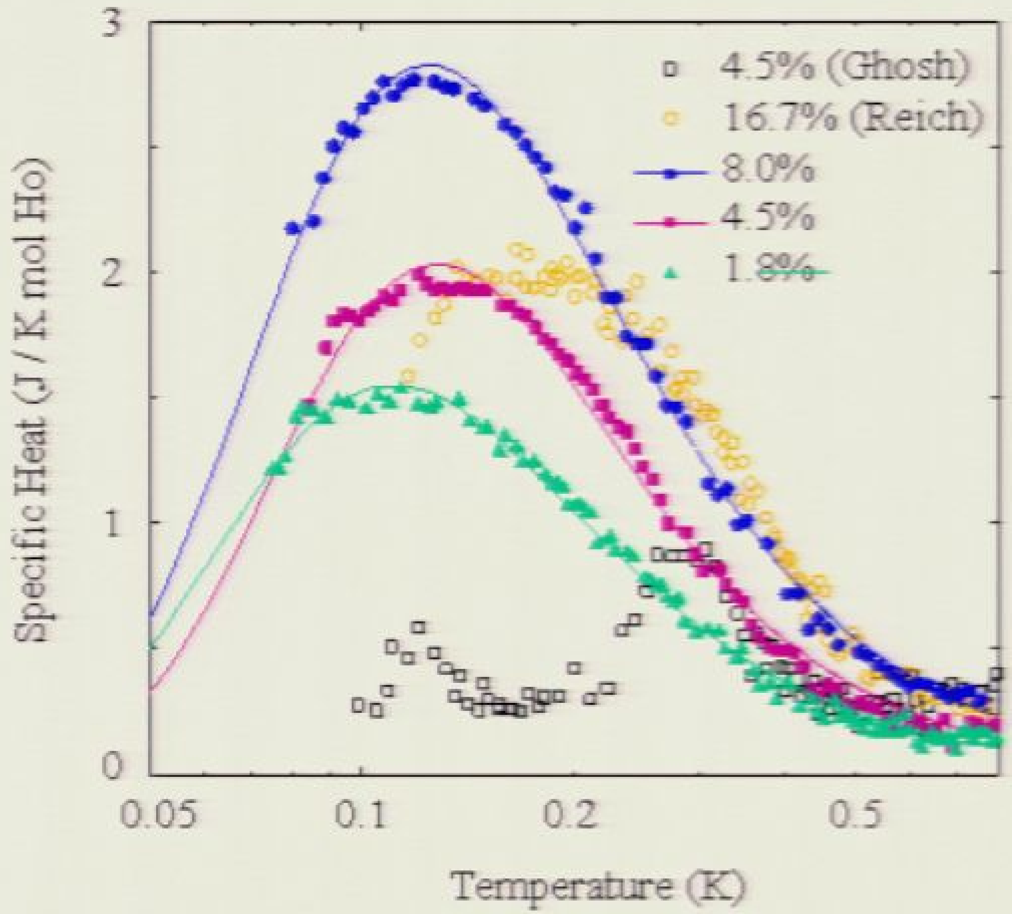
300 mK

Reich et al. PRB (1990)

Our χ” broadens as temperature decreases.
Consistent with a spin glass.
Not consistent with antiglass.

Comparison with Previous Results

- Our results do not reproduce the unusually sharp features observed by Ghosh *et al*. in 4.5% Ho:YLF
- Thermal conductivity of 4% sample also saw no sharp features (Nikkel & Ellman CondMat 0504269)
- Data is qualitatively consistent with the 16.7% sample measured by Reich *et al.*
- We account for much more of the expected entropy in the system (*R*ln2)
- Heat capacity does not give us a measure of the dynamics of the system so cannot say whether "anti-glass" or not.



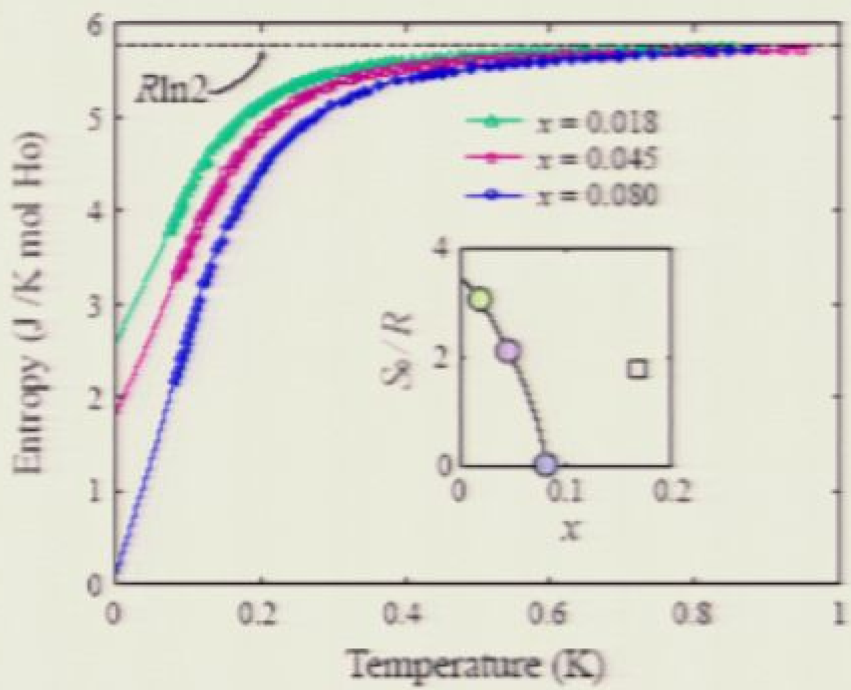
Reich *et al*. PRB 42, 4631 (1990)
Ghosh *et al*. Science 296, 2195 (2002)

Residual Entropy?

- Entropy can be determined with numerical integral

$$S(T) = \int_0^T dT \frac{\Delta C(T)}{T}$$

- Total entropy should be $R \ln 2$
- Possibility of residual entropy S_0
- Integral done here with linear extrapolation to $T = 0$
- S_0 is increasing with decreasing x

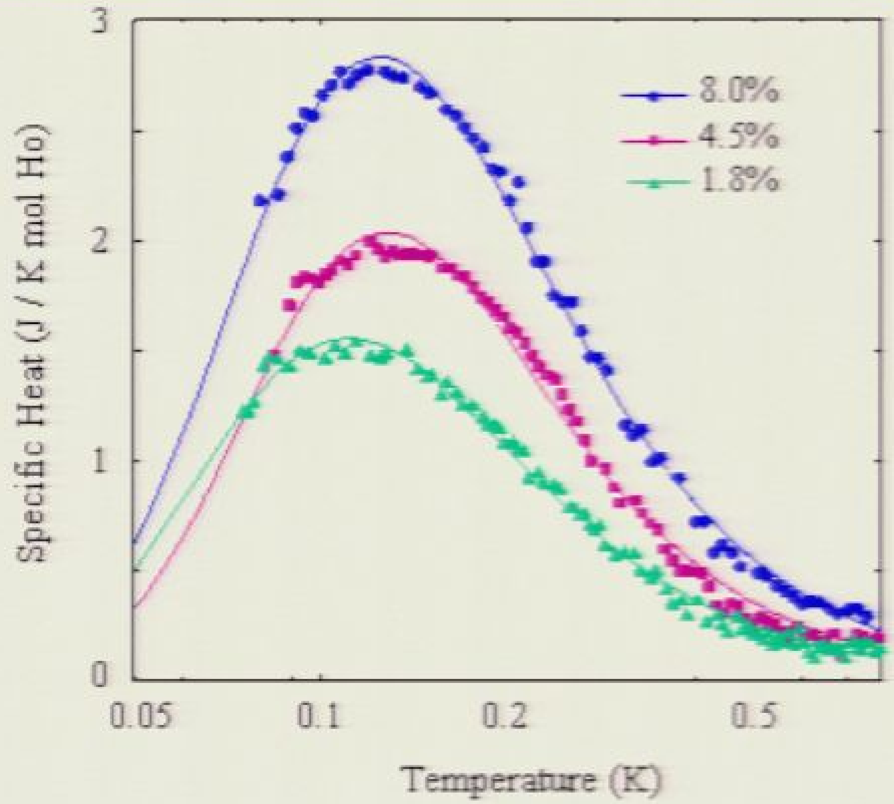


After Subtraction of Nuclear Specific Heat

- Non-interacting C_N subtracted to give electronic part ΔC
- Broad feature remains which is consistent with a spin glass for all 3 samples
- Spin glass C does not have a sharp feature at T_0
- Indicative of excitations above the transition
- Simplest model: 1 excited energy level with degeneracy n w.r.t. ground state (fits)

$$\Delta C \propto \frac{(E_1/kT)^2 e^{-E_1/kT}}{(1+ne^{-E_1/kT})^2}$$

- More low-temperature data required to look for linear temperature dependence



Parameter	1.8% sample	4.5% sample	8.0% sample	16.7% sample [1]
C_0 (J/K mol Ho)	4.06	3.45	7.31	2.85
E_1/k_B (K)	0.26	0.32	0.29	0.46
n	0.85	1.43	0.86	1.89
T_{peak} (K)	0.11	0.13	0.12	0.17
FWHM/T_{peak}	1.7	1.6	1.7	1.5
S_0/R	0.31	0.21	0.00	0.18

[1] Reich *et al.* PRB 42, 4631 (1990).