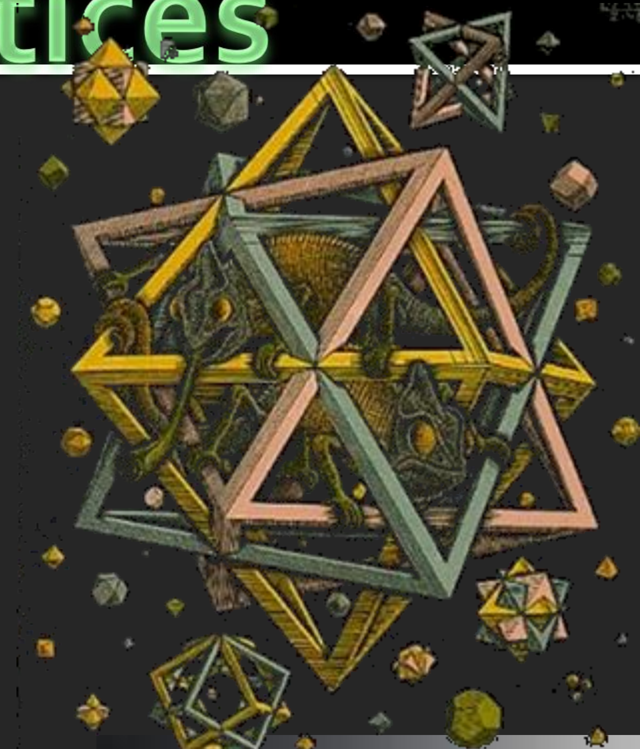
Impact of Perturbations on Frustrated Lattices



Oren Ofer and Amit Keren

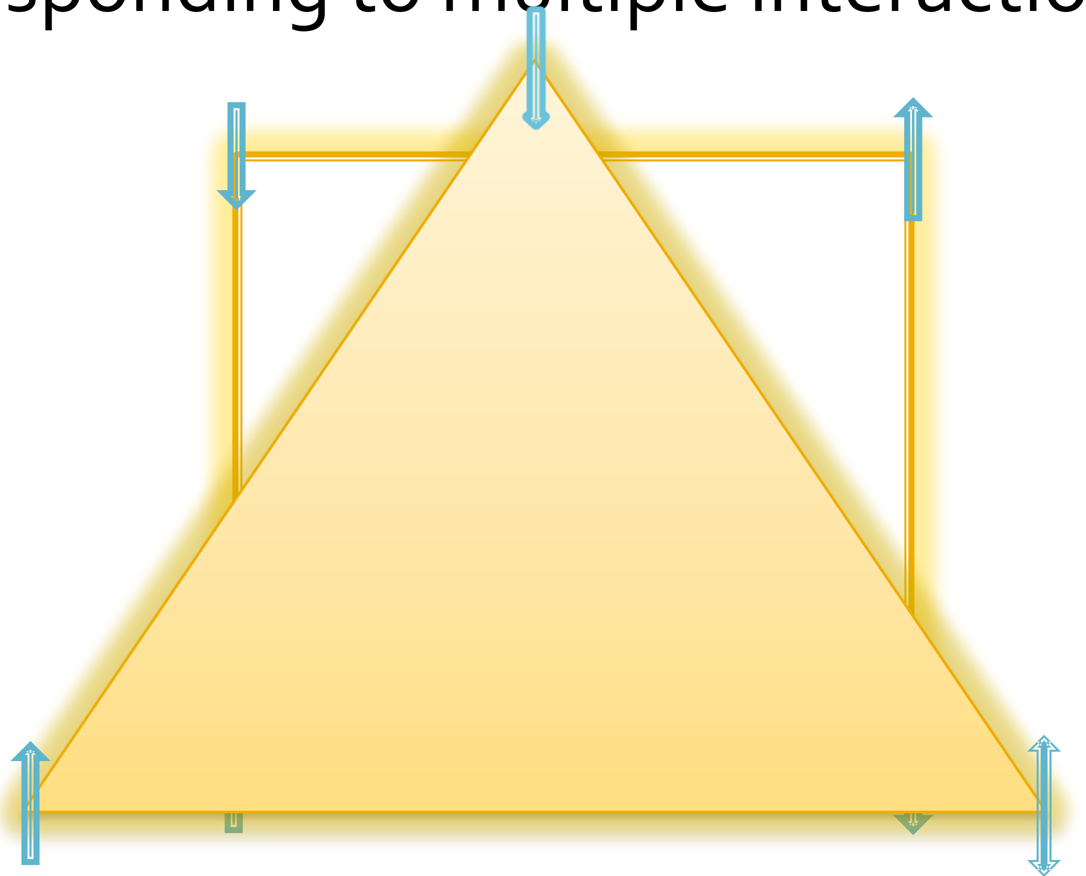
O.O and A.K, J. Phys: Conden. Matter **19**, 145270 (2006)
O.O and A.K., cond-mat/0610540 (2007)
O.O and A.K., cond-mat/0804.4781 (2008)
O.O and A.K, *in preparations*

Outline

- What is Geometrically Frustrated Magnet?
- Two lattices: the Kagome ($ZnCu_3(OH)_6Cl_2$) and the Pyrochlore ($Y2Mo2O7$ /$Tb2Ti2O7$).
- 3 Possible perturbations.
- Experimental results.

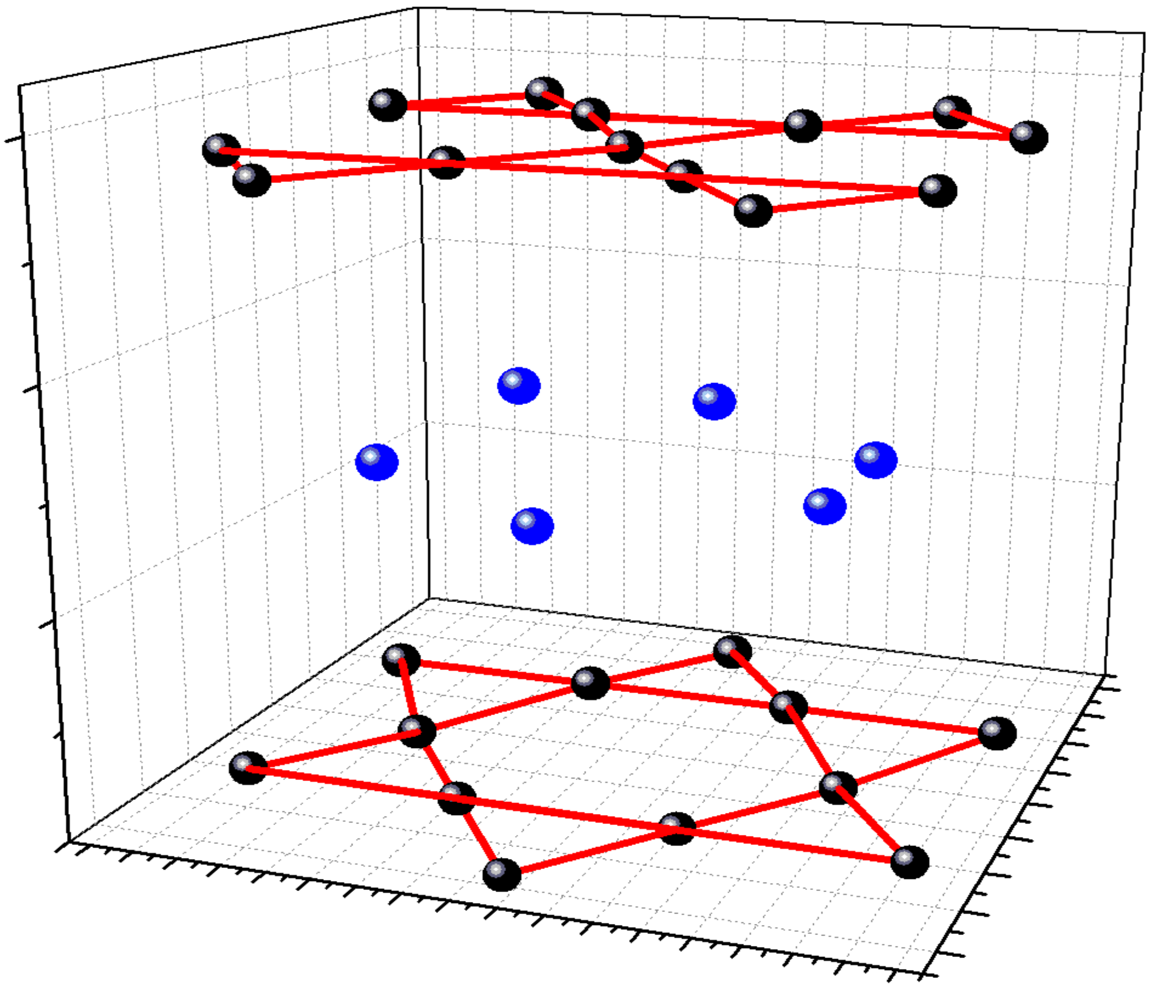
Geometric Frustration: unitcell

- The inability to minimize different energies corresponding to multiple interactions.



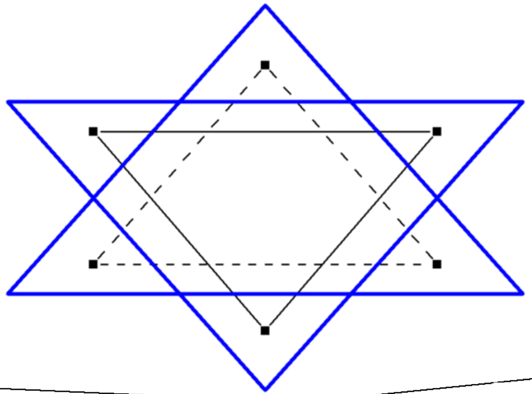
Extended Systems I: Kagome

- 2D: Kagome



Extended Systems II: Pyrochlore

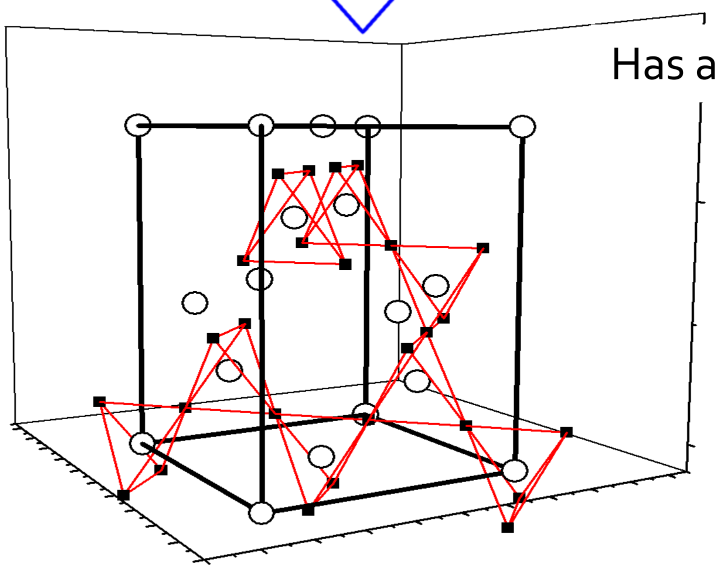
- 3D: Pyrochlore

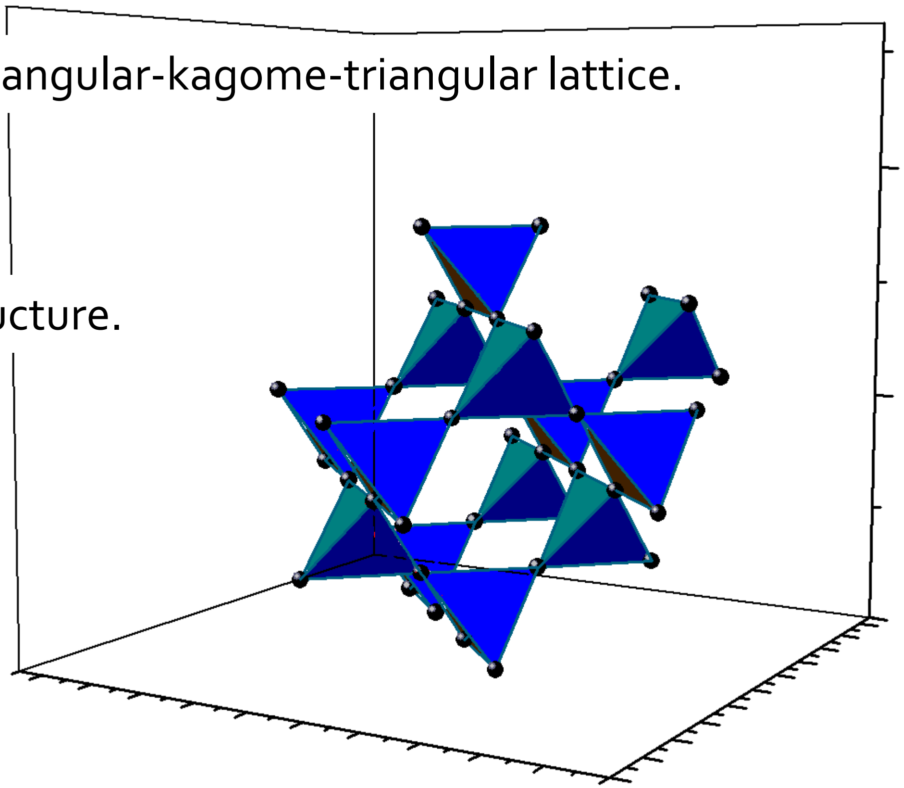


Corner Sharing Tetrahedra = Pyrochlore.

A stacked triangular-kagome-triangular lattice.

Has a FCC structure.





Ground State Degeneracy

- Classical n.n AF Heisenberg Hamiltonian,

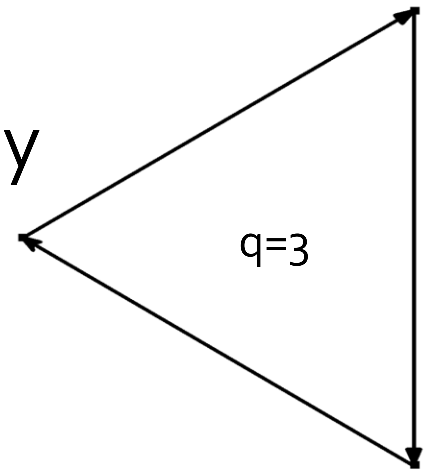
$$\mathcal{H} = J\sum_{\langle i,j\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

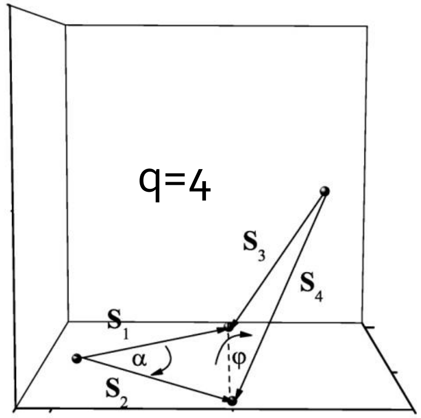
- In corner sharing systems of q mutually interacting spins,

$$\mathcal{H} = \frac{1}{2}J\sum_{\Delta}\left(\sum_{i=1}^{q}\mathbf{S}_i\right)^2 - \frac{J}{2}\sum_i S_i^2$$

$$\propto \frac{J}{2}\sum_{\Delta}\mathbf{L}^2$$

where $\mathbf{L} = \sum_{i=1}^{q}\mathbf{S}_i$.





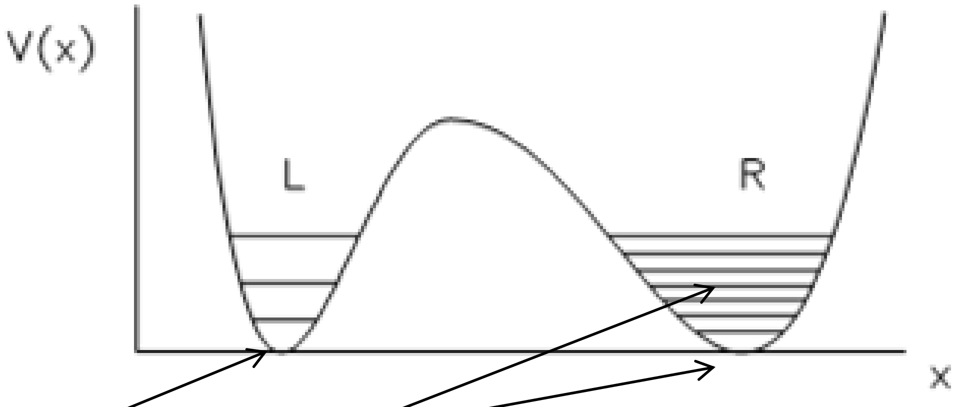
Ground State Degeneracy

- Heisenberg Hamiltonian predicts, a macroscopic ground state degeneracy with no Long range order.
- Breaking the GS degeneracy - Every perturbation will have a huge effect on the GS
- GFM ARE IDEAL TO GO BEYOND THE HEISENBREG MODEL.

Our aim is to find possible perturbations in various geometrically frustrated magnets.

Order from disorder

- A single quantum particle in a non-symmetric double potential well.

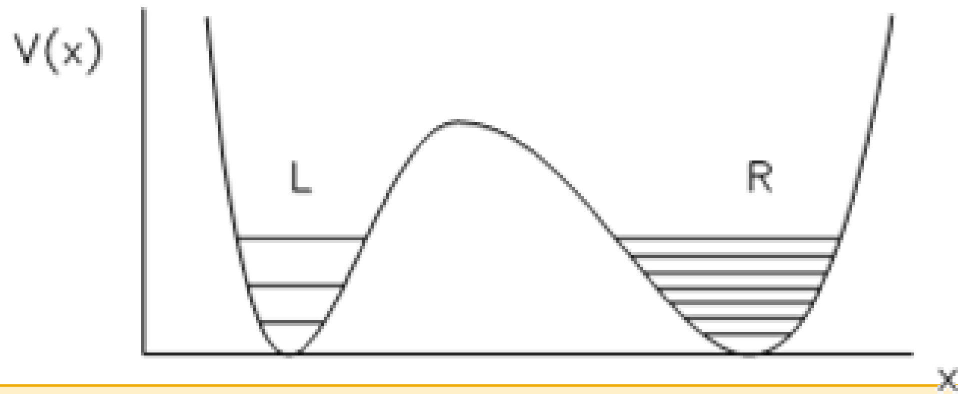


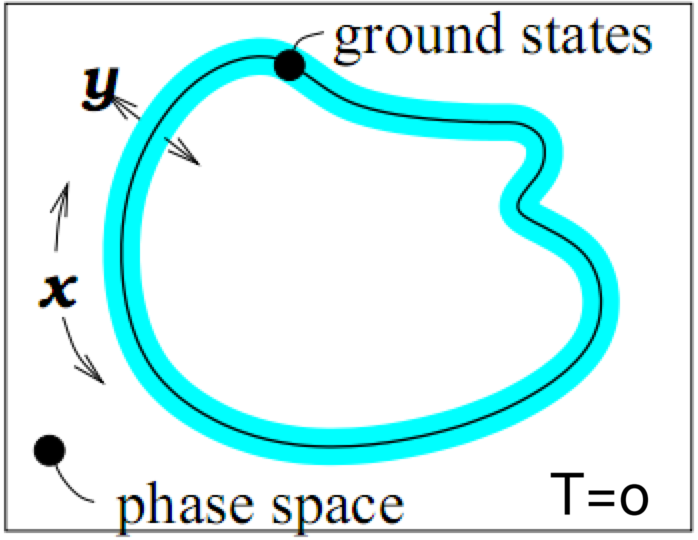
At T=0 both sides have the same energy.

At finite T, the particle will likely to be in R.

Villain *et al.*, Journal de Physique **41**, 1263 (1980).

Order from disorder in frustrated lattices



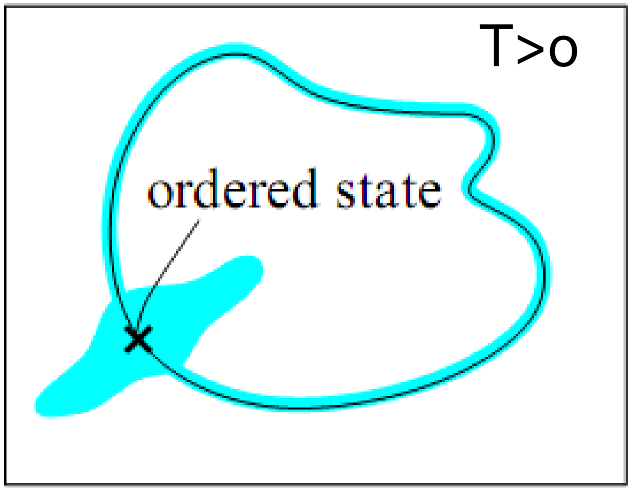


"Order from disorder" is possible in kagome lattices *not* in pyrochlores.

The density of excited states is highest near a LRO configuration.

The softer the fluctuations are, the larger phase space accessible to our particle.

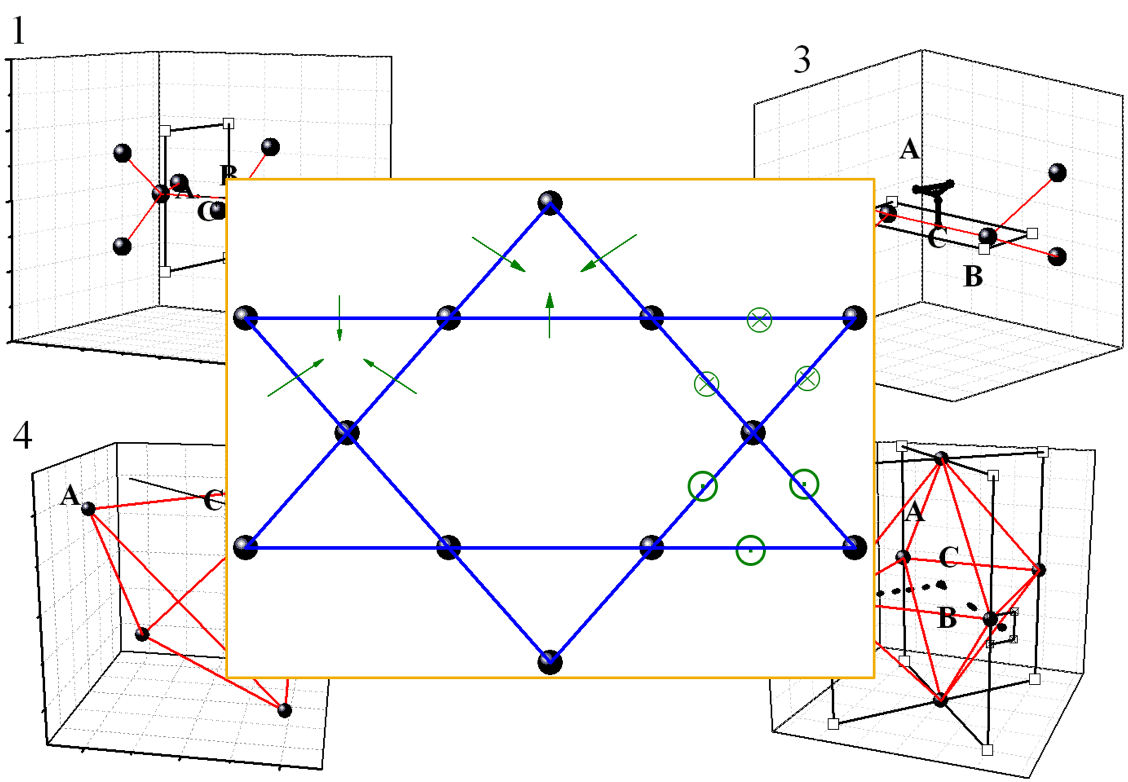
Thus it is probable that soft modes can cause the system to enter a LRO configuration from a thermal-disordered tendency.



Villain *et al.*, Journal de Physique **41**, 1263 (1980).

Breaking the GS degeneracy

1. Dzyaloshinsky Moriya Interactions



- DMI bi-linear

$$\mathcal{H}_{DM} = \sum_{<i,j>} \mathbf{D}_{ij} \left(\mathbf{S}_i \times \mathbf{S}_j \right)$$

DMI rules:

1. Center of inversion at C, D=0.
2. Mirror plane⊥AB through C, D||AB or D⊥AB.
3. Mirror plane in AB, D⊥AB.
4. 2-fold (180°) rotation axis ⊥AB through C, D⊥axis.
5. n-fold (360°/n) rotation along AB, D||AB.

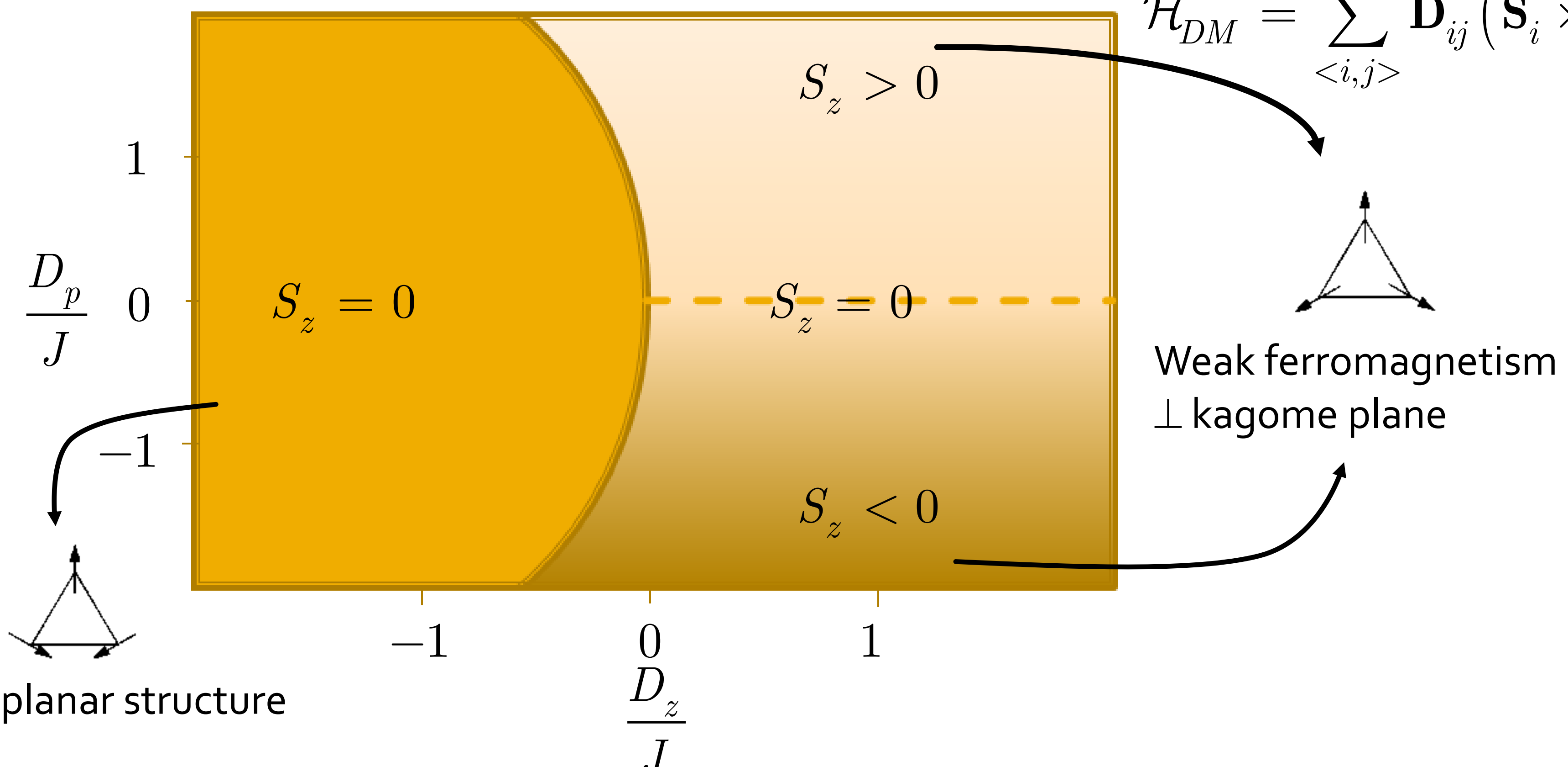
Tôru Moriya, Phys. Rev. Lett. **4**, 228 (1960)

Breaking the GS degeneracy

1. Dzyaloshinsky Moriya Interactions

- DMI bi-linear

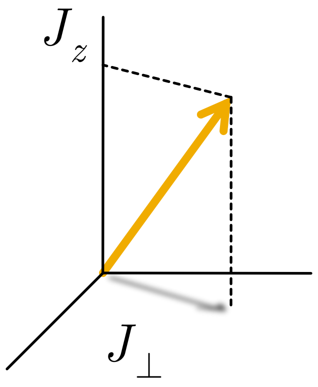
$$\mathcal{H}_{DM} = \sum_{<i,j>} \mathbf{D}_{ij} \left(\mathbf{S}_i \times \mathbf{S}_j \right)$$



Elhajal *et al.*, Phys. Rev. B **66**, 014422 (2002)

Breaking the GS degeneracy

2. Exchange Anisotropy



- *J* Anisotropy linear $J \rightarrow \mathbf{J} = J_z + J_\perp$

$$J_z \neq J_\perp$$

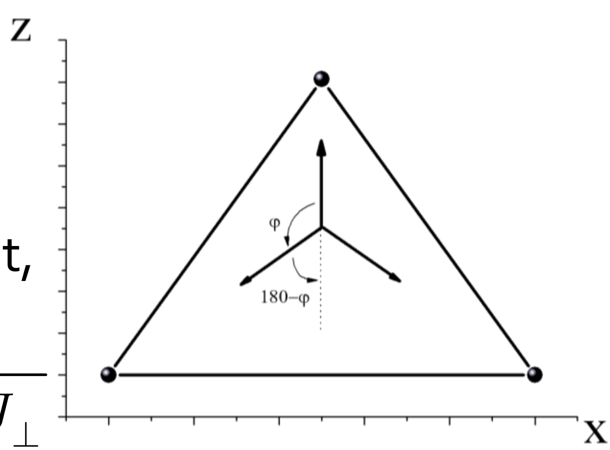
$$\mathcal{H} = \sum_{<i,j>} J_z S_i^z S_j^z + J_\perp S_i^\perp S_j^\perp$$

- In mean-field this yields a dramatic change in the susceptibility,

$$\chi_{\perp,z}^{-1} = \frac{3k_B}{(g\mu_B)^2 S(S+1)}(T + \theta_{\perp,z})$$

- *Suprisingly*, Could also induce FM with average moment,

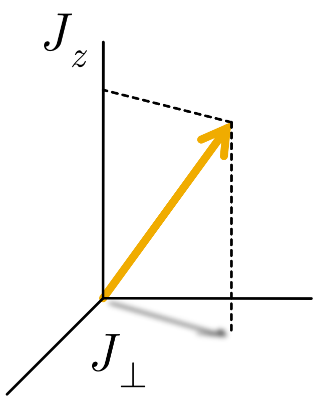
$$\left\langle \mathbf{M}\cdot\widehat{\mathbf{H}} \right\rangle = \frac{\mu_B}{6}(1 + 2\cos\varphi) \qquad \cos\varphi = \frac{J_z}{J_z + J_\perp}$$

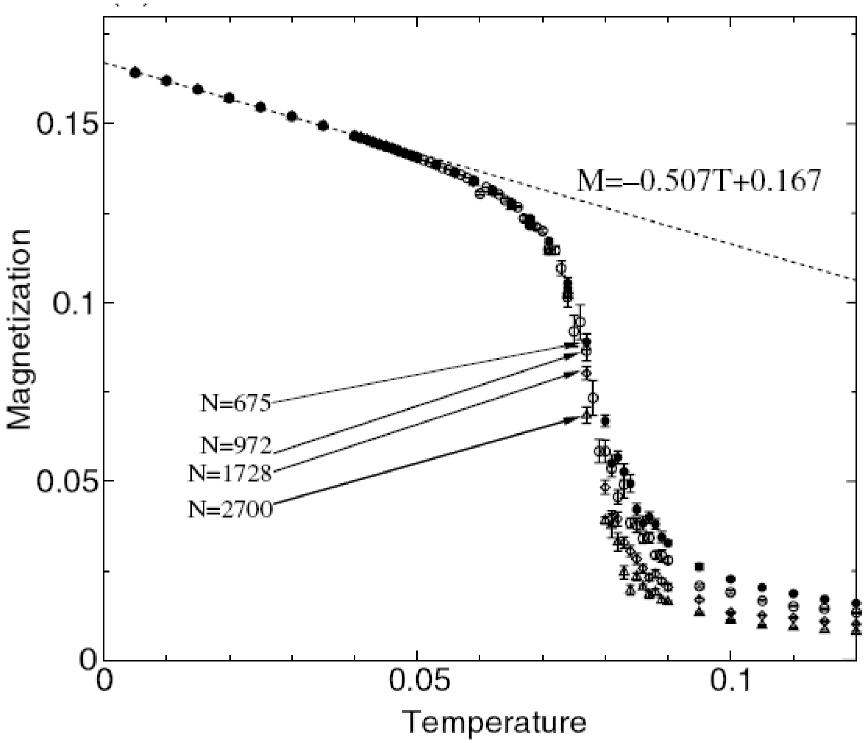


Breaking the GS degeneracy

2. Exchange Anisotropy

- J Anisotropy $\mathcal{H} = \sum_{<i,j>} J_z S_i^z S_j^z + J_\perp S_i^\perp S_j^\perp$
- At $T<T_c$ a FM order emerges

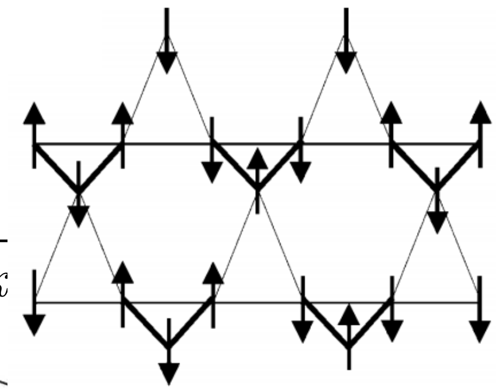




Tanaka and Myashita, J. Phys.: Condens. Matter **19**, 145256 (2007)

Breaking the GS degeneracy

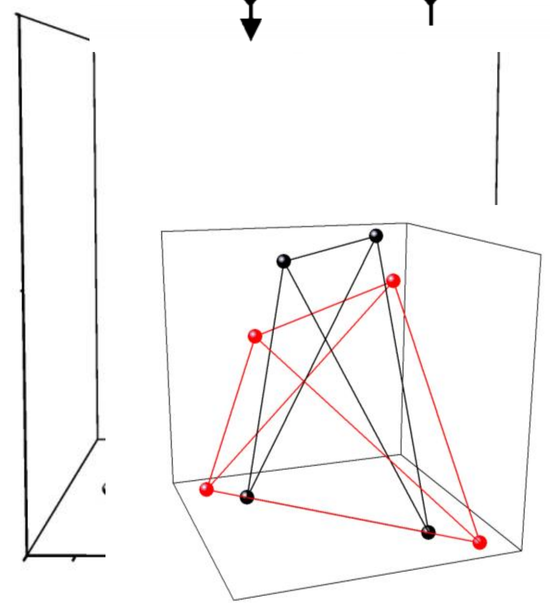
3. Lattice Deformations

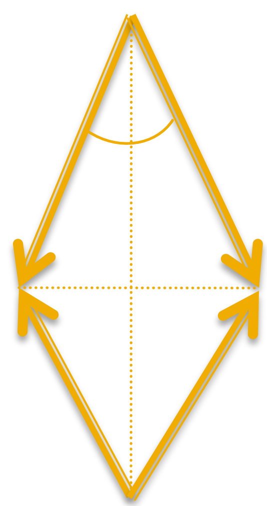


- Magneto-elastic bi-quadratic coupling $\mathcal{H}_{me} = \frac{1}{3k}$

  The term $\mathbf{S}_i \cdot \mathbf{S}_j$ favors

coplanarity $\varphi = 180^{\circ}$

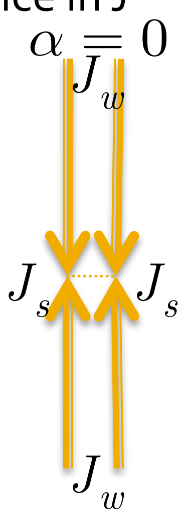




A difference in *J*

$J_w < J$

$J_s > J$



Breaking the GS degeneracy

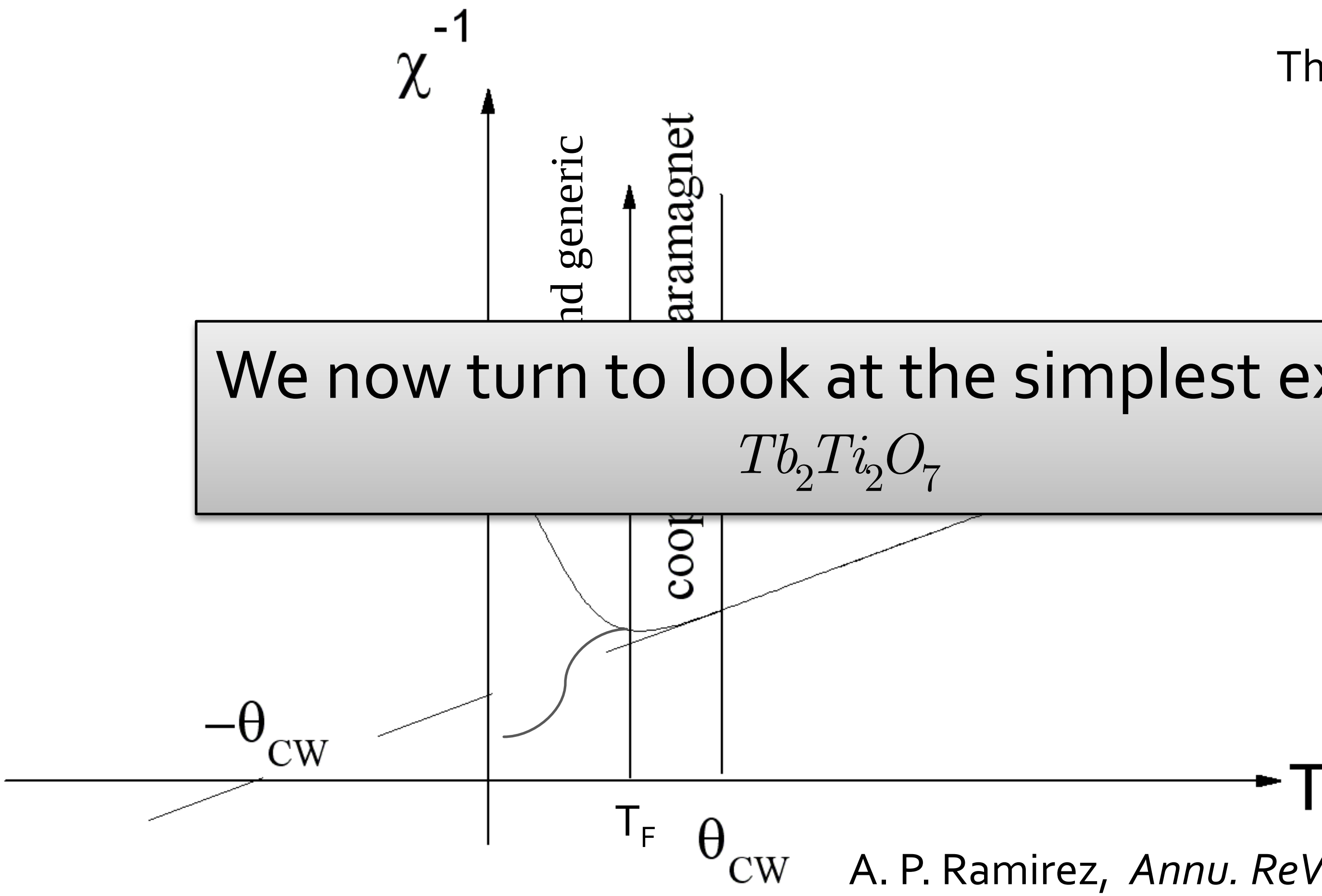
- The full Hamiltonian,

$$\mathcal{H} = \sum_{<i,j>} J_z S_i^z S_j^z + J_\perp S_i^\perp S_j^\perp + \mathbf{D}_{ij}\left(\mathbf{S}_i \times \mathbf{S}_j\right) + \frac{1}{3k}\left(\frac{dJ}{dr}\right)^2 \sum_{i>j}\left(\mathbf{S}_i \cdot \mathbf{S}_j\right)^2$$

Our aim is to find experimental indications to J_z and $J_\perp$, or $\mathbf{D}_{ij}$, or dJ/dr.

Experimental Fingerprint

- Susceptibility



The frustration parameter,

$$f = \frac{|\theta_{CW}|}{T_f}$$

ates the
rustration

We now turn to look at the simplest example:

$Tb_2Ti_2O_7$

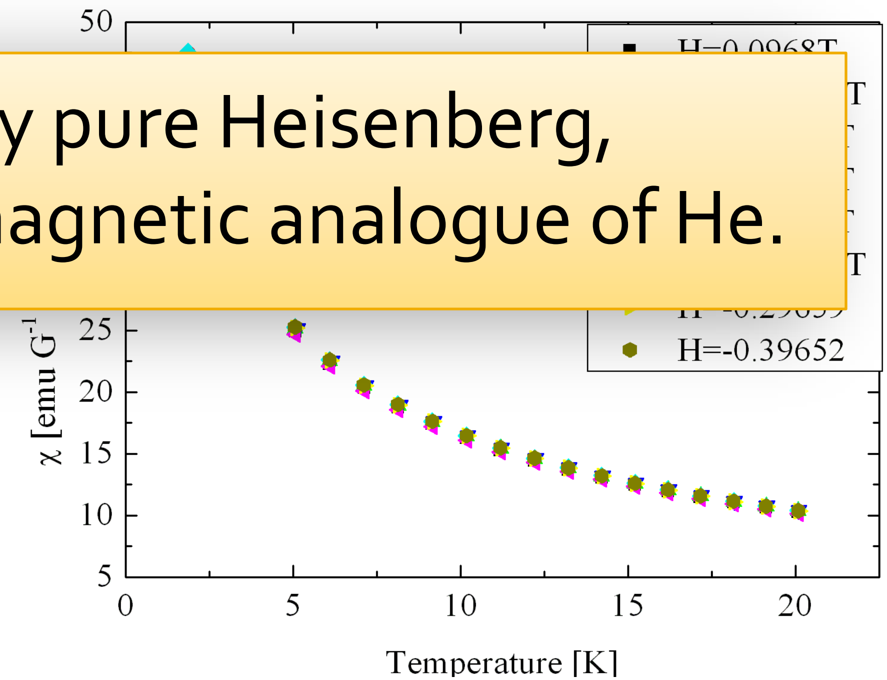
A. P. Ramirez, *Annu. ReV. Mater. Sci.* **1994, 24, 453**

Cooperative Paramagnet Tb2Ti2O7
χ

- Tb^{3+} ions creates magnetic pyrochlore.
- Doesn’t exhibits spin freezing down to 50mK.

The Hamiltonian is probably pure Heisenberg, might be considered as a magnetic analogue of He.

$$\theta_{CW} \approx -20\text{K} \rightarrow f > 400.$$



We will return to this compound later on.

Herbertsmithite

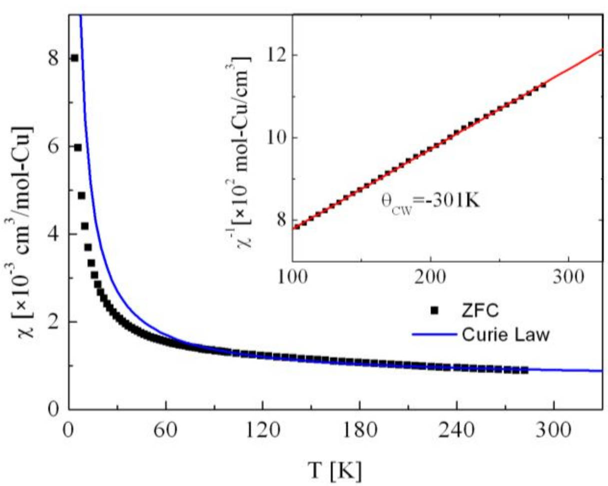
- Structurally perfect quantum S= ½ Kagome :

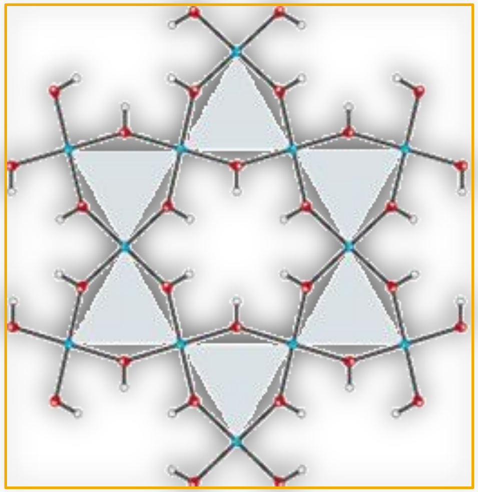
1. Cu^{+2} atoms carrying S=½ mediated through super-exchange $(OH)^{-}$.
2. Zn atoms sepa

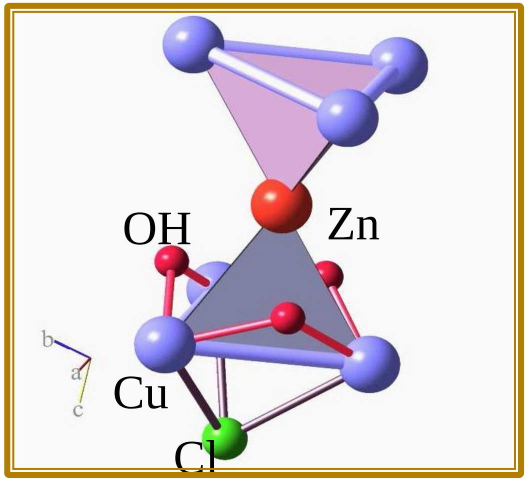
no 'Order from disorder' for T>2K!!!

3. XRD reveal perfect kagome structure, best model achieved when 100% Cu in the kagome, 100% Zn in the interlayer.

$$f > 157$$

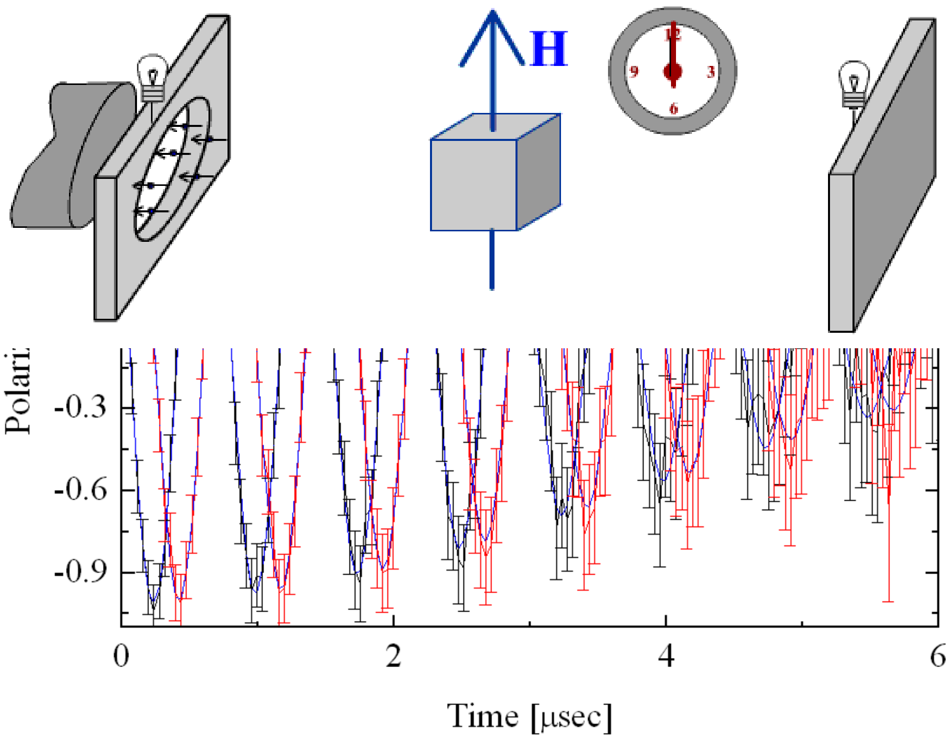






Herbertsmithite: Magnetic Characterization - μSR

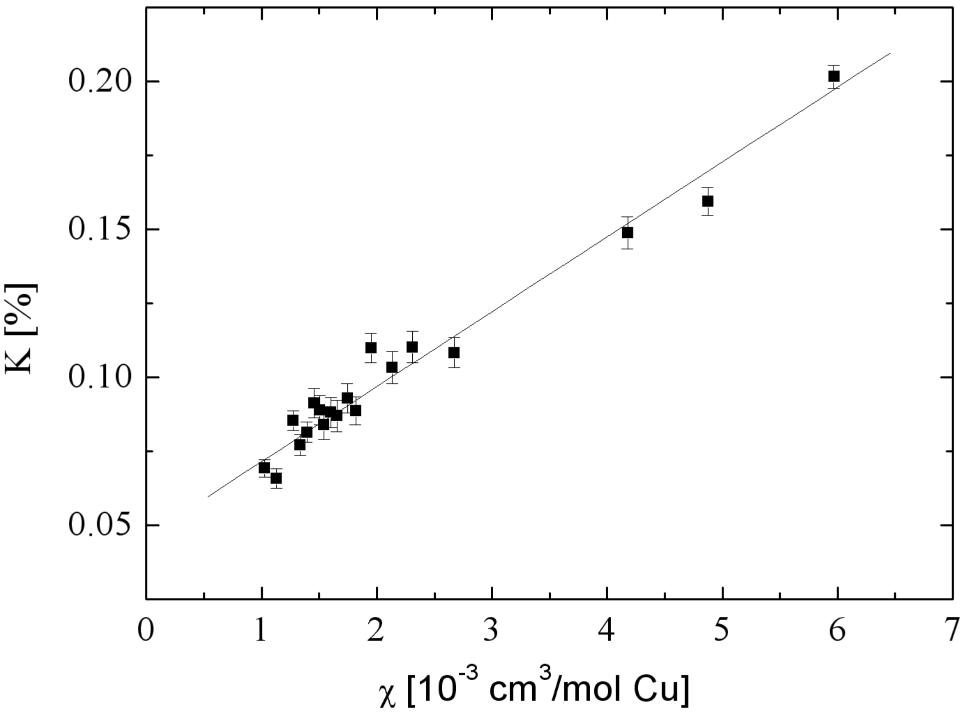
- We measure rotation frequency and relaxation of the muon spin.
- The frequency shift is a result of the sample magnetization.
- The TF relaxation is the result of static field inhomogeneties.



Herbertsmithite: Magnetic Characterization - μSR

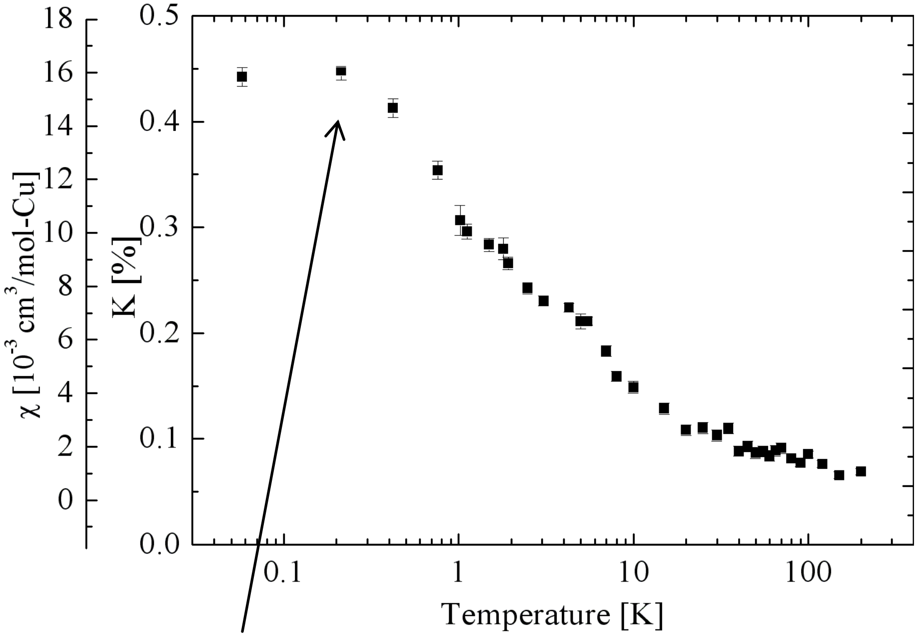
- K_μ calibration:

K and χ exhibit linear relation, thus allowing calibration.



Herbertsmithite: Magnetic Characterization - μSR

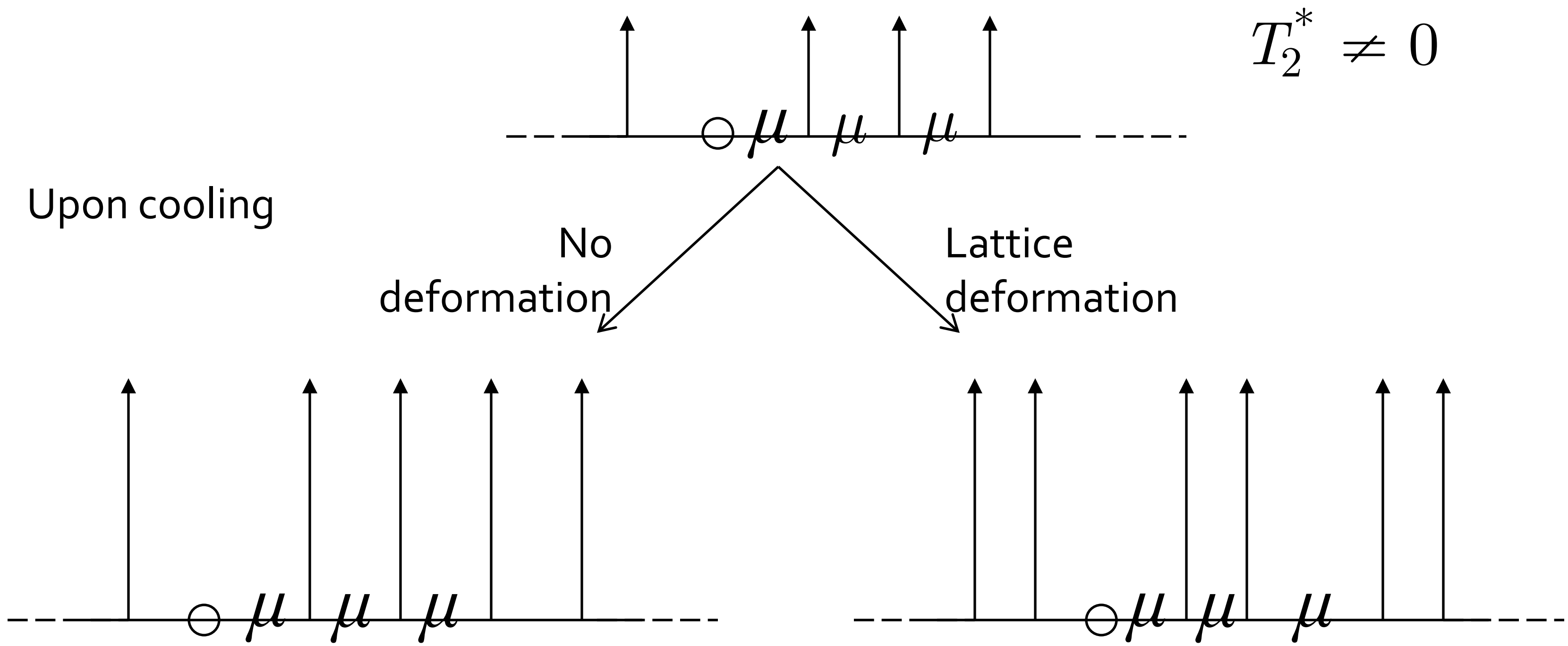
- *K* measured (hence χ) down to 60mK.
- Saturation of χ at *T*~200mK.
- As with the susceptibility, μSR does not detect spin freezing.



$$\frac{g\mu_B H}{k_B} = 0.268\text{K}$$

Herbertsmithite: Magnetic Characterization - μSR

- T_2^* **Interpretation**



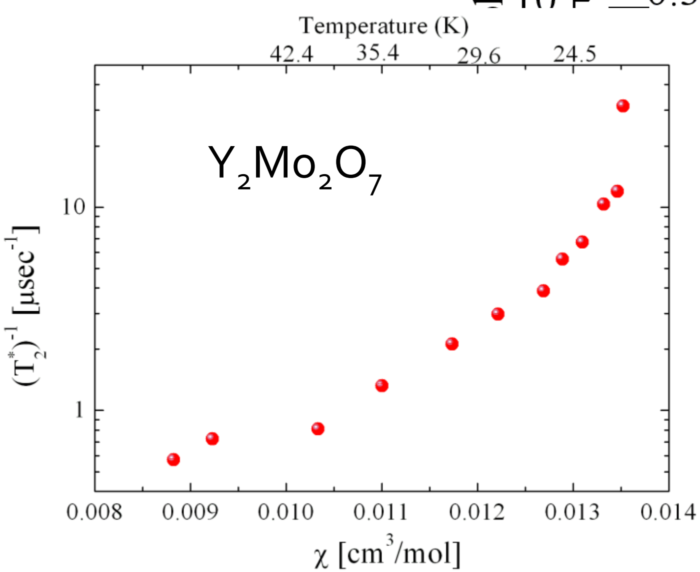
$$\frac{1}{T_2^*}(T) = C \cdot \chi(T)$$

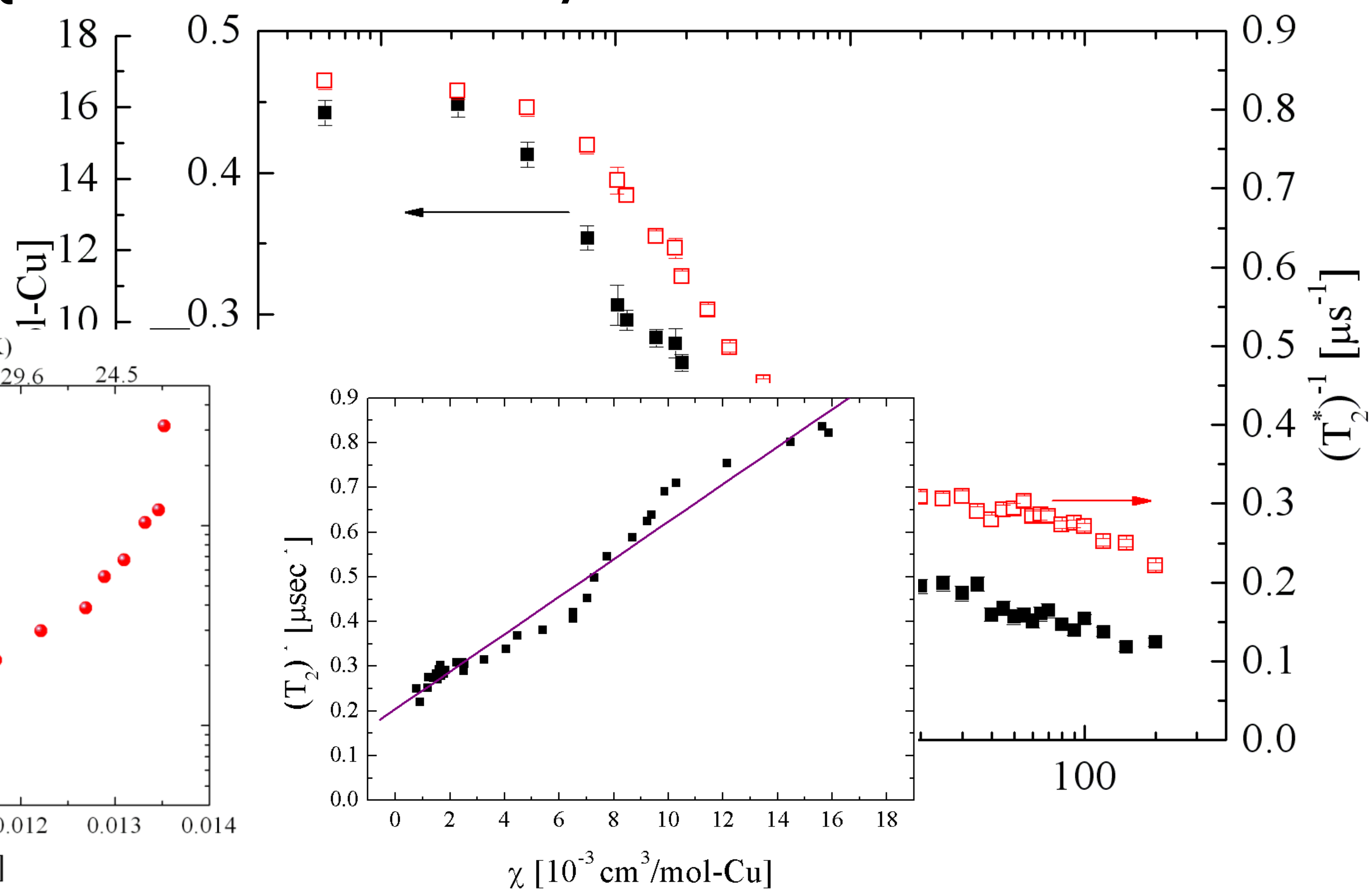
$$\frac{1}{T_2^*}(T) \neq C \cdot \chi(T)$$

Additional relaxation mechanism.

Herbertsmithite: Magnetic Characterization - μSR

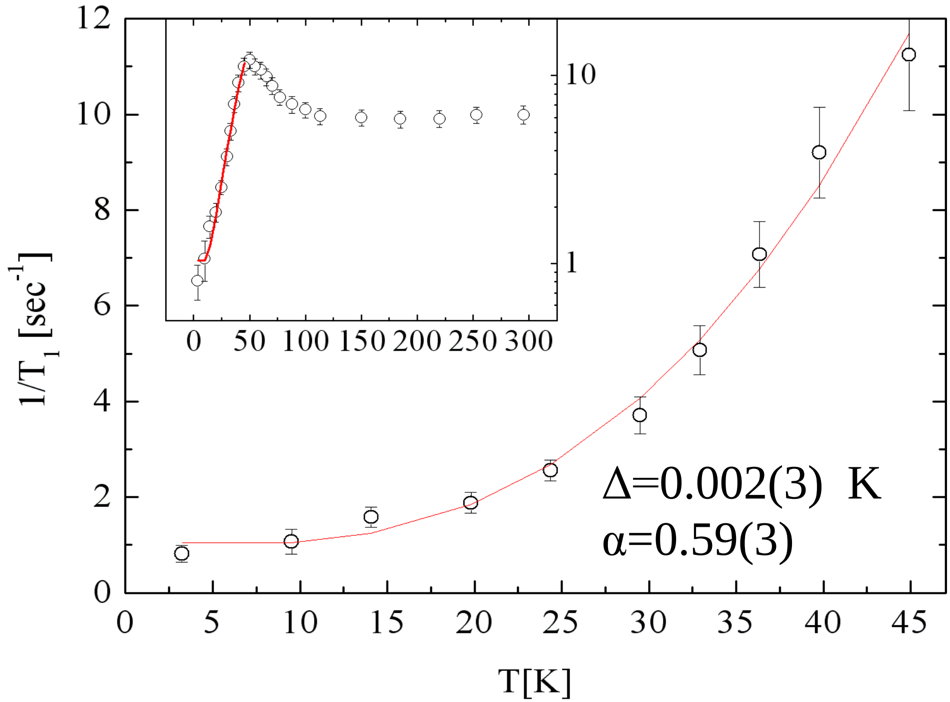
- Absence of Lattice deformation: $(T_2^*)^{-1}$ and χ behave similarly





Herbertsmithite: Magnetic Characterization - Cl NMR

- Cl line shape broadens as *T* decreases.
- $^{35/37}Cl\ T_1$ increases down to T~50K and slowly decreases.



- Spin lattice relaxation can be interpreted as a Bosonic excitation,

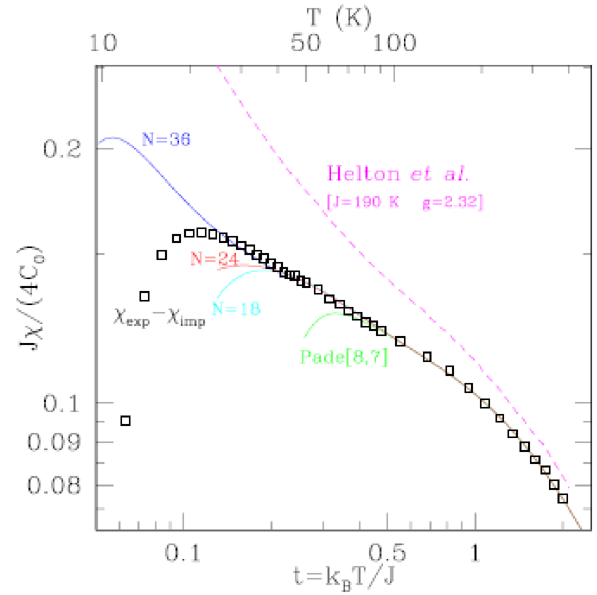
$$\left(T_1\right)^{-1} = A\gamma^2 \int_{\Delta}^{\infty} \rho^2(E)\cdot n(E)\cdot[n(E)+1]dE.$$

($\rho \propto E^{\alpha}$)

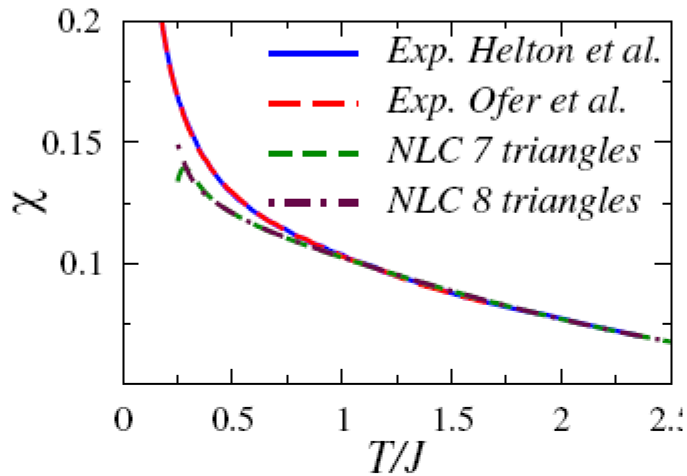
T. Imai *et al.*, PRL **100**, 077203 (2008)

Herbertsmithite: The Magnetization Problem

- Several theoretical work suggest,
  1. DMI causes the canting of the spins.
  2. 3.7% Weakly interacting impurities.
  3. Exchange anisotropy.



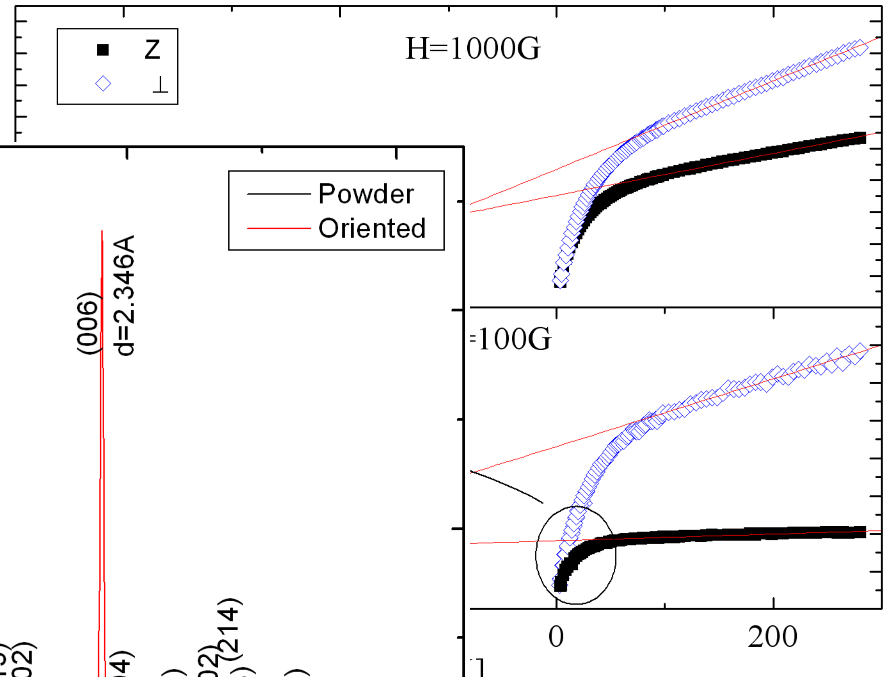
However, none could fully describe the χ behavior.

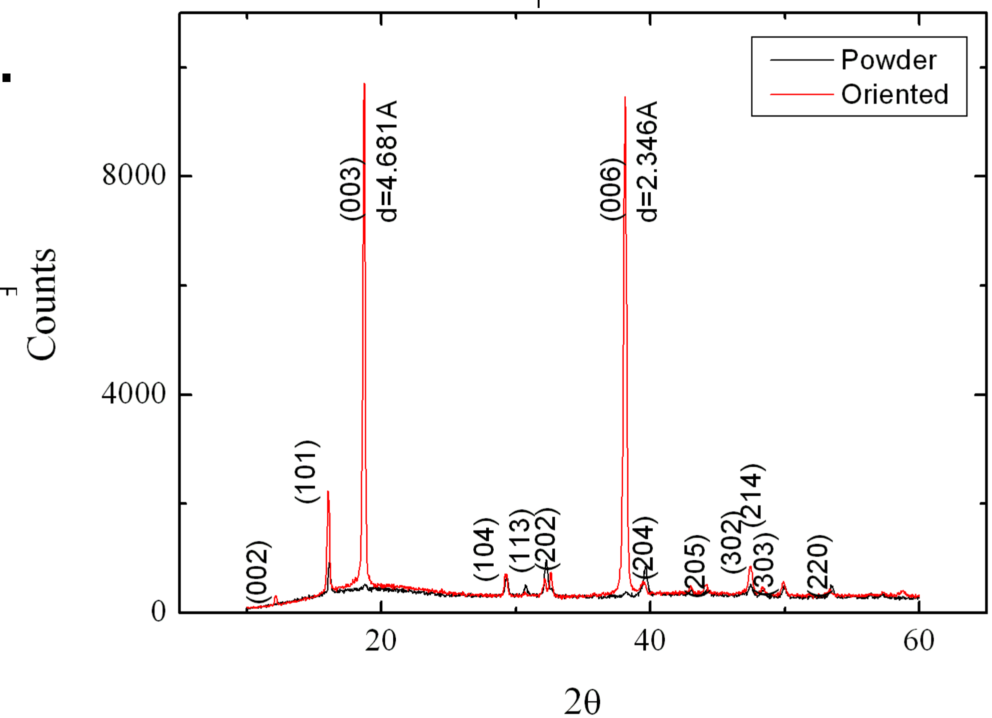


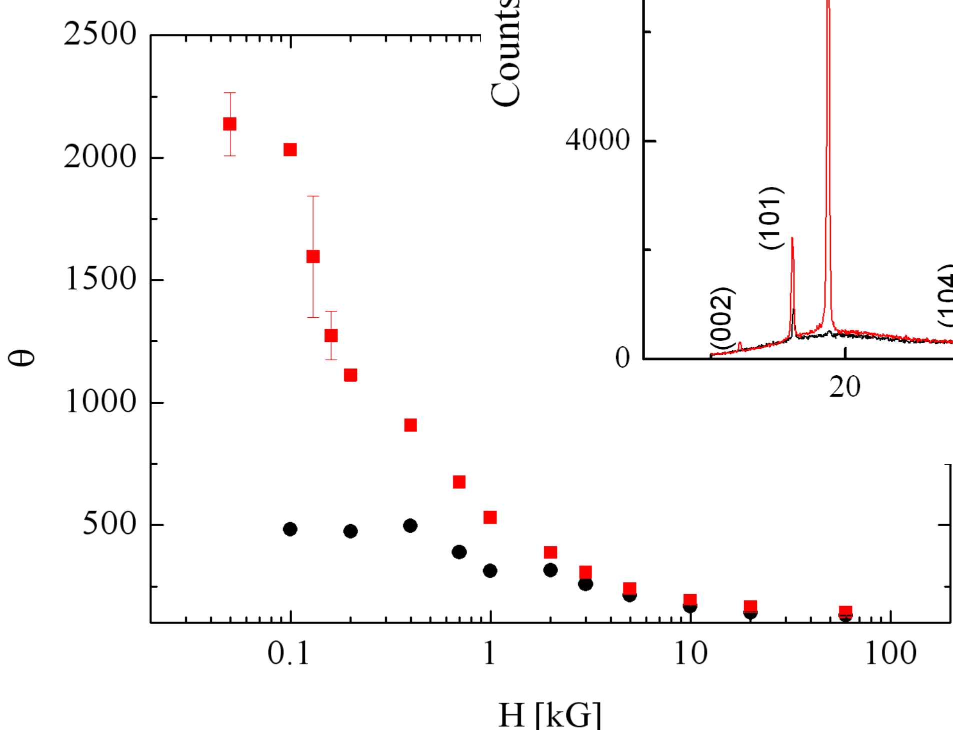
Rigol and Singh, PRB **76**, 184403 (2007).
Rigol and Singh, PRL **98**, 207204 (2007).
Misguich and Sindzingre, Eur. Phys. J. B **59** 305, (2007).
Chern and Tsukamoto, arXiv:cond-mat/0710.1334 (2008).

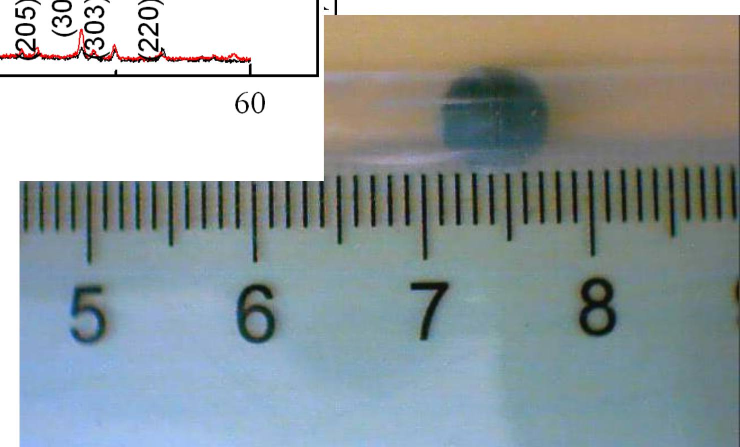
Herbertsmithite: The Magnetization Problem

- Oriented powder spheres.







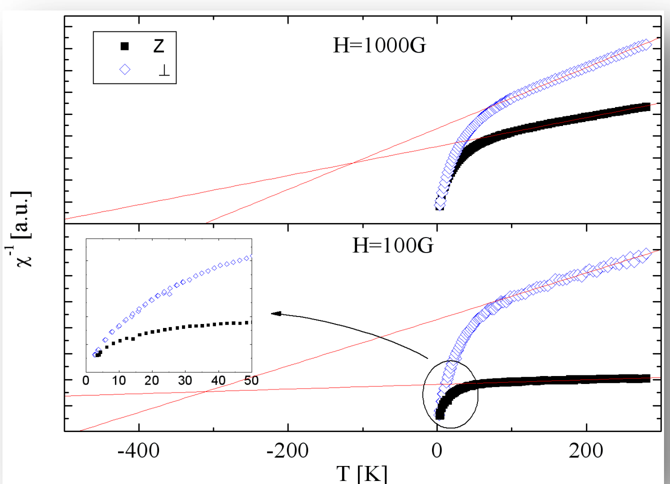


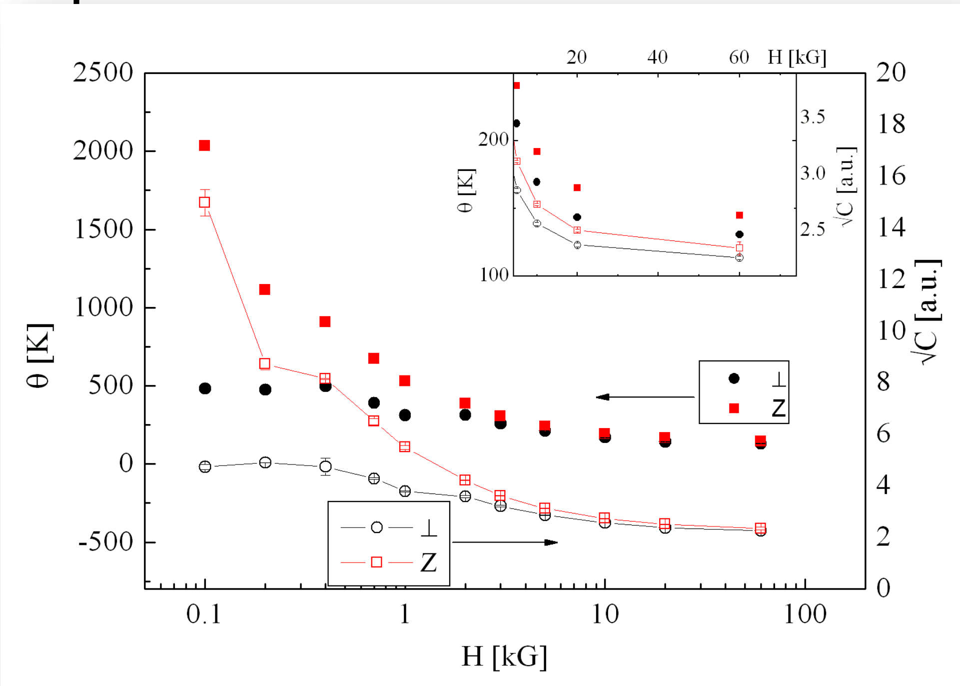
Herbertsmithite: The Magnetization Problem

Recalling, $$\chi^{-1}_{\perp,z} = \frac{3k_B}{(g\mu_B)^2 S(S+1)}(T+\theta_{\perp,z}) \quad .$$

Therefore, high-*T* high-*H* fitting would yield $J_z = k_B\theta_z$, $J_z > J_\perp$

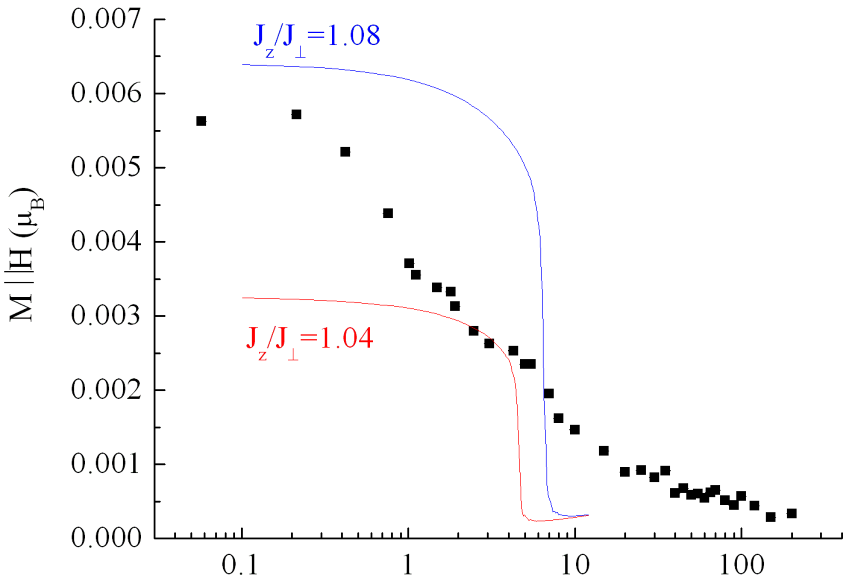
However, the experimental picture calls for additional g-factor anisotropy.

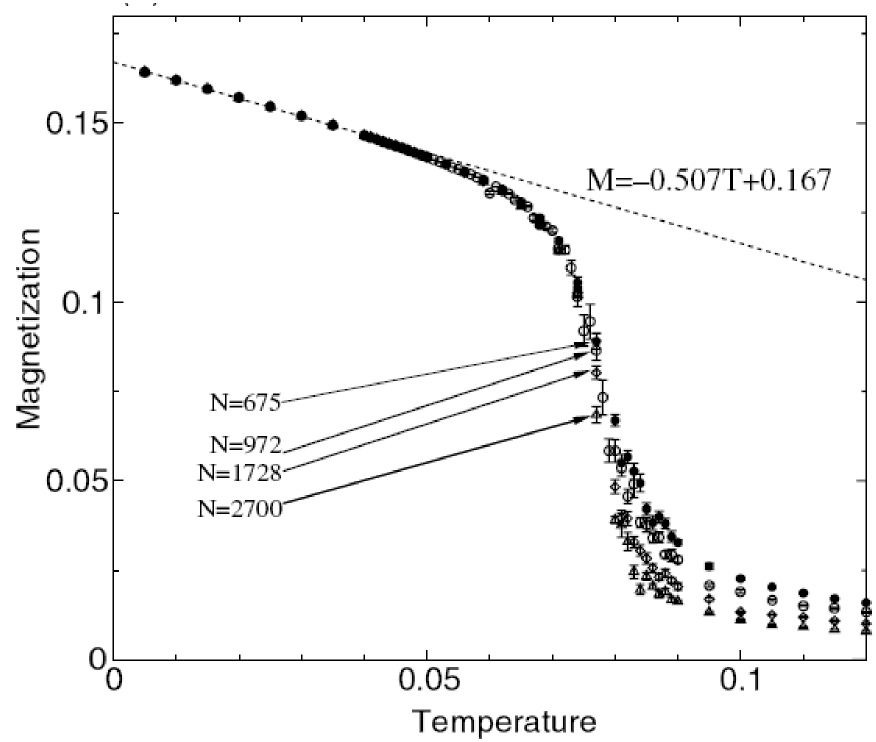




Herbertsmithite: The Magnetization Problem

- Classical AFM with J-anisotropy on the kagome, develops FM order below T_c.





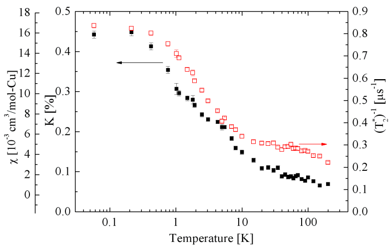
J-anisotropy explains high *T* extreme low *T*, misses at intermediate *T*

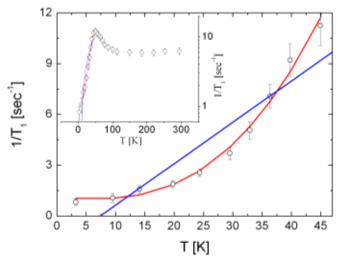
Tanaka and Miyashita, J. Phys.: Condens. Matter **19**, 145256 (2007)

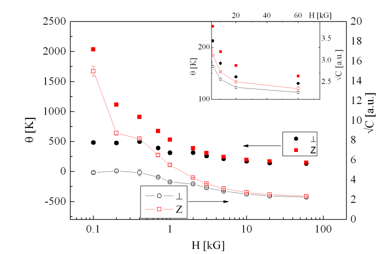
Tovar *et al.*, arXiv:cond-mat/0809.0031 (2008)

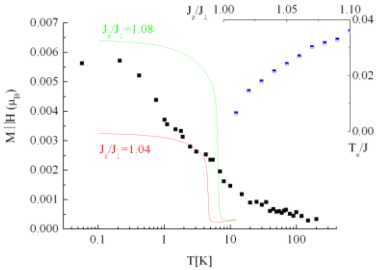
Herbertsmithite: Summary

- Herbertsmithite doesn't freeze, spins still active at 60mK.
- Saturation of χ, indicating no phase transition or singlet formation.
- No sign of lattice deformation.
- Negligible gap to excitations.
- χ and *g* anisotropy. Mean-field suggests $J_z > J_\perp$.
- Behavior suggest Exchange anisotropy and DMI.



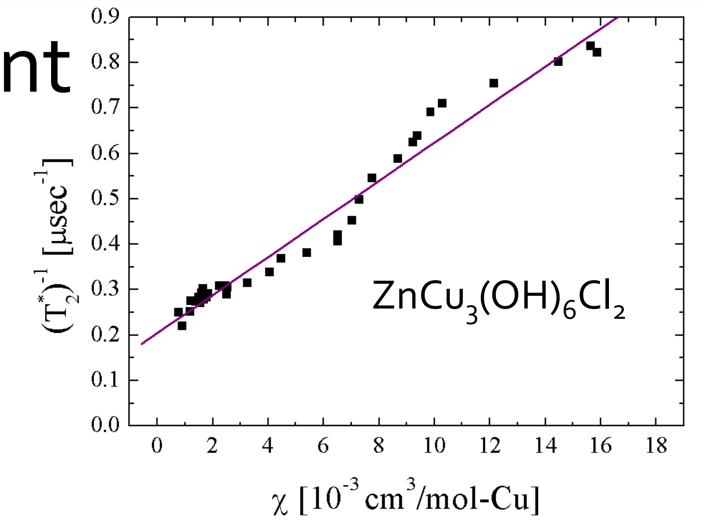




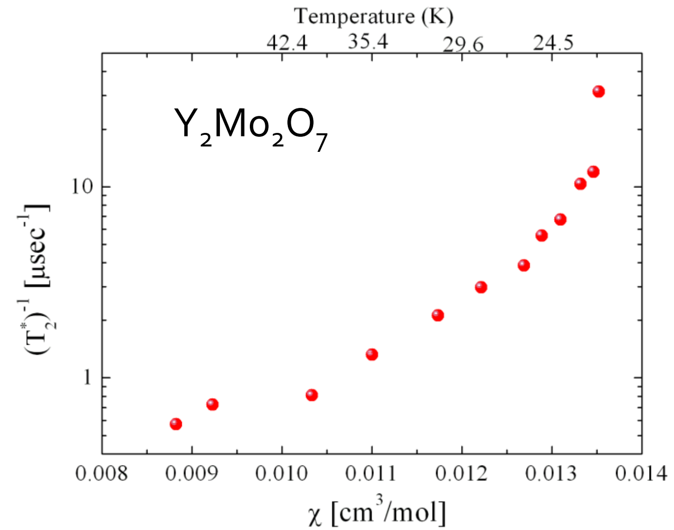


Lattice distortions

- Lattice deformations were absent in the kagome Herbertsmithite, were found in the pyrochlore $Y_2Mo_2O_7$.



Out to look for additional confirmation from NMR

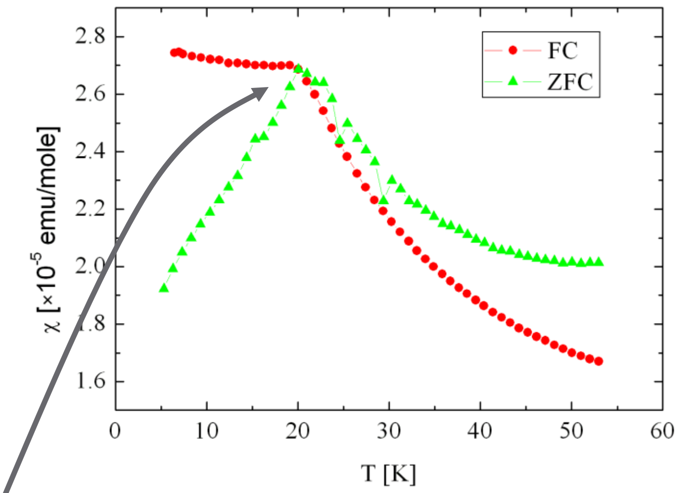


E.S. O.O and A.K., PRL **94**, 237202 (2005).

Spin Glass Pyrochlore Y2Mo2O7

- Mo^{4+} (S=1) forming a pyrochlore.
- Remarkable SG characteristics, with $T_g \approx 20\text{K}.$
- Data indicate high frustration,

$$\theta_{CW} \approx -200\text{K} \to f = 10.$$

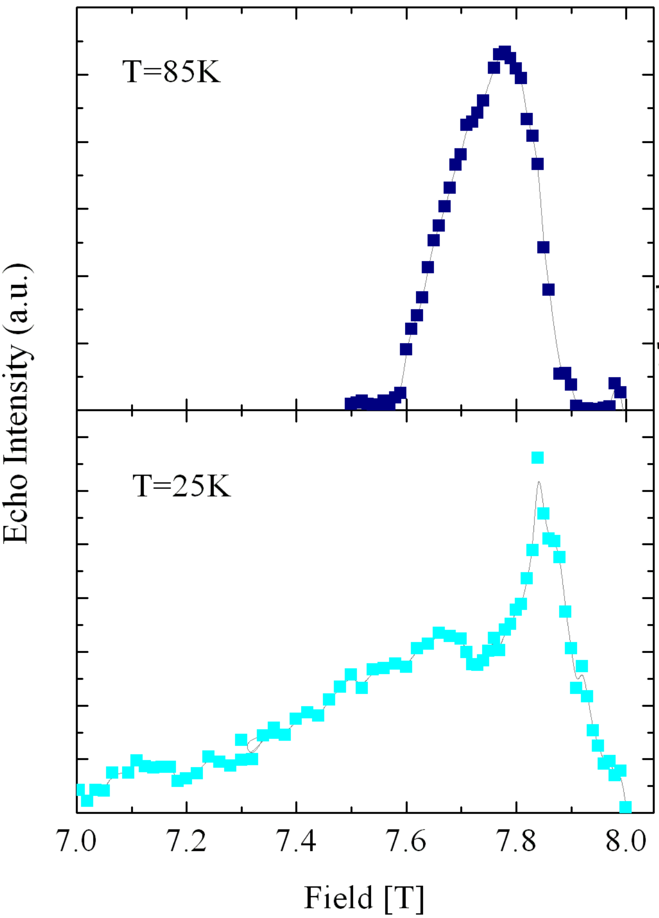


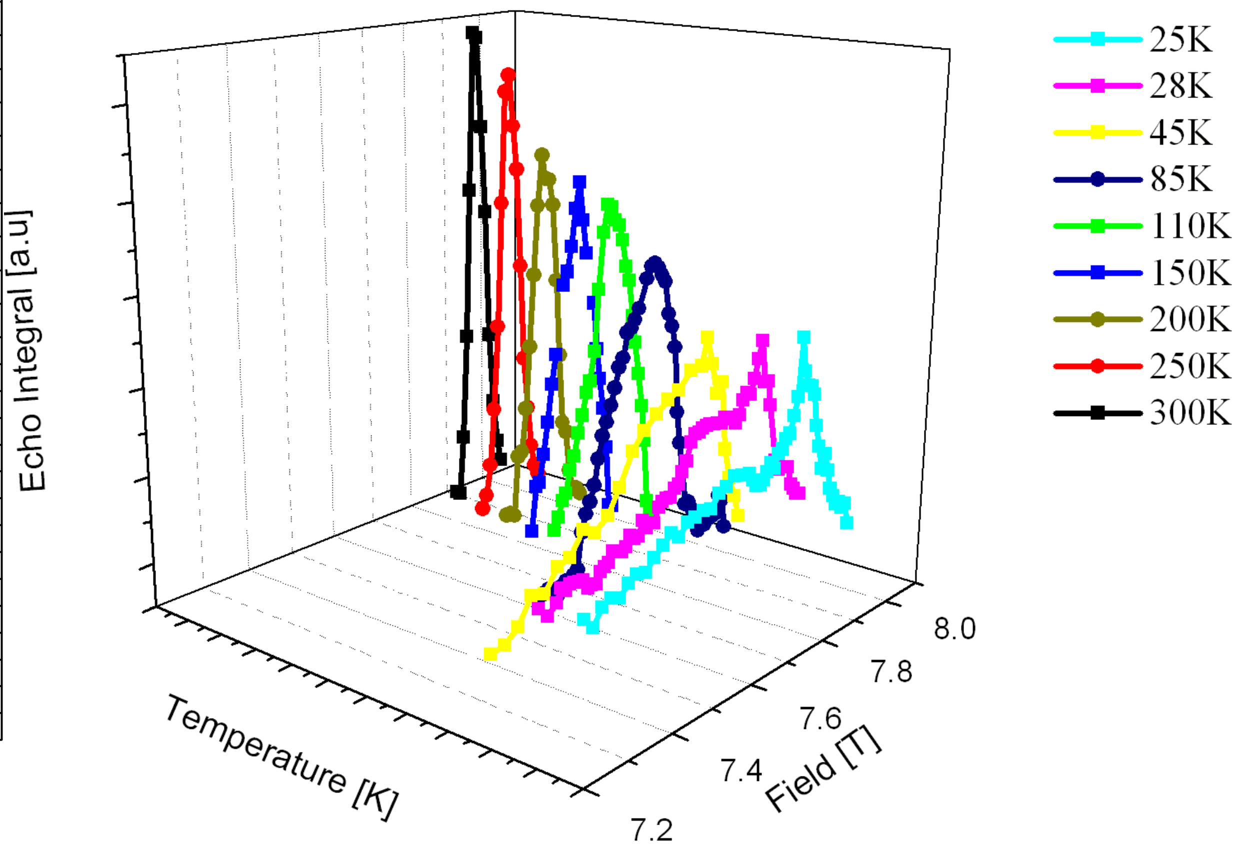
Freezing is surprising,

This should not happen in a pure Heisenberg model.

Spin Glass Pyrochlore Y2Mo2O7 Y NMR

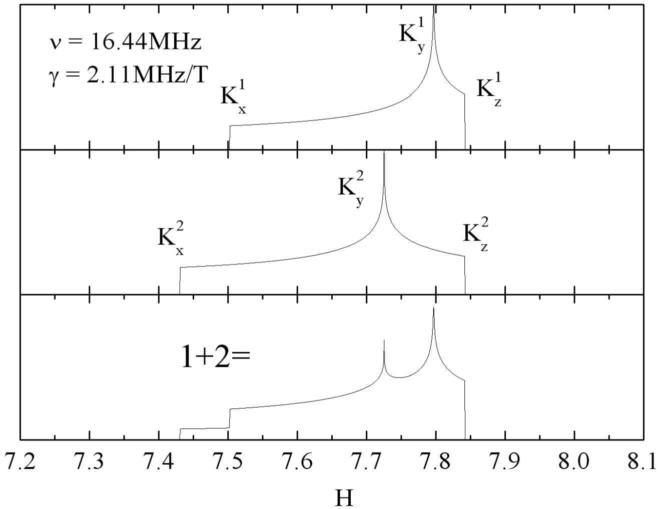
- ^{87}Y NMR line shape vs T.

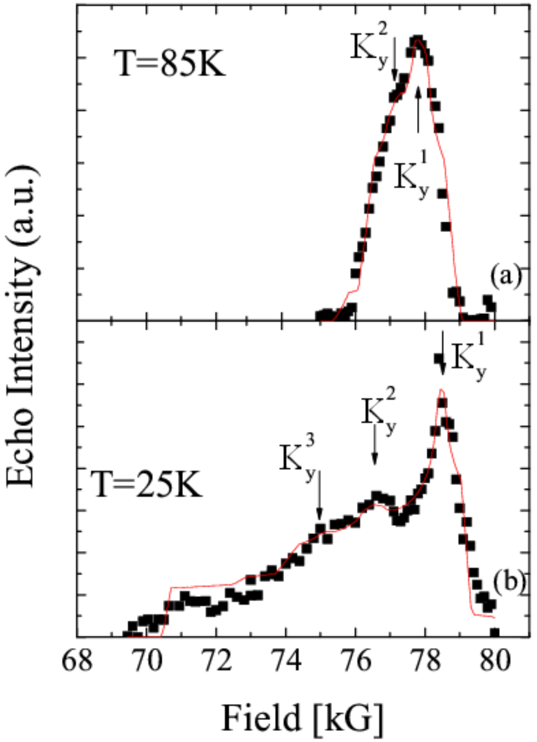




Spin Glass Pyrochlore $Y_2Mo_2O_7$ Powder Average NMR

- The NMR line powder-average,

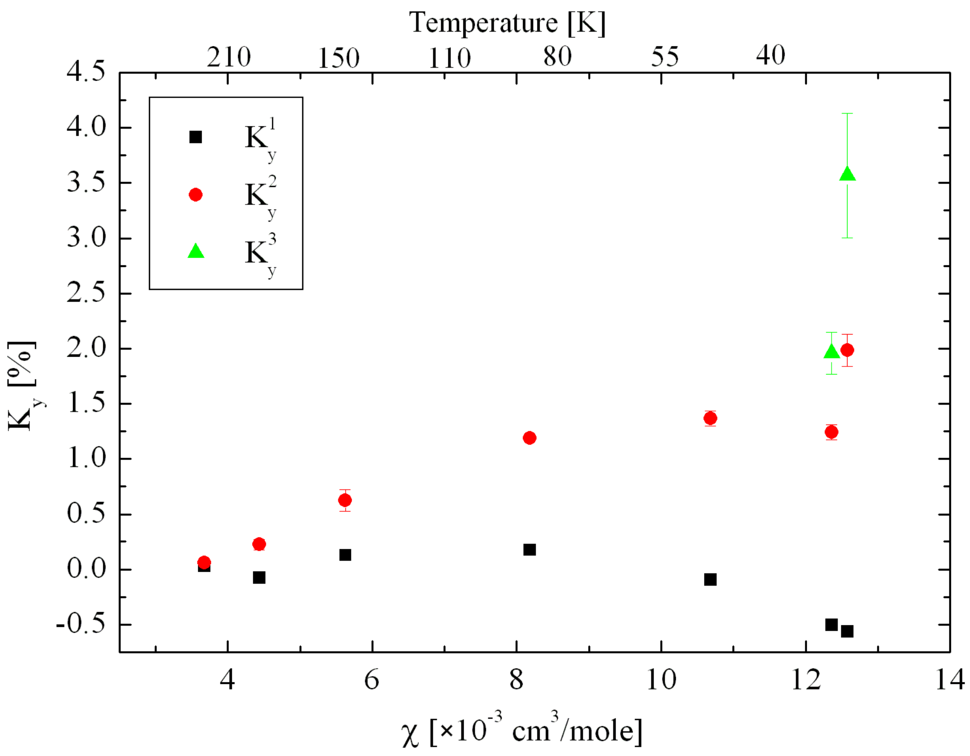


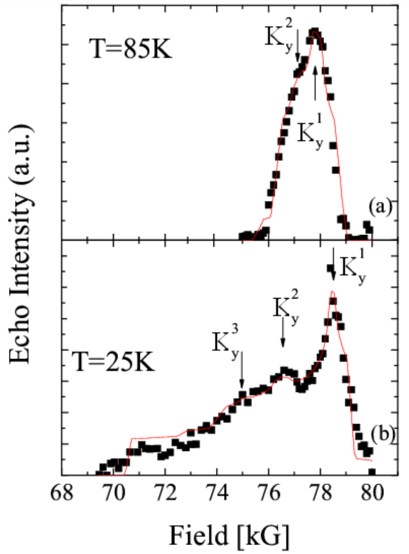


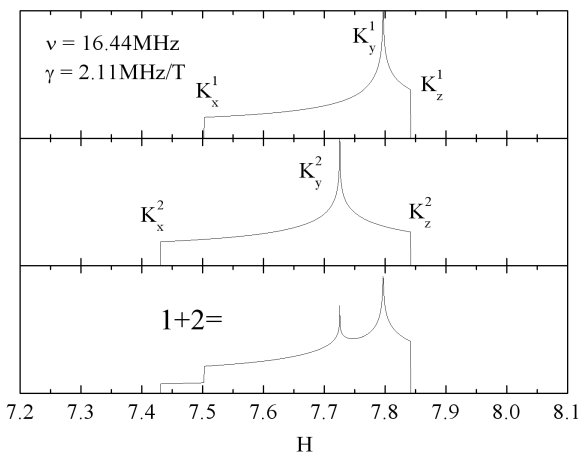
- Thus, for each *T*, it is possible to extract **K**.

Spin Glass Pyrochlore Y2Mo2O7 Y NMR

- *K* is not linear with χ indicating lattice deformation.

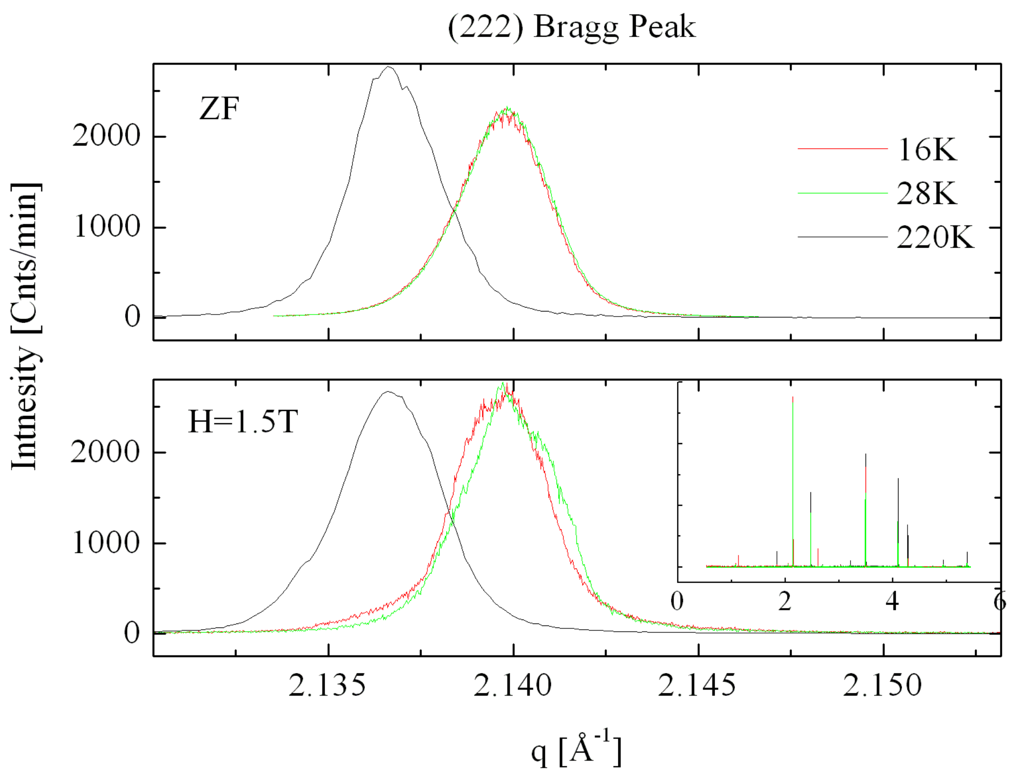


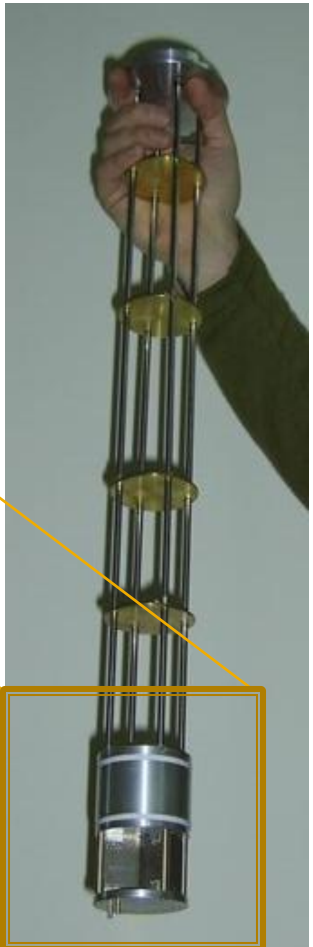


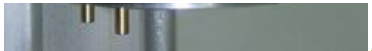


Spin Glass Pyrochlore Y2Mo2O7 Field dependent X-ray diffraction

- High resolution x-ray powder diffraction reveal a single phase for any *T*.







Spin Glass Pyrochlore Y2Mo2O7 Data frustration

- $Y_2Mo_2O_7$:
  Unexpected freezing,
  X-ray data conflicts μSR,NMR data

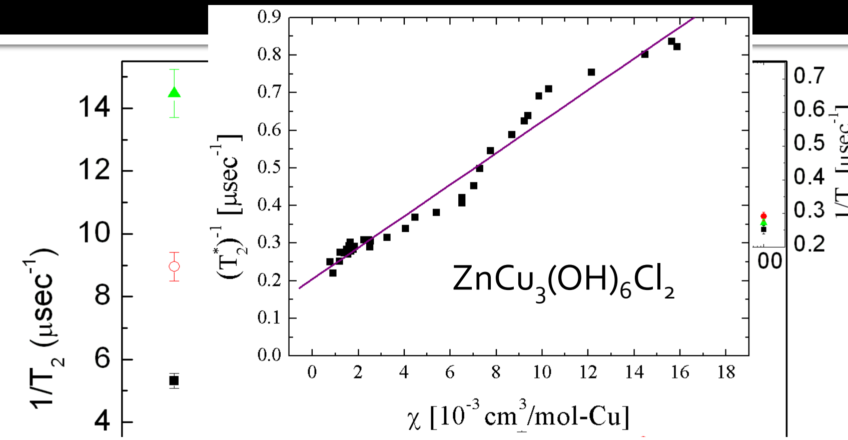
- Z

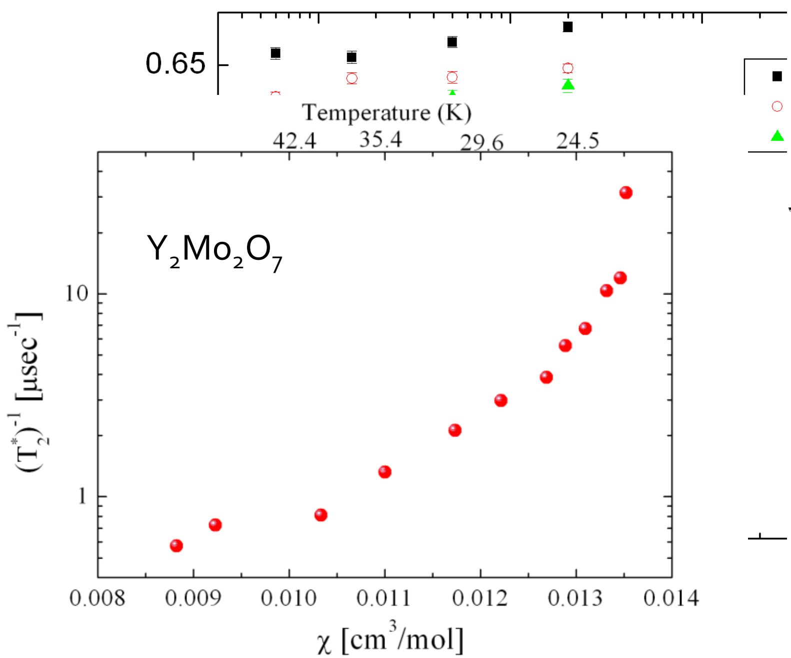
We return to our simplest example, $Tb_2Ti_2O_7$ and confirm that indeed it doesn't experience lattice deformation.

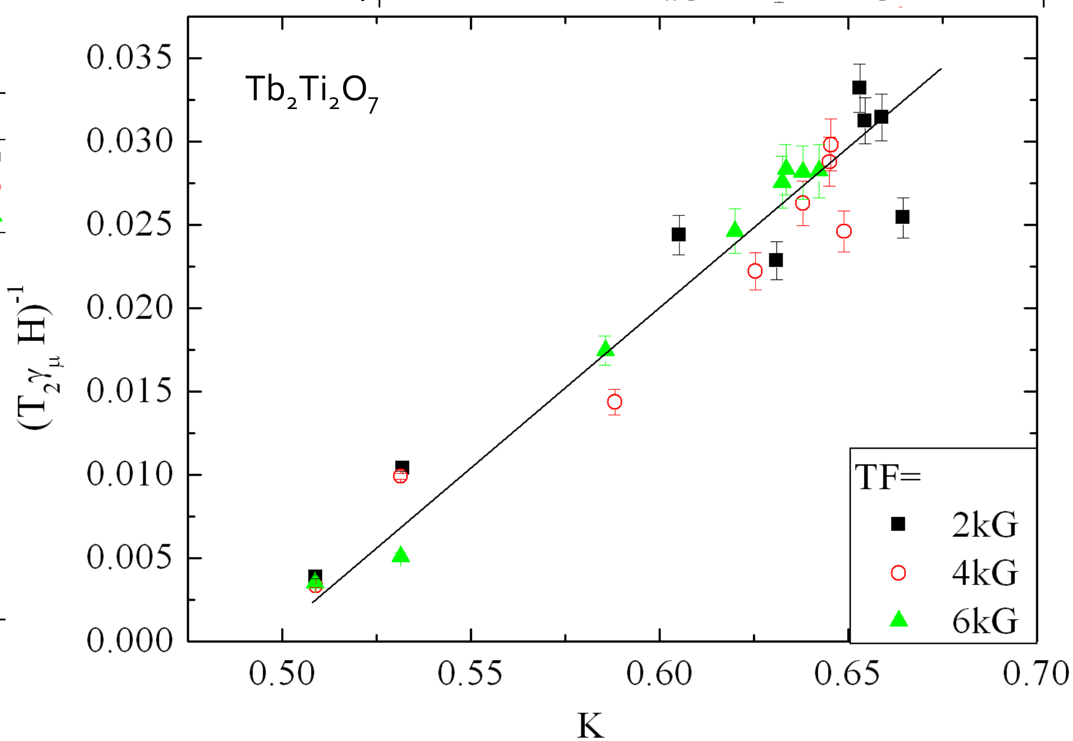
Peculiar Low-*T* susceptibility.

Cooperative Paramagnet Tb2Ti2O7 μSR

- Observing the TF relaxation and the muon shift K.







Pyrochlores $Y_2Mo_2O_7$, $Tb_2Ti_2O_7$: Summary

$Y_2Mo_2O_7$:

- Spin-glass transition at T_g=22K.
- Formation of 2 (3?) Y inequivalent sites as $T \rightarrow T_g$ suggesting a magneto-elastic deformation.

$Tb_2Ti_2O_7$:

- Cooperative Paramagnetism (no spin freezing) at T>60mK.
- No μSR indication for lattice deformation.

Summary

- Geometrically Frustrated Magnets posses an exotic playground where interactions beyond Heisenberg can be examined.
- There are more experimental dilemmas than answers.