KITP – **Moments and Multiplets in Mott Materials (August 13 - December 14, 2007)**

Experimentally Motivated Problems in Some Highly Frustrated Rare-Earth Magnetic Materials

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Collaborators

Theory

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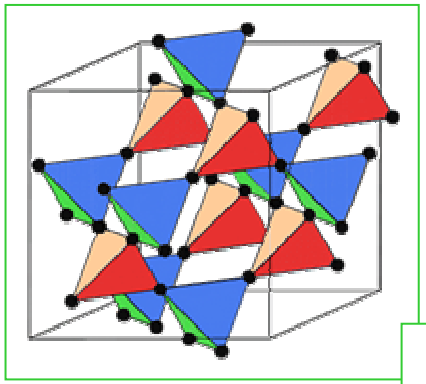
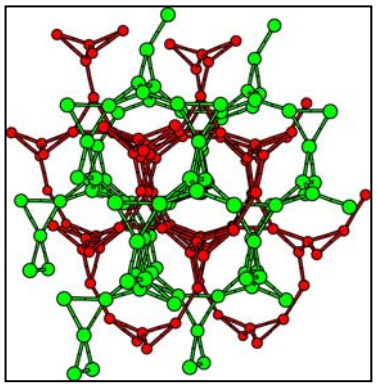
- Pawel Stasiak (Waterloo)
- Taras Yavors'kii (Waterloo)
- Hamid Molavian (Waterloo)
- Adrian del Maestro (Harvard)
- Francois Vernay (Waterloo → PSI)
- Matt Enjalran (USC)
- Ying-Jer Kao (Nat. U. Taiwan,Tapei)
- Jean-Yves Fortin (CNRS, Strasbourg)
- Benjamin Canals (CNRS, Grenoble)

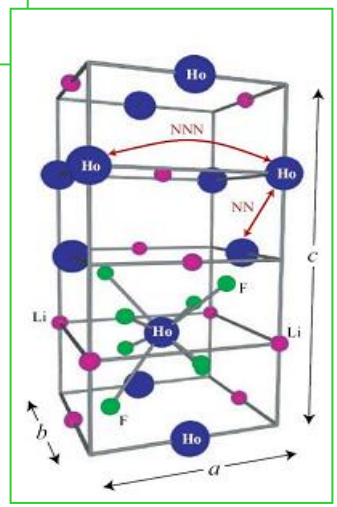
Experiment

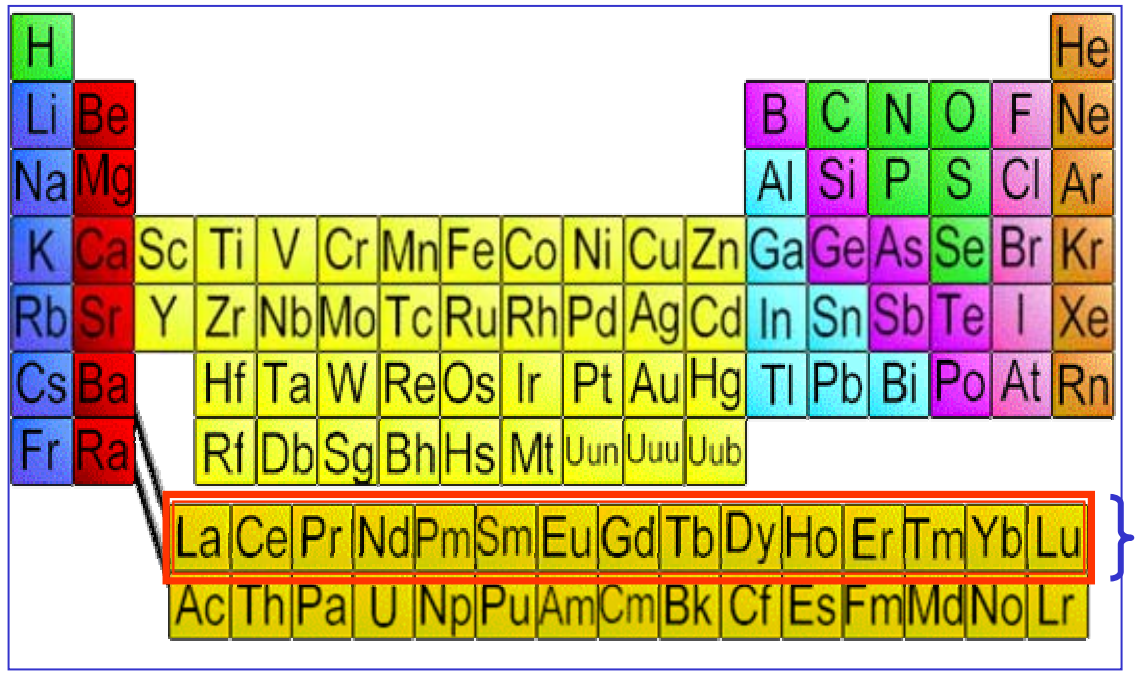
- Steve Bramwell (U.Coll. London)
- Tom Fennell (U. Coll. London)
- Jan Kycia (Waterloo)
- Jeff Quilliam (Waterloo)
- Kate Ross (Waterloo)
- Linton Corruccini (UC Davis)
- Oleg Petrenko (Warwick)

Insulating Rare-Earth Magnetic Materials









- 4f orbitals are burried under 5s, 6s, 5d orbitals: exchange interactions J_{ij} are "small": $\theta_{CW} \sim 10^0 — 10^1$ K
- Re^{3+} can have large magnetic moments $\mu \sim 10^0 — 10^1\ \mu_B$
- Magnetic dipolar interactions, $D \sim 10^{-1} — 10^0$ K $\sim \theta_{CW}$

GENERAL SPIN HAMILTONIAN

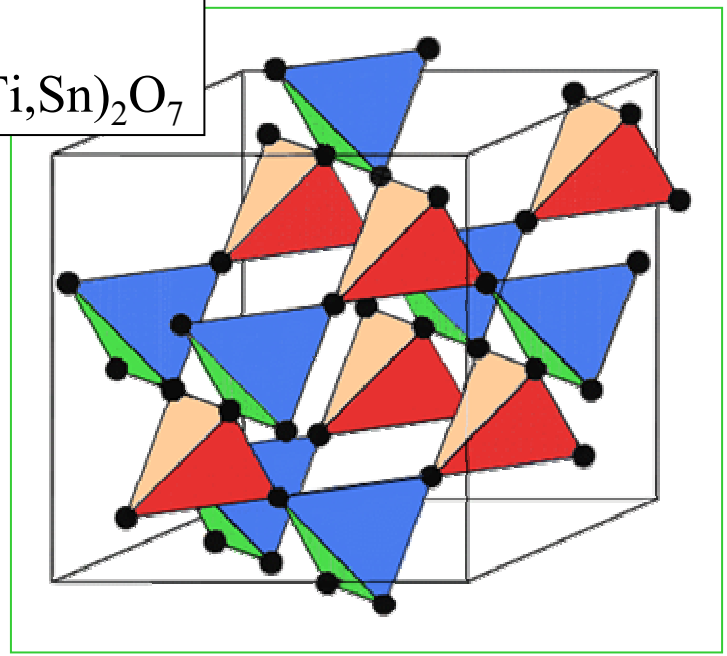
$$H = -\sum_{j>i} J_{ij}\ \vec{\mathrm{J}}_i \bullet \vec{\mathrm{J}}_j$$

$$+\ \frac{\mu_0}{4\pi}(g\mu_{\mathrm{B}})^2 \sum_{j>i} \frac{\vec{\mathrm{J}}_i \bullet \vec{\mathrm{J}}_j}{\left|\vec{r}_{ij}\right|^3} - \frac{3(\vec{\mathrm{J}}_i \bullet \vec{r}_{ij})(\vec{\mathrm{J}}_j \bullet \vec{r}_{ij})}{\left|\vec{r}_{ij}\right|^5}$$

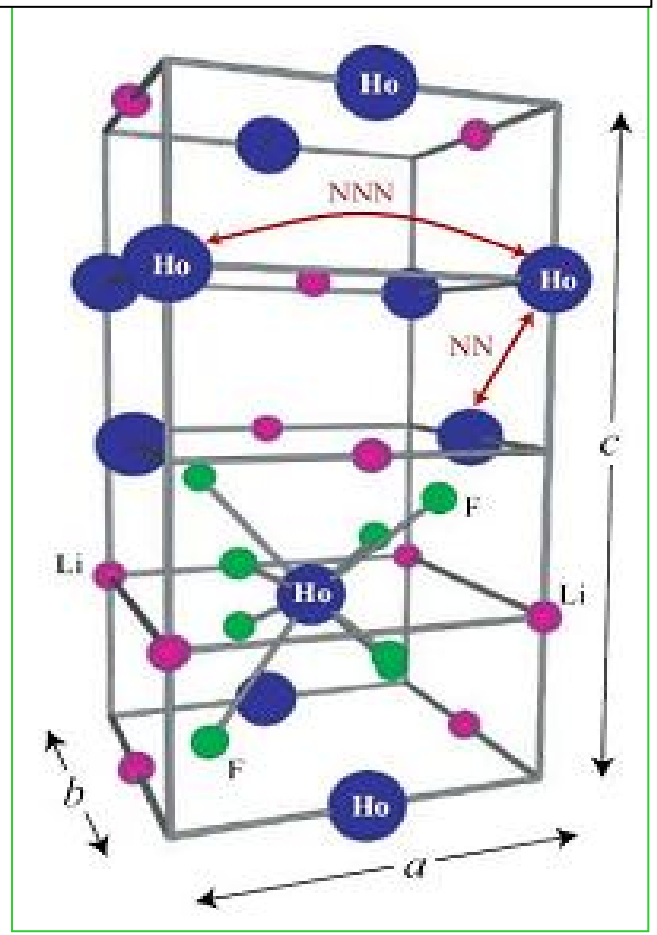
$$+\ V_{\mathrm{CF}}(\mathrm{J}_i^{\alpha}) - g\mu_{\mathrm{B}} \sum_i \vec{\mathrm{J}}_i \bullet \vec{\mathrm{B}}$$

Crystal field part of *H*. This is a single-particle part of the Hamiltonian. it describes how the local electrostatic/chemical environment lifts the otherwise (2J+1) degeneracy of the otherwise free rare-earth ion.

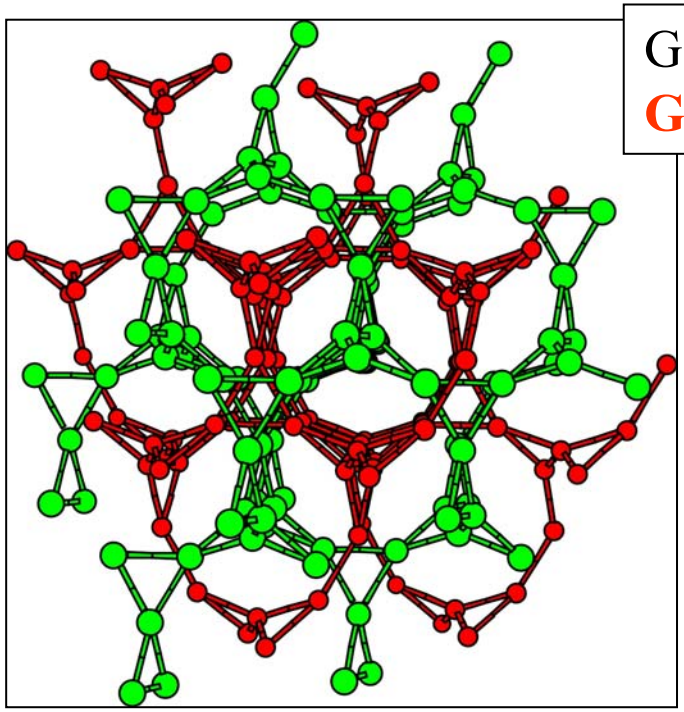
Pyrochlore lattice:
$(\mathbf{Gd,Tb,Ho,Dy})_2(Ti,Sn)_2O_7$



Body-centered tetragonal lattice:
$Li\mathbf{Ho}_xY_{1-x}F_4$

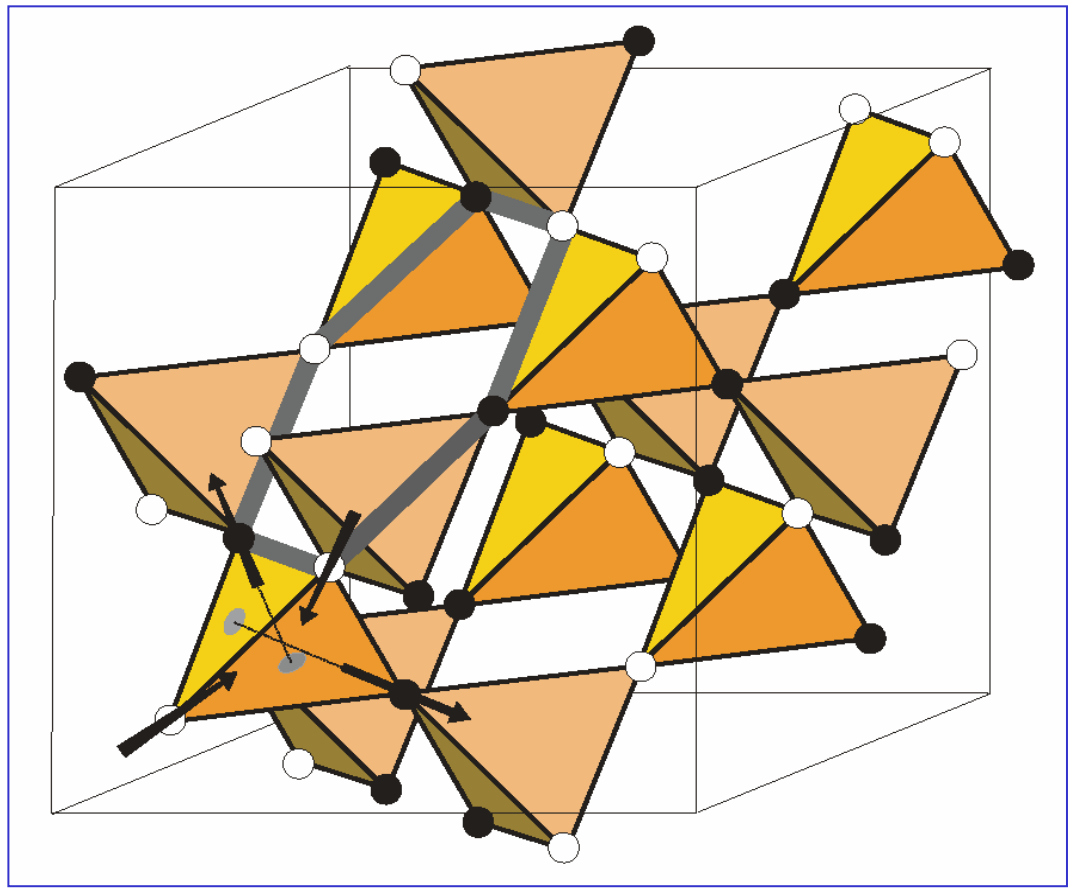


Garnet lattice:
$\mathbf{Gd}_3Ga_5O_{12}$



Spin Ice

Pyrochlore lattice:
$(\mathbf{Ho,Dy})_2(Ti,Sn)_2O_7$

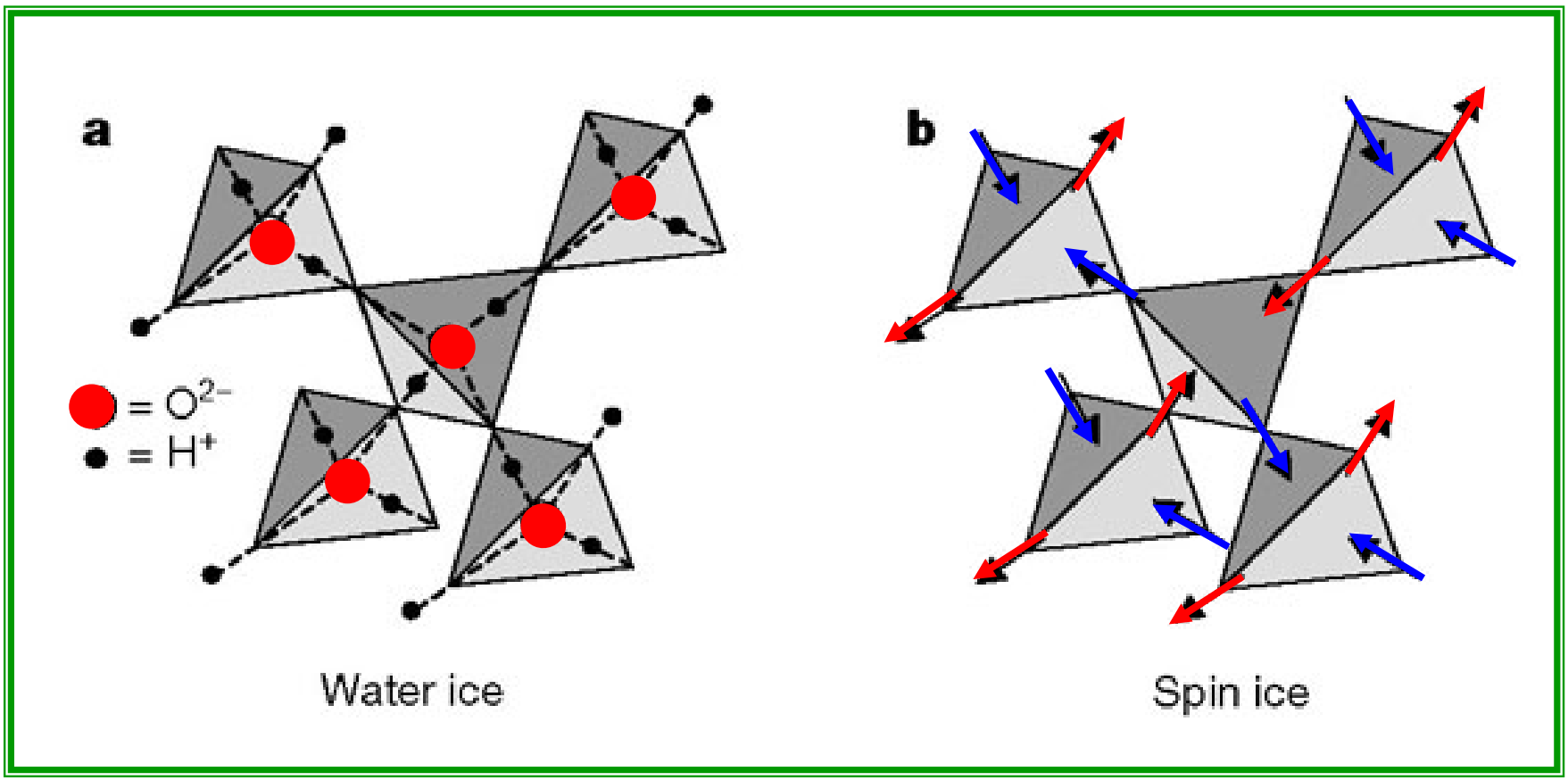


VOLUME 79, NUMBER 13 PHYSICAL REVIEW LETTERS 29 SEPTEMBER 1997

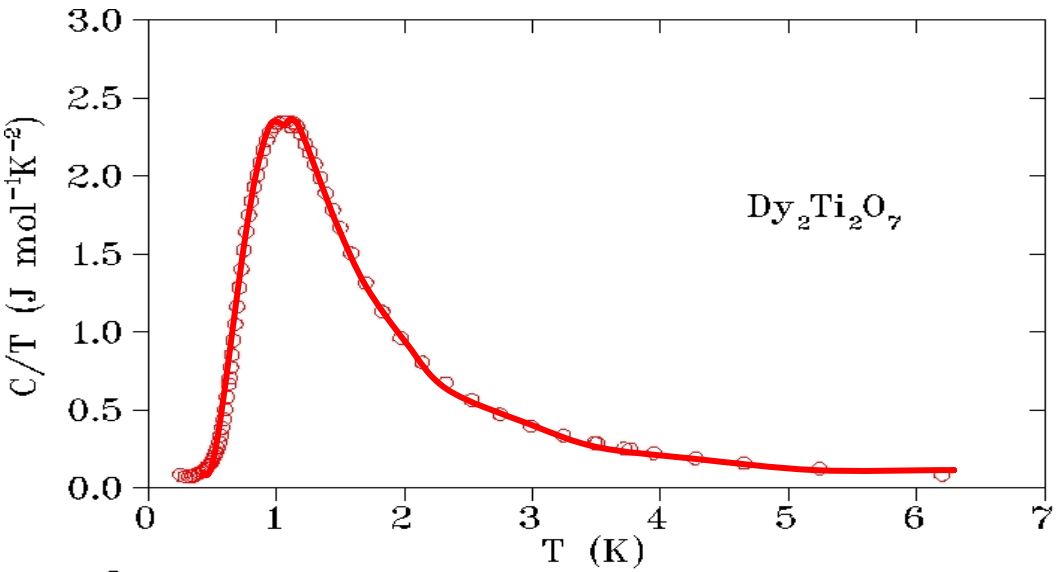
Geometrical Frustration in the Ferromagnetic Pyrochlore $Ho_2Ti_2O_7$

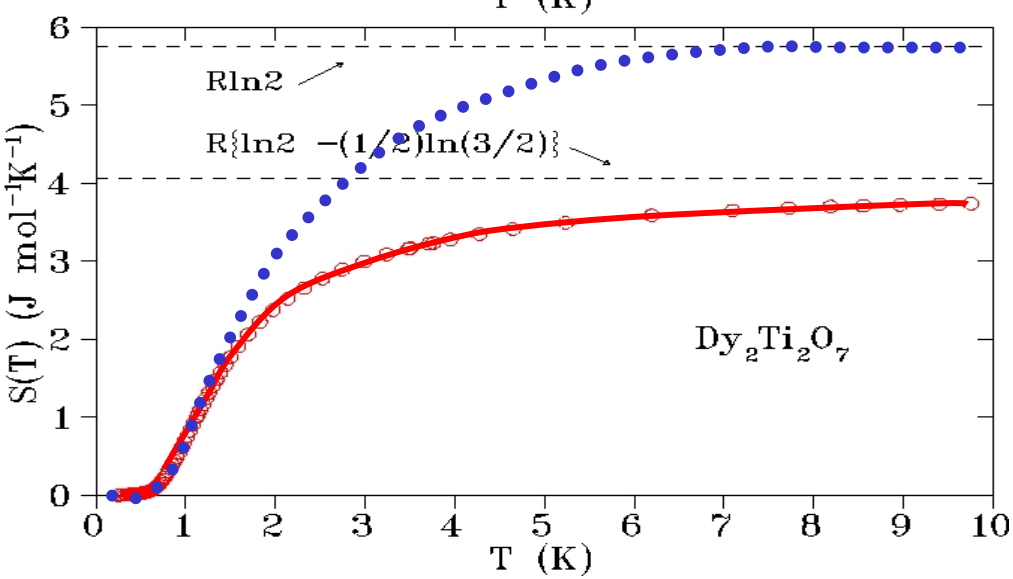
M. J. Harris,[1] S. T. Bramwell,[2] D. F. McMorrow,[3] T. Zeiske,[4] and K. W. Godfrey[5]

$Ho_2Ti_2O_7$, $Dy_2Ti_2O_7$, $Ho_2Sn_2O_7$, $Dy_2Sn_2O_7$,



Real materials show manifestations of Pauling's ground state entropy magnetic analogues of water ice





$$S(T_2) - S(T_1) = \int_{T_1}^{T_2} \frac{C(T)}{T} dT$$

Ramirez *et. al*, Nature **399**, 333 (1999)

Dipolar Ising Spin Ice Model

$$H = -J \sum_{\langle i,j \rangle} \left(\hat{z}_i \bullet \hat{z}_j \right) \sigma_i^{z_i} \sigma_j^{z_j} +$$

$$D \sum_{i>j} \frac{\hat{z}_i \bullet \hat{z}_j}{\left| \vec{r}_{ij} \right|^3} - \frac{3(\hat{z}_i \bullet \vec{r}_{ij})(\hat{z}_j \bullet \vec{r}_{ij})}{\left| \vec{r}_{ij} \right|^5} \sigma_i^{z_i} \sigma_j^{z_j}$$

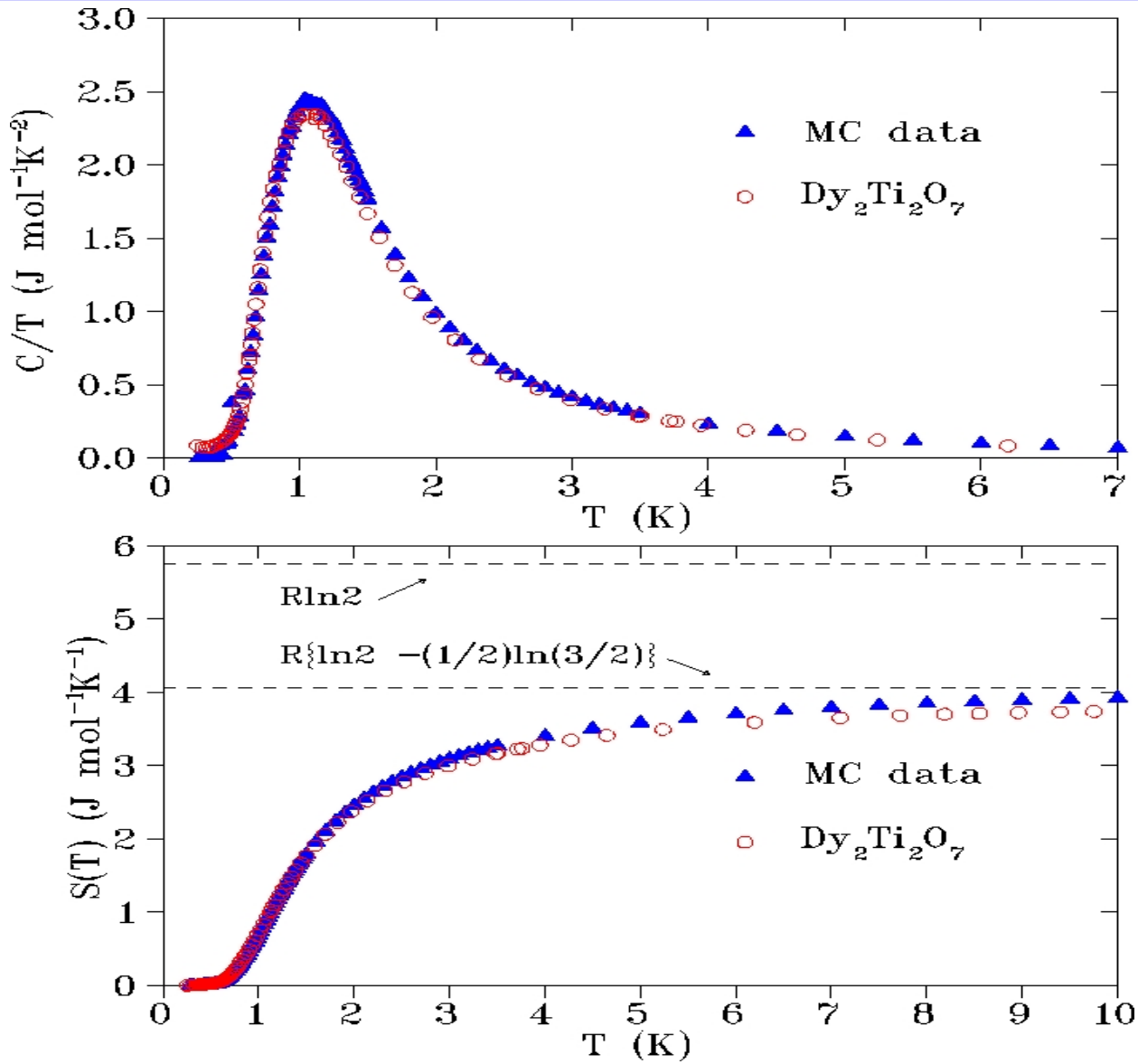
Not known

Known

Moved from J=15/2 → S=1/2 effective classical Ising spin

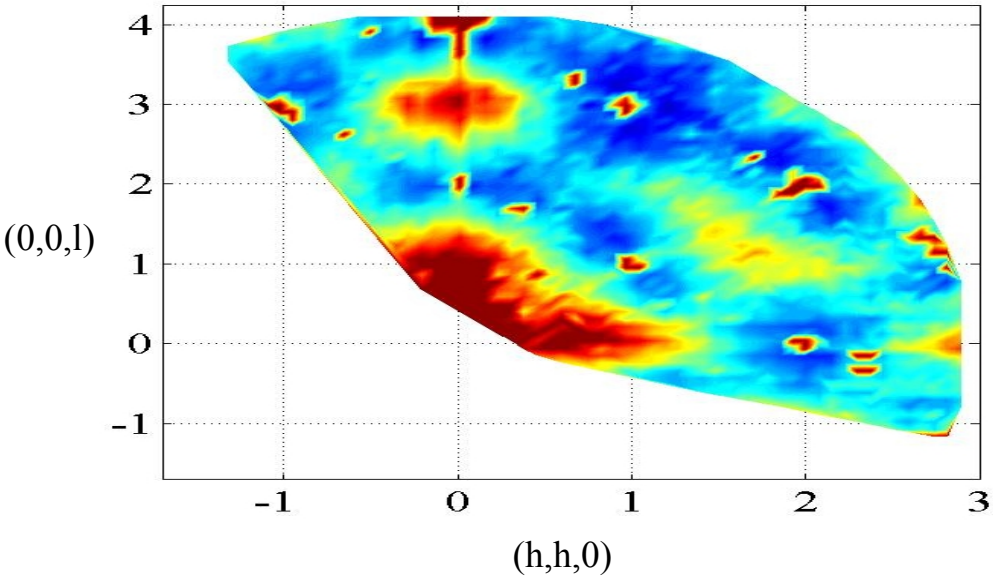
den Hertog and Gingras, *Phys. Rev. Lett.* **84**, 3430 (2000).

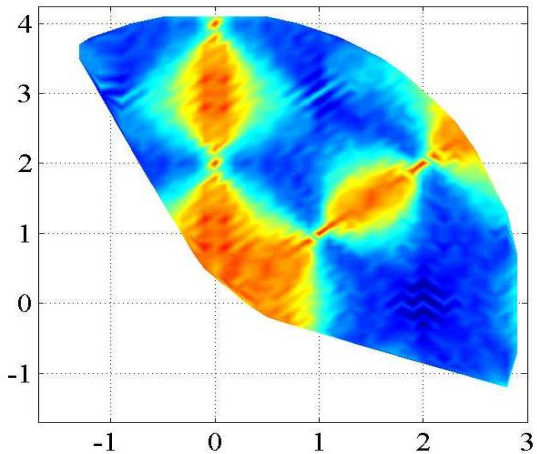
Real materials show manifestations of Pauling's ground state entropy magnetic analogues of water ice



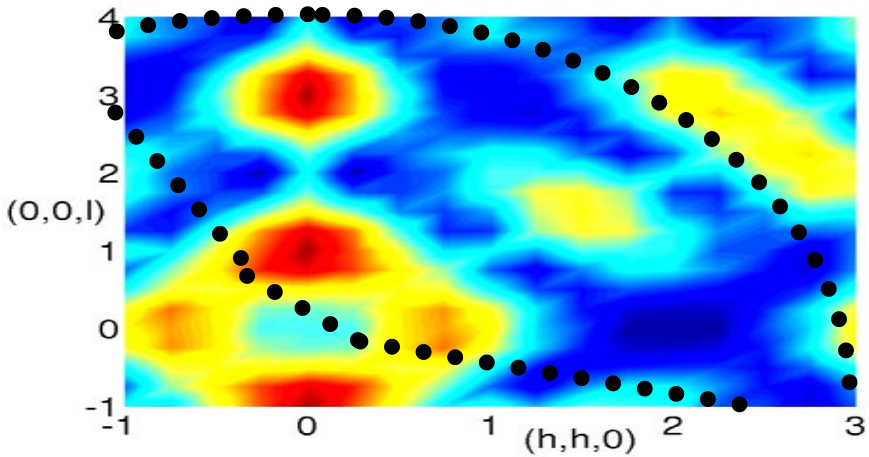
den Hertog and Gingras, *Phys. Rev. Lett.* **84**, 3430 (2000).

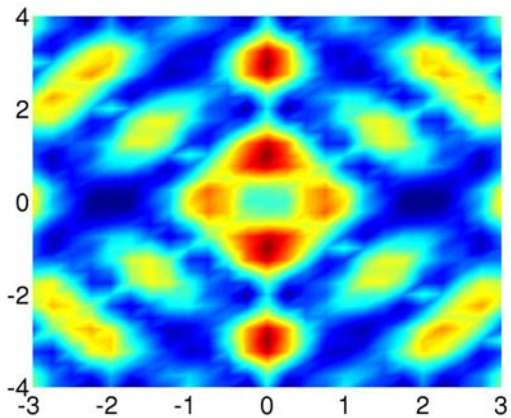
Neutron scattering in $Ho_2Ti_2O_7$





nn-spin ice

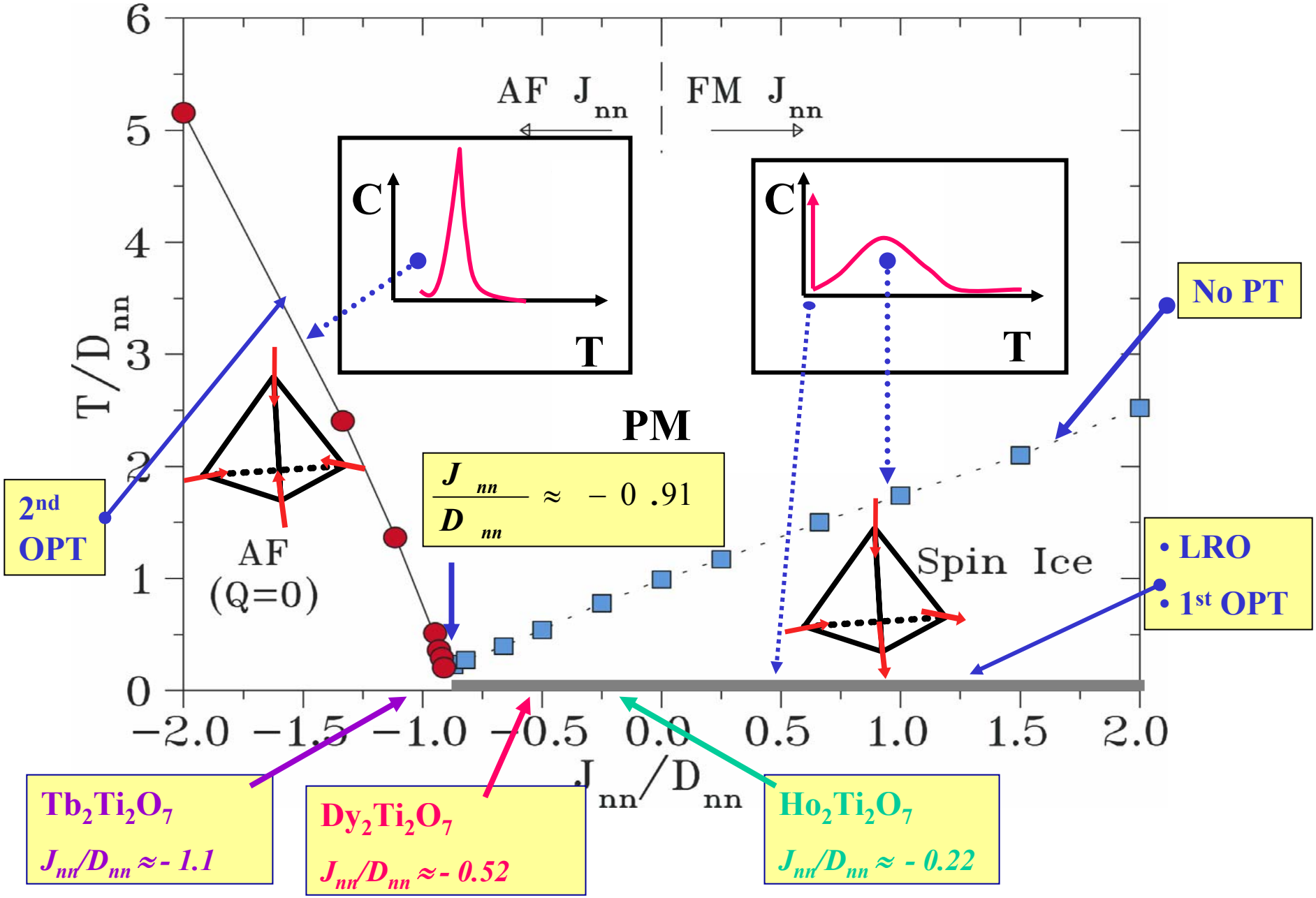




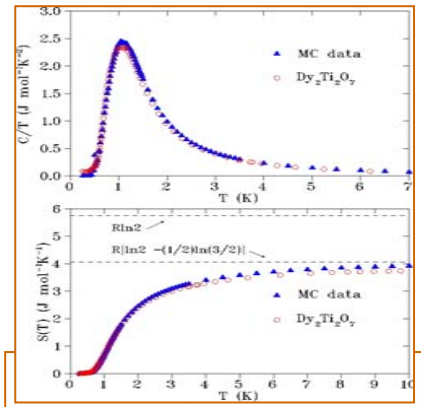
dipolar spin ice

S. T. Bramwell, *et al.*, Phys. Rev. Lett. **87**, 047205 (2001).

Monte Carlo Phase Diagram of the Dipolar Spin Ice Model



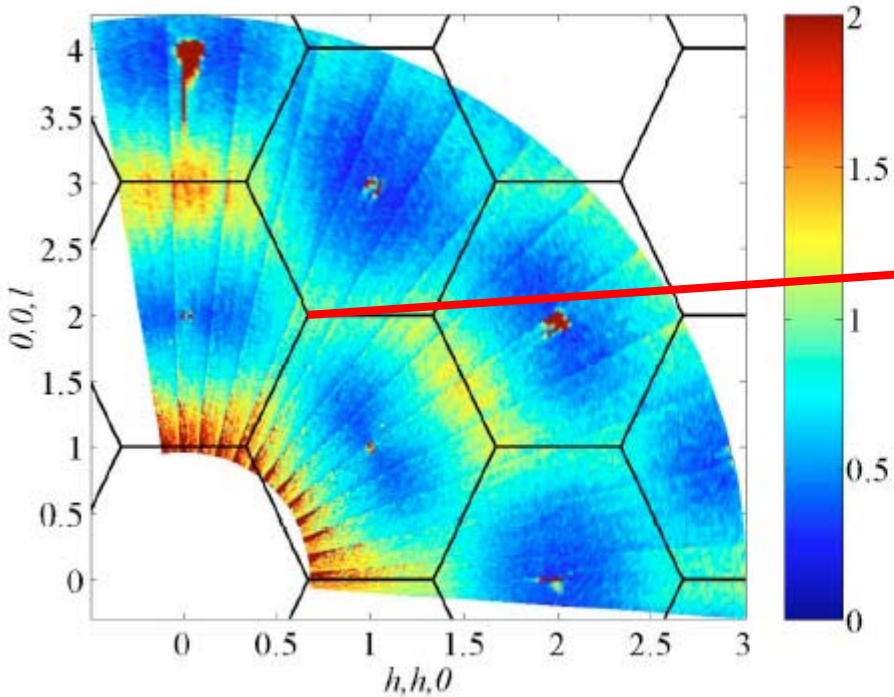
Neutron Scattering in $Dy_2Ti_2O_7$

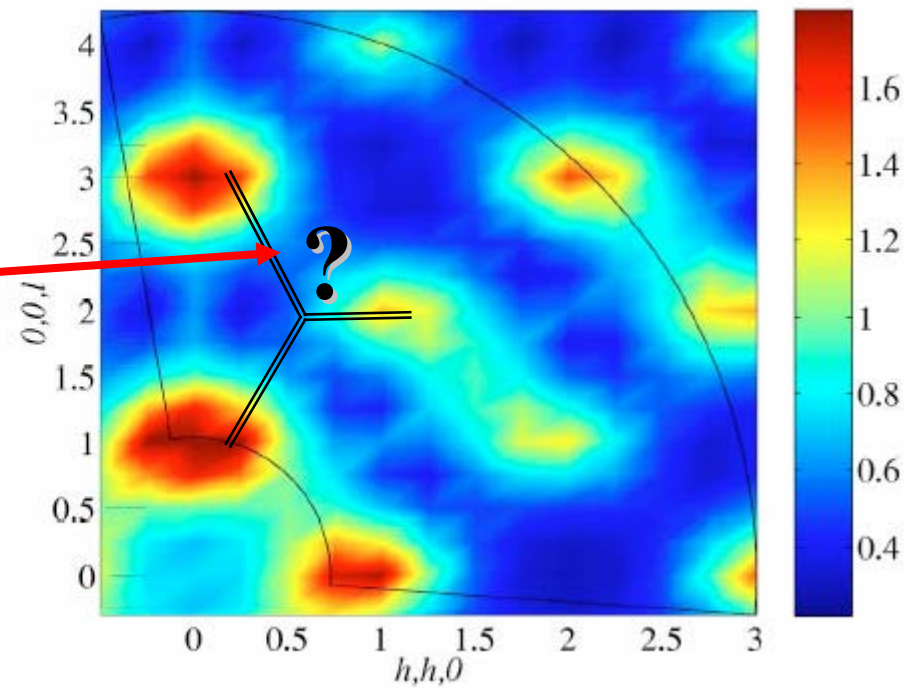


Gives exchange J_{nn}

Brillouin zone boundary scattering

T=400 mK





Experiment

Monte Carlo

NATURE | VOL 418 | 22 AUGUST 2002 | www.nature.com/nature

Emergent excitations in a geometrically frustrated magnet

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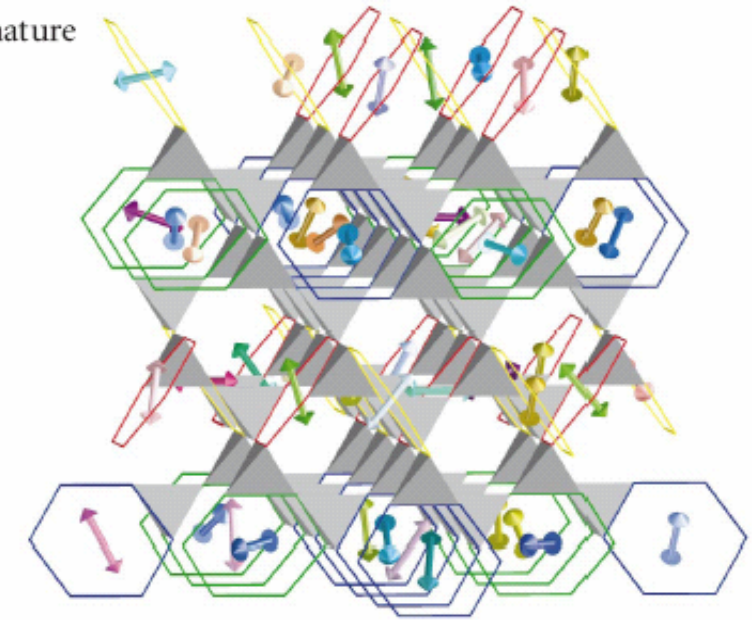
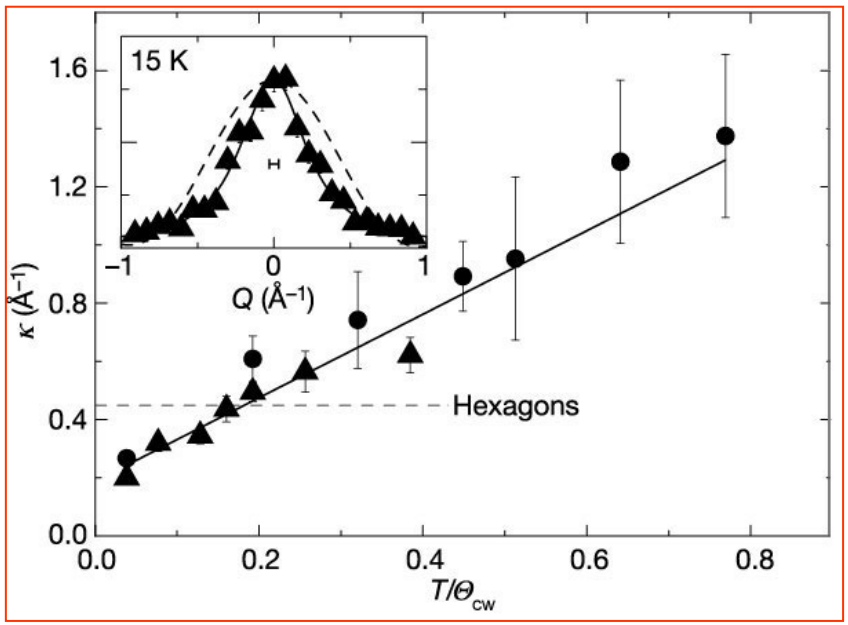
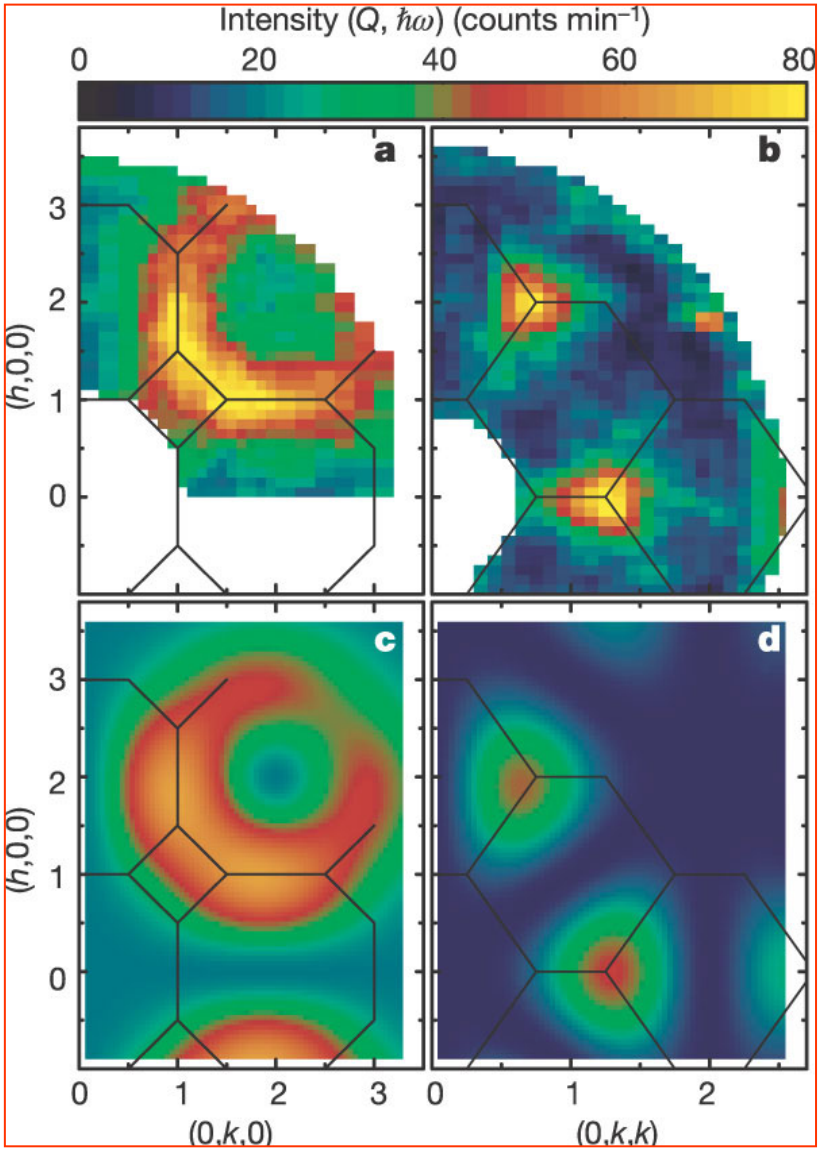


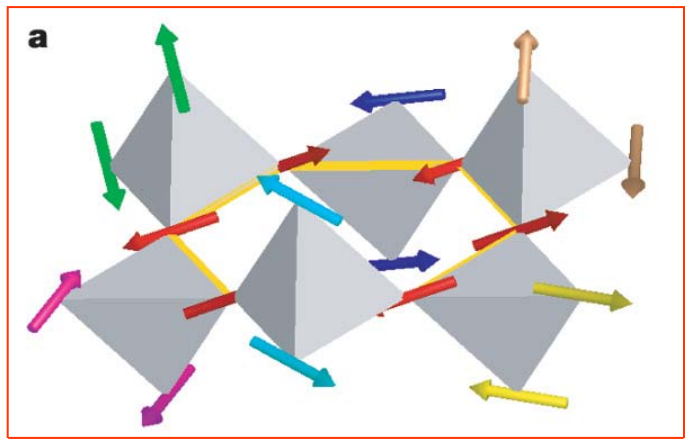


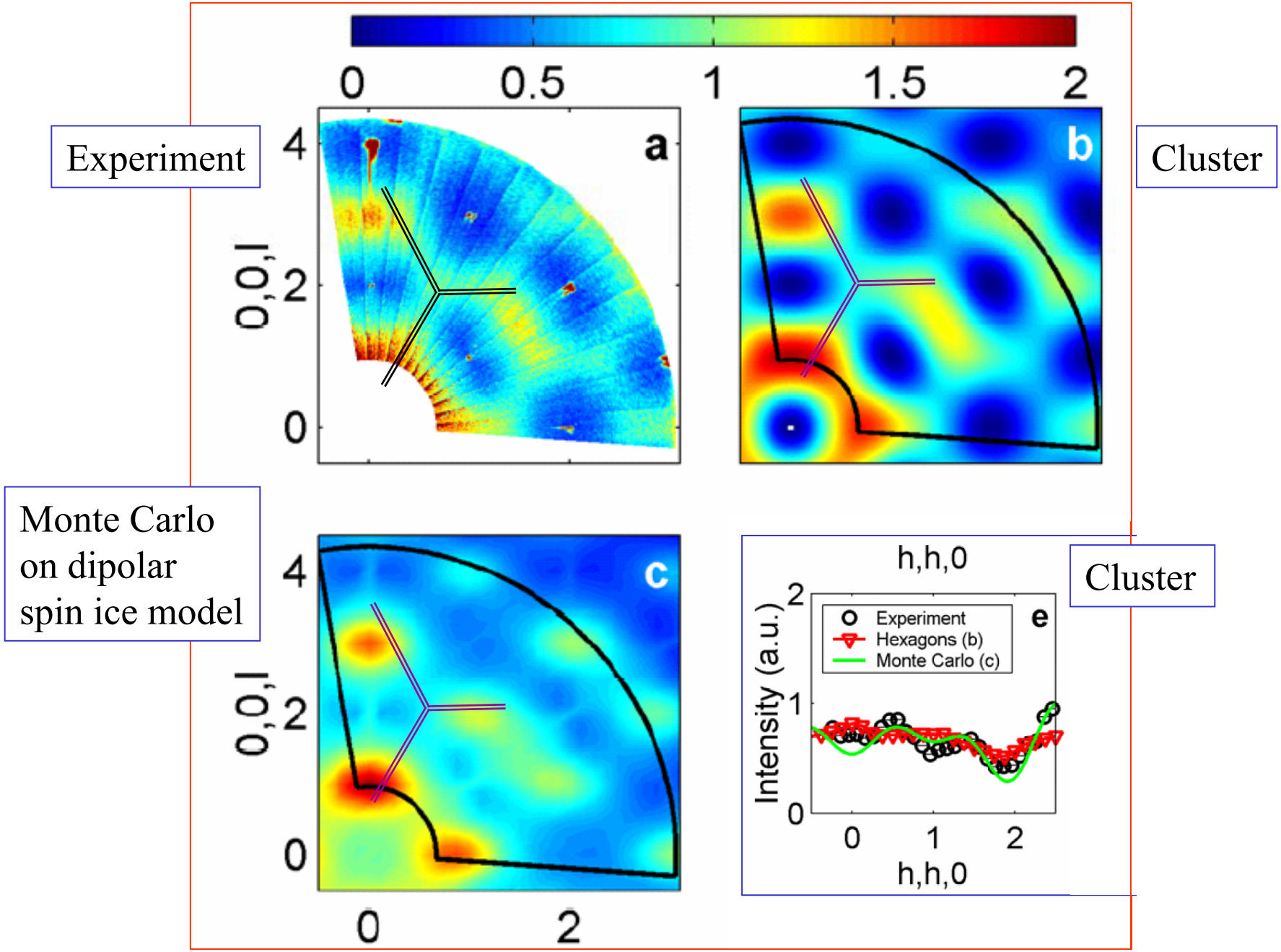
$ZnCr_2O_4$ spinel

$\theta_{CW} \sim -390$ K
$T_c = 12.5$ K

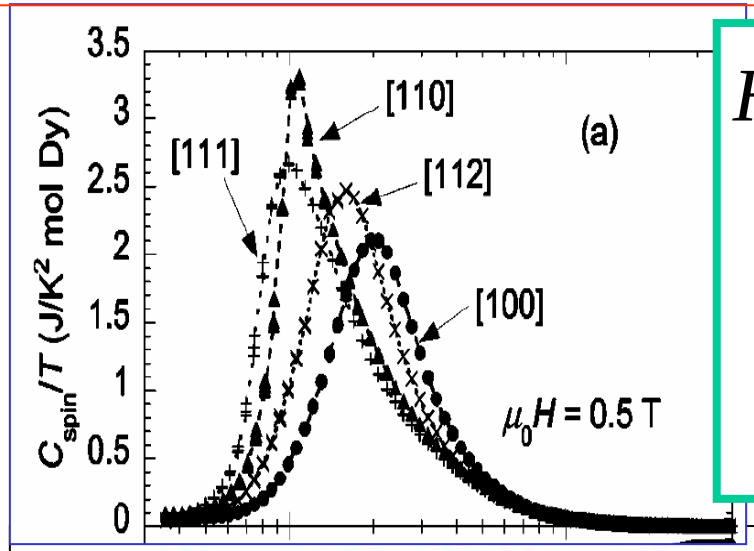


$$|F_6(\mathbf{Q})|^2 \propto \left\{\sin\frac{\pi}{2}h\cdot\left(\cos\frac{\pi}{2}k-\cos\frac{\pi}{2}l\right)\right\}^2 + \left\{\sin\frac{\pi}{2}k\cdot\left(\cos\frac{\pi}{2}l-\cos\frac{\pi}{2}h\right)\right\}^2 + \left\{\sin\frac{\pi}{2}l\cdot\left(\cos\frac{\pi}{2}h-\cos\frac{\pi}{2}k\right)\right\}^2$$

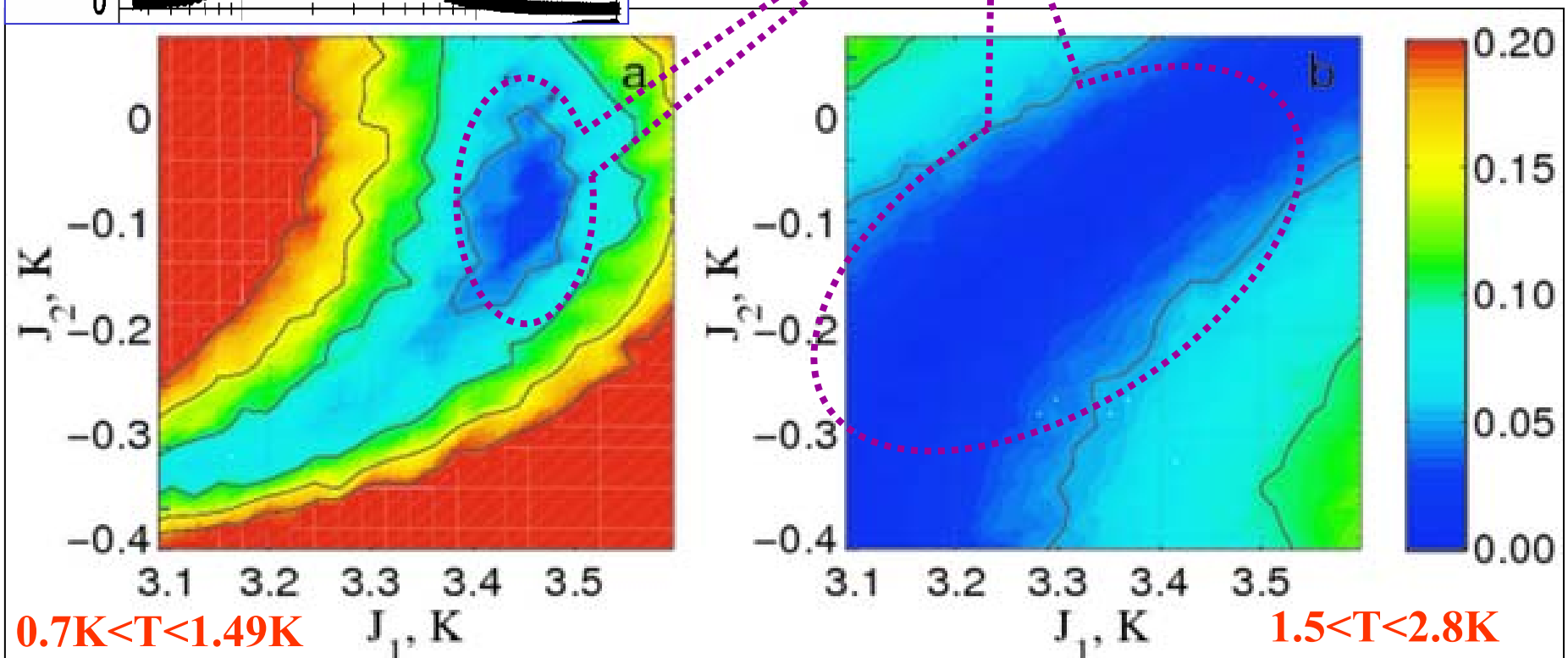


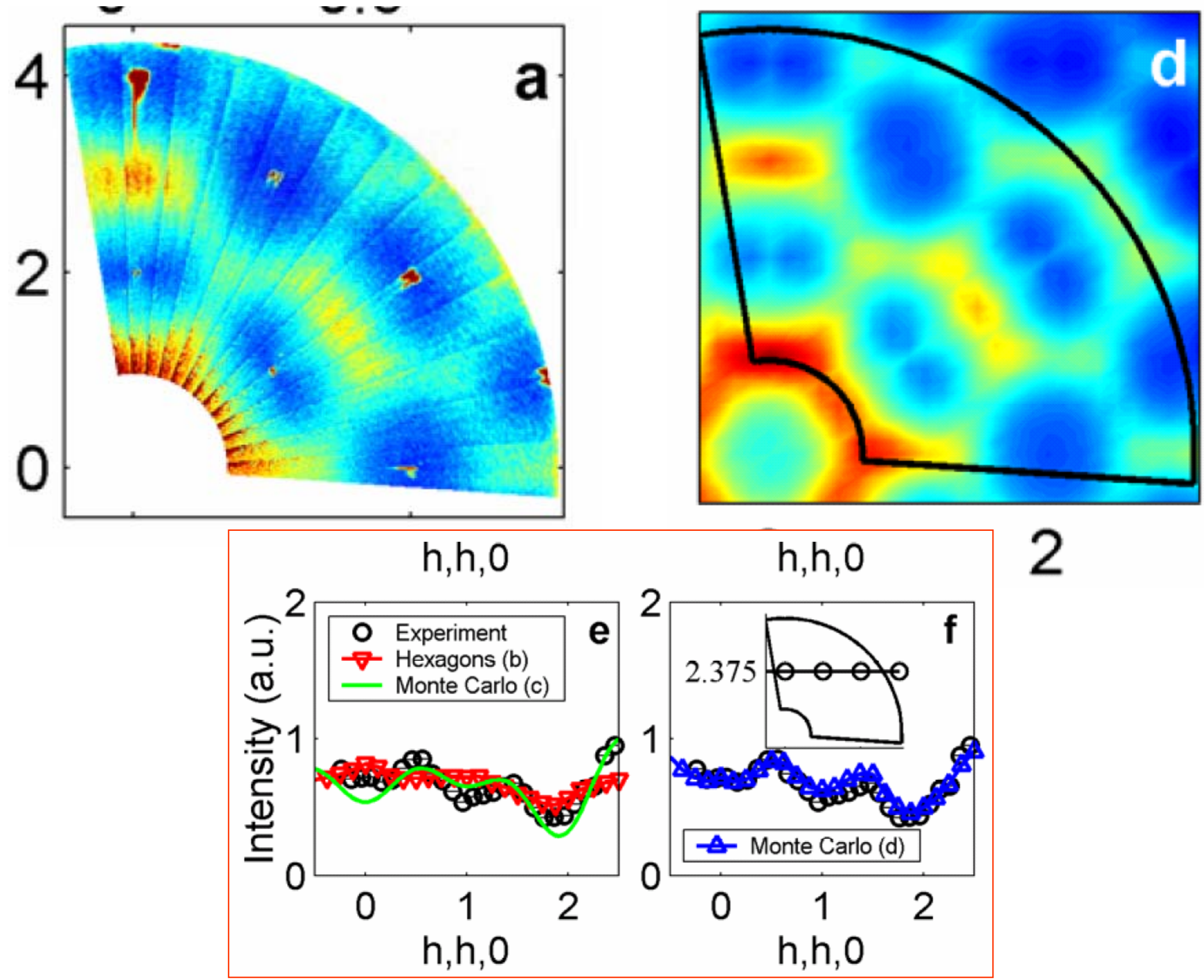

Experiment
Cluster
Monte Carlo on dipolar spin ice model
Cluster
0
0.5
1
1.5
2
a
b
c
e
0,0,l
h,h,0
Intensity (a.u.)
Experiment
Hexagons (b)
Monte Carlo (c)

Fit to many experiments: $M(\mathbf{H})$, $C_v(T,H)$



$$H = -\sum_{\langle i,j \rangle} J_{ij} \left(\hat{z}_i \bullet \hat{z}_j \right) \sigma_i^{z_i} \sigma_j^{z_j} + D \sum_{i>j} \frac{\hat{z}_i \bullet \hat{z}_j}{\left| \vec{r}_{ij} \right|^3} - \frac{3(\hat{z}_i \bullet \vec{r}_{ij})(\hat{z}_j \bullet \vec{r}_{ij})}{\left| \vec{r}_{ij} \right|^5} \sigma_i^{z_i} \sigma_j^{z_j}$$




a
d
4
2
0
2
h,h,0
h,h,0
e
f
Experiment
Hexagons (b)
Monte Carlo (c)
Monte Carlo (d)
2.375
Intensity (a.u.)
2
1
0
0
1
2
h,h,0
h,h,0

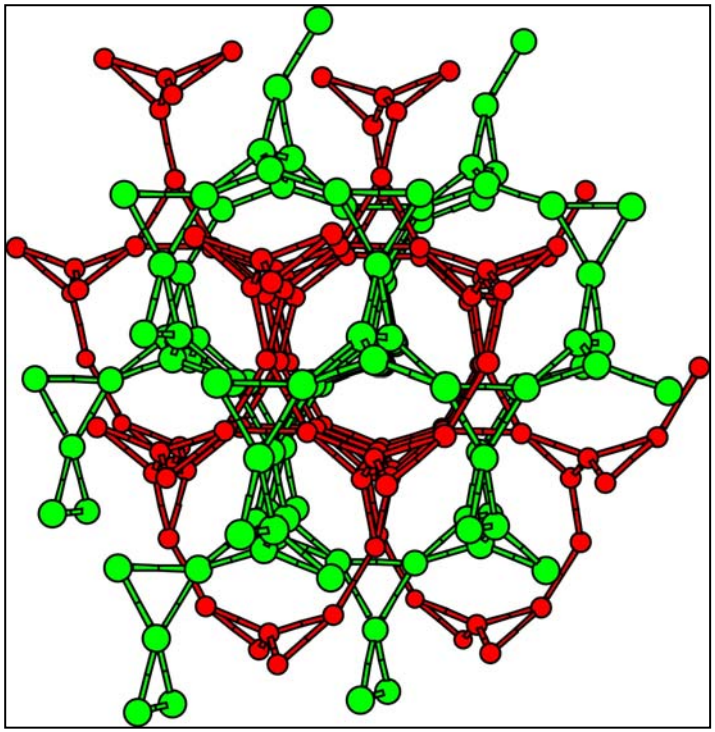
Conclusion about $Dy_2Ti_2O_7$ Spin Ice

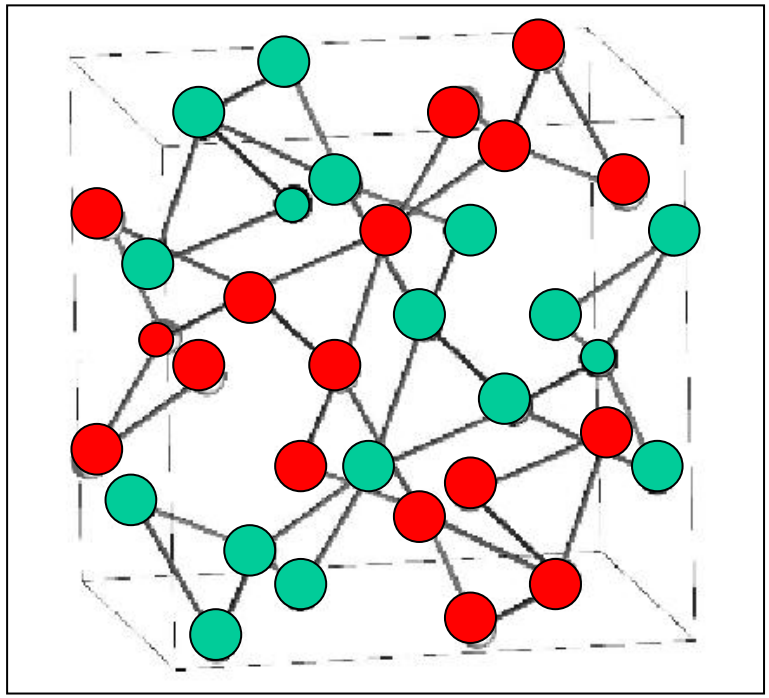
- At least in this system, it appears that "cluster-like" / "composite-spin" – like correlations are not emergent.
- They are <u>accidentally</u> <u>fined-tuned</u> by Nature.
- They are the signature of the development of "superstructure" in S(**q**) arising from ancillary perturbations to H_0 in the "spin liquid" state.

Yavorsk'ii, Fennell, Gingras and Bramwell; arXiv:0707.3477

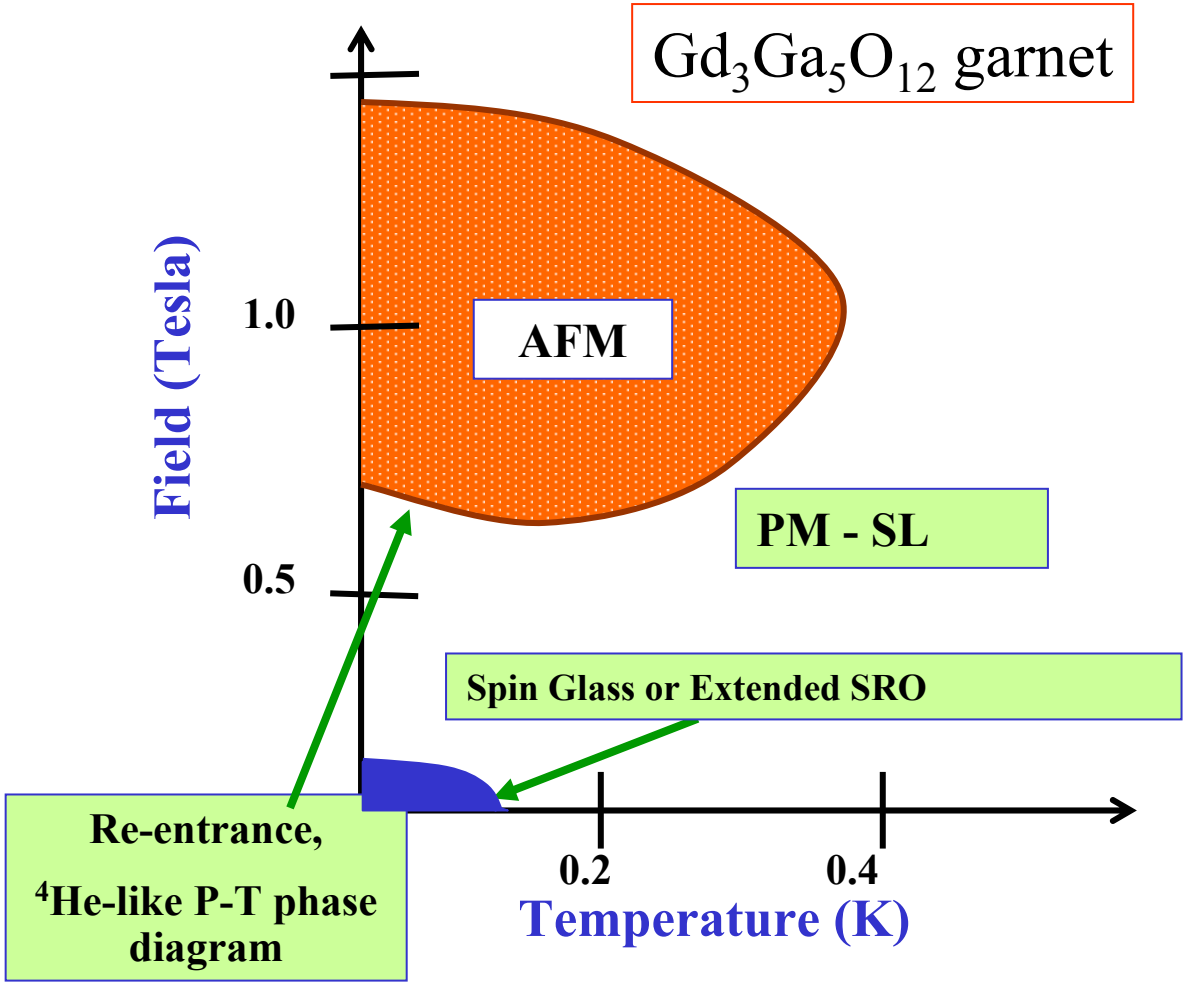
$Gd_3Ga_5O_{12}$ Garnet (GGG)

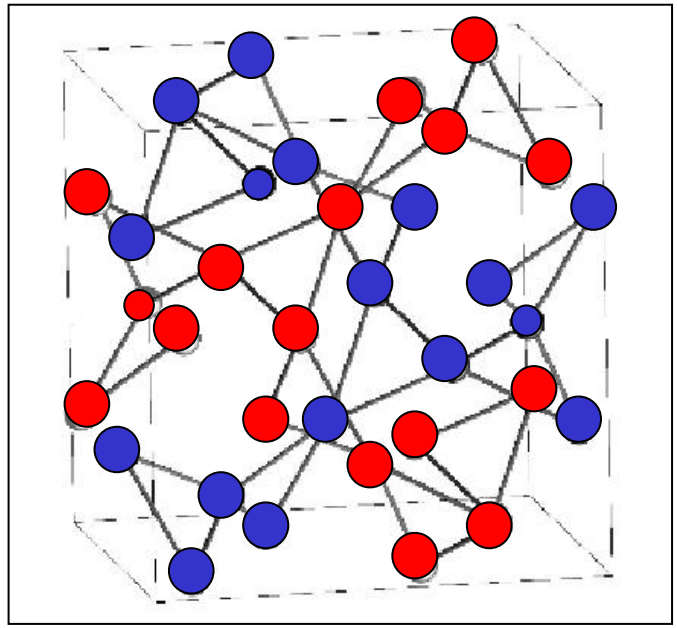
•Complex lattice with 24 atoms in conventional cubic unit cell





Yavors'kii, Enjalran and Gingras; Phys. Rev. Lett. **97,** 267203 [2006]





- H~0T, T< 180mK, spin glass phase or extended SR correlations, ξ=100A?
- No LRO at H=0.
- Spin Liquid phase at intermediate fields.
- AFM phase at stronger fields.
- Reentrance resembles ^{4}He melting.

VOLUME 85, NUMBER 16 PHYSICAL REVIEW LETTERS 16 OCTOBER 2000

Low Temperature Spin Dynamics of the Geometrically Frustrated Antiferromagnetic Garnet $Gd_3Ga_5O_{12}$

S. R. Dunsiger,[1] J. S. Gardner,[2,*] J. A. Chakhalian,[1] A. L. Cornelius,[3] M. Jaime,[4] R. F. Kiefl,[1,5] R. Movshovich,[4] W. A. MacFarlane,[1,†] R. I. Miller,[1] J. E. Sonier,[1,‡] and B. D. Gaulin[2]

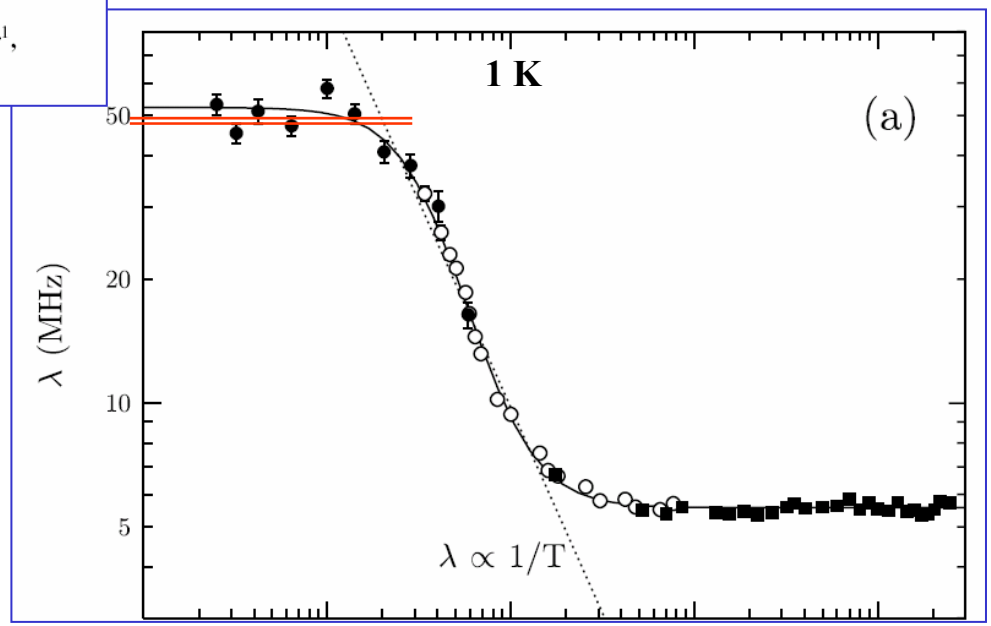
INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **14** (2002) L157–L163 PII: S0953-8984(02)28459-3

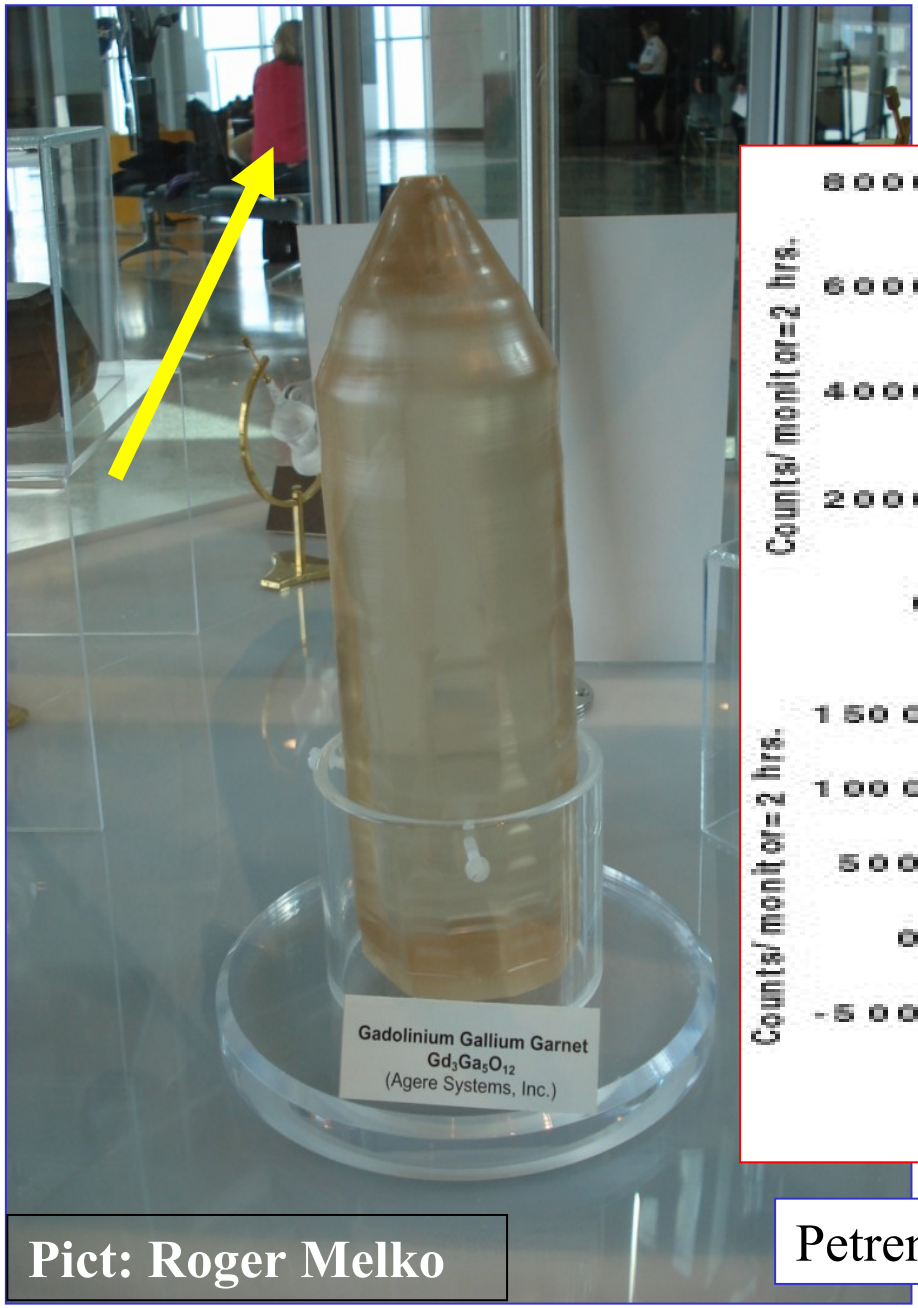
LETTER TO THE EDITOR

A muon-spin relaxation (μSR) study of the geometrically frustrated magnets $Gd_3Ga_5O_{12}$ and $ZnCr_2O_4$

I M Marshall[1], S J Blundell[1,3], F L Pratt[2], A Husmann[1], C A Steer[1], A I Coldea[1], W Hayes[1] and R C C Ward[1]

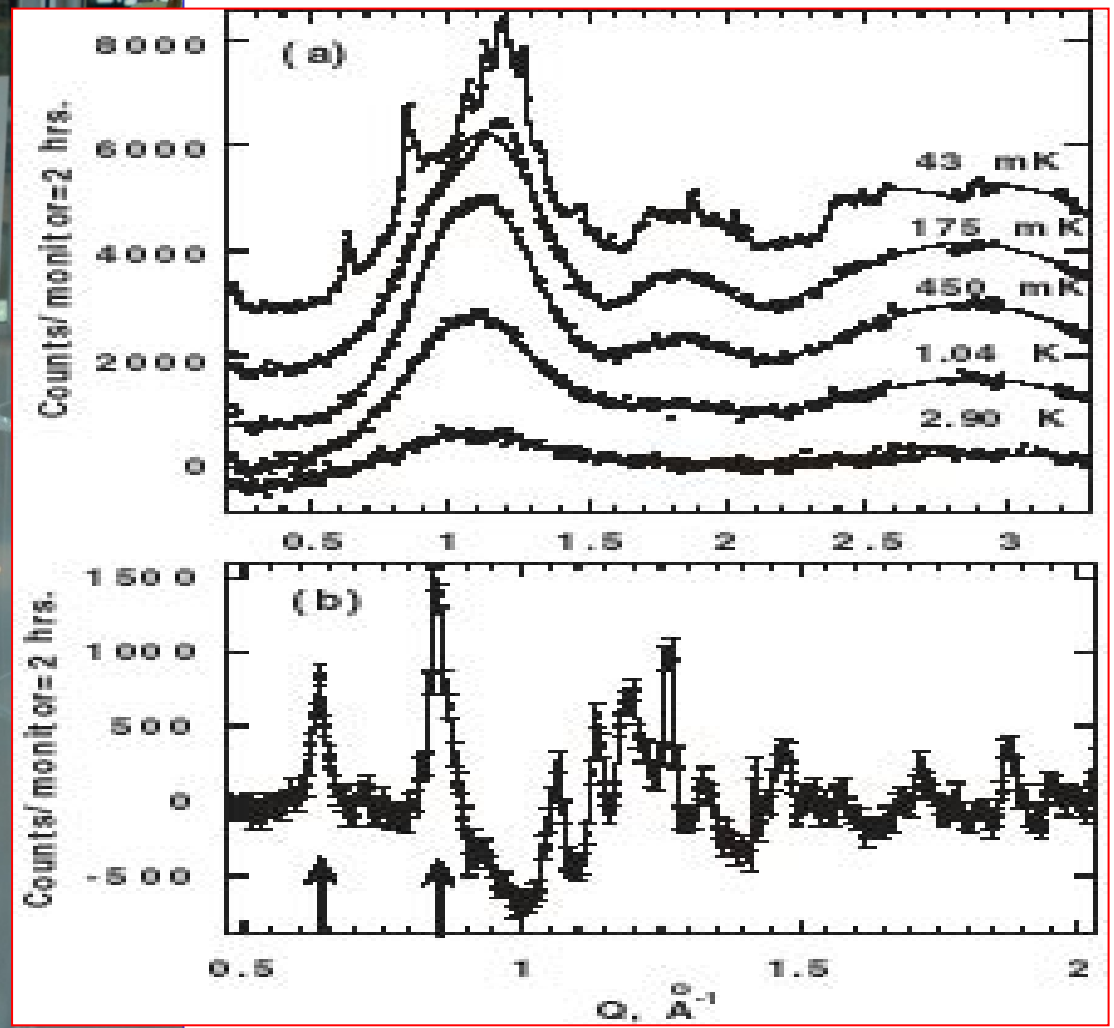


Zero field experimental picture of: neutrons



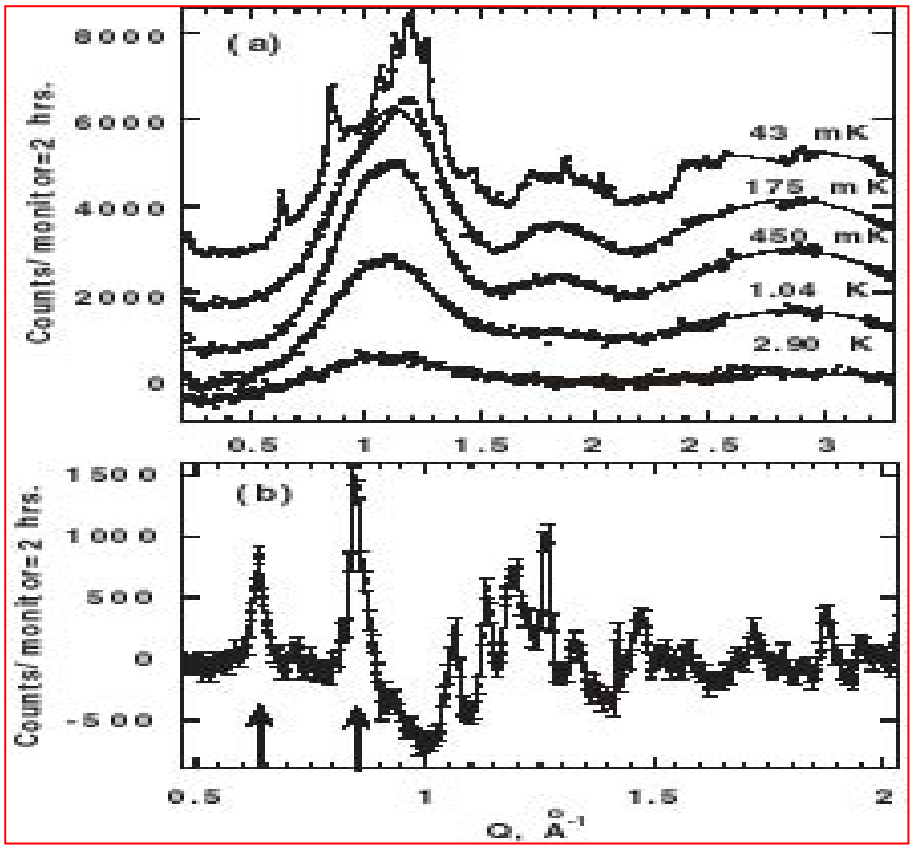
Pict: Roger Melko

Isotopically enriched ^{160}Gd



Petrenko *et* al., Phys. Rev. Lett. **80**, 4570 (1998)

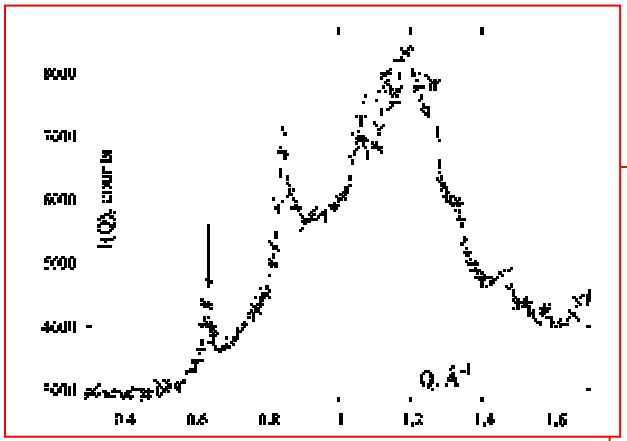
Zero field experimental picture of : neutrons

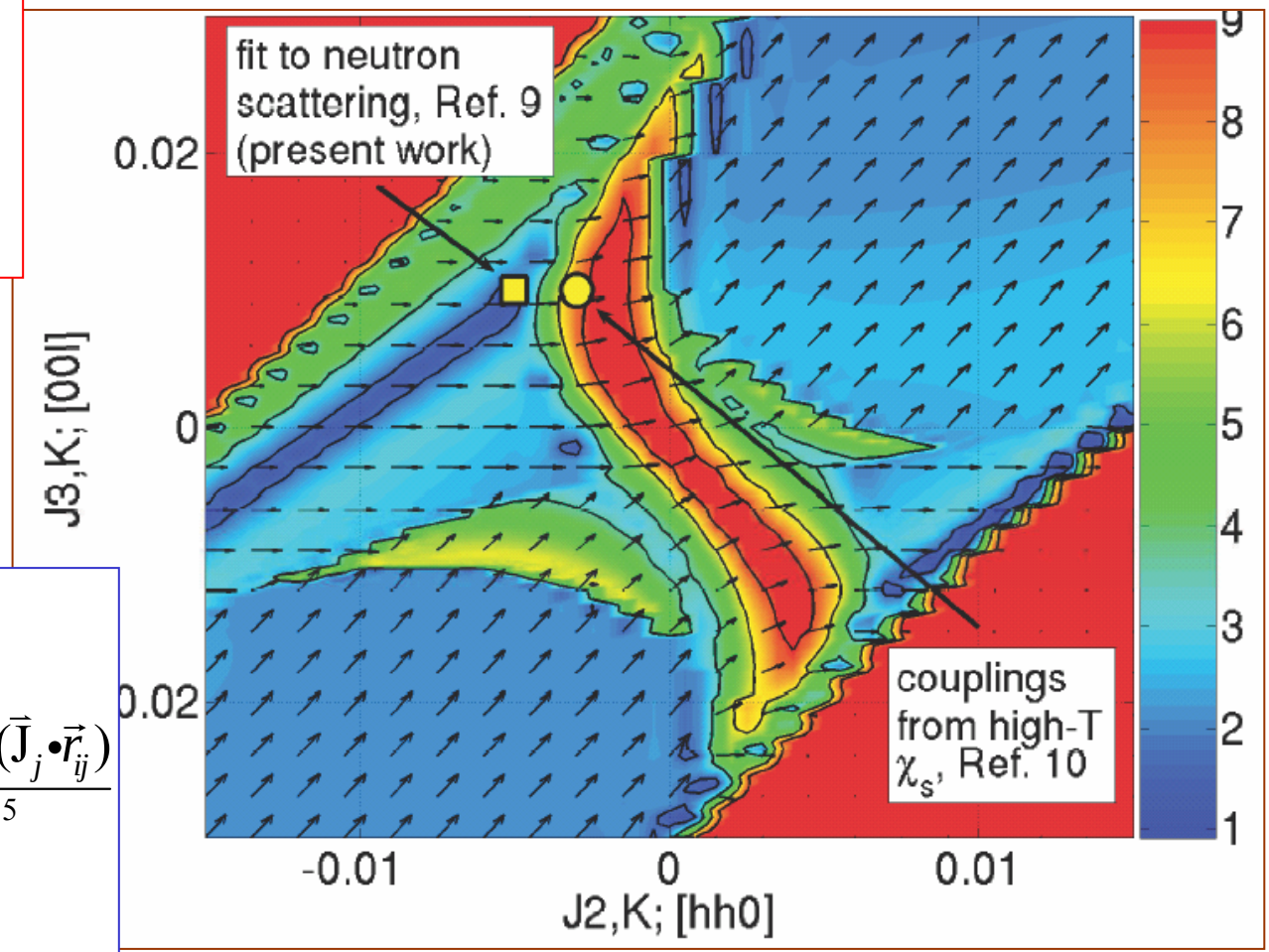


1. How can the existence of sharp peaks in neutron scattering be understood in the context of the bulk measurements finding glassy behavior and muon spin relaxation finding persistent spin dynamics down to 20 mK?

2. There has been some skepticism in the field as to the correctness/accuracy/intrinsicness of those neutron data.

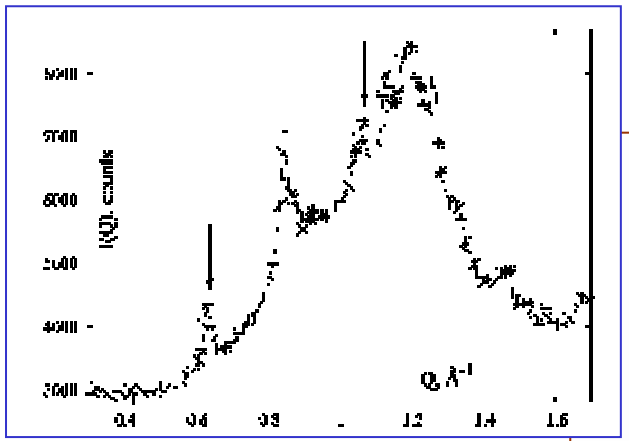
1) Determine the ordering wavevector using Gaussian approximation (i.e.) mean-field theory

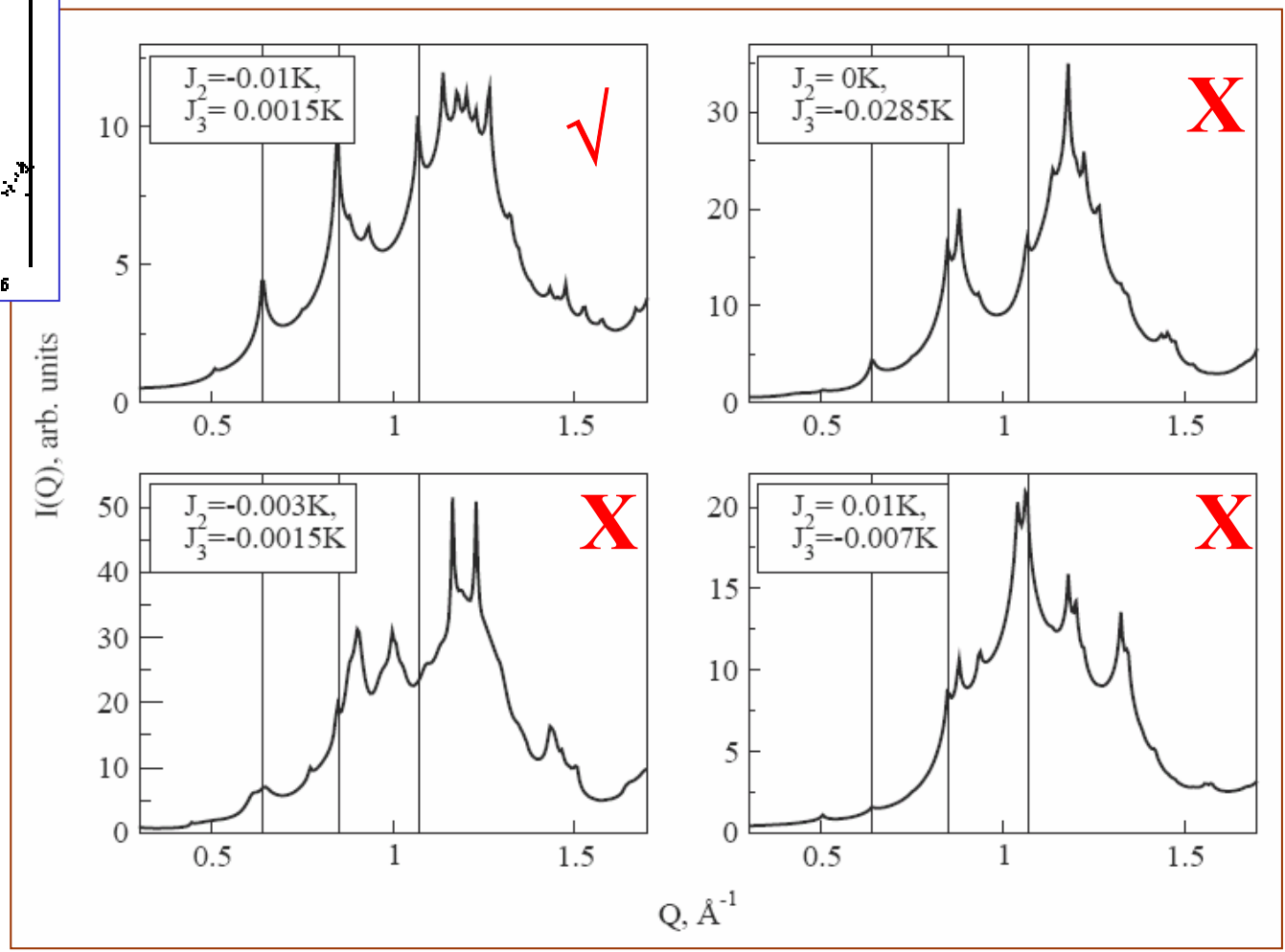




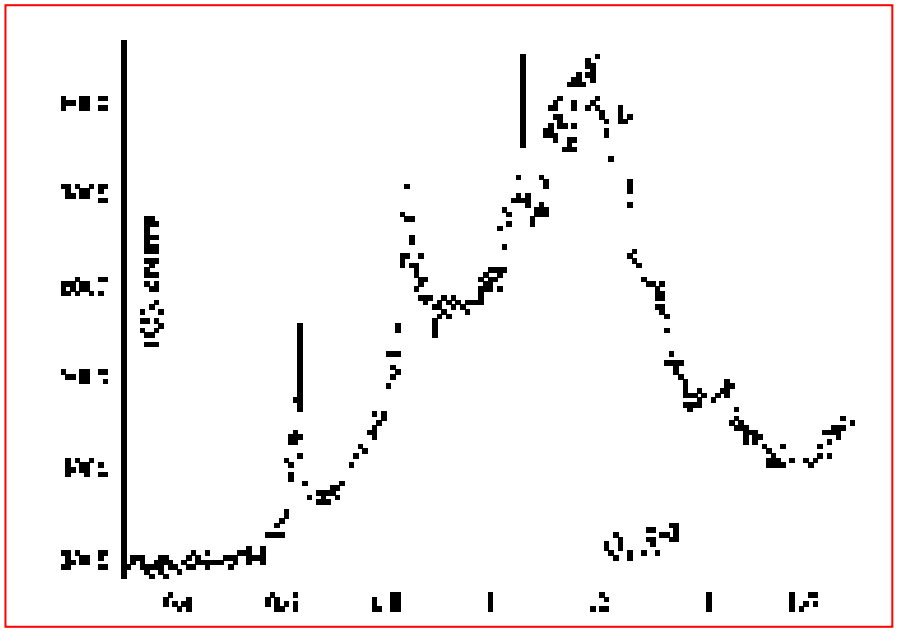
$$H = -\sum_{j>i} J_{ij}\ \vec{J}_i \bullet \vec{J}_j + \frac{\mu_0}{4\pi}(g\mu_B)^2 \sum_{j>i} \frac{\vec{J}_i \bullet \vec{J}_j}{\left|\vec{r}_{ij}\right|^3} - \frac{3(\vec{J}_i \bullet \vec{r}_{ij})(\vec{J}_j \bullet \vec{r}_{ij})}{\left|\vec{r}_{ij}\right|^5}$$

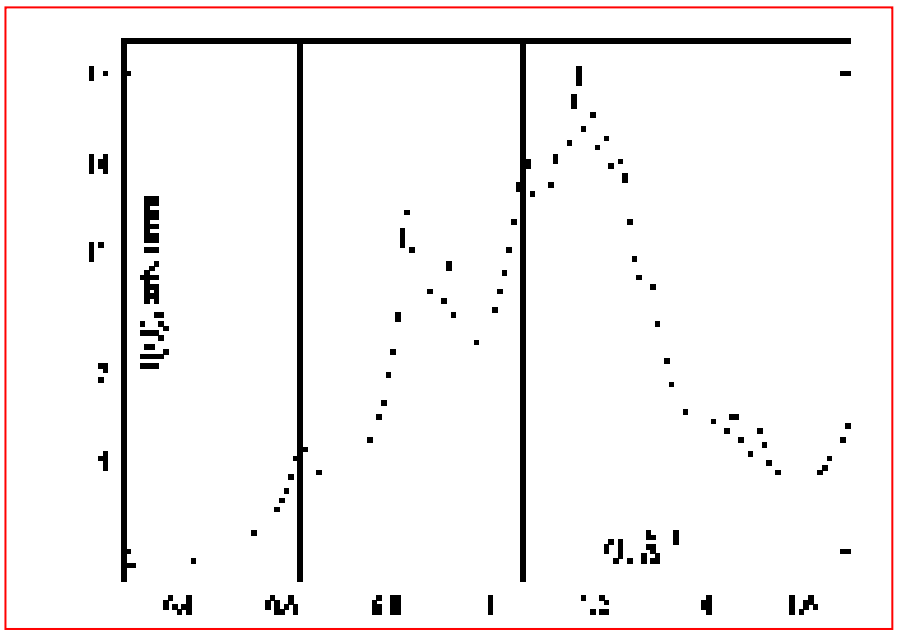
2) Determine the right J_2 and J_3 by comparing the experimental vs theoretical critical neutron scattering calculated using MFT [similar conclusion reached by considering paramagnetic $S(\mathbf{q})$]





Theoretical Calculations





By allowing a fine-tuning of the exchange interactions beyond nearest neighbors, we are able to suggest that the incommensurate ordering seen in $Gd_3Ga_5O_{12}$ garnet (GGG) may be "intrinsic" to the would-have been "idealized/disorder-free system".

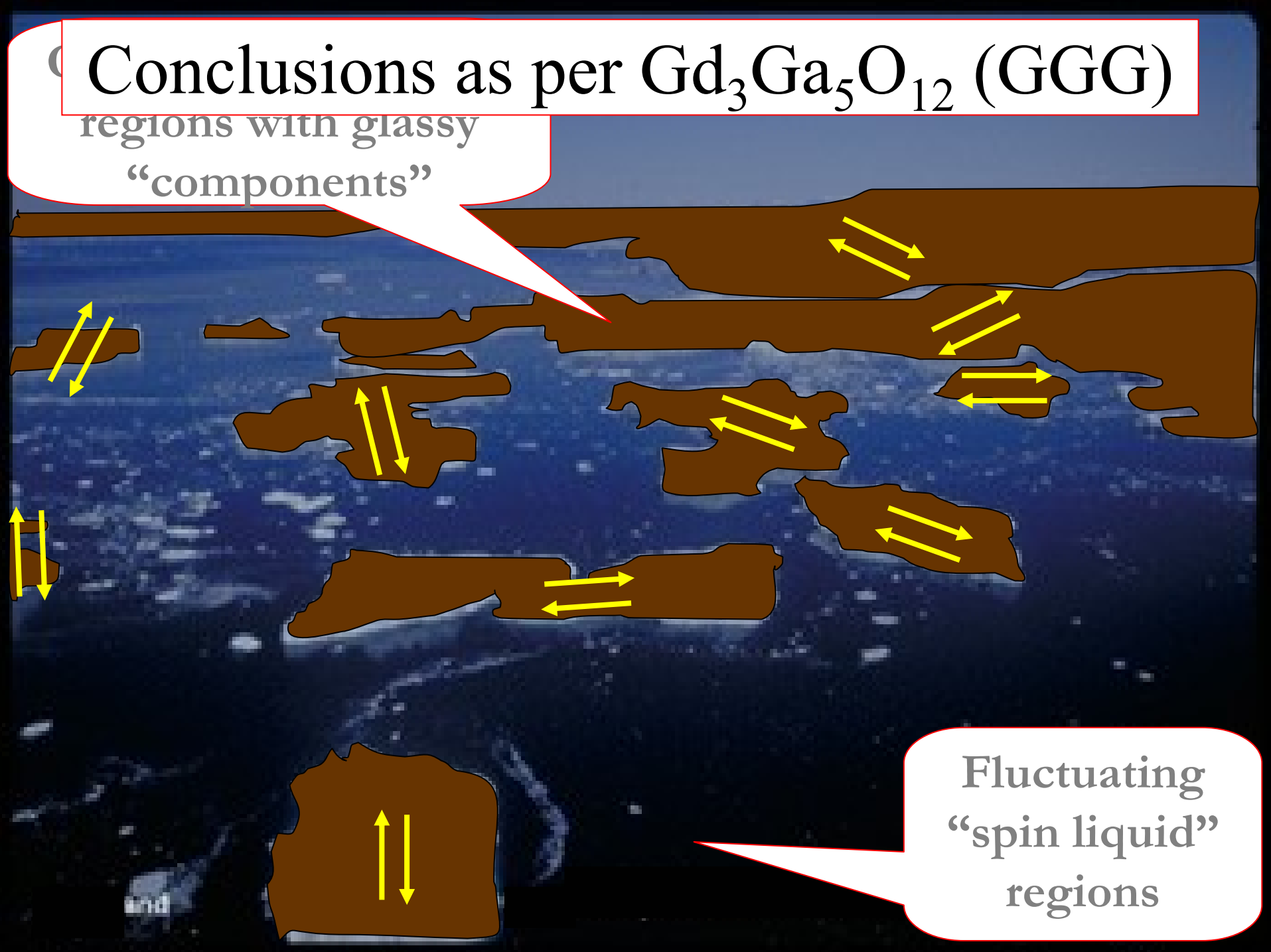
$$\vec{q}_{\text{ord}} \cong \frac{2\pi}{a}(0.29, 0.29, 0)$$

Similar results reached with MC
T_c^{MC} ~0.2K / T_c^{exp} ~0.14K

What does that mean?
What kind of state is GGG in at low temperatures?

T. Yavors'kii, M. Enjalran, and M. J. P. Gingras
Physical Review Letters **97**, 267203 (2006)

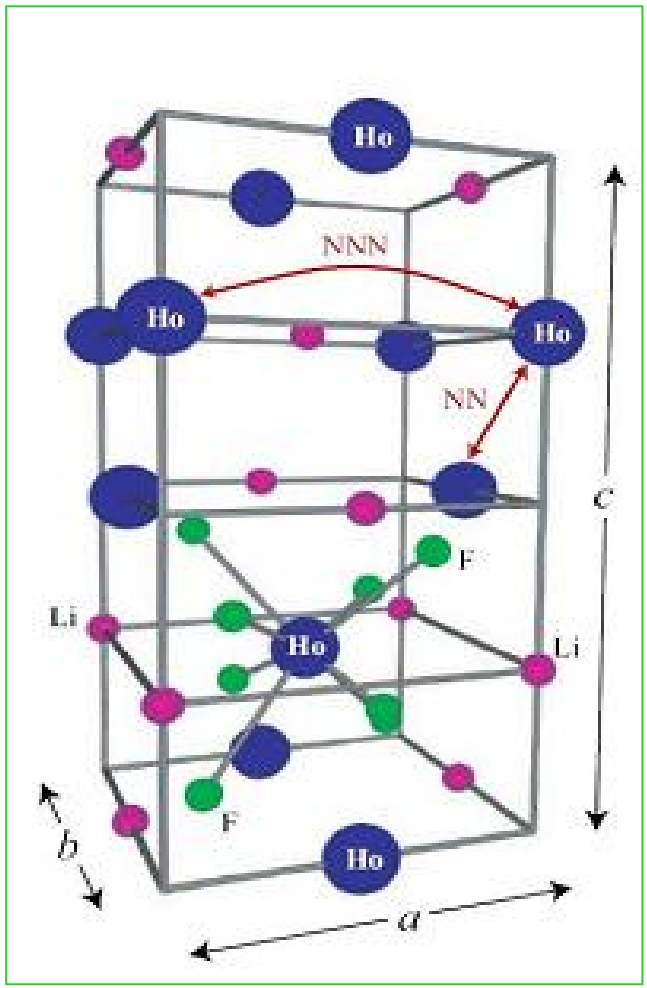
Conclusions as per $Gd_3Ga_5O_{12}$ (GGG)



Conclusion about $Gd_3Ga_5O_{12}$

- $Gd_3Ga_5O_{12}$ is displaying incommensurate order – system is on the verge …
- Bulk is akin to a *spin slush*…

Transverse Field $LiHo_xY_{1-x}F_4$



$\mathbf{B_x}$

The Transverse-Field Ising Model

Pierre-Gilles de Gennes
Paris, France 1932

PGdG introduced the TFIM to describe collective low energy proton excitations & *tunneling* effects in ferroelectrics:

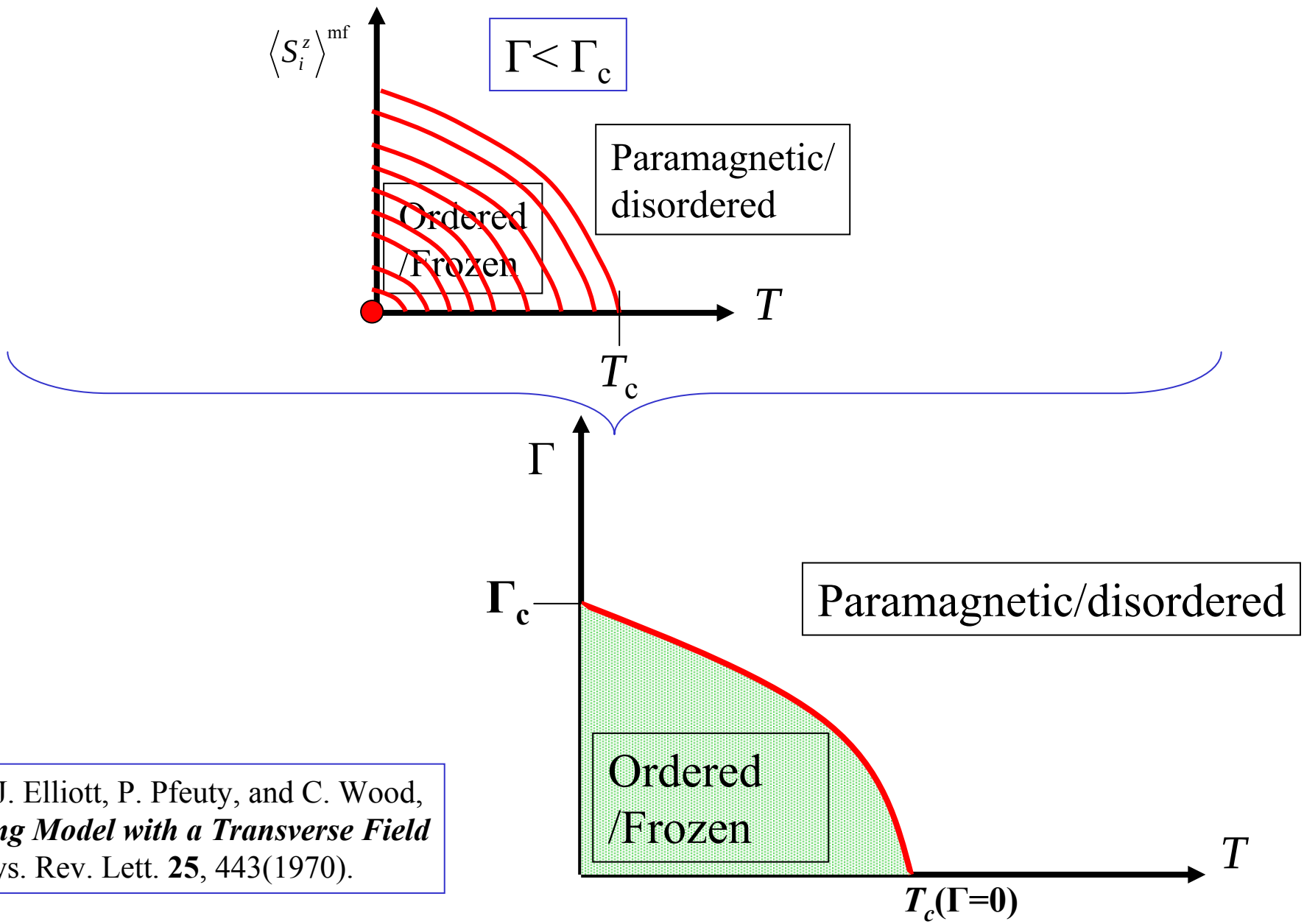
$$H = -\sum_{\langle i,j \rangle} J_{ij} S_i^z S_j^z - \Gamma \sum_{\mathrm{i}} S_i^x$$

Quantum mechanics (tunneling between the "up" and "down" direction) is introduced by the *transverse field* term (proportional to Γ)

$$\left[S_i^x , S_i^z \right] \neq 0$$

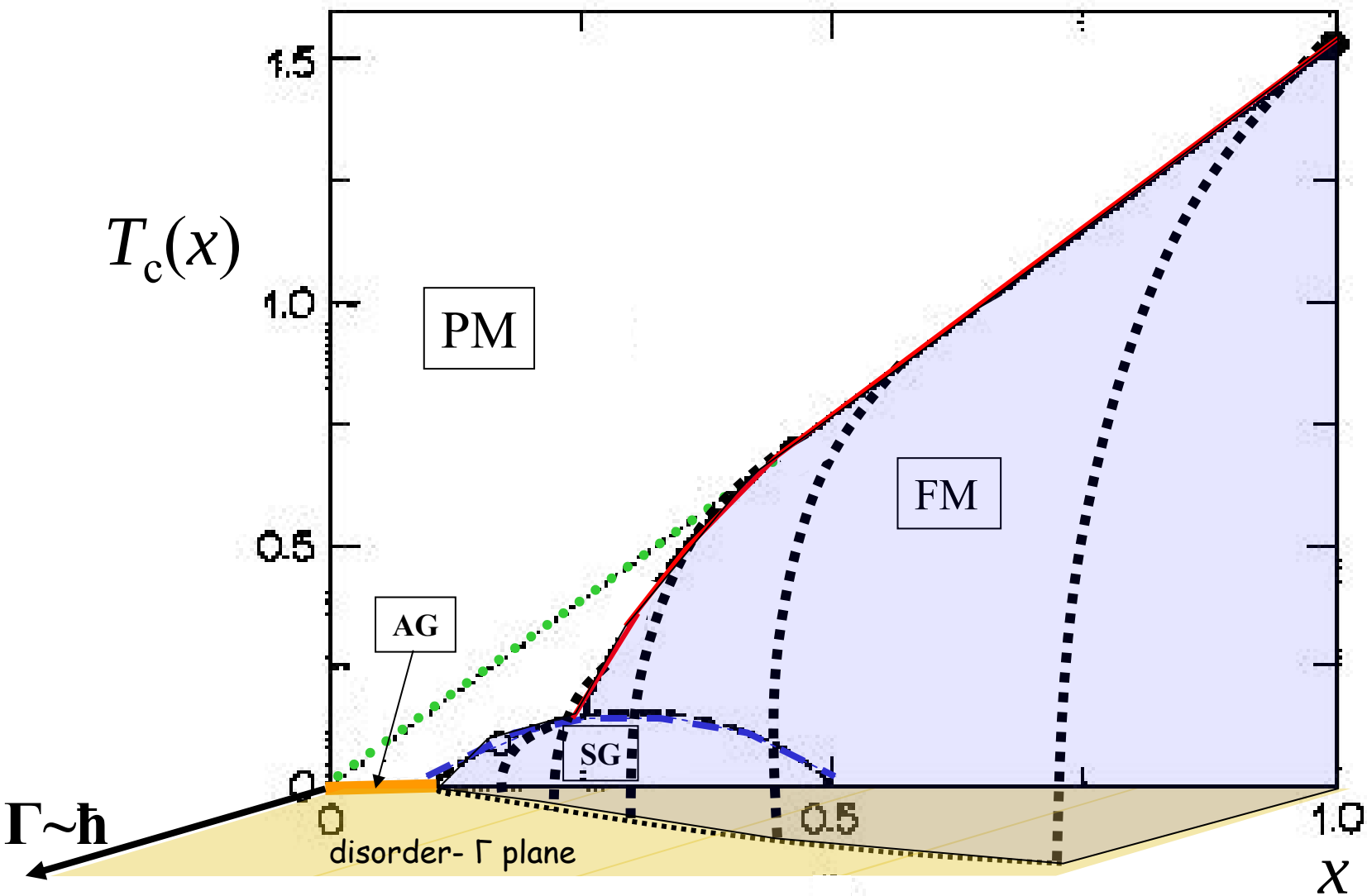
P.G. de Gennes,
Collective Motions of Hydrogen Bonds
Solid. State. Comm. **1,** 132 (1963).

Essentials of Γ-*T* Phase Diagram



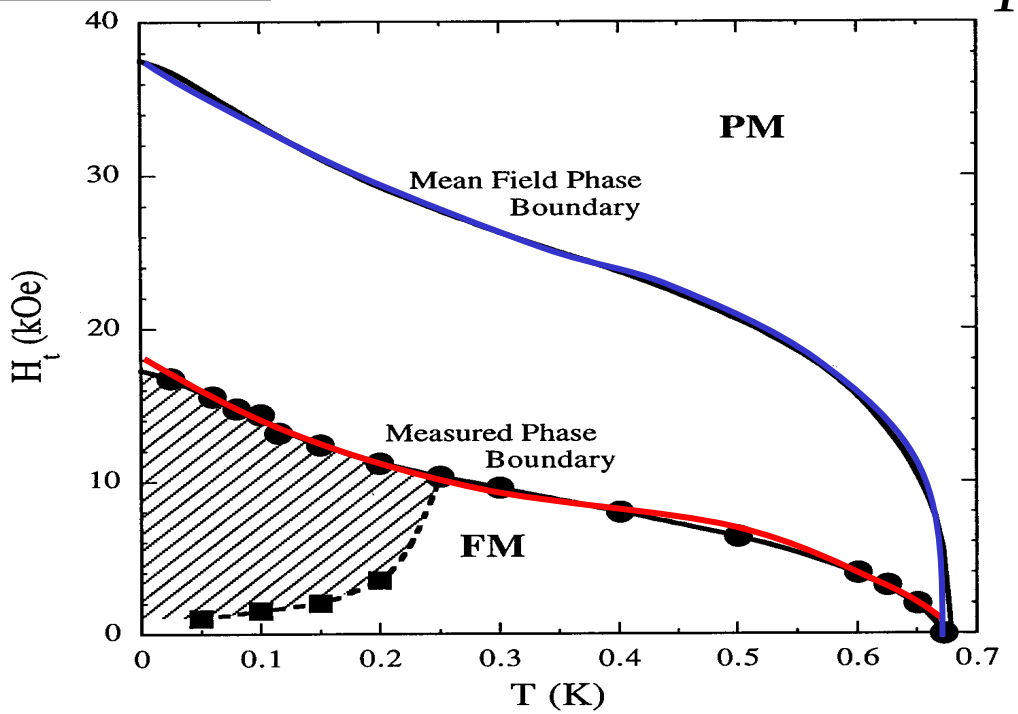
R. J. Elliott, P. Pfeuty, and C. Wood,
Ising Model with a Transverse Field
Phys. Rev. Lett. **25**, 443(1970).

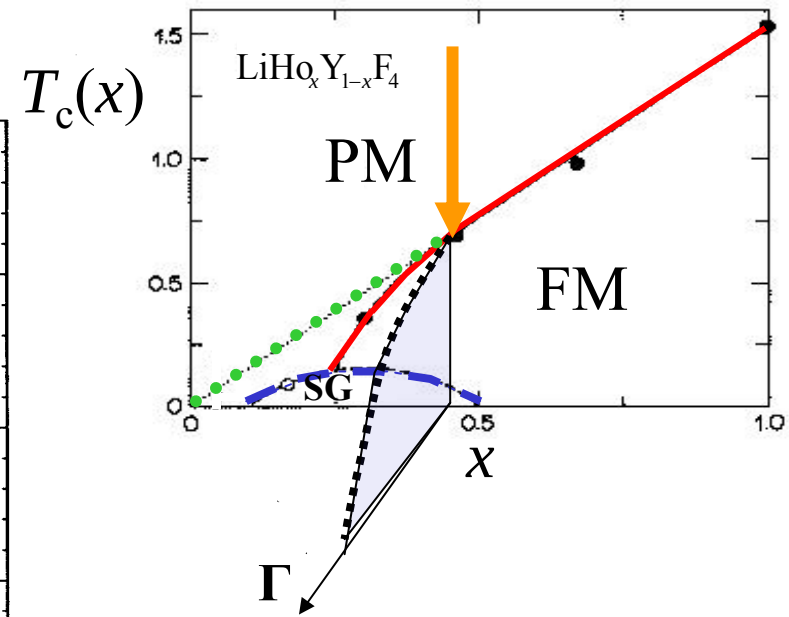
x-T Phase Diagram of $LiHo_xY_{1-x}F_4$



Transverse field (Γ) vs temperature (T) phase diagram of $LiHo_xY_{1-x}F_4$; x=0.44

"ferroglass" composition





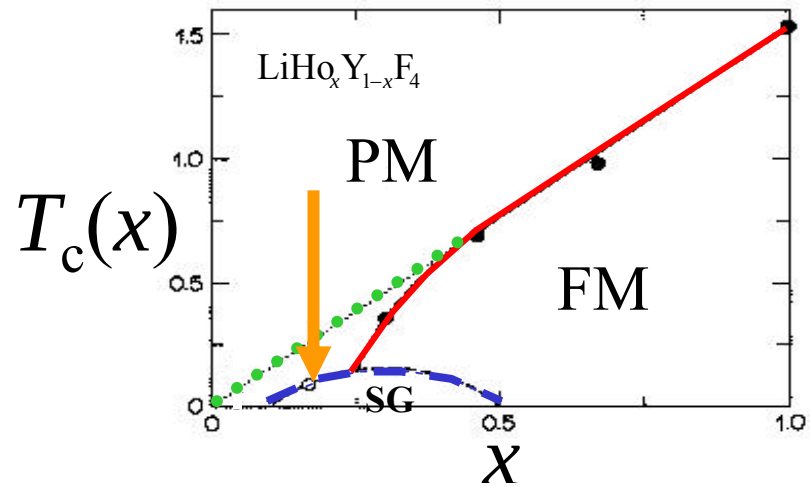
J. Brooke, U. Chicago; Ph.D Thesis.

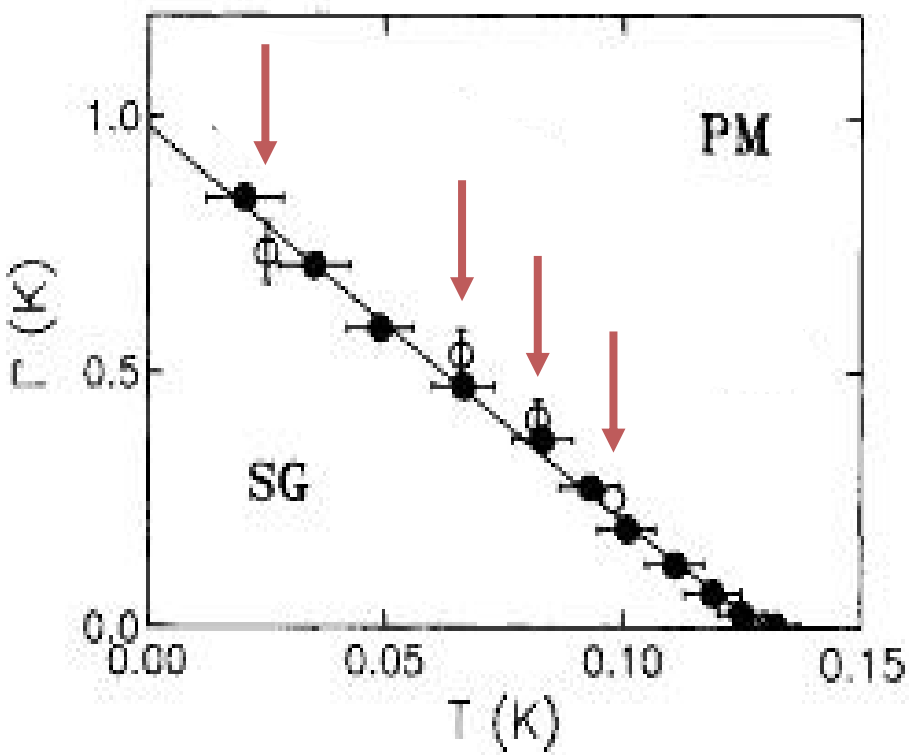
Figure 2.3: The phase diagram of $LiHo_xY_{1-x}F_4$ as measured by the peak in $\chi'(H_t)$ when ramped from $H_t = 24$ kOe to $H_t = 0$ kOe. Solid circles are data, and the line through the data is simply a guide to the eye. Upper curve is a single-ion mean-field calculation. The excellent agreement at the classical $T_c = 0.670$ K does not hold in the presence of a transverse field.

Transverse field (Γ) vs temperature (T) phase diagram of $LiHo_xY_{1-x}F_4$; x=0.167

Spin glass composition

$$M(T,H) = \chi_1(T)H - \chi_3(T)H^3 + \ldots$$





Microscopic origin of transverse field Ising model in $LiHo_xY_{1-x}F_4$

$$H = \tilde{V}_{CEF}(\{J_i^\alpha\}) + \sum_{\langle ij \rangle} J_{ex}^z S_i^z S_j^z + \sum_{\langle i,j \rangle} \vec{J}_i \bullet \vec{J}_j$$

$$+ \tilde{D} \sum_{i \neq j} \left(\frac{\vec{J}_i \bullet \vec{J}_j}{|\vec{r}_{ij}|^3} - \frac{3(\vec{J}_i \bullet \vec{r}_{ij})(\vec{J}_j \bullet \vec{r}_{ij})}{|\vec{r}_{ij}|^5} \right) S_i^z S_j^z$$

$$- g\mu_B \sum_i \sum_i S_i J_i^x B_x$$

!!!

Remember: moved from J=8 → S=1/2 effective spin

Leading Interactions in $LiHo_xY_{1-x}F_4$: long-range magnetic dipole-dipole <u>REVISITED:</u>

$$H = DR_{nn}^3 \sum_{i>j} \varepsilon_i \varepsilon_j \left(\vec{J}_i \bullet \vec{J}_j - 3\vec{J}_i \bullet \hat{r}_{ij} \hat{r}_{ij} \bullet \vec{J}_j \right) r_{ij}^{-3}$$

- In presence of the randomness, there will be random terms of the form:

$$\varepsilon_i \varepsilon_j r_{ij}^x r_{ij}^z J_i^x J_i^z$$

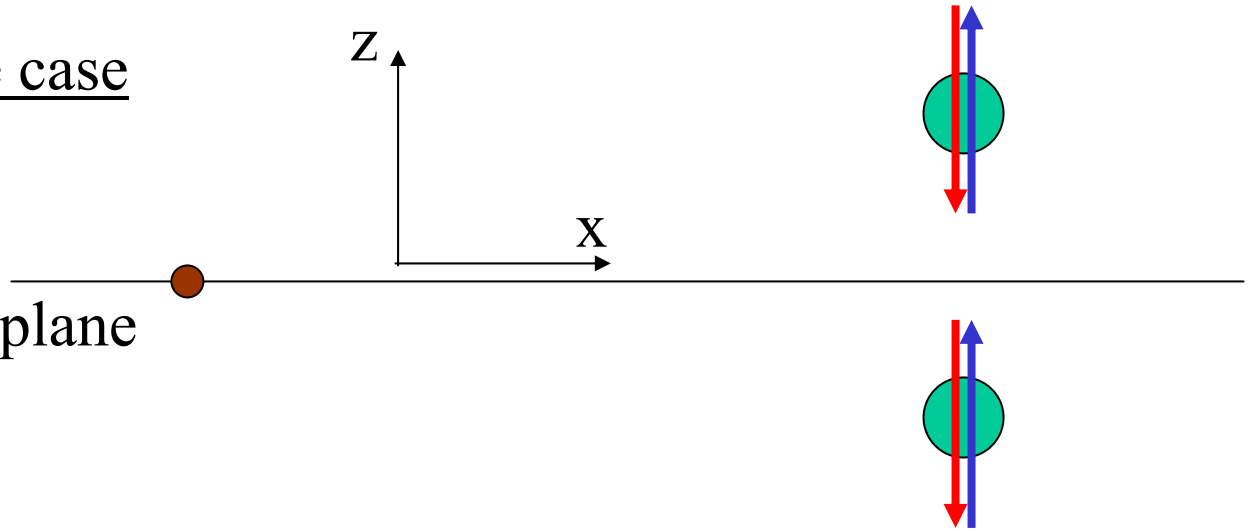
- Since there is broken symmetry along the x direction by the applied transverse field, the x-component of J_i will be nonzero:

$$\sum_j \varepsilon_j r_{ij}^x r_{ij}^z \left\langle J_j^x \right\rangle J_i^z \approx h_i^z J_i^z$$

- This means that there is a ***random*** field along the z direction introduced by the combination of (***i***) the applied transverse field along the x direction and (***ii***) the random dilution of $Ho^{3+} \rightarrow Y^{3+}$.

Generation of Longitudinal Random Fields

a) Pure case

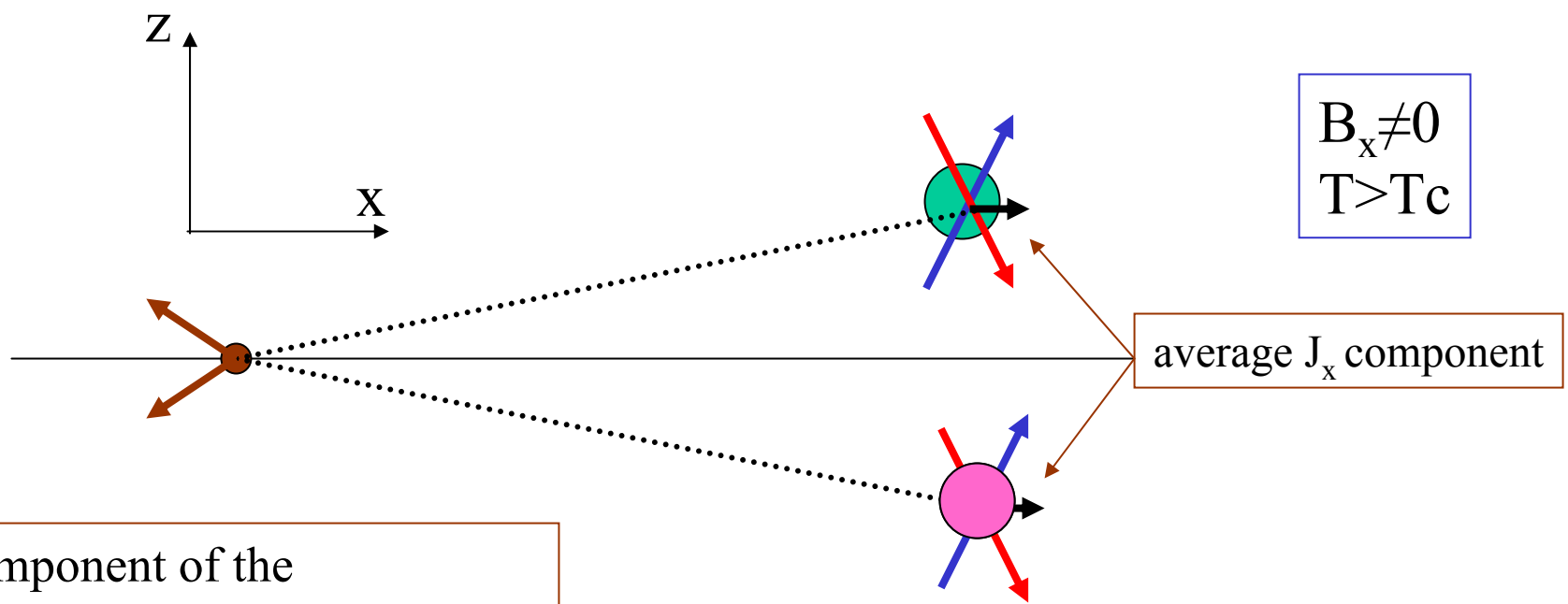


mirror plane

$B_x=0$
$T>T_c$

Generation of Longitudinal Random Fields

b) Diluted case



A component of the
local (magnetic) field is induced
along *z* at reference point

Hence, the minimal model that we have is:

- **<u>Not only</u>** random Ising systems only in a transverse field:

$$H = -\sum_{\langle i,j \rangle} J_{ij} \sigma_i^z \sigma_j^z - \Gamma \sum_{\mathrm{i}} \sigma_i^x$$

Effective Hamiltonian Method

$$H = D\sum_{j>i}\left(\varepsilon_i\varepsilon_j\right)\left(\frac{\vec{\mathrm{J}}_i \bullet \vec{\mathrm{J}}_j}{R_{ij}^{\ 3}} - 3\frac{(\vec{\mathrm{J}}_i \bullet \vec{R}_{ij})(\vec{\mathrm{J}}_j \bullet \vec{R}_{ij})}{R_{ij}^{\ 5}}\right) + V_c$$

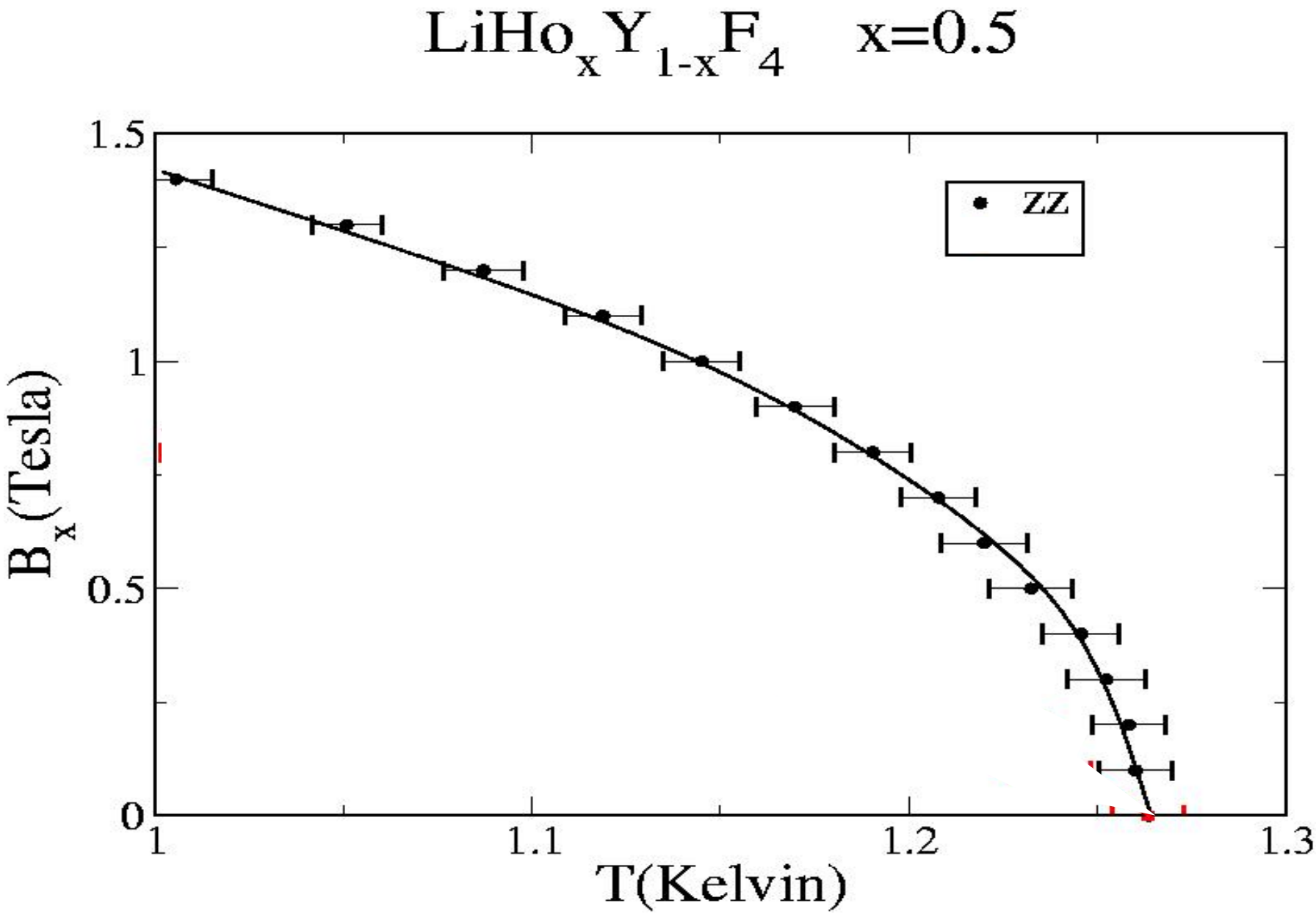
$$\mathrm{P} = \sum_{\alpha\in\mathrm{P}} |\alpha\rangle\langle\alpha|$$

$$R = \sum_{\beta\notin P}\frac{|\beta\rangle\langle\beta|}{E_0^{\alpha} - E_0^{\beta}}$$

$$H_{\mathrm{eff}} = PHP + PHRHP + \cdots$$

$$H_{\mathrm{eff}} = -\frac{1}{2}\sum_{i,j} J_{ij} S_i^z S_j^z - \frac{1}{2}\sum_{i,j} K_{ij} S_i^x S_j^z$$
$$-\Gamma\sum_i S_i^x - h_{0z}\sum_i S_i^z - \sum_i h_{ri} S_i^z$$

Mean-Field Phase Diagram

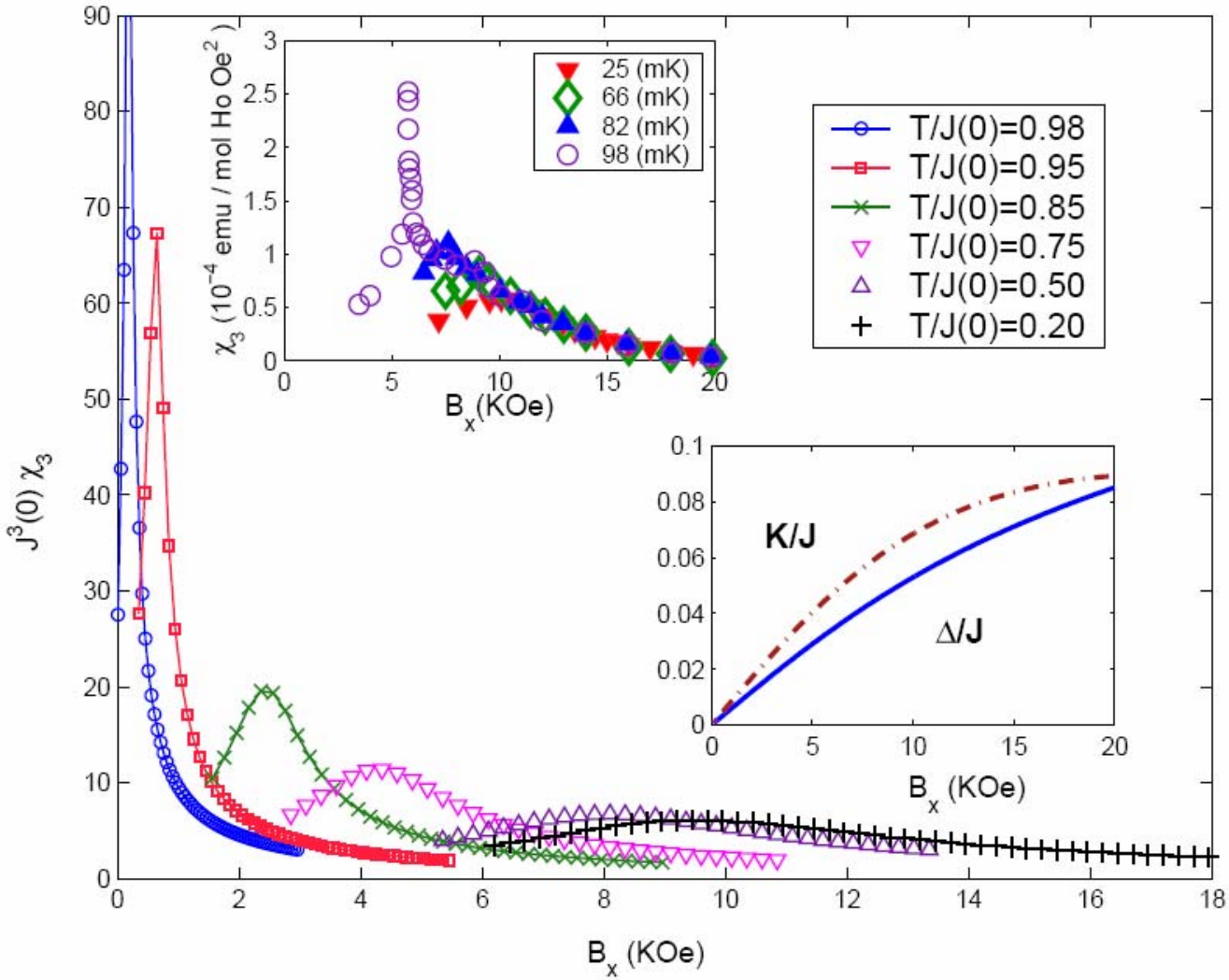


Toy Model

$$H = -\frac{1}{2}\sum_{i,j} J_{ij} S_i^z S_j^z - \frac{1}{2}\sum_{i,j} K_{ij} S_i^x S_j^z - \Gamma \sum_i S_i^x - h_{0z}\sum_i S_i^z - \sum_i h_{ri} S_i^z$$

$$\begin{cases} P(J_{ij}) = \left(\dfrac{N}{2\pi J^2}\right)^{1/2} \exp\left(-NJ_{ij}^2/2J^2\right) \\ P(K_{ij}) = \left(\dfrac{N}{2\pi K^2}\right)^{1/2} \exp\left(-NK_{ij}^2/2K^2\right) \\ P(h_{ri}) = \left(\dfrac{1}{2\pi\Delta}\right)^{1/2} \exp\left(-h_{ri}^2/2\Delta\right) \end{cases}$$

$\frac{K}{J}$, $\frac{\Delta}{J^2}$ and $\frac{\Gamma}{J}$ are functions of B_x (external transverse magnetic field)



Tabei, Gingras, Kao *et al.*, Phys. Rev. Lett., **97**, 237203 (2006).

Conclusion about $LiHo_xY_{1-x}F_4$ in a Transverse Field

- This system is a new realization in a magnetic context of the random field Ising model.

  Yavors'kii *et al.*, Phys. Rev. Lett. **97,** 267203 [2006]

- Similar conclusion reached by Schechter *et al*.

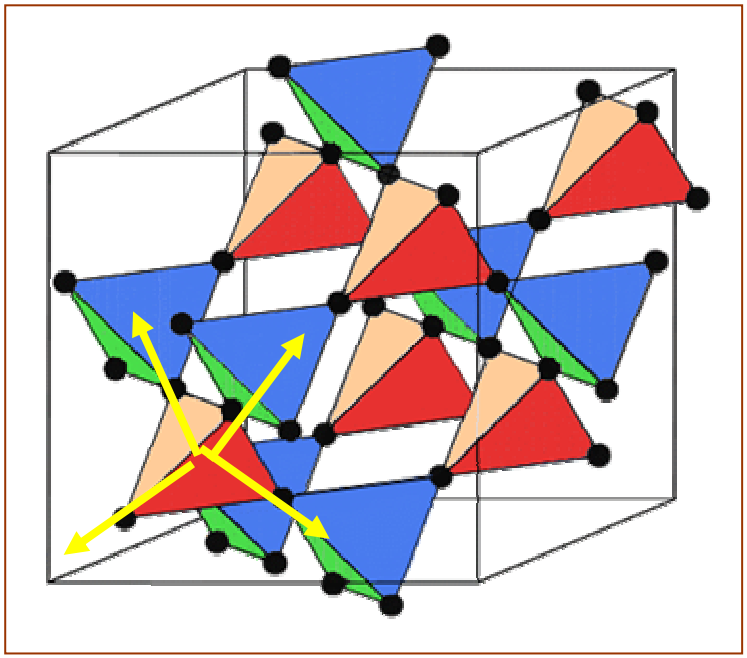
  M. Schechter and N. Laflorencie; Phys. Rev. Lett. 97, 137204 (2006)

- Some evidence from experiment that RFIM is at work in $LiHo_xY_{1-x}F_4$ in FM regime of x:

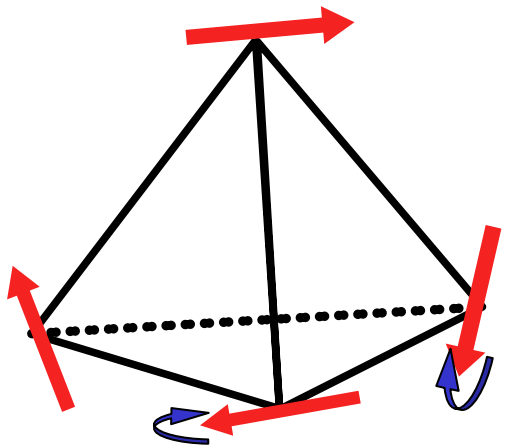
  D. M. Silevitch *et al.; Nature* **448**, 567-570 (Aug. 2007)

$Tb_2Ti_2O_7$ Pyrochlore

•Complex lattice with 16 atoms in conventional cubic unit cell



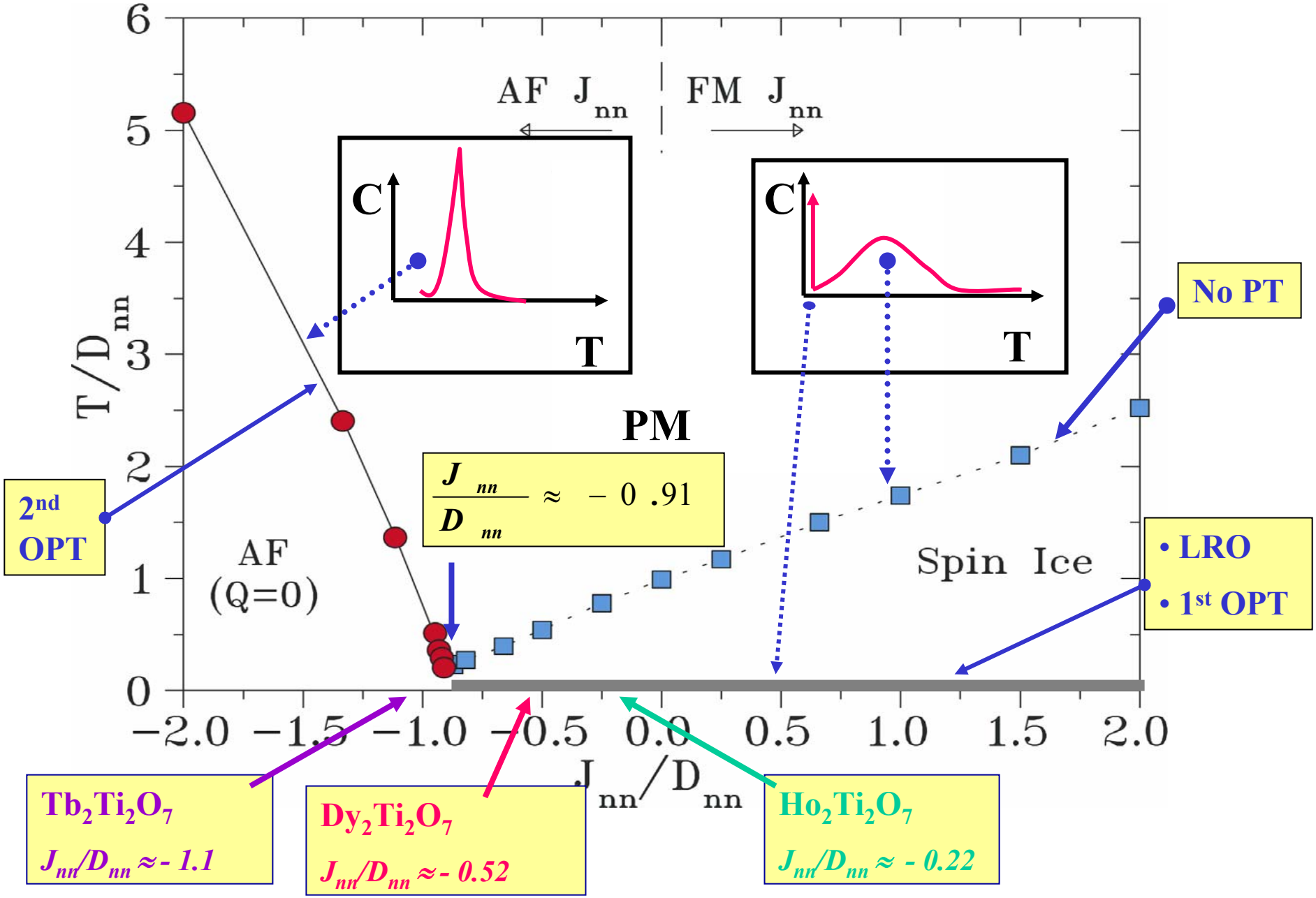
$$\vec{S}_{\Delta,1} + \vec{S}_{\Delta,2} + \vec{S}_{\Delta,3} + \vec{S}_{\Delta,4} = 0$$



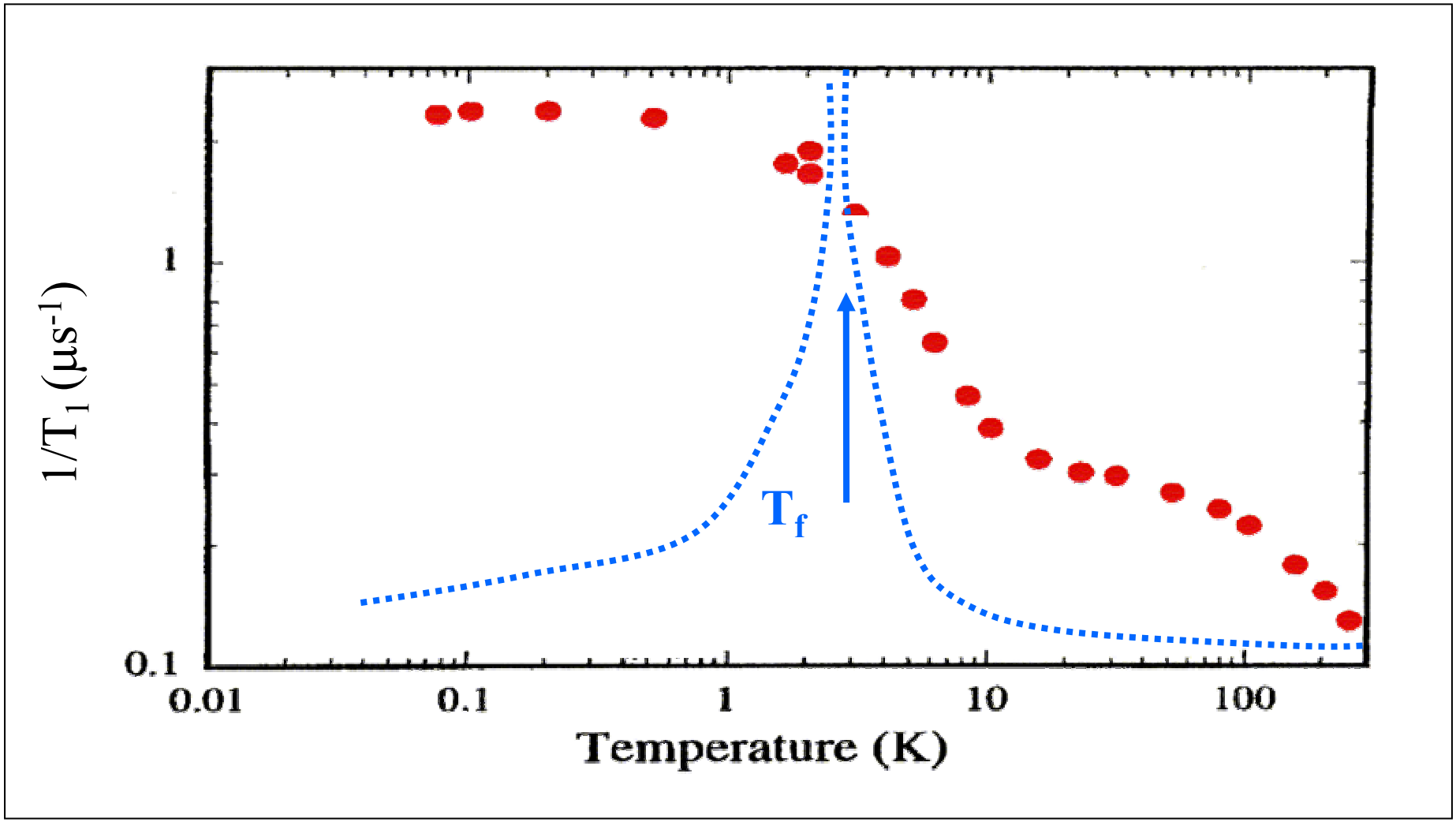
However, crystalline field effects and dipolar interactions play an important role in $Re_2Ti_2O_7$ pyrochlores.

H. Molavian, B. Canals and M.J.P. Gingras , Phys. Rev. Lett. **98**, 157204 (2007).

Monte Carlo Phase Diagram of the Dipolar Spin Ice Model

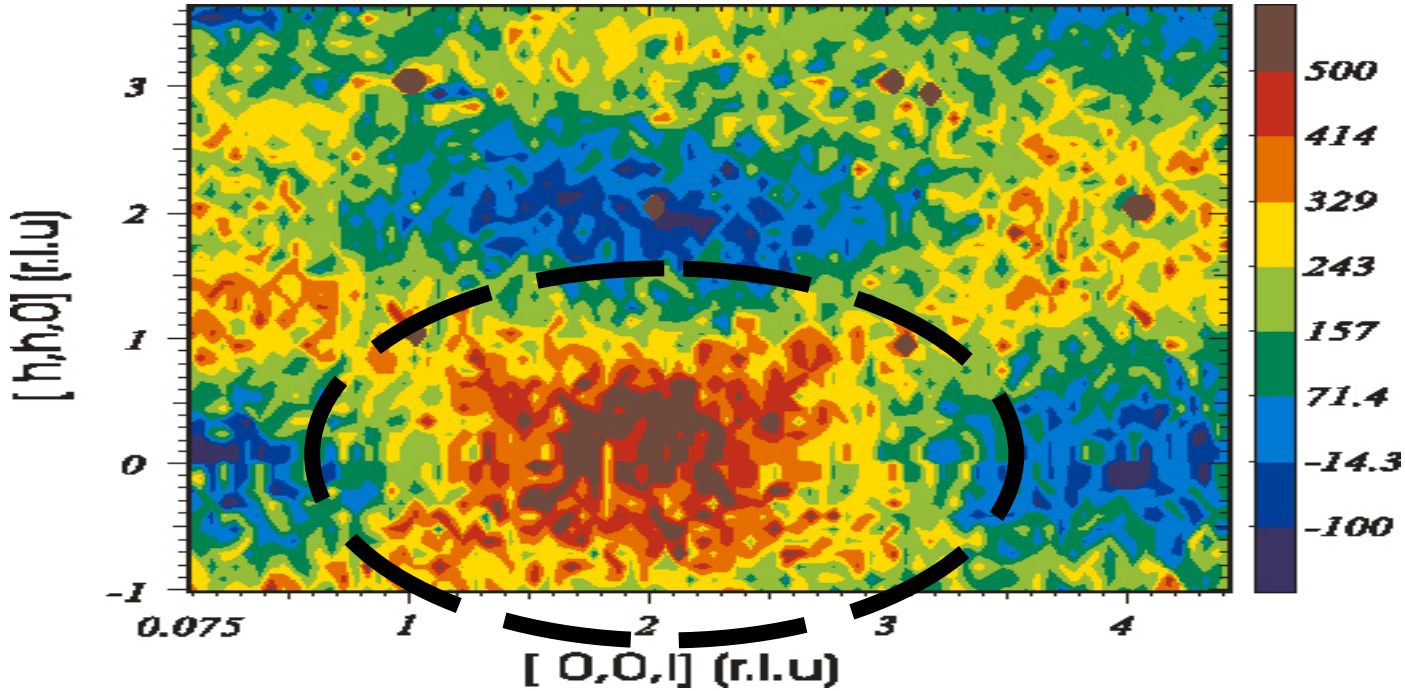


Muon Spin Relaxation Study of $Tb_2Ti_2O_7$



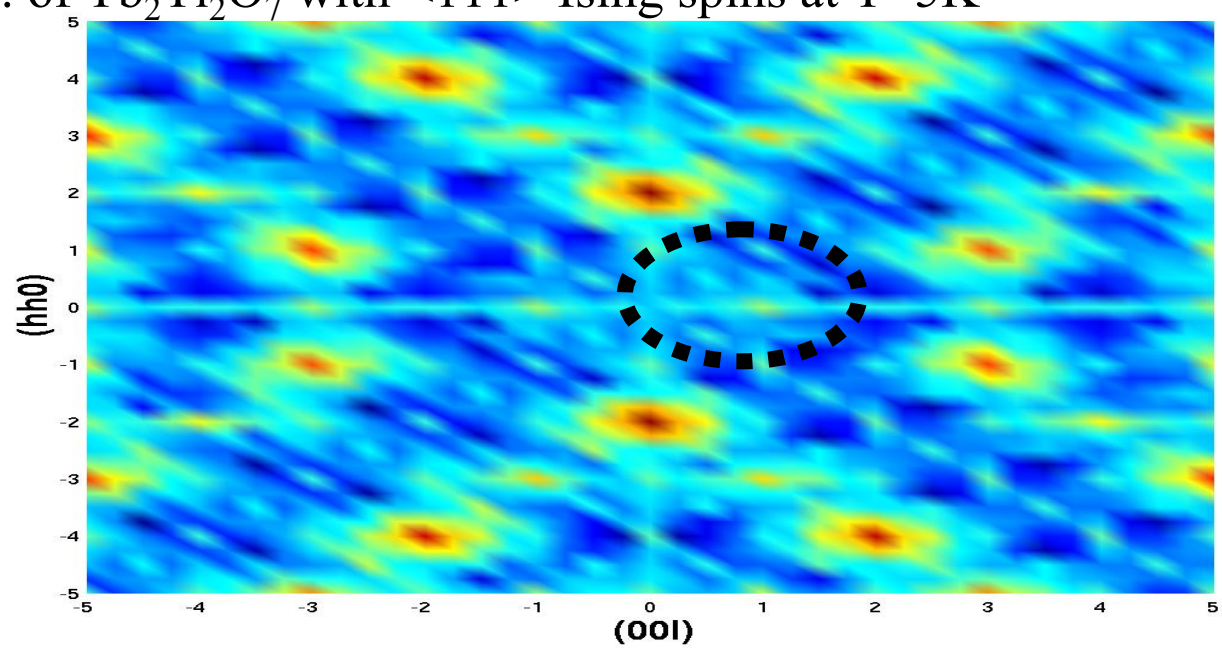
J.S. Gardner *et al.* Phys. Rev. Lett. **82**, 1012 (1999)

Experiment



M.C. of $Tb_2Ti_2O_7$ with <111> Ising spins at T=5K

Monte Carlo



$|m_J\rangle$ wavefunction decomposition

Dy^{3+} (J=15/2)

~ 350 K

$|\pm 15/2\rangle + O(10^{-1})$

Ho^{3+} (J=8)

~ 280 K

$|\pm 8\rangle + O(10^{-1})$

Tb^{3+} (J=6)
(should be a nonmagnetic singlet, as Tm^{3+} !)

$\{ |+3\rangle, |-3\rangle, |+6\rangle, -6\rangle\}$

~ 20 K

$\{ |-3\rangle, |+6\rangle, -6\rangle\}$

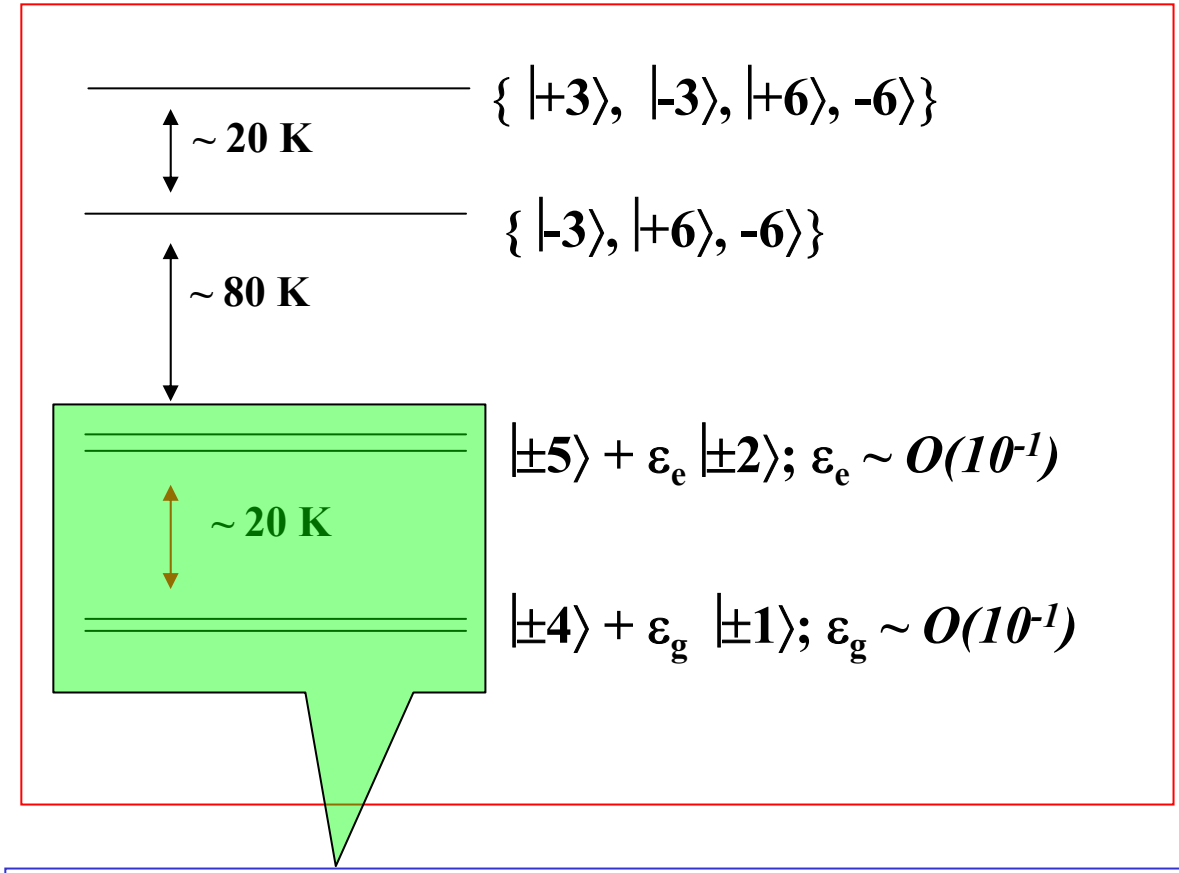
~ 80 K

$|\pm 5\rangle + \varepsilon_e |\pm 2\rangle; \varepsilon_e \sim O(10^{-1})$

~ 20 K

$|\pm 4\rangle + \varepsilon_g |\pm 1\rangle; \varepsilon_g \sim O(10^{-1})$

Gingras *et al.*, Phys. Rev. B **61**, 6496 (2000)

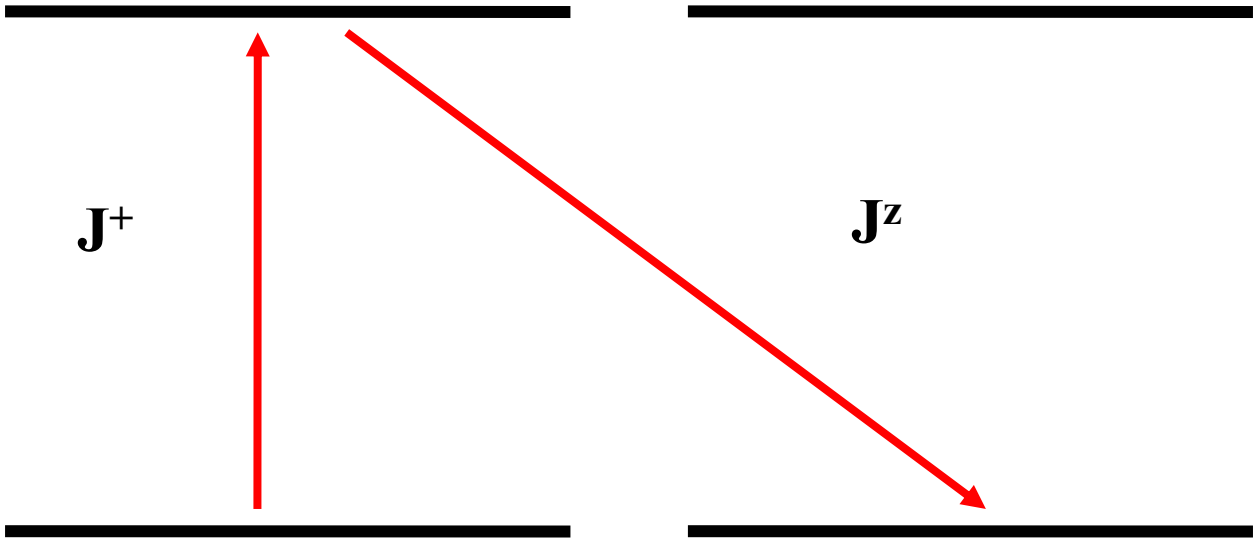


- The issue is that this gap is not very much larger than the exchange and dipolar interactions, H_{int}.
- Hence, H_{int} induces admixing via virtual transitions among CEF levels.
- This problem is not unrelated to the one of multispin (ring-exchange) interactions generated via double-occupancy in the Hubbard model.

Gingras *et al.*, Phys. Rev. B **61**, 6496 (2000)

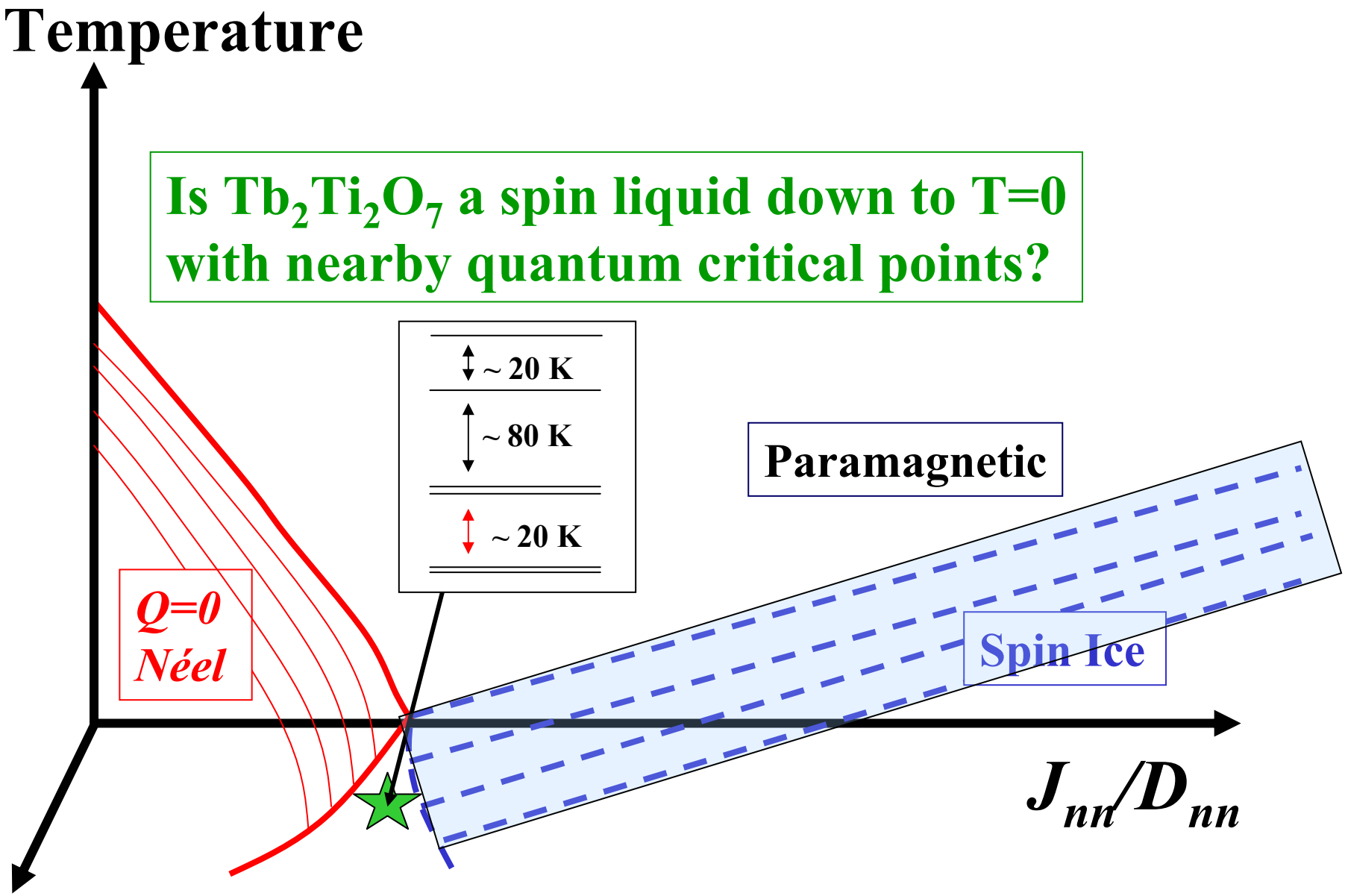
Quantum Fluctuations via Excited States

$$|\psi_e\rangle_1 \approx |5\rangle + \beta|-4\rangle \qquad |\psi_e\rangle_2 \approx -|-5\rangle + \beta|4\rangle$$



$$|\psi_0\rangle_1 \approx |4\rangle - \alpha|-5\rangle \qquad |\psi_0\rangle_2 \approx |-4\rangle + \alpha|5\rangle$$

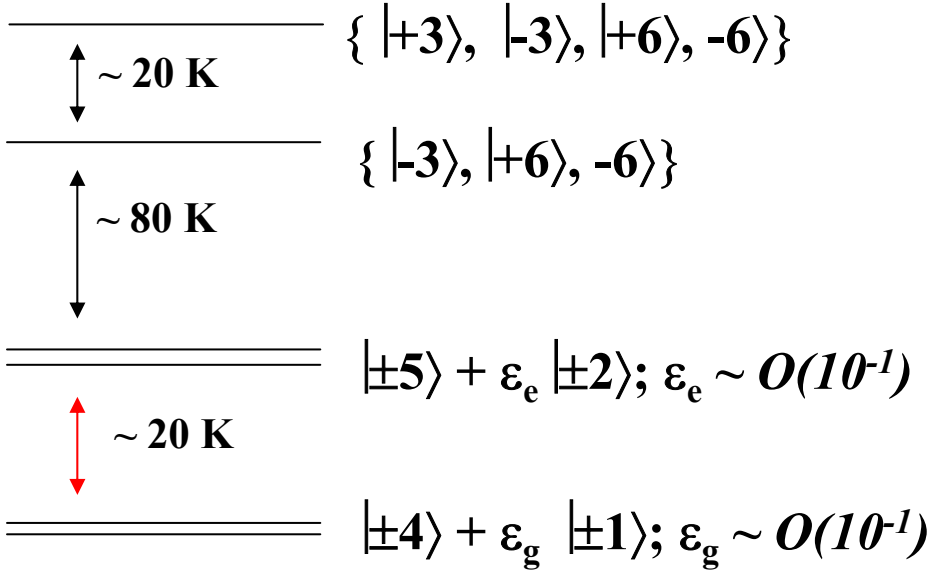
There is a mechanism for spin flips, and some isotropy can be restored.

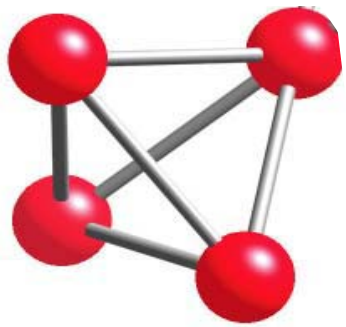


The "third axis" (e.g. finite anisotropy {crystal field gap Δ} controls quantum fluctuations)

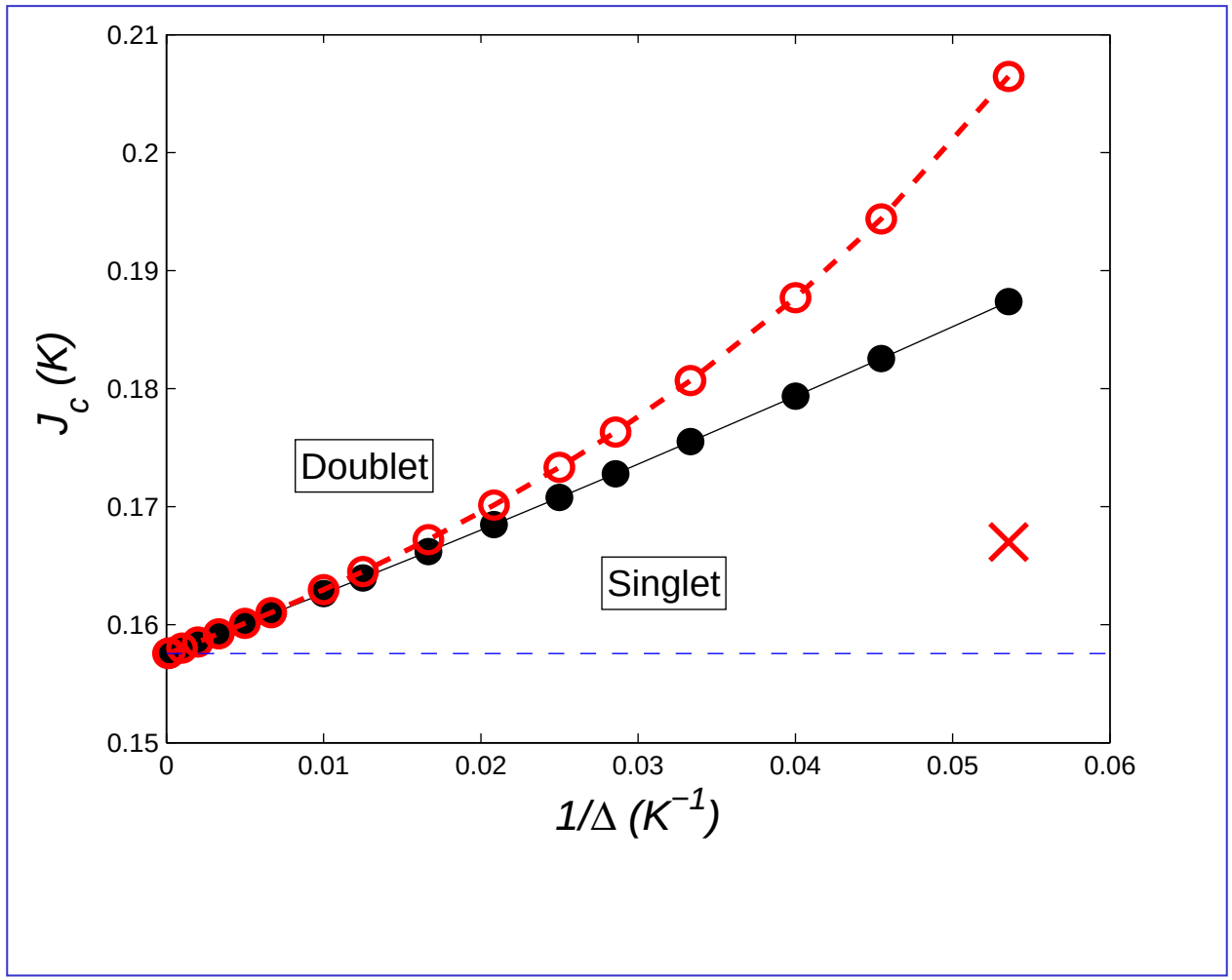
Exact Diagonalization of Single Tetrahedron

Shed some light on quantum fluctuation channels





Single Tetrahedron Exact Diagonalization



Effective Hamiltonian Method

$$H = J\sum_{\langle i,j\rangle}\vec{\mathrm{J}}_i \bullet \vec{\mathrm{J}}_j + D\sum_{j>i}\left(\frac{\vec{\mathrm{J}}_i \bullet \vec{\mathrm{J}}_j}{R_{ij}^{\ 3}} - 3\frac{(\vec{\mathrm{J}}_i \bullet \vec{R}_{ij})(\vec{\mathrm{J}}_j \bullet \vec{R}_{ij})}{R_{ij}^{\ 5}}\right)$$

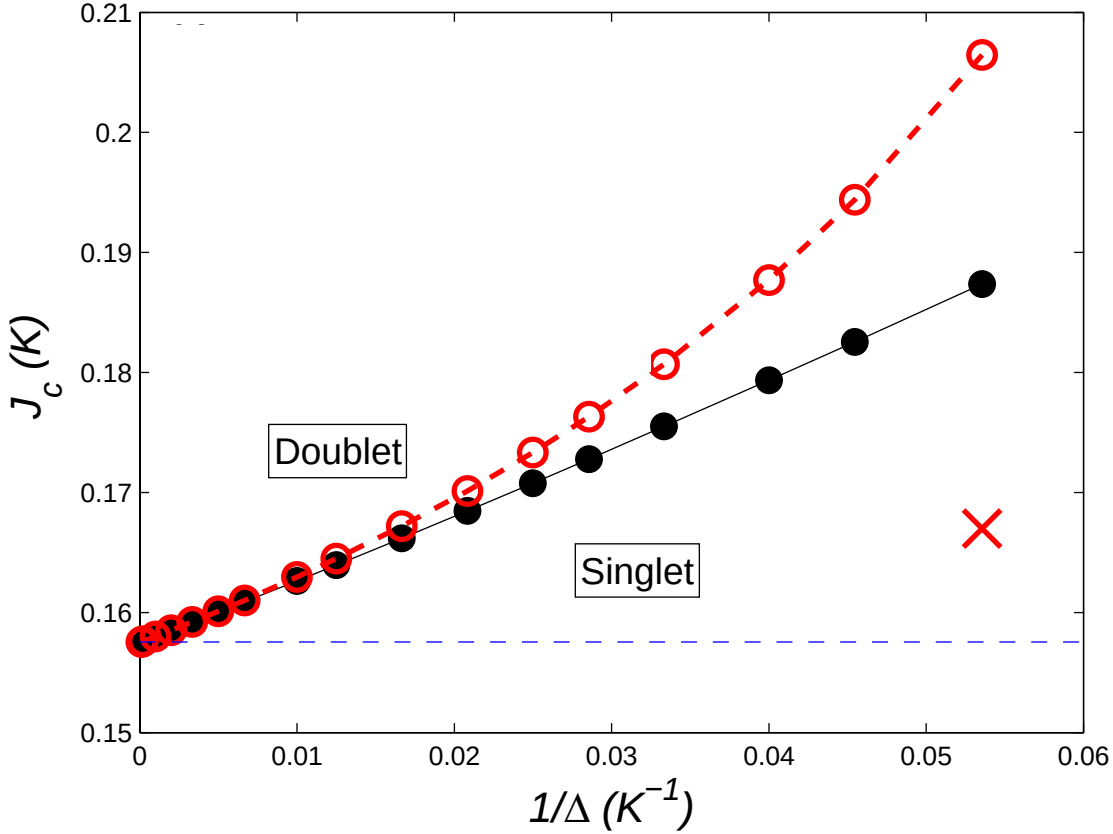
$$\mathrm{P} = \sum_{\alpha\in \mathrm{P}} |\alpha\rangle\langle\alpha|$$

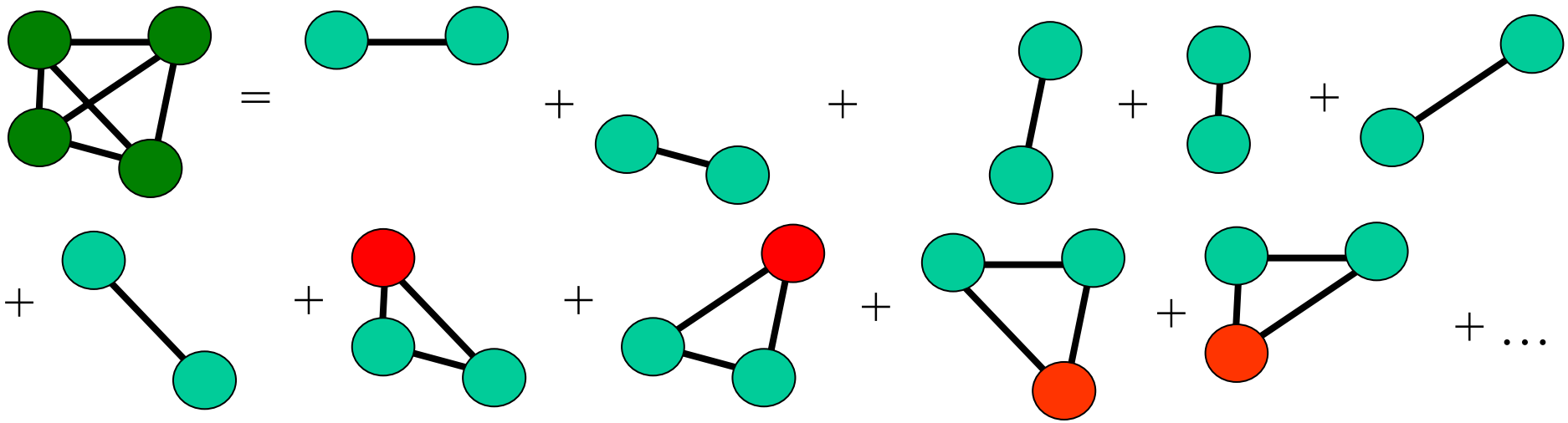
$$R = \sum_{\beta\notin P}\frac{|\beta\rangle\langle\beta|}{E_0^{\alpha} - E_0^{\beta}}$$

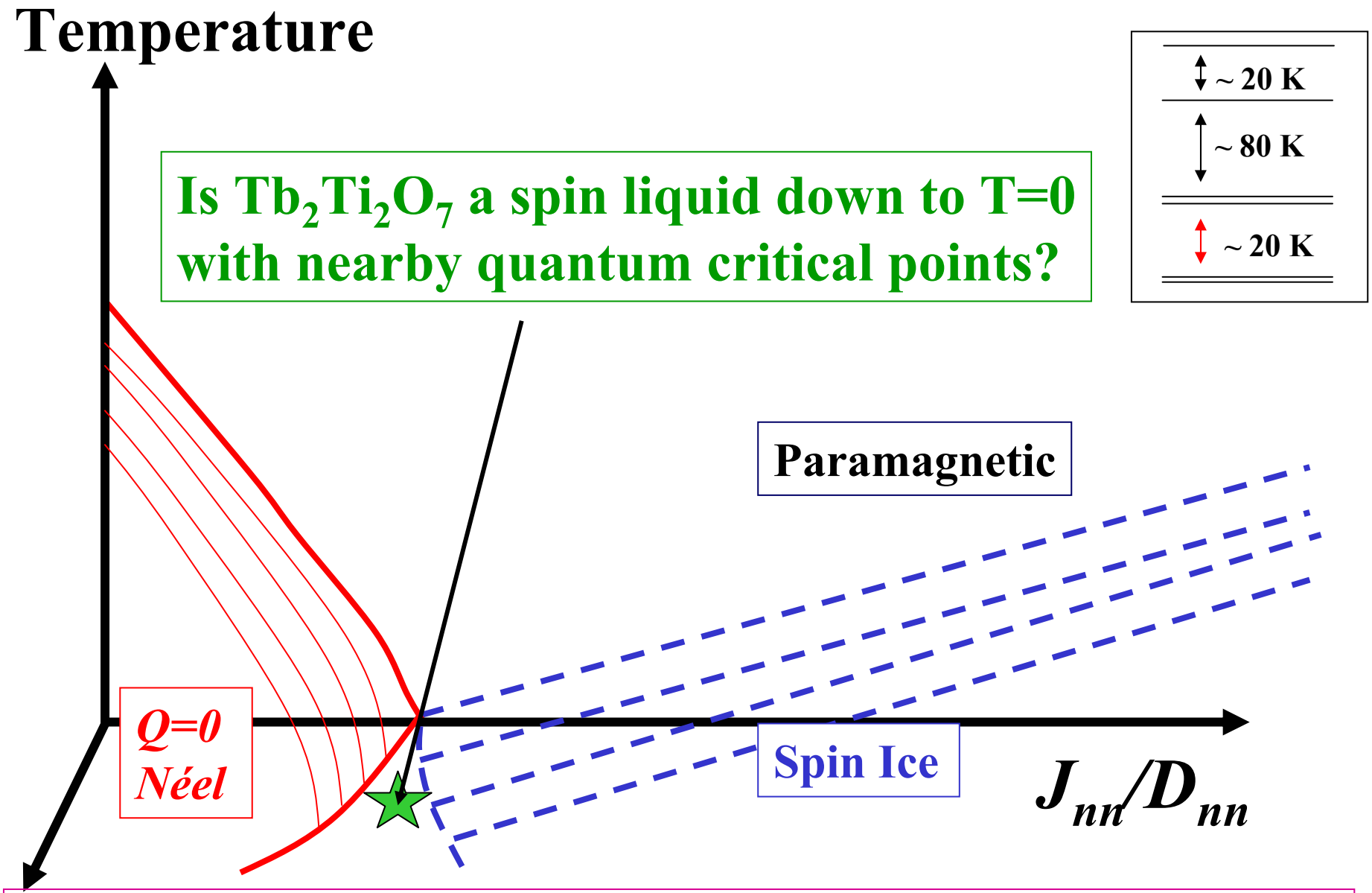
$$H_{\mathrm{eff}} = PHP + PHRHP + \cdots$$

$$H_{\mathrm{eff}} = PH_{\mathrm{ex}}P + PH_{\mathrm{dip}}P + PH_{\mathrm{ex}}RH_{\mathrm{ex}}P$$
$$+(PH_{\mathrm{ex}}RH_{\mathrm{dip}}P + PH_{\mathrm{dip}}RH_{\mathrm{ex}}P) + PH_{\mathrm{dip}}RH_{\mathrm{dip}}P$$

N-body quantum effects play an important role in the renormalization of the Néel – spin-ice boundary.







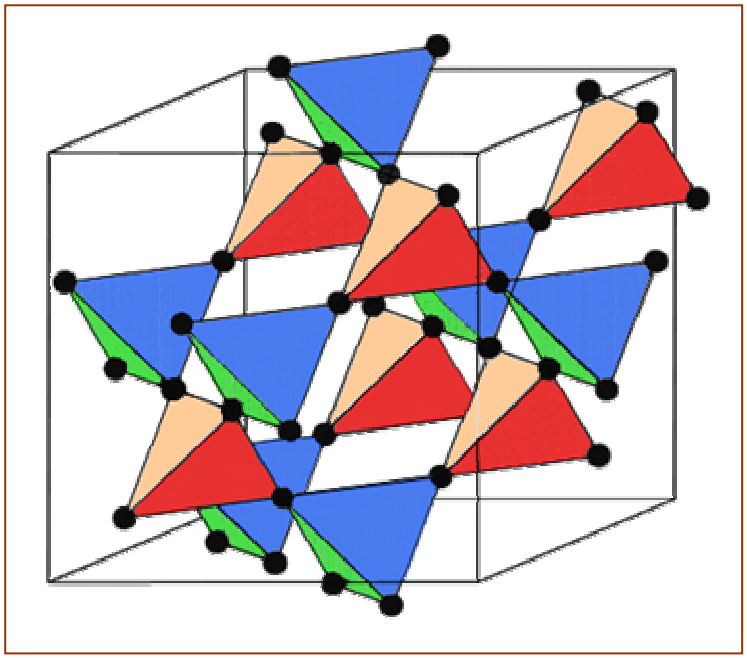
The "third axis" (e.g. finite anisotropy {1 over the *crystal field gap* Δ} *controls quantum fluctuations)*

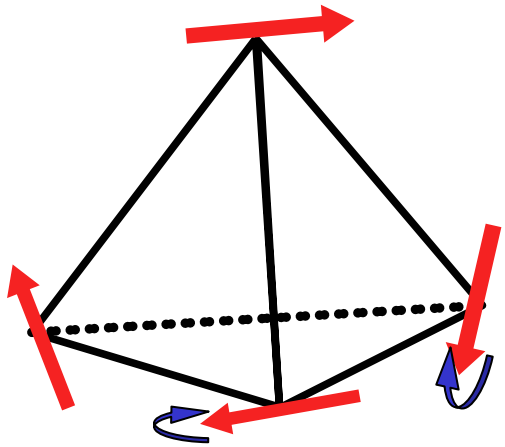
Conclusion about $Tb_2Ti_2O_7$

- $Tb_2Ti_2O_7$ is an exotic material system – akin to a genuine 3D spin liquid.

  Why? Perhaps is pushed into a quantum disordered "*quantum spin ice*" regime. More experiments and calculations are needed.

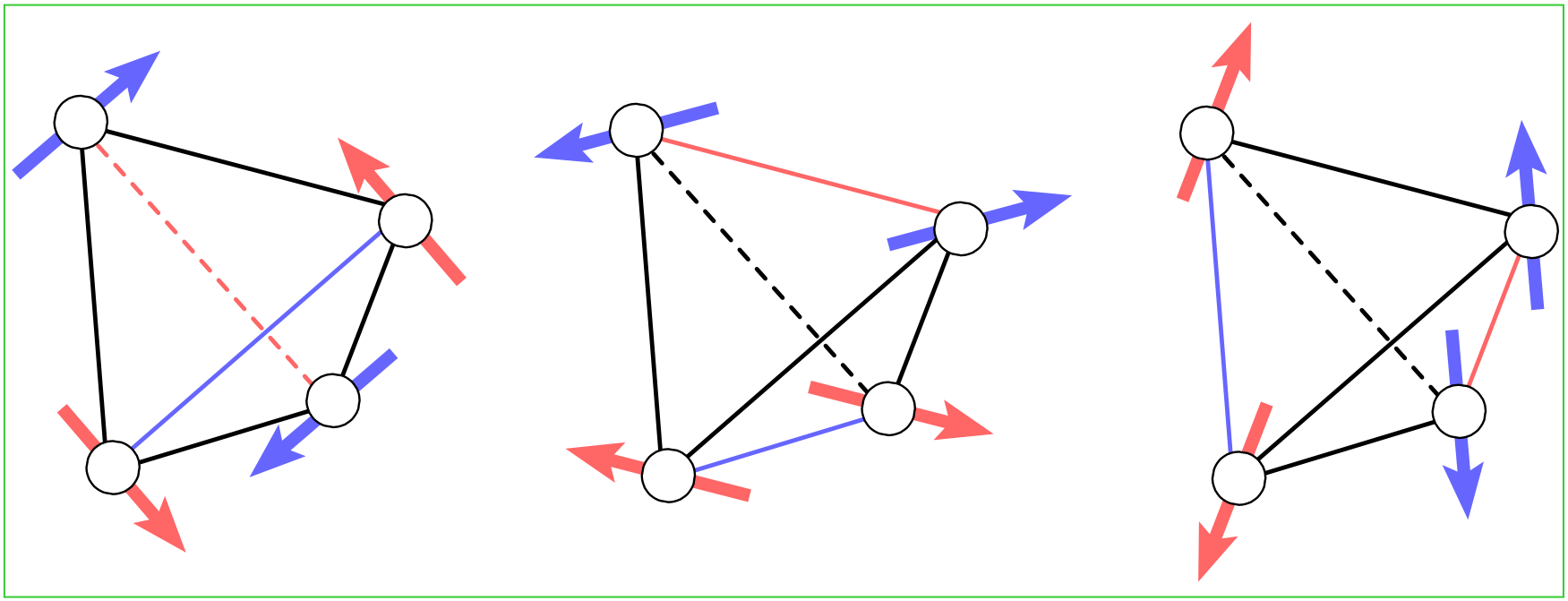
$Gd_2Sn_2O_7$ Pyrochlore

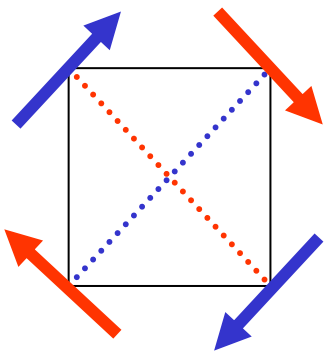




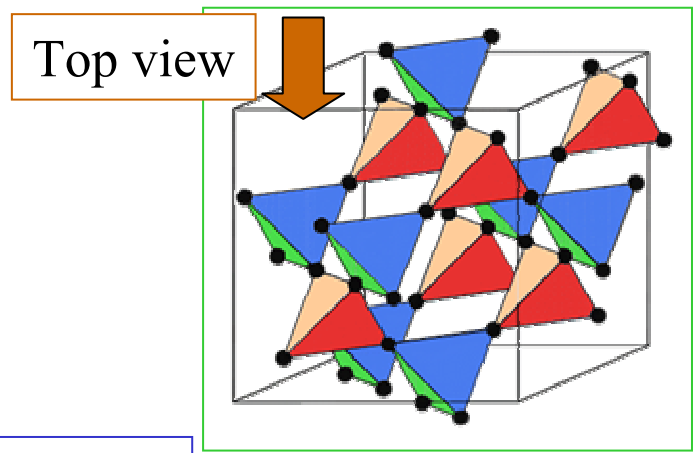
- A. Del Maestro, M. J. P. Gingras; Phys. Rev. B **76**, 064418 (2007).
- J.A. Quilliam, K.A. Ross, A.G. Del Maestro, M.J.P. Gingras, L.R. Corruccini and J.B. Kycia; Phys. Rev. Lett. **99**, 097201 (2007).

The neutron diffraction below T=1 K is well described by the so-called Palmer-Chalker ground states (3 discrete g.s. (×2))





"top" (001) view



S. E. Palmer and J. T. Chalker, Phys. Rev. B **62**, 488 (2000).

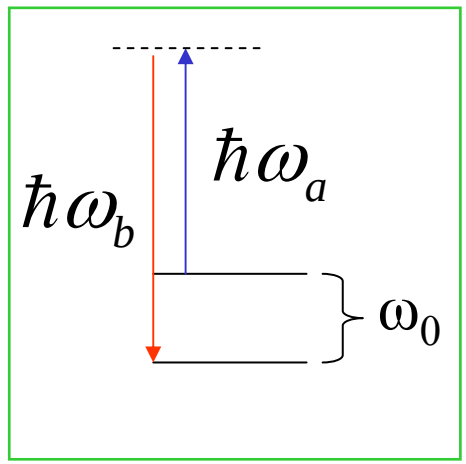
NMR/muSR Relaxation Regimes

For $T \ll T_c$, Raman process

$$\lambda \sim \int_{\Delta}^{\infty} \left[n(\varepsilon / k_{\mathrm{B}} T) \right] \times \left[n(\varepsilon / k_{\mathrm{B}} T) + 1 \right] \left(g(\varepsilon) \right)^2 d\varepsilon$$

$$\lambda \sim T^{\mu} \quad ; \Delta \ll T \ll T_c$$

$$\lambda \sim \exp(-\Delta / T) \; ; \quad T \ll \Delta$$



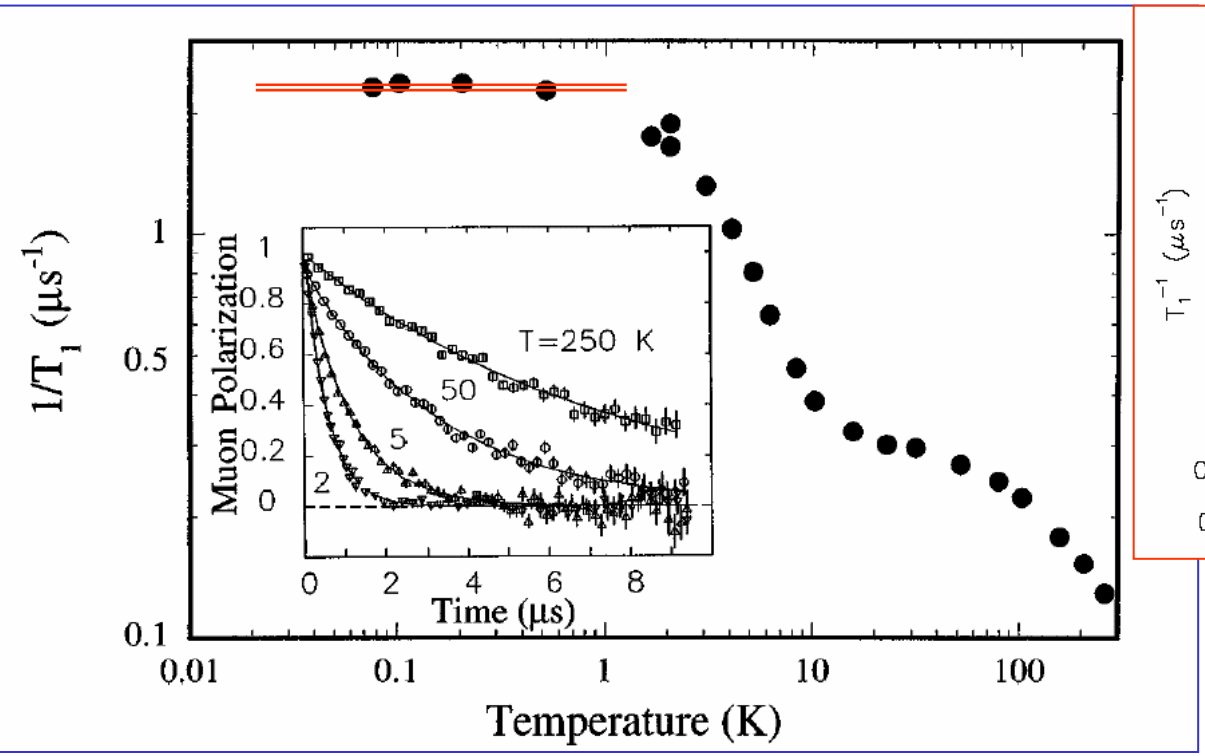
Conversely,

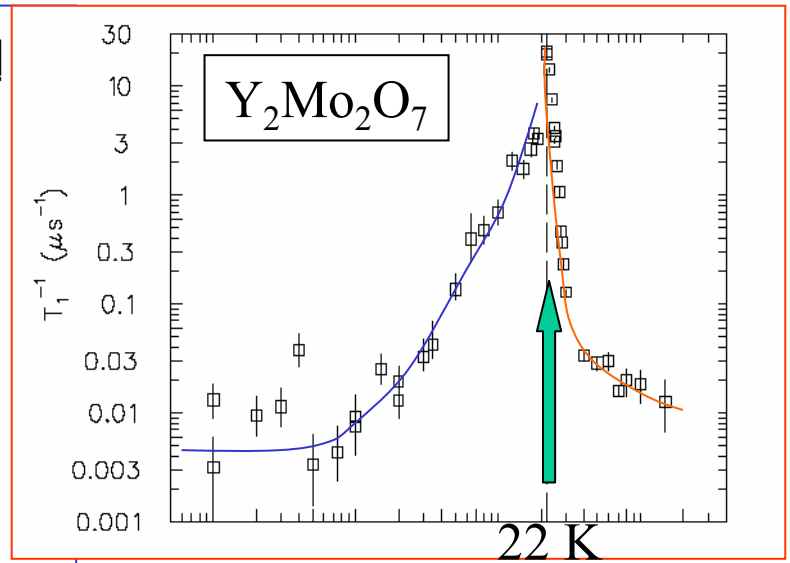
$$\lambda \sim \Lambda^2 / \omega_{\mathrm{ex}} \; ; \; \omega_{\mathrm{ex}} \sim \sqrt{J_{\mathrm{ex}} z S(S+1) / \hbar^2} \; ; T \gg T_c$$

VOLUME 82, NUMBER 5 PHYSICAL REVIEW LETTERS 1 FEBRUARY 1999

Cooperative Paramagnetism in the Geometrically Frustrated Pyrochlore Antiferromagnet $Tb_2Ti_2O_7$

J. S. Gardner,[1] S. R. Dunsiger,[2] B. D. Gaulin,[1] M. J. P. Gingras,[3] J. E. Greedan,[4] R. F. Kiefl,[2] M. D. Lumsden,[1] W. A. MacFarlane,[2] N. P. Raju,[4] J. E. Sonier,[2] I. Swainson,[5] and Z. Tun[5]



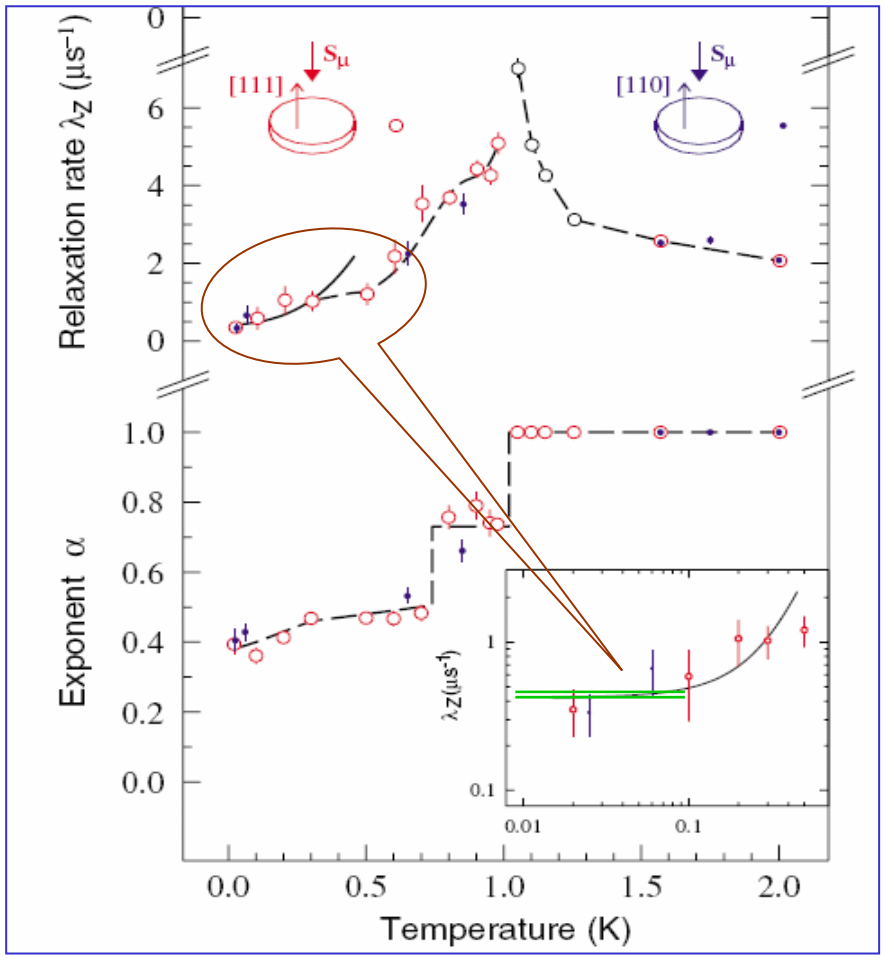


No order seen via any techniques down to ~ 50 mK

PRL 95, 047203 (2005) PHYSICAL REVIEW LETTERS week ending 22 JULY 2005

Magnetic Density of States at Low Energy in Geometrically Frustrated Systems

A. Yaouanc,[1] P. Dalmas de Réotier,[1] V. Glazkov,[1] C. Marin,[1] P. Bonville,[2] J. A. Hodges,[2] P. C. M. Gubbens,[3] S. Sakarya,[3] and C. Baines[4]



PHYSICAL REVIEW B 73, 172418 (2006)

Magnetic field dependence of muon spin relaxation in geometrically frustrated $Gd_2Ti_2O_7$

S. R. Dunsiger,[1,*] R. F. Kiefl,[2,3] J. A. Chakhalian,[4] J. E. Greedan,[5] W. A. MacFarlane,[6,3] R. I. Miller,[3] G. D. Morris,[3] A. N. Price,[7] N. P. Raju,[8] and J. E. Sonier[9]

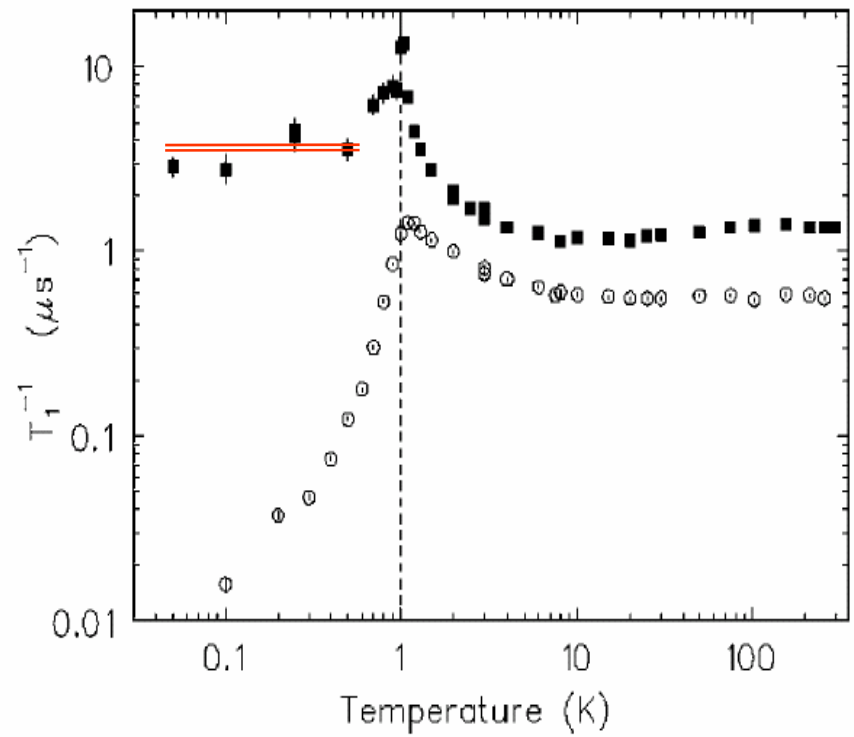


FIG. 2. Spin relaxation rate in $Gd_2Ti_2O_7$ as a function of temperature in longitudinally applied magnetic fields of 5 mT (filled squares) and 4 T (open circles).

- Two-step transition to long-range order.
- Mechanisms and ground state not fully understood.
- **Yet,** neutron sees long-range order below 700 mK.

PRL 96, 127202 (2006) PHYSICAL REVIEW LETTERS week ending 31 MARCH 2006

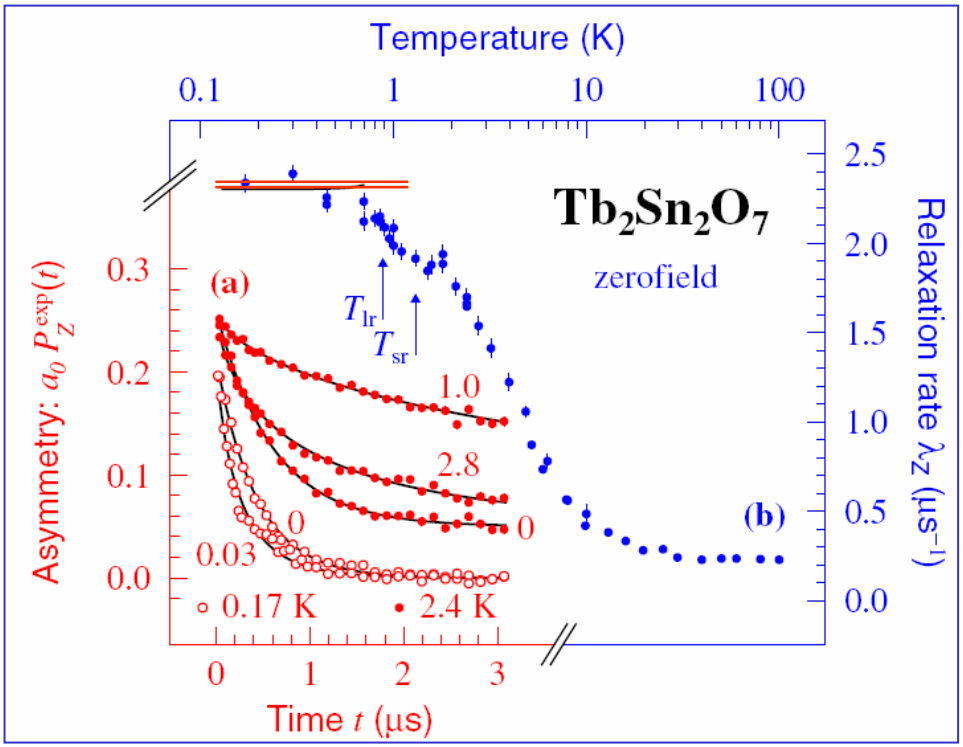
Spin Dynamics and Magnetic Order in Magnetically Frustrated $Tb_2Sn_2O_7$

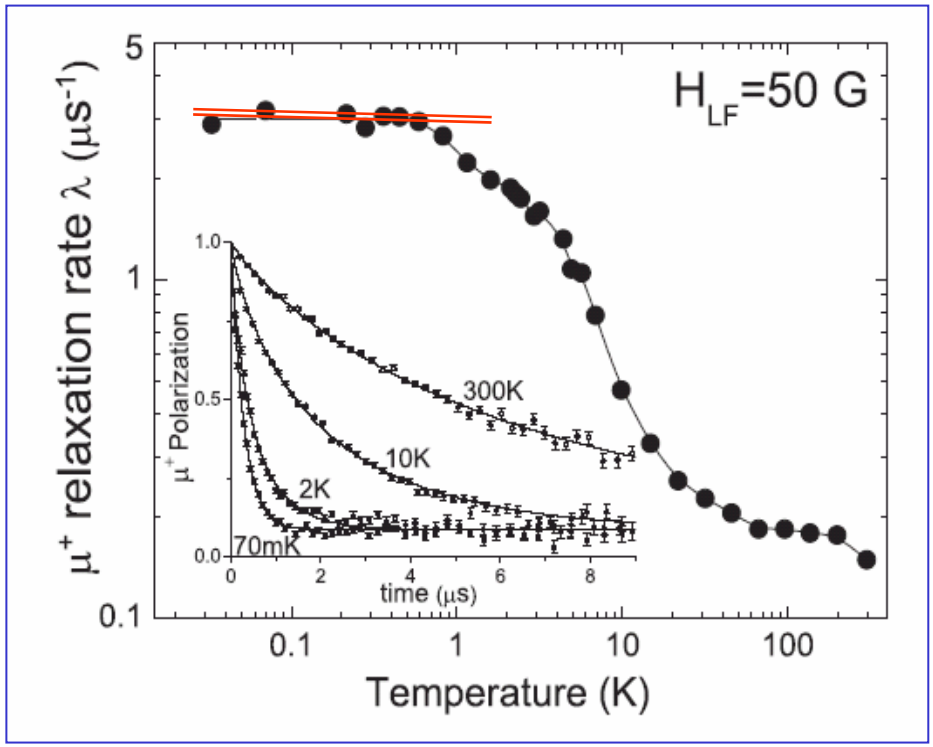
P. Dalmas de Réotier,[1] A. Yaouanc,[1] L. Keller,[2] A. Cervellino,[2,*] B. Roessli,[2] C. Baines,[3] A. Forget,[4] C. Vaju,[1] P.C.M. Gubbens,[5] A. Amato,[6] and P.J.C. King[7]

PRL 97, 117203 (2006) PHYSICAL REVIEW LETTERS week ending 15 SEPTEMBER 2006

Direct Evidence for a Dynamical Ground State in the Highly Frustrated $Tb_2Sn_2O_7$ Pyrochlore

F. Bert,[1] P. Mendels,[1] A. Olariu,[1] N. Blanchard,[1] G. Collin,[2] A. Amato,[3] C. Baines,[3] and A. D. Hillier[4]



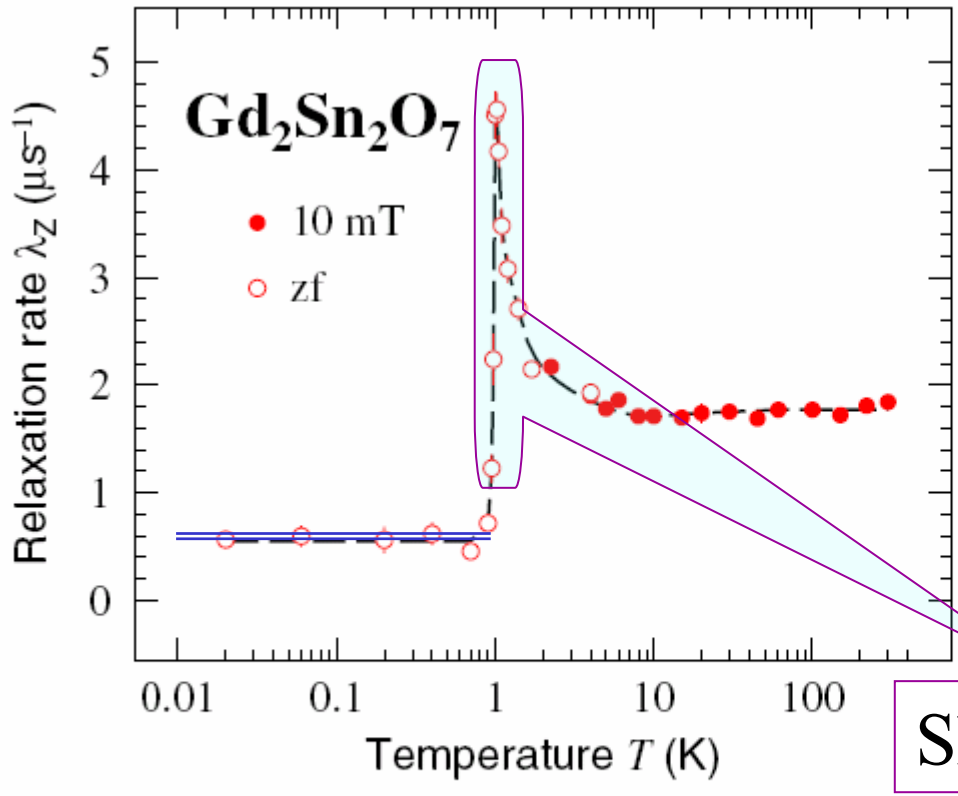


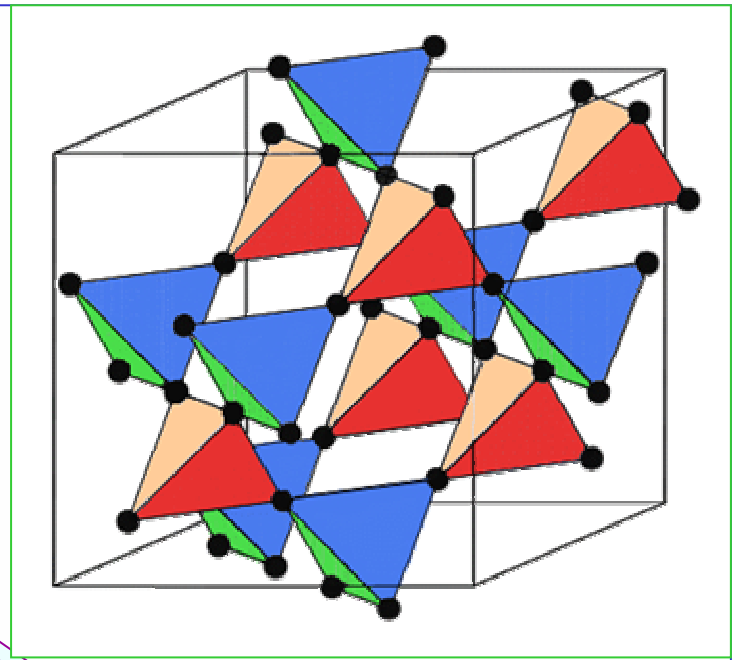
But neutron scattering sees a long-range ordered spin ice state: Mirebeau *et al.*, Phys. Rev. Lett. **94**, 246402 (2005).

Summary

- $(Tb,Gd,Er,Yb)_2(Ti/Sn)_2O_7$ pyrochlores display $\lambda>0$ down to the lowest temperature.
- This persistent spin dynamics suggests unconventional ground state and/or excitations.
- "Psychologically", one may be able to accept $\lambda>0$ in the previous materials since the ground state is all of them lacks a complete understanding.
- **Not so** for the next material ($Gd_2Sn_2O_7$) since neutron scattering reveals long range order.

MuSR on $Gd_2Sn_2O_7$



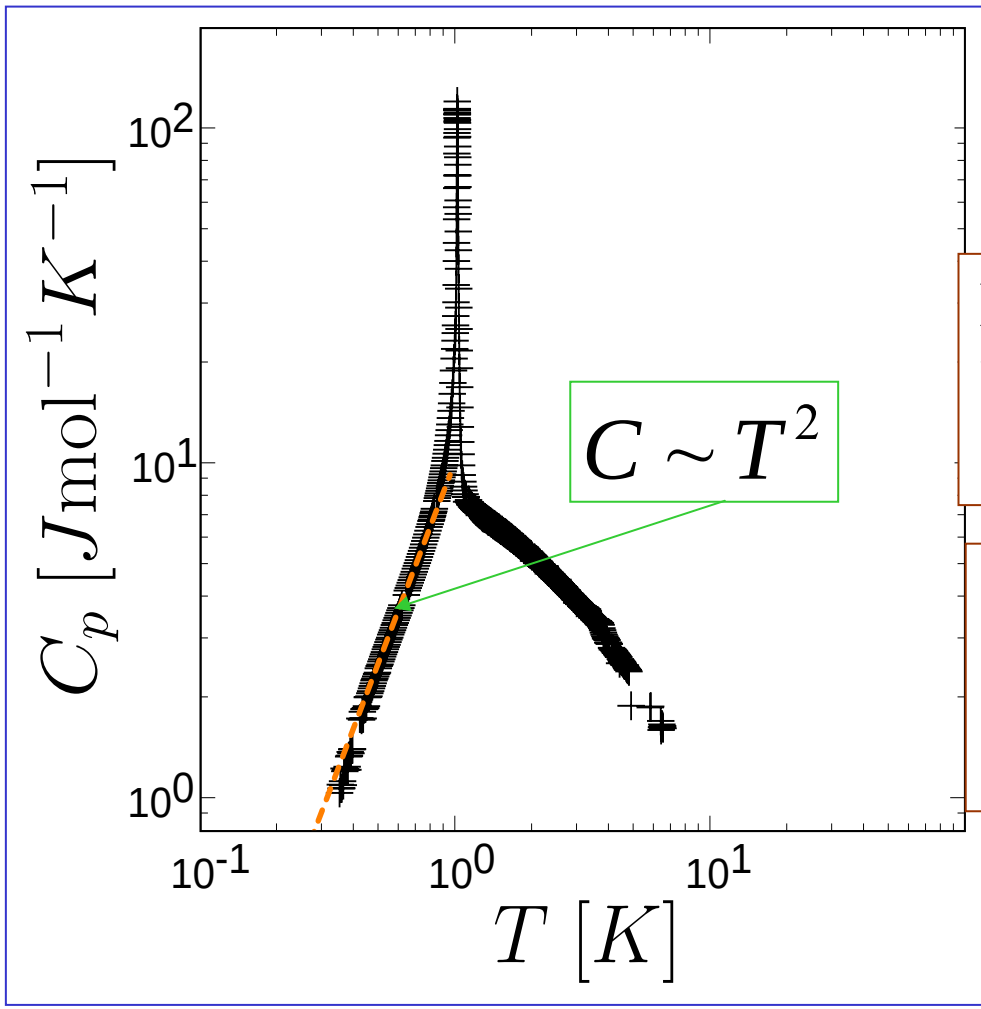


Sharp transition

Figure 7. Spin–lattice relaxation rate λ_Z versus temperature for $Gd_2Sn_2O_7$ measured for two values of B_{ext}: 0 and 10 mT. The line is a guide to the eyes.

P. Dalmas de Réotier, P. C. M. Gubbens and A. Yaouanc
J. Phys.: Condens. Matter **16**, S4687(2004).

Interestingly … the specific heat has also been interpreted as unconventional



In conventional three-dimensional long-range ordered antiferromagnets one expects T:

$$C \sim T^3 \ ; \Delta \ll T \ll T_c$$

$$C \sim \exp(-\Delta / T) \ ; T \ll \Delta$$

P. Bonville *et al*, J. Phys. Condens. Matter **15,** 7777 (2003).

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

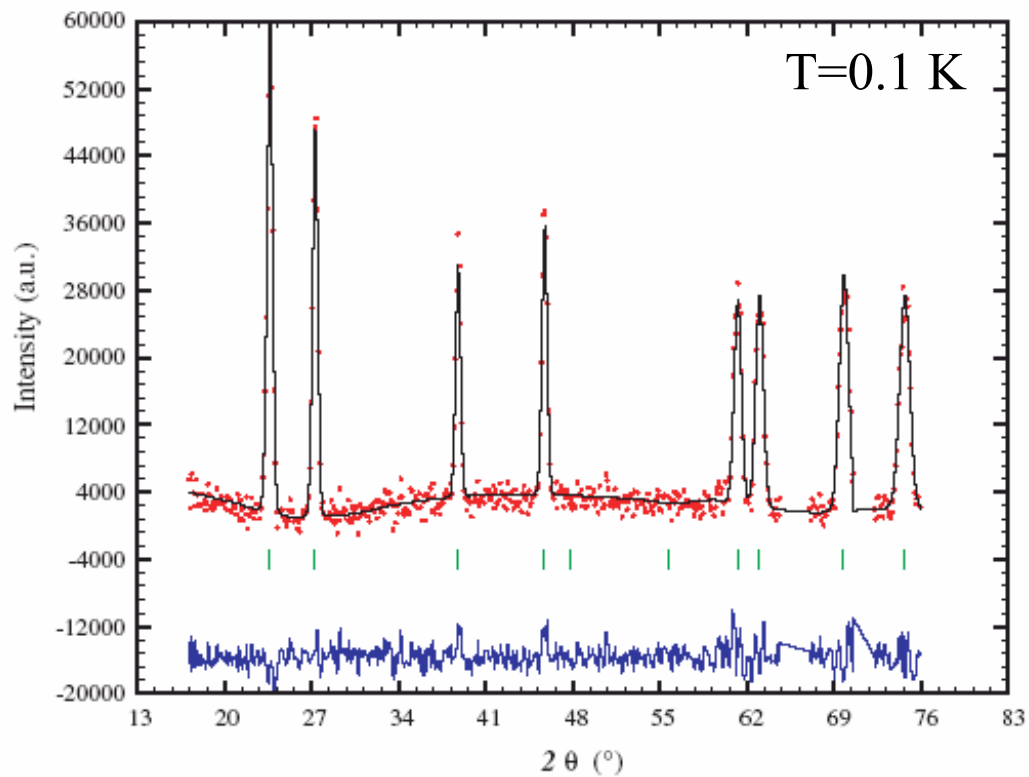
J. Phys.: Condens. Matter **18** (2006) L37–L42 doi:10.1088/0953-8984/18/3/L02

LETTER TO THE EDITOR

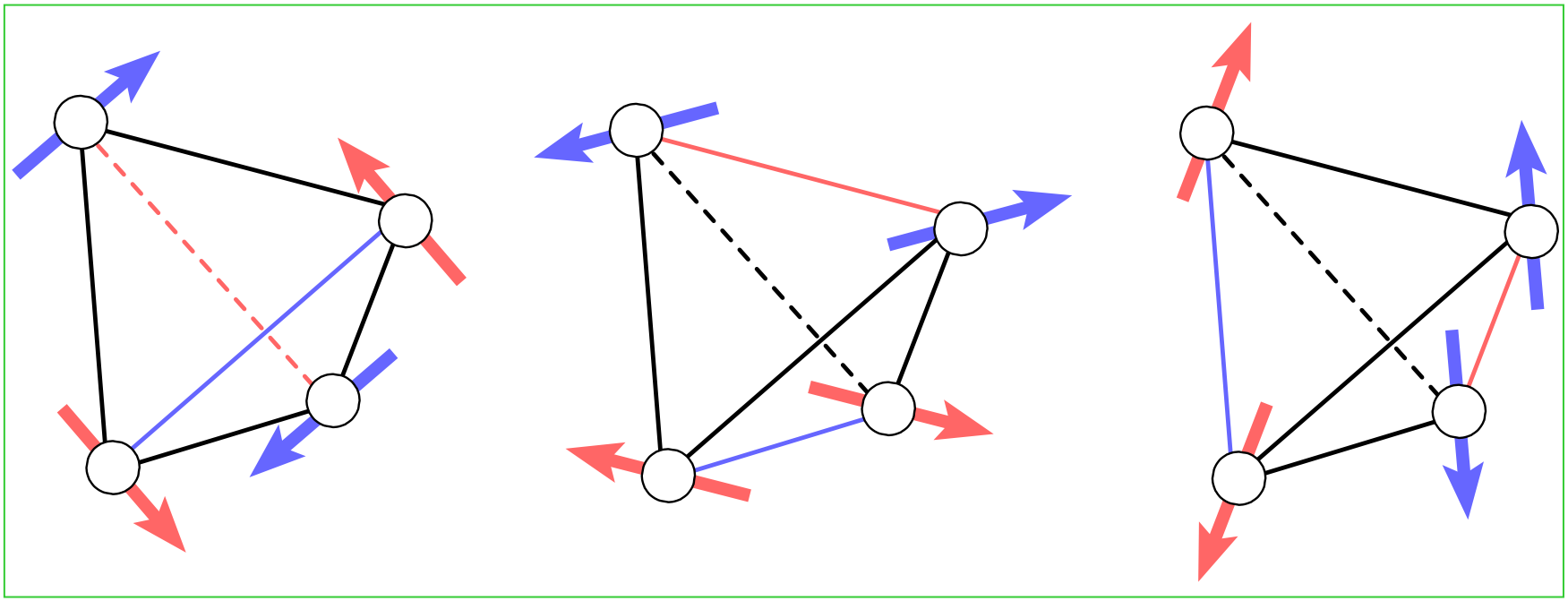
Magnetic ordering in $Gd_2Sn_2O_7$: the archetypal Heisenberg pyrochlore antiferromagnet

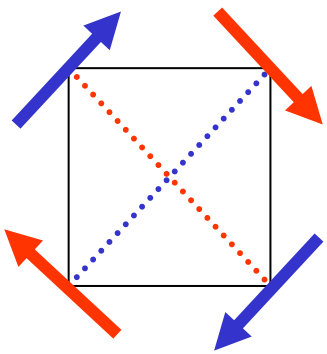
A S Wills[1,2], M E Zhitomirsky[3], B Canals[4], J P Sanchez[3], P Bonville[5], P Dalmas de Réotier[3] and A Yaouanc[3]

But neutron scattering sees long-range order below 1 K:

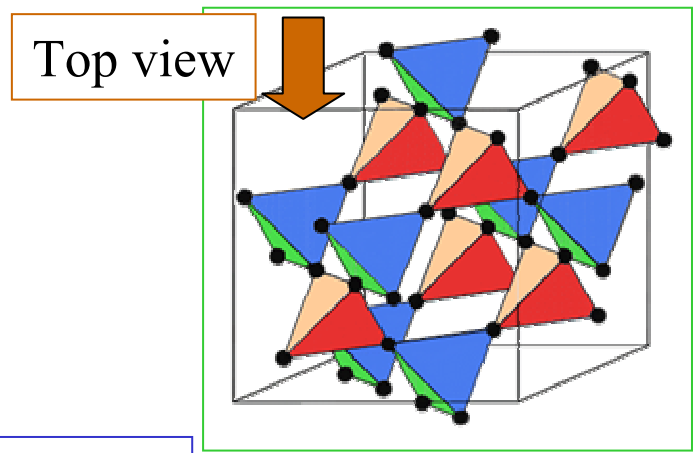


The neutron diffraction below T=1 K is well described by the so-called Palmer-Chalker ground states (3 discrete g.s. (×2))





"top" (001) view

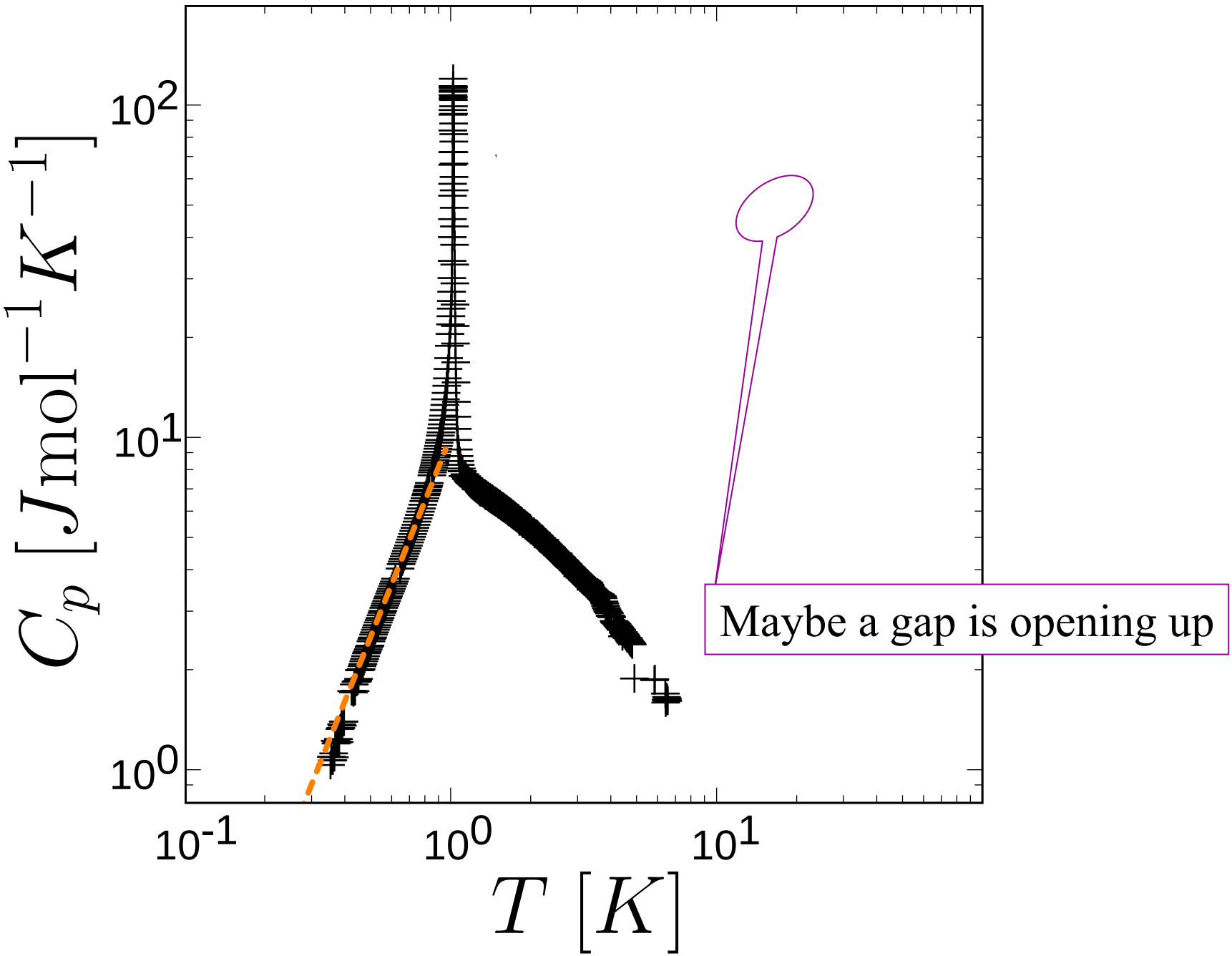


S. E. Palmer and J. T. Chalker, Phys. Rev. B **62**, 488 (2000).

This is “interesting” …

The identification of the PC ground state via neutron diffraction motivates a microscopic description.

A. G. del Maestro and M. J. P. Gingras; “*Low temperature specific heat and possible gap to magnetic excitations in the Heisenberg pyrochlore antiferromagnet* $Gd_2Sn_20_7$”; Phys. Rev. B **76**, 064418 (2007).


$C_p\ [J\mathrm{mol}^{-1}K^{-1}]$
10^2
10^1
10^0
10^{-1}
10^0
10^1
$T\ [K]$
Maybe a gap is opening up

Magnetic Interactions Involved

$$H = -\sum_{j>i} J_{ij}(|\vec{r}_{ij}|)\,\vec{\mathrm{J}}_i \bullet \vec{\mathrm{J}}_j$$

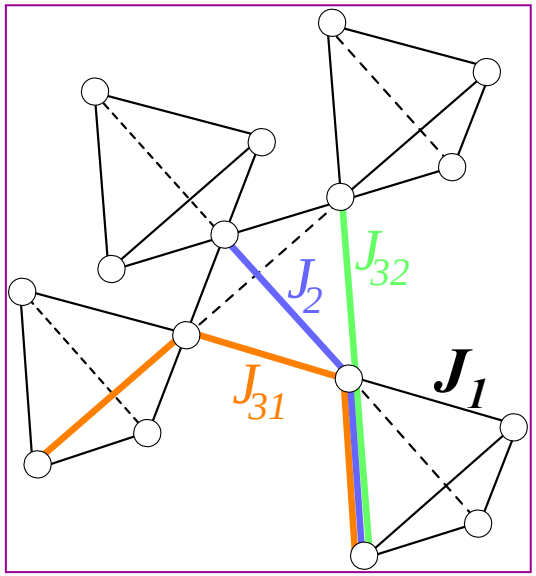
$$+\ \frac{\mu_0}{4\pi}(g\mu_{\mathrm{B}})^2 \sum_{j>i} \frac{\vec{\mathrm{J}}_i \bullet \vec{\mathrm{J}}_j}{\left|\vec{r}_{ij}\right|^3} - \frac{3(\vec{\mathrm{J}}_i \bullet \vec{r}_{ij})(\vec{\mathrm{J}}_j \bullet \vec{r}_{ij})}{\left|\vec{r}_{ij}\right|^5}$$

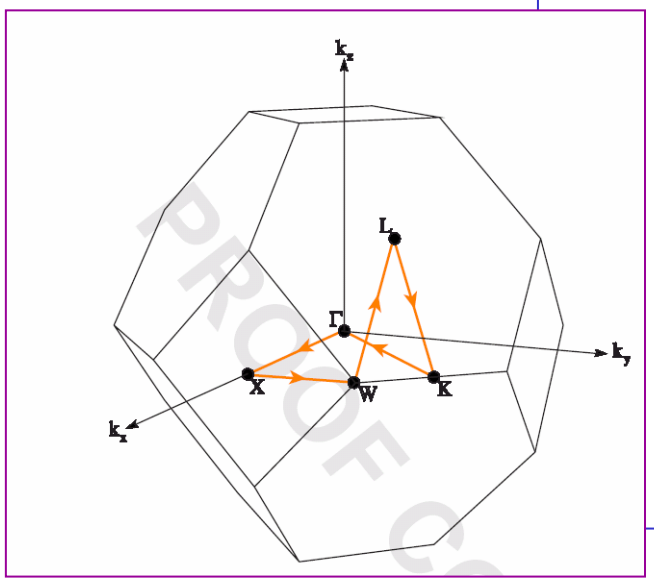
$$+\ V_{\mathrm{CF}}(\mathrm{J}_i^{\alpha})$$

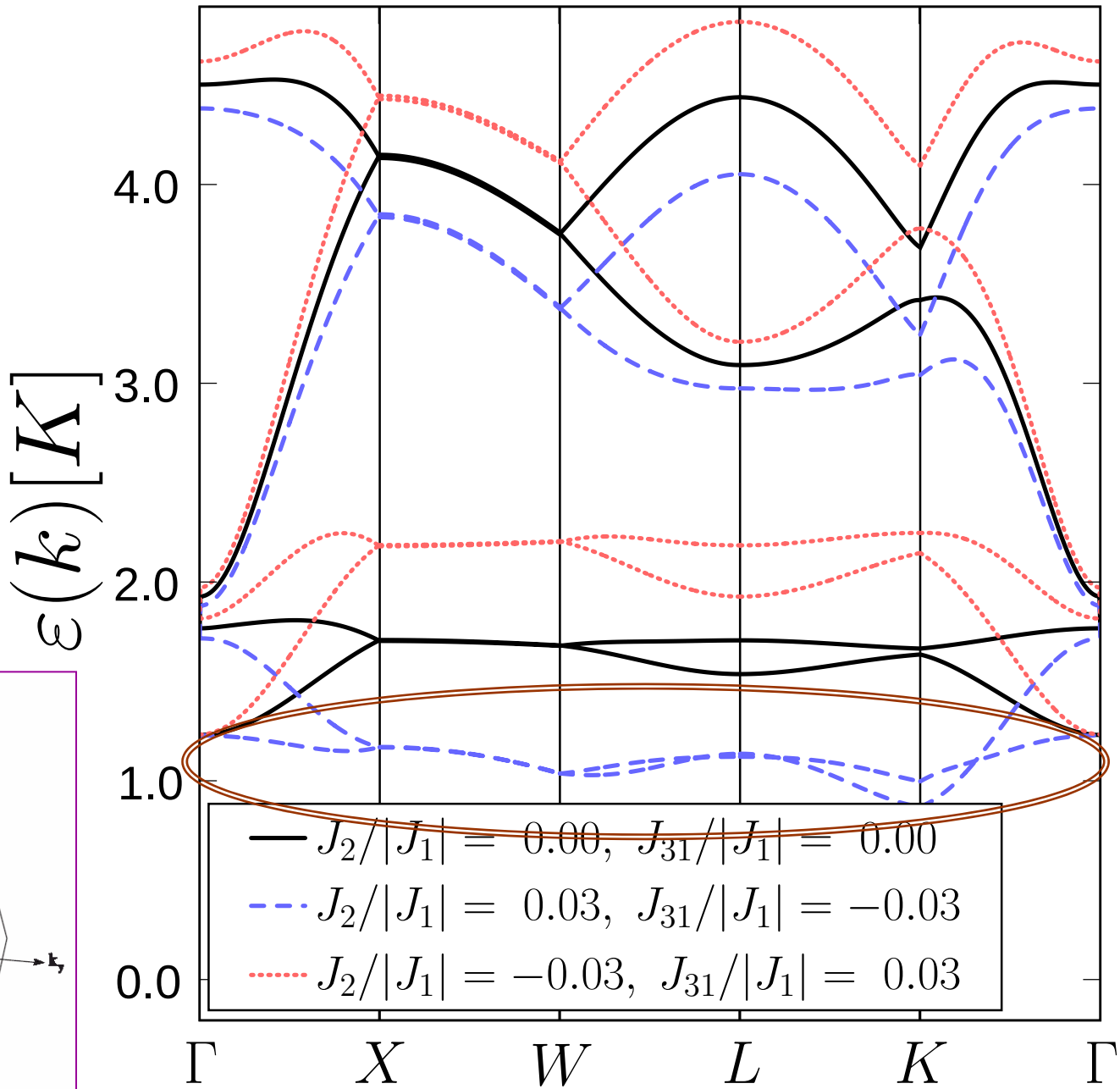
Crystal field part of *H*. This is a single-particle part of the Hamiltonian. it describes how the local electrostatic/chemical environment lifts the otherwise (2J+1) degeneracy of the otherwise free rare-earth ion.

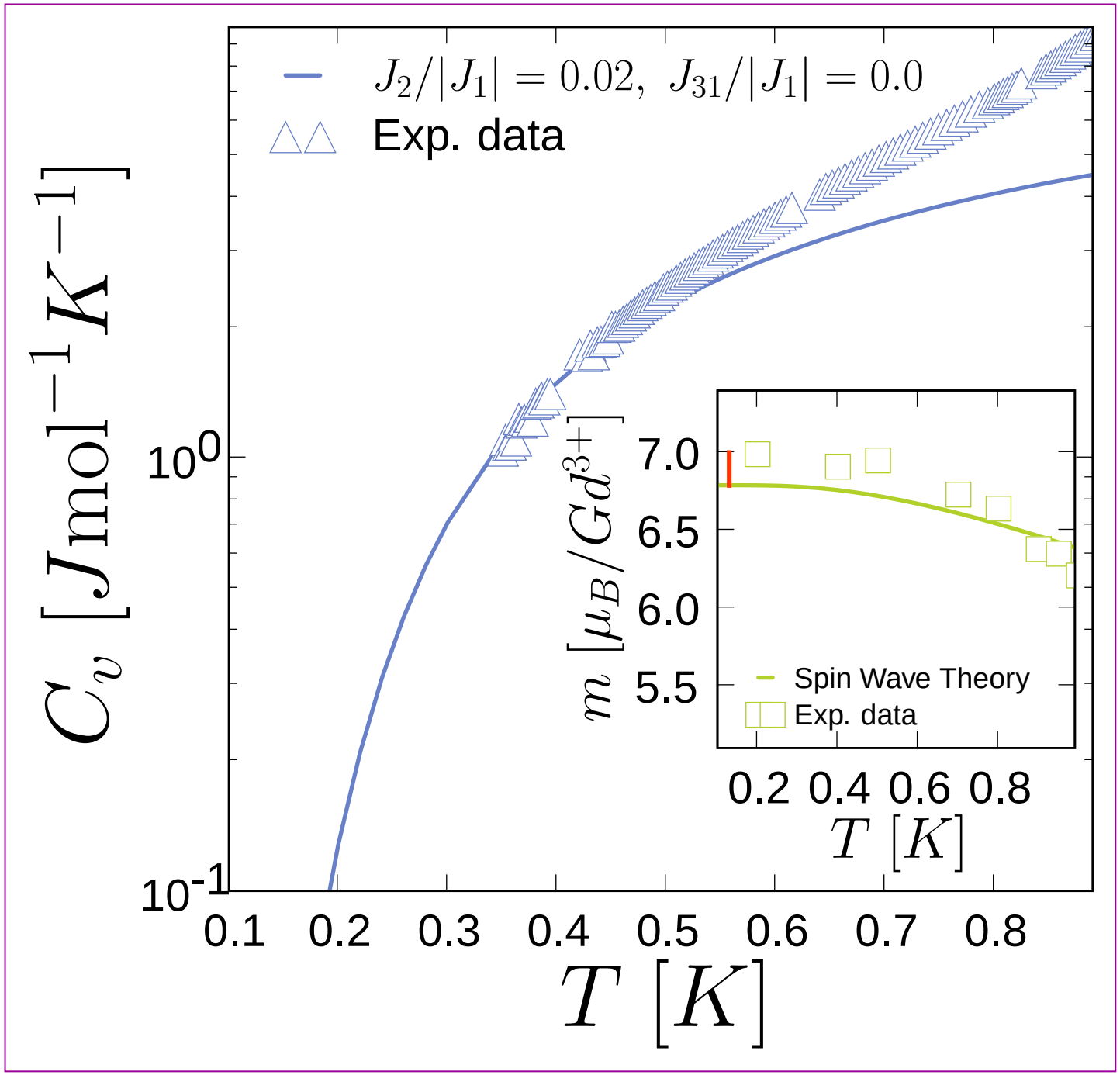
Knowing the classical ground state of H, one can do $1/S$ spin-wave expansion away from it. After some work … one can recast H as:

$$H = H_0 + \sum_{\vec{k}} \sum_{\alpha} \hbar\omega_\alpha(\vec{k}) \left[a_{\alpha,\vec{k}}^{\dagger} a_{\alpha,\vec{k}} + 1/2 \right]$$

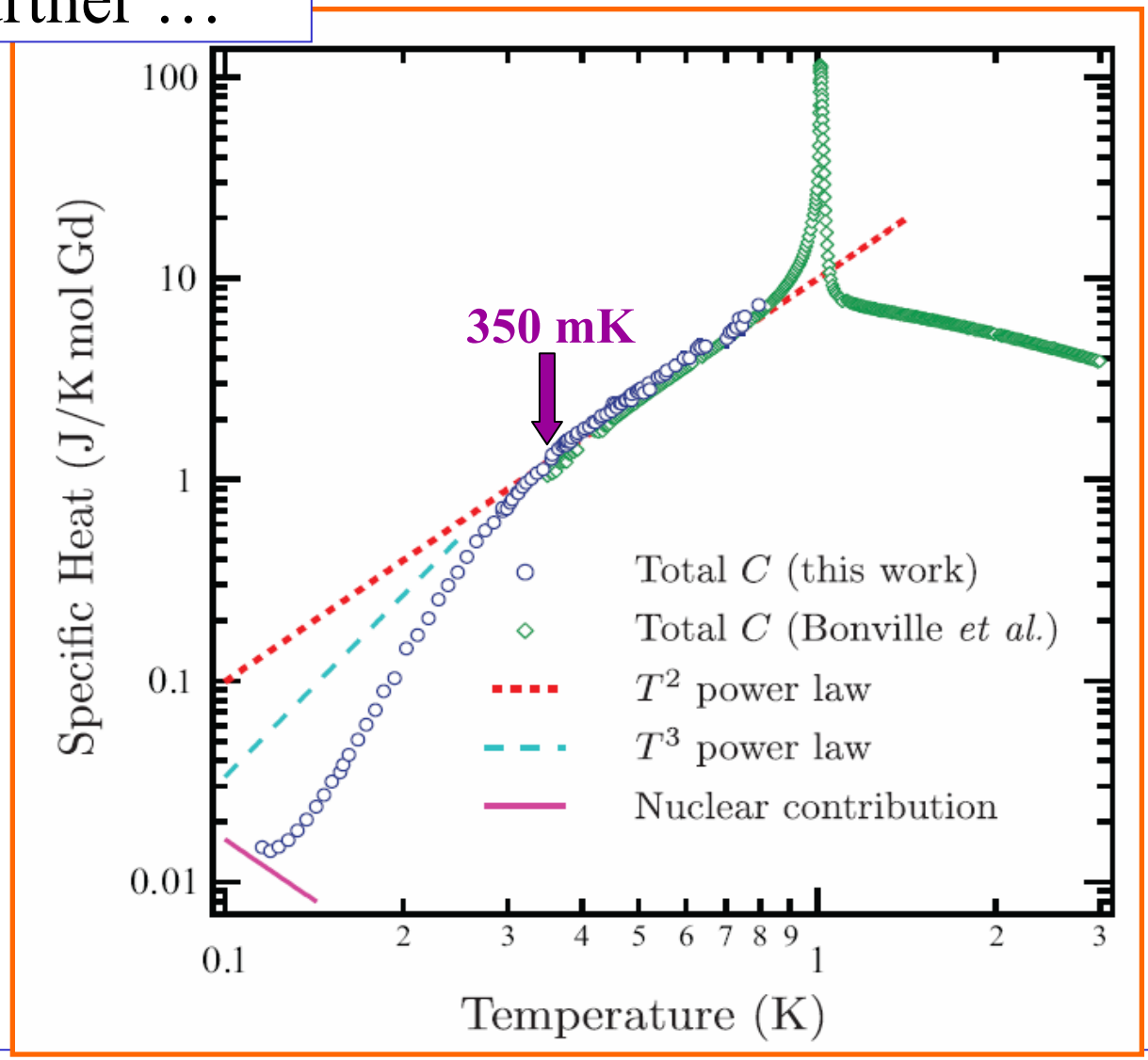

J_2
J_{32}
J_{31}
J_1


k_z
k_y
k_x
L
Γ
X
W
K


$\varepsilon(k)\,[K]$
4.0
3.0
2.0
1.0
0.0
Γ
X
W
L
K
Γ
$J_2/|J_1| = 0.00,\ J_{31}/|J_1| = 0.00$
$J_2/|J_1| = 0.03,\ J_{31}/|J_1| = -0.03$
$J_2/|J_1| = -0.03,\ J_{31}/|J_1| = 0.03$


$J_2/|J_1| = 0.02,\ J_{31}/|J_1| = 0.0$
Exp. data
$C_v\ [J\,\mathrm{mol}^{-1}K^{-1}]$
10^0
10^{-1}
0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8
$T\ [K]$
$m\ [\mu_B/Gd^{3+}]$
7.0
6.5
6.0
5.5
Spin Wave Theory
Exp. data
0.2 0.4 0.6 0.8
$T\ [K]$

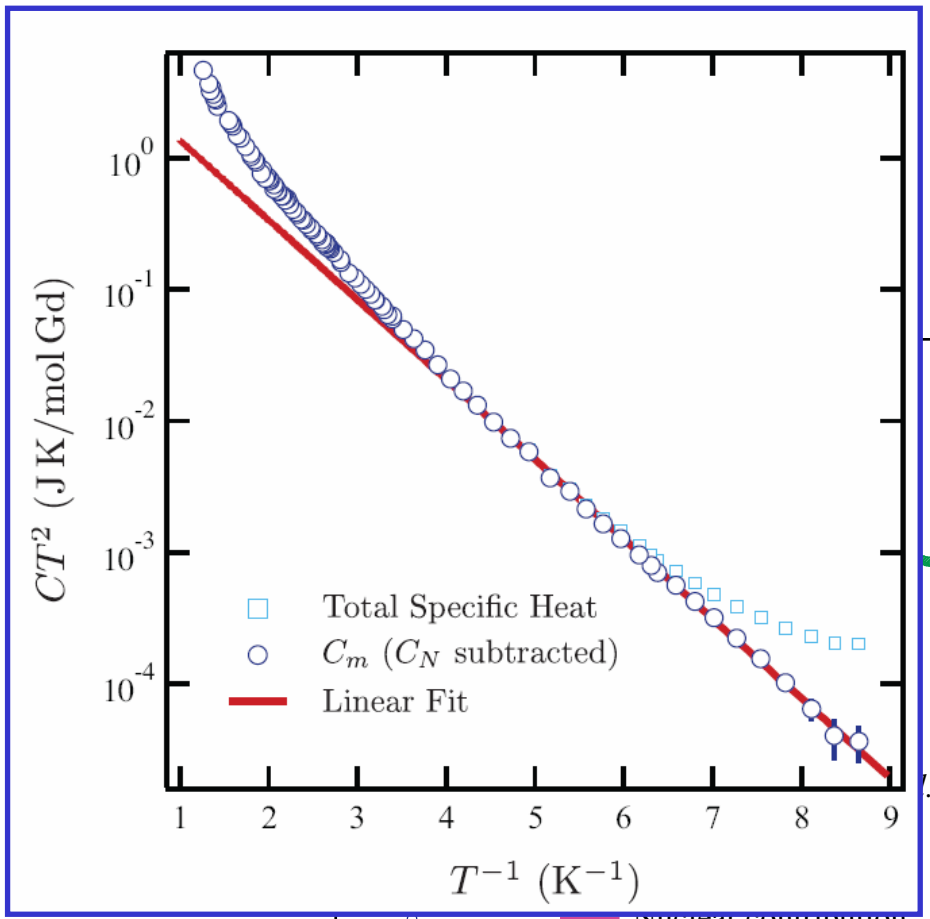
Pushing the study further …

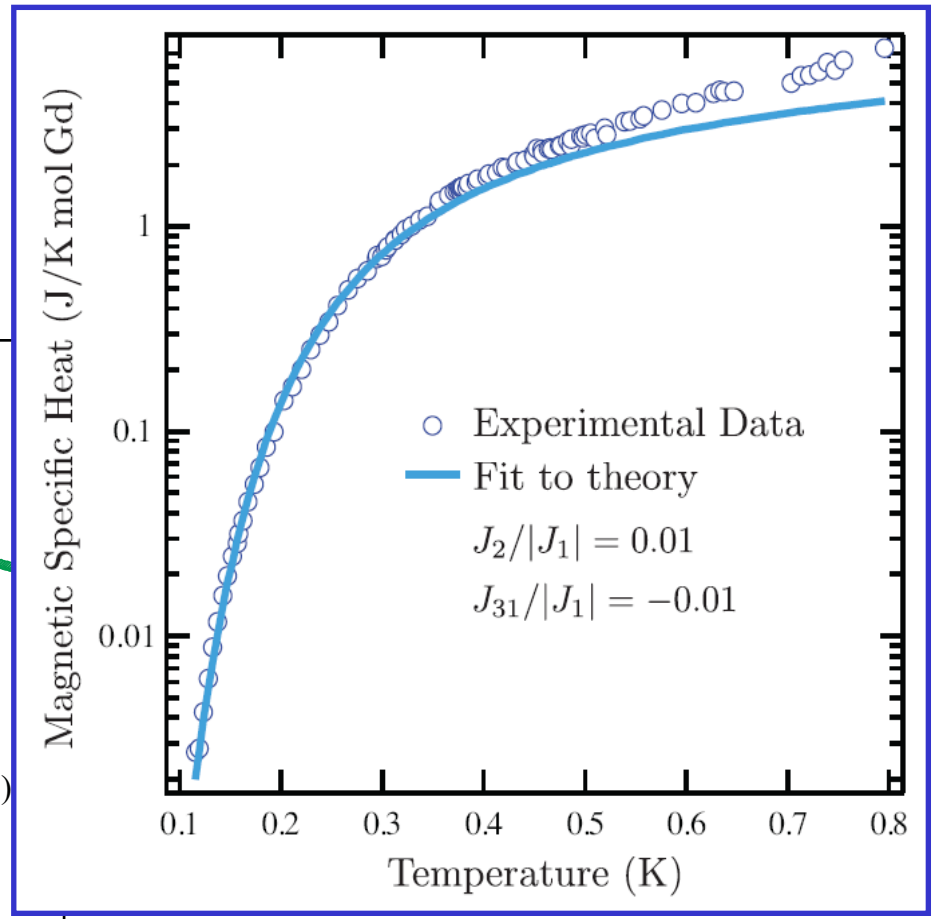


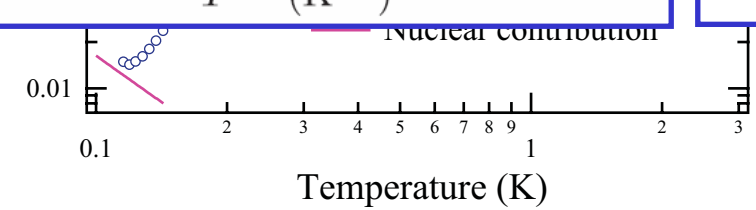
"Evidence for gapped spin-wave excitations in the frustrated $Gd_2Sn_2O_7$ pyrochlore antiferromagnet from low-temperature specific heat measurements"
J. A. Quilliam, K. A. Ross, A. G. Del Maestro, M. J. P. Gingras, L. R. Corruccini, and J. B. Kycia (Phys. Rev. Lett. **99** 097201 [2007]).

Gap-like behavior, $C \sim \exp(-\Delta/T)$

Fit to spin-wave theory







Conclusions as per $Gd_2Sn_2O_7$ pyrochlore

- Neutron seemingly "sees" long range order.
- That order is semi-classical (very weak quantum fluctuations ~ 3%).
- That order agrees with simplest theoretical expectations.
- Specific heat reveals gapped magnetic excitations below ~350 mK
- The magnetic excitations are spin-wave like.
- These quantitatively characterize the experimental specific heat.
- **Yet** muSR "sees" persistent spin dynamics down to the lowest temperature.
- What is it that gives rise to this persistent spin dynamics in this material?
- **Most/more interestingly**, what is the influence of this "cause" in $Gd_2Sn_2O_7$ on the observed PSD in other (rare-earth oxide) HFMs?

THE END