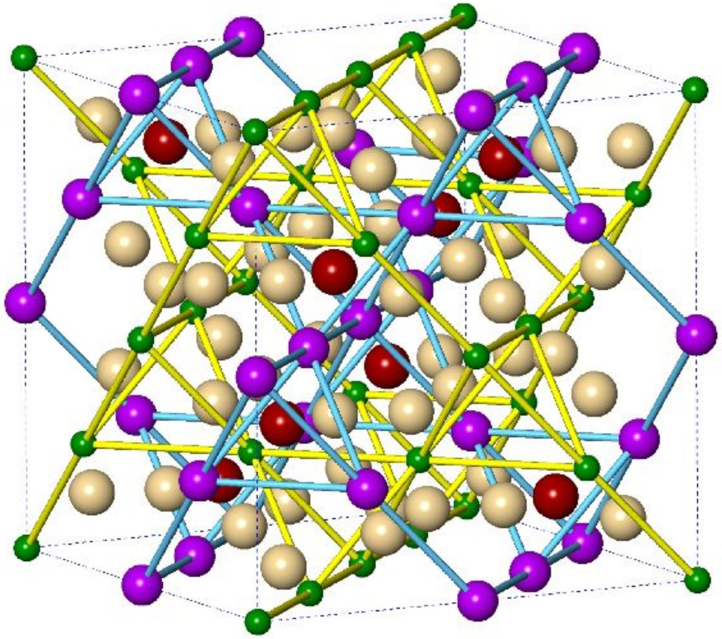
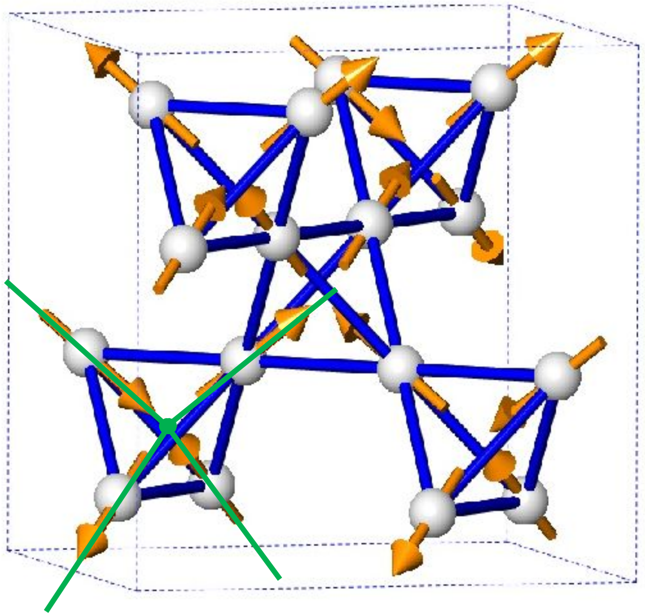
"Oxydes de Pyrochlore Magnétique: Fin de l'Esclavage du Modèle d'Ising"



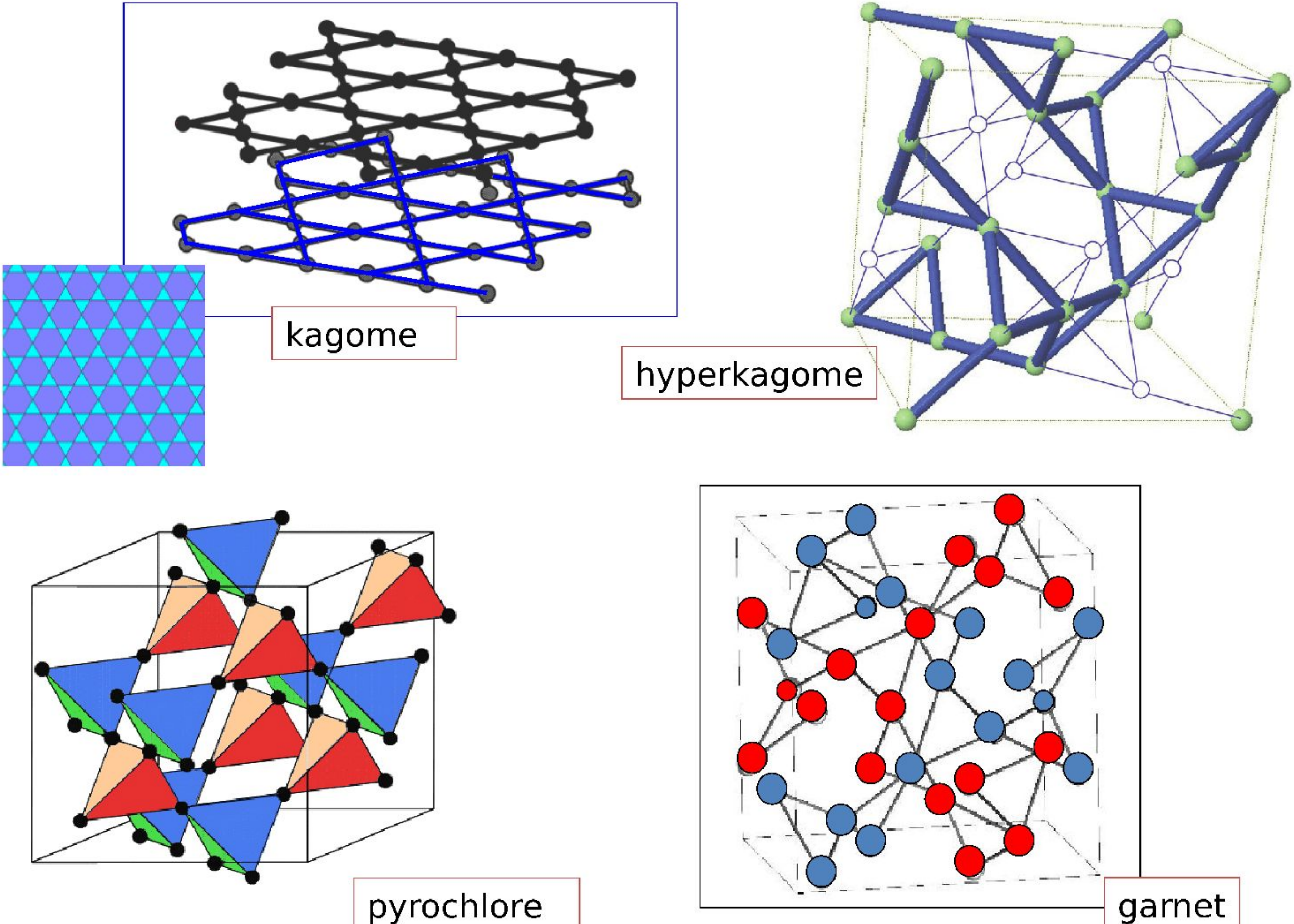


Michel Gingras

Department of Physics & Astronomy, University of Waterloo, Waterloo, Ontario, Canada

&

Canadian Institute for Advanced Research/Quantum Materials Program



kagome
hyperkagome
pyrochlore
garnet

What is the issue at stake? (I)

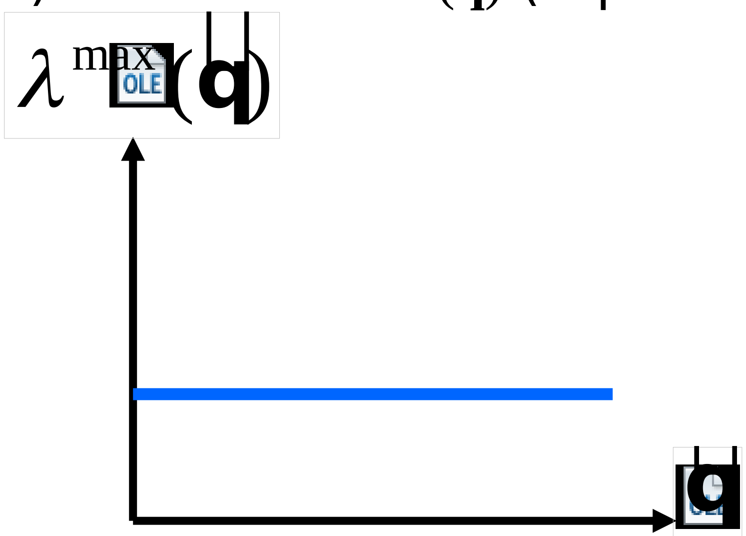
1) Mean-field: staggered susceptibility $\chi(\mathbf{q})$

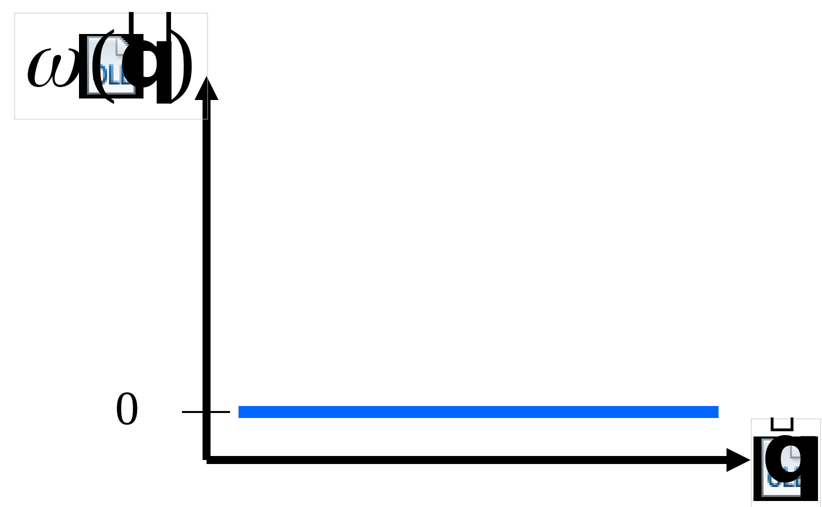
$$\chi(\mathbf{q}) \sim \sum_{\alpha} \frac{1}{1 - \lambda_{\alpha}(\mathbf{q})/3T}$$

- $\chi(\mathbf{q}_{ord})$ is a maximum.
- $\lambda_\alpha(\mathbf{q})$ maximum at $\mathbf{q}_{ord}$, $\lambda_{max}(\mathbf{q}_{ord})$.
- $T_c^{MF} = \lambda_{max}(\mathbf{q}_{ord})/3$.

→ $\lambda(\mathbf{q})$ ~ Fourier transform of the spin-spin interactions $J(\mathbf{q})$

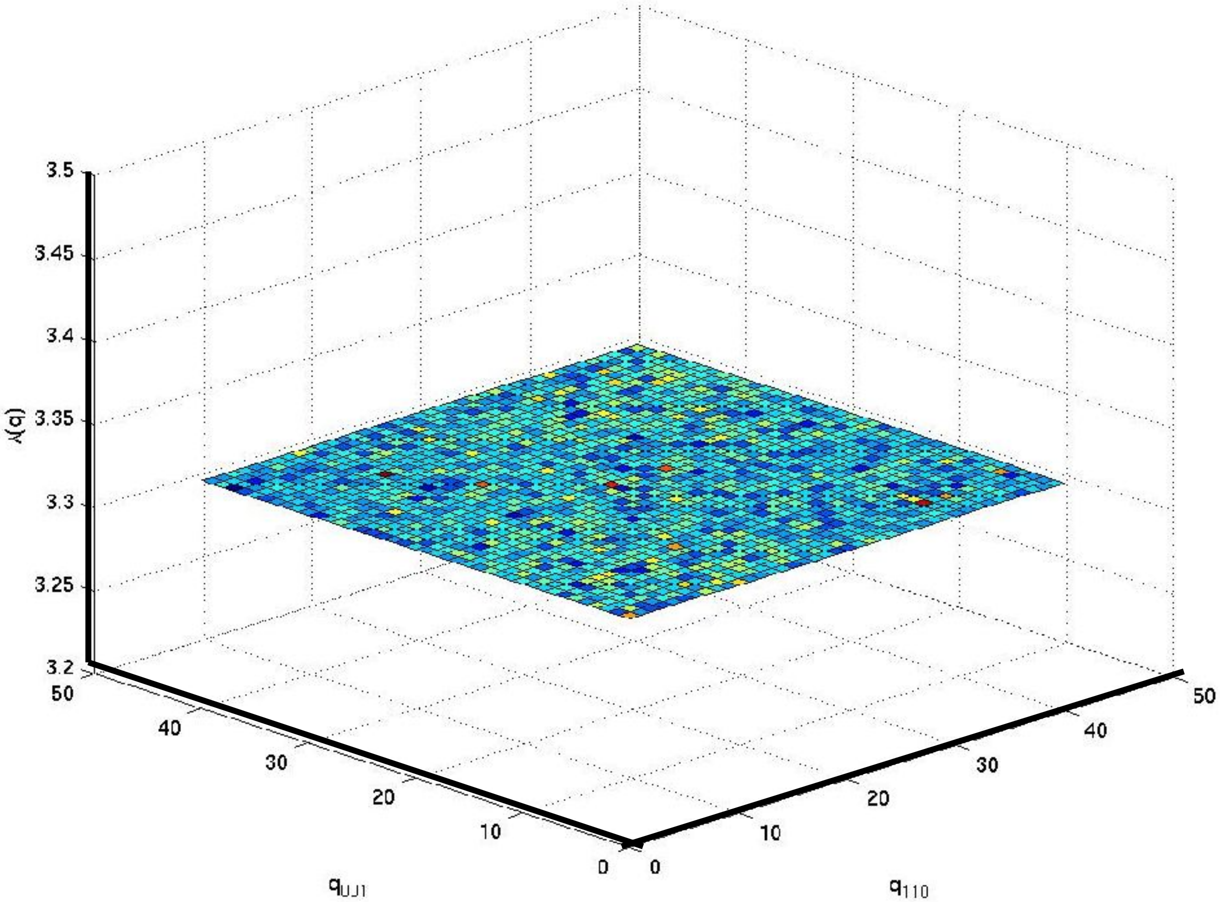
2) Excitations $\omega(\mathbf{q})$ (“spin waves”) around classical ground states





Example: Nearest-neighbor Ising AF on pyrochlore lattice (or, equivalently, n.n. spin ice)

1nn cut-off



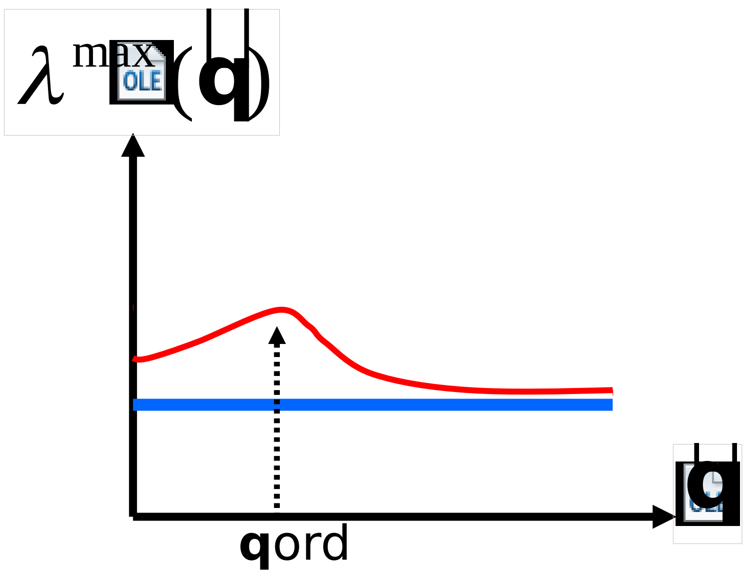
What is the issue at stake? (II)

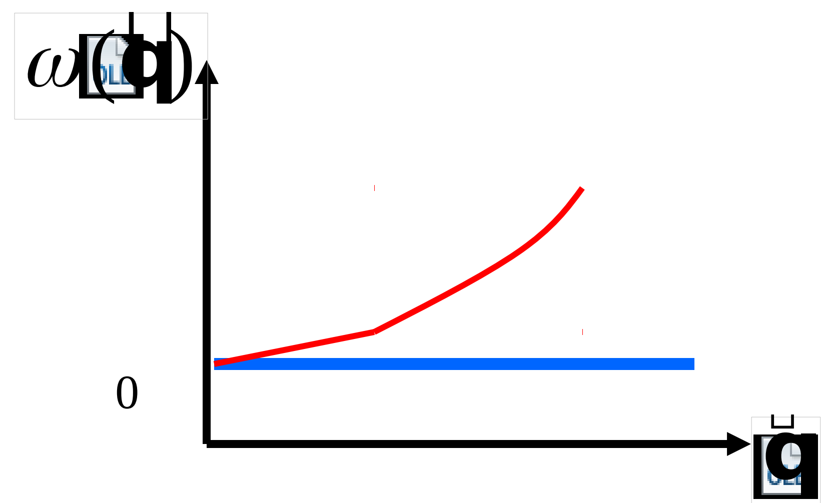
$$H = H_0 + H'$$

H':

- *J*ij(|**rij**|) beyond nearest neighbor
- single-ion anisotropy
- spin-phonon coupling
- random disorder
- long-range dipole-dipole interaction

 In highly frustrated magnets, 1/S fights against *H'*





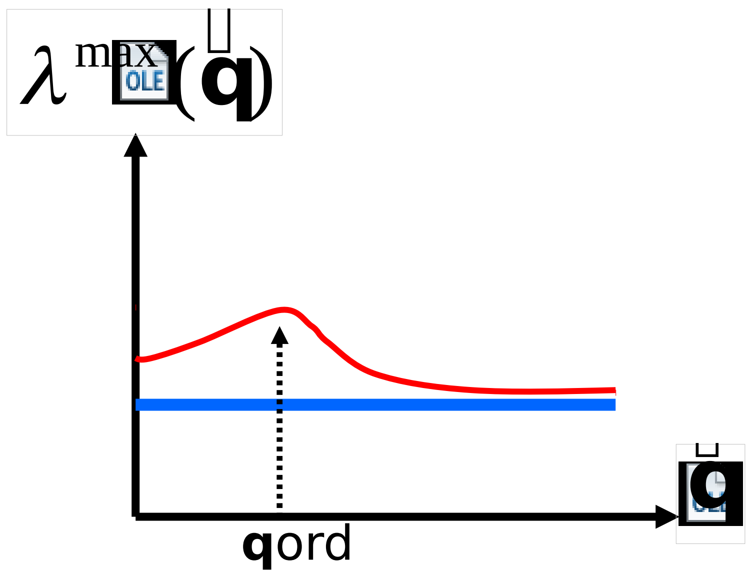
What is the issue at stake? (II)

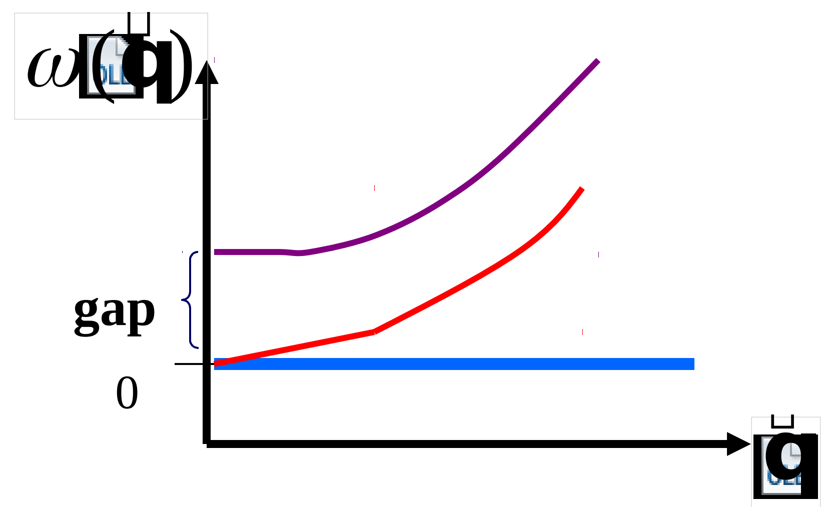
$$H = H_0 + H'$$

H':

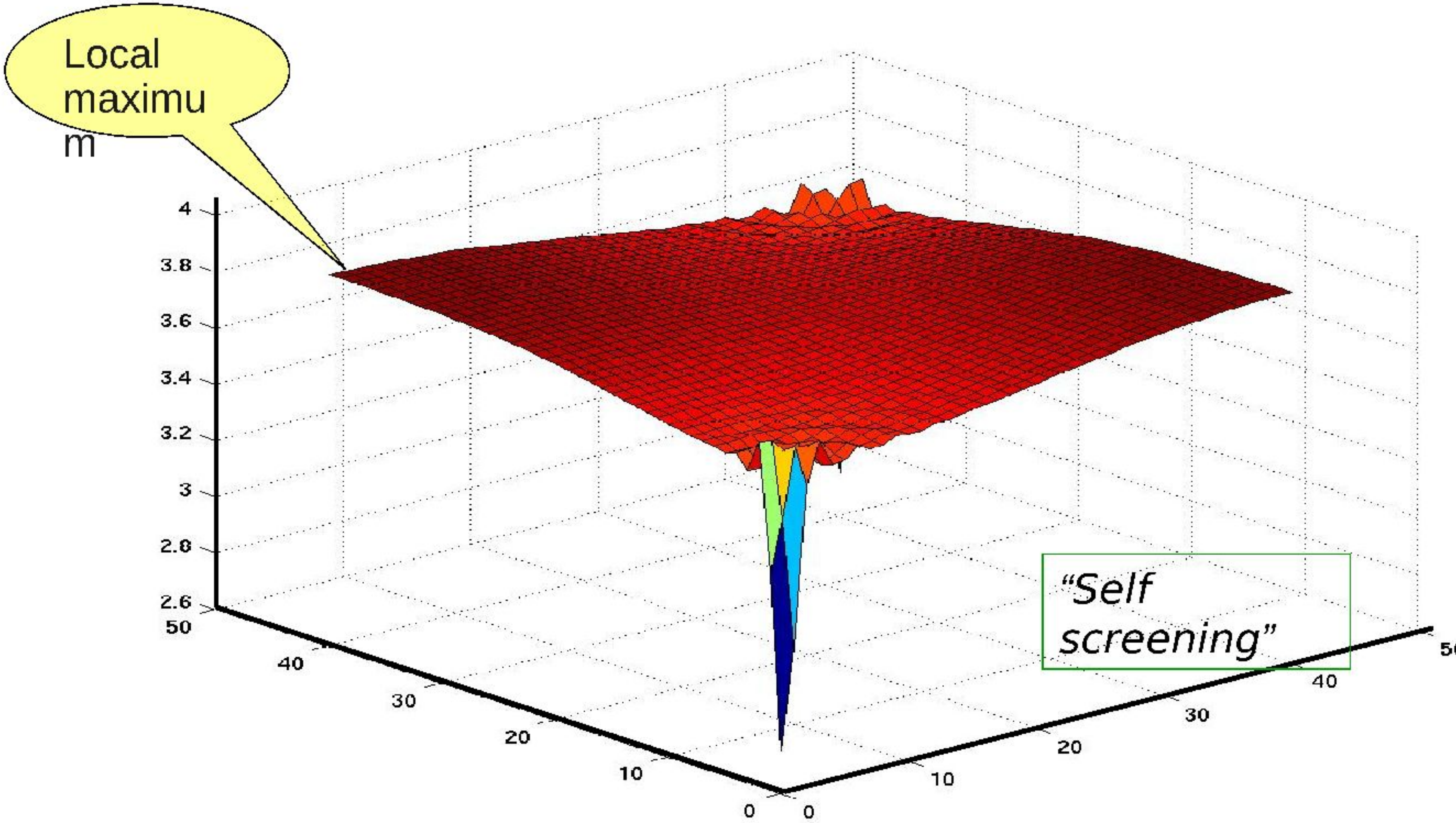
- Jij(|**rij**|) beyond nearest neighbor
- single-ion anisotropy
- spin-phonon coupling
- random disorder
- long-range dipole-dipole interaction

In highly frustrated magnets, 1/S fights against H'

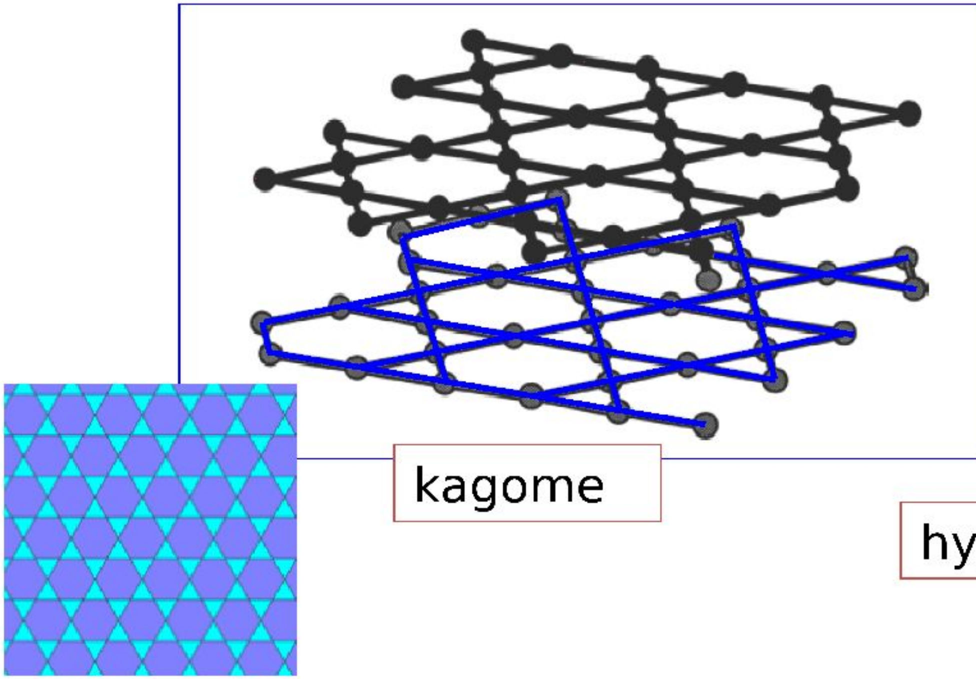




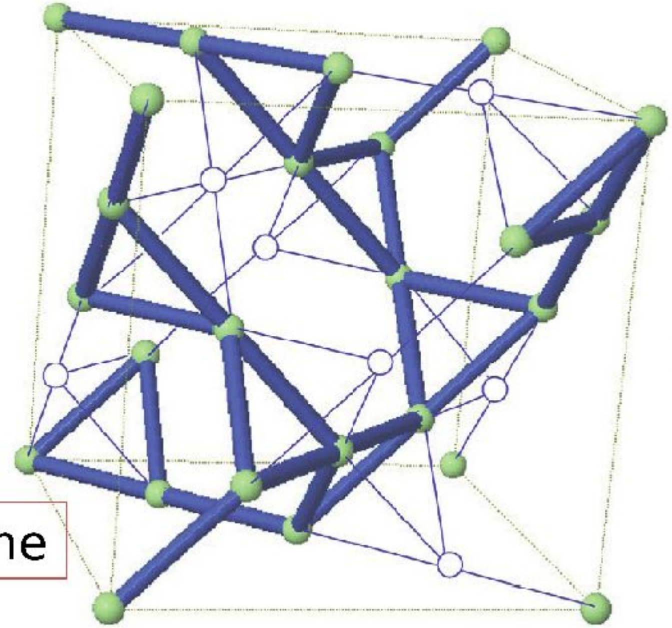
Example: Long-range dipolar spin ice



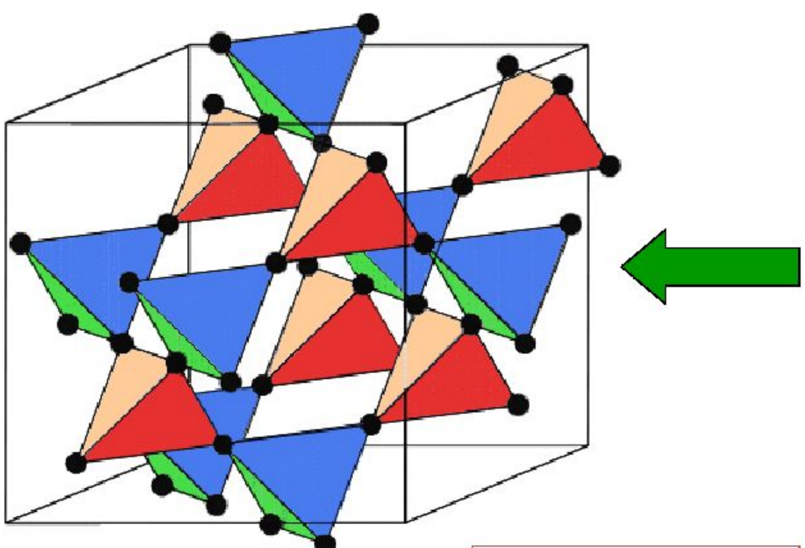
M.J.P. Gingras and B.C. den Hertog, Can. J. Phys. **79**, 1339 (2001)



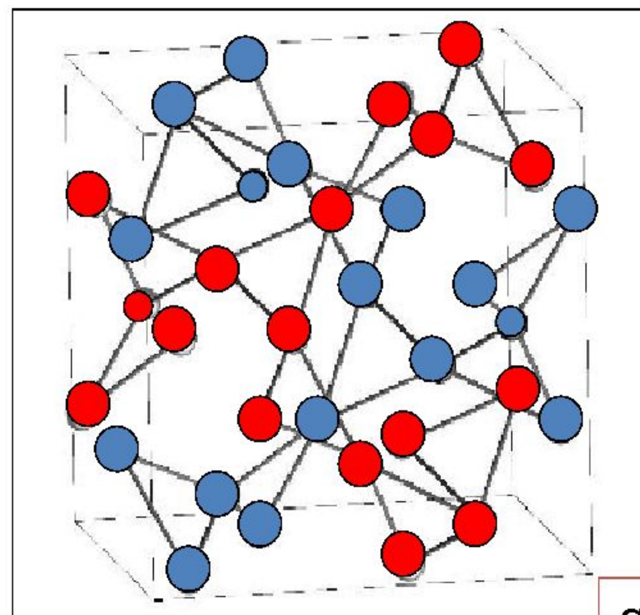
kagome



hyperkagome



pyrochlore



garnet

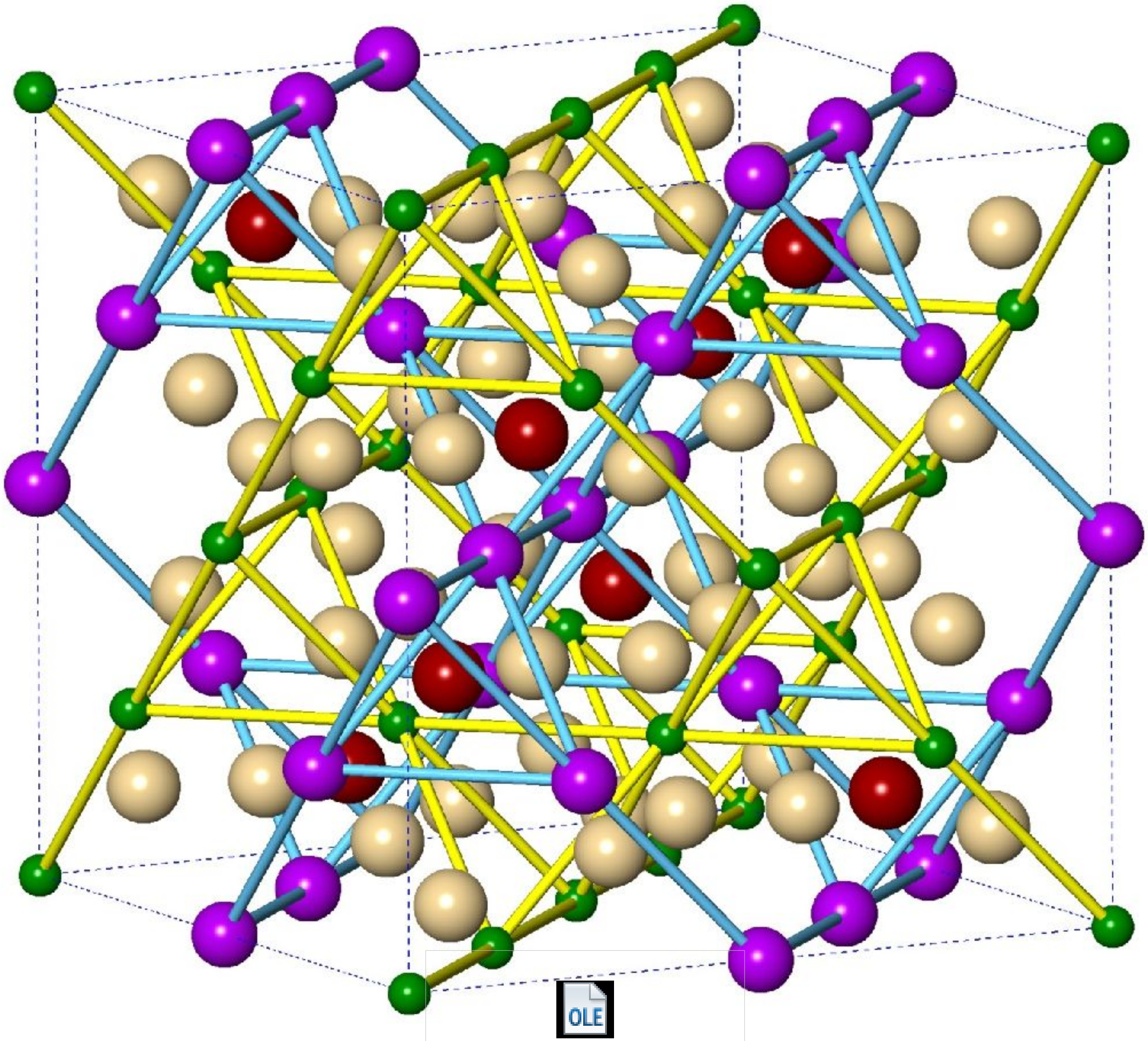
Outline

1. **Frustrated rare-earth pyrochlore oxides**
   - Hamiltonian, crystal field, effective Hamiltonian
2. **Examples of phenomena**
   - Ising – (Ho,Dy)2(Ti,Sn,Ge)2O7 : spin ice
   - Ising – Tb2Ti2O7 : spin ice/quadrup. fluct.
   - Heisenberg" – Gd2Ti2O7 : multiple transitions
   - XY AF – Er2Ti2O7 : order-by-disorder
   - XY FM – Yb2Ti2O7 : quantum spin ice (?)
3. **Conclusion**

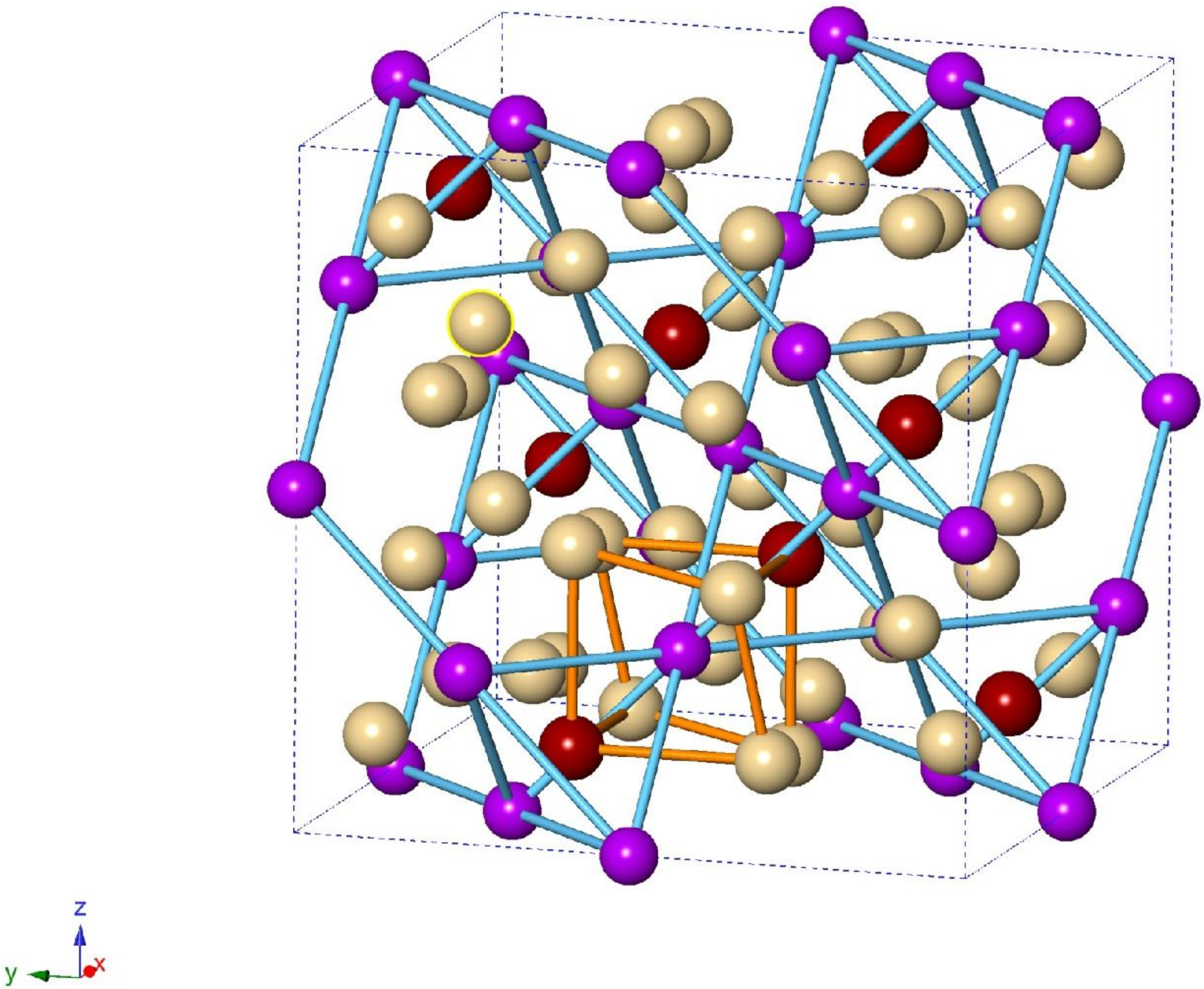
Smorgasbord of Phenomena

Magnetic pyrochlore oxides

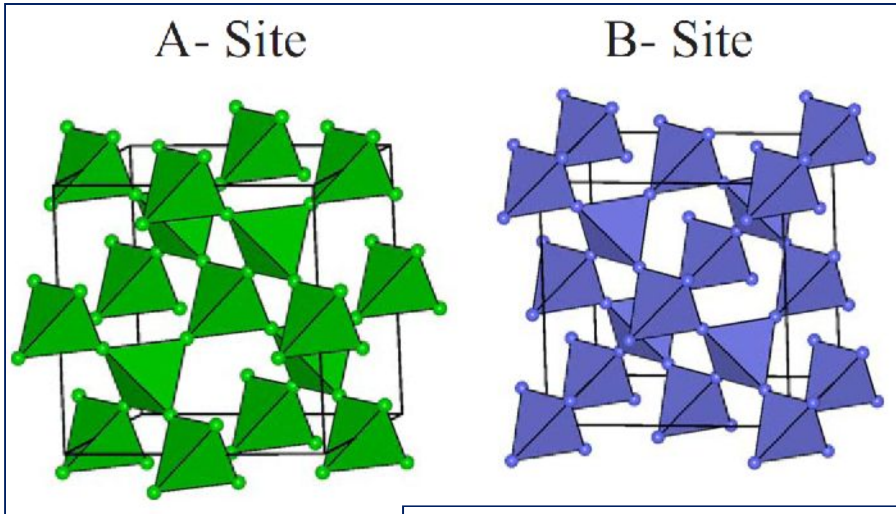
Jason S. Gardner, Michel J. P. Gingras, and John E. Greedan
Rev. Mod. Phys. **82**, 53 (2010).



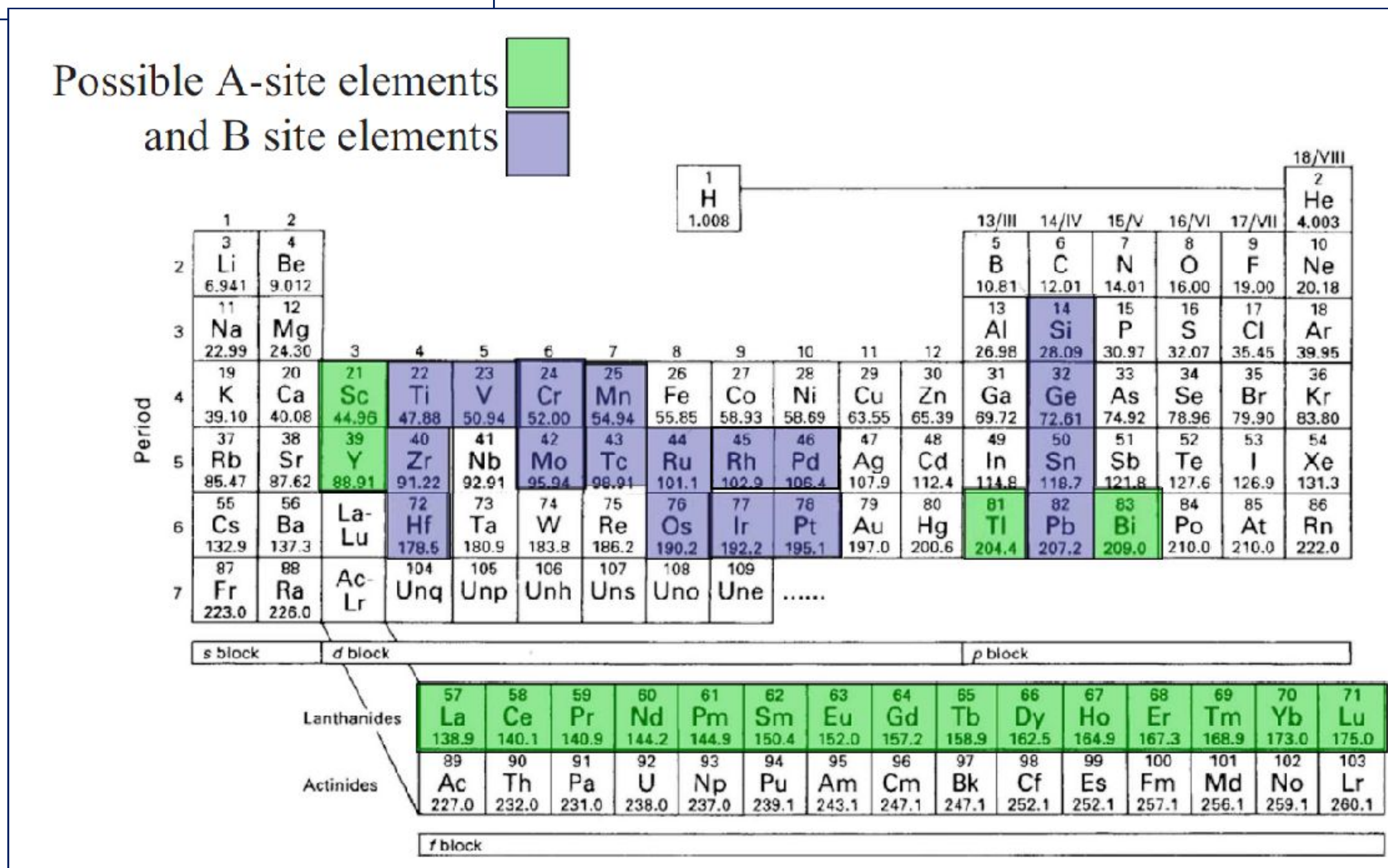
$a \sim 10$ Å

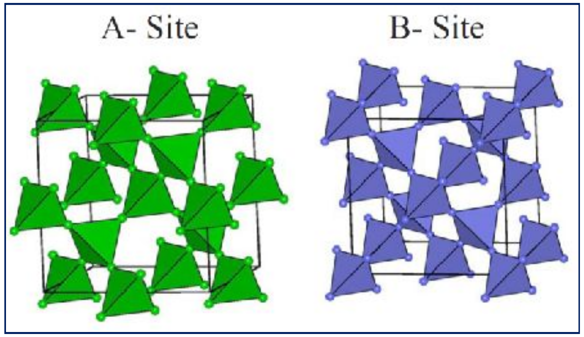


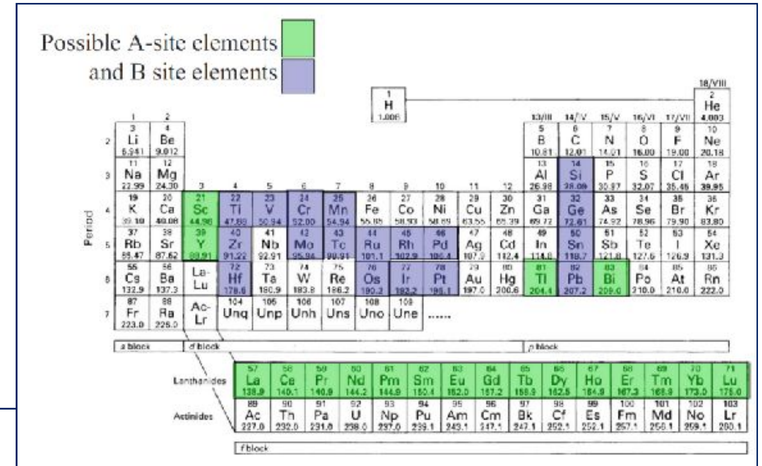
z
y
x

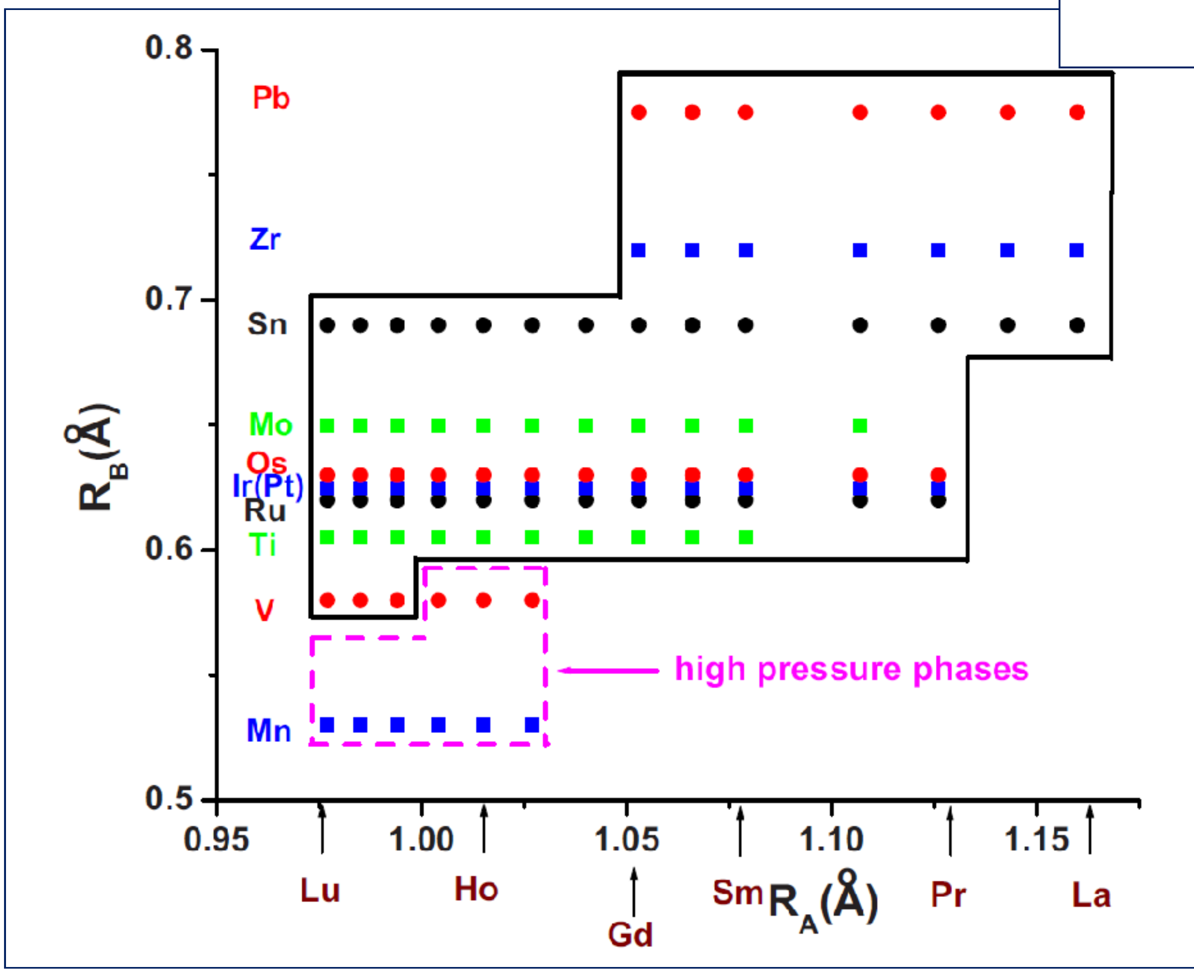

A- Site
B- Site

$A_2B_2O_7$

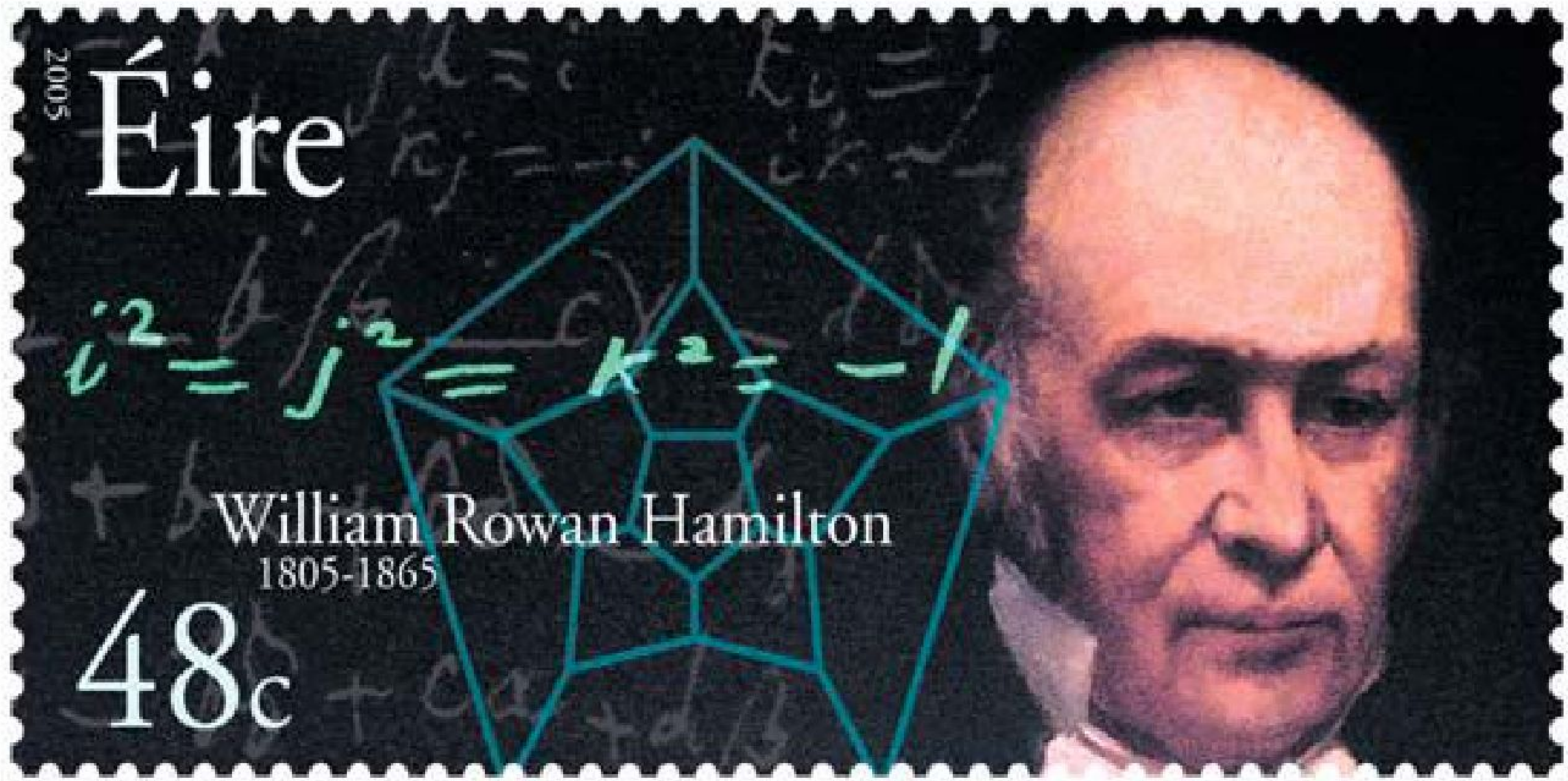

Possible A-site elements
and B site elements
Period
Lanthanides
Actinides
s block
d block
p block
f block


A- Site
B- Site


Possible A-site elements
and B site elements


0.8
0.7
0.6
0.5
R_B(Å)
Pb
Zr
Sn
Mo
Os
Ir(Pt)
Ru
Ti
V
Mn
high pressure phases
0.95
1.00
1.05
1.10
1.15
R_A(Å)
Lu
Ho
Gd
Sm
Pr
La

HAMILTONIAN … HAMILTONIAN … HAMILTONIAN



What are we talking about?

For rare-earth RE3+ ions one typically has:

$$H_{\text{Spin-orbit}} >> H_{\text{crystal-field}} >> H_{\text{interactions}}$$

$$\vec{J} = \vec{L} + \vec{S}$$

is a good quantum number

- Free (in vacuum) RE3+ ions have 2J+1 fold degenerate groundstate state
- “Environment” (crystal-field) lift that degeneracy

What are we talking about?

$$H = -\sum_{j>i} I_{ij}\, \vec{J}_i \cdot \vec{J}_j + \frac{\mu_0}{4\pi}(g\mu_B)^2 \sum_{j>i} \frac{\vec{J}_i \cdot \vec{J}_j}{|\vec{r}_{ij}|^3} - \frac{3(\vec{J}_i \cdot \vec{r}_{ij})(\vec{J}_j \cdot \vec{r}_{ij})}{|\vec{r}_{ij}|^5} + V_{CF}(J_i^{\alpha}) - g\mu_B \sum_i \vec{J}_i \cdot \vec{B}$$

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.

What are we talking about?

$$H = -\sum_{j>i} I_{ij}^{\alpha\beta} \mathrm{J}_i^{\alpha} \mathrm{J}_j^{\beta} + \frac{\mu_0}{4\pi}(g\mu_{\mathrm{B}})^2 \sum_{j>i} \frac{\vec{\mathrm{J}}_i \cdot \vec{\mathrm{J}}_j}{|\vec{r}_{ij}|^3} - \frac{3(\vec{\mathrm{J}}_i \cdot \vec{r}_{ij})(\vec{\mathrm{J}}_j \cdot \vec{r}_{ij})}{|\vec{r}_{ij}|^5} + V_{\mathrm{CF}}(\mathrm{J}_i^{\alpha}) - g\mu_{\mathrm{B}} \sum_i \vec{\mathrm{J}}_i \cdot \vec{\mathrm{B}}$$

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.

What are we talking about?

$$H = \sum_{i>j} \sum_{K,K'} \sum_{Q,Q'} I_{KK'}^{QQ'} O_K^Q(\mathrm{J}_i) O_{K'}^{Q'}(\mathrm{J}_j)$$

$$\text{where } K \leq 2\mathrm{J}_i,\; K' \leq 2\mathrm{J}_j;$$

$$Q = [-K, K];\; Q' = [-K', K']$$

Constraints on the $I_{KK'}^{QQ'}$ coefficients are imposed by symmetry.

Moment	Symmetry	Operator
Dipoles	Γ_4	J_x
		J_y
		J_z
Quadrupoles	Γ_3	$O_{3z^2-r^2}=3J_z^2-J(J+1)\equiv\hat{O}_2^0$
		$O_{x^2-y^2}=J_x^2-J_y^2\equiv\hat{O}_2^2$
	Γ_5	$O_{xy}=\overline{J_xJ_y}/2\equiv\hat{O}_2^{-2}$
		$O_{yz}=\overline{J_yJ_z}/2\equiv\hat{O}_2^{-1}$
		$O_{zx}=\overline{J_zJ_x}/2\equiv\hat{O}_2^{1}$
Octupoles	Γ_2	$T_{xyz}=(\sqrt{15}/6)\overline{J_xJ_yJ_z}$
	Γ_4	$T_x^\alpha=J_x^3-(\overline{J_xJ_y^2}+\overline{J_z^2J_x})/2$
		$T_y^\alpha=J_y^3-(\overline{J_yJ_z^2}+\overline{J_x^2J_y})/2$
		$T_z^\alpha=J_z^3-(\overline{J_zJ_x^2}+\overline{J_y^2J_z})/2$
	Γ_5	$T_x^\beta=\sqrt{15}(\overline{J_xJ_y^2}-\overline{J_z^2J_x})/6$
		$T_y^\beta=\sqrt{15}(\overline{J_yJ_z^2}-\overline{J_x^2J_y})/6$
		$T_z^\beta=\sqrt{15}(\overline{J_zJ_x^2}-\overline{J_y^2J_z})/6$

TABLE I. Operator equivalents describing active multipoles within a cubic Γ_8 quartet. Bars over symbols indicate the sum with respect to all the possible permutations of the indices, e.g., $\overline{J_xJ_y^2}=J_xJ_y^2+J_yJ_xJ_y+J_y^2J_x$. The five $\hat{O}_2^Q$ are the usual Stevens' operator equivalents (see below). Adapted from Shiina *et al.* (1998).

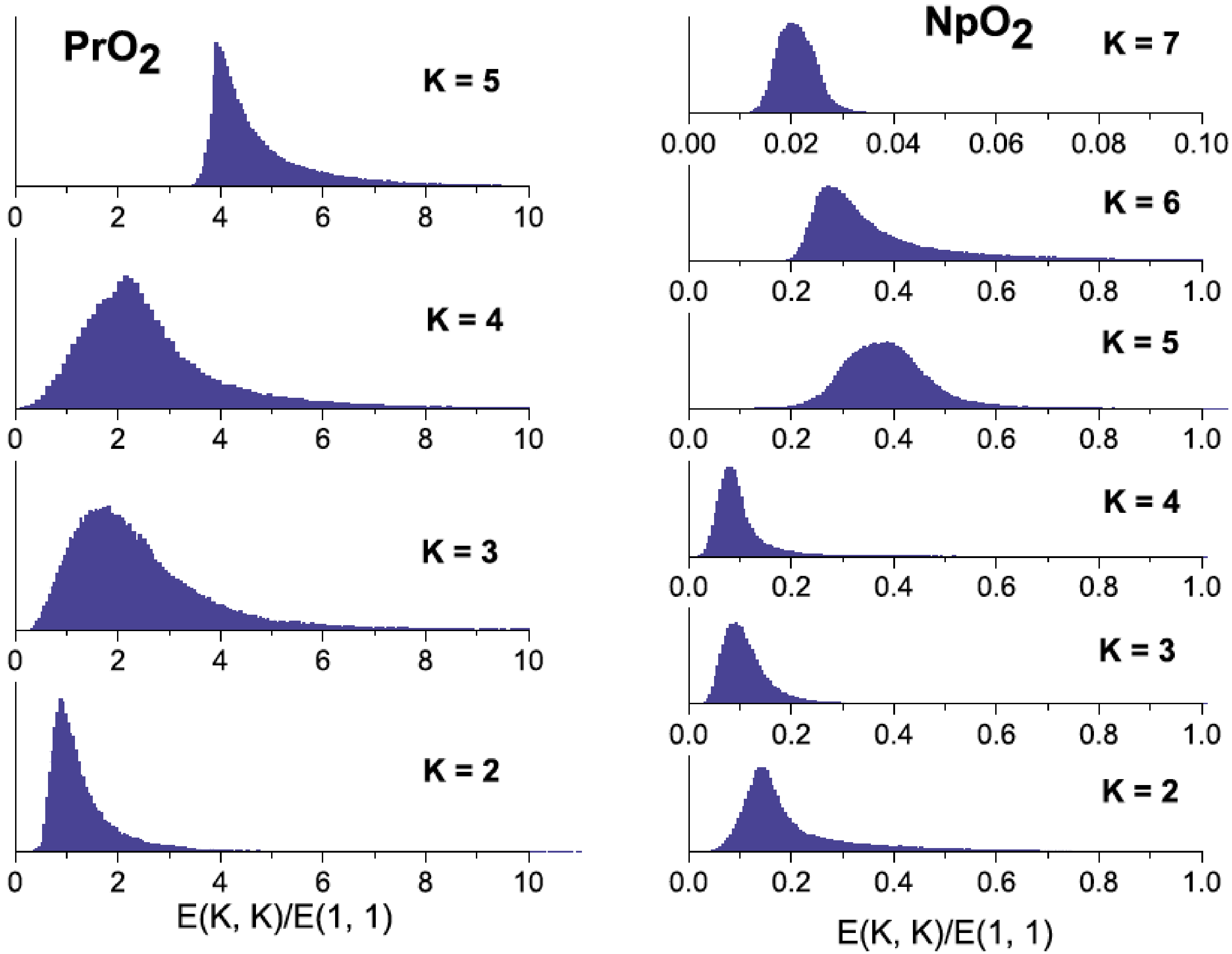


FIG. 2. (Color online) Distribution of the ratio $E(K,K)/E(1,1)$, with $E(K_i,K_j)$ the ground-state energy of a dimer (C_{2v} bond) of Pr^{4+} and Np^{4+} ions coupled with the part of Eq. (35) associated with a specific pair of ranks K_i, K_j.

What are we talking about?

$$H = -\sum_{j>i} I_{ij}^{\alpha\beta} \, J_i^{\alpha} J_j^{\beta} + \frac{\mu_0}{4\pi}(g\mu_{\mathrm{B}})^2 \sum_{j>i} \frac{\vec{J}_i \cdot \vec{J}_j}{|\vec{r}_{ij}|^3} - \frac{3(\vec{J}_i \cdot \vec{r}_{ij})(\vec{J}_j \cdot \vec{r}_{ij})}{|\vec{r}_{ij}|^5} + V_{\mathrm{CF}}(J_i^{\alpha}) - g\mu_{\mathrm{B}} \sum_i \vec{J}_i \cdot \vec{B}$$

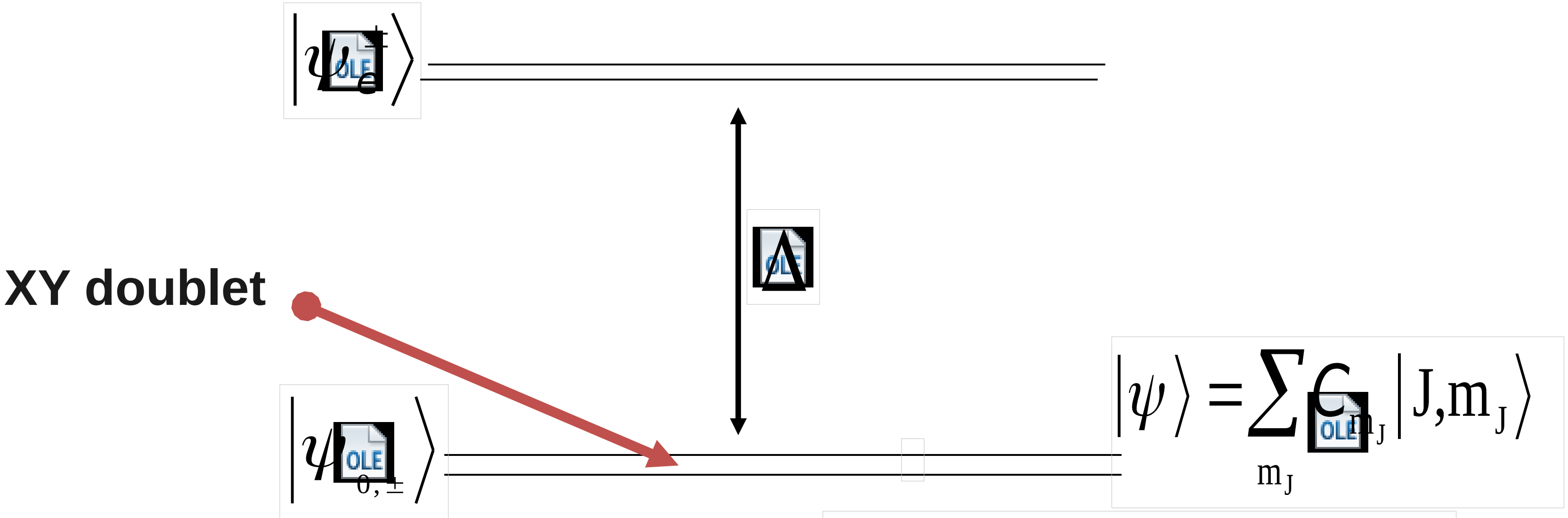
Crystal field part of *H*. This is a single-particle part of the Hamiltonian. it describes how the local electrostatic/chemical environment lifts the otherwise (2J+1) degeneracy of the otherwise free rare-earth ion.

What are we talking about?

$$H = -\sum_{j>i} I_{ij}^{\alpha\beta}\, J_i^{\alpha} J_j^{\beta} + \frac{\mu_0}{4\pi}(g\mu_B)^2 \sum_{j>i}\left[\frac{\vec{J}_i \cdot \vec{J}_j}{|\vec{r}_{ij}|^3} - \frac{3(\vec{J}_i \cdot \vec{r}_{ij})(\vec{J}_j \cdot \vec{r}_{ij})}{|\vec{r}_{ij}|^5}\right] + V_{CF}(J_i^{\alpha}) - g\mu_B \sum_i \vec{J}_i \cdot \vec{B}$$

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.

Crystal field effects in rare-earth pyrochlore oxides



$$\langle \psi_{0,+} | J_i^{\pm} | \psi_{0,-} \rangle = 0 \qquad \langle \psi_{0,\pm} | J_i^{z} | \psi_{0,\pm} \rangle \neq 0$$

$Dy_2Ti_2O_7$ & $Dy_2Sn_2O_7$: $\Delta \approx 320$ K spin ice (s.i.)
$Ho_2Ti_2O_7$ & $Ho_2Sn_2O_7$: $\Delta \approx 280$ K spin ice (s.i.)
$Tb_2Ti_2O_7$ & $Tb_2Sn_2O_7$: $\Delta \approx$ 20 K "soft Ising" ??

Quantum spin ice: Molavian *et al.*: Phys. Rev. Lett. **98**, 157204 (2007); arXiv: 0912.2957;
J. Phys.: Condens. Matter **21**, 172201 (2009).

A comment in anticipation …

$$H = \sum_{i>j} \sum_{K,K'} \sum_{Q,Q'} I^{QQ'}_{KK'} O^{Q}_{K}(\mathrm{J}_i) O^{Q'}_{K'}(\mathrm{J}_j)$$

$$\text{where } K \le 2\mathrm{J}_i,\ K' \le 2\mathrm{J}_j;$$

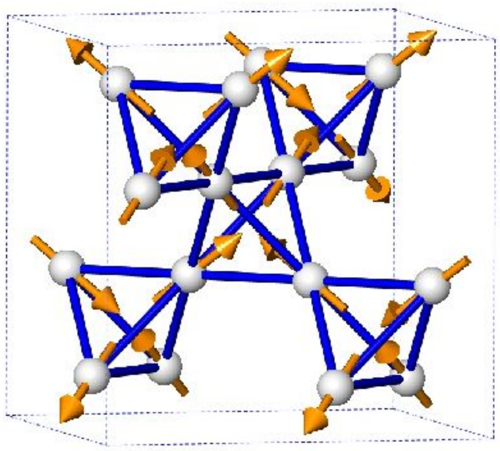
$$Q = [-K, K];\ Q' = [-K', K']$$

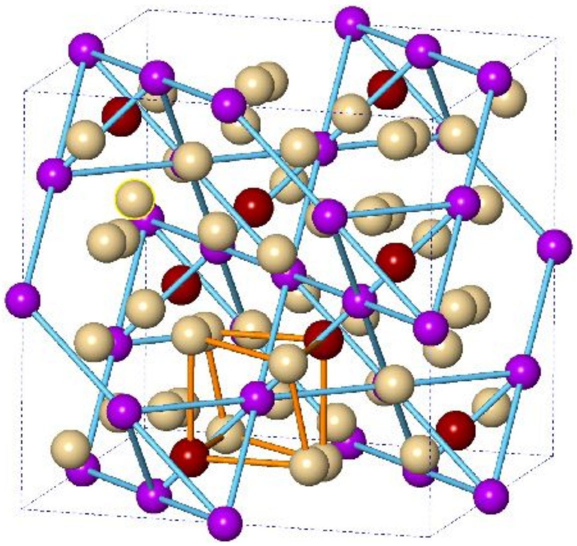
Constraints on the $I^{QQ'}_{KK'}$ coefficients are imposed by symmetry.

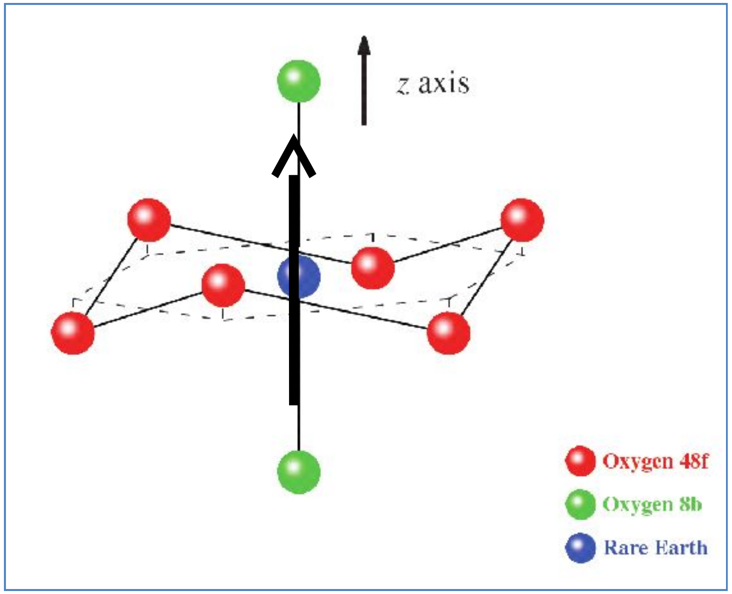
<u>THE POINT IS</u>:

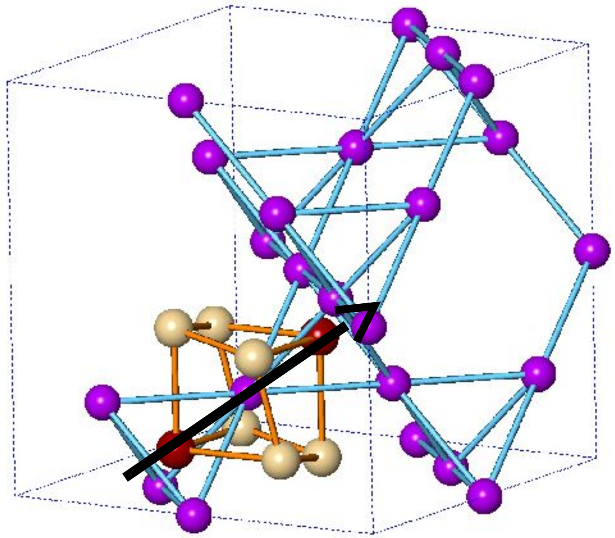
- Quantum fluctuations are “no longer” (not) forbidden as soon as multipolar interactions are considered, even in “classical” Dy2Ti2O7 and Ho2Ti2O7
- This point has been emphasized for Pr2(Sn,Zr)2O7 by Onoda and Tanaka [Phys. Rev. B **83**, 094411 (2011)]

Ho2Ti2O7 & Dy2Ti2O7

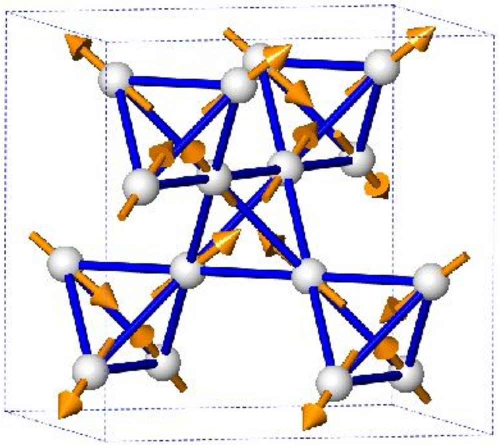


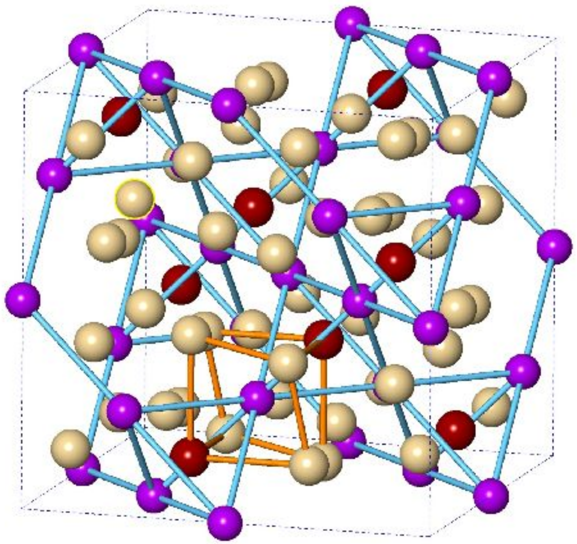


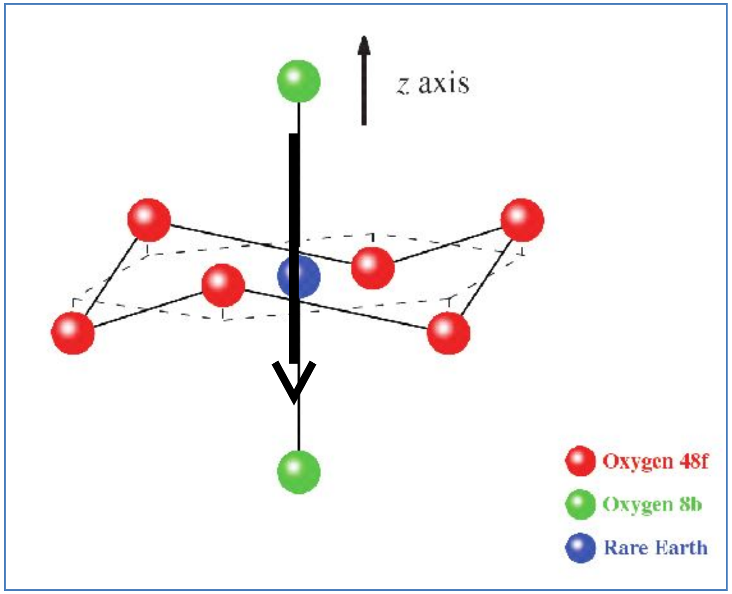


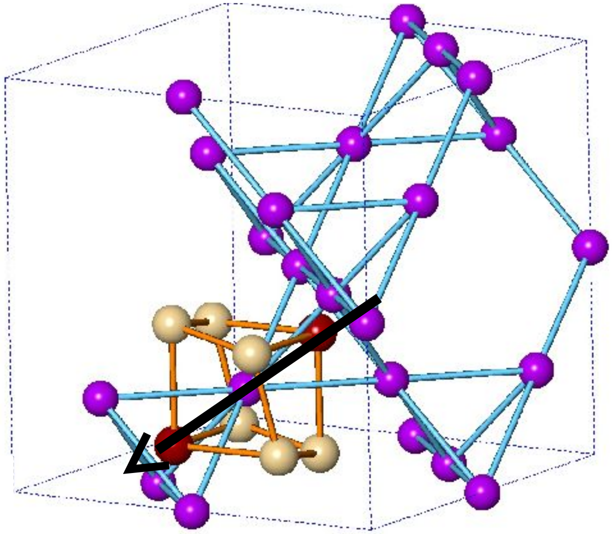


Ho2Ti2O7 & Dy2Ti2O7

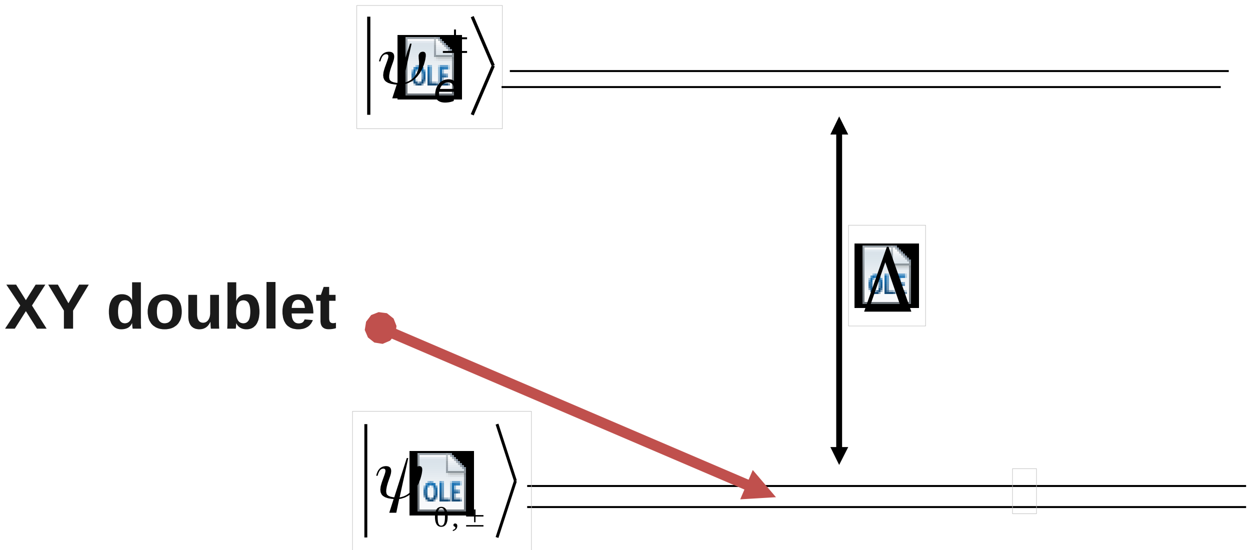








Crystal field effects in rare-earth pyrochlore oxides



$$|\psi\rangle = \sum_{m_J} C_{m_J} |J, m_J\rangle$$

$$\langle \psi_{0,+} | J_i^{x,y} | \psi_{0,-} \rangle = g_{\perp}$$

$$\langle \psi_{0,\pm} | J_i^{z} | \psi_{0,\pm} \rangle = g_{z}$$

$Er_2Ti_2O_7$ & $Er_2Sn_2O_7$: $\Delta \approx$ 80 K; AF XY
$Yb_2Ti_2O_7$ & $Yb_2Sn_2O_7$: $\Delta \approx$ 620 K; FM XY

Projection … Projection … Projection

$|m_J\rangle$ wavefunction decomposition

Dy2Ti2O7

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

~ 350 K

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

Ho2Ti2O7

Ho^{3+} (J=8)

~ 280 K

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

Tb2Ti2O7

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)

Effective Hamiltonian Method

$$H = H_{cef} + H_{int}$$

$$H_{int} = \sum_{i>j} \sum_{K,K'} \sum_{Q,Q'} I^{QQ'}_{KK'} O^{Q}_{K}(J_i) O^{Q'}_{K'}(J_j)$$

$$H_{eff} = P H_{int} P + P H_{int} Q H_{int} P + \cdots$$

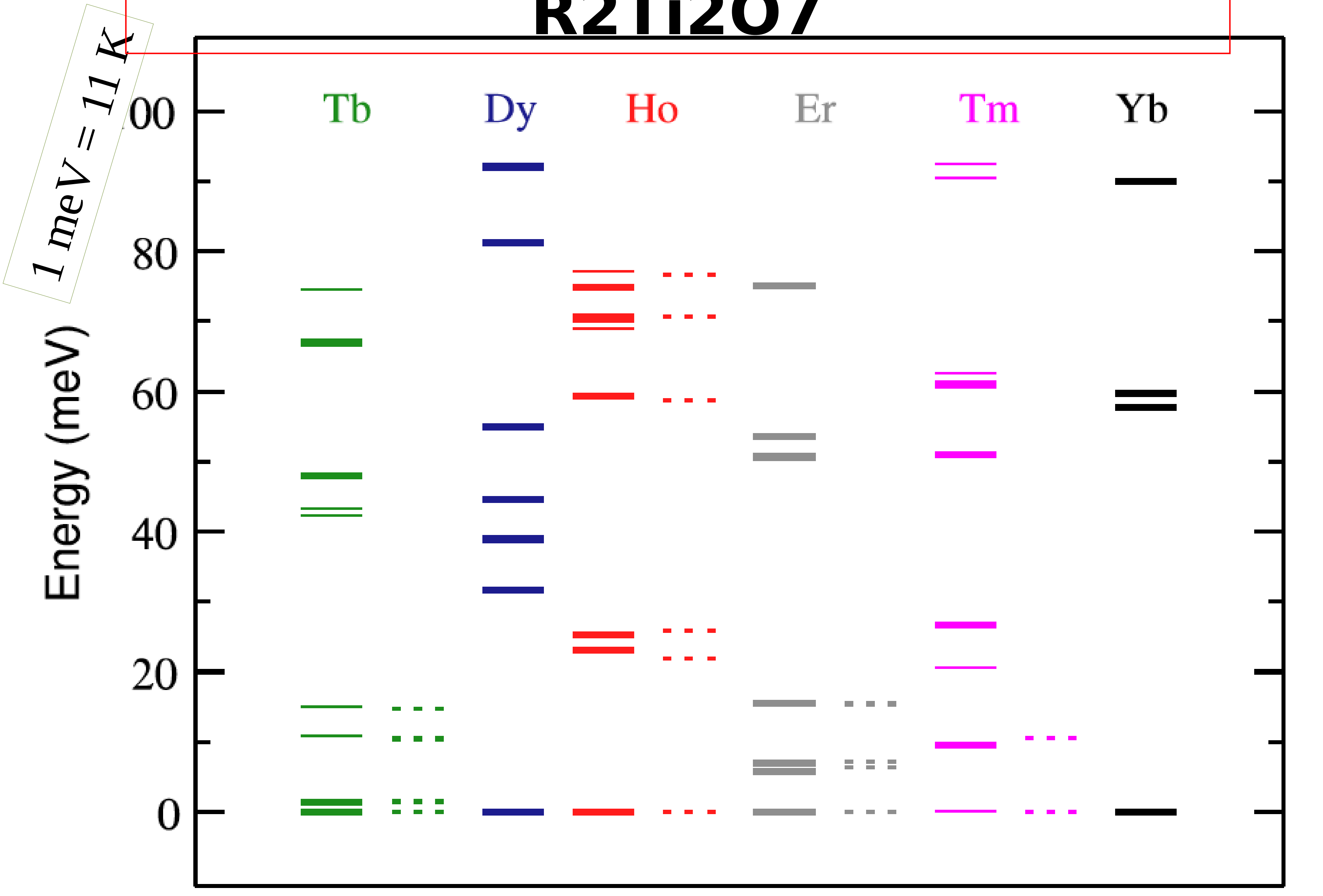
$$Q = \sum_{\beta \notin P} \frac{|\beta\rangle\langle\beta|}{E_0^{\alpha} - E_0^{\beta}}$$

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

Large denominator compared to the energy scale of H'

$$H_{eff} = P H_{int} P + \cancel{P H_{int} Q H_{int} P}$$

Crystal field energy levels in R2Ti2O7



Symmetry Allowed Hamiltonian on Pyrochlore Lattice

$$H = J_{zz} \sum_{\langle i,j \rangle} S_i^z S_j^z - J_{\pm} \sum_{\langle i,j \rangle} \left(S_i^+ S_j^- + S_i^- S_j^+ \right)$$

$$+ J_{z\pm} \sum_{\langle i,j \rangle} \left[S_i^z \left(\zeta_{ij} S_j^+ + \zeta_{ij}^* S_j^- \right) + i \leftrightarrow j \right]$$

$$+ J_{\pm\pm} \sum_{\langle i,j \rangle} \left(\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^- \right) \quad S = \tfrac{1}{2} \text{ operator}$$

S.H. Curnoe, Phys. Rev. B **78**, 094418 (2008)

Symmetry Allowed Hamiltonian on Pyrochlore Lattice

$$H = J_{\text{Ising}} \sum_{\langle i,j \rangle} S_i^z S_j^z + J_{\text{iso}} \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j +$$

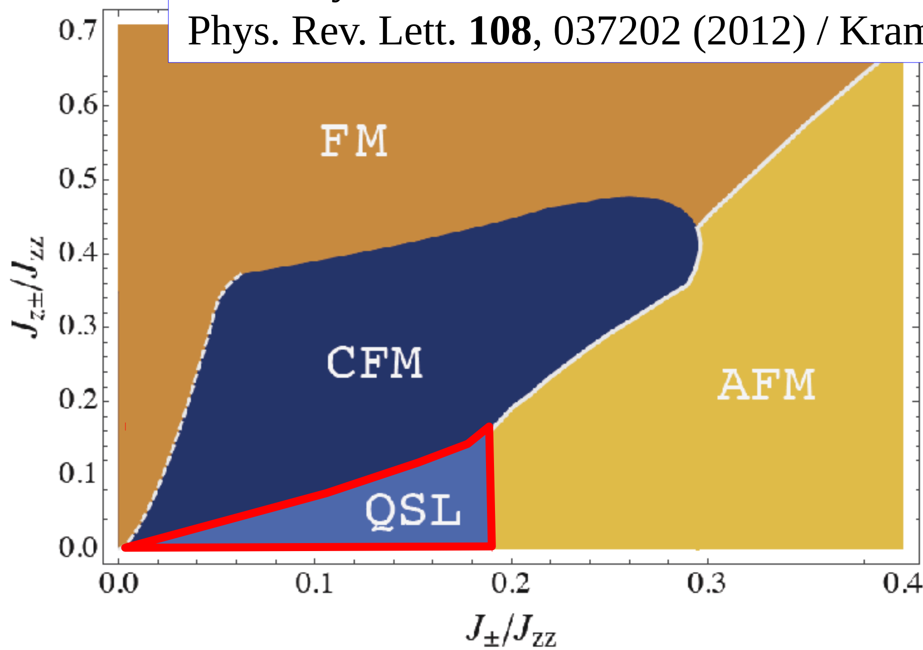
$$J_{\text{pd}} \sum_{\langle i,j \rangle} \left(\vec{S}_i \cdot \vec{S}_j - 3 \vec{S}_i \cdot \hat{r}_{ij} \hat{r}_{ij} \cdot \vec{S}_j \right) +$$

$$J_{\text{DM}} \sum_{\langle i,j \rangle} \vec{d}_{ij} \cdot \vec{S}_i \times \vec{S}_j$$

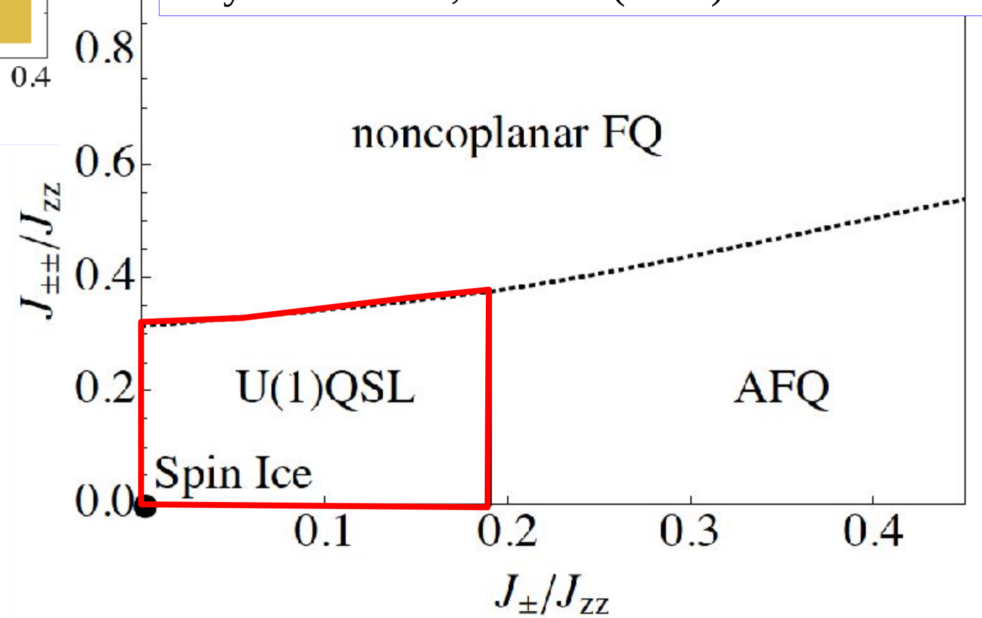
P. A. McClarty, S. H. Curnoe, M. J. P. Gingras;
Journal of Physics Conference Series **145**, 012032 (2009)

What is possible?

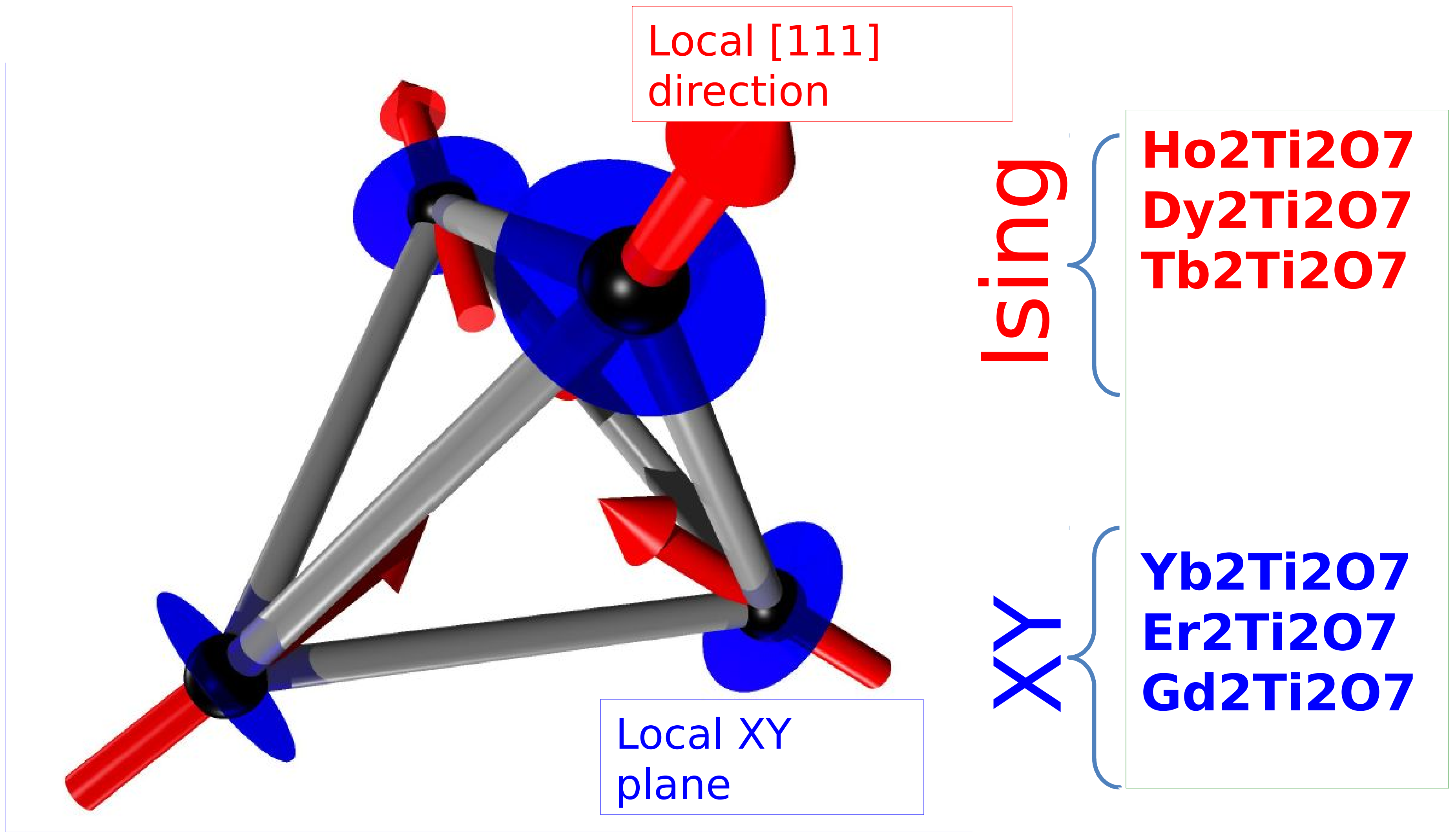
L. Savary and L. Balents,
Phys. Rev. Lett. **108**, 037202 (2012) / Kramers



S.-B. Lee, S. Onoda, and L. Balents,
Phys. Rev. B **86**, 104412 (2012) / non-Kramers



<u>*Review:*</u> *"Quantum Spin Ice: A Search for Gapless Quantum Spin Liquids in Pyrochlore Magnets"*
P.A. McClarty and M.J.P. Gingras, arXiv:1311.1817


Local [111]
direction
Local XY
plane
Ising
Ho2Ti2O7
Dy2Ti2O7
Tb2Ti2O7
XY
Yb2Ti2O7
Er2Ti2O7
Gd2Ti2O7

R2Ti2O7 vs R2Sn2O7

L: Ho2Ti2O7
R: Ho2Sn2O7

L: Dy2Ti2O7
R: Dy2Sn2O7

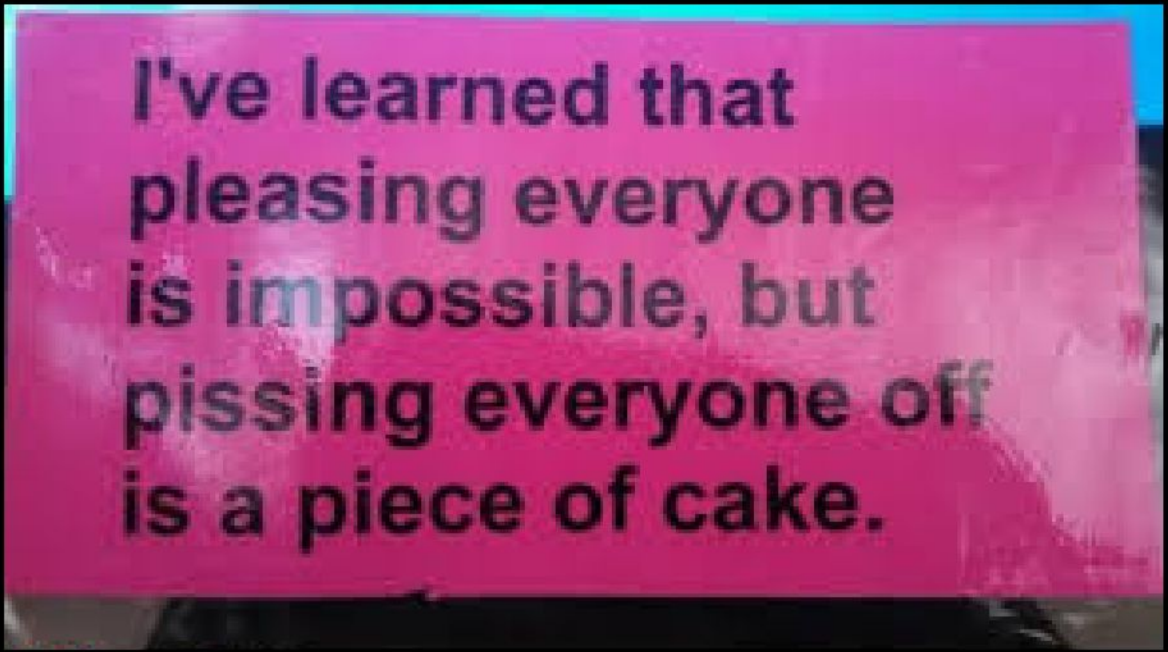
L: Pr2Sn2O7
R: Pr2Zr2O7

L: Tb2Ti2O7
R: Tb2Sn2O7

L: Yb2Sn2O7
R: Yb2Ti2O7

L: Gd2Ti2O7
R: Gd2Sn2O7

RE: on references of other people's work provided henceforth

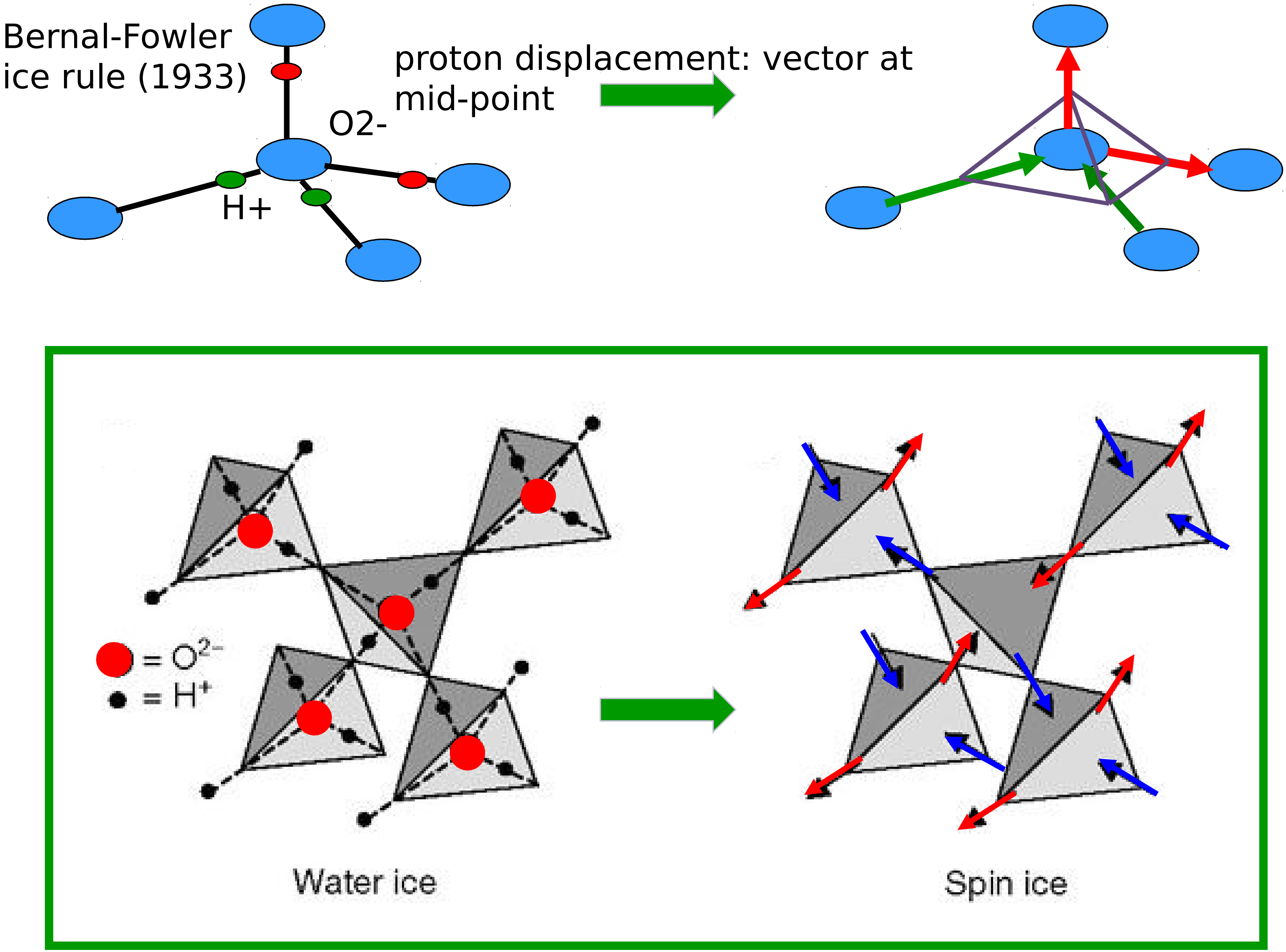


So, in order to be fair, I'll try to p… o… everybody equally and endeavour to refer as much as possible only to my own work.

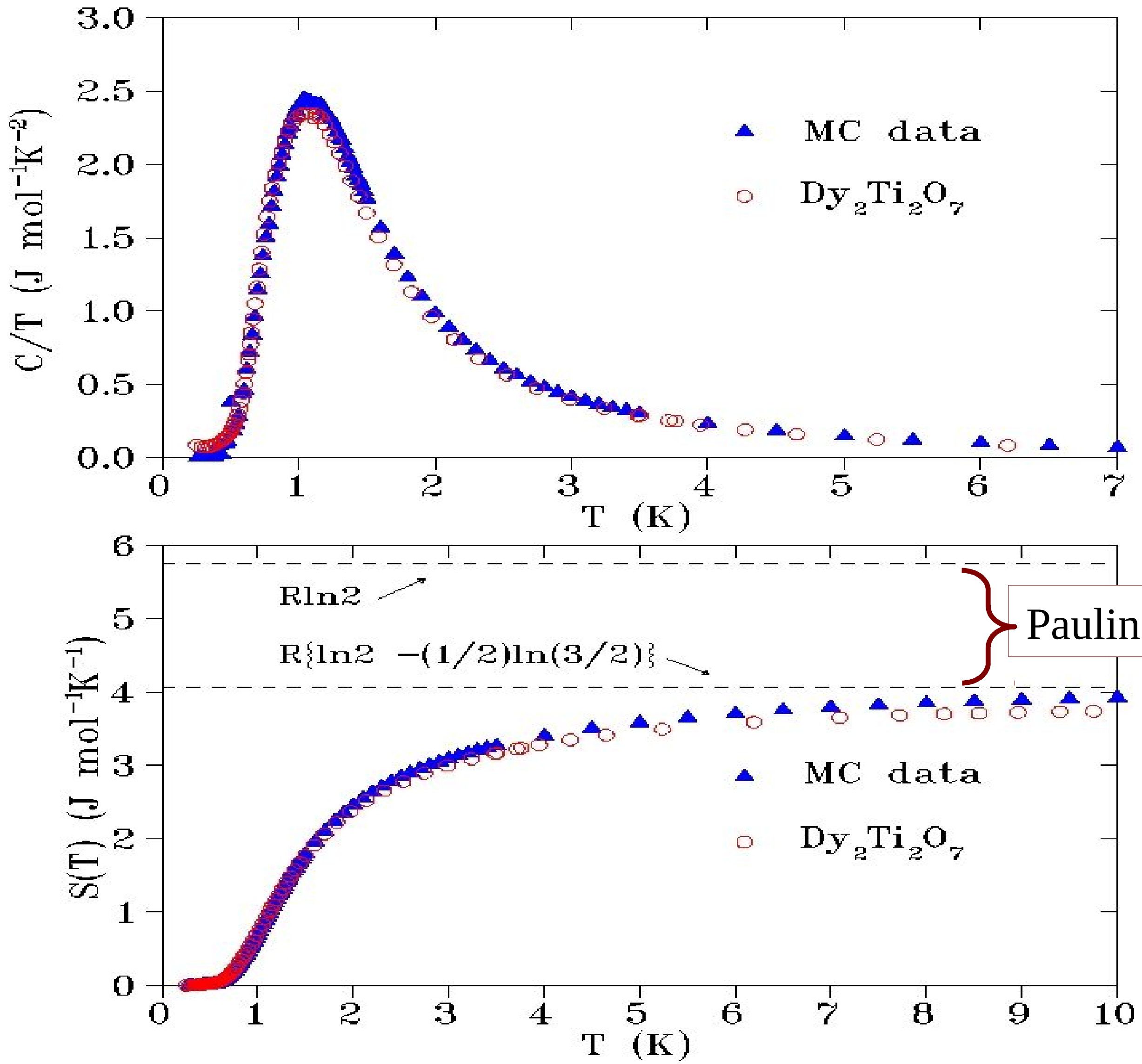
$(Dy,Ho)_2(Ti,Sn,Ge)_2O_7$

Phenomenology:

Classical dipolar spin ices


Bernal-Fowler
ice rule (1933)
O2-
H+
proton displacement: vector at
mid-point
= O2-
= H+
Water ice
Spin ice

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



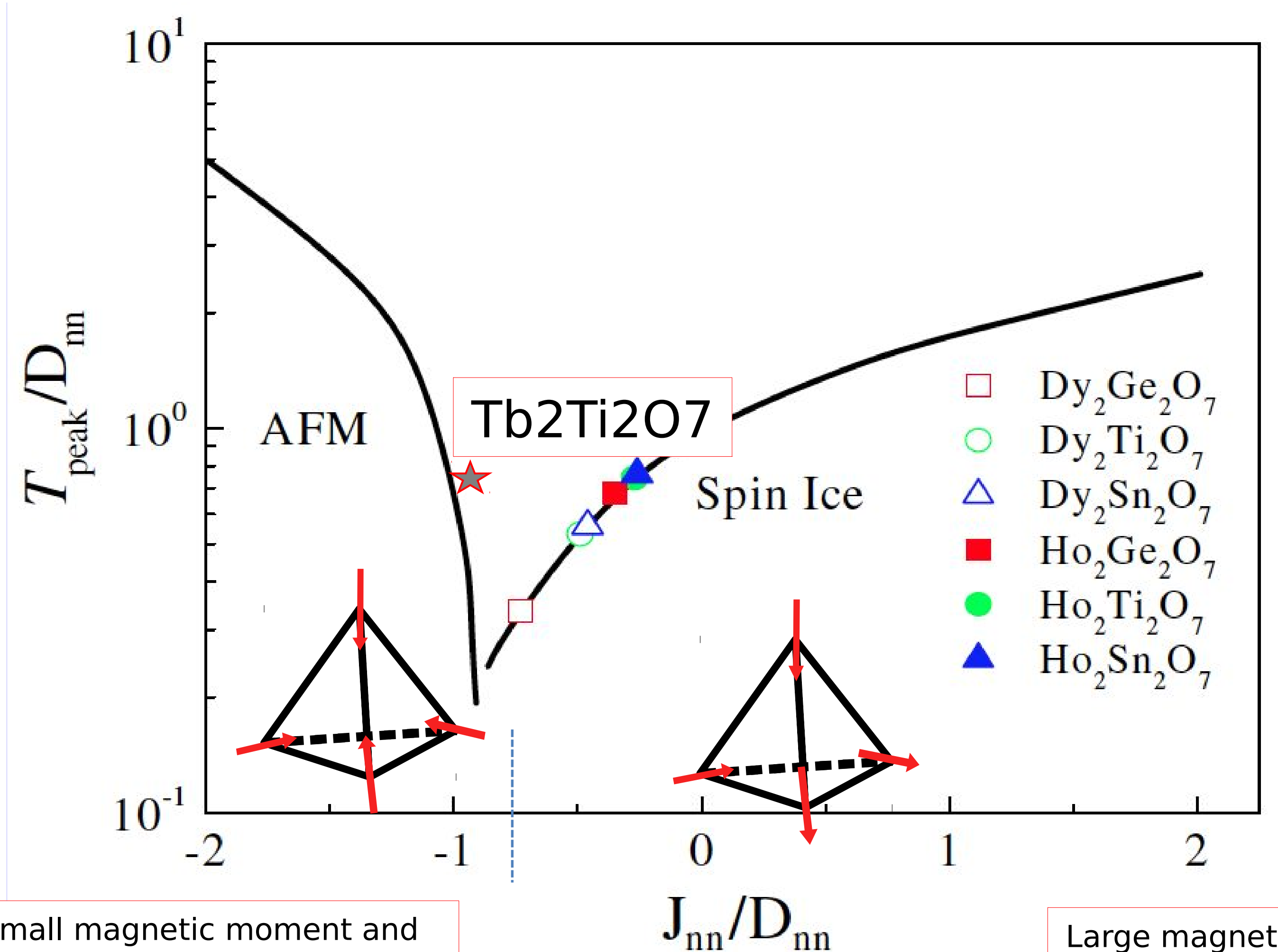
Pauling entropy= (kB/2)ln(3/2)

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

PRL 108, 207206 (2012) PHYSICAL REVIEW LETTERS week ending 18 MAY 2012

Chemical Pressure Effects on Pyrochlore Spin Ice

H. D. Zhou,[1,*] J. G. Cheng,[2] A. M. Hallas,[3] C. R. Wiebe,[1,3,4] G. Li,[1] L. Balicas,[1] J. S. Zhou,[2] J. B. Goodenough,[2] J. S. Gardner,[5,6] and E. S. Choi[1]



Small magnetic moment and effective antiferromagnetic exchange

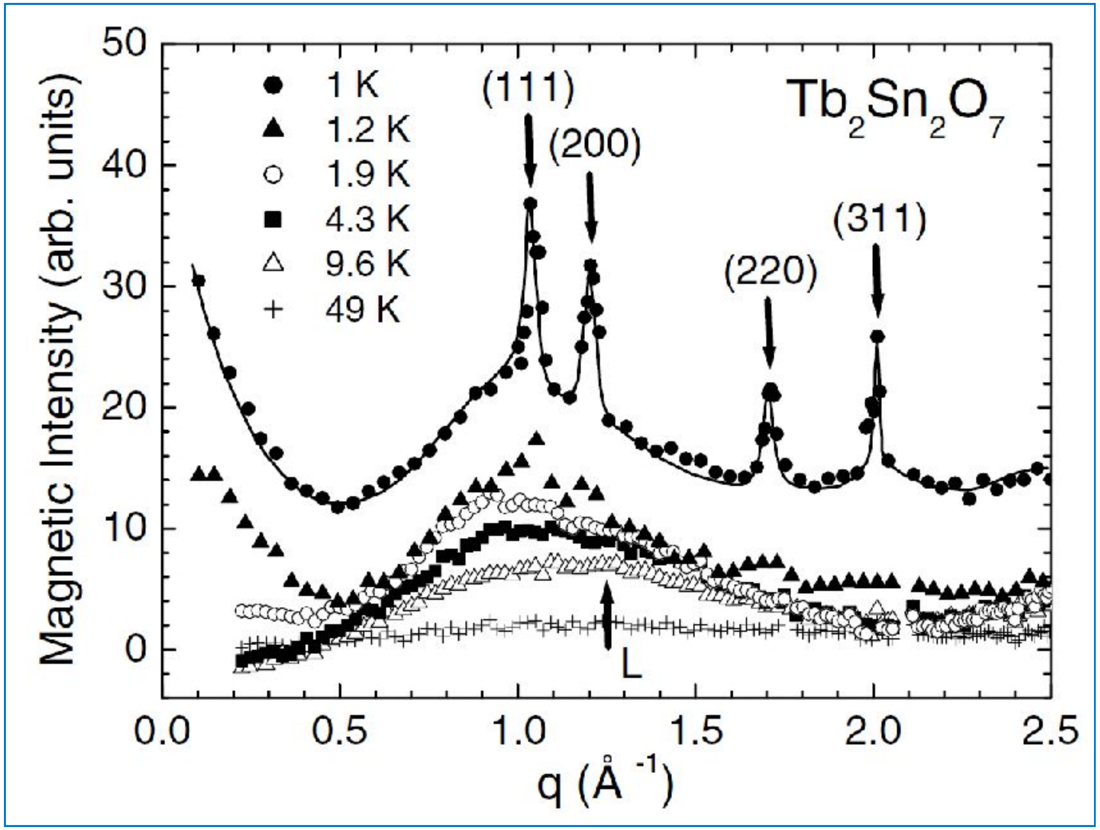
Large magnetic moment or effective ferromagnetic exchange

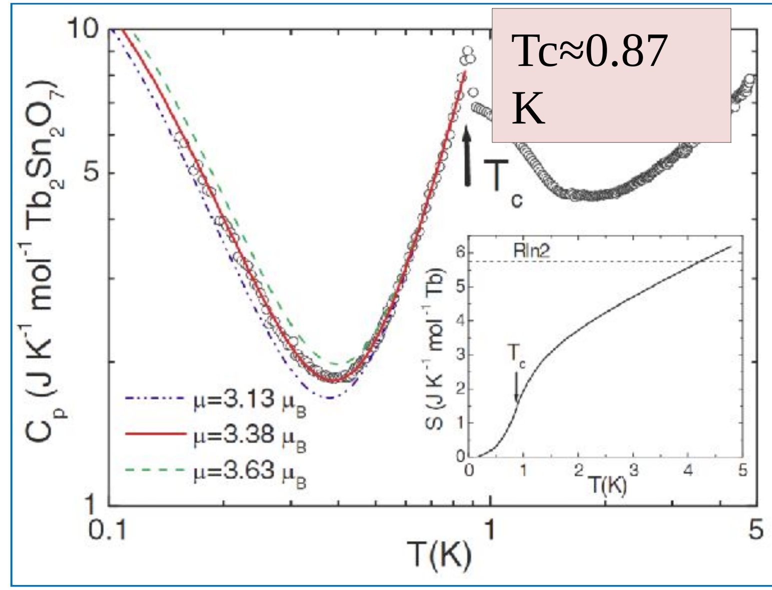
Tb2Ti2O7 and Tb2Sn2O7

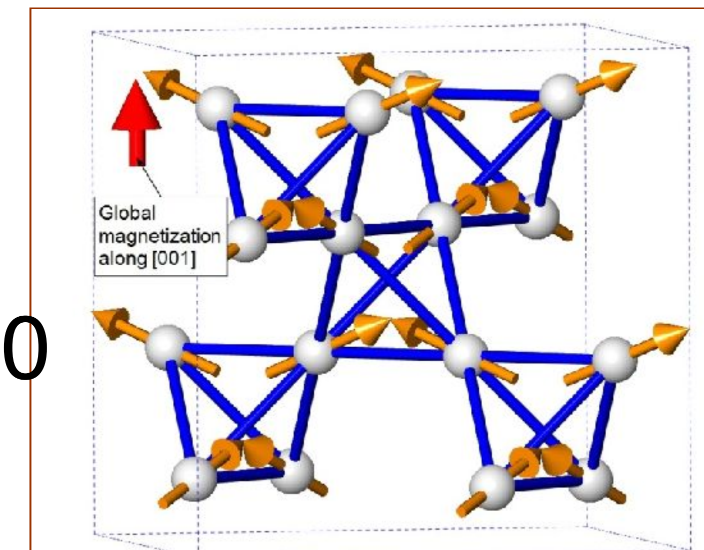
Phenomenology:

Well, it's a mess alright!

Meanwhile: Tb2Sn2O7 has a long range ordered spin ice







$\boldsymbol{q}$=000

“Ordered Spin Ice State and Magnetic Fluctuations in Tb2Sn2O7”; Mirebeau *et al*. Phys. Rev. Lett. **94**, 246402 (2005).

PRL 98, 157204 (2007) PHYSICAL REVIEW LETTERS week ending 13 APRIL 2007

Dynamically Induced Frustration as a Route to a Quantum Spin Ice State in $Tb_2Ti_2O_7$ via Virtual Crystal Field Excitations and Quantum Many-Body Effects

Hamid R. Molavian,[1] Michel J. P. Gingras,[1,2] and Benjamin Canals[1,3]

PRL 109, 017201 (2012) PHYSICAL REVIEW LETTERS week ending 6 JULY 2012

Power-Law Spin Correlations in the Pyrochlore Antiferromagnet $Tb_2Ti_2O_7$

T. Fennell,[1,*] M. Kenzelmann,[2] B. Roessli,[1] M. K. Haas,[3] and R. J. Cava[3]

PHYSICAL REVIEW B 87, 094410 (2013)

Antiferromagnetic spin ice correlations at $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ in the ground state of the pyrochlore magnet $Tb_2Ti_2O_7$

K. Fritsch,[1] K. A. Ross,[1,2,3] Y. Qiu,[3,4] J. R. D. Copley,[3] T. Guidi,[5] R. I. Bewley,[5] H. A. Dabkowska,[6] and B. D. Gaulin[1,6,7]

PRL 111, 087201 (2013) PHYSICAL REVIEW LETTERS week ending 23 AUGUST 2013

Anisotropic Propagating Excitations and Quadrupolar Effects in $Tb_2Ti_2O_7$

Solène Guitteny,[1] Julien Robert,[1] Pierre Bonville,[2] Jacques Ollivier,[4] Claudia Decorse,[3] Paul Steffens,[4] Martin Boehm,[4] Hannu Mutka,[4] Isabelle Mirebeau,[1] and Sylvain Petit[1]

- Evidence for complex correlations, both in the dynamics and quasi-static
- Evidence for development of extended correlations with peaks at (1/2,1/2,1/2)
- Evidence for concomitant suggesting quantum spin-ice correlations.

PHYSICAL REVIEW B 87, 060408(R) (2013)

Long-range order and spin-liquid states of polycrystalline $Tb_{2+x}Ti_{2-x}O_{7+y}$

T. Taniguchi,[1] H. Kadowaki,[1] H. Takatsu,[1] B. Fåk,[2] J. Ollivier,[3] T. Yamazaki,[4] T. J. Sato,[5] H. Yoshizawa,[5] Y. Shimura,[4] T. Sakakibara,[4] T. Hong,[6] K. Goto,[1] L. R. Yaraskavitch,[7] and J. B. Kycia[7]

But…
evidence for extreme sample sensitivity

$(Gd)_2(Ti,Sn)_2O_7$

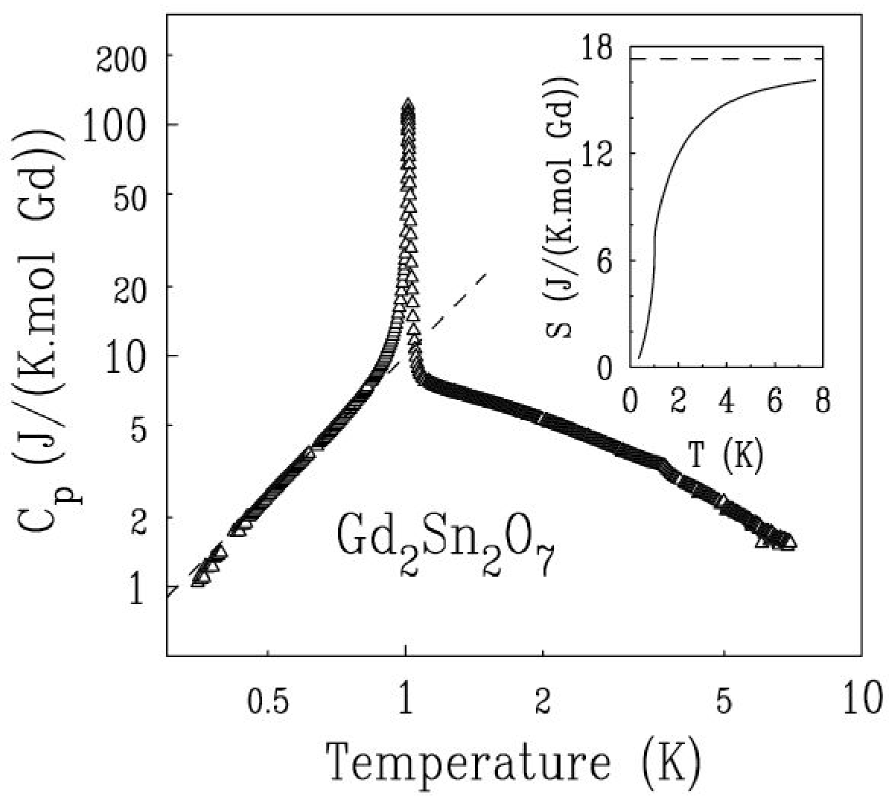
Phenomenology:

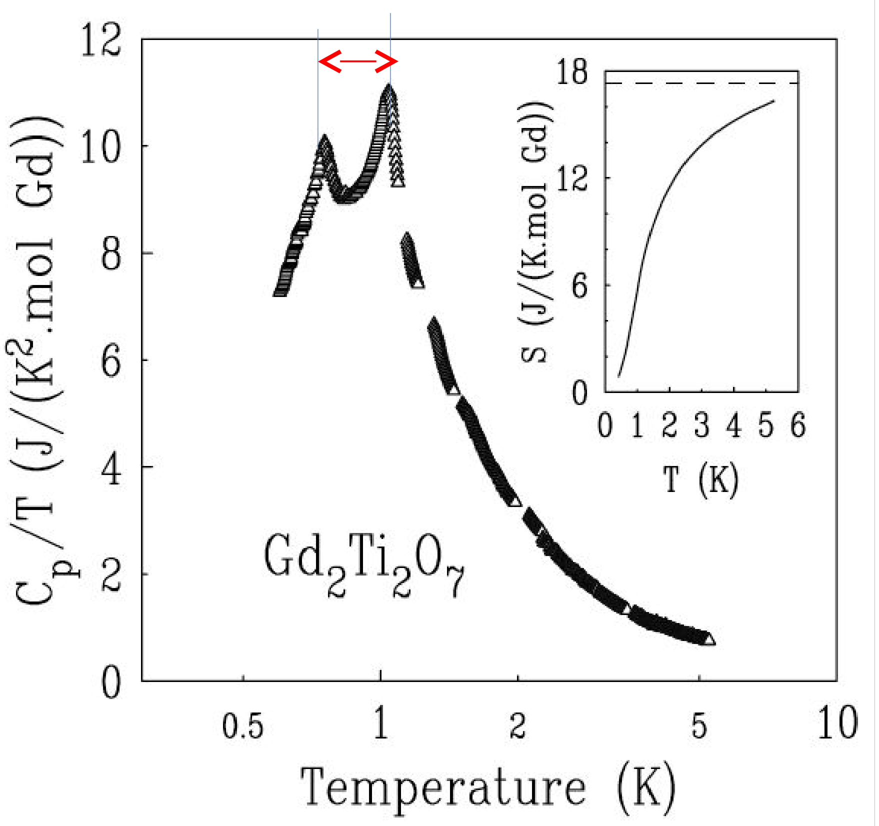
- $Gd_2Sn_2O_7$: “simple” long-range order. [Phys. Rev. Lett. **99**, 097201(2007)]
- $Gd_2Ti_2O_7$:
  - Multiple phase transitions
  - Partially ordered intermediate 4-***k*** state
  - Order-by-disorder at *Tc*
  - Unusual criticality at *Tc*

arXiv:1310.5146; B. Javanparast, Z. Hao, M. Enjalran, M. J. P. Gingras

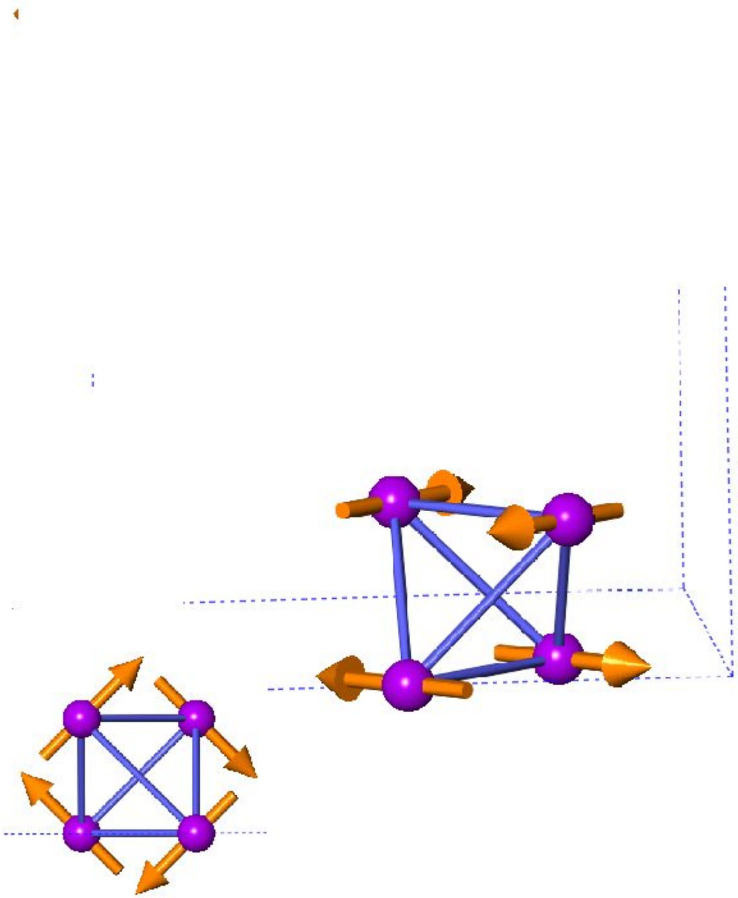
“Fluctuation-Driven Selection at Criticality in a Frustrated Magnetic System: the Case of Multiple-k Partial Order on the Pyrochlore Lattice”

Gd2Ti2O7 vs Gd2Sn2O7

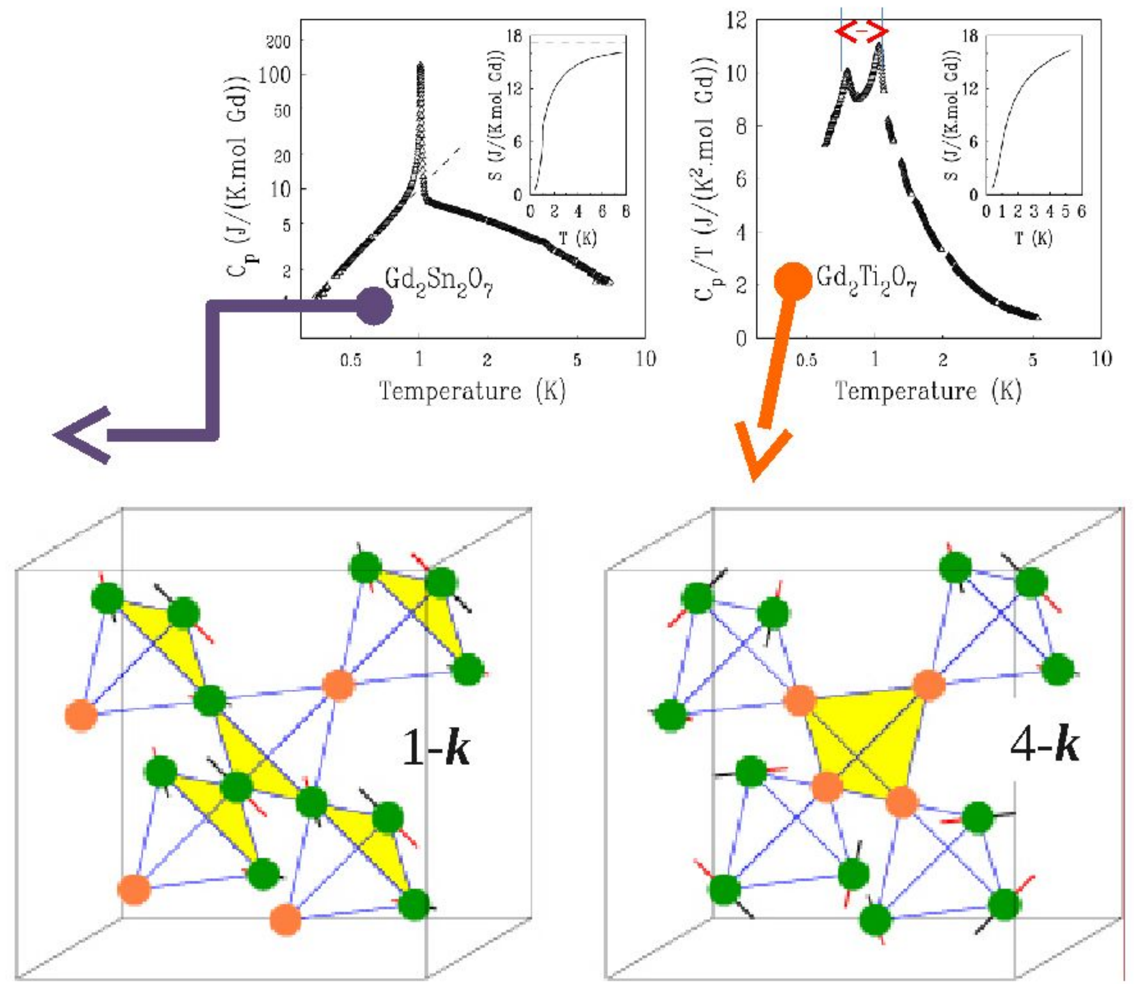




Gd2Ti2O7 vs Gd2Sn2O7



Palmer & Chalker.
Phys Rev B **62**, 488
(2000)



arXiv:1310.5146; B. Javanparast, Z. Hao, M. Enjalran, M. J. P. Gin

"Fluctuation-Driven Selection at Criticality in a Frustrated Magnetic Syste the Case of Multiple-k Partial Order on the Pyrochlore Lattice"

Er2Ti2O7 and Er2Sn2O7

Phenomenology:

- Er2Ti2O7: most likely concurrent quantum & thermal *order-by-disorder*

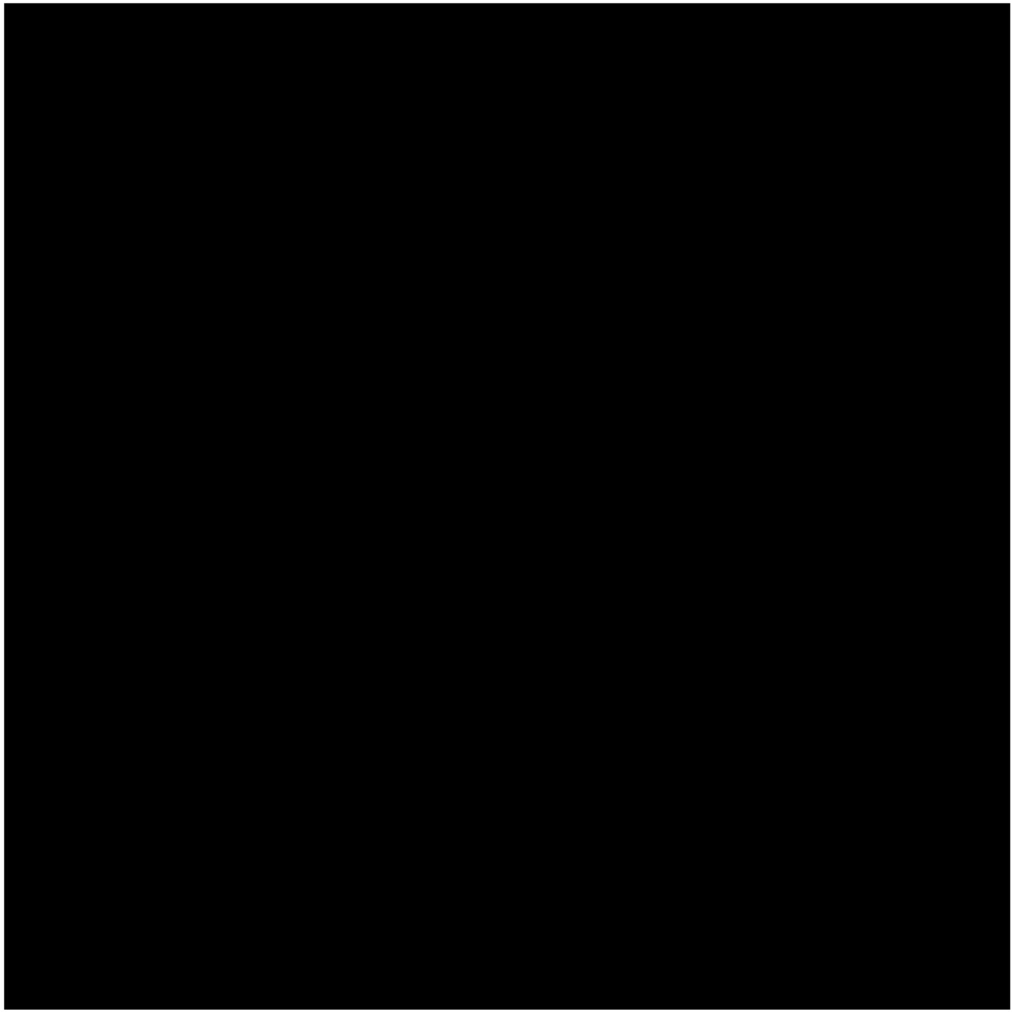
See Mike hitomirsky's talk this afternoon

  - Quantum Order by Disorder and Accidental Soft Mode
    V. Gvozdikova, P. C. W. Holdsworth, and R. Moessner,
    (2012)
  - Order by Quantum Disorder in Er2Ti2O7 Lucile Savary, Kate A. Ross, Bruce D. Gaulin, Jacob P. C. Ruff, and Leon Balents, Phys. Rev. Lett. **109**, 167201 (2012)
  - Ground state phase diagram of generic *XY* pyrochlore magnets with quantum fluctuations, Anson W. C. Wong, Zhihao Hao, and Michel J.P. Gingras Phys. Rev. B **88**, 144402 (2013)
  - Phase transition and thermal order-by-disorder in the pyrochlore quantum antiferromagnet Er2Ti2O7, J. Oitmaa, R.R.P. Singh, B. Javanparast, A.G.R. Day, B.V. Bagheri, M.J.P. Gingras; arXiv:1305.2935 (to appear in PRB/RC)
  - Living on the edge: ground-state selection in quantum spin-ice pyrochlores ,
    H. Yan, O. Benton, L..D.C. Jaubert, N. Shannon, arXiv:1311.3501

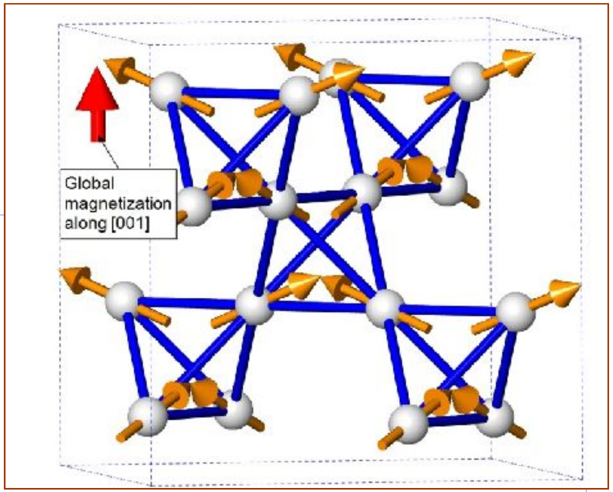
- Er2Sn2O7: local short-range order akin to Gd2Sn2O7, but not true long-range order down to 100mK
  - Palmer-Chalker correlations in the *XY* pyrochlore antiferromagnet Er2Sn2O7
    S. Guitteny, S. Petit, E. Lhotel, J. Robert, P. Bonville, A. Forget, and I. Mirebeau; Phys. Rev. B **88**, 134408 (2013)

$Yb_2Ti_2O_7$ and $Yb_2Sn_2O_7$

Phenomenology:



Yb2Ti2O7 and Yb2Sn2O7



Phenomenology:

- Yb2Sn2O7: seems to be a canted (FM) (akin to Tb2Sn2O7)
  - Dynamical Splayed Ferromagnetic Ground State in the Quantum Spin Ice Yb2Sn2O7, A. Yaouanc *et al.* Phys. Rev. Lett. **110**, 127207 (2013).

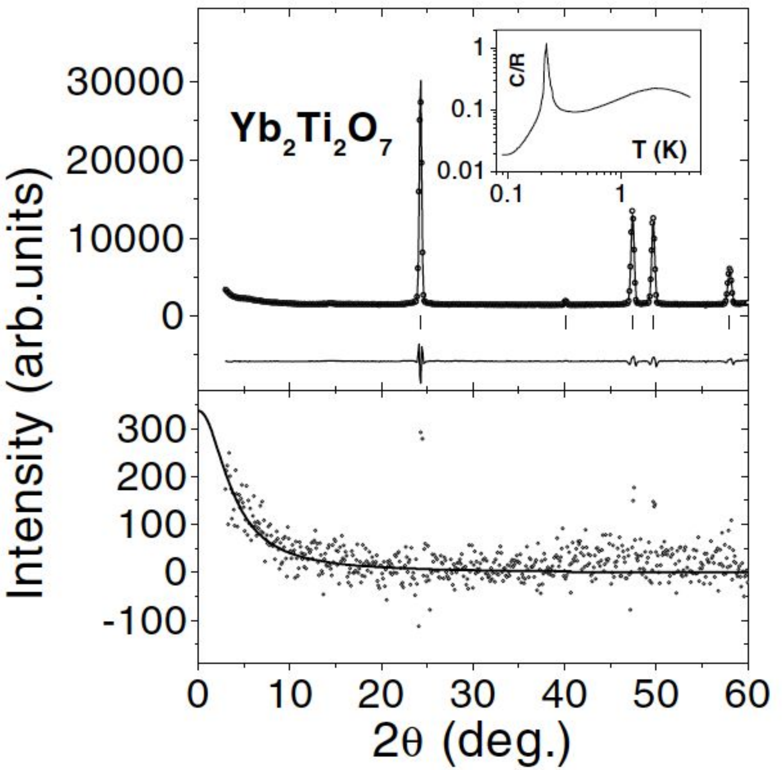
- Yb2Ti2O7: no agreement yet
  - Long-range (ferrimagnetic) order at *T*<~240 mK (Yasui *et al.* JPSJ, Chang *et al.* Nat. Comm.)

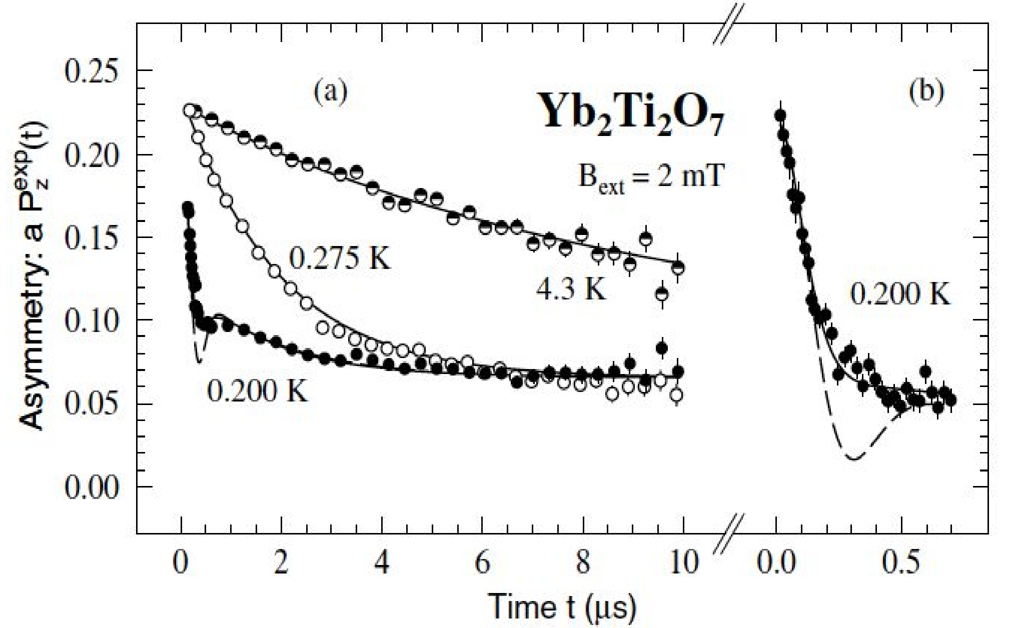
  VS

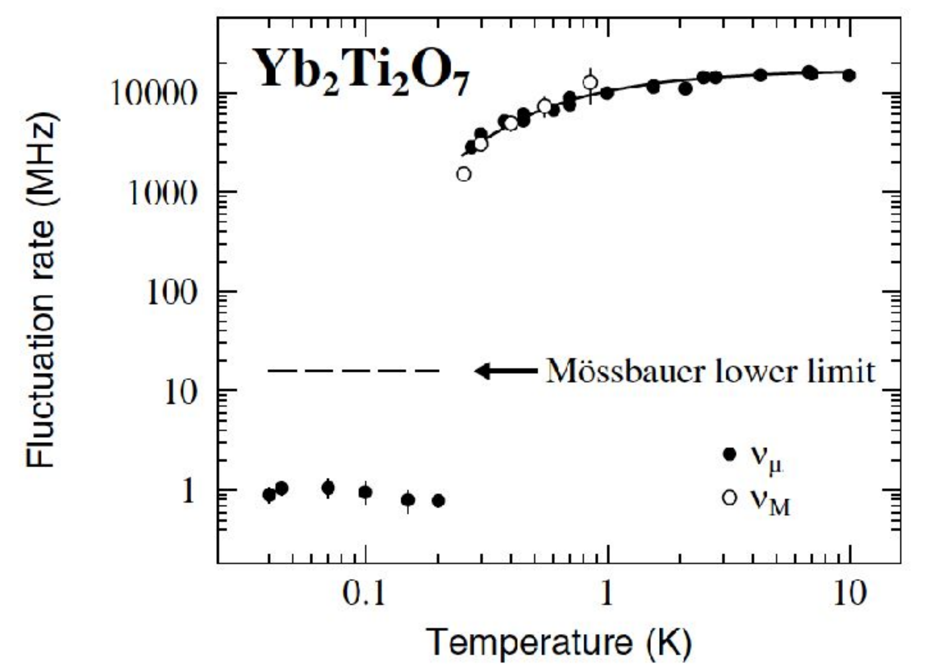
  - No transition down to 20 mK (perhaps related to U(1) QSL of a quantum spin ice?)

Most recent experimental work (?) is:

Unconventional magnetic ground state in Yb2Ti2O7 R. M. D'Ortenzio *et al.* Phys. Rev. B **88**, 134428 (31 October 2013) / See references therein.







Hodges *et al.*
Phys. Rev. Lett. **88**, 077204 (2002)

Very confused about Yb2Ti2O7

It would seem that we know the effective S=1/2 Hamiltonian (from inelastic neutron scattering in strong field – Ross *et al.* Phys

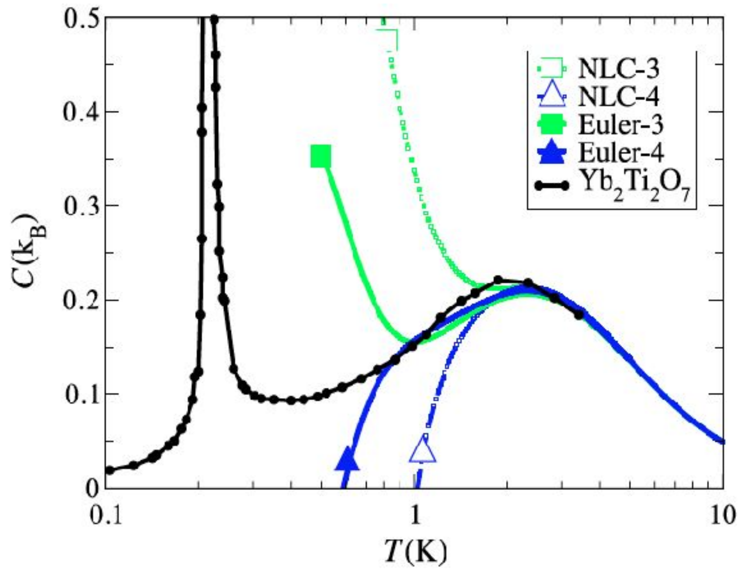
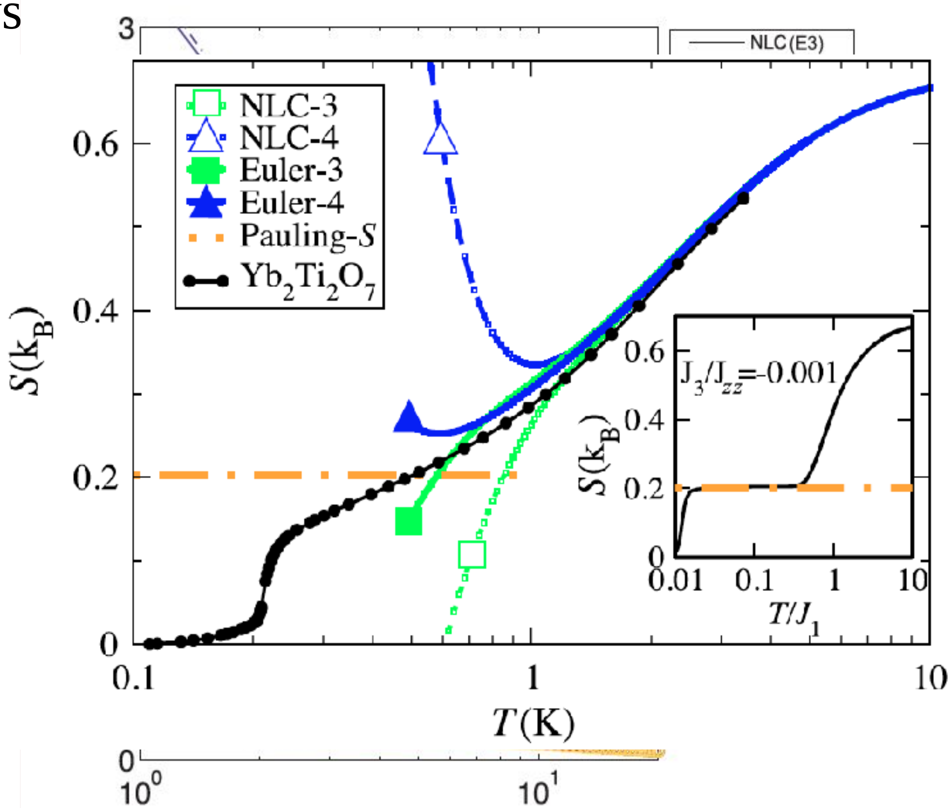
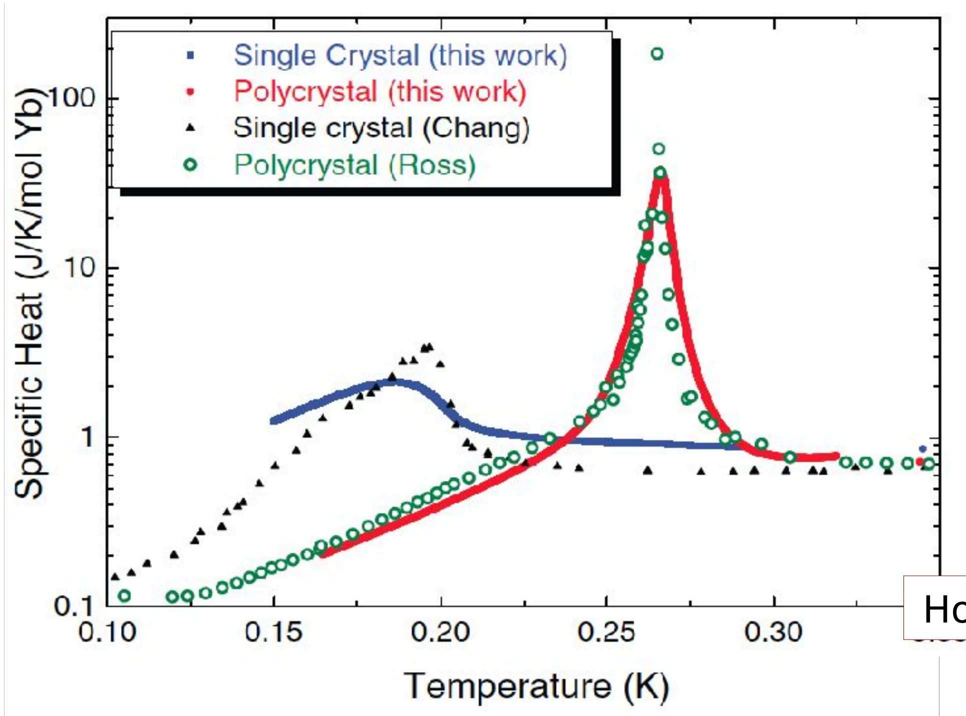


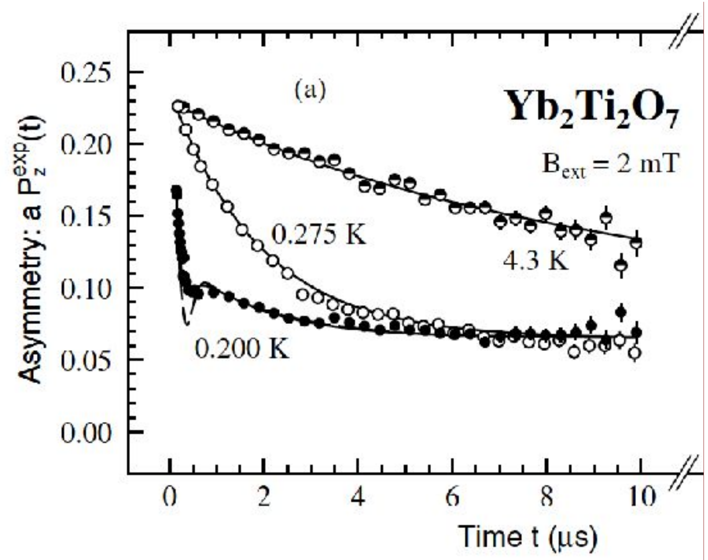
FIG. 2 (color online). Heat capacity $C(T)$ per mole of Yb for the model parameters in Ref. [16], in units of the Boltzmann constant k_B, calculated via NLC (up to the fourth order NLC together with Euler extrapolations) are compared with experimental data for $Yb_2Ti_2O_7$. The filled small black circles are data from Ref. [25].



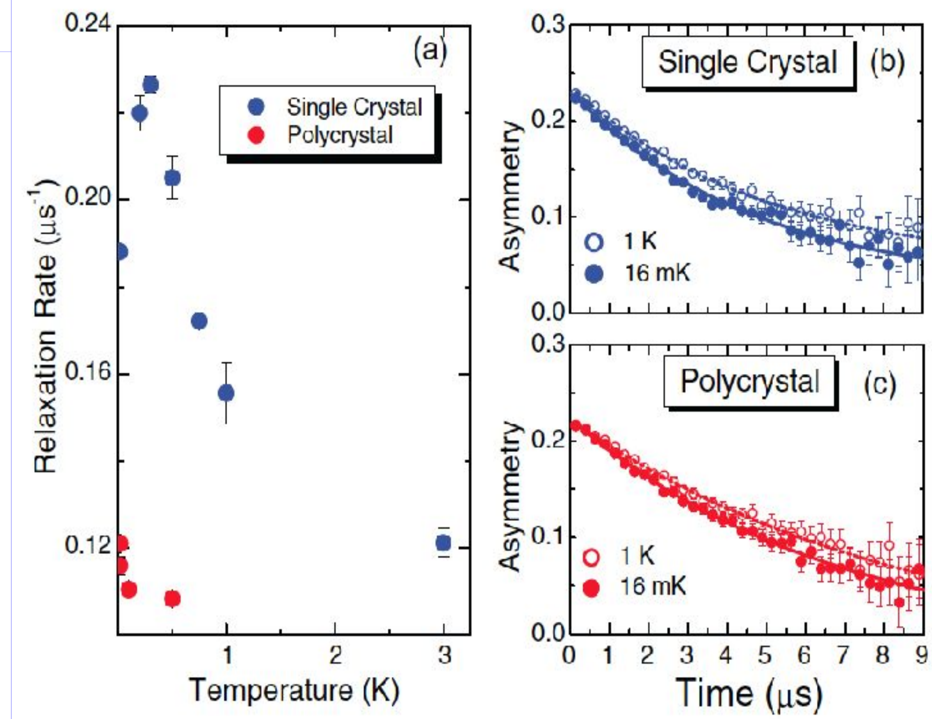
Vindication of Yb2Ti2O7 as a Model Exchange Quantum
Applegate, Hayre, Singh, Lin, Day and Gingras
Phys. Rev. Lett. **109**, 097205 (2012).

Thermodynamic properties of Yb2Ti2O7 pyrochlore as a function of temperature and magnetic field: Validation of a quantum spin ice exchange Hamiltonian; Hayre, Ross, Applegate, Lin, Singh, Gaulin and Gingras, Phys. Rev. B **87**, 184423 (2013).

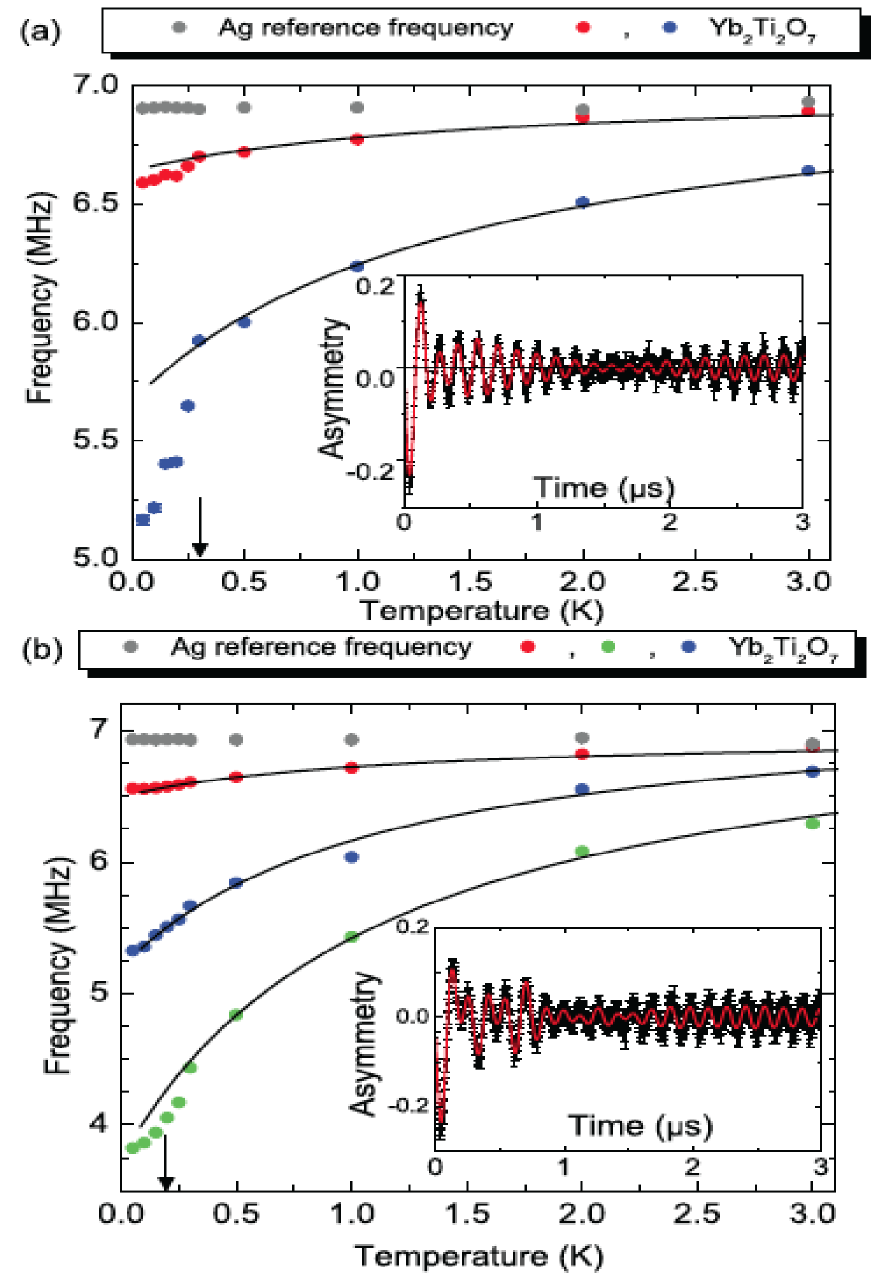




Hodges *et al.* Phys. Rev. Lett. **88**, 077204 (2002)



R. M. D'Ortenzio *et al.*
Phys. Rev. B **88**, 134428 (2013)



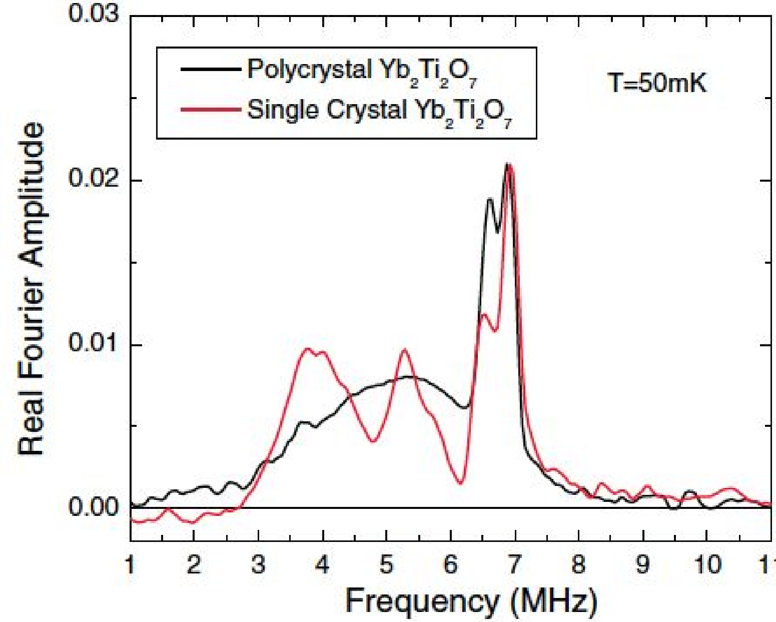
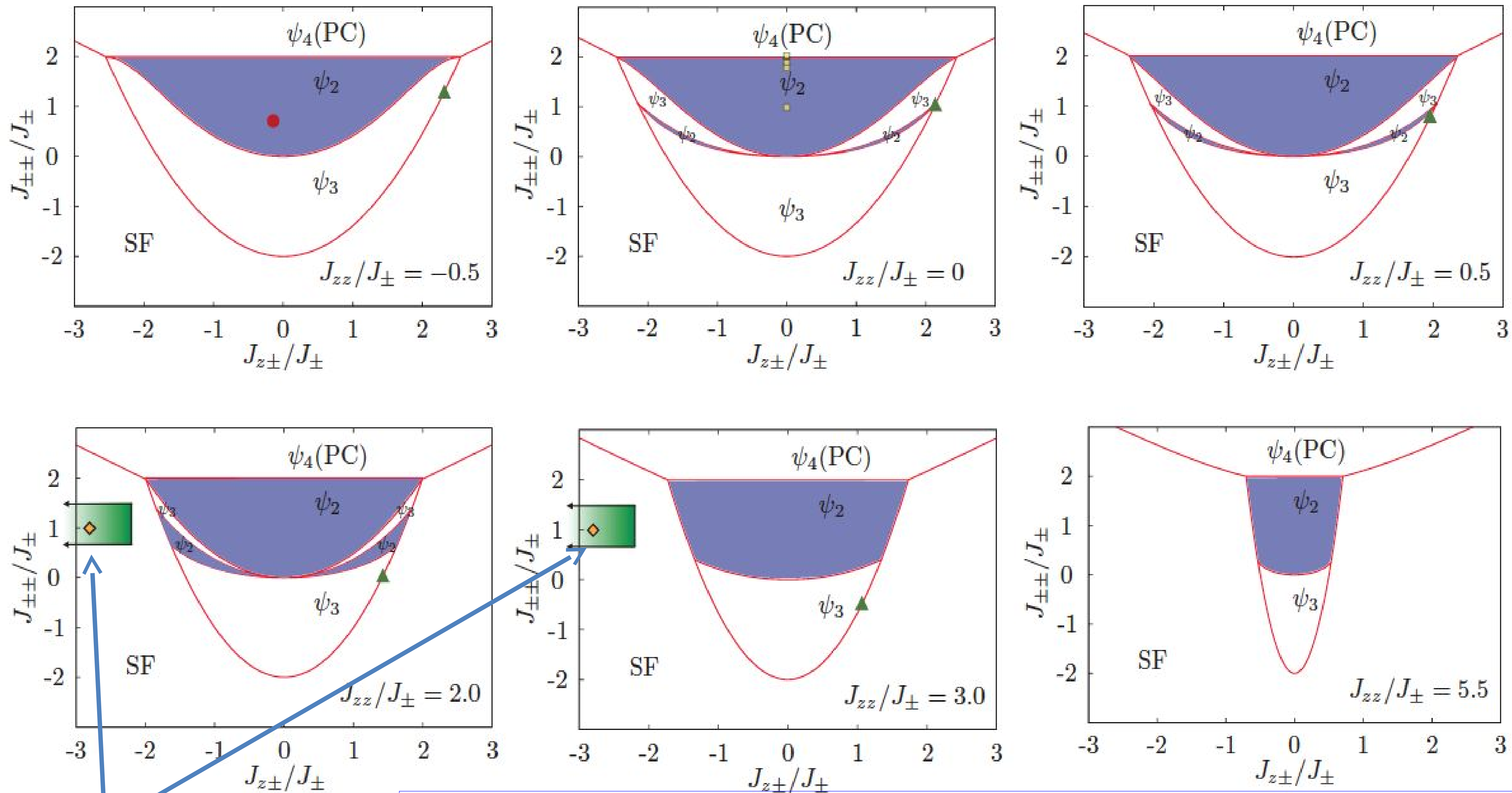
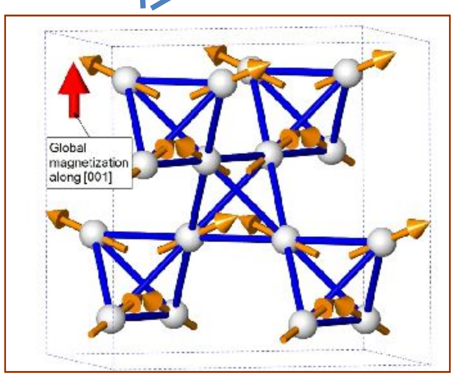


FIG. 3. (Color online) Fourier transform of μSR asymmetry spectra in a transverse field of 50 mT at $T = 50$ mK. The single-crystal $Yb_2Ti_2O_7$ shows four resolvable frequencies (red) and the polycrystalline $Yb_2Ti_2O_7$ shows three (black). This motivates $n = 4$ for the single-crystal and $n = 3$ in the polycrystalline $Yb_2Ti_2O_7$ for Eq. (1). The sharp signal at approximately 7 MHz for both samples reflects the precession of the muons in the Ag cryostat tails.

R. M. D'Ortenzio *et al.*
Phys. Rev. B **88**, 134428 (2013)





Ground state phase diagram of generic *XY* pyrochlore magnets with quantum fluctuations, A.W.C. Wong, Zhihao Hao, and M.J.P. Gingras,
Phys. Rev. B **88**, 144402 (2013)

BUT DIFFERENT POINT OF VIEW IN:

Living on the edge: ground-state selection in quantum spin-ice pyrochlores, H. Yan, O. Benton, L.D.C. Jaubert, N. Shannon, arXiv:1311.3501

Conclusion

- Rare earth pyrochlore oxides display a smorgasbord of phenomena
- Much has been understood in the past 2 years (in particular for $Er_2Ti_2O_7$, perhaps for $Gd_2Ti_2O_7$ as well).
- Confusion remains as per $Tb_2Ti_2O_7$ & $Yb_2Ti_2O_7$
  - These two compounds may be related to a *quantum spin ice state* – this is a most exciting prospect. Namely a discovery a U(1) quantum spin liquid. This remains to be established beyond doubt (in $Pr_2(Sn,Zr)_2O_7$ as well)
  - Both materials display extreme sample variation sensitivity (at least in image furnace grown single-crystals)
- So, more experiments & more theory is needed
- A definite resolution of “all” significant problems presented by insulating $R_2M_2O_7$ systems may be in reached within a foreseeable future

Morale de l'histoire …

- "These rare-earth systems are of no-interest in the search for exotic quantum states of matter (e.g. quantum spin liquid) because J [illegible] 1/2"
- Not so… What matters is the existence and details of the effective spin dynamics/algebra in the low-energy sector.