Geometrical Frustration

Materials, Physics, and the Need for New Probes

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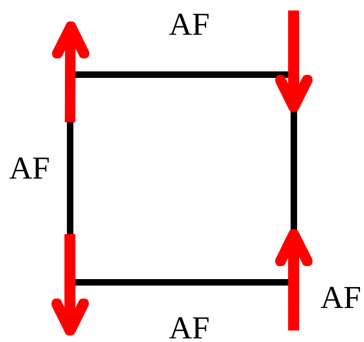
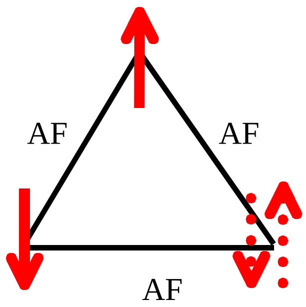
Outline

- Basic Ideas
- Materials Specifications for High Frustration
- Spin Liquid and Spin Ice in Gd, Dy systems
- Need for Different Probe – Magnetic X-rays?

1st Materials Specification – Triangle Subunit

Unfrustrated and Frustrated Spins





Local interaction

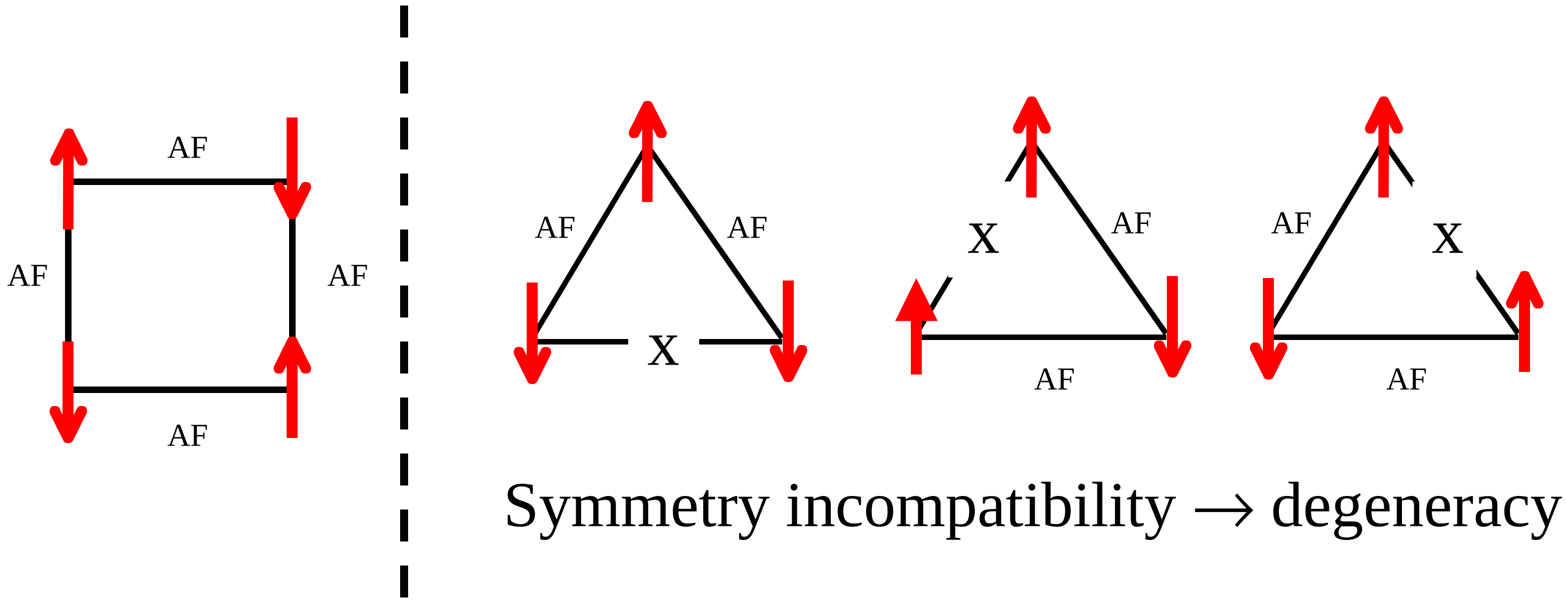
ß

Order Parameter symmetry

ß

Symmetry incompatibility – OP Û Space group

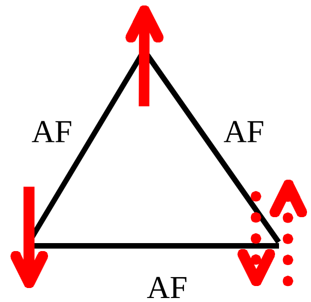
Geometrical Frustration *is* spectral weight downshift



Symmetry incompatibility → degeneracy

A. Ramirez, in Handbook of Magnetic Materials, 2001

Geometrical Frustration – Materials Considerations

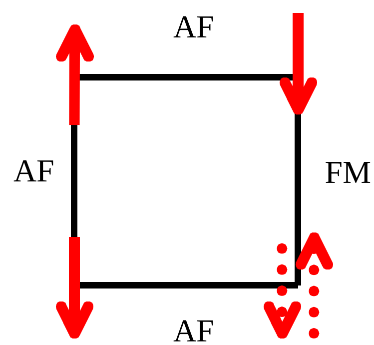


Frustration →

Disorder ↓

Antiferromagnetism Ferromagnetism Metamagnetism Ferrimagnetism	Geometrical Frustration
percolation, random fields	Spin glass

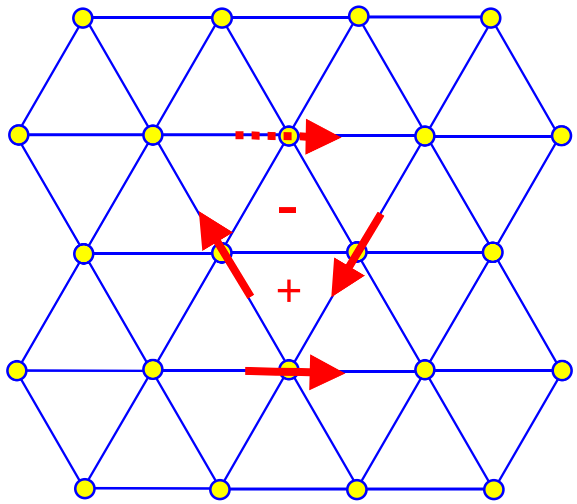
$$H=\Sigma S_i \cdot S_j$$



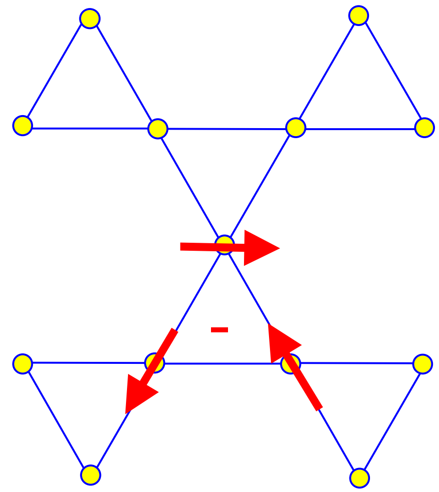
2nd Materials Criterion – Marginal Constraint

For vector spins

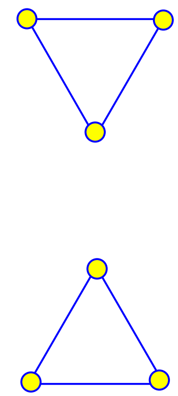
e.g., in 2 Dimensions



Triangular lattice
$D_M/N = -2$



Kagome
$D_M/N = 0$

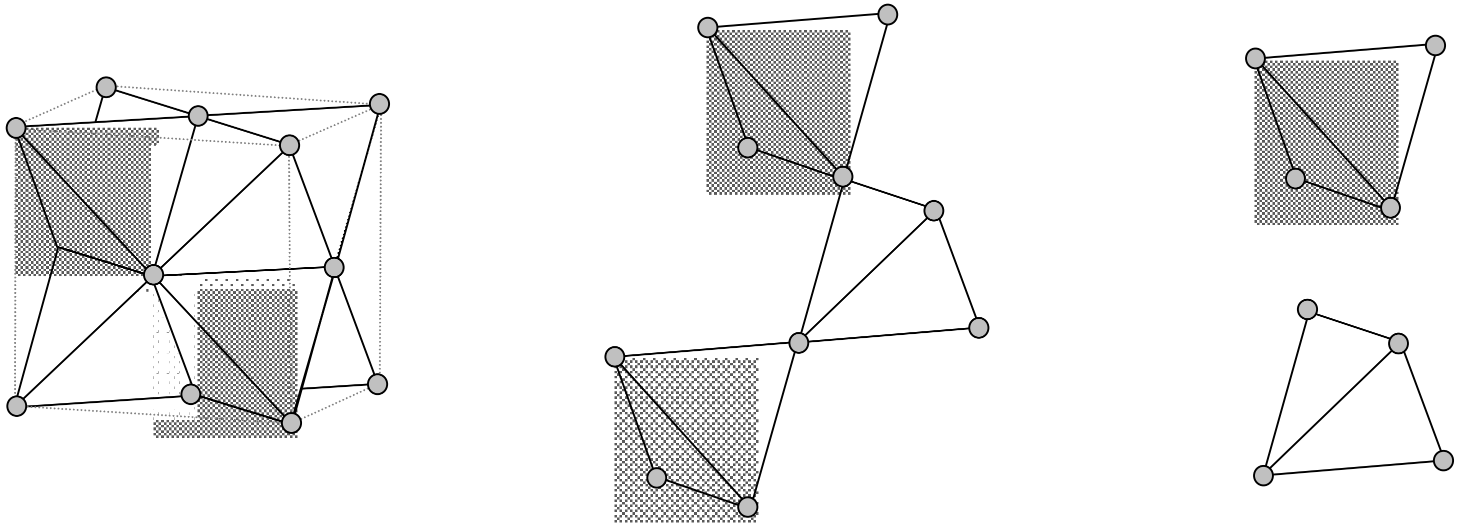


Triangle
$D_M/N = 1$

\ **Need a marginally-constrained system in general**

R. Moessner & J. Chalker, *Phys. Rev. B*, 1998

Role of *Underconstraint* in 3 Dimensions

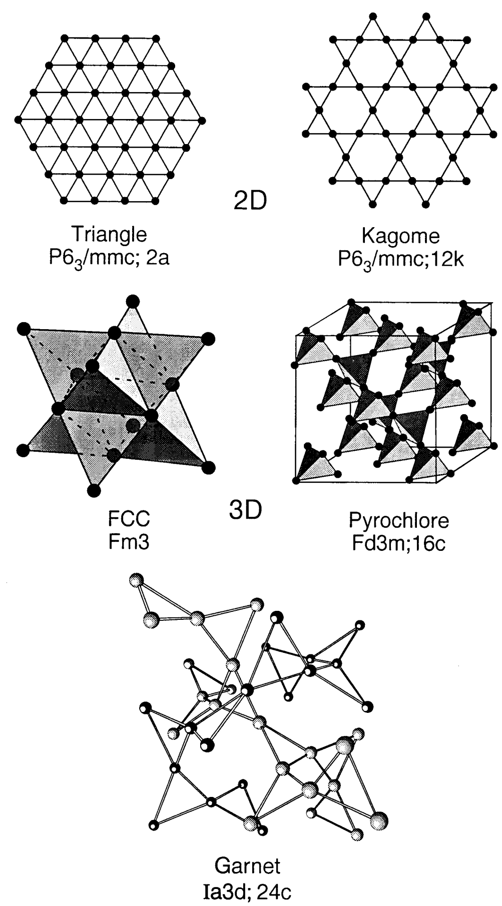


FCC
$D_M/N = -2$

Pyrochlore
$D_M/N = 1$

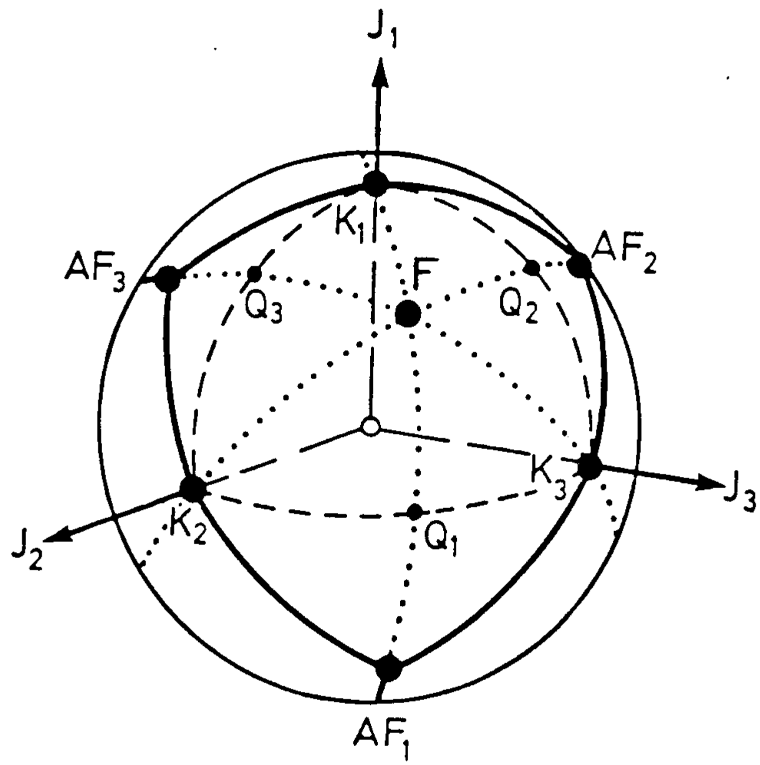
Tetrahedron
$D_M/N = 5$

The Major Frustrating Lattices



3rd Materials Specification – Isotropic Spin

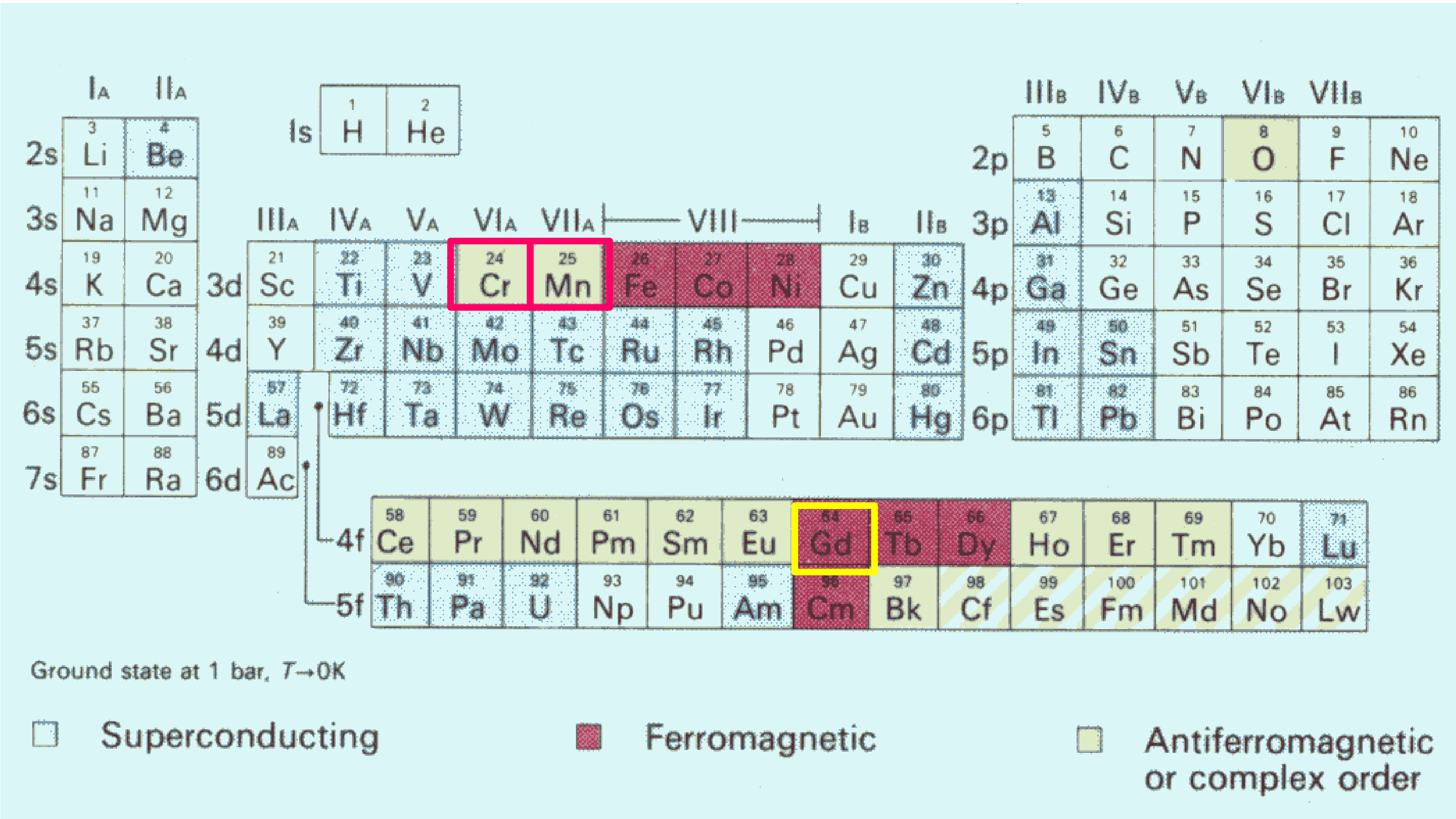
Frustration Point Achieved only with Most Isotropic Interactions



Liebmann, 1980

What are the Most Isotropic Spins*

* in octahedral crystal field



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Why Focus on Rare-Earth Spins?

Accessible phase diagrams –

$$\mathbf{H}_{\mathbf{Zeeman}} \text{ (achievable)} \sim \mathbf{J}_{\mathbf{nn}}$$

3*d*-spins – superexchange dominates – $10\text{ K} < \mathbf{J}_{\mathbf{nn}} < 1000\text{K}$

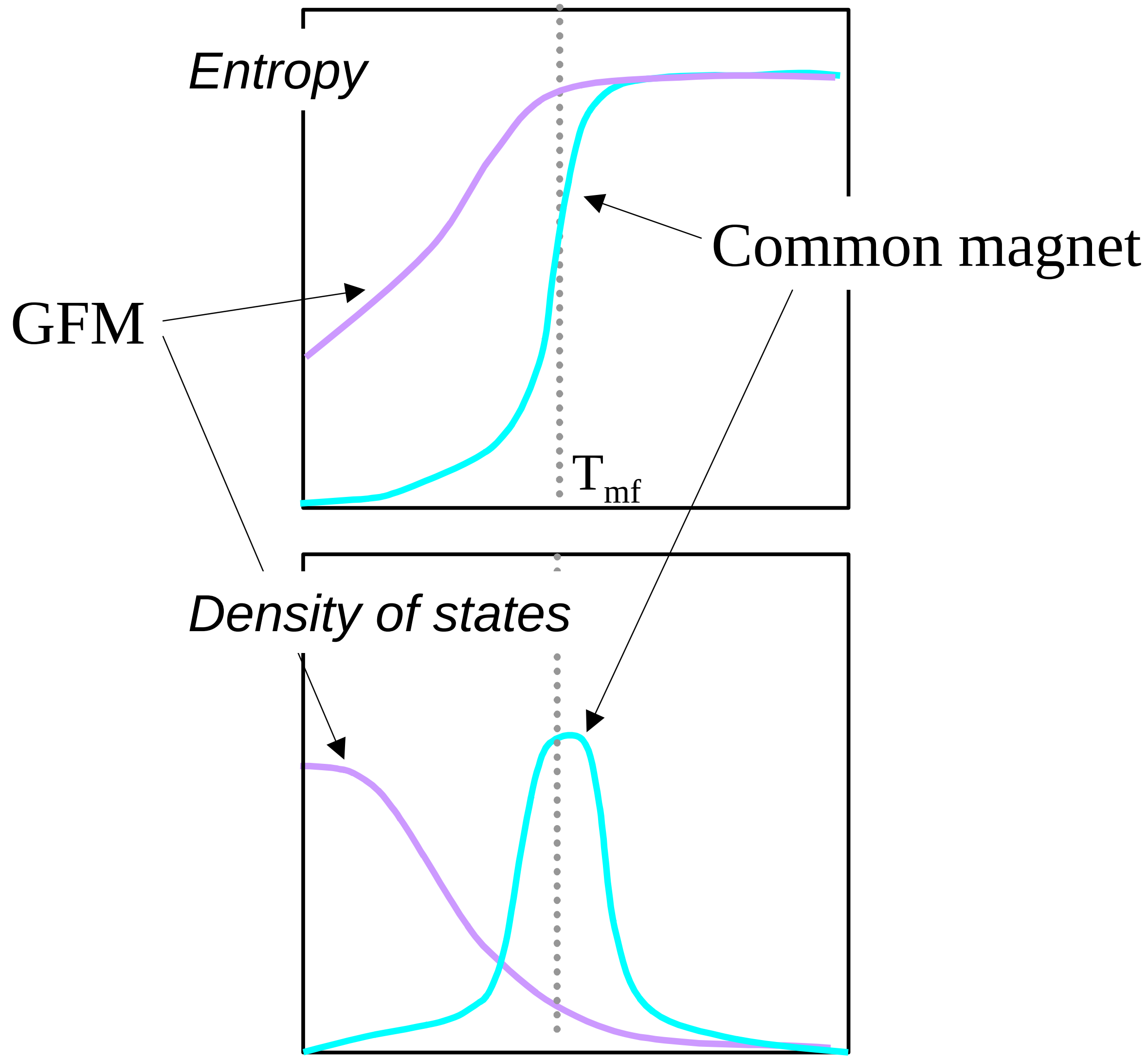
4*f*-spins – sup. exch. Or dipole-dipole – $0.1\text{K} < \mathbf{J}_{\mathbf{nn}} < 10\text{K}$

Table 1 Strongly Geometrically Frustrated Compounds

Compound	magnetic lattice	$-\theta_W$ (K)	T_c (K)	f	Order Type	Elect. Config.	Reference
2D MAGNETS							
VCl_2	triangular	437	36	12	AF	$3d^3$	(Hirakawa and al. 1983)
$NaTiO_2$	triangular	1000	<2	>500	—	$3d^1$	(Hirakawa, Kadowaki et al. 1985)
$LiCrO_2$	triangular	490	15	33	AF	$3d^3$	(Tauber, Moller et al. 1972)
$Gd_{0.8}La_{0.2}CuO_2$	triangular	12.5	0.7	16	SG	$4f^7$	(Ramirez, Jager-Waldau et al. 1991)
$SrCr_8Ga_4O_{19}$	kagome	515	3.5	150	SG	$3d^3$	(Ramirez, Espinosa et al. 1990)
$KCr_3(OH)_6(SO_4)_2$	kagome	70	1.8	39	AF	$3d^3$	(Townsend, Longworth et al. 1986)
3D MAGNETS							
$ZnCr_2O_4$	B-spinel	390	16	24	AF	$3d^3$	(Fiorani, Viticoli et al. 1983; Fiorani 1984; Fiorani, Viticoli et al. 1984; Fiorani, Dormann et al. 1985)
K_2IrCl_6	FCC	32.1	3.1	10	AF	$5d^5$	(Cooke, Lazenby et al. 1959)
FeF_3	pyrochlore	240	15	16	AF	$3d^5$	(DePape and Ferey 1986; Ferey, DePape et al. 1986)
$CsNiFeF_6$	pyrochlore	210	4.4	48	SG	$3d^8$,$3d^5$	(Alba, Hammann et al. 1982)
$MnIn_2Te_4$	zinc-blende	100	4	25	SG	$3d^5$	(Doll, Anghel et al. 1991)
$Gd_3Ga_5O_{12}$	garnet	2	0.1	20	SG	$4f^7$	(Hov, Bratsberg et al. 1980; Schiffer, Ramirez et al. 1994)
Sr_2NbFeO_6	perovskite	840	28	30	SG	$3d^4$	(Rodriguez and al. 1985)
$Gd_2Ti_2O_7$	pyrochlore	10	1.0	10	AF	$4f^7$	(Cashion, Cooke et al. 1968)

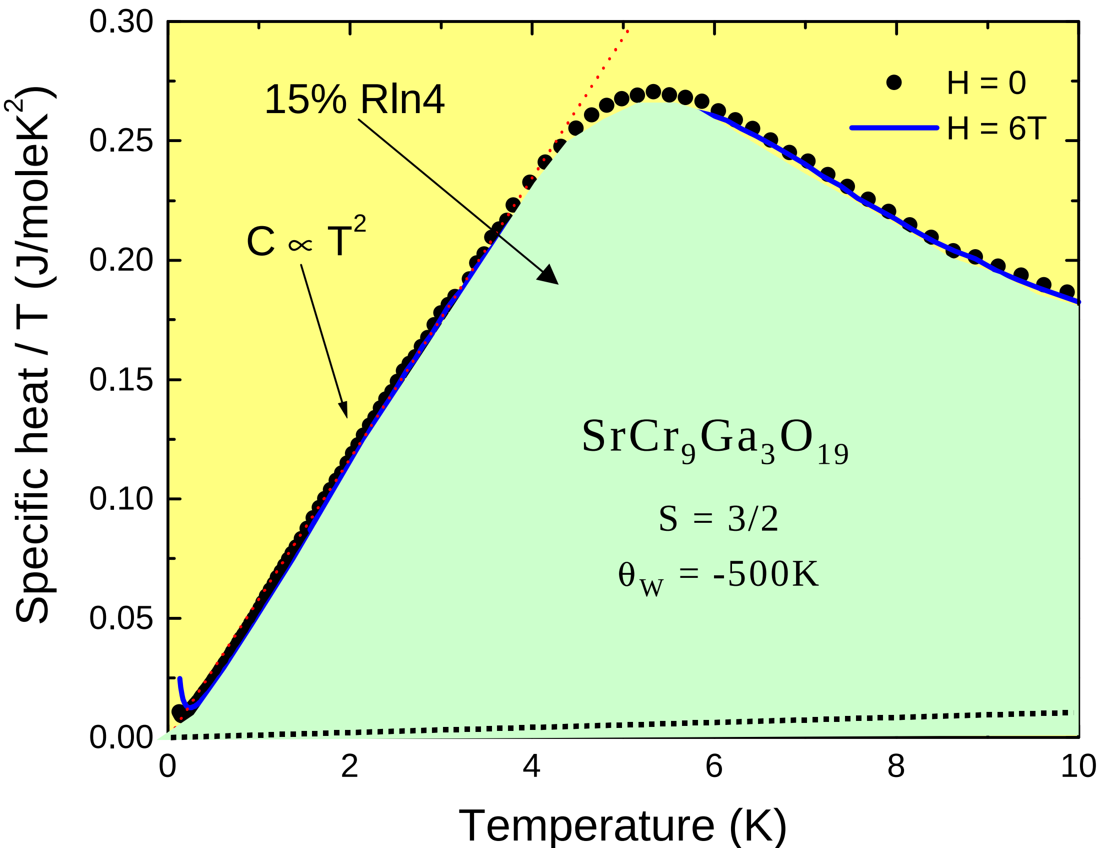
A. Ramirez, in Handbook of Magnetic Materials, 2001

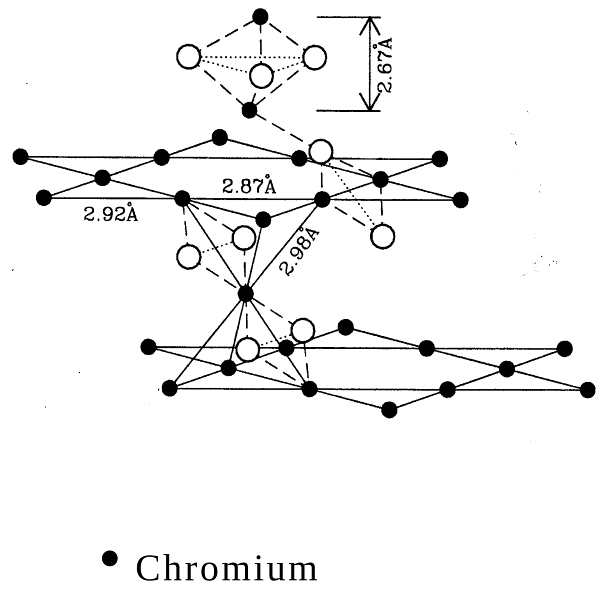
Hallmark of geometrical frustration - *Spectral Weight Downshift*



New Condensed Matter Physics From Frustrated Magnets

Spectral weight downshift in a kagome magnet - $SrCr_9Ga_3O_{19}$

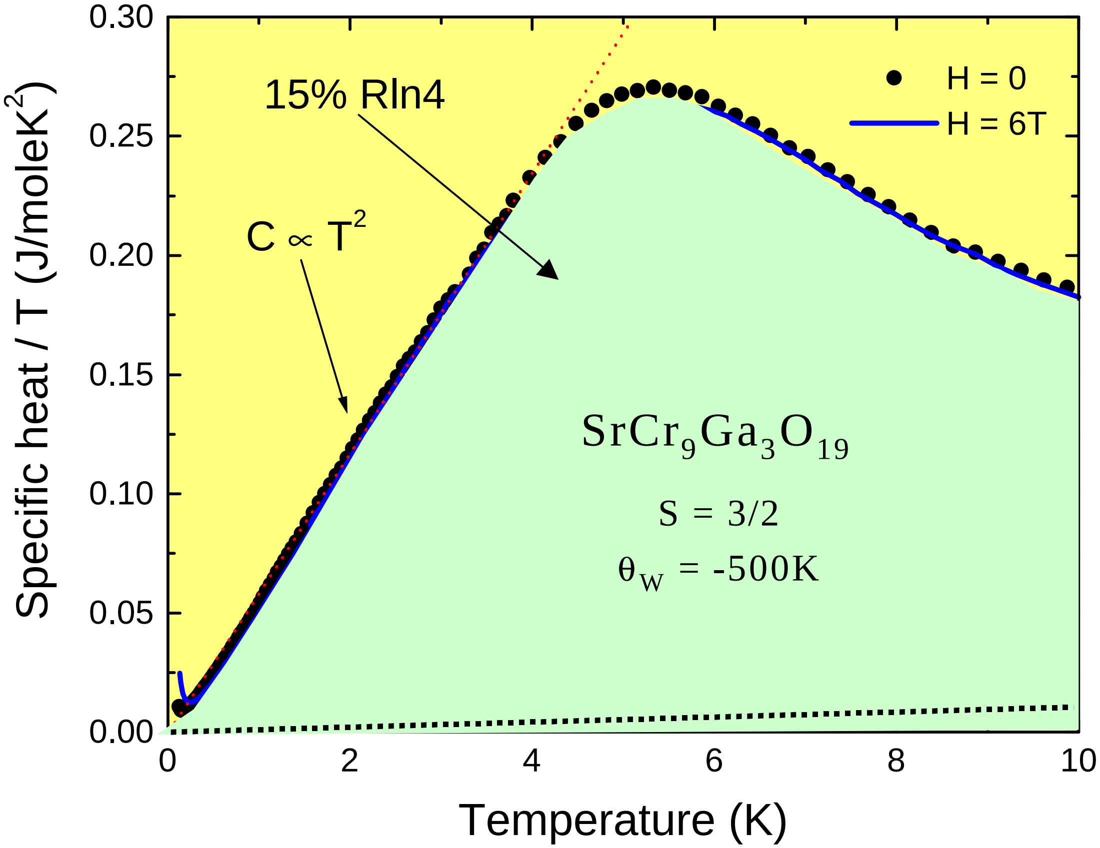




!! a strongly-interacting system !!

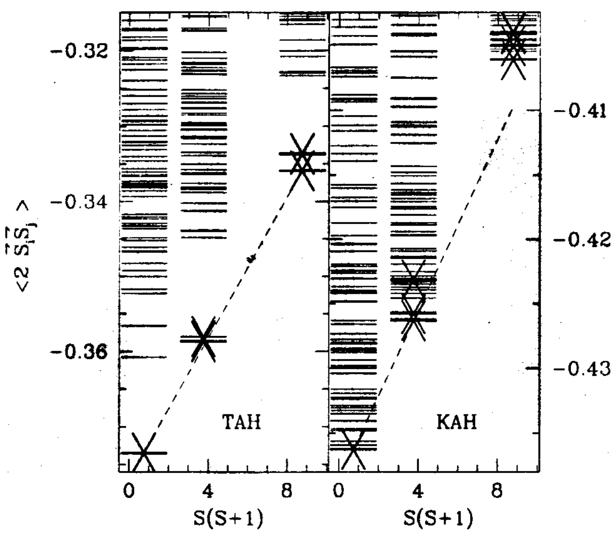
A. P. Ramirez et al, *Phys. Rev. Lett.*, (1990)

Spectral weight downshift in a kagome magnet - $SrCr_9Ga_3O_{19}$



A. Ramirez et al, *Phys. Rev. Lett.*, (1990)

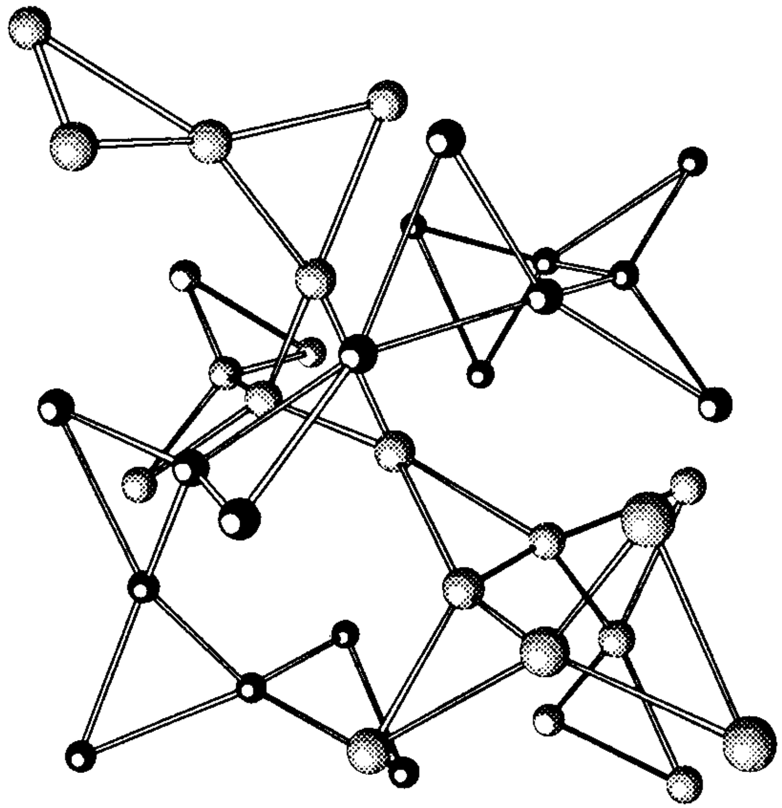
Exact diagonalization, N = 27



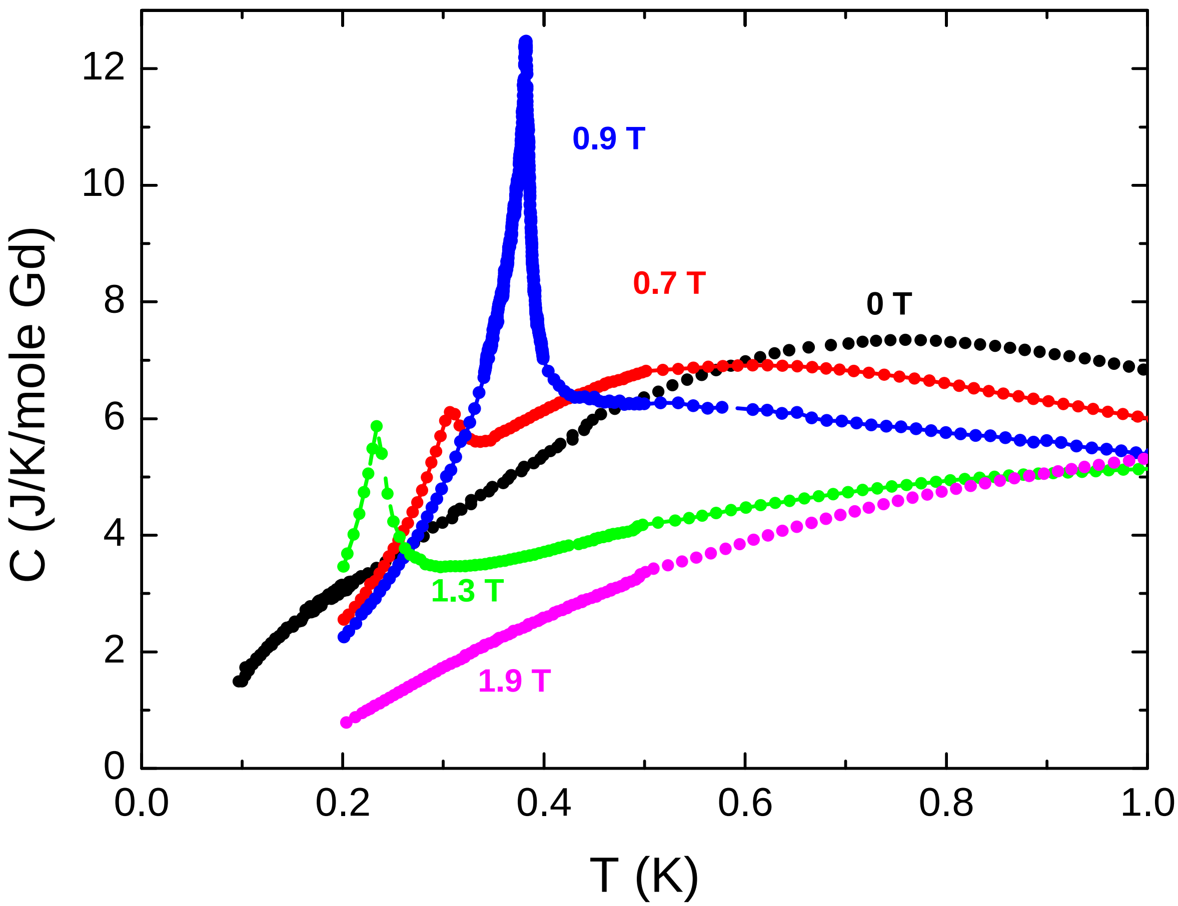
P. Sindzingre et al., *Phys. Rev. Lett.*, 2000

$Gd_3Ga_5O_{12}$ (GGG) – The *Hyper*kagome Structure

GGG – Two interpenetrating lattices of corner-sharing triangles of Gd

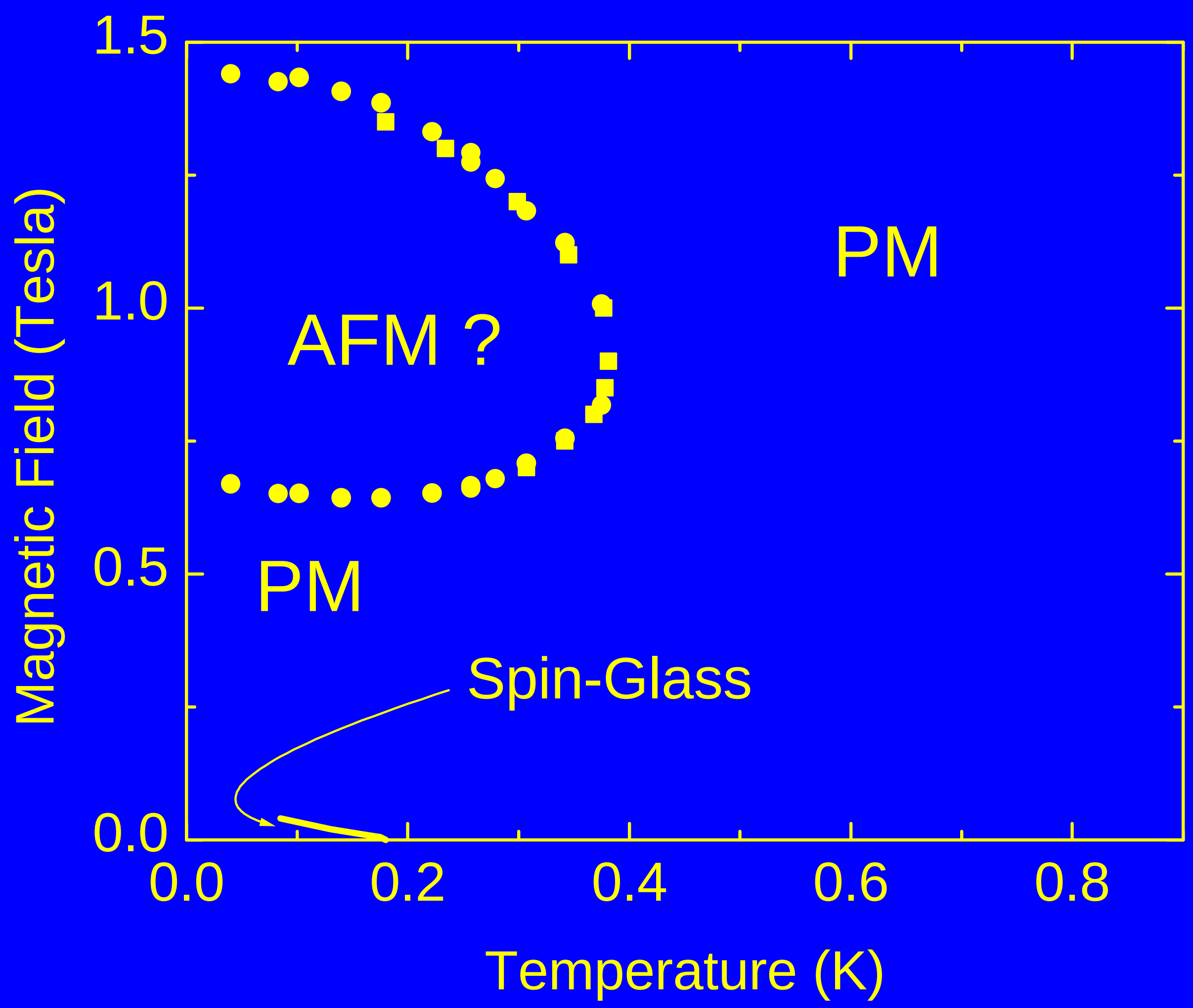


Specific Heat - GGG



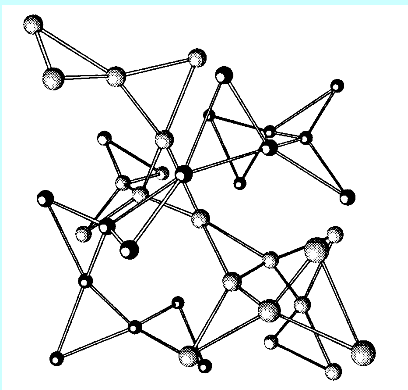
P. Schiffer et al., *Phys. Rev. Lett.* 1994

Resulting Phase Diagram - GGG

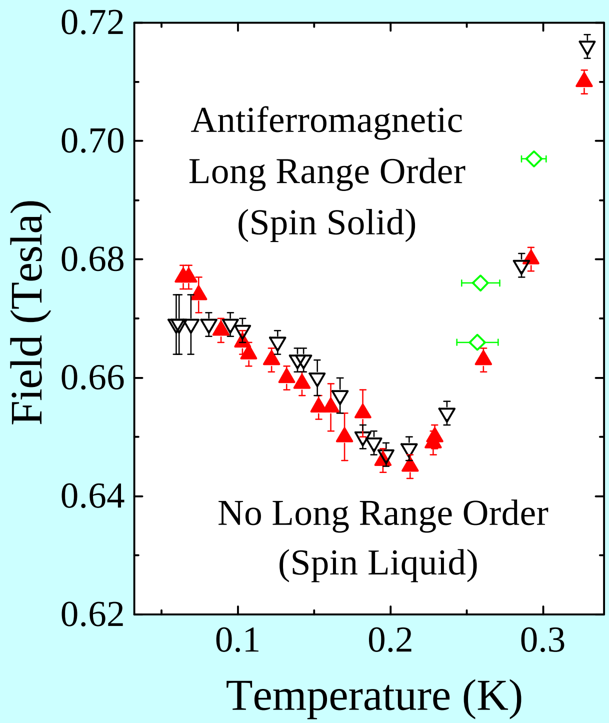


Loss of shear mode in GGG – Evidence of Spin Liquid

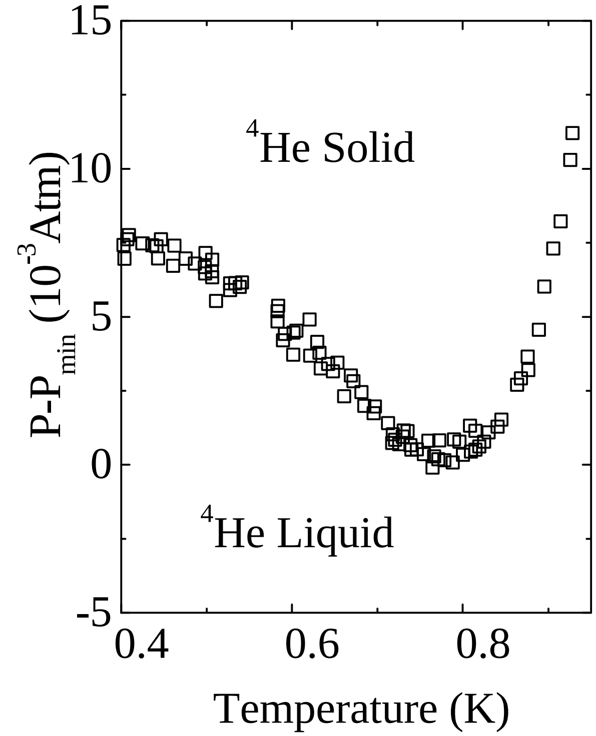
$Gd_3Ga_5O_{12}$



Garnet structure



Helium-4

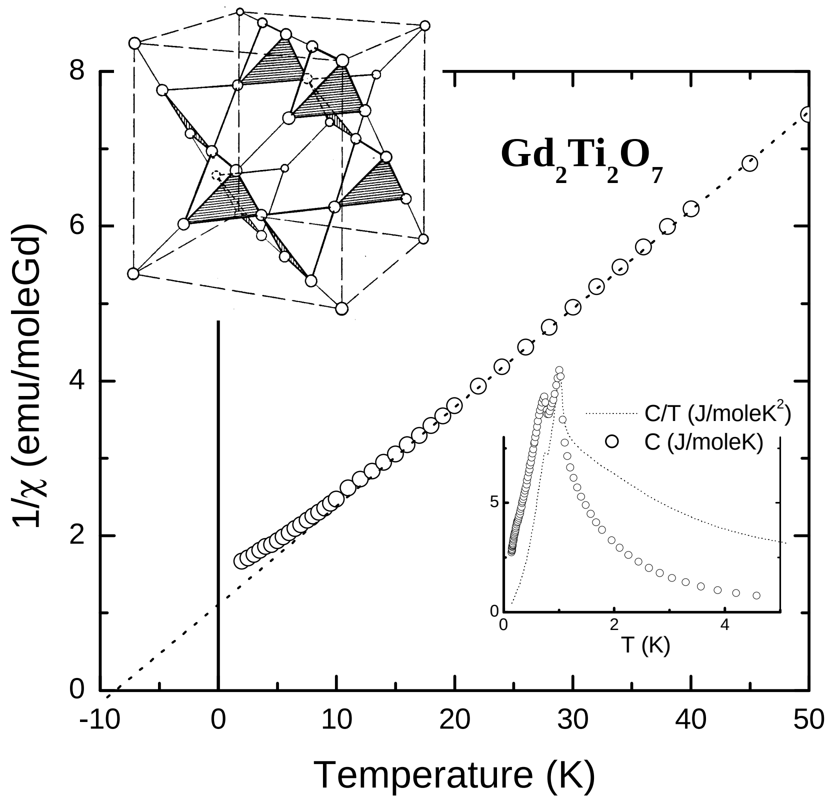


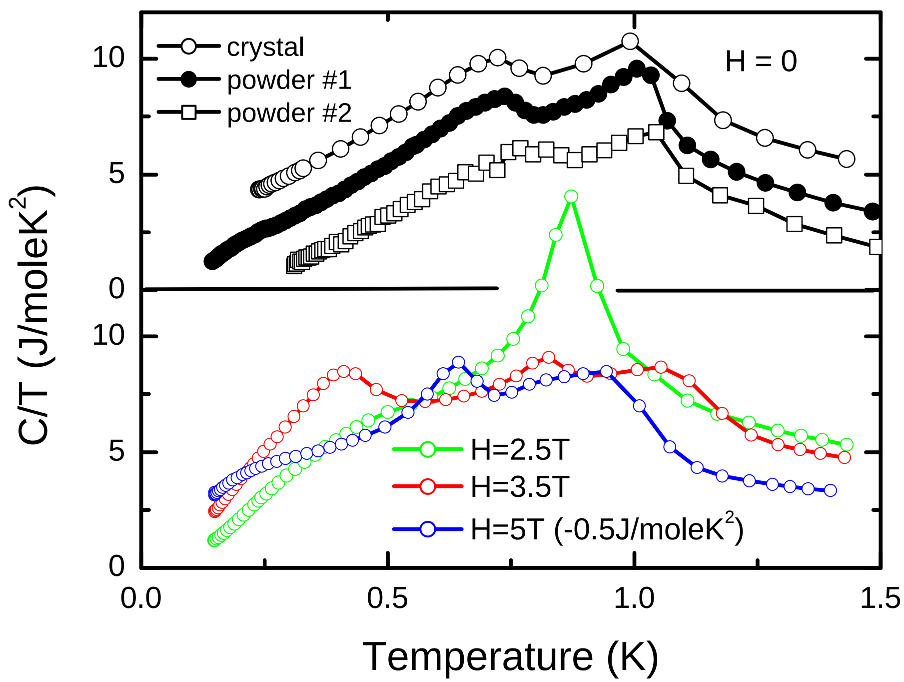
Possible spin liquid?

P. Schiffer et al., *Phys. Rev. Lett.*, 1994, 1999

$Gd_2Ti_2O_7$ – A Dipolar Heisenberg Pyrochlore

Magnetic Field/GF Competition in a Dipolar Pyrochlore Magnet $Gd_2Ti_2O_7$

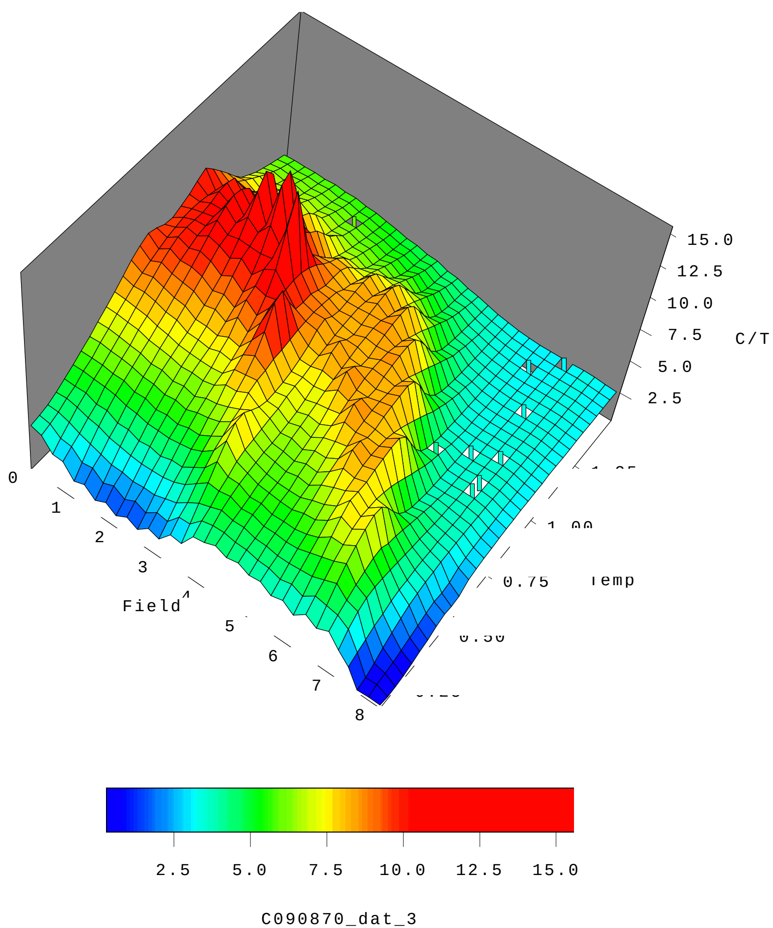




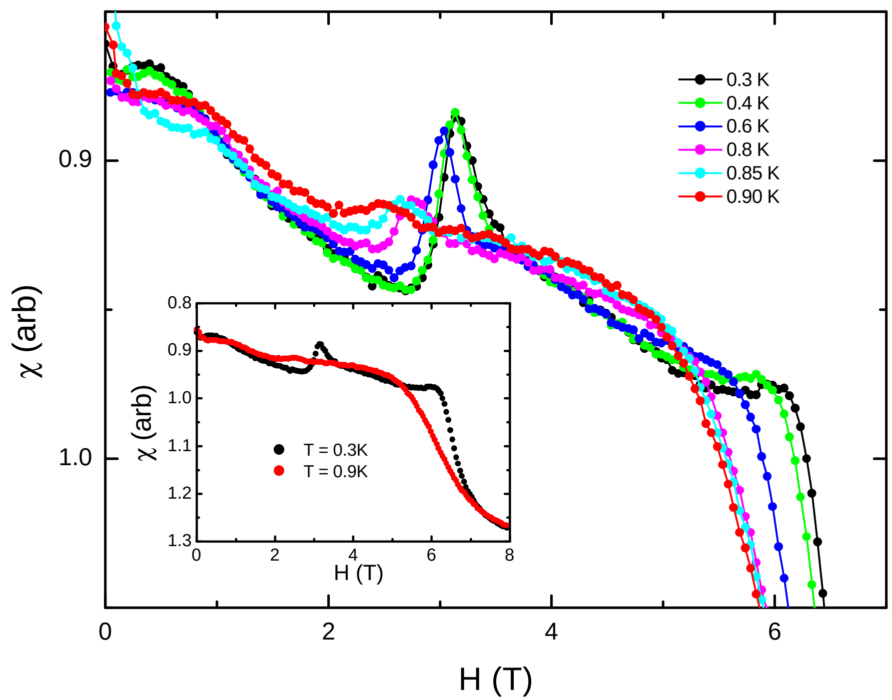
A. Ramirez et al., Phys. Rev. Lett., 2002

$Gd_2Ti_2O_7$

Specific Heat



Magnetic Susceptibility



A. Ramirez et al., Phys. Rev. Lett., 2002

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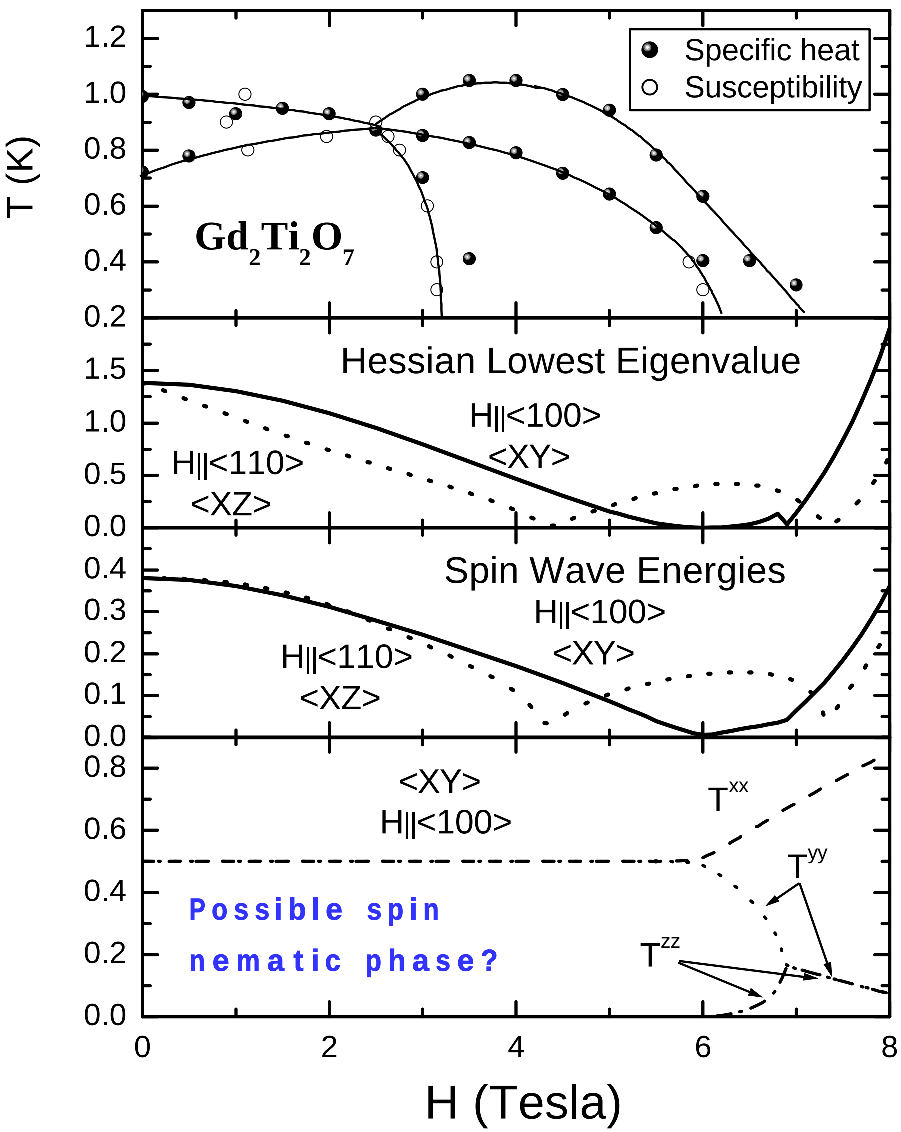
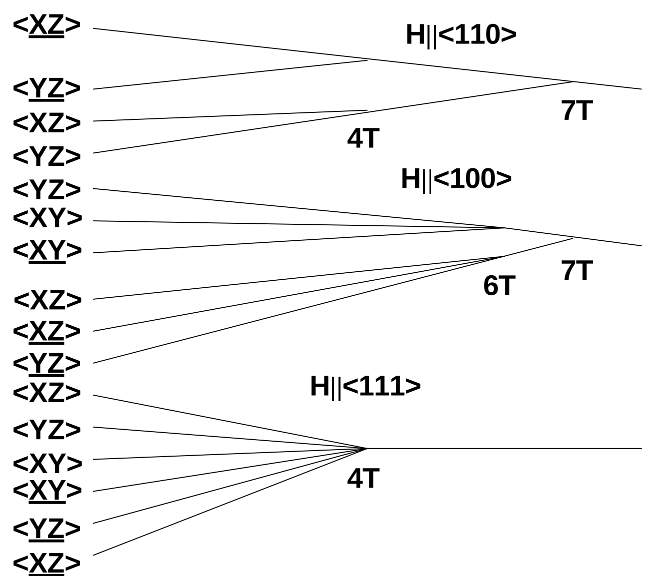
Phase Diagram – polycrystalline $Gd_2Ti_2O_7$



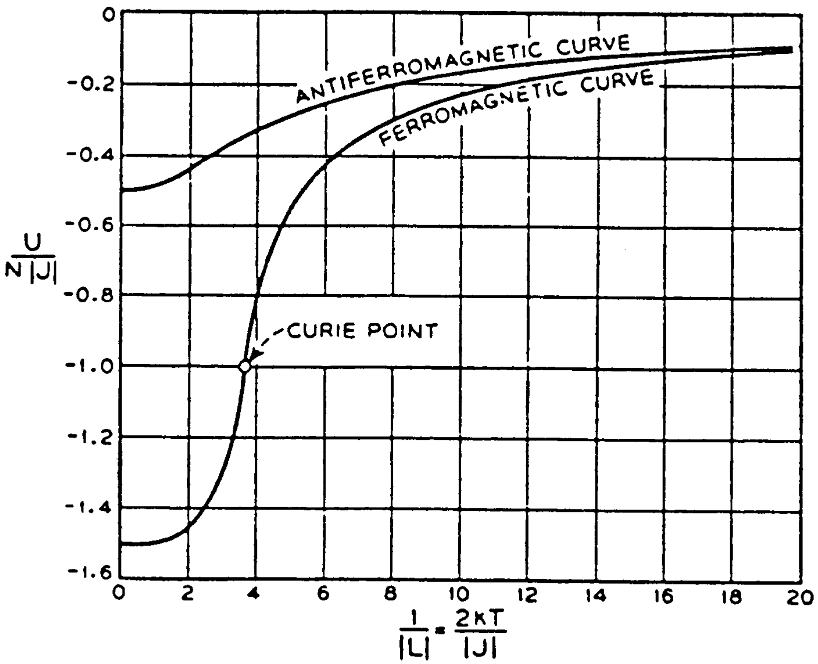
$$4F/N = +JS^2 \sum_{j,k}' \vec{\eta}(j)\cdot\vec{\eta}(k) - g\mu_B \sum_j \vec{\eta}(j)\cdot\vec{B} + (g\mu_B S)^2/(2a^3) \sum_{i\neq j} \sum_{a,b=x,y,z} \eta^a(i) D^{ab}(i,j)\eta^b(j)$$

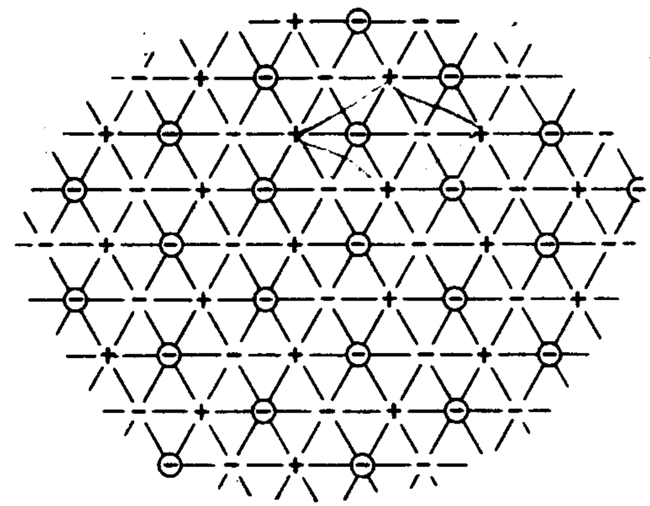


Ramirez et al., Phys. Rev. Lett., 2002

$Dy_2Ti_2O_7$ – An Ising Pyrochlore

Geometrical Frustration for 2D Ising spins





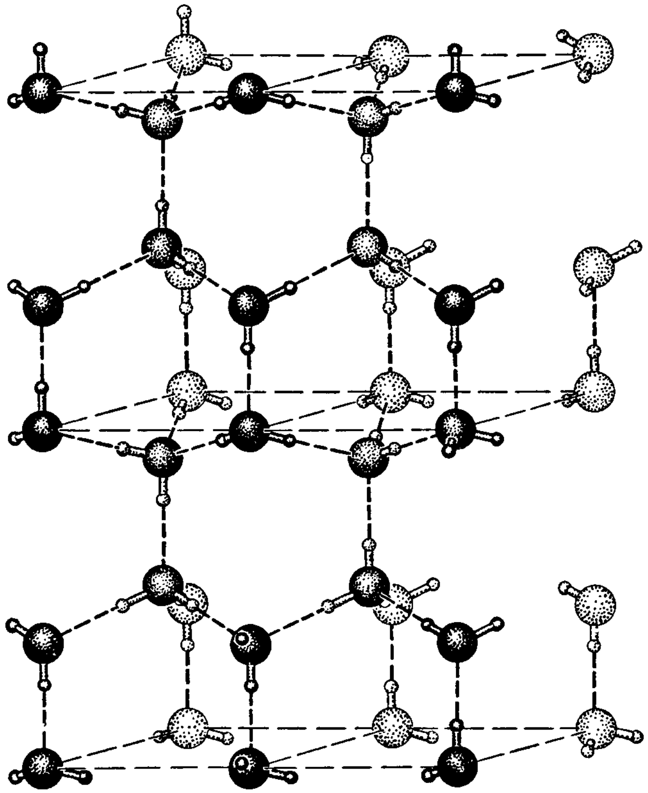
For the Ising triang. AF,

$S_0 = 0.323R = 0.47S_\infty$

Wannier, PR **79**, 357 (1950)

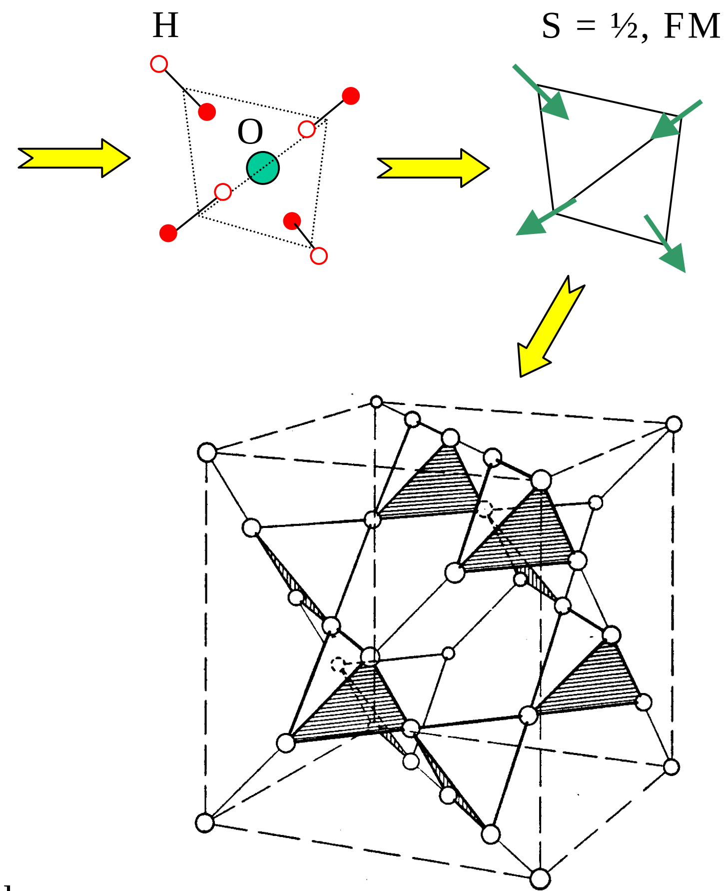
Houtappel, Physica, **16**, 425 (1950).

Example of Geometrical Frustration – Ice & Spin Ice



Pauling, The Nature of the Chemical Bond

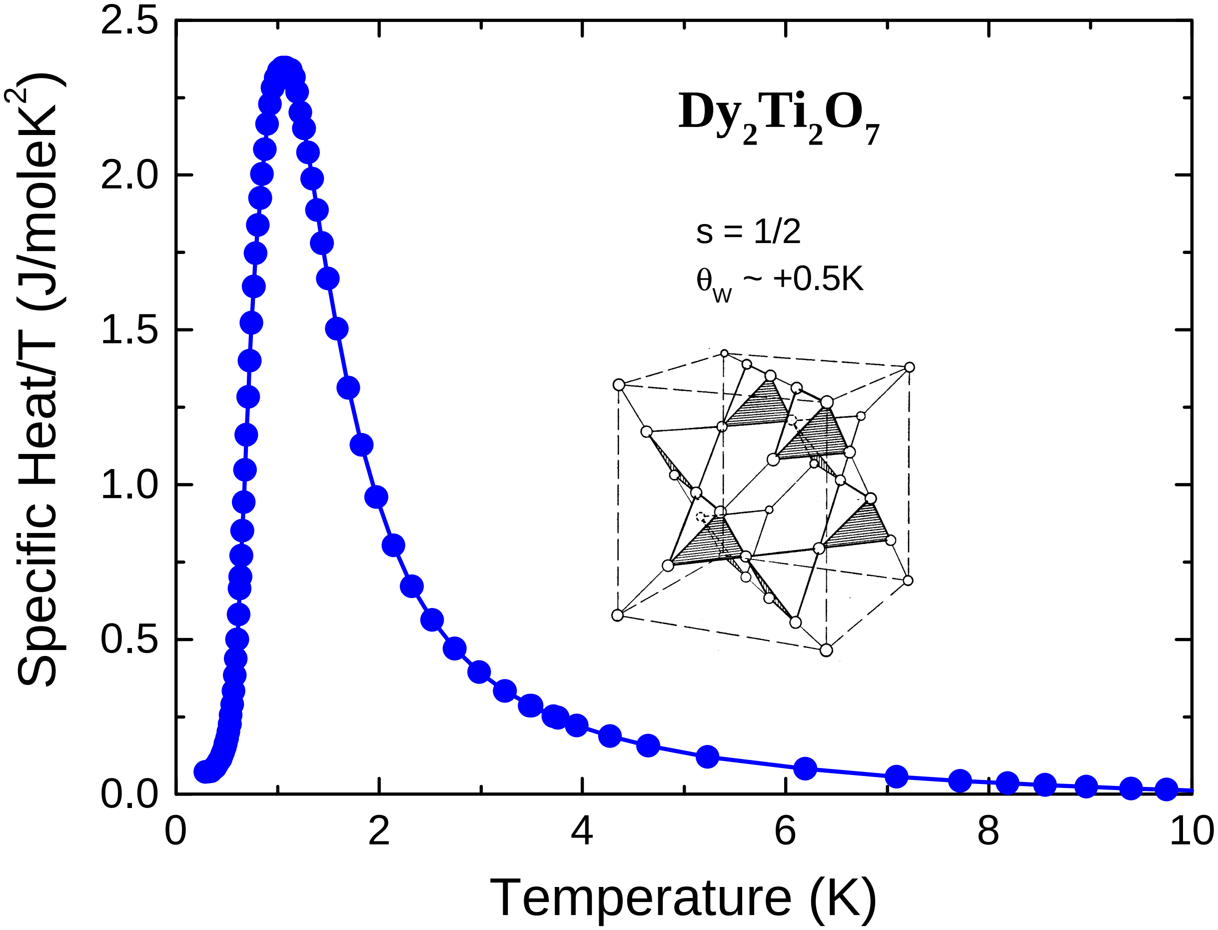
$S_0 = R\ln 3/2$



B-spinel, or pyrochlore

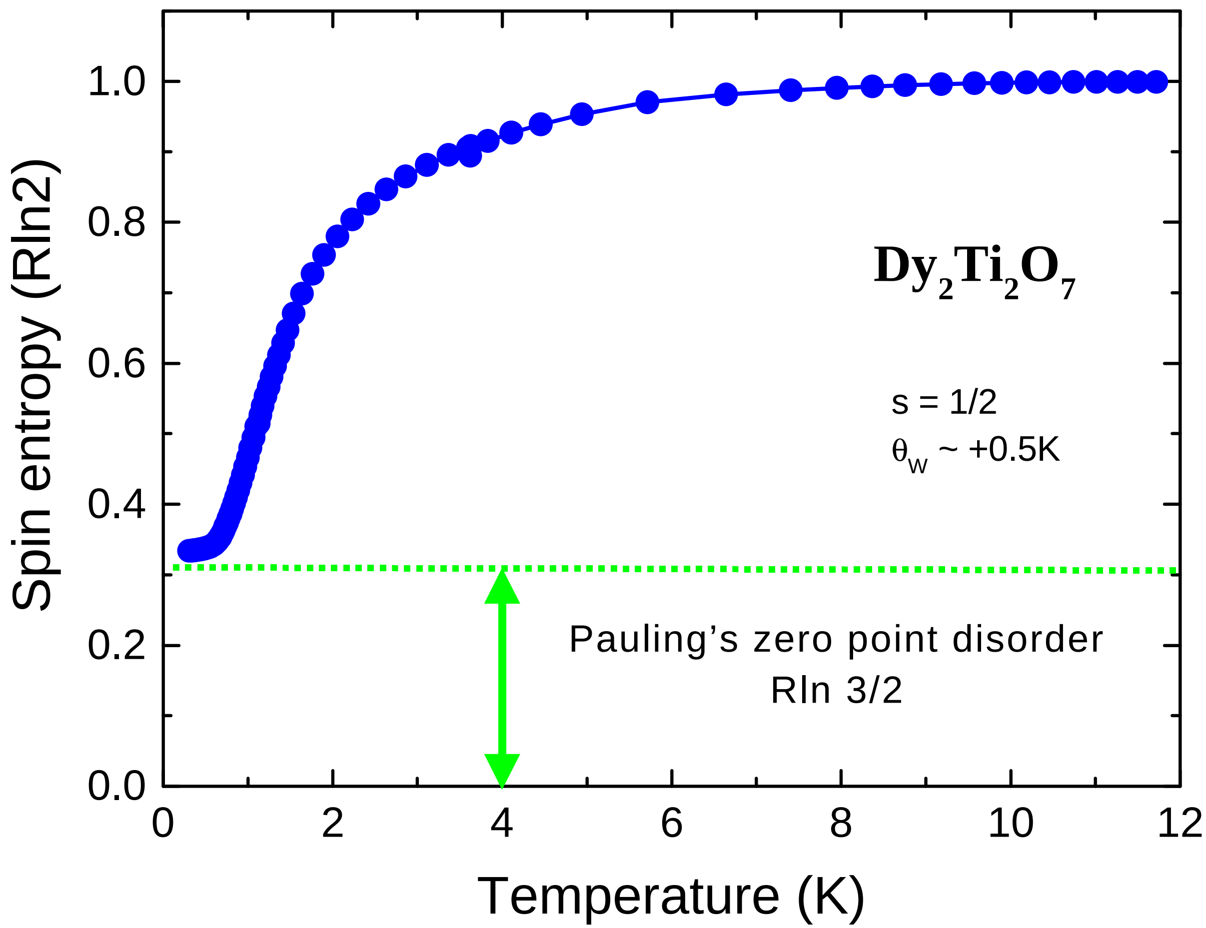
M. Harris, S. Bramwell, et al. PRL, 1997

Specific heat - Ising s = 1/2 pyrochlore with FM interactions



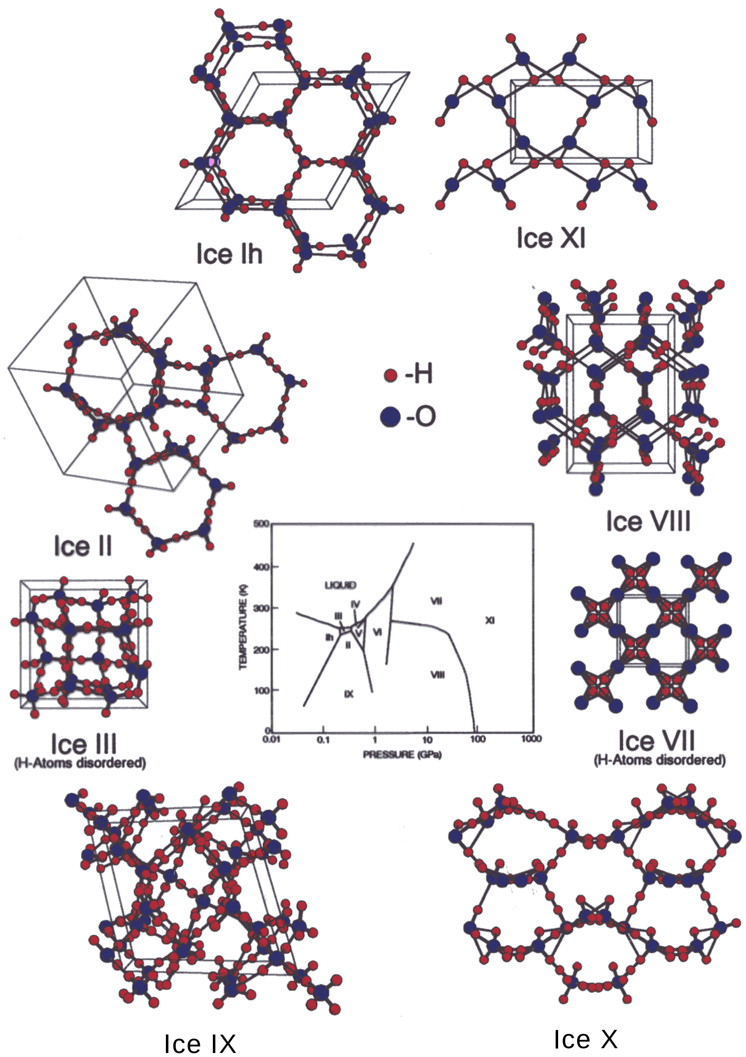
Observation of Zero Point disorder in *Spin Ice*

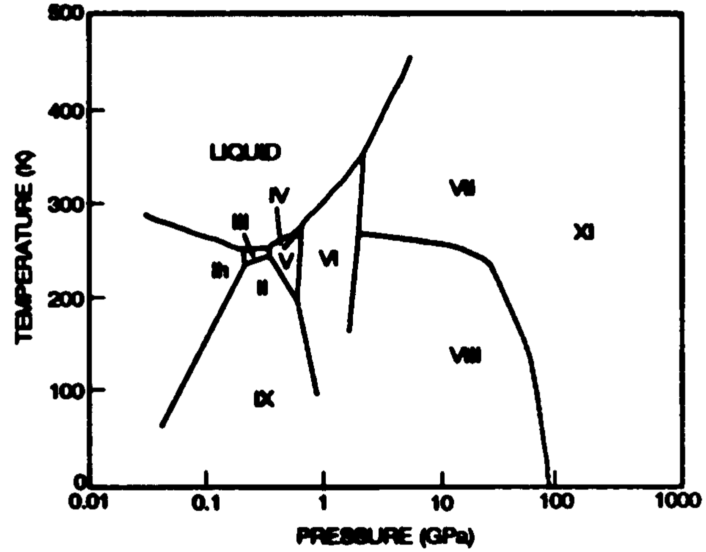
$Dy_2Ti_2O_7$



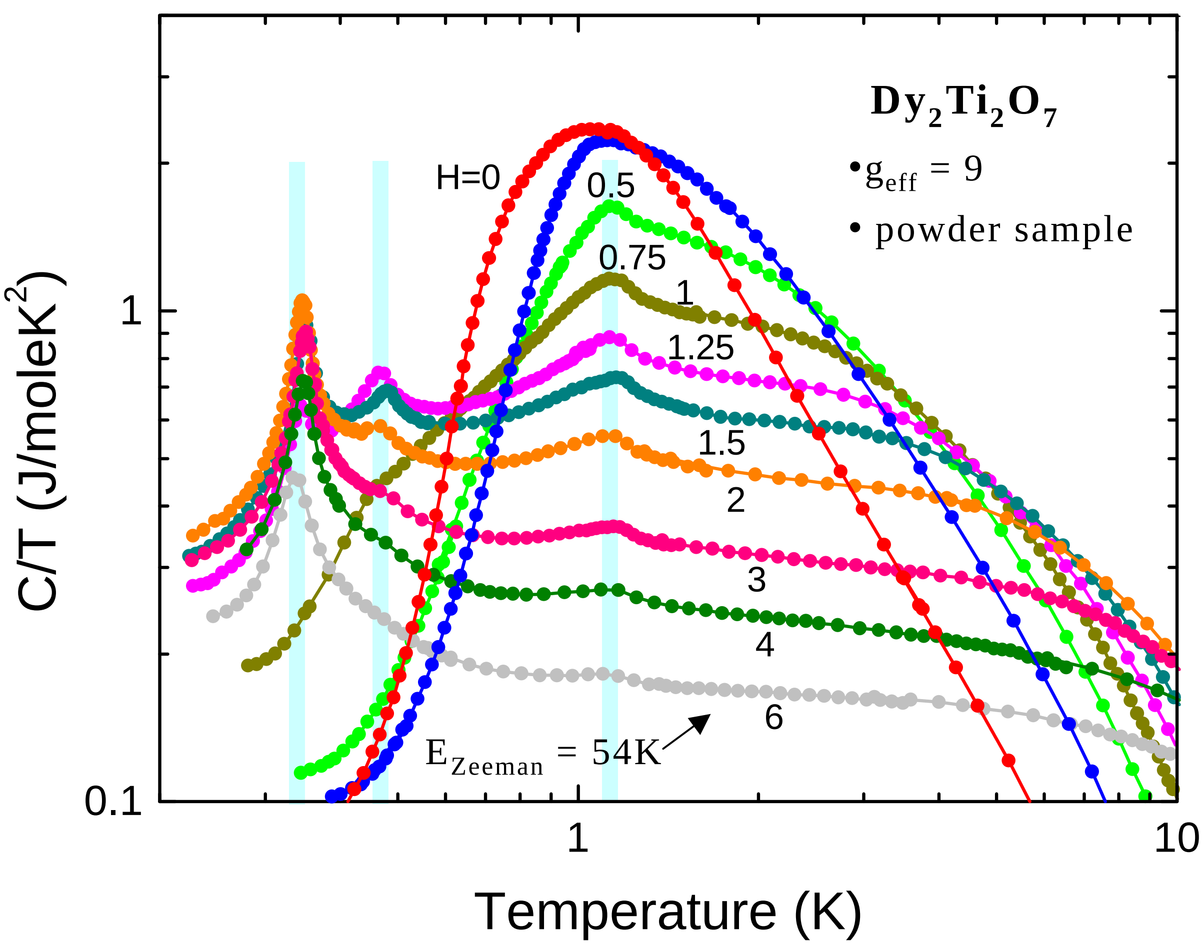
A. Ramirez et al., Nature, 1999

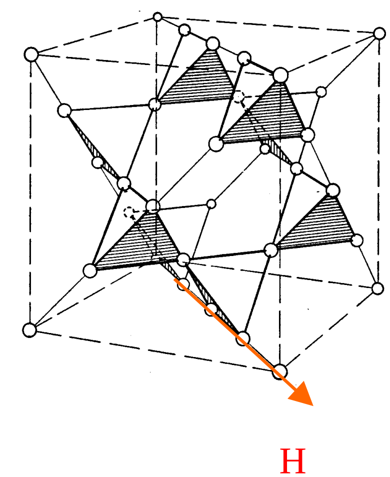
Crystal Structures of H_2O





High-field behavior of specific heat





A. Ramirez et al., *Nature*, 1999

Conclusions

• **Completely new and poorly understood phases of strongly correlated matter exist in GFMs:**

spin-liquid
spin-nematic
spin-ice

• **These states, and field-probes dictate study of Gd, Dy compounds.**

• **Neutrons are inadequate – need to measure same samples used in bulk measurements.**

\ Need to look at other techniques – magnetic x-rays?

Collaborators

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