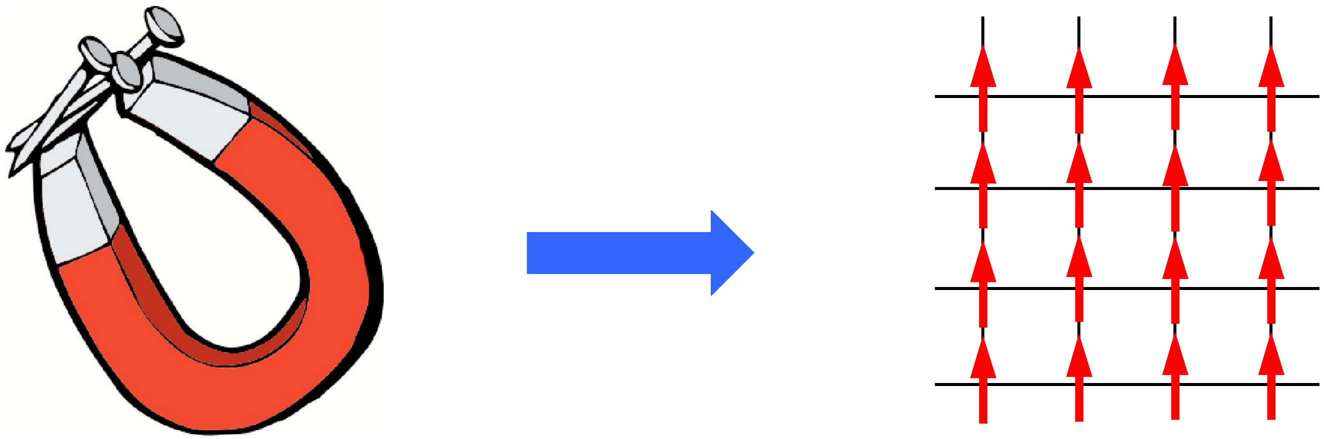
Geometrical Frustration In Solids

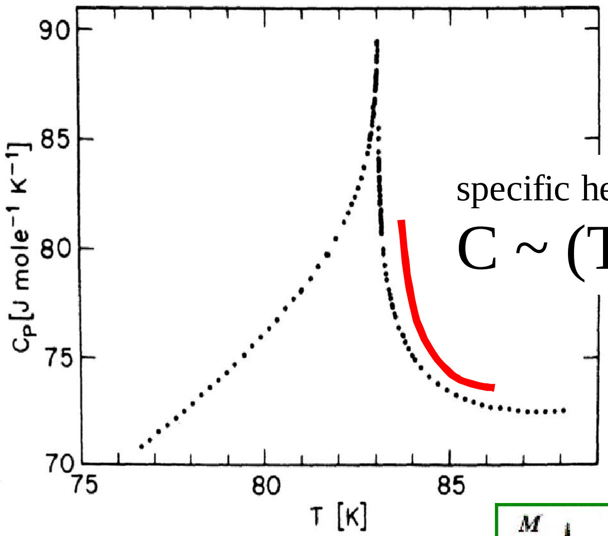
Understanding the ground state from the local interactions



ferromagnet

Try to describe matter with a single energy scale

Critical Phenomena at phase transitions - universality

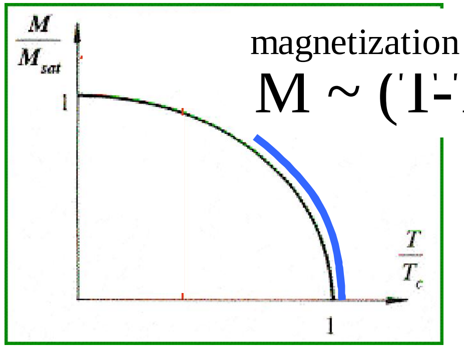


Specific heat of $RbMnF_3$
Kornblit and Ahlers, 1973

specific heat

$$C \sim (T-T_c)^{-\alpha}$$

Transition	Order parameter
Ferromagnet	magnetization
Superconductor	Δ – gap parameter
Liquid-Gas	$\rho-\rho_c$
Ferroelectric	polarization



magnetization

$$M \sim (T-T_c)^{\beta}$$

Critical exponents, α, β, ν, … depend on spatial dimensionality, spin dimensionality

correlation length

$$\xi \sim (T-T_c)^{-\nu}$$

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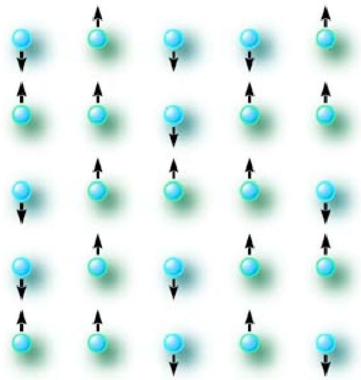
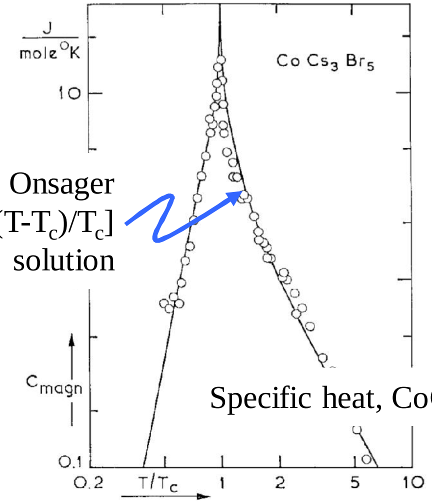
Only one analytic solution for critical exponents

…in 2D

Lars Onsager



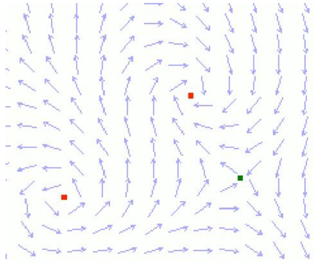
2D square Ising model
1944



Onsager ln [(T-T_c)/T_c] solution

Specific heat, $CoCs_3Br_5$

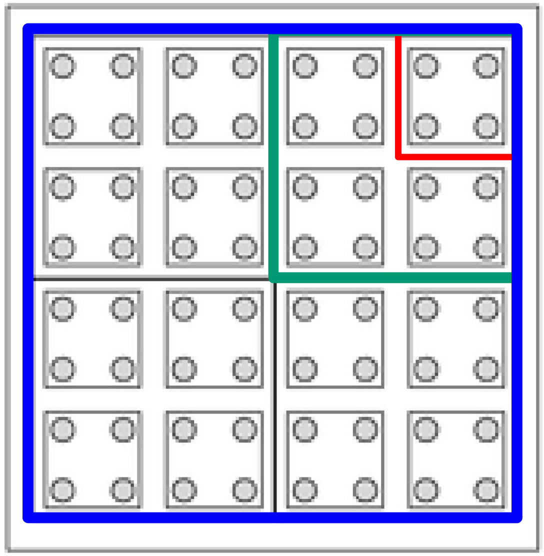
…in 3D



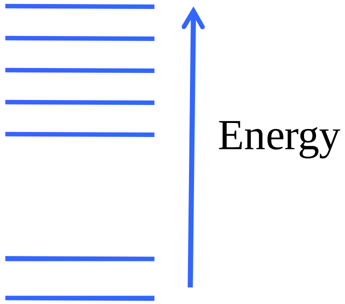
Exponent (3D)	Heisenberg	Ising
α (spec ht)	-0.14	0.013
β (magnetization)	0.38	0.31
ν (correl length)	0.70	0.64

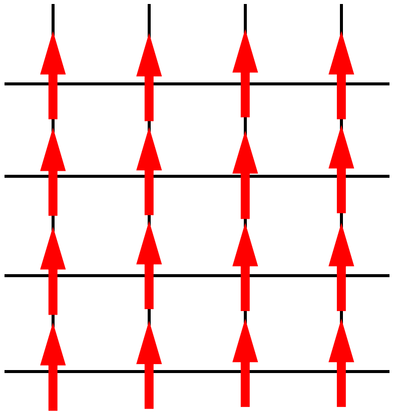
Critical Phenomena and scale invariance

Ken Wilson
b. 1936
Nobel Prize 1982
Renormalization Group



Kadanoff block spin contruction



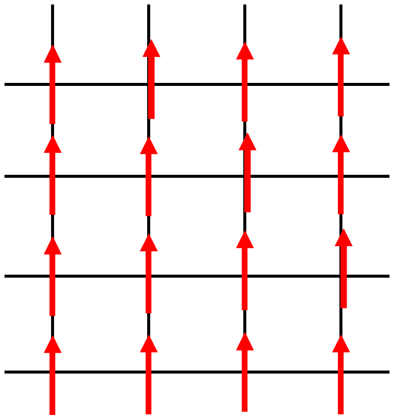


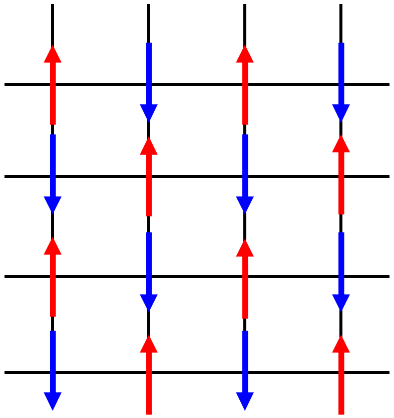
ferromagnet

Antiferromagnetism – not *quite* so simple

→

→





Antiferromagnetism sees the lattice, no need for compatibilty

Louis Néel
1904-2000
Nobel Prize 1970

Antiferromagnets vs. Ferromagnets – easy to distinguish

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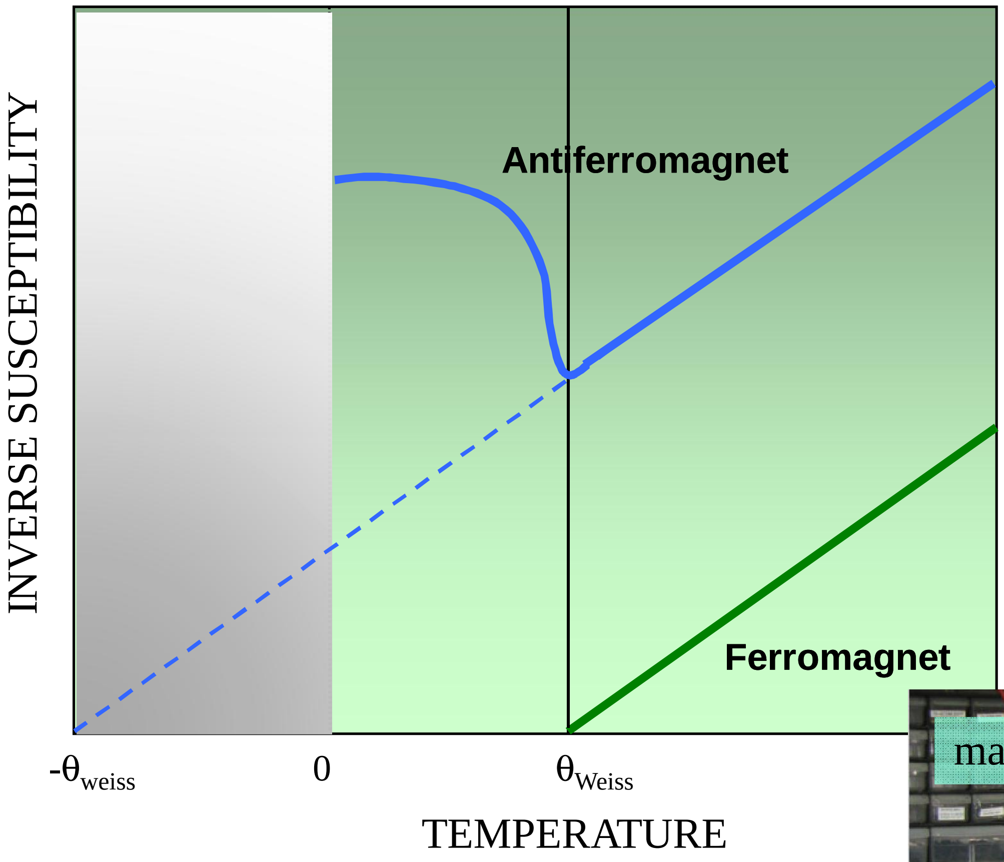
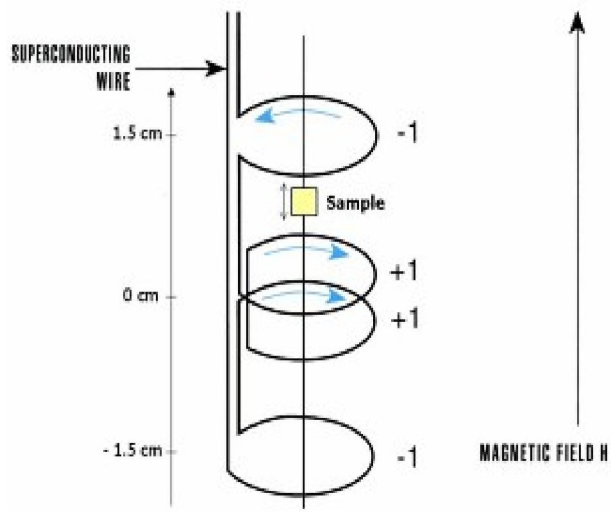
$$1/\chi \sim T - \underbrace{zSJ}_{\theta_{Weiss}}$$





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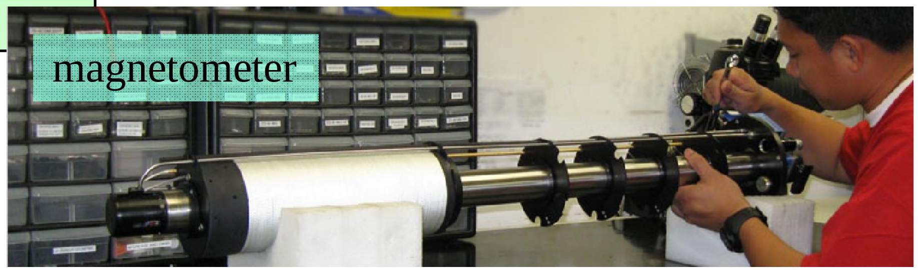


Antiferromagnets – Deviation from MF-T_c is common

Row no.	Substance	Chem. structure	Crystal sym. $T > T_N$	Mag. cat. structure	n_{eff}	T_N, °K	$-\theta_a/T_N$
1	VO	Rock salt	Cubic	f.c.c.	( )[a]	117	
2	CrN	Rock salt	Cubic	f.c.c.	( )[a]	~273	
3	MnO	Rock salt	Cubic	f.c.c.	5.95	122	5.0
4	α-MnS	Rock salt	Cubic	f.c.c.	5.6	130	3.1
5	β-MnS	Zinc blende	Cubic	f.c.c.	5.82	160	6.1
6	MnSe	Rock salt	Cubic	f.c.c.	5.7	~173	2.1
7	$Li_{0.1}Mn_{0.9}Se$	Rock salt	Cubic	f.c.c.	4.76	71[b]	−0.8
8	FeO	Rock salt	Cubic	f.c.c.	4.0[d]	198	~1.0[d]
9	CoO	Rock salt	Cubic	f.c.c.	5.1	291	1.1
10	NiO	Rock salt	Cubic	f.c.c.	4.6	520[c]	~5
11	TbP	Rock salt	Cubic	f.c.c.		9	
12	ErP	Rock salt	Cubic	f.c.c.		3.1	
13	TbAs	Rock salt	Cubic	f.c.c.		12	
14	TbSb	Rock salt	Cubic	f.c.c.	9.9	14	
15	HoSb	Rock salt	Cubic	f.c.c.		9	
16	ErSb	Rock salt	Cubic	f.c.c	9.8	3.7	
17	γ-Mn	f.c.c.	Cubic	f.c.c.		660	
18	MnS_2	Pyrite	Cubic	f.c.c.	6.30	<77	>8
19	$MnSe_2$	Pyrite	Cubic	f.c.c.	5.93	~100	~4.8
20	$MnTe_2$	Pyrite	Cubic	f.c.c	6.22	80	6.5
20a	FeS_2	Pyrite	Cubic	f.c.c.			
20b	CoS_2	Pyrite	Cubic	f.c.c.	1.85	$T_c = 110$	
20c	NiS_2	Pyrite	Cubic	f.c.c.	3.19		
21	CrF_2	Dist. rut.	Mono.	b.c. mono.	4.9	53	
22	$CrCl_2$	Dist. rut.	Ortho.	b.c. ortho.	5.1	40[h]	2.7
23	MnF_2	Rut.	Tet. ($c/a < 1$)	b.c. tet.	5.7	72	1.6
24	FeF_2	Rut.	Tet. ($c/a < 1$)	b.c. tet.	5.6	79	1.5
25	CoF_2	Rut.	Tet. ($c/a < 1$)	b.c. tet.	5.13	37	1.4
25a	CuF_2	Dist. rut.	Mono.	b.c. mono.		78	
26	NiF_2	Rut.	Tet. ($c/a < 1$)	b.c. tet.	3.5	78.5–83	~2.0
27	VO_2	Rut.	Tet. ($c/a < 1$)	b.c. tet.	1.73	343	2.1

from J. B. Goodenough, “Magnetism and the Chemical Bond”,

Some systems do not (or cannot) order

(at Temperature > 0)

- glass
- 1-dimensional systems
- systems below percolation threshold
- systems at high magnetic field

PHYSICAL REVIEW VOLUME 79, NUMBER 2 JULY 15, 1950

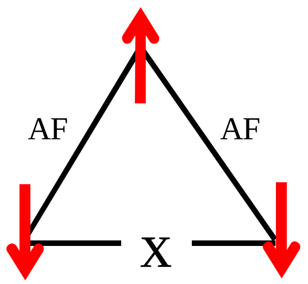
Antiferromagnetism. The Triangular Ising Net

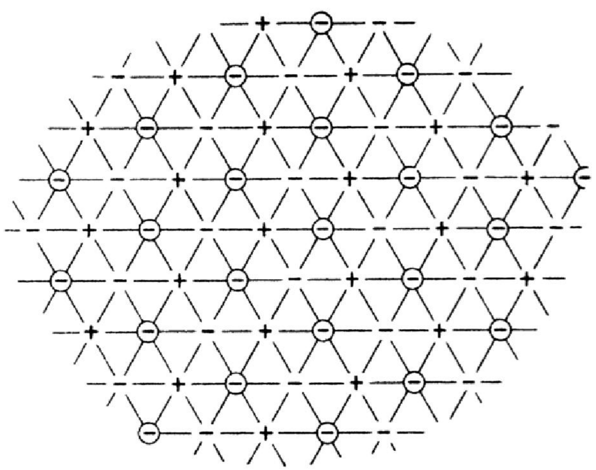
G. H. WANNIER
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received February 11, 1950)

In this paper the statistical mechanics of a two-dimensionally infinite set of Ising spins is worked out for the case in which they form either a triangular or a honeycomb arrangement. Results for the honeycomb and the ferromagnetic triangular net differ little from the published ones for the square net (Curie point with logarithmically infinite specific heat). The triangular net with antiferromagnetic interaction is a sample case of antiferromagnetism in a non-fitting lattice. The binding energy comes out to be only one-third of what it is in the ferromagnetic case. The entropy at absolute zero is finite; it equals

$$S(0)=R\frac{2}{\pi}\int_0^{\pi/3}\ln(2\cos\omega)d\omega=0.3383R.$$

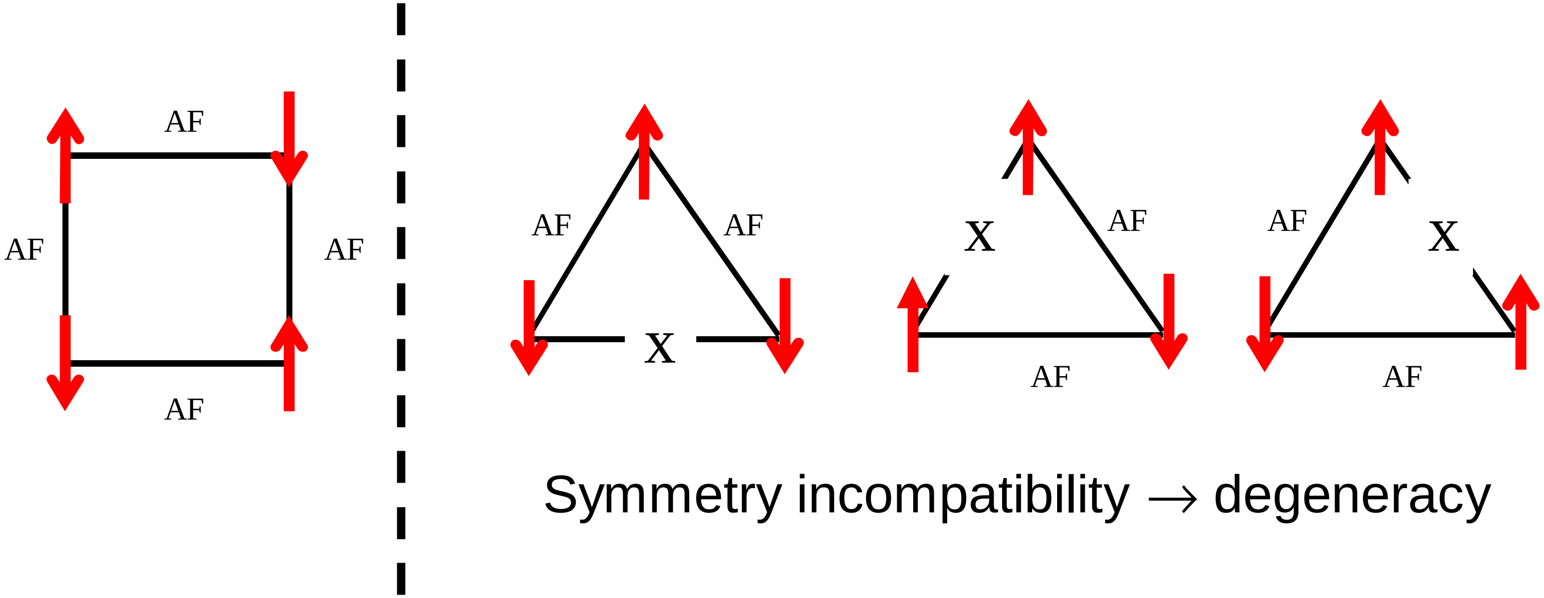
The system is disordered at all temperatures and possesses no Curie point.





Bell Labs photo book 1960

When the local interactions give no clue to the macroscopic ground state: "geometrical frustration"



Symmetry incompatibility → degeneracy

Kadanoff block construction won't work

Wannier's influence on Phil Anderson

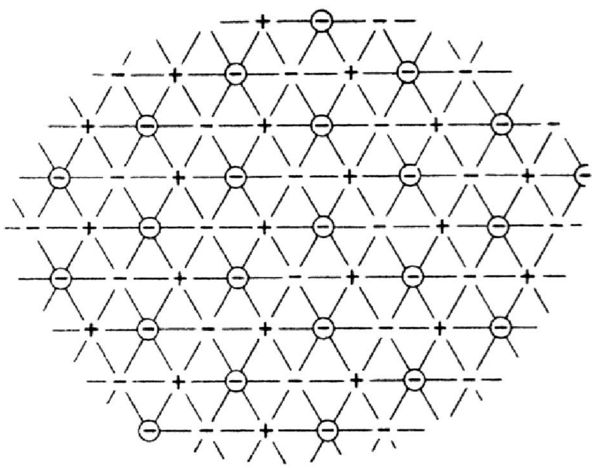
PHYSICAL REVIEW VOLUME 102, NUMBER 4 MAY 15, 1956

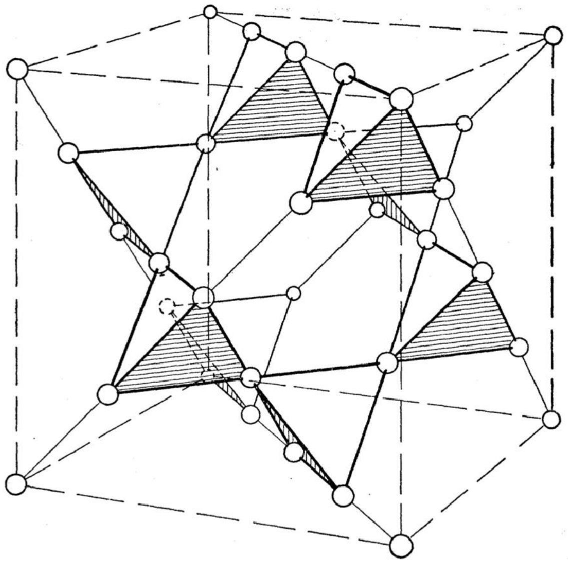
Ordering and Antiferromagnetism in Ferrites

P. W. Anderson
Bell Telephone Laboratories, Murray Hill, New Jersey

(Received January 9, 1956)

The octahedral sites in the spinel structure form one of the anomalous lattices in which it is possible to achieve essentially perfect short-range order while maintaining a finite entropy. In such a lattice nearest-neighbor forces alone can never lead to long-range order, while calculations indicate that even the long-range Coulomb forces are only 5% effective in creating long-range order. This is shown to have many possible consequences both for antiferromagnetism in "normal" ferrites and for ordering in "inverse" ferrites.





(a)

Anderson, P. W. 1111

Bell Labs photo book 1960

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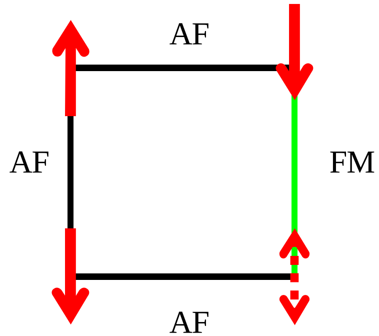
Evolution of frustration due to Anderson



THE CONCEPT OF FRUSTRATION IN SPIN GLASSES*

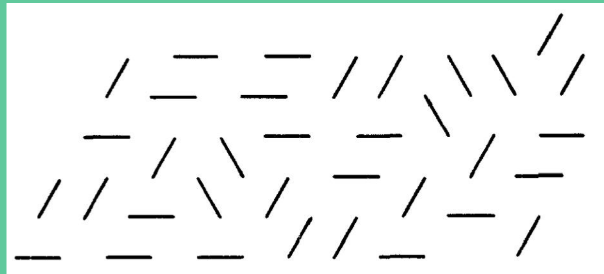
P. W. ANDERSON

Bell Telephone Laboratories Incorporated, 600 Mountain Avenue, Murray Hill, N. J. 07974 and Princeton University, Princeton, N.J. 08540 (U.S.A.)

(Received June 19, 1978)

RESONATING VALENCE BONDS: A NEW KIND OF INSULATOR?*

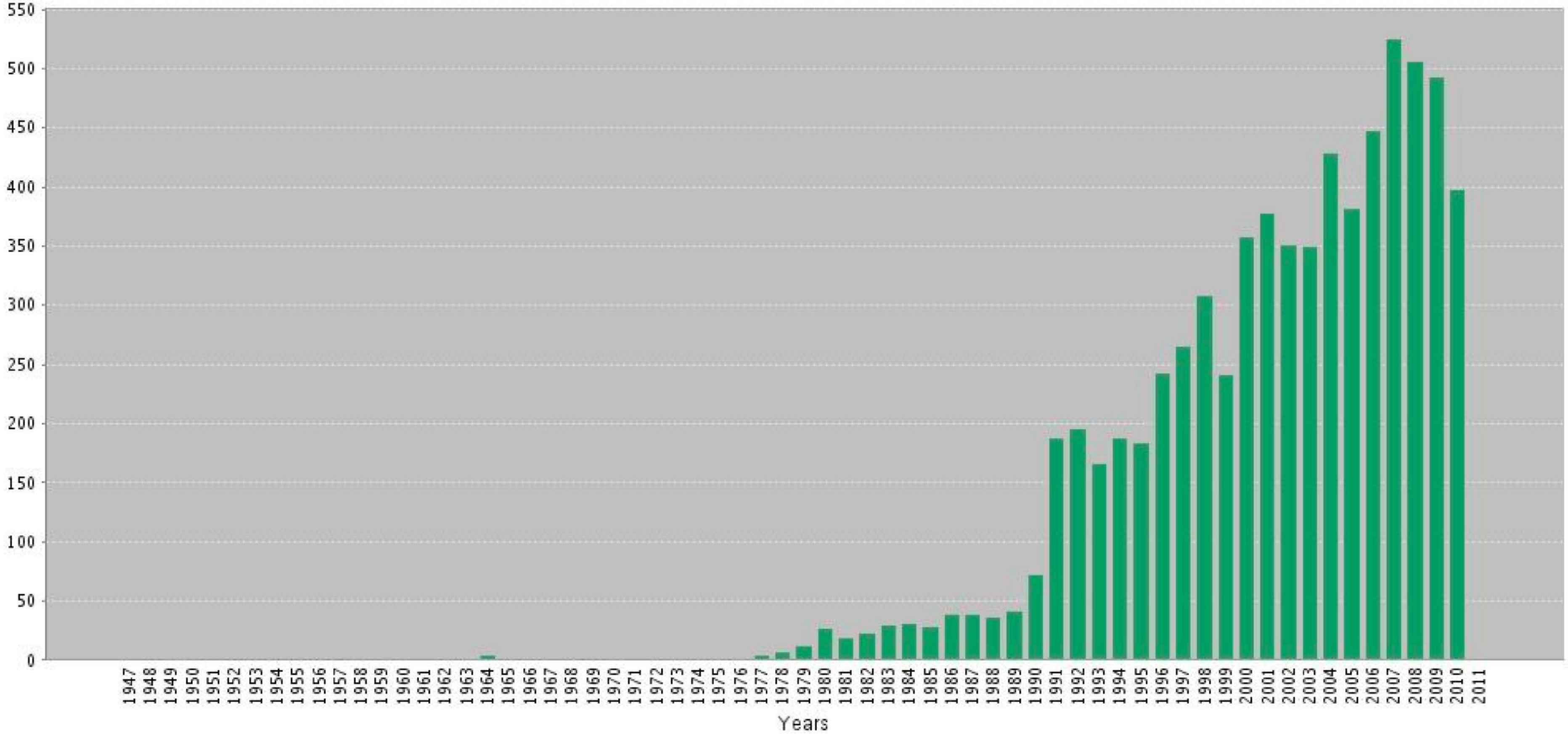
P. W. Anderson
Bell Laboratories, Murray Hill, New Jersey 07974
and
Cavendish Laboratory, Cambridge, England



The Resonating Valence Bond State in La_2CuO_4 and Superconductivity

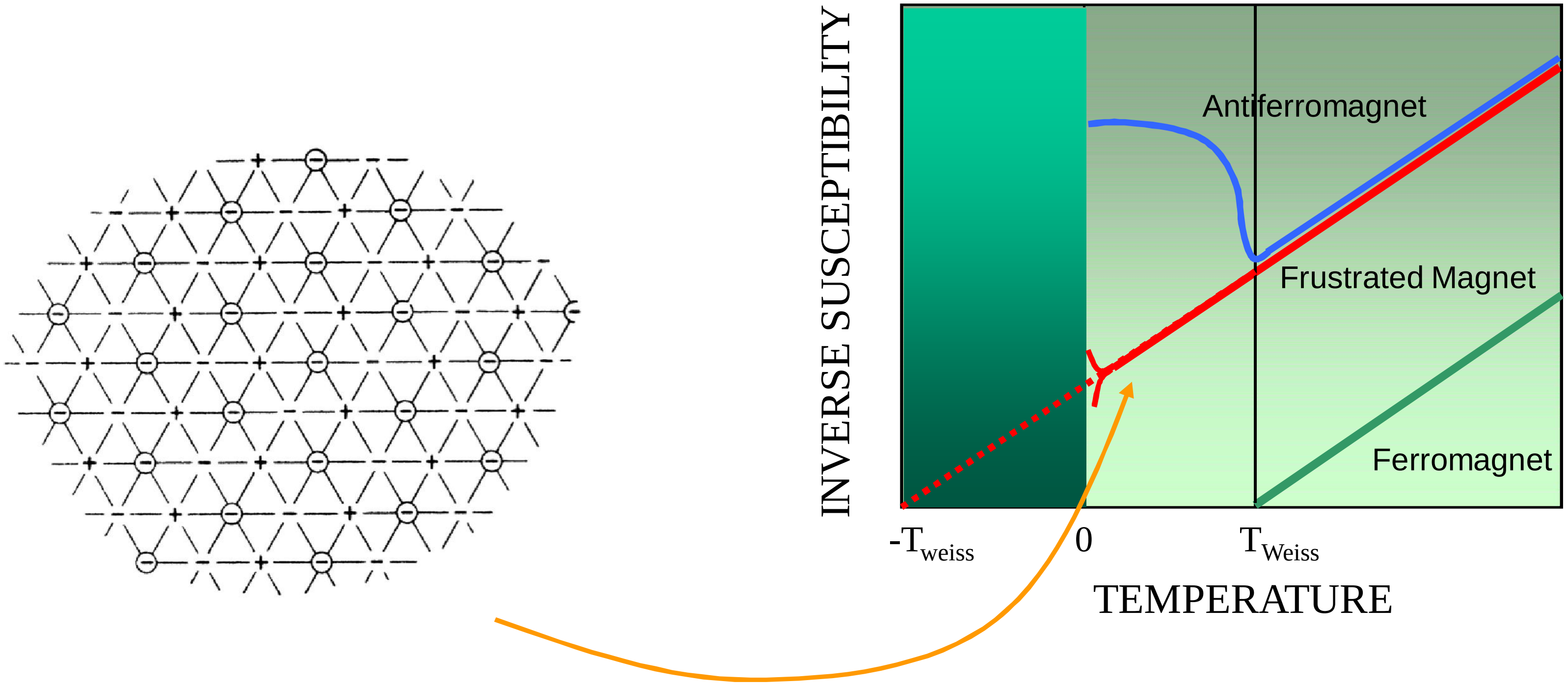
P. W. ANDERSON

Papers with key word "frustration"

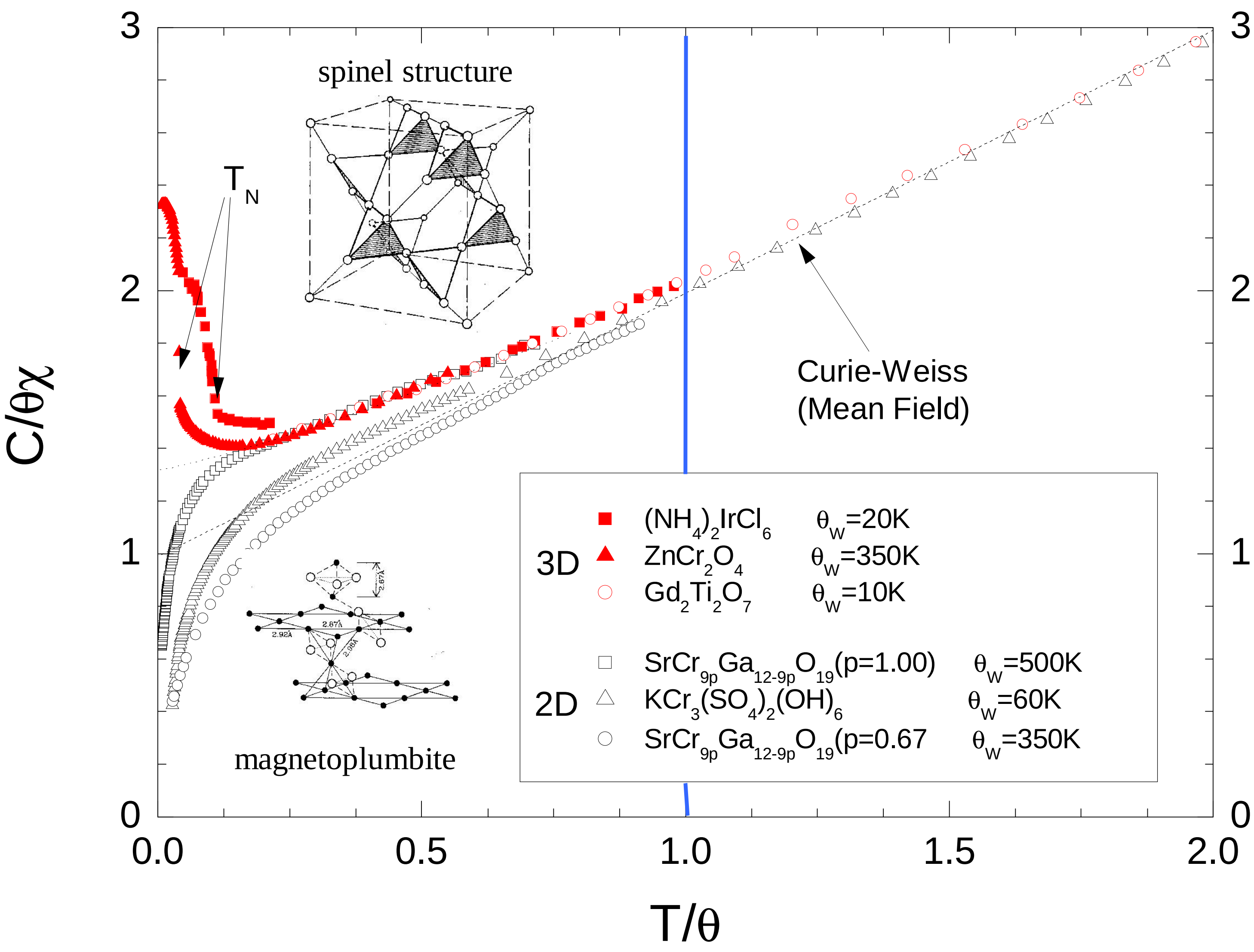


Source: ISI Web of Science

The bulk signature of geometrical frustration



Geometrical frustration seen in many systems



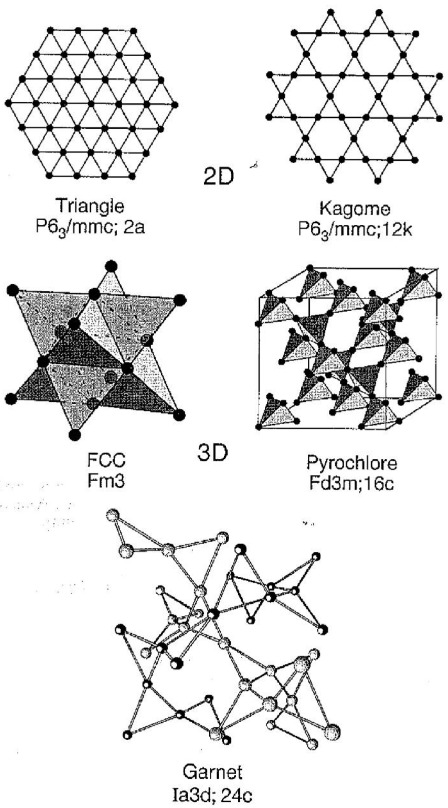
G-F Materials Commonalities

A Class of systems with:

§ Triangle-based lattices

§ Isotropic spins

§ θ_{Weiss}/T_c >10 (Anderson, Ter Haar & lines)



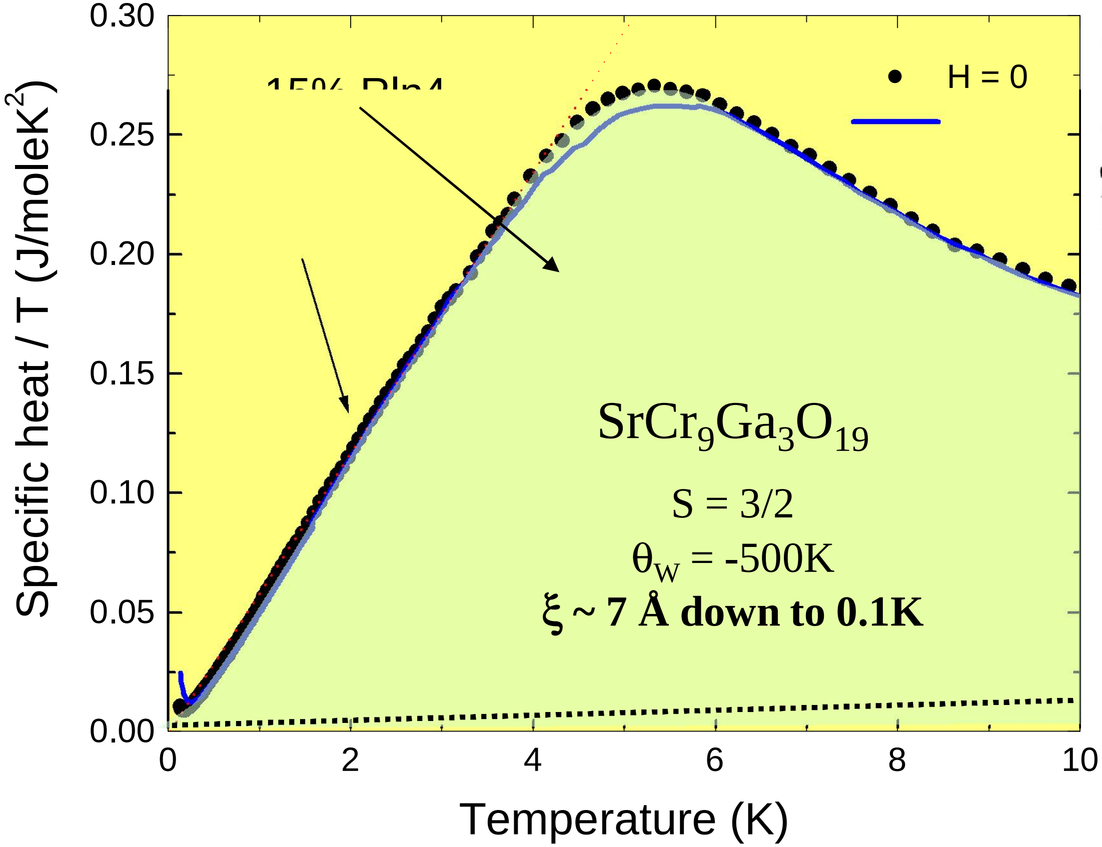
Strongly geometrically frustrated compounds

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

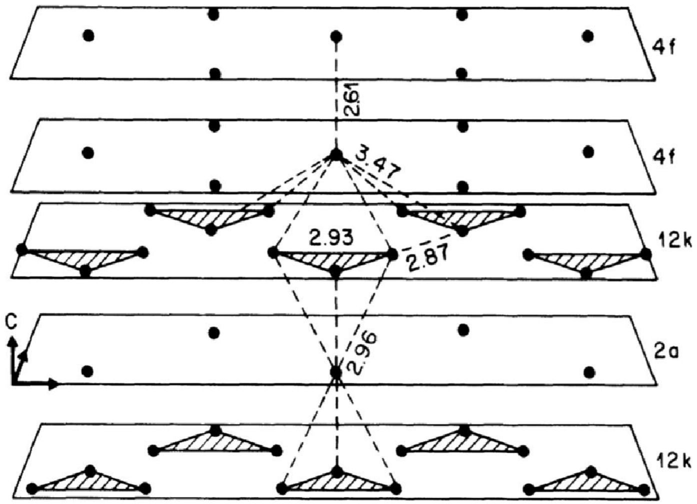
A. Ramirez, in Handbook of Magnetic Materials, 2001

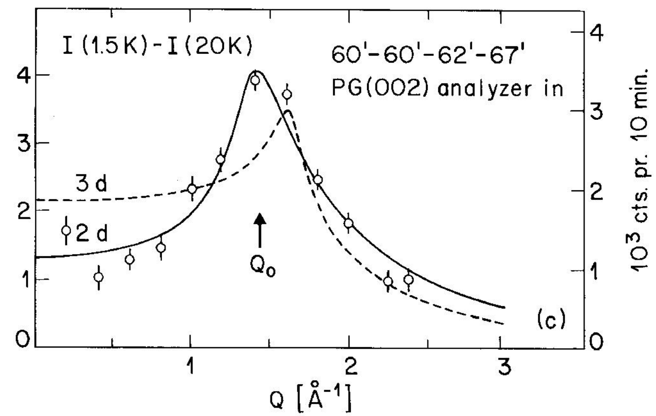
Ordering is necessary but not sufficient for GF

Spectral weight downshift in the kagome magnet $SrCr_9Ga_3O_{19}$



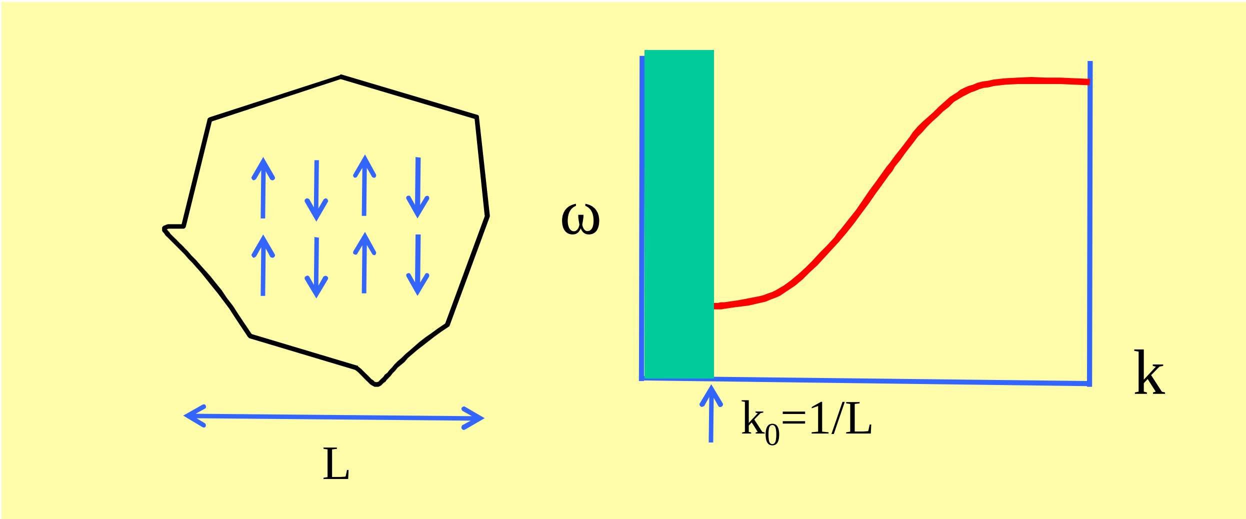
APR et al., PRL, **64**, 2070 (1990)

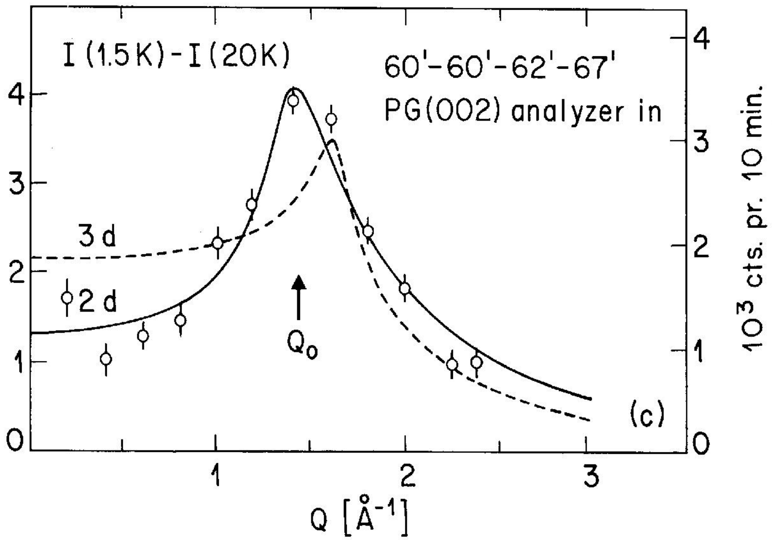




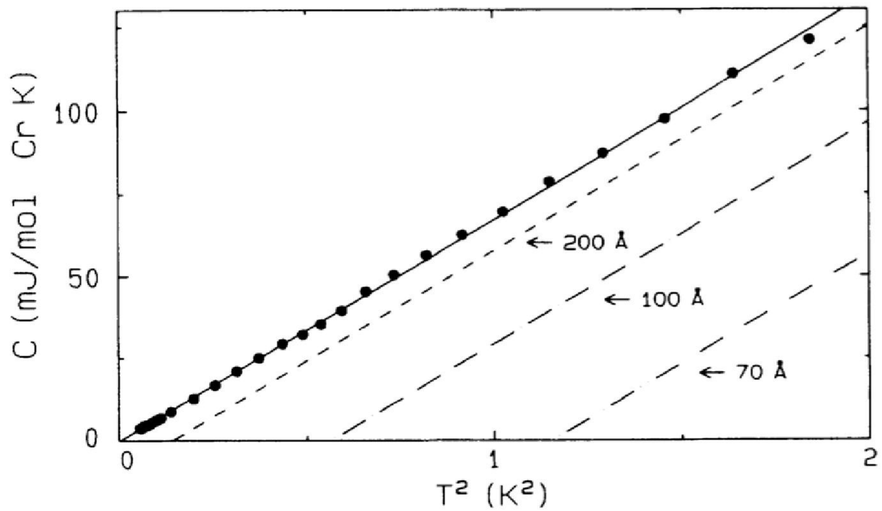
Broholm, Aeppli et al
Neutron scattering – liquid like structure factor

Coherence of the elementary excitations in a kagome AF



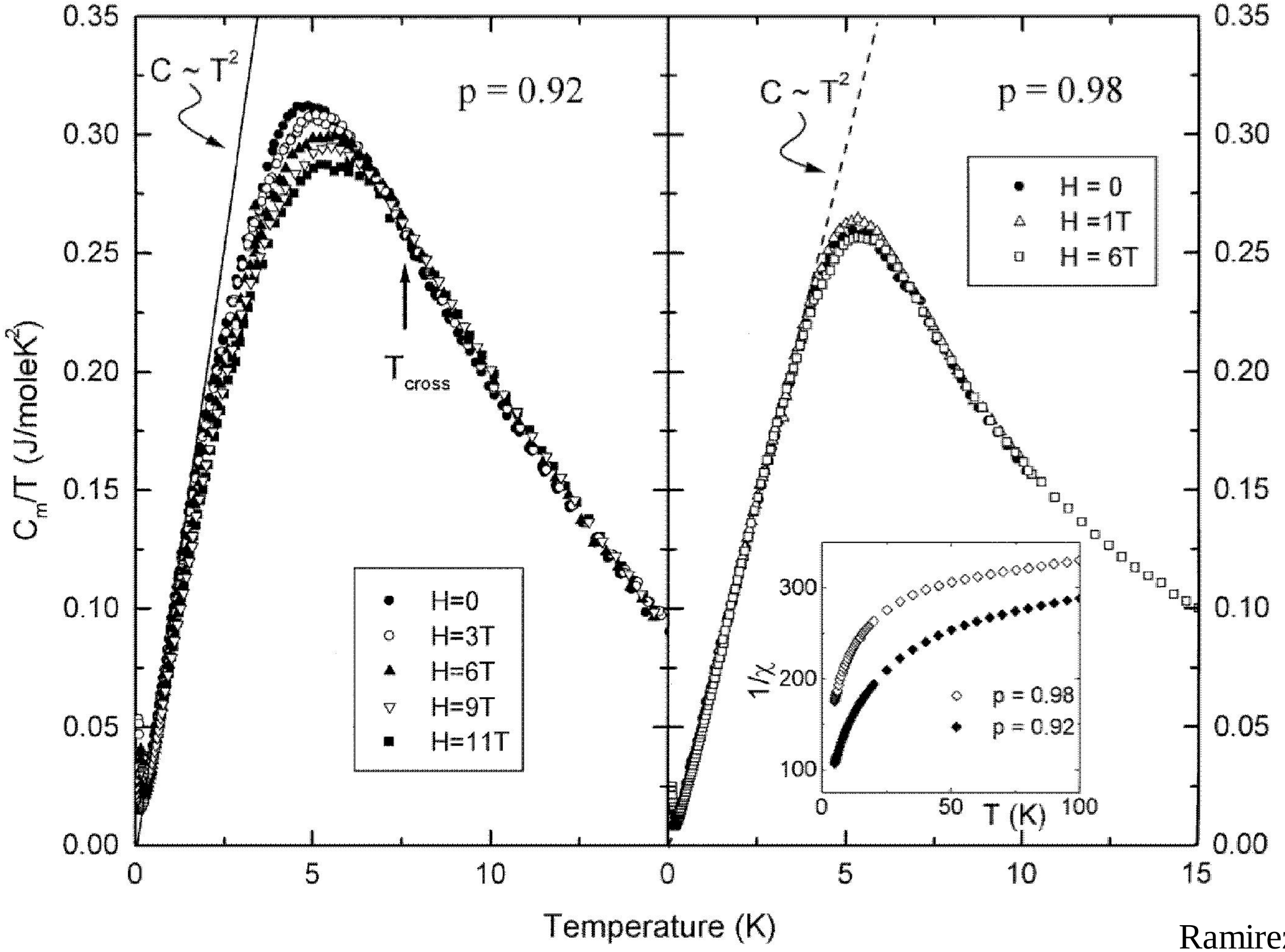


Broholm, Aeppli et al



Ramirez et al, 1992

B-field independence è singlet modes



Ramirez et al., 2000

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Interplay of lattice symmetry and underconstraint

D = total # degrees of freedom

R.Moessner & J. Chalker

K = # constraints in the ground state

F = D - K = # degrees of freedom available to the ground states

Example: cluster of 4 Heisenberg spins, e.g. on a tetrahedron

$$H = J/2\left(\sum_i S_i\right)^2$$

q = # spins per cluster = 4

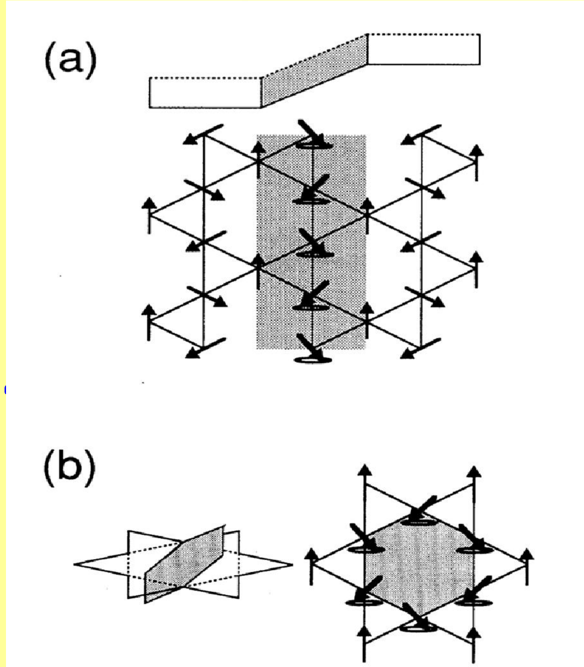
D=2q

$$\sum_i S_i = 0 \quad \rightarrow \quad \textbf{K=3}$$

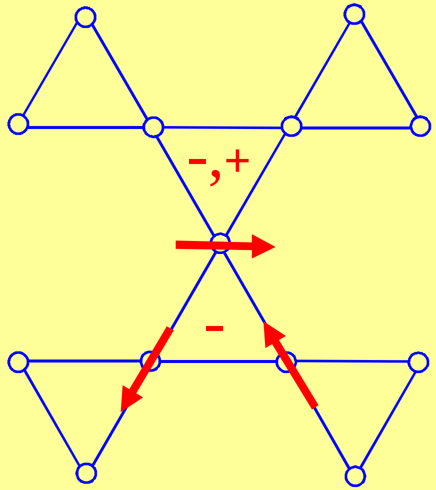
$\therefore$ **F = 2q-3**

So, for q = 4, cluster is underconstrained

Example - connected clusters in a Potts model



Ritchey, Coleman, Chandra

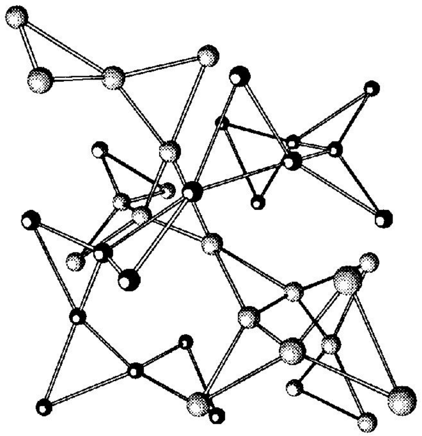


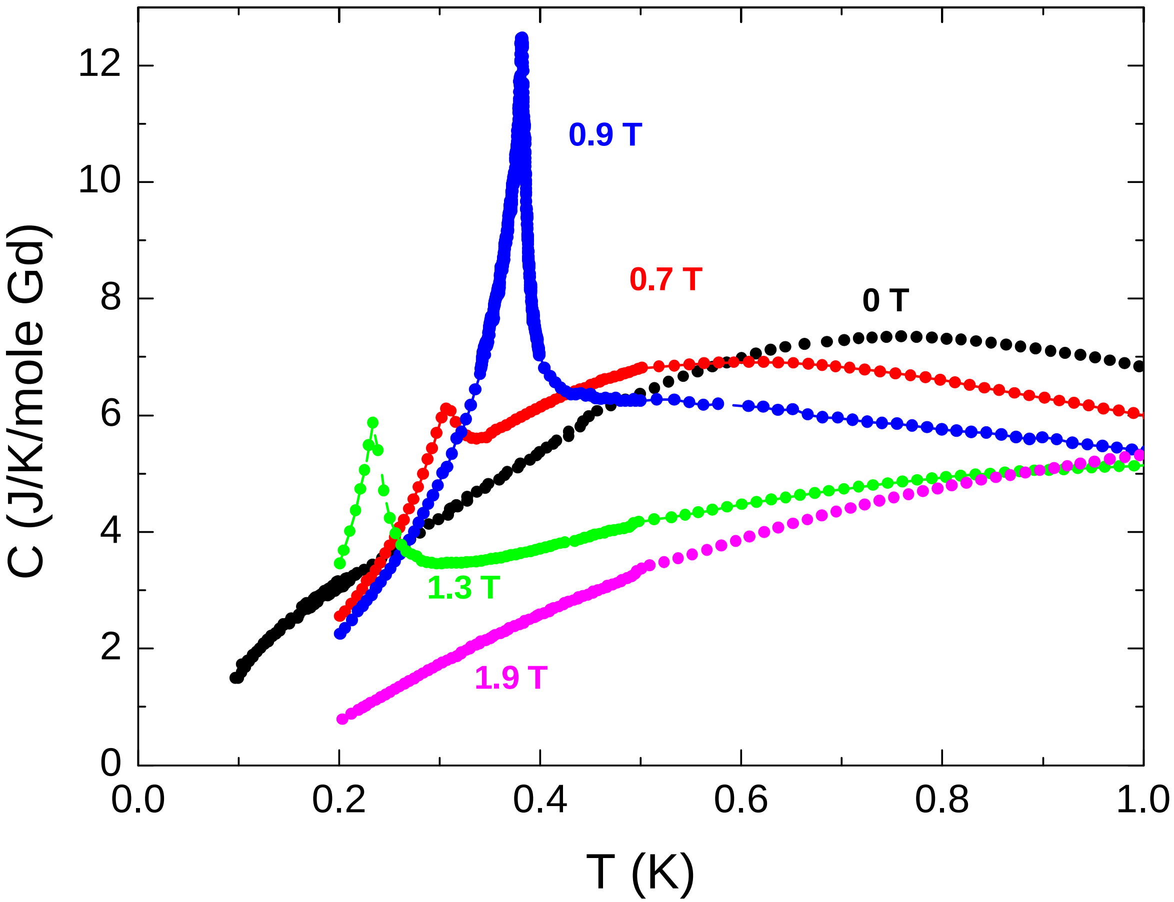
Kagome

So, the degeneracy can also depend on the connectivity

GF in 3D – $Gd_3Ga_5O_{12}$ "GGG"

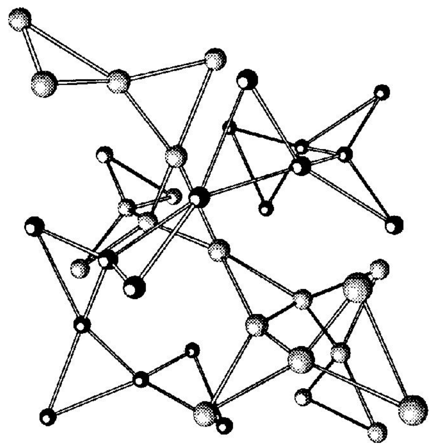
Interplay with magnetic field

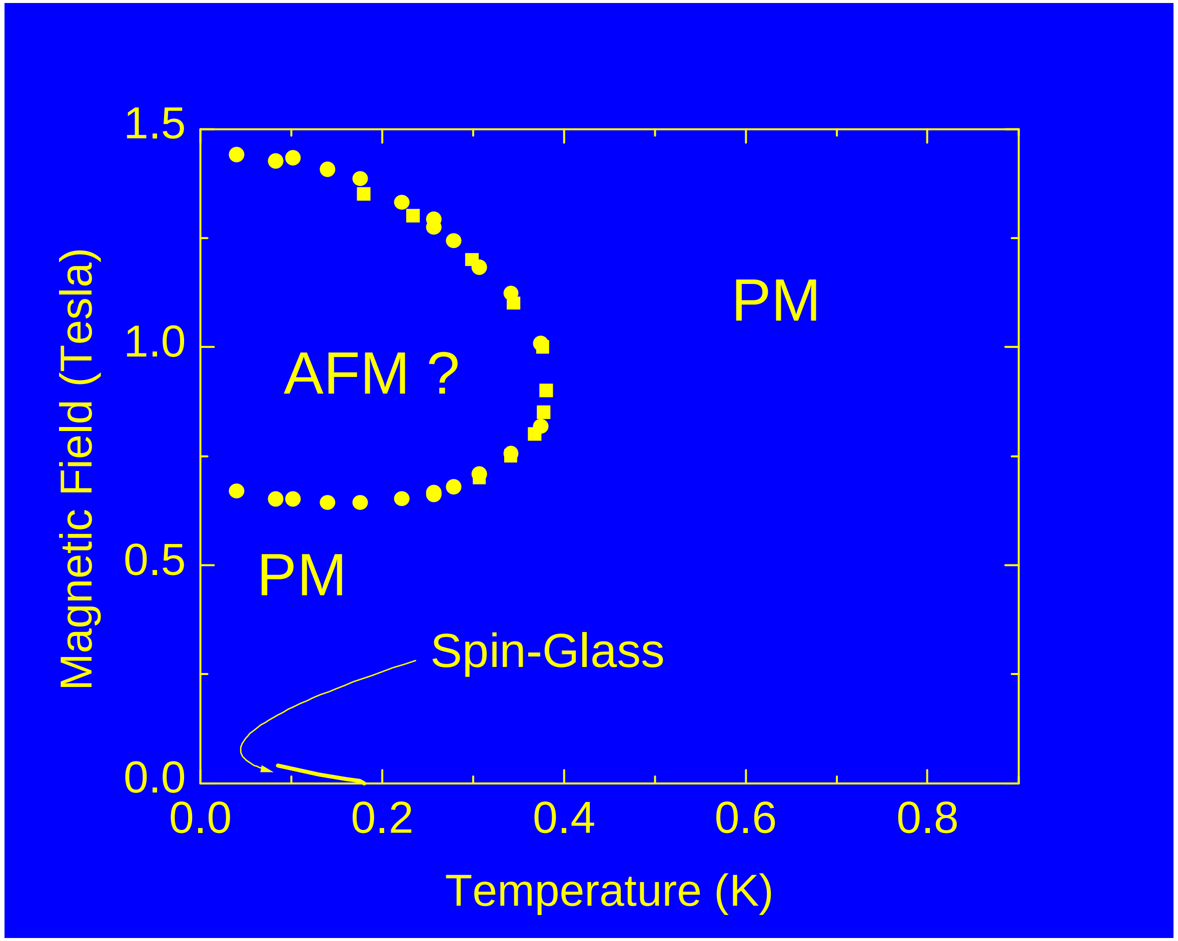




Schiffer, Huse, APR, *PRL* 1994

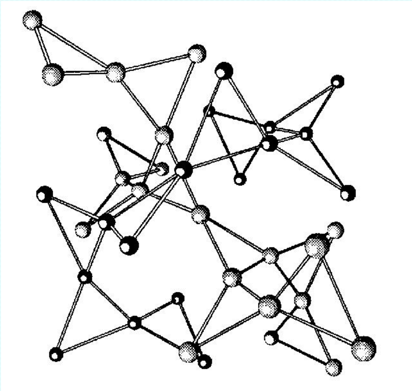
GGG – Phase Diagram



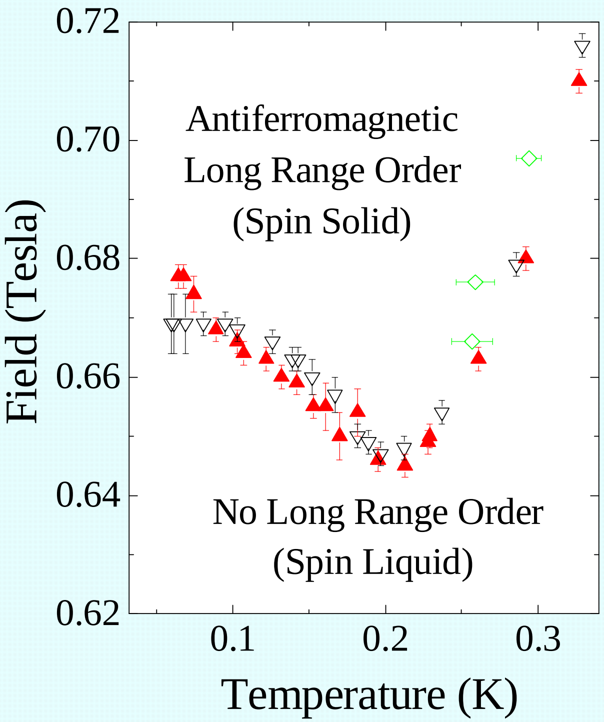


Phase diagram and loss of transverse mode in a GFM - $Gd_3Ga_5O_{12}$

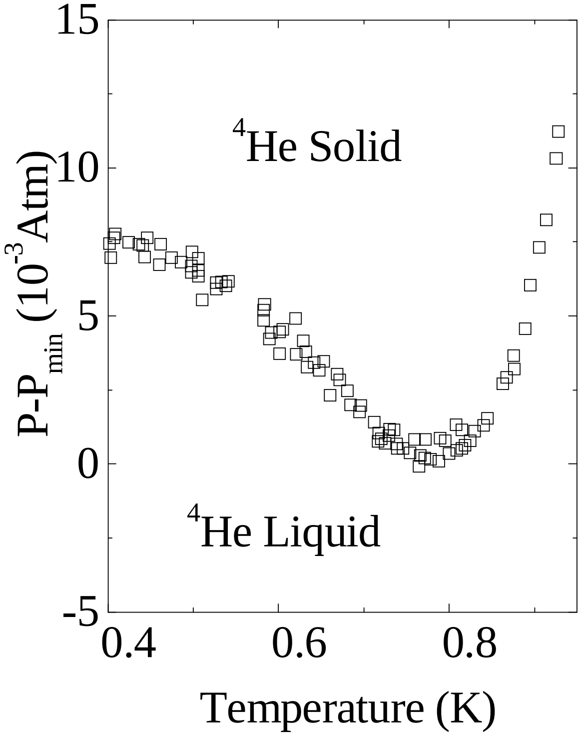
$Gd_3Ga_5O_{12}$



Garnet structure



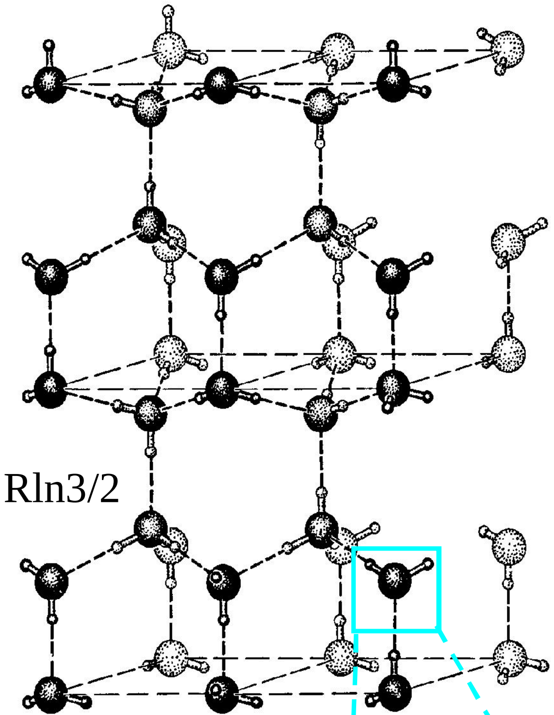
Helium-4



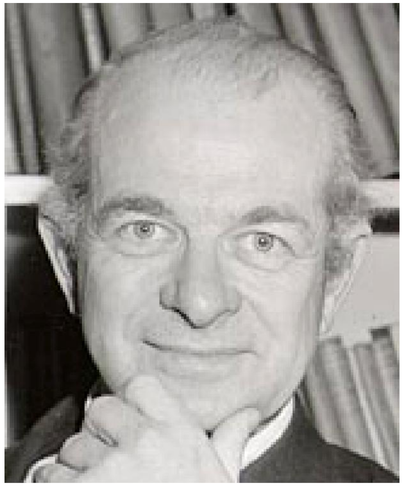
Schiffer et al, *PRL*, 1999

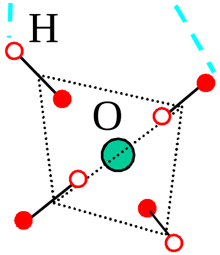
Ice & Spin Ice

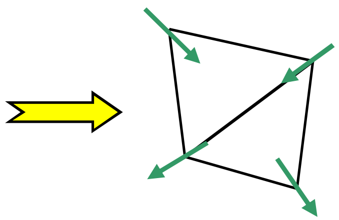
G-F for Ising Degrees of Freedom – Ice & Spin Ice



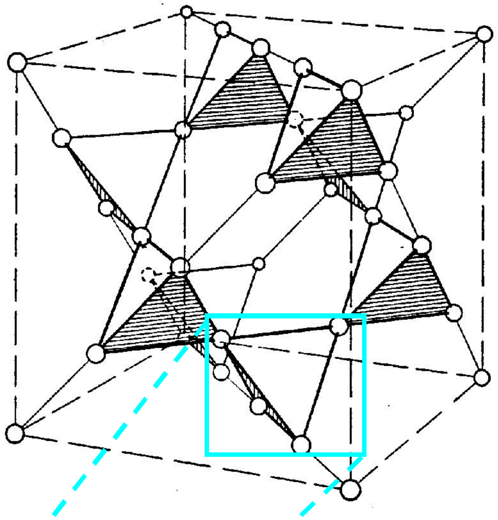
Pauling: $S_0 = R\ln 3/2$







B-spinel, or pyrochlore



Anderson, 1956

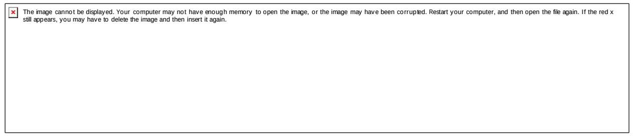
Harris, Bramwell, et al, 1997

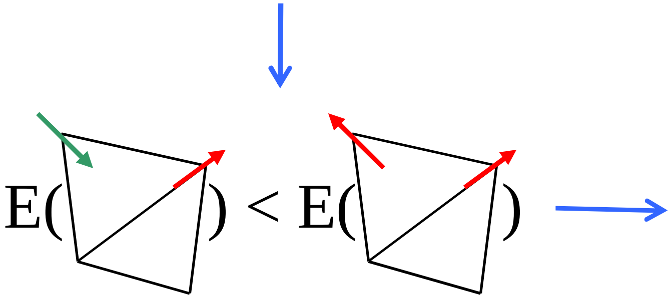
Liebmann, 1981

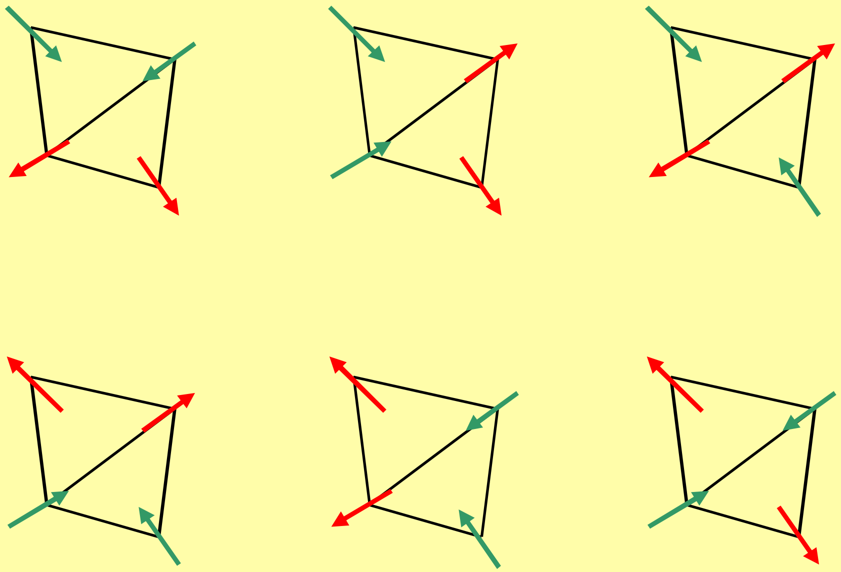
Dominant Energy in rare-earth systems is dipole-dipole

Atomic number	Symbol	Name	Atomic mass	Electrons per shell
57	La	Lanthanum	138.90547	2 8 18 18 9 2
58	Ce	Cerium	140.116	2 8 18 19 9 2
59	Pr	Praseodymium	140.90765	2 8 18 21 8 2
60	Nd	Neodymium	144.242	2 8 18 22 8 2
61	Pm	Promethium	(145)	2 8 18 23 8 2
62	Sm	Samarium	150.36	2 8 18 24 8 2
63	Eu	Europium	151.964	2 8 18 25 8 2
64	Gd	Gadolinium	157.25	2 8 18 25 9 2
65	Tb	Terbium	158.92535	2 8 18 27 8 2
66	Dy	Dysprosium	162.500	2 8 18 28 8 2
67	Ho	Holmium	164.93032	2 8 18 29 8 2
68	Er	Erbium	167.259	2 8 18 30 8 2
69	Tm	Thulium	168.93421	2 8 18 31 8 2
70	Yb	Ytterbium	173.054	2 8 18 32 8 2
71	Lu	Lutetium	174.9668	2 8 18 32 9 2

§ superexchange small
§ moment big







“Ice Rules” for Ising spins on a tetrahedron

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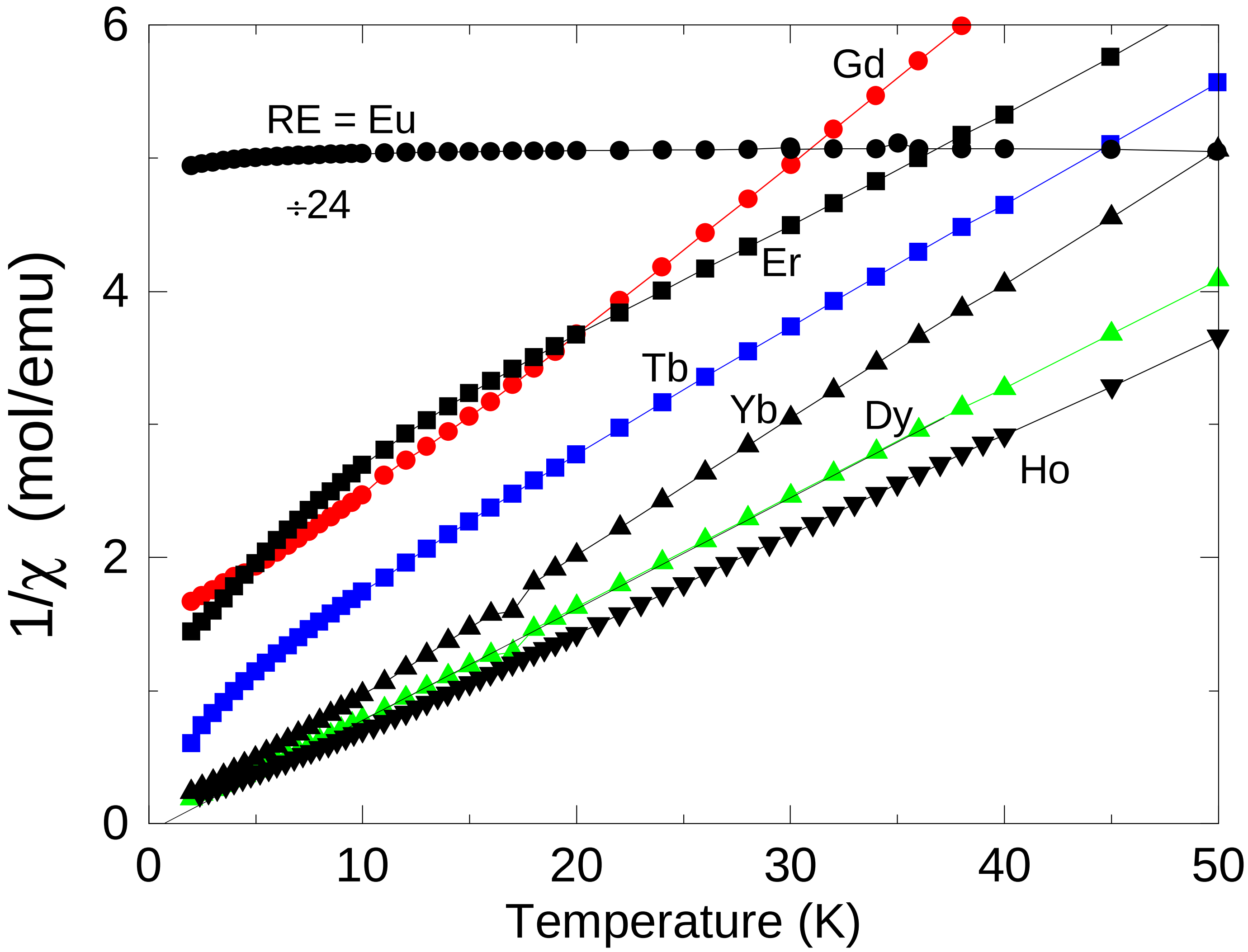
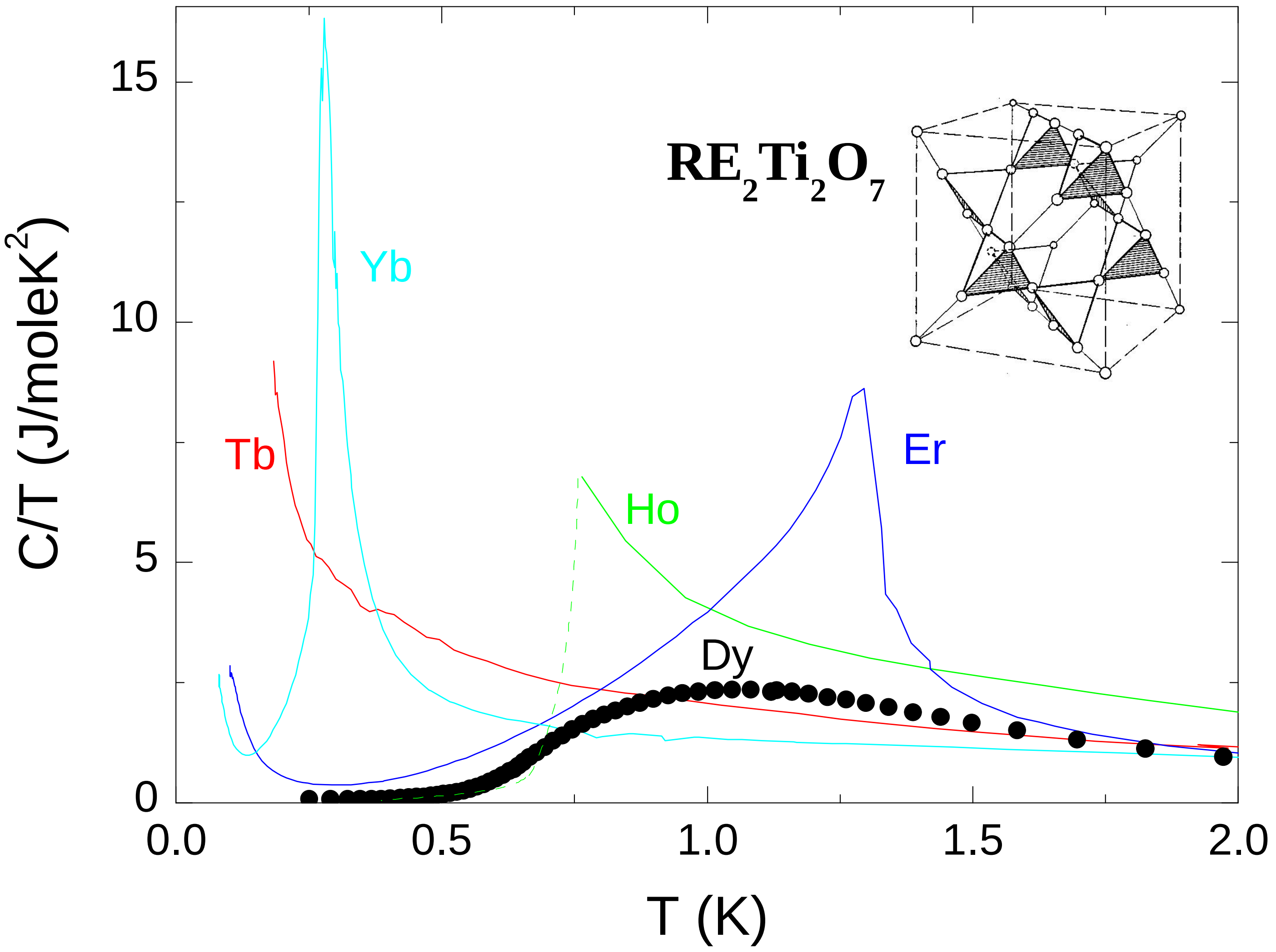
$1/\chi$, $RE_2Ti_2O_7$



Specific heat comparison - RE-titanate pyrochlores



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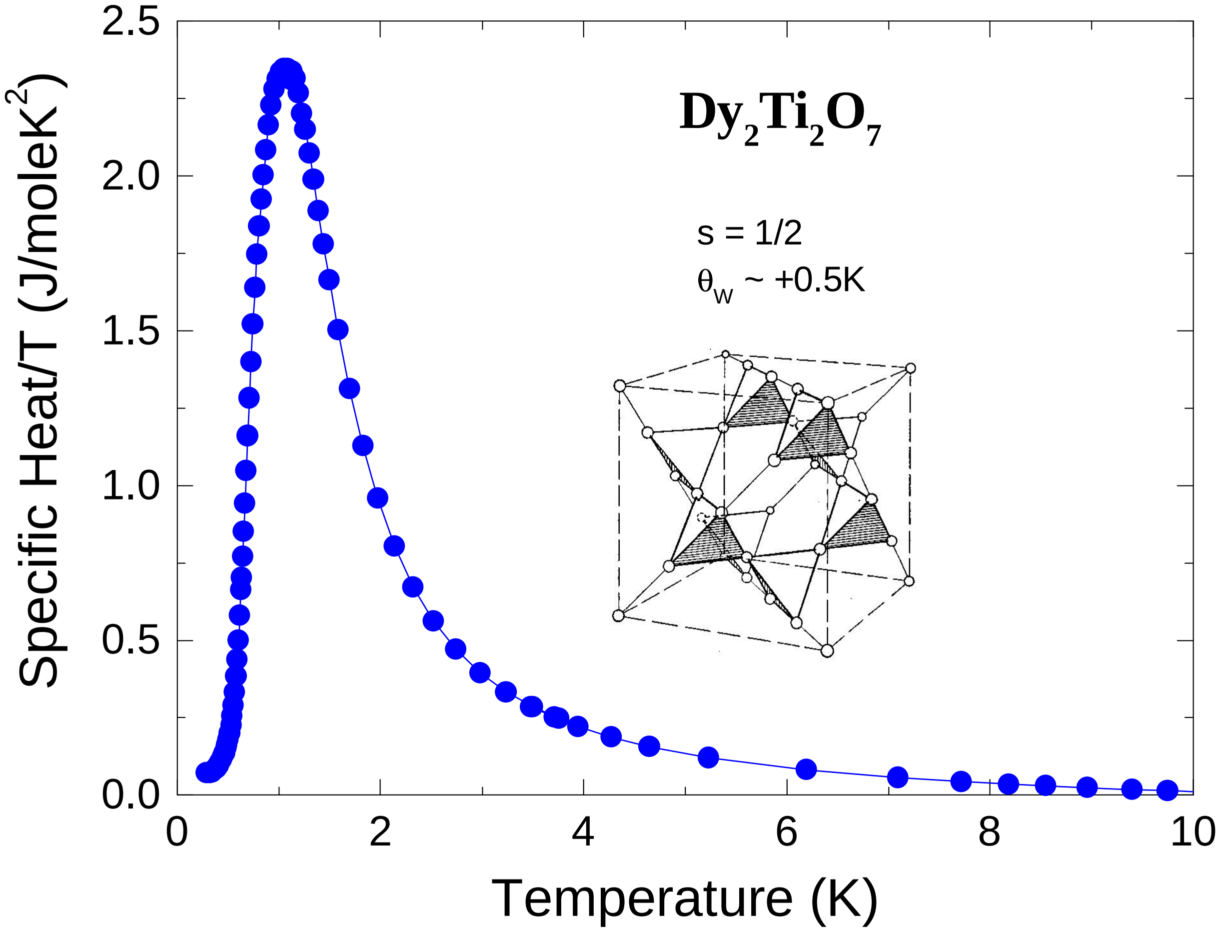
Specific heat – Ising, s = ½, FM, pyrochlore



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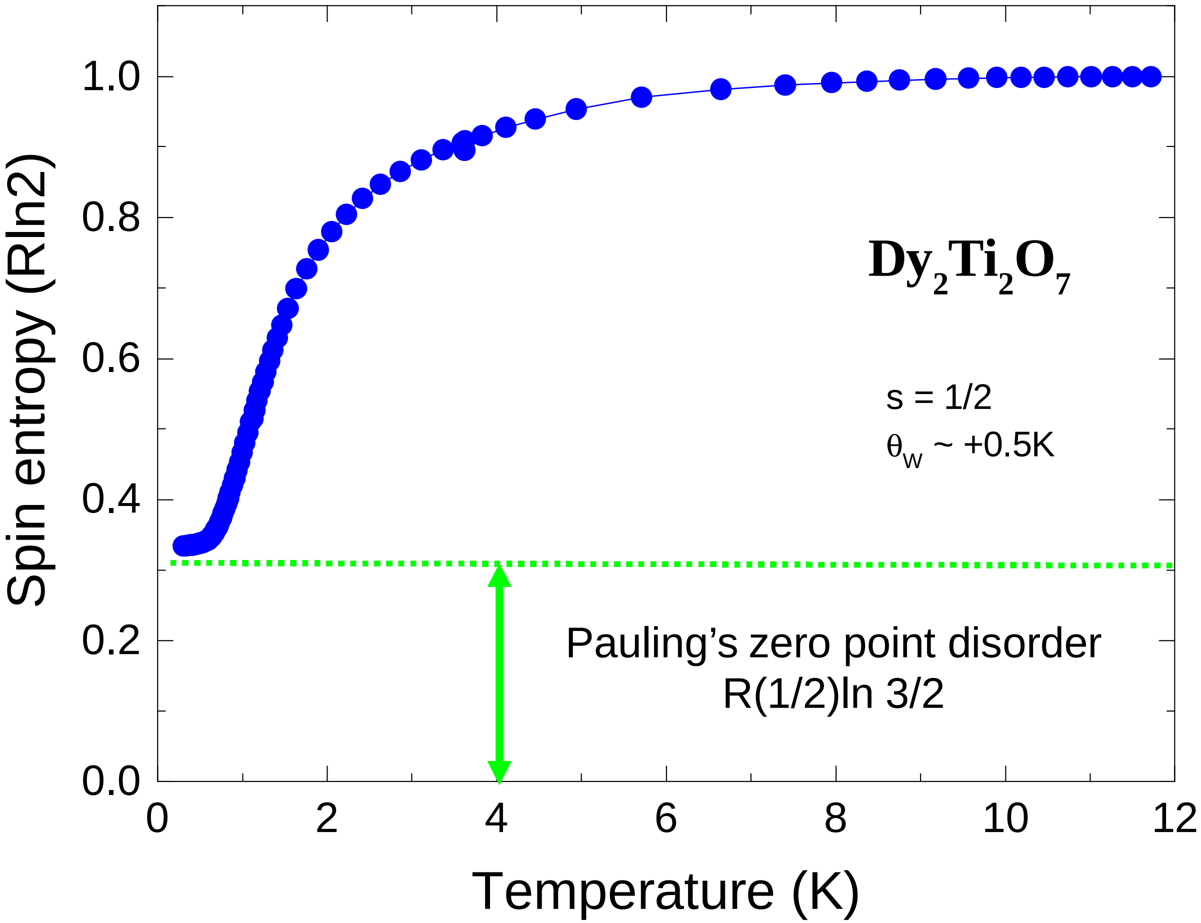
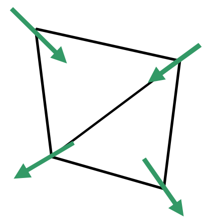
Observation of Zero Point disorder in *Spin Ice* - $Dy_2Ti_2O_7$





1) # tetrahedrons = N
2) # states per tetrahedron = 2^4 = 16
3) # states with low energy = 6 è (3/8)^N
4) #spin configs = 2^{2N}
5) # states=$2^{2N}(3/8)^N$
6) S_0=ln(3/2) per tetra.
7) S_0=1/2ln(3/2) per Dy

Ramirez, Siddarthan, Shastry, Cava, Nature **399**, 333 (1999)

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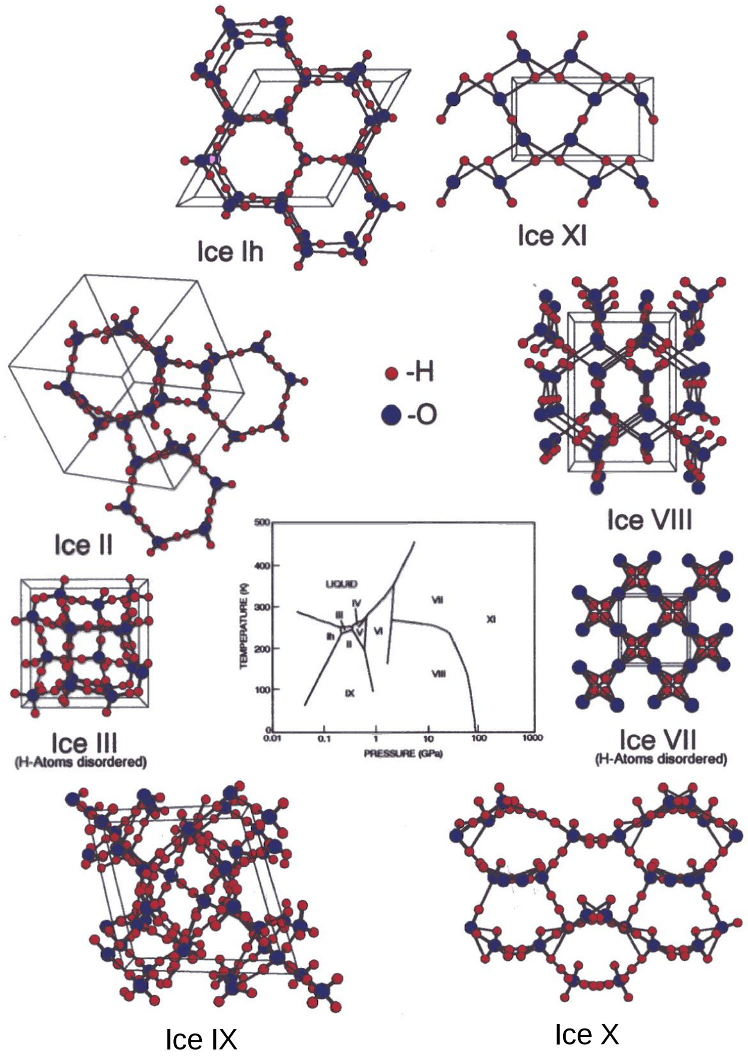
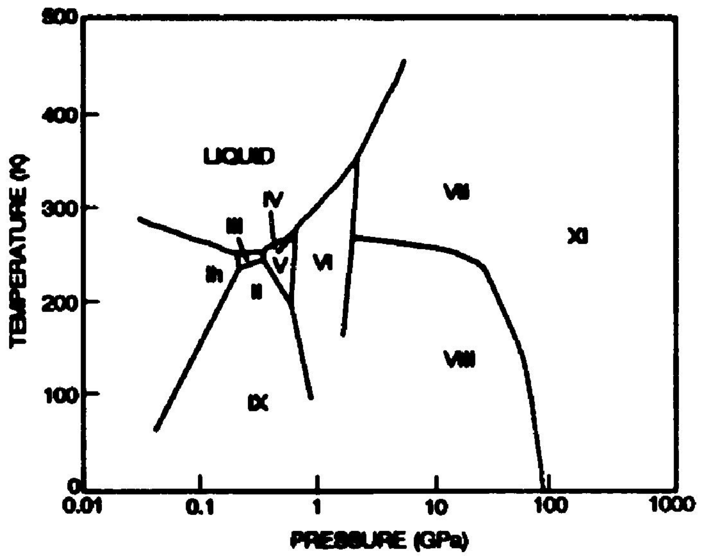
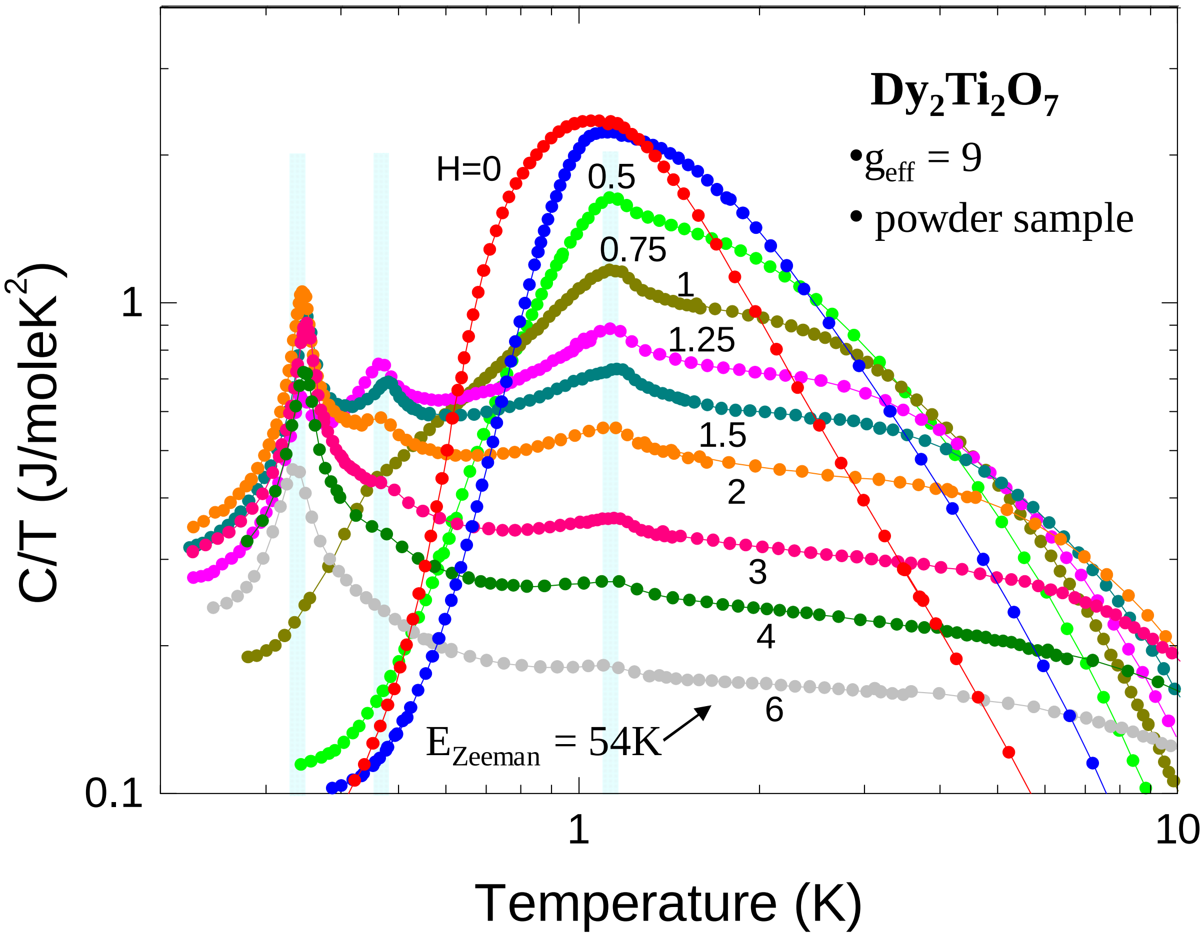
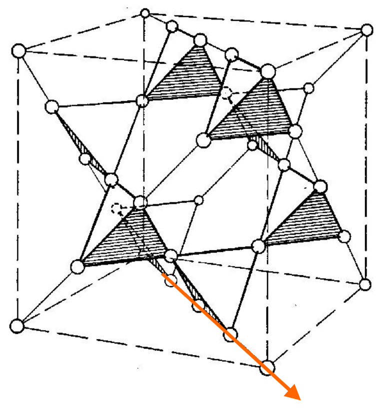
Crystal Structures of H_2O





Breaking the ice-rules with magnetic field





Nature **399**, 333 (1999)

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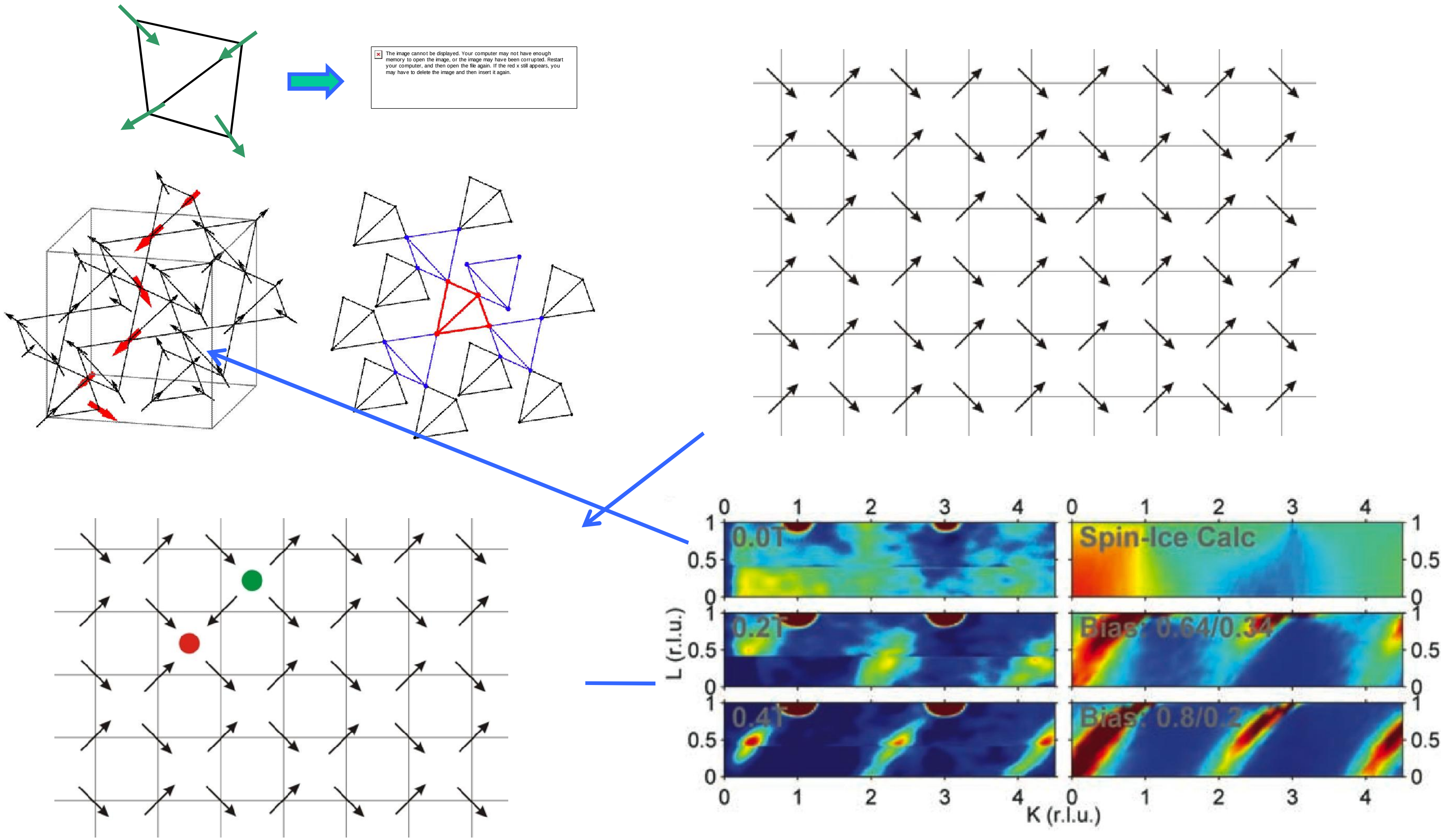
Electrostatics of *Spin Ice* – Emergence of Monopoles



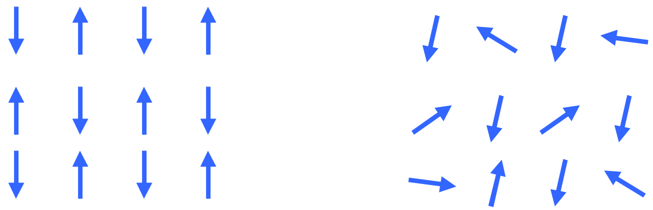
Morris et al, Science, 2009

Castelnovo, Moessner, Sondhi, Nature 2008

Geometric Frustration Beyond Magnetism

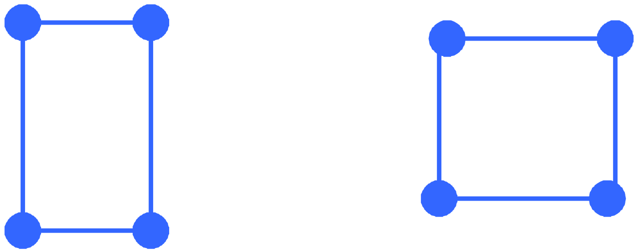
Comparison – Magnetic vs. Structural Transitions

Generic Magnetic Transition – Order-Disorder

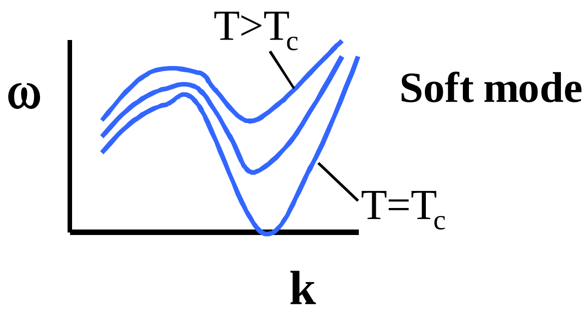


below T_c above T_c

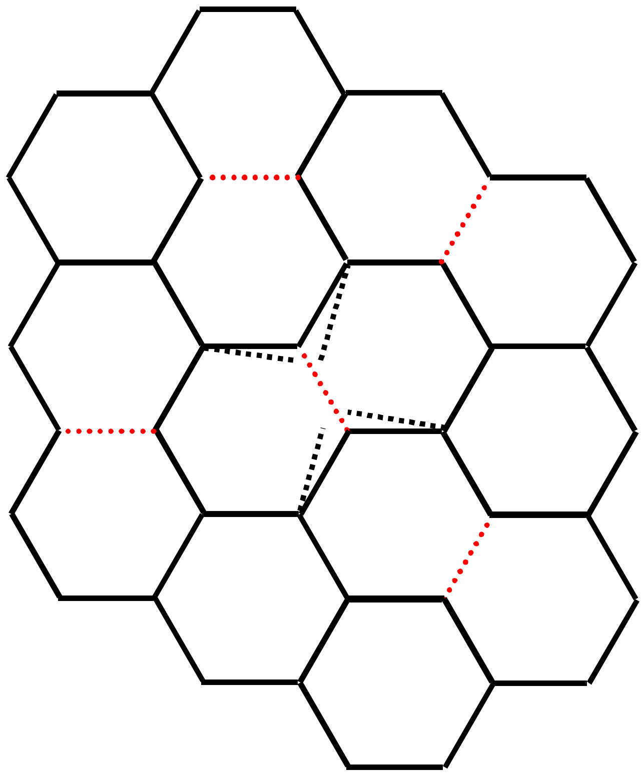
Generic Structural Transition – Displacive



below T_c above T_c



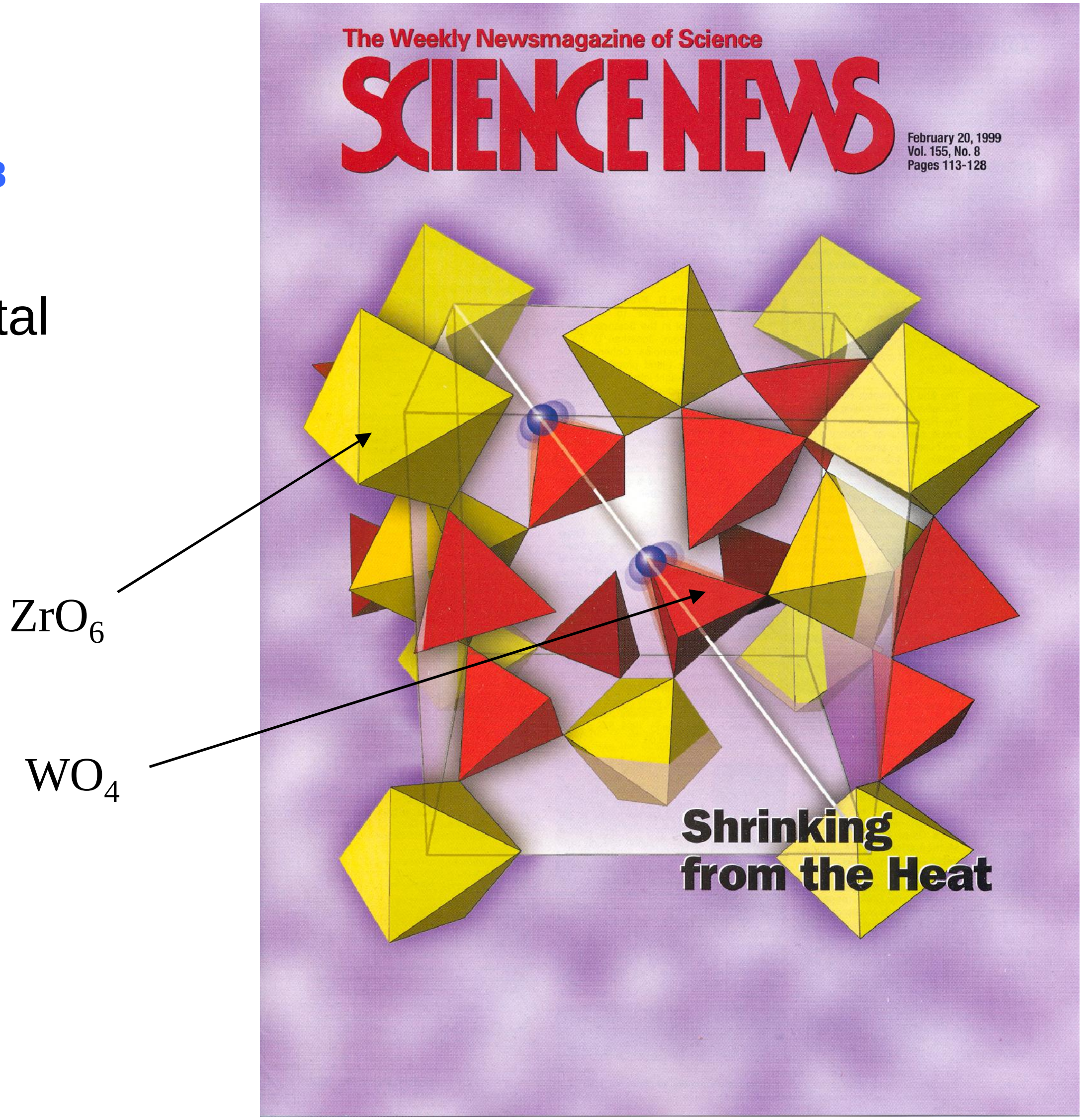
Can there be a “Frustrated Soft Mode”?



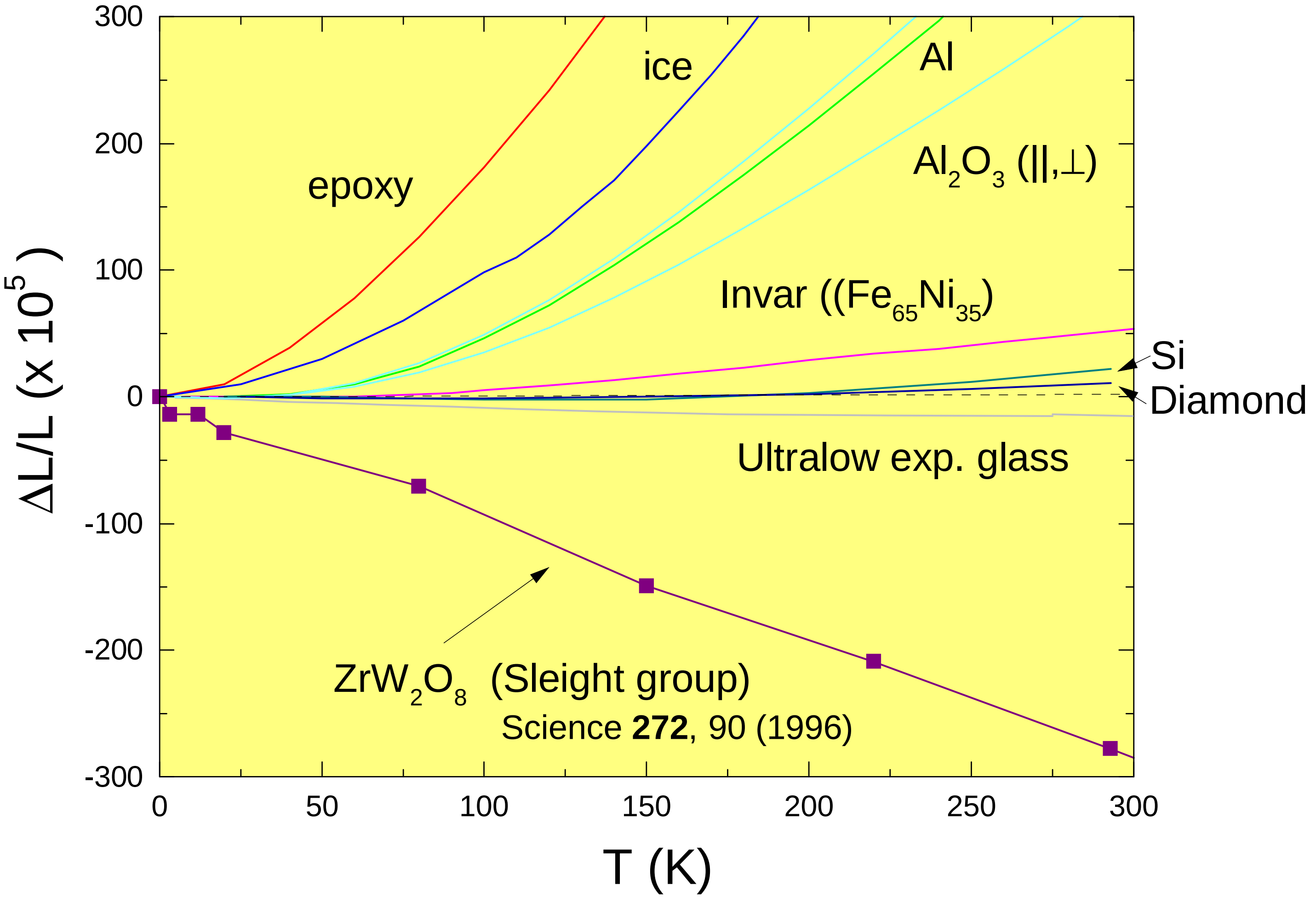
Frustrated Soft Modes – Negative Thermal Expansion

Negative thermal expansion in ZrW_2O_8

Unusual, highly underconstrained crystal structure



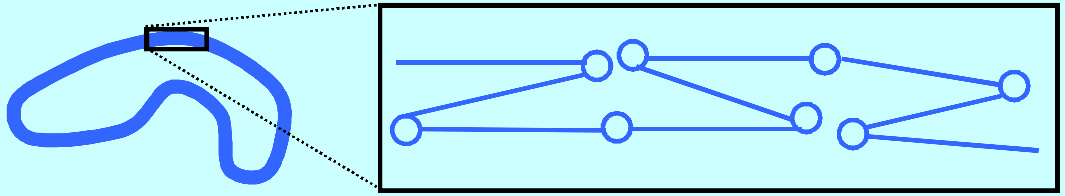
Thermal expansion - materials comparison

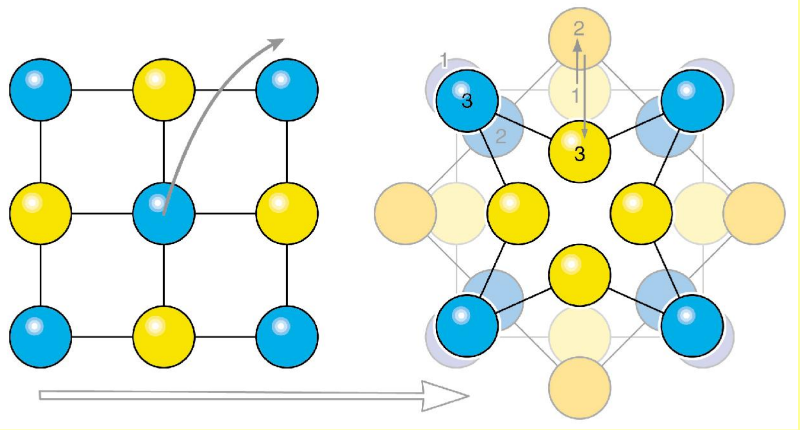


Negative thermal expansion

$$\alpha = \frac{1}{3V}\frac{\partial V}{\partial T}\bigg|_P = \frac{\gamma C}{3B} \qquad \gamma_{k,s} = \frac{\partial \ln \omega_s(k)}{\partial \ln V}$$

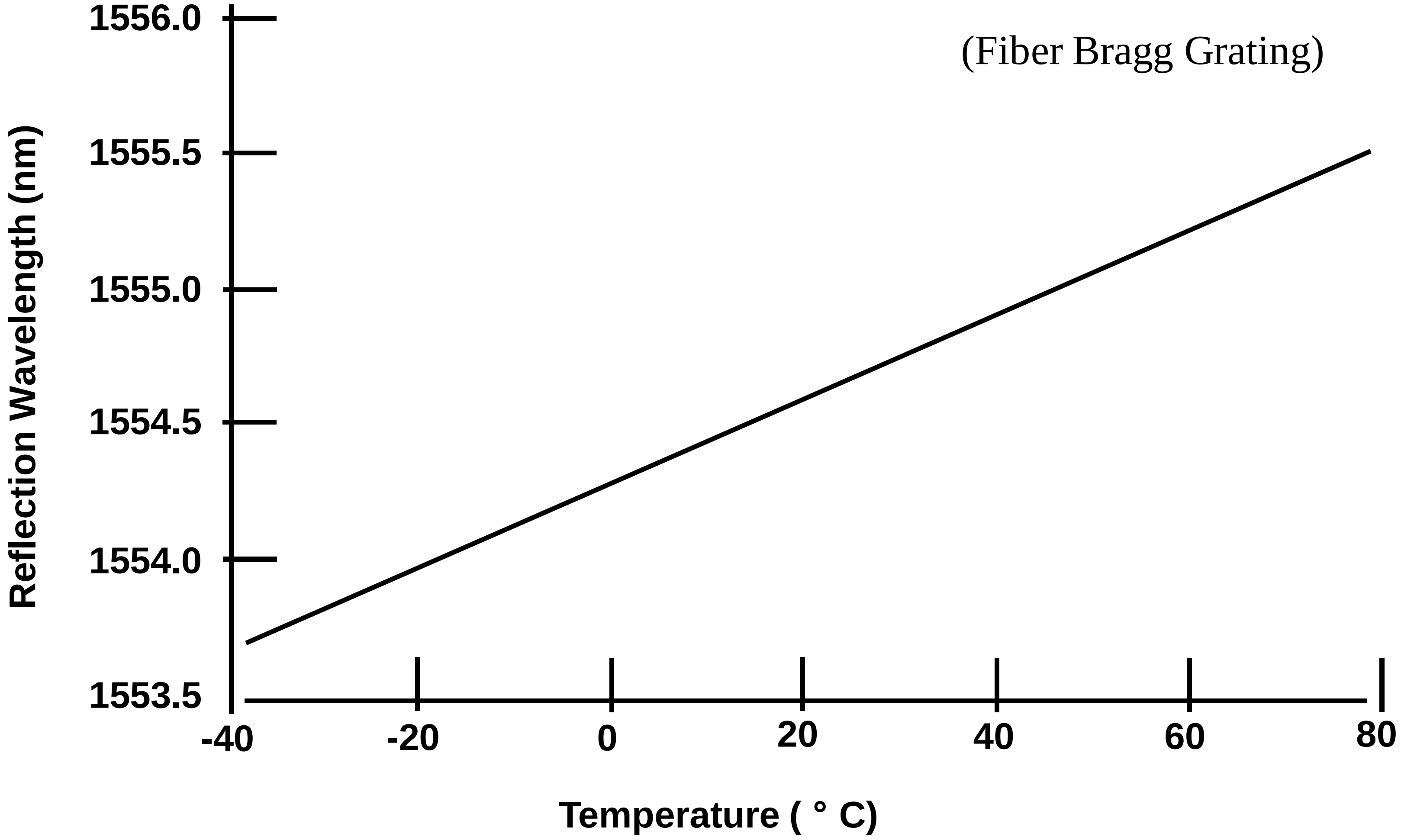
Examples



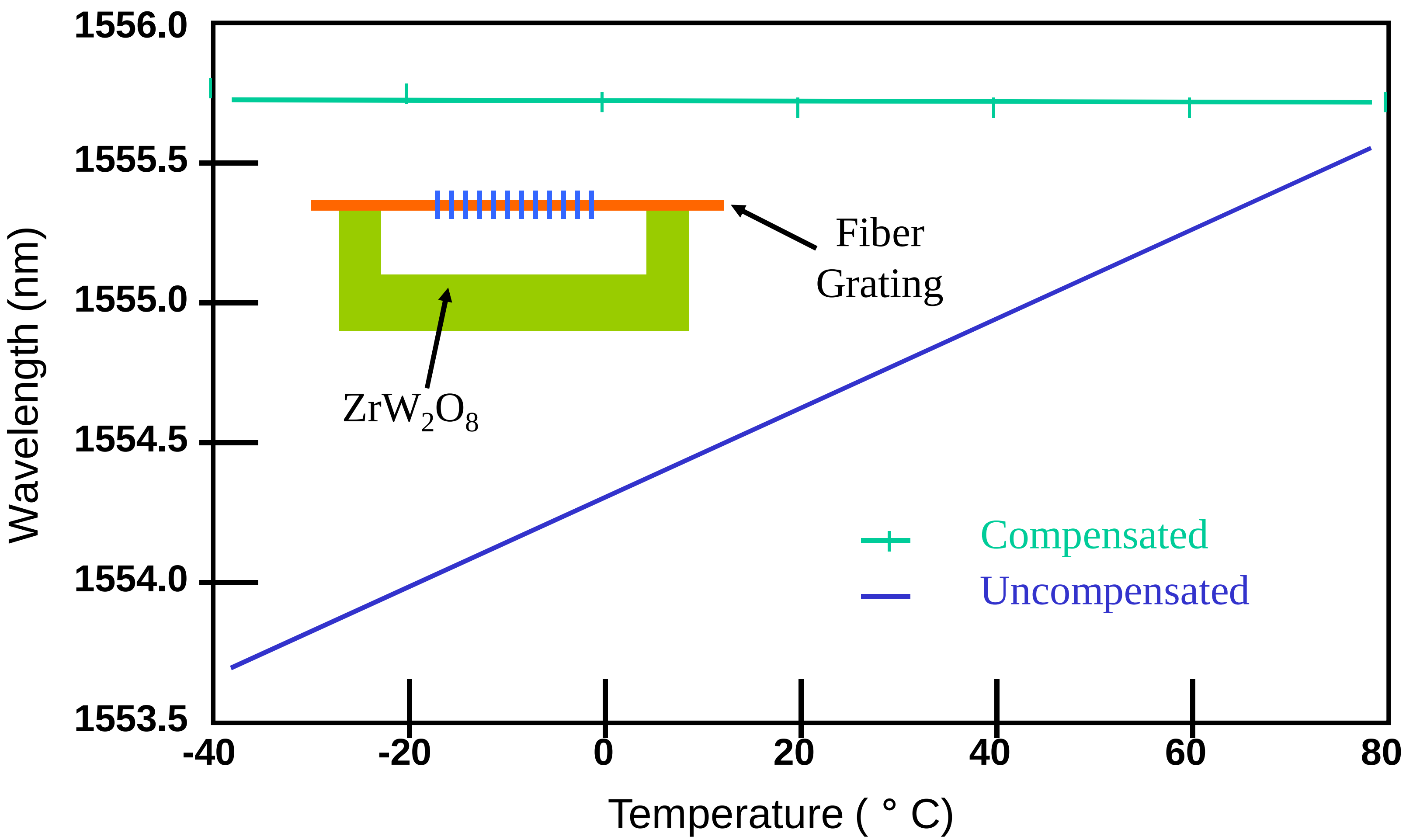


Application of NTE

Temperature dependence of index of refraction in a FBG



Solution to problem - bond FBG to ZrW_2O_8/ZrO_2 composite



Aside – any connection to magnetism?

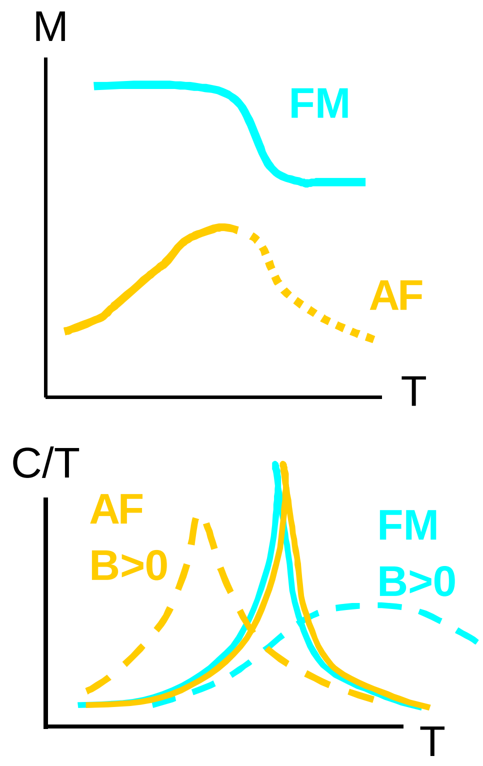
$$\alpha = \frac{1}{3}\frac{\partial \ln V}{\partial T} = \frac{1}{B}\left(\frac{\partial S}{\partial V}\right)_T$$

- In a magnet, V is replaced by -M, and P by H, ask what is analogy for low-energy behavior?

NTE $\rightarrow$ $\alpha < 0$ means…….$\partial V/\partial T < 0$ $\rightarrow$ $\partial M/\partial T > 0$

…………………….. and $\partial S/\partial V < 0$ $\rightarrow$ $\partial S/\partial M > 0$

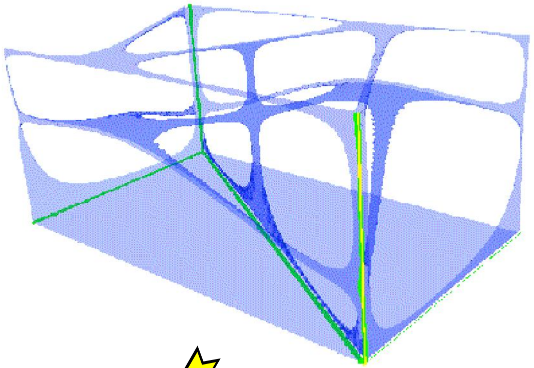
Negative Thermal Expansion *is* Antiferromagnetism



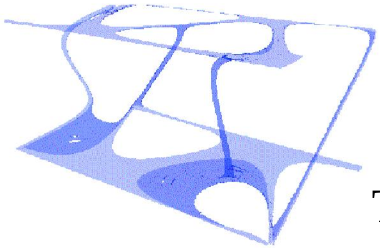
Structure implies *underconstraint* - what about *symmetry*?

Look at low-energy modes in reciprocal space by the split-atom method (Dove et al.)

Quartz

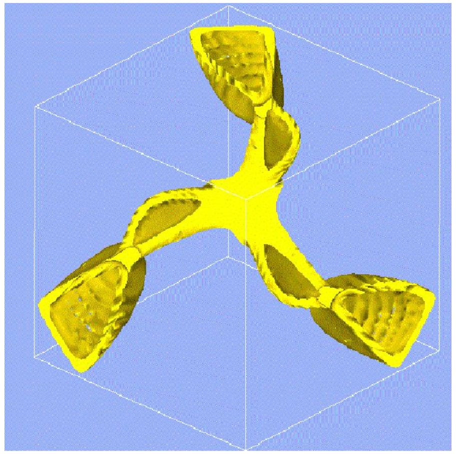


T > T_c



T < T_c

ZrW_2O_8

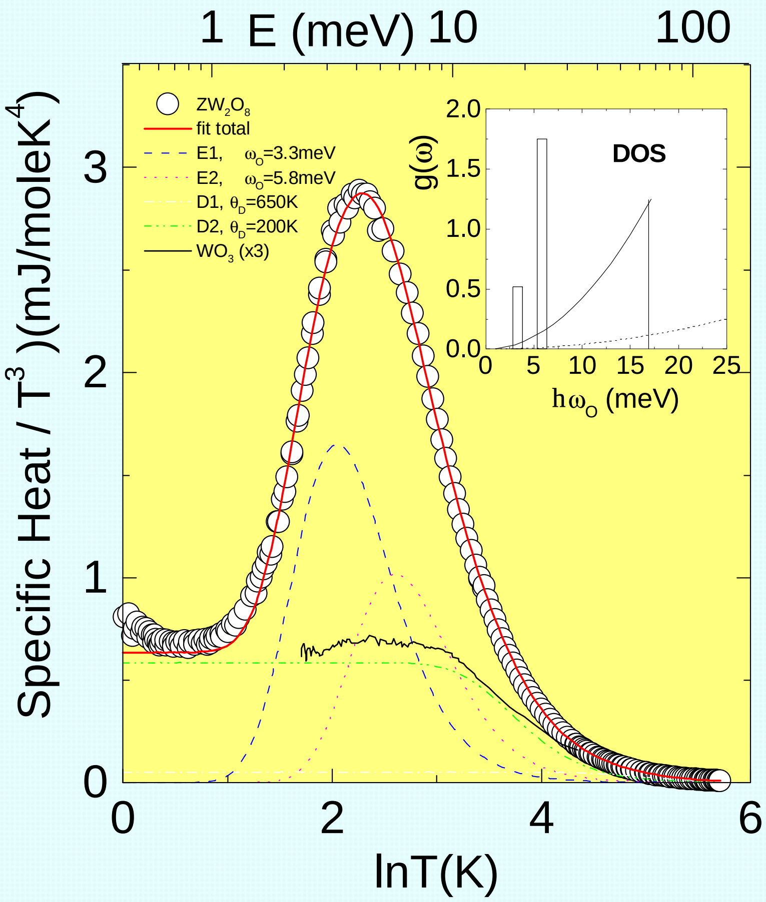


T = 300K

??? Is there evidence for low-energy modes?

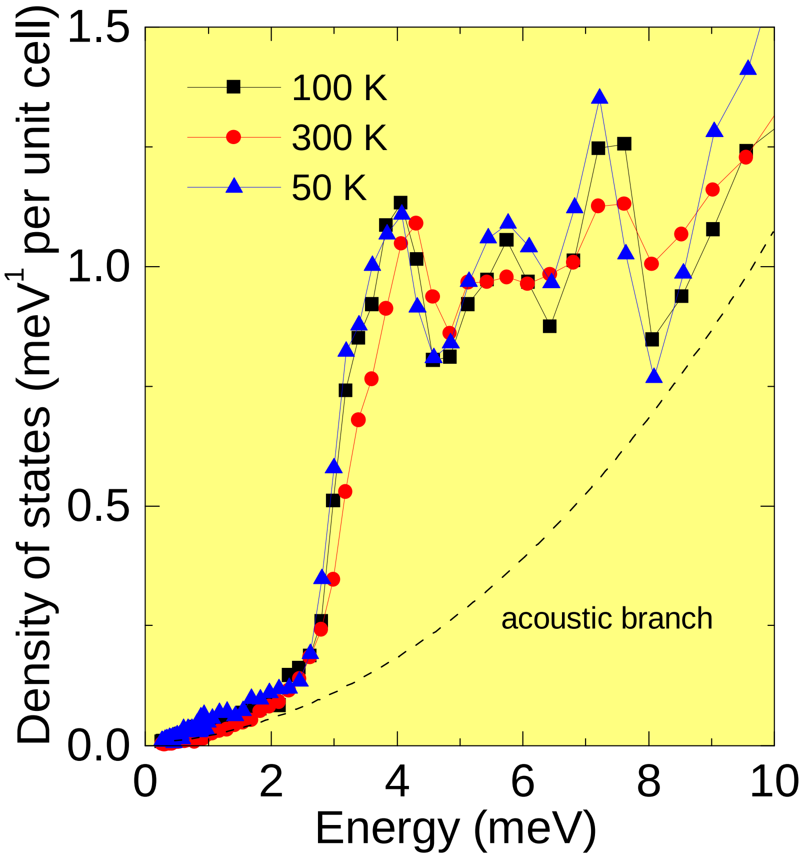
Yes - low-energy lattice modes in ZrW_2O_8

Specific heat



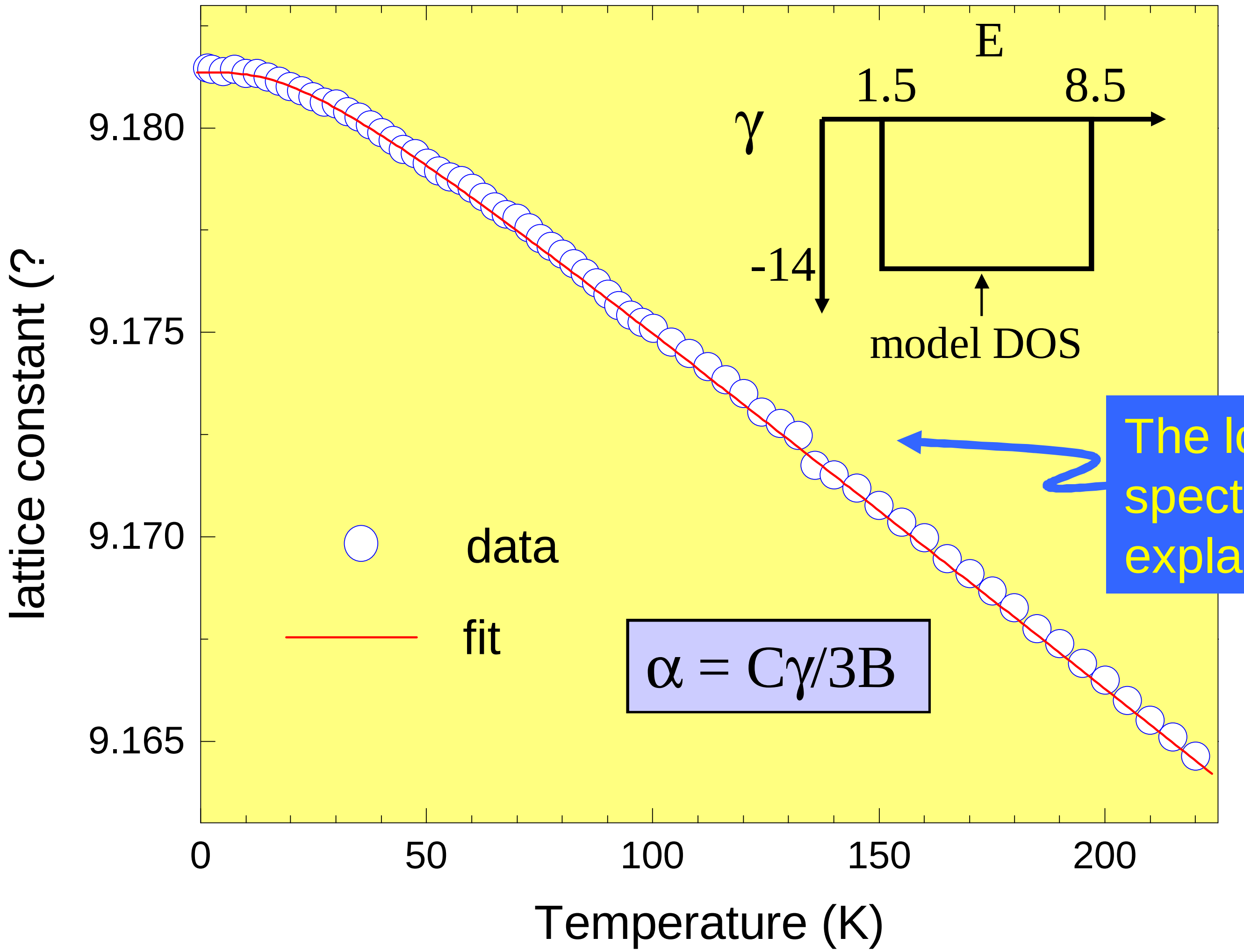
PRL **80**, 4903 (1998)

Neutron scattering DOS



Nature **396**, 147 (1998)

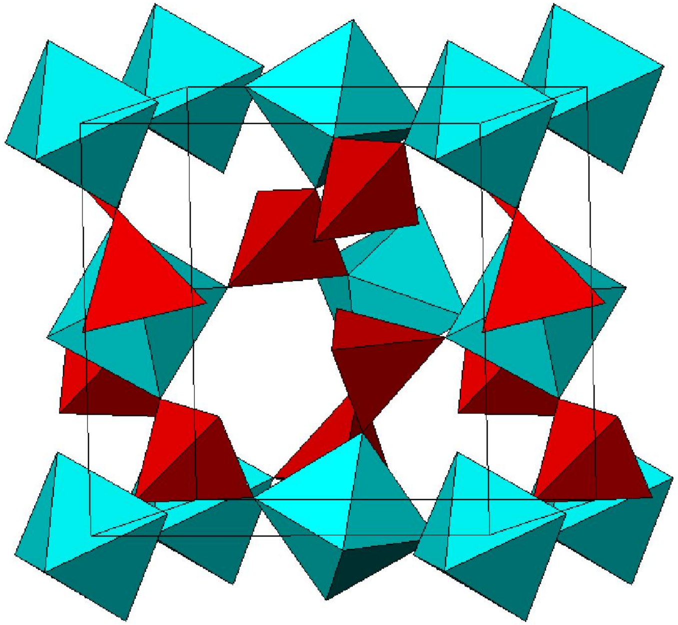
Low-energy modes in ZrW_2O_8 and NTE

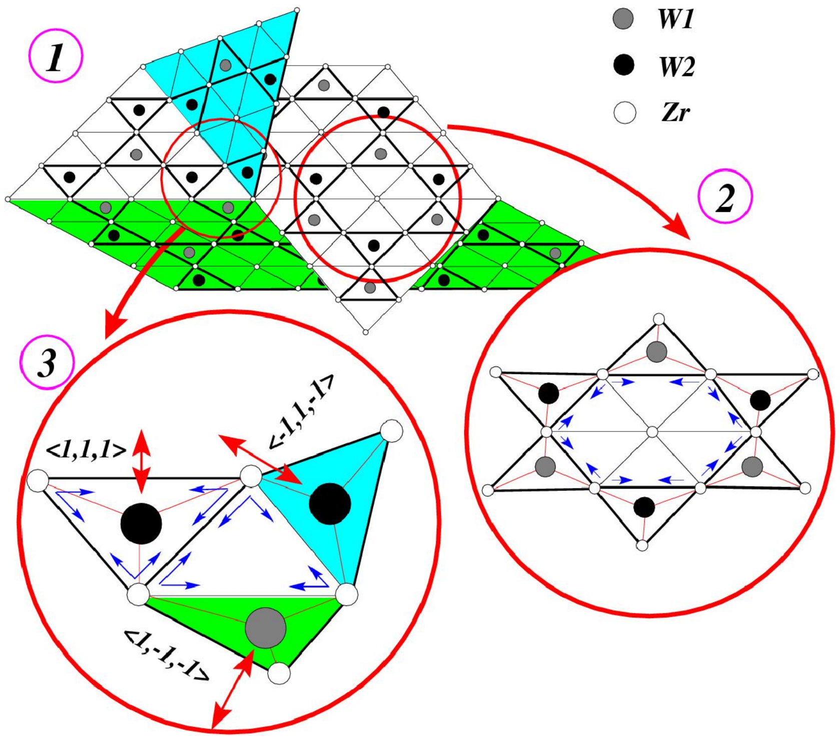


The low energy spectral weight explains NTE

Nature **396**, 147 (1998)

XAFS atomic motion in ZrW_2O_8

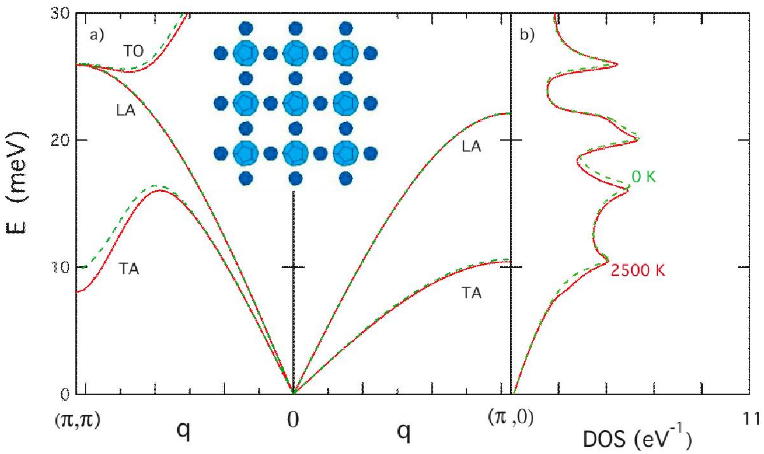


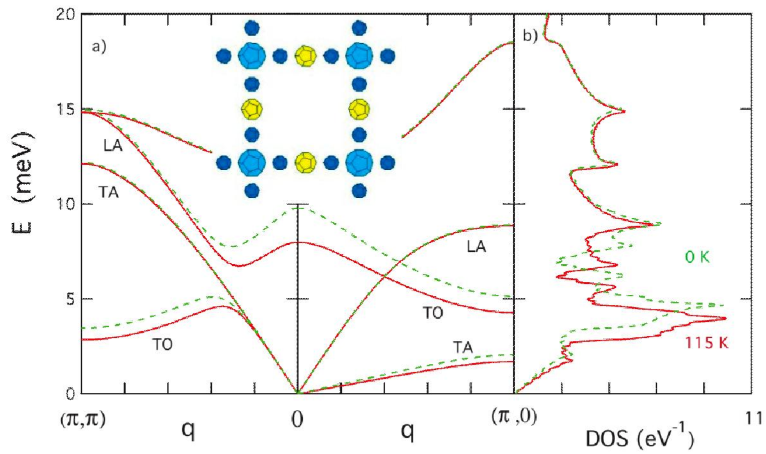


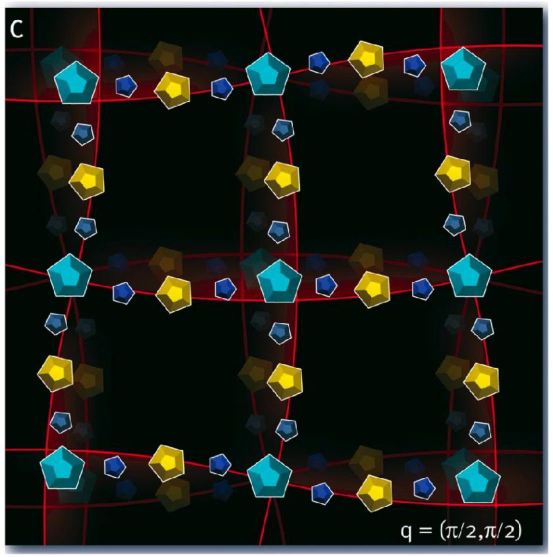
Cao, Bridges, Kowach & Ramirez – PRL 2002

Soft Manifold Dynamics behind Negative Thermal Expansion

Z. Schlesinger,[1] J. A. Rosen,[1] J. N. Hancock,[2] and A. P. Ramirez[3]
[1]*Physics Department, University of California Santa Cruz, Santa Cruz, California 95064, USA*
[2]*Geballe Laboratory for Advanced Materials and SSRL, Stanford University, Stanford, California 94305, USA*
[3]*Bell Laboratories, Alcatel-Lucent, 600 Mountain Avenue, Murray Hill, New Jersey 07974, USA*
(Received 13 October 2006; published 30 June 2008)







Frustration and Magnetoelectricity

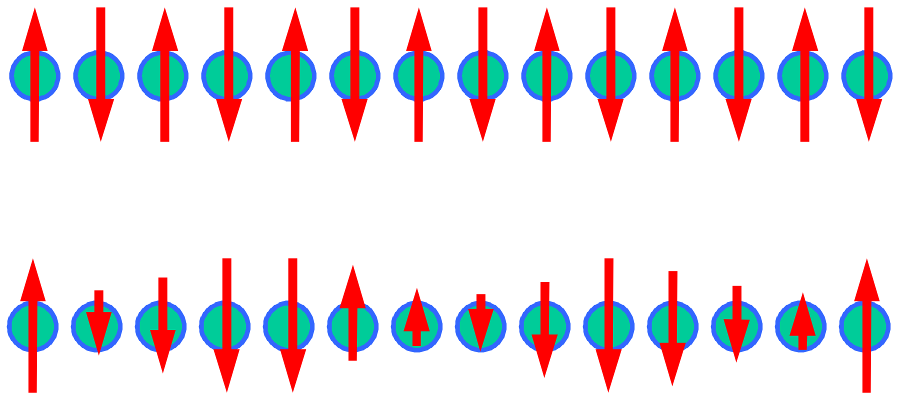
Antiferromagnets – Deviation from MF-T_c is common

Row no.	Substance	Chem. structure	Crystal sym. $T > T_N$	Mag. cat. structure	n_{eff}	T_N, °K	$-\theta_a/T_N$
1	VO	Rock salt	Cubic	f.c.c.	( )[a]	117	
2	CrN	Rock salt	Cubic	f.c.c.	( )[a]	~273	
3	MnO	Rock salt	Cubic	f.c.c.	5.95	122	5.0
4	α-MnS	Rock salt	Cubic	f.c.c.	5.6	130	3.1
5	β-MnS	Zinc blende	Cubic	f.c.c.	5.82	160	6.1
6	MnSe	Rock salt	Cubic	f.c.c.	5.7	~173	2.1
7	$Li_{0.1}Mn_{0.9}Se$	Rock salt	Cubic	f.c.c.	4.76	71[b]	−0.8
8	FeO	Rock salt	Cubic	f.c.c.	4.0[d]	198	~1.0[d]
9	CoO	Rock salt	Cubic	f.c.c.	5.1	291	1.1
10	NiO	Rock salt	Cubic	f.c.c.	4.6	520[c]	~5
11	TbP	Rock salt	Cubic	f.c.c.		9	
12	ErP	Rock salt	Cubic	f.c.c.		3.1	
13	TbAs	Rock salt	Cubic	f.c.c.		12	
14	TbSb	Rock salt	Cubic	f.c.c.	9.9	14	
15	HoSb	Rock salt	Cubic	f.c.c.		9	
16	ErSb	Rock salt	Cubic	f.c.c	9.8	3.7	
17	γ-Mn	f.c.c.	Cubic	f c.c.		660	
18	MnS_2	Pyrite	Cubic	f.c.c.	6.30	<77	>8
19	$MnSe_2$	Pyrite	Cubic	f.c.c.	5.93	~100	~4.8
20	$MnTe_2$	Pyrite	Cubic	f.c.c	6.22	80	6.5
20a	FeS_2	Pyrite	Cubic	f.c.c.			
20b	CoS_2	Pyrite	Cubic	f.c.c.	1.85	$T_c = 110$	
20c	NiS_2	Pyrite	Cubic	f.c.c.	3.19		
21	CrF_2	Dist. rut.	Mono.	b.c. mono.	4.9	53	
22	$CrCl_2$	Dist. rut.	Ortho.	b.c. ortho.	5.1	40[h]	2.7
23	MnF_2	Rut.	Tet. ($c/a < 1$)	b.c. tet.	5.7	72	1.6
24	FeF_2	Rut.	Tet. ($c/a < 1$)	b.c. tet.	5.6	79	1.5
25	CoF_2	Rut.	Tet. ($c/a < 1$)	b.c. tet.	5.13	37	1.4
25a	CuF_2	Dist. rut.	Mono.	b.c. mono.		78	
26	NiF_2	Rut.	Tet. ($c/a < 1$)	b.c. tet.	3.5	78.5–83	~2.0
27	VO_2	Rut.	Tet. ($c/a < 1$)	b.c. tet.	1.73	343	2.1

from J. B. Goodenough, “Magnetism and the Chemical Bond”,

UC SANTA CRUZ

One way for GFMs to order - Incommensurately



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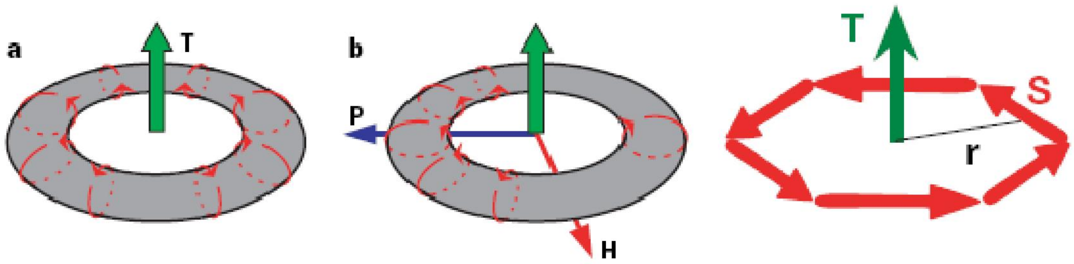
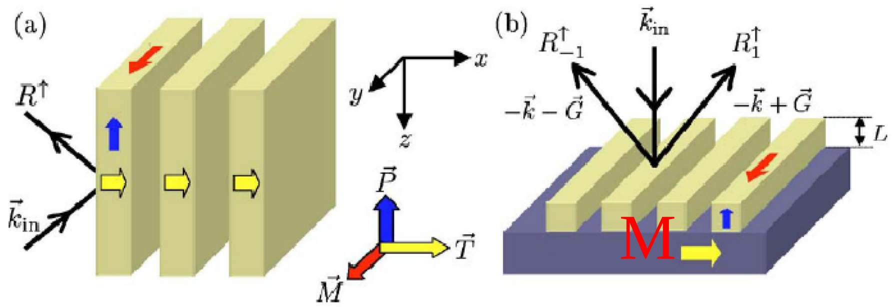
High interest in magnetoelectric materials

<u>Toroidal moments</u>



Fiebig et al

<u>Magneto-chiral effect</u>



Sawada & Nagaosa

<u>Electrical modulation of magnetic functionality</u>: magneto-optics, spin transistors →

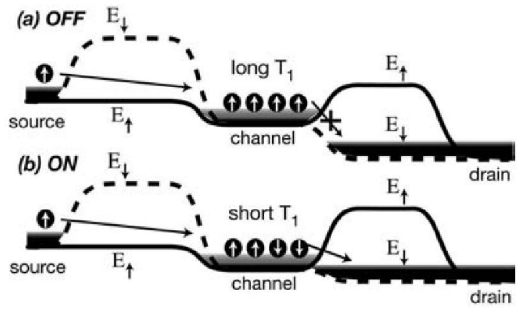
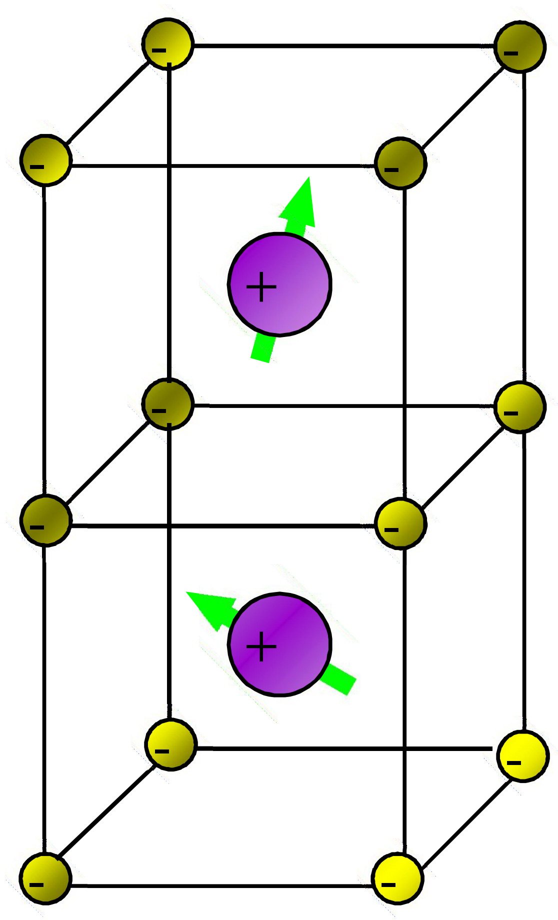
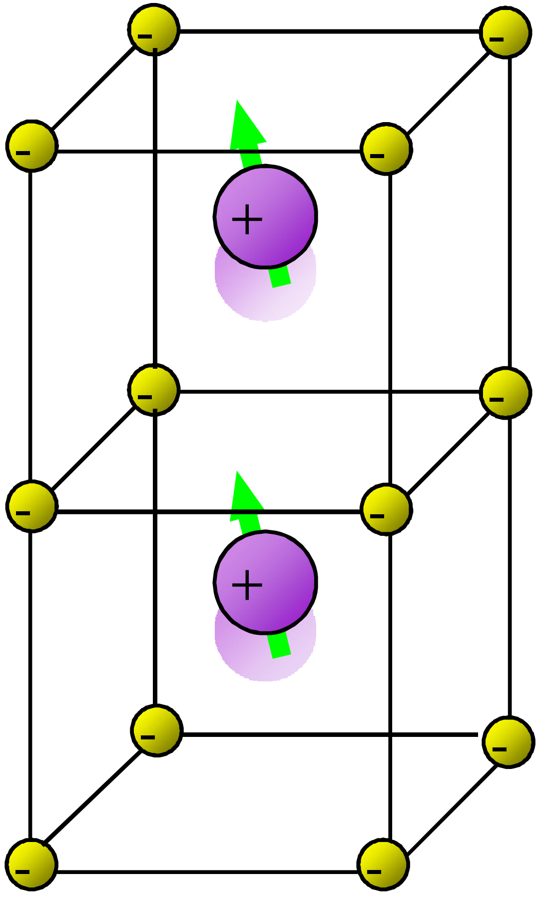


FIG. 2. Spin transistor in the (a) off and (b) on configurations. Hall & Flatte

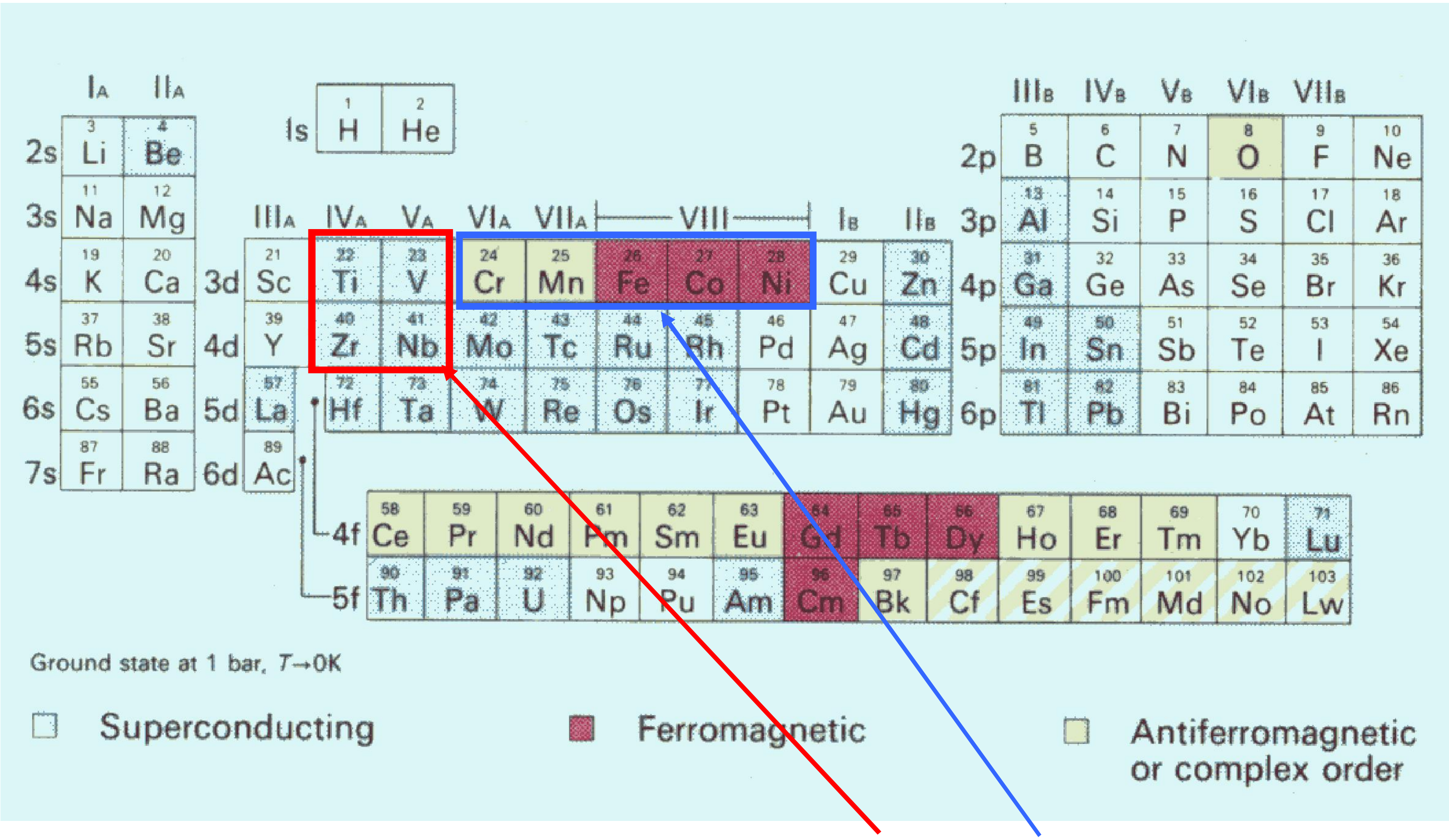
Uniform charge, disordered spins



Shifted charge, ordered spins



Multiferroics are Rare

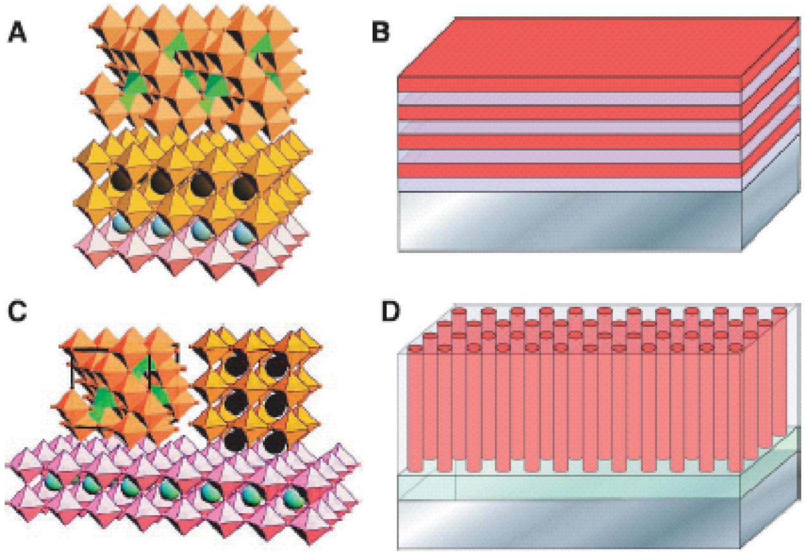


Look at common mineral types that combine FE and FM ions

Spinel: AB_2O_4; Perovskite: ABO_3; Pyrochlore: $A_2B_2O_7$ - hard to find A^{4+} and $B^{2,3+}$.

One approach to engineering new ME materials

Composite Multiferroic Materials



Multiferroic $BaTiO_3$-$CoFe_2O_4$ Nanostructures

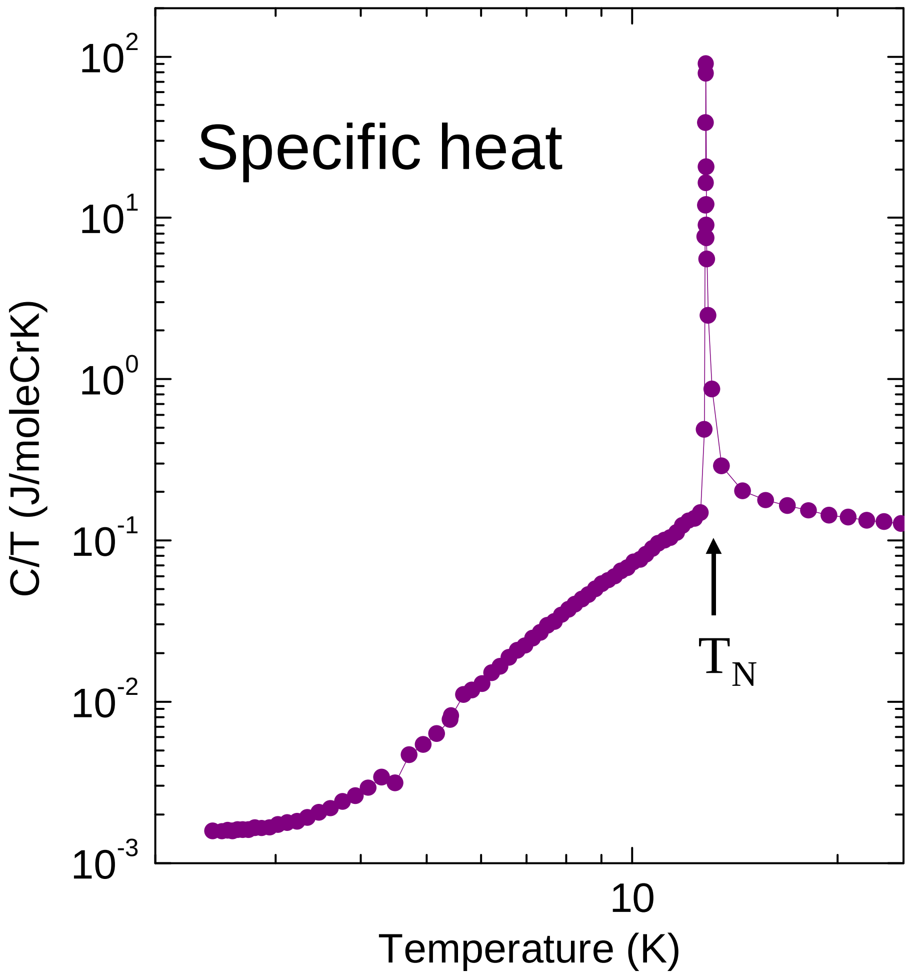
H. Zheng,[1] J. Wang,[1] S. E. Lofland,[3] Z. Ma,[1] L. Mohaddes-Ardabili,[1] T. Zhao,[1] L. Salamanca-Riba,[1] S. R. Shinde,[2] S. B. Ogale,[2] F. Bai,[4] D. Viehland,[4] Y. Jia,[5] D. G. Schlom,[5] M. Wuttig,[1] A. Roytburd,[1] R. Ramesh[1,2]

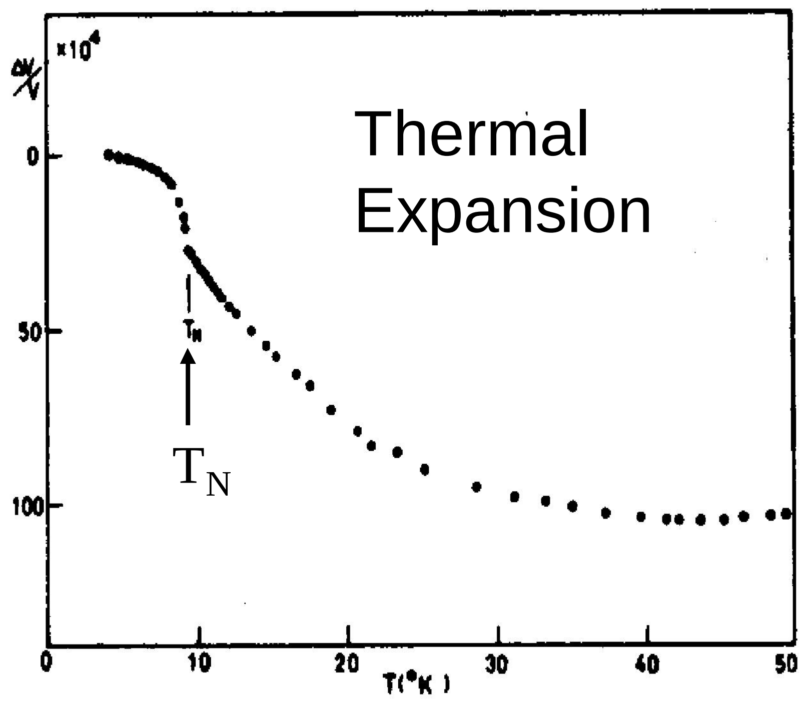
Science, 2004

Can one find an intrinsic mechanism for large ME?

Can a GF magnet induce a large lattice change?

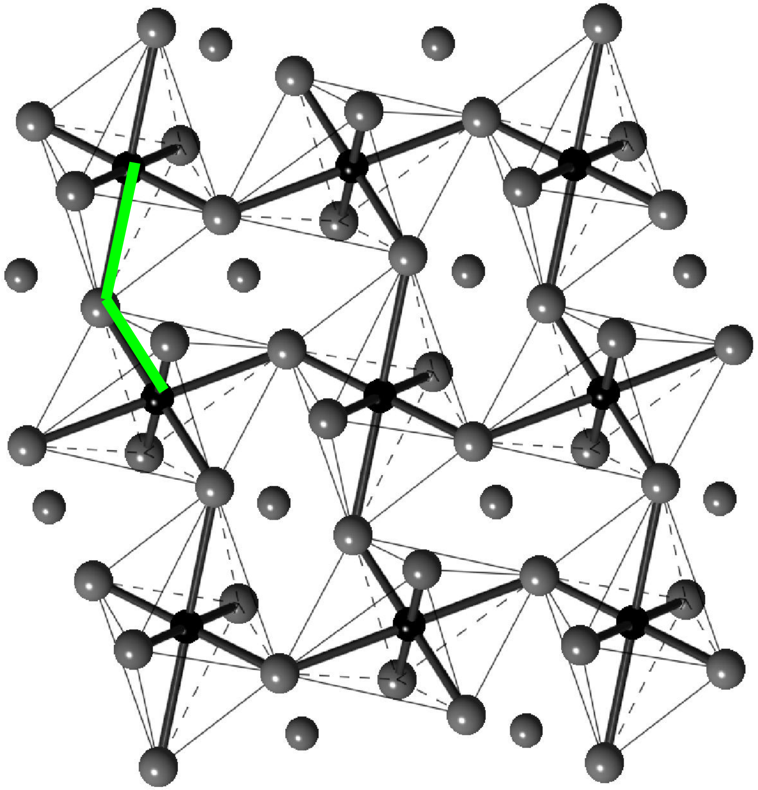
$ZnCr_2O_4$ $\theta_W \sim 400K$

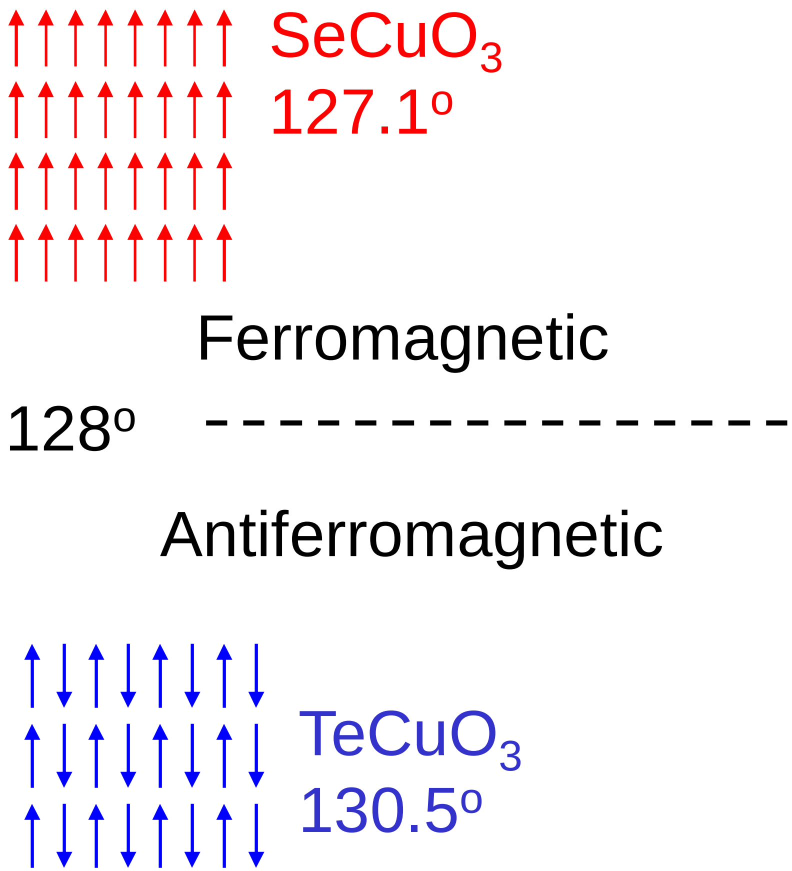




UC SANTA CRUZ

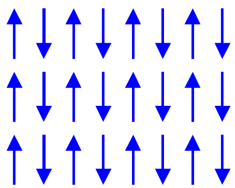
Can magnetism itself induce a non-centrosymmetric lattice? Look at $SeCuO_3$ and $TeCuO_3$

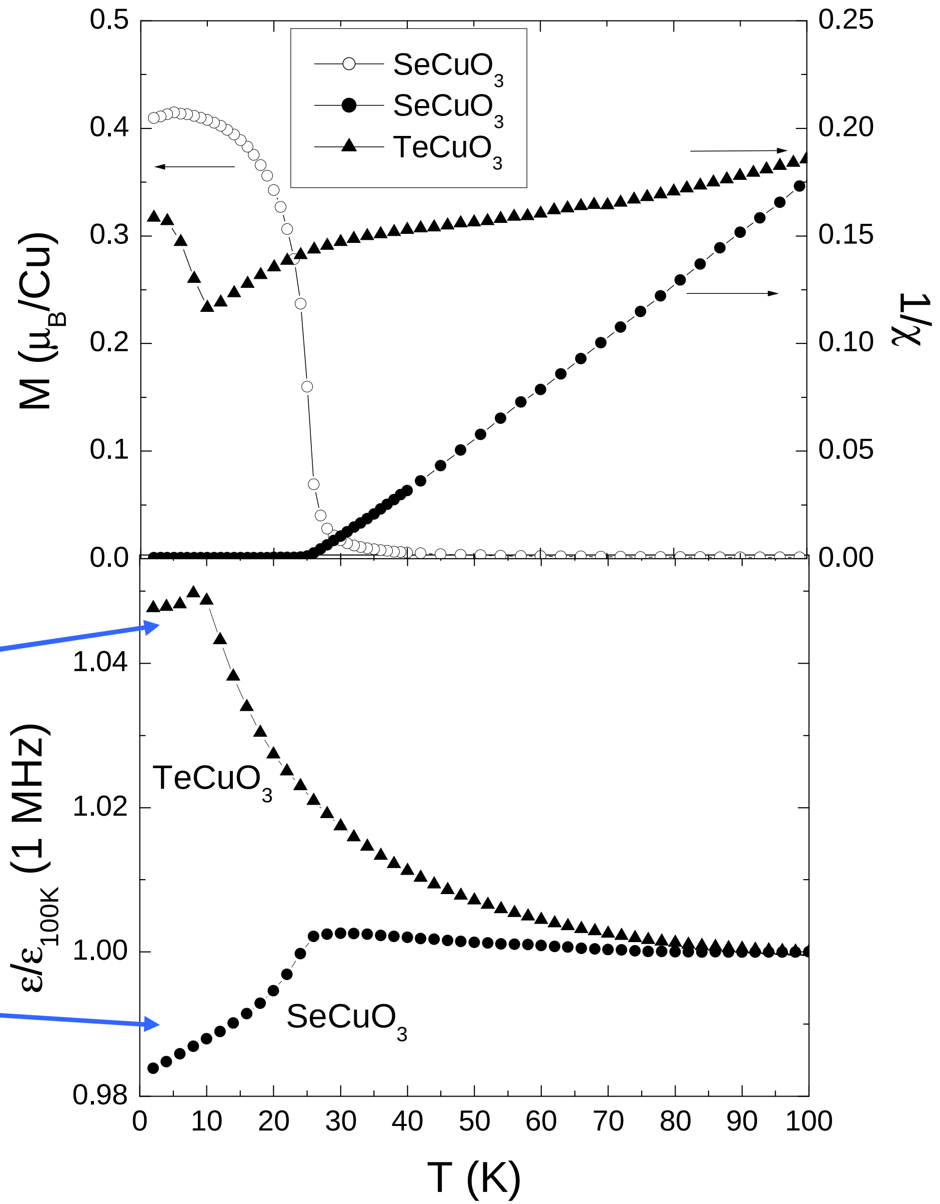


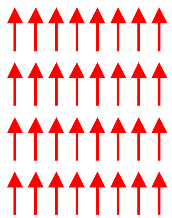


M Subramanian, APR, W. Maschall, *PRL*, 1999
G. Lawes, C. M. Varma, M. A. Subramanian, APR, *PRL* 2003

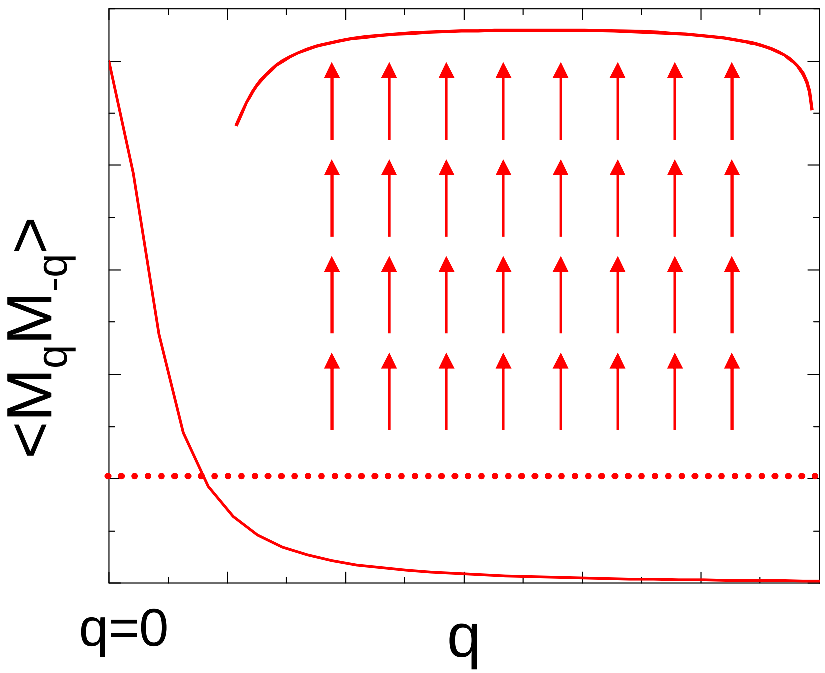
Magnetization and Dielectric Constant



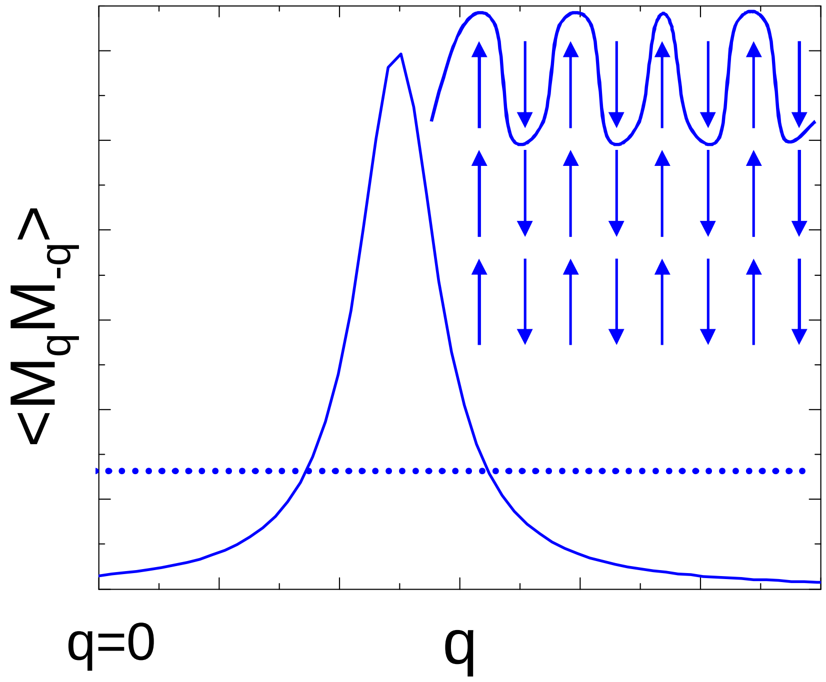




Spin Fluctuation Spectral Weight for FM and AF Ordering



Ferromagnet



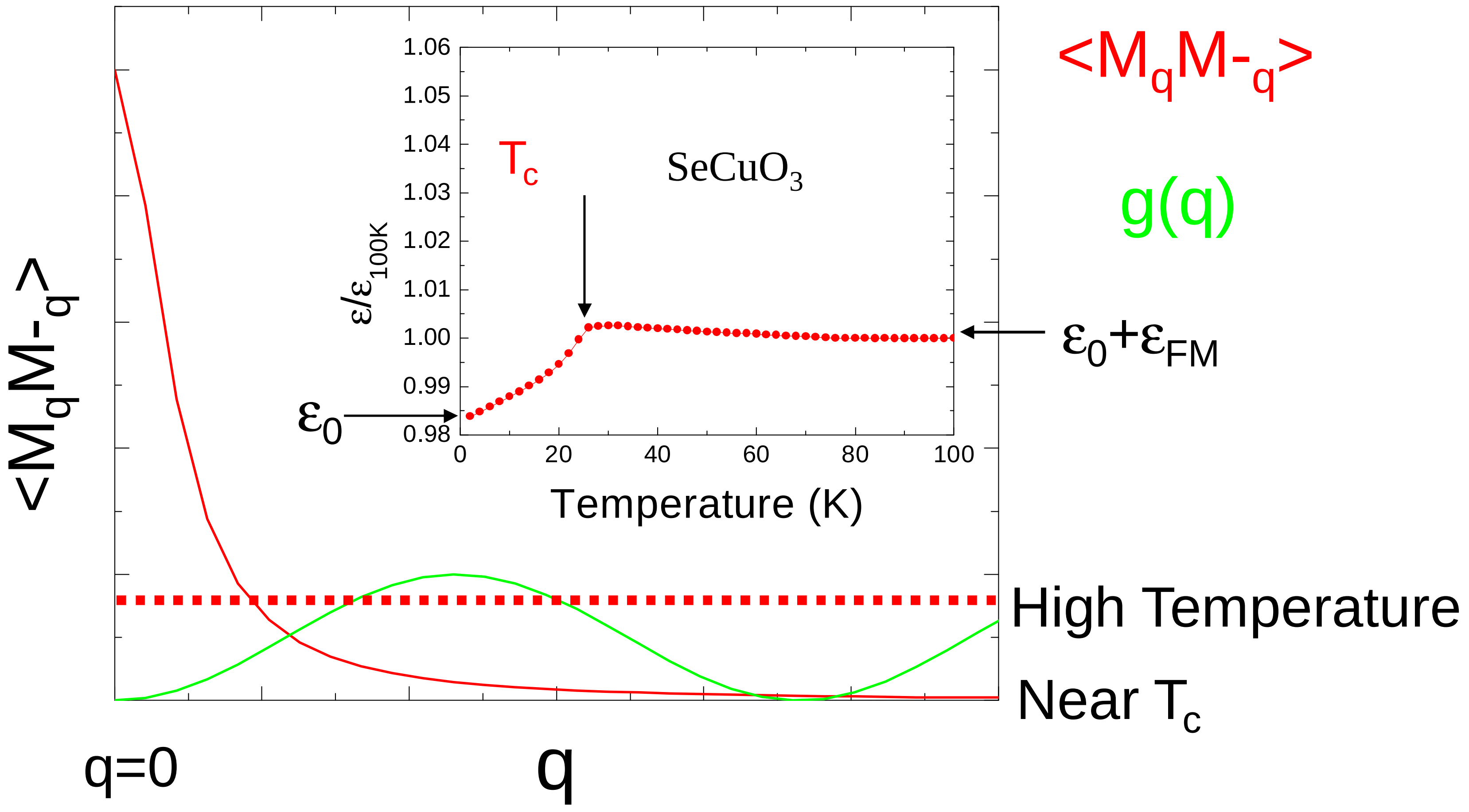
Antiferromagnet

Model for Magnetodielectric Coupling

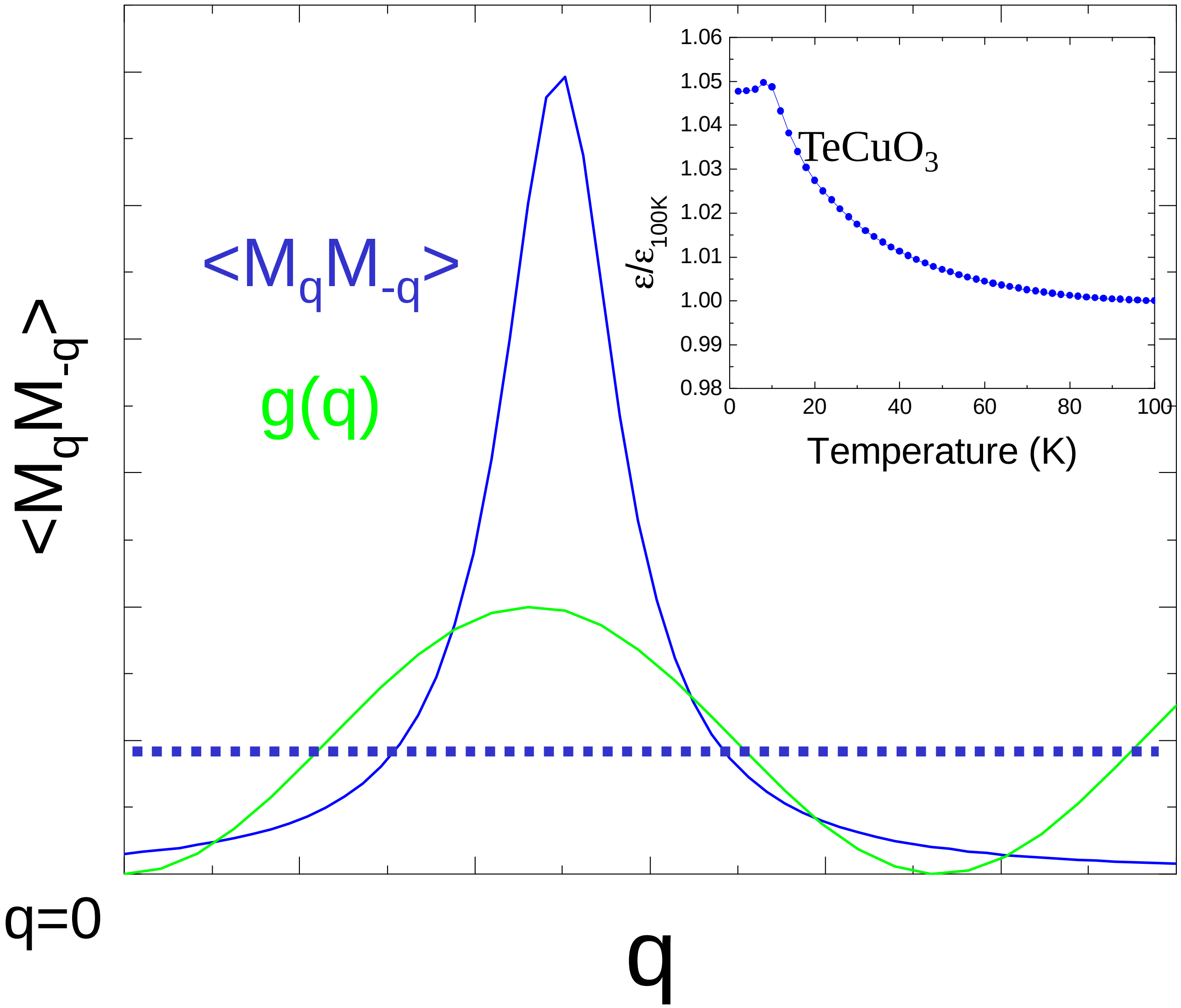
- "Standard" coupling: $F_{MDE}(\mathbf{M},\mathbf{P})=\alpha M^2P^2$
- Allow magnetodielectric coupling parameter to have q-dependence.

C. Varma

Ferromagnetic spin correlations and g(q)



Antiferromagnetic spin correlations and g(q)



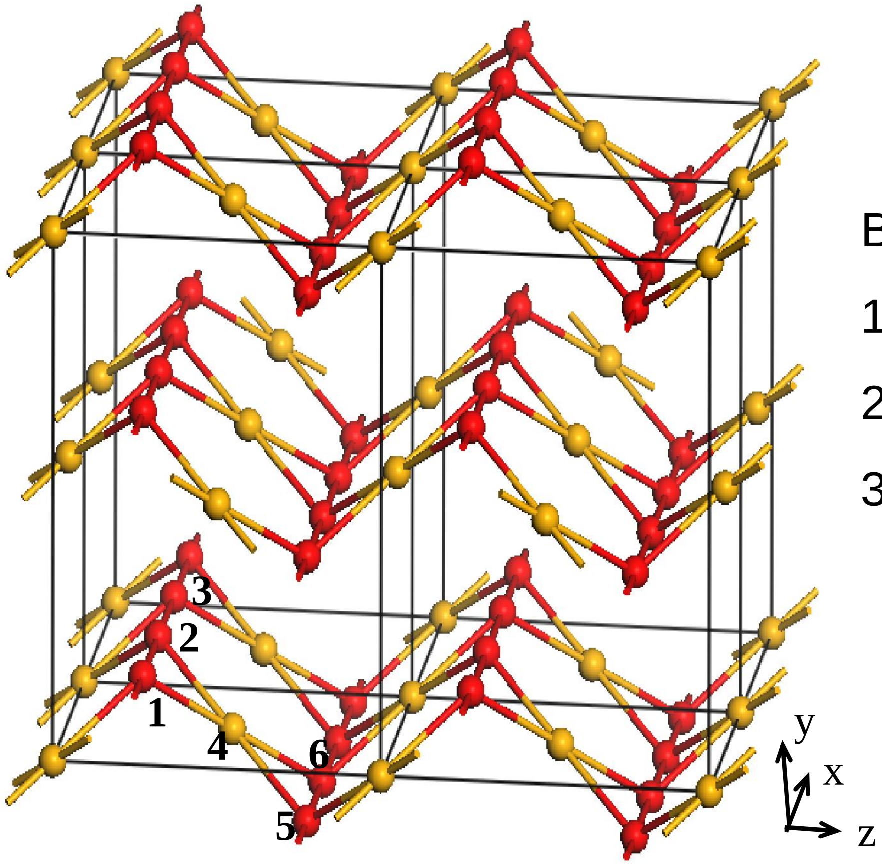
UC SANTA CRUZ

Can we find a magnetic system where the AF-like ordering induces non-centrosymmetric lattice displacements?

Competing Phases on a Kagome Staircase

$Ni_3V_2O_8$

Ni sites only shown

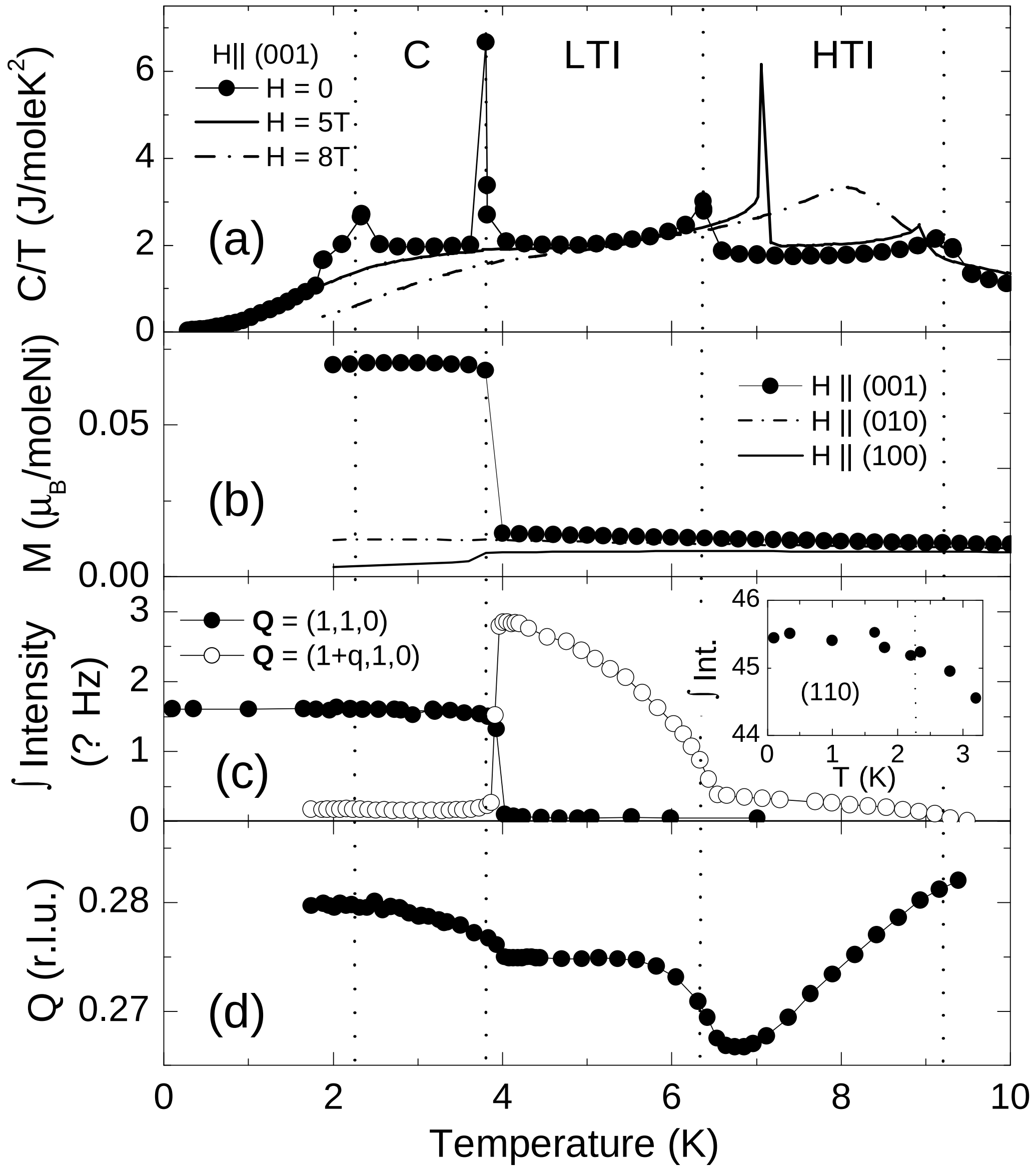


Buckling introduces

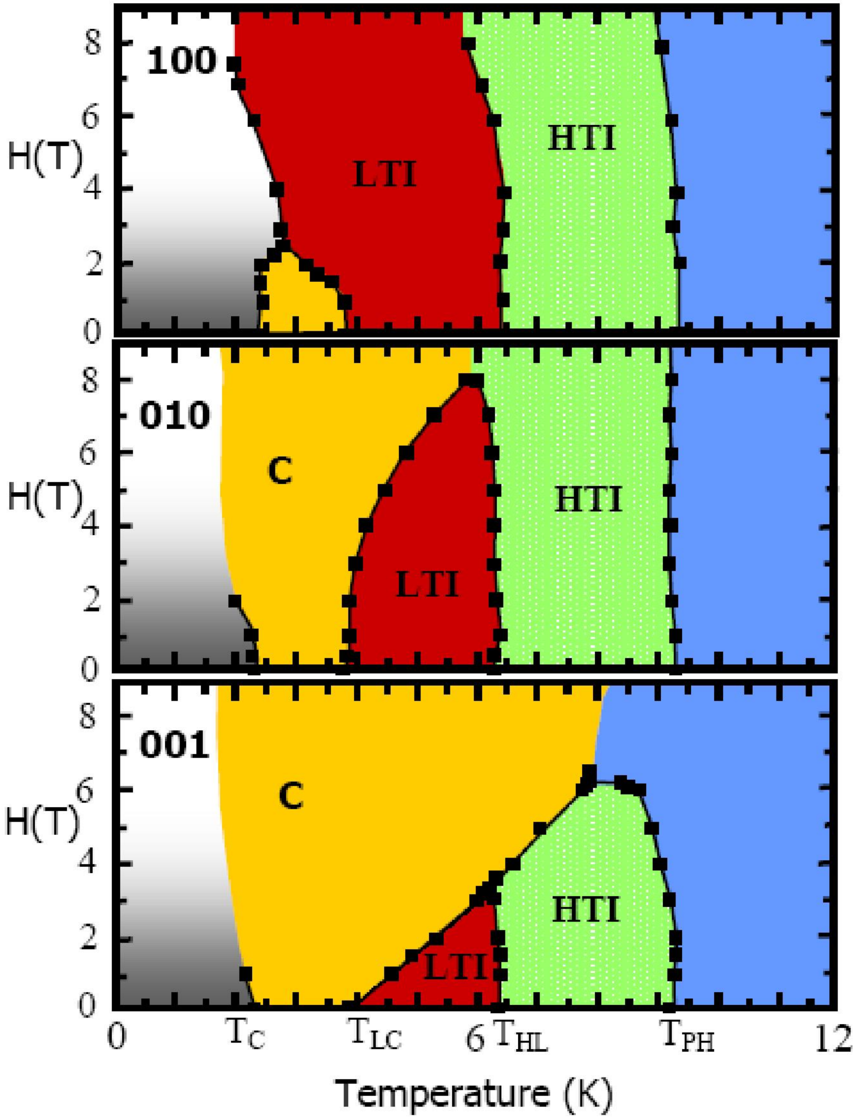
1) AF-FM
2) DM
3) Pseudo-DP

G. Lawes, M. Kenzelman, N. Rogado, K.H. Kim, G. Jorge, R. J. Cava, A. Aharony, O. Entin-Wohlman, A. B. Harris, T. Yildirim, Q. Z. Huang, C. Broholm, and APR, *PRL* 2004

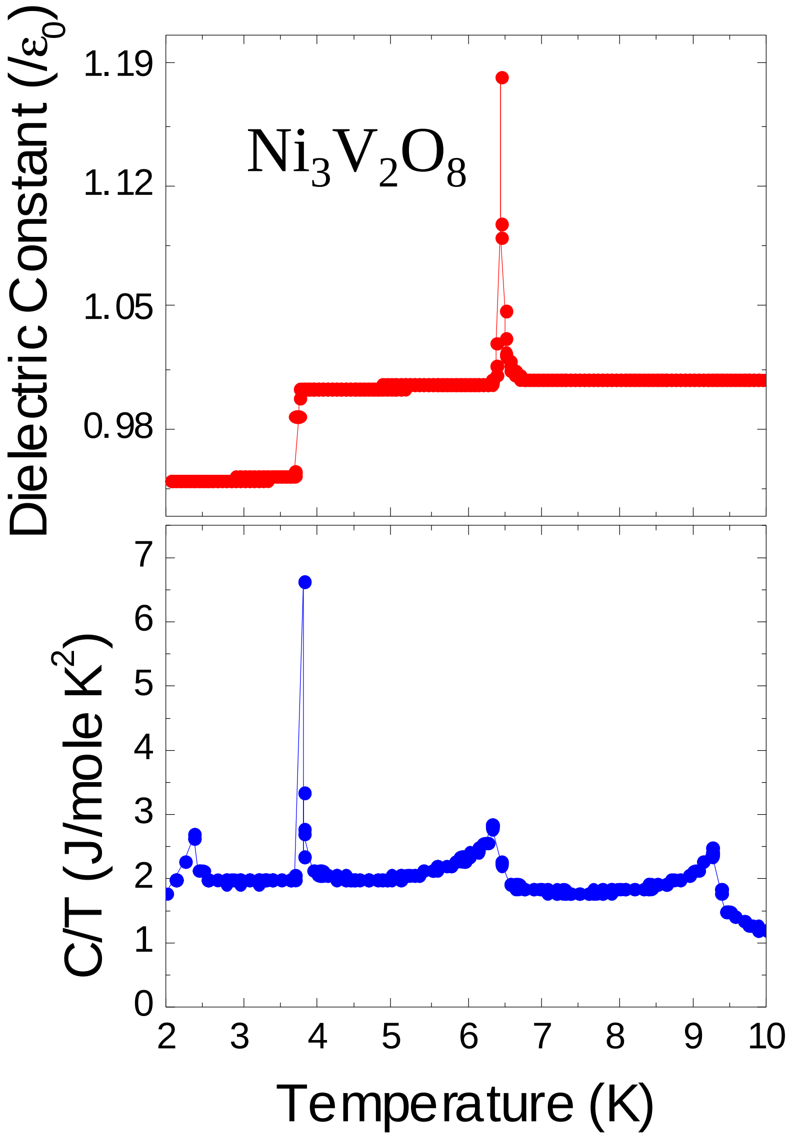
Thermal, Magnetic and Neutron Properties of $Ni_3V_2O_8$



Magnetic Phase Diagram of $Ni_3V_2O_8$



So have IC state – what about the dielectric response?



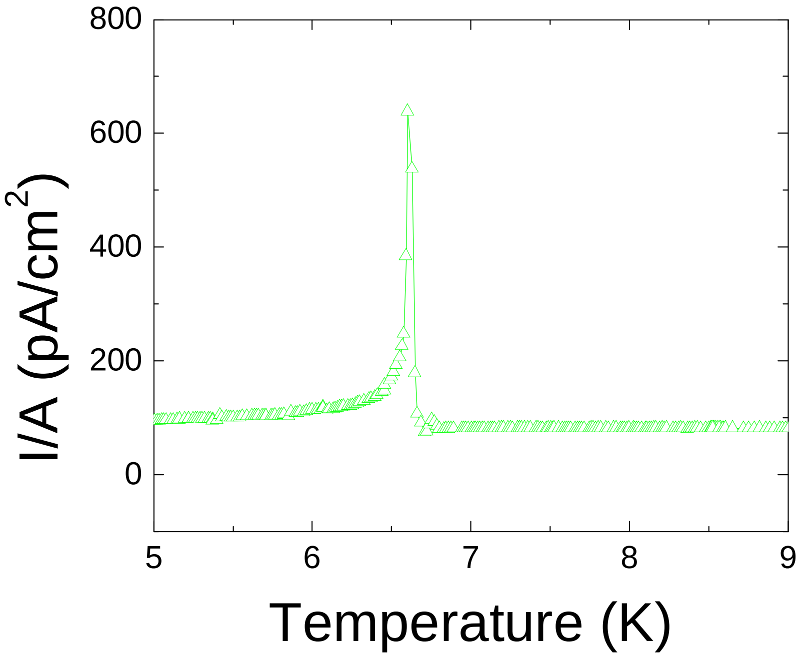
Two features in dielectric constant:

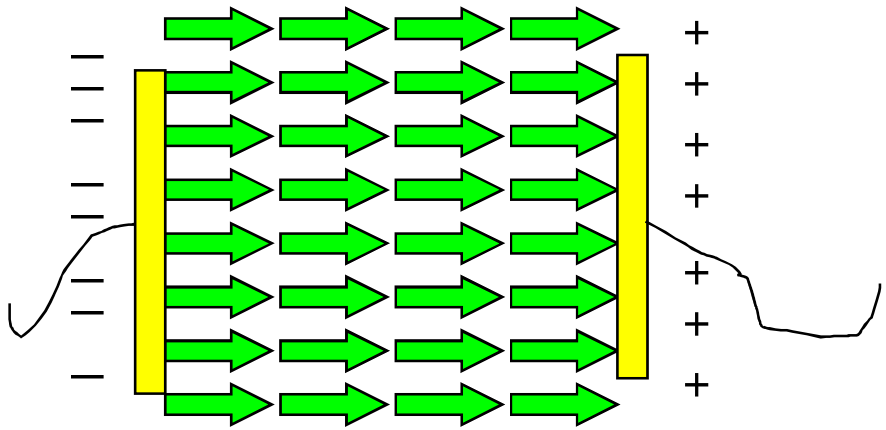
•Divergent electric susceptibility at T=6.4K

•Sharp drop at T=3.9K (consistent with magnetodielectric effect at ferromagnetic ordering)

Measuring Ferroelectricity

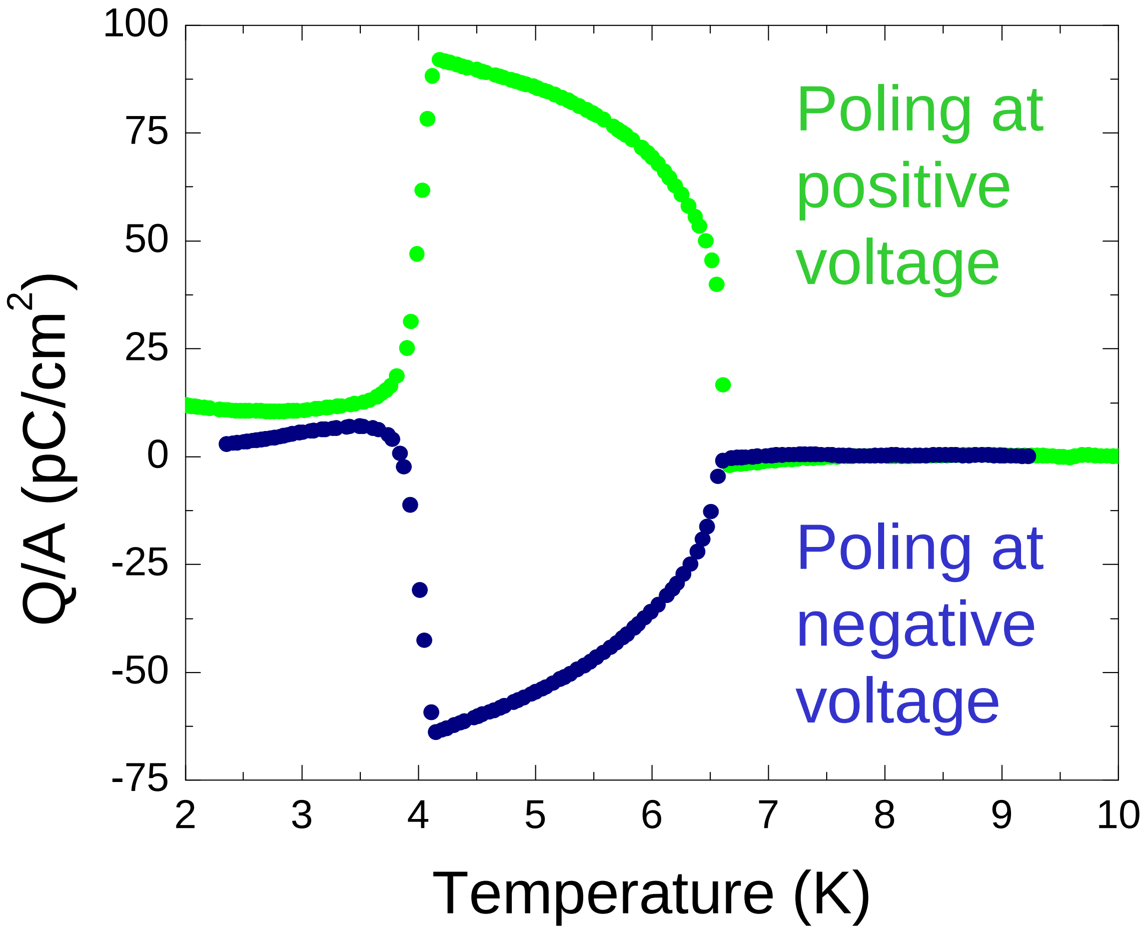
•The spontaneous polarization at a ferroelectric leads to a charge buildup at the boundaries.
•This produces a temperature dependent current.





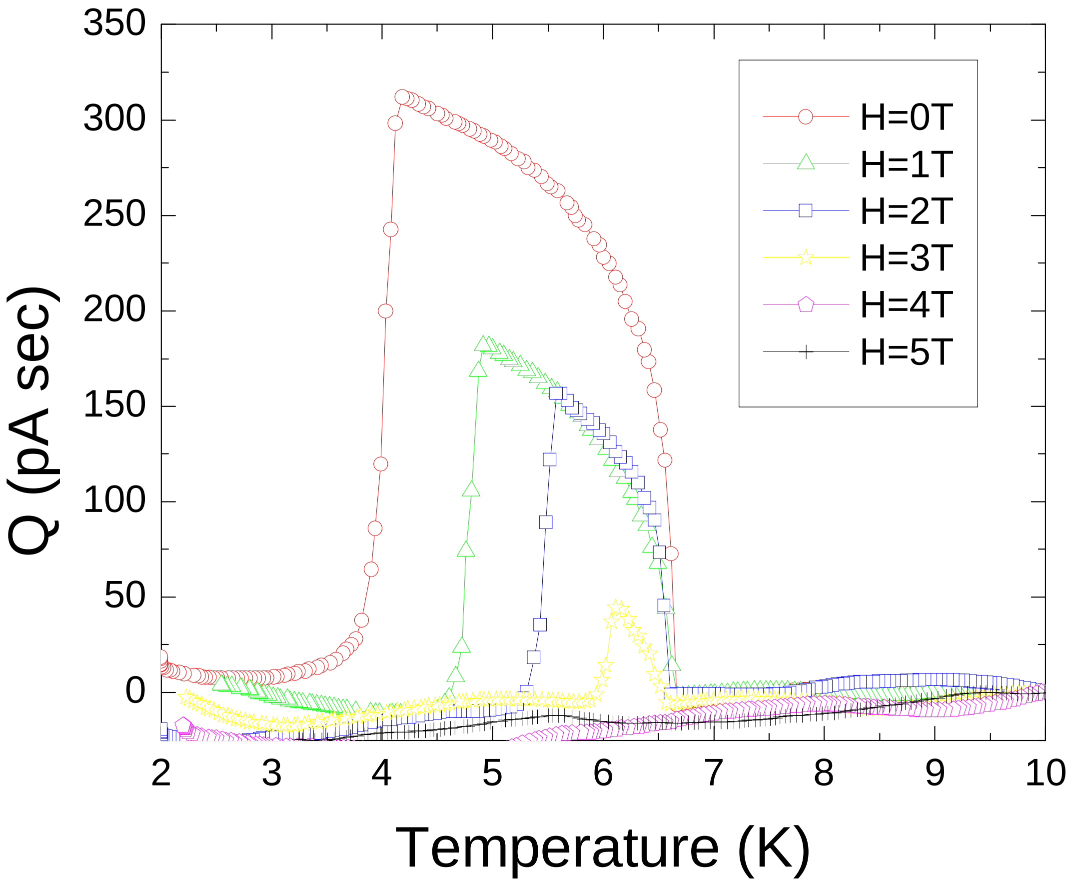
•Integrating this pyrocurrent against time gives the spontaneous polarization.
•Necessary to pole (align) ferroelectric domains

Spontaneous Polarization of $Ni_3V_2O_8$



- Spontaneous polarization switches direction upon reversing electric field

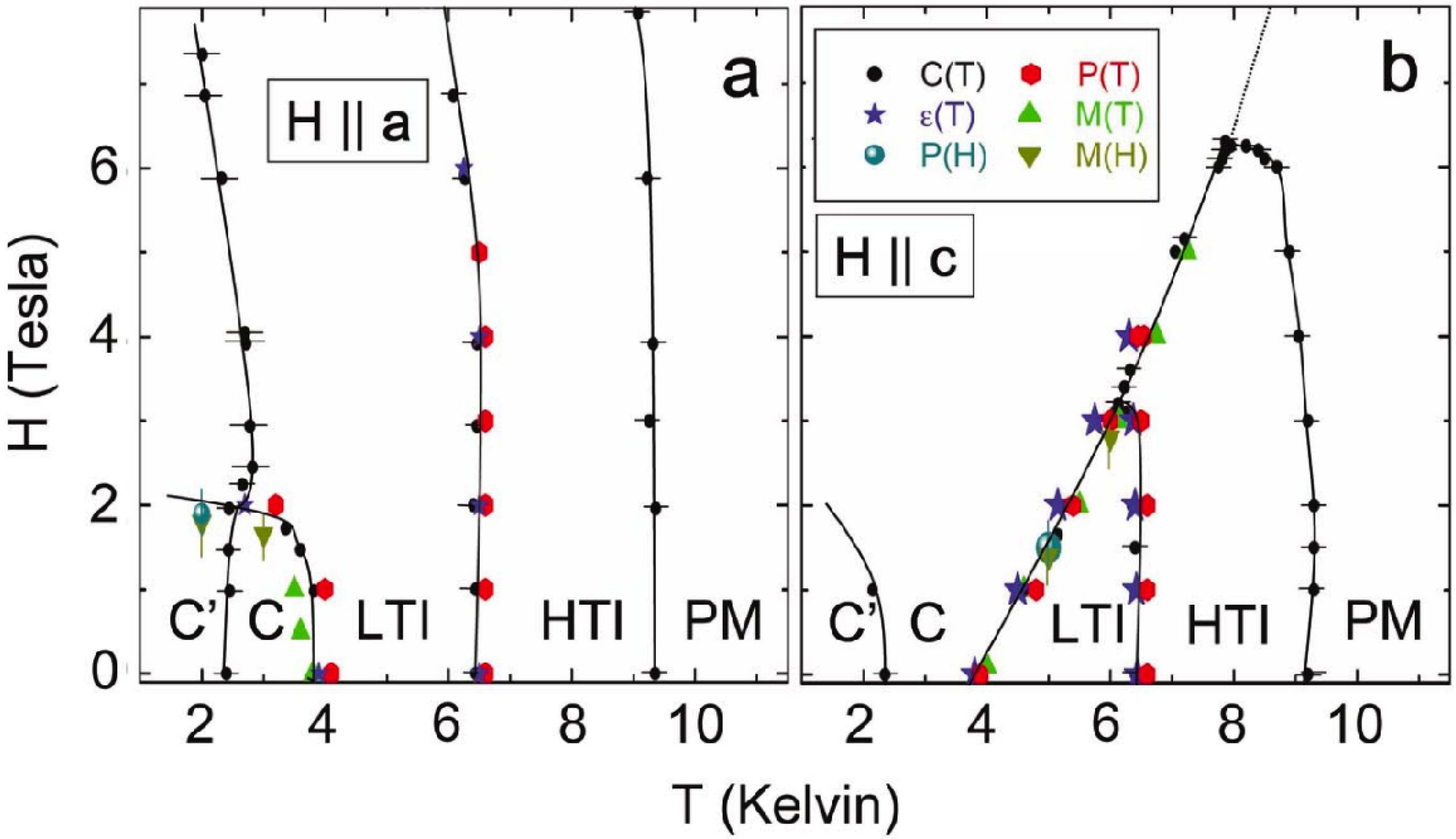
Magnetic Field Dependence of Ferroelectric Transition



- Ferroelectric transition in $Ni_3V_2O_8$ is very sensitive to magnetic field.
- Ferroelectric order is suppressed completely for fields larger than H=4T.

UC SANTA CRUZ

Induced multiferroic (FE/FM) phase boundary



UC SANTA CRUZ

How to understand the FE state in $Ni_3V_2O_8$?

Magnetic state breaking inversion symmetry:

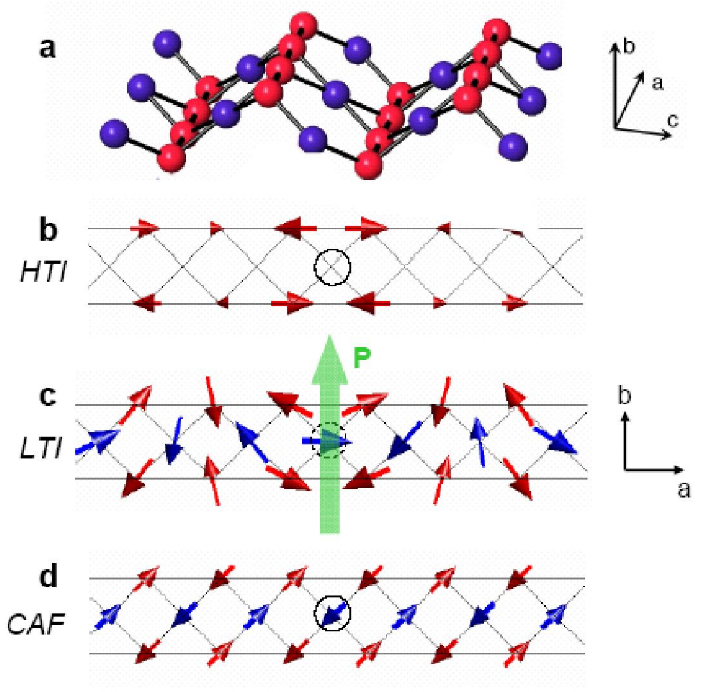
-Axial/Non-Axial Parity Breaking (B. Harris)

-Wavevector conservation forbids bilinear coupling

-Need a trilinear term in a Landau expansion

-

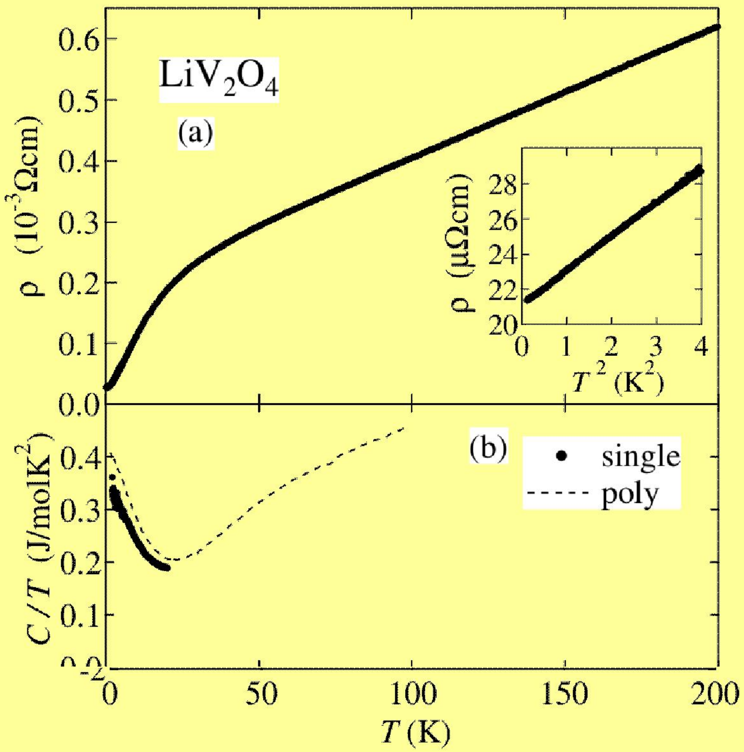
$$V_{LTI} = -\sum_{ij\gamma}\left[a_{ij\gamma}\sigma_{H;i}(k)\sigma_{L;j}(-k) + a^*_{ij\gamma}\sigma_{H;i}(-k)\sigma_{L;j}(k)\right]P_\gamma$$



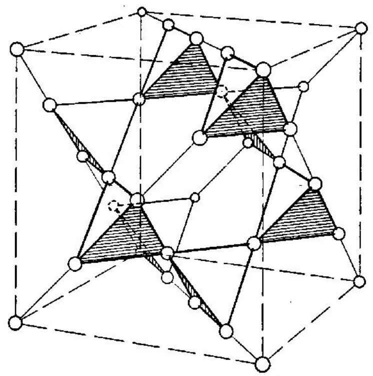
G. Lawes, A. B. Harris, T. Kimura, N. Rogado, R. J. Cava, A. Aharony, O. Entin-Wohlman, T. Yildirim, M. Kenzelmann, C. Broholm, and A. P. Ramirez, *PRL*

Frustration and Itinerant Charge

Heavy Fermions from GF Magnetism?



al, Takagi, et al, PRL 2000



Cf: also $LiTi_2O_4$ T_c~14K

feature article

The quantum spin Hall effect and topological insulators

Xiao-Liang Qi and Shou-Cheng Zhang

In topological insulators, spin–orbit coupling and time-reversal symmetry combine to form a novel state of matter predicted to have exotic physical properties.

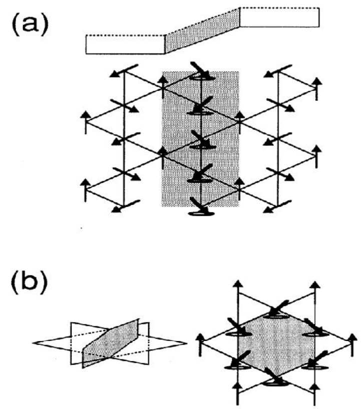
PHYSICAL REVIEW B **78**, 125104 (2008)

Band touching from real-space topology in frustrated hopping models

Doron L. Bergman,[1] Congjun Wu,[2] and Leon Balents[3]

Three-Dimensional Topological Insulators on the Pyrochlore Lattice

H.-M. Guo and M. Franz
Department of Physics and Astronomy, University of British Columbia, Vancouver, BC, Canada V6T 1Z1
(Received 5 August 2009; published 13 November 2009)



Ritchey, Coleman, Chandra

Our main finding here is that, quite generically, whenever electrons hopping on the pyrochlore lattice acquire a band gap from SO interactions the resulting state is either a STI or a weak topological insulator (WTI), defined as a

Summary

- Geometrical frustration is a paradigm for strongly correlated matter
- Emergent phenomena include new states of matter; spin liquid, ice
- Novel excitations; monopoles, Dirac strings
- Unique responses to external perturbations, NTE, magnetoelectrics
- Possible route to topological insulators

Collaborators:

GFM:
G. Aeppli
C. Broholm
E. Bucher
R. Cava
P. Schiffer
C. Kloc
G. Espinosa
A. S. Cooper
S. W. Cheong
D. Huse
D. Bishop
P. Gammel
S. Shastry
S. Rosenkranz
G. Lawes

NTE:
G. Ernst
G. Kowach
F. Bridges
C. Varma
C. Broholm

Large-ε:
M. Subramanian
G. Blumberg
S. Shapiro
T. Vogt
C. Varma
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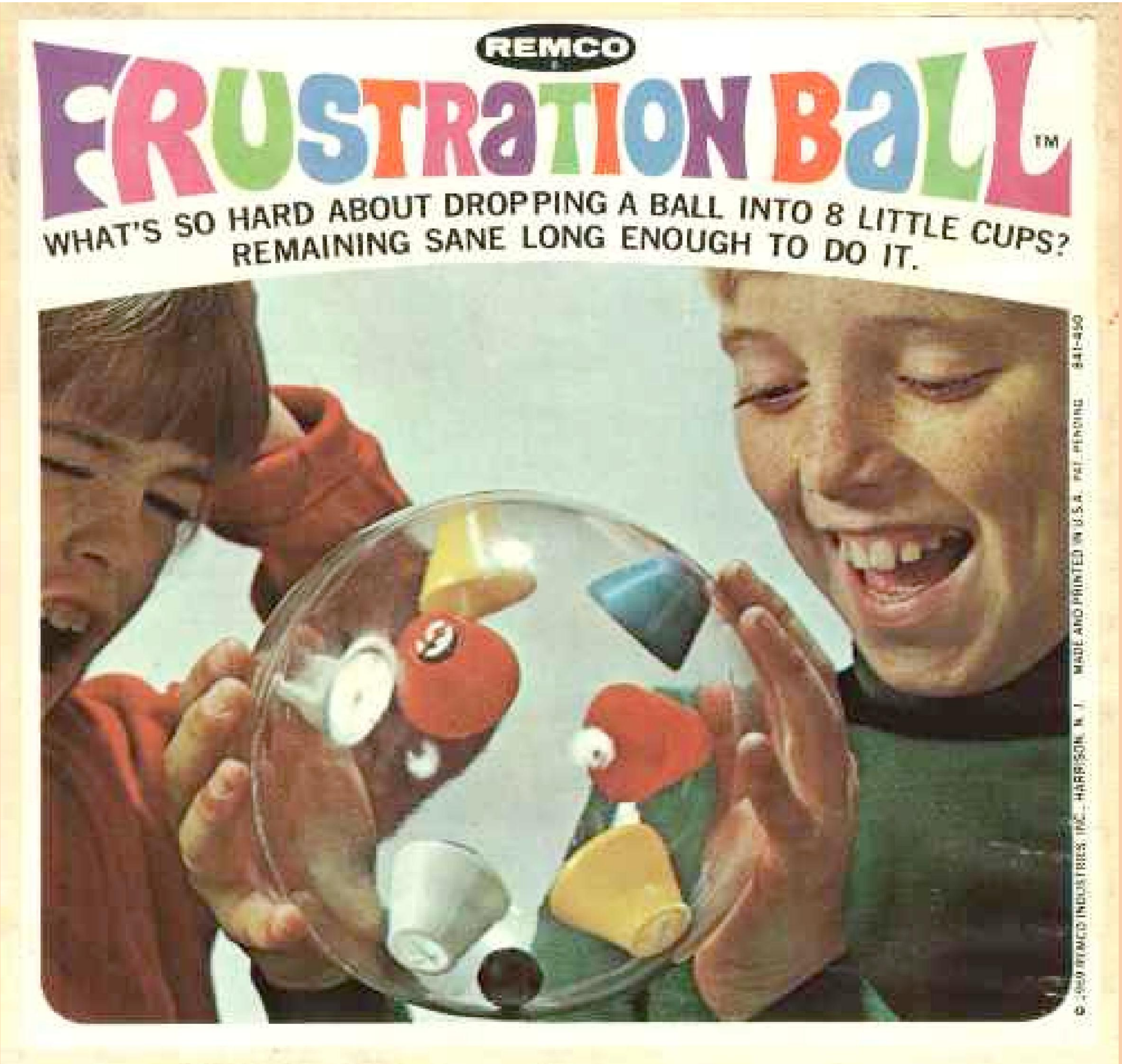
Multiferroics:
G. Lawes
T. Kimura
C. Broholm
A. Aharony
A. B. Harris
O. Entin-Wohlman
M.Kenzelmann
T. Yildirim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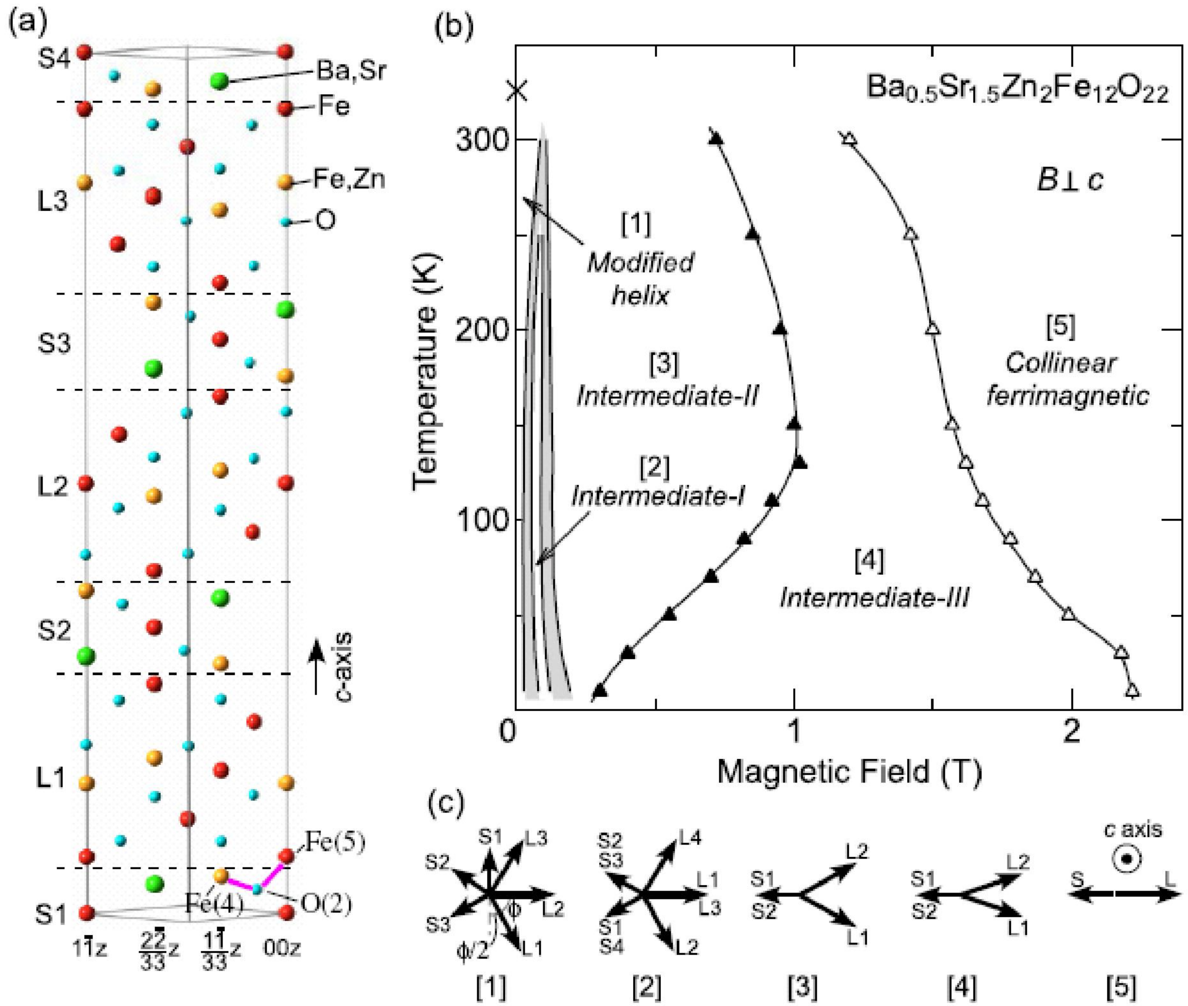
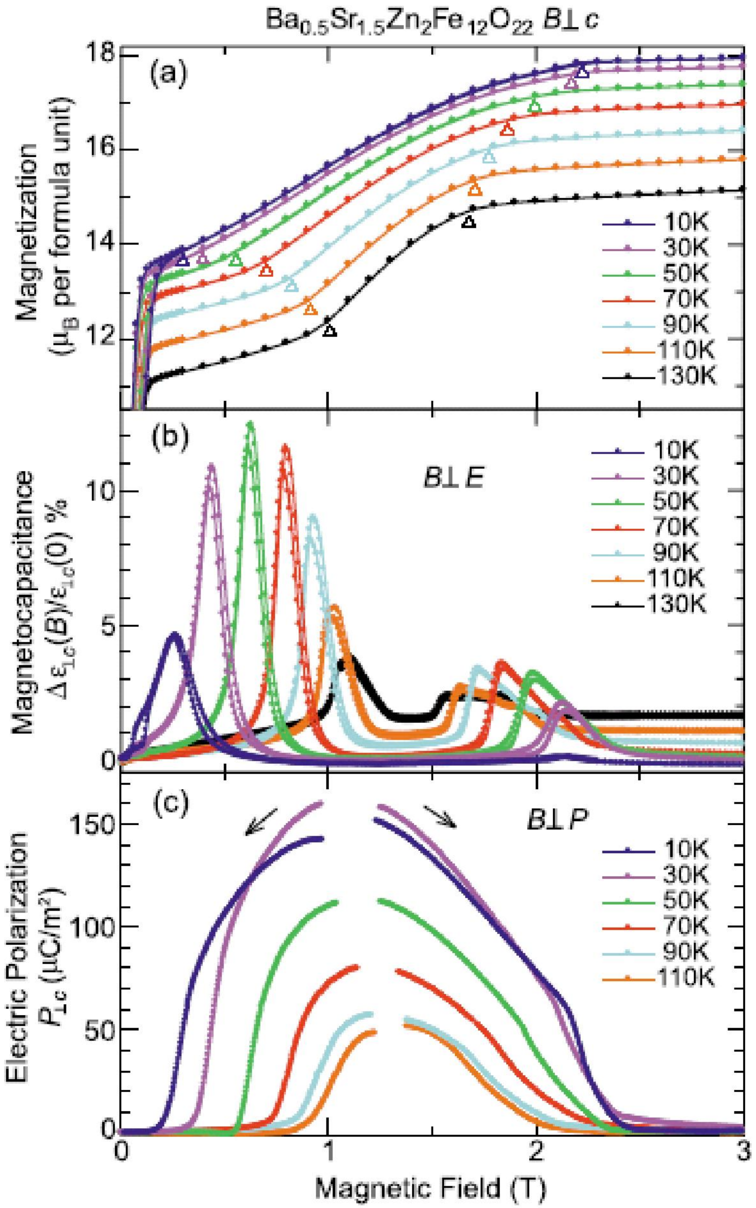
REMAINING SANE LONG ENOUGH TO DO IT.
REMCO INDUSTRIES INC., HARRISON, N. J.
MADE AND PRINTED IN U.S.A.

Above examples were all at low-T – Can we find higher-T manifestations?

Search for higher-T Incommensurate magnets



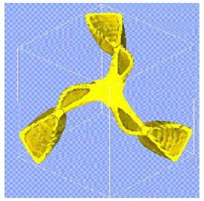
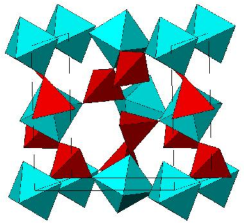
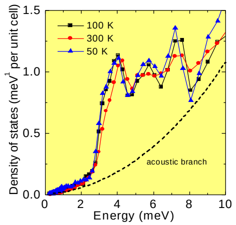
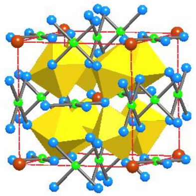
T. Kimura, G. Lawes, APR, *PRL*, 2005



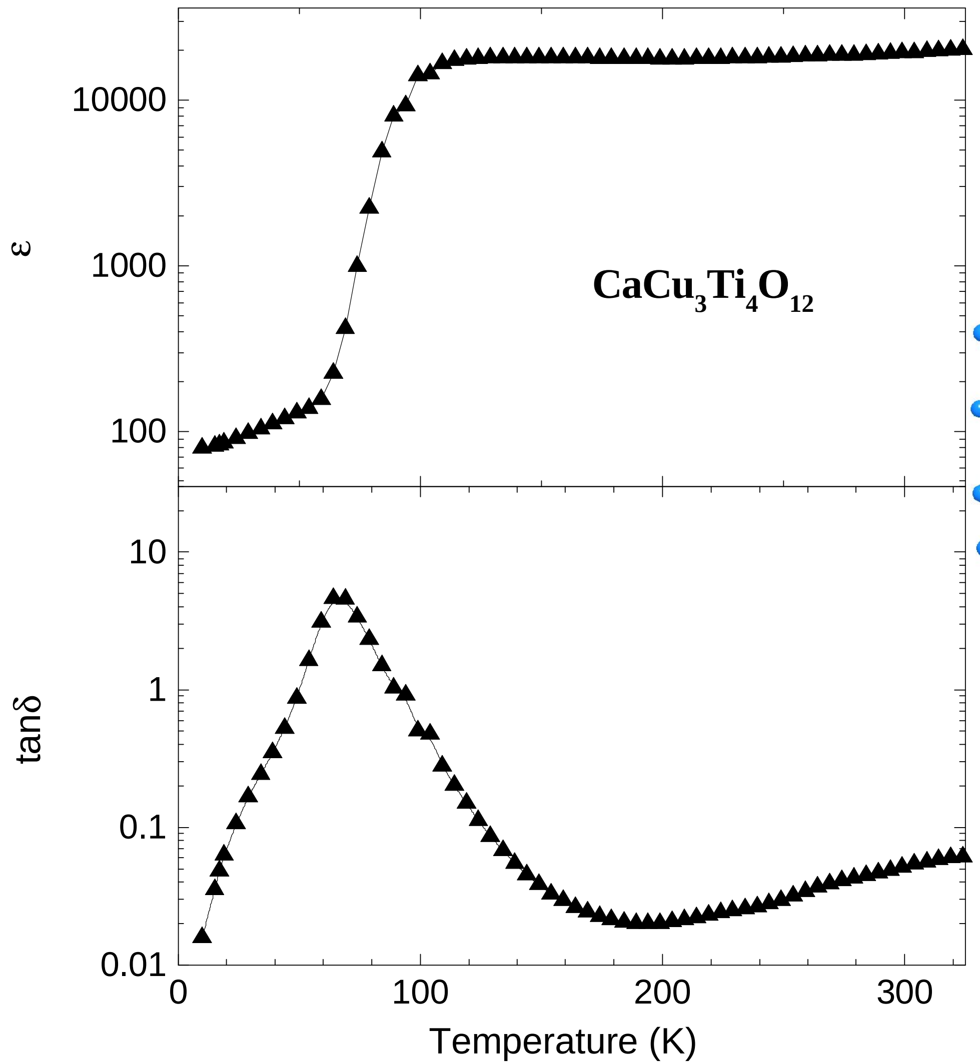
More work to do in understanding materials possibilities and potential application of multiferroics.

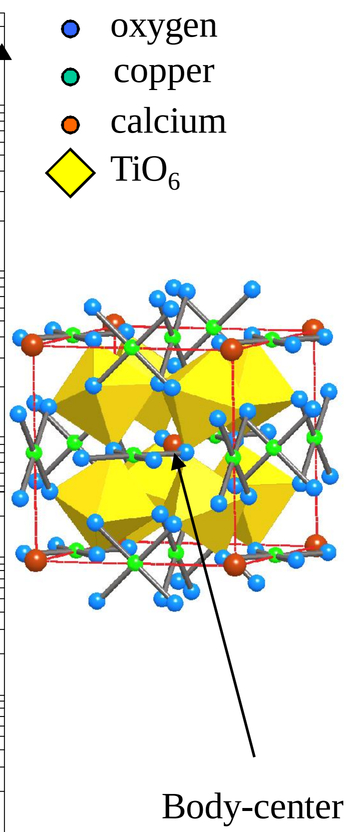
Summary

Summary and Conclusions

	symmetry incompatibility	marginal constraint	spect. weight downshift
GFMs	yes	yes	yes
Neg. Th. Exp. ZrW_2O_8			
Large Diel. Const. $CaCu_3Ti_4O_{12}$		?	?

Dielectric response of $Cu_3Ti_4O_{12}$





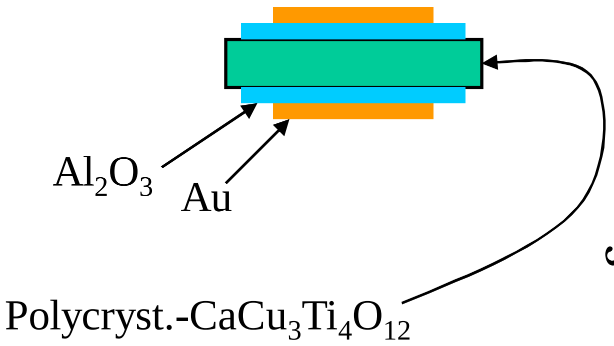
SIA/ITRS Tables of Requirements

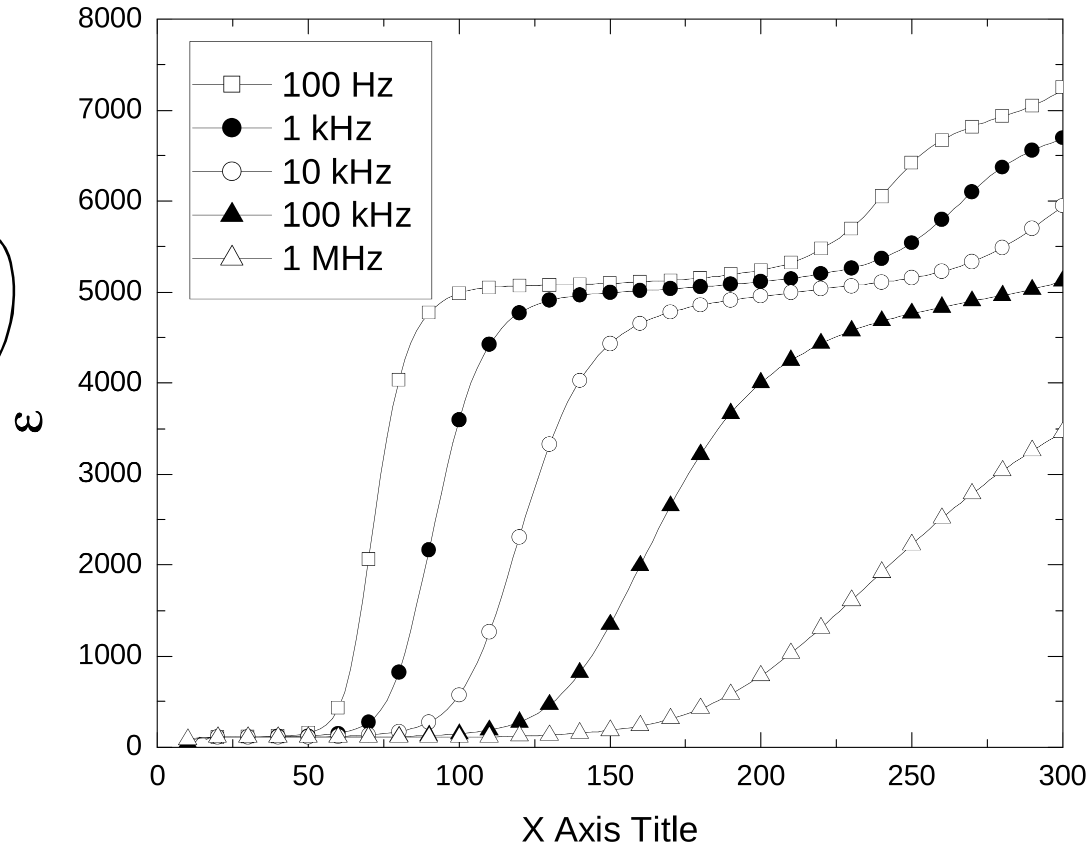
- ITRS consists of 65 tables of requirements: Red Indicates that there are “No Known Solutions”

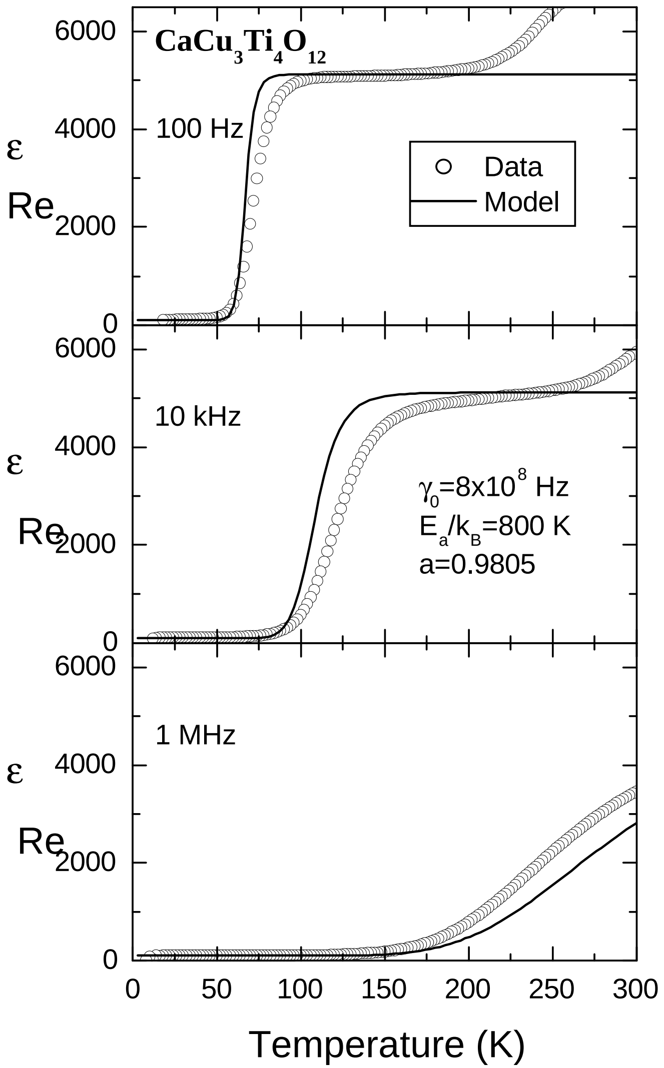
Year of First Product Shipment Technology Generation	***1999 180 nm***	***2000 165 nm***	***2001 150 nm***	***2002 130 nm***	***2003 120 nm***	***2004 110 nm***	***2005 100 nm***
DRAM Half Pitch (nm)	180	165	150	130	120	110	100
MPU Gate Length (nm)	140	120	100	85	80	70	65
MPU / ASIC Half Pitch (nm)	230	210	180	160	145	130	115
ASIC Gate Length (nm)	180	165	150	130	120	110	100
Min. Logic V_{dd} (V) (desktop)	**1.5 - 1.8**	**1.5 - 1.8**	**1.2 - 1.5**	**1.2 - 1.5**	**1.2 - 1.5**	**0.9 - 1.2**	**0.9 - 1.2**
Tox equivalent (nm)	**1.9–2.5**	**1.9–2.5**	**1.5-1.9**	**1.5-1.9**	**1.5-1.9**	**1.2-1.5**	**1.2-1.5**
Nominal I_{on} @25 °C (µA/µm) [NMOS/PMOS] High Perf.	**750/350**	**750/350**	**750/350**	**750/350**	**750/350**	**750/350**	**750/350**

		2008 70 nm	***2011 50 nm***	***2014 35 nm***
1	DRAM Half Pitch (nm)	70	50	35
2	MPU Gate Length (nm)	45	32	22
3	MPU / ASIC Half Pitch (nm)	80	55	40
4	ASIC Gate Length (nm)	70	50	35
5	Min. Logic V_{dd} (V) (desktop)	**0.6 - 0.9**	**0.5 - 0.6**	**0.3 - 0.6**
6	Tox equivalent (nm)	**0.8-1.2**	**0.6-0.8**	**0.5-0.6**
7	Nominal I_{on} @25 °C (µA/µm) [NMOS/PMOS] High Perf.	**750/350**	**750/350**	**750/350**

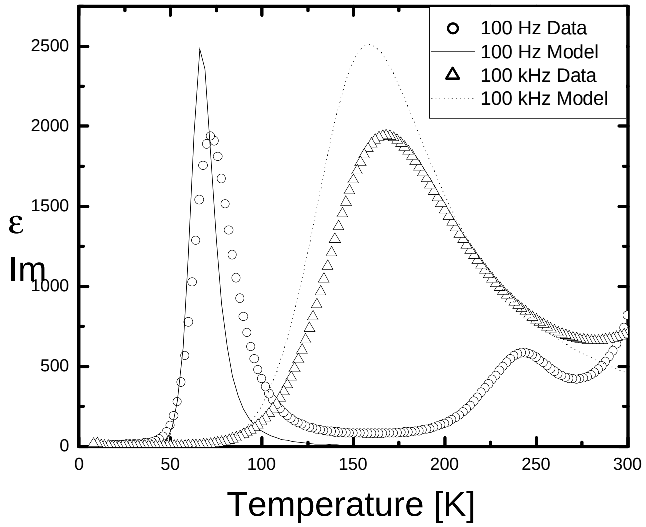
Polycrystalline sample - Schottky barrier removed





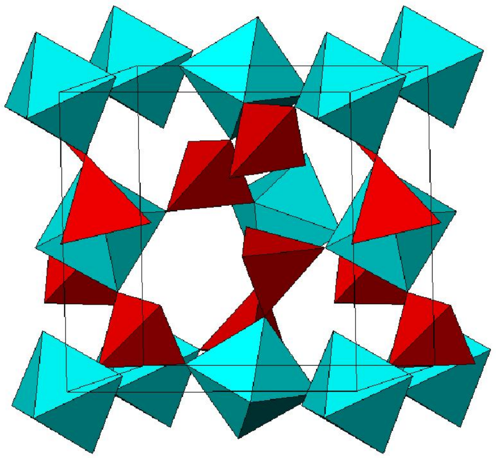


$CaCu_3Ti_4O_{12}$
100 Hz
Data
Model
10 kHz
γ_0=8x10[8] Hz
E_a/k_B=800 K
a=0.9805
1 MHz
ε Re
Temperature (K)

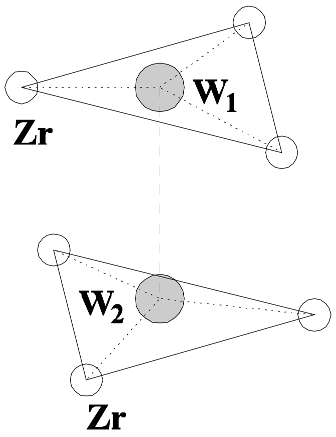


100 Hz Data
100 Hz Model
100 kHz Data
100 kHz Model
ε Im
Temperature [K]

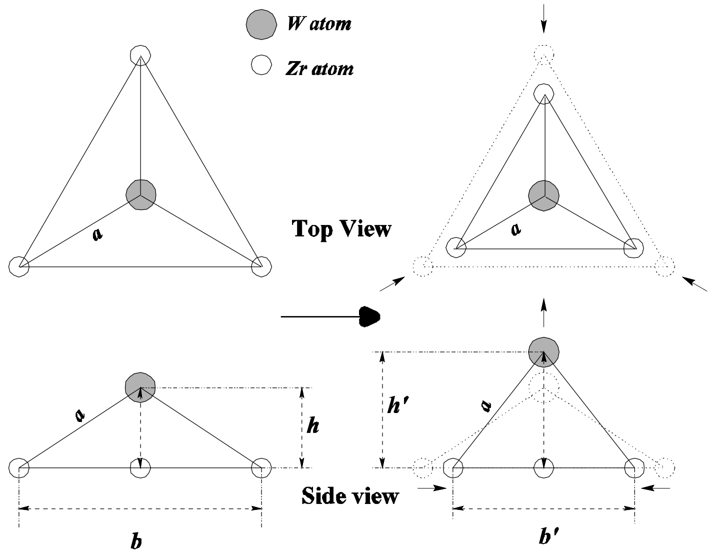
The Umbrella Model for W-Zr Motion



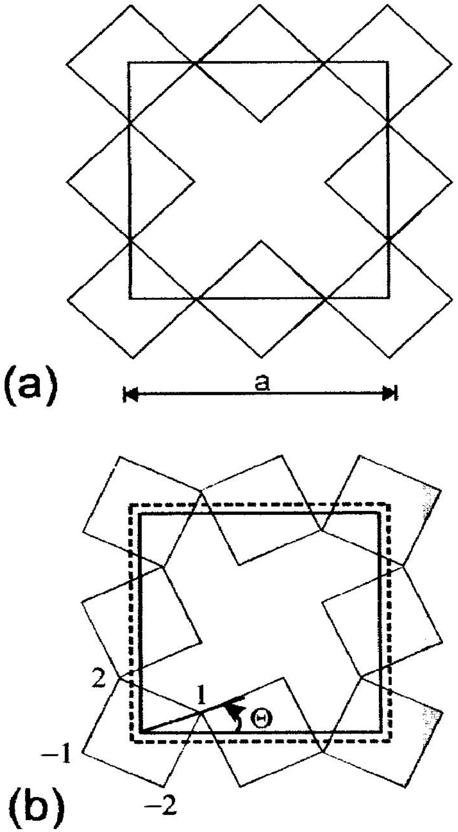
A:



B:



Possible relevance of a new field theory of lattice distortions



$$\nabla\cdot\mathbf{u} = 2(\cos\theta-1)-3b^2\nabla\cdot\nabla(\cos\theta) = J_0(\theta)$$

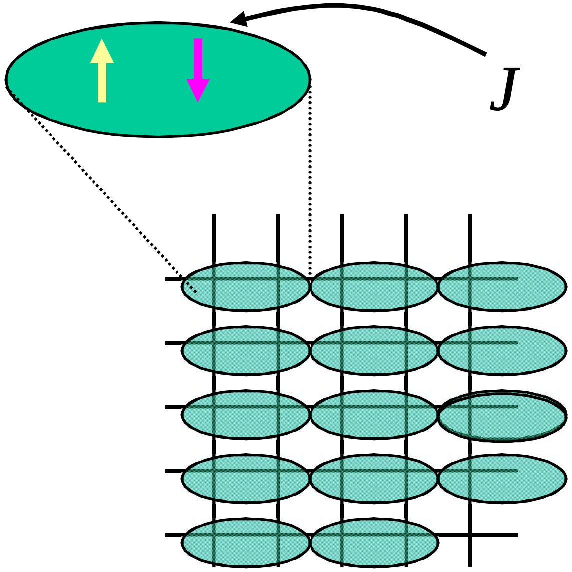
$$\nabla\times\mathbf{u} = 2\sin\theta + 3b^2\nabla\cdot\nabla(\sin\theta) = J_1(\theta)$$

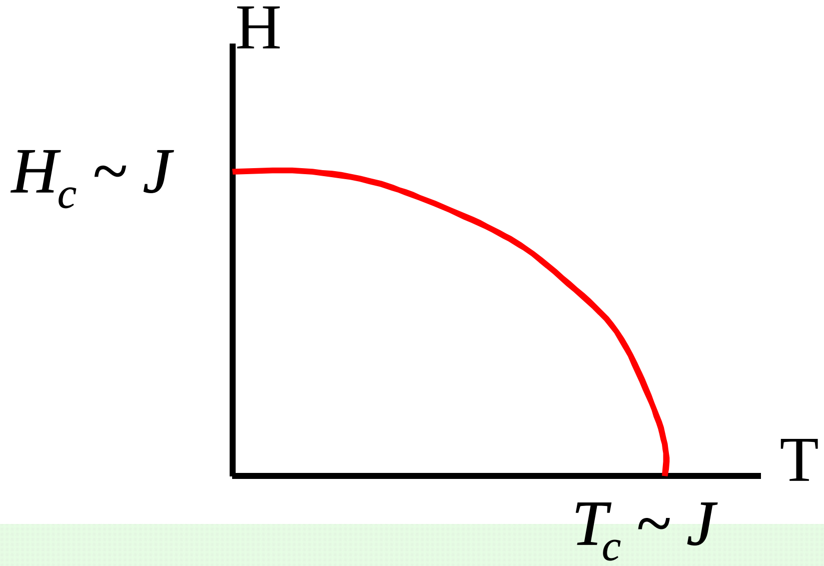
⇓

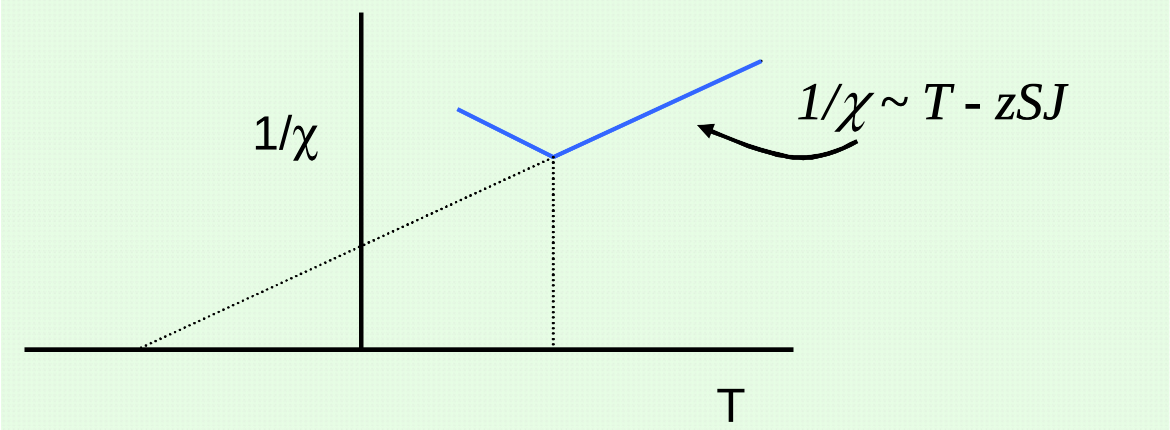
Zero shear velocity at T=0

M. E Simon and C. Varma, Phys. Rev. Lett. 2001

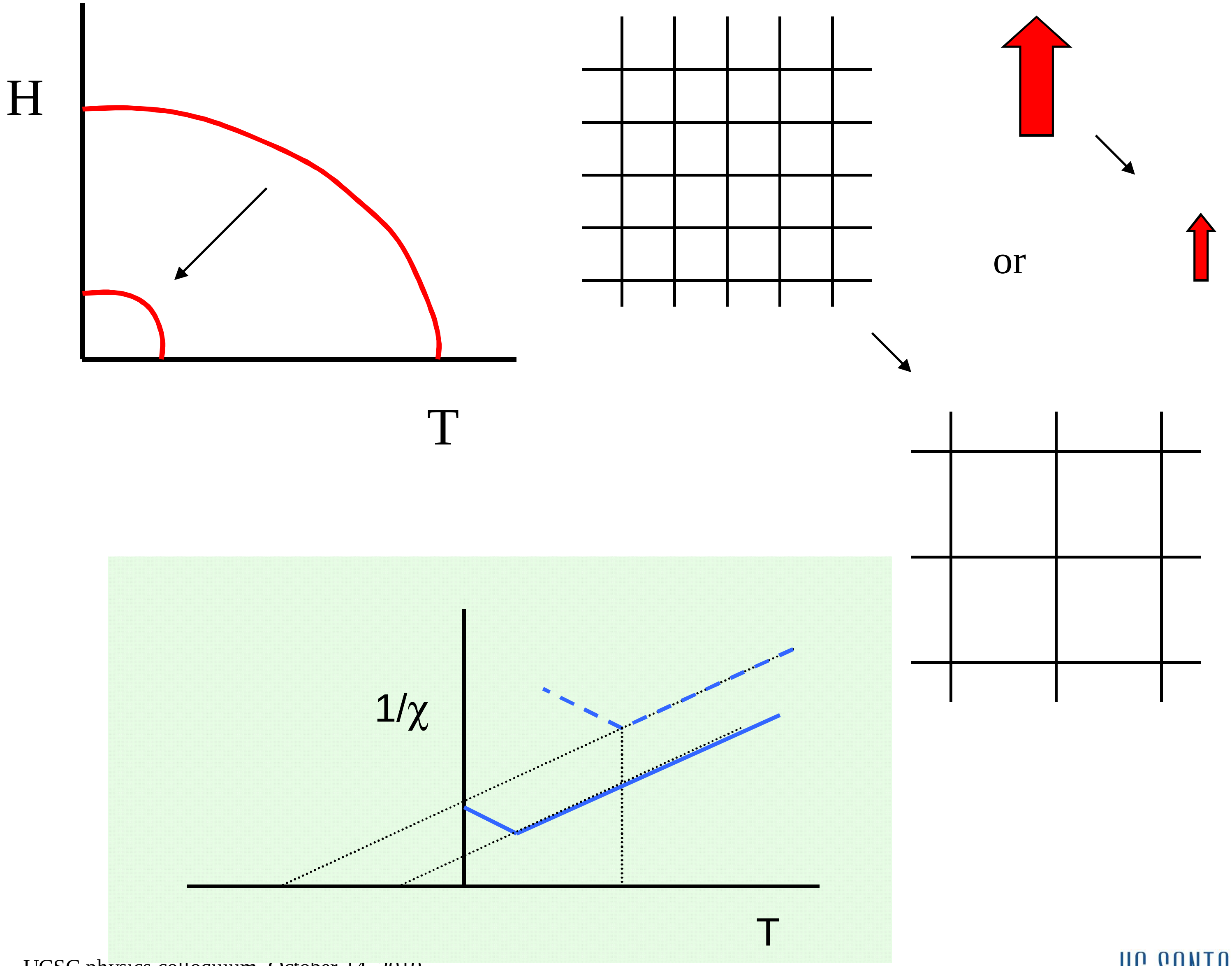
Mean Field Theory in Magnetism



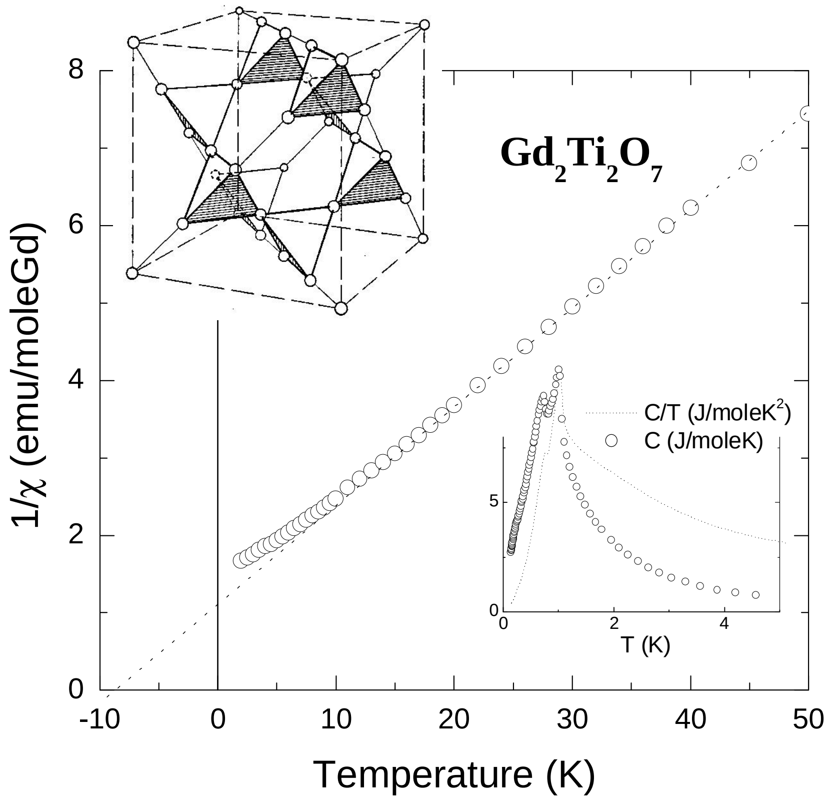


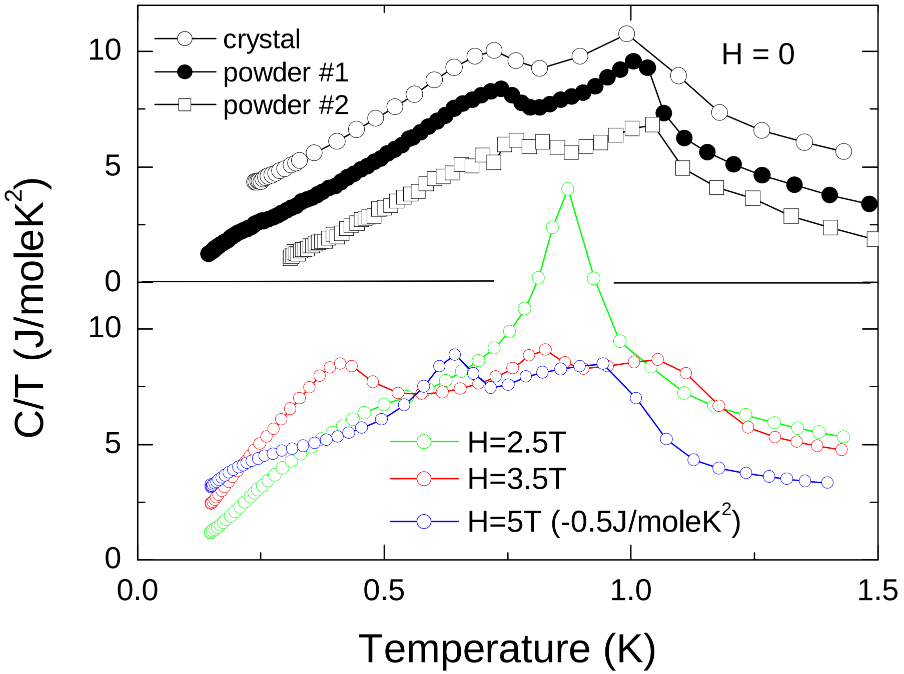


How can T_c be reduced?



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

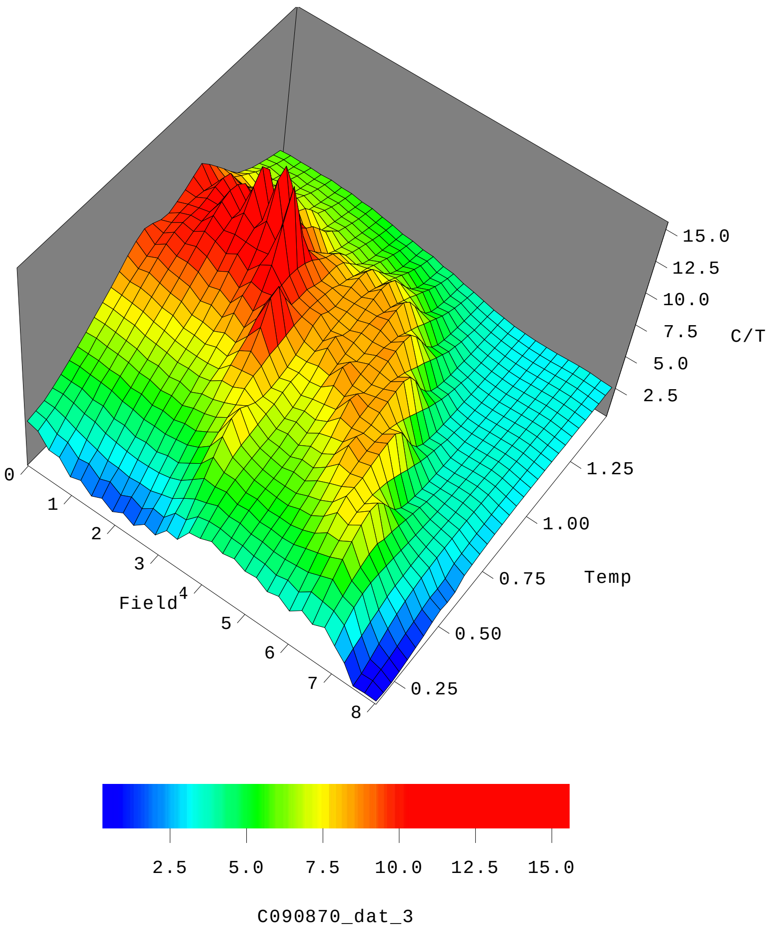




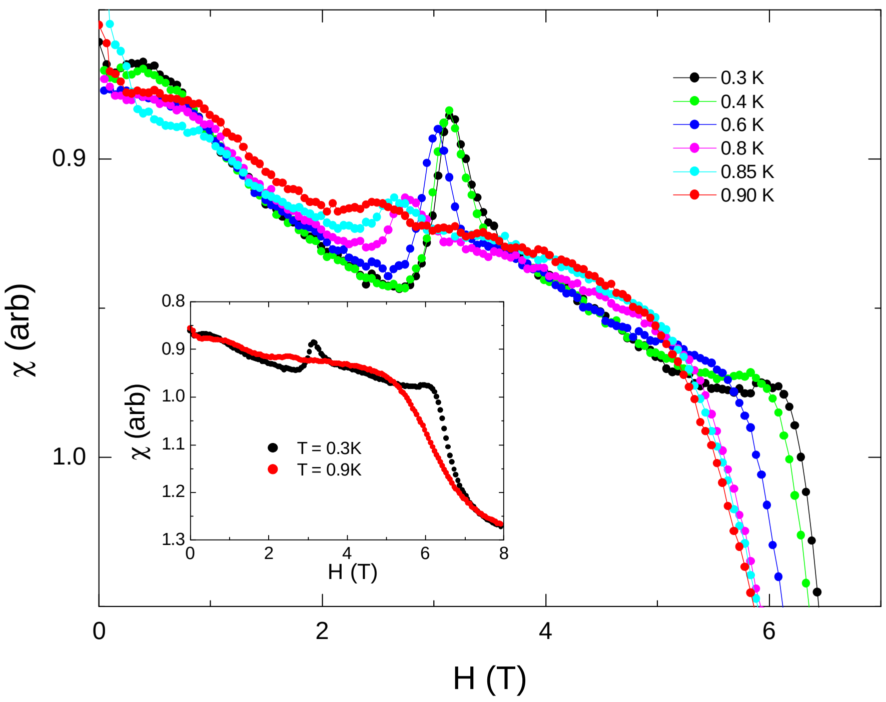
PRL, 2002

$Gd_2Ti_2O_7$

Specific Heat

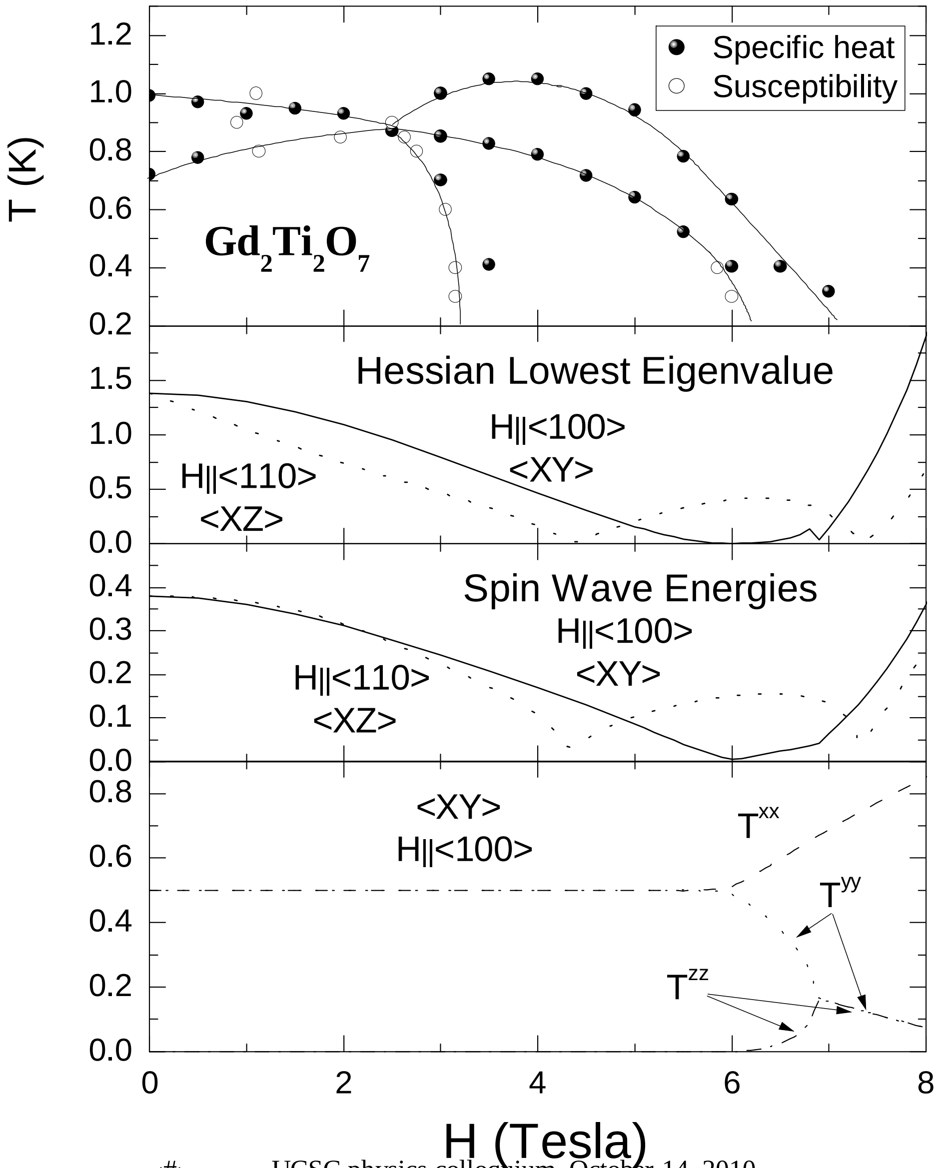


Magnetic Susceptibility



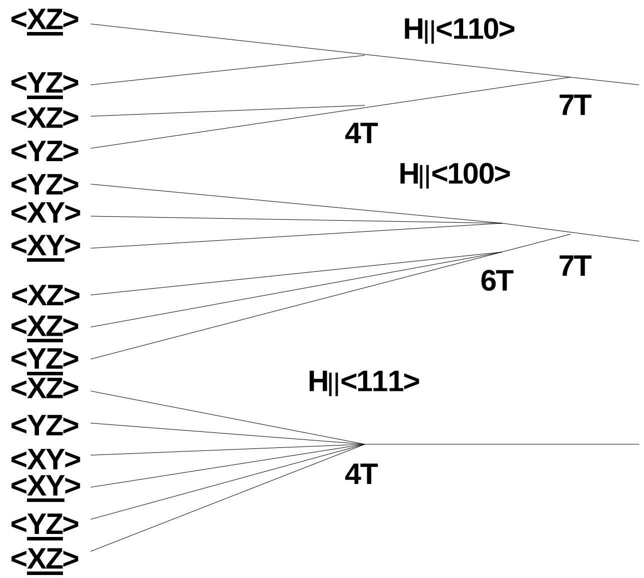
PRL, 2002

Phase Diagram – $Gd_2Ti_2O_7$

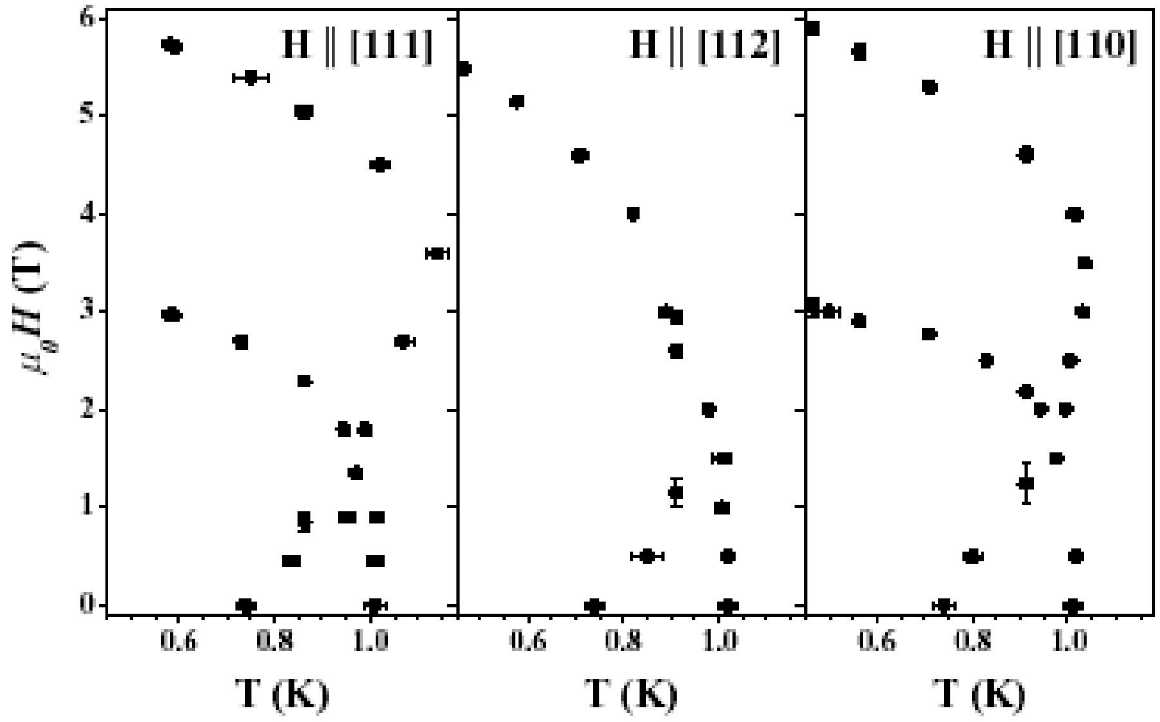


$$4F/N = +JS^2 \sum_{j,k} \vec{\eta}(j)\cdot\vec{\eta}(k) -$$

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



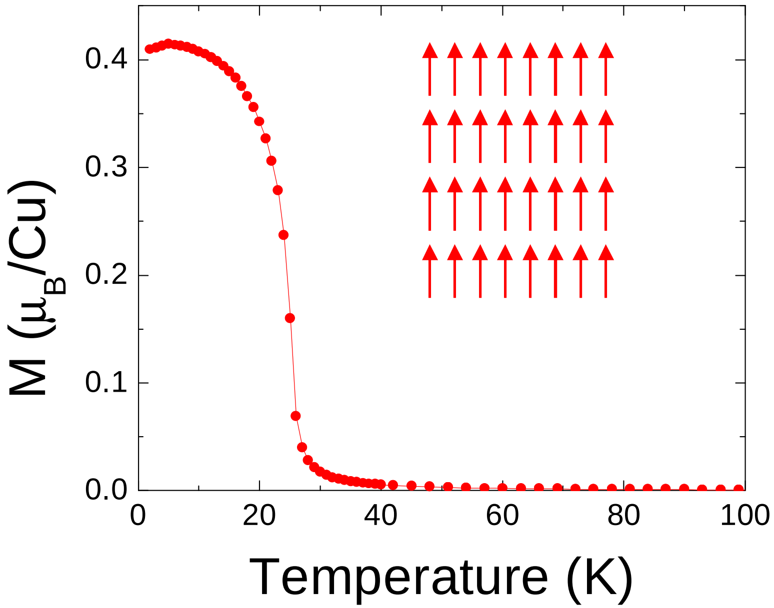
PRL, 2002



O. Petrenko et al, 2004

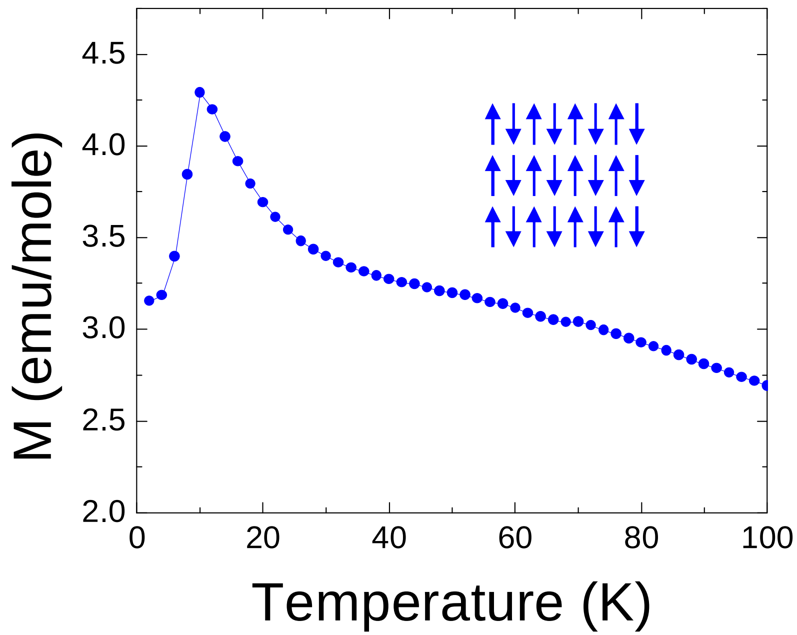
Magnetization

$SeCuO_3$



T_c=25K

$TeCuO_3$



T_N=9K

Functional Form for Coupling Strength

$$F_{MDE} = \alpha P^2 \sum_q g(q) \left\langle M_q M_{-q} \right\rangle$$

$$H_{ex} = \sum_{i<j} J_{ij} M_i \cdot M_j$$

$$\approx \sum \left[J_{ij}(R_{ij}^0) + \vec{J}' \cdot (u_i - u_j) + J''(u_i - u_j)^2 + \dots \right] (M_i \cdot M_j)$$

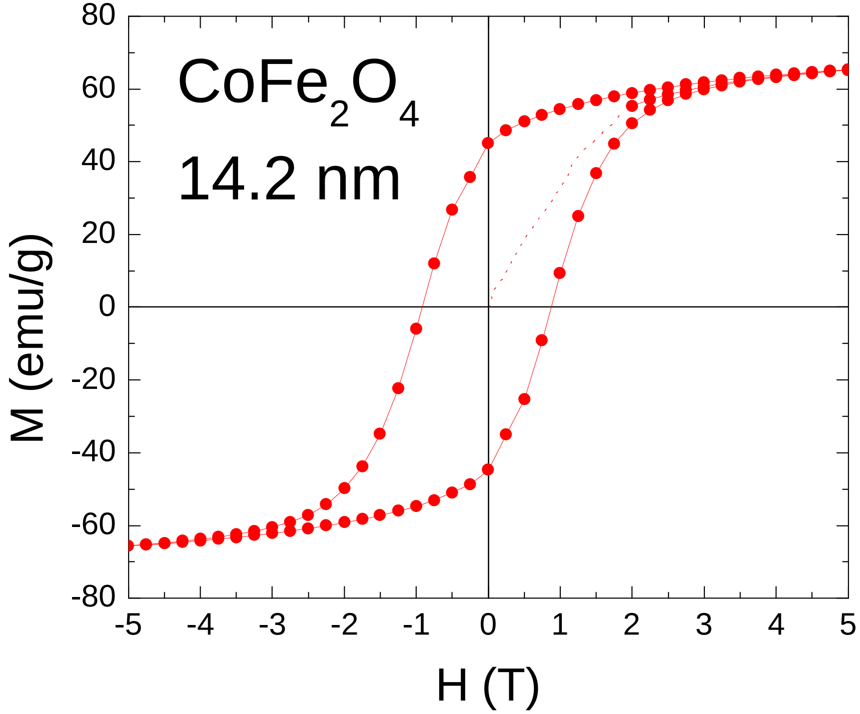
$$H_{phonon-M} = J'' u_0^2 \sum_q (1 - \cos q \cdot R_{ij}^0) \left\langle M_q M_{-q} \right\rangle$$

$$\mathrm{g}(q) \propto \left(1 - \cos qR\right)$$

Ferromagnetism

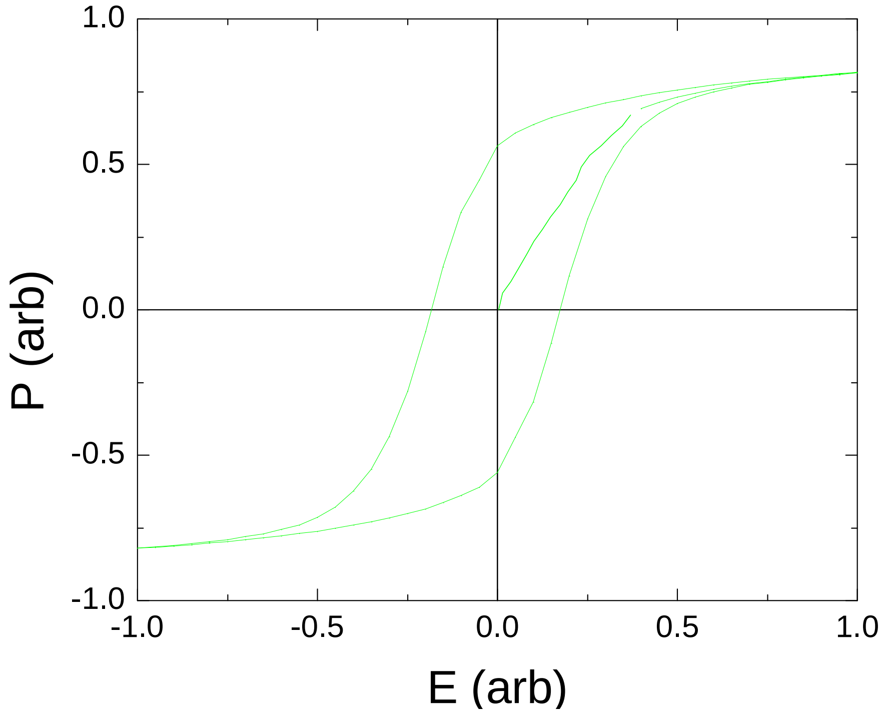
- Spin ordering transition
- T_c < 100K

- Below T_c there is a spontaneous magnetization
- Hysteresis loop in M vs H (arising from domains)



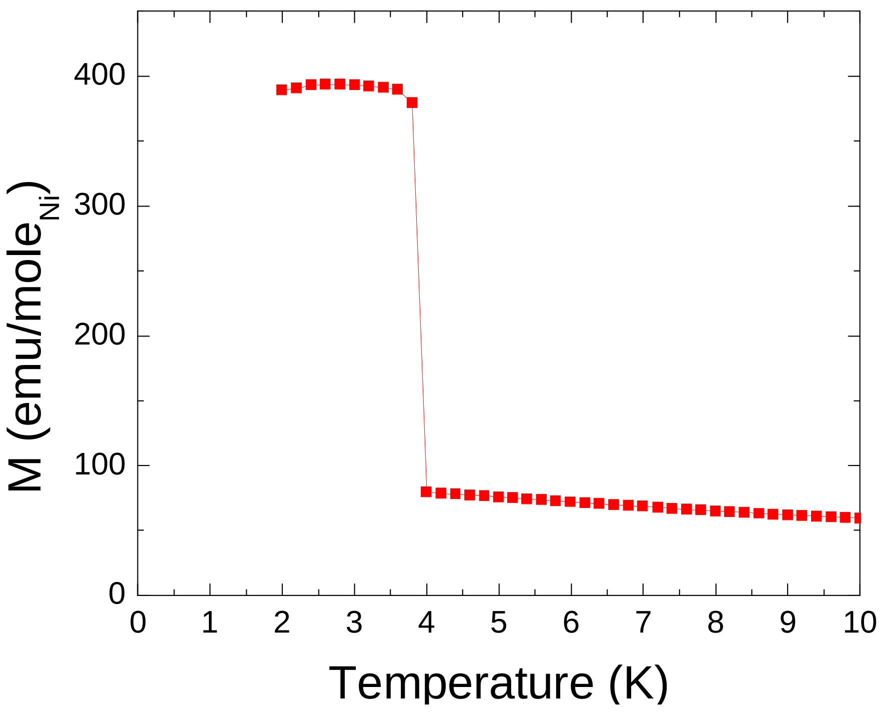
Ferroelectricity

- Electric dipole ordering transition
- T_c typically > 300K

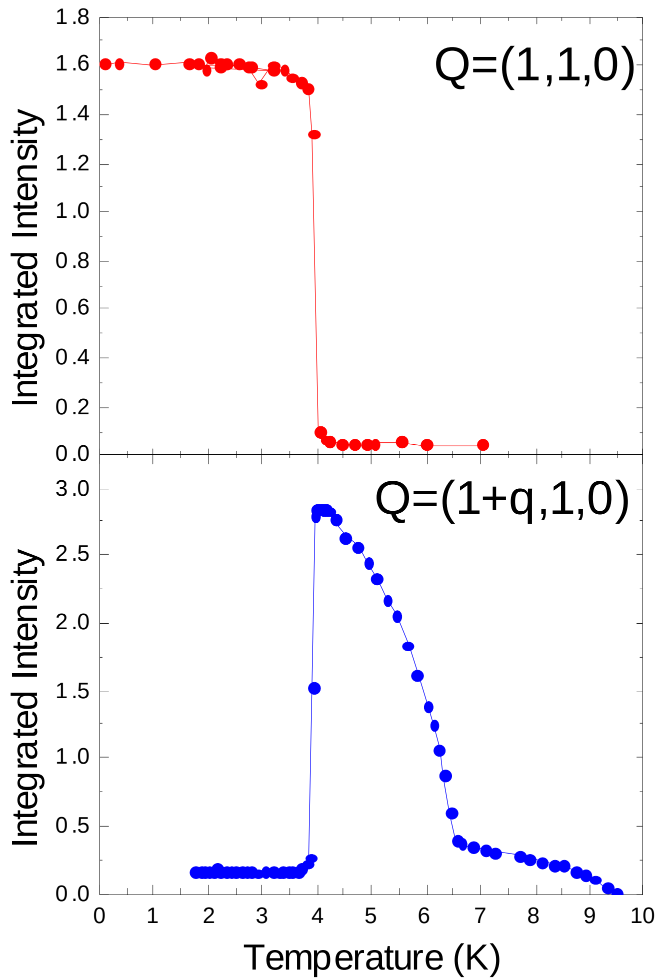


- Below T_c there is a spontaneous polarization
- Hysteresis loop in P vs E (arising from domains)

Magnetic Properties of $Ni_3V_2O_8$

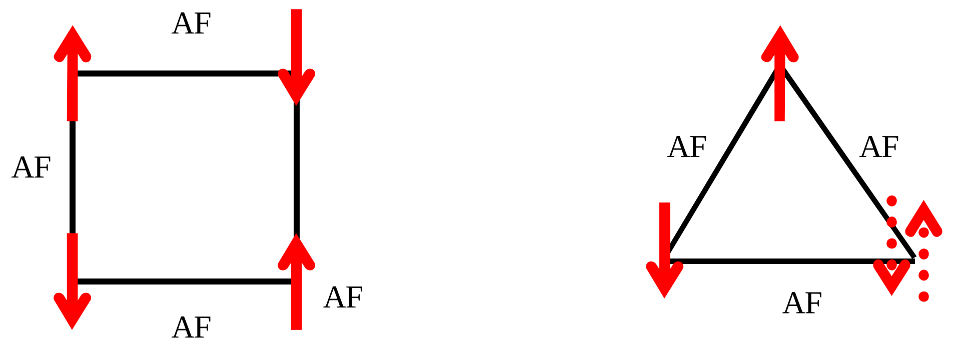


•Incommensurate magnetic order between 6.5K and 4K
•Canted antiferromagnetic order below 4K



Neutron data from M. Kenzelmann and C. Broholm

Unfrustrated and Frustrated Spins



Local interaction

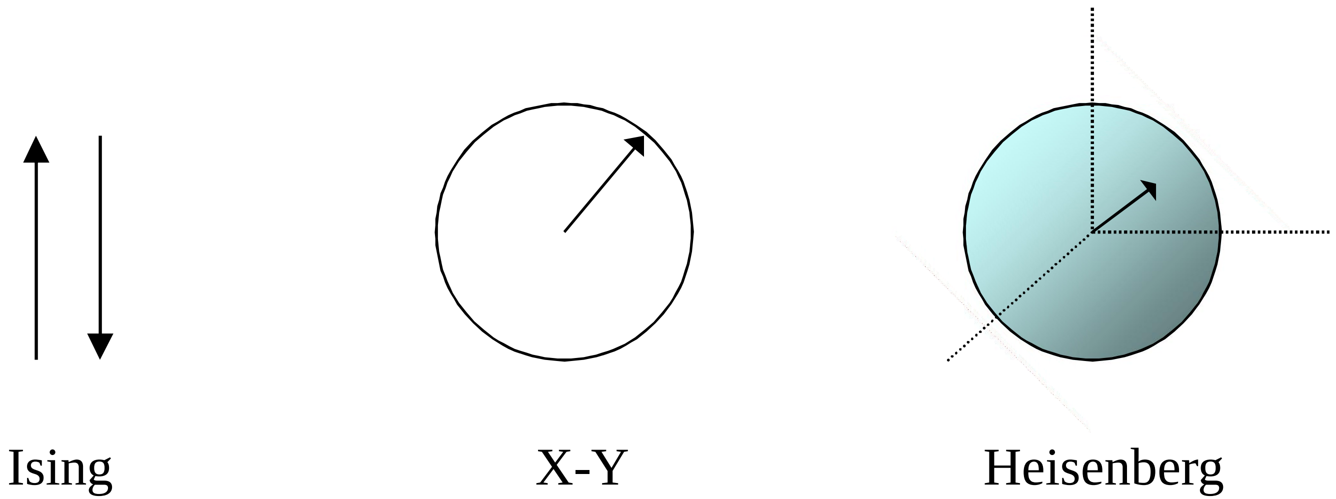
⇓

Order Parameter symmetry

⇓

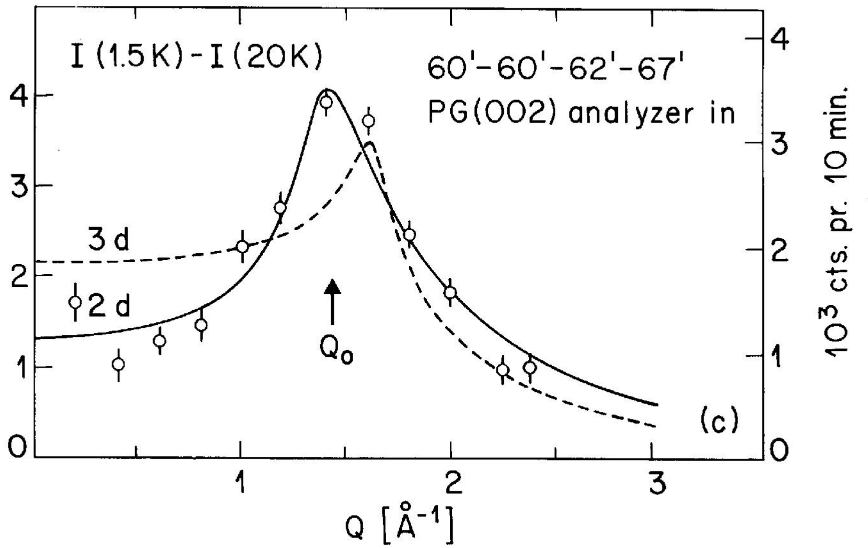
Symmetry incompatibility – OP ⇔ Space group

Different spin types have different GF conditions



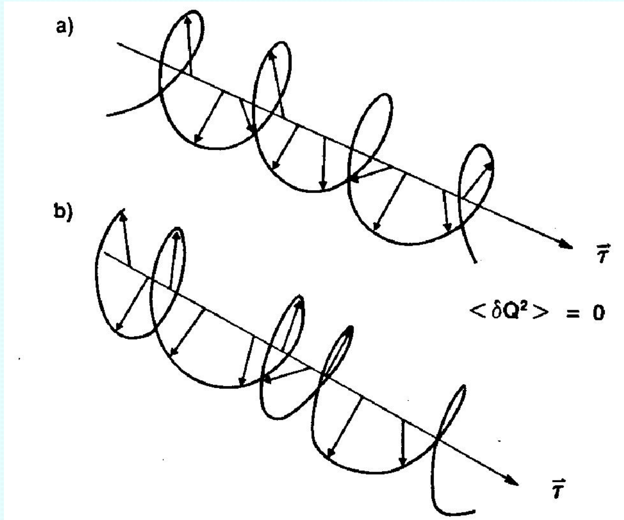
Moessner & Chalker

Liquid-like structure factor in a kagome system $SrCr_8Ga_4O_{19}$



Broholm, Aeppli et al., PRL, **65**, 3173 (1990)

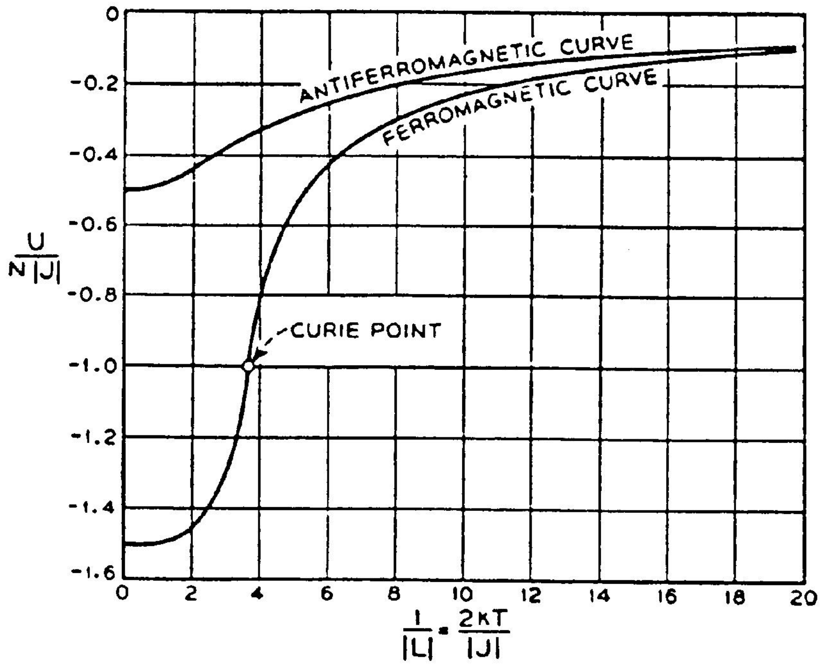
Spin Nematic

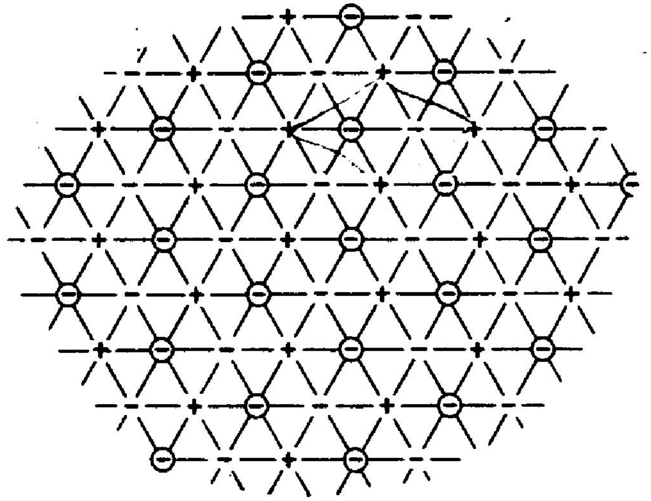


Chandra & Coleman, PRL, **66**, 100 (1991)

UC SANTA CRUZ

Geometrical Frustration for 2D Ising spins





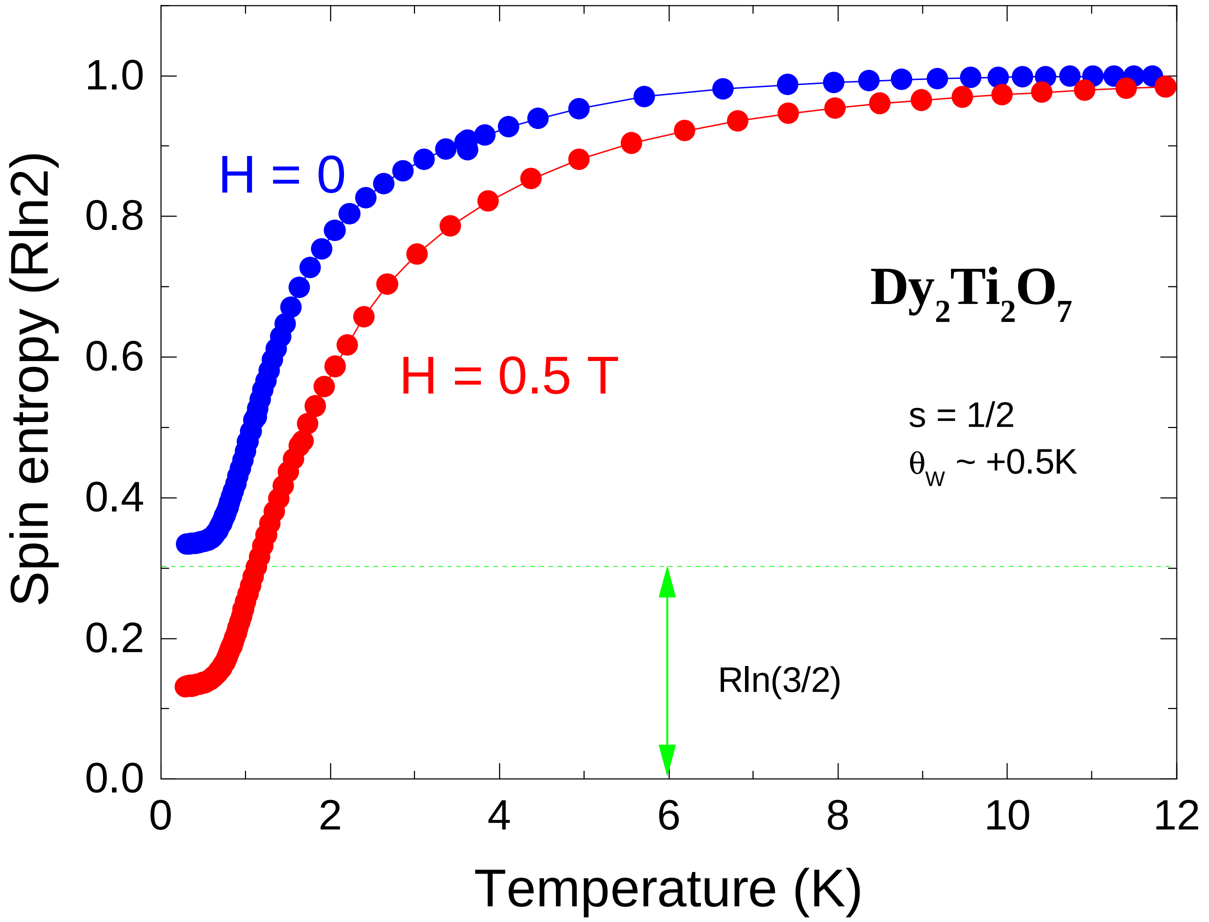
For the Ising triang. AF,

$S_0 = 0.323R = 0.47S_\infty$

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

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

Effect of magnetic field on zero-point disorder



MultiFerroics – The Ultimate Magnetoelectric Materials

- Ferroelectrics are usfeul – sensors, actuators, memory
- Ferromagnets are useful – same as above
- Materials with both should be useful – “Multiferroics”
- Need the following:

  1) Materials with both at microscopic level

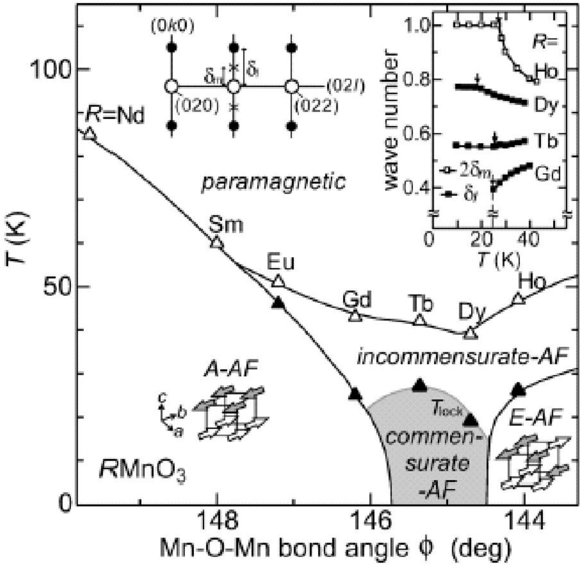
  2) Symmetry breaking of ionic positions (FE) as well as magnetic orientation

  3) Strong coupling between FE and FM(AF) – magnetoelastics

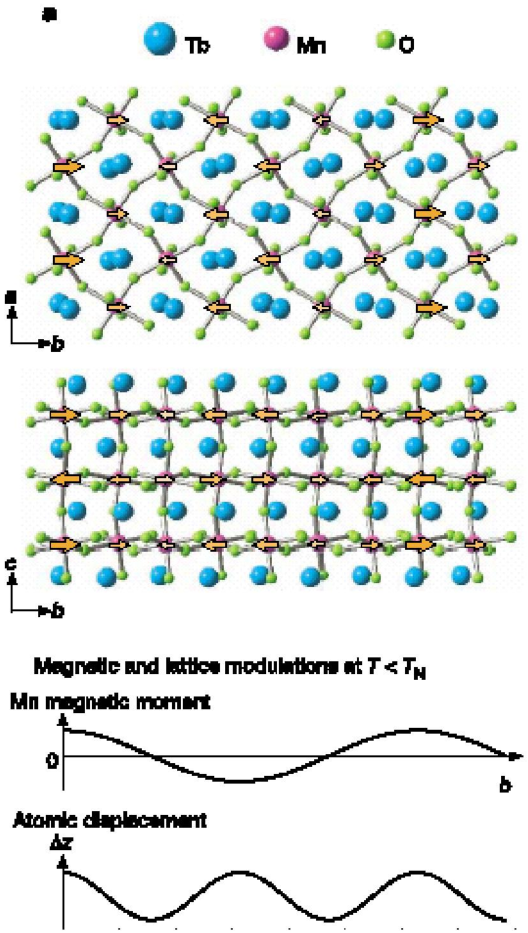
Observation of Ferroelectricity in the Magnet $TbMnO_3$

Magnetic control of ferroelectric polarization

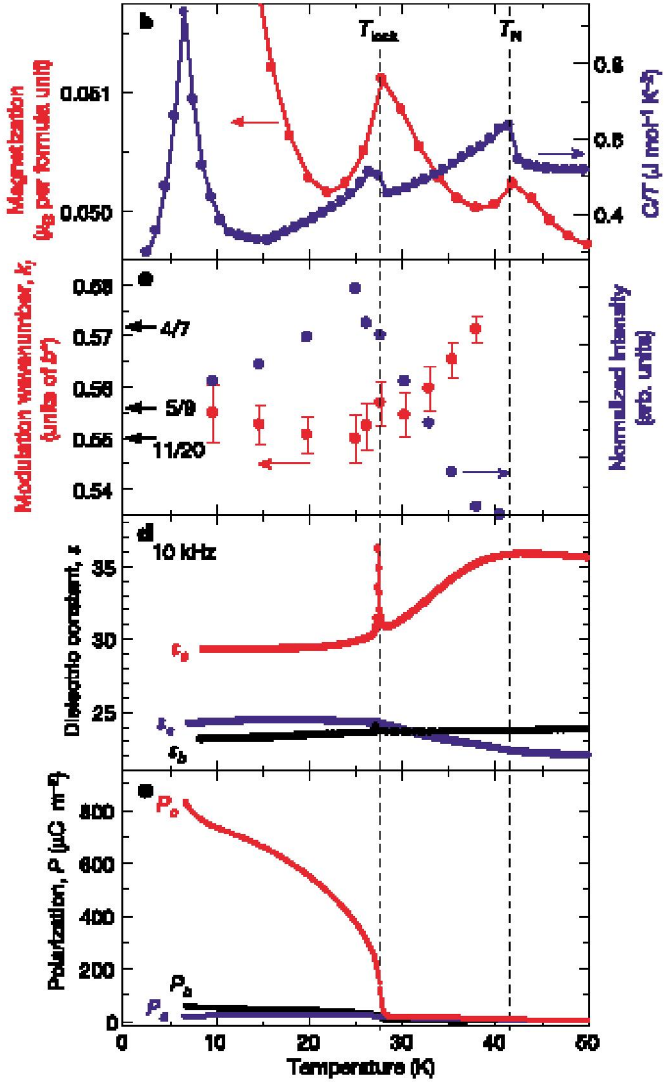
T. Kimura[1,*], T. Goto[1], H. Shintani[1], K. Ishizaka[1],



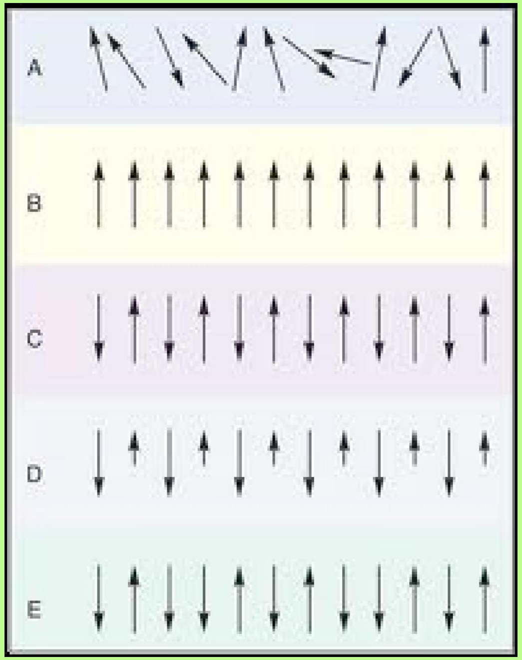
Goto, Kimura, Lawes, Tokura, APR, *PRL*, 2004



Kimura et al, *Nature* 2003



Antiferromagnetism



Paramagnetic

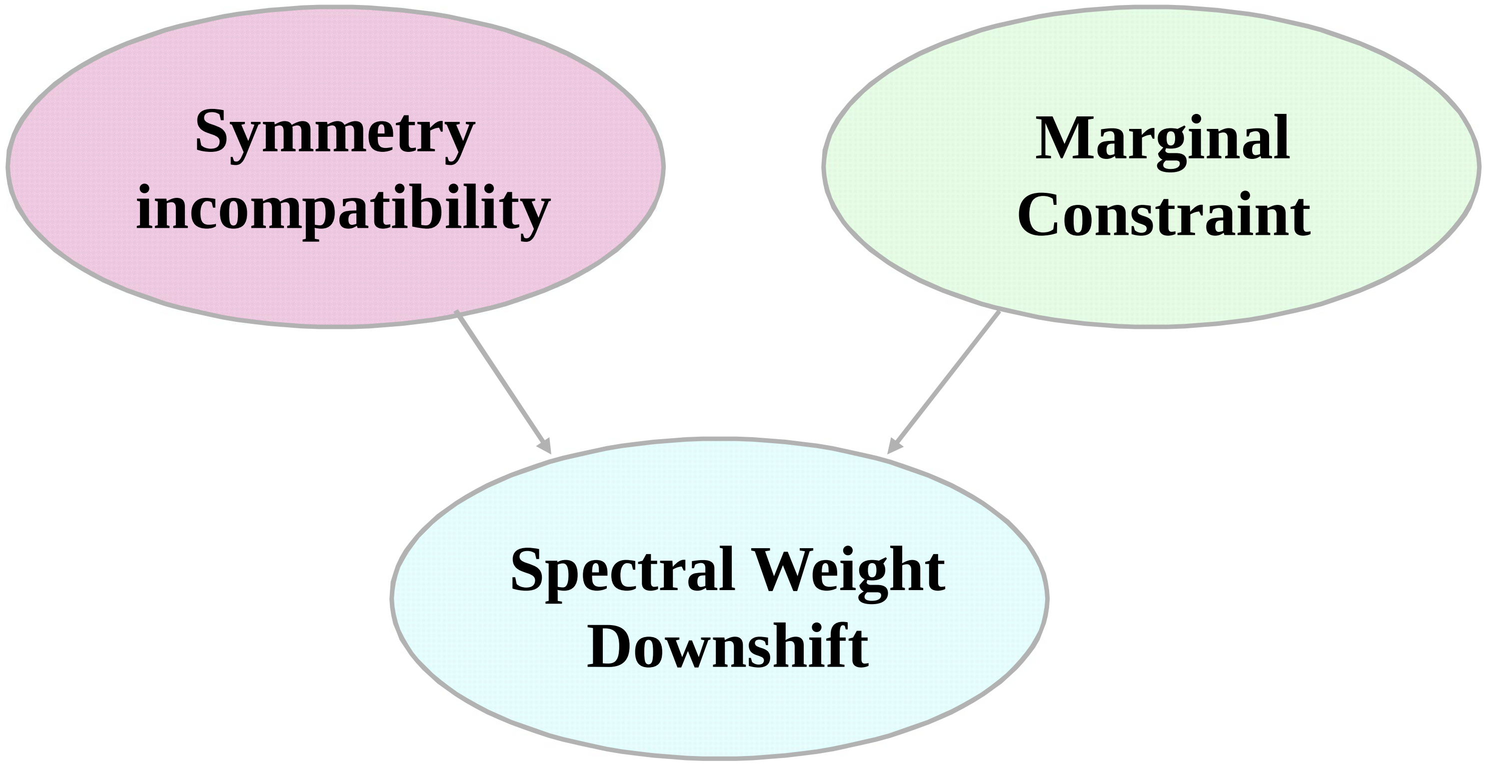
Ferromagnetic

Antiferromagnetic (Néel state)

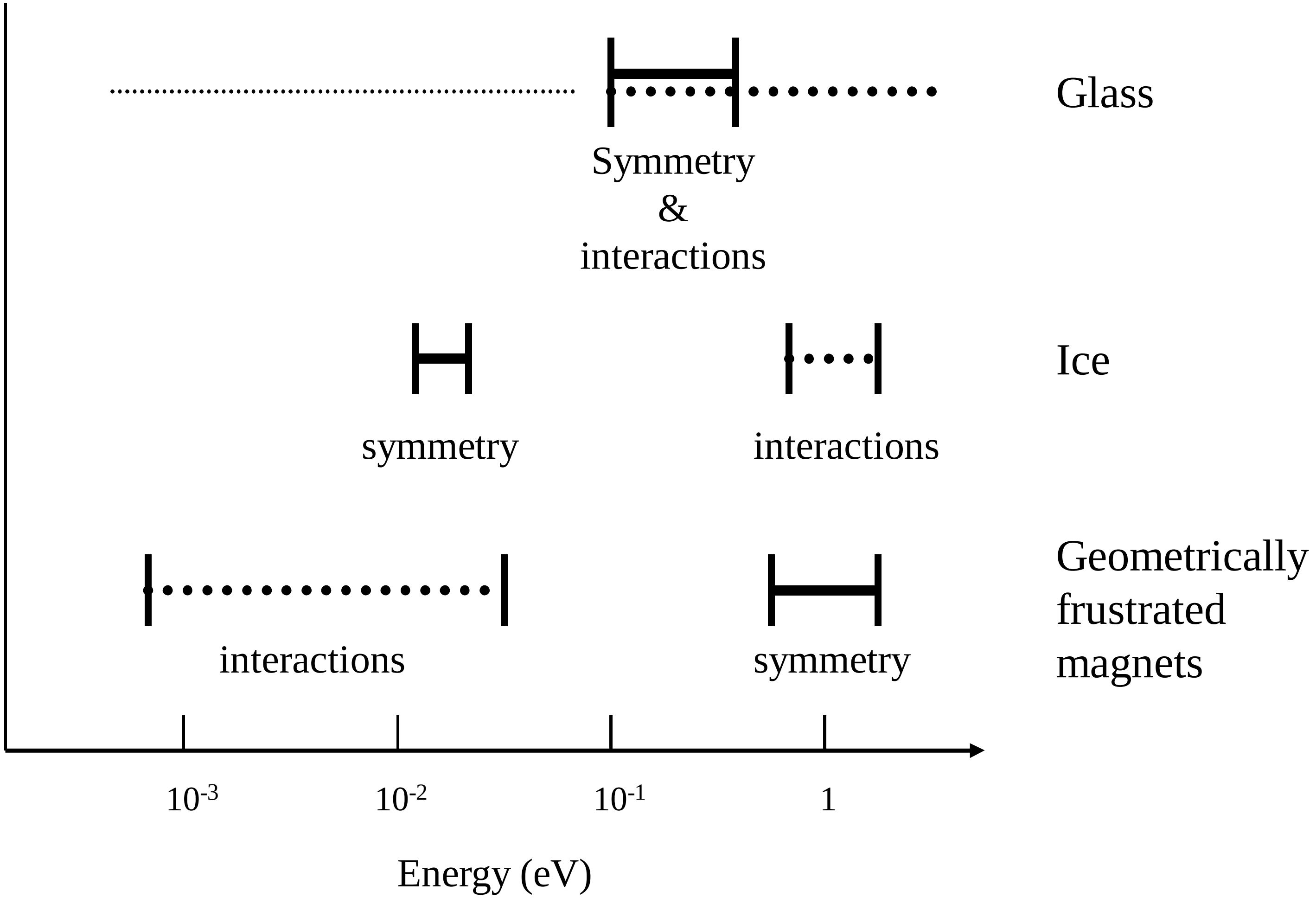
Ferrimagnetic

Incommensurate

Conventional magnetic ground states


Symmetry incompatibility
Marginal Constraint
Spectral Weight Downshift

Glass/Spin-Glass…Ice/Spin-Ice



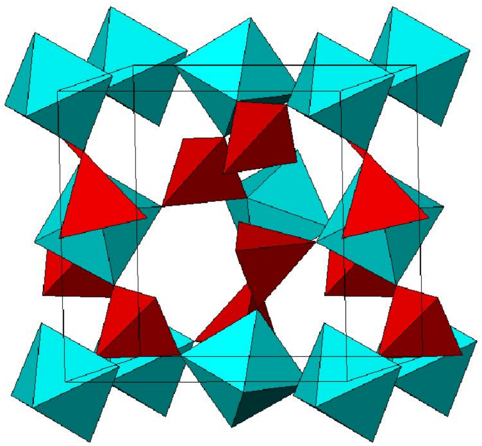
So, we see a parallel set of G-F non-magnetic materials

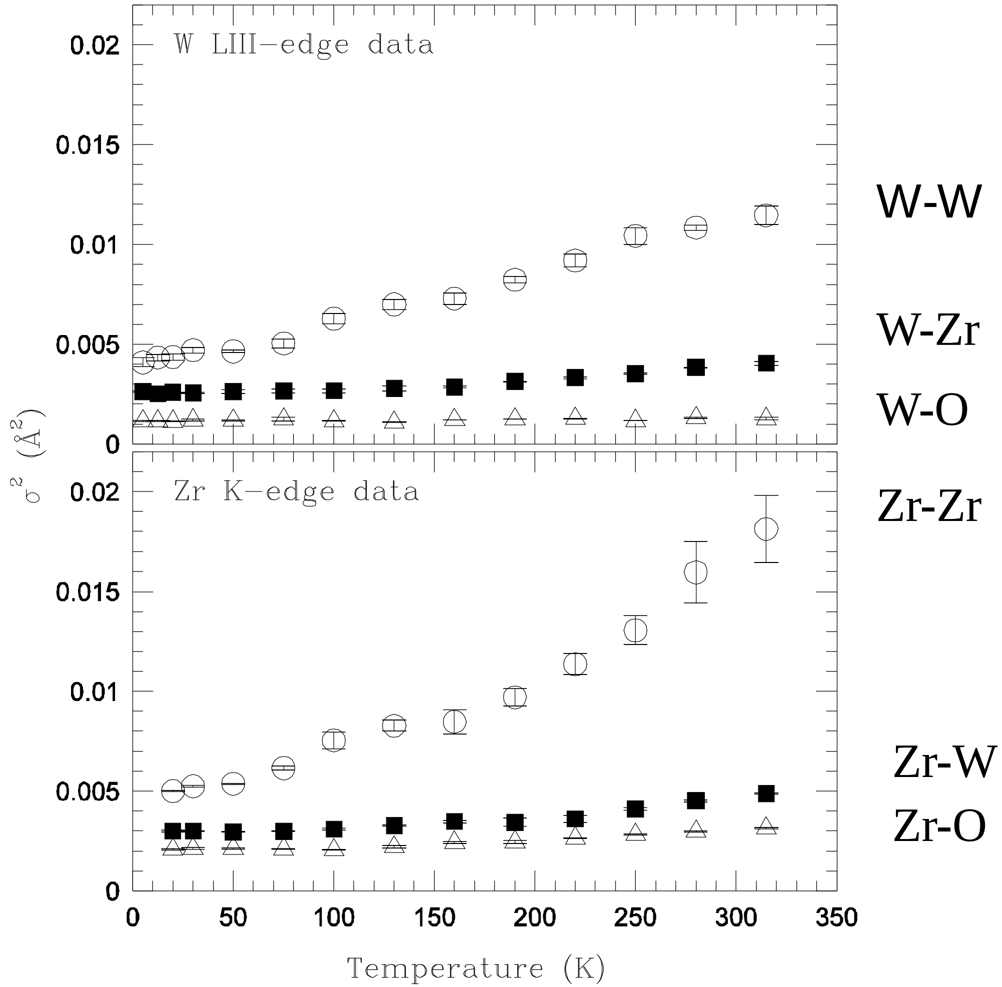
Frustration →

Disorder ↓

Ferromagnets Antiferromagnets	Geometrical frustration
Bravais lattice	Negative thermal expansion
Random fields percolation	Spin glass
Rigidity percolation	Relaxor ferroelectric

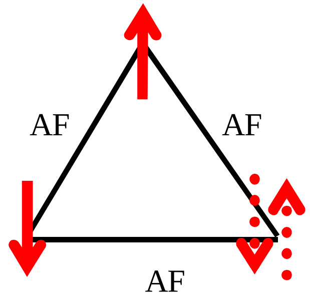
- **Spectral weight in ZrW_2O_8 - What's the origin?**
- **Look at the local bond lengths with XAFS**





D. Cao, F. Bridges, G. Kowach, APR, PRL 2002

Geometrical Frustration – Materials Considerations

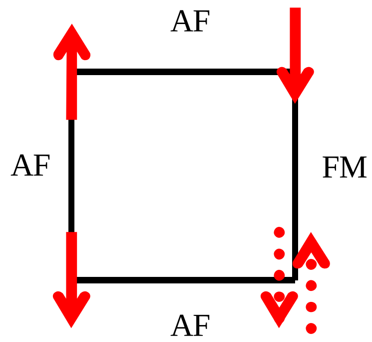


Frustration →

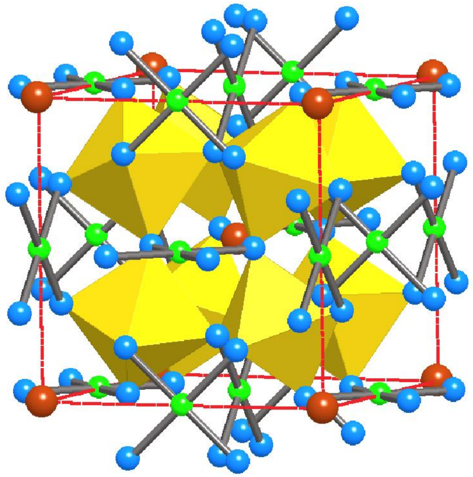
Disorder ↓

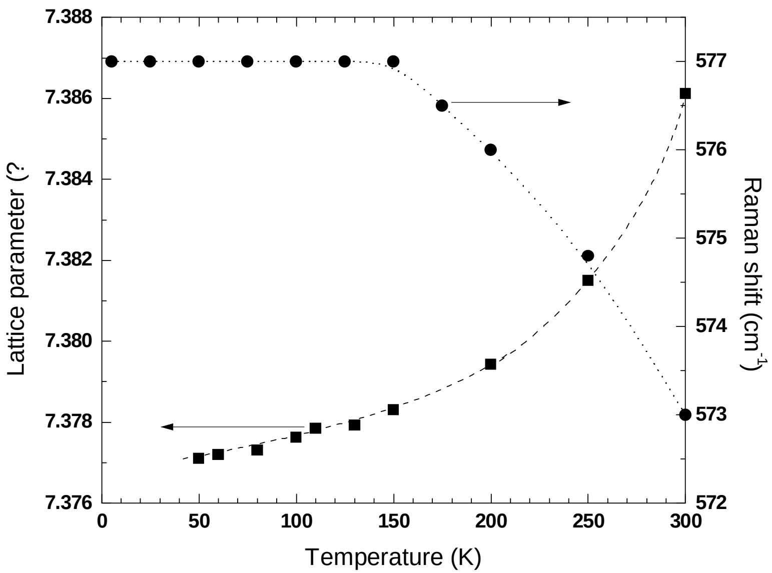
	Antiferromagnetism Ferromagnetism Metamagnetism Ferrimagnetism	Geometrical Frustration
	percolation, random fields	Spin glass

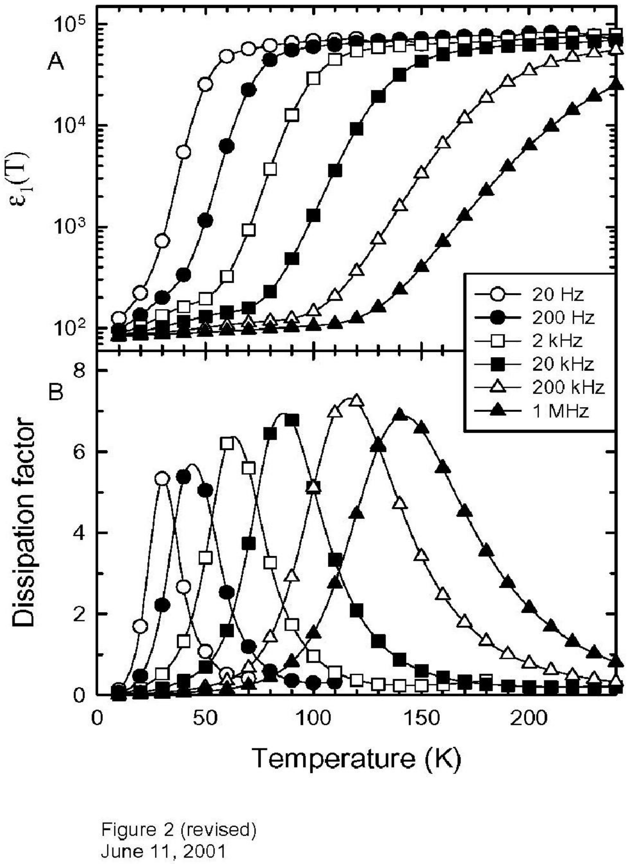
$$H=\Sigma S_i \cdot S_j$$



Possible example of a GF ferroelectric - $CaCu_3Ti_4O_{12}$

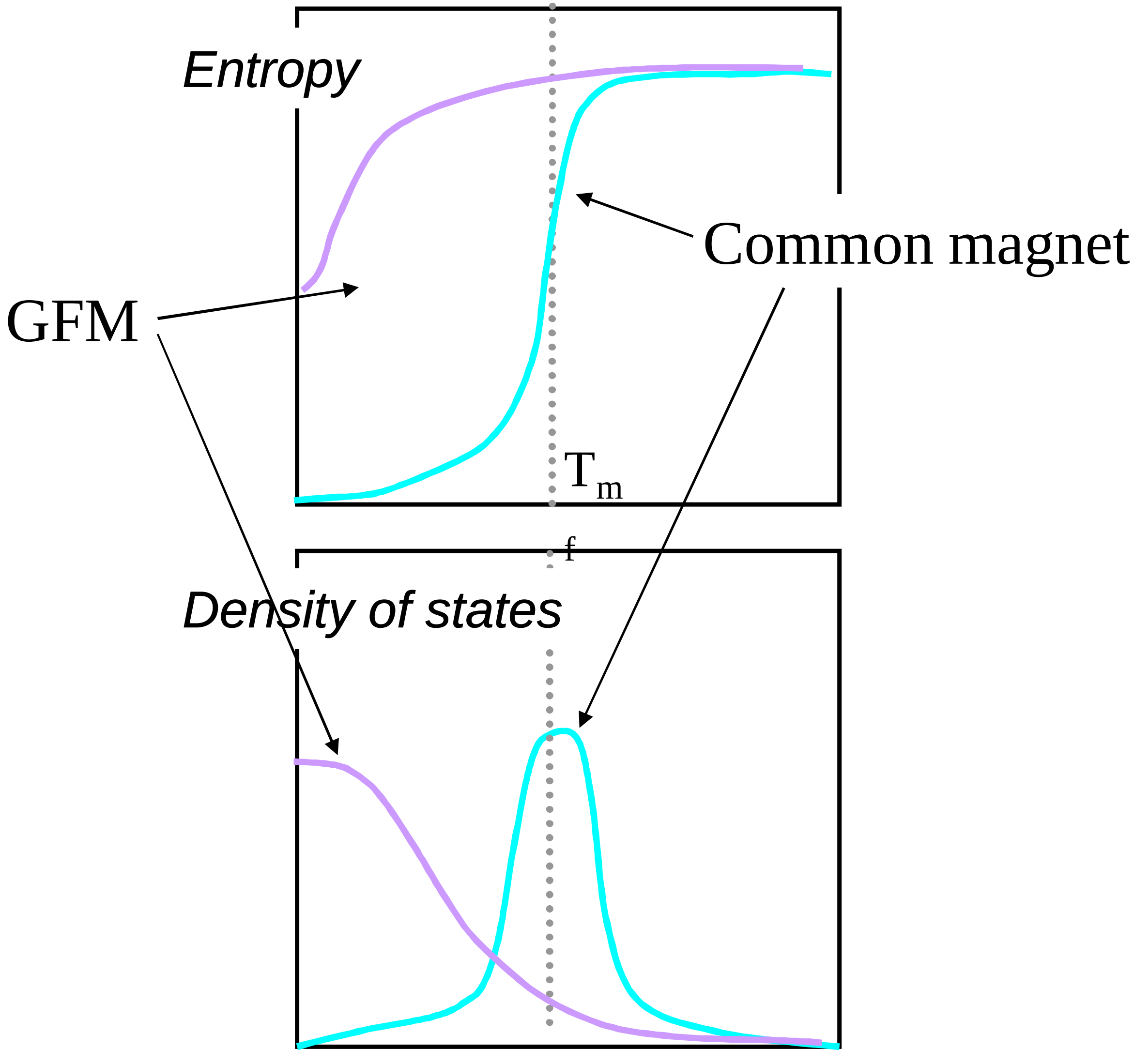




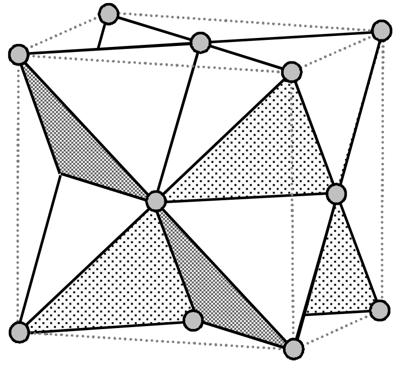


APR et al, SSC (200), Homes et al. Science (2001)

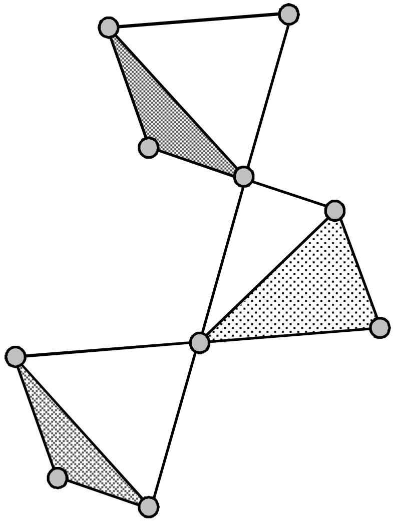
More on *Spectral Weight Downshift*



3 Dimensions



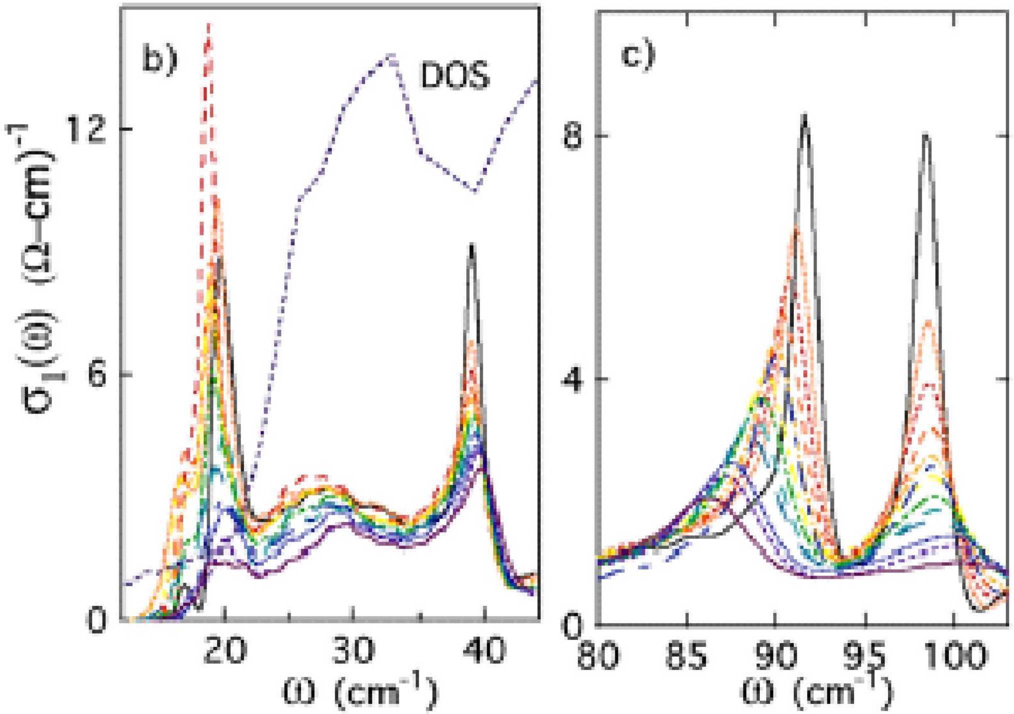
FCC

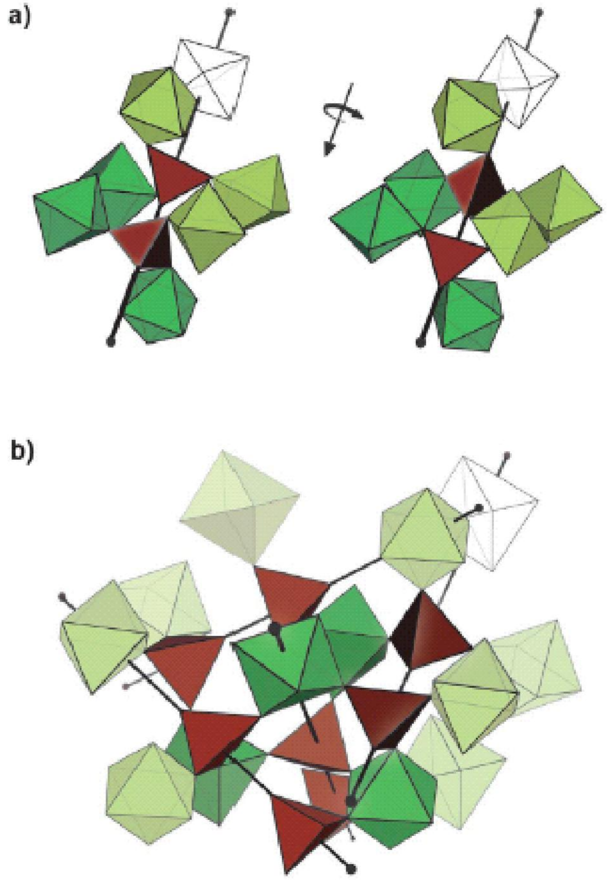


Pyrochlore

Let's look at a simple Ising-type system where the degeneracy is countable

IR Conductivity





Hancock, Turpen, Schlesinger, Kowach, Ramirez, PRL, 2004

Hoberman sphere – a “negative Poisson ratio” material