Half-magnetization plateaux in Cr Spinels

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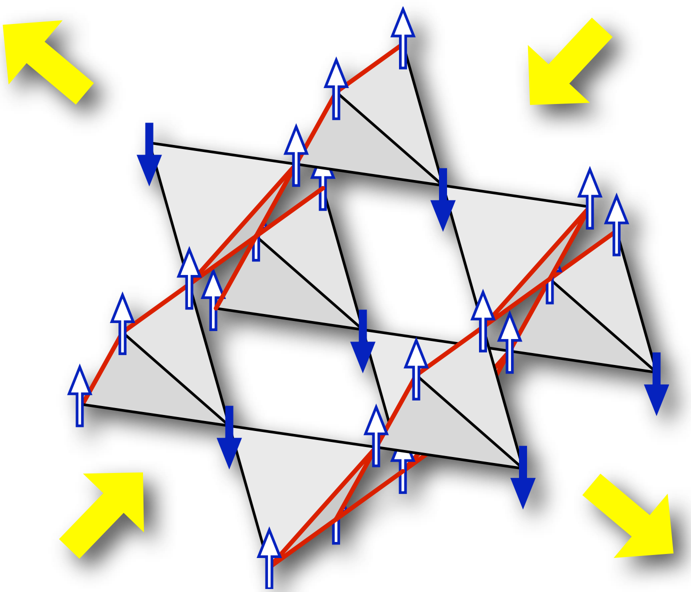

Moments and Multiplets
in Mott Materials

October 11, 2007
KITP, Santa Barbara

In collaboration with :

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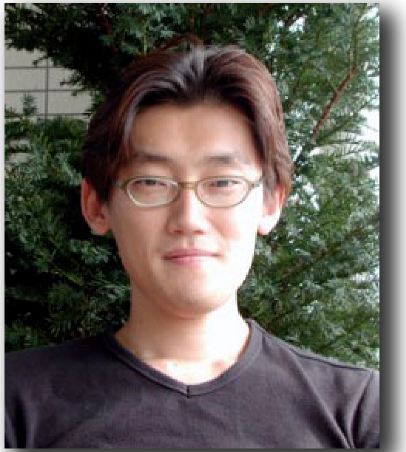

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(Riken, Tokyo)

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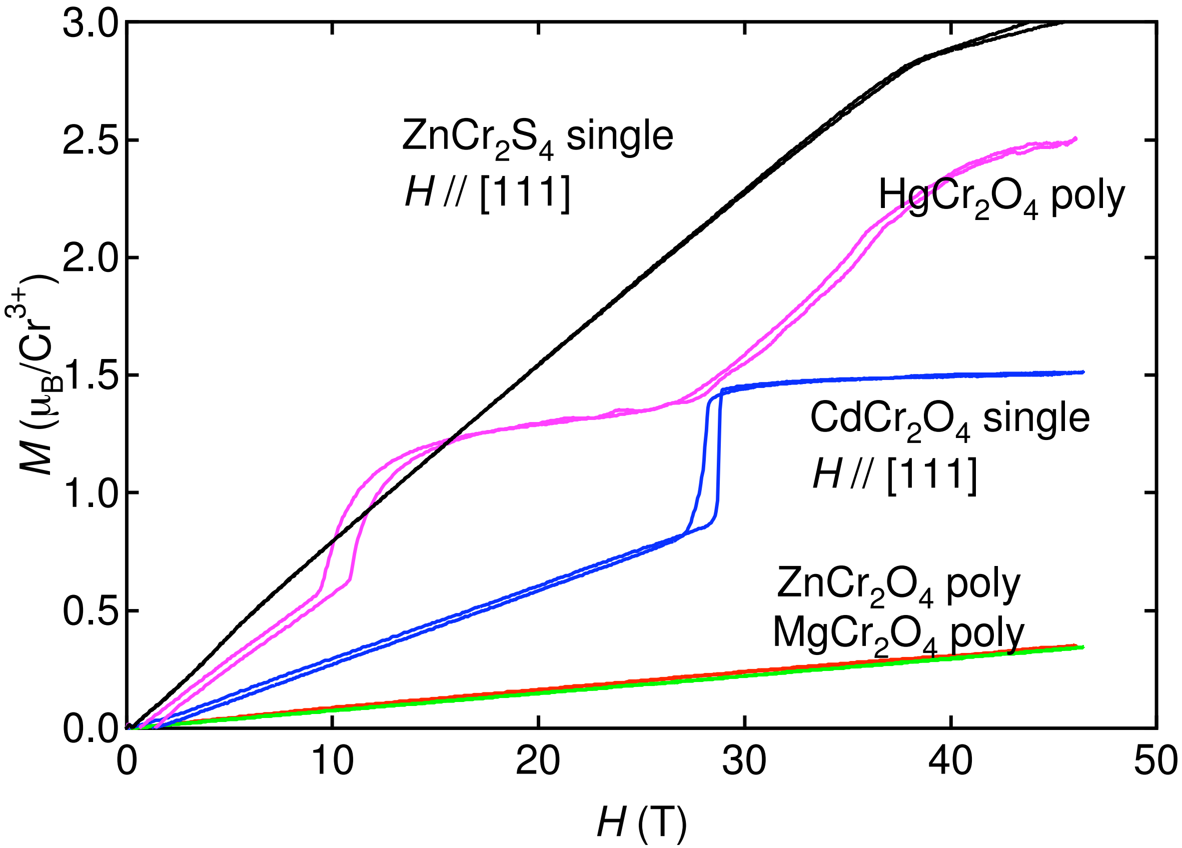
- Riken/CREST

Guide map of "simple" spinel oxides (after Takagi)

charge frustration	$d^{0.5}$ $LiTi_2O_4$ BCS SC	$d^{1.5}$ LiV_2O_4 heavy fermion	$d^{2.5}$ AlV_2O_4 charge ordered insulator	$d^{3.5}$ $LiMn_2O_4$
spin frustration (insulators)	d^1 $MgTi_2O_4$ valence bond crystal	d^2 ZnV_2O_4 MgV_2O_4 CdV_2O_4 spin+orbital ordering	d^3 $ZnCr_2O_4$ $MgCr_2O_4$ $CdCr_2O_4$ spin driven structural phase transition	d^4 $ZnMg_2O_4$

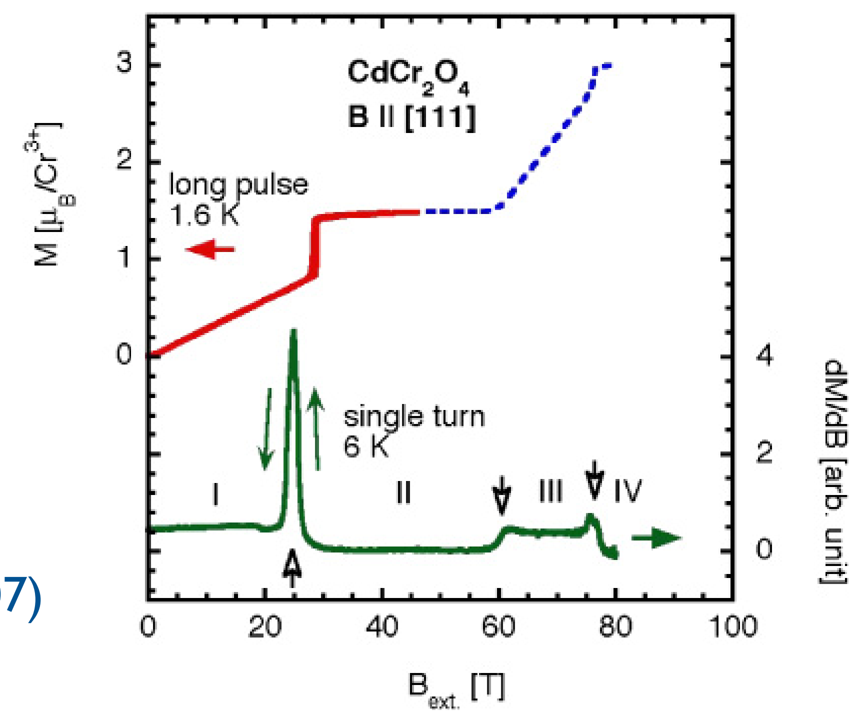
Where does the large magnetization plateau in Cr spinel compounds comes from?

H. Ueda et al.,PRL **94**, 047202 (2005)



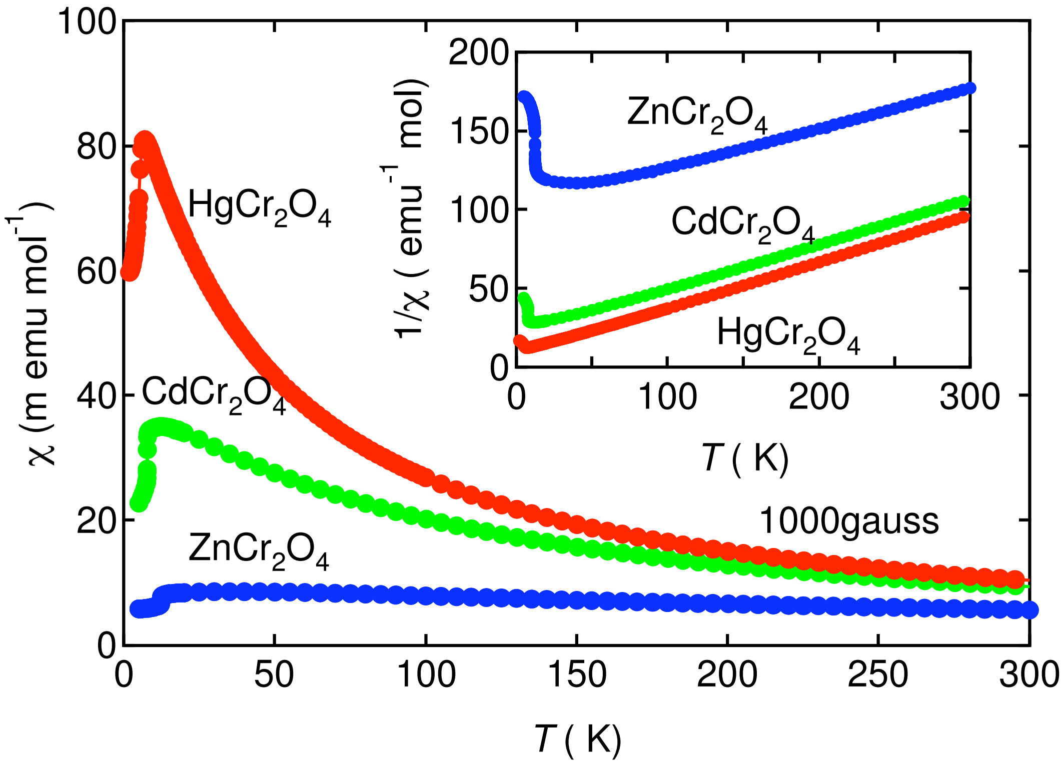
full S=3/2 moment

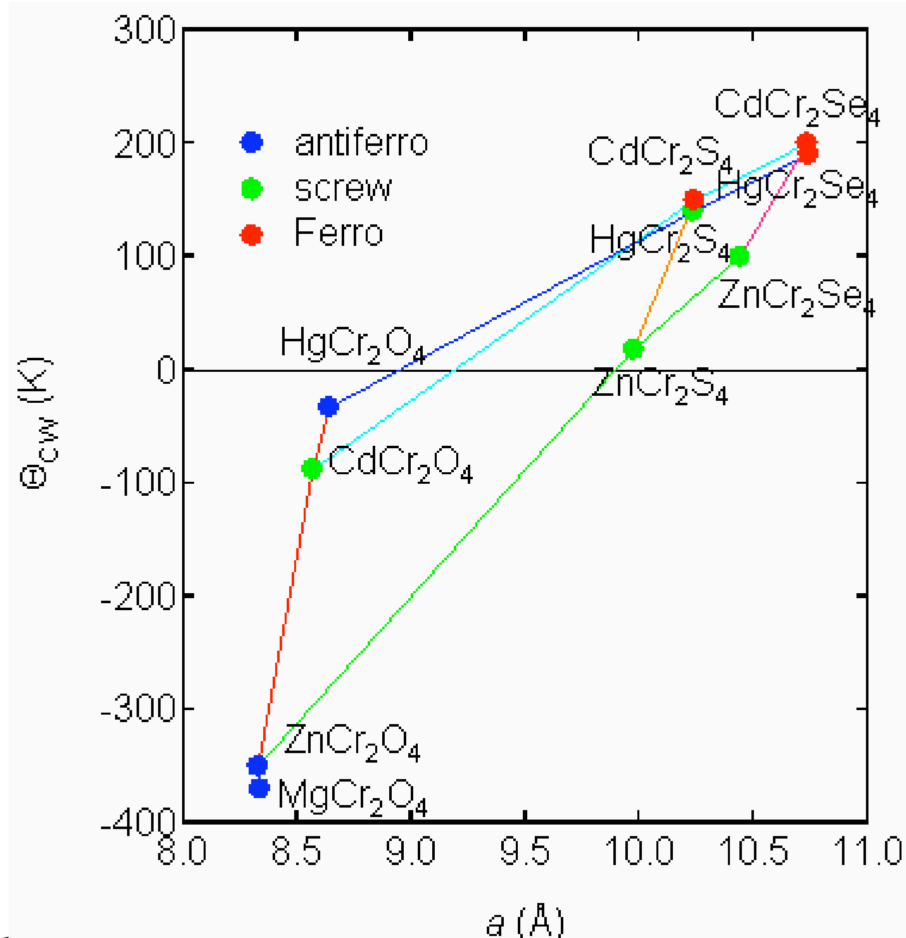
half S=3/2 moment



H. Mitamura et al., JPSJ **76**, 085001 (2007)

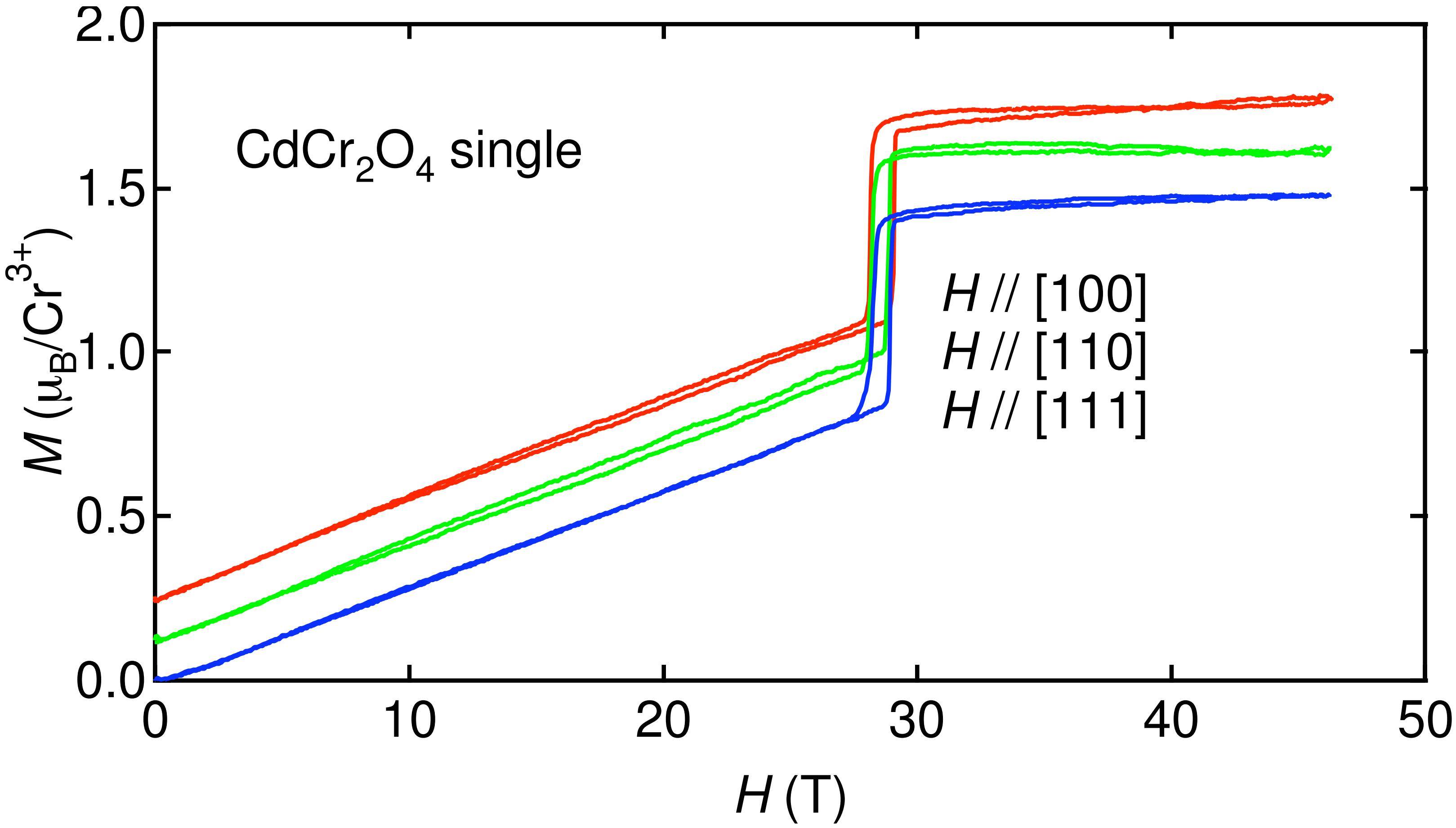
Experimental evidence for frustrated AF interactions





	$MgCr_2O_4$	$ZnCr_2O_4$	$CdCr_2O_4$	$HgCr_2O_4$
T_N(K)	12.5	12	8	6
Θ(K)	−370	−390	−70	−32
$T_N/\|\Theta\|$	0.03	0.03	0.11	0.19

Plateau is independent of the direction of magnetic field - anisotropy negligible



(though anisotropy may be relevant to explain some fine details observed in neutron and ESR [M Yoshida JPSJ 2006] experiments)

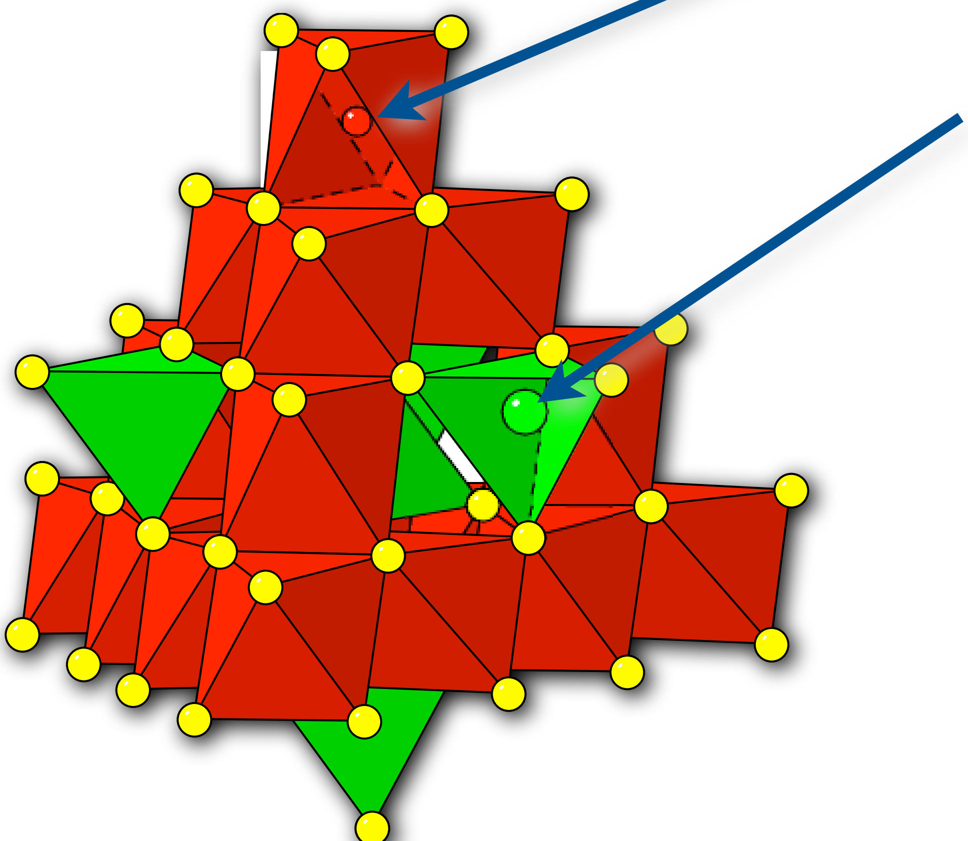
A brief introduction to Cr spinels

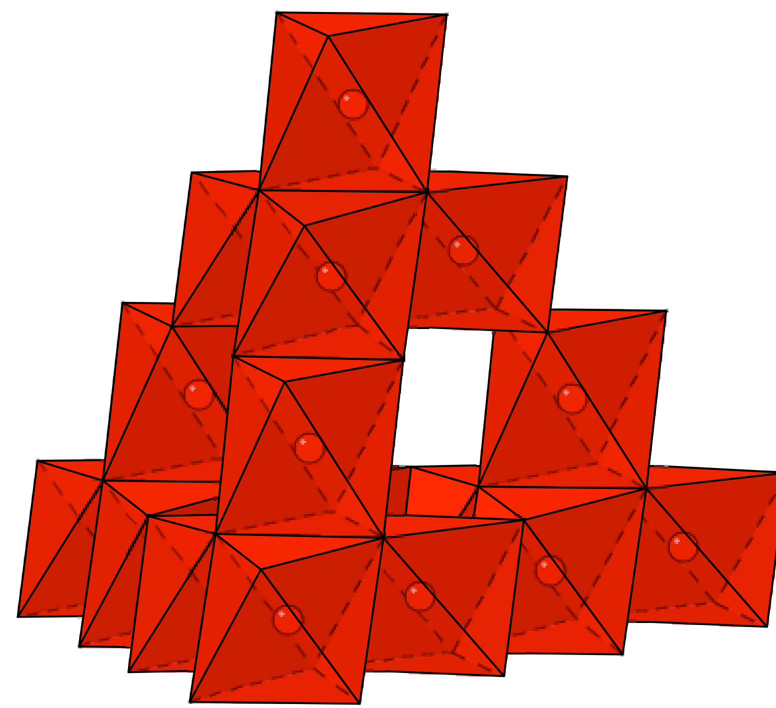
spinel ACr_2X_4

(space group *Fd-3m*)

octahedrally coordinated **magnetic Cr**-site

tetrahedrally coordinated **nonmagnetic A**-site





A brief introduction to Cr spinels

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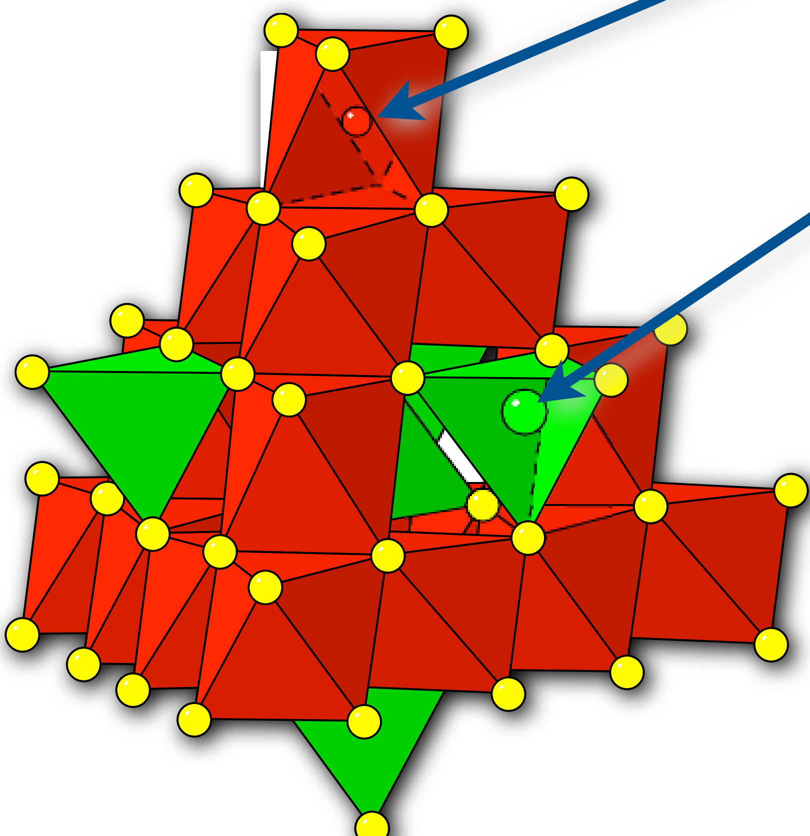

octahedrally coordinated **magnetic Cr**-site

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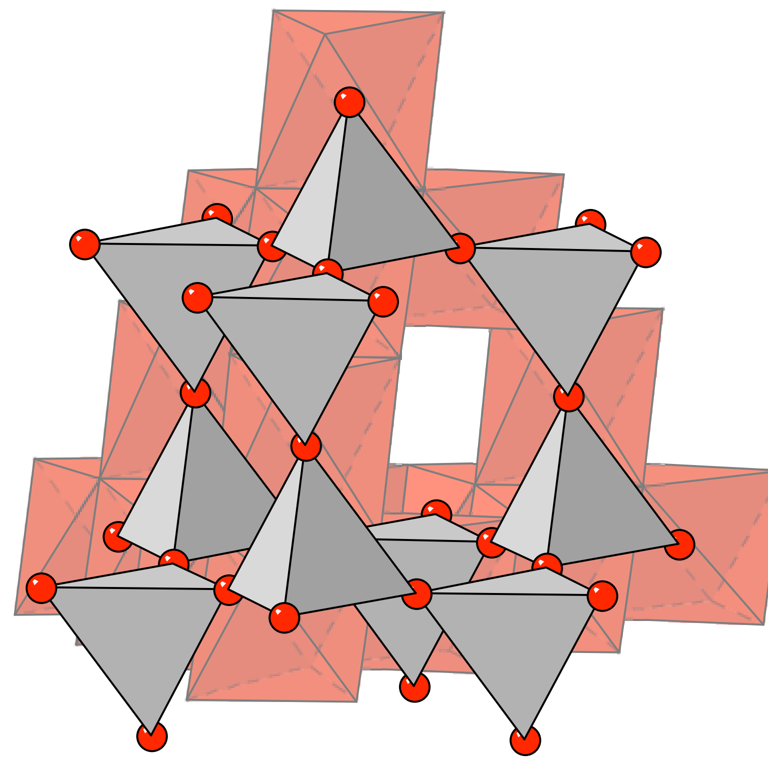
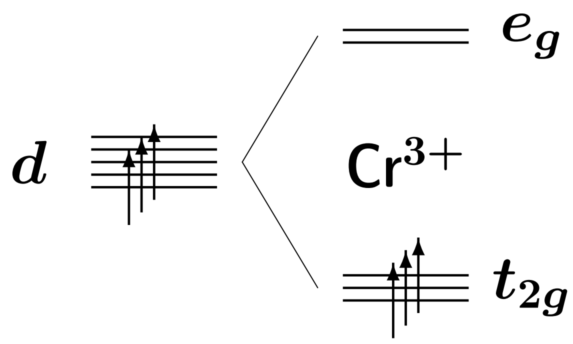


Cr-sublattice: pyrochlore lattice

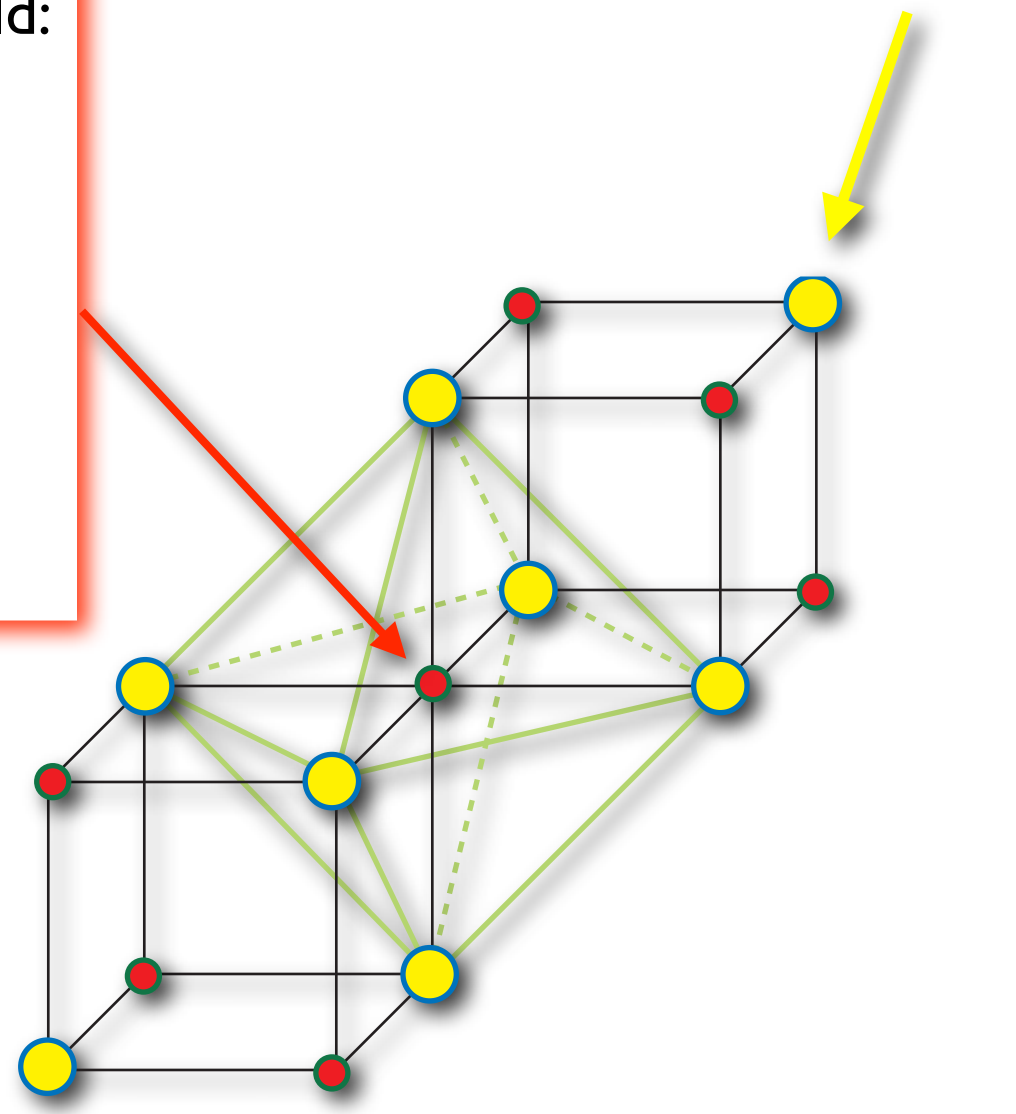
Microscopic structure

in cubic crystal field:



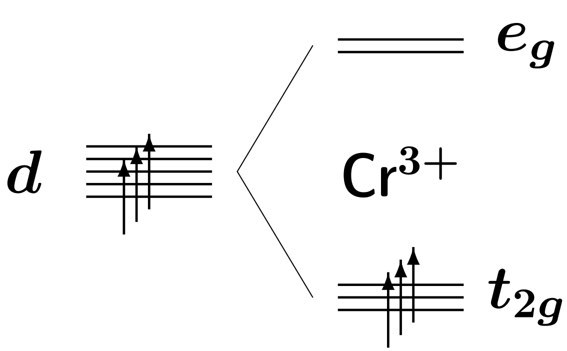
S=3/2, no orbital degrees of freedom

O^{2-} $(2p^2)$

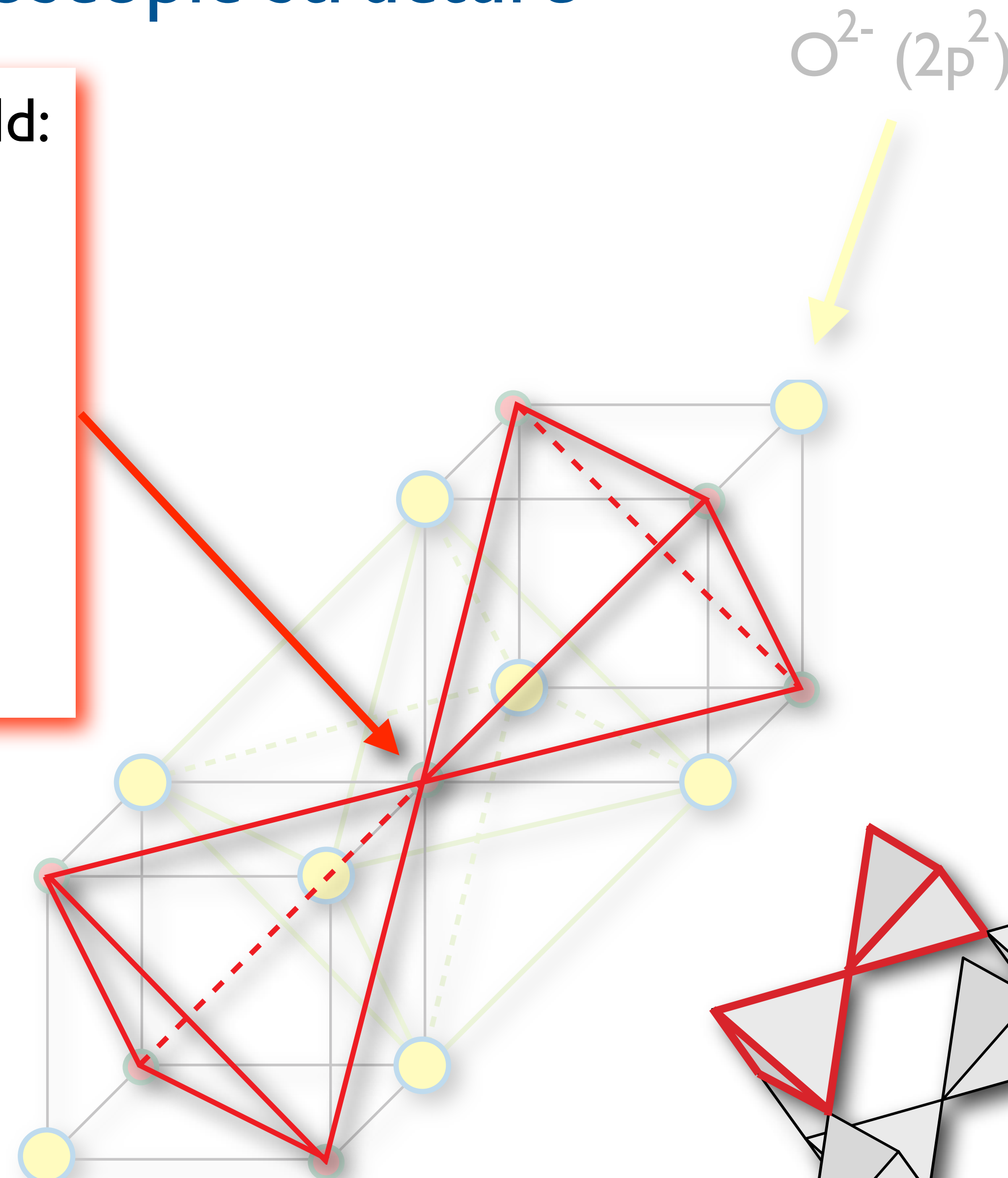


Microscopic structure

in cubic crystal field:



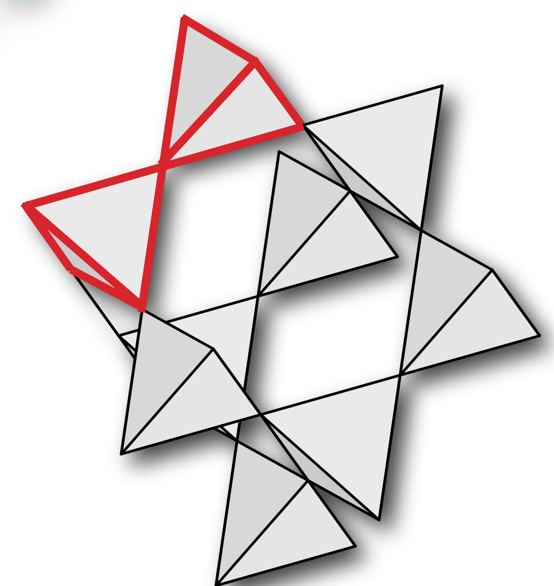
S=3/2, no orbital degrees of freedom



minimal magnetic model :

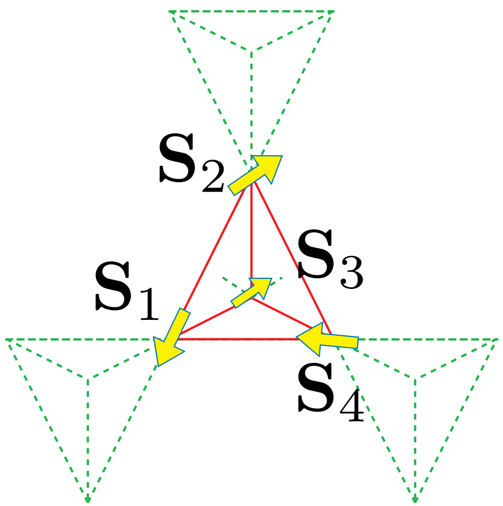
$$\mathcal{H} = J \sum_{\langle i,j \rangle} \mathbf{S}_i \mathbf{S}_j$$

on pyrochlore lattice



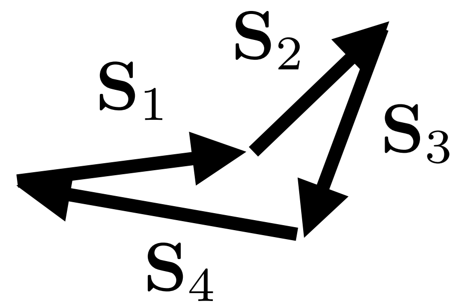
Classical AF on pyrochlore lattice

Energy is a sum of squares :

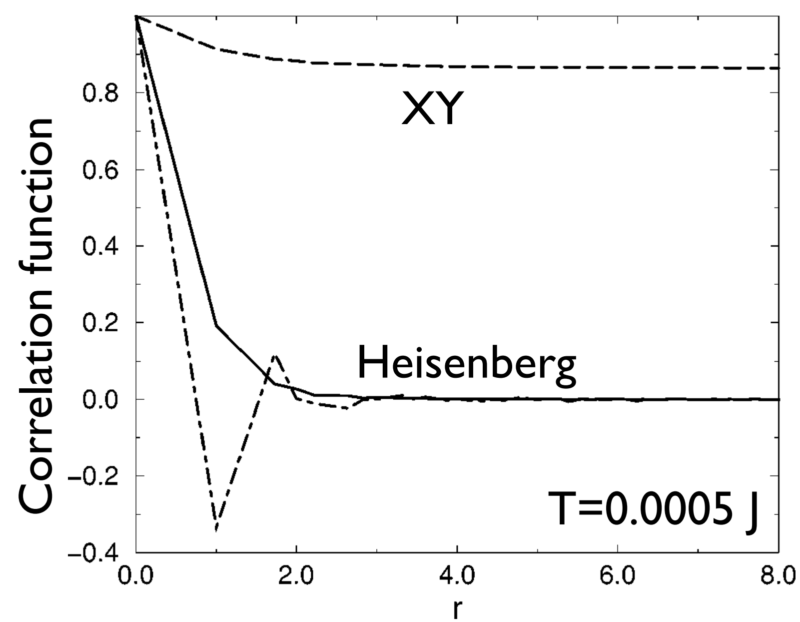


$$\mathcal{H} = J \sum \mathbf{S}_i \mathbf{S}_j$$
$$= \frac{J}{2} \left(\mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3 + \mathbf{S}_4 \right)^2 + \dots$$
$$= 4J \sum_{\text{tet.}} \mathbf{M}^2$$

Ground state manifold on a single tetraheron defined by **M**=0



Due to the residual degeneracy the system remains disordered [Moessner & Chalker, (1998)].

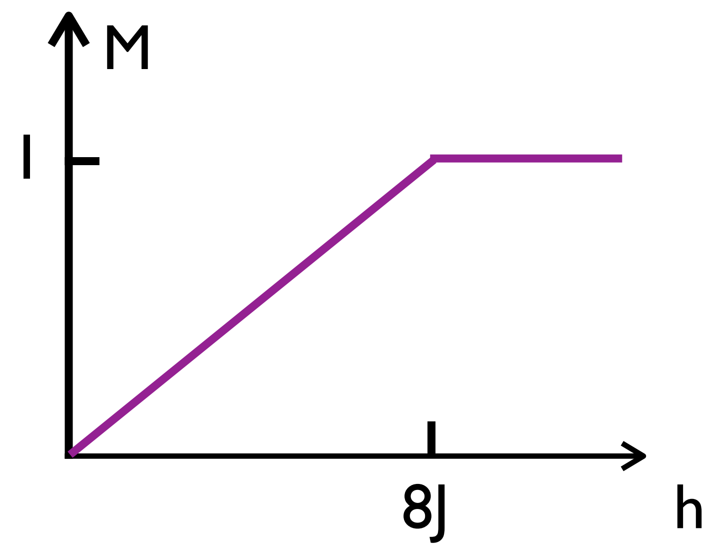


Classical ground state manifold in applied field (T=0)

$$\mathcal{H} = 8J \sum_{\text{tetr.}} \mathbf{M}^2 - 2 \sum_{\text{tetr.}} \mathbf{hM} = 8J \sum_{\text{tetr.}} \underbrace{\left(\mathbf{M} - \frac{\mathbf{h}}{8J} \right)}_{\text{GS if }=0}{}^2 - \sum_{\text{tetr.}} \frac{h^2}{8J}$$

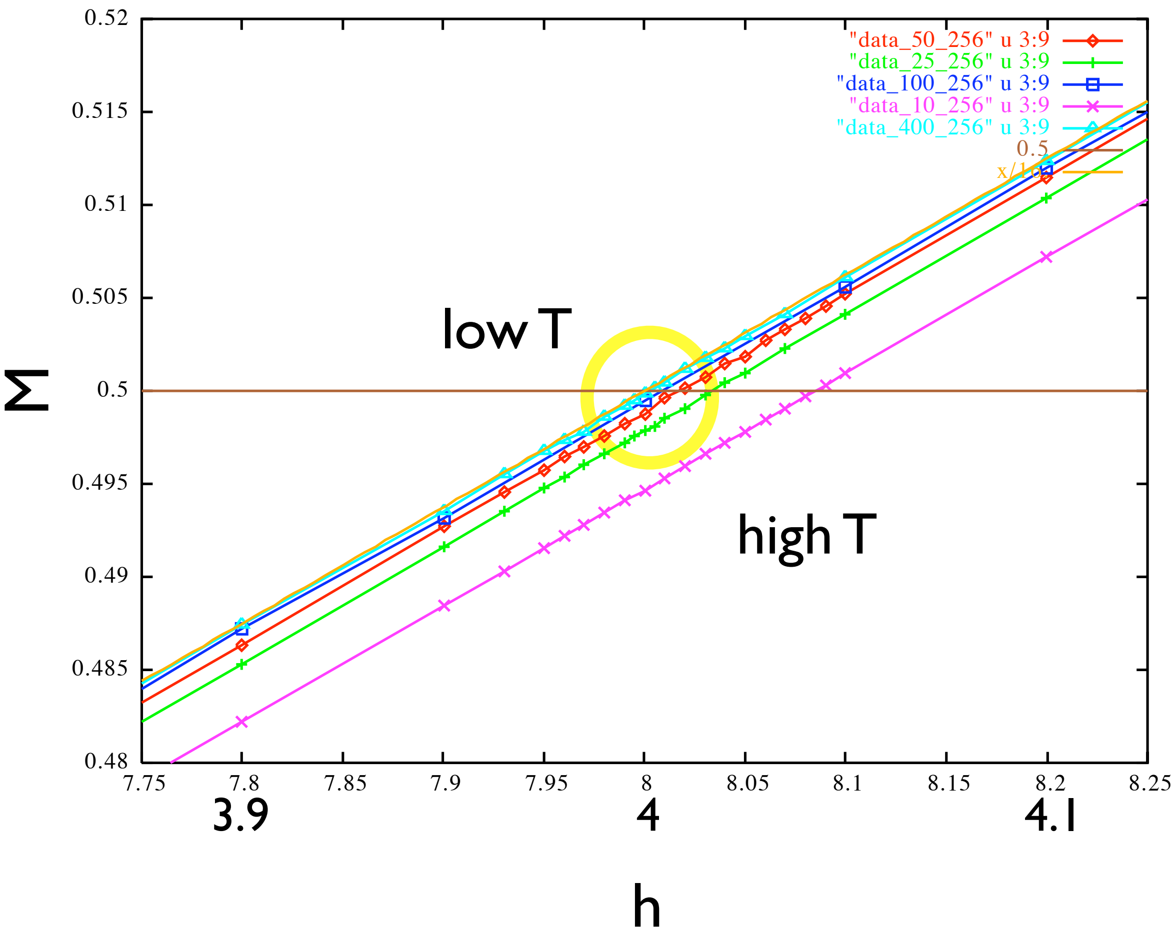
For each tetrahedron, require : $\mathbf{M} = \dfrac{\mathbf{h}}{8J}$

Ground state degeneracy survives and magnetization is linear up to saturation.



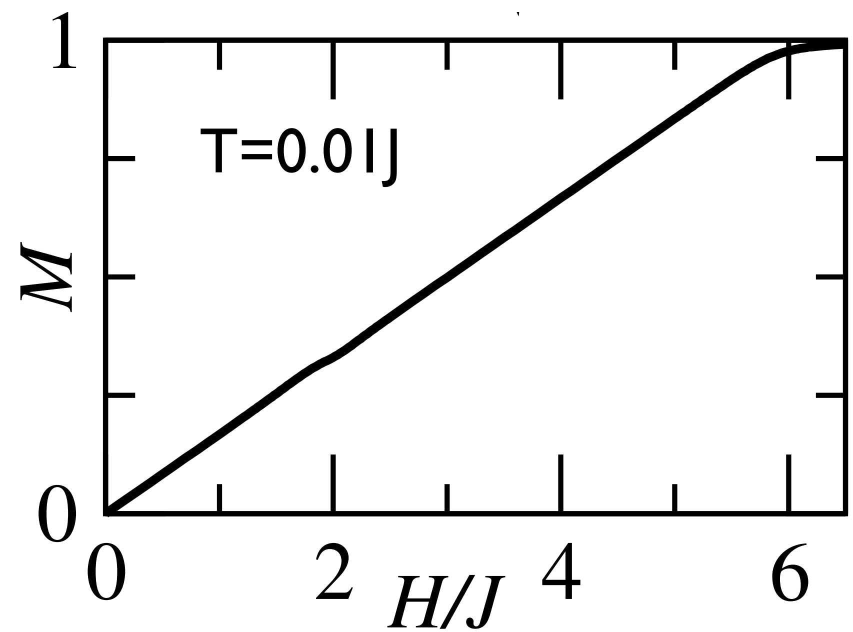
finite T: order by disorder scenario does not work

simple MC simulations of classical pyrochlore model



Thermal fluctuations do not stabilize a collinear state (plateau)

MC simulations of classical kagome model [Zhitomirsky, PRL **88**, 057204 (2002)]

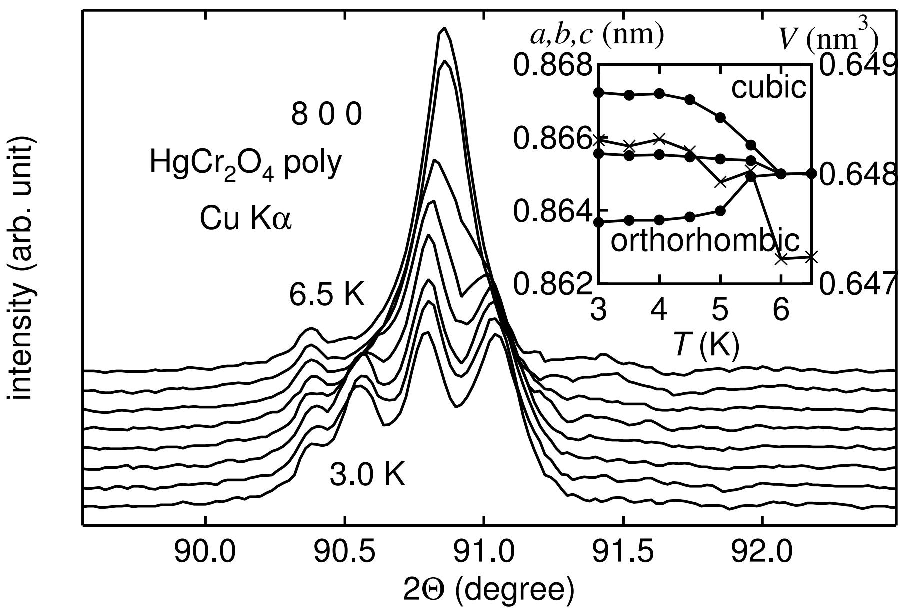


What is missing ?

magnetoelastic coupling: h=0

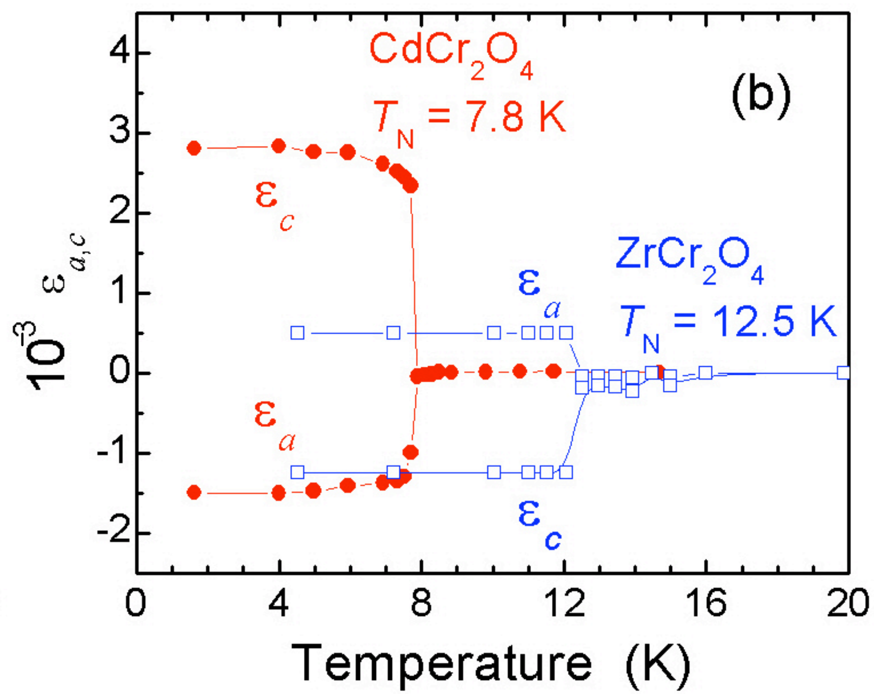
magnetic ordering accompanied with a structural transition:

orthorhombic ← cubic



H. Ueda *et al.*, PRB **73**, 094415 (2006)

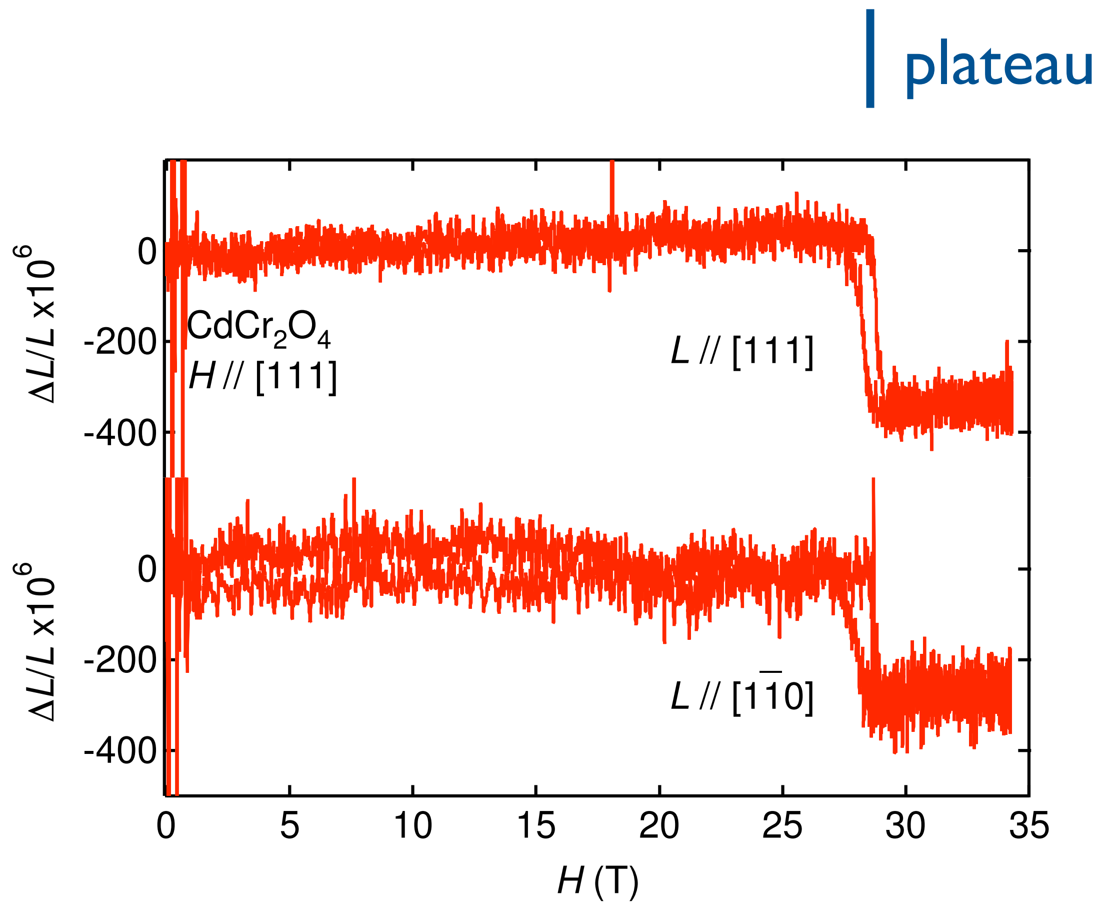
tetragonal ← cubic



J-H Chung *et al.*, PRL **95**, 247204 (2005)

magnetoelastic coupling: h>0

strong magnetostriction entering the plateau phase:



H. Ueda et al.,PRL **94**, 047202 (2005)

Coupling to lattice distortions for h=0

YMn_2: Terao JPSJ **65**, 1413 (1996), Canals & Lacroix PRB **61**, 1149 (2000)

$Y_2M_2O_7$: Keren & Gardner PRL **87**, 177201 (2001)

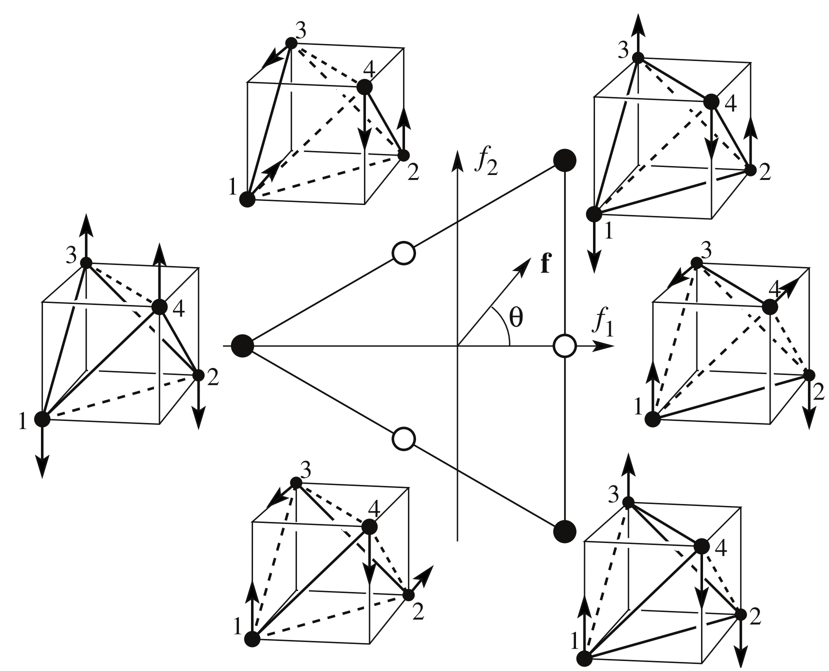
Yamashita & Ueda, PRL **85**, 4960 (2000):

In a single tetrahedron of S=1/2 spins ground state 2x degenerate, E irrep.
Coupled tetrahedra: VBS-like theory of ZnV_2O_4

Tchernyshyov, Moessner, & Sondhi,
PRL **88**, 067203 (2002):

"Order by Distortion"

Landau-like theory of the spin-Peierls mechanism in the E irrep.



How does lattice distortion affect magnetic order ?

spin exchange depends on distance: $$J(r) = J(r_0) + \left.\frac{\partial J}{\partial r}\right|_{r_0} \delta r = J(1 + \alpha\rho)$$

Consider generalized "spin-Peierls" Hamiltonian :

$$\mathcal{H} = \sum_{\langle i,j \rangle} \left[J(1 - \alpha\rho_{i,j})\mathbf{S}_i\mathbf{S}_j + \frac{K}{2}\rho_{i,j}^2 \right] - \mathbf{h}\sum_i \mathbf{S}_i$$

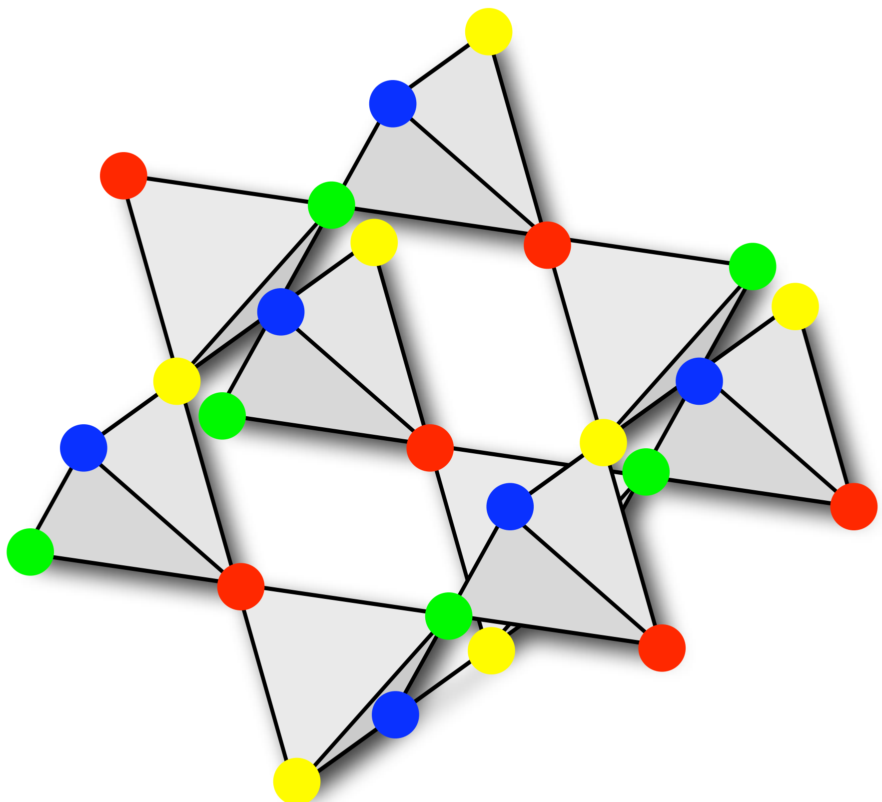
spin-lattice coupling

elastic energy

the elastic energy is quadratic - we can integrate it out:

- it leads to long range spin-spin effective interaction
- for realistic description we shall take realistic phonon modes
- we want to understand the basic mechanism, so we look at the simplest case (affine deformations)

Minimal symmetry breaking solution

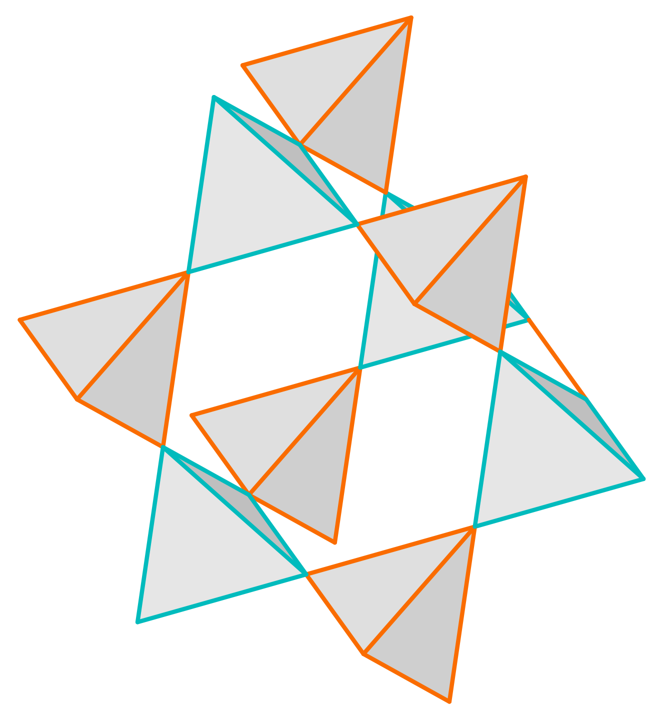


The four-sublattice ordering does not break the translational symmetry (uniform q=0 distortions).
The point group symmetry is broken.

The four-sublattice ordering can be stabilized e.g. by AF J2 or FM J3.

full point group is $O_h = T_d \times \{1, I\}$

site-factorized wave function is invariant under inversion I
$\Rightarrow$ only T_d remains



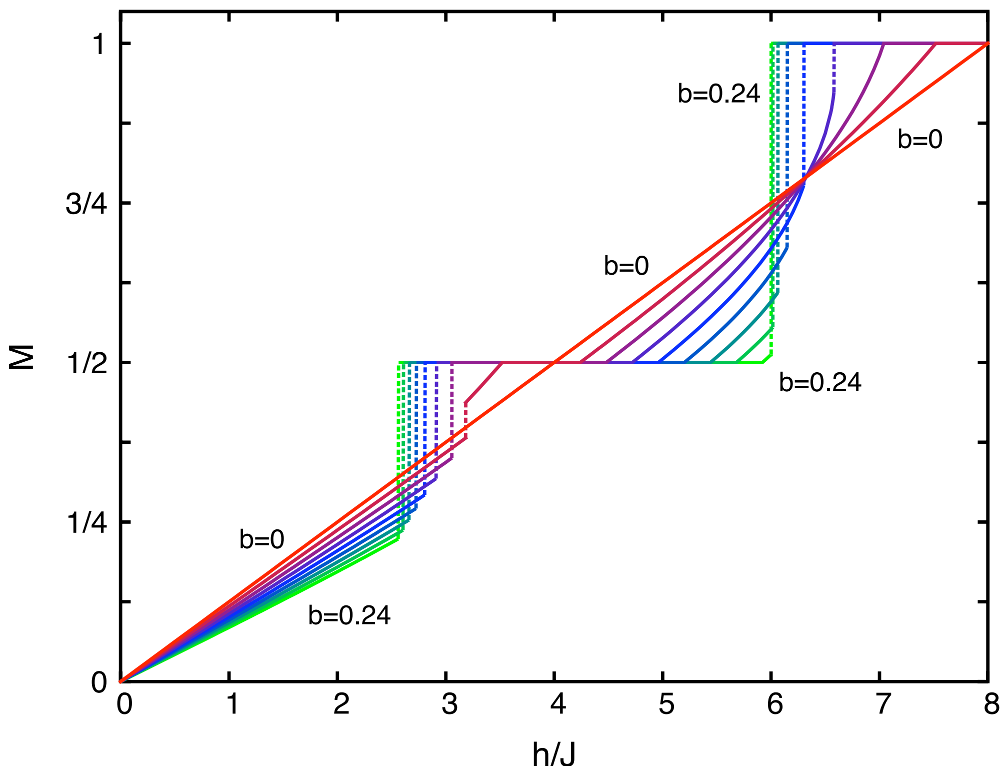
Minimazing the energy of a tetrahedron (4LRO state)

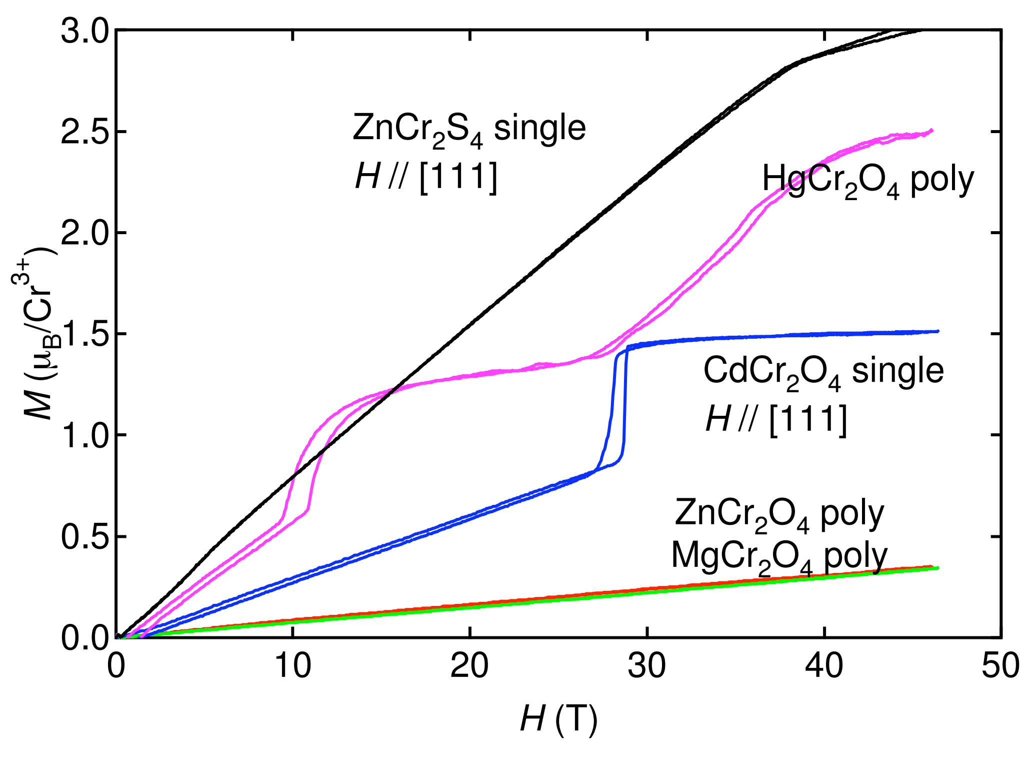
The ground state problem is reduced to pure spin energy (assuming 4 sublattice LRO):

$$\mathcal{H} = \sum_{\langle i,j \rangle} J \left[\mathbf{S}_i \mathbf{S}_j - b(\mathbf{S}_i \mathbf{S}_j)^2 \right] - \mathbf{h} \sum_i \mathbf{S}_i$$

favours collinear spin configurations !

T=0 classical



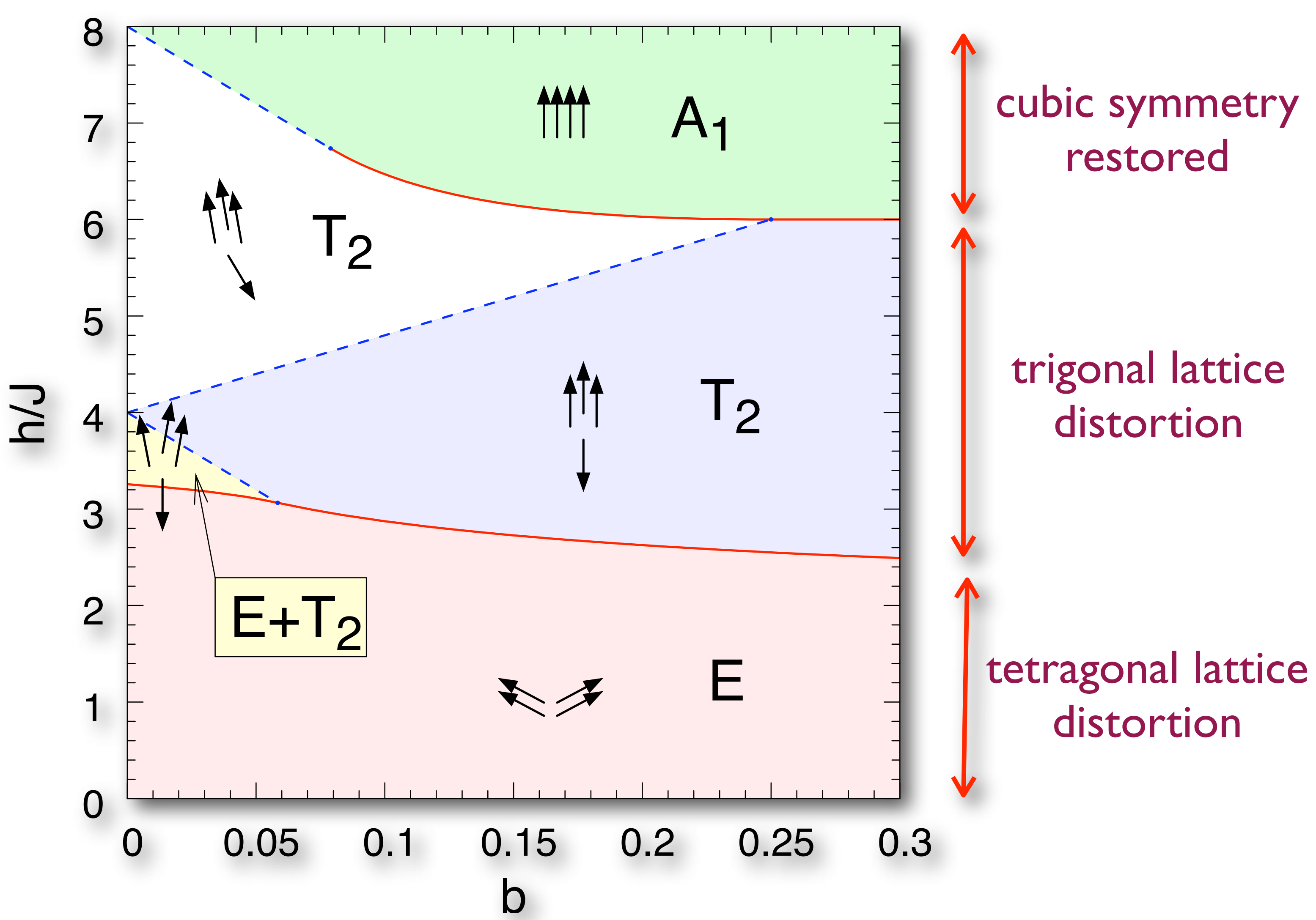


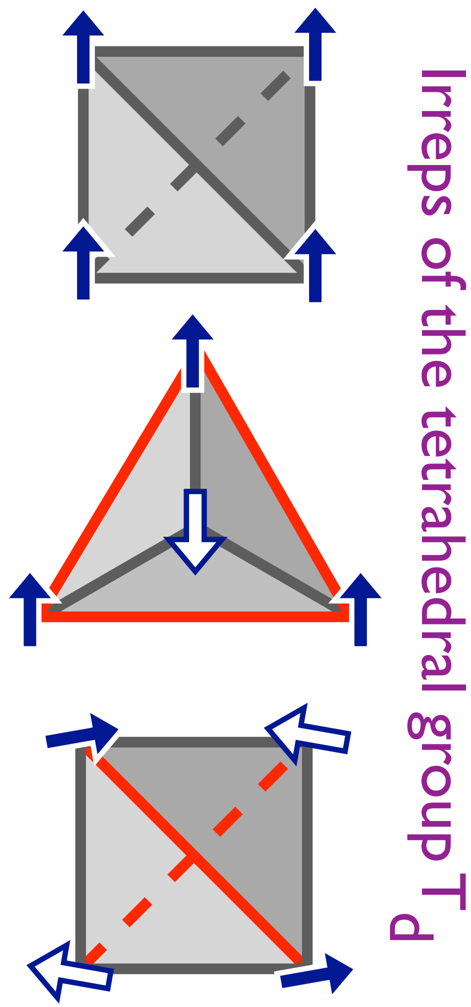
Irreps of tetrahedral symmetry group Td :

$$\begin{pmatrix} \rho_{\mathsf{A}_1} \\ \rho_{\mathsf{E},1} \\ \rho_{\mathsf{E},2} \\ \rho_{\mathsf{T}_2,1} \\ \rho_{\mathsf{T}_2,2} \\ \rho_{\mathsf{T}_2,3} \end{pmatrix} = \begin{pmatrix} \frac{1}{\sqrt{6}} & \frac{1}{\sqrt{6}} & \frac{1}{\sqrt{6}} & \frac{1}{\sqrt{6}} & \frac{1}{\sqrt{6}} & \frac{1}{\sqrt{6}} \\ \frac{1}{\sqrt{3}} & \frac{-1}{2\sqrt{3}} & \frac{-1}{2\sqrt{3}} & \frac{-1}{2\sqrt{3}} & \frac{-1}{2\sqrt{3}} & \frac{1}{\sqrt{3}} \\ 0 & \frac{1}{2} & -\frac{1}{2} & -\frac{1}{2} & \frac{1}{2} & 0 \\ 0 & 0 & \frac{-1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 & 0 \\ 0 & \frac{-1}{\sqrt{2}} & 0 & 0 & \frac{1}{\sqrt{2}} & 0 \\ \frac{-1}{\sqrt{2}} & 0 & 0 & 0 & 0 & \frac{1}{\sqrt{2}} \end{pmatrix} \begin{pmatrix} \rho_{1,2} \\ \rho_{1,3} \\ \rho_{1,4} \\ \rho_{2,3} \\ \rho_{2,4} \\ \rho_{3,4} \end{pmatrix}$$

A_1	E	T_2
1 dim.	2 dim.	3 dim.

The phase diagram





PRL **93**, 197203 (2004)

Why are these particular phases stable ?

Irreps of tetrahedral symmetry group Td :

$$\begin{pmatrix} \Lambda_{\mathrm{A}} \\ \Lambda_{\mathrm{E},1} \\ \Lambda_{\mathrm{E},2} \\ \Lambda_{\mathrm{T}_2,1} \\ \Lambda_{\mathrm{T}_2,2} \\ \Lambda_{\mathrm{T}_2,3} \end{pmatrix} = \begin{pmatrix} \frac{1}{\sqrt{6}} & \frac{1}{\sqrt{6}} & \frac{1}{\sqrt{6}} & \frac{1}{\sqrt{6}} & \frac{1}{\sqrt{6}} & \frac{1}{\sqrt{6}} \\ \frac{1}{\sqrt{3}} & \frac{-1}{2\sqrt{3}} & \frac{-1}{2\sqrt{3}} & \frac{-1}{2\sqrt{3}} & \frac{-1}{2\sqrt{3}} & \frac{1}{\sqrt{3}} \\ 0 & \frac{1}{2} & \frac{-1}{2} & \frac{-1}{2} & \frac{1}{2} & 0 \\ 0 & 0 & \frac{-1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 & 0 \\ 0 & \frac{-1}{\sqrt{2}} & 0 & 0 & \frac{1}{\sqrt{2}} & 0 \\ \frac{-1}{\sqrt{2}} & 0 & 0 & 0 & 0 & \frac{1}{\sqrt{2}} \end{pmatrix} \begin{pmatrix} \vec{S}_1 \cdot \vec{S}_2 \\ \vec{S}_1 \cdot \vec{S}_3 \\ \vec{S}_1 \cdot \vec{S}_4 \\ \vec{S}_2 \cdot \vec{S}_3 \\ \vec{S}_2 \cdot \vec{S}_3 \\ \vec{S}_3 \cdot \vec{S}_4 \end{pmatrix}$$

In terms of these, the Hamiltonian reads :

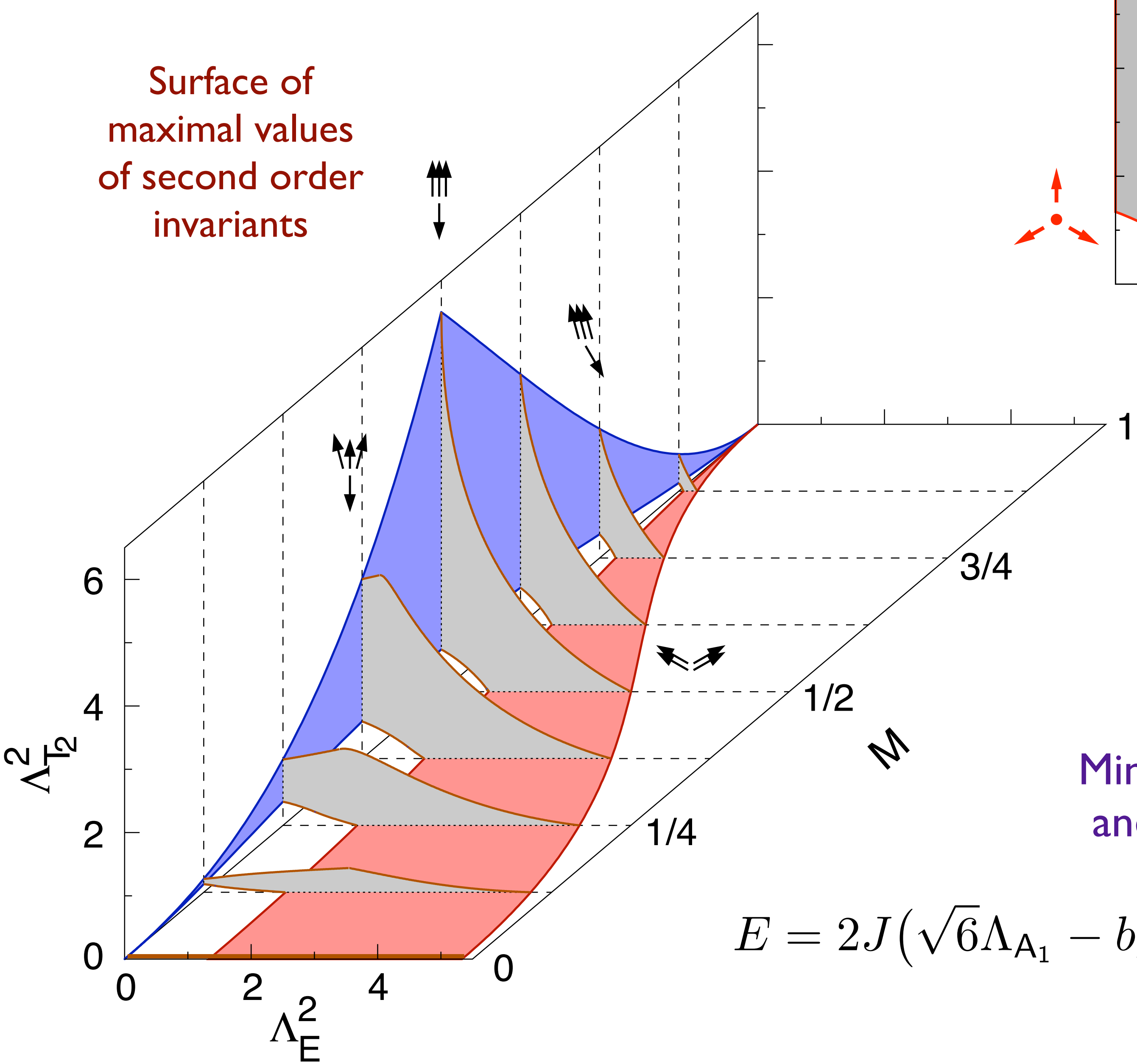
$$\begin{aligned} \mathcal{H} \;&=\; 2\sqrt{6}J\Lambda_{\mathsf{A}} - 2\alpha J\left(\Lambda_{\mathsf{A}}\rho_{\mathsf{A}} + \mathbf{\Lambda}_{\mathsf{E}}\boldsymbol{\rho}_{\mathsf{E}} + \mathbf{\Lambda}_{\mathsf{T}_2}\boldsymbol{\rho}_{\mathsf{T}_2}\right) \\ &\quad + K\left(\rho_{\mathsf{A}}^2 + \rho_{\mathsf{E}}^2 + \rho_{\mathsf{T}_2}^2\right) - 4\mathbf{hM} \end{aligned}$$

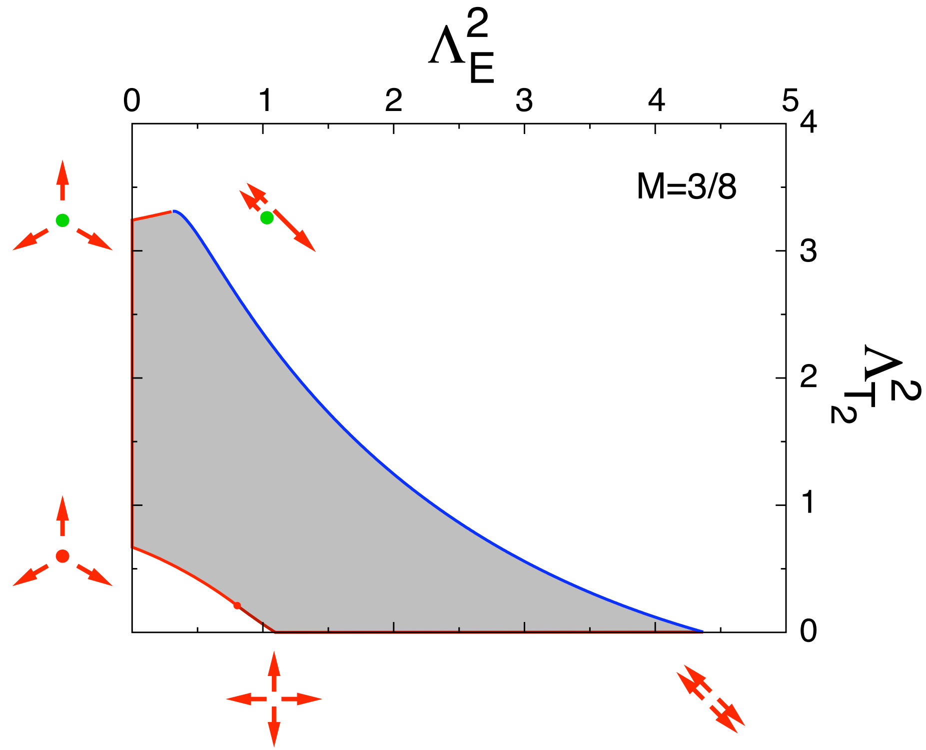
Eliminating the distances:

$$E = 2J\left(\sqrt{6}\Lambda_{\mathsf{A}_1} - b_{\mathsf{A}_1}\Lambda_{\mathsf{A}_1}^2 - b_{\mathsf{E}}\Lambda_{\mathsf{E}}^2 - b_{\mathsf{T}_2}\Lambda_{\mathsf{T}_2}^2\right) - 4\mathbf{hM}$$

Why are these particular phases stable ?

Surface of maximal values of second order invariants





Minimize energy as a function of M and look to see which irrep wins.

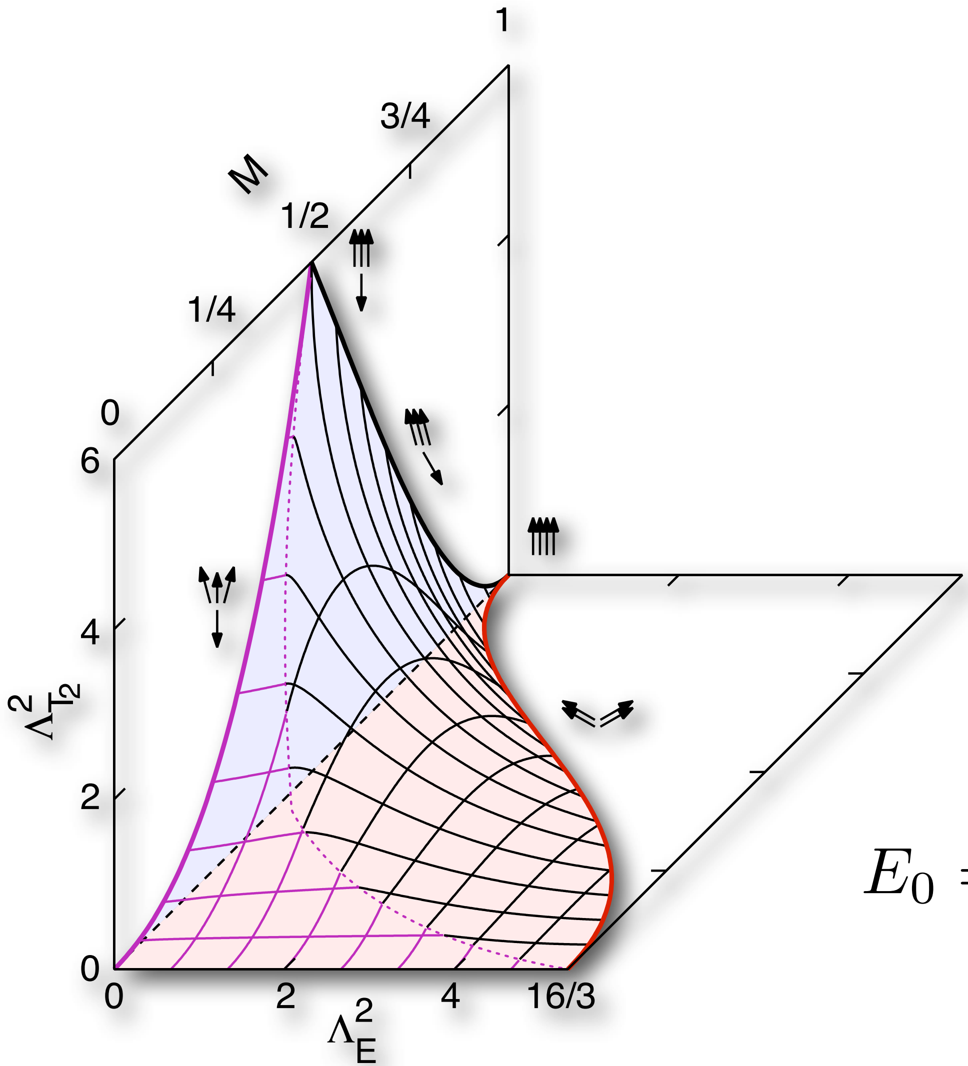
$$E = 2J\left(\sqrt{6}\Lambda_{A_1} - b_{A_1}\Lambda_{A_1}^2 - b_E\Lambda_E^2 - b_{T_2}\Lambda_{T_2}^2\right) - 4\mathbf{hM}$$

cusp $\Rightarrow$ stable plateau with T_2 symmetry

Energy as a function of magnetization :

$$E = E_0 - 4\mathbf{h}\mathbf{M}$$

$$E = E_0$$



E symmetry state

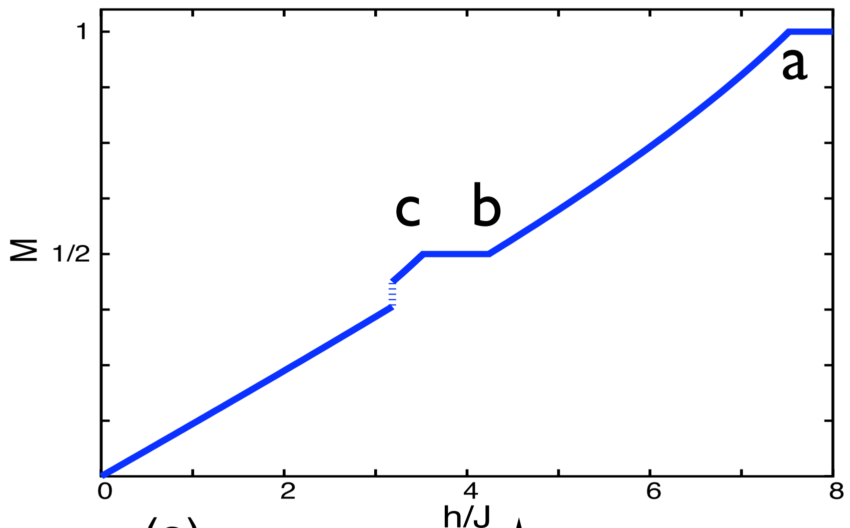
T2 symmetry state

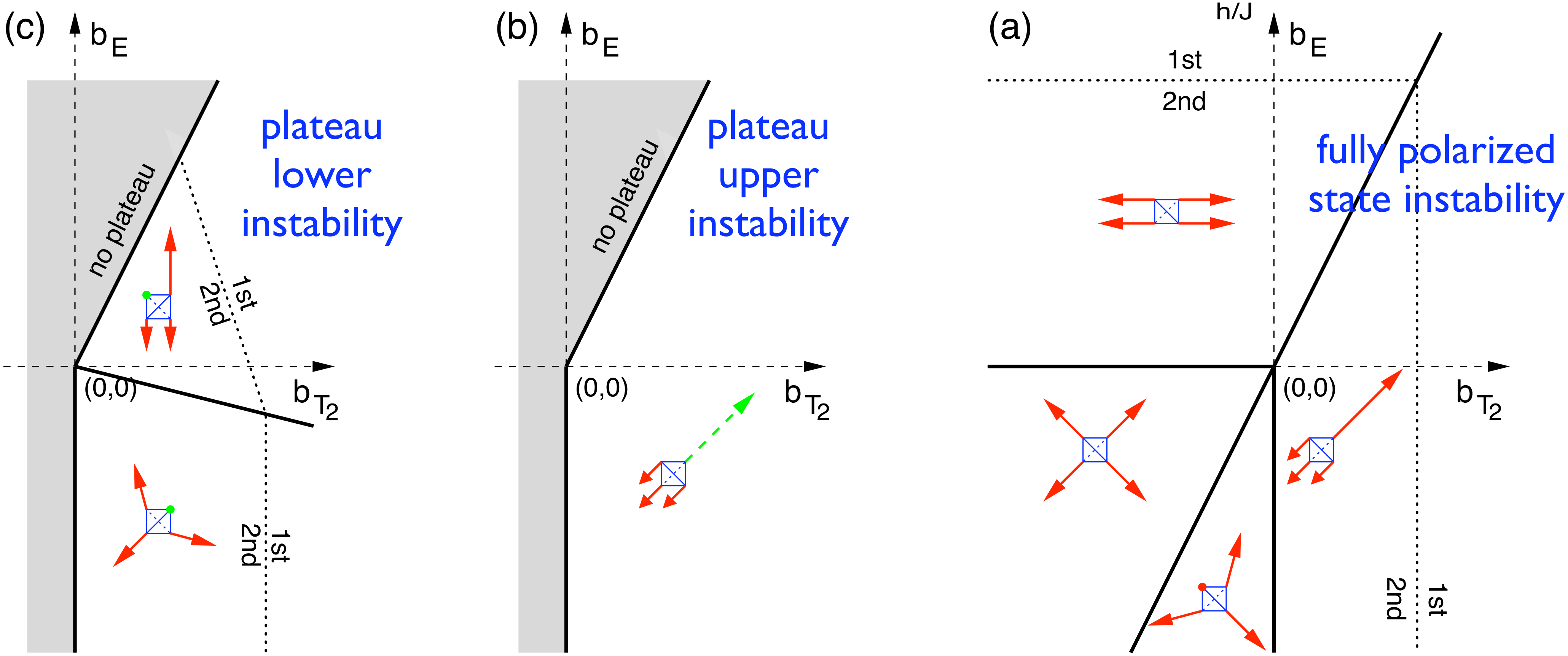
$$E_0 = 2J\left(\sqrt{6}\Lambda_{A_1} - b_{A_1}\Lambda^2_{A_1} - b_E\Lambda^2_E - b_{T_2}\Lambda^2_{T_2}\right)$$

Local instability of collinear states

$$E = 2J\left(\sqrt{6}\Lambda_{A_1} - b_{A_1}\Lambda_{A_1}^2 - b_E\Lambda_E^2 - b_{T_2}\Lambda_{T_2}^2\right) - 4\mathbf{hM}$$

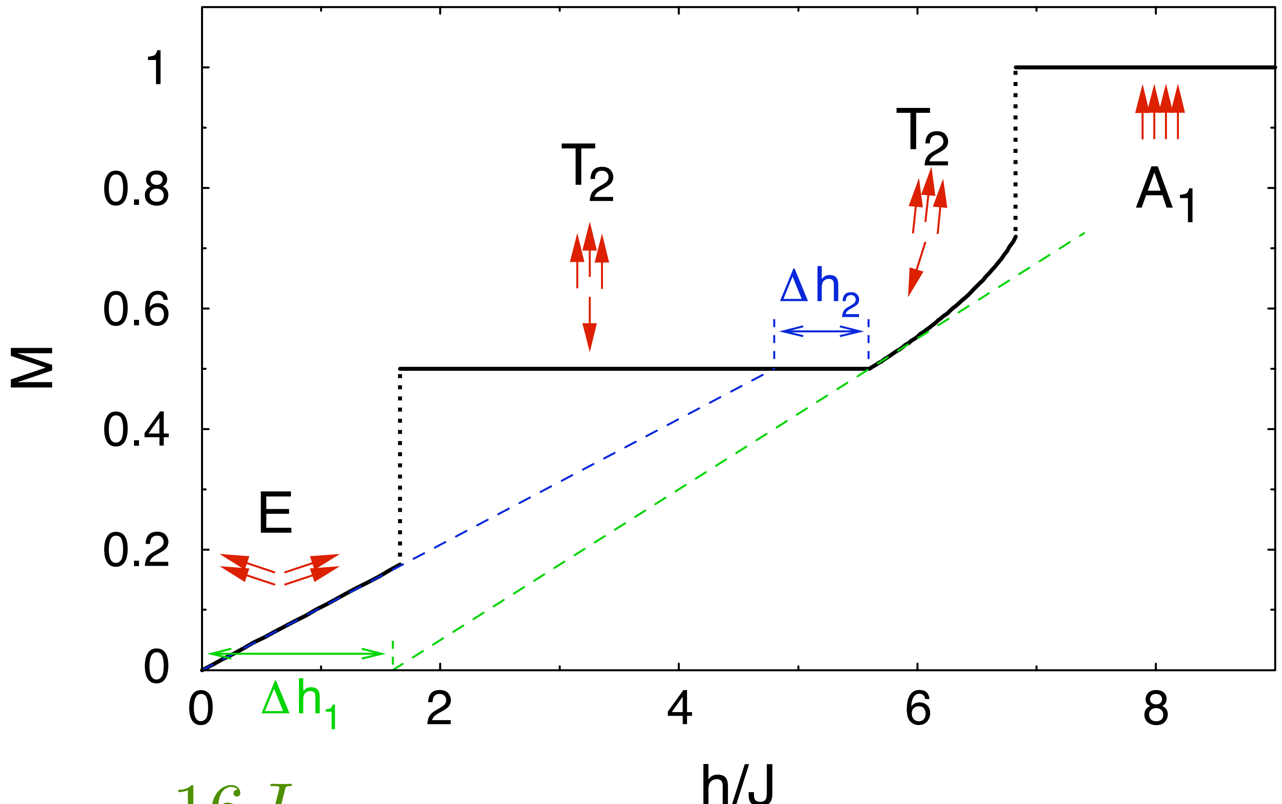
J. Phys.: Condens. Matter **19**, 145267 (2007).





Magnetization curve and b's

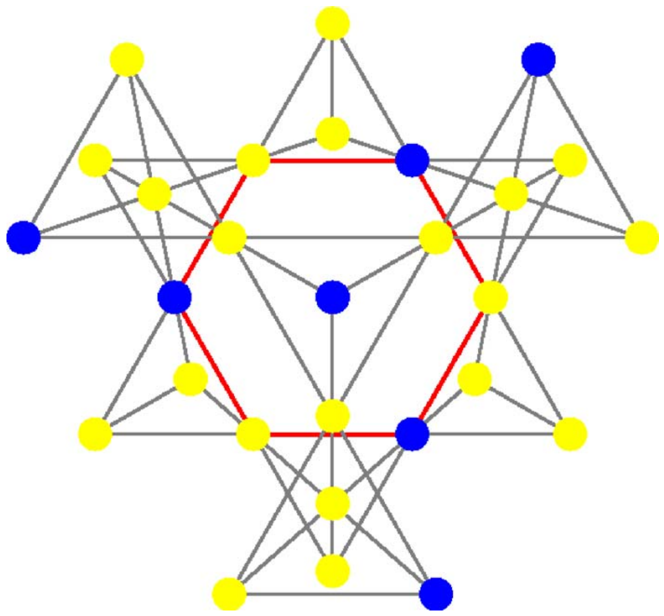
$$\Delta h_2 = \frac{8J}{3}(3b_{\mathsf{T}_2} - b_{\mathsf{A}_1} - 2b_{\mathsf{E}})$$



$$\Delta h_1 = \frac{16J}{3}(b_{\mathsf{A}_1} + b_{\mathsf{T}_2})$$

$$h_u = 4J(1 + 2b_{\mathsf{T}_2})$$

Back to spinels



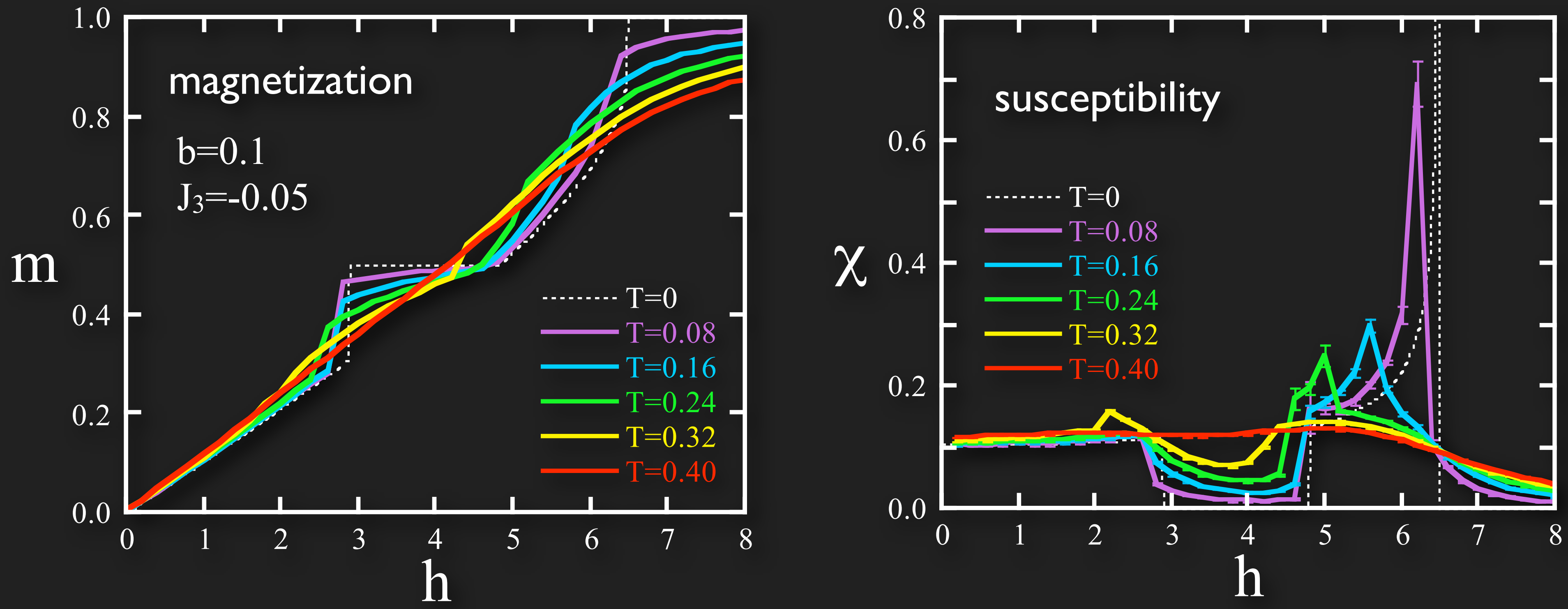
In the real material ($HgCr_2O_4$, Matsuda *et al*., Nature Physics **3**, 397 (2007)), the plateau state is not a 4 sublattice, but a more complicated, 16 sublattice state.

Phonons: Bergman *et al*. Phys. Rev. B **74**, 134409 (2006) : Einstein model incorporating local site distortions can lead to 16 sublattice plateau state.

Exchanges: Longer range exchanges can also select the 16 sublattice state.

Magnetoelastic coupling does not lead necessarily to plateau: in $ZnCr_2Se_4$ magnetostriction, but M linear up to saturation (Hemberger et al., PRL **98**, 147203 (2007)).

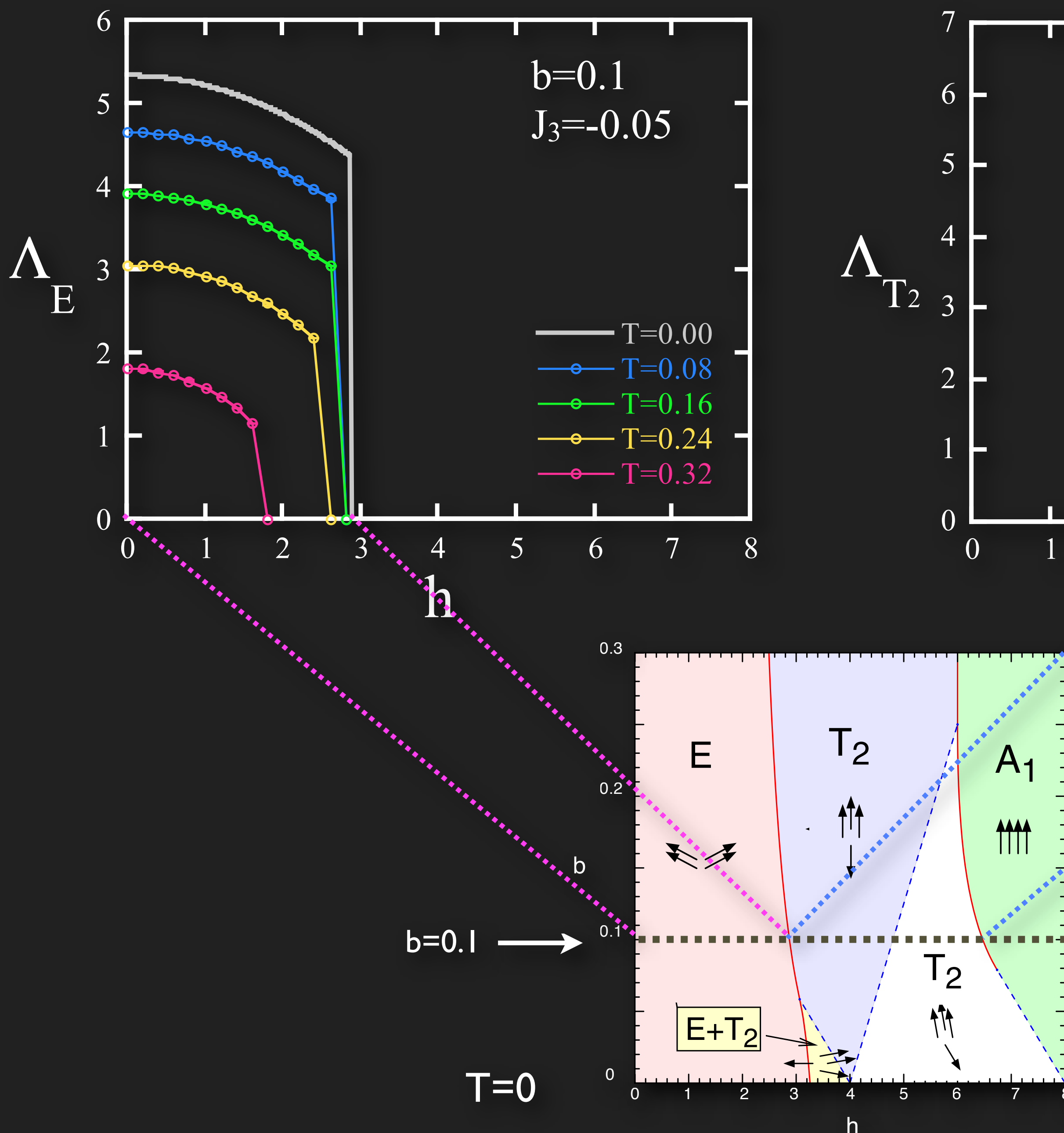
Monte Carlo Results at finite T (biquadratic effective model)



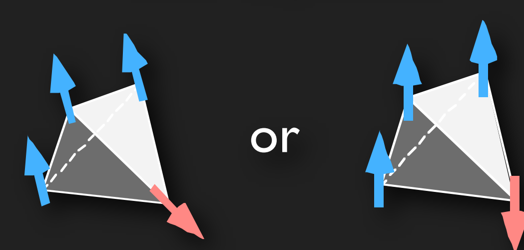
half-magnetization plateau survives at finite temperatures

J. Magn. Magn. Mater. **300**, 57 (2006)

Order Parameters - I



How to distinguish two T_2 phases?

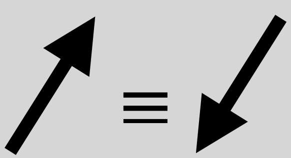


Order Parameters - II

nematic order parameter, measures coplanarity:

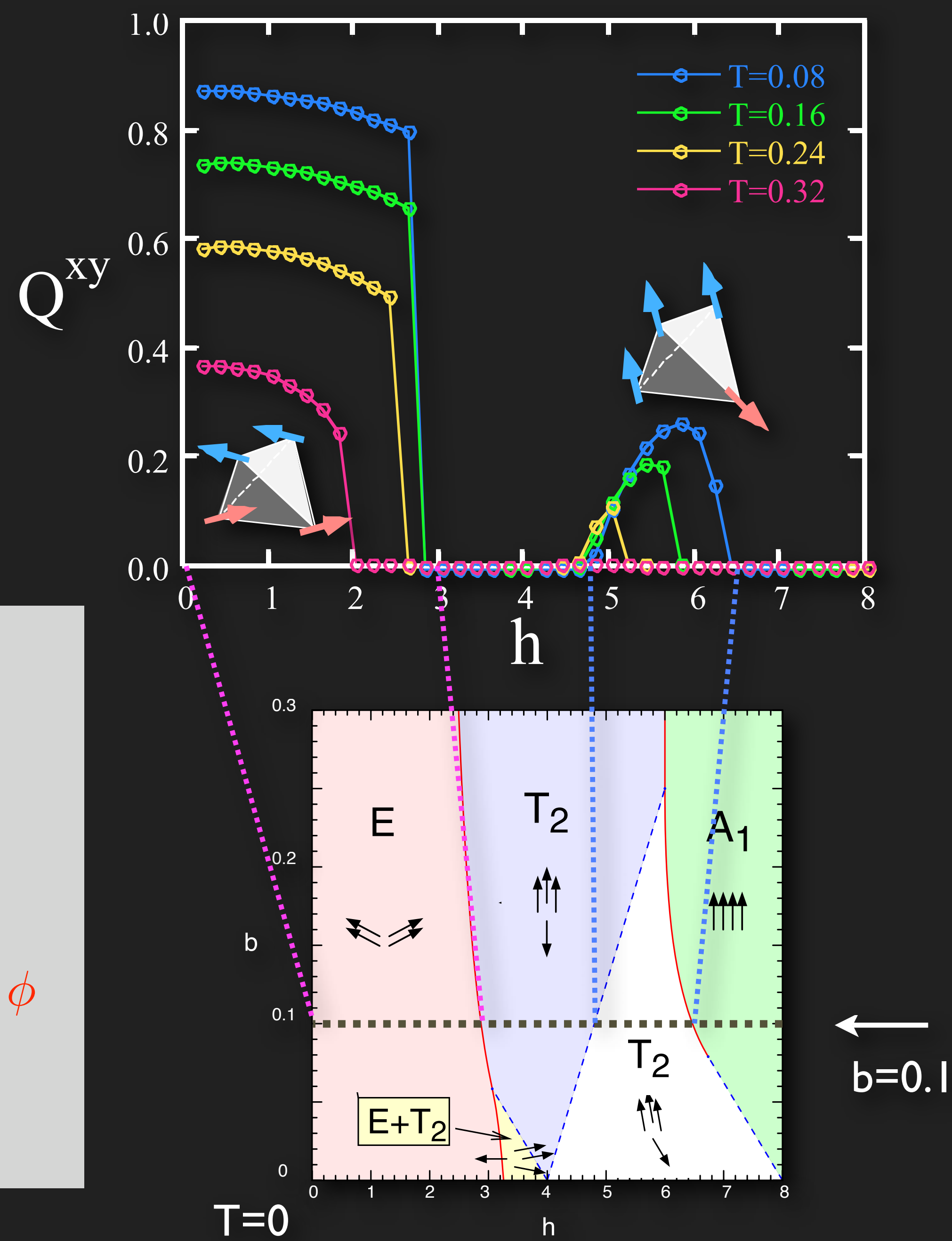
$$Q^{x^2-y^2} = \langle S^x S^x - S^y S^y \rangle$$

$$Q^{xy} = \langle 2 S^x S^x \rangle$$

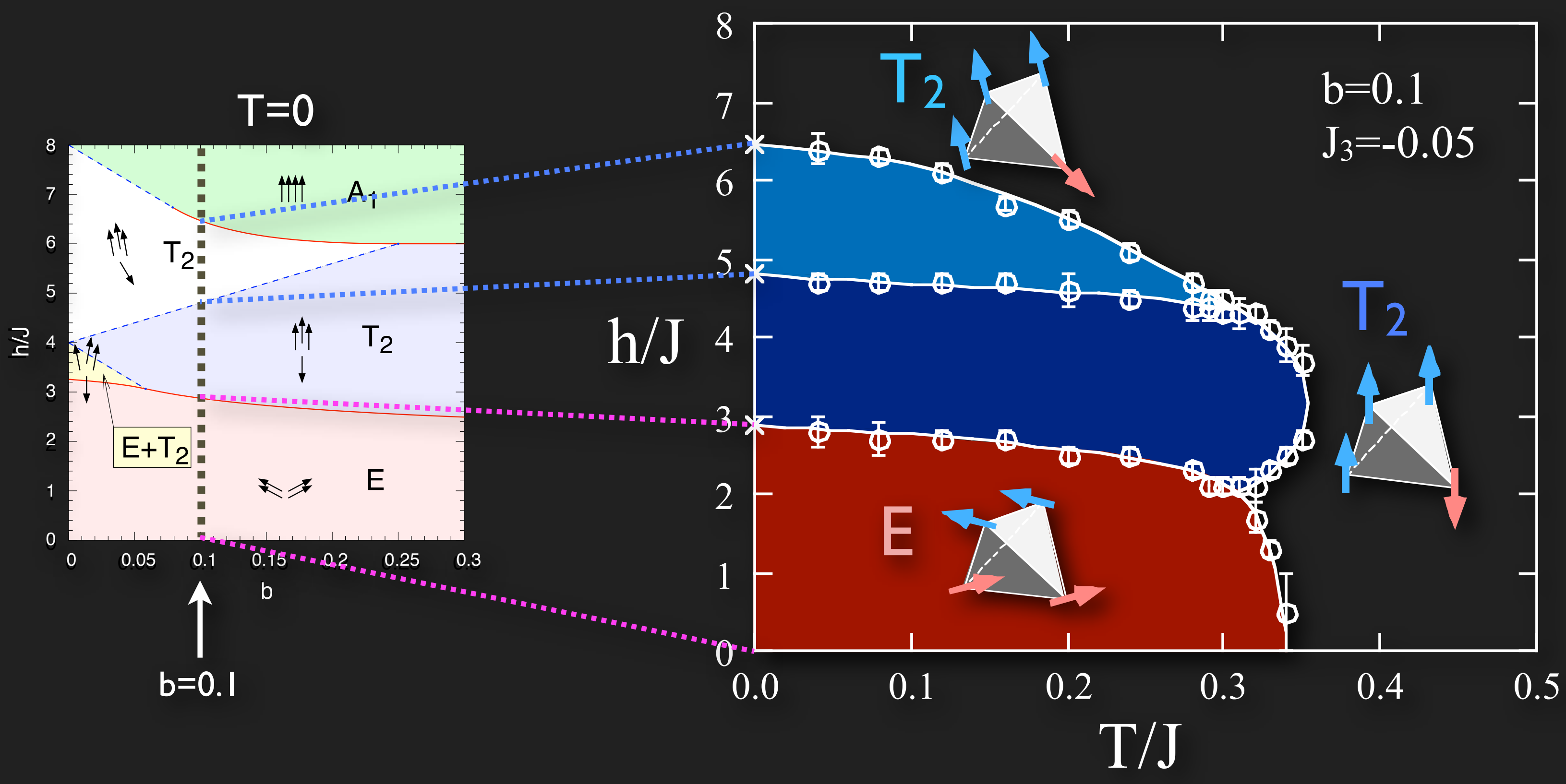


$$\phi \qquad \phi + \pi$$

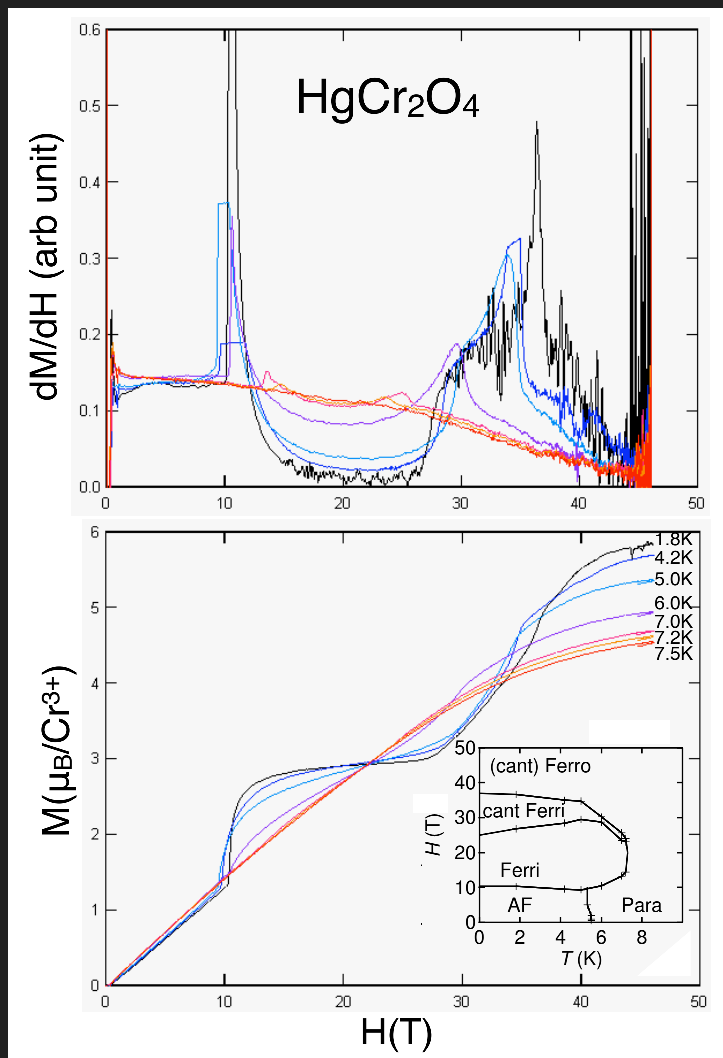
$$\begin{aligned} \exp 2i\phi &= \cos 2\phi + i \sin 2\phi \\ &= \cos^2\phi - \sin^2\phi + i\, 2\sin\phi\cos\phi \\ &= S_x^2 - S_y^2 + i\, 2 S_x S_y \\ &= Q_{x^2-y^2} + i\, Q_{xy} \end{aligned}$$



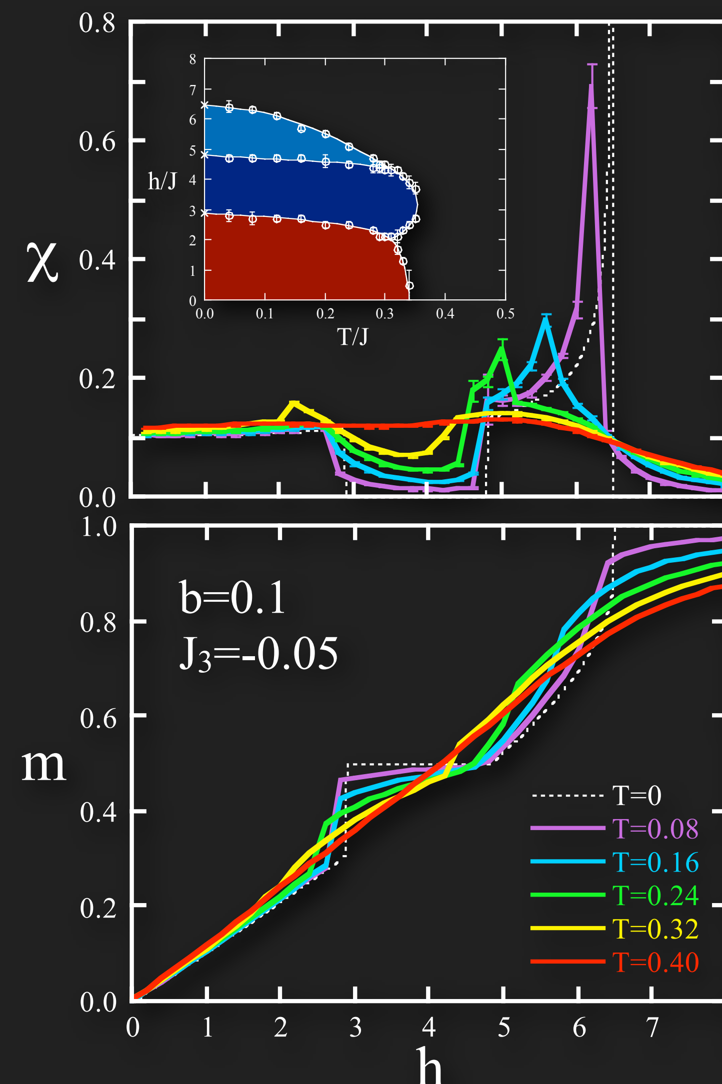
Phase Diagram at Finite T



Comparison with Experiments

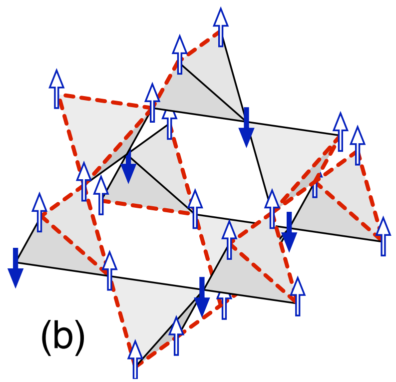


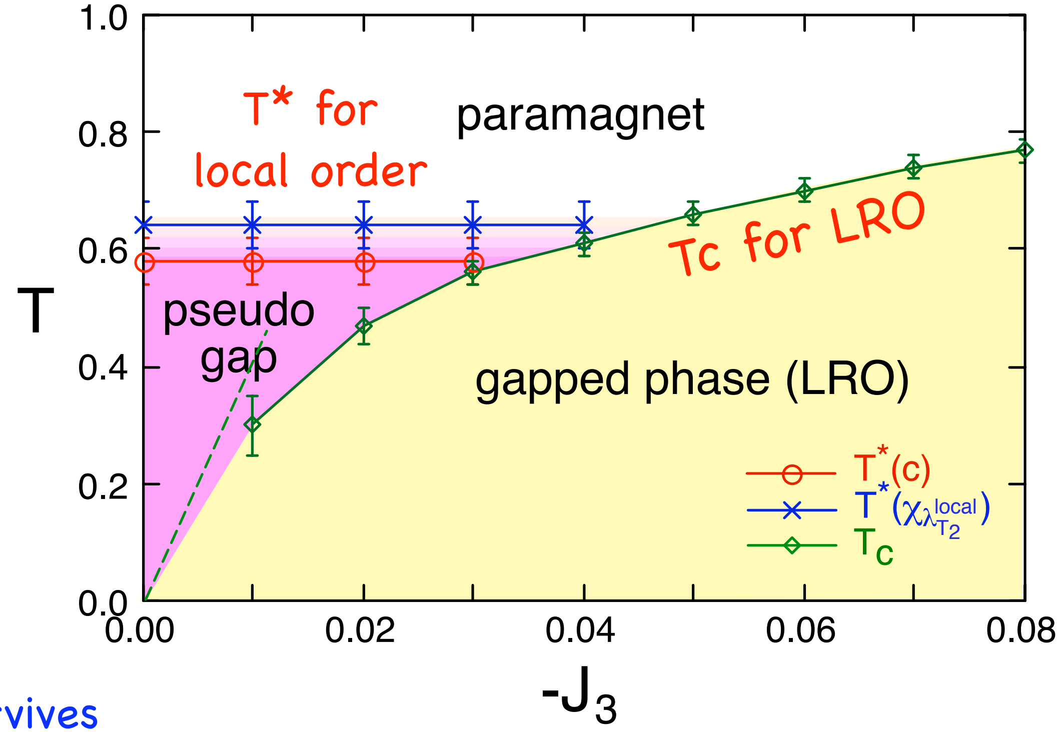
H. Ueda *et al.*, unpublished



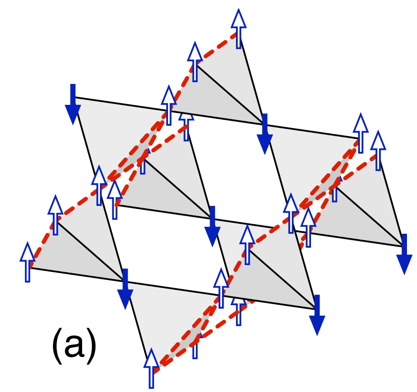
Liquid plateau

"plateau liquid"



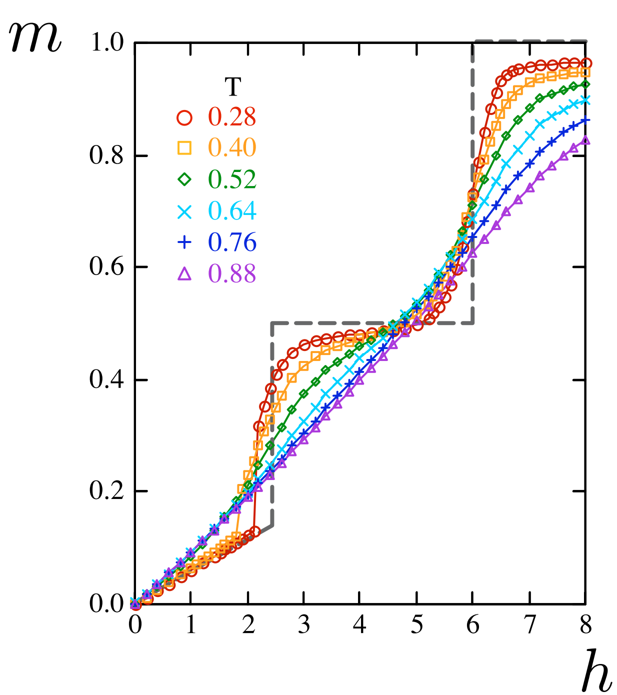


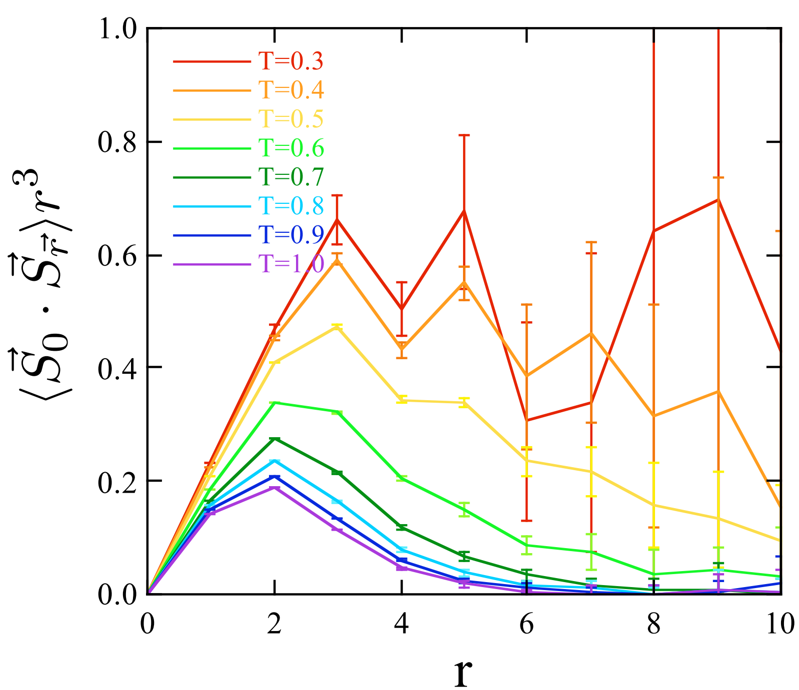
b=0.6



"plateau solid"

magnetization plateau survives





Coulomb phase in the disordered plateau ?

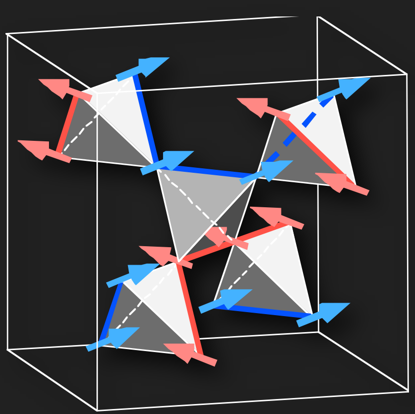
Phase Diagram for J_3=0

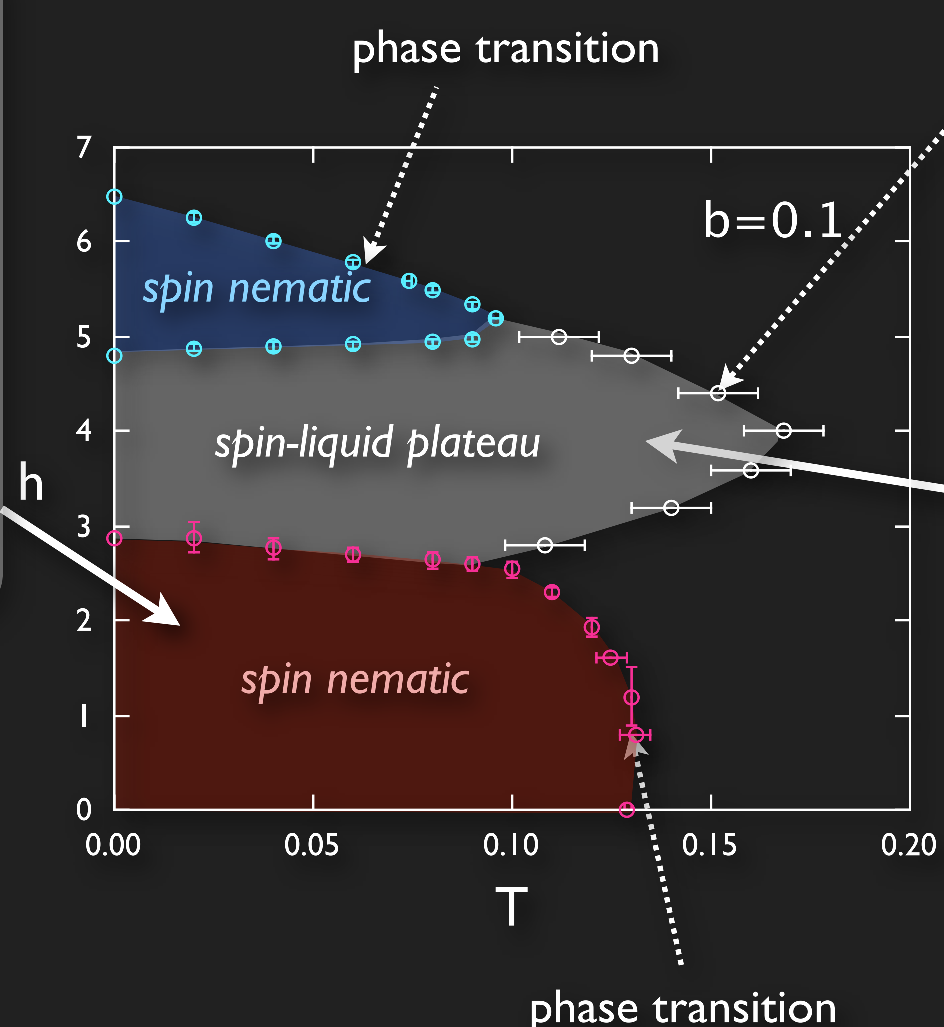
2:2 E-symmetry in each tetrahedron

macroscopic degeneracy

spin nematic order

power-law decay of spin correlation



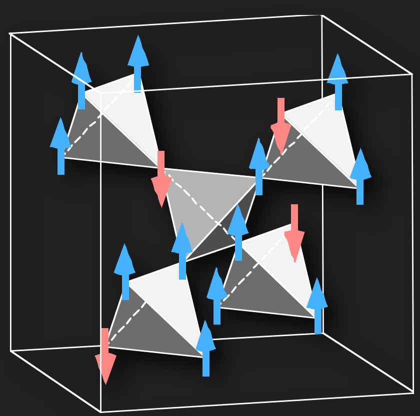


3up-1down T_2 symmetry in each tetrahedron

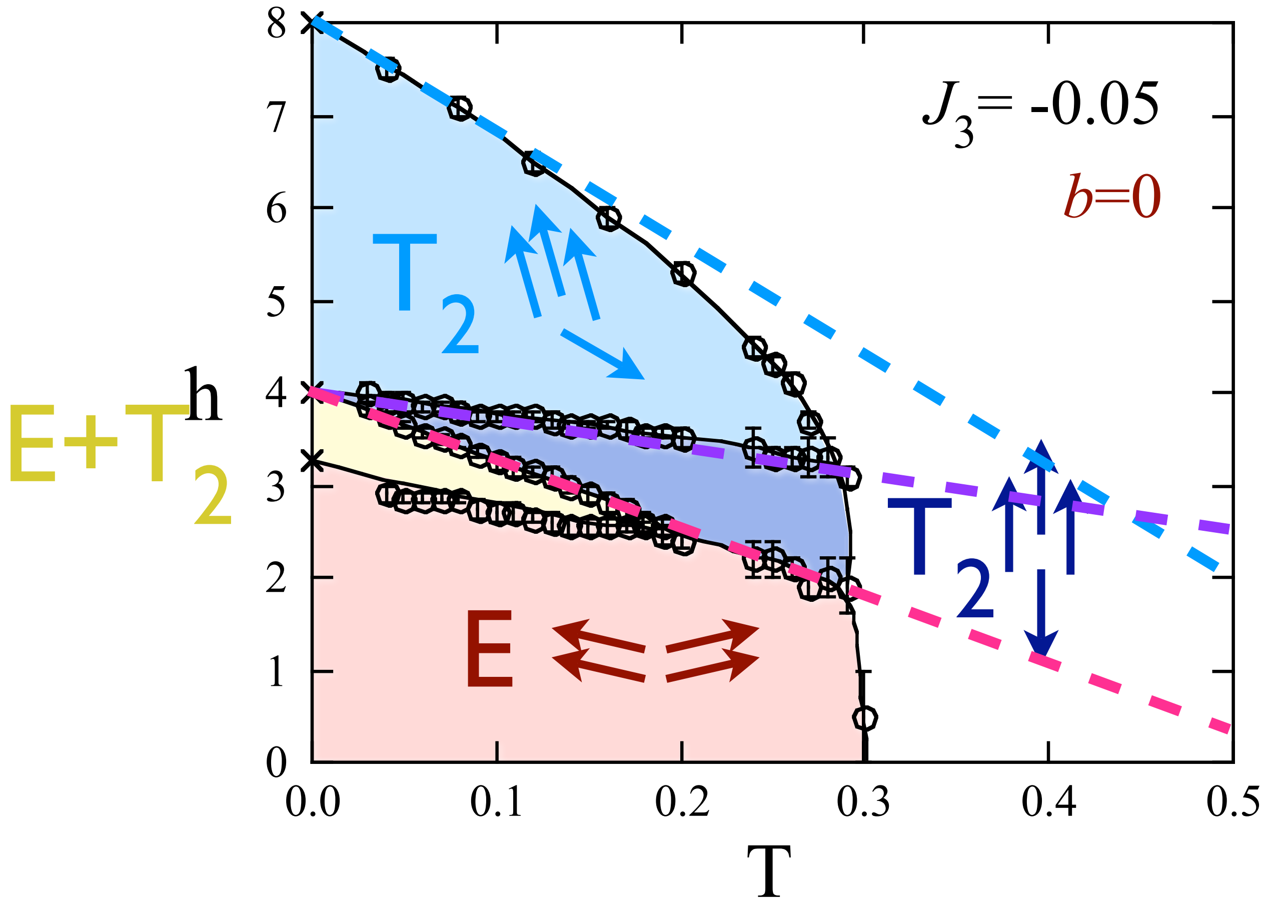
macroscopic degeneracy

robust plateau

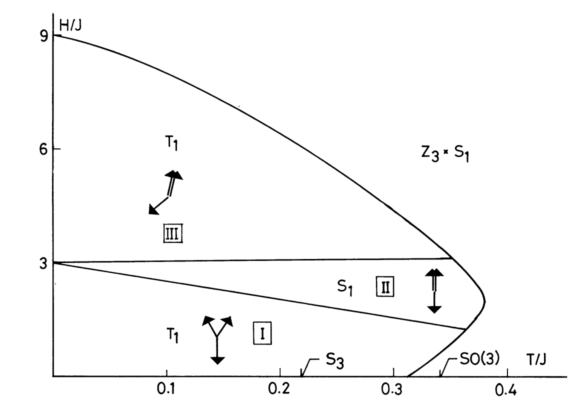
spin pseudo-gap



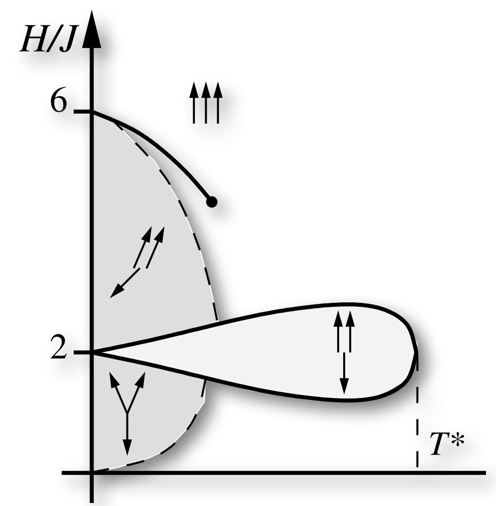
Phase diagram for b = 0 : order by disorder revisited



$$\Delta\mathcal{F} \sim -\frac{T}{J_3}(\mathbf{S}_i\mathbf{S}_j)^2$$



H. Kawamura and S. Miyashita, 1985



M. Zhitomirsky, 2002

Conclusions

- Coupling to lattice distortions provides a very efficient mechanism for magnetization plateaux in frustrated and degenerate AF's.
- The phase diagram and order parameters determined.
- Plateau can survive without long range order
- At finite T : order-by-disorder possible with some help