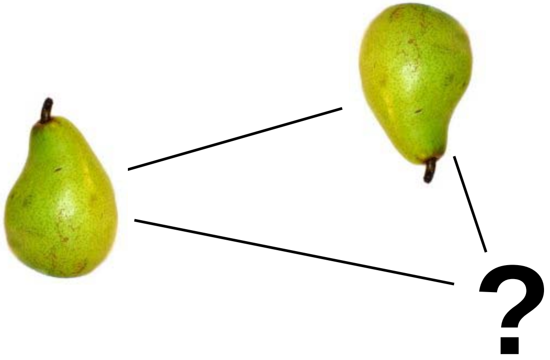
Frustrated pairs: magnon BEC in geometrically frustrated spin dimer compounds



I. R. Fisher
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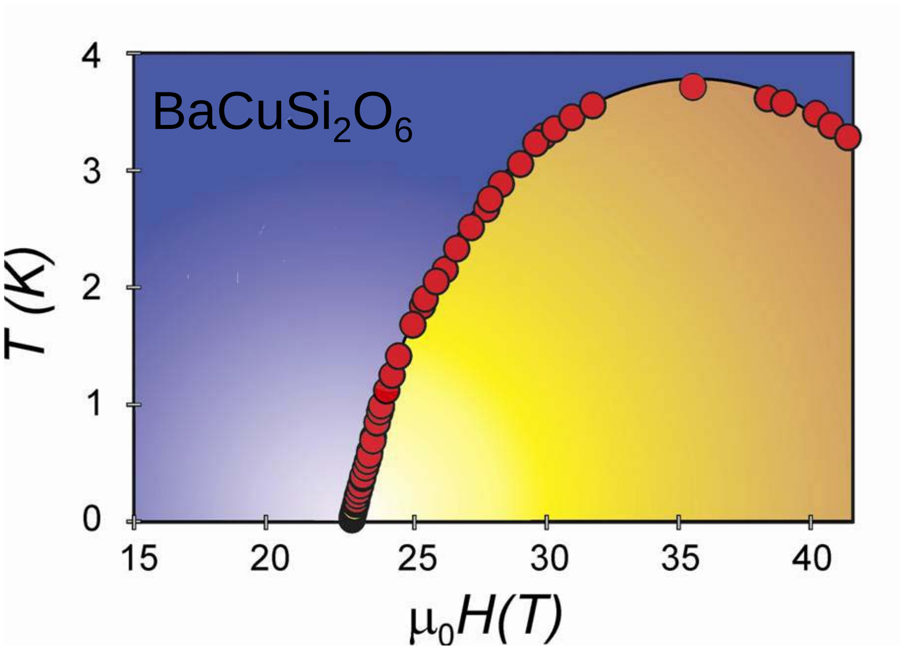

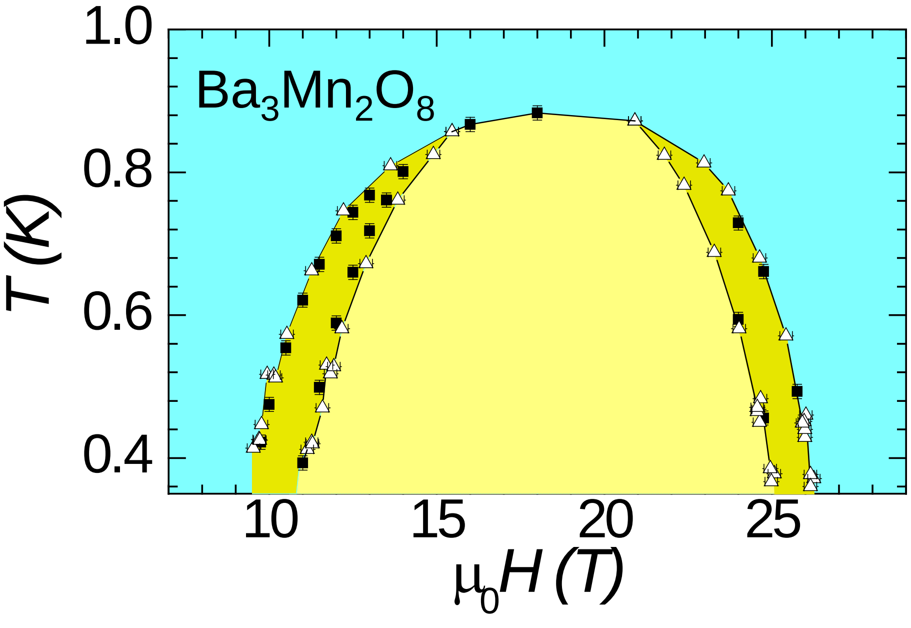

University of Toronto (2008)

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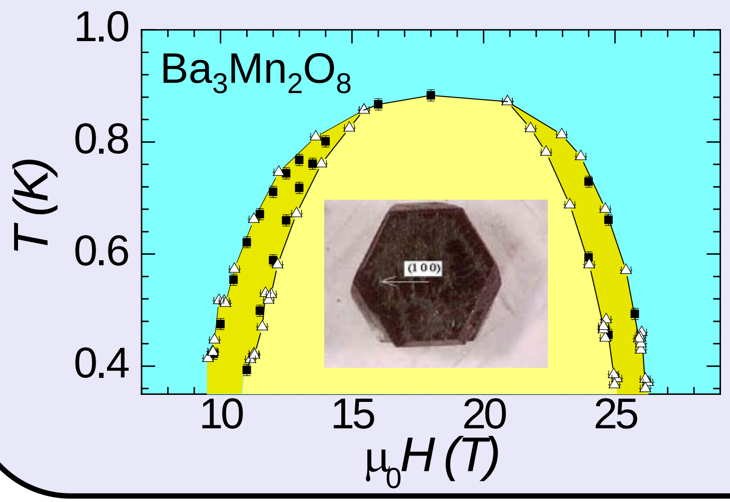
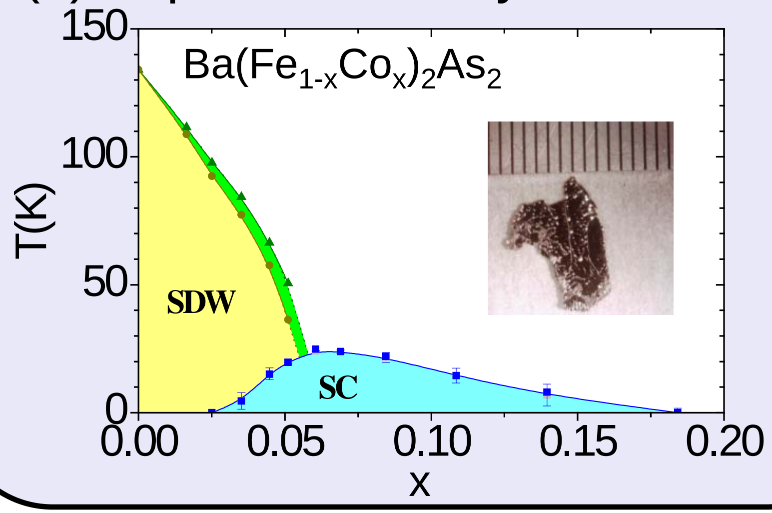
Research program:

New materials – unconventional magnetic & electronic ground states & phase transitions

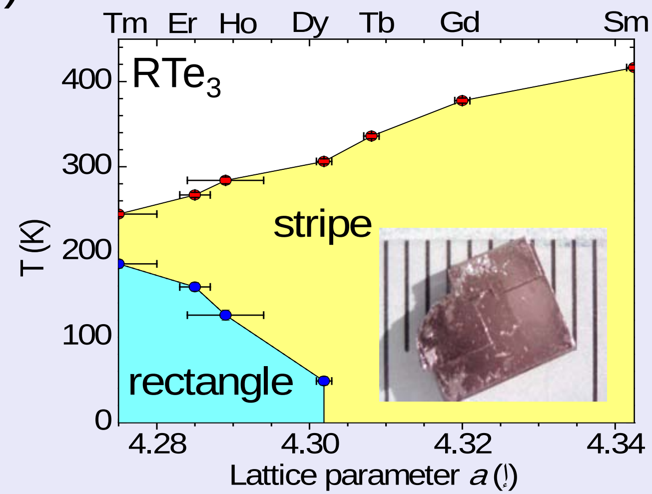
(1) Quantum magnetism:



(2) Superconductivity:



(3) Low dimensional materials:



http://www.stanford.edu/group/Fisher

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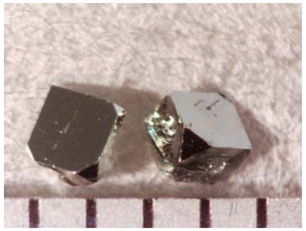
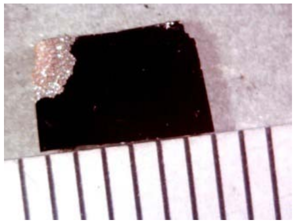
Crystal growth

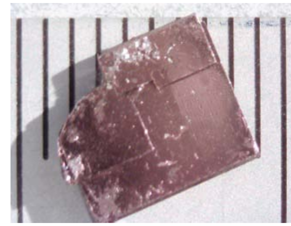


$Pb_{1-x}Tl_xTe$

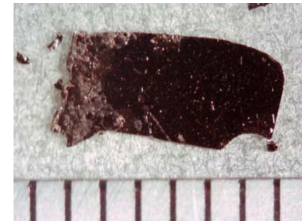


$BaPb_{1-x}Bi_xO_3$

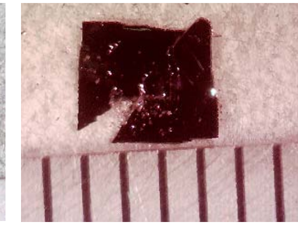
$BaFe_2As_2$



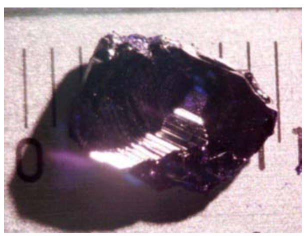
RTe_3



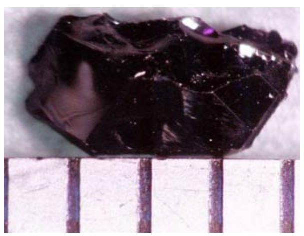
RTe_2



R_2Te_5

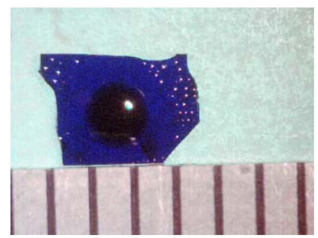


$BaCuSi_2O_6$

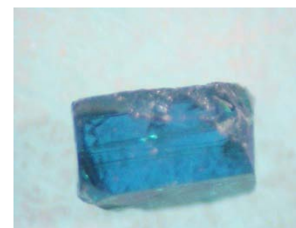


$Sr_2Cu(BO_3)_2$

$Ba_2Cu(BO_3)_2$

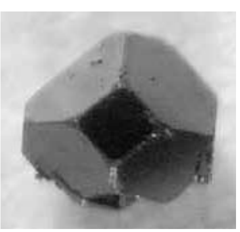


$BaCu_2Si_2O_7$

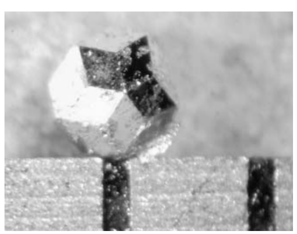


$BaCuB_2O_5$

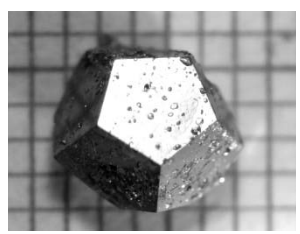
$Ba_3Mn_2O_8$



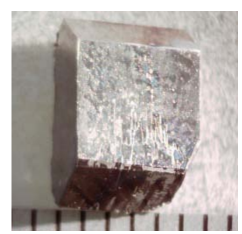
Ba_2OsNaO_6



R-Mg-Cd



Al-Pd-Re



$R_6Mo_4Al_{43}$

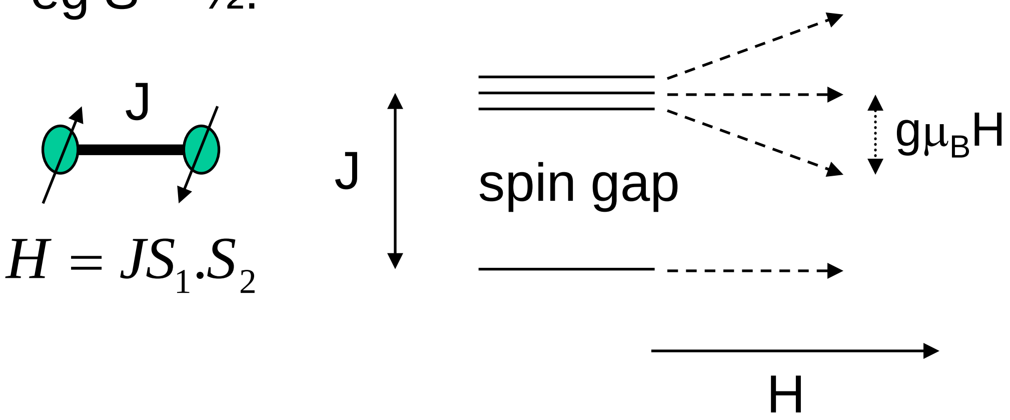
Outline

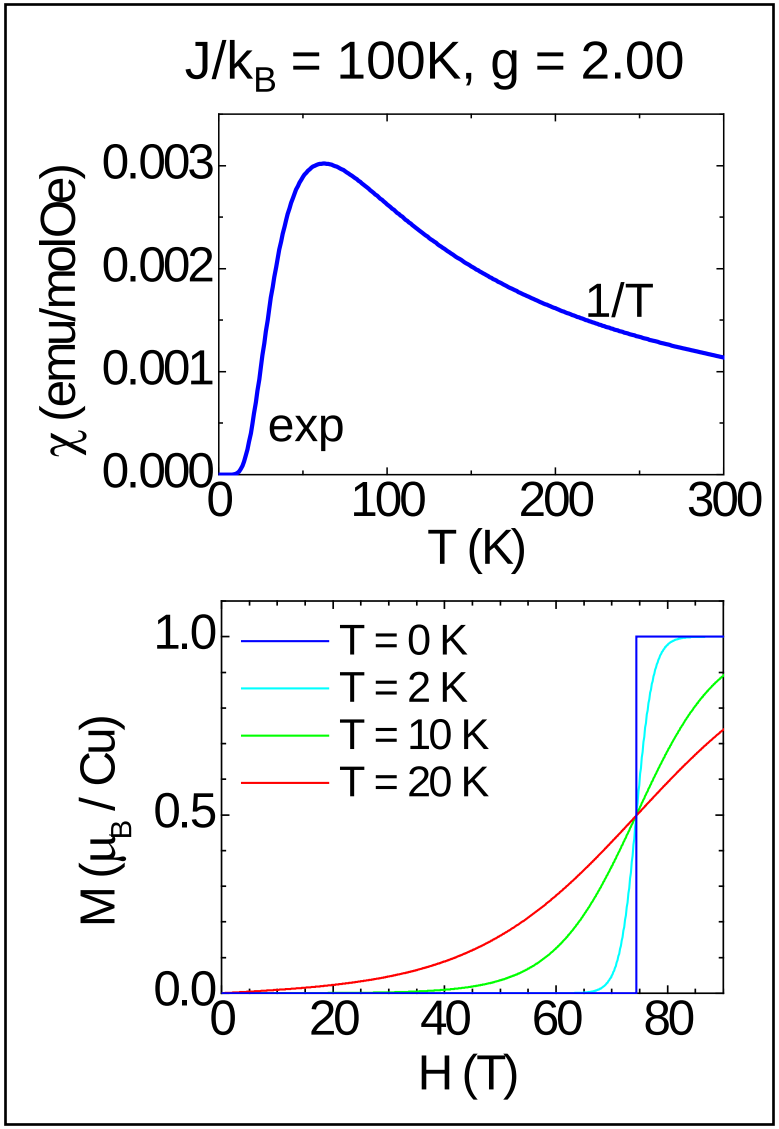
(1) A short introduction to spin dimer compounds:
- what they are, and why we should care

(2) "Frustrated pairs": two neat examples for which frustration plays a crucial role…
- bct lattice ($BaCuSi_2O_6$)
- triangular lattice ($Ba_3Mn_2O_8$)

Properties of an isolated pair

eg S = ½:





Low field properties:

- thermodynamic distribution
- calculate C_p, M, χ…

$$\chi = \frac{N(g\mu_B)^2}{k_B T\left(3 + \exp(J/T)\right)}$$

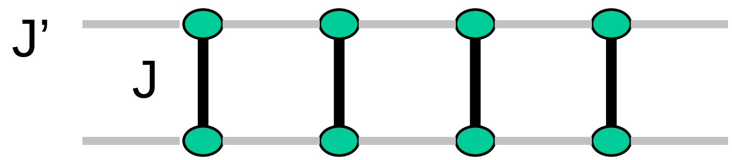
High field properties:

- $g\mu_B H_c = J$
- thermal broadening at finite T

Question:

- what happens for weakly interacting spin dimers?

Insight from the two leg ladder



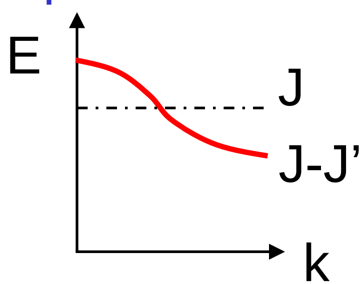
$$H = J\sum_{i=1}^{N} S_{1,i}.S_{2,i} + J'\sum_{\alpha=1}^{2}\sum_{i=1}^{N} S_{\alpha,i}.S_{\alpha,i+1} - h\sum_{\alpha=1}^{2}\sum_{i=1}^{N} S^{z}_{\alpha,i}.$$

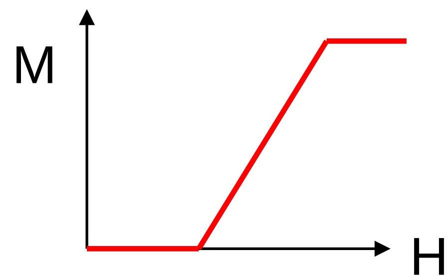
- for J >J', treat perturbatively

$$S \equiv \left|\tilde{\downarrow}\right\rangle \qquad T_{-} \equiv \left|\tilde{\uparrow}\right\rangle$$

$$H_{eff} = J'\sum_{i}\left[\tilde{S}^{x}_{i}\tilde{S}^{x}_{i+1} + \tilde{S}^{y}_{i}\tilde{S}^{y}_{i+1} + \tfrac{1}{2}\tilde{S}^{z}_{i}\tilde{S}^{z}_{i+1}\right] - \tilde{h}_{eff}\sum_{i}\tilde{S}^{z}_{i} + C$$

- delocalized triplet excitations (triplon)





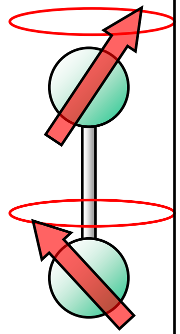
$$H_{eff} \equiv t\sum_{i,\alpha}\left(b^{\dagger}_{i+\hat{e}_\alpha} b_i + h.c.\right) + V\sum_{i,\alpha} n_i n_{i+\hat{e}_\alpha} + \mu\sum_{i} n_i$$ where $t = V = \frac{J'}{2}$ & $\mu = h - J$

- i.e. realization of a lattice gas of hard core bosons, for which B controls μ

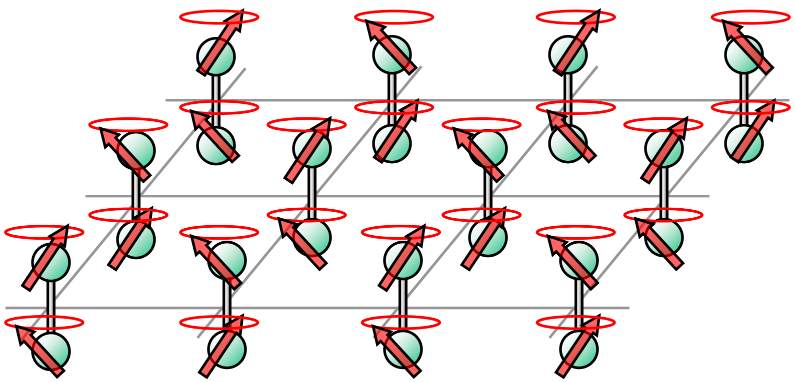
F. Mila, Euro Phys. J. B. **6**, 201 (1998).
T. Giamarchi & A. M. Tsvelik, PRB **59**, 11398 (1999).
S. Sachdev in "Quantum Magnetism" (2004).
T. Matsubara & H. Matsuda, Prog. Theor. Phys. **16**, 569 (1956).

Ground states: KE vs. PE

- Ordered state is a product of individual dimer terms of form:

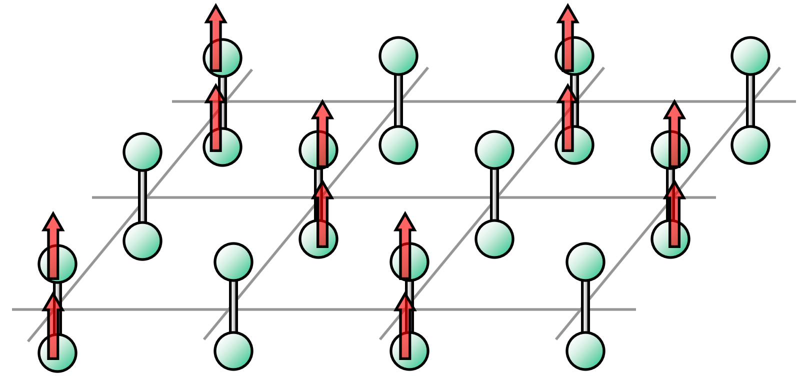


$$\left|\psi_i\right\rangle = \cos\theta_i\left|s\right\rangle + \sin\theta_i e^{i\phi_i}\left|t\right\rangle$$

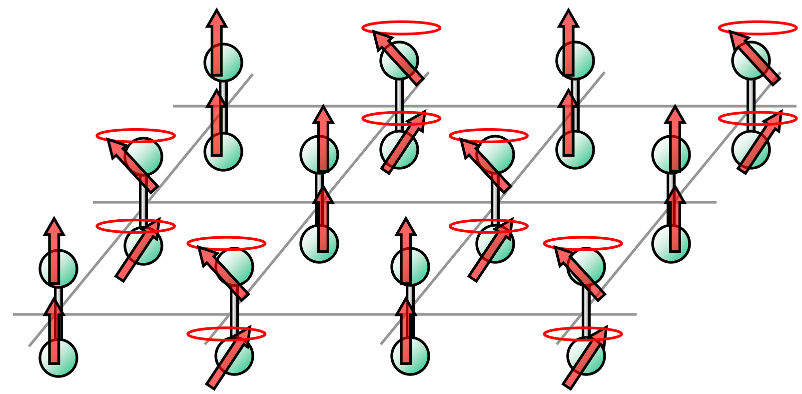


- macroscopic occupancy of minimum of triplet band $(\pi/a, \pi/a)$
- triplet density $\rho = \langle s_z \rangle = m_z$
- amplitude of order parameter = m_{xy}; phase = angle of spins in plane
- spontaneously broken U(1) symmetry

eg $TlCuCl_3$



eg $SrCu_2(BO_3)_2$

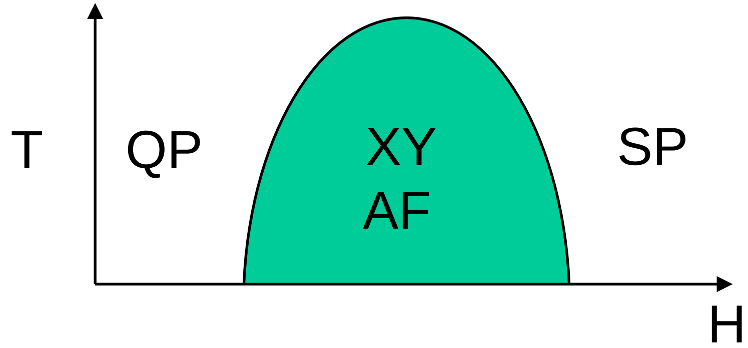


no candidate to date

So why study spin dimer compounds?

(1) Can access lattice gas models which would be unphysical for simple AFs:
i.e. in the limit of weak spin orbit coupling, can "engineer" Hamiltonian with highly anisotropic effective exchange…
… and potentially access to some rather unusual quantum phases of matter

(2) Tunable by magnetic field, so can explore entire quantum phase diagram:

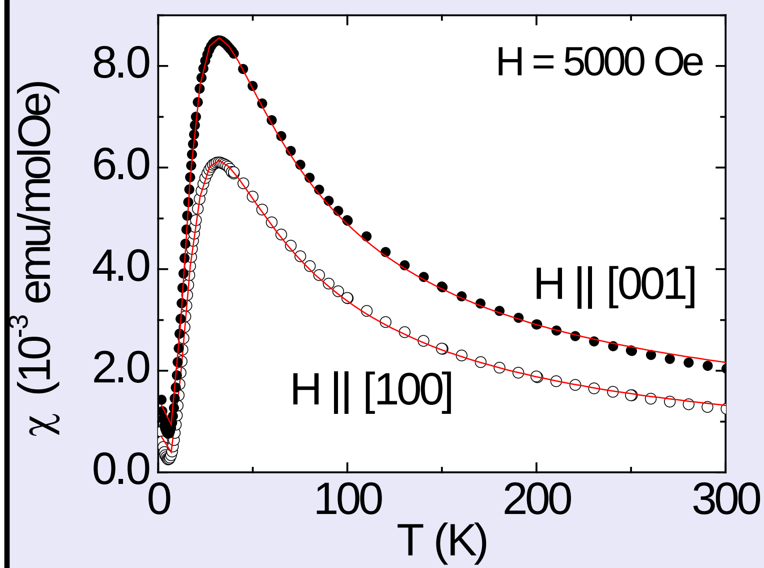


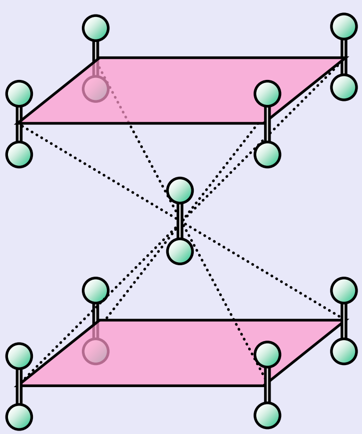
(3) "Protected" against symmetric anisotropies:
terms like DS_z^2 in effective Hamiltonian can only mix singlet and triplet states in second order → terms which break axial symmetry are suppressed relative to a simple AF

(4) Relatively few candidate materials…

This talk: magnon BEC in two neat compounds with geometrically frustrated lattices

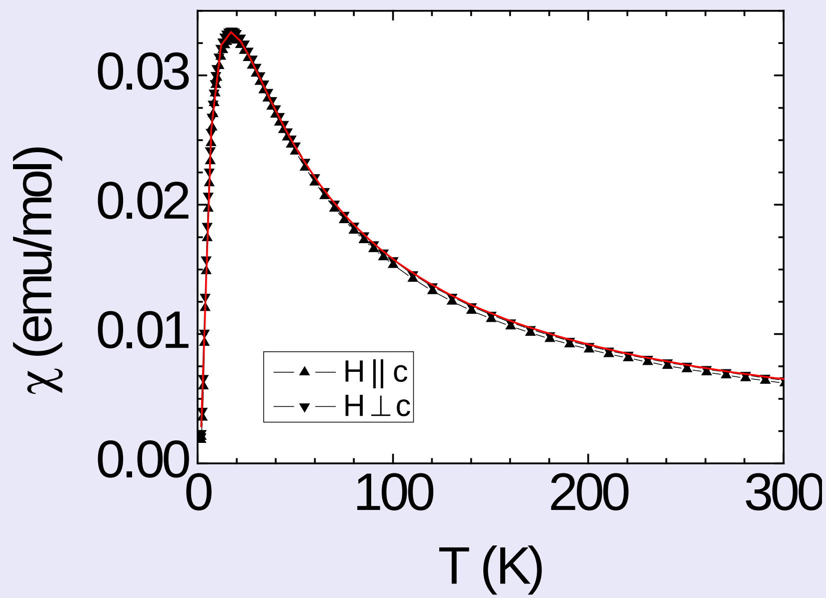
(1) $BaCuSi_2O_6$

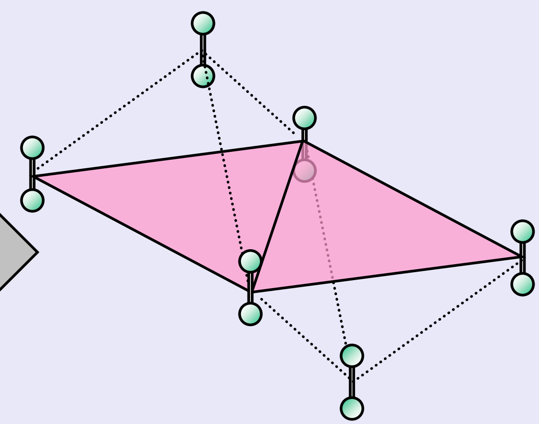




more frustrated **?**

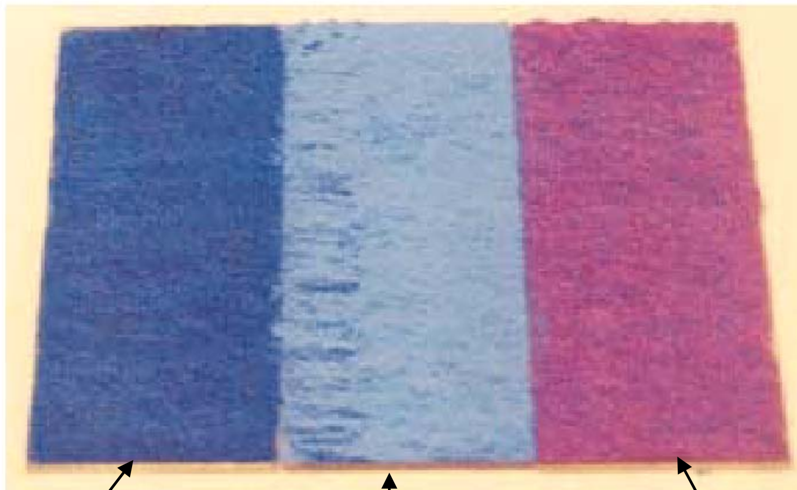
(2) $Ba_3Mn_2O_8$





New materials?

Anthropogenic pigments:



Egyptian blue
$CaCuSi_4O_{10}$

Chinese blue
$BaCuSi_4O_{10}$

Han purple
$BaCuSi_2O_6$

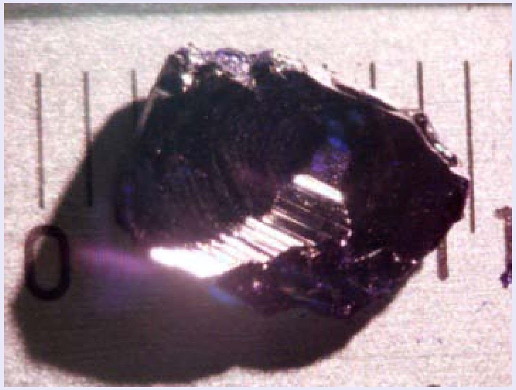
“Chemistry in ancient times:
the development of blue and purple pigments”
H. Berke, Angew. Chem. Int. Ed. **41**, 2483 (2002).

Crystal growth

(1) $BaCuSi_2O_6$

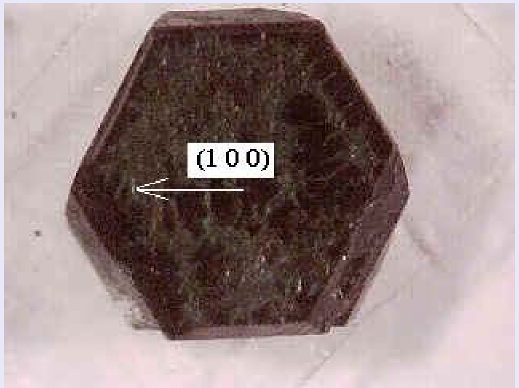
(a) Evidence for Pb in archeological samples
- PbO flux works

(b) but $LiBO_2$ better

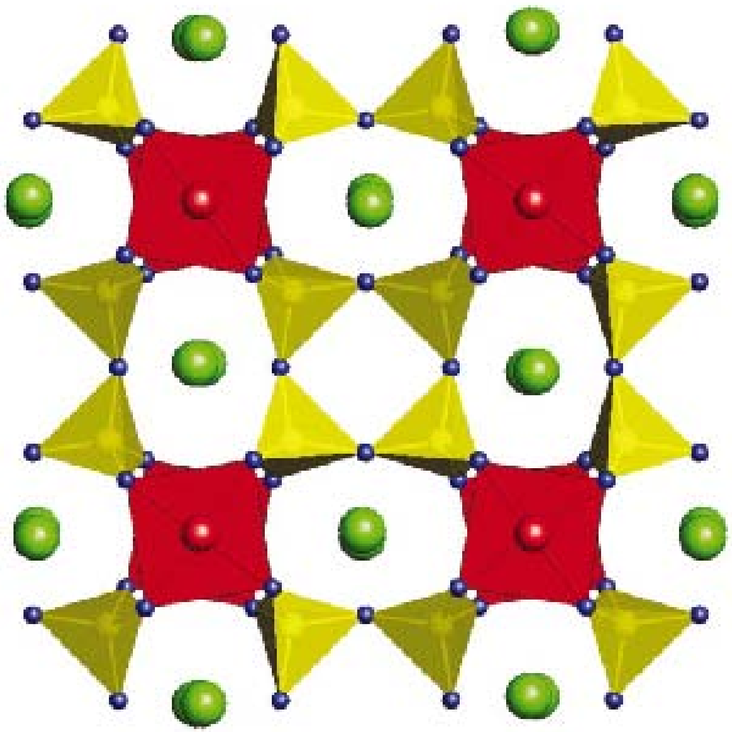


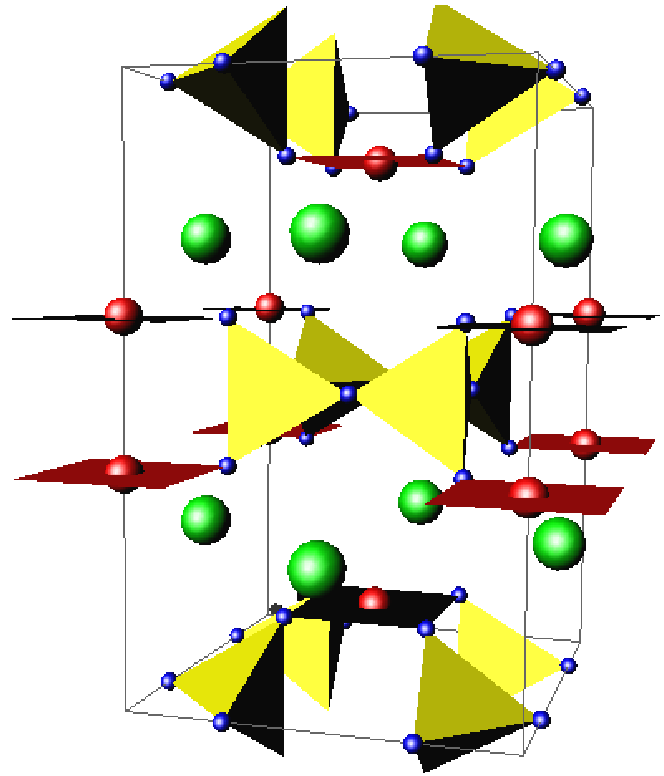
(2) $Ba_3Mn_2O_8$

Mn^{5+} valence unusual
Requires strongly oxidizing flux
NaOH works very well



Case 1: *bct* lattice - $BaCuSi_2O_6$





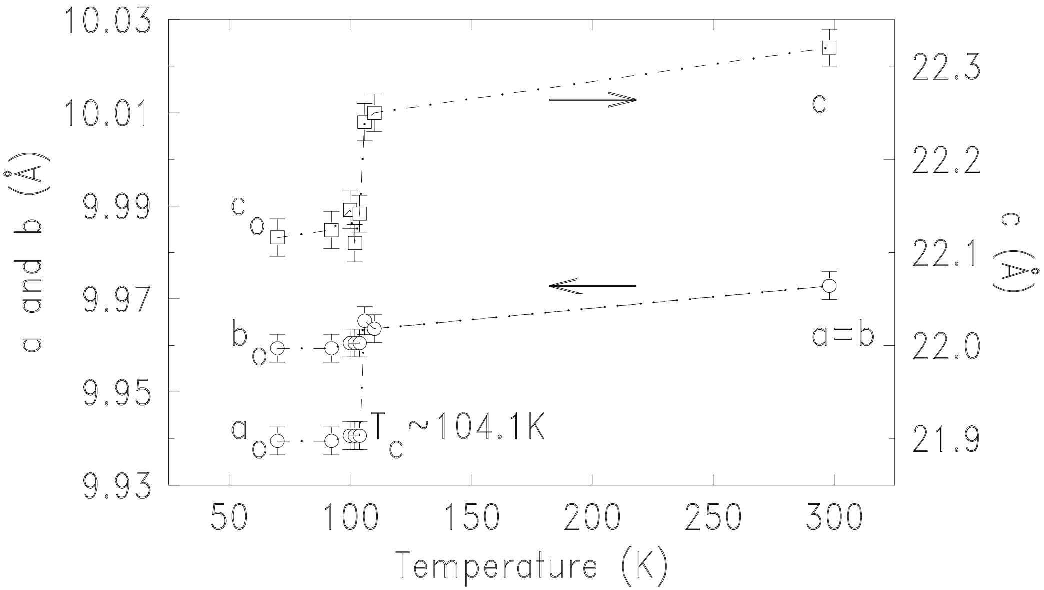
Sparta & Roth, Acta Cryst. **B60**, 491 (2004).

Inelastic neutron scattering (Ch. Ruegg):

- $J \sim 4.5$ meV; $J' \sim 0.5$ meV
- c-axis bandwidth finite but small (< 0.05 meV)
- spin gap $\Delta = 3.1$ meV → $H_{c1} = \Delta/g\mu_B \sim 23.5$ T for H || [001]

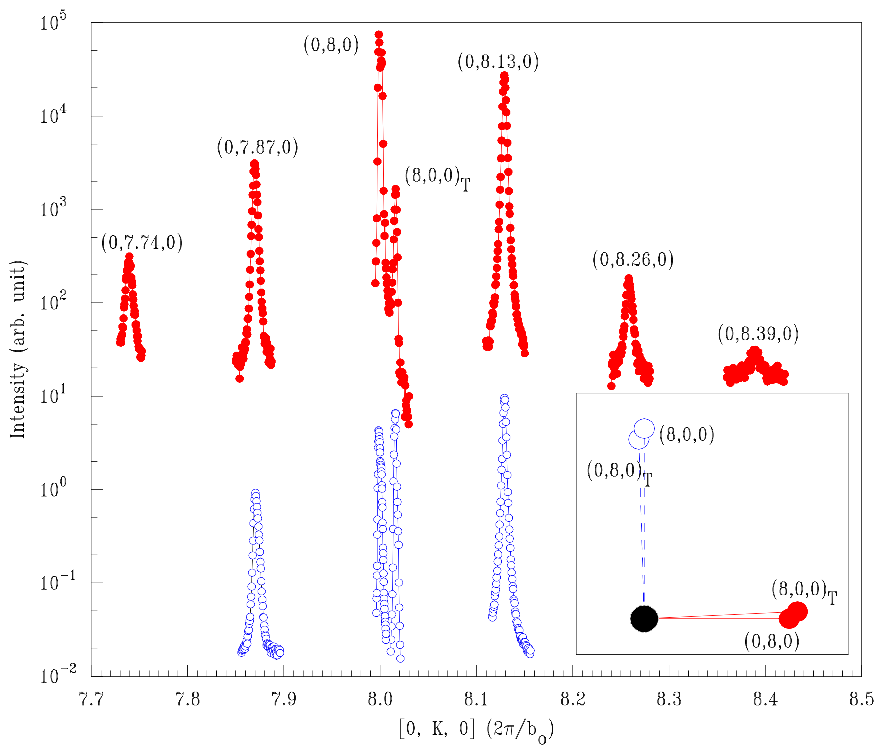
Ch. Ruegg *et al.*, PRL **98**, 017202 (2007).

Structural phase transition (with Z. Islam, APS)



- weak orthorhombic distortion at ~ 100 K

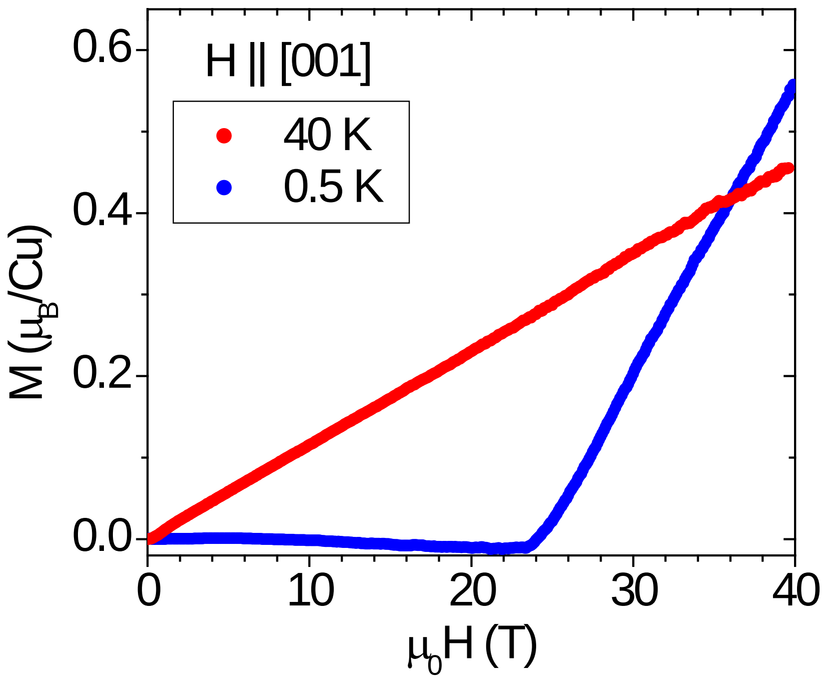
$$\frac{b-a}{\frac{1}{2}(b+a)}=0.2\,\%$$

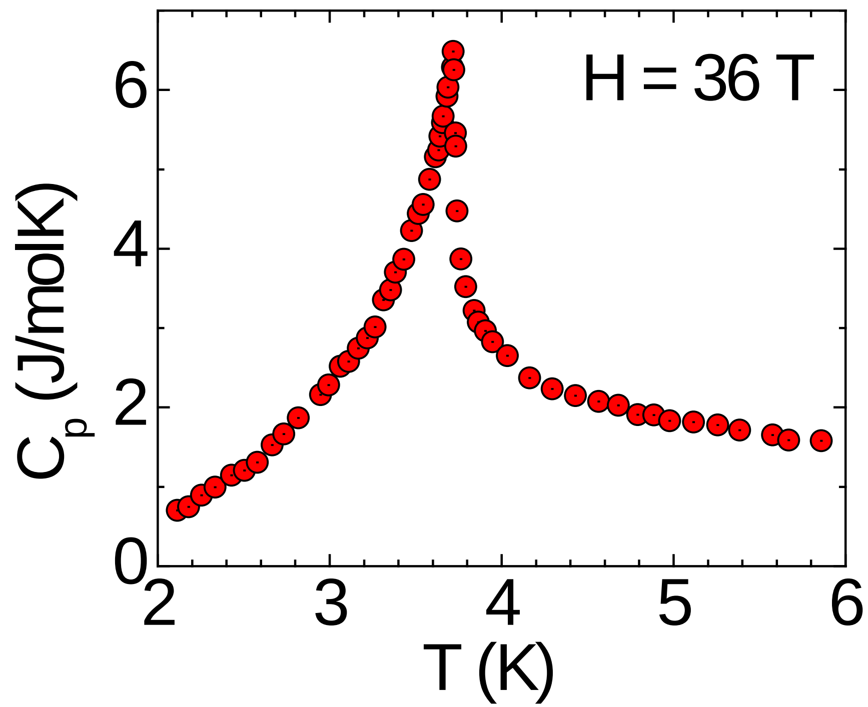


- incommensurate lattice modulation
- q_{IC} = 0.129 b*
- presumably driven by rigid rotation of SiO_4 tetrahedra
- lack a complete structural model
- subtle effect, and doesn't affect subsequent analysis

E. Samulon, Z. Islam *et al.*, PRB **73**, 100407(R) (2006).

High field behavior (with M. Jaime & N. Harrison, NHMFL)





- does this correspond to BEC of triplons as conceived theoretically?
- address by looking at critical scaling (characteristic of universality class)
- thermal phase transition: $C_p \sim (T-T_c)^{-\alpha}$
- quantum phase transition: $T_c \propto (H - H_{c1})^{\nu}$
- BEC universality class: $\nu = 2/d$
- 3D Ising: $\nu = 1/2$

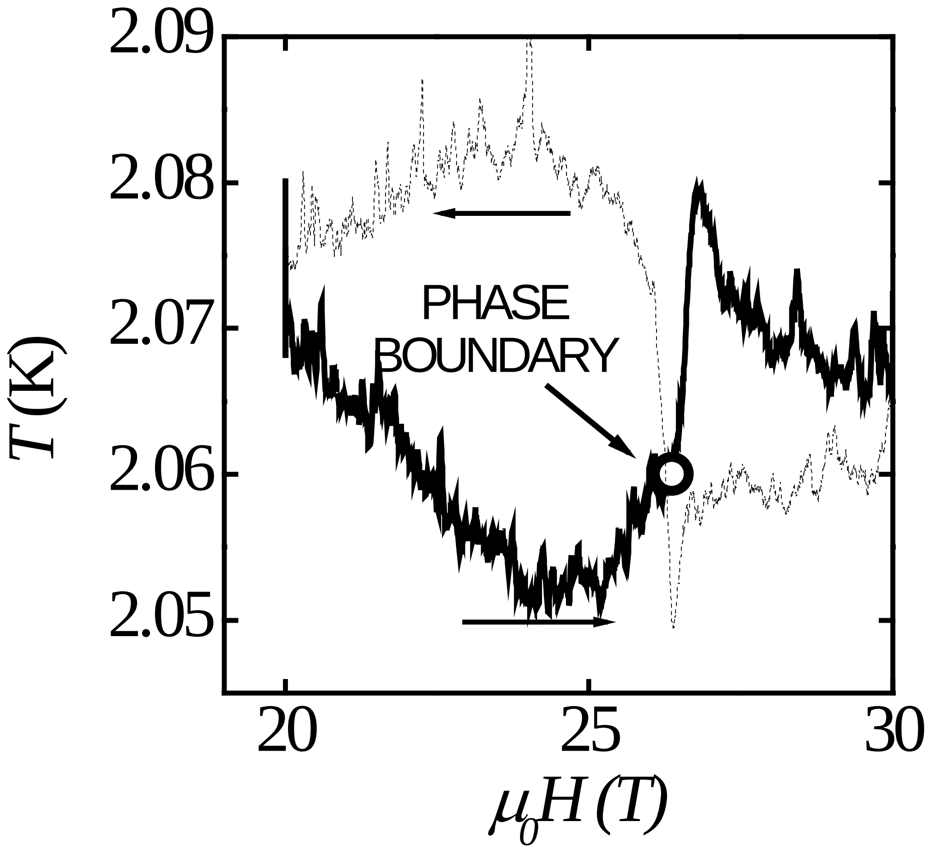
Experimental determination of $T_c(H)$ close to the QCP

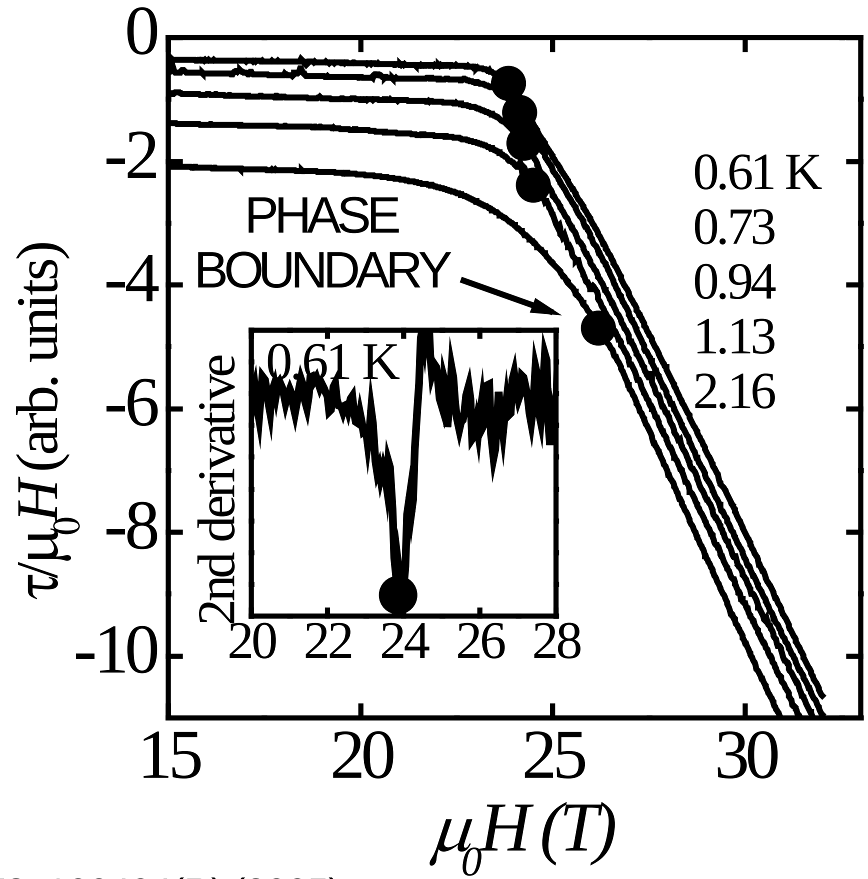
(a) Magneto-caloric effect: (with Marcelo Jaime, NHMFL)

- jump in T of sample on entering/leaving ordered state

(b) Cantilever torque magnetometry: (with N. Harrison, L. Balicas, NHMFL)

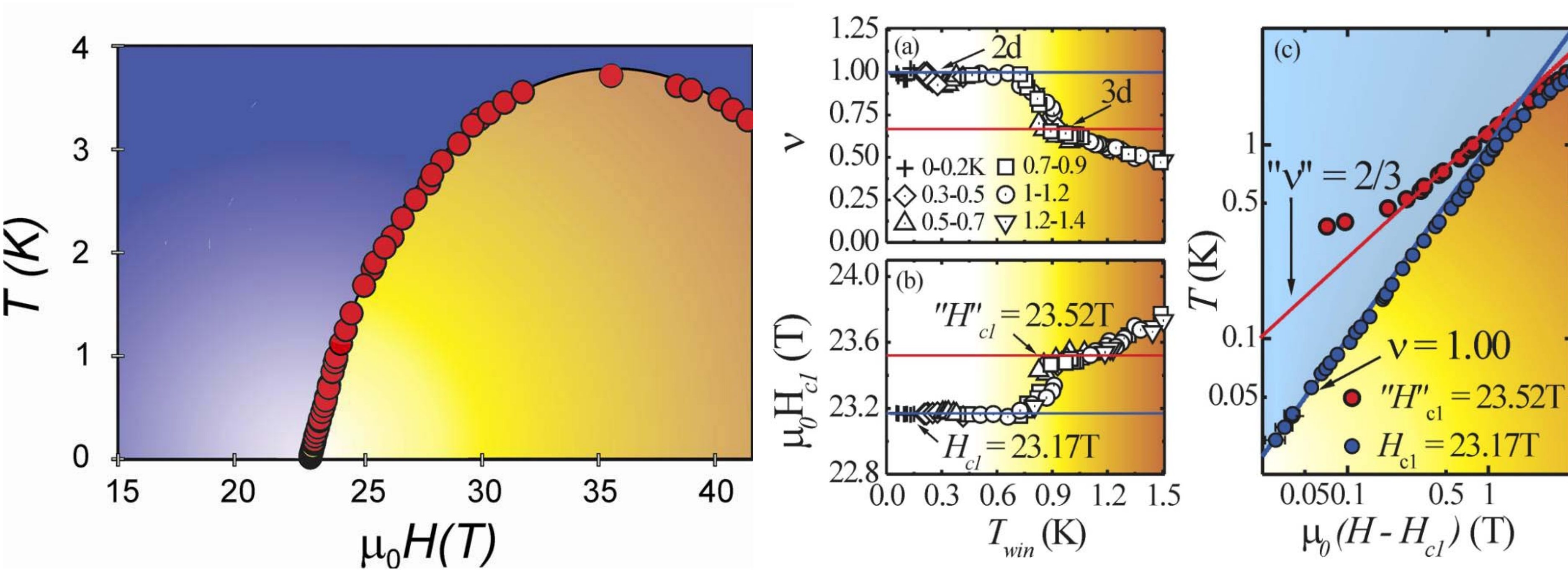
- divergence in 2nd derivative of magnetization





S. Sebastian *et al.*, Phys. Rev. B. **72**, 100404(R) (2005).

Phase diagram & critical scaling analysis



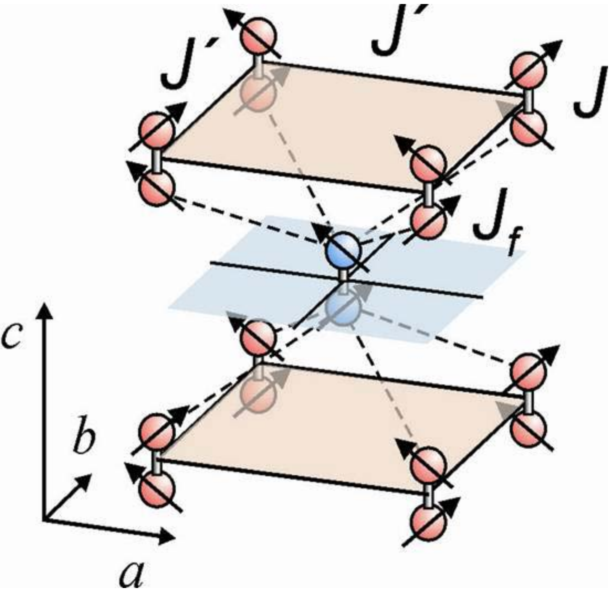
- Two parameter fit to $T_c \propto (H - H_{c1})^{\nu}$ with a sliding window

- BEC scaling exponents observed ($\nu = 2/d$)
- Surprise - cross-over to 2D exponent approaching the QCP!

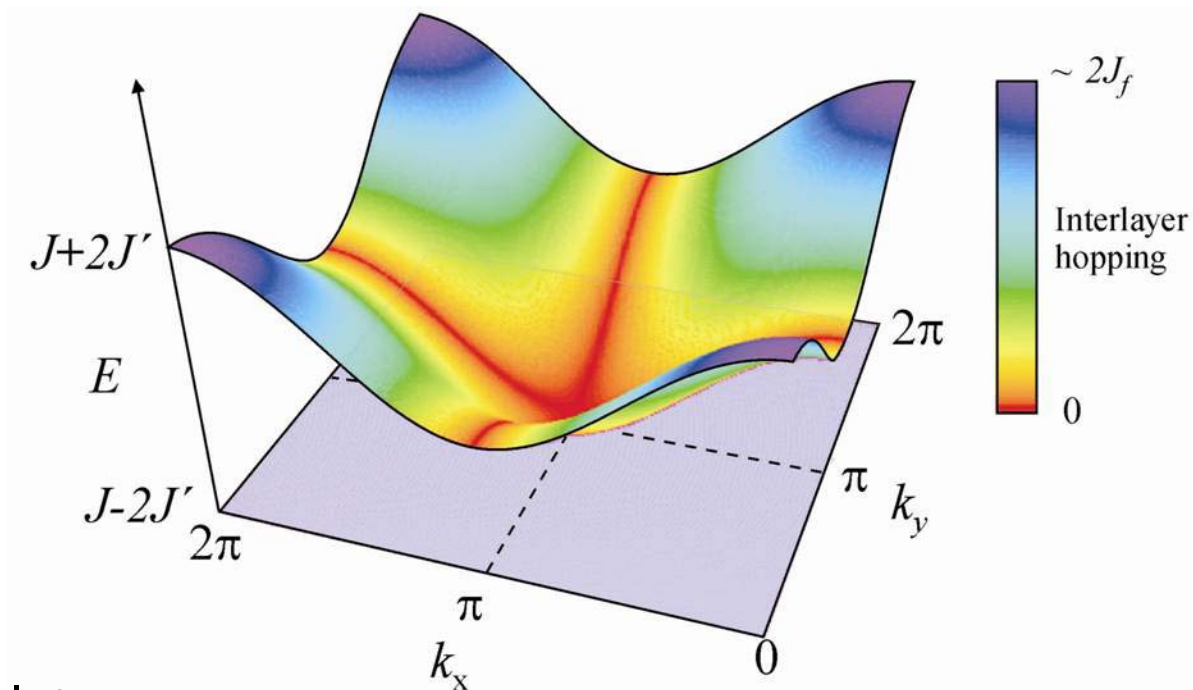
Question: Individual triplets can move in 3D, so why is the collective behavior at the QCP in just 2D?

S. Sebastian, Harrison, Jaime, Balicas, Batista, IRF, *et al*., Nature **441**, 617 (2006).

Dispersion relation for perfect *bct* lattice



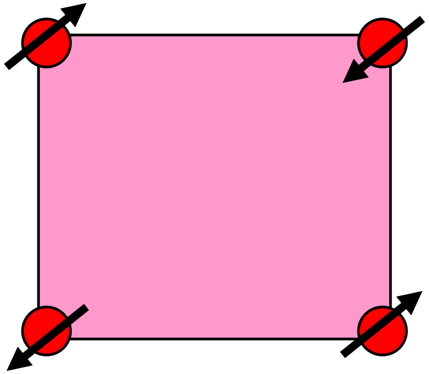
$$E = J'(\cos k_x + \cos k_y) + 2J_f \cos\frac{k_x}{2}\cos\frac{k_y}{2}\cos k_z$$



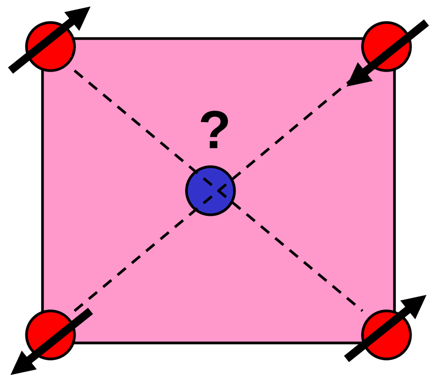
Case of non-interacting triplets:

- At T = 0 all particles will be in the condensate at k = (π,π)
- But there is no interlayer hopping for k = (π,π) !
- → independent 2d condensates

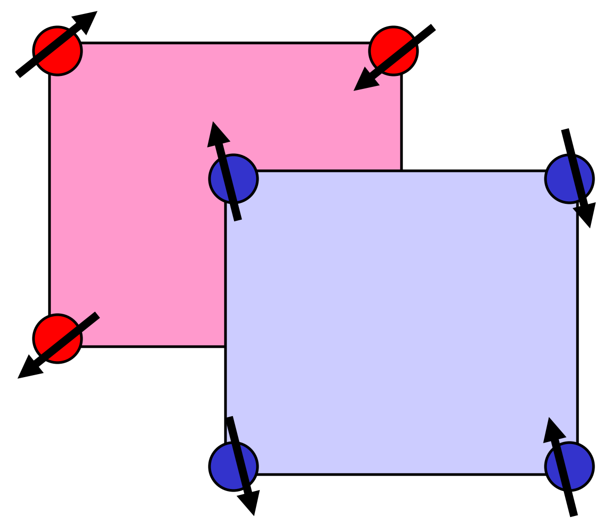
Origin of vanishing dispersion: geometric frustration



- in-plane ordering wave-vector = (π,π)



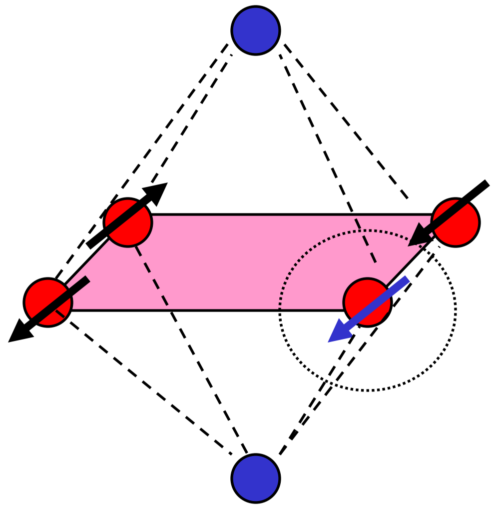
- body centered tetragonal lattice
- geometric frustration
- adjacent planes decoupled!



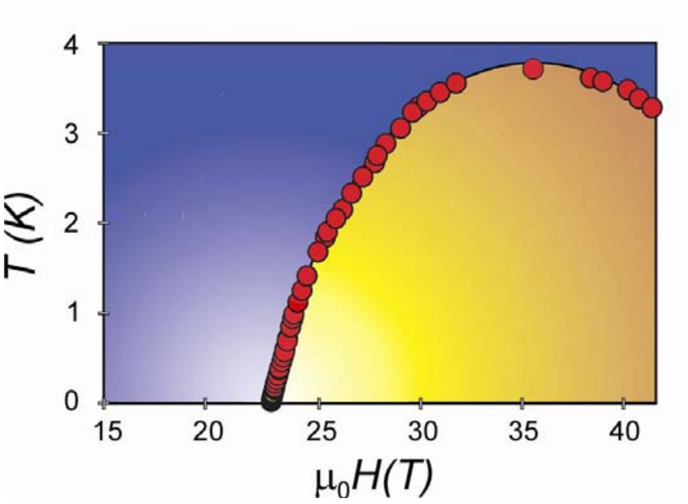
Order from disorder?

Theory: C. Batista, LANL
& J. Schmalian, Ames

- Must consider the effect of phase fluctuations on this delicate frustration…



- leads to an effective unfrustrated biquadratic interlayer hopping / coupling $K \sim \rho^2$
- restores 3D phase coherence for finite triplet concentrations ρ



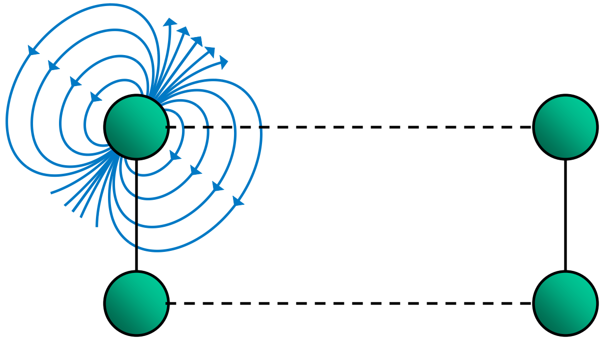
- but at the QCP, $\rho \rightarrow 0$ so $K \rightarrow 0$
- 2D fixed point determines universal scaling

Maltseva & Coleman, Phys. Rev. B **72**, 174415 (2005).
Batista, Schmalian *et al.*, PRL **98**, 257201 (2007).

U(1) symmetry?

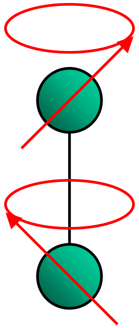
- BEC implies a spontaneously broken axial symmetry
- What effects might break this apparently delicate symmetry?

- For $BaCuSi_2O_6$, it turns out that the largest effect is from dipolar interactions…

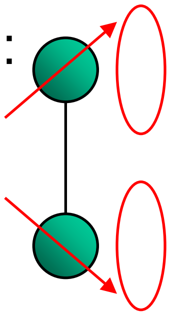


- Dipolar energy ~ $1/r^3$, so consider first the intradimer dipolar coupling…

H || dimer axis:
(no anisotropy)



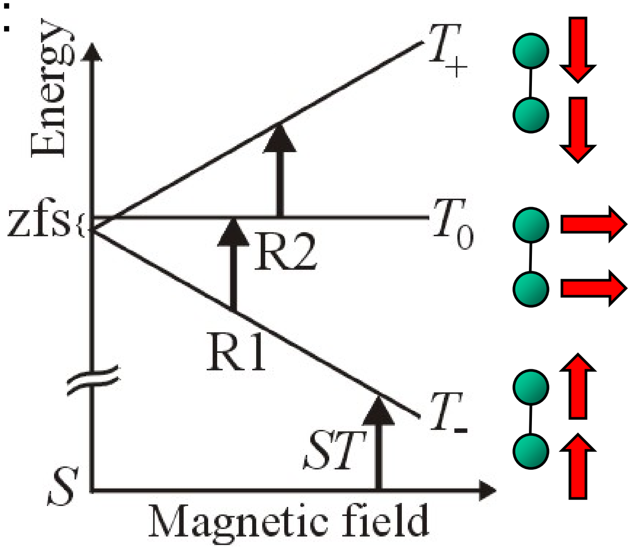
H ⊥ dimer axis:
(anisotropy)

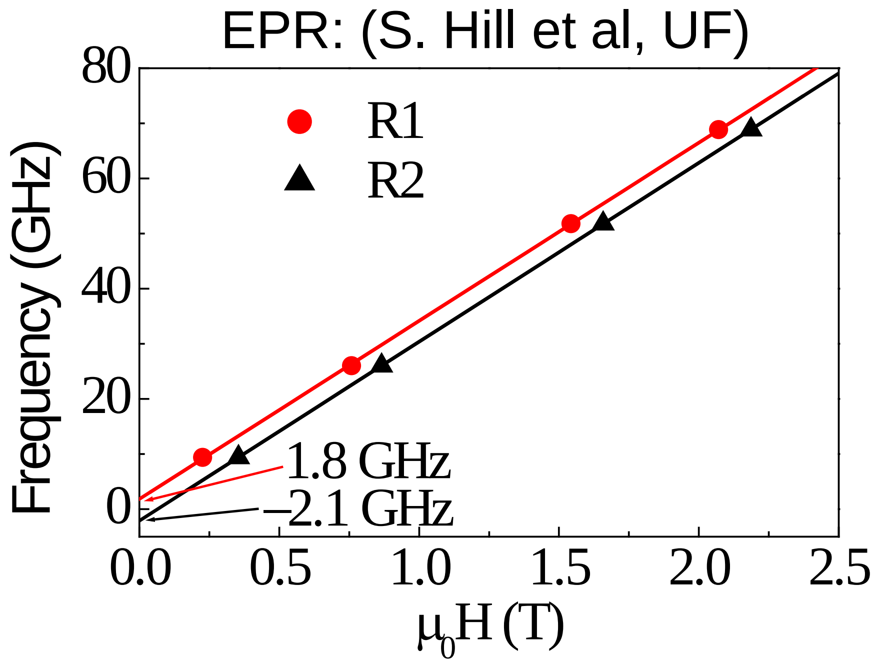


U(1) symmetry?

- Intradimer dipolar coupling causes zfs of the triplet…

H || c:





S. Sebastian et al
PRB **74**, 180401(R) (2006).

$$zfs = D = \frac{\mu_0}{16\pi r^3}\left(2g_{\parallel}^2 + g_{\perp}^2\right)\mu_B^2 \cong 0.11K \qquad H_{dip} = DS_z^2 \qquad D_{obs} = 0.10(1)\,K$$

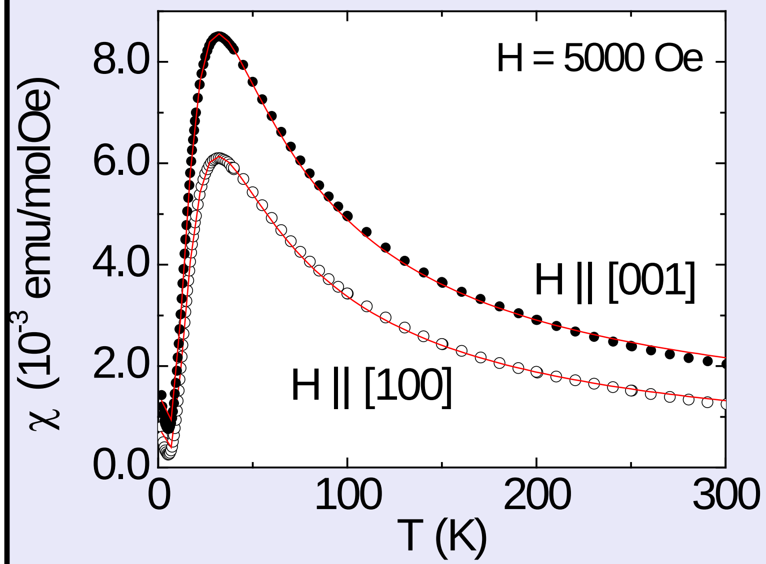
- For H ⊥ dimer axis → effective anisotropy energy (gap to "Goldstone" mode)

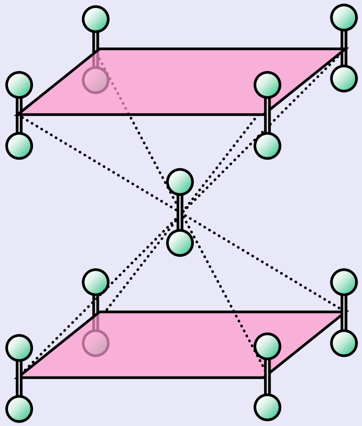
$$\approx \frac{DJ'}{J} \approx 10\,mK$$

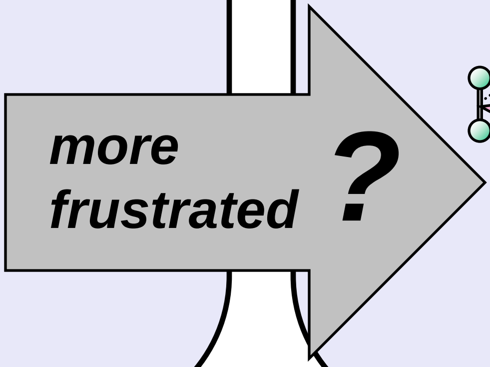
- i.e. the axial symmetry required for BEC is remarkably robust ("protected")

Case 2: triangular lattice

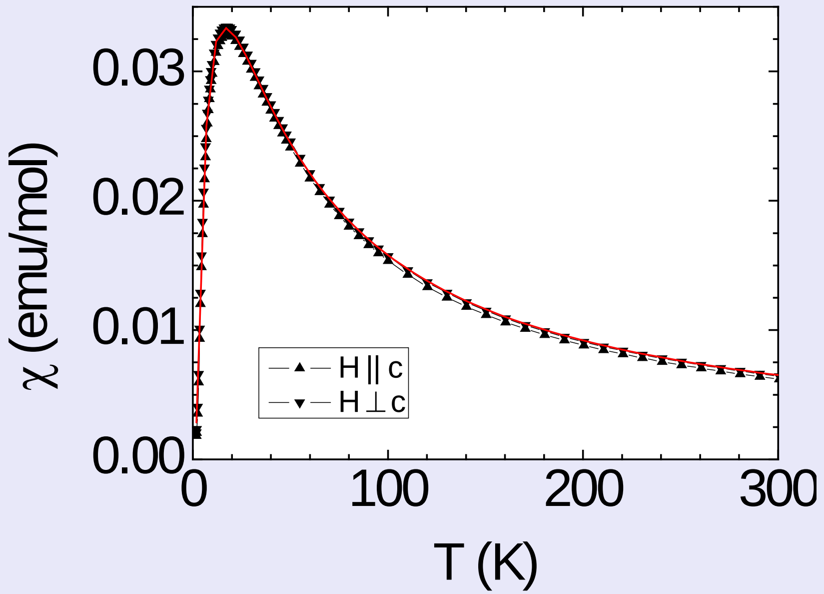
(1) $BaCuSi_2O_6$

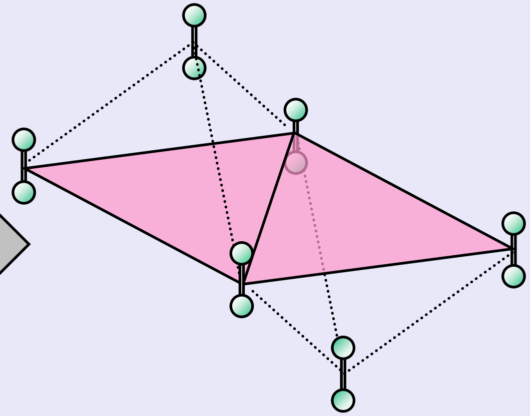






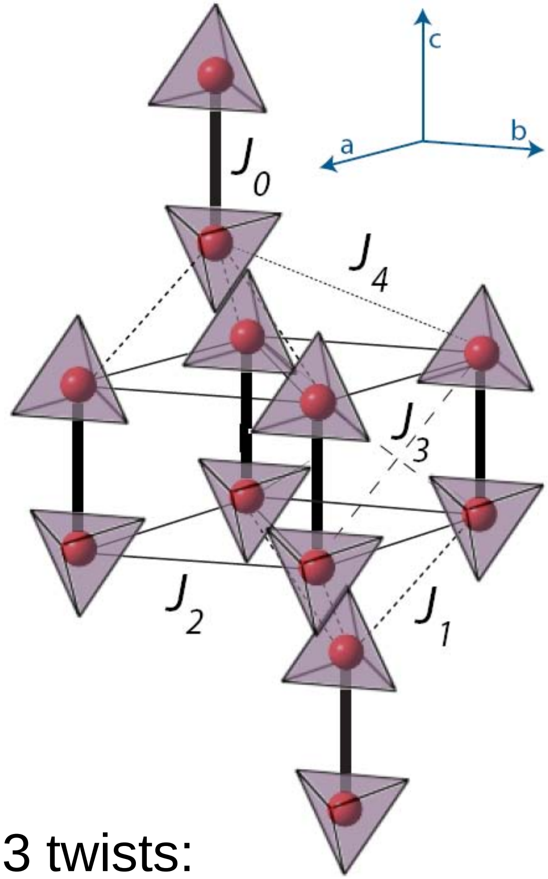
(2) $Ba_3Mn_2O_8$

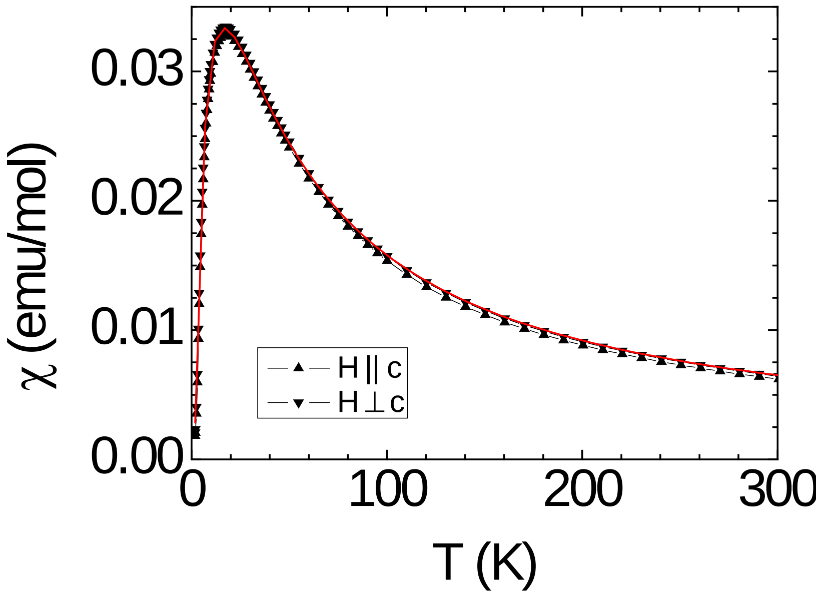


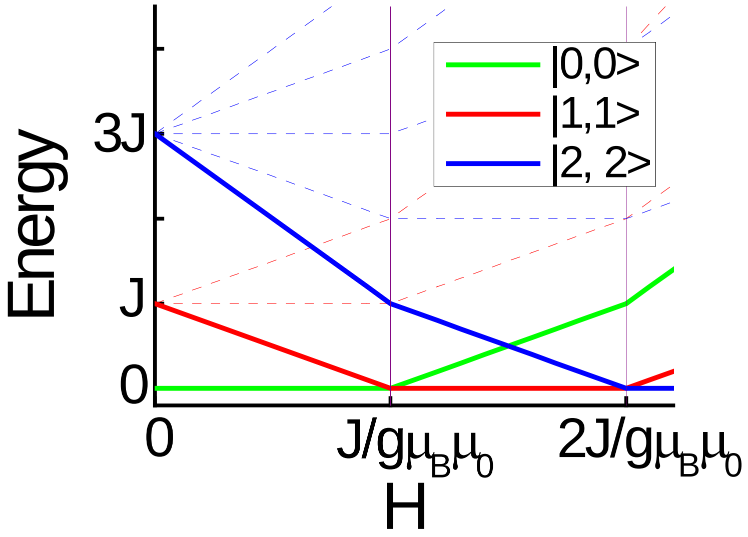


$Ba_3Mn_2O_8$

- $R\bar{3}m$
- Mn^{5+}, $3d^2$, s=1 dimers







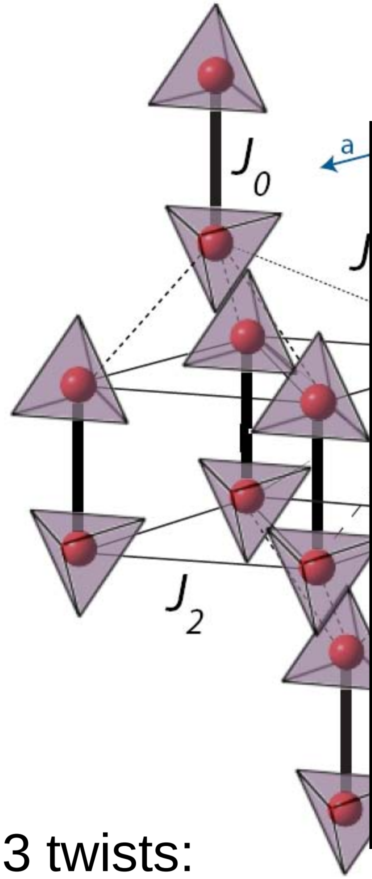
E. Samulon et al, PRB **77**, 214441 (2008).

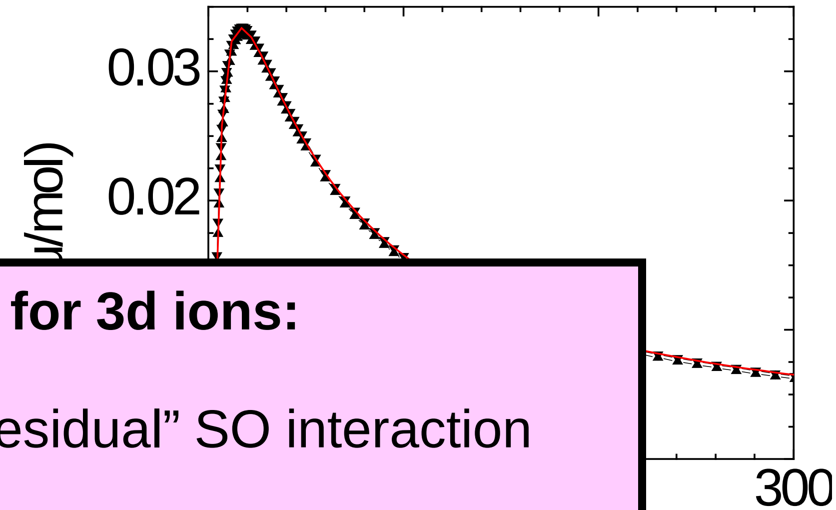
3 twists:

- in-plane frustration
- single ion anisotropy
- triplet and quintuplet condensation

$Ba_3Mn_2O_8$

- $R\bar{3}m$
- Mn^{5+}, $3d^2$, s=1 dimers



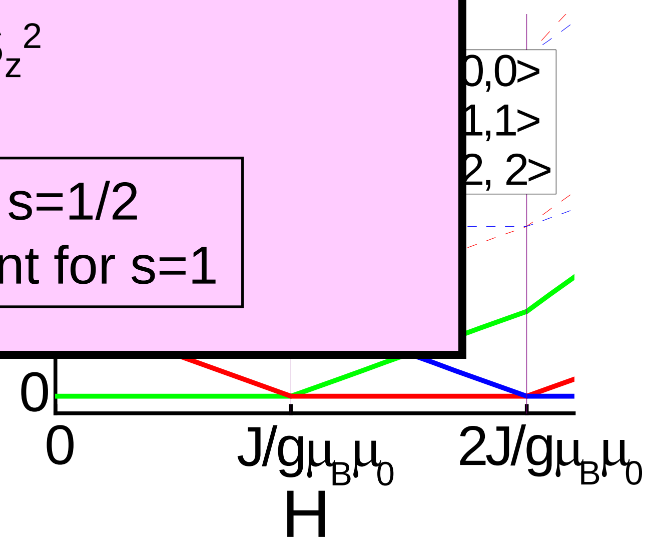


Spin-orbit coupling for 3d ions:

L is quenched, but “residual” SO interaction leads to…

- g anisotropy: g → $g_{\mu\nu}$
- anisotropy energy: DS_z^2

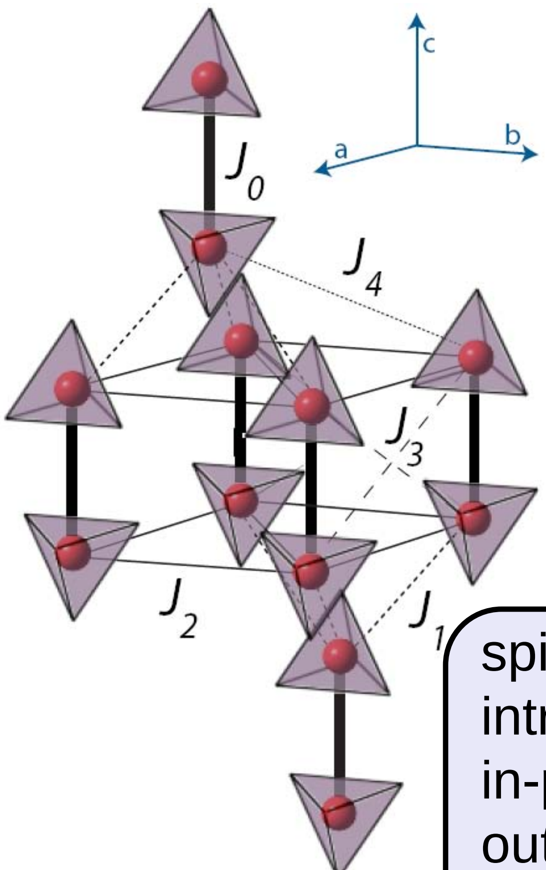
no effect for s=1/2
but significant for s=1

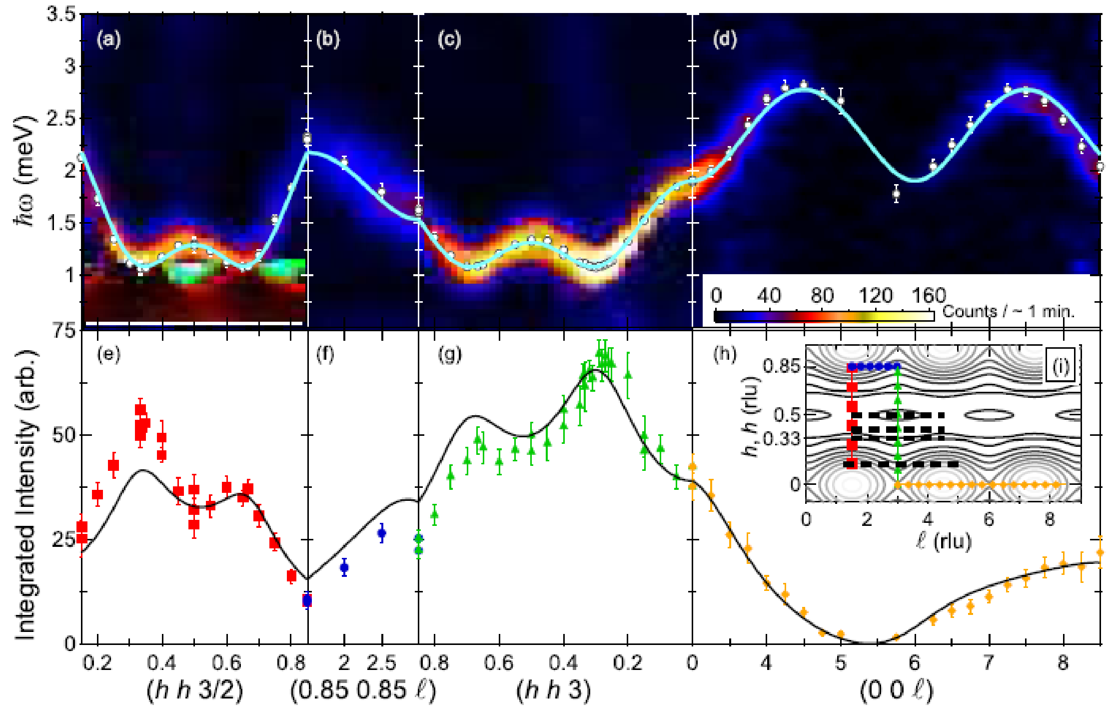


3 twists:
- in-plane frustration
- single ion anisotropy
- triplet and quintuplet condensation

E. Samulon et al, PRB **77**, 214441 (2008).

Triplon dispersion (M. Stone & M. Lumsden, ORNL)





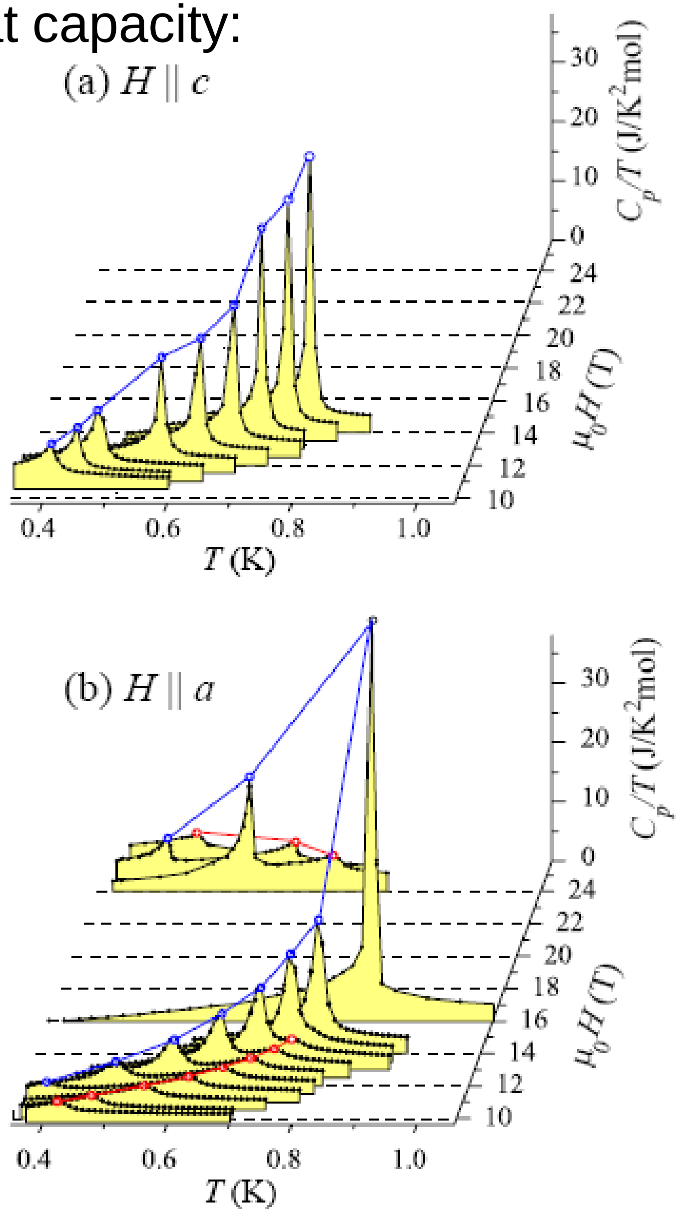
M. Stone et al, PRB **77**, 134406 (2008).
& PRL **100**, 237201 (2008).

spin gap: Δ = 1.081 meV
intradimer: J_0 = 1.642meV
in-plane interdimer: J_2 = 0.256meV (& J_3 = 0.142meV)
out-of-plane interdimer: J_1 = -0.118meV (& J_4 = -0.037meV)
single ion anisotropy: D = -0.032meV (from EPR)

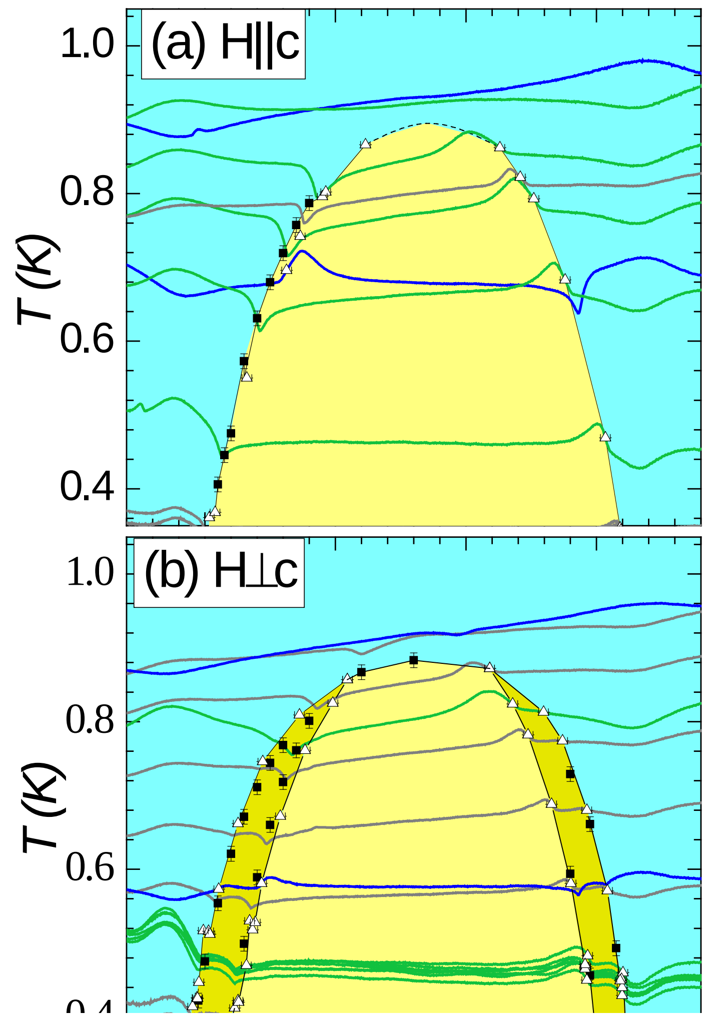
- i.e. quasi-2D material in which planes of vertical dimers arranged on triangular layers interact weakly in the perpendicular direction

High field behavior: triplet ordered states

Heat capacity:



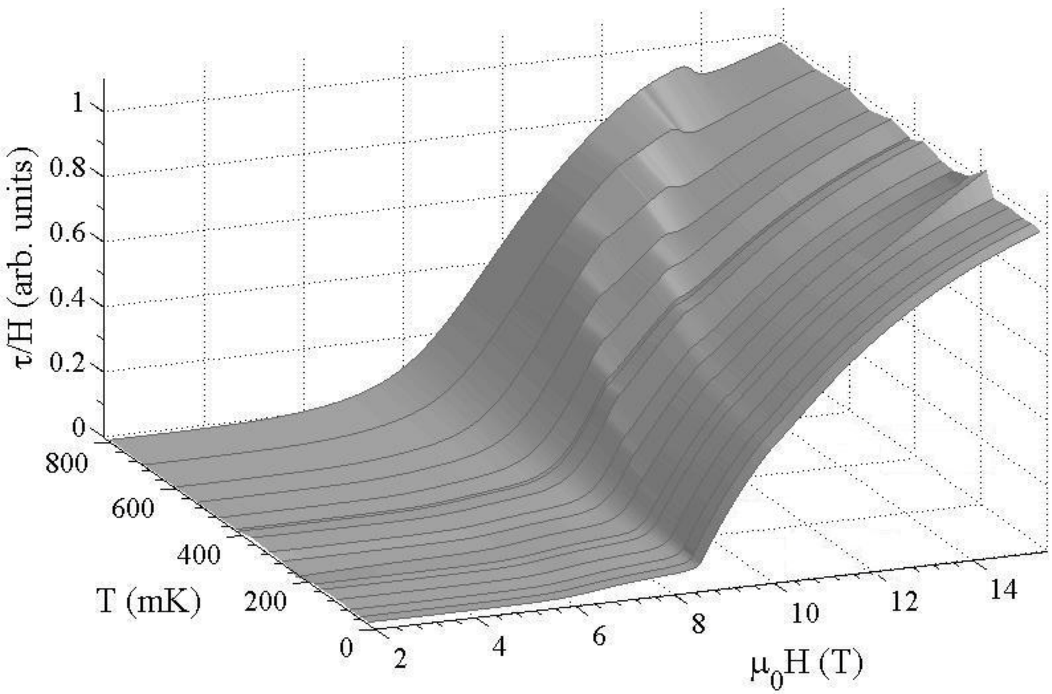
MCE (with M. Jaime, NHMFL):

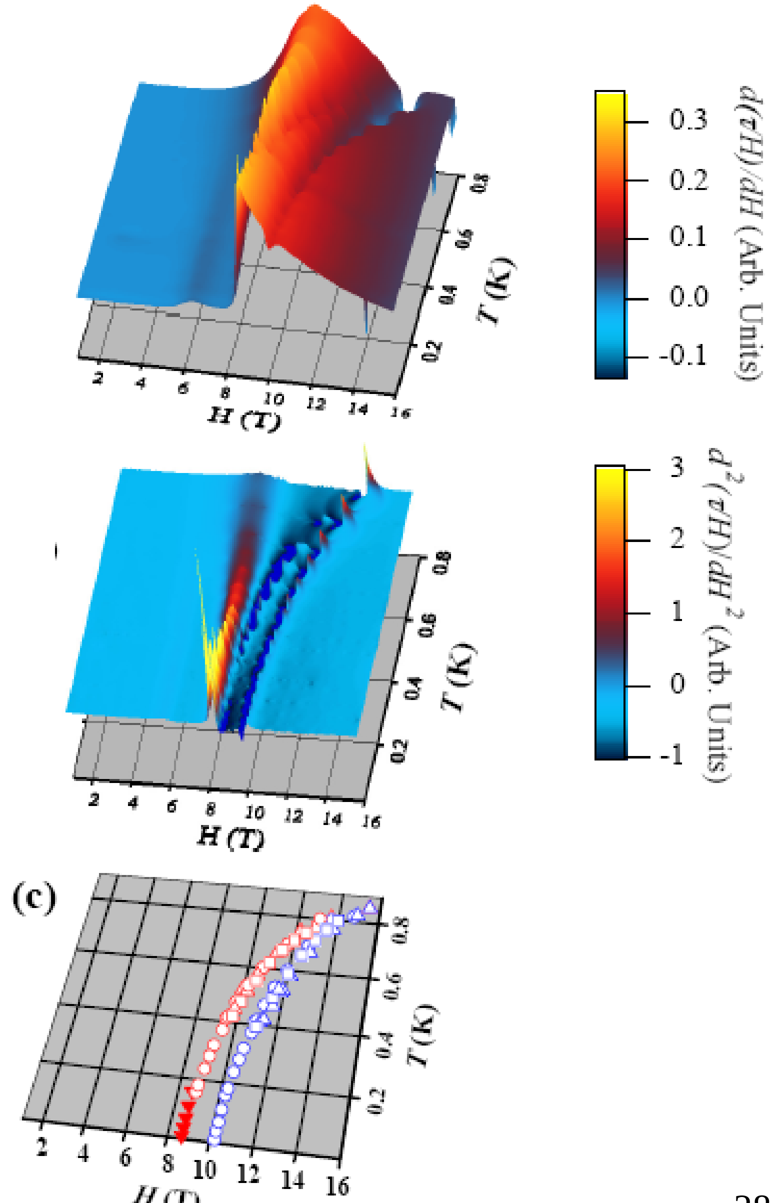


E. Samulon et al, PRB **77**, 214441 (2008).

High field behavior: triplet ordered states

Torque (with L. Balicas & Y.-J Jo, NHMFL):

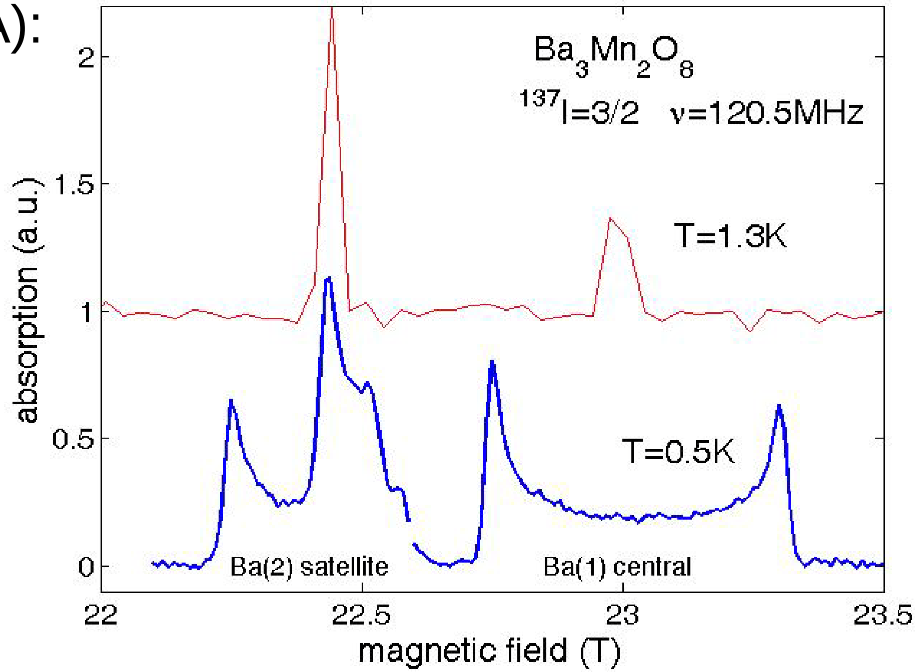


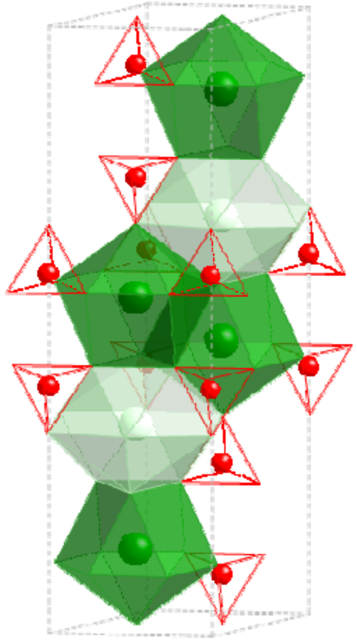


E. Samulon et al, PRB **77**, 214441 (2008).

Magnetic structure?

(a) NMR (H||c):
(S. Brown, UCLA):





(b) Neutron scattering (H||a):
(M. Stone & M. Lumsden, ORNL)

- in-plane wave vectors are both very close to (2π/3a, 2π/3a)

- in absence of other information, resort to an analysis of the spin Hamiltonian for a qualitative understanding of the ground states…

Spin Hamiltonian for $Ba_3Mn_2O_8$

Theory: Cristian Batista (LANL)

For $Ba_3Mn_2O_8$ the minimal spin Hamiltonian is:

$$H = \sum_{i,j,\mu,\nu} \frac{J_{i\mu j\nu}}{2} S_{i\mu} \cdot S_{j\nu} + D \sum_{i,\nu} \left(S_{i\mu}^{\eta}\right)^2 - g_{\alpha\alpha}\mu_B H \sum_{i\mu} S_{i\mu}^z$$

Where i,,j designate coordinates of dimer and m,n = {1,2} denote each of the two spins on a given dimer
D = -0.032 meV (EPR: S. Hill)

The resulting effective Hamiltonian is:

$$H = \frac{4J_1}{3} \sum_{l\langle\langle i,j\rangle\rangle} \left[\mathbf{s}_{i,l} \cdot \mathbf{s}_{j,l+1} - \frac{13}{16} s_{i,l}^z s_{j,l+1}^z \right] + \frac{8J_2}{3} \sum_{l\langle i,j\rangle} \left[\mathbf{s}_{i,l} \cdot \mathbf{s}_{j,l} - \frac{13}{16} s_{i,l}^z s_{j,l}^z \right] +$$

$$+ J_1\alpha(\eta) \sum_{l\langle\langle i,j\rangle\rangle} \left(s_{i,l}^x s_{j,l+1}^x - s_{i,l}^y s_{j,l+1}^y \right) + 2J_2\alpha(\eta) \sum_{l\langle i,j\rangle} \left(s_{i,l}^x s_{j,l}^x - s_{i,l}^y s_{j,l}^y \right)$$

$$- B \sum_{l,i} s_{i,l}^z$$

Where l is the layer index, <i,,j> indicates NN on same layer, and << i,,j >> denotes NN on adjacent layers,

$$B = g_{\alpha\alpha}\mu_B H - J_0 - 3J_2/2 - 3J_1/4 - D\delta_{\eta z}/6$$

$a(x) = -8D/[3(3J_0 - 2g_{\alpha\alpha}\mu_B H)]$ and $a(z) = 0$

i.e. anticipate canted AF, with xy components similar to triangular Heisenberg AF

Form approximate eigenstates from linear combination of singlet and triplet:

$$|\psi_{i,l}\rangle = \cos\theta_{i,l}|00\rangle + \sin\theta_{i,l} e^{i\phi_{i,l}}|11\rangle \longrightarrow |\psi_{\vec{r}}\rangle = \sqrt{\frac{1 \mp \xi(\vec{r})}{2}}|00\rangle + \sqrt{\frac{1 \pm \xi(\vec{r})}{2}} e^{i\phi_{\vec{r}}}|11\rangle$$

Where $\xi(\vec{r}) = \sqrt{\cos^2 2\theta + \sin^2 2\theta \sin^2\gamma \sin^2 \vec{q}.\vec{r}}$ & $\tan\phi_{\vec{r}} = \cos\gamma \tan\vec{q}.\vec{r}$
& cos(γ) describes anisotropy in xy plane

Determine the ground states by minimizing E with respect to each parameter

Spin Hamiltonian for $Ba_3Mn_2O_8$ — Theory: Cristian Batista (LANL)

For $Ba_3Mn_2O_8$ the minimal spin Hamiltonian is:

$H = \sum_{i,j,}$ …

Where i, j designate coordinates of dimer … e each of the two … r … : S. Hill)

The resu…

$H = \frac{4J}{3}$ …

$+ J_1 \alpha(\eta$ …

$- B \sum_{l,i} S$ …

…ndex, <i,j> indicates …nd << i,j >> denotes …rs, … $/2 - 3J_1/4 - D\delta_{\eta z}/6$ … $\mu_B H)]$ and $a(z) = 0$

i.e. antici… …senberg AF

Form app… …triplet:

$|\psi_{i,l}\rangle =$ … $e^{i\phi_{\vec{r}}}|11\rangle$

Aside: Classical ground state for a Heisenberg AF on a triangular lattice

For a single triangular plaquette:

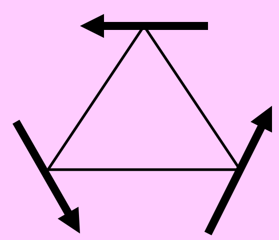
$$E = J\left(\vec{S}_1 \cdot \vec{S}_2 + \vec{S}_2 \cdot \vec{S}_3 + \vec{S}_3 \cdot \vec{S}_1\right) \equiv \frac{J}{2}\left(\vec{S}_1 + \vec{S}_2 + \vec{S}_3\right)^2 - \frac{3}{2} J S^2$$

From which the energy is minimized when…

$$\vec{S}_1 + \vec{S}_2 + \vec{S}_3 = 0$$

… corresponding to a 120° structure:

q = (2π/3, 2π/3)

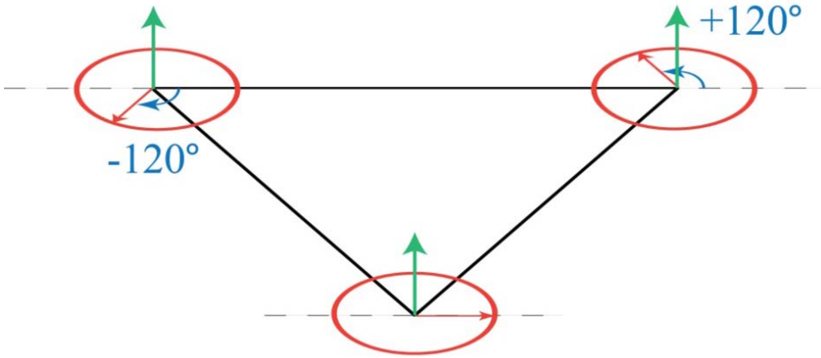


Where $\xi(\vec{r}) = \sqrt{\cos^2 2\theta + \sin^2 2\theta \sin^2 \gamma \sin^2 \vec{q}.\vec{r}}$ & $\tan\phi_{\vec{r}} = \cos\gamma \tan\vec{q}.\vec{r}$
& cos(γ) describes anisotropy in xy plane

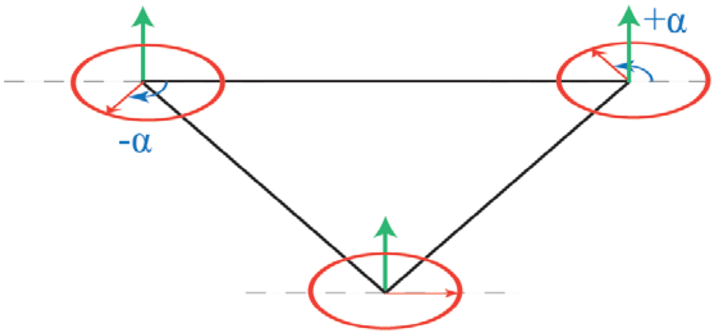
Determine the ground states by minimizing E with respect to each parameter

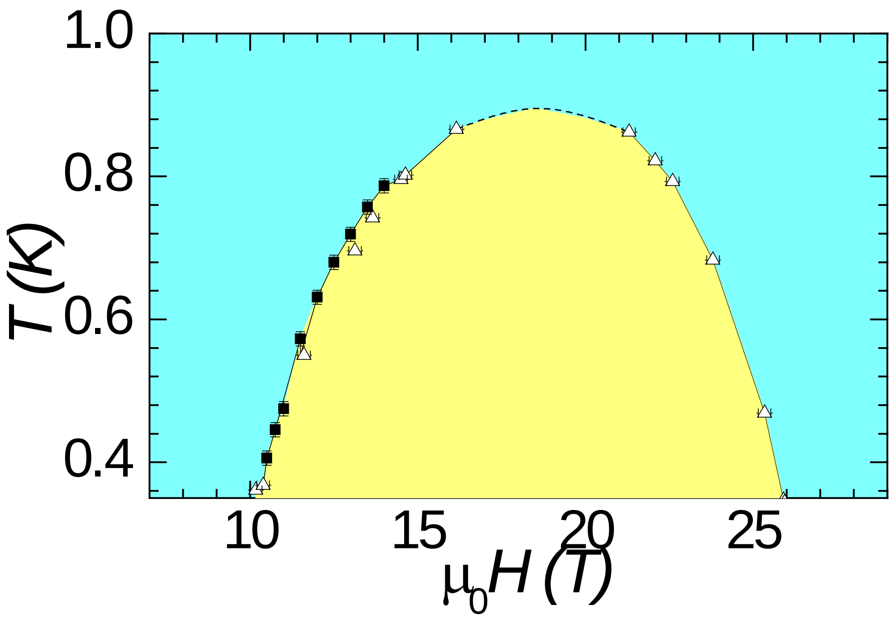
Classical groundstates: H || c

(a) no interlayer coupling (J_1 = 0):



(b) finite interlayer coupling (J_1 > 0):





ΔE due to deviation from 120°:

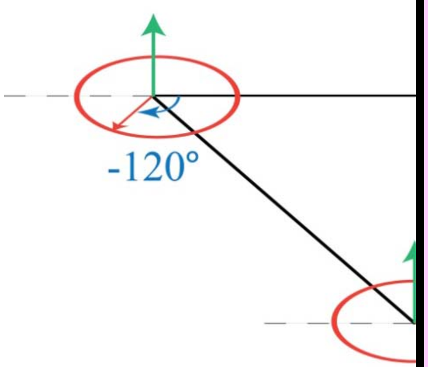
in-plane coupling: even (quadratic)

out-of-plane coupling: odd (linear)

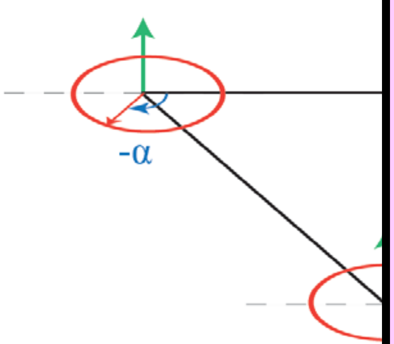
$$\alpha = \cos^{-1}\left(-\frac{1}{2}+\frac{J_1}{4J_2}\right)\approx 117^\circ$$

Classical groundstates: H || c

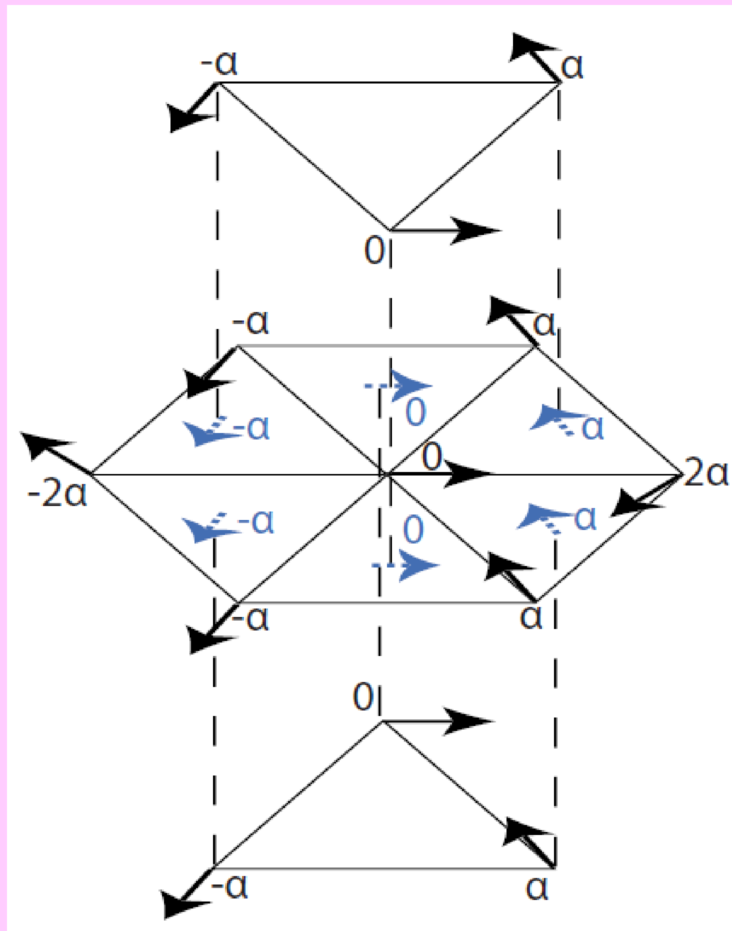
(a) no interlayer

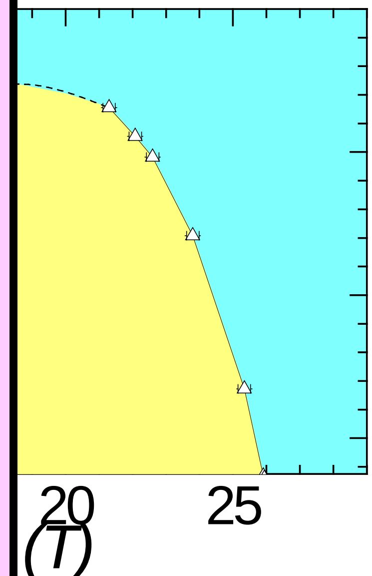


(b) finite interlay



Ferromagnetic interlayer coupling
→ in-plane spiral structure:





from 120°:

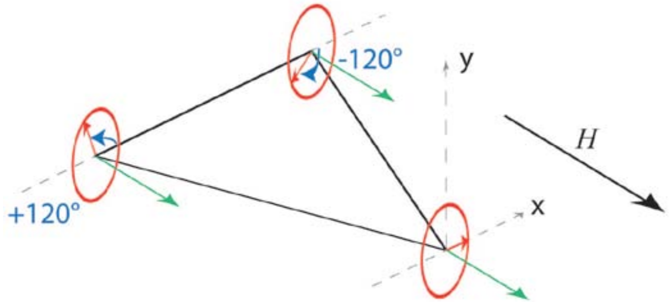
ven (quadratic)

g: odd (linear)

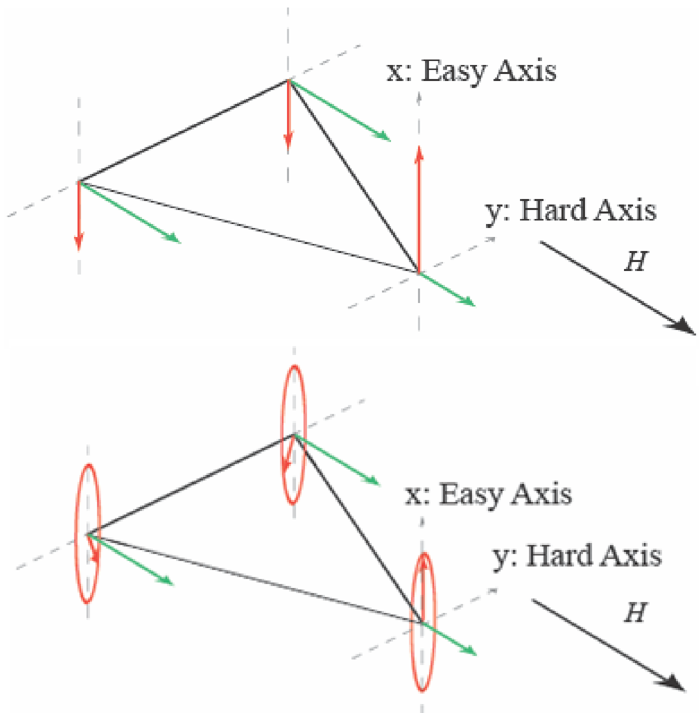
$$\frac{J_1}{J_2}\Bigg) \approx 117°$$

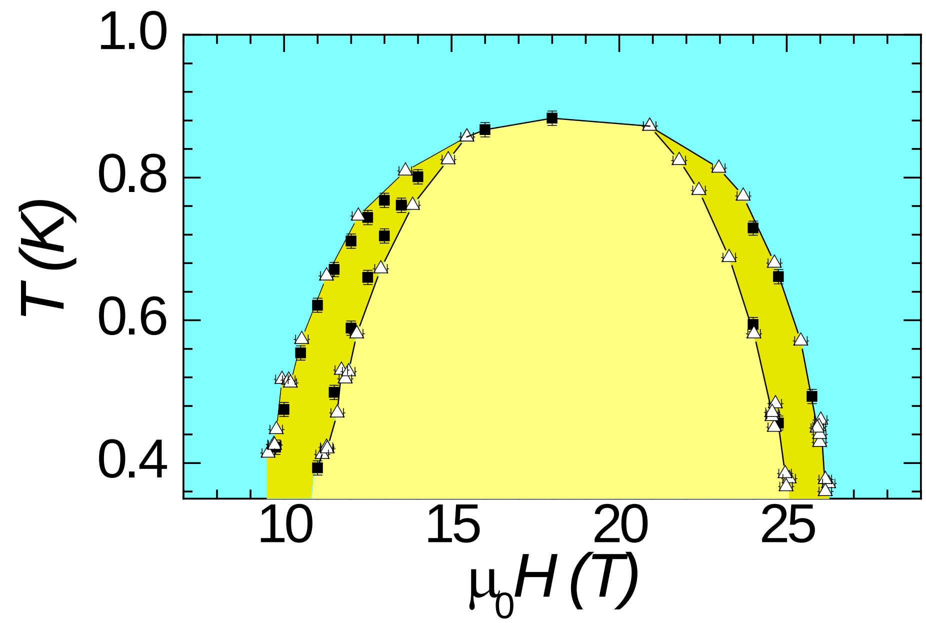
Classical groundstates: H ⊥ c

(a) D = 0, J_1 = 0:



(b) D < 0, J_1 > 0:



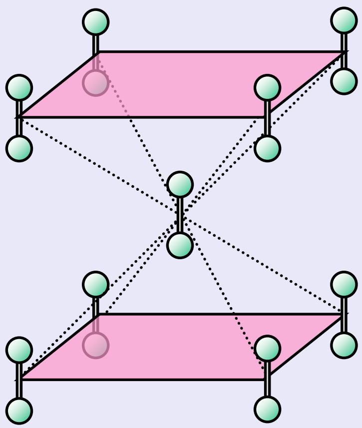


- easy axis in x-y plane
- sum of xy components ~ 0
- → modulated structure
- but modulation costs energy…

$$\frac{8J_2}{3}\sum_{l<i,j>}\left[\mathbf{s}_{i,l}\cdot\mathbf{s}_{j,l}-\frac{13}{16}s^z_{i,l}s^z_{j,l}\right]$$

What about dimensional reduction?

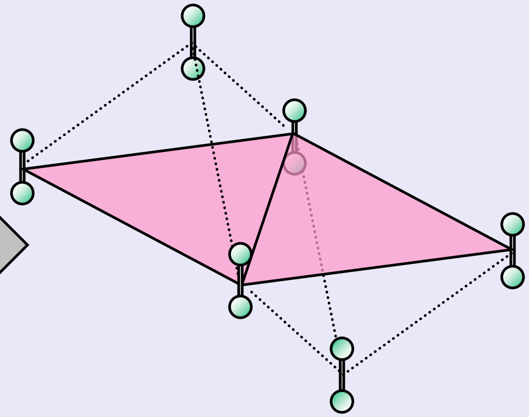
(1) $BaCuSi_2O_6$



- q = (π/a, π/a)
- frustrates interlayer coupling

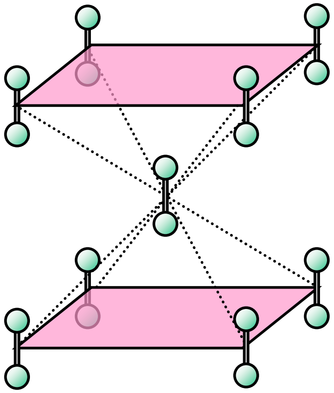
***more frustrated* ?**

(2) $Ba_3Mn_2O_8$



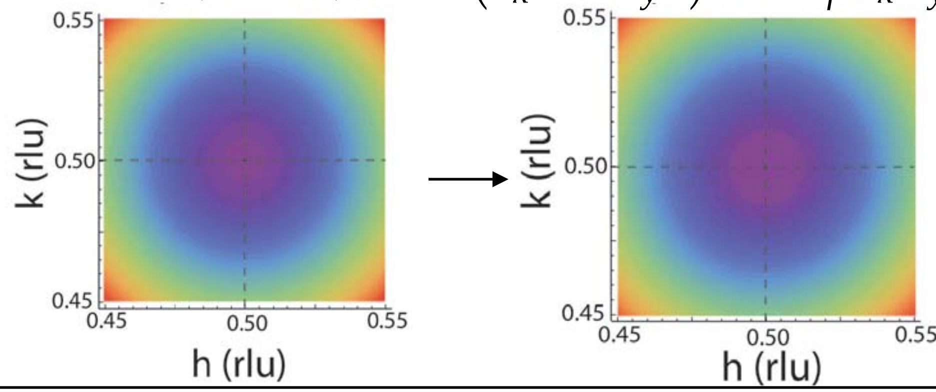
- if q = (2π/3a, 2π/3a), then interlayer coupling is also perfectly frustrated
- but q ≠ (2π/3a, 2π/3a)…

$BaCuSi_2O_6$ vs $Ba_3Mn_2O_8$: symmetry

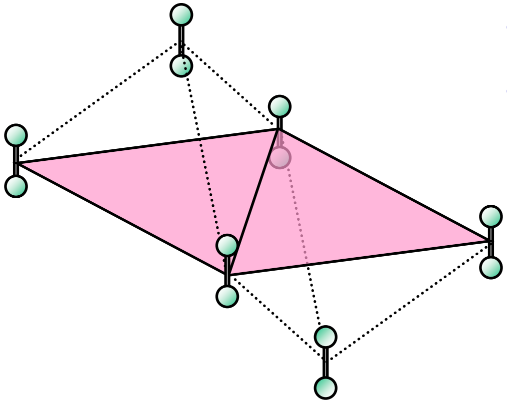


- Planar inversion symmetry about Q= (π/a, π/a)
- Interlayer coupling can only introduce terms quadratic in k

$$E(Q+k) \approx J'\left(k_x^2+k_y^2\right)+2J_f k_x k_y$$

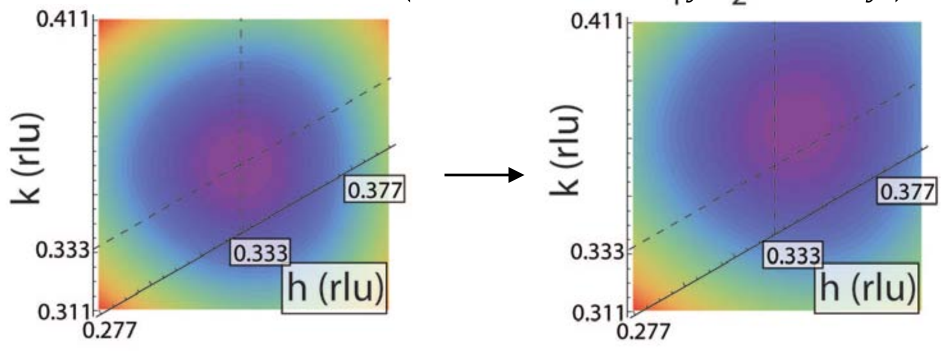


Q= (π/a, π/a) ⇒ layers decouple



- No inversion symmetry about Q=(2π/3a, 2π/3a)
- Interlayer coupling introduces terms *linear* in k

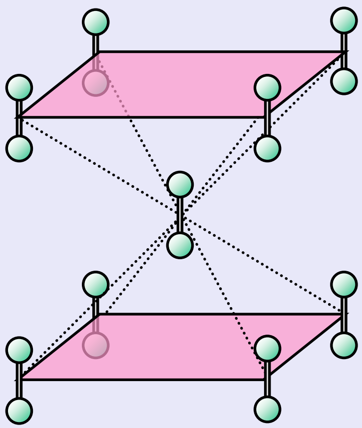
$$E(Q+k) \propto J_2\left(-3+k_x^2+k_y^2+k_x k_y\right)+\frac{\sqrt{3}}{2}J_1\left(k_x+k_y\right)$$



Q≠(2π/3a,2π/3a) ⇒ 3D spiral

So which lattice is more frustrated?

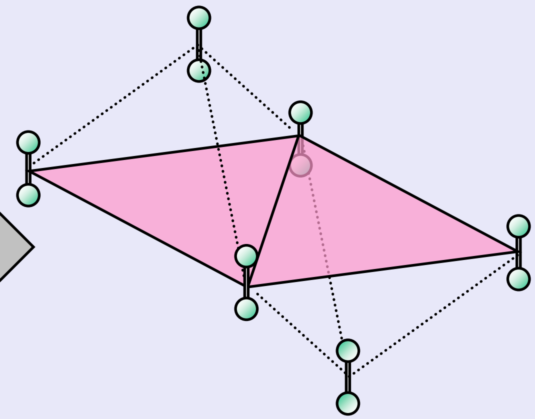
(1) $BaCuSi_2O_6$



robust interlayer frustration
→ dimensional reduction at T = 0

less frustrated !

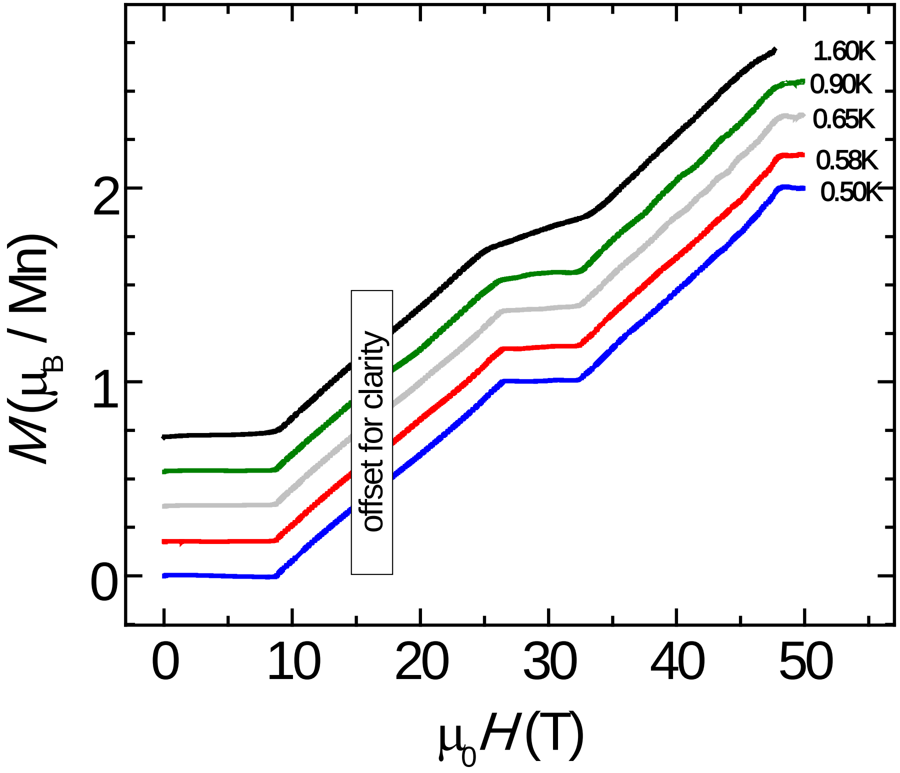
(2) $Ba_3Mn_2O_8$



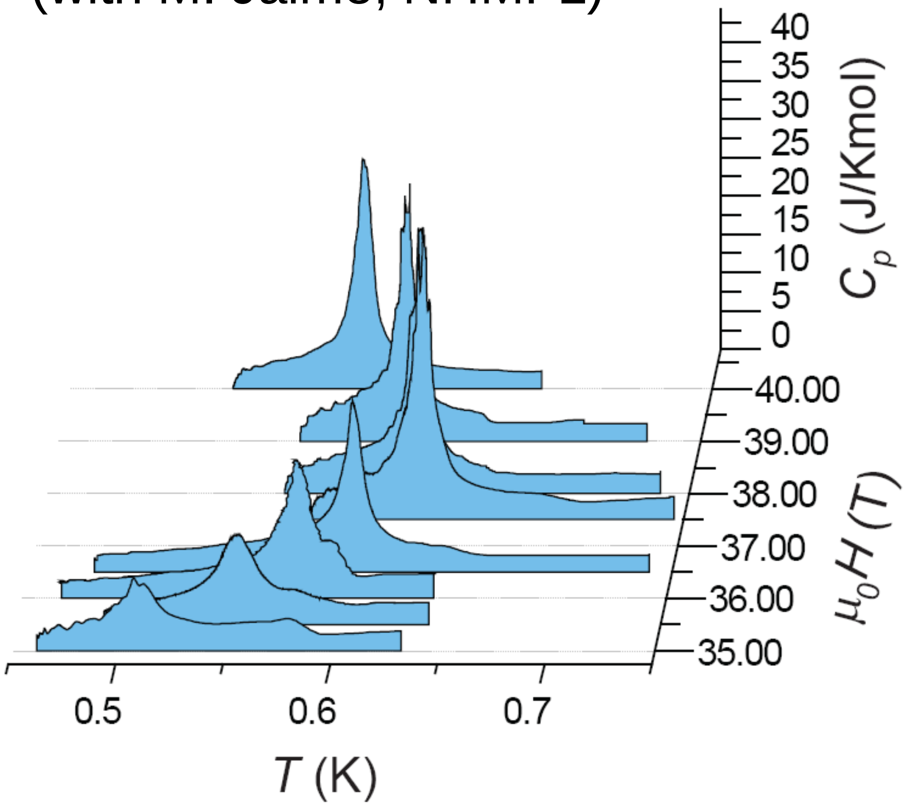
perfect frustration is not protected
→ 3d incommensurate spiral

What about the quintuplet states of $Ba_3Mn_2O_8$?

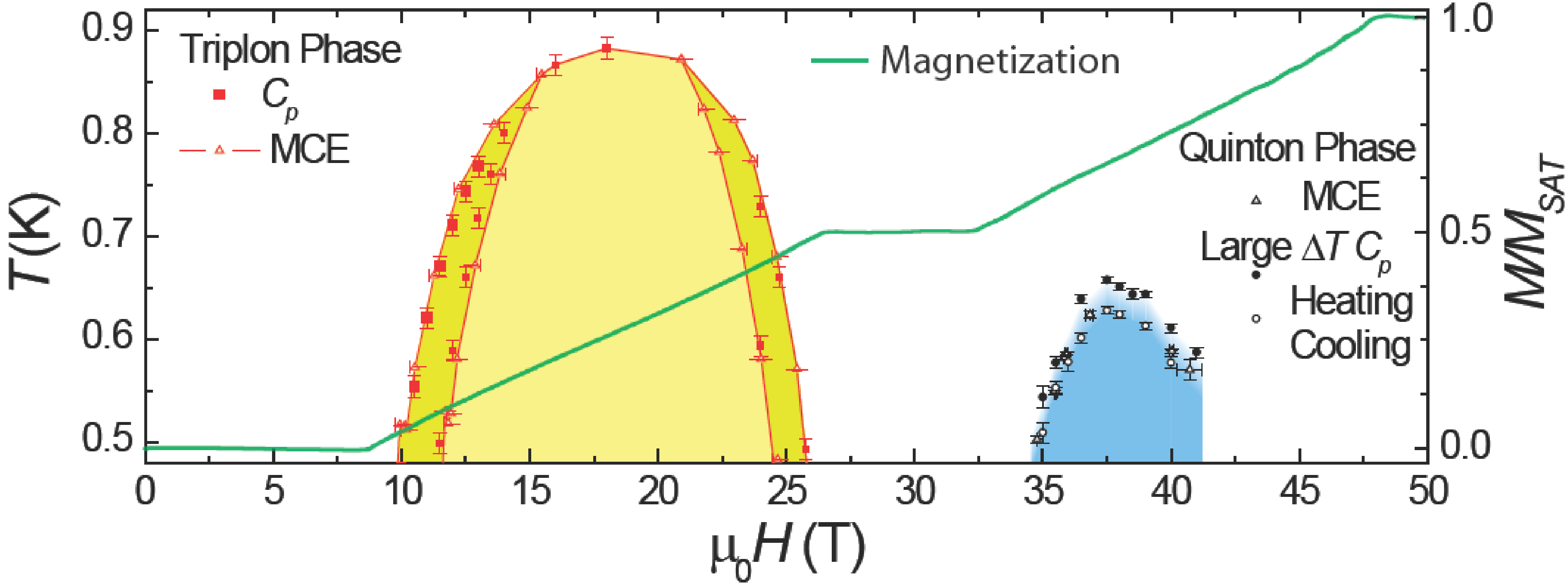
Magnetization:
(with R. McDonald, NHMFL)



C_p & MCE:
(with M. Jaime, NHMFL)



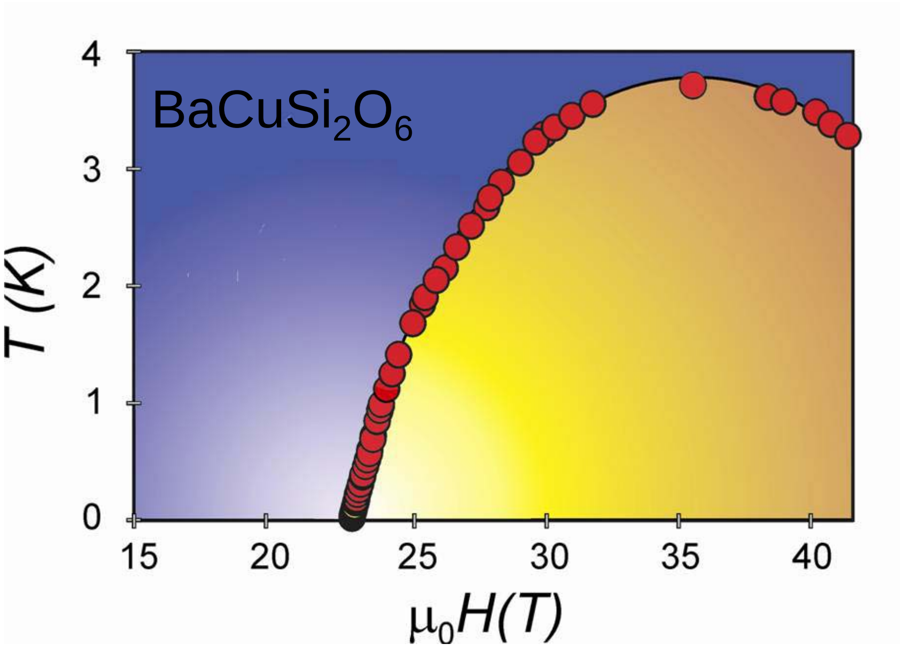
What about the quintuplet states of $Ba_3Mn_2O_8$?

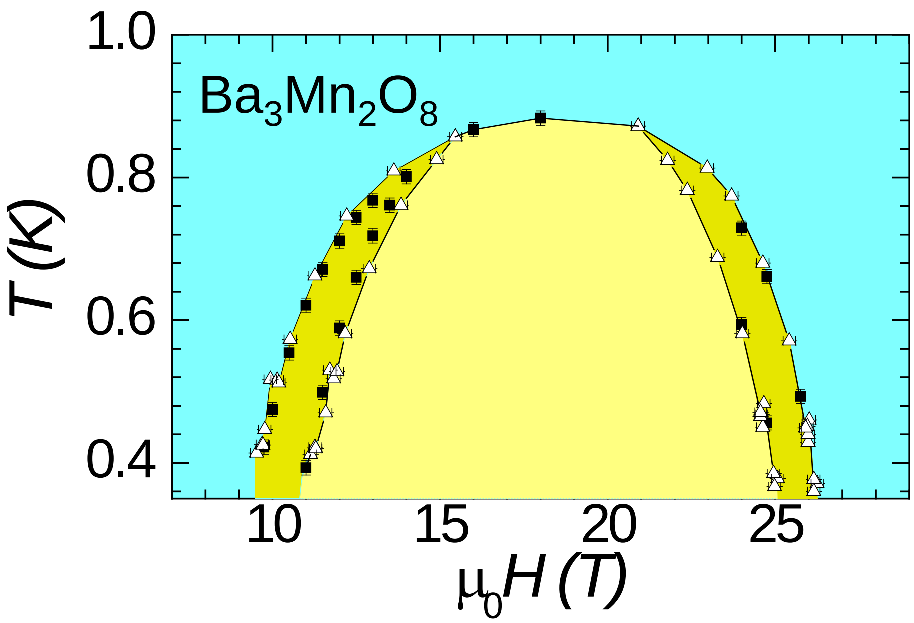


- Second region of LRMO
- Can be understood following a similar treatment of the effective Hamiltonian

Summary

Spin dimer compounds provide access to some exciting physics and some beautiful magnetic structures...





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