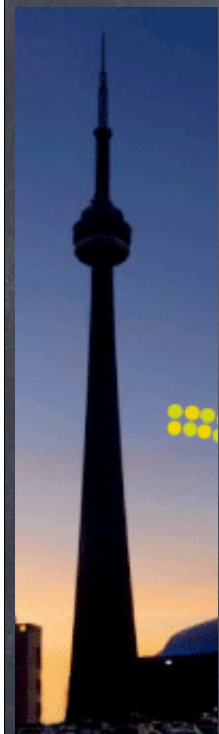


A quantum critical point in an orbitally degenerate insulator: $FeSc_2S_4$

Leon Balents

Quantum Critical Phenomena, Toronto 9/08

The David and Lucile Packard Foundation

People

The original

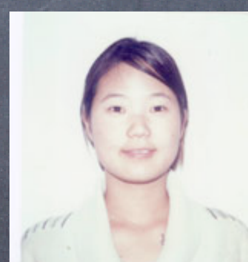
Doron Bergman

Jason Alicea

Emanuel Gull

Simon Trebst

The next generation



Sungbin Lee

Lucile Savary

the final frontier

Gang Chen

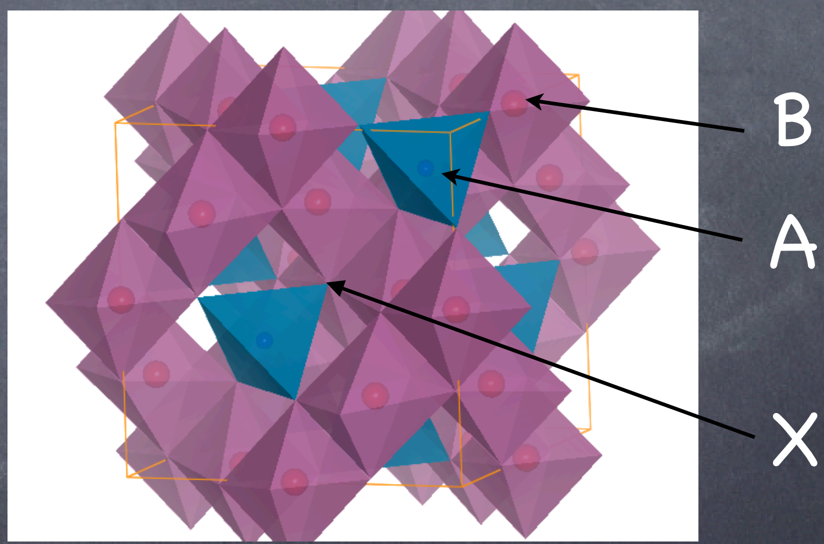
Outline

- Classical diamond lattice antiferromagnets
  - Frustration by competing interactions
- Spin-orbital physics in orbitally degenerate A-site spinels
  - A possible QCP in $FeSc_2S_4$
  - Remarkably SOI can drive spin-orbital singlet formation even for “large” S=2 spins!

AB_2X_4 spinels

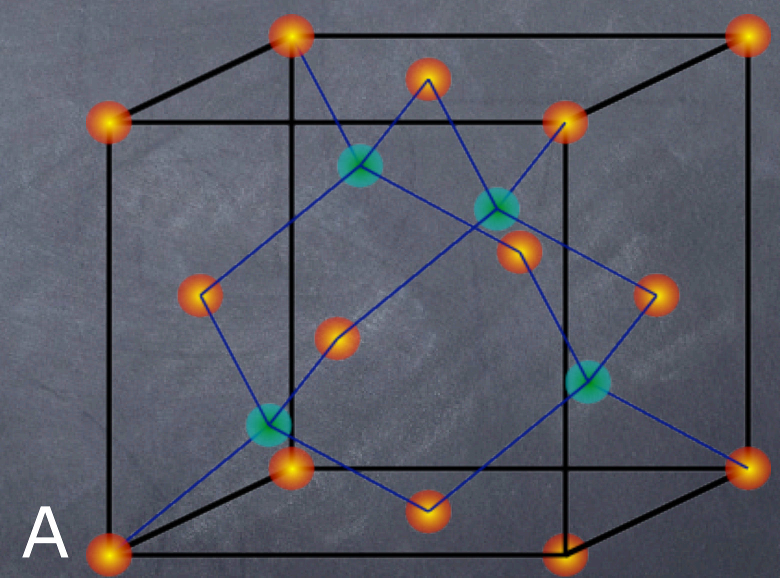
- One of the most common mineral structures
- Common valence:
  - A^{2+}, B^{3+}, X^{2-}
  - X=O,S,Se

cubic $Fd\bar{3}m$

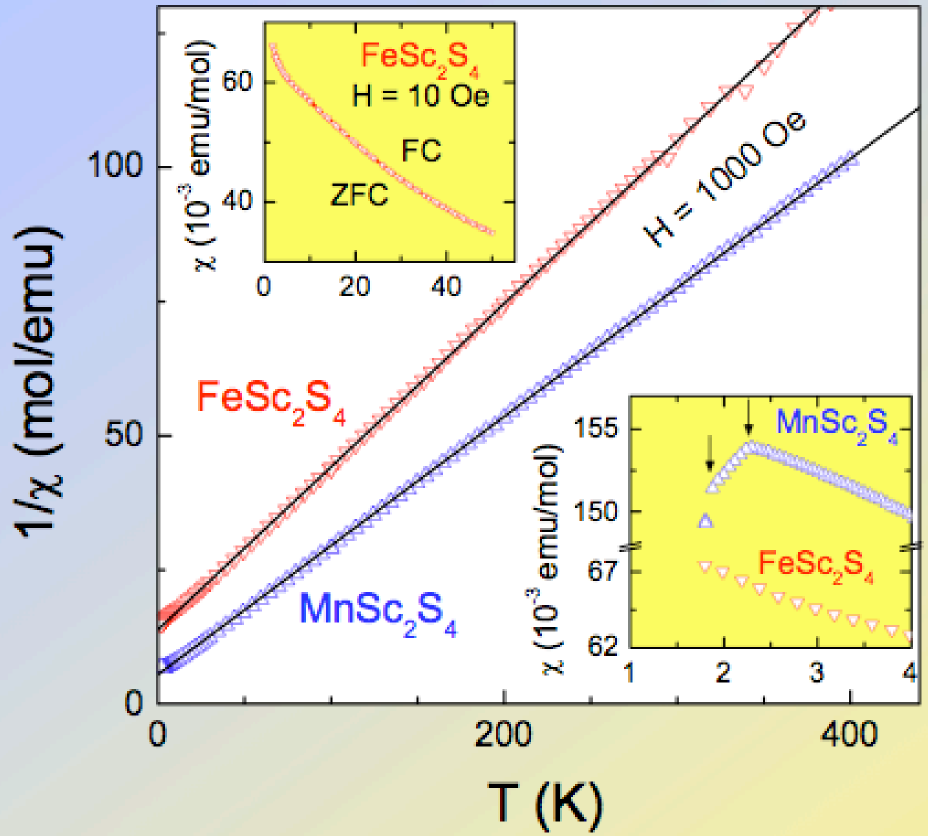


Deconstructing the spinel

- A atoms: diamond lattice
  - Bipartite: not *geometrically* frustrated



Frustrated diamond spinels



$FeSc_2S_4$: θ_{CW} = 50 K

T > 30 mK:

no long-range magnetic order

no spin-glass

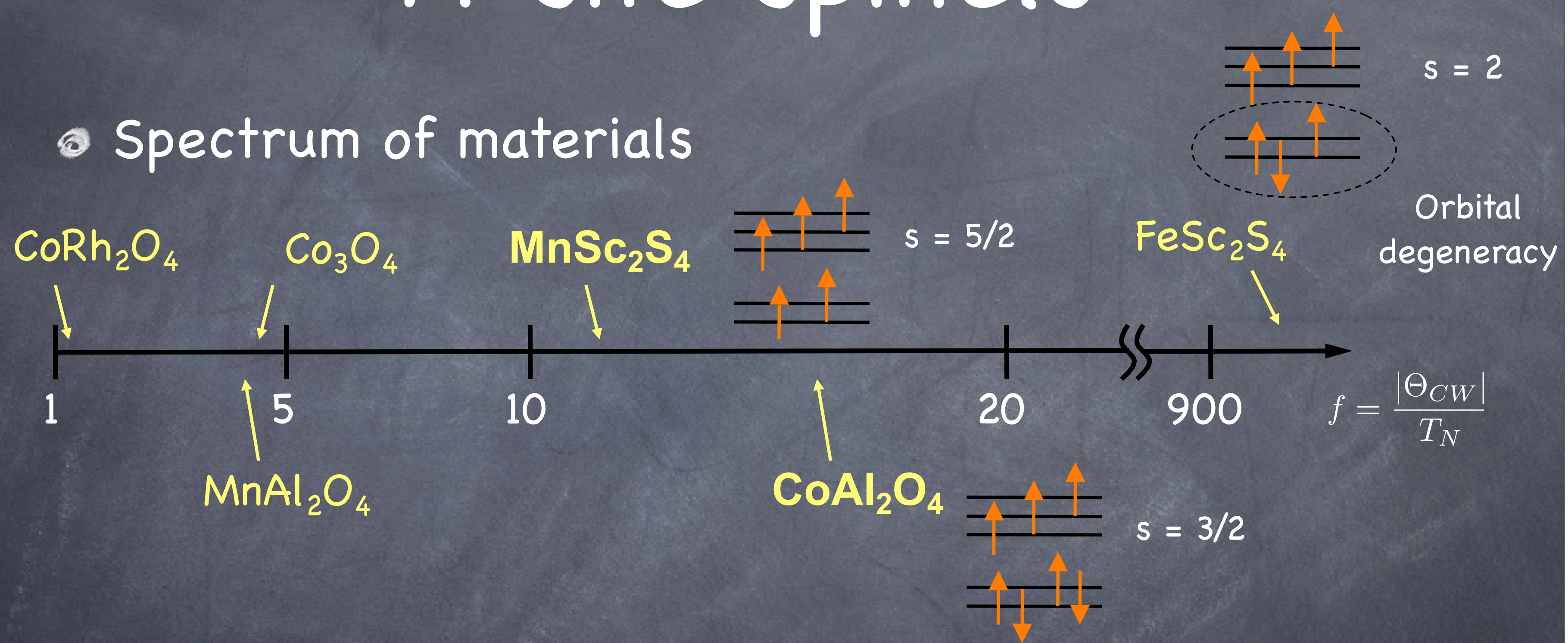
$MnSc_2S_4$: θ_{CW} = 25 K

AFM transition @ 2 K

Fritsch *et al.*, PRL **92**, 116401, 2004

A-site spinels

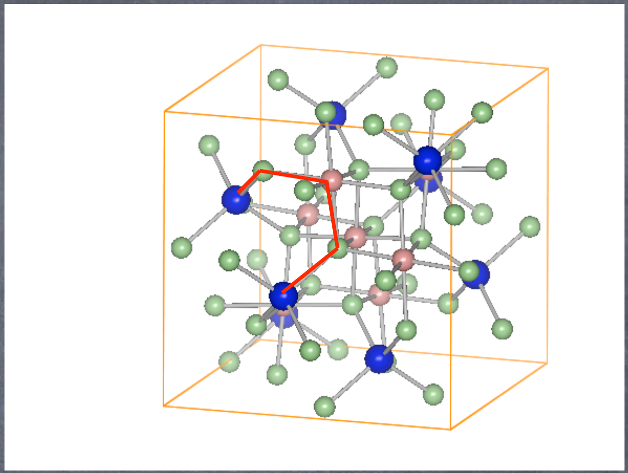
- Spectrum of materials

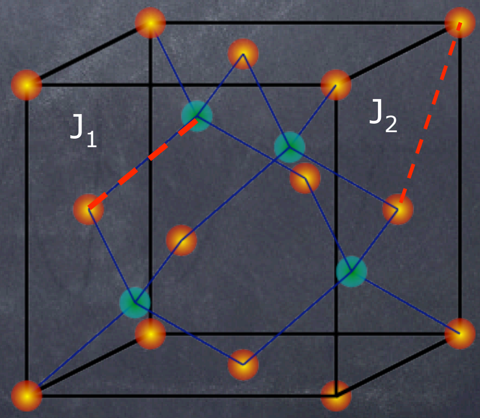


V. Fritsch et al. PRL **92**, 116401 (2004); N. Tristan et al. PRB **72,** 174404 (2005); T. Suzuki *et al.* (2006)

Why frustration?

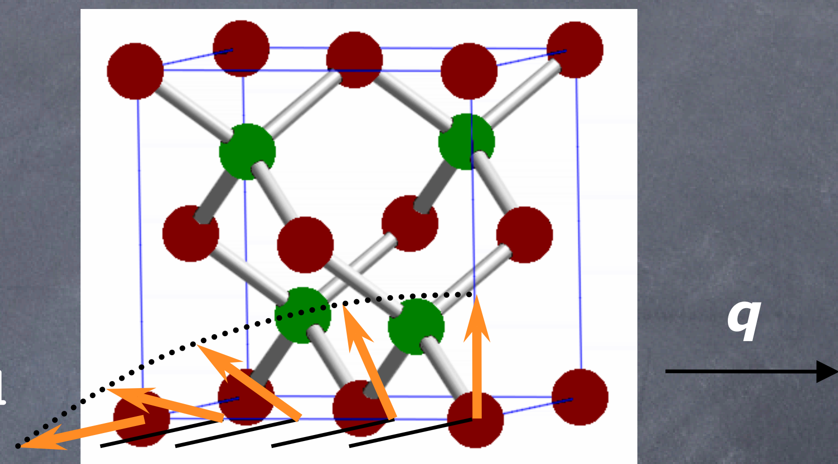
- Roth, 1964: 2nd and 3rd neighbor exchange not necessarily small
  - Exchange paths: A-X-B-X-B comparable
- Minimal model
  - J_1-J_2 exchange



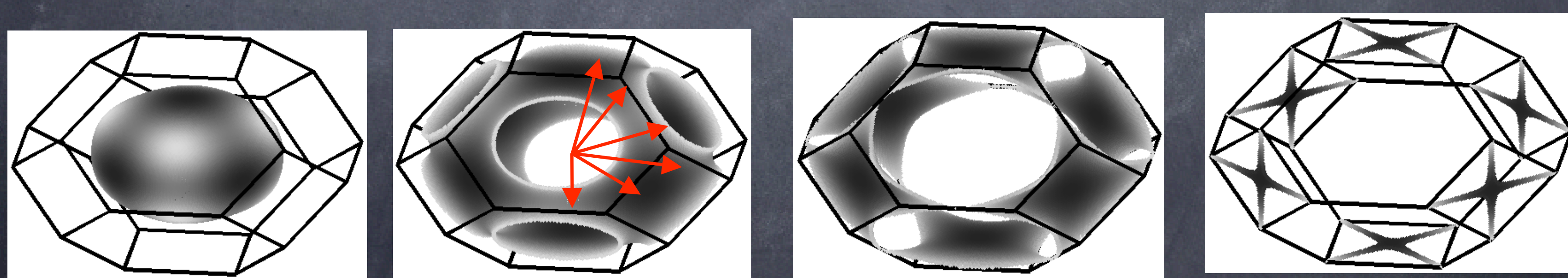


Ground state evolution

- Coplanar spirals



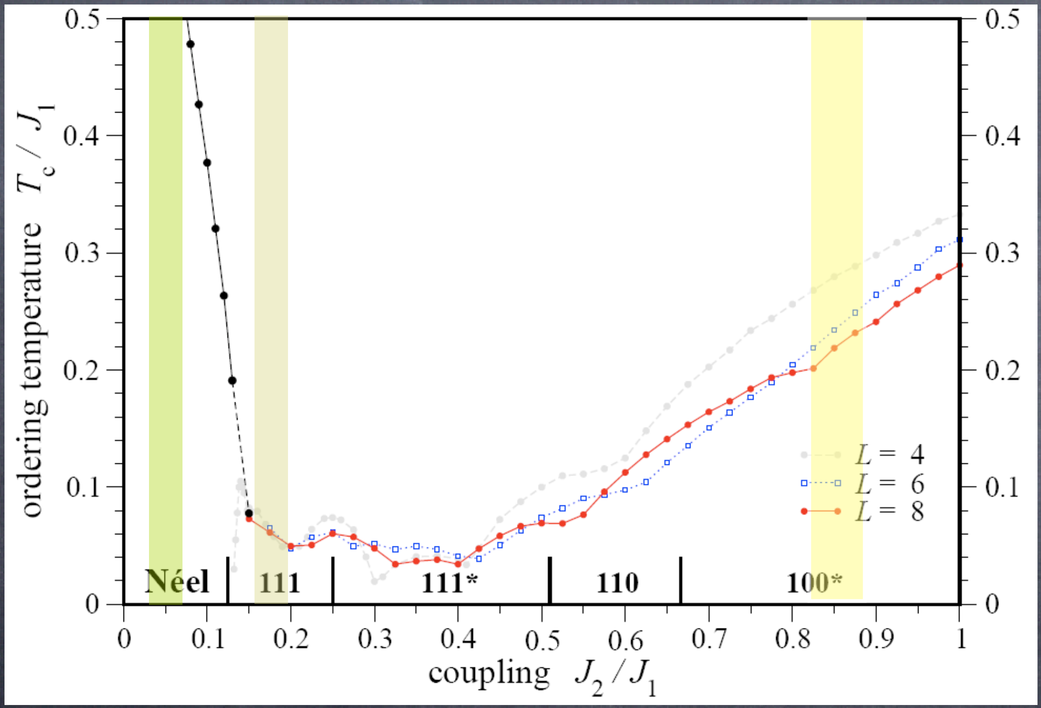
- Spiral surfaces:



$J_2/J_1 = 0.2$ $J_2/J_1 = 0.4$ $J_2/J_1 = 0.85$ $J_2/J_1 = 20$

Monte Carlo

$MnAl_2O_4$ $CoAl_2O_4$ $MnSc_2S_4$

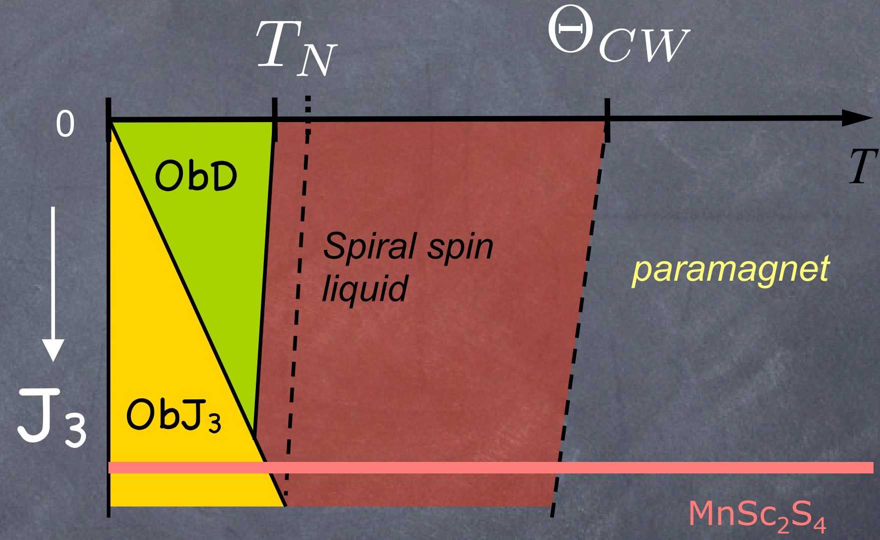


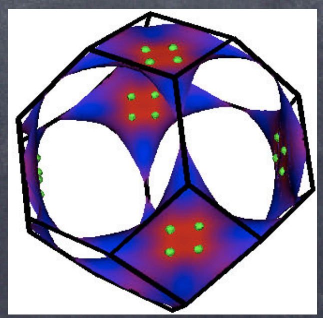
f = 11 at
$J_2/J_1 = 0.85$

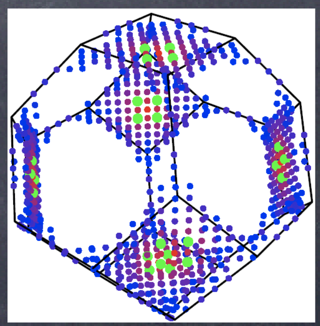
Ordering strongly suppressed by fluctuations amongst spirals

Phase Diagram

- Entropy and J_3 compete to determine ordered state
- Spiral spin liquid regime has intensity over entire spiral surface





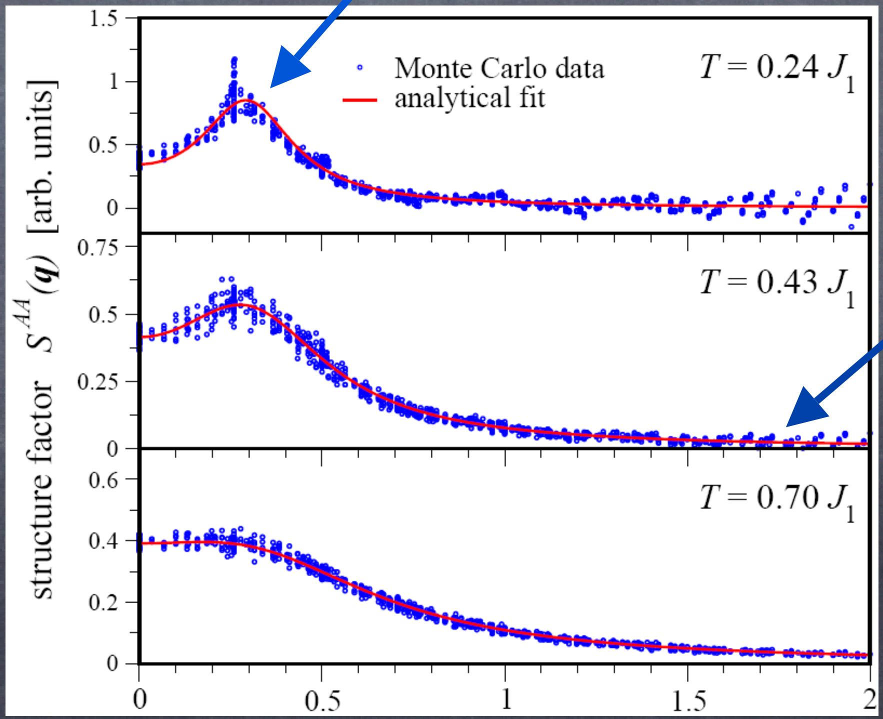


Capturing Correlations

- Spherical model
  - Predicts data collapse

$MnSc_2S_4$

Structure factor for one FCC sublattice



Peaked near surface

Quantitative agreement! (except very near T_c)

Nontrivial experimental test, but need single crystals...

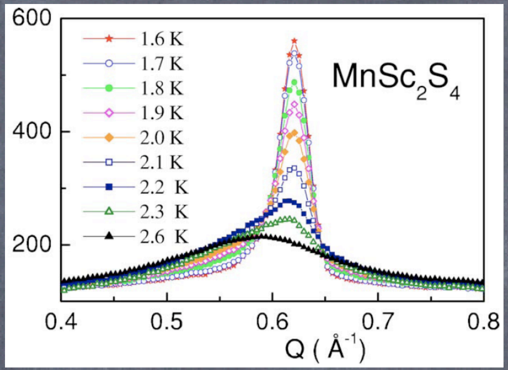
$$\Lambda(q) = 2\left[\cos^2\frac{q_x}{4}\cos^2\frac{q_y}{4}\cos^2\frac{q_z}{4} + \sin^2\frac{q_x}{4}\sin^2\frac{q_y}{4}\sin^2\frac{q_z}{4}\right]^{1/2}$$

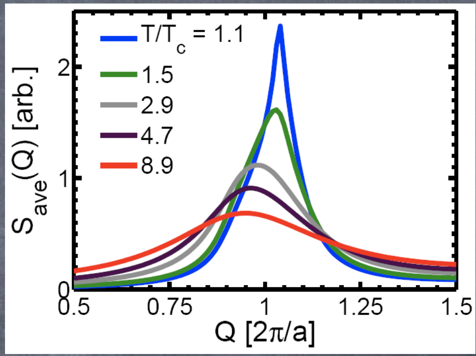
Comparison to Expts.

Expt.

Theory

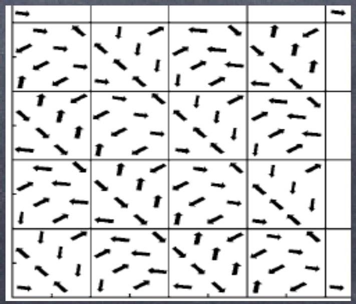
- Diffuse scattering





A. Krimmel et al, 2006

- Ordered state
  - (qq0) spiral
- Specific heat?



agrees with theory for FM J_1

J.-S. Bernier, M.J. Lawler and Y.B. Kim, 2008

Other Aspects of Spin-Only Materials

Sungbin Lee

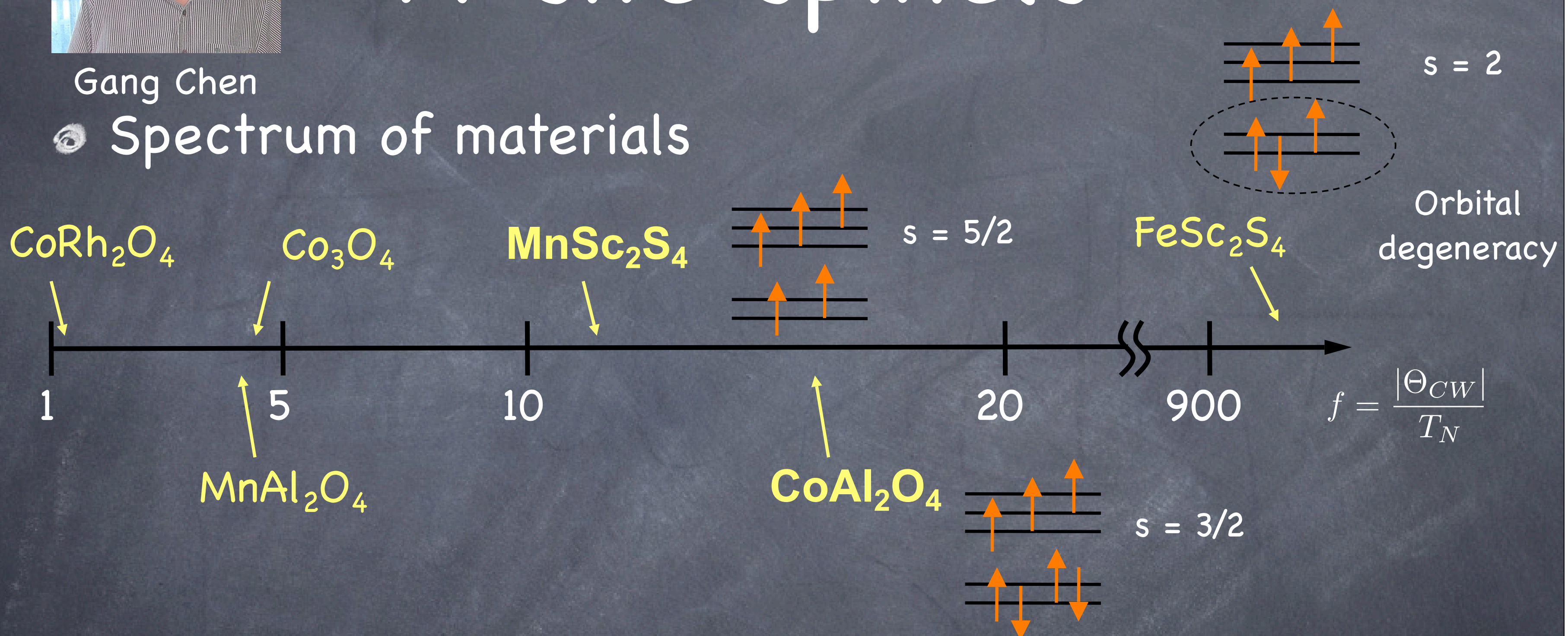
- Anisotropy (spin-orbit/dipolar) effects
  - Choice of spin plane
  - Commensurate-incommensurate transition
  - Spin flop/metamagnetic transition in field
- Disorder effects
  - Sensitivity to disorder near Lifshitz point ($CoAl_2O_4$)
- Staggered magnetization
  - Unusually large spin-wave suppression
  - Suppression by disorder

Lucile Savary

A-site spinels

Gang Chen

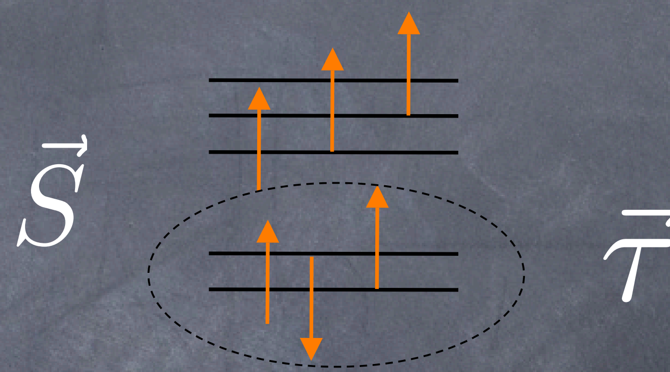
- Spectrum of materials

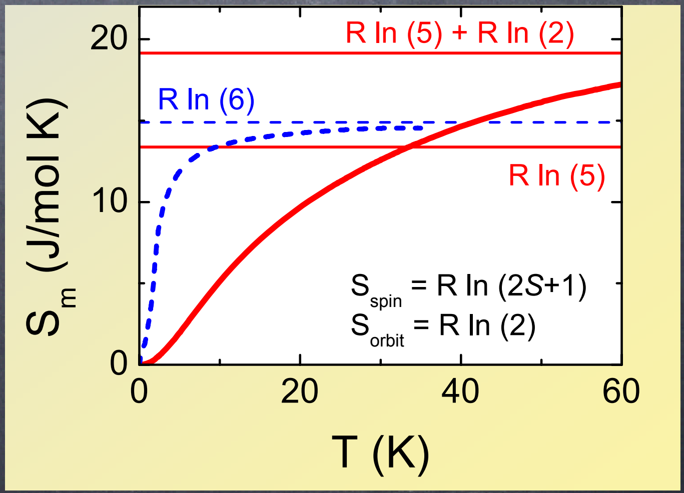


V. Fritsch et al. PRL **92**, 116401 (2004); N. Tristan et al. PRB **72**, 174404 (2005); T. Suzuki *et al*. (2006)

Orbital degeneracy in $FeSc_2S_4$

- Chemistry:
  - Fe^{2+}: $3d^6$
  - 1 hole in e_g level
- Spin S=2
- Orbital pseudospin 1/2
- Static Jahn-Teller does not appear





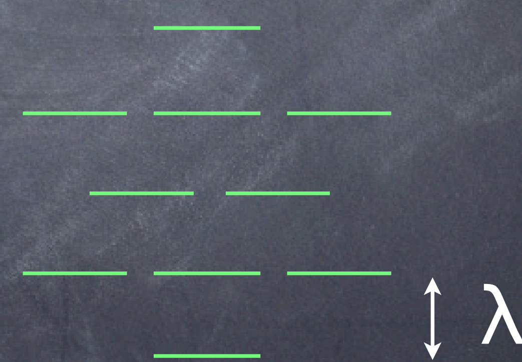
Atomic Spin Orbit

- Separate orbital and spin degeneracy can be split!

$$H_{SO} = -\lambda\left(\frac{1}{\sqrt{3}}\tau^x\left[(S^x)^2-(S^y)^2\right] + \tau^z\left[(S^z)^2 - \frac{S(S+1)}{3}\right]\right)$$

- Energy spectrum: singlet GS with gap = λ
- Microscopically,

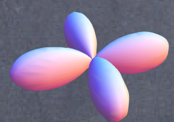
$$\lambda = \frac{6\lambda_0^2}{\Delta}$$



- Naive estimate λ ≈ 25K
  - should be reduced by dynamic JT

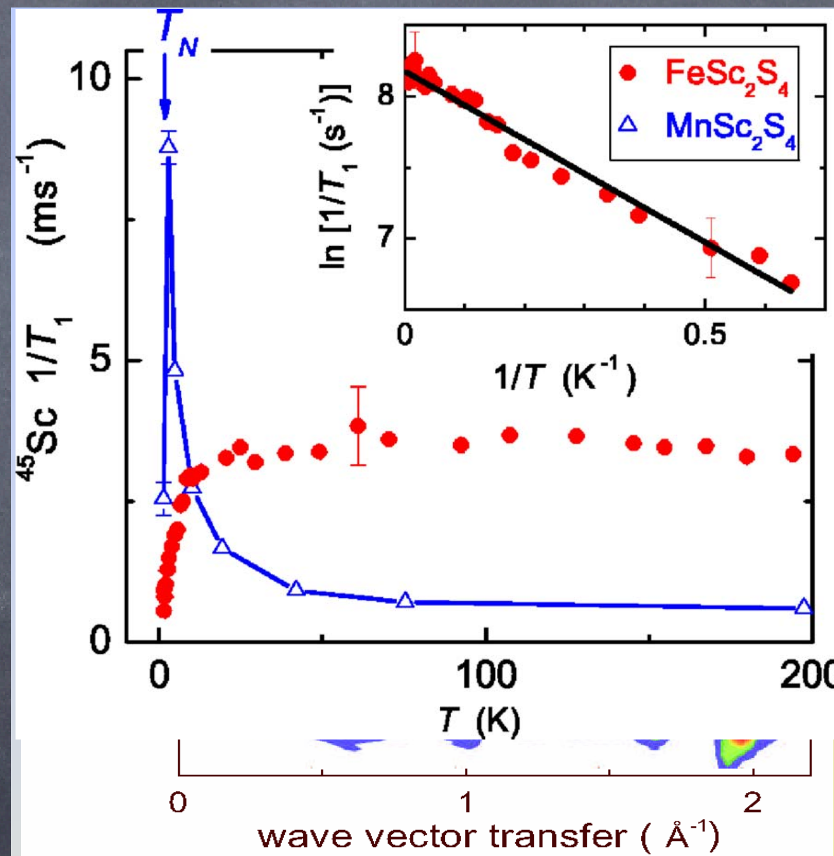
Spin orbital singlet

- Ground state of λ>0 term:

$|$  $\rangle|S^z{=}0\rangle - \frac{1}{\sqrt{2}}\,|$  $\rangle\big(|S^z{=}2\rangle + |S^z{=}{-2}\rangle\big)$

Exchange

- Inelastic neutrons show significant dispersion indicating exchange
- Bandwidth ≈ 20K similar order as Θ_{CW} and estimated λ
- Gap (?) 1-2K
  - Small gap is classic indicator of incipient order



A. Krimmel et al., Phys. Rev. Lett. 94, 237402, 2005

N. Büttgen et al., PRB 73, 132409 (2006)

Exchange

- Most general symmetry-allowed form of exchange coupling (neglecting SOI)

$$H_{ex} = \frac{1}{2}\sum_{ij}\Big\{J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j + K_{ij}\boldsymbol{\tau}_i\cdot\boldsymbol{\tau}_j + \tilde{K}_{ij}\tau_i^y\tau_j^y$$

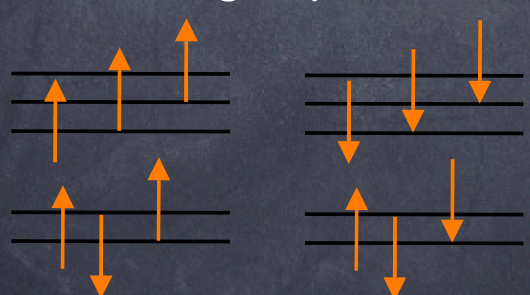
$$+\Big[L_{ij}\boldsymbol{\tau}_i\cdot\boldsymbol{\tau}_j + \tilde{L}_{ij}\tau_i^y\tau_j^y\Big]\mathbf{S}_i\cdot\mathbf{S}_j\Big\}$$

Exchange

- Neglecting SOI, a simplified superexchange calculation gives

$$H_{ex} = \frac{1}{2}\sum_{ij}\left\{J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j + K_{ij}\left(4+\mathbf{S}_i\cdot\mathbf{S}_j\right)\left(1+4\boldsymbol{\tau}_i\cdot\boldsymbol{\tau}_j\right)\right\}$$

- Largest coupling is AF spin interaction $\mathbf{S}_i\cdot\mathbf{S}_j$
  - More exchange processes

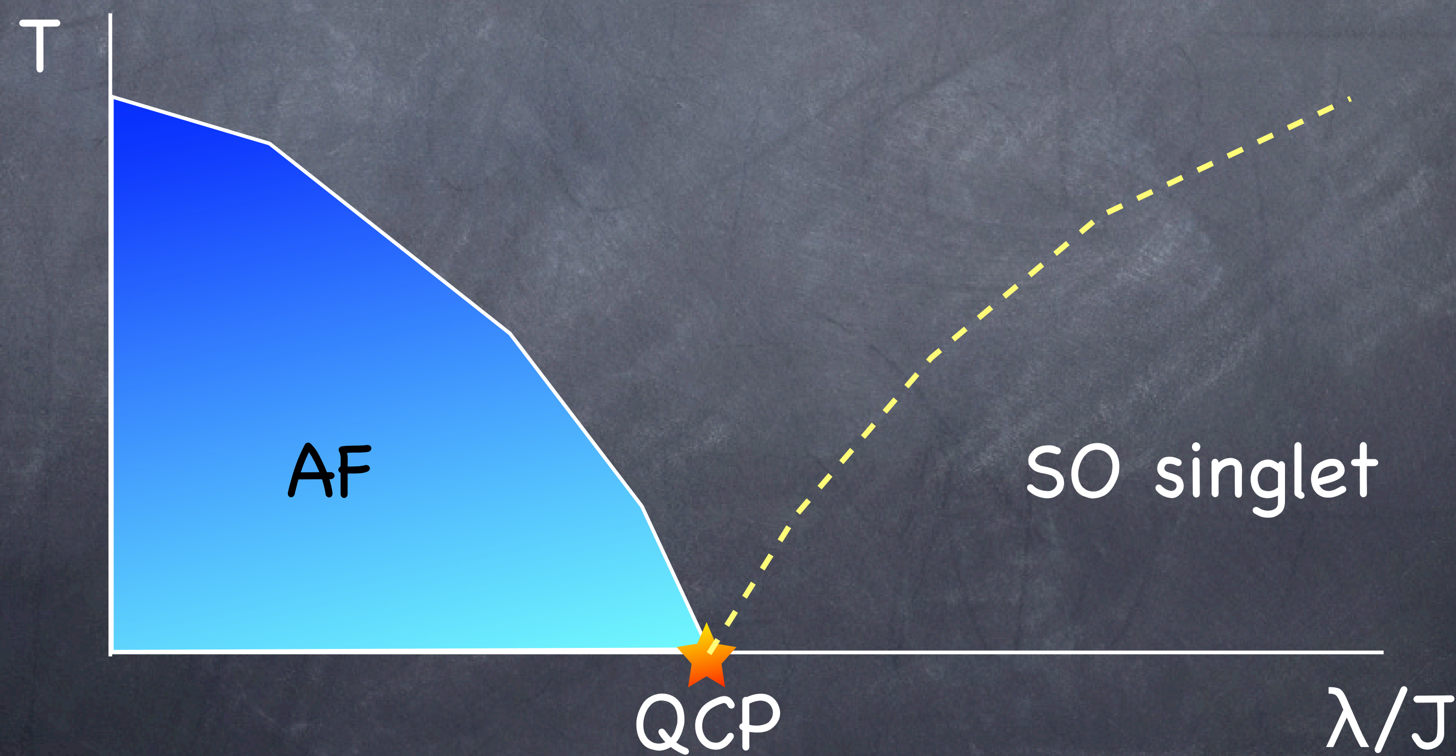


Ordered Phase (J >> λ)

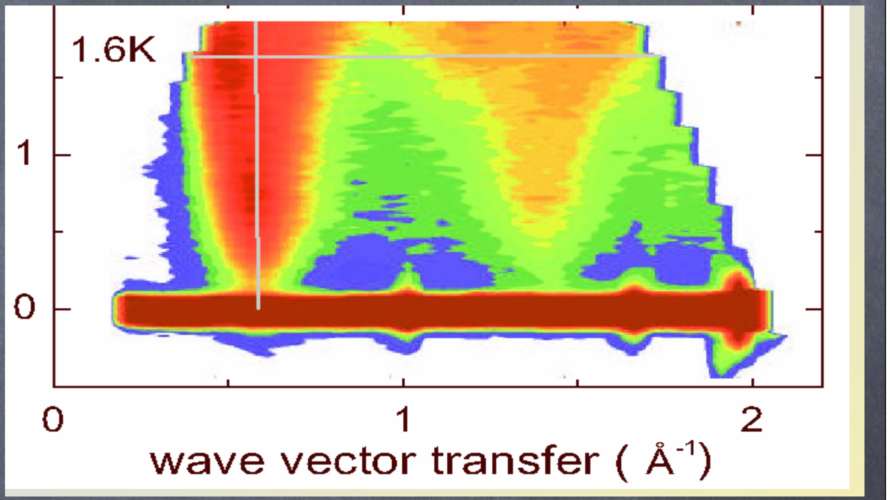
- Ground state of H_{ex} is almost certainly ordered
  - $\mathbf{S}_i \cdot \mathbf{S}_j$ coupling is strongest
  - Complex multi-spiral ground states possible
- Inclusion of weak SOI λ favors simpler commensurate “cubic” spin arrangements
  - spin order leads to induced orbital order

Quantum Critical Point

- Full Hamiltonian $H = H_{SO} + H_{ex}$



Minimal Model

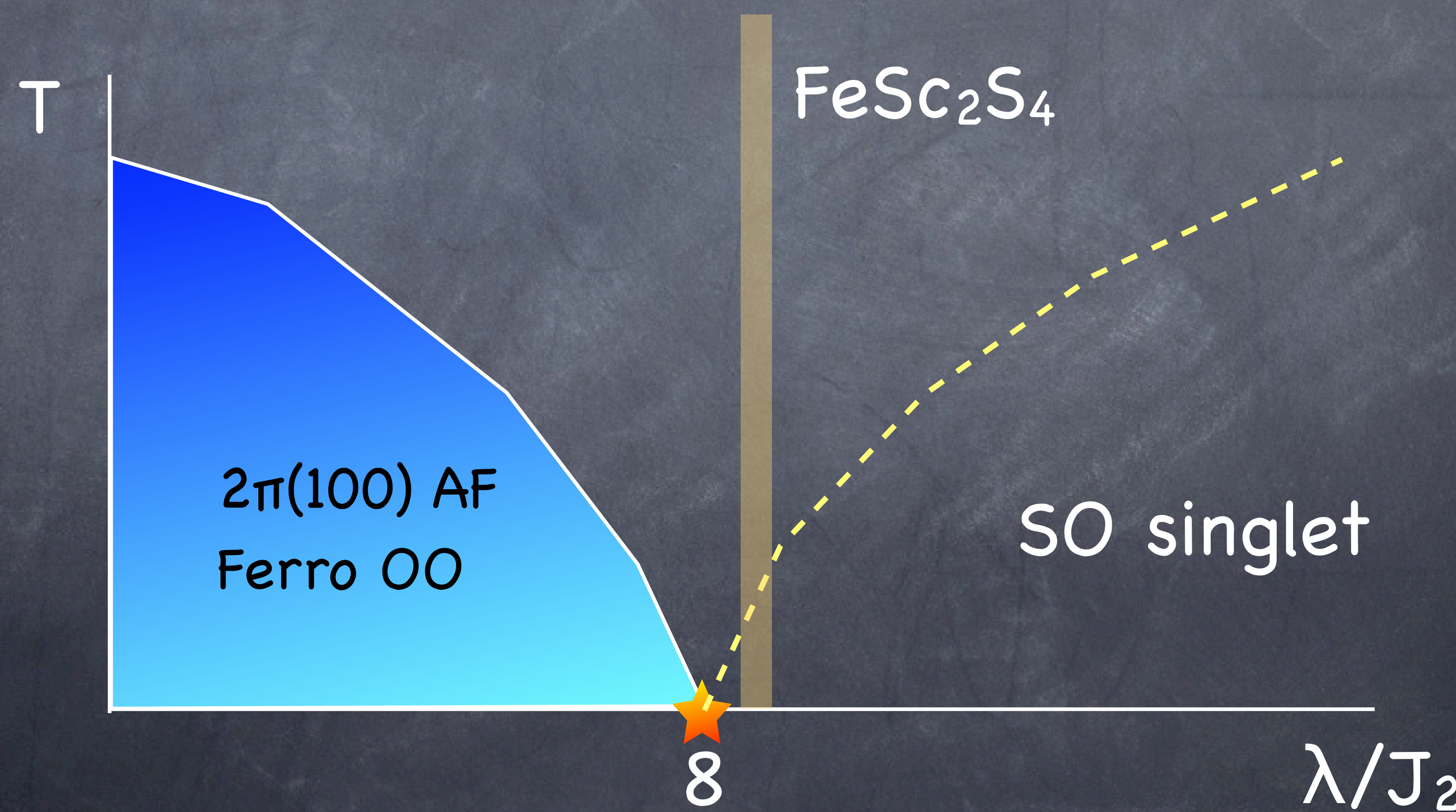


- Neutron scattering suggests peak close to 2π(100)
- Indicates $J_2 >> J_1$

$$H_{min} = J_2 \sum_{\langle\langle ij \rangle\rangle} \mathbf{S}_i \cdot \langle \mathbf{S}_j \rangle + H_{SO}$$

Quantum Critical Point

- Mean field phase diagram



Consequences of QCP

- Power-law spin correlations
- Scaling form for $(T_1T)^{-1} \sim T\, f(\Delta/T)$
- Specific heat $C_v \sim T^3\, f(\Delta/T)$
- Possibility of pressure-induced ordering
- Impurity effects?
- Behavior in field? Can triplet be made to condense?

To Do List

- Effects of J_1 on QCP
  - transitions to incommensurate states?
- Phonons and Jahn-Teller effects
- More in-depth study of phases
  - Higher order expansion about J=0
  - Spin-wave corrections for ordered states
- Effects of exchange anisotropy

Summary

- Rich physics in A-site antiferromagnetic spinels arises from "tunable frustration" due to complex exchange paths
  - We need single crystal neutron studies of spiral spin liquid!
- Orbital degeneracy and spin orbit provides an exciting route to quantum paramagnetism an quantum criticality
  - entangled spin-orbital singlet ground state in an S=2 magnet!