Frustrated Magnets (3)
Materials Survey

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Boulder summer school, 2008

The David and Lucile Packard Foundation

Recent Collaborators

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- Oleg Starykh
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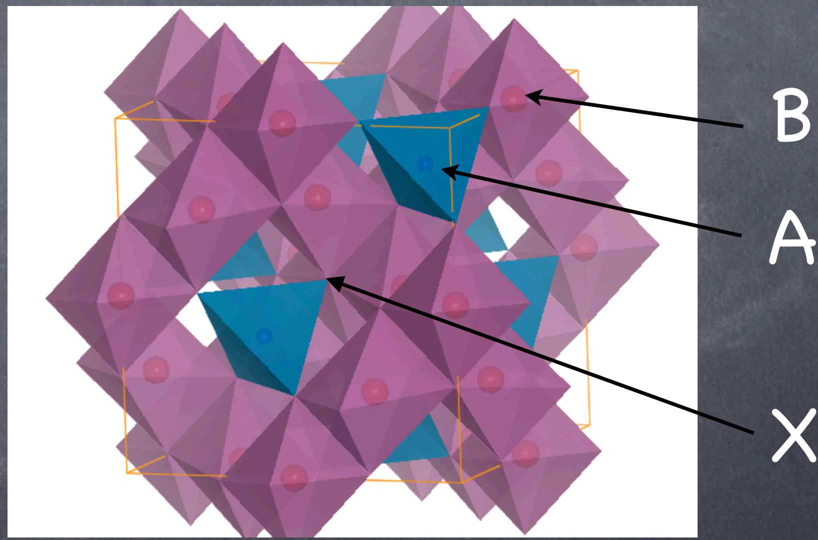
What do we look for?

- Is it an insulator?
- Is it a magnet? Curie law
- Signs of frustration
  - f>>1:
    - Θ_{CW} (χ)
    - T_N: signs of transition in χ, C_v, ...
  - low T entropy, low energy excitations
    - C_v, $1/T_1$,...
- Identify the states
  - nature of correlations?
  - ordering if it occurs
- Compare with some theoretical expectations

AB_2X_4 spinels

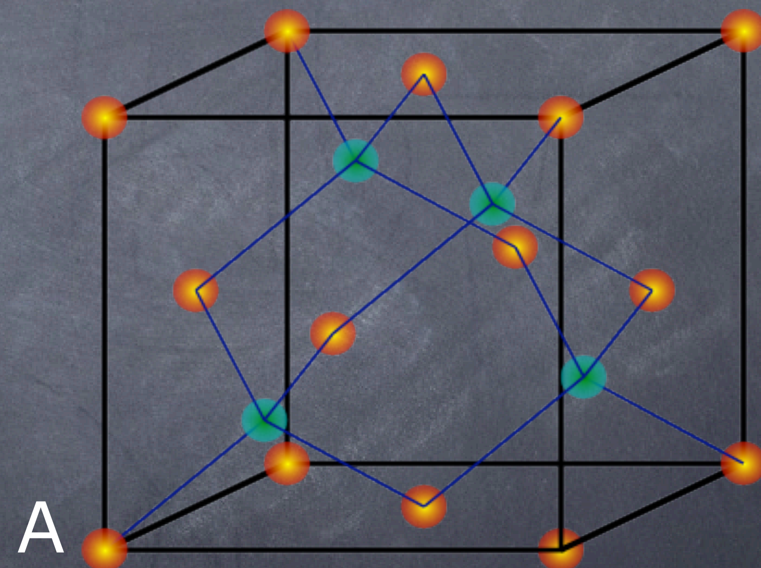
- One of the most common mineral structures
- Common valence:
  - A^{2+}, B^{3+}, X^{2-}
  - X=O,S,Se

cubic $Fd\bar{3}m$

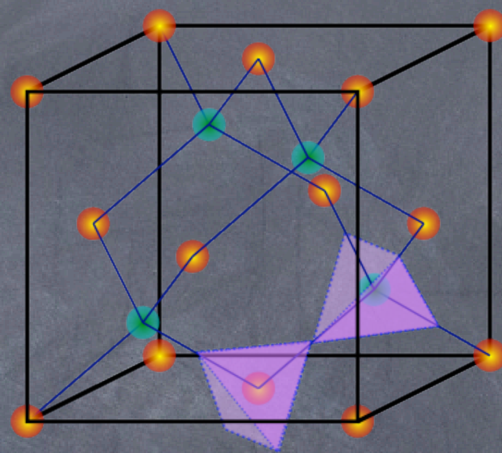


Deconstructing the spinel

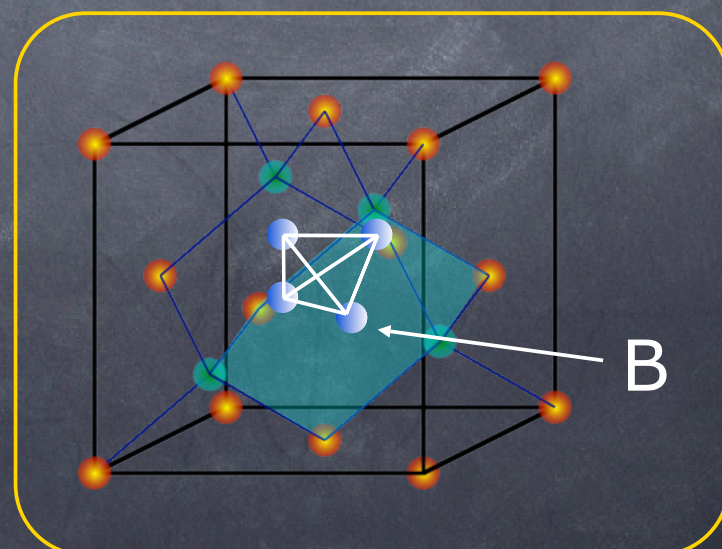
- A atoms: diamond lattice
  - Bipartite: not *geometrically* frustrated



Deconstructing the spinel

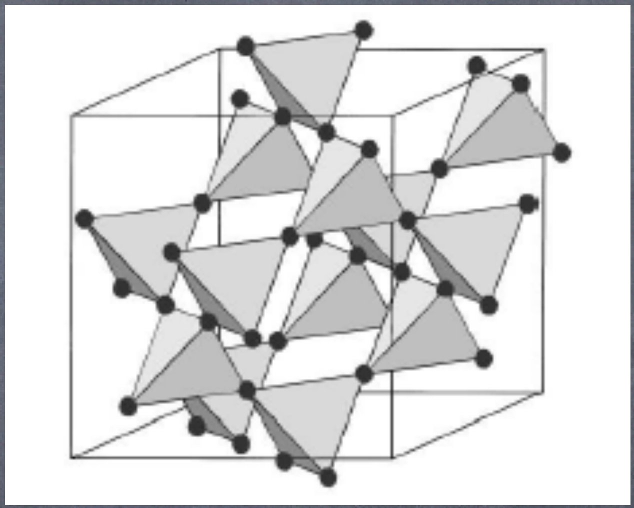


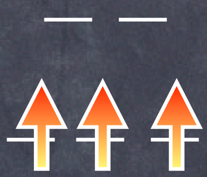
- B atoms: pyrochlore
  - decorate the plaquettes of the diamond lattice



ACr_2O_4 spinels

- pyrochlore lattice
- S=3/2 Isotropic moment
- X=O spinels: B-B distance close enough for direct overlap
  - dominant AF nearest-neighbor exchange



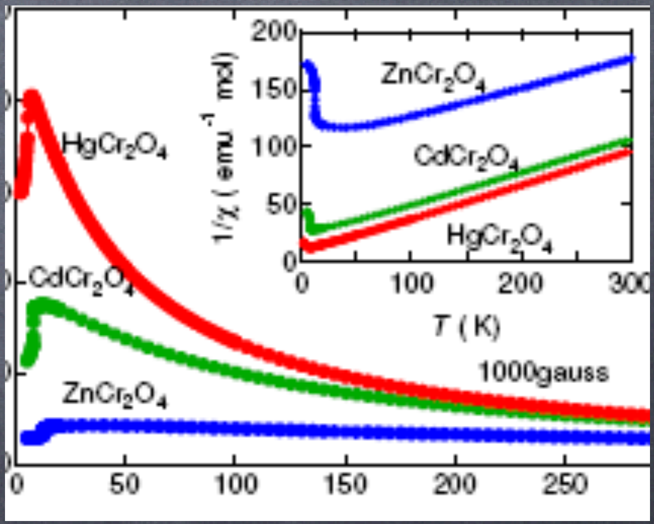


H=0 Susceptibility

- Frustration:

	Zn	Cd	Hg
Θ_{CW} (K)	-390	-70	-32
T_N (K)	12	7.8	5.8
f	33	9	6

χ



T

H. Ueda et al

Degeneracy

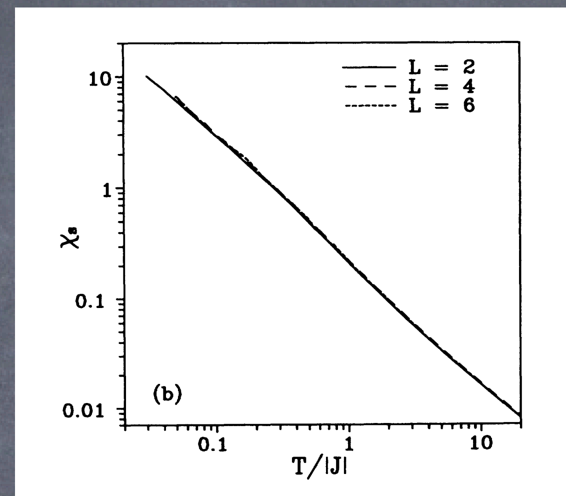
- Heisenberg model

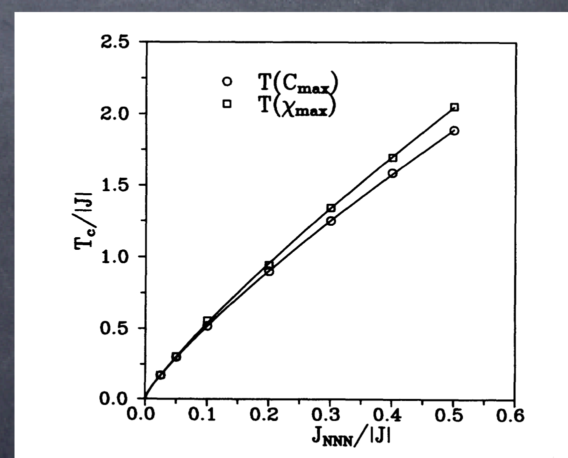
$$H = \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j = \frac{1}{2} \sum_t \left(\sum_{i \in t} \vec{S}_i \right)^2 + \text{const.}$$

- Ground state constraint: total spin 0 per tetrahedron
  - Quantum mechanically: not possible

Classical spin liquid

- No LRO (Reimers)

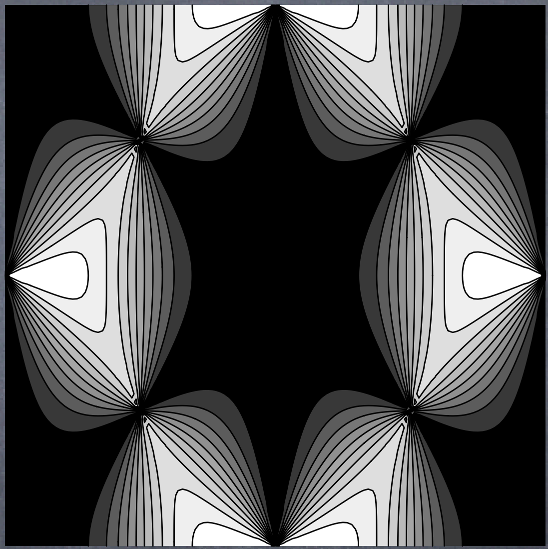




Classical spin liquid

- No LRO (Reimers)
- Dipolar correlations

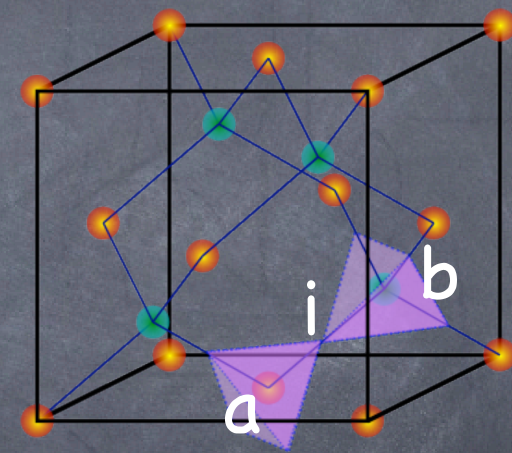
(Youngblood+Axe,Henley, Isakov et al...)



$$S_i^\mu = b_{ab}^\mu$$

Classical spin liquid

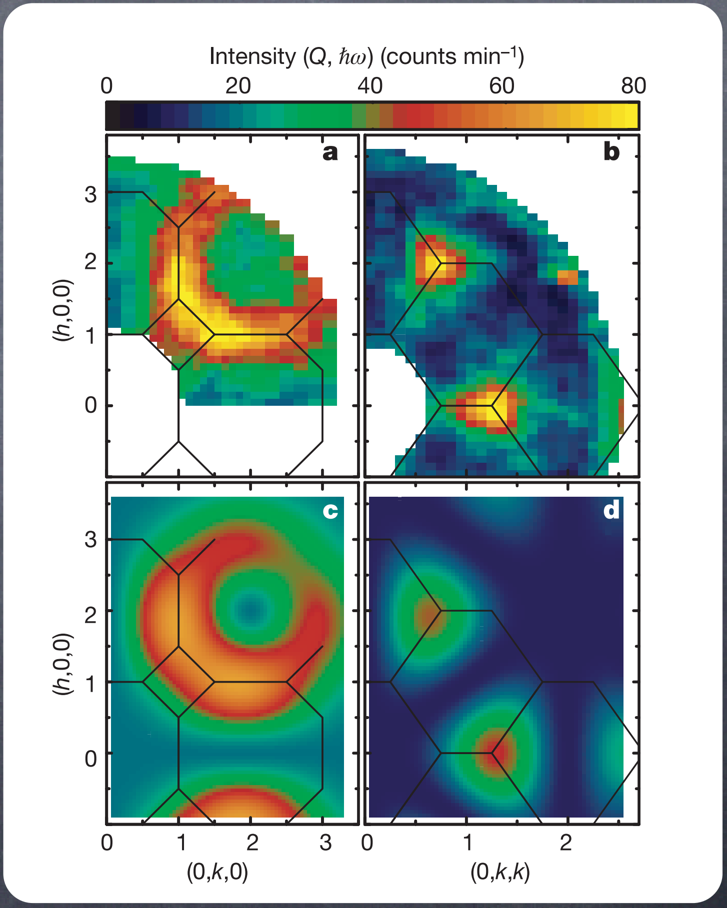
- No LRO (Reimers)
- Dipolar correlations



$$S_i^\mu = b_{ab}^\mu$$

Classical spin liquid

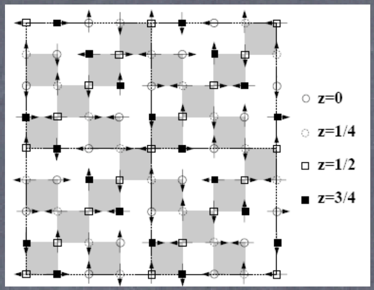
- Unusual "ring" correlations seen in $CdCr_2O_4$ related
- $Y_2Ru_2O_7$: J. van Duijn et al, 2007



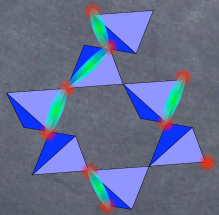
Broholm et al

Ordering

- Many perturbations important for ordering:
  - Spin-lattice coupling
  - Further exchange
  - Spin-orbit effects
  - Quantum corrections

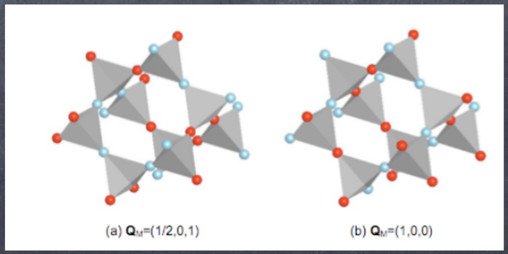


$ZnCr_2O_4$



$CdCr_2O_4$

JH Chung et al, 2005

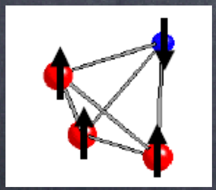


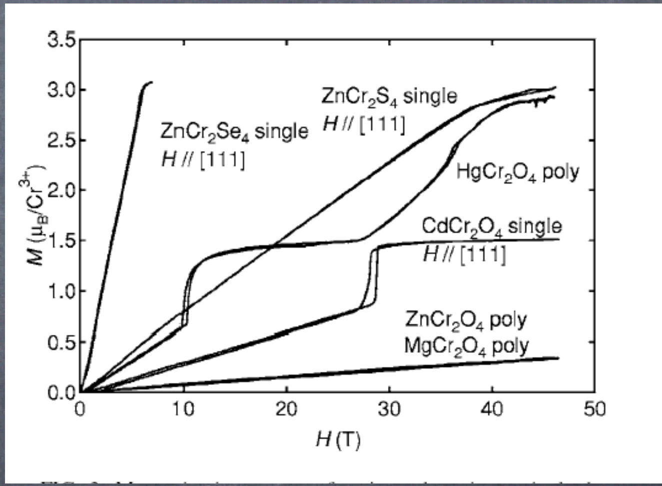
$HgCr_2O_4$

S.H. Lee + many others

Magnetization Plateaus

- Classically: $M = M_s H/H_s$
- Plateau indicates 3:1 structure

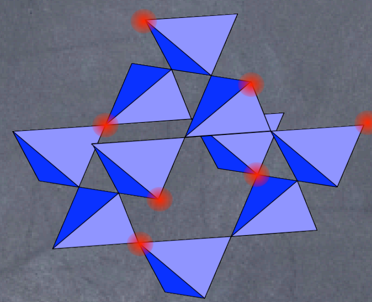




H. Ueda at al, 2005/6

Magnetization Plateaus

- Plateau mechanism:
  - spin-lattice coupling favors collinearity
- Order on plateau may be selected by
  - spin-lattice
  - quantum effects

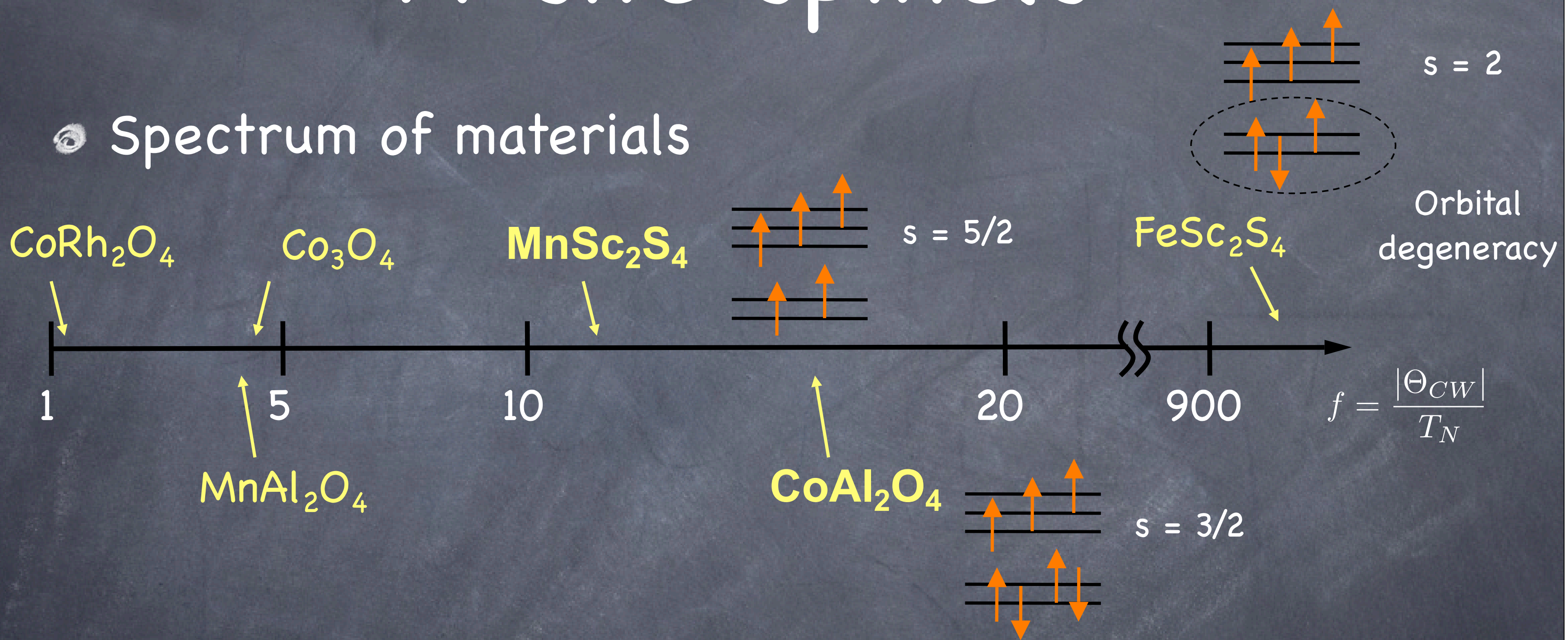


"R" state observed in neutrons

Matsuda et al

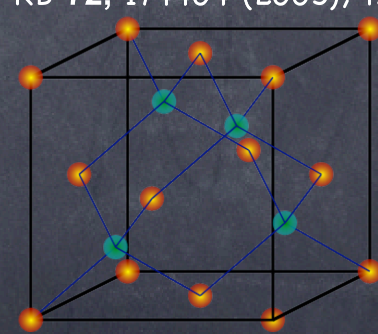
A-site spinels

- Spectrum of materials



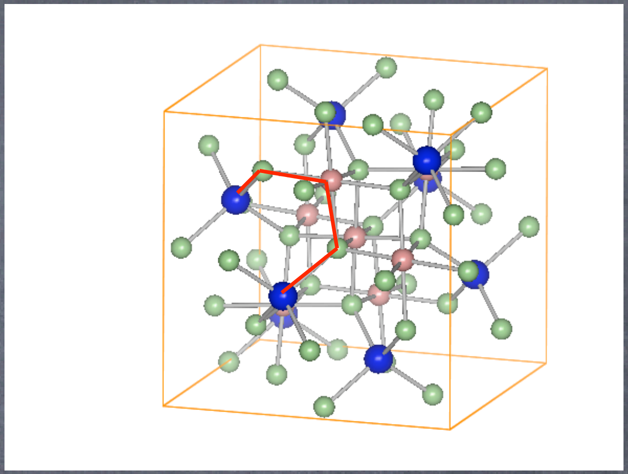
V. Fritsch et al. PRL **92**, 116401 (2004); N. Tristan et al. PRB **72**, 174404 (2005); T. Suzuki *et al.* (2006)

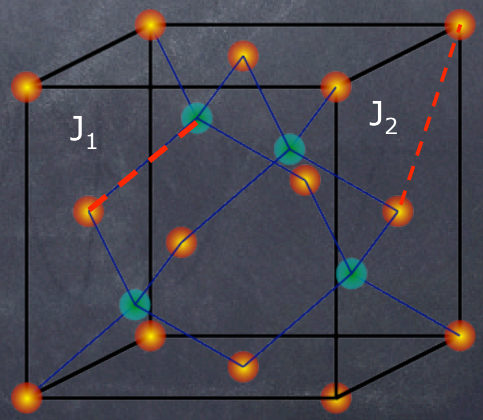
- Naively unfrustrated



Why frustration?

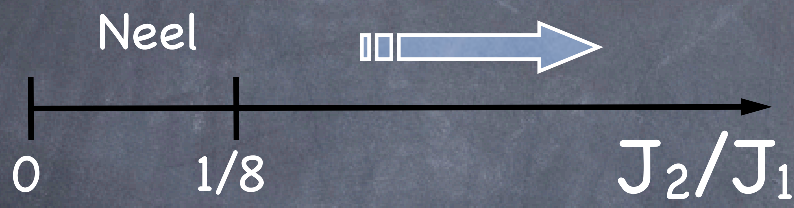
- Roth, 1964: 2nd and 3rd neighbor exchange not necessarily small
  - Exchange paths: A-X-B-X-B comparable
- Minimal model
  - J_1-J_2 exchange

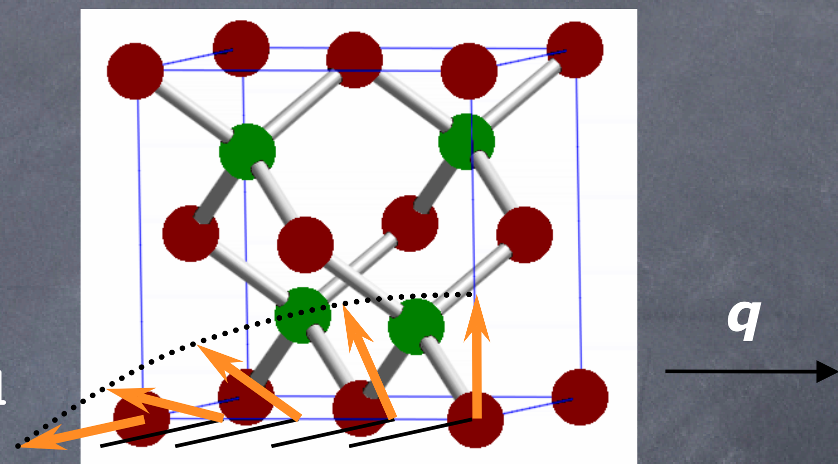




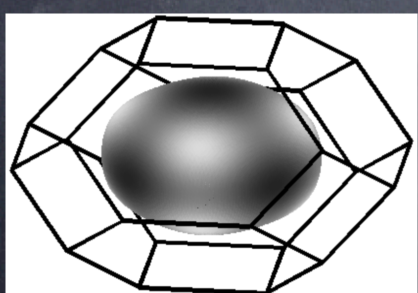
Ground state evolution

- Coplanar spirals

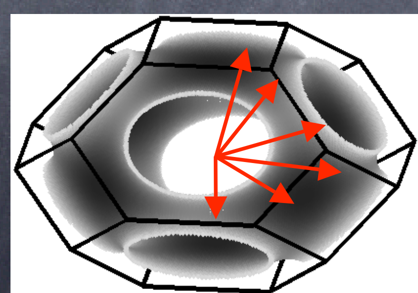




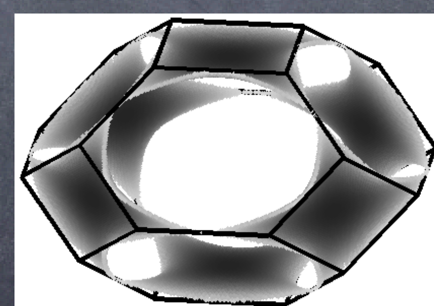
- Spiral surfaces:



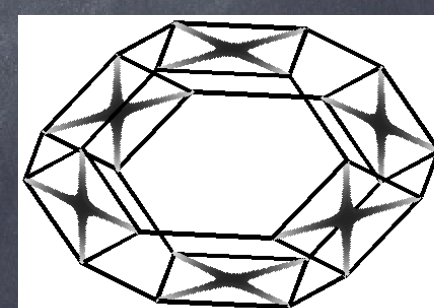
$J_2/J_1 = 0.2$



$J_2/J_1 = 0.4$



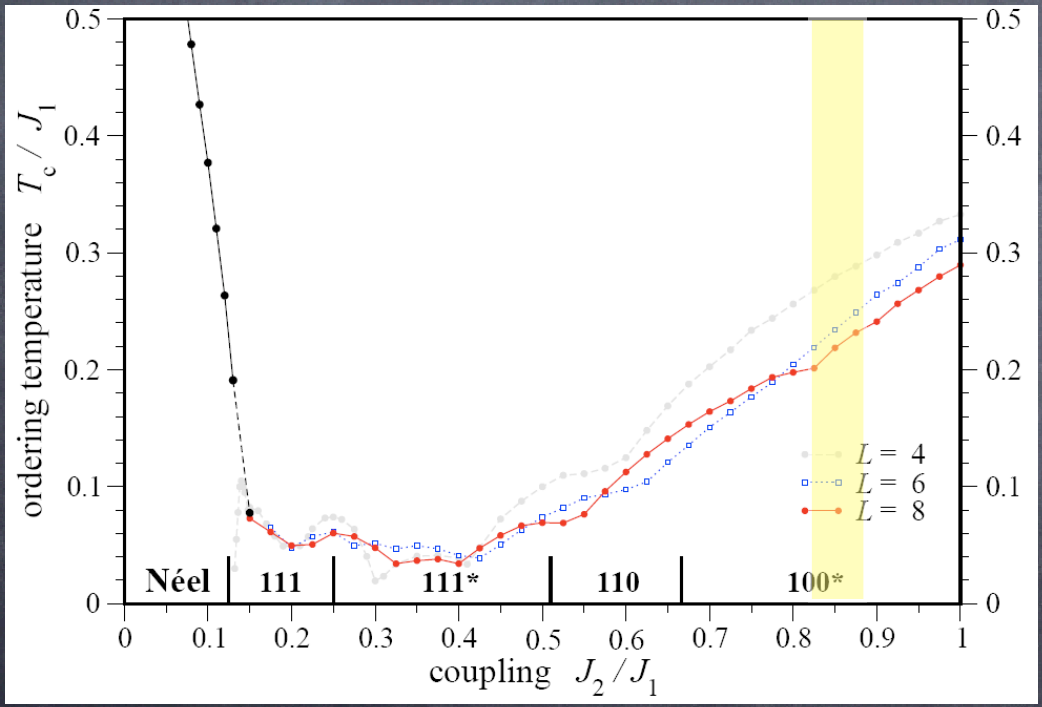
$J_2/J_1 = 0.85$



$J_2/J_1 = 20$

Monte Carlo

$MnSc_2S_4$

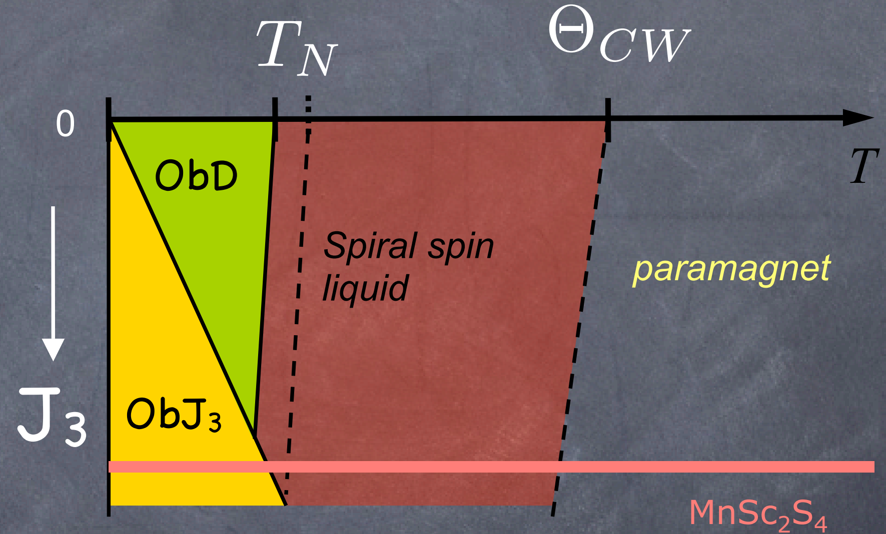


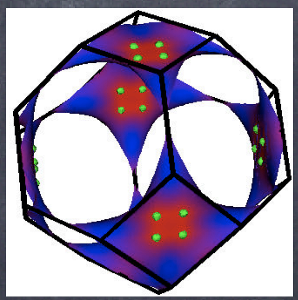
f = 11 at

$J_2/J_1 = 0.85$

Phase Diagram

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

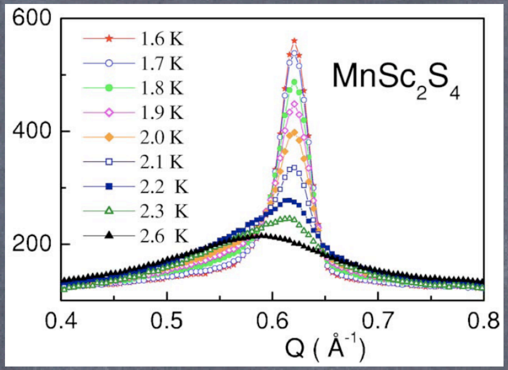


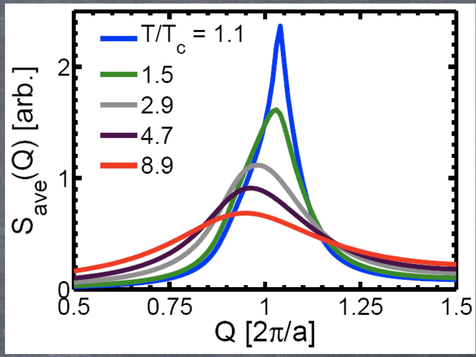


Comparison to Expt.

Expt. Theory

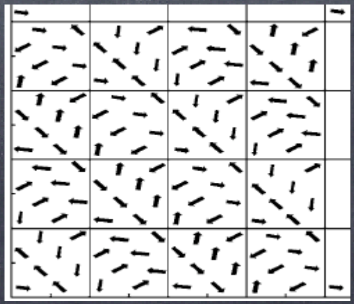
- Diffuse scattering





A. Krimmel et al, 2006

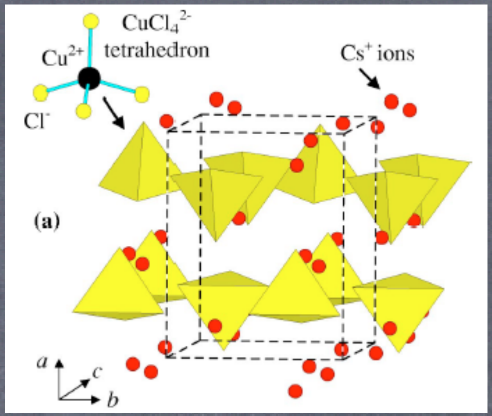
- Ordered state
  - (qq0) spiral
- Specific heat?



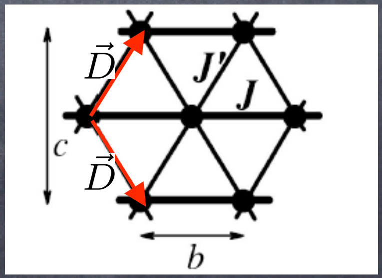
agrees with theory for FM J_1

Cs_2CuCl_4

- Spatially anisotropic triangular lattice
    - Cu^{2+} spin-1/2 spins



$$H = \frac{1}{2}\sum_{ij}\left[J_{ij}\vec{S}_i \cdot \vec{S}_j - \vec{D}_{ij} \cdot \vec{S}_i \times \vec{S}_j\right]$$



$$\vec{D} = D\hat{a}$$

- couplings:

J=0.37meV

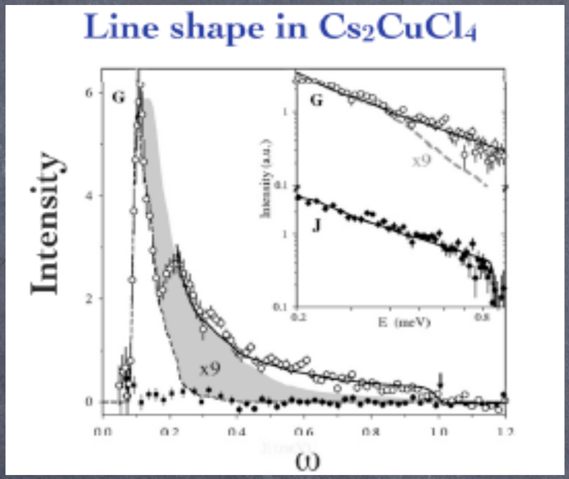
J'=0.3J

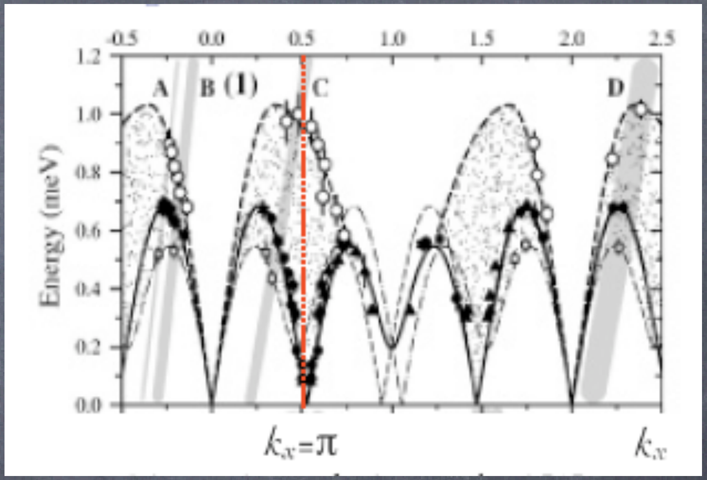
D=0.05J

R. Coldea et al

Neutron scattering

- Coldea et al, 2001/03: a 2d spin liquid?



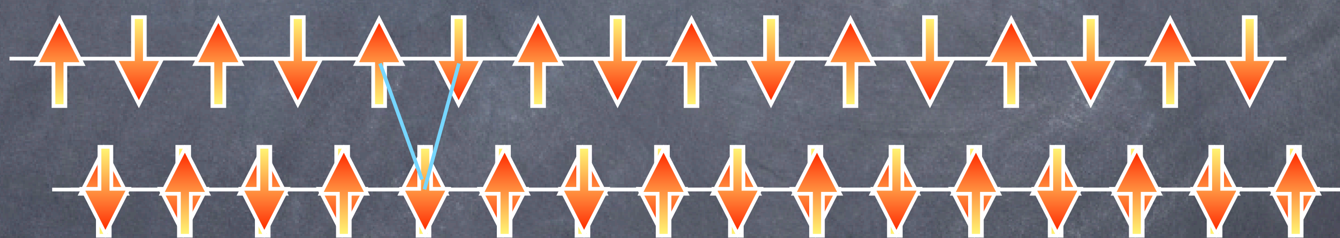


Very broad spectrum similar to 1d (in some directions of k space). Roughly fits power law.

Fit of "peak" dispersion to spin wave theory requires adjustment of J,J′ by 40% - in opposite directions!

Dimensional reduction?

- Frustration of interchain coupling makes it less "relevant"
  - First order energy correction vanishes

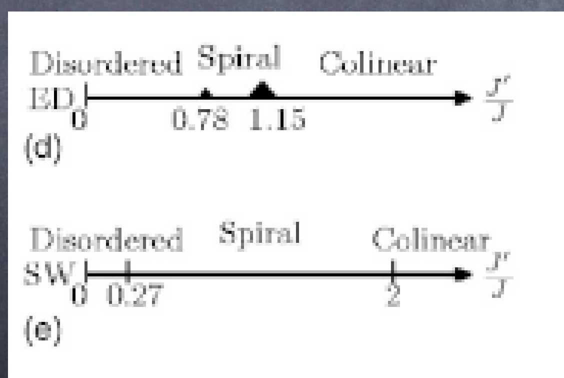


  - Leading effects are in fact $O[(J')^4/J^3]$!

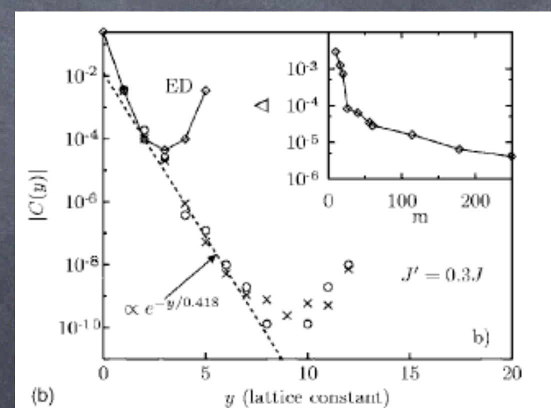
Dimensional reduction?

- Frustration of interchain coupling makes it less "relevant"
  - First order energy correction vanishes.
  - Numerics: J'/J < 0.7 is "weak"

Weng et al, 2006



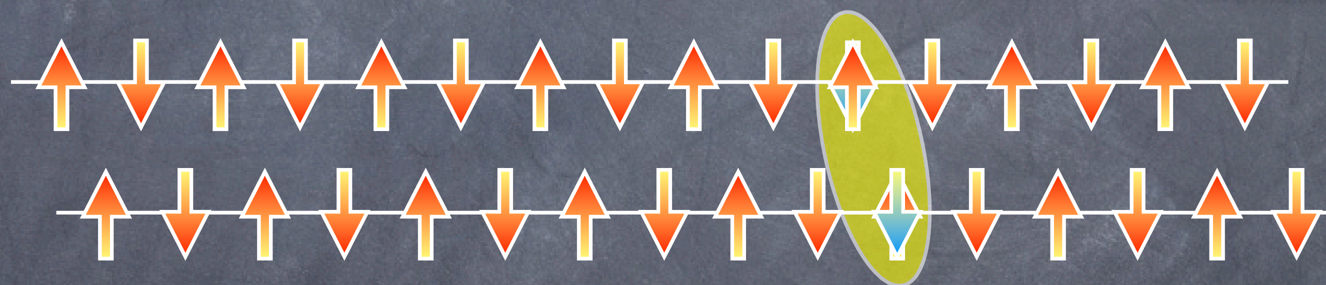
Very different from spin wave theory



Very weak inter-chain correlations

Excitations

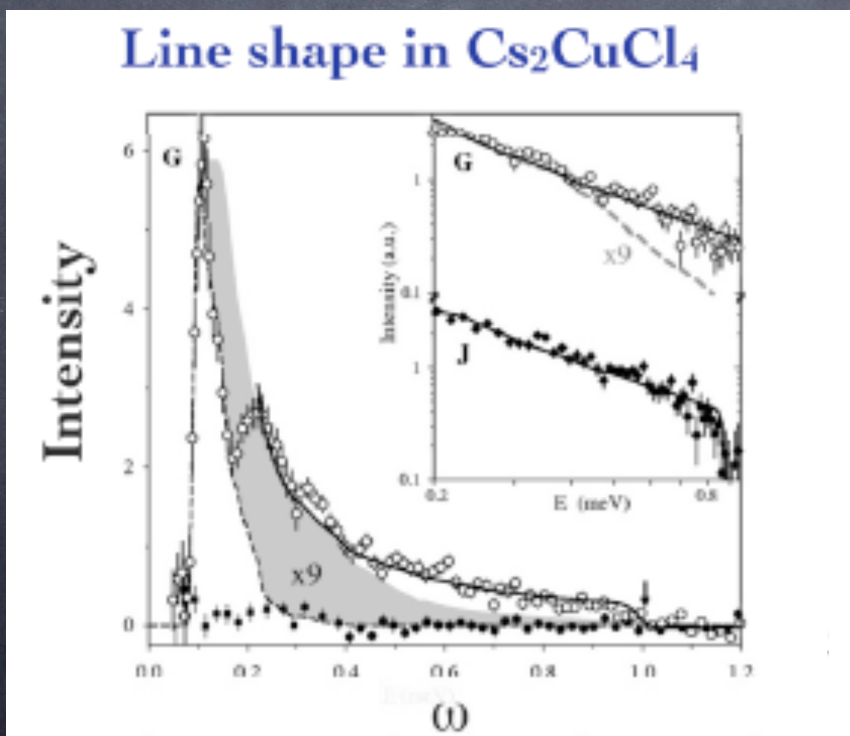
- Build 2d excitations from 1d spinons
  - Exchange: $\frac{J'}{2}\left(S_i^+ S_j^- + S_i^- S_j^+\right)$

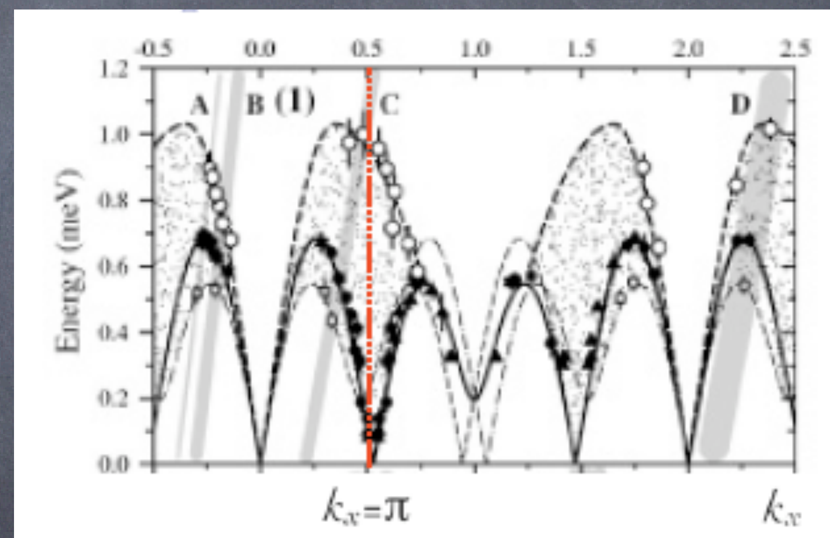


- Expect spinon binding to lower inter-chain kinetic energy
  - Use 2-spinon Schroedinger equation

Broad lineshape: "free spinons"

- "Power law" fits well to free spinon result
  - Fit determines normalization

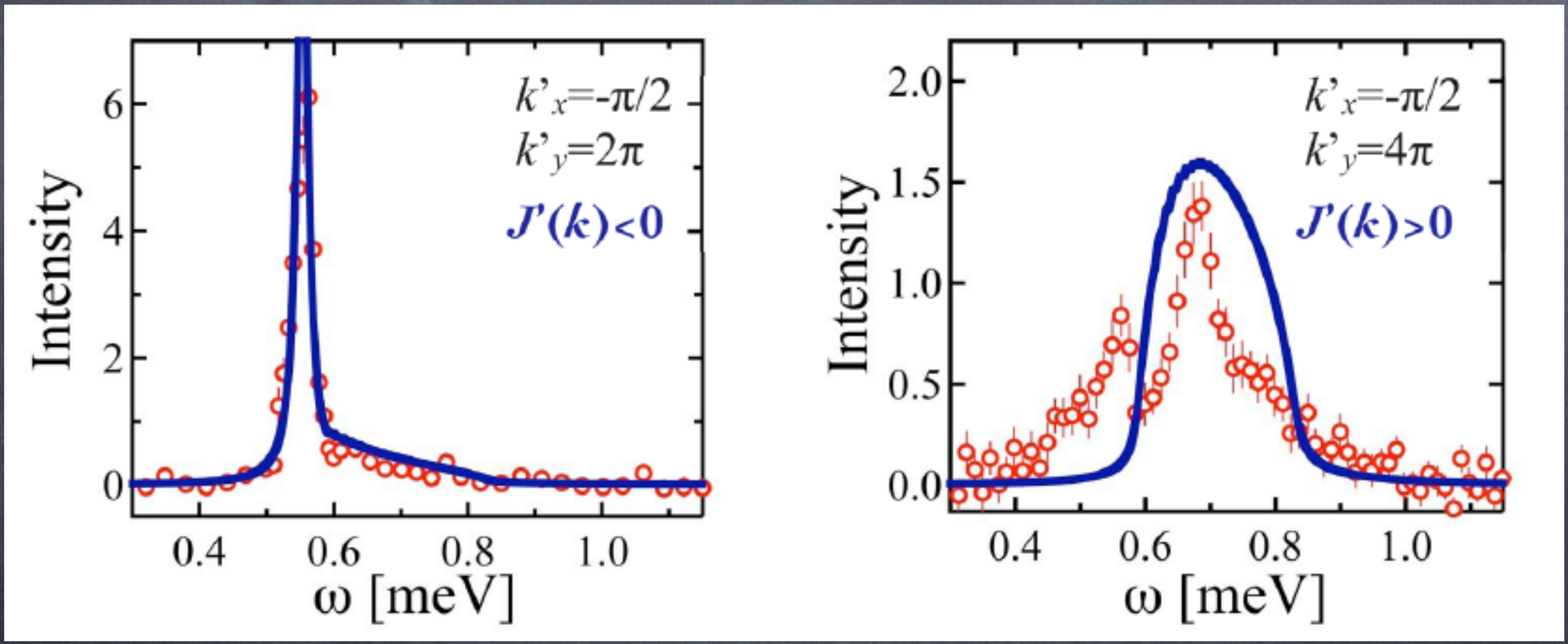




J'(k)=0 here

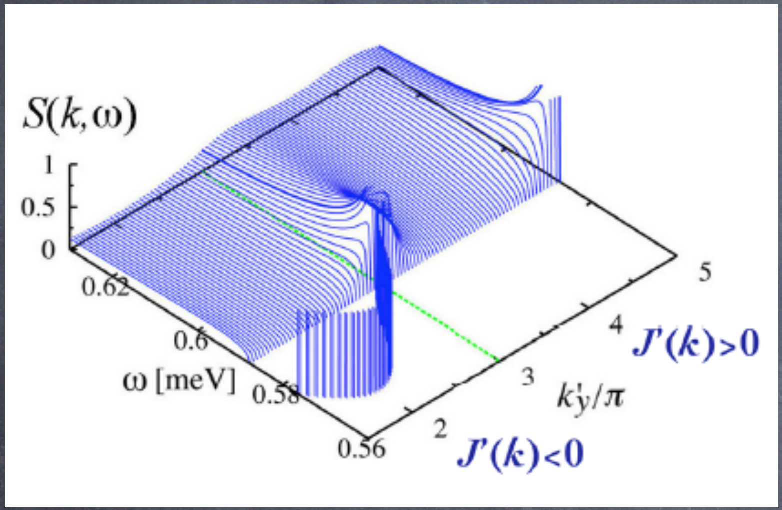
Bound state

- Compare spectra at J'(k)<0 and J'(k)>0:

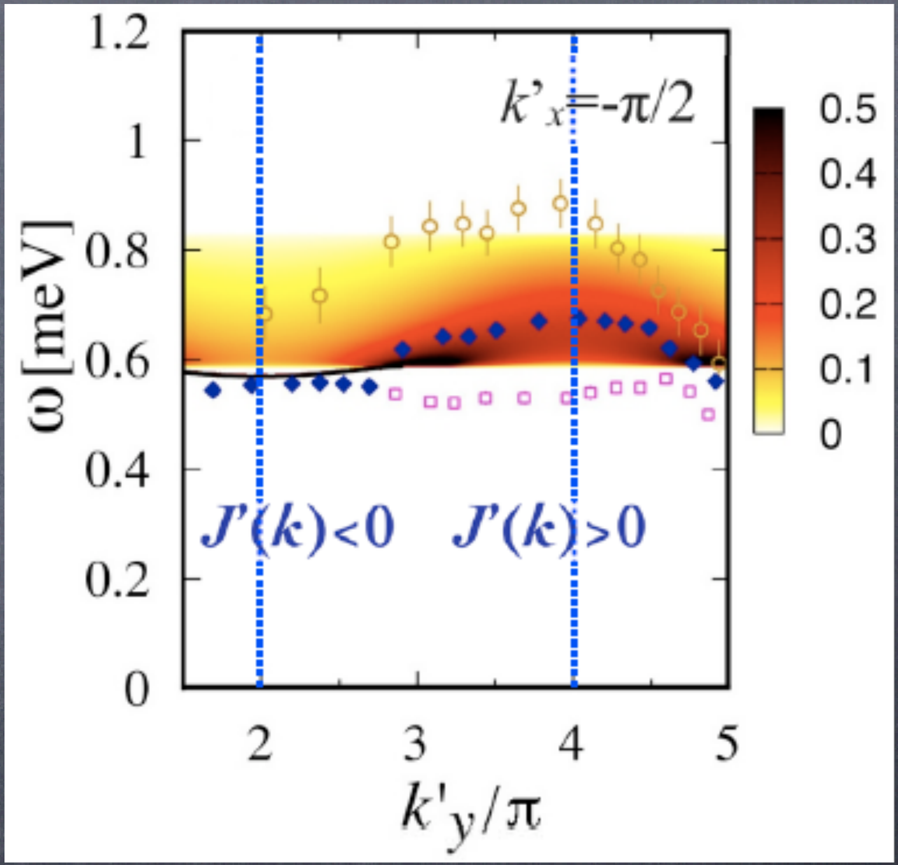


- Curves: 2-spinon theory w/ experimental resolution
- Curves: 4-spinon RPA w/ experimental resolution

Transverse dispersion

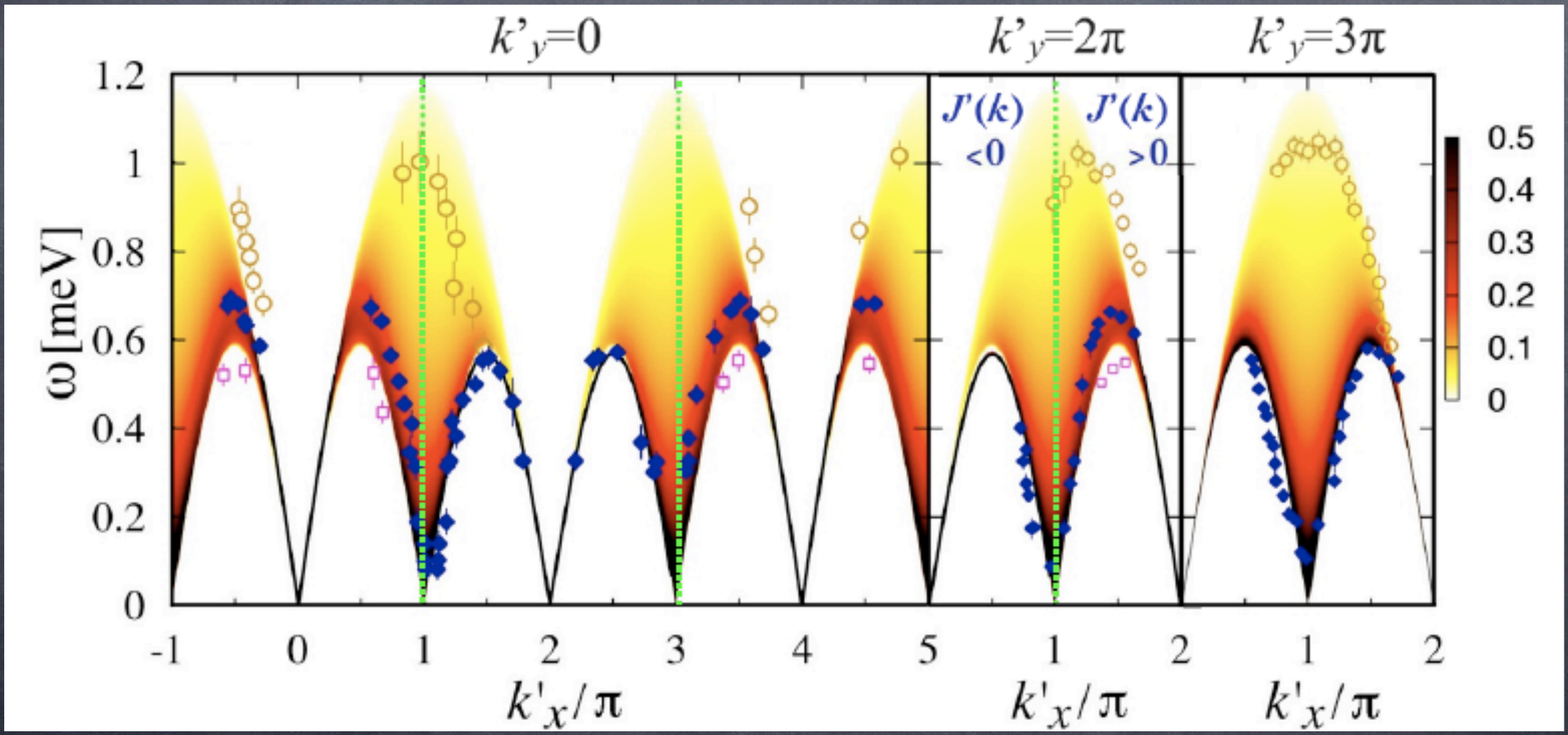


Bound state and resonance



Solid symbols: experiment
Note peak (blue diamonds) coincides with bottom edge only for J'(k)<0

Spectral asymmetry

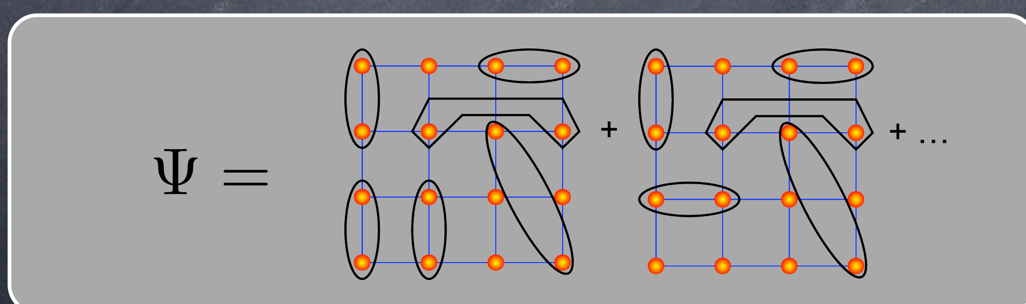


- Vertical lines: J’(k)=0.

Quantum Spin Liquids

Ultimate frustration?

- Can quantum fluctuations prevent order even at T=0: f=∞?
- Many theoretical suggestions since Anderson (73)
  - "Resonating Valence Bond" QSL states



Search for QSLs

- Where do we look?
  - Spin-1/2 frustrated magnets
  - Intermediate correlation regime (near the Mott transition)

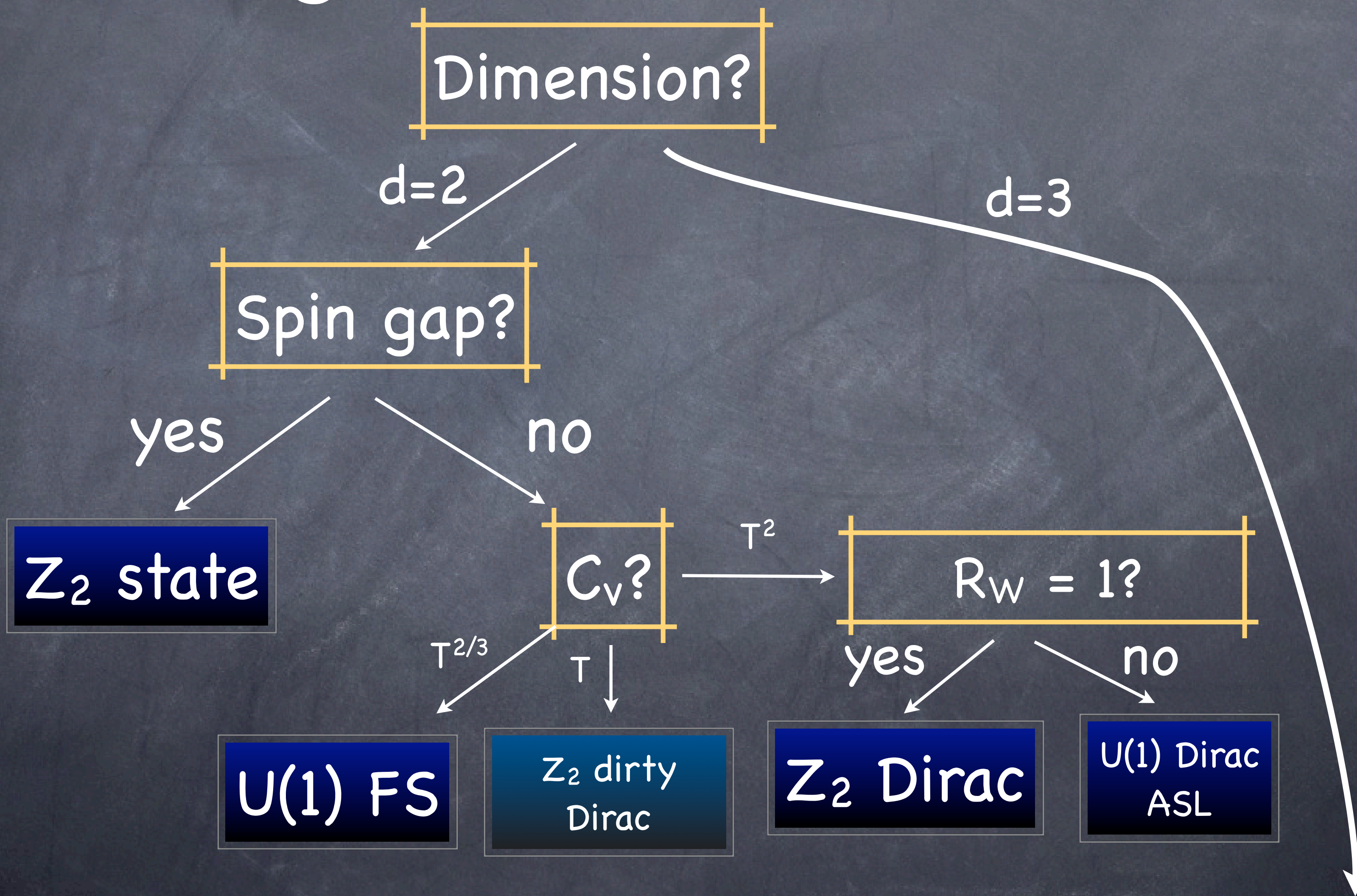
Search for QSLs

- $1/f=T_c=0$: no ordering (magnetic or otherwise!)
- No spin freezing (hysteresis, NMR, μSR)
- Structure of low energy excitations
  - $\chi(T)$, $C_v(T)$, $1/T_1$, inelastic neutrons
  - theoretical guidance helpful!
- Smoking gun?

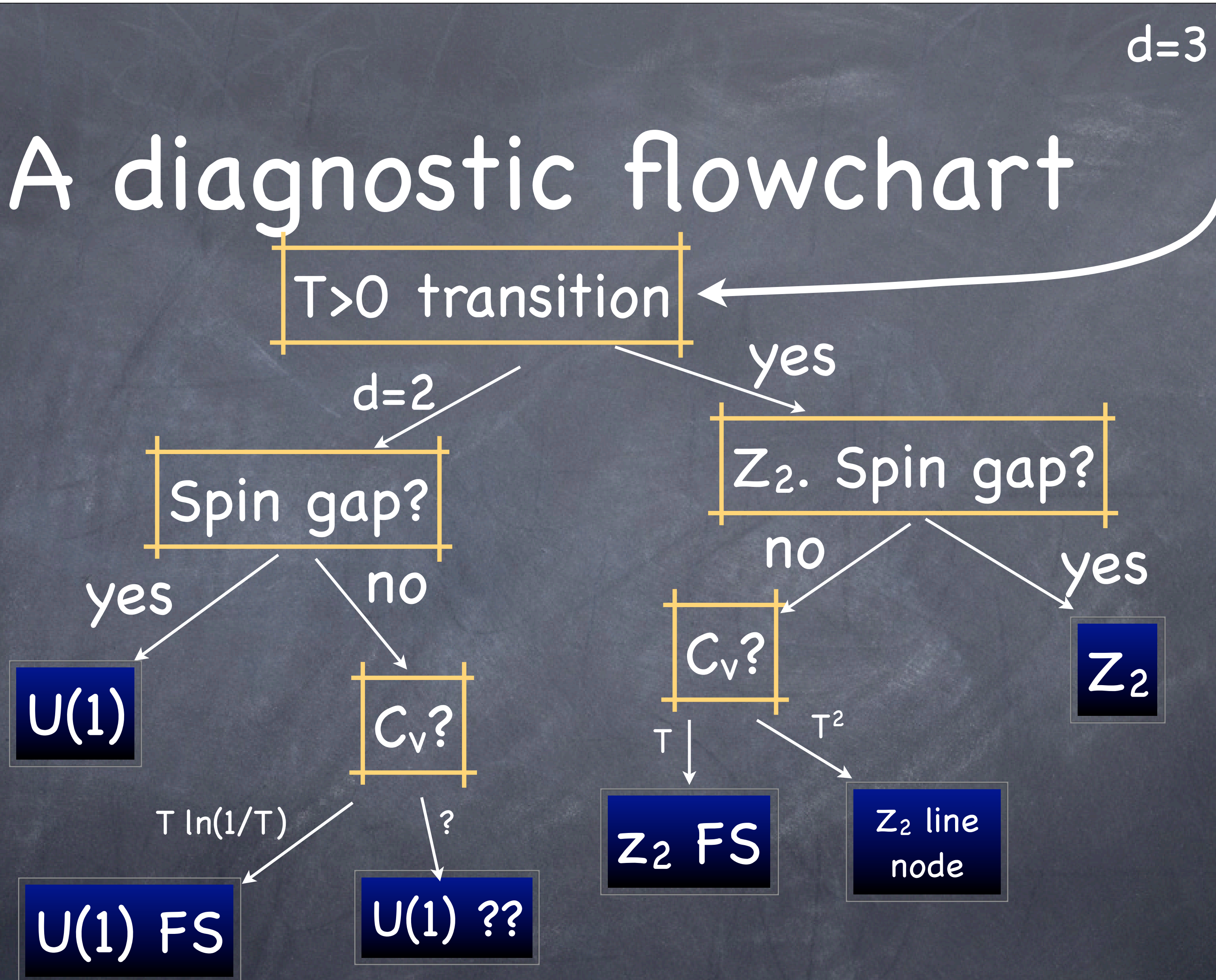
QSL Family Tree

- U(1) states
  - spinons unpaired
  - strong gauge fluctuations
  - spinons must be gapless in d=2
  - stable in d=3 at T=0 only
- Z_2 states
  - spinons paired
  - weak gauge fluctuations
  - stable in d=2
  - T>0 Ising transition in d=3

A diagnostic flowchart



A diagnostic flowchart

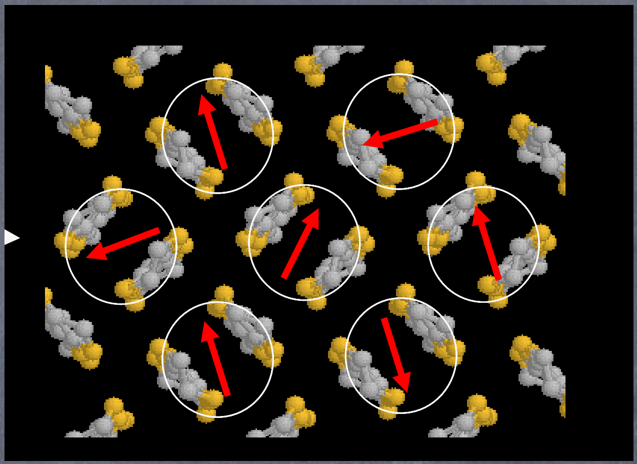


disordered possibilities neglected

QSL candidates

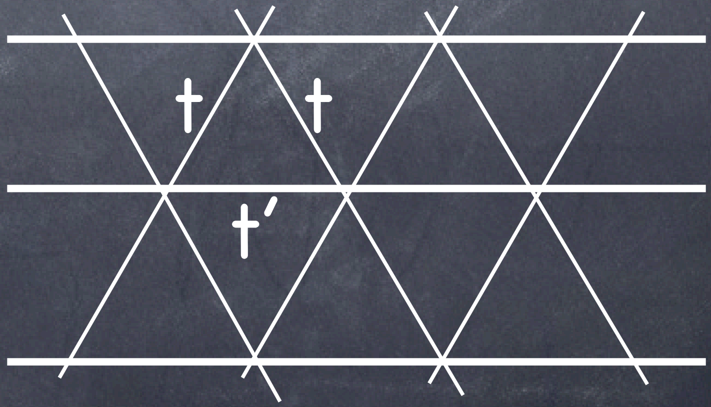
- ~~$NiGa_2S_4$ - spin 1 triangular lattice~~
- κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ - triangular lattice organic
- $EtMe_3Sb[Pd(dmit)_2]_2$ - triangular lattice organic
- $Na_4Ir_3O_8$ - hyperkagome
- $ZnCu_3(OH)_6Cl_2$ - kagome

κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$



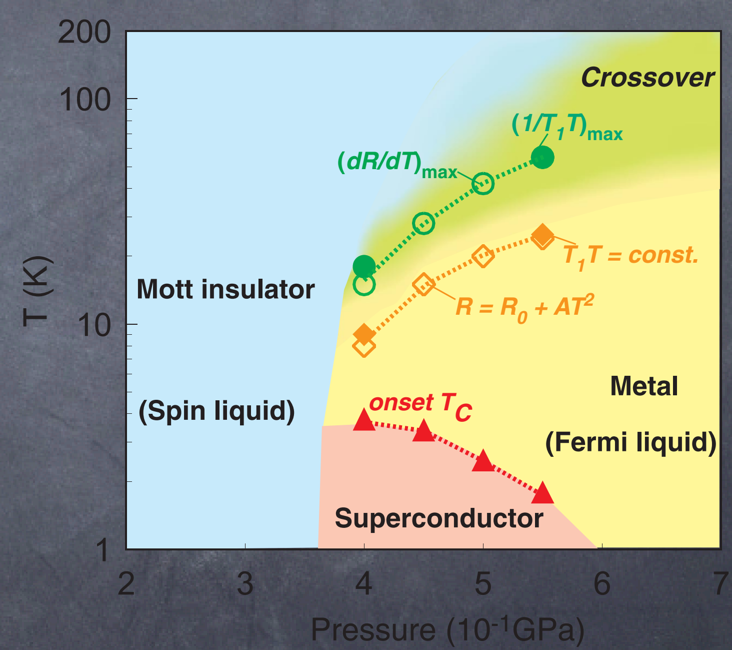
- Organic
- S=1/2 triangular lattice
- Nearly isotropic Hubbard-like with $t'/t = 1.06$

K. Kanoda group



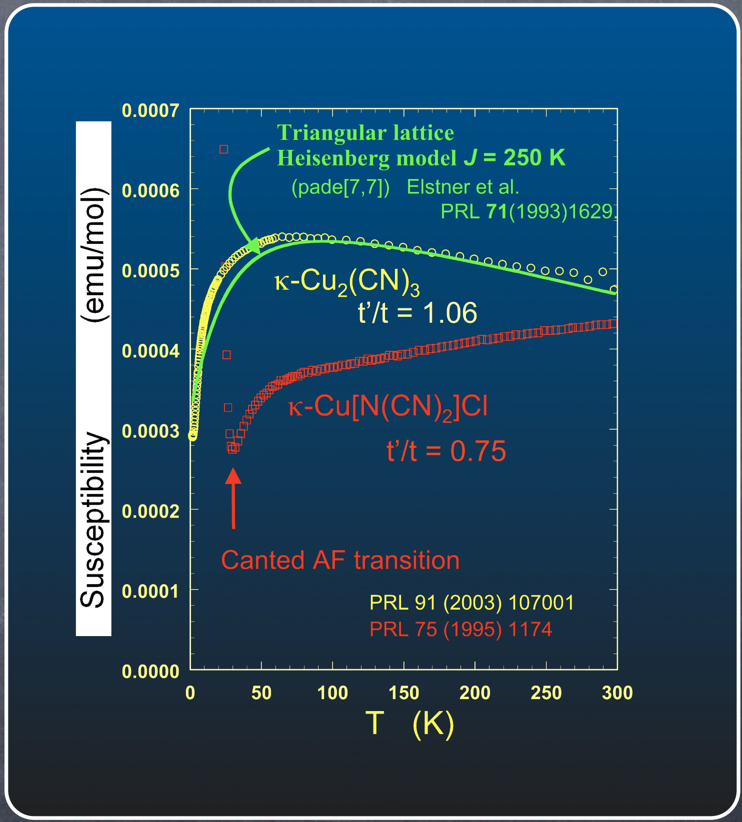
κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

- Material is proximate to a Mott transition
- Non-activated transport
- Optical pseudogap



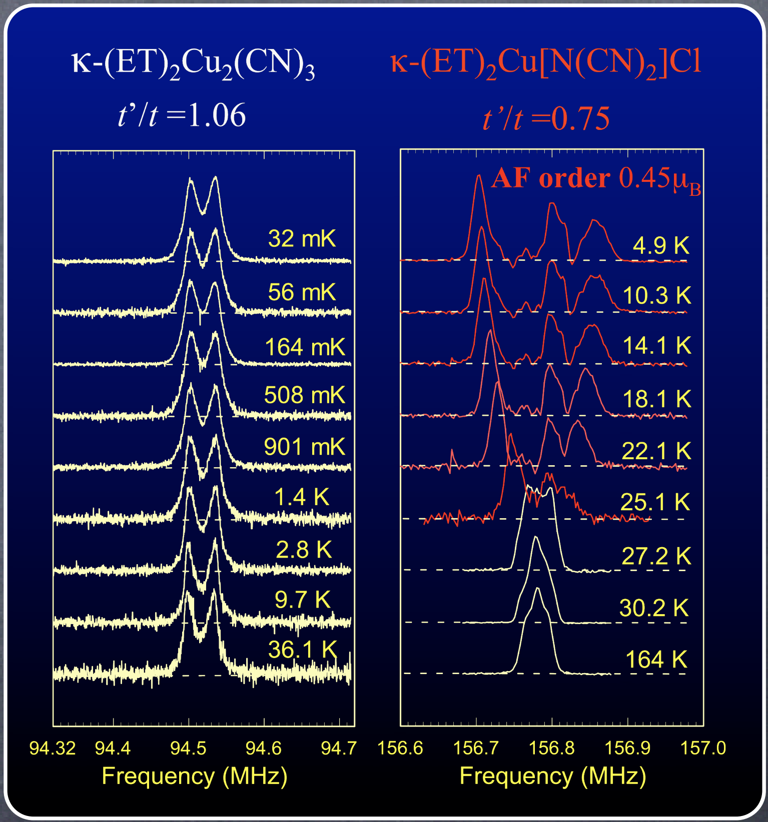
κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

- Susceptibility similar to Heisenberg triangular lattice
- χ(T=0) finite
- No ordering



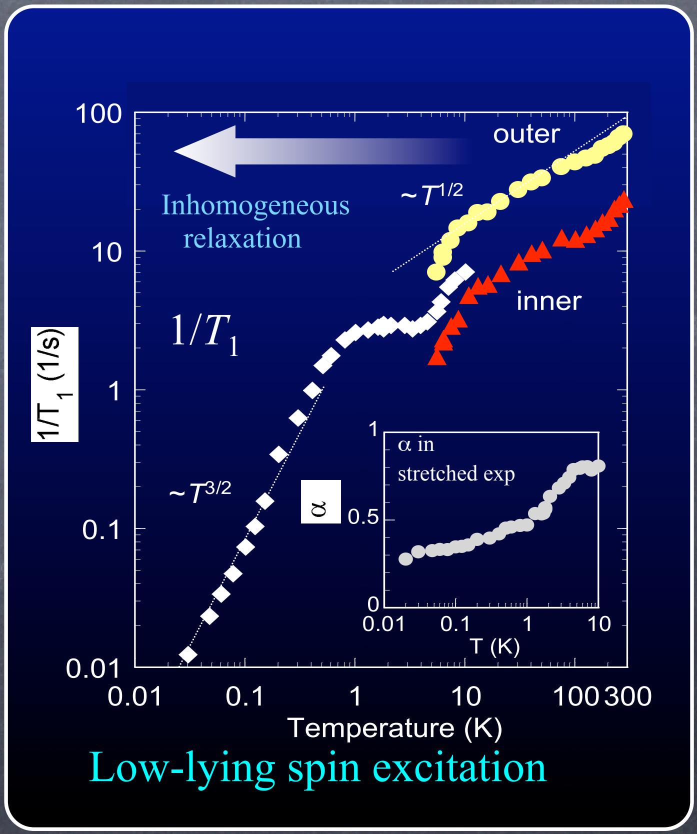
κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

- No ^{1}H NMR line splitting down to 32 mK - no internal fields
- No ordering



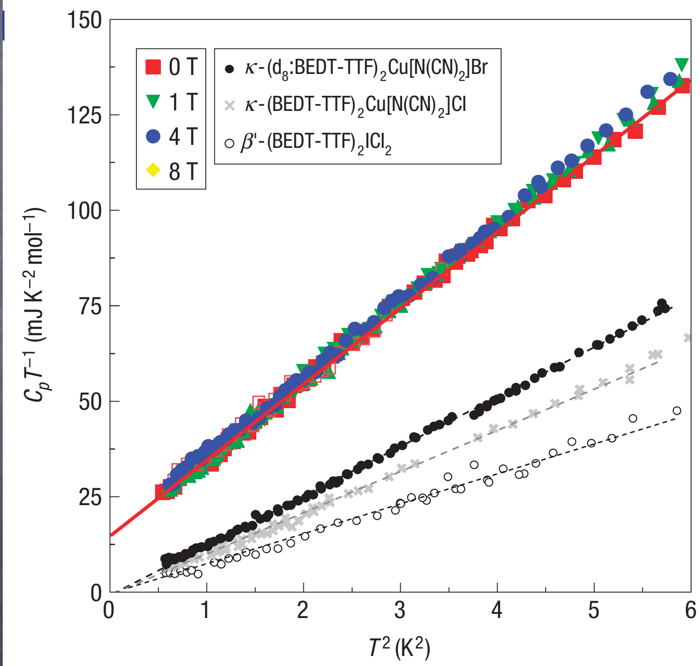
κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

- 1/T_1 relaxation rate power law at low temperature indicating gapless excitations



κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

- Linear specific heat
  - γ=15mJ/K^2 mol
  - field independent
- Wilson ratio T χ/γ is O(1)



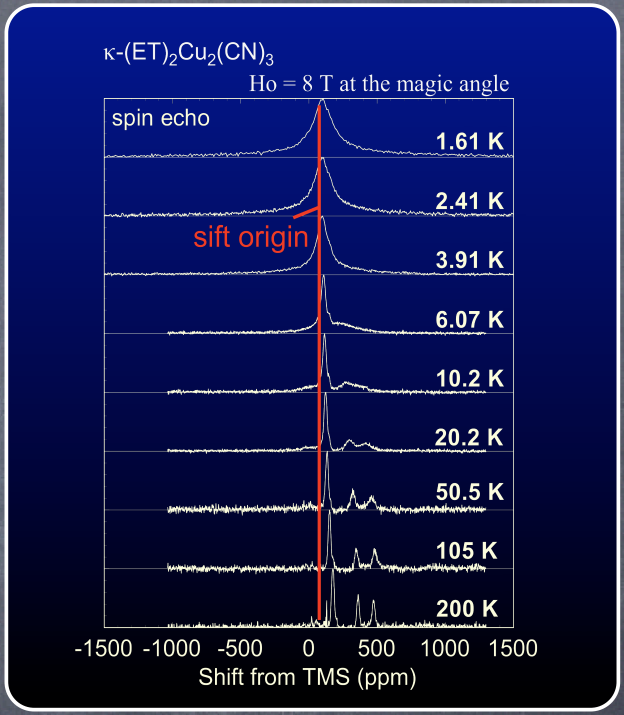
Interpretation?

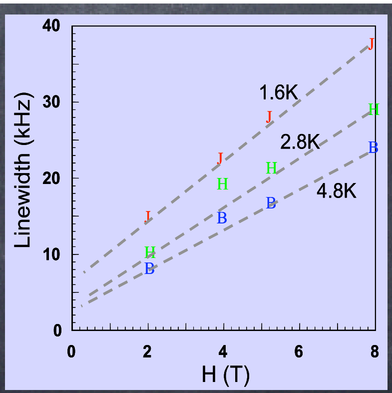
- Theoretical suggestion (Motrunich) - U(1) spin liquid with spinon Fermi surface
  - Good variational energy for triangular lattice Hubbard model
  - Large susceptibility ✔
  - Linear specific heat ☹
    - theory predicts $C_v = AT^{2/3}$
  - Spinon pairing?
    - features visible around T=5K. related?

SS Lee et al

κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

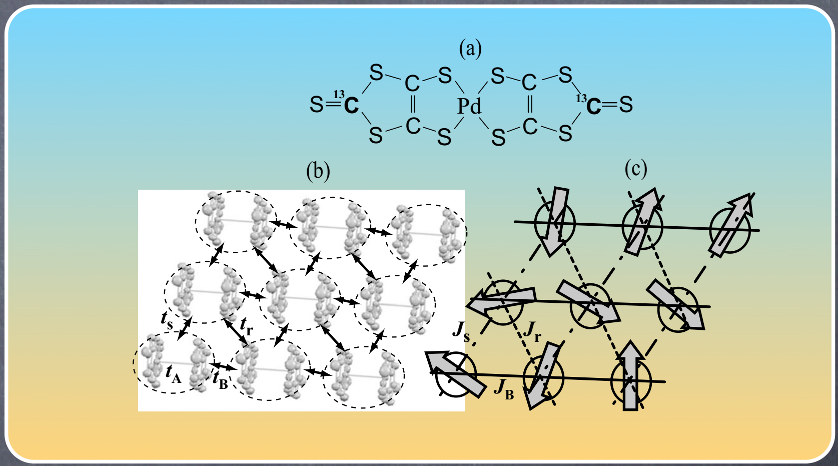
- ^{13}C NMR: line broadening at low temperature in a field
  - indicates inhomogenous AF moments induced by field





$EtMe_3Sb[Pd(dmit)_2]_2$

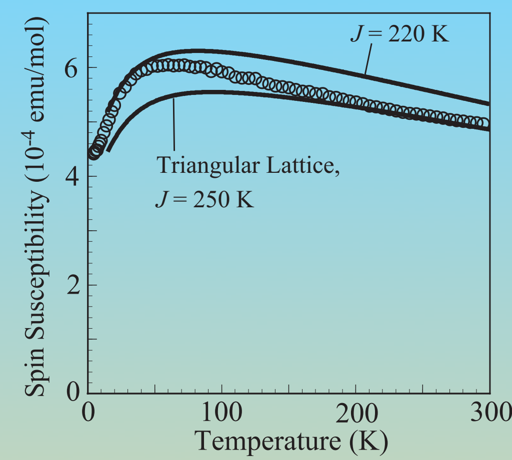
- another organic triangular lattice Mott insulator!

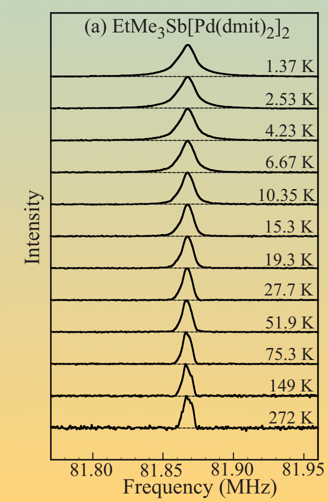


R. Kato group

$EtMe_3Sb[Pd(dmit)_2]_2$

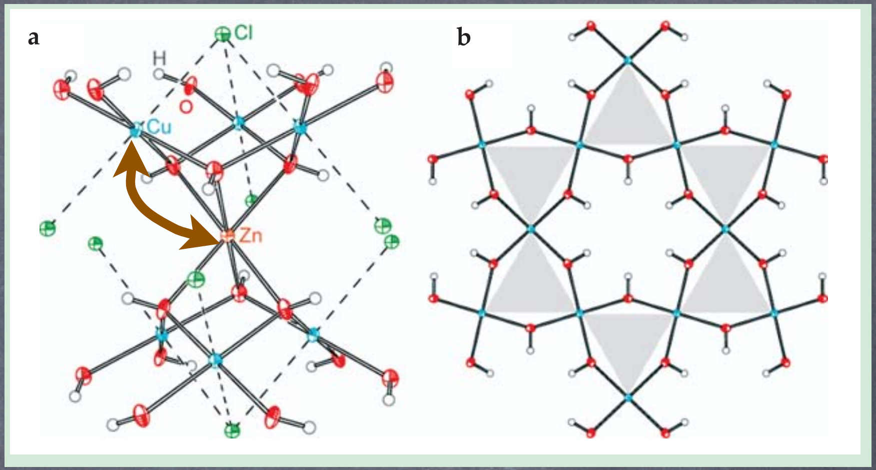
- Susceptibility very similar to κ-(ET)
- No line broadening from static moments



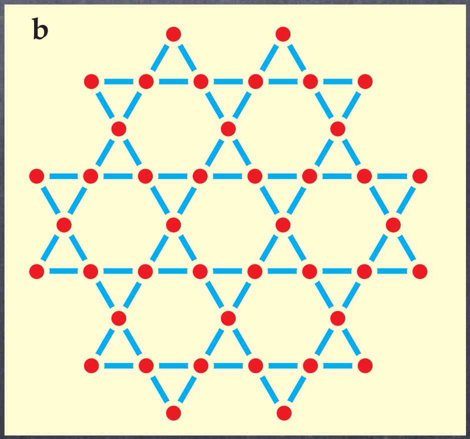


$ZnCu_3(OH)_6Cl_2$

Cu and Zn
can “invert”



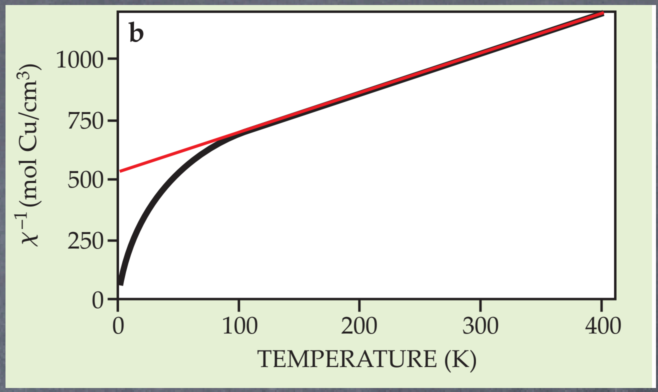
- Herbertsmithite - a 2d s=1/2 kagome antiferromagnet

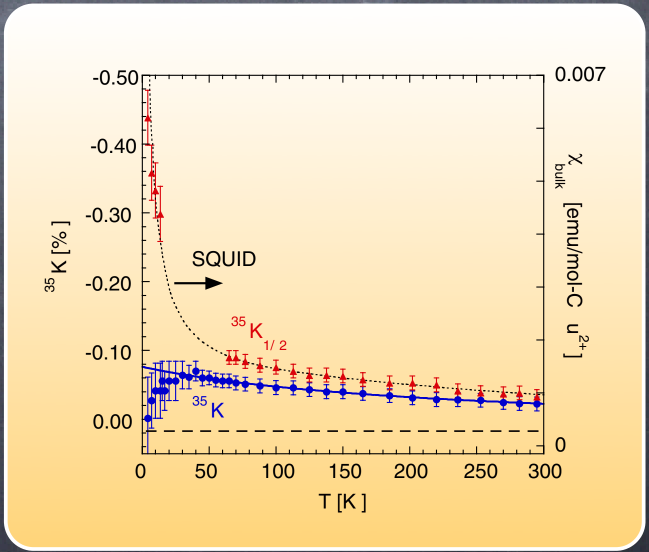


D. Nocera, Y.S. Lee groups

$ZnCu_3(OH)_6Cl_2$

- Upturn of χ below 50K, probably due to defect spins
- Curie-Weiss temperature $\Theta_{CW} \approx$ -240K from ^{35}Cl NMR, $\Theta_{CW} \approx$ -300K from χ
- No order down to T=50mK

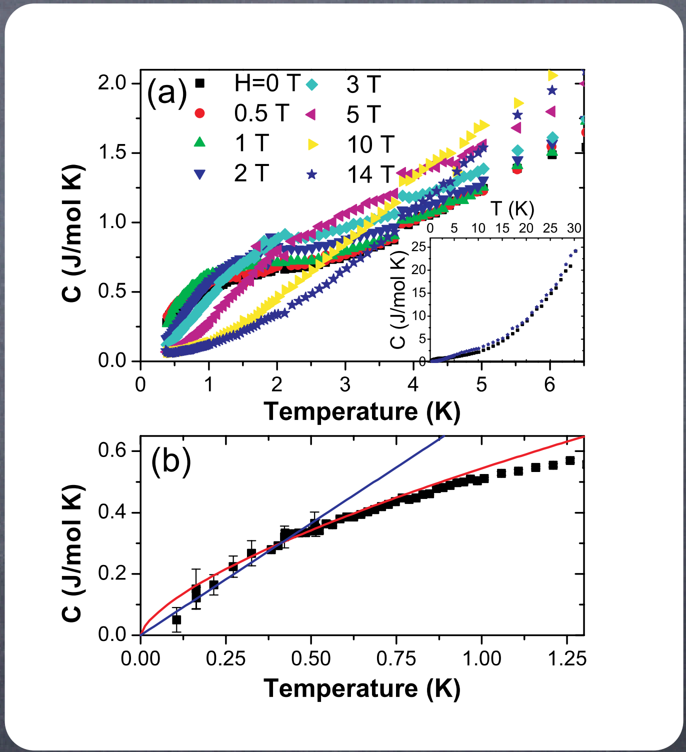




T. Imai et al

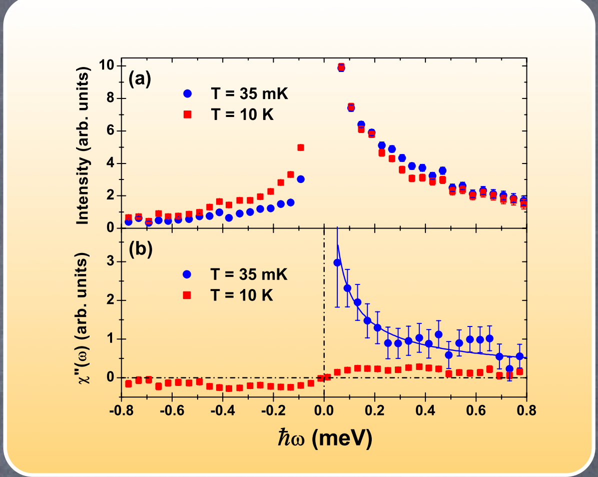
$ZnCu_3(OH)_6Cl_2$

- Specific heat is dominated by magnetic contribution below only ≈ 1K
- This appears roughly power law $C \sim T^{\alpha}$ with $\alpha = 0.5\text{-}1$
- Indicates many low energy excitations

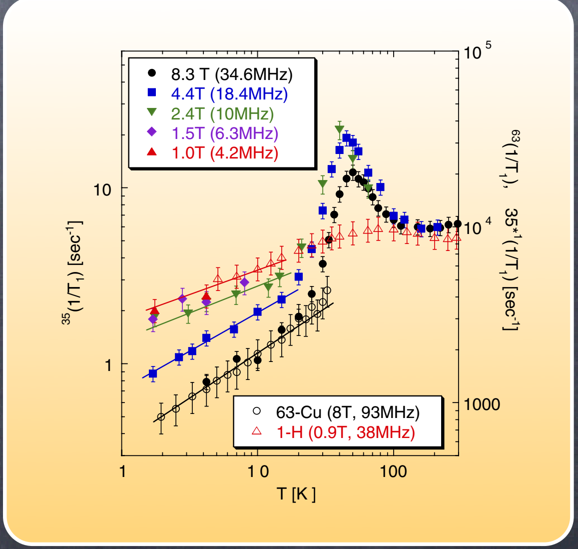


$ZnCu_3(OH)_6Cl_2$

- Evidence of gapless spin excitations:
  - low-energy $\chi''(E)$ in neutrons
  - Similar behavior observed in $1/T_1 \propto T\,\chi''(0^+,T)$



$\chi''(E)$



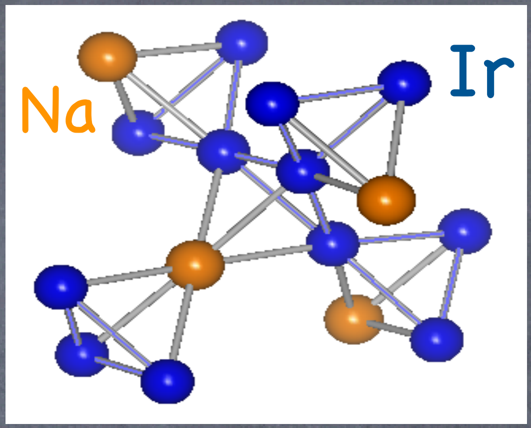
$1/T_1$

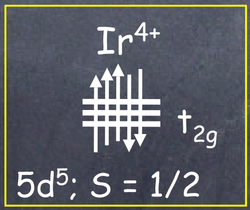
Theory

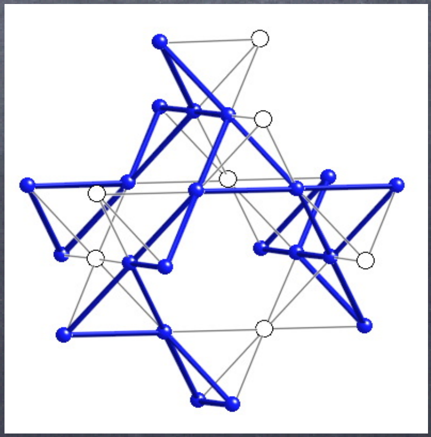
- U(1) Dirac ASL proposed (Y. Ran, M. Hermele et al)
  - Predicts $\chi \sim T$, $C_v \sim T^2$ in pure system
  - Can be reconciled to $\chi \sim$ const, $C_v \sim T$ by impurities
- A large concentration of 5-10% of disorderd inverted Zn and Cu ions makes interpretation difficult

$Na_4Ir_3O_8$

- An “hyperkagome” lattice of Ir^{4+} spins
- Expect S=1/2 spin state - orbital state unclear?



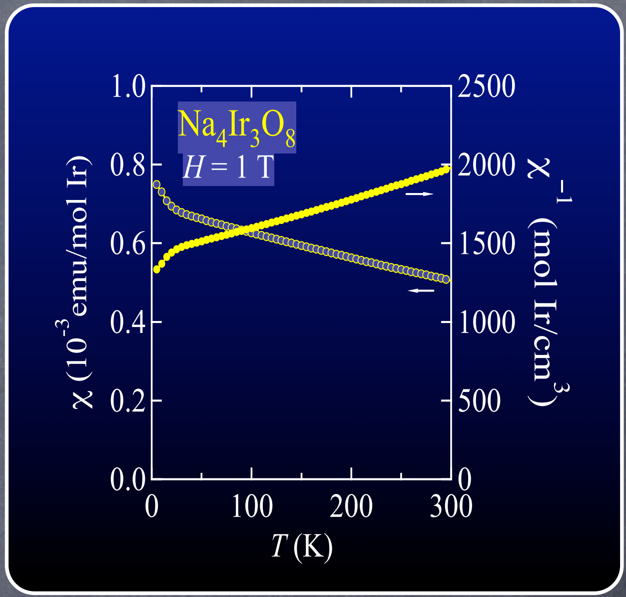


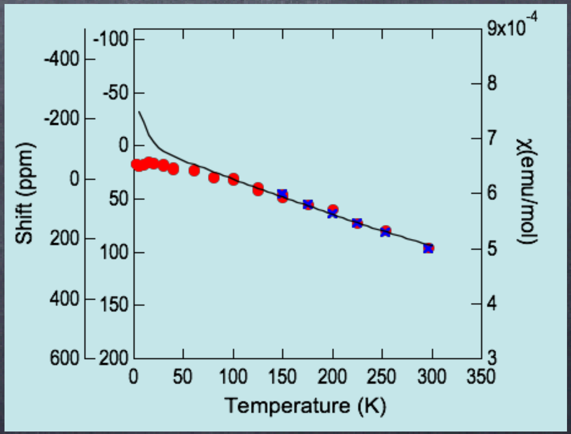


Takagi group

$Na_4Ir_3O_8$

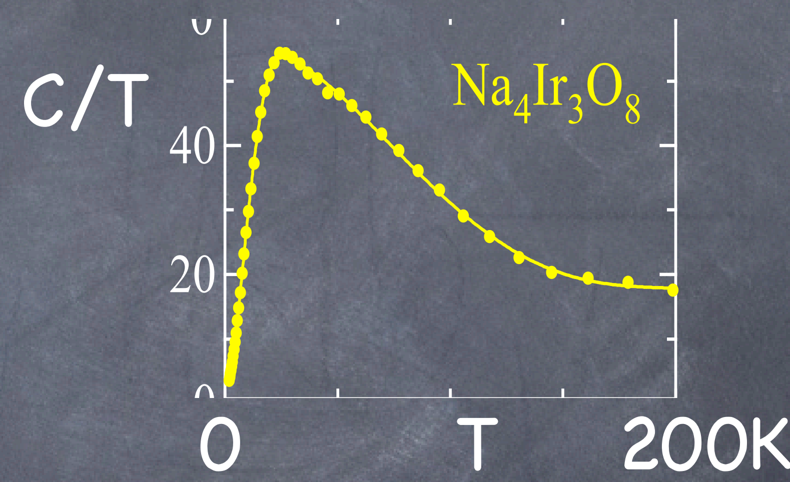
- Susceptibility
  - Curie-Weiss temperature $\Theta_{CW} \approx$ -650K
  - Large χ at low T
  - μ_{eff} = 1.96 μ_B/Ir ≈ 1.73 μ_B/Ir (s=1/2)
- Consistent with Knight shift
  - low-T upturn not seen in K: extrinsic

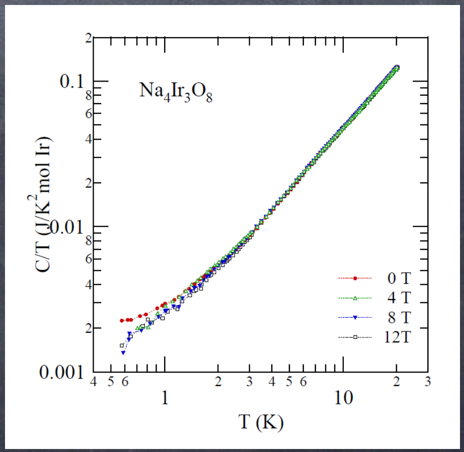




$Na_4Ir_3O_8$

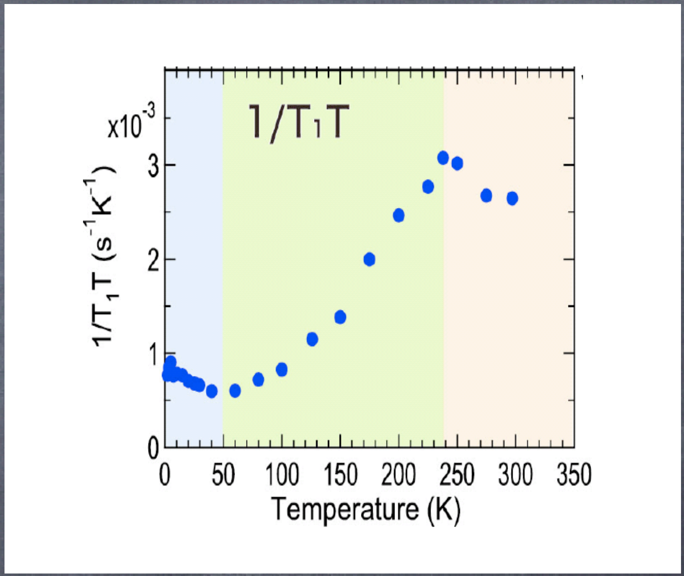
- Specific Heat
  - broad peak around 30K
  - power-law (between T and T^2) at low T indicates many low energy excitations





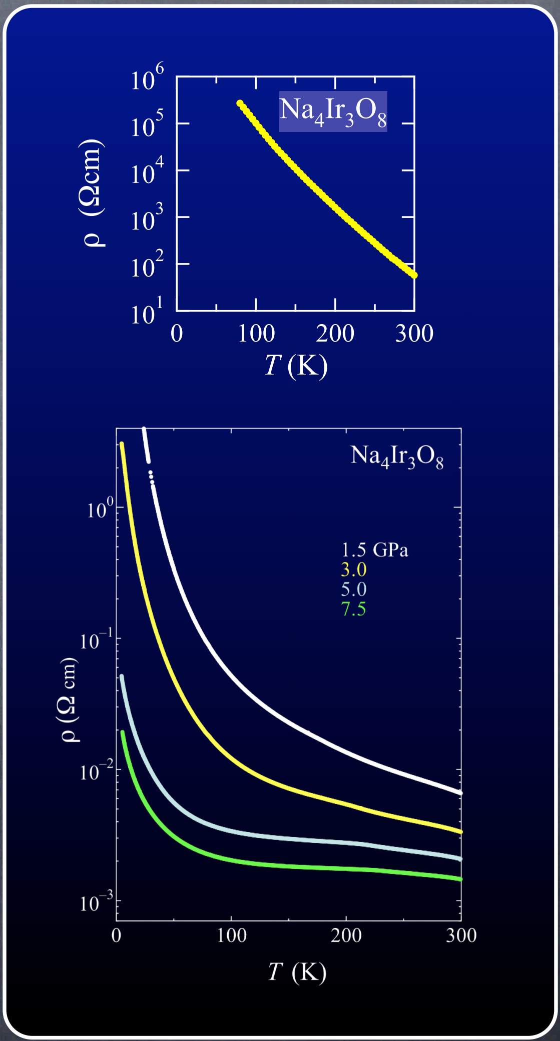
$Na_4Ir_3O_8$

- NMR $1/T_1$ rate is power law for $50<T<200$, suggestive of low energy excitations



$Na_4Ir_3O_8$

- Transport
  - Mott insulator
  - but...close to a Mott transition
- Perhaps this proximity may be important



Theory

- Heavy Ir (Z=77) has strong spin-orbit
  - expect j=1/2 spin with g=-2
  - Hamiltonian may be Heisenberg plus Dzyaloshinskii-Moriya corrections
  - Probably explains large $\chi(T=0)$

G. Chen+LB

- Large size differences between Na, Ir, O suggests little disorder
- Gapless specific heat suggests gapless spinons
  - Two QSL proposals can roughly fit specific heat but both have some difficulties
  - Perhaps resolved by itinerancy?

Y. Zou et al
M. Lawler et al

Conclusions

- Frustrated magnets provide a rich variety of phenomena including a number of promising new quantum spin liquid candidates
- For QSLs, what is needed is a combined effort of innovative experimental and theoretical work, with attention of the latter paid to the former!