Evora, 11/2008

Spin Liquid States in Frustrated Antiferromagnets

Leon Balents
KITP

The David and Lucile Packard Foundation

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Looking for

Spin Liquid States in Frustrated Antiferromagnets

and finding surprises

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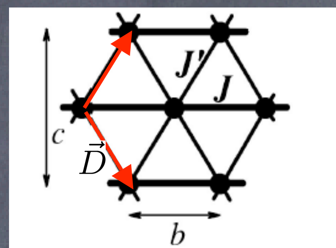
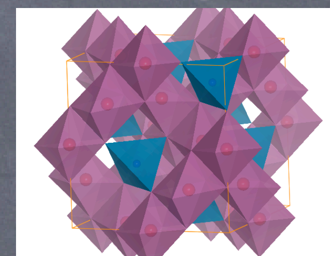
Collaborators



- Oleg Starykh, U. Utah

- Masanori Kohno, NIMS, Japan

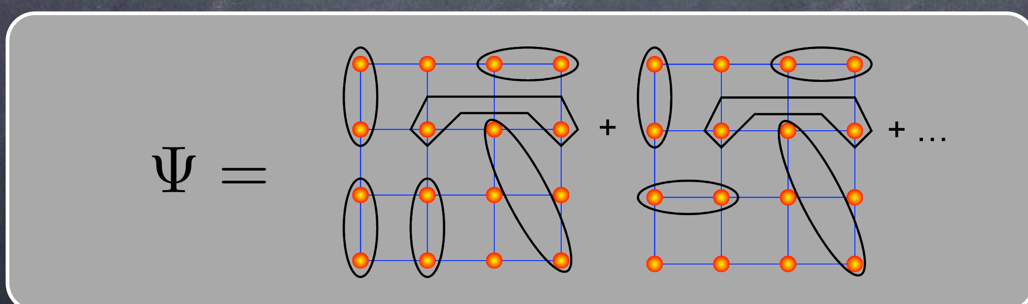


- Gang Chen, UCSB

- Andreas Schnyder, KITP

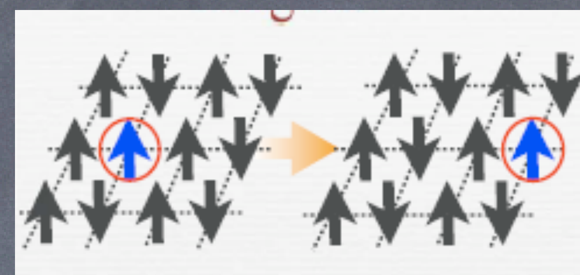
Quantum Spin Liquids

- QSL: a state of a magnet in which quantum fluctuations prevent order even at T=0.
- Many theoretical suggestions since Anderson (73)
  - "Resonating Valence Bond" QSL states

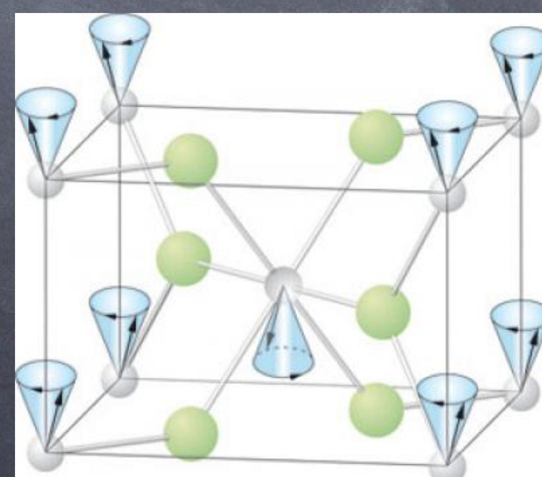


Magnons

- Basic excitation: spin flip
  - Carries "S^z"=$\pm$ 1



- Periodic Bloch states: spin waves
  - Quasi-classical picture: small precession



MnF_2

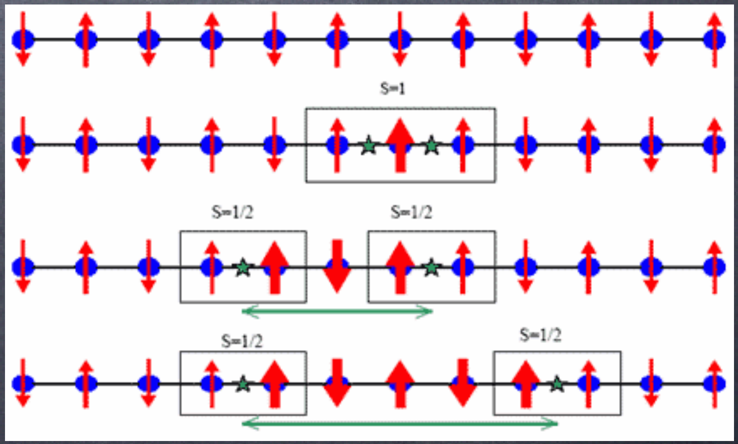
$\vec{k}$

$\epsilon = \hbar\omega(\vec{k})$

Image: B. Keimer

One dimension

- Heisenberg model is a *spin liquid*
  - No magnetic order $\langle \vec{S}(x) \cdot \vec{S}(x') \rangle \sim \frac{(-1)^{x-x'}}{|x-x'|} + \cdots$
  - Power law correlations of spins and dimers
- Excitations are s=1/2 *spinons*
  - General for 1d chains

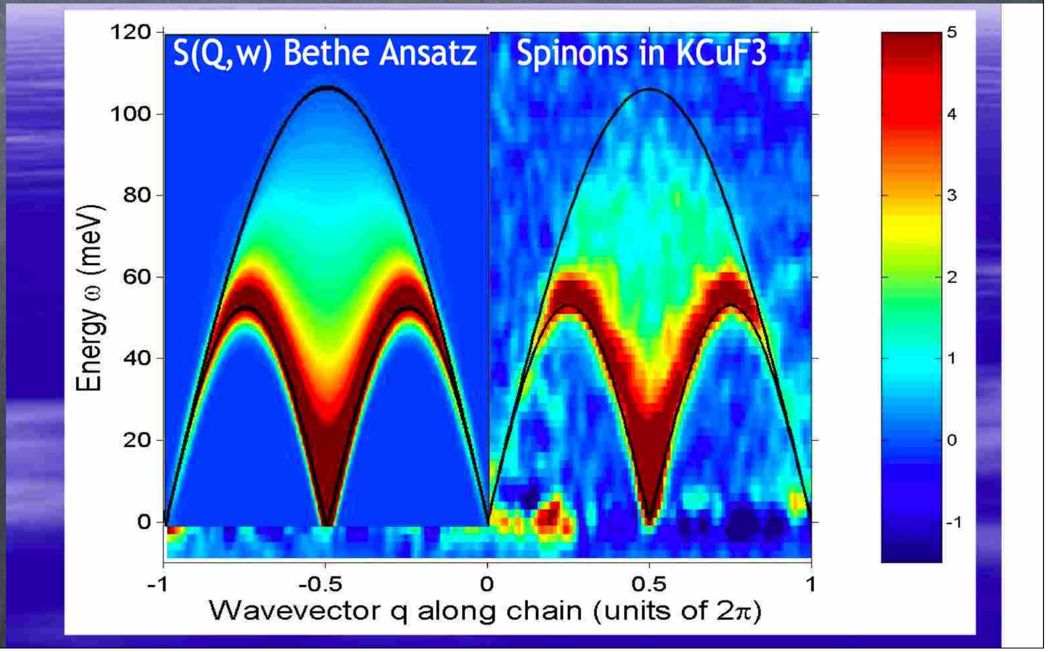


Spinons by neutrons

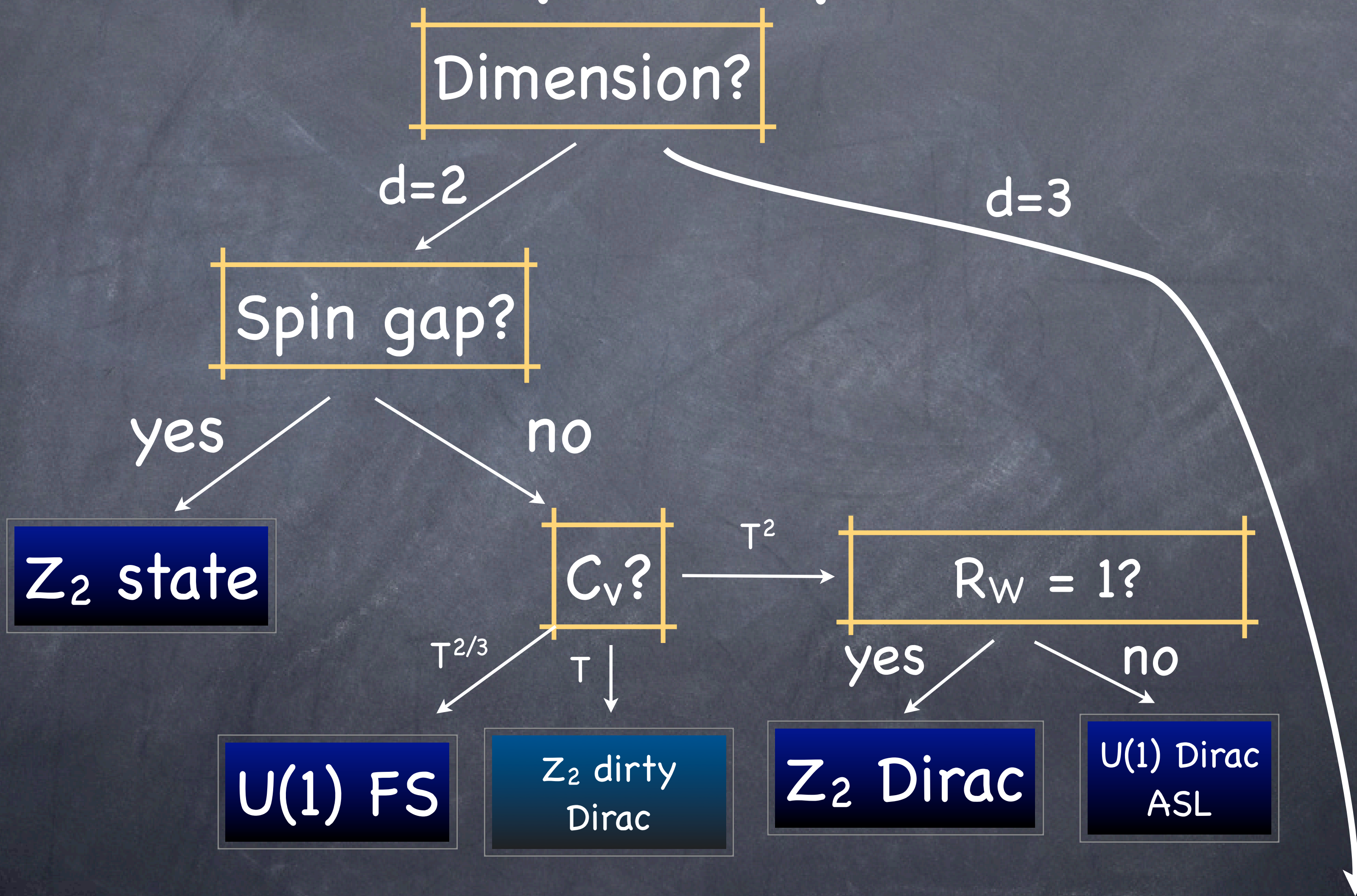
- Bethe ansatz: $\epsilon_s(k) = \frac{\pi J}{2} |\sin k|$
  - Spinon energy $k = k_1 + k_2$
  - Spin-1 states $\epsilon = \epsilon_s(k_1) + \epsilon_s(k_2)$

  } 2-particle continuum

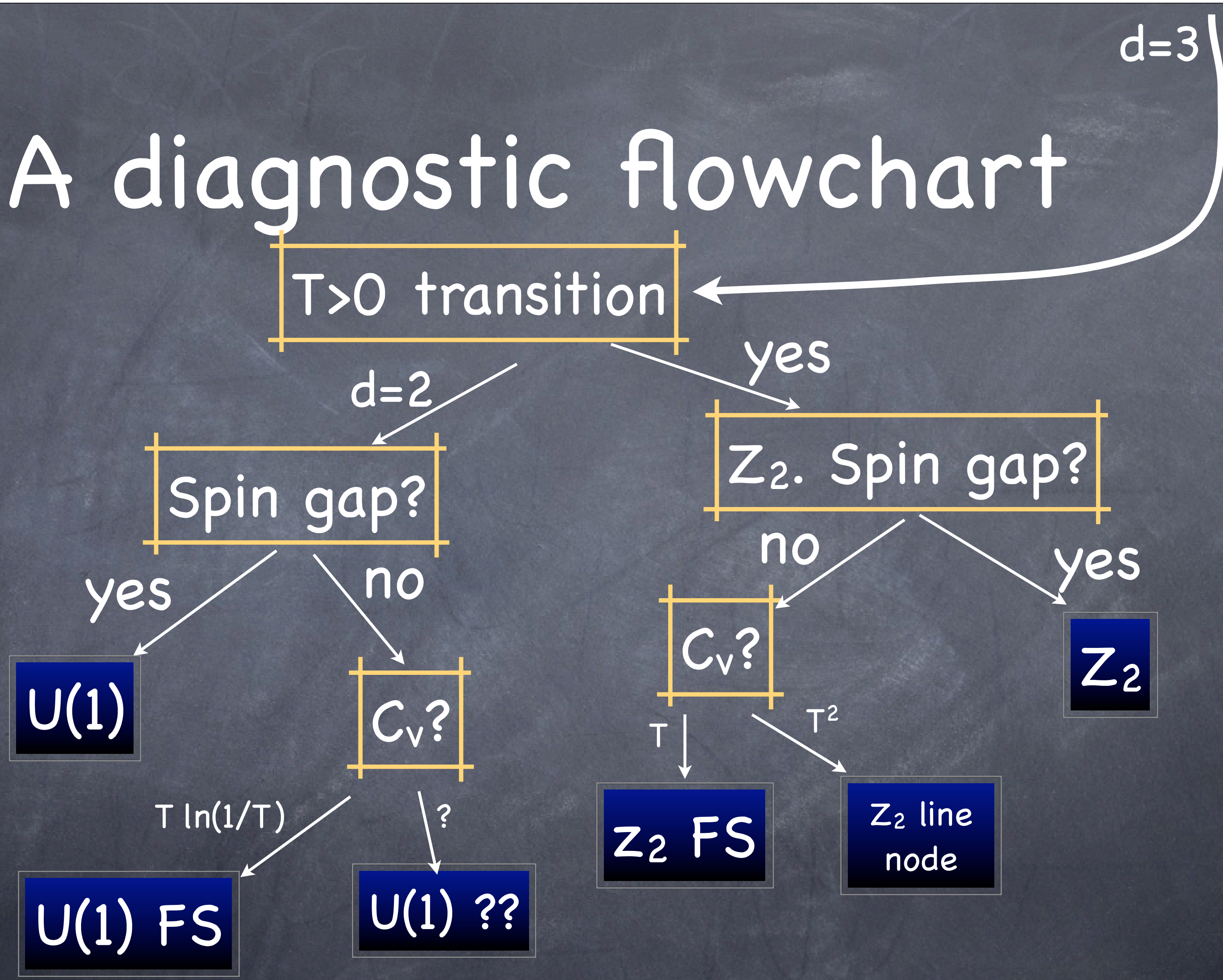
- Theory vs experiment for $KCuF_3$ with anisotropy ≈30
  - B. Lake, HMI



Many spin liquids in theory!



A diagnostic flowchart



disordered possibilities neglected

QSL candidates

- $CsCu_2Cl_4$ - spin-1/2 anisotropic triangular lattice

?

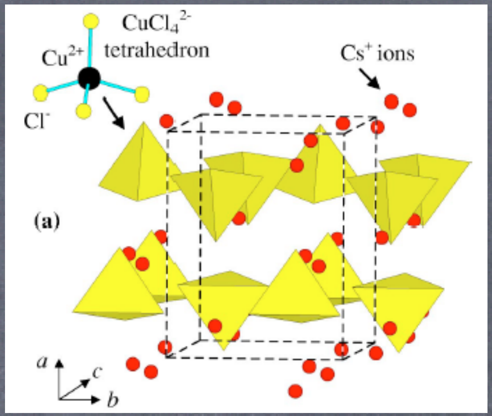
- $NiGa_2S_4$ - spin-1 triangular lattice
- κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ - triangular lattice organics
- $FeSc_2S_4$ - orbitally degenerate spinel

?

- $Na_4Ir_3O_8$ - hyperkagome
- $ZnCu_3(OH)_6Cl_2$ - kagome

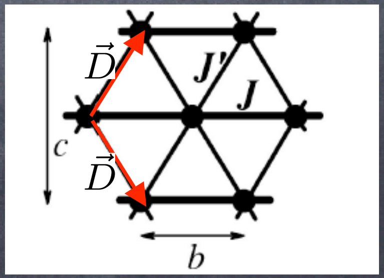
Cs_2CuCl_4

- 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:

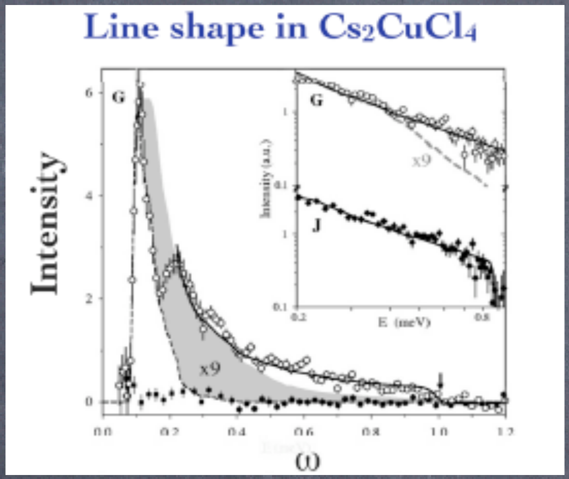
J=0.37meV
J'=0.3J

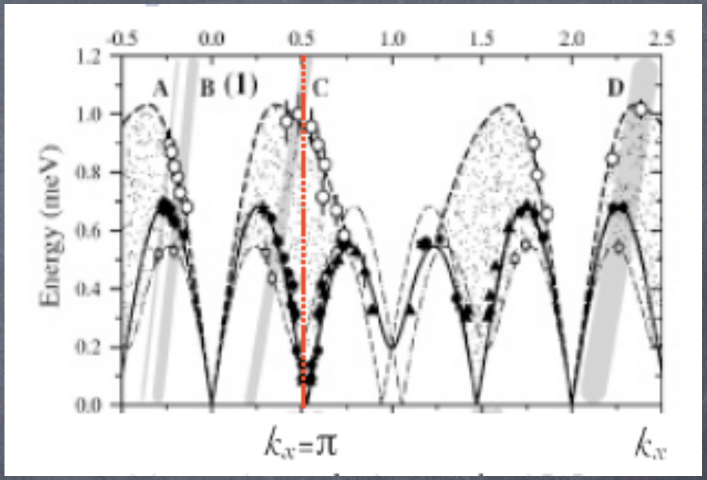
D=0.05J

R. Coldea et al

Neutron scattering

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



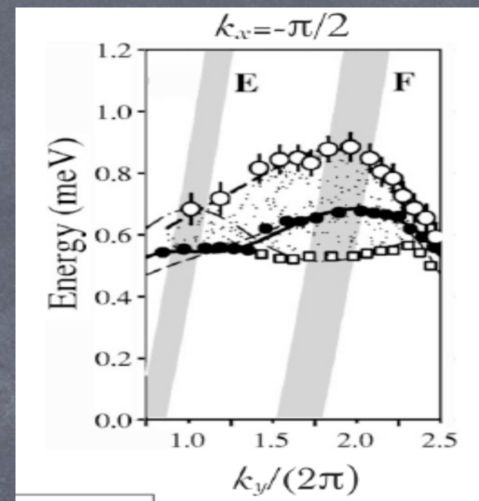


Very broad spectrum similar to 1d (in some directions of k space). Roughly fits power law.

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

2d theories

- Arguments for 2d:
  - J'/J = 0.3 *not very small*
  - Transverse dispersion
- Exotic theories:



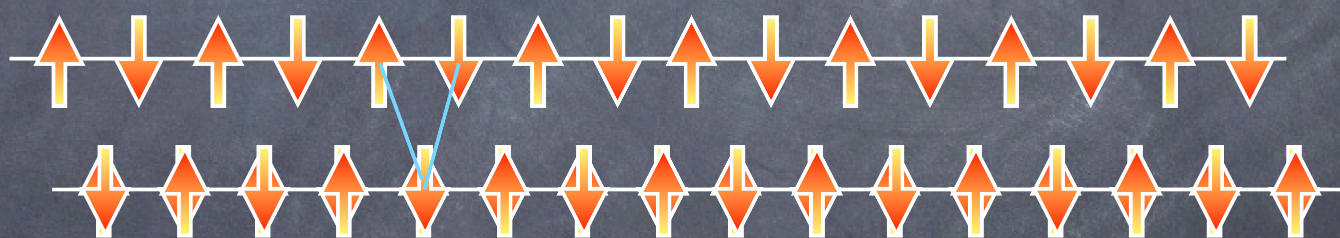
- J.Alicea, O.I.Motrunich & M.P.Fisher: Phys. Rev. Lett. **95**, 247203 (2005).
- S.V.Isakov, T.Senthil & Y.B.Kim: Phys. Rev. B **72**, 174417 (2005).
- Y.Zhou & X.-G.Wen: cond-mat/0210662.
- F.Wang & A.Vishwanath: Phys. Rev. B **74**, 174423 (2006).
- C.-H.Chung, K.Voelker & Y. B. Kim: Phys. Rev. B **68**, 094412 (2003).

- Spin waves

- M.Y.Veillette, A.J.A.James & F.H.L.Essler: Phys. Rev. B **72**, 134429 (2005).
- D.Dalidovich, R.Sknepnek, A.J.Berlinsky, J.Zhang & C.Kallin: Phys. Rev. B **73**, 184403 (2006).
- R.Coldea, D.A.Tennant & Z.Tylczynski: Phys. Rev. B **68**, 134424 (2003).

Dimensional reduction?

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

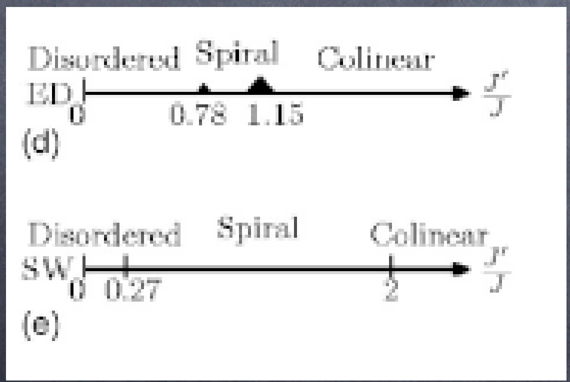


  - Leading effects on correlations are in fact $O[(J')^4/J^3]$!

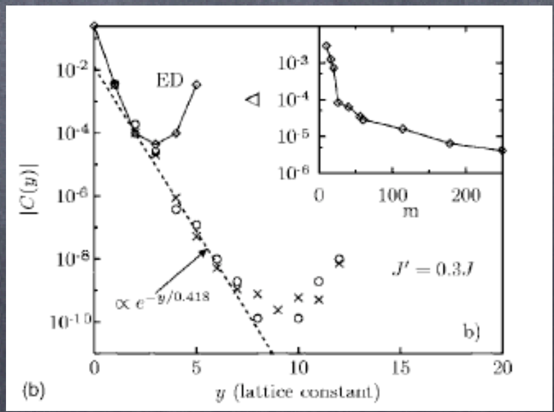
Dimensional reduction?

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

Weng et al, 2006



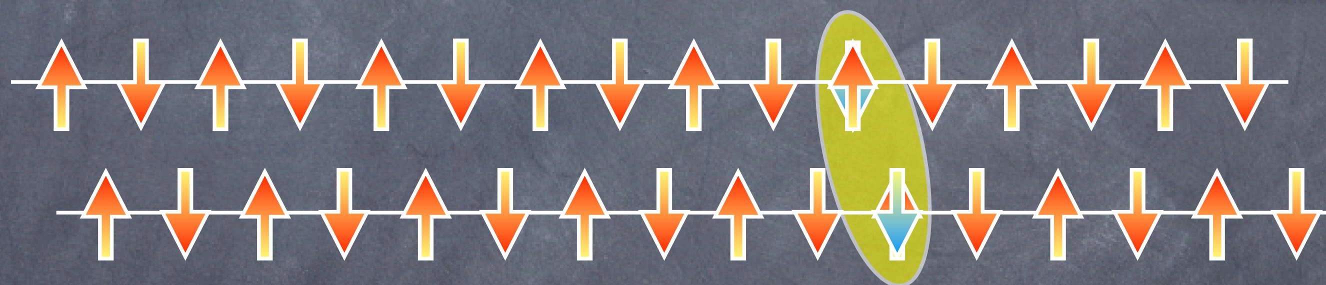
Very different from spin wave theory



Very weak inter-chain correlations

Excitations

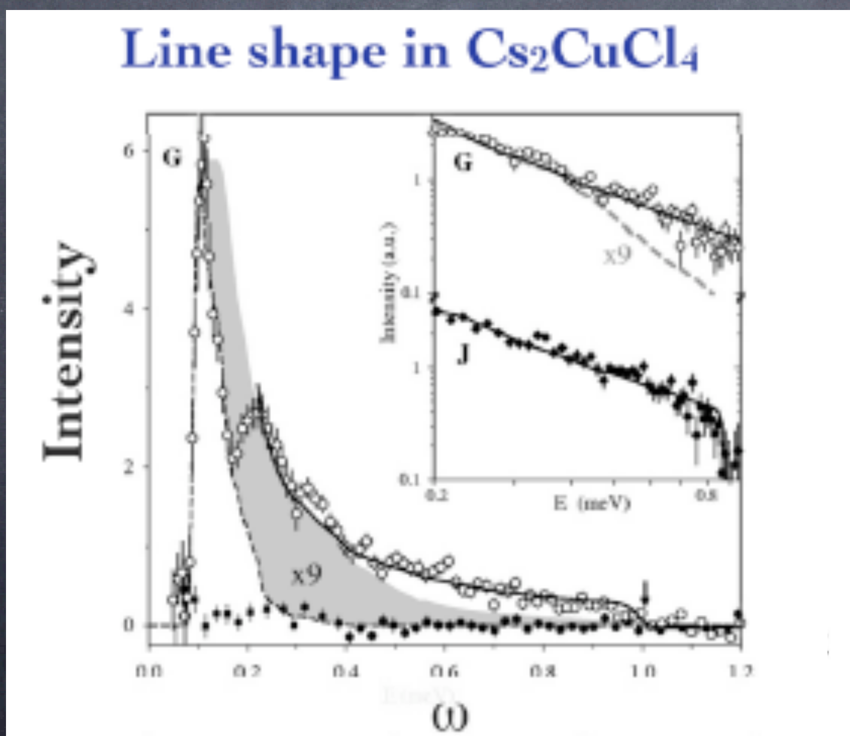
- Build 2d excitations from 1d spinons
  - Exchange: $\frac{J'}{2}\left(S_i^+ S_j^- + S_i^- S_j^+\right)$

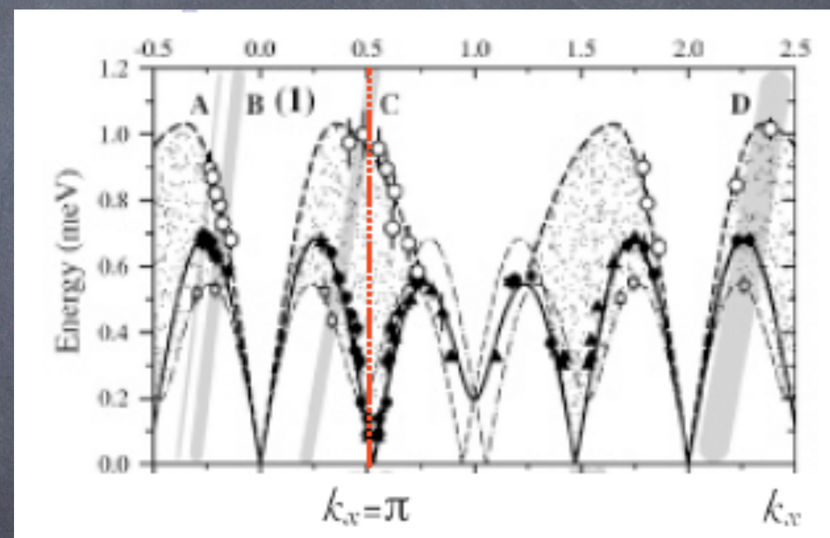


- Expect spinon binding to lower inter-chain kinetic energy
  - Use 2-spinon Schroedinger equation

Broad lineshape: “free spinons”

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

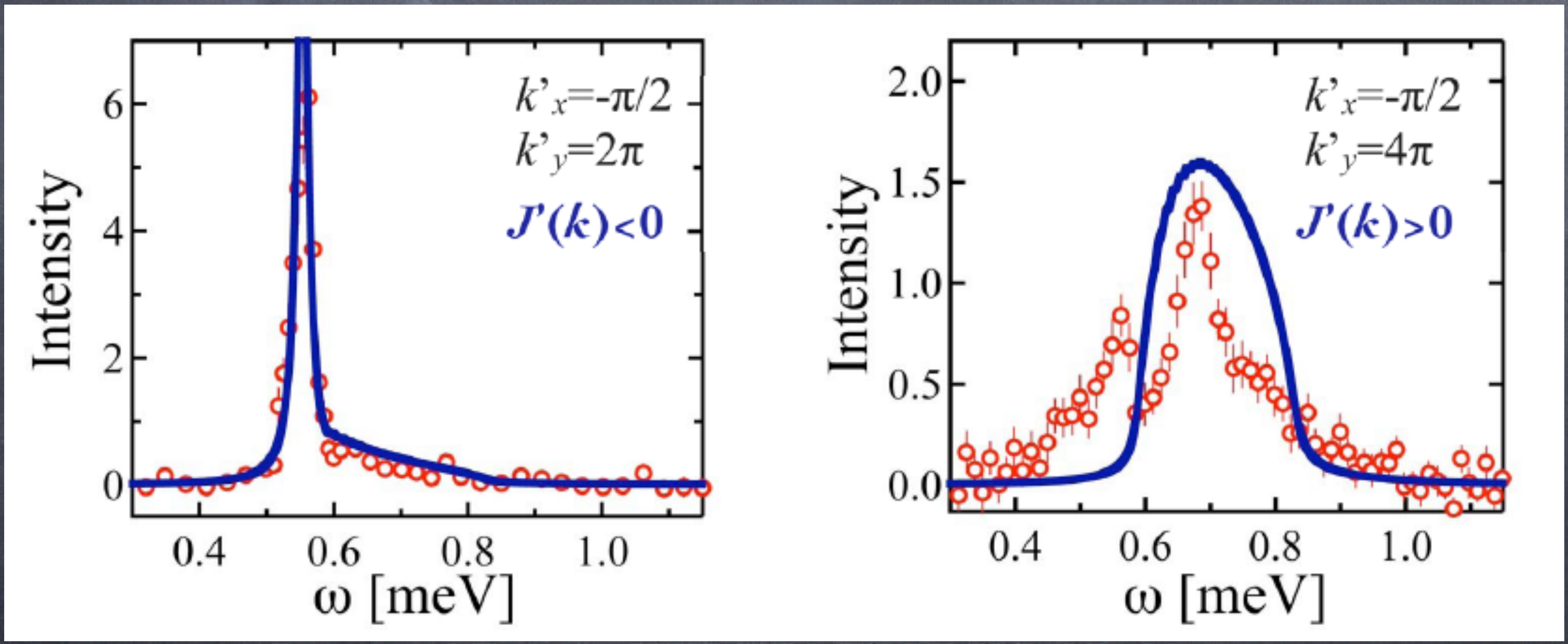




J’(k)=0 here

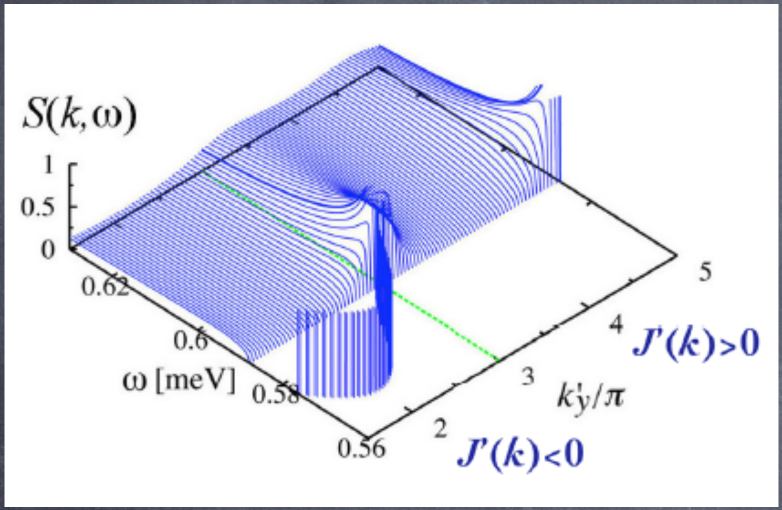
Bound state

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

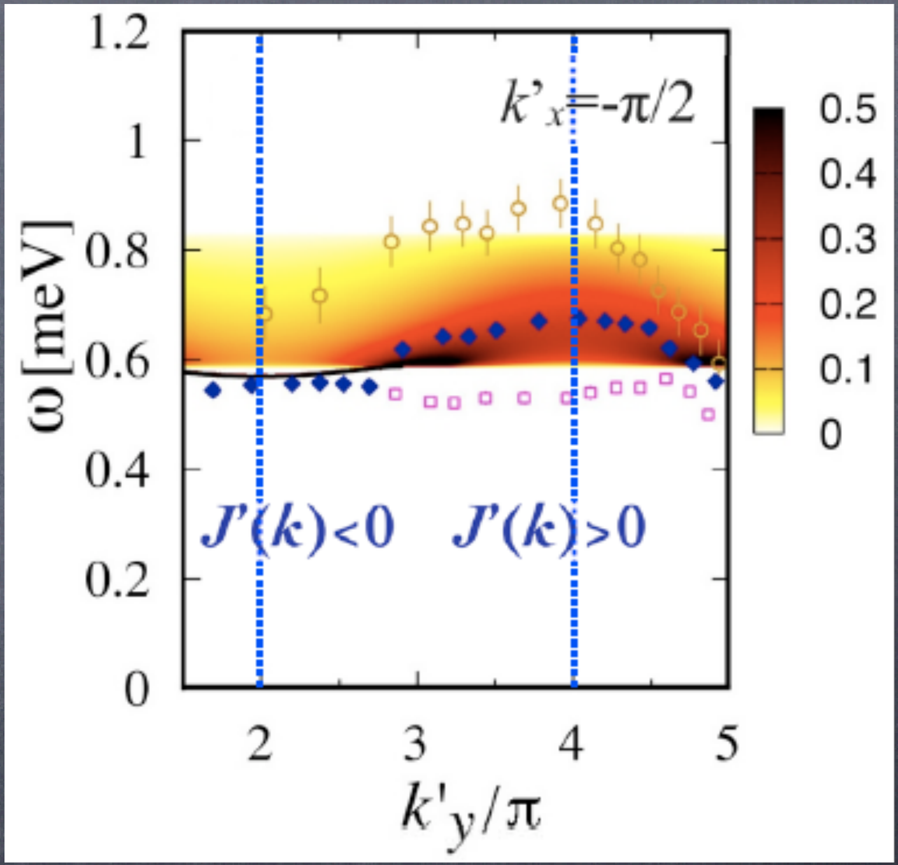


- Curves: 2-spinon theory w/ experimental resolution
- Curves: 4-spinon RPA w/ experimental resolution

Transverse dispersion

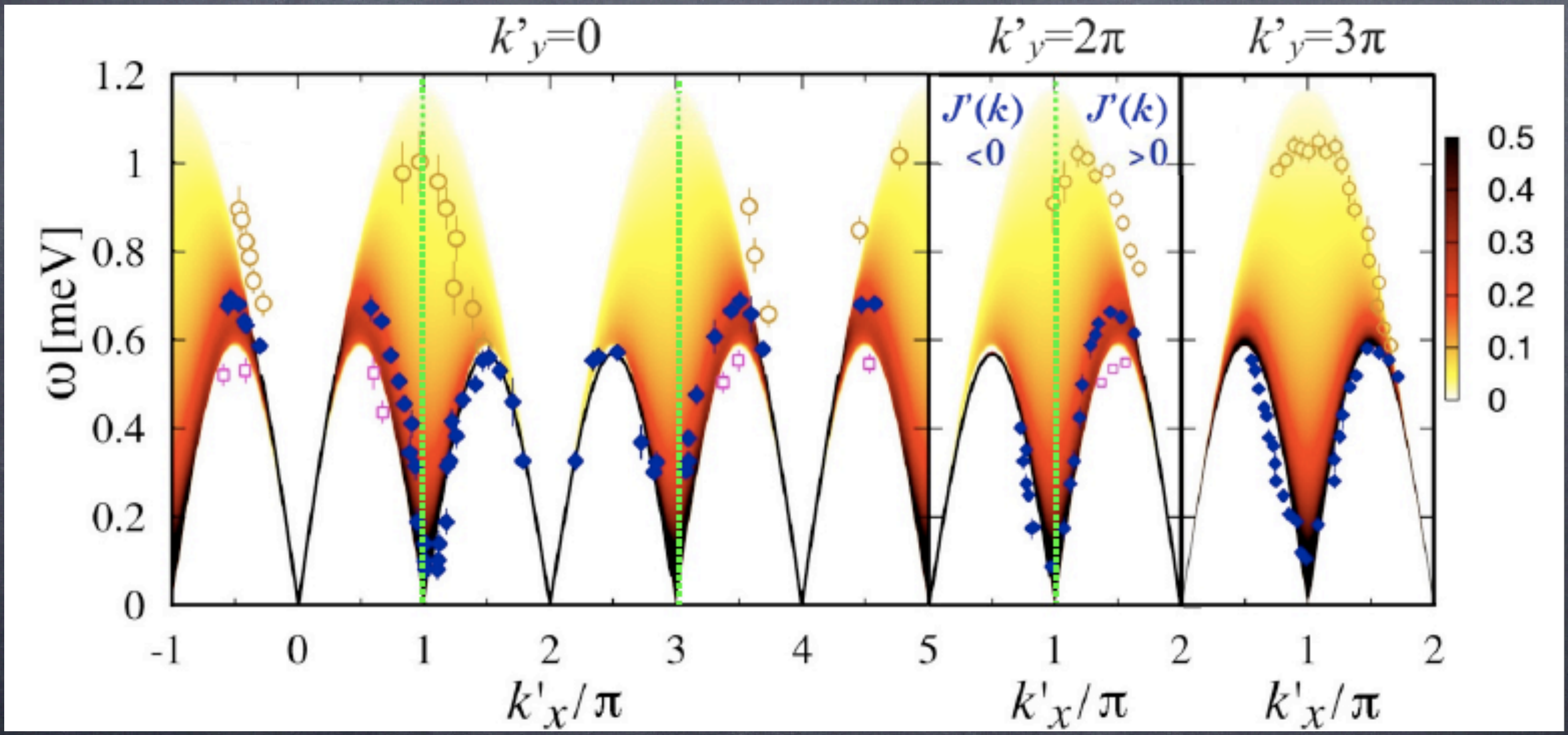


Bound state and resonance



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

Spectral asymmetry



- Vertical lines: J’(k)=0.

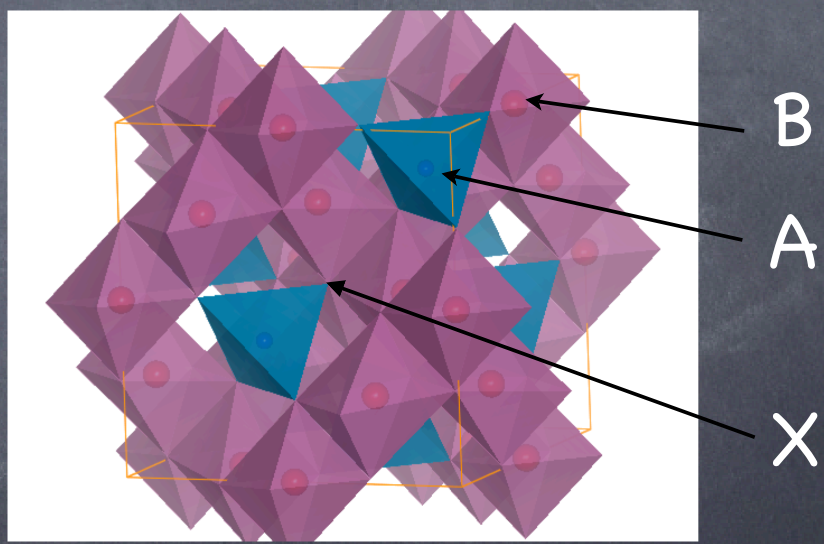
Conclusions on Cs_2CuCl_4

- Simple theory works well for frustrated quasi-1d antiferromagnets
  - Frustration leads to a strong enhancement of one-dimensionality
- The mystery of Cs_2CuCl_4 should be considered solved
  - Many (nearly all) other details of diverse experiments on this material may be understood in the same framework

AB_2X_4 spinels

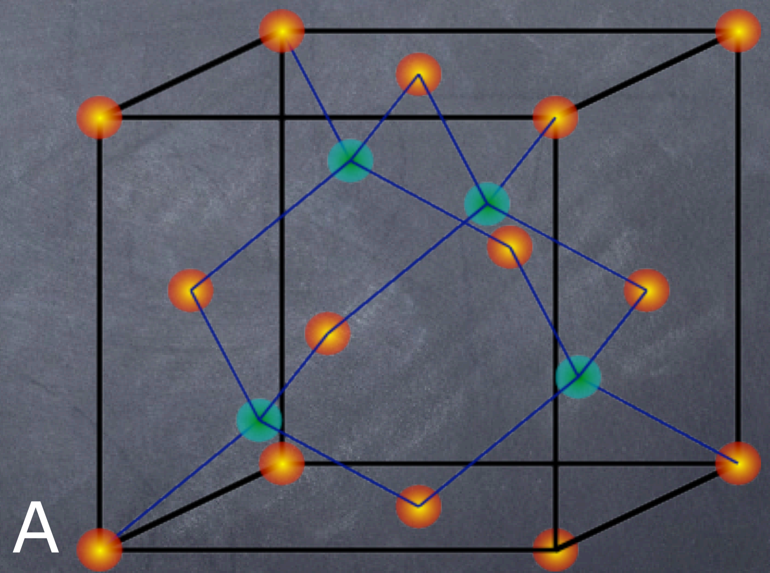
cubic $Fd\bar{3}m$

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

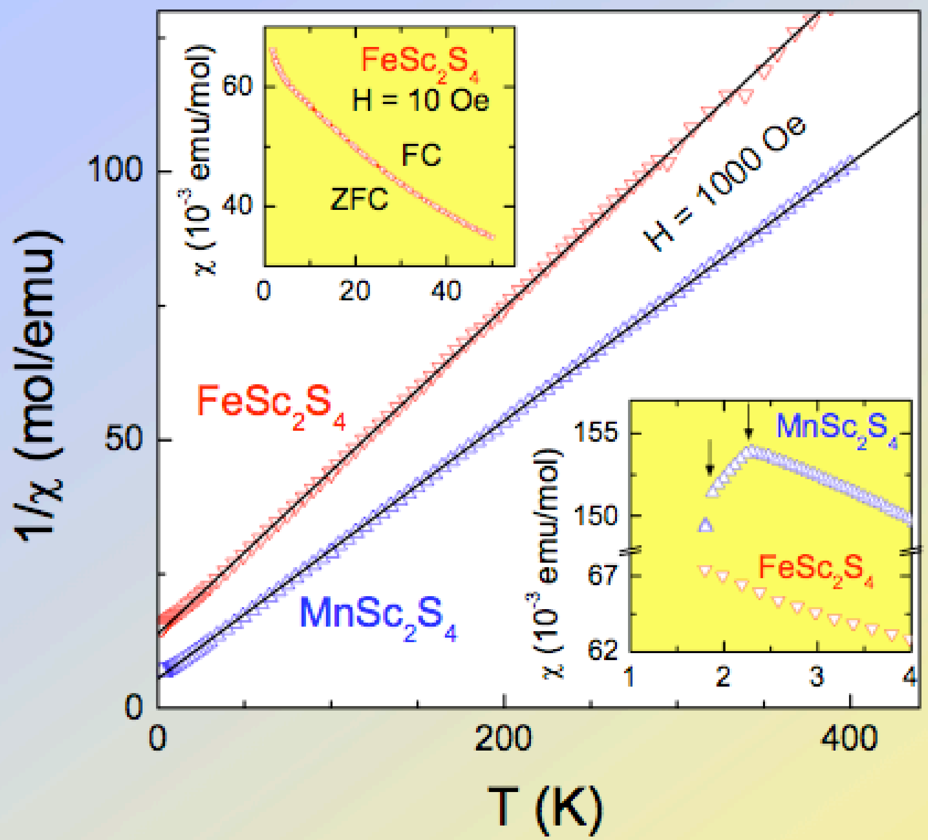


Deconstructing the spinel

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



Frustration Signature



$FeSc_2S_4$: θ_{CW} = 50 K

T > 30 mK:

no long-range magnetic order

no spin-glass

$MnSc_2S_4$: θ_{CW} = 25 K

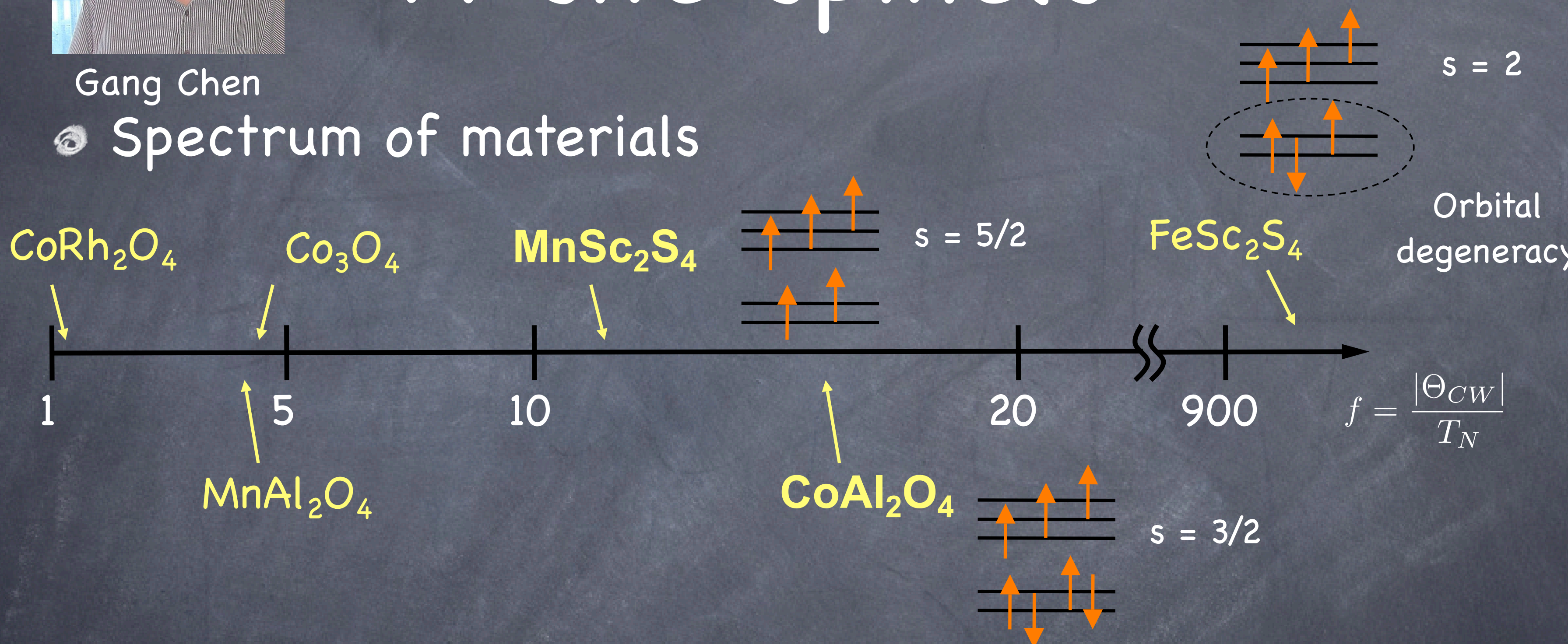
AFM transition @ 2 K

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

Gang Chen

A-site spinels

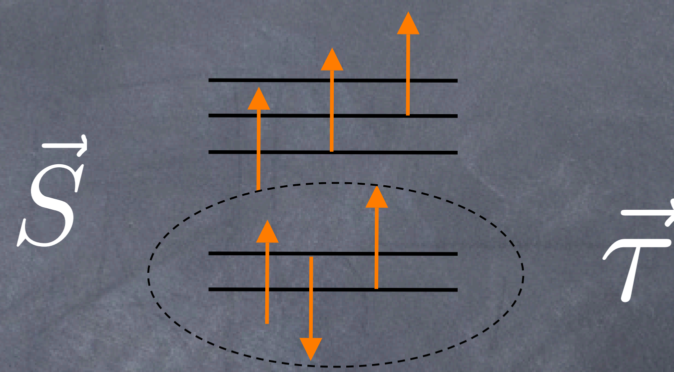
- Spectrum of materials

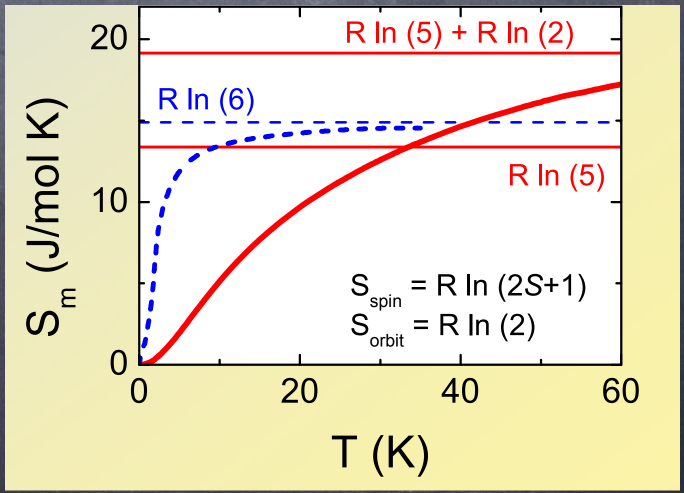


V. Fritsch et al. PRL **92**, 116401 (2004); N. Tristan et al. PRB **72**, 174404 (2005); T. Suzuki *et al.* (2006)

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





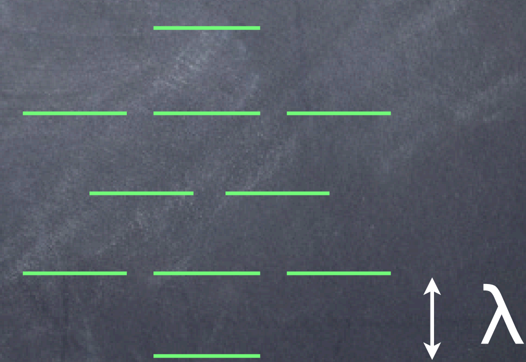
Atomic Spin Orbit

- 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,

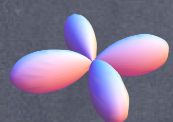
$$\lambda = \frac{6\lambda_0^2}{\Delta}$$



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

Spin orbital singlet

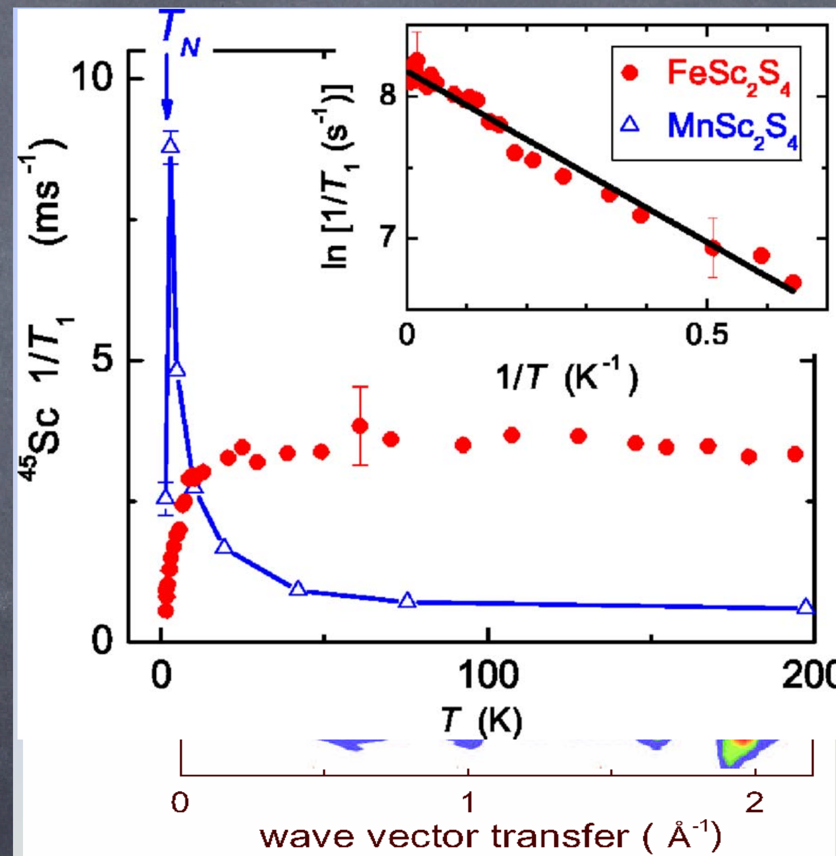
- Ground state of λ>0 term:

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

- Due to gap, there is a stable SOS phase for λ >> J.

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)

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

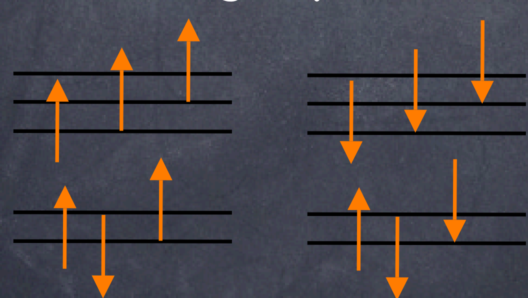
$$+\Big[L_{ij}\boldsymbol{\tau}_i\cdot\boldsymbol{\tau}_j + \tilde{L}_{ij}\tau_i^y\tau_j^y\Big]\mathbf{S}_i\cdot\mathbf{S}_j\Big\}$$

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

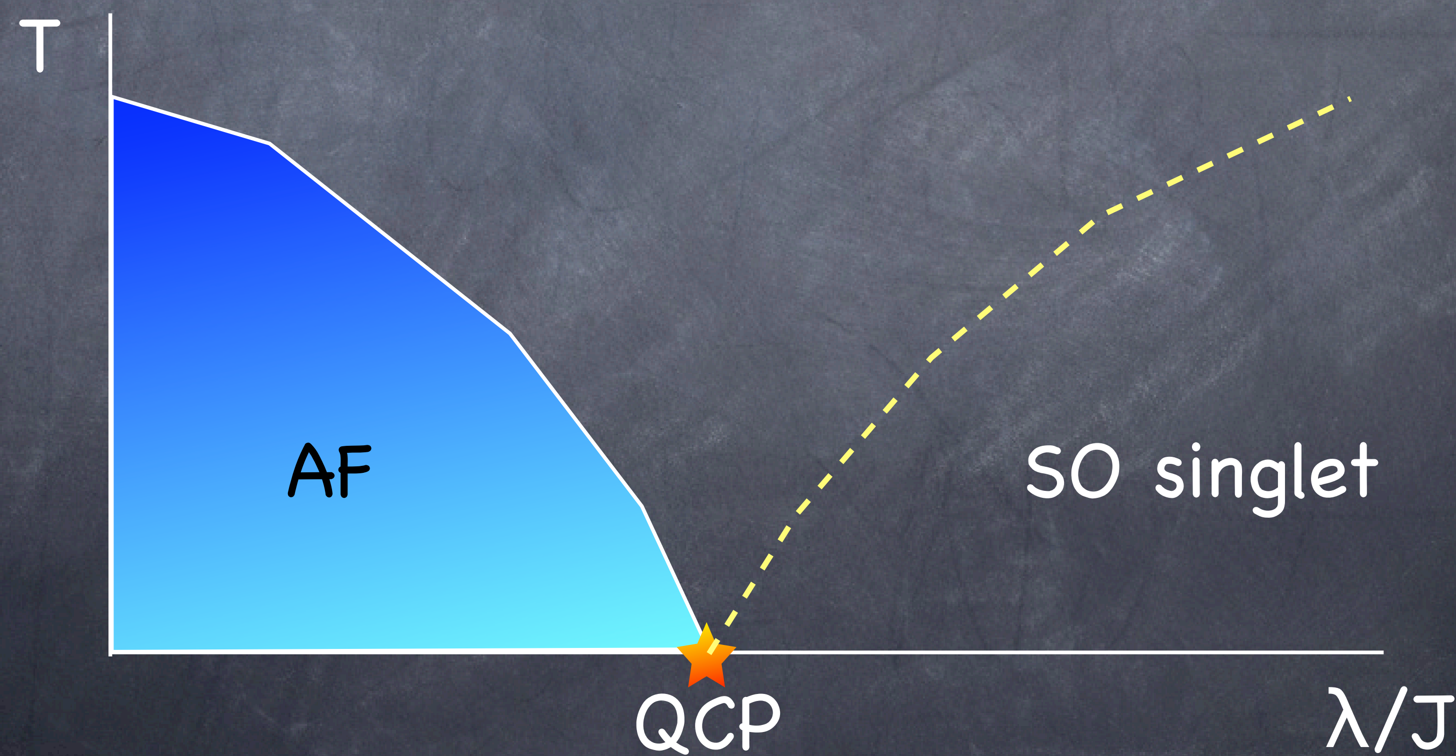


Ordered Phase (J >> λ)

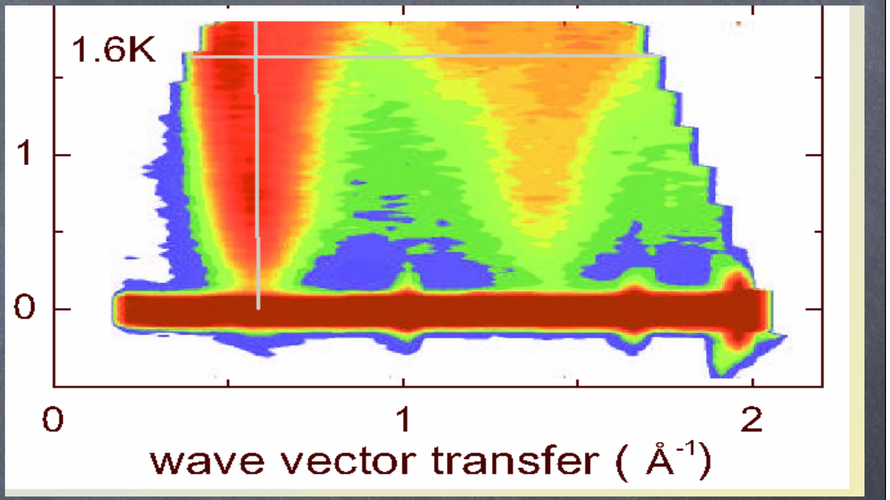
- Ground state of H_{ex} is almost certainly ordered
  - $\mathbf{S}_i \cdot \mathbf{S}_j$ coupling is strongest
  - Complex multi-spiral ground states possible
- Inclusion of weak SOI λ favors simpler commensurate "cubic" spin arrangements
  - spin order leads to induced orbital order

Quantum Critical Point

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



Minimal Model

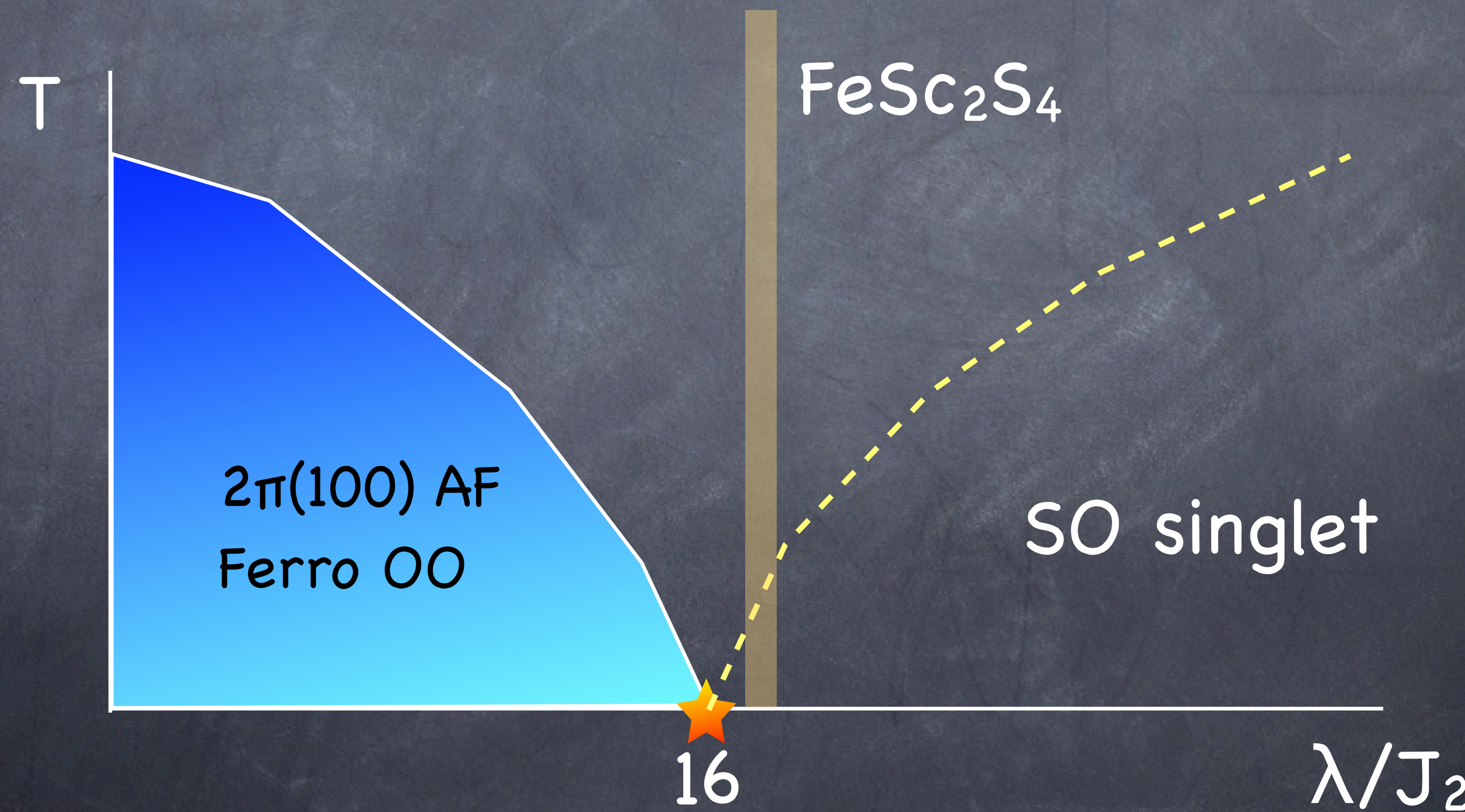


- Neutron scattering suggests peak close to 2π(100)
- Indicates $J_2 >> J_1$

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

Quantum Critical Point

- Mean field phase diagram

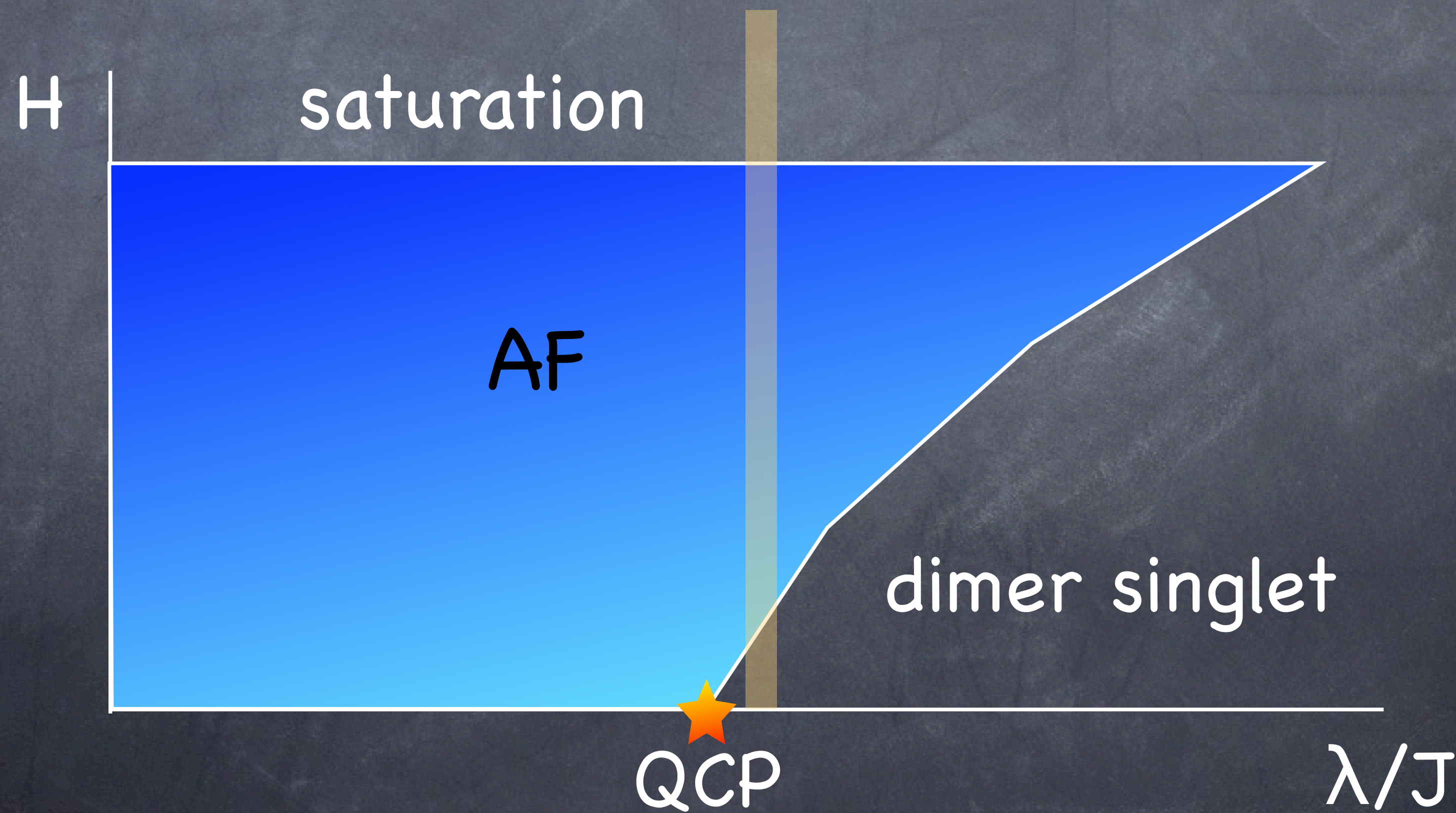


Consequences of QCP

- Power-law spin correlations
- Scaling form for $(T_1T)^{-1} \sim f(\Delta/T)$
- 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?

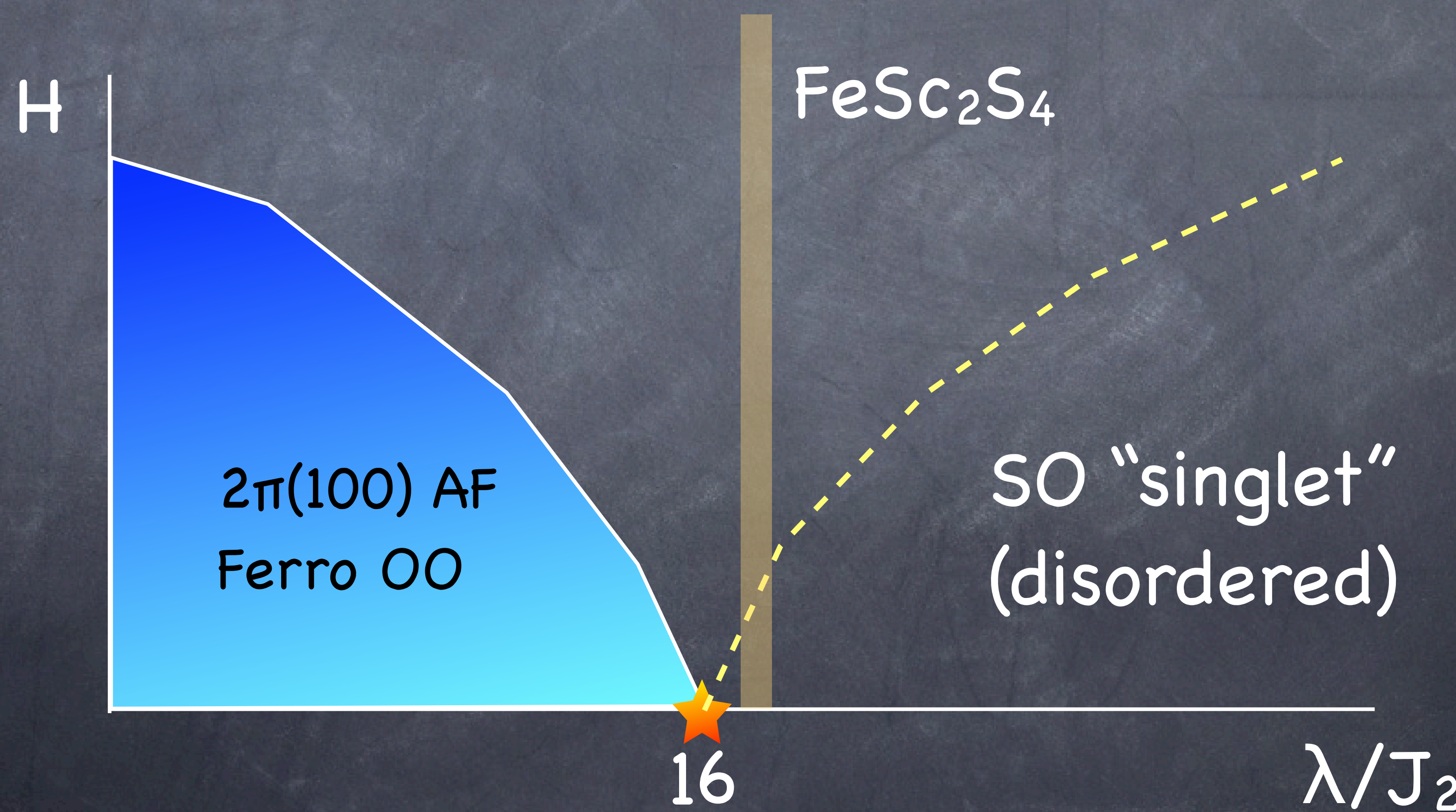
Behavior in Field

- c.f. dimer antiferromagnet



Behavior in field

- This model



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
- Disorder and fields

Conclusions on $FeSc_2S_4$

- Orbital degeneracy and spin orbit provides an exciting route to quantum paramagnetism and quantum criticality
  - entangled spin-orbital singlet ground state in an S=2 magnet!

More for the future!

✔ - $CsCu_2Cl_4$ - spin-1/2 anisotropic triangular lattice

? - $NiGa_2S_4$ - spin-1 triangular lattice

- κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ - triangular lattice organics

✔ - $FeSc_2S_4$ - orbitally degenerate spinel

? - $Na_4Ir_3O_8$ - hyperkagome

- $ZnCu_3(OH)_6Cl_2$ - kagome