Spin fluid and spin nematic states in frustrated quantum magnets

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Quantum magnetism of Mott insulators

Electronic Mott insulators - charges localize below some energy scale "U"

Active low energy degree of freedom - electron spin

Fate of local moments at low temperature ??

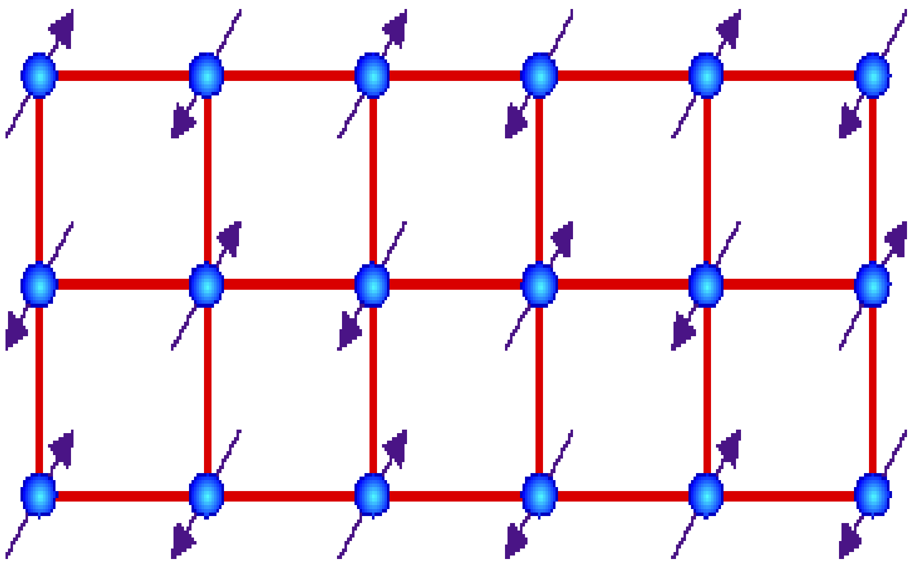
Effective Hamiltonian $H_{eff} \approx J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j + \underbrace{\cdots\cdots}_{}$

(Typically $J > 0$)

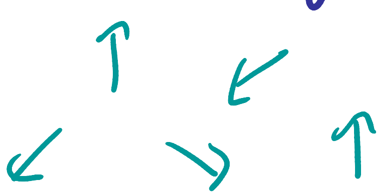
longer range interaction, ring exchange, etc.

Traditional fate – magnetic ordering at low T

- Neel ordered state



Other ordering patterns depending on details

Eg:  non-collinear state on Δ lattice.

Can quantum fluctuations destroy Neel magnetic ordering at T = 0?

Yes - well known in $d = 1$.

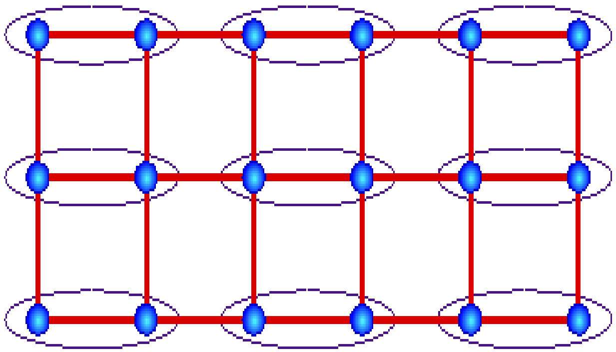
$d > 1$: Long standing theoretical question.

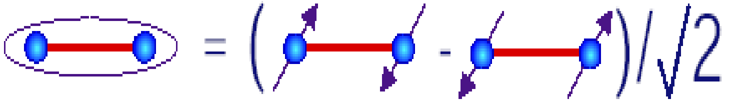
Intense activity in last $\approx$ 20 years.

Possible non-Neel phases

QUANTUM PARAMAGNETS

- Simplest: Valence bond solids (spin-Peierls)
- Ordered pattern of valence bonds breaks lattice translation symmetry.
- Elementary spinful excitations have S = 1 above spin gap.





Seen in many model calculations.

Are there any genuinely 2d or 3d experimental realizations?

Other ordered non-Neel phases - quantum spin nematics

$$\langle \vec{S} \rangle = 0 \quad \text{but} \quad \langle SS \rangle \neq 0$$

$\Rightarrow$ Spontaneous generation of spin anisotropy without any ordered moment ("Moment-free magnetism" – Coleman, Chandra '90)

Eg: $\left\langle S_\alpha S_\beta + S_\beta S_\alpha - \frac{S(S+1)}{3}\delta_{\alpha\beta} \right\rangle \neq 0$

(Spontaneous single ion anisotropy)

or $\langle \vec{S}_i \times \vec{S}_j \rangle \neq 0$ (spontaneous Dzyaloshinskii-Moriya)

Where might stabilize a spin nematic?

1. Magnets with sizeable biquadratic interactions

$$H \approx J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j \underbrace{- K \sum_{\langle ij \rangle} (\vec{S}_i \cdot \vec{S}_j)^2}_{\text{favors nematic}}$$

[Known since $\simeq 1970$]

2. Geometrical frustration

Eg: $S=1$ Kagome AF with strong easy axis anisotropy

$$H = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j - D \sum_i S_{iz}^2$$

(Damle, TS '06)

$D \gg J$: $\langle \vec{S} \rangle = 0$ but $\langle (S_i^+)^2 \rangle \neq 0$

Spin liquids and other exotica in quantum magnets

- Traditional quantum magnetism: Ordered ground states

(Neel, spin Peierls, …………)

Notion of broken symmetry

Modern theory (last 2 decades): Possibility of `spin liquid' states

(well-known in d = 1, but also possible in any d).

Eg: Mott insulators with 1 electron/unit cell with no broken symmetry

Excitations with fractional spin (spinons),

Emergent gauge structure, notion of `topological order'

Maturing theoretical understanding -

extensive developments in last few years

But where are the spin liquids?

Almost no clear experimental sightings in d > 1 so far.

Hints from theory

- Geometrically frustrated quantum magnets

- ``Intermediate'' correlation regime

Eg: Mott insulators that are not too deeply into the insulating regime ("weak" Mott insulators)

- More subtle: Intermediate scale physics of doped Mott insulators (in cuprates?)

This talk – focus on specific candidate materials.

Three triangular lattice Mott insulators

1. Cs_2CuCl_4 – spin-1/2 on anisotropic Δ lattice

Ordered spiral but close to a spin liquid?

2. $\kappa(ET)_2Cu_2(CN)_3$ – "weak" Mott insulator.

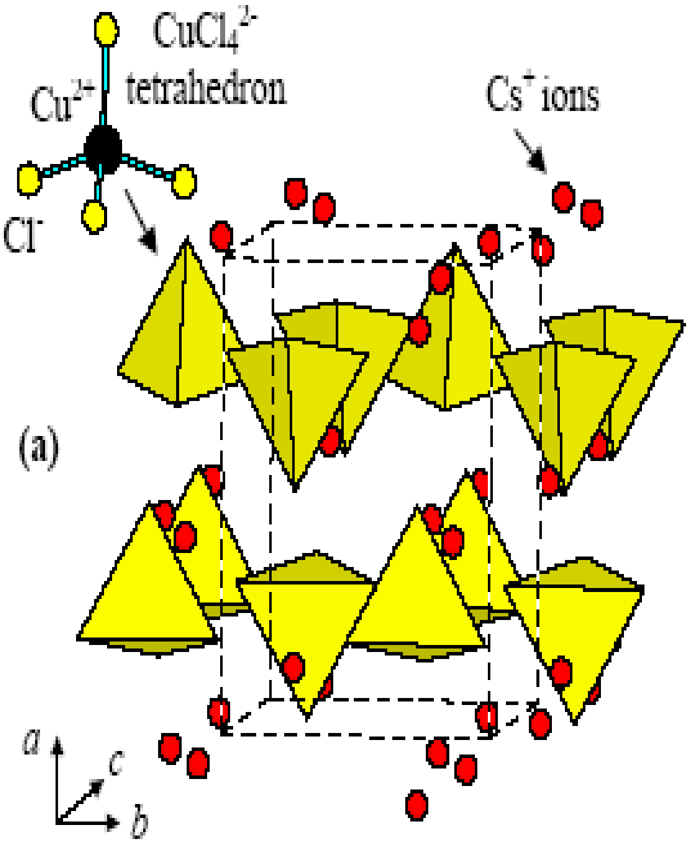
A genuine spin liquid?

3. $NiGa_2S_4$ – spin-1 on isotropic Δ lattice

Spin liquid or spin nematic?

Very promising candidate for exotica - Cs_2CuCl_4

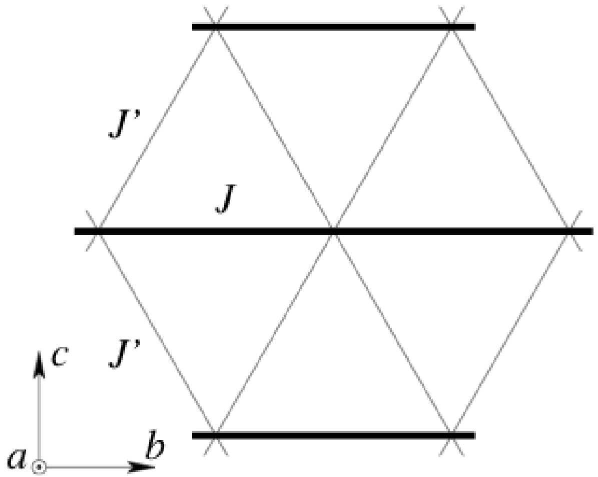
- Transparent layered Mott insulator
- Spin ½ per Cu site on anisotropic triangular lattice



Experiments

Radu Coldea's group
@ Oxford 1996 - present

Known microscopic spin Hamiltonian

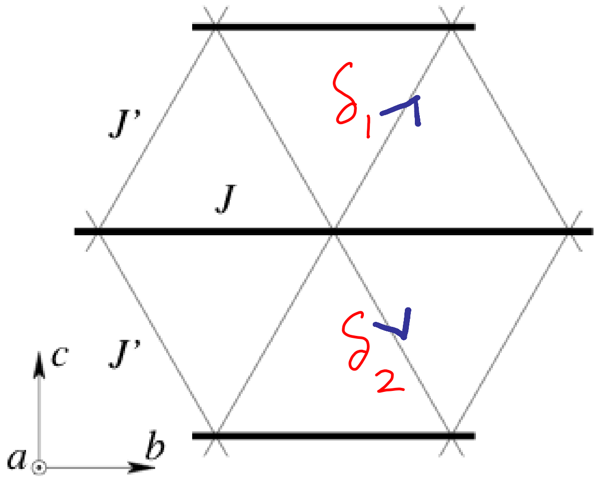


$J \approx 0.375\text{meV}$

$J' \approx J/3$

$J'' \approx 0.045\ J$ (weak interlayer exchange)

Known microscopic spin Hamiltonian



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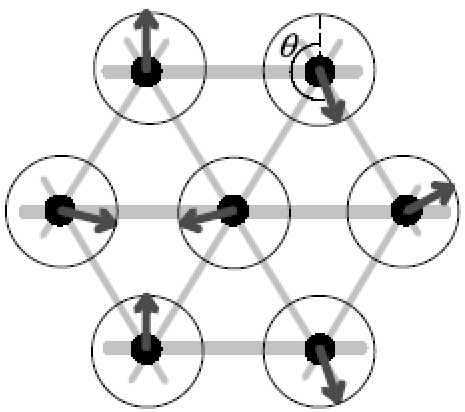
Weak Dzyaloshinski-Moriya interaction along zigzag bonds

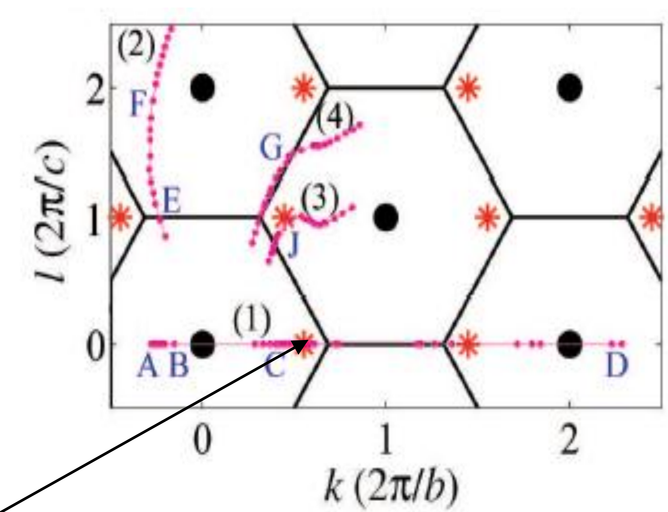
$$H_{DM} = -\mathbf{D} \cdot \sum_{\mathbf{r}} \mathbf{S}_{\mathbf{r}} \times (\mathbf{S}_{\mathbf{r}+\boldsymbol{\delta}_1} + \mathbf{S}_{\mathbf{r}+\boldsymbol{\delta}_2})$$

$$D \approx 0.02meV \approx 0.05J$$

Ordering at low T

Magnetic long range spiral order
below T=0.62K with incommensurate wave vector





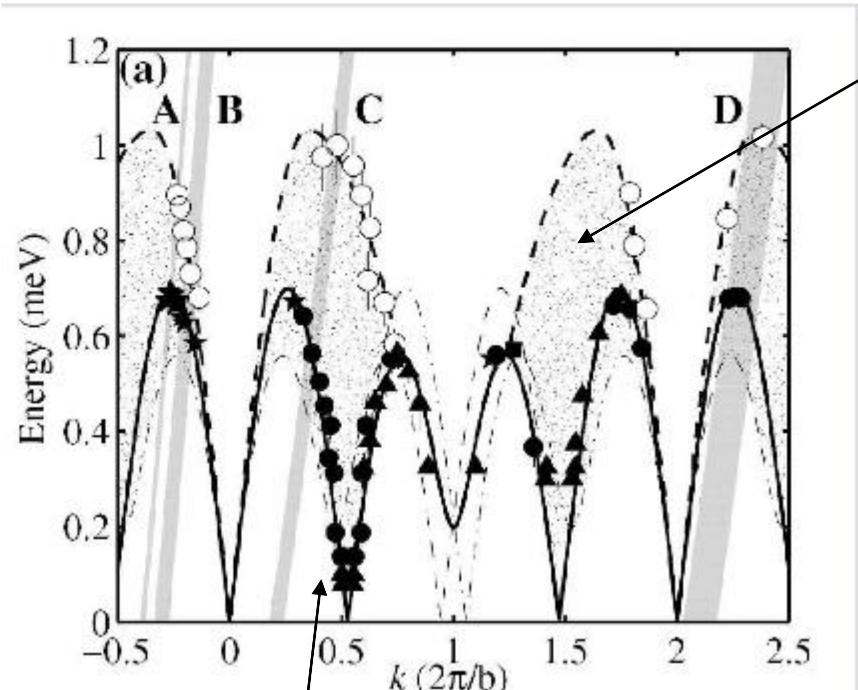
$$\mathbf{Q} = (0.5 + \epsilon_0)\mathbf{b}^*$$

$$\epsilon_0 = 0.030(2)$$

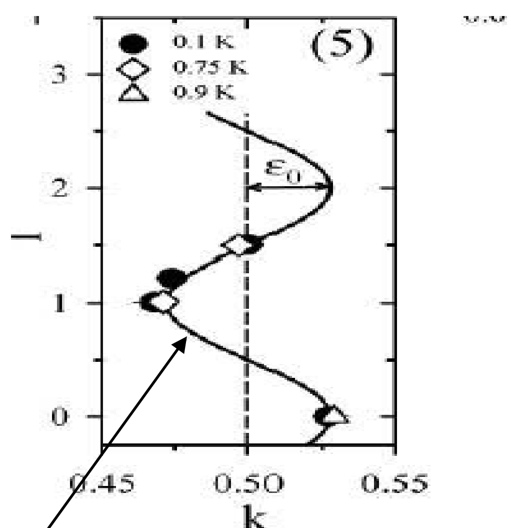
$$\mathbf{b}^* = (2\pi/b, 0, 0)$$

But many unusual phenomena en route!

Spin fluctuation spectrum



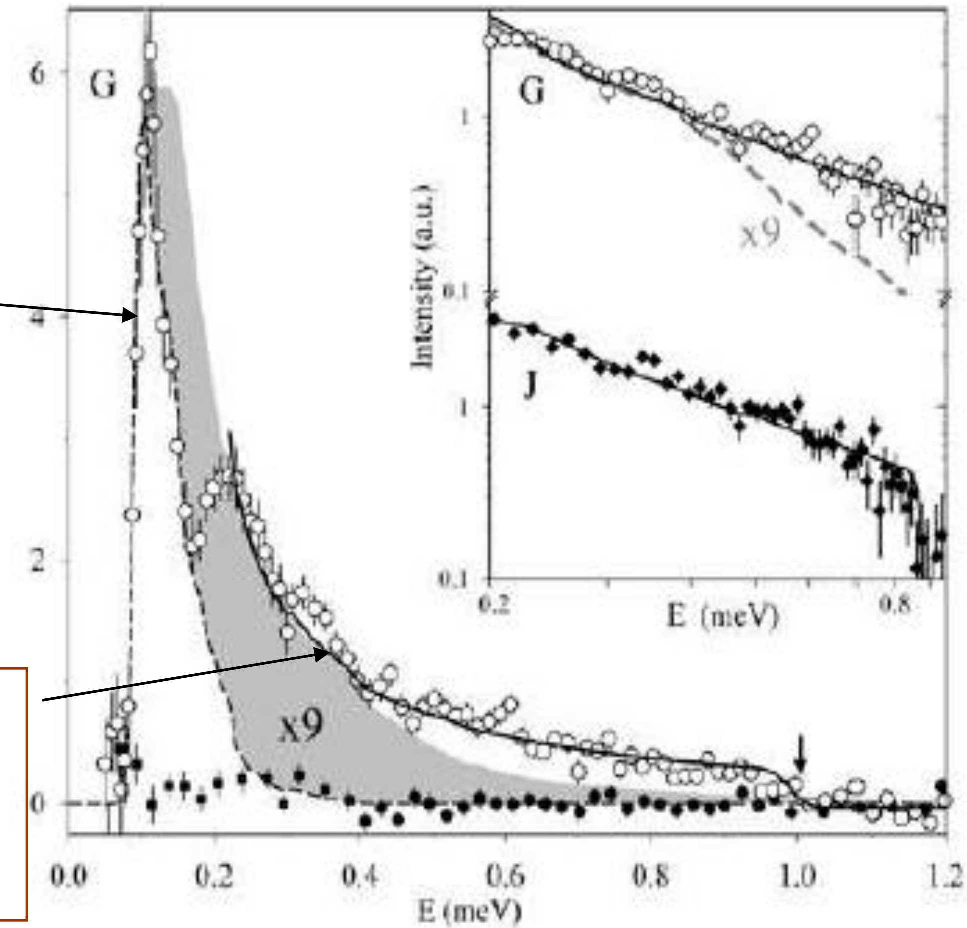
Large high energy continuum
(not contained in spin wave theory)



Low energy gapless magnon (as expected) – two dimensional dispersion

Inelastic line shape – failure of spin wave theory

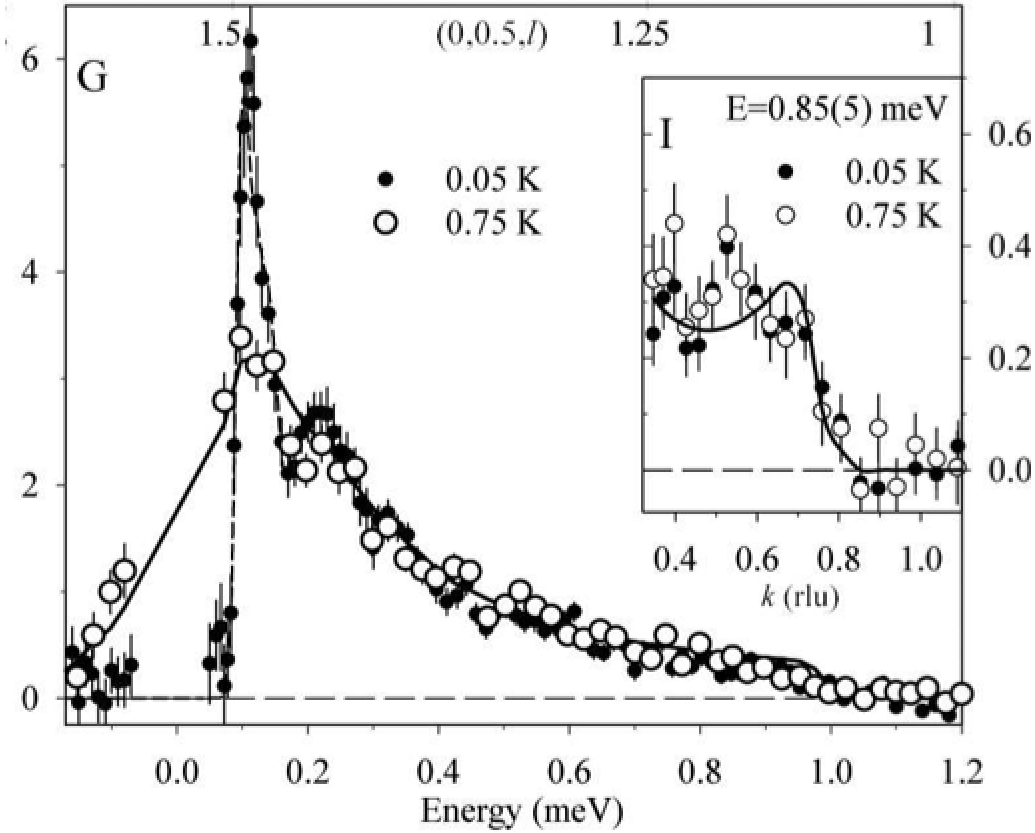
Spin wave peak



Broad inelastic background from 0.2 meV to 1 meV

Possibly power law, fit to estimate exponent.

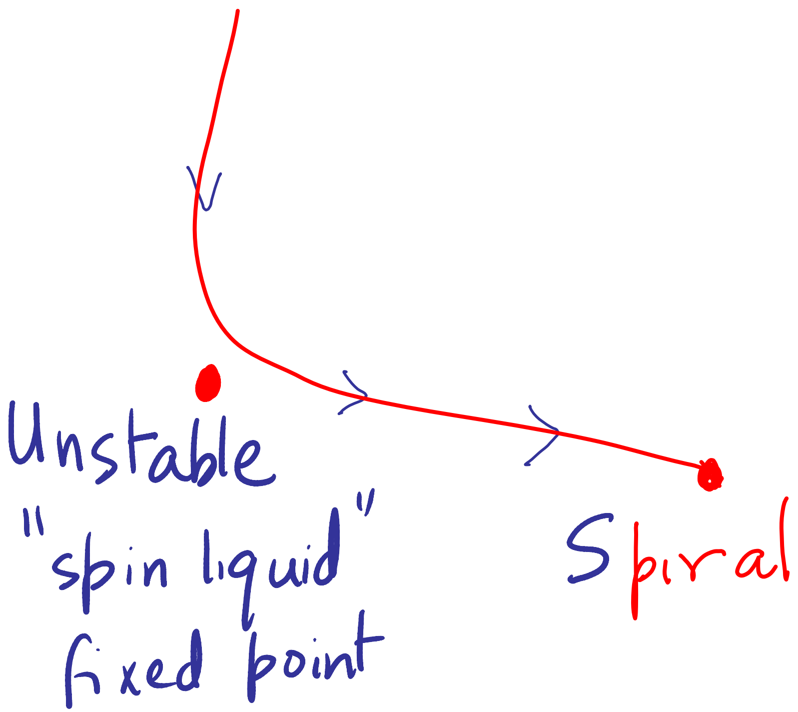
Temperature dependence



Magnon shoots out of broad background on cooling below T_N.

General qualitative similarity to antinodal ARPES in underdoped cuprates.!

General viewpoint on the experiments



Unstable fixed point controls
broad continuum scattering.

??Nature and description??

General framework:

1. Two dimensional but anisotropic

2. Spin SU(2) invariant
(DM small effect in this energy range)

3. Scale invariant?

Candidates (in order of increasing sophistication)

1. Decoupled 1d chains (Essler, Tsvelik,……)

2. Proximate quantum critical point
to gapped spin liquid (Isakov, TS, Kim)

3. `Algebraic spin liquid'
(Gapless fermionic spinons coupled
to fluctuating gauge field) (Zhou, Wen)

4. Algebraic vortex liquids (Alicea, Motrunich, Fisher)

5. Proximate quantum critical point to
dimer ordered (spin Peierls) state (No theory yet!)

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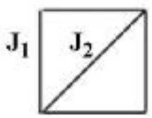
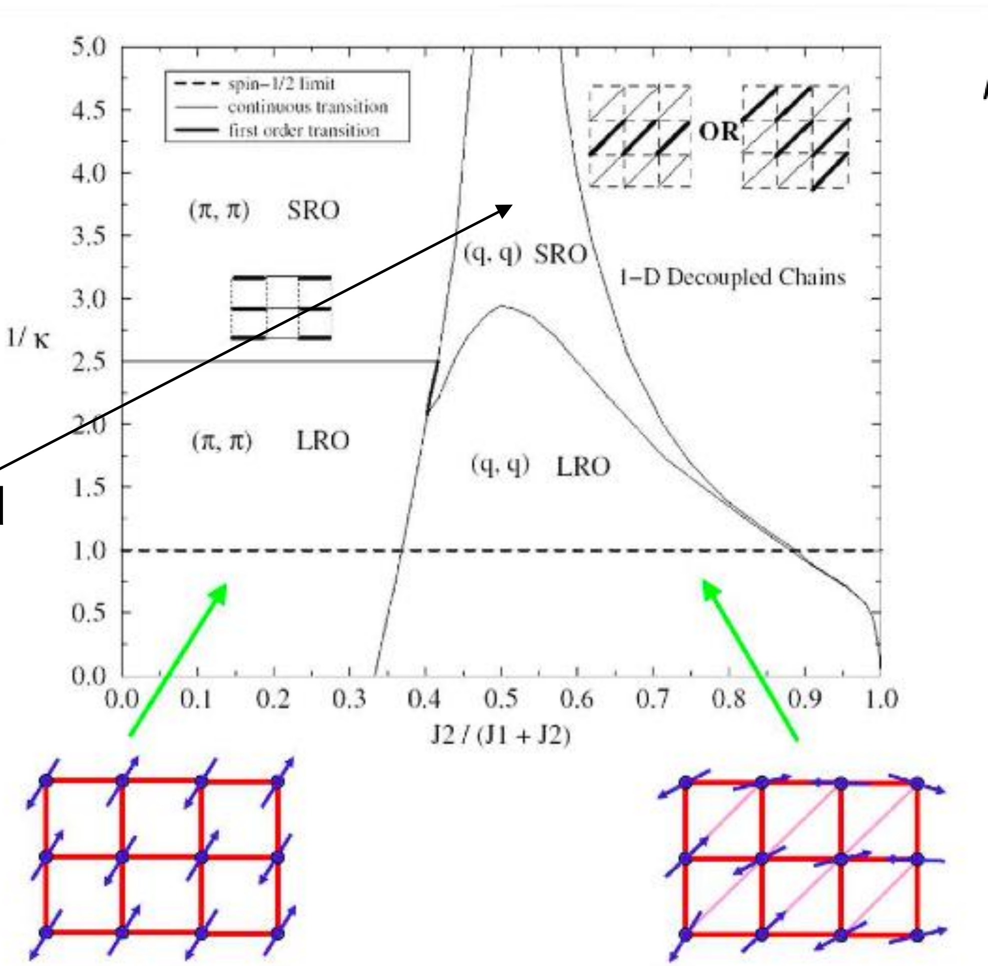
Large-N Phase Diagram - Anisotropic Triangular Lattice

Chung et al
2001, 03





Spin liquid

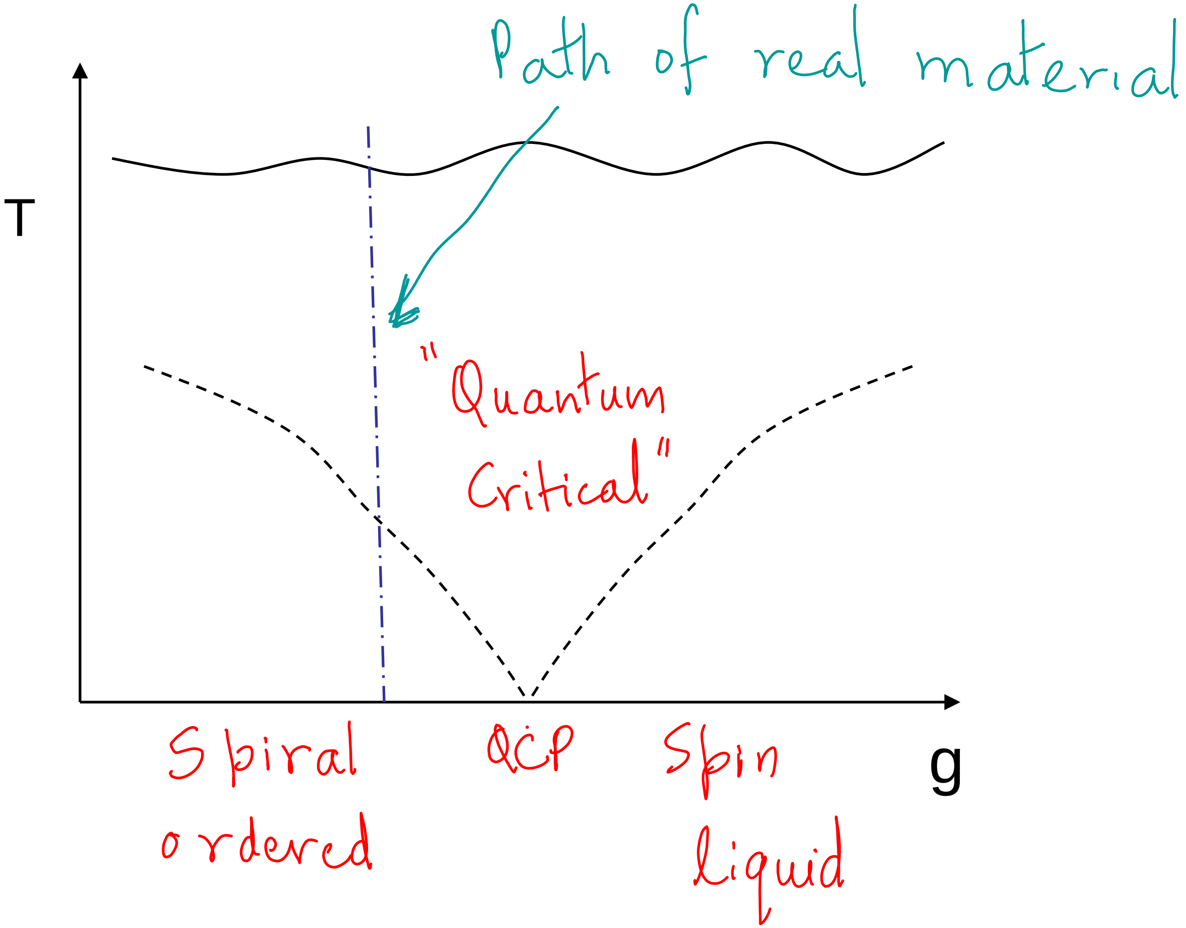
$\kappa = \text{“}2S\text{”}$

controls quantum fluctuations

$\mathbf{Q} = (q_x, q_y)$

magnetic ordering wave vector

Crossovers near phase boundary to spin liquid



Structure of spiral ordering

$$\underline{S}(\underline{r}) \sim \hat{n}_1 \cos(\underline{Q}\cdot\underline{r}) + \hat{n}_2 \sin(\underline{Q}\cdot\underline{r})$$

$$\hat{n}_1^2 = \hat{n}_2^2 = 1 \quad ; \quad \hat{n}_1 \cdot \hat{n}_2 = 0$$

Order parameter = rotation matrix R

$\in SO(3)$

Topological defects in ordered state

$$SO(3) \sim \frac{SU(2)}{Z_2} \Rightarrow \text{order parameter lives in } S^3/Z_2$$

$$\pi_1\left(S^3/Z_2\right) = Z_2 \Rightarrow \text{point } Z_2 \text{ vortices}$$

Liberating spinons

Chubukov, Sachdev, TS '94

Quantum disorder spiral without proliferating Z_2 vortices

Isakov, TS, Kim '05

$\Rightarrow$ redundancy in $SU(2)$ representation of $SO(3)$ matrix unimportant

$SU(2)$ matrix $U = \begin{bmatrix} z_1 & z_2^* \\ z_2 & -z_1^* \end{bmatrix}$

(z_1, z_2) = spin-$1/2$ spinons!

Structure of the spin liquid

Paramagnetic phase :

z_α free & gapped (bosonic statistics)

Z_2 vortex survives with gap

Effective low energy theory

- deconfined Z_2 gauge theory

Topologically ordered Z_2 spin liquid

Spiral – spin liquid phase transition

Phase transition – condensation of z_α

$\begin{bmatrix} z_1 \\ z_2 \end{bmatrix}$ lives on S^3

Asymptotic critical theory

$$S = \int d^2x \, d\tau \, e^{-\frac{1}{2g}\left[(\partial_\mu z_1)^2 + (\partial_\mu z_2)^2\right]}$$

All anisotropies on S^3 irrelevant

$\Rightarrow$ classical $O(4)$ fixed point in $D=2+1$ space-time dimensions

Scaling of spin fluctuations

Physical spin ~ bilinear of spinons

$\Rightarrow$ Broad inelastic scattering

For $q \approx Q$, small ω, T

$$\chi''(q, \omega, T) \sim \frac{1}{\omega^{2-\eta}} F\left(\frac{|q-Q|}{T}, \frac{\omega}{T}\right)$$

$\eta \approx 1.37$ from Monte-Carlo (large!)

Experiments – hard to measure η

Rough estimate – large η (≈ 0.74) qualitatively consistent with theory

Useful future experiments

1. Careful measurement of inelastic line shape for various fixed $q \approx Q$.

2. NMR Relaxation

$$1/T_1 \sim T^{\eta} \approx T^{1.37}$$ in quantum critical region

Direct measure of η.

More dramatic consequence -enhanced O(4) symmetry

(Isakov, TS, Kim '05)

Under O(4) can rotate $\hat{n}_1$ or $\hat{n}_2$ to

$$\hat{n}_3 = \hat{n}_1 \times \hat{n}_2$$

(3rd column of rotation matrix)

$$\Rightarrow \langle \hat{n}_3(x)\cdot\hat{n}_3(0)\rangle = \langle \hat{n}_1(x)\cdot\hat{n}_1(0)\rangle = \langle \hat{n}_2(x)\cdot\hat{n}_2(0)\rangle$$

$$\sim \frac{1}{x^{1+\eta}} \quad \text{with } \eta \simeq 1.35$$

Microscopics: $\hat{n}_3 \sim$ "vector spin chirality"

$$\sim \vec{S}(\underline{r}) \times \vec{S}(\underline{r}+\underline{a})$$

Detecting vector spin chirality fluctuations
-polarized inelastic neutron scattering
(Maleyev'95, Isakov,TS,Kim,'05)

Polarization dependent part

- antisymmetric component of spin structure factor $\sim \langle \vec{S}(0\,0) \times \vec{S}(\underline{r}\,t) \rangle$

Zero with full SU(2) spin symmetry

Non-zero due to weak Dzyaloshinski-Moriya D

To 1st order in D, measure vector spin chirality correlations

Expected result in quantum critical regime

For $q = Q$, $\omega \gg T$,

polarization dependent part

$$R_p(\omega) \sim \frac{1}{T^{2-\eta}\,\omega^{\frac{5-\eta}{2}}} \qquad (\eta \approx 1.37)$$

(to linear order in D)

Difficult but doable experiment.

Summary on Cs_2CuCl_4

- Concrete version of general idea that Cs_2CuCl_4 is proximate to a spin liquid

Many testable consequences.

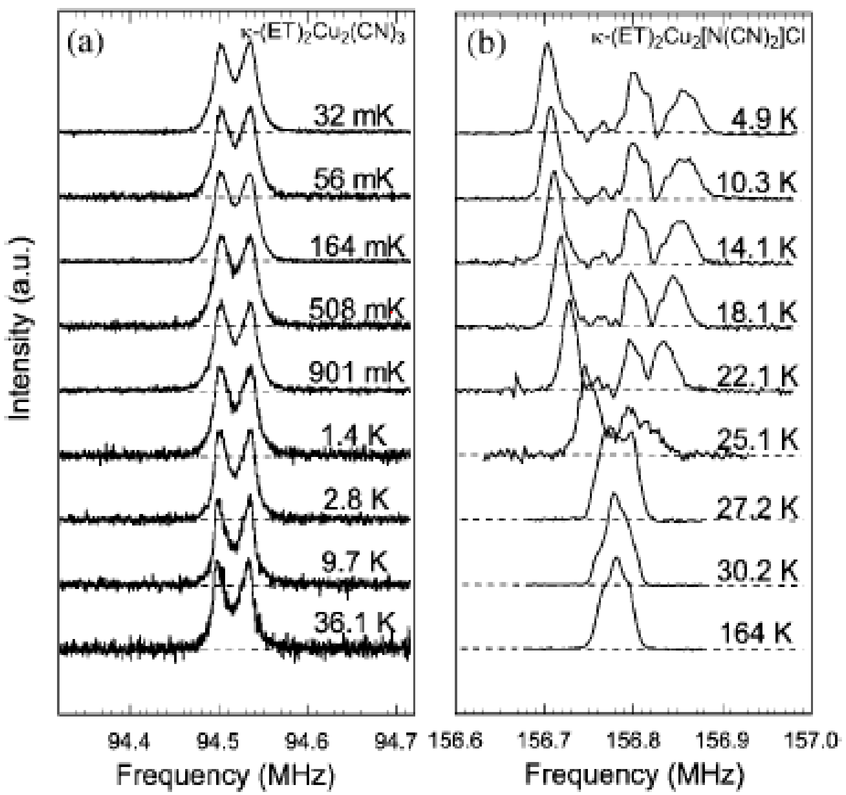
Wish-list for experiments:

1. Careful frequency scans at fixed wavevector
2. NMR relaxation
3. Polarized inelastic neutron

Future theory: Direct second order spiral – dimer transition?

$\kappa(ET)_2Cu_2(CN)_3$ – LONG SOUGHT SPIN LIQUID?

(Expt: Kanoda, 2002 - present)



. Weak Mott insulator close to Mott transition.

$\approx$ Isotropic Δ lattice

. No ordering to 32 mK $\ll J \approx 250$ K

but $\chi \to$ const.

$\frac{1}{T_1 T} \to$ const.

Promising candidate spin liquid state

Gutzwiller projected Fermi sea (Motrunich '05)

$P_G \,|\, \text{Fermi sea} \rangle$

remove double occupancy

½-filling : Spin wavefunction for particular spin liquid state.

Rough description : "Electrons which have lost their charge"

Fermi surface of neutral fermionic $S = \frac{1}{2}$ spinons.

PRECISE DESCRIPTION

(Motrunich '05, Lee & Lee '05)

Effective theory: Fermi sea of spinons coupled to fluctuating U(1) gauge field.

$$H = \int d^2x \; f^\dagger \left(-\frac{(-i\vec{\nabla} - \vec{a})^2}{2m} - \mu \right) f + (\vec{\nabla} \times \vec{a})^2 + \vec{e}^{\,2}$$

$\vec{a}$ = internal vector potential

$\vec{e}$ = " electric field

Well studied in many different contexts

(Holstein et. al. '74, Reizer '89, Lee & Nagaosa, Ioffe, Millis, Altshuler, Polchinski ....)

Expected properties

$\chi \to$ const. at low-T

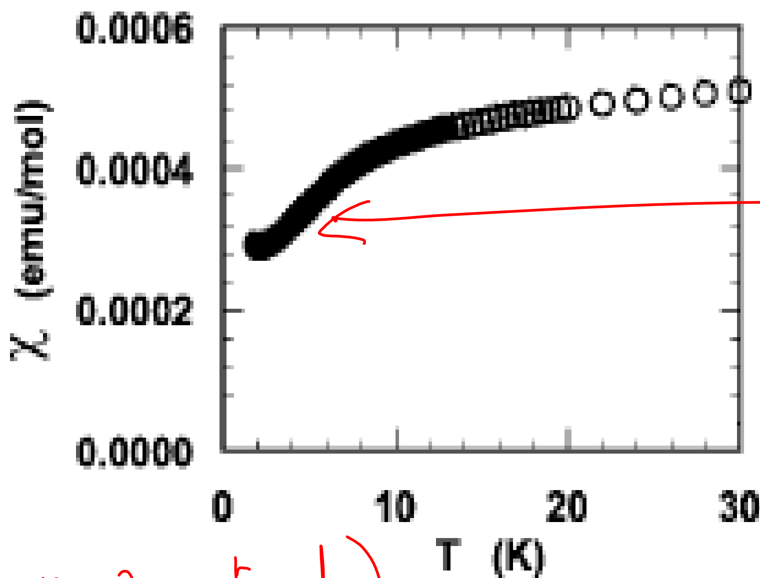
$C \sim T^{2/3}$ (enhanced by gauge fluctuations)

More entropy than in a metal at low-T!

Singular "$2k_F$" spin response.

Anamolous low-T thermal conductivity $\kappa \sim T^{1/3}$

Discrepancy at very low temperature



(Shimuzu et. al. '03)

Rapid decrease below 10 K before saturating as $T \to 0$

Specific heat (Nakazawa et. al.)

$C/T \to$ const. as $T \to 0$ (& not $T^{-1/3}$)

But low-T Wilson ratio ≈ 1

Instability of the spinon Fermi surface state?

(Lee, Lee, TS)

χ, $\frac{C}{T}$ $\rightarrow$ const. in insulator

Gapless spinons but no gapless gauge field ?

Natural route : Pairing $\langle ff \rangle \neq 0$

gaps out gauge field but may preserve

gapless spinons.

Amperean pairing?

Gauge induced interaction - repulsive for $(\vec{k}, -\vec{k})$ fermions but attractive for fermions with $\parallel$ momenta (Ampere's law)

Mean field theory: Find pairing solution

with $\Delta_{\vec{Q}}(\vec{p}) = \langle f_{\uparrow}(\vec{Q}+\vec{p}) f_{\downarrow}(\vec{Q}-\vec{p}) - f_{\uparrow}(\vec{Q}-\vec{p}) f_{\downarrow}(\vec{Q}+\vec{p}) \rangle$

$\neq 0$

$\vec{Q} \in$ maximal curvature points on Fermi surface.

Properties of Amperean paired spin liquid

1. $\langle f f \rangle \neq 0 \Rightarrow$ U(1) gauge field gapped
(get Z_2 spin liquid)

2. Pairing only in some patches of FS
– "gapless" weakly interacting spinons at "residual FS"
$\Rightarrow C \sim T$ at low-T.

3. Break lattice symmetries – incommensurate valence bond solid order coexisting with spin liquid.

4. $\kappa \sim T$ like in ordinary metal.

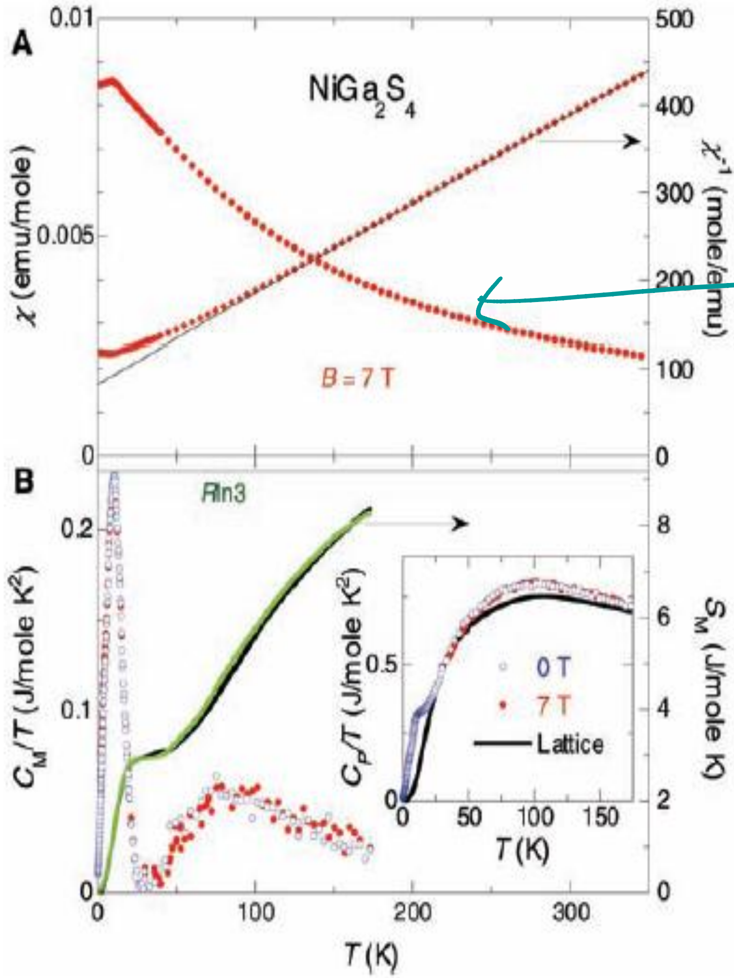
Crucial future experiment

Thermal transport $\kappa \sim T$ will be remarkable in an electrical insulator!

(Can distinguish from alternate scenario of Anderson insulator).

NiGa2S4: spin-1 triangular magnet

(Nakatsuji et. al., '05)



Curie Weiss of $S=1$;

$\theta_W \simeq -80$ K

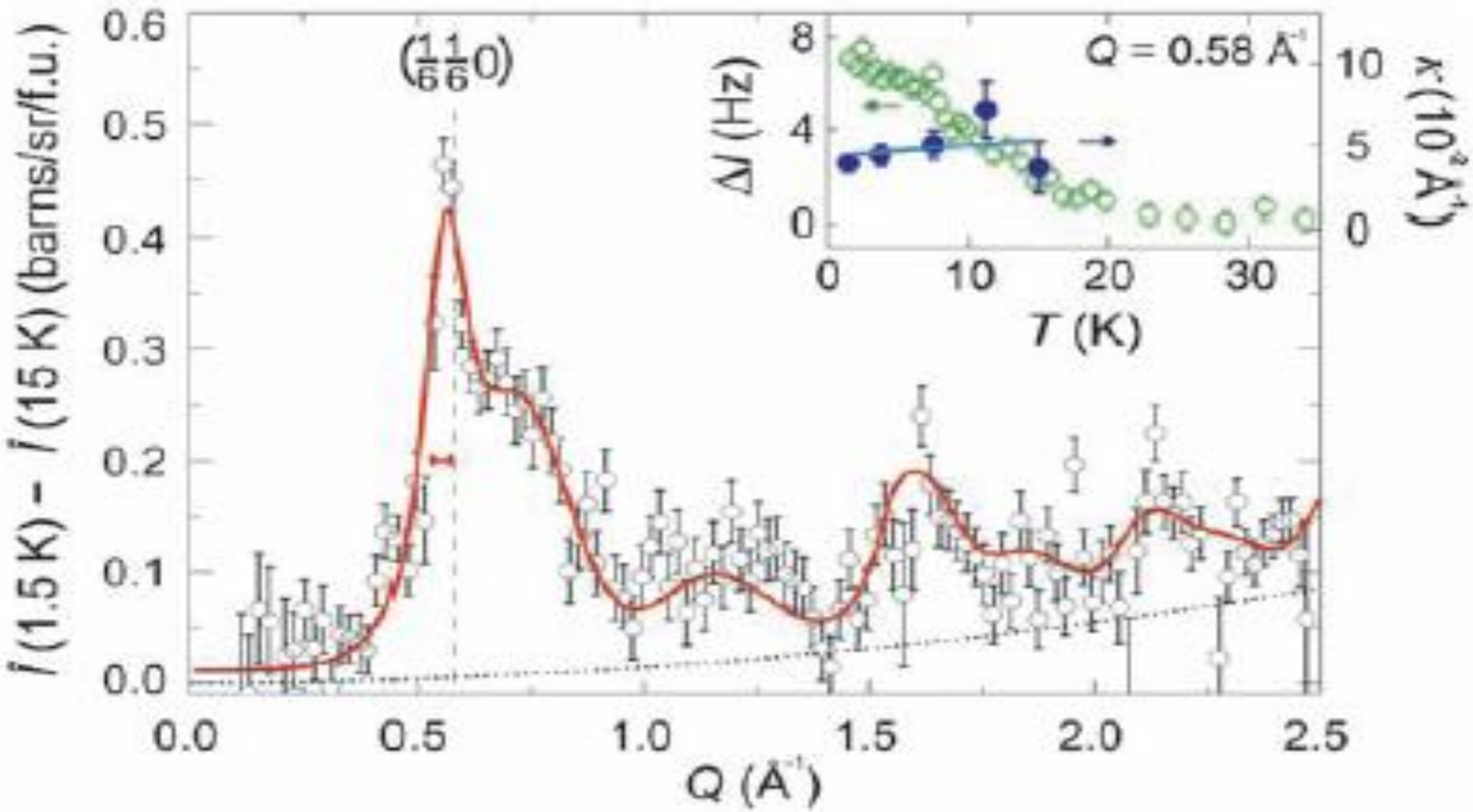
$\chi(T \to 0) \to$ const.

$C \sim T^2$ at low $T \lesssim 10$ K

1/3 of spin entropy recovered by ≈ 50 K

(Note: Polycrystalline samples)

Powder neutron scattering



No Bragg peak ; only short range order !

Quantum spin liquid or nematic?

No obvious (to me!) spin liquid explanation

Is this a spin nematic?

$\langle \vec{S} \rangle = 0 \Rightarrow$ No Bragg peaks in neutron expt

$$\left\langle \frac{\{S_\alpha, S_\beta\}}{2} - \frac{2}{3}\delta_{\alpha\beta} \right\rangle \neq 0 = q\left(n^\alpha n^\beta - \frac{1}{3}\delta^{\alpha\beta}\right)$$

$\Rightarrow$ Broken spin rotations & corresponding Goldstone mode gives T^2 specific heat.

Specific proposals

Non-collinear nematic (Tsunetsugu, Arikawa)

"Director" $\hat{n}$ along 3 $\perp$r directions on 3 sublattices

Collinear ferro-nematic (Lauchli et. al., Bhattacharjee, Shenoy, TS)

$\hat{n}$ uniform independent of site

How to distinguish?

Inelastic neutron scattering :

Collinear ferro-nematic : Spins fluctuate in plane $\perp$r to director $\Rightarrow$ anisotropic dynamic susceptibility

Non-collinear nematic : No single common plane for all spins.

Summary

- Many interesting new quantum magnets with unusual low temperature physics

- Future – most promising setting to establish the experimental validity of some modern ideas in strong correlation physics (fractionalized quantum liquids, emergent gauge theories, topological order, etc).