Quantum phases and transitions of spatially anisotropic triangular antiferromagnets

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Introduction to quantum magnetism on triangular and anisotropic triangular lattices

- Magnetic order, spin liquid, ...

- Cs$_2$CuCl$_4$, dimensional reduction, and low temperature phases of anisotropic triangular lattice antiferromagnets
Frustration and triangles

- Simplest idea: pairwise exchange interactions cannot be simultaneously satisfied

- But this is a bit simplistic, and overstates the problem

“geometric frustration”
Triangular Ising model

- Wannier (1950): thermodynamic ground state degeneracy

\[ \Omega = e^{S/k_B} \]

\[ S \approx 0.34Nk_B \]

- Classical Ising model does not order at any \( T>0 \)

- but very sensitive, and no known examples of this
Heisenberg Systems

\[ H = J \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j \]

- Unlike the Ising model, this is not intrinsically classical

- However, it turns out that the semi-classical “spin wave” expansion tends to work well even for small S (unfortunately!)

- We may still ask about the degeneracy in the classical limit
Classical ground state

- Spins must sum to zero
Classical ground state

Degrees of freedom: 2
Classical ground state

Degrees of freedom: 2+1
Classical ground state

Degrees of freedom: 2+1
order parameter is SO(3) matrix

\[\langle \vec{S}_i \rangle = \Re \left[ \psi(\hat{n}_1 + i\hat{n}_2)e^{iQ \cdot r_i} \right] \]
Quantum Spin Liquid (QSL)

- Anderson (1973) proposed Resonating Valence Bond (RVB) state of S=1/2 Heisenberg magnets

- Fazekas + Anderson (1974) made first calculations suggesting this for the triangular lattice in particular

\[ \Psi = \begin{align*}
\end{align*} \]

- Valence bond = spin singlet

\[ |V B\rangle = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle) \]
Reality: 120° state

- Semi-classical ordered state has been verified for $S=1/2$ spins by exact diagonalization and other numerics.
  - best estimate $M_s = 0.205 \pm 0.015$
  - compare full moment $M_s=0.5$, spin wave theory gives $M_s=0.24$
Hubbard Model

* If charge fluctuations are strong, reduction to the Heisenberg model is not accurate

\[
H = t \sum_{\langle ij \rangle} c_{i\alpha}^\dagger c_{j\alpha} + U \sum_i n_i(n_i - 1)
\]

\[
H = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j + K \sum_4 \left( P_{1234} + P_{1234}^\dagger \right)
\]

* Calculations (Imada, Motrunch, Lee) suggest QSL state at intermediate \( t/U \) (\( K/J > 0.15 \))
$\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ and EtMe$_3$Sb[Pd(dmit)$_2$]$_2$

- Organics proximate to a Mott transition
- Non-activated transport
- Optical pseudogap
Specific Heat

- Linear specific heat as expected for *free fermions* - approximately as expected for QSL - but very strange for an insulator

\[
\frac{C_p}{T} = \alpha T + \beta T^3
\]

\[
\frac{C_p}{T} = \gamma T^2
\]

S. Yamashita *et al.*, 2008
Challenges

- Thermal conductivity shows a gap
- Not consistent with uniform RVB state
- Consistency with specific heat?
- Different behavior seen for other organic very recently

M. Yamashita et al, 2008
Challenges

very small or no optical gap (pseudogap)

Készsmárki et al, 2006
Challenges

- Dramatic dielectric anomalies observed at T<60K
- Points to molecular dipoles in individual organic “dimers” - not taken into account by RVB theory

M. Abdel-Jawad et al, 2010
Spin $S=1$ antiferromagnets allow biquadratic exchange $K$

$$H = \sum_{\langle ij \rangle} \left[ J \vec{S}_i \cdot \vec{S}_j - K (\vec{S}_i \cdot \vec{S}_j)^2 \right]$$

This can induce quadrupolar order, or "spin nematic" state

$$\langle \vec{S}_i \rangle = 0$$
$$\langle S_i^\mu S_i^\nu \rangle = q \ n_i^\mu n_j^\nu$$
**NiGa$_2$S$_4$**

- Very 2d isotropic $S=1$ material

- Considered a candidate for spin-nematic state
  - Tsunetsugu *et al.*, 2006;
  - Bhattacharjee *et al.*, 2006;
  - Läuchli *et al.*, 2006;
  - Stoudenmire, LB, 2009

---

S. Nakatsuji *et al.*, 2005

![Graphs showing magnetic susceptibility and heat capacity](image)
T>0 Heisenberg Systems

- Mermin-Wagner theorem implies that 2d Heisenberg magnet cannot have magnetic order at T>0

- Non-linear sigma model: \( \xi \sim \exp \left( \frac{cJ}{T} \right) \), stiffness=0

- smooth crossovers seen in measured quantities

figures from classical MC simulations by Kawamura et al, 2010
T>0 Heisenberg Systems

- Mermin-Wagner theorem implies that 2d Heisenberg magnet cannot have magnetic order at T>0

figures from classical MC simulations by Kawamura et al, 2010

- Kawamura + Miyashita (1984) proposed a “topological transition” due to Z_2 vortices, point defects of the SO(3) order parameter

- existence of phase transition is controversial
Spatially anisotropic case

- Issues:
  - Is there a QSL state?
  - Magnetization plateaux in strong magnetic field?
Theories (zero field)

- Spin wave theory
  - $J'/J$
  - 0 0.27 1

- DMRG
  - disordered spiral
  - 0 0.8 1

- Variational QMC
  - 1d SL 2d SL spiral
  - 0 0.6 0.85 1

- Series expansion
  - spiral
  - 0 1

- Coupled cluster method
  - collinear spiral
  - 0 0.55 1
Materials

- \( \text{Cs}_2\text{CuCl}_4 : J'/J = 0.34 \)
- \( \text{Cs}_2\text{CuBr}_4 : J'/J = 0.5-0.7 \)
- \( \kappa-(\text{ET})_2X; X=\text{Cu}_2(\text{CN})_3, \text{Cu}[\text{N(CN)}_2]\text{Cl} : J'/J \geq 1 \)
- \( X[\text{Pd(dmit)}_2]_2; X=\text{EtMe}_3\text{Sb}, \text{EtMe}_3\text{P}, \text{Me}_4\text{P}... : J'/J? \)
- \( \text{NaTiO}_2 ? \)
- \( ^3\text{He on graphite?} \)
- \( \text{spiral AF} \)
- \( \text{spiral AF} \)
- \( \text{AF and spin liquid} \)
- \( \text{AFs, spin liquid, dimerized} \)
- \( \text{little studied} \)
- \( \text{spin liquid at 4/7 coverage} \)
Materials

- $\text{Cs}_2\text{CuCl}_4: J'/J = 0.34$
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- NaTiO$_2$?
- $^3\text{He}$ on graphite?

Organics close to the Mott transition. Hamiltonian must include charge fluctuations, and is not well understood.

- AF and spin liquid
- AFs, spin liquid, dimerized
- little studied
- spin liquid at 4/7 coverage
Materials

- $\text{Cs}_2\text{CuCl}_4 : J'/J = 0.34$
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- $\text{NaTiO}_2$
- $^{3}\text{He}$ on graphite?

- Spiral AF
- Spiral AF
- AF and spin liquid
- AFs, spin liquid, dimerized
- Little studied
- Spin liquid at 4/7 coverage

Orbital quasi-degeneracy probably is important.
Materials

- $\text{Cs}_2\text{CuCl}_4 : J'/J = 0.34$
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- $\kappa-(\text{ET})_2X; X=\text{Cu}_2(\text{CN})_3, \text{Cu}[\text{N}(\text{CN})_2]\text{Cl} : J'/J \geq 1$
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- $\text{NaTiO}_2$
- $^3\text{He on graphite?}$
- Spiral AF
- $\text{AF and spin liquid}$
- Experimental probes are limited, and there are competing models. Likely significant multi-spin ring exchange is important
- Spin liquid at 4/7 coverage
Materials

- $\text{Cs}_2\text{CuCl}_4 : J'/J = 0.34$
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- $\text{NaTiO}_2$?
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- spiral AF
- spiral AF
- AF and spin liquid
- AFs, spin liquid, dimerized
- little studied
- spin liquid at 4/7 coverage
Cs$_2$CuCl$_4$

- Cu$^{2+}$ spin-1/2 spins

- Couplings measured by measuring single-magnon spectrum in polarizing magnetic field (9T)

  \[ J = 0.37 \text{meV} \quad \text{chains} \]

  \[ J' = 0.34J \quad \text{diagonals} \]

  \[ J'' = 0.045J \quad \text{inter-layer} \]

  \[ D = 0.053J \quad \text{Dzyaloshinskii-Moriya} \]

R. Coldea et al, 2002
Frustration of interchain coupling makes it less “relevant”

First order energy correction vanishes

Interchain correlations are established only at $O[(J')^4/J^3]$!

Other smaller interchain interactions can dominate

Elementary excitations of 1d chains - spinons - form a good basis for excitations of the coupled system
Dimensional reduction?

* Numerics show that correlations are weak for $J'/J < 0.7$

Very different from spin wave theory

Very weak inter-chain correlations

Weng et al, 2006
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 $J'(k)>0$:
Low energy phases - zoology?

(a) $T < 0.2$ K

(b) $T < 0.1$ K

(c) $B \parallel a$: only one phase

$B \perp a$: many phases

$E_{\text{incommensuration}} = \frac{2\pi}{B(0)}$

$S_\alpha$ (arb. units)

$E_{\text{meV}}$

$B(T) \parallel a$

$B(\alpha, \beta)$

$S_\alpha$ (arb. units)

$B(0)$

$0, 0.5 - \epsilon, 1$

$T < 0.1$ K

$B_\parallel a$

$B_\parallel c$

$\text{Paramagnetic}$

$\text{SRO}$

$\text{Spiral}$

$\text{Cone}$

$\text{Ferromagnetic}$

$\text{Cs}_2\text{CuCl}_4$

$B_\parallel b$

$B_\parallel c$

Y. Tokiwa et al., 2006

R. Coldea et al., 2002
Strategy

- Quantitative understanding of high energy neutron scattering implies that a description in terms of weakly correlated spin chains is appropriate.

- Use the *low energy* field theory of the *decoupled* spin chains to study the low energy physics when they are coupled.

- This field theory is *critical* (i.e. the system is gapless), with conformal invariance.

- Use renormalization group methods to study the coupling.
Operators

- For example, in zero magnetic field:
  - \( N \): Néel vector
  - \( \varepsilon \): staggered dimerization

\( N > 0 \) \( \varepsilon < 0 \)
Inter-chain exchange

- Translates into operator couplings

- e.g. rectangular lattice

\[ H' \sim J' \int dx \mathbf{N}_y \cdot \mathbf{N}_{y+1} \]

- Triangular lattice

\[ H' \sim J' \int dx \mathbf{N}_y \cdot \partial_x \mathbf{N}_{y+1} \]

reflection symmetry
Power counting

- Operators “scale” like $L^{-\Delta}$, with scaling dimension $\Delta$
- e.g. $\Delta(N) = 1/2$.
- Dimensionless coupling, measured with respect to $v k \sim v/L$:
  - e.g. rectangular lattice $H' \sim J' \int dx \mathbf{N}_y \cdot \mathbf{N}_{y+1}$
  - $g'(L) \sim \frac{J' L \times L^{-2\Delta}}{v/L} \sim \frac{J'}{v} L^{2-2\Delta} \sim \frac{J'}{v} L$
- “relevant” interaction: generates Néel order at $kT \sim v/L \sim J'$
Power counting

* Operators “scale” like $L^{-\Delta}$, with scaling dimension $\Delta$

  * e.g. $\Delta(N)=1/2$.

* Dimensionless coupling, measured with respect to $v \kappa \sim v/L$:

  * triangular lattice

    $$H' \sim J' \int dx \mathbf{N}_y \cdot \partial_x \mathbf{N}_{y+1}$$

    $$g'(L) \sim \frac{J' L \times L^{-2\Delta-1}}{v/L} \sim \frac{J'}{v}$$

  * “marginal” interaction: exponentially weak effects?
Fluctuations

- Relevant couplings between second neighbor chains are allowed

\[ H'' \sim g_2 \int dx \mathbf{N}_y \cdot \mathbf{N}_{y+2} \]

- Interactions of this type are generated by fluctuations

  - A nasty calculation shows \( g_2 \sim (J')^4 / v^3 \)
  
  - Leads to commensurate, collinear AF order with \( kT_c \sim g_2 \)

O. Starykh + LB, 2007
## Theories (zero field)

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</table>
Cs$_2$CuCl$_4$ has a *spiral* ground state!

This is due to Dzyaloshinskii-Moriya interactions

\[
H_D = D \hat{z} \cdot S_{x,y} \times \left( S_{x-\frac{1}{2},y+1} - S_{x+\frac{1}{2},y+1} \right)
\]

This leads to spiral order with

\[
H_D \sim D \int dx \hat{z} \cdot \mathbf{N}_y \times \mathbf{N}_{y+1}
\]

\[
kT \sim D \sim 0.05J >> (J')^4/v^3 \sim (0.3)^4J \sim 0.008J
\]
Behavior in a field

- The approach is readily generalized to arbitrary magnetic fields
- Need to understand the operator content of the Heisenberg chain in a field
- Crucial fact: anisotropic correlations

\[ N = \begin{pmatrix} N_x \\ N_y \\ N_z \end{pmatrix} \]

\[ N^\pm \quad \Delta^\pm < 1/2 \]

- XY-like correlations transverse to field

\[ S^z_{\pi \pm 2\delta} \quad \Delta_z > 1/2 \]

- Incommensurate SDW correlations parallel to field

\[ 2\delta = 2\pi M \]
Ideal 2d model: J-J’ only

* Competition between longitudinal (less frustrated) and transverse (less fluctuating) couplings

\[ T \approx \frac{(J')^4}{v^3} \quad M = \frac{1}{3} M_{sat} \quad \sim 0.7 H_{sat} \quad H_{sat} \]

Plateau
Ideal 2d model: J-J’ only

- Competition between longitudinal (less frustrated) and transverse (less fluctuating) couplings

\[ T \approx \frac{(J')^4}{v^3}, \quad M = \frac{1}{3} M_{sat}, \quad \sim 0.7H_{sat}, \quad H_{sat} \]

up, up, down state
**Cs$_2$CuBr$_4$**

- Isostructural to Cs$_2$CuCl$_4$, but with larger $J'/J = 0.5 - 0.8$
- Shows a well-formed magnetization plateau at $M_{sat}/3$

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T. Ono *et al*, 2004

Fortune *et al*, 2009
But Cs$_2$CuCl$_4$ looks different

B $\parallel$ a: only one phase
B $\perp$ a: many phases
But Cs$_2$CuCl$_4$ looks different

Field and DM share the same “z” axis

$$H_D \sim D \int dx \, N_y^+ N_{y+1}^- + \text{h.c.}$$

The DM-induced order is strengthened by the field, because $\Delta$ decreases

“length” $|N^\pm|$ decreases approaching saturation
In-plane field

- DM and field compete to establish the “z” axis
- When $g \mu_B B \gg D$, the field establishes the axis:

$$H_D \sim D \int dx \ N_y^+ S_{2\delta, y+1} e^{i2\delta x} + \text{h.c.}$$

- Due to the oscillation, D is now strongly irrelevant
- Expect to see ideal J-J’ behavior?
No!

Reason:

* $J''$ exchange *between triangular planes* is unfrustrated and not suppressed by the field

* another strongly relevant perturbation

* It is only slightly weaker than $D$, and takes over when $D$ is washed out by the field

Y. Tokiwa et al, 2006
Due to small $J'' \ll J, J'$, it is natural to view $\text{Cs}_2\text{CuCl}_4$ in terms of correlated $b$-$c$ planes of spins.
Correlated planes?

* But actually, when the magnetic field is in the b-c plane, the spins are more correlated in a-b planes *perpendicular* to the triangular layers!
Extreme sensitivity

- Order in the a-b planes further amplifies the effects of very weak but unfrustrated interactions (of 1% of $J$ or so)
  - Second neighbor exchange $J_2$
  - Fluctuation-generated 4-spin interactions
  - Dzyaloshinskii-Moriya coupling $D_c$ on the chain bonds
Results vs. Experiment

For the ideal two-dimensional anisotropic triangular lattice antiferromagnet in a magnetic field and with a very weak second-neighbor and effective "biquadratic" exchange nontrivially competing for these regions we do not have reliable theoretical predictions at this point. A characteristic of frustrated systems. However, the existence of multiple ground states is a common feature of these systems. We must somehow explain major outstanding disagreements, which leave little room for doubt of their correctness with respect to the available neutron diffraction results. The anomalies discussed above are shown in Fig. 10.

The remainder of the paper is organized as follows. In Sec. II, we present some necessary background, including the renormalization group and chain mean-field theories. Based on the "standard" model, we are able to distinctly identify the gCMFTh and theoretical phase diagrams of the material in magnetic fields. To account for some finite-temperature effects and for very weak symmetry-breaking terms, we have determined the ground state variety of very weak second-neighbor and effective "biquadratic" exchange. This approach has been quite successful for triangular antiferromagnets. Nevertheless, in some field orientations over most of the range of applied magnetic fields, the model appropriate to Cs$_2$CuCl$_4$ may be as large as 35% of the CuCl$_2$ layers. In the former, we find spin density wave spiral ground state and no discontinuity is seen in the increasing field and disappears above 6 T. Taking into account these correlations is crucial to obtaining the proper low-temperature phase diagrams. They lead to an enhanced sensitivity to some steps in the magnetization normalized by the applied field. This indicates strong zero-point quantum fluctuations.

FIG. 2: Schematic phase diagrams in the temperature-field plane. The spin correlations for 6 T, with experiments on Cs$_2$CuCl$_4$, are shown in Fig. 1, (a) and (b) for parallel fields B/a, B/b, and (c) for parallel field B/c. The former are characterized by the presence of a family of tetrahedral cone states and in the SDW a family of commensurate coplanar antiferromagnetic states. In the latter, we find spin density wave spiral ground state and no discontinuity is seen in the increasing field and disappears above 6 T. Taking into account these correlations is crucial to obtaining the proper low-temperature phase diagrams. They lead to an enhanced sensitivity to some steps in the magnetization normalized by the applied field. This indicates strong zero-point quantum fluctuations.
a commensurate state stabilized by very weak (fluctuation-generated) 4-spin interactions (and possibly J₂)
The AF state has a period of 4 chains in the triangular plane consistent with neutron scattering for $B \parallel c$ which observed a commensurate state with this periodicity (R. Coldea).

Commensurate state also observed by NMR in this field range (M. Takigawa)
Results vs. Experiment

An incommensurate state, in which the AF order is deformed by $D_c$.
Incommensurate state deformed from AF one by spiraling spins in opposite directions on even and odd chains.

Evidence for incommensurate state seen in NMR (Takigawa).

Neutron data looks very similar to AF state. However, wavevector of incommensurability is expected to be very small (<2%), which may be too small to resolve.
Summary

- Frustration has strong effects on $S=1/2$ spatially anisotropic triangular antiferromagnets
  - It greatly enhances the regime of quasi-one-dimensionality
  - It leads to tremendous sensitivity to weak perturbations, producing a very complex phase diagram
  - Some additional source of fluctuations (other interactions, charge fluctuations...) is needed to obtain a true QSL state
Summary


![Phase diagrams](image)

(a) $h \parallel a$

(b) $h \parallel b$

(c) $h \parallel c$

(d) Ideal 2d model