Quantum phases and transitions of spatially anisotropic triangular antiferromagnets

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Collaborators

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- Masanori Kohno, NIMS, Tsukuba, Japan
- Hosho Katsura, KITP
Outline

- Introduction: quantum spins on the triangular lattice and the search for the Quantum Spin Liquid
- \( S=1/2 \) materials
- Spatial anisotropy, dimensional reduction, and a phase diagram
- Extreme sensitivity and \( \text{Cs}_2\text{CuCl}_4 \)
Bizarre Love Triangles
Too bizarre...
Triangular Ising model

- Wannier (1950): thermodynamic ground state degeneracy

\[ \Omega = e^{S/k_B} \approx 0.34 N k_B \]

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

- Similar to spin ice

1 frustrated bond per triangle
Heisenberg Systems

\[ H = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j \]
Heisenberg Systems

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

• Classically

• but small Heisenberg spins are not intrinsically classical
Classical ground state

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

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

* Resonating Valence Bond (RVB) state
  * Valence bond = spin singlet
  \[ |VB\rangle = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle) \]

\[ \Psi = \]

* no broken symmetries but “topological” character

Fig. 3: Random arrangements of pair bonds on a triangle lattice. (a) Shows a regular arrangement with 2N/4 alternative distinct configurations (“rhombus” approximation). (b) An arbitrary arrangement. While only \((S_1S_3)\) gives a matrix element of opposite sign. Thus we can always gain energy by linearly combining different configurations in which such bonds are interchanged. Since there are in any random configuration like Fig. 3b great numbers of sets of parallel bonds, one can arrive at any configuration from any other; and return to the original one by very many paths. What is not clear is that one will return to the same state in the same phase by traversing different paths. If one did, the state would be essentially a Bose condensed state of pair-bonds with a form of ODLRO. This would be closely related to the liquid state of pair-bonds with a form of ODLRO.
Reality: 120° state

* 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 $M_s=0.24$

Bernu et al, 1992+...
Other parameters

* To find exotic states, one needs to expand the phase space...
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, Motrunich, Lee\textsuperscript{2}) suggest QSL state at intermediate \( t/U \) (\( K/J > 0.15 \))
Organics proximate to a Mott transition

Some suggestive experiments support QSL behavior but many questions exist...a subject for another talk!
To find exotic states, one needs to expand the phase space...
**S=1 Systems**

- 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

\[
\langle \vec{S}_i \rangle = 0
\]

\[
\langle S_i^{\mu} S_i^{\nu} \rangle = q n_i^{\mu} n_j^{\nu}
\]

Läuchli *et al.*, 2006
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
- However, gradual and partial freezing below 8K is not understood, and there are other scenarios
  c.f. H. Kawamura

S. Nakatsuji et al, 2005
Other parameters

- To find exotic states, one needs to expand the phase space...

\[ S = \frac{1}{2} \]

Heisenberg
T>0 Heisenberg Systems

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

![Graphs showing correlation length ξ vs. T/J](image)

- Non-linear sigma model: ξ ~ exp (c J / T), 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
Other parameters

- To find exotic states, one needs to expand the phase space...

\[ S = \frac{1}{2} \tag{Heisenberg} \]

\[ \text{spin } s \]

\[ \text{spatial anisotropy} \]

\[ \text{itinerancy } (t/U) \]
Other parameters

* To find exotic states, one needs to expand the phase space...
Spatially anisotropic case

"chains"
Spatially anisotropic case

\[ \frac{J'}{J} \]

1d chains

\[ B, B_{\text{sat}} \]

spiral

?
Spatially anisotropic case
Theories (zero field)

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

- DMRG, ED
  - disordered, spiral
  - $0$, $0.8$, $1$

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

- Series expansion
  - spiral
  - $0$, $1$

- RG, coupled cluster method
  - collinear, spiral
  - $0$, $0.55$, $1$
Materials

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

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

Organics close to the Mott transition. Hamiltonian must include charge fluctuations. Proper model is still controversial.

- AF and spin liquid
- AFs, spin liquid, dimerized

Experimental Talks:
- T. Ito, Tues. 15:00
- R. Kato, Wed. 9:50
- M. Yamashita, W 12:00
Materials

- Cs$_2$CuCl$_4$ : $J'/J = 0.34$
- Cs$_2$CuBr$_4$ : $J'/J = 0.5-0.7$
- $\kappa$-(ET)$_2$X; X=Cu$_2$(CN)$_3$, Cu[N(CN)$_2$]Cl : $J'/J \geq 1$
- X[Pd(dmit)$_2$]$_2$; X= EtMe$_3$Sb, EtMe$_3$P, Me$_4$P... : $J'/J$?
- NaTiO$_2$?
- $^3$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

- $\text{Cs}_2\text{CuCl}_4 : J'/J = 0.34$
- $\text{Cs}_2\text{CuBr}_4 : J'/J = 0.5-0.7$
- $\kappa-(\text{ET})_2\text{X} ; \text{X} = \text{Cu}_2(\text{CN})_3 , \text{Cu}[\text{N(CN)}_2]\text{Cl} : J'/J \geq 1$
- $\text{X}[\text{Pd(dmit)}_2]^+ ; \text{X} = \text{EtMe}_3\text{Sb}, \text{EtMe}_3\text{P}, \text{Me}_4\text{P}... : J'/J$
- $\text{NaTiO}_2$
- $^3\text{He on graphite}$
- Experimental probes are limited, and there are competing models. Likely significant multi-spin ring exchange is important
- Spin liquid at 4/7 coverage
Materials

<table>
<thead>
<tr>
<th>Material</th>
<th>Exchange Ratio</th>
<th>Phase</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cs₂CuCl₄</td>
<td>J'/J = 0.34</td>
<td>spiral AF</td>
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<tr>
<td>κ-(ET)₂X; X=Cu₂(CN)₃, Cu[N (CN)₂]Cl</td>
<td>J'/J ≥ 1</td>
<td>AF and spin liquid</td>
</tr>
<tr>
<td>X[Pd(dmit)₂]₂; X= EtMe₃Sb, EtMe₃P, Me₄P...</td>
<td>J'/J?</td>
<td>AFs, spin liquid, dimerized</td>
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<td>NaTiO₂</td>
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<td>³He on graphite</td>
<td></td>
<td>spin liquid at 4/7 coverage</td>
</tr>
</tbody>
</table>
Spatially anisotropic case

Can we approach from the 1d regime?
Dimensional reduction

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

\[ \text{Interchain correlations are established only at } O(J')^4/J^3! \]

- 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 $J'/J < 0.7$

   ![Graph showing correlation strength for different coupling ratios](image)

   - Very different from spin wave theory
   - Can we check this experimentally?

   Ref. 3–4

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   Ref. 4–8


   Department of Physics and Astronomy, California State University, Northridge, California 91330, USA

   Center for Advanced Study, Tsinghua University, Beijing 100084, China

   The isotropic spin-1/2 Heisenberg model on a triangular lattice was a candidate for the realization of a disordered spin-liquid phase, which may be described by a minimal model with an exponential-decay spin correlator along the weaker coupling direction, consistent with the very weak inter-chain correlations.

   The characterization of such a spin-liquid ground state as revealed by recent numerical studies based on exact diagonalization for the magnetic phase diagram of the HAFM model in the space of nearest-neighbor couplings may also have a spin-liquid intermediate range three-sublattice spiral Néel ordered phase occurring at an energy to serve as a direct relevant to the quantum magnet in the Cs$_2$CuCl$_4$ anisotropic compounds, which may be described by a minimal model with an exponential-decay spin correlator along the weaker coupling direction, consistent with the very weak inter-chain correlations.

   Furthermore, outside this region, PBC always gives a smaller ground state energy than the energy from the ED calculation. By keeping up to an intermediate size of 8 lattice agrees quite well with the DMRG results for 6 lattice, similar error bars were obtained for both six-leg and eight-leg models, excluding the possibility of the system developing an exponential-decay behavior of the spin correlator, we establish the existence of the spin-liquid phase demands further theoretical studies.
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
Excitations

* Theory: 2d excitations are built from small numbers of 1d spinons

* Diagonalize inter-chain coupling (J’) in this basis

* Leads to direct calculation of neutron spectra with no adjustable parameters

Broad lineshape: “free spinons”

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

peak locations well-described by quasi-1d theory
Bound state

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

![Graphs showing spectra comparison](image)
Spatially anisotropic case

1d chains

1d theory and its excitations form a good basis for studying systems with $J'/J < 0.6$
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
    - \( \varepsilon > 0 \)
    - \( \varepsilon < 0 \)
  - \( \varepsilon \): staggered dimerization
Inter-chain exchange

* Translates into operator couplings

* e.g. rectangular lattice

\[ H' \sim J' \int dx \, N_y \cdot N_{y+1} \]
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 $v k \sim v/L$:
  - Triangular lattice
    \[ H' \sim J' \int dx \, N_y \cdot \partial_x N_{y+1} \]
  - \[ g'(L) \sim \frac{J' L \times L^{-2\Delta-1}}{v/L} \sim \frac{J'}{v} \]
- “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
Spatially anisotropic case

- $B_{\text{sat}}$
- $J' / J$
- $1d$ chains

- Cs$_2$CuCl$_4$
- Cs$_2$CuBr$_4$

- Collinear AF
- Spiral

- $J' / J$
- $0$
- $0.34$
- $0.5 - 0.7$
- $1$
## Theories (zero field)

<table>
<thead>
<tr>
<th>Theory</th>
<th>$J'/J$</th>
<th>$0$</th>
<th>$0.27$</th>
<th>$0.8$</th>
<th>$0.6$</th>
<th>$0.85$</th>
<th>$1$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spin wave theory</td>
<td></td>
<td>?</td>
<td>spiral</td>
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<td>DMRG</td>
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<td>variational QMC</td>
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<td>series expansion</td>
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</table>

- $J'$ is the effective exchange interaction parameter.
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

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

\[ \Delta_\pm < \frac{1}{2} \]

XY-like correlations transverse to field

\[ S_{\pi \pm 2\delta} \]

\[ \Delta_z > \frac{1}{2} \]

incommensurate SDW correlations parallel to field

\[ 2\delta = 2\pi M \]
Spatially anisotropic case

1d chains

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

$0.34$ $0.5-0.7$

collinear AF

cone

SDW

plateau

spiral

$M/M_{\text{sat}} = 1/3$

$B$ $B_{\text{sat}}$

$1$ $J'/J$

$J'$ $J$

$J$
For the anisotropic system, particularly near spin-wave expansion is, however, not suitable for studying overshadow those of spatial anisotropy. The standard Planar order can then emerge only if quantum effects however, reveal decidedly different behavior. In fields di-

While the UUD phase is well established for the iso-

A. Chubukov, 2001
J. Alicea et al, 2009
Spatially anisotropic case

May expect 1/3 plateau for all anisotropy
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 $M_{\text{sat}}/3$

T. Ono et al, 2004

Fortune et al, 2009
Spatially anisotropic case

May expect 1/3 plateau for all anisotropy
Low energy phases - \( \text{Cs}_2\text{CuCl}_4 \)

Not much similarity to theory!

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

\( B \perp a: \) many phases
Frustration: low energy behavior is *extremely sensitive* to weak interactions.
Other parameters

- Frustration: low energy behavior is extremely sensitive to weak interactions.

\[ S = \frac{1}{2} \]

Heisenberg

\[ D_a, J'', D_c < 0.05J \]
In zero field, Cs$_2$CuCl$_4$ has a *spiral* - not collinear - ground state!

This is due to Dzyaloshinskii-Moriya interactions

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

\[ H_D \sim D \int dx \ N_y^+ N_{y+1}^- + h.c. \]

This leads to spiral order with

\[ kT \sim D \sim 0.05J \gg (J')^4/v^3 \sim (0.3)^4J \sim 0.008J \]
Out of plane field

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

B \parallel a: only one phase
In-plane field

* DM and field compete to establish the “z” axis

* When $g \mu_B B \gg D$, the field establishes the axis:

$$H_D \sim D \int dx \, N^y_+ S^z_{2\delta, y+1} e^{i2\delta x} + \text{h.c.}$$

* 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

B⊥a: many phases
Correlated planes?

- Due to small $J'' << J, J'$, it is natural to view $\text{Cs}_2\text{CuCl}_4$ in terms of correlated b-c planes of spins
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

(b) $h \parallel b$

(c) $h \parallel c$

$B \perp a$: many phases

Y. Tokiwa et al, 2006
Results vs. Experiment

A commensurate state stabilized by very weak (fluctuation-generated) 4-spin interactions (and possibly $J_2$)

(b) $h \parallel b$

(c) $h \parallel c$

$B \perp a$: many phases
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

(b) \( h \parallel b \)

(c) \( h \parallel c \)

an *incommensurate* state, in which the AF order is deformed by \( D_c \)

B \( \perp a \): many phases
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


(a) \( h \parallel a \)

(b) \( h \parallel b \)

(c) \( h \parallel c \)

(d) Ideal 2d model

\begin{align*}
\text{Cone} & \quad \text{FP} \\
\text{DM spiral} & \quad \text{AF} \quad \text{Cone} \quad \text{FP} \\
\text{DM spiral} & \quad \text{AF} \quad \text{SDW} \quad \text{Cone} \quad \text{FP}
\end{align*}