Valence Bond Crystal Order in Kagome Lattice Antiferromagnets

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OUTLINE

• Motivation and Perspective
• Dimer Expansions: General Remarks
• Valence Bond Crystal (VBC) Order
• Honeycomb VBC State
• Excitation Spectra
• Experimental Relevance?
• Conclusions
MOTIVATION
Kagome Lattice Antiferromagnets

• A Lattice of Corner sharing triangles
• Several Experimental Realizations
  He3 on Graphite  (Elser, ……)
  Volborthite, Paratacamite, Kapellasite, ….

  Herbertsmithites

• Many variations/decorations
  Distortions, Further-neighbor exchanges, Ring exchanges, DM interactions, Impurities, …

• Quantum Spin Liquids?
Kagome Lattice Heisenberg Model
Important Theoretical Model

\[ H = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j \]

- On the verge of many instabilities
  - Magnetic order by disorder
  - Trimerization (Mila, Mambrini)
  - Valence Bond Crystals
  - Quantum Spin-Liquids of various kinds
- Mother of all Competing Orders? (MPA Fisher)
Magnetic Long Range Order

Many Candidates

TLM [root(3)by root(3)]
Q=0
Doubled Unit Cell along Y
Answer appears to be NO for spin-half KLHM

- Spectra from exact diagonalization
- Series expansions
- Other numerics
Exact Diagonalization of finite clusters (Elser, French Group, DMRG(Sheng))

GS Energy = -0.433, Triplet Gap ~0.055

About 200 singlet states below triplet for 36-sites
SINGLETS: RVB

- P. W. Anderson (and Fazekas) (TLHM)
- Quantum Dimer Models: (low energy subspace)
  - Hubbard ---- Heisenberg -----Dimers
  - Sutherland, Rokhsar-Kivelson, Moessner-Sondhi, Mila, …..
- Misguich, Serban and Pasquier
- Ralko, Trousselet, Poilblanc (Zeng-Elser)

KLHM maybe close to a Phase Transition
Quantum Spin-liquid Proposals

Variational (Projected) Wave Functions

- Marston/Zeng, Hastings
- Hermele, Ran, Lee and Wen
- Ryu, Motrunich, Alicea and Fisher
  Dirac Fermions, No spin-gap
- Ryu, Motrunich, Alicea and Fisher
  Gapless, Critical Spin-Liquids
- Hao and Tchernyshyov: Spinons are fermions but bound in pairs at low energies
  Spin-gap
QDM often lead to VBC
Many Possibilities Here Too
Large N: Max-Perfect Hexagons

Honeycomb
Stripes
Both have 36-site unit cells (Need different PBC)

Other VBC Budnik+Auerbach, Syromyatnikov and Maleev
Dimer Expansions
Perturbation Theory in Singlet/Triplet Basis
of a given DIMER COVERING

\[ 2^{2N} \text{ states} \quad \longrightarrow \quad (1+3)^{N/2} \text{ states} \]

Unperturbed Ground State

One Particle States \( (\text{Energy } J) \) (Triplons)

Two Particle States \( (\text{Energy } 2J) \) (2-Triplons)
Dimer Expansion: Illustrations

- Square
- Hexagon
- Octagon
- Linear Chain (Alternating Heisenberg Chain)
- Square Lattice (Columnar Dimer) (Read-Sachdev, ....)
- Majumdar-Ghosh Model
Single Square

2 singlets only
Easy to solve
Series expansion for small Loops
14th order Series

Square Compared with exact

Square, Hexagon, Octagon
Square, Hexagon, Octagon

Absolute Value of Coefficients

Partial sums after n-terms

Partial sums oscillate around zero
One-d Alternating Chain

More regular series but Power-Law singularity

Square-Lattice Heisenberg Model: Phase Transition to Neel Order RRPS, Gelfand, Huse
Majumdar Ghosh Model: Exact Ground State, No change
Dimer Expansion for KLHM
Empty Triangles are Key
The rest are in local ground state

Kagome Lattice

Shastry-Sutherland Lattice
Series Expansion around arbitrary Dimer Configuration

Graphs defined by triangles

All graphs to 5th order
Degeneracy Lifts in 3\textsuperscript{rd}/4\textsuperscript{th} Order But Not Completely

3\textsuperscript{rd} Order: Bind 3Es into H

4\textsuperscript{th} Order: Honeycomb over Stripe

Leftover: Pinwheels

24 \times 2^{(N/36)} Low energy states
Series show excellent Convergence

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<tr>
<th>Order</th>
<th>Honeycomb</th>
<th>Stripe VBC</th>
<th>36-site PBC</th>
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Ground State Energy per site

Estimated H-VBC energy: -0.433(1) (ED, DMRG)

36-site PBC: Energy=-0.43837653

Variational state of Ran et al (Hastings)-0.429
In this Scheme Auerbach and Maleev VBCs not favored.

VBC of Pinwheels has no Resonating Hexagons
36-site PBC wraps around
New graphs start contributing in 4th order
Closed Loops of 4 triangles
## Dimer Order Parameter

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<td>-.258</td>
<td>-.326</td>
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Resonance within hexagons appears not restored
Both strong and weak are stronger than mean
Mean energy per bond = -0.217

Pinwheels remain virtually unperturbed
Dimerization inside hexagons

Dimerization within Perfect Hexagons

Order of series

Absolute Value of Coefficients
Partial Sums
Excitation Spectra
First Block Diagonalize The Hamiltonian

Finite System or thermodynamic limit
Diagonalizing the Block Hamiltonian

In general:

- One-Particle states obtained by Fourier Transformation
- Two-Particle States: Need to solve the Schrodinger equation in relative coordinates

In all cases the spectra and spectral weight can be calculated by Linked-cluster methods
Large Unit Cell
One particle spectra: $18 \times 18$ Matrix

Heavy Triplets
and
Light Triplets
Shastry/Sutherland model

Yang, Kim, Yu, Park (Center Pinwheel)
9 By 9 matrix for Mobile Triplets

First Order,
z1, z3 are phase factors for q nonzero
Degenerate Perturbation Theory until Degeneracy Lifts
Then Non-degenerate Perturbation Theory

Q=0; First and Second order:

4 degenerate lowest energy states

(1,1,1, 0,0,0, 0,0,0) First Hexagon (local) (R3=1)
(0,0,0, 0,0,0, 1,1,1) Second Hexagon (local) (R3=1)
(2,-1,-1, 3,-3,0, 1,1,-2) (R3= +/-)
(0,-1,-1 1,1,-2, 1,-1,0) (R3= +/-)

Q nonzero: Only 3-degenerate states
Triplets localized at all scales

Loop: Hop along String of Green and Black
Cannot exit, has lowest energy
Yang, Kim, Yu and Park
Treat triplets as Bosons

Low energy structure agrees completely:
3 flat and one dispersive states that crosses them
Main difference is gap + small dispersion
Spin Spectra

Four-fold Degeneracy Lifts in 3^{rd} order:
Structure for Lowest 4-states
(very close in energy at q=0)
• One nearly flat and one dispersive band that meet at q=0
• Two nearly flat bands (hops from hexagon to hexagon)
Spin Gap

- Lowest triplet at q=0 (reduced zone)
  Gap Series:
  \[1 \quad -0.5 \quad -0.875 \quad +0.440625 + 0.07447 - 0.04347 - 0.02336\]
  Estimated Gap = 0.08 (0.02) (agrees with ED/DMRG)

- Lowest triplet for 36-site PBC
  Gap Series:
  \[1 \quad -0.5 \quad -0.875 \quad +0.440625 + 0.486458 - 0.16984 - \ldots\]
  Poorer convergence for 36-site
  Estimated Gap = 0.2 (cf 0.164 exact answer)
Is VBC Order consistent with ED?

Problems:
1. 200 states below triplet
2. Symmetry of Low Lying States: Includes R3 noninvariant ones
3. Dimer subspace Has continuous spectra

However:
Energy of PBC is much larger than separation (0.006 vs 0.001)
States are mixed strongly
Dimer order has not yet set in at this scale

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How many singlets below spin-gap?
Ground State manifold has 48 states
2-Triplet Bound states?

Pair of Light Triplets form stronger Bound States
Two-particle bound states on the 36-site PBC cluster

- Degenerate Perturbation theory
- Light-Heavy Bound states (9 x 9 matrix)
- Light-Light Bound states (36 x 36 matrix)
- Many bound states in 1st order
- 1st order Light-Heavy and Light-Light strongest bound states are very close in energy
- Second order: Only Light-Light remain at lowest energies
Two-particle bound states

- Many singlet bound states for 36-site PBC
- Four bound states are so tightly bound their energy falls below the triplet (~0.2 for PBC)
- \( E_b = 2*E(1p) - E(2p) \)
- 0.33, 0.29, 0.25, 0.25, 0.13, 0.13, 0.11, 0.11

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Low energy singlets

• Upto 48x5 states below Triplet (compare from about 200)
• What are the quantum numbers in the unbroken state? (requires a study of mixing which we have not done)
• What do the singlets look like?
Two triplets in separate Hexagons
Two-triplets in one hexagon

Two-particle Bound state in same hexagon can recover staggered dimerization of the perfect hexagons
Bound states with lowest energy triplets

Both triplets move around.

Rarely in same hexagon.

Mostly one in a Hexagon one on the bridges

May ultimately be the lowest excited state of all!

Made of Lowest triplets and No level repulsion

May explain quantum numbers of lowest states in ED
Recent work of Laeuchli and Lhuillier provides further support for VBC in Lowest Order

\begin{align*}
\text{g} & \quad 19.2 \\
\text{e} & \quad 0 \\
\text{f} & \quad 11.2 \\
\text{h} & \quad 2.4 \\
\text{d} & \quad 6.4
\end{align*}
Let Entangle!!

G. Vidal and co

MERA

Tensor Product based Annealing

VBC recovered variationally!!
Some similarity to Kagome Stripes

Azaria et al

DMRG
S. R. White and RRPS
Small gap (0.01) but robust dimerization
Experimental Relevance?

- Raman (Lhuillier, …)
- Long Range Order is fragile
  Temperature, Impurity, Phonons, DM …
- Short-Range Order: What is more robust?

Fluctuating interconnected hexagons with Light Triplets or
Pristine Pinwheels with Heavy Triplets

Pinwheels are Delta-chains protected from rest
Delta chains ubiquitous in all Dimer configurations
(Tchernyshyov)
Experimental Signature?

- Recent Neutron Spectra by DeVries et al
  ---- Spectral weight spread over wide range
  ---- from below 4 meV to above 30 meV
  ---- But with Q dependence essentially of Dimer
  ---- Pinwheel triplets ---localized two-spinons
  ---- Have spectral weight with Q dependence of dimer but spread over J/4 to 9 J/4
Heavy Triplets on Pinwheels break into kink-antikink pairs
Pinwheel Spectra (q-dependence of Dimer)
Single Dimer would be at one energy
Finite Kagome Clusters also show a similar continuum.

Laeuchli and Lhuillier

Short-range Valence Bond order
Dimer Liquid with short-range VB Order?

- Spectra also temperature insensitive
- Short range VB Order
- Classical Dimer Liquid?

\[ T_x = \frac{\Delta E}{\Delta S} \]

\[ = 0.375 \frac{J}{(\ln(2)-\ln(1.26))} = 0.8 \ J \]

\[ T_{VBC} = \frac{(.012 - - -.06)}{\ln(1.26)} = .04 - - 0.2 \ J \]

- Delta chains ubiquitous in Dimer phase
Summary

• VBC state has energy close to GS
• Dimerization weak inside the Hexagon but robust overall
• Spin-gap is small compared to J
• Lowest spin states: weak dispersion
• Singlet gap much smaller (bands?)
• Energy difference between VBC phases tiny
• VBC may be very fragile
• VB Liquid – Robust
• A robust signature: spectral features of Delta Chain?
THE END
Summary: Many Open Questions

- Is the ground state magnetically ordered?
- Is there a Quantum Spin-Liquid?
- Is the ground state a VBC?
- Is there an algebraic (power-law) phase?
- Is there a spin-gap?
- Are there fractional/topological excitations?
- Spinons and Visons

\[ H = \sum_{\langle ij \rangle} J_{ij} \hat{S}_i \cdot \hat{S}_j \]
Summary

Many Questions remain:

• Phase Transition: Order and Temperature?
• Susceptibility at low temperatures?
  ---Peak temperature set by gap?
• Sensitivity to Perturbations (Lattice Vibrations, Further Neighbor Js, Spatial anisotropy, DM, Impurity, Finite U …..)
• If there is an exotic phase, how can it be conclusively shown?
Calculations at finite T (HTE,NLC,ED)

Elstner and Young
Rigol and RRPS
Good Fit with
J=170K, g=2.19
CW is not asymptotic

Sharp upturn at low T not consistent with Kagome-HAFM
Dzyloshinski-Moria Interactions
Cross Product between spins: $D_z$

Both $D_z$ and $D_p$ are of order 10% of $J$ in structurally related Fe-based spin-5/2 material

$D_z$ can order the system!

Planar $+$ preferred helicity

Selects a unique Classical Ground state in the 120-degree subspace
Dzyaloshinski-Moria Interactions: Dp

\( Dp \) rotates from bond to bond

No spin symmetry left

Cannot be satisfied in 120-degree subspace

Classically a small \( Dp \) leads to canting—like a FM Ising anisotropy!
Susceptibility with Dp and Dz

Dz lowers susceptibility  
Dp increases susceptibility  
Both lead to anisotropy enhancing z-susceptibility
Entropy and Specific Heat

Dz: Entropy drops rapidly

Dp: No discernible change

What does Dp do to states?
Experimental Status Remains Fluid

- Number of recent papers claim large antisite disorder (6-10 percent)
- Number of very recent papers (Mendel, Keren) claim substantial anisotropy—DM and or Exchange (10 percent or larger)
- Some evidence of lattice freezing (NMR)
- Large magnetic entropy at low T not of isolated impurities—what is the origin?
- Direct probe of singlet sector?
Conclusions

- Kagome HM likely has a VBC ground state
- Very small energy scales between different phases
  What are the implications at finite T?
- If there is a transition what and how big would be the signature in entropy/specific heat?
- DM interactions are allowed—Will be there in any material realization—how large?
- Dz and Dp are quite different—the latter is more intriguing for avoiding LRO
- If there is an exotic state (such as Dirac spin-liquid) how can it be established?
Classical Heisenberg Models: Kagome Remains Highly Degenerate

Triangular-Lattice:
Edge sharing triangles

Kagome-Lattice:
Corner sharing triangles

Triangular Lattice has a unique Ground State up to Symmetries
Momentum Dependence
36-site PBC

• Four q points- 0(Gamma), K, Q, M
• Minimum triplet gap at q=M (French Group)
• Zeng+Elser Spin and Dimer Correlations (Huse+RRPS)
• Largest eigenvalues of correlation matrices

<table>
<thead>
<tr>
<th>q</th>
<th>spin-spin (3X3)</th>
<th>dimer-dimer(6X6)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.49998</td>
<td>0.4013</td>
</tr>
<tr>
<td>Q</td>
<td>0.35856</td>
<td>0.3375</td>
</tr>
<tr>
<td>M</td>
<td>0.43806</td>
<td>0.6736</td>
</tr>
</tbody>
</table>
Fluidity: Isolated spin-half objects must be free to flow

Deconfinement is the key property

Deconfined spinons and topological order

RVB state with free spinons

**P. Pukropas and P.W. Anderson, Phil Mag 30, 23 (1974).**

www.nordita.dk/~sylju/Nordic/meeting2/sachdev.ppt

May (must) have Topological Degeneracies
Spin-Liquids in 1D (Special Case)

- 1D QHM has a spin-liquid ground state
- As does the Majumdar-Ghosh Model

\[ \begin{align*}
\text{Heisenberg Ising} \\
\text{MG model}
\end{align*} \]
Triangular-Kagome Lattice Magnets
Canonical frustrated 2D spin systems

Triangular-Lattice:
Edge sharing triangles

Kagome-Lattice:
Corner sharing triangles

Kagome-Lattice is much more frustrated
(eg Ising entropy and correlation Length at T=0)
Quantum Heisenberg Model

\[ H = \sum_{<ij>} J_{ij} \hat{S}_i \cdot \hat{S}_j \]

\[ J_{ij} > 0 \]

\[ [\hat{S}^x_j, \hat{S}^y_j] = i\hbar \hat{S}^z_j \]

\[ \hat{S}^2_i = S(S + 1) \hbar^2 \]

Spin is a good quantum number

Pair of spins like to form rotationally invariant singlets state (Valence Bond)

Most interest in spin-half case
Specific heat sublinear at low-T?

Highly sensitive to magnetic field!

No clear sign of a transition

Or spin gap
Is the upturn due to impurities?
Not in a simple way

Rigol+RRPS
\[ c=0.04 \text{ Agrees to } 0.3 \text{ J} \]

Misguich+sindzingre
FM CW constant 6.5K
Agrees to 0.1 J
Where does the Kagome HAFM susceptibility peak? (T=0.1!)

Rigol+RRPS, Misguich+Sindzingre

Crossover to Reduced # of Localized Triplets?
Entropy and experiments: Assuming no frozen entropy, impurity

Misguich and Sinzindgre
High-T expansions

Lowering of entropy due to DM Interactions
Low Lying Spectra (Lhuillier)

Spectra of $H = \Sigma S_i \cdot S_j$
(per spin)

Square $N=36$

Kagomé $N=36$

triangular $N=36$

Extremely high density of low lying excitations
$\rightarrow$ Extra low temperature dynamics
Finite-T studies with DM Interactions (Rigol+RRPS)

- **D_z:**
  - Reduces entropy
  - Reduces uniform susceptibility
  - May lead to long-range XY order

- **D_p**
  - No change in entropy over a wide range of T
  - Increases susceptibility, makes it highly anisotropic
  - May lead to a non ordered ground state
Physics behind large number of singlets?

Fitting system-size data (upto \(N=36\)):
Exponentially large number of singlets? \(1.15^{\ast N}\)

Mila:
Short-range RVB subspace grows as: \(1.26^{\ast N}\)
There is a smaller subspace that grows as \(1.15^{\ast N}\)
Dimerize the up-triangles: $1.15^{**N}$

A triangle has four ground states: Pair of them have a unique ground state
Trimerized Models: Bands with varying number of defect triangles

\[ J'/J = 0 \]

\[ J'/J = 0.1 \]
Structure absent for $J'/J=1$

Structure like that of full diagonalization

Triplet energy is not a special mark

Mila’s conclusion: Physics in SR-RVB subspace but with no broken symmetries (caveat: size 36)
CORE: Auerbach and Buzdin
VBC Order at long scales

3SL

CD

\[ e_{12} \quad e_{21} \]
Second proposal for Large Number of Low-Lying Singlets

Kagome Stripes: Azaria et al

Gapless Singlet Modes and LSM Theorem

DMRG: SR White+RRPS : Spin-gap, VBC Order
Trimerization: Mambrini and Mila

Mila (PRL)
First attempt to explain Number of states growing as 1.15**N

Studied trimerized HM
Character of states changes close to uniform limit