Quantum Spin Liquids in Frustrated Magnets

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SB, C. Lhuillier, and L. Messio,
Phys. Rev. B 93, 094437 (2016).

SB, L. Messio, B. Bernu, and C. Lhuillier,
Phys. Rev. B 92, 060407(R) (2015).

L. Messio, **SB**, C. Lhuillier, and B. Bernu,
arXiv:1701.01253 (2017)

B. Fåk, **SB**, et al., Phys. Rev. B 95, 060402(R) (2017).

Reviews: Balents, Nature 464, 199 (2010); Norman, RMP 88, 041002 (2016); Zhou et al., arXiv:1607.03228 (RMP)

Collaborations

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Outline

- Ordinary liquids and quantum (spin) liquids
- Quantum spin liquids: general properties; RVB states
- Projective symmetry group classification: generalities
- Experimental candidates
- Kagome Heisenberg system: kapellasite
- Kagome with Dzyalioshinskii-Moiya term (herbertsmithite)
- Triangular Spin S=1 QSL in "A-BaNiSbO"
- Conclusion

Liquids?

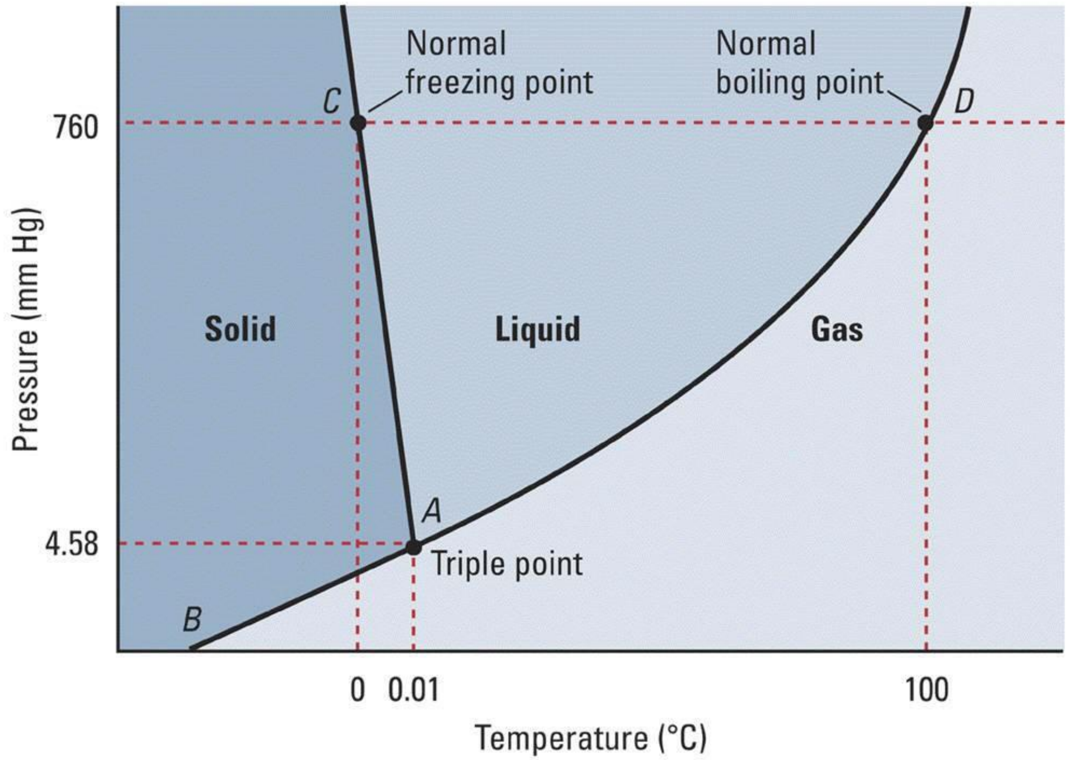
or solids ?

Solids

- Described by local order parameters and symmetry breaking
- Rich structure; classification: 230 space groups in 3D; 17 in 2D

Liquids (water)

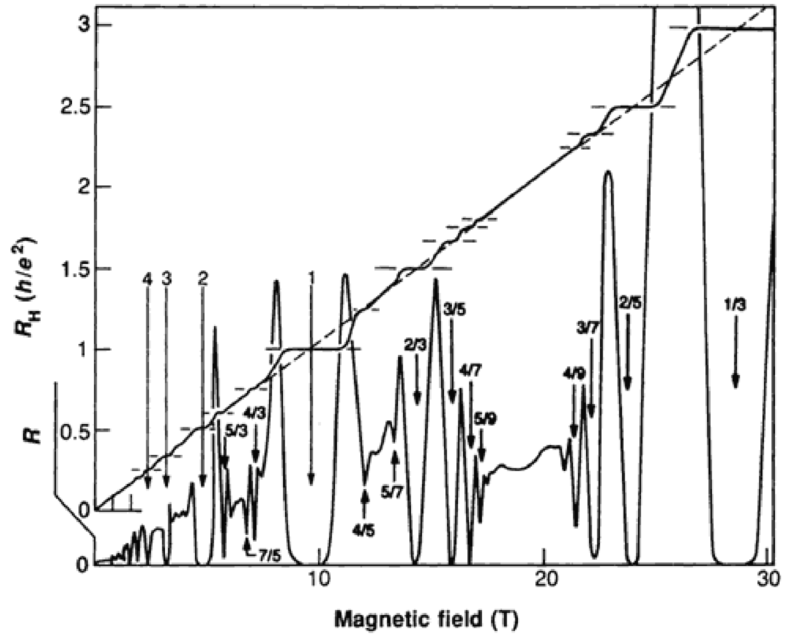
- Unbroken space symmetries
- Characterized by short distance correlations, dynamical properties
- Much more subtle to classify than solids
- Crossover between "phases"



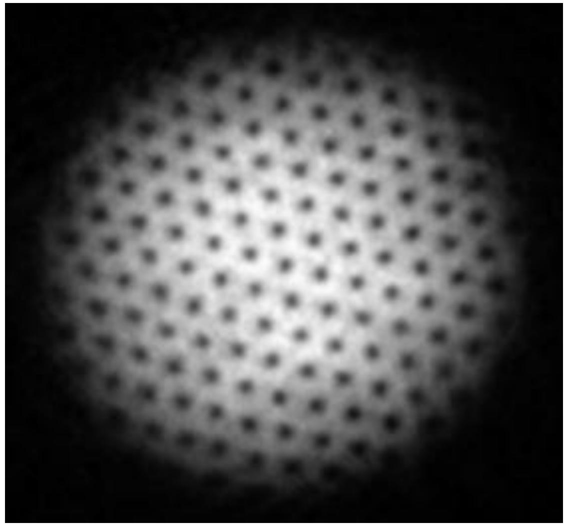
Quantum liquids

- Prominent examples:

Quantum Hall effect



Superfluidity



Here: Liquids beyond Landau / topological order

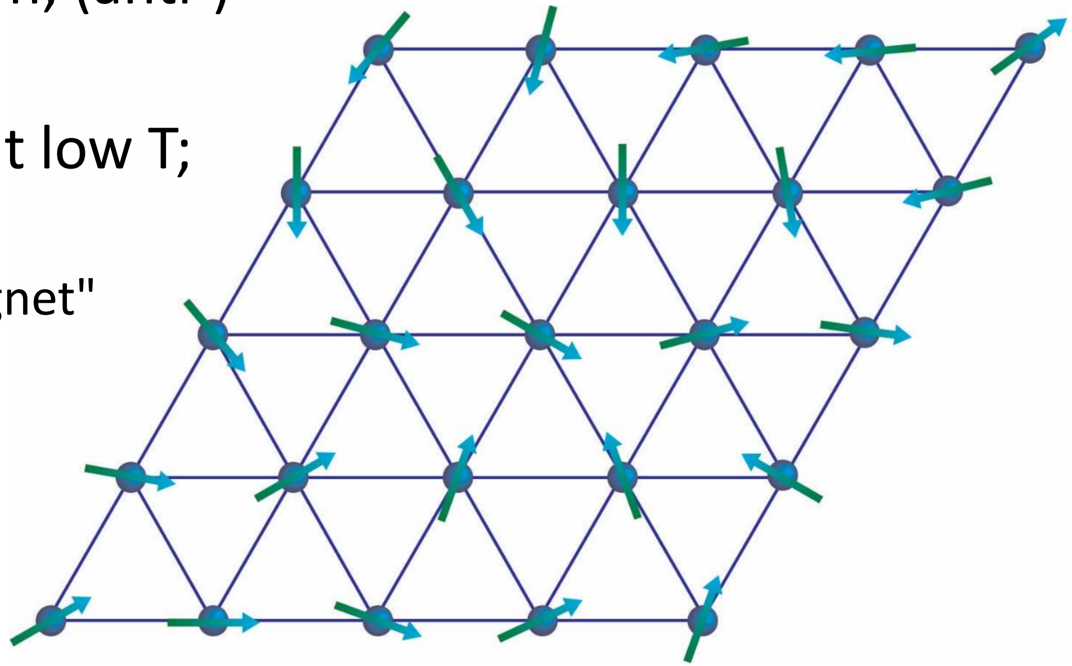
- No breaking of (continuous, global) symmetry as T -> 0
- Absence of local order parameter

Similar phases in magnetic systems

Phases:

- **Spin gas**: Independent spins point in random directions; high-T paramagnetic phase.
- **Spin solid**: Freezing of spins to a regular pattern; (anti-) ferromagnetic phase.
- **Spin liquid?** Interacting and fluctuating spins at low T; no ordering and no symmetry breaking

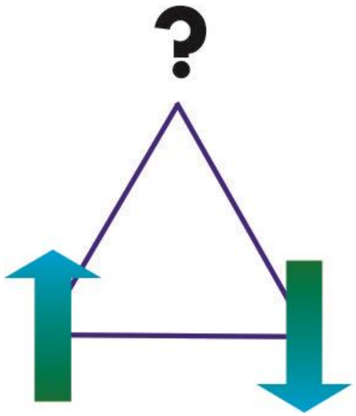
"Cooperative paramagnet"



Geometric frustration

$$H = J S_i^z S_j^z \ , \ J > 0$$

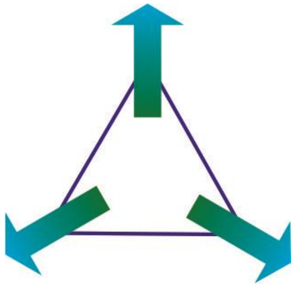
- Two Ising spins:  = Happy
- Three Ising spins with antiferromagnetic interaction:



→Degeneracy of classical ground state.

Triangular Ising lattice [Wannier 1950]

- Classical Heisenberg spins:

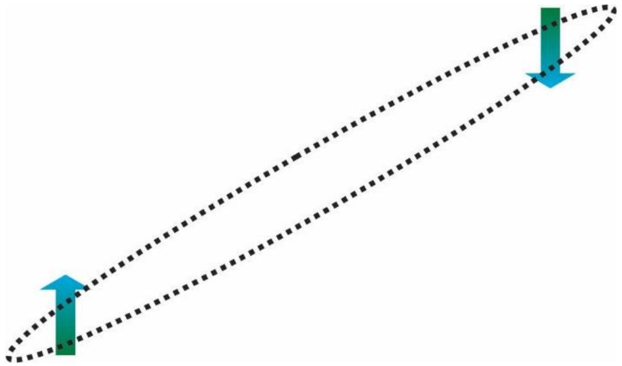


→ Quantum spins?
→ More involved interactions?

Quantum spin liquids: general properties

- Absence of global spin rotation breaking at T=0 (negative definition)
- Certainly happens in 1D spin models

  What about 2D or 3D? Néel order or disordered GS?
- Long-range entanglement [Levin, Wen; Kitaev, Preskill 06]; State that cannot be approximated by any finite-region product wave function.

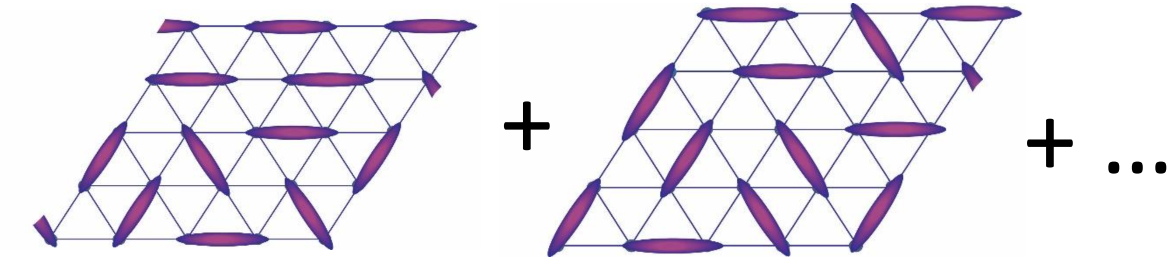


Resonating valence bonds (RVB)

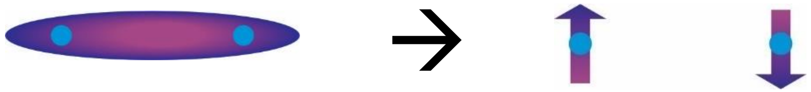
- Valence bond singlet: $|\mathrm{VB}\rangle = \frac{1}{\sqrt{2}}[|\uparrow_1\downarrow_2\rangle - |\downarrow_1\uparrow_2\rangle]$ =  $\langle \boldsymbol{S}_1 \cdot \boldsymbol{S}_2 \rangle = -3/4$

  Néel:  $\langle \boldsymbol{S}_1 \cdot \boldsymbol{S}_2 \rangle = -1/4$

- Anderson 1973: Quantum superposition of valence bonds may beat Néel order

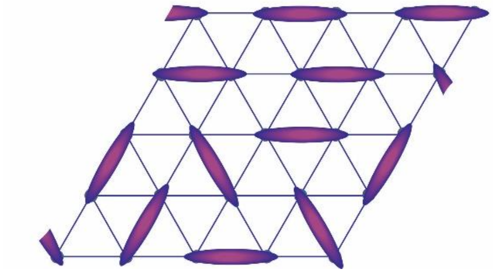


- Anderson 1987: **High-temperature superconductivity** can naturally emerge from RVB states (under doping) [Lee, Nagaosa, Wen, RMP 78, 17 (2006)]
- Spinon excitation (spin-1/2); broken valence bond ($\Delta E = J/2$)



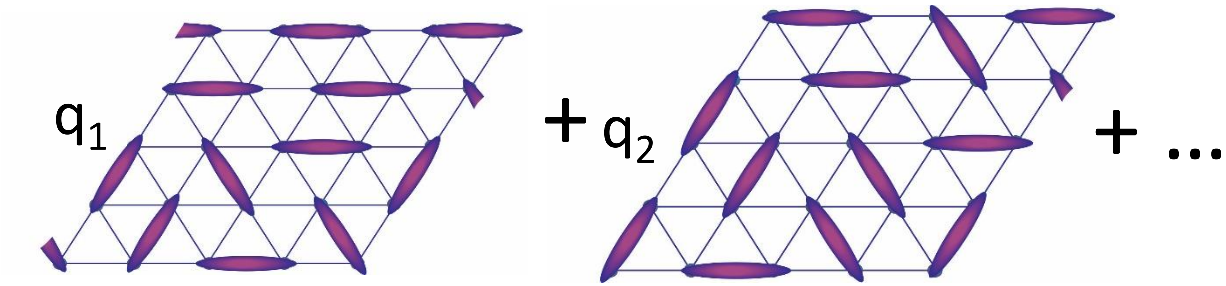
Types of valence bond states

- Valence bond solid



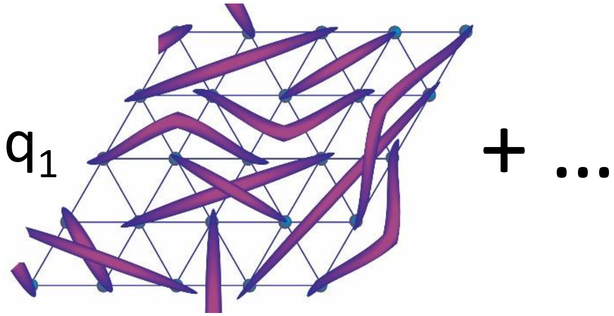
→ Lattice symmetry breaking
→ Product state of valence bonds
→ No long-range entanglement

- Liquid of "short" valence bonds



→ No lattice symmetry breaking
→ Long-range entangled
→ Gapped (S=1/2) spinon excitation
→ Spinless vortex excitation (visons)
→ Topological order; group cohomology classification [Chen, Gu, Wen 12]

- Liquid of "long" valence bonds



→ No lattice symmetry breaking
→ Long-range entangled
→ Low-energy spinon excitations
→ Algebraic/critical correlations (ASL)
→ Refined classification more subtle

Gutzwiller construction of RVB states

- *A priori* it is difficult to make the RVB picture quantitative
- Take simple long-range entangled state – the Fermi gas: $|FS\rangle = \prod_{\epsilon_k<\mu} c^\dagger_{k\downarrow} c^\dagger_{k\uparrow}|0\rangle$

$$P_G\,|FS\rangle = q_1|\uparrow,\downarrow,\downarrow,\uparrow,\ldots\rangle + q_2|\downarrow,0,\uparrow\!\!\!\!/,\downarrow,\ldots\rangle + q_3|\downarrow,\uparrow,\uparrow\!\downarrow\!\!\!\!/,\downarrow,\ldots\rangle + \ldots$$

- Projection (P_G) can efficiently be done (for Fermions) using Monte Carlo tec.
- Liquid character not destroyed by projection ?

  [Grover, Vishwanath 11; Tao Li, EPL 13]
- Auxiliary degree of freedom [slave particles, partons (spinons)] $c_{j\sigma}$
- Emergent local (gauge) symmetry

→ parton construction; PSG

Projective symmetry group

- How to classify RVB spin states beyond symmetry breaking?
  - Broken symmetry: Bragg-peaks; Landau theory
- X.-G. Wen: Parton classification [PRB 65, 165113 (2002)]
- Parton classification of *chiral* spin liquid states [**SB** et al., PRB 93, 094437 (2016)]

Parton construction & classification

Spin-1/2 Heisenberg model: $H = \sum_{ij} J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$

(a) Fractionalize spin into spinons f_α , carrying ΔS = 1/2 (magnons ΔS=1)
(f_α : "Abrikosov fermion" creation operator)

spinon doublet: $\boldsymbol{f} = (f_\uparrow, f_\downarrow)^T$ $\quad 2S_a = \boldsymbol{f}^\dagger \sigma_a \boldsymbol{f}$ $\quad \boldsymbol{S}^2 = \frac{3}{4} n[2-n]$

enlarged local Hilbert space:

$\{|\uparrow\rangle, |\downarrow\rangle\} \Rightarrow \{|0\rangle, |\uparrow\rangle, |\downarrow\rangle, |\uparrow\downarrow\rangle\}$ constraint/physical subspace: $n = \boldsymbol{f}^\dagger \boldsymbol{f} \equiv 1$

gauge doublet: $\boldsymbol{\psi} = (f_\uparrow, f_\downarrow^\dagger)^T$

gauge transformation: $\boldsymbol{\psi} \mapsto g\boldsymbol{\psi},\ g \in \mathrm{SU}(2)$: leaves spin S_a invariant

Affleck et al, PRB 38, 745 (1988).
Marston et al, PRB 39, 11538 (1989).

Emergent SU(2) symmetry is local:
(gauge sym)

$$\boldsymbol{\psi} = (f_\uparrow, f_\downarrow^\dagger)^T$$

$$\boldsymbol{\psi} \mapsto g\boldsymbol{\psi}, \; g \in \mathrm{SU}(2)$$

Projective symmetry group:
How can actual symmetries be represented in the spinon Hilbert space?

[Wen, PRB 65, 165113 (2002)]

e.g., time-reversal: $\Theta(\boldsymbol{\psi}) = \varepsilon \boldsymbol{\psi}^* \xrightarrow{g_\Theta = \varepsilon^T} \boldsymbol{\psi}^*$ $\quad \varepsilon = i\sigma_2$

Algebraic relations among symmetries must be *respected* by the representation (up to gauge transformations) !

Parton Ansatz

$$H = \sum_{ij} J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j \qquad 2S_a = \boldsymbol{f}^\dagger \sigma_a \boldsymbol{f}$$

+ Hubbard-Stratonovich
or MF decoupling

⇨ (b) Quadratic spinon Hamiltonian (= singlet "ansatz")

$$H_0 = \sum_{ij} \xi_{ij} f^\dagger_{i\alpha} f_{j\alpha} + \Delta_{ij} f_{i\uparrow} f_{j\downarrow} + \text{h.c.} = \sum_{ij} \boldsymbol{\psi}^\dagger_i u_{ij} \boldsymbol{\psi}_j + \text{h.c.} \qquad \boldsymbol{\psi} = (f_\uparrow, f^\dagger_\downarrow)^T$$

Ansatz: $u = \{u_{ij}\}$

$$u_{ij} = \begin{pmatrix} \xi_{ij} & \Delta_{ij} \\ \Delta^*_{ij} & -\xi^*_{ij} \end{pmatrix}$$

Interpretations/uses of $H_0(u)$:

(i) Low-energy effective theory;
Invariant gauge group (IGG_u): U(1) [$f_j \mapsto e^{i\varphi} f_j$] or $\mathbb{Z}_2$ [$f_j \mapsto -f_j$]

(ii) Self-consistent saddle point solutions for H

(iii) Tool for constructing spin w.f. by Gutzwiller projection : $|\psi\rangle = \prod_j n_j[2-n_j]|\psi_0\{u_{ij}\}\rangle$

Variational Monte Carlo (VMC) method

Projective symmetry group (PSG)

1. Algebraic PSG: Representation classes of the symmetry group SG in the gauge group $\mathcal{G}=\{g\},\ g=\otimes g_j\ ,\ g_j\ \in\mathrm{SU}(2)$

$$Q\colon \mathrm{SG} \mapsto \mathcal{G}$$
$$x \mapsto g_x$$

Equivalence of reps:

$$Q^1 \sim Q^2 \iff \exists g \in \mathcal{G}\ \text{ s.t. }\ Q^1 = gQ^2g^\dagger$$

Algebraic relations in SG respected *up to the IGG*, e.g.: reflection $\sigma^2 = 1 \implies g_\sigma(\boldsymbol{r})g_\sigma(\sigma\boldsymbol{r}) \in \mathrm{IGG}\ \{\pm 1\}$

IGG: Invariant Gauge Group (subgrp of $\mathcal{G}$)
(here: $\mathbb{Z}_2$ classification)

2. Invariant PSG: Ansatz u respecting SG for each PSG class

action of symmetry x on Ansatz: $Q_x(u_{ij}) = (-)^{\tau_x} g_x(i)u_{x^{-1}(ij)}[g_x(j)]^\dagger$

$$Q_x(u) = u \qquad \text{for all } x \text{ in SG}$$

PSG: Kagome

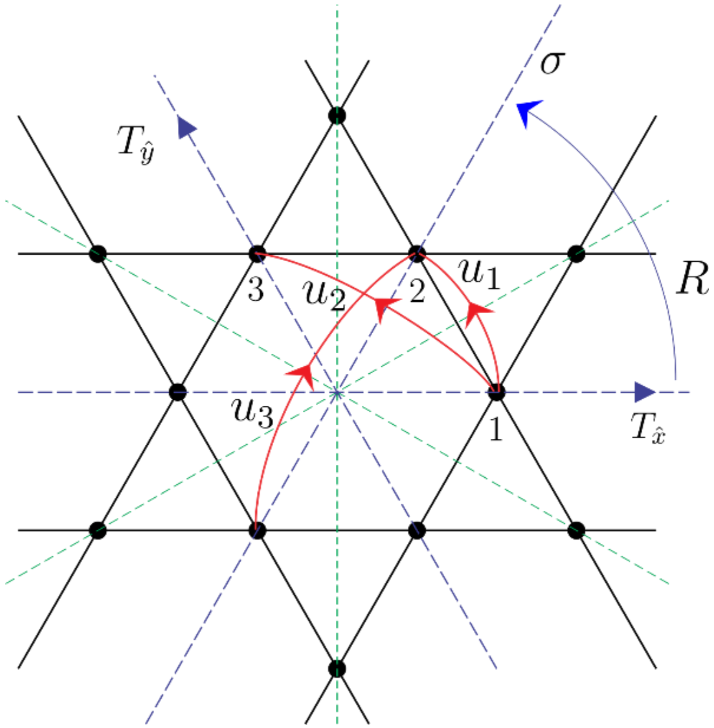
SB et al., Phys. Rev. B 92, 060407(R) (2015)

Symmetries: $\mathrm{SG}_{\tau_\sigma,\tau_R} = \{T_{\hat{x}}, T_{\hat{y}}, \sigma\Theta^{\tau_\sigma}, R\Theta^{\tau_R}\}$

$\tau_R = 0, \tau_\sigma = 0$: Symmetric QSL

$\tau_R = 0, \tau_\sigma = 1$: "Kalmeyer-Laughlin" CSL

$\tau_R = 1$: Staggered-flux CSL



$g_x = \mathbb{1}_2$

$g_y = (\epsilon_2)^x \mathbb{1}_2$

$g_\sigma(x,y) = (\epsilon_2)^{xy} g_\sigma$

$g_R(x,y) = (\epsilon_2)^{xy+y(y+1)/2} g_R$

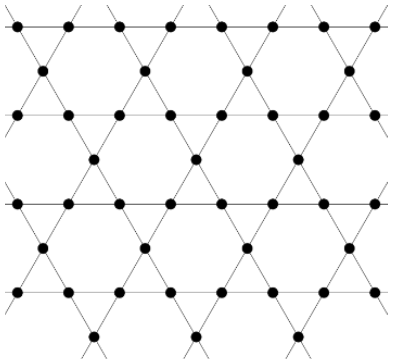
$\epsilon_2 = \pm 1$

no.	g_σ	g_R	ϵ_σ	$\epsilon_{R\sigma}$	ϵ_R	sym
1	$\mathbb{1}_2$	$\mathbb{1}_2$	+	+	+	SU(2)
2	$i\sigma_3$	$\mathbb{1}_2$	−	−	+	U(1)
3	$\mathbb{1}_2$	$i\sigma_3$	+	−	−	U(1)
4	$i\sigma_3$	$i\sigma_3$	−	+	−	U(1)
5	$i\sigma_2$	$i\sigma_3$	−	−	−	$\mathbb{Z}_2$

⇨ **10 PSG classes** on Kagome

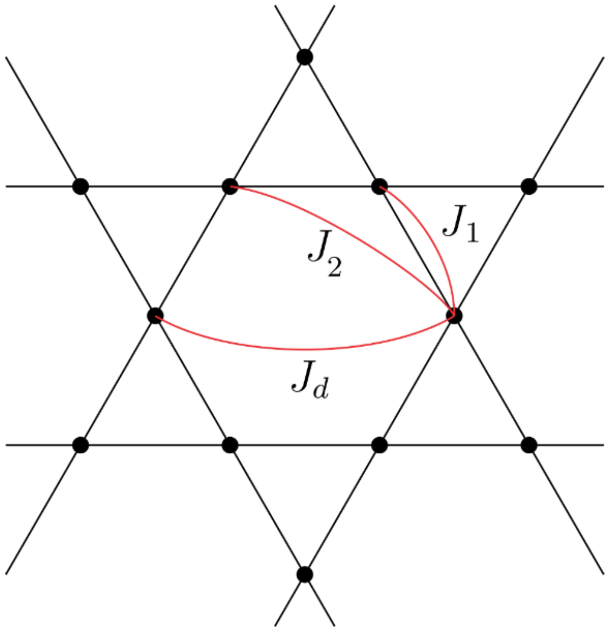
Physical realizations of QSLs

- In recent years, a number of experimental QSL candidate materials have been discovered:
  - Kagome lattice [$ZnCu_3(OH)_6Cl_2$] spin-1/2
    - Herbertsmithite [Nocera 05; Y. Lee 12]
    - Kapellasite [Wills 08; Fak 12]
    - Vanadite [Bert 13], ...
  - Triangular lattice spin-1/2 : k-ET, dMIT (organics) [Saito 03; Kato 07, ...]; $YbMgGaO_4$ [Li 15, ...]
  - Triangular lattice spin-1 : $Ba_3NiSb_2O_9$ [Balicas 11; Quilliam; Darie; Fak 17]
  - 3D candidates: $Yb_2Ti_2O_7$, $Na_4Ir_3O_8$, … (pyrochlore, hyperkagome) [Mendels 15, ...]



Kapellasite [$ZnCu_3(OH)_6Cl_2$]

- No ordering down to mK, gapless continuum of spin excitations
- Weak ferro Curie-Weiss temp Θ_{CW} ~ 9 K
- Farther-neighbor Heisenberg exchange: J_1 ~ -12 K, J_2 ~ -4 K, J_d ~ 16 K
- Powder samples



R. H. Colman et al, C.M. 20, 6897 (2008); 22, 5774 (2010).

O. Janson at al, PRL 101, 106403 (2008).

H. O. Jeschke et al, PRB 88, 075106 (2013).

E. Kermarrec et al, PRB 90, 205103 (2014).

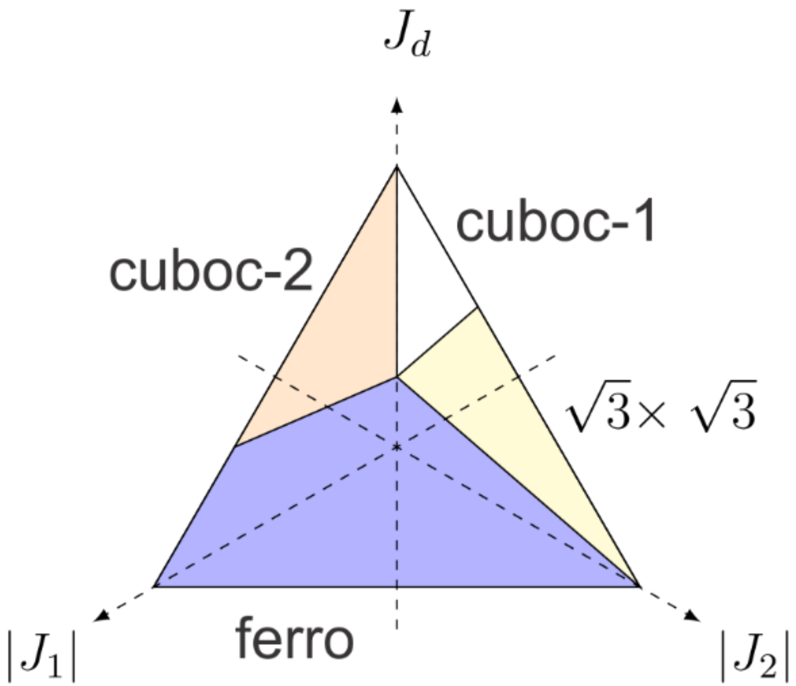
B. Fåk et al, PRL 109, 037208 (2012).

B. Bernu et al, PRB 87, 155107 (2013).

$$H = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_d \sum_{\langle i,j \rangle_d} \mathbf{S}_i \cdot \mathbf{S}_j$$

→ Experimental evidence for gapless quantum spin liquid ground state

Phase diagram of *classical* J_1-J_2-J_d kagome Heisenberg model



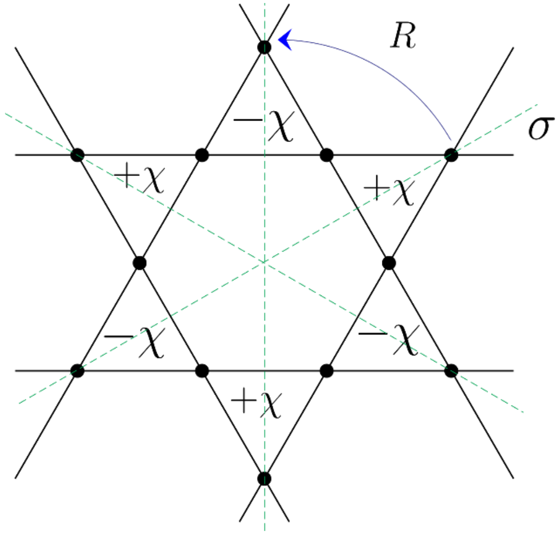
$$|J_1|+|J_2|+J_d = 1$$

$$J_1 < 0,\ J_2 < 0,\ J_d > 0$$

Messio, Lhuillier, Misguich, PRB 83, 184401 (2011).

cuboc-1,-2: non-planar spin order with $\chi = \boldsymbol{S}_1\cdot(\boldsymbol{S}_2 \times \boldsymbol{S}_3) \neq 0$
12 site unit cell

Spontaneous breaking of time-reversal, (up to) lattice reflection and rotation



What happens in the case of quantum spin S=1/2?

Is the elusive chiral spin liquid realized in kapellasite?

Kalmeyer and Laughlin, PRL 59, 2095 (1987).

Wen, Wilczek, Zee, PRB 39, 11413 (1989).

Yang, Warman, Girvin, PRL 70, 2641 (1993).

Quantum phase diagram

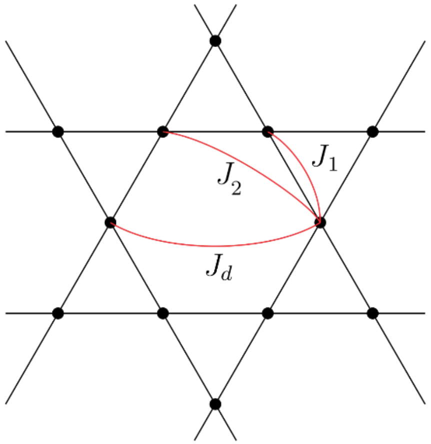
SB et al, PRB 92, 060407 (2015)

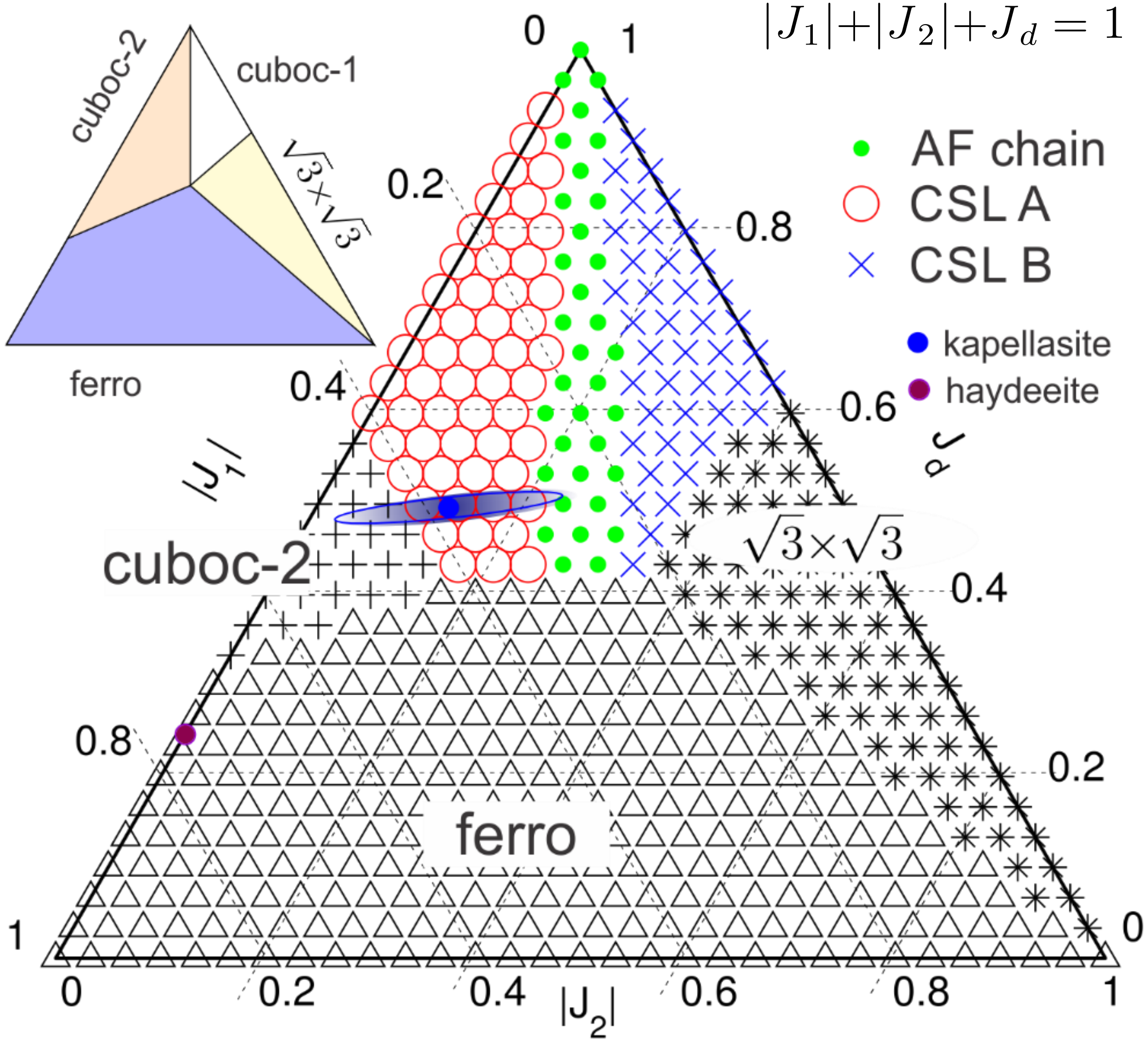
Energy comparison of projected U(1) CSL w.f.

$$|\psi\rangle = \prod_j n_j[2-n_j]|\psi_0\rangle$$

with correlated Néel states

$$|\mathrm{Neel}\rangle = \exp\{\sum_{ij} \mathcal{J}_{ij} S_i^z S_j^z\} \prod_k |S_k\rangle$$





Spin model

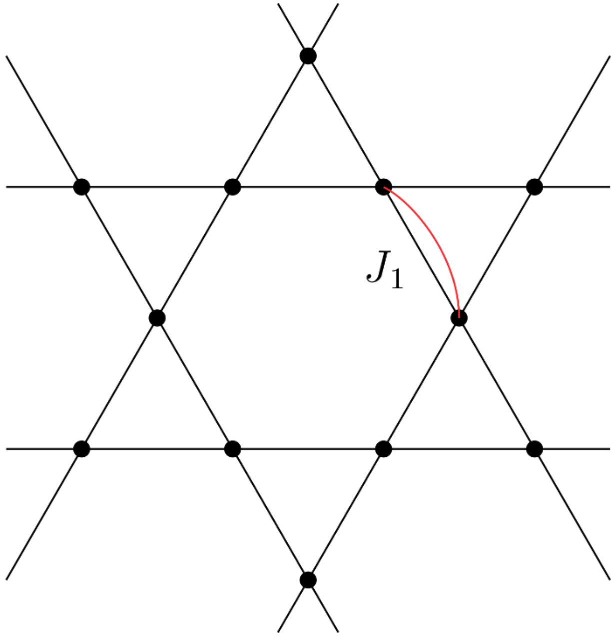
$$H = J_1 \sum_{\langle i,j\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle i,j\rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_d \sum_{\langle i,j\rangle_d} \mathbf{S}_i \cdot \mathbf{S}_j$$

with $J_1 < 0,\ J_2 < 0,\ J_d > 0$

see also Iqbal, PRB 92, 220404 (2015);
Gong, PRB 94, 035154 (2016) ?
[work in progress w. R. Pereira @ IIP]

Herbertsmithite [$ZnCu_3(OH)_6Cl_2$]

- No ordering down to mK, gapless/gapped (?) spin excitations
- Strong AF Curie-Weiss, dominant $J_1 = J \sim 200$ K ; $J_2 = J_d = 0$
- Single crystals [Young Lee (MIT), now also France (Ph. Mendels)]
- Perturbations; e.g. Dzialoshinskii-Moriya: $D_z \sim J/12$



T.-H. Han et al, Nature 492, 406 (2012).

A. Zorko et al, PRL 118, 017202 (2017).

A. Zorko et al, PRL 101, 026405 (2008).

I. Dzyaloshinsky, J. Phys. Chem. Solids 4, 241 (1958).

T. Moriya, PRL 4, 228 (1960).

L. Shekhtman et al, PRL 69, 836 (1992).

Spin-orbit coupling/Dzyaloshinskii-Moriya (DM) Interaction:

$$H = J \sum_{\langle i,j \rangle} h_{ij}$$

$$h_{ij} = \boldsymbol{S}_i' \cdot \boldsymbol{S}_j'$$

DM-vector: $\boldsymbol{D}_{ij} = J\,\theta_{ij}\,\hat{\boldsymbol{d}}_{ij}$

$$\boldsymbol{S}_i' = \boldsymbol{S}_i \text{ rotated by } -\theta_{ij} \text{ around } \hat{\boldsymbol{d}}_{ij}$$

$$\boldsymbol{S}_j' = \boldsymbol{S}_j \text{ rotated by } +\theta_{ij} \text{ around } \hat{\boldsymbol{d}}_{ij}$$

Schwinger-boson mean-field theory (for DM)

L. Messio, **SB**, et al, arXiv:1701.01253

(a) Fractionalize spin into bosonic ΔS = 1/2 spinons b_α

(b_α: “Schwinger boson” creation operator)

$$2S_a = \boldsymbol{b}^\dagger \sigma_a \boldsymbol{b} \qquad \boldsymbol{b} = (b_\uparrow, b_\downarrow)^T \qquad \boldsymbol{S}^2 = \frac{1}{4} n(2+n)$$

enlarged local Hilbert space: $\{|\uparrow\rangle,\ |\downarrow\rangle\} \Rightarrow \{|0\rangle, |1,0\rangle, |0,1\rangle, |1,1\rangle, \ldots\}$

constraint/physical subspace:

$$n = \boldsymbol{b}^\dagger \boldsymbol{b} \equiv 2S$$

emergent gauge symmetry: $\boldsymbol{b} \mapsto g\boldsymbol{b}$, $\quad g \in \mathrm{U}(1)$

Quadratic spinon theories:

$$h_{ij} = :B^\dagger_{ij} B_{ij}: \ - A^\dagger_{ij} A_{ij}$$

$$A_{ij} = e^{-i\theta_{ij}} b_{i\uparrow} b_{j\downarrow} - e^{i\theta_{ij}} b_{i\downarrow} b_{j\uparrow}$$

$$B_{ij} = e^{i\theta_{ij}} b^\dagger_{i\uparrow} b_{j\uparrow} - e^{-i\theta_{ij}} b^\dagger_{i\downarrow} b_{j\downarrow}$$

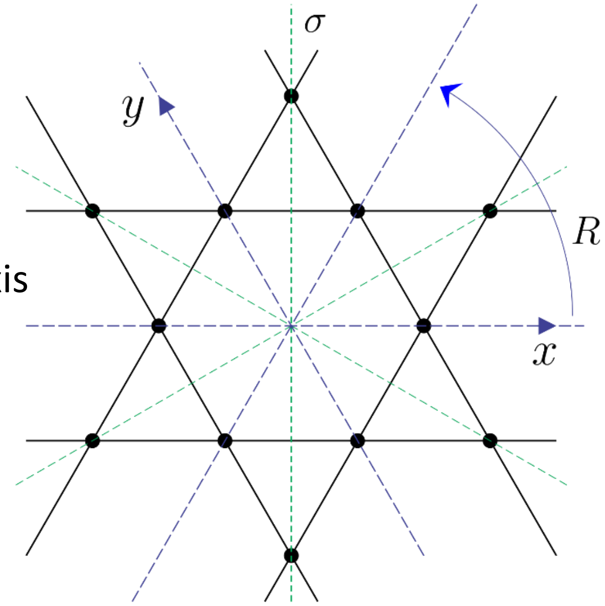
$$\Rightarrow \quad h^{\mathrm{MF}}_{ij} = \mathcal{B}^*_{ij} B_{ij} - \mathcal{A}^*_{ij} A_{ij} + \mathrm{h.c.}$$

$$\mathrm{SG} = \{T_{\hat{x}}, T_{\hat{y}}, R\Theta^{\tau_R}, \sigma S_{\pi x} \Theta^{\tau_\sigma}\}$$

$S_{\pi x}$: spin-rotation π around x-axis

$\tau_R = 1$ or $\tau_\sigma = 1$

$\Rightarrow$ spont. breaking of pt group and TR: CSL



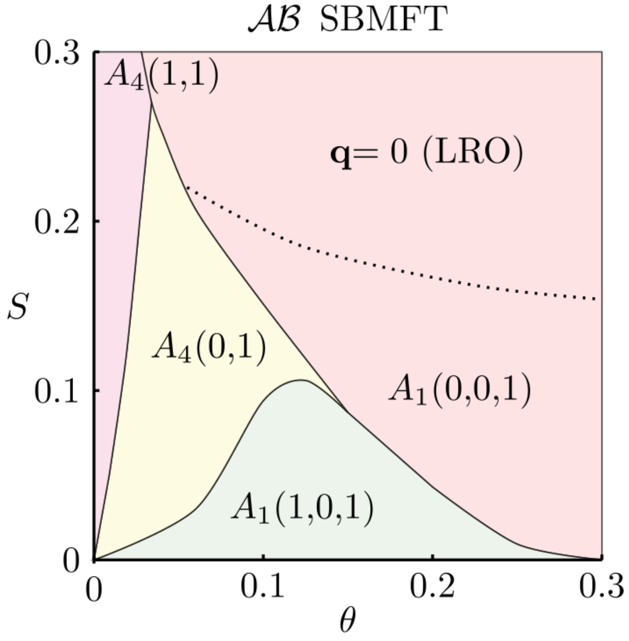
(b) PSG classification of MF states:
L. Messio et al, PRB 83, 184402 (2011).

Self-consistent solutions

$$\mathcal{A}_{ij} = \langle A_{ij} \rangle \qquad A_{ij} = e^{-i\theta_{ij}} b_{i\uparrow} b_{j\downarrow} - e^{i\theta_{ij}} b_{i\downarrow} b_{j\uparrow}$$

$$\mathcal{B}_{ij} = \langle B_{ij} \rangle \qquad B_{ij} = e^{i\theta_{ij}} b^\dagger_{i\uparrow} b_{j\uparrow} - e^{-i\theta_{ij}} b^\dagger_{i\downarrow} b_{j\downarrow}$$

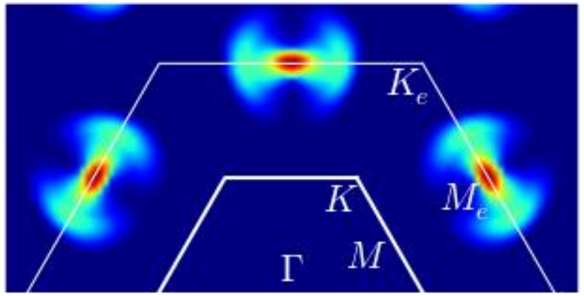
$$2S = \sum_j \langle n_j \rangle$$



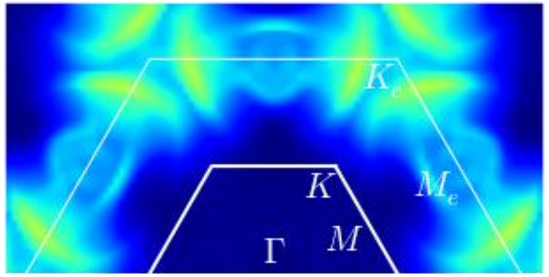
L. Messio, **SB**, C. Lhuillier, B. Bernu, arXiv:1701.01253

A4(1,1) phase: L. Messio et al, PRL 108, 207204 (2012)

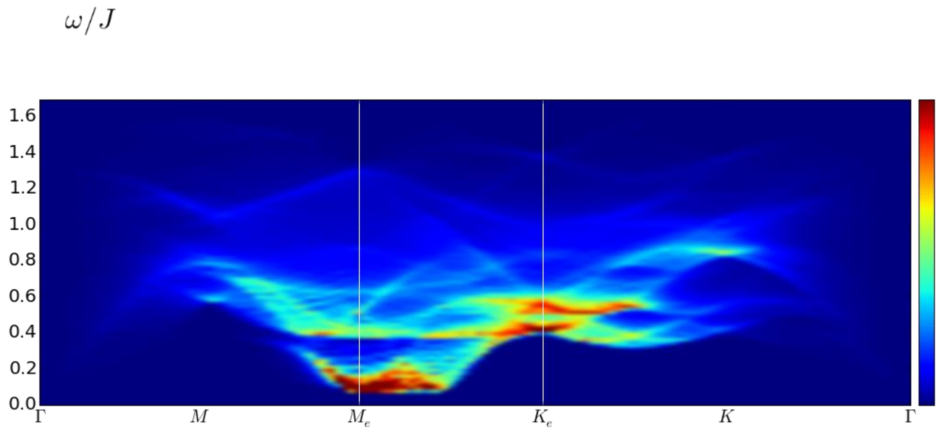
$S(\boldsymbol{q},\omega)$ in new CSL phase $A_4(0,1)$



$\omega = 0-0.15J$



$\omega = 0.3-0.45J$



$Ba_3NiSb_2O_9$

PRL 107,197204 (2011) PHYSICAL REVIEW LETTERS week ending 4 NOVEMBER 2011

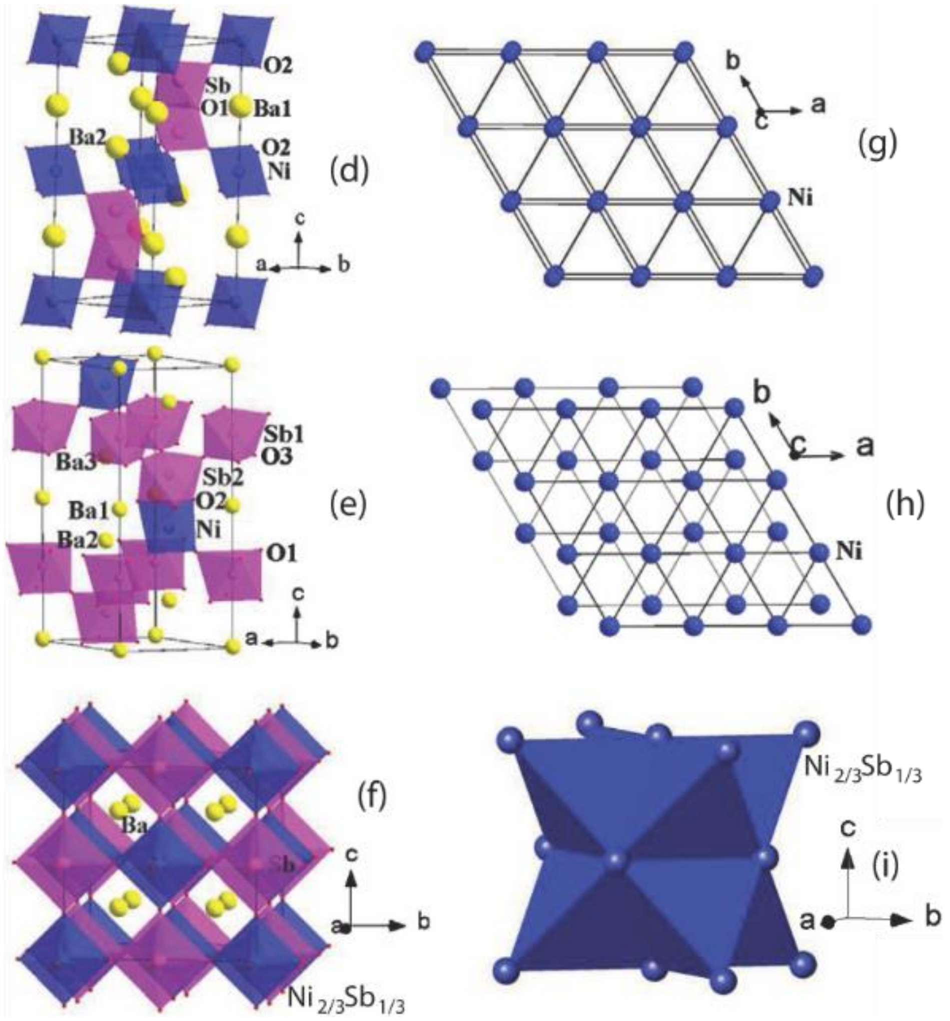
High-Pressure Sequence of $Ba_3NiSb_2O_9$ Structural Phases: New $S = 1$ Quantum Spin Liquids Based on Ni^{2+}

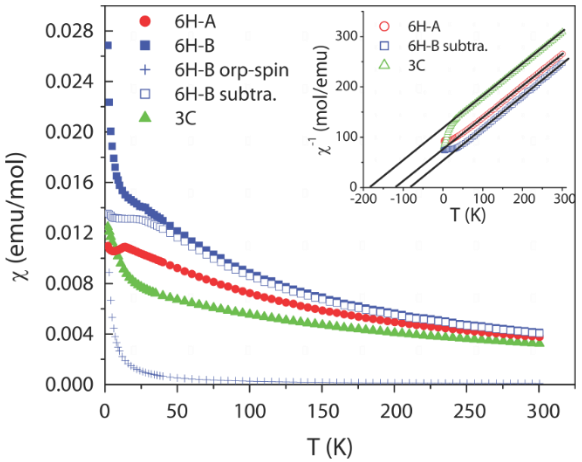
J. G. Cheng,[1] G. Li,[2] L. Balicas,[2] J. S. Zhou,[1] J. B. Goodenough,[1] Cenke Xu,[3] and H. D. Zhou[2,*]

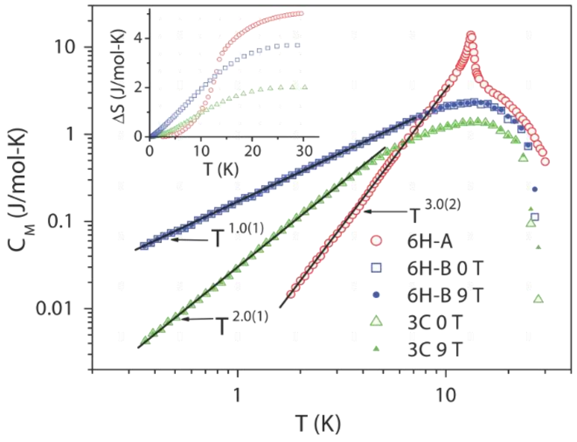
6H-A

6H-B

3C







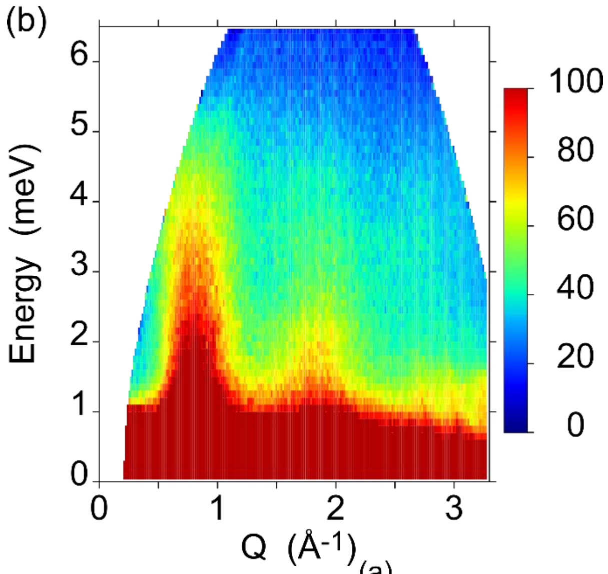
Theories for intriguing 6H-B phase:

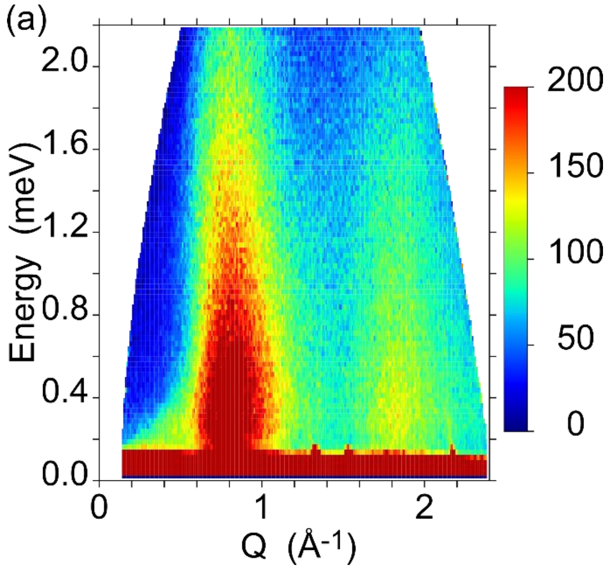
Serbyn et al, PRB 84, 180403 (2011)
SB et al, PRB 86, 224409 (2012)
Xu et al, PRL 108, 087204 (2012)
Chen et al, PRL 109, 016402 (2012)
Hwang et al, PRB 87, 235103 (2013)

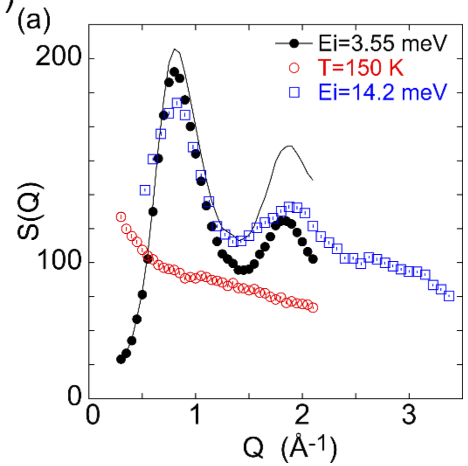
B-$Ba_3NiSb_2O_9$ (INS)

B. Fåk, **SB**, et al., Phys. Rev. B 95, 060402(R) (2017).

Inelastic neutron scattering on 6H-B phase (powder):





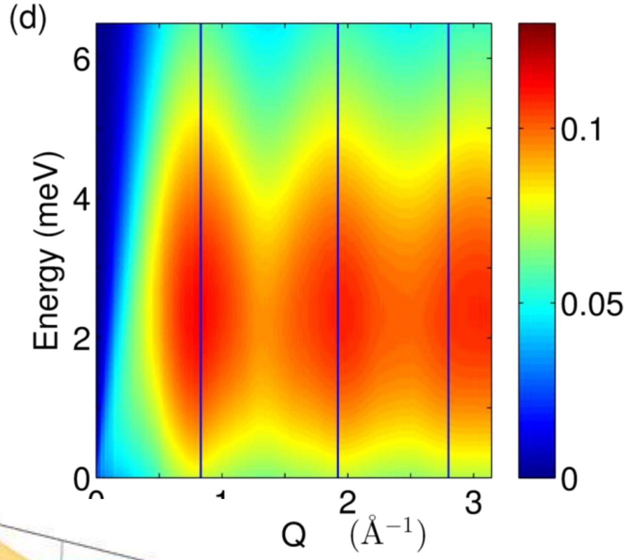


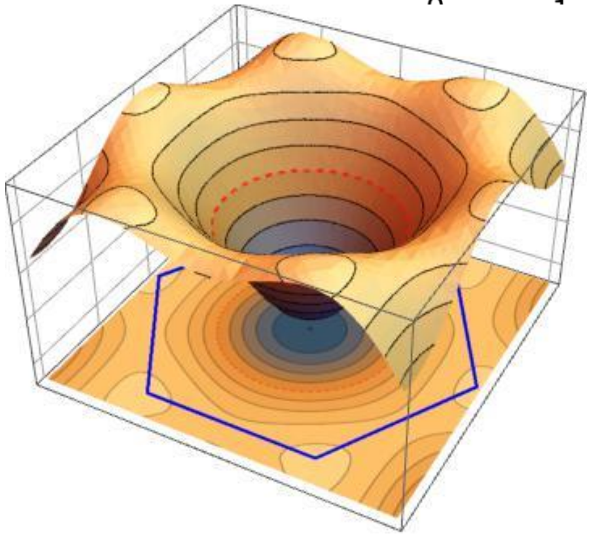
NMR: Quilliam et al, PRB 93, 214432 (2016).

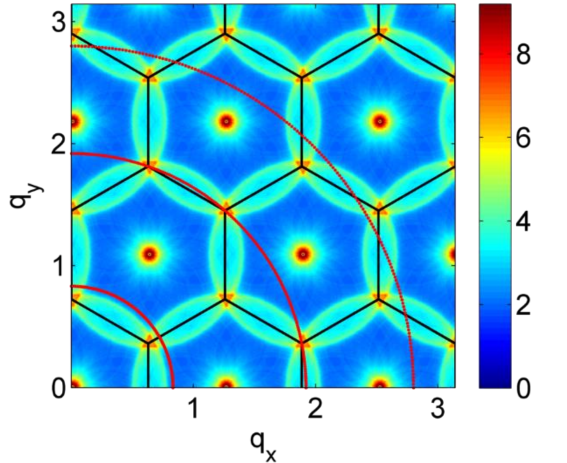
spin fractionalization:

$$\boldsymbol{S} = i\boldsymbol{f}^\dagger \wedge \boldsymbol{f} = i(\varepsilon^{abc} f_b^\dagger f_c), \ a = x, y, z$$

$S(Q,\omega)$ for Fermi sea of spinons at 1/3 filling







Conclusion & outlook

- PSG classification for QSLs
- Exhaustive list of fermonic parton CSLs (kagome, triangular)
- Quasi-1D phase in a S=1/2 kagome system (kapellasite)
- New CSL in kagome Heisenbg. model with DM term (herbertsmithite)
- Evidence for spinon Fermi surface in S=1 triangular QSL (B-BaNiSbO)
- Outlook:
  - Fermionic approach to Dzyaloshinskii-Moriya
  - Ring exchange, honeycomb
  - 3D lattices (hyperkagome, ...)

Thank you!

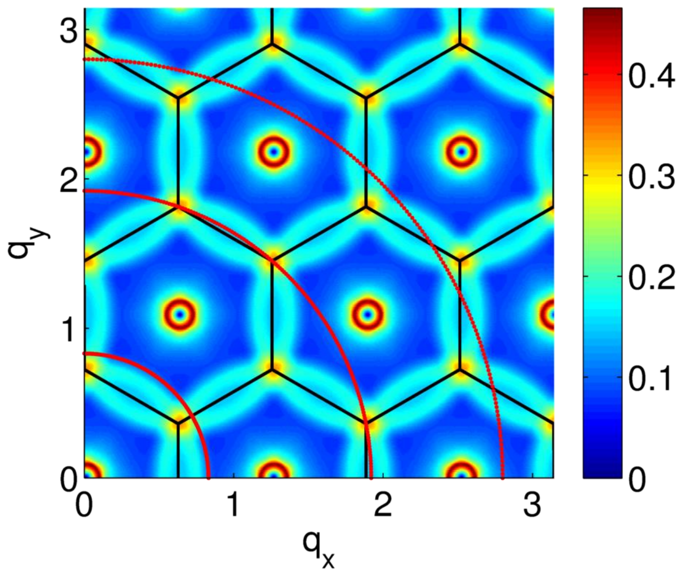
Characterization: Spinon spectrum; Spin structure factor

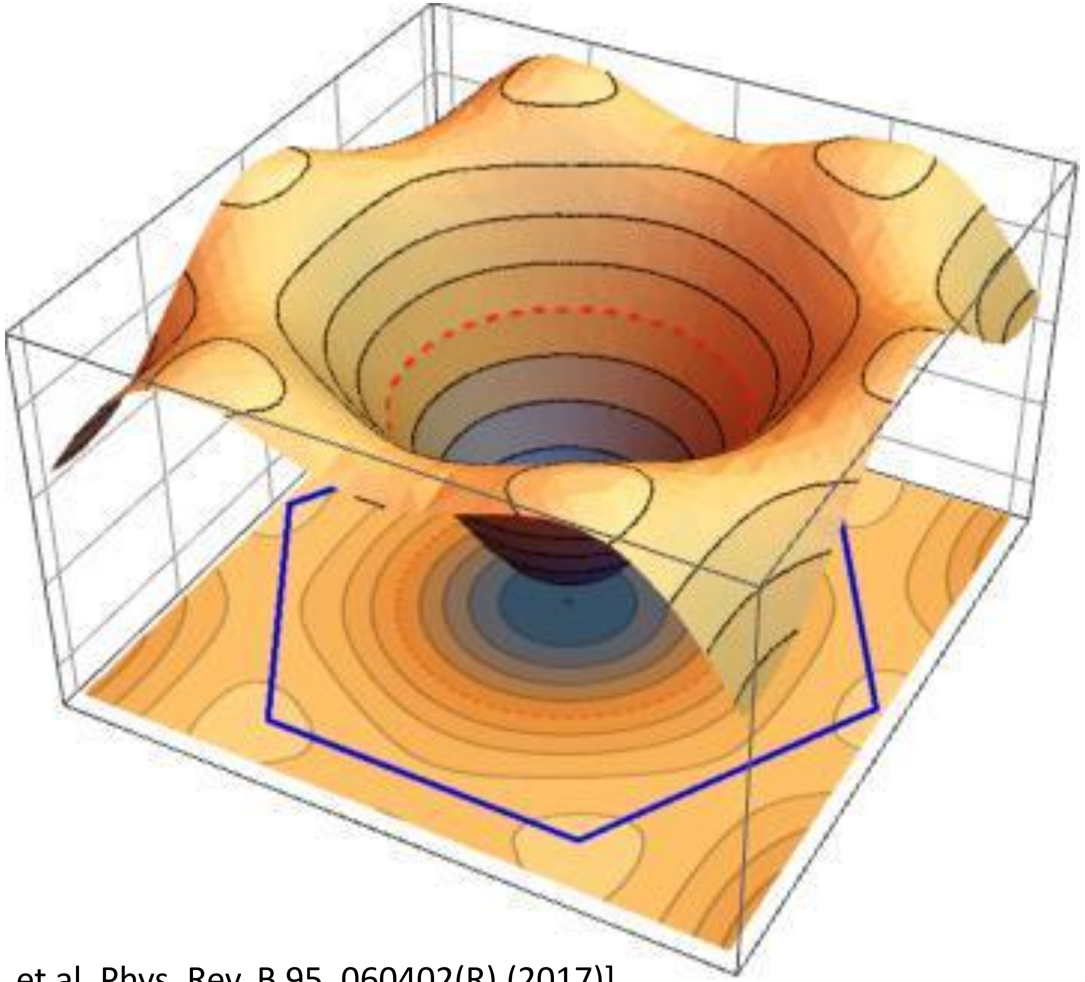
E.g. spinon Fermi surface
(note: This is a Mott insulator!)

$$S \sim \langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j \rangle$$

$$S(\boldsymbol{q}) \sim |\boldsymbol{q}|^{\alpha}\ , \text{ as } |\boldsymbol{q}| \to 0$$ ("algebraic SL")

$$S(\boldsymbol{q}, \omega \sim 0) \text{ features at } \boldsymbol{q} \sim 2\boldsymbol{k}_F$$





[B. Fåk, **SB**, et al, Phys. Rev. B 95, 060402(R) (2017)]