Variational perspective on quantum spin liquids in triangular-lattice Heisenberg models for spin $1/2$ and spin 1

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Abstract

We propose novel gapless quantum spin liquid (QSL) states that may explain the phenomenology of recently discovered experimental spin liquid candidates in spin $S = 1/2$ and $S = 1$ layered triangular lattice compounds. These states have a number of theoretically very interesting and appealing properties. We propose microscopic Heisenberg models with ring-exchange terms where these new phases can be realized as ground states. Using variational Monte Carlo calculations, we compare the energetics of a wide range of correlated spin wave functions. We find that our exotic spin states are indeed stabilized in some parameter range. For the organic compounds ($S = 1/2$), the parameters are realistic, and our theoretical scenario therefore presents a serious possibility.

Motivation

Recent experimental discoveries of the quantum spin liquid candidate $Ba_3NiSb_2O_9$ [3], and the organic compounds κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ [4] and EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ [5]. Key facts are

- Layered 2d structure of isotropic triangular geometry
- No ordering transition observed down to 0.1 K and below
- Spin carried by Nickel is effective $S = 1$, and by Copper is $S = 1/2$
- AF Mott insulators: $J \simeq 250$ K (organics), $\Theta_{\text{CW}} \simeq -80$ K (Ni)
- Finite spin susceptibility χ_0, and linear $C_V = \gamma T$ at low T
- Weak site disorder
- Powder samples only, no large crystals so far

⇒ Quite robust experimental indications that two-dimensional gapless quantum spin liquid states are realized in these materials at low temperature.

Spin fractionalization

We use a "parton construction" [6] with fermionic spinons to fractionalize the quantum spin operators. For spin 1/2 it is

$$S_a = \frac{1}{2}\boldsymbol{f}^\dagger \sigma_a \boldsymbol{f}\,, \tag{1}$$

with $\boldsymbol{f} = (f_\uparrow, f_\downarrow)^T$. The emergent gauge structure is SU(2).

For spin $S = 1$, at least *three* spinons are needed (3d rep); we use

$$\boldsymbol{S} = -i\boldsymbol{f}^\dagger \wedge \boldsymbol{f}\,, \tag{2}$$

with $\boldsymbol{f} = (f_x, f_y, f_z)^T$. $f_z = if_0$ is a "nematic" quasiparticle, while $f_{\uparrow,\downarrow} = (f_x \mp if_y)/\sqrt{2}$ carry spin. Gauge structure is U(1)$\rtimes\mathbb{Z}_2$.

Possible QSL scenarios at the mean-field level are

1. The "mother" of gapless QSLs, the U(1) state with spinon Fermi sea: $H_0 = \sum_{ij\alpha} t_{ij} f^\dagger_{i\alpha} f_{j\alpha}$
   ⇒ emergent U(1) gauge Boson (photon)
2. Pairing instability, Anderson-Higgs mechanism; $\mathbb{Z}_2$ spin liquids: $H_0 = \sum_{ij}\sum_\alpha t_{ij} f^\dagger_{i\alpha} f_{j\alpha} + \sum_{\alpha\beta}\Delta^{\alpha\beta}_{ij} f_{i\alpha} f_{j\beta} + \text{h.c.}$
   ⇒ gauge Boson is gapped and has no effect at low energy.
3. Ordering instabilities (SDW) of the U(1) QSL, spinon confinement: $H_0 = \sum_{ij\alpha} t_{ij} f^\dagger_{i\alpha} f_{j\alpha} - h\sum_{j\alpha\beta} \hat{n}_{j\alpha}\hat{n}^*_{j\beta} f^\dagger_{j\alpha} f_{j\beta}$

Space group, time reversal, and spin rotation symmetries may be broken or unbroken in these states.

⇒ Are there realistic microscopic spin models that exhibit deconfined spinon quasiparticles at low energy?

Construction of microscopic variational spin states:

- Fermionic wave functions [7]

$$|\psi\rangle = P_d|\psi_0(t_{ij}, \Delta_{ij}, \ldots)\rangle\,, \tag{3}$$

where $|\psi_0\rangle$ is the ground state of a quadratic spinon Hamiltonian H_0, and $P_d = \prod_j n_j(n_j - 2)$ the Gutziller projector to the physical spin space; $n = \sum_\alpha f^\dagger_\alpha f_\alpha$.

- Huse-Elser [8] type construction

$$|\psi\rangle = \exp\{\sum_{ij} \mathcal{J}_{ij} S^z_i S^z_j + \mathcal{K}_{ij}(S^z_i S^z_j)^2\}\prod_k |\alpha_k\rangle\,. \tag{4}$$

⇒ Variational Monte Carlo allows evaluation of expectation values in theses highly correlated wave functions to an arbitrary precision on large clusters (here up to 10^3 sites).

Spin 1/2

The ring-exchange model ($P_{ij} = 2\boldsymbol{S}_i\cdot\boldsymbol{S}_j + 1/2$)

$$H = \sum_{\langle i,j\rangle}\boldsymbol{S}_i\cdot\boldsymbol{S}_j + J_2\sum_{\langle\langle i,j\rangle\rangle}\boldsymbol{S}_i\cdot\boldsymbol{S}_j + K\sum_{\langle i,j,k,l\rangle} P_{ijkl} + \text{h.c.} \tag{5}$$

on the triangular lattice for $J_2 = 0$ is known to exhibit 120° AFM order when $K \lesssim 0.1$ [9]. For $K \gtrsim 0.3$, it supports a U(1) QSL, $H_0 = \sum_{ij\sigma} f^\dagger_{i\sigma} f_{j\sigma}$, a half-filled Fermi sea of spinons [10].

⇒ What about intermediate values of K? What is the effect of next-neighbor $J_2 > 0$?

We consider variationally all nearest-neighbor singlet and triplet pairing instabilities: s-, p_x+ip_y, d_x+id_y, f-wave ($\varphi = n\pi/3$; $n = 0,1,2,3$), nodal d-wave ($d^2_x - d^2_y$), and deformations; finite-momentum pairing ("amperean" state [11]). Furthermore, we check all known planar orders (120° AFM, columnar, spiral).

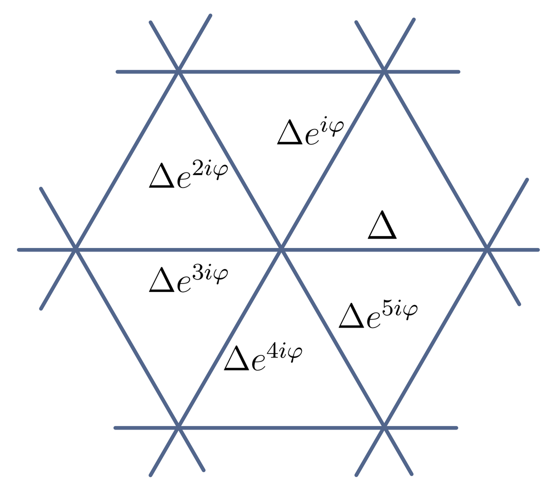


FIGURE 1: Rotation invariant nearest-neighbor parings $\Delta^{\sigma\bar\sigma}_{ij}$

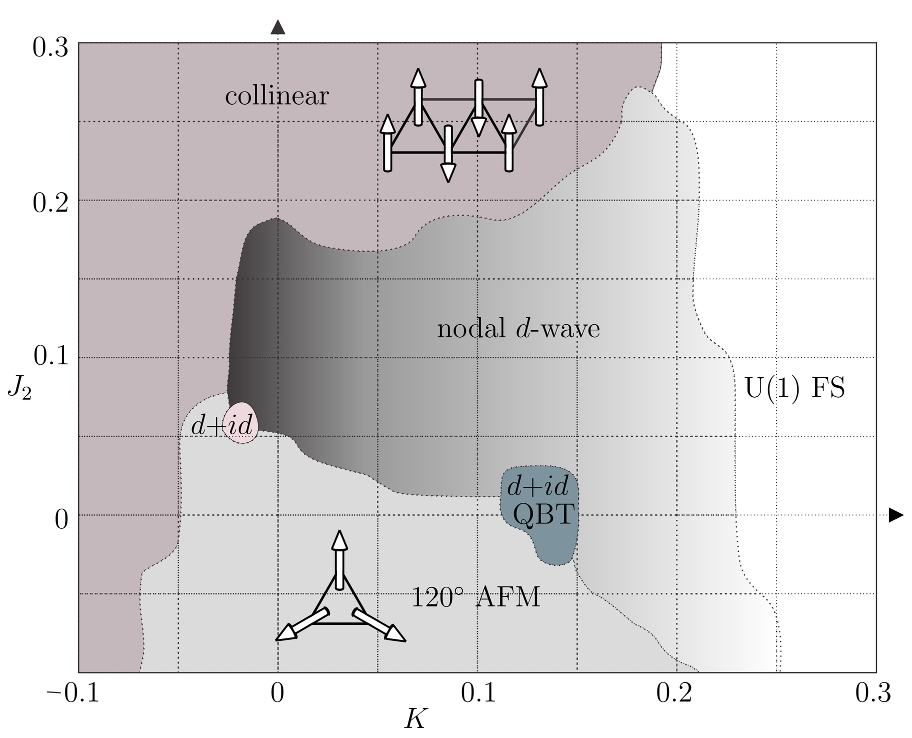


FIGURE 2: Variational phase diagram of model (5). A pure $d+id$ pairing state w. quadratic bands touching is stabilized at $K \simeq 0.13$.

⇒ We find a novel $\mathbb{Z}_2$ QSL state with quadratic bands touching (QBT) at $\boldsymbol{k} = 0$ as $\Delta\to\infty$, stabilized around $K \simeq 0.13$, $J_2 \simeq 0$.

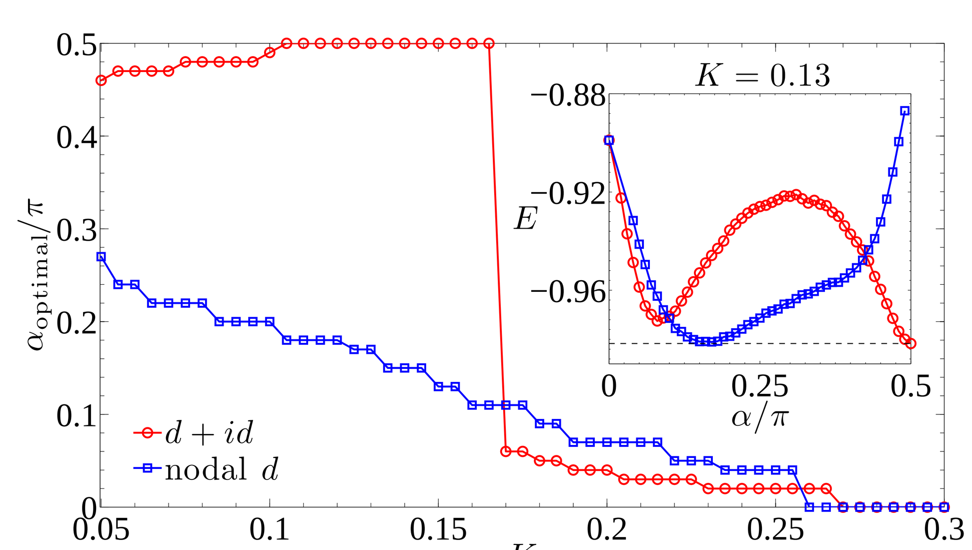


FIGURE 3: Optimal $\alpha = \arctan\Delta$, for $d+id$ and d-wave at $J_2 = 0$.

Conclusion

Described QBT state has several appealing properties, and it provides a new explanation for the physics of organic QSL candidate materials:

- Lowest energy among wide range of states in a realistic spin model
- No broken symmetries (T-rev, S-rot, lattice)
- $\mathbb{Z}_2$ state, so controlled weak-coupling calculations are possible, i.e. no technical issues due to strong gauge fluctuations
- Quadratic band touching of fermionic spinons, constant DOS
- Marginal perturbations may open a small gap (seen in thermal conductivity measurements in κ-ET)

Spin 1

Fractionalization into *three* spinons (2) leads to new and surprising possibilities. Pairing instabilities of the U(1) Fermi sea QSL are

- Singlet (equal flavor) pairing: $\Delta^{xx} = \Delta^{yy} = \Delta^{zz}$; $p+ip$-wave (fully gapped) and f-wave (nodal) $\mathbb{Z}_2$ QSLs.
- Triplet: $\Delta^{xy} = -\Delta^{yx}$, $\Delta^{za} = 0$; s- and $d+id$-wave. The gauge boson is "higgsed", but f_z spinon must remain unpaired in the presence of lattice symmetries: emergence of a free spinon Fermi surface!

While the first option leads to conventional topological or nodal QSL states, the second represents a novel class: $\mathbb{Z}_2$ QSLs in a Higgs phase *and* exhibiting an extended Fermi surface; this is impossible for $S = 1/2$.
⇒ Is this exotic phase stable in a microscopic Heisenberg model?

1st trial: Bilin-biquad. Heisenberg model with single-ion anisotropy

$$H_{KD} = \sum_{\langle i,j\rangle}\boldsymbol{S}_i\cdot\boldsymbol{S}_j + K(\boldsymbol{S}_i\cdot\boldsymbol{S}_j)^2 + D\sum_j S^2_{zj}\,, \tag{6}$$

for $D = 0$ exhibits Néel ($K < 1$), and spin nematic ($K > 1$) order [12]. What happens at finite D?

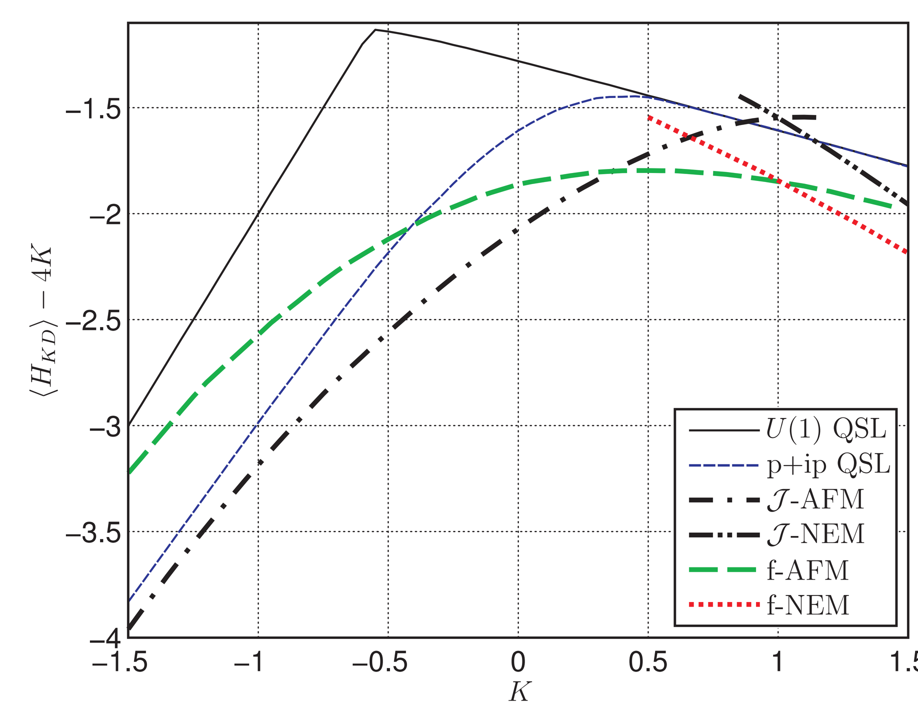


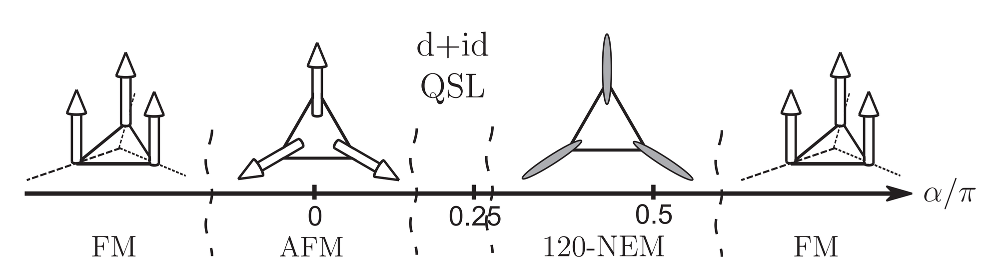
FIGURE 4: Variational energies per site in model (6) at $D = -0.4$. AFM states are realized for $K < 1$, quadrupolar states for $K > 1$.

⇒ Tsunetsugu-Arikawa three-sublattice ordered states [13] at $D = 0$ are not destroyed, just deformed for $D \neq 0$ ($|D| < 1.5$).

Next, we focus on the SU(3) symmetric point $K = 1$ and $D = 0$. For spin 1, the swap operator is $P_{ij} = \boldsymbol{S}_i\cdot\boldsymbol{S}_j + (\boldsymbol{S}_i\cdot\boldsymbol{S}_j)^2 - 1$. A natural ring-exchange model to consider is

$$H_\alpha = \cos\alpha\sum_{\langle i,j\rangle} P_{ij} + \sin\alpha\sum_{\langle i,j,k\rangle} P_{ijk} + \text{h.c.}\,, \tag{7}$$

where $\langle i,j,k\rangle$ are elementary triangles, and $P_{ijk} = P_{ij}P_{jk}$.



⇒ The $d+id$ f_z-Fermi surface QSL is the best variational state at $\alpha \simeq \pi/4$. For $\alpha \gtrsim 0.3\pi$, SU(3) is spontaneously broken to SU(2) and a Néel order is realized.

Conclusion

It is difficult to stabilize spin-1 QSLs in SU(2) Heisenberg models on the triangular lattice. Strong model deformations are necessary.
⇒ More experiments are necessary to challenge/refine the result on the Nickel compound [3]. Our results on the previously unexplored SU(3) model (7) may find interesting applications in systems of cold atoms.

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