Exotic \( S=1 \) spin-liquid state with fermionic excitations on the triangular lattice
Exotic $S = 1$ spin-liquid state with fermionic excitations on the triangular lattice

Maksym Serbyn, T. Senthil, and Patrick A. Lee

Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA

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Motivated by recent experiments on the material Ba$_3$NiSb$_2$O$_9$, we consider a spin-one quantum antiferromagnet on a triangular lattice with the Heisenberg bilinear and biquadratic exchange interactions and a single-ion anisotropy. Using a fermionic “triplon” representation for spins, we study the phase diagram within mean-field theory. In addition to a fully gapped spin-liquid ground state, we find a state where one gapless triplon mode with a Fermi surface coexists with $d + id$ topological pairing of the other triplons. Despite the existence of a Fermi surface, this ground state has fully gapped bulk spin excitations. Such a state has linear-in-temperature specific heat and constant in-plane spin susceptibility, with an unusually high Wilson ratio.

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The spin liquid (SL) is a long sought exotic state of matter proposed by Anderson,\textsuperscript{1} where long-range magnetic order is destroyed by quantum fluctuations at zero temperature. Some materials have been discovered which are promising candidates for the $S = 1/2$ SL. More recently, possible SL materials with $S = 1$ have been discussed. One example is the insulating spin-1 quantum magnet on a triangular lattice, NiGa$_2$S$_4$, reported by Nakatsuji \textit{et al.}\textsuperscript{7} This material motivated a number of theoretical papers proposing different microscopic realizations of $S = 1$ SL.\textsuperscript{8-11} Recently high-pressure synthesis of the two-dimensional triangular magnet Ba$_3$NiSb$_2$O$_9$ (Ref. 12) has produced two phases which possibly realize two- and three-dimensional $S = 1$ SL.

In particular the 6H-B phase, described as a triangular lattice of $S = 1$ Ni$^{2+}$ ions, shows no magnetic ordering down to $T = 350$ mK, well below the Curie-Weiss temperature scale $\theta_{CW} = -75.5$ K. Such behavior, combined with the frustration of a triangular lattice, suggests the possibility of the SL phase. The spin susceptibility saturates to a constant at low temperatures; specific heat is linear in temperature over a wide range, $T = 0.35–7$ K, with a high coefficient and Wilson ratio $R_W = 5.6$. Such observations are highly unusual for a magnetic insulator and point to a SL with gapless fermionic excitations. Indeed, to the best of our knowledge, the only other example where such behavior has been seen is the organic magnetic insulator and point to a SL with gapless fermionic excitations on the triangular lattice

$\langle \hat{S} \rangle = 0$, but full spin rotation symmetry is broken down to rotations around an axis specified by the director vector $d$ (see Refs. 16 and 17 and the discussion below). For positive $K > J$ the ground state is described by antiferromagnetic (AFM) order. In this state the director vectors $d_i$ on three different sublattices are orthogonal to each other (see Fig. 1), thus breaking the lattice translation symmetry. In the extreme case of easy-plane anisotropy ($D \gg J, |K|$), the GS is a trivial product of states of $S^z = 0$ on all sites, corresponding to the trivial single-site FN order. For large but negative $D$ the system favors collinear ferronematic (FN) order, i.e., nematic order that does not break the lattice translational symmetry. In this state the average spin vanishes $\langle \hat{S} \rangle = 0$.

Even within the framework of SL with fermionic excitations, finding a state describing the experiment is a nontrivial problem. For example, a Fermi surface of neutral spin-carrying excitations is strongly coupled to a $U(1)$ gauge field,\textsuperscript{13,14} and the specific heat is expected to behave as $T^{-2/3}$. On the other hand, paired SL states in the absence of impurities will typically have $C/T \to 0$ in the $T \to 0$ limit. In the present Rapid Communication we propose a candidate SL ground state with a Fermi surface coexisting with fermion pairing which gaps out the gauge field. As a result, this state exhibits the exotic physical properties observed in the experiment. Within the mean field we find our state to be a ground state of a simple Hamiltonian. Our goal is not to find a Hamiltonian which describes the material or to find the ground state of that Hamiltonian. Rather, we are interested in exploring ground states which can explain the specific-heat and susceptibility data and point to further experimental probes of this material.
\[ D/J \]

\[ \langle |S^z = 0 \rangle \]

\[ FN \]

\[ AF \]

\[ AF-XXZ \]

\[ XXZ \]

FIG. 1. Schematic representation of the ground state in different limits of the Hamiltonian (1). The white arrows represent average spin; the arrows with disks indicate the director of the nematic order parameter. Details are discussed in the text.

\[ D \] seems plausible. Likewise, it is not known what sign of the biquadratic exchange \( K \) is realized, even though negative \( K \) can be obtained from the large \( U \) expansion of a certain multiorbital Hubbard model or from coupling to phonons. Therefore, in what follows we study the phase diagram of Hamiltonian (1) for both signs of \( D \) and \( K \) but will assume \( |D|, |K| < J \). Except for very small \( |D| \), this is outside of the regions of the known GSs shown in Fig. 1. In order to get access to the [resonating-valence-bond (RVB)-like] state with fermion excitations, we use the fermion representation of the spin.\( ^{10} \) After this we study the resulting phase diagram in the mean-field approximation.

**Fermion representation.** The spin operator is conveniently represented via a set of three operators called triplons which were bosons, but here we use fermions\( ^{10} \) written as a vector \( \vec{f}_i = (f_{ix}, f_{iy}, f_{iz})^T \),

\[ \vec{S}_i = -i \vec{f}_i \times \vec{f}_i, \quad \vec{f}_i \cdot \vec{f}_i = 1. \]  

In terms of the \( |S^z \rangle \) eigenstates, we used the following basis to represent the states of \( |S = 1, |x \rangle = \sqrt{(1 + | - 1)}/\sqrt{2}, |y \rangle = (|1) - | - 1)}/\sqrt{2}, |z \rangle = -i|0 \rangle \), since it facilitates the handling of the biquadratic term in the Hamiltonian. Equation (2) also imposes a constraint of single occupation in order to exclude unphysical states from the Hilbert space. In the mean-field theory this constraint will be relaxed to hold only on average. There are two possible choices of constraint for the spin-one system: the particle representation that we used above and the hole representation \( \vec{f}_i \cdot \vec{f}_i = 2 \). In contrast to the case of \( S = 1/2 \), these are not equivalent. Nevertheless, they can be mapped into each other by a particle-hole transformation plus a change of the sign of hopping. Therefore, we consider only the particle representation but do not restrict hopping to be positive to include the hole representation.\( ^{26} \)

The chosen spin representation has a remaining \( U(1) \) redundancy:\( ^{10,26} \) One can multiply \( \vec{f}_i \) by a phase factor, leaving the spin intact. In addition, in the absence of \( D \) there is a spin rotation symmetry, realized by the simultaneous rotation of the vectors \( \vec{f}_i \) and \( (\vec{f}_i)^T \). Nonzero anisotropy \( D \) breaks full spin rotation symmetry to rotation symmetry in the \( xy \) plane, supplemented by the reflection of spin along the \( z \) axis.

The bilinear term is expressed via fermions as \( \vec{S}_i \cdot \vec{S}_j = \langle \vec{f}_i \cdot \vec{f}_j \rangle \), using the constraint \( \vec{f}_i \cdot \vec{f}_i = 1 \), the biquadratic term also can be expressed as a product of four fermion operators\( ^{15} \). Adding a Lagrange multiplier to enforce the single occupancy constraint (2) on average, we have

\[ H = \sum_{\langle ij \rangle} \left[ J (\vec{f}_i \cdot \vec{f}_j)^2 + (J - K) (\vec{f}_i \cdot \vec{f}_j)(\vec{f}_i \cdot \vec{f}_j) + K \right] + \sum_i (\mu (1 - \vec{f}_i^\dagger \cdot \vec{f}_i) + D (1 - \vec{f}_i^\dagger \vec{f}_i)). \]  

*Mean-field results.* Having expressed the Hamiltonian via fermion operators, we study the mean-field phase diagram of our model. To unambiguously decouple quartic fermion terms, we use the Feynman variational principle,\( ^{21,22} \) which is equivalent to the trial wave-function approach. We define an action based on the Hamiltonian (3) \( S = \int_0^\beta d\tau \left[ \sum_j f_{ja}(\partial_\tau - \mu) f_{ja} + H_c \right] \), as well as the trial quadratic action \( \bar{S} \), with \( H \) replaced by \( \bar{H} \):

\[ \bar{H} = \sum_{\langle ij \rangle} \left[ \tilde{f}_i^\dagger T_{ij} \tilde{f}_j + \tilde{f}_i^\dagger A_{ij} \tilde{f}_j + H_c \right] + \sum_i \tilde{f}_i^\dagger i \epsilon_i \tilde{f}_i. \]  

The mean-field parameters \( T_{ij}, A_{ij}, \) and \( \epsilon_i \) are determined from the stationary points of the functional \( \Psi(\tilde{S}) = \langle S - \bar{S} \rangle S - \log Z \):

\[ T_{ij}^{ab} = -J \delta_{ab} (f_{ja}^\dagger f_{ja}) + (J - K) (f_{ja}^\dagger f_{ja}), \]

\[ A_{ij}^{ab} = -J (f_{ja} f_{ja}) + (J - K) \delta_{ab} (f_{ja} f_{ja}), \]

\[ \epsilon_i^{ab} = \sum_{\langle ij \rangle} (J (f_{ja}^\dagger f_{ja}) - (J - K) (f_{ja}^\dagger f_{ja})) - D \delta_{ab} \delta_{\alpha z}. \]

For \( T = 0 \), we get the estimate for the ground-state energy \( E_{g.s.} \)\( ^{18} \) for the trial wavefunction \( \bar{H} \), where

\[ E_{g.s.} = \sum_{\langle ij \rangle} \left[ T_{ij}^{ab} (f_{ja}^\dagger f_{ja}) + A_{ij}^{ab} (f_{ja}^\dagger f_{ja}) \right] \]

\[ + \frac{1}{2} \sum_i \left[ \epsilon_i^{ab} (f_{ja}^\dagger f_{ja}) - D (f_{ja}^\dagger f_{ja}) + 6K + 2D \right]. \]  

We search for self-consistent solutions to the mean-field equations that do not break any additional symmetries other than \( T \) reversal. When the full spin rotation symmetry is present, the only possible pairing order parameter is \( \Delta_0 \sim (\vec{f}_i \cdot \vec{f}_j)z \). Such pairing preserves full rotational symmetry in space, with the resulting state being a spin singlet. We call this pairing an odd channel, since it is possible only with an odd orbital momentum, i.e., \( p, f \)-wave pairing. Since in Hamiltonian (1), only in-plane rotational symmetry is present for \( D \neq 0 \), the pairing in the even channel with order parameter \( \Delta_0 \sim (f_{ix} \times f_{iz})x \) is allowed. However, the presence of two order parameters simultaneously violates the symmetry with respect to rotations of \( \pi \) around the \( x \) or \( y \) axis.
Both the aforementioned types of pairing were considered by Liu et al.\textsuperscript{10} in a similar system, however, without anisotropy but with a competing third-nearest neighbor $J$. Their treatment of biquadratic exchange also differs from ours. The result of Ref. 10 was that the pairing in the odd channel always wins. Below, after establishing the mean-field equations for each type of pairing, we identify the region in phase space where even-channel pairing has a lower energy than the odd-channel pairing.

Pairing in an odd channel. We introduce the mean-field parameters $\chi^a$, $n^a$, and $\Delta^a_{\alpha}$, $\alpha = x, y, z$, defined as

$$
\chi^a = \langle f^a_i \bar{f}_{i+e_\alpha} \rangle, \quad n^a = \langle f^a_i \bar{f}_i \rangle, \quad \Delta^a_{\alpha} = \langle f^a_i \bar{f}_{i+e_\alpha} \rangle.
$$

(7)

The vectors $e_1 = (1, 0)$, $e_2 = (1/2, \sqrt{3}/2)$, and $e_3 = e_2 - e_1$ specify the link orientation. The hopping is the same on all links, whereas the pairings for the remaining two orientations are $\langle f^{a_1}_i \bar{f}_{i+e_\alpha} \rangle = \Delta^a_{\alpha} e^{2i\beta}$, $\langle f^{a_1}_i \bar{f}_{i+e_\alpha} \rangle = \Delta^a_{\alpha} e^{i\pi/3}$, where the pair angular momentum $l = 1, 2, 3$ for $p + ip$, $d + id$, and $f$-wave pairing, respectively. Spin rotation symmetry in the $xy$ plane requires $\chi^x = \chi^y$, $n^x = n^y$, $\Delta^z = \Delta^x$. The Hamiltonian in momentum space (modulo nonessentially constant terms) can be rewritten as

$$
\hat{H} = \sum_{k, \alpha} \chi^a_{\alpha} \bar{f}_{i,k} f_{i,k} + \Delta^a_{\alpha} \bar{f}_{i,k} f_{i,k} + \Delta^a_{\alpha} f_{-i,k} f_{i,k},
$$

(8)

with mean-field parameters

$$
\chi^a_{\alpha} = 2\gamma(k)(J - K)\chi^a - J(\chi^x + \chi^y + \chi^z) + 6K n^a - \mu - \delta_{\alpha,z} D,
$$

(9)

$$
\Delta^a_{\alpha} = \psi(k)(J - K)(\Delta^a_{\alpha} - \Delta^x_{\alpha} + \Delta^y_{\alpha} - J \Delta^z_{\alpha}).
$$

(10)

The function $\gamma(k)$ is a sum over nearest neighbors $\gamma(k) = \cos k \cdot e_1 + \cos k \cdot e_2 + \cos k \cdot e_3$. On the other hand, $\psi(k)$ depends on the type of pairing under consideration. Note that $p$-wave pairing breaks the lattice rotational symmetry. Therefore, we consider $p + ip$-wave and $f$-wave pairings: $\psi^p(k) = \left[\sin k \cdot e_1 - \sin k \cdot e_2 + \sin k \cdot e_3\right]$, $\psi^{fwp}(k) = i\left[\sin k \cdot e_1 + e^{2i\pi/3} \sin k \cdot e_2 + e^{4i\pi/3} \sin k \cdot e_3\right]$. Equation (8) is solved with the Bogolyubov transformation acting separately on each fermion species. This results in the spectrum $E_{\alpha} = \sqrt{(\chi^x_{\alpha}/2)^2 + (\Delta^0_{\alpha})^2}$, and mean-field equations

$$
\chi_{\alpha} = \frac{1}{N} \sum_k \frac{1}{6} \gamma(k) \left[ 1 - \frac{\chi^x_{\alpha}}{2E_{\alpha}} \right],
$$

(11a)

$$
\Delta_{\alpha} = \frac{1}{N} \sum_k \frac{1}{3} \psi(k) \frac{\Delta_{\alpha}}{2E_{\alpha}},
$$

(11b)

$$
n^a = \frac{1}{N} \sum_k \frac{1}{2} \left[ 1 - \frac{\chi^x_{\alpha}}{2E_{\alpha}} \right],
$$

(11c)

supplemented by the constraint equation $(f^{\dagger}_i) \cdot (\bar{f}^{\dagger}_i) = 1$.

Pairing in an even channel. Hoppings are defined as in (7), whereas pairing is $\Delta^x = 1/2(f_{i+1,1} f_{i+2,2} - f_{i,1} f_{i+2,2})$. The Hamiltonian is

$$
\hat{H} = \sum_{k, a} \chi^a_{\alpha} \bar{f}_{i,k} f_{i,k} + \Delta^a_{\alpha} \bar{f}_{i,k} f_{i,k} + \Delta^{xy}_{\alpha} f_{-i,k} f_{i,k},
$$

with $\chi^a_{\alpha}$ given by Eq. (9), and $\Delta^{xy} = 2J \psi(k) \Delta^x$. Note, that the $f_z$ band is unpaired and retains its Fermi surface.

We consider $s$-wave and $d + id$-wave pairings (the $d$ wave violates lattice symmetry and higher orbital momentum pairing requires inclusion of further neighbors). For the case of $s$-wave pairing, the function $\psi(k) = \gamma(k)$. For $d + id$-wave pairing we have $\psi^{adi}(k) = \cos k \cdot e_1 + e^{2i\pi/3} \cos k \cdot e_2 + e^{-2i\pi/3} \cos k \cdot e_3$. The Bogolyubov spectrum is $E^s_k = E^s_k = \sqrt{(\chi^x_{\alpha})^2 + (\Delta^0_{\alpha})^2}$, $E^p_k = \chi^x_{\alpha}$. Self-consistent mean-field equations for the $x$ and $y$ components are given by Eq. (11) with the new expressions for the spectrum and gap functions. For the $z$ component we have

$$
\chi^z = \frac{1}{N} \sum_k \frac{1}{3} \gamma(k) n_f(\chi^z_k), \quad n^z = \frac{1}{N} \sum_k n_f(\chi^z_k).
$$

Our mean-field approach automatically includes on-site FN order. The on-site nematic order is described by the order parameter tensor $Q^{opp} = 1/2(\delta^x \delta^x + \delta^y \delta^y) - 2/3 \delta^z \delta^z$. For a single site with $S = 1$ all states with zero average spin ($\langle \hat{S}_z \rangle = 0$) can be characterized by the unit director vector $d$,\textsuperscript{20} in the basis defined earlier, $|d| = d_x|\chi| + d_y|\gamma| + d_z|\delta|$. For this state $Q^{opp}$ is expressed via $d$ as $Q^{opp} = 1/3d_x d_y - d_y d_z$. For example, $d_3$ corresponds to the state $|\delta| = 0$, and the nematic order is diagonal, $Q^{opp} = diag(1/3, 1/3, -2/3)$. In our model we also have states with vanishing spin order and diagonal on-site nematic order. However, since our GS is RVB like with long-range entanglement, $Q^{opp}$ cannot be described by the above simple form. We have to introduce the magnitude $q$, $Q^{opp} = q(1/3d_x d_y - d_y d_z)$. Calculating the nematic order parameter tensor in our model, we have $Q^{opp} = \delta_{opp}[1/3 - n^z]$, where $n^z$ is the average occupation of corresponding fermion. Since $n^z = n^x$, we have nematic order with $d||z$, with a magnitude given by $q = n^z - n^x$, varying from 1 for $n^z = 1$ (state $|\delta| = 0$) to $-1/2$ for $n^z = 0$. Nonzero anisotropy $D \neq 0$ causes $n^a$ to be different from 1/3, and therefore directly coupled to FN order along the $z$ axis.

Having studied the energies of all the aforementioned states using Eq. (6), we found that the main competition is between states with $p + ip$ and $d + id$-wave pairings, with all other states being higher in energy. As one increases $K$, the effective coupling for the odd-channel pairing decreases, whereas for even pairing it remains the same. Finally, for $K \approx 0.45 J$, singlet pairing wins. The resulting phase diagram is shown in Fig. 2. The boundary between the two states appears to be weakly dependent on $D$.

Physical properties of the $d + id$ state. The $d + id$ state breaks the time-reversal symmetry. The chiral order parameter associated with this broken symmetry $\langle \tilde{S}_j \cdot (\tilde{S}_{j+1, \times} \times \tilde{S}_{j+1, \times}) \rangle \propto \gamma^2(\Delta^0)^2$ is proportional to the magnitude of the pairing gap squared. In addition, pairing with $d + id$ gap symmetry in two dimensions is topological,\textsuperscript{21} resulting in the existence of a pair of zero-energy edge modes at the boundaries. The physics of these modes will be discussed elsewhere.

The combination of gapless excitations with topological pairing gives rise to a number of unusual physical properties that may explain the results of the recent experiment.\textsuperscript{12} Due to ungapped $f_z$ excitations, the specific heat depends linearly on temperature near $T = 0$, $C = \pi^2 k_B^2 \nu_z T/3$, where $\nu_z$ is the density of states of $f_z$ at the Fermi surface. Due to the Higgs mechanism, the gauge field is massive and does not
susceptibility exhibits more exotic behavior: Due to the pairing
$D/J = 8/3 \approx 2.66$ for the case of small anisotropy, and $R_W \rightarrow 16/3 \approx 5.33$ for large anisotropy. Note that we
take the average susceptibility $\tilde{\chi} = 2/3 \chi_{xx}$ to account for the
polycrystalline nature of the sample. The latter value
gives surprisingly good agreement with the Wilson ratio observed experimentally, $R_W \approx 5.63$. We also calculated the
imaginary part of the spin susceptibility. Since two out of three
fermions are gapped, $\text{Im}\chi_{xx}(\omega, q)$ vanishes for temperatures and
frequencies smaller than the gap for all $\alpha$. This implies that the NMR relaxation $1/(T, T)$ is exponentially small for
temperatures below the pairing scale. These results tell us that the
Fermi surface associated with $f_1$ [see Fig. 2(b)] should be viewed very differently than the spinon Fermi surface in the $S = 1/2$ SL, which carries spin-1/2 quantum numbers and leads to gapless spin-1 excitations. In our case $S' = 1$ excitations are gapped even though the static spin susceptibility
$\chi_{xx, xx} \neq 0$ and the specific heat has a linear $T$ dependence.

Finally, we discuss experiments that could confirm the
proposed ground state. Measurement of the spin susceptibility for single-crystal or oriented powder samples is of great interest in order to test our prediction of strong anisotropy. We also predict an exponentially activated behavior for $1/(T, T)$ which may be surprising in view of the linear $T$ behavior of the specific heat.

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