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

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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.

Our model system consists of quantum $S = 1$ spins forming a triangular lattice. For simplicity, we consider only nearest-neighbor interactions. The general form of the Hamiltonian can be written as

$$H = \sum_{\langle ij \rangle} \left[ J \vec{S}_i \cdot \vec{S}_j + K (\vec{S}_i \cdot \vec{S}_j)^2 \right] + D \sum_{i} (\vec{S}_i^z)^2,$$

where we included the Heisenberg exchange interaction with coupling $J > 0$ and the biquadratic exchange with coupling $K$. In addition, we allow easy-plane or easy-axis type of anisotropy controlled by the parameter $D$, but we neglect this anisotropy in the couplings $J$ and $K$ since it is presumably small for transition metals. The Hamiltonian (1) has been considered in the literature in the limits when the anisotropy is either zero or dominates over other couplings, or there are longer-range competing exchange couplings. Figure 1 summarizes known results for the ground-state (GS) phase diagram in a schematic way. There are three different phases on the line of zero anisotropy $D = 0$: 15–18 In the range $K = -0.4J \cdots J$, GS is a 120$\degree$ antiferromagnet (AFM). For larger negative $K$ the system favors collinear ferromagnetic (FN) order, i.e., nematic order that does not break the lattice translational symmetry. In this state the average spin vanishes $\langle |\vec{S}| \rangle = 0$, 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 anisotropic (AFN) order. In this state the director vectors $d_{ij}$ 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 $|S^z = 0\rangle$ on all sites, corresponding to the trivial single-site FN order. For large but negative $D$, implying extreme easy-axis anisotropy, only two states with $S^z = \pm 1$ on each site survive. This system can be described by a spin-$1/2$ XXZ model with all exchange couplings being antiferromagnetic if $2J > K > 0$ or with $J^z$ being frustrating and $J^\perp$ ferromagnetic if $K < 0$. In both cases there is spin-density-wave ordering of the $z$ component of the spin in the GS, supplemented by planar AFN order in the former case and collinear nematic order in the latter case.19

Physically for Ba$_3$NiSb$_2$O$_9$ we may expect the exchange coupling $J$ to be the largest with $J > |K|/|D|$. Both signs of
spin; the arrows with disks indicate the director of the nematic order $K$ seems plausible. Likewise, it is not known what sign of MAKSYM SERBYN, T. SENTHIL, AND PATRICK A. LEE PHYSICAL REVIEW B 84 $\alpha$ represents the states of can be obtained from the large multiorbital Hubbard model or from coupling to phonons. rotation symmetry, realized by the simultaneous rotation of $D$ imposes a constraint of single occupation in order to exclude the stationary points of the functional $\Psi[S] = \langle S - \bar{S} \rangle S - \log \tilde{Z}$:

$$T_{ij}^{a\beta} = -J \delta_{a\beta}(f_j^\dagger f_i),$$

$$A_{ij}^{a\beta} = -J \langle f_j f_i \rangle + (J - K) \delta_{a\beta}(f_j^\dagger f_i),$$

$$t_i^{a\beta} = \sum_{(j)} [T_{ij}^{a\beta} f_{j\alpha} + A_{ij}^{a\beta} f_{i\alpha}] + D \delta_{a\beta} \delta z.$$

For $T = 0$, we get the estimate for the ground-state energy $E_{\text{gs}} \lesssim \langle H \rangle_\bar{S}$, where

$$E_{\text{gs}} = \sum_{(i)} \left[ T_{ij}^{a\beta} f_{j\alpha} + A_{ij}^{a\beta} f_{i\alpha} f_{j\beta} \right] + \frac{1}{2} \sum_i \left[ t_i^{a\beta} f_{i\alpha} f_{i\beta} - D \langle f_{i\alpha} f_{i\beta} \rangle + 6K + 2D \right].$$

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 $\Delta_\alpha \sim \langle f_i^\dagger f_j \rangle$. Such pairing preserves full rotational symmetry in the $xy$ plane, 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 $\Delta_\alpha \sim \langle f \times f \rangle_z = \langle f_i^\dagger f_j - f_i f_j^\dagger \rangle$ 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. 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

$$\chi^a = \langle f_{ia}^\dagger f_{i+\ve{e}_a} \rangle, \quad n^a = \langle f_{ia}^\dagger f_{ia} \rangle, \quad \Delta^a_{\alpha} = \langle f_{ia} f_{i+\ve{e}_a} \rangle. \quad (7)$$

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 $\langle f_{ia} f_{i+\ve{e}_a} \rangle = \Delta^a_{\alpha} e^{i\beta}, \quad \langle f_{ia} f_{i+\ve{e}_a} \rangle = \Delta^{a^\prime} e^{j\beta}$, 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 $\chi^x = \chi^y, n^x = n^y, \Delta^x_{\alpha} = \Delta^y_{\alpha}$. The Hamiltonian in momentum space (modulo nonessential constant terms) can be rewritten as

$$\hat{H} = \sum_{k, \alpha} \chi^a f_{ka}^\dagger f_{ka} + \Delta^a_{\alpha} f_{ka}^\dagger f_{-k\alpha} + \Delta^-_{\alpha} f_{-k\alpha}^\dagger f_{ka}. \quad (8)$$

with mean-field parameters

$$\chi^a = 2\gamma(k)(J - K)\chi^a - J(\chi^x + \chi^y + \chi^z), \quad + 6K n^a - \mu - \delta^{ad}\zeta D, \quad (9)$$

$$\Delta^a_{\alpha} = \gamma(k)\left[(J - K)(\Delta^a_{\alpha} + \Delta^a_{\beta} + \Delta^a_{\gamma}) - J \Delta^a_{\delta}\right]. \quad (10)$$

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, $\psi$ 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: $\psi(k) = i(\sin k \cdot e_1 - \sin k \cdot e_2 + \sin k \cdot e_3), \quad \psi(k) = i(\sin k \cdot e_1 + e^{i2\pi/3} \sin k \cdot e_2 + e^{i4\pi/3} \sin k \cdot e_3). \quad (8)$

Equation (8) is solved with the Bogolyubov transformation acting separately on each fermion species. This results in the spectrum

$$E_k = \sqrt{(\chi^a/2)^2 + |\Delta^a_{\alpha}|^2}, \quad \text{mean-field equations for} \quad \chi^a = 1/N \sum_k 1/3 \gamma(k) \left[1 - \chi^a_{2E_k^a}\right], \quad (11a)$$

$$\Delta^a_{\alpha} = 1/N \sum_k 1/3 \psi(k) \Delta^a_{\alpha}/E_{2k}, \quad (11b)$$

$$n^a = 1/N \sum_k 1/2 \left[1 - \chi^a_{2E_k^a}\right]. \quad (11c)$$

supplemented by the constraint equation \((f_{i+\ve{e}_a}^\dagger \cdot f_i^\dagger) = 1\).

Pairing in an even channel. Hoppings are defined as in (7), whereas pairing is $\Delta^S_{xy} = 1/2(f_{1i+ej_{i+ej_{i+e1}}})$. The Hamiltonian is

$$\hat{H} = \sum_{k, a} \chi^a f_{ka}^\dagger f_{ka} + \Delta^a_{\alpha} f_{ka}^\dagger f_{-k\alpha} + \Delta_{\alpha}^{S_{xy}} f_{-kj}^\dagger f_{kj},$$

with \(\chi^a_{k}\) given by Eq. (9), and $\Delta_{\alpha}^{S_{xy}} = 2J \psi(k) \Delta_{\alpha}^{S_{xy}}$. 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 $\psi^S(k) = \gamma(k)$. For $d + id$-wave pairing we have $\psi^d(k) = \cos k \cdot e_1 + e^{i2\pi/3} \cos k \cdot e_2 + e^{i2\pi/3} \cos k \cdot e_3$. The Bogolyubov spectrum is $E^S_k = (\chi^a_{2E_k^a})^2 + |\Delta^a_{\alpha}|^2$, $E^d_k = \chi_{k}^a$. 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

$$\chi^z = 1/N \sum_k 1/3 \gamma(k) n_f(\chi^z), \quad n^z = 1/N \sum_k n_f(\chi^z).$$

Our mean-field approach automatically includes on-site FN order. The on-site nematic order is described by the order parameter tensor $Q^{\alpha\beta} = 1/2(S^\alpha S^\beta + S^\beta S^\alpha) - 2i/3\delta^{\alpha\beta}$. For a single site with $S = 1$ all states with zero average spin ($\bar{S} = 0$) can be characterized by the unit director vector $d$, in the basis defined earlier, \(|d| = d_1(|\chi| + d_1(|\gamma| + d_1(|\delta|). For this state $Q^{\alpha\beta}$ is expressed via $d$ as $Q^{\alpha\beta} = \bar{1}/3\delta_{\alpha\beta}d_\alpha d_\beta$. For example, $d_3$ corresponds to the state $|4S^z = 0\rangle$, and the nematic order is diagonal, $Q^{\alpha\beta} = \bar{1}/3(1, 1, 2, 3)$. 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, $Q^{\alpha\beta}$ cannot be described by the above simple form. We have to introduce the magnitude $q$, $Q^{\alpha\beta} = q(1/3\delta_{\alpha\beta} - d_\alpha d_\beta)$. Calculating the nematic order parameter tensor in our model, we have $Q^{\alpha\beta}_{nF} = \delta_{\alpha\beta}[1/3 - n^a]$, where $n^a$ is the average occupation of corresponding fermion. Since $n^a = n^z$, 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 $|S^z = 0\rangle$) to $-1/2$ for $n^z = 0$. Nonzero anisotropy $D \neq 0$ causes $n^a$ to be different from 1/3, and therefore directly couples 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 $(\vec{S}_f \cdot (\vec{S}_{i+ej_{i+ej_{i+e1}}}) \propto \chi^z|\Delta^S_{xy}|^2$ is proportional to the magnitude of the pairing gap squared. In addition, pairing with $d + id$ gap symmetry in two dimensions is topological, 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. Due to ungapped $f_{z}$ excitations, the specific heat depends linearly on temperature near $T = 0$, $C = \pi^2 k_B v_t T/3$, where $v_t$ 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
The behavior of $\tilde{\chi}_{xx}$ is shown in Fig. 2(a). We calculate the Wilson ratio, defined as $R_W = (4\pi^2 k_F^2)/(3g^\mu_B^2)\langle \tilde{\chi}(T) \rangle / C$, and obtain $R_W = 8/3 \approx 2.66$ 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, $\text{Im}\chi(\omega, q)$ vanishes for temperatures and frequencies smaller than the gap for all $\alpha$. This implies that the NMR relaxation $1/(T_S T)$ is exponentially small for temperatures below the pairing scale. These results tell us that the Fermi surface associated with $f_s$ [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.

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 $1/(T_S T)$ which may be surprising in view of the linear $T$ behavior of the specific heat.

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20. This is different from Ref. 10, where authors use a combination of particle and hole constraints in order to preserve particle-hole symmetry. Our treatment violates particle-hole symmetry from the very beginning.