Spontaneous Spin Ordering of a Dirac Spin Liquid in a Magnetic Field

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Spontaneous Spin Ordering of a Dirac Spin Liquid in a Magnetic Field

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The Dirac spin liquid was proposed to be the ground state of the spin-1/2 kagomé antiferromagnets. In a magnetic field B, we show that the state with Fermi pocket is unstable to the Landau level (LL) state. The LL state breaks the spin rotation around the axis of the magnetic field. We find that the LL state has an in-plane 120° q = 0 magnetization M which scales with the external field M ∼ Bα, where α is a universal number of the Dirac spin liquid. We discuss the related experimental implications which can be used to detect the possible Dirac spin liquid phase in herbertsmithite ZnCu₃(OH)₆Cl₂.

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Spin liquids (SL), defined as the ground states of spin systems with half integer spins per unit cell which does not order magnetically and/or break translation symmetry, are believed to contain fundamentally new physics beyond Landau’s symmetry breaking characterization of phases. After years of search, a promising candidate finally emerged in the spin-1/2 kagomé system herbertsmithite ZnCu₃(OH)₆Cl₂ [1–3]. Despite an antiferromagnetic (AFM) exchange J = 170–190 K [4,5], the system does not order down to 50 mK [1–3]. Theoretically the Dirac SL is proposed [6] as the ground state of the nearest neighbor Heisenberg antiferromagnetic model on kagomé lattice. Unfortunately the spin susceptibility χ₀ is consistent with ~4% magnetic impurities [4], and possible Dzyaloshinskii-Moriya (DM) interaction has also been proposed [5]. These may explain the hump of the specific heat C around 2 K and also obscure the C ∼ T² and χ₀ ∼ T behaviors predicted by the Dirac SL. In this Letter we propose a unique signature of the Dirac SL in an external magnetic field, which can be used to detect the Dirac SL phase in experiment and numerical simulations.

The Dirac SL experiences no orbital effect in the external magnetic field because it is an insulator. The Zeeman coupling gμ_BB · ∑Sz polarizes spin along the field direction, breaks time reversal, and breaks SU(2) spin rotation down to U(1). Let us denote the direction of B as the z direction (note that the xy plane does not have to be the plane of the two-dimensional kagomé system). We find that the Dirac SL will spontaneously break the remaining Sz-U(1) and form a staggered magnetization M [see Fig. 2(b)] in the xy plane which scales with B: M ∼ Bα.

The positive exponent α is an intrinsic universal number of the Dirac SL phase and in principle calculable.

This unique signature of the Dirac SL can be compared with a regular coplaner AFM ordered phase. In a small magnetic field B, the magnetization of the regular AFM phase would rotate into the xy plane to maximize the susceptibility along the B direction. As B is tuned to zero, the in-plane magnetization remains finite. In contrast, for the Dirac SL we predict that M vanishes as Bα. This suggests that the Dirac SL can be viewed as an AFM phase whose long-range AFM order has been destroyed by the quantum fluctuations.

We begin with writing down the low-energy theory of the Dirac SL [6], which includes four flavors of fermions coupled with compact U(1) gauge field in 2 + 1 dimension (QED2):

\[
S = \int d^3x \left[ \frac{1}{g^2} (e_A \mu_\alpha \partial_\mu \psi_\sigma)^2 + \sum_\sigma \bar{\psi}_+ (\partial_\mu - i a_\mu) \tau_\mu \psi_+ + \sum_\sigma \bar{\psi}_- (\partial_\mu + i a_\mu) \tau_\mu \psi_- \right] + \cdots,
\]

where the two-component fermionic Dirac spinon fields are denoted by ψ±, where ± label the two inequivalent nodes and σ the up or down spins. In the presence of a magnetic field, the simplest guess is that the Dirac points will change into Fermi pockets due to Zeeman splitting. We will call this state Fermi pocket (FP) state. But, can there be other states whose energy may be lower?

The FP state has many gapless spinon excitations near the Fermi pockets and in general is not energetically favorable. One natural way to gap out the Fermi pocket state is to induce (internal) gauge fluxes and develop Landau levels (LL). Here we take advantage of the appearance of zero-energy Landau levels when Dirac particles are subject to a magnetic field, as is well known in the recent studies of graphene [7,8]. If the gauge flux is adjusted in such a way that the zero-energy Landau level is fully filled for up-spin, and fully empty for down-spin, then the spinons are fully gapped (Fig. 1). We will call this state Landau level state.

In the following we show that LL state has lower energy than the FP state. We first compare the energies of the LL state and the FP state at the mean-field level (ignoring the gauge fluctuation) with fixed Sz polarization. We set aside the Zeeman energy which is common between the two states. The density of spin imbalance is

\[
\Delta n = \frac{\Delta N}{A} = \frac{N_+ - N_-}{A} = \frac{1}{4\pi} \left( \frac{\mu_B B}{v_F} \right)^2,
\]

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\]

where the two-component fermionic Dirac spinon fields are denoted by ψ±, where ± label the two inequivalent nodes and σ the up or down spins.
FIG. 1. The spinon is gapless in FP state with an electronlike Fermi pocket for spin-up spinons and holelike pocket for spin-down spinons. The spinons are fully gapped in the LL state. Each Landau level is doubly degenerate due to two inequivalent Dirac nodes.

The mean-field level the energy cost of magnetic field is between the sum of the energies of all negative Landau energy of the mean-field LL state is just the difference (3) and (4) we have

\[ \Delta e_{\text{FP}} = \Delta E_{\text{FP}} / A = 2\sqrt{\pi} v_F (\Delta n)^{3/2}, \]

where \( N_1 \) and \( N_\parallel \) are the number of up and down spins, \( \Delta E \) is the energy increase compared to Dirac point state, \( A \) is the system area, \( B \) is the magnetic field, and \( v_F \) is the mean-field Fermi velocity. The factor 4 in Eq. (2) is from the fact that there are two spin degeneracy and two nodal degeneracy. We choose our units such that \( h = 1 \).

To make sure the LL state has the same spin imbalance as the FP state, each Landau level should contain \( \Delta n A/2 \) states; i.e., the induced gauge magnetic field \( b \) satisfies \( \Delta n = \frac{2b}{2\pi} \), where the \( 2\pi \) is gauge flux quantum. At the mean-field level the energy cost of magnetic field is entirely due to the fermion band contribution [9], namely, the energy of the mean-field LL state is just the difference between the sum of the energies of all negative landau levels, and the filled Fermi sea:

\[ \Delta e_{\text{LL}} = 4v_F \left( \frac{b}{2\pi} \sum_{n=1}^{\infty} \sqrt{2n} b - \frac{\int dk^2}{(2\pi)^3} (-k) \right) = \frac{3}{2\pi} b^{3/2} v_F = \frac{3}{2\pi} (3/2)^{3/2} v_F. \]

The same result was obtained in Refs. [10,11]. From Eqs. (3) and (4) we have \( \Delta e_{\text{FP}} / \Delta e_{\text{LL}} = \frac{2\sqrt{\pi} (3/2)^{3/2}}{3} \approx 1.134 \); i.e., the LL state has lower mean-field energy than the FP state.

To include the effect of gauge fluctuations and to go beyond the mean-field theory, we will use the Gutzwiller projected wave function to calculate the energies of LL and FP states. To obtain the Gutzwiller projected wave function we first write down the mean-field Hamiltonian on lattice:

\[ H_{\text{mean}} = \sum_{ij} \chi_{ij} f^\dagger_{i\sigma} f_{j\sigma}, \]

where \( f_{i\sigma} \) are the fermionic spinons. The mean-field ground state \( \{|\Psi_{\text{mean}}(\chi_{ij})\rangle\} \) with mean-field parameters \( \chi_{ij} \) is a spin singlet. The projected wave function \( \{|\Psi_{\text{proj}}(\chi_{ij})\rangle\} = \{|\Psi_{\text{mean}}(\chi_{ij})\rangle\} \) removes the unphysical states and becomes a spin state; here, \( P = \prod_i (1 - \gamma_i \gamma_i) \) is the projection operator ensuring one fermion per site. The physical observables can be measured on \( \{|\Psi_{\text{proj}}(\chi_{ij})\rangle\} \) by a Monte Carlo approach [12]. The Dirac SL is characterized by \( \chi_{ij} \) such that \( |\chi_{ij}| = \chi \) is a bond independent constant and there are \( \pi \) fluxes through the hexagonal plaquettes and zero fluxes through the triangular plaquettes [6]. Note that the projected Dirac SL state has no tunable parameter since \( \chi \) only gives the wave function an overall factor. On a 16 × 16 unit cell lattice, the energetics of the FP and LL states are given in Table I.

Is the \( \Delta e \sim \Delta n^{3/2} \) law still valid after projection? The answer is positive because the \( S_z \) and energy are conserved and have no anomalous dimension. The energies in Table I can thus be fitted [Fig. 2(a)] as

\[ \Delta e_{\text{FP}}^{\text{proj}} = 0.33(2) \Delta n^{3/2} + 0.00(4) \Delta n^2, \]

(5)

\[ \Delta e_{\text{LL}}^{\text{proj}} = 0.223(6) \Delta n^{3/2} + 0.03(1) \Delta n^2, \]

(6)

where the units are chosen such that the unit cell spacing \( a = 1 \) and \( J = 1 \) and we include the first order correction to scaling, the \( \Delta n^2 \) term. From the coefficient 0.33(2) in Eq. (5) we are able to read off the effective Fermi velocity \( v_F = \frac{3^{3/2} \Delta n^{1/2}}{2\pi} \approx 0.28(2) \frac{a}{\sqrt{h}} \) by fitting to the free fermion result Eq. (3). \( v_F \) is almost twice the mean-field value \( v_F = \frac{a}{\sqrt{h}} \) with \( \chi = 0.221 \) [13]. For herbertsmithite this means \( v_F^\ast = 4.9 \times 10^3 \text{ m/s} \) assuming \( J = 200 \text{ K} \). This is very close to the Fermi velocity we found by a projected band structure study [14], where only one particle-hole excitation is considered. The closeness of the two results implies that the gauge interactions between many particle-hole excitations may only give small corrections for energetics.

From Eqs. (5) and (6) we see that the LL state has a lower energy than the FP state. Therefore the LL state may be the true ground state in the presence of a magnetic field. Because of the presence of internal gauge flux, one can easily see that the LL state breaks parity (mirror reflection). Surprisingly, it turns out that the LL state breaks the \( S_z \) spin rotation as well. Since the spinons \( \psi \) are gapped due to the Landau levels, the gauge field \( a_{\mu} \) is the only low-energy excitation. We note that the Hall conductance of the spin-up spinons is +1 and the Hall conductance for the spin-down spinons is −1. Thus the total Hall conductance is zero and there is no Chern-Simons term for the gauge field \( a_{\mu} \). As a result, the dynamics of the gauge field is cons-

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<td>−0.41994(2)</td>
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vector normal to \( r \) between the compact Bose condensation. The argument relies on the duality superfluidity, where the phonon mode is the signature of energy density \( C_1 \). It corresponds to the Goldstone mode of the ordered vortex in the \( XY \) field and spin imbalance density \( C_1 \). We also present the relative phase of \( S_i^+ \) operator computed by projected Landau level state in unit of \( \pi \) [see the paragraph following Eq. (7)]. Note that in computing \( (S_i^+)/(S_j^+) \) we fix the left-bottom corner site as the site \( i \) and scan the site \( j \) for the nine sites depicted.

trolled by the Maxwell term \( f_{\mu
u} \), which arise from integrating out high-energy fermions, and gives rise to a linear dispersed gapless photon mode.

To understand the meaning of the gapless mode, we note that due to the nonzero Hall conductances \( \pm 1 \) for the spin-up and spin-down spinons, a flux quantum of the gauge field \( a_{\mu} \) will induce one spin-up spinon and minus one spin-down spinon. Thus a flux quantum carries a spin quantum number \( S_z = 1 \). The “magnetic field” \( b = \epsilon_{ij}\partial_i a_j \) is nothing but the \( S_z \) density. Therefore, the linear gapless mode of \( a_{\mu} \) is actually the density fluctuations of \( S_z \). The appearance of a linear gapless mode of a conserved density as the only low lying excitation implies that the corresponding symmetry is spontaneously broken in the ground state [15,16]. The more familiar example is that of superfluidity, where the phonon mode is the signature of Bose condensation. The argument relies on the duality between the compact \( U(1) \) gauge theory and the \( XY \) model \( \mathcal{L} = \sum_{\mu}(\partial_{\mu}\theta)^2 \) in two dimensions [17,18], where \( b \) corresponds to \( \theta \) and the electric field \( \mathbf{e} \) corresponds to a vector normal to \( \nabla \theta \) (to be precise, \( e_i = \epsilon_{ij}\partial_j \theta \)). The latter relation implies that electric charge which is the source of \( 1/r \) electric field in the gauge theory corresponds to a vortex in the \( XY \) ordered phase. In the deconfined (Coulomb) phase, the photon is the only gapless excitation. It corresponds to the Goldstone mode of the ordered \( XY \) model [17].

By definition the insertion of a monopole at time \( \tau \) creates a \( 2\pi \) flux. In our case the additional flux implies the addition of \( S_z = 1 \). Since the total spin is conserved, the appearance of free monopoles and antimonopoles is strictly forbidden and we are in the Coulomb phase. On the other hand we can insert by hand a monopole at position \( i \) and remove it at position \( j \). The action of the monopole-antimonopole pair in \( 2 + 1 \) dimensions is the same as the Coulomb energy between charges in three spatial dimensions and goes as \( 1/r \). Hence \( \langle V(r)^\dagger V(r') \rangle \sim \exp(-|r - r'|^{-1}) \) has long-range correlation. Since the slowly varying field operator \( V^\dagger \) increases \( S_z \) by one, it is related to the lattice-scale spin operator \( S_i^+ \), but with a possible staggering pattern. That is, we may write \( V^\dagger \sim \sum_{i} e^{i\theta} S_i^+ \), where the sum is restricted to the lattice sites in the vicinity of where the flux is inserted. The above reasoning thus indicates that the spin is ordered in the \( xy \) plane.

What is the spatial pattern of the spin ordering? We have calculated spin-spin correlation function \( \langle S_i^+ S_{i'}^- \rangle \) of the projected LL state described earlier above Eq. (5) and found the \( XY \) ordered pattern as shown in Fig. 2(b). This order pattern is referred to as the \( q = 0 \) magnetic order [19]. To verify this in a different way, we compute

\[
\langle S_i^+ \rangle \equiv \langle n + 1|P S_i^+ P|n \rangle,
\]

where \( P|n \rangle \) denotes the projection of a LL state with \( n \) flux quanta. Since \( S_i^+ \) necessarily increases the flux by \( 2\pi \), the matrix element in Eq. (7) must connect \( P(n) \) and \( P(n + 1) \). The long-range order in \( V^\dagger \) implies nonzero \( \langle S_i^+ \rangle \) in the thermodynamic limit. The relative phase between spins at different sites is computed using \( \langle S_i^+ \rangle/\langle S_j^+ \rangle = e^{-i(\theta_{ij} - \theta_i)} \).

The \( q = 0 \) pattern corresponds to a relative phase of \( 0, \pm 2\pi/3 \) alternating between the three sublattices. We performed the Monte Carlo calculation on a finite \( 6 \times 6 \) torus with \( n = 4 \). The results are shown in Fig. 2(b), in perfect agreement with the direct computation of spin-spin correlation [20].

Because \( V^\dagger \) is in fact the magnetization operator, we conclude that the in-plane staggered magnetization \( M \sim B^a \) scales with the external magnetic field, where the exponent \( a \) is the scaling dimension of the monopole operator at the Dirac SL fixed point, because \( B \) is dimension one. This exponent is computable by numerical simulations of QED3 or the field theory techniques such as \( 1/N \) expansions. It is known that \( a \propto N \) in the leading order of the \( 1/N \) expansion, where \( N \) is the number of flavors of fermions [22], but the proportionality constant is highly nontrivial. The details on this exponent \( a \) are subject to further study.

So far we show that the FP state is unstable towards the \( S_z \) broken LL state by numerical arguments. In the following we present an analytical argument that the \( S_z \) symmetry is broken by studying the low-energy effective theory. The FP state is characterized by an electronlike Fermi pocket of spin-up spinons \( f_{k\uparrow} \) and a holelike Fermi pocket of spin-down spinons \( f_{k\downarrow} \). After doing a particle-hole transformation on the spin-down Fermi pocket only \( f_{k\downarrow} \rightarrow -h_{k\downarrow} \), we have \( f_{k\uparrow} \) and \( h_{k\downarrow} \) carry same spin but opposite gauge charges. The “Coulomb” attraction between the two particles will cause a pairing instability \( \langle f_{k\uparrow}^\dagger h_{-k\downarrow} \rangle = \langle f_{k\uparrow}^\dagger f_{k\downarrow} \rangle \neq 0 \). This is a triplet excitonic insulator and this has been discussed in the context of graphene [23,24]. Besides Coulomb attraction, there is also an Amperean attraction [25] between the currents of \( f_{k\uparrow}^\dagger \) and \( h_{-k\downarrow} \) excitations. The
“Coulomb” attraction and the Amperean attraction are cooperating for the same condensation. Note that $f_{k_1}^\dagger f_{k_1}$ is a $S_z$ spin-1 object.

Finally, we discuss the consequences of this spontaneous spin ordering in experiment. First, the in-plane $q = 0$ magnetization pattern and its scaling law $M \sim B^\alpha$ are observable by neutron scattering. Second, since the ground state breaks the parity and $S_z$ rotation symmetry, it is separated from the high temperature paramagnetic phase by at least one finite temperature phase transition. This transition can be first order or continuous. If the transition is a continuous one that restores the $S_z$-$U(1)$ symmetry, we expect it to be the Kosterlitz-Thouless universality. Here we simply estimate the transition temperature $T_c \sim \frac{\mu_B B}{k_B}$ when $\mu_B B \leq \chi\rho$. This is because the external magnetic field $B$ is the only energy scale if it is much smaller than the spinon band width $\chi\rho$. Since these are intrinsic properties of the Dirac SL in magnetic field, they can be used to experimentally detect the possible Dirac SL ground state in herbertsmithite and other materials where a Dirac SL may be realized. This XY spin ordering can also serve as a way to detect Dirac SL in numerical studies of the kagomé lattice Heisenberg model such as exact diagonalization.

There are other proposals for the possible nonmagnetic valence bond solid (VBS) ground states in the spin-1/2 Heisenberg model on kagomé lattice [26,27]. If one of those VBS states is the ground state of herbertsmithite, it will surely not break $S_z$ symmetry in a small magnetic field because the VBS phases are fully gapped. The $S_z$ symmetry can be broken at an external magnetic field larger than the spin gap due to the triplon condensation. However, the XY magnetic order generated in this fashion is unlikely to be the $q = 0$ pattern because the VBS orders themselves break translation and we expect their XY orders also break translation and have a large unit cell. Therefore the spontaneous spin ordering we present in this work can be used to differentiate the $U(1)$ Dirac SL from VBS states experimentally.

It is likely that in the current herbertsmithite compound there is an energy scale below which the significant amount of impurities and/or the Dzyaloshinskii-Moriya interaction start to play an important role. Recent experiments estimate the strength of the DM interaction to be $\sim 15$ K [28]. In a strong magnetic field $\sim 30$ T we expect that it is possible to suppress their effects and reveal the intrinsic property of the Dirac SL.

We thank M. Hermele, T. M. Rice, T. Senthil, and F.-C. Zhang for helpful discussions. This research is supported by NSF Grants No. DMR-0706078 and No. DMR-0804040.

[9] In a Schwinger-fermion treatment, the gauge field dynamics entirely comes from integrating out fermion modes. At the mean-field level there are no gauge fluctuations (i.e., no electric field term), the gauge field variables $a_{ij}$, which are the phases of the bond variables $\chi_{ij}$, are treated as complex numbers, and the energy cost of varying $\chi_{ij}$ is completely given by the variation of the fermion band structure.
[20] The relative phase that we computed describes the simultaneous spin ordering we present in this work can be used to differentiate the $U(1)$ Dirac SL from VBS states experimentally.