Algebraic Vortex Liquid in Spin-1/2 Triangular Antiferromagnets: Scenario for Cs_2CuCl_4

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Motivated by inelastic neutron scattering data on Cs_2CuCl_4, we explore spin-1/2 triangular lattice antiferromagnets with both spatial and easy-plane exchange anisotropies, the latter due to an observed Dzyaloshinskii-Moriya interaction. Exploiting a duality mapping followed by a fermionization of the dual vortex degrees of freedom, we find a novel critical spin-liquid phase described in terms of Dirac fermions with an emergent global SU(4) symmetry minimally coupled to a noncompact U(1) gauge field. This ‘‘algebraic vortex liquid’’ supports gapless spin excitations and universal power-law correlations in the dynamical spin structure factor which are consistent with those observed in Cs_2CuCl_4. We suggest future neutron scattering experiments that should help distinguish between the algebraic vortex liquid and other spin liquids and quantum critical points previously proposed in the context of Cs_2CuCl_4.

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The search for two-dimensional (2D) spin liquids has been one of the most tantalizing pursuits in condensed matter physics. Among the most promising systems for realizing such states is the spin-1/2 triangular antiferromagnet, as both the low spin and geometric frustration suppress magnetic ordering. This was appreciated over three decades ago by Anderson, who postulated a quantum-disordered ‘‘resonating-valence-bond’’ (RVB) ground state in the spin-1/2 Heisenberg triangular antiferromagnet [1]. Anderson's RVB concept matured in the high-T_c era, with the triangular lattice often center stage. Using slave bosons, Sachdev explored an Sp(N) generalization of the Heisenberg antiferromagnet and obtained a quantum-disordered ground state, the Z_2 spin liquid, which breaks no symmetries [2]. More recently, Moessner and Sondhi realized a Z_2 spin liquid in a quantum dimer model on the triangular lattice [3]. By exploiting a correspondence between the triangular antiferromagnet and hard-core bosons in a magnetic field, Kalmeyer and Laughlin introduced a ‘‘chiral’’ spin liquid [4] which violates time-reversal symmetry. Both the Z_2 and chiral spin liquids admit gapped, fractionalized $s = 1/2$ excitations—*spinons*—which are bosonic in the former case and ‘‘semionic’’ in the latter.

In spite of these theoretical advances, experimental spin-liquid candidates have only recently appeared. One promising material is the spin-1/2 triangular antiferromagnet Cs_2CuCl_4, which has anisotropic exchange energies $J = 4.3$ K and $J' = 0.34J$ (see Fig. 1). Although Cs_2CuCl_4 develops long-range spiral order below the Néel temperature $T_N = 0.62$ K [5], unusual features reminiscent of spin-liquid physics are manifested in its *dynamics*, probed via inelastic neutron scattering [5,6]. Most notably, in addition to the sharp low-energy spin-wave peaks observed in the ordered phase, neutron scans at higher energies reveal ‘‘critical’’ power laws in the dynamical structure factor. This enhanced scattering persists in a range of temperatures above T_N, where the magnons are absent, and is suggestive of spinon deconfinement characteristic of spin liquids.

This remarkable behavior has attracted much theoretical interest in Cs_2CuCl_4, and several scenarios for the origin of the anomalous scattering have been proposed. Spin-wave theory [7–9] and series expansion studies [10,11] have yielded an important quantitative connection with experiment. Quasi-1D effects have been explored by approaching the triangular lattice by coupling 1D chains [12,13]. Sachdev's slave boson approach was generalized to the anisotropic triangular antiferromagnet by Chung *et al.* [14], and Isakov *et al.* [15] explored the possibility that the Cs_2CuCl_4 phenomenology may be controlled by a quantum critical point separating the Z_2 spin liquid and the spiral state. Using slave fermions, Zhou and Wen [16] alternatively suggested the presence of a critical algebraic spin liquid.

Here, we pursue a new theoretical approach to the triangular antiferromagnet. We consider an easy-plane XXZ spin-1/2 system reformulated in terms of *fermionized-vortex* degrees of freedom using Chern-Simons flux attach-

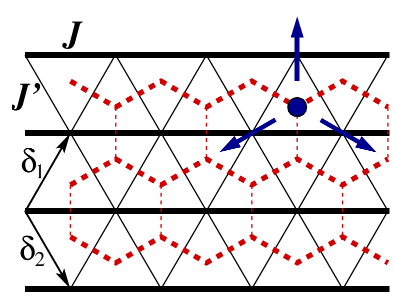

FIG. 1 (color online). Triangular lattice and the dual honeycomb on which vortices reside. Spins shown illustrate a vortex. The dominant spin coupling/vortex hopping occurs along the corresponding bold links. With our convention the vortices are at half filling; e.g., in the spiral state the vortex number is one (zero) on up (down) triangles.

ment. Remarkably, this approach leads naturally to a new critical spin liquid—the "algebraic vortex liquid"—which we explore and then apply to Cs_2CuCl_4.

Algebraic vortex liquid.—Consider an easy-plane spin-$1/2$ antiferromagnet on the anisotropic triangular lattice shown in Fig. 1. We return below to the appropriateness of the easy-plane assumption for Cs_2CuCl_4. We follow closely a dual approach employing *fermionized vortices*, developed for integer-spin systems in Ref. [17]. Implementing the standard duality mapping, one obtains a theory for interacting bosonic vortices on the dual honeycomb lattice (see Fig. 1). A crucial feature is that the vortices are at *half filling* due to the spin frustration. Moreover, because S^z is a half-integer, the vortices "see" an average background of π flux through each hexagon. In terms of a vortex number operator $N_{\mathbf{x}}$ and its conjugate phase $\theta_{\mathbf{x}}$, the vortex Hamiltonian is [17]

$$\mathcal{H}_{\rm dual} = -\sum_{\langle \mathbf{x}\mathbf{x}'\rangle} t_{\mathbf{x}\mathbf{x}'} \cos(\theta_{\mathbf{x}} - \theta_{\mathbf{x}'} - a_{\mathbf{x}\mathbf{x}'} - a^0_{\mathbf{x}\mathbf{x}'}) + \sum_{\mathbf{x}\mathbf{x}'} (N_{\mathbf{x}} - 1/2) V_{\mathbf{x}\mathbf{x}'} (N_{\mathbf{x}'} - 1/2) + \mathcal{H}_a. \quad (1)$$

Here $V_{\mathbf{x}\mathbf{x}'}$ encodes a logarithmic vortex repulsion; $a^0_{\mathbf{x}\mathbf{x}'}$ is a static field producing π flux per hexagon; and $\mathcal{H}_a$ describes the dynamics of the dual gauge field $a_{\mathbf{x}\mathbf{x}'}$ residing on honeycomb links. The S^z spin component appears here as a dual gauge flux, $S^z_{\mathbf{r}} \sim (\Delta \times a)_{\mathbf{r}}/(2\pi)$. The vortex hopping amplitudes $t_{\mathbf{x}\mathbf{x}'}$ are anisotropic to reflect the lattice anisotropy of the exchanges.

Since the vortices are at half filling, the dual theory appears as intractable as the original spin model. One can, however, make significant progress by *fermionizing* the vortices. While similar approaches employing fermionized spins have been pursued [18], our treatment is appealing because vortex density fluctuations are greatly suppressed by their logarithmic repulsion. Following Ref. [17], we perform flux attachment to convert the bosonic vortices into fermions coupled to a Chern-Simons gauge field. Taking a "flux-smeared" mean-field state, the Chern-Simons flux averages to 2π per hexagon, which can be ignored on the lattice. Thus, the mean-field Hamiltonian describes half-filled fermionic vortices hopping on the honeycomb in a background of π flux (due to $a^0_{\mathbf{x}\mathbf{x}'}$). The corresponding band structure reveals four gapless Dirac points. Focusing on low-energy excitations in the vicinity of the four nodes and including gauge fields, we obtain the Euclidean Lagrangian density

$$\mathcal{L} = \bar{\psi}_\alpha(\not\partial - i\not a - i\not A)\psi_\alpha + \frac{1}{2e^2}(\epsilon_{\mu\nu\lambda}\partial_\nu a_\lambda)^2 + \frac{i}{4\pi}\epsilon_{\mu\nu\lambda}A_\mu\partial_\nu A_\lambda + \mathcal{L}_{4f}. \quad (2)$$

Equation (2) describes four flavors of two-component Dirac fermions ψ_α, $\alpha = 1, \ldots, 4$, minimally coupled to a noncompact U(1) gauge field a_μ and a Chern-Simons field A_μ. The gauge field a_μ mediates the logarithmic vortex repulsion, while the Chern-Simons terms implement the flux attachment. Here and below, the nodal velocity anisotropies are absorbed in the scaling of coordinates, while the Maxwell term is only schematic. Symmetries of the spin model preclude all possible mass terms in Eq. (2).

The term $\mathcal{L}_{4f}$ represents symmetry-allowed four-fermion terms arising from short-range vortex interactions. If sufficiently strong, such terms can drive spontaneous fermion mass generation, leading to various symmetry-broken states [19]. For example, giving the fermions a mass by introducing a staggered chemical potential on the honeycomb induces a vortex density wave corresponding to the spiral spin-ordered state. On the other hand, the state with a spontaneously generated $\bar{\psi}_\alpha\psi_\alpha$ mass corresponds to the Kalmeyer-Laughlin chiral spin liquid.

In addition to broken-symmetry states, Eq. (2) allows us to access a new critical spin liquid. To this end, we rewrite the Lagrangian in terms of $\tilde{a}_\mu \equiv a_\mu + A_\mu$ and integrate out the Chern-Simons field to obtain $\mathcal{L} = \mathcal{L}_{\rm QED3} + O(\partial^3\tilde{a}^2)$, with

$$\mathcal{L}_{\rm QED3} = \bar{\psi}_\alpha(\not\partial - i\not{\tilde{a}})\psi_\alpha + \frac{1}{2e^2}(\epsilon_{\mu\nu\lambda}\partial_\nu\tilde{a}_\lambda)^2 + \mathcal{L}_{4f}. \quad (3)$$

Remarkably, up to higher-derivative terms which are henceforth dropped, we arrive at noncompact quantum electrodynamics in $2+1$ dimensions (QED3), with $N=4$ flavors of two-component Dirac fermions [20]. Physically, vortex density fluctuations are suppressed so strongly by interactions that exchange statistics play only a minor role.

QED3 has been widely studied (e.g., see references in [17]) and in the large-N limit realizes a nontrivial stable critical phase. For $N < N_c$, with some unknown N_c, it is believed that four-fermion terms become relevant and spontaneously generate fermion masses, destroying criticality (except at fine-tuned critical points). Here, we proceed with the assumption that $N_c < 4$, implying the presence of a stable critical phase for our dual fermionized-vortex theory. We now explore some of the properties of this algebraic vortex liquid (AVL).

Critical spin correlations in the AVL.—The AVL respects all symmetries of the microscopic spin system, exhibiting no magnetic or other types of order. Since the Dirac fermions are gapless, power-law spin correlations are expected. Consider first the in-plane spin components. The spin raising operator $S^+_{\mathbf{r}}$ adds $S^z = 1$ and hence 2π dual gauge flux. Near this flux insertion the fermionic Hamiltonian has four zero-energy modes, one for each flavor. Physical (gauge-invariant) states are obtained by occupying two of these, so there are six distinct such "monopole insertions." Following the procedure of Ref. [17], we determine the momenta carried by the monopoles: two occur at the spiral ordering wave vectors $\pm\mathbf{Q}$ and three occur at the midpoints $\mathbf{M}_{1,2,3}$ of the Brillouin zone (BZ) edges (see Fig. 2). Numerical diagonalization suggests that the sixth monopole, which carries zero momentum, does not have the same symmetry as S^+ [19], and thus by itself will not contribute to the in-plane spin correlations.

Monopole insertions are known to have nontrivial power-law correlations in large-N QED3. Their scaling dimension, Δ_m, can be estimated from the leading large-N result [21], $2\Delta_m \approx 0.53N$. Extrapolating to $N = 4$ yields $\eta_m \equiv 2\Delta_m - 1 \approx 1.12$. Since all monopoles have the same scaling dimension, S^+ is expected to exhibit the same universal power-law correlations at all five momenta $\mathbf{K}_j$ shown in Fig. 2. Specifically, near each $\mathbf{K}_j$, the dynamic spin structure factor is predicted to scale as

$$S^{+-}(\mathbf{k} = \mathbf{K}_j + \mathbf{q}, \omega) = A_{\mathbf{K}_j} \frac{\Theta(\omega^2 - \mathbf{q}^2)}{(\omega^2 - \mathbf{q}^2)^{1-\eta_m/2}}. \quad (4)$$

The amplitudes $A_{\mathbf{K}_j}$ are sensitive to short-distance physics and can differ significantly among the five wave vectors. In particular, with $J \gg J'$ the amplitude at wave vector $\mathbf{M_3}$ with $k_x = 0$ is expected to be much suppressed compared to the other four momenta, as the latter are near $k_x = \pi$ where the dominant antiferromagnetic correlations occur along the nearly decoupled chains.

The S^z spin correlation behaves rather differently. At zero momentum, the correlation is that of the conserved dual gauge flux. However, a more prominent power law occurs at the spiral ordering wave vectors $\pm\mathbf{Q}$. These arise because an expression for S^z in terms of continuum fields allows a term $e^{i\mathbf{Q}\cdot\mathbf{r}}\bar{\psi}_\alpha W_{\alpha\beta}\psi_\beta$ + H.c. with a fermionic bilinear $\bar{\psi}\hat{W}\psi$ whose correlation is enhanced by gauge field fluctuations [19]. The $S^{zz}(\mathbf{k}, \omega)$ dynamical spin structure factor near $\pm\mathbf{Q}$ has a form similar to Eq. (4), except the corresponding anomalous dimension is that of an enhanced fermionic bilinear in QED3. Using the $1/N$ result [22], $\eta_{enh} \approx 3 - 128/(3\pi^2 N)$, we estimate $\eta_{enh} \approx 1.92$. At other wave vectors the dynamical correlation $S^{zz}(\mathbf{k}, \omega)$ exhibits subdominant power laws.

Cs_2CuCl_4 *Hamiltonian.*—To address the possible experimental relevance of the AVL phase, we now consider the measured Cs_2CuCl_4 Hamiltonian. Besides the Heisenberg exchange with $J = 4.3$ K, $J' = 0.34J$, there is also a Dzyaloshinskii-Moriya (DM) interaction:

$$H_{DM} = -\sum_{\mathbf{r}} \mathbf{D} \cdot \mathbf{S}_\mathbf{r} \times (\mathbf{S}_{\mathbf{r}+\delta_1} + \mathbf{S}_{\mathbf{r}+\delta_2}). \quad (5)$$

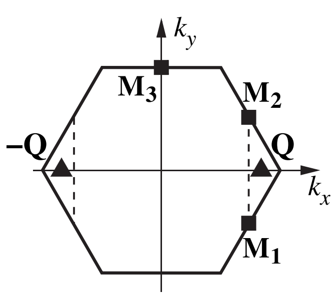

FIG. 2. Momenta in the triangular BZ at which S^{+-} exhibits dominant power-law correlations in the AVL. We predict the *same power law* at all five momenta. For S^{zz}, the dominant scattering is at $\pm\mathbf{Q}$ and has a faster-decaying power law. Dashed lines show $k_x = \pm\pi$ characteristic of a 1D system.

The vectors $\delta_{1,2}$ connect spins on neighboring chains as in Fig. 1, and $\mathbf{D} = D\hat{\mathbf{z}}$ is oriented perpendicular to the triangular layers, with $D = 0.053J$. Although there is a small interlayer coupling [8], we focus on the 2D system.

Significantly, the DM term provides an easy-plane anisotropy, breaking the SU(2) spin symmetry of the Heisenberg exchange down to U(1), and also violates inversion symmetry $\mathbf{r} \rightarrow -\mathbf{r}$. Thus for $T < T_N = 0.62$ K the DM term determines both the spin ordering plane and the sign of the ordering wave vector, which along with D changes sign from one layer to the next. Despite the small value of D, the easy-plane anisotropy is amplified since the DM interaction is not frustrated near the dominant antiferromagnetic wave vector along the chains (whereas the J' coupling is frustrated). To quantify this, we briefly consider a *classical* 2D spin system with Cs_2CuCl_4 parameters. Without the DM term, a $J - J'$ Heisenberg spin system remains disordered at all temperatures. With DM coupling the system has only U(1) spin symmetry and thus exhibits a low-temperature phase with quasi-long-range order (QLRO). We performed a classical Monte Carlo study and found this transition at $T_c \approx 0.27JS^2$. Taking $S^2 = 3/4$ appropriate for spin-1/2, we estimate that a single layer would obtain QLRO below $T_c = 0.87$ K. Once each layer has QLRO, an arbitrarily small interlayer coupling would induce 3D long-range order, even though the interlayer coupling is frustrated in Cs_2CuCl_4 due to the alternating sign of D [8]. This suggests that the observed spiral ordering at T_N in Cs_2CuCl_4 is primarily driven by the easy-plane character of spins in each layer. As the classical treatment neglects quantum fluctuations, it is reasonable that the estimated T_c somewhat exceeds T_N. These considerations suggest that vortices acquire integrity as degrees of freedom in Cs_2CuCl_4.

Scenarios for AVL in Cs_2CuCl_4.—To specifically address the applicability of the AVL to Cs_2CuCl_4, we consider the effect of the DM term on the AVL. In the fermionized-vortex Lagrangian (2), the DM interaction appears as an inversion-breaking fermion mass term corresponding to a staggered vortex chemical potential, with bare mass $m \sim D$. This mass drives the system to the spiral state as indicated by the vertical flow in Fig. 3. Thus, with the DM interaction the observed spiral ground state of Cs_2CuCl_4 emerges naturally out of the AVL.

Since the DM term induces an easy-plane spin anisotropy *and* breaks inversion symmetry, applicability of the AVL to Cs_2CuCl_4 requires a delicate balance. For the AVL to apply on intermediate energy scales, the DM interaction must first produce sufficient easy-plane anisotropy for the description in terms of vortices to be appropriate, before destabilizing the AVL state toward the spiral order. This is scenario 1 in the schematic flow diagram of Fig. 3. The alternative scenario 2 does not approach the easy-plane fixed point but is driven directly to the magnetic order. In the latter case, intermediate energy scales may be governed by an (unknown) SU(2)-invariant criticality indicated with

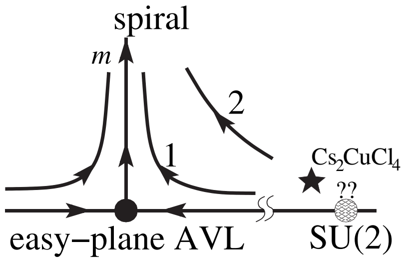

FIG. 3. Schematic renormalization group flows near the AVL fixed point in the presence of the DM interaction. The vertical axis is the inversion-breaking fermion mass m arising from the DM term. The bare Cs_2CuCl_4 Hamiltonian is indicated with the star. For flow trajectory 1, the dynamics at intermediate energy scales is governed by the AVL theory. For trajectory 2, intermediate energy scales are controlled by an (unknown) SU(2)-invariant criticality.

question marks in the figure. Below, we pursue the consequences of scenario 1.

Comparison with experiments.—The spin dynamics in Cs_2CuCl_4 was measured in neutron scattering experiments by Coldea *et al.* [5,6]. For $T < T_N$, Cs_2CuCl_4 has well-defined spin waves, gapless near zero momentum but observed with a small gap near the ordering wave vectors $\pm\mathbf{Q}$, presumably due to weak violations of SU(2) spin symmetry by the DM term. Experiments also see fairly small spin-wave gaps at momenta $\mathbf{M_{1,2}}$ of Fig. 2. Broad continuum scattering is observed above the spin-wave gaps near momenta $\pm\mathbf{Q}$ and $\mathbf{M_{1,2}}$, and persists even for $T > T_N$. Notably, Ref. [6] reports power-law line shapes in scans near these momenta (scans J and G in Ref. [6]), each with the *same* exponent, $\eta_{\rm exp} = 0.74$. The AVL also admits gapless spin-1 excitations with power-law scaling at these momenta, with the leading estimate of $\eta_{\rm m} = 1.12$ somewhat larger than experiment. These momentum space locations of enhanced scattering are determined by physics on the scale of several lattice spacings. The AVL theory captures this aspect of the Cs_2CuCl_4 phenomenology rather well.

Since the above data are near $k_x = \pi$, which is the dominant wave vector in the quasi-1D limit, some caution is necessary. With $J \approx 3J'$ strong contributions on intermediate energy scales from antiferromagnetic correlations along the chains are not unexpected. A measurement midway between points $\mathbf{Q}$ and $\mathbf{M_1}$ would help determine the transverse k_y dependence of the continuum scattering and clarify whether it is meaningful to speak of enhanced scattering near discrete momenta in the 2D BZ. The AVL also predicts enhanced $S^{+-}(\mathbf{k}, \omega)$ near $\mathbf{M_3}$, albeit with a smaller amplitude. Being less influenced by quasi-1D effects, further measurements near $\mathbf{M}_3$ would be useful. Polarized neutron experiments to search for the easy-plane character of the AVL would also be interesting.

Competing 2D theoretical proposals with full SU(2) spin symmetry include an algebraic spin liquid ("U1C") proposed by Zhou and Wen [16] and the quantum critical point scenario of Isakov *et al.* [15]. Each makes distinct predictions for the momenta of the low-energy spin-1 excitations, which also differ from the AVL. Such characterizations can in principle be used to discriminate among different theories, although the limited energy window for observing the continua and the material's strong anisotropy are unavoidable complications. Further experimental and theoretical studies should help clarify the true "spin-liquid" nature of this interesting material.

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