Algebraic vortex liquid in spin-1/2 triangular antiferromagnets: Scenario for Cs$_2$CuCl$_4$

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Motivated by inelastic neutron scattering data on Cs$_2$CuCl$_4$, 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 non-compact 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$_2$CuCl$_4$. 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$_2$CuCl$_4$.

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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. 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. More recently, Moessner and Sondhi realized a $Z_2$ spin liquid in a quantum dimer model on the triangular lattice. By exploiting a correspondence between the triangular antiferromagnet and hard-core bosons in a magnetic field, Kalmeyer and Laughlin introduced a “chiral” spin liquid 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 and “semionic” in the latter.

In spite of these theoretical advances, experimental spin-liquid candidates have only recently appeared. One promising system is the spin-1/2 triangular antiferromagnet Cs$_2$CuCl$_4$, which has anisotropic exchange energies $J = 4.3$K and $J' = 0.34$K (see Fig. 1). Although this material develops long-range spiral order below the Neel temperature $T_N = 0.62$K, unusual features reminiscent of spin-liquid physics are manifested in its dynamics, probed via inelastic neutron scattering. 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$_2$CuCl$_4$, and several scenarios for the origin of the anomalous scattering have been proposed. Spin-wave theory and series expansion studies have yielded important quantitative connection with experiment. Quasi-1D effects have been explored by approaching the triangular lattice by coupling 1D chains. Sachdev’s slave boson approach was generalized to the anisotropic triangular antiferromagnet by Chung et al., and Isakov et al. explored the possibility that the Cs$_2$CuCl$_4$ 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 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 attachment. Remarkably, this approach leads naturally to a new “critical” spin liquid—the “algebraic vortex liquid”—which we explore and then apply to Cs$_2$CuCl$_4$.

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$_2$CuCl$_4$. 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 $\pi$ flux through each hexagon. In terms of a vortex number operator $N_x$ and its conjugate phase $\theta_x$, the vortex Hamiltonian is

$$\mathcal{H}_{\text{dual}} = -\sum_{\langle xx' \rangle} t_{xx'} \cos(\theta_x - \theta_{xx'} - \theta_{xx'}^0)$$

$$+ \sum_{xx'} (N_x - 1/2)V_{xx'}(N_{xx'} - 1/2) + \mathcal{H}_a. \quad (1)$$
Here $V_{\alpha\alpha'}$ encodes a logarithmic vortex repulsion; $a_{\alpha\alpha'}^0$ is a static field satisfying $(\Delta \times a^0)^r = \pi$, where $(\Delta \times a^0)^r$ is a lattice curl around the hexagon encircling triangular lattice site $r$; and $H_a$ describes the dynamics of the dual gauge field $a_{xx'}$ residing on honeycomb links. The $S^2$ spin component appears here as a dual gauge flux, $S_{S}^2 \sim (\Delta \times a^0)^r/(2\pi)$. The vortex hopping amplitudes $t_{xx'}$ are anisotropic to reflect the lattice anisotropy of the exchanges. Thus, vortices hop more easily across weak spin links $J'$ than across strong links $J$.

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 suppressed so strongly by interactions that exchange statistics play only a minor role.

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 $S^+_{S}$ adds $S^2 = 1$ and hence $2\pi$ 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 $\pm Q$ and three occur at the midpoints $M_{1,2,3}$ 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 $S^x$([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 the large-$N$ QED3 theory. The scaling dimension, $\Delta_m$, of the monopoles can be estimated from the leading large-$N$ result([20], $2\Delta_m \approx 0.53N$). A naive extrapolation to $N = 4$ yields $\eta_m \equiv 2\Delta_m - 1 \approx 1.12$. Since all monopoles have the same scaling dimension, $S^z$ is expected 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 $L = L_{QED3} + O(\partial^3 \tilde{A}^2)$, with

$$L_{QED3} = \bar{\psi}_{\alpha}(\tilde{\gamma} - i\tilde{A})\psi_{\alpha} + \frac{1}{2\pi^2}(\epsilon_{\mu\nu\lambda}\partial_\nu \tilde{a}_\lambda)^2 + L_{4f}. \quad (3)$$

Remarkably, up to higher-derivative terms which are henceforth dropped, we arrive at non-compact quantum electrodynamics in 2+1 dimensions (QED3), with $N = 4$ flavors of two-component Dirac fermions([22], 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 refs. 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).
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

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

(4)

The amplitudes $A_{\mathbf{K}_j}$ are sensitive to short-distance physics and can differ significantly among the five wave vectors, particularly in the anisotropic system. 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 $e^{iQr^z}W_{\alpha\beta}\psi^\beta$ with a fermionic bilinear $\overline{\psi}W\psi$ whose correlation is enhanced by gauge field fluctuations. Here, $W$ can be obtained by considering a perturbation to the fermionic hopping Hamiltonian that adds static gauge flux through the layers, with bare mass $m$.

The vectors $\delta_{1,2}$ connect spins on neighboring chains as in Fig. 1 and $D = D\hat{z}$ is oriented perpendicular to the triangular layers, with $D = 0.053J$. Although there is a small interlayer coupling, 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 $T < T_N = 0.62K$ the DM term determines both the ordering plane, which coincides with the triangular layers, 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 $\text{Cs}_2\text{CuCl}_4$ 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 $T_c \approx 0.27 J S^2$. A simple classical ground state analysis indicates that about half of the “phase stiffness” originates from the DM coupling itself. Taking $S^2 = 3/4$ appropriate for spin-1/2, we estimate that a single layer would obtain QLRO below $T_c = 0.84K$. Once each layer has QLRO, an arbitrarily small interlayer coupling would induce 3D long-range order, even though the interlayer coupling is frustrated in $\text{Cs}_2\text{CuCl}_4$ due to the alternating sign of $D$. This suggests that the observed spiral ordering at $T_N$ in $\text{Cs}_2\text{CuCl}_4$ 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, required to drive the 2D ordering, acquire integrity as degrees of freedom in $\text{Cs}_2\text{CuCl}_4$.

**Scenarios for AVL in $\text{Cs}_2\text{CuCl}_4$.** To specifically address the applicability of the AVL to $\text{Cs}_2\text{CuCl}_4$, 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 with chirality dictated by the sign of $D$. Thus, with the DM interaction the observed spiral ground state of $\text{Cs}_2\text{CuCl}_4$ emerges naturally out of the critical AVL.

Since the DM term induces an easy-plane spin anisotropy and breaks inversion symmetry, applicability of the AVL to $\text{Cs}_2\text{CuCl}_4$ 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 question marks in the figure. Below, we pursue the consequences of scenario 1.

Comparison with experiments. — The spin dynamics in Cs$_2$CuCl$_4$ was measured in neutron scattering experiments by Coldea et al.\cite{6} For $T > T_N$, Cs$_2$CuCl$_4$ has well-defined spin waves, gapless near zero momentum and observed with a small gap near the ordering wave vectors $\pm Q$, presumably due to weak violations of SU(2) symmetry by the DM term. Experiments also see fairly small spin-wave gaps at momenta $M_{1,2}$ of Fig. 2. (Series expansion studies of the Heisenberg model show that the gaps near these momenta and also $M_3$ deviate strongly from spin-wave theory.\cite{14}) Broad continuum scattering is observed above the spin-wave gaps near momenta $\pm Q$ and $M_{1,2}$, 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, $\eta_{\text{exp}} = 0.74$. The AVL also admits gapless spin-1 excitations with power-law scaling at these momenta, with the leading estimate of $\eta_{\text{AVL}} = 1.12$ somewhat larger than experiment. These momentum space locations of enhanced scattering are determined by physics on the scale of several lattice spacings, and provide important constraints on theory. The AVL theory captures this aspect of the Cs$_2$CuCl$_4$ 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. Specifically, with $J \approx 3J'$ strong contributions on intermediate energy scales from antiferromagnetic correlations along the chains are not expected. A measurement midway between points $Q$ and $M_1$ 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 $s^{--}(k, \omega)$ near $M_3$, albeit with a smaller amplitude. Being less influenced by quasi-1D effects, further measurements near $M_3$ would be useful. Polarized neutron experiments to search for the distinct easy-plane character of the AVL would also be very interesting.

Competing 2D theoretical proposals with full SU(2) spin symmetry include an algebraic spin liquid ("U1C") proposed by Zhou and Wen\cite{16} and the quantum critical point (QCP) scenario of Isakov et al.\cite{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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