Critical spin liquid at $\frac{1}{3}$ magnetization in a spin-$\frac{1}{2}$ triangular antiferromagnet

Jason Alicea[1] and Matthew P. A. Fisher[2]
[1]*Physics Department, University of California, Santa Barbara, California 93106, USA*
[2]*Kavli Institute for Theoretical Physics, University of California, Santa Barbara, California 93106, USA*
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Although magnetically ordered at low temperatures, the spin-$\frac{1}{2}$ triangular antiferromagnet Cs_2CuCl_4 exhibits remarkable spin dynamics that strongly suggest proximity to a spin-liquid phase. Here we ask whether a proximate spin liquid may also occur in an applied magnetic field, leaving a similar imprint on the dynamical spin correlations of this material. Specifically, we explore a spatially anisotropic Heisenberg spin-$\frac{1}{2}$ triangular antiferromagnet at $\frac{1}{3}$ magnetization from a dual vortex perspective, and indeed find a "critical" spin-liquid phase described by quantum electrodynamics in (2+1)-dimensions with an emergent SU(6) symmetry. A number of nontrivial predictions follow for the dynamical spin structure factor in this "algebraic vortex liquid" phase, which can be tested via inelastic neutron scattering. We also discuss how well-studied "up-up-down" magnetization plateaus can be captured within our approach, and further predict the existence of a stable gapless solid phase in a weakly ordered up-up-down state. Finally, we predict several anomalous "roton" minima in the excitation spectrum in the regime of lattice anisotropy where the canted Néel state appears.

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The quest for an unambiguous experimental realization of a quantum spin liquid remains a central pursuit in condensed matter physics, despite a long history dating back to Anderson's suggestion that the nearest-neighbor Heisenberg triangular antiferromagnet may realize a "resonating valence bond" ground state.[1] Quite generally, the theoretical search for models realizing such exotic quantum ground states has focused primarily on frustrated magnets in zero magnetic field. The central goal of this paper is to take a first step at analyzing the situation when a finite magnetic field is present and ask to what extent spin liquids may occur in this broader setting. Naively speaking, this may seem somewhat misguided, since sufficiently strong magnetic fields quench quantum fluctuations entirely and lead to a simple ferromagnetically ordered ground state. However, at moderate-strength fields, it is conceivable that the presence of numerous competing phases arising from the geometric frustration may lead rather to an *enhanced* role of quantum fluctuations by the field.

The spin-$\frac{1}{2}$ triangular antiferromagnet provides a simple and experimentally relevant test case for these ideas. One noteworthy example is the spatially anisotropic material Cs_2CuCl_4,[2–4] which has garnered much attention as a promising experimental realization of a spin liquid at zero magnetic field. Though Cs_2CuCl_4 exhibits long-range spiral order at the lowest temperatures, inelastic neutron-scattering experiments reveal broad regions of continuum scattering at intermediate energies throughout the Brillouin zone.[4] This anomalous scattering coexists with well-defined spin waves in the ordered phase and, significantly, also persists at temperatures above the Néel temperature where the spin waves are absent. The origin of this continuum scattering presents one of the foremost challenges for understanding the behavior of this interesting material. While two groups find that nonlinear spin-wave theory can account for much of the observed weight in the ordered phase,[5,6] spin-liquid physics has been widely invoked as a possible explanation for the continuum.[7–14]

Low-temperature magnetic order develops in the presence of a magnetic field as well, leading to a rich phase diagram.[15] Motivated in part by the observed zero-field phenomenology, we will explore the following question here. Can spin-liquid phases appear at nonzero magnetic field, which influence the dynamics of Cs_2CuCl_4 at intermediate energies, just as it appears to be the case in the absence of a field? Several experimental features[15] that make this scenario plausible are worth noting. First, a broad temperature range characterized by short-range order persists up to sizable fields of around 6 T. Second, the ordering temperature initially *decreases* with the magnetic-field strength, thereby broadening this short-range order regime. Finally, there is a stark contrast in the experimentally determined phase diagrams for moderate fields applied along the *b* and *c* axes in the plane of the triangular layers, implying a high sensitivity of ordering to small perturbations. These features together strongly point to the presence of many nearly degenerate states and a corresponding enhancement of quantum fluctuations by intermediate-strength fields.

To explore possible field-induced spin-liquid phases relevant for Cs_2CuCl_4, we study an anisotropic Heisenberg spin-$\frac{1}{2}$ triangular antiferromagnet tuned to $\frac{1}{3}$ magnetization using a well-studied duality mapping.[16] The present paper significantly extends an earlier study of a (easy-plane) spin model in the absence of a magnetic field.[11] At zero field, it was argued that reformulating the spin model in terms of *fermionized* vortices leads naturally to a spin liquid of the "critical" (versus topological) variety. This algebraic vortex liquid (AVL) is a promising candidate for explaining the zero-field Cs_2CuCl_4 phenomenology. As we describe below, generalizing to the case of $\frac{1}{3}$ magnetization again leads naturally to a critical AVL phase, which like its zero-field predecessor supports gapless spin excitations and power-law spin correlations. The different AVL characterized here is described by quantum electrodynamics in (2+1)-dimensions (QED3) with an emergent SU(6) symmetry, which has important implications for the spin dynamics. Specifically, it

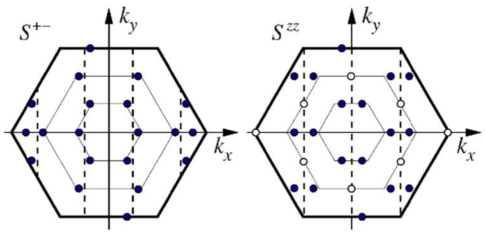

FIG. 1. (Color online) Filled circles indicate momenta at which the components of the spin structure perpendicular to the field (S^{+-}) and along the field (S^{zz}) exhibit enhanced universal correlations with the same power-law decay in the AVL. In the canted Néel phase, anomalous "roton" minima in the excitation spectrum are predicted at the momenta denoted by open circles.

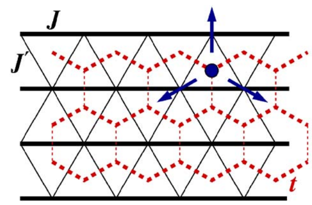

FIG. 2. (Color online) Triangular lattice and the dual honeycomb on which vortices reside. Spins shown illustrate a vortex.

follows that both the components of the dynamical spin structure factor along the field and perpendicular to it exhibit enhanced universal correlations with *identical* power laws at various momenta in the Brillouin zone (i.e., the filled circles in Fig. 1). Such anomalous scattering would be quite interesting to search for in Cs_2CuCl_4 via inelastic neutron scattering. On a technical level, we emphasize two appealing features of this AVL. First, the field explicitly breaks the SU(2) spin symmetry down to U(1), so that our dual description is valid even in the absence of a Dzyaloshinskii-Moriya interaction, thereby avoiding subtleties encountered at zero field. Second, the QED3 theory describing this AVL is "larger-N" than in the zero-field case, and thus, even more likely to exist as a stable phase.

We also discuss the so-called up-up-down (UUD) $\frac{1}{3}$ magnetization plateaus from our dual perspective. Such UUD states have been well studied for the isotropic triangular antiferromagnet using spin-wave theory[17] and exact diagonalization,[18] and have recently been observed in the anisotropic material Cs_2CuBr_4.[19] We show how the $\frac{1}{3}$ magnetization plateau can be captured within our approach when the sublattice magnetization is near full polarization, and further predict the presence of a critical solid when the sublattice magnetization is weak. Signatures of this gapless solid phase may be accessible in exact diagonalization and/or series expansion studies by examining the excitation spectrum in an *XXZ* model with spin-space anisotropy tuned near the transition to the UUD plateau.

Finally, we discuss the range of lattice anisotropy where the ground state is the canted square-lattice Néel phase. In this "frustrated square lattice" limit, we predict several anomalous minima in the excitation spectrum at the momenta indicated by open circles in Fig. 1, which are due to vortex-antivortex "roton" excitations. These excitations can be difficult to capture within spin-wave theory, but appear naturally in our dual framework. Such features were also predicted at zero field, in agreement with earlier series expansions,[20] and would be interesting to probe here as well to (perhaps) further substantiate the vortex interpretation of those results.

Turning to the details, the Hamiltonian we consider is

$$\mathcal{H} = \sum_{\langle \mathbf{r}\mathbf{r}' \rangle} J_{\mathbf{r}\mathbf{r}'} \mathbf{S}_{\mathbf{r}} \cdot \mathbf{S}_{\mathbf{r}'} + h \sum_{\mathbf{r}} S^z_{\mathbf{r}}, \tag{1}$$

where the magnetic field h lies in the triangular planes and the anisotropic exchanges are as shown in Fig. 2. Throughout we assume that the field is tuned so that the system is at $\frac{1}{3}$ magnetization. Furthermore, we will ignore the small interplane and Dzyaloshinskii-Moriya couplings present in Cs_2CuCl_4, which is appropriate for the field orientations of interest.[21] Since the Hamiltonian has U(1) spin symmetry, Eq. (1) can be mapped onto a quantum rotor model and dualized using the standard transformation.[16] Readers interested in the details of this transformation and the analysis to follow are referred to Refs. 12 and 22, where the approach we employ has been well developed in similar settings. Here we will apply this technique to the different physical situation of a $\frac{1}{3}$-magnetized triangular antiferromagnet, highlighting the nontrivial physics it enables us to access but dispensing with the unnecessary formalism developed elsewhere.

The quantum rotor mapping is implemented by replacing $S^+_{\mathbf{r}} \rightarrow e^{i\varphi_{\mathbf{r}}}$ and $S^z_{\mathbf{r}} \rightarrow n_{\mathbf{r}} - \frac{1}{2}$, where $n_{\mathbf{r}}$ is an integer-valued boson number and $\varphi_{\mathbf{r}}$ is the conjugate phase. While not exact, such a transformation is expected to be inconsequential for describing the universal physics, which is our focus. The rotor Hamiltonian can then be expressed as

$$\mathcal{H}' = \sum_{\langle \mathbf{r}\mathbf{r}' \rangle} J_{\mathbf{r}\mathbf{r}'} \cos(\varphi_{\mathbf{r}} - \varphi_{\mathbf{r}'}) + U \sum_{\mathbf{r}} \left(n_{\mathbf{r}} - \tfrac{1}{3}\right)^2 + \sum_{\langle \mathbf{r}\mathbf{r}' \rangle} J_{\mathbf{r}\mathbf{r}'} \left(n_{\mathbf{r}} - \tfrac{1}{3}\right)\left(n_{\mathbf{r}'} - \tfrac{1}{3}\right), \tag{2}$$

where the U term above enforces energetically the constraint of having either 0 or 1 boson per site as appropriate for modeling a spin-$\frac{1}{2}$ system. In the quantum rotor language, the condition of $\frac{1}{3}$ magnetization translates into having, on average, one boson for every three sites, which is manifested in the above Hamiltonian.

The duality mapping applied to Eq. (2) proceeds in an identical fashion as in Refs. 12 and 22; the only difference between these references and the present system is that the bosons are now at a different mean filling. In the dual picture, one equivalently reformulates the rotor model in terms of quantum mechanical, bosonic vortex degrees of freedom, which are topological defects in which the spins wind around triangular plaquettes as shown in Fig. 2. These vortices are mobile, pointlike particles that hop on the dual honeycomb lattice (see Fig. 2) in a background of a fluctuating gauge field $a_{\mathbf{x}\mathbf{x}'}$, whose flux encodes the boson number (or equivalently the S^z component of spin, along the field), $n^z_{\mathbf{r}} \sim (\Delta \times a)_{\mathbf{r}}/(2\pi)$. Thus, the magnetic field manifests itself as a nontrivial background flux "felt" by the vortices, which at $\frac{1}{3}$ magnetization is a commensurate $2\pi/3$ flux per dual hexagon on average. This background flux is where the present study departs from the zero-field analysis of Refs. 11 and 12, where the vortices see π flux, and is responsible for the different physics we obtain here. The vortices interact via a logarithmic repulsion mediated by the gauge field and, importantly, are at half-filling due to the underlying frustration in the original spin model. In terms of a vortex number operator $N_{\mathbf{x}}$ and its conjugate phase $\theta_{\mathbf{x}}$, the dual Hamiltonian takes the form

$$\mathcal{H}_{\text{dual}} = -\sum_{\langle \mathbf{x}\mathbf{x}' \rangle} t_{\mathbf{x}\mathbf{x}'} \cos(\theta_{\mathbf{x}} - \theta_{\mathbf{x}'} - a_{\mathbf{x}\mathbf{x}'}) + \sum_{\mathbf{x}\mathbf{x}'} \left(N_{\mathbf{x}} - \tfrac{1}{2}\right) V_{\mathbf{x}\mathbf{x}'} \left(N_{\mathbf{x}'} - \tfrac{1}{2}\right) + \mathcal{H}_a. \tag{3}$$

The first term allows vortices to hop across nearest-neighbor honeycomb sites. The hopping amplitudes are generally anisotropic since vortices hop more easily across weak spin links; thus we take $t'/t \sim J'/J$ (see Fig. 2). The second term encodes the vortex repulsion, while the last describes the gauge-field dynamics.

The large vortex density poses a significant challenge for analyzing the dual theory as it stands. The strong vortex interactions, however, actually make the problem more tractable, as they strongly suppress vortex density fluctuations and lead to an incompressible vortex fluid. Vortex exchange statistics is therefore of secondary importance, and one can proceed by utilizing a formally exact mapping to convert the bosonic vortices into fermions bound to 2π flux tubes. This statistical flux does not alter the average flux seen by the vortices, since it averages to 2π per hexagon (which is equivalent to zero flux). Furthermore, as argued in detail in Ref. 12, the incompressibility renders the flux attachment *irrelevant* for describing low-energy physics of the vortex fluid. This statistical transmutation at low energies from bosonic to fermionic vortices is at the heart of our approach.

Physically, working with fermions is advantageous because the Pauli principle allows one to first focus on the vortex kinetic energy while still maintaining a good interaction energy. Thus, we initially consider a mean-field state where we “smear” the $2\pi/3$ background flux (due to the $\frac{1}{3}$ magnetization) uniformly across the lattice, though we will relax this assumption later. Fluctuations around this flux-smeared state can be systematically controlled as discussed below. Within this mean field, one can work out the vortex band structure, which captures the important intermediate length-scale physics. In particular, we find that for $t'/t > 2^{1/3}$ the vortices are gapped, while for $t'/t < 2^{1/3}$ they form a critical state with six gapless Dirac points. The former gapped state corresponds to the canted “square lattice” Néel phase expected when J' is dominant, while the latter is the mean-field description of the AVL which will be our main focus.

We discuss the gapless regime first. Expanding around the Dirac points and including gauge fluctuations around the flux-smeared state, one obtains a low-energy effective theory describing six flavors of two-component Dirac fermions ψ_α, $\alpha = 1, \ldots, 6$, which are minimally coupled to a U(1) gauge field a_μ that mediates the vortex repulsion. The low-energy effective theory so obtained is identical to QED3,

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

(Inclusion of the statistical 2π flux tubes merely leads to interactions that are irrelevant by power counting in the above, as claimed earlier.) The first term encodes the linearly dispersing kinetic energy for our six flavors of vortices. The second is the usual Maxwell term, while $\mathcal{L}_{4f}$ represents symmetry-allowed four-fermion interactions. Of central importance is whether the critical nature of the mean-field state survives in the full interacting theory above. A partial answer to this question can be obtained by asking whether any fermion mass terms, which would drive some type of ordering in the original spin model and lead to a gap in the vortex spectrum, are allowed by symmetry. Despite the loss of time-reversal symmetry by the magnetic field, the answer is “no”—the remaining symmetries of the original spin model are still sufficient to preclude all possible mass terms in Eq. (4). This is not the full story, however, since the four-fermion interactions above, if relevant in the renormalization group sense, could still potentially lead to ordering via spontaneous mass generation. Hence to proceed, we must assess the role of these terms in the theory.

In the limit of a large number N of fermion flavors, all such four-fermion interactions are indeed known to be irrelevant, so that QED3 realizes a nontrivial stable critical phase. (See QED3 references in Ref. 12) While the critical value of N above which this holds is uncertain, calculations to leading order in $1/N$ suggest that $N=6$ relevant here is large enough. Hence, we proceed with the assumption that Eq. (4) indeed describes a stable critical phase for our vortices, with an emergent SU(6) symmetry due to the presumed irrelevance of $\mathcal{L}_{4f}$. In terms of the original spin model, this can be qualitatively understood as follows. The presence of numerous gapless Dirac points implies that there are many competing orders in the spin model. With sufficiently many competing orders (i.e., at large enough N), quantum fluctuations can be so strong as to disorder the system even at zero temperature. The resulting critical phase is precisely the AVL, which respects all symmetries of the original spin model and supports gapless vortex excitations and, in turn, gapless spin excitations as we now discuss.

The key experimental prediction for this phase lies in the behavior of the dynamical spin structure factor, since this can be directly probed with inelastic neutron scattering. We first discuss the spin correlations of $S^\pm$, transverse to the field. Recalling that $S^z + \frac{1}{2} \sim (\Delta \times a)/(2\pi)$, since S^+ adds $S^z = 1$, it follows that the corresponding dual operators are “monopoles” which add 2π gauge flux. The added flux gives rise to six additional vortex zero modes, one for each fermion flavor, and half of these must be filled to produce a physical state. Thus there are 20 leading monopole excitations, which can be shown to carry the momenta $\mathbf{\Pi}_j$ displayed on the left side of Fig. 1.[12] Such monopoles exhibit nontrivial power-law correlations, each with identical scaling dimension due to the emergent SU(6) symmetry. Consequently, near each $\mathbf{\Pi}_j$, the transverse spin structure factor scales as

$$\mathcal{S}^{+-}(\mathbf{k}=\mathbf{\Pi}_j+\mathbf{q},\omega)\sim\frac{\Theta(\omega^2-\mathbf{q}^2)}{(\omega^2-\mathbf{q}^2)^{1-\eta_{+-}/2}}. \quad (5)$$

Using the leading large-N result,[23] the anomalous dimension is $\eta_{+-}\approx 0.54N-1\approx 2.2$ for each $\mathbf{\Pi}_j$.

Consider now the correlations of S^z along the field. Near zero momentum S^z appears as the dual gauge flux $\Delta\times a$. But the vortex band structure also allows naturally for "particle-hole" excitations which correspond to vortex currents, and such currents generate modulated gauge flux that contributes to S^z at other wave vectors. These appear in the continuum as fermion bilinears, which provide the leading S^z spin correlations at the 18 momenta denoted by filled circles on the right side of Fig. 1. Near each of these momenta, the structure factor S^{zz} scales as in Eq. (5), but with a larger anomalous dimension $\eta_{zz}\approx 2.3$ estimated from the leading $1/N$ result.[24] Note that the momenta denoted by filled circles in Fig. 1 shift along k_x as the lattice anisotropy changes, lining up along the dashed lines in the one-dimensional limit and residing symmetrically around the hexagons shown in the isotropic limit. We also mention that both η_{+-} and η_{zz} are estimated to be larger than 2, in which case a cusp rather than a divergence occurs in the structure factor. Enhanced scattering should, nevertheless, be observable near the above momenta, and moreover, in Cs_2CuCl_4 such divergences would be cut off at the lowest energies due to the onset of magnetic order.

We now shift our attention to $\frac{1}{3}$ magnetization plateaus, focusing for simplicity on the isotropic limit $J'=J$. Returning to the "flux-smeared" mean-field level, we now weakly modulate the gauge flux away from $2\pi/3$ such that UUD order is assumed at the outset. Introducing this flux modulation, surprisingly, merely shifts the locations of the Dirac nodes. Furthermore, the remaining symmetries of the spin model are *still* sufficient to protect the gaplessness of this state against small perturbations. That is, the critical nature of the AVL is preserved upon introducing weak UUD order, implying the interesting possibility of having a stable *gapless* solid phase.

Where, then, is the gapped magnetization *plateau*? Such a spin state is described by a $\nu=1$ quantum Hall state for the fermionic vortices. To see this, note that the first quantized wave function for the bosonic vortices is $\Psi_{\text{bos}}=\prod_{i<j}e^{-i\Theta(\mathbf{x}_i-\mathbf{x}_j)}\mathbf{\Psi}_{\text{ferm}}$, where $\Theta(\mathbf{x})$ denotes the angle formed by the vector $\mathbf{x}$ with respect to a fixed axis and Ψ_{ferm} is the fermionic vortex wave function. With Ψ_{ferm} the usual $\nu=1$ wave function, it has been shown[25] that Ψ_{bos} exhibits (quasi) off-diagonal long-range order. Hence, the bosonic vortices form a "superfluid," which corresponds to an "insulating" state for the original spin model. Indeed, due to the accompanying dual "Meissner effect," the gauge field $a_{\mathbf{xx}'}$ picks up a Higgs mass, and there are thus no gapless excitations in this phase. This is the desired UUD plateau as claimed. Such a $\nu=1$ quantum Hall state can be driven by adding an imaginary second neighbor vortex hopping with strength t_2 larger than a critical value t_{2c}, which decreases as the sublattice magnetization increases toward full polarization. This can be explicitly demonstrated by computing the Chern numbers for the occupied bands.[26] Second neighbor hoppings with $t_2 < t_{2c}$ lead to symmetry breaking, and are thus precluded in accordance with the above discussion.

Finally, let us discuss the frustrated square lattice limit where J' is dominant and the vortices are gapped. Again, this regime corresponds to the expected canted Néel phase. Consider the correlations of S^z at momentum $\mathbf{q}$. Aside from spin waves, the spin structure factor S^{zz} receives contributions from vortex-antivortex roton excitations. In spin language, these correspond to vortex currents generating modulated S^z as discussed above in the AVL. Such excitations are analogous to Feynman's rotons in He-4, and likewise should appear as minima in the structure factor. The energy required to create a roton with momentum $\mathbf{q}$ is simply given by the minimum energy required to promote a fermionic vortex in an occupied band with momentum $\mathbf{k}$ to an unoccupied band with momentum $\mathbf{k}$-$\mathbf{q}$.[12] As J increases, enhancing frustration, the vortex band gap shrinks leading to a sharp reduction in the minimum roton energy at the commensurate wave vectors denoted by open circles in Fig. 1. When the gap closes, signaling the destruction of the canted Néel order, the roton energy becomes *gapless* at these momenta. (For larger J, one enters the AVL phase, and the additional momenta on the right side of Fig. 1 denoted by filled circles then branch out from these roton minima.) The presence of these low-energy rotons should lead to dramatic deviations from linear spin-wave theory.

To conclude, we have provided a concrete theoretical proposal for a spin liquid which may influence the intermediate-energy dynamics of Cs_2CuCl_4 in a magnetic field. The prospect of observing the spin-liquid physics described here is an exciting one, and we hope experiments in this direction will be pursued. Our nontrivial predictions for the dynamic spin structure factor can be tested with inelastic neutron scattering by measuring the lower edge of the continuum scattering at the momenta specified in Fig. 1. Polarized neutrons, in particular, would provide a useful probe for the markedly different correlations identified parallel and transverse to the field. We also identified a stable gapless phase with weak UUD order, which would be interesting to search for via exact diagonalization and series expansions. A renewed look at the excitation spectrum in the UUD plateau as one adds easy-plane anisotropy to suppress the solid order may prove fruitful. Series expansion studies to search for the predicted rotons in the frustrated square lattice limit with J' dominant would also be interesting. More generally, the spin liquid presented here suggests that it may be worthwhile to widen the search for such exotic phases in other frustrated systems by incorporating a finite magnetic field.

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