Ordering in Cs_2CuCl_4: Possibility of a proximate spin liquid

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The layered spiral magnet Cs_2CuCl_4 displays several interesting properties that have been suggested as evidence of proximity to a two-dimensional quantum spin liquid. In this paper we study a concrete version of this proposal and suggest experiments that can potentially confirm it. We study universal critical properties of two-dimensional frustrated quantum magnets near the quantum phase transition between a spiral magnetic state and a spin liquid state with gapped bosonic spinons in the framework of an O(4)-invariant critical theory proposed earlier [A. Chubukov, T. Senthil, and S. Sachdev, Phys. Rev. Lett. **72**, 2089 (1994)]. Direct numerical calculation of the anomalous exponent in spin correlations shows that the critical scattering has broad continua qualitatively similar to experiment. More remarkably we show that the enlarged O(4) symmetry leads to the same slow power-law decay for the vector spin chirality and the Néel correlations. We show how this may be observed through polarized-neutron scattering experiments. A number of other less dramatic consequences of the critical theory are outlined as well.

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I. INTRODUCTION

Despite the great amount of progress made in our theoretical understanding of spin liquid phases[2–4] of quantum magnets, there are to date no convincing experimental examples of such phases in spatial dimension $d \geqslant 2$. A particularly promising candidate—much discussed recently—is the material Cs_2CuCl_4. This is a layered Mott insulator whose spin physics is accurately modeled by a spin-1/2 Heisenberg antiferromagnet on an anisotropic triangular lattice. Neutron scattering measurements have revealed several unusual properties of the spin excitations[5] which have motivated intensive theoretical research works.[6–10] There is incommensurate spiral magnetic order at low temperatures (below 0.62 K). The low-energy excitations of this ordered state are spin waves. However, spin-wave theory fails to quantitatively describe the spin-wave linewidth seen in inelastic neutron scattering. In particular there are long tails that extend to reasonably high frequencies in the inelastic scattering spectrum. This structure may be understood as indicative of a broad continuum of excited states. The continuum survives upon heating above the magnetic phase transition (though the spin wave itself does not). In addition the phase diagram in an external magnetic field shows several interesting features.

The broad continuum is reminiscent of spinon excitations in one-dimensional Heisenberg antiferromagnetic chains. However, the continuum disperses in both spatial directions so that a two-dimensional description of the physics may be more appropriate. A number of workers have therefore suggested interpretations of the neutron data in terms of fractional spin (spinon) excitations in two dimensions into which the magnons decay. In particular Coldea *et al.*[5] suggested that the material may be viewed as being close to a quantum phase transition between a spiral Néel state and a spin liquid state. The purpose of the present paper is to explore this possibility in greater detail and to suggest concrete experimental tests to confirm (or rule out) this proposal.

We begin with some general considerations. It is by now established that a number of different kinds of spin liquid phases are theoretically possible in two-dimensional quantum magnets. We will restrict attention to a particular kind of spin liquid phase whose excitations consist of *bosonic* spin-1/2 spinons with a full spin gap. In addition there are gapped Z_2 vortices (visons) which act as sources of π flux for the spinons. As shown many years ago in Ref. 1 such a spin liquid phase admits a direct second-order transition to the spiral Néel state which is simply driven by condensation of the bosonic spinons. Specifically the transition was argued to be in the universality class of the classical O(4) fixed point in three dimensions. Here we point out several remarkable consequences of this theory which may be used to test its applicability to Cs_2CuCl_4 or other materials. Perhaps most interestingly we show that the large extra O(4) symmetry that emerges at the critical fixed point unifies seemingly different competing orders. The Néel vector correlations have the same slow power-law decay as the vector spin chirality. This nontrivial prediction can potentially be probed in polarized-neutron scattering experiments.

In passing we note that other more exotic spin liquid phases with (possibly gapless) fermionic spinons could potentially exist as stable phases[11–14] but the transitions to the Néel state have not been studied. The corresponding critical theories are also likely to be more exotic—we will therefore defer consideration of such spin liquid phases and the corresponding critical points for the future and focus here on the simpler case with gapped bosonic spinons.

A controlled calculation in which both the magnetically ordered state and such a gapped spin liquid state appear is provided by considering a large-N generalization of the $S=1/2$ Heisenberg model[7] on the anisotropic triangular lattice. Specifically, the large-N limit of a bosonic Sp(N) Heisenberg model[15,16] was used to obtain the mean-field phase diagram as a function of the ratio of intraplane anisotropic couplings,

J'/J, and the strength of quantum fluctuations, $\kappa=$"$2S$", where κ plays the same role as $2S$ (=value of spin at each site) in the SU(2) limit. It was found that, for a range of J'/J, the semiclassical limit (large κ) leads to a spiral magnetic order. As the quantum fluctuations become stronger (small κ), a spin liquid state (Z_2 spin liquid) with gapped deconfined bosonic spinons emerges.

The possibility that Cs_2CuCl_4 is proximate to the quantum critical point between the incommensurate spiral and spin liquid states will affect the behavior at intermediate energy and length scales. As mentioned above Chubukov *et al.*[1] showed quite generally that the transition was in the universality class of the O(4) fixed point in three Euclidean dimensions. This is much higher symmetry than in the original microscopic model. This enlarged O(4) symmetry acts naturally on spinon degrees of freedom which thus emerge as the useful variables already at the transition to the spin liquid. The theory predicts a large anomalous exponent $\bar{\eta}$ for the spin-spin correlation function since the spins are composite operators in terms of spinon operators. Such composite operators usually have large anomalous dimensions. A qualitative physical picture is simply that the spin-1 magnons decay rapidly into the spinons, thereby leading to broad line shapes in the inelastic neutron scattering. However, right at the critical point the spinons are not free particles. From the large-N calculation extrapolated to the physical case of SU(2) spins, the anomalous exponent may be estimated to be 1.54.[1] A more accurate value of $\bar{\eta}$ can be obtained directly by classical Monte Carlo simulations of the O(4) nonlinear σ model in three dimensions. In the present work and in Ref. 17, it is found that $\bar{\eta}=1.373$. It is worth noting that the neutron experiments[5] in Cs_2CuCl_4 were fit to functional forms that also suggest large anomalous dimension $\bar{\eta}_E$ in the range 0.7–1. There is, however, reason to question these measurements of the actual numerical value of $\bar{\eta}$ as we discuss in Sec. V. Nevertheless, the large anomalous dimension predicted by the theory is qualitatively consistent with the broad line shapes seen in experiment.

Thus it is certainly desirable to have other qualitatively distinct predictions of the critical theory. It is our hope that the enlarged O(4) symmetry and the consequent enhanced vector spin chirality correlations will provide such a sharp test. We also outline some other less dramatic consequences of the extra O(4) symmetry that too may be useful.

From a theoretical point of view our considerations are rather similar to those in a recent study[18] of "algebraic spin liquid" phases where too the low-energy theory is characterized by nontrivial enlarged symmetry as compared to the microscopic model. There as in the present problem this enlarged symmetry acts naturally on "spinon" degrees of freedom and leads to nontrivial relationships between the fluctuations of rather different competing orders. Perhaps such phenomena are common in correlated systems.

The rest of this paper is organized as follows. In Sec. II, we introduce the model that we use to describe Cs_2CuCl_4. We briefly review the O(4)-invariant critical theory in Sec. III and discuss the anomalous exponent in Sec. IV A. In Sec. IV B, we study the vector-spin-chirality and scalar-chirality correlation functions. In Sec. V, we discuss possible experiments to test our predictions and the critical theory. Finally, we summarize our results and conclude in Sec. VI.

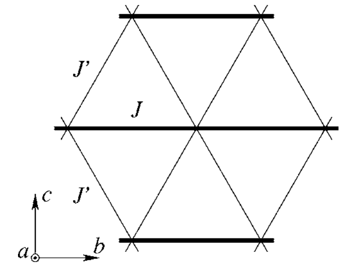

FIG. 1. Schematic representation of the triangular lattice in the b-c plane. The a direction is perpendicular to the plane. There are two coupling constants: J along the horizontal chains and J' along the zigzag bonds.

II. MODEL

The magnetic ions Cu^{2+} in Cs_2CuCl_4 carry $S=1/2$ spin moments that reside on a stack of triangular layers.[5] The intralayer antiferromagnetic interaction (the interaction in the b-c plane; see Fig. 1) is anisotropic with two coupling constants $J\approx 0.375$ meV along the b direction and $J'\approx J/3$ along the zigzag bonds[5] (see Fig. 1). The interlayer interaction (the interaction along the a direction) is weak[5] with a coupling constant $J''=0.045J\ll J$. Therefore the system is quasi two dimensional and can be modeled by a two-dimensional frustrated Heisenberg Hamiltonian on the triangular lattice. The main part of the Hamiltonian reads

$$H=J\sum_{\langle i,j\rangle}\mathbf{S}_i\cdot\mathbf{S}_j+J'\sum_{\langle\langle i,j\rangle\rangle}\mathbf{S}_i\cdot\mathbf{S}_j. \tag{1}$$

Here $\mathbf{S}_i$ are spin-1/2 operators at the sites i of a two-dimensional triangular lattice. The first sum runs over bonds in the b direction, and the second sum runs over the zigzag bonds. There is also a weak Dzyaloshinskii-Moriya interaction.[19] We will neglect this term when we consider the critical theory and discuss later its role in experiments.

The weak interlayer interaction stabilizes magnetic long-range order below $T=0.62$ K.[20] The order is incommensurate because of the frustrated anisotropic interactions in the triangular planes and occurs at an incommensurate wave vector $\mathbf{Q}=(0.5+\epsilon_0)\mathbf{b}^*$ with $\epsilon_0=0.030(2)$ and $\mathbf{b}^*=(2\pi/b,0,0)$. The corresponding classical ordering wave vector is $\mathbf{Q}=(0.5+\epsilon_c)\mathbf{b}^*$ with $\epsilon_c=(1/\pi)\arcsin(J'/2J)=0.053$. The substantial difference is due to large quantum fluctuations that renormalize the ordering wave vector.

Order parameter

The spiral ordering pattern may be represented using two orthonormal unit vectors $\mathbf{n}_1$ and $\mathbf{n}_2$ through

$$\langle\mathbf{S}(\mathbf{r}_i)\rangle\propto\mathbf{n}_1\cos(\mathbf{Q}\cdot\mathbf{r}_i)+\mathbf{n}_2\sin(\mathbf{Q}\cdot\mathbf{r}_i), \tag{2}$$

where $\mathbf{n}_1$ and $\mathbf{n}_2$ satisfy

$$\mathbf{n}_1\cdot\mathbf{n}_2=0,\quad \mathbf{n}_1^2=\mathbf{n}_2^2=1. \tag{3}$$

The $\mathbf{n}_{1,2}$ are clearly vectors under global spin rotation, and together they define the order parameter for the spiral state.

In addition to global spin rotations the spin Hamiltonian, Eq. (1), is invariant under various lattice symmetries and time reversal. It is useful and straightforward to work out the transformation properties of $\mathbf{n}_{1,2}$ under these symmetries. We find

$$T_{\hat{\mathbf{l}}}:(\mathbf{n}_1 + i\mathbf{n}_2) \to (\mathbf{n}_1 + i\mathbf{n}_2)e^{i\mathbf{Q}\cdot\hat{\mathbf{l}}},$$

$$R_c, I:(\mathbf{n}_1 + i\mathbf{n}_2) \to (\mathbf{n}_1 - i\mathbf{n}_2),$$

$$T:(\mathbf{n}_1 + i\mathbf{n}_2) \to -(\mathbf{n}_1 - i\mathbf{n}_2). \quad (4)$$

Here $T_{\hat{\mathbf{l}}}$ is a unit translation along a lattice vector $\mathbf{l}$ direction, R_c is reflection about the c axis, I is lattice inversion, and T is time reversal. Other symmetry operations may be obtained as combinations of the above.

III. THEORY OF THE QUANTUM TRANSITION

In this section we briefly review the theory of Ref. 1 for the quantum transition between the Néel and spin liquid states. We will also use this as an opportunity to clarify several possible confusions with other superficially similar results in the literature.

The set $\mathbf{n}_1, \mathbf{n}_2$ defines the order parameter for the spiral state. To study phase transitions out of the spiral state it is necessary to allow fluctuations where $\mathbf{n}_{1,2}$ vary slowly as a function of space and time. The symmetry properties in Eq. (4) actually imply that the resulting continuum theory has an extra U(1) symmetry over and above that of spin rotations. This may be seen by considering translations along the $\mathbf{b}$ direction. We have

$$(\mathbf{n}_1 + i\mathbf{n}_2) \to (\mathbf{n}_1 + i\mathbf{n}_2)e^{imQ_x} \quad (5)$$

for a translation by m lattice sites. With incommensurate Q_x, this effectively translates into a full U(1) symmetry that rotates between the $\mathbf{n}_1$ and $\mathbf{n}_2$ fields.

The order parameter manifold defined by $\mathbf{n}_{1,2}$ allows for topological vortex defects with a discrete Z_2 character.[21] These Z_2 vortices have energy logarithmic in the system size in the ordered state due to the long-distance distortion of the order parameter. Now consider disordering the spiral order while keeping the core energy of these Z_2 vortices finite. Reference 1 argues that the resulting state is a fractionalized spin liquid with bosonic spin-1/2 spinons. The Z_2 vortices survive into the spin liquid phase but now only cost finite (not divergent) energy.

A theory for this transition is obtained by writing

$$\mathbf{n}_1 + i\mathbf{n}_2 = \epsilon_{\alpha\gamma} z_\gamma \boldsymbol{\sigma}_{\alpha\beta} z_\beta, \quad (6)$$

where ϵ is the antisymmetric tensor, σ^a are the Pauli matrices, and $z_\alpha=(z_\uparrow, z_\downarrow)$ is a two-component complex unit vector satisfying

$$z^\dagger z = 1. \quad (7)$$

It is easy to check that the parametrization (6) satisfies the constraints given by Eq. (3). The z_α transform as spinors under the SU(2) spin rotation and describe spin-1/2 spinons. This representation clearly has a Z_2-gauge redundancy associated with changing the sign of z at any point in space-time. Thus a reformulation in terms of z may be fruitfully viewed as a theory of the z fields coupled to a Z_2-gauge field. The corresponding Z_2-gauge flux is associated with the Z_2 vortices discussed above. These vortices stay gapped at the transition between the Néel and spin liquid states and may hence be ignored for a low-energy description of the critical point. Thus we may obtain the critical behavior by focusing only on the spinons and ignoring their coupling to the Z_2-gauge field. Detailed arguments show that the critical theory is in fact in the O(4) universality class in $D=2+1$ dimensions where there is full rotational symmetry between the four real numbers described by $(z_\uparrow, z_\downarrow)$. In effect the extra U(1) symmetry of rotation between $\mathbf{n}_1$ and $\mathbf{n}_2$ has been enlarged to SU(2). Combined with the SU(2) spin rotations the full symmetry is $SU(2)\times SU(2) \sim O(4)$. Thus the critical properties may be computed using the Euclidean action

$$\mathcal{S} = \int d^2x d\tau \sum_{\mu=x,\tau} \frac{1}{g} \partial_\mu z^*_\alpha \partial_\mu z_\alpha. \quad (8)$$

The relationship between the z and $\mathbf{n}_{1,2}$ fields may also be expressed in a different way that will be fruitful later. Let us introduce an SU(2) matrix U built out of z:

$$U = \begin{pmatrix} z_\uparrow & z^*_\downarrow \\ z_\downarrow & -z^*_\uparrow \end{pmatrix}. \quad (9)$$

Then U satisfies

$$U^\dagger \sigma^a U = R^{ab}\sigma^b, \quad (10)$$

where R is a 3×3 rotation matrix. Clearly,

$$R^{ab} = \frac{1}{2} tr(U^\dagger \sigma^a U \sigma^b). \quad (11)$$

It is readily checked that $\mathbf{n}_1$ and $\mathbf{n}_2$ are the first and second columns, respectively, of this rotation matrix. The third column represents the unit vector

$$\mathbf{n}_3 = \mathbf{n}_1 \times \mathbf{n}_2, \quad (12)$$

which is orthogonal to both $\mathbf{n}_1$ and $\mathbf{n}_2$. Symmetry under physical spin rotations corresponds to *left* multiplication of U by an SU(2) spin rotation matrix:

$$U \to VU, \quad (13)$$

with $V \in SU(2)$. The enlarged O(4) symmetry implies that *right* multiplication

$$U \to UV \quad (14)$$

is also a symmetry of the critical fixed point.

Several comments are in order on these results. Prior to Ref. 1, analysis of a continuum nonlinear σ model appropriate for noncollinear magnets had led to the suggestion of enlarged O(4) symmetry in $2+\epsilon$ space-time dimensions.[22] However, numerical calculations on stacked triangular lattices near their finite-temperature ordering transition failed to observe the predicted O(4) universality class.[23] Rather the evidence supports a transition in the "chiral" universality class of Refs. 23 and 24. How then are we to reconcile this with our claims about the quantum-Néel–spin-liquid transition being in the O(4) universality class? The answer lies in the nature of the paramagnetic phase. In the classical stacked triangular lattice the natural paramagnetic phase is the trivial one that is smoothly connected to the high-temperature limit. The transition to this phase from the ordered state is obtained by proliferating the Z_2 vortices (which are line defects in the three-dimensional classical model). In principle an exotic paramagnetic phase is also possible which would be the classical three-dimensional analog of the spin liquid: this requires destroying the magnetic order without proliferating the Z_2 vortices. This paramagnet will be topologically ordered and will be separated from the trivial very-high-temperature paramagnet by a phase transition. In this classical context, the arguments of Ref. 1 apply to the transition between the Néel state and this topologically ordered paramagnet which will indeed be in the O(4) universality class. However, neither this nontrivial paramagnet nor the corresponding transition was apparently accessed in the numerical calculations.[23] Equally it is hard to decide which of the two possible paramagnetic phases were accessed in the $2+\epsilon$ calculations of Ref. 22. The distinction between the two phases is topological and hence sensitive to spatial dimension. This is hard to disentangle in the ϵ expansion.

Turning to the quantum problem at hand it differs from the classical stacked magnet in an important way. There are extra Berry phases that are sensitive to the microscopic spin at each lattice site (i.e., spin 1/2 or spin 1, etc.). A close and familiar analogy is from the theory of collinear quantum antiferromagnets in two dimensions where such Berry phases spoil any general direct mapping to classical collinear magnets in one higher dimension. In that case in the semiclassical limit the Berry phases are associated entirely with singular topological configurations known as hedgehogs. A similar result also holds for the noncollinear quantum magnets of interest in this paper. The Berry phases are associated entirely with the topological Z_2 vortex configurations. As these Z_2 vortices are gapped across the transition to the spin liquid the Berry phases play no role in the low-energy universal critical physics. Indeed the O(4) universality class will describe the Néel–spin-liquid transition for all spin-S magnets regardless of the value of S. On the other hand, the phase obtained when the Néel state is disordered by proliferating the Z_2 vortices will be strongly influenced by the Berry phases. For spin 1/2 it is expected that the Berry phases will lead to broken lattice symmetries in the resulting paramagnet.[25] The nature of the transition between such a valence bond solid (VBS) ordered paramagnet and the Néel state on the triangular lattice is not presently understood. Preliminary analysis suggests that an interesting "Landau-forbidden" second-order transition may be possible. However, in the present paper we restrict ourselves to studying the transition to the spin liquid.

IV. CONSEQUENCES OF THE CRITICAL THEORY

A. Spin correlations

We begin by considering the spin-spin correlation function at the ordering wave vector which is readily accessed in neutron scattering experiments. This can be expressed in terms of the correlation functions of the vectors $\mathbf{n}_1$ and $\mathbf{n}_2$:

$$\langle \mathbf{S}(\mathbf{r},\tau)\cdot\mathbf{S}(0,0)\rangle = \langle \mathbf{n}_1(\mathbf{r},\tau)\cdot\mathbf{n}_1(0,0)\rangle \cos(\mathbf{Q}\cdot\mathbf{r}). \quad (15)$$

We note that due to the symmetry of rotations between $\mathbf{n}_1$ and $\mathbf{n}_2$, they will both have the same correlations. Close to the critical point, the correlation function has the usual power-law behavior

$$\langle \mathbf{S}(-\mathbf{q},\omega)\cdot\mathbf{S}(\mathbf{q},\omega)\rangle \sim \frac{1}{(\omega^2-k^2)^{1-\bar{\eta}/2}}, \quad (16)$$

where $\mathbf{k}=\mathbf{q}-\mathbf{Q}$. Since $\mathbf{n}_1$ and $\mathbf{n}_2$ (and $\mathbf{S}$) are composite operators in terms of the $\bar{z}_\alpha$ fields, one can expect that the spin-spin correlation function has a large anomalous exponent $\bar{\eta}$. Indeed, the large-N calculation of $\bar{\eta}$ for an O($2N$) theory gives[1]

$$\bar{\eta} = 1 + \frac{32}{3\pi^2 N}. \quad (17)$$

For the physical case $N=2$, we have $\bar{\eta}\approx 1.54$. This is a large number compared to anomalous exponents of noncomposite operators.

To improve the large-N estimate of the anomalous exponent, one can alternately perform Monte Carlo simulations of the classical O(4) nonlinear σ model in three dimensions since this model is in the same universality class as the transition of interest. Ballesteros *et al.*[17] measured the anomalous exponent η_T of the tensorial magnetization, which is defined as

$$M_T = \langle \sqrt{\mathrm{Tr}\mathcal{M}^2}\rangle,$$

$$\mathcal{M}_{\alpha\beta} = \sum_i \left(\phi_{i\alpha}\phi_{i\beta} - \frac{1}{4}\delta_{\alpha\beta}\right),$$

where $\phi_{i\alpha}$ are four components of the O(4) unity vector at the site i in three dimensions. They found $\eta_T=1.375(5)$. This result is directly related to our case because of the following argument. Using Eq. (6) and writing $z_\uparrow=(\phi_1+i\phi_2)$ and $z_\downarrow=(\phi_3+i\phi_4)$, where ϕ_α are now real fields, we have

$$\mathbf{n}_1 = \begin{pmatrix} \phi_2^2+\phi_3^2-\phi_1^2-\phi_4^2 \\ 2(\phi_1\phi_2+\phi_3\phi_4) \\ 2(\phi_1\phi_3-\phi_2\phi_4) \end{pmatrix}, \quad (18)$$

$$\mathbf{n}_2 = \begin{pmatrix} 2(\phi_3\phi_4-\phi_1\phi_2) \\ \phi_2^2+\phi_4^2-\phi_1^2-\phi_3^2 \\ 2(\phi_1\phi_4+\phi_2\phi_3) \end{pmatrix}. \quad (19)$$

We can see that the tensorial magnetization is related to $\mathbf{n}_1$ and $\mathbf{n}_2$ by symmetry and therefore the anomalous exponent of the tensorial magnetization is equal to the anomalous exponent of the vectors $\mathbf{n}_1$ or $\mathbf{n}_2$. Then, from Eq. (15), it is the same as the anomalous exponent of the correlation function at wave vector $\mathbf{Q}$.

We have confirmed this conclusion by computing the anomalous exponent of $\mathbf{n}_1$ directly. We simulate the O(4) nonlinear σ model in three dimensions on the simple cubic lattice using a cluster algorithm.[26] We measure the anomalous exponent $\bar{\eta}$ in the following way. We calculate the total z component $n_1^z=2(\phi_1\phi_3-\phi_2\phi_4)$ of the vector $\mathbf{n}_1$:

$$M=\sum_i n_{1i}^z, \tag{20}$$

where the sum runs over all lattice sites. In our simulations, we measure the "magnetization" and "susceptibility"

$$\bar{m}=\frac{1}{L^3}\langle M\rangle,$$

$$\bar{\chi}=\langle M^2\rangle,$$

where L is the system size. We perform our simulations only at the critical temperature found in Ref. 17 with very high accuracy, $\beta_c=1/T_c=0.935\,861(8)$. To find the critical exponents, we use finite-size scaling analysis. Close to the critical point, observables satisfy the general scaling form

$$\mathcal{O}=L^{\rho/\nu}F_{\mathcal{O}}(tL^{1/\nu},L^{-\omega}), \tag{21}$$

where $\mathcal{O}$ can be $\bar{m}$ or $\bar{\chi}$, ρ is the scaling exponent corresponding to the operator $\mathcal{O}$ $[\rho=-\bar{\beta}=-\nu(1+\bar{\eta})/2$ for $\bar{m}$ and $\rho=\bar{\gamma}=\nu(2-\bar{\eta})$ for $\bar{\chi}]$, $F_{\mathcal{O}}$ is a universal function, $t=|T-T_c|$, and ω is a universal exponent related to the leading irrelevant operator. Exactly at the critical point $t=0$, one can write

$$\mathcal{O}=L^{\rho/\nu}(a+cL^{-\omega}). \tag{22}$$

We assume that the correction to scaling is negligible for large lattice sizes and fit to a simplified scaling form

$$\mathcal{O}=aL^{\rho/\nu}. \tag{23}$$

We have performed simulations for large enough lattices (up to $L_{\max}=96$). Fitting from $L_{\min}=20$ to $L_{\max}$, we obtain $\bar{\eta}=1.373(3)$.

The large value of $\bar{\eta}$ is a reflection of the emergence of spinons as useful degrees of freedom at the critical point.

B. Other competing orders

The extra O(4) symmetry actually has more striking consequences for the critical properties. It implies that operators other than the natural magnetic order parameter will have enhanced power-law correlators. Consider the magnetic order parameters $\mathbf{n}_{1,2}$. As discussed in Sec. III, they may be regarded as the first and second columns of a rotation matrix R. The third column of the rotation matrix is $\mathbf{n}_3=\mathbf{n}_1\times\mathbf{n}_2$. The enlarged O(4) symmetry at the critical point implies that both left and right multiplications by an orthogonal matrix of this rotation matrix R are symmetries of the critical theory. Left multiplication is just physical spin rotation. However, symmetry under right multiplication implies that $\mathbf{n}_{1,2,3}$ will all have the same correlations. In particular $\mathbf{n}_3$ will have the same slow power-law decay as $\mathbf{n}_{1,2}$ calculated in the previous subsection. We can express the vector $\mathbf{n}_3$ in terms of the spin operator as follows. Consider $\mathbf{S}_{\mathbf{r}+\hat{\mathbf{l}}}\times\mathbf{S}_{\mathbf{r}}$, where $\hat{\mathbf{l}}$ is a unit lattice vector. Using Eq. (2) and expanding the first spin operator around $\mathbf{r}$, we have

$$\mathbf{S}_{\mathbf{r}+\hat{\mathbf{l}}}\times\mathbf{S}_{\mathbf{r}}\sim\mathbf{n}_3\sin(\mathbf{Q}\cdot\hat{\mathbf{l}})+\text{derivative terms}. \tag{24}$$

We can ignore the derivatives terms since their contribution to the correlation function is negligible. Consider the chirality vector

$$\mathbf{C}_{\mathbf{r}}=\sum_{\mathbf{r}'}\mathbf{S}_{\mathbf{r}}\times\mathbf{S}_{\mathbf{r}'}, \tag{25}$$

where the oriented sum is over an elementary plaquette. Using Eq. (24), it is easy to show that

$$\mathbf{C}=\left[\sin Q_x-2\sin\frac{Q_x}{2}\cos\frac{\sqrt{3}Q_y}{2}\right]\mathbf{n}_3\propto\mathbf{n}_3. \tag{26}$$

Thus the vector spin-chirality correlation function

$$\langle\mathbf{C}(\mathbf{r},\tau)\cdot\mathbf{C}(\mathbf{0},0)\rangle\propto\langle\mathbf{n}_3(\mathbf{r},\tau)\cdot\mathbf{n}_3(0,0)\rangle=\langle\mathbf{n}_1(\mathbf{r},\tau)\cdot\mathbf{n}_1(0,0)\rangle \tag{27}$$

has the same power-law behavior at zero wave vector as those of spins at the wave vector $\mathbf{Q}$. This highly nontrivial statement possibly provides a way to test the theory of Ref. 1 in experiments and in model numerical calculations.

C. Conserved quantities

Some less dramatic effects of the enlarged O(4) symmetry are also worth noting. The O(4) group has six generators, and these will all be conserved at the critical fixed point. Three of these are just the conserved total spin $\mathbf{S}_{tot}=\Sigma_i\mathbf{S}_i$—they are the generators of left rotations of U. The remaining three are conserved only in the low-energy critical theory (though not in the original microscopic model). They correspond to right rotations of U. We will denote these K_a, $a=1,2,3$. The K_a transform as vectors under the group $SU(2)_R$ of right rotations of U. They generate rotations of $\mathbf{n}_a$ among one another so that

$$[\mathbf{n}_a,K_b]=i\epsilon_{abc}\mathbf{n}_c. \tag{28}$$

As is well known such conserved quantities have scaling dimension $d_{\text{scale}}=2$ with no anomalous dimension.[27] Thus, at the critical point at zero temperature,

$$\langle K_a(\mathbf{r},\tau)K_b(\mathbf{0},0)\rangle\sim\frac{\delta_{ab}}{(|\mathbf{r}|^2+\tau^2)^2}, \tag{29}$$

where $\mathbf{r}$ is the spatial coordinate and τ is the (imaginary) time coordinate. A similar result also holds for the conserved total spin. Away from the critical point at finite temperature in the quantum critical region, the K_a will continue to be approximately conserved. The exact conservation will be spoiled by irrelevant operators that break the O(4) symmetry down to SU(2)×lattice space group. Thus at $T>0$ in the quantum critical region we expect that the K_a will diffuse up to a long length and time scale which will be determined by T and the distance to the critical point. It is possible that the presence of such extra nearly diffusive modes can also be looked for in experiments.

But what do the K_a correspond to in terms of the underlying spins? A useful guess for the answer is provided by examining the transformation properties of K_a under the microscopic symmetries of the original lattice system. It is reasonable that any lattice operator that has the same transformation properties will have some overlap in the continuum theory with the K_a. With the further assumption that there are no other operators in the continuum theory with smaller scaling dimension that also have the same transformation, the K_a will give the dominant correlations of such lattice operators. Similar considerations were also used in Ref. 18.

The transformation properties of K_a under all physical symmetries are fixed by the commutation relation, Eq. (28). The transformation properties of K_a are related to the transformation properties of the matrix R. First, we list the latter ones:

$$\mathrm{SU}(2)_{\mathrm{spin}}: \quad R \to OR,$$

$$T_{\hat{\mathbf{l}}}: \quad R \to R \begin{pmatrix} \cos(\mathbf{Q}\cdot\hat{\mathbf{l}}) & \sin(\mathbf{Q}\cdot\hat{\mathbf{l}}) & 0 \\ -\sin(\mathbf{Q}\cdot\hat{\mathbf{l}}) & \cos(\mathbf{Q}\cdot\hat{\mathbf{l}}) & 0 \\ 0 & 0 & 1 \end{pmatrix},$$

$$R_c, I: \quad R \to R \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & -1 \end{pmatrix},$$

$$T: \quad R \to R \begin{pmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 1 \end{pmatrix},$$

where O is an O(3) matrix and $T_{\hat{\mathbf{l}}}$ is the lattice translation along the lattice vector $\hat{\mathbf{l}}$. We can use these transformations and the condition that K_a are vectors under $\mathrm{SU}(2)_R$ to obtain the transformations of K_a:

$$\mathrm{SU}(2)_{\mathrm{spin}}: \quad (K_1,K_2,K_3) \to (K_1,K_2,K_3),$$

$$T_{\hat{\mathbf{l}}}: \quad (K_1,K_2,K_3) \to (K_1,K_2,K_3)M,$$

$$R_c, I: \quad (K_1,K_2,K_3) \to (K_1,-K_2,-K_3),$$

$$T: \quad (K_1,K_2,K_3) \to (K_1,K_2,K_3),$$

where

$$M = \begin{pmatrix} \cos(\mathbf{Q}\cdot\hat{\mathbf{l}}) & -\sin(\mathbf{Q}\cdot\hat{\mathbf{l}}) & 0 \\ \sin(\mathbf{Q}\cdot\hat{\mathbf{l}}) & \cos(\mathbf{Q}\cdot\hat{\mathbf{l}}) & 0 \\ 0 & 0 & 1 \end{pmatrix}. \tag{30}$$

We can construct local spin operators that have the same symmetry properties as K_a. We find that

$$K_1(\mathbf{r}) \sim \cos(\mathbf{Q}\cdot\mathbf{r})\mathbf{S}(\mathbf{r})\cdot[\mathbf{S}(\mathbf{r}-\hat{\mathbf{x}})+\mathbf{S}(\mathbf{r}+\hat{\mathbf{x}})], \tag{31}$$

$$K_2(\mathbf{r}) \sim \sin(\mathbf{Q}\cdot\mathbf{r})\mathbf{S}(\mathbf{r})\cdot[\mathbf{S}(\mathbf{r}-\hat{\mathbf{x}})+\mathbf{S}(\mathbf{r}+\hat{\mathbf{x}})], \tag{32}$$

$$K_3(\mathbf{r}) \sim \mathbf{S}(\mathbf{r}-\hat{\mathbf{x}})\cdot[\mathbf{S}(\mathbf{r})\times\mathbf{S}(\mathbf{r}+\hat{\mathbf{x}})]. \tag{33}$$

Thus K_1+iK_2 transforms identically with the Fourier transform of the bond energy at wave vector $\mathbf{Q}$ while K_3 can be identified with the scalar spin chirality along the chains.

V. EXPERIMENTAL IMPLICATIONS

A. Spin correlations

Quite generally in the quantum critical region the spin correlations at the ordering wave vector must satisfy scaling—for instance, as a function of ω/T. Cs_2CuCl_4 orders at low temperature. Consequently data at the lowest temperatures and frequencies must be excluded from the scaling analysis. Furthermore, it is precisely the very-low-T and low-ω range that will also be most sensitive to the presence of weak spin anisotropies (such as Dzyaloshinski-Moriya interactions). However, the data at intermediate T and ω will be less affected by such spin anisotropies or by the low-energy long-range order.

The value of $\bar{\eta}$ found from Monte Carlo simulations can be compared to $\bar{\eta}_E$ estimated in experiments using a fitting procedure.[5] The experimental values fall in a range between 0.7 and 1. Thus the Monte Carlo result is still larger than the experimentally estimated value. It is important to note the following caveat on this comparison. The existing measurements of $\bar{\eta}_E$ were performed in inelastic neutron scattering experiments where *both the wave number and frequency transfer to the sample were simultaneously varied*. In particular typical data sets (such as for instance scan G of Ref. 5) involve increasing the frequency transfer while the momentum transfer varies from points in the two-dimensional Brillouin zone far away from the ordering wave vector $\mathbf{Q}$ to points close to $\mathbf{Q}$. In the context of the ideas explored in this paper the frequency dependence at fixed $\mathbf{q}$ far away from $\mathbf{Q}$ will not be dominated by the singular long-distance critical fluctuations and will be highly nonuniversal. In contrast the frequency dependence for $\mathbf{q}$ close to $\mathbf{Q}$ will be determined by the long-distance critical fluctuations and will be universal. It is these latter fluctuations that are described by the calculated exponent $\bar{\eta}$. Thus the existing measurements of $\bar{\eta}_E$ are quite possibly severely contaminated by nonuniversal short-distance effects. Hence the lack of quantitative agreement between $\bar{\eta}$ and $\bar{\eta}_E$ is not surprising. However, the qualitative observation of broad magnon linewidths is consistent with the large $\bar{\eta}$ obtained in the theory. Future experiments will hopefully directly probe the fluctuations near the ordering wave vector, thereby allowing for quantitative comparison.

As noted already in Ref. 1, NMR experiments may be a useful way to directly measure $\bar{\eta}$. For the nuclear relaxation rate we have

$$\frac{1}{T_1} \sim T^{\bar{\eta}}. \tag{34}$$

Again this behavior will only obtain at intermediate temperatures.

B. Detection of vector spin-chirality fluctuations

A dramatic consequence of the critical theory studied in this paper is the enhanced vector spin-chirality correlations discussed in Sec. IV B. How can they be detected in experiments? In this section we show how this may be done through polarized neutron scattering. Our proposal builds on and generalizes the pioneering ideas of Maleyev[28] in the context of classical noncollinear magnets.

The part of the neutron scattering rate R_P that depends on the incoming neutron polarization $\mathbf{P}_i$ is given by

$$R_P(\mathbf{k}_i,\mathbf{P}_i \to \mathbf{k}_f) \sim (\mathbf{P}_i \cdot \hat{q})(\hat{q} \cdot \mathbf{b}), \tag{35}$$

where $\mathbf{q}=\mathbf{k}_f-\mathbf{k}_i$ is the three-momentum transferred to the sample and $\hat{q}$ is the corresponding unit vector. The vector $\mathbf{b}$ is given by

$$b_\alpha = \frac{\epsilon_{\alpha\beta\gamma}}{2i} S_{\beta\gamma}, \tag{36}$$

where $S_{\beta\gamma}$ is the spin structure factor. Thus the polarization-dependent part probes the *antisymmetric* part of the structure factor. By the usual arguments it is clear that b_α can be obtained from a calculation of the imaginary-time Green function (here $0<\tau<\beta=1/T$)

$$\mathbf{g}(\mathbf{r},\tau) = \langle \mathbf{S}(\mathbf{r},\tau) \times \mathbf{S}(\mathbf{0},0)\rangle, \tag{37}$$

$$\mathbf{g}(\mathbf{q},i\omega_n) = \int_{r\tau} e^{i\mathbf{q}\cdot\mathbf{r}-i\omega_n\tau}\mathbf{g}(\mathbf{r},\tau). \tag{38}$$

Specifically we have

$$\mathbf{b}(\mathbf{q},\omega) = \frac{-1}{1-e^{-\beta\omega}}\left(\frac{\mathbf{g}(\mathbf{q},i\omega_n \to \omega + i0^+) - \mathbf{g}(\mathbf{q},\omega + i0^-)}{2}\right). \tag{39}$$

In the context of this paper using Eq. (2) we find

$$\mathbf{g}(\mathbf{r},\tau) = \langle \mathbf{n}_1(\mathbf{r},\tau) \times \mathbf{n}_1(\mathbf{0},0)\rangle \cos(\mathbf{Q}\cdot\mathbf{r}) + \langle \mathbf{n}_2(\mathbf{r},\tau) \times \mathbf{n}_1(\mathbf{0},0)\rangle \sin(\mathbf{Q}\cdot\mathbf{r}).$$

Clearly such correlators are zero if there is full spin isotropy. In the specific case of Cs_2CuCl_4 the presence of a weak Dzyaloshinski-Moriya (DM) interaction will lead to a nonzero result. It is known that the DM interaction is nonzero along oriented zigzag bonds. For a single triangular layer,

$$H_{DM} = -\mathbf{D}\cdot\sum_{\mathbf{r}} \mathbf{S}_{\mathbf{r}} \times (\mathbf{S}_{\mathbf{r}+\delta_1} + \mathbf{S}_{\mathbf{r}+\delta_2}). \tag{40}$$

Here $\delta_{1,2}$ are unit vectors along the oriented zigzag bonds as shown in Fig. 2. In addition H_{DM} is also staggered between different layers. The DM vector is oriented along the a axis and has magnitude $D \approx 0.02$ meV $\approx 0.05\ J$. In the continuum theory we may again use Eq. (2) to write

$$H_{DM} \approx -\mathbf{d}\cdot\int d^2r\,\mathbf{n}_1 \times \mathbf{n}_2 \tag{41}$$

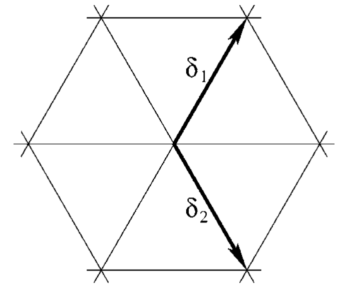

FIG. 2. Dzyaloshinski-Moriya interaction is nonzero along oriented zigzag bonds that are denoted by vectors δ_1 and δ_2.

$$= -\mathbf{d}\cdot\int d^2r\,\mathbf{n}_3. \tag{42}$$

Here $\mathbf{d} \propto \mathbf{D}$. Let us now try to evaluate $\mathbf{g}$ to linear order in $\mathbf{d}$. We get

$$\mathbf{g}(x) = \int d^3x' \langle \mathbf{n}_1(x) \times \mathbf{n}_1(0)[\mathbf{d}\cdot\mathbf{n}_3(x')]\rangle \cos(\mathbf{Q}\cdot\mathbf{r}) + \int d^3x' \langle [\mathbf{n}_2(x) \times \mathbf{n}_1(0)][\mathbf{d}\cdot\mathbf{n}_3(x')]\rangle \sin(\mathbf{Q}\cdot\mathbf{r}).$$

Here $x=(\mathbf{r},\tau), x'=(\mathbf{r}',\tau')$ are space-time coordinates. The averages are evaluated in the isotropic theory. The first average vanishes due to the symmetry $\mathbf{n}_3 \to -\mathbf{n}_3, \mathbf{n}_1 \to \mathbf{n}_1$ present in the action. We are therefore left with

$$\mathbf{g}(x) = d_\alpha \int d^3x' \langle \mathbf{n}_2(x) \times \mathbf{n}_1(0) n_{3\alpha}(x')\rangle \sin(\mathbf{Q}\cdot\mathbf{r}). \tag{43}$$

Thus $\mathbf{g}$ is determined by the three-point correlation function of $\mathbf{n}_{1,2,3}$. Using spin rotation invariance we have

$$\mathbf{g}(\mathbf{r},\tau) = \frac{\mathbf{d}}{3}\sin(\mathbf{Q}\cdot\mathbf{r}) \int d^3x' \langle \mathbf{n}_2(x) \times \mathbf{n}_1(0)\cdot\mathbf{n}_3(x')\rangle. \tag{44}$$

Such three-point correlators of scaling fields are severely restricted by conformal invariance.[29] As $\mathbf{n}_{1,2,3}$ all have the same scaling dimension $\Delta=(1+\bar{\eta})/2$, we have (at $T=0$)

$$\langle \mathbf{n}_2(x) \times \mathbf{n}_1(0)\cdot\mathbf{n}_3(x')\rangle \sim \frac{1}{x^\Delta x'^\Delta |x-x'|^\Delta}. \tag{45}$$

We now have to integrate over x' which is the coordinate of $\mathbf{n}_3$. It is easy to see that this integral is infrared (IR) divergent. This is because at large x', the integrand behaves as $1/x'^{2\Delta}$. With $2\Delta=1+\bar{\eta}\approx 2.37$, the integral over x' will diverge in the infrared. Formally this just means that perturbation theory in the DM interaction diverges at $T=0$ and the correct answer will involve some fractional power of the DM vector. However, for our purposes it is much more meaningful and simpler to go to finite temperature where this divergence will be cut off. Before doing that it is useful to understand the origin of this divergence at $T=0$ better.

The divergence comes from large $x' \gg x$. To discuss this limit let us keep x' fixed and bring x close to 0. Then we need the operator product expansion (OPE) of $\mathbf{n}_2 \times \mathbf{n}_1$. This is a vector in spin space, and the leading term will just be $\mathbf{n}_3$. Scaling requires the equation

$$\mathbf{n}_2(x) \times \mathbf{n}_1(0) \sim \frac{1}{x^{\Delta}} \mathbf{n}_3(x). \qquad (46)$$

There is only one power of Δ on the right-hand side so that both sides will scale identically. Now, if we calculate the correlator with $\mathbf{n}_3(x')$, we will reproduce the limit of the exact result, Eq. (45), above when x' is large. It is now clear that the infrared divergence in the integral over x' actually is nothing but the divergence of the uniform susceptibility of the vector spin chirality at zero temperature.

Armed with this insight let us discuss $T>0$. We will assume that T is large enough that perturbation theory in D is meaningful. This requires T bigger than an energy scale ω_D set by D. For small D, it is easy to see from scaling that $\omega_D \sim D^{1/\Delta}$. Here we will also assume that we are interested in the scattering at an external frequency $\omega \gg T$. This simplifies things because then the sole effect of $T>0$ is to cut off the IR divergence without affecting the rest of the correlations.

Using the OPE it is now clear that the answer is the Fourier transform of

$$\frac{1}{x^{\Delta}} \chi_{ch}(T), \qquad (47)$$

where $\chi_{ch}(T)$ is the uniform vector spin-chirality susceptibility at temperature T. From scaling we have

$$\chi_{ch}(T) \sim \frac{1}{T^{3-2\Delta}}. \qquad (48)$$

Using this we may straightforwardly calculate the inelastic scattering rate. For scattering right at the ordering wave vector $\mathbf{Q}$ and for $\omega \gg T$, we get

$$R_P(\omega) \sim \frac{(\mathbf{P}_i \cdot \hat{\mathbf{q}})(\hat{\mathbf{q}} \cdot \mathbf{D})}{\omega^{3-\Delta} T^{3-2\Delta}}, \qquad (49)$$

with $2\Delta = 1 + \bar{\eta} \approx 2.37$.

This is a definite prediction that can possibly be tested. In the actual experiments it will be necessary to take into account the staggering of the DM interaction between the different layers. Thus it is best to choose the a component of the three-vector $\mathbf{q}$ to be π. We note that in the same range $\omega \gg T$, the intensity in unpolarized neutron scattering behaves as

$$R(\omega) \sim \frac{1}{\omega^{3-2\Delta}}. \qquad (50)$$

Therefore, the dimensionless ratio of the polarization-dependent part to the polarization independent part behaves as

$$\frac{R_P(\omega)}{R(\omega)} = (\mathbf{P}_i \cdot \hat{\mathbf{q}}) \left(\hat{\mathbf{q}} \cdot \frac{\mathbf{D}}{J} \right) \left(\frac{J}{\omega} \right)^{\Delta} \left(\frac{J}{T} \right)^{3-2\Delta} F\left(\frac{J'}{J} \right). \qquad (51)$$

Here, without loss of generality, we have used J to convert all energy scales into dimensionless numbers. The function $F(J'/J)$ is an as-yet undetermined function of the dimensionless ratio of J' and J. This equation is correct to linear order in D. We expect that, for Cs_2CuCl_4 where $J' \approx J/3$, F will be a number of order 1. We note again that this is correct for $\omega \gg T \gg \omega_D$.

In practice successful confirmation of these predictions will require a large enough window of energy scales where the asymptotic critical behavior controlled by the O(4) fixed point is visible. It is an open question whether such a window is available in Cs_2CuCl_4 or not.

VI. SUMMARY AND CONCLUSIONS

In this paper we have pursued a concrete version of the idea that Cs_2CuCl_4, though magnetically ordered at low temperature, may nevertheless be proximate to a spin liquid phase. Such proximity suggests that the intermediate length- and time-scale physics of Cs_2CuCl_4 may be governed by a quantum critical point between a magnetic spiral and a genuine two-dimensional quantum spin liquid. A theory for such a quantum phase transition was obtained in Ref. 1 for a simple (Z_2) spin liquid with gapped bosonic spinons. We showed that the spin correlations at the quantum critical point are characterized by a large anomalous exponent $\bar{\eta} \approx 1.37$. This is qualitatively consistent with the broad power-law tails observed in inelastic neutron scattering in Cs_2CuCl_4. Further we showed that the enlarged O(4) symmetry[1] at the critical fixed point has some remarkable consequences. The vector spin chirality has the same slow power-law decay as the natural magnetic order parameter. This sharp qualitative observation should be of great use in confirming (or ruling out) the applicability of the theory of Ref. 1 to Cs_2CuCl_4. Building on Ref. 28, we showed how polarized inelastic neutron scattering can be used to directly detect the vector spin-chirality correlations. It is our hope that future experiments will be able to use these results to clarify the physics behind the interesting properties of Cs_2CuCl_4.

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