Gapless Spin Liquids on the Three-Dimensional Hyperkagome Lattice of $Na_4Ir_3O_8$

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Recent experiments indicate that $Na_4Ir_3O_8$, a material in which $s = 1/2$ Ir local moments live on a three-dimensional "hyperkagome" lattice of corner-sharing triangles, may have a quantum spin liquid ground state with gapless spin excitations. Using a combination of exact diagonalization, symmetry analysis of fermionic mean field ground states and Gutzwiller projected variational wave functions studies, we propose a quantum spin liquid with spinon Fermi surfaces as a favorable candidate for the ground state of the Heisenberg model on this lattice. We point out implications of this proposal for thermodynamic properties and discuss possible weak instabilities of the spinon Fermi surfaces.

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Introduction.—$Na_4Ir_3O_8$ is a recently discovered three-dimensional (3D) frustrated quantum magnet [1]. The Ir atoms in this insulating compound have $s = 1/2$ local moments and form a 3D network of corner-sharing triangles called a "hyperkagome" lattice [1], a cubic lattice whose unit cell is shown in Fig. 1. High temperature magnetic susceptibility (χ) measurements in this material suggest that the Ir moments have strong antiferromagnetic correlations with a Curie-Weiss temperature $\Theta_W \sim -650$ K. The observation of a large χ and entropy at low temperature indicates that gapless spinful excitations survive for $T \ll \Theta_W$. At the same time, χ and specific heat measurements reveal no signatures of magnetic order or any other symmetry breaking down to $T \sim 0.5$ K, nearly 3 orders of magnitude lower than Θ_W, suggesting that $Na_4Ir_3O_8$ may be the first example of a 3D quantum spin liquid which does not order down to $T = 0$. It joins a small but growing list of recently discovered frustrated $s = 1/2$ quantum magnets [2] which appear to have quantum disordered ground states.

These experiments motivated a study of the classical Heisenberg antiferromagnet on the hyperkagome lattice [3]. This model was found to order into a coplanar "classical nematic" state at low temperatures, $T \lesssim J/1000$, where J is the nearest neighbor antiferromagnetic exchange coupling. However, quantum effects are clearly significant at such low temperatures. A subsequent study of the quantum Heisenberg model, using an Sp(N) mean field theory, uncovered a candidate quantum spin liquid ground state with Z_2 topological order [4]. However, this "bosonic" spin liquid has a nonzero spin gap which is at odds with recent observations, that gapless spin excitations survive down to $T \sim 0.5$ K [5], unless the spin gap is anomalously small. Another difficulty of this proposal is that there should be a finite temperature transition from the Z_2 spin liquid to the higher temperature paramagnetic phase while there is no signature of such a phase transition in thermodynamic measurements [1].

Here we pursue a completely different line of attack and attempt to build a "fermionic" spin liquid theory of the hyperkagome Heisenberg model. This formulation has the virtue that gapless spin liquids emerge as stable phases at mean field level and beyond without any need for fine tuning [6,7]. The main results of our Letter are as follows. (i) We find that of a number of candidate spin liquid ground states we have explored, a particularly simple fermionic spin liquid state, one which supports Fermi surfaces of spinons, emerges as a promising candidate for the ground state of the nearest neighbor Heisenberg model on the hyperkagome lattice. This result is obtained by a combination of exact diagonalization, a projective symmetry group (PSG) analysis [6] of mean field ground states, and Gutzwiller projected variational wave function calculations. (ii) We then show, using a Gutzwiller renormalized mean field theory [8], that the specific heat of this spin liquid state is quite similar to the experimentally observed

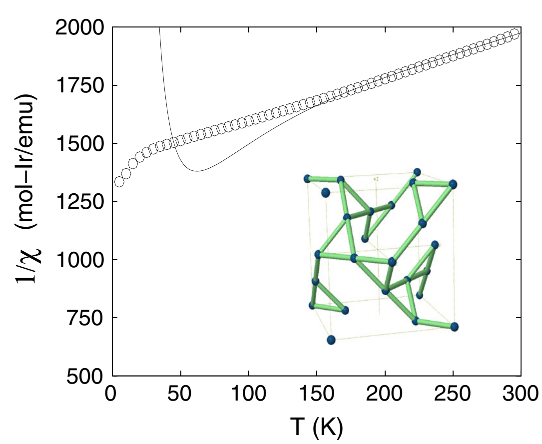

FIG. 1 (color online). Exact diagonalization (ED) results (solid line) for the inverse uniform magnetic susceptibility $1/\chi$ compared with experiments (open circles). The ED was done on a single unit cell of the hyperkagome lattice (inset) with $J = 304$ K chosen to reproduce the high temperature experimental $1/\chi$.

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specific heat of $Na_4Ir_3O_8$ for $T \gtrsim 5$ K. This spinon Fermi surface state therefore seems to be a good starting point to understand the physics of this material over a wide range of temperatures in the same way that Fermi liquid theory is a good starting point to understand conventional metals. However, as in conventional metals, the Fermi surfaces could be unstable, at very low temperature, due to small additional interactions. (iii) We considered a symmetry analysis of possible low temperature instabilities of the spinon Fermi surface state which yields candidate states with line nodes in the spinon dispersion. We discuss implications of these states for the specific heat data.

Model and exact diagonalization.—We begin with an exact diagonalization (ED) study of the nearest neighbor $s = 1/2$ Heisenberg model $H = \frac{1}{2}\sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$, on a single 12-site unit cell of the hyperkagome lattice formed by the Ir sites in $Na_4Ir_3O_8$ (see inset of Fig. 1). Here J_{ij} is the exchange coupling on the bond ij, and we keep only the nearest neighbor antiferromagnetic exchange interaction, $J > 0$. Figure 1 displays the ED result for $\chi^{-1}(T)$ with a choice of $J = 304$ K which, as shown, reproduces the experimental data in the range 150–300 K. Over the limited temperature range $T = 200$–300 K, the $\chi^{-1}(T)$ from ED can be fit by an apparent "Curie-Weiss" law with $\Theta_W \approx -730$ K. [The upturn in $\chi^{-1}(T)$ in the ED for $T \lesssim 50$ K arises from a nonzero spin gap on a single unit cell.] ED calculations of χ^{-1} with next nearest neighbor Heisenberg exchange J' suggest that $|J'|$ is unlikely to be larger than $\sim 0.1J$.

Specific heat experiments, after subtraction of the phonon part to extract the magnetic contribution, find a broad peak in C/T at $T_p \approx 25$ K with a peak height $(C/T)_{\max} \approx 55\ \mathrm{mJ}/K^2/\mathrm{mol\ Ir}$ [1]. While finite size effects are clearly important in the ED calculations at low T, it is nevertheless encouraging that the ED result for C/T of the Heisenberg model with $J = 304$ K, shown in Fig. 3, has a broad peak at $T_p \approx 20$ K with a peak height of $(C/T)_{\max} \approx 70\ \mathrm{mJ}/K^2/\mathrm{mol\ Ir}$.

Since the $s = \frac{1}{2}$ Heisenberg model appears to capture aspects of the experimental data on $Na_4Ir_3O_8$, we turn to an analysis of variational candidates for the ground state of this model in order to understand the emergence of a gapless quantum spin liquid in $Na_4Ir_3O_8$.

Hyperkagome spin liquid states.—We begin by representing spin operators in terms of fermionic spinors, as $\vec{S}_i = \frac{1}{2} f^\dagger_{i\alpha} \vec{\sigma}_{\alpha\beta} f_{i\beta}$, where each site is constrained to have a single fermion, i.e., $f^\dagger_{i\sigma} f_{i\sigma} = 1$. The Hamiltonian H, written in terms of these fermions, can be decoupled in both the hopping and pairing channels at mean field level leading to a mean field Hamiltonian

$$H_{\mathrm{MF}} = \frac{3}{8}\sum_{\langle ij\rangle} J_{ij}\left[\frac{1}{2}\mathrm{Tr}(\mathbf{U}^\dagger_{ij}\mathbf{U}_{ij}) - \Psi^\dagger_i \mathbf{U}_{ij}\Psi_j + \mathrm{H.c.}\right] + \sum_i \vec{a}_i \cdot \Psi^\dagger_i \vec{\tau} \Psi_i, \tag{1}$$

which is given in a manifestly SU(2) invariant form where $\Psi^T_i = (f_{i\uparrow}, f^\dagger_{i\downarrow})^T$ is a Nambu spinor,

$$\mathbf{U}_{ij} = \begin{pmatrix} \chi_{ij} & \Delta_{ij} \\ \Delta^*_{ij} & -\chi^*_{ij} \end{pmatrix} \tag{2}$$

are the mean field hopping and pairing amplitudes, and $\vec{a}_i$ is a Lagrange multiplier which enforces, on average and in an SU(2) invariant manner, the single occupancy constraint $\langle f^\dagger_{i\sigma} f_{i\sigma}\rangle = 1$.

Guided by experiment, we assume that the ground state of H is a spin liquid, and therefore seek variational mean field fermion ground states which preserve all lattice symmetries, global spin-rotation symmetry, and time-reversal symmetry. The identification of such distinct symmetric spin liquid candidates requires a "projective symmetry group" (PSG) analysis [6] that takes into account the space group symmetries of the lattice together with all possible gauge transformations which change the mean field fermion state but leave the spin wave function intact. Under a PSG transformation, spinors transform as $\Psi_i \to \mathbf{G}^X_i \Psi_{X(i)}$ where $\mathbf{G}^X_i$ is an SU(2) gauge transformation associated with the space group transformation X. We require that transforming a mean field ansatz by

$$U_{ij} \to \mathbf{G}^X_i U_{X(i)X(j)} \mathbf{G}^X_j; \qquad \vec{a}_i \cdot \vec{\tau} \to \mathbf{G}^X_i (\vec{a}_{X(i)} \cdot \vec{\tau}) \mathbf{G}^X_i \tag{3}$$

leaves H_{MF} invariant. We have constructed a systematic classification of spin liquid ground states by constructing all PSGs with nontrivial $\mathbf{G}^X_i$ associated with the point group of the hyperkagome lattice. This group turns out to be equivalent to the octahedral group O and consists of twofold rotations about each site, threefold rotations for each triangle and fourfold screw rotations for each thread (see Ref. [4]). Details of our calculation will be presented elsewhere [9].

Here, for simplicity, we focus on the family of states

$$U_{ij} = \chi_{ij}\tau_3 + \Delta_{ij}\tau_1, \qquad \vec{a}_i = -\mu\hat{z}, \tag{4}$$

where χ_{ij} is real and positive, Δ_{ij} is real but alternates sign as discussed below and only the bonds ij that have a finite exchange J_{ij} have finite U_{ij}. This family of states covers most of the states resulting from our PSG analysis. χ_{ij} and Δ_{ij} are chosen to be invariant under translations and threefold rotations through each triangle. However, twofold rotations about each site and the fourfold screw rotations both need to be followed by the gauge transformation $\mathbf{G}^X_i = i\tau_3$, where X is either of these transformations. This second requirement fixes the sign of the pairing fields Δ_{ij}. In addition to these spatial symmetries, we have imposed time-reversal (T) invariance by requiring that a T transformation followed by $\mathbf{G}^T_i$ commute (or anticommute) with the spatial transformations. Since T sends $U_{ij} \to -U_{ij}$, we found $\mathbf{G}^T_i = i\tau_2$ satisfies all requirements. The combination of all these symmetries completely determines the form of U_{ij} in Eq. (4).

It turns out that due to enhanced symmetry in special limits, the ansatz of Eq. (4) describes three different spin liquid states depending on the variational parameter the U (1)-uniform state, the U(1)-staggered state, and the Z_2 state. The U(1) uniform state has $\chi_{ij} > 0$, no pairing ($\Delta_{ij} = 0$), and a U(1) phase invariance. On the other hand, the U(1) staggered state has no hopping $\chi_{ij} = 0$ and finite $\Delta_{ij} \neq 0$ which alternates sign on adjacent triangles. The Z_2 family of states arises when both pairing and hopping are present.

In general, we need not keep the time-reversal symmetry of the ansatz in Eq. (4). If we let $\chi_{ij} = u_{ij}\cos\theta_{ij}$ and $\Delta_{ij} = u_{ij}\sin\theta_{ij}$, so that $\mathrm{sgn}(\theta_{ij}) = \mathrm{sgn}(\Delta_{ij})$, we can extend the ansatz to

$$U_{ij} = iu_{ij}\exp\left\{-i\frac{\nu}{2}\hat{n}_{ij}\cdot\vec{\tau}\right\}, \qquad \vec{a}_i = -\mu\hat{z}, \tag{5}$$

where $\hat{n}_{ij} = \hat{z}\cos\theta_{ij} + \hat{x}\sin\theta_{ij}$. This extended form then has all the same spatial symmetries of Eq. (4) but recovers time-reversal invariance only at $\nu = \pi$. To determine which of these states are viable candidates for the ground state of the Heisenberg model, we next compute the energies of these different states as a function of the angle θ, which is the same up to a sign on all bonds, and ν, with $\nu = \pi$ for the time-reversal invariant states.

Energetics of candidate spin liquid states.—We have computed the ground state energy for the above class of states in mean field theory as well as by a numerical Gutzwiller projection of the mean field states which yields a physical spin wave function. The Gutzwiller projected energy is computed using the variational Monte Carlo (VMC) method [7,10,11].

(i) *U(1)-uniform state:* The mean field ground state energy per spin is $E^{\mathrm{mf}}_{\mathrm{unif}} = -0.144J$. After Gutzwiller projection, we find a variational energy $E^{\mathrm{proj}}_{\mathrm{unif}} \approx -0.424J$, so that $E^{\mathrm{proj}}_{\mathrm{unif}}/E^{\mathrm{mf}}_{\mathrm{unif}} \approx 3$. The energy of the projected state compares favorably with the ED result on a single unit cell, $E^{\mathrm{ed}} = -0.454J$. In the preprojected state, three spinon bands cross the Fermi level. One Fermi surface is electronlike and centered at $\vec{K} = (0, 0, 0)$, while the other two are holelike and centered about $\vec{K} = (\pi, \pi, \pi)$. All three have $k_F \approx 0.2\pi$.

(ii) *U(1)-staggered state:* The mean field energy of this state is $E^{\mathrm{mf}}_{\mathrm{stag}} = -0.122J$. Because of flat bands at the chemical potential in this state, the energy of the projected wave function depends somewhat on our selection of the subset of the flat band states we fill with fermions in the preprojected state. For various choices that we have explored the estimated VMC energy is about $E^{\mathrm{proj}}_{\mathrm{stag}} \sim -0.37J$, significantly higher than the uniform state.

(iii) Z_2 *state*: As seen from Fig. 2, the mean field energy of the Z_2 states parametrized by the variational parameter $\theta = |\theta_{ij}|$ is higher than that of the U(1) uniform state (which corresponds to $\theta = 0$). Even after projection, the U(1) uniform state appears to have the lowest energy,

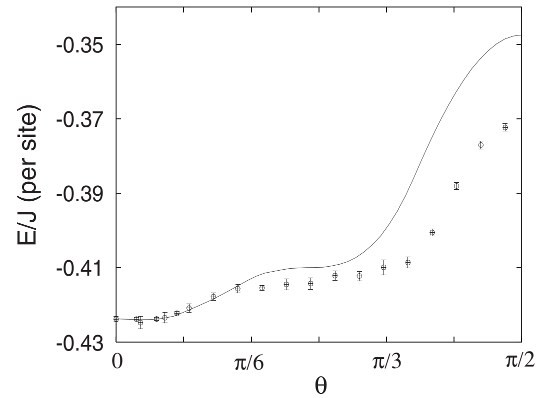

FIG. 2. Energetics of the Z_2 family of states parametrized by θ. Points show VMC data and the solid line is the mean field result multiplied by $g_J \approx 3$. The uniform state ($\theta = 0$) appears to have the lowest energy in this class of states.

although the energy is quite flat as a function of θ for $\theta \lesssim 0.1\pi$ as in the mean field theory.

(iv) *Chiral states*: We have also checked the energetics of the time-reversal symmetry broken chiral U(1) spin liquid ansatz. The uniform U(1) state is stable against such symmetry breaking. The staggered U(1) state energy is lowered by breaking time-reversal symmetry; however, the lowest energy thus obtained, $E^{\mathrm{proj}}_{\mathrm{chir}} \sim -0.39J$, is still higher than the uniform U(1) state energy.

In summary, the U(1) uniform state appears to be the most favorable candidate for the ground state of the nearest neighbor Heisenberg model on the hyperkagome lattice. As seen from Fig. 2, the energy is a rather flat function of θ for small values of $\theta \lesssim 0.1\pi$. Small further neighbor couplings may therefore favor Z_2 states with a small pair amplitude. We discuss this further in the concluding section. We note that spin liquids with spinon Fermi surfaces have also been proposed recently for some quasi-two-dimensional frustrated magnets [11].

Application to the specific heat of $Na_4Ir_3O_8$.—Motivated by our variational ground state calculations we next turn to specific heat of the uniform U(1) state for the nearest neighbor Heisenberg model in order to compare with the data on $Na_4Ir_3O_8$. Since we cannot implement the Gutzwiller projection exactly for computing finite temperature properties, we will resort to renormalized mean field theory (RMFT) [8] which relies on simple renormalization factors to account for the effect of projection. For instance, $\langle\vec{S}_i\cdot\vec{S}_j\rangle_{\mathrm{proj}} = g_J\langle\vec{S}_i\cdot\vec{S}_j\rangle_{\mathrm{mf}}$ defines the renormalization factor g_J for the energy. From our calculations, we find that $E^{\mathrm{proj}}/E^{\mathrm{mf}} \approx 3$ which implies $g_J \approx 3$. We therefore set $g_J = 3$ to renormalize the mean field quasiparticle dispersion and compute the RMFT result for the specific heat of the U(1) uniform state.

As seen from Fig. 3, the computed specific heat is in broad agreement with experiment for $T \gtrsim 5$ K. Remarkably, for 5 K $\lesssim T \lesssim$ 25 K, we find C/T shows a strong, almost linear, T dependence similar to experiment

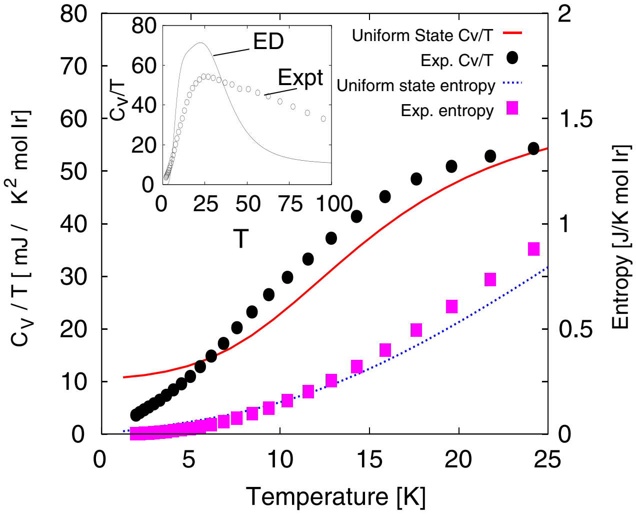

FIG. 3 (color online). Comparison of the specific heat coefficient C/T and entropy $S(T)$ of the U(1) uniform state with measurements on $Na_4Ir_3O_8$ (after phonon subtraction). The inset compares the phonon subtracted C/T with the ED result for $J = 304$ K.

arising simply from the energy dependence of the spinon density of states in the uniform U(1) state. The entropy of this state is also in reasonable agreement with the higher temperature data as seen from Fig. 3. We emphasize that these are zero-parameter fits which thus serve as nontrivial tests of our proposal. For $T \lesssim 5$ K, the spinon Fermi surfaces lead to a saturation of C/T with a small $\gamma \approx 10$ mJ/K^2/mol Ir. A more precise estimate of γ requires projection of excited states—this is complicated due to the many bands and was not attempted.

Discussion.—We have shown, based on mean field theory and projected wave function studies, that the U(1) uniform state which supports three spinon Fermi surfaces is a viable candidate for the ground state of the $s = \frac{1}{2}$ hyperkagome Heisenberg model. This state provides a reasonable description of the specific heat of $Na_4Ir_3O_8$ over a broad temperature range $T \gtrsim 5$ K. Such a Fermi surface state would have a constant low temperature spin susceptibility. Knowledge of the spinon Fermi surfaces of the mean field Hamiltonian also allows us to construct the (gauge invariant) wave vectors connecting different points on these Fermi surfaces which determines the wave vector dependence of triplet excitations.

At lower temperatures $T \ll 5$ K, experiments may be consistent with $C/T \sim T$, suggesting that the spinon Fermi surface could be unstable at low T due to various smaller exchange couplings which we have ignored here. For instance, small further neighbor interactions $J' \sim \pm 0.1J$ would tend to favor weak next neighbor pairing terms. We have checked that such a pairing term which transforms nontrivially under point group operations can gap out most of the spinon Fermi surface leading to a Z_2 state which has line nodes where the [110] plane (and symmetry related planes) intersect the spinon Fermi surfaces. Such a line-node state would lead to $C/T \sim T$ at sufficiently low T. Any Z_2 spin liquid state would, however, likely undergo a phase transition to the high T paramagnetic phase. No such transition was observed in experiment for $T \gtrsim 5$ K; however we cannot rule out such a transition at lower T.

Finally, turning to the very low temperature behavior of χ, it has been recently argued that the experimental observation of a constant $\chi(T \to 0)$ cannot be reconciled with a specific heat $C/T \sim T$ as $T \to 0$ unless spin-orbit interactions are taken into account [12]. It was shown that, despite rather strong atomic spin-orbit coupling on Ir, the effective spin model is likely still of Heisenberg type with spin-orbit induced Dzyaloshinskii-Moriya (DM) corrections. For small DM (relative to J), our results for the energetics and specific heat would remain unchanged. So the intermediate T state could still be a spinon Fermi surface state, with a possible low T instability into a line-node state as discussed. The DM coupling would, however, strongly modify the susceptibility of such a line-node state, which is naïvely expected to behave as $\chi(T \to 0) \sim T$, and could bring it into better agreement with experiment. The clarification of these issues is a promising direction for future research.

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Note Added.—During the final stages of the preparation of this manuscript, we received a preprint [13] which discusses related issues.

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