Gapless spin liquids on the three dimensional hyper-kagome lattice of Na$_4$Ir$_3$O$_8$

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Recent experiments indicate that Na$_4$Ir$_3$O$_8$, a material in which $s$=1/2 Ir local moments form a three dimensional network 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 wavefunction studies, we propose a quantum spin liquid with spinon Fermi surfaces as a favorable candidate for the ground state of the Heisenberg model on the hyper-kagome lattice of Na$_4$Ir$_3$O$_8$. We present a renormalized mean field theory of the specific heat of this spin liquid and also discuss possible low temperature instabilities of the spinon Fermi surfaces.

**Introduction:** Na$_4$Ir$_3$O$_8$ 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 ‘hyper-kagome’ lattice [1], a cubic lattice whose unit cell is shown in Fig. 1. High temperature magnetic susceptibility ($\chi$) 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 $\chi$ and entropy at low temperature indicates that gapless spinful excitations survive for $T \ll \Theta_W$. At the same time, $\chi$ and specific heat measurements reveal no sign of any magnetic order or any other symmetry breaking down to $T \sim 0.5 K$, nearly three orders of magnitude lower than $\Theta_W$, suggesting that Na$_4$Ir$_3$O$_8$ 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 possibly supporting fractionalized excitations.

These experiments motivated a study of the classical Heisenberg antiferromagnet on the hyper-kagome 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 as well as an interesting magnetically ordered state which is proximate to this spin liquid [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 $T \sim 0.5 K$ [3], 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 clear signature of 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 hyper-kagome 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 paper 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 hyper-kagome 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 wavefunction 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 specific heat of Na$_4$Ir$_3$O$_8$ 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. Some of the states resulting from this analysis support line nodes for spinon excitations. We discuss implications of such line node states for the specific heat data.

Model and exact diagonalization: We begin with an exact diagonalization (ED) study of the nearest neighbor Model and exact diagonalization: implications of such line node states for the specific heat analysis support line nodes for spinon excitations. We discuss possible low temperature instabilities of the spinon Fermi interactions. (iii) We considered a symmetry analysis of in conventional metals, the Fermi surfaces could be un-

$H = \frac{1}{2} \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j,$

on a single 12-site unit cell of the hyper-kagome lattice formed by the Ir sites in Na$_4$Ir$_3$O$_8$ (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, $J > 0$. Fig. 1 displays the ED result for $\chi^{-1}(T)$ with a choice of $J = 304K$ which, as shown, reproduces the experimental data at high temperatures. We find that for $T = 200–300K$, the $\chi^{-1}(T)$ from ED can be fit by a “Curie-Weiss” law with $\Theta_W \approx -730K$. This is only an apparent Curie-Weiss behavior in the sense that it is only valid for a limited range of temperature around $J$ and not for $T \gg J$ where one recovers the usual Curie-Weiss law with $\Theta_W \approx -zJ/4$, with the coordination number $z = 4$ on the hyper-kagome lattice. The upturn in $\chi^{-1}(T)$ in the ED for $T \lesssim 50K$ arises from a nonzero spin gap on a single unit cell.

Experiments observe a broad peak in $C/T$ at $T_p \approx 25K$ with a peak height $(C/T)_{max} \approx 55mJ/K^2/mol-Ir$ [1]. While finite size effects are clearly important in the ED calculations at low temperatures, it is nevertheless encouraging that the ED result for $C/T$ of the Heisenberg model with $J = 304K$ shows a broad peak at $T_p \approx 20K$ with a peak height of $(C/T)_{max} \approx 70mJ/K^2/mol-Ir$.

Since the $s = 1/2$ Heisenberg model appears to capture some aspects of the experimental data on Na$_4$Ir$_3$O$_8$, we now turn to an analysis of possible fermionic spin liquid candidates for the ground state of this model in order to see if we can understand the emergence of a gapless spin liquid ground state in Na$_4$Ir$_3$O$_8$.

Hyper-kagome spin liquid states: We begin by representing spin operators in terms of fermionic spinors with the constraint of one fermion per site. The Hamiltonian $H$ can then be decoupled in both the hopping (Hartree-Fock) and pairing (Bogoliubov) channels within a mean field approximation. This decoupling leads to the mean field Hamiltonian

$$H_{MF} = \frac{3}{8} \sum_{ij} J_{ij} \left[ \frac{1}{2} \text{Tr}(U^\dagger_{ij} U_{ij}) - \Psi_i^\dagger \cdot \Psi_j + h.c. \right]$$

written in a manifestly SU(2) invariant form where $\Psi_i^T = (f_{i\uparrow}, f_{i\downarrow})^T$ is a Nambu spinor,

$$U_{ij} = \left( \frac{\chi_{ij}}{\Delta_{ij}} \right)$$

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

We seek the ground state of $H$. Guided by experiment, assume the ground state is a spin liquid, that it does not break any lattice symmetries, global spin-rotation symmetry or time-reversal symmetry. Remarkably, even aside from the known SU(2) gauge redundancy, imposing all these symmetries on the mean field ground state does not single out a unique choice for $U_{ij}$ and $\bar{\alpha}_i$. The identification of non-redundant spin liquid phases, therefore, requires careful examinations of the combined space group and gauge transformations. These fundamental symmetry relations, proposed by Wen [2] and dubbed “projective symmetry group (PSG)” can be used to classify all possible symmetric spin liquid phases. Under a PSG transformation, spinors transform as $\Psi_i \rightarrow G^X_i \cdot \Psi_{X(i)}$ where $G^X_i$ is an SU(2) gauge transformation associated with the space group transformation $X$. Transforming a mean field ansatz by:

$$U_{ij} \rightarrow G^X_i \cdot U_{X(i)X(j)} G^X_j, \quad \bar{\alpha}_i \rightarrow G^X_i (\bar{\alpha}_{X(i)} \cdot \tau) G^X_i$$

then leaves $H_{MF}$ invariant. Naturally, these PSGs are subgroups of the combined space and gauge groups, which leads to strong constraints on the possible choices of $G^X_i$.

We have constructed a systematic classification of spin liquid ground states by constructing all PSGs with non-trivial $G^X_i$ associated with the point group of the hyper-kagome lattice. This group turns out to be equivalent to the octahedral group $O$ and consists of two-fold rotations about each site, three-fold rotations for each triangle and four-fold screw rotations for each thread (see Ref. [4] for a discussion of threads). Details of our calculation will be presented in a future publication [11]. Here, for simplicity, we focus on the family of states

$$U_{ij} = \chi_{ij} \tau_3 + \Delta_{ij} \tau_1, \quad \bar{\alpha}_i = -\mu \hat{z}$$

where $\chi_{ij}$ is real and positive, $\Delta_{ij}$ 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.

From a symmetry perspective, $\chi_{ij}$ and $\Delta_{ij}$ are chosen to be invariant under translations and three-fold rotations through each triangle. However, two-fold rotations about each site and the four-fold screw rotations both need to be followed by the gauge transformation $G^X_{ij} = \tau_X$, where $X$ is either of these transformations. This second requirement fixes the sign of the pairing fields $\Delta_{ij}$. In addition to these spatial symmetries, we have imposed time reversal (T) invariance by requiring that a T transformation followed by $G^T_{ij}$ commute (or anticommute) with the spatial transformations. Since T sends $U_{ij} \rightarrow -U_{ij}$, we found $G^T_{ij} = i\tau_2$ satisfies all requirements. The combination of all these symmetries completely determines the form of $U_{ij}$ in Eq. ([3]).

It turns out that due to enhanced symmetry in special limits, the ansatz of Eq. ([3]) describes three different spin liquid states: 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. In this state, the U(1) phase invariance can be understood after noting that an SU(2) gauge transformation can rotate this pure pairing state into a pure hopping state (with hopping $\chi_{ij} = \Delta_{ij}$). Lastly, the Z$_2$ state arises when both pairing and hopping are present.

In general, we need not keep the time reversal symmetry of the ansatz in Eq. ([3]). If we let $\chi_{ij} = u_{ij} \cos \theta_{ij}$ and $\Delta_{ij} = u_{ij} \sin \theta_{ij}$, so that $\text{sign}(\theta_{ij}) = \text{sign}(\Delta_{ij})$, we can extend the ansatz to

$$U_{ij} = iu_{ij} \exp\{ -i \frac{\theta_{ij}}{\tau} \hat{n}_{ij} \cdot \tau \}, \quad \hat{a}_i = -\mu \hat{\zeta}$$

where $\hat{n}_{ij} = \hat{\zeta} \cos \theta_{ij} + \hat{\zeta} \sin \theta_{ij}$. This extended form then has all the same spatial symmetries of Eq. ([3]) but recovers time reversal invariance only at $\nu = \pi$.

Having discussed the fermionic projective symmetry group approach to construct a set of distinct spin liquid states on the hyper-kagome lattice we would like to know which of them are viable candidates for the ground state of the Heisenberg model. In order to address this issue, we compare the energies of these different 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 wavefunction. The Gutzwiller projected energy is computed using the variational Monte Carlo (VMC) method [3].

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

- **U(1)-staggered state**: The mean field energy of this state is $E_{\text{stag}}^{\text{mf}} = -0.122J$. Due to flat bands at the chemical potential in this state, the energy of the projected wavefunction 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 $E_{\text{stag}}^{\text{proj}} \approx -0.37J$, significantly higher than the uniform state.

- **Z$_2$ state**: As seen from Fig. 2 the mean field energy of the Z$_2$ states parametrized by $\theta = |\theta_{ij}|$ 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, although the energy is quite flat as a function of $\theta$ for $\theta \lesssim 0.1\pi$ as in the mean field theory. The projected energy differs from the mean field value by about a factor of three over a wide range of $\theta$.

- **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. However, the staggered U(1) state energy is lowered by breaking time reversal symmetry. Nevertheless, the lowest energy obtained in this manner, $E_{\text{chir}}^{\text{proj}} \approx -0.39J$, 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 hyper-kagome lattice. However, as seen from Fig. 2 the energy is a rather flat function of $\theta$ for small values of $\theta \lesssim 0.1\pi$. We therefore cannot rule out the possibility that further neighbor couplings or an extended variational ansatz with more
variational parameters will not favor such a $Z_2$ state with a small pair amplitude. We discuss this further in our concluding section.

Application to the specific heat of Na$_4$Ir$_3$O$_8$: Motivated by our variational ground state calculations we next turn to specific heat of the uniform U(1) state in order to compare with the data on Na$_4$Ir$_3$O$_8$. Since we cannot implement the Gutzwiller projection exactly for computing finite temperature properties in any simple manner, we will use a renormalized mean field theory (RMFT) [8] to make progress. The RMFT analysis suggests that a part of the effect of projection can be taken into account on top of the mean field theory by Gutzwiller renormalization factors. For instance, $\langle \vec{S}_i \cdot \vec{S}_j \rangle_{\text{proj}} = g_J \langle \vec{S}_i \cdot \vec{S}_j \rangle_{\text{mf}}$ defines the renormalization factor $g_J$ for the energy. This renormalizes the mean field spinon bandwidth. From our calculations, we find that $E_{\text{proj}}/E_{\text{mf}} \approx 3$ which implies $g_J \approx 3$.

We next set $g_J = 3$ and compute the quasiparticle contribution to the specific heat of the U(1) uniform state in the RMFT. Fig. 3 shows the heat capacity computed this way. We emphasize that this is a zero-parameter fit. As seen, the overall behavior of $C/T$ is in broad agreement with the data. Remarkably, for $5K \lesssim T \lesssim 25K$, we find $C/T$ shows a strong, almost linear, $T$-dependence similar to experiment arising simply from the spinon dispersion in the uniform U(1) state. However, as expected, the computed $C/T$ eventually saturates below about $5K$ due to the spinon Fermi surfaces, leading to a nonzero $\gamma \approx 10$ mJ/K$^2$/mol-Ir. A more precise estimate of $\gamma$ requires the projection of excited states — this is numerically complicated due to the many bands in this system and was not attempted in this work. We emphasize that the broad agreement with experimental data already provides a nontrivial check of our theory. Integrating $C/T$ we also present a comparison between the entropy of this state which also shows reasonable agreement with the higher temperature data.

Discussion: We have argued, based on mean field theory and projected wavefunction studies, that the U(1) uniform state which supports three spinon Fermi surfaces is an energetically viable candidate for the ground state of the $s=1/2$ hyper-kagome Heisenberg model. We have shown that it provides a reasonable overall description of the specific heat of Na$_4$Ir$_3$O$_8$ over a broad temperature range $T \gtrsim 5K$. Such spin liquids with spinon Fermi surfaces have also been proposed recently for some quasi-two-dimensional frustrated magnets [10]. The spinon Fermi surfaces have direct physical implications for the low energy spin excitations. Specifically, from the mean field Hamiltonian, we can construct the gauge-invariant wavevectors connecting the surfaces, which dictate the wavevector dependence of triplet $s = 1$ states.

At lower temperatures, there could be Fermi surface instabilities upon the inclusion of various small perturbations. For example, our analysis above did not include the effect of possible further neighbor interactions (small next neighbor antiferromagnetic exchange $J' \sim 0.1$–0.2$J$ cannot be ruled out from our diagonalization study). We have checked that extending our $Z_2$ ansatz to include next neighbor pairing terms, which would arise from further neighbor interactions in a mean field theory, leads to an unconventional pairing state with line nodes. These line nodes exist where the [110] plane (and symmetry related planes) intersect the spinon Fermi surfaces. Such a line-node state would lead to a low temperature specific heat $C \sim T^2$. As we mentioned, however, any $Z_2$ spin liquid state would most likely undergo a phase transition to the higher temperature paramagnetic phase, which was not observed in the experiment above 0.5$K$. Moreover, from Fig. 3 it seems that one need not appeal to such a paired state to explain the linear-$T$ behavior of $C/T$ between $5K - 20K$. If such a transition does exist, it is likely to occur at much lower temperatures.

Finally, it has been recently argued that the weak temperature dependence of $\chi$ at low $T$ and the specific heat $C \sim T^2$ cannot be reconciled unless spin orbit interactions are present [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 the dominant effect of spin-orbit induced Dzyaloshinskii-Moriya (DM) corrections. For sufficiently small DM (relative to $J$), our results for the energetics and specific heat would remain unchanged. However, the DM coupling can strongly effect the spin susceptibility, especially in the line-node state. Such effects could potentially bring the susceptibility of the line-node state, which naively behaves as $\chi(T) \sim T$, into better agreement with experiment. The clarification of these issues is a promising direction for future research.

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