Ground state of the spin-1 antiferromagnet on the kagome lattice

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We study the phase diagram of the spin-1 quantum antiferromagnet on the kagome lattice with nearest neighbor bilinear and biquadratic interactions using exact diagonalization (ED) and the density matrix renormalization group (DMRG) algorithm. Our results indicate that at the Heisenberg point, the ground state is trimerized, in contrast to previous proposals.

The discovery of experimental realizations of kagome antiferromagnets [1, 2] and indications that they have exotic ground states has spurred immense theoretical activity in the last few years. Even for the simplest realistic model i.e. the nearest neighbor spin 1/2 Heisenberg antiferromagnet, the nature of the ground state has remained a contentious issue for the last three decades [3–10]. However, with advances in numerical algorithms, recent progress has been made in the study of this system [10–15], though several open issues remain.

In contrast to the spin 1/2 case, little has been definitively established for the ground state of the spin > 1/2 case, even though several realizations exist for these systems [16]. For example, KV\textsubscript{2}Ge\textsubscript{2}O\textsubscript{6} [17] and BaNi\textsubscript{3}(OH)\textsubscript{2}(VO\textsubscript{3})\textsubscript{2} [18] have been recently reported to be candidates for spin-1 kagome and the chromium-jarosite (KCr\textsubscript{3}(OH)\textsubscript{6}(SO\textsubscript{4})\textsubscript{2}) has been found to be a spin 3/2 kagome antiferromagnet [19]. While it is known that the spin 5/2 compound KFe\textsubscript{3}(OH)\textsubscript{6} has magnetization plateaus close to a spin-1 KHAF [19], the situation for intermediate spins: spin 1 [20–22] and spin 3/2 [23] has not been so clear. Previous numerical studies of the spin-1 XXZ model [24, 25] have shed light on the phase diagram of the spin-1 kagome antiferromagnet, but the approach is limited around the Heisenberg point (owing to the Quantum Monte Carlo "sign problem"). A recent coupled cluster calculation by Götze et al. [22] has shown that the spin 1 kagome Heisenberg antiferromagnet (henceforth abbreviated as KHAF) has no long range antiferromagnetic order, in contrast to previous analytic results [21].

Based on exact diagonalization of the spin-1 KHAF, Hida proposed that the ground state is a Hexagonal Singlet Solid (HSS) with a sizeable spin gap [20]. The HSS state is a translationally invariant state that is described by an AKLT [26] type wavefunction: all the spin ones fractionalize into two translationally invariant state that is described by an AKLT [26]. The HSS has a competitive energy (\(\sim -1.36\) per site) in comparison to the DMRG results (\(\sim -1.41\) per site), it is not the correct qualitative picture of the ground state. Instead, we find that the ground state is trimerized (see Fig. 1(a)). Our calculations also show that the ground state has no long range spin-spin correlations and has a finite spin gap of \(\sim 0.28J\) and non-magnetic gap of \(\sim 0.1\) to 0.2J. Based on energetics, we suggest that it is unlikely that a recently proposed \(Z_2\) spin liquid [31] is the ground state of this system as its reported energy is higher than both the HSS and the trimerized state found in DMRG.

In order to validate this finding, we have considered additional nearest neighbor biquadratic interactions (given by coupling strength \(J_{bq}\)) and thus the Hamiltonian we study is,

\[
H = J_{bl} \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_{bq} \sum_{\langle ij \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j)^2
\]

where \(\langle ij \rangle\) refers to nearest neighbor pairs and \(J_{bl} > 0\) is the Heisenberg coupling, which we will set to \(J_{bl} = 1\). It has been previously shown [32] that \(J_{bl} = J_{bq}\) leads to the ground state being a simplex solid [33], a symmetry broken state where the three spin ones living on each up (or equivalently down) pointing triangles trimerize into singlets as shown in Fig. 1(a). However, it is not clear if this trimerization will survive on reducing the magnitude of \(J_{bq}\) all the way to zero. Here we provide evidence in favor of the trimerization picture and observe a quantum phase transition to a ferroquadrupolar spin nematic only around \(J_{bq} \sim -0.16\). This calls for a theoretical review of this picture.

In this Letter, we shed further light on the nature of the ground state of the idealized spin-1 KHAF. Using Exact Diagonalization for clusters of sizes up to 21 sites and the Density Matrix Renormalization Group (DMRG) algorithm [29] for cylindrical geometries [30], we show that even though the HSS has a competitive energy (\(\sim -1.36\) per site) in comparison to the DMRG results (\(\sim -1.41\) per site), it is not the correct qualitative picture of the ground state. Instead, we find that the ground state is trimerized (see Fig. 1(a)). Our calculations also show that the ground state has no long range spin-spin correlations and has a finite spin gap of \(\sim 0.28J\) and non-magnetic gap of \(\sim 0.1\) to 0.2J. Based on energetics, we suggest that it is unlikely that a recently proposed \(Z_2\) spin liquid [31] is the ground state of this system as its reported energy is higher than both the HSS and the trimerized state found in DMRG.

The Heisenberg point— We begin by considering \(J_{bq} = 0\) i.e. the Heisenberg point and assess the quality of the HSS with respect to exact calculations. Following Hida [20], we associate two spin 1/2 degrees of freedom with each spin 1, \(|+1\rangle \equiv \frac{\psi_{1/2,1/2}}{\sqrt{2}}|0\rangle \equiv \psi_{1/2,-1/2}\) \n and have \(|1\rangle \equiv \psi_{-1/2,-1/2}\) \n where \(\psi_{\alpha,\beta} \equiv \frac{1}{\sqrt{2}}(\psi_{\alpha}\psi_{\beta} + \psi_{\beta}\psi_{\alpha})\) where \(\alpha, \beta\) correspond to spin half degrees of freedom and have value \(\pm 1/2\). Then the HSS wavefunction is defined to be,

\[
\phi_{\text{HSS}} = \otimes_{\alpha,\beta} \prod_{\gamma} (\delta_{\alpha,\gamma} + \delta_{\beta,\gamma}) \int dq_{\gamma} dq_{\gamma',l_{p},l_{p}',n_{p}} \langle q_{\gamma} | q_{\gamma'} l_{p} l_{p}' n_{p} \rangle
\]
where \( p \) is a label used to distinguish the hexagons and \( i_p \) through \( n_p \) refer to the sites on the elementary hexagon (in contiguous order) and \( \gamma_{i_p} \) through \( \gamma_{n_p} \) correspond to the spin 1/2 degrees on those sites. \( \gamma_{i_p} \gamma_{j_p} \gamma_{k_p} \gamma_{l_p} \gamma_{m_p} \gamma_{n_p} \) is the coefficient of the lowest energy singlet state of a spin 1/2 Heisenberg model on a hexagon.

Table I shows the energy of the HSS, the Resonating AKLT state (a recently proposed spin liquid \([31]\)) and exact wavefunctions. \([34]\). It is difficult to calculate the value of the HSS energy on much larger lattices, even by Monte Carlo sampling, as the wavefunction becomes exponentially hard to compute. Despite this limitation, we estimate, based on the trends seen in Table I and using the size extensivity of the HSS state, the energy to be around -1.36 per site in the thermodynamic limit. This is comparable to the exact energy of \( \approx -1.4 \).

In order to develop understanding of the ground state of the spin-1 KHAF for larger systems, we employed density matrix renormalization group (DMRG) calculations. As has been shown in Figure 1(c), our choice of geometries for simulating this system is cylinders with periodic boundary conditions in the short (width) direction and open boundary conditions in the long (length) direction. In order to have complete hexagons we choose widths which are always even.

DMRG calculations were performed with the number of renormalized states (denoted by \( m \)) which were typically 2000, 3000 and 4000 for widths 4, 6 and 8 respectively. On cylinders with widths 4 and 6, and odd lengths (these have equal number of up and down pointing triangles), one sees a pattern of strong and weak trimers propagate from the left and right edges. In the bulk (i.e. near the center of the sample), the bond energies are relatively uniform i.e. translational invariance is restored due to the superposition of the two competing trimer patterns. On the even length cylinders (with an excess of down triangles over up triangles), there is dimerization at the left end, effectively decoupling it from the rest of the system. Thus, the even length cylinders have similar bulk properties as the odd length cylinders.

While there appear to be appreciable finite size effects for the smaller cylinders, a robust and relatively uniform trimerization pattern is observed throughout the bulk for width 8 cylinders. For the odd lengths too, DMRG breaks the symmetry between the up and down pointing triangles, which we take to be evidence that the system prefers to trimerize. (Note that this is a "finite \( m \)" effect, as in any exact calculation for the odd length cylinders, one should obtain a superposition of both trimer states).

To get an estimate of the energy per bond (and in turn the energy per site) in the thermodynamic limit we tried two different fitting procedures. In the first method, we consider the entire recorded energy and fit to

\[
E/N = E_0 + a/L + b/L^2
\]  

In the second method, we sum up the bond energies only on a central feature (such as the bowtie or "star" consisting of three up and three down triangles). This is what we refer to as "bulk energy". For every cylinder width, we considered various lengths and extrapolated the energy of the bulk of the system to infinite length, either analysis gave us similar estimates. Both of these measures have been plotted in Fig. 2.

For the width 4, 6 and 8 cylinders the energy per bond is \(-0.7117(1), -0.7067(1)\) and \(-0.7058(4)\). The energy varies very slightly with width and thus an estimate the energy per bond is \( \approx -0.706(1) \), which in terms of the energy per site is \(-1.412(2)\). This estimate of the energy is comparable to (and slightly lower than) the extrapolated coupled cluster re-
Figure 2. (Color online): (a) Ground state energy per bond for cylinders of odd lengths and different widths. The total energy per bond is extrapolated to infinite length and the fit is shown by solid lines. The bulk energy is also estimated (see text) and plotted in the figure. (b) shows the spin gap for various cylinder widths and lengths.

Results [22] ($E_0 = -1.4031$) [35].

Next, we also verified the presence of a spin gap in the thermodynamic limit (see Fig. 2(b)). We checked that the excitation is a bulk one by observing the magnetization profile of the $S_z = 1$ ground state. The gap is estimated to be roughly 0.28$J$ and there appears to be some variance depending on the aspect ratio.

Since the KHAF is gapped, one can ask whether there is a Hamiltonian in its vicinity, which is also gapped but perhaps easier to analyze (i.e. its order parameter is easier to establish analytically or detect numerically). If there are no energy crossings while tuning the Hamiltonian towards the Heisenberg point, one can be reasonably sure that the physics at the two points is qualitatively similar.

For example, Cai et al. [36] introduced additional second-nearest and third-nearest neighbor exchange couplings on the kagome lattice, and analytically showed the existence of a set of parameters that give a trimerized ground state [37]. We considered an alternate Hamiltonian, the bilinear-biquadratic (BLBQ) model (see equation (1)). For this model, it has been previously shown (via exact diagonalization and tensor network state calculations [32]) at the $SU(3)$ point ($J_{bl} = J_{bq} = 1$) that the kagome lattice trimerizes. Thus, we can use $J_{bq}$ as a knob to connect the Heisenberg point to the $SU(3)$ point.

The bilinear-biquadratic (BLBQ) model— We performed an exact diagonalization calculation on a 21 site sample with periodic boundary conditions (see Supplemental Material). Such a calculation gives us access to multiple low energy excited states which have been plotted in Figure 3. As this figure shows, we see no energy crossings on tuning $J_{bq}$ from 1 towards 0. However, at $J_{bq} \approx -0.16$, we see a crowding of energies and a relatively small finite size gap indicative of a quantum phase transition.

In order to strengthen our interpretation, we also look for signatures of possible transitions with the "fidelity metric" [38], defined to be the overlap of two wavefunctions $F \equiv \langle \psi_1 | \psi_2 \rangle$. We have computed the wavefunctions for the 12 and 21 site clusters for $J_{bq}/J_{bl}$ in the range of (-0.5 to +1.0).

In Figure 4, we show fidelities with respect to the ground state wavefunction of the Heisenberg model and that of the $SU(3)$ model. While it is known that the fidelity alone can not solely detect phase transitions reliably (especially those that are second or higher order), the absence of any unusual features in the range of $J_{bq}/J_{bl}$ from 0 to 1, suggests that the Heisenberg model is trimerized.

The observations and inferences from exact diagonalization can be verified on a larger sample using DMRG, by consider-
but still breaks the spin rotational symmetry. This is verified by the observation that $\langle S_i^+ S_i^- \rangle \neq (\langle S_i^z \rangle)^2$ and that $\langle (S_i^z)^2 \rangle$ abruptly changes from 0.66 (= 2/3) to around 0.4 at the critical point.

**Conclusion**—We have performed ED and DMRG calculations on the spin 1 antiferromagnet with bilinear and biquadratic terms. We have found that the kagome lattice is trimerized at the Heisenberg point, which is not consistent with the hexagonal-singlet state (HSS) picture [20], nor with the $\sqrt{3} \times \sqrt{3}$ order predicted by 1/S methods [21]. However, there is no long range magnetic order, consistent with recent coupled cluster calculations [22].

We have provided evidence that the Heisenberg model is trimerized and have obtained the approximate location of the phase transition from the trimerized state to the spin nematic phase, which we estimate to be around $J_{bq}/J = -0.16$. Since the trimerization order parameter is small: adding sufficiently strong longer range interactions at the Heisenberg point may destabilize this order.

Recently, Li et al. [31] have proposed a spin liquid ground state for the spin-1 KHAF. The state is a uniform superposition of all possible “AKLT-loops” (consider a covering of the lattice by loops and perform the AKLT construction along each such loop) and hence has been called the Resonating AKLT state (RAL). On an 18 site lattice, the RAL has an energy marginally lower than the HSS, but its energy in the infinite lattice limit is significantly higher [31]. A plausible reason for this effect is that the RAL is dominated by long loops that are still relatively short on an 18 site lattice. Presumably, if the longest loops in the RAL are penalized (i.e. a loop tension is added in the wavefunction), the energy could improve significantly. However, it is not clear if such a modification of the wavefunction preserves its spin liquid properties, or whether it is driven to a confining phase (such as the trimerized phase). In summary, it will be interesting to see if exotic phases such as the HSS or RAL can be stabilized in some part of Hamiltonian parameter space.

Finally, we comment on the possible experimental consequences of our finding. Since trimerization does not change the magnetic unit cell structure of the kagome lattice, we still expect to see the 1/3 magnetization plateau in an ideal kagome lattice (based on the Oshikawa-Yamanaka-Affleck criterion [40]). However prominent magnetization plateaus seen in the experiment with m-MPYNN-BF$_4$ (which also has a slight $\sqrt{3} \times \sqrt{3}$ distortion) [27] correspond to 1/2 and 3/4 which is indicative of a magnetic unit cell with 12 atoms. Thus, we intend to understand the effective low energy Hamiltonian better to resolve this issue.

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Supplemental Material: Ground state of the spin-1 antiferromagnet on the kagome lattice

The kagome geometries used for exact diagonalization (referenced in the main text) are shown here. Periodic boundary conditions are assumed.

Figure S1. Kagome geometries used for exact diagonalization.