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Numerical linked-cluster expansion for the distorted kagome lattice Heisenberg model

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Abstract. Motivated by experimental results for the thermodynamic properties of the $\text{Rb}_2\text{Cu}_3\text{SnF}_{12}$ material and the discovery of its valence-bond solid ground state, we utilize the numerical linked-cluster expansions (NLCEs) and devise an expansion tailored to solve the Heisenberg model on a pinwheel-distorted kagome lattice. Using the exchange interactions that are relevant to $\text{Rb}_2\text{Cu}_3\text{SnF}_{12}$, we calculate its uniform spin susceptibility and find a very good agreement with experiment. Next, we focus on the ground state of a simplified model of the distorted kagome lattice and take advantage of a zero-temperature Lanczos-based NLCE to study the approach to the ground state of the uniform kagome lattice Heisenberg model using a tuning parameter. Together with results from exact diagonalization of finite clusters, we find evidence that a phase transition occurs before the uniform limit is reached.

1. Introduction

Frustrated magnets are systems in which the combination of lattice geometry and the nature of interactions between spins prevent simultaneous minimization of the energy on all the bonds for any arrangement of spins (in a classical picture). Examples of these are the antiferromagnetic (AF) spin models on the kagome, triangular, checkerboard and pyrochlore lattices. They all share a common feature. That is, they consist of frustrating plaquettes (triangles or tetrahedra). In the (classical) Ising model, frustration results in an exponentially large ground state degeneracy and thus, a finite zero-temperature entropy [1, 2]. In the quantum mechanical models, such as the AF Heisenberg model, the large number of low lying states often leads to unusual low-temperature properties and exotic ground states, such as the resonating valence bond (RVB) spin-liquids [3, 4, 5, 6, 7, 8, 9].

Among different frustrated geometries, the kagome lattice Heisenberg model (KLHM) is of particular interest since, according to numerical calculations, it has energy gaps that are an order of magnitude smaller than for the other geometries and exponentially large number of singlet states below the lowest triplet state [10, 11, 4]. On the other hand, experiments on materials with almost perfectly decoupled kagome layers, such as Herbertsmithite, show no magnetic order down to very low temperatures and no evidence of a spin gap [12, 13, 14, 15]. However, when studying these materials, one has to be careful about the role of magnetic impurities and Dzyaloshinskii-Moria anisotropy, which might ultimately determine the nature of the ground state [16, 17, 18].

Numerical studies have played an important role since the beginning of the search for the nature of the ground of the KLHM. Most recently, a large-scale density matrix renormalization

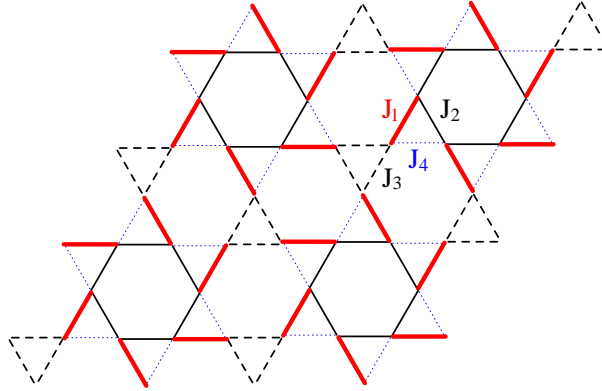


Figure 1. The anisotropic kagome lattice, representing the model for $\text{Rb}_2\text{Cu}_3\text{SnF}_{12}$ with four different exchange interactions, $J_1 \dots J_4$.

group (DMRG) study by Yan *et al.* [9], suggested that the ground state of this model in the thermodynamic limit is a fully gapped singlet. This is despite the fact that previous series expansion studies had found indications of the 36-site unit cell valence-bond solid (VBS) being the ground state [6, 7]. By introducing a parameter, λ , which takes one from the distorted kagome to the uniform lattice, they predicted that such dimer order is picked up according to the perturbative expansion of the energies in powers of λ . However, in this scheme, other VBS orders can, in principle, be energetically favored in higher orders of λ .

In this work, we first focus on the finite-temperature properties of the material $\text{Rb}_2\text{Cu}_3\text{SnF}_{12}$, which is known to have distorted kagome layers and a gapped valence-bond solid (VBS) ground state with a 12-site unit cell [19, 20]. We report on the results from a numerical linked-cluster expansion (NLCE) [21, 22] that is designed to capture not only the high temperature properties of this material in the thermodynamic limit, but also the low-temperature physics associated with its VBS order. The advantage of the NLCE used here, over the regular series expansions, is that in NLCE the properties of linked-clusters in the series are calculated exactly, as opposed to perturbatively, using exact diagonalization techniques. We find that using the experimental values for the four different exchange interactions in the model for $\text{Rb}_2\text{Cu}_3\text{SnF}_{12}$ (shown in Figure 1), our NLCE yields a uniform spin susceptibility that is in very good agreement with the experiment at low temperatures, while results from exact diagonalization of small periodic clusters exhibit very large finite-size effects and lead to a different result. In order to approach the ground state of the uniform KLHM, we simplify the model Hamiltonian and use a tuning parameter, λ , that takes one from the distorted to the uniform KLHM and study the evolution of an order parameter for the VBS as λ is varied. Together with results from finite clusters, we find evidence for a phase transition before the uniform limit is reached and discuss its implications for the search for the properties of the KLHM ground state.

2. The Model

We write the KLHM Hamiltonian as:

$$\hat{H} = \sum_{\langle ij \rangle_\alpha} J_\alpha \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j \quad (1)$$

where $\langle \dots \rangle$ denotes nearest neighbors, $\hat{\mathbf{S}}_i$ is the spin-1/2 vector on site i , and α is the bond type. We consider up to four different bond types as depicted in Figure 1. The unit of energy is set to the largest exchange interaction (J_1).

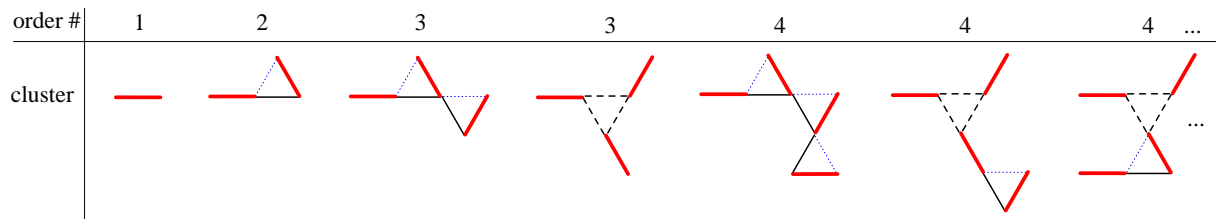


Figure 2. Topological clusters in the expansion with up to four pinwheel dimers (fourth order). Bond types are the same as in Fig. 1.

order number	1	2	3	4	5	6	7	8	9	10	11	12	13
number of clusters	1	1	2	3	7	14	25	50	102	216	442	929	1969

Table 1. Number of topological clusters in each order of the expansion.

3. Numerical Linked-Cluster Expansion

Linked-cluster expansions are methods in which an extensive property of the lattice model is expanded in terms of contributions from all the clusters, up to a certain size, that one can embed in the lattice [23]. This can be summarized in the following equation:

$$P = \sum_c L(c)W_{\mathcal{P}}(c), \quad (2)$$

where P is the property per site in the thermodynamic limit, $L(c)$ is the number of ways a cluster c can be embedded in the lattice, per site, and $W_{\mathcal{P}}(c)$ is the weight of cluster c for property \mathcal{P} . While $L(c)$ depends only on the geometry of the lattice, to calculate the weights, one has to compute the property of interest for different linked clusters, and use the inclusion-exclusion principle:

$$W_{\mathcal{P}}(c) = \mathcal{P}(c) - \sum_{s \subset c} W_{\mathcal{P}}(s), \quad (3)$$

where the sum is over all the subclusters of c . In high temperature expansions, $\mathcal{P}(c)$ are calculated using a perturbative expansion in powers of the inverse temperature. In that case, at low temperatures, the series can diverge even if the correlations in the model are short-ranged. A similar perturbative approach has also been used to calculate the ground state properties of a pinwheel distorted KLHM by using λ as the small parameter [6, 7]. Thus, the contributions of the clusters to each power of λ are properly added to the series.

In contrast, in NLCE, $\mathcal{P}(c)$ is calculated exactly using full diagonalization techniques and so, λ does not have to be small. The convergence of NLCE is controlled instead by the correlations in the system. Namely, NLCE converges if the correlation lengths, or the fluctuations around an ordered state, remain short-ranged. Moreover, the properties of each cluster are computed in all orders of λ , therefore, by increasing the size of the clusters in the series, one can systematically improve the results. There are many ways one can generate the topological clusters in the NLCE. Examples of that for the kagome lattice are, the site expansion, where the building block for generating clusters is a single site, bond expansion, triangular expansion, etc [21, 22].

For the pinwheel-distorted KLHM, one way to capture the low-temperature physics, associated with the VBS order, is to use the dimers of the pinwheel (strong bonds in Figure. 1, indicated by J_1) as the building blocks and generate the clusters in the expansion by attaching together more of those dimers through the other three types of bonds. In Figure. 2, we show all the topologically distinct clusters in our expansion that have four dimers or less. In Table 1, we enumerate the number of clusters in each order up to the 13th order. In the above expansion, we

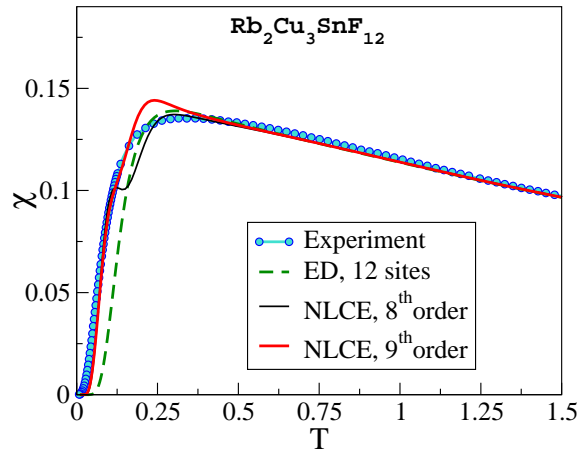


Figure 3. Comparison of the uniform spin susceptibility per site from our expansion to the experimental results (from Ref. [19]). The exchange interactions used are $J_2 = 0.95J_1$, $J_3 = 0.85J_1$ and $J_4 = 0.55J_1$, consistent with experimental finding for $\text{Rb}_2\text{Cu}_3\text{SnF}_{12}$. We have also included results from ED on a periodic 12-site cluster.

have restricted ourselves to those clusters that have the dimers connected through complete triangles. This is because at zero temperature, the contribution from any other cluster is zero [6, 7]. Even though this is not necessarily true at finite temperatures, this choice helps significantly reduce the number of topological clusters, which can be a limiting factor in large orders, while providing a consistent expansion scheme.

The limit on the number of orders in the expansion that can be treated by exact diagonalization is set either by memory or time requirements. In each order, the size of the clusters is twice the number of dimers, i.e., we need to solve the Heisenberg model on 18-site clusters in the 9th order. We use full diagonalization at finite temperatures and carry out the expansion to the 9th order. After block-diagonalizing the Hamiltonian matrix using the conservation of total spin in the z direction, the largest matrix belongs to the spin sector with equal number of spin-ups and spin-downs. More specifically, in the 9th order alone, we have to diagonalize matrices of size $\binom{18}{9} = 48,620$, or less, 102 times (the number of topological clusters in the 9th order) for each parameter set. For the zero-temperature calculations, we are interested only in the lowest eigenenergy and its eigenfunction. Therefore, we utilize the Lanczos algorithm to compute the ground state properties. Unlike for the finite-temperature case, here, we are not restricted by memory, but rather by the exponentially larger number of clusters in the higher orders and by the time it takes to diagonalize each of them. We carry out the calculations to the 13th order where there are 1969 clusters to diagonalize and the Hilbert space for their largest spin sector is $\binom{26}{13} = 10,400,600$.

4. Results

Employing NLCE with a pinwheel dimer expansion as explained above, we first explore the uniform susceptibility of $\text{Rb}_2\text{Cu}_3\text{SnF}_{12}$ that has been measured in experiments [19]. In Figure 3, we show the comparison between the experimental results and the results from NLCE with exchange interactions that are believed to be appropriate for this material ($J_2 = 0.95J_1$, $J_3 = 0.85J_1$ and $J_4 = 0.55J_1$) [19, 20]. We use $J_1 = 234\text{K}$, from experiments and match the two results at the highest temperature available experimentally with the g -factor, $g = 2.46$. We observe that NLCE converges at high temperatures and down to $T \sim 0.5$, as well as in the low-temperature window of $0 < T \lesssim 0.1$. Convergence in the latter region was expected since our NLCE is

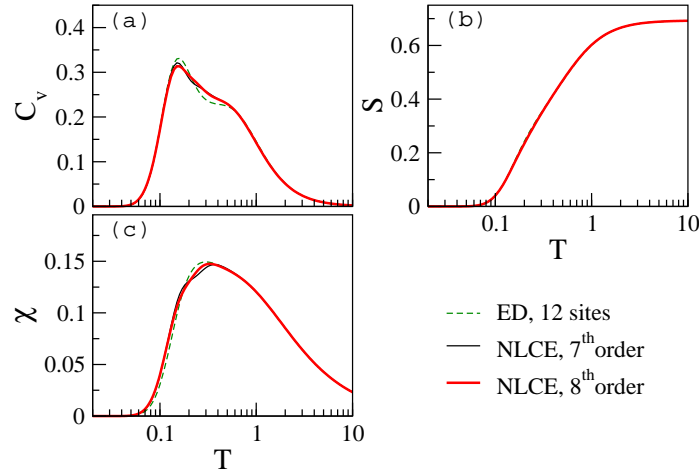


Figure 4. Specific heat, entropy, and uniform spin susceptibility per site for the pinwheel distorted Kagome lattice of Fig. 1 with $J_2 = J_3 = J_4 = \lambda J_1$ and $\lambda = 0.70$ using NLCE. Dashed lines are results from ED for the periodic 12-site cluster.

tailored to capture the VBS ground state of this material. However, to achieve convergence in the intermediate temperature region of $0.1 < T < 0.5$, where the thermal fluctuations around the VBS phase grow and eventually destroy it, one needs to consider higher orders (larger clusters) in NLCE, something that is computationally beyond our reach at this point. By comparing the ED results for a periodic 12-site cluster with the same exchange interactions used in NLCE, one can see that the discrepancy between ED and experiments at low temperatures is not due to the inaccurate exchange values, or their temperature dependence, but rather is caused by finite-size effects, which are absent in NLCEs.

To be able to define a tuning parameter that systematically transforms the pinwheel-distorted KLHM with VBS ground state to the uniform KLHM, we simplify the model and assume $J_2 = J_3 = J_4 = \lambda J_1$ [24]. When λ is sufficiently small, the ground state will still be the pinwheel VBS order with a 12-site unit cell. $\lambda = 1$ on the other hand, corresponds to the uniform KLHM. We are interested in the evolution of the thermodynamic properties of this model as λ is tuned from small values to close to unity. In Figure 4, we show the specific heat, entropy and the susceptibility for the case of $\lambda = 0.7$. The NLCE clearly converges at almost all temperature regions with this λ . We also point out the clear deviation of the ED results with the 12-site cluster from NLCE at $0.1 < T < 0.5$ [see e.g., Figure 4(a)]. The results for other values of λ [25] show that the convergence in NLCE at low temperatures is lost before λ reaches unity. Therefore, it is natural to ask, what is the value of λ beyond which NLCE no longer converges at low temperatures? and, what does that tell us about the robustness of the VBS order and the nature of the ground state of the KLHM?

To answer these questions, we focus on the ground state only and take advantage of the Lanczos algorithm to speed up the diagonalization process (since we are interested only in the lowest energy state), reduce the required memory, and therefore, be able to carry out the series to up to the 13th order in the dimer expansion. We calculate the bond energies, B_α , for the four different types of bonds that exist on the lattice. The difference between B_1 (corresponding to the pinwheel dimers) and any other bond can be used as the VBS order parameter [25]. Here, we choose this difference to be between B_1 and the average of the other three, $(B_2 + B_3 + B_4)/3 - B_1$. In the small- λ region, $|B_1| > |(B_2 + B_3 + B_4)/3|$ and so, the order parameter is finite. On the other hand, at $\lambda = 1$, by symmetry, all B_α are the same and the order parameter for the uniform KLHM has to be zero.

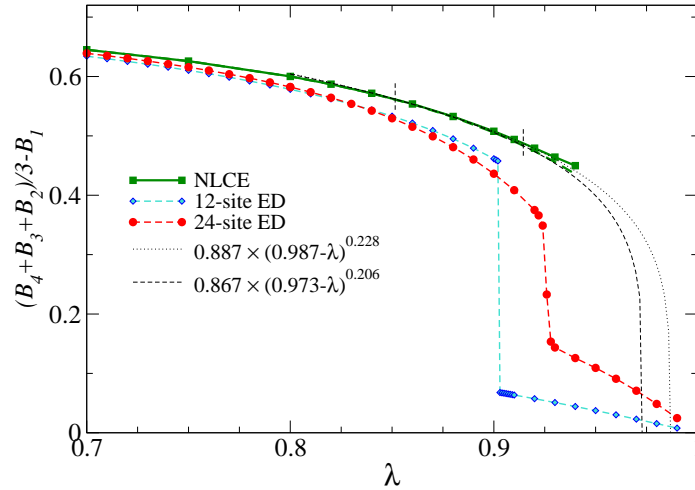


Figure 5. Order parameter at $T = 0$ vs the anisotropy parameter, λ , for $J_2 = J_3 = J_4 = \lambda J_1$. We use the Wynn sum with four cycles of improvement [21, 22] in NLCE. Results for the last two orders of the former are fit to $A(\lambda_c - \lambda)^B$ (thin dotted and dashed lines). Vertical solid lines show the region used for the fit.

The behavior of the order parameter by increasing λ at $T = 0$ is shown in Figure 5. As expected, the NLCE results converge well for small λ . However, beyond $\lambda = 0.9$, the series do not converge anymore. This signals that the VBS may not be the ground state phase in the thermodynamic limit when the pinwheel distortion is small. Moreover, the results from ED with the 12-site and 24-site periodic clusters show a clear first-order phase transition (level crossing) to another phase at or slightly above $\lambda = 0.9$. These findings provide strong evidence that in the thermodynamic limit, a phase transition likely occurs at a critical λ_c as the distortion is decreased and before the uniform KLHM limit is reached. We point out that there is also the possibility for a second-order, or weak first-order, phase transition at a λ_c . For this reason, we fit the high- λ region of the NLCE results to a power-law form to deduce λ_c in such a scenario and find that $\lambda_c < 1$ (see Figure 5).

Note that if in fact the transition is first order, λ_c will be smaller than that of the second-order transition found above. Moreover, if the transition happens at $\lambda = 1$, it is likely to a gapless spin liquid, since in that case, any dimerization perturbation of this type would immediately take the system from the spin liquid to the VBS phase. However, such scenario is not in agreement with the recent DMRG study [9] that argues the ground state of the KLHM is a *gapped* spin liquid.

5. Conclusions

In conclusion, we implemented a numerical linked-cluster expansion in terms of strong bonds on a pinwheel-distorted kagome lattice with a 12-site unit cell that describes the material $\text{Rb}_2\text{Cu}_3\text{SnF}_{12}$. Using the appropriate exchange parameters for this material, we calculate the uniform susceptibility that is in very good agreement with the experimental results. We define a tuning parameter, λ , that takes one from the distorted to the uniform kagome lattice in a simplified model and focus on the properties of the ground state as λ is varied. NLCE results, along with the results from exact diagonalization of small clusters with periodic boundary condition, point to a phase transition that likely occurs in the thermodynamic limit before the uniform limit is reached. This implies that the KLHM may not spontaneously dimerize in such pattern and that the ground state phase is stable to small dimerization perturbations of this type.

Acknowledgments

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