p6 chiral resonating valence bonds in the kagome antiferromagnet

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The kagome Heisenberg antiferromagnet is mapped onto an effective Hamiltonian on the star superlattice by contractor renormalization. A comparison of ground-state energies on large lattices to density matrix renormalization group (DMRG)11,12 shows that the thermodynamic limit into a chiral ground state. This chirality is understood as the effect of three-star interactions. Classical mean field theory on the effective Hamiltonian explains this symmetry breaking. A two-dimer chirality order parameter is defined on the microscopic kagome model. We propose an experimental signature of this broken symmetry in the phonon spectrum: a splitting of symmetry-protected degeneracy between two zone center optical modes. Finally, we add weak ferromagnetic next nearest neighbor interactions J2, and find that it eliminates the chirality at J2 ≈ −0.1.

CORE procedure. Previous CORE calculations for the kagome model18,19 started with up-triangle blocking, and did not reach sufficient convergence at range 3. Here we use much larger and more symmetric blocks of 12-site (“Star of David”) stars which form a triangular superlattice. In each star, we retain just the two degenerate singlet ground states |L⟩ and |R⟩, depicted in Fig. 1, which form a pseudospin-1/2

FIG. 1. (Color online) The CORE-blocking scheme on the kagome lattice. R and L denote the two pinwheel ground states of the 12-site stars, and the arrows (pseudospins) denote the symmetrized R basis which spans the reduced Hilbert space of $H_{\text{CORE}}$ [see Eq. (2)].
basis:

\[
|\uparrow_i\rangle = \frac{1}{\sqrt{2+1/16}}(|R_i\rangle + |L_i\rangle),
\]

\[
|\downarrow_i\rangle = \frac{1}{\sqrt{2-1/16}}(|R_i\rangle - |L_i\rangle).
\]

Note that the two states are \(C_0\) invariant, and have opposite parity under all \(D_6\) reflections.

The CORE effective Hamiltonian on a superlattice of size \(N_s\) stars is defined by the cluster expansion

\[
H_{\text{CORE}}^{n_0} = \sum_{i \leq j} h^{n_0}_{ij} + \sum_{i \leq j} h^{n_1}_{ij} + \cdots + \sum_{i \leq j} h^{n_N_{s}}_{ij},
\]

\[
h^{n_0}_{ij} \equiv H^{(2)} - \sum_{m<n} \beta(nm)\beta(nm). \tag{3}
\]

Here \(\beta(nm)\) is a connected subcluster of size \(m\) in a cluster \(\alpha(n)\) of size \(n\) stars, and \(h^{n_0}_{ij}\) is defined to be an interaction of range \(n\). The operators \(H^{n_0}_{ij}\) are constructed by ED of Eq. (1) on a kagome cluster \(\alpha\):

\[
H^{n_0}_{\alpha} = \sum_{\nu} e^{\nu}_{\sigma} |\Psi^{\nu}_{\sigma}\rangle |\Psi^{\nu}_{\sigma}\rangle. \tag{4}
\]

Here \((e^{\nu}_{\sigma}, |\Psi^{\nu}_{\sigma}\rangle)\) are the exact \(2^n\) lowest singlet energies and wave functions. The states \(|\Psi^{\nu}_{\sigma}\rangle\) are an orthogonal basis constructed by sequential projections of \(|\Psi^{\nu}_{\sigma}\rangle\) by lattice reflection symmetries is obtained sequentially by using the Gram-Schmidt procedure.

If interactions of all ranges \(n \leq N_s\) are included, then \(H_{\text{CORE}}^{n_0}\) has the identical low-energy singlet spectrum as Eq. (1) on the equivalent kagome lattice. However, the ED cost to compute \(h^{n_0}\) grows exponentially with \(n\).

The error in the ground-state energy \(\Delta E = \Delta E_{\text{CORE}}\) of \(36\) spins with open boundary conditions (OBCs) is estimated by subtracting the ground-state energy of \(H^{\text{CORE}}_{\text{3}}\) from that of \(H^{\text{CORE}}_{\text{3}}\) on the three-star triangular cluster. This required \(O(36)\) computations.

Lattice translations. Our choice of stars for the reduced Hilbert space nominally breaks lattice translational symmetry as seen in Fig. 1. The microscopic spin correlations are computed by a functional differentiation of the CORE ground-state energy with respect to source terms. In principle, one must compute the effective interactions to all ranges to restore full translational symmetry. Nevertheless, symmetry breaking artifacts decrease with the truncation range \(n\). We can therefore identify any spontaneous translational symmetry breaking which significantly exceeds the truncation error.

CORE range 2. We start with the lowest-order truncation at range 2. The general form of the two-star interactions allowed by lattice reflection symmetries is

\[
H_{\text{CORE}}^{n_1} = N c_0 + h \sum_{i} a_{i} + J^{0} \sum_{(ij)} a_{i} a_{j}, \tag{5}
\]

where \(i\) labels sites, and \((ij)\) nearest neighbor bonds on the triangular lattice. \(a_{\sigma}, \alpha = x,y,z,\) are Pauli matrices.

### Table I. Parameters of the CORE range-2 Hamiltonian, by exact diagonalization and second order perturbation theory (Ref. 21).

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>(c_0)</td>
<td>-6.26391</td>
</tr>
<tr>
<td>(h)</td>
<td>0.13818</td>
</tr>
<tr>
<td>(J^0)</td>
<td>0.00713</td>
</tr>
<tr>
<td>(J^1)</td>
<td>-0.00105</td>
</tr>
<tr>
<td>(J^2)</td>
<td>-0.00045</td>
</tr>
<tr>
<td>PT</td>
<td>-5.268</td>
</tr>
<tr>
<td>0</td>
<td>0.046</td>
</tr>
<tr>
<td>0</td>
<td>-0.00025</td>
</tr>
<tr>
<td>0</td>
<td>-0.00175</td>
</tr>
</tbody>
</table>

The parameters derived from the lowest four eigenstates of 24 spins are computed by the Lanczos algorithm, and are listed in Table I. It is instructive to compare the ED parameters to the second order perturbation theory (PT) in the interstar bonds, as was calculated by Syromyatnikov and Maleyev. Second order PT in the connecting bonds is not very accurate when the connecting bonds have exchanges equal to 1. For example, PT misses the important \(J^1\) interactions. The dominant interaction of \(H_{\text{CORE}}^{n_1}\) is the field \(h = 0.138\), which would yield in the thermodynamic system a ferromagnetic ground state polarized in the \(\downarrow\) direction. In terms of kagome spins, the ground state would be a product of \(a\) symmetric superposition of pinwheel states, with local \(\uparrow\) fluctuations generated by the \(xx\), \(yy\) terms.

Within CORE2, the connecting bonds energy is \(E_{\text{inter}} = -0.21283\) versus the intrastar bonds at \(E_{\text{intra}} = -0.2225\). Interestingly, the modulation is already diminished from 100% to 4.3% with range-2 interactions.

How accurate is \(H_{\text{CORE}}^{n_1}\)? Unfortunately, it is not as accurate as needed. The exact ground-state energy per site of \(\mathcal{H}\) for 36 sites is \(E_{\text{DD}} = -0.412\) while the CORE2 energy per site for three stars is \(E_{\text{CORE}}^{n_2} = -0.4277\). This error in energy per site on the triangular lattice is \(72|E_{\text{DD}} - E_{\text{CORE}}^{n_2}| = 1.0757\). This amounts to a large correction, just from range 3, of 780% of the CORE2 field term \(h = 0.138\) (see Table I). Hence, we must add the range-3 interactions.

CORE range 3. To obtain \(H_{\text{CORE}}^{n_2}\), we compute the interactions \(H_{\text{CORE}}^{n_2}\) on the three-star triangular cluster. This required \(O(36)\) computations.

Lattice translations. Our choice of stars for the reduced Hilbert space nominally breaks lattice translational symmetry as seen in Fig. 1. The microscopic spin correlations are computed by a functional differentiation of the CORE ground-state energy with respect to source terms. In principle, one must compute the effective interactions to all ranges to restore full translational symmetry. Nevertheless, symmetry breaking artifacts decrease with the truncation range \(n\). We can therefore identify any spontaneous translational symmetry breaking which significantly exceeds the truncation error.
estimate of the total magnitude of neglected interactions to the size of the dominant fields in $H^\text{CORE}_3$, which are $\hbar$ and $J_{zzz}$, we estimate that total neglected terms of all ranges $>3$ are at most of order 14% of the most important interaction couplings.

The effects of the truncated interactions on the ground state depend on the energy spacing and frustration of $H^\text{CORE}_3$. We shall soon see that the latter yields a nonfrustrated canted ferromagnet, and excitation energies of magnitude 0.1. Therefore the neglected interactions of order 0.008 are not expected to modify the ground-state correlations and symmetry breaking of $H^\text{CORE}_3$. Thus, we believe that CORE truncation at range 3 is sufficiently accurate to predict the correct thermodynamic phase.

$p6$ chirality. The ED spectrum of $H^\text{CORE}_3$ is evaluated on lattices of up to $N_s = 27$ stars (324 kagome sites) with PBC. The most striking feature on lattices larger than $N_s = 27$ stars is the emergence of ground-state degeneracy of two singlets with opposite parity under reflections. In the pseudospin representation, even (odd) parity states include only an even (odd) number of stars with antisymmetric $|\downarrow\rangle$ states. These degeneracies signal a spontaneous reflection symmetry breaking $p6\bar{m} \rightarrow p6$ in the thermodynamic limit.

A mean field (MF) energy of $H^\text{CORE}_3$ in spin-1/2 coherent states $|\Omega\rangle$ is

$$E_{\text{MF}} = N c_0 + h \sum_i \cos \theta_i + \sum_{(ij),a} J_{aa} \Omega^a_i \Omega^a_j,$$

where $\Omega_i = (\sin \theta_i \cos \phi_i, \sin \theta_i \sin \phi_i, \cos \theta_i)$. In Table II we see that for $J_z = 0$, the dominant couplings are the field $\hbar$ and the $J_z$ and $J_{zzz}$ exchanges. The last coupling is responsible for the chiral symmetry breaking, as it pulls the spins in the $\pm \hat{z}$ direction.

TABLE II. Interaction parameters of CORE range 3, with three values of $J_z$.  

<table>
<thead>
<tr>
<th>$J_z$</th>
<th>$J_z = 0$ (kagome)</th>
<th>$J_z = +0.1$</th>
<th>$J_z = -0.1$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$c_0$</td>
<td>$-5.24629$</td>
<td>$-5.17068$</td>
<td>$-5.48631$</td>
</tr>
<tr>
<td>$h$</td>
<td>$-0.069224$</td>
<td>$0.059323$</td>
<td>$-0.362797$</td>
</tr>
<tr>
<td>$J_x$</td>
<td>$-0.009028$</td>
<td>$-0.015421$</td>
<td>$0.001123$</td>
</tr>
<tr>
<td>$J_y$</td>
<td>$0.011879$</td>
<td>$0.001832$</td>
<td>$-0.017699$</td>
</tr>
<tr>
<td>$J_z$</td>
<td>$0.021056$</td>
<td>$0.003686$</td>
<td>$0.020141$</td>
</tr>
</tbody>
</table>


\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|}
\hline
Number of stars & $E_{\text{CORE}}$ & $E_{\text{DMRG}}$ & Error \\
\hline
2 x 2 & $-0.418452$ & $-0.417213$ & $-0.001239$ \\
2 x 3 & $-0.423953$ & $-0.422336$ & $-0.001617$ \\
3 x 4 & $-0.431150$ & $-0.428046$ & $-0.003104$ \\
3 x 5 & $-0.432688$ & $-0.429191$ & $-0.003497$ \\
\hline
\end{tabular}
\end{table}

\textbf{Minimizing $E_{\text{MF}}$}, we find a ferromagnetic state depicted in Fig. 2(a). The $z$ polarization $M_z^{\text{MF}} = \frac{1}{2} \cos \bar{\theta}$ is compared to ED in Table IV. For $J_z = 0$, we find that the chirality order is substantial with $\frac{1}{2} \sin \bar{\theta} = 0.424$ by MF and $\frac{1}{2} \langle \sigma^z \rangle = 0.397$ by ED.

In Fig. 2(b) we depict a typical dimer configuration which contributes to the $p6$ chiral RVB state. One can see the predominance of $R$ pinwheel chirality over $L$. The most local order parameter for this chirality is the two-dimer correlation depicted in Fig. 2(c),

$$C_i = \sum_d (S_d^a S_d^{\eta(d)} - S_d^a S_d^{\eta'(d)}),$$

where the dimer singlet projectors are

$$S_d = 1/4 - S_d^i \cdot S_d^j,$$

and $\eta'(\mathbf{d}) = \eta(\mathbf{d})$ is the bond emanating from $i$ at angle $\pi/3 (2\pi/3)$ relative to the dimer bond opposing $i$. The two terms in $C$ measure parts of pinwheels of opposite chirality.

\textbf{Translational symmetry}. At range 3, the energy of internal triangles $E_{\Delta_{\text{tri}}}$ is $-0.686$ and connecting triangles is $E_{\Delta_{\text{con}}} = -0.665$ (depicted by solid and dashed lines, respectively, in Fig. 1). This relative modulation of about 3.0% lies within the truncation error. Thus we can affirm that CORE$_3$ ground state...
is consistent with translational invariance, in agreement with DMRG.\textsuperscript{11,12}

\textbf{Singlet excitations.} In the 27-star lattice, the lowest singlet excitation above the two degenerate ground states is $\Delta E_{S=0} = 0.28$, which has a nonzero wave vector. This excitation gap does not vary much with lattice size. Within the pseudospin Hamiltonian, it can be understood as a local spin flip from the ferromagnetic ground state. We note that the singlet gap is slightly higher than two $S=1$ magnons at energies $E_{S=1} = 0.13$. This conclusion differs from that obtained by ED on the 36-site PBC, which found a large number of singlets below the spin gap.\textsuperscript{27} Since our effective Hamiltonian describes excitations on much larger lattices, we are inclined to associate these low singlets with the smaller PBC lattice geometry.

\textit{Experimentally}, fluctuating two-dimer correlations are tricky to observe directly. Fortunately, real compounds have a sizable magnetoelastic coupling between the ions and the dimer singlets. While, on average, dimer density and bond lengths are uniform in the RVB state, \textit{dimer density fluctuations}, $\delta \rho_d$ governed by the characteristic singlet energy scale, are linearly coupled to the ionic displacements. In Fig. 3, the effect of a temporary excess of dimers on a triangle is shown. In the chiral phase, imbalance between the left and right bonds emanating out of the triangle produces a chiral force on the ions, as depicted by the arrows. Integrating out the dimer density fluctuations results in a chiral perturbation to the phonon dynamical matrix.\textsuperscript{17} By symmetry, the degeneracy between two optical modes is removed at the zone center, as shown in Fig. 3. These phonons are polar, and therefore accessible to infrared spectroscopy but not to Raman scattering.\textsuperscript{29}

Finite $J_2$. We have added next nearest neighbor interactions with coupling $J_2$ to Eq. (1), and calculated the parameters of $H_{\text{CORE}}$, as shown in Table II. For $J_2 = 0.1$, we find the same doublet degeneracies and chirality as for the pure model $J_2 = 0$.\textsuperscript{14} In contrast, for a weak negative $J_2 = -0.1$ the spectrum changes dramatically: The doublets are removed, and the ground state is fully polarized in the $\uparrow$ direction. The precise nature of this phase needs to be explored. Interestingly, we notice that, in proximity to the parameters of Table II, one finds the Ising antiferromagnet in the field. Its ground state contains ferromagnetic hexagons with reversed spins in their center. It represents the hexagonal valence bond solid state, previously shown to have low variational energies,\textsuperscript{5} and proposed for $J_z \approx -0.1$.\textsuperscript{30}

\textbf{Summary.} Using CORE we arrived at an effective Hamiltonian, whose accuracy was determined to be sufficiently high so as to trust its predictions for the thermodynamic limit. Its ground state is consistent with a translationally invariant RVB phase, but with broken $p_6$ chiral symmetry. A two-dimer chiral order parameter is defined, which may be numerically explored on large lattices. Experimentally, it may be detected by splitting of optical phonon degeneracy.

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22. We order the clusters in terms of their radii. Thus three-star lines are considered longer range than the triangle. Three-star 120° lines are not included in the cluster expansion, as they get canceled in embedding range-4 rhombi (Ref. 20 and E. Altman, Ph.D. thesis, Technion-Israel Institute of Technology, 2002).
25. We use open boundary conditions on these clusters, and keep up to get a discarded weight smaller than 10^{-5}. Note that these variational DMRG energies give exact upper bounds on the ground-state energy (Ref. 11).