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*p*6 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 justifies truncation of effective interactions at range 3 (36 sites). Within our accuracy, magnetic and translational symmetries are not broken (i.e., a spin liquid ground state). However, we discover doublet spectral degeneracies which signal the onset of *p*6 chirality symmetry breaking. This is understood by a simple mean field analysis. Experimentally, the *p*6 chiral order parameter should split the optical phonon degeneracy near the zone center. The addition of weak next to nearest neighbor coupling is discussed.

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The antiferromagnetic Heisenberg model on the kagome lattice,

$$\mathcal{H} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j, \quad J > 0, \quad S = \frac{1}{2}, \tag{1}$$

is a much studied paradigm for frustrated quantum magnetism. In the classical approximation $S \to \infty$, this model exhibits macroscopic ground-state degeneracy which encumbers semiclassical approximations. There is evidence, both numerical and experimental [in $ZnCu_3(OH)_6Cl_2$ (Ref. 1)], that quantum fluctuations lead to a paramagnetic "spin liquid" ground state.[2]

Exact diagonalization (ED) studies,[3] and ED in the variational dimer singlet subspace,[4] have not approached the thermodynamic limit due to severe computer memory limitations. Many methods have proposed paramagnetic ground states, including lattice symmetry breaking "valence bond crystals,"[5–8] algebraic spin liquids,[9] and a time reversal symmetry breaking, chiral spin liquid.[10]

To date, the lowest energy on long cylinders has been found by the density matrix renormalization group (DMRG).[11,12] The DMRG ground state is a translationally invariant singlet, with apparently no broken translational or rotational symmetries. This state is consistent with a resonating valence bond (RVB) state[13] with a spin gap $\Delta_{S=1} = 0.13$ (henceforth we express energies in units of J) and Z_2 topological order.[12,14] It is still unclear, however, what are the low-energy singlet excitations of this state,[11,15] and whether or not any other symmetry of $\mathcal{H}$ may be broken in the infinite two-dimensional limit.

This Rapid Communication reports a surprising result: The thermodynamic ground state appears to break reflection symmetries, and to possess two-dimensional *p*6 chirality (not to be confused with "spin chirality" which also breaks time reversal symmetry).[10] Our conclusion is obtained by contractor renormalization (CORE)[16] with 12-site star blocking (see Fig. 1). The star scheme is found to reach sufficient accuracy with range-3 (36 sites) interactions. This is evidenced by comparing ground-state energies of the effective Hamiltonian H^{CORE_3} to high-precision DMRG on large lattices. The small modulation of bond energies is consistent, within our accuracy, with a translationally invariant singlet state as deduced by DMRG.[11,12]

H^{CORE_3} is diagonalized on up to 27 stars (effectively 324 kagome sites). The spectra exhibit doublet degeneracies between states with opposite parity under reflection.[17] These signal an unexpected spontaneous symmetry breaking in 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 J_2, and find that it eliminates the chirality at $J_2 \approx -0.1$.

CORE procedure. Previous CORE calculations for the kagome model[18,19] 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 $|L_i\rangle$ and $|R_i\rangle$, depicted in Fig. 1, which form a pseudospin-1/2

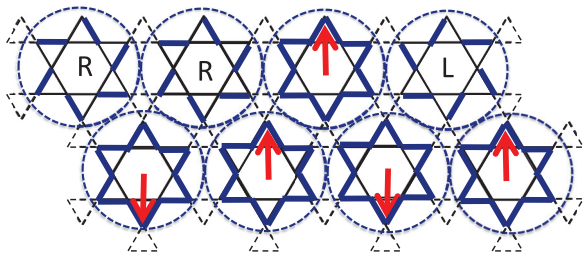

FIG. 1. (Color online) The CORE-blocking scheme on the kagome lattice. *R* and *L* denote the two pinwheel ground states of the 12-site stars, and the arrows (pseudospins) denote the symmetrized Ising basis which spans the reduced Hilbert space of H^{CORE} [see Eq. (2)].

basis:

$$|\uparrow_i\rangle = \frac{1}{\sqrt{2+1/16}}(|R_i\rangle + |L_i\rangle),$$
$$|\downarrow_i\rangle = \frac{1}{\sqrt{2-1/16}}(|R_i\rangle - |L_i\rangle). \quad (2)$$

Note that the two states are C_6 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

$$H^{\mathrm{CORE}} = \sum_{i=1}^{N_s} h_i^{(1)} + \sum_{i_1 i_2} h_{i_1,i_2}^{(2)} + \cdots + \sum_{i_1,\ldots,i_N} h_{i_1,\ldots,i_{N_s}}^{(N_s)},$$
$$h_\alpha^{(n)} \equiv H_\alpha^{(n)} - \sum_{m<n} \sum_{\beta(m)\in\alpha(n)} h_\beta^{(m)}. \quad (3)$$

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

$$H_\alpha^{(n)} = \sum_{\nu}^{2^n} \epsilon_\nu^\alpha \left|\tilde{\Psi}_\nu^\alpha\right\rangle\left\langle\tilde{\Psi}_\nu^\alpha\right|. \quad (4)$$

Here $(\epsilon_\nu^\alpha, \Psi_\nu^\alpha)$ are the exact 2^n lowest singlet energies and wave functions. The states $|\tilde{\Psi}_\nu\rangle$ are an orthogonal basis constructed by sequential projections of $|\Psi_\nu^\alpha\rangle, \nu = 1,2,\ldots,2^n$ onto the pseudospin states. After projection, the states are orthogonalized sequentially by using the Gram-Schmidt procedure.

If interactions of all ranges $n \leqslant N_s$ are included, then H^{CORE} has the identical low-energy singlet spectrum as Eq. (1) on the equivalent kagome lattice. However, the ED cost to compute $h^{(n)}$ grows exponentially with n. Thus, the success of CORE depends on the ability to truncate the cluster expansion at feasible n while maintaining sufficient accuracy in the truncated Hamiltonian.

The error in the ground-state energy $\delta E_0^{\mathrm{CORE}_n}$ can be computed by comparison to high-precision DMRG on large lattices with $m > n$ stars. This error should be much smaller than the important interactions in H^{CORE_n}.

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.[20] 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

$$H^{\mathrm{CORE}_2} = Nc_0 + h\sum_i \sigma_i^z + \sum_{\langle ij\rangle} J^\alpha \sigma_i^\alpha \sigma_j^\alpha, \quad (5)$$

where i labels sites, and $\langle ij\rangle$ nearest neighbor bonds on the triangular lattice. σ^α, $\alpha = x,y,z$, are Pauli matrices.

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

	c_0	h	J^x	J^y	J^z
ED	−6.26391	0.13818	0.00713	−0.00105	−0.00045
PT	−5.268	0.046	0	−0.00025	−0.00175

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.[21] 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^x interactions. The dominant interaction of H^{CORE_2} is the field $h = 0.138$, which would yield in the thermodynamic system a ferromagnetic ground state polarized in the $|\downarrow\rangle$ direction. In terms of kagome spins, the ground state would be a product of *antisymmetric* superposition of pinwheel states, with local $\uparrow$ fluctuations generated by the xx, yy terms.

Within CORE_2, the connecting bonds energy is $E_{\mathrm{inter}} = -0.212\,83$ versus the intrastar bonds at $E_{\mathrm{intra}} = -0.2225$. Interestingly, the modulation is already diminished from 100% to 4.3% with range-2 interactions.

How accurate is H^{CORE_2}? Unfortunately, it is not as accurate as is needed. The exact ground-state energy per site of $\mathcal{H}$ for 36 sites is $E_0^{\mathrm{ED}} = -0.412\,76$ while the CORE_2 energy per site for three stars is $E_0^{\mathrm{CORE}_2} = -0.4277$. The error in energy per site on the triangular lattice is $72|E_0^{\mathrm{ED}} - E_0^{\mathrm{CORE}_2}| = 1.0757$. This amounts to a large correction, just from range 3, of 780% of the CORE_2 field term $h = 0.138$ (see Table I). Hence, we must add the range-3 interactions.

CORE range 3. To obtain H^{CORE_3} we compute the interactions $h^{(3)}$ on the three-star triangular cluster.[22] This required ED of Eq. (1) of 36 spins with open boundary conditions (OBCs). For verification, we ran both a standard Lanczos routine on a supercomputer, and the memory-economical Lanczos singular value decomposition (SVD) routine[23] on a desktop computer. Adding contributions from ranges 1 to 3 we obtain the following effective Hamiltonian:

$$H^{\mathrm{CORE}_3} = Nc_0 + \sum_i h\sigma_i^z + \sum_{\langle ij\rangle,\alpha} J_\alpha \sigma_i^\alpha \sigma_j^\alpha + \sum_{\langle ijk\rangle_\Delta,\alpha} J_{z\alpha\alpha}\sigma_i^z \sigma_j^\alpha \sigma_k^\alpha, \quad (6)$$

where $\langle ijk\rangle_\Delta$ label nearest neighbor triangles on the triangular lattice. The interaction parameters are listed in Table II. We do not list terms that cancel in the superlattice summation with periodic boundary conditions (PBCs).

The magnitude of the truncated interactions is estimated by subtracting the ground-state energy of H^{CORE_3} from that of high-precision DMRG[24,25] on clusters of ranges up to 15 stars. In Table III we see that these interactions contribute less than <0.004 per site. If we extrapolated CORE_3 ground-state energy to the thermodynamic limit, we get -0.447 which underestimates the extrapolated DMRG result -0.439 (Ref. 12) by at most -0.008 per site. When we compare this

TABLE II. Interaction parameters of CORE range 3, with three values of J_2.

J_2	$J_2 = 0$ (kagome)	$J_2 = +0.1$	$J_2 = -0.1$
c_0	−5.24629	−5.17068	−5.48631
h	−0.069224	0.059323	−0.362797
J_x	−0.009028	−0.015421	0.001123
J_y	−0.011879	0.001832	−0.017699
J_z	0.021056	0.003686	0.020141
J_{zxx}	−0.027920	−0.019649	−0.009524
J_{zyy}	0.004550	−0.004749	0.002394
J_{zzz}	0.000660	−0.001410	0.010495

estimate of the total magnitude of neglected interactions to the size of the dominant fields in H^{CORE_3}, which are h and J_{xzz}, 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^{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^{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^{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 $N_s = 9$ 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 $p6m \rightarrow p6$ in the thermodynamic limit.

A mean field (MF) energy of H^{CORE_3} in spin-1/2 coherent states $|\mathbf{\Omega}_i\rangle$ is

$$E^{\mathrm{MF}} = Nc_0 + h\sum_i \cos\theta_i + \sum_{\langle ij\rangle,\alpha} J_\alpha \mathbf{\Omega}_i^\alpha \mathbf{\Omega}_j^\alpha, + \sum_{\langle ijk\rangle,\alpha} J_{z\alpha\alpha}\cos\theta_i \mathbf{\Omega}_j^\alpha \mathbf{\Omega}_k^\alpha, \quad (7)$$

where $\mathbf{\Omega}_i = (\sin\theta_i\cos\phi_i, \sin\theta_i\sin\phi_i, \cos\theta_i)$. In Table II we see that for $J_2 = 0$, the dominant couplings are the field h and the J_z and J_{zxx} exchanges. The last coupling is responsible for the chiral symmetry breaking, as it pulls the spins in the $\pm\hat{x}$ direction.

TABLE III. Ground state energies per site of H^{CORE_3} and comparison to DMRG (Ref. 26) on equivalent kagome clusters (with OBC).

Number of stars	$E_0^{\mathrm{CORE}_3}$	E_0^{DMRG}	Error
2×2	−0.418452	−0.417213	−0.001239
2×3	−0.423953	−0.422336	−0.001617
3×4	−0.431150	−0.428046	−0.003104
3×5	−0.432688	−0.429191	−0.003497

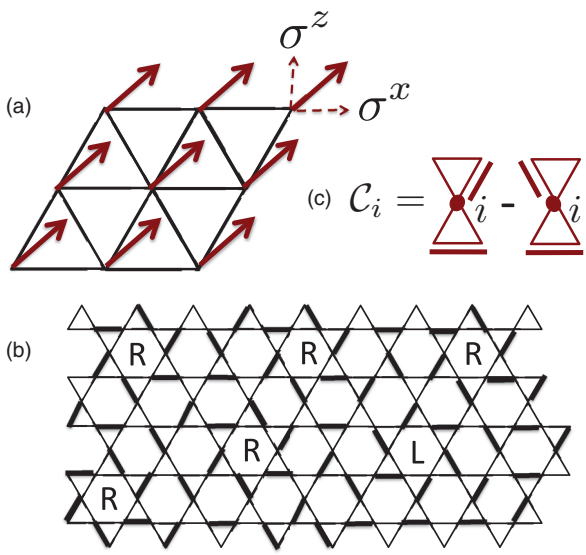

FIG. 2. (Color online) (a) The mean field ground state of H^{CORE_3} exhibiting $\langle\sigma^x\rangle > 0$ order, which corresponds to two-dimensional chirality. (b) A typical singlet configuration in the corresponding ground state of the kagome lattice. Notice that there is no translational order, but that there are more pinwheel configurations of $|R\rangle$ than $|L\rangle$. (c) The two-dimer chiral order parameter defined in Eq. (8).

Minimizing E^{MF}, we find a ferromagnetic state depicted in Fig. 2(a). The z polarization $M_z^{\mathrm{MF}} = \frac{1}{2}\cos\bar{\theta}$ is compared to ED in Table IV. For $J_2 = 0$, we find that the chirality order is substantial with $\frac{1}{2}\sin\bar{\theta} = 0.424$ by MF and $\frac{1}{2}\langle\sigma^x\rangle = 0.397$ 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),

$$\mathcal{C}_i = \sum_{\mathbf{d}} (\mathcal{S}_{\mathbf{d}}\mathcal{S}_{\boldsymbol{\eta}^r(\mathbf{d})} - \mathcal{S}_{\mathbf{d}}\mathcal{S}_{\boldsymbol{\eta}^l(\mathbf{d})}), \quad (8)$$

where the dimer singlet projectors are

$$\mathcal{S}_{\mathbf{d}} = 1/4 - \mathbf{S}_{\mathbf{d}_1}\cdot\mathbf{S}_{\mathbf{d}_2}, \quad (9)$$

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

Translational symmetry. At range 3, the energy of internal triangles $E_{\Delta_{\mathrm{inter}}} = -0.686$ and connecting triangles is $E_{\Delta_{\mathrm{intra}}} = -0.665$ (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

TABLE IV. Ground state z polarization of H^{CORE_3} on a 27-star lattice.

J_2	M_z^{MF}	M_z^{ED}
0.0	0.2647	0.2257
+0.1	0.1390	0.1476
−0.1	0.5	0.4999

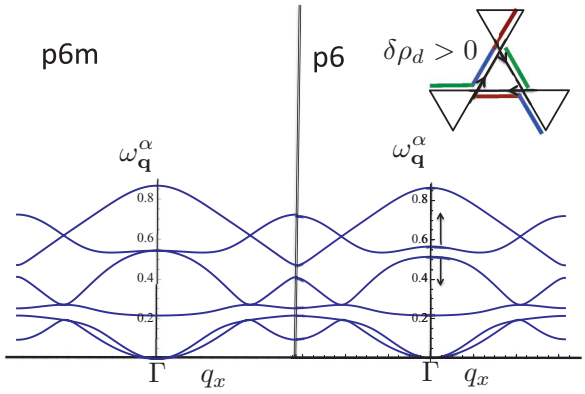

FIG. 3. (Color online) Kagome phonon spectra in the *p6m* phase and *p*6 chiral phase calculated within a nearest neighbor spring constant model given in Ref. 28. In the top right, the dimer chiral correlations induce a linear coupling between excess dimer density $\delta\rho_d$ and chiral ionic displacements, as depicted by the arrows. This adds a chiral term to the dynamical matrix which splits the degeneracy of the optical phonons at the zone center.

is consistent with *translational invariance*, in agreement with DMRG.[11,12]

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.[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.

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, *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.[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.[29]

Finite J_2. We have added next nearest neighbor interactions with coupling J_2 to Eq. (1), and calculated the parameters of H^{CORE_3}, as shown in Table II. For $J_2 = 0.1$, we find the same doublet degeneracies and chirality as for the pure model $J_2 = 0$.[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 ↑ 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,[5] and proposed for $J_2 \simeq -0.1$.[30]

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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[17]See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.87.161118 for details of the exact diagonalization of $CORE_3$ on 27 stars, and the effects of the chiral order parameter on the optical phonons of the kagome ions.

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