Geometric criticality between plaquette phases in integer-spin kagomé $XXZ$ antiferromagnets

Cenke Xu$^1$ and J. E. Moore$^{1,2}$

$^1$Department of Physics, University of California, Berkeley, CA 94720
$^2$Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720

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The phase diagram of the uniaxially anisotropic $s=1$ antiferromagnet on the kagomé lattice includes a critical line exactly described by the classical three-color model. This line is distinct from the standard geometric classical criticality that appears in the classical limit ($s \to \infty$) of the 2D $XY$ model; the $s=1$ geometric $T=0$ critical line separates two unconventional plaquette-ordered phases that survive to nonzero temperature. The experimentally important correlations at finite temperature and the nature of the transitions into these ordered phases are obtained using the mapping to the three-color model and a combination of perturbation theory and a variational ansatz for the ordered phases. The ordered phases show sixfold symmetry breaking and are similar to phases proposed for the honeycomb lattice dimer model and $s=1/2$ $XXZ$ model. The same mapping and phase transition can be realized also for integer spins $s \geq 2$ but then require strong on-site anisotropy in the Hamiltonian.

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I. INTRODUCTION

The geometric constraints that determine the infinite set of ground states in classical highly frustrated magnets are known in several celebrated instances to give rise to power-law or “critical” correlations at vanishing temperature. A frustrated magnet is one in which not all interaction energies can be simultaneously optimized; here we follow convention in describing by “highly frustrated” the class of frustrated magnets that have an infinite number of classical ground states, even after removing global symmetries. An example of a frustrated magnet that is not highly frustrated is the triangular lattice $XY$ antiferromagnet, which has a unique ground state once the three spins on a single triangle are fixed.

The classical $XY$ antiferromagnet on the (2D) kagomé lattice and the classical Heisenberg antiferromagnet on the (3D) pyrochlore lattice are special members of the highly frustrated class: they show power-law correlations determined by a critical two-component height model in the first case$^{1,2}$ and by 3D dipolar correlations in the second$^{3}$. A general highly frustrated magnet, such as the Ising antiferromagnet on the kagomé or triangular lattice, has only short-ranged correlations, i.e., an exponential decay of correlations beyond a finite “correlation length.” Geometrically critical frustrated magnets can be thought of as on the boundary between disordered phases with short-ranged correlations and long-ranged ordered phases, but rather than being tuned by an external parameter, the criticality is a consequence of the fixed geometry of the lattice. A major focus of current research in frustrated magnetism is how quantum effects in real magnets with finite spin $S$ modify these classical critical points, leading possibly to spin liquid$^{4,5,6,7,8}$, emergent gauge symmetries$^9$, and novel ordered phases.

The first goal of this paper is to show that the uniaxially anisotropic spin-1 antiferromagnet on the kagomé lattice has a range of parameters over which the ground states are exactly determined by the “three-color model”$^{10}$, which is the same geometrically critical model that appears in the classical $XY$ model on the same lattice$^{11}$. The $s=1$ kagomé antiferromagnet is thus a rare example where exact information can be obtained on a frustrated quantum magnet. Uniaxial ($XXZ$) anisotropy is generic in kagomé materials because their layered quasi-2D structure implies $XXZ$ anisotropy. A typical kagomé compound is the $s=3/2$ material SrCr$_5$Ga$_{4+x}$O$_{12+1}$; nickel kagomé compounds have $s=1$. A recent neutron scattering study$^{12}$ of the iron jarosite KFe$_3$(OH)$_6$(SO$_4$)$_2$, which has $s=5/2$, observed, in addition to expected classical Heisenberg kagomé physics, several effects induced by Dzyaloshinskii-Moriya terms resulting from additional ions that break the inversion symmetry of the kagomé lattice.

The second part looks at the plaquette-ordered phases separated by this critical line, which serves as a solvable starting point for perturbation theory. The critical line occurs for reasons quite different from the critical point of the classical $XY$ model on this lattice; in particular, the $s=1$ problem has a nonvanishing gap above the ground-state manifold in the thermodynamic limit, unlike the large-$S$ case. Section II reviews the connection between the classical $XY$ antiferromagnet and the three-color model and introduces the $XXZ$ model for spin-1 antiferromagnets on the kagomé lattice. From this we derive the existence of a critical line related to the three-color model. Section III discusses the ordered phases separated by this critical line and the nature of the phase transition and concludes by developing a gauge theory description in order to compare this system to other recently studied models of frustrated magnetism.
II. THE KAGOMÉ ANTI-FERROMAGNET AND
THE THREE-COLOR MODEL

The ground states of the classical XY model on the
kagomé lattice (Fig. 1) are equivalent\textsuperscript{12,13,15} up to an over-
all $O(2)$ rotation, to the states of the honeycomb lattice
“three-color model”\textsuperscript{10}. Once one spin is fixed, all other
spins form angles of 0 or $\pm 2\pi/3$ with this spin: labeling
these three directions with colors, the ground states have
one spin of each color around each small triangle (Fig. 1).
This is equivalent to coloring the bonds of a honeycomb
lattice with three colors in such a way that each vertex
joins bonds of all three colors. The ground-state degener-
acy and chiral susceptibility were obtained by Baxter\textsuperscript{10}.
Correlations within this set of (equally weighted) ground
states, which determine the thermodynamics in the limit
of low temperature, are determined by a two-component
height model with an $SU(3)$ symmetry\textsuperscript{15}; one quantity
with critical correlations is the chirality $\chi = \pm 1$ of colors
around a vertex\textsuperscript{10}:

\[
\langle \chi(r_1)\chi(r_2) \rangle \sim \frac{1}{|r_1 - r_2|^4}.
\] (1)

Experimental observation of spin chirality in a kagomé
compound with approximate Heisenberg symmetry\textsuperscript{15} is
discussed in\textsuperscript{22}.

Now consider $s = 1$ spins on the sites of the kagomé
lattice, with antiferromagnetic, uniaxially anisotropic in-
teractions $J_{xy} \neq J_z$ between nearest neighbors.
Once the interactions are anisotropic, symmetry requires the
inclusion of on-site anisotropy at the same order:

\[
H = \sum_{\langle ij \rangle} [J_{xy}(S_x^i \cdot S_x^j + S_y^i \cdot S_y^j) + J_z S_z^i \cdot S_z^j] + D \sum_i (S_z^i)^2.
\] (2)

Note that the on-site anisotropy term $D$ would be con-
stant in an $s = 1/2$ XXZ model. Terms omitted for an
isolated kagomé layer are long-ranged (beyond nearest-
neighbor), involve more than 2 spin operators, or break
time-reversal symmetry, so it is appropriate when ex-
change interactions are dominant and time-reversal is not
explicitly broken. Dzyaloshinskii-Moriya terms absent in
\textsuperscript{22} are allowed if the nonmagnetic lattice surrounding
the kagomé layers breaks inversion symmetry, as in the
compound studied in\textsuperscript{12}.

We start with the case $J_{xy} = 0$, in which the model is
effectively classical (i.e., $S_z$ on each site commutes with
the Hamiltonian). Rewriting the Hamiltonian as a sum
over the small triangles in the kagomé lattice gives (here
1, 2, 3 label the three spins in a triangle)

\[
H = \sum_{\triangle} \frac{J_z}{2} (S_z^1 + S_z^2 + S_z^3)^2 + \frac{D - J_z}{2} ((S_z^1)^2 + (S_z^2)^2 + (S_z^3)^2).
\] (3)

The first term is minimized if $\sum_{\triangle} S_z^i = 0$, while the sec-
ond term favors $S_z = 0$ for $D > J_z$ and $S_z = \pm 1$ for
$D < J_z$. This paper concentrates on the case of weak
positive $D$, but first we briefly list the other possibilities.
For $D > J_z$, the ground state is simply $S_z = 0$ every-
where; the effective $U(1)$ gauge theory for low-energy ex-
citations when $J_{xy} \neq 0$ is added to this case has been
studied by Wen\textsuperscript{15}. For $D < 0$, the ground state has
$S_z = \pm 1$ everywhere and the problem reduces to the
classical Ising kagomé antiferromagnet, which is strongly
disordered (i.e., the correlation functions in the equally
weighted set of ground states fall off exponentially with
distance).

The intermediate range, $0 < D < J_z$, is classically crit-
ical: a ground state has on every triangle one spin with
$S_z = 1$, one with $S_z = 0$, and one with $S_z = -1$, and
this condition is an exact statement of the three-color
problem for the bonds of the honeycomb lattice. Even
though the Hamiltonian $H$ does not have the additional
$\mathbb{Z}_3$ color symmetry of the three-state Potts antiferromag-
net or the classical XY antiferromagnet, the ground state
family of states $doses$ have this symmetry. Note that the
additional symmetry is $\mathbb{Z}_3$ rather than the permutation
group of three colors $\mathbb{S}_3$ because there is always a time-
reversal symmetry in the Hamiltonian: the full permuta-
tion group of the three colors is made of this twofold
symmetry that interchanges $S = +1$ and $S_z = -1$, plus
three chirality-preserving rotations of the colors (isomor-
phic to $\mathbb{Z}_3$). The icelike $T \rightarrow 0$ entropy per triangle of
the kagomé lattice is half the entropy per hexagon of the
coloring problem, $S_{\Delta} = 0.189557k_{\text{B}}\textsuperscript{10}$.

The key difference between this $s = 1$ realization of the
d不宜, the standard classical XY realization is the existence of an energy gap above the ground.

FIG. 1: A portion of the kagomé lattice, with the associated honeycomb lattice (dotted lines). For $0 < D < J_z$, $J_{xy} = 0$, and $T = 0$, the three spins around every small triangle include one with $S_z = +1$, one with $S_z = -1$, and one with $S_z = 0$. The bottom right shows breakup of a single bond defect into two vertex defects (large dots): the two defective vertices of the honeycomb lie on a loop containing only $S_z = 0, +1$. 

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state manifold in the $s = 1$ model. The existence of a critical line (a range of values of $D$ over which the model is critical) is simply a consequence of this gap: the set of ground states is exactly the same for all values of $D$, but the energy gap to defects varies with $D$ as now calculated. Clearly the energy gap to these defects collapses at the two endpoints: below we obtain the correlation length at low temperatures by the inverse correlation volume:

$$\xi(T) \sim \frac{D}{kT}.$$  

(These formulas describe the exponential part of the behavior: it is possible that there are power-law dependences in the prefactors.) The chirality correlator will show power-law behavior for $r = \phi_1 - \phi_2 \ll \xi$, and then decrease as $\exp(-r/\xi)$ once $r \gg \xi$. A small numerical transfer-matrix study (up to 9 bonds per unit cell) confirms qualitatively the prediction that $f_s$ for a small system of size $L \ll \xi(T)$ goes as $\exp(-D/kT)$ when $kT \ll D$ with a temperature-dependent prefactor that increases with system size: in the thermodynamic limit, this prefactor gives rise to the different scaling in $\xi$.

Recent interest in the classical three-color model has focused on its unusually slow dynamics at low temperature, which invalidate local Monte Carlo methods\cite{13,14}: the $s = 1$ anisotropic kagomé antiferromagnet may provide an experimental system in which the dynamical phenomena predicted for this classical model can be observed. The rest of this paper concentrates on the quantum model: nonzero $J_{xy}$ generates two different quantum ordered phases separated by the classical line ($J_{xy} = 0.0 < D < J_z$) with critical correlations. Unlike the power-law correlations on the critical line, the order parameters in these phases are predicted to survive to $T > 0$. The following section starts with a heuristic derivation of the ordered phases, then develop a lattice $U(1)$ gauge theory to justify some assumptions and connect to other models.

III. PLAQUETTE-ORDERED PHASES NEAR THE CRITICAL LINE

This section starts by finding states near the critical line using a standard perturbation-theoretic approach within the degenerate manifold of ground states found above. The first splitting of the ground-state degeneracy of the three-color model occurs at third order in perturbation theory in $J_{xy}/J_z$: the effective Hamiltonian is

$$H_{eff} = -t \sum_\circ (S_{1\circ}^1 S_{2\circ}^2 S_{3\circ}^3 + S_{1\circ}^1 S_{2\circ}^4 S_{3\circ}^5 + \text{h.c.})$$

where $t = (3J_{xy}^3)/(2J_z^2)$, and the numbers 1 to 6 refer to consecutive spins around a hexagon of the kagomé lattice; spins live on bonds of the hexagons in Fig. 1. Similar loop terms occur in many models when an effective $H$
is derived for states in the constrained space. A similar perturbation theory has been carried out for the XXZ antiferromagnet on the triangular lattice\textsuperscript{21}.

\( H_{\text{eff}} \) will induce resonances when a hexagon is “flippable”, i.e., will superpose the two configurations \( S_z^i = (1, 0, 1, 0, 1, 0) \) and \((0, 1, 0, 1, 0, 1)\) around a hexagon, or superpose \((-1, 0, -1, 0, -1, 0)\) and \((0, -1, 0, -1, 0, -1)\). Note that the third type of alternating hexagon, e.g. \((-1, 1, -1, 1, -1)\), is not flippable, so that at this lowest nontrivial order of perturbation theory the \( \mathbb{Z}_3 \) symmetry of the classical critical line is broken. The existence of two kinds of hexagons that can resonate will lead to an additional symmetry breaking in the ordered phases we find, compared to predictions for the \( s = 1/2 \) XXX model\textsuperscript{10,22} and the honeycomb quantum dimer model\textsuperscript{16,23}.

For nonzero \( t \), we start by assuming that the system will favor a state with as many flippable hexagons as possible. This leads to the so-called \( \sqrt{3} \times \sqrt{3} \) state, in which 2/3 of the hexagons are flippable, as a classical starting point. The hexagons in the honeycomb lattice form a triangular lattice with three sublattices: in one \( \sqrt{3} \times \sqrt{3} \) state, for example, sublattice \( A \) has spins \((1,-1,1,-1,-1)\), sublattice \( B \) has spins \((1,0,1,0,1,0)\), and \( C \) has \((-1,0,-1,0,1,0)\). Now all hexagons of sublattices \( B \) and \( C \) are flippable. To minimize the energy starting from this state, one expects resonance in the \( B \) and \( C \) sublattices, but both cannot simultaneously resonate without violating the three-color constraint. Consider a variational wavefunction in which only sublattice \( B \) resonates. The spins with \( S_z = -1 \) are fixed and do not lie on the hexagons of sublattice \( B \); call these sites \( \bar{B} \). The other spins resonate: the lowest-energy superposition is

\[
\Psi_{\text{var}} = \prod_{i \in \bar{B}} |S_z^i = -1\rangle \\
\quad \otimes \prod_{O \in B} \left( \frac{|O = \begin{pmatrix} 0 & 0 \\ 1 & 0 \\ 0 \end{pmatrix}\rangle + |O = \begin{pmatrix} 0 & 0 \\ 0 & 1 \\ 0 \end{pmatrix}\rangle}{\sqrt{2}} \right). \tag{8}
\]

Here the sign in the superposition is the same for all hexagons, and for lowest energy has the same sign as \( t \) and \( J_{xy} \): we label the state with \( t < 0 \) as \( P1 \) and that with \( t > 0 \) as \( P2 \). This state has \( \langle H_{\text{eff}} \rangle = -|t|/3 \) per hexagon of the whole lattice, since only 1/3 of hexagons resonate. It is an eigenstate of \( H_{\text{eff}} \) if the Hilbert space is restricted to states satisfying the three-color constraint.

These states correspond to sixfold symmetry breaking: hexagons on one of three sublattices, containing either \( S_z = +1, 0 \) or \( S_z = 0, -1 \), resonate, and the other two sublattices are symmetric. This combined sublattice and time-reversal symmetry breaking is expected to survive to finite temperature, with a transition temperature \( T_c \) of order \(|t|\) (Fig. 2). At finite temperature or by including additional terms in the model, it may be possible to separate this sixfold symmetry breaking into two transitions. The zero-temperature transition between \( P1 \) and \( P2 \) is unusual in that it is a first-order transition by most definitions (the state changes discontinuously at \( t = 0 \)), but with algebraic correlations at the transition point determined by the \( SU(3) \) theory of the three-color model: effectively the kagomé lattice is fine-tuned so that this scenario occurs. For integer spin \( s \geq 2 \), the three-color problem will again describe the \( J_{xy} = 0 \) model if sufficiently strong \( S_z^4 \) anisotropy is added to the Hamiltonian.

A theory of two compact \( U(1) \) gauge fields on the bonds of the honeycomb lattice gives insight into how the ordered phases and critical line of this model connect to recent work on 2D and 3D frustrated magnetism. It is possible to use a single \( U(1) \) gauge field, following the compact QED description of the quantum dimer model\textsuperscript{24}: an integer-valued variable \( E^i = S_z^i \) is assigned to each bond \( i \) of the honeycomb lattice to represent spin, with a gauge constraint that the sum of integers around a vertex is 0, and an energetic constraint that restricts \( E^i = -1, 0, 1 \). However, the theory now given in terms of two gauge fields better captures the symmetries of the three-color line at \( t = 0 \): the two gauge fields are related to the two components of the height-model description.

First let two scalar fields \( E^i_1, a = 1, 2 \), be assigned to each bond \( i \): the values for the three spin possibilities are

\[
(E^i_1, E^i_2) = \begin{cases} 
(1, 0) & \text{if } S_z^i = 0 \\
(-1/2, \sqrt{3}/2) & \text{if } S_z^i = 1 \\
(-1/2, -\sqrt{3}/2) & \text{if } S_z^i = -1.
\end{cases} \tag{9}
\]

These three possible states can be selected by allowing \( (E_1, E_2) \) to range over the sites of a triangular lattice centered on the origin with an additional energy \( \frac{1}{\sqrt{3}} \sum_i [(E_1^i)^2 + (E_2^i)^2] \), \( k \to 0 \). The Hilbert space is constrained to have the sum of \( E \) fields vanish for the bonds around a vertex, creating two \( U(1) \) gauge symmetries.

Next, note that a 2D unit vector \( \hat{n}_i \) can be assigned parallel or antiparallel to each bond so that vertices of one sublattice of the honeycomb have 3 incoming bonds, while those of the other sublattice have 3 outgoing bonds. Now define two-component vector fields on bonds: \( E_a = E_a \hat{n}_i, a = 1, 2 \). Then the three-color constraint is Gauss’s law with no background charges: \( \nabla \cdot E_a = 0 \). Two height fields \( h_a \) on the faces are defined so that scalar \( E_a \) on a bond is equal to \( h_a - h_a^* \), where \( (r, l) \) indicate right and left w.r.t. the orientation on bonds: \( E_a = (\hat{z} \times \hat{n}) \cdot \nabla h_a \), which guarantees Gauss’s law around each vertex. This definition of height variables corresponds to the standard definition in the classical 3-color model\textsuperscript{25}.

The last step is to represent the ring-exchange terms, which break the \( \mathbb{Z}_3 \) symmetry. Define conjugate operators on each bond \( \langle A^\dagger_a, A^\dagger_j \rangle \) with commutation relations

\[
[A^\dagger_a, E^b_k] = i\delta_{jk}\delta_{ab}. \tag{10}
\]

Then \( T^a_j \equiv \exp(iA^\dagger_a) \) acts as a raising operator: it increases the quantum number \( E^a_j \) by 1. This enables a compact representation of the ring-exchange terms proportional to \( t \): on bond \( j \), \( \exp(iA^j_a\delta^{(1)}_a) \) will raise
\[ S_{z}^{j} = 0 \text{ to } S_{z}^{j} = 1 \text{ if } l^{(1)} = (-3/2, \sqrt{3}/2). \] Similarly, if \( l^{(2)} = (-3/2, -\sqrt{3}/2) \) then \( \exp(iA_{a}^{l^{(2)}}) \) takes \( S_{z}^{j} = 0 \) to \( S_{z}^{j} = -1 \). Defining vector fields \( \mathbf{A}_{j}^{l} = A_{j}^{l} \mathbf{n}_{j} \), the ring-exchange terms around hexagons become

\[
H' = -2t \left[ \cos(\nabla \times \mathbf{A}_{a}^{l^{(1)}}) + \cos(\nabla \times \mathbf{A}_{a}^{l^{(2)}}) \right].
\] (11)

Here, as usual in gauge theories of lattice spin models, the meaning of \( \cos(\nabla \times \mathbf{A}) \) is that one takes the lattice circulation around a plaquette: for \( \mathbf{v}_{j} \) an clockwise assignment of unit vectors along the bonds around a hexagon,

\[
\cos(\nabla \times \mathbf{A}) = \cos(A_{1} - A_{2} + A_{3} - A_{4} + A_{5} - A_{6}) = \cos \left( \sum_{j=1,...,6} \mathbf{v}_{j} \cdot \mathbf{A}_{j} \right).
\] (12)

Note that the two ring-exchange terms only act in one of two cases: either the spins around the hexagon alternate between \( S_{z} = 0 \) and \( S_{z} = 1 \), or between \( S_{z} = 0 \) and \( S_{z} = -1 \). The sixfold symmetry breaking in the plaquette-ordered phases results from the absence of the possible ring-exchange term with \( l^{(3)} = (0, \sqrt{3}) \). Adding this term restores a \( Z_{3} \) color symmetry, with likely nine-fold symmetry breaking in the ordered phases. The similarity of this gauge model to compact QED supports the conclusion that there is a gapped ordered state for \( t \neq 0 \).

A quantum height model can be obtained from this gauge theory: using the definition of height variables given above, the ring exchange term becomes

\[
\sum_{l=1}^{2} -2t \cos(\pi A_{a}^{l^{(2)}}), \text{ where } \pi_{a} \text{ is the operator conjugate to height.}
\]

The quantum height Hamiltonian can be used to study a simple variational wave function that gives the same ordered state as \( \mathbf{B} \). A generalization that can be studied using this technique is to an \( s = 1/2 \) XXZ kagomé bilayer in a magnetic field, with coupling between the layers that induces a three-color point: this system has columnar order rather than plaquette order.

### IV. Conclusions

We have used the proximity to a nontrivial classically solvable line to study part of the phase diagram of the \( s = 1 \) antiferromagnet on the kagomé lattice, including phases where \( J_{xy} \neq 0 \) and quantum Monte Carlo studies are impractical. The solvable line at zero temperature has an emergent \( Z_{3} \) symmetry of the three colors that does not exist in the full spectrum. A promising way to extend this approach closer to the isotropic point \( (J_{xy} = J, D = 0) \) is via numerical series expansion in \( J_{xy} / J \) from the critical line, to map out the extent of the plaquette-ordered phase. Finding other cases where frustrated magnets have solvable critical points for finite \( s \) similar to those in the kagomé and pyrochlore antiferromagnets at \( s = \infty \) is an important direction for future research.

The unusual phase diagram of the spin-1 antiferromagnet on the kagomé lattice should be verifiable by experimental neutron scattering studies of such compounds: in particular, confirmation of the plaquette ordered state should be feasible if it survives for a significant range of values of \( J_{xy} / J \). The special low-temperature physics of the three-color model at \( J_{xy} = 0 \) also can be discerned by sensitive thermodynamic measurements in principle, but away from the special three-color point, the zero-field thermodynamic signature of the phase transition into the plaquette-ordered phase will be similar to standard second-order phase transitions at finite temperature.

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