Kagome Antiferromagnet: A Chiral Topological Spin Liquid?

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Inspired by the recent discovery of a new instability towards a chiral phase of the classical Heisenberg model on the kagome lattice, we propose a specific chiral spin liquid that reconciles different, well-established results concerning both the classical and quantum models. This proposal is analyzed in an extended mean-field Schwinger boson framework encompassing time reversal symmetry breaking phases, which allows both a classical and a quantum phase description. At low temperatures, we find that quantum fluctuations favor this chiral phase, which is stable against small perturbations of second- and third-neighbor interactions. For spin-1/2, this phase may be, beyond the mean field, a chiral gapped spin liquid. Such a phase is consistent with the density matrix renormalization group results of Yan *et al.* [Science **332**, 1173 (2011)]. Mysterious features of the low-lying excitations of exact diagonalization spectra also find an explanation in this framework. Moreover, thermal fluctuations compete with quantum ones and induce a transition from this flux phase to a planar zero flux phase at a nonzero value of the renormalized temperature ($T/\mathcal{S}^2$), reconciling these results with those obtained for the classical system.

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With the extensive degeneracy of its classical ground state, the antiferromagnetic Heisenberg model on the kagome lattice was recognized early on as the paradigm of a quantum spin liquid phase [1]. The recent experimental discovery of Herbersmithite, a kagome compound fluctuating down to temperatures thousands of times lower than the coupling constant, strengthens this speculation [2]. Despite numerous efforts, the nature of the ground state (GS) of the spin-1/2 Heisenberg antiferromagnetic kagome model (AFKM) remains controversial. Exact quantum approaches point to the absence of long-range order [3–5]. Although exact diagonalizations (ED) on small samples (up to $N = 36$ sites) leave open the question of the criticality [6,7], density matrix renormalization group (DMRG) calculations [8] support the idea of a true gapped spin liquid.

Recently, a new instability of the degenerate *classical* model towards a chiral phase has been discovered [9]. In this Letter, we show at a mean-field level that the hypothesis of a chiral spin liquid holds and is consistent with numerous robust results accumulated during the last 20 years, both for spin-1/2 and in the classical limit [10–13].

The properties of chiral spin states, with simultaneously and spontaneously broken space reflection (P) and time reversal (T) symmetries, were largely debated at the end of the 1980s in the wake of the quantum Hall effect. A revival of these topics has occurred, thanks to graphene and flat band insulators. Wen *et al.* [14] defined the chiral phases through the fluxes of the underlying gauge fields, and Kalmeyer and Laughlin [15] proposed to describe spin liquids by Laughlin wave functions. Yang *et al.* [16] suggested that the Heisenberg model on the kagome lattice might be in a chiral spin liquid state. We reexamine this suggestion inspired by the knowledge of the classical nonplanar spin order, described in Fig. 1, and propose a specific *chiral spin liquid* as the GS of the spin-1/2 AFKM.

The kagome lattice can be viewed as a lattice of corner sharing triangles. The classical ground state on a single triangle is planar, with three spins at 120 degrees. Fixing the spin plane on a triangle does not fix the planes on adjacent ones, hence the extensive ground-state degeneracy. This degeneracy can be lifted by couplings beyond nearest neighbors. We consider the following J_1-J_2-J_{3h} Hamiltonian with $J_1 = 1$:

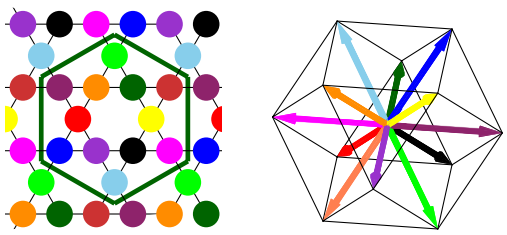

FIG. 1 (color online). Description of the *cuboc1* order. Left: on the kagome lattice, each color corresponds to a different magnetic sublattice. The thick line indicates the 12-site unit cell. Right: arrows are the spin orientations, with the same color coding as in the left figure. The black lines connecting the ends of the vectors form a cuboctahedron. On each triangle, the spins are coplanar at 120 degrees; for opposite sites on each hexagon, spins are antiparallel. The triple products (determinant) of three spins of the hexagons, either first or second neighbors, are nonzero and measure the chirality of the phase. They change sign in a mirror symmetry or in a spin flip.

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

where the coupling constants and *exact* classical phase diagram are given in Fig. 2. AFKM refers to this model when $J_2 = J_{3h} = 0$. For $J_{3h} = 0$, numerous studies of quantum states have been motivated by the two planar classical states denoted $\mathbf{q} = 0$ ($J_2 > 0$) and $\sqrt{3} \times \sqrt{3}$ ($J_2 < 0$) [11,12,17–19]. An infinitesimal antiferromagnetic third-neighbor interaction across hexagons ($J_{3h} > 0$) lifts the degeneracy of the classical AFKM to a 12-sublattice magnetic state where the spins point toward the corners of a cuboctahedron (Fig. 1), hence the name *cuboc1*. This order was first introduced by Janson *et al.* [20], who claim that this $J_{3h} > 0$ interaction should be of experimental relevance. This order is chiral: it breaks mirror symmetry.

Monitoring the evolution under the effect of quantum fluctuations from the classical limit to the disordered spin-1/2 system remains a challenge. The Schwinger boson mean-field theory (SBMFT) is an approximate but versatile method to study, in an unified framework, both long-range ordered (LRO) and gapped spin liquid phases, from the classical to the quantum limit. In a first enlightening work, Sachdev [17] showed that, amongst the planar states, the $\sqrt{3} \times \sqrt{3}$ is more stable than the $\mathbf{q} = 0$ at the AFKM point. In order to study the present model [Eq. (1)] around the AFKM point, we extend Sachdev's work in several directions, as will be seen below.

The Schwinger boson operator $\hat{b}^\dagger_{i\sigma}$ ($\sigma = \uparrow$ or $\downarrow$) creates a spin-1/2 on lattice site i. A physical spin s at site i is represented by $2s$ bosons. After a mean-field decoupling, the Hamiltonian reads

$$H_{\mathrm{MF}} = \sum_{\langle i,j\rangle_\alpha} J_\alpha (\mathcal{B}_{ij} \hat{B}^\dagger_{ij} - \mathcal{A}_{ij} \hat{A}^\dagger_{ij}) + \mathrm{H.c.} - \sum_i \lambda_i \hat{n}_i + \epsilon_0, \quad (2)$$

where the bond operators are defined by $2\hat{A}_{ij} = \hat{b}_{i\uparrow}\hat{b}_{j\downarrow} - \hat{b}_{i\downarrow}\hat{b}_{j\uparrow}$ and $2\hat{B}_{ij} = \hat{b}^\dagger_{i\uparrow}\hat{b}_{j\uparrow} + \hat{b}^\dagger_{i\downarrow}\hat{b}_{j\downarrow}$. $\mathcal{A}_{ij}$ and $\mathcal{B}_{ij}$ are the associated complex mean-field parameters to be determined by the self-consistency equations. The $\{\mathcal{A}_{ij}, \mathcal{B}_{ij}\}$ set is called an *Ansatz*. λ_i are Lagrange multipliers to constrain the mean boson number: $\langle \hat{n}_i \rangle = 2\mathcal{S}$ and $\epsilon_0 = \sum_{\langle i,j\rangle_\alpha} J_\alpha (|\mathcal{A}_{ij}|^2 - |\mathcal{B}_{ij}|^2) + 2\mathcal{S}\sum_i \lambda_i$, where $\mathcal{S}$ is a continuous real positive mean-field parameter.

Most SBMFT studies use only one of the two types of parameter ($\mathcal{A}$ or $\mathcal{B}$). Recently, taking both fields, Mezio *et al.* [21] found a much better description of the excitation spectrum of frustrated systems. More specifically, the $\mathcal{A}$ fields describe the singlet amplitudes, whereas the $\mathcal{B}$ fields allow the description of boson hopping amplitudes, which are a fundamental ingredient, to describe the mixing of spin singlets and triplets on each bond, a mechanism that is central in quantum frustrated magnets [22]. In addition, because both ferromagnetic and antiferromagnetic interactions are treated on an equal footing, the phase diagram can be explored continuously around the origin, regardless of the sign of the coupling parameters.

Solving the full problem with two complex parameters per link and one real Lagrange multiplier per site is too numerically demanding for large lattices. Looking for spin liquids or regular LRO [9], we assume $\lambda_i = \lambda$, and $\mathcal{A}$ and $\mathcal{B}$ are invariant under lattice symmetries up to a local gauge transformation. Using projective symmetry groups (PSGs) [23], Wang and Vishwanath [18] obtained four *Ansätze*, where physical observables are invariant under lattice symmetries. They are defined by the fluxes of the $\hat{A}$ operators on specific loops $(\phi_\bigcirc, \phi_\square) = (0,0)$, $(\pi, 0)$, $(0, \pi)$, and (π, π). $\sqrt{3} \times \sqrt{3}$ and $\mathbf{q} = 0$ are associated with the first two choices, respectively.

The chiral *cuboc1 Ansatz*, however, cannot be obtained within this first PSG approach because in chiral states the lattice symmetries are respected only up to a time reversal symmetry. In all previous studies, the mean-field parameters were chosen as real, the fluxes equal to 0 or π, thus excluding chiral *Ansätze*. The extension of the PSG to include both the symmetric and the chiral spin liquids will be described in a longer paper [24]. In short, the new *Ansätze* are defined by complex fields with specific constraints on the moduli and arguments. Thanks to the PSG analysis, the number of parameters at the AFKM point is limited to 2 moduli of bond fields for $\mathbf{q} = 0$ or $\sqrt{3} \times \sqrt{3}$, plus a phase $\theta_{A_{1b}}$ for *cuboc1*. Other bond fields of the 6-spin unit cell are fixed by algebraic constraints (see Fig. 3 and Table I). Nonzero fluxes [25] (modulo π) induced by $\theta_{A_{1b}}$ are an indirect mean-field measure of the

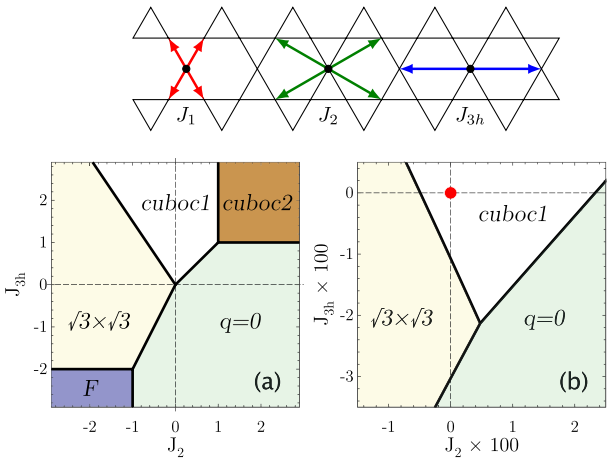

FIG. 2 (color online). Phase diagram of the model with up to third-neighbor interactions at $T = 0$ and $J_1 = 1$. (a) Exact classical phase diagram (the method used to obtain this phase diagram and all orders is described in [9]). The point AFKM: $(J_2, J_{3h}) = (0, 0)$ is a tricritical point. (b) SBMFT phase diagram for $\mathcal{S} = 0.5$. The tricritical point stands at $(J_2, J_{3h}) = (0.0049, -0.021)$, and the AFKM point (red circle) is now inside the *cuboc1* phase.

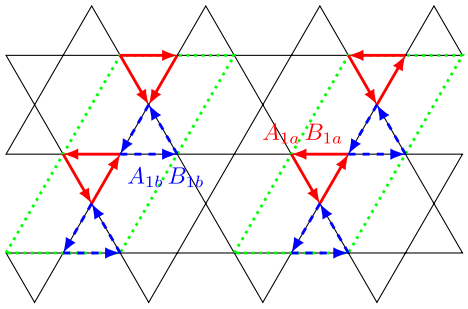

FIG. 3 (color online). Unit cell (in dotted green lines) of the *cuboc1 Ansatz* (left) and $\mathbf{q} = 0$ and the $\sqrt{3} \times \sqrt{3}$ *Ansätze* (right) at the AFKM point. A_{1a}, A_{1b}, B_{1a}, and B_{1b} are complex link parameters with constraints on their moduli: $|A_{1a}| = |A_{1b}|$ and $|B_{1a}| = |B_{1b}|$ and, on their arguments, $\theta_{A_{1a}} = 0$ and $\theta_{B_{1a}} = \theta_{B_{1b}} = \pi$. The constraint on $\theta_{A_{1b}}$ depends on the *Ansatz*: it is 0 for $\mathbf{q} = 0$, π for $\sqrt{3} \times \sqrt{3}$, and is not fixed for *cuboc1*.

chirality of *cuboc1* ($\theta_{A_{1b}}$ is 0 or π for $\mathbf{q} = 0$ or $\sqrt{3} \times \sqrt{3}$, respectively).

The numerical solution of the mean-field equation is found from a descent method minimizing the sum Σ of the squares of the (free) energy derivatives with respect to the field parameters, each single energy evaluation being maximized with respect to λ (we stop the descent when $\Sigma < 10^{-8}$). We keep the solutions with hessians of the correct sign, positive for the $\mathcal{A}$ fields and negative for the $\mathcal{B}$ if $J > 0$, and the opposite if $J < 0$. These requirements imply that our solutions are stable against Gaussian fluctuations.

The resulting phase diagram at $T = 0$ and $\mathcal{S} = 0.5$ is given in Fig. 2(b): at the AFKM point, the *cuboc1 Ansatz* is more stable than any other regular *Ansatz*, with an energy per site of -0.4717 [26]. The numerical values of the parameters at this point are given in Table I. The parameter range of stability of the *cuboc1* phase increases when the spin decreases. For $J_{3h} = 0$ and $\mathcal{S} = 0.5$, it is $J_2 \in [-0.005; 0.025]$ and, for $\mathcal{S} = 0.366$, it is enlarged to $J_2 \in [-0.008; 0.045]$. This increase is another proof of the role of quantum fluctuations in the stabilization of the *cuboc1* phase.

TABLE I. Values of the self-consistent SBMFT parameters for the three competing *Ansätze* near the AFKM point.

	$\mathcal{S}$	$\lvert A_1\rvert$	$\lvert B_1\rvert$	$\theta_{A_{1b}}$
$\mathbf{q} = 0$	1/2	0.516 24	0.180 36	0
	∞	$\sqrt{3}\mathcal{S}/2$	$\mathcal{S}/2$	0
$\sqrt{3} \times \sqrt{3}$	1/2	0.517 06	0.177 90	π
	∞	$\sqrt{3}\mathcal{S}/2$	$\mathcal{S}/2$	π
cuboc1	1/2	0.516 60	0.176 16	1.9525
	∞	$\sqrt{3}\mathcal{S}/2$	$\mathcal{S}/2$	1.9106[a]

[a]The exact value is $\pi - \arctan\sqrt{8}$.

The dimensionless free energy difference ($\Delta F/\mathcal{S}^2$) between the $\sqrt{3} \times \sqrt{3}$ phase and the *cuboc1* phase is given as a function of $\mathcal{S}$ in Fig. 4(a). At $T = 0$, it is of the order of 10^{-3} in favor of *cuboc1*. In the classical limit, $\mathcal{S} \to \infty$, the two phases are degenerate, as they should be. The comparison with the $\mathbf{q} = 0$ state is not shown, as it always has a much higher energy at the AFKM point.

Decreasing $\mathcal{S}$ leads to a second-order phase transition from a gapless LRO *cuboc1* phase to a fully gapped chiral spin liquid at a critical value $\mathcal{S}_c = 0.4$. One should not hastily conclude that the true spin-1/2 system has Néel long-range order. In this mean-field approach, the on-site number of bosons fluctuates: it is only fixed on average, $\mathcal{S}$ is a parameter, and $\langle \hat{\mathbf{S}}^2 \rangle = 3\mathcal{S}(\mathcal{S}+1)/2$ [27]. For spin-1/2, the good quantum number is $\langle \hat{\mathbf{S}}^2 \rangle = 3/4$. To recover this good quantum number, we should use the parameter $\mathcal{S} = (\sqrt{3} - 1)/2 \sim 0.366$. With $\mathcal{S} = 0.5$, the phase is gapless and finite-size scaling shows a very small stiffness, while, with $\mathcal{S} \sim 0.366$, the system is a gapped spin liquid compatible with the results of Yan *et al.*.

Now, we turn to the effect of thermal fluctuations at the AFKM point. Figure 4(b) shows how they destabilize the *cuboc1* phase in favor of the $\sqrt{3} \times \sqrt{3}$ one. The renormalized transition temperature $T_c/\mathcal{S}^2$ from the *cuboc1* to the $\sqrt{3} \times \sqrt{3}$ phase decreases to zero with increasing $\mathcal{S}$ [see the inset of Fig. 4(b)]. This is consistent with classical numerical simulations showing a selection of a planar state by thermal fluctuations [11–13,19].

This chiral order hypothesis also explains why the ED spectra for sizes up to $N = 36$ have a large number of singlets below the triplet gap. Let us consider a cell of 12 spin-1/2 describing some short-range order (SRO). Assuming three spin directions (as for $\sqrt{3} \times \sqrt{3}$ and $\mathbf{q} = 0$ SRO), one finds a single $S = 0$ ground state derived from the coupling of three spin-2 dressed by quantum fluctuations. With a 12-sublattice *cuboc1* SRO, one finds 132 singlets built, starting from the angular addition of 12 spin-1/2, many of which are low energy states. This crude picture makes it possible to understand why there are so

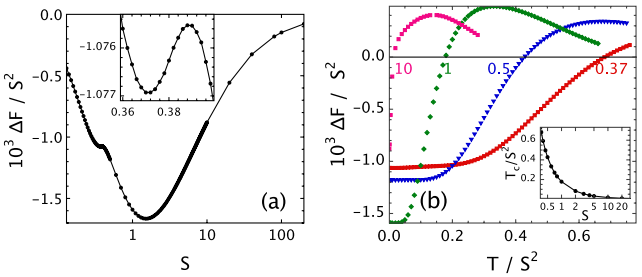

FIG. 4 (color online). (a) Difference of SBMFT free energy between the *cuboc1* and $\sqrt{3} \times \sqrt{3}$ *Ansätze* as a function of $\mathcal{S}$ at $T = 0$. Inset: zoom of the domain around critical spins $\mathcal{S}_c$. (b) Same quantity as a function of T for different $\mathcal{S}$. Inset: value of $T_c/\mathcal{S}^2$ versus $\mathcal{S}$ (see the text).

many (63) singlet states below the triplet gap of the $N = 36$ sample. Moreover, this property may explain, through resonances, the stabilization of *cuboc1* relative to the $\sqrt{3} \times \sqrt{3}$ SRO.

On the other hand, with the hypothesis of a chiral spin liquid, we expect an eightfold GS degeneracy on a 2 torus (a factor of 2 for the chirality times a factor of 4 for the topological degeneracy) at the thermodynamic limit. Thus, the large number of low-lying singlets seen in ED spectra should be restricted to small size samples. Such an evolution has already been observed in the J_1-J_2 model on the triangular lattice in the parameter range where the classical ground state has a 4-sublattice unit cell [28]. Note that the DMRG results do not exhibit a large number of singlet states below the triplet gap [8].

Spin-1/2 ED results on the $N = 36$ sample are also compatible with *cuboc1* short-range order: (i) the first $S = 1$ state is at the softest k vector of the *cuboc1* short-range order, as can be seen in Fig. 4 of Ref. [6]; (ii) the dynamical and static structure factors [29] have relatively larger values at the wave vector of the *cuboc1* order than near the quasisoft points of the $\sqrt{3} \times \sqrt{3}$ and $\mathbf{q} = 0$ orders. Thus, correlation functions in large-scale DMRG computations and/or characterization of low energy excitations by ED for 48 site samples would be an essential complement to further support or discard the present proposal.

Moreover, the first spin-1/2 states of small samples (with an odd-integer number of sites) have nonzero Chern numbers [4]. This quantum number, first introduced in such a context by Haldane and Arovas [30], is a topological index (and thus a robust property) characterizing the chiral character of a wave function. This property, which has never been explained in other approaches, could be understood for chiral spinons [15]. In the same spirit, the classical chirality defined by the determinant of three spins (nonzero on hexagons for the classical *cuboc1*) is generalized for quantum spins as the imaginary part of the cyclic permutation operator of spins on closed contours [14]. This quantity (Wilson loop operator), computed in the ED GS for different contours, obeys the law expected for a chiral liquid [24].

We have shown that, within mean-field Schwinger boson approximation, the kagome antiferromagnet has a chiral ground state. Spin-1/2 exact results both from ED and DMRG give some support to this hypothesis. In such a system, we expect a low temperature chiral symmetry breaking phase with topologically protected edge states, as in the quantum Hall systems. The extent of this low temperature phase, the nature of the phase transition, and the role of defects—questions already addressed in classical systems [31,32]—remain open questions.

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