Spin- $\frac{1}{2}$ Heisenberg antiferromagnet on an anisotropic kagome lattice

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We use the coupled-cluster method to study the zero-temperature properties of an extended two-dimensional Heisenberg antiferromagnet formed from spin- $\frac{1}{2}$ moments on an infinite spatially anisotropic kagome lattice of corner-sharing isosceles triangles, with nearest-neighbor bonds only. The bonds have exchange constants $J_1 > 0$ along two of the three lattice directions and $J_2 \equiv \kappa J_1 > 0$ along the third. In the classical limit, the ground-state (GS) phase for $\kappa < \frac{1}{2}$ has collinear ferrimagnetic (Néel') order where the J_2 -coupled chain spins are ferromagnetically ordered in one direction with the remaining spins aligned in the opposite direction, while for $\kappa > \frac{1}{2}$ there exists an infinite GS family of canted ferrimagnetic spin states, which are energetically degenerate. For the spin- $\frac{1}{2}$ case, we find that quantum analogs of both these classical states continue to exist as stable GS phases in some regions of the anisotropy parameter κ , namely, for $0 < \kappa < \kappa_{c1}$ for the Néel' state and for (at least part of) the region $\kappa > \kappa_{c2}$ for the canted phase. However, they are now separated by a paramagnetic phase without either sort of magnetic order in the region $\kappa_{c1} < \kappa < \kappa_{c2}$, which includes the isotropic kagome point $\kappa = 1$ where the stable GS phase is now believed to be a topological ($\mathbb{Z}_2$) spin liquid. Our best numerical estimates are $\kappa_{c1} = 0.515 \pm 0.015$ and $\kappa_{c2} = 1.82 \pm 0.03$.

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I. INTRODUCTION

Low-dimensional quantum magnets, especially those defined on regular two-dimensional (2D) lattices, have been the subject of intense study in recent years (and see, e.g., Refs. 1 and 2 for recent reviews). In particular, it is known that highly frustrated 2D quantum antiferromagnets display a bewilderingly rich panoply of ground-state (GS) phases, which often have no classical counterparts. Examples include various valence-bond crystalline and spin-liquid phases. Among the parameters that determine the zero-temperature ($T = 0$) phase diagram of such systems are the dimensionality and structure (e.g., the coordination number) of the crystallographic lattice on whose sites the magnetic ions are situated, the spin quantum number s of the ions, and the type and range of the magnetic bonds between the ions that often compete for differing forms of order, thereby leading to frustration. We have learned too that the quantum versions of classical models that have massively degenerate ground states, especially those with a nonzero ($T = 0$) GS entropy, are prime candidates for systems with novel GS phases.

Among all such candidate spin-lattice systems, therefore, those with periodic arrays of vertex-sharing structures, each of which is itself magnetically frustrated, occupy a special niche. These include the three-dimensional (3D) pyrochlore lattice of vertex-sharing tetrahedra and the 2D kagome lattice of corner-sharing triangles. Of these, the spin- $\frac{1}{2}$ Heisenberg antiferromagnet (HAF) on the 2D kagome lattice has been the

subject of intensive study in recent years.^{2–52} Even after several decades of research, the nature of the GS phase of the spin- $\frac{1}{2}$ HAF on the spatially isotropic kagome lattice has remained uncertain until very recently. Various outcomes, ranging from states with magnetic order to valence-bond solids or quantum spin liquids of different types, have been proposed.

Although many such studies agree on the finding that the GS phase lacks magnetic long-range order (LRO), there has remained uncertainty over its precise character. For example, some studies have favored a gapless critical spin liquid of one type or another, while others have favored one or other valence-bond crystals with an abundance of low-lying spin-singlet excited states. Only in the last year or so has compelling numerical evidence been provided,³⁸ due to advances in the density-matrix renormalization group (DMRG) technique, that the ground state is both gapped and is without any signal of either valence-bond or magnetic order at the largest finite-size systems that could be studied. In the past few months, further convincing evidence has come from different large-scale DMRG studies^{49,52} that this GS phase is indeed a topological ($\mathbb{Z}_2$) spin liquid, as we discuss further in Sec. V when we discuss our own results. We should note, however, that despite these recent findings, no final consensus has yet been reached within the community as to whether the spin-liquid ground state is actually gapped or gapless.

Theoretical interest in spin- $\frac{1}{2}$ kagome HAFs heightened considerably in the last few years with the discovery of

several candidate materials for their experimental realizations. Chronologically, the first promising such candidate was the mineral herbertsmithite (also known as Zn-paratacamite) γ - $\text{Cu}_3\text{Zn}(\text{OH})_6\text{Cl}_2$,^{53–55} for which it has been shown that the spin- $\frac{1}{2}$ Cu^{2+} ions are antiferromagnetically coupled and lie on the vertices of well-separated and structurally undistorted kagome-lattice planes. Although the underlying kagome planes in herbertsmithite appear to be essentially structurally perfect, there does appear to be an appreciable amount of antisite disorder due to a mixing of the spin- $\frac{1}{2}$ Cu^{2+} ions and the diamagnetic Zn^{2+} ions between the Cu and Zn sites. This disorder acts to introduce a coupling between the kagome planes, thereby effectively destroying the local 2D nature of the system. Thus, while herbertsmithite is structurally perfect, these impurities, together with a spin-orbit coupling that can be modeled by a Dzyaloshinskii-Moriya interaction with a non-negligible strength parameter, act to complicate the comparison of theory with experiment. As a consequence, herbertsmithite has lost some of its initial promise as an almost perfect, spin- $\frac{1}{2}$, isotropic kagome HAF.

A more recently discovered candidate for that role is another member of the atacamite family, namely, the polymorph kapellasite α - $\text{Cu}_3\text{Zn}(\text{OH})_6\text{Cl}_2$ (Refs. 56–59) of herbertsmithite. Although they share the same chemical composition, the two minerals have a different crystallographic structure. Interestingly, however, they both display distinct kagome structures, although in different ways. Thus, in kapellasite the spin- $\frac{1}{2}$ kagome lattice is obtained by the regular doping of a 2D triangular Cu^{2+} metal-site sublattice with diamagnetic Zn^{2+} ions. By contrast, in herbertsmithite, it is obtained by a similar diamagnetic dilution with Zn^{2+} ions of the 3D pyrochlorelike sublattice. It is asserted⁵⁹ for kapellasite that while the Cu/Zn mixing leads to some intralayer disorder within the kagome planes, it can not induce any appreciable interlayer coupling, unlike in herbertsmithite.

It should be noted, however, that a theoretical electronic study using density functional theory (DFT) within the local density approximation,⁵⁷ of both the material kapellasite and its relative haydeeite $\text{Cu}_3\text{Mg}(\text{OH})_6\text{Cl}_2$, has revealed significant non-nearest-neighbor (non-NN) exchange coupling strengths, especially those corresponding to bonds across the diagonals of the hexagons on the kagome lattice. Furthermore, recent high-temperature series expansion fits to the measured dc magnetic susceptibility $\chi_{\text{dc}}(T)$, as a function of temperature T , for kapellasite,⁵⁹ seem to give a nearest-neighbor exchange interaction (J_1) on the kagome planes that is *ferromagnetic* in nature (i.e., $J_1 < 0$), with the overall antiferromagnetic behavior of the material explained by large positive further-neighbor interactions.

A spatially anisotropic version of the spin- $\frac{1}{2}$ kagome HAF has also been suggested to have been realized experimentally in the minerals volborthite $\text{Cu}_3\text{V}_2\text{O}_7(\text{OH})_2 \cdot 2\text{H}_2\text{O}$ (Refs. 60–66) and vesignieite $\text{BaCu}_3(\text{VO}_4)_2(\text{OH})_2$.^{64,66–69} In both of these materials, the Cu sites form a slightly distorted kagome network, resulting in two inequivalent Cu sites per triangle, namely, one Cu1 site and two Cu2 sites per triangle. For example, volborthite has a monoclinic distortion that deforms the equilateral triangles of the isotropic kagome network into isosceles triangles. In this material, the difference in

the Cu1–Cu2 and Cu2–Cu2 bond lengths is about 3%. In turn, theoretical modeling of the thermodynamic properties then leads to a suggested difference between two of the NN (Cu1–Cu2) exchange constants (J_1) and the third (Cu2–Cu2) one (J'_1) on each triangle of the kagome lattice. In volborthite, this magnetic anisotropy is around 20%. The anisotropy is much less pronounced in vesignieite where the difference in bond length is less than 0.1%, and the material is closer to being structurally isotropic. Another recently discovered spin- $\frac{1}{2}$ deformed kagome-lattice antiferromagnet is the material $\text{Rb}_2\text{Cu}_3\text{SnF}_{12}$.^{70–72}

Although both volborthite and vesignieite have a reduced symmetry compared with the structurally perfect herbertsmithite, they do offer some advantages. As noted above, this latter compound shows antisite disorder with up to 10% of the magnetic Cu^{2+} ions exchanged by Zn^{2+} ions, thereby leading to a weak interlayer magnetic coupling between the kagome planes as well as magnetic vacancies within them. By contrast, in both volborthite and vesignieite, their intermediate layers between the kagome planes contain V^{5+} ions, and hence antisite disorder of the Cu ions is prevented. Although vesignieite is much less anisotropic than volborthite, it suffers in practice, like herbertsmithite, from low sample quality. One of the main experimental advantages of studying the more anisotropic volborthite over either of herbertsmithite or vesignieite is that it is much easier to prepare with fewer impurities. Nevertheless, we note that the nature of the magnetic couplings in this material has been questioned in a recent study,⁷³ where it is pointed out that the local environments of the two inequivalent types of Cu sites differ essentially in important ways. DFT is then used to show that volborthite should not be modeled as an anisotropic J_1 – J'_1 kagome-lattice HAF, but rather as a J'_1 – J'_2 – J_1 model, more reminiscent of coupled *frustrated* chains, in which two-thirds of the kagome sites (viz., the Cu2 sites) are considered as J'_1 – J'_2 chains [i.e., with *ferromagnetic* NN exchange $J'_1 < 0$ and frustration induced by next-nearest-neighbor (NNN) exchange $J'_2 > 0$], and with the chains coupled via NN exchange bonds of strength J_1 between the Cu2 and remaining Cu1 sites.

Although it is thus often very uncertain as to whether a given real material does or does not provide an experimental realization of a particular theoretical model, such as the kagome-lattice HAFs considered here, there is still much to be gained by a systematic theoretical comparison between such models. It is of particular interest in such comparisons to use, wherever possible, the same theoretical technique. Among the relatively few widely applicable and systematically improvable (within a well-defined hierarchical approximation scheme) such tools is the coupled-cluster method (CCM).^{74–78} Our intention here is to use the CCM to further the study of the HAF, with NN interactions only, on an anisotropic kagome lattice.^{79–84}

By now, the CCM has been used to study a huge number of quantum spin-lattice problems (see, e.g., Refs. 19,36,48,76–78,85–106 and references cited therein). Among these, of particular interest here are applications of the CCM to the frustrated spin- $\frac{1}{2}$ J_1 – J_2 HAF model on the square lattice^{87,94–96,100} with NN bonds (of strength $J_1 > 0$) competing with NNN bonds (of strength $J_2 > 0$), and various models related to it

FIG. 1. (Color online) (a) The interpolating kagome-square model with (black) solid bonds J_1 , (green) dashed bonds J'_1 , and (red) dashed-dotted bonds J_2 , showing the canted state, and the equivalent anisotropic kagome model when $J'_1 = 0$, showing (b) the coplanar ferrimagnetic canted state and (c) the collinear ferrimagnetic semistriped state. In all cases, the (blue) arrows represent spins located on lattice sites (solid bullets).

by removal of some of the NNN J_2 bonds. When half of the J_2 bonds are removed, these include an interpolating square-triangle HAF (or spatially anisotropic triangular HAF),⁹⁸ the Union Jack lattice model,¹⁰¹ and the anisotropic planar pyrochlore (or checkerboard) HAF (also known as the crossed chain model).¹⁰⁶

A further modification of the original square-lattice J_1 - J_2 model is now to remove another half of the J_2 bonds in such a way as to leave half of the fundamental square plaquettes with one J_2 bond and the other half with none. One way of doing this in a regular fashion results in the Shastry-Sutherland model¹⁰⁷ in which no J_2 bonds meet at any lattice site and every site is five-connected (by four NN J_1 bonds and one J_2 bond). The CCM has also been successfully applied to this model.^{91,99}

Another similar such model, of particular interest here, arises from removing alternate diagonal lines of J_2 bonds from the interpolating square-triangle HAF (which itself arises from the square-lattice J_1 - J_2 HAF model by removing all of the diagonal lines of J_2 bonds in the same direction). It differs from the Shastry-Sutherland model primarily in that the square lattice now breaks into two square sublattices of A and B sites, respectively, such that the A sites are all six-connected (by four NN J_1 bonds and two NNN J_2 bonds), while the B sites are all four-connected (by four NN J_1 bonds only). A particularly relevant generalization of the model for present purposes arises from introducing an additional anisotropy in the NN bonds such that along alternating rows and columns, the NN bonds are allowed to have the strength J'_1 , as shown in Fig. 1(a). Clearly, when $J'_1 = 0$, the model reduces to the anisotropic kagome-lattice HAF, shown equivalently in Fig. 1(b), which is the subject of this paper.

The spin- $\frac{1}{2}$ interpolating kagome-square model described above and shown in Fig. 1(a) was studied by us in an earlier paper, using the CCM.³⁶ In that paper, we were mainly interested in the quantum phase transition line in the J'_1 - J_2 plane (with $J_1 \equiv 1$) in the model between the two quasiclassical states with antiferromagnetic Néel order and ferrimagnetic canted order. By contrast, in this paper we focus purely on the $J'_1 \equiv 0$ case corresponding to the anisotropic kagome-lattice HAF that has been suggested as one possibility to describe the magnetic properties of volborthite

and vesignieite, as discussed above. Our main aim is not so much to shed light on the structure of the paramagnetic GS phase of the isotropic spin- $\frac{1}{2}$ kagome-lattice HAF, but to determine the boundaries of this phase as the anisotropy is varied.

After first describing the model in Sec. II, we apply the CCM to investigate its GS properties. The CCM is itself described briefly in Sec. III, and our results are then presented in Sec. IV. We conclude in Sec. V with a discussion of the results.

II. THE MODEL

As mentioned in Sec. I, in a previous paper³⁶ we considered a depleted (and anisotropic) variant of the archetypal and much-studied J_1 - J_2 model in which three-quarters of the J_2 bonds are removed from it in the pattern shown in Fig. 1(a). The square-lattice representation of the model shown in Fig. 1(a) contains the two square sublattices of A and B sites, respectively, and each of these in turn contains the two square sublattices of A_1 and A_2 , and B_1 and B_2 sites, respectively, as shown. It is very illuminating to consider the anisotropic variant (viz., the interpolating kagome-square model or J_1 - J'_1 - J_2 model) in which half of the J_1 bonds are allowed to have the strengths $J'_1 > 0$ along alternating rows and columns, as shown in Fig. 1(a). All of the bonds joining sites i and j are of standard Heisenberg type, i.e., proportional to $\mathbf{s}_i \cdot \mathbf{s}_j$, where the operators $\mathbf{s}_k = (s_k^x, s_k^y, s_k^z)$ are the quantum spin operators on lattice site k , with $\mathbf{s}_k^2 = s(s+1)$ and $s = \frac{1}{2}$ for the quantum case considered here.

The spin- $\frac{1}{2}$ HAF's on the 2D kagome and square lattices are represented, respectively, by the limiting cases $\{J_1 = J_2, J'_1 = 0\}$ and $\{J_1 = J'_1, J_2 = 0\}$. The limiting case $\{J_1 = J'_1 = 0; J_2 > 0\}$ represents a set of uncoupled one-dimensional (1D) HAF chains. The case $J'_1 = 0$ with $J_2 \neq J_1$ represents a spatially anisotropic kagome HAF considered recently by other authors,^{79–84} especially in the quasi-1D limit where $J_2/J_1 \gg 1$.^{82,84} It is this latter model where $J'_1 = 0$ that is considered here (although, for technical reasons, we note that we actually set J'_1 to be a small positive value, henceforth chosen to be $J'_1 = 10^{-5}$). Our model is thus equivalently shown

in the kagome-lattice geometry of Fig. 1(b). It thus comprises parallel chains of spins on A sites coupled in a NN fashion along the chains by bonds of strength J_2 , with the chains then cross linked via B_2 sites with NN bonds of strength J_1 . The totality of sites arranged in a regular kagome lattice on which each component triangle is comprised of two J_1 bonds and one J_2 bond.

The Hamiltonian of the resulting anisotropic kagome-lattice model is thus

$$H = J_1 \sum_{\substack{\langle i,j \rangle \\ i \in A, j \in B_2}} \mathbf{s}_i \cdot \mathbf{s}_j + J_2 \sum_{\substack{\langle i,j \rangle \\ i \in A_1, j \in A_2}} \mathbf{s}_i \cdot \mathbf{s}_j, \quad (1)$$

where the sum on $\langle i, j \rangle$ runs over all NN pairs (of the sort specified in each sum), counting each bond once and once only. Henceforth, we consider the model where the spins on all lattice sites have spin quantum number $s = \frac{1}{2}$, and where both types of bonds are antiferromagnetic in nature (i.e., $J_1 \geq 0$, $J_2 \geq 0$) and hence act to frustrate one another. With no loss of generality, we may then choose the energy scale by setting $J_1 \equiv 1$. We are interested in the infinite-lattice limit $N_K \equiv \frac{3}{4}N \rightarrow \infty$, where N_K is the number of sites on the kagome lattice and N is the number of sites on the square lattice of Fig. 1(a) before the $(\frac{1}{4}N)$ B_1 sites have been removed.

Considered as a classical model (corresponding to the case where the spin quantum number $s \rightarrow \infty$), the interpolating kagome-square model of Fig. 1(a) (i.e., with $J'_1 \neq 0$) has only two GS phases separated by a continuous (second-order) phase transition at $J_2 = J_2^{\text{cl}} \equiv \frac{1}{2}(J_1 + J'_1)$. For $J_2 < J_2^{\text{cl}}$, the system is Néel ordered on the square lattice, while for $J_2 > J_2^{\text{cl}}$ the system has noncollinear (but coplanar) canted order as shown in Fig. 1(a), in which the spins on each of the A_1 and the A_2 sites are canted, respectively, at angles $(\pi \mp \phi)$ with respect to those on the B sublattice, all of the latter of which point in the same direction. The lowest-energy state in the canted phase is obtained with $\phi = \phi_{\text{cl}} \equiv \cos^{-1}(J_2^{\text{cl}}/J_2)$. The Néel state, for $J_2 < J_2^{\text{cl}}$, simply corresponds to the case $\phi_{\text{cl}} = 0$.

For the case of the anisotropic kagome-lattice HAF considered here (i.e., with $J'_1 = 0$), the classical ($s \rightarrow \infty$) ground states are those spin configurations that satisfy the condition that for each elementary triangular plaquette of the kagome lattice in Fig. 1(b) the energy is minimized. If we take the angle ϕ to be such that the middle spin (viz., that on a B_2 site) of a given triangular plaquette forms angles $(\pi \pm \phi)$ with the other two (chain) spins of the same plaquette (viz., those on A_2 and A_1 sites, respectively), the total energy of the lattice, for classical spins of length s , is $E = \frac{2}{3}N_K s^2 [2J_1 \cos(\pi - \phi) + J_2 \cos(2\phi)]$.

For $J_2 < \frac{1}{2}J_1$ this energy is minimized with $\phi = 0$, and the classical GS phase is thus collinear and unique, with the spins (on the A sites) along the J_2 -bond chains aligned in one direction and the remaining spins (on the B_2 sites) on the kagome lattice aligned in the opposite direction. As a convenient shorthand notation, we henceforth refer to this collinear ferrimagnetic state as the Néel' state. Indeed, the Néel' state of the anisotropic kagome-lattice HAF is precisely equivalent to the Néel state of the interpolating kagome-square model of Fig. 1(a) (i.e., before the removal of the B_1 sites in the limiting case $J'_1 = 0$) from which it is derived. The total spin of this classical collinear Néel' ferrimagnetic state is thus

$S_{\text{tot}} = \frac{1}{3}N_K s$ where each spin has magnitude s . In terms of the saturation magnetization (i.e., in the ferromagnetic state with all spins aligned in the same direction), $M_{\text{sat}} \equiv N_K s$, the total magnetization in this collinear ferrimagnetic state is $M^{\text{tot}} = \frac{1}{3}M_{\text{sat}}$. For the quantum case, the Marshall-Lieb-Mattis theorem^{108,109} may also be used to show, for the limiting case $J_2 = 0$ only, that the exact ground state has the same value $S_{\text{tot}} = \frac{1}{3}N_K s$ of the total spin as its classical counterpart.

By contrast, for $J_2 > \frac{1}{2}J_1$, the classical GS energy is minimized with the canting angle $\phi = \phi_{\text{cl}} \equiv \cos^{-1}(\frac{J_1}{2J_2})$. We expect that coplanar canted states will then be favored by either thermal or quantum fluctuations, and henceforth we only consider coplanar states from among this degenerate manifold that includes noncoplanar states. The total magnetization of this canted ferrimagnetic state is $M^{\text{tot}} = \frac{1}{3}(2 \cos \phi - 1)N_K s = \frac{1}{3}(\frac{J_1}{J_2} - 1)M_{\text{sat}}$. The ground state of the HAF on the isotropic kagome lattice (i.e., with $J_2 = J_1$) falls in this regime, and has the canting angle $\phi = \frac{\pi}{3}$ demanded by symmetry. Only for this case does the total classical magnetization vanish, $M^{\text{tot}} = 0$. The classical ensemble of degenerate coplanar states is now characterized by two variables for each triangular plaquette, namely, the angle ϕ , and the two-valued chirality variable $\chi = \pm 1$ that defines the direction (anticlockwise or clockwise) in which the spins turn as one transverses the plaquette in the positive (anticlockwise) direction. For a given value of $J_2 > \frac{1}{2}J_1$, the different degenerate canted states arise from the various possible ways to assign positive or negative chiralities to the triangular plaquettes of the lattice. (Appendix A of Ref. 80 gives a good description of the constraints that these chiralities need to satisfy.)

We note that the well-known $q = 0$ and $\sqrt{3} \times \sqrt{3}$ states of the (isotropic) kagome-lattice HAF are the special cases, respectively, where all of the chiralities are the same, and where basic triangular plaquettes joined by a vertex have opposite chiralities. The $q = 0$ state is one in which the spins on each of the three sublattices (of A_1 , A_2 , and B_2 sites, respectively) are parallel to one another, and make an angle of 120° with the spins on the other two sublattices, while the $\sqrt{3} \times \sqrt{3}$ state contains nine sublattices and is obtained by deleting $\frac{1}{4}$ of the sites (viz., the B_1 sites) of the ordered spins of a triangular lattice to form the kagome lattice (and see also, e.g., Refs. 4, 9, and 43 for further details). Clearly, the limiting case $J'_1 \rightarrow 0$ of the classical ground state of the interpolating kagome-square model shown in Fig. 1(a) is just the state shown in Fig. 1(b), which corresponds to the $q = 0$ state for the isotropic ($J_2 = J_1$) kagome-lattice HAF.

The HAF on the isotropic kagome lattice (i.e., with $J_2 = J_1$) is especially interesting since for this case, with $\phi = \frac{\pi}{3}$, the number Ω of degenerate classical spin configurations grows exponentially with the number N_K of spins, so that even at zero temperature the system has a nonzero value of the entropy per spin. A previous high-order CCM study⁴⁸ of the isotropic kagome-lattice HAF showed that for the extreme quantum case $s = \frac{1}{2}$, the $q = 0$ state is energetically favored over the $\sqrt{3} \times \sqrt{3}$ state, while for any $s > \frac{1}{2}$ the $\sqrt{3} \times \sqrt{3}$ state is selected over the $q = 0$ state. For both the $\sqrt{3} \times \sqrt{3}$ and the $q = 0$ states, it was further found that the magnetic order is strongly suppressed by quantum fluctuations. In particular, the

order parameter (viz., the average local onsite magnetization or sublattice magnetization) M_K was found to vanish for both $s = \frac{1}{2}$ and 1, while nonzero values for M_K were found for $s = \frac{3}{2}, 2, \frac{5}{2}$, and 3.

By contrast, for the anisotropic case (with $J_2 \neq J_1$), the classical degeneracy Ω has been shown⁸⁰ to grow exponentially with $\sqrt{N_K}$ [i.e., $\Omega \propto \exp(c\sqrt{N_K})$], so that the GS entropy per spin vanishes in the thermodynamic limit. Clearly, since in the limit $J_2 \rightarrow J_1$ the anisotropic model approaches the isotropic model, the anisotropic model must have an appropriately large number of low-lying excited states that become degenerate with the ground state in the isotropic limit $J_2 \rightarrow J_1$.

Of course, it is not clear, once the kagome lattice is allowed to become anisotropic (i.e., with $J_2 \neq J_1$), that the $q = 0$ state should necessarily remain the lowest-energy state among the (subextensive) ensemble of classically degenerate states. Continuity would clearly suggest, however, that this should be the case as long as J_2 is not too different from J_1 . We have partially checked this within our own CCM calculations by showing that the $q = 0$ state remains lower in energy than the $\sqrt{3} \times \sqrt{3}$ state, for example, over the range of anisotropy studied here.

We also note that as $J_2 \rightarrow \infty$, the classical canting angle $\phi_{cl} \rightarrow \frac{1}{2}\pi$, and the spins on the A sublattice chains become antiferromagnetically ordered, as is expected, and these spins are orientated at 90° to those on the B_2 sublattice. For the particular ordering in Fig. 1(b), which arises from that in Fig. 1(a) in the limit $J'_1 \rightarrow 0$, the spins on the B_2 sublattice are themselves parallel and hence ferromagnetically ordered. Of course, there is complete degeneracy at the classical level in this decoupled-chain limit (i.e., when $J_2 \rightarrow \infty$) between all states for which the relative ordering directions for spins on the A and B_2 sublattices are arbitrary. In the same limit, the quantum spin- $\frac{1}{2}$ problem considered here should also comprise decoupled antiferromagnetic chains on the A-sublattice sites. We expect that this degeneracy in relative orientation might be lifted by quantum fluctuations by the well-known phenomenon of *order by disorder*.¹¹⁰ Since it is also true that quantum fluctuations generally favor collinear ordering, a preferred state is thus likely to be the so-called ferrimagnetic semistriped state shown in Fig. 1(c) where the A sublattice is now Néel ordered in the same direction as the B_2 sublattice is ferromagnetically ordered. [Note that in the square-lattice geometry of Fig. 1(a) and where the B_1 sites and the J'_1 bonds are retained, alternate rows (and columns) are thus ferromagnetically and antiferromagnetically ordered in the same direction in the semistriped state, which is the origin of its name.]

III. COUPLED-CLUSTER METHOD

We now apply the CCM (see, e.g., Refs. 74–78 and references cited therein) to the spin- $\frac{1}{2}$ anisotropic kagome-lattice HAF discussed in Sec. II above. At a very general level, the method provides one of the most versatile techniques now available in quantum many-body theory. At attainable levels of computational implementation, it has been shown to provide some of the most accurate results ever obtained for a large number of quantum many-body systems in quantum

chemistry, as well as in condensed matter, atomic, molecular, nuclear, and subnuclear physics.^{74,75} More specifically for present purposes, it has been very successfully applied by now to a large number of systems of interest in quantum magnetism (see, e.g., Refs. 19,36,48,76–78,85–106 and references cited therein), as we have already noted in Sec. I.

The method of applying the CCM to quantum magnets has been described in detail elsewhere (see, e.g., Refs. 19, 76, 78, 88, and 92, and references cited therein). It relies on building multispin correlations on top of a suitably chosen, normalized, GS model (or reference) state $|\Phi\rangle$ in a systematic hierarchy of approximations that we described below. The reference state $|\Phi\rangle$ is required only to be a fiducial vector for the system in the sense that all possible states of the system can be described in terms of it as a linear combination of states obtained from it by acting on it with members of some suitably chosen complete set of mutually commuting multispin creation operators $C_I^+ \equiv (C_I^-)^\dagger$. In this way, $|\Phi\rangle$ acts as a generalized vacuum state with respect to the set of operators $\{C_I^+\}$. It is often chosen as a classical ground state of the model under investigation, and for the present anisotropic kagome-lattice HAF we use mainly the canted (coplanar) ferrimagnetic state shown in Fig. 1(b) (including the collinear Néel' state, which is its limiting form when the canting angle $\phi \rightarrow 0$), although we also discuss briefly in Sec. V the use of the semistriped (collinear) ferrimagnetic state shown in Fig. 1(c) as a CCM model state.

Once the set $\{|\Phi\rangle, C_I^+\}$ has been suitably chosen, the exact GS ket and bra wave functions of the system are parametrized within the CCM, in terms of them, in the *exponential* forms that are the hallmark of the method,

$$|\Psi\rangle = e^S |\Phi\rangle, \quad S = \sum_{I \neq 0} S_I C_I^+, \quad (2)$$

$$\langle\tilde{\Psi}| = \langle\Phi|\tilde{S}e^{-S}, \quad \tilde{S} = 1 + \sum_{I \neq 0} \tilde{S}_I C_I^-, \quad (3)$$

where we define $C_0^+ \equiv 1$. It is clear from Eqs. (2) and (3) that the normalization has been chosen so that $\langle\tilde{\Psi}|\Psi\rangle = \langle\Phi|\Phi\rangle = 1$. The complete set of GS CCM correlation coefficients $\{S_I, \tilde{S}_I\}$ ($\forall I \neq 0$) is then obtained by the requirement that the states $|\tilde{\Psi}\rangle$ and $|\Psi\rangle$ obey the GS Schrödinger equations $\langle\tilde{\Psi}|H = E\langle\tilde{\Psi}|$ and $H|\Psi\rangle = E|\Psi\rangle$, respectively. The resulting equations

$$\langle\Phi|C_I^- e^{-S} H e^S |\Phi\rangle = 0, \quad \forall I \neq 0 \quad (4)$$

$$\langle\Phi|\tilde{S}e^{-S}[H, C_I^+]e^S |\Phi\rangle = 0, \quad \forall I \neq 0 \quad (5)$$

may, completely equivalently, be derived from the requirement that the GS energy functional

$$\bar{H} \equiv \langle\tilde{\Psi}|H|\Psi\rangle \quad (6)$$

be stationary with respect to variations in all members of the set $\{S_I, \tilde{S}_I; I \neq 0\}$.

Once the CCM correlation coefficients have been found from solving Eqs. (4) and (5), it is easy to see that the GS energy E is given purely in terms of the ket-state coefficients $\{S_I\}$ as $E = \langle\Phi|e^{-S} H e^S |\Phi\rangle$. Clearly, however, for a more general operator $\hat{O}$, the evaluation of its GS expectation value $\bar{O} \equiv \langle\tilde{\Psi}|\hat{O}|\Psi\rangle$ requires knowledge of the set of bra-state correlation

coefficients $\{\tilde{S}_I\}$ as well as of the corresponding set of ket-state coefficients $\{S_I\}$.

It is very convenient in practice to perform a rotation of the local spin axes of each of the spins in the system (i.e., we define a different set of spin axes on every lattice site) such that all spins in the reference state align along the negative z axes of the local coordinates. In practice, this simply means that the Hamiltonian has to be reexpressed in terms of these local axes for each choice of reference state used. The big advantage of so doing is that in these local coordinates we have

$$|\Phi\rangle = |\downarrow\downarrow\downarrow\dots\rangle, \quad C_I^+ = s_{k_1}^+ s_{k_2}^+ \dots s_{k_n}^+, \quad n = 1, 2, 3, \dots \quad (7)$$

where $s_k^+ \equiv s_k^x + i s_k^y$, the indices k_n denote arbitrary lattice sites, and the components of the spin operators are defined in the local rotated coordinate frames. We note that for spins of quantum number s , each site index k_n in each multispin configuration set index $I = \{k_1, k_2, \dots, k_n\}$ in Eq. (7) can be repeated up to a maximum of $2s$ times. Thus, for the present case $s = \frac{1}{2}$, all individual spin-site indices in each set index I are different.

The magnetic order parameter (viz., the average local onsite magnetization) is now given by $M = -\frac{1}{N} \sum_{i=1}^N \langle \tilde{\Psi} | s_i^z | \Psi \rangle$, where s_i^z is expressed in the local spin coordinates defined above, and $N (\rightarrow \infty)$ is the number of lattice sites. We denote by M here the order parameter defined for the general interpolating kagome-square model of Fig. 1(a) (i.e., before the $\frac{1}{4}N$ number of B_1 sites have been removed to yield the anisotropic kagome model). The corresponding order parameter M_K for the anisotropic kagome-lattice HAF considered here is similarly given by $M_K = -\frac{1}{N_K} \sum_{i=1}^{N_K} \langle \tilde{\Psi} | s_i^z | \Psi \rangle$, where the sum is taken over only the kagome-lattice sites. Thus, in the limiting case $J'_1 = 0$ considered here of the anisotropic kagome-square model, the spins on the non-kagome B_1 sites are frozen to have their spins aligned exactly along their local negative z axis. Hence, for the anisotropic kagome-lattice limit (i.e., when $J'_1 = 0$) of the interpolating kagome-square model, we have the simple relation

$$M_K = \frac{4}{3} M - \frac{1}{6}. \quad (8)$$

The parametrizations of Eqs. (2) and (3) yield, in principle, the exact GS eigenstate when the complete sets of multispin creation and destruction operators C_I^+ and C_I^- , respectively, is retained. In practice, of course, it is necessary to make approximations by truncating the complete set of multispin configuration set indices $\{I\}$. In that case, the results for physical quantities such as the GS energy E and order parameter M will naturally depend both on the particular truncation (i.e., on the configurations specified by the set indices I that are retained), as well as on the specific choice of model state to which those multispin configurations are referred. We note, however, that the CCM always exactly obeys the Goldstone linked-cluster theorem at every such level of approximation.⁷⁵ Thus, the CCM approach always yields results directly in the thermodynamic limit $N \rightarrow \infty$ from the outset, and no finite-size scaling is required. The results at all levels of approximation are guaranteed to be size extensive.

Although the CCM is fully (bi)variational, as discussed above in connection with the stationarity of $\tilde{H}$ in Eq. (6), we note, however, that it does not lead to strict upper bounds

for the energy due to the lack of explicit Hermiticity in the parametrizations of the GS ket and bra wave functions in Eqs. (2) and (3). This minor drawback is more than compensated for by the fact that the CCM parametrizations exactly obey the important Hellmann-Feynman theorem at all levels of approximation.⁷⁵

For the present spin- $\frac{1}{2}$ model, we employ the so-called LSUB m (or lattice-animal-based subsystem) approximation scheme to truncate the expansions of S and $\tilde{S}$ in Eqs. (2) and (3). In this very widely tested scheme, one includes from the full set of multispin configurations specified by the set indices I in Eqs. (2), (3), and (7) only those involving m or fewer correlated spins in all arrangements (or lattice animals in the language of graph theory) which span a range of no more than m contiguous lattice sites. In this context, a set of sites is defined to be contiguous if every site has at least one other in the set as a nearest neighbor, and where it is clearly necessary to include a definition in the geometry (or better, topology) of the lattice of which pairs of sites are considered to be NN pairs. For example, we choose here to work in the triangular-lattice geometry of the anisotropic kagome-square model in which the B sublattice sites of Fig. 1(a) are defined to have four NN sites joined to them by either J_1 bonds or J'_1 bonds, and the A sublattice sites are defined to have the six NN sites joined to them by J_1 , J'_1 , or J_2 bonds. If we had chosen instead to work in the square-lattice geometry, every site would have four NN sites. The former triangular-lattice geometry leads in the limiting case when $J'_1 = 0$ to the natural kagome-lattice geometry of Fig. 1(b) in which any two of the A_1 , A_2 , and B_2 sites forming the basic triangular plaquettes are considered to be NN pairs. Clearly, this would not be the case in the latter square-lattice geometry. For more details of the LSUB m scheme, the reader is referred, for example, to Refs. 76–78.

The astute reader will have noted the close connection between the CCM parametrization of the ket-state wave function given by Eqs. (2) and (7), when the multispin cluster configurations I are restricted to those between two spins only (as in the case of the LSUB m scheme used here with $m = 2$, where only NN pair correlations are included), with that of (lowest-order) self-consistent spin-wave theory (SWT).¹¹¹ Nevertheless, the two methods are still not identical, due partly to the lack of explicit Hermiticity between the CCM ket and bra parametrizations, as in Eqs. (2) and (3), and partly due to the way that self-consistency is incorporated within SWT. For example, in many cases where SWT is unstable (i.e., gives a negative magnetic order parameter M), the CCM LSUB2 result is usually stable (i.e., gives a positive value for M). The interested reader is referred to the literature¹¹² for a detailed discussion of the relationships between SWT and low-order implementations of the CCM.

Clearly, the LSUB m truncations scheme provides a fully systematic approximation hierarchy in the sense that each time the truncation index m is increased, more of the Hilbert space is sampled, such that as $m \rightarrow \infty$ the approximation becomes exact. Although, as we have already indicated, we never need to perform any finite-size scaling, since all CCM approximations are automatically performed from the outset in the infinite-lattice limit $N_K \rightarrow \infty$, where N_K is the number of lattice sites, we do still need as a last step in a CCM

calculation to extrapolate to the exact $m \rightarrow \infty$ limit in the LSUB m truncation index m , at which the complete (infinite) Hilbert space is reached. By now, there is a great deal of experience available regarding how one should extrapolate the GS energy per site $e_0(m) \equiv E(m)/N_K$ and the magnetic order parameter $M(m)$.

Thus, for the GS energy per spin $e_0(m)$, we use the well-tested empirical scaling ansatz (and see, e.g., Refs. 88,91,92, 94–96,98,100–102,106)

$$e_0(m) = a_0 + a_1 m^{-2} + a_2 m^{-4}. \quad (9)$$

For the GS magnetic order parameter, different extrapolation rules have been used depending on the features of the spin-lattice system at hand. For highly frustrated spin-lattice systems, a well-tested rule (and see, e.g., Refs. 94–96,100, 102–106) is

$$M(m) = b_0 + b_1 m^{-1/2} + b_2 m^{-3/2}. \quad (10)$$

An alternative rule that is useful, for example, for situations when there is some frustration present, but when it is not too large, is^{91,106}

$$M(m) = c_0 + c_1 m^{-\nu}, \quad (11)$$

which, as an advantage, leaves the leading exponent open for determination. The disadvantage of this scheme is clearly that it involves only the leading two terms in the asymptotic expansion, compared to the (inherently more accurate) three terms used in Eq. (10). Clearly, however, when the exponent ν in Eq. (11) is found by fitting to the CCM LSUB m results to be close to the value 0.5, as is usually found for highly frustrated systems, we can then revert to the more accurate form of Eq. (10).

In this paper we present extrapolated results based on LSUB m data sets with $m = \{2, 4, 6, 8\}$. To check the robustness of the extrapolation rules and to estimate the associated error bars in the extrapolations, we have also performed extrapolations using LSUB m data sets with $m = \{2, 4, 6\}$ and $\{4, 6, 8\}$. We find in general that the extrapolated results from all three sets of data are very similar.

The number of independent fundamental LSUB m clusters (i.e., those that are inequivalent under the symmetries of the Hamiltonian and of the model state) that are retained in the expansions of Eqs. (2) and (3) for the CCM correlation operators S and $\tilde{S}$ increases rapidly with the truncation index m . For example, the number of such fundamental clusters for the canted model state of the interpolating kagome-square model of Fig. 1(a) is 201 481 at the LSUB8 level of approximation in the triangular-lattice geometry used here where J_2 bonds are considered to join NN pairs, and this is the highest level for the present model that we have been able to attain with available computing power. In order to solve the corresponding coupled sets of CCM bra- and ket-state equations, we use an efficient parallelized CCM code,¹¹³ and typically employ around 600 processors simultaneously.

Finally, we note that our CCM calculations based on the canted phase of the anisotropic kagome-lattice HAF shown in Fig. 1(b) do not assume for the $s = \frac{1}{2}$ model considered here that the canting angle ϕ takes the same value $\phi_{cl} \equiv \cos^{-1}(\frac{J_1}{2J_2})$ as in the classical ($s \rightarrow \infty$) case. Rather, calculations are first performed for an arbitrary choice of canting angle ϕ .

We then minimize the corresponding LSUB m approximation for the energy $E_{\text{LSUB}m}(\phi)$ with respect to ϕ to yield the corresponding approximation to the quantum canting angle $\phi_{\text{LSUB}m}$. Generally (for $m > 2$), the minimization must be carried out computationally in an iterative procedure. Results for the canting angle $\phi_{\text{LSUB}m}$ are presented in Sec. IV.

IV. RESULTS

We now present our CCM results for the anisotropic spin- $\frac{1}{2}$ J_1 - J_2 HAF on the kagome lattice of Eq. (1), where we use the canted ferrimagnetic state shown in Fig. 1(a) (and with $J'_1 \rightarrow 0$) as the CCM model (or reference) state $|\Phi\rangle$ in the representations of Eqs. (2) and (3) of the exact GS ket and bra wave functions. Without loss of generality, but simply to set the energy scale, we henceforth set $J_1 \equiv 1$. (Equivalently, when more convenient to do so, we quote results in terms of the ratio $\kappa \equiv J_2/J_1$, assuming always that $J_1 > 0$.) We first show in Fig. 2 the GS energy per spin E/N_K as a function of the canting angle ϕ . Although results are shown at the LSUB4 level of approximation, results for other LSUB m levels are qualitatively similar. Curves such as those shown in Fig. 2 show that at this LSUB4 level of approximation, with $J_1 = 1$, the GS energy is minimized at $\phi = 0$ for $J_2 < J_2^{\text{LSUB4}} \approx 0.60$ and at a value $\phi \neq 0$ for $J_2 > J_2^{\text{LSUB4}}$. Hence, these first results indicate the possibility of a slight shift of the critical point to $J_2 = J_2^{cl} \equiv J_2^{\text{LSUB}\infty}$ between the quantum ferrimagnetic Néel' and canted phases, from the classical value $J_2^{cl} = 0.5$ when $J_1 = 1$. The fact that Néel' order survives here, at least at finite orders of LSUB m approximation, into the regime where it would be classically unstable against the formation of canted order is an example of a phenomenon that has

FIG. 2. (Color online) Ground-state energy per spin E/N_K of the spin- $\frac{1}{2}$ J_1 - J_2 HAF on the anisotropic kagome lattice of Eq. (1) (with $J_1 \equiv 1$), using the LSUB4 approximation of the CCM with the ferrimagnetic canted model state shown in Fig. 1(b), versus the canting angle ϕ for various selected values of the anisotropy parameter J_2 . For $J_2 \lesssim 0.6$, the minimum is at $\phi = 0$ (Néel' order) at this level of approximation, whereas for $J_2 \gtrsim 0.6$, the minimum occurs at $\phi = \phi_{\text{LSUB4}} \neq 0$, thereby providing evidence of a phase transition at $J_2 \approx 0.6$ in this approximation. We show results for those values of ϕ for which the corresponding CCM equations have real solutions.

FIG. 3. (Color online) The angle $\phi_{\text{LSUB}m}$ that minimizes the GS energy $E_{\text{LSUB}m}(\phi)$ of the spin- $\frac{1}{2}$ J_1 - J_2 HAF on the anisotropic kagome lattice of Eq. (1) (with $J_1 \equiv 1$) versus the anisotropy parameter J_2 . The LSUB m approximations with $m = \{2, 4, 6, 8\}$, using the ferrimagnetic canted state of Fig. 1(b) as CCM model state, are shown. The corresponding classical result $\phi_{\text{cl}} \equiv \cos^{-1}(\frac{1}{2J_2})$ is also shown for comparison.

been observed in many other magnetic systems, namely, the tendency for quantum fluctuations themselves to favor collinear over noncollinear order. Nevertheless, we leave until later in this section a discussion of extrapolating these results for the critical point $J_2^{\text{LSUB}m}$ to the $m \rightarrow \infty$ limit, and hence also of a comparison of the quantum $s = \frac{1}{2}$ case with its classical ($s \rightarrow \infty$) counterpart.

In Fig. 3, we now show the canting angle $\phi_{\text{LSUB}m}$ that minimizes the GS energy $E_{\text{LSUB}m}(\phi)$ using the ferrimagnetic canted state of Fig. 1(b) as CCM model state, at various CCM LSUB m levels, with $m = \{2, 4, 6, 8\}$. We see from Fig. 3 that, at each LSUB m level shown, the canting angle $\phi_{\text{LSUB}m}$, which minimizes the corresponding estimate $E_{\text{LSUB}m}(\phi)$ for the GS energy of the model, approaches zero smoothly, but with infinite slope, from the canted state (with $\phi_{\text{LSUB}m} \neq 0$) side of the phase transition, as the anisotropy parameter J_2 is reduced, to the corresponding estimate for the critical point $J_2^{\text{LSUB}m}$, and that it then remains zero for all $J_2 < J_2^{\text{LSUB}m}$ on the Néel' state side of the transition. This behavior is completely analogous to that seen in the classical version of the model, also shown in Fig. 3. The evidence so far, therefore, is that both the classical and spin- $\frac{1}{2}$ versions of the HAF on the anisotropic kagome lattice show second-order phase transitions between ferrimagnetic states with collinear Néel' order and noncollinear canted order. We will see in the following, however, that this picture changes when the magnetic order parameter is considered too.

We note first though that, by contrast, the corresponding behavior observed in the isotropic version (i.e., when $J'_1 = J_1 \equiv 1$) of the interpolating kagome-square HAF model (or J_1 - J'_1 - J_2 model) of Fig. 1(a), (and for which the present HAF model on the anisotropic kagome lattice is just the special case with $J'_1 = 0$), is quite different. Thus, in the spin- $\frac{1}{2}$ J_1 - J'_1 - J_2 model with $J'_1 = J_1 \equiv 1$, it was observed³⁶ that at each LSUB m level there is a finite jump in $\phi_{\text{LSUB}m}$ at the corresponding LSUB m approximation for the phase transition

TABLE I. The critical value $\kappa_{c_1}^{\text{LSUB}m}$ at which the transition between the Néel' phase ($\phi = 0$) and the canted phase ($\phi \neq 0$) occurs in the LSUB m approximation using the CCM with the (Néel) or canted state shown in Fig. 1(a) (and with $J'_1 \rightarrow 0$) as model state for the spin- $\frac{1}{2}$ J_1 - J_2 HAF on the anisotropic kagome lattice of Eq. (1).

Method	$\kappa_{c_1}^{\text{LSUB}m}$
LSUB2	0.685
LSUB4	0.600
LSUB6	0.568
LSUB8	0.559

at $J_2 = J_2^{\text{LSUB}m}$ between the Néel state (with $\phi_{\text{LSUB}m} = 0$) and the canted state (with $\phi_{\text{LSUB}m} \neq 0$). This may be compared with the smooth behavior of the classical canting angle $\phi_{\text{cl}} \equiv \cos^{-1}(\frac{1}{J_2})$ for this case for $J_2 > J_2^{\text{cl}} = 1$ for that model. For this latter J_1 - J'_1 - J_2 model in the isotropic case with $J'_1 = J_1$ the evidence was that the phase transition between states with Néel and canted order was first order for the spin- $\frac{1}{2}$ case compared with its second-order classical counterpart. On the other hand, one should note that for that case³⁶ we could not completely rule out the possibility that as $m \rightarrow \infty$, with increasing level of LSUB m approximation, the phase transition at $\kappa = \kappa_{c_1} \equiv \kappa_{c_1}^{\text{LSUB}\infty}$ becomes of second-order type, although a weakly first-order one seemed more likely on the basis of the available numerical evidence.

Returning now to the question of estimating the phase transition point at $\kappa = \kappa_{c_1}$ in the present model, we note that previous empirical experience^{36,101} shows that the LSUB m estimates at $\kappa = \kappa_{c_1}^{\text{LSUB}m}$ fit well to an extrapolation scheme $\kappa_{c_1}^{\text{LSUB}m} = \kappa_{c_1}^{\text{LSUB}\infty} + cm^{-1}$. For the present anisotropic spin- $\frac{1}{2}$ J_1 - J_2 model on the kagome lattice of Eq. (1), our phase transition estimates for $\kappa_{c_1} \equiv \kappa_{c_1}^{\text{LSUB}\infty}$ are shown in Table I. Using the above extrapolation scheme and the whole data set $m = \{2, 4, 6, 8\}$ gives the estimate $\kappa_{c_1} = 0.514 \pm 0.003$, while the corresponding estimate from using the data set $m = \{4, 6, 8\}$ is $\kappa_{c_1} = 0.515 \pm 0.007$. In both cases, the quoted error estimates are simply the standard deviations from the associated least-squares fits. Our best estimate from combining all of our results is $\kappa_{c_1} = 0.515 \pm 0.015$, which may be compared with the corresponding classical value of $\kappa_{\text{cl}} = 0.5$. Clearly, it is not excluded that $\kappa_{c_1} = \kappa_{\text{cl}}$, such that the transition point above which collinear Néel' order disappears occurs at exactly the same value $\kappa = 0.5$ of the anisotropy parameter for both the extreme quantum and classical limiting cases of the spin quantum number s . We also discuss the nature of the phase transition at κ_{c_1} for the spin- $\frac{1}{2}$ model in more detail in the following.

We note from Fig. 3 that as $J_2 \rightarrow \infty$, the canting angle $\phi \rightarrow \frac{1}{2}\pi$ faster than does the classical analog ϕ_{cl} . We also note that for the special case $J_2 = 1$ (or, equivalently, $\kappa = 1$) of the isotropic kagome lattice, the CCM LSUB m estimates for the canting angle ϕ take the value $\phi_{\text{LSUB}m} = \frac{\pi}{3}$ for all values of m , as expected by symmetry, and exactly as in the classical version of the model.

In Fig. 4, we show our CCM results for the GS energy per spin E/N_K as a function of J_2 for the present anisotropic

FIG. 4. (Color online) Ground-state energy per spin E/N_K versus J_2 for the spin- $\frac{1}{2}$ J_1 - J_2 HAF on the anisotropic kagome lattice of Eq. (1) (with $J_1 \equiv 1$), using the generic ferrimagnetic canted model state shown in Fig. 1(b) as CCM model state, and with the canting angle $\phi = \phi_{\text{LSUB}m}$ chosen to minimize the corresponding LSUB m estimate for the energy $E_{\text{LSUB}m}(\phi)$, at each value of J_2 . The CCM LSUB m results with $m = \{2, 4, 6, 8\}$ are shown, together with the corresponding extrapolated LSUB ∞ result from Eq. (9).

spin- $\frac{1}{2}$ J_1 - J_2 model on the kagome lattice of Eq. (1). The CCM LSUB m results with $m = \{2, 4, 6, 8\}$ and the corresponding extrapolated LSUB ∞ results obtained from Eq. (9) are shown. As explained previously, in each LSUB m approximation, we choose the canting angle $\phi = \phi_{\text{LSUB}m}$ for each separate value of the parameter J_2 that minimizes the corresponding CCM estimate for the energy $E_{\text{LSUB}m}(\phi)$.

At the isotropic kagome point (i.e., when $J_2 = J_1 = 1$), our present best estimate for the GS energy per spin, based on the extrapolation with the data set $m = \{4, 6, 8\}$, is $E/N_K \approx -0.4352$. This is lower than two recent rigorous upper bounds.^{37,38} Thus, Evenbly and Vidal³⁷ used the multiscale entanglement renormalization ansatz to evaluate exactly (up to floating point round-off errors) the energy of a wave function of the so-called honeycomb valence-bond crystalline type (with a 36-site unit cell), to give the rigorous bound $E/N_K < -0.4322$. Similarly, using a simple cluster product state for the infinite kagome lattice based on a fundamental cluster of 576 sites, for which the interior of the cluster has the uniform valence-bond patterning expected of a spin-liquid state, Yan, Huse, and White³⁸ have recently given an improved rigorous upper bound of $E/N_K < -0.4332$. The same authors³⁸ also use a large-scale density-matrix renormalization group (DMRG) technique to provide what is certainly one of the most accurate estimates currently available for the energy per site of the isotropic kagome-lattice HAF, namely, $E/N_K = -0.4379 \pm 0.0003$. This estimate is itself consistent with the best available large-scale Lanczos exact diagonalization (ED) results for finite clusters of up to $N = 42$ sites.⁴⁰ It is also in excellent agreement with that from another very recent large-scale DMRG study,⁵² namely, $E/N_K = -0.4386 \pm 0.0005$. Our own present best estimate, cited above, is clearly below both of the rigorous upper bounds and in good agreement with the DMRG results.

FIG. 5. (Color online) Ground-state magnetic order parameter M_K versus J_2 for the spin- $\frac{1}{2}$ J_1 - J_2 HAF on the anisotropic kagome lattice of Eq. (1) (with $J_1 \equiv 1$), using the generic ferrimagnetic canted model state shown in Fig. 1(b) as CCM model state, and with the canting angle $\phi = \phi_{\text{LSUB}m}$ chosen to minimize the corresponding LSUB m estimate for the energy $E_{\text{LSUB}m}(\phi)$ at each value of J_2 . The CCM LSUB m results with $m = \{2, 4, 6, 8\}$ are shown, together with the corresponding extrapolated LSUB ∞ results using both data sets $m = \{2, 4, 6, 8\}$ and $\{2, 4, 6\}$ for comparison purposes. Extrapolated results are calculated using Eq. (11) for the Néel' state and Eq. (10) for the canted state, respectively; and the (gray) dashed vertical line at $J_2 = 0.515$ represents our best estimate for the termination point above which collinear Néel' order disappears, at $J_2 = J_2^c \equiv J_2^{\text{LSUB}\infty}$, as discussed in the text.

Our extrapolated result for the energy is also in very good agreement with previous CCM estimates⁴⁸ for the spin- $\frac{1}{2}$ HAF on the isotropic kagome lattice that used the kagome geometry itself to define the fundamental clusters of the LSUB m configurations rather than the triangular lattice geometry of Fig. 1(a) that we use here, as discussed above in Sec. III, in which the B_1 sites are retained. Thus, using the kagome-lattice geometry the extrapolated result $E/N_K \approx -0.4357$ was found with the LSUB m set $m = \{4, 5, 6, 7, 8, 9, 10\}$ and $E/N_K \approx -0.4372$ with the LSUB m set $m = \{6, 7, 8, 9, 10\}$.⁴⁸

Figure 4 shows weak signals of the phase transition point (at $\kappa = \kappa_{c_1} \equiv \kappa_{c_1}^{\text{LSUB}\infty}$) in each of the LSUB m curves, where a discontinuity in the first derivative of the energy is observed at the corresponding value $\kappa = \kappa_{c_1}^{\text{LSUB}m}$. As usual, the transition is seen more clearly in the behavior of the average local onsite magnetization $M_K \equiv -\frac{1}{N_K} \sum_{i=1}^{N_K} \langle s_i^z \rangle$, where the sum is taken over all N_K sites of the kagome lattice and where again the spins are defined in the local, rotated spin axes in which all spins in the CCM model state point in the negative z direction.

Thus, in Fig. 5 we show the magnetic order parameter M_K , as a function of J_2 , for the present anisotropic spin- $\frac{1}{2}$ J_1 - J_2 model on the kagome lattice of Eq. (1). CCM results are shown for LSUB m approximations with $m = \{2, 4, 6, 8\}$, together with various LSUB ∞ extrapolations. As discussed in Sec. III, for the strongly frustrated regime in which the canted state is the stable ground state (i.e., for $J_2 > J_2^c$), we use the well-tested and established scheme of Eq. (10), whereas for the less frustrated regime in which the Néel' state is the stable

ground state (i.e., for $J_2 < J_2^{c1}$), we use the scheme of Eq. (11). Since for the Néel' state the LSUB m results converge (with increasing values of m) much faster than those for the canted state, as can clearly be seen from Fig. 5, it is evident that the use of these different schemes for the two regimes is justified. Since the approximate transition point at $J_2 = J_2^{\text{LSUB}m}$ (when $J_1 = 1$) between the two phases depends slightly on the CCM truncation index m , as has already been noted above, and as can be seen clearly in Fig. 5 (and, more explicitly in Table I), it is clear that the region very near the transition is inherently difficult to extrapolate accurately.

To illustrate the sensitivity of our extrapolations to the approximations used, we show in Fig. 5 the corresponding extrapolations in the two regimes using both the data sets $m = \{2, 4, 6, 8\}$ and $\{2, 4, 6\}$. Since the (most accurate) LSUB8 scheme is computationally expensive, results are shown only at the limited set of J_2 values indicated by the symbols in Fig. 5. It is very encouraging that the extrapolated curves for M_K are very steep near J_2^{c1} and that they become steeper still as higher LSUB m approximations are included. For example, the extrapolated LSUB ∞ curve for M_K obtained from the set $m = \{2, 4, 6\}$ becomes zero at $J_2 \approx 0.67$ (with $J_1 \equiv 1$), while that obtained from the set $m = \{2, 4, 6, 8\}$ becomes zero at $J_2 \approx 0.63$ (with $J_1 \equiv 1$). It seems reasonable to assume that the actual LSUB ∞ curve will become vertical at the point $J_2 = J_2^{c1} \equiv J_2^{\text{LSUB}\infty}$, as is seen in Fig. 5 by the proximity of the extrapolations to the vertical line at $J_2 = 0.515$, which represents our best estimate for the transition point between the Néel' and canted states.

Figure 5 shows the existence of a clear window in the parameter κ in which $M_K < 0$, and hence in which the canted order present in the model state has vanished. This (paramagnetic) region includes the point $\kappa = 1$ corresponding to the isotropic kagome HAF, and it seems reasonable to assume that the phase present in this regime is the same as the paramagnetic GS phase of the isotropic model. As discussed above, the lower boundary of this window seems to coincide with the point $\kappa = \kappa_{c1}$ above which the collinear Néel' order disappears.

Thus, the evidence from the magnetization data shown in Fig. 5 now indicates that the transition at $\kappa = \kappa_{c1}$ is actually between the Néel' and paramagnetic phases, rather than between the Néel' and canted phases as in the classical case at the corresponding value $\kappa = \kappa_{cl}$. Nevertheless, we can not completely exclude the possibility of a very narrow strip of canted phase between the Néel' and paramagnetic phases confined to the region $0.5 < \kappa \lesssim 0.6$.

We denote by κ_{c2} the corresponding critical value of κ that marks the upper boundary of the window in which $M_K < 0$, and that hence marks the transition between the paramagnetic and canted phases. From Fig. 5 we find estimates $\kappa_{c2} \approx 1.83 \pm 0.02$ from the extrapolated LSUB ∞ curve using the data set $m = \{2, 4, 6, 8\}$ and $\kappa_{c2} \approx 1.80 \pm 0.02$ from the corresponding curve using the data set $m = \{2, 4, 6\}$.

In Fig. 6 we also show, in various approximations, the separate average onsite magnetizations M_A and M_{B_2} on the A and the B_2 sites, respectively, of Fig. 1(b), as well as their average value on the kagome lattice $M_K = \frac{2}{3}M_A + \frac{1}{3}M_{B_2}$. We recall that the A sites are connected by two J_1 bonds and two J_2 bonds, while the B_2 sites are connected by four J_1 bonds. In particular, we note that as $J_2 \rightarrow \infty$ (with $J_1 \equiv 1$), the model

FIG. 6. (Color online) Various ground-state magnetic order parameters versus J_2 for the spin- $\frac{1}{2}$ J_1 - J_2 HAF on the anisotropic kagome lattice of Eq. (1) (with $J_1 \equiv 1$), using the generic ferrimagnetic canted model state shown in Fig. 1(b) as CCM model state, and with the canting angle $\phi = \phi_{\text{LSUB}m}$ chosen to minimize the corresponding LSUB m estimate for the energy $E_{\text{LSUB}m}(\phi)$ at each value of J_2 . Results are shown for the average local onsite magnetizations M_A and M_{B_2} on the A and the B_2 sites, respectively, of Fig. 1(b), as well as for their average value on the kagome lattice $M_K = \frac{2}{3}M_A + \frac{1}{3}M_{B_2}$. LSUB4 results are shown for M_{B_2} , and extrapolated LSUB ∞ results are shown for M_K using both data sets $m = \{2, 4, 6, 8\}$ and $\{2, 4, 6\}$, and for M_A using the data set $m = \{2, 4, 6\}$. Extrapolated results are calculated using Eq. (11) for the Néel' state and Eq. (10) for the canted state, respectively; and the (gray) dashed vertical line at $J_2 = 0.515$ represents our best estimate for the termination point above which collinear Néel' order disappears, at $J_2 = J_2^{c1} \equiv J_2^{\text{LSUB}\infty}$, as discussed in the text.

reduces to one of independent linear HAF chains of alternating A_1 and A_2 sites. In this limit, our extrapolated CCM result for the energy using the ferrimagnetic canted state as model state (and see Fig. 4) is $E/N_K \approx -0.2954J_2$. Since in this limit the B_2 sites become irrelevant, we may reexpress the result in terms of the number $N_c = \frac{2}{3}N_K$ of A sites that form the independent 1D chains in this limit, as $E/N_c \approx -0.4431J_2$. This compares extremely well with the exact result for the 1D HAF, obtained from the Bethe ansatz solution $E/(N_c J_2) = \frac{1}{4} - \ln 2 \approx -0.443147$.

As $J_2 \rightarrow \infty$ (with $J_1 \equiv 1$), all of our CCM LSUB m approximations give $M_{B_2} \rightarrow 0.5$, as expected. The LSUB4 result for M_{B_2} shown in Fig. 6 is typical of the behavior of the entire LSUB m set. By contrast, in the same limit, the individual LSUB m approximations for M_A approach different constant (positive) values. The extrapolated LSUB ∞ result for M_A , shown in Fig. 6, however, approaches a value very close to zero, again fully consistent with the exact behavior of 1D HAF chains.

Our results so far have indicated the presence of a paramagnetic state in the range $\kappa_{c1} < \kappa < \kappa_{c2}$ between the quasiclassical states with Néel' order (for $\kappa < \kappa_{c1}$) and canted order (for $\kappa > \kappa_{c2}$). We have seen too that as $\kappa \rightarrow \infty$, the model reduces consistently to the limit of uncoupled isotropic HAF 1D chains. There remains still the question raised in Sec. II as to whether the canted state illustrated in Fig. 1(b)

remains the stable GS phase all the way out to $\kappa \rightarrow \infty$, where the canting angle $\phi \rightarrow 90^\circ$, or whether there might exist a further phase transition at a value $\kappa = \kappa_{c_3} > \kappa_{c_2}$ to some other phase. One such possibility is the semistriped state illustrated in Fig. 1(c). We have argued that such a phase might be stabilized due to the quantum fluctuations possibly lifting the infinite degeneracy, which exists in the classical counterpart between the orientation of the antiferromagnetically aligned spins on the A sites and the orientation of the ferromagnetically aligned spins on the B₂ sites (that become decoupled from those on the A sites in this limit), by the order-by-disorder mechanism. We consider this possibility further in Sec. V, where we also discuss our results and compare them with those of others.

V. DISCUSSION AND CONCLUSIONS

We have used the CCM to investigate the effects of quantum fluctuations on the zero-temperature GS properties and phase diagram of the spin- $\frac{1}{2}$ J_1 - J_2 HAF on the anisotropic kagome lattice of Eq. (1), and as illustrated in Fig. 1(b). The system contains spatially anisotropic NN exchange couplings on the kagome net, with coupling $J_2 \equiv \kappa J_1 > 0$ in one of the three equivalent spatial directions of the lattice and coupling $J_1 \equiv 1$ along the other two directions. The model has only two classical GS phases. For $\kappa < \kappa_{cl} \equiv \frac{1}{2}$, the GS phase has collinear ferrimagnetic Néel' order, in which the J_2 -chain spins on the A sites of the lattice are aligned in one direction and the middle spins on the remaining B₂ sites are aligned in the opposite direction. At $\kappa = \kappa_{cl} \equiv \frac{1}{2}$, the classical GS configuration changes from the essentially unique collinear ferrimagnetic Néel' state to an infinite ensemble of degenerate states. These include a still infinite number of coplanar canted ferrimagnetic states that are expected to be selected from among the rest by thermal or quantum fluctuations to comprise the stable GS phase for all $\kappa > \kappa_{cl} \equiv \frac{1}{2}$.

For a given value of $\kappa > \kappa_{cl}$, all of these states are characterized by a canting angle ϕ such that on each A₁A₂B₂ triangular plaquette of the kagome net, the two J_2 -chain spins (i.e., those on the A₁ and A₂ sites) form angles $(\pi \pm \phi)$ with respect to the middle spin (i.e., that on the B₂ site). Clearly, the Néel' state is just the special case with $\phi = 0$. Each triangular plaquette in the ensemble of coplanar states also carries a chirality variable $\chi = \pm 1$, defined to be the direction (anticlockwise or clockwise, respectively) in which the spins rotate as one traverses the plaquette in the positive (anticlockwise) direction. The different degenerate coplanar canted states then correspond to different ways of assigning chiralities to the individual plaquettes.

In this paper, we have used for the spin- $\frac{1}{2}$ model the generic ferrimagnetic canted model state shown in Fig. 1(b) as our CCM model state, which is that member of the classically degenerate ensemble in which all of the triangular plaquettes have the same (here positive) chirality. This state thus corresponds to the $q = 0$ state of the isotropic (i.e., when $J_2 = 1$) kagome-lattice HAF, in which $\phi = \frac{\pi}{3}$ by symmetry. At each LSUB m level of approximation, we have chosen the value of the canting angle ϕ that minimizes the corresponding LSUB m estimate for the GS energy. We note again parenthetically that in a previous recent CCM analysis⁴⁸ of the isotropic kagome

HAF in which both the $q = 0$ state and the $\sqrt{3} \times \sqrt{3}$ state (that corresponds to that member of the classically degenerate ensemble in which the chiralities alternate, such that triangular plaquettes joined by a vertex have opposite values of χ) were employed as CCM model states, it was found that for the extreme quantum case considered here, with $s = \frac{1}{2}$, the $q = 0$ state is energetically favored over the $\sqrt{3} \times \sqrt{3}$ state, while for any $s > \frac{1}{2}$ the $\sqrt{3} \times \sqrt{3}$ state is selected over the $q = 0$ state.

We note too that previous CCM studies of many other strongly correlated and highly frustrated models in quantum magnetism have shown that the calculated positions of phase boundaries are rather insensitive to the choice of CCM model state where several competing possibilities exist that lie close in energy to one another. In the present case, we have repeated the calculations performed here, for the $q = 0$ state as CCM model state, with the $\sqrt{3} \times \sqrt{3}$ state so chosen, and have found that the effect on the resulting value of κ_{c_2} is basically within our stated error bars.

In a very interesting recent paper, Masuda *et al.*⁵¹ show that a first-order phase transition, which has no counterpart in the isotropic case ($\kappa = 1$), occurs in the *classical* ($s \rightarrow \infty$) anisotropic kagome model (with $\kappa > 1$) at a very low but finite temperature. They conclude that thermal fluctuations tend to favor, by the order-by-disorder mechanism,¹¹⁰ an incommensurate spiral phase from among the massively degenerate ensemble of classical ground states for values of $\kappa > 1$. This spiral state reduces to the $\sqrt{3} \times \sqrt{3}$ state in the isotropic limit $\kappa \rightarrow 1$. Such a state, however, must be extremely fragile to small perturbations in the Hamiltonian. For example, even an addition of the J'_1 bonds shown in Fig. 1(a) with an infinitesimal (positive) strength acts to stabilize, for all values of $\kappa > \frac{1}{2}$, the classical canted state shown in the figure, which is just the $q = 0$ state used as our CCM model state. The use of this spiral state as CCM model state would again be very unlikely to alter our results for κ_{c_2} for reasons cited above.

We found here that the canting angle ϕ_{LSUBm} that minimizes the GS energy $E_{LSUBm}(\phi)$ for the spin- $\frac{1}{2}$ model, at a given LSUB m level, becomes nonzero for values of the anisotropy parameter $\kappa > \kappa_{c_1}^{LSUBm}$, where $\kappa_{c_1}^{LSUBm}$ is generally somewhat higher than the corresponding classical value $\kappa_{cl} \equiv \frac{1}{2}$, as shown in Table I. On the other hand, these critical values converge to an extrapolated value $\kappa_{c_1} \equiv \kappa_{c_1}^{LSUB\infty}$ that is close to $\kappa_{cl} \equiv \frac{1}{2}$. Indeed, our best estimate is $\kappa_{c_1} = 0.515 \pm 0.015$. Although they can not entirely exclude the possibility of a small regime of canted phase in a very narrow strip immediately above κ_{c_1} , our corresponding CCM LSUB m results for the magnetization shown in Figs. 5 and 6 give compelling evidence, however, that the transition for the spin- $\frac{1}{2}$ model at $\kappa = \kappa_{c_1}$ from a GS phase with Néel' order is *not* to one with canted order, as in the corresponding classical model at $\kappa = \kappa_{cl} = 0.5$, but rather to a paramagnetic state with no canted order. Our evidence is that this paramagnetic GS phase persists over the anisotropy range $\kappa_{c_1} < \kappa < \kappa_{c_2}$, before the canted state becomes the stable GS phase for $\kappa > \kappa_{c_2}$. Our best estimate for this upper critical point of the paramagnetic phase is $\kappa_{c_2} = 1.82 \pm 0.03$.

Since the isotropic kagome-lattice point $\kappa = 1$ is contained within the parameter range $\kappa_{c_1} < \kappa < \kappa_{c_2}$ of this paramagnetic phase, the natural conclusion is that this phase of the

anisotropic model shares the same order as the GS phase of the isotropic model. As we discussed in Sec. I, the isotropic spin- $\frac{1}{2}$ HAF on the kagome lattice has been greatly studied in the past. The most direct results, namely, those from the exact diagonalization (ED) of finite lattices,^{14,16,34,40,42} seem to provide strong evidence for a spin-liquid GS phase. This conclusion is supported by the results of block-spin approaches^{15,18} and by those from various other studies too.^{5,10,17,26,28,31,32}

Very recent ED studies^{40,42} have examined the GS energies and spin gaps (specifically between the GS singlet level and the lowest-lying triplet level) of many isotropic kagome clusters of sizes up to $N = 42$. In their study, for example, Nakano and Sakai⁴² further claim that, from the result of their analysis of larger clusters, the isotropic HAF on the kagome lattice is gapless, in contradiction with other recent DMRG studies^{38,49,52} that find it to be gapped. Whereas earlier ED studies also attempted to resolve the spin-gap issue, the data on clusters of sizes up to $N = 36$ was deemed³⁴ to be insufficient to distinguish between a gapless system and one with a very small gap.

On the other hand, conflicting results have been found by other authors^{3,21,22,24,27,30,37} who have proposed various valence-bond solid states as the GS phase of the isotropic HAF on the kagome lattice. A detailed comparison of the exact spectrum of a 36-site finite lattice sample of the isotropic kagome HAF against the excitation spectra allowed by the symmetries of the various proposed valence-bond crystal states has, however, cast very strong doubts on their validity as stable GS phases.²⁹ Over the past year or so, this muddled and confused picture of the nature of the GS phase of the isotropic spin- $\frac{1}{2}$ HAF on the kagome lattice has been seemingly resolved in favor of a topological spin liquid.

In particular, as indicated in Sec. I, the results of two independent and very recent large-scale DMRG studies of the spin- $\frac{1}{2}$ isotropic HAF on the kagome lattice^{49,52} have provided compelling *positive* evidence that this GS phase is a topological quantum spin liquid. The simplest such state that preserves all symmetries is the $\mathbb{Z}_2$ spin liquid, and by explicitly calculating the topological entanglement entropy, both recent DMRG studies provide strong positive evidence that the spin-liquid state does indeed have $\mathbb{Z}_2$ topological order, with a finite spin (triplet) gap.⁵²

It has not been our aim here to investigate the order properties of the paramagnetic GS phase of antiferromagnetically coupled $s = \frac{1}{2}$ spins on the infinite kagome lattice, but rather to investigate the stability of the phase as the lattice is spatially distorted. Clearly, however, it is natural to expect that over the entire range $\kappa_{c_1} < \kappa < \kappa_{c_2}$ in which the paramagnetic phase persists (as manifested here by a negative, and hence unphysical, value of the calculated local magnetic order parameter), it remains a spin liquid with the same topological order.

We are reticent to make specific claims of the direct relevance of our results to such real materials as volborthite. Naturally, we would like to be able to claim that the paramagnetic region $\kappa_{c_1} < \kappa < \kappa_{c_2}$ in which we have found that the classical ground states are unstable has applicability to the spin-glass phase observed experimentally in volborthite. Indeed, if volborthite could certainly be described by the present anisotropic kagome model, then its value for κ would

fall within the paramagnetic region we have found, and such a claim might be justified. Unfortunately, however, as we discussed in Sec. I, the nature of the magnetic couplings in volborthite has recently been questioned.⁷³ Thus, it was pointed out that the local environments of the two inequivalent types of Cu sites (that were previously used to justify the use of the present anisotropic model) differ in essential ways. A DFT study⁷³ was then used to show that a better model of this material might be more akin to one involving coupled frustrated chains in which some of the NN bonds are actually *ferromagnetic* in nature.

The spin- $\frac{1}{2}$ HAF on the spatially anisotropic kagome lattice has also been studied by several other authors recently using a variety of techniques. These have included large- N expansions of the $\text{Sp}(N)$ -symmetric generalization of the actual $\text{SU}(2)$ model,^{79,80} a block-spin perturbation approach to the trimerized kagome lattice,⁸⁰ various semiclassical calculations (appropriate to the limit of large spin quantum number s) that include (a) studying an effective chirality Hamiltonian derived from a low-temperature classical nonlinear spin-wave expansion,⁸¹ and (b) keeping terms of order $1/n$ in the large- n limit of the $\text{O}(n)$ generalization of the classical $\text{O}(3)$ model together with a high-temperature expansion,⁸¹ field-theoretical techniques appropriate to quantum critical systems in one dimension (and which are hence appropriate here for the case $\kappa \equiv J_2/J_1 \gg 1$ of weakly coupled chains),⁸² and a renormalization-group analysis in the same quasi-1D limit but now also in the presence of a Dzyaloshinskii-Moriya interaction.⁸⁴

Since many of these calculations employ perturbation theories of one kind or another in some “artificial” small parameter, direct comparison is difficult. Thus, for example, in the large- N $\text{Sp}(N)$ expansion, the effective smallness parameter is $\alpha \equiv n_b/N$, where n_b is the number of bosons on each site. While in the physical $\text{SU}(2)$ model (which corresponds to the case $N = 1$) we have $\alpha = 2s$, the comparison in the large- N limit actually studied is lost. Yavors’kii *et al.*⁸⁰ argue that the value of α that corresponds to the $s = \frac{1}{2}$ under study must be somewhat less than 0.5. Similarly, in the block-spin perturbation approach of Yavors’kii *et al.*⁸⁰ the assumption is made that the kagome lattice is trimerized such that the spins on the downward-pointing triangles, say, are strongly coupled, whereas the couplings on the bonds of the upward-pointing triangles are weaker by a factor γ . The approximate GS phases of this trimerized model are then studied in different regimes of the anisotropy parameter κ in a perturbation expansion with respect to γ , while the physical model corresponds to the case $\gamma = 1$.

Complementary to such essentially perturbative studies have been various more controlled analyses of the quasi-1D limit of the model.^{82,84} The latter studies, by their nature of focusing on the large-anisotropy ($\kappa \gg 1$) limit, also lose sight of the intermediate paramagnetic (spin-liquid) phase that has been the focus of this study. For example, Zyuzin *et al.*⁸⁴ explicitly state that they do not find a spin-liquid ground state in any regime that they study.

As we have seen, much of the previous work on the spin- $\frac{1}{2}$ HAF on the spatially anisotropic kagome lattice has approached the quantum limit only very indirectly, either from the classical side or in such slave particle approaches as the

Schwinger boson technique applied in the large- N $\text{Sp}(N)$ approach. The only direct $s = \frac{1}{2}$ approach seems to be a small-scale ED study of up to $N_K = 24$ spins (in a 4×2 unit-cell arrangement) with periodic boundary conditions.⁸¹ The numerical evidence from the ED study seems to indicate very clearly that for values of the anisotropy parameter $\kappa < 0.5$, the GS phase has nonzero total spin. Indeed, its value for the few finite-size lattices studied is precisely what is expected for the classical collinear Néel' ferrimagnetic state, namely, $S_{\text{tot}} = \frac{1}{3}N_K s$, thereby agreeing fully with our own findings. By contrast, the numerical evidence for values $\kappa > 0.5$ is far less clear. While the evidence seems to be that for all lattice sizes up to 24 sites the GS phase is a spin singlet for all values $\kappa > 0.5$, there is a clear tendency for a state of nonzero spin at the Γ point to drop in energy on moving away from the isotropic point $\kappa = 1$, perhaps indicating a tendency to develop a net moment again. Nevertheless, the evidence from such small-scale ED studies seems to leave completely open the nature of the GS phase for $\kappa > 0.5$.

The two semiclassical approaches of Wang *et al.*⁸¹ also concur that for $\kappa < 0.5$ the GS phase is the collinear Néel' ferrimagnetic state. For the case $\kappa > 1$, both approaches also indicate a canted ferrimagnetic state of the classical type, but where the infinitely degenerate manifold of coplanar states is lifted by the order-by-disorder mechanism, which now seems to favor the so-called *chirality stripe* state as the GS phase, in which all spins on the interstitial B_2 sites are ferromagnetically aligned, and where pairs of triangles on the kagome lattice that share either an A_1 or A_2 vertex have the same value of the chirality parameter χ , while pairs sharing a B_2 vertex have opposite values of χ . [We note parenthetically that this state is the only other state, apart from the $q = 0$ state of Fig. 1(b), that is allowed by the chirality constraints in the general case $\kappa \neq 1$ and in which all of the B_2 spins are aligned parallel to one another.] By contrast, in the intermediate regime $0.5 < \kappa < 1$, the semiclassical models do not give clear indications of which GS ordering is favored. Thus, while the $\sqrt{3} \times \sqrt{3}$ order seems to be favored in the spin-wave expansion for the isotropic case $\kappa = 1$, the comparable analysis for the $\kappa < 1$ case seems to depend very sensitively on both the choice of unphysical parameters and on the number of chirality-chirality couplings included in the analysis. By contrast, the large- n saddle-point solution of the $O(n)$ generalization of the classical $O(3)$ model seems to favor the $q = 0$ GS ordering of spins for the case $0.5 < \kappa < 1$.

The large- N $\text{Sp}(N)$ expansion analysis of the model^{79,80} indicates that the actual spin- $\frac{1}{2}$ HAF on the spatially anisotropic kagome lattice has a GS phase with collinear Néel' ferrimagnetic order for small values of the anisotropy parameter κ , which gives way at larger values of κ to an incommensurate (spiral) GS phase with no LRO, in general agreement with our findings. In turn, as κ is increased further, this GS phase then gives way to a phase in which the chains are completely decoupled, while the interstitial spins (i.e., those on B_2 sites) show some short-range spin-spin correlations. For reasons already noted above, this $\text{Sp}(N)$ analysis is unable to give quantitative estimates for the corresponding two critical values of κ for the actual spin- $\frac{1}{2}$ $\text{SU}(2)$ model.

Finally, we note that the canted ferrimagnetic phase that we have found to be the stable GS phase for $\kappa > \kappa_{c_2}$, after

the disappearance of the paramagnetic (spin-liquid) phase is not likely to remain the GS phase for sufficiently large values of κ , as we have already noted in Sec. II, since in the limit $\kappa \rightarrow \infty$ the canting angle $\phi \rightarrow \frac{\pi}{2}$, and the chain spins become perpendicular to the interstitial spins. Since in this limit the relative orientation of the chain spins and the interstitial spins becomes irrelevant, and since quantum fluctuations generally prefer collinear spin configurations in such a degenerate situation, we thus expect a third phase transition at a value $\kappa = \kappa_{c_3}$ to a phase that eventually becomes the decoupled 1D HAF chain phase in the asymptotic limit $\kappa \rightarrow \infty$. The precise nature of this fourth phase is by no means settled.

We have alluded in Sec. II to one such candidate being the collinear ferrimagnetic semistriped phase shown in Fig. 1(c). We are currently investigating whether this phase might become energetically favored at sufficiently large values of κ . We intend to report on this in a separate future paper.

An alternative candidate state for the large- κ phase has been suggested by Yavors'kii *et al.*⁸⁰ from their block-spin trimerized version of the model. In the subsequent small- γ perturbative limit they find a tentative ground state in the large-anisotropy case ($\kappa \gg 1$) that is a collinearly ordered antiferromagnet in which the interstitial (B_2) spins are Néel ordered and the spins on the A chains form singlet dimers (i.e., the spins on each A_1 - A_2 NN pair of sites on, say, each downward-pointing triangle on the kagome lattice form a spin-singlet state).

Yet another analysis of the large- κ limit, by Schnyder *et al.*,⁸² suggests that all of the spins order with a (generally noncoplanar) configuration in which the interstitial spins on B_2 sites and the chain spins on A sites each separately form predominantly coplanar spirals with a wave vector $(q, 0)$, but with a reduced $[O(1/\kappa)]$ static moment on the J_2 -coupled chains. These authors find that the chain spins are weakly canted out of the plane, with the $[O(1/\kappa^2)]$ normal components being ordered in an antiferromagnetic fashion. While their analysis could not determine q reliably, it is expected that $q \ll 1$ and, indeed, the authors suggest that $q = 0$ is a real possibility, in which case the state becomes coplanar. Nevertheless, even this state differs from both the chirality stripe state considered by Wang *et al.*⁸¹ and the dimerized state considered by Yavors'kii *et al.*,⁸⁰ although there are some similarities with each.

It is clear that the large- κ limit of the spin- $\frac{1}{2}$ HAF on the anisotropic kagome lattice is far from settled. Our own work presented here has mainly been concerned to investigate the stability with respect to anisotropy κ of the spin-liquid state that has convincingly been found in recent work to be the stable GS phase of the isotropic ($\kappa = 1$) model. Nevertheless, we hope to return in the future to the quite separate question of whether or not there is a further transition at some value $\kappa = \kappa_{c_3} > \kappa_{c_2}$ of the anisotropy parameter, from the canted state discussed here to some other state with or without collinear order.

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