The ground-state phase diagram of the XXZ spin-$s$ kagome antiferromagnet: A coupled-cluster study

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We use the coupled cluster method to high orders of approximation in order to calculate the ground-state phase diagram of the XXZ spin-$s$ kagome antiferromagnet with easy-plane anisotropy, i.e., the anisotropy parameter $\Delta$ varies between $\Delta = 1$ (isotropic Heisenberg model) and $\Delta = 0$ (XY model). We find that for the extreme quantum case $s = 1/2$ the ground state is magnetically disordered in the entire regime $0 \leq \Delta \leq 1$. For $s = 1$ the ground state is disordered for $0.818 \leq \Delta < 1$, it exhibits $\sqrt{3} \times \sqrt{3}$ magnetic long-range order for $0.281 \leq \Delta < 0.818$, and $q = 0$ magnetic long-range order for $0 \leq \Delta < 0.281$. We confirm the recent result of Chernyshev and Zhitomirsky (Phys. Rev. Lett. 113, 237202 (2014)) that the selection of the ground state by quantum fluctuations is different for small $\Delta$ ($XY$ limit) and for $\Delta$ close to one (Heisenberg limit), i.e., $q = 0$ magnetic order is favored over $\sqrt{3} \times \sqrt{3}$ for $0 \leq \Delta < \Delta_c$ and vice versa for $\Delta_c < \Delta \leq 1$. We calculate $\Delta_c$ as a function of the spin quantum number $s$.

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Introduction. The investigation of the ground state (GS) of the quantum antiferromagnet on the kagome lattice is one of the most challenging problems in the field of frustrated quantum magnetism. Over many years numerous theoretical methods has been applied to understand the GS properties of the kagome antiferromagnet (KAFM), see, e.g., Refs. 1, 31 and references therein.

While it became clear very early that GS magnetic long-range order (LRO) is absent for the $s = 1/2$ Heisenberg KAFM, there was a longstanding debate on the nature of the non-magnetic quantum GS. Recent large-scale numerics provide strong arguments for a gapped $Z_2$ topological spin-liquid GS for spin quantum number $s = 1/2$.

Other recent investigations have been focused on higher spin $s > 1/2$ and also on the anisotropic XXZ-model. Both modifications have relevance for the experimental research, see, e.g., Refs. 32, 40. Moreover, anisotropic spin models are of great interest with respect to engineering models of quantum magnetism on optical lattices, see, e.g., Refs. 14, 15. Since higher spin quantum numbers as well as spin anisotropy, in general, lead to a reduction of quantum fluctuations, see, e.g., Refs. 43, 50, we use the coupled cluster method (CCM) for the KAFM might be facilitated. However, for the isotropic $s = 1$ Heisenberg KAFM there is clear evidence that there is no magnetic LRO. (Note, however, that in Ref. 44 $\sqrt{3} \times \sqrt{3}$ GS LRO for integer spin quantum numbers including $s = 1$ was reported.) For $s > 1$ several approaches lead to indications for $\sqrt{3} \times \sqrt{3}$ GS LRO. On the other hand, recent density matrix group (DMRG) calculations have demonstrated that for $s = 1/2$ the XXZ KAFM remains in a magnetically disordered GS for the entire range of the anisotropy parameter $\Delta$ between the $XY$ point ($\Delta = 0$) and the isotropic Heisenberg point ($\Delta = 1$).

Motivated by the recent Letter of Chernyshev and Zhitomirsky we use the coupled cluster method (CCM) in high orders of approximation to calculate the $s - \Delta$ GS phase diagram of the spin-$s$ XXZ KAFM with easy-plane anisotropy. The corresponding Hamiltonian is

$$H = \sum_{|i,j|} \left( s_i^x s_j^x + s_i^y s_j^y + \Delta s_i^z s_j^z \right), \quad 0 \leq \Delta \leq 1, \quad (1)$$

where the sum runs over all nearest-neighbor pairs. The CCM is a very general ab initio many-body technique that has been successfully applied to strongly frustrated quantum magnets, see, e.g., Refs. 12, 20, 46, 48–60. In particular, in Ref. 18 it has been demonstrated that the CCM GS energy for the $s = 1/2$ isotropic Heisenberg KAFM is close to best available DMRG results.

The coupled cluster method (CCM). For the sake of brevity we illustrate here only some relevant features of the CCM. At that we follow strictly the lines given in Ref. 18, where the CCM was applied to the isotropic spin-$s$ Heisenberg KAFM. For more general information on the methodology of the CCM, see, e.g., Refs. 51, 67. We first mention that the CCM approach yields results directly in the thermodynamic limit $N \to \infty$, where $N$ is the number of lattice sites.

First we choose a normalized reference state $|\Phi\rangle$ that is typically a classical GS of the model. From a quasi-classical point of view the coplanar $\sqrt{3} \times \sqrt{3}$ states are favored candidates among the massively degenerate manifold of classical ground states (see, e.g., Refs. 2, 4, 6). Consequently, we use both states as reference states. Then we perform a rotation of the local axes of each of the spins such that all spins in the reference state align along the negative $z$ axis in this new set of local spin coordinates we define a complete set of mutually commuting multispin creation operators $C_I^+ \equiv (C_I^-)^\dagger$ related to this reference state:

$$|\Phi\rangle = \left| \underline{\downarrow}\downarrow\downarrow \cdots \right>; \quad C_I^+ = s_i^x s_j^x + s_i^y s_j^y + s_i^z s_j^z, \quad (2)$$

where $s_n^z \equiv s_n^x + is_n^y$. In Eq. (2) the components of the spin operators are defined in the local rotated coordinate frames, and the indices $n, m, k, \ldots$ denote arbitrary
lattice sites, where each site index in each configuration index \( I \) in Eq. 2 can be repeated up to a maximum of 2\( s \) times. With the set \( \{ |\Phi\rangle, C_I^\dagger \} \) thus defined, the CCM parametrizations of the ket and bra GS eigenvectors |\( \Psi \rangle \) and \( \langle \bar{\Psi} | \) of the spin system are given by

\[
|\Psi\rangle = e^{S}|\Phi\rangle, \quad S = \sum_{I \neq 0} a_I C_I^\dagger \tag{3}
\]
\[
\langle \bar{\Psi} | = \langle \Phi | \bar{S} e^{-S} \rangle, \quad \bar{S} = 1 + \sum_{I \neq 0} \bar{a}_I C_I^{-}. \tag{4}
\]

The correlation coefficients, \( a_I \) and \( \bar{a}_I \), contained in the CCM correlation operators, \( S \) and \( \bar{S} \), are determined by the CCM ket-state and bra-state equations

\[
\langle \Phi | C_I^{-} e^{-S} H e^{S} |\Phi\rangle = 0 \; ; \forall I \neq 0 \tag{5}
\]
\[
\langle \Phi | \bar{S} e^{-S} [H, C_I^{-}] e^{S} |\Phi\rangle = 0 \; ; \forall I \neq 0. \tag{6}
\]

Equations (5) and (6) are fully equivalent to the GS Schrödinger equations for the ket and bra states. Each ket-state or bra-state equation belongs to a certain configuration index \( I \), i.e., it corresponds to a certain set (configuration) of lattice sites \( n, m, k, \ldots \), as in Eq. 2.

Using the Schrödinger equation, \( H |\Psi\rangle = E_0 |\Psi\rangle \), we can now write the GS energy as \( E_0 = \langle \Phi | e^{-S} H e^{S} |\Phi\rangle \). The magnetic order parameter (sublattice magnetization) is given by \( M = -\frac{1}{N} \sum_{i=1}^{N} \langle \Psi | s_i^z |\Psi\rangle \), where \( s_i^z \) is expressed in the transformed coordinate system, and \( N(\rightarrow \infty) \) is the number of lattice sites.

For the quantum many-body system under consideration we have to use an appropriate approximation scheme in order to truncate the expansions of \( S \) and \( \bar{S} \) in Eqs. 3 and 4. For that we use the well established SUB\( n \)-\( n \) approximation scheme, cf., e.g., Refs. 12, 18, 20, 46–48, 60. In the SUB\( n \)-\( n \) scheme we include no more than \( n \) spin flips spanning a range of no more than \( n \) conjugate lattice sites. Using an efficient parallelized CCM code \(^{29,30} \) we are able to solve the CCM equations up to SUB10-10 for \( s = 1/2 \) and up to SUB8-8 for \( s > 1/2 \). We have calculated the GS energy per spin \( e_0 = E_0/N \) and the magnetic order parameter \( M \) for spin quantum numbers \( s = 1/2, 3/2, \ldots, 9/2, 5 \). The maximum number of ket-state equations which we have to take into account is 416193 for \( s = 5 \). Following Ref. 18 we extrapolate the ‘raw’ SUB\( n \)-\( n \) data to the limit \( n \rightarrow \infty \) using \( n = 4, 5, \ldots, 10 \) (\( n = 4, 5, \ldots, 8 \)) for \( s = 1/2 \) (\( s > 1/2 \)). For that we use the well-tested extrapolation ansätze\(^ {29,30,33,37} \)

\[
e_0(n) = a_0 + a_1(1/n)^2 + a_2(1/n)^4 \quad \text{and} \quad M(n) = b_0 + b_1(1/n)^{1/2} + b_2(1/n)^{3/2}.
\]

Results. As already mentioned in the introduction, we want to calculate the GS s − \( \Delta \) phase diagram. Such a phase diagram has been very recently presented by Chernyshev and Zhitomirsky\(^ {24} \), using nonlinear spin-wave and real-space perturbation theories. To have the necessary information available for the comparison of Chernyshev’s and Zhitomirsky’s results with our CCM results reported below, let us first briefly report the main findings of Ref. 24. It is well known\(^ {24,48,57} \) that in the large-s limit for \( \Delta = 1 \) quantum fluctuations select the \( \sqrt{3} \times \sqrt{3} \) state. In Ref. 24 it was found that this GS selection is preserved for weak easy-plane anisotropy, i.e., for \( \Delta_c < \Delta \leq 1 \). However, the main and unexpected result of Ref. 24 is the selection of the \( q = 0 \) GS for smaller values of \( \Delta \) down to the XY point, i.e., for \( 0 \leq \Delta < \Delta_c \). This finding is contrary to the selection trend by thermal fluctuations for the classical KAFM, where for \( \Delta = 1 \) and for \( \Delta = 0 \) the \( \sqrt{3} \times \sqrt{3} \) state is asymptotically selected\(^ {22,23} \). Hence, for the XY antiferromagnet on the kagome lattice we are faced with an example that quantum and thermal fluctuations may act very differently. The term in the nonlinear spin-wave theory (SWT) responsible for the GS selection is of order \( O(1) \), and, therefore, the critical anisotropy \( \Delta_c = 0.72235 \) is found to be independent of the spin quantum number \( s \) within this approximation\(^ {24} \).
The real-space perturbation theory provides insight in the mechanism of the quantum selection of the ground state: Some relevant seventh-order processes change their sign as varying $\Delta$. The magnetic order parameter calculated in harmonic approximation shows a clear trend to magnetic LRO as lowering $\Delta$. However, this trend is substantially overestimated by SWT, since already for $\Delta = 1$ the linear SWT yields a vanishing order parameter for any value of $s$. For comparison we also show the corresponding large-$s$ results of Ref. 24 (labeled by 'NLSWT') obtained by non-linear SWT. The inset shows an enlarged scale of that region of $\Delta$, where $\delta e$ changes its sign.

We now discuss our CCM results. In Fig. 1 and Fig. 2, we show the GS energy per spin $e_0$ and the magnetic order parameter $M$ extrapolated to $n \to \infty$ for both the $\sqrt{3} \times \sqrt{3}$ and the $q = 0$ reference states as a function of $\Delta$ for spin quantum numbers up to $s = 5$. In general, the $\sqrt{3} \times \sqrt{3}$ and the $q = 0$ cases behave very similar. The GS energy for $s = 1/2$ is almost linearly growing with decreasing $\Delta$. For larger $s$ the $e_0(\Delta)$-curve noticeably deviates from linearity, particularly near $\Delta = 1$. This trend that the influence of the anisotropy $\Delta$ becomes exceedingly large near the isotropic Heisenberg limit at $\Delta = 1$ is more pronounced for the order parameter, see Fig. 2. For spin quantum numbers $s \geq 3/2$ there is a drastic downturn in the $M(\Delta)$-curve as approaching $\Delta = 1$ and there is a strong increase in the slope $(dM/d\Delta)|_{\Delta=1}$ with growing $s$. Note that a special behavior for $\Delta \to 1$ is also present within the spin wave approach, where the $1/s$ corrections diverge for $\Delta \to 1$. The cases $s = 1/2$ and $s = 1$ are different from the cases $s > 1$. For $s = 1/2$ our CCM approach leads to a disordered GS in the entire region of the anisotropy parameter $0 \leq \Delta \leq 1$ in accordance with recent DMRG calculations. For $s = 1$ we find a finite region of disorder, $\Delta^* \leq \Delta \leq 1$, for both reference states, where $\Delta^* = 0.818$ ($\Delta^* = 0.945$) for the $\sqrt{3} \times \sqrt{3}$ ($q = 0$) reference state. Note, however, that for anisotropies around $\Delta^*$ the CCM GS energy for the $\sqrt{3} \times \sqrt{3}$ reference state is lower than that for the $q = 0$ reference state, see below. We mention also that a table of values of the GS energy and the order parameter for $\Delta = 1$ and for spin quantum numbers up to $s = 3$ can be found in Ref. 18. We present corresponding values for the $XY$ limit ($\Delta = 0$) in Table I of the present paper.

Next we discuss the GS selection. For that we consider the energy difference $\delta e = e^{q=0}_0 - e^{\sqrt{3} \times \sqrt{3}}_0$, see Fig. 3. The main common feature of the curves shown in Fig. 3 is the change of the sign of $\delta e$, i.e., in accord with Ref. 24 we find a change in the GS selection from the $\sqrt{3} \times \sqrt{3}$ state to the $q = 0$ state at a critical value $\Delta_c$, as varying the anisotropy from $\Delta = 1$ to $\Delta = 0$. The magnitude of $\delta e$ is small, in particular for smaller $\Delta$. For larger $s$ it agrees very well with the SWT data of Ref. 24 up to $\Delta \sim 0.9$. The stronger deviation for $\Delta$ close to one can be attributed to the divergence of the $1/s$ corrections for $\Delta \to 1$.

As already mentioned above, the large-$s$ spin-wave approach yields a critical value $\Delta_c$ that is independent of $s$.
TABLE I. Extrapolated CCM results for GS energy per spin, $e_0|_{\Delta \rightarrow \infty}$, and the GS sublattice magnetization, $M|_{\Delta \rightarrow \infty}$, using the $\sqrt{3} \times \sqrt{3}$ and the $q = 0$ reference states for $\Delta = 0$ (XY model). Note that corresponding tables for $\Delta = 1$ (isotropic Heisenberg model) can be found in Ref. 13.

| $\sqrt{3} \times \sqrt{3}$ | $q = 0$ | $\frac{e_0}{s^2}$ | $M/s$ | $\frac{e_0}{s^2}$ | $M/s$ |
|-------------------------|----------|------------------|-------------|------------------|-------------|
| $s = 1/2$            | $-1.1986$ | $< 0$            | $-1.968$    | $< 0$            | $-1.1986$   |
| $s = 1$              | $-1.0578$ | $0.8602$         | $-1.0349$   | $0.8589$         | $-1.0578$   |
| $s = 3/2$             | $-1.0347$ | $0.9402$         | $-1.0349$   | $0.9368$         | $-1.0347$   |
| $s = 2$              | $-1.0252$ | $0.9570$         | $-1.0253$   | $0.9556$         | $-1.0252$   |
| $s = 5/2$             | $-1.1018$ | $0.9664$         | $-1.0199$   | $0.9656$         | $-1.1018$   |
| $s = 3$              | $-1.0163$ | $0.9723$         | $-1.0164$   | $0.9719$         | $-1.0163$   |
| $s \rightarrow \infty$ | $-1$      | $1$              | $-1$        | $1$              | $-1$        |

of the spin quantum number $s$. However, $\Delta_c$ certainly depends on $s$. Chernyshev and Zhitomirsky\textsuperscript{22} suggested that $\Delta_c$ may increase for smaller spins from the large-$s$ value $\Delta_c = 0.72325$ of $s \rightarrow \infty$. Our CCM approach yields directly $\Delta_c$ as a function of $s$, see the black open circles in Fig. 4. Contrary to the conjecture of Chernyshev and Zhitomirsky, see Fig. 4b in Ref. 24, we find that $\Delta_c$ increases for larger $s$. The smallest value, $\Delta_c = 0.281$, is found for $s = 1$. Applying $g(x) = a + bx + cx^2$, $x = 1/s$, to extrapolate the CCM data of the critical anisotropy to $s \rightarrow \infty$ yields $\lim_{s \rightarrow \infty} \Delta_c = 0.727$, where we have used $s = 3, 7/2, 4, 9/2, 5$ for the extrapolation. This CCM estimate of the large-$s$ limit of $\Delta_c$ is in excellent agreement with the large-$s$ spin-wave result.\textsuperscript{24}

To get the full relationship to the $s - \Delta$ phase diagram given in Fig. 4 of Ref. 24 we may consider the spin quantum number $s$ as a continuous variable. We determine that (fictional) value of $s$, for which the GS becomes magnetically disordered. For that we fit the data for the order parameter $M/s$ as function of $s$ and $\Delta$ by the fitting function $f(s, \Delta) = b_0(\Delta) - b_1(\Delta)s^{-1/2} - b_2(\Delta)s^{-1} - b_3(\Delta)s^{-3/2} - b_4(\Delta)s^{-2}$, where $s = 1/2$ is excluded from the fit. From $f(s_c, \Delta) = 0$ we obtain the corresponding phase boundary $s_c(\Delta)$ between magnetically disordered and ordered GS phases. This phase boundary is shown by the red solid line in Fig. 4. The resulting value of $s_c$ for $\Delta = 1$ ($\Delta = 0$) is $s_c \sim 1.34$ ($s_c \sim 0.537$).

As already mentioned above for $s = 1/2$ the GS is always magnetically disordered. For $s = 1$ there are three GS phases: the disordered state for $0.818 < \Delta \leq 1$, the ordered $\sqrt{3} \times \sqrt{3}$ state for $0.281 < \Delta < 0.818$, and the ordered $q = 0$ state for $0 \leq \Delta < 0.281$. For spin quantum numbers $s > 1$ there are two magnetically ordered GS phases, where the phase boundary is given by the black solid line in Fig. 4.

Summary. We summarize our findings by comparing our GS $s - \Delta$ phase diagram (Fig. 4) with that of Ref. 24. Most importantly, we get the same trend as Ref. 24, namely the GS selection changes from the $\sqrt{3} \times \sqrt{3}$ state to the $q = 0$ state at a critical $\Delta_c$, as varying the anisotropy parameter $\Delta$ from $\Delta = 1$ to $\Delta = 0$. The energy difference $\delta e$ between the $\sqrt{3} \times \sqrt{3}$ and the $q = 0$ state calculated by the CCM, e.g. for $s = 5$, is in excellent agreement with the large-$s$ SWT data of Ref. 24. Moreover, we find also in accordance with Ref. 24 that the region of disorder in the GS $s - \Delta$ phase diagram grows with growing $\Delta$.

Since our CCM approach is not limited to large $s$, we can overcome some limitations of the SWT of Ref. 24. The CCM result for the critical $\Delta_c$ depends on $s$ by contrast to the $s$-independent SWT value. For $s = 1/2$ the SWT yields GS magnetic LRO for $\Delta \leq 0.95$, whereas the CCM (in agreement with DMRG results\textsuperscript{25}) yields always a disordered GS.

Knowing these limitations of the SWT, Chernyshev and Zhitomirsky\textsuperscript{22} proposed a tentative GS $s - \Delta$ phase diagram, see Fig. 4b of Ref. 24. However, our phase diagram determined from the CCM results differs from that conjectured by Chernyshev and Zhitomirsky. We find that $\Delta_c$ increases with increasing $s$, whereas Chernyshev and Zhitomirsky speculate that $\Delta_c$ may decrease for larger $s$. Hence our results indicate that the region of $\sqrt{3} \times \sqrt{3}$ GS LRO is much larger than that proposed in Ref. 24. As a consequence, the (fictional) transition from the magnetically disordered GS to a state with magnetic LRO obtained by a continuous increase of spin quantum number $s$ starting from the extreme quantum case $s = 1/2$ is always a transition to the state with $\sqrt{3} \times \sqrt{3}$ LRO, while in Ref. 24 it is suggested that the transition is to the state with $q = 0$ LRO. Moreover, for $s = 3/2$ we find $\sqrt{3} \times \sqrt{3}$ LRO for $0.525 < \Delta < 1$, while in Ref. 24 $q = 0$ LRO is suggested for the entire region $0 \leq \Delta < 1$.

We conclude, that the interplay of frustration, quantum fluctuations and anisotropy leads to a rich ground-state phase diagram of the XXZ spin-$s$ KAFM. Bearing in mind the numerous investigations of the isotropic Heisenberg KAFM, see, e.g., Refs. 31 and references therein, the anisotropic model provides a challenging playground to apply the toolbox of frustrated quantum magnetism on this so far little investigated problem.

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2 A. B. Harris, C. Kallin and A. J. Berlinsky, Phys. Rev. B 45, 2899 (1992).
For technical reasons it is convenient to start with a Hamiltonian, where the anisotropy parameter $\Delta$ stands in front of the $y$-components of the spin instead of the $z$-components, see also Ref. 24.

Since for $s = \frac{1}{2}$ typically the notation LSUB$n$ is used, it is in order to mention here that for $s = \frac{1}{2}$ the LSUB$n$ scheme is identical to the SUB$n$-$n$ scheme.

For the numerical calculation we use the program package ’The crystallographic CCM’ (D. J. J. Farnell and J. Schulenburg).

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