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 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 Δ 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 region $0 \leqslant \Delta \leqslant 1$. For $s = 1$ the ground state is disordered for $0.818 < \Delta \leqslant 1$, it exhibits $\sqrt{3}\times\sqrt{3}$ magnetic long-range order for $0.281 < \Delta < 0.818$, and $\mathbf{q} = \mathbf{0}$ magnetic long-range order for $0 \leqslant \Delta < 0.281$. 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 Δ close to 1 (Heisenberg limit), i.e., $\mathbf{q} = \mathbf{0}$ magnetic order is favored over $\sqrt{3}\times\sqrt{3}$ for $0 \leqslant \Delta < \Delta_c$ and vice versa for $\Delta_c < \Delta \leqslant 1$. We calculate the critical anisotropy parameter Δ_c as a function of the spin quantum number s.

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I. 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 have been applied to understand the GS properties of the kagome antiferromagnet (KAFM); see, e.g., Refs. [1–32]. 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 nonmagnetic quantum GS. Recent large-scale numerics [14,15,19] provide strong arguments for a gapped $\mathbb{Z}_2$ topological spin-liquid GS for spin $s = 1/2$. Other recent investigations have been focused on higher spin $s > 1/2$ [18,25,28–32] and also on the anisotropic XXZ model [10,25,26]. Both modifications have relevance for the experimental research; see, e.g., Refs. [33–41]. Moreover, anisotropic spin models are of great interest with respect to engineering models of quantum magnetism on optical lattices, see, e.g., Refs. [42–44]. Since higher spin as well as anisotropy, in general, lead to a reduction of quantum fluctuations, see, e.g., Refs. [45–51], GS magnetic LRO for the KAFM might be facilitated. However, for the isotropic $s = 1$ Heisenberg KAFM there is clear evidence that there is no magnetic LRO [18,29–32]. For $s > 1$ several approaches lead to indications for $\sqrt{3}\times\sqrt{3}$ GS LRO [3,4,6,18,25,28]. On the other hand, recent density matrix group (DMRG) calculations [26] have demonstrated that for $s = 1/2$ the XXZ KAFM remains in a magnetically disordered GS for the entire range of the anisotropy parameter Δ between the XY point ($\Delta = 0$) and the Heisenberg point ($\Delta = 1$).

Motivated by the recent Letter of Chernyshev and Zhitomirsky (CZ) [25] we use the coupled cluster method (CCM) to high orders of approximation to calculate the s-Δ GS phase diagram of the spin-s XXZ KAFM with easy-plane anisotropy. The corresponding Hamiltonian is

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

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. [12,18,20,47,49–61]. 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 the best available DMRG results [14,19].

II. THE COUPLED CLUSTER METHOD

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. [62–68]. We first mention that the CCM yields results directly in the thermodynamic limit.

First we choose a normalized reference state $|\Phi\rangle$ that is typically a classical GS of the model. From a quasiclassical point of view the coplanar $\sqrt{3}\times\sqrt{3}$ and $\mathbf{q} = \mathbf{0}$ states are favored candidates among the massively degenerate manifold of classical GS's (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 [69]. 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: $|\Phi\rangle = |\downarrow\downarrow\downarrow\cdots\rangle$; $C_I^+ = s_n^+, s_n^+ s_m^+, s_n^+ s_m^+ s_k^+, \ldots, s_n^+ \equiv s_n^x + i s_n^y$, where the spin operators are defined in the local rotated coordinate frames, and the indices $n,m,k,\ldots$ denote arbitrary lattice sites. Then the CCM parametrizations of the ket and bra GS eigenvectors $|\Psi\rangle$ and $\langle\tilde{\Psi}|$ of the spin system are given by $|\Psi\rangle = e^S|\Phi\rangle$, $S = \sum_{I\neq 0} a_I C_I^+$; $\langle\tilde{\Psi}| = \langle\Phi|\tilde{S}e^{-S}$, $\tilde{S} = 1 + \sum_{I\neq 0} \tilde{a}_I C_I^-$. The coefficients a_I and $\tilde{a}_I$ contained in the CCM correlation operators, S and $\tilde{S}$, are determined by the CCM ket-state and bra-state equations $\langle\Phi|C_I^- e^{-S} H e^S|\Phi\rangle = 0$; $\langle\Phi|\tilde{S}e^{-S}[H, C_I^+]e^S|\Phi\rangle = 0; \forall I \neq 0$. Each ket-state or bra-state equation belongs to a certain configuration index I, i.e., it corresponds to a certain configuration of lattice sites $n,m,k,\ldots$. Using the Schrödinger equation, $H|\Psi\rangle = E_0|\Psi\rangle$, we can now write the GS energy as $E_0 = \langle\Phi|e^{-S}He^S|\Phi\rangle$. The magnetic order parameter (sublattice magnetization) is 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 transformed coordinate system, and $N(\to\infty)$ is the number of lattice sites. We have to use an appropriate approximation scheme in order to truncate the expansions of S and $\tilde{S}$. For that we use the well established SUBn-n approximation scheme; cf., e.g., Refs. [12,18,20,47,49–61]. In the SUBn-n scheme we include no more than n spin flips spanning a range of no more than n contiguous lattice sites [70]. Using an efficient parallelized CCM code [71] 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 $s = 1/2, 1, \ldots, 9/2, 5$. The maximum number of ket-state equations considered here is 416 193 for $s = 5$. Following Ref. [18] we extrapolate the "raw" SUBn-n data to the limit $n \to \infty$ 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 [12,18,20,49–58] $e_0(n) = a_0 + a_1(1/n)^2 + a_2(1/n)^4$ and $M(n) = b_0 + b_1(1/n)^{1/2} + b_2(1/n)^{3/2}$.

III. RESULTS

We use the CCM to calculate the GS s-Δ phase diagram. Such a phase diagram has been very recently presented by CZ [25] using nonlinear spin-wave theory (SWT) and real-space perturbation theory (RSPT). To have the necessary information available for the comparison between the results of CZ and our CCM results reported below, let us first briefly report the main findings of Ref. [25]. It is well known [2,4,6,18] that in the large-s limit for $\Delta = 1$, quantum fluctuations select the $\sqrt{3}\times\sqrt{3}$ state. In Ref. [25] it was found that this GS selection is preserved for weak easy-plane anisotropy, i.e., for $\Delta_c < \Delta \leqslant 1$. However, the main and unexpected result of Ref. [25] is the selection of the $\mathbf{q} = \mathbf{0}$ GS for smaller values of Δ, i.e., for $0 \leqslant \Delta < \Delta_c$. 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 [2,5,72–74]. 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 SWT responsible for the GS selection is of order $\mathcal{O}(1)$, and, therefore, the critical anisotropy $\Delta_c = 0.72235$ is found to be independent of the spin s within this approximation [25]. The RSPT provides insight into the mechanism of the quantum selection of the GS: Some relevant seventh-order processes change their sign as varying Δ [25]. The magnetic order parameter calculated in linear SWT shows a clear trend to magnetic LRO as lowering Δ. However, this trend is substantially overestimated by SWT, since already for $s = 1/2$ the GS is found to exhibit magnetic LRO for $\Delta < 0.95$. On the other hand, for the $\Delta = 1$ it is known that the linear SWT yields a vanishing order parameter for any value of s [1,25]. Both findings for the order parameter are in contradiction to recent results obtained by large-scale numerical approaches [18,26].

We now discuss our CCM results. In Figs. 1 and 2 we show the GS energy per spin e_0 and the magnetic order parameter M, respectively, extrapolated to $n \to \infty$ for both reference states as a function of Δ for $s = 1/2, \ldots, 5$. In general,

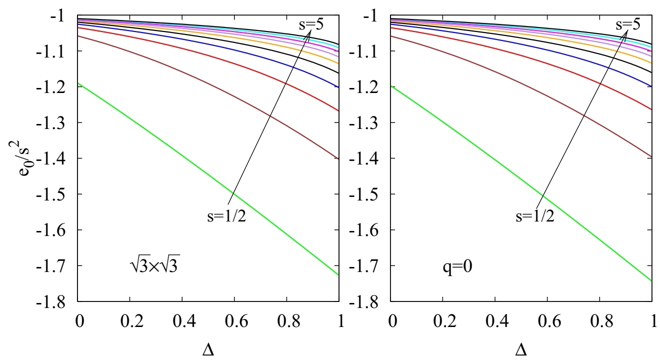

FIG. 1. (Color online) The extrapolated CCM GS energy per spin $e_0|_{n\to\infty}$ using the $\sqrt{3}\times\sqrt{3}$ reference state (left) and the $\mathbf{q} = \mathbf{0}$ reference state (right) as a function of the anisotropy parameter Δ for spin $s = 1/2, 1, 3/2, \ldots, 9/2, 5$.

the $\sqrt{3}\times\sqrt{3}$ and the $\mathbf{q} = \mathbf{0}$ cases behave very similarly. The GS energy for $s = 1/2$ is almost linearly growing with decreasing of Δ. For larger s the $e_0(\Delta)$ curve noticeably deviates from linearity, particularly near $\Delta = 1$. This trend that the influence of the anisotropy Δ becomes exceedingly large near the Heisenberg limit is more pronounced for the order parameter; see Fig. 2. For spin $s \geqslant 3/2$ 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 SWT, where the $1/s$ corrections diverge for $\Delta \to 1$ [25]. 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 $0 \leqslant \Delta \leqslant 1$ in accordance with recent DMRG calculations [26]. For $s = 1$ we find a finite region of disorder, $\Delta^* \leqslant \Delta \leqslant 1$, for both reference states, where $\Delta^* = 0.818\,(\Delta^* = 0.945)$ for the $\sqrt{3}\times\sqrt{3}\,(\mathbf{q} = \mathbf{0})$ state. Note, however, that for anisotropies around Δ^* the CCM GS energy for the $\sqrt{3}\times\sqrt{3}$ reference state is lower than that for the $\mathbf{q} = \mathbf{0}$ reference state; see below. We mention also that a table of values for e_0 and M at $\Delta = 1$ and for spin up to $s = 3$ can be found in Ref. [18]. We show corresponding values for the XY limit ($\Delta = 0$) in Table I of the present paper.

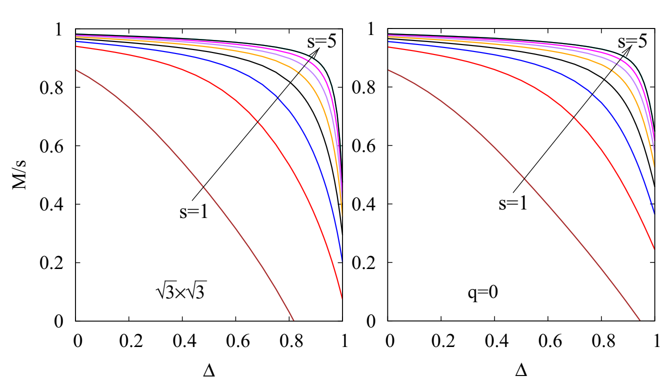

FIG. 2. (Color online) The extrapolated CCM GS sublattice magnetization $M|_{n\to\infty}$ using the $\sqrt{3}\times\sqrt{3}$ reference state (left) and the $\mathbf{q} = \mathbf{0}$ reference state (right) as a function of the anisotropy parameter Δ for spin $s = 1, 3/2, \ldots, 9/2, 5$. Note that for $s = 1/2$ no data are shown, since there is no magnetic LRO in the entire region $0 \leqslant \Delta \leqslant 1$.

TABLE I. Extrapolated CCM results for GS energy per spin $e_0|_{n\to\infty}$ and the GS sublattice magnetization $M|_{n\to\infty}$ using the $\sqrt{3}\times\sqrt{3}$ and the $\mathbf{q}=\mathbf{0}$ reference states for $\Delta=0$.

	$\sqrt{3}\times\sqrt{3}$		$\mathbf{q}=\mathbf{0}$	
	e_0/s^2	M/s	e_0/s^2	M/s
$s=1/2$	−1.1896	< 0	−1.1968	< 0
$s=1$	−1.0578	0.8602	−1.0583	0.8589
$s=3/2$	−1.0347	0.9402	−1.0349	0.9368
$s=2$	−1.0252	0.9570	−1.0253	0.9556
$s=5/2$	−1.0198	0.9664	−1.0199	0.9656
$s=3$	−1.0163	0.9723	−1.0164	0.9719

Next we discuss the GS selection. For that we consider the energy difference between the $\mathbf{q}=\mathbf{0}$ and the $\sqrt{3}\times\sqrt{3}$ states $\delta e = e_0^{q=0} - e_0^{\sqrt{3}\times\sqrt{3}}$; see Fig. 3. The main common feature of the $\delta e(\Delta)$ curves is the change of the sign of δe, i.e., in accord with Ref. [25] we find a change in the GS selection from the $\sqrt{3}\times\sqrt{3}$ state to the $\mathbf{q}=\mathbf{0}$ state at a critical value Δ_c as varying the anisotropy from $\Delta=1$ to $\Delta=0$. The magnitude of δe is small, in particular for smaller Δ. For larger s it agrees very well with the SWT data of Ref. [25] up to $\Delta\sim 0.9$. The stronger deviation for Δ close to 1 can be attributed to the divergence of the $1/s$ corrections for $\Delta\to 1$ [25].

As already mentioned above, the large-s SWT [25] yields a critical value Δ_c that is independent of s. However, Δ_c certainly depends on s. CZ [25] suggested that Δ_c may increase for smaller spins from the large-s value $\Delta_c = 0.72235$ of $s\to\infty$. Our CCM approach yields directly Δ_c as a function of s; see the black open circles in Fig. 4. Contrary to the conjecture of CZ, see Fig. 4(b) in Ref. [25], we find that Δ_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 $s\to\infty$ yields $\lim_{s\to\infty}\Delta_c = 0.727$, where we have used $s=3,7/2,4,9/2,5$ for the extrapolation. This CCM estimate

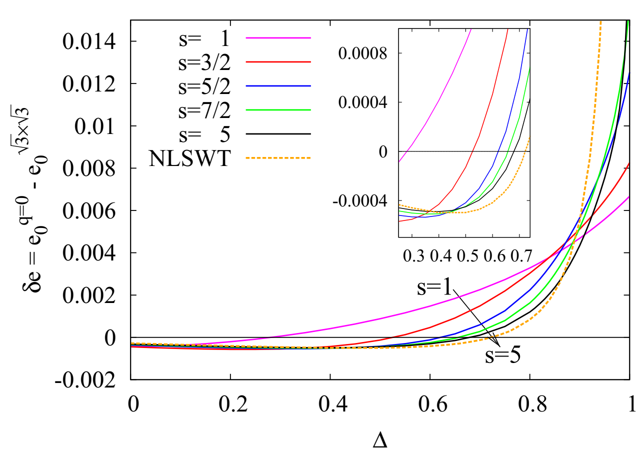

FIG. 3. (Color online) Difference $\delta e = e_0^{q=0} - e_0^{\sqrt{3}\times\sqrt{3}}$ of the extrapolated GS energies of the $\sqrt{3}\times\sqrt{3}$ and the $\mathbf{q}=\mathbf{0}$ states as a function of the anisotropy parameter Δ for spin $s=1,3/2,5/2,7/2,5$. We also show the corresponding large-s results of Ref. [25] (labeled by "NLSWT") obtained by nonlinear SWT. The inset shows an enlarged scale of that region of Δ, where δe changes its sign.

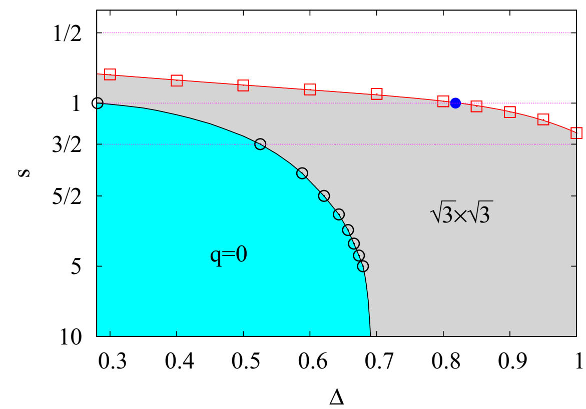

FIG. 4. (Color online) GS s-Δ phase diagram of the XXZ spin-s kagome antiferromagnet. The open circles connected by the black solid line show the critical anisotropy $\Delta_c(s)$, where the GS selection changes between the $\sqrt{3}\times\sqrt{3}$ state and $\mathbf{q}=\mathbf{0}$ state. The upper red line connecting the open squares is the fictional transition line between GS disorder and GS LRO, which is obtained by considering the spin s as a continuous quantity. The blue point indicates the transition point $\Delta^*=0.818$ between the disordered GS and the GS with $\sqrt{3}\times\sqrt{3}$ LRO for $s=1$ obtained directly from the $M(\Delta)$ curve shown in Fig. 2.

of the large-s limit of Δ_c is in excellent agreement with the large-s SWT result [25].

To get the full relationship to the GS s-Δ phase diagram given in Fig. 4 of Ref. [25] we may consider the spin 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 Δ 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, which 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 $0.818<\Delta\leqslant 1$, the ordered $\sqrt{3}\times\sqrt{3}$ state for $0.281<\Delta<0.818$, and the ordered $\mathbf{q}=\mathbf{0}$ state for $0\leqslant\Delta<0.281$. For spin $s>1$ there are two magnetically ordered GS phases, where the phase boundary is given by the black solid line in Fig. 4.

IV. SUMMARY

We summarize our findings by comparing our GS s-Δ phase diagram (Fig. 4) with that of Ref. [25]. Most importantly, we get the same trend as Ref. [25], namely, the GS selection changes from the $\sqrt{3}\times\sqrt{3}$ state to the $\mathbf{q}=\mathbf{0}$ state at a critical Δ_c as varying the anisotropy Δ from $\Delta=1$ to $\Delta=0$. The energy difference δe between the $\sqrt{3}\times\sqrt{3}$ and the $\mathbf{q}=\mathbf{0}$ state calculated by the CCM, e.g., for $s=5$, is in excellent agreement with the large-s SWT data of Ref. [25]. Moreover, we find also in accordance with Ref. [25] that the region of disorder in the GS s-Δ phase diagram decreases with shrinking Δ. This narrowed region of disorder can be attributed to reduced quantum fluctuations as increasing anisotropy (lowering Δ); see, e.g., Ref. [47].

Since our CCM approach is not limited to large s, we can overcome some limitations of the SWT of Ref. [25]: The CCM result for the critical Δ_c depends on s by contrast to the s-independent SWT value. For $s = 1/2$ the SWT yields GS magnetic LRO for $\Delta \lesssim 0.95$, whereas the CCM (in agreement with DMRG results [26]) yields always a disordered GS.

Knowing these limitations of the SWT, CZ [25] proposed a tentative GS s-Δ phase diagram; see Fig. 4(b) of Ref. [25]. However, our CCM phase diagram differs from that conjectured by CZ. We find that Δ_c increases with increasing s, whereas CZ speculate that Δ_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. [25]. As a consequence, the (fictional) transition from the magnetically disordered GS to a state with magnetic LRO obtained by a continuous increase of spin 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. [25] it is suggested that the transition is to the state with $\mathbf{q} = \mathbf{0}$ LRO. Moreover, for $s = 3/2$ we find $\sqrt{3} \times \sqrt{3}$ LRO for $0.525 < \Delta \leqslant 1$, while in Ref. [25] $\mathbf{q} = \mathbf{0}$ LRO is suggested for the entire region $0 \leqslant \Delta \leqslant 1$.

We conclude, that the interplay of frustration, quantum fluctuations, and anisotropy leads to a rich GS phase diagram of the XXZ spin-s KAFM. Bearing in mind the numerous investigations of the isotropic Heisenberg KAFM, see, e.g., Refs. [1–32], 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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[1] C. Zeng and V. Elser, Phys. Rev. B **42**, 8436 (1990).
[2] A. B. Harris, C. Kallin, and A. J. Berlinsky, Phys. Rev. B **45**, 2899 (1992).
[3] S. Sachdev, Phys. Rev. B **45**, 12377 (1992).
[4] A. Chubukov, Phys. Rev. Lett. **69**, 832 (1992).
[5] J. N. Reimers and A. J. Berlinsky, Phys. Rev. B **48**, 9539 (1993).
[6] C. L. Henley and E. P. Chan, J. Magn. Magn. Mater. **140–144**, 1693 (1995).
[7] C. Waldtmann, H.-U. Everts, B. Bernu, C. Lhuillier, P. Sindzingre, P. Lecheminant, and L. Pierre, Eur. Phys. J. B **2**, 501 (1998).
[8] S. Capponi, A. Läuchli, and M. Mambrini, Phys. Rev. B **70**, 104424 (2004).
[9] R. R. P. Singh and D. A. Huse, Phys. Rev. B **76**, 180407(R) (2007).
[10] O. Cepas, C. M. Fong, P. W. Leung, and C. Lhuillier, Phys. Rev. B **78**, 140405(R) (2008).
[11] P. Sindzingre and C. Lhuillier, Europhys. Lett. **88**, 27009 (2009).
[12] R. F. Bishop, P. H. Y. Li, D. J. J. Farnell, and C. E. Campbell, Phys. Rev. B **82**, 104406 (2010).
[13] G. Evenbly and G. Vidal, Phys. Rev. Lett. **104**, 187203 (2010).
[14] S. Yan, D. A. Huse, and S. R. White, Science **332**, 1173 (2011).
[15] A. M. Läuchli, J. Sudan, and E. S. Sørensen, Phys. Rev. B **83**, 212401 (2011).
[16] Y.-M. Lu, Y. Ran, and P. A. Lee, Phys. Rev. B **83**, 224413 (2011).
[17] H. Nakano and T. Sakai, J. Phys. Soc. Jpn. **80**, 053704 (2011).
[18] O. Götze, D. J. J. Farnell, R. F. Bishop, P. H. Y. Li, and J. Richter, Phys. Rev. B **84**, 224428 (2011).
[19] S. Depenbrock, I. P. McCulloch, and U. Schollwöck, Phys. Rev. Lett. **109**, 067201 (2012).
[20] P. H. Y. Li, R. F. Bishop, C. E. Campbell, D. J. J. Farnell, O. Götze, and J. Richter, Phys. Rev. B **86**, 214403 (2012).
[21] Y. Iqbal, F. Becca, S. Sorella, and D. Poilblanc, Phys. Rev. B **87**, 060405(R) (2013).
[22] R. Suttner, C. Platt, J. Reuther, and R. Thomale, Phys. Rev. B **89**, 020408 (2014).
[23] Z. Y. Xie, J. Chen, J. F. Yu, X. Kong, B. Normand, and T. Xiang, Phys. Rev. X **4**, 011025 (2014).
[24] I. Rousochatzakis, Y. Wan, O. Tchernyshyov, and F. Mila, Phys. Rev. B **90**, 100406(R) (2014).
[25] A. L. Chernyshev and M. E. Zhitomirsky, Phys. Rev. Lett. **113**, 237202 (2014).
[26] Y. C. He and Y. Chen, Phys. Rev. Lett. **114**, 037201 (2015).
[27] W. Zhu, S. S. Gong, and D. N. Sheng, arXiv:1410.4883 [cond-mat.str-el].
[28] O. Cépas and A. Ralko, Phys. Rev. B **84**, 020413 (2011).
[29] H. J. Changlani and A. M. Läuchli, arXiv:1406.4767 [cond-mat.str-el].
[30] T. Liu, W. Li, A. Weichselbaum, J. von Delft, and G. Su, arXiv:1406.5905 [cond-mat.str-el].
[31] S. Nishimoto and M. Nakamura, arXiv:1409.5870 [cond-mat.str-el].
[32] W. Li, A. Weichselbaum, J. von Delft, and H.-H. Tu, arXiv:1412.7123 [cond-mat.str-el].
[33] A. S. Wills, A. Harrison, C. Ritter, and R. I. Smith, Phys. Rev. B **61**, 6156 (2000).
[34] B. Fak, F. C. Coomer, A. Harrison, D. Visser, and M. E. Zhitomirsky, Europhys. Lett. **81**, 17006 (2008).
[35] S.-H. Lee, C. Broholm, M. F. Collins, L. Heller, A. P. Ramirez, Ch. Kloc, E. Bucher, R. W. Erwin, and N. Lacevic, Phys. Rev. B **56**, 8091 (1997).
[36] Y. J. Uemura, A. Keren, K. Kojima, L. P. Le, G. M. Luke, W. D. Wu, Y. Ajiro, T. Asano, Y. Kuriyama, M. Mekata, H. Kikuchi, and K. Kakurai, Phys. Rev. Lett. **73**, 3306 (1994); B. Martinez, F. Sandiumenge, A. Rouco, A. Labarta, J. Rodriguez-Carvajal, M. Tovar, M. T. Causa, S. Gali, and X. Obradors, Phys. Rev. B **46**, 10786 (1992).
[37] D. E. Freedman, R. Chisnell, T. M. McQueen, Y. S. Lee, C. Payen, and D. G. Nocera, Chem. Commun. **48**, 64 (2012).
[38] K. Matan, D. Grohol, D. G. Nocera, T. Yildirim, A. B. Harris, S. H. Lee, S. E. Nagler, and Y. S. Lee, Phys. Rev. Lett. **96**, 247201 (2006).
[39] A. Zorko, S. Nellutla, J. van Tol, L. C. Brunel, F. Bert, F. Duc, J.-C. Trombe, M. A. de Vries, A. Harrison, and P. Mendels, Phys. Rev. Lett. **101**, 026405 (2008).
[40] A. Zorko, F. Bert, A. Ozarowski, J. van Tol, D. Boldrin, A. S. Wills, and P. Mendels, Phys. Rev. B **88**, 144419 (2013).

[41] T. Ono, K. Matan, Y. Nambu, T. J. Sato, K. Katayama, S. Hirata, and H. Tanaka, J. Phys. Soc. Jpn. **83**, 043701 (2014); K. Katayama, N. Kurita, and H. Tanaka, arXiv:1412.5770 [cond-mat.str-el].
[42] S. V. Isakov, S. Wessel, R. G. Melko, K. Sengupta, Yong Baek Kim, Phys. Rev. Lett. **97**, 147202 (2006).
[43] I. Bloch, J. Dalibard, and W. Zwerger, Rev. Mod. Phys. **80**, 885 (2008).
[44] J. Struck, M. Weinberg, C. Ölschläger, P. Windpassinger, J. Simonet, K. Sengstock, R. Höppner, P. Hauke, A. Eckardt, M. Lewenstein, and L. Mathey, Nat. Phys. **9**, 738 (2013).
[45] T. Pardini and R. R. P. Singh, Phys. Rev. B **77**, 214433 (2008).
[46] H. Nakano, S. Todo, and T. Sakai, J. Phys. Soc. Jpn. **82**, 043715 (2013).
[47] R. F. Bishop, P. H. Y. Li, R. Darradi, J. Schulenburg, and J. Richter, Phys. Rev. B **78**, 054412 (2008).
[48] E. A. Ghioldi, A. Mezio, L. O. Manuel, R. R. P. Singh, J. Oitmaa, and A. E. Trumper, arXiv:1411.5298 [cond-mat.str-el].
[49] R. F. Bishop, P. H. Y. Li, R. Darradi, and J. Richter, Europhys. Lett. **83**, 47004 (2008).
[50] P. H. Y. Li and R. F. Bishop, Eur. Phys. J. B **85**, 25 (2012).
[51] R. F. Bishop, P. H. Y. Li, and C. E. Campbell, Phys. Rev. B **89**, 214413 (2014).
[52] N. B. Ivanov, J. Richter, and D. J. J. Farnell, Phys. Rev. B **66**, 014421 (2002).
[53] D. Schmalfuß, R. Darradi, J. Richter, J. Schulenburg, and D. Ihle, Phys. Rev. Lett. **97**, 157201 (2006).
[54] R. Darradi, O. Derzhko, R. Zinke, J. Schulenburg, S. E. Krüger, and J. Richter, Phys. Rev. B **78**, 214415 (2008).
[55] R. F. Bishop, P. H. Y. Li, R. Darradi, and J. Richter, J. Phys.: Condens. Matter **20**, 255251 (2008).
[56] D. J. J. Farnell, R. Zinke, J. Schulenburg, and J. Richter, J. Phys.: Condens. Matter **21**, 406002 (2009).
[57] J. Richter, R. Darradi, J. Schulenburg, D. J. J. Farnell, and H. Rosner, Phys. Rev. B **81**, 174429 (2010).
[58] D. J. J. Farnell, R. F. Bishop, P. H. Y. Li, J. Richter, and C. E. Campbell, Phys. Rev. B **84**, 012403 (2011).
[59] J. Reuther, P. Wölfle, R. Darradi, W. Brenig, M. Arlego, and J. Richter, Phys. Rev. B **83**, 064416 (2011).
[60] P. H. Y. Li, R. F. Bishop, and C. E. Campbell, Phys. Rev. B **88**, 144423 (2013).
[61] J. J. Jiang, F. Tang, and C. H. Yang, arXiv:1406.4317 [cond-mat.str-el].
[62] C. Zeng, D. J. J. Farnell, and R. F. Bishop, J. Stat. Phys. **90**, 327 (1998).
[63] R. F. Bishop, in *Microscopic Quantum Many-Body Theories and Their Applications*, edited by J. Navarro and A. Polls, Lecture Notes in Physics Vol. 510 (Springer, Berlin, 1998), p. 1.
[64] R. F. Bishop, D. J. J. Farnell, and C. Zeng, Phys. Rev. B **59**, 1000 (1999).
[65] R. F. Bishop, D. J. J. Farnell, S. E. Krüger, J. B. Parkinson, J. Richter, and C. Zeng, J. Phys.: Condens. Matter **12**, 6887 (2000).
[66] D. J. J. Farnell, R. F. Bishop, and K. A. Gernoth, J. Stat. Phys. **108**, 401 (2000).
[67] D. J. J. Farnell and R. F. Bishop, in *Quantum Magnetism*, edited by U. Schollwöck, J. Richter, D. J. J. Farnell, and R. F. Bishop, Lecture Notes in Physics Vol. 645 (Springer, Berlin, 2004), p. 307.
[68] S. E. Krüger, R. Darradi, J. Richter, and D. J. J. Farnell, Phys. Rev. B **73**, 094404 (2006).
[69] For technical reasons it is convenient to start with a Hamiltonian, where the anisotropy parameter Δ stands in front of the y components of the spin instead of the z components; see also Ref. [25].
[70] Since for $s = 1/2$ typically the notation LSUBn is used, it is in order to mention here that for $s = 1/2$ the LSUBn scheme is identical to the SUBn-n scheme.
[71] For the numerical calculation we use the program package by D. J. J. Farnell and J. Schulenburg, THE CRYSTALLOGRAPHIC CCM, http://www-e.uni-magdeburg.de/jschulen/ccm/
[72] D. A. Huse and A. D. Rutenberg, Phys. Rev. B **45**, 7536 (1992).
[73] C. L. Henley, Phys. Rev. B **80**, 180401(R) (2009).
[74] S. E. Korshunov, Phys. Rev. B **65**, 054416 (2002).