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Phys. Rev. B **93**, 014424 — Published 19 January 2016

DOI: [10.1103/PhysRevB.93.014424](https://doi.org/10.1103/PhysRevB.93.014424)

Competing orders in spin-1 and spin-3/2 XXZ Kagome antiferromagnets: A series expansion study

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(Dated: December 30, 2015)

We study the competition between $\sqrt{3} \times \sqrt{3}$ (RT3) and $q = 0$ (Q0) magnetic orders in spin-one and spin-3/2 Kagome-lattice XXZ antiferromagnets with varying XY anisotropy parameter Δ , using series expansion methods. The Hamiltonian is split into two parts: an H_0 which favors the classical order in the desired pattern and an H_1 , which is treated in perturbation theory by a series expansion. We find that the ground state energy series for the RT3 and Q0 phases are identical up to sixth order in the expansion, but ultimately a selection occurs, which depends on spin and the anisotropy Δ . Results for ground state energy and the magnetization are presented. These results are compared with recent spin-wave theory and coupled-cluster calculations. The series results for the phase diagram are close to the predictions of spin-wave theory. For the spin-one model at the Heisenberg point ($\Delta = 1$), our results are consistent with a vanishing order parameter, that is an absence of a magnetically ordered phase. We also develop series expansions for the ground state energy of the spin-one Heisenberg model in the trimerized phase. We find that the ground state energy in this phase is lower than those of magnetically ordered ones, supporting the existence of a spontaneously trimerized phase in this model.

PACS numbers:

Kagome lattice antiferromagnets have been studied extensively both theoretically and experimentally over the last few decades^{1,2}. There is, by now, very strong numerical evidence that the ground state of the nearest-neighbor spin-half Heisenberg model on the Kagome-lattice is a quantum spin-liquid and has no long-range magnetic order³. However, the more general XXZ model for larger spin and with XY anisotropy may well have long-range magnetic order⁴. Indeed, several experimental Kagome systems with large spin are known to have magnetic long-range order⁵.

Competing magnetic orders in these models were investigated recently by Chernyshev and Zhitomirsky⁶ using non-linear spin-wave theory and real-space perturbation theory, where they found a phase diagram with competing $\sqrt{3} \times \sqrt{3}$ (RT3) and $q = 0$ (Q0) magnetic orders at different spin and anisotropy Δ values. The models have also been studied recently using the coupled cluster method by Gotze and Richter⁷ who found a similar but not identical phase diagram to spin-wave theory. The main difference in the phase diagram was that in the coupled-cluster calculations the RT3 phase occupies a significantly bigger region of the phase diagram at the expense of the Q0 phase. The purpose of this paper is to study the competing magnetic phases by series expansion methods. Our numerical results appear much closer to the spin-wave theory.

We consider the antiferromagnetic XXZ model on the Kagome lattice with Hamiltonian

$$\mathcal{H} = \sum_{\langle i,j \rangle} (S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z) \quad (1)$$

where the sum is over the nearest neighbor pairs and Δ is the anisotropy parameter. We will study spin-one and spin-3/2 model with various Δ values less than or equal to unity corresponding to XY anisotropy ($\Delta < 1$) and Heisenberg symmetry ($\Delta = 1$).

To carry out the series expansion around a particular non-collinear ordered state, we rotate our axis of quantization at each site so the local z axis points along the ordering direction, and reexpress the Hamiltonian in this rotated basis^{8,9}. In this basis the ferromagnetic zz coupling will lead to order in the desired classical pattern. Thus by splitting the Hamiltonian into such an Ising term and calling it the unperturbed Hamiltonian and treating the rest of the Hamiltonian by perturbation theory, we can calculate the properties of the system in the ordered phase^{10,11}. The Hamiltonian we end up with takes the form:

$$\mathcal{H} = H_0 + \lambda H_1 + t(1 - \lambda) \sum_i S_i^z, \quad (2)$$

with

$$H_0 = -\frac{1}{2} \sum_{\langle ij \rangle} S_i^z S_j^z \quad (3)$$

and

$$\begin{aligned} H_1 = & -\frac{1}{8}(1 + 2\Delta) \sum_{\langle ij \rangle} (S_i^+ S_j^+ + S_i^- S_j^-) \quad (4) \\ & -\frac{1}{8}(1 - 2\Delta) \sum_{\langle ij \rangle} (S_i^+ S_j^- + S_i^- S_j^+) \\ & + \frac{\sqrt{3}}{4} \sum_{\langle ij \rangle} \eta_{ij} \{ S_i^z (S_j^+ + S_j^-) - (S_i^+ + S_i^-) S_j^z \} \end{aligned}$$

with

$$\begin{aligned} \eta_{ij} = +1 & \quad A \rightarrow B, B \rightarrow C, C \rightarrow A \\ -1 & \quad B \rightarrow A, C \rightarrow B, A \rightarrow C \end{aligned} \quad (5)$$

where A,B, and C are the three sublattices. The XXZ model of interest only arises at $\lambda = 1$. Thus, the parameter t can be varied to improve convergence as it does not play any role at $\lambda = 1$. We have studied the model for different spin and anisotropy Δ .

Our interesting finding is that regardless of spin, anisotropy Δ and redundant field value t , the ground state energy for RT3 and Q0 phases are identical to 6th order in the series expansion. This high degree of degeneracy is reminiscent of the q -independence of the high temperature susceptibility for the classical Kagome antiferromagnet to high orders¹² and the high-temperature order-parameter susceptibility degeneracy for the XY pyrochlore antiferromagnets¹³. Here, the degeneracy is for

the ground state energy. The degeneracy is lifted in 7th order. The difference between the ground state energy for RT3 and Q0 phases are given order by order in Table I for spin-one models and in Table II for spin-3/2 models.

The degeneracy to 6th order and its lifting at 7th order can be understood in terms of the general real-space perturbation theory arguments provided by Chernyshev and Zhitomirsky⁶. These authors showed that for a general 120 degree classical ground state order, bilinear real space perturbations can not lift the degeneracy below 7th order. In order to lift the degeneracy virtual spin-flips must be created, propagate along a closed loop and then annihilate. On the Kagome lattice, the smallest such ‘tunneling’ paths are the hexagons for which the lowest order in perturbation theory, in which the degeneracy can be lifted, is 7th order. Thus, our series expansion results are a special case of this deep ‘topological’ or ‘protected’ nature of the ground state degeneracy of all 120 degree ordered states on the Kagome lattice.

TABLE I: Difference between ground state energy series of RT3 and Q0 phases for $S = 1$ model

Δ	t	$n = 7$	$n = 8$	$n = 9$	$n = 10$	$n = 11$	$n = 12$
1.0	0.0	1.79281056E-05	-.00228441779	.00485266477	-.0131501048	.0244393622	-.048490238
1.0	0.5	-1.99373138E-05	-.000359989266	-4.05357305E-05	-.000541912718	-.000190168854	-.000584500274
1.0	1.0	-1.00496807E-05	-9.61299295E-05	-.000130394373	-.00017789055	-.000217321242	-.0002516233
0.8	0.0	.000337078044	-.00123165186	.0025634538	-.00520062285	.00822960621	-.0138676947
0.8	0.5	6.06764982E-05	-9.81439873E-05	4.17426967E-05	-.000109903463	-4.91074816E-05	-.000138445195
0.8	1.0	1.58188574E-05	-5.15123324E-06	-1.36613326E-05	-2.32793478E-05	-3.57282811E-05	-5.02830945E-05
0.6	0.0	.000368046153	-.000640326014	.00145292023	-.00238946797	.00383705524	-.00610563131
0.6	0.5	7.40771254E-05	4.96763165E-06	8.15631813E-05	1.48517701E-05	5.09012595E-05	-5.98002479E-06
0.6	1.0	2.09523323E-05	2.48510186E-05	3.090978E-05	3.26457591E-05	3.16691472E-05	2.69258504E-05
0.4	0.0	.00028381678	-.000321290585	.00085742526	-.00125112059	.00222202346	-.00350982038
0.4	0.5	5.95457596E-05	3.10517426E-05	8.18210584E-05	4.17878108E-05	7.49216123E-05	3.20756294E-05
0.4	1.0	1.73025349E-05	2.68002386E-05	3.72787257E-05	4.24146522E-05	4.52699282E-05	4.47170232E-05
0.2	0.0	.000179532034	-.000154891221	.000493208144	-.00066481917	.0012891238	-.00201744976
0.2	0.5	3.88092673E-05	2.72091805E-05	6.11630232E-05	3.75408522E-05	6.30259464E-05	3.42523708E-05
0.2	1.0	1.14936167E-05	1.93171026E-05	2.83962037E-05	3.35527207E-05	3.72330216E-05	3.83276157E-05
0.0	0.0	9.51120353E-05	-6.93197882E-05	.000254673693	-.000318821724	.000660552769	-.0010122032
0.0	0.5	2.1177613E-05	1.64290641E-05	3.6276253E-05	2.44400256E-05	4.05063144E-05	2.45502426E-05
0.0	1.0	6.39053891E-06	1.10355978E-05	1.6780207E-05	2.0390407E-05	2.33497084E-05	2.47684216E-05

Examining Table 1 and Table II closely, it is clear that $t = 0$ does not have good convergence so we need to look at higher t values. In this case all terms of the difference series become negative for $\Delta = 1$, where as all terms are positive for $\Delta \leq 0.6$ for both spin-one and spin-3/2. In other words, for $\Delta = 1$ the energy is lower for the RT3 phase whereas for $\Delta \leq 0.6$ the energy is lowered for the Q0 phase. For both spin values $\Delta = 0.8$ is at the boundary between the two phases as all terms in the difference series do not have the same sign. However, adding up all the terms shows that the energy difference is still negative for both $S = 1$ and $S = 3/2$. This implies that

$\Delta = 0.8$ is still in the RT3 phase. This suggests a phase diagram in the $\Delta - S$ plane which runs roughly at a constant Δ separating the two phases with a critical Δ value a little below 0.8. This is in remarkably good agreement with the non-linear spin wave calculation of Chernyshev and Zhitomirsky⁶, who find that the phase boundary occurs at $\Delta_c \approx 0.72$. The coupled cluster calculation of Gotze and Richter find a much larger extent of the RT3 phase. For spin-one they find that Q0 phase exists only for Δ less than about 0.3, while for $S = 3/2$ they find that the Q0 phase only exists for Δ less than about 0.5. Clearly the series expansion results are much closer to

TABLE II: Difference between ground state energies of RT3 and Q0 phases for $S = 3/2$ model.

Δ	t	$n = 7$	$n = 8$	$n = 9$	$n = 10$
1.0	0.0	-.000114472141	-.00121771703	.00246072529	-.00806875991
1.0	0.5	-5.29672613E-05	-.000398665752	.000174231254	-.00110052262
1.0	1.0	-2.56833861E-05	-.000162609093	-9.51666575E-05	-.000300829022
0.8	0.0	.000180186675	-.000626753577	.0012997734	-.00278227259
0.8	0.5	5.97067161E-05	-.00012385424	.000137564896	-.000258061125
0.8	1.0	2.339967E-05	-2.44231392E-05	6.79724913E-06	-3.93092026E-05
0.6	0.0	.000238660164	-.000315525754	.000746629417	-.0011861483
0.6	0.5	8.41645616E-05	-1.25172465E-05	.000119584595	-3.55451801E-05
0.6	1.0	3.46495049E-05	2.42397017E-05	4.47287003E-05	3.3561785E-05
0.4	0.0	.000197086688	-.00015596158	.000454414143	-.000547182914
0.4	0.5	7.0831025E-05	2.0197586E-05	9.64105041E-05	1.41819151E-05
0.4	1.0	2.95715969E-05	3.15245715E-05	4.76339757E-05	4.39917413E-05
0.2	0.0	.000130919478	-7.51355574E-05	.000273391632	-.000286281148
0.2	0.5	4.76753252E-05	2.16031853E-05	6.80384984E-05	2.10691061E-05
0.2	1.0	2.00976062E-05	2.41489131E-05	3.61438392E-05	3.53424359E-05
0.0	0.0	7.27922323E-05	-3.39489158E-05	.000149188932	-.000138119912
0.0	0.5	2.68531517E-05	1.42218295E-05	4.05437452E-05	1.64788846E-05
0.0	1.0	1.14277712E-05	1.43851135E-05	2.19416679E-05	2.23838979E-05

the non-linear spin-wave theory.

The ground state energies, estimated by the use of Padé approximants, are shown in Table-3 and Table-4. In general, the energy difference between the two ordered phases is very small. The results are consistent with simply examining the series term by term. The ground state energy is lower in the RT3 phase for $\Delta = 1.0$ and 0.8 , and it is lower in the Q0 phase for $\Delta \leq 0.6$.

The sublattice magnetization series is analyzed by first using a change of variables¹⁴ to remove the square-root singularity caused by spin-waves and then using Padé approximants. Plots of the sublattice magnetization for the phase with the lowest energy are shown in Fig. 1. The results from linear spin-wave theory are also shown^{6,15}. For the XY model, our results of $M/S = 0.86$ for spin-one and $M/S = 0.94$ for spin-3/2 are in excellent agreement with the results of the coupled cluster calculations⁷. For the spin-one Heisenberg model our results suggest a vanishing sublattice magnetization or an absence of the magnetically ordered phase. For the spin-3/2 Heisenberg model, we find non-zero long range order. Our estimate for the sublattice magnetization of $M = 0.14 \pm 0.03$ is in good agreement with earlier coupled cluster results of 0.112 by Gotze et al¹⁶. This provides strong support for the existence of long-range order for the spin-3/2 Heisenberg model.

For the spin-one Heisenberg model, several candidate ground state phases have been proposed¹⁷. Recent exact diagonalization and density matrix renormalization group (DMRG) studies by Changlani and Lauchli¹⁸ presented strong evidence for a spontaneously trimerized phase in the model. These results were further supported by Numerical Linked Cluster expansions done by Ixert, Tischler and Schmidt¹⁹. Motivated by these studies, we calculate the ground state energy of the trimerized phase by series expansions.

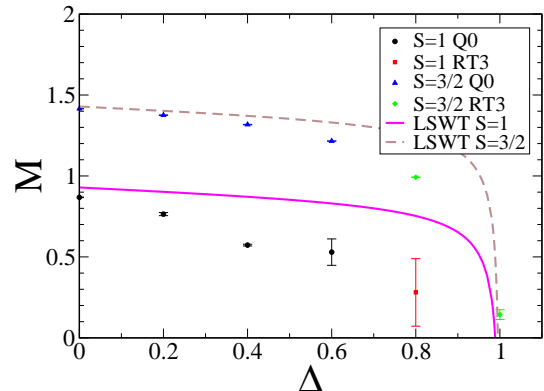


FIG. 1: Sublattice magnetization for the spin-one and spin-3/2 XXZ Kagome antiferromagnets as a function of the XY anisotropy parameter Δ . $\Delta = 1$ corresponds to the Heisenberg model, where the results for the spin-one model is consistent with a vanishing order parameter. Also shown are results from linear spin-wave theory^{6,15}.

To carry out the expansion for the trimerized phase of the spin-one model, we consider all bonds in up pointing triangles to have exchange constant of unity, where as all bonds in down pointing triangles have exchange constant of α . At $\alpha = 0$, this system breaks into disconnected triangles. For spin $S = 1$, each triangle of spins has a unique ground state. Series expansions can be calculated for ground state properties in powers of α by non-degenerate perturbation theory^{10,11}. The ground state energy per site, e_0 , has a series expansion

$$3e_0 = -3 - 2\alpha^2 + \frac{2}{3}\alpha^3 + \frac{11}{18}\alpha^4 - 0.33757716\alpha^5 - 0.36266528\alpha^6 - 0.75868273\alpha^7 + \dots \quad (6)$$

TABLE III: Ground state energy for $S = 1$ model. The mean value of the Padé estimates for the ground state energy and the spread in the values of the different approximants are shown

Phase	Δ	t	mean	spread
RT3	1.0	0.0	-1.3950	.0013
RT3	1.0	0.5	-1.3928	.00014
RT3	1.0	1.0	-1.3910	.00002
Q0	1.0	0.0	-1.3903	.0006
Q0	1.0	0.5	-1.3890	.0004
Q0	1.0	1.0	-1.3877	.00012
RT3	0.8	0.0	-1.3033	.0002
RT3	0.8	0.5	-1.3019	.0002
RT3	0.8	1.0	-1.3012	.0005
Q0	0.8	0.0	-1.3016	.0001
Q0	0.8	0.5	-1.3001	.00003
Q0	0.8	1.0	-1.2992	.00005
RT3	0.6	0.0	-1.2215	.0003
RT3	0.6	0.5	-1.2214	.0003
RT3	0.6	1.0	-1.2208	.0006
Q0	0.6	0.0	-1.2221	.0002
Q0	0.6	0.5	-1.2213	.0002
Q0	0.6	1.0	-1.2206	.0002
RT3	0.4	0.0	-1.1534	.00009
RT3	0.4	0.5	-1.1535	.0003
RT3	0.4	1.0	-1.1531	.00011
Q0	0.4	0.0	-1.1544	.0002
Q0	0.4	0.5	-1.1541	.0002
Q0	0.4	1.0	-1.1536	.0002
RT3	0.2	0.0	-1.0987	.00013
RT3	0.2	0.5	-1.0989	.00012
RT3	0.2	1.0	-1.0990	.0003
Q0	0.2	0.0	-1.0995	.00014
Q0	0.2	0.5	-1.0995	.00007
Q0	0.2	1.0	-1.0996	.00008
RT3	0.0	0.0	-1.0563	.00003
RT3	0.0	0.5	-1.0562	.00009
RT3	0.0	1.0	-1.0563	.0001
Q0	0.0	0.0	-1.0568	.00002
Q0	0.0	0.5	-1.0568	.00008
Q0	0.0	1.0	-1.0568	.00009

We use Dlog Padé approximants to estimate the sum of the series. The $[2/4]$, $[1/5]$, $[3/3]$ and $[2/3]$ approximants give -4.1555 , -4.3801 , -4.1391 , -4.1236 , respectively. Upon averaging, this give an energy per site of $e_0 = -1.40$, which is indeed lower than our estimate for the energy of the ordered phases. This supports the results by Changlani and Lauchli¹⁸ that the spin-one Heisenberg model has a spontaneously trimerized ground state.

We have also developed series expansions for the trimerized phase of the spin-one model with $\Delta < 1$.²⁰ We find that the energy of the trimerized phase becomes even more stabilized with anisotropy and its ground state energy is always lower than the energy of the magnetically ordered state until the series begin to diverge below $\Delta \approx 0.2$. This suggests that the trimerized phase may extend well beyond just the Heisenberg model in the spin-one case.

TABLE IV: Ground state energy for $S = 3/2$ model. The mean value of the Padé estimates for the ground state energy and the spread in the values of the different approximants are shown

Phase	Δ	t	mean	spread
RT3	1.0	0.0	-2.8193	.004
RT3	1.0	0.5	-2.8250	.004
RT3	1.0	1.0	-2.8175	.005
Q0	1.0	0.0	-2.8185	.004
Q0	1.0	0.5	-2.8229	.003
Q0	1.0	1.0	-2.8162	.004
RT3	0.8	0.0	-2.6661	.002
RT3	0.8	0.5	-2.6674	.002
RT3	0.8	1.0	-2.6664	.0008
Q0	0.8	0.0	-2.6660	.002
Q0	0.8	0.5	-2.6671	.002
Q0	0.8	1.0	-2.6662	.0006
RT3	0.6	0.0	-2.5481	.0005
RT3	0.6	0.5	-2.5476	.0007
RT3	0.6	1.0	-2.5469	.0002
Q0	0.6	0.0	-2.5485	.0004
Q0	0.6	0.5	-2.5481	.0006
Q0	0.6	1.0	-2.5475	.00005
RT3	0.4	0.0	-2.4559	.0001
RT3	0.4	0.5	-2.4553	.0004
RT3	0.4	1.0	-2.4548	.00012
Q0	0.4	0.0	-2.4564	.00011
Q0	0.4	0.5	-2.4563	.0006
Q0	0.4	1.0	-2.4552	.0002
RT3	0.2	0.0	-2.3830	.00012
RT3	0.2	0.5	-2.3834	.0006
RT3	0.2	1.0	-2.3826	.00006
Q0	0.2	0.0	-2.3834	.00014
Q0	0.2	0.5	-2.3838	.0006
Q0	0.2	1.0	-2.3830	.00009
RT3	0.0	0.0	-2.3261	.00003
RT3	0.0	0.5	-2.3261	.0002
RT3	0.0	1.0	-2.3261	.00003
Q0	0.0	0.0	-2.3264	.00004
Q0	0.0	0.5	-2.3266	.0003
Q0	0.0	1.0	-2.3264	.00002

In conclusion, in this paper we have studied the competing ground state phases of spin-one and spin-3/2 Kagome Lattice antiferromagnets with XY anisotropy. We find that near the XY limit the $q = 0$ magnetically ordered phase is obtained, whereas near the Heisenberg model the $\sqrt{3} \times \sqrt{3}$ phase is realized. Our phase diagrams are in remarkably good agreement with spin-wave theory. The degeneracy between the ground state energy of the two states is lifted only in 7th order of perturbation theory, which can be understood in terms of the mechanism discussed by Chernyshev and Zhitomirsky⁶, requiring creation, propagation and annihilation of virtual excitations around closed loops. For the spin-one Heisenberg model, the ground state is not magnetically ordered. We presented evidence that in this case the ground state is spontaneously trimerized. For spin-3/2 Heisenberg model, our estimate for long-range order is in good agreement with earlier coupled-cluster results of

Gotze et al¹⁶, providing strong support to the existence of an ordered phase in this model.

Acknowledgments

We thank A. Chernyshev, M. Zhitomirsky, J. Richter and Z. Zhu for many valuable communications and discussions. This work is supported in part by NSF grant number DMR-1306048, and by the computing resources provided by the Australian (APAC) National facility.

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