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

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(Dated: October 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 $\Delta$, 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 $\Delta$. 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 order [3]. However, the more general XXZ model for larger spin and with XY anisotropy may well have long-range magnetic order [4]. Indeed, several experimental Kagome systems with large spin are known to have magnetic long-range order [5].

Competing magnetic orders in these models were investigated recently by Chernyshev and Zithomirsky [6] using non-linear spin-wave 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 $\Delta$ values. The models have also been studied recently using the coupled cluster method by Gotze and Richter [7], 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

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

where the sum is over the nearest neighbor pairs and $\Delta$ is the anisotropy parameter. We will study spin-one and spin-3/2 model with various $\Delta$ 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. 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 [8, 9]. 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

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

and

$$
H_1 = -\frac{1}{4}(1 + 2\Delta) \sum_{<ij>} (S_i^+ S_j^- + S_i^- S_j^+) \quad (4)
$$

$$
+ \frac{\Delta}{4} \sum_{<ij>} \eta_{ij} \left( S_i^+ S_j^- - S_i^- S_j^+ \right)
$$

$$
+ \frac{\Delta}{4} \sum_{<ij>} \eta_{ij} \left( S_i^+ S_j^- - S_i^- S_j^+ \right)
$$

$$
+ \frac{\Delta}{4} \sum_{<ij>} \eta_{ij} \left( S_i^+ S_j^- - S_i^- S_j^+ \right)
$$
with
\[ \eta_{ij} = \begin{cases} +1 & A \to B, B \to C, C \to A \\ -1 & B \to A, C \to B, A \to C \end{cases} \] (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 \( \Delta \).

Our interesting finding is that regardless of spin, anisotropy \( \Delta \) 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 orders\(^{10}\) and the high-temperature order-parameter susceptibility degeneracy for the XY pyrochlore antiferromagnets\(^{11}\). 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.

| \( \Delta \) | \( t \) | \( n = 7 \) | \( n = 8 \) | \( n = 9 \) | \( n = 10 \) | \( n = 11 \) | \( n = 12 \) |
|---|---|---|---|---|---|---|---|
| 1.0 0.0 | 1.79281056E-05 | -0.0228441779 | 0.0485266477 | -0.0131501048 | 0.0244393622 | -0.048490238 |
| 1.0 0.5 | -1.99373138E-05 | -0.0038998796 | -0.05357305E-05 | -0.005541912718 | -0.00190168854 | -0.00584502974 |
| 1.0 1.0 | -1.0049807E-05 | -0.96129929E-05 | -0.00130394373 | -0.000177890555 | -0.00217321242 | -0.0002516233 |
| 0.8 0.0 | 0.00337078044 | -0.0123165168 | 0.0025684538 | -0.0062066228 | 0.0082990621 | -0.0138679947 |
| 0.8 0.5 | 0.60676498E-05 | -0.81433973E-05 | 4.17426967E-05 | -0.00100993463 | -0.91047816E-05 | -0.0003845195 |
| 0.8 1.0 | 1.58188574E-05 | -5.15123324E-05 | -1.36613326E-05 | -2.32793478E-05 | -5.7282811E-05 | -5.02830945E-05 |
| 0.6 0.0 | 0.00368046153 | -0.00640326014 | 0.0145292023 | -0.0023896797 | 0.0038375524 | -0.0061563131 |
| 0.6 0.5 | 0.74772154E-05 | 4.96763165E-06 | 8.15631813E-05 | 1.48577108E-05 | 5.09012958E-05 | -5.9802479E-06 |
| 0.6 1.0 | 2.09523323E-05 | 2.48510186E-05 | 3.09097891E-05 | 3.26457591E-05 | 3.16691472E-05 | 2.69268504E-05 |
| 0.4 0.0 | 0.0038381678 | -0.00321290585 | 0.0085742526 | -0.00125111059 | 0.00223520346 | -0.0050982038 |
| 0.4 0.5 | 5.95457596E-05 | 3.10517426E-05 | 8.18210585E-05 | 4.17878108E-05 | 7.4921612E-05 | 3.20756294E-05 |
| 0.4 1.0 | 1.73025349E-05 | 2.68002386E-05 | 3.72782575E-05 | 4.2146522E-05 | 4.56299282E-05 | 4.47107232E-05 |
| 0.2 0.0 | 0.00179532034 | -0.00154691221 | 0.000493208144 | -0.0006481917 | 0.0012891238 | -0.0020174976 |
| 0.2 0.5 | 3.88092673E-05 | 2.72901805E-05 | 6.11630223E-05 | 3.75408522E-05 | 6.30259464E-05 | 3.42537024E-05 |
| 0.2 1.0 | 1.14936167E-05 | 1.93170126E-05 | 2.83962037E-05 | 3.35527207E-05 | 3.72393212E-05 | 3.83276157E-05 |
| 0.0 0.0 | 9.51120335E-05 | -6.93169878E-05 | 0.00254673693 | -0.000318821724 | 0.00660552769 | -0.001122032 |
| 0.0 0.5 | 2.1177673E-05 | 1.64290641E-05 | 3.6276235E-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 I 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 \( \Delta \) separating the two phases with a critical \( \Delta \) value a little below 0.8. This is in remarkably good agreement with the non-linear spin wave calculation of Chernyshev and Zhilomirsky\(^{8}\), 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 \( \Delta \) less than about 0.3, while for \( S = 3/2 \) they find that the Q0 phase only exists for \( \Delta \) less than about 0.5. Clearly the series expansion results are much closer to 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 variables\(^{12}\) to remove the square-root singularity caused by spin-waves and then using Padé approximants. Plots of the sublattice magnetization for...
the ground state energy of the trimerized phase by series expansions. The ground state energy of the trimerized phase by series expansions. Recent exact diagonalization and density matrix renormalization group (DMRG) studies by Changlani and Lauchli [14] presented strong evidence for a spontaneously trimerized ground state. For the spin-one Heisenberg 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 \( \alpha \). 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 \( \alpha \) by non-degenerate perturbation theory [8, 9]. The ground state energy per site, \( e_0 \), has a series expansion

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

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 \( \alpha \). 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 \( \alpha \) by non-degenerate perturbation theory [8, 9]. The ground state energy per site, \( e_0 \), has a series expansion

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-0.36266528\alpha^6 - 0.75868273\alpha^7 + \ldots
\]

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 Lauchli [14] that the spin-one Heisenberg model has a spontaneously trimerized ground state.

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. 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.

This work is supported in part by NSF grant number

\[
\begin{array}{cccc}
\Delta & t & n = 7 & n = 8 & n = 9 & n = 10 \\
1.0 & 0.0 & -0.00114472141 & -0.00121771703 & -0.00246072529 & -0.0080675901 \\
1.0 & 0.5 & -5.29672613E-05 & -0.00398665752 & -0.00174231254 & -0.0011005262 \\
1.0 & 1.0 & -2.56383618E-05 & -0.001626090993 & -9.51666575E-05 & -0.00300829022 \\
0.8 & 0.0 & -0.00218867675 & -0.00626753577 & -0.0012997734 & -0.0027822759 \\
0.8 & 0.5 & -5.97067161E-05 & -0.0012385424 & -0.00137564896 & -0.0025806125 \\
0.8 & 1.0 & -2.339967E-05 & -2.42431392E-05 & 6.79724913E-06 & -3.93092026E-05 \\
0.6 & 0.0 & -0.002386360164 & -0.00315525754 & -0.00146629417 & -0.0011861438 \\
0.6 & 0.5 & 8.41645616E-05 & -1.25172465E-05 & -0.00119584595 & -3.5541801E-05 \\
0.6 & 1.0 & 3.4695049E-05 & 2.42390717E-05 & 4.7287003E-05 & 3.3561785E-05 \\
0.4 & 0.0 & -0.00197086688 & -0.0015596158 & 0.00045414143 & -0.000547182014 \\
0.4 & 0.5 & 7.0831025E-05 & 2.0197586E-05 & 9.64105041E-05 & 1.4819151E-05 \\
0.4 & 1.0 & 2.95715096E-05 & 3.15245715E-05 & 4.76339757E-05 & 3.3561785E-05 \\
0.2 & 0.0 & -0.00130919478 & -7.51355574E-05 & 0.000273391632 & -0.00286281148 \\
0.2 & 0.5 & 4.76753252E-05 & 2.16031853E-05 & 6.80349894E-05 & 2.10691061E-05 \\
0.2 & 1.0 & 2.00070602E-05 & 2.41849131E-05 & 3.61408392E-05 & 3.5342359E-05 \\
0.0 & 0.0 & 7.27922323E-05 & -3.94891518E-05 & 0.00149188932 & -0.001381199012 \\
0.0 & 0.5 & 2.68531517E-05 & 1.42218295E-05 & 4.05437452E-05 & 1.64788846E-05 \\
0.0 & 1.0 & 1.14277712E-05 & 1.43851135E-05 & 2.19416679E-05 & 2.2383979E-05 \\
\end{array}
\]

FIG. 1: Sublattice magnetization for the spin-one and spin-
3/2 XXZ Kagome antiferromagnets as a function of the XY
anisotropy parameter \( \Delta \). \( \Delta = 1 \) corresponds to the Heisenberg model, where the results for the spin-one model is consistent with a vanishing order parameter.
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 | ∆t | mean | spread |
|-------|-----|------|--------|
| RT3   | 1.0 | -1.3950 | .0013  |
| RT3   | 1.0 | -1.3928 | .0014  |
| RT3   | 1.0 | -1.3910 | .0002  |
| Q0    | 1.0 | -1.3903 | .0006  |
| Q0    | 1.0 | -1.3890 | .0004  |
| Q0    | 1.0 | -1.3877 | .0012  |
| RT3   | 0.8 | -1.3033 | .0002  |
| RT3   | 0.8 | -1.3014 | .00014 |
| RT3   | 0.8 | -1.3010 | .00002 |
| Q0    | 1.0 | -1.3001 | .0003  |
| Q0    | 0.5 | -1.3016 | .0001  |
| Q0    | 1.0 | -1.3001 | .0003  |
| RT3   | 0.6 | -1.2215 | .0003  |
| RT3   | 0.6 | -1.2214 | .0003  |
| RT3   | 0.6 | -1.2208 | .0006  |
| Q0    | 0.6 | -1.2221 | .0002  |
| Q0    | 0.6 | -1.2213 | .0002  |
| Q0    | 0.6 | -1.2206 | .0002  |
| RT3   | 0.4 | -1.1534 | .0009  |
| RT3   | 0.4 | -1.1535 | .0003  |
| RT3   | 0.4 | -1.1531 | .0011  |
| Q0    | 0.4 | -1.1544 | .0002  |
| Q0    | 0.4 | -1.1541 | .0002  |
| Q0    | 0.4 | -1.1536 | .0002  |
| RT3   | 0.2 | -1.0987 | .0013  |
| RT3   | 0.2 | -1.0989 | .0012  |
| RT3   | 0.2 | -1.0990 | .0003  |
| Q0    | 0.2 | -1.0995 | .0014  |
| Q0    | 0.2 | -1.0995 | .0007  |
| Q0    | 0.2 | -1.0996 | .0008  |
| RT3   | 0.0 | -1.0563 | .0003  |
| RT3   | 0.0 | -1.0562 | .0009  |
| RT3   | 0.0 | -1.0563 | .0001  |
| Q0    | 0.0 | -1.0568 | .0002  |
| Q0    | 0.0 | -1.0568 | .0008  |
| Q0    | 0.0 | -1.0568 | .0009  |

DMR-1306048, and by the computing resources provided by the Australian (APAC) National facility.

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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 | ∆t | mean | spread |
|-------|-----|------|--------|
| RT3   | 1.0 | -2.8193 | .004   |
| RT3   | 1.0 | -2.8250 | .004   |
| RT3   | 1.0 | -2.8175 | .005   |
| Q0    | 1.0 | -2.8185 | .004   |
| Q0    | 1.0 | -2.8229 | .003   |
| Q0    | 1.0 | -2.8162 | .004   |
| RT3   | 0.8 | -2.6661 | .002   |
| RT3   | 0.8 | -2.6674 | .002   |
| RT3   | 0.8 | -2.6664 | .0008  |
| Q0    | 0.8 | -2.6660 | .002   |
| Q0    | 0.8 | -2.6671 | .002   |
| Q0    | 0.8 | -2.6662 | .0006  |
| RT3   | 0.6 | -2.5481 | .0005  |
| RT3   | 0.6 | -2.5476 | .0007  |
| RT3   | 0.6 | -2.5469 | .0002  |
| Q0    | 0.6 | -2.5485 | .0004  |
| Q0    | 0.6 | -2.5481 | .0006  |
| Q0    | 0.6 | -2.5475 | .0005  |
| RT3   | 0.4 | -2.4559 | .0001  |
| RT3   | 0.4 | -2.4553 | .0004  |
| RT3   | 0.4 | -2.4548 | .0012  |
| Q0    | 0.4 | -2.4564 | .0011  |
| Q0    | 0.4 | -2.4563 | .0006  |
| Q0    | 0.4 | -2.4552 | .0002  |
| RT3   | 0.2 | -2.3830 | .0012  |
| RT3   | 0.2 | -2.3834 | .0006  |
| RT3   | 0.2 | -2.3826 | .0006  |
| Q0    | 0.2 | -2.3834 | .0014  |
| Q0    | 0.2 | -2.3838 | .0006  |
| Q0    | 0.2 | -2.3830 | .0009  |
| RT3   | 0.0 | -2.3261 | .00003 |
| RT3   | 0.0 | -2.3261 | .0002  |
| RT3   | 0.0 | -2.3261 | .00003 |
| Q0    | 0.0 | -2.3264 | .0003  |
| Q0    | 0.0 | -2.3266 | .0008  |
| Q0    | 0.0 | -2.3264 | .00002 |

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