Three-Sublattice Order in Triangular- and Kagomé-Lattice Spin-Half Antiferromagnets

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We study the possibility of $\sqrt{3}\times\sqrt{3}$ antiferromagnetic order in the $S=\frac{1}{2}$ triangular- and Kagomé-lattice Heisenberg models. An Ising-like anisotropy is introduced into the Hamiltonian, which picks a pair of ground states out of the manifold of the classically ordered states. To study properties of the Heisenberg model, we develop series expansions around one ordered state. We find that the Kagomé-lattice model is disordered whereas the triangular-lattice model is very close to the critical point for antiferromagnetism; if ordered, the latter has an order parameter much smaller than that predicted by spin-wave theory.

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The magnetic properties of two-dimensional frustrated quantum-spin Hamiltonians have been of considerable interest for a long time. Early work goes back to Anderson and co-workers [1], who proposed that the ground state of the spin-half triangular antiferromagnet should have no long-range order. They proposed a special type of ground state due to strong quantum fluctuations which was called a resonating valence bond state. Several numerical studies including exact diagonalizations [2] were interpreted in favor of this picture. Another notable work is the proposed mapping by Kalmeyer and Laughlin [3] between the triangular-lattice Heisenberg model and a two-dimensional electron gas in a strong magnetic field. On that basis they suggested a disordered ground state with an excitation gap. Huse and Elser [4] carried out a variational wave-function study of the triangular antiferromagnet. They found that they could construct wave functions which had energy substantially lower than that of Kalmeyer and Laughlin. Within their variational scheme the ground-state energy was minimized by a wave function that had a large (about 68% of the classical value) antiferromagnetic order parameter.

One technique for studying magnetic order in the presence of quantum fluctuations is the spin-wave or large-S expansion [5]. Recent numerical studies [6] of the square-lattice Heisenberg model found that the spin-wave expansion was quantitatively accurate even for $S=\frac{1}{2}$. For example, the estimated sublattice magnetization and spin-wave velocity agree with the spin-wave answer truncated at order $1/S$ [7]. Such $1/S$ expansions have also been developed [8] for the triangular lattice, and predict an order parameter about 48% of the classical value.

Another approach that has provided a lot of insight into the magnetically ordered and disordered phases of these systems is the large-N expansion of Read and Sachdev [9]. For the square lattice, they predicted the existence of Néel and dimer ordered phases. Numerical evidence for such phases was also obtained by series expansions [10] and by exact diagonalization studies [11]. For the triangular and Kagomé lattices, where the classical order is noncollinear, these large-N expansions predict a nondimerized quantum disordered ("spin liquid") phase for sufficiently small S. However, this theory cannot predict accurately the value of the spin where the transition from magnetic order to disorder takes place.

The Kagomé-lattice antiferromagnet has generated tremendous interest lately from both an experimental [12,13] and a theoretical point of view [14–18]. A simple way to visualize the Kagomé lattice is to regard the triangular lattice as consisting of four sublattices and remove the spins on one of the sublattices. As shown, e.g., by Broholm *et al.* [12], the classical ground state is highly degenerate in this case, with nonzero entropy of the degeneracy per spin. However, the entropy of classical zero-point fluctuations gives different relative Boltzmann weights to the various ground states such that the classical Heisenberg model on the Kagomé lattice apparently has true long-range antiferromagnetic order in the limit of vanishing temperature [18]. Thus the question naturally arises as to whether this order survives the strong quantum fluctuations of the spin-half system. The spin correlations found in exact ground states of small clusters suggest that the spin-half system does not have magnetic order [19], but a more systematic approach to address this question is clearly called for.

In this Letter we study the spin-half models by series-expansion methods. We introduce an Ising-type anisotropy, which allows us to develop systematic series expansions for the ground-state properties. The series are extrapolated by standard methods. The results of the extrapolations indicate that the Kagomé antiferromagnet is not antiferromagnetically ordered. Thus this system may well have a true spin-liquid ground state. For the triangular antiferromagnet, on the other hand, we can only conclude that it is very close to its critical point. If long-range ordered, it has an order parameter much smaller than given by spin-wave theory; if disordered it has a large correlation length. Given the length of our series we cannot resolve whether or not it is ordered. However, we do obtain a significantly improved estimate of the

ground-state energy.

The spin-half Heisenberg models are defined by the Hamiltonian

$$\mathcal{H} = 4\sum_{\langle i,j\rangle} \mathbf{S}_i \cdot \mathbf{S}_j , \tag{1}$$

where the sum is over all nearest-neighbor pairs of spins. In order to develop Ising-type expansions around a classical ground state, which is noncollinear, we need to introduce an anisotropy parameter in the Hamiltonian that selects a pair of ground states out of the classical ordered states. For the triangular lattice and for the Kagomé lattice, this corresponds to the three-sublattice 120° order (also called $\sqrt{3}\times\sqrt{3}$ order). Let us choose a ground state with the spins on the A sublattice pointing along the z axis, and the spins on the B and C sublattices rotated 120° either way in the x-z plane from the z axis. We choose new variables $\boldsymbol{\sigma}_i = 2\mathbf{S}_i$ on A, $2R_+\mathbf{S}_i$ on B, and $2R_-\mathbf{S}_i$ on C, where the operator $R_\pm$ is a rotation around the y axis by $\pm 120°$. In these new variables our reference classical ground state has all the $\boldsymbol{\sigma}_i$ pointing along the z axis (ferromagnetic order). The transformed Hamiltonian is

$$\mathcal{H} = \sum_{i\rightarrow j}\left[-\frac{1}{2}(\sigma_i^z\sigma_j^z+\sigma_i^x\sigma_j^x) + \frac{\sqrt{3}}{2}(\sigma_i^z\sigma_j^x-\sigma_i^x\sigma_j^z)+\sigma_i^y\sigma_j^y\right]. \tag{2}$$

Here $i\rightarrow j$ goes from sublattice A to B, B to C, and C to A. Taking as the unperturbed Hamiltonian

$$\mathcal{H}_0 = -\tfrac{1}{2}\sum_{i\rightarrow j}\sigma_i^z\sigma_j^z , \tag{3}$$

and writing the full Hamiltonian as

$$\mathcal{H}' = \mathcal{H}_0 + J_\perp(\mathcal{H}-\mathcal{H}_0) , \tag{4}$$

we can generate power-series expansions in the variable $J_\perp$. The Heisenberg antiferromagnet corresponds to $J_\perp = 1$.

We generate the expansions by the cluster method [20,21]. As in a low-temperature expansion, we define our clusters as sets of sites. A cluster of N sites only contributes in Nth or higher order. To calculate the contribution of a given cluster, we include all terms in the Hamiltonian which flip spins only in the cluster. All clusters which are related to each other by a symmetry of the lattice can be grouped together; because of the directionality of the bonds in (2) we cannot group clusters together by their topology. For the triangular lattice we find 182510 clusters in eleventh order. For the Kagomé lattice we consider the decomposition of the triangular lattice into four sublattices and keep only the clusters which do not fall on all four sublattices. The lattice constants of the clusters for the Kagomé lattice can be obtained from the triangular ones. Straight, linear clusters have their lattice constant reduced to $\frac{2}{3}$ of the triangular value while all other permitted clusters have it reduced to $\frac{1}{3}$. In twelfth order we find only 26079 clusters for the Kagomé lattice. This allows us to carry out the expansions for the Kagomé lattice to higher order than for the triangular case [22]. Let us define the order parameter M and the structure factor S by the relations

$$M = \langle\sigma_0^z\rangle, \quad S = \sum_i (\langle\sigma_0^z\sigma_i^z\rangle - \langle\sigma_0^z\rangle\langle\sigma_i^z\rangle) , \tag{5}$$

where the brackets denote ground-state expectation values. The expansion coefficients are presented in Table I.

We first consider the extrapolation of the order-parameter series assuming long-range order at the Heisenberg point. In this case, there will be a square-root singularity at $J_\perp = 1$. If there were no other singularities in the complex plane within the unit circle, the partial sum of the first n terms in the series should asymptotically converge as $1/\sqrt{n}$, allowing for a systematic estimate of the sum of the infinite series [6]. However, this is not true if there are other singularities in the complex plane with $|J_\perp| < 1$. From the Hamiltonian, we expect an additional singularity on the negative real axis near $J_\perp = -0.5$, where the system may order ferromagnetically along the y axis. Such a singularity is clearly present, causing the partial sums to alternate and diverge before $J_\perp = 1$. To move this singularity away, we use Euler transforms to change variables from $J_\perp$ to $x = J_\perp/[a+(1-a)J_\perp]$. For very small a the series coefficients become extremely small, making extrapolations harder. For a close to unity, the coefficients are hardly changed from the untransformed series. Good compromises appear to be $a = \frac{1}{2}$ and $a = \frac{1}{3}$, which map $J_\perp = -1$ and $J_\perp = -\frac{1}{2}$ to infinity, respectively. The resulting plots of the partial sums are shown in Fig. 1. The Kagomé antiferromagnet is clearly extrapolating to a negative value

TABLE I. Expansion coefficients for the ground-state energy E_0, the order parameter M, and the structure factor S.

		Triangular			Kagome	
m	E_0	M	S	E_0	M	S
0	-1.5	1	0	-1	1	0
1	0	0	0	0	0	0
2	-0.675	-0.27	1.08	-0.75	-0.5	2
3	0.135	0.108	-0.432	0.125	0.1666667	-0.6666667
4	-0.1773482	-0.2726916	1.4341347	-0.08020833	-0.3245833	1.7695833
5	0.0817034	0.1717951	-0.9583433	0.02412326	0.1749051	-1.0925162
6	-0.1133163	-0.3315263	2.0593548	-0.02719836	-0.2906697	1.9308204
7	0.1261394	0.4110737	-2.7696352	0.06731218	0.4679179	-3.6055475
8	-0.1906393	-0.7382203	5.2916370	-0.1751062	-1.1833128	8.9988224
9	0.2740349	1.1781303	-8.9566042	0.1891826	1.5674260	-12.9259803
10	-0.4101784	-2.0109889	16.0740098	-0.2419287	-2.5738747	21.7897165
11	0.6262086	3.4012839	-28.4769501	0.3815669	4.3229068	-39.2884503
12				-0.6701061	-8.0770009	76.5639246

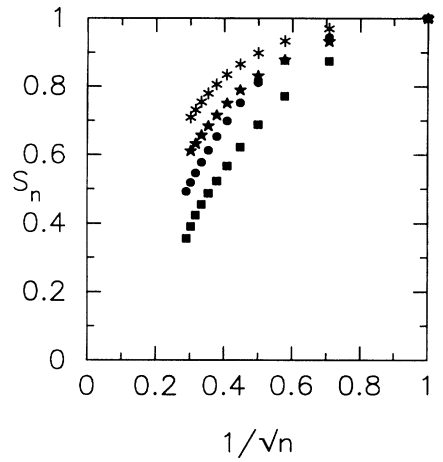

FIG. 1. Partial sums for the order-parameter series. The five- and six-point stars are for the triangular lattice with Euler transform parameters $a=\frac{1}{2}$ and $a=\frac{1}{3}$, respectively. The squares and the circles are for the Kagomé lattice with $a=\frac{1}{2}$ and $a=\frac{1}{3}$, respectively.

suggesting that it is disordered, while the triangular-lattice model appears to extrapolate to a value of about 20% of the classical moment. This should be compared with the spin-wave estimate of 48%.

Since the order-parameter estimate for the triangular lattice is so small, the analysis assuming long-range order cannot be fully relied upon. Unfortunately, unbiased analysis shows poor convergence. Hence we look for consistency with different scenarios. If the system disorders before reaching the Heisenberg point, we expect the critical point to be in the classical 3D Ising universality class. On the other hand, if it disorders right at the Heisenberg point we expect it to have the universality of the classical Heisenberg antiferromagnet on the stacked triangular lattice [23]. In the first case we expect to see the magnetization vanish at the critical point with the exponent $\beta_I \approx \frac{1}{3}$, while in the latter case it should be $\beta_{HI}=\beta_H/\phi_I$, where β_H is the exponent for the stacked triangular Heisenberg model for disordering with temperature, and ϕ_I is the crossover exponent for Ising anisotropy. Kawamura quotes [23] $\beta_H \approx 0.3$, $\phi_I < \gamma \approx 1.1$. Alternative scenarios for the stacked triangular lattice give similar estimates [24]. Thus β_{HI} is not far from $\frac{1}{3}$. To investigate these different possibilities we raise the magnetization series to different powers, such that the resulting series may have a simple zero. We then use Padé extrapolations to estimate the location of the zero. These estimates of the zeros of M^3 are shown in Table II. The closeness of the estimates to Table I shows just how near the Heisenberg system is to the critical point. For the Kagomé lattice we find that all high order ($N+M>8$), near diagonal ($|N-M|<6$) estimates of the zero are in the range 0.83–0.94, showing that it disorders well before the Heisenberg point.

We also studied the order-parameter series by biased

TABLE II. $[N/M]$ Padé estimate for the zero of $(M^{\dagger})^3$ for the triangular lattice. A value of 1 corresponds to the Heisenberg model.

M/N	2	3	4	5	6	7	8
1	1.385	1.205	1.062	1.019	1.015	1.003	1.002
2	1.004	0.955	0.910	1.015	1.019	1.001	1.003
3	0.960	0.827	0.949	1.001	0.997	1.008	1.014
4	0.923	0.954	0.974	0.998	1.000	1.012	
5	1.125	1.005	1.005	1.020	0.978		
6	0.875	1.005	1.005	0.999			
7	1.191	1.016	0.997				
8	0.863	1.037					
9	1.337						

differential approximants. We constructed d-log Padé approximants for the series which were biased to have a power-law singularity at $J_\perp=1$. The approximants can then be used to estimate the exponent and the value of the function at that point. For the triangular lattice, we found that those approximants which gave exponent estimates around $\frac{1}{2}$ gave an order-parameter estimate around 20%, as in our previous analysis. However, an equal number of approximants gave exponent estimates in the range 0.3 to 0.4. These had values very close to zero. For the Kagomé lattice these biased approximants did not give sensible results, as might be expected if the singularity really occurs well before $J_\perp=1$.

As long as the triangular system does not disorder much before $J_\perp=1$, the ground-state energy per spin E_I for the Heisenberg model can be estimated from the series. Since the singularity in the ground-state energy is weak (a $\frac{3}{2}$ power in spin-wave theory), a reasonable estimate can be obtained by direct Padé approximants. Different approximants showed excellent agreement with each other for the triangular lattice. From these we would estimate $E_I=-2.204\pm0.005$, where the uncertainties reflect the spread in the Padé estimates. However, we expect a systematic error in this estimate. If the Heisenberg model is disordered, there is a singularity at $J_\perp<1$, and we cannot reliably predict this systematic error. However, if the singularity is at the Heisenberg point (which will occur for long-range ordered and critical cases), we can also estimate the energies by biased differential approximants. We find that a number of approximants show a very weak singularity (exponent ≥ 3), and they have energy estimates in the range quoted earlier. However, a few approximants which give exponent values close to $\frac{3}{2}$ suggest an energy estimate slightly lower. If the Heisenberg model is critical, we expect the exponent to be larger than $\frac{3}{2}$. Thus our best overall estimate for the ground-state energy of the triangular-lattice Heisenberg model is

$$E_I=-2.21\pm0.01\,. \tag{6}$$

This can be compared with the results from exact diago-

nalizations [2] of $E_t = -2.20 \pm 0.04$. On the other hand, the extrapolations for the Kagomé-lattice energy series are ill-behaved as expected. A similar extrapolation was done for the structure factor to ascertain its location and power of divergence. The convergence was not as good, but the results were consistent with earlier conclusions.

To summarize, in this Letter we have developed systematic Ising-type expansions for the $S = \frac{1}{2}$ Kagomé- and triangular-lattice antiferromagnets. We find that the Kagomé-lattice Heisenberg model is magnetically disordered, while the triangular-lattice Heisenberg model is very close to its critical point: If ordered it has an order parameter much smaller than predicted by spin-wave theory; if disordered it has a large correlation length.

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