Finite Temperature Properties of the Spin-$\frac{1}{2}$ Heisenberg Antiferromagnet on the Triangular Lattice

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We have studied the spin-$\frac{1}{2}$ Heisenberg antiferromagnet on the triangular lattice by high temperature series expansions. From an analysis of the antiferromagnetic structure factor and correlation length, we deduce that the ground state has small but nonzero long range antiferromagnetic order. We also determine the temperature dependence of the uniform susceptibility and the specific heat.

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The behavior of frustrated quantum-spin systems in two dimensions is of considerable interest, particular attention being given to whether quantum fluctuations destroy the long range magnetic order (LRMO) which is generally present in the classical ground state. The simplest frustrated quantum-spin system is probably the spin-$\frac{1}{2}$ Heisenberg antiferromagnet (HAF) on the triangular lattice, which was the original model for Anderson's proposed resonant valence bond state [1]. More recently Kalmeyer and Laughlin (KL) [2] proposed a different ground state for this model, also without long range magnetic order but with chiral symmetry breaking. However, Huse and Elser [3] found variational states with a large antiferromagnetic order parameter, which had significantly lower energy than the KL states, which suggests that the ground state may have LRMO. This possibility is also consistent with spin-wave theory, for which the lowest order approximation [4] predicts the staggered magnetization to be about 48% of the classical value and the next order correction actually increases this a little [5]. Exact diagonalization of small clusters [6, 7] suggested no LRMO, while two recent calculations, which included the cluster with $N = 36$ sites [8, 9], have come to opposite conclusions about the existence of LRMO. Another useful approach is series expansions. One of us (R.R.P.S.) and D. Huse [10] investigated the HAF on the triangular and kagomé lattices by introducing an Ising-type anisotropy into the Hamiltonian. Ground state properties were determined by expanding away from the ground state in the Ising limit. Extrapolation of the results to the Heisenberg model indicated that the ground state is close to the critical point for antiferromagnetism, so if there is long range order, it is very small compared with the classical value.

In this paper we investigate the spin-$\frac{1}{2}$ HAF on the triangular lattice by high temperature series expansions. It seems a particularly useful approach for the HAF on the triangular lattice, since other methods have been rather inconclusive and quantum Monte Carlo simulations have sign problems [11]. Furthermore, there is at least one corresponding experimental system, $NaTiO_2$ [12]. Experiments are done at finite temperatures, so it is important to have accurate results for the *temperature dependence* of measurable quantities. By contrast, most of the earlier calculations just investigated ground state properties. Although there is no long range magnetic order at *finite* temperature for two-dimensional systems with continuous symmetry, the temperature dependence of certain quantities, discussed below, indicates whether the ground state is ordered [13, 14]. The aim is to compute enough terms in the expansion to determine these quantities down to temperatures where the asymptotic low temperature behavior has set in. Our results imply that there is a *small* amount of magnetic order so the system is *close* to a quantum critical point as found by Singh and Huse [10], but our calculations indicate that it lies just in the regime with LRMO. We also determine the temperature dependence of the susceptibility and specific heat.

The Hamiltonian is

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

where the $\mathbf{S}_i$ are spin-$\frac{1}{2}$ operators and the sum is over all nearest-neighbor (NN) pairs of sites, the NN interaction, J, having been set to unity. In the classical ground state, the spins form three sublattices, with the spins in different sublattices lying at 120° to each other [4,5]. The antiferromagnetic ordering wave vector $\mathbf{Q}$ is at the corner of the hexagonal Brillouin zone of the triangular lattice, so the antiferromagnetic structure factor is given by

$$S(\mathbf{Q}) = \sum_j \langle S_i^z S_j^z\rangle \exp[i\mathbf{Q}\cdot(\mathbf{R}_i - \mathbf{R}_j)]. \tag{2}$$

The phase factors are unity if the two sites are on the same sublattice and $-\frac{1}{2}$ if they are on different sublattices. The antiferromagnetic correlation length, ξ, is given by

$$\xi^2 = \frac{1}{4S(\mathbf{Q})} \sum_j (\mathbf{R}_i - \mathbf{R}_j)^2 \langle S_i^z S_j^z\rangle \exp[i\mathbf{Q}\cdot(\mathbf{R}_i - \mathbf{R}_j)], \tag{3}$$

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TABLE I. For each quantity, A, we define coefficients, a_n, by $A = \sum_{n=0} \frac{a_n}{n!} \left(\frac{\beta}{4}\right)^n$. The table shows the values of the a_n, which are all integers, for various quantities: $S(\mathbf{Q})$ is the antiferromagnetic structure factor, ξ is the antiferromagnetic correlation length, $\chi(0)$ is the uniform susceptibility, and E is the energy per site.

n	$4S(\mathbf{Q})$	$4\xi^2$	$\chi(0)$	$4E$
0	1	0	0	0
1	3	3	1	−9
2	12	24	−12	18
3	−102	−114	144	306
4	−864	−840	−1632	−3240
5	26496	49944	18000	−49176
6	97152	−672576	−254016	1466640
7	−14345712	−21018048	5472096	13626000
8	81196032	1187206272	−109168128	−1172668032
9	12374094336	−4809277056	818042112	−75256704
10	−254475179520	−1713907044864	17982044160	1392243773184
11	−14573708001792	60758490092544	778741928448	−18426692664576
12	673457798762496	1891818602827776	−90462554542080	−2213592367094784
13	19900415187750912	−225402231854284800	829570427172864	
14			181683850768637952	

which is normalized to agree with the Ornstein-Zernicke form, $S(\mathbf{q}) = S(\mathbf{Q})/[1 + \xi^2(\mathbf{q} - \mathbf{Q})^2]$, for $\mathbf{q} - \mathbf{Q}$ small. In addition, we have generated series for the energy per spin, E, and the uniform susceptibility, $\chi(0)$, defined by $\chi(0) = T^{-1} \sum_j \langle S_i^z S_j^z \rangle$.

Using the cluster method [15] we have generated series for $S(\mathbf{Q}), \xi^2, T\chi(0)$ and the logarithm of the partition function up to order 13 in powers of $\beta \equiv 1/T$. The thirteenth order calculation, which took about 12 days of CPU time on an IBM RS6000 workstation, involved clusters of 35 714 different topologies with a total of 2 269 064 363 embeddings on the lattice. Writing the series for $4S(\mathbf{Q}), 4\xi^2, \chi(0)$, and $4E$ as $\sum_n a_n(\beta/4)^n/n!$, then the coefficients a_n are integers and are given in Table I. The series were analyzed by the method of Padé approximants [16].

We found that the series for the logarithm of $S(\mathbf{Q})$ and ξ^2 behaved better than the series for the quantities themselves. In Fig. 1 we plot results for the structure factor on a double logarithmic scale obtained by analyzing the series for $\ln[4S(\mathbf{Q})]$ [17]. For this plot a Euler transformation was made to a new variable $u = 1/(T + 0.08)$ and the Padé analysis was done on the series for u. However, the results in this and the other figures do not depend greatly on the use of the Euler transformation. Figure 1 shows that the structure factor stays small for temperatures well below the mean field transition temperature, $T_c^{\mathrm{MF}} = 0.75$, but then appears to grow quite rapidly at lower temperatures. The slopes of the curves increase (in magnitude) as the temperature is lowered, consistent with a weak exponential divergence as $T \to 0$. The inset shows a plot of the correlation length, which is also very small even well below T_c^{MF}.

In order to analyze the series, we need to discuss further the expected behavior at low temperatures. If there is long range order, the correlation length should grow as [13, 14]

$$\xi = A_\xi T^x \exp(C_\xi/T)\,[1 + O(T)]\,. \tag{4}$$

For a collinear antiferromagnet, C_ξ is just 2π times the stiffness coefficient ρ_s for twisting the spins [14]. However, here, the classical ground state is noncollinear and, as a result, there are two distinct stiffnesses [18]. The factor, C_ξ, in the exponential in Eq. (4) has been determined for the classical model by Azaria *et al.* [19] who find $C_\xi = 6.994S^2$. They also find the power of T in Eq. (4) to be $x = -0.5$. In general, the structure factor varies with the correlation length as $S(\mathbf{Q}) \sim \xi^{2-\eta}$. Here $\eta = 0$ [13, 14], so

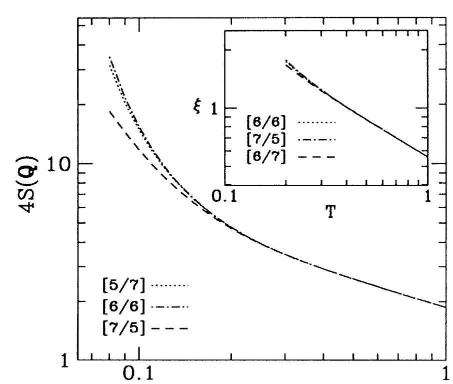

FIG. 1. A double logarithmic plot of $S(\mathbf{Q})$ against T. The data were obtained from Padés for $\ln[4S(\mathbf{Q})]$ after performing a Euler transformation to the variable $u = 1/(T + 0.08)$. The different curves refer to different Padé approximants: $[L, M]$ means that the series is represented by an Lth order polynomial in the numerator and an Mth order polynomial in the denominator. The inset shows results for the correlation length ξ.

$$S(\mathbf{Q}) = A_S T^y \exp(2C_\xi/T)\,[1 + O(T)]\,. \qquad (5)$$

Note, however, that, in general, $y \neq 2x$ because the relation $S(\mathbf{Q}) \sim \xi^2$ only holds for the *leading* exponential variation [14]. It would be very instructive to determine the value of y analytically for the HAF on the triangular lattice. To determine C_ξ, in Eqs. (4) and (5) it is useful to consider $T\ln[S(\mathbf{Q})]$ because this tends to $2C_\xi$ as $T \to 0$. For the correlation length, the power law prefactor in Eq. (4) is $x = -0.5$ [19] so this can be incorporated by considering $T\ln(T\xi^2)$, which also extrapolates to $2C_\xi$ for small T. For a system in its classical ground state [19], $2C_\xi = 2 \times 6.994 \times S^2 = 3.497$. In fact, $2C_\xi$ will be reduced below this value because of quantum fluctuations. If quantum fluctuations destroy LRMO in the ground state, then $C_\xi = 0$.

Figure 2 plots [17] $T\ln[4S(\mathbf{Q})]$ and $T\ln(16T\xi^2/3)$ against T. The Padé approximations for the structure factor are the better behaved and converge down to a temperature of about 0.15, which is $0.2T_c^{\rm MF}$. We believe that this is low enough for the system to show the asymptotic low temperature critical behavior, since Monte Carlo simulations on the corresponding classical model [20, 21] show that the low temperature regime sets in at about $T = 0.35T_c^{\rm MF}$. If there is no crossover to different behavior for $T < 0.15$, then the data for both $T\ln[4S(\mathbf{Q})]$ and $T\ln(16T\xi^2/3)$ extrapolate to a value of about $2C_\xi = 0.2$ as $T \to 0$, apparently nonzero but *far below* the classical value of 3.497. The results for the correlation length break down at a somewhat higher temperature, but this plot has the advantage that the expected power law prefactor is incorporated. It is therefore gratifying that both the structure factor and the square of the correlation length appear to extrapolate to about the same value as $T \to 0$.

Recently there has been much interest in quantum critical (QC) behavior in antiferromagnets with stiffness small compared with J [14, 22, 23]. It has been argued that there may be an intermediate temperature region in these systems, where the magnetic behavior is dominated by the $T = 0$ quantum critical point and various physical quantities have power law behavior [14, 22, 23] in T. For the square lattice, evidence for QC behavior has been found [23] in the range $0.3 < T < 0.6$. Since the stiffness for the triangular lattice appears much smaller than J, one might expect a wider QC regime for this case than for the square lattice. However, we do not find evidence for QC behavior down to $T = J/3$, perhaps because the correlation length is still only about one lattice spacing at this temperature; see the inset to Fig. 1. It is interesting to speculate on whether vortices [21] might play a role in reducing the range of the QC regime.

In Fig. 3, we show results for the susceptibility and specific heat. In both cases, the series converge down to temperatures below the peak. For the susceptibility, the zero temperature limit is rather uncertain, but the diagonal Padés give the following estimates: [4, 4] 0.184; [5, 5] 0.465; [6, 6] 0.199; [7, 7] 0.216. The

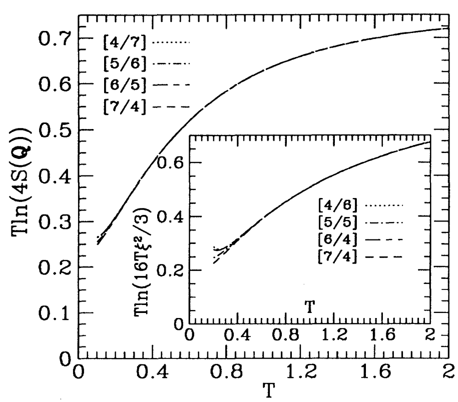

FIG. 2. $T\ln[4S(\mathbf{Q})]$ against T. The data were obtained by performing a Euler transformation, changing variables $u = 1/(T + 0.08)$, and doing a Padé analysis. The results tend to the value 0.75 ($\equiv T_c^{\rm MF}$) as $T \to \infty$ and appear to extrapolate to a value of about 0.2 as $T \to 0$. The zero temperature limit is $2C_\xi$, where C_ξ is the coefficient of $1/T$ in the exponential divergence of the correlation length; see Eq. (5). The inset shows $T\ln(16T\xi^2/3)$ against T. The data were obtained by performing a Euler transformation, changing variables to $u = 1/(T + 0.2)$, and doing a Padé analysis. The results tend to the value 1.0 as $T \to \infty$ and appear to extrapolate to a value of about 0.2 as $T \to 0$, the same as for the structure factor as expected.

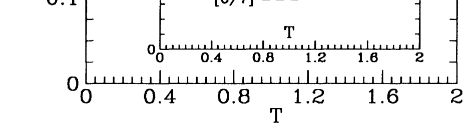

FIG. 3. Results for the uniform susceptibility (×4), against T, obtained by doing the Euler transformation $u = 1/(T + 0.08)$ followed by a Padé analysis. At high temperatures, $4\chi(0) \to 1/T$. At lower temperatures there is a broad peak at $T \simeq 0.35$ while the zero temperature limit appears to be between about 0.20 (see text). The inset shows results for the specific heat, C, obtained after doing a Euler transformation to $u = 1/(T + 0.20)$. There is a peak at $T \simeq 0.55$, and, of course, $C \to 0$ as $T \to 0$. The unphysical spike which appears on the curves at $T \simeq 1$ for the [6/7] and [5/8] Padés (but not for the [5/7] Padé) is due to the presence of a pole and a zero close by. Since the curves for *all three* Padés otherwise agree very well with each other down to $T \simeq 0.2$, we believe that the results are reliable down to this temperature if the spike is ignored.

[5, 5] result seems to be much too high so, based on the other results, we estimate a zero temperature limit of about 0.2. The specific heat vanishes, of course, at $T = 0$.

To conclude, our results indicate that the spin-$\frac{1}{2}$ HAF on the triangular lattice has small but nonzero LRMO. The only possible escape from this conclusion is if there is crossover to some other behavior at extremely low temperatures. If this is the case, other methods would also have difficulty in determining the asymptotic low temperature behavior. It might be possible to drive the system through the zero temperature critical point into the quantum disordered state by adding an antiferromagnetic second neighbor coupling. We have also accurately determined the temperature dependence of the specific heat and uniform susceptibility down to temperatures below the peaks in these quantities. Our results are significantly more precise in this region than those of Imada [6] who used a different technique. Experimental results on $NaTiO_2$ for $\chi(0)$ [12] show a gradual increase as the temperature is reduced, which may correspond to the increase shown in Fig. 3 at temperatures above the peak, then a rapid increase at low T, apparently due to impurities. Neutron scattering did not show a *strong* antiferromagnetic Bragg peak, consistent with our results, but it would be interesting to look for scattering from fluctuations in the staggered magnetization, which is proportional to $S(\mathrm{Q})$ and could therefore be compared with our results.

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