Spin Dependence of Correlations in Two-Dimensional Square-Lattice Quantum Heisenberg Antiferromagnets

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We present a series expansion study of spin-S square-lattice Heisenberg antiferromagnets. The numerical data are in excellent agreement with recent neutron scattering measurements. Our key result is that the correlation length ξ for $S > 1/2$ strongly deviates from the exact $T \to 0$ (renormalized classical) scaling prediction for all experimentally and numerically accessible temperatures. We note basic trends with S of the experimental and series expansion correlation length data and propose a sequence of crossovers from renormalized classical to classical to Curie-Weiss regimes to explain them.

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In recent years much attention has focused on two-dimensional (2D) square-lattice quantum Heisenberg antiferromagnets, described by the Hamiltonian

$$H = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j \,, \tag{1}$$

where $\langle ij \rangle$ denotes summation over all pairs of nearest neighbors. In their seminal work [1], Chakravarty, Halperin, and Nelson (CHN), utilizing a mapping of the low-energy spectrum of Eq. (1) onto the quantum nonlinear sigma model (QNLσM), have shown that the low-temperature properties of these systems obey renormalized classical (RC) scaling, where the correlation length $\xi \approx 0.31(c/2\pi\rho_s)\exp(2\pi\rho_s/T)$ for $T \ll \rho_s$ (ρ_s is the $T = 0$ spin stiffness and c the spin-wave velocity). Subsequently, Hasenfratz and Niedermayer calculated for the 2D QNLσM the exact value of the prefactor and the leading $O(T/2\pi\rho_s)$ correction [2]:

$$\xi_{\mathrm{HN}} = \frac{e}{8} \frac{c}{2\pi\rho_s} \exp\!\left(\frac{2\pi\rho_s}{T}\right) \times \left[1 - \frac{T}{4\pi\rho_s} + O\!\left(\frac{T}{2\pi\rho_s}\right)^2\right]. \tag{2}$$

The neutron scattering measurements of $\xi(T)$ in $S = 1/2$ layered Heisenberg antiferromagnets such as La_2CuO_4 [3] and $Sr_2CuO_2Cl_2$ [4] reveal a remarkable agreement with Eq. (2). At first sight, one would expect the RC description to improve as the value of the spin is increased. If S is formally regarded as a continuous variable, then the Néel order is expected to vanish for some $S < 1/2$. At the critical point, where ρ_s vanishes, Eq. (2) no longer applies and the correlation length is, in fact, inversely proportional to the temperature; this is the quantum critical regime (QC), discussed in Refs. [1,5]. [It was argued earlier [5,6] that the $S = 1/2$ Heisenberg model (1) exhibits certain signatures of QC behavior for $T > 0.5J$; we do not discuss RC to QC crossover effects in this Letter.] Naively, increasing the value of spin would move the system away from this limit so that the RC behavior would be more pronounced.

However, such an expectation does not hold. Recently, Greven and co-workers [4,7] have reported a significant discrepancy between the neutron scattering measurements of the correlation length in the $S = 1$ systems K_2NiF_4 and La_2NiO_4 and the RC prediction. Preliminary experiments on the $S = 5/2$ system Rb_2MnF_4 reveal an even larger discrepancy [8]. As is evident from Fig. 1, series expansion results for $S = 1$ [11] are in excellent agreement with the experimental data in the region of overlap, and also deviate substantially from the RC prediction (here, ξ is in units of the lattice constant a).

In order to investigate the origin of this deviation, we have calculated high-temperature expansions for the Fourier transform of the spin-spin correlation function $\langle S^z_{-\mathbf{q}} S^z_{\mathbf{q}} \rangle$ for all spin values in the range from $S = 1/2$ to $S = 5/2$. We present the data for the antiferromagnetic structure factor $S_0 = S(\mathbf{Q})$, where $\mathbf{Q} = (\pi/a, \pi/a)$, and for the second moment correlation length ξ, defined by $\xi^2 = -\partial^2 S(\mathbf{Q} + \mathbf{q})/\partial q^2|_{q=0}/2S_0$. The series are analyzed either by performing a direct Padé approximation or by taking the logarithm of the series first and then calculating the Padé approximants. The latter is likely to show better convergence if the correlation length increases exponentially fast at low temperatures. We restrict ourselves to temperature ranges where different methods of extrapolation agree within a few percent.

The ratio between the calculated ξ and the Hasenfratz and Niedermayer formula ξ_{HN} given by Eq. (2) is plotted in Fig. 2 for different spins as a function of T/ρ_s. Contrary to the naive expectation that the RC behavior becomes more pronounced as S increases, we find that for the range of temperatures probed here ξ monotonically

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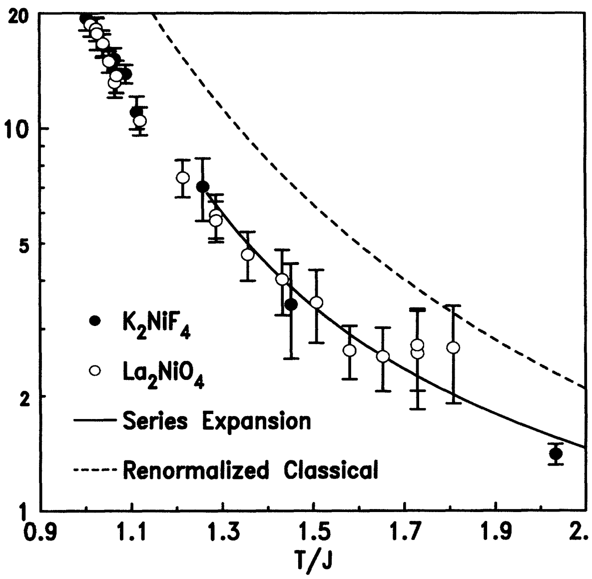

FIG. 1. Semilogarithmic plot of the series expansion result for the correlation length in the $S = 1$ Heisenberg antiferromagnet versus T/J (solid line) together with the experimental data for K_2NiF_4 (solid circles, [4]) and La_2NiO_4 (open circles, [7]). Numerical and experimental results agree with each other, and all deviate from the exact RC prediction [2] evaluated using known values for c and ρ_s [9,10] (dashed line).

deviates from ξ_{HN} as S increases. As noted above, this same systematic trend is seen experimentally [3,4,7,8].

In an attempt to understand these data, we have considered various possible scaling scenarios. To highlight the expected spin dependence, we note that for a square-lattice nearest-neighbor Heisenberg antiferromagnet $\rho_s = Z_\rho(S)JS^2$ and $c = 2^{3/2}aZ_c(S)JS$. The quantum renormalization factors $Z_\rho(S)$ and $Z_c(S)$ are known from spin-wave theory [9] (for all values of S), $T = 0$ series expansion [10] (for $S = 1/2$ and $S = 1$), and Monte Carlo studies [12] (for $S = 1/2$). For $S = 1/2$ and $S = 1$ these different methods yield good agreement.

Equation (2) may then be written as

$$\frac{S\xi}{a} = \frac{eZ_c}{2^{5/2}\pi Z_\rho}\exp\!\left(\frac{2\pi Z_\rho}{T/JS^2}\right)\left[1 - \frac{T/JS^2}{4\pi Z_\rho} + \cdots\right], \tag{3}$$

which suggests plotting $S\xi/a$ vs T/JS^2 to elucidate the dependence on S. The results so obtained are shown in Fig. 3. For $S = 1/2$ we also show Monte Carlo data [13], which extend to somewhat lower temperatures than the series expansion result. Surprisingly, over the range of $S\xi/a$ from 1 to at least 50, the data to a good approximation fall on the same curve. For the sake of clarity, we have omitted experimental data for La_2CuO_4 ($S = 1/2$), La_2NiO_4 ($S = 1$), and Rb_2MnF_4 ($S = 5/2$), all of which fall on the approximate "scaling" curve of Fig. 3 to within experimental error.

It was demonstrated in Ref. [4] that Eq. (3) describes the correlation length data in absolute units in the $S = 1/2$ system $Sr_2CuO_2Cl_2$ extremely well. Interpreted naively, Fig. 3 would then suggest that RC behavior holds for all S, but with quantum renormalization factors $Z_\rho(S)$ and $Z_c(S)$ that are nearly S independent for $1/2 \leq S \leq$

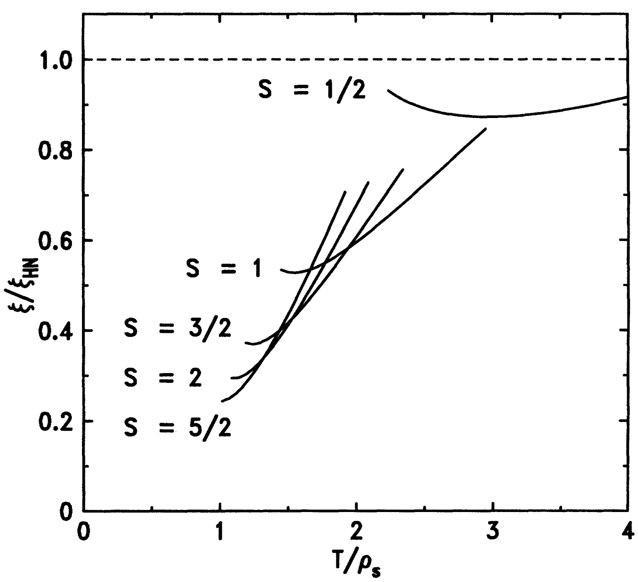

FIG. 2. Series expansion results for the correlation length plotted as ξ/ξ_{HN} vs T/ρ_s. Here ξ_{HN} is the exact RC prediction [2], Eq. (2), with c and ρ_s from Refs. [9,10]. The ratio ξ/ξ_{HN}, which is fairly close to unity for $S = 1/2$, gradually decreases as S increases, suggesting increasing disagreement with RC theory for larger S.

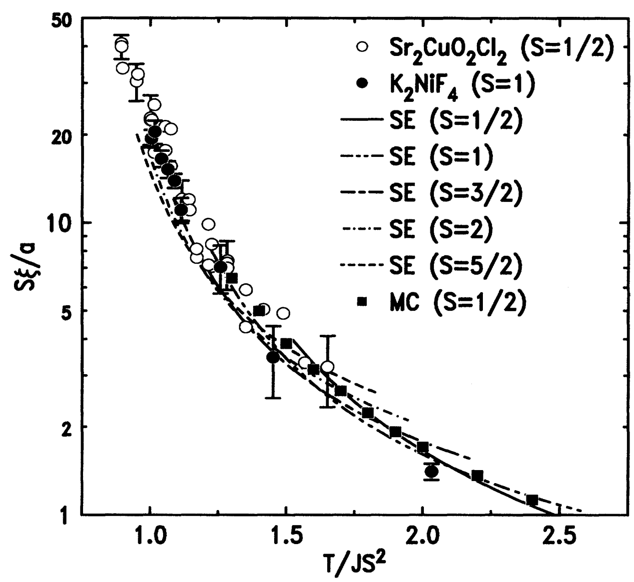

FIG. 3. Semilogarithmic plot of $S\xi/a$ vs T/JS^2 of $Sr_2CuO_2Cl_2$ ($S = 1/2$, open circles, [4]) and K_2NiF_4 ($S = 1$, solid circles, [4]) together with the series expansion results ($1/2 \leq S \leq 5/2$, represented by the various lines) as well as Monte Carlo data ($S = 1/2$, solid squares, [13]). However impressive, this data collapse is inconsistent with Eq. (2) with c and ρ_s from Ref. [9].

5/2 and close to their values at $S = 1/2$, $Z_\rho(1/2) \approx 0.72$ and $Z_c(1/2) \approx 1.18$. *However, this is very unlikely.* Spin-wave theory [9] predicts a substantial S dependence: $Z_\rho(5/2) \approx 0.95$ and $Z_c(5/2) \approx 1.03$, which is already close to the classical limit ($S \to \infty$) in which $Z_\rho, Z_c \to 1$. Serious errors in these values seem very unlikely, given their good agreement for $S = 1/2$ and $S = 1$ with those obtained by other methods [10,12].

In an attempt to understand these data, it is helpful to recall that at any fixed T/ρ_s RC theory will inevitably fail for sufficiently large values of spin. Indeed, a straightforward application of Eq. (2) to the $S \to \infty$ limit taken at $T/JS^2 \sim 1$ would predict $\xi = 0$. However, this limit corresponds to the classical Heisenberg magnet, where ξ/a is known to be nonzero and of order unity for $T \sim JS^2$.

One may understand *where* Eq. (2) might fail by following CHN in their derivation of the leading asymptotic behavior in the RC regime, but taking into account that S may be large. CHN have shown that for $T \ll \rho_s$ the magnetic correlations can be calculated using classical dynamics, except that all wave vector integrations should be limited to $|\mathbf{q}| \lesssim q_c = T/c$ rather than taken over the whole Brillouin zone. Here "classical dynamics" simply means that for $|\mathbf{q}| \lesssim q_c$, all Bose factors for spin waves can be approximated assuming $cq \ll T$. A key result of CHN is to show that such a calculation yields correct $\xi(T)$ and other observables for the quantum Heisenberg model. In other words, $\xi(T)$ for $T \ll \rho_s$ can be obtained from the expression for the correlation length $\xi_{\mathrm{CL}}(T)$ of the *classical* Heisenberg model by substituting the lattice spacing of the classical model, a_{CL}, by const $\times$ (c/T). We now evaluate the cutoff wave vector as

$$q_c \sim \frac{T}{c} \sim \frac{\rho_s}{c}\frac{T}{\rho_s} \sim \frac{S}{a}\frac{T}{\rho_s}. \qquad (4)$$

This dependence of q_c on S arises because the spin stiffness is proportional to the square of S, but the spin-wave velocity is only linear in S.

For $S \gg 1$ and $T \sim \rho_s \sim JS^2$, the cutoff wave vector $q_c \sim S/a \gg \pi/a$ is *outside* the Brillouin zone. Hence, the requirement that $cq \lesssim T$, or equivalently $q \lesssim q_c$, places no further restrictions on the q integrations which are already limited by the Brillouin zone. In this case all of the integrals are the same as those of the classical Heisenberg magnet, and the classical $S \to \infty$ limit is recovered.

The crossover temperature T_{cr} between the RC and classical regimes depends on S, and its order of magnitude can be estimated as the temperature where $q_c \sim a^{-1}$. This yields

$$T_{\mathrm{cr}} \sim \frac{c}{a} \sim JS, \quad \text{while} \quad \rho_s \sim JS^2. \qquad (5)$$

By substituting T_{cr} into Eq. (2), one concludes that the crossover from RC behavior at low temperatures to classical behavior at higher temperatures should occur for a $\xi_{\mathrm{cr}} = \xi(T_{\mathrm{cr}})$ that is larger for larger S.

In order to test this scenario, we plot the correlation length as a function of $T/JS(S+1)$ in Fig. 4, where $JS(S+1)$ is the *classical* (not $T = 0$) spin stiffness. In replacing S^2 by $S(S+1)$, we follow a purely empirical observation that the correlation length at $T \gg JS$ for $S > 3/2$ depends on S primarily through the combination $S(S+1)$ (for $T \ll JS$, ξ depends on S only through ρ_s and c). We find from our series expansion results that for $S > 1$ at intermediate temperatures $T \gtrsim JS$

$$\xi(S,T) \approx \xi_{\mathrm{CL}}(T_{\mathrm{CL}}), \quad \text{where} \quad T_{\mathrm{CL}} = T/JS(S+1), \qquad (6)$$

as shown in Fig. 4. This result supports the hypothesis that the deviations from asymptotic RC behavior evident in Figs. 1 and 2 are primarily driven by RC to classical crossover effects.

Before we conclude, we would like to point out some puzzling features regarding the Lorentzian amplitude of the spin-spin correlator, S_0/ξ^2, where S_0 is the correlator magnitude at $Q = (\pi/a, \pi/a)$, defined such that $S_0 = S(S+1)/3$ for $T \to \infty$. This ratio has only a power-law temperature dependence, and is therefore less sensitive to the model parameters than ξ or S_0 separately. The scaling prediction for this quantity is [1] $S_0/\xi^2 = Z_3 2\pi N_0^2 (T/2\pi\rho_s)^2$, where Z_3 is a universal number and N_0 is the $T = 0$ sublattice magnetization

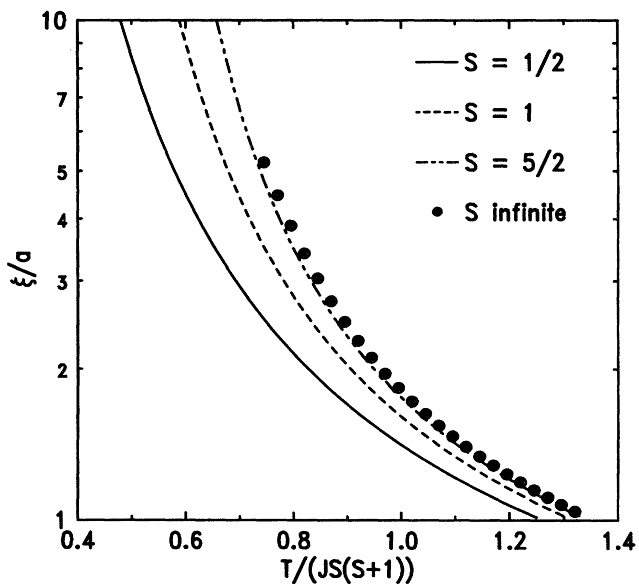

FIG. 4. Semilogarithmic plot of the correlation length obtained from series expansion plotted versus $T/JS(S+1)$ for $S = 1/2, 1$, and $5/2$. For larger spins ξ/a is close to the classical ($S \to \infty$) limit, which provides evidence that classical ($S \to \infty$) magnetic behavior holds for $JS \ll T \ll JS^2$, in agreement with our proposed scaling crossover scenario. Note, that in most of the temperature range shown, the $S \to \infty$ model is not in the scaling limit and its correlation length deviates from the expected $T \to 0$ behavior $\xi/a \approx$ const $\times$ $(T/JS^2)\exp(2\pi JS^2/T)$.

defined such that in the classical limit $N_0 = S$. The value of Z_3 can be estimated by substituting the numerical data for the two limiting cases $S = 1/2$ [11,13] and $S \to \infty$ [14,15] into the above formula. Here we estimate Z_3 at the lowest temperature accessible to us, $T_{\min}$. We get $Z_3 \approx 3.2$ for $S = 1/2$ at $T_{\min} = 0.35J$, and $Z_3 \approx 6.6$ for $S \to \infty$ at $T_{\min} \simeq 0.8JS^2$. This strong disagreement for a parameter, which was shown in Refs. [1,5] to be universal, implies that at least one of the models, and possibly both of them, are not in the scaling limit for temperatures of order their respective $T_{\min}$ [16].

Here we speculate on two possible causes of this discrepancy. First, if the classical model is in the classical scaling limit and the $S = 1/2$ model is not in the RC limit, this discrepancy may be due to the vicinity of the classical to RC crossover. In this case, $S_0/\xi^2 T^2$ for the $S = 1/2$ model should increase at temperatures lower than those studied numerically, in order to reach its presumed larger scaling limit for $T \to 0$. This scenario may be consistent with neutron scattering measurements which give $S_0/\xi^2 \sim$ const [3,4,7].

Alternatively, the finite size of the Brillouin zone (i.e., lattice corrections) may be important in determining the ratio S_0/ξ^2 for *all* values of spin at any numerically or experimentally accessible temperature, including the temperature range where the correlation length for $S = 1/2$ is itself in agreement with the universal RC prediction.

In summary, we present and analyze high-temperature series expansion data for the spin-S square-lattice Heisenberg model for all S in the range $S = 1/2$ to $S = 5/2$. In agreement with neutron scattering measurements, we find that the correlation length deviates from the low-temperature RC prediction of Eq. (2) for $S > 1/2$, and that the deviation becomes larger for larger S. We suggest that this deviation results from a sequence of crossovers, observed for large spin, from renormalized classical [$T \ll JS$, Eq. (2)], to classical ($JS \ll T \ll JS^2$), to Curie-Weiss ($T \gg JS^2$) regime. We find that in the proposed classical regime, the correlation length is given by Eq. (6), i.e., it is nearly equal to the correlation length of the classical $S \to \infty$ model with the same lattice spacing, when plotted versus $T/JS(S+1)$.

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[16] Our estimate of Z_3 from the $S \to \infty$ model agrees with earlier Monte Carlo estimates; see P. Kopietz and S. Chakravarty, Phys. Rev. B **40**, 4858 (1989), for discussion and references. Z_3 can also be calculated directly using $1/N$ expansion. As pointed out in a recent preprint by A. Sandvik, A. V. Chubukov, and S. Sachdev, $Z_3^{1/N} = 2.15$, which is 40% smaller than our estimate based on the $S = 1/2$ model. With increasing S the agreement becomes worse: there is about a factor of 3 difference between $Z_3^{1/N}$ and our $S \to \infty$ estimate.