Spin Dependence of Correlations in Two-Dimensional Quantum Heisenberg Antiferromagnets

N. Elstner\textsuperscript{1}, A. Sokol\textsuperscript{2}, R.R.P. Singh\textsuperscript{3}, M. Greven\textsuperscript{4}, and R.J. Birgeneau\textsuperscript{4}

\textsuperscript{1}Service de Physique Théorique, CEA-Saclay, 91191 Gif-sur-Yvette Cedex, France
\textsuperscript{2}Department of Physics, University of Illinois at Urbana-Champaign, Urbana, IL 61801-3080 and L.D. Landau Institute, Moscow, Russia
\textsuperscript{3}Department of Physics, University of California, Davis, CA 95616
\textsuperscript{4}Department of Physics, Massachusetts Institute of Technology, Cambridge, MA 02139

(February 27, 1995)

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 $\xi$ for $S > 1/2$ strongly deviates from the exact $T \to 0$ (renormalized classical, or RC) 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 scaling crossover scenario to explain them.

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

$$H = J \sum_{\langle ij \rangle} S_i S_j,$$

where $\langle ij \rangle$ denotes summation over all pairs of nearest neighbors. In their seminal work \cite{1}, Chakravarty, Halperin, and Nelson (CHN), utilizing a mapping of the low-energy spectrum of Eq.(1) onto the quantum nonlinear sigma model (QNL$\sigma$M), have shown that the low-temperature properties of these systems obey renormalized classical (RC) scaling, where the correlation length $\xi \approx 0.37 \times (c/\rho_s) \exp(2\pi\rho_s/T)$ for $T \ll \rho_s$ (here $\rho_s$ is the $T \to 0$ spin stiffness, and $c$ is the spin-wave velocity). Subsequently, Hasenfratz and Niedermayer calculated for the 2D QNL$\sigma$M the exact value of the prefactor and the leading $O(T/2\pi\rho_s)$ correction \cite{2}:

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

The neutron scattering measurements of $\xi(T)$ in $S = 1/2$ layered Heisenberg antiferromagnets such as La$_2$CuO$_4$ \cite{2} and Sr$_2$CuO$_2$Cl$_2$ \cite{3} reveal a remarkable agreement with Eq.(2). At first sight, one would expect the RC description to improve as the value of the spin increases. 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 $\rho_s$ vanishes, Eq.(1) fails and the correlation length is, in fact, inversely proportional to the temperature; this is the quantum critical regime (QC), discussed in Ref. \cite{4} (it was argued earlier \cite{5} that the $S = 1/2$ Heisenberg model \cite{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 moves the system away from this limit so that the RC behavior would be more pronounced.

![FIG. 1. Semi-log 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$_2$NiF$_4$ (solid circles, \cite{6}) and La$_2$NiO$_4$ (open circles, \cite{7}). Numerical and experimental results agree with each other, and all deviate from the exact RC prediction \cite{2} evaluated using known values for $c$ and $\rho_s$ \cite{8} (dashed line).]

However, such an expectation does not hold. Recently, Greven and co-workers \cite{4,7} have reported a significant discrepancy between the neutron scattering measurements of the correlation length in the $S = 1$ systems...
K$_2$NiF$_4$ and La$_2$NiO$_4$ and the RC prediction. Preliminary experiments on the $S = 5/2$ system Rb$_2$MnF$_4$ reveal an even larger discrepancy [10]. As is evident from Fig. 2, our series expansion result for $S = 1$ [1] is in excellent agreement with the experimental data in the region of overlap, and also deviates substantially from the RC prediction (note, that $\xi$ is expressed 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 $\langle S_{-q}^{-}S_{q}^{-}\rangle$ for all spins in the range $S = 1/2$ to $S = 5/2$. The generation of the series expansions will be discussed elsewhere. We present the data for the $T = 0$ structure factor $S_0 = S(Q)$, where $Q = (\pi/a, \pi/a)$ is the staggered ordering wavevector, and the second moment correlation length $\xi$ is defined by $\xi^2 = -\langle \partial^2 S(Q + q)/\partial q^2 \rangle \bigg|_{q=0}/2S_0$. The series are analyzed by either performing 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 per cent.

The ratio between the calculated $\xi$ and the Hasenfratz and Niedermayer formula $\xi_{HN}$ given by Eq. (3), is plotted in Fig. 2 for different spins as a function of $T/\rho_s$. 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 $\xi$ monotonically deviates from $\xi_{HN}$ as $S$ increases. As noted above, this same systematic trend is seen experimentally [11].

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, $\rho_s = Z_{\rho}(S)J/2$ and $c = 2^{3/2}aZ_c(S)J/2$. The quantum renormalization factors $Z_{\rho}(S)$ and $Z_c(S)$ are known from spin-wave theory [12] (for all values of $S$), $T = 0$ series expansion [13] (for $S = 1/2$ and $S = 1$), and Monte Carlo studies [14] (for $S = 1/2$). For $S = 1/2$ and $S = 1$ these different methods yield good agreement.

Eq. (3) may then be written as

$$\frac{S\xi}{a} = \frac{cZ_c}{2^{5/2}aZ_{\rho}} \exp \left( 2\pi \frac{Z_{\rho}}{T/JS^2} \right) \left[ 1 - \frac{T/JS^2}{4\pi Z_{\rho}} + \ldots \right],$$

which suggests plotting $S\xi/a$ versus $T/JS^2$ 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 [12], 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$_2$CuO$_4$ ($S = 1/2$), La$_2$NiO$_4$ ($S = 1$), and Rb$_2$MnF$_4$ ($S = 5/2$), all of which fall on the approximate “scaling” curve of Fig. 3 to within experimental error.

It was demonstrated in Ref. [3] that Eq.(3) describes the correlation length data in absolute units in the $S = 1/2$ system Sr$_2$CuO$_2$Cl$_2$ 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 $1/2 \leq S \leq 5/2$ and close to their values at $S = 1/2$, $Z_\rho(1/2) \approx 0.72$ and $Z_c(1/2) \approx 1.2$. However, this is very unlikely. Spin-wave theory [8] 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 ($S \rightarrow \infty$) in which $Z_\rho$, $Z_c \rightarrow 1$. 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 [13].

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

One can understand where Eq.(2) may 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 wavevector integrations should be limited to $|\mathbf{q}| \lesssim q_c = T/c$ rather than taken over the whole Brillouin zone. Here the words “classical dynamics” simply mean that for $|\mathbf{q}| \lesssim q_c$, all Bose factors for spin-waves can be approximated assuming $cq \ll T$. The key result of CHN is to show that such a calculation yields the correct spin correlations for the quantum Heisenberg model. We now evaluate the cutoff wavevector as

$$q_c \sim \frac{T}{c} \sim \frac{\rho_s}{\rho_s} \frac{T}{\rho_s} \sim \frac{S}{\alpha} \frac{T}{\rho_s}. \tag{4}$$

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 wavevector $q_c \sim S/a \gg \pi/a$ is outside the Brillouin zone. Hence, the requirement that $cq \ll T$, 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 $S \rightarrow \infty$ 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

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

By substituting $T_{cr}$ into Eq.(3), one concludes that the crossover from RC behavior at low temperatures to classical behavior at higher temperatures should occur for a $\xi_{cr} = \xi(T_{cr})$ that is larger for larger $S$.

In order to test this scenario, we plot the correlation length obtained from series expansion plotted versus $T/(JS(S+1))$ for $S = 1/2, 1$, and $5/2$. For larger spins $\xi/a$ is close to the classical ($S \rightarrow \infty$) limit, which provides evidence that classical ($S \rightarrow \infty$) 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 $S \rightarrow \infty$ model is not in the scaling limit and its correlation length deviates from the expected $T \rightarrow 0$ behavior $\xi/a \approx \text{const} \times (T/JS^2) \exp(2\pi JS^2/T)$.

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 $S > 3/2$ seems to depend on $S$ primarily through the combination $S(S+1)$ (for $T \ll JS$, $\xi$ depends on $S$ only.

FIG. 4. Semi-log 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 $\xi/a$ is close to the classical ($S \rightarrow \infty$) limit, which provides evidence that classical ($S \rightarrow \infty$) 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 $S \rightarrow \infty$ model is not in the scaling limit and its correlation length deviates from the expected $T \rightarrow 0$ behavior $\xi/a \approx \text{const} \times (T/JS^2) \exp(2\pi JS^2/T)$.
through $\rho_s$ and $c$). We find that $\xi$ gradually approaches the classical result as $S$ increases, and the difference between $\xi$ for the largest spin in our study ($S = 5/2$) and the classical antiferromagnet ($S \to \infty$) is already very small. This result supports the hypothesis that the deviations from asymptotic RC behavior evident in Figs. 1 and 2 are driven primarily by RC to classical crossover effects.

Another important observable of the scaling theory is the Lorentzian amplitude of the spin-spin correlator, $S_0/\xi^2$, where $S_0$ is the correlator magnitude at $Q = (\pi/a, \pi/a)$. This ratio has only a power-law temperature dependence, and is therefore less sensitive to the model parameters than $\xi$ or $S_0$ separately. A detailed discussion of this quantity is beyond the scope of this publication. However, we note some important issues which need to be clarified before the scaling behavior of $S_0/\xi^2$ can be fully understood.

The RC scaling prediction for this quantity is

$$
S_0/\xi^2 = Z_3 N_0^2 / 2\pi \left( T / \rho_s \right)^2,
$$

where $Z_3$ is a universal number and $N_0$ is the $T = 0$ sublattice magnetization defined such that in the classical ($S \to \infty$) limit $N_0 = S$, and $S_0$ is defined such that $S_0 = S(S + 1)/3$ for $T \to \infty$. The value of $Z_3$ can be estimated independently using the numerical data for the two limiting cases $S = 1/2$ and $S \to \infty$.

$Z_3$ is estimated by substituting series expansion data at the lowest temperature where calculation is possible, $T_{\text{min}}$, into Eq. (6). The data so-obtained for $S = 1/2$ and $S \to \infty$ differ by a factor of two. $Z_3 \approx 3.2$ for $S = 1/2$ when estimated at $T_{\text{min}} = 0.35 J$, and $Z_3 \approx 6.6$ for $S \to \infty$ when estimated at $T_{\text{min}} \approx 0.8 J S^2$. The disagreement of the $S = 1/2$ and $S \to \infty$ results for this universal parameter suggests that at least one of the models is not in the scaling limit at the respective $T_{\text{min}}$.

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 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 $S_0/\xi^2 \sim \text{const}$. Second, for either of the models the true $T \to 0$ scaling form for large $q \xi$ ($q$ is a deviation from $Q = (\pi/a, \pi/a)$),

$$
S(q) \approx S_0 \frac{1 + (B_f/2) \log(1 + q^2 \xi^2)}{1 + q^2 \xi^2},
$$

and the hydrodynamic result, valid for $\xi^{-1} \ll q \ll T/c$, cannot hold simultaneously at the temperatures studied for any $q$ inside the Brillouin zone. The reason is that the RC scaling form (3), which has to match the hydrodynamic expression (6) for $q \xi \to \infty$, is numerically found (11,12) to approach its asymptotic large-$q$ limit quite slowly. In the temperature range studied, it still has large deviations from this limit for any $q$ inside the Brillouin zone ($q \lesssim a^{-1}$), hence Eqs. (6) and (8) are not equal and at least one of them must be inconsistent with the data.

However, both Eqs. (6) and (8) are derived using essentially the same assumptions that (i) $c/\xi \ll T$ and that (ii) $T$ is much smaller than other energy scales. This might indicate the importance of lattice corrections (i.e. finite size of the Brillouin zone) to the ratio $S_0/\xi^2$ in the temperature range studied, in which $\xi$ is itself consistent with our proposed crossover scaling scenario for any $S$.

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. (3) for $S > 1/2$, and that the deviation becomes larger for larger $S$. We suggest that this deviation primarily reflects a crossover to classical (as opposed to renormalized classical, or RC) behavior in the temperature range $c/a \ll T \ll \rho_s$, or equivalently $J S \ll T \ll J S^2$, where the correlation length is nearly equal to that of the classical $S \to \infty$ model. In a subsequent publication (17), which is now in preparation, we propose to use comparisons between AFMs and FM's with the same value of $S$ to study the role of quantum effects as a function of spin; the results obtained in (17) using this method are consistent with the crossover scenario proposed here. Finally, we discuss the behavior of the staggered structure factor $S_0$, which measures the strength of magnetic correlations, and speculate on possible causes of its apparent deviation from the $T \to 0$ scaling limit.

We would like to thank S. Chakravarty, A.V. Chubukov, B.I. Halperin, M.A. Kastner, M.S. Makivic, D.R. Nelson, S. Sachdev, and U.-J. Wiese for helpful discussions. We thank J. Tobochnik and D.N. Lambeth for providing their numerical data for comparisons.

A.S. is supported by an A.P. Sloan Research Fellowship. R.R.P.S. is supported by the NSF under Grant No. DMR 93-18537. The work at MIT was supported by the NSF under Grant No. DMR 93-15715 and by the Center for Materials Science and Engineering under NSF Grant No. DMR 94-00334.
[1] S. Chakravarty, B.I. Halperin, and D.R. Nelson, Phys. Rev. B 39, 2344 (1989).
[2] P. Hasenfratz and F. Niedermayer, Phys. Lett. B 268, 231 (1991); Z. Phys. B 92, 91 (1993).
[3] B. Keimer et al., Phys. Rev. B 46, (1992) 14034; R.J. Birgeneau et al. (unpublished).
[4] M. Greven et al., Phys. Rev. Lett. 72, 1096 (1994); Z. Phys. B (in press).
[5] A.V. Chubukov, S. Sachdev, and J. Ye, Phys. Rev. B 49, 11919 (1994); A.V. Chubukov and S. Sachdev, Phys. Rev. Lett. 71, 169 (1993).
[6] A. Sokol, R.L. Glenister, and R.R.P. Singh, Phys. Rev. Lett., 72, 1549 (1994).
[7] K. Nakajima et al., Z. Phys. B (in press).
[8] T. Oguchi, Phys. Rev. 117, 117 (1960); J. Igarashi, Phys. Rev. B 46, 10763 (1992); C.J. Hamer, Z. Weihong, and J. Oitmaa, Phys. Rev. B 50, 6877 (1994).
[9] R.R.P. Singh, Phys. Rev. B 39, 9760 (1989); R.R.P. Singh and D. H. Huse, Phys. Rev. B 40, (1990) 7247; C.J. Hamer, Z. Weihong, and J. Oitmaa, Phys. Rev. B 50, 6877 (1994).
[10] Y.S. Lee, M. Greven, and R.J. Birgeneau (unpublished).
[11] N. Elstner, R. L. Glenister, R. R. P. Singh, and A. Sokol, to appear in Phys. Rev. B (March, 1995).
[12] We used the data by M. Greven, U.-J. Wiese, and R.J. Birgeneau (unpublished); similar results were earlier obtained by M.S. Makivic and H.-Q. Ding, Phys. Rev B 43, 3562 (1991).
[13] U.-J. Wiese and H.-P. Ying, Z. Phys. B 93, 147 (1994)
[14] M. Lüscher and P. Weisz, Nucl. Phys. B 300, 325 (1988); D.N. Lambeth and H.E. Stanley, Phys. Rev. B 12, 5302 (1975); S.H. Shenker and J. Tobochnik, Phys. Rev. B 22, 4462 (1980).
[15] S. Tyč, B.I. Halperin, and S. Chakravarty, Phys. Rev. Lett. 62, 835 (1989). The parameter $B_s$ discussed in this publication is equal to $B_s = 6\pi Z_3$.
[16] Our estimate of $Z_3$ from the $S \to \infty$ model agrees with several earlier Monte Carlo estimates; see P. Kopietz and S. Chakravarty, Phys. Rev. B 40, 4858 (1989), for discussion and a list of references. $Z_3$ can also be calculated directly using $1/N$ expansion, without having to employ Eq.(6). As pointed out in a recent preprint by A. Sandvik, A.V. Chubukov, and S. Sachdev, $Z_1^{1/N} = 2.15$, which is 40% smaller than our estimate based on the $S = 1/2$ model. We find that with increasing $S$ the agreement becomes worse; there is about a factor of three difference between $Z_1^{1/N}$ and our $S \to \infty$ estimate.
[17] A. Sokol, N. Elstner, and R.R.P. Singh, to be published.