Low Temperature Behavior and Crossovers of the Square Lattice Quantum Heisenberg Antiferromagnet

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We present thermodynamic measurements of various physical observables of the two-dimensional $S = 1/2$ isotropic quantum Heisenberg antiferromagnet on a square lattice, obtained by quantum Monte Carlo methods. The results are in excellent agreement with field-theoretical predictions. The issue of the existence of a crossover from quantum critical to renormalized classical regime is clarified. [S0031-9007(98)05597-5]

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The square lattice quantum Heisenberg antiferromagnet (QHA) is important in condensed matter physics since it describes the critical behavior of undoped insulating parent compounds of the high-T_c superconductors. Some materials believed to be well described by these models are La_2CuO_4 [1], Sr_2CuO_2Cl [2], La_2NiO_4 [3] and K_2NiF_4 [2]. Most existing theoretical treatments of the long wavelength, low energy behavior of the QHA are actually based on an effective field theory, the (2 + 1)D $O(3)$ nonlinear σ model (NLσM) [4–6].

Although the mapping from the QHA to the NLσM may be justified on the general grounds of universality, it is rigorous only for sufficiently large values of the magnitude of the quantum spin (S). There is also a subtle problem due to the existence of Berry phase terms, which are present in the effective field theory of the QHA [7], but not in the NLσM. This term is not believed to be relevant [6,8], but this is still a matter of debate [9].

The results of previous experimental and numerical studies on the $S = 1/2$ QHA show a leading exponential temperature dependence of the correlation length, in agreement with the prediction of the theories [4–6]. There, nevertheless exist some systematic discrepancies between the observed values and theoretical predictions: The two-loop order formula of the correlation length deviates from both measurements [2,3] and numerical simulations [10,11]. The deviation becomes worse as the value of S increases [11], in contrast to naive expectations. Recent neutron scattering experiments on $Sr_2CuO_2Cl_2$ [2] show systematic deviations from the theoretical prediction for the peak value of the static structure factor.

On the theoretical side a crossover from renormalized classical (RC) behavior at very low temperatures T to quantum critical (QC) behavior at intermediate temperatures was predicted for a planar antiferromagnet near a quantum critical point. In the RC regime where $T \ll 2\pi\rho_s$ (ρ_s is the spin stiffness), the QHA maps to a classical antiferromagnet with renormalized couplings. At higher temperatures, $T \sim 2\pi\rho_s$ a crossover to QC behavior, controlled by the properties of the quantum critical point where $\rho_s = 0$ is expected. It was proposed theoretically [4,6] that this crossover can even be observed in the spin-1/2 square lattice QHA, although it is not very close to the quantum critical point. The presence of this crossover remains controversial with differing claims reported in the literature [2,10,12].

In this Letter we present results of a very large scale quantum Monte Carlo (QMC) study on the $S = 1/2$ square lattice QHA up to the linear size of the lattice $L = 1000$. Our study confirms the validity of the theory [4–6] and resolves the issue of the existence of a crossover from RC to QC behavior.

Using the continuous time version [13] of the loop algorithm [14] that eliminates the systematic error due to finite Suzuki-Trotter number, we measure the thermodynamic values (infinite volume limit values) of various physical observables such as the uniform susceptibility (χ_u), staggered susceptibility (χ_{st}), second moment correlation length (ξ), peak value of the staggered structure factor $S_{\mathbf{Q}}$ at $\mathbf{Q} = (\pi, \pi)$, and internal energy ($\mathcal{E}$). For each T and L we performed $10^6 - 10^7$ single-loop updates after thermalization. Despite the large system sizes of up to $L = 1000$ no critical slowing down was observed in the simulations using the loop algorithm. The autocorrelation times always remained shorter than 1. Computer memory imposed the only limitation on the system size.

In order to monitor finite size effects in our measurements, we repeated the measurements at each temperature with varying lattice size and found that the measured values of ξ, χ_{st}, and $S_{\mathbf{Q}}$ become size independent under the condition $L/\xi \gtrsim 7$, within the typical relative statistical errors of 0.3% or better. This condition is very restrictive for the measurements of the infinite volume value of those quantities. It turns out, however, that χ_u and $\mathcal{E}$ can be measured reliably on much smaller lattices. The uniform susceptibility could be measured up to $\beta \equiv J/T = 40$, where ξ is of order $\mathcal{O}(10^{20})$. Varying L from 20 to 120, we found that the data are already size independent for $L \geq 80$ within our statistical error.

A selection of our data is shown in Table I. The complete results will be published in more detail in a forthcoming paper [15]. In the following we discuss our results and compare them to theoretical predictions.

Uniform susceptibility.—Chiral perturbation theory [5], for χ_u as $T \to 0$, predicts for the RC regime (with convention $J = \hbar = g\mu_B = k_B = 1$ hereafter)

$$\chi_u^{\mathrm{HN}} = \frac{2}{3}\chi_\perp \left[1 + \frac{T}{2\pi\rho_s} + \left(\frac{T}{2\pi\rho_s}\right)^2\right], \quad (1)$$

where $\chi_\perp = \rho_s/c^2$. It turns out that the fit of our data to Eq. (1) becomes stable only for data with $\beta \geq 4.5$. The estimated values of the parameters from the fit are

$$\chi_\perp = 0.06549(2), \qquad \rho_s = 0.178(2), \qquad c = 1.65(1). \quad (2)$$

These values are in agreement with QMC results based on finite size scaling formulas [13,16] and are also in remarkably good agreement with spin wave theory [17].

We can also observe a crossover to the QC behavior [6]

$$\chi_u = \frac{1}{c^2}[A_u T + B_u(\rho_s)] \quad (3)$$

at higher temperatures. The constant $A_u = 0.26 \pm 0.01$ [8] is universal [6]. As shown in Fig. 1, χ_u is linear in T with the universal slope A_u in a reasonably broad range $0.3 \lesssim T \lesssim 0.5$. The offset $B_u \approx 0.47\rho_s$, reasonably close to the leading order estimate of a $1/N$ expansion $B_u \approx 0.57\rho_s$ [6].

Internal energy.—The energy $\mathcal{E}$ becomes size independent under the condition $L/\xi \gtrsim 3$, allowing us to measure it for $\beta \leq 5.5$. At low temperatures the energy is the T^3 bosonic contribution of the two magnon branches with first corrections appearing only in fifth order in T [5]:

$$\mathcal{E}(T) = E_0 + \frac{2\zeta(3)}{\pi c^2}T^3 + E_5 T^5. \quad (4)$$

Because of considerable uncertainties in three parameter fits, we here fix the c to the value obtained in the above fit of χ_u, i.e, $c = 1.652$, rather than treating it as a fitting parameter. It turns out that only data for $\beta \gtrsim 4.25$ fit reasonably well to Eq. (4) with $\chi^2/N_{NF} \simeq 1.0$. We observe, however, that the fit is still slightly unstable in the sense that the values of the fitting parameters change mildly with the range of T selected for the fit. The ground state energy we have extracted,

$$E_0 = -0.66953(4), \quad (5)$$

should thus be regarded as a weak lower bound of the correct value. It may be compared with $E_0 = -0.6693 \sim -0.6694$ obtained from the ground state properties of QHA [18,19].

Correlation length.—At low temperatures, ξ diverges exponentially, similar to a classical antiferromagnet but with renormalized parameters (thus "renormalized classical" behavior) [5]

$$\xi_{\mathrm{HN}} = \frac{e}{8}\frac{c}{2\pi\rho_s}\exp\left(\frac{2\pi\rho_s}{T}\right)\left[1 - \frac{T}{4\pi\rho_s}\right]. \quad (6)$$

We measured the infinite volume limit ξ over the inverse temperature range $0.25 \leq \beta \leq 4.95$, corresponding to $0.289(2) \leq \xi \leq 120.5(4)$, and fitted it to Eq. (6) in various temperature ranges. We find that the fits are unstable, i.e., the value of ρ_s $[c]$ decreases [increases] systematically as the data at higher T are removed in the fit. Considering only data with $\xi \geq 39.2(1)$ in the fit, we find

$$\rho_s = 0.185(1), \qquad c = 1.442(3). \quad (7)$$

Because of the systematic tendency, this value of ρ_s $[c]$ should be regarded as the upper [lower] bound of the correct value.

Although these bounds are consistent with other estimates, our correlation length data strongly deviate from the asymptotic expression (Fig. 2) in our temperature range $\beta \leq 4.95$. However, we wish to note that both our data and the experimental measurements [2] can be fitted quite well by Eq. (6) with the correct ρ_s if one leaves the prefactor a free fitting parameter.

We can also resolve the controversy about a crossover to QC behavior, given by $c/\xi(T) = A_{\mathrm{QC}}\rho_s + B_{\mathrm{QC}}T$. A previous Monte Carlo study [10] claimed that all ξ fit a simple exponential form even for β as low as 0.25, which is inconsistent with our higher precision data. They thus concluded that there is no crossover. On the other hand a series expansion study for $\xi \lesssim 10$ claimed to see a crossover [12]. We indeed observe that $1/\xi$ is linear in T over a small range $1.273(6) \leq \xi \leq 3.25$,

TABLE I. A selection of thermodynamic data for the 2D QHA in the range $0.25 \leq \beta \leq 4.95$.

β	0.25	1.75	2.25	2.75	3.25	3.75	4.25	4.75	4.95
ξ	0.289(2)	2.37(1)	4.46(1)	8.38(1)	15.7(1)	29.0(1)	52.8(2)	95.7(3)	120.5(4)
χ_{st}	0.0811(2)	4.185(7)	12.54(3)	38.2(1)	117.3(3)	357.5(9)	1082(3)	3230(10)	4980(17)
χ_u	0.0487(1)	0.0840(1)	0.0735(1)	0.0656(1)	0.0604(1)	0.0572(1)	0.0550(1)	0.0535(1)	0.0531(1)
$\mathcal{E}$	−0.0986(1)	−0.5609(1)	−0.6162(1)	−0.6432(1)	−0.6559(1)	−0.6618(1)	−0.6648(1)	−0.6663(1)	−0.6676(1)
S_Q	0.329(1)	2.63(1)	5.96(1)	14.40(2)	36.91(5)	95.7(2)	254.8(5)	682(2)	1007(5)

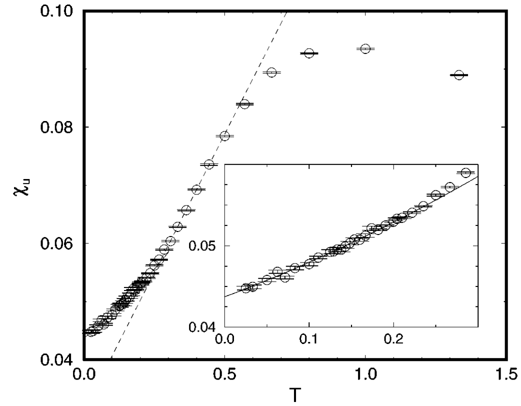

FIG. 1. The uniform susceptibility χ_u versus T. The inset shows the low-T region enlarged. The fit to the low-T prediction [Eq. (1)] is drawn as a solid line, and the fit to the quantum critical linear behavior is shown as a dashed line. In the latter fit the slope is determined completely by the value of c, extracted from the first fit.

but the measured values of A_{QC} and B_{QC} are definitely inconsistent with the universal theoretical predictions [4,6]. Moreover, we observe such a linearity even in the 2D classical Heisenberg model, where there should be no such crossover. Our data are not consistent with the one-loop order equations for the crossover regime [4,6,9] either. Thus we conclude that no QC behavior can be observed in the correlation length.

A final point that has to be mentioned is that the theoretical prediction, Eq. (6), is actually for the correlation length obtained from the real space decay of the correlation function, while the second moment correlation length is obtained from the second moment of the structure factor. However, the deviations of the structure factor from a Lorentzian are small [6,20], leading to a difference between the two definitions of about $(1-2)\%$. These small differences are thus not the reason for the discrepancies.

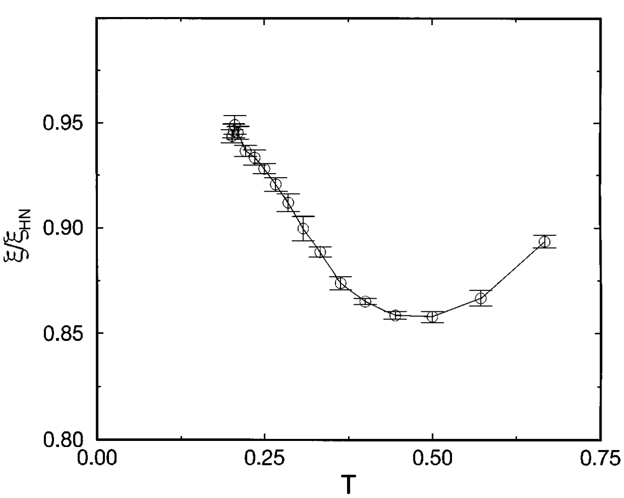

FIG. 2. Ratio of the measured and predicted correlation length, i.e., $\xi/\xi_{\mathrm{HN}}(\rho_s, c)$ with the values of ρ_s and c chosen as the mean of our values in Eq. (2) and those of Ref. [13].

The peak value of the static structure factor.—Theory [4,6,21] predicts, for low T,

$$\frac{S_{\mathbf{Q}}(T)}{\xi^2(T)} = 2\pi A M^2 \left(\frac{T}{2\pi\rho_s}\right)^2 \left(1 + C\,\frac{T}{2\pi\rho_s}\right), \quad (8)$$

with a universal constant A. $M \approx 0.307$ [19] is the ground state magnetization. However, the experimental data suggested [2,3] that $S_{\mathbf{Q}}(T)/\xi^2(T)$ is temperature independent over the temperature range accessible in the experiment.

We find that the data with $\beta \geq 2.75$ fit with a correction of $C = -0.5(1)$. For the universal prefactor, we obtain the estimate $A \approx 4.0$. A series expansion study [11] obtained $A^s_{1/2} \approx 3.2$ for spin $S = 1/2$ and $A^s_\infty \approx 6.6$ for spin $S = \infty$. Our result clearly shows that, as conjectured in Ref. [11], these values do not agree because the models are not yet in the low-T scaling regime. As even our fits at lower temperatures are unstable in the sense that A increases as we leave out data at higher T in the fit we view our estimate as a lower bound for this universal number.

Our data are definitely in agreement with the theory but not with the analysis from experiment [2]. In our opinion the deviations are caused by problems in the fits of the experimental measurements.

The staggered susceptibility.—In the classical high-T, as well as in the low-T, renormalized classical regime the staggered susceptibility is expected to be related to the staggered structure factor as [4,6]

$$\frac{S_{\mathbf{Q}}}{T\chi_{\mathrm{st}}} = 1\,, \quad (9)$$

because in both limits the spins are perfectly correlated along the imaginary time (β) axis. In the QC regime this ratio is expected to also be constant but, due to quantum fluctuations, with a different value of [6,12]

$$\frac{S_{\mathbf{Q}}}{T\chi_{\mathrm{st}}} \simeq 1.09\,. \quad (10)$$

Our data, shown in Fig. 3, are in complete agreement with previous results by Sandvik *et al.* [22]. At high and low temperatures, this ratio tends to 1. However, instead of an expected plateau with a value of 1.09 in the QC regime the data shows only a broad peak with a maximum at about $T \sim 0.8$ [22].

While the peak value is close to the predicted quantum critical value, no plateau of constant value could be seen and the temperature is outside the range where quantum critical behavior is observed in the uniform susceptibility. Thus we conclude that, although the behavior is not very different from the expected QC behavior also in this quantity, no QC regime can be found.

Conclusion and discussion.—We have presented, for a set of thermodynamic data which displays the asymptotic behavior predicted by field-theoretical approaches a variety of observables. Our estimates of ρ_s and c from the

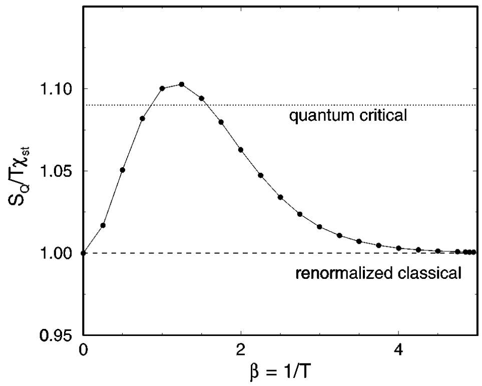

FIG. 3. Ratio of the staggered structure factor and susceptibility $\frac{S_{\mathbf{Q}}}{T\chi_{\mathrm{st}}}$ versus $\beta = J/T$. This ratio should be 1 (dashed line) *both* in the renormalized classical ($T \rightarrow 0$) and high temperature regimes ($T \rightarrow \infty$). In the quantum critical regime it is expected to be 1.09.

analysis of χ_u may be compared with previous estimates based on the size dependence formula near the ground state, i.e., $\rho_s = 0.185(2)$ and $c = 1.68(1)$ [13,16]. The values also agree well with those given by the spin wave theory of the QHA. Our results thus strongly confirm the validity of the mapping from the QHA to the NLσM in describing the long distance behavior of the former.

The fact that the asymptote of χ_u manifests itself only at very low temperatures may account for previous puzzles raised in studies of the correlation length. The deviation of ξ from Eq. (6) is reduced to approximately few percent at $\beta \simeq 4.9$ from the 20% deviation seen previously at $\beta = 2.5$. In fact, this is similar to the 2D classical Heisenberg model as demonstrated by recent numerical studies [23,24]. Our data of $S_{\mathbf{Q}}$ and χ_u also indicate that one needs to probe data with much lower T for the correction term to be safely ignored.

We also clarify the issue of the existence of the crossover. As suggested in Ref. [6] the uniform susceptibility, where all logarithmic corrections in the theory cancel, shows a clear crossover from a quantum critical to the renormalized classical regime at about $T \sim J/3$. In other quantities, e.g., in ξ, no evidence for the crossover could be found. It was, however, noted in Ref. [6] that the crossovers for different observables, though governed by the variation of the same ratio T/ρ_s, do not necessarily occur at the same temperature. It is then possible that the width of the QC region is too small to be unambiguously detected from the data.

Finally, in comparison with experiments, we wish to note that experimental measurements of the uniform susceptibility in La_2CuO_4 [25] also show QC behavior above the Néel temperature and agree perfectly with QMC data.

After completing our simulations, we became aware of a recent related preprint [26] that also addresses the correlation length at low temperatures and comes to similar conclusions regarding that quantity.

The simulations were performed in 250 000 hours of CPU time on the 1024-node Hitachi SR2201 massively parallel computer of the Computer Center of the University of Tokyo. M. T. wishes to acknowledge the Aspen Center for Physics that enabled helpful discussions with S. Chakravarty, A. Chubukov, S. Sachdev, and R. Singh.

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