Instantaneous Spin Correlations in La$_2$CuO$_4$

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We have carried out a neutron scattering study of the instantaneous spin-spin correlations in La$_2$CuO$_4$ ($T_N = 325$ K) over the temperature range 337 K to 824 K. Incident neutron energies varying from 14.7 meV to 115 meV have been employed in order to guarantee that the energy integration is carried out properly. The results so-obtained for the spin correlation length as a function of temperature when expressed in reduced units agree quantitatively both with previous results for the two dimensional (2D) tetragonal material Sr$_2$CuO$_2$Cl$_2$ and with quantum Monte Carlo results for the nearest neighbor square lattice $S=1/2$ Heisenberg model. All of the experimental and numerical results for the correlation length are well described without any adjustable parameters by the behavior predicted for the quantum non-linear sigma model in the low temperature renormalized classical regime. The amplitude, on the other hand, deviates subtly from the predicted low temperature behavior. These results are discussed in the context of recent theory for the 2D quantum Heisenberg model.

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The physics of low dimensional quantum Heisenberg antiferromagnets has been the subject of research ever since the advent of modern quantum and statistical mechanics [1,2]. Interest in two dimensional (2D) systems was heightened by the discovery of high temperature superconductivity in the lamellar copper oxides [3]. Specifically, it was realized early on that the parent compounds such as La$_2$CuO$_4$ correspond to rather good approximations to the $S=1/2$ 2D square-lattice quantum Heisenberg antiferromagnet (2DSLQHA) [4,5]. It seems at least possible that the 2D magnetism may in some way be essential to the superconductivity in the charge carrier doped cuprates. Further, the magnetism itself is of fundamental interest as a quantum many body phenomenon in lower dimensions.

Early experiments by Endoh and co-workers [5] showed that over a wide range of temperatures above the three dimensional Néel ordering transition in La$_2$CuO$_4$$_{4+y}$ (that is, La$_2$CuO$_4$ with a small amount of excess oxygen) the instantaneous spin-spin correlations were purely two dimensional and that the correlation length diverged exponentially in $1/T$. This led to a flurry of theoretical activity [2] including most especially work based on the quantum non-linear sigma model (QNLSM) by Chakravarty, Halperin and Nelson (CHN) [6] and Hasenfratz and Niedermayer (HN) [7]. These theories are all based on the 2D Heisenberg Hamiltonian which for nearest neighbor ($nn$) interactions alone takes the form

$$H = J \sum_{<i,\delta_n>} \vec{S}_i \cdot \vec{S}_{i+\delta_n}$$

where the summation is over $nn$ pairs on a square lattice.

In La$_2$CuO$_4$, for temperatures below the tetragonal (I4/mmm) - orthorhombic (Bmab) structural phase transition temperature of $T_{st} = 530$ K, the leading terms in the spin Hamiltonian [8,9] are

$$H = J \sum_{<i,\delta_n>} \vec{S}_i \cdot \vec{S}_{i+\delta_n} + \alpha_{nn} \sum_{<i,\delta_{nnn}>} \vec{S}_i \cdot \vec{S}_{i+\delta_{nnn}} + \alpha_{xy} \sum_{<i,\delta_{nnn}>} \vec{S}_i^x \vec{S}_{i+\delta_{nnn}}^y + \sum_{<i,j>} \alpha_{1} \vec{S}_i \cdot \vec{S}_{i+\delta_{1}} + \alpha_{DM} \sum_{<i,\delta_{nn}>} (-)^i \vec{a} \cdot \vec{S}_i \times \vec{S}_{i+\delta_{nn}}.$$  \hspace{1cm} (2)

Here, $\alpha_{nn}$, $\alpha_{xy}$, $\alpha_{1}$, and $\alpha_{DM}$ represent the reduced next nearest neighbor in-plane Heisenberg exchange coupling, XY anisotropy, interlayer coupling and Dzyaloshinskii-Moriya antisymmetric exchange, respectively, and $\vec{S}_i^c$ is the $c$ component of the spin at site $i$. The fourth term in Eq. (2) explicitly includes the two different out-of-plane neighbors at $\delta_{11}$ and $\delta_{12}$. Note that, as was implicit in the work of Thio et al. [8], the sign of the antisymmetric term changes on opposite sublattices because of the opposite rotation of the CuO$_6$ octahedra. This Dzyaloshinski-Moriya term originates from a small rotation of the CuO$_6$ octahedra about the $\vec{a}$ axis. In the tetragonal phase $\alpha_{DM} = 0$ and the nearest neighbor out-of-plane effective coupling vanishes since $\alpha_{1} = \alpha_{12}$.

The most complete experimental study to-date is on the material Sr$_2$CuO$_2$Cl$_2$ [10] rather than La$_2$CuO$_4$. The reasons for this are twofold: First, Sr$_2$CuO$_2$Cl$_2$ is very
difficult to dope so that there are no complications arising from the effects of doped electrons or holes on the spin correlations. Second, since Sr$_2$CuO$_2$Cl$_2$ is tetragonal down to the lowest temperatures measured ($< 10$ K), $\alpha_{DM}$=0 and the nearest neighbor interplanar coupling vanishes to leading order, that is $\alpha_{11} = \alpha_{12}$. As shown in Table (1) there is a small XY anisotropy. In addition, from results in Sr$_2$Cu$_3$O$_4$Cl$_2$ [11], we infer that there is a next nearest neighbor in-plane Heisenberg exchange coupling which is about 8% of the nearest neighbor value. To first order, the latter should simply lead to a slight renormalization of the effective $J$ in Eq. (1). The XY anisotropy will lead to a crossover from Heisenberg to XY behavior for correlation lengths $\xi/a \gtrsim 100$. Thus, Sr$_2$CuO$_2$Cl$_2$ should be a good realization of the S=1/2 2DSLQHA for length scales $\gtrsim 100$. This has, in fact, been confirmed in detail experimentally [10]; specifically, over a wide range of length scales the 2D correlation length measured in Sr$_2$CuO$_2$Cl$_2$ agrees quantitatively with results from quantum Monte Carlo (QMC) calculations carried out on the Hamiltonian Eq. (1) with S=1/2 [12-14]. The value for $J$ for Sr$_2$CuO$_2$Cl$_2$ listed in Table (1) is deduced from two magnon Raman scattering measurements [15].

Both the QMC and the Sr$_2$CuO$_2$Cl$_2$ experimental results for the correlation length in turn are quantitatively predicted by theory based on the QNL$\sigma$M in the low temperature renormalized classical (RC) regime [6, 7]. This comparison again involves no adjustable parameters. Surprisingly, this agreement holds for correlation lengths as short as a few lattice constants. This is far outside of the temperature range where the QNL$\sigma$M-RC theory should hold. A plausible explanation for this unexpected agreement has been given by Beard et al.[13].

In spite of the fact that the progenitor of this work was the discovery of high temperature superconductivity in La$_{2-x}$Ba$_x$CuO$_4$ [3] together with the early work on the 2D spin correlations in La$_2$CuO$_4$ [5], our prior knowledge of the spin correlations in stoichiometric La$_2$CuO$_4$ is rather limited. The primary correlation length data for La$_2$CuO$_4$ originate from the neutron scattering study of Keimer et al. [9] on a carrier-free single crystal of La$_2$CuO$_4$ with $T_N = 325$ K. The Keimer et al. [9] data on the correlation length and structure factor extend up to 550 K. Their measurements are generally consistent with the Sr$_2$CuO$_2$Cl$_2$, QMC and QNL$\sigma$M-RC results, but there appear to be systematic discrepancies at the limit of the error bars for the correlation length at both low and high temperatures. These neutron experiments were carried out using a single incident neutron energy of 31 meV. It seems likely that the discrepancies are an experimental artifact originating from the use of a single incident neutron energy over a wide range of temperatures. Alternatively, they could represent a real effect originating from the antisymmetric exchange and interplanar coupling terms in Eq. (2) for La$_2$CuO$_4$. Clearly, therefore, it is important to carry out a more complete study of the spin-spin correlations in La$_2$CuO$_4$ in order to characterize fully the magnetism in this parent compound of the monolayer high temperature superconductors. Such data would also be valuable for the interpretation of NQR results in La$_2$CuO$_4$ [16]. Finally, there have been some important advances in our understanding of the theory for the 2DSLQHA since the work of Greven et al.[10] on Sr$_2$CuO$_2$Cl$_2$ and it is therefore of value to re-examine the relationships between the results of experiments in real systems and theory.

The experiments were carried out primarily on the H7 triple-axis spectrometer at the High Flux Beam Reactor at Brookhaven National Laboratory. The measurements utilized the same single crystal of La$_2$CuO$_4$ as employed by Keimer et al. [9]; this crystal had a volume of about 1.5 cm$^3$. Throughout this paper we use $bma$orthorhombic axes; at $T_N = 325$ K the lattice constants are $a = 5.338$ Å, $b = 5.406$ Å, and $c = 13.141$ Å. We show in Fig. 1 the temperature dependence of the $(0 1 2)$ nuclear superlattice peak intensity together with the reduced orthorhombic splitting $(b/a)/(b+a)$ [17]. As evident from Fig. 1, the sample of La$_2$CuO$_4$ shows a sharp tetragonal-orthorhombic structural phase transition at $T_m = 530.5 \pm 0.5$ K. The sharpness of the transition in turn reflects the microscopic homogeneity of this sample.

The magnetic neutron scattering experiments were carried out in the energy-integrating two axis mode. For 2D systems the integration over energy is carried out automatically in a two axis experiment provided that the outgoing neutron wave vector $k_f$ is perpendicular to the 2D planes and provided that the neutron energy is significantly larger than the characteristic energy $\omega_0$ of the spin fluctuations at a given temperature [18]. From the theory of CHN [6, 19] one has

$$\omega_0^{CHN} = \frac{c}{\xi} \left(\frac{T}{2\pi\rho_s}\right)^{1/2}$$

|          | La$_2$CuO$_4$ | Sr$_2$CuO$_2$Cl$_2$ |
|----------|---------------|----------------------|
| $S$      | 1/2           | 1/2                  |
| $T_N$ (K) | 325           | 256.5                |
| $J$ (meV) | 135           | 125                  |
| $\alpha_{nn}$ | $\sim 0.08$  | $\sim 0.08$          |
| $\alpha_{DM}$ | $1.5 \times 10^{-2}$ | $-$              |
| $\alpha_{XY}$ | $-5.7 \times 10^{-4}$ | $-5.3 \times 10^{-4}$ |
| $\alpha_{11} - \alpha_{12}$ | $5 \times 10^{-5}$ | $\sim 10^{-8}$ |

TABLE I. Néel temperature, superexchange energy, and corrections to the 2D Heisenberg Hamiltonian for La$_2$CuO$_4$ [9] and Sr$_2$CuO$_2$Cl$_2$ [10]. $\alpha_{DM}$ and $\alpha_{XY}$ are larger than the values quoted in references [9] and [10] by factors of $(Z_s/Z_n)$ and $(Z_s/Z_n)^2$ respectively. Here $Z_s(1/2) \approx 1.17$ and $Z_n(1/2) \approx 0.6$ are the quantum renormalization factors for the spin wave velocity and spin wave gap respectively.
FIG. 1. Orthorhombic splitting and (0,1,2) superlattice peak intensity versus temperature; the data are normalized relative to each other in the temperature region of overlap. The solid line is the result of a fit to a power law \( A(T_{st}-T)^{-\beta} \) with \( \beta = 0.31 \pm 0.01 \) and \( T_{st} = 530.5 \pm 0.5 \) K.

where \( c \) and \( \rho_s \) are the zero temperature spin wave velocity and spin stiffness respectively. For \( \text{La}_2\text{CuO}_4 \) this becomes [20]

\[
\omega_{GHN}^0 \simeq \frac{850 \text{ meV} \sqrt{T}}{\xi} \sqrt{1800} \tag{4}
\]

Quantum Monte Carlo calculations by Makivić and Jarrell [21] at intermediate temperatures generally are well described by Eq. (4) but with an amplitude that is approximately twice as large. Specifically, Makivić and Jarrell [21] find that between 550 K and 800 K for \( J = 135 \) meV, as in \( \text{La}_2\text{CuO}_4 \) [20], \( \omega_0 \) varies from \( \sim 25 \) meV to \( \sim 63 \) meV. Accordingly, the following protocol was used in our measurements. Neutrons with incident energies \( E_i = 14.7 \) meV were used at the lower temperatures. With increasing temperature and hence decreasing \( \xi \) the incoming neutron energy was progressively raised to 41 meV, 90 meV and 115 meV. To ensure that the energy integration was carried out correctly, it was required that the results for the correlation length in the temperature regions of overlap agreed with each other to well within the experimental errors.

We show first in Fig. 2 preparatory data taken at a temperature of \( T = 328 \) K which is just above the 3D Néel temperature of 325 K. The incident neutron energy was \( E_i = 3.6 \) meV which results in very high momentum resolution. The two peaks evident in Fig. 2 originate from the two rods of scattering which are along (1,0, l) and (0,1,l), respectively. The equi-intensity of the two peaks implies that at 328 K the 2D spin fluctuations have at least XY symmetry, that is, at 328 K there is no measurable in-plane anisotropy induced by the antisymmetric exchange terms in Eq. (2).

We now compare the results in Fig. 5 for the correlation length in \( \text{La}_2\text{CuO}_4 \) with the predictions of various theories. We begin with the results of QMC calculations

where \( q_{2D} \) is the 2D deviation in wave vector from the positions of the (1,0,l) and (0,1,l) rods, convolved with the instrumental resolution function of the spectrometer.

The results so-obtained for the inverse correlation length \( \xi^{-1} \) are shown in Fig. 5. These data are consistent within the errors with the earlier results of Keimer et al. [9], but are much more precise and cover a wider range of temperatures. The solid line is the predicted behavior for the QNLσM in the renormalized classical regime [6, 7]; this will be discussed below. The results for the Lorentzian amplitude \( S(0)/\xi^2 \) are shown in Fig. 6. The four sets of data are normalized to unity over the temperature interval 450 K \( \leq T \leq 550 \) K.
FIG. 3. Representative energy-integrating two-axis scans in La$_2$CuO$_4$ with $E_i = 41$ meV and collimations $20' - 10' - S - 10'$. The solid lines are the result of fits to a 2D Lorentzian scattering function Eq. (5) convolved with the resolution function of the spectrometer.

for Eq. (1) with $S = 1/2$. Because of both advances in computational techniques and the implementation of finite-size scaling methods, QMC data now exist for $\xi/a$ for the $S = 1/2$ nn 2DSLQHA for length scales varying from 1 to 350,000 lattice constants. QMC results of Beard et al. [13] and Kim and Troyer [14] are plotted in Fig. 7 together with our experimental results in La$_2$CuO$_4$. The data are plotted in the reduced form $\xi/a$ vs. $J/T$. It is evident that the QMC and La$_2$CuO$_4$ results agree in absolute units over the complete temperature range ($337$ K $< T < 824$ K) or equivalently, length scale range ($3 \lesssim \xi/a \lesssim 115$). Thus, over this range the 2D spin correlations in La$_2$CuO$_4$ are entirely determined by the leading near-neighbor Heisenberg couplings and the anisotropic in-plane plus interplanar terms in Eq. (2) have no measurable effect to within the uncertainty of our experiments. Specifically, the tetragonal-orthorhombic structural phase transition at 530 K does not manifest itself in the temperature dependence of the correlation length.

We now consider the predictions of various analytical theories. A low-temperature theory for the 2DSLQHA was formulated by Chakravarty, Halperin, and Nelson [6], in which they obtained the static and dynamic properties of the 2DSLQHA by mapping it onto the 2D quantum non-linear $\sigma$ model. The 2D QNL$\sigma$M is the simplest continuum model which reproduces the correct spin-wave spectrum and spin-wave interactions of the 2DSLQHA at long wavelengths and low energies. First, CHN argued that for $S \geq 1/2$ the nn 2DSLQHA corresponds to the region of the 2D QNL$\sigma$M in which the ground state is ordered - the renormalized classical regime. Then, CHN used perturbative renormalization group arguments to derive an expression for the correlation length to two-loop order, showing a leading exponential divergence of $\xi$ versus inverse temperature. Later, Hasenfratz and Nie-dermayer [7] employed chiral perturbation theory to calculate the correlation length more precisely to three-loop order. In the RC scaling regime, the correlation length is given by

$$\frac{\xi}{a} = \frac{e}{8} \frac{c/a}{2\pi \rho_s} e^{2\pi \rho_s/T} \left[ 1 - \frac{1}{2} \left( \frac{T}{2\pi \rho_s} \right) + O \left( \frac{T}{2\pi \rho_s} \right)^2 \right],$$

(6)
which we refer to as the CHN-HN formula. The parameters \( \rho_s \) and \( c \) are the macroscopic \( T = 0 \) spin stiffness and spin-wave velocity, respectively. For the nearest-neighbor 2DSLQHA, they are related to the microscopic parameters \( J, S \) and the lattice constant \( a \) according to
\[
\rho_s = Z_\rho(S) S^2 J, \quad c = Z_c(S) 2\sqrt{2}aSJ.
\]
The coefficients \( Z_\rho(S) \) and \( Z_c(S) \) are quantum renormalization factors, which can be calculated using spin-wave theory (\( S \geq 1/2 \)) [22, 23], series expansion (\( S = 1/2, 1 \)) [23], and Monte Carlo techniques (\( S = 1/2 \)) [12-14]. For \( S = 1/2 \), the spin-wave approximation gives \( Z_\rho(1/2) \approx 0.699 \) and \( Z_c(1/2) \approx 1.18 \) [22, 23]. The most precise values for \( S = 1/2 \) currently available come from the QMC study of Beard et al. [13] who find \( c = 1.657(2)Ja \), and \( \rho_s = 0.1800(5)J \) and, for the \( T=0 \) sublattice magnetization, \( M_s = 0.30797(3)/a^2 \). These correspond to \( Z_c(1/2) = 1.172 \) and \( Z_\rho(1/2) = 0.72 \). The CHN-HN prediction for the Lorentzian amplitude \( S(0)/\xi^2 \) is
\[
\frac{S(0)}{\xi^2} = A2\pi M_s^2 \left( \frac{T}{2\pi \rho_s} \right)^2 \left[ 1 + C_1 \frac{T}{2\pi \rho_s} + O \left( \frac{T}{2\pi \rho_s} \right)^2 \right],
\]
(7)

It is of interest to compare the QNL\( \sigma \)M predictions with the corresponding predictions of the classical spin model for the \( nn \) 2DSLHA at low temperatures [2,6,24,25].

FIG. 5. Inverse magnetic correlation length of La\(_2\)CuO\(_4\). The solid line is Eq. (6) with \( J = 135 \) meV. The Néel and structural transition temperatures are indicated by arrows.

FIG. 6. Lorentzian amplitude, \( S(0)/\xi^2 \) versus temperature. The data for the different incident neutron energies are normalized to unity in the temperature range 450 K \( \lesssim T \lesssim \) 550 K.

FIG. 7. The logarithm of the reduced magnetic correlation length \( \xi/a \) versus \( J/T \). The closed circles are data for La\(_2\)CuO\(_4\) plotted with \( J = 135 \) meV, the open circles are data for Sr\(_2\)CuO\(_2\)Cl\(_2\) plotted with \( J = 125 \) meV [10], and the open squares are the results of the Monte Carlo computer simulations [12-14]. The solid line is the theoretical prediction without adjustable parameters of the 2DQNL\( \sigma \)M for the renormalized classical regime Eq. (6).
\[
\frac{\xi}{a} = 0.0125 \frac{T}{2\pi \rho_{ct}} e^{2\pi \rho_{ct}/T} \left[ 1 - b_1 \frac{T}{2\pi \rho_{ct}} + O \left( \frac{T}{\rho_{ct}} \right)^2 \right]
\]

(8)

and

\[
\frac{S(0)}{\xi^2} = A 2\pi M_s^2 \left( \frac{T}{2\pi \rho_{ct}} \right)^2 \left[ 1 + C_1 \frac{T}{2\pi \rho_{ct}} + O \left( \frac{T}{\rho_{ct}} \right)^2 \right]
\]

(9)

where for classical unit vector spins, \( \rho_{ct} = J \). For large \( S \) quantum spins, one finds that the classical limit is approached smoothly as a function of \( S \) provided that temperature is measured in units of \( JS(S+1) \), implying one that should take \( \rho_{ct} = JS(S+1) \) [2]. This choice for \( S = 1/2 \), gives \( \rho_{ct} = 0.75J \) compared with \( \rho_s = 0.18J \). The arguments in the exponents in Eq. (6) and (8) then differ by more than a factor of 4 – a very dramatic difference between renormalized classical and classical scaling behavior.

An alternative theoretical analysis of the 2DSLQHA has been carried out by Cuccoli et al. [26] in which they treat quantum fluctuations in a self-consistent Gaussian approximation separately from the classical contribution. In their approach, which they label the purely-quantum self-consistent harmonic approximation (PQSCHA), the quantum spin Hamiltonian is rewritten as an effective classical Hamiltonian, where the temperature scale is renormalized due to quantum fluctuations, and the new classical spin length appears as \( S + 1/2 \). Defining the reduced temperature as \( t = T/(JS(S+1/2)^2) \), the correlation length for the 2DSLQHA is then simply given by

\[
\xi(t) = \xi_{cl}(t_{cl}) \quad \text{with} \quad t_{cl} = \frac{t}{\theta^4(t)}.
\]

(10)

Here, \( \xi_{cl} \) is the correlation length for the corresponding classical 2D square-lattice on Heisenberg model, which is given by Eq. (8) at low temperatures and may be obtained from classical Monte Carlo calculations at higher temperatures [27], and \( \theta^4(t) \) is a temperature renormalization parameter. The PQSCHA is most accurate in the limit where quantum fluctuations are weak, and correspondingly \( \theta^4 \) is near unity. This is the case for large spin, for example, \( S = 5/2 \), and indeed this has recently been studied experimentally by Leheny et al. [28] who have measured \( S(q_{2D}) \) in the \( S = 5/2 \) 2DSLQHA material Rb\(_2\)MnF\(_4\). They follow a field-temperature trajectory which approaches the bicritical point in the phase diagram and which accordingly should show pure 2D Heisenberg behavior. In this case the PQSCHA predicts the correlation length precisely with no adjustable parameters over the inverse temperature range \( 0.5 < \rho_{ct}/T < 2 \) or equivalently, the length scale range \( 1 < \xi/a < 100 \). We note from Eq. (8) and (9) that for the classical model \( T \) always appears scaled by \( \rho_{ct} \). Thus the quantum effects in the PQSCHA can be thought of simply as a temperature dependent renormalization of \( \rho_{ct} \), that is, \( \rho_{ct} \to \theta^4(t) \rho_{ct} \).

Finally, for the QNL\( \sigma \)M there may be a crossover from renormalized classical to quantum critical behavior with increasing temperature [6]. In the QC regime heuristically one expects

\[
\frac{\xi}{a} = 0.8 \frac{c/a}{T - T_{QC}}
\]

(11)

with \( T_{QC} \geq 0 \) adjustable [2,6,10]. We emphasize that this anticipated crossover is a property of the QNL\( \sigma \)M and it may or may not occur for quantum spins on a 2D lattice.

The solid line in Fig. 5 is the QNL\( \sigma \)M-RC prediction, Eq. (6) with \( c \) and \( \rho_s \) from Beard et al. [13] As observed previously for Sr\(_2\)CuO\(_2\)Cl\(_2\) [10], Eq. (6) describes the measured correlation length of La\(_2\)CuO\(_4\) extremely well without adjustable parameters over the temperature range \( 337 \, K < T < 824 \, K \), or equivalently, the length scale range, \( \sim 3 \lesssim \xi/a \lesssim 110 \). All of the data for \( \xi/a \) from each of quantum Monte Carlo, Sr\(_2\)CuO\(_2\)Cl\(_2\) and La\(_2\)CuO\(_4\) together with Eq. (6) are plotted in the universal form \( \xi/a \) vs. \( J/T \) in Fig. 7. The evident universal behavior is, of course, both pleasing and reassuring. The good agreement of all of the experimental and numerical results with the low temperature QNL\( \sigma \)M-RC predictions down to very small values of \( J/T \) at first appears to be quite puzzling. The QMC study, Ref. [13], suggests that this agreement is, at least in part, accidental. Specifically, in crossing over from the low temperature continuum QNL\( \sigma \)M to the discrete lattice \( S = 1/2 \) Heisenberg model the higher order terms in Eq. (6) conspire such that over the measured temperature range the deviation of \( \xi/a \) from Eq. (6) is never more than 15% which is well within the experimental error.

We now focus on the high temperature behavior in La\(_2\)CuO\(_4\). We show in Fig. 8 the La\(_2\)CuO\(_4\) correlation length data together with the predictions from QNL\( \sigma \)M-RC (Eq. (6)). QNL\( \sigma \)M-QC Eq. (11), high temperature series expansion [29] and the PQSCHA which involves Eq. (10) combined with results of classical Monte Carlo simulations [27]. As observed previously for Sr\(_2\)CuO\(_2\)Cl\(_2\) [10] as well as for both \( S = 1/2 \) 2DSLQHA QMC calculations [14] and high temperature series expansion results [29], the QNL\( \sigma \)M-QC prediction, Eq. (11), disagrees strongly with the experimental results in La\(_2\)CuO\(_4\). This is, perhaps, not surprising given the extremely short length scales at the relevant temperatures. Specifically, at these short distances, the continuum QNL\( \sigma \)M approach which underlies the possible QC behavior is probably no longer valid. By contrast the PQSCHA which corresponds to classical scaling for the pure 2D
The temperature dependent correction factors (c.f. Eq. (7)) beyond the leading \( \xi^2 \) divergence are problematic. Specifically, Greven et al. [10] find in their measurements in \( \text{Sr}_2\text{CuO}_2\text{Cl}_2 \) that over the length scale \( 5 \leq \xi/a \leq 200 \), \( S(0)/\xi^2 \) is independent of temperature to within the errors. By contrast, QMC [12-14] and high temperature series expansion [29,30] studies of the \( S=1/2 \) an 2DSLQHA find \( S(0)/\xi^2 \sim T^2 \) over about the same range of length scales. In the \( S = 5/2 \) 2DSLQHA material \( \text{Rb}_2\text{MnF}_4 \), Leheny et al. [28] find a clear crossover at \( \xi/a \sim 4 \) from \( S(0)/\xi^2 \sim T^2 \) behavior to a much weaker dependence of \( S(0)/\xi^2 \) on \( T \). The data for \( S(0)/\xi^2 \) in \( \text{La}_2\text{CuO}_4 \) shown in Fig. 5 are clearly inconsistent with \( T^2 \) behavior over the complete temperature range but would allow a gradual crossover as found in \( \text{Rb}_2\text{MnF}_4 \). This lack of universality in \( S(0)/\xi^2 \) seems surprising given the robust universality of the behavior for \( \xi/a \) (Fig. 7). Of course, departures from the low temperature QNL\( \sigma \)M-RC behavior may occur at different temperatures for different quantities. It is also possible that the terms in Eq. (2) beyond the nearest neighbor Heisenberg coupling will effect \( S(0)/\xi^2 \) more than they effect \( \xi \) itself.

In summary, we have carried out a neutron scattering study of the instantaneous spin-spin correlations in \( \text{La}_2\text{CuO}_4 \) (\( T_N = 325 \text{ K} \)) over the temperature range 337 K to 824 K. Incident neutron energies varying from 14.7 meV to 115 meV have been employed in order to guarantee that the energy integration is carried out properly. The results so-obtained for the spin correlation length as a function of temperature when expressed in reduced units agree quantitatively both with previous results for the 2D tetragonal material \( \text{Sr}_2\text{CuO}_2\text{Cl}_2 \) and with quantum Monte Carlo results for the nearest neighbor square lattice \( S = 1/2 \) Heisenberg model. All of the experimental results for the correlation length are well described without any adjustable parameters by the behavior predicted for the quantum non-linear sigma model in the low temperature renormalized classical regime. The structure factor, on the other hand, deviates subtly from the predicted low temperature behavior although the leading \( \xi^2 \) behavior is confirmed. The correlation length data at high temperature agree reasonably well with predictions of the PQSCHA which corresponds to classical scaling with quantum corrections for the 2D Heisenberg model. We therefore hypothesize that in \( \text{La}_2\text{CuO}_4 \) there is a gradual crossover from renormalized classical to classical scaling with increasing temperature.

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References

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1. H. A. Bethe, Z. Phys. 71, 205 (1931).
2. For reviews see: E. Manousakis, Rev. Mod. Phys. 63, 1 (1991); N. Elstner, Int. J. Mod. Phys. B11, 1753 (1997).
3. J. G. Bednorz and K. A. Müller, Z. Phys. B64, 189 (1986).
4. P. W. Anderson, Science 235, 1196 (1987).
5. Y. Endoh, K. Yamada, R. J. Birgeneau, D. R. Gabbe, H. P. Jennsen, M. A. Kastner, C. J. Peters, P. J. Picone, T. R. Thurston, J. M. Tranquada, G. Shirane, Y. Hidaka, M. Oda, Y. Enomoto, M. Suzuki, and T. Murakami, Phys. Rev. B37, 7443 (1988).
6. S. Chakravarty, B.I. Halperin, and D.R. Nelson, Phys. Rev. B39, 2344 (1989). For a detailed discussion of the quantum critical region see also A. Chubukov, S. Sachdev and Y. Ye, Phys. Rev. B49, 2344 (1994).
7. P. Hasenfratz and F. Niedermayer, Phys. Lett. B 268, 231 (1991).
8. T. Thio, T. R. Thurston, N. W. Preyer, P. J. Picone, M. A. Kastner, H. P. Jennsen, D. R. Gabbe, C. Y. Chen, R. J. Birgeneau, and A. Aharony, Phys. Rev. B 38, 905 (1988).
9. B. Keimer, N. Belk, R. J. Birgeneau, A. Cascarano, C. Y. Chen, M. Greven, M. A. Kastner, A. Aharony, Y. Endoh, R. W. Erwin, and G. Shirane, Phys. Rev. B 46, 14034 (1992).
10. M. Greven, R. J. Birgeneau, Y. Endoh, M. A. Kastner, M. Matsuda, and G. Shirane, Z. Phys. B 96, 465 (1995).
11. F. C. Chou, A. Aharony, R. J. Birgeneau, O. Entin-Wohlman, M. Greven, A. B. Harnig, M. A. Kastner, Y.-J. Kim, D. S. Kleinberg, Y. S. Lee, and Q. Zhu, Phys. Rev. Lett. 78, 535 (1998); Y.-J. Kim (private communication).
12. M. Makivić and H.-Q. Ding, Phys. Rev. B43, 3562 (1991).
13. B. Beard, R. J. Birgeneau, M. Greven, and U.-J. Wiese, Phys. Rev. Lett 80, 1742 (1998).
14. J.-K. Kim and M. Troyer, Phys. Rev. Lett. 80, 2705 (1998).
15. Y. Tokura, S. Koshihara, T. Arima, H. Takagi, S. Ishibashi, T. Ido, and S. Uchida, Phys. Rev. B41, 11657 (1990).
16. T. Imai, C. P. Schlieter, K. Yoshimura, M. Katoh, and K. Kosuge, Phys. Rev. Lett. 71, 1254 (1993).
17. R. J. Birgeneau, C. Y. Chen, D. R. Gabbe, H. P. Jennsen, M. A. Kastner, P. J. Picone, T. Thio, T. R. Thurston, and H. L. Tuller, Phys. Rev. Lett. 59, 1329 (1987).
18. R. J. Birgeneau, J. Skalyo, Jr., and G. Shirane, Phys. Rev. B3, 1736 (1971).
19. S. Tyč, B. I. Halperin, and S. Chakravarty, Phys. Rev. Lett. 62, 835 (1989).
20. G. Aeppli, S. M. Hayden, H. A. Mook, Z. Fisk, S. W. Cheong, D. Rytz, J. P. Remeika, G. P. Espinosa, and A. S. Cooper, Phys. Rev. Lett. 62, 2052 (1989).
21. M. Makivić and M. Jarrell, Phys. Rev. Lett. 68, 1770 (1992).
22. J. Igarashi, Phys. Rev. B46, 10763 (1992).
23. R.R.P. Singh, Phys. Rev. B39, 9760 (1989); C. Hamer, Z. WeiHong, and J. Oitmaa, Phys. Rev. B 50, 6877 (1994).
24. Th. Jolicoeur and J. C. LeGouillou, Mod. Phys. Lett. B5, 593 (1991).
25. D. R. Nelson and R. A. Pelcovits, Phys. Rev. B16, 2191 (1977).
26. A. Cuccoli, V. Tognetti, R. Vaia, and P. Verrucchi, Phys. Rev. B56, 14456 (1997).
27. J.-K. Kim, Phys. Rev. D50, 4663 (1994).
28. R. L. Leheny, R. J. Christianson, R. J. Birgeneau, and R. W. Erwin, Phys. Rev. Lett. 82, 418 (1999).
29. A. Sokol, R.L. Glenister and R.R.P. Singh, Phys. Rev. Lett., 72, 1549 (1994).
30. N. Elstner, A. Sokol, R. R. P. Singh, M. Greven, and R.J. Birgeneau, Phys. Rev. Lett. 75, 938 (1995).
31. See for example L.C. LeGuillon and J. Zinn-Justin,Phys. Rev. B21, 3976 (1980).

32. O.A. Starykh, A.W. Sandvik, and R.R.P. Singh,Phys. Rev. B55, 14953 (1997).