Spin Correlations in the 2D Heisenberg Antiferromagnet $Sr_2CuO_2Cl_2$: Neutron Scattering, Monte Carlo Simulation, and Theory

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(Received 23 July 1993)

We report a neutron scattering study of the spin correlations in the model 2D, $S=\frac{1}{2}$, square-lattice Heisenberg antiferromagnet $Sr_2CuO_2Cl_2$. The spin correlation lengths obtained agree quantitatively with values deduced from Monte Carlo simulations over a wide range of temperature. The combined data, which cover the length scale from 1 to 200 lattice constants, are predicted accurately with no adjustable parameters by renormalized classical theory for the quantum nonlinear sigma model.

PACS numbers: 75.10.Jm, 75.25.+z, 75.40.−s

The discovery of high temperature superconductivity in the lamellar copper oxides in 1986 has led to a renaissance not only in the field of superconductivity but also in the field of lower dimensional quantum magnetism. The reason for the latter is that the parent compounds such as La_2CuO_4 correspond to rather good approximations to the $S=\frac{1}{2}$ two-dimensional (2D) square-lattice Heisenberg antiferromagnet (2DSLHA) [1,2]. Prior to 1986 this 2D quantum system represented one of the major unsolved problems in quantum statistical physics. As a result of symbiotic interactions between neutron scattering experiments [1,2], Monte Carlo simulations [3], and theory [4–6], a coherent picture has emerged for the low temperature properties of the $S=\frac{1}{2}$ 2DSLHA. It is now generally agreed that the spin-spin correlation length diverges exponentially with decreasing temperature leading to true long range order at $T=0$. Furthermore, the correlation lengths in real systems such as La_2CuO_4 are predicted rather well in absolute units by theory [2]. There are, nevertheless, persistent discrepancies between experiment and theory for various physical quantities [2,7] especially at intermediate and high temperatures, $T \geq 0.36J$, where J is the nearest neighbor antiferromagnetic exchange. One possible explanation for these discrepancies is that they signal a crossover with increasing temperature from "renormalized classical" to "quantum critical" behavior. Not surprisingly, there are also alternative explanations [2]. In order to test the theories properly one requires precise correlation length data covering as wide a temperature range as possible in a system which is described accurately by the ideal 2D $S=\frac{1}{2}$ Heisenberg Hamiltonian.

In this paper we report energy-integrating neutron scattering studies of the 2D Cu^{2+} $S=\frac{1}{2}$ instantaneous spin-spin correlations in the material $Sr_2CuO_2Cl_2$ [8]. As we shall discuss below, this system is the best experimental realization found to date of the $S=\frac{1}{2}$ 2DSLHA. Further, the exchange coupling J is known reasonably well from two-magnon Raman scattering measurements [9]. Our experiments cover the range $0.19 \leq T/J < 0.4$. Monte Carlo simulations by Makivić and Ding [3] give the correlation length for the nearest neighbor $S=\frac{1}{2}$ 2DSLHA for the temperature range $0.27 \leq T/J < 2$. We find that the $Sr_2CuO_2Cl_2$ data and the Monte Carlo data agree in absolute units in the region of overlap. The combined Monte Carlo and $Sr_2CuO_2Cl_2$ correlation length data cover the distance range from 200 lattice constants to <1 lattice constant. Recently, Hasenfratz and Niedermayer (HN) [5], building on the pioneering theoretical work of Chakravarty, Halperin, and Nelson (CHN) [4], have given an exact expression for the correlation length of the 2D quantum nonlinear sigma model (2DQNLσM) in the renormalized classical regime; they parametrize the QNLσM in terms of the spin wave velocity c and the spin stiffness ρ_s. Further, from recent theory for the nearest neighbor $S=\frac{1}{2}$ 2DSLHA the proportionality constants between c, ρ_s, and J are accurately known [10–12]. Thus it is possible to make a no-adjustable-parameter comparison between the theory for the 2DQNLσM and the combined $Sr_2CuO_2Cl_2$–Monte Carlo data. We find that the renormalized classical theory describes the combined data accurately over the temperature range $0.19 \leq T/J \leq 1$, or, equivalently, the length scale from 200 to ~ 1 lattice constant.

The experiments were carried out on the H7 and H8 triple-axis spectrometers at the Brookhaven High Flux Beam Reactor. The spectrometers were operated in the two-axis, energy-integrating mode with collimator sequence 10′-10′-S-10′. The scattering geometry was chosen such that the outgoing neutrons were perpendicular to the CuO_2 planes thence integrating over energy without changing the in-plane momentum transfer. In order to optimize the momentum resolution the incoming neutron energy was varied from 5.0 to 14.7 to 30.5 to 41.0 meV with increasing temperature. The essential concern in energy-integrating two-axis neutron measurements is that the incoming energy must be such that the experiment integrates properly over the relevant dynamic

0031-9007/94/72(7)/1096(4)$06.00

fluctuations. To ensure that this condition was satisfied we carried out simulations using the theory of Tyč, Halperin, and Chakravarty [13] for the dynamics of the $S=\frac{1}{2}$ 2DSLHA. This theory predicts that the characteristic energy $\omega_0(T)$ scales inversely with the correlation length $\xi(T)$. Thus with increasing temperature as ξ decreases, the incoming neutron energy must be proportionately increased. In all cases, we carried out experiments over an overlapping range of temperatures using a given neutron energy and the next highest neutron energy in the above sequence, and we explicitly verified that the two measurements yielded the same value for the instantaneous correlation length.

Single crystals of $Sr_2CuO_2Cl_2$ with $T_N=256.5\pm1.5$ K were grown following the same synthesis procedure as used by Miller *et al.* [8]; typical sample dimensions were $1\times1\times0.1$ cm^3. In order to obtain a satisfactory signal we aligned three crystals; the resultant overall mosaicity was $\sim0.15°$ half width at half maximum (HWHM). The crystals were mounted with a $(1\bar{1}0)$ axis perpendicular to the scattering plane in either a Dewar or an air furnace for measurements below and above room temperature, respectively. $Sr_2CuO_2Cl_2$ has the K_2NiF_4 crystal structure, space group $I4/mmm$, with square sheets of CuO_2 separated by two intervening sheets of SrCl. The material is tetragonal down to at least 10 K; this high symmetry makes the magnetic properties much simpler than those of La_2CuO_4. The room temperature lattice constants are $a=3.967$ Å and $c=15.59$ Å. The Cu^{2+} $S=\frac{1}{2}$ 2D spin Hamiltonian is given simply by

$$H=\sum_{\langle i,j\rangle}J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j+\sum_{\langle i,j\rangle}J_{ij}^{XY}S_i^zS_j^z\,. \tag{1}$$

From overlap considerations we expect that the isotropic and anisotropic exchange interactions are overwhelmingly between nearest neighbor Cu^{2+} spins alone. From the two-magnon Raman scattering measurements of Tokura *et al.* [9] we deduce for the nearest neighbor exchange $J=125\pm6$ meV. From measurements of the out-of-plane spin-wave gap we find $J^{XY}/J\sim1.4\times10^{-4}$. Because of the perfect tetragonal symmetry with its resultant cancellation of the nearest neighbor interplanar isotropic exchange, the net 3D coupling is reduced by several orders of magnitude below the XY anisotropy. Thus $Sr_2CuO_2Cl_2$ is indeed a very good approximation to the ideal $S=\frac{1}{2}$ 2DSLHA. Finally, we note that $Sr_2CuO_2Cl_2$ is difficult to dope chemically with either electrons or holes. Thus compared with, for example, La_2CuO_4 the magnetism in the CuO_2 sheets in $Sr_2CuO_2Cl_2$ is relatively immune to the effects of intrinsic or extrinsic carriers [2].

We show in Fig. 1 representative two-axis scans for $E_i=14.7$ and 41.0 meV at several temperatures. The background which is Q independent arises primarily from Cl nuclear incoherent scattering. In order to extract the intrinsic peak widths we fitted our data to the form

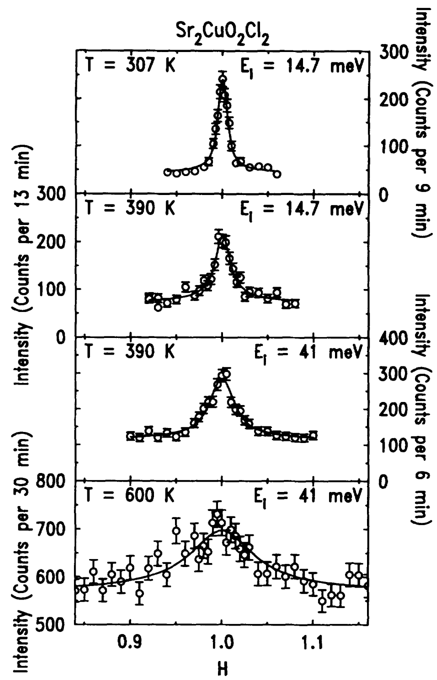

FIG. 1. Two-axis scans along the direction $(H/2,H/2,L)$ with L chosen such that $\mathbf{k}_f\|\mathbf{c}$ for $E_i=14.7$ meV and $E_i=41.0$ meV. For the 41.0 meV data the first collimator was increased to 20′. The solid lines are the results of least squares fits by a 2D Lorentzian, Eq. (2), convoluted with the instrumental resolution function. Note that upon deconvolution the two scans at 390 K yield the same value for the inverse correlation length κ.

$$S(q_{2D})=\frac{S(0)}{1+q_{2D}^2/\kappa^2} \tag{2}$$

convoluted with the instrumental resolution function. Here $\mathbf{q}_{2D}=(Q_x-\frac{1}{2},Q_y-\frac{1}{2},0)$ where $\mathbf{Q}$ is the momentum transfer in reciprocal lattice units. It is clear from Fig. 1 that the 2D Lorentzian form, Eq. (2), describes the measured profiles rather well. We will discuss the results for $S(0)$ in a future publication. In this paper we confine our attention to the behavior of the correlation length. The results for the inverse correlation length κ are shown in Fig. 2. It is evident from Fig. 2 that the correlation lengths obtained with different incoming neutron energies in the overlap region agree very well with each other, thence confirming that the energy integration is indeed being carried out properly experimentally.

Before discussing the theory we first compare our data with the results of the Monte Carlo simulations of Makivić and Ding [3] on the $S=\frac{1}{2}$ 2DSLHA with pure nearest neighbor interactions. The data may be compared quantitatively in reduced form without any adjustable parameters by plotting ξ/a vs J/T. The result is shown in Fig. 3. There is a substantial region of overlap covering the length range from about 6 to 28 lattice constants. As is evident in Fig. 3 the $Sr_2CuO_2Cl_2$ and Monte Carlo results agree within experimental error. This abso-

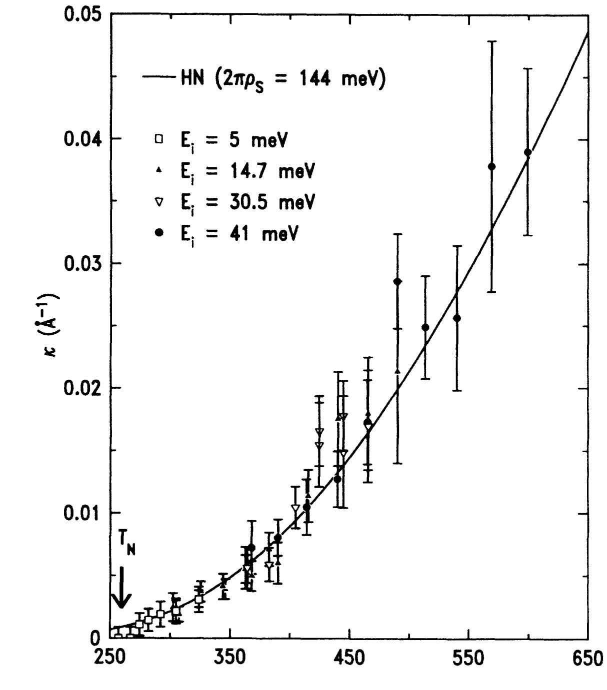

FIG. 2. Inverse correlation length versus temperature in $Sr_2CuO_2Cl_2$. The solid line is Eq. (5) with $J = 125$ meV.

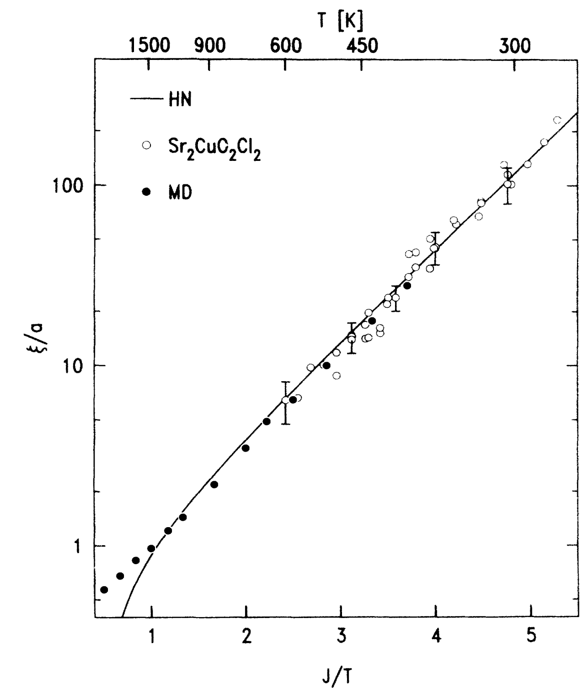

FIG. 3. Semilog plot of the reduced magnetic correlation length ξ/a versus J/T. The open circles are the data for $Sr_2CuO_2Cl_2$ from Fig. 2 plotted with $J = 125$ meV, and the filled circles are the results of the Monte Carlo simulations by Makivić and Ding [3]. The solid line is the theoretical prediction of the 2DQNLσM, Eq. (5). We emphasize that in this plot Eq. (5) has no adjustable parameters.

lute agreement by itself is gratifying; further it serves to verify the essential correctness of our approach.

As noted above, pioneering theoretical work on the 2DSLHA has been carried out by CHN [4]. They computed the static and dynamic properties of the 2DSLHA by first mapping it into the 2DQNLσM. The 2DQNLσM is the simplest continuum model which reproduces the correct spin wave spectrum and spin wave interactions of the 2DSLHA at long wavelengths [4–6]. The effective Euclidean action of the 2DQNLσM may be written ($k_B = \hbar = 1$)

$$S_{\rm eff} = \frac{\rho_s^0}{2} \int_0^{1/T} d\tau \int d^2x \left[[\nabla \cdot \mathbf{\Omega}]^2 + \frac{1}{c^2} \left| \frac{\partial \mathbf{\Omega}}{\partial \tau} \right|^2 \right], \quad (3)$$

where $\mathbf{\Omega}$ is a three-component vector field ($|\Omega| = 1$) and the space integrals are carried out to a maximum wave vector Λ; ρ_s^0 is the bare stiffness constant and c is the spin-wave velocity. Equation (3) is conveniently reparametrized in terms of a dimensionless coupling constant $\tilde{g}_0 = \Lambda c/\rho_s^0$ and c. At low temperatures CHN [4] find that there are three basic regimes depending upon the value of the renormalized coupling constant $\tilde{g}$ relative to a critical value $\tilde{g}_c$. For $\tilde{g} < \tilde{g}_c$ the system should exhibit renormalized classical behavior with the correlation length diverging exponentially in $2\pi\rho_s/T$. For $\tilde{g} \simeq \tilde{g}_c$ the system is predicted to exhibit quantum critical behavior with $\xi \sim 0.8c/T$; finally, for $\tilde{g} > \tilde{g}_c$ the system should exhibit quantum disordered behavior with ξ finite as $T \to 0$; in the quantum disordered phase there should be a gap in the excitation spectrum. CHN argue that the $S = \frac{1}{2}$ 2DSLHA with predominantly nearest neighbor interactions should exhibit renormalized classical behavior at low temperatures and this prediction was shown to be in agreement with experiments in La_2CuO_4 [1,2]. Generally, it is expected that at higher temperatures a crossover will occur from renormalized classical to quantum critical behavior but the crossover length and temperature have not been explicitly calculated and the crossover function itself is only known to the one-loop approximation.

The CHN model [4] has been refined by HN [5]. They show that in the renormalized classical region the correlation length is given rigorously by

$$\frac{\xi}{a} = \frac{e}{8} \frac{c/a}{2\pi\rho_s} e^{2\pi\rho_s/T} \left[1 - \frac{1}{2} \left(\frac{T}{2\pi\rho_s} \right) + O\left(\frac{T}{2\pi\rho_s} \right)^2 \right]. \quad (4)$$

In order to compare the predictions of Eq. (4) for the 2DQNLσM with the data shown in Figs. 2 and 3 it is necessary to know the relationships between c, ρ_s, and J. Fortunately these have been determined rather well by recent theory and Monte Carlo simulations [10–12]. The spin wave velocity of the $S = \frac{1}{2}$ 2DSLHA is given simply by $c = Z_c \sqrt{2} Ja$ where in the spin wave approximation $Z_c = 1.18 + O(1/2S)^3$ [10]. An identical result for Z_c is found in Monte Carlo simulations [11]. Related calculations give $2\pi\rho_s = 1.15J$ [10,12] with an uncertainty in the proportionality constant of about 2%. Substitution of these values into Eq. (4) then yields

$$\frac{\xi}{a} = 0.493 e^{1.15J/T}\left[1 - 0.43\left(\frac{T}{J}\right) + O\left(\frac{T}{J}\right)^2\right]. \tag{5}$$

The solid lines in Figs. 2 and 3 correspond to Eq. (5). In Fig. 2 we have used the measured value $J = 125$ meV. It is evident from Fig. 2 that Eq. (5) describes the inverse correlation length $\kappa = \xi^{-1}$ in $Sr_2CuO_2Cl_2$ very well. The comparison between the CHN-HN theory and the combined $Sr_2CuO_2Cl_2$–Monte Carlo results shown in Fig. 3 is especially notable. The renormalized classical theory describes the combined correlation length data accurately over the inverse temperature range $1 < J/T < 5.2$ or, using the measured exchange for $Sr_2CuO_2Cl_2$, $275 < T < 1450$ K. Concomitantly, the theory works for length scales from 200 lattice constants down to 1 lattice constant with no adjustable parameters. We reiterate that technically Eq. (4) is the result for the 2DQNLσM rather than the $S = \frac{1}{2}$ 2DSLHA. Figure 3 thus proves that the isomorphism between the two models is valid down to very short length scales.

A number of issues remain to be addressed both experimentally and theoretically. First, at high temperatures higher order terms in T/J in Eq. (5) should become important. Clearly, it is essential that the HN theory be extended to calculate the coefficients of the $(T/J)^2$ and possibly $(T/J)^3$ correction terms. We have confirmed that by including small terms in $(T/J)^2$ and $(T/J)^3$ in Eq. (5) with fitted coefficients the agreement up to $T/J \sim 1$ can be made essentially perfect. Second, at intermediate T/J there is no apparent evidence for a crossover from renormalized classical to quantum critical behavior [7,14,15]. We remind the reader that in the quantum critical regime $\xi \sim J/T$ rather than $\xi \sim e^{1.15J/T}$. In several recent papers it has been suggested that certain results in La_2CuO_4 may signal such a crossover for $T/J \geq 0.36$ [7,14,15]. However, the arguments are complicated [14,15] and the experimental evidence is largely indirect [7]. The combined data for ξ/a vs T in Fig. 3 provide a more direct testing ground for any such theories. At the minimum, any claims of a crossover from renormalized classical to quantum critical behavior must be supported by explicit calculations of a crossover function which, in turn, must agree with the data shown in Fig. 3. We note that the most accurate current estimate [6] of the slope in $\xi/a = (0.8c/a)(T - T\rho_s)^{-1}$ in the quantum critical regime disagrees by a factor of 2 with that which best describes the data in Fig. 3 for $500 < T < 800$ K (see also Ref. [14]). Finally, we expect a crossover from 2D Heisenberg to 2D XY behavior when the correlation length exceeds $\sqrt{J/J^{XY}} \sim 85$ lattice constants. As is evident in Figs. 2 and 3 there is no signature of such a crossover in the correlation length data for length scales up to ~ 200 lattice constants which corresponds to $T \simeq 275$ K. There is, however, some evidence that between $T = 275$ K and $T_N = 256.5$ K new behavior may be observed. We note that the NMR relaxation data of Borsa *et al.* [16] indeed imply a crossover to critical behavior below $T_N + 17$ K. Clearly precise scattering measurements very near T_N, possibly using synchrotron magnetic x-ray scattering techniques, will be required to probe the spin correlations in the XY regime.

We would like to thank S. Chakravarty, B. I. Halperin, D. C. Johnston, S. K. Sinha, A. Sokol, and U.-J. Wiese for helpful discussions. This work was supported by the U.S.-Japan Cooperative Neutron Scattering Program. The work at Tohoku University was supported by a Grant-In-Aid for Scientific Research from the Japanese Ministry of Education, Science and Culture. The work at BNL was supported by the Division of Materials Science, the Office of Basic Energy Science of the U.S. Department of Energy, under Contract No. DE-AC02-76CH-00016. The work at MIT was supported by the U.S. National Science Foundation under Contracts No. DMR 90-22933 and No. DMR 90-07825.

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