Critical Spin Dynamics of the 2D Quantum Heisenberg Antiferromagnets $Sr_2CuO_2Cl_2$ and $Sr_2Cu_3O_4Cl_2$

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We report a neutron scattering study of the long-wavelength dynamic spin correlations in the model two-dimensional $S = 1/2$ square lattice Heisenberg antiferromagnets $Sr_2CuO_2Cl_2$ and $Sr_2Cu_3O_4Cl_2$. The characteristic energy scale, $\omega_0(T/J)$, is determined by measuring the quasielastic peak width in the paramagnetic phase over a wide range of temperature ($0.2 \lesssim T/J \lesssim 0.7$). The obtained values for $\omega_0(T/J)$ agree *quantitatively* between the two compounds and also with values deduced from quantum Monte Carlo simulations. The combined data show scaling behavior, $\omega \sim \xi^{-z}$, over the entire temperature range with $z = 1.0(1)$, in agreement with dynamic scaling theory.

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Since the discovery of high temperature superconductivity in 1986 a great number of studies have been devoted to understanding this fascinating yet difficult problem. Although the superconducting mechanism itself in these lamellar copper oxides is still elusive, tremendous progress in condensed matter physics has resulted as a byproduct. One such example is our understanding of the two-dimensional quantum Heisenberg antiferromagnet (2DQHA) on a square lattice, which had been a long-standing problem in theoretical physics even before the discovery of high temperature superconductivity. Since the parent compounds of the copper oxide superconductors, such as La_2CuO_4, are very good representations of the $S = 1/2$ square-lattice Heisenberg antiferromagnet, the study of the 2DQHA has received renewed interest from both theorists and experimentalists [1]. The synergistic efforts of theoretical, numerical, and experimental investigations have yielded a comprehensive picture of the static magnetic properties of the 2DQHA over the past decade [2–4].

However, less progress has been made in studies of the dynamic properties of the 2DQHA. In particular, there are only a small number of analytic theoretical models and numerical simulations on this problem, while systematic experimental investigation is lacking for the $S = 1/2$ 2DQHA. One of the most fundamental questions we can ask in the study of dynamic critical behavior of the 2DQHA is whether or not dynamic scaling is obeyed. The dynamic scaling hypothesis proposed by Ferrell *et al.* [5] and developed for magnetic systems by Halperin and Hohenberg [6], is that the dynamic structure factor is completely determined by the static properties with the scaling frequency given by $\omega_0 \sim \xi^{-z}$. The theoretical value for the dynamic critical exponent $z = d/2$ of the Heisenberg antiferromagnet has been known for many years [7]. Using this dynamic scaling hypothesis, Chakravarty, Halperin, and Nelson (CHN) have argued that in the $S = 1/2$ 2DQHA the characteristic energy scale is given by $\omega_0 \propto \xi^{-1} c_0 (T/2\pi\rho_s^0)^{1/2}$, where c_0 is the spin-wave velocity and ρ_s^0 is the spin stiffness constant, both at zero temperature [2]. We set $\hbar = k_B = 1$, so that temperature T, and frequency ω have the units of energy.

The characteristic energy scale, ω_0, for the 2DQHA is equivalent to the relaxation rate of the order parameter, which can be determined by measuring the damping of the quasielastic peak at the antiferromagnetic wave vector (π, π) via inelastic neutron scattering. One of the main reasons for the scarcity of neutron experiments is the large energy scale of the magnetism in the copper oxide materials. In magnetic systems, the primary energy scale is set by the nearest neighbor exchange coupling J, and in La_2CuO_4 or $Sr_2CuO_2Cl_2$ (2122) this is about $J \sim 130$ meV. Although thermal neutron scattering is one of the most powerful tools in probing excitations in magnetic systems, it is not normally suitable for studying the physics at such a high energy scale. As a result, neutron scattering studies of the long-wavelength spin dynamics of $S = 1/2$ 2DQHA's such as La_2CuO_4 are very limited [8,9]. The recent discovery of the system $Sr_2Cu_3O_4Cl_2$ (2342), which contains a square lattice Cu_{II} subsystem that has an order of magnitude smaller J, has opened the door for a quantitative study of the spin dynamics of the $S = 1/2$ 2DQHA [10]. In this Letter, we report systematic measurements of ω_0 as a function of T/J in the model $S = 1/2$ 2DQHA's, 2122 and 2342. We show that dynamic scaling is obeyed over the entire temperature range probed in our experiment, $0.2 \lesssim T/J \lesssim 0.7$, which corresponds to the correlation length range $100 \gtrsim \xi/a \gtrsim 2$.

The neutron scattering experiments were carried out at the Center for Neutron Research at the National Institute of Standards and Technology. Large single crystal samples of both 2122 and 2342 were grown by the flux method; typical sample dimensions were $20 \times 20 \times 5$ mm^3, and the sample mosaicity was $\sim 0.2°$ half width at half maximum

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(HWHM). Various experimental configurations at the thermal beam line BT9 were employed in the measurements. Progressively coarser resolution was used to accommodate the wider energy width as the temperature was increased and, correspondingly, ξ decreased. Pyrolytic graphite filters were used to reduce the effects of $\lambda/2$ neutrons. The magnetic structure factor at (π, π) as a function of energy transfer, ω, was measured at various temperatures. The lower bound of the temperature range was governed by the Néel ordering temperature ($T_N \approx 256$ K for 2122 and $T_N \approx 40$ K for 2342), below which the spin dynamics is no longer relaxational at (π, π) [3,11]. Typical scans are shown in Fig. 1 for both 2122 and 2342. The energy resolution is also plotted in each panel as a horizontal bar.

To determine ω_0 these scans were fitted to the following form for the dynamic structure factor convoluted with the instrumental resolution function:

$$S(\mathbf{q}, \omega) = \frac{\omega}{1 - e^{-\omega/T}} \frac{S(0)}{1 + (q\xi)^2} \times \left[\frac{\Gamma_\mathbf{q}}{(\omega - cq)^2 + \Gamma_\mathbf{q}^2} + \frac{\Gamma_\mathbf{q}}{(\omega + cq)^2 + \Gamma_\mathbf{q}^2} \right]. \quad (1)$$

Here $\mathbf{q}$ is defined as the deviation in wave vector from the 2D antiferromagnetic position (π, π), and c is the spin wave velocity. We have used $\Gamma_\mathbf{q} \equiv \omega_0[1 + \mu(q\xi)^2]^{1/2}$ as proposed by Tyč, Halperin, and Chakravarty (THC) [12]. Our fits turned out to be insensitive to the choice of the phenomenological parameter μ, and, accordingly μ was set equal to zero. In Eq. (1), we define the characteristic energy scale as the HWHM of the quasielastic peak, that is, $\omega_0 \equiv \Gamma_{\mathbf{q}=0}$. It should be noted that Eq. (1) is a simplified version of the dynamic structure factor obtained by THC from their molecular dynamics simulation of a classical lattice rotor model [12]. Specifically, we ignore the logarithmic corrections ($\log[1 + (q\xi)^2]$) in the dynamic structure factor of THC, since we focus only on the small $\mathbf{q}$ regime.

We plot the results for ω_0/J versus the inverse correlation length ξ^{-1} in Fig. 2. The values of the correlation length in these systems have been determined previously with high accuracy [3,11]. Therefore, ω_0 and the overall amplitude $S(0)$ are the only adjustable parameters in the fits to Eq. (1). In order to compare the two different systems, as well as with the quantum Monte Carlo results, we scale ω_0 by J, and ξ by the lattice constant a. Each data set obtained with a different experimental setup is plotted in a different symbol. Filled and open symbols are used for 2122 and 2342, respectively. In their study of the spin dynamics of La_2CuO_4, Hayden and co-workers obtained

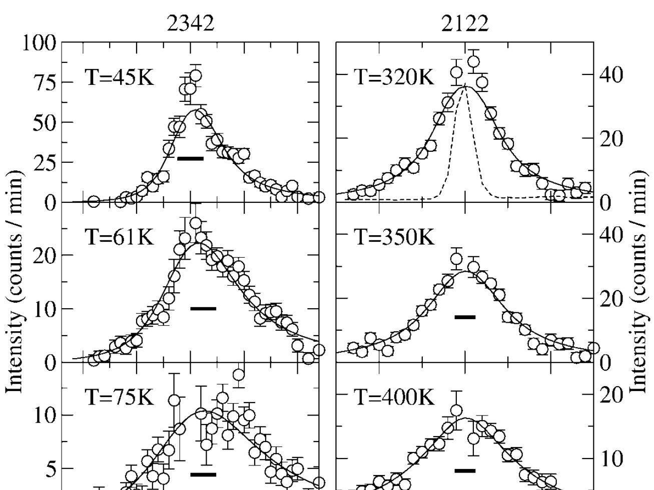

FIG. 1. Representative ω scans at the (π, π) position, which corresponds to (1/2 1/2 0) for both $Sr_2Cu_3O_4Cl_2$ and $Sr_2CuO_2Cl_2$, following the notation of Refs. [3,11]. The background from incoherent scattering, measured far away from the (π, π) position, has been subtracted from the raw data. A representative scan of the incoherent scattering, multiplied by 1/5, is shown as a dashed line in the top right panel. A fixed final neutron energy of 30.5 meV and collimations of 40′-48′-sample-44′-40′ have been used. The solid lines are the results of least square fits to Eq. (1) convoluted with the instrumental resolution function.

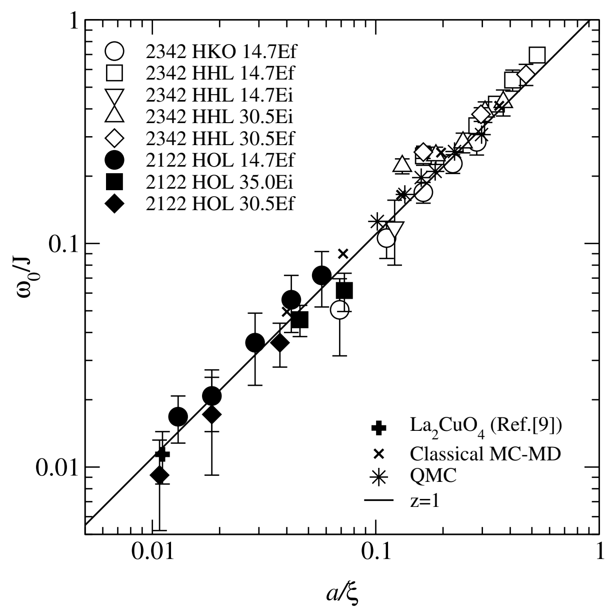

FIG. 2. Logarithmic plot of the reduced characteristic energy scale ω_0/J versus the inverse of reduced spin correlation length a/ξ. The open symbols are the data for $Sr_2Cu_3O_4Cl_2$ plotted with $J = 10.5$ meV, and the filled symbols are the data for $Sr_2CuO_2Cl_2$ plotted with $J = 125$ meV. Note that we use different symbols for each experimental configuration. Here HKO denotes the scattering plane in reciprocal space. We also plot the result of Ref. [9] for La_2CuO_4 at $T = 320$ K. The solid line is the dynamic scaling prediction with $z = 1$, that is, $\omega_0 \sim \xi^{-1}$. The results of the quantum Monte Carlo simulations of the $S = 1/2$ 2DQHA by Makivić and Jarrell [13] are plotted as $*$, while Wysin and Bishop's classical Monte Carlo result [14] are plotted as $\times$ with the energy scaled by $JS(S + 1)$.

ω_0 at $T = 320$ K [9], which is also plotted in Fig. 2. It is quite remarkable that the experimental results from all three systems fall on a single straight line without any adjustable parameter. This clearly shows the dynamic scaling behavior of ω_0. If we fit the data to the scaling form $\omega_0 \sim \xi^{-z}$, we obtain $z = 1.0(1)$, in excellent agreement with the theoretical value for the 2DQHA. The solid line is a fit of the experimental data to the scaling form with fixed $z = 1$: $\omega_0/J = 1.1(1)a\xi^{-1}$.

In order to compare our experimental results with the results of numerical simulations, Makivić and Jarrell's quantum Monte Carlo results [13] for the $S = 1/2$ 2DQHA are plotted in Fig. 2. Wysin and Bishop's Monte Carlo molecular dynamics calculation results for the classical 2D Heisenberg antiferromagnet are also plotted [14]. Not surprisingly, the quantum Monte Carlo results are in an excellent agreement with our neutron scattering results. However, it is interesting to note that the values from the classical Monte Carlo calculation also agree with our neutron scattering results within experimental error bars. Since Wysin and Bishop have calculated ξ as well as ω_0, there is no adjustable parameter except the $J \rightarrow JS(S+1)$ scaling. The good agreement between quantum and classical dynamics presumably reflects the fact that the zero temperature spin dynamics of the $S = 1/2$ 2DQHA are well described by the classical spin-wave picture with a uniform frequency renormalization of only 17%.

Two important comments regarding the data analysis are in order. First, due to the nonzero resolution width in both energy and momentum transfer, scattered neutrons with $\mathbf{q} \neq \mathbf{0}$ are invariably included in our data shown in Fig. 1. However, from numerical simulations, we have verified that the $\mathbf{q} \neq \mathbf{0}$ dynamical contribution to the total quasielastic scattering intensity for 2342 is insignificant and yields a correction to the energy width that is much smaller than the experimental error bars. On the other hand, 2122 has an order of magnitude larger spin wave velocity (820 meV Å) than that of 2342 (95 meV Å), and accordingly 2122 has a very steep dispersion relation. For 2122, the nonzero $\mathbf{q}$ dynamical contribution thus gives an apparent peak width in energy that is slightly larger than the intrinsic value. Although this is already taken into account in our fitting processes, relatively large error bars are generated for 2122 as a result. We have employed a number of different experimental configurations to ensure that the extracted ω_0 is intrinsic in both 2122 and 2342. Second, we have defined the characteristic energy scale as the HWHM of the dynamic structure factor at $\mathbf{q} = \mathbf{0}$. In Monte Carlo studies of Refs. [13] and [14], a different definition of ω_0 was used, and the values given in these studies are converted to fit our definition by multiplying by a constant. We have obtained this constant multiplication factor of 0.682 by fitting the raw data in Ref. [13] to Eq. (1).

The consistency among the experimental results and the numerical simulation results in *absolute* units gives strong credence to the combined data as a testing ground, against which various analytic theories can be examined. Since the pioneering work by CHN, there have been a number of theoretical studies to determine the characteristic energy scale of the 2DQHA. They all seem to indicate $z = 1$ dynamic scaling behavior of ω_0, but differ in the temperature dependence of R_ω, which is defined as a dimensionless ratio $R_\omega \equiv \omega_0\xi/c_0$. Specifically, $R_\omega \sim T^{1/2}$ is predicted by CHN, while Grempel [15] has carried out a conventional mode-mode coupling calculation and obtained the explicit prefactor of the $T^{1/2}$ dependence:

$$R_\omega \equiv \frac{\omega_0\xi}{c_0} = 1.3573\left(\frac{1}{\pi}\frac{1}{Z_c^2 Z_\chi}\frac{T}{J}\right)^{1/2} \approx 0.9\sqrt{\frac{T}{J}}\,. \qquad (2)$$

This result is close to the expression obtained by THC in their numerical simulation: $R_\omega = 0.85\sqrt{T/2\pi\rho_s} \approx 0.8\sqrt{T/J}$. However, Auerbach and Arovas, in their Schwinger boson mean field theory [16], have obtained a T-independent R_ω. Quantum Monte Carlo calculations by Makivić and Jarrell [13] as well as Nagao and Igarashi"s self-consistent theory [17] also show a T-independent R_ω, although these results are limited to a somewhat narrow temperature range.

In Fig. 3, we plot the dimensionless ratio R_ω obtained from our measurements as a function of reduced temperature. Note that c_0 for the $S = 1/2$ 2DQHA is well known from both Monte Carlo [4] and series expansion studies [18]: $c_0 = 1.657Ja$. Therefore, Fig. 3 also does not contain any adjustable parameter. Results from Monte Carlo calculations are also plotted in the figure, where the same symbols as in Fig. 2 are used. The analytic theory by Grempel, Eq. (2), is plotted as a solid line, while a fit to a presumed constant R_ω is plotted as a dashed line.

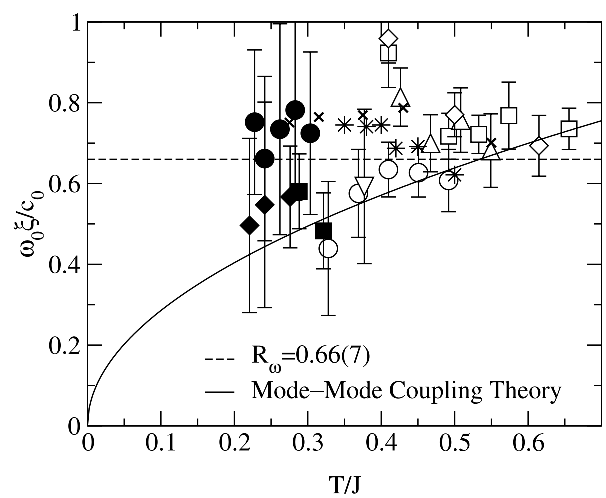

FIG. 3. The dimensionless ratio $R_\omega \equiv \omega_0\xi/c_0$ as a function of reduced temperature. The same symbols as in Fig. 2 are used in the plot. The solid line is the result from Grempel's mode-mode coupling theory, Eq. (2). The dashed line is a fit to a constant, $R_\omega = 0.66(7)$.

We should emphasize that the dominant T dependence of ω_0 is $\sim\xi^{-1}$, which has a strong exponential T dependence, so that Fig. 3 shows only the weak T dependence of the prefactor of the leading ξ^{-1} behavior. The salient feature to recognize in this figure is that the coupled mode theory of Grempel is in a reasonable agreement with our experimental results in *absolute* units. Considering that the mode-mode coupling approximation used by Grempel ignores vertex renormalizations, the agreement in this intermediate temperature range appears to be quite good. The large error bars on our 2122 data, however, prevent us from concluding that R_ω indeed has the predicted $\sqrt{T}$ dependence. A temperature independent R_ω describes our data equally well, if not better. Another analytic theory that predicts a T-independent R_ω is quantum critical theory [19]. However, quantitative comparison with our data is difficult, since the temperature dependence of ω_0 in the quantum critical regime is not known theoretically.

It turns out that the task of experimentally determining the functional form of R_ω is very difficult. Since $\sqrt{T}$ is a weak function of T, one has to probe at very low temperatures to distinguish between $\sqrt{T}$ and constant behavior. Although $Sr_2CuO_2Cl_2$ has the smallest T_N/J among model $S = 1/2$ 2DQHAs known to this date, its large nearest neighbor coupling makes a high-resolution neutron scattering study very difficult. To elucidate further the temperature dependence of the correction term R_ω, a quantum Monte Carlo study of the spin dynamics over a wide temperature range is highly desirable. It should be noted that in the bicritical $S = 5/2$ 2DQHA system Rb_2MnF_4, R_ω appears to depend strongly on T; indeed in that case the observed temperature dependence is much more rapid than $T^{1/2}$ [20].

In summary, a neutron scattering study of the long-wavelength dynamic spin correlations in the model $S = 1/2$ 2DQHA's $Sr_2CuO_2Cl_2$ and $Sr_2Cu_3O_4Cl_2$ has been presented. We have measured the characteristic energy scale as a function of temperature, and have shown that dynamic scaling is valid over a wide temperature range ($0.2 \lesssim T/J \lesssim 0.7$) with the dynamic critical exponent $z = 1$. This temperature range corresponds to the magnetic correlation length of $100 \gtrsim \xi/a \gtrsim 2$, for which dynamic scaling is expected to hold according to theoretical predictions. Our attempt to determine the weak temperature dependence of any possible corrections to this $z = 1$ scaling is, however, inconclusive, and remains to be addressed in future studies.

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