Spin-Spin Correlation in the Quantum Critical Regime of La_2CuO_4

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We report measurements of the ^{63}Cu nuclear spin echo decay rate $^{63}1/T_{2G}$ in the paramagnetic state of a quasi-two-dimensional antiferromagnet La_2CuO_4 up to 900 K, and from it deduce the temperature dependence of the spin-spin correlation length ξ. ξ shows a crossover from $\xi \sim \exp(J/k_BT)$ in the renormalized classical regime ($T \lesssim 600$ K) to $\xi \sim J/k_BT$ in the quantum critical regime ($T \gtrsim 600$ K), where J is the exchange interaction. Our finding that the ratio $^{63}T_1T/^{63}T_{2G}$ is temperature independent, where $^{63}T_1$ is the ^{63}Cu nuclear spin-lattice relaxation time, gives clear evidence for quantum critical scaling.

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The discovery of high-T_c superconductivity in doped La_2CuO_4 promoted strong interest in the physical properties of the undoped antiferromagnetic parent material because of its low dimensionality and strong quantum phenomena [1]. It is firmly established that the magnetism of La_2CuO_4 can be described very well based on the two-dimensional Heisenberg model with a nearest neighbor exchange interaction J (~ 1500 K) [2],

$$H = \sum_{i \neq j} J S_i S_j , \tag{1}$$

where S_i is a copper d spin ($S = \frac{1}{2}$) at a lattice site i. A weak intralayer coupling induces a three-dimensional antiferromagnetic ordering at $T_N \sim 300$ K. Neutron scattering experiments by the Brookhaven Collaboration carried out up to 540 K demonstrated that the copper spin-spin antiferromagnetic correlation length ξ shows a very strong temperature dependence even far above T_N [3]. Chakravarty, Halperin, and Nelson [4] showed theoretically that there are two regimes in the paramagnetic state of La_2CuO_4. One is *the renormalized classical regime*, $T_N < T \lesssim 2\rho_s$ ($\rho_s = 0.18J \sim 300$ K is the spin stiffness constant), where ξ is shown to satisfy an exponential temperature dependence [4,5]

$$\frac{\xi}{a} = \frac{0.498}{1 + 0.5(2\pi\rho_s)/T} \exp\left[\frac{2\pi\rho_s}{T}\right] . \tag{2}$$

a is the in-plane lattice constant (3.8 Å), k_B is the Boltzmann constant. The other is *the quantum critical regime* ($2\rho_s \lesssim T$), where ξ satisfies

$$\xi/a \sim c_Q \hbar c / k_B T . \tag{3}$$

c_Q is a constant of order of unity, $\hbar c = \sqrt{2} J a Z_c$ ($Z_c = 1.158$) is the spin wave velocity.

Very recently Imai *et al.* [6] reported that the ^{63}Cu nuclear spin-lattice relaxation rate $^{63}1/T_1$ in La_2CuO_4 satisfies a simple relation $^{63}1/T_1 \sim T^{1.5}\xi \sim T^{1.5}\exp(2\pi\rho_s/k_BT)$ in the renormalized classical regime as predicted by Chakravarty and Orbach [7]. Their observation, as well as neutron scattering experiments by Yamada *et al.* [8], strongly supports the dynamical scaling theory [4] employed by Chakravarty, Halperin, and Nelson to relate the dynamical structure factor $S(\mathbf{q},\omega)$ with the equal time correlation function $S(\mathbf{q})$, where $\mathbf{q}$ and ω are wave vector and frequency, respectively. The ^{35}Cl NMR experiments for a similar material $Sr_2CuO_2Cl_2$ by Borsa *et al.* also showed the validity of the dynamical scaling at low temperatures [9]. However, Imai *et al.* also found that $^{63}1/T_1$ levels off to $\sim 2.8 \times 10^3$ above 650 K, which clearly indicates the breakdown of the renormalized classical behavior. Moreover they found that even the Sr-doped high-temperature superconducting phase $La_{1.85}Sr_{0.15}CuO_4$ exhibits essentially the same behavior at high temperatures. Chubukov and Sachdev [10] pointed out that these results can be accounted for by quantum critical scaling [4,10] which relates $S(\mathbf{q},\omega)$ and $S(\mathbf{q})$ differently from the case of dynamical scaling. More recently Sokol and Pines [11] showed that quantum critical scaling can be verified if the ratio $[^{63}1/T_{2G}]/[^{63}1/T_1T] = {}^{63}T_1T/{}^{63}T_{2G}$ is temperature independent, where $^{63}1/T_{2G}$ is the Gaussian component of the ^{63}Cu nuclear spin echo decay rate, and used it as a base for their discussion of the unified picture of the phase diagram of La and Y cuprates.

Prior to the present work, there has been no experiment in the quantum critical regime of La_2CuO_4 other than our $^{63}1/T_1$ measurements [6] because of technical difficulties at high temperatures. In the present paper, we report the temperature dependence of $^{63}1/T_{2G}$ between 450 and 900 K, i.e., in *both* the renormalized classical and quantum critical regimes. As explained below, $^{63}1/T_{2G}$ is known to give quantitative information regarding $\chi'(\mathbf{q})$, the real part of the wave vector $\mathbf{q}$ dependent static spin susceptibility, and hence allows us to determine the correlation length ξ [12]. Our results are the first measurements of the temperature dependence of ξ in

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the quantum critical regime of La_2CuO_4. We test Eq. (3) for ξ in the quantum critical regime, and demonstrate that the linear temperature dependence of $1/\xi$ [4] with positive intercept [10], $1/\xi \sim T+\text{const}$, indeed holds. By comparing the results of ${}^{63}1/T_{2G}$ $[\sim\sqrt{\sum_{\mathbf{q}}\{\chi'(\mathbf{q})\}^2}$, see below] and ${}^{63}1/T_1$ $[\sim T\sum_{\mathbf{q}}\chi''(\mathbf{q},\omega_n)\sim\sum_{\mathbf{q}}S(\mathbf{q},\omega_n)$ [13], where $\chi''(\mathbf{q},\omega)$ is the imaginary part of the dynamical electron spin susceptibility and $\omega_n \sim 33$ MHz is the nuclear quadrupole resonance (NQR) frequency], we will also present an unambiguous experimental evidence that ${}^{63}T_1T/{}^{63}T_{2G}$ is temperature independent, as predicted by Sokol and Pines [11], so that $\chi''(\mathbf{q},\omega)$ and $\chi'(\mathbf{q})$ exhibit quantum critical scaling [4,10].

The ^{63}Cu NQR and NMR experiments were carried out utilizing the standard pulsed NMR spectrometer of our laboratory. In our previous study, a tiny amount of oxygen ($\delta \sim 0.0035$ per unit cell) was absorbed by the polycrystalline sample warmed up in air, resulting in *the motional narrowing effect* [14] above 600 K possibly due to the fast motion of the excess oxygen atoms. This did not allow us to measure the temperature dependence of ${}^{63}1/T_{2G}$ [6]. To avoid oxygen absorption, our new sample ($T_N = 308$ K) was sealed in a quartz vacuum tube whose diameter is 5 mm. The temperature dependence of the ^{63}Cu NQR frequency did not reveal any anomaly around 750 K, indicating that no absorption of the oxygen into the sample took place in the process of our measurements in contrast to our previous report. The refined data of ${}^{63}1/T_1$ measured by NQR showed perfect agreement with our published results [6]. Comparison of the $1/T_1$ measured for ^{63}Cu and ^{65}Cu isotopes indicated that the spin-lattice relaxation is completely dominated by magnetic relaxation processes in the entire temperature range.

The Gaussian component of the ^{63}Cu nuclear spin echo decay rate ${}^{63}1/T_{2G}$ was obtained by fitting the time evolution of the integrated intensity of the spin echo $M(2t)$ as a function of the separation time t between the exciting and the refocusing pulses to the following formula with two independent parameters M_0 and T_{2G},

$$M(2t) = M_0 \exp\left(\frac{-2t}{T_{2R}}\right)\exp\left[-\frac{(2t)^2}{2(T_{2G})^2}\right]. \quad (4)$$

The contribution of the spin-lattice relaxation processes to the spin echo decay, ${}^{63}1/T_{2R}$, was determined by use of the Redfield theory [14] as ${}^{63}1/T_{2R} = (\beta+R)({}^{63}1/T_1)$ [15]. The value of β is 2 and 3 for NQR and NMR measurements, respectively, while R is the anisotropy of ${}^{63}1/T_1$, $R = 3.6 \pm 0.2$. As shown in Fig. 1, we clearly observed a contribution of a Gaussian decay as reported previously for $YBa_2Cu_3O_7$ [16,17]. Our key result, the temperature dependence of ${}^{63}1/T_{2G}$ measured by NQR, is presented in Fig. 2. To facilitate comparisons, we converted our NQR data to the values expected for the NMR measurements of the $\frac{1}{2}$ to $-\frac{1}{2}$ transition by multiplying our raw data of ${}^{63}1/T_{2G}$ by a factor $1.07/\sqrt{2}$ [18].

As stressed by some of us before [17], ${}^{63}1/T_{2G}$ can be measured correctly only when the linewidth of the resonance line Δf is sufficiently narrow compared with the strength of the rf pulses H_1. The NQR technique can be applied to La_2CuO_4 merely because Δf, which is primarily dominated by the mechanism of *homogeneous broadening* [14], is exceptionally narrow [6]. In fact the temperature-dependent NQR linewidth Δf is an order of magnitude narrower at high temperatures ($\Delta f = 24 \pm 2$ kHz at 900 K) than that of high-temperature superconducting materials whose NQR linewidth is dominated by *inhomogeneous broadening* induced by disordering. Since Δf is nearly proportional to ${}^{63}1/T_{2G}$ and diverges with lowering temperature, we checked at 500 K that the fitted value of ${}^{63}1/T_{2G}$ does not change even if we reduce H_1 by 50%.

If only the nuclear dipole-dipole interaction contributes to ${}^{63}1/T_{2G}$, the results should be temperature independent with a much slower rate, ~ 1800 sec^{-1}. Pennington *et al.* [16] concluded from their single crystal NMR experiments for $YBa_2Cu_3O_7$ that the c-axis component of *the indirect nuclear spin-spin coupling* [14] dominates ${}^{63}1/T_{2G}$ in high-T_c cuprates. In such a case, Pennington and Slichter showed that ${}^{63}1/T_{2G}$ measured for the $\frac{1}{2}$ to

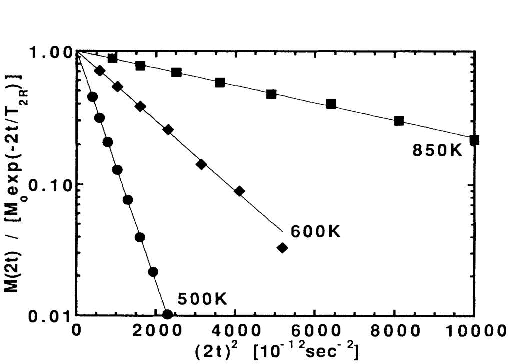

FIG. 1. The time evolution of the Gaussian component of the integrated intensity of ^{63}Cu NQR spin echo, $M(2t)/[M_0 \times\exp(-2t/T_{2R})]$. The straight line is the best fit by Eq. (4).

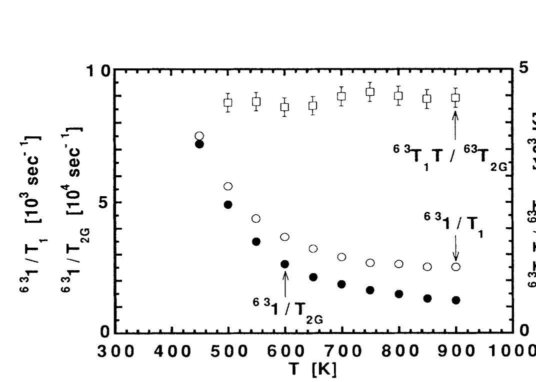

FIG. 2. ●: ${}^{63}1/T_{2G}$. ○: ${}^{63}1/T_1$. Experimental errors are about the size of the symbols. □: The ratio ${}^{63}T_1T/{}^{63}T_{2G}$.

$-\frac{1}{2}$ transition by NMR can be related to $\chi'(\mathbf{q})$ by the following relation [12,19]:

$$\left(\frac{1}{T_{2G}}\right)^2=\frac{0.69}{2\hbar^2}\left[\frac{1}{N}\sum_{\mathbf{q}}f_c(\mathbf{q})^4\chi'(\mathbf{q})^2-\left(\frac{1}{N}\sum_{\mathbf{q}}f_c(\mathbf{q})^2\chi'(\mathbf{q})\right)^2\right]. \quad (5)$$

The factor 0.69 originates from the natural abundance of ^{63}Cu. $f_c(\mathbf{q})=A_c+2B[\cos(q_xa)+\cos(q_ya)]$ is the form factor originating from the hyperfine interaction between the nuclear spin and the surrounding electron spins [20]. A_c and B are the c-axis component of the nearest neighbor and the transferred hyperfine interaction, respectively. The physical origin of the relation between $^{63}1/T_{2G}$ and $\chi'(\mathbf{q})$ results from the following: The hyperfine field that originated from a nuclear spin polarizes surrounding electron spins through nonlocal electron spin susceptibility $\chi'(\mathbf{r}',\mathbf{r})=(1/N)\sum_{\mathbf{q}}e^{i\mathbf{q}(\mathbf{r}'-\mathbf{r})}\chi'(\mathbf{q})$, and the polarized electron spins in turn interact with another nuclear spin. Since the range of $\chi'(\mathbf{r}',\mathbf{r})$ is the correlation length ξ, the temperature dependence of ξ can be determined by the measurement of $^{63}1/T_{2G}$. Since the form factor $f_c(\mathbf{q})$ is peaked at $\mathbf{q}=(\pi/a,\pi/a)$ in the present case [20], $^{63}1/T_{2G}$ is dominated by the short wavelength component of $\chi'(\mathbf{q})$. The observed monotonic increase of $^{63}1/T_{2G}$ in Fig. 2 therefore indicates that $\chi'(\mathbf{q})$ around the staggered wave vector $\mathbf{q}=(\pi/a,\pi/a)$ increases monotonically with lowering temperature due to the growing antiferromagnetic correlation. It is worthwhile noting that the long wavelength susceptibility $\chi'(\mathbf{q}=0)$ decreases in the same temperature range [1,2].

If one has an explicit form for $\chi'(\mathbf{q})$, Eq. (5) can be evaluated. If the functional form depends on parameters such as T and ξ, one can deduce the temperature dependence of ξ from the data of $^{63}1/T_{2G}$. Chakravarty and co-workers [4] showed that $\chi'(\mathbf{q})$ can be expressed by a nearly Lorentzian formula in the renormalized classical regime ($T\lesssim 600$ K),

$$\chi'(\mathbf{q})=\frac{g^2\mu_B^2N_0^2B_s\xi^2}{3k_BT(2\pi\rho_s/T+1)^2}\,\frac{1+(2\pi/B_s)\ln(1+\mathbf{q}^2\xi^2)}{1+\mathbf{q}^2\xi^2}, \quad (6)$$

where $B_s\approx 10^{1-2}$, $N_0\approx 0.31$, $g=2$, and μ_B is the Bohr magneton. A subsequent Monte Carlo study by Makivić and Jarrel [21] found Eq. (6) is valid even in higher temperatures with $B_s\approx 55$. More recently the numerical calculations by Glenister, Singh, and Sokol [22] based on the high-temperature expansion technique showed that, to a good approximation, a simple Lorentzian formula $\chi'(\mathbf{q})\approx\xi^2/(1+q^2\xi^2)$ reproduces their results in the quantum critical regime ($T\gtrsim 600$ K). Since the temperature-dependent coefficient $1/T(2\pi\rho_s/T+1)^2$ of Eq. (6) is nearly temperature independent (within $\sim 17\%$) between 600 and 900 K, we may use Eq. (6) in the entire temperature range of Fig. 2. We note that our definition of ξ is based on the width of static susceptibility $\chi'(\mathbf{q})$. Our definition coincides in the renormalized classical regime with that of neutron scattering experiments, in which ξ is defined as the width of the equal time correlation function $S(\mathbf{q})$, because $\chi'(\mathbf{q})=S(\mathbf{q})/k_BT$ holds in the limit of dynamical scaling.

In the inset to Fig. 3, we show a fit of $^{63}1/T_{2G}T$ by Eq. (6) using Eq. (2). We inserted the neutron value $2\pi\rho_s=1.13J=1730k_B$ ($J=0.132$ eV [23]), and normalized the integral by picking $B_s=39$ to fit the value of $^{63}1/T_{2G}$ below 600 K. The deviation from the renormalized classical behavior makes the fit increasingly poor in the quantum critical regime above 600 K. Plotted in the main panel of Fig. 3 is the temperature dependence of a/ξ determined by using Eqs. (5) and (6) for the value of B_s determined by Monte Carlo calculations $B_s=55$ [21]. Essentially all that is assumed here is a Lorentzian-like $\mathbf{q}$ dependence. The results do not change appreciably even if we neglect the log term in Eq. (6) as shown in Fig. 3. A different choice for the value of B_s does not change the qualitative aspects of the temperature dependence of a/ξ in Fig. 3. Evidently, the results in the renormalized classical regime agree very well with neutron data by Keimer *et al.* [3]. Above 600 K, the observed temperature dependence of a/ξ can be approximated very well by $a/\xi=\zeta(T-T_0)$, where $T_0\sim 400$ K and $\zeta\sim 2\times 10^{-4}$. This T-linear behavior with positive intercept is consistent with the theoretical prediction based on the quantum critical scaling theory [4,10] and high-temperature expansion [22]. We also note that the first-principles calculations in Ref. [22] reproduce our data of $^{63}1/T_{2G}$ *quantitatively* without any adjustable parameters.

Another striking feature of our experimental results may be seen in Fig. 2, where we plotted the ratio of $^{63}1/T_1T$ and $^{63}1/T_{2G}$. We find that $^{63}T_1T/^{63}T_{2G}\approx 4.2\times 10^3$ K is temperature independent to a high degree of

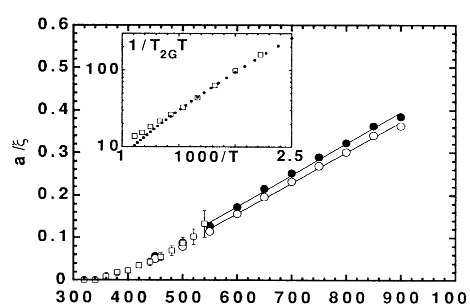

FIG. 3. ●: The temperature dependence of a/ξ determined by $^{63}1/T_{2G}$ using Eq. (6) for $B_s=55$ and $B_f=0.23$. ○: a/ξ when the log term of Eq. (6) is neglected (i.e., for $B_s=55$ and $B_f=0$). □: a/ξ determined by neutron scattering (after Ref. [3]). Solid lines are the best fits by $a/\xi=\zeta(T-T_0)$. Inset: □, $^{63}1/T_{2G}T$ (sec^{-1}K^{-1}) as a function of $1000/T$ (K^{-1}). The dotted curve is a fit by Eq. (6) with exponentially temperature-dependent ξ described by Eq. (2).

precision between 450 and 900 K. This indicates that there is a scaling relation between $\chi'(\mathbf{q})$ and $\chi''(\mathbf{q},\omega)$ as discussed below. The results, however, disagree with the prediction of the dynamical scaling theory, $^{63}1/T_1T \sim T^{1.5}\xi$ and $^{63}1/T_{2G} \sim T\xi$ [4,7], hence $^{63}T_1T/^{63}T_{2G} \sim T^{0.5}$. On the other hand, Sokol and Pines [11] predicted very recently that

$$^{63}T_1T/^{63}T_{2G} = \text{const} + O(\rho_s/T)\,, \tag{7}$$

based on the quantum critical scaling theory [4,10]. Sokol and Pines also showed using the finite cluster method that the quantity $^{63}T_1T/^{63}T_{2G} \sim 4.3\times10^3$ K is nearly temperature independent around $T \sim 750$ K. This means that the correction term $O(\rho_s/T)$ in Eq. (7) is actually very small, and the temperature independence of $^{63}T_1T/^{63}T_{2G}$ is a criterion for quantum critical scaling. We therefore conclude that the quantum critical scaling holes in the quantum critical regime ($T \gtrsim 600$ K) of La_2CuO_4. The fact that $^{63}T_1T/^{63}T_{2G}$ is temperature independent down to 450 K suggests that the crossover from the renormalized classical regime [$^{63}T_1T/^{63}T_{2G} \sim T^{0.5}$ and $\xi \sim \exp(2\pi\rho_s/T)$ at $T \ll 2\rho_s$, where the dynamical scaling hypothesis should hold] to the quantum critical regime ($^{63}T_1T/^{63}T_{2G} \sim$ const and $\xi \sim 1/T$ at $T \gtrsim 2\rho_s$) takes place smoothly in the interval of a broad temperature range.

In conclusion, we reported the temperature dependence of the ^{63}Cu nuclear spin echo decay rate $^{63}1/T_{2G}$ in *both* the renormalized classical and quantum critical regimes of a quasi-two-dimensional antiferromagnet La_2CuO_4. The temperature dependence of the antiferromagnetic correlation length ξ was determined from the data of $^{63}1/T_{2G}$. The results in the renormalized classical regime agreed with neutron scattering data. The temperature dependence of ξ showed a crossover above 600 K from an exponential law to a quantum critical behavior, $1/\xi \sim T+$const. Moreover the ratio $^{63}T_1T/^{63}T_{2G}$ was found to be temperature independent. These results unambiguously establish that the high-temperature properties of La_2CuO_4 can be explained by quantum critical scaling.

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