^{17}O NMR Study of Undoped and Lightly Hole Doped CuO_2 Planes

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Using ^{17}O NMR, we probed the short wavelength excitations in the CuO_2 planes of insulating and weakly metallic high T_c cuprates. We measured the spin wave damping for an $S = 1/2$ 2D quantum Heisenberg antiferromagnet for the first time. The results establish the nearly free behavior (*asymptotic freedom*) of the high energy spin waves, even without long range magnetic order. Light hole doping dramatically enhances the low energy excitation spectrum below 300 K. [S0031-9007(97)03483-2]

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The discovery of high T_c superconductivity in hole doped CuO_2 planes has focused strong interest on the spin $S = 1/2$ two-dimensional (2D) Heisenberg antiferromagnet, $H = J\sum \mathbf{S}_i \cdot \mathbf{S}_j$, where J ($\approx$1500 K) is the exchange interaction [1]. Previous experimental studies of the 2D Heisenberg model by ^{63}Cu NMR [2] and neutron scattering [3] have concentrated on the sharp peak of the dynamical structure factor $S(\mathbf{q}, \omega)$ located at the corners of the first Brillouin zone, $\mathbf{Q} = (\pm\pi/a, \pm\pi/a)$, where $\mathbf{q}$ is the wave vector, ω is the frequency, and a is the lattice constant. This antiferromagnetic peak has a momentum width $\sim 1/\xi$, the inverse of the spin correlation length. Accordingly, these experiments have probed the long wavelength properties ($\gtrsim \xi$) near $\mathbf{q} = \mathbf{Q}$.

On the other hand, virtually nothing is known experimentally at short wavelengths ($\ll \xi$) in the paramagnetic state, except for the dispersion of the high energy spin excitations [4]. In 2D, the high energy spin waves can exist even without long range magnetic order because of the strong short range spin correlations for $T \ll J$ [1]. Theoretically, the renormalization group analysis of the nonlinear sigma model by Chakravarty, Halperin, and Nelson predicted that the spin waves in the 2D Heisenberg antiferromagnet behave as *noninteracting free particles* at short distance scales even in the paramagnetic state [5]. This intriguing prediction for 2D has a close analogy in QCD known as the *asymptotic freedom* of quarks [6] and is in remarkable contrast to the 3D system, in which the spin wave excitations are highly damped above the bulk Néel ordering temperature T_N^{3d} [7]. Unfortunately, regardless of the strong theoretical interest, many believed that probing the nature of the short wavelength elementary excitations was *beyond the current experimental technology* [1,8(a)]. Among the unresolved questions are how these spin waves are damped by thermal effects [8] and how mobile holes doped in the CuO_2 planes contribute to the elementary excitations [9].

To shed light on these issues from experiments, we report in this Letter an ^{17}O NMR study of magnetic excitations in undoped and lightly hole doped (weakly metallic) CuO_2 planes of high T_c cuprates in an extremely broad range of temperature ($0.2 \leq T/J \leq 0.5$). We deduce, for the first time, the magnon damping, Γ, of the $S = 1/2$ 2D Heisenberg antiferromagnet at short wavelengths, and test theoretical predictions [5,8]. Moreover, we demonstrate the progressive evolution of the low-energy quasiparticle excitation spectrum across the insulator-metal transition with hole doping.

As an experimental system for the undoped 2D Heisenberg model, either ^{17}O isotope enriched single crystals, uniaxially aligned powder, or partially aligned powder samples of $Sr_2CuO_2Cl_2$ [3] were used depending on experimental necessities. We determined the bulk Néel temperature as $T_N^{3d} = 257$ K based on ^{35}Cl NMR measurements, in agreement with Suh *et al.* [10]. The major advantage of $Sr_2CuO_2Cl_2$ is that the magnetic anisotropy is so weak ($J_{xy}/J \sim 10^{-4}$ [3]) that the isotropic 2D Heisenberg behavior is robust even down to $\approx$280 K (see Fig. 1 and [3,10]). For the study of lightly hole doped CuO_2 planes, we used ^{17}O isotope enriched single crystals of $La_{2-x}Sr_xCuO_4$ with $x = 0.025$ and 0.035 [11]. All ^{17}O NMR measurements were conducted for clearly resolved NMR transitions with the typical line width 2 to 15 KHz.

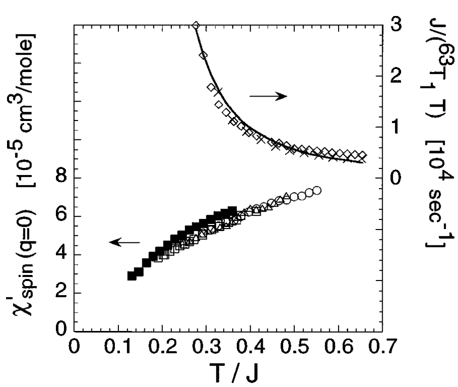

FIG. 1. Temperature dependence of uniform spin susceptibility $\chi'(\mathbf{q} = \mathbf{0})$ (left axis) for $Sr_2CuO_2Cl_2$ from $^{17}K_{bond}$ (□), $^{63}K_{ab}$ (○), and bulk measurement (△), and for $La_{1.965}Sr_{0.035}CuO_4$ from $^{17}K_{bond}$ (■). ^{63}Cu $J/(^{63}T_1T)$ measured by NQR (right axis) is shown for $Sr_2CuO_2Cl_2$ (◇, this work) and La_2CuO_4 (×, [2]). The solid line shows theoretical results based on Eq. (5).

In what follows, we will deduce information regarding spin waves and electron-hole pair excitations based on the measurements of the nuclear spin-lattice relaxation rate $1/T_1$. $1/T_1$ is related to $S(\mathbf{q},\omega) = \chi''(\mathbf{q},\omega)/[1 - \exp(-\hbar\omega/k_B T)]$, where $\chi''(\mathbf{q},\omega)$ is the imaginary part of the dynamical electron spin susceptibility, as [12]

$$^{17,63}\left(\frac{1}{T_1}\right) = \frac{^{17,63}\gamma_n^2}{\mu_B^2\hbar} \sum_{\mathbf{q}} {}^{17,63}F(\mathbf{q})^2 S(\mathbf{q},\omega_n)\,, \qquad (1)$$

where ω_n is the NMR frequency, γ_n is the nuclear gyromagnetic ratio. The quantization axis of our measurements of $1/T_1$ is along the crystal c axis. $^{17,63}F(\mathbf{q})$ is the wave vector dependent *hyperfine form factor* [13]

$$^{17}F(\mathbf{q})^2 = 4(C_{\text{bond}}^2 + C_{\text{perp}}^2)\cos^2(q_x a/2)\,, \qquad (2a)$$

$$^{63}F(\mathbf{q})^2 = 2\{A_{ab} + 2B[\cos(q_x a) + \cos(q_y a)]\}^2 \qquad (2b)$$

for the planar ^{17}O [Eq. (2a)] and ^{63}Cu [Eq. (2b)] sites. The hyperfine interaction tensors, A_α, B, and C_α, were determined from measurements of the NMR shift and uniform susceptibility as described below. The subscript, $\alpha =$ bond, c, or perp, refers to the direction of the magnetic field, along the Cu-O bond axis, along the crystal c axis, or perpendicular to both the bond and c axis, respectively. Since $S(\mathbf{q},\omega)$ is the space-time Fourier transform of the spin-spin correlation function, and the NMR frequency is very low ($\hbar\omega_n/k_B \approx 1$ mK), $1/T_1$ probes the $\mathbf{q}$ summation of the low energy part of the elementary excitation spectrum, or slow spin dynamics. In Fig. 1, we present the uniform spin susceptibility $\chi'(\mathbf{q}=\mathbf{0})$ deduced from the measurements of bulk susceptibility and ^{17}O NMR shifts [14],

$$^{17}K_\alpha = \frac{2C_\alpha}{N_A\mu_B}\chi'(\mathbf{q}=\mathbf{0}) + {}^{17}K_{\alpha,\text{chem}}\,. \qquad (3)$$

$^{17}K_{\alpha,\text{chem}}$ is the temperature independent chemical shift, and determined in $YBa_2Cu_3O_{6.63}$ to be 0.04% for the Cu-O bond axis ($\alpha =$ bond) [15]. At $T/J > 0.35$, we also deduced $\chi'(\mathbf{q}=\mathbf{0})$ from ^{63}Cu NMR shifts $^{63}K_{ab}$ in the ab plane by matching the results with $^{17}K_{\text{bond}}$ between 425 and 550 K. We also found that the anisotropy of ^{17}K in $Sr_2CuO_2Cl_2$ and $La_{2-x}Sr_xCuO_4$ (not shown) is very close to that in $YBa_2Cu_3O_x$ [15]. Applying the standard K-χ plot analysis [14] to $Sr_2CuO_2Cl_2$, we determined $C_{\text{bond}} = 83 \pm 7$, $C_c = 40 \pm 4$, $C_{\text{perp}} = 53 \pm 11$ kOe/μ_B. We also obtained $A_{ab} = 31 \pm 45$, $A_c = -84 \pm 36$, $B = 37 \pm 8$ kOe/μ_B, and $^{63}K_{ab,\text{chem}} = 0.27\%$.

In Figs. 1 and 2, we present the results of $^{63}1/(T_1T)$ and $^{17}1/(T_1T)$ measured for the ^{63}Cu and ^{17}O sites. To facilitate the quantitative comparison possible between $Sr_2CuO_2Cl_2$ ($J_{2122} = 1450$ K [16]) and Sr-doped La_2CuO_4 ($J_{214} = 1530$ K [4]) independent of slightly different values of J, the values of $1/(T_1T)$ are multiplied by J, because at finite temperatures, $1/(T_1T) \propto 1/J$ [17]. The most striking feature is that $^{17}1/T_1$ and $^{63}1/T_1$ show completely different dependencies on temperature. Because of the critical slowing down of the long wavelength, antiferromagnetic spin fluctuations at $\mathbf{q} \approx \mathbf{Q}$, $^{63}1/T_1$ for the undoped 2D Heisenberg antiferromagnet diverges exponentially toward $T = 0$ $(= T_N^{2D})$ following $^{63}1/T_1 \approx T^{1.5}\exp(1.13J/T)$ [2,18]. Light hole doping mildly suppresses the divergent behavior [2]. On the other hand, since $^{17}F(\mathbf{q}=\mathbf{Q}) = 0$, $^{17}1/T_1$ is insensitive to the long wavelength critical dynamics around $\mathbf{q}=\mathbf{Q}$ [13], and senses the short wavelength mode ($1/\xi \lesssim \mathbf{q} < \mathbf{Q}$) of $S = 1/2$ Cu spin fluctuations. Also, ^{89}Y $1/T_1$ as reported by Ohno *et al.* [19] for $YBa_2Cu_3O_6$ should be sensitive to the short wavelength spin fluctuations and appears to be qualitatively consistent with this picture over their limited temperature range.

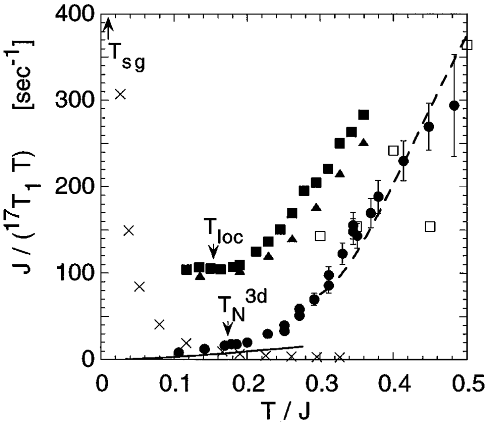

FIG. 2. Temperature dependence of ^{17}O $J/(^{17}T_1T)$ at 9 T for undoped $Sr_2CuO_2Cl_2$ (•), and doped $La_{2-x}Sr_xCuO_4$ for planar oxygen sites ($x = 0.025$ ▲, $x = 0.035$ ■) and apical oxygen sites ($x = 0.035$, ×). We also show theoretical predictions for the undoped 2D Heisenberg antiferromagnet by low T spin wave theory (solid line, [18]), high T expansion (dashed line, [17]), and Monte Carlo (□, [20]).

According to the renormalization group analysis, the elementary excitations at short wavelengths in 2D Heisenberg antiferromagnets are spin waves with asymptotic freedom [5]. This property allowed Chakravarty *et al.* to use a spin wave expansion to calculate $^{17}1/T_1$ at $T_N < T \ll J$ as [18,20]

$$^{17}\left(\frac{1}{T_1}\right) = \frac{2\pi C^2 a}{3\hbar^2 c}\left(\frac{Ta}{\hbar c}\right)^3\left[1 + C_o^{(2)}\left(\frac{T}{2\pi\rho_s}\right) + O(T^2)\right], \qquad (4)$$

where $c = \sqrt{2}Z_c Ja/\hbar$ is the spin wave velocity with $Z_c \approx 1.18$, and $C_o^{(2)} \approx -1.88$ [20]. This theoretical prediction is shown in Fig. 2 (solid line) together with a high temperature expansion (dashed line) by Singh and Gelfand [17] and Monte Carlo results (open squares) by Sandvik and Scalapino [21]. These parameter-free theoretical predictions agree well with our data.

Our results of $^{17}1/T_1$ indicate that free spin waves are indeed a good description of the quasiparticle excitations at short wavelengths even at $T \gtrsim T_N^{3d}$. Then, we can deduce the effective thermal damping Γ of the spin waves at finite temperatures from ${}^{17}1/T_1$ as follows. For damped spin waves, we can express $\chi''(\mathbf{q}, \omega)$ as

$$\chi''(\mathbf{q}, \omega) = \chi'(\mathbf{q})\left(\frac{\omega\Gamma(\mathbf{q})}{[\omega - \omega(\mathbf{q})]^2 + \Gamma(\mathbf{q})^2} + \frac{\omega\Gamma(\mathbf{q})}{[\omega + \omega(\mathbf{q})]^2 + \Gamma(\mathbf{q})^2}\right), \quad (5)$$

where $\omega(\mathbf{q}) = 2JZ_c\sqrt{1 - [\cos(q_x a) + \cos(q_y a)]^2/4}$ is the spin wave dispersion ($\gg \omega_n$), and $\Gamma(\mathbf{q})$ represents the spin wave damping [4,8]. The $\mathbf{q}$ dependence of $\chi'(\mathbf{q})$ is known analytically [22], and we normalize the absolute value of $\chi'(\mathbf{q})$ using the results in Fig. 1. Then the only unknown parameter in Eq. (1) is $\Gamma(\mathbf{q})$. However, Monte Carlo results by Makivic and Jarrell [8(c)] and analytic results by Kopietz [8(b)] indicate that $\Gamma(\mathbf{q})$ shows little dependence on the wave vector $\mathbf{q}$ in most of the Brillouin zone except near $\mathbf{q} = \mathbf{0}$ and $\mathbf{Q}$. Therefore, we let $\Gamma(\mathbf{q}) = \Gamma$ be independent of $\mathbf{q}$ except for the regions near $\mathbf{q} = \mathbf{0}$ and $\mathbf{Q}$, and consider Γ as the wave vector averaged damping at short wavelengths. The $\mathbf{q}$ dependence of $\Gamma(\mathbf{q})$ used for the calculation is shown schematically in the inset to Fig. 3. Near $\mathbf{q} = \mathbf{Q}$ where the dynamical scaling form of the damping $\gamma(\mathbf{q})$ [23] satisfies $\gamma(\mathbf{q}) < \Gamma$, we let $\Gamma(\mathbf{q}) = \gamma(\mathbf{q})$. This guarantees that *the same form of* $\chi''(\mathbf{q}, \omega)$ used to fit ${}^{17}1/T_1$ reproduces the low temperature behavior ($T \ll J$) of ${}^{63}1/T_1$ as shown in Fig. 1, consistent with the earlier finding for La_2CuO_4 [2]. For the region near $\mathbf{q} = \mathbf{0}$, we neglect the contribution from $q < 1/\xi$ [24]. Monte Carlo calculations by Sandvik and Scalapino [21] indicate that this contribution is $\lesssim 25\%$ at $T = 0.5J$ and decreases with lowering temperature ($\lesssim 10\%$ at $T = 0.4J$). Including this contribution would lower our calculated value of the damping Γ by the amount of the contribution (but within the error bars). Essentially, we are measuring the damping Γ by the fact that the size of the tail of $\chi''(\mathbf{q}, \omega)$ at $\omega = \omega_n \approx 0$ is determined by the energy width Γ. For the short wavelength region, the contribution to ${}^{17}1/(T_1T)$ is $\propto \Gamma$. We note that the deviation of $\chi''(\mathbf{q}, \omega)$ from the Lorentzian form of Eq. (5) can change our estimate of the magnitude of Γ [25], but this does not affect the conclusions of this paper.

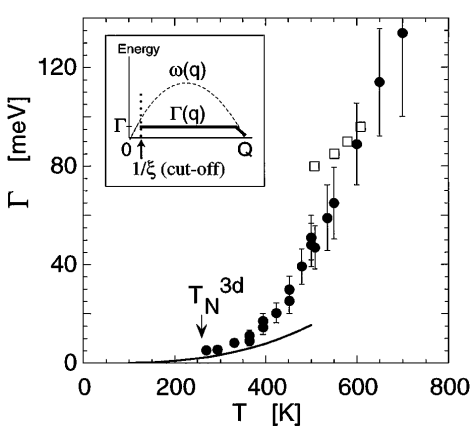

FIG. 3. Temperature dependence of the spin wave damping, Γ (•). The solid line indicates a low temperature expansion by Kopietz [8(b)], and the squares show Monte Carlo results of Makivic and Jarrell (□) [8(c)]. The inset shows the $\mathbf{q}$ dependence of $\Gamma(\mathbf{q})$ used in Eq. (5).

We plot the temperature dependence of Γ deduced from Eq. (5) in Fig. 3. We emphasize two key findings. First, the damping Γ is smaller than the highest excitation energy $\omega(\mathbf{q}) \approx 0.3$ eV [4] of the short wavelength magnons in the entire temperature range we studied. This observation establishes that magnons are well defined (long lifetime) elementary excitations for short wavelengths even in the paramagnetic state up to $T \approx 0.4J$ in $S = 1/2$ 2D quantum Heisenberg antiferromagnets. Second, the value of Γ is in good agreement with theoretical predictions based on low temperature analytic calculations, $\hbar\Gamma \sim 3J(T/J)^3$ [8(b)], and high temperature Monte Carlo simulations [8(c)].

Next, we would like to address another fundamental question: how do the elementary excitations evolve when we dope holes into the CuO_2 planes and transform the system to metallic behavior? The measurement of ${}^{17}1/T_1$ is best suited to answering this question, because ${}^{17}O$ NMR is efficient in probing the electron-hole pair excitations at the Fermi surface, while ${}^{63}1/T_1$ is dominated by the divergently large, long wavelength collective spin dynamics at $\mathbf{q} \approx \mathbf{Q}$. The results of ${}^{17}1/(T_1T)$ in hole doped $La_{2-x}Sr_xCuO_4$ are compared with the results of undoped $Sr_2CuO_2Cl_2$ in Fig. 2. At these doping levels, previous studies established that the resistivity is linear in temperature down to T_{loc} (<300 K) [26,27], where holes begin to localize, followed by the spin-glass transition at T_{sg} (≈ 10 K) [28]. We identified T_{loc} as ≈ 250 K from the onset of the dramatic increase of ${}^{17}1/T_{1,apex}$ at the apical oxygen sites. At T_{sg}, ${}^{17}1/T_{1,apex}$ diverged (see Fig. 2 for the results observed for $x = 0.035$). In what follows, we will focus our attention on the temperature dependence of ${}^{17}1/(T_1T)$ at the planar site above T_{loc}, where carriers are mobile and resistivity is roughly proportional to temperature.

The temperature dependence of ${}^{17}1/(T_1T)$ is surprisingly similar for the undoped and lightly hole doped samples, but with a weakly temperature dependent increase of ${}^{17}1/(T_1T)$ for the doped samples. We estimate this increase as ${}^{17}1/(T_1T)_{e\text{-}h} = 0.05$–$0.065\ \mathrm{sec}^{-1}\,\mathrm{K}^{-1}$ and 0.06–$0.085\ \mathrm{sec}^{-1}\,\mathrm{K}^{-1}$ for $x = 0.025$ and 0.035, respectively. It is also interesting to recall that Reven *et al.* observed ${}^{17}1/(T_1T) \approx 0.4\ \mathrm{sec}^{-1}\,\mathrm{K}^{-1}$ at $T \lesssim 300$ K in optimally doped $La_{1.85}Sr_{0.15}CuO_4$ [29]. The increase in ${}^{17}1/(T_1T)$ appears to be proportional to the amount of hole doping x. Angle resolved photo emission experiments for undoped $Sr_2CuO_2Cl_2$ by Wells *et al.* show that the highest occupied band is peaked at $\mathbf{q} = (\pm\pi/2a, \pm\pi/2a) = \mathbf{Q}/2$

with a bandwidth $(2.2 \pm 0.5)J$ [30]. Therefore, the simplest interpretation is that the electron-hole pair (Stoner) excitations with wave vectors, $(0,0)$, $(\pm\pi/a,0)$, $(0,\pm\pi/a)$, $(\pm\pi/a,\pm\pi/a)$, connecting the hole pockets at $\mathbf{Q}/2$ give rise to an additional contribution to $^{17}1/(T_1T)$ without changing the spin wave contribution. However, our calculations based on the rigid band picture showed that the Stoner continuum is very close to the spin wave dispersion over most of the Brillouin zone except near $\mathbf{q} = (\pm\pi/a,0)$ and $(0,\pm\pi/a)$ because the width of the spin wave dispersion $\approx 2.36J$ is within experimental error equal to the bandwidth. The conventional wisdom for the spin waves in metals is that the electron-hole pair excitations damp the spin waves if the dispersion of the spin wave merges into the Stoner continuum [31]. This suggests that the magnons and the electron-hole pair excitations will interact strongly, perhaps resulting in complete renormalization of the damping, $\Gamma(\mathbf{q})$. We note that the temperature T and doping x dependence of $^{17}1/(T_1T)$ is quite similar to that of $1/\xi(T,x)$ observed by Keimer *et al.* [27]. Knowing that $^{17}1/(T_1T) \propto \Gamma$, this suggests that the renormalized quasiparticle damping, $\Gamma(T,x)$, is related to $\xi(T,x)$ as $\Gamma(T,x) \propto c/\xi(T,x)$.

To summarize, we deduced the effective damping Γ of the short wavelength magnons of the $S = 1/2$ 2D Heisenberg antiferromagnet in a broad range of temperature $(0.2 \leq T/J \leq 0.5)$, contrary to the prevailing perception in the community that Γ was not measurable with current technology. Our results establish the asymptotic freedom of paramagnetic spin waves in 2D, and the temperature dependence of the damping. The low-energy excitations in the hole doped, weakly metallic CuO_2 planes show a similar temperature dependence to the undoped sample, but with a weakly temperature dependent increase from the addition of electron-hole pair excitations. We suggest that the spin waves may interact strongly with electron-hole pair excitations.

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