

Nuclear relaxation in the spin-1/2 antiferromagnetic chain compound Sr_2CuO_3 : Comparison between theories and experiments

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The NMR relaxation data on Sr_2CuO_3 [Phys. Rev. Lett. **76**, 4612 (1996)] are reexamined and compared with the analytic theory of the dynamic susceptibility in the $S=1/2$ antiferromagnetic Heisenberg chain including multiplicative logarithmic corrections [Phys. Rev. Lett. **78**, 539 (1997)]. Comparisons of the spin-lattice and the Gaussian spin-echo decay rates ($1/T_1$ and $1/T_{2G}$) and their ratio all show good quantitative agreement. Our results demonstrate the importance of the logarithmic corrections in the analysis of experimental data for quasi-one-dimensional systems and indicate that the dynamics of Sr_2CuO_3 is well described by an $S=1/2$ one-dimensional Heisenberg model with a nearest-neighbor exchange. [S0163-1829(97)02741-0]

The spin-1/2 one-dimensional antiferromagnetic Heisenberg model

$$\mathcal{H} = J \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+1} \quad (1)$$

continues to reveal novel many-body quantum effects^{1,2} in spite of its simplicity and extensive studies over many decades. In particular, a good understanding of the dynamic properties at finite temperature has been obtained only recently, using field theories^{3,1,4,5} and advanced numerical techniques.⁶ Experimentally, there are generally two complementary methods to observe spin dynamics. The spin correlation function over a wide frequency (ω) and momentum (q) range can be measured by neutron scattering experiments.⁷ The nuclear spin-lattice and the Gaussian spin-echo decay rates ($1/T_1$ and $1/T_{2G}$) measured by nuclear magnetic resonance (NMR) experiments⁸ provide accurate information on q -averaged low frequency dynamic and static susceptibilities, respectively. Since it is the static and low frequency ($\hbar\omega \ll J$) properties at low temperatures ($T \ll J$) that can be treated most accurately by field theories, NMR is particularly suitable to test them.

A remarkable consequence of the field theories is the quantum critical scaling behavior,³ i.e., scaling of the dynamic susceptibility $\chi(q, \omega, T)$ in the variables $c(q - \pi)/T$ and $\hbar\omega/T$ ($c = \pi J/2$ is the spinon velocity). This implies a divergence of the antiferromagnetic correlation length and the characteristic time scale of spin fluctuations as $1/T$ at low temperatures and reflects the critical nature of the ground state. The scaling determines the temperature dependences of the NMR relaxation rates as $1/T_1 = \text{const}$ and $1/T_{2G} \propto 1/\sqrt{T}$.⁴ This behavior has indeed been observed in the experiments

on Sr_2CuO_3 ,⁸ which has the most ideal one-dimensional character known to date.⁹⁻¹¹ The theory also shows that the scaling is correct only approximately and there is a multiplicative correction with logarithmic temperature dependence.^{12,13} However, including this correction to the highest order actually degrades the agreement between theory and the data on Sr_2CuO_3 .⁸

Very recently, an improved analytic theory of the logarithmic corrections has been proposed by Starykh, Singh, and Sandvik,⁵ generalizing the scaling ansatz and the conformal mapping to include the logarithmic factors. In this paper, we reexamine the NMR data on Sr_2CuO_3 reported in Ref. 8 and compare them with this theory. In the following, we first describe the theoretical results for the NMR relaxation rates both with and without the logarithmic corrections. We then reanalyze the spin-echo decay data to extract $1/T_{2G}$ more accurately, taking the nuclear spin fluctuations due to spin-lattice relaxation into account, and also examine the appropriate value of J in Sr_2CuO_3 . From the comparison, we found that including the logarithmic corrections considerably improves the quantitative agreement.

Let us start with the expression for $1/T_1$ (Ref. 14) and $1/T_{2G}$ (Refs. 15-17) due to the magnetic hyperfine interaction

$$\mathcal{H}_{\text{hf}} = \sum_{\alpha, i, j} A_{\alpha}^{ij} I_{i\alpha} S_{j\alpha} \quad (\alpha = a, b, \text{ and } c) \quad (2)$$

between a nuclear spin at i site and an electron spin at j site,

$$\frac{1}{T_1} = \frac{k_B T}{\hbar^2} \int \frac{dq}{2\pi} \{A_a^2(q) + A_b^2(q)\} \frac{\text{Im}\chi(q, \omega_0)}{\omega_0}, \quad (3a)$$

$$\left(\frac{1}{T_{2G}}\right)^2 = \frac{p}{8\hbar^2} \left[\int \frac{dq}{2\pi} A_c^4(q) \chi^2(q) - \left\{ \int \frac{dq}{2\pi} A_c^2(q) \chi(q) \right\}^2 \right]. \quad (3b)$$

Here $\chi[(q, \omega)]$ [$\chi(q)$] is the dynamic (static) susceptibility per spin in units of $(g\mu_B)^2$, ω_0 is the NMR frequency, and $A_\alpha(q) = \sum_j A_\alpha^{ij} \exp(iqr_{ij})$. The magnetic field is assumed to be applied along the c direction. The prefactor in Eq. (3b) is chosen for nuclei with spin 3/2 and is valid when the spin-echo decay is measured on one of three NMR lines split by the quadrupole interaction. The isotopic abundance p is 0.69 for ^{63}Cu nuclei. Since $\chi(q)$ and $\text{Im}\chi(q, \omega)$ are strongly peaked at $q = \pi$ (the antiferromagnetic wave vector), $A_\alpha(q)$ in the integral can be replaced by $A_\alpha(\pi)$.

An analytic result for $\chi(q, \omega)$ at finite temperatures was first obtained by Schultz,³ using the bosonization technique to transform the Heisenberg model to a free boson Hamiltonian. The result satisfies the quantum critical scaling,

$$\chi(q, \omega) = D f_1(\tilde{q}, \tilde{\omega}) / k_B T, \quad (4)$$

where $\tilde{q} = c(q - \pi) / k_B T$, $\tilde{\omega} = \hbar \omega / k_B T$, and D is an unknown constant determining the overall magnitude of $\chi(q, \omega)$. It immediately follows from Eqs. (3) and (4) that $1/T_1 = \text{const}$ and $1/T_{2G} \propto 1/\sqrt{T}$. In order to eliminate dependence on material parameters, we define the following normalized dimensionless NMR rates, which should be universal functions of T/J and can be compared directly with theories,

$$(1/T_1)_{\text{norm}} = \frac{2\hbar J}{T_1 \{A_x^2(\pi) + A_y^2(\pi)\}}, \quad (5a)$$

$$(\sqrt{T}/T_{2G})_{\text{norm}} = \left(\frac{k_B T}{pJ}\right)^{1/2} \frac{\hbar J}{A_z^2(\pi) T_{2G}}. \quad (5b)$$

Sachdev used the result by Schulz to calculate the NMR rates and obtained⁴

$$(1/T_1)_{\text{norm}} = 2D, \quad (\sqrt{T}/T_{2G})_{\text{norm}} = 1.1908D, \quad (6)$$

with their ratio being a universal number $(T_{2G}/T_1 \sqrt{T})_{\text{norm}} = 1.680$, which can be tested experimentally. The T dependence of $1/T_1$ and $1/T_{2G}$ and the value of $T_{2G}/(\sqrt{T}T_1)$ indeed agree quite well with the experimental data on Sr_2CuO_3 .⁸

However, the free boson theory neglects the marginally irrelevant operator in the original Heisenberg Hamiltonian, describing umklapp scattering processes, that leads to a multiplicative logarithmic correction to $\chi(q, \omega)$. Sachdev has shown that to the leading order both $(1/T_1)_{\text{norm}}$ and $(\sqrt{T}/T_{2G})_{\text{norm}}$ acquire an identical multiplicative factor of $\ln^{1/2}(T_0/T)$, where T_0 is the high energy cutoff of the order of J .⁴ However, the agreement with the experimental data becomes worse if the factor $\ln^{1/2}(J/T)$ is included.⁸ Recently Starykh, Singh, and Sandvik have proposed a more elaborate theory of the logarithmic corrections⁵ by adopting a simple ansatz generalizing the finite-size scaling and the conformal mapping to correlation functions with multiplicative logarithmic factors. They showed that the susceptibility takes the form

$$\chi(q, \omega) = D \ln^{1/2}(T_0/T) f_2(\tilde{q}, \tilde{\omega}, \Delta) / k_B T, \quad (7)$$

with $\Delta = (1/4)\{1 - [2 \ln(T_0/T)]\}$. In addition to the multiplicative factor, the logarithmic T dependence of Δ also breaks the scaling. The equal-time real-space correlation function obtained from this formula agrees very well with the quantum Monte Carlo (QMC) result for $8 \leq J/T \leq 32$ if $T_0/J \approx 4.5$ and $D \approx 0.075$. The theoretical expressions for the NMR rates are⁵

$$(1/T_1)_{\text{norm}} = 2D \times 2^{5/2-2\Delta} \sin(2\pi\Delta) I_1(\Delta) \ln^{1/2}(T_0/T) / \pi^2, \quad (8a)$$

$$(\sqrt{T}/T_{2G})_{\text{norm}} = D \times 2^{-5/2+2\Delta} \sin(2\pi\Delta) \Gamma^2(1-2\Delta) I_2(\Delta) \times \ln^{1/2}(T_0/T) / \pi^{3/2} \quad (8b)$$

with $I_1(\Delta) = \int_0^\infty dx [x/(\sinh x)]^{4\Delta}$ and $I_2(\Delta) = 4 \int_0^\infty dx |\Gamma(\Delta - ix)/\Gamma(1 - \Delta - x)|^4$.

The above finite- T QMC estimate of the scale factor D compares well with recent $T=0$ numerical calculations [0.067 89 (Ref. 18) and 0.065 (Ref. 19)], but certainly may contain some errors arising from nonasymptotic contributions in the temperature regime studied. The high energy cut-off T_0 has also been determined from fits of QMC data for the static staggered susceptibility and structure factor to their corresponding analytic expressions.⁶ The results are $T_{0,x} = 3.9 \pm 0.3$ from the susceptibility data and $T_{0,s} = 5.1 \pm 0.2$ from the structure factor. Within the accuracy of the procedure these numbers can be considered being in good agreement with the value $T_0 = 4.5$ quoted above.

We now turn to the experimental results. NMR experiments on ^{63}Cu nuclei in Sr_2CuO_3 have been performed by Takigawa *et al.*⁸ They have measured $1/T_1$ along the three crystal axes and $1/T_{2G}$ along the c direction. A static approximation was used to obtain $1/T_{2G}$ from the spin-echo decay data, namely time dependence of I_z due to a spin-lattice relaxation process was neglected. Although this is a reasonable approximation in Sr_2CuO_3 , for which $(1/T_1)_c$ is more than an order of magnitude smaller than $(1/T_{2G})_c$, it is desirable to take the I_z fluctuation effects into account to obtain more accurate values of $1/T_{2G}$. Recently, Curro *et al.*²⁰ have derived a highly accurate analytic expression of spin-echo decay in the presence of the I_z fluctuations, using the Gaussian approximation proposed by Recchia *et al.*²¹ We fitted the spin-echo decay data in Sr_2CuO_3 to this expression and extracted $1/T_{2G}$ as defined in Eq. (3b).

In order to convert the measured NMR rates to the normalized units defined in Eq. (5), we need the values of $A_\alpha(\pi)$ and J . The values of $A_\alpha(\pi)/\sqrt{J}$ were determined from the width of the characteristic broad background of the NMR spectra due to a field-induced staggered magnetization near impurities,²² using the calculation of alternating local susceptibility near chain ends by Eggert and Affleck.² The accuracy of these values depends on how closely the actual impurities (most likely the holes in the Zhang-Rice singlet states²³ due to excess oxygen) behave as chain ends. We consider it to be of the order of 10%. The anisotropy of A_α can be determined much more accurately, with an uncertainty of a few percent.

The exchange J was estimated to be 2200 ± 200 K (Ref. 11) by fitting the temperature dependence of the uniform

susceptibility to the theory by Eggert, Affleck, and Takahashi.¹ This value was used in the analysis of the previous NMR experiments.^{8,22} Suzuura *et al.*, on the other hand, measured the optical absorption spectrum at $T = 32$ K,²⁴ which was interpreted to be due to simultaneous phonon and spin excitations. Lorenzana and Eder²⁵ analyzed the spectrum and obtained $J = 2850$ K. Since this value is directly determined from the sharp peak of the spectrum located exactly at $\pi J/2$ (the top of the des Cloiseaux-Pearson mode) except for a small shift corresponding to the optic phonon frequency (~ 0.08 eV), this latter value should be more reliable.

We speculate that the discrepancy may be resolved if one takes into account lattice degrees of freedom. A temperature dependence of J naturally results from the thermal lattice expansion. In fact, we recognize a slight systematic deviation between the $\chi(T)$ data and the theoretical curve assuming a constant J (Fig. 4 in Ref. 11). Since the measured susceptibility includes the constant orbital and diamagnetic contributions, which are not known well, we can choose $J = 2850$ K at $T = 0$ and allow J to change with temperature to reproduce the $\chi(T)$ data. We found that the $\chi(T)$ data can be explained if J is reduced to 2530 K at $T = 800$ K, which is the highest temperature of the measurements. Since $J \propto t^4$ and $t \propto d^{-3.5}$, where t is the Cu to O transfer integral and d is the Cu-O distance,²⁶ thermal expansion of 0.8% over 800 K is enough to account for this change of J . This seems quite plausible.

A recent QMC study of the Heisenberg chain including dynamic (fully quantum mechanical) phonons indicates that the fluctuations in J may furthermore lead to an apparent shift of J as obtained from $\chi(T)$. A fit to the Heisenberg $\chi(T)$, assuming a constant J , gives a result which is lower than the actual average spin-spin coupling in the spin-phonon model.²⁷ This effect may be the reason for the different values of J obtained from $\chi(T)$ and the optical absorption. We use $J = 2850$ K in the following analysis but show also the results for $J = 2200$ K to indicate how sensitively the results depend on the value of J .

Finally, we are at a position to compare the experimental results with the theory. Figure 1 shows the results for $(1/T_1)_{\text{norm}}$. Since the ratio $A_\alpha(\pi)/\sqrt{J}$ is determined from experiments, $(1/T_1)_{\text{norm}}$ is independent of the choice of J . The solid line shows the result of Eq. (8a) with $T_0/J = 4.5$ and $D = 0.062$, while the dotted line is the $\ln^{1/2}(J/T)$ dependence with a magnitude chosen arbitrarily. It is clear that the theory by Starykh *et al.* shows much weaker T dependence than $\ln^{1/2}(J/T)$ and is closer to the experimental data. The value $D = 0.062$ agrees with the value $D = 0.075$ determined from the comparison with the QMC results of the real-space correlation function⁵ within the uncertainty due to errors in $A_\alpha(\pi)/\sqrt{J}$ and numerical determination of D .

Figure 2 shows the results for $(\sqrt{T}/T_{2G})_{\text{norm}}$. The open (filled) circles are the experimental results for $J = 2850$ K ($J = 2200$ K) adjusted for the I_z fluctuations. The crosses are the results for $J = 2850$ K in the static approximation. The correction for the I_z fluctuations reduces the value of $1/T_{2G}$ by 6–12%. The solid and the dotted lines are the theoretical results of Eq. (8b) for $D = 0.062$ and $D = 0.07$, respectively ($T_0/J = 4.5$). We remark that only the first term in Eq. (3b), which we call the ‘‘scaling part,’’ can be calculated reliably

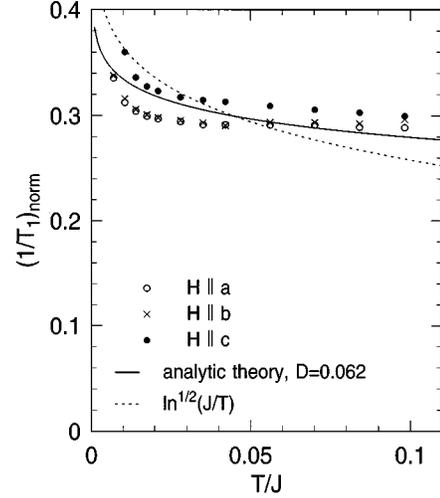


FIG. 1. Results for $(1/T_1)_{\text{norm}}$. Symbols are the experimental data for different field directions. The solid and dotted lines show the analytic theory Eq. (8a) with $D = 0.062$ and the $\ln^{1/2}(J/T)$ dependence, respectively.

by the field theory, giving the results of Eq. (8b). The second term in Eq. (3b) is the square of the local susceptibility, which cannot be handled properly by continuum effective field theory.⁵ It is smaller than the scaling part by a factor $(T/J)\ln^2(T_0/T)$. QMC calculations²⁸ are useful to examine the relative magnitude of the second term. The filled and open triangles in Fig. 2 show the QMC results for the full and the scaling part, respectively. The QMC results for the scaling part agree very well with the analytic results with $D = 0.07$. The analytic results for $D = 0.062$ (the best value to fit the $1/T_1$ data) are in good agreement with the experimental results for $J = 2850$ K (filled circles) if the negative contribution from the second term in Eq. (3b) (the difference between the two sets of QMC results) is taken into account.

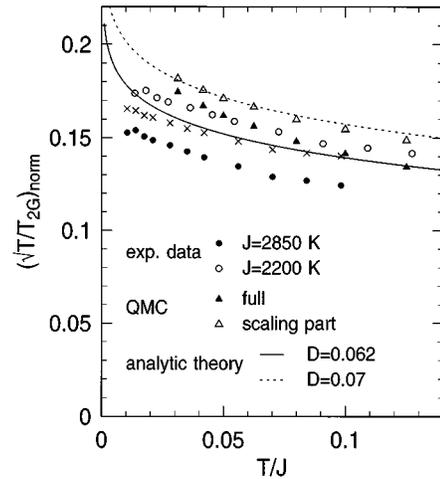


FIG. 2. Results for $(\sqrt{T}/T_{2G})_{\text{norm}}$. The filled and open circles are the experimental data for different values of J corrected for the I_z fluctuations. The crosses are the data for $J = 2850$ K without the corrections. The filled and open triangles show the QMC results for the full and the scaling [the first term in Eq. (3b)] parts, respectively. The results of the analytic theory Eq. (8b) for the scaling part is shown by the solid ($D = 0.062$) and the dotted ($D = 0.07$) lines.

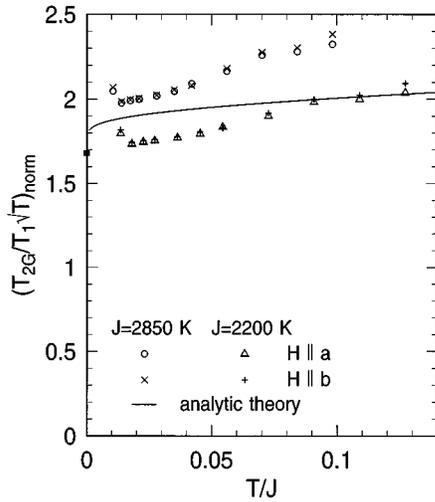


FIG. 3. Results for the ratio of $(1/T_1)_{\text{norm}}$ and $(\sqrt{T}/T_{2G})_{\text{norm}}$. Symbols are the experimental data for different values of J and field directions. The result of the analytic theory [the ratio of Eqs. (8a) and (8b)] is shown by the line. Its value at $T=0$ (square) coincides the scaling result.

Figure 3 shows the ratio of Eqs. (5a) and (5b). We used the $1/T_1$ data for $H||a$ and $H||b$, since the hyperfine form factor $A_x^2(q) + A_y^2(q)$ for $H||c$ is relatively small at $q = \pi$. This ratio provides a particularly stringent test for theories,

since uncertainties in the values of D and $A_\alpha(\pi)$ cancels out and the only parameter that needs to be known is the exchange J . The results of the analytic theory are between the two sets of experimental data for different values of J . It predicts a weak logarithmic T dependence, consistent with the experimental data, with an infinite slope at $T=0$. The value at $T=0$ shown by the square in Fig. 3 is equal to the scaling result 1.680, which is temperature independent. Once again, including the logarithmic corrections improves the agreement with the experimental data.

In summary, $1/T_1$, $1/T_{2G}$, and their ratio in Sr_2CuO_3 all show good quantitative agreement with the results of the analytic field theory that takes the logarithmic corrections into account. Our results demonstrate the importance of the logarithmic corrections in analyzing experimental data for quasi-1D systems, and that the dynamics in Sr_2CuO_3 is well described by a $S=1/2$ 1D Heisenberg model with a nearest-neighbor exchange. We suggest that remaining small discrepancies, such as the different estimates of J from the magnetic susceptibility and optical absorption experiments, may be due to spin-phonon interactions.

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