RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 51, NUMBER 14 1 APRIL 1995-II

Spin dynamics of La_2CuO_4 and the two-dimensional Heisenberg model

Anders W. Sandvik* and Douglas J. Scalapino
Department of Physics, University of California, Santa Barbara, California 93106
(Received 31 October 1994)

The spin-lattice relaxation rate $1/T_1$ and the spin-echo decay rate $1/T_{2G}$ for the two-dimensional Heisenberg model are calculated using quantum Monte Carlo and maximum-entropy analytic continuation. The results are compared with recent experiments on La_2CuO_4, as well as predictions based on the nonlinear σ model.

The CuO_2 planes of the undoped high-T_c cuprates are good physical realizations of the two-dimensional (2D) antiferromagnetic Heisenberg model.[1] The mapping of this lattice model onto the nonlinear σ model (NLσM) in 2+1 dimensions has led to detailed predictions for various experimentally measurable quantities.[2–4] For $T<600$ K (above the 3D ordering temperature) the correlation length of La_2CuO_4 grows exponentially as the temperature is lowered.[5] The behavior is in close agreement with quantum Monte Carlo results for the 2D Heisenberg model with a nearest-neighbor coupling $J\approx 1500$ K,[6] and corresponds to the NLσM in the low-temperature "renormalized classical" (RC) regime.[2] It was recently suggested[3,4,7,8] that the high-temperature behavior of the cuprates corresponds to the "quantum critical" (QC) regime of the NLσM, where the leading temperature dependence of the inverse correlation length is linear. Experimental evidence supporting this scenario has been provided by Imai *et al.*, who measured the spin-lattice relaxation rate $1/T_1$ and the Gaussian component of the spin-echo decay rate $1/T_{2G}$ at temperatures as high as $T=900$ K.[9,10] In particular, it was found that $1/T_1$ and the ratio T_1T/T_{2G} were both temperature independent at high temperatures, as predicted for the QC regime.[8] These experiments were recently repeated by Matsumura *et al.*[11] Their results for $1/T_1$ are almost identical to the earlier ones, but for $1/T_{2G}$ the temperature dependence obtained is different at high temperature, causing TT_1/T_{2G} to be temperature dependent, in disagreement with the QC scenario. In addition to this discrepancy, an open question is the reason for the absence of the minimum in $1/T_1$ at $T\approx 750$ K, theoretically predicted by Chakravarty and Orbach.[12] In order to settle these questions we have calculated both $1/T_1$ and $1/T_{2G}$ for the 2D Heisenberg model using quantum Monte Carlo simulation and the maximum-entropy analytic continuation method.[13,14] This enables us to compare directly the spin dynamics of the Heisenberg model and La_2CuO_4, as well as to assess rigorously the accuracy of the predictions based on the NLσM. Lower temperatures can be reached than with high-temperature series expansions[15,16] and calculations on small clusters,[17] and the approximations necessary with these methods can be avoided.

Overall our results are in good agreement with the experiments on La_2CuO_4. However, $1/T_{2G}$ for the 2D Heisenberg model decreases faster than the rate reported by Imai *et al.* above 750 K. The temperature dependence is $\sim T^{-2}$ for $0.45<T/J<1$, in disagreement with the QC prediction $\sim T^{-1}$. For $1/T_1$ our results are in good agreement with the experiments. We find that while $1/T_1$ exhibits a minimum for a local contact hyperfine coupling of the type used by Chakravarty and Orbach,[12] this minimum is absent when the experimentally known on-site and near-neighbor interaction is used.

The spin-lattice relaxation rate and the spin-echo decay rate for a given nucleus provide information on the spin susceptibility through the direct and transferred hyperfine couplings of the nuclear spin to surrounding electronic spins. Here we consider the standard 2D Heisenberg Hamiltonian

$$\hat{H}=J\sum_{i=1}^{N}\sum_{\delta}\vec{S}_i\cdot\vec{S}_{i+\delta}, \tag{1}$$

where $\vec{S}_i$ is a spin-$\frac{1}{2}$ operator, and δ runs over the nearest neighbors of site i. For a ^{63}Cu nuclear spin $\vec{I}_0$ at site 0, the coupling to the electronic spins $\vec{S}_i$ is given by the hyperfine Hamiltonian[19,20]

$$^{63}\hat{H}=A_\perp(I_0^xS_0^x+I_0^yS_0^y)+A_\parallel I_0^zS_0^z+B\sum_\delta \vec{I}_0\cdot\vec{S}_\delta. \tag{2}$$

The constants $A_\perp$, $A_\parallel$, and B are known from Knight shift measurements.

With the external field in the direction α, the NMR spin-lattice relaxation rate is given by[21]

$$\frac{1}{T_1}=\frac{1}{N}\sum_{\alpha'}\sum_q |A_q^{\alpha'}|^2S(q,\omega_N), \tag{3}$$

where α' denotes the two axes perpendicular to α, and $A_q^{\alpha'}$ is the Fourier transform of the α' component of the hyperfine coupling. The dynamic structure factor $S(q,\omega)$ is related to the imaginary part of the spin susceptibility; $S(q,\omega)=\chi''(q,\omega)/(1-e^{-\beta\omega})$. Since the resonance frequency ω_N is small compared to J, $1/T_1$ effectively measures $S(q,\omega\rightarrow 0)$, averaged with the hyperfine form factor $|A_q^{\alpha'}|^2$. In terms of the inverse Fourier transform $S_{mn}=S(m\vec{x}+n\vec{y},\omega\rightarrow 0$ of $S(q,\omega)$, $1/T_1$ with the external field perpendicular to the CuO_2 planes is given by

$$^{63}\left(\frac{1}{T_1}\right)_\perp=2(A_\perp^2+4B^2)S_{00}+16A_\perp BS_{10}+16B^2S_{11}+8B^2S_{20}. \tag{4}$$

This is the rate measured in the experiments by Imai *et al.* and Matsumura *et al.* $S_{mn}(\omega)$ can be obtained from the imaginary-time correlation function

$$C_{mn}(\tau)=\langle S^z_{m\vec{x}+n\vec{y}}(\tau)S^z_0(0)\rangle, \tag{5}$$

where $S^z_{\vec{r}}(\tau)=e^{\tau\hat{H}}S^z_{\vec{r}}e^{-\tau\hat{H}}$, by inverting the relation

$$C_{mn}(\tau)=\frac{1}{\pi}\int_{-\infty}^{\infty}d\omega S(m\vec{x}+n\vec{y},\omega)e^{-\tau\omega}. \tag{6}$$

Here $C_{mn}(\tau)$ is computed using a quantum Monte Carlo technique,[22] and the inversion of (6) is carried out with the maximum-entropy method.[13,14]

The rate $1/T_{2G}$ is related to the interactions between the nuclear spins. The coupling (2) induces an indirect nuclear spin-spin interaction, which dominates the direct dipole-dipole interactions. For the external magnetic field applied perpendicular to the CuO_2 planes, Pennington and Slichter derived the following expression for $1/T_{2G}$:[23]

$$^{63}\left(\frac{1}{T_{2G}}\right)_{\perp}=\left(\frac{0.69}{2\hbar}\sum_i J_z^2(\vec{x}_i)\right)^{1/2}. \tag{7}$$

Here $J_z(\vec{x}_i)$ is the z component of the induced interaction at distance $\vec{x}_i$, given by

$$J_z(\vec{x}_i)=A_{\parallel}F_z(\vec{x}_i)+B\sum_{\delta}F_z(\vec{x}_{i+\delta}), \tag{8}$$

with

$$F_z(\vec{x}_i)=-\frac{1}{2}\left(A_{\parallel}\chi(\vec{x}_i)+B\sum_{\delta}\chi(\vec{x}_{i+\delta})\right), \tag{9}$$

where $\chi(\vec{x}_i)$ is the static response at separation $\vec{r}=\vec{x}_i$, given by the Kubo formula

$$\chi(\vec{x}_i)=\int_0^{\beta}d\tau\langle S^z_i(\tau)S^z_0(0)\rangle. \tag{10}$$

The factor 0.69 in (7) is the natural abundance of the ^{63}Cu isotope.

We have used a recently improved variant of the Handscomb quantum Monte Carlo technique[22] to calculate the necessary correlation functions. Unlike standard methods,[24] this technique is free from the systematical errors associated with the Trotter breakup. For the analytic continuation of the imaginary-time data necessary to obtain $1/T_1$, we have implemented the so-called "classic" maximum-entropy procedure as described in a recent unpublished work by Jarrell and Gubernatis.[14] We have studied systems of $N=64\times 64$ spins with periodic boundary conditions, at temperatures $T/J=0.25-1.0$. At these temperatures the correlation length is smaller than the lattice size, and there are virtually no finite-size effects.

The calculation of $1/T_{2G}$ is straightforward, as it involves only the static susceptibility (10). We use the relation $A_{\parallel}=-4B$, experimentally known to hold quite accurately.[19,20] We are then left with J and B as fitting parameters, that can be checked against other experiments. The best agreement with the experimental data for $1/T_{2G}$ is obtained with $J=1580$ K and $B=3.4\times 10^{-7}$ eV ($\approx$ 37 kOe/μ_B), both consistent with other estimates.[5,6,20] The Monte Carlo results with these parameters are shown in Fig. 1, along with the experimental data.[25] Although the overall agreement is good, a notable feature is that for $T>750$ K the data of Imai *et al.*[10] are flatter than both the Monte Carlo results and those of Matsumura *et al.*[11] This flatness cannot be reproduced for the Heisenberg model with any reasonable values of J and B and, if correct, must be associated with physics not described by this model alone. On the other hand, the data of Matsumura *et al.* are well reproduced at high temperatures. Figure 1 also shows the theoretical form derived by Chubukov *et al.*[26] for the QC regime, which for the hyperfine coupling used here becomes

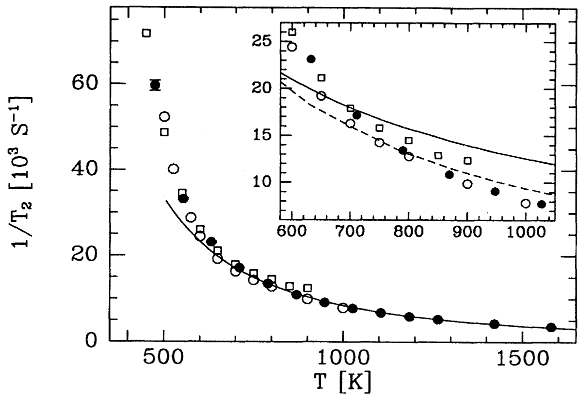

FIG. 1. Monte Carlo results for $1/T_{2G}$ (solid circles) and experimental results by Imai *et al.* (Ref. 10) (open squares) and Matsumura *et al.* (Ref. 11) (open circles). The solid line in the main figure is of the form $\sim T^{-2}$. Inset: Same as the main figure with the QC prediction by Chubukov *et al.* (Ref. 26) (solid curve) and including the temperature dependence of the spin-wave velocity (Ref. 18) (dashed curve).

$$\frac{1}{T_{2G}}\approx 0.49\ \xi(T)\times 10^4\ \mathrm{s}^{-1}, \tag{11}$$

where the QC correlation length is given by[3,4]

$$\xi=c/(1.04T)\quad \text{(QC regime)}, \tag{12}$$

and the spin-wave velocity $c\approx 1.68$.[29] As noted by Chubukov *et al.*,[26] the overall magnitude of $1/T_{2G}$ at high temperature is well reproduced with this formula, but the slope is not. Actually, the Monte Carlo results for $1/T_{2G}$ in the regime $0.45<T<1$ are well described by a T^{-2} behavior, a quite significant deviation from (11). Elstner *et al.*[18] recently suggested that the leading lattice corrections to the NLσM can be taken into account via a temperature-dependent spin-wave velocity. The velocity calculated from Monte Carlo results for the static structure factor and the static susceptibility agrees well[27] with the high-temperature series expansion results by Elstner *et al.*,[18] and when used in Eq. (12) slightly improves the agreement with the Monte Carlo results for $1/T_{2G}$ at high temperatures.

We now turn to the calculation of $1/T_1$, which is more complicated as it relies on a numerical analytic continuation

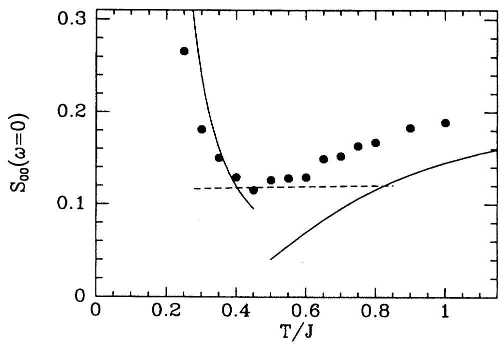

FIG. 2. Maximum-entropy results for S_{00} vs T (solid circles) compared to the RC form (13) and the high-temperature form derived in Ref. 12 (solid curves). The dashed curve is the QC form (15).

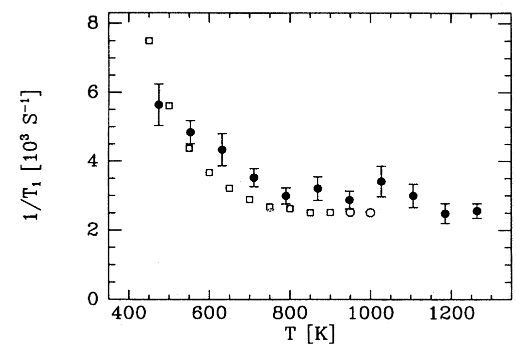

FIG. 3. Maximum-entropy results for $1/T_1$ vs T (solid circles) compared to the experimental results by Imai *et al.* (Ref. 9) (open squares) and Matsumura *et al.* (Ref. 11) (open circles).

of imaginary-time correlation functions. For the local correlation function $C_{00}(\tau)$ the relative statistical errors in our data are typically as low as 10^{-4}, and the continuation of this quantity is relatively stable. For C_{10}, C_{11}, and C_{20} the relative errors are typically on the order of 10^{-3} and an accurate determination of $1/T_1$ using the full extended hyperfine coupling (2) is therefore more difficult than with a strictly local interaction ($B=0$).

In Fig. 2, the $\omega\to 0$ limit of S_{00} is graphed versus the temperature. For a strictly local coupling, this quantity is proportional to $1/T_1$. Repeating the analytic continuation procedure for different subsets of the Monte Carlo data, we estimate the statistical errors to be a few percent (any bias due to the maximum entropy procedure itself is of course not captured this way). A broad minimum around $T/J=0.5$ is observed, in agreement with the prediction by Chakravarty and Orbach,[12] who deduced this feature by contrasting the behavior of $S_{00}(\omega\to 0)$ in the RC regime and the high-temperature limit. The RC expression is[12,4]

$$S_{00}^{\rm RC}(\omega\to 0)=\frac{\lambda N_0^2}{\sqrt{6}}\frac{\xi}{c}\left(\frac{T}{2\pi\rho_s}\right)^{3/2}\left(\frac{1}{1+T/2\pi\rho_s}\right)^2, \quad (13)$$

where the correlation length is given by[2,28]

$$\xi=\frac{e}{8}\frac{c}{2\pi\rho_s}\left(1-\frac{T}{4\pi\rho_s}\right)e^{2\pi\rho_s/T} \quad \text{(RC regime)}. \quad (14)$$

The spin stiffness $\rho_s\approx 0.18$,[29,30] and the ordered moment is $N_0\approx 0.31$.[30] The constant λ has not been calculated rigorously, but an estimate based on fitting the NLσM scaling forms to numerical results is $\lambda N_0^2=0.61$.[12,4] The rather poor agreement with our result for S_{00} shown in Fig. 2 indicates that this value is too large. It should be noted, however, that even the lowest temperatures studied here correspond to the crossover regime to RC behavior,[18] and perfect agreement with the RC expression cannot be expected. The high-temperature form derived in Ref. 12 is also shown in Fig. 2, and deviates from the Monte Carlo result by 25% at $T/J=1$. In the regime $0.4<T/J<0.6$, $S_{00}(\omega\to 0)$ is rather flat, as predicted for the QC regime. The expression derived by Chubukov *et al.* is[4]

$$S_{00}^{\rm QC}(\omega\to 0)=\frac{N_0^2}{\rho_s}\left(\frac{3T}{2\pi\rho_s}\right)^{\eta}R_1, \quad (15)$$

where the the 3D classical Heisenberg exponent $\eta\approx 0.03$, and R_1 is a constant for which Chubukov *et al.* estimated $R_1\approx 0.22$ (there are certain complications in estimating R_1).[4] Equation (15) with this value of R_1 describes the behavior in the intermediate temperature regime reasonably well.

A minimum in $1/T_1$ has not been observed experimentally.[9,11] In Fig. 3 we show results obtained with the full hyperfine coupling (2), using the same values of J and B as in the fit to $1/T_{2G}$ in Fig. 1. For $A_\perp/B$ we take the experimental value 0.84.[19,20] The statistical errors are rather large, as discussed above, but a clear difference from the temperature dependence of Fig. 2 can be noted, and the agreement with the results by Imai *et al.*[9] and Matsumura *et al.*[11] is reasonably good. In particular, $1/T_1$ is temperature independent at high temperatures and the minimum found above for $S_{00}(\omega\to 0)$ at $T\approx J/2$ is absent.

To summarize our results, we note that $1/T_{2G}$ for the Heisenberg model agrees well with the experimental results for La_2CuO_4 by Imai *et al.*[9,10] and Matsumura *et al.*[11] for $T<750$ K. However, for $T>750$ K the Heisenberg result decays considerably faster than the data by Imai *et al.*, but fits well the data by Matsumura *et al.*. The temperature dependence is close to T^{-2} in a wide temperature regime. For $1/T_1$ our results are also in reasonable agreement with the experiments and, in particular, are almost temperature independent at high temperatures, as predicted for the QC regime.[4] For the q-integrated dynamic susceptibility $S_{00}(\omega\to 0)$ (i.e., $1/T_1$ with a constant hyperfine coupling $A_q=A$) we find a clear minimum around $T/J\approx 0.5$. This was predicted by Chakravarty and Orbach,[12] who also speculated that the crossover between the RC and high-temperature forms is related to QC behavior. This is confirmed by the close agreement of our numerical result with the QC prediction[4] in the regime $0.4<T/J<0.6$, which is also approximately the regime in which the uniform susceptibility exhibits QC behavior.[3,4] As a consequence of the behavior of $1/T_{2G}$, the ratio T_1T/T_{2G} is not constant at high temperatures, in disagreement with the QC prediction.[8] The temperature dependence is, however, relatively weak in the regime 600–800 K.

The deviations from QC behavior at very high temperatures are caused by lattice effects, not present for the continuum NLσM. Elstner *et al.* have argued that the dominant lattice effects can be absorbed into a temperature-dependent spin-wave velocity, and that the correlation length of the Heisenberg model then shows QC behavior above $T/J \approx 0.6$. In the regime $0.4 < T/J < 0.6$ there is a crossover to RC behavior. Using the temperature-dependent $c(T)$, the deviations from the QC predictions found here for $1/T_{2G}$ and the ratio TT_1/T_{2G} can be accounted for to some extent. However, it should be noted that the uniform susceptibility, the temperature dependence of which is in remarkable agreement with the QC prediction with a constant $c=c(0)$ for $0.35 < T/J < 0.55$,[3,4] does not follow the form expected with $c=c(T)$. Hence, the use of $c(T)$ in the QC formulas can be questioned.

Our study reaffirms that effects of the proximity to the critical point of a quantum phase transition are manifest in the 2D Heisenberg model. However, the size and location of the regime where QC behavior can be observed depends strongly on the quantity considered, and there does not seem to exist a temperature regime where the QC scaling formulas can be applied universally.

We would like to thank A. Chubukov, H. Monien, R.R.P. Singh, and A. Sokol for very helpful conversations. This work was supported by the Department of Energy under Grant No. DE-FG03-85ER45197.

---

*Present address: National High Magnetic Field Laboratory, 1800 E. Paul Dirac Dr., Tallahassee, FL 32306. On leave from Department of Physics, Åbo Akademi University, Åbo, Finland.

[1] See, e.g., E. Manousakis, Rev. Mod. Phys. **63**, 1 (1991).

[2] S. Chakravarty, B. I. Halperin, and D. R. Nelson, Phys. Rev. Lett. **60**, 1057 (1988); Phys. Rev. B **39**, 2344 (1989).

[3] S. Sachdev and J. Ye, Phys. Rev. Lett. **69**, 2411 (1992); A. Chubukov and S. Sachdev, *ibid.* **71**, 169 (1993).

[4] A. Chubukov, S. Sachdev, and J. Ye, Phys. Rev. B **49**, 11 919 (1994).

[5] G. Shirane *et al.*, Phys. Rev. Lett. **59**, 1613 (1987); Y. Endoh *et al.*, Phys. Rev. B **37**, 7443 (1988); B. Keimer *et al.*, *ibid.* **46**, 14 034 (1992).

[6] H.-Q. Ding and M. S. Makivić, Phys. Rev. Lett. **64**, 1449 (1990); M. S. Makivić and H.-Q. Ding, Phys. Rev. B **43**, 3562 (1991).

[7] A. J. Millis and H. Monien, Phys. Rev. Lett. **70**, 2810 (1993).

[8] A. Sokol and D. Pines, Phys. Rev. Lett. **71**, 2813 (1993).

[9] T. Imai *et al.*, Phys. Rev. Lett. **70**, 1002 (1993).

[10] T. Imai *et al.*, Phys. Rev. Lett. **71**, 1254 (1993).

[11] M. Matsumura, H. Yasuoka, Y. Ueda, H. Yamagata, and Y. Itoh, J. Phys. Soc. Jpn. **63**, 4331 (1994).

[12] S. Chakravarty and R. Orbach, Phys. Rev. Lett. **64**, 224 (1990).

[13] J. E. Gubernatis *et al.*, Phys. Rev. B **44**, 6011 (1991).

[14] M. Jarrell and J. E. Gubernatis (unpublished).

[15] M. P. Gelfand and R. R. P. Singh, Phys. Rev. B **47**, 14 413 (1993).

[16] A. Sokol, R. L. Glenister, and R. R. P. Singh, Phys. Rev. Lett. **72**, 1549 (1994).

[17] A. Sokol, E. Gagliano, and S. Bacci, Phys. Rev. B **47**, 14 646 (1993).

[18] N. Elstner, R. L. Glenister, R. R. P. Singh, and A. Sokol, Phys. Rev. B **51**, 8984 (1995).

[19] F. Mila and T. M. Rice, Physica C **157**, 561 (1989).

[20] A. J. Millis, H. Monien, and D. Pines, Phys. Rev. B **42**, 167 (1990).

[21] T. Moriya, Prog. Theor. Phys. **28**, 371 (1962).

[22] A. W. Sandvik and J. Kurkijärvi, Phys. Rev. B **43**, 5950 (1991); A. W. Sandvik, J. Phys. A **25**, 3667 (1992).

[23] C. H. Pennington and C. P. Slichter, Phys. Rev. Lett. **66**, 381 (1991).

[24] J. E. Hirsch *et al.*, Phys. Rev. B **26**, 5033 (1982).

[25] At high temperatures our results for $1/T_{2G}$ are in good agreement with the high-T series expansion results of Ref. 16 (taking into account the slightly different values used for B and J). At low temperature we find a considerably faster increase, however.

[26] A. V. Chubukov, S. Sachdev, and A. Sokol, Phys. Rev. B **49**, 9052 (1994).

[27] A. W. Sandvik, R. R. P. Singh, and A. Sokol (unpublished).

[28] P. Hasenfratz and F. Niedermayer, Phys. Lett. B **268**, 231 (1991); Z. Phys. B **92**, 91 (1993).

[29] R. R. P. Singh, Phys. Rev. B **39**, 9760 (1989).

[30] J. D. Reger and A. P. Young, Phys. Rev. B **37**, 5987 (1988); U.J. Wiese and H.P. Ying, Z. Phys. B **93**, 147 (1994).