Spin Dynamics and Magnetic Correlation Length in Two-Dimensional Quantum Heisenberg Antiferromagnets

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The correlated spin dynamics and temperature dependence of the correlation length $\xi(T)$ in two-dimensional quantum ($S = 1/2$) Heisenberg antiferromagnets (2DQHAF) on a square lattice are discussed in light of experimental results of proton spin lattice relaxation in copper formiate tetradeuterate. In this compound the exchange constant is much smaller than the one in recently studied 2DQHAF, such as La_2CuO_4 and $Sr_2CuO_2Cl_2$. Thus the spin dynamics can be probed in detail over a wider temperature range. The NMR relaxation rates turn out to be in excellent agreement with a theoretical mode-coupling calculation. The deduced temperature behavior of $\xi(T)$ is in agreement with high-temperature expansions, quantum Monte Carlo simulations, and the pure quantum self-consistent harmonic approximation. Contrary to the predictions of the theories based on the nonlinear σ model, no evidence of crossover between different quantum regimes is observed.

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One of the most appealing issues in quantum magnetism is the temperature and doping behavior of a $S = 1/2$ planar Heisenberg antiferromagnet (2DQHAF). The attention towards this aspect has been triggered by La_2CuO_4 and $Sr_2CuO_2Cl_2$. In fact, the former, besides being the parent of high-temperature superconductors, has represented the first experimental realization of a 2DQHAF. For both of the above systems the in-plane superexchange coupling constant is $J \simeq 1500$ K while the interplane interaction J' is no more than $10^{-4}J$. Soon after their discovery a large number of experimental studies were carried out to clarify the basic aspects of 2DQHAF and, in particular, the temperature dependence of the in-plane correlation length $\xi(T)$ [1–6].

Unfortunately, the high value of J in La_2CuO_4 and $Sr_2CuO_2Cl_2$ has not allowed one to perform experiments above a temperature range $T/J \simeq 0.6$, thus inhibiting the test of relevant theoretical predictions for 2DQHAF's, for instance, the possible crossover to a quantum critical (QC) regime of spin fluctuations. Thus, in spite of great experimental and theoretical efforts this issue is still controversial [7–9].

In this paper we present new experimental data for $\xi(T)$ in copper formiate tetradeuterate (CFTD) as obtained by NMR experiments. Furthermore, we show how a mode-coupling approach can be successfully employed to analyze the temperature behavior of the longitudinal relaxation time, thus allowing one to extract reliable information on the static and dynamical properties of the system. CFTD is a good prototype of 2DQHAF with $J'/J \simeq 10^{-4}$, but with $J \simeq 80$ K. Thus $\xi(T)$ can be determined over a wide temperature range (up to $T/J \simeq 1.5$) and consistently compared to a series of recent theoretical approaches, in particular by investigating the occurrence of quantum criticality and the predictions of the quantum nonlinear σ model (QNLσM).

The 2DQHAF, with nearest-neighbor interaction on a square lattice, is described by the Hamiltonian

$$\mathcal{H} = \frac{J}{2}\sum_{\mathbf{j},\mathbf{a}} \mathbf{S}_{\mathbf{j}} \cdot \mathbf{S}_{\mathbf{j}+\mathbf{a}}\,, \tag{1}$$

with $J > 0$ and $\mathbf{a} = (0, \pm a), (\pm a, 0)$, a being the lattice constant. The 2DQHAF has been commonly described in terms of the two-dimensional quantum nonlinear σ model [10], with the action given by

$$S = \frac{1}{2g}\int_{-\infty}^{\infty}\int_{0}^{u} d\mathbf{x}\, d\tau\, (|\nabla \mathbf{n}|^2 + |\partial_\tau \mathbf{n}|^2), \quad |\mathbf{n}|^2 = 1\,, \tag{2}$$

where $\mathbf{n}(\mathbf{x})$ is a unitary 3D vector field, and $g = c_s\Lambda/\rho_s$ and $u = c_s\Lambda/T$ are the coupling and the imaginary-time cutoff, respectively. This mapping is rigorous only in the large-spin limit, where ρ_s and c_s are the spin stiffness and the spin-wave velocity of the corresponding magnetic model. This approach can be extended to $S = 1/2$ provided that c_s and ρ_s are introduced as fitting parameters. A rich and interesting phase diagram can then be devised [11–13]. Accordingly, the QNLσM has risen to great popularity, both for basic quantum magnetism studies as well as for being a good candidate to describe the modifications in the microscopic properties of a 2DQHAF when it becomes a high-T_c superconductor upon charge doping. However, it should be remarked that the QNLσM is expected to model a 2DQHAF when the correlation length is very large, i.e., at low temperature, but it has not yet clearly stated which is the actual temperature upper limit where the currently available results are reliable. In particular, it is still the object of strong debate whether or not the predictions of the QNLσM can be usefully employed in the temperature range where experimental data on 2DQHAF are available.

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Below a finite value of g, by increasing the temperature, QNLσM calculations predict that the system should cross from the *renormalized classical* to the *quantum critical* regime and eventually to the classical behavior [11,13]. In the region where the quantum critical regime is expected to occur, namely, $0.4 \leq T/J \leq 0.8$, one should find $\xi \propto 1/T$.

The temperature behavior of the static correlation length $\xi(T)$ can be directly estimated by neutron scattering [1–3] or by the longitudinal NMR relaxation rate, which provides an indirect measure of ξ [5,6]. While, for small ξ ($\simeq a$), neutron scattering studies become difficult and accuracy problems arise, it has been proved [5] that NMR relaxation, driven by the correlated spin dynamics, can be a suitable tool to derive the temperature and doping dependence of the magnetic correlation length. Neutron scattering experiments in antiferromagnets with larger S have been recently carried out with good accuracy [2,3] and no evidence of crossover has been found in a large range of T/J. Indeed, these data are in agreement with high-temperature expansion (HTE) results [14], quantum Monte Carlo (QMC) data [15], and with the results of the pure quantum self-consistent harmonic approximation (PQSCHA) recently obtained by some of us [16,17]. The latter approach is reliable for any realistic temperature range for $S \geq 1$, and it gives good results starting from $T/J \gtrsim 0.35$ for $S = 1/2$ [17].

With NMR relaxation, one can suitably explore the temperature range where the correlation length is of the same order of the lattice constant a. ^{1}H NMR experiments have been performed on a $5 \times 5 \times 4$ mm^3 single crystal of copper formiate tetradeuterate $[Cu(HCO_2)_2 \cdot 4D_2O]$ [18]. The spin-lattice relaxation rate $1/T_1$ has been measured in an 8 kG magnetic field ($g\mu_B H \ll J$) applied perpendicular to the ab plane, by using standard pulse sequences. The experimental data for $1/T_1$ are reported in Fig. 1.

The relaxation rates can be calculated, through the mode-coupling approach [19–21]. We start by calculating the normalized Kubo relaxation function [22],

$$F_{\mathbf{k}}(t) = \frac{R_{\mathbf{k}}(t)}{R_{\mathbf{k}}(0)}, \qquad R_{\mathbf{k}}(t) = \int_0^{1/T} \langle S^{\alpha}_{\mathbf{k}}(t + i\lambda) S^{\alpha}_{\mathbf{k}} \rangle \, d\lambda , \tag{3}$$

by solving the following integro-differential equation [19–21]:

$$\frac{d}{dt} F_{\mathbf{q}}(t) = -\frac{2}{N} \sum_{\mathbf{k}} (J_{\mathbf{k}} - J_{\mathbf{q}-\mathbf{k}})(J_{\mathbf{k}} - J_{\mathbf{q}}) \times \frac{T}{\lambda + J_{\mathbf{k}}} \int_0^t F_{\mathbf{k}}(t - t') F_{\mathbf{q}-\mathbf{k}}(t - t') F_{\mathbf{q}}(t') , \tag{4}$$

where $J_{\mathbf{k}} = J \sum_{\mathbf{a}} \exp(\mathbf{k} \cdot \mathbf{a})$, and λ is a temperature-dependent parameter which fixes the static susceptibility of the system and can be directly related to $\xi(T)$ [21].

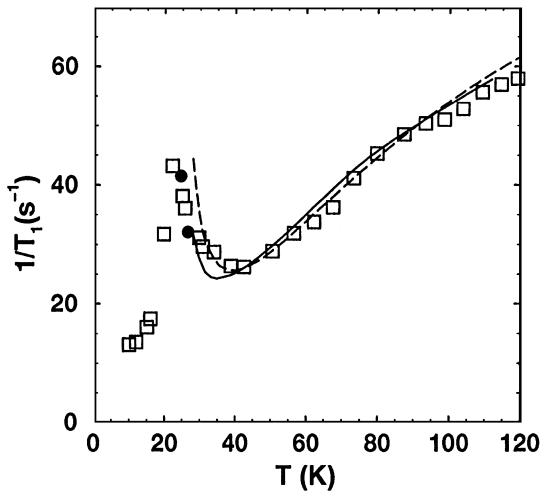

FIG. 1. Proton spin-lattice relaxation $1/T_1$ as a function of temperature. Squares: experimental data; dashed line: theoretical mode-coupling results using the static quantities of the spherical model; solid line: mode-coupling results using the correlation length given by the PQSCHA for $T/J > 0.35$; filled circles: mode-coupling results using the correlation length given by QMC simulations for $T/J < 0.35$ [8].

By means of the spectral theorem the dynamic cross section $S_{\mathbf{k}}(\omega)$ can be obtained from the Fourier transform of $F_{\mathbf{k}}(t)$. Thus the longitudinal relaxation time is given by [23]

$$1/T_1 = \frac{\gamma^2}{2N} \sum_{\mathbf{k}} S_{\mathbf{k}}(\omega) \{A^2 + B^2[\cos(k_x a) + \cos(k_y b)]\} , \tag{5}$$

where $\gamma = 2\pi \times 42.576 \times 10^2$ s^{-1} G^{-1}. The form factors $\{A^2 + B^2[\cos(k_x a) + \cos(k_y b)]\}$ have been derived by assuming the hyperfine coupling of ^{1}H nuclei with the two Cu^{2+} nearest neighbors only. The coupling constants have been estimated from the rotation pattern by rotating the crystal around the a axis in a field of 94 kG. In this way the transferred hyperfine interaction constant was directly obtained. The dominant dipolar part was determined through lattice sums (details will be published elsewhere [24]). The two coupling constants turned out $A = 2.31$ kG and $B = 1.46$ kG.

The analysis in terms of the theoretical models requires the knowledge of the exchange constant J. Neutron and light scattering experiments devised to estimate J have been performed in La_2CuO_4 [25]. At the moment, in CFTD we can assume a value about $J = 80$ K. Estimates based on the velocity of the spin waves [26] are in the same range, as well as old data [27] (within 10%), in agreement with the dispersion curve determined by neutron scattering near the zone boundary [28].

The mode-coupling scheme requires the static quantities as an external input. For this purpose the simplest and most self-consistent approach is to use those pertaining to the two-dimensional spherical model [21]. An alternative method is to rely on the results obtained for $\xi(T)$ by

the PQSCHA to determine the parameter λ appearing in Eq. (4), for $T/J \gtrsim 0.35$. The two theoretical curves are reported in Fig. 1. Furthermore, we have used the more precise QMC data for $\xi(T)$ [15] to calculate the relaxation rate at lower temperatures, down to the value of $T \simeq 20$ K, where three-dimensional ordering effects are revealed by the NMR experimental findings. According to currently available calculations based on the QNLσM, the relaxation rate $1/T_1$ should be weakly temperature dependent in the quantum critical regime [13], without any observable minimum at about $T/J = 0.5$ ($T = 40$ K) [29]. Instead the experimental data do present such a minimum, which is completely reproduced by our approach [30]. The minimum in $1/T_1$ is related to the $\mathbf{k}$ dependence of the hyperfine form factor in Eq. (5), which is strongly peaked at the center of the Brillouin zone (BZ) but nonzero at the AF wave vector $\mathbf{k} = (\pi/a, \pi/a)$. One has to notice that in the temperature range far from the transition the spherical model seems to be a good approximation, as shown also by the temperature dependence of the correlation length in Fig. 2.

The remarkable agreement between mode-coupling calculations and the experimental results allows us to derive the correlation length $\xi(T)$ from the experimental data through an *inversion* of the mode-coupling approach. The results are shown in Fig. 2, together with the theoretical predictions. The success of the mode-coupling approach gives further support to the validity of the scaling hypothesis, and the correlation length can be extracted with satisfactory accuracy from the relaxation rate by means of a procedure which has already yielded reliable quantitative estimates in La_2CuO_4 (see Refs. [5,6] for details). Indeed, when $\xi(T) \gg a$, classical scaling arguments for the generalized susceptibility and the decay constants lead to the following equation [5]:

$$1/T_1 = \gamma^2 \epsilon \frac{S(S+1)}{3} \left(\frac{\xi}{a}\right)^{z+2} \frac{\beta^2 \sqrt{2\pi}}{\omega_e} \left(\frac{a^2}{4\pi^2}\right) \int_{\mathrm{BZ}} d\mathbf{q} \times \frac{\{A^2 + B^2[\cos(q_x a - \pi) + \cos(q_y a - \pi)]\}}{(1 + q^2\xi^2)^2}, \quad (6)$$

thus establishing a one-to-one correspondence between $1/T_1$ and $\xi(T)$. In Eq. (6) the 2D wave vector $\mathbf{q}$ starts from the BZ boundary $(\pi/a, \pi/a)$, corresponding to the AF ordering wave vector; ω_e is the Heisenberg exchange frequency describing the fluctuations in the limit of infinite temperature, $z = 1$ is the dynamical scaling exponent, $\epsilon = 0.33$ takes into account the reduction of the amplitude due to quantum fluctuations [31], and β is a normalization factor preserving the sum rule for the amplitude of the collective modes. Also, the data for $\xi(T)$ obtained by means of this procedure are reported in Fig. 2. It is apparent that the values for $\xi(T)$ are close to the ones deduced by the mode-coupling approach, the main differences being at low values of ξ, as expected.

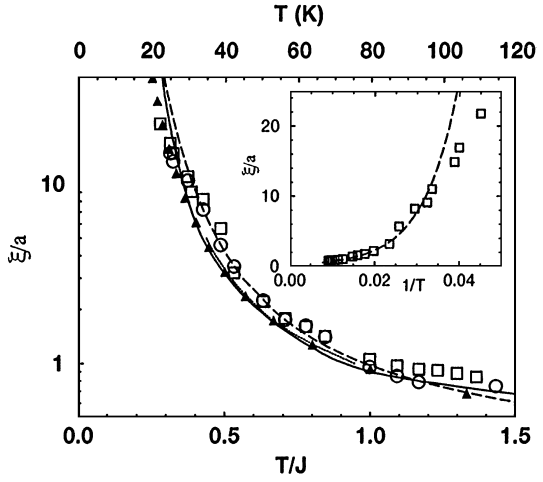

FIG. 2. Correlation length ξ vs temperature. Squares: experimental data deduced from $1/T_1$ by inversion of Eq. (6); circles: experimental data deduced from $1/T_1$ by inversion of mode-coupling results leaving λ as a free parameter; dashed line: spherical model; solid line: PQSCHA; triangles: QMC simulations [8]. The solid line in the range $0.45 < T/J < 1$ shows the HTE results [14]. In the inset we report the corresponding data for $\xi(T)$ derived from Eq. (6) and from the spherical model as a function of the inverse temperature. According to the predictions of the QNLσM, the QC regime should occur for $0.4 < T/J < 0.8$ (i.e., $0.016 \lesssim 1/T \lesssim 0.036$), with a correspondent linear behavior of $\xi(T)$ vs $1/T$, at variance with the experimental findings.

From the comparison with the theoretical estimates, the permanence of the system in the renormalized classical regime, on moving towards the classical one at highest temperatures, is shown. No evidence of crossover to the quantum critical regime appears, as is clearly shown in the inset of Fig. 2. Finally, we point out that, although we used a value of $J = 80$ K in all of our calculations, the main conclusions derived regarding the absence of a crossover towards a quantum critical regime do not depend on J.

In summary, we have used a single crystal of a proper 2DQHAF which allows one to analyze the behavior of the in-plane correlation length $\xi(T)$ over a wide temperature range in light of several theoretical models. $\xi(T)$ has been derived from ^{1}H NMR relaxation rates, having verified also that the mode-coupling theory is able to reproduce successfully the temperature dependence of $1/T_1$. It turns out that $\xi(T)$ is well described by QMC, PQSCHA, and HTE developed on a discrete lattice. In particular, at variance with other recent theories for 2DQHAF, no crossover to the quantum critical regime occurs, up to temperatures $T/J \simeq 1.5$, where the classical behavior has already been set. Furthermore, Eq. (6), which is based on the validity of the scaling hypothesis, is shown to yield reliable estimates of $1/T_1$ even for $\xi \simeq a$. Finally, after the submission of this paper, a neutron scattering study of the temperature dependence of the magnetic correlation length in CFTD was published [32]. A comparison of the data extracted

with the two techniques is now possible. The temperature dependence of ξ and the numerical values are in strict agreement, thus supporting our theoretical estimate based on the mode coupling. The lack of any crossover to a QC regime is hence also manifested by the neutron scattering data.

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