# Universal behavior of the spin-echo decay rate in $La_2CuO_4$

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We present a theoretical expression for the spin-echo decay rate $1/T_{2G}$ in the quantum-critical regime of square-lattice quantum antiferromagnets. Our results are in good agreement with recent experimental data by Imai *et al.* [Phys. Rev. Lett. **71**, 1254 (1993)] for $La_2CuO_4$.

## I. INTRODUCTION

Part of the recent interest in high-$T_c$ superconductivity has been devoted to the study of low-temperature magnetic phases of undoped and weakly doped antiferromagnetic $La_2CuO_4$ and related compounds. Neutron scattering measurements of the correlations length at $T \leq 500$ K (Ref. 1) have established that it agrees very well with the theoretical result for the low-temperature renormalized-classical (RC) region[2–4] (we set $k_B = 1$)

$$\xi \sim 0.34 \left(\frac{\hbar c}{2\pi\rho_s}\right) \exp(2\pi\rho_s/T) \left[1 - \frac{T}{4\pi\rho_s} + \cdots\right]. \quad (1)$$

In this region, the correlation length is determined predominantly by classical, thermal fluctuations, while quantum fluctuations only renormalize the values of input $T = 0$ parameters—the spin stiffness $\rho_s$ and the spin-wave velocity $c$. The RC value of the correlation length was also successfully used[5] to fit the data on the longitudinal spin-lattice relaxation rate, $1/T_1$, for $400 < T < 600$ K.[6]

More recently, significant attention has also been devoted to a region of intermediate temperatures where the temperature becomes larger than the energy scale associated with the spin stiffness at $T = 0$. Under these circumstances, the system behaves almost as if $\rho_s = 0$, i.e., as if it is at the quantum transition point separating the Néel and quantum-disordered states. This region, where the temperature is the largest energy scale in the problem, was first identified in Ref. 3 as the quantum-critical (QC) region, and then studied in detail in Refs. 7 and 8. Clearly, in the QC region, all Bose factors are of order of unity, and hence quantum and classical fluctuations are equally important.

It seems appropriate at this point to review some of the general ideas on the RC-QC crossover, and to present the aims of the theoretical comparisons with experiments. All two-dimensional magnets with a Néel ordered ground state are characterized by three energy scales: $\rho_s$, $T$, and a near-neighbor exchange constant $J$. It was pointed out in Ref. 8 that when $\rho_s/J$ and $T/J$ are small, the properties of such antiferromagnets are characterized by *universal* functions of $T/\rho_s$. The physics is a smooth function of $T/\rho_s$, especially simple in two limits: the RC limit ($T/\rho_s \to 0$) and the QC limit ($T/\rho_s \to \infty$). While only approximate $N = \infty$ results, where $N$ is the number of components of the order parameter, have been obtained for the full functional dependence on $T/\rho_s$, rather precise numerical predictions are available in the two limits. At intermediate values of $T/\rho_s$, deciding which of the two limits is appropriate is subject to interpretation, and different measurements may lead to different conclusions: we shall present a case for our choices below. A second, important issue is that of the corrections of order $\rho_s/J$ and $T/J$. These corrections are in fact *nonuniversal*, depend upon the nature of the lattice cutoff, and impossible to calculate precisely in an analytical theory. As theorists, the best we can do is to calculate the universal parts, compare with the experiments and numerical simulations, and then determine whether the discrepancies can be attributed to these nonuniversal corrections. Clearly, $T/J$ corrections will eventually become important at high enough temperatures—this will then impose a model-dependent upper boundary of the QC region. It turns out, however, that in the temperature range of experimental observations, the universal terms agree surprisingly well with the data indicating that these nonuniversal corrections are in fact quite small. Why this should be so is not completely understood, but we can offer the following plausibility arguments: (i) for small $T/\rho_s$, Hasenfratz and Niedermayer[4] have shown

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that there are no $\rho_s/J$ corrections to the leading terms in the low-temperature expansions of observables; (ii) numerical studies of the dynamical susceptibility, $\chi(k,\omega)$, have shown[9] that in the whole temperature range of experiments it remains strongly peaked at $(\pi,\pi)$, the local susceptibilities related to NMR and NQR measurements are then given by momentum integrals confined to $k \ll \Lambda$ where $\Lambda$ is the upper cutoff in momentum space; (iii) recent numerical analysis of the cutoff-dependent, two-loop equations of the $\sigma$ model[10] shows almost no cutoff dependence in the QC results until $T \sim 0.5c\Lambda$. We will argue below that the upper boundary of the universal QC region in the spin-1/2 square lattice Heisenberg model (above which $T/J$ terms cannot be neglected) is about $0.6J$ ($\sim 900$ K in $La_2CuO_4$, we use $J = 1500$ K). This temperature approximately coincides with the maximum temperature of experimental measurements.

We begin by presenting our scenario of where the RC-QC crossover occurs in the universal functions of $T/\rho_s$. Despite the fact that in Eq. (1) the dimensionless ratio is $T/2\pi\rho_s$, two of us have recently shown explicitly[8] that the parameter which governs the crossover behavior between RC and QC regimes is three times larger: $x = 3T/2\pi\rho_s$. The reason for the extra factor of 3 is the following. From renormalization-group studies, it is known that the effective spin stiffness, which has to be compared with the temperature, is $\rho_s^{\rm eff} = 2\pi\rho_s/\widetilde{N}$ where $\widetilde{N}$ is the effective number of components of the order parameter which contribute to the coupling constant renormalization.[2] Deep in the RC region, longitudinal fluctuations are suppressed, and the running coupling constant diverges at the scale of correlation length only due to *interactions* between $N-1$ transverse spin-wave modes [for $O(N)$ systems]. In this situation, $\widetilde{N} = N-2$, i.e., $\widetilde{N} = 1$ for the $O(3)$ case. On the contrary, in the crossover region near the quantum phase transition, all fluctuation modes contribute equally to the correlation length and we have simply $\widetilde{N} = N$. For $O(3)$ magnets then $\rho_{\rm eff} = 2\pi\rho_s/3$, and therefore $x = T/\rho_{\rm eff} = 3T/2\pi\rho_s$. It is the variation of $\rho_s^{\rm eff}$ vs $T/\rho_s$ which eventually gives rise to $N \to N-2$ substitution for $\rho_{\rm eff}$ deep in the RC region, as was observed in Ref. 8.

For the $S = 1/2$ antiferromagnet on a square lattice, both perturbative[11] and numerical[12] studies yield $\rho_s = 0.18J$, $c = 1.67Ja$, so that $x = T/0.38J$. On general grounds, we expect the crossover between RC and QC regimes to occur around $x \sim 1$, although not necessarily at the same $x$ for all observables. The upper boundary of the universal QC region is, we said, around $0.6J$. Then, in the undoped antiferromagnets, the temperature range of the QC behavior should be $0.4J < T < 0.6J$, which for $La_2CuO_4$ corresponds to $600 < T < 900$ K—a range which is accessible to experimental studies (a wider QC region is expected in the doped cuprates[13]). The uniform susceptibility data in this range[14,15] was compared to both RC (Refs. 4 and 8) and QC (Ref. 8) formulas, and very good agreement with the QC result was found. Furthermore, the measured spin-lattice relaxation rate, $1/T_1$, was found to be nearly temperature independent above 700 K, which is consistent with the QC behavior; the measured constant value of $1/T_1 \sim 2.7 \times 10^3$ sec$^{-1}$ is also quantitatively reproduced by the QC theory of Ref. 8 which predicts $1/T_1 \sim (3.2 \pm 0.5) \times 10^3$ sec$^{-1}$.

Very recently, Imai *et al.*[16] reported results on the Gaussian component of the spin-echo decay rate $1/T_{2G}$ in the temperature range between 450 and 900 K. Much can be learned from their results, but we will need theoretical predictions for $T_{2G}$ in the QC region, which are computed in the following section.

## II. $1/T_{2G}$ IN THE QUANTUM-CRITICAL REGION

The Gaussian component of the spin-echo decay rate $1/T_{2G}$ is related to the *static* susceptibility, $\chi(\mathbf{q}) \equiv \chi(\mathbf{q},\omega=0)$, by[17]

$$\left(\frac{1}{T_{2G}}\right)^2 = \frac{1}{a^2}\int \frac{d^2q}{4\pi^2} A_\perp^4(\mathbf{q})\,[\chi(\mathbf{q})]^2 - \left[\int \frac{d^2q}{4\pi^2} A_\perp^2(\mathbf{q})\,\chi(\mathbf{q})\right]^2, \qquad (2)$$

where $a$ is the interatomic spacing and $A_\perp(\mathbf{q})$ is a form factor in the direction perpendicular to the $CuO_2$ plane, which early studies[18] estimated as $A_\perp^2(\pi,\pi) = 4.85\times 10^7$ K/sec. In the temperature region of interest, $\chi(\mathbf{q})$ is strongly peaked at $\mathbf{q} = (\pi,\pi)$. The second term in (2) is nonuniversal in two dimensions, but it contains one less power of the correlation length compared to the first term, and thus can safely be neglected at low temperatures where $\xi \gg a$. To the same accuracy, we have to take the form factor exactly at $\mathbf{q} = (\pi,\pi)$, in which case $(1/T_{2G})^2$ measures simply the local static $\chi^2$.

Deep in the quantum-critical region, $1/T_1 \propto T^\eta$, $\chi(q) \propto q^{-2+\eta} f(q/T)$,[7] where $f(\infty)$ is a constant, and $\eta = 0.028$ (Refs. 19 and 20) is the critical exponent for the spin correlations at criticality. A straightforward analysis then yields[21] $1/T_{2G} \propto T^{-1+\eta}$. One may combine this with previous results on $1/T_1$, and the complete scaling forms (including prefactors) obeyed by the staggered susceptibility[8] to obtain

$$\frac{TT_1}{T_{2G}} = \left(\frac{A_\perp(\pi,\pi)}{A_\parallel(\pi,\pi)}\right)^2 \frac{\hbar c}{a}\mathcal{R}, \qquad (3)$$

where $\mathcal{R}$ is a *universal* number, computable in the $1/N$ expansion. Here $A_\parallel(\pi,\pi)$ is the in-plane hyperfine form factor which appears in the $1/T_1$ measurements of Ref. 6 The ratio $TT_1/T_{2G}$ is indeed found to be temperature independent in the data of Imai *et al.*[16] However for *quantitative* comparisons of $1/T_{2G}$ data with the theory, and for the estimate of the correlation length, Imai *et al.* used a RC expression for $\chi(q)$ (Ref. 3) modified by finite-temperature corrections in a manner first discussed by Shenker and Tobochnik.[22] Below we take an alternative approach and compare the experimental data to the QC formula for $1/T_{2G}$ which we derive here. We will see that the agreement between our theory and experiment is rather good, and thus confirm the original conclusion of Refs. 21 and 16 that the data on $1/T_{2G}$ favor QC behavior at intermediate temperatures.

As input for our calculations, we need the expression for the static susceptibility near the antiferromagnetic wave vector. In the QC region, the only energy scale is the temperature, and the scaling function for the susceptibility depends only on $\bar{q} = \hbar c q/T$. Using the results of Ref. 8, we then obtain

$$\chi(q) = \frac{N_0^2}{\rho_s}\left(\frac{NT}{2\pi\rho_s}\right)^{\eta}\left(\frac{\hbar c}{T}\right)^2 \Phi(\bar{q}), \tag{4}$$

where $N_0$ is the value of on-site magnetization at $T = 0$, and $\Phi(\bar{q})$ is a universal function given by

$$\Phi(\bar{q}) = \frac{Z}{\bar{q}^2 + m^2 + \Sigma(\bar{q})}. \tag{5}$$

Here $Z$ is a rescaling factor, $m$ is proportional (but not exactly equal) to inverse correlation length, and $\Sigma(\bar{q})$ is a self-energy given by

$$\Sigma(\bar{q}) = T\sum_n \int \frac{d^2\bar{k}}{4\pi^2}\frac{G(\bar{k}+\bar{q},\bar{\omega}_n) - G(\bar{k},\bar{\omega}_n)}{\Pi(\bar{k},\bar{\omega}_n)}, \tag{6}$$

where $\bar{\omega}_n = \hbar\omega_n/T$, $G(\bar{k},\bar{\omega}_n) = 1/(\bar{k}^2 + \bar{\omega}_n^2 + m^2)$, and $\Pi(\bar{k},\bar{\omega}_n)$ is a polarization operator.

Below we calculate $1/T_{2G}$ in the two leading orders in $1/N$ expansion for $O(N)$ magnets. The physical case will be considered at the end by setting $N = 3$. To first order in $1/N$ we have[8]

$$\begin{aligned} Z &= 1 + \eta \ln\frac{\Lambda\pi}{8T}, \\ m^2 &= \Theta^2\left(1 + \eta\ln\frac{\Lambda}{T} + \frac{0.231}{N}\right), \\ \Sigma(\bar{q}) &= \bar{q}^2\left(\eta\ln\frac{\Lambda}{T}\right) + \tilde{\Sigma}(\bar{q}). \end{aligned} \tag{7}$$

Here $\Theta = 2\ln[(\sqrt{5}+1)/2]$, $\Lambda$ is an upper cutoff, and $\tilde{\Sigma}(\bar{q}) \propto 1/N$ stands for the regular part of the self-energy term. Substituting Eq. (7) into Eq. (5), we observe that all $\Lambda$-dependent terms disappear as they should, so that the scaling function for $\chi(\bar{q})$ is universal. Performing then the momentum integration in Eq. (2) and numerically evaluating the contribution from $\tilde{\Sigma}(\bar{q})$, we obtain after some algebra

$$\frac{1}{T_{2G}} = \frac{A_\pi^2}{a\sqrt{4\pi}\Theta}\frac{N_0^2}{\rho_s}\left(\frac{NT}{2\pi\rho_s}\right)^{\eta}\frac{\hbar c}{T}\left(1 + \frac{0.22}{N}\right) \times\left[1 + \mathcal{O}\left(\frac{2\pi\rho_s}{NT}\right)^{1/\nu}\right], \tag{8}$$

where $\nu \sim 0.7$ is the critical exponent for correlation length. It is then convenient to reexpress the result for $1/T_{2G}$ in terms of the actual correlation length defined from the exponential ($e^{-r/\xi}$) decay of the spin-spin correlation function, or, equivalently, from the pole of the static structure factor on the imaginary $q$ axis. From the large $N$ theory of Ref. 8, deep in the QC region we have

$$\xi^{-1}(T) = \frac{T}{\hbar c}\,\Theta\left(1 + \frac{0.237}{N}\right)\left[1 + \mathcal{O}\left(\frac{2\pi\rho_s}{NT}\right)^{1/\nu}\right]. \tag{9}$$

Using Eq. (9), we can rewrite Eq. (8) as

$$\frac{1}{T_{2G}} = \frac{A_\pi^2}{\sqrt{4\pi}}\frac{N_0^2}{\rho_s}\left(\frac{NT}{2\pi\rho_s}\right)^{\eta}\frac{\xi}{a}\left(1 + \frac{0.46}{N}\right) \times\left[1 + \mathcal{O}\left(\frac{1}{N}\frac{2\pi\rho_s}{NT}\right)\right]. \tag{10}$$

The advantage of using Eq. (10) is in the form of the correction term which now has an extra factor of $1/N$; this is because at $N = \infty$ all corrections related to a deviation from pure criticality are already absorbed into the correlation length. We will assume that the remaining corrections are small and neglect them below.

We now use the values of $N_0$, $\rho_s$ and $c$ for the $S = 1/2$ Heisenberg antiferromagnet, the same value of the form factor as in Refs. 18 and 21, and rewrite the result for $N = 3$ as

$$\frac{1}{T_{2G}} \approx 0.546\,\frac{\xi}{a}\times 10^4\ \text{sec}^{-1}. \tag{11}$$

Finally for the ratio $T_1T/T_{2G}$ deep in the QC region, we obtain, using the previous theoretical result[8] for $1/T_1$,

$$\frac{T_1T}{T_{2G}} \approx 3.36\times 10^3\ \text{K}. \tag{12}$$

Note however, that $1/T_1$ is itself of the order $1/N$, and $1/N$ corrections to $1/T_1$ have not been calculated.

## III. DISCUSSION

We now turn to a comparison with the experimental results of Imai *et al.*[16] We first use Eq. (11) and infer from the data the values of the correlation length. Figure 1 presents our theoretical result obtained with no adjustable parameters together with neutron scattering data available at $T < 560$ K (Ref. 1) and the data of numerical simulations.[15,9] We see that above 700 K (where $1/T_1$ levels off to a constant value expected in the QC regime), the data inferred from the $T_{2G}$ measurements using the QC formula are in good agreement with numerical results. At lower temperatures, the actual correlation length increases faster than in Eq. (11), which is a clear signature of a crossover into the RC regime.

Next, we compare the experimental data directly with our Eq. (8) for $1/T_{2G}(T)$. The comparison requires some caution because the $\mathcal{O}(0.38J/T)$ corrections in Eq. (8) are not expected to be small. However, previous studies of $1/T_1$ have shown[8,23] that between 700 and 900 K, the experimental data are surprisingly well described by the pure QC formula with no temperature-dependent corrections. We calculated $1/T_{2G}$ in the same way, and again found surprisingly good agreement with the experimental data above 700 K (see Fig. 2). At present we have no ex-

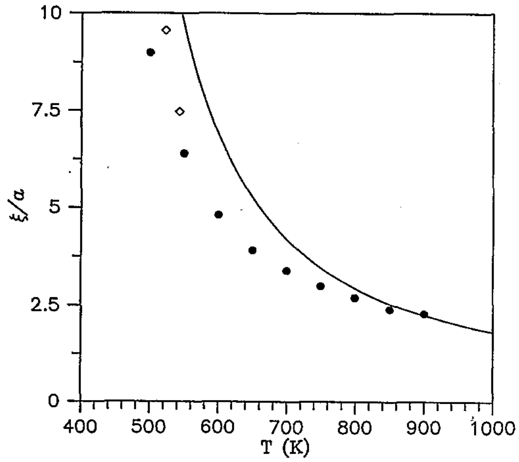


FIG. 1. The correlation length vs the temperature. Solid circles are the data inferred from the experiments on $1/T_{2G}$ (Ref. 16) using the QC formula, Eq. (11). Diamonds are the neutron-scattering data of Ref. 1. The line represents the results of numerical studies (Refs. 15, 26, and 9) at $J = 1500$ K.

planation why $\rho_s/T$ corrections to $T_1$ and $T_{2G}$ are small at $T \sim 0.5J$; it is however difficult to be more quantitative without calculating $1/N$ corrections to subleading term, which has not been done. The comparison of the slope of $1/T_{2G}(T)$ with our QC formula requires more caution as on one side, at $T < 650$ K, the system crosses over to the renormalized-classical regime, while on the other, numerical studies of $1/T_{2G}$ in an $S = 1/2$ Heisenberg antiferromagnet[9] show that above 900 K, lattice corrections become important. Between these two temperatures, the slope of the theoretical $1/T_{2G}(T)$ agrees with the data (see Fig. 2), though the agreement is not as good as for the magnitude.

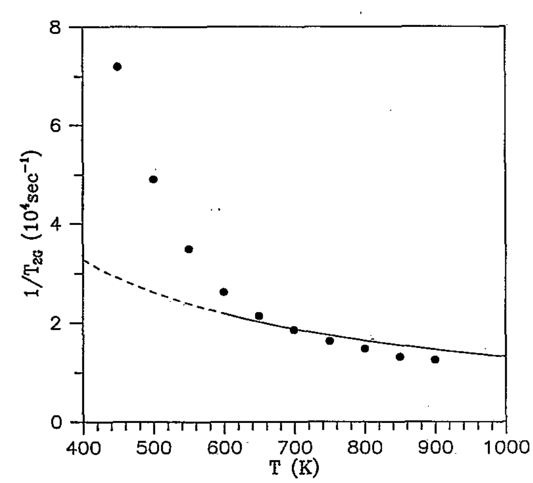


FIG. 2. Experimental and theoretical results for the spin-echo decay rate $1/T_{2G}$. The solid circles are the experimental data of Imai *et al.* (Ref. 16). The asymptotic QC result, Eq. (8), is shown as solid line in the QC region, $T > 600$ K, where the agreement with the experiment is expected.

The better agreement we found for the magnitude of $T_{2G}(T)$ than for the slope is consistent with numerical results which indicate that the lattice corrections to the slope of $1/T_{2G}(T)$ are relatively larger than the corrections to its magnitude. The numerical results are presented in Fig. 3. The two curves in Fig. 3 are the data for $T_{2G}$ vs the temperature, obtained using the full Eq. (2) (these data agree with the experiment for all temperatures measured), and its truncated version without nonuniversal corrections due to the second term and to the momentum dependence of the form factor: $\widetilde{T}_2 = aA_\pi^{-2}[\int d^2q\ \chi^2(q)/4\pi^2]^{-1/2}$. We recall that these nonuniversal corrections can be neglected only if the correlation length well exceeds the interatomic spacing.

It is clear from the data that in the region of spin-echo decay measurements $T_{2G}$ and $\widetilde{T}_2$ are quite close to each other, so that the absolute value of $T_{2G}$ (and hence $\xi$) inferred from the experimental data should be consistent with the long-wavelength description. At the same time, the slopes of $T_{2G}(T)$ and $\widetilde{T}_2(T)$ differ already by the factor of 1.7 at $0.6J$ ($\sim$ 900 K) which means that it is less reasonable to compare the experimentally measured slope of $T_{2G}(T)$ with the theoretical formula than to compare its magnitude.

Finally, our theoretical result for $T_1T/T_{2G}$, Eq. (12), also agrees satisfactorily with the experimental value $T_1T/T_{2G} \sim 4.3 \times 10^3$ K. The difference is chiefly due to the theoretical result for $1/T_1$, which is a bit larger than the experimental result.[8] Note also that the experimental values of $T_1T/T_{2G}$ remain temperature independent even at smaller temperatures, where the correlation length already fits the RC formula. At the same time, deep in the RC region, one has $T_1T/T_{2G} \propto T^{1/2}$.[3,21] This disagreement is not surprising as the RC result for $T_1T/T_{2G}$ assumes a temperature-independent uniform susceptibility $\chi_u$.[3] Numerical studies however indicate[15] that $\chi_u$ does not saturate until very low $T \leq 0.2J \sim 300$ K. It is therefore likely that in the temperature range of experimental comparisons here, the highly nontrivial downturn renormalization of the spin-wave velocity, which leads to $c(T) \sim \sqrt{T}$ at $T \to 0$,[3] does not occur and one has $T_1T/T_{2G} = \text{const}$ even when $\xi$ is given by Eq. (1).

We now address the issue of whether it is possible to extend the QC behavior above $0.6J$. This issue is probably irrelevant for experimental studies, as no experiments

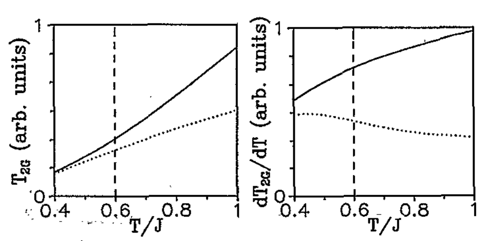


FIG. 3. The series expansion results (Ref. 9) for the spin-echo decay rates $T_{2G}$, calculated using Eq. (2) (solid line), and $\widetilde{T}_2$, calculated without the nonuniversal second term in (2) and without the momentum dependence of the prefactor (dotted lin ).

have been done above 900 K, but it is nevertheless important for the interpretation of numerical data. In particular, it has been recently shown that the numerical data for the correlation length at $0.6J < T < J$ can be fitted by either by a modified RC expression,[24,25] or an inverse linear dependence as in the QC regime.[8,9] However, the slope of the inverse linear dependence in the QC fit was nearly twice as large as in Eq. (9). Our point of view is that above $T \sim 0.6J$, a mean-field description, similar in spirit to the $N = \infty$ approach is possible, but, as we discussed in the Introduction, it should definitely include nonuniversal $T/J$ terms. There are at least two sets of data which support the above conjecture. The first set are the data for the uniform susceptibility[14,15,26] which clearly indicate that above $T \sim 0.6J$ the susceptibility tends to approach a broad maximum produced by short-wavelength fluctuations. The second set of data, shown in Fig. 3, are the numerical results for $T_{2G}$.[9] We see that above $0.6J$, not only the slopes but also the absolute values of $T_{2G}$, $\widetilde{T}_2$ begin to differ substantially and their ratio reaches a value of $\sim 1.7$ at $T = J$. A similar nonuniversal behavior is likely to hold for the correlation length above $0.6J$, although we cannot also exclude the possibility that the RC formula for the correlation length extends to higher temperatures than for other observables. The latter is however unlikely in view of present results and recent numerical results for doped antiferromagnets.[9]

To conclude, in this paper we have presented the theoretical expression for the spin-echo decay rate in the QC region of 2D antiferromagnets. We compared our QC result with the experimental data of Imai *et al.*[16] and found good quantitative agreement in the temperature range between $700 < T < 900$ K. The temperature dependence of the correlation length inferred from the $T_{2G}$ data is in good agreement with neutron scattering and numerical data. We have also argued that above $T \sim 0.6J$, lattice effects are relevant and the use of the universal low-temperature expressions for observables is unlikely to be justified.

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