

Nearly critical ground state of $\text{LaCuO}_{2.5}$

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Using a combination of analytical techniques and quantum Monte Carlo simulations we investigate the coupled spin-ladder system $\text{LaCuO}_{2.5}$. At a critical ratio of the interladder to intraladder coupling $(J'/J)_c \approx 0.11$ we find a quantum phase transition between a Néel ordered and a disordered state. At criticality the uniform susceptibility behaves as $\chi(T) = aT^2$ with a universal prefactor. At intermediate temperatures the system crosses over to a ‘‘decoupled ladders regime’’ with pseudogap-type behavior, similar to uncoupled ladders. This can explain the gaplike experimental data for the magnetic susceptibility of $\text{LaCuO}_{2.5}$ despite the presence of long-range Néel order. [S0163-1829(97)50910-6]

The unusual normal-state magnetic properties of doped high- T_c cuprates have led to enhanced interest in zero-temperature order-disorder transitions of quantum magnets. In particular, detailed predictions have been made about the behavior of a two-dimensional (2D) Heisenberg antiferromagnet by mapping it to the nonlinear sigma model.¹ They are in good agreement with experimental measurements on La_2CuO_4 . In addition to various mechanisms proposed for 2D spin systems, long-range Néel order at $T=0$ can also be destroyed if a 3D antiferromagnet approaches the 1D limit due to spatially anisotropic exchange. Then, quantum critical behavior and a disordered spin-liquid phase should be observed in three spatial dimensions.

Recently, a suitable system for such type of behavior, $\text{LaCuO}_{2.5}$, has been synthesized.² The copper atoms in this compound form an array of coupled spin-1/2 two-chain ladders. Isolated spin ladders have a spin-liquid ground state and show signs of superconducting pairing with a d -wave order parameter upon doping³. However, a marked transition to a metallic phase takes place in $\text{LaCuO}_{2.5}$ under Sr doping, but no sign of superconducting pairing was observed down to 5 K.² In contrast superconductivity was recently found in the ladder compound $\text{Sr}_{0.4}\text{Ca}_{13.6}\text{Cu}_{24}\text{O}_{41.84}$,⁴ which has weak and frustrated interladder couplings. This observation makes it quite important to study in detail the influence of the interladder coupling on the magnetic properties of the undoped insulating phase.

First, susceptibility measurements on $\text{LaCuO}_{2.5}$ were interpreted as showing a spin gap in the excitation spectrum.² Subsequent NMR and μSR studies indicated, in contrast, antiferromagnetic ordering below $T_N \sim 110$ K.^{5,6} Normand and Rice⁷ suggested that the magnetic state could be close to a transition to spin-liquid phase. In this paper we expand on this idea and show that the apparently conflicting experimental results can be reconciled.

The basic model for understanding these properties of $\text{LaCuO}_{2.5}$ is a spin-1/2 Heisenberg Hamiltonian for coupled ladders⁷

$$\hat{\mathcal{H}} = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J' \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (1)$$

which are shown schematically in Fig. 1. We assume for simplicity equal rung and leg exchange constants J in each ladder and different exchange J' between ladders. Notice that the crystalline structure of $\text{LaCuO}_{2.5}$ is more complicated, having four spins per unit cell.² However, we may choose a simpler, topologically equivalent lattice structure having only two spins per unit cell. For $J' \approx J$ the spin system is three-dimensional and has Néel order at low temperatures because the interladder coupling does not introduce

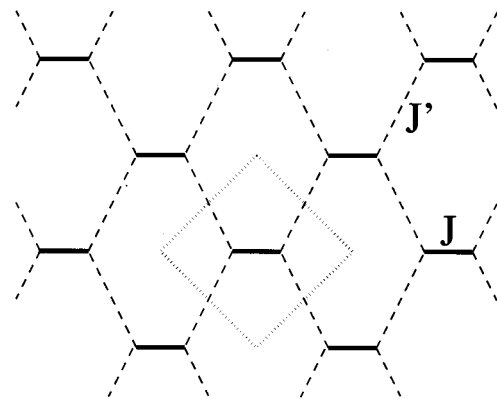


FIG. 1. Cross section of the lattice structure of the model. The ladders run perpendicular to the paper plane. Solid lines are the rungs of the ladders with a coupling J . Dashed lines are the interladder couplings J' . The dotted lines indicate the unit cell used.

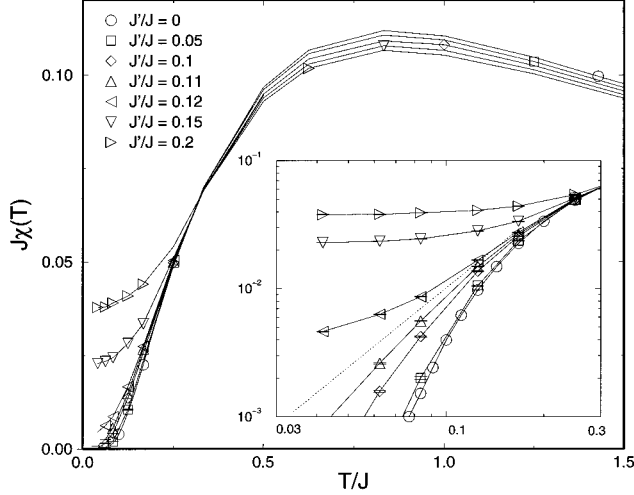


FIG. 2. Uniform susceptibility calculated by QMC for some representative ratios of the couplings. Error bars were omitted where the relative error was less than 1%. The inset is the same data in a double logarithmic plot. The dotted line is added as a guide to the eye, indicating the critical T^2 behavior clearly.

frustration. Quantum fluctuations become more and more significant as one approaches the quasi-1D limit. Since the 1D phase is a spin liquid with a finite gap, the magnetic order is destroyed at some finite J' .

We examine the following points: (i) the critical ratio of $(J'/J)_c$ for the order-disorder transition, (ii) the low- T behavior of the uniform susceptibility χ at the critical point, and (iii) $\chi(T)$ in the whole temperature range and for arbitrary J'/J . For this we employ a combination of analytical and numerical techniques. With the help of the renormalized spin-wave theory⁸ and the bond-operator method⁹ we obtain lower and upper bounds for the transition point: $0.05 < (J'/J)_c < 0.12$. The quantum critical behavior of the uniform susceptibility for a 3D spin system has been predicted from scaling arguments by Chubukov *et al.*¹ as $\chi(T) = aT^2$. We calculate a universal factor in this law. Employing a quantum Monte Carlo cluster algorithm¹⁰ (QMC) we then obtain a better estimate for the critical coupling: $0.11 < (J'/J)_c < 0.12$. Next we calculate the temperature dependence of the uniform susceptibility $\chi(T)$ for the whole temperature range and various coupling ratios, shown in Fig. 2. Finally we show that the susceptibility measurements of Hiroi and Takano² can be fitted perfectly by the predicted form for a nearly critical ordered system, thus resolving the apparent contradiction between the susceptibility and magnetic resonance measurements.

A natural approach to the Hamiltonian (1) from the side of strong interladder coupling $J' \sim J$ is the renormalized spin-wave theory of antiferromagnets.⁸ Following a slightly different procedure, we express the two spins per unit cell via two types of boson operators a_i and b_i using the antiferromagnetic Dyson-Maleev transformation. Interaction terms with four bosons are then treated in the mean-field approximation by introducing boson averages: $m = \langle a_i^\dagger a_i \rangle$, $\Delta_1 = \langle a_i b_i \rangle$, $\Delta_2 = \langle a_i a_{i+z} \rangle$, $\Delta_3 = \langle a_i b_{i+x} \rangle$, which are determined by solving self-consistent equations. The corresponding spin-wave spectrum consists of two branches,

$$\omega_{\mathbf{k}} = \sqrt{A^2 - (B_{\mathbf{k}} \pm |C_{\mathbf{k}}|)^2}, \quad (2)$$

$$A = J(S - m + \Delta_1) + 2J(S - m + \Delta_2) + 2J'(S - m + \Delta_3),$$

$$B_{\mathbf{k}} = 2J(S - m + \Delta_2) \cos k_z,$$

$$C_{\mathbf{k}} = J(S - m + \Delta_1) + 2J'(S - m + \Delta_3)(e^{ik_x} + e^{iky}),$$

each having zero-frequency mode at $k_z = 0$ or π . At the isotropic point $J' = J$, our calculations predict for $S = 1/2$ only a small reduction of the sublattice magnetization: $\langle S \rangle = 0.40$. Quantum fluctuations destroy the magnetic order for the critical coupling $J'_c \approx 0.05J$. (The result by the linear spin-wave theory is an order of magnitude smaller.) From general arguments we expect that the renormalized spin-wave theory overestimates the stability region of the ordered phase and, hence, $J'_c \approx 0.05J$ presents a *lower* bound for the exact critical value.

In the ordered phase the uniform magnetic susceptibility becomes anisotropic with two components parallel and perpendicular to the staggered moments. The parallel component χ^{\parallel} vanishes at $T=0$. We calculate its low-temperature behavior in the framework of the present approach by using

$$\chi^{\parallel} = \frac{1}{T} \sum_j \langle S_i^z S_j^z \rangle. \quad (3)$$

In the limit $T \rightarrow 0$ we find in agreement with Oguchi's results:⁸ $\chi^{\parallel} = T^2/6c_{\parallel}c_{\perp}^2$, where c_{\parallel} and c_{\perp} are the two spin-wave velocities determined from Eq. (2). The numerical coefficient in the square-law behavior of $\chi(T)$ increases by a factor of 20 between $J' = J$ and J'_c .

Describing correctly transverse oscillations in the ordered phase, spin-wave theory fails, however, in the vicinity of J'_c since at the critical point excitation spectrum has the same triplet degeneracy as in the disordered singlet phase for $J' < J'_c$. To study the order-disorder transition from the opposite side, we use the bond operators formalism.⁹ This method describes a single spin ladder fairly well for strong enough rung coupling.⁷ It has also been applied to a 3D array of ladders in $\text{LaCuO}_{2.5}$ at $T=0$, but the result of Ref. 7 is different from ours.

The two spins ($n=1,2$) belonging to the same ladder's rung with the lattice index i are expressed in terms of dimer states as

$$S_{n,i}^{\alpha} = \frac{(-1)^n}{2} (s_i^{\dagger} t_{\alpha,i} + t_{\alpha,i}^{\dagger} s_i) - \frac{i}{2} e^{\alpha\beta\gamma} t_{\beta,i}^{\dagger} t_{\gamma,i}, \quad (4)$$

where s_i and $t_{\alpha,i}$ are singlet and triplet boson operators subject to the constraint $s_i^{\dagger} s_i + \sum_{\alpha} t_{\alpha,i}^{\dagger} t_{\alpha,i} = 1$. This relation is enforced by a chemical potential μ . Also, a site independent condensate of singlets $\langle s_i \rangle = \bar{s}$ is assumed. In the quadratic approximation we keep only the terms with two triplet operators. Diagonalizing the remaining Hamiltonian by the Bogoliubov transformation we obtain two self-consistent equations $\langle \partial \mathcal{F}_{\text{quad}} / \partial \mu \rangle = 0$ and $\langle \partial \mathcal{F}_{\text{quad}} / \partial \bar{s} \rangle = 0$ on the parameters μ and \bar{s} . They can be reduced to a single equation on the new parameter $d = 2J\bar{s}^2/(J/4 - \mu)$:

$$d = 5 - 6 \sum_{\mathbf{k}} \frac{1}{\sqrt{1 + d\nu_{\mathbf{k}}}} \left(n_{\mathbf{k}} + \frac{1}{2} \right), \quad (5)$$

where $\nu_{\mathbf{k}} = \cos k_z - J'/2J(\cos k_x + \cos k_y)$, magnon dispersion is $\omega_{\mathbf{k}} = (J/4 - \mu)\sqrt{1 + d\nu_{\mathbf{k}}}$, and $n_{\mathbf{k}}$ is a Bose factor. We first solve Eq. (5) at $T=0$. The gap becomes zero for $d = 1/(1 + J'/2J)$. Substituting this value into Eq. (5) we find that the critical coupling corresponding to vanishing gap and to the transition to the ordered phase is $J'_c = 0.121J$. The mean-field theory should again overestimate the stability region of the corresponding phase. Therefore, we conclude that the above value is an *upper* bound for the exact value of J'_c , which lies between 0.05 and 0.12. We will find below from QMC that the exact critical coupling is very close to the upper bound. The spectrum of low-lying excitations in the disordered phase near $(0,0,\pi)$ has the form $\omega_{\mathbf{k}} = c_{\parallel}\sqrt{k_z^2 + p^2k_{\perp}^2 + m^2}$, where c_{\parallel} and $c_{\perp} = pc_{\parallel}$ are spin-wave velocities parallel and perpendicular to ladders, $c_{\parallel} \approx 1.16J$ (at $J' = J'_c$), and $p = \sqrt{J'/2J}$. The mass m and the gap $\Delta = c_{\parallel}m$ behave like $(J'_c - J')^{1/2}$ close to the critical point.

The isotropic susceptibility in the spin singlet state can be calculated by Eq. (3), which after substitution of Eq. (4) takes the form

$$\chi = \frac{1}{T} \sum_{\mathbf{k}} (n_{\mathbf{k}}^2 + n_{\mathbf{k}}), \quad (6)$$

where summation is performed over one of the three magnon branches only.

If the temperature is smaller than the gap, one can use the zero-temperature spectrum. In this quantum disordered regime the asymptotic behavior of the susceptibility found from Eq. (6) is

$$\chi(T) = \frac{\Delta^{3/2} T^{1/2}}{(2\pi)^{3/2} c_{\parallel}^2 c_{\perp}^2} e^{-\Delta/T}, \quad (7)$$

which differs by its prefactor from the analogous results for magnetically disordered phases in 1D and 2D.¹¹

At $J' = J'_c$ the mass m is generated by thermal fluctuations. It can be found from the self-consistency equation at finite T . In contrast to the 2D case,¹² variation of the zero-point fluctuation term in Eq. (5) becomes logarithmically divergent on the upper limit, and is, therefore, lattice dependent. Accordingly, m is a linear function of T with a logarithmically small prefactor computed by evaluating lattice sums:

$$\frac{c^2 m^2}{T^2} = \frac{2\pi^2}{3 \ln(0.7J/T)}. \quad (8)$$

To calculate the universal behavior of the uniform susceptibility in the quantum critical region $\Delta \ll T \ll J'$ we should neglect logarithmically small mass and substitute the gapless dispersion into Eq. (6). As a result, the universal form for the susceptibility coincides with the result for χ^{\parallel} obtained in the spin-wave theory:

$$\chi(T) = \frac{T^2}{6c_{\parallel}c_{\perp}^2}. \quad (9)$$

Notice that nonuniversal corrections to the prefactor in the above expression have only logarithmic smallness.

Analogous calculations for the specific heat predict $C(T) = 2\pi^2 T^3 / 5c_{\parallel}c_{\perp}^2$ at the critical point. The temperature dependence coincides again with the behavior in the ordered phase. However, the prefactor is multiplied by 3/2 according to the different number of gapless modes in the two phases. Consequently, a crossover between these two regimes should exist for a ‘‘nearly critical’’ ordered spin system.

Critical behavior can be also studied using a sigma model description of quantum antiferromagnets.¹ Predictions of that method have been compared with bond-operator results for a 2D magnet in Ref. 12. By analogy we argue that the limit $N \rightarrow \infty$ of the $O(N)$ quantum nonlinear σ model in 3+1 dimensions should give the same universal factor as in Eq. (9). This is quite natural since both approaches use mean-field approximation. Calculation of leading $1/N$ corrections to the mean-field prediction remains an open question.

Using QMC we can obtain a better estimate for the critical coupling. We have calculated the uniform susceptibility $\chi(T)$ for various couplings on lattices up to 10×10 ladders of length 40 (8000 spins) and periodic boundary conditions at temperatures down to $\beta J = 24$. The results are shown in Fig. 2. We estimate the critical coupling by varying the coupling ratio and looking for the predicted T^2 behavior at criticality. Taking into account the shift of the critical point due to finite-size effects¹³ we estimate: $0.11 < (J'/J)_c < 0.12$, very close to the bond-operator estimate.

Additionally we use self-consistent field boundary conditions¹⁴ to probe the occurrence of Néel order and to estimate Néel temperatures. We find $T_N \approx 0.38(3)J$ at $J'/J = 0.25$, $T_N \approx 0.27(3)J$ at $J'/J = 0.15$ and no indication for order down to $T = J/16$ at $J'/J = 0.1$. These results are consistent with the above estimates and show that the Néel temperature of about 110 K ($\approx J/10$) observed in the experiments is realized *very close* to the critical point.

Next we want to discuss $\chi(T)$ for the whole temperature and coupling range and compare with the experimental measurements. For all couplings the Curie behavior at high temperatures changes over into a broad maximum at temperatures of the order of J , caused by local spin singlet formation on the individual ladders, just as in uncoupled ladders.¹¹ The single ladder then shows a steep decrease with lowering the temperature, following an exponential decay $\chi(T) \sim T^{-1/2} e^{-\Delta/T}$ (Ref. 11) with a gap of about $0.5J$ at low temperatures.³

A weak coupling between the ladders does not destroy the spin gap. At high and intermediate temperatures we observe the same behavior and a steep exponential decrease with a *pseudogap* similar to the gap of the single ladder. Only at temperatures of the order of J' does a crossover to the 3D quantum disordered behavior Eq. (7), an exponential decay with the *actual gap*, take place.

When Δ becomes smaller than J' upon approaching the $T=0$ transition point, the quantum critical region¹ with its T^2 law for the susceptibility appears between the quantum disordered and decoupled ladders regimes. Note, that existence of the 3D-type quantum critical behavior is restricted to quite low temperatures $T < J'$. At $T > J'$, when interladder coupling can be neglected, $\chi(T)$ still shows a remarkable similarity to the single ladder.

In the ordered phase close to criticality we find the same pseudogap behavior, but the susceptibility goes to a small but nonzero value at zero temperature. The crossover occurs at temperatures of the order of the Néel temperature (compare $J'/J=0.15,0.2$ in Fig. 2).

Let us now fit the susceptibility measurements on $\text{LaCuO}_{2.5}$. Hiroi and Takano have fitted them to an exponential form plus a Curie contribution due to impurity spins, and thus concluded a disordered ground state. But, as the magnetic resonance measurements indicate an ordered ground state the correct low-T behavior is

$$\chi(T) = C/(T - \Theta) + \chi_0 + aT^2/J^3, \quad (10)$$

where $a \approx 0.33(3) \text{ emu mol}^{-1}$ estimated from QMC, and χ_0 is the sum of the temperature independent core susceptibility, and van Vleck susceptibility, and the small zero-temperature spin susceptibility. The fit is excellent, as shown in Fig. 3, with fitting parameters $C = 1.8(1) \times 10^{-3} \text{ emu mol}^{-1}$, $\Theta = -6.0(4) \text{ K}$, $\chi_0 = -6.2(2) \times 10^{-6} \text{ emu mol}^{-1}$. $J = 1340 \pm 150 \text{ K}$, similar to the value in the planar copper oxide compounds.

We see that the uniform susceptibility measured by Hiroi and Takano² is indeed compatible with a gapless ordered ground state close to quantum criticality, as suggested by Normand and Rice.⁷ We remark that due to the dominance of quantum fluctuations in this nearly critical system no anomaly can be observed at the Néel temperature.

Measurements of the total susceptibility suffer from the Curie contribution of impurity spins at low temperatures, which make the extraction of the asymptotic $T \rightarrow 0$ behavior

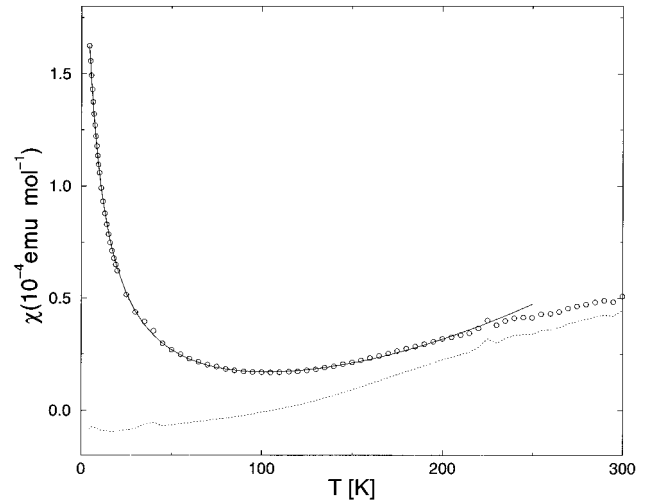


FIG. 3. Fit of the susceptibility measurements by Hiroi and Takano (Ref. 2) to Eq. (10). The circles denote the measurements, the solid line the fit, and the dashed line the measurements after subtraction of the Curie term.

difficult. Thus measurements which are not sensitive to impurities, such as NMR or μSR are much better in distinguishing nearly critical ordered magnetic materials from disordered ones.

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