Universal Magnetic Properties of $La_{2-\delta}Sr_{\delta}CuO_4$ at Intermediate Temperatures

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We present the theory of two-dimensional, clean quantum antiferromagnets with a small, positive, zero temperature (T) stiffness ρ_s, but with the ratio k_BT/ρ_s arbitrary. Universal scaling forms for the uniform susceptibility (χ_u), correlation length (ξ), and NMR relaxation rate ($1/T_1$) are proposed and computed in a $1/N$ expansion and by Monte Carlo simulations. For large k_BT/ρ_s, $\chi_u(T)/T$ and $T\xi(T)$ asymptote to universal values, while $1/T_1(T)$ is nearly T independent. We find good quantitative agreement with experiments and some numerical studies on $La_{2-\delta}Sr_{\delta}CuO_4$.

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The last few years have seen extensive theoretical and experimental studies of two-dimensional quantum Heisenberg antiferromagnets, with particular attention to the antiferromagnetism in the cuprate compounds [1,2]. On the theoretical side, most notable has been the work of Chakravarty, Halperin, and Nelson [3], who focused mainly on the low temperature (T) properties of systems with well established long-range Néel order at $T = 0$; their most detailed results were in a regime in which the fully renormalized, $T = 0$, spin stiffness ρ_s was not too small, while the temperature satisfied $k_BT \ll \rho_s$. Under these conditions, the antiferromagnet could be treated as a classical system, with all effects of quantum fluctuations being absorbed into renormalization of the couplings. At low T, there has been good agreement between their results and experiments on La_2CuO_4 [3]. However, the experimental results at higher T remain poorly understood—there are clear deviations from the classical behavior and it is expected that quantum fluctuations will play a more fundamental role. Besides, in the lightly doped cuprates, ρ_s is likely to be quite small, thus decreasing the T range over which the renormalized-classical (RC) behavior will hold. Finally, there are experimental realizations of frustrated two-dimensional Heisenberg antiferromagnets [4], which, in all likelihood, have a very small value of ρ_s.

Our understanding of the experiments would clearly be improved by precise theoretical predictions in low temperature regimes other than $k_BT \ll \rho_s$. To this end, we discuss here some universal properties of clean two-dimensional quantum Heisenberg antiferromagnets with nearest-neighbor exchange J, in which the stiffness ρ_s is "small," but nonzero. We will study the physics when $0 < \rho_s \ll J$, $k_BT \ll J$, but the ratio k_BT/ρ_s is allowed to be *arbitrary*. The system is then controlled by renormalization-group flows near the $T = 0$ quantum fixed point separating the Néel-ordered and quantum-disordered phases. Our main new result will be that, in this regime, the absolute values of the entire long-wavelength, low-frequency, uniform and staggered spin susceptibilities are completely universal functions of just three thermodynamic parameters: ρ_s, c, and the ordered staggered moment N_0. The universal functions depend *only* on the symmetry of the order parameter, and sensitivity to all lattice-scale physics arises only through the values of ρ_s, c, and N_0. For small k_BT/ρ_s (the RC region), the T dependence of our results is similar to those already obtained in Ref. [3]. For large k_BT/ρ_s [the quantum-critical (QC) region of Ref. [3]], most of our results are new. We will show that they are consistent with the available experimental [5–7] and some of the numerical [8–10] data on the uniform susceptibility, correlation length, and NMR relaxation rate for undoped and weakly doped $La_{2-\delta}Sr_{\delta}CuO_4$. We thus argue that the use of a small ρ_s point of view is not unreasonable even for the pure square lattice, spin-1/2, Heisenberg antiferromagnet; while ordered at $T = 0$, this system is evidently close to the point where long-range order vanishes.

Our results follow from some very general properties of the $T = 0$ quantum fixed point separating the magnetic-ordered and quantum-disordered phases. These properties are expected to be valid in both undoped and doped antiferromagnets, though not in the presence of randomness [11,12]. They are as follows: (i) The fixed point is described by a continuum (2+1)-dimensional field theory which is Lorentz invariant, and the spin-wave velocity, c, remains nonsingular through the phase transition. (ii) At $T = 0$, on the magnetic-ordered side, there is a Josephson correlation length ξ_J which diverges at the quantum fixed point; near this fixed point ρ_s equals $\hbar c\Theta/\xi_J$, where Θ is a universal number [13,14]. (iii) Turning on a small T places the critical field theory in a "slab" geometry which is infinite in the two spatial directions, but of finite length $L_\tau = \hbar c/k_BT$, in the imaginary time (τ) direction—its consequences therefore follow from finite-size scaling [3].

Uniform susceptibility, χ_u.—We first consider the response of the antiferromagnet to a static, spatially uniform, external magnetic field (the extension to a field at finite wave vector k or frequency ω will be omitted here for brevity). Such a field causes a uniform precession of all the spins, which can be removed by transforming to a rotating reference frame at the price of a twist in the boundary conditions along the τ direction [15]. The response of the system to this twisted boundary condition defines a stiffness, ρ_τ, which equals χ_u. However, the fixed point is Lorentz invariant, and hence χ_u has the same scaling properties as ρ_s. Application of finite-size scaling [16] then yields the following T dependence for χ_u,

$$\chi_u(T) = \left(\frac{g\mu_B}{\hbar c}\right)^2 k_B T\, \Omega_Q(x) \quad , \quad x \equiv \frac{N k_B T}{2\pi \rho_s} , \qquad (1)$$

where $g\mu_B/\hbar$ is the gyromagnetic ratio, N is the number of components of the order parameter, and $\Omega_Q(x)$ is a *universal* function. Note $x \propto \xi_J/L_\tau$, the length ratio expected in finite-size scaling functions. We have computed $\Omega_Q(x)$ in a $1/N$ expansion for the O(N) nonlinear sigma model in 2+1 dimensions [17]. The O(3) model describes the low-energy dynamics of two-dimensional Heisenberg antiferromagnets on a square lattice. The antiferromagnet also carries Berry phases, not present in the σ model, but these have been argued to be irrelevant at the quantum fixed point [11]. At $N = \infty$, the scaling function $\Omega_Q(x)$ can easily be calculated:

$$\Omega_Q^{N=\infty}(x) = \frac{1}{\pi x} + \frac{\sqrt{4+e^{-2/x}}}{\pi e^{-1/x}} \operatorname{arcsinh}\left(\frac{e^{-1/x}}{2}\right) . \quad (2)$$

Of particular interest is the behavior of χ_u for large x. The function $\Omega_Q^{N=\infty}(x)$ is analytic at $x = \infty$, and the general principles of finite-size scaling [16] suggest that this remains true at finite N. Thus we expect that $\Omega_Q(x \to \infty) = \Omega_\infty + \Omega_1/x + \cdots$ with Ω_∞, Ω_1 universal numbers. Combined with (1), this implies that a plot of $\chi_u(T)$ vs T will be a straight line at large T/ρ_s with universal slope and intercept, whose values are related to Ω_∞ and Ω_1, respectively. At $N = \infty$ we obtain from (2) $\Omega_\infty = (\sqrt{5}/\pi)\ln[(\sqrt{5}+1)/2] \approx 0.3425$ and $\Omega_1 = 4\Omega_\infty/5$. We have computed the first $1/N$ correction to Ω_∞ and indeed found that it is a universal, regularization-independent number. We obtained

$$\Omega_Q(x = \infty) \equiv \Omega_\infty = 0.3425(1 - 0.619/N + \cdots). \quad (3)$$

We have also performed Monte Carlo simulations of a *classical* $D = 3$ Heisenberg ferromagnet on a cubic lattice, whose phase transition is expected to be in the same universality class as the O(3) sigma model. We used a lattice of size $L \times L \times L_\tau$ ($L \le 30$, $L_\tau \le 10$) at its known critical coupling [18] and computed ρ_τ. It then follows that $\Omega_\infty = \lim_{L_\tau \to \infty} \lim_{L \to \infty} L_\tau \rho_\tau$, where the order of limits is crucial. The result was $\Omega_\infty = 0.25 \pm 0.04$, in good agreement with the $1/N$ result at $N = 3$. Finally, there is an analogy between Ω_∞ and another universal number discussed recently—the universal conductivity, σ_Q, at the superfluid-insulator transition [14,19].

We turn next to small x. The $N = \infty$ result (2) gives the leading term $\Omega_Q(x \to 0) = 1/\pi x$, which implies $\chi_u(T \to 0) = 2g^2\mu_B^2\rho_s/\hbar^2c^2N \equiv (2/N)\chi_\perp$, where $\chi_\perp$ is the transverse susceptibility. This is in fact equal to the exact result expected from rotational averaging of an ordered quantum O(N) sigma model [3,20]—we have indeed found no corrections in the $1/N$ expansion at $T = 0$. Furthermore, it was shown in Ref. [3] that there are no T-dependent corrections to the isotropic χ_u in a classical, $d = 2$, lattice rotor model. In contrast, for our quantum O(N) sigma model, the $N = \infty$ result contains a term linear in T at small T. In the $1/N$ expansion of this quantum model, the classical contributions to various observables appear as $\ln x/N$ terms; however, as expected, all $\ln x/N$ contributions to χ_u were found to cancel among each other. We then calculated the regular $1/N$ corrections and found

$$\Omega_Q(x \to 0) = 1/\pi x + \Gamma_N + \cdots , \qquad (4)$$

where $\Gamma_N = (N-2)/\pi N$ is a universal number.

Correlation length, ξ.—The scaling dimension of ξ is -1, and the finite-size scaling result for $\xi(T)$ is therefore

$$\xi^{-1}(T) = (k_B T/\hbar c) X_Q(x), \qquad (5)$$

where $X_Q(x)$ is a universal function. As for χ_u, there are no nonuniversal factors on the right-hand side. The numerical results for X_Q depend on the precise definition chosen for ξ: we follow Ref. [3] and define the correlation length from the long distance $e^{-r/\xi}$ decay of the equal time order parameter correlation function. Equivalently, one can define ξ as κ^{-1}, where κ is the location of the pole of the staggered structure factor $S(k)$ closest to the real k axis. At $N = \infty$, we have simply

$$X_Q^{N=\infty}(x) = 2\operatorname{arcsinh}[(1/2)e^{-1/x}] . \qquad (6)$$

For large x, the properties of ξ^{-1} are similar to those of χ_u. The function $X_Q(x)$ is expected to be analytic at $x = \infty$ with $X_Q(x \to \infty) = X_\infty + X_1/x + \cdots$; a plot of $\xi^{-1}(T)$ vs T will be a straight line at large T with a universal slope and intercept, whose values are related to X_∞ and X_1, respectively. We have also computed the $1/N$ correction to X_∞ and found $X_\infty = 0.962(1 + 0.237/N)$. From (6) it follows that $X_1 = -0.894 + O(1/N)$.

For small x, the $N = \infty$ result (6) gives $X_Q(x \to 0) = e^{-1/x}$. However, unlike χ_u, the RC spin fluctuations make a strong contribution to ξ, thus requiring careful consideration of the $\ln x/N$ terms in the $1/N$ expansion. We identified terms to order $(\ln x/N)^2$, exponentiated them, and found

$$X_Q(x \to 0) = Y_N x^{-1/(N-2)} e^{-N/(N-2)x} , \qquad (7)$$

where $Y_N = 1 + (3\ln 2 - 1 + C)/N + O(1/N^2)$ is a universal number (C is the Euler constant, $C \approx 0.5772$). The T dependence of this result agrees with that of Ref. [3]. The prefactor was also obtained by another method in Ref. [21].

Note that Ref. [3] obtained an interpolation formula for ξ in an ϵ expansion which, when reexpressed in terms of x, becomes $\xi \sim \operatorname{arcsinh}(\frac{\epsilon}{2}\, e^{-N/(N-2)x})$. Our approach shows that the $N/(N-2)$ factor in the exponent is present only at small x, where the perturbative series is logarithmic.

NMR relaxation rate, $1/T_1$.—The relaxation of nuclear spins coupled to the antiferromagnetic order parameter (e.g., Cu nuclear spins in La_2CuO_4) is given by $1/T_1(T) = \lim_{\omega\to 0} 2\tilde{A}_\pi^2 (k_BT/\hbar\omega) \int (d^2k/4\pi^2)\chi''(k,\omega)$, where $\chi''(k,\omega)$ is the imaginary part of the dynamic staggered susceptibility of the underlying quantum antiferromagnet, and $\tilde{A}_\pi$ is the bare hyperfine coupling. This determines the scaling dimension of $1/T_1$ at the quantum fixed point to be η, the critical exponent associated with spin correlations at criticality: $\eta = 8/3\pi^2N - 512/27\pi^4N^2 + \cdots$ in a $1/N$ expansion [22] and the best current value at $N = 3$ is $\eta \approx 0.028$ [18]. The finite-size scaling form for $1/T_1$ can be shown to be

$$1/T_1(T) = (2\tilde{A}_\pi^2 N_0^2/\rho_s)x^\eta R_Q(x) , \qquad (8)$$

where $R_Q(x)$ is a completely universal function. Note the complete absence of nonuniversal normalization factors in (8).

We now consider the limiting behavior of $R_Q(x)$ for large and small x. As before, at large x, $R_Q(x\to\infty) = R_\infty$, a positive constant; the small value of η then implies that $1/T_1$ is *essentially T independent* at high T. To leading order in $1/N$, we estimate R_∞ from the result for $\chi''(k,\omega \le T)$ in Ref. [11] to be $R_\infty = 0.66/N$. Note the factor of $1/N - \chi''(k,\omega)$ is finite at $\omega\to 0$ only due to the self-energy corrections. At small x, dynamical scaling [3,23] predicts that $R_Q(x) \propto \xi(x)$. The $1/N$ expansion is again singular when $x \ll 1$ and we will not discuss it here.

Comparison with numerical and experimental results.—We have so far presented general scaling forms for the magnetic properties of a two-dimensional quantum antiferromagnet which has $\rho_s \ll J$. Explicit scaling functions can be calculated at $N=\infty$, and examination of $1/N$ corrections has been limited to those for Ω_∞. These corrections were, however, quite small, and we expect, in general, that $1/N$ expansion is robust and numerically quite accurate for large values of x. On the other hand, at small x, the $1/N$ expansion is logarithmically singular, and eventually changes the leading singularity in some of the scaling functions at $x=0$; the final low-T behavior is the same as that in the RC scaling theory of Ref. [3]. The crossover between small and large x should occur for x around unity. Thus for $x \ge 1$ (but such that the long-wavelength description is still valid), it is quite likely that $1/N$ expansion will describe the experimental data better than the RC theory, which, strictly speaking, is valid only for $x \ll 1$. In a square lattice, nearest-neighbor, $S = 1/2$ Heisenberg antiferromagnet, $2\pi\rho_s \approx 1.13J$ (Ref [24]) and we therefore expect that our large-N, large x results should work for $x \ge 1$, i.e., for $k_BT \ge 0.35J$.

The absence of any RC corrections to χ_u makes it an ideal candidate for testing our theory; the $1/N$ expansion should become accurate even at fairly small values of x. We start with the numerical results for $\chi_u(T)$ on the square lattice $S = 1/2$ antiferromagnet. There have been high-T series expansions [8], quantum Monte Carlo calculations [9], and finite cluster calculations [10]. Their results all show that $\chi_u(T)$ obeys a Curie-Weiss law at high T, reaches a maximum at $k_BT \sim J$, and then falls to a finite value at $T=0$, which is rather close to the rotationally averaged $1/S$ result $(\hbar/g\mu_B)^2\chi_u(T=0) \approx 0.04/Ja^2$, where a is the lattice spacing. For $0.35J < T < 0.55J$, both series expansions [8] and Monte Carlo [9] calculations report a *linear* T dependence of $\chi_u(T)$ (Fig. 1). Also plotted is our theoretical prediction of Eqs. (2) and (3) which, over the range of x values used in the figure, is well approximated by $(\hbar/g\mu_B)^2[Ja^2\chi_u(T)] \approx 0.037x(1+\alpha/x)$, $\alpha = 0.8 + O(1/N)$. This is remarkably close to the best fit to the data of Ref. [9] which gives $0.037x(1+0.775/x)$. Moreover, the theoretical x-dependent term in the RC region is $0.014x$, in clear disagreement with the numerical data at $T > 0.35J$.

We consider next measurements of $\chi_u(T)$ in weakly doped $La_{2-\delta}Sr_\delta CuO_4$. The interpretation of the experimental data even above the zero-doping T_N requires caution because one has to subtract Van-Vleck, core, and diamagnetic contributions from the measured $\chi_u(T)$. Nevertheless, after subtraction was carried out, it was found [5,25] that at small doping concentration the susceptibility is *linear* in T; the slope of $(\hbar/g\mu_B)^2[Ja^2\chi_u(T)]$ vs x is about 0.043, which is very close to our result (0.037). Thus for $La_{1.95}Sr_{0.05}CuO_4$, χ_u scales with T in the temperature interval 100 ± 400 K (note that for 5% doping, ρ_s is already very small and the QC region is shifted to lower T).

Now the ^{63}Cu spin-lattice relaxation rate, $1/T_1$, in La_2CuO_4. At low T (x small), the theory of Ref. [23] predicts that $1/T_1 \propto x^{3/2}e^{-3/x}$; this is in very good agree-

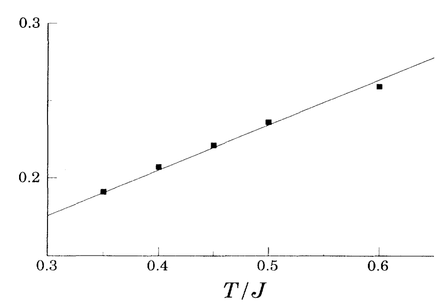

FIG. 1. Monte Carlo [9] (squares) and theoretical (line) results for the uniform susceptibility $\overline{\chi}_u = [3J(a\hbar/g\mu_B)^2]\chi_u$ of a square lattice spin-1/2 Heisenberg antiferromagnet (a is the lattice spacing). The experimental results for $La_{1.95}Sr_{0.05}CuO_4$ are very close to the Monte Carlo data [5]. There are *no* adjustable parameters in the theoretical result (1). Over the range of x plotted, the function $\Omega_Q(x)$ is very close to its large-x behavior $\Omega_Q(x) \approx \Omega_\infty(1+0.8/x)$. We used this large-$x$ result with Ω_∞ from (3) at $N=3$. The theoretical and experimental slopes agree remarkably well. The good agreement in the intercept is somewhat surprising as its theoretical value ($= 0.8$) is known only at $N=\infty$.

ment with recent observations [7]. At larger T, Ref. [23] predicted a crossover to a QC behavior. Complementing this, the present theory predicts that $1/T_1$ becomes nearly T independent for $x > 1$ or $T > 0.35J$. This has in fact already been observed in series expansions [8] and finite cluster calculations [10] for the square lattice antiferromagnet. More importantly, a flattening in $1/T_1(T)$ has recently been observed in the experiments on La_2CuO_4 [7]. We also calculated, from our results above, the limiting large-T value of $1/T_1$ for the same values of parameters as were used in the low-T fit [23] and found $1/T_1 \approx 3.3 \times 10^3$ sec^{-1}; this is in good agreement with the experimental result $1/T_1 \approx 2.7\times 10^3$ sec^{-1}. Furthermore, the experimental T range over which $1/T_1$ is nearly T independent increases upon doping. This is consistent with our results because ρ_s is expected to decrease with doping, thus pushing the system into larger x for the same T.

Finally, the correlation length ξ. Detailed measurements of $\xi(T)$ in La_2CuO_4 have been performed at low T, where the system is in the RC region [6]. At the highest experimentally accessible T (= 560 K for J = 1460 K), our result $\xi^{-1} = 0.023$ Å^{-1} is not far from the experimental value of $\xi^{-1} = 0.03 \pm 0.004$ Å^{-1}. At finite doping, we expect the crossover between two regimes to occur at lower T; QC behavior should therefore be observable at T even below 500 K. We fitted the data of [6] at $x = 0.04$ by Eq. (6) and found satisfactory agreement with the data over the T range between 300 and 550 K. At lower T, the experimental results on the dynamical local susceptibility [6,11] clearly show that the effects of randomness are relevant. We also compared our results with the numerical data for ξ at higher T [9,26]. For $T > 0.35J$ these data obey quite well $\xi^{-1} \propto x(1 - \gamma/x)$ where γ is close to 1. However, the overall factor in ξ^{-1} in the fit is close to twice our $N = \infty$ result. This discrepancy is probably due to the fact that the strong singular corrections in $X_Q(x)$ at small x cause the crossover from small- to large-x behavior to occur at a larger x. Note, however, that the Monte Carlo calculations in the QC region [27] yield the value of $X_\infty = 1.25$, which is close to our result to order $1/N$ at $N = 3$ of $X_\infty = 1.038$.

To conclude, we have considered in this paper the magnetic properties of two-dimensional quantum antiferromagnets. We focused attention on the T range where the classical low-T description is no longer valid and the behavior of observables is governed by the renormalization-group flows near the $T = 0$ quantum fixed point. A comparison with the experimental data for the uniform susceptibility and ^{63}Cu spin-lattice relaxation rate shows that the intermediate behavior has been observed in the range $0.35J < T < 0.55J$ in La_2CuO_4.

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