Low-Temperature Behavior of Two-Dimensional Quantum Antiferromagnets

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Recent neutron-scattering data for the spin-correlation length in La_2CuO_4 can be fitted quantitatively with an analysis of the quantum mechanical nonlinear σ model in two space dimensions. The coupling constant must be chosen in the range where an isolated CuO_2 layer has antiferromagnetic order at $T=0$. The parameters are consistent with the spin-wave theory for the nearest-neighbor spin-$\frac{1}{2}$ Heisenberg antiferromagnet on a square lattice.

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Recent neutron measurements by Endoh and coworkers[1,2] in single-crystal La_2CuO_4 have added a new dimension to the current excitement surrounding high-temperature superconductivity. Although the sample itself is not superconducting, it exhibits novel two-dimensional antiferromagnetic behavior. The energy scale of spin fluctuations is large, and it appears that quantum fluctuations play an essential role. Deeper understanding of this magnetic behavior may be important to the understanding of the possible new mechanism involved in these superconducting systems.[3] In this paper, we present a low-temperature renormalization-group analysis which is in quantitative agreement with the neutron experiments.

Our model is essentially a $(d+1)$-dimensional nonlinear σ model,[4] where the thickness in the extra dimension (imaginary-time direction) is proportional to the inverse temperature β of the system. The dimensionless coupling constant $\tilde{g}_0$ of the model is chosen to reproduce the results of the zero-temperature spin-wave theory for the $s=\frac{1}{2}$ nearest-neighbor Heisenberg model in $d=2$ dimensions.[5] (With this choice of $\tilde{g}_0$, the two-dimensional system has a nonzero staggered magnetization at $T=0$, although it is reduced considerably by quantum fluctuations.) There is then a single dimensional parameter, the spin-wave velocity c, which we have chosen to be $\hbar c \approx 0.425$ eV Å, close to the lower bound quoted in Ref. 1. The computed correlation length $\xi(T)$ is then in excellent agreement with the data quoted in Ref. 2 for their "best" sample (i.e., the sample with highest Néel temperature, $T_N=195$ K), in the entire temperature range $T_N < T \lesssim 550$ K. The occurrence of long-range antiferromagnetic order for $0<T<T_N$ is attributed to a very weak coupling between the Cu planes which we estimate to be of order 10^{-5} times smaller than the coupling within the planes. Such a small interlayer coupling has little effect on the spin correlations for $T>T_N$, and the staggered magnetization observed[1,6] at low temperatures should be very close to the staggered magnetization of an isolated layer at $T=0$. Indeed, the largest staggered magnetizations observed in La_2CuO_4 are close to the spin-wave estimates of $\approx 0.6\mu_B$ per site.

If the coupling constant of the $(2+1)$-dimensional σ model is increased (e.g., as a result of frustrating next-nearest-neighbor interactions), one can enter a quantum disordered regime ($\tilde{g}_0 > \tilde{g}_c$), where the isolated layer has only finite-range spin correlations at $T=0$ similar to the resonating valence-bond state in the version proposed by Kivelson, Rokhsar, and Sethna.[7] If one chooses $\tilde{g}_0=\tilde{g}_c$, the model at $T=0$ has spin correlations that fall off as an inverse power of the separation, resembling somewhat the resonating valence-bond state espoused by Anderson and others.[8,9] However, we are unable to obtain a reasonable fit to the neutron-scattering data if we choose $\tilde{g}_0 \geq \tilde{g}_c$ so that the two-dimensional system lacks long-range magnetic order at $T=0$.

The effective Euclidean action of the nonlinear σ model may be written in the form[4,10]

$$\hbar^{-1} S_{\text{eff}} = \frac{\rho_s^0}{2\hbar} \int_0^{\beta\hbar} d\tau \int d^d x \left[|\nabla \cdot \boldsymbol{\Omega}|^2 + \frac{1}{c^2} \left| \frac{\partial \boldsymbol{\Omega}}{\partial \tau} \right|^2 \right], \tag{1}$$

where $\boldsymbol{\Omega}$ is a three-component vector field with the constraint $|\boldsymbol{\Omega}|=1$, the space integrals are carried out up to a maximum wave vector Λ, τ is the imaginary time variable, ρ_s^0 is the bare spin stiffness constant on the length scale Λ^{-1}, and c is the spin-wave velocity, which is not renormalized at long wavelengths in this model. The combination $\rho_s^0 c^{-2} \equiv \chi_\perp^0$ may be identified as the local uniform magnetic susceptibility, in the direction perpendicular to the local staggered magnetization (in units where $2\mu_B/\hbar = 1$).[11] The average of $\boldsymbol{\Omega}$ in this ensemble is proportional to the staggered magnetization. For convenience, we define a dimensionless coupling constant $\tilde{g}_0$ by $g_0 = \hbar c/\rho_s^0 \equiv \tilde{g}_0 \Lambda^{1-d}$.

Renormalization-group equations for this model can be derived with the methods described by Hertz,[12] Young,[12] Polyakov,[13] and Nelson and Pelcovits,[13] and are (up to one-loop order)

$$d\tilde{g}/dl = (1-d)\tilde{g} + \tfrac{1}{2} K_d \tilde{g}^2 \coth(\tilde{g}/2\tilde{t}), \tag{2}$$

$$d\tilde{t}/dl = (2-d)\tilde{t} + \tfrac{1}{2} K_d \tilde{g}\tilde{t} \coth(\tilde{g}/2\tilde{t}). \tag{3}$$

Here e^l is the length rescaling factor, $K_d^{-1}=2^{d-1}\times\pi^{d/2}\Gamma(d/2)$, and the initial values of the dimensionless coupling constant $\tilde{g}(l)$ and temperature scale $\tilde{t}(l)$ are $\tilde{g}_0=\hbar c\Lambda^{d-1}/\rho_s^0$ and $\tilde{t}_0=k_BT\Lambda^{d-2}/\rho_s^0$. To describe a square lattice with lattice constant a when $d=2$, we shall take Λa equal to $(2\pi)^{1/2}$. This choice conserves the area of the Brillouin zone for the antiferromagnetically ordered state. Note that $\tilde{g}/\tilde{t}=\beta\hbar c\Lambda$ is the dimensionless "slab thickness" of (1) in the timelike direction, and obeys the simple recursion relation $d(\tilde{g}/\tilde{t})/dl=-\tilde{g}/\tilde{t}$.

The renormalization-group flows show that at $T=0$ there is a nontrivial fixed point at $\tilde{g}=\tilde{g}_c$ for $d>1$. This describes a *quantum* phase transition with the critical exponents of a *classical* $(d+1)$-dimensional Heisenberg model.[4] For $d\leq 2$, there are no finite-temperature fixed points, while for $d>2$, there is a fixed point describing a classical finite-temperature d-dimensional ordering transition. In this paper we shall concentrate on $d=2$. At $T=0$, Néel order persists up to $\tilde{g}_c$, where $\tilde{g}_c=4\pi$ from Eq. (2); for $\tilde{g}>\tilde{g}_c$ there is a quantum disordered phase at $T=0$ with a gap Δ in the excitation spectrum.

Although the system is disordered at all finite temperatures, we can nevertheless identify three regions separated by crossover lines as shown in Fig. 1. The central *quantum critical* region is controlled by the $T=0$ fixed point at $\tilde{g}_c$. In the *renormalized classical* region, for $\tilde{g}<\tilde{g}_c$, the correlation length ultimately diverges exponentially fast as $T\rightarrow 0$.[13] Specifically, we shall find, within our approximations,

$$\xi\approx 0.9(\hbar c/k_BT)\exp(2\pi\rho_s/k_BT), \tag{4}$$

where ρ_s is the actual spin-stiffness constant at $T=0$, renormalized by the quantum fluctuations. Since ρ_s will vanish when $\tilde{g}_0\rightarrow\tilde{g}_c$, it is clear that proximity to the $T=0$ fixed point at $\tilde{g}_c$ can greatly reduce the rate of growth of ξ. The prefactor $(\hbar c/k_BT)$ in Eq. (4) is the thermal de Broglie wavelength of the spin waves (divided by 2π). In the *quantum disordered* region $(\tilde{g}>\tilde{g}_c)$ the correlation length becomes temperature independent as $T\rightarrow 0$, and is given by $\xi(T=0)\approx(\tilde{g}_0/\tilde{g}_c-1)^{-\nu_{d+1}}$. Our one-loop calculation yields $\nu_{d+1}=1$, instead of $\nu_{d+1}\approx 0.7$, the correct result for Heisenberg models in $2+1$ dimensions. Precisely at $\tilde{g}_c$, the system will behave like a three-dimensional classical spin system at its critical point, for length scales less than the effective "slab thickness" $\beta\hbar c$. The order will be broken up by two-dimensional fluctuations on larger scales, and so we conclude that $\xi(T)\approx\hbar c/k_BT$, for $\tilde{g}_0=\tilde{g}_c$.

Next we consider the crossover lines in Fig. 1. For $\tilde{g}_0>\tilde{g}_c$, $T_x\approx\Delta/k_B\propto(\tilde{g}_0/\tilde{g}_c-1)^{\nu_{d+1}}$. For $\tilde{g}_0<\tilde{g}_c$, we obtain the crossover temperature by first defining[14] a correlation length $\xi_\perp^0\approx(\hbar c/\rho_s)^{1/(d-1)}$ which separates, at $T=0$, the long-wavelength antiferromagnetic magnons from the shorter-wavelength critical fluctuations important near $\tilde{g}_c$. This gives $T_x'\sim(1-\tilde{g}_0/\tilde{g}_c)^{\nu_{d+1}}$ by our equating $\xi_\perp^0$ to the thermal length $\hbar c/k_BT$.

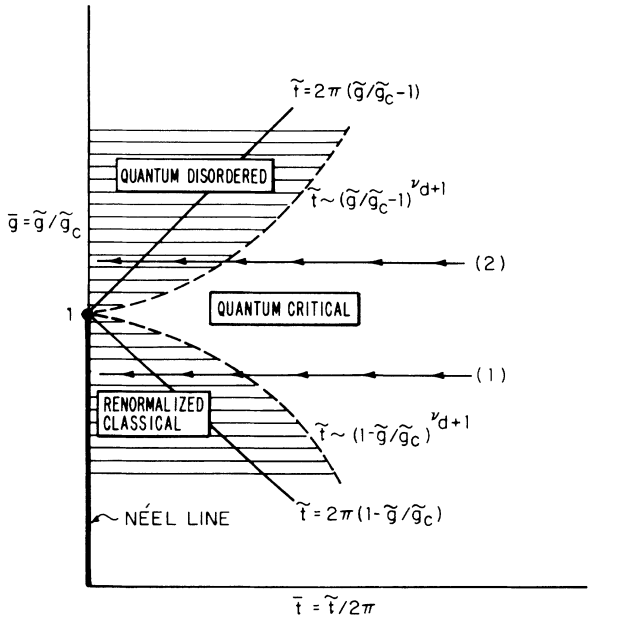

FIG. 1. Crossover phase diagram for $d=2$. $\nu_{d+1}=0.7$ for $d=2$. $\tilde{g}_c$ is the critical point of the $(d+1)$-dimensional nonlinear σ model.

The correlation length can be calculated from our recursion relations by integration of them until the renormalized correlation length $\xi(l)=e^{-l}\xi$ equals the lattice constant. Both in the renormalized classical and in the quantum critical regions, where $\tilde{t}(l)$ grows faster than $\tilde{g}(l)$, it suffices to choose l^* such that $\tilde{t}(l^*)=2\pi$. (Our results depend only weakly on the precise matching condition; see below.) Using the exact solutions of Eqs. (2) and (3) for the case $d=2$, we obtain for $\xi=ae^{l^*}$

$$\xi^{-1}=(\bar{t}_0/a\bar{g}_0)\sinh^{-1}\{\sinh(\bar{g}_0/\bar{t}_0)\exp[-(1-\bar{t}_0)/\bar{t}_0]\}, \tag{5}$$

where $\bar{g}_0\equiv\tilde{g}_0/\tilde{g}_c$, and $\bar{t}_0\equiv\tilde{t}_0/2\pi$. The prefactor $\bar{t}_0/a\bar{g}_0=(2/\pi)^{1/2}(k_BT/\hbar c)$ is not explicitly dependent on a. Other choices of the matching condition would change the coefficient $(2/\pi)^{1/2}$. At low temperatures, for $\bar{g}_0<1$, Eq. (5) becomes equivalent to Eq. (4), with $\rho_s=\rho_s^0\times(1-\bar{g}_0)$, which is the renormalized spin stiffness constant at $T=0$, within the one-loop approximation.

In the quantum region $\tilde{g}(l)$ grows faster than $\tilde{t}(l)$, and it is convenient to choose l^* such that $\bar{g}(l^*)=2$; i.e., $\tilde{g}(l^*)=2\tilde{g}_c$. This yields an implicit equation for e^{l^*}. As $\bar{t}_0\rightarrow 0$ $(\bar{g}_0>1)$ ξ approaches its $T=0$ value with corrections of order $\exp(-\Delta/k_BT)$.

To proceed further, we must determine the coupling constant $\bar{g}_0$. If we choose $\bar{g}_0<1$, and set $T=0$, then the spin stiffness ρ_s and the magnetic susceptibility $\chi_\perp=\rho_sc^{-2}$ approach finite values in the long-wavelength

limit. From the defining relation for $\tilde{g}$, we have then, for $d=2$,

$$\chi_\perp = \lim_{l\to\infty} [\hbar\Lambda e^{-l}/c\tilde{g}(l)].$$

If one integrates Eq. (2), at $T=0$, one finds that $\tilde{g}(l)=4\pi\bar{g}_0 e^{-l}/[1-\bar{g}_0(1-e^{-l})]$. Hence, we obtain $\bar{g}_0=(1+4\pi\chi_\perp c/\hbar\Lambda)^{-1}$. For a spin-$s$ antiferromagnet on a square lattice, with nearest-neighbor exchange constant J, one finds $\chi_\perp \equiv \hbar^2 Z_\chi(s)/8Ja^2$, and $\hbar c \equiv \sqrt{8}\times JsaZ_c(s)$, where the correction factors are given by[5] $Z_\chi \approx 1-0.552/2s$, and $Z_c \approx 1+0.158/2s$. Oguchi[5] finds that the $O((1/2s)^2)$ term in Z_c is small even for $s=\frac{1}{2}$, and we have made preliminary estimates which suggest that the same is true for Z_χ. Having chosen $\Lambda=(2\pi)^{1/2}/a$, we find that $\bar{g}_0=(1+\sqrt{\pi}Z_\chi Z_c/2)^{-1}=0.685$.

The value $\bar{g}_0<1$ implies that, in the absence of impurities or other sources of frustration, experiments on La_2CuO_4 should take path (1) instead of path (2) in Fig. 1; i.e., *the ground state of an isolated Heisenberg layer is Néel ordered.*

Figure 2 shows our fits to the experimental data for the inverse correlation length. With $\bar{g}_0=0.685$ (the spin-wave result) the "best fit" is obtained with $\hbar c=0.425$ eV Å; the choices $\hbar c=0.45$ eV Å and $\hbar c=0.4$ eV Å bracket the experimental uncertainties. We could have obtained comparable results directly from Eq. (4), using the formula $\rho_s=\hbar c s Z_c Z_\chi/a\sqrt{8}$. We can also fit the data allowing (20–30)% variations in Z_χ, and some further variation in $\hbar c$. In Fig. 2, we show our fit for $\hbar c=0.8$ eV Å and $\bar{g}_0=0.85$.[15] A fit for $\bar{g}_0=1$ is also shown in Fig. 2. In this case a rather large value for $\hbar c$ (>2 eV Å) is necessary to obtain the overall magnitude of ξ as seen in the experiment. However, the temperature dependence, $\xi^{-1}\propto T$, would disagree with the experiment. The disagreement is worse for $\bar{g}_0>1$.

We obtained ξ by setting $\tilde{t}(l^*)=2\pi$. If instead we used the matching condition $\tilde{t}(l^*)=4\pi$ we could fit the data with $\bar{g}_0=0.685$ and $\hbar c=0.36$ eV Å. On the other hand, for $t(l^*)=\pi$ we would need $\hbar c=0.58$ eV Å with $\bar{g}_0=0.685$. Although we have not attempted detailed calculations of the *staggered* susceptibility χ_{st}, it is easy to see that $k_B T\chi_{st}(T)\sim\xi^{1-\eta_{d+1}}$ when $\bar{g}=1$ and $\chi_{st}(T)\sim\exp[4\pi\rho_s/k_BT]$ for $\bar{g}<1$.

Calculations *beyond* one-loop order were carried out by Brézin and Zinn-Justin[16] for the classical model at $d=2$. A similar analysis of our quantum model in the renormalized classical regime suggests that in the limit $T\to 0$, for fixed $\bar{g}_0<1$, the pre-exponential factor in Eq. (4) for ξ will be relaced by const$\times\hbar c/2\pi\rho_s$. Excellent fits to the data can be obtained with this form, e.g., with $2\pi\rho_s=1175$ K, corresponding to $\hbar c\approx 0.67$ eV Å in the spin-wave theory, and const$=0.6$. Moreover, since the experimental data are all within the renormalized classical regime of Fig. 1, our analysis should not be seriously affected by the incorrect value of ν_{d+1} (which determines the shape of the crossover line) inherent in a one-loop calculation.

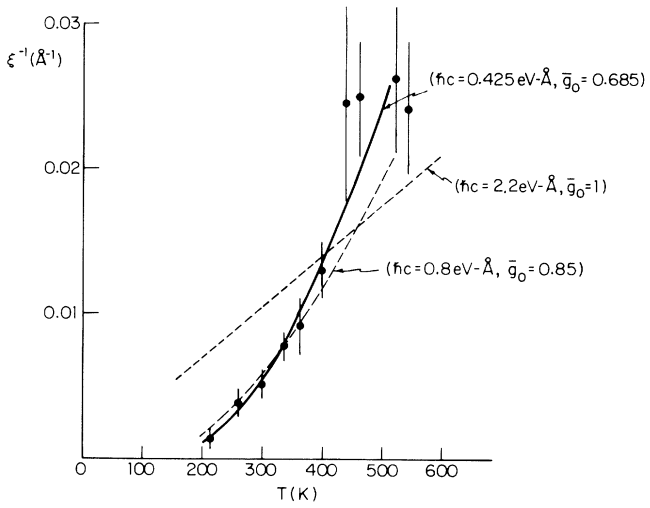

FIG. 2. Inverse correlation length as a function of temperature. The solid curve is our best fit to the data (Ref. 2) (solid circles). The dashed lines are attempts to fit the data with other values of $\hbar c$ and $\bar{g}_0$.

The experiments also show a transition to *three*-dimensional Néel order at $T_N=200$ K, which is presumably triggered by a weak exchange coupling J' between the CuO_2 planes. The transition should occur when $J'(M^2/M_0^2)(\xi^2/a^2)\approx k_B T_N$, where M/M_0 is the reduction in the $T=0$ staggered magnetization relative to the Néel value induced by 2D quantum fluctuations at length scales shorter than the in-plane correlation length ξ. Because the magnetic eigenvalues in one-loop low-temperature renormalization groups are rather inaccurate,[13] we estimate M/M_0 from $M^2\approx\rho_s$, which is a valid approximation if $\eta_{d+1}\approx 0$.[17] Taking $\bar{g}_0=0.685$, we find $M^2/M_0^2\approx 1-\bar{g}_0\approx 0.315$. (Alternatively, one could use the estimate of spin-wave theory for an isolated layer at $T=0$, which is $M^2/M_0^2\approx 0.36$.) Using the experimental results $T_N=200$ K and $\xi(T_N)\approx 200a$, we find $J'=0.015$ K. A simple mean-field treatment shows that such a small value of $J'\approx 2\times 10^{-5}J$ has a negligible effect on our estimates for ρ_s, $\chi_\perp$, and M/M_0 at $T=0$, except very close to $\tilde{g}_c$.

If we had assumed $g_0\geq 1$, so that the isolated layer has $M=0$, then it would have been necessary to choose a very large value of J', comparable to J, in order to account for the actual staggered magnetization in La_2CuO_4. [Scaling predicts that $(M/M_0)\propto(J'/J)^{(1+\eta_{d+1})/(4-2\eta_{d+1})}$, for $\bar{g}_0=1$.] A large value of J' is inconsistent with the fact that the observed spin correlations are two dimensional for $T>T_N$.

We close with a word of caution regarding our comparison between theory and experiment. Because the Néel temperature of La_2CuO_4 is extremely sensitive to

impurities and defects (e.g., oxygen vacancies), it might be incorrect to ignore the effects of impurities even in the best samples which have been studied so far. Quenched impurities become defect *rods* in the timelike direction of the effective action [Eq. (1)] which destroys the Lorentz invariance of the model. We expect that at the quantum transition point there will be new critical exponents dominated by randomness.[18]

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[9]The nonlinear σ model does not distinguish between the cases of integer and half-integer spin, so that there is some question whether its description of the quantum-disordered phase is correct in detail. [See, e.g., I. Affleck, Phys. Rev. B (to be published).] Nevertheless, it seems to us unlikely that any theoretical modification could enable one to fit the neutron-scattering data if an isolated layer were quantum disordered at $T=0$.

[10]Equation (1) is obtained from the effective $T=0$ Lagrangean proposed in Ref. 4. This is the simplest possible continuum model of quantum antiferromagnets with the correct spin-wave spectrum $\omega \propto k$ and incorporating the nonlinearities dictated by rotational invariance. For $d>2$ and $T>0$, it can be shown that this model leads to the same hydrodynamic description as that of the Heisenberg antiferromagnet developed by B. I. Halperin and P. C. Hohenberg, Phys. Rev. **188**, 898 (1969). We remark that although the ferromagnet and antiferromagnet have identical thermodynamic properties classically, their different spin-wave spectra lead to different behaviors in the quantum regime.

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