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Quantum disordered regime and spin gap in the cuprate superconductors

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We discuss the crossover from the quantum critical, $z=1$, to the quantum disordered regime in high-T_c materials in relation to the experimental data on the nuclear relaxation, bulk susceptibility, and inelastic neutron scattering. In our scenario, the spin excitations develop a gap $\Delta \sim \Delta_\xi = c/\xi$ well above T_c, which is supplemented by the quasiparticle gap below T_c. The above experiments yield consistent estimates for the value of the spin gap, which for small doping increases as the correlation length decreases.

The underdoped cuprates $YBa_2Cu_3O_{6.63}$, $YBa_2Cu_4O_8$, and $La_{1.85}Sr_{0.15}CuO_4$ display strikingly different magnetic behavior from that measured for the fully doped system, $YBa_2Cu_3O_7$. In $YBa_2Cu_3O_7$, the uniform spin susceptibility, χ_0, is essentially temperature independent, and the product $^{63}T_1T$ of the nuclear spin-lattice relaxation time and the temperature has monotonic temperature dependence; in the underdoped compounds χ_0 is temperature dependent while the temperature dependence of $^{63}T_1T$ is nonmonotonic. Recent measurements of the spin-echo decay time, T_{2G},[1] for $YBa_2Cu_3O_{6.63}$,[2] together with the earlier measurements for $YBa_2Cu_3O_{6.9}$,[3] provide additional valuable information on such *spin gap* phenomena in the underdoped systems.

Alternative physical origins of the spin gap have been proposed recently by Millis and Monien[4] and by Sokol and Pines,[5] hereafter SP. Millis and Monien[4] argued that the decreases in magnetic susceptibility measured in $YBa_2Cu_3O_{6.63}$ and $La_{1.85}Sr_{0.15}CuO_4$ are of different physical origin, with that in $YBa_2Cu_3O_{6.63}$ arising from a magnetic coupling between adjacent CuO_2 layers, while for $La_{1.85}Sr_{0.15}CuO_4$ it is attributed to the spin density wave fluctuations of a metal. On the other hand, SP applied scaling arguments to the analysis of the NMR experiments to determine the magnetic phase diagram of the cuprates. They argued that at high temperatures these materials, and the closely related system, $YBa_2Cu_4O_8$, are in the quantum critical (QC), $z = 1$, regime (see Refs. 6 and 7), characterized by a temperature-independent ratio $^{63}T_1T/T_{2G}$, and a linear dependence on T of $^{63}T_1T$,[8,7] while the common physical origin of the spin gap behavior these systems exhibit at lower temperatures is the suppression of the spectral weight of spin waves characteristic of the quantum disordered (QD) regime, for frequencies $\omega < \Delta \sim \Delta_\xi = c/\xi$. In the quantum critical regime,[6,7] $\bar{\omega} \sim \xi^{-1} \sim T$ and $z=1$, while in the quantum disordered regime ξ saturates (ξ is the antiferromagnetic correlation length, $\bar{\omega}$ the characteristic energy scale, and z the dynamical exponent). In the present paper we show that the spin susceptibility and neutron-scattering measurements on these underdoped cuprates are consistent with the SP magnetic phase diagram, suggest a way to reconcile the low-frequency spin dynamics measured by NMR with the results of neutron-scattering experiments, and give estimates of Δ and ξ.

We consider first the Knight shift and bulk susceptibility measurements. As shown in Ref. 7, in the QC, $z = 1$, regime, χ_0 is linear in T, in agreement with numerical calculations[9] as well as experiment for La_2CuO_4. In the renormalized classical regime, χ_0 is finite at $T=0$; on the other hand, in the QD regime, the susceptibility rapidly (exponentially for the insulator) decreases due to the presence of the gap in the spin excitation spectrum, and tends to zero as $T \to 0$.[7] As may be seen in Fig. 1, at high temperatures $\chi_0(T)$ displays the expected linear in T behavior for $La_{1.85}Sr_{0.15}CuO_4$, $YBa_2Cu_3O_{6.63}$, and $YBa_2Cu_4O_8$ with a magnitude which increases and a slope which decreases with increasing doping. These changes reflect the influence of the holes on the "spin-only" system.

The beginning of the crossover from the QC to the QD regime at T^* is clearly visible in the experimental data shown in Fig. 1. Below T^* (which increases with increased hole doping) χ_0 rapidly (although not exponentially in the doped case) decreases with decreasing T, reflecting the suppression of the low-frequency spin excitations. The deviation from nearly linear behavior of $\chi_0(T)$ begins at almost the same temperature as that of $^{63}T_1T$. It follows that, as expected, the spin gap influences the low-frequency dynamics both near $\mathbf{q}=\mathbf{0}$ and $\mathbf{q}=(\pi,\pi)$. The experimental finding that the ^{17}O

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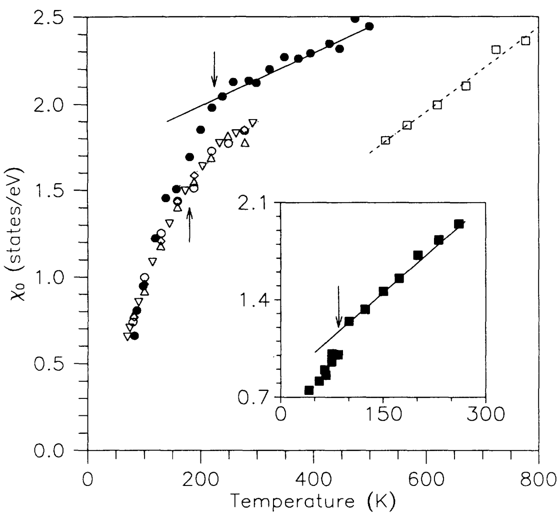

FIG. 1. The spin susceptibility for: □, La_2CuO_4 (Ref. 32), from the bulk susceptibility; ■, $La_{1.85}Sr_{0.15}CuO_4$ (Ref. 33), from the Cu Knight shift; ●, $YBa_2Cu_3O_8$ (Ref. 34), from the Cu Knight shift; ○ ($^{63}K_{ab}$), △ ($^{17}K_{ax}$), ▽ ($^{17}K_c$), ◇ ($^{17}K_{iso}$), $YBa_2Cu_3O_{6.63}$ (Ref. 10), from the Knight shift. The dotted and solid lines show linear fits to the high-temperature parts of the respective data.

spin lattice relaxation rate in $YBa_2Cu_3O_{6.63}$ (Ref. 10) and $YBa_2Cu_4O_8$ (Ref. 11) follows the temperature dependence of $\chi_0(T)$ $[(^{17}T_1T)^{-1} \sim \chi_0(T)]$ is thus consistent with our scenario. If one assumes, with SP, that the effect of the quasiparticles on the $QNL\sigma$ model parameters is somewhat less for $La_{1.85}Sr_{0.15}CuO_4$ than in $YBa_2Cu_3O_{6.63}$, and ascribes this to a smaller hole density in the former material, and also, as Dupree *et al.*[12] argue, that $YBa_2Cu_4O_8$ possesses a hole density comparable to that found in $YBa_2Cu_3O_{6.8}$, we see that the temperature at which the downturn occurs increases with hole concentration, going from $\sim$ 85 K for $La_{1.85}Sr_{0.15}CuO_4$ to $\sim$ 180 K for $YBa_2Cu_3O_{6.63}$ and $\sim$ 225 K for $YBa_2Cu_4O_8$. Moreover, since the magnitude of $\chi_0(T)$ in the QC and QD regime may be expected to increase with hole doping, this trend is also evident in Fig. 1, once one assigns a hole density in $La_{1.85}Sr_{0.15}CuO_4$ similar to that found for $YBa_2Cu_3O_{6.55}$, and takes its orbital Knight shift to be in the range 0.02% to 0.076%.

A close examination of the data shows that *two* successive downturns are visible in Fig. 1, with only the second corresponding to the superconducting transition. This supports the view that in the superconducting state of QD materials, two separate gaps are present, a charge gap which reflects the pairing of quasiparticles in the superconducting state, and a spin gap, which reflects the QD behavior.

We turn next to the evidence for a spin gap and a crossover from the QC to the QD regime found in the neutron-scattering experiments on the $YBa_2Cu_3O_{6+x}$ system. A simple phenomenological expression which can be used to parametrize both neutron-scattering and NMR data is

$$\chi(\mathbf{q},\omega) = \frac{\chi_{\mathbf{Q}}}{1+q^2\xi^2 - \frac{\omega^2}{\Delta^2} - i\frac{\omega}{\omega_{SF}}}; \tag{1}$$

in the low-frequency limit it reduces to the form introduced earlier by Millis, Monien, and Pines.[13] For $YBa_2Cu_3O_{6.6}$ the measurements of Ref. 14 show that the q-integrated (i.e. local) dynamical response function, $\chi_L''(\omega) = \int d^2\mathbf{q}\,\chi''(\mathbf{q},\omega)$, is a universal function of ω/T,[14] which behaves as $\chi_L'' \sim \omega/T$ for high temperatures (i.e. small ω/T). This behavior is consistent with the QC scaling found for this compound in the analysis of the NMR experiments.[5] Moreover, the value of $\bar{\chi}_L(\bar{\omega}) = T\chi_L(\omega)$ inferred from experiment[15] is consistent with the NMR results on $^{63}T_1$. For low temperatures, the suppression of χ_L'' for $YBa_2Cu_3O_{6.6}$, which is seen as an abrupt deviation from the universal behavior of χ_L'' as a function of ω/T, is an indication of the spin gap phenomenon. On using Eq. (1) to analyze the experimental results of Tranquada *et al.*[14] we conclude that the magnitude of the spin gap for $YBa_2Cu_3O_{6.6}$ at low temperatures is $\Delta_{T=0} \simeq 8$ meV, while from the experimental results of Rossat-Mignod *et al.*[16] for $YBa_2Cu_3O_{6.69}$ we find $\Delta_{T=0} \simeq 14$ meV. As may be seen in Fig. 2, these values are slightly smaller than those inferred by the experimentalists who used a different method of analysis.

Our conclusion that for small doping the spin gap increases as the doping increases is somewhat unexpected in view of previous analyses,[17] but may be easily understood in terms of the $QNL\sigma$ model, where $\Delta = c/\xi$, provided ξ increases faster than c as the doping decreases. One can, moreover, combine the results of neutron-scattering and NMR experiments to obtain an estimate of the doping dependence of c and ξ.

We consider first $YBa_2Cu_3O_{6.63}$, for which we estimate $\Delta_{T=0} \cong 10$ meV by interpolating between the experimental results for $YBa_2Cu_3O_{6.6}$ and $YBa_2Cu_3O_{6.69}$. This value is somewhat less than that found by applying scaling arguments to the experimental results

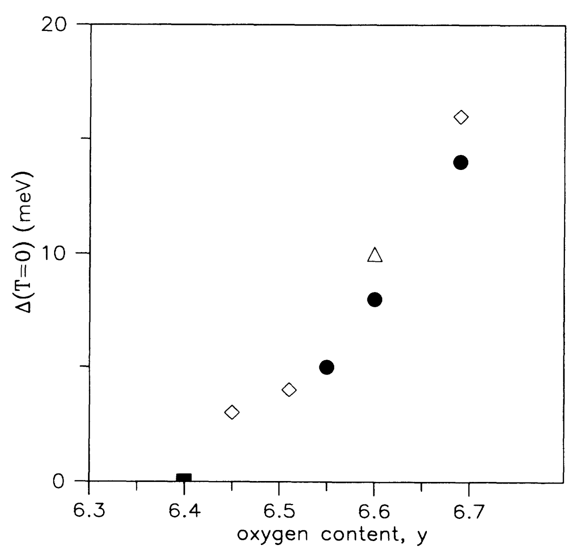

FIG. 2. The values of the spin gap in $YBa_2Cu_3O_y$ at different oxygen concentrations y determined in inelastic-neutron-scattering experiments, see the text for discussion; ●, our analysis based on Eq. (1); ■, Sato *et al.* (Ref. 15); ◇, Rossat-Mignod *et al.* (Ref. 16); △, Tranquada *et al.* (Ref. 14).

for $T_{2G}(T)$, shown in Fig. 3. Applying the finite size scaling approach,[7,18] one finds that in the QC regime, $T_{2G}(T) \sim T\left[1 + C_1(\Delta/T)^{1/\nu}\right]$, where C_1 is a universal number and $\nu = 0.7$. In the region of experimental comparison $T_{2G}(T)$ is nearly linear in temperature; similar T-linear behavior of T_{2G} in a doped antiferromagnet has been recently reported by Glenister, Singh, and one of us (A.S.).[19] On taking this universal number as that found in the leading order of $1/N$ expansion, $C_1 \simeq 0.4647$, one finds $\Delta_{T=0} = 14$ meV; the discrepancy between the two results plausibly reflects the influence of quasiparticles on scaling behavior.

A different approach is to make use of the scaling result $\chi_Q \sim \xi^{2-\eta}$, $\eta \ll 1$, and neglect any possible temperature dependence of α in $\chi_Q = \alpha\xi^2$. To the extent this holds, the full temperature dependence of $\xi(T)$ may be determined directly from T_{2G}, as shown in Fig. 3. For convenience, we introduce a dimensionless quantity, α measured in units of 15 states/eV, the value estimated by Thelen and Pines[20] for $YBa_2Cu_3O_{6.9}$; thus $\bar{\alpha}_{15} = \alpha/(15$ states/eV). We find that at low temperatures, in the QD regime, $\xi \simeq 4.14\bar{\alpha}_{15}^{-1}$, while in the QC regime, one has $\xi^{-1} = \bar{\alpha}_{15} \times 0.1(1+T/100\ \mathrm{K})$.

We examine next the extent to which the neutron-scattering experimental results on $La_{1.85}Sr_{0.15}CuO_4$ are consistent with the NMR results, the magnetic phase diagram of SP, and our proposed variation of $\Delta_{T=0}$ with hole concentration. Here there are two issues. First, while the experiments reported in Ref. 21 show no clear evidence for such a gap, from the analysis based on Eq. (1) of the experiments of Thurston *et al.*[22] we find an upturn near $\omega \simeq 5$ meV in $\chi''_L(\omega)$ at 35 K measured by Thurston *et al.*,[22] as might be expected if $\Delta_{T=0} \sim 5$ meV for this material. Second, while the incommensurate spin fluctuation peaks, at $(\pi, \pi \pm \delta)$ and $(\pi \pm \delta, \pi)$, where $\delta = 0.245\pi$, reported in Ref. 21, are not incompatible with our picture, such a large value of δ is incompatible with NMR experiments. The results of Ref. 23 confirm the prediction[24] that the hyperfine constants for the 2:1:4 systems are quite close to those inferred for the 1:2:3 systems and specifically, for $YBa_2Cu_3O_{6.63}$.[25] On taking the neutron results for $\chi''(\mathbf{q},\omega)$ it is straightforward to show that at 100 K, the copper-to-oxygen relaxation rate ratio, $({}^{17}T_1/{}^{63}T_{1,\parallel}) \lesssim 35$ while the ^{63}Cu relaxation rate anisotropy ratio, $({}^{63}T_{1,\parallel}/{}^{63}T_{1,\perp}) \sim 4.5$. These results, which reflect the leakage of correlations away from (π,π), brought about by the comparatively large value of δ, are to be compared to those found experimentally: $({}^{17}T_{1,\parallel}/{}^{63}T_{1,\parallel}) \gtrsim 80$ (Ref. 26) and $({}^{63}T_{1,\parallel}/{}^{63}T_{1,\perp}) \sim 2.6$.

Can the NMR and neutron-scattering results be reconciled? The problem is not with the magnitude of the measured spin fluctuation peaks. The value of ${}^{63}T_{1,\parallel}T$ is $\sim 5.5 \times 10^{-2}$ K sec at $T = 35$ K, when calculated from the neutron-scattering results of Ref. 21 for q-integrated intensity $\chi_L(\omega)$, assuming commensurability. This compares favorably with the NMR result, ${}^{63}T_{1,\parallel}T \sim 5.9 \times 10^{-2}$ K sec; indeed the slightly larger value at 35 K may reflect the continued upturn possibly associated with a spin gap ~ 5 meV. The problem rather lies in the assumption that the peaks are incommensurate. If instead, as suggested independently by Slichter[27] and Phillips,[28] what is being observed is discommensuration, associated with the formation of domains in the Cu-O plane, the NMR and neutron-scattering experiments can be reconciled, as shown in recent calculations by Monthoux.[29] Another experimental result lends support to this proposal: the appearance of a second ^{63}Cu resonance line in both the Sr-doped 2:1:4 systems[30] and $La_2CuO_{4.032}$,[31] which would plausibly be associated with ^{63}Cu nuclei located in or near domain walls.

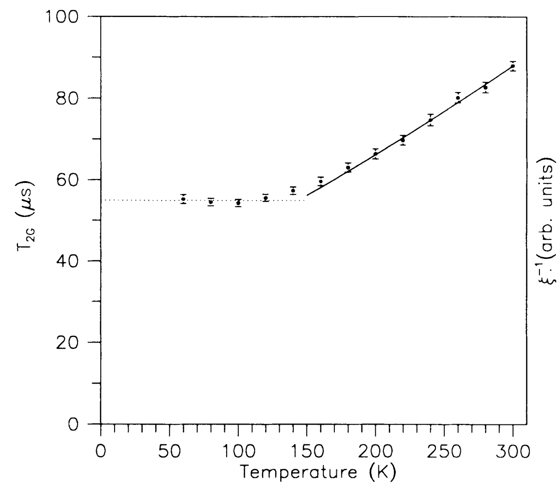

FIG. 3. The Gaussian spin-echo decay time for $YBa_2Cu_3O_{6.63}$, after Ref. 2. The solid line is the high-temperature fit to the expression: $T_{2G} \sim T[1 + C_1(\Delta/T)^{1/\nu}]$ (QC regime); the dotted line represents its saturation in the QD regime. An alternate axis shows the temperature dependence of ξ^{-1} as given by the scaling relation $T_{2G} \sim 1/\xi$.

In summary, we have shown that NMR, bulk susceptibility, and neutron-scattering data in the underdoped materials are consistent with the picture of Sokol and Pines,[5] where the spin gap onset is associated with the crossover from the QC, $z = 1$, regime to the low-temperature QD regime. The spin gap magnitudes we have deduced for $La_{1.85}Sr_{0.15}CuO_4$, $YBa_2Cu_3O_{6.63}$, and $YBa_2Cu_3O_{6.69}$ from different experiments are consistent with each other. For small doping, the magnitude of the spin gap gradually *increases* as doping increases; since the quasiparticle density also increases, this eventually leads to the smearing of the gap and a crossover to the overdamped, $z = 2$, regime in the fully doped materials.

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