Spin Gaps and Spin Dynamics in $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{7-\delta}$

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A magnetic susceptibility which decreases with decreasing temperature is observed in all CuO_2 based superconductors with less than optimal doping. We propose that in $La_{2-x}Sr_xCuO_4$ this is due to spin density wave ordering which is prevented by the low spatial dimensionality, while in $YBa_2Cu_3O_{6.6}$ it is due to the interplay between antiferromagnetic fluctuations within a plane and singlet pairing of electrons between nearest neighbor planes.

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How unusual is the normal (i.e., $T>T_c$) state of the high-T_c CuO_2 superconductors? There are two points of view: that an essentially Fermi-liquid-like picture applies, or that a fundamentally new non-Fermi-liquid physics is required. One important piece of *experimental* evidence for the existence of non-Fermi-liquid behavior is the temperature (T) dependence of the spin susceptibility χ_s. In "underdoped" high-T_c materials (i.e., those with fewer carriers than the number which optimizes T_c), χ_s *decreases* with decreasing T, in sharp contrast to the χ_s of a conventional Fermi liquid, which would be T independent.

In this paper we explain the physical origin of the observed $\chi_s(T)$. We focus on the two best studied CuO_2 systems: $La_{2-x}Sr_xCuO_4$, in which the basic structural unit is a single CuO_2 plane weakly coupled to other CuO_2 planes, and $YBa_2Cu_3O_{7-\delta}$, in which the basic structural unit is a pair of coupled CuO_2 planes. We determine the magnitude of χ_s in $La_{2-x}Sr_xCuO_4$ (about which there are conflicting assertions in the literature). Then, by analyzing and contrasting the static and dynamic spin susceptibilities in the two systems we show that in $La_{2-x}Sr_xCuO_4$ the $\chi_s(T)$ observed for $x<0.15$ is caused by a spin density wave (SDW) instability, which is prevented from developing into true long-range order by the low dimensionality and the onset of superconductivity. However, in the underdoped members of the $YBa_2Cu_3O_{7-\delta}$ series novel non-Fermi-liquid behavior occurs because of spin singlet pairing of electrons in adjacent CuO_2 planes.

The ideas we present here are related to previous work on the high-T_c problem. Johnston argued early on that incipient magnetic order explained the $La_{2-x}Sr_xCuO_4$ data [1]. SDW correlations are also important in the spin-bag model [2] and other theories of $La_{2-x}Sr_xCuO_4$ [3]. Our analysis of $YBa_2Cu_3O_{6.6}$ is related to previous work of Altshuler and Ioffe as discussed below [4], and also to the work of DaGotto, Riera, and Scalapino [5]. Magnetic correlations between planes in $YBa_2Cu_3O_{6.6}$ have been observed by Tranquada *et al.* [6]. Many authors have studied models which predict "spin-gap" behavior for electrons moving in a single CuO_2 plane, and have argued that because these theories produce a $\chi_s(T)$ similar to that observed in $YBa_2Cu_3O_{6.6}$ they must be relevant to CuO_2 materials [7]. Our analysis implies, on the contrary, that they are not.

We now turn to the data. χ_s may be determined from the measured bulk susceptibility $\chi_b=\chi_s+\chi_c+\chi_{VV}+\chi_{dia}$ if the core (χ_c), van Vleck (χ_{VV}), and Landau diamagnetic (χ_{dia}) contributions are known. In $YBa_2Cu_3O_7$, χ_{dia} is less than $\frac{1}{6}$ of χ_s [8]. We neglect χ_{dia} in our analysis. χ_s for the $YBa_2Cu_3O_{7-\delta}$ materials is known [9]. For $La_{2-x}Sr_xCuO_4$, however, there are conflicting assertions in the literature [10–13]. We estimate that in the La_2CuO_4 $(\chi_c+\chi_{VV})\cong 1.0$ states/(eV Cu) for fields parallel to the CuO_2 plane and -1.3 states/(eV Cu) for fields perpendicular to it by comparing the measured [10] susceptibility (at temperatures of order 600–800 K, well above the 3D Néel ordering temperature) to the known [14] values for the 2D $S=\frac{1}{2}$ quantum Heisenberg model [15]. Our estimates of $(\chi_c+\chi_{VV})$ for La_2CuO_4 agree with [10]. The example of the $YBa_2Cu_3O_{7-\delta}$ series [9] and the near equality [10] of the difference between the a-b plane and c axis susceptibilities of La_2CuO_4 and $La_{1.925}Sr_{0.075}CuO_4$ shows that it is reasonable to assume that χ_{VV} and χ_c do not change much with doping; we use this assumption to extract χ_s from χ_b.

Figure 1 shows $\chi_s(T)$ for some high-T_c materials, and for the $S=\frac{1}{2}$ 2D Heisenberg model with $J=0.13$ eV. It is clear that the spin susceptibility of $YBa_2Cu_3O_{6.6}$ is smaller in magnitude and larger in relative temperature dependence than that of all of the others and is the only one which extrapolates to zero at $T=0$. To obtain information about the dynamic susceptibility $\chi''(q,\omega)$ we consider nuclear relaxation rates $1/T_1T$. These are determined by weighted averages over q of the low frequency limit of $\chi''(q,\omega)$. Different nuclei have different weighting factors and probe different regions of a space. In all high-T_c superconductors the copper (Cu) and oxygen (O) relaxation rates have different T dependences [9]. The difference has been successfully explained by a model of T-dependent antiferromagnetic (AF) spin fluctuations which relax the Cu but not the O nuclei [16]. Figure 2 shows the copper nuclear relaxation rate $(1/T_1T)$ data [17,18]. The relaxation rates in $La_{2-x}Sr_xCuO_4$ are larger and more strongly temperature and doping dependent

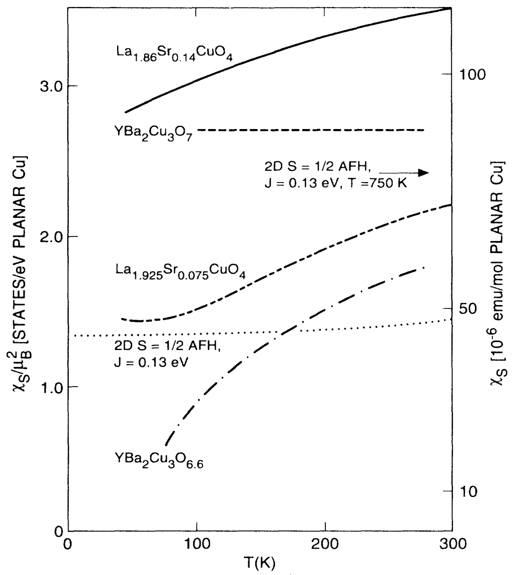

FIG. 1. Spin susceptibilities of high-T_c materials obtained from data as described in the text, along with the theoretical susceptibility of the 2D $S=\frac{1}{2}$ Heisenberg model [15].

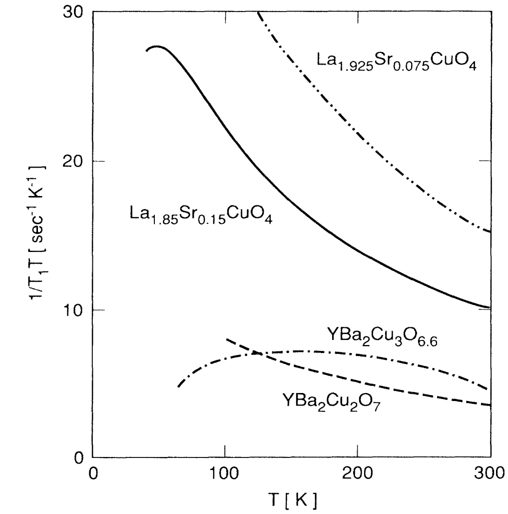

FIG. 2. Copper nuclear-quadrupole-resonance relaxation rates of high-T_c materials from Refs. [17,18]. Reference [18] used a normalization convention which differs from one used here by a factor of 3. We have reexpressed the data of Ref. [18] accordingly.

than in $YBa_2Cu_3O_{7-\delta}$. Further, in $La_{2-x}Sr_xCuO_4$ the Cu $1/T_1T$ increases as T decreases except in a small region below about $T=50$ K (which we suspect is dominated by superconducting fluctuations), in contrast to $YBa_2Cu_3O_{6.6}$ where the Cu $1/T_1T$ has a broad maximum at $T=150$ K. In $YBa_2Cu_3O_{6.6}$ the oxygen $1/T_1T$ has nearly the same T dependence as $\chi_s(T)$ [9].

The distinctive behaviors of $La_{2-x}Sr_xCuO_4$, namely, a decrease, but *not to zero*, of χ_s and a monotonic increase in the Cu $1/T_1T$ as $T\rightarrow 0$, are signatures of SDW antiferromagnetism [19]. To see this, note that mean-field theory predicts a transition at a temperature $T_{\rm MF}$ to a phase with nonzero staggered magnetization $\mathbf{N}$. In two spatial dimensions, thermal fluctuations prevent long-range order for $T>0$ [20]; for $T<T_{\rm MF}$ the appropriate picture is of slowly fluctuating domains, with $\mathbf{N}$ nonzero, but random from domain to domain. χ_s in this situation may be reasonably approximated by rotationally averaging the mean-field result, and rounding out the singularity at $T_{\rm MF}$. This leads to a susceptibility which drops by a factor of $\frac{2}{3}$ between $T_{\rm MF}$ and 0 [21]. These arguments imply that antiferromagnetic fluctuations at $T<T_{\rm MF}$ lead to a $\chi_s(T)$. However, a mode-coupling analysis of antiferromagnetic fluctuations at $T>T_{\rm MF}$ yields a negligible T dependence of χ_s [22]. Thus we propose that in the $La_{1.925}Sr_{0.075}CuO_4$ sample the $T_{\rm MF}$ is rather above room temperature while in the $La_{1.86}Sr_{0.14}CuO_4$ sample it is somewhat below. This may be consistent with neutron scattering experiments on $La_{1.86}Sr_{0.14}CuO_4$ [23] in which quite sharp peaks are observed emerging at low T. For still larger Sr concentrations, $T_{\rm MF}<T_c$ and χ_s in our model would not decrease with decreasing T, in agreement with data [10]. The $\chi_s(T)$ in $La_{2-x}Sr_xCuO_4$ was previously argued to be consistent with that of the 2D $S=\frac{1}{2}$ Heisenberg model with an x dependent $J\sim 300$–400 K at $x=0.14$ [1]. A difficulty with this interpretation is that the χ_s $1/T$ behavior of the Heisenberg model at $T>J$ is not observed [10]. We feel the SDW interpretation is more reasonable.

In our picture the Cu NMR relaxation is dominated by the relaxational dynamics of the AF fluctuations, so $1/T_1T\sim\sum_q \xi^{2-\eta+z} f(q\xi)/g(q\xi)\sim\xi^{z-\eta}$, where ξ is the correlation length, η and z are critical exponents, and f and g are scaling functions for the staggered susceptibility and spin fluctuation energy scale, respectively. Note this form is not multiplied by $\chi_s(T)$ in contrast to Ref. [13]. The "MMP" form proposed [16] for $YBa_2Cu_3O_7$ corresponds to $\eta=0$, $z=2$, and $f=g^{-1}=(1+q^2\xi^2)$. For $T\gtrsim T_{\rm MF}$, the MMP forms are appropriate; as $T\rightarrow 0$ the function must cross over to the 2D AF scaling forms, in which $z=1$. The oxygen relaxation rate, however, is due to small-q spin fluctuations [9]. In an ordered antiferromagnet relaxation is due to spin waves; at low T the number of thermally excited spin waves is small and the projection of these onto the small-q fluctuations relaxing the oxygen site vanishes as $q\rightarrow 0$, so that the oxygen $1/T_1T\sim T^3$ [24]. Similarly in a 2D SDW below $T_{\rm MF}$ the formation of antiferromagnetic domains will lead to an oxygen $1/T_1T$ which drops more rapidly than $\chi_s(T)$, in contrast to the prediction in [13]. If our analysis of $\chi_s(T)$ is accepted then this behavior has already been observed [11,25].

Now consider $YBa_2Cu_3O_{7-\delta}$. The basic structural unit is a pair of CuO_2 planes, separated from the next pair of CuO_2 planes by the relatively inert CuO chains [26]. Neutron scattering measurements on $YBa_2Cu_3O_{6.6}$ reveal that at least the $\mathbf{q}=(\pi,\pi)$ spin fluctuations on near neighbor CuO_2 planes are perfectly anticorrelated up to 40 meV and room temperature, implying the planes are magnetically coupled [27]. We argue that this coupling leads to the formation of singlet pairs with one member in each plane. We study this effect in a simple model of two coupled planes of antiferromagnetically correlated spins introduced to describe the insulating antiferromagnetic phase of $YBa_2Cu_3O_{6+x}$ [6]. The Hamiltonian is

$$H = J_1 \sum_{\langle i,j \rangle, a} \mathbf{S}_{i(a)} \cdot \mathbf{S}_{j(a)} + J_2 \sum_i \mathbf{S}_{i(1)} \cdot \mathbf{S}_{i(2)} . \qquad (1)$$

Here i and j label nearest neighbor sites in a two-dimensional square lattice and $a \in \{1,2\}$ labels the two different planes. This model has a $T=0$ phase transition between a large J_2 singlet state and a small J_2 antiferromagnetic state. Reference [6] gave a classical spin-wave analysis of some properties of the ordered phase. The spin-wave analysis cannot be used to study the disordered phase and the transition; we use instead the Schwinger boson method [28]. Applying this to Eq. (1) we find the second order transition of interest here is preempted by a first order transition. In the mean-field analysis, sums over the momentum q occur; these may be replaced by an integral over an energy times a density of states which, for the model of Eq. (1), is constant near the band edges and logarithmically divergent at band center. Replacing this density of states by a constant yields a model with a second order $T=0$ transition (of the 3D Heisenberg universality class) at $J_2^* = 4.48 J_1$.

We have computed $\chi_s(T)$ and the oxygen, yttrium, and copper NMR relaxation rates for various $J_2 > J_2^*$ using Eq. (1), the constant density of states, and a simplified version of the standard NMR form factors [16] in which the Cu transferred hyperfine coupling B was set to 0. Some results are shown in Fig. 3 for $J_2 - J_2^* = 0.3 J_1$; the resemblance of the curves for Cu and O to the data for $YBa_2Cu_3O_{6.6}$ is evident, and suggests that the physics of Eq. (1) is relevant to this material. Note in particular

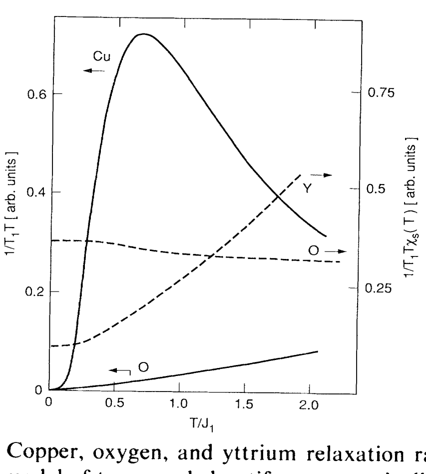

FIG. 3. Copper, oxygen, and yttrium relaxation rates calculated for a model of two coupled antiferromagnetically correlated planes using Schwinger boson mean-field analysis of Eq. (1) for $J_2 - J^* = 0.3$. The left ordinate shows the Cu and O relaxation rates $1/T_1T$ (solid lines); the right ordinate shows the ratio of the O and Y $1/T_1T$ to the calculated spin suceptibility χ_s.

the existence of two temperature scales; a higher one, of order J_2, at which χ_s and the oxygen $1/T_1T$ begin to drop, and a lower scale of order $T^* \sim (J_2 - J^*) < J_2$, at which the Cu $1/T_1T$ begins to drop. The different T dependences of the Cu and O $1/T_1T$ in our model are due to both a growing correlation length and different size spin gaps in different regions of q space, in contrast to a previous model [29] in which the difference was due only to a T-dependent correlation length. The difference between the O and Y relaxation rates arises in our model because the Y nucleus is relaxed only by fluctuations symmetric under interchange of the two planes; these are the most strongly suppressed by the tendency to form singlets. The experimental situation is not settled: Published Y relaxation data indicate Y and O relaxation rates have the same temperature dependence [30] but a very recent preprint reports that the Y rate falls faster than the O rate in $YBa_2Cu_3O_{6.6}$ [31].

We have computed the dynamical susceptibility $\chi''(q,\omega)$ at in-plane $\mathbf{q} = (\pi,\pi) = Q_{AF}$. For $T < T^*$ this shows an onset at $\omega \sim 2T^*$, and has no other low energy structure. Neutron scattering data show at low T an onset of scattering at 9 meV [27] or 16 meV [32] (i.e., at $\omega \sim T^*$) and additional structure at higher ω. We have also computed the dependence of $\chi''(q,\omega)$ on the momentum transfer between planes. We find that for in-plane $q = Q_{AF}$ the observed [27] "bilayer modulation" persists up to $\omega \sim \max[J_2, (J_1 J_2)^{1/2}]$ independent of the value or existence of the spin gap, so we believe the modulation is evidence that the planes are coupled but is not a consequence of the existence of the spin gap. We have also computed the static susceptibility at $q = Q_{AF}$, $\chi_{AF}(T)$. For all $J_2 > J_2^*$ we find χ_{AF} decreases with T for T less than the temperature at which $1/T_1T$ for Cu has its maximum. This is inconsistent with recent T_2 measurements on $YBa_2Cu_4O_8$ [33] (which has the same spin-gap behavior as $YBa_2Cu_3O_{6.6}$). These imply that $\int dq [\chi'(q)]^2 \sim \chi_{AF}$ increases monotonically and smoothly by a factor of 2 between 300 and 100 K, so that as T decreases spin fluctuation weight is not only pushed away from low frequencies but also pulled down from high frequencies. Equation (1) does not contain this physics.

A realistic theory must incorporate itinerant carriers, and as in $La_{2-x}Sr_xCuO_4$ a Fermi-surface-instability description of the magnetic dynamics is required. One possibility, a model of two planes of fermions, with a J_2 and with direct hopping from plane to plane forbidden, was shown in Ref. [4] to lead to a crossover below a temperature T_{pair} to a "superconducting" state in which every Cooper pair has one member in each plane. This is a mathematical representation of the between-planes singlet produced by the J_2 interaction of Eq. (1); it need not imply the presence of true superconducting order [4]. Because the itinerant carriers suppress the magnetism, very much smaller values of the coupling J_2 than were required in the spin-only model of Eq. (1) will produce appropriate interplane pairing. We have not yet fully incorporated antiferromagnetism in the formalism of Ref. [4], but have shown that the requirement $J_2 > J_1$ of the insulating model is replaced by $T_{\text{pair}} > T_{\text{SDW}}$, and that coherent three-dimensional transport acts as a pair breaker [22]. This provides a possible explanation of the difference between $YBa_2Cu_3O_{6.6}$ and $YBa_2Cu_3O_7$: In the latter material the much larger hole density permits three-dimensional transport which is strong enough to destroy the interplane pairing.

In this paper we have proposed models for the magnetic dynamics of underdoped cuprates. A crucial datum is the $T \to 0$ extrapolation of $\chi_s(T)$. We have argued that this is nonzero and indeed large in $La_{2-x}Sr_xCuO_4$. If it is small, then a one-plane quantum disordered phase must be considered for $La_{2-x}Sr_xCuO_4$, and the evidence for interplane pairing in $YBa_2Cu_3O_{6.6}$ is weakened. Two important consequences are (a) in $La_{2-x}Sr_xCuO_4$ samples with a T-dependent χ_s the oxygen $1/T_1T$ should decrease more rapidly than χ_s with decreasing T and (b) in $YBa_2Cu_3O_{6.6}$ the Y relaxation rate should drop more rapidly than the O relaxation rate as T decreases.

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