Properties that change as superconductivity disappears at high-doping concentrations in $La_{2-x}Sr_xCuO_4$

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In the system $La_{2-x}Sr_xCuO_4$, as x and p (the $[CuO_2]^{p-2}$ charge) are increased, the superconducting transition temperature first increases, then peaks becoming nonsuperconducting for $x \gtrsim 0.26$. We report here a search for changes in physical properties at the values of x where T_c is observed to change its behavior. The in-plane lattice constants and the normal resistivity both show a continued monotonic decrease over this entire region, suggesting that no major electronic changes occur. The tetragonal-to-orthorhombic transition temperature T_s also decreases with increasing x and becomes unobservable for $x \gtrsim 0.19$, suggesting that this structural transition itself is unrelated to the disappearance of superconductivity that occurs at higher doping levels. The magnetic spin susceptibility $\chi_{spin}(T)$ generally rises gradually with increasing doping (reflecting decreasing spin-spin interactions), reaches a maximum near $x \sim 0.25$, and then decreases. There is a weak peak in $\chi_{spin}(T)$ as a function of temperature at $T = T_{max}$. As a function of increasing x, T_{max} falls to zero near $x \sim 0.25$. These two observations might be related to the disappearance of superconductivity, since all three occur near the same value of Sr content x.

The system $La_{2-x}Sr_xCuO_4$ is potentially ideal for studying high-temperature superconductivity[1] due to its simple structure and wide range of solid solution,[2] in which the K_2NiF_4 structure is maintained over the range $0 \le x \le 1.33$. Corresponding to this large range of doping, there is potentially a wide range of values of the charge[3] p on the $[CuO_2]^{p-2}$ sheets, over which high-temperature superconductivity can be studied. In practice, however, as the Sr content is increased, oxygen vacancies start to appear and severely limit[4] this range to $p < 0.15$, depending on the preparation conditions. Preparing samples under a high pressure (100 bars) of oxygen has recently been shown[5] to extend the range of accessible charge up to $p = 0.4$. From now on, we shall restrict our discussion to samples containing no oxygen vacancies and (for reasons discussed later) we shall characterize them by their Sr content x. As a function of x the behavior of the observed superconducting transition temperature T_c is shown in Fig. 1(a). At low x, superconductivity was found[4,6] to appear for $x > 0.06$, with T_c rising with increasing x until maximum values were reached for both T_c (36 K) and x (0.15) on those samples. Measurements on samples with higher x more clearly reveal that T_c levels off at $T_c = 36$ K until $x = 0.19$, beyond which T_c surprisingly decreases, until about $x = 0.26$, above which superconductivity is no longer observed.[5] This behavior has recently been confirmed by Takagi *et al.*[7] Most of the work[6] on this system has focused on the region of low x, where superconductivity first starts to appear. This region is complicated by the competition with the antiferromagnetic state[8] near $x = 0$, by spin-glass effects,[9] by the presence of another phase,[10] and by the conductivity which shows evidence of partial localization.[6] In this paper, we concentrate on the region of large x, where the conductivity is metallic and yet superconductivity anomalously disappears.[5] We present measurements of the in-

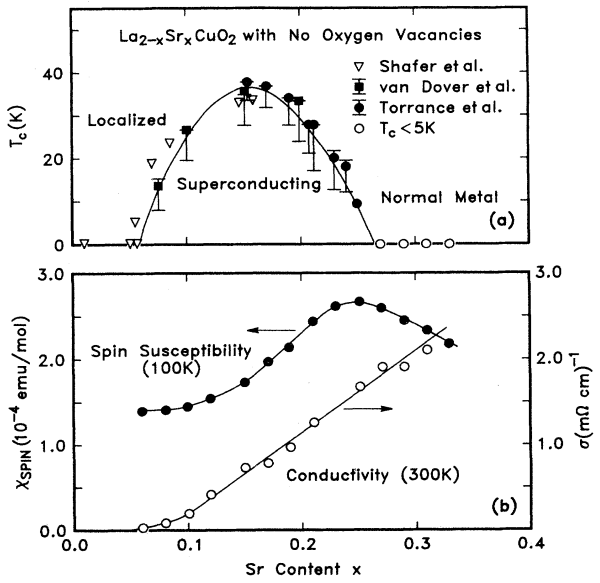

FIG. 1. (a) T_c vs x for $La_{2-x}Sr_xCuO_4$ [data from Refs. 4 and 5, and van Dover *et al.*, Phys. Rev. B **35**, 5337 (1987)]; (b) the x dependence of the conductivity at 300 K and spin susceptibility at 100 K.

plane lattice constants, orthorhombic distortion, conductivity, and magnetic susceptibility over the range of x where superconductivity appears, levels off, and then disappears, in hope of finding a correlation between these properties.

For the original $La_{2-x}Sr_xCuO_{4-\delta}$ series of samples,[5] the in-plane lattice constants at 300 K are shown in Fig. 2. At low Sr content, the structure is orthorhombic, with axes $a' \neq b'$. As the Sr content increases, this orthorhombic distortion at 300 K decreases and becomes unobservable above $x = 0.07$. In the tetragonal phase, the lattice constant continues to decrease, until near $x = 0.28$, where the lattice constant of samples prepared in one bar of O_2 suddenly increases. From measurements of the charge p on these samples, this abrupt change has been shown[5] to be caused by the sudden appearance of oxygen vacancies at this value of x. Samples prepared under a pressure of 100 bars of O_2 do not show this sudden increase and exhibit a continuing decrease of a until $x = 0.4$, as seen in Fig. 2. This decrease suggests that charge is being continuously removed from antibonding Cu-O orbitals. Also shown in Fig. 2 is a data point[11] for the $x = 1$ compound $LaSrCuO_4$, prepared under extremely high oxygen pressures (3 kbars). In the remainder of this paper, we shall only discuss physical measurements on samples without oxygen vacancies, as determined by measuring either p or the lattice constants, or both (as in our case).

Undoped La_2CuO_4 is orthorhombic at low temperatures, but has a transition at $T_s = 530$ K to a tetragonal phase. As the system is doped with Sr, T_s decreases, and somewhere near $x \sim 0.2$ it goes to zero.[12,13] It has been suggested that the orthorhombicity observed at low temperatures may be essential to the onset of high-temperature superconductivity,[14] and thus superconductivity will disappear at high x, where the samples are tetragonal at low temperatures. In order to test this conjecture, we carried out high-resolution x-ray scattering measurements on the series of $La_{2-x}Sr_xCuO_4$ samples studied in Ref. 5. For four of these samples ($x = 0.16$, 0.19, 0.21, and 0.23), we studied the (110) and (103) tetragonal phase diffraction peaks as a function of temperature for $15 < T < 300$ K. The tetragonal-to-orthorhombic transition is characterized by a splitting of the (110) peak, while the (103) peak remains unchanged. For the $x = 0.16$ sample, we find $T_s \approx 170$ K (as shown in Fig. 3). For the $x = 0.19$ sample we find no evidence for a tetragonal-to-orthorhombic transition, although we do observe a 35% increase in the width of the (110) peak between 170 and 60 K, while the width of the (103) peak remains essentially constant. This observation suggests that although the sample remains tetragonal, significant orthorhombic strains are present at lower temperatures ($T < 100$ K). Finally, for $x = 0.21$ and 0.23 samples we find no evidence for either a structural transition or orthorhombic strain above 15 K—this despite the fact that they become superconducting at 27.5 and 20 K, respectively. As shown schematically in Fig. 4, T_s falls to zero temperature for $x \sim 0.18$, whereas T_c disappears near $x \sim 0.26$, as shown in Fig. 1. Thus, the long-range orthorhombic or tetragonal nature of the samples appears unrelated to their superconducting behavior.

The electrical resistivity[15] ρ of the original samples from Ref. 5 is plotted versus temperature in Fig. 5. The samples are all metallic with approximately the same resistance ratios, $\rho(300\text{ K})/\rho(50\text{ K}) \sim 4$. The temperature dependence of $\rho(T)$ is roughly linear, although there are substantial deviations from linearity evident for large x. The magnitude at the room temperature conductivity is plotted in Fig. 1(b). Its behavior is very simple: The conductivity increases monotonically with increasing Sr content x, indicating that increasing doping continuously improves the conductivity. This monotonic behavior along with that found for the a-lattice constant (Fig. 2) give no indication of anything complex going on as a function of increasing x, such as the conductivity becoming dominated by holes from another band, for example.

The behavior of the magnetic spin susceptibility[16] χ as a function of temperature is shown in Figs. 6(a) and 6(b). In Fig. 1(b), we show the x dependence of χ_{spin} (100 K), i.e., the susceptibility at $T = 100$ K adjusted for effects of

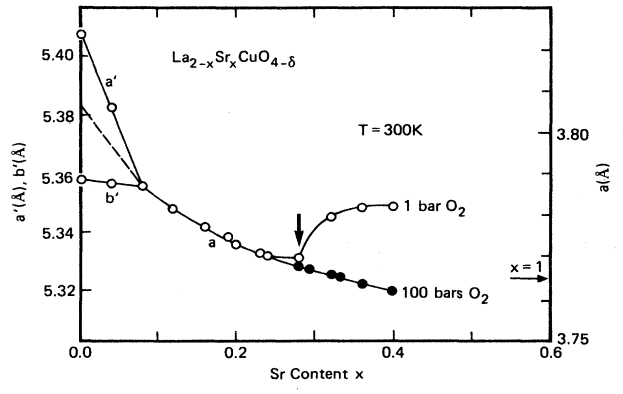

FIG. 2. In-plane lattice constants of $La_{2-x}Sr_xCuO_{4-\delta}$ at 300 K, in the tetragonal (a) and orthorhombic (a',b') phases. Note that the samples prepared under 1 bar of oxygen show an onset of oxygen (○) vacancies near $x \sim 0.28$, while those prepared under 100 bars (●) show no indications of vacancies up to $x \sim 0.4$.

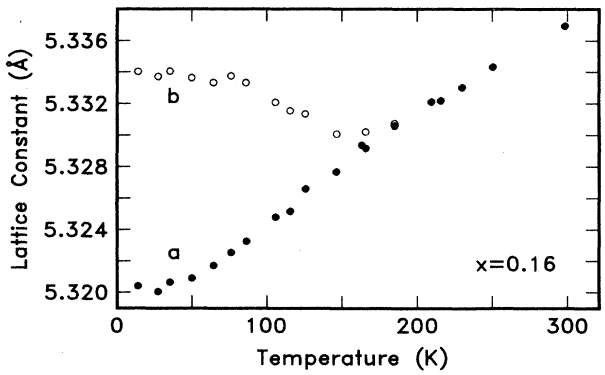

FIG. 3. The lattice constants a and b for a sample with $x = 0.16$, showing the tetragonal-to-orthorhombic transition for $T_s \sim 170$ K.

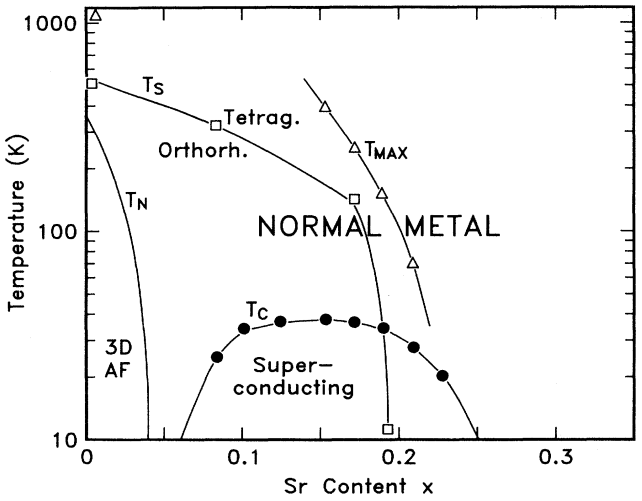

FIG. 4. Schematic figure of the variation with x of the 3D Néel temperature (T_N), the superconducting transition temperature (T_c), the temperature (T_s) of the orthorhombic-to-tetragonal transition, and the temperature (T_{max}) at which $\chi(T)$ shows a maximum as a function of temperature.

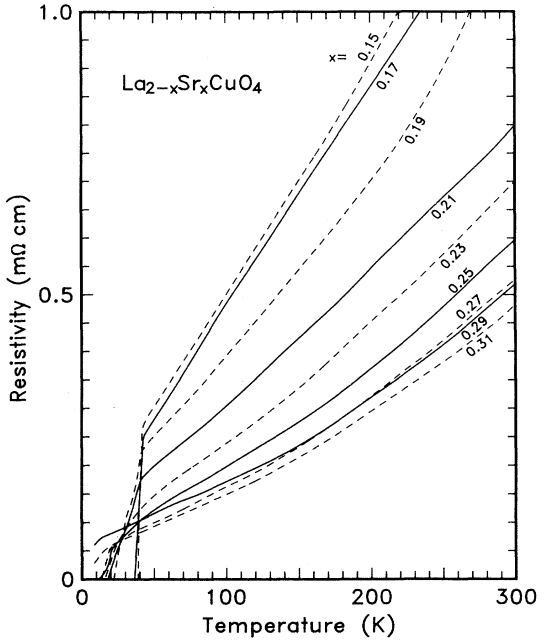

FIG. 5. The resistivity vs temperature, showing metallic behavior and decreasing resistance for increasing x.

the diamagnetic contribution (1×10^{-4} emu/mol) of the ionic cores. There are important differences between these data and those reported earlier by Schneemeyer *et al.*,[17] where the oxygen vacancy content was not determined. Our data and those of Takagi *et al.*[7] are in excellent agreement. For the lowest x ($x<0.02$, not shown in Fig. 6), muon spin rotation and neutron scattering experiments have shown the existence of a three-dimensional antiferromagnetic (AF) ordering of the Cu^{2+} moments with strong two-dimensional correlations above the Néel temperature T_N.[18–20] As shown schematically in Fig. 4, doping with holes destroys the Néel state and the large, dominant anomaly in $\chi(T)$ associated with it, but short-range two-dimensional AF correlations are retained in the Cu-O planes.[21–23] For $x>0.02$, $\chi(T)$ is dominated by these 2D correlations, and it is their evolution with increasing Sr content x that we want to discuss.

The main characteristic of such a two-dimensional AF spin system with a strong exchange interaction is expected[24,25] to be the absence of long-range order and the presence of a relatively weakly temperature-dependent $\chi(T)$. Furthermore, the value of the magnetic moment is strongly reduced from its Néel value because of zero-point deviations. It reaches 60% of $S=\frac{1}{2}$ in the case of a quadratic Heisenberg antiferromagnet, according to recent Monte Carlo calculations or spin-wave theory.[24,25] In the region $0.02<x<0.14$, $\chi(T)$ is only weakly temperature dependent, consistent with the behavior of exchanged coupled Cu^{2+} spins with a large in-plane AF exchange interaction J. The most reliable discussion of these data centers on relating the magnitude of χ_{spin} to this 2D exchange coupling J, i.e., χ_{spin} is expected to be inversely proportional to J in the region where $T<J$. To be more quantitative,[24,25] for a 2D system near T_{max} the

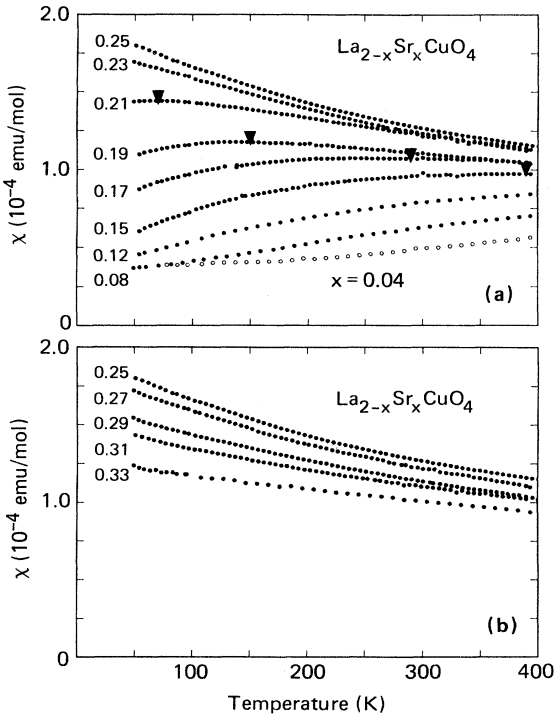

FIG. 6. Normal-state magnetic susceptibility of $La_{2-x}Sr_xCuO_4$ compounds which, for increasing x, (a) increases for $0.04<x<0.25$, and (b) decreases for $0.25<x<0.33$. The filled triangles mark the temperatures where χ is a maximum.

magnitude of $\chi_{spin}=0.047Ng^2\mu_B^2/J=(0.14\ \text{emu/mol})/J$. From our data at 400 K (Fig. 6), $\chi_{spin}\sim 1.6-2.2\times10^{-4}$ emu/mol, which corresponds to $J\sim650-900$ K, somewhat reduced from the value of 1000 K estimated for the undoped compound from light scattering and inelastic neutron diffraction experiments.[8,26] Thus, in general the increasing magnitude of χ_{spin} [Fig. 1(b)] is interpreted as reflecting a general weakening of the exchange interactions as x increases. The data in Fig. 1(b) correspond to a weakening of J by approximately 40%, although data at higher temperatures would indicate a smaller reduction. The decrease in χ_{spin} (100 K) above $x\sim0.26$ might correspond to a renewed strengthening of the interaction.

While we believe that the magnitude of χ is the most reliable quantity to discuss, nevertheless, it is interesting to examine the behavior of T_{max}, the temperature of the maximum in $\chi(T)$, which in the simplest model is related to J by $kT_{max}=0.95J$, as calculated from the high-temperature series expansion for $S=\frac{1}{2}$.[24] For the lowest doping levels, data taken at high temperatures[27] suggest a maximum above 800 K. Upon doping, this maximum appears[27] to broaden and shift to lower temperatures, so that for $x>0.14$, the broad maximum in $\chi(T)$ at T_{max} can be seen in the $0<T<400$ K range of our experiments. The values of T_{max}, represented by the solid triangles in Fig. 6, are tabulated in Table I and plotted in Fig. 4. It is interesting to note that T_{max} falls to zero near the same $x\sim0.26$ where superconductivity disappears, although it is not clear how much significance we should assign to this observation. For interpretation, there are two points of view: The first is that we should not regard T_{max} in these data as a reliable measure of J, since we would be forced to conclude that the exchange interaction had vanished near $x\sim0.26$, and that would not be reasonable for the following reasons: if J had vanished, the magnitude of $\chi\sim1/J$ would have increased much more dramatically, and a significant Curie contribution would have been observed for $x\geq0.25$ (assuming there are still localized Cu^{2+} spins remaining). Furthermore, in undoped samples, the exchange is large (~1000 K) because of the short Cu-O interactions, which strengthen upon doping (Fig. 2). According to the second point of view, it could be argued that as the system becomes more delocalized, the localized Cu^{+2} spins continue to diminish, eventually resulting in an effective vanishing of both these moments and J. This point of view has been recently pursued by Johnston *et al.*[28] in their analysis of their $\chi(T)$ data. It is not clear how to interpret the detailed temperature and x dependence of $\chi(T)$ in Figs. 6(a) and 6(b). Nevertheless, from the localized point of view, we would speculate that as x is increased and J is weakened, that the weakening is not uniform and there is a distribution of J's, some of which are relatively low. The increasing number of these low J's may be responsible for the increasing "Curie-Weiss" component to $\chi(T)$ as the hole concentration is increased.

Our data for T_c as a function of x [Fig. 1(a)] correspond well with the recent data of Takagi *et al.*,[7] and our measurement of T_s at $x=0.16$ agrees with the data of Moret *et al.*[13] However, our samples have higher values of p, and when the above data are compared as a function of p, there is poor agreement. This observation suggests that the extra oxygen (~0.04) present in our samples,[5] which gives the extra contribution to p, does not strongly affect the properties, perhaps because the extra oxygen is incorporated in the form of peroxide[29,30] (rather than oxide). This is consistent since peroxide would contribute to the titration measurement, but would not dope the CuO_2 sheets and hence not contribute to the $[CuO_2]^{p-2}$ charge. For this reason, we believe that the hole concentration in the sheets is better represented by x than by p.

Another issue involved in this system is sample inhomogeneity. This inhomogeneity could take on at least two forms: (i) random fluctuations in Sr content; or (ii) phase separation.[31] In the first type, the randomness in the Sr concentration would normally be averaged out by the delocalized electrons, but the superconducting pairs in these materials have such an extremely short coherence length (~10 Å) that they do not average out these fluctuations. There are two examples of the second type of inhomogeneity: Samples of $La_2CuO_{4+\delta}$ have been shown[10] to contain a two-phase mixture of an insulating phase (with $\delta\sim0$) and a superconductivity phase (with $\delta\sim0.14$). Such two-phase mixtures can be identified by their characteristic behavior observed in Meissner mea-

TABLE I. Sr doping (x), hole concentration (p), symmetry of structure (ss) at 15 K, superconducting transition temperature (T_c), Meissner fraction (%), conductivity (σ) in $(\text{m}\Omega\,\text{cm})^{-1}$, temperature ($T_{max}$), where normal-state magnetic susceptibility (χ) in (10^{-4} emu/mol) is a maximum, and the values of χ at T_{max} and 100 K, for the $La_{2-x}Sr_xCuO_4$ series.

x	p	ss	T_c	%	σ (300 K)	T_{max}	χ (T_{max})	χ (100 K)
0.15	0.208	*O*	37.5	30	0.73	390	0.97	0.72
0.17	0.226	*O*	36.5	30	0.78	260	1.06	0.96
0.19	0.244	*T*	34	30	0.95	150	1.17	1.14
0.21	0.263	*T*	27.5	30	1.25	70	1.43	1.42
0.23	0.281	*T*	20	20	1.43	20	1.74	1.60
0.25	0.299		<5	0	1.67	0	1.95	1.66
0.27	0.318		<5	0	1.90	0	0	1.59
0.29	0.336		<5	0	1.90	0	0	1.44
0.31	0.354		<5	0	2.11	0	0	1.33
0.33	0.373		<5	0		0	0	1.16

surements: a relatively constant $T_c \sim 30$ K and a strongly varying Meissner fraction (from 10^{-3} to $\sim$ 40%). A second example is given by the large x samples of van Dover *et al.*[32] These samples show a sharp drop in resistivity at about the same temperature ($\sim$37 K) for $x = 0.15$, 0.20, 0.30, and 0.50, while the Meissner fraction decreases by more than an order of magnitude. In this case, the inhomogeneity is undoubtedly due to oxygen vacancies in their samples (since at this early stage the importance of these vacancies was not known). Such a drop in resistivity is not observed in our samples in this range. In both of these examples of phase inhomogeneity, the oxygen content is the physical quantity which has the potential to be inhomogeneous, i.e., different phases can have different oxygen contents. In the first example, it is the extra, interstitial oxygen, while in the second it is the oxygen vacancies. In the samples of Takagi *et al.*[7] (which give essentially identical results to ours), the oxygen content is equal to 4, and phase separation due to oxygen content is not possible. Furthermore, the Meissner data[5,7] in our and their samples are not characteristic of phase inhomogeneity; there is a clear change in T_c, by more than a factor of 2. Variations in the Meissner fraction can be less reliable due to complications with flux pinning and field dependence. Further evidence for the phase homogeneity of this system is provided by the tetragonal-to-orthorhombic phase transition, which has a narrow temperature width and is a continuous function of x over much of this range.[13]

In summary, annealing a series of $La_{2-x}Sr_xCuO_{4-\delta}$ samples under high oxygen pressure allows us to obtain samples without oxygen vacancies up to $x = 0.36$. This increase in the Sr content does not give rise to further increases in T_c, as had been inferred by some theoretical predictions. On the contrary, T_c peaks and superconductivity surprisingly disappears for $x \sim 0.26$, whereas the normal-state conductivity continues to increase. In the $YBa_2Cu_3O_y$-like systems, a strong dependence of T_c on charge p in the $[CuO_2]^{p-2}$ sheets is also observed,[33] with a maximum T_c similarly near $p \sim 0.2$, and with some recent evidence[34] that T_c decreases for $p > 0.3$. Recent data[35] on some bismuth-copper-oxide compounds also indicate that T_c falls with increasing oxygen content (increasing p). In the $La_{2-x}Sr_xCuO_4$ system, we have measured a variety of properties over the wide range $0 < x \leq 0.35$. We should recognize that this range is part of the broader $0 < x \leq 1$ range. For low Sr content the carriers will be holes, whereas for large x they will be electrons. As x is increased over the broader range, we should expect a maximum in the conductivity, a generally decreasing χ_{spin}, which vanishes for $x = 1$, and a change in the sign of the Hall effect. For increasing x, the conductivity and in-plane lattice constant appear to behave monotonically and smoothly, giving no indication of dramatic changes in the electronic or charge states. Our x-ray data indicate that the tetragonal-to-orthorhombic phase transition is observed only for $x \geq 0.19$, whereas superconductivity is observed until $x \sim 0.26$, as shown in Fig. 4. For the $x = 0.21$ and 0.23 samples, we find clear evidence for a tetragonal superconductor, further supporting the view that these two transitions are unrelated. As a function of increasing hole concentration, the magnetic susceptibility generally increases, reflecting a weakening of the in-plane exchange interaction induced by the holes. Although $\chi(T)$ shows no dramatic changes in behavior over the region of x studied, it does exhibit two distinct changes which might be correlated with the disappearance of superconductivity: First, there is a peak in the magnitude of χ_{spin} ($T = 100$ K) as a function of x near $x \sim 0.25$ [Fig. 1(b)] and second, the temperature of T_{max}, where $\chi(T)$ is maximum, falls to zero near $x \sim 0.25$ (Fig. 3). The fact that both of these effects occur near the value of x at which superconductivity disappears suggests some correlation between high-temperature superconductivity and magnetism. Whichever theory will account for the dramatic dependence [Fig. 1(a)] of T_c on x in $La_{2-x}Sr_xCuO_4$ must also account for the behavior of the other properties reported here.

Note added in proof. We have recently become aware of interesting results on the pressure dependence[36] of T_c, the magnetic susceptibility,[37] changes[38] in the coherence length with x, and a comparison with other systems.[39]

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