Superconductor-to-nonsuperconductor transition in $(La_{1-x}Sr_x)_2CuO_4$ as investigated by transport and magnetic measurements

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Transport and magnetic properties of $(La_{1-x}Sr_x)_2CuO_4$ are systematically investigated over a wide composition range up to $x=0.175$ including a nonsuperconducting metal phase in the heavily doped region. Remarkable changes associated with the superconductor-to-nonsuperconductor transition are observed both in the Hall coefficient and the magnetic susceptibility, suggesting the modification of both charge and spin states.

I. INTRODUCTION

While the mechanism of the high-T_c superconductivity in Cu oxides is not understood yet, much effort has been made for specifying the characteristics which are essential for the occurrence of high-T_c superconductivity. Among the high-T_c cuprates, the $(La_{1-x}Sr_x)_2CuO_4$ system is the simplest and can be a prototype of these materials. La_2CuO_4, for which the average valence of the (Cu-O) complex is just zero, is regarded as the parent compound of the system. The intriguing feature of the parent compound is that it is an antiferromagnetic insulator as a consequence of the strong on-site Coulomb interaction on Cu site.[1] The high-T_c superconductivity appears in the vicinity of this antiferromagnetism. The increase of the average valence of the (Cu-O) complex with doping leads to the transition from insulator to superconductor. In this system, T_c increases monotonously up to $x=0.075$. Above this composition, however, the physical properties reported so far have been quite ambiguous. This has been ascribed to the presence of oxygen vacancies which will be introduced more easily in the high-Sr concentration range. Recently, Torrance *et al.* have carefully synthesized a series of specimens without oxygen vacancies and have established the relationship between the average valence of (Cu-O) complex and T_c.[2] It was pointed out in the earlier stage of the study that the higher-average valence led to the higher T_c. They have, however, unambiguously demonstrated a saturation of T_c against the average valence and the disappearance of the superconductivity at a higher-valence state, posing a new test on the proposed mechanisms.

In order to elucidate the novel electronic states of the cuprate superconductors, the detailed cooperative study on both the charge and spin states in the superconductor and nonsuperconductor would be useful. At the present stage, the $(La_{1-x}Sr_x)_2CuO_4$ system is the only system for which we can trace the change in the electronic states over a wide range of the average valence of the (Cu-O) complex which covers from the antiferromagnetic state to the nonsuperconducting-metal phase through the superconducting phase. Furthermore, its simple K_2NiF_4-type structure consisting of a single CuO_2 layer and the precisely controllable average valence should allow much simpler interpretation for the physics behind them than the other Cu oxide systems. In these contexts, we have investigated the effect of the Sr doping on the transport and magnetic properties of $(La_{1-x}Sr_x)_2CuO_4$ in detail, with emphasis on the characteristic change in the physical properties associated with the composition induced transition from superconductor-to-nonsuperconducting metal.

II. SAMPLE PREPARATION AND EXPERIMENTAL PROCEDURE

In order to investigate the details of the doping effect, it is essentially important to prepare the samples with good homogeneity and precisely controlled stoichiometry. We have employed the spray-dry technique in order to satisfy the above requirement. Furthermore, particular attention was paid for incorporating oxygen in the heavily doped region. Here, we describe the details of the sample preparation procedure.

The acetic solution of La^{3+}, Sr^{2+}, and Cu^{2+} with prescribed cation ratio was prepared. The stoichiometry was strictly controlled by means of the titration. The solution was sprayed through a nozzle and dried by hot air. Thus, we can obtain fine-powder mixture of the acetates of composite cations. The advantage of this technique versus the conventional powder mixing is to produce very homogeneous starting materials, since the cations have distributed as ions in the solution.

The obtained starting materials were first heat treated at 400 °C in air for 5 h in order to remove water. The products were ground and calcined at 900 °C for 5 h. After regrinding, the powders were pressed into pellets and sintered at 1050 °C for 50 h under O_2 gas flow, followed by cooling at the rate of 60 °C/h down to room temperature. Finally, the pellets were annealed at 400–500 °C for 50 h under O_2 gas flow. Since the oxygen

deficiency can be easily introduced in the heavily doped region, the annealing under oxygen high pressure of 80 bars at 500 °C was done for the samples $x > 0.10$. Judging from the x-ray-diffraction pattern and the magnitude of resistivity, very little difference was seen for $x < 0.175$ between the samples annealed under oxygen high pressure and those annealed in O_2 gas flow for a prolonged time. Therefore, in this paper, we will report the measurements on the samples annealed under O_2 gas flow.

These series of samples were confirmed to be single phase K_2NiF_4-type structures by the powder x-ray diffraction. The lattice constants at room temperature were plotted against the Sr content in Fig. 1, which agrees well with the previous reports. The observed systematic change of the lattice parameters upon doping guarantees the well-controlled stoichiometry. Any trace of line broadening was not observed over the whole composition range. The doping-induced orthorhombic-to-tetragonal transition was located at around $x = 0.05$.

The oxygen content was determined by the iodometric titration technique described by Nazzal *et al.*[3] Oxygen content is kept at almost four within our resolution, at least up to $x = 0.175$. Because of the difficulty in controlling the stoichiometry, in particular the oxygen content, it has been pointed out that the chemically determined valence of the (Cu-O) complex describes the system as a better variable than the Sr content. In this paper, however, we employ the nominal Sr content as a variable, since we did not detect any substantial difference between the chemically determined valence and that expected from the nominal Sr content. We have measured the electrical resistivity, the Hall coefficient, and the magnetic susceptibility on 30 samples in the range $0 \leq x \leq 0.175$. In order to check the effect of the anisotropy, we have prepared the single crystals in the heavily doped region from the CuO-rich melt. The Sr content of the obtained single crystals was determined as $x = 0.17$ by the inductively coupled plasma (ICP) technique.

For the resistivity and the Hall coefficient measurements, electrodes were formed by applying the heat-treatment-type gold paste. The Hall coefficient was measured under the magnetic field of $H = 50$ kOe. The magnetization measurement was carried out by using a SQUID magnetometer.

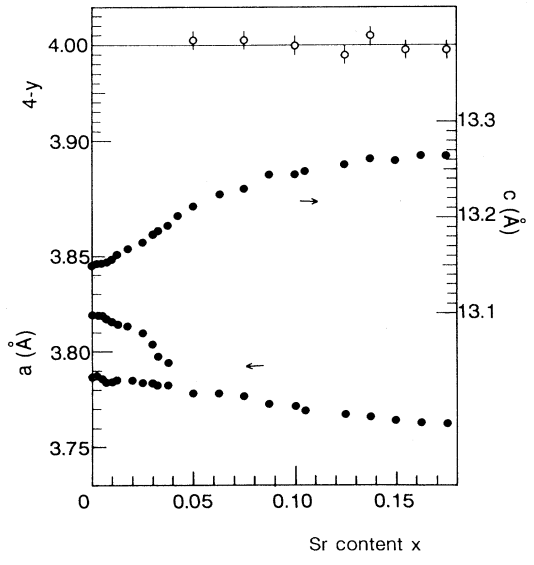

FIG. 1. Sr content x dependence of the lattice parameters a and c and oxygen content $4-y$ determined by the iodometric titration. For the orthorhombic samples below $x = 0.05$, normalized lattice parameters $a/\sqrt{2}$, $b/\sqrt{2}$, and c are shown.

III. RESULTS AND DISCUSSIONS

A. Electrical resistivity

First, we show the resistivity data in order to see the gross feature of the successive phase change of the system. Reflecting the well-controlled stoichiometry, the resistivity changes very systematically with doping, as shown in Fig. 2. As is well known, La_2CuO_4 shows semiconducting behavior over the measured temperature range, except for a trace of very minor superconductivity at low temperatures. With only 1% or less Sr doping, the resistivity drastically decreases to the order of 10^{-3} Ω cm and T-linear resistivity dominates at high temperature, indicating mobile carriers are supplied by a very small amount of doping. This feature is quite distinct from isostructural La_2NiO_4 or La_2CoO_4.[4] In these materials Sr seems to form localized electronic states within an insulating gap.

At the low-doping level below $x = 0.030$, the supplied carriers tend to be localized at low temperatures, which is evidenced by the variable-range-hopping-type resistivity behavior. As seen from the systematic decrease of the onset temperature of the low-temperature resistivity increase, the localization is suppressed by doping, which can be ascribed to the screening of the random potential by doped carriers. At around $x = 0.03$, the superconductivity sets in. T_c increases rapidly with doping, having a maximum $T_c = 39$ K at around $x = 0.075$.

An intriguing event occurs above $x = 0.075$.[2] In contrast to the case for $x < 0.075$, T_c decreases rapidly with dopant concentration, although the magnitude of the resistivity is still decreasing. Above $x = 0.130$, we cannot see superconducting transition any more down to 1.8 K. The system becomes a nonsuperconducting metal. No discontinuous change associated with the disappearance of the superconductivity is seen in the temperature dependence of the resistivity. However, with further increase in doping above $x = 0.150$, the resistivity increases and the upturn of the resistivity at low temperatures becomes significant. This might be ascribed to the scattering due to the disorder produced by a very small amount of oxygen vacancy which we cannot detect within our resolution.

The transition temperature T_c determined from the Meissner measurement is summarized in Fig. 3 as a function of the Sr content. The superconductivity is seen in the composition range from $x = 0.030$ to 0.125. Since the magnitude of the Meissner signal is remarkably small at

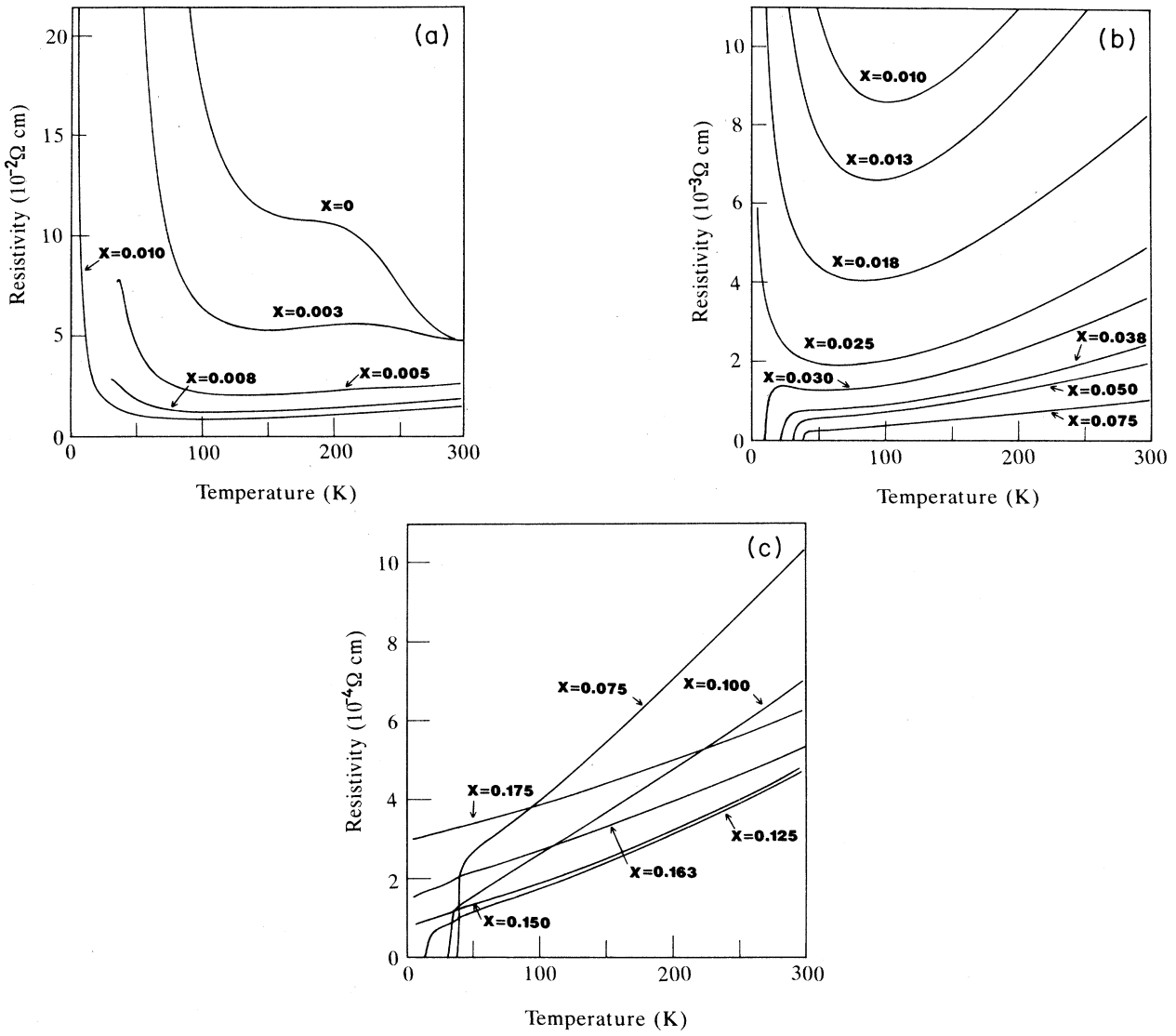

FIG. 2. Temperature dependence of the resistivity for $(La_{1-x}Sr_x)_2CuO_4$ with various Sr content.

around $x=0.03$ and 0.125 as shown in Fig. 4, the presence of compositional inhomogeneity seems to make the boundary obscured. Therefore, the real superconducting phase is possibly confined to narrower composition range than that experimentally observed. Recently, Moodenbaugh *et al.* have found an anomaly in the composition dependence of T_c in $(La_{1-x}Ba_x)_2CuO_4$. T_c shows a singular decrease at around $x=0.06$.[5] In the present Sr-doped system, however, we do not see such pronounced anomaly at around $x=0.06$, although a small trace of the degradation of the superconductivity is observed both in the resistivity and the Meissner signal. Therefore, the anomaly seems to be more or less specific to the Ba-doped system, supposedly originating from the larger difference of ionic radius between La^{3+} and Ba^{2+}. Hereafter, we denote the composition ranges $0 \le x < 0.010$, $0.010 \lesssim x < 0.030$, $0.030 \lesssim x < 0.125$, and $x \gtrsim 0.125$ as the antiferromagnetic phase, the intermediate region, the super-

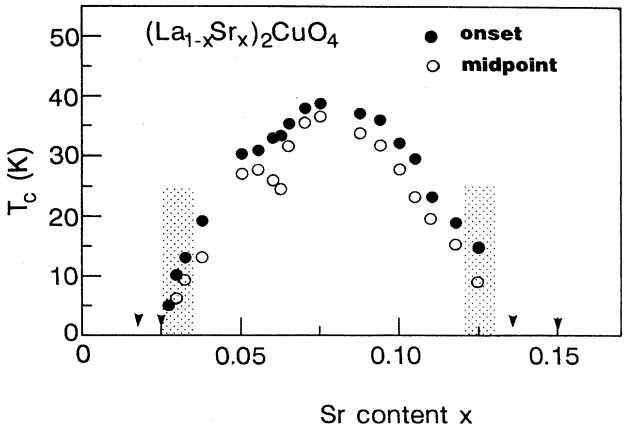

FIG. 3. Sr content x dependence of the superconducting transition temperature T_c. T_c was determined by the Meissner measurements shown in Fig. 4. The midpoint temperature were defined as half of the low-temperature saturated value.

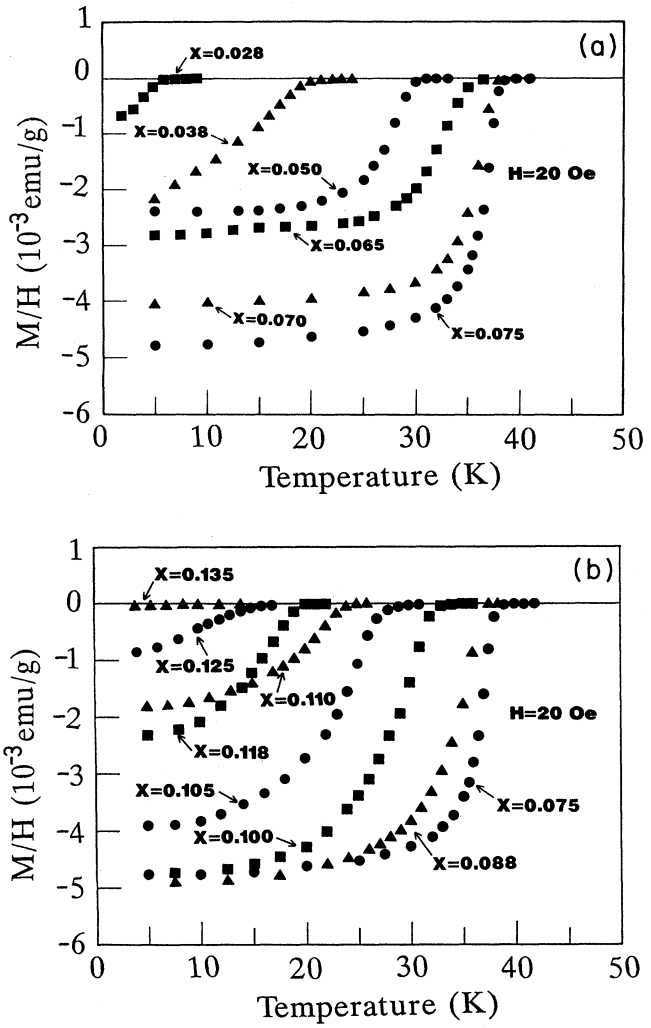

FIG. 4. Temperature dependence of the Meissner signal for $(La_{1-x}Sr_x)_2CuO_4$ with various Sr content. The measurement was performed by cooling the sample under constant field of $H=20$ Oe.

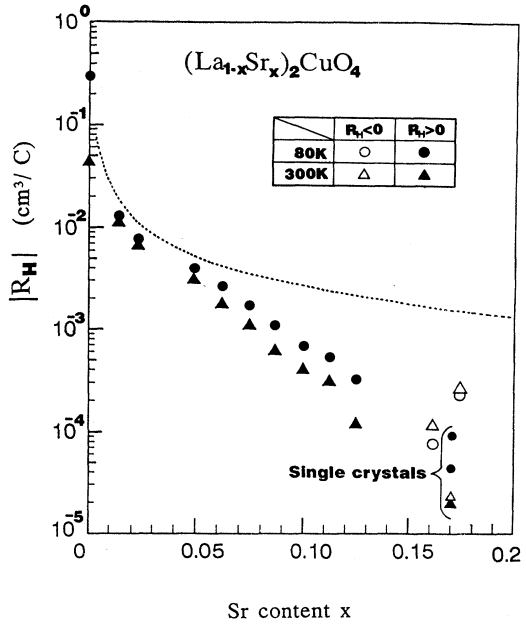

FIG. 5. Sr content x dependence of the Hall coefficient R_H at 80 K (circles) and 300 K (triangles). The sign of R_H is positive for $x<0.15$ and negative for $x>0.15$, respectively. The data for single crystals with $x=0.17$ are also plotted.

conducting phase, and the nonsuperconducting-metal phase, respectively.

B. Hall effect

We have measured the Hall coefficient in order to see how the electronic states of La_2CuO_4 are modified with doping. In Fig. 5, the Hall coefficients at 80 and 300 K are shown against the Sr content. As already observed in the earlier stage of the study,[6,7] the positive Hall coefficient decreases almost in proportion to the reciprocal Sr content $1/x$ in the low-composition range. This behavior has been explained on the basis of the Mott-Hubbard picture in which a Sr doping provides one mobile hole in the lower-filled band. With further doping $x>0.050$, the Hall coefficient decreases more rapidly than expected from the above picture. In the nonsuperconducting-metal phase, the Hall coefficient becomes as small as of the order of 10^{-4} cm^3/C or less and changes its sign from positive to negative. This rapid decrease was first reported by Ong *et al.*[7] In their report, however, the deviation from $1/x$ was significant at around $x=0.1$ and the decrease is much steeper than the present result, which might be ascribed to the oxygen vacancies.

It has been argued whether the $1/x$ dependence of the Hall coefficient can be understood within the framework of the Fermi-liquid theory. Although attempts have been made, any theory based on the Fermi-liquid description has not succeeded in explaining the $1/x$ dependence satisfactorily. This has been casting a doubt about the applicability of the Fermi-liquid description.[8] However, even if we stand on the view that the Mott-Hubbard picture is appropriate in the low-x region, the rapid decrease of the Hall coefficient indicates the breakdown of the simple Mott-Hubbard picture, at least in its simplest form in the heavily doped region. A naive interpretation for this might be the two-carrier description. We believe, however, that the reason for the rapid decrease should be sought to the modification of the electronic states. If the holes were introduced to the another band in the heavily doped region, it could not reverse the sign of the Hall coefficient. Therefore, the two-hole description cannot explain the rapid decrease nor the change of the sign. On the other hand, as far as the heavily doped region is concerned, the very small or negative Hall coefficient seems not to be so different from what is expected from Fermi-liquid theory.[8] One of the plausible explanations might

be that the rapid decrease of the Hall coefficient indicates a crossover from the Mott-Hubbard picture to the Fermi-liquid-like picture. The high-T_c superconductivity seems to be realized just in the crossover region between the two.

We should comment on an alternative explanation for the rapid decrease of the Hall coefficient. In this paper, we have assumed that the Hall coefficient of the present polycrystals is dominated by the component within the plane with the current parallel to the plane and the magnetic field perpendicular to the plane because of the large anisotropy of the resistivity. In the low-x region, the anisotropy has been clearly demonstrated by the resistivity and the upper critical field measurement on single crystal.[9,10] If the conduction becomes remarkably less anisotropic with doping in the highly doped region, it is possible that the negative contribution from the other direction brings about the rapid decrease of the Hall coefficient. Recently Suzuki has reported the Hall coefficient of single-crystalline thin film with various Sr compositions.[11] In his report, the decrease of the Hall coefficient in the heavily doped region is not as rapid as the present result though it is qualitatively similar to the present result. In order to clarify this point, we have measured the Hall coefficient with the current parallel to the plane and the magnetic field perpendicular to the plane for the heavily doped single crystals with $x=0.17$. The results were plotted in Fig. 5. Although the data is somewhat sample dependent, perhaps due to the compositional inhomogeneity, the results are qualitatively in good agreement with those for polycrystals. This clearly indicates that the rapid decrease of the Hall coefficient is not due to an artifact of the anisotropic transport properties.

One further remark on the Hall coefficient is the temperature dependence shown in Fig. 6. In the antiferromagnetic phase, the Hall coefficient exhibits strongly semiconducting temperature dependence with the activation energy of the order of 10 meV. In the intermediate region, it is temperature independent like a typical metal, consistent with appearance of the T-linear resistivity. In contrast to the intermediate region, however, the Hall coefficient again shows a negative temperature coefficient ($dR_H/dT<0$) in spite of the metallic resistivity in the superconducting phase. For $x=0.075$, the Hall coefficient increases by a factor of 2 on decreasing temperature from 300 to 50 K. The negative coefficient seems to be correlated with the appearance of the superconductivity. The observation of the negative temperature coefficient in the metallic phase is common among the high-T_c cuprate systems. It is most pronounced in $Ba_2YCu_3O_{7-y}$, where the Hall coefficient fits well to the empirical relationship $R_H=A/T$.[12] At the present stage, the origin of this peculiar temperature dependence is not clear, although several mechanisms have been proposed. They are the skew scattering as a consequence of the scattering by the localized moment,[13] two-carrier model in which two types of carriers with different temperature-dependent mobilities contribute to the conduction,[14] and so on. As for the skew scattering, we cannot see any evidence for the magnetic scattering in the resistivity as in the heavy

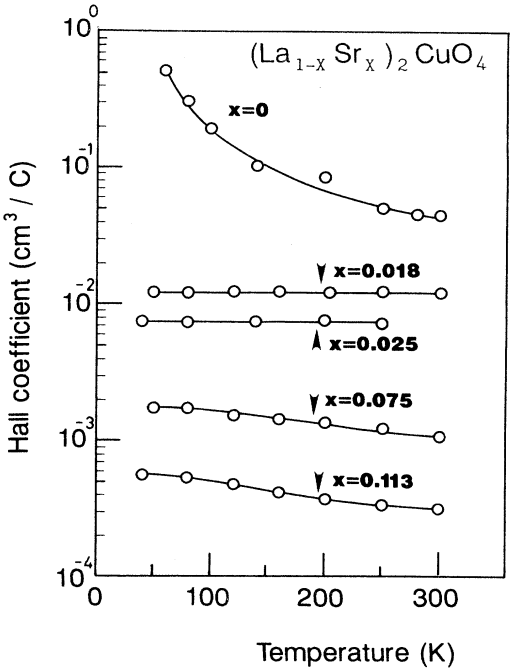

FIG. 6. Temperature dependence of the Hall coefficient for $(La_{1-x}Sr_x)_2CuO_4$ with various Sr content.

fermion system.[15] In the case of the $Ba_2YCu_3O_{7-y}$ system, it has been proposed that the presence of inequivalent Cu sites result in the two-carrier contribution. Apparently, it is not the primary origin, since the present system which does not contain the Cu-O chain shows the same temperature dependence.

C. Magnetic susceptibility

We proceed to show the effect of doping on the magnetic susceptibility. All the samples investigated here show a weak paramagnetism of the order of 10^{-7} emu/g as shown in Fig. 7. La_2CuO_4 is an antiferromagnet with $T_N\sim240$ K, as demonstrated by the presence of the peak in the temperature dependence of the susceptibility. The obvious change with Sr doping is the disappearance of the peak, indicating the suppression of the antiferromagnetic order. The peak temperature rapidly shifts to the lower temperatures and disappears at around $x=0.01$, which agrees with the previous study by Fujita *et al.*[16]

In the intermediate region, the susceptibility first shows the Curie-like temperature dependence. As the doping proceeds, the susceptibility is dominated by the component which increases with raising temperature. On the appearance of the superconductivity at around $x=0.03$, no remarkable change can be seen in the temperature dependence of the magnetic susceptibility. The susceptibility remains an increasing function of temperature, i.e., having a positive temperature coefficient in the superconducting phase. This is consistent with the results of the neutron scattering measurements which suggest that the magnetism does not change appreciably

across the insulator to superconductor transition.[17]

For higher doping, a notable change occurs which seems to be correlated with the disappearance of the superconductivity. In the superconducting phase, the increasing magnetic susceptibility is seen over the measured temperature range up to 400 K for $x < 0.075$. For $x \geq 0.075$, a broad peak appears in the temperature dependence. The broad peak shifts to the lower temperature with further doping and cannot be seen for x larger than 0.110, where T_c rapidly decreases and the superconductivity fades out. Judging from the shift of the peak temperature with doping, it is likely that the samples with $x < 0.075$ should show a peak above 400 K. Actually, the presence of the broad peak was reported for small x by Johnston *et al.*[18]

In the nonsuperconducting-metal phase, the susceptibility shows Curie-like increase with lowering temperature. Since the oxygen vacancies can be easily introduced in the heavily doped region, we have to take account of the paramagnetic moments created by the oxygen vacancies as the origin of the Curie-like behavior. We rule out the contribution of the oxygen vacancies in the following way. In Fig. 8 the temperature dependences of the magnetic susceptibility of the identical sample with $x = 0.125$ after various heat treatments is shown. As seen from the figure, after the heat treatment under the reduced atmosphere which reduces the oxygen content in the sample, the Curie-like behavior disappears and instead the temperature coefficient becomes positive like the samples with smaller x. Obviously, the oxygen vacancies are not the origin of the Curie-like behavior, but seem to act as donors and to reduce x effectively.

We ascribe the temperature-dependent magnetic susceptibility primarily to the Cu spin contribution. The extensive inelastic neutron experiments have shown that localized moments are present on Cu sites. The localized Cu spins instantaneously couple antiferromagnetically over the scale of the coherence length ξ within Cu-O plane. How is the contribution to the susceptibility from these localized Cu spins? The mother compound La_2CuO_4 can be regarded as a two-dimensional (2D) Heisenberg system of $S = \frac{1}{2}$. For the 2D $S = \frac{1}{2}$ Heisenberg system with exchange coupling $J < 0$, the calculation

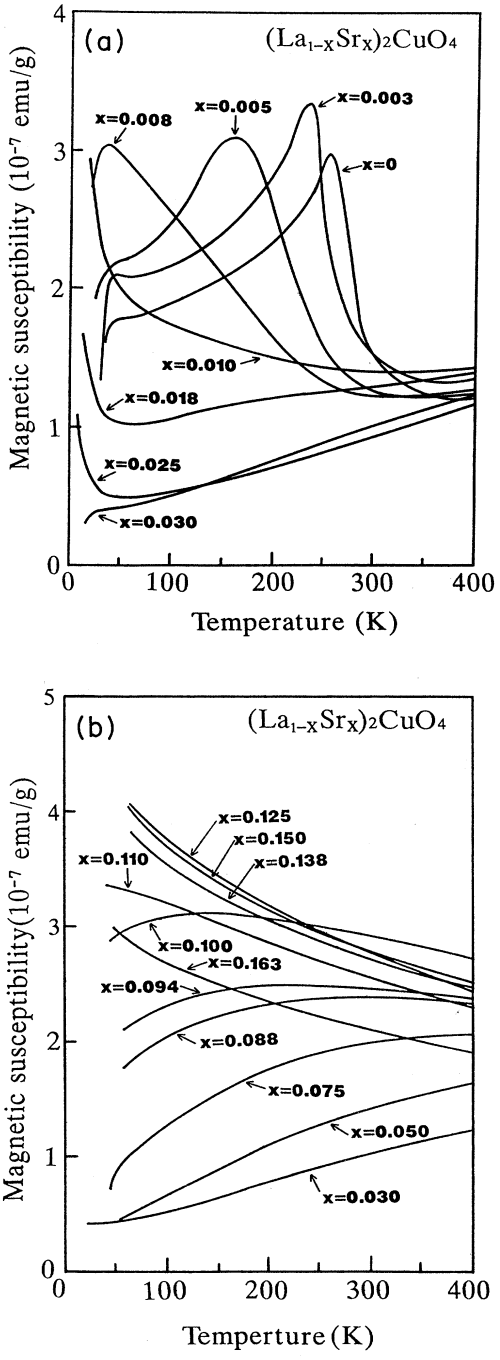

FIG. 7. Temperature dependence of the magnetic susceptibility for $(La_{1-x}Sr_x)_2CuO_4$ with various Sr content. The measurement was performed under the constant field of $H = 10$ kOe.

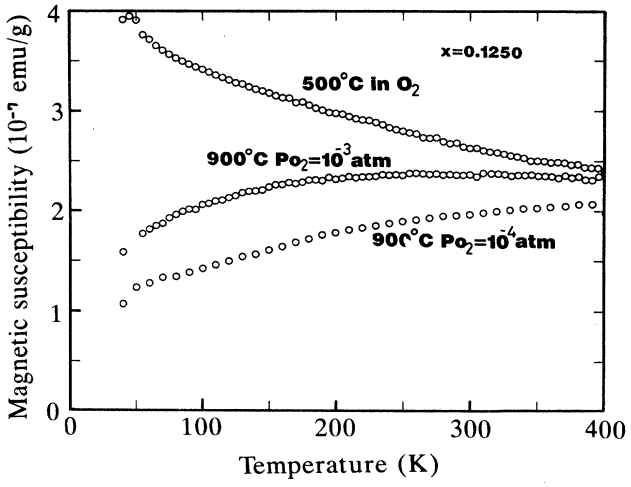

FIG. 8. Temperature dependence of the magnetic susceptibility for $x = 0.125$, (a) as prepared and annealed under Ar-O_2 gas mixture with (b) $P_{O2} = 10^{-3}$ atm and (c) 10^{-4} atm at 900 °C for 24 h.

based on the high-temperature expansion predicts that the susceptibility shows a peak at $T=0.95J/k_B$ like the 1D $S=\frac{1}{2}$ Heisenberg system.[19] This has actually been observed experimentally in some Cu quadratic layer systems. In the case of undoped La_2CuO_4, the susceptibility below 400 K is now understood to be dominated by the contribution from the very small component perpendicular to the plane produced by the orthorhombic lattice distortion.[20] In the tetragonal phase above 500 K, Johnston *et al.* have demonstrated that the susceptibility continues to increase at least up to 1000 K.[18] This should come from the contribution from the 2D antiferromagnetically coupled spin system, since the in-plane exchange coupling J is of the order of 1000 K and the susceptibility is expected to have a peak above 1000 K. We further note that related compounds with Cu-O layered structure such as $Ca_{0.86}Sr_{0.14}CuO_2$ (Ref. 21) and $Sr_2CuO_2Cl_2$,[22] which presumably possess comparable magnitude of the exchange coupling J, show similar behavior of the magnetic susceptibility at high temperatures.[23]

At present, theoretical work which addresses the susceptibility of the doped 2D $S=\frac{1}{2}$ Heisenberg system is not satisfactory. Nevertheless, it is likely that the behavior is not so different from the undoped system as far as dopant concentration is not large. Therefore, we suppose that the increasing susceptibility observed over a wide composition range originates from the antiferromagnetically coupled Cu spins. The lowering of the peak temperature with doping can be interpreted as a result of weakened antiferromagnetic correlation upon doping. Based on the neutron experiment on single crystals of $(La_{1-x}Sr_x)_2CuO_4$, Birgeneau *et al.* have shown that the correlation length is almost identical to the average distance between doped holes, i.e., $\xi \sim 3.9/\sqrt{2x}$ (Å).[17] For $x>0.110$ where the broad peak disappears, ξ is as small as ~ 7.8 Å which is about twice the nearest-neighbor Cu-Cu distance. In this case the antiferromagnetic correlation might be too weak to overcome the Curie-like contribution in the magnetic susceptibility.

Since the preceding picture proposed from the neutron experiments—the destruction of antiferromagnetic correlation in localized Cu^{2+} spin system by doped O $2p$ holes—is highly idealized, we speculate that the modification of the electronic states going to a heavily doped region, as suggested from the Hall coefficient, might also be responsible for this drastic change of the magnetic susceptibility which is suggestive of weakened antiferromagnetic correlation. This point should be clarified both theoretically and experimentally.

The observed correlation between superconductivity and magnetism seems favorable for the spin mechanism of the superconductivity. In these theories, the coexistence of the mobile carriers and the antiferromagnetic correlation between Cu spins is essential for the high-T_c superconductivity. For example, in the magnetic frustration model proposed by Birgeneau *et al.*,[24] T_c is determined by two competing effects, the increase of the density of states and the destruction of the antiferromagnetic order with doping. Thus, in the heavily doped region, T_c decreases because of the dominant contribution of the latter effect.

IV. SUMMARY

We have shown how the Hall coefficient and the magnetic susceptibility change with doping in the prototype cuprate system $(La_{1-x}Sr_x)_2CuO_4$. Particularly, we have found the modification in both charge and spin states in the heavily doped region. The characteristic features are summarized below for various composition regions. Theoretical models for the high-T_c superconductivity have to explain these changes upon doping.

(i) $0 \leq x < 0.01$. The system is an insulator. The long-range antiferromagnetic order is formed which is rapidly suppressed with doping.

(ii) $0.01 \lesssim x < 0.03$. Metallic temperature dependence of the resistivity dominates with the temperature-independent Hall coefficient like a typical metal. At low temperatures, however, carriers are localized. After the disappearance of the long-range antiferromagnetic order, the susceptibility increasing with temperature dominates which we ascribe to the persistence of 2D antiferromagnetic correlation between Cu spins.

(iii) $0.03 \lesssim x < 0.12$. The bulk superconductivity is observed. The positive Hall coefficient decreases more rapidly than that expected from the simple picture that substituted Sr supplies a hole per atom, indicating the breakdown of the Mott-Hubbard picture. The antiferromagnetic correlation is still strong enough to preserve the positive temperature coefficient of the magnetic susceptibility, although it was weakened rapidly with doping. On the appearance of the superconductivity, the Hall coefficient becomes temperature dependent, increasing with lowering temperature, which is common among high-T_c oxides.

(iv) $0.12 \lesssim x$. In this region the compound is metallic but not superconducting. The Hall coefficient decreases to the order of 10^{-4} cm^3/C, changing its sign from positive to negative. In the framework of the Fermi-liquid description, the negative sign can be understood more easily than the $1/x$ dependence in the low-x region. Possibly because the antiferromagnetic correlation length is as short as the order of the nearest-neighbor Cu-Cu distance, the magnetic susceptibility does not exhibit the negative temperature coefficient anymore. Instead, the temperature dependence is characterized by the Curie-like behavior.

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