Pressure dependence of out-of-plane conduction in $La_{2-x}Sr_xCuO_4$

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Pressure dependence of out-of-plane resistivity ρ_c has been investigated in an optimally-doped high-T_c cuprate $La_{2-x}Sr_xCuO_4$. By applying pressure, we found a drastic decrease in ρ_c which reaches 50% at $P=8$ GPa. This surprisingly large P dependence implies that the thermal contraction along the c axis is significantly reflected in the $\rho_c(T)$ curve, leading to a metal-like term; hence $La_{2-x}Sr_xCuO_4$ is essentially characterized as a two-dimensional metal modified with nonmetallic conduction between CuO_2 layers. Tunneling is suggested for a possible mechanism for ρ_c. [S0163-1829(96)05138-7]

It is known that the high-T_c cuprates are characterized by strong electron-electron correlations and by two-dimensional character of the electronic states, the latter of which is enhanced from that expected from band calculations.[1] To investigate the details of the out-of-plane conduction, which is closely related to the two dimensionality, provides a crucial test for various theories of high-T_c superconductivity.[2–6] A nonmetallic increase in the out-of-plane resistivity ρ_c at low temperatures has often been considered to reflect the exotic electronic states described in terms of the resonating-valence-bond[7–9] or the marginal-Fermi-liquid[10] theories. However, the actual temperature dependence of ρ_c, especially T-linear metallic conduction at high temperatures, which suggests a crossover from three-dimensional metallic conduction at high temperatures to two-dimensional one at low temperatures, has not been understood.

The electrical and structural properties of $La_{2-x}Sr_xCuO_4$ (LSCO), which is known as a model system of high-T_c superconductors, have extensively been investigated over the wide range of carrier concentration. However, for LSCO with $x<0.2$ the presence of structural transition prevents us from observing the generic temperature dependence of ρ_c. A change of slope is clearly observed in $\rho_c(T)$ curve at a temperature corresponding to the structural transition temperature T_d from a tetragonal to an orthorhombic phase.[11] In the tetragonal phase above T_d, ρ_c exhibits T-linear metallic conduction, but in the orthorhombic phase below T_c, ρ_c shows nonmetallic dependence. On the other hand, the in-phase resistivity ρ_{ab} shows the metallic conduction at all temperatures investigated up to 800 K.[12] These suggest that the crossover from three-dimensional to two-dimensional metallic conduction occurs as a result of the structural transition. In contrast, the optical conductivity along the c axis has been reported to show a non-Drude type, nonmetallic spectrum[13] over the temperature range in which the electrical resistivity along the c axis shows the metallic conduction. In order to resolve this controversy and to understand better the intrinsic conduction along the c axis in LSCO, it is desirable to clarify the temperature dependence of ρ_c without the influence of the structural transition.

Our thermal expansion study[14] and the reported result of x-ray diffraction under quasihydrostatic pressure[15] indicate that the suppression rate of T_d with applying hydrostatic pressure is $dT_d/dP \approx -120$ K/GPa for optimally-doped single crystalline LSCO ($x=0.15$). Therefore, the structural transition at $T_d=160$–180 K for LSCO with $x=0.15$ can be completely suppressed by applying hydrostatic pressure $P \geqslant 1.5$ GPa. We report here the pressure and temperature dependence of the electrical resistivity along the c axis at pressures up to 8 GPa.

LSCO crystals with the tetragonal $\langle 110 \rangle$ direction along the rod were grown by a traveling-solvent floating-zone method.[16] The Sr concentration of the crystals used in this work is $x=0.145$, as determined by electron-probe microanalysis. The value is consistent with x expected from the observed T_d of 170 K. The x-ray rocking curve measurement with Cu$K\alpha$ radiation gives the full width at half maximum of $0.09 \pm 0.01°$ for the (006) reflection. To our knowledge, the mosaic spread of the c axis direction indicated by this width is less than that ever reported for LSCO.[17] In order to minimize the strains and oxygen deficiencies, the crystals were annealed in flowing O_2 atmosphere at 900°C for 24 h and 500° C for 50 h. As a result, the oxygen deficiencies of the crystals, which were determined by an iodine-titration method, were 0.01 or less. Zero resistivity was observed below $T_c \approx 35$ K.

The pressure was generated by using a cubic-anvil device.[18] Nearly hydrostatic compression was achieved with a pressure transmitting medium: a mixture of Fluorinert FC70 and FC77.[19] The resistivity measurements were performed both on cooling and heating at each pressure, after the applied pressure was changed at room temperature. The pressure applied to the anvil unit was controlled to be constant within 3% during the temperature excursion.

The resistivity along the c axis ρ_c was measured by a standard four-probe method under hydrostatic pressure P up to 8 GPa in the temperature range of 4.2–300 K. The crystals were cut into rectangles with dimensions of $L_{110} \times L_{110} \times L_{001} = 0.34 \times 0.34 \times 1.2$ mm^3 in the tetragonal notation. As electrical leads, gold wires of 25 μm diameter were attached with indium solder to the electrodes made of gold paste with a post heat treatment. The $\rho_c(T)$ curves un-

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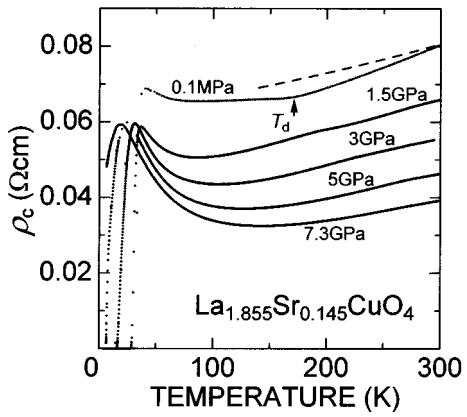

FIG. 1. Temperature dependence of ρ_c measured in the presence of pressures up to 7.3 GPa. Broken line shows the variation of ρ_c estimated simply from the thermal contraction. Arrow shows the structural transition temperature $T_d=170$ K under ambient pressure.

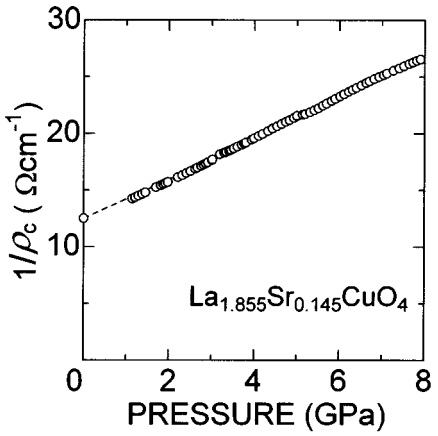

FIG. 2. The P dependence of inverse ρ_c at 297 K. The inverse ρ_c increases almost linearly up to 8 GPa. Dotted line is a guide to the eye.

der the ambient pressure before and after the high-pressure measurements agree well with each other, indicating that applied pressure did not damage the sample. It is necessary to correct for a change of the sample dimension under pressure, when we evaluate the resistivity from the resistance. From the compressibility of LSCO with $x=0.15$,[20] the correction of ρ_c amounts to about -1.6% at 8 GPa.

Figure 1 shows the temperature dependence of ρ_c for LSCO with $x=0.145$ measured on heating in the presence of P up to 7.3 GPa. A change in the slope of $\rho_c(T)$ curve measured under ambient pressure is evident at around $T_d=170$ K. By applying $P\geqslant 1.5$ GPa, which should suppress the structural phase transition completely, this feature in $\rho_c(T)$ actually disappeared. A small hump observed for $P=1.5$ GPa around 200 K is attributable to a freezing point of the transmitting medium. Even in the absence of the structrual transition, however, the nonmetallic ρ_c is observed below about 100 K. A minimum in the $\rho_c(T)$ curve at around 60 K under ambient pressure shifts to higher temperature with increasing P. These results clearly indicate that the appearance of the nonmetallic ρ_c at low temperatures does not require the orthorhombicity. The $\rho_c(T)$ of LSCO is therefore composed of three contributions: the one described by $AT+B$ at high temperatures, behavior related to the orthorhombic phase, and semiconductorlike increase at low temperatures. We note that T_c is suppressed by applying P. This suppression is reasonably explained by the uniaxial-stress effects[21,22] along the c axis, which is probably caused by a small deviation from hydrostatic stress at low temperatures.

The P dependence of ρ_c was measured at 297 K. With increasing P, we found a drastic decrease in ρ_c which reaches as small as 50% at 8 GPa. The inverse ρ_c increases almost linearly without saturation, as shown in Fig. 2. Since the slope A of the T-linear contribution at high temperatures remains nearly unchanged for P between 1.5 and 7.3 GPa, this surprisingly large P dependence is mainly in the T-independent term B. This term would be usually regarded as residual resistivity, attributable to elastic scattering processes. Such a large decrease in ρ_c could occur[23] if the carrier number changes drastically by applying P. However, it has been reported from the measurements of the Hall coefficient that the carrier number of LSCO changes little with P.[24] This large P dependence observed in the T-independent term is a key feature to be understood but seems to have been overlooked in previous studies.

Pressure causes a variation of the lattice parameters. The reported P dependence of ρ_{ab} up to 2 GPa (Ref. 25) is much smaller than that of ρ_c. The observed P dependence of ρ_c up to 8 GPa is too large to be interpreted in terms of a variation of the lattice parameters based on band theories;[26] therefore, we need a mechanism other than that predicted by band calculations.

Let us discuss this large P dependence of ρ_c in association with the variation of the lattice parameter c, or the distance between CuO_2 layers. The P dependence of the lattice parameter c at room temperature for LSCO with $x=0.15$ is approximated by $c(P)=c_0(1-k_cP+\beta_cP^2)$, in which $c_0=13.225$ Å is the lattice parameter at room temperature under ambient pressure, and the compressibility along the c axis $k_c=2.3\times10^{-3}$ $(\mathrm{GPa})^{-1}$ and the coefficient $\beta_c=4.5\times10^{-5}$ $(\mathrm{GPa})^{-2}$ are known from an x-ray diffraction measurement[20] under hydrostatic pressure up to 11 GPa. The pressure of 8 GPa reduces the lattice parameter c by 1.6%. In Fig. 3, the P dependence of ρ_c at 297 K is plotted as a function of the lattice parameter c: a small variation of the lattice parameter c induces the large change in ρ_c.

We should note that the lattice parameter c depends also on temperature. Consequently, a variation of c due to thermal contraction with the expansion coefficient $\alpha_c\approx1.7\times10^{-5}$ K^{-1} is expected to contribute equally to the change in ρ_c. The broken line in Fig. 1 shows the variation of ρ_c estimated simply from the thermal contraction and the dependence of ρ_c on c inferred from the results of the present measurements under pressure. The estimated ρ_c shows rather precise T-linear dependence at the rate of $d\rho_c/dT\approx0.8\times10^{-4}\Omega$ cm/K, while the T-linear term in the observed ρ_c has a slope of $d\rho_c/dT\approx1.1\times10^{-4}\Omega$ cm/K at ambient pressure and $0.8\times10^{-4}\Omega$ cm/K at 1.5 GPa. For the estimated value at ambient pressure, it is necessary to take account of the extra effect associated with the structural transition. Nevertheless, the observed values nearly agree with the estimated one, though it is necessary for the careful com-

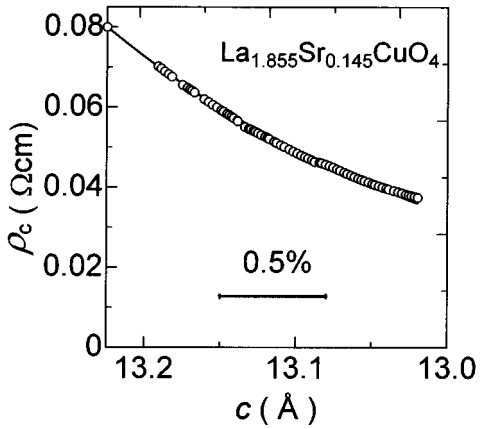

FIG. 3. The P dependence of ρ_c at 297 K plotted as a function of the lattice parameter c. A small variation of c induces the large change in ρ_c. A change in $c(P,T)$ by -0.5% correspond to the change of temperature by 300 K or applying $P\approx 2$ GPa. Solid line shows $\rho_c \approx \rho_0 \exp\{2c(P,T)/\Lambda_n\}$ with $\Lambda_n \sim 0.5$ Å based on a tunneling process.

parison to consider the change of electronic states caused by the variation of in-plane lattice parameters. This result indicates that the observed T-linear contribution in ρ_c is predominantly determined by the effect of thermal contraction along the c axis. Therefore, the intrinsic interlayer transport mechanism of LSCO is essentially nonmetallic and it is probably not necessary to introduce a metal-nonmetal crossover mechanism to describe $\rho_c(T)$.

The next question to be discussed is about the conduction mechanism along the c axis. For optimally-doped and lightly-doped high-T_c cuprates, the interpretation in terms of coherent conduction can be excluded from the considerations, because the T dependence of out-of-plane conduction for LSCO with $x=0.15$ is essentially nonmetallic and the interlayer distance is much longer than the mean-free path along the c axis at all temperatures.[5,27] Instead, high-T_c cuprates may be modeled by parallel CuO_2 layers which are weakly coupled with each other by tunneling or quantum hopping. The large T-independent term, which is characterized by strong dependence on pressure, can be understood in terms of such a tunneling process.

An exponential change in ρ_c is probably induced by a variation of c with tunneling between CuO_2 layers. The ρ_c is proportional to $t_\perp^{-2}$ with an interlayer tunneling matrix element $t_\perp$ approximately given by $t_\perp \sim \exp\{-c(P,T)/\Lambda_n\}$, where $c/2$ is the interlayer distance and Λ_n is a characteristic penetration length into the barrier. Consequently, ρ_c is expressed by $\rho_c \approx \rho_0 \exp\{2c(P,T)/\Lambda_n\}$, where ρ_0 is a constant. Here the pressure and temperature dependence of the lattice parameter c is expressed by $c(P,T)=c_0\{1-k_cP+\beta_cP^2-\alpha_c(295\ \mathrm{K}-T)\}$. The observed pressure dependence of $\rho_c[c(P)]$ at room temperature is in good agreement with that estimated from the above expression with $\Lambda_n \sim 0.5$ Å as shown in Fig. 3 by a solid line. We note here that the above expression for $\rho_c(T)$ is approximated by $AT+B$ in the wide temperature range.

High-temperature $\rho_c(T)$ is successfully reproduced by the tunneling model, which is governed by the distance between CuO_2 layers. The semiconductorlike behavior of $\rho_c(T)$ at lower temperatures, however, cannot be explained solely by such a tunneling process unless the decrease of Λ_n at lower temperatures is considered. In order to understand the out-of-plane conduction over the whole temperature reange, it is necessary to clarify quantitatively the nonmetallic increase of $\rho_c(T)$ at lower temperatures. Recently, a logarithmic increase of $\rho_c(T)$ in the temperature range of 0.3 to 30 K has been reported from the measurements in which the superconductivity is suppressed by a large magnetic field.[28] Although the presence of magnetoresistance and the behavior related to the orthorhombic phase, including the leak current through the twin boundaries,[29] may complicate the observation of the intrinsic semiconductorlike increase, the above experiment no doubt involves an important clue to the T dependence of Λ_n.

We next discuss the implication of the x dependence based on the present results, though our investigation so far has been concentrated on optimally-doped crystals. The conduction for $x<0.2$ is characterized by large anisotropy ρ_c/ρ_{ab} and the two dimensionality. The large T-independent term in ρ_c, which is interpreted in terms of the interlayer tunneling, increases with decreasing x, while the T-linear variation is attributable to the thermal contraction. On the other hand, the conduction for $x>0.2$ is characterized by bandlike transport, because the optical conductivity along the c axis recovers a Drude-type metallic spectrum.[13] As a result, P dependence of ρ_c in this region of x would probably be small. This inference can be confirmed by resistivity measurements under P for LSCO with the wide x range, which we are now promoting.

T-linear ρ_c is observed for optimally doped $YBa_2Cu_3O_{7-\delta}$, another model of high-T_c cuprates. To the best of our knowledge, $\rho_c(T)$ under high pressure has not been reported, and it is worth investigation. One important difference between $YBa_2Cu_3O_{7-\delta}$ and the present system is that the carrier density of the former changes with pressure. Hence the interpretation of the result would not be as straightforward as that presented here.

In conclusion, pressure dependence of ρ_c revealed that a small variation of the lattice parameter c induces a large change of ρ_c in optimally-doped LSCO. The observed T-linear term in ρ_c is apparent metallic conduction, because it can be regeared as a predominant contribution of thermal contraction along the c axis. As far as we know, this apparent metallic behavior has never been reported before. The nonmetallic conduction along the c axis can be described in terms of the tunneling or quantum hopping between adjacent CuO_2 layers. In this sense optimally-doped LSCO is a two-dimensional metal over the whole temperature range.

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