Pressure-Induced Dimensional Crossover and Superconductivity in the Hole-Doped Two-Leg Ladder Compound $Sr_{14-x}Ca_xCu_{24}O_{41}$

T. Nagata,[1] M. Uehara,[1] J. Goto,[1] J. Akimitsu,[1] N. Motoyama,[2] H. Eisaki,[2] S. Uchida,[2] H. Takahashi,[3]
T. Nakanishi,[3] and N. Môri[4]
[1]*Department of Physics, Aoyama-Gakuin University, Chitosedai, Setagaya-ku, Tokyo 157, Japan*
[2]*Department of Superconductivity, The University of Tokyo, Bunkyo-ku, Tokyo 113, Japan*
[3]*Department of Physics, College of Humanities and Sciences, Nihon University, Sakurajosui, Setagaya-ku, Tokyo 156, Japan*
[4]*Institute for Solid State Physics, The University of Tokyo, Minato-ku, Tokyo 106, Japan*
(Received 13 March 1998)

Anisotropic electrical resistivity under high pressure was measured for a single crystal of $Sr_{2.5}Ca_{11.5}Cu_{24}O_{41}$, a hole-doped two-leg ladder compound. Superconductivity was observed between 3.5 and 8 GPa, accompanied by metallic resistivity between the ladders, which indicates semiconducting behavior at ambient pressure. This in turn strongly suggests that the application of pressure brings about a dimensional crossover from one to two, and that superconductivity in this system is a consequence of an insulator to superconductor transition in the anisotropic two-dimensional system. [S0031-9007(98)06739-8]

PACS numbers: 74.25.Fy, 74.72.Jt

One of the most active areas stemming from high-T_c superconductivity research is the study of ladder materials [1]. Such interest has been accelerated by the theoretical prediction that hole-doped two-leg ladder compounds could have a superconducting ground state [2–4]. Numerous experiments have since been carried out in search of superconductivity, and have led to the recent discovery of superconductivity in $Sr_{0.4}Ca_{13.6}Cu_{24}O_{41.84}$ at $T_c = 12$ K under high pressure [5]. At present, the $Sr_{14-x}Ca_xCu_{24}O_{41}$ system is the only known superconducting cuprate without a two-dimensional (2D) CuO_2 plane.

$Sr_{14-x}Ca_xCu_{24}O_{41}$ is composed of alternating stacks of a plane consisting of an edge-sharing CuO_2 chain, a Sr/Ca layer, and a plane consisting of a two-leg Cu_2O_3 ladder. From optical conductivity measurements, we know that holes are transferred from the CuO_2 chains to Cu_2O_3 ladders by substituting Sr with Ca [6]. And, consequently, a relatively high Cu valence on the ladder site is realized at large x, i.e., +2.2 at $x = 11$. For $x = 11$ at ambient pressure, ρ_c, the resistivity along the ladder (c axis) is characterized by metallic, nearly T-linear dependence over a wide temperature region. In contrast, ρ_a, the resistivity between the ladder (a axis) within the ladder plane, is characterized by a negative temperature coefficient whose absolute value is about 2 orders of magnitude larger. Such findings indicate that, at ambient pressure, charge dynamics is essentially one dimensional (1D) and carriers are confined within each ladder, reminiscent of charge dynamics of high-T_c cuprates in which the carriers are confined within a CuO_2 plane [7]. Another similarity to high-T_c cuprates is the existence of a spin gap. That is, NMR [8,9] and inelastic neutron scattering [10] studies clearly show the existence of a finite gap ($\Delta = 33$ meV) in magnetic excitation even at $x = 11.5$, for which metallic ρ_c is observed. Since the spin gap behavior is a universal feature of underdoped high-T_c copper oxides, and the 2D charge dynamics is considered to be related to the existence of the spin gap [11], this makes it highly reasonable to surmise that the spin gap plays a crucial role in the 1D charge dynamics of the $Sr_{14-x}Ca_xCu_{24}O_{41}$ system.

Towards a better understanding of the nature of superconductivity in this system, charge dynamics at pressures high enough to bring about superconductivity should be clarified. As a conjecture, one might expect two possible scenarios. In the first, 1D charge dynamics is preserved under high pressure so that superconductivity is produced by the development of the pairing correlation within each ladder such that the interladder coupling plays a minor role. This scenario corresponds to original theoretical arguments which neglect interladder coupling. As for the second scenario, application of high pressure increases interladder coupling such that a dimensional crossover from one to two is eventually induced. The relaxation of charge confinement and hence coherent a-axis charge dynamics would be the result. If this were the case, interladder interactions should be taken into account explicitly, and then superconductivity might be regarded as a phenomenon in the two dimensions.

To determine which of these scenarios is indeed true, we measured the temperature dependence of the resistivity of a $Sr_{2.5}Ca_{11.5}Cu_{24}O_{41}$ single crystal under high pressure up to 8 GPa. The results show that superconductivity under high pressure is accompanied by coherent charge transport between the ladders. Such evidence strongly suggests that the application of pressure triggers a dimensional crossover from one to two, and that the superconductivity in this system can be better described as an anisotropic 2D phenomenon occurring between an array of the Cu_2O_3 ladders.

Single crystal $Sr_{2.5}Ca_{11.5}Cu_{24}O_{41}$ was grown by the traveling-solvent floating zone method under oxygen gas at 10 atm. A cubic-anvil-type apparatus was employed to generate hydrostatic pressure [12]. Resistivity was measured using a standard dc four-probe technique, and the bulk nature of the superconducting transition was confirmed by an ac susceptibility measurement.

Figures 1(a) and 1(b) show temperature dependent resistivities ρ_c and ρ_a of $Sr_{2.5}Ca_{11.5}Cu_{24}O_{41}$ at various pressures up to 4.5 GPa. At ambient pressure, ρ_c shows nearly linear temperature dependence above 130 K, while below 80 K it shows a sharp upturn indicative of carrier localization. With increasing pressure, both ρ_c and ρ_a rapidly decrease. For instance, at 3.0 GPa, the value of ρ_c at room temperature (RT) is nearly half that at ambient pressure, a large pressure effect that has not been observed in 2D cuprates [13]. Note that the resistivity upturn is weakened with increasing pressure, eventually vanishing at 4.5 GPa, and where a resistive superconducting transition shows a minimum at 9 K. There is no metallic nonsuperconducting region between the localized and superconducting regions, thus indicating that this pressure-induced superconductivity is accompanied by a superconductor-insulator (SI) transition. This is a general aspect of 2D superconductors showing a SI transition triggered by an increase in the sheet resistance, being the case of the underdoped high-T_c cuprates in which an insulating phase is realized by impurity (Zn) substitution [14]. At pressures higher than 5 GPa [inset of Fig. 1(a)], a reduction T_c is observed, qualitatively consistent with previous results for polycrystal samples [5].

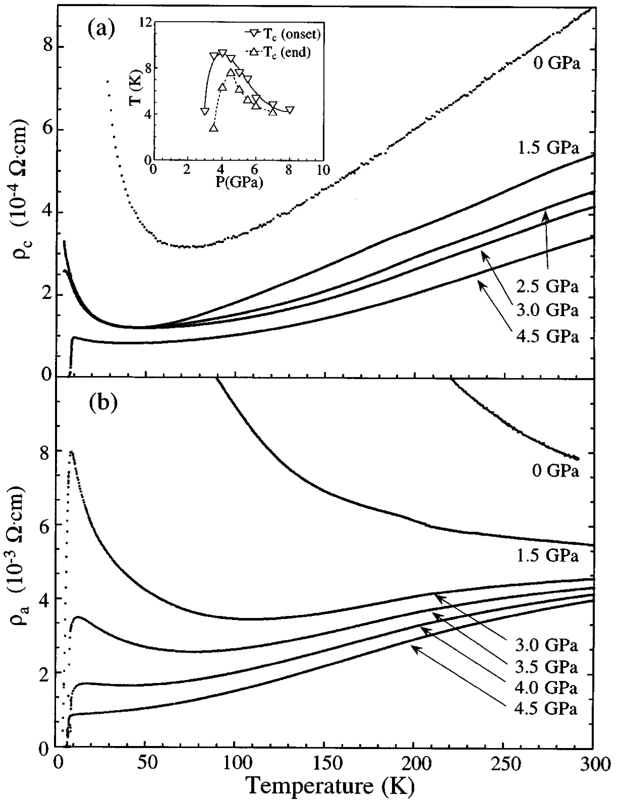

FIG. 1. Effect of pressure on temperature dependence of the resistivity (a) along the ladder direction (ρ_c) and (b) across (perpendicular to) the ladder in the ladder plane (ρ_a) of single crystal $Sr_{2.5}Ca_{11.5}Cu_{24}O_{41}$ at indicated pressures. Inset shows the pressure dependence of T_c.

The pressure dependence of ρ_a is more remarkable. At ambient pressure, it is characterized by a negative temperature coefficient indicating incoherent charge dynamics between ladders. As pressure is raised, the temperature coefficient changes to positive and the temperature region of the positive T coefficient widens. Note that at 4.5 GPa, ρ_a shows metallic behavior down to T_c, which indicates that increasing pressure induces not only superconductivity but also coherent charge dynamics perpendicular to the ladder. A resistivity drop is usually seen for pressures higher than 3 GPa as shown in Fig. 1(b). However, according to the ac susceptibility measurement, only a small volume fraction is superconducting at 3 GPa, and bulk superconductivity is realized at pressure higher than 3.5 GPa. Therefore, the onset pressure for superconductivity was defined to be 3.5 GPa in the $Sr_{2.5}Ca_{11.5}Cu_{24}O_{41}$ single crystal.

Figure 2 shows the temperature dependent anisotropy ratio ρ_a/ρ_c at various pressures, to illustrate how the anisotropy evolves with pressure. The ratio at RT is about 10 and almost independent of pressure, while at ambient pressure it gets larger with decreasing temperature, reaching 85 at 50 K owing to metallic ρ_c and insulating ρ_a. With increasing pressure, however, the enhancement in ρ_a/ρ_c is less pronounced, and the ρ_a/ρ_c vs T curve is flattened out at about 4.0 GPa where superconductivity appears. This result is again reminiscent of the evolution

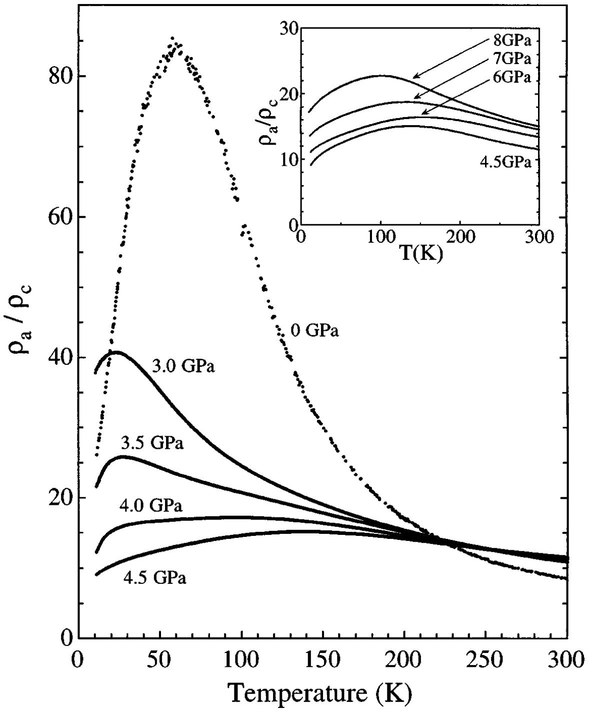

FIG. 2. Effect of pressure on temperature dependence of the anisotropic ratio ρ_a/ρ_c of single crystal $Sr_{2.5}Ca_{11.5}Cu_{24}O_{41}$ at indicated pressures. Inset shows the anisotropic ratio ρ_a/ρ_c above 4.5 GPa.

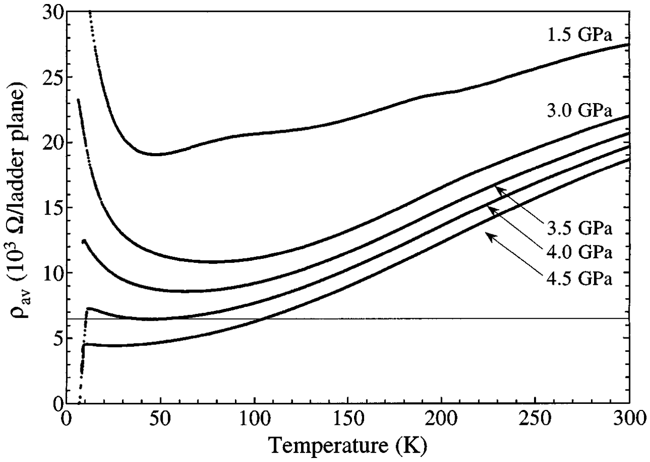

FIG. 3. Effect of pressure on temperature dependence of the average resistivity $[\rho_{\mathrm{av}} = (\rho_a^* \rho_c)^{1/2}]$ of single crystal $Sr_{2.5}Ca_{11.5}Cu_{24}O_{41}$ at indicated pressures. The horizontal line represents the universal value, $h/4e^2 = 6.5$ kΩ, i.e., the critical sheet resistance for SI transition in typical 2D superconductors.

of the anisotropic resistivity with doping in high-T_c cuprates [7] and suggests that the charge dynamics in the superconducting phase is 2D rather than 1D in the a-axis (interladder) hopping. It is noteworthy that the effect of increasing pressure is obviously distinct from that of Ca substitution. Although both ρ_a and ρ_c also decrease with Ca substitution, Ca works opposite for the evolution of ρ_a/ρ_c, enhancing anisotropy [15]. The Ca substitution is regarded as a chemical pressure because of its smaller ionic radius, leading to the lattice contraction, and its major effect on the ladders is to increase the hole density [6]. The application of pressure also has an effect of increasing carrier density in the ladders as inferred from the decrease in the slope of the ρ_c vs T curve shown in Fig. 1. However, the contrasting evolution of ρ_a/ρ_c appears to point toward another more relevant effect of pressure, possibly an enhancement of interladder hopping, which would be more crucial for the superconductivity in this system.

As to whether or not the charge dynamics under high pressure is anisotropic 2D, the following evidence might clarify. The first and strongest evidence is that ρ_a and ρ_c show quite similar temperature dependence, which indicates that both parameters are subject to the same scattering mechanism. Moreover, the recent band structure calculation by Arai and Tsunetsugu [16] estimates nearest-neighbor interladder hopping to be 5%–20% of intraladder hopping; hence, the anisotropic ratio of one-particle band mass (m_a/n_c) is from 5 to 20, in agreement with the value of ρ_a/ρ_c at high pressures. This in turn suggests that the anisotropy is mainly due to the anisotropic effective mass rather than the anisotropic scattering mechanism.

Another evidence for the 2D charge dynamics is a SI transition at around 4 GPa boundary. Figure 3 shows the temperature dependence of 2D sheet resistance [calculated from the geometrical average, $\rho_{\mathrm{av}} = (\rho_a^* \rho_c)^{1/2}$] per one Cu_2O_3 ladder plane at various pressures. Note that the critical resistivity separating insulator from superconductor nearly coincides with the universal value $h/4e^2 = 6.5$ kΩ, namely, the critical sheet resistance of SI transition in 2D superconductors [14,17,18]. This coincidence also supports the fact that the superconductivity in this system is 2D in nature, and that the role of pressure is to transform the electronic state from 1D to 2D, appearing to be indispensable for superconductivity.

Regarding the origin of 1D charge dynamics at ambient pressure, a possible scenario is that holes doped into the spin ladders form pairs (bipolarons). They are confined with a ladder but can move coherently along it. However, it is not straightforward to conclude that the superconductivity is a consequence of Bose condensation of these "preformed" pairs, since the actual situation is much more complicated.

From the present result, one may guess that the occurrence of coherent a-axis charge transport at high pressures is due to single holes dissolved from the hole pairs presumably as a consequence of an increase in the interladder coupling. The recent high-pressure NMR experiments by Mayaffre *et al.* [19] on $Sr_2Ca_{12}Cu_{24}O_{41}$ have shown that the spin gap existing at ambient pressure is filled in with increasing pressure, forming a pseudogap in the superconducting region. It is reasonable to say that spin gap filling is correlated with the progressive dissociation of hole pairs with pressure. The theory addressing the occurrence of superconductivity by the condensation of preformed pairs should accordingly be reconsidered although these pairs likely exist at ambient pressure.

Nagata *et al.* [20] have recently discovered, by the use of specific heat and neutron scattering measurements, a distinct antiferromagnetic order at $T_N \approx 2.1$ K in $Sr_{2.5}Ca_{11.5}Cu_{24}O_{41}$ at ambient pressure. While a detailed magnetic structure remains to be elucidated, it is likely that the magnetic ordering is due to some of the unpaired holes breaking up spin singlet states, and that at low temperatures the revived spins order antiferromagnetically. Accordingly, the ground state of this system at ambient pressure might be one in which both paired and unpaired holes coexist and holes and spins float in a "spin liquid sea," basically moving along the c axis at high temperature until the holes are localized at low temperature and the spins finally order at $T_N \approx 2.1$ K.

Degradation of T_c with "overpressure" is also significant. The T_c vs P curve has a dome shape, peaking at 4.5 GPa with $T_c = 9$ K, while ongoing from 5 to 8 GPa, T_c decreases to 4 K [see inset of Fig. 1(a)]; this reminds us of the overdoping effect in high-T_c cuprates. Some classes of high-T_c cuprates show a peak in T_c vs pressure, which is interpreted as being overdoped due to an increase in carrier density with pressure [21]. In light of the present results, however, a major effect of pressure on the ladder cuprate is to increase the interladder coupling, and so this behavior implies that overly strong interladder coupling does not favor superconductivity. Indeed, in $La_{1-x}Sr_xCuO_{2.5}$ discovered by Hiroi and Takano [22],

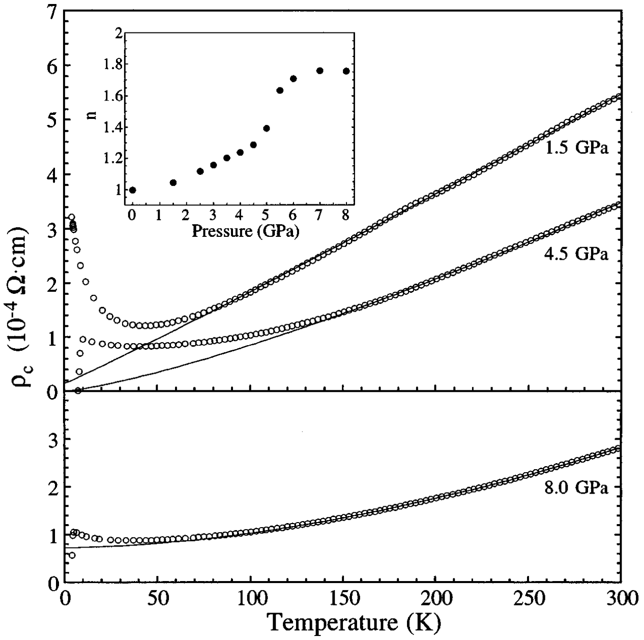

FIG. 4. Effect of pressure on temperature dependence of resistivity along the ladder direction ρ_c of a $Sr_{2.5}Ca_{11.5}Cu_{24}O_{41}$ single crystal at indicated pressures. Solid lines show the best fitted curves with power law at indicated pressures. Inset shows the pressure dependence of the power law exponent n, where $\rho = \rho_0 + AT^n$.

no sign of superconductivity exists even though holes are successfully doped. The most plausible explanation for the absence of superconductivity is a strong interladder coupling which makes this system a 3D Fermi liquid. In fact, experiments on single crystal $La_{1-x}Sr_xCuO_{2.5}$ showed that resistivity follows the characteristic T^2-temperature dependence in every direction as expected in a Fermi liquid [23]. Similar resistivity behavior is also observed in the present system at high pressures where T_c decreases. Figure 4 shows the typical examples of the T dependence of ρ_c at 1.5, 4.5, and 8.0 GPa, which were fitted with the simple power law,

$$\rho = \rho_0 + AT^n \ldots . \tag{1}$$

Solid lines in the figure indicate the best fitted curves of Eq. (1) with $n = 1.04$ (1.5 GPa), 1.29 (4.5 GPa), and 1.76 (8.0 GPa) to the experimental data above 150 K. The inset shows the pressure dependence of the exponent n, and suggests that as pressure is increased the temperature dependence changes from T linear to almost T^2 dependence, i.e., at 8.0 GPa, the T^2 dependence persists from 50 K to RT, which suggests that strong interladder interactions transform the system into a Fermi liquid with two or three dimensions as in the case of $La_{1-x}Sr_xCuO_{2.5}$.

In summary, anisotropic electrical resistivity under high pressure was measured for a single crystal of $Sr_{2.5}Ca_{11.5}Cu_{24}O_{41}$, a hole-doped two-leg ladder compound. An insulator-to-superconductor transition was observed at ~4.0 GPa, accompanied by incoherent-to-coherent crossover of the transverse (interladder) charge transport. The application of pressure therefore triggers dimensional crossover in the charge dynamics from one to two, and the superconductivity in this ladder compound might be a phenomenon in a 2D anisotropic electronic system.

This work was partly supported by a Grant-in-Aid for Scientific Research and a COE grant from the Ministry of Education, Science and Culture, Japan, and by a Grant from the New Energy and Institute Technology Development Organization (NEDO).

[1] E. Dagotto and T. M. Rice, Science **271**, 618 (1996).
[2] E. Dagotto, J. Riera, and D. Scalapino, Phys. Rev. B **45**, 5744 (1992).
[3] T. M. Rice, S. Gopalan, and M. Sigrist, Physica (Amsterdam) **199&200B**, 378 (1994).
[4] M. Sigrist, T. M. Rice, and F. C. Zhang, Phys. Rev. B **49**, 12 058 (1994).
[5] M. Uehara, T. Nagata, J. Akimitsu, H. Takahashi, N. Môri, and K. Kinoshita, J. Phys. Soc. Jpn. **65**, 2764 (1996).
[6] T. Osafune, N. Motoyama, H. Eisaki, and S. Uchida, Phys. Rev. Lett. **78**, 1980 (1997).
[7] Y. Nakamura and S. Uchida, Phys. Rev. B **47**, 8369 (1993).
[8] K. Magishi, S. Matsumoto, Y. Kitaoka, K. Ishida, K. Asayama, M. Uehara, T. Nagata, and J. Akimitsu, Phys. Rev. B **57**, 11 533 (1998).
[9] S. Tsuji, K. Kumagai, M. Kato, and Y. Koike, J. Phys. Soc. Jpn. **65**, 3474 (1996).
[10] R. S. Eccleston, M. Uehara, J. Akimitsu, H. Eisaki, N. Motoyama, and S. Uchida, Phys. Rev. Lett. (to be published).
[11] For a review, S. Uchida, Physica (Amsterdam) **282-287C**, 12 (1997).
[12] N. Môri, Y. Okayama, H. Takahashi, Y. Haga, and T. Suzuki, Jpn. J. Appl. Phys. **8**, 182 (1993).
[13] F. Nakamura, M. Kodama, S. Sakita, Y. Maeno, T. Fujita, H. Takahashi, and N. Môri, Phys. Rev. B **54**, 10 061 (1996).
[14] Y. Fukuzumi, K. Mizuhashi, K. Takenaka, and S. Uchida, Phys. Rev. Lett. **76**, 684 (1996).
[15] N. Motoyama *et al.* (unpublished).
[16] M. Arai and H. Tsunetsugu, Phys. Rev. B **56**, R4305 (1997).
[17] T. Pang, Phys. Rev. Lett. **62**, 2176 (1989).
[18] D. B. Haviland, Y. Liu, and A. M. Goldman, Phys. Rev. Lett. **62**, 2180 (1989).
[19] H. Mayaffre, P. Auban-Senzier, D. Jérome, D. Poilblanc, C. Bourbonnais, U. Ammerahl, G. Dhalenne, and A. Revcolevschi, Science (to be published).
[20] T. Nagata *et al.* (unpublished).
[21] H. Takahashi and N. Môri, Studies of High Temperature Superconductors **16**, 1 (1996).
[22] Z. Hiroi and M. Takano, Nature (London) **377**, 41 (1995).
[23] S. Miyasaka, T. Tonogai, M. Nohara, and H. Takagi (unpublished).