Dimensionality of the Cu-O double-chain site of $PrBa_2Cu_4O_8$

S. Horii* and H. Takagi
Department of Advanced Materials Science and Applied Chemistry, University of Tokyo, Hongo 7-3-1, Bunkyo-ku, Tokyo 113-8656, Japan

H. Ikuta
Center for Integrated Research in Science and Engineering, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan

N. E. Hussey
H. H. Wills Physics Laboratory, University of Bristol, Tyndall Avenue, Bristol, BS8 1TL, United Kingdom

I. Hirabayashi
Superconductivity Research Laboratory, ISTEC, Mutsuno 2-4-1, Atsuta-ku, Nagoya 456-8587, Japan

U. Mizutani
Department of Crystalline Materials Science, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan
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We report measurements of the resistivity ρ, a-axis magnetoresistance $\Delta\rho_a/\rho_a$ and Hall coefficient R_H of $PrBa_2Cu_4O_8$ (Pr124) single crystals. The in-plane anisotropy ρ_a/ρ_b in Pr124 is extremely large, reflecting the quasi-one-dimensional nature of the highly conducting Cu-O chains oriented along the b axis. Interchain resistivity, however, also shows metallic behavior below a peak at $T=140$ K, where intriguingly, $R_H(T)$ has a minimum. $\Delta\rho_a/\rho_a$ $(B//c)$ is large, positive and obeys Kohler's rule below 100 K. Moreover, the T dependence of square root of $\Delta\rho_a/\rho_a(T)$ follows closely that of $l_{\mathrm{chain}}(T)$ where $l_{\mathrm{chain}}(T)$ is the mean-free-path of the carriers along the chain direction determined from $\rho_b(T)$. Collectively, these results prove the existence of a finite and coherent interchain charge transfer along the a-axis direction at low temperature, in the absence of mobile carriers within the CuO_2 planes.

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I. INTRODUCTION

The ground state of one-dimensional (1D) metals, the so-called Tomonaga-Luttinger (TL) liquid, is theoretically predicted to be dramatically different from that of a 3D Fermi liquid. Most notably, the spin and charge degrees of freedom are separated.[1,2] Experimental studies on carrier-doped 1D systems are frequently aimed at seeking evidence for such departure from FL physics. The Cu-O chain structure found in some of the high-temperature superconductors and related materials is an excellent candidate for a 1D conductor provided enough mobile carriers are doped. Indeed, experimental evidence for TL behavior has surfaced in recent years. Kim *et al.* studied angle-resolved photoemission spectroscopy (ARPES) in $SrCuO_2$ and claimed evidence for spin-charge separation.[3] Mizokawa *et al.* also reported ARPES data for the Cu-O chain site of $PrBa_2Cu_3O_7$ (Pr123) and revealed two structures that could be interpreted as holon and spinon bands.[4] It is well known, however, that the chain structure in both $SrCuO_2$ and Pr123 is insulating. On the other hand, the behavior of Cu-O chains is totally different when they are doubly stacked. While the only essential difference in the structure of $PrBa_2Cu_4O_8$ (Pr124) and Pr123 is the Cu-O chain site that is running parallel to the b-axis as shown in Fig. 1, resistivity measurements in polycrystalline Pr124 (Refs. 5–7) and $YBa_2Cu_4O_8$ (Y124) single crystals[8] suggest that the Cu-O double-chain site is a good conductor with self-doped, mobile carriers. The striking difference in the electronic conduction of the Cu-O chain structures in the 123 and 124 compounds might stem from the structural stability, i.e., the carriers on the single chain site are more easily localized due to oxygen defects.

In Pr124, electronic conduction and superconductivity in the CuO_2 planes are suppressed, presumably due to the hybridization of Pr $4f$ and O $2p$ orbitals.[9] Therefore, Pr124 offers a unique opportunity to directly probe the metallic state of the Cu-O structure without being influenced by carriers in the plane site. Obviously, high quality single crystals are necessary for extracting the behavior of the metallized chains and studying the anisotropic transport properties. Recently, we have successfully grown single crystals of Pr124 by means of a flux method under high oxygen pressure.[10] Resistivity measurements revealed a large in-plane anisot-

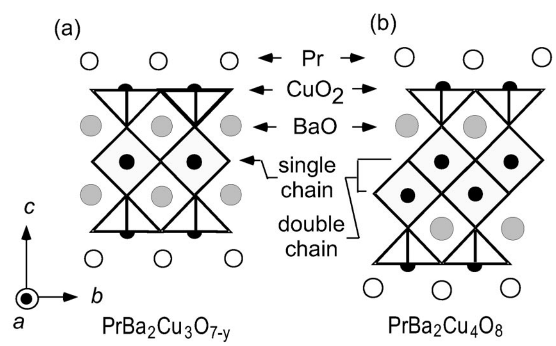

FIG. 1. Schematic pictures of the crystal structure of Pr123 (a) and Pr124 (b).

ropy and the highly conducting nature of the Cu-O double chain site down to low temperatures. However, the dimensionality of the chain structure remained an open question. The *a*-axis resistivity (ρ_a) exhibits metallic behavior at low temperature in the sense that it decreases with temperature after passing a peak.[10] A possible scenario is that the Cu-O site undergoes a 1D to 2D (or 3D) dimensional crossover upon lowering the temperature, similar to some organic compounds. Terasaki *et al.* reported some evidence of such crossover at about 100 K from a study using a magnetically aligned Pr124 polycrystalline sample.[11] The polarized optical spectrum measured at 300 K can be interpreted within the framework of a TL liquid[12] while ARPES data at 10 K do not reconcile with the TL liquid picture,[13] which seems to support the dimensional crossover scenario.

To shed more light on the dimensionality of the Cu-O chain site of Pr124, we have studied the *a*-axis magnetoresistance (MR) and Hall effect (with $I//b$) of Pr124 single crystals. Our results give a consistent picture that supports the establishment of a FL ground state in Pr124 at low temperature with coherent interchain conductivity along the *a* axis.

II. EXPERIMENTAL

Single crystals of Pr124 were grown by a self-flux method in MgO crucibles under high-pressure oxygen gas of 11 atm, as reported previously.[10] The typical size of the crystals was approximately $0.5\ \mathrm{mm}\times 0.2\ \mathrm{mm}\times 0.04\ \mathrm{mm}$. The direction of each crystallographic axis was determined by using a polarizing microscope or a precession x-ray camera. Microscopic observations revealed no indication of twinning. *a*-axis and *b*-axis resistivity measurements were carried out between 2 and 300 K using a standard ac four-probe method. The *a*-axis MR measurements were conducted in a 9 T magnetic field aligned along each crystallographic axis. $R_H(T)$ was measured in a six-probe configuration, with two pairs of voltage electrodes attached to the same crystal to ensure consistency (see the inset to Fig. 5). The magnetic field in the Hall effect measurements was swept from -5 T to 5 T with an interval of 1 T.

III. RESULTS AND DISCUSSION

The results of resistivity measurements of Pr124 along *a*- and *b*-axes were reported in our previous paper.[10] These data are replotted as a function of T^2 in Figs. 2(a) and 2(b) for $\rho_a(T)$ and $\rho_b(T)$, respectively. The inset of each figure shows the linear plot reported in the previous paper.[10] As shown schematically in Fig. 1, *a* and *b* axes are running perpendicular and parallel to the chains, respectively. Superconductivity was not detected down to 2 K for all crystals that were examined. The value of $\rho_a \sim 4$ mΩ cm at 300 K, indicating that the number of mobile carriers within the CuO_2 plane is significantly less than that of optimally doped high-temperature superconductors. $\rho_a(T)$ possesses a maximum at $T_{\max}\sim 140$ K, below which ρ_a decreases with decreasing temperature down to 2 K, reaching a value ~ 4 mΩ cm [inset of Fig. 2(a)]. As can be seen from Fig.

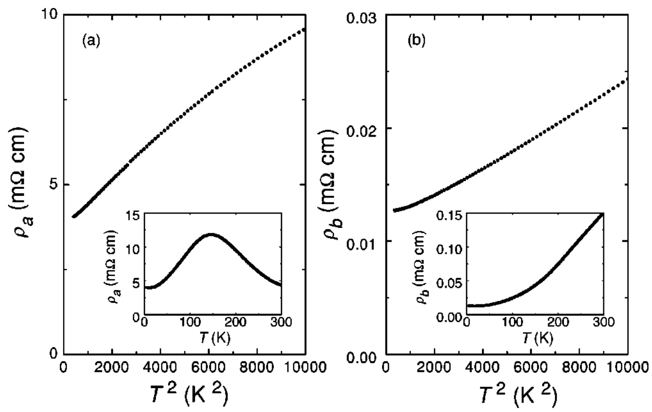

FIG. 2. Resistivity of Pr124 along *a* (a) and *b* axes (b) as a function of T^2. Each inset shows the same $\rho_a(T)$ and $\rho_b(T)$ data as a function of *T*, as previously reported in Ref. 10.

2(a), $\rho_a(T)$ follows fairly well the T^2 law expected for a Fermi liquid at low temperature (below about 60 K). A similar behavior was reproduced in all crystals for which ρ_a was measured.

$\rho_b(T)$ has a metallic temperature dependence in the whole temperature range down to 2 K, as shown in the inset of Fig. 2(b). The residual resistivity of ρ_b of this crystal is estimated to be 12 $\mu\Omega$ cm by extrapolating the low-temperature data, though residual resistivities as low as 4 $\mu\Omega$ cm have been observed.[14] Similarly to $\rho_a(T)$, $\rho_b(T)$ also followed well the T^2 law at low temperature, as shown in Fig. 2(b). Intriguingly, this behavior of $\rho_b(T)$ in Pr124 is consistent with the behavior of the resistivity along the Cu-O double chain site of Y124 extracted from the in-plane resistivity via a parallel resistor model.[15]

The resistive anisotropy ρ_a/ρ_b is roughly 25 at 300 K, then increases with decreasing temperature, exceeding 300 below $T_{\max}$. The large anisotropy between the *a*- and *b*-axis directions implies that the metallic behavior of ρ_b is governed solely by the double chain site. We pointed out in our previous paper[10] that there are at least two possibilities for the origin of the peak in $\rho_a(T)$. First, the conduction along the CuO_2 planes dominates that across the CuO chains for the whole temperature range, and the peak in $\rho_a(T)$ simply indicates a change in the conduction mechanism intrinsic to the plane site. The second possibility is that the peak signifies a switching of the dominant conduction path from CuO chain to CuO_2 plane with increasing temperature. In the latter interpretation, conduction along *a* and *b* axes are both governed by the Cu-O chain site at low temperature, and the Fermi-liquid behavior can be attributed to the two- (or three-) dimensionality of the Cu-O chain site. In this regard, it should be noted that we have recently observed a metallic *c*-axis transport in Pr124, with a similar magnitude to $\rho_a(T)$ and a T^2-dependence at low temperatures.[14]

Figure 3 shows the *T* dependence of $\Delta\rho_a/\rho_a(T)$ in a magnetic field of 9 T applied along the *b* and *c* axes. The isotropic contribution to MR has been removed from the raw data by subtracting the longitudinal MR measured with $B//a$. For $B//b$, the magnitude of MR was comparable with our experimental resolution for the whole temperature range,

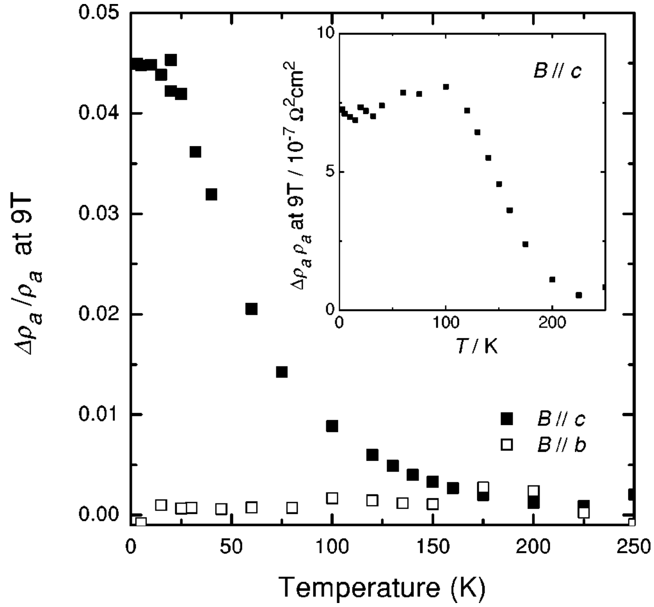

FIG. 3. Temperature dependence of a axis MR $\Delta\rho_a/\rho_a$ under magnetic field of 9 T applied parallel to b axis (open squares) and c axis (closed squares). Inset: Kohler analysis of the orbital a axis MR data for Pr124, as explained in the text.

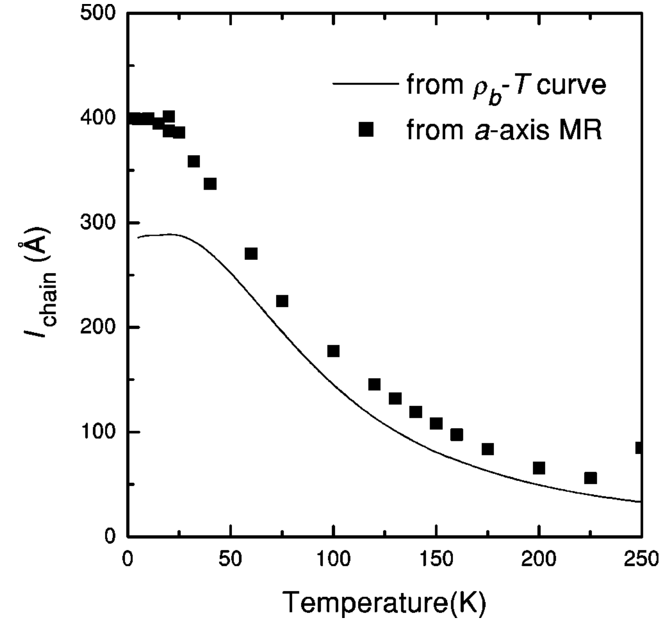

FIG. 4. Temperature dependence of the mean-free-path of the chain site of Pr124 estimated from $\rho_b(T)$ (solid line) and $\Delta\rho_a/\rho_a(T)$ (closed squares).

and similarly with $B//c$ above $T=150$ K. However, below 150 K, a sizable MR develops for $B//c$, which increases monotonically down to 3 K. The value of $\Delta\rho_a/\rho_a \sim 4.5\%$ at 3 K, where $\Delta\rho_a$ is defined by the difference in ρ_a at 9 T and zero field; $\rho_a(B=9\text{ T})-\rho_a(0)$. If the carriers are moving coherently across the chains, a finite positive $\Delta\rho_a$ is expected for $B//c$ since the Lorentz force $F=e[v_{\rm chain}\times B]$ is largest in this configuration. For $B//b$, the field is parallel to the main velocity component $v_{\rm chain}$, the Lorentz force is a minimum and the orbital MR is correspondingly small. Hence, the MR data plotted in Fig. 3 can be understood, at least qualitatively, if a-axis transport is dominated by the interchain carriers and the CuO_2 plane site is irrelevant for a-axis transport.

Further insight into the electron dynamics can be gained from performing Kohler analysis on the orbital MR data. In a conventional metal, Kohler's rule is written as $\Delta\rho/\rho \sim f(B/\rho)\sim(B/\rho)^2$ for an orbital MR varying as B^2. Thus, at a fixed field, the product $\Delta\rho_a\cdot\rho_a$ should be constant, independent of temperature. In one sense, Kohler's rule implies that all the carriers that determine the form of the zero-field resistivity are susceptible to the same scattering mechanisms in a magnetic field. In the inset of Fig. 3 we have plotted $\Delta\rho_a\rho_a$ (at 9 T) as a function of temperature. As can be seen, Kohler's rule is obeyed below 100 K, indicating coherent a-axis conduction in this temperature range, but at higher temperatures, deviations from Kohler's rule appear as interchain conduction becomes incoherent and additional contributions to the (zero-field) resistivity emerge. Indeed, optical reflectivity data have revealed that the electronic state of the Cu-O chain site has a strong 1D character at $T=300$ K.[12] Intriguingly, this crossover temperature is much smaller than $T_{\rm max}$ ($\sim$140 K), implying that these temperatures are uncorrelated.

For a more quantitative argument about the nature of the interchain conduction, we checked the consistency of $l_{\rm chain}$ estimated by two different methods. If the double-chain site has a quasi-1D electronic structure and the chain bands are represented in reciprocal space by two undulatory sheets along a- and c-axes, one obtains the following equation from the linearized Boltzmann equation in the low magnetic field limit for $B//c$:

$$l_{\rm chain}=\frac{\hbar}{eBa}\sqrt{\frac{\Delta\rho_a}{\rho_a}}, \tag{1}$$

where a is the interchain distance along a axis ($\sim$3.88 Å).[16] Similarly, $l_{\rm chain}$ is related to ρ_b by

$$l_{\rm chain}=\frac{ac\pi\hbar}{2e^2}\,\frac{1}{\rho_b}, \tag{2}$$

where c represents the inter chain distance along the c-axis direction ($\sim$13.7 Å).[17]

The temperature dependence of $l_{\rm chain}$ estimated by these two methods is compared in Fig. 4. The a axis MR for $B//c$ in Fig. 3 is very small above 150 K, and therefore one can hardly discuss the consistency of $l_{\rm chain}$ estimated using the two methods. Below 150 K, on the other hand, we can safely conclude that the agreement between the two methods is quite satisfactory taking into account that $l_{\rm chain}$ at low temperature is affected by the residual resistivity that is sample dependent. This agreement appears to confirm that $\rho_a(T)$ is indeed governed by the conduction across the Cu-O chains at low temperature. It is also interesting to note that we observed quite recently a metallic behavior for c-axis resistivity

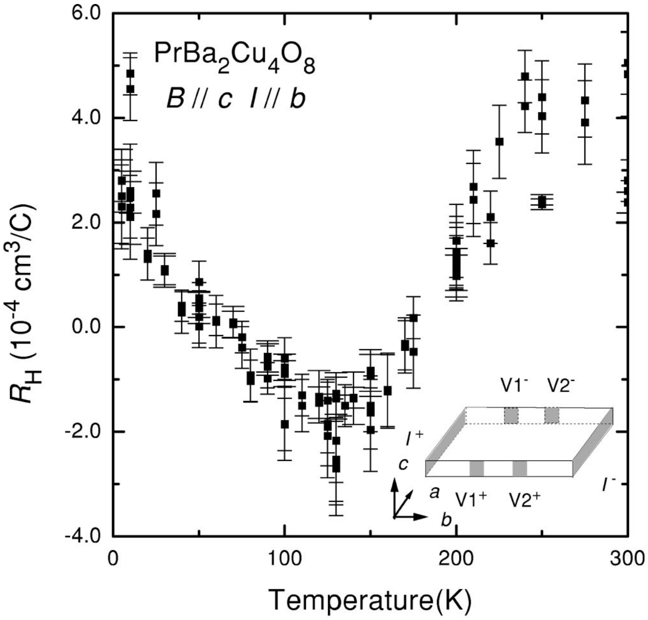

FIG. 5. Temperature dependence of the Hall coefficient along the *a*-axis direction ($B//c$, $I//b$). A six-probe configuration as schematically shown in the inset was adopted that facilitates an easy double check of the results.

of Pr124,[14] and a magnetic field induced metal-insulator transition of $\rho_c(T)$ at approximately 20 T with $B//a$,[18] similarly to the one observed for Y124.[19] These results suggest that the conduction along the *c*-axis direction is also governed by interchain conduction, and the Cu-O chain site is essentially 3D with a Fermi liquid ground state. As discussed above, Kohler's rule is violated in the present compound above approximately 100 K (inset to Fig. 3). Therefore, we think that the conduction across the chains starts to lose coherency above 100 K, but still the interchain transport dominates that of the CuO_2 plane up to, at least, 150 K.

We also measured the Hall effect in a geometry in which the Lorentz force exerted on the electrons is directed along the *a* axis, i.e., $I//b$ and $B//c$. We measured the T dependence of $R_H(T)$ for three different crystals, two of which were double-checked with two pairs of Hall voltage electrodes. Figure 5 summarizes these five data sets, and the excellent reproducibility shows that the results reflect the intrinsic $R_H(T)$. The value of R_H is roughly 4.0×10^{-4} cm^3/C at 300 K, which is extremely small compared with the in-plane R_H of optimally doped cuprates. R_H monotonically decreases with decreasing temperature down to about 130 K, changing sign at $T = 170$ K. Below 130 K, R_H begins to become more positive, changing sign once more and at 5 K, $R_H \sim 3.0 \times 10^{-4}$ cm^3/C. This value is rather small, given the size of k_{F_b} estimated from ARPES (Ref. 13) and optical measurements,[12] and may reflect an additional (small) contribution from the plane carriers or a reduced Hall voltage parallel to the *a* axis due simply to the low interchain conductivity.

Although the peculiar T dependence of $R_H(T)$ is not understood, it is intriguing to note that the temperature where $R_H(T)$ has its minimum corresponds closely with T_{max} in the inset of Fig. 2(a), suggesting that a drastic change in the transport behavior occurs around this temperature. From our *a*-axis MR data, we have inferred that coherent *a*-axis conduction is generated from carriers on the double chain site but that coherence may be lost around 100 K. Collectively, the MR data and the $R_H(T)$ data suggest that there are two relevant temperature scales in the problem, i.e., $T = 100$ K and $T_{max} = 140$ K, with the latter more likely reflecting an emerging contribution from the CuO_2 plane carriers at higher temperatures. Indeed, in untwinned semiconducting Pr123, where the chains play no role in the conduction process, $\rho_a \sim 10$ mΩ cm at $T = 300$ K (Ref. 20) compared with $\rho_a \sim 4$ mΩ cm for Pr124. This suggests that the CuO_2 planes do indeed start to contribute to the *a*-axis transport in Pr124 at high temperatures, even though they are nonmetallic.

IV. CONCLUSION

In summary, we have studied the resistivity, magnetoresistance, and Hall effect of Pr124 single crystals. The conduction along the *a*- and *b*-axis directions is very anisotropic, with $\rho_a/\rho_b \sim 300$ at 3 K, reflecting the highly 1D nature of the Cu-O chains running parallel to the *b*-axis direction. Nevertheless, both ρ_a and ρ_b followed well the T^2 law that is expected for a Fermi-liquid state at low temperature. The *a*-axis MR data were quantitatively analyzed in terms of a quasi-1D band model, and the results support the presence of a finite and coherent interchain interaction along the *a*-axis direction at low temperature. Moreover, the MR data followed Kohler's rule below 100 K. Hence, both the resistivity and MR data indicate that the chain site is essentially 2D (or 3D) in nature, and the low temperature state is indeed a Fermi liquid. Finally, we have presented evidence that the CuO_2 planes may begin to play some role in the *a*-axis conduction at higher temperatures.

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*Present address: Department of Applied Chemistry, Graduate School of Engineering, University of Tokyo, Hongo 7-3-1, Bunkyo-ku, Tokyo 113-8656, Japan

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[17] When an electric field E_y is applied along b-axis, the current density J_{yy} along this direction is given by $J_{yy}=\int e v_k g_k d^3k$, where $g_k(=-\tau e E_y v_{yy} df/d\epsilon)$ is proportional to the difference of the Fermi distribution function between the equilibrium state and the steady state with E_y applied. Assuming a linear energy dispersion near the Fermi energy along b direction and cosine bands along the other two directions yields then Eq. (2).

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