Normal-state transport properties of slightly overdoped $YBa_2Cu_3O_y$ crystals prepared by a crystal-pulling technique

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Anisotropic transport properties of high-temperature superconductors were extensively investigated for $YBa_2Cu_3O_y$ crystals prepared by a crystal-pulling technique. Since the crystals are as large as $4\times4\times2$ mm^3, we have successfully measured their resistivity, Hall coefficient, and thermopower along both the in-plane and out-of-plane directions, by using samples cut from the same crystal. The measured in-plane data have revealed that our samples are slightly overdoped when highly oxygenated. This means that our samples have the least anisotropic resistivities between the in-plane and out-of-plane directions among high-T_c cuprates. The Hall coefficient and the thermopower along the out-of-plane direction qualitatively agree with the band calculation, but the scattering time along the out-of-plane direction is anomalously short. Thus, it is concluded that the two-dimensional nature in the electronic states of high-T_c cuprates is mainly characterized by the anomalously short scattering time along the out-of-plane direction, not by the anisotropic effective masses.

I. INTRODUCTION

As is widely accepted, high-temperature superconductivity occurs in the doped CuO_2 plane whose electronic states are highly anisotropic between the in-plane and out-of-plane directions.[1] Owing to this nature, it has been discussed for a long time whether the electronic states of high-T_c superconducting cuprates (HTSC) are essentially two-dimensional (2D) or three-dimensional (3D). If they are 2D, we need to consider exotic concepts such as the resonating-valence-bond (RVB) theory.[2] In this case we have to regard the electronic states of HTSC as *non-Fermi liquid*. On the other hand, if 3D, we can employ a *Fermi-liquid* approach based on a conventional band picture, no matter how much it might be modified. To discuss the dimensionality one must measure and analyze various quantities along both the in-plane and out-of-plane directions.

Among the anisotropic properties of HTSC, normal-state transport is of particular importance. One reason is that it shows anomalous temperature dependence, which a Fermi liquid is unlikely to show.[3] A prime example is that in-plane resistivity (ρ_{ab}) and out-of-plane resistivity (ρ_c) have different temperature dependence. In particular, ρ_c in underdoped HTSC increases with decreasing temperature, while ρ_{ab} shows metallic conduction.[4–8] In terms of ρ_c/ρ_{ab} that is often regarded as a measure of anisotropy,[9] highly oxygenated $YBa_2Cu_3O_y$ is least anisotropic, whereas $Bi_2Sr_2Ca_{n-1}Cu_nO_y$ is most anisotropic.

There are two ways to study the anisotropy in HTSC. One is to investigate the most anisotropic HTSC, from which one can find the 2D nature inherent in HTSC. For this purpose, $Bi_2Sr_2Ca_{n-1}Cu_nO_y$ and $La_{2-x}Sr_xCuO_4$ are appropriate. The other way is to investigate the least anisotropic HTSC, from which one can examine to what extent a Fermi-liquid picture is valid. In this case, highly oxygenated $YBa_2Cu_3O_y$ is the most suitable material. We have studied the anisotropic transport of $YBa_2Cu_3O_y$ and discussed the dimensionality of its electronic states,—by following the latter strategy.[10,11]

Moreover there are many advantages in the studies of $YBa_2Cu_3O_y$. First, high-quality crystals with narrow transition width (less than 0.5 K) are available. Second, $YBa_2Cu_3O_y$ has been studied most extensively among HTSC including neutron experiments, which have detected a gap structure in the spin excitation spectrum.[12,13] Thus the enormous amount of compiled data is available for the discussion of our results. Third, a large single crystal of $YBa_2Cu_3O_y$ has been grown by a crystal-pulling (modified Czochralski) technique,[14] which enables us to measure the out-of-plane transport as precisely as the in-plane one.

In this paper we report on the measurements and analyses of the normal-state transport properties of $YBa_2Cu_3O_y$ along the in-plane and out-of-plane directions. We prepared two kinds of $YBa_2Cu_3O_y$ crystals, grown by a crystal-pulling technique and by a conventional CuO flux technique. We used the former crystals for the study of anisotropy, and the latter ones as a reference. Although we used twinned crystals, twin structures do not seriously affect the discussion of the anisotropy between the in-plane and out-of-plane directions. This paper is organized as follows: In Sec. II we briefly mention the sample preparation and the experimental procedure; in Sec. III we present the results to specify what is anomalous in the normal-state transport of $YBa_2Cu_3O_y$. Finally we quantitatively discuss three topics: the doping level of our samples, the analysis based on a band theory, and the anomaly in the scattering time.

II. EXPERIMENTAL

We prepared single crystals of $YBa_2Cu_3O_y$ by a crystal-pulling technique.[14] A typical dimension of the as-grown crystals was $4\times4\times2$ mm^3, which is much larger than that of single crystals by a flux technique. We cut the crystals into a rectangular shape, and painted gold paste to make good contacts (a typical contact resistance was less than 1 Ω). Then we highly oxygenated the samples by annealing in an oxygen flow at 400 °C for 4–10 days. For comparison, we also prepared $YBa_2Cu_3O_y$ crystals by a flux technique using gold crucibles,[15] and annealed them under the same conditions. Since the crystals by the flux technique were very thin along the out-of-plane direction (typically less than 50 μm), we

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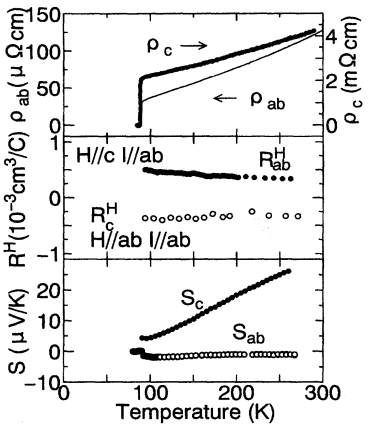

FIG. 1. Normal-state transport of highly oxygenated $YBa_2Cu_3O_y$ grown by a crystal-pulling technique. In-plane (ρ_{ab}) and out-of-plane (ρ_c) resistivities are shown in the top panel; Hall coefficient with magnetic field parallel to the c axis (R^H_{ab}) and that with magnetic field perpendicular to the c axis (R^H_c) are shown in the middle panel; in-plane (S_{ab}) and out-of-plane (S_c) thermopowers are shown in the bottom panel.

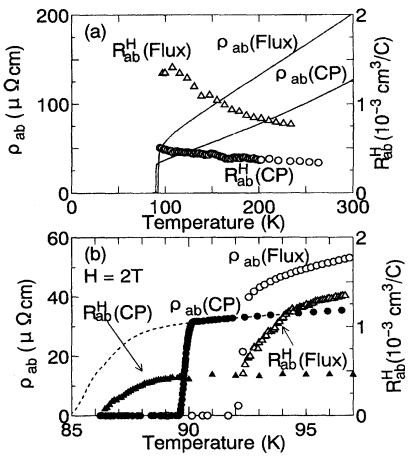

FIG. 2. In-plane resistivity (ρ_{ab}) and Hall coefficient (R^H_{ab}) for two $YBa_2Cu_3O_y$ crystals prepared by a crystal-pulling (CP) technique and a conventional CuO-flux (Flux) technique. (a) $0 \leq T \leq 300$ K; (b) near T_c. Note that the dotted curve represents the resistive transition of the CP sample in 2 T.

measured only the resistivity and the Hall coefficient along the in-plane direction. To distinguish these two kinds of crystals, we will call those by the crystal-pulling technique "CP samples," and those by the flux technique "flux samples."

We measured resistivity by a standard four-probe method with applied current density I of 0.1–10 A/cm^2. We employed a six-probe configuration for Hall-coefficient measurement in magnetic field H of 8 T, and eliminated zero-field signals due to the misalignment of the voltage leads by sweeping magnetic fields from -8 to 8 T or by rotating samples in a static field. As for thermopower, we pasted one edge of the sample to a copper block working as a heat bath, and the other edge to a resistance heater. A typical temperature gradient was 0.1–0.5 K/mm. The contribution from the voltage leads was carefully subtracted.

III. RESULTS

The normal-state transport of highly oxygenated CP samples is summarized in Fig. 1. Both ρ_{ab} and ρ_c exhibit a superconducting transition near 90 K with a transition width of 0.4 K. The observed resistivities are qualitatively consistent with a preliminary report on the CP samples.[16] The in-plane Hall coefficient (R^H_{ab}) increases with decreasing temperature, and its magnitude (5×10^{-4} cm^3/C just above T_c) is nearly half of a typical value for a flux sample. The out-of-plane Hall coefficient (R^H_c) (Ref. 17) is negative and essentially independent of temperature. The in-plane thermopower (S_{ab}) is negative, while the out-of-plane thermopower (S_c) is positive. In addition, S_{ab} and S_c have different temperature dependence.

First of all, we emphasize that the data in Fig. 1 are quantitatively different from typical data for a highly oxygenated flux sample. We attribute the difference to different doping levels, that is, the CP sample is slightly overdoped, whereas a highly oxygenated flux sample is optimally doped. To see the difference more clearly, we compare ρ_{ab} and R^H_{ab} for the CP sample with the data for the flux sample in Fig. 2(a). There are three notable differences between the two ρ_{ab}'s. One is that ρ_{ab} of the CP sample is lower. The second difference is that ρ_{ab} of the CP sample draws a slightly positive curvature as a function of temperature, whereas the flux sample exhibits the T-linear resistivity. The last one is seen in the superconducting transition. While the resistivity for the flux sample has a characteristic rounding (deviation from the T-linear resistivity) well above T_c, the CP sample has a sudden drop of ρ_{ab} just above T_c. The rounding has been explained in terms of superconducting fluctuation,[18] and accordingly the fluctuation less affects ρ_{ab} in the CP sample. These three features in ρ_{ab} of the CP sample are characteristic of overdoping. It is also confirmed by Hall coefficients: R^H_{ab} is smaller and its temperature dependence is weaker in the CP sample than in the flux sample. As is seen in the Tl-based cuprates,[19] R^H_{ab} rapidly decreases in magnitude and its temperature dependence becomes weaker when doping goes from the optimal to the overdoped region. Thus we regard the observed R^H_{ab} for the CP sample as further evidence for overdoping. The negative S_{ab} of the CP sample also suggests overdoping. It has been reported that the thermopower in 90-K phase $YBa_2Cu_3O_y$ is extremely sensitive to the doping levels.[20,21] By comparing the reported thermopower of 90-K phase $YBa_2Cu_3O_y$,[20] we estimate the oxygen content y in our highly oxygenated CP sample to be 6.93–6.95.

We point out an additional feature suggestive of overdoping. Ito, Takenaka, and Uchida have reported that R^H_{ab} of $YBa_2Cu_3O_y$ takes a maximum near a certain temperature T^*,[22] at which an energy gap opens in the magnetic excitation spectrum.[23] As doping proceeds (y increases), T^* is lowered towards T_c. In the highly oxygenated flux sample the maximum of R^H_{ab} is somewhat above T_c (around 100 K), as shown in Fig. 2(a). On the contrary, R^H_{ab} of the CP sample monotonically increases with decreasing temperature until superconductivity sets in. To see this more clearly we replot R^H_{ab} and ρ_{ab} near T_c in Fig. 2(b). As temperature lowers, R^H_{ab}

of the flux sample decreases from 96 K down to T_c, while R_{ab}^H of the CP sample does not decrease above 89 K, well below zero-field T_c.[24] This implies not only the overdoped nature, but also the absence of the spin gap above T_c in the CP sample.

Optical properties also indicate the overdoped nature. The out-of-plane optical conductivity of a highly oxygenated CP crystal showed anomalous excitation below T_c, which was attributed to the collective excitation of unpaired carriers.[25] A similar spectrum has been observed in overdoped $La_{2-x}Sr_xCuO_4$.[26] A Raman-active phonon gives evidence for the high doping level in a CP sample. The phonon energy related to the apical oxygen was observed at 517 cm^{-1} in a highly oxygenated CP sample [with $x(zz)x$ polarization],[27] and at 502 cm^{-1} in a highly oxygenated flux sample.[28] Since this energy monotonically increases with doping, the extremely high phonon energy implies that carriers are more doped in the CP sample than in the flux sample.

Next we will have a closer look at the out-of-plane transport. The out-of-plane resistivity is indicative of the overdoped nature. The magnitude of ρ_c in Fig. 1 (2 mΩ cm just above T_c) is smaller than a typical ρ_c in highly oxygenated flux samples (3–4 mΩ cm just above T_c).[7,29] It should be also noted that ρ_c in Fig. 1 decreases with decreasing temperature, whereas ρ_c of flux samples often shows a slight upturn around T_c.[30] These results indicate that ρ_c for the CP sample is more conductive than that for the optimally doped $YBa_2Cu_3O_y$. The high conductivity along the out-of-plane direction is also seen in the infrared optical spectra, where a Drude-like conductivity has been clearly observed.[25] We emphasize that, among HTSC, only our highly oxygenated CP samples show a clear Drude-like conductivity along the out-of-plane direction. In spite of the high conductivity, the anisotropy in the Hall coefficient and the thermopower still remains the same as in other HTSC. A positive S_c has been reported in $La_{2-x}Sr_xCuO_4$ (Ref. 5) and $Tl_2Ba_2CuO_6$,[31] and a negative R_c^H has been observed in $La_{2-x}Sr_xCuO_4$ (Ref. 32) and $Bi_2Sr_2CuO_6$.[33] We further note that the magnitudes and temperature dependence of S_c and R_c^H are also similar among various HTSC. This suggests that R_c^H and S_c are independent of the magnitude of ρ_c.

The positive R_{ab}^H and the negative R_c^H in Fig. 1 require us to abandon a simple parabolic band with the dispersion of $\epsilon(k)=\hbar^2k_x^2/2m_x+\hbar^2k_y^2/2m_y+\hbar^2k_z^2/2m_z$ (m_i is the effective mass along the i direction). In other words, R_{ab}^H is no longer equal to $(nec)^{-1}$ (n is the carrier density). Moreover the thermopower, which should have the same sign as R^H in the simple parabolic band picture, has the opposite sign. The calculated band based on the actual crystal structure, in contrast, explains their signs,[34] which seems to show the correctness of the band picture.

The band calculation, however, cannot reproduce the anisotropy between ρ_c and ρ_{ab}. Even though both ρ_{ab} and ρ_c show metallic behavior, only ρ_c seems to have a large residual resistivity, or equivalently, ρ_c/ρ_{ab} increases with decreasing temperature. We think that the temperature dependence of the resistivity is mainly dominated by scattering time (τ). This is most directly seen in the temperature dependence of the infrared conductivity spectra, where the Drude weight is nearly independent of temperature.[25,35] Another example is seen in ρ_{ab} and ρ_c for Zn-substituted HTSC;[29,36,37]

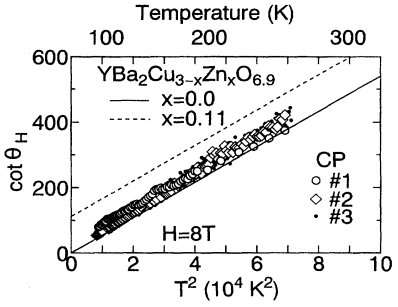

FIG. 3. The cotangent of Hall angle cot θ_H ($\equiv\rho_{ab}/HR_{ab}^H$) in 8 T plotted as a function of the square of temperature. Sample No. 1 is the same as in Figs. 1 and 2, and the data for two CP samples Nos. 2 and 3 are added for comparison. The solid and dotted lines correspond to cot θ_H for the flux samples of $YBa_2Cu_{3-x}Zn_xO_{6.9}$ crystal from Ref. 36. The solid (dotted) line is for the $x=0$ ($x=0.11$) sample.

the Zn substitution adds residual resistivity to both ρ_{ab} and ρ_c and leaves $d\rho/dT$ unchanged. Thus the different temperature dependence between ρ_{ab} and ρ_c reduces to *the different temperature dependence of* τ. At first glance one may think it natural that τ can be anisotropic in an anisotropic material, but it is quite difficult to derive such a large anisotropy from a 3D Fermi liquid. Later we will see the difficulty quantitatively, and show that the residual resistivity in ρ_c does not come from the impurity scattering.

IV. DISCUSSION

A. Slightly overdoped $YBa_2Cu_3O_y$

As already mentioned, $YBa_2Cu_3O_y$ crystals by the crystal-pulling technique are slightly overdoped when highly oxygenated. The facts suggesting the overdoped nature are (1) the small magnitude of ρ_{ab} and ρ_c, (2) the superlinear dependence of ρ_{ab}, (3) the small contribution of superconducting fluctuation near T_c, (4) the small magnitude of R_{ab}^H, and (5) the negative S_{ab}.

As shown in Fig. 2(b), the CP sample has a lower T_c than the flux sample. We will examine whether the lower T_c is due to overdoping or not. There are many causes of lowering T_c of HTSC, e.g., overdoping, underdoping, pair breaking by Pr,[38] disorder,[39] and inhomogeneity.[40] In the present sample, we regard overdoping and disorder as candidates suppressing T_c (we ruled out inhomogeneity because of the reasonably sharp transition). For the evaluation of the amount of disorder, the cotangent of Hall angle (cot $\theta_H\equiv\rho_{ab}/HR_{ab}^H$) is quite useful.[36] The relation of cot $\theta_H=AT^2+B$ is widely observed in HTSC,[41] which has been regarded as evidence for the anomalous transport in HTSC. Here we note that the parameters A and B give valuable information on sample quality; the former is related to doping level, and the latter is proportional to the amount of impurity. Figure 3 shows cot θ_H calculated from the measured ρ_{ab} and R_{ab}^H (as denoted No. 1), together with the data for other two CP samples (Nos. 2 and 3). Figure 3 clearly indicates the relation of AT^2+B with a small B. Within experimental errors we estimated B/H to be at most 5 T^{-1}, which corresponds to the disorder induced by the substitution of less than 1 at. % Zn for Cu. Since the

sample (No. 1) with the largest B (=40) and the sample (No. 2) with the smallest B (=0) have the same T_c, the low T_c is likely to come from overdoping rather than disorder.[42]

Here we propose a possible origin for overdoping, which is related to the growth mechanism in the crystal-pulling technique.[14] A CP crystal is grown in the Ba-Cu-O liquid where Y cations are transported by thermal convection from the Y_2BaCuO_5 precipitate located at the bottom of the crucible. In short, the CP crystal is grown in the solution containing less Y than the stoichiometry, which may cause some Y^{3+} deficiencies. In fact, inductively coupled-plasma mass spectroscopy (ICP) showed Y:Ba:Cu~0.95:1.98:3 in CP samples (within the errors of 3%), that is, Y was deficient. In addition, the ICP analysis detected a small amount of Ca ($\leq$5 at. %), which possibly came from the BaO source of 99.9% purity. Accordingly it is most probable that Ca^{2+} cations work as acceptors by substituting for Y^{3+}. We also note that the oxygen content of the CP sample was estimated to be 6.92 ± 0.02 with iodometric titration, which is nearly the same value as in highly oxygenated flux samples.[43] It is well established that an overdoped $YBa_2Cu_3O_y$ can be made by substituting Ca for Y,[44] which supports our explanation. Very recently the c-axis-oriented $R_{1-x}Ca_xBa_2Cu_3O_y$ films have been grown, where R represents rare-earth cations such as Y (Ref. 45) and Sm.[46] The transport measurements of the films have shown a small ρ_{ab}, a small R^H_{ab}, and a negative S_{ab}. The observed R^H_{ab} is quantitatively consistent with our data, but ρ_{ab} of these films is higher, suggesting that the films contain more disorder. It should be emphasized that there are only a few reports on single crystals of overdoped $YBa_2Cu_3O_y$,[47] and no reports on its out-of-plane transport.

At the end of this subsection, we briefly mention the contribution of the CuO chain. As is widely accepted, the CuO chain is conductive in highly oxygenated samples, and hence resistivity,[7,48] thermopower,[49] and optical reflectivity[35,50] are anisotropic between the a and b directions. In particular, the temperature dependence of the CuO-chain resistivity is proportional to $\alpha T^2+\rho_0$, different from the T-linear resistivity.[51] Then one would doubt whether the temperature dependence of ρ_{ab} in the twinned CP sample is due to the CuO chain, not due to overdoping. Suppose that the observed ρ_{ab} is mainly due to the CuO chains; this implies that the resistivity of the CuO_2 plane is much smaller than that of the CuO chain. Such a small ρ_a naturally suggests the overdoped nature. We also note that R^H_{ab} will not be influenced by the CuO chain because of Onsager's relation of $R^H_{abc}=-R^H_{bac}$.[52] Thus we expect that R^H_{ab} in the twin-free CP sample will have the same value as the present data that suggest the overdoped nature.

B. Comparison with band calculation

In the studies of HTSC, ρ_c/ρ_{ab} has been used as a measure of anisotropy. In the same doping level, $YBa_2Cu_3O_y$ has the smallest ρ_c/ρ_{ab} among HTSC. As doping proceeds, ρ_c/ρ_{ab} monotonically decreases. Consequently the overdoped $YBa_2Cu_3O_y$ has the least anisotropy between ρ_c and ρ_{ab} among all high-T_c cuprates, being expected to be most three dimensional. We then expect that a band theory could most satisfactorily explain the data for the overdoped $YBa_2Cu_3O_y$. In this subsection, we examine the validity of band calculation.

TABLE I. Various physical parameters at room temperature. ρ_i, R^H_i, S_i, τ_i, ω^p_i, and l_i are resistivities, Hall coefficients, thermopowers, scattering times, plasma frequencies, and mean free paths along the i directions, respectively. The experimental data for transport properties are the same as in Fig. 1, and those for optical properties are taken from Ref. 25. The calculated data based on the band theory are taken from Ref. 34. Note that the quantities calculated along the a and b directions are properly averaged, as shown at the bottom of the table.

Parameters	Experiment	Calculation	Units
ρ_{ab}	125	26.5[a]	$\mu\Omega$ cm
ρ_c	4250	260	$\mu\Omega$ cm
R^H_{ab}	4	2	10^{-4} cm^3/C
R^H_c	−3.5	−7.5[b]	10^{-4} cm^3/C
S_{ab}	−2	−15[c]	μV/K
S_c	25	22	μV/K
$\hbar/\tau_a$	0.026[d]	0.052[e]	eV
$\hbar/\tau_c$	0.35[f]	0.052[e]	eV
ω^p_a	1.9	2.9	eV
ω^p_c	0.71	1.1	eV
l_{ab}	75[g]	85	Å
l_c	5.8[g]	26	Å

[a]Averaged as $(\rho_a+\rho_b)/2$.
[b]Averaged $(R^H_{bca}+R^H_{cab})/2$.
[c]Averaged $(S_a+S_b)/2$.
[d]k_BT (T=300 K) is regarded as $\hbar/\tau_a$.
[e]$2\pi\lambda k_BT$ with $\lambda=0.32$ is regarded as $\hbar/\tau_a$.
[f]The data at 50 cm^{-1} in Fig. 4 of Ref. 25 is used.
[g]Calculated from $v^F_i\tau_i$, where v^F_i is evaluated from $(\rho_i)^{-1}=N(0)(v^F_i)^2\tau_i/3$.

There are several calculations about transport parameters in the studies of the band structure.[34,53–55] Allen, Pickett, and Krakauer have calculated various transport parameters based on the local-density-functional theory.[34] We compare our results with their calculation, as listed in Table I. Since we used twinned samples, the calculated quantities are properly averaged between the a and b axes. It is noteworthy that the signs of the calculated R^H and S are in good agreement with the observed results. This means that the signs of R^H and S can be understood as the results from the complicated shape of the Fermi surface, and that an effective-mass model with a parabolic dispersion is too simple to use. In addition, the magnitudes of the calculated R^H_c and S_c reproduce the experimental data fairly well. We plot the experimental data and the calculation for R^H and S in Fig. 4, where the calculation qualitatively explains the out-of-plane data. The calculation, however, does not successfully fit the observed R^H_{ab} and S_{ab}.

A serious disagreement between the band theory and the experiments is seen in the resistivities. The calculated ρ_c/ρ_{ab}(~10) is independent of temperature, while the observed ρ_c/ρ_{ab} is much larger and dependent on temperature (35 at room temperature and 70–80 just above T_c). We think that this disagreement arises from τ. Within the framework of band theory, τ is treated as anisotropic in $\mathbf{k}$ space in the lowest-order approximation, where the anisotropy in resistivity is attributed to the anisotropic Drude weights, being independent of temperature. The experimental results, how-

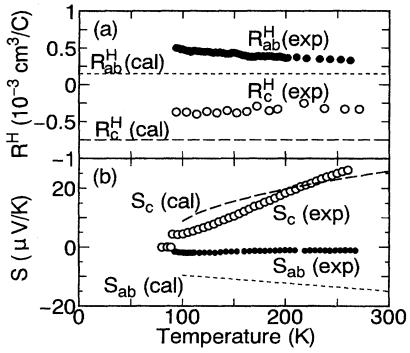

FIG. 4. The comparison between experiments (exp) and band calculation (cal). The experimental data are the same as shown in Fig. 1, and the results of the band calculation are taken from Ref. 34. (a) Hall coefficient (R^H). Since the calculation neglects the contribution of the scattering time to R^H, the calculated Hall coefficient is drawn as temperature independent. (b) Thermopower (S). Note that the calculated S strongly depends on the conductivitylike function $\sigma(\varepsilon)$. The data correspond to $\sigma(\varepsilon)\propto\{\omega^p(\varepsilon)\}^2$.

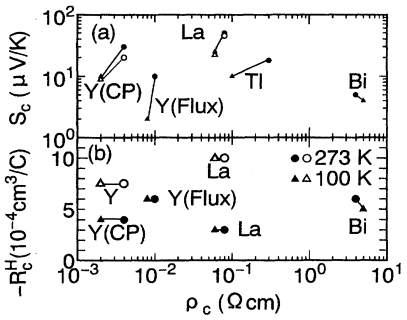

FIG. 5. The out-of-plane transport for various overdoped high-T_c cuprates plotted as a function of the out-of-plane resistivity (ρ_c). The closed (open) symbols are experimental (calculated) data. The circles and triangles are the 300- and 100-K data, respectively. Note that the calculated data are plotted at the same ρ_c as observed in the experiment. (a) The out-of-plane thermopower (S_c). The labels in the figure are as follows. Y(CP): the present result; Y(Flux): a flux sample of $YBa_2Cu_3O_{6.9}$ taken from Ref. 59; La: $La_{1.8}Sr_{0.2}CuO_4$ from Ref. 5; Tl: $TlBa_2CuO_6$ from Ref. 31; Bi: $Bi_2Sr_2CuO_6$ from Ref. 60. The results of band calculation are taken from Ref. 34. (b) The out-of-plane Hall coefficient (R^H_{ab}). The labels in the figure are as follows. Y(CP): the present result; Y(Flux): a flux sample of $YBa_2Cu_3O_{6.9}$ taken from Ref. 61; La: $La_{1.7}Sr_{0.3}CuO_4$ from Ref. 32; Bi: $Bi_2Sr_2CaCu_2O_8$ from Ref. 33. The calculation for $YBa_2Cu_3O_7$ and $La_{1.7}Sr_{0.3}CuO_4$ are taken from Refs. 34 and 54, respectively.

ever, strongly suggest anisotropic τ's.[56] Actually the optical study of $YBa_2Cu_3O_y$ has revealed that the in-plane scattering time (τ_a),[35] is more than 10 times longer than the out-of-plane one (τ_c).[25] A recent microwave study for 90-K phase $YBa_2Cu_3O_y$ has also shown that τ_{ab} is 10 times longer than τ_c in the normal state.[57] As a result, the out-of-plane mean free path ($l_c\sim5.8$ Å) at room temperature is shorter than the c-axis lattice constant (~12 Å),[58] while the in-plane ($l_{ab}\sim75$ Å) is much longer than the a- or b-axis constant (~4 Å). Thus the out-of-plane transport is in the dirty limit, and the in-plane transport is in the clean limit. Such a large anisotropy in τ or l has never been observed in conventional metals, and we regard this as clear evidence for the breakdown of the band calculation in $YBa_2Cu_3O_y$.

Assuming that only τ is anomalous, we can easily understand why different kinds of slightly overdoped HTSC have similar R^H_c and S_c. Hall coefficient and thermopower are, in principle, less affected by τ. The Hall coefficient in metals is weakly dependent on temperature, because the contribution of τ is cancelled in the lowest-order approximation. The temperature dependence of thermopower in metals is mainly determined by the ratio of k_BT to the chemical potential, and the correction by τ is usually small. R^H_c and S_c (Refs. 5, 32, 33, 35, 53, and 59–61) for various HTSC are plotted as a function of ρ_c in Fig. 5, where the experimental data and calculated values are plotted with the full and open symbols, respectively. (The calculated ρ_c is much smaller than the observed ρ_c, but we plot R^H_c and S_c at the same ρ_c as observed in the experiment.) Figure 5 shows that R^H_c and S_c have no clear relation to ρ_c. Furthermore the temperature dependence of R^H_c and S_c qualitatively agrees with the band calculation.

As long as we assume that the band calculation can explain R^H_c and S_c, we naturally encounter the question why it cannot explain R^H_{ab} and S_{ab} well. This leads us to the conclusion that the in-plane transport is completely different from the out-of-plane transport. In other words, the electronic states of $YBa_2Cu_3O_y$ are essentially 2D, and the band calculation is applicable only in a limited way.

Even non-Fermi-liquid theories are, more or less, based on the results of the band calculation. In the RVB theory, for example, the band dispersion survives as the dispersion of spinons. Tanamoto, Kohno, and Fukuyama have estimated the transfer energies from the band calculation to use them in their mean-field treatment of the RVB state.[62] The RVB theory predicts that the carriers doped in the CuO_2 plane dissociate into spinons and holons, which are responsible for the spin and charge degrees of freedom, respectively. Then the in-plane transport is explained in terms of these two, which results in the non-Fermi-liquid behavior. In the out-of-plane transport, however, a holon accompanies a spinon to hop as one physical hole from one CuO_2 plane to another. Consequently the out-of-plane transport is dominated by the physical hole, and seems to be Fermi-liquid-like. This is a similar situation seen in photoemission spectra, which give a large Fermi surface consistent with the band calculation.[63] These features of the RVB theory are qualitatively consistent with the present experiment.

C. Out-of-plane scattering time

From the critical comparison between the experiments and the band calculation, we have pointed out the anomaly in ρ_c and τ_c. Here we will consider more quantitatively what it means.

First of all, we will demonstrate that a conventional Boltzmann theory, which is valid in a 3D Fermi liquid, cannot explain the anomalously large anisotropy in τ. Within the framework of the Boltzmann transport, ρ_{ab} and ρ_c are calculated as

$$1/\rho_{ab}\sim\sigma_{ab}=\frac{2e^2}{(2\pi)^3}\int d^3k\{v_{ab}(\mathbf{k})\}^2\tau(\mathbf{k})\frac{df_0}{d\varepsilon}, \quad (1)$$

$$1/\rho_c\sim\sigma_c=\frac{2e^2}{(2\pi)^3}\int d^3k\{v_c(\mathbf{k})\}^2\tau(\mathbf{k})\frac{df_0}{d\varepsilon}, \quad (2)$$

where $v_i(\mathbf{k})\equiv\hbar^{-1}\partial\varepsilon(\mathbf{k})/\partial k_i$ is the Fermi velocity along the i direction, and f_0 is the Fermi distribution function. In a conventional metal, $df_0/d\varepsilon$ is nearly identical to the δ function at low temperatures. Equations (1) and (2) show that $\tau(\mathbf{k})$ is averaged with the weight of $\{v_i(\mathbf{k})\}^2$ in the Brillouin zone, and thus the anisotropy between ρ_{ab} and ρ_c is mainly determined by $\{v_i(\mathbf{k})\}^2$. For example, assuming the dispersion of $\varepsilon(\mathbf{k})=\hbar^2(k_x^2+k_y^2)/2m_{ab}+\hbar^2k_z^2/2m_c$, we get ρ_c/ρ_{ab} as

$$\frac{\rho_c}{\rho_{ab}}\sim\frac{m_c}{m_{ab}}\frac{\int\tau(\theta,\varphi)\sin^3\theta\cos^2\varphi d\theta d\varphi}{\int\tau(\theta,\varphi)\cos^2\theta\sin\theta d\theta d\varphi}\sim\frac{m_c}{m_{ab}}\frac{C_0\bar{\tau}}{C_0'\bar{\tau}}. \quad (3)$$

where $\bar{\tau}$ is the average of $\tau(\mathbf{k})$ at $k=k_F$ and C_0 (C_0') is a constant of the order of unity. Consequently Eq. (3) reduces to the relation of $\rho_c/\rho_{ab}\sim m_c/m_{ab}$. Although Eq. (3) is modified to reflect their band structures in general, ρ_c/ρ_{ab} roughly scales with Ω_{ab}/Ω_c, (Ω_i is the Drude weight along the i direction), and τ_c is of the order of τ_{ab}. Thus we conclude that an anisotropic 3D Boltzmann theory is seriously incompatible with the observed values of $\tau_c/\tau_{ab}\sim5$–10 in the slightly overdoped $YBa_2Cu_3O_y$ crystal. This indicates that its electronic states cannot be treated as an anisotropic 3D Fermi liquid, even though the Drude weight is less anisotropic than that for Sr_2RuO_4 [$(\omega_{ab}^p/\omega_c^p)^2\sim10^3$] whose electronic states can be treated as an anisotropic 3D Fermi liquid.[64]

Another notable discrepancy from the Fermi liquid picture is that ρ_c seems to have a large residual resistivity. In the Boltzmann theory, the residual resistivity arises from impurity scattering, which is isotropic in $\mathbf{k}$ space in the lowest-order approximation. Then we can put τ out of the integrals in Eqs. (1) and (2), and get $\rho_c/\rho_{ab}=(\omega_{ab}^p/\omega_c^p)^2$ at $T=0$. If the residual ρ_c is attributed to impurity scattering, the fact that ρ_c at 0 K is about 1 mΩ cm requires ρ_{ab} at 0 K to be about 140 $\mu\Omega$ cm, which is larger than the observed ρ_{ab} at all temperatures. Thus *the residual resistivity in ρ_c is not attributable to impurity scattering*. The same consideration is valid even in exotic theories. For instance, Monthoux and Pines have calculated a quite small residual resistivity in ρ_c within the theory for 2D metal with antiferromagnetic fluctuation.[65] Nagaosa has proposed $\rho_c\propto\sqrt{T}$, which indicates no residual resistivity, on the basis of the gauge-field theory in the t-J model.[66] Instead of the residual resistivity, Anderson and Zou have suggested $\rho_c=C_1T+C_2/T$ by combining the RVB theory with incoherent interplane hopping.[67] Varma and Littlewood have proposed the same expression of ρ_c from the viewpoint of the marginal Fermi liquid (MFL).[68]

Discarding the 3D Boltzmann transport, we will examine whether a highly anisotropic (effectively 2D) Fermi-liquid picture is applicable or not. In a highly anisotropic Fermi liquid, where an interplane hopping parameter (t_c) is much smaller than an intraplane one (t_{ab}), the out-of-plane transport can be incoherent under certain conditions.[69] When $t_c\hbar/\tau_{ab}\ll1$, Kumar and Jayannavar[70] have calculated the anisotropy in the resistivity as

$$\frac{\rho_c}{\rho_{ab}}=4na^2c\left(\frac{a}{c}\right)^2\left(\frac{\omega_a^p}{\omega_c^p}\right)^4, \quad (4)$$

where a and c are the lattice constants of the a and c axes, respectively. By substituting $(\omega_a^p/\omega_c^p)^2=7$, $a=3.8$ Å, $c=11.7$ Å, and $na^2c=0.15$–0.2 in Eq. (4), we estimate $\rho_c/\rho_{ab}\sim3$–4, which is much smaller than the observed ρ_c/ρ_{ab}. Rojo and Levin[71] have considered the interplane disorder that scatters a carrier in one CuO_2 plane to the neighboring plane, and have derived the anisotropic conductivity as

$$\sigma_c=A_0T+A_1+\frac{1}{A_2T+A_3} \quad (5)$$

$$\sigma_{ab}=\frac{1}{B_1T+B_2}, \quad (6)$$

where A_i and B_i are constants. Note that Eq. (5) gives $\rho_c\to(A_0T+A_1)^{-1}$ as $T\to\infty$. This means that ρ_c always *decreases* with increasing T at high temperatures. Contrary to this, the experimental results indicate that $d\rho_c/dT$ is positive at high temperatures in various HTSC, even if it might be negative near T_c. We also note that the observed finite R_c^H is difficult to explain within these theories. If the electronic states were highly anisotropic, the Fermi surface would be cylindrical to give $R_c^H=0$.

The anomalously short τ_c can be also explained by the RVB theory. After a holon with a phase θ and a spinon with a phase $-\theta$ hop together as one physical hole from one CuO_2 plane to another, it dissociates again into a holon with a phase θ' and a spinon with a phase $-\theta'$. Even though this process is a coherent hopping for the physical hole, it loses the memory of the phases of the holon and the spinon. As a result, the out-of-plane dephasing length for holons are roughly equal to the c-axis lattice constant, that is, the out-of-plane transport is in the dirty limit. A similar situation would occur in exotic theories other than the RVB theory. If elementary excitations in the CuO_2 plane (e.g., polarons) are essentially 2D, the interplane hopping may lose the memory of their phase. From the present study, therefore, we cannot specify which non-Fermi-liquid theory is correct; what we can conclude is that the electronic states of HTSC are essentially 2D, which are characterized by the anomalously short τ_c, not by the magnitude of ρ_c or t_c. Further study, both experimentally and theoretically, is necessary to evaluate τ_c more quantitatively.

Finally we will discuss the temperature dependence of ρ_c. The expressions for ρ_c proposed so far are listed as follows:

$$\rho_c=C_1T+C_2/T, \quad (7)$$

$$\rho_c=C_1\sqrt{T}, \quad (8)$$

$$\rho_c=C_1T+C_2+C_3\exp(C_4/T), \quad (9)$$

where C_i is a constant. Equation (7) is for RVB (Ref. 67) and MFL (Ref. 68) pictures, while Eq. (8) is calculated using the gauge-field theory of the uniform RVB state.[66] Recently Eq. (9) has been suggested by Ong and co-workers,[3,8] involving a true gap [C_4 in Eq. (8)]. Since T in Eqs. (7)–(9) arises from the T-linear ρ_{ab}, we generalize this term as

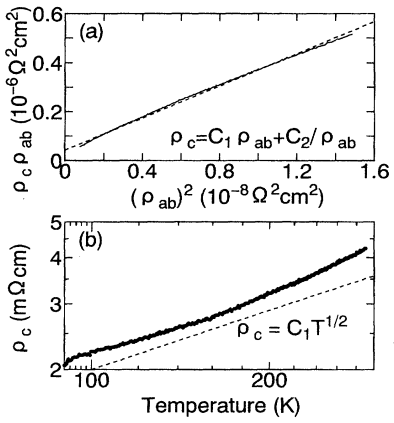

FIG. 6. (a) $\rho_c\rho_{ab}$ plotted as a function of $\{\rho_{ab}\}^2$. The observed linear relation indicates the relation of $\rho_c=C_1\rho_{ab}+C_2/\rho_{ab}$. The parameters C_1 and C_2 are estimated to be 32.8 and 4.31×10^{-8} Ω^2 cm^2, respectively. (b) ρ_c plotted as a function of ρ_{ab} in log-log scale.

$$\rho_c=C_1\rho_{ab}+C_2/T, \tag{7'}$$

$$\rho_c=C_1\rho_{ab}+C_2/\rho_{ab}, \tag{7''}$$

$$\rho_c=C_1\{\rho_{ab}\}^p \quad (1/2\leq p\leq 2), \tag{8'}$$

$$\rho_c=C_1\rho_{ab}+C_3\exp(C_4/T). \tag{9'}$$

This generalization enables us to fit the present data, because ρ_{ab} is no longer T linear in the slightly overdoped $YBa_2Cu_3O_y$.

We have examined all the expressions, and have found that Eqs. (7), (7′), and (7″) fit the observed ρ_c better than Eqs. (8) and (8′).[72] In Fig. 6(a), $\rho_c\rho_{ab}$ is plotted as a function of $\{\rho_{ab}\}^2$, where Eq. (7″) requires a linear relation. The expected linear relation is clearly seen, from which the parameters are obtained as $C_1=32.8$ and $C_2=4.31\times10^{-8}$ Ω^2 cm^2. On the other hand, Eq. (8′) cannot fit the data well, as shown in Fig. 6(b), where ρ_c vs ρ_{ab} is plotted in log-log scale. We have found that the data are well fitted by Eqs. (9) and (9′), but we cannot determine the fitting parameters uniquely. Moreover Yan, Harris, and Ong have speculated that the true gap in Eq. (9) is characteristic of bilayer materials,[8] but we think that there are no remarkable differences in temperature dependence of ρ_c between bilayer ($YBa_2Cu_3O_y$) and single-layer ($La_{2-x}Sr_xCuO_4$) materials. Thus we propose that Eq. (7), (7′), or (7″) is a promising candidate for the expression of ρ_c.

Equation (7) tells us that τ_{ab} also dominates ρ_c, and that ρ_c is essentially proportional to ρ_{ab} with the coefficient of C_1. The spurious residual resistivity in ρ_c comes from the additional anomalous contribution characterized by C_2. This contribution is related to disorder in the MFL picture, while it is associated with incoherent hopping in the RVB theory. For further discussion it is necessary to study ρ_c for the Zn-substituted samples, which will give valuable information on the effect of disorder. Leaving the anomalous term of C_2, we can find the parameter C_1 to be anomalously larger ($\sim$33) than expected from $(\omega_a^p/\omega_c^p)^2=7$. This suggests that τ_c or t_c is reduced by a factor of 5 through the interplane hopping. In either case, the dephasing length is as short as the c-axis length to make the interplane hopping nearly incoherent. The short dephasing length along the out-of-plane direction is presumably the evidence for carrier confinement in the CuO_2 plane.

V. SUMMARY

To study anisotropic transport of high-T_c cuprates, we prepared two kinds of $YBa_2Cu_3O_y$ crystals, by a crystal-pulling technique and by a conventional flux technique, and measured the in-plane and out-of-plane transport parameters. The in-plane transport has revealed that the highly oxygenated samples by the crystal-pulling technique are slightly overdoped with T_c of 90 K that is slightly lower than a typical T_c of 92 K for crystals grown from flux. The evidence for overdoping is the low resistivity (30 $\mu\Omega$ cm along the in-plane direction and 2 mΩ cm along the out-of-plane direction just above T_c), the small in-plane Hall coefficient (5×10^{-4} cm^3/C just above T_c), the negative in-plane thermopower, and the temperature dependence of the resistivity and the Hall coefficient.

We have examined to what extent a band theory fits the present data. Since our samples are (slightly) overdoped $YBa_2Cu_3O_y$, the out-of-plane resistivity is lowest among high-T_c cuprates. Accordingly the electronic states of our samples are expected to be most three dimensional. Nevertheless we have shown that their scattering time along the out-of plane direction is still too short for a band theory. This means that the overdoped $YBa_2Cu_3O_y$ is still two dimensional, and we have concluded that the two-dimensional nature of high-T_c cuprates is characterized not by the magnitude of the out-of-plane resistivity, but by the anisotropic scattering times.

The out-of-plane scattering time is so short that the out-of-plane transport is in the dirty limit, while the in-plane transport is in the clean limit. Since the evaluated mean free path is shorter than the c-axis lattice constant, the short out-of-plane scattering time might be related to the carrier confinement in the CuO_2 plane. The present study has shown that the anomaly in the scattering time is incompatible with a conventional Boltzmann transport, or the 2D Fermi-liquid approach involving incoherent interplane hopping. The residual resistivity in ρ_c cannot be ascribed to impurity scattering, and instead the empirical relation of $\rho_c=C_1\rho_{ab}+C_2/\rho_{ab}$ fits the data fairly well.

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