Intrinsic Hall response of the CuO$_2$ planes in a chain-plane composite system of YBa$_2$Cu$_3$O$_y$

Kouji Segawa and Yoichi Ando

Central Research Institute of Electric Power Industry, Komae, Tokyo 201-8511, Japan

(Received 4 August 2003; revised manuscript received 14 November 2003; published 24 March 2004)

The Hall coefficient is measured in YBa$_2$Cu$_3$O$_y$ untwinned single crystals for a wide range of doping. We show that the Hall conductivity and the Hall angle of the CuO$_2$ *planes* in YBa$_2$Cu$_3$O$_y$ can be extracted from measurable transport properties regardless of the conduction of the Cu-O chains and the in-plane anisotropy of the CuO$_2$ planes. The present analysis allows us to discuss the genuine Hall effect in the CuO$_2$ planes alone in YBa$_2$Cu$_3$O$_y$ without any complications due to the Cu-O chains.

I. INTRODUCTION

The origin of the peculiar normal-state properties of the high-T_c superconductors is not fully understood. The strongly temperature-dependent Hall coefficient (R_H) is one of the best-known peculiar features in the normal state of high-T_c cuprates.[1] While R_H shows a complicated temperature dependence, the cotangent of the Hall angle (cot Θ_H $\equiv E_x/E_y = \rho_{xx}/\rho_{yx}$) was reported to show a simple T^2 dependence in YBa$_2$Cu$_3$O$_y$ (YBCO) (Ref. 2); it is widely believed that the peculiar behavior of R_H comes from an anomalous coexistence of the T^2 law in cot Θ_H and the pronounced T linear resistivity. This fact suggests that two kinds of scattering times dominate charge transport in high-T_c cuprates.[3–12]

However, the YBCO system has the Cu-O chains which are likely to show one-dimensional (1D) electronic conduction,[13] which is believed to cause the in-plane resistivity anisotropy in the highly doped region.[13–15] On the other hand, in the underdoped region, the 1D conduction of the Cu-O chains is expected to diminish quickly with reducing oxygen content, because oxygens are removed from the Cu-O chains, creating vacancies in the chains, and 1D systems are known to be very sensitive to disorder.[16] However, we have demonstrated that the in-plane anisotropy does not disappear in the underdoped region, which indicates that the CuO$_2$ plane itself is anisotropic.[15] In such a material with in-plane anisotropy as well as the chains, resistivity and cot Θ_H measured in twinned crystals are mixtures of the intrinsic transport properties. The original report of the T^2 law in cot Θ_H (Ref. 2) was based on the result in twinned single crystals, and therefore the conductive Cu-O chains may have affected the temperature dependences of cot Θ_H and resistivity. Detwinning the crystals allows us to observe the in-plane anisotropy in resistivity; however, even the results in untwinned single crystals[17] are not free from contribution of the Cu-O chains, because the Cu-O chains can indirectly modify the Hall coefficient, as described later. Therefore, measurement on untwinned crystals and estimating the contribution from the Cu-O chains are necessary to obtain intrinsic transport properties in YBCO in the whole doping region.

In this paper, we report the way to deduce the genuine Hall response of the CuO$_2$ *planes* in YBa$_2$Cu$_3$O$_y$ untwinned single crystals. We show that the Hall conductivity (σ_{xy}) and the cotangent of the Hall angle (cot Θ_H) of the CuO$_2$ *planes* can be obtained from measurable transport properties such as the $a(b)$-axis in-plane resistivity $\rho_{a(b)}$ and the Hall coefficient R_H. The result shows that the electronic state of the CuO$_2$ planes is responsible for the observed "60 K anomalies" in YBCO, such as a decrease in the Hall coefficient just above T_c and the enhancement of the Hall conductivity and the Hall mobility in samples with $y \sim 6.65$.

II. EXPERIMENTS

The YBCO single crystals are grown in Y$_2$O$_3$ crucibles by a conventional flux method.[18] A wide range of oxygen contents are achieved by annealing under various conditions as shown in Table I and quenching from the set temperatures of the annealing. Slightly overdoped ($y \simeq 7.00$) and optimally doped ($y \simeq 6.95$) crystals are obtained by annealing in oxygen atmosphere for a long time. Samples with $y = 6.70$–6.85 are obtained by annealing in air for 12–48 h at temperatures which increase with decreasing oxygen contents. For more underdoped samples, annealing in air is not applicable because it is difficult to quench samples quickly enough for preventing absorption of oxygen onto the surface of the crystals at very high temperatures. Instead, in order to obtain oxygen contents of $y \lesssim 6.65$ we seal crystals in a quartz tube together with polycrystalline powders with controlled oxygen contents. The amount of the buffer powder should be as large as possible for obtaining homogeneous oxygen distribution; however, if the amount of powder is too large (e.g., more than 100 mg in ~ 7 cc sealed quartz tube) we observe superconducting transition of minor phases at around 60 K. Therefore, we use ~ 50 mg of powder for each annealing, keeping the amount of single crystals less than 1% of powder. Only samples with $y = 6.30$ are annealed by using special furnace in which we can control the oxygen partial pressure. We keep samples at a room temperature for at least a week after any heat treatments, because the room-temperature annealing effect[19] is observed in the time scale of a few days. All crystals are detwinned at $\leqslant 210\,°C$ in flowing nitrogen *after* the annealing, because annealing at high temperatures above $\sim 600\,°C$ destroys an untwinned state. We confirmed that the experimental results are not affected by the order of annealing and detwinning for samples with $y = 6.80$ and 6.85 (Ref. 20), which can be detwinned either before or after the annealing. We decrease the detwin-

TABLE I. Annealing conditions for our YBCO crystals. T_{quench} is the temperature at which polycrystalline YBCO powders are quenched in air, and the polycrystalline powders are sealed in a quartz tube together with YBCO single crystals for tuning the oxygen contents.

y	T_{anneal} (°C)	Keeping time	Atmosphere	T_{quench} (°C) of powder	T_c (K)
7.00	400	$\geqslant$10 days	$\sim$2 atm O_2		91
6.95	485	$\geqslant$7 days	1 atm O_2		93
6.85	535	48 h	Air		83
6.80	565	24 h	Air		69
6.75	600	12 h	Air		62
6.70	625	12 h	Air		60
6.65	550	24 h	Sealed	620	58
6.60	600	12 h	Sealed	640	57
6.55	600	12 h	Sealed	660	55
6.50	600	12 h	Sealed	675	35
6.45	600	12 h	Sealed	700	20
6.35	700	6 h	Sealed	800	
6.30	550	36 h	3.5×10^{-4} atm O_2		

ning temperature with decreasing oxygen contents; for example, the detwinning temperature for $y = 7.00$ samples is $\sim$210 °C, and that for $y = 6.30$ samples is $\sim$120 °C under a uniaxial pressure of $\sim$0.1 GPa. The oxygen content y is determined by iodometric titration[21] on powders which are annealed with the crystals, because the volume of the crystals themselves is too small for the titration. The error in y is $\pm$0.02 (Ref. 18).

The Hall coefficient data are taken by sweeping the magnetic field (along the c axis) to both plus and minus polarities up to 10–14 T at fixed temperatures. For each composition, the R_H data are presented only for those temperatures at which the Hall voltage is perfectly proportional to the magnetic field.[22] Figure 1 shows the field dependences of the Hall resistivity for an optimally doped sample at various

temperatures. The origin for each temperature is shifted along y axis by 0.5 $\mu\Omega$ cm for 93–100 K and 0.25 $\mu\Omega$ cm for 100–300 K for clarity. We cannot extract R_H from the data for 93 and 95 K, because the field dependences deviate from the H-linear behavior by superconducting fluctuation.

III. EXPERIMENTAL RESULTS

A. In-plane anisotropy of the Hall coefficient

According to the Onsager's reciprocal relation[23] $[\rho_{yx}(H) = \rho_{xy}(-H)]$, when the magnetic field is applied along the c axis, R_H measured in a sample with the current along the a axis (ρ_a sample) is the same as that with the current along the b axis (ρ_b sample) unless time-reversal symmetry is broken. However, YBCO is a complicated composite system containing 2D CuO_2 planes and 1D Cu-O chains and thus it is not self-evident whether the Onsager relation is satisfied in YBCO. Harris *et al.* mentioned that the Onsager relation was accurately satisfied,[24] but no actual data were shown in their paper. Figure 2 shows the temperature dependences of R_H^a and R_H^b in an optimally doped sample, which were measured with the current along the a axis and the b axis; it should be

FIG. 1. Magnetic-field dependence of ρ_{yx} for a YBCO crystal with $y = 6.95$ (optimum doping) at various temperatures.
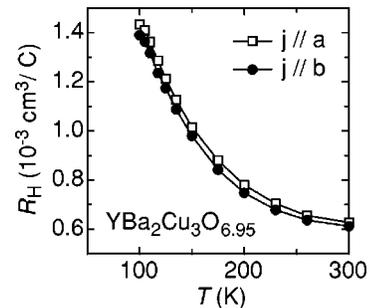
FIG. 2. $R_H(T)$ for two measurements on the same crystal at $y = 6.95$ (optimum doping), with $j\|a$ (open squares) and $j\|b$ (solid circles).
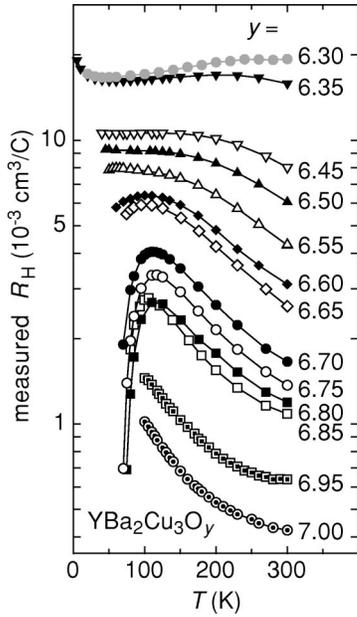
FIG. 3. T dependences of R_H^{meas} in untwinned YBCO crystals where the magnetic field is along the c axis.

FIG. 4. T dependences of V/NeR_H^{meas} in untwinned YBCO crystals with $y = 6.30$–6.55.

noted that these data were measured for the same sample with the same electrodes by re-detwinning the crystal without changing the oxygen content. Therefore, we have eliminated the uncertainty in the estimation of the sizes of the crystal and electrodes from comparison. The two curves shown in Fig. 2 do not coincide but are very close to each other. R_H^b is slightly (at most $\sim 4\%$) smaller than R_H^a, but this difference is most likely due to an extrinsic effect of the Hall measurement on a finite-sized sample combined with the resistivity anisotropy.[25] Therefore, we can say that the Onsager relation indeed holds within an error of $\sim 5\%$.

B. Measured Hall coefficient

Figure 3 shows the temperature dependences of the measured Hall coefficient (R_H^{meas}) for samples with various oxygen contents in a semilog plot. In general, the magnitude of R_H^{meas} increases with decreasing oxygen content. At high temperatures for most samples R_H^{meas} increases with decreasing temperature, while at low temperatures various behaviors are observed. For slightly underdoped samples with $y = 6.60$–6.85, R_H^{meas} shows a decrease at low temperatures below ~ 120 K unlike highly doped samples with $y = 6.95$–7.00. The observed decrease appears to be the strongest in samples with $y = 6.75$–6.80. These results are roughly consistent with the previously published data for twinned crystals with $y \geq 6.45$ (Ref. 26). For less doped samples the low-temperature decrease becomes weaker and eventually almost constant R_H^{meas} is observed at low temperatures for samples with $y = 6.45$–6.55. In the lightly doped nonsuperconducting samples with $y = 6.30$ and 6.35, R_H^{meas} increases with decreasing temperature at low temperatures, which is due to the charge localization observed in the in-plane resistivity.[15]
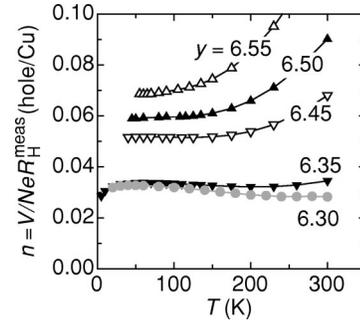
In the lightly doped region, nearly constant R_H^{meas} may allow us to discuss carrier concentrations extracted from the Hall coefficients. Figure 4 shows the temperature dependence of the apparent carrier concentration which is calculated by V/NeR_H^{meas}, where V is the volume of a unit cell and N is the number of Cu atoms per unit cell.[27] The data show plateaus at low temperatures, where values of V/NeR_H^{meas} tell us the actual carrier concentrations; we can estimate the carrier concentration of the samples with $y = 6.30(6.35)$, 6.45, 6.50, and 6.55 to be $\sim 3\%$, $\sim 5\%$, $\sim 6\%$, and $\sim 7\%$, respectively. Validity of this method in estimating the carrier concentration is confirmed in a recent experiment in La$_{2-x}$Sr$_x$CuO$_4$ (Ref. 28). The carrier concentration at the superconductor-insulator boundary in YBCO appears to be a reasonable value of $\sim 4.5\%$ and thus the Hall coefficient in this region is apparently not affected by the conduction of the Cu-O chains. However, for more doped samples R_H shows the complicated temperature dependence, which is possibly modified by a contribution of the Cu-O chains if the Cu-O chains are conductive. Next we try to extract the Hall coefficient in the CuO$_2$ planes for samples with possibly conductive Cu-O chains.

IV. ANALYSIS AND DISCUSSION

In this section, we extract the physical properties of the CuO$_2$ planes by evaluating the effect of 1D conduction of the Cu-O chains. Our analysis is based on the parallel resistor model (PRM) along the b axis,[13] by which a T^2 law in the resistivity of the Cu-O chains was successfully extracted in optimally doped YBCO (Ref. 13) and YBa$_2$Cu$_4$O$_8$ (Y124) (Ref. 29). Excellent T^2 law is also observed in the resistivity of the Cu-O chains (ρ^{ch}) in our slightly overdoped samples with $y = 7.00$ as shown in Fig. 5(a). In addition, the value of the residual resistivity for ρ^{ch} (35.7 $\mu\Omega$ cm) is roughly a third of the previously reported value,[13] which evinces the cleanliness of our crystals. In the PRM we assume that (1) the Cu-O chains show electronic conduction along their direction, (2) interchain conduction is negligible, and (3) conduction bands of the Cu-O chain layer and the CuO$_2$ planes do not mix. Since the Cu-O chains run along the b axis, the total conductivity along the b axis is

$$\sigma_b^{tot} = \sigma^{pl} + \sigma^{ch},$$
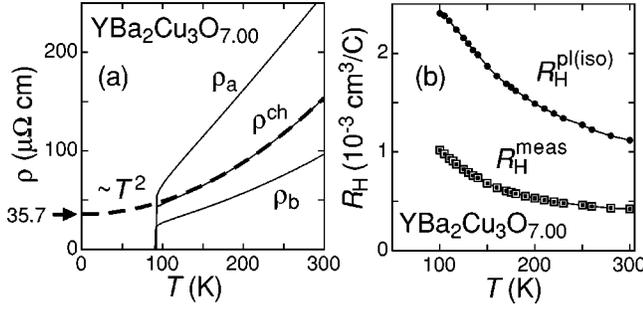
FIG. 5. (a) T dependences of ρ_a, ρ_b, and ρ^{ch} for samples with $y=7.00$, where ρ^{ch} is obtained by using the formula $1/\rho^{ch}=1/\rho_b - 1/\rho_a$. The dashed line is a fit to $\rho^{ch}(T)$ by aT^2+b. (b) T dependences of $R_H^{pl(iso)}$ and R_H^{meas} with $y=7.00$.

where σ^{pl} and σ^{ch} are the conductivity of the CuO_2 planes and the Cu-O chains, respectively. On the other hand, the total conductivity along the a axis is

$$\sigma_a^{tot}=\sigma^{pl},$$

since the conduction perpendicular to the Cu-O chains is neglected. The in-plane anisotropy of the electrical conduction of the CuO_2 planes is neglected for the moment (we will discuss the case of anisotropic CuO_2 planes later).

A. Hall coefficient

First we extract the Hall coefficient of the CuO_2 planes in the ρ_b sample because the process is simpler than that in the ρ_a sample. In the ρ_b sample the current flows separately through the CuO_2 planes and the Cu-O chains so that the current flowing through the CuO_2 planes (j_x^{pl}) is given by

$$j_x^{pl}=j_x^{tot}\left(\frac{\sigma^{pl}}{\sigma^{pl}+\sigma^{ch}}\right)=j_x^{tot}\left(\frac{\sigma_a^{tot}}{\sigma_b^{tot}}\right)=j_x^{tot}\left(\frac{\rho_b}{\rho_a}\right),$$

where j_x^{tot} is the total current. In the PRM it is expected that the Hall voltage is produced only by this current j_x^{pl} and the Cu-O chains do not affect the Hall electric field. Although some finite Hall coefficient is observed in quasi-1D systems,[30,31] as we discuss in Sec. IV E, contribution from the Cu-O chains to R_H can be neglected in the case of YBCO. Therefore, the Hall coefficient of the CuO_2 planes which are assumed to be isotropic, $R_H^{pl(iso)}$, is calculated as

$$R_H^{pl(iso)}=R_H^{meas}\left(\frac{j_x^{tot}}{j_x^{pl}}\right)=R_H^{meas}\left(\frac{\rho_a}{\rho_b}\right). \tag{1}$$

The same result is obtained also in the ρ_a sample. The longitudinal current j_x^{tot} flows only through the CuO_2 planes in the ρ_a sample, and thus $j_x^{pl}=j_x^{tot}$ and $j_x^{ch}=0$ are satisfied along the a axis. Naively, the measured Hall coefficient itself may seem to signify the Hall coefficient of the CuO_2 planes; however, the observed Hall voltage is reduced by the short-circuiting effect of the Cu-O chains in the ρ_a sample, because the transverse electric field E_y takes effect not only in the CuO_2 planes but also in the Cu-O chains. Since E_y is

parallel to the Cu-O chains, a current j_y^{ch} inevitably flows through the Cu-O chains. Therefore, in the Cu-O chains

$$E_y=\rho^{ch}j_y^{ch}$$

is satisfied. On the other hand, a countercurrent of the same magnitude of j_y^{ch} must flow through the CuO_2 planes because of the condition $j_y^{tot}=0$, where j_y^{tot} is the transverse current of the total system along the y axis. Therefore, E_y in the CuO_2 planes is calculated as

$$E_y=E_H-\rho^{pl}j_y^{ch},$$

where E_H is the Hall electric field which is generated to compensate the Lorentz force and E_H is given by

$$E_H=\rho_{yx}^{pl(iso)}j_x^{pl},$$

where $\rho_{yx}^{pl(iso)}$ is the Hall resistivity of the isotropic CuO_2 planes. Since E_y is identical in the CuO_2 planes and in the Cu-O chains,

$$E_H-\rho^{pl}j_y^{ch}=\rho^{ch}j_y^{ch}$$

is satisfied. We can express j_y^{ch} by using measurable properties as

$$j_y^{ch}=\frac{E_y}{\rho^{ch}}=E_y\left(\frac{1}{\rho_b}-\frac{1}{\rho_a}\right).$$

Therefore, we obtain

$$E_H=E_y+\rho_a j_y^{ch}=E_y\left(\frac{\rho_a}{\rho_b}\right),$$

and by dividing both sides of this equation by $j_x^{tot}(=j_x^{pl})$ Eq. (1) is obtained (using $\rho_{yx}^{pl(iso)}=E_H/j_x^{pl}$ and $\rho_{yx}^{meas}=E_y/j_x^{tot}$).

In the above analysis, the anisotropy of the resistivity in the CuO_2 planes is ignored. In other words, the global in-plane anisotropy is assumed to be caused only by the Cu-O chains. In recent measurements, however, we observed considerable anisotropy in the in-plane resistivity in oxygen deficient YBCO (Ref. 15). It is unlikely that oxygen deficient Cu-O chains show good 1D conduction, and thus the CuO_2 planes themselves must be anisotropic in the underdoped YBCO. If we recalculate Eq. (1) under the assumption of the anisotropic CuO_2 planes, we obtain

$$R_H^{pl}=R_H^{meas}\left(\frac{\rho_a}{\rho_b}\right)\left(\frac{\rho_b^{pl}}{\rho_a^{pl}}\right), \tag{2}$$

where R_H^{pl} is the Hall coefficient of the CuO_2 planes which can be anisotropic and $\rho_{a(b)}^{pl}$ is the resistivity of the CuO_2 planes along the $a(b)$ axis. Unfortunately, ρ_b^{pl} is not measurable and thus R_H^{pl} cannot be obtained if the CuO_2 planes are anisotropic and the Cu-O chains are conductive.

Figure 5(b) shows the temperature dependence of R_H^{meas} and $R_H^{pl(iso)}$ with $y=7.00$. $R_H^{pl(iso)}$ becomes more than two times larger than R_H^{meas} in the slightly overdoped samples. Figure 6(a) shows the temperature dependence of $R_H^{pl(iso)}$ for
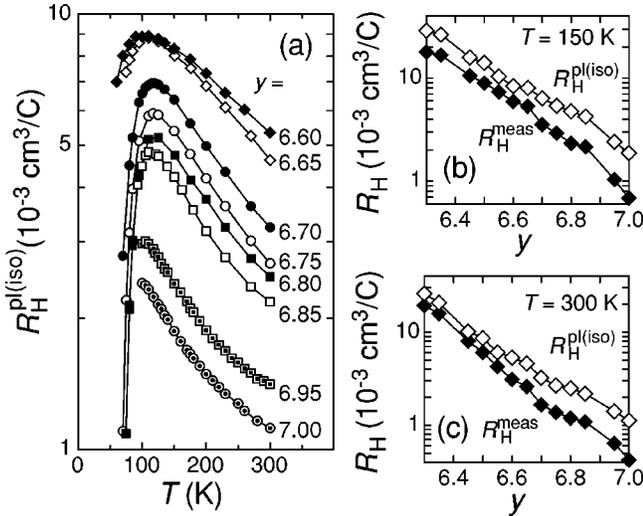
FIG. 6. (a) T dependences of $R_H^{pl(iso)}$ for samples with $y \geq 6.60$. (b),(c) y dependences of R_H^{meas} and $R_H^{pl(iso)}$ at 150 and 300 K.

samples with $y \geq 6.60$. The significant decrease at low temperatures in samples with $y \sim 6.75$ we noted in Fig. 3 does not diminish in $R_H^{pl(iso)}$. Figures 6(b) and 6(c) show the y dependences of R_H^{meas} and $R_H^{pl(iso)}$ at 150 K and 300 K, respectively. One should note that the obtained $R_H^{pl(iso)}$ just gives the maximum possible value of R_H^{pl} in the limit where the Cu-O chains are conductive and the CuO₂ planes are isotropic. On the other hand, if the Cu-O chains are insulating, R_H^{pl} becomes equal to R_H^{meas}. Therefore, the above analysis of the Hall coefficient provides a range of the R_H^{pl} values. In the heavily underdoped samples where chains are broken, the chain conduction is expected to be negligible and the observed peculiar anisotropy is most likely due to the planes;[15] in this case, one can see from Eq. (2) that R_H^{meas} is a true measure of R_H of the planes.

B. Hall conductivity

In contrast to the Hall resistivity (Hall coefficient), we can unambiguously obtain the Hall *conductivity* of the CuO₂ planes regardless of anisotropy of the CuO₂ planes. Since the Hall conductivity is calculated by $\sigma_{xy} = \rho_{yx}/(\rho_{xx}\rho_{yy} - \rho_{yx}\rho_{xy}) \simeq \rho_{yx}/(\rho_{xx}\rho_{yy})$, in YBCO we calculate it as

$$\sigma_{xy}^{pl} = \frac{\rho_{yx}^{pl}}{\rho_a^{pl}\rho_b^{pl}}, \tag{3}$$

where σ_{xy}^{pl} is the Hall conductivity of the CuO₂ planes and ρ_{yx}^{pl} is the Hall resistivity of the CuO₂ planes. We can transform Eq. (3) by using Eq. (2) into

$$\sigma_{xy}^{pl} = \frac{\rho_{yx}^{meas}}{\rho_a\rho_b}.$$

Note that this result tells us that the Hall conductivity in the CuO₂ planes is the same as that of the total system regardless of the anisotropy of the CuO₂ planes, which means that
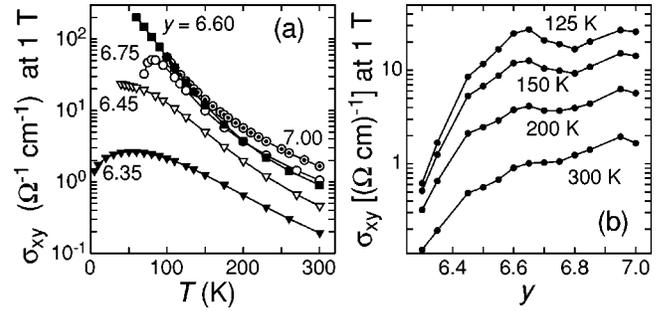
FIG. 7. (a) T dependences of σ_{xy} for samples with five representative y values. (b) y dependences of σ_{xy} at 125, 150, 200, and 300 K.

$\sigma_{xy}^{pl} = \sigma_{xy}$ always holds in YBCO. Thus, in the following we do not discriminate σ_{xy}^{pl} from σ_{xy}.

Figure 7(a) shows the temperature dependences of σ_{xy} for samples with five representative compositions in a semilog plot. We omitted other samples only for clarity. For the samples with $y \sim 6.75$ the low-temperature decrease remains in σ_{xy} and thus the observed decrease in R_H at low temperatures is not an artifact of some feature of the Cu-O chains but is an intrinsic feature of the CuO₂ planes. The y dependence of σ_{xy} at fixed temperatures is shown in Fig. 7(b). σ_{xy} at low temperatures appears to be suppressed at the oxygen content of ~ 6.75 (Ref. 20); this anomaly also has nothing to do with the Cu-O chains. As we discussed in Ref. 20, this anomaly must be due to a peculiar doping dependence of the mobility of the carriers.

C. Hall angle

Next, let us analyze the cotangent of the Hall angle ($\cot \Theta_H$). As mentioned above, the Hall coefficient R_H appears to show no in-plane anisotropy as far as magnetic fields are applied along the c axis, which is expected from the Onsager relation. On the other hand, the in-plane resistivity is observed to be anisotropic[15] in a whole doping range in YBCO. The parameter $\cot \Theta_H$ is calculated by ρ_{xx}/ρ_{yx}, and thus in general $\cot \Theta_H$ is expected to be anisotropic. With careful considerations, it turns out that one can obtain the Hall angle of the CuO₂ planes from measurable properties only in the ρ_b sample. We can express $\cot \Theta_H$ ($\equiv E_x/E_y = \rho_{xx}/\rho_{yx}$) of the CuO₂ planes along the b axis as

$$\cot \Theta_{H(b)}^{pl} = \frac{\rho_b^{pl}}{\rho_{yx}^{pl}},$$

where $\Theta_{H(b)}^{pl}$ is the Hall angle of the CuO₂ planes with the current along the b axis, and from Eq. (2) we obtain

$$\cot \Theta_{H(b)}^{pl} = \frac{\rho_b}{\rho_{yx}^{meas}},$$

where the nonmeasurable property ρ_b^{pl} is canceled out.

Figure 8(a) shows a $\cot \Theta_{H(b)}^{pl}$ vs T^2 plot for samples with $y = 6.30 - 7.00$, where the origin for each sample is shifted for clarity. In highly doped samples with $y = 6.95$ and
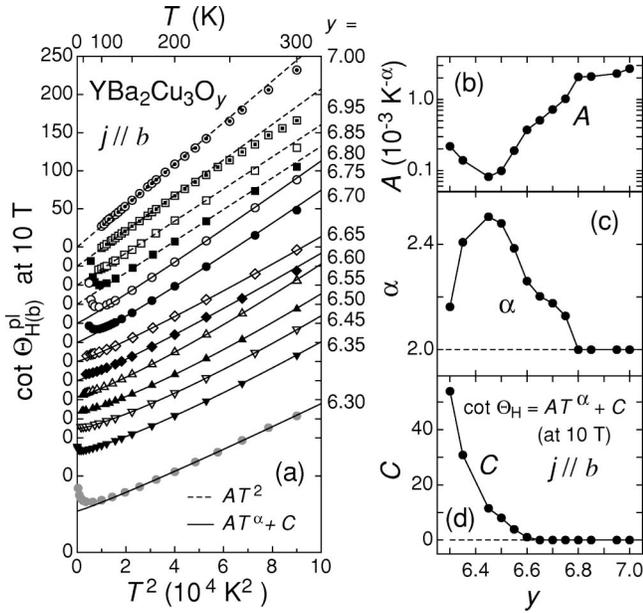
FIG. 8. (a) $\cot \Theta_{H(b)}^{pl}$ vs T^2 plot for $y = 6.30$–7.00, where dashed lines are fits to the data by AT^2 and solid lines are fits to the data by $AT^\alpha + C$. (b),(c),(d) y dependences of the fitting parameters A, α, and C, respectively.

FIG. 9. (a) $\cot \Theta_{H(a)}$ vs T^2 plot for $y = 6.30$–7.00, where solid lines are fits to the data by $AT^\alpha + C$. (b),(c),(d) y dependences of the fitting parameters A, α, and C, respectively.

7.00, $\cot \Theta_H$ appears to be fitted well by a function of AT^2 except for high temperatures (dashed lines in the figure). Note that only one fitting parameter is used for the fitting and the residual component is zero. This result is consistent with the reported result of ρ_b / ρ_{yx} for a 90 K sample.[17] The data for $y = 6.85$ and 6.80 can also be fitted by the function of AT^2 (dashed lines in the figure); however, at low temperatures some upward deviation is observed. The low-temperature deviation seen in samples with $y = 6.60$–6.85 is clearly corresponding to the decrease in R_H at low temperatures and thus this anomaly seems to be one of the 60 K anomalies rather than an effect of the pseudogap.[22,32] The T dependences of $\cot \Theta_H$ for samples with $y \leqslant 6.75$ can be fit by a function of $AT^\alpha + C$ with $\alpha > 2$ (solid lines in the figure) except for the upturn at low temperatures. Figures 8(b)–8(d) show y dependences of the fitting parameters. The power α is 2 for $y \geqslant 6.80$ and increases with decreasing oxygen content from 6.75. In the slightly doped region $\alpha(y)$ shows a peak at $y \sim 6.45$ and decreases with decreasing y; α is shown to be 2 in the low doping limit.[28] The "residual" component C becomes finite only for y below ~ 6.60, and this is reminiscent of the result of the normal-state orbital magnetoresistance,[33] in which the residual component b in its temperature dependence $(aT^2 + b)^{-2}$ becomes finite for $y \leqslant 6.60$.

Of course we can also calculate $\cot \Theta_H$ with the current along the a axis ($\cot \Theta_{H(a)}$), which may contain contribution from the Cu-O chains if the Cu-O chains are conductive. Figure 9(a) shows a $\cot \Theta_{H(a)}$ vs T^2 plot for samples with $y = 6.30$–7.00 (the origin for each sample is shifted for clarity). In highly doped samples with $y \geqslant 6.85$, $\cot \Theta_H$ appears to be fitted well by a function of $AT^2 + C$ except for high temperatures (dashed lines in the figure); however, unlike
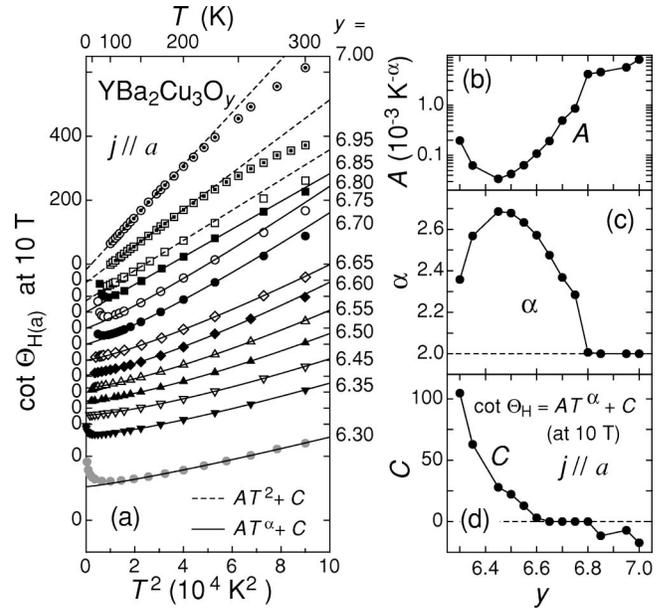
$\cot \Theta_{H(b)}^{pl}$, the residual component C becomes negative. The solid lines are fits to the data by a function $AT^\alpha + C$ with $\alpha > 2$. For $y = 6.70$–6.80 the data can be fitted only below ~ 200 K, whereas for more underdoped samples with $y \leqslant 6.65$ the data are fitted well. The y dependences of the fitting parameters are shown in Figs. 9(b)–9(d). Observed tendency is qualitatively similar to that in $\cot \Theta_{H(b)}^{pl}$ except for the residual component for highly doped samples with $y \geqslant 6.85$.

From the Hall angle we can extract the Hall mobility μ_H; Figs. 10(a) and 10(b) show the y dependences of μ_H with the current along the b axis and the a axis, respectively, at various temperatures. Of course, the meaning of μ_H with the current along the b axis is more transparent, but both data of μ_H are qualitatively consistent. At low temperatures the Hall mobility increases with decreasing y from 6.80 to 6.65. This enhancement of the mobility compensates the effect of decreasing carrier concentration and thus an overlapping of $\rho_a(T)$ is observed for samples with $y = 6.65$–6.80 (Ref. 20).
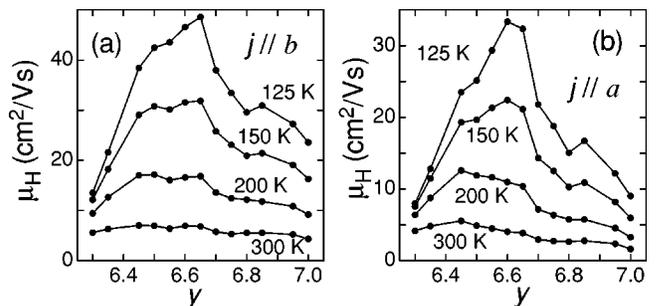
FIG. 10. y dependences of the Hall mobility μ_H at 125, 150, 200, and 300 K with the current along the b axis (a) and the a axis (b).
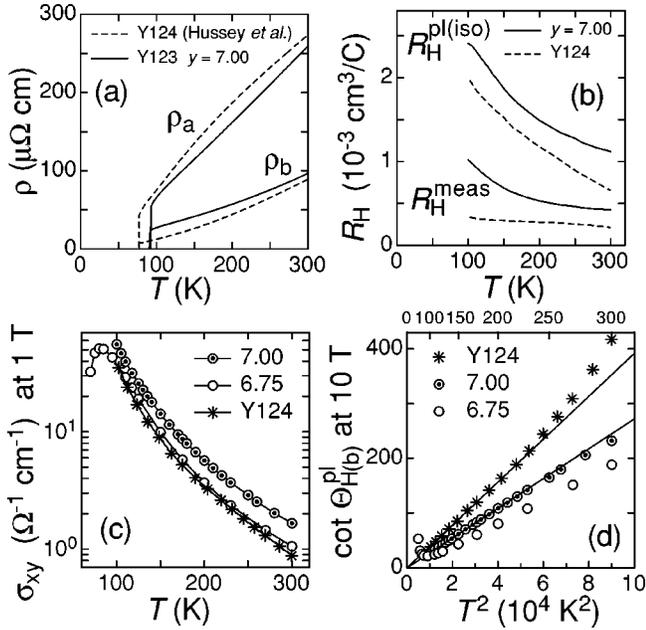
FIG. 11. The temperature dependences of (a) ρ_a and ρ_b, (b) R_H^{meas} and $R_H^{\text{pl(iso)}}$ for Y124 and YBCO samples with $y=7.00$. (c) T dependences of σ_{xy} and (d) $\cot\Theta_{H(b)}^{\text{pl}}$ vs T^2 plot for Y124 and YBCO samples with $y=7.00$ and 6.75.

D. Comparison with YBa$_2$Cu$_4$O$_8$

Y124 can be a good reference system of YBCO, because Y124 has doubled Cu-O chains which are believed to show good conduction. Figures 11(a)–11(d) show the temperature dependences of ρ_a, ρ_b, R_H^{meas}, $R_H^{\text{pl(iso)}}$, σ_{xy}, and $\cot\Theta_{H(b)}^{\text{pl}}$ for our YBCO samples ($y=7.00$ and/or 6.75) together with those for Y124, which are extracted from the data in published papers.[29,34] Figure 11(a) shows the temperature dependences of ρ_a and ρ_b. While these data are not very different between YBCO$_{7.00}$ and Y124, the anisotropy of the resistivity in Y124 is larger than that in YBCO$_{7.00}$; this strong anisotropy is considered to come from the doubled Cu-O chains. The temperature dependence of the measured Hall coefficient is very different [Fig. 11(b)]; that of Y124 is much weaker than YBCO. However, when we calculate $R_H^{\text{pl(iso)}}$ from the data, it shows qualitatively similar behavior in both YBCO$_{7.00}$ and Y124. This fact suggests that the very conductive Cu-O chains mask a strong temperature dependence of the Hall coefficient in Y124. Figure 11(c) shows $\sigma_{xy}(T)$ in Y124 and YBCO samples with $y=7.00$ and 6.75. The Hall conductivity of Y124 appears to be very close to that of YBCO$_{6.75}$ rather than YBCO$_{7.00}$. This is understandable because Y124 is naturally underdoped. The cotangent of the Hall angle along the b axis is shown in Fig. 11(d). Data for Y124 can be fitted well by a function of AT^2 below ~ 230 K (solid lines). Thus, the transport properties of the CuO$_2$ planes in Y124, which are extracted by the analysis presented in this paper, appear to be similar to those in YBCO, and this fact gives confidence in the validity of our analysis.

E. Finite Hall coefficient in quasi-1D systems

Our analysis assumes that the Cu-O chains do not contribute to the Hall electric field, whereas in actual quasi-1D sys-

tems the Hall resistivity has been observed to be not negligible.[30,31] This is mostly due to the large resistivity in the direction perpendicular to the chains, ρ_{yy}. One can easily understand this situation by noticing that the Hall resistivity ρ_{yx} is expressed as

$$\rho_{yx} = \sigma_{xy}\rho_{xx}\rho_{yy},$$

where σ_{xy} is the Hall conductivity of the quasi-1D system and ρ_{xx} is the resistivity along the chain direction. (In quasi-1D metals, $\rho_{yy} \gg \rho_{xx}$.) This formula tells us that a sizable ρ_{yx} can be observed for a very small σ_{xy}, if ρ_{yy} is very large. In fact, if we calculate σ_{xy} for the case of PrBa$_2$Cu$_4$O$_8$ (Pr124) using published data,[31] it turns out that σ_{xy} of Pr124 is only 10^{-4} of that of YBCO. This estimate indicates that the contribution of σ_{xy} of the Cu-O chains to the total Hall conductivity is negligible in YBCO. More generally speaking, in a chain-plane-composite system, the Hall conductivity of the total system, σ_{xy}^{tot}, is given by

$$\sigma_{xy}^{\text{tot}} = \sigma_{xy}^{\text{pl}} + \sigma_{xy}^{\text{ch}},$$

where σ_{xy}^{pl} and σ_{xy}^{ch} are the individual Hall conductivity of the planes and the chains. In the chain layer, σ_{xy}^{ch} is always very small even when ρ_{yx}^{ch} is sizable, as we discussed above. Therefore, one can safely neglect the contribution of σ_{xy}^{ch} in the composite system and consider that the behavior of σ_{xy}^{tot} is governed almost solely by σ_{xy}^{pl}.

F. Anomalous decrease in R_H at low tempeartures

Finally, let us discuss the pronounced decrease in R_H observed only for samples near $y \simeq 6.75$ at low temperatures. We emphasize that the present data are measured only at $T > T_c$ by sweeping the magnetic fields up to at least 10 T, where the field dependence of the Hall resistivity is H linear in the whole region. Therefore, the decrease in R_H reflects the normal-state property and is irrelevant to the negative Hall anomaly[35] which is observed only at low fields and below T_c. Our observation is somewhat reminiscent of the decrease in R_H observed in La$_{2-x-y}$Nd$_y$Sr$_x$CuO$_4$ (Ref. 36), and thus the present result may actually be related to charged stripes. In fact, the carrier concentration of YBCO$_{6.75}$ is consistent with the hole concentration $p \sim 1/8$ (per Cu atom in the CuO$_2$ planes) (Refs. 33, 37 and 38), where so-called "1/8 anomaly" is observed in cuprate superconductors; therefore, it is likely that the observed anomaly in R_H is related to the "1/8 problem." Note, however, that the in-plane resistivity anisotropy is observed to become *small* in samples with $y \sim 6.75$ at low temperatures.[15] Such a result may seem to imply that one dimensionality of charge dynamics in the CuO$_2$ planes is minimal, which is inconsistent with the picture of static stripes at $p \sim 1/8$. However, the possibility of fluctuating charge stripes is not excluded, because the stripe liquid can be in an "isotropic" phase;[39] in this case, the decrease in R_H can be caused by a realization of the particle-hole symmetry when the hole concentration becomes $\sim 1/8$ and the stripes become 1/4 filled.[40,41]

V. SUMMARY

In summary, we show how to extract the Hall conductivity and the cotangent of the Hall angle of the CuO_2 planes in YBCO regardless of conduction of the Cu-O chains and in-plane anisotropy of the CuO_2 planes. The analysis is applied to the data of untwinned YBCO for a wide range of doping as well as the available data of Y124 in the literature. The present analysis provides a legitimate way to discuss the Hall effect of the CuO_2 planes in YBCO on the same ground as that in other high-T_c cuprates.

[1] N.P. Ong, in *Physical Properties of High Temperature Superconductors*, edited by D.M. Ginsberg (World Scientific, Singapore, 1990), Vol. 2, p. 459.

[2] T.R. Chien, Z.Z. Wang, and N.P. Ong, Phys. Rev. Lett. **67**, 2088 (1991).

[3] P.W. Anderson, Phys. Rev. Lett. **67**, 2092 (1991).

[4] A. Carrington, A.P. Mackenzie, C.T. Lin, and J.R. Cooper, Phys. Rev. Lett. **69**, 2855 (1992).

[5] A.S. Alexandrov, A.M. Bratkovsky, and N.F. Mott, Phys. Rev. Lett. **72**, 1734 (1994).

[6] G. Kotliar, A. Sengupta, and C.M. Varma, Phys. Rev. B **53**, 3573 (1996).

[7] P. Coleman, A.J. Schofield, and A.M. Tsvelik, Phys. Rev. Lett. **76**, 1324 (1996).

[8] B.P. Stojković and D. Pines, Phys. Rev. B **55**, 8576 (1997).

[9] L.B. Ioffe and A.J. Millis, Phys. Rev. B **58**, 11 631 (1998).

[10] A.T. Zheleznyak, V.M. Yakovenko, and H.D. Drew, Phys. Rev. B **59**, 207 (1999).

[11] H. Kontani, K. Kanki, and K. Ueda, Phys. Rev. B **59**, 14 723 (1999).

[12] C.M. Varma and E. Abrahams, Phys. Rev. Lett. **86**, 4652 (2001).

[13] R. Gagnon, C. Lupien, and L. Taillefer, Phys. Rev. B **50**, 3458 (1994).

[14] K. Takenaka, K. Mizuhashi, H. Takagi, and S. Uchida, Phys. Rev. B **50**, R6534 (1994).

[15] Y. Ando, K. Segawa, S. Komiya, and A.N. Lavrov, Phys. Rev. Lett. **88**, 137005 (2002).

[16] L. Hoffmann, A.A. Manuel, M. Peter, E. Walker, M. Gauthier, A. Shukla, B. Barbiellini, S. Massidda, Gh. Adam, S. Adam, W.N. Hardy, and R. Liang, Phys. Rev. Lett. **71**, 4047 (1993).

[17] J.P. Rice, J. Giapintzakis, D.M. Ginsberg, and J.M. Mochel, Phys. Rev. B **44**, 10 158 (1991).

[18] K. Segawa and Y. Ando, Phys. Rev. B **59**, R3948 (1999).

[19] A. Lavrov and L. Kozeeva, Physica C **253**, 313 (1995), and references therein.

[20] K. Segawa and Y. Ando, Phys. Rev. Lett. **86**, 4907 (2001).

[21] K. Kishio, J. Shimoyama, T. Hasegawa, K. Kitazawa, and K. Fueki, Jpn. J. Appl. Phys., Part 2 **26**, L1228 (1987).

[22] Y. Abe, K. Segawa, and Y. Ando, Phys. Rev. B **60**, R15 055 (1999).

[23] L. Onsager, Phys. Rev. **38**, 2265 (1931).

[24] J.M. Harris, N.P. Ong, P. Matl, R. Gagnon, L. Taillefer, T. Kimura, and K. Kitazawa, Phys. Rev. B **51**, R12 053 (1995).

[25] For estimating the distribution of the current, it is necessary to map an anisotropic sample to an equivalent isotropic sample [L.J. van der Pauw, Philips Res. Rep. **16**, 187 (1961)]. In YBCO ρ_b is always smaller than ρ_a and thus the ρ_b sample is effectively shorter and wider than the ρ_a sample. R_H becomes smaller when electrodes get closer to a current contact of a sample, because the current contact is made on the whole surface on which the Hall voltage is short circuited. Since in the ρ_b sample electrodes become effectively closer to the current contacts than in the ρ_a sample, R_H^b becomes slightly smaller than R_H^a.

[26] T. Ito, K. Takenaka, and S. Uchida, Phys. Rev. Lett. **70**, 3995 (1993).

[27] Y. Ando, Y. Hanaki, S. Ono, T. Murayama, K. Segawa, N. Miyamoto, and S. Komiya, Phys. Rev. B **61**, R14 956 (2000).

[28] Y. Ando, Y. Kurita, S. Komiya, S. Ono, and K. Segawa (unpublished).

[29] N.E. Hussey, K. Nozawa, H. Takagi, S. Adachi, and K. Tanabe, Phys. Rev. B **56**, R11 423 (1997).

[30] G. Mihály, I. Kézsmárki, F. Zámborszky, and L. Forró, Phys. Rev. Lett. **84**, 2670 (2000); J. Moser, J.R. Cooper, D. Jérome, B. Alavi, S.E. Brown, and K. Bechgaard, *ibid.* **84**, 2674 (2000).

[31] S. Horii, U. Mizutani, H. Ikuta, Y. Yamada, J.H. Ye, A. Matsushita, N.E. Hussey, H. Takagi, and I. Hirabayashi, Phys. Rev. B **66**, 054530 (2002).

[32] Z.A. Xu, Y. Zhang, and N.P. Ong, cond-mat/9903123 (unpublished).

[33] Y. Ando and K. Segawa, Phys. Rev. Lett. **88**, 167005 (2002).

[34] B. Bucher, P. Steiner, J. Karpinski, E. Kaldis, and P. Wachter, Phys. Rev. Lett. **70**, 2012 (1993).

[35] T. Nagaoka, Y. Matsuda, H. Obara, A. Sawa, T. Terashima, I. Chong, M. Takano, and M. Suzuki, Phys. Rev. Lett. **80**, 3594 (1998), and references therein.

[36] T. Noda, H. Eisaki, and S. Uchida, Science **286**, 265 (1999).

[37] Y. Ando and K. Segawa, J. Phys. Chem. Solids **63**, 2253 (2002).

[38] K. Segawa and Y. Ando, J. Low Temp. Phys. **131**, 821 (2003).

[39] S.A. Kivelson, E. Fradkin, and V.J. Emery, Nature (London) **393**, 550 (1998).

[40] V.J. Emery, E. Fradkin, S.A. Kivelson, and T.C. Lubensky, Phys. Rev. Lett. **85**, 2160 (2000).

[41] P. Prelovšek, T. Tohyama, and S. Maekawa, Phys. Rev. B **64**, 052512 (2001).