

Universal transport anomaly in $\text{YBa}_2\text{Cu}_3\text{O}_7$ -type systems with reduced carrier density

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A systematic study of the $\text{YBa}_2\text{Cu}_3\text{O}_7$ -type (1:2:3) compounds with various carrier densities has revealed a universal presence of an anomaly in the transport coefficients at around 120 K when the carrier number per copper is 0.10–0.15. A comparison with a similar anomaly reported in $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ at a comparable carrier density ($x=0.125$) suggests that it is an inherent property of the CuO_2 plane. The transport anomaly consists of a decrease in the Hall coefficient and an upturn of the resistivity as the temperature is lowered below 120 K. It suggests the destruction of a part of the Fermi surface by gap formation. Possible origins of such Fermi-surface instabilities and other possibilities for the transport anomaly are discussed.

INTRODUCTION

One of the central issues in the physics of high- T_c superconductors is the change of their electronic states with carrier doping. In the case of $\text{YBa}_2\text{Cu}_3\text{O}_7$ -type (1:2:3) compounds, the hole density can be varied by various substitutions and/or oxygen stoichiometry.

Among the high- T_c superconductors, T_c as a function of doped carrier density appears to give similar curves,^{1,2} where the maximum of T_c is located at a carrier number per copper ~ 0.2 . The carrier density can be estimated by chemical methods such as iodometric titration on one hand, and the Hall measurement on the other hand. Although the carrier density estimated from the Hall coefficient by assuming a single-band conduction generally shows a reasonable agreement with a chemical estimate in the low doping region, the discrepancy becomes significant at higher carrier densities.

As the carrier density is varied, the transport coefficients drastically change both magnitudes and their temperature dependences. Although the behavior of the transport coefficients should, in principle, provide useful information about the nature of the normal state from which the high- T_c superconductivity evolves, a coherent picture is yet to be obtained. In particular, interpretation of the Hall effect remains a stumbling block.

The Hall coefficient in high- T_c superconductors often shows a strong temperature dependence. A fully oxygenated 1:2:3 compound, whose Hall coefficient varies roughly as $1/T$ down to T_c , is the most conspicuous in this regard. It is obvious that such a temperature-dependent Hall coefficient is inconsistent with a simple single-band picture. Various attempts have been made to account for the temperature dependence of the Hall coefficient with the ubiquitous T -linear resistivity, including multiband conduction,³ narrow-band conduction,⁴ magnetic skew scattering,⁵ and resonating valence bond-(RVB) based transport mechanisms.⁶

Generally, the temperature dependence becomes less pronounced as the hole density and, hence, T_c is lowered. However, at a lower hole density, a broad maximum of

the Hall coefficient as a function of temperature was widely observed among high- T_c superconductors. This kind of behavior was first found by Tamegai *et al.* for $\text{Nd}_{2-x+y}\text{Ce}_x\text{Ba}_{2-y}\text{Cu}_3\text{O}_{10+\delta}$ (2:2:3 phase) having a T_c of ~ 30 K.⁷ Ikegawa *et al.*⁸ studied the temperature dependence of the Hall coefficient for various high- T_c superconductors and suggested a correlation of the broad maximum of the Hall coefficient with the presence of fluorite-type layers in the crystal structure.

To gain more insight into the origin of the anomalous temperature dependence of the Hall coefficient, we have performed a systematic study of the 1:2:3 compounds having a wide range of carrier densities. In the case of 1:2:3 compounds, the coexistence of the CuO_2 plane and CuO chain brings another complication. In the present work, two different methods have been employed to change the hole density in the 1:2:3 compounds: (1) Y- and Ba-site solid solution, $\text{Nd}_{1+x}\text{Ba}_{2-x}\text{Cu}_3\text{O}_{7+y}$; (2) chain-site substitution, $\text{YBa}_2(\text{Cu}_{1-x}\text{Co}_x)_3\text{O}_{7+y}$. Isoelectronic plane Cu-site substitution, $\text{YBa}_2(\text{Cu}_{1-x}\text{Zn}_x)_3\text{O}_{7-y}$, was also made for a comparison. We observed a maximum in the Hall coefficient accompanied by an upturn of the resistivity when the carrier number is reduced to a certain value. These findings, together with a similar anomaly reported in the 2:1:4 system, can be understood as a universal anomaly in the CuO_2 planes at a certain carrier number per copper.

EXPERIMENTS AND RESULTS

Two series of 1:2:3 compounds, $\text{Nd}_{1+x}\text{Ba}_{2-x}\text{Cu}_3\text{O}_{7+y}$ and $\text{YBa}_2(\text{Cu}_{1-x}\text{Co}_x)_3\text{O}_{7+y}$, were prepared by a standard solid-state reaction. Starting powders of oxides and BaCO_3 were mixed thoroughly and fired for more than 60 h with a few intermediate grindings at 930°C and 900°C for $\text{Nd}_{1-x}\text{Ba}_{2-x}\text{Cu}_3\text{O}_{7+y}$ and $\text{YBa}_2(\text{Cu}_{1-x}\text{Co}_x)_3\text{O}_{7+y}$, respectively. After being pressed into pellets, they were fired 30°C above the previous setting for 24 h in oxygen flow. To equilibrate the oxygen content in atmospheric oxygen, pellets were slowly cooled down to 300°C in 12 h. The range of the step of x for each series was chosen

so that obtained samples cover the range from a 90 K superconductor to the one near the metal-insulator transition. Samples of $\text{YBa}_2(\text{Cu}_{1-x}\text{Zn}_x)_3\text{O}_{7-y}$ were also prepared in the same way as $\text{YBa}_2(\text{Cu}_{1-x}\text{Co}_x)_3\text{O}_{7+y}$.

In the high- T_c superconductors, the Hall resistivity was typically 3 or 4 orders of magnitude smaller than the diagonal resistivity even in the magnetic field of 5 T. In order to perform a precise Hall measurement, it was essential to minimize the misalignment of the Hall arms and also to achieve a low contact resistance. For this purpose, a sample was cut from a pellet into a six-terminal Hall-bar shape as illustrated in the inset of Fig. 1(a) by use of a numerically controlled milling machine with a diamond-tip bit. Gold was evaporated onto the pads and then gold wires were attached with silver paste. Subsequent curing at 400 °C further reduced the contact resistances. The final contact resistance depended on the bulk resistivity of the sample, but was typically 0.1 Ω .

The Hall measurements were made at a field of 5 T generated by a split-type superconducting magnet. Current density passed through the sample was typically 20 A/cm². The standard procedure of taking data for

both polarities of the current and magnetic field to cancel out thermal emf and resistance component was followed. Reversing the polarity of the magnetic field was done by rotating the sample by 180° about the vertical axis by a computer-controlled stepping motor. Data were taken automatically as the sample was cooled slowly (0.1–0.2 K/min) in a gas-flow-type insert Dewar. The temperature dependence of the resistivity at zero field was measured in a separate run.

The temperature dependence of the Hall coefficient and the resistivity (at zero field) for $\text{Nd}_{1+x}\text{Ba}_{2-x}\text{Cu}_3\text{O}_{7+y}$ are shown in Figs. 1(a) and 1(b). With increasing Nd content, the superconducting transition temperature (T_c) decreases and the metallic conduction gradually turns into a semiconducting one. With lowering temperature from room temperature, the Hall coefficient initially increases for all samples. But, the low-temperature behavior strongly depends on the content of Nd; the Hall coefficients for higher- T_c samples ($x=0, 0.1$) show the $1/T$ dependences down to T_c . In samples with lower T_c ($x=0.4, 0.5$), the Hall coefficients are roughly constant below 100 K. Samples with moderate T_c ($x=0.2, 0.3$) show anomalous temperature dependences of the Hall coefficient. They show a maximum in the Hall coefficient around 120 K which is well above the T_c of these samples (56 K for $x=0.2$, 38 K for $x=0.3$). In Fig. 2, the Hall coefficients and the resistivity are plotted simultaneously for $\text{Nd}_{1.2}\text{Ba}_{1.8}\text{Cu}_3\text{O}_{7+y}$, which showed the most pronounced maximum in the Hall coefficient. It is clear that the maximum in the Hall coefficient is accompanied by the upturn of the resistivity. If we consider only one band for this system, the decrease in the Hall coefficient means an increase in the carrier density, which is hard to be reconciled with the increase in the resistivity.

The anomaly in the transport coefficients strongly suggests a change in the electronic structure at the temperature. But it is necessary to check whether extrinsic effects might be responsible for the anomaly. First of all,

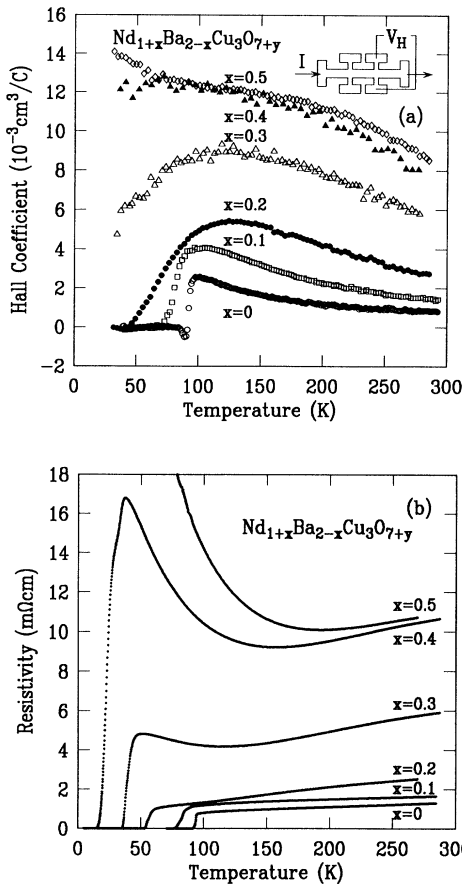


FIG. 1. Temperature dependence of (a) the Hall coefficient and (b) the resistivity in $\text{Nd}_{1+x}\text{Ba}_{2-x}\text{Cu}_3\text{O}_{7+y}$. The inset shows the shape of the sample with Hall arms used for the Hall coefficient measurements.

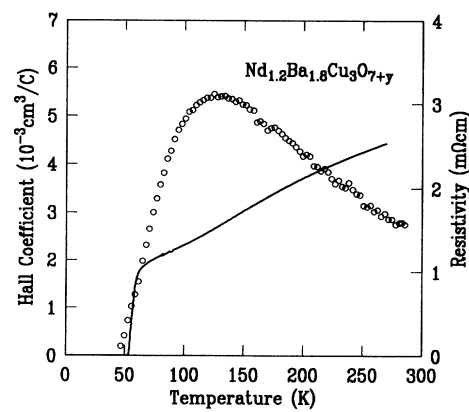


FIG. 2. Temperature dependence of the Hall coefficient (open circle) and the resistivity (solid line) in $\text{Nd}_{1.2}\text{Ba}_{1.8}\text{Cu}_3\text{O}_{7+y}$. Note the steep decrease in the Hall coefficient accompanied by the upturn of the resistivity below 120 K.

compositional inhomogeneity might result in a segregation of partially higher- T_c regions and hence decrease the Hall coefficient above bulk T_c , because, in the superconducting state, the Hall coefficient should go to zero. Secondly, strong two-dimensional superconducting fluctuation would suppress the Hall coefficient.⁹ Both of them, however, are inconsistent with increase in the resistivity. In order to see more clearly what is happening around 120 K, we measured the magnetic field dependence of the transport coefficients.

Figure 3 shows the magnetic field dependence of the Hall resistance (V_H/I , where V_H is the Hall voltage and I is the current passed through the sample) and the normalized magnetoresistance in $\text{Nd}_{1.2}\text{Ba}_{1.8}\text{Cu}_3\text{O}_{7+y}$. The Hall voltage is essentially linear in the magnetic field at least down to 90 K, which is much below the temperature of the maximum in the Hall coefficient. Magnetoresistance is small but shows a quadratic magnetic field dependence at higher temperature, which is expected from the fluctuation theory for $T \gg T_c$. Below about 90 K, it shows a marked increase at low fields. This is probably due to the presence of higher- T_c regions with weak links whose superconductivity is easily destroyed by a weak magnetic field. These observations support that the

anomaly at 120 K we found in the present study is intrinsic to the material.

In Figs. 4 and 5, the temperature dependence of the Hall coefficient and the resistivity are shown for $\text{YBa}_2(\text{Cu}_{1-x}\text{Co}_x)_3\text{O}_{7+y}$ and $\text{YBa}_2(\text{Cu}_{1-x}\text{Zn}_x)_3\text{O}_{7-y}$, respectively. The variation of the Hall coefficient and the resistivity in the $\text{YBa}_2(\text{Cu}_{1-x}\text{Co}_x)_3\text{O}_{7+y}$ is very similar to that in $\text{Nd}_{1+x}\text{Ba}_{2-x}\text{Cu}_3\text{O}_{7+y}$. Similar data for the Co-substituted 1:2:3 system above 80 K have already been reported by Clayhold *et al.*¹⁰ They reported an increase in the Hall coefficient and a decrease in the temperature derivative of it with x . We observed the similar trend at higher temperature, but the presence of the transport anomaly is also evident in this system, which is more remarkable for $x=0.05$ ($T_c=57$ K). On the other hand, in the $\text{YBa}_2(\text{Cu}_{1-x}\text{Zn}_x)_3\text{O}_{7-y}$ system, the absolute value of the Hall coefficient does not change appreciably with x , but it increases monotonically down to just above T_c without any maximum with lowering temperature. Similar data for the Zn-substituted system have also been obtained by Affronte *et al.*¹¹ It is clear that, in contrast with $\text{YBa}_2(\text{Cu}_{1-x}\text{Co}_x)_3\text{O}_{7+y}$, the resistivity in the Zn system shows no sudden upturn at any temperature.

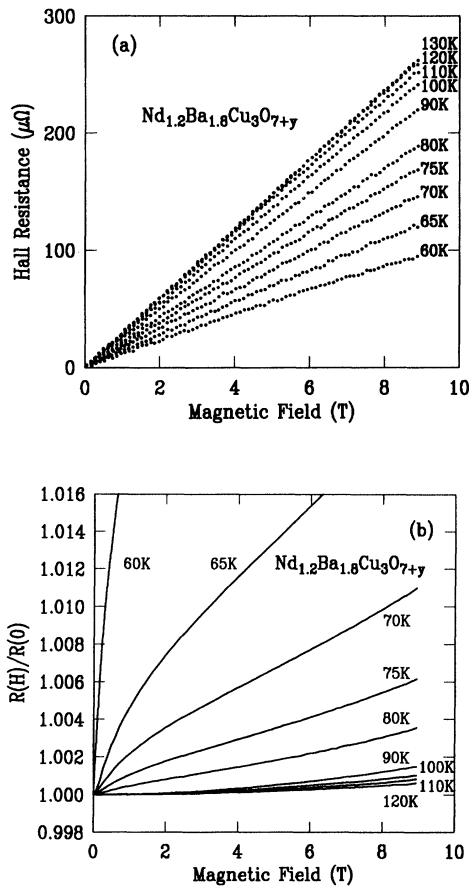


FIG. 3. (a) The Hall resistance and (b) the magnetoresistance as a function of the magnetic field in $\text{Nd}_{1.2}\text{Ba}_{1.8}\text{Cu}_3\text{O}_{7+y}$.

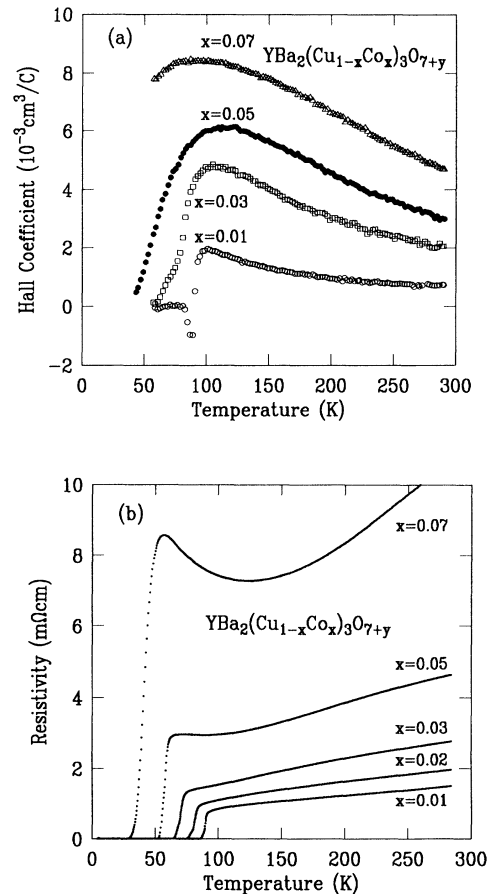


FIG. 4. Temperature dependence of (a) the Hall coefficient and (b) the resistivity in $\text{YBa}_2(\text{Cu}_{1-x}\text{Co}_x)_3\text{O}_{7+y}$.

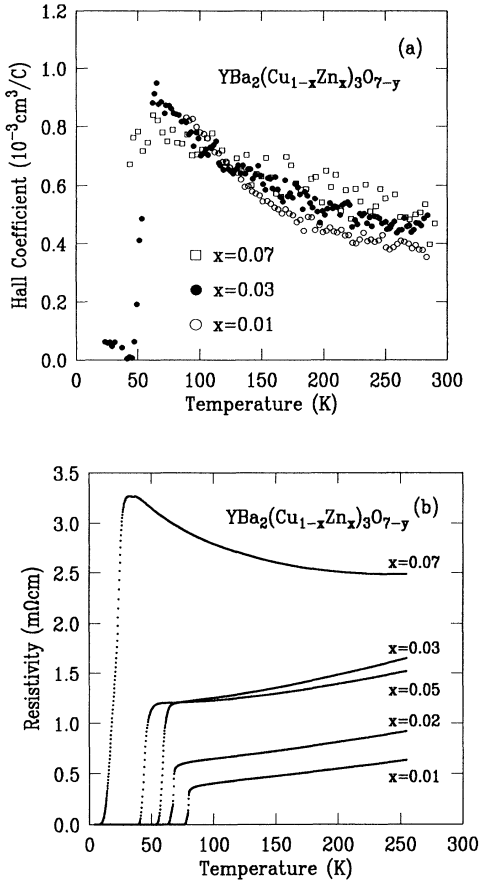


FIG. 5. Temperature dependence of (a) the Hall coefficient and (b) the resistivity in $\text{YBa}_2(\text{Cu}_{1-x}\text{Zn}_x)_3\text{O}_{7-y}$.

DISCUSSION

The fact that we observed a broad maximum of the Hall coefficient in the 1:2:3 systems, which do not have fluorite-type layers, is a counterexample for the suggestion by Ikegawa *et al.*⁸ that their presence was a necessary condition for the broad maximum in the Hall coefficient. The anomaly found in the present transport measurements shares some features in common with the anomaly found in $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ with $x=0.125$ at the orthorhombic to low-temperature tetragonal structural phase transition around 60 K.¹²⁻¹⁵ The anomalies found in $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ are summarized as follows: (1) structural change, (2) increase in the resistivity, (3) decrease in the Hall coefficient, (4) decrease in the Seebeck coefficient, and (5) increase in the magnetic susceptibility. Among these anomalies, we reported (2) and (3) in the present study for the 1:2:3 compounds. (4) is reported by van Woerden *et al.* for $\text{Nd}_{1+x}\text{Ba}_{2-x}\text{Cu}_3\text{O}_{7+y}$.¹⁶ In the case of $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ with $x=0.125$, the anomaly in the transport properties are closely related with its structural aspect. However, various kinds of substitution experiments in the 2:1:4 system show that the transport anomaly in this system only occurs when the carrier

number per copper is around 0.125.^{17,18} These results suggest that the transition is not a simple structural transition but is driven by an electronic instability at a certain carrier density.

As we have described, the universal transport anomalies in the 1:2:3 systems were also observed in a certain range of x in both $\text{Nd}_{1+x}\text{Ba}_{2-x}\text{Cu}_3\text{O}_{7+y}$ and $\text{YBa}_2(\text{Cu}_{1-x}\text{Co}_x)_3\text{O}_{7+y}$. Trivalent Co is known to substitute for Cu in the chain site and introduce excess oxygen ions. The oxygen uptake is not sufficient to compensate the decrease in hole number due to Co substitution, and leads to a net decrease in the hole number as in the case of $\text{Nd}_{1+x}\text{Ba}_{2-x}\text{Cu}_3\text{O}_{7+y}$. On the other hand, Zn is known to substitute mainly the Cu in the plane. Since Zn is isoelectronic with Cu, it does not affect the hole number in the system. In two of the present 1:2:3 systems, in which transport anomalies were found, the anomalies were most pronounced for samples having a T_c of ~ 60 K. Using the relation between T_c and p_s (hole number per copper in the CuO_2 plane) by Takita *et al.*¹⁹ for the 1:2:3 systems, $T_c=60$ K corresponds to $p_s=0.15$. The estimate of the hole coefficient using the maximum value of the Hall coefficient gives $p_s=0.10$ for $\text{Nd}_{1.2}\text{Ba}_{1.8}\text{Cu}_3\text{O}_{7+y}$ and $p_s=0.09$ for $\text{YBa}_2(\text{Cu}_{0.95}\text{Co}_{0.05})_3\text{O}_{7+y}$. We have not determined the hole number based on the chemical analysis because the assumption by Tokura *et al.*² of the carrier distribution between plane and chain sites depends on the ordering of the oxygen in the chain sites.²⁰ In any case, the value is comparable to $p_s=0.125$ in the case of $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$. The presence of transport anomalies in two kinds of high- T_c systems with similar hole number per copper suggests that the anomalies are the inherent property of the two-dimensional CuO_2 planes.

Since high- T_c superconductors are made up of two-dimensional CuO_2 planes, most of them would have similar Fermi surfaces if the carrier numbers per copper are the same. In low-dimensional systems, Fermi surfaces are known to be susceptible to $2k_F$ instabilities, which open a gap at the Fermi surface. As we have already mentioned, we need to postulate the coexistence of hole and electron bands in order to reconcile the anomalous decrease in the Hall coefficient with the increase in the resistivity. The gap formation in the hole band would result in the decrease in the Hall coefficient. As a result of the net decrease in the carrier density, the resistivity will increase. Actually, a decrease in the density of states has recently been reported from low-temperature specific-heat measurements in $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ by Okajima *et al.*²¹ They found a decrease in the density of states around $x \sim 0.12$, where the anomaly is most pronounced.

Although no evidence of lattice distortion has been found so far in 1:2:3 systems, it may be due to the short-range nature of the ordering. We performed a preliminary low-temperature x-ray-diffraction experiment on $\text{Nd}_{1.2}\text{Ba}_{1.8}\text{Cu}_3\text{O}_{7+y}$ in order to check whether the transport anomalies found in the present study for 1:2:3 systems were also accompanied by a structural change or not. Lattice parameters were found to change continuously as the temperature was lowered without any anom-

ally around 120 K. But there still remains a possibility for the formation of a superlattice, which can be weak and short range. Experimental probes which are sensitive to the local structural change, e.g., Mössbauer spectroscopy and extended x-ray-absorption fine-structure (EXAFS) measurement, may be useful. Actually, recent Mössbauer spectroscopy data in a carrier-reduced 1:2:3 system, $\text{YBa}_2(\text{Cu}_{0.983}\text{Fe}_{0.017})_3\text{O}_{6.8}$, by Afanas'ev *et al.*²² shows a structural change around 110 K, which is close to the temperature of the transport anomaly. Although the T_c for this specimen, 82 K, is slightly higher than 60 K, the fact that the carrier-reduced sample shows this structural change suggests that it is closely related with the transport anomaly.

A possible structural change in the 1:2:3 system is suggested by Rey *et al.*²³ from the structural analysis of $\text{LaBa}_2\text{Cu}_2\text{NbO}_8$, which has the 1:2:3 structure with Y replaced by La and the CuO chain replaced by a NbO_2 plane. They showed that $\text{LaBa}_2\text{Cu}_2\text{NbO}_8$ has a modulated structure with zigzag folding of the NbO_6 octahedra and with lattice parameters of $a = \sqrt{2}a_0$, $b = \sqrt{2}b_0$, $c = 2c_0$, where a_0 , b_0 , and c_0 are the lattice parameters of the 1:2:3 system.

An anomalous isotope effect in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ with the same hole number as discussed in this paper is reported by Crawford *et al.*²⁴ This experiment demonstrates the presence of a strong electron-phonon interaction in high- T_c superconductors. Tsuei *et al.* proposed that the anomalous isotope effect is due to the presence of a van Hove-type singularity which is pinned near the Fermi surface.²⁵ The presence of the van Hove singularity in the half-filled two-dimensional band has been discussed as the mechanism of the high- T_c superconductivity and possible charge-density wave formation.^{26,27}

The transport anomaly in the 1:2:3 system is gradual compared with the sharp anomaly in the LaBaCuO system. As we suggested, if the mechanism for the anomaly is related with the two-dimensionality of the system, it is understandable by considering the difference of the anisotropy for both systems. The anisotropies of the resistivity are reported as ~ 30 and ~ 1000 for $\text{YBa}_2\text{Cu}_3\text{O}_7$ (Ref. 28) and $\text{La}_{1.86}\text{Sr}_{0.14}\text{CuO}_4$,²⁹ respectively. Unfortunately, there are no data available to compare the anisotropy for both systems when the hole number per copper is about 0.125. It is probable that the presence of the chain site in the 1:2:3 system makes the anisotropy in it lower than in the 2:1:4 system even in this carrier number range. However, it should be stressed that the two-dimensionality is not the only factor for the anomaly because it is well established that the anomaly is strongly suppressed in $\text{La}_{1.875}\text{Sr}_{0.125}\text{CuO}_4$,³⁰ which is considered to have a similar anisotropy to $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$. The difference in the phonon spectra coupled with the electronic instability may make the anomaly quite different in both compounds.

Finally, we want to comment on the anomalous $1/T$ dependence of the Hall coefficient. Among many theories, magnetic skew scattering is the most direct way

to explain the anomalous temperature dependence. The spin-orbit interaction causes an unbalance of the scattering probability of carriers to the left and the right, and produces an effective Hall voltage which is proportional to the magnetic moment. Aronov *et al.* studied this effect in the case of high- T_c materials and concluded that it could give rise to a considerable contribution compared with ordinary materials.³¹ However, the most serious problem for this theory is the origin of the spin-orbit interaction, because the static magnetic susceptibility in $\text{YBa}_2\text{Cu}_3\text{O}_7$ is known to be almost constant above T_c . Incidentally, it is interesting to note a similarity of the temperature dependence of the static susceptibility at the antiferromagnetic wave vector ($\chi(Q)$, $Q = (\pi, \pi)$) for 1:2:3 systems as determined by analyses of nuclear spin-relaxation experiments.³² Namely, $\chi(Q) \propto 1/T$ for $\text{YBa}_2\text{Cu}_3\text{O}_7$ and $\chi(Q)$ shows a maximum around ~ 150 K for $\text{YBa}_2\text{Cu}_3\text{O}_{6.63}$ with a T_c of 60 K.³² While such a similarity could be accidental, it might give a clue to the origin of the anomalous temperature dependence of the Hall coefficient. Admittedly, however, how the antiferromagnetic susceptibility, rather than the uniform susceptibility, can be reflected in the dc transport is not clear at all.

CONCLUSIONS

We have found a universal transport anomaly in the 1:2:3 compounds when the hole number per copper is 0.1–0.15. The anomaly consists of a decrease in the Hall coefficient at ~ 120 K accompanied by an upturn of the resistivity as temperature is lowered below 120 K. It calls for the presence of two bands with different polarity. The opening of a gap at the hole band can explain the anomaly consistently. The fact that the anomaly occurs in both the 1:2:3 and the 2:1:4 systems at a similar carrier number per copper suggests that it reflects an intrinsic instability of the two-dimensional CuO_2 planes at this carrier number. A similarity of the temperature dependence of the Hall coefficient and the antiferromagnetic component of susceptibility was pointed out. To check possible origins of the anomaly, including gap formation, optical or tunneling spectroscopy would be valuable. A search for a low-temperature structural change by electron-diffraction, x-ray-diffraction, neutron-diffraction, ultrasonic, Mössbauer spectroscopy, and EXAFS measurements would be useful to clarify the universality of the anomaly.

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