

Transport Anomalies and the Role of Pseudogap in the 60-K Phase of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$

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We report the result of our accurate measurements of the a - and b -axis resistivity, Hall coefficient, and the a -axis thermopower in untwinned $\text{YBa}_2\text{Cu}_3\text{O}_y$ single crystals in a wide range of doping. It is found that both the a -axis resistivity and the Hall conductivity show anomalous dependences on the oxygen content y in the 60-K phase below the pseudogap temperature T^* . The complete data set enables us to narrow down the possible pictures of the 60-K phase, with which we discuss a peculiar role of the pseudogap in the charge transport.

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$\text{YBa}_2\text{Cu}_3\text{O}_y$ (YBCO) is one of the most intensively studied systems among the high- T_c cuprates. Although it is known that an increase in the oxygen content y from 6 to 7 causes the hole doping into the CuO_2 planes and leads to superconductivity, the dependence of T_c on y is nontrivial and there is a plateau at $T_c \approx 60$ K with y of around 6.7 ("60-K plateau" or "60-K phase"). The origin of this plateau remains controversial and two different explanations have been discussed: one [1] is to consider (partial) oxygen ordering in the Cu-O chain layers and asserts that the average valence of the Cu ions (and thus the hole concentration n) in the CuO_2 planes is unchanged in a certain range of y ; the other [2,3] assumes that n is continuously changing and relates the plateau to the "1/8 anomaly," which is a suppression of T_c at the hole doping of 1/8 per Cu due to a charge-density-wave instability (in other words, charged stripe formation [4]). Because of this controversy and also because of the difficulty in determining the actual hole concentrations in the CuO_2 planes, the understanding of the normal and superconducting states of underdoped YBCO remains far from satisfactory, which has been a source of extra complications in elucidating the high- T_c mechanism.

The difficulty in clarifying the origin of the 60-K plateau lies partly in the essentially inhomogeneous nature of the oxygen distribution in the Cu-O chain layers [5]; note that for a given y the actual hole doping can differ depending on the arrangement of the O atoms [6], and the O atoms in the Cu-O chains can rather easily rearrange at room temperature, which causes the room-temperature (RT) annealing effect [7]. These facts often make it difficult to achieve reproducibility in the experiments, and therefore very careful experiments armed with high-quality samples are necessary for obtaining reliable results. Also, when measuring the in-plane transport properties, the use of untwinned crystals is indispensable for extracting the intrinsic behavior of the CuO_2 planes. Previous transport studies of untwinned YBCO crystals (e.g., Refs. [8,9]) did not provide enough information on the y dependence in the 60-K plateau region to discuss its origin.

In this Letter, we report the result of our careful study of the a -axis resistivity ρ_a of high-quality untwinned YBCO

single crystals for a wide doping range, where we paid particular attention to determining the absolute magnitude of the resistivity to the accuracy of 5%. Since the Cu-O chains run along the b axis, in the ρ_a measurements the electric current flows only in the CuO_2 planes, and thus ρ_a is not complicated with the conductivity of the chains [10]. We also measure the b -axis resistivity ρ_b , Hall coefficient R_H , and the a -axis thermopower at 290 K, $S_a(290\text{ K})$, all of which help elucidating the transport anomalies in the 60-K phase as a function of y . We found that ρ_a becomes remarkably independent of y in the 60-K phase when the pseudogap opens, indicating that one of the following two situations is realized: (i) the hole concentration in the planes, n , is essentially unchanged, or (ii) a change in n is compensated by a change in the scattering time. Both possibilities bear intriguing implications on not only the peculiar electronic state in the 60-K phase but also the role of the pseudogap in the charge transport.

The YBCO single crystals are grown in Y_2O_3 crucibles by a conventional flux method [11]. The high purity of our crystals can be inferred from the optimum zero-resistance T_c , which is as high as 93.4 K observed for $y = 6.95$ (the transition width is less than 0.5 K). Before detwinning, the crystals are annealed to be tuned to the targeted oxygen content (both the annealing atmosphere and temperature should be varied to best tune the oxygen content over a wide range). The crystals reported here are in the range of $y = 6.45$ –7.0. The crystals are always quenched at the end of the high-temperature annealing. Detwinning is performed at temperatures below 220 °C under a uniaxial pressure of ~ 0.1 GPa while monitoring the crystal surface with a polarized-light microscope. We measure only samples that are perfectly detwinned. The exact oxygen content is determined by iodometry.

After the preparations are finished, the samples are left at room temperature for at least a week for the oxygen arrangement to equilibrate. (We do observe the RT annealing effect in the time scale of a few days, which slightly reduces the resistivity and increases T_c .) Note that during the RT annealing the total oxygen content y do not change, but the oxygen atoms tend to order locally to form longer Cu-O chains. To check for the impact of the order

of annealing/detwinning, we prepared both predetwinned (annealed after detwinning) and postdetwinned (reverse order) crystals for $y = 6.65, 6.75,$ and 6.80 ; differences in ρ_a and R_H between the pre- and postdetwinned crystals are confirmed to be within the experimental error. This indicates that the extent of the oxygen ordering is reproducible in our samples as long as the annealing conditions are unchanged.

Measurements of ρ_a (ρ_b) are done with a standard ac four-probe method using samples that are at least 2 times longer in the a (b) direction. (We note that it is easier to detwin crystals for ρ_a measurements, for which the uniaxial pressure is applied to a smaller cross-sectional area.) The Hall data are taken by sweeping the magnetic field to both plus and minus polarities at fixed temperatures. All the R_H data shown here are measured with the electric current along the a axis, and we confirmed that R_H is essentially identical when the current is along the b axis (i.e., Onsager's relation holds) [12]. The thermopower is measured with a standard steady-state technique with a reversible temperature gradient of ~ 1 K.

First we demonstrate the reproducibility and the accuracy of our measurement in Fig. 1, which shows the result of $\rho_a(T)$ measurements on three different samples, A, B, and C, all at $y = 6.75$. As depicted in the inset of Fig. 1, the voltage is measured on the two sides of the crystals, so each sample yields two sets of $\rho_a(T)$ data; this is a good practice for checking the homogeneity of the sample. The thickness of the crystals is accurately determined by measuring the weight with $0.1 \mu\text{g}$ resolution. The largest source of error is the separation between the voltage contacts, which causes the uncertainty in the overall magnitude; use of a long sample can reduce this error

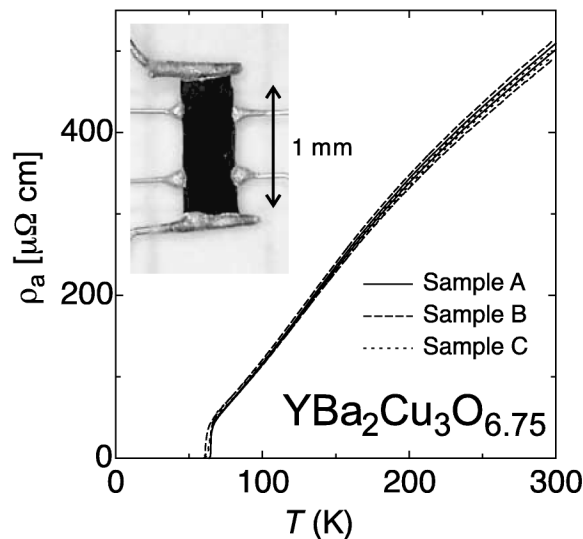


FIG. 1. Six data sets of $\rho_a(T)$ measured on three samples at $y = 6.75$, which represent the reproducibility and accuracy of our measurements. Inset: Photograph of a typical sample with contacts.

to about 5%, because our voltage contacts are defined by narrow gold pads whose width is $\sim 50 \mu\text{m}$. Typically, our samples have the voltage-contact separation of $0.6\text{--}1$ mm, width of 0.3 mm, and the thickness of $60 \mu\text{m}$. As can be understood from Fig. 1, the reproducibility of the $\rho_a(T)$ data is very good and the scatter of the magnitude is consistent with the estimated error. T_c is also very reproducible, and its variation for a given y is less than 5 K. We note that $\rho_a(T)$ is measured on at least three crystals for each composition and the data are reproducible within the same order of accuracy as is demonstrated in Fig. 1. We pick up the data which sit in the middle of the spectrum to be representative of a given composition.

Figure 2 shows the temperature dependence of ρ_a for the oxygen contents $y = 6.45\text{--}7.0$; this figure is a summary of the measurements of more than 30 samples. The $y = 7.0$ crystal ($T_c = 92.0$ K) is slightly overdoped, while the $y = 6.95$ crystal ($T_c = 93.4$ K) is optimally doped and shows a strictly linear T dependence with a negative intercept. In the underdoped region, our samples show essentially similar behavior as was reported before [8,9]; however, what is remarkable here is that the $\rho_a(T)$ data for $y = 6.65\text{--}6.80$ show clear overlap below ~ 130 K. Note that in the underdoped YBCO the pseudogap opening can be inferred from a downward deviation from the high-temperature T -linear dependence below T^* [8], and thus the data in Fig. 2 suggest that *the overlapping of $\rho_a(T)$ is observed in the pseudogapped state*, although T^* inferred from the data is already above 300 K for $y = 6.65$. Figure 3 shows the corresponding evolution of $\rho_b(T)$ with y . Note that ρ_b is generally smaller than ρ_a for the same y , which is believed to be

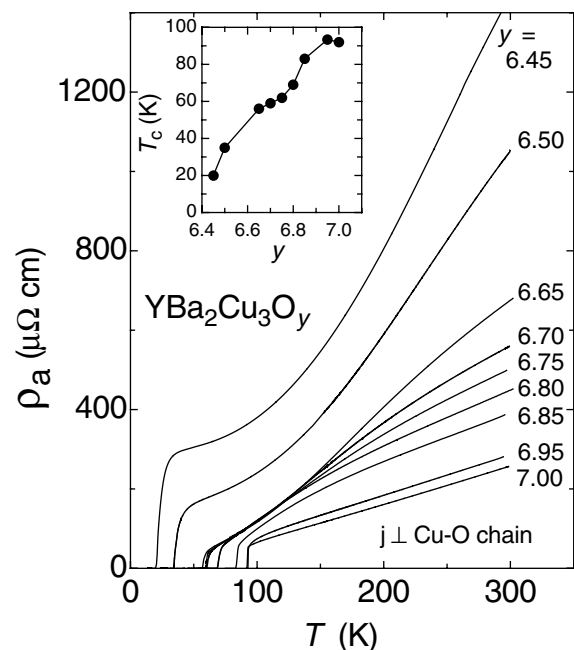


FIG. 2. T dependences of ρ_a for untwinned YBCO crystals in 0 T. Inset: Phase diagram of zero-resistance T_c vs y .

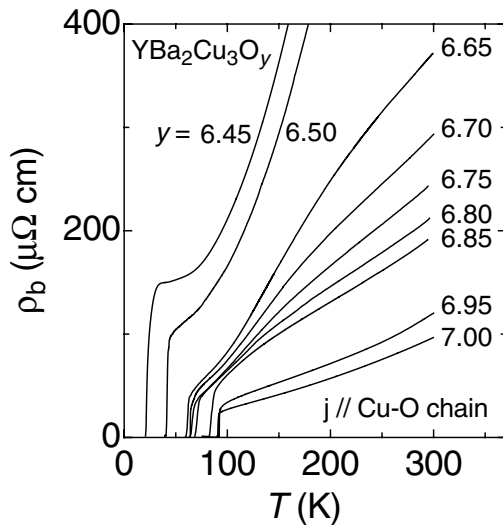


FIG. 3. T dependences of ρ_b for untwinned YBCO crystals in 0 T. Note the different scale compared to Fig. 2.

due to the finite chain conductivity [10]. The $\rho_b(T)$ data do not show as clear overlap in the 60-K plateau region as the $\rho_a(T)$ data.

The Hall channel in the in-plane transport is also found to show an anomaly in the 60-K phase, which can be seen in the Hall conductivity σ_{xy} . In YBCO, it is expected that the Cu-O chains contribute little to σ_{xy} because of their one dimensionality, so σ_{xy} is governed by the properties of the planes. On the other hand, the Hall resistivity ρ_{xy} is expressed as $\rho_{xy} \approx \sigma_{xy}/(\sigma_{xx}\sigma_{yy})$ [13] (where $\sigma_{xx} \approx 1/\rho_a$ and $\sigma_{yy} \approx 1/\rho_b$ in our case) and thus the Hall coefficient $R_H (= \rho_{xy}/B)$ reflects the properties of not only the planes but also the chains. Therefore, σ_{xy}

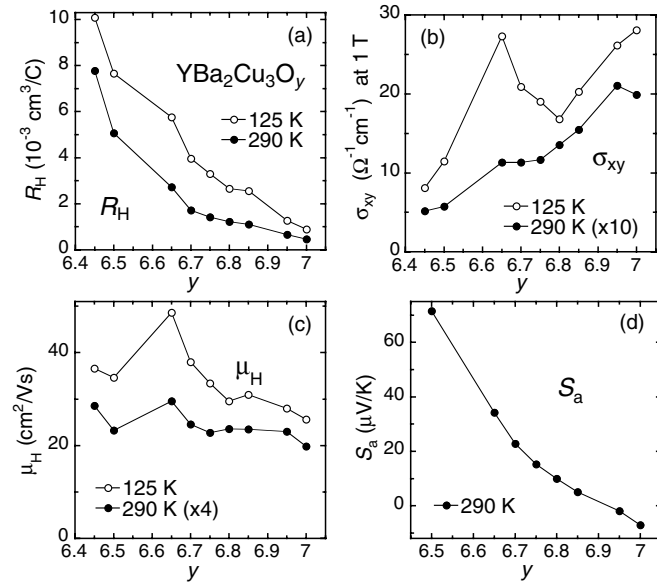


FIG. 4. y dependences of (a) raw R_H , (b) Hall conductivity σ_{xy} (calculated for $B = 1$ T), (c) Hall mobility μ_H , at 125 and 290 K, and (d) a -axis thermopower S_a at 290 K.

is a better indicator of the properties of the planes compared to R_H [14]. The raw R_H and the calculated σ_{xy} [which is well approximated by $\rho_{xy}/(\rho_a\rho_b)$] are shown in Figs. 4(a) and 4(b), respectively, as functions of y for 125 and 290 K. In Fig. 4(b), a nonmonotonic y dependence of σ_{xy} is apparent at 125 K [where $\sigma_{xx} (\approx 1/\rho_a)$ is unusually y independent], while the raw R_H data [Fig. 4(a)] show relatively smooth change with y . (Detailed account on the Hall data of our untwinned YBCO will be published elsewhere [12].) The nature of this anomaly in the Hall channel is probably best understood by the plot of the Hall mobility in the planes, $\mu_H = \sigma_{xy}/(B\sigma_{xx})$ [Fig. 4(c)], which in principle does not include n . One can clearly see that μ_H is anomalously enhanced near $y = 6.65$, particularly at 125 K (in the pseudogapped state).

It is generally believed that the room-temperature thermopower $S(290$ K) reflects the change in the hole concentration and thus may be used as a guide to estimate n [2]. Figure 4(d) shows the y dependence of the thermopower measured along the a axis at 290 K, $S_a(290$ K). The $S_a(290$ K) data show a continuous change across the 60-K phase, which is suggestive of n changing with y ; however, we cannot draw a definite conclusion from this, because there is a possibility that the density of states of the Cu-O chains gives some contribution to S_a .

Now let us discuss the implications of the above results. First, we note that the y independence of ρ_a (observed below ~ 130 K in the 60-K phase) has two possible origins: (i) both the carrier concentration n and the scattering time τ remain unchanged with y , or (ii) a change in n is compensated by a change in τ . Since it is difficult to conclusively identify the origin based on the data presented here, we should fully discuss the possible nature of the 60-K phase for the two cases.

If case (i) is true, we must understand why at high temperatures ρ_a changes with y . In Fig. 2, it looks like the $\rho_a(T)$ data for $y = 6.65$ – 6.80 gradually converge to a single curve as the temperature is lowered below T^* ; this trend can be interpreted to mean that there is some y -dependent scattering above T^* that is gradually wiped out in the pseudogapped state. Remember that the pseudogap is a partial gap near $(\pi, 0)$ and $(0, \pi)$ on the Fermi surface [15], and therefore any scattering that is concentrated near these regions of the Fermi surface is expected to be wiped out when the pseudogap opens. Since the scattering caused by the AF fluctuations is concentrated in the “hot spots” [16], which correspond to the gapped regions in the pseudogapped state, the magnetic scattering could cause ρ_a to be y dependent above T^* even when τ remains unchanged with y below T^* .

If this is really the case, the strength of the magnetic fluctuations must be changing with y in the 60-K phase while n stays unchanged; we can construct an argument for this by recalling the role of the oxygen in the Cu-O chains. Within the oxygen ordering scenario [1], an addition of oxygen converts Cu^{1+} on the chains into Cu^{2+} ,

adding spins onto the chain layers instead of adding holes into the CuO_2 planes in the 60-K phase. These Cu spins on the chains mediate the magnetic coupling between the bilayers [17], causing a suppression of the AF fluctuations in the CuO_2 planes. We can thus argue for case (i) that the increase in y may lead to weaker AF fluctuations, which cause the quasiparticles in the hot spots to be scattered less, leading to a decrease in $\rho_a(T)$ at high temperatures. Note that the peak in μ_H at $y = 6.65$ seems to be inconsistent with the assumption of y -independent τ below T^* , but this inconsistency might be resolved by considering a scattering-time separation [18,19].

Next, if case (ii) is true, the transport properties demonstrate an unusual variation of the scattering events in the 60-K phase. In this case, we can interpret that μ_H directly reflects the variation of τ with changing n ; namely, we can infer that τ at 125 K is notably enhanced as y is decreased from 6.8 to 6.65 in the 60-K phase. If this enhancement in τ compensates a decrease in n , ρ_a becomes y independent as is observed in Fig. 2. Therefore, in this scenario, the anomalous y dependence of μ_H and the unusual overlap of $\rho_a(T)$ have a common physical origin. The fact that the compensation is visible only below T^* indicates that the anomalous enhancement of τ takes place only in the pseudogapped state, which suggests that this anomaly is of electronic origin. This observation reveals a novel aspect of the pseudogap in YBCO, which might help elucidating the origin of the pseudogap itself. We note that the y dependence of $S_a(290 \text{ K})$ seems to support this scenario, although it is not conclusive.

Within the picture of case (ii), the 60-K plateau in T_c may be understood to be caused by the anomalous y dependence of μ_H in this doping region, because T_c is expected to be reduced when the carrier scattering is enhanced (which leads to smaller μ_H). Therefore, if case (ii) is true, the 60-K phase corresponds to a particular doping region where τ in the pseudogapped state is anomalously enhanced as n is decreased, and the peculiar features in ρ_a , σ_{xy} , and T_c can essentially be understood by the unusual doping dependence of τ . If, on the other hand, the oxygen ordering causes the case (i) to be realized, the 60-K phase of YBCO offers a unique case where the hole concentration is essentially unchanged while the strength of the AF fluctuations keeps changing with y ; within this scenario, the plateau in T_c suggests that the charge carriers are *protected* from the bare AF fluctuations once the pseudogap opens and the occurrence of superconductivity is governed by n . In both cases, our data bear fundamental implications on the electronic state in YBCO.

Lastly, we comment that our result does not exclude the possibility that the hole doping happens to be $1/8$ per Cu in the planes for $y \approx 6.7$. In view of the reports that suggest the existence of $1/8$ -anomaly-like features in Ca-doped

YBCO [2,3], it is perhaps sensible to assume that both an unusual electronic state and the $1/8$ doping are realized in the 60-K plateau region.

In summary, we found a remarkable overlap of the $\rho_a(T)$ data of untwinned $\text{YBa}_2\text{Cu}_3\text{O}_y$ crystals in the 60-K phase below $\sim 130 \text{ K}$. Moreover, the transport in the Hall channel is also found to show anomalous behavior in the same region. We discuss that two different scenarios can potentially explain the observed anomalies, and in both scenarios the pseudogap appears to play a key role; in any case, it is clear that an unusual electronic state is responsible for the anomalies in the 60-K phase. Further studies of this unusual region is clearly desirable, and the set of data presented here would serve as a touchstone to test any model for the 60-K phase.

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