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Diverging resistivity anisotropy with decreasing temperature in 60-K $YBa_2Cu_3O_{7-y}$

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The in-plane resistivity ρ_{ab} of 60-K $YBa_2Cu_3O_{7-y}$ is equal to (340 ± 30) $\mu\Omega$ cm at 290 K and varies approximately linearly with temperature T. The anisotropy ρ_c/ρ_{ab} increases from ~ 100 to 2000 as T decreases from 290 to 80 K. In the four crystals studied ρ_c is observed to increase monotonically with decreasing T. This is an intrinsic property of the normal state which has not been explained by conventional transport theory.

In the high-T_c oxides the two-dimensional nature of the transport properties is one of the striking anomalies of the normal-state properties. Recent studies[1–4] have shown that the conductivity anisotropy is much greater than anticipated by band-structure calculations, or indicated by optical measurements.[5] In 90-K crystals of $YBa_2Cu_3O_{7-y}$ (1:2:3) Hagen *et al.*[2] report that the anisotropy of the resistivity ρ_c/ρ_{ab} varies from ~ 80 at 290 K to ~ 250 near T_c, whereas "realistic" band-structure calculations[6] predict 10 to 20. In "two-plane" $Bi_2Sr_2CaCu_2O_{8+y}$ (Bi 2:2:1:2), Martin *et al.*[3] find that ρ_c/ρ_{ab} grows to 10^5 as T decreases.

Even more interesting than the large anisotropy is the disparity in sign of the derivatives $d\rho_c/dT$ and $d\rho_{ab}/dT$. As is well known, the in-plane resistivity ρ_{ab} is linear (or very close to linear) in the hole-type superconductors built up of CuO_2 planes. In contrast, the out-of-plane resistivity ρ_c increases as T decreases. This was first observed by Tozer *et al.*[1] for 90-K 1:2:3, and confirmed by Hagen *et al.*[2] These results were contested by other groups[7,8] who report a strictly metallic variation in ρ_c in their "best" crystals.

The clarification of this point is important because it is clearly not possible within normal Fermi-liquid theory and the usual Bloch-Boltzmann approach to explain such a disparate T dependence in the two directions. The qualitatively different T dependence and the divergence of the ratio ρ_c/ρ_{ab} may be signatures of the peculiar normal state of these oxides, so it is important to investigate this point systematically. We have now extended the study of the resistivity anisotropy to the 60-K version $YBa_2Cu_3O_{7-y}$, with the goal of studying the temperature dependence of both ρ_c and ρ_{ab}, and also to compare accurately the value of ρ_{ab} in the 60- and 90-K states of 1:2:3.

Following the procedure worked out for the ceramic samples,[9] we have reduced the oxygen content of crystals of the 90-K phase, which are grown using a BaO-CuO flux.[10] The starting crystals all have very sharp transitions (measured both resistively and magnetically) between 91 and 93 K, and have been extensively studied.[2,10] ρ_{ab} is approximately 150 $\mu\Omega$ cm at 290 K while ρ_c is of the order of 10 mΩ cm^2. Below 150 K, ρ_c shows a slight increase as T decreases towards T_c. To obtain the 60-K system, the 90-K crystals were reduced in an Ar atmosphere mixed with 8% O_2 at a temperature of 573 °C for between 7 to 13 days. The ac susceptibility shows a superconducting transition between 60 and 70 K, and no trace of superconducting anomalies at higher T. The width of the transition is, however, much wider than in the original crystals, with the resistivity onset as high as 80 K (Sample 5). We note that, by starting with good homogeneous 90-K crystals, and reducing down to the 60-K phase, we arrive at an oxygen distribution which, if inhomogeneous, would be higher in the core. However, because of our long anneal times, and the susceptibility and resistivity results, we have reason to believe that the oxygen distribution is homogeneous. Fixing the annealing temperature and percentage of O_2 in the reducing gas, we have doubled the annealing time and found very similar values for ρ_{ab} (Samples 4 and 5). In earlier attempts executed at lower annealing T or with shorter annealing times, we could detect a diamagnetic step between 80 and 90 K which indicated that part of the core remained in the O_7 phase. Large decreases in ρ_{ab} above 80 K were also present in the partially reduced samples. Thick crystals (>80 μm along c) also tend to show such evidence of incomplete reduction. Our annealing process has been selected to achieve an oxygen content of 6.5–6.6 per unit cell. However, technical difficulties prevent the determination of the actual oxygen content in each crystal.

We used the Montgomery method[11] to measure ρ_{ab} and ρ_c separately. In some crystals we also used the van der Pauw method[11] to measure ρ_{ab}. Indium solder was used to attach the leads, but because the surface quality is quite

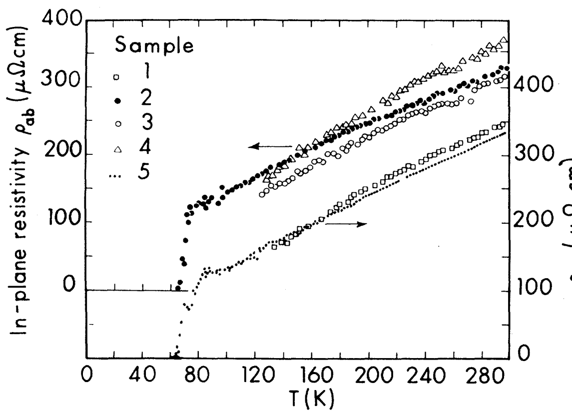

FIG. 1. The temperature dependence of ρ_{ab} in five samples of $YBa_2Cu_3O_{7-y}$ reduced to the 60-K phase. Samples 1–4 are measured using Montgomery's method. Sample 5 is measured using the van der Pauw technique. ρ_{ab} at 290 K lies between 315 and 370 $\mu\Omega$ cm. Data points for Samples 1, 3, and 4 are not reliable below 120 K because R_1 is suppressed below our resolution (see text). Samples 1–5 were annealed at 573 °C for 8, 8, 10, 7, and 13 days, respectively.

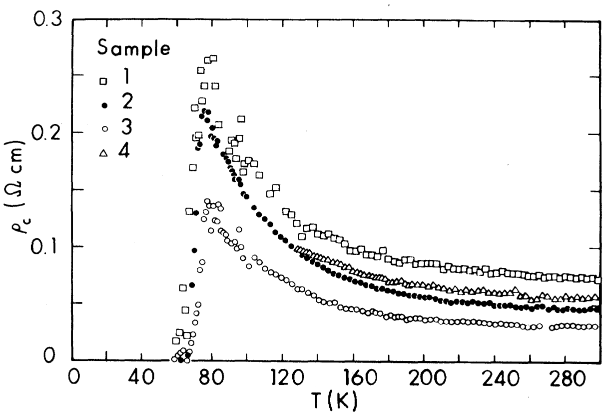

FIG. 2. Variation of ρ_c with T in four 60-K crystals of $YBa_2Cu_3O_{7-y}$, showing monotonic increase with decreasing T. The increased scatter below 120 K is caused by suppression of R_1.

poor after reduction, the contact resistance (50–200 Ω) is much higher than in the 90-K crystals. Gentle scratching of the surface with a needle often improved the contacts. One of the major difficulties in this study is caused by the well-known exponential suppression of the lower resistance in the Montgomery technique.[2] This was a less serious problem in the 90-K phase because the anisotropy is smaller. Here, the larger anisotropy often suppresses the resistance in the direction nominally parallel to the CuO_2 planes to the extent that it becomes unobservable. Thus, in some crystals, the data for ρ_{ab} only extend to 120 K. The van der Pauw method has been used to supplement our results for ρ_{ab} since it is free of this restriction. In all measurements the current is reversed to eliminate the contribution of thermal emf.

Figure 1 shows the temperature dependence of the in-plane resistivity for the five samples studied. ρ_{ab} is close to being linear in T in all samples, although it extrapolates to a finite value at $T=0$. In Samples 1, 3, and 4 the suppression of the in-plane potential drop is severe, so that the calculated ρ_{ab} shows very large fluctuations below 120 K. In all samples, the room-temperature value of ρ_{ab} is between 320 and 370 $\mu\Omega$ cm (average over the five samples is 340 $\mu\Omega$ cm). In view of the different measuring techniques, we consider this to be a fairly reliable estimate of the absolute resistivity in the 60-K phase. The value for the 90-K phase is 150 $\mu\Omega$ cm at 290 K.

The variation in the out-of-plane resistivity value is larger, ranging from 30 to 75 mΩ cm at 290 K (compared with $\sim$10 mΩ cm for the 90-K crystals). However, all four crystals studied show a strongly increasing ρ_c as T decreases. In Samples 1, 2, and 3 ρ_c goes through a sharp peak near 80 K before decreasing to zero (see Fig. 2). We regard the peak as spurious, since the onset of superconducting behavior near 80 K greatly alters the current distribution, making the Montgomery separation of the contributions to the observed resistances highly unreliable.

Previous studies of the electrical anisotropy of 1:2:3 crystals focused on getting oxygen into highly deficient samples. The reduction of the peak in the ρ_c vs T profile with increasing oxygen content led some groups[7,8] to conclude that the peak was associated with an insulating core. We will argue that this interpretation is incorrect. By starting with crystals with ρ_{ab} much lower than those obtained even after long oxygen annealing, we reduce the oxygen content. This leads to an oxygen concentration which, if not uniform, must be larger in the core and smaller at the two *ab* faces. If we attribute the observed "semiconducting" behavior of ρ_c to one (or more) *uninterrupted* layers of insulating phase between the two faces, these layers must lie at the surface, i.e., directly under the contact pads. In the Montgomery technique the resistance is calculated with the current first nominally parallel, and then perpendicular, to the *a*-*b* face (R_1 and R_2 respectively). It is clear that the putative existence of the insulating layers at the surface must affect both R_1 and R_2, so that ρ_c and ρ_{ab} will show semiconducting behavior, in disagreement with our results. (This argument is inapplicable if the layer is in the core.) The observation of the same qualitative ρ_c vs T profile in four samples (with annealing times spanning 7 to 13 days) also makes it highly unlikely that the data are distorted by a pathological current distribution. We conclude that the ρ_c vs T profile is reproducible, and an intrinsic property of the 60-K phase. Whereas in the 90-K phase the upturn in ρ_c only occurs below 150 K, $d\rho_c/dT$ here is negative even at 290 K. The magnitude of the anisotropy is also larger in the present system, increasing from $\sim$100 at 290 K to over 2000 at 80 K (Fig. 3). These numbers suggest that by removing oxygen from the chains, the system becomes more "two-dimensional," and the qualitatively different behaviors of ρ_c and ρ_{ab} become more transparent. The rapid divergence of ρ_c with decreasing T implies the remarkable result that as $T \rightarrow 0$, the system tends to an insulator in the out-of-plane direction while the in-plane conduction remains metallic. The onset of superconduc-

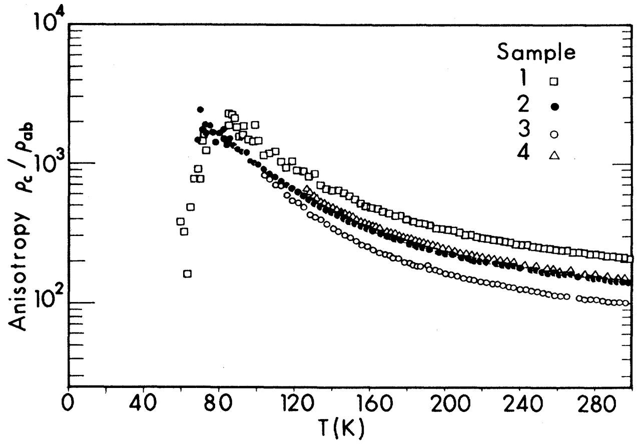

FIG. 3. The anisotropy of the resistivity in four crystals of 60-K $YBa_2Cu_3O_{7-y}$.

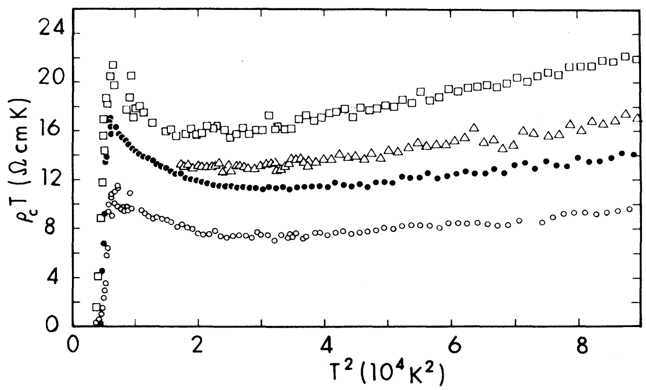

FIG. 4. Plot of $\rho_c T$ vs T^2 for four crystals of 60-K $YBa_2Cu_3O_{7-y}$ to compare data with the expression $\rho_c = AT + B/T$. Symbol identification is the same as in Fig. 3.

tivity interrupts this trend, converting the system to a superconductor with sizable "effective-mass" anisotropy, but nowhere comparable with ρ_c/ρ_{ab} just above T_c.

As previously discussed for the 90-K phase, the different signs of $d\rho_c/dT$ and $d\rho_{ab}/dT$ in the high-T_c oxides is unusual (if not unique) in highly anisotropic metals. There appears to be no way to account for such disparate behavior in conventional Bloch transport theory, even if one presupposes disorder leading to localization along c. The present study shows that ρ_c rises much more dramatically with decreasing T in the 60-K phase compared with either the 90-K phase[2] of 1:2:3 or Bi 2:2:1:2,[3] so that the difficulties with band-structure approaches are even more severe. Rather than the usual conventional Bloch-Boltzmann theory, the out-of-plane transport calls for a more sophisticated theory. (This is also the case for the thermopower[12] along c.) Anderson and Zou[13] proposed a mechanism based on the merging of spinons and holons which leads to a $1/T$ behavior for ρ_c. The calculation itself has been criticized by Kallin and Berlinsky.[14] However, it remains of interest to check if ρ_c obeys the phenomenological expression $\rho_c = AT + B/T$. In Fig. 4 we plot $\rho_c T$ vs T^2 to compare the present results with the 90-K data. The plot shows that the increase in ρ_c below ~140–200 K is steeper than $1/T$. Above 140–200 K, however, the data fall on a straight line as previously found for the 90-K system. The comparison shows that the phenomenological expression, as it stands, fails to describe the behavior of ρ_c over the whole temperature range. The deviation suggests a new process operating below 140–200 K, or that an entirely different expression is needed.

We note that the conductivity along c (0.2–0.3 Ω cm at 80 K) is 20 to 30 times smaller than the Mott-Ioffe-Regel (MIR) limit. Why is the conductivity along c so poor? Explanations based on Anderson localization[15] along c are unconvincing. To our knowledge, the coexistence of localization along c, with intraplane itinerant behavior has not been theoretically demonstrated for an anisotropic metal. To explain the present data, we would require very strong disorder along c (to produce the steep increase in ρ_c). Yet, this disorder should not produce potential fluctuations strong enough to cause in-plane localization. We judge it highly unlikely that this highly specific pattern of disorder has been realized in the four annealed crystals. In any case, these are not strongly disordered solids (the in-plane conductivity is more consistent with a clean crystal).

The evidence points instead to a mechanism intrinsic to the unusual electronic states which strictly confines the charge carriers to each CuO_2 plane. In the normal state, interplane transport occurs only when the confining mechanism is violated, for instance, by strong thermal fluctuations at elevated T. We believe that the strictly monotonic increase of ρ_c as T decreases requires the existence of such a confining mechanism. In Zou and Anderson's theory the mechanism is the specific nature of the holes which can only exist in the planes. Interplane charge transfer requires scattering from spin excitations, which increase with T. Thus, increasing T leads to an enhancement of the out-of-plane transport, *and* a degradation of the inplane conduction. (From recent studies of the thermal conductivity anisotropy, Hagen, Wang, and Ong[16] find that the electron-phonon coupling along c is very weak, so that phonons are not a likely candidate for the out-of-plane scattering.)

In summary, we have found that the out-of-plane resistivity increases with decreasing T in the 60-K phase of $YBa_2Cu_3O_{7-y}$. The larger anisotropy (~2000 at 80 K) is consistent with the removal of oxygen from the chain sites. The increase is much more dramatic than in the 90-K phase.[1,2] Iye *et al.*[7] found that their crystals with higher T_c's have a metallic ρ_c while those with lower T_c's have a larger and "semiconducting" ρ_c. The general trend of $d\rho_c/dT$ vs y in Ref. 7 is consistent with our finding, whereas the T dependence of ρ_c in the $y = 7$ phase remains controversial.[17] Because of the conditions of sample preparation in the present work we are persuaded that the negative sign of $d\rho_c/dT$ is an intrinsic property of the 60-K phase, rather than the result of "bad samples." The evidence in favor of an *intrinsic* difference in signs for $d\rho_c/dT$ and $d\rho_{ab}/dT$ in the superconducting oxides based on CuO_2 is now much stronger. This unusual property may offer an important clue to the nature of the normal state.

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[17]It is instructive to compare Sample A of Iye *et al.* (Ref. 7) with the data of Ref. 2 on 90-K crystals. In both studies, ρ_c is ~ 10 mΩ cm at 100 K which is close to the MIR limit. However, in Iye *et al.*, the putative metallic behavior of ρ_c forces it to attain *twice* this value at 290 K with no sign of saturation. In Ref. 2, ρ_c is weakly T dependent, but shows a distinct increase with decreasing T near T_c. We suggest that there is significant contribution of the in-plane ρ_{ab} to Iye's measurement of ρ_c. Recent work on $Nd_{2-x}Ce_xCuO_{4-y}$ crystals in which the anisotropy exceeds 10^4 persuades us that, due to such contamination, it is easy to measure an apparently metallic ρ_c even when the deduced magnitude exceeds 5 Ω cm (500 times the MIR limit).