PHYSICAL REVIEW B VOLUME 56, NUMBER 18 1 NOVEMBER 1997-II

Anisotropic resistivity of $YBa_2Cu_4O_8$: Incoherent-to-metallic crossover in the out-of-plane transport

N. E. Hussey,* K. Nozawa, and H. Takagi
Institute for Solid State Physics, University of Tokyo, Roppongi 7-22-1, Minato-ku, Tokyo 106, Japan

S. Adachi and K. Tanabe
Superconducting Research Laboratory, International Superconductivity Technology Center, Shinonome 1-10-13, Koto-ku, Tokyo 135, Japan

(Received 5 May 1997; revised manuscript received 14 July 1997)

The resistivities of high-quality single crystals of $YBa_2Cu_4O_8$ have been measured along all three crystallographic axes up to 450 K. Both the in-plane and out-of-plane resistivities show a distinct change in behavior on cooling from high temperatures. The chain resistivity $\rho_{\rm chain}(T)$, by contrast, obeys a T^2 law over the entire temperature range. The c-axis resistivity $\rho_c(T)$ is almost T independent at high T, but as the temperature is reduced, the slope $d\rho_c/dT$ gradually increases and the c-axis transport crosses over from incoherent to metallic, implying the onset of *three*-dimensional conductivity at low T. These measurements reveal that the nonmetallic behavior of $\rho_c(T)$ in underdoped cuprates is *not* universal. [S0163-1829(97)50242-6]

It is now well established that underdoped high-T_c cuprates are characterized by the presence of a pseudogap E_g in the normal-state excitation spectrum.[1] This pseudogap, or normal-state gap, has been observed by a variety of experimental probes and is found to affect many normal-state properties in an unusual and complex way. For underdoped $YBa_2Cu_3O_{7-\delta}$,[2,3] for example, the in-plane resistivity $\rho_{\rm plane}(T)$ shows marked deviations from the high-T T-linear behavior below a characteristic T^*, well above T_c, whose value decreases with increasing hole concentration p in a manner similar to $E_g(p)$. In contrast, $\rho_c(T)$ *increases* sharply below T^*.[2,3] This increase in $\rho_c(T)$ appears to be correlated with the development of a pseudogap in the out-of-plane optical conductivity $\sigma_c(\omega)$ which suppresses the low-frequency spectral weight and transfers it to a higher ω.[4,5] However, in all underdoped cuprates where $\rho_c(T)$ is nonmetallic below T^*, doping introduces considerable disorder between the CuO_2 planes (e.g., oxygen vacancies and/or cation substitution), which may have a significant effect on the out-of-plane conductivity, yet the importance of which has so far been rather overlooked.

Underdoped $YBa_2Cu_4O_8$ ($T_c=78$ K) is naturally stoichiometric and in principle can be prepared with negligible interplane disorder. It is also untwinned, thus allowing the possibility to distinguish the effects of the normal-state gap on both the plane and chain charge carriers. Evidence for the normal-state gap has been observed in the spin-lattice relaxation rate,[6] Knight shift,[7] in-plane resistivity,[8] and thermopower[9] with 160 K$\leqslant T^* \leqslant$250 K. A reduction in the in-plane scattering rate[10] and a suppression of $\sigma_c(\omega)$ (Ref. 11) have also been observed in optical spectra below a characteristic frequency ω^*. Recently, however, Zhou *et al.*[12] reported a *maximum* in the c-axis resistance $R_c(T)$ around $T=140$ K, which appears to contradict the observation of a suppression of $\sigma_c(\omega)$ at low ω.

Here we report the resistivities of high-quality $YBa_2Cu_4O_8$ single crystals along all three crystallographic axes up to 450 K. The quality of the crystals is confirmed by the observation of a negligibly small residual resistivity for $\rho_{\rm chain}(T)$. We find that the maximum in $\rho_c(T)$ reported by Zhou *et al.*[12] is not intrinsic to the temperature dependence of $\rho_c(T)$, although we confirm that in contrast to other underdoped cuprates, $\rho_c(T)$ in $YBa_2Cu_4O_8$ does become increasingly more metallic as we approach T_c. This observation represents an important challenge to existing theoretical models of the c-axis conductivity of high-T_c cuprates and suggests that interplanar disorder may have a significant effect on the behavior of $\rho_c(T)$ in other underdoped cuprates.

$YBa_2Cu_4O_8$ crystals were grown by a flux method in Y_2O_3 crucibles and an Ar/O_2 mixture at 2000 bar, with a partial oxygen pressure of 400 bar.[13] $\rho_a(T)$ and $\rho_b(T)$ were measured by a direct (four-probe) method, while $\rho_c(T)$ was measured using a quasi-Montgomery technique on near-cubic crystals. A large number of crystals were studied and the data shown here were reproduced on several other crystals for all three current directions (up to 300 K).[14] The transition temperatures varied from 76 to 79 K (defined as the temperature of zero resistance) and the resistive transitions had a typical width of 0.5–1.0 K.

Because of the resistive anisotropy within the plane, $\rho_b(T)$ data were taken on needlelike crystals to ensure 1D current flow along the b axis. For $\rho_c(T)$, large current and voltage pads were mounted across the top and bottom so as to short out the a-axis and maximize the c-axis dimension of the effective ''isotropic'' crystal. As previously mentioned, Zhou *et al.*[12] reported c-axis resistance $R_c(T)$ data for $YBa_2Cu_4O_8$, which showed a maximum around $T=140$ K. However, for their crystal, $R_c(300\ \text{K})<0.003\ \Omega$, which we believe is far too small to represent a genuine $\rho_c(T)$ measurement. This maximum in $R_c(T)$ most likely arises from a change in the current distribution inside the crystal as the anisotropy ρ_c/ρ_b decreases and the effective ''isotropic'' crystal becomes thinner. To illustrate this point further, Fig. 1 shows $R_c(T)$ data for several crystals of different aspect

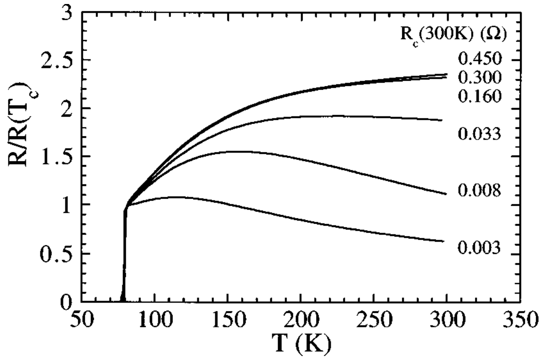

FIG. 1. c-axis resistance $R_c(T)$ data for $YBa_2Cu_4O_8$ crystals with different R_c (300 K) values. The data have been normalized to the value at $T=T_c$. For R_c (300 K)$=0.30\ \Omega$ and $0.45\ \Omega$, the data overlap.

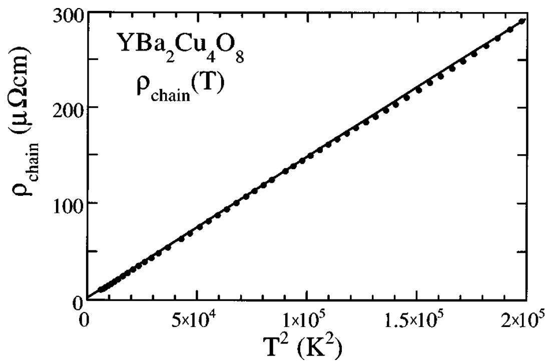

FIG. 3. $\rho_{\rm chain}(T)$ of $YBa_2Cu_4O_8$ plotted versus T^2. The solid line is a fit to the expression $\rho_{\rm chain}(T)=0.5+0.00147T^2$ $\mu\Omega$ cm. Note that there is no signature of a crossover in behavior with decreasing T.

ratios [and hence different $R_c(300$ K) values]. The data are normalized to R_c $(T=T_c)$. For crystals with R_c (300 K) $<0.07\ \Omega$, we also observed a maximum in $R_c(T)$ which shifted to lower T as R_c (300 K) decreased. For R_c (300 K) between 0.2 and 0.7 Ω, however, the T dependences and absolute magnitudes of $\rho_c(T)$ were found to be identical within experimental accuracy, at least up to 300 K. The $\rho_c(T)$ data discussed in this report were taken on a crystal of (190 μm)^2 area and 140 μm thickness with R_c (300 K)$=0.3$ Ω. No maximum was observed in $\rho_c(T)$ up to 450 K. Errors in the absolute magnitudes of the resistivities, mainly due to finite contact dimensions, are estimated to be around 25% for $\rho_a(T)$ and $\rho_b(T)$ and around 10% for $\rho_c(T)$.

Figure 2 shows all three resistive components of $YBa_2Cu_4O_8$ below 450 K. The a axis is perpendicular to the double CuO chains and so $\rho_a(T)$ represents the resistive component of the CuO_2 planes only. $\rho_a(T)$ decreases essentially linearly with T from 450 K, but as T falls, the slope $d\rho_a/dT$ gradually increases. This change of slope has been interpreted as a ''kink'' in $\rho_a(T)$ around $T^*=160-180$ K, which is thought to correlate with the opening of the normal-state gap and is attributed to the removal of the spin scattering channel within the plane around $T=T^*$.[8] Although T^* is itself a debatable quantity, as we shall discuss later, it is clear that there is a marked reduction in the in-plane scattering rate over a wide temperature range well above T_c.

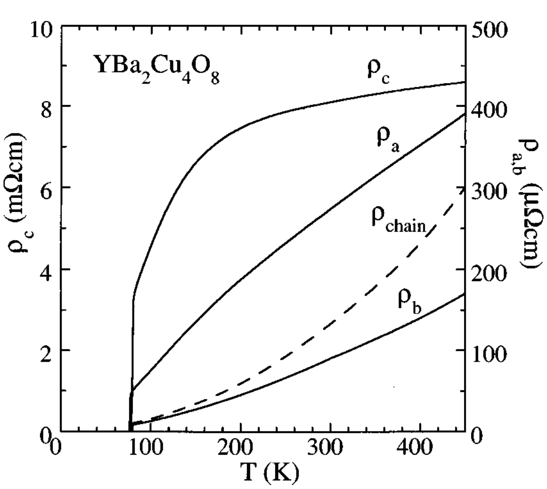

FIG. 2. The anisotropic resistivity components of $YBa_2Cu_4O_8$ up to 450 K. Solid lines represent $\rho_a(T)$, $\rho_b(T)$, and $\rho_c(T)$. The dashed line represents $\rho_{\rm chain}(T)$ (right-hand scale) derived from the expression $1/\rho_{\rm chain}=1/\rho_b-1/\rho_a$.

$\rho_b(T)$ contains contributions from both the CuO_2 planes and the double CuO chains and therefore has a lower resistivity than $\rho_a(T)$. The anisotropy at $T=T_c$ $(\rho_a/\rho_b\approx 6)$ is in excellent agreement with the square of the anisotropy of the in-plane penetration depth $(\lambda_a/\lambda_b)^2=6.25$,[15] implying that the scattering rates $1/\tau_{\rm chain}$ and $1/\tau_{\rm plane}$, are essentially equivalent at the superconducting transition. NMR and NQR measurements have shown that spin susceptibilities on the planes and chains are markedly different,[7,16,17] suggesting that the two conducting channels may act independently of one another. Assuming isotropic conductivity within the CuO_2 planes, we can extract $\rho_{\rm chain}(T)$ using the expression for two conductors in parallel, i.e., $1/\rho_{\rm chain}(T)=1/\rho_b(T)-1/\rho_a(T)$. The resultant $\rho_{\rm chain}(T)$, plotted as a dashed line in Fig. 2, can be fitted extremely well to the form $\rho_{\rm chain}(T)=\rho_0+AT^2$, as shown in Fig. 3, with $\rho_0=0.5\mu\Omega$ cm and $A=1.47$ $n\Omega$ cm/K^2.[18] A T^2 dependence has also been reported for $\rho_{\rm chain}(T)$ in $YBa_2Cu_4O_8$ (Ref. 19) and untwinned $YBa_2Cu_3O_{6.9}$ (Ref. 20) with ρ_0 values of 10 $\mu\Omega$ cm and 100 $\mu\Omega$ cm respectively. Since quasi-1D structures such as CuO chains are extremely susceptible to disorder, the remarkably low value of ρ_0 reported here implies that there is negligible disorder or oxygen vacancies along the chains and confirms the very high level of purity of these crystals. The T^2 resistivity, together with the observation of a Korringa law in $1/T_1T$ for the Cu(1) chain site in $YBa_2Cu_4O_8$,[6,16] suggests that the chain carriers are itinerant electrons of Fermi liquid character. Furthermore, Knight shift data for the Cu(1) (Refs. 7,17) and O(1)[17] chain sites are essentially T independent with no clear evidence of the gap in the spin spectra above T_c that is seen within the plane.[7,16,17] Importantly, $\rho_{\rm chain}(T)$ shows no deviation from its T^2 behavior between 80 and 450 K, providing further evidence that the behavior of $\rho_a(T)$ is intimately linked to the opening of the normal-state gap.

We now turn our discussion to $\rho_c(T)$. At 300 K, $\rho_c\approx 8$ $m\Omega$ cm and $\rho_c/\rho_a\approx 30$. These values are significantly lower than those for $YBa_2Cu_3O_{6.8}$,[3] which has a similar T_c and hole doping level to $YBa_2Cu_4O_8$, and suggests that there is a considerably stronger c-axis coupling in $YBa_2Cu_4O_8$,

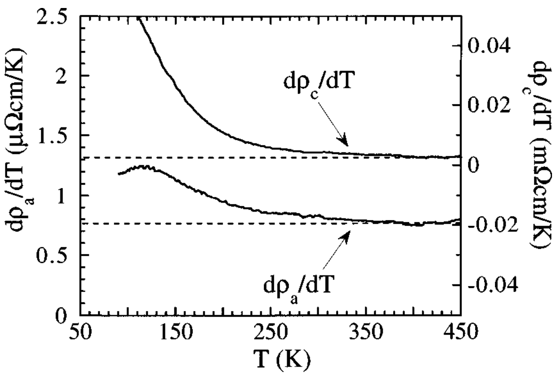

FIG. 4. The derivative of the in-plane ($d\rho_a/dT$) and out-of-plane resistivity ($d\rho_c/dT$) of $YBa_2Cu_4O_8$. Note that the derivative starts to increase in the same temperature range for both directions and that there is no kink in the derivatives at lower T which could be ascribed to T^*.

presumably due to the presence of the fully loaded double chains. Above around 350 K, $\rho_c(T)$ shows a very flat T dependence with a small positive slope $d\rho_c/dT = 3$ $\mu\Omega$ cm/K, which extrapolates to a very large apparent residual resistivity at 0 K. Similar behavior has also been observed in underdoped $YBa_2Cu_3O_{7-\partial}$ (Refs. 2,3) and $La_{2-x}Sr_xCuO_4$ (Ref. 21). Although this high-T resistivity looks ''metallic,'' measurements of the pressure dependence of $\rho_c(T)$ in $La_{2-x}Sr_xCuO_4$ (Ref. 22) have shown that this positive $d\rho_c/dT$ can be attributed almost entirely to the effects of thermal expansion on some form of incoherent tunneling or hopping process along the c axis. It seems reasonable to assume that the almost T-independent $\rho_c(T)$ observed here is also a signature of nonmetallic c-axis conductivity in $YBa_2Cu_4O_8$ at high T. As T falls, $\rho_c(T)$ starts to decrease more rapidly. The slope in the resistivity changes continuously down to T_c, and by 100 K, $\rho_c(T)$ has reduced to almost half its value at 350 K. Thermal expansion is essentially linear below 300 K (Ref. 23) and hence it is most unlikely that this dramatic decrease in $\rho_c(T)$ can be attributed to any type of expansion effect. Therefore, we conclude that this dramatic change in slope reflects a gradual crossover from 2D to 3D electronic transport in $YBa_2Cu_4O_8$ with decreasing T.[24]

This crossover at lower T is strikingly different from what is observed in underdoped $YBa_2Cu_3O_{7-\partial}$ (Refs. 2,3) and $La_{2-x}Sr_xCuO_4$ (Ref. 21) where $d\rho_c/dT$ changes its sign from positive to negative as T falls. It is perhaps even more remarkable when one compares $YBa_2Cu_4O_8$ and $YBa_2Cu_3O_{6.8}$ (Ref. 3) where the T dependence and magnitude of $\rho_a(T)$ are almost identical, yet $\rho_c(T)$ deviates in opposite directions at low T. It is significant to note too that for $YBa_2Cu_4O_8$, ρ_c (100 K)≈ 8 $m\Omega$ cm is on the boundary of the Mott-Ioffe-Regel criterion for metallic c-axis conductivity.[25] In this sense, $\rho_c(T)$ of $YBa_2Cu_4O_8$ is similar to that of the quasi-2D metal Sr_2RuO_4, which has a broad maximum around 130 K and shows a gradual evolution to a metallic T^2 dependence below 30 K where $\rho_c(T) < 10$ $m\Omega$ cm.[26]

In Fig. 4 we plot the temperature derivatives $d\rho_c/dT(T)$ and $d\rho_a/dT(T)$, in order to illustrate some common misconceptions about the behavior of the normal-state resistivity in underdoped cuprates. First, $\rho_a(T)$ first deviates from linearity at a much higher T than the value of T^* that is commonly quoted for $YBa_2Cu_4O_8$. Indeed, in the vicinity of T^* (160 K$<T^*<$200 K), it is clear that there is no additional feature in $d\rho_a/dT$ and the characteristic ''kink'' at T^* should really be dismissed as a crude visual interpretation of the data. Second, the change of slope in both current directions is a very gradual, continuous process and does not appear, as is often suggested, as some form of phase transition. Finally, the deviation from linearity appears to begin within the same range for both $\rho_a(T)$ and $\rho_c(T)$, suggesting that the crossover to metallic conduction along the c axis coincides with a reduction of the in-plane scattering rate. Whether the onset of metallic conduction along the c axis is actually correlated with the opening of the normal-state gap is of course still debatable and further studies, such as the effects of hole doping on $\rho_c(T)$, will be required in order to provide a more definitive answer to this question.

The behavior of $\rho_c(T)$ also raises another important issue, namely the existence of a genuine pseudogap in $\sigma_c(\omega)$ in $YBa_2Cu_4O_8$, as reported by Basov *et al.*[11] This pseudogap was believed to show the generic features of the pseudogap state observed in other underdoped cuprates[4,5,27] and it was assumed that the spectral weight that is suppressed below 200 cm^{-1} is transferred to higher energies. However, in order for $\rho_c(T)$ and $\sigma_c(\omega)$ to be consistent, there must be an enhancement in $\sigma_c(\omega)$ at low frequencies. Hence, in $YBa_2Cu_4O_8$, the missing spectral weight may be transferred to *lower* frequencies, at least in part.

There has been considerable theoretical effort[28–34] in trying to understand the origin of the nonmetallic $\rho_c(T)$ of underdoped high-T_c cuprates at low T. In particular, this behavior has been viewed as strong evidence for charge confinement within the CuO_2 planes, possibly associated with a non-Fermi liquid ground state and/or spin singlet pair formation in cuprate superconductors.[28,29,32,34] This claim has been strengthened by the observation of a low-T metallic c-axis response in Sr_2RuO_4 which is isostructural with La_2CuO_4 but shows many properties consistent with a highly anisotropic Fermi-liquid description.[35] The observation of similar behavior in underdoped $YBa_2Cu_4O_8$ now represents an important challenge to the charge confinement picture.

We may draw two possible scenarios for the distinct $\rho_c(T)$ behavior in $YBa_2Cu_4O_8$. First, we note here that $YBa_2Cu_4O_8$ and Sr_2RuO_4 are stoichiometric compounds with effectively no disorder between the conducting planes. In contrast, as we have already pointed out, the other cuprates (in particular underdoped $YBa_2Cu_3O_{7-\partial}$), which show a pseudogap in $\sigma_c(\omega)$ and a nonmetallic $\rho_c(T)$ at low T, contain a significant amount of interplane disorder in the form of oxygen vacancies and substituted cations. These impurities must act as an extra blocking layer for out-of-plane transport and reduce the mean free path along the c axis. This effect will be substantially enhanced when the transfer integral perpendicular to the planes is very small and may ultimately inhibit the formation of a 3D metallic band at low T. Within this simple picture, the ''charge confinement'' in $YBa_2Cu_3O_{6.8}$ would originate from the highly disordered blocking layer situated between the planes (with 20% oxygen vacancies) rather than from any exotic non-Fermi liquid property of the CuO_2 plane carriers themselves.

An alternative scenario is that the metallic c-axis conductivity does not show a generic behavior of layered cuprates but is observed only in $YBa_2Cu_4O_8$ due to the presence of the double chain Fermi sheets. It may be argued, within a two-channel picture, that only the chain channel forms a coherent band along the c axis and therefore it may still be possible that the carriers in the plane band remain confined. However, this appears to contradict the apparent correlation between the onset of metallic behavior in $\rho_c(T)$ and the reduction of the *in-plane* scattering rate. Further transport studies, where disorder is introduced onto the planes or onto the chains, should help to clarify which scenario is the most applicable to $YBa_2Cu_4O_8$.

In conclusion, we have measured all three resistive components of $YBa_2Cu_4O_8$ up to 450 K. $\rho_c(T)$ shows a striking crossover from incoherent to metallic behavior with decreasing T. Since $YBa_2Cu_4O_8$ is one of the cleanest high-T_c cuprates, its c-axis response may represent the *intrinsic* behavior of the out-of-plane conductivity in underdoped cuprates and suggests that interplane disorder plays a much more critical role in determining the behavior of $\rho_c(T)$ than has previously been acknowledged. Furthermore, these observations cast a serious challenge against the generally accepted picture of charge confinement in underdoped cuprates below T^*.

We would like to acknowledge enlightening discussions with N. Nagaosa, S. Tajima, J. L. Tallon, S. Uchida, and T. Xiang. We would also like to thank M. Kibune for experimental assistance. This work was supported by Core Research for Evolutional Science and Technology (CREST) and NEDO for R&D of Industrial Science and Technology Frontier Program.

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*On leave from Interdisciplinary Research Centre in Superconductivity, University of Cambridge, Madingley Road, Cambridge, CB3 0HE, U.K.

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