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Temperature behavior of the out-of-plane resistivity in single-crystal $YBa_2Cu_3O_{7-\delta}$

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Analysis of anisotropic transport data of $YBa_2Cu_3O_{7-\delta}$ shows that the out-of-plane resistivity ρ_c changes from nonmetallic to metallic following a reduction in ρ_c. The metallic ρ_c arises from a conventional relaxation mechanism within the framework of the band theory, while the nonmetallic ρ_c is dominated by hopping conduction. Whether the Mott-Ioffe-Regel rule is satisfied by the out-of-plane transport draws a distinction between relaxation transport and hopping conduction. The results can account for experimental data observed on single-crystal $YBa_2Cu_3O_{7-\delta}$ as well as other high-T_c copper oxides.

Large anisotropy in the normal-state transport properties of high-T_c copper oxides arises from their layered structures and the two-dimensional electronic nature of their CuO planes. Information on anisotropic resistivity has been obtained from a large number of measurements on single-crystal samples of $YBa_2Cu_3O_{7-\delta}$ (Y-1:2:3),[1-9] as well as other copper oxides.[10-12] For in-plane resistivity ρ_{ab} (along the CuO plane), there is unanimity on linear T-dependence, $\rho_{ab} \propto T$, over a rather wide range of temperatures.[13] For the out-of-plane resistivity ρ_c (perpendicular to the CuO plane), however, there are contradictory reports on T dependence. Both metallic and nonmetallic behaviors have been observed by different groups. Some groups[1-5,10] reported ρ_c to have a semiconductorlike upturn as T decreases (before dropping to zero at T_c). Others[6-9,11,12] found that ρ_c for a number of high-quality samples shows a metallic T dependence, similar to ρ_{ab}. Owing to the disagreement in experimental data, whether the nonmetallic or the metallic behavior of ρ_c is intrinsic has become an outstanding question.

Let us first analyze experimental data of ρ_c of high-T_c copper oxides reported by several groups and try to discover a phenomenological law. The ρ_c observed in the Y-1:2:3 single crystals has three types of T dependences,[8] depending upon the different types of samples. The first type of samples exhibits a strong, nearly exponential increase of ρ_c with decreasing T.[5,8] The second type of ρ_c shows a weak T dependence with a shallow minimum around about 150 K.[1-4] In both cases the T dependence of ρ_c is nonmetallic. By contrast, for the third type of samples, ρ_c has a nearly linear T dependence from T_c to room temperature.[6-9] Its temperature behavior is metallic, similar to that of ρ_{ab}, and therefore the anisotropic ρ_c/ρ_{ab} has only a weak T dependence. In Table I we summarize the main features of ρ_c in the three types of single-crystal samples of Y-1:2:3. These data show that among the three types of samples, type I has the highest absolute values of ρ_c, type II takes second place, and type III has the lowest in ρ_c. It has been suggested that high-quality single crystals of Y-1:2:3, which exhibit a sharp superconducting transition, show a linear ρ_c T dependence and at the same time have lower values of ρ_c as compared to those with a nonmetallic T dependence.[6-8] Recently, Friedmann *et al.*[9] reported that twin-free single crystals of Y-1:2:3 show linear T dependence of the resistivities along all the three orthorhombic axes. Their samples have the lowest resistivities reported so far and may approximate intrinsic behavior. In sum, there seems to be a phenomenological law that the T dependence of ρ_c changes from semiconductorlike to metallic following a reduction in ρ_c.

Similar phenomena have been observed in other high-T_c copper oxides. Earlier work on the Bi compounds[10] shows ρ_c to have a nonmetallic character. Very recently, Xiang *et al.*[11] used iodine intercalation to alter the interplane interaction of the Bi compound $Bi_2Sr_2CaCu_2O_x$. They found that in the normal state above T_c, the metallic CuO_2-sheet resistance is unaffected by intercalation, while ρ_c changes from semiconductorlike to metallic. Anisotropic resistivity measurements of $Tl_2Ba_2CaCu_2O_8$ single crystals[12] also indicate that ρ_c decreases with decreasing T, exhibiting a metallic character, although it is still more than 100 times larger than ρ_{ab}.

It was generally accepted that the nonmetallic ρ_c is the intrinsic behavior of the high-T_c copper oxides before the linear T dependence of ρ_c was found. It is difficult to interpret simultaneously the nonmetallic behavior of ρ_c and the linearity of ρ_{ab} in the same temperature region. It does not appear possible to explain such a disparity of T behavior between ρ_{ab} and ρ_c within the Bloch-Boltzmann approach. Furthermore, the possible effect of localization on the anisotropic transport can also be excluded, because the scaling theory of localization to anisotropic systems[14] does not permit anisotropic localization, i.e., the localization transition occurs at the same time in all the directions, regardless of anisotropy.

We propose that the intrinsic behavior of ρ_c may change from nonmetallic to metallic as ρ_c decreases with

TABLE I. Anisotropic resistivities for three types of single crystals of $YBa_2Cu_3O_{7-\delta}$.

Type	ρ_c (100 K) mΩ cm	ρ_c (100 K)/ρ_c (300 K)	ρ_c/ρ_{ab} at 100 K	ρ_c/ρ_{ab} at 300 K	Reference
I	420–480	2.4			5
	170–350	3.1–3.2	210–1300	16–20	8
	38	2.0	48	10	7
II	19–23	0.8	26–31	16–20	8
	19.5	1.0	95	50	4
	17	1.5			6
	16	1.2	80	30	1
	9–12	0.7–0.9	200–250	56–110	2
III	8–10	0.4	35–40	35–40	7
	3.5–6.7	0.4	52–58	33–41	8
	3	0.65	75	30	6
	2.3–3.4	0.5–0.6	75–87	45	9

the increase of interplane coupling. The metallic transport is bandlike and relaxive, while the nonmetallic conduction may arise from a hopping mechanism. It should be stressed that the relative transport in which carriers undergo scattering during their band motion is based on the band theory. In order to guarantee the band approximation suitable for charge transport, the Mott-Ioffe-Regel (MIR) rule[15] must be satisfied. It follows from the argument of Mott and Davis[15] that the minimum distance over which the phase memory of the wave function may be lost due to scattering is the distance between atoms and thus the interatomic spacing s is the minimum of the mean free path l. The condition $l>s$ is always well satisfied for conventional metals. For the layered oxide systems having narrow band and high resistivity, however, the MIR rule is likely to be violated, particularly for the out-of-plane transport. In this case the coherent band transport breaks down, and so hopping conduction may dominate charge transport. Thus, whether the MIR rule $l>s$ is satisfied can be considered as a criterion which distinguishes between relaxive transport and hopping conduction.

Owing to the anisotropy of high-T_c layered oxides, the MIR rule requires $l_{ab}>a$ and $l_c>d$ where l_{ab} and l_c are the in-plane and out-of-plane mean free path, respectively, and a and d are the in-plane and out-of-plane lattice constants. If both conditions $l_{ab}>a$ and $l_c>d$ are satisfied the band approximation is valid for the in-plane and the out-of-plane transport, and thus the system can be regarded as a three-dimensional (3D) anisotropic metal. In this case ρ_c has a metallic T dependence, the same as ρ_{ab}. On the other hand, on condition that $l_{ab}>a$ but $l_c<d$, the band approximation is valid only for the in-plane transport, and so the dominant mechanism of ρ_c can be hopping conduction. As was proposed by us,[16] the nonmetallic T dependence of ρ_c may be explained by a phonon-assisted hopping mechanism of electrons between adjacent CuO planes. Such an argument accounts for the disparate types of T dependence of ρ_c observed in the high-T_c copper oxides.

Let us start from a 3D anisotropic metal. Its band structure is modeled as

$$\varepsilon_{\mathbf{k}}=(\hbar\mathbf{k}_{ab})^2/(2m_{ab})-2t_c\cos(k_c d)\ , \tag{1}$$

where $\mathbf{k}_{ab}$ and k_c are the wave vectors parallel to the CuO plane and along the c axis, respectively. m_{ab} is the effective mass of carriers for the motion within the CuO plane and t_c is the interplane coupling. For the energy-wave-vector relationship given in Eq. (1), the group velocity of carriers associated with a state $\mathbf{v}$ is given by $\mathbf{v}_{ab}(\mathbf{k}_{ab})=\hbar\mathbf{k}_{ab}/m_{ab}$, and $v_z(k_z)=(2t_c d/\hbar)\sin(k_c d)$, so that we have $l_{ab}\simeq v_F\tau$, and $l_c\simeq(t_c d/\hbar)\tau$, with v_F the Fermi velocity and τ the relaxation time. Thus, the MIR conditions $l_{ab}>a$ and $l_c>d$ can be rewritten as $\tau>a/v_F$ and $\tau>\hbar/t_c$, respectively, for the in-plane and the out-of-plane transport.

On the other hand, an expression for anisotropic resistivities can be derived by solving the linear Boltzmann equation in the relaxation-time approximation

$$e\mathbf{E}\cdot\mathbf{v}(\mathbf{k})\frac{\partial f_0}{\partial\varepsilon}=\frac{f-f_0}{\tau}\ , \tag{2}$$

and by calculating the electric current density

$$\mathbf{J}=2e\int\mathbf{v}(\mathbf{k})(f-f_0)d^3k/(2\pi)^3\ . \tag{3}$$

Here f_0 is the equilibrium distribution function and $f-f_0$ is the correction to the distribution function due to the electric field $\mathbf{E}$ and the scattering which transfers carriers from one band state to the others. Substituting the expressions for $\mathbf{v}(\mathbf{k})$ into Eqs. (2) and (3), we obtain for the anisotropic resistivities

$$\rho_{ab}=m_{ab}/(ne^2\tau) \tag{4}$$

and

$$\rho_c=\hbar^2\varepsilon_F/(2ne^2\tau t_c^2 d^2)\ , \tag{5}$$

with n as the density of charge carriers. It is interesting to notice that the above result is the same as that obtained recently by Kumar and Jayannavar,[17] even though each has a different physical picture.

Since both ρ_{ab} and ρ_c given in Eqs. (4) and (5) are inversely proportional to τ, they must have the same T

dependence. However, it should be emphasized that this result is entirely dependent on the validity of the band theory and the Bloch-Boltzmann transport picture. Substituting Eqs. (4) and (5) into inequalities $\tau > a/v_F$ and $\tau > \hbar/t_c$, we obtain the following form of the MIR rule:

$$\rho_{ab} < \rho_{ab}^* \equiv \frac{m_{ab} v_F}{ne^2 a} , \tag{6}$$

$$\rho_c < \rho_c^* \equiv \left[\frac{\hbar}{2ne^2a^2} \right] \left[\frac{m_c}{m_{ab}} \right] \left[\frac{\varepsilon_F}{t_{ab}} \right] , \tag{7}$$

with the effective mass anisotropy $m_c/m_{ab} = (t_{ab}/t_c)(a/d)^2$ and t_{ab} the in-plane transfer matrix element. Inequalities (6) and (7) are the major result of the present paper, in which ρ_{ab} and ρ_c are the measured values of the in-plane and out-of-plane resistivities, respectively, and ρ_{ab}^* and ρ_c^* stand for the corresponding critical values. All physical parameters which appear in the definitions of ρ_{ab}^* and ρ_c^* can be deduced from a variety of measurements. For example, n is usually determined by the Hall effect or by iodometric titration. The effective mass ratio can be estimated from $m_c/m_{ab} = (\lambda_c/\lambda_{ab})^2$ where λ_c/λ_{ab} is the anisotropic ratio of the London penetration depths.[17]

For $YBa_2Cu_3O_7$ single crystals, with the typical values of the following parameters: $n \simeq 5\times 10^{21}/\text{cm}^3$, $a \simeq 3.85$ Å, $v_F \simeq 10^7$ cm/sec, and $m_{ab} \simeq 5m_e$ (m_e being the free-electron mass), ρ_{ab}^* is estimated to be about 1 mΩ cm. Taking into account that $\varepsilon_F/t_{ab} \simeq 0.8$ and $m_c/m_{ab} \simeq 25-64$ ($\lambda_c/\lambda_{ab} \simeq 5-8$ in the low-temperature limit), we estimate ρ_c^* at 100 K (just above T_c) to be 5.5–14 mΩ cm. This estimation for ρ_c^* seems to be reasonable. As has been shown in Table I, the third type of ρ_c with metallic T dependence ranges between 2 and 10 mΩ cm at 100 K. In this case, since both conditions $\rho_{ab} < \rho_{ab}^*$ and $\rho_c < \rho_c^*$ are satisfied, Eqs. (4) and (5) can be used to account for the same linear T dependence of both ρ_c and ρ_{ab} provided that $1/\tau \propto T$. In Table I the second type of ρ_c with nonmetallic and weak T behavior ranges from 9 to 23 mΩ cm at 100 K. Since these values for ρ_c are slightly greater than ρ_c^*, inequality (7) cannot be well satisfied. Consequently, Eq. (5) for ρ_c is no longer valid even though Eq. (4) for ρ_{ab} still holds, causing ρ_{ab} and ρ_c, respectively, to have metallic and nonmetallic T dependences. The first type of ρ_c is always much greater than ρ_c^*, and so exhibits strong T behavior which may be dominated by the hopping mechanism.[16]

Equation (7) shows that ρ_c^* is inversely proportional to the interplane coupling t_c. So, the value for ρ_c^* may vary from sample to sample and depends sensitively on the value of the oxygen deficiency δ in $YBa_2Cu_3O_{7-\delta}$ samples. Structure studies have established that the vacancies introduced by removing oxygen atoms from the O_7 phase of $YBa_2Cu_3O_{7-\delta}$ lie primarily in the Cu(1) chains rather than the Cu(2) planes. The removal of oxygen atoms from the chains makes electrons transfer from the Cu(1) chains to the Cu(2) planes, which has two effects on the anisotropic transport: charge compensation eliminates the hole density n on the Cu(2) planes; and charge transfer enhances the height of the potential barrier between the adjacent Cu(2) planes so as to decrease t_c. It follows from Eq. (5) that both effects lead to an increase in ρ_c. At the same time, the latter effect of oxygen deficiency (with increasing δ) is unfavorable to the inequality $\tau > \hbar/t_c$ which is necessary to the metallic transport along the c axis. Therefore, almost all the ρ_c vs T profiles with a metallic character are observed in single-crystal samples of fully oxygenated $YBa_2Cu_3O_{7-\delta}$ ($\delta < 0.1$), correlated with T_c in excess of 90 K.[6,9] It can be also understood that in the 60 K Y-1:2:3 with δ near 0.4, not only is ρ_c much greater than in the 90 K Y-1:2:3, but it always increases monotonically with decreasing temperature.[18]

We now use the above arguments to discuss a recent report on the anisotropic transport measurements[11] for the iodine-intercalated $IBi_2Sr_2CaCu_2O_x$ compounds in which the iodine intercalation between the Bi-O bilayers causes a 23% expansion of the crystal along the c axis and a suppression of T_c by 5–10 K. In the normal state, the metallic in-plane resistance was unaffected by intercalation, but ρ_c was dramatically changed from semiconductorlike to metallic, following a reduction in ρ_c. Since at a constant carrier density ρ_c varies inversely as the square of t_c, the observed reduction in ρ_c with intercalation results from the increase of t_c. At the same time, the increase of t_c is favorable to the condition $\tau > \hbar/t_c$ necessary to the metallic conduction along the c axis. Thus, the present theory, along with the assumption that the iodine intercalation leads to an increase in t_c may explain the metallic T dependence for ρ_c obtained in the $IBi_2Sr_2CaCu_2O_x$ crystals. This is contrary to the assumption that t_c decreases with intercalation,[11] which was made to account for the depression in T_c observed in the $IBi_2Sr_2CaCu_2O_x$ crystals, but it is very difficult to adequately explain the decrease in ρ_c, as well as the metallization of the ρ_c vs T profile. It appears that T_c may be correlated with t_c by a more complicated mechanism than that considered in Ref. 11. Their correlation is expected to depend, to a great extent, on the mechanism for the high-T_c superconductivity which is not yet clear.

We now discuss the case of $\rho_{ab} > \rho_{ab}^*$ and $\rho_c > \rho_c^*$, in which the band theory is not suitable for either the in-plane or the out-of-plane transport, and the hopping mechanism will dominate conduction in all directions. This situation may occur at high temperatures where ρ_{ab} due to inelastic scattering increases and exceeds ρ_{ab}^*. The deviation of the ρ_{ab} vs T profile from linearity at high temperatures has been observed in $La_{1.825}Sr_{0.175}CuO_4$ and $YBa_2Cu_3O_7$ samples.[19] Although this deviation is closely related to the loss of oxygen at high temperatures, its behavior can be explained in part by the violation of the MIR rule given in Eq. (6). As has been estimated above, $\rho_{ab}^* \simeq 1$ mΩ cm for $YBa_2Cu_3O_7$. According to this estimation, as $\rho_{ab} < 1$ mΩ cm the resistivity will deviate from linearity and dramatically increase. This prediction seems to be consistent with the experimental data given in Ref. 19. For $La_{1.85}Sr_{0.175}CuO_4$, the resistivity at which the ρ_{ab} vs T profile starts to deviate from linearity is found to be higher than that for $YBa_2Cu_3O_7$. It can be understood that the $La_{1.825}Sr_{0.175}CuO_4$ system has lower n, and so has higher ρ_{ab}^{*} as compared with $YBa_2Cu_3O_7$.

Finally, we wish to point out that the copper oxides are strongly correlated systems. The strong Coulomb correlations may be responsible for the metal-insulator transition as well as the high-T_c mechanism. In the present approach, the important effect of strong Coulomb correlations was not explicitly considered. Consequently, we limit ourselves to a phenomenological study of ρ_c changing from semiconductorlike to metallic without discussing its microscopic origins. The latter is an open question and awaits further investigation into strongly correlated systems.

In summary we have proposed that there may be two different criteria for distinguishing the conduction mechanism for the in-plane and the out-of-plane transport in layered oxide compounds. If the interplane coupling t_c is large enough for the band theory to be true for the out-of-plane conduction, the layered system can be regarded as a 3D anisotropic metal. In this case both ρ_{ab} and ρ_c are dominated by the scattering mechanism, exhibiting the same metallic T dependence. In the opposite case of small t_c, the Bloch-Boltzmann approach is not suitable for the out-of-plane transport, and the hopping mechanism will cause ρ_c to exhibit nonmetallic behavior, even though ρ_{ab} is still metallic. The present arguments can be used to account for the different types of T dependences of ρ_c observed in the high-T_c copper oxides.

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