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Interplane charge transport in $YBa_2Cu_3O_{7-y}$: Spin-gap effect on in-plane and out-of-plane resistivity

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We have investigated the anisotropic resistivities of $YBa_2Cu_3O_{7-y}$ using detwinned crystals with various oxygen contents ($6.68 \leq 7-y \leq 6.93$). The out-of-plane resistivity ρ_c shows a crossover from high-T metallic to low-T semiconducting behavior while the in-plane resistivity ρ_a deviates in the low-T region from T-linear dependence. We find that the crossover in ρ_c is linked with the onset of nonlinearity in ρ_a, which seems to be associated with the "spin gap" suggested by neutron and NMR studies.

Recently, an explanation of the in-plane charge transport in high-T_c cuprates in relation to the spin excitations has been put forth, based on the results of the in-plane resistivity $\rho_{ab}(\rho_a)(T)$ and Hall coefficient $R_H(T)$ measured on $YBa_2Cu_3O_{7-y}$ (Ref. 1) and $YBa_2Cu_4O_8$.[2] In both systems, ρ_a for the crystals in the underdoped regime deviates from the T-linear behavior and R_H from the $1/T$ one below a certain temperature well above the superconducting transition temperature (T_c). The deviation was found to correspond to the gap formation in the spin excitation spectrum suggested by neutron and NMR studies.[3,4]

The out-of-plane resistivity ρ_c of $YBa_2Cu_3O_{7-y}$ has been investigated by many researchers, which revealed that both the magnitude and T dependence of ρ_c are very sensitive to the oxygen content[5,6] and that ρ_c shows semiconducting temperature dependence except for $y \sim 0$. However, the origin of the semiconducting out-of-plane conduction is still in dispute. In order to get insight into this problem, we have measured the anisotropic resistivities using untwinned crystals of $YBa_2Cu_3O_{7-y}$ ($6.68 \leq 7-y \leq 6.93$) and established how the magnitudes and T dependences of in-plane and out-of-plane resistivity vary with carrier concentration. The present work reveals that the metallic-to-semiconducting crossover in ρ_c correlates with the deviation from the T-linear behavior in ρ_a.

Single crystals of $YBa_2Cu_3O_{7-y}$ were grown by a self-flux method using a Y_2O_3 crucible. We obtained single-domain crystals by picking up as-grown twin-free crystals from the crucible or by applying uniaxial stress on the twinned crystals. The oxygen content of crystals was controlled by annealing them at 600 °C for 12 h in a sealed quartz tube together with about 10 g of polycrystalline $YBa_2Cu_3O_{7-y}$ which had a prescribed oxygen content. After annealing, the crystals in a quartz tube were slowly cooled in order to reduce oxygen disorder in the chain site.[1] The annealing condition mentioned above was crucial to avoid introducing twins into the twin-free crystals. The observation by the polarized optical microscope and the single-crystalline x-ray diffraction confirm that the detwinning was perfect. These procedures enable us to obtain untwinned crystals for any oxygen content, which produce highly reproducible and systematic transport data.

In this experiment, in-plane resistivities ρ_a and ρ_b were measured on an *untwinned* crystal by the two-dimensional Montgomery method and out-of-plane resistivity ρ_c (and ρ_{ab}) was measured on a *twinned* crystal by the tetragonal-symmetry Montgomery method.[7] In the latter case, since the domain size is much smaller than the sample dimensions, we can safely treat the sample as a pseudotetragonal one. We measured several crystals with the same oxygen content to confirm that the scattering of the data is within the dimensional errors. All of the experimental results are shown in Figs. 1 and 2. With increasing oxygen content, the resistivities in all the directions decrease. Evidences for high quality

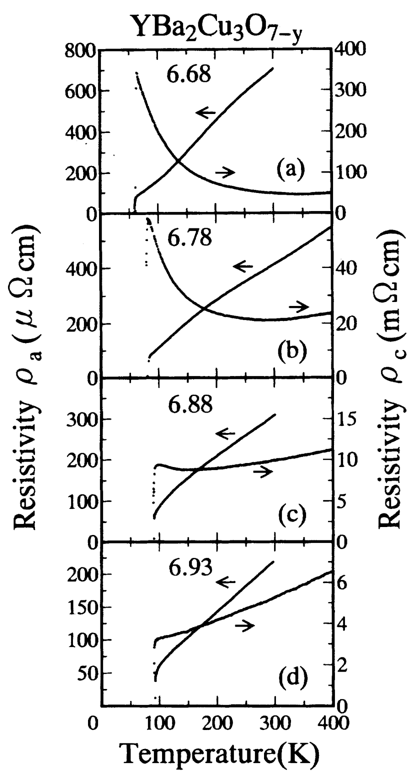

FIG. 1. In-plane (ρ_a) and out-of-plane (ρ_c) resistivity of $YBa_2Cu_3O_{7-y}$ plotted as a function of temperature for various oxygen contents $7-y \sim$ 6.68, 6.78, 6.88, and 6.93.

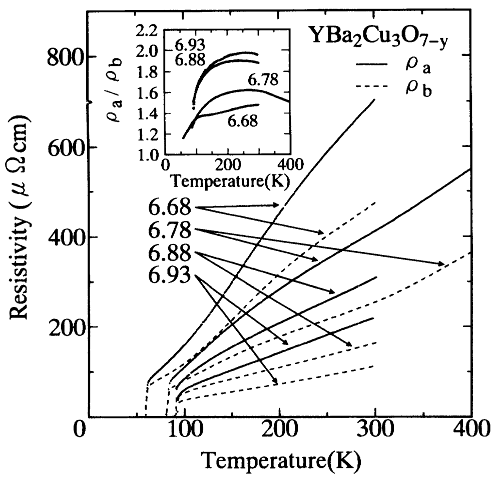

FIG. 2. Temperature dependences of two in-plane resistivity components, ρ_a (solid line) and ρ_b (broken line), measured on untwinned $YBa_2Cu_3O_{7-y}$ with various oxygen contents. The inset shows the temperature variation of anisotropic resistivity ratio ρ_a/ρ_b.

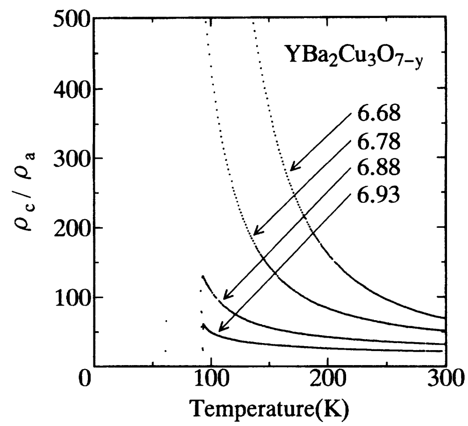

FIG. 3. Anisotropic resistivity ratio ρ_c/ρ_a plotted as a function of temperature for various oxygen contents.

of the present crystals are given by a narrow superconducting transition, a large *a-b* anisotropy, and the smallest residual resistivity components in all directions ($T_c \sim 92$ K with $\Delta < 1$ K for $7-y\sim$ 6.93 and 6.88, ~ 82 K with $\Delta < 2$ K for $\sim$ 6.78, and ~ 63 K with $\Delta < 3$ K for $\sim$ 6.68).

First, we describe the results of in-plane components of resistivity. ρ_a is the in-plane resistivity component perpendicular to the CuO chain, and therefore, is expected to contain purely the CuO_2 plane contribution. For the fully oxygenated crystal ($7-y\sim 6.93$ in the present case), ρ_a shows the T-linear behavior from above T_c to room temperature. As reported previously, ρ_a of reduced crystals deviates from this T-linear behavior at low temperatures.[1] The onset of the deviation, T_{a0}, increases with decreasing oxygen content. Thus, although the T-linear resistivity over a wide T range is considered to be characteristic of high-T_c cuprates,[8] it is observed only near the optimum composition.

Both ρ_a and ρ_b appear to show similar temperature and oxygen content dependence, though ρ_a is always larger than ρ_b. However, the value of ρ_a/ρ_b decreases as the temperature and/or the oxygen content decrease (Fig. 2). These differences can be interpreted by considering the chain contribution; the decrease of ρ_a/ρ_b with decreasing oxygen content is due to the decrease in the effective number of the complete CuO chains and/or the increase of the disorder in each chain caused by the oxygen deficiencies, and the decrease in ρ_a/ρ_b at low temperatures can be explained by the tendency for localization of carriers in the chains.[9,10]

The difference between ρ_a and ρ_b is also seen at high temperatures. It is impressive that ρ_b shows an appreciable upturn above room temperature whereas the feature is weaker in ρ_a. Reflecting this difference, ρ_a/ρ_b decreases as the temperature increases beyond 300 K. This upturn may originate possibly from oxygen rearrangement in the chains, as suggested by de Fontaine *et al.*[11] This is also suggested by the much larger upturn in ρ_b than in ρ_a because this rearrangement is expected to introduce defects into the chain. The slight upturn in ρ_a is probably an indirect effect of the oxygen rearrangement, for example, a slight change of the lattice parameters and/or of the hole density in the plane. Therefore, the T dependence of ρ_a would essentially be T linear to higher temperatures. This speculation is reinforced by the absence of a similar upturn in $YBa_2Cu_4O_8$,[2] in which the double-chain structure is much more stable against oxygen rearrangement.

Turning to the out-of-plane resistivity ρ_c, the magnitude of ρ_c is much more sensitive to the oxygen content than that of ρ_a, and the temperature dependence of ρ_c is characterized by a crossover from the high-T metallic ($d\rho_c/dT>0$) to the low-T semiconducting ($d\rho_c/dT<0$) regime. Even in the reduced 60-K crystal, we can see metallic T dependence above 300 K. As evident from Fig. 1, the crossover temperature, T_{c0}, decreases with increasing oxygen content, and the fully oxygenated sample does not show the semiconducting behavior down to T_c. ρ_c in the metallic region is roughly linear in T without showing any appreciable change at the temperature where the upturn is observed in the in-plane resistivity.

The band-structure calculation predicts that ρ_c/ρ_a is about 7 for $YBa_2Cu_3O_7$ and not very dependent on temperature and/or oxygen content.[12] However, our results show that ρ_c/ρ_a is much larger than the calculated value and strongly depends upon both temperature and oxygen content (Fig. 3). This suggests that the out-of-plane conduction is dominated by a different mechanism, owing to the highly two-dimensional electronic state.

For the sample with the lowest oxygen content ($7-y\sim 6.68$) in the present experiment, ρ_c in the semiconducting regime shows a T dependence with an activation energy $\sim$ 300 K in some temperature range but it becomes weaker at low temperatures—ρ_c in the semiconducting regime rather fits with a power-law T dependence, $\rho_c \sim T^{-\alpha}$ with α increasing with reducing oxygen content ($\alpha \sim 2$ for the 60-K crystal).

With increasing oxygen content, ρ_c rapidly decreases and the metallic T dependence dominates over a wide T range. ρ_c no longer shows a definite activation behavior. As a consequence, the anisotropic ratio ρ_c/ρ_a approaches the value predicted by the band-structure theory and becomes nearly T independent for the highest hole density. It seems that, as doping proceeds, the interplane conduction tends to restore

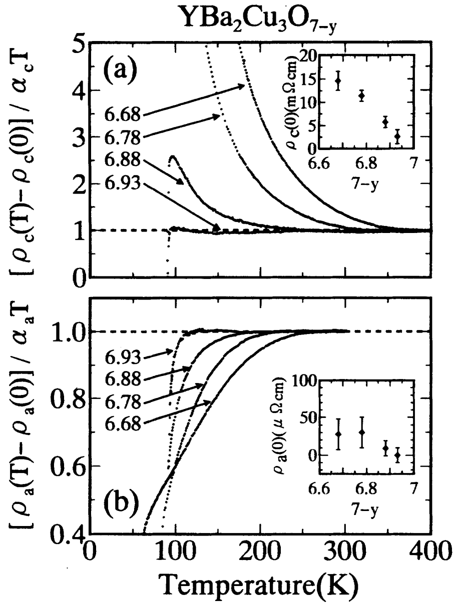

FIG. 4. Plot of the out-of-plane and in-plane resistivity data as $[\rho_i(T) - \rho_i(0)]/\alpha_i T$ vs T, $i=c$ in (a) and $i=a$ in (b). Here, α_i and $\rho_i(0)$ are the slope and intercept, respectively, when the metallic part of ρ_i is approximated with a T-linear one. The inset shows how $\rho_i(0)$ varies with oxygen content. Note that $\rho_c(0)$ radically increases with reduced oxygen content, whereas $\rho_a(0)$ does not with vanishingly small values for higher oxygen contents.

the coherent motion and the electronic system becomes three-dimensional.

In order to examine any correlation between in-plane and out-of-plane conduction, we plot the data as $[\rho_{c(a)}(T) - \rho_{c(a)}(0)]/\alpha_{c(a)}T$ vs T in Fig. 4. Here, $\alpha_{c(a)}$ and $\rho_{c(a)}(0)$ are slope and intercept, respectively, when the metallic part of $\rho_{c(a)}$ is approximated by the formula $\rho_{c(a)}(T) = \alpha_{c(a)}T + \rho_{c(a)}(0)$. From these figures we notice that the crossover to the nonmetallic regime in ρ_c is obviously linked with the the crossover from the T-linear to the nonlinear regime in ρ_a. The onsets of the anomaly in ρ_a and ρ_c, T_{a0} and T_{c0}, do not exactly coincide—T_{c0} is higher than T_{a0}, perhaps because ρ_c is more sensitive to an underlying mechanism which causes these anomalies or the above approximation for the metallic part is not good for ρ_c. In any case, this correlation is suggestive of the common origin for these crossovers, which leads to the rapid increase of ρ_c/ρ_a in the low-temperature region for oxygen-reduced compounds. It is curious that one physical effect acts on the in-plane and out-of-plane conduction in the opposite direction.

In a previous paper[1] we have given a possible explanation of the in-plane charge transport, based on the correspondence between the deviation from the T-linear dependence in ρ_a and the gap formation in the spin excitation spectrum. The present experimental results, therefore, suggest that the spin gap might be responsible also for the crossover behavior in $\rho_c(T)$. In the region where the spin gap closes, at high temperatures or at higher doping levels, spin fluctuations would give rise to the T-linear in-plane resistivity as well as the metallic T dependence of the interplane conduction. When the spin gap opens (or spin pseudogap deepens), the suppressed spin fluctuations would reduce in-plane scattering, leading to the decrease in ρ_a. However, it is not straightforward that the spin gap might lead to the semiconducting ρ_c.

One possibility is the scenario of the resonating valence bond (RVB) theory,[13,14] where one electron dissociates into a spinon-holon pair in the CuO_2 plane, and the in-plane current is carried by holons which are scattered by spinons. On the other hand, the c-axis transport is dominated by hopping of one physical electron between the planes; a holon combines with a spinon to hop between the planes and again dissociate into the spinon-holon pair in another plane. Thus, a gap (or more appropriately a pseudogap) in the spinon density of states, suppresses the interplane hopping and gives rise to semiconducting $\rho_c(T)$. Recent optical experiments on the 60-K material by Homes *et al.*[15] have revealed that the out-of-plane optical conductivity spectrum $\sigma_c(\omega)$ shows a clear pseudogap behavior—σ_c below $\sim 200\ \mathrm{cm}^{-1}$ is depressed as the temperature decreased below 300 K—while it is not observed in the in-plane spectrum. The pseudogap in the $\sigma_c(\omega)$ spectrum deepens with decreasing temperature and is consistent with the T dependence of the dc transport.

A highly anisotropic Fermi-liquid picture which incorporates interplanar disorder, both static and dynamic, has recently been proposed as a counter model of the non-Fermi-liquid pictures of high-T_c cuprates.[16,17] In this scenario, the c-axis electronic conduction is determined by the competition between direct and random hopping; the former originates from interplane hopping matrix $t_\perp$ which is reduced, e.g., by electronic interactions, the latter from interplanar disorders V, respectively. The interplanar disorders would predominantly arise from the static defects in the CuO chains and from the dynamic out-of-plane phonons (or c-axis spin-fluctuation mode). This picture explains the overall features of ρ_c observed in the present experiment: The interplanar disorder decreases as one approaches $YBa_2Cu_3O_7$, so the direct hopping process would dominate at the optimal doping, giving a metallic ρ_c. Contrary to this, the hopping assisted by the dynamical interplanar disorder, such as phonons, would dominate in the underdoped regime, and as a consequence a negative $d\rho_c/dT$ would result in the T range above T_c. In the same T range, the dynamical interplanar disorder contributes to the in-plane transport as an additional scattering process, making ρ_a nonlinear with T. However, it is not clear whether this mechanism can explain the crossover to metallic T dependence of ρ_c at high temperatures.

There are other mechanisms proposed to explain the nonmetallic conduction in the c direction with emphasis on in-plane fluctuations. One of them incorporates in-plane superconducting fluctuations.[18,19] In a layered superconductor, the in-plane resistivity is reduced, while the out-of-plane hopping is suppressed near T_c, due to superconducting fluctuations. However, the effect of the superconducting fluctuations should become apparent at higher temperature for higher-T_c material, which is just opposite to the observed trend of the crossover. Leggett has proposed an explanation of the c-axis charge dynamics based on the "dynamical dephasing" model.[20] In a highly anisotropic system where the in-plane (t_{ab}) hopping matrix is much larger than the interplane (t_c) one, in-plane thermal fluctuations with energy larger than t_c break the band degeneracy in adjacent planes, therefore, coherent c-axis transport is destroyed. In this scenario, the dephasing occurs for $k_BT \gtrsim t_c$, so it cannot explain the high-T metallic regime of ρ_c either.

We should note that similar features are observed also in $La_{2-x}Sr_xCuO_4$:[21] nonlinear T dependence of ρ_{ab} in the underdoped regime and the high-T metallic to low-T semiconducting crossover in ρ_c at a certain temperature which rapidly decreases with increasing x. In this case the increase in hole density, the Sr substitution, introduces disorder in the LaO layers, contrary to the case of $YBa_2Cu_3O_{7-y}$ where the increase in hole density reduces the disorder in the CuO chains. Thus, the observed progression of the c-axis conduction with doping appears to be a property of the high-T_c cuprates, irrelevant to how disorders are introduced into the system. Different from the present system, the crossover in $La_{2-x}Sr_xCuO_4$ coincides with the structural phase transition[22] and the spin gap such as that in $YBa_2Cu_3O_{7-y}$ has not so far been identified.[23] Detailed investigations of the spin excitations and the connection with the structural distortions are necessary for the $La_{2-x}Sr_xCuO_4$ system.

In summary, we have presented anisotropic transport data of $YBa_2Cu_3O_{7-y}$ for various oxygen contents. Our results reveal the correlation between the semiconducting behavior in ρ_c and the nonlinearity in ρ_a. This correlation is most likely ascribed to the spin gap and suggests that the out-of-plane conduction in $YBa_2Cu_3O_{7-y}$ can be determined predominantly by spin fluctuations.

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