

## Pressure effects on anisotropic resistivity in detwinned $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ : Unconventional carrier doping

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We investigated pressure dependence of the in-plane, chain, and out-of-plane resistivity of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  under hydrostatic pressure  $P$  of up to 2 GPa using detwinned crystals. A dramatic decrease in all the resistive components and the disappearance of  $\rho_c(T)$  upturn with  $P$  can be explained by assuming that applied pressure induces additional carriers in both the  $\text{CuO}_2$  planes and the  $\text{CuO}$  chains, as is similar to the chemical doping. Therefore, the conventional model of the carrier transfer from the chains to the planes is to be reexamined. [S0163-1829(99)50646-2]

One characteristic of the high- $T_c$  cuprates is the quasi-two-dimensional (quasi-2D) electronic state, which is well described as “a confinement of carriers within the  $\text{CuO}_2$  planes” in the normal state and typically seen in the anisotropic resistivity. For example, in the underdoped regime, the out-of-plane resistivity exhibits a steep semiconductorlike upturn at low temperatures,<sup>1,2</sup> whereas the in-plane resistivity remains metallic. In order to clarify the nature of the peculiar quasi-2D electronic state, it is of great importance to understand the mechanism of such a unique conduction, which may be closely related to the pairing mechanism of high- $T_c$  superconductivity.

For the above purpose, applying pressure  $P$  is expected to be a powerful technique, because pressure can control the electronic dimensionality through a lattice compression. In the high- $T_c$  cuprates, almost all the pressure studies have concentrated on how  $T_c$  changes with  $P$ .<sup>3</sup> The  $P$  dependence of  $T_c$  strongly depends on the carrier doping level, where, in many cases, pressure increases the doping level via a carrier redistribution. In  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (Y-123), a source for the carrier redistribution has been considered as a charge transfer of holes from the  $\text{CuO}$  chains to the  $\text{CuO}_2$  planes. This is the so-called pressure-induced charge transfer (PICT) model.<sup>4</sup> Contrary to this, Tozer *et al.*<sup>5</sup> reported that the  $T_c$  of a Y-123 crystal reaches 95.2 K at 11.3 GPa, exceeding the ambient-pressure maximum of 93.5 K. This obviously shows that  $T_c$  is not determined solely by the doping level, and that some key parameters for  $T_c$  other than the doping level are also modified by pressure.<sup>6</sup> Thus the PICT model as well as the  $P$  dependence of physical properties in Y-123 should be further examined.

As is widely accepted, the resistivity of the high- $T_c$  cuprates is a sensitive measure for the doping level. With doping, the magnitude monotonically decreases, and the temperature dependence exhibits a characteristic change.<sup>7,8</sup> Thus precise measurements of the  $P$  dependence of the resistivity will give crucial information on how many holes per  $\text{CuO}_2$  plane are supplied by pressure. In spite of this prospect, there are only a few studies on the  $P$  dependence of the resistivity. Here we report the  $P$  dependence of the in-plane, chain, and

out-of-plane resistivities of detwinned Y-123 crystals under hydrostatic pressure of up to 2 GPa.

We grew high-quality large single crystals of Y-123 using a crystal pulling technique and detwinned them as described previously.<sup>9</sup> Crystals were cut into a rectangular shape and annealed in flowing  $\text{O}_2$  atmosphere for two weeks at 480 and 340 °C for optimal and overdoped samples, respectively.  $T_c$  determined by the midpoint of the sharp resistive drop is 93.1 K for optimal doping and 88.2 K for overdoping at ambient pressure.

The resistivity was measured using a standard four-probe method. The typical size of samples which were cut from a detwinned crystal was  $1 \times 0.3 \times 0.1 \text{ mm}^3$  for  $\rho_a$  and  $\rho_b$ , and  $0.2 \times 0.2 \times 0.4 \text{ mm}^3$  for  $\rho_c$ , respectively. Gold paste was used for electrodes and was cured in air at 300 °C for 5 min. Gold leads were attached to the electrodes with silver paste. We attained a contact resistance of  $< 1 \Omega$ . Pressure was generated by using a piston-cylinder device made of Be-Cu and W-C alloys. Samples are immersed in a fluid pressure-transmitting medium of an equal mixture of Fluorinert FC70 and FC77 in a Teflon cell. This device achieves quasi-hydrostatic pressure. Pressures were calibrated using  $T_c$  of Pb as a standard. Resistivity measurements at each pressure were carried out on heating, after the applied pressure was set at room temperature. In order to keep the pressure constant during the temperature excursion, the load was maintained constant by controlling the oil press equipment. The resistivities after releasing pressure are consistent with the initial ones, which eliminates any deformations or defects in the samples by applying pressure.

Figure 1 shows the  $P$  dependence of  $T_c$  of the two crystals, optimally doped (OpD) and overdoped (OvD) samples. With increasing  $P$ ,  $T_c$  changes linearly up to our highest pressure of 1.93 GPa from 93.1 to 94.5 K with a rate of +0.8 K/GPa for the OpD crystal and from 88.2 to 87.7 K with a rate of -0.3 K/GPa for the OvD one. Interestingly, the transition width becomes narrower with increasing  $P$  for optimal doping, while it becomes broader for overdoping, as shown in the inset of Fig. 1. The transition width of  $< 1 \text{ K}$  at our highest pressure for optimal doping suggests a good uniformity of the applied pressure.

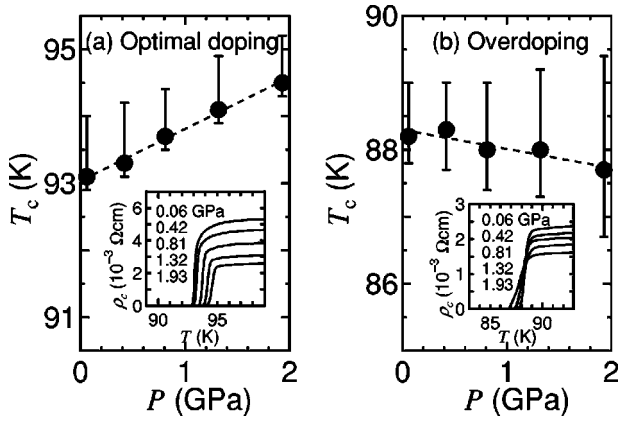


FIG. 1. The variation of  $T_c$  with pressure.  $T_c$  is determined by a midpoint of sharp resistive drop. Each end of the error bars indicates an onset or offset of the transition.

For the OpD sample, we cannot find any indication of saturation of  $T_c$  even at 1.93 GPa, although  $T_c$  exceeds the ambient-pressure maximum of  $T_c$  ( $=93.5$  K) reported so far (see Fig. 1). The observed  $dT_c/dP$  for  $P \rightarrow 0$  is nearly the same as that reported by Tozer *et al.*<sup>5</sup> As they pointed out, this is the direct evidence that  $T_c$  is not determined solely by the doping level. A similar situation is seen in the Hg-based cuprates, where  $T_c$  of an optimally doped sample increases from 130 to 160 K.<sup>10</sup> This increase in  $T_c$  is yet to be understood, and various theoretical explanations are proposed.<sup>3</sup> Since the expression of the  $T_c$  of high- $T_c$  cuprates is open and controversial at present, it will need further compilation of the data.

As mentioned above, since the main purpose of this paper is to see the  $P$  dependence of the resistivity, we measured all three resistive components  $\rho_a$ ,  $\rho_b$ , and  $\rho_c$  of the Y-123 crystals under hydrostatic pressures. Because  $\rho_b$  contains contributions from both the  $\text{CuO}_2$  planes ( $\parallel ab$  plane) and the  $\text{CuO}$  chains ( $\parallel b$ ), with an assumption of isotropic conduction within the  $\text{CuO}_2$  planes, we extract  $\rho_{chain}$  using the expression for two conduction paths in parallel, i.e.,  $1/\rho_{chain} = 1/\rho_b - 1/\rho_a$ . The temperature dependence of  $\rho_{plane}$  ( $=\rho_a$ ),  $\rho_{chain}$ , and the out-of-plane resistivity  $\rho_c$  at several pressures is plotted in Fig. 2 for (a) the OpD sample and (b) the OvD one. In the upper panels,  $\rho_{plane}$  appreciably decreases with  $P$  in both crystals. The decreasing ratio of  $\rho_{plane}$  between ambient pressure and 1.9 GPa is  $-15\%$  for the OpD crystal, and  $-10\%$  for the OvD one, which are almost independent of temperature. These values are much larger than those for simple metals (e.g.,  $-1.9\%/GPa$  for Cu)<sup>11</sup> which is mainly due to the pressure-induced lattice stiffening. Note that in Y-123, the change in carrier density expected from lattice compression is less than  $-1\%/GPa$ ,<sup>4</sup> which is too small to explain the whole magnitude of the decrease in  $\rho$ . We also have observed a large decrease of the Hall coefficient  $R_H$  with  $P$  using samples cut from the same detwinned OpD crystal ( $d \ln R_H/dP = -18\%/GPa$ ).<sup>12</sup> These results clearly indicate that pressure induces additional carriers in the  $\text{CuO}_2$  planes.

The most important finding is that  $\rho_{chain}$  also decreases with  $P$ , which suggests that pressure induces carriers in the  $\text{CuO}$  chains. The decreasing ratio of  $\rho_{chain}$  between ambient pressure and 1.9 GPa is  $-25\%$  for the OpD crystal, and

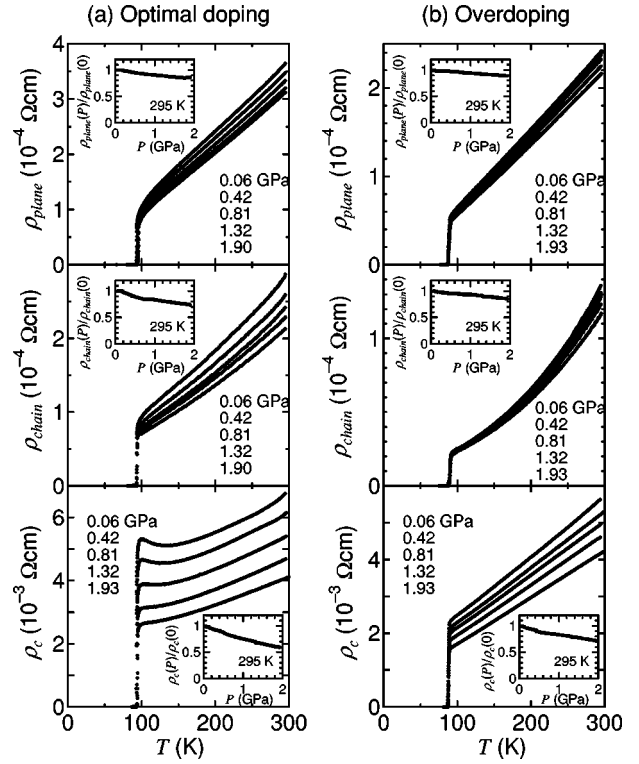


FIG. 2. The temperature dependence of  $\rho_{plane}$ ,  $\rho_{chain}$ , and  $\rho_c$  for (a) the optimal doping and (b) the overdoping at several pressures. The inset shows the pressure dependence of  $\rho_{plane}$ ,  $\rho_{chain}$ , and  $\rho_c$  at 295 K.

$-13\%$  for the OvD one, which are almost independent of temperature. In a naive interpretation following the PICT model, the hole transfer from the chain site to the plane site results in an increase of  $\rho_{chain}$  and a decrease of  $\rho_{plane}$  with  $P$ , which is seriously incompatible with our results. Thus, instead of the PICT model, we propose that the pressure-induced doping is equivalent to chemical doping such as an increase in the oxygen content.

It should be emphasized that the decreasing  $\rho_{chain}$  with  $P$  can be deduced even if we take into account the anisotropy within the  $\text{CuO}_2$  planes. Putting the anisotropy ratio within the planes as  $A = \rho_{plane}^a / \rho_{plane}^b$ ,  $\rho_{chain}$  can be estimated from the relation  $1/\rho_{chain} = 1/\rho_b - A/\rho_a$ . We found that our results of  $\rho_a$  and  $\rho_b$  give the conclusion that  $\rho_{chain}$  decreases with  $P$  for  $A < 2.5$ . In the lowest-order approximation, the  $\text{CuO}_2$  plane will be more resistive for the  $b$  direction, because the lattice parameter for the  $b$  axis ( $3.89 \text{ \AA}$ ) is longer than that for the  $a$  axis ( $3.82 \text{ \AA}$ ). Gagnon *et al.*<sup>13</sup> estimated the upper limit of the anisotropy is 2.0, where  $A = 1/2$ . From the band calculation, Massidda *et al.*<sup>14</sup> found that the  $\text{CuO}_2$  plane of Y-123 is more conductive along the  $b$  direction owing to the existence of the  $\text{CuO}$  chain. Even in this case,  $A = 1.8$  which is less than 2.5.

The substantial difference in  $d\rho/dP$  between the OpD and OvD samples can be explained only by assuming a constant change in the carrier density  $n$  with  $P$ . Since the net change in  $n$  induced by  $P$  is roughly written by  $\Delta n \propto \Delta \sigma \sim -\Delta \rho / \rho^2$ , we found that  $\Delta \rho / \rho^2$  for these two are almost the same. In addition, we found that  $(\Delta \rho / \rho^2)_{chain}$  is two or three times larger than  $(\Delta \rho / \rho^2)_{plane}$ , which is the same as the case for chemical doping reported by Takenaka *et al.*<sup>8</sup>

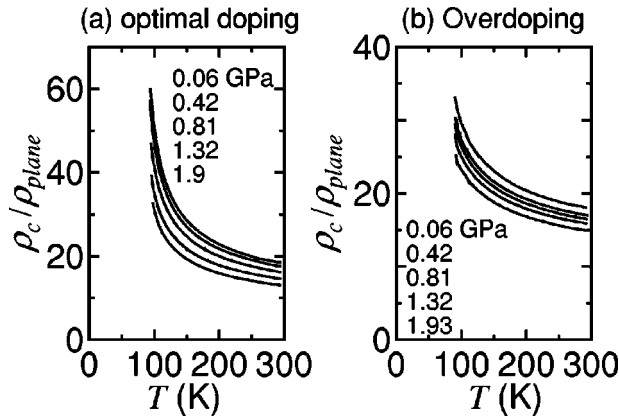


FIG. 3. The variation  $\rho_c/\rho_{plane}$  with  $T$  at several pressures.

According to their data,  $\rho_{plane}$  at 300 K changes from 300 to 200  $\mu\Omega$  cm with increasing oxygen contents from 6.88 to 6.92. Hence we may say that the applied pressure of 1 GPa is roughly equivalent to the increased oxygen contents of 0.01. This is qualitatively consistent with a bond-valence argument that pressure induces +0.0065 holes/GPa.<sup>4</sup>

Another important feature in Fig. 2 is that the decrease of resistivity with  $P$  is more pronounced in  $\rho_c$  than in  $\rho_{plane}$  and  $\rho_{chain}$ . While it is partly explained by an anisotropic compressibility, it also reflects an anisotropic doping dependence in the three resistive components. In fact, the variation of  $\rho_c$  with oxygen contents in Y-123 is more than two times larger than those of  $\rho_{plane}$  and  $\rho_{chain}$ .<sup>8</sup> Moreover, it is worth noting that, for the OpD crystal, the semiconducting upturn at low temperatures diminishes with  $P$ . Since doping by chemical substitutions gives a similar change in  $\rho_c(T)$ , the change in  $\rho_c$  with  $P$  provides the second support for the pressure-induced doping. We can further say that the  $\rho_c$  upturn does not originate from any kind of disorder. This could be a constraint for models of the out-of-plane conduction.

The third support for the pressure-induced doping is given by the change in the anisotropy of  $\rho_c/\rho_{plane}$  with  $P$  (see Fig. 3). Although the increase in the anisotropy ratio with reducing  $T$  indicates that a 2D metallic state with the carrier confinement within the  $\text{CuO}_2$  planes survives even in our OvD sample, the noticeable decrease in anisotropy ratio with  $P$  suggests the increased doping level.

Next, we examine whether pressure affects the scattering rate  $1/\tau(T)$  or not. Figure 4 shows that normalizing  $\rho(T)$  at 295 K gives excellent scaling behaviors for  $\rho_{plane}$  and  $\rho_{chain}$ , while  $\rho_c$  for the OpD sample shows a qualitative change. This suggests that the scattering mechanisms along both the planes and the chains do not change under high pressures. Namely, the pressure does not change the  $T$  dependence of  $1/\tau(T)$ , but increases  $n/m^*$  or the Drude weight. This is consistent with the case of chemical doping reported by Ito *et al.*<sup>7</sup> Note that  $\Delta n$  is as small as 0.005–0.01 per  $\text{CuO}_2$ , which implies that the change in  $1/\tau$  by  $\Delta n$  is too small to be detected.

Note that the excellent scaling behavior implies that the residual resistivity  $\rho_0$  changes with  $P$ .  $\rho_0$  is proportional to  $l_0 S_F$ ,<sup>15</sup> where  $l_0$  is a mean-free path for impurity scattering and  $S_F$  is a Fermi-surface area. Therefore, normally  $\rho_0$  in the Fermi-liquid metals is independent of  $P$ , even in strongly correlated systems. As is seen in  $\text{La}_{2-x}\text{Sr}_x\text{TiO}_3$ ,<sup>16</sup>

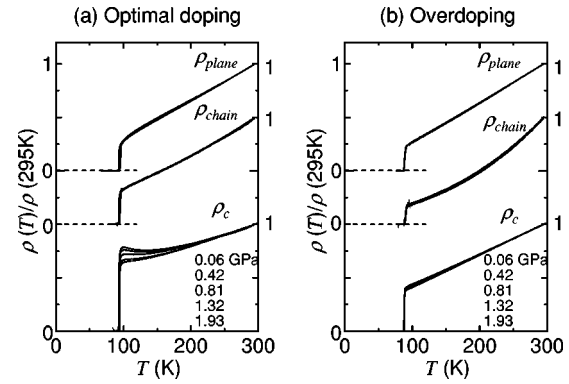


FIG. 4. The variation  $\rho_{plane}$  and  $\rho_{chain}$  divided by each room-temperature value with  $T$  at several pressures.

$\text{Ni}(\text{S}_{1-x}\text{Se}_x)_2$ ,<sup>17</sup> and organic conductors,<sup>18</sup> applied pressure corresponds to an increase of the one-electron bandwidth  $W$  and thus a decrease of the effective electronic correlation  $U/W$ . We propose that the change in  $\rho_0$  of Y-123 is a hallmark of the unconventional pressure effect.

Finally we will make brief comments on unsolved problems. (i) Zhou *et al.*<sup>19</sup> have reported that the PICT model is successful for the pressure effects in  $\text{YBa}_2\text{Cu}_4\text{O}_8$  (Y-124). Their work seems incompatible, particularly in  $\rho_c$ , with a recent work of Hussey *et al.*<sup>20</sup> According to the latter, the conduction band for the chain site governs the transport properties along the  $c$  axis in Y-124. Then, if  $P$  did not change  $\rho_b$ , it would not change  $\rho_c$  either. Since the size of Y-124 crystals is very small, larger crystals are needed to step further. (ii) At present we have no clear answer to the question from where the carriers are induced by  $P$ . A possible candidate is the pressure-induced change of the ionic state or the Madelung potential.

In summary, we have measured and analyzed the pressure effects on the resistivity for the detwinned crystals of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . The decreases of both  $\rho_{plane}$  and  $\rho_{chain}$  with  $P$  are difficult to explain by the model of the pressure-induced charge transfer from the chains to the planes, which has been believed so far. We propose an alternative interpretation of the pressure-induced doping which is similar to chemical doping. This consistently explains our observations that pressure decreases (1) both  $\rho_{plane}$  and  $\rho_{chain}$ , (2) the low-temperature upturn in  $\rho_c$ , (3) the anisotropy of  $\rho_c/\rho_{plane}$ , (4) the residual resistivity  $\rho_0$ , and (5) the Hall coefficient  $R_H$ . This would provide a method for changing a doping level without introducing any substitutions which might cause disorder in the system. In contrast to the normal-state transport properties, the enhanced maximum  $T_c$  clearly shows that applying pressure induces additional changes other than an increase of the doping level, which may provide a crucial insight to a mechanism responsible for the  $T_c$  value.

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