Observation of metallic resistivity behavior following a $1/\rho_{300\ K}$ dependence of T_c in a $YBa_2Cu_3O_{7-x}$ thin film with varying oxygen deficiency

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The normal-state resistivity as a function of T_c of an expitaxial $YBa_2Cu_3O_{7-x}$ thin film has been measured. The superconducting transition temperature T_c of the film was varied by annealing the sample to obtain different oxygen deficiencies. The film showed metallic resistivity behavior even when T_c was reduced to 26 K. We also found that the T_c is inversely proportional to the room-temperature resistivity of the sample ($T_c \propto 1/\rho_{300 K}$). These experimental results are compared with a theoretical prediction based on the Coulomb coupling model and two-dimensional analyses.

As the sample quality of high-T_c superconducting copper oxides has been improved, a remarkable linear temperature dependence of the normal-state resistivity in the Cu-O plane (ρ_{ab}) has been observed. The resistivity can often be approximated by the equation $\rho_{ab}(T)=\rho_0+\alpha T$, where ρ_0 is the residual resistivity. For optimized (fully oxidized) $YBa_2Cu_3O_{7-x}$ (YBCO) samples, many experimental results show that the ρ_0 tends to be zero.[1,2] Consequently, the linear temperature dependence of ρ_{ab} with $\rho_0=0$ has been suggested to be an intrinsic property of the optimized YBCO material, and a deviation from this behavior, if observed, is attributed to extrinsic effects. Obtaining a correlation between normal-state properties and superconducting properties is of great interest in order to gain a better understanding of the superconductivity in layered materials. By making YBCO samples oxygen deficient[3] or by doping or substituting with metal elements such as Zn (Ref. 4) or Pr (Ref. 5) the superconducting transition temperature T_c can be reduced. However, most experimental results show that as T_c is reduced, the temperature dependence of the resistivity deviates from linear characteristics. One usually observes a decrease in α and an increase in ρ_0 with decreasing T_c. Eventually at low temperature, a region develops where ρ increases with decreasing temperature and goes through a peak above T_c (so-called semiconductorlike behavior). Recently two independent experimental results[6,7] have shown metallic resistivity behavior of YBCO even when T_c was reduced to below 30 K, which suggests that the observed semiconductorlike behavior was probably due to extrinsic effects caused by sample preparation. In order to study the intrinsic resistivity of the T_c-reduced YBCO, it is important to perform systematic and careful measurements on high-quality, nearly "ideal" samples.[2]

In this paper, we report on experimental results from a study of ρ_{ab} as a function of T_c from one epitaxial $YBa_2Cu_3\ O_{7-x}$ thin-film sample. Different T_c values were obtained by varying the oxygen content in the same sample through annealing under different conditions. We found that the sample showed metallic behavior with ratios of $\rho_{300\ K}/\rho_{100\ K}$ close to 3 even when T_c was reduced to 26 K. The T_c of the oxygen deficient film was found to be proportional to $1/\rho_{300\ K}$. In this paper we will also briefly compare the experimental results with a theoretical model[2] based on Coulomb coupling and two-dimensional treatment.

The *c*-axis oriented film used in this study was deposited on a 10×10 mm $LaAlO_3$ (100) substrate using our standard *in situ* off-axis sputtering process.[8] A bridge of 0.5 mm in length and 50 μm in width was etched from the 100-nm-thick film for four-probe measurements of critical current density J_c (using a 1 μV criterion), T_c, and resistivity ρ_{ab}. The thickness of the film was determined using both a dectac surface profiler and cross-sectional transmission-electron microscopy. The uncertainty is estimated to be less than 10%. Considering that a thin layer close to the substrate and a layer on the surface of the film might be degraded,[9] measured resistivity data are an upper limit of the true value when using the total film thickness to calculate the resistivity. Silver contacts were evaporated onto the film and gold-coated springs were used to connect the silver on the film and the measuring wires. In this way, contamination from each measurement can be largely limited, which is critical to perform repeated measurements on the same sample. One part of the film (5×6 mm) on the same substrate was reserved for monitoring the crystalline unit-cell length changes by x-ray-diffraction mapping, and it was confirmed that the film was of high epitaxial quality with no detectable high-angle grain boundaries.[10] The as-deposited film (fully oxidized) had a T_c of 88.5 K and a J_c of 3.3×10^6 A cm^{-2} at 77 K. The T_c was determined as the temperature at which the resistivity of the sample vanished (the measurement accuracy is 10^{-5} of the $\rho_{300\ K}$). In order to reduce the oxygen content in a controlled way, the film was annealed in vacuum (1×10^{-5} Torr) at 200 °C for different times and cooled to 80 °C in 30 min. Figure 1 shows the annealing conditions and their effects on the resistivity. By repeated experiments we confirmed that such an annealing process did not degrade the crystalline quality of the film and the original superconducting properties could be recovered by reannealing the sample at 500 °C in 200 Torr pure oxygen.

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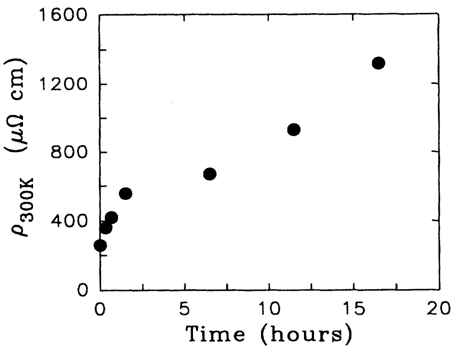

FIG. 1. Room-temperature resistivity $\rho_{300\ K}$ vs isothermal annealing time (cumulated time) curve for the film sample at 200 °C in a vacuum of 1×10^{-5} Torr. The seven data points in this figure can be easily traced in the following figures.

Figure 1 also demonstrates that the annealing is not an equilibrium process but controlled by diffusion. However, using an equilibrium annealing method,[7] we obtained the same results as shown in this report. We therefore confirm that the measured results are not significantly affected by the suspected inhomogeneity of oxygen distribution. We also did not find any room-temperature aging effects as reported by Claus *et al.*[11] for the sample in this study. Since all measured results presented in this paper were from the same thin-film sample, measurement error from sample geometry was avoided.

The resistivity as a function of temperature is presented in Fig. 2 for the same sample after different anneals. Since the film is c axis oriented, the measured resistivity data represent ρ_{ab}. As can be seen from the normalized resistivity data in Fig. 3, the ratio $\rho_{300\ K}/\rho_{100\ K}$ is close to 3 for all measurements. The resistivity curves with T_c values higher than 60 K show roughly a straight line above T_c, and the ρ_0 tends to be zero. When the T_c of the film is reduced to a lower value, an increased deviation from the straight line can be observed. This deviation is, however, smaller than that previously observed. We believe the reason is due to less extrinsic effects introduced during sample preparation in our case. It is not possible for us to speculate if the resulting deviation from

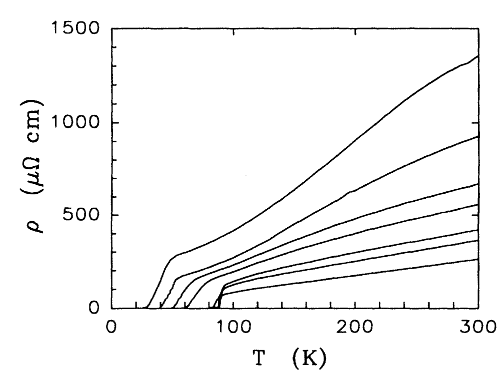

FIG. 2. Resistivity ρ_{ab} vs temperature curves for the thin-film sample.

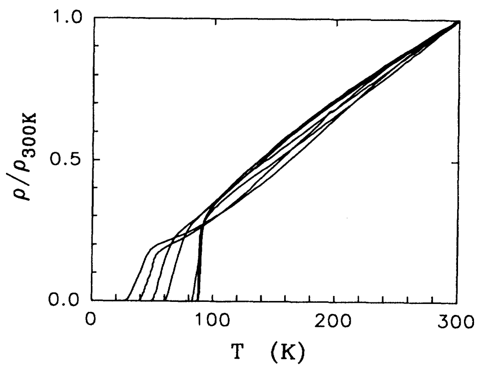

FIG. 3. Normalized resistivity vs temperature curves for the thin-film sample.

linear behavior is due to intrinsic or extrinsic effects. However, we do think it is safe to conclude that intrinsic deviation from linear behavior is not larger than what is observed in our measurements. Therefore, we consider that the equation of $\rho_{ab}=\rho_0+\alpha T$ (with $\rho_0=0$) is an applicable approximation for the intrinsic resistivity behavior of the T_c-reduced YBCO.

The correlation between T_c and $1/\rho_{300\ K}$ is shown in Fig. 4, and the crystal lattice parameters as a function of T_c are shown in Fig. 5. On reducing the oxygen content T_c was initially constant, while the resistivity and the c-axis length increased. This agrees with the observation of the 90 K plateau.[3,12] It is interesting to point out that the J_c value of the film decreased from 3.3 to 0.35×10^6 A cm^{-2} (at 77 K), while the T_c was kept at 88.5 K. The fact that the annealing temperature was rather low suggests that the change in J_c was only caused by oxygen deficiency which in turn produces a decrease in carrier density and/or changes in pinning behavior.

As seen in Fig. 4, when the T_c of the sample starts to decrease, a correlation of T_c proportional to $1/\rho_{300\ K}$ can be observed. Results from studies using Pr-doped bulk single crystals[5] are also included in this figure for comparison. The tendency of their results is similar to ours. The commonly observed semiconductorlike behavior

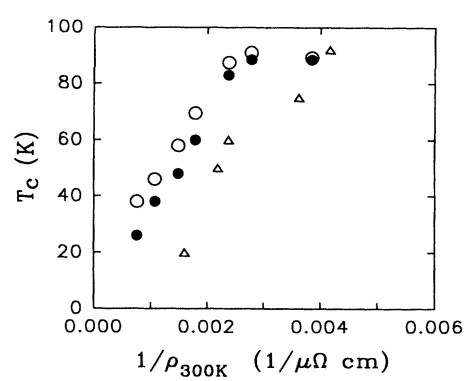

FIG. 4. A plot of T_c as a function of $1/\rho_{300\ K}$. (●) $T_{c0}(\rho=0)$; (○) T_{cm} (the midpoint of the superconducting transition); (△) data from bulk single crystals (Ref. 5).

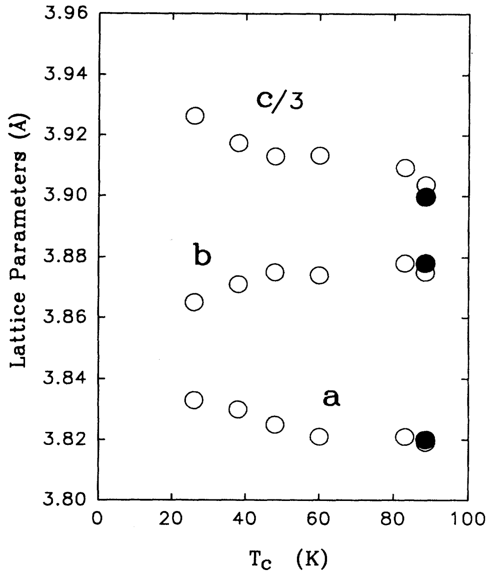

FIG. 5. Lattice parameters as a function of T_c for the thin-film sample. The filled circles represent the parameters of the as-deposited.

caused by extrinsic effects will be significant only at low temperature, and the measured transport properties will be less sensitive to the sample quality at room temperature. Therefore, we believe that the $1/\rho_{300\text{ K}}$ dependence of T_c of the thin-film sample shown in Fig. 4 is a close reflection of an intrinsic property of the YBCO material, and we expect that this behavior can be commonly observed.

Since the temperature dependence of the resistivity of the T_c-reduced film is close to linear, it is interesting to compare our results with a theoretical model that predicts linear behavior. Based on the Coulomb coupling model and two-dimensional treatment, Harshman and Mills[2] gave a relation of $T_c \propto E_F^{2D}$ for optimized high-T_c superconductors, where E_F^{2D} is the two-dimensional Fermi energy. From this they expected a correlation of $\varepsilon\delta/T_c \propto d\rho/dT$ for materials having $\rho_0=0$, where ε is an average dielectric constant and δ is the distance between the two Cu-O planes of the YBCO. If we assume that the resistivity of the T_c-reduced YBCO follows the equation $\rho_{ab}=\rho_0+\alpha T$ with $\rho_0=0$, then our observed relation $T_c \propto 1/\rho_{300\text{ K}}$ becomes $1/T_c \propto d\rho/dT$, and the resistivity can be expressed as $\rho=(\beta/T_c)T$. From this assumption it also follows that β is a constant independent of T and T_c, and that the resistivity just above T_c will be equal to β, that is, $\rho_{T_c}=\beta$, while Harshman and Mills predict $\rho_{T_c}=\varepsilon\delta\beta$. In the experimental data presented in this paper we come relatively close to a constant value of ρ_{T_c}. However, the differences in ρ_{T_c} are still significant; ρ_{T_c} increases by a factor of 3 when the T_c of the film is reduced from 88.5 to 26 K. Based on the observations of the Cu-O plane spacing by Jorgensen[12] and our x-ray-diffraction measurements of the c axis, we conclude that the variation of δ could not be larger than 5% in our case. On the other hand, we do not know if the change in ε can be as large as a factor of 3 when T_c is reduced. Moreover, as mentioned above, we are not able to confirm that our samples show complete intrinsic properties. So far, we cannot make any final conclusion on this comparison with the predictions of Harshman and Mills. Further experiments such as measurements of penetration depth are needed, and are currently in progress.

In conclusion, our experimental results show that the resistivity of T_c-reduced YBCO exhibits a temperature dependency close to linear behavior. The correlation between room-temperature resistivity and T_c has been found to be $T_c \propto$ to $1/\rho_{300\text{ K}}$. We consider such a dependency of T_c as a close reflection of the $YBa_2Cu_3O_{7-x}$ high-T_c superconductor.

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