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Anisotropy of the ion-beam radiation effect in $YBa_2Cu_3O_{7-\delta}$ (110) thin films

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The anisotropy of the radiation effect on normal-state resistivity has been studied on $YBa_2Cu_3O_{7-\delta}$ (110) thin films irradiated by 200-keV proton beams. The resistivity along the Cu-O planes increases with the ion dose while the out-of-plane resistivity decreases and becomes metal-like at a very low dose, where the superconducting transition temperature and the in-plane resistivity show little change. This suggests that the *c*-axis transport properties are sensitive to the structural defects and there exist no direct correlations between the unusual *c*-axis transport behavior and the superconductivity mechanism in $YBa_2Cu_3O_{7-\delta}$ (110) thin films.

The investigation of normal-state transport properties is of crucial importance in an attempt to understand the superconductivity in high-temperature superconductors (HTS). One of the puzzling normal-state transport properties of HTS is the electrical transport anisotropy. All experiments have agreed on a metallic linear temperature dependence of the resistivity along the Cu-O planes (*ab* plane).[1] The results, however, are ambiguous and confusing in the out-of-plane (*c*-axis) direction. Even though most experiments showed semiconductorlike behavior,[2] metallic temperature dependence of resistivity was reported recently on fully oxygenated[3] $YBa_2Cu_3O_{7-\delta}$ (YBCO) and intercalated[4] Bi compounds. It is, therefore, suspected that the unusual *c*-axis transport behavior is only a structural effect and it may have no intimate relation to the superconducting mechanism in YBCO.

Ion-beam irradiation provides a promising method to study this issue on thin-film samples by systematically introducing point defects to perturb the electrical conduction of materials. When the target material is anisotropic, this perturbation effect could be substantially different along each crystalline axis. The investigation of this anisotropic radiation effect, therefore, can provide important clues to an understanding of the physical properties of the target materials. For YBCO, it has been found that resistivity along the *ab* plane can be described[5] approximately by Matthiessen's rule after low-dose ion irradiation, which experiences a metal-insulator transition when the dose is high. A similar result has also been reported on the direct impurity-doped YBCO single-crystal bulk samples.[6] The study of the radiation effect or impurity-doping effect on the *c*-axis conduction, however, is absent because single-crystal thin-film samples with the *c* axis lying in the plane have not been available. Recently, we have successfully fabricated high-quality YBCO (110) thin films.[7,8] One of the unique features of these films is the in-plane alignments of the *c* axis and the [110] axis (or *ab* plane). Direct transport measurements along these two directions can be conducted on the same sample. In this paper, we present data of the radiation effect on the normal-state resistivity of the proton-irradiated YBCO (110) films. Our experiments showed that the *c*-axis conduction is changed from semiconductorlike to metal-like after very low dose proton irradiation, at which T_c and the *ab*-plane conduction stay almost unperturbed. It then suggests that the *c*-axis transport behavior in the normal-state is very sensitive to the structural defects and irrelevant to the superconducting mechanism in YBCO (110) thin films.

Four YBCO (110) thin films made by pulsed-laser ablation on (110) $SrTiO_3$ substrates were used in this experiment. They all showed similar results. The details of the crack-free film fabrication and characterization have been reported elsewhere.[7,8] On each sample, two perpendicular microbridges, with one along the *ab* plane and the other, along the *c* axis, were defined photolithographically and etched by Ar-ion milling. These bridges were 1 mm long and 10 or 20 μm wide. Normal-state resistivity was measured by the standard four-probe technique. The basic features of these samples before irradiation are listed in Table I. In all these samples, the resistivity along the *ab* plane (ρ_{ab}) shows a metallic linear T dependence and that along the *c* axis, semiconductorlike behavior. As we reported earlier,[7] T_c (zero resistance) is the same in both directions. In this study, we used a 200-keV proton beam which has a range (~ 1 μm) much larger than the thickness of the film (~ 0.2 μm) so that the proton-beam-induced point-defects[9] were uniformly distributed throughout the film. Extended defects were significant only for very high-dose irradiation where the nearby point defects agglomerated.

The resistivity along the *ab* plane shows typically metallic behavior after the proton irradiation (Fig. 1). In a

TABLE I. Basic features of four YBCO (110) thin-film samples used in this experiment.

	Sample 1	Sample 2	Sample 3	Sample 4
Phase (110)[a]	97%	99%	99%	98%
T_c (K)	83	84	86.7	86.0
Thickness (Å)	1700	1500	1900	2000
ρ_{ab} (300 K) (mΩ cm)	0.29	0.23	0.22	0.24
ρ_c (300 K) (mΩ cm)	6.7	5.1	4.8	5.6

[a]Impurity phase is mostly (103) YBCO.

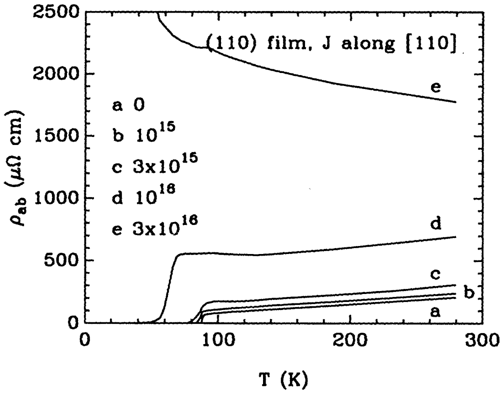

FIG. 1. Temperature dependence of the resistivity along the [110] axis in sample 2 after bombardment with protons at doses of $(0, 1, 3, \text{and } 10)\times10^{15}$ ions/cm^2.

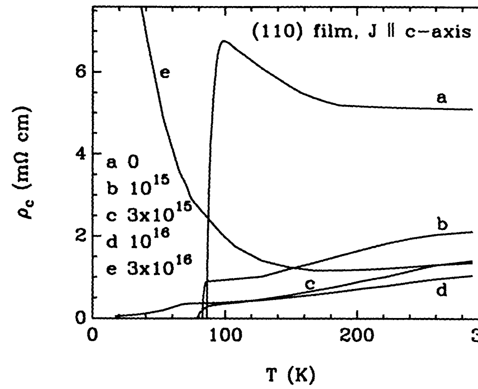

FIG. 2. Temperature dependence of the resistivity along the c axis in sample 2 after bombardment with protons at doses of $(0, 1, 3, \text{and } 10)\times10^{15}$ ions/cm^2.

fairly low-dose range (up to 10^{16} ions/cm^2), ρ_{ab} is metallic and has a finite T_c. The ion-beam-induced point-defects act primarily to increase the residual resistivity which was found to be almost linearly proportional to the proton dose, or the point-defect concentration as in the case of conventional metals. Meanwhile, T_c decreases and finally disappears at a dose of 3×10^{16} ions/cm^2 accompanied by a metal-insulator transition. This observation is not surprising and it agrees well with the measurements on the irradiated c-oriented YBCO thin films[5] and the Zn-doped YBCO single crystals.[6]

What surprises us is the radiation effect on c-axis resistivity. In sharp contrast to the result shown in Fig. 1, ρ_c decreases monotonically as the ion dose increases up to 10^{16} ions/cm^2. At a dose of 10^{15} ions/cm^2, where T_c drops 3.5 K in both directions, ρ_{ab} (300 K) increases 13%, but ρ_c (300 K) decreases about 60%. Moreover, the upper turn near T_c disappears and ρ_c shows a metal-like linear T dependence. In Figs. 1 and 2, T_c is approximately the same for the ab plane and c axis in curves (a), (b), and (c), but different in curve (d) where the proton dose is 10^{16} ions/cm^2. T_c along the ab plane is about 50 K while that along the c axis is impossible to define because of residual resistance. It should be noted that the onset of the superconducting transition is always the same in both directions. One plausible explanation for this T_c anomaly is that the effect of the agglomeration of point defects may be anisotropic so that the percolation of the superconducting phase disappears at different ion doses in these two directions. At a high dose (3×10^{16} ions/cm^2), ρ_c shows a similar insulator behavior to that along the ab plane.

Material amorphization cannot explain this observation. The investigation in a very low-dose range ($10^{13}-10^{15}$ ions/cm^2) indicates a continuous transition of ρ_c, from semiconductorlike to metal-like, as shown in Fig. 3. This transition is completed at 6×10^{14} ions/cm^2 where T_c and ρ_{ab} show little change. It implies that only a small number of point defects are produced which have little influence on the in-plane conduction and pair interaction. The diameter of these defects is estimated to be ~3 Å from the two-dimensional percolation model[12] so they are basically point defects. Based on the Monte Carlo simulation[10] assuming a displacement energy of 20 eV, the displacement per atom (DPA) at this dose is about 0.0035 or one defect per 30 unit cells of YBCO. This DPA is more than one order of magnitude lower than the amorphization threshold (~0.15) suggested[11] for YBCO so that the changeover of ρ_c is not due to material amorphization.

One may argue that some large-size cascades can be generated during the bombardment and they may act as shorts between the Cu-O planes. However, these shorts could contribute to the conduction only if their volume portion was high. It is well known that the probability of the cascade formation is very small for a 200-keV proton beam. Taking the DPA of 0.0035 and the defect dimension of 3 Å, the total volume of defects is 0.3% at a proton dose of 6×10^{14} ions/cm^2. Even if all defects were large-size cascades, it is still unlikely that such a small quantity of cascades could form any kind of connected route based on the percolation theory. Their contributions to the c-axis conduction, therefore, could not be significant. A TEM study[12,13] has confirmed this analysis. It shows no visible defects within the resolution

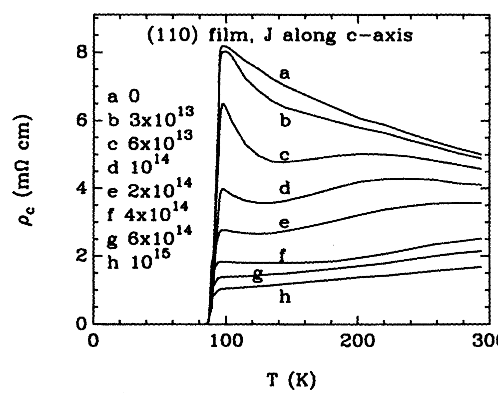

FIG. 3. Resistivity as a function of temperature along the c axis of sample 3 at low doses of $(0, 3, 6, 10, 20, 40, 60, \text{and } 100)\times10^{13}$ ions/cm^2.

($\sim$nm) at 10^{15} ions/cm^2 and only a small number of localized strains at a dose of 3×10^{16} ions/cm^2. The possibility of macroscopic shorts can thus be excluded.

Our experimental results show that electrical transport properties along the *c* axis are very sensitive to the small number of point defects. Since YBCO is a compound involving four elements, there are different kinds of point-defect, vacancies and interstitials due to the displacement of individual Y, Ba, Cu, and O atoms. It has been found[1,3] that $\rho_c(T)$ can be changed from metallic to semiconductorlike by removing oxygen. In the (110) thin films, the slightly low T_c's and the semiconductorlike $\rho_c(T)$ imply a slight oxygen deficiency before the irradiation. The proton beam further knocks out some O (and Y, Ba, and Cu as well) and the number of these point defects is proportional to the scattering cross sections, composition ratio, and the proton dose. Based on the argument of oxygen removal, $\rho_c(T)$ should become more insulating instead of metallic. Our observations, therefore, suggest that the *c*-axis transport mechanism is more sensitive to the metal-element-related point defects than to the oxygen deficiency. Furthermore, the little change in both T_c and ρ_{ab} indicates that the *c*-axis transport behavior has no direct correlation with the superconducting mechanism.

Our experimental results reveal that the electrical transport mechanism along the *ab* plane of YBCO is completely different from that along the *c* axis. The increase of the residual resistance with the point-defect concentration implies that charge carriers transport coherently along the *ab* plane of YBCO as in conventional metals. The abnormal behavior of ρ_c under irradiation is consistent with an incoherent transport mechanism along the *c* axis suggested by theoretical calculations[14,15] in which the strong intralayer correlation in the *ab* plane is found restricting the coherent conduction across the plane, leading to an incoherent transport along the *c* axis. In this case, a small number of point defects introduced by ion-beam irradiation could locally destroy the strong intralayer correlation so that the *c*-axis conduction could be enhanced.

In summary, we have observed the anisotropic radiation-effect on normal-state resistivity in YBCO (110) thin films. Along the *ab* plane, ion-beam-induced point defects act basically to increase the residual resistance at a low-to-medium ion dose. On the other hand, resistivity along the *c* axis decreases as the ion dose increases and starts showing a metal-like linear *T* behavior at a dose as low as 6×10^{14} ions/cm^2. A metal-insulator transition follows in both directions after a high-dose irradiation. Our experiment indicates (1) charge-carrier conduction is coherent along the *ab* plane and differs from that along the *c* axis, (2) a small number of point defects dramatically increase the *c*-axis conduction and change its *T* dependence with little influence on the T_c and the *ab* plane transport properties, and (3) the unusual *c*-axis transport properties have no direct correlation with the superconducting mechanism in YBCO (110) thin films.

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