Superconducting and Normal Conducting Characteristics of $(Y_xPr_{1-x})Ba_2Cu_3O_{7-\delta}$ Thin Films

Masayuki Miyazaki and Yoichi Okabe
Research Center for Advanced Science and Technology, The University of Tokyo
4-6-1, Komaba, Meguro-ku, Tokyo 153, Japan

Abstract—**The ratio of Yttrium (Y) to Praseodymium (Pr) is varied in $(Y_xPr_{1-x})Ba_2Cu_3O_{7-\delta}$ (YPBCO) thin films to consider the relation between superconducting $YBa_2Cu_3O_{7-\delta}$ (YBCO) and insulating $PrBa_2Cu_3O_{7-\delta}$ (PBCO) thin films. The critical temperature (T_c) of the superconducting YPBCO thin film is decreased by substituting more Pr in place of Y, because Pr reduces the carrier density on the CuO_2 plane in YPBCO. The electrical characteristic of the YBCO film can be explained by the weakly coupled chain model. The addition of Pr reinforces the superconducting links among the grains in the YPBCO thin film. The normal state YPBCO film shows two types of transport systems. Behavior similar to impurity semiconductors appears at high temperatures and the variable range hopping mechanism governs their conductance at low temperatures.**

I. Introduction

Most cuprates with a perovskite structure e.g. $YBa_2Cu_3O_{7-\delta}$ (YBCO) show superconducting transitions. Only $PrBa_2Cu_3O_{7-\delta}$ (PBCO) is insulating and not metallic or superconducting among the entire rare-earth 1:2:3 family [1]. Since the perovskite cuprates transfer carriers mainly on the CuO_2 planes, the carrier density on the planes decide their conducting properties. A system with strongly localized carriers shows variable range hopping (VRH) transport properties. Materials in these systems reveal a transition from metallic or superconducting phase to insulating phase depending on the carrier concentration [2]. Therefore, there is a difference of conducting properties between YBCO and PBCO because Yttrium (Y) supplies carriers easily to the CuO_2 plane of YBCO but Praseodymium (Pr) provides few carriers to the plane of PBCO. Nevertheless, the interface between PBCO and YBCO makes a good electrical contact and YBCO/PBCO/YBCO type Josephson junction can show long range coupling [3]–[5]. Recently there have been some reports that the CuO chain of PBCO has the same Fermi surface as YBCO [6] and dominates the electrical properties along with the b-axis [7]. Consequently, it is important for high T_c superconductor research to investigate the roles of Y and Pr in the perovskite cuprates.

We have examined the superconductivity and normal conductivity of $(Y_xPr_{1-x})Ba_2Cu_3O_{7-\delta}$ (YPBCO) thin films. The composition of Y and Pr was varied every 10 % to change the carrier density on the CuO_2 plane and the crystal aspect. As another method to reduce the carrier density of the plane, a deoxygenation process by annealing in vacuum was adopted for a YBCO film. Resistivity properties and AC susceptibilities were measured for YPBCO thin films to estimate the conducting characteristics and crystal structures.

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II. Experimental

$(Y_xPr_{1-x})Ba_2Cu_3O_{7-\delta}$ thin films were prepared where x was controlled from 0.0 to 1.0 in steps of 0.1 concentration. The films were fabricated by RF magnetron sputtering. The conditions for depositions were the following. Stoichiometric targets were used to make each type of film. The orientation was (001) on MgO (100) substrate measured by an X-ray diffractometer. The substrate temperature was 670 °C during the deposition. The total pressure was 30 Pa (Ar : O_2 = 4 : 1). RF power was 150 W. The film thickness was 240 nm. Resistivity versus temperature was measured with the bias current of 10 μA to investigate the electrical characteristics of the YPBCO thin films. The bias current flows along the film surface i.e. parallel to the CuO_2 plane. AC susceptibility was used to characterize the superconductivity of YPBCO thin films. The standard conditions to measure the susceptibility were that the peak value of applied magnetic field was 2.6 μT and the frequency was 313 Hz. The magnetic field was applied perpendicular to the film surface and the induced currents due to the diamagnetic response flow parallel to the CuO_2 plane.

III. Results and Discussion

A. Superconducting $(Y_xPr_{1-x})Ba_2Cu_3O_{7-\delta}$ thin films

The resistivity of the superconducting YPBCO films was measured as a function of temperature as shown in

Fig. 1. The resistivity is normalized at 300 K. The films exhibit a resistivity of about 4×10^{-4} Ω·cm at 300 K. Because Pr provides fewer carriers than Y, it reduces the carrier density on the CuO_2 plane in YBCO superconductor. The critical temperature (T_c) of YBCO is degraded as Pr concentration increases.

The magnetic field amplitude and frequency dependence of susceptibilities were measured for the superconducting YPBCO thin films in order to investigate the quality of weaklinks between grains. The diamagnetic signals obtained from a YBCO film vary with applied field amplitude as shown in Fig. 2. On the other hand, the signals do not vary with the frequency. Such responses are explained with a model reported by Ishida, et al [8]. That is, the YBCO film has many micro scaled grains e.g. twin boundaries, and each grain boundary makes a junction. The model is named the weakly coupled chain model. There exist two phases in YBCO bulk samples as usual [9]–[11]. One is the bulk phase and the other is the coupling phase. The YBCO thin film proves to be a single phase of linked boundaries.

Fig. 3 shows susceptibilities of a YPBCO film with various field where x equals 0.7. The field amplitude reliance disappears in the presence of Pr included in YBCO. There are two possibilities for the result. The first possibility is that Pr among YBCO grains connects the coupling more strongly. The second is that Pr breaks the chain style junctions of YBCO grains. Our argument will be discussed below.

It is clarified how Pr acts in YBCO in comparison with the oxygen deficient YBCO film and the YPBCO film. Then a YBCO film is annealed in 400 °C and 10 Pa oxygen atmosphere to reduce the T_c. Fig. 4 shows the susceptibility of such a YBCO film. The degraded YBCO film shows susceptibilities independent of the applied field similar to the YPBCO film.

The slope of the susceptibility curve between the T_c and the saturating temperature indicates a crystal uniformity of a sample [12]. A sharper slope indicates better coupling between grains. The susceptibility curves of the YBCO and YPBCO films saturate within 10 K as shown in Fig. 2 and 3. The curves of deoxidized YBCO are broader over 30 K as shown in Fig. 4. Therefore, the mechanism of oxygen reducing and Pr substituting are different. Of course, the deficient YBCO makes the crystal uniformity worse and decouples the grains. On the other hand, Pr contained in YBCO acts to reinforce the connection of the grains.

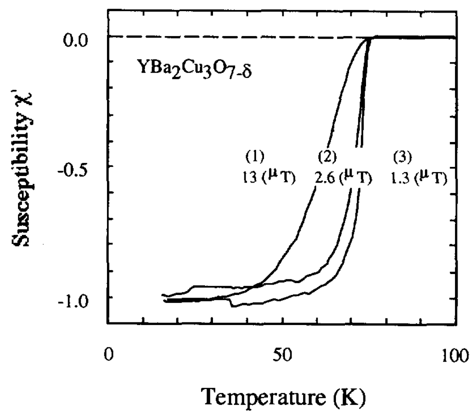

Fig. 2. Real (χ') susceptibilities as functions of temperature for a $YBa_2Cu_3O_{7-\delta}$ thin film. The magnetic field conditions are (1) 13 μT as peak and 313 Hz, (2) 2.6 μT and 313 Hz and (3) 1.3 μT and 313 Hz.

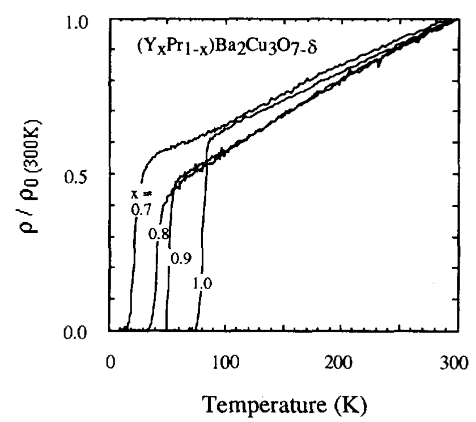

Fig. 1. Normalized resistivity at 300 K vs. temperature curves of $(Y_xPr_{1-x})Ba_2Cu_3O_{7-\delta}$ thin films. Curves show the data for various Y concentration ratios (x) of 70, 80, 90 and 100 % from the left.

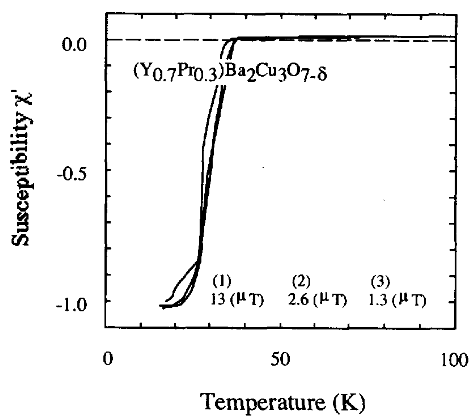

Fig. 3. Real (χ') susceptibilities as functions of temperature for a $(Y_{0.7}Pr_{0.3})Ba_2Cu_3O_{7-\delta}$ thin film. The magnetic field conditions are (1) 13 μT as peak and 313 Hz, (2) 2.6 μT and 313 Hz and (3) 1.3 μT and 313 Hz.

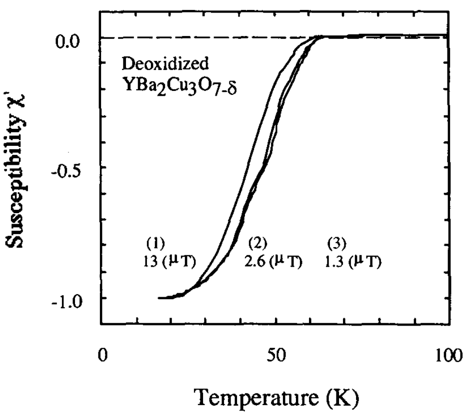

Fig. 4. Real (χ') susceptibilities as functions of temperature for a deoxidized $YBa_2Cu_3O_{7-\delta}$ thin film. The magnetic field conditions are (1) 13 μT as peak and 313 Hz, (2) 2.6 μT and 313 Hz and (3) 1.3 μT and 313 Hz.

B. Normal state (Y_xPr_{1-x})$Ba_2Cu_3O_{7-\delta}$ thin films

The normal state YPBCO films can be considered as a p-type impurity semiconductor at high temperatures. The resistivity for conventional doped semiconductor is a function of temperature [13] given by

$$\rho(T) = \rho_0 \exp\left(\frac{\varepsilon_i}{k_B T}\right) \qquad (1)$$

where ρ_0 is the fitting parameter, ε_i is the ionization energy of acceptor and k_B is the Boltzmann's constant. Fig. 5 shows the resistivity as a function of the inverse temperature ($1/T$) for the YPBCO film substituted Pr for Y. The ionization energy indicated in the resistivity curve of the higher temperature region above 100 K is: 60 meV at x = 0.0; 35 meV at $x = 0.1$; 27 meV at $x = 0.2$; 18 meV at $x = 0.3$ and 11 meV at $x = 0.4$, respectively. Y reduces the ionization energy along with providing carriers on the CuO_2 plane in the YPBCO film.

The VRH mechanism is dominant at low temperatures in the normal state YPBCO film, since the carrier density is decreased thermally. In the two-dimensional case, the resistivity dependence on temperature [14], [15] is given by

$$\rho(T) = \rho_0 \exp\left(\frac{T}{T_0}\right)^{-1/3} \qquad (2)$$

$$T_0 = \frac{8\alpha^2}{k_B N(E_f)\, d} \qquad (3)$$

where ρ_0 and T_0 are the fitting parameters, α^{-1} is the localization radius of states near the Fermi level, $N(E_f)$ is the localized density of states at the Fermi level and d is the film's thickness. Fig. 6 shows the resistivity of the normal state YPBCO films as a function of temperature

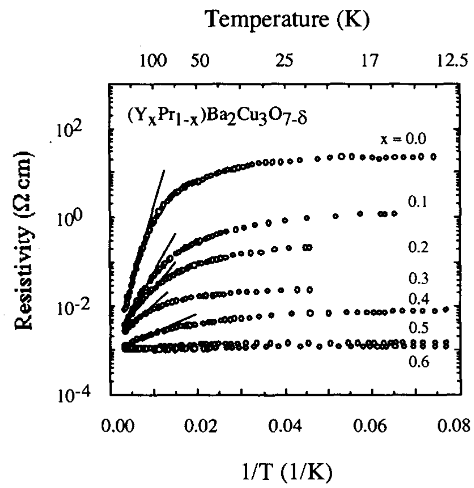

Fig. 5. Semilog plot of resistivity vs. inverse temperature ($1/T$) curves of (Y_xPr_{1-x})$Ba_2Cu_3O_{7-\delta}$ thin films, for various Y concentration ratios (x) varying from 0 % to 60 %.

($T^{-1/3}$) in the two dimensional VRH system. The fitting parameter T_0 presents about the same value of 10^2 K in the lower temperature region below 100 K. T_0 is in inverse proportion to $N(E_f)$. Thus, the density of states $N(E_f)$ is independent of Y and Pr concentration in the YPBCO films. That means there might be also a different conducting phase from the CuO_2 plane transport. It could reveal not only the VRH transmission path on the CuO_2 plane but also the path in the CuO chain.

IV. Conclusions

The resistivity and susceptibility properties of YPBCO thin films are measured in order to examine the effect of Y and Pr on their conductivities and connection between grains.

Pr reduces T_c of YBCO thin films, since the carrier density on the CuO_2 plane determines the superconducting transition and Pr provides fewer carriers on the plane. A YBCO thin film shows a dependence of susceptibility on the amplitude of applied magnetic field. Then the YBCO film includes many grain boundaries coupled as a junction named the weakly coupled chain. The substitution of Y for Pr in superconducting YPBCO film diminishes the dependence on the applied field and keeps the shapes of the susceptibility curves sharp. This indicates the enhanced coupling of grains due to the presence of Pr. Deoxidation of a YBCO film releases carriers from the CuO_2 plane and shows a similar T_c decrease as YPBCO films. However the connection between grains is different between the YPBCO film and the reduced YBCO film. It is apparent from the susceptibility measurement of degraded YBCO that oxygen deficiency makes the YBCO

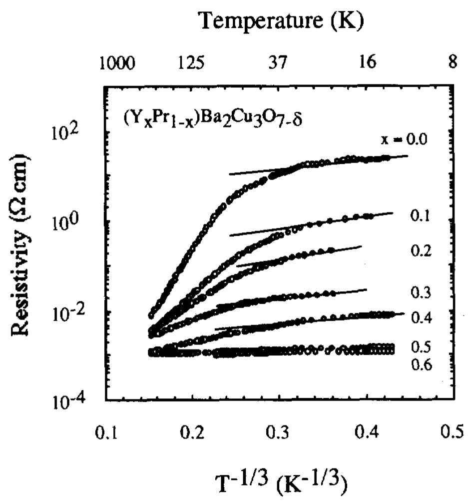

Fig. 6. Semilog plot of resistivity vs. temperature ($T^{-1/3}$) curves of $(Y_xPr_{1-x})Ba_2Cu_3O_{7-\delta}$ thin films, for various Y concentration ratios (x) varying from 0 % to 60 %.

grain links weaker.

The normal state YPBCO films show the impurity semiconductor properties at high temperatures and the VRH characteristics at lower temperatures. The ionization energy decreases as Y is substituted with Pr in the higher temperature region over 100 K. On the contrary, the YPBCO film creates no variation in the localized density of states dependent on Y concentration in the temperature region below 100 K.

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