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Synthesis of low resistive polycrystalline $PrBa_2Cu_3O_7$

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Abstract

We have studied difference in electrical transport properties of polycrystalline $PrBa_2Cu_3O_7$ (Pr123) by changes in synthesis conditions. As raw material we used the carbon free 3N-BaO_2 and the tetravalent Pr free 3N-Pr_2O_3. These raw materials are mixed with CuO and reacted in Ar atmosphere with low oxygen concentration. The temperature dependence of resistivity shows almost semiconductive behavior for all the samples. However, when the samples were prefired in Ar atmosphere with 0.2–0.4% oxygen concentration, the absolute value of resistivity at room temperature decreased to the value close to that of $YBa_2Cu_3O_7$ superconductor. From these results we think that the control of site occupation of Pr atoms is important to give the low resistive Pr123.

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Keywords: Semiconductors; Powder metallurgy; X-ray diffraction; Electronic transport

1. Introduction

$RBa_2Cu_3O_7$ (R123) compounds with R = Y or rare earth element are 90 K superconductors except for R = Pr, Ce and Tb. The suppression of superconductivity by the substitution of Pr123 has attracted great interest since the discovery of the R123 series [1–3]. Our previous study shows that the physical properties of Pr123 are very sensitive to growth condition [4]. We found that some single crystals grown by floating zone method show superconductivity below 80 K. The value of resistivity decreases by the prefiring in Ar with low oxygen concentration [5]. However, we have not yet obtained the growth condition of superconducting samples. One of the origins of growth condition dependence of physical properties may be Pr-on-Ba-site defects. So it is important to make clear the sample preparation process by which Pr atoms perfectly occupy the Pr-site. This time we have tried to make the sample using the carbon free 3N-BaO_2 and the tetravalent Pr free 3N-Pr_2O_3 and succeeded to obtain the polycrystalline sample with the smallest resistivity value.

2. Experiment

Polycrystalline samples were synthesised by the solid-state-reaction method. We used the carbon free raw material 3N-BaO_2 and the tetravalent Pr free 3N-Pr_2O_3. These materials are mixed with CuO and reacted in Ar atmosphere with low oxygen concentration. The oxygen concentration changed from 0.2 to 0.6%. We repeated firing and grinding two times in the atmosphere of low oxygen concentration. After the final firing, the samples were annealed in 100% O_2 atmosphere at 350 °C.

The X-ray powder diffraction measurement was carried out to determine the crystal structure and the quality of samples. We performed measurements of susceptibility by SQUID magnetometer and of resistivity by four-probe method in the range of 5–300 K.

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3. Results and discussion

Fig. 1 shows the dependence of lattice parameters and unit cell volume as a function of prefired oxygen concentration. For 0.2 and 0.3% of oxygen concentration, the crystal structure changes from tetragonal to orthorhombic after the samples were annealed in 100% O_2 atmosphere. It shows that oxygen was in the site of a chain from this result. On the other hand, for over 0.3%, it already becomes orthorhombic structure before annealing.

Temperature dependence of resistivity for samples prepared by different prefired oxygen concentration is shown in Fig. 2. The resistivity of the sample prepared in reduced oxygen atmosphere undoubtedly differs from that of the sample prepared in the air. The results show that the value of resistivity becomes minimum at 0.4% oxygen concentration.

The resistivities at 300 K as a function of prefired oxygen concentration are shown in Fig. 3. The minimum absolute value of resistivity is obtained, when the prefired oxygen concentration ∼0.4% in present work. The previous data show that the minimum resistivity is at 0.2%. The minimum value of this time is over two times smaller than that of previous time [5].

The origins of synthesis condition dependence of transport properties may be Pr-on-Ba-site defects. So we synthesised Pr123 polycrystalline samples in order to reduce the disordering of Pr and Ba, which disorder arrangement delocalizes holes in the narrow band [6] and the resistivity becomes high. We find the synthesis in low oxygen atmosphere and using high quality materials is useful to make lower resistive polycrystalline Pr123 than Y123.

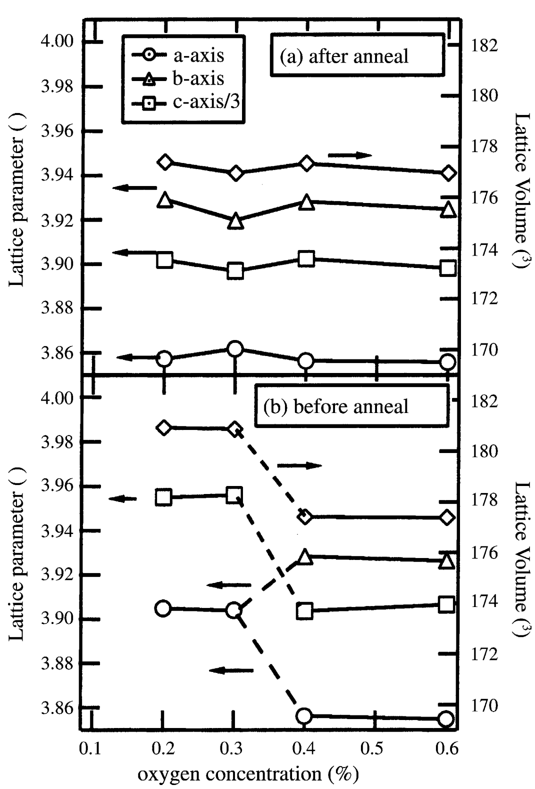

Fig. 1. Lattice parameters and cell volume of orthorhombic unit cell and tetragonal unit cell as a function of prefired oxygen concentration.

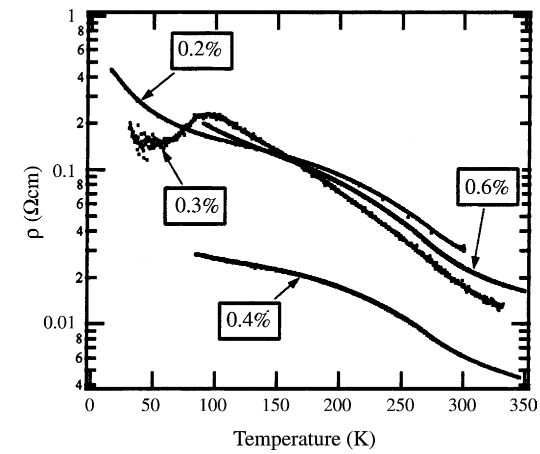

Fig. 2. Temperature dependence of resistivity for different prefired oxygen concentrations.

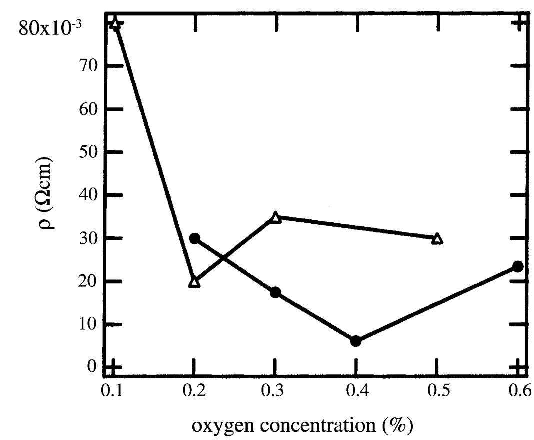

Fig. 3. Resistivity at 300 K as a function of prefired oxygen concentration. The present results of resistivity are plotted as closed circles and the previous results [5] are plotted as open triangles.

4. Conclusion

We have studied the synthesis condition to make the low resistive polycrystalline $PrBa_2Cu_3O_7$. We synthesised the sample with the carbon free BaO_2 and the tetravalent Pr free Pr_2O_3, and reacted in Ar atmosphere with low oxygen concentration. When the samples were prefired in Ar atmosphere with 0.4% oxygen concentration, the value of resistivity decreased to the value close to that of $YBa_2Cu_3O_7$ superconductor. From these results we think that the control of site occupation of Pr atoms is important to obtain the low resistive Pr123.

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