Superconductivity of $(Y_{1-x}RE_x)Ba_2Cu_3O_{7-\delta}$ system

KYUN NAHM, JUNG SOO KIM, KYEONG BOCK LEE, DEOUG WHA SHONG, BO YOUNG CHA, CHUL KOO KIM
Department of Physics, Yonsei University, Seoul 120-749 Korea

Polycrystalline samples of the superconducting oxides $(Y_{1-x}RE_x)Ba_2Cu_3O_{7-\delta}$ (RE = Er and Dy) with $x = 0$, 0.2, 0.4, 0.6, 0.8 and 1.0 were prepared. From the X-ray diffraction and the electrical resistivity measurements, it was found that T_c and the unit cell volume have common peaks near $x \simeq 0.4$. Close examination of previously published data on the $(Y_{1-x}RE_x)Ba_2Cu_3O_{7-\delta}$ system also indicate that an anomaly exists near $x \simeq 0.4$. This anomaly is ascribed to an anomalous change of δ in oxygen near $x \simeq 0.4$.

1. Introduction

Since the discovery of high T_c superconductivity in Y–Ba–Cu-oxide [1, 2], intensive studies have been directed towards the substitution effect of yttrium with other rare earth (RE) elements [3–5] and copper with other 3-d elements [6, 7] in order to understand the correct mechanism leading to the high T_c superconductivity.

In this paper, the temperature dependence of the electrical resistivity and the room-temperature unit cell volume in the series $(Y_{1-x}RE_x)Ba_2Cu_3O_{7-\delta}$ with $x = 0$, 0.2, 0.4, 0.6, 0.8, 1.0, are reported. We have observed that both T_c and the unit cell volume show common peaks near $x = 0.4$ in the series. This finding contradicts earlier reports that either T_c is independent or varies monotonically as a function of x in the $(Y_{1-x}RE_x)Ba_2Cu_3O_{7-\delta}$ [3, 8] system. Thus, we have re-examined the earlier reports and found that the earlier results also exhibit an anomaly near $x \simeq 0.4$ to 0.5.

2. Experimental results and discussion

The samples were prepared following the conventional powder mixing and pressing method under flowing oxygen. The mixture of Y_2O_3, Er_2O_3, CuO and $BaCO_3$ powders was pressed into pellets 1 to 2 mm thick and of radius 3 mm. The pellets were sintered at 950° C for 12 h in flowing oxygen at a rate 200 $cm^3\,min^{-1}$, and cooling from 950 to 350° C was performed very slowly over 8 h in flowing oxygen at a rate of $10^3\,cm^3\,min^{-1}$.

X-ray diffraction pattern measurements were made at room temperature using Cu$K\alpha$ X-rays. The electrical resistivity for the rectangular-shaped samples was measured using a standard four-probe low-frequency a.c. (80 Hz) technique [9].

From the assignment of the 2θ X-ray diffraction patterns, the structure of the main phase in our sample is identified as orthorhombic. Fig 1a shows the relationship between erbium and dysprosium concentration and the unit cell volumes obtained from the lattice constants measurements. Here we observe that the unit cell volume exhibits a maximum around $x = 0.4$ for erbium and $x = 0.6$ for dysprosium.

In Fig. 1b we also observe that T_c has maximums near $x = 0.4$ for both cases. Although T_c or the cell volume has not been shown so far to exhibit a peak as a function of x, it is known that T_c has a nominally linear relationship with the cell volume [10]. Fig. 2 shows that the present data also satisfy this nominal relationship. Therefore, one interesting problem raised by our observation is why some $(Y_{1-x}RE_x)Ba_2Cu_3O_{7-\delta}$ show an anomaly near $x \simeq 0.4$, whereas others have not been reported to possess such a property. To answer this question, we re-examined earlier data on $(Y_{1-x}Pr_x)Ba_2Cu_3O_{7-\delta}$ [8], $(Y_{1-x}Nd_x)Ba_2Cu_2O_{7-\delta}$ [3] and $(Y_{1-x}Lu_x)Ba_2Cu_3O_{7-\delta}$ [11]. The present findings are compared with earlier results on the cell volume as a function of x in Fig. 3. We observe that $(Y_{1-x}Pr_x)Ba_2Cu_3O_{7-\delta}$ has a kink, and $(Y_{1-x}Lu_x)Ba_2Cu_3O_{7-\delta}$ shows an anomalous behaviour around $x = 0.5$.

For the neodymium-doped system, data on the cell volume are too scattered to draw any conclusion. However, the data on T_O (O resistance) in $(Y_{1-x}Nd_x)Ba_2Cu_3O_y$ show that a weak maximum exists near $x \simeq 0.5$ (Table I of [3]). Therefore, we can draw a tentative conclusion that the anomaly around $x \simeq 0.4$ is not an artifact and represents a real physical situation.

The physical origin of this anomaly is a rather difficult question to answer at present. However, the study made by Okai *et al.* [8] on $(Y_{1-x}Pr_x)Ba_2Cu_3O_y$ sheds some light on this question. First of all, they noticed that y has a peak near $x \simeq 0.4$. They also found that T_c decreases monotonically as a function of x and $(Y_{1-x}Pr_x)_{1+\delta}Ba_{2-\delta}Cu_3O_y$ ceases to be superconducting beyond $x \simeq 0.5$. The measured valence of copper also decreases linearly up to $x \simeq 0.4$ and shows an anomalous behaviour beyond that. Therefore, it is quite obvious that the T_c and the cell volume peak we have found in $(Y_{1-x}Er_x)Ba_2Cu_3O_{7-\delta}$ are closely related to the anomalous change of δ in

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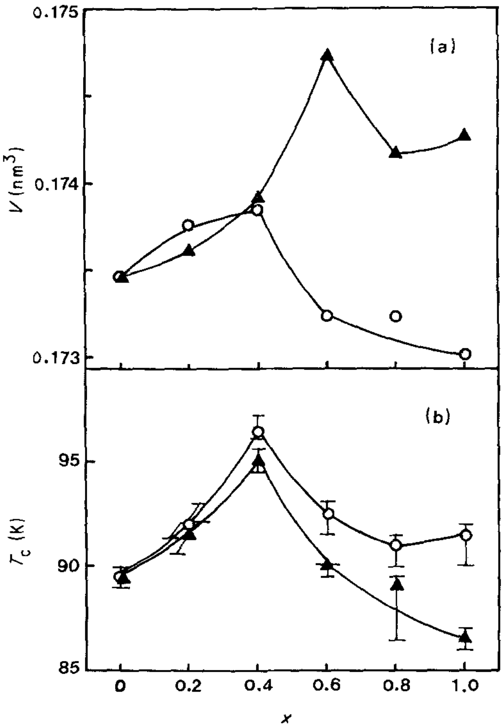

Figure 1 (a) The unit cell volume plotted against (○) Er and (▲) Dy concentration. (b) Superconducting transition temperature, T_c, plotted against (○) Er and (▲) Dy concentration. The upper ends of the bars indicate the onset temperatures (○, ▲) Midpoints of the transition. The lower ends of the bars represent the zero resistance temperatures.

oxygen. However, we do not know at present why T_c decreases again beyond $x \simeq 0.4$ in $(Y_{1-x}Er_x)Ba_2Cu_3O_{7-\delta}$ and $(Y_{1-x}Dy_x)Ba_2Cu_3O_{7-\delta}$, whereas in $(Y_{1-x}Pr_x)_{1+\delta}Ba_{2-\delta}Cu_3O_y$ T_c changes monotonically. It may be argued that the change in barium concentration may have something to do with this behaviour, because the bonding nature of barium may be different for the two cases. We believe that future experiments on the oxygen content, the valence of copper,

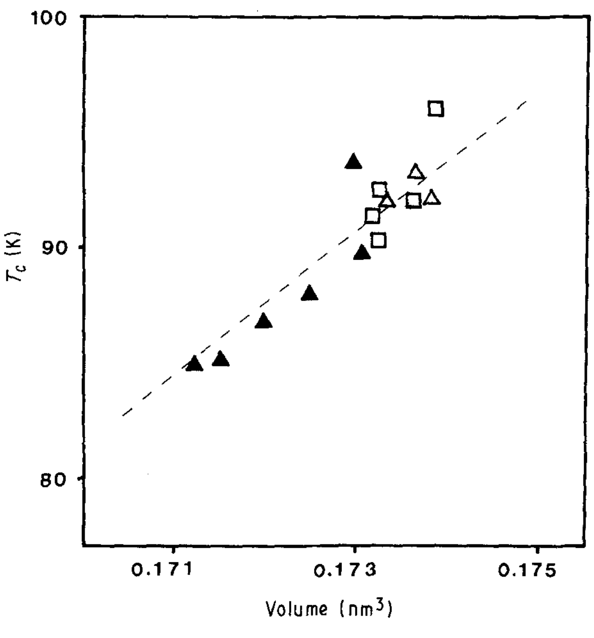

Figure 2 T_c plotted against the cell volume. (□) Present work, (▲), data from $Y(Ba_{1-x}Sr_x)_2Cu_3O_{7-\delta}$, taken from [10], (△) earlier data on $REBa_2Cu_3O_{7-\delta}$ (Re = Y, Er, Dy) from [10].

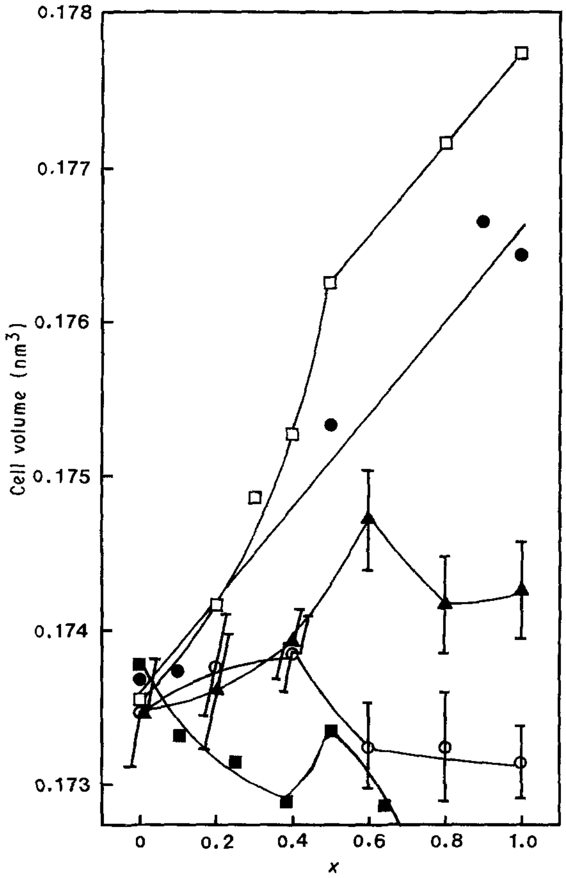

Figure 3 Cell volumes plotted against the rare earth concentration, x. Erbium and dysprosium points are from the present work, praseodymium from [8], neodymium from [3] and lutetium from [11]. (○) Er, (▲) Dy, (□) Pr, (●) Nd, (■) Lu.

and other properties as functions of the oxygen contents in these systems, will provide further clues to this problem. Indeed, preliminary results on the normal state magnetic susceptibility indicate that the paramagnetic Curie temperature and the effective magnetic moment have peaks near $x = 0.4$.

In conclusion, we have found that in $(Y_{1-x}RE_x)Ba_2Cu_3O_{7-\delta}$ (Re = Er, Dy), T_c and the cell volume possess peaks at $x \simeq 0.4$. Re-examination of other data reveals that weaker yet anomalous, behaviour exists in systems with praseodymium and neodymium. We ascribe this anomaly to an anomalous behaviour of δ in the $(Y_{1-x}RE_x)Ba_2Cu_3O_{7-\delta}$ system.

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