

Comparison of the Sr substitution effect on $\text{Nd}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{6+\delta}$ and $\text{Pr}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{6+\delta}$ prepared under reducing atmosphere

Tuerxun Wuernisha^{a,b,*}, Fumihiko Sano^b, Yumiko Takahashi^b, Takashi Kamiyama^a,
Yoshiki Takano^b, Kazuko Sekizawa^b

^a Institute of Materials Structure Science, High Energy Accelerator Research Organization (KEK), 1-1 Oho, Tsukuba, Ibaraki 305-0801, Japan

^b Department of Physics, College of Science and Technology, Nihon University, 1-8 Kanda-Surugadai, Chiyoda-ku, Tokyo 101-8308, Japan

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Abstract

We report on the results of systematic studies of Sr substitution effect for $\text{R}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{6+\delta}$ (where R = Nd, Pr) with $0 \leq x \leq 0.5$, prepared under reducing atmosphere. Single phases were obtained for a wide range of x for both systems of $\text{Nd}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{6+\delta}$ and $\text{Pr}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{6+\delta}$ by means of synthesis at high-temperature under reducing atmosphere (Ar:O₂ = 100:0.1). In particular, solubility as high as $x = 0.5$ was successfully achieved for the first time in the $\text{Pr}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{6+\delta}$. For R = Nd, the superconducting transition temperature, T_c , decreases from 95 to 50 K as x increases from 0 to 0.5. From the Rietveld refinement of powder X-ray diffraction (XRD) data, occupancy of Sr at the Nd site is found, $g_{\text{Sr}/\text{Nd}} (=2 \times g_{\text{Nd}/\text{Ba}})$, and the value of $g_{\text{Sr}/\text{Nd}}$ increases with x from 0 ($x = 0$) to 0.11 ($x = 0.5$). Occupancy of O(4), $g_{\text{O}(4)}$, decreases and O(5), $g_{\text{O}(5)}$, increases with x . It is demonstrated that the depression of T_c with x is caused by the replacement of Ba^{2+} by Sr^{2+} and Nd^{3+} and also by the replacement of Nd^{3+} by Sr^{2+} . Disorder in the BaO plane induce the disorder in the CuO plane, which results in the decrease of the orthorhombicity. For R = Pr, both of the electrical resistivity and antiferromagnetic ordering temperature, T_N , increase with x until $x = 0.3$, then decreases with x . Refinement results do not show intermixing of Sr and Pr; Sr ions occupy only the Ba site and Pr ions does not occupy the Ba site. The dependence of the orthorhombicity on x is quite different from that of Nd-system. The crystal becomes tetragonal at $x = 0.4$, which is smaller than that of Nd-system. The hybridization between the Pr4f and the O2p orbitals which prefers the tetragonal symmetry is considered to advances the transition to the tetragonal structure.

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1. Introduction

Extensive research efforts have been directed toward the study of high-temperature superconducting cuprates ever since its discovery in early 1986 [1]. It is well recognized

that the physical properties of such compounds are closely related to the preparation conditions of them.

High T_c superconductivity is carried by holes on the CuO_2 plane, in which the holes show strong correlations between them. The electronic states of the CuO_2 plane are delicate and influenced by structural characteristics, such as the radius, valence and distribution of surrounding ions, the oxygen content and the corrugation of the plane itself. Then, it is interesting to clarify the effects of the above characteristics, especially the effect of the distribution of ions. In addition, the distribution of ions is strongly

* Corresponding author. Address: Institute of Materials Structure Science, High Energy Accelerator Research Organization (KEK), 1-1 Oho, Tsukuba, Ibaraki 305-0801, Japan. Tel.: +81 29 864 5609; fax: +81 29 864 3202.

E-mail address: urnisa@post.kek.jp (T. Wuernisha).

influenced by heat-treatments applied during the course of sample preparation. In particular, when R is a light rare-earth element, such as La, Pr or Nd, various effects are expected because of large ionic radii of the R ions. $\text{PrBa}_2\text{Cu}_3\text{O}_y$ is not superconducting, and the mechanism of the suppression of superconductivity has not yet been clarified. Both $\text{LaBa}_2\text{Cu}_3\text{O}_y$ and $\text{NdBa}_2\text{Cu}_3\text{O}_y$ show superconductivity with T_c values higher than 90 K only when they are prepared under an optimum condition. For example, in $\text{LaBa}_2\text{Cu}_3\text{O}_y$ synthesized in an inert gas [2] its T_c could be achieved up to 98 K.

So far, many studies have been made on Sr substituted $\text{R}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{6+\delta}$ [3–7]. For a given R ion, T_c and the orthorhombicity, $(b-a)/(a+b)$, decreases with increasing Sr concentration [3,4,8]. For given Sr concentration, both the crystal structure and T_c depend on the ionic size of the R ions [9]. However, details of the previously reported experimental data did not agree with each other, probably because of the different ion distribution due to the different heat-treatment condition. For example, in the case of $\text{Pr}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{6+\delta}$, the Sr substitutions were limited to 0.4 or less by the solid solubility [6,10,11]. We have recently reported the systematic study on the effects of Sr substitution in $\text{Nd}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{6+\delta}$ prepared under various sintering conditions [7]. The optimum sintering condition to give the highest T_c was sintering at as high temperature as possible under a reducing atmosphere. Sintering under reducing atmosphere improve the cation ordering. Thus, we are interested in the correlation between the physical properties and the ion distribution in the Sr substituted compounds of Nd and Pr which are on the broche line for the occurrence of superconductivity; prepared under the condition considered to be best for the ionic ordering.

In the present study, the samples have been synthesized at high temperature under reducing atmosphere ($\text{Ar}:\text{O}_2 = 100:0.1$) in order to obtain higher solubility of Sr in $\text{R}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{6+\delta}$ (where R = Nd, Pr). The powder XRD measurement, thermogravimetric analysis (TGA), electrical resistivity and DC magnetization measurements have been carried out. Correlation among T_c , hybridization of Pr4f with O2p and crystal structure will be discussed.

2. Experimental

Polycrystalline samples of $\text{Nd}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{6+\delta}$ (Nd-system) and $\text{Pr}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{6+\delta}$ (Pr-system) with $0 \leq x \leq 0.5$ were synthesized by the solid-state reaction under a reducing atmosphere ($\text{Ar}:\text{O}_2 = 100:0.1$). The ingredients Nd_2O_3 , BaCO_3 , SrCO_3 , CuO and Pr_6O_{11} of 4 N purity were weighted in composition ratio and well mixed. They were then preheated in Ar in a temperature range of 950–980 °C (R = Nd) and 900–925 °C (R = Pr) for 20 h, and furnace cooled to room temperature. The sintered powders were ground, pressed into pellets at 800 MPa and heated again under the same conditions. This

process was repeated at least twice with intermediate grinding. The sintering temperatures were gradually increased with increasing Sr amount from 950 °C (for $x = 0$) to 980 °C (for $x = 0.5$) for R = Nd, and 900 °C (for $x = 0$) to 925 °C (for $x = 0.5$) for R = Pr. In the final sintering and oxygenation, samples were heat treated in a flowing $\text{Ar}:\text{O}_2 = 100:0.1$ gas at 950 °C (R = Nd) or 900 °C (R = Pr) for 10 h, slowly cooled to 440 °C in the same atmosphere, and kept at this temperature for 60 h under flowing pure oxygen and then cooled to room temperature.

Powder XRD data were collected at room temperature by using a diffractometer Rigaku RINT1100 with a $\text{CuK}\alpha$ radiation source. The crystal structure has been refined by Rietveld analysis of powder XRD data. The electrical resistance was measured by employing a standard dc four-probe technique. Data were taken from 300 down to 4 K. T_c was determined from the mid-point of the resistivity change at the transition. For each sample, the total amount of oxygen was determined by TGA under a reducing gas flow (10 at.% $\text{H}_2 + 90$ at.% He). DC magnetization was measured in the temperature range from 3 to 303 K using Quantum Design PPMS 7N system under the magnetic field of 1 T. The antiferromagnetic ordering temperature, T_N , was determined by the point where the gradient, $d\chi_m/dT$, changed.

3. Results and discussion

Fig. 1(a) and (b) shows the powder XRD patterns of the Nd- and Pr-systems, respectively. These diffraction patterns confirmed that the Sr substituted sample up to $x = 0.5$ in both systems was still a single phase, and no impurity phase was observed. It should be noted that, the solubility range of Sr in $\text{Pr}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{6+\delta}$ is $0 \leq x \leq 0.5$. As described later in detail, the value of the solubility ($x = 0.5$) found in this study is much higher than the other reports [6,10,11]; it is the highest solubility obtained until now.

According to our knowledge, several reports are available concerning $\text{Pr}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{6+\delta}$. However, it has to be mentioned that the processing conditions were not yet optimized in the previous work. For example, Zhao et al. [6] reported that the solubility range of Sr in $\text{Pr}(\text{Ba}_{1-x}\text{Sr}_x)_2\text{Cu}_3\text{O}_{6+\delta}$ was $0 \leq x \leq 0.4$ with an orthorhombic-to-tetragonal (O–T) phase transition at $x = 0.1$. Song et al. [10] reported that the solubility range was $0.1 \leq x \leq 0.3$ (the impurity phase BaCuO_2 appeared at $x = 0, 0.1$) with the O–T transition in $0.15 \leq x \leq 0.3$. That is, the highest value is $x = 0.3$ under their experimental conditions (in air). Furthermore, Sonobe and Yoshida [11] reported that $x = 0.25$ is the solubility limit of Sr at the Ba site with the O–T transition at $x = 0.2$ (in air), or without the O–T transition (Ar annealing). In the present study, we have attained the highest solubility limit of $x = 0.5$ with the O–T transition at $x = 0.3$ by the final sintering under reducing atmosphere ($\text{Ar}:\text{O}_2 = 100:0.1$). It will probably be the optimal condition for the cation ordering for the Pr-system. These results suggest that the reducing

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