An anomalous dip in thermoelectric power of $Nd_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$

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Abstract

The thermoelectric power, S, has been studied for sintered samples of $Nd_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ with $0 \leq x \leq 0.30$ in the temperature range from the superconducting T_c to room temperature. S increases with decreasing temperature, and has a broad maximum at T^{max} in the region around 120 K before decreasing strongly when T_c is approached. Several properties indicate a decrease of charge concentration with decreasing doping, x, from $S(x, 290K)$, T^{max}, and the resistivity $\rho(x, 290K)$. An anomaly has been observed in $S(T)$ for $x \geq 0.20$ in the form of a dip at 78 K of order 15% of S. The origin of this feature is not known.

Key words: Superconductivity ;Thermoelectric power ;Pr doping ;

1. Introduction

In the RE-123 (RE=rare earth element) superconductors the RE's can be exchanged for each other with only small effect on the critical temperature T_c with the exception of Pr substitution which decreases T_c dramatically. Different explanations [1–4] for the depression of T_c have been suggested. Varying results for the valence of Pr is one reason. Recent neutron diffraction results in Nd(Pr)-123 [5] and analysis of T_c in Y(Pr)-123 [6] have suggested that hole localisation in the Pr^{4+} site is the main reason for the suppression of superconductivity by Pr. The observation of superconductivity in single crystals of $PrBa_2Cu_3O_{7-\delta}$ [7,8] further complicates the understanding of the role played by Pr in the suppression of T_c. In our previous work [5], resistivity measurements in $Nd_{1-x}Pr_xBa_2Cu_3O_7$ showed a decreasing metallic behaviour with increased resistivity and decreased slope of the normal state resistivity vs. temperature with increasing x. Neutron diffraction data indicated that the Cu1-O4 and Cu2-O4 distances and the oxygen content were independent of Pr doping in Nd(Pr)-123. Bond Valence Sum (BVS) calculations showed a constant Cu2 valence but a decrease of the total hole concentration in the CuO_2 plane. To further investigate the effect of Pr in $Nd_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ system, we have measured the thermoelectric power $S(x, T)$.

2. Sample characterisation and experimental

Samples of $Nd_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ (with $x = 0, 0.05, 0.10, 0.15, 0.20$, and 0.30) were prepared by standard solid-state methods. Starting materials were high purity Nd_2O_3, $BaCO_3$, CuO, and Pr_6O_{11}. The samples were pressed into pellets and calcinated in air at 900, 920, and 920 °C with intermediate grindings. They were then annealed in flowing oxygen at 460 °C for 3 days and the temperature was finally decreased to room temperature at a rate of 12 °C/hr.

The samples were characterised by X-ray powder diffraction (XRD). The XRD patterns were recorded in a Guinier-Hägg focusing camera using CuK_α radiation with Si as an internal standard [9]. The XRD results for Pr-doped Nd-123 samples displayed single-phase behaviour. All XRD patterns were indexed with an orthorhombic unit cell.

The electrical resistivity was measured with a standard dc four-probe method. Electrical leads were at-

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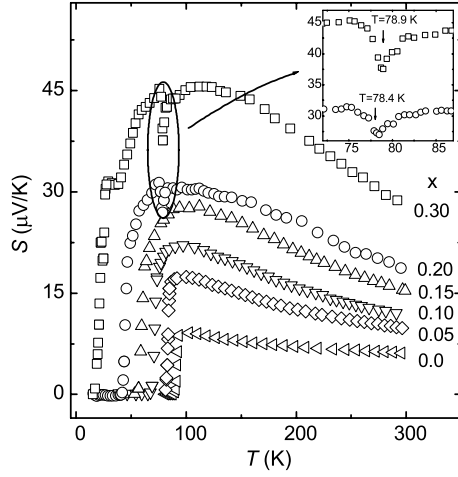[1] Corresponding author. E-mail address:ghorbani@ftf.kth.se

Fig. 1. Thermoelectric power S as a function of temperature and Pr doping for $Nd_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$. Inset: $S(T)$ in the temperature range 72-87 K, where an anomaly has been observed in the form a dip, for x=0.20 and 0.30.

Fig. 2. $S(290K)$ (left scale) and $\rho(290K)$ (right scale) vs. Pr doping, x.

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tached to the sample by silver paint and heat treated at 300 ^{o}C in flowing oxygen for half an hour, which gave contact resistances of order 1-2 Ω . Thermoelectric power measurements were made on sintered bars of typical dimensions $0.5 \times 2.5 \times 10 \ mm^3$, using a small, reversible temperature difference of 1.5 K.

3. Results and discussion

Figure 1 shows the thermoelectric power S as a function of temperature and Pr concentration for $Nd_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$. S first increases with increasing temperature towards a broad maximum at T^{max} above the superconducting T_c, and then decreases up to room temperature. Both S and T^{max} increase with increasing x. As further illustrated in the inset of Fig. 1, a dip in $S(T)$ was observed at a temperature in the range 78-79 K. This feature only occured for $x \geq 0.20$. These anomalies are reproducible. The origin of the anomaly is not known. Except for the dips, all studied properties of the samples with $x \geq 0.20$ (e.g. $S(290K)$, $\rho(290K)$, T_c, T^{max}, and BVS calculations) are in agreement with the trends expected from the results for $x < 0.20$.

The doping concentration dependence of the room temperature $S(290K)$ and resistivity $\rho(290K)$ are plotted in Fig. 2. Both $S(290K)$ and $\rho(290K)$ show a continous increase with increasing Pr doping. The results for $S(290K)$, T^{max}, and $\rho(290K)$ suggest that Pr reduces hole concentration in the CuO$_2$ plane. This is well in agreement with BVS calcu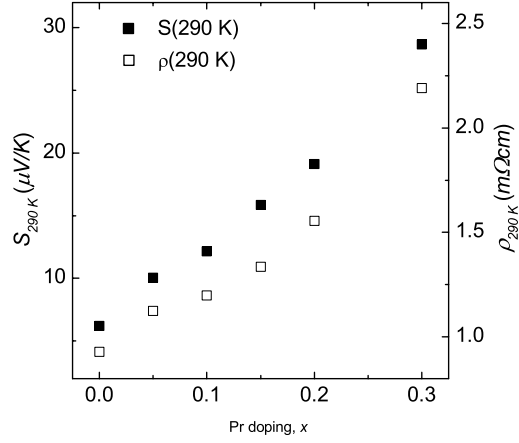lations from neutron diffraction data [5].