An anomalous dip in thermoelectric power of Nd$_{1-x}$Pr$_x$Ba$_2$Cu$_3$O$_{7-\delta}$

S. R. Ghorbani$^a$,1, ¨O. Rapp$^a$

$^a$Solid State Physics, Department of Microelectronics and Information Technology, KTH Electrum 229, SE-164 40 Kista, Sweden

Abstract

The thermoelectric power, $S$, has been studied for sintered samples of Nd$_{1-x}$Pr$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ 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 $T^{\text{max}}$ 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 $S(x, 290\text{K})$, $T^{\text{max}}$, and the resistivity $\rho(x, 290\text{K})$. 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$_2$Cu$_3$O$_{6-\delta}$ [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$_x$Ba$_2$Cu$_3$O$_7$ 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$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ system, we have measured the thermoelectric power $S(x, T)$.

2. Sample characterisation and experimental

Samples of Nd$_{1-x}$Pr$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ (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$_2$O$_3$, BaCO$_3$, CuO, and Pr$_6$O$_{11}$. The samples were pressed into pellets and calcinated in air at 900, 920, and $920^\circ\text{C}$ 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$_\alpha$ 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-
attached to the sample by silver paint and heat treated at 300 °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 0.5×2.5×10 mm³, 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$_x$Ba$_2$Cu$_3$O$_{7-δ}$. $S$ first increases with increasing temperature towards a broad maximum at $T_{\text{max}}$ above the superconducting $T_c$, and then decreases up to room temperature. Both $S$ and $T_{\text{max}}$ 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 occurred 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. $S(290\,\text{K})$, $\rho(290\,\text{K})$, $T_c$, $T_{\text{max}}$, 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 $S(290\,\text{K})$ and resistivity $\rho(290\,\text{K})$ are plotted in Fig. 2. Both $S(290\,\text{K})$ and $\rho(290\,\text{K})$ show a continuous increase with increasing Pr doping. The results for $S(290\,\text{K})$, $T_{\text{max}}$, and $\rho(290\,\text{K})$ suggest that Pr reduces hole concentration in the CuO$_2$ plane. This is well in agreement with BVS calculations from neutron diffraction data [5].

Acknowledgements

We would like to thank I. Bryntse and M. Valldor, Stockholm University, for help with sample preparation and X-ray analysis. Financial support from the Swedish Agencies Vetenskapsrådet and the SSF Oxide Consortium, and from the Iranian Ministry of Science, Research, and Technology are gratefully acknowledged.

References