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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 $0 \leqslant x \leqslant 0.30$ 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 increasing doping, x, from $S(x, 290\ K)$, T^{max}, and the resistivity $\rho(x, 290\ K)$. An anomaly has been observed in $S(T)$ for $x \geqslant 0.20$ in the form of a dip at 78 K of order 15% of S. The origin of this feature is not known.

Keywords: 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 three days and the temperature was finally decreased to room temperature at a rate of 12 °C/h.

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

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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 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 $0.5 \times 2.5 \times 10$ mm^3, using a small, reversible temperature difference of 1.5 K.

3. Results and discussion

Fig. 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 occurred for $x \geqslant 0.20$. The proximity to the boiling point of liquid nitrogen might suggest an experimental artefact from nitrogen boil off. Such an effect appears to be unlikely however, since reproducible dips in S were observed in repeated experiments only for $x \geqslant 0.20$ and not for the other samples. The origin of the anomaly is not known. Except for the dips, all studied properties of the samples with $x \geqslant 0.20$ (e.g. S(290 K), ρ(290 K), 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(290 K) and resistivity ρ(290 K) are

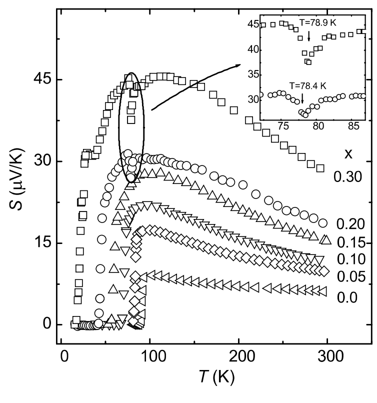

Fig. 1. Thermoelectric power S as a function of temperature and Pr doping for $Nd_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$. Inset: $S(T)$ in the temperature range 72–87 K, where an anomaly has been observed in the form a dip, for $x = 0.20$ and 0.30.

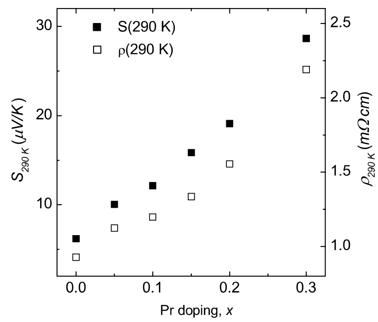

Fig. 2. S(290 K) (left scale) and ρ(290 K) (right scale) vs. Pr doping, x.

plotted in Fig. 2. Both S(290 K) and ρ(290 K) show a continuous increase with increasing Pr doping. The results for S(290 K), T^{max}, and ρ(290 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].

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