Non-ohmic resistance characteristic of inhomogeneous structure in Pr-doped cuprate Nd-123

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Abstract. Electrical characteristics of the Pr-doped cuprate Nd-123 (NdPr-123) have been investigated in reference to an inhomogeneous structure consisting of Nd-123 and Pr-123 phases. The superconductor-insulator (S-I) transition takes place there when Pr-concentration \( x \) exceeds a threshold point \( x_c \approx 0.32 \). At \( x \) just below the threshold point \( x_c \), the resistance has been found to become highly non-ohmic, appearing as an increasing function of the current \( I \). In contrast, at \( x \) much higher than \( x_c \), the resistance turns into a decreasing function of \( I \). The non-ohmic resistance having such dual aspects was observed also for another series of samples of NdPr-123 that was prepared in a different way and had a much lower threshold \( x_c \approx 0.14 \). For this degraded samples, the resistance becomes non-ohmic more easily, indicating that the structural inhomogeneity is closely related to the non-ohmic behaviour. The non-ohmic resistance can be explained in terms of the current distribution governed by a percolation process pertain to the inhomogeneous structure consisting of unit cells of Nd-123 and Pr-123.

1. Introduction

The Y-123 type cuprate \( \text{NdBa}_2\text{Cu}_3\text{O}_7-\delta \) (Nd-123) is a superconductor having \( T_c \) (onset) \( =96 \) K, while \( \text{PrBa}_2\text{Cu}_3\text{O}_7-\delta \) (Pr-123) is an anti-ferromagnetic insulator having semiconductor-like resistance as a decreasing function of temperature \( T \). In the chemical aspect, both Nd-123 and Pr-123 are isomorphic in crystallography, and so merge well with each other [1, 2]. Therefore, Pr-doped cuprate \( \text{Nd}_{1-x}\text{Pr}_x\text{Ba}_2\text{Cu}_3\text{O}_7-\delta \) (NdPr-123) forms a solid solution where the unit cells of Nd-123 and Pr-123 mix with each other on nanometric scale. This nanostructural mixture can be regarded as a percolation system consisting of unit-cells of Nd-123 and Pr-123 [1], although its percolation process is not necessarily identical to the classical one as seen for usual composites [3, 4]. Common to such Pr-doped curates of type Y-123, the superconductivity degrades with increasing Pr-concentration \( x \), and eventually disappears entirely at the percolation threshold \( x_c \). This superconductor-insulator (S-I) transition just refers to the percolation transition with this threshold \( x_c \).

In early studies this superconducting behaviour was described mainly in terms of the depletion in the hole concentration or the pair breaking by the magnetic moment of Pr ion [5, 6]. Those models are based on the assumption that the doped cuprates have homogeneous structures. However, such conventional view seems to be unable to describe a variety of the transport properties of the doped cuprates. The following non-ohmic resistance is one of such typical phenomena that are unable to describe if we assume the homogeneous structure. As reported previously, we have shown that in such Pr-doped curates (typically in GdPr-123) the resistance noticeably depends on the supplied current \( I \),...
which appears as an increasing function of $I$ in the range $x<x_c$ and as a decreasing function of $I$ in the range $x>x_c$, and pointed out that the structural inhomogeneity is responsible for this non-ohmic phenomenon [7, 8].

In the present work, we have investigated the transport properties of Pr-doped cuprate NdPr-123 with two series of samples that were prepared in different ways; one series of samples of high quality (samples-A) shows the zero-resistance temperature $T_0=91$ K for $x=0$ and the S-I transition at $x_c=0.32$, while another series of low quality (samples-B) exhibits $T_c=70$ K for $x=0$ and the S-I transition at $x_c=0.14$. By comparison between two series of samples-A and -B, we aim at confirming the validity of the structural inhomogeneity responsible for the non-ohmic resistance.

2. Experimental procedure
A series of NdPr-123 samples (samples-A) was synthesized by the solid reaction method from a stoichiometric mixture of raw powders Nd$_2$O$_3$, Pr$_2$O$_{11}$, BaCO$_3$, and CuO, through the cyclic process (three-times repeated) of pulverizing, mixing and subsequent calcining at 925°C for 24 hour in airs. The resultant specimens were then sintered at 930°C for 24 hour in an oxygen flow and subsequently annealed at 400°C for 24 hours in the same furnace. The Pr-concentration $x$ ranged as $0.82 \leq x \leq 0.0$. The series of samples-A has $T_0=91$ K (onset $T_c=95.5$ K) for $x=0$ and exhibits the superconductor-insulator (S-I) transition at the Pr concentration $x_c=0.32$. Although not shown here, the XRD pattern exhibited the structure inherent to the Y-123 type cuprates, indicating no vestige of impurity phases like BaCuO$_2$ and PrBaO$_3$. From SEM images the grain size was observed to range from 10 to 20 μm. For the sake of comparison, another series of samples (samples-B) of NdPr-123 was also prepared in a different way from the same powders, through the cyclic process (two times) of pulverizing, mixing and subsequent calcining at 840°C for 24 hour in airs. The resultant specimens were then sintered at 930°C for 24 hour in an oxygen flow and subsequently annealed at 400°C for 24 hours in the same furnace. The XRD pattern was found to be identical to that for samples-A, without significant vestige of such impurity phases as stated above. The series of samples-B has $T_0=70$ K ($T_c=80$ K) for $x=0$ and shows the S-I transition at $x_c=0.14$. Thus, the samples-B are of low quality in comparison with the samples-A.

3. Results and discussion
Figure 1 shows the Pr-concentration ($x$) dependence of the zero-resistance temperature $T_0$, at which the resistivity reaches zero. For the high quality samples (-A), the $T_0$ descends from 91 K at $x=0$ to 0 K at $x_c=0.32$, where the S-I transition takes place. This value of $x_c$ agrees closely with observations reported in the literature [9]. For the low qualified samples (-B), the initial $T_0$ at $x=0$ is as low as 70 K, and the S-I transition ($T_0=0$) occurs at $x_c=0.14$, which is much lower than that for samples-A. Since the

![Figure 1. Pr-concentration dependence of the zero-resistance temperature $T_0$ for NdPr-123 samples-A and -B. The data extrapolation is given by broken lines to obtain $x_c$. However, the plot for samples-B is determined only by two experimental points, giving a rough estimation of $x_c$ in comparison with samples-A.](image-url)
Figure 2. Behaviour of resistivity ($\rho$) on varying the current ($I$): (a) non-ohmic resistance for samples-A with $x=0.3$; (b) ohmic resistance for samples-A with $x=0.45$; (c) non-ohmic (negative) resistance for samples-A with $x=0.8$; (d) negative resistance for samples-B with $x=0.3$. The intensity $I=1$ mA corresponds to the current density $J$: (a) $J=10^3$, (b) $J=100$, (c) $J=159$, (d) $J=285$ (A/m$^2$).

Structures of both samples-A and -B are identical to each other as confirmed from the XRD patterns, the superconducting degradation of the samples-B is probably caused by the oxygen depletion due to the heat treatment in sample preparation, which was different to that for the samples-A [10].

Figures 2 displays resistivity ($\rho$) vs. temperature ($T$) curves for samples-A and -B. As depicted in figure 2(a), $\rho$ at $x$ near the percolation threshold ($x_c=0.32$ for samples-A) exhibits a strong dependence on the current $I$, as an increasing function of $I$. In general, its current-dependence becomes considerable as $x$ approaches the percolation threshold, as typically observed for YPr-123 at $x=0.55$ [1]. It is to be noted that such current-dependent resistivity is generally observed also when the superconducting paths contains weak-links between adjacent grains [11]. The above current dependence strongly reflects the breaking process of the superconducting network that consists of Nd-123 unit cells only, where the structural neck (hot spot) that is crucial for completing the network is broken in superconductivity when the current passing there exceeds its critical current [7]. The stepwise breaking at such hot spots with increasing $I$ produces the non-ohmic resistance as an increasing function of $I$. This current dependence vanishes when $x$ increases further, as seen for $x=0.45$ in figure 2(b). The resistance becomes ohmic, independent of $I$. This current independence persists over a relatively wide range of $x$ above $x_c$. When $x$ departs far away from $x_c$, however, the resistance...
Figure 3. Model of the negative resistance: finite conducting clusters are embedded in the semi-conducting matrix of Pr-123 and the supplied current $I$ passes mainly along a route of the lowest resistance (bold arrows), elevating a local temperature $T$ at each of the highly resistive separations between clusters. The dashed arrows indicate faint branch-currents.

comes to depend again on $I$, but changes into a decreasing function of $I$, as shown in figure 2(c); we refer this behaviour as to negative resistance. Figure 2(d) shows that for samples-B the negative resistance appears much faster than for samples-A, as seen at concentrations as low as $x=0.3$. The easy occurrence of the negative resistance just reflects the extremely low percolation threshold $x_c \approx 0.14$.

As exhibited in figure 3, at $x$ sufficiently higher than $x_c$, the superconducting network is strongly broken into small clusters consisting of Nd-123 unit cells that are embedded in the semiconducting matrix of Pr-123 unit cells. The current path preferably threads those highly conducting clusters in order to take a route of the lowest resistance across the sample. The observed resistance comes mainly from the highly resistive separation between neighbouring clusters. When the spatial distribution of these clusters is dilute as expected for high $x$, the freedom of forming the current path is spatially restricted, so that the current is especially condensed along the selected route, elevating a local Joule heat at the separation, that is, giving a local rise in temperature and thereby reducing the resistance there [7]. The ambient temperature $T_0$ that we can observe is lower than the local temperature $T$. As the current increases, the local temperature rises at the separations so that the overall resistance decreases. For the moderately high range of $x$, the conducting clusters distribute densely, so that the current can take many routes. Then, each of the separations only bears a low current density, yielding no significant rise in the local temperature. Thus, the overall resistance is substantially kept ohmic.

In conclusion, the non-ohmic resistance having the dual aspects demonstrates a distinct correspondence between the electrical properties and the structural inhomogeneity in the Pr-doped Nd-123. Under appropriate conditions, the negative resistance can produce an anomalous voltage that decreases with increasing $I$ [8], giving a potential for functional applications. Finally, the authors would like to thank Mr Hayasaka for his experimental support.

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