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Electrical and thermal transport properties of $Dy_{0.95}Pr_{0.05}Ba_2(Cu_{1-x}M_x)_3O_{7-\delta}$ with (M=Fe, Co, Ni and Zn) bulk superconductors

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ABSTRACT

In the present communication, measurements of transition temperature, thermal conductivity, thermoelectric power and specific heat have been carried out on pristine and co-doped samples of $Dy_{0.95}Pr_{0.05}Ba_2(Cu_{0.98}M_{0.02})_3O_{7-\delta}$ system [M=Fe, Co, Ni and Zn]. The electrical resistivity results show that all the samples exhibit metallic behavior. The thermal conductivity results illustrate that for pristine sample of $DyBa_2Cu_3O_{7-\delta}$, the expected pronounced hump in thermal conductivity is seen below the transition temperature. The hump is suppressed with 5% Pr-doping and this is further suppressed with co-doping. The thermoelectric power measurements show that the pristine sample exhibits hole-like behavior and this continues for doped as well co-doped samples. Pristine sample shows the expected jump in specific heat and with Pr-doping a slope change is observed near the transition temperature.

1. Introduction

The discovery of superconductivity in $Y_1Ba_2Cu_3O_7$ (Y-123) opened a new era to discover new superconductors with higher superconducting transition temperatures T_C [1]. Over last two and a half decades, exhaustive study has been done on the substitution at the Cu site which has helped in understanding the mechanism of superconductivity [2–5]. Studies in the past, over a last decade, have suggested that the other rare earth based systems such as $RE_1Ba_2Cu_3O_7$ (RE=Nd, Sm, Eu, Gd, Dy etc.) (RE-123), have better applicability compared to Y-123 system [6–10]. They show a higher transition temperature, better surface morphology and also better performance under external magnetic field. Reports have shown that rare earths such as Nd, Sm, and Eu which have their ionic radii similar to that of Ba^{2+}, lead to more point defects due to the higher possibility of RE/Ba substitution, that act as strong vortex pinning centers [9]. On the other hand, RE-based materials with smaller radii such as (Er, Yb), have been more advantageous as they require lower processing temperatures and possess wider processing windows, unlike the larger RE-123 materials [6–10].

It is well-known that RE-123 with RE=Ce, Tb, Pm and Pr, are non-superconducting, however, Pr-123 is the only compound iso-structural to RE-123. It remains semiconducting down to very low temperatures. The destruction of superconductivity in Pr-123 has been attributed to many debatable mechanisms such as, magnetic pair breaking involving a strong exchange interaction between the Pr magnetic moments and the conducting electron, hole filling in conducting CuO_2 planes and the localization of states owing to disorder in the CuO_2 planes. While the valence of Pr in $RE_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ has been a subject of immense controversy, it is seen that Pr doping quenches the superconductivity bringing about a metal–insulator transition. An extensive study on the effects of Pr substitution in RE-123 system, in addition to providing an insight into the mechanism of superconductivity [11–16], has shown an increase in J_C [17–19].

A good amount of work has been done to understand the combined effect of the partial substitution of Pr ion at the RE site along with the substitution of Fe, Co, Zn and Ni at the Cu site [20–32]. These studies were basically intended to understand the interaction between Pr *4f* electrons and the CuO_2 valence band by varying the electronic state of the CuO_2 planes [20–29]. In most cases these studies are limited to routine characterizations and the other measurements including IR spectroscopy, Raman scattering and Mössbauer effect [20,21,23–29]. There are reports on electrical and thermal studies on co-substitution of Y, Gd and Eu [30–32], no work seems to have been done on the co-doped Dy-123 system. In particular, Dy-123 system is of great importance as the oxygen pressure and the substrate temperature do not play significant

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roles for good quality Dy-123 films. In addition to this, both T_C and critical current density J_C, are not very sensitive to oxygen pressure and the substrate temperature. This property of Dy-123 makes it a superior material for fabrication of long-length conductors where the processing parameters fluctuation is inevitable [6]. Recent reports suggest that Dy-based low-thermal-conductive composite superconducting materials are suitable for applications such as power leads [33]. Melt-processed Dy-123 superconductors have high J_C, at 77 K and are desirable for high field applications such as superconducting permanent magnets and current leads [34,35].

It was thus desirable to investigate electrical and thermal properties of Dy-based co-doped system. In this communication we have carried out electrical resistivity, thermal conductivity, thermo-power and specific heat on co-doped system $Dy_{0.95}Pr_{0.05}Ba_2(Cu_{1-x}M_x)_3O_7$ (M=Fe, Co, Ni and Zn). We have chosen low concentration of Pr (5%) and low doping levels (2%) of Fe, Co, Ni and Zn. A low content of the transition metals is chosen so that the disorder may not be the dominating effect of impurity, instead the electronic effect will be dominant [36].

2. Experimental details

The samples of $Dy_{0.95}Pr_{0.05}Ba_2(Cu_{0.98}\ M_{0.02})_3O_{7-\delta}$ system [M=Fe,Co,Ni and Zn] were prepared using the solid state reaction technique. The stoichiometric amount of the required constituent materials Dy_2O_3, $BaCO_3$, CuO, Pr_6O_{11}, $Fe\ _2O_3$, Co_3O_4, NiO, and ZnO were ground in an agate mortar and then calcined in air at 930 °C for 12 h. The process of calcination and grinding was repeated three times to ensure the homogeneity of the sample stoichiometry. The calcined powder was pressed into rectangular bars of dimensions approximately 14 mm × 7 mm × 2 mm using a hydraulic press. The samples were then annealed in flowing oxygen at 950 °C for 36 h, 630 °C for 24 h and 450 °C for 48 h and finally slow cooled to room temperature. The oxygen content of all the samples was estimated using the standard iodometric titration. The room temperature X-ray diffraction data was obtained using the Rigaku Miniflex 600 X-ray diffractometer, with Cu-$K_{\alpha1}$ (λ=1.5405 A) radiation as a source, in the 2θ range 5–110° in step of 0.02° and the structural and phase analysis was done. The XRD patterns show that the samples are single phased.

The electrical resistivity $\rho(T)$ was measured in the temperature (T) range 10–300 K using the standard four-probe method in a closed cycle refrigerator (CCR). Lakeshore temperature controller-Model 325 was used to measure and control the temperature of the sample. Keithley current source (Model 6221) was used to maintain constant current through the current leads, and the voltage across the voltage leads was measured by Keithley nano-voltmeter (model 2182A). The thermal conductivity $\kappa(T)$ and the thermo-electric power S (T) measurements were simultaneously done in the range 10–300 K in a CCR, using a direct-pulse technique. Both these measurements were made in the warming cycle at a controlled rate of ≤ 20 K/h, with reproducibility over 2%. Specific heat measurements were carried out using a high resolution ac calorimeter, using chopped light as a heat source. The details of the measurement technique are given elsewhere [32].

3. Results and discussion

3.1. Transition temperature

Fig. 1 shows temperature dependent resistivity of pristine and co-doped superconducting samples of $Dy_{0.95}Pr_{0.05}Ba_2(Cu_{0.98}\ M_{0.02})_3O_{7-\delta}$ system [M=Fe, Co, Ni and Zn]. It is seen that all the samples under present studies exhibit metallic trend. Normal state

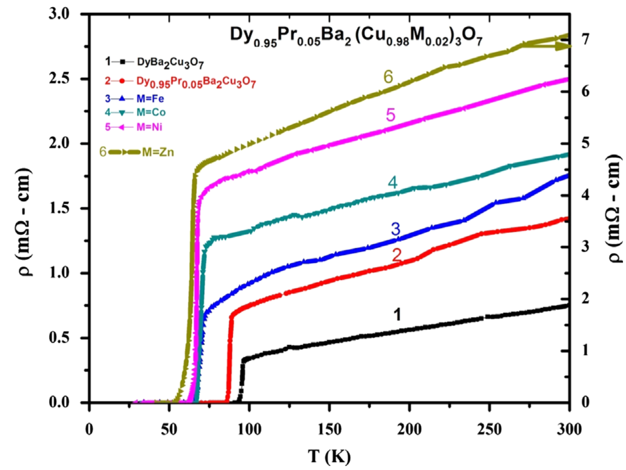

Fig. 1. (Color online) Resistivity versus temperature plots of $DyBa_2Cu_3O_{7-\delta}$ and $Dy_{0.95}Pr_{0.05}Ba_2Cu_{2.94}M_{0.06}O_{7-\delta}$ samples for M=Fe, Co, Ni and Zn.

resistivity is observed to increase with 5%Pr doping at Y-site. With co-doping at Cu-site, the normal state resistivity is observed to increase further. The increase in normal state resistivity is attributed due to additional scattering centers which are produced through doping. To the best of our knowledge, there appear to be no studies done on co-doped samples of Dy–Pr. However; similar trends are seen in co-doped RE-based superconducting compounds [30–32]. It is further found that the superconducting transition temperature, T_C decreases with co-doping, on contrary, the width in transition, ΔT_C, is found to increase with co-doping. For example, the pristine sample of $DyBa_2Cu_3O_{7-\delta}$ has a T_C of about 95 K with ΔT_C=3.5 K, whereas, for $Dy_{0.95}Pr_{0.05}Ba_2Cu_3O_{7-\delta}$ compound, it is found to decrease to 87 K with associated transition width of about 4 K. These results are in good agreement with those reported in literature [33]. With co-doping with transition elements, a further decrease is seen in T_C. Among all the co-dopants, Zn-doped sample has the lowest T_C and highest ΔT_C values. The increase in these values may be attributed to an increased degree of in-homogeneity and possible disorders in these specimens. The present results are in fairly good agreement with those reported in literature [37]. Table 1 gives T_C and ΔT_C of various samples investigated in present studies.

3.2. Thermal conductivity

The temperature dependent thermal conductivity, $\kappa(T)$, of $Dy_{0.95}Pr_{0.05}Ba_2(Cu_{0.98}\ M_{0.02})_3O_{7-\delta}$ system [M=Fe, Co, Ni and Zn] in the temperature range of 10–300 K is shown in Fig. 2. It is expected that one always observes a hump in thermal conductivity below the superconducting transition temperature T_C. This hump signifies the existence of electron–phonon interactions which is a consequence of the lattice theory of superconductivity. In addition to this factor, the increase in $\kappa(T)$ is an indication of possible enhancement of the quasi-particle contribution to the heat conduction thereby there is an increase in quasi-mean free path in these high temperature superconductors [31]. In the present studies, we have observed the expected hump for the pristine sample of $DyBa_2Cu_3O_{7-\delta}$ below its transition temperature. For the Pr-doped sample ($Dy_{0.95}Pr_{0.05}Ba_2Cu_3O_{7-\delta}$), the expected hump below its transition temperature is seen, however, there is a decrease in the height of hump in κ (T). This decrease in hump is basically due to an enhancement of scattering processes at low temperatures via Pr-doping. Similar results are reported in literature [31].

With co-doping with transition metals at Cu site, the hump structure shows a different trend. For co-dopants such as Fe and