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Electrical and thermal transport properties of $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$

Ashok Rao[1,7], Anirban Das[2], Tirthankar Chakraborty[3], Bhasker Gahtori[4], S K Agarwal[4], Chandan Kumar Sarkar[3], K M Sivakumar[5], K K Wu[6] and Y K Kuo[6,7]

[1] Department of Physics, Manipal Institute of Technology, Manipal 576104, India
[2] Department of Physics, Sikkim Manipal Institute of Technology, Majitar, Rangpo, Sikkim 737132, India
[3] Department of Electronics and Telecommunication Engineering, Jadavpur University, Kolkata 700032, India
[4] Superconductivity and Cryogenics Division, National Physical Laboratory, Dr K S Krishnan Road, New Delhi 110012, India
[5] Department of Physics, RIRT, Radharaman Group of Institutes, Badbadha Road, Ratibad, Bhopal 462046, MP, India
[6] Department of Physics, National Dong-Hwa University, Hualien 974, Taiwan

E-mail: ashokanu_rao@rediffmail.com and ykkuo@mail.ndhu.edu.tw

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Abstract

In this communication, we present the measurements of electrical resistivity (ρ), thermal conductivity (κ), thermopower (S), and specific heat (C_P) on Mn doped compounds $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$ for $0 \leqslant x \leqslant 0.02$. The transition temperature T_C and the upper critical field $H_{C2}(0)$ in this series of compounds are found to decrease quasi-linearly as the Mn concentration increases. All the currently investigated samples show a hump in thermal conductivity below their transition temperatures. Thermopower measurements exhibit an electron-like behaviour for pure samples; however, a hole-like behaviour is seen in all the doped samples. A small jump in specific heat is observed in the pure samples. It is seen that replacing 0.5% Cu by Mn has little effect on the transition temperature, but there is a nearly twofold suppression in the C_P jump for this sample. These results demonstrate that the impurities are being incorporated as a whole and not as a cluster.

1. Introduction

There are a number of reports on partial and full substitution effects in high T_C superconductors and such substitutions in general, found to have an influence on the superconducting as well as normal state properties [1–8]. The primary aim of such studies is to rationalize the underlying mechanism behind the principle of superconductivity by altering the composition of these compounds. Understanding the changes in their corresponding physical and mechanical properties is the key to a possible enhancement of the efficiency of these ceramic superconductors for technological and industrial applications. As far as the thermal and electric transport properties of pristine and doped ceramic superconductors are concerned, $YBa_2(Cu_{1-x}M_x)_3O_{7-\delta}$ (where M = transition metal) is the most widely investigated system.

In the last decade or so, there have been several efforts to study other rare-earth-based superconductors $REBa_2Cu_3O_{7-\delta}$ (RE = Eu, Sm, Nd etc) [9–16]. Compared to the Y-123 system, it has been reported that other RE-based systems have a better applicability. For instance, Eu-based superconducting films show a better surface morphology, and high transition temperature [9, 16]. These properties are important for thin film devices and coated conductors. Besides, Eu^{3+} has an ionic size almost the same as that of Ba^{2+} and this may lead to the introduction of point defects into the RE-123 system. This is advantageous for the application in an external magnetic field, as large densities of point defects can act as strong pinning centres in the RE-123 systems [15]. Even though the Eu-based compounds represent a superior potential in applications,

[7] Authors to whom any correspondence should be addressed.

relatively few studies have been done on electrical and thermal transport properties of pure as well as doped $EuBa_2Cu_3O_{7-\delta}$ superconductors.

In the present work we have carried out low temperature electrical resistivity (with and without applied fields), thermal conductivity, thermopower, and specific heat measurements on pure and Mn doped $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$ samples. Mn doping is of significant importance as far as the theoretical point of view is concerned, because Mn in the Y-123 system shows a distinctly different behaviour as compared to other transition metals. In fact, in the Y-123 system, Mn behaves like Fe as far as the occupancy of the copper chain site is concerned [1, 2]; on the other hand, it behaves like Zn or Ni as far as the retention of orthorhombic structure is concerned [3–5]. Recently we have reported electrical and thermal transport properties of Mn doped Y-123 and Gd-123 systems [17, 18]. The motivation behind the present investigation is to study the effect of Mn doping in the Eu-123 system and compare it with the thermal and electrical properties of Mn doped Y-123 and Gd-123 systems.

2. Experimental details

Samples of the $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$ system in the nominal doping range $0\% \leqslant x \leqslant 2\%$ were prepared by the standard solid-state reaction technique. High purity (>99.99%)Eu_2O_3, $BaCO_3$, CuO, and MnO were thoroughly mixed and calcined at 930 °C in air for 12 h. This calcination process was repeated thrice to improve the homogeneity. The powder was then pressed into pellets followed by annealing at 930 °C under oxygen atmosphere for 72 h, and was slowly cooled to 450 °C and kept at this temperature for about 6 h. The x-ray diffraction confirmed that all the samples prepared in the present investigation are of single phase (orthorhombic) with no traceable impurities. The transition temperatures of the samples were determined by electrical resistivity using the standard four-probe technique. Magneto-resistance studies were carried out on all the samples in the temperature range of 1.5–300 K by varying external magnetic field in the range of 0–8 T. Both the thermal conductivity (κ) and thermopower (S) measurements were carried out in the range 10–300 K in a closed-cycle refrigerator, using a direct-pulse technique. Both these measurements were performed during the warming cycle at a slow rate of $\leqslant$20 K h^{-1}. The reproducibility of $S(T)$ and $\kappa(T)$ measurements is better than 2%. Relative specific heats were performed with a high-resolution ac calorimeter, using chopped light as a heat source. The details of the experimental techniques can be found elsewhere [17].

3. Results and discussion

3.1. Electrical resistivity

The temperature dependence of the resistivity of the investigated samples is shown in figure 1. One can observe that all the samples show metallic behaviour. We also observed that the resistivity increases with Mn content. To the best of our knowledge, there are no reports on Mn doping in the $EuBa_2Cu_3O_{7-\delta}$ (Eu-123) system. We have thus compared the present results with that reported for Mn doped samples of $YBa_2Cu_3O_{7-\delta}$ (Y-123) and $GdBa_2Cu_3O_{7-\delta}$ (Gd-123) systems [17]. It is observed that for 1% Mn doping the resistivity of the Eu-123 sample at 200 K is about 0.26 mΩ cm, whereas for the same doping levels of Mn in Y-123 and Gd-123 systems it is about 5 mΩ cm and 11 mΩ cm, respectively [17].

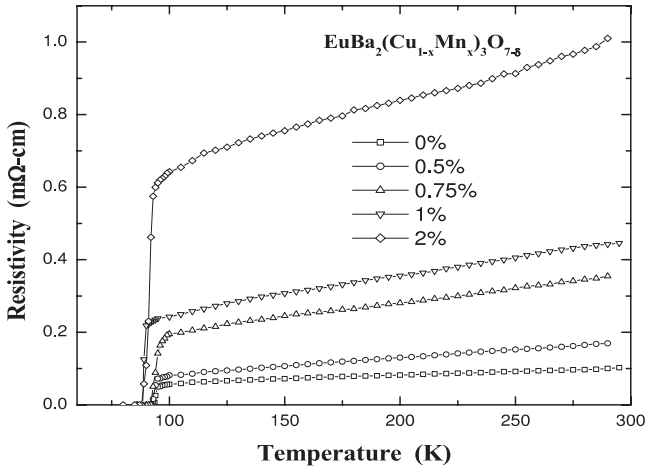

Figure 1. Resistivity versus temperature plots of the samples $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$ for various values of x.

Figure 2(a) depicts the transition temperatures of Mn doped samples of Eu-123 as a function of Mn concentration. It is found that the transition temperature decreases from 93 to 88 K as the dopant level is increased from 0 to 2%, with a depression rate $D = dT_C/dx \approx -300$. The decrease in transition temperature is possibly due to the combination of two effects, namely transfer of electrons from Mn ions to the CuO_2 layer and transfer of electronic spectral weight from higher energy states to low energy states associated with a shift in chemical potential. It is worthwhile mentioning that in the Y-123 system only a marginal change in transition temperature, whereas in Gd-123 system a noticeable increase in T_C, have been observed [17]. Magneto-resistance measurements were performed in the temperature range of 1.5–300 K and magnetic field was varied from 0 to 8 T. The upper critical field $H_{C2}(0)$ was estimated by standard procedures and the behaviour of $H_{C2}(0)$ as a function of Mn content is depicted in figure 2(b). It is observed that $H_{C2}(0)$ decreases almost linearly with Mn content. This is due to the fact that impurities in general break Cooper pairs and hence as a result there is a decrease in $H_{C2}(0)$.

3.2. Thermal conductivity

The thermal conductivity of the $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$ system in the temperature range 10–300 K is displayed in figure 3. We have observed a hump in all the samples below their respective transition temperatures. The hump signifies the occurrence of electron–phonon interaction as per the lattice theory of thermal conductivity [18]. However, Aksan *et al* [19, 20] have shown that the increase in κ just below T_C is an indication for the enhancement of the quasiparticle contribution to the heat conductivity and so an

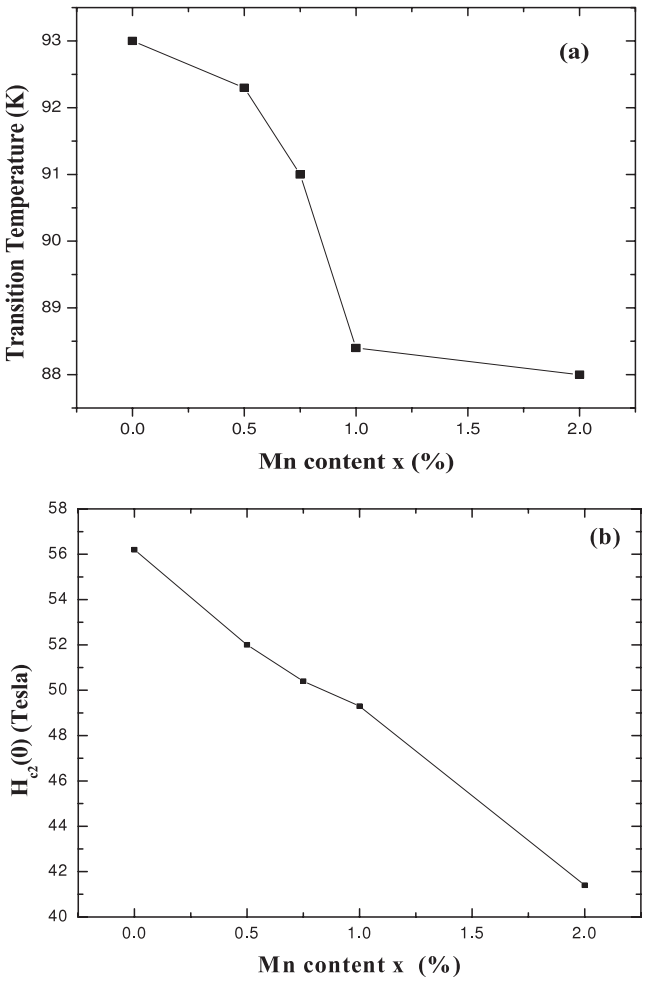

Figure 2. (a) Variation of transition temperature of $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$ compounds with Mn concentration (x%). (b) H_{C2} versus Mn content.

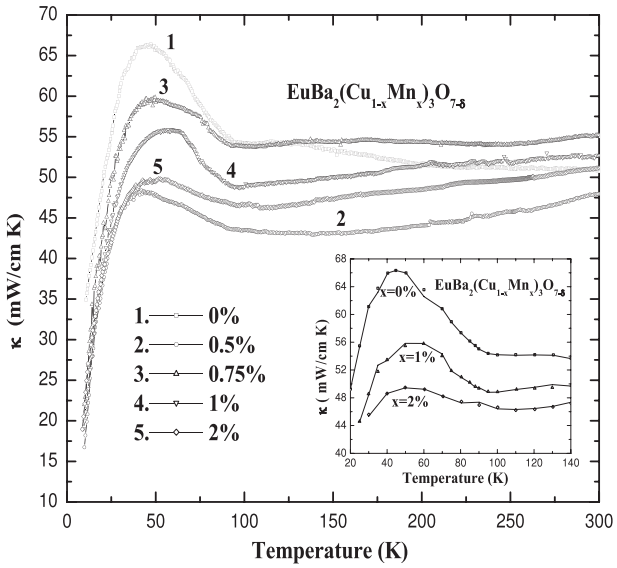

Figure 3. Variation of thermo-conductivity of $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$ for $x \leqslant 0.02$ with the temperature up to 300 K. The inset shows the comparison of experimental results (points) and the fitting (line).

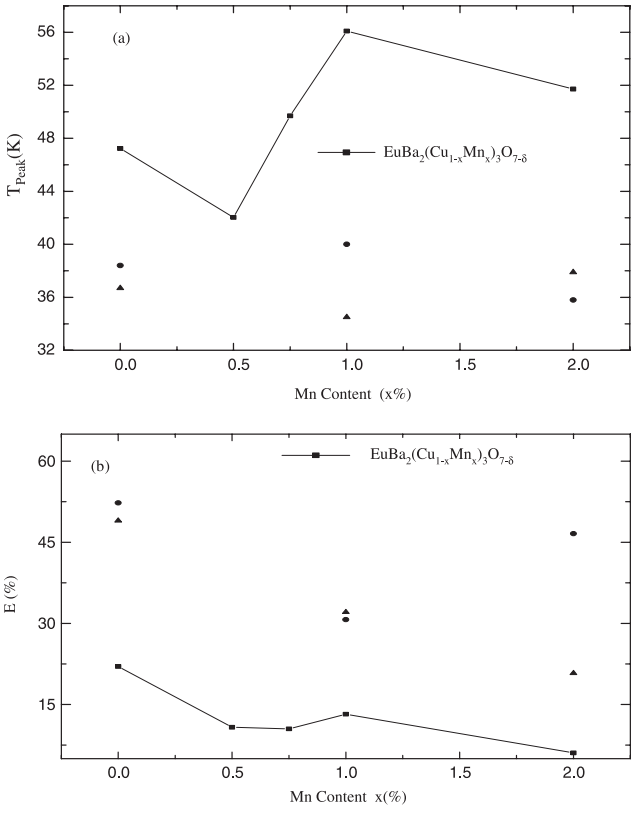

Figure 4. (a) Variation of T_{peak} with Mn content. (b) Variation of E (%) for the $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$ system for various Mn contents. For comparison, these values for $YBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$ and $GdBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$ systems are shown by the solid circle and solid triangle, respectively.

increase of the quasiparticle mean free path in the high-T_C materials. Therefore, an alternative possibility for the thermal conductivity hump is the electronic contribution, as the quasiparticle scattering is the main source in the superconducting state of high-T_C materials. In addition, it is found that the height of the observed hump in $\kappa(T)$ decreases gradually with increasing substitution level, except for the $x = 0.5\%$ sample. The deviation from the systematic trend for $x = 0.5\%$ is certainly not intrinsic to the crystal structure and could be originated from the point-like defects or tunnelling states present in the sample. The observed suppression of the hump suggests an enhancement of low temperature scattering processes with increasing Mn content.

We now analyse the hump structure by determining the peak value of thermal conductivity κ_{peak} and the peak temperature T_{peak} at which the maximum values of κ occur and the percentage peak enhancement, $E(\%)$. The details can be seen in our earlier communication [18]. Figures 4(a) and (b) show the behaviour of T_{peak} and $E(\%)$ as a function of Mn concentration, respectively. It is observed that T_{peak}, in general has higher values than that seen in Y-123 and Gd-123 systems, while the values of $E(\%)$ for the Eu-123 system appear to be lower [18].

In the lattice model, it is well known that the thermal conductivity of a solid is contributed mainly by five processes, which are (1) boundary scattering of the phonons, (2) scattering

Table 1. Values of parameters B, α, β, η, θ_D and U according to the fitting equation with the thermal conductivity of $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$, for various x. Here B, α, β, η and U are expressed in cm K mW^{-1} and θ_D is expressed in K.

Sample	B	α	β	η	θ_D	U
$EuBa_2Cu_3O_{7-\delta}$	0.21	100.2	5.02	6.72	379	9.8
$EuBa_2(Cu_{0.995}Mn_{0.005})_3O_{7-\delta}$	0.24	109.2	6.4	5.9	378	6.2
$EuBa_2(Cu_{0.9925}Mn_{0.0075})_3O_{7-\delta}$	0.53	94.2	8.5	13.3	382	5.6
$EuBa_2(Cu_{0.99}Mn_{0.01})_3O_{7-\delta}$	0.51	74.2	9.4	13.8	384	7.8
$EuBa_2(Cu_{0.98}Mn_{0.02})_3O_{7-\delta}$	0.64	16.2	11.4	19.2	392	6.4

of phonons from point defects, (3) scattering of phonons from the strain field of sheet-like faults, (4) electron–phonon scattering and (5) phonon–phonon scattering. We have used the standard procedure [21, 22] to obtain the expression for the thermal conductivity within the lattice model. For all the samples, we have fitted the experimentally observed thermal conductivity. There is a good agreement between experimentally observed thermal conductivity and that obtained by fitting, which is shown in the inset of figure 3. Table 1 shows the values of the parameters B, α, β, η, θ_D and U. These parameters have been defined in our earlier communication [18].

3.3. Thermopower

Figure 5(a) depicts the behaviour of thermopower as a function of temperature for the pristine as well as doped samples. The pristine sample $EuBa_2Cu_3O_{7-\delta}$ shows an electron-like behaviour, whereas with Mn doping it shifts to a hole-like behaviour. Present results are qualitatively similar to that observed earlier in $GdBa_2Cu_3O_{7-\delta}$. To analyse the observed temperature variation of thermopower, we have applied the theoretical model proposed by Gasumyants *et al* [23] using the narrow band picture of cuprates. In this model one describes S in terms of three parameters, namely n, W_D and W_σ. Here, n, W_D and W_σ denote band filling, total effective bandwidth of the electronic states and the effective width of an energy interval for electronic conduction, respectively. In the present investigation, we have limited the temperature range to 300 K. Hence, we have used only two parameters. The two-parameter version of the expression for S as given by Gasumyants *et al* [23] may be written as

$$S = \frac{k_B}{e \sinh W_\sigma^*}\left[W_\sigma^* \sinh\mu^* + \mu^*(\cosh\mu^* + \exp W_\sigma^*) + \left(\cosh\mu^* + \cosh W_\sigma^* \ln\frac{1+e^{W_\sigma^*-\mu^*}}{1+e^{W_\sigma^*+\mu^*}}\right)\right] \quad (1)$$

where

$$\mu^* = \frac{\mu}{k_B T} = \ln\left\{\frac{\sinh(nW_D^*)}{\sinh[(1-n)W_D^*]}\right\}, \qquad W_\sigma^* = \frac{W_\sigma}{2k_B T} \quad \text{and} \quad W_D^* = \frac{W_D}{2k_B T}. \quad (2)$$

In equation (1), e is the electronic charge of the carrier. In equation (2), μ is the chemical potential and one can notice that μ is temperature dependent for given values of n and W_D. In order to avoid this problem, one can notice that, in the limit $2k_BT \ll W_D$, μ^* approaches $(2n-1)W_D/2k_BT$. Thus, μ becomes independent of T. Hence, for low temperatures we have estimated values of μ for different samples in the framework of the narrow band picture. Notice that S is an odd function of μ. Using equation (1) we have fitted the experimental values of thermopower and thus obtained the values of μ and W_σ for different samples. Figure 5(b) shows the comparison of experimental data (points) with theoretical results obtained from the above mentioned fitting

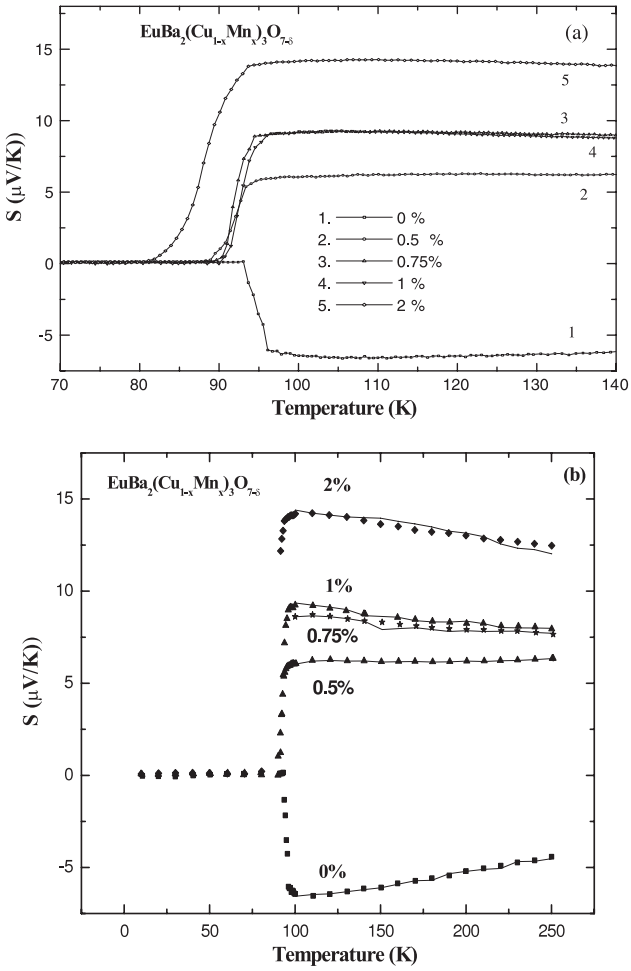

Figure 5. (a) Variation of thermopower (S) of $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$ for various x values with the temperature up to 300 K. (b) Comparison of experimental results (points) and the fitting (line).

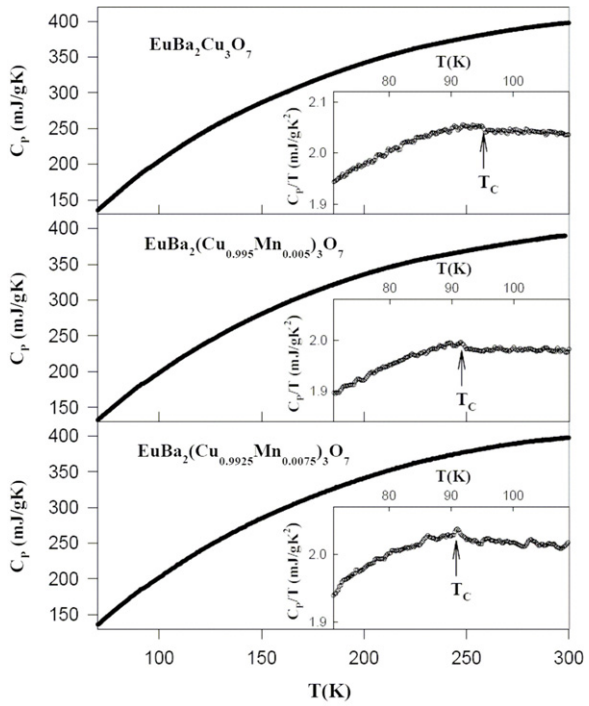

Figure 6. The temperature dependence of the specific heat of pure and Mn doped $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$ samples with $x = 0.5\%$ and 0.75%. The insets show temperature dependent variations of C_P/T in the vicinity of the transition temperature.

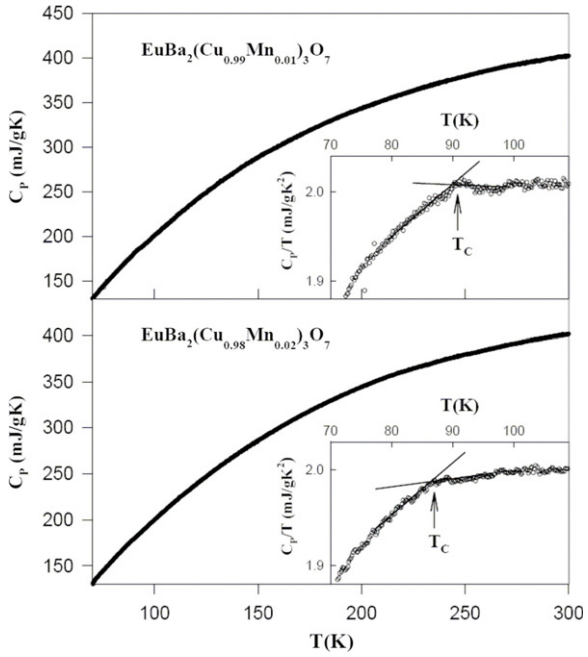

Figure 7. The temperature dependence of the specific heat of Mn doped $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$ samples with $x = 1\%$ and 2%. The insets show temperature dependent variations of C_P/T in the vicinity of the transition temperature.

(solid line). One can observe that there is a good match between experimental data and theoretical results.

3.4. Specific heat

Figure 6 illustrates the temperature dependent specific heat of $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$ ($0\% \leqslant x \leqslant 0.75\%$). We present the specific heat behaviour as a function of temperature in figure 7 for two specimens with Mn doping of 1 and 2%. The insets show the specific heat data in the vicinity of the transition temperature of the respective samples. It may be mentioned that the present results of specific heat do not show a sharp discontinuity around the transition temperature. Instead, C_P increases gradually as the specimen undergoes the transition from normal to superconducting state. In a conventional superconductor, a sharp discontinuity at T_C is generally observed in specific heat. This difference is essentially due to the broad transition widths (2–3 K) in high T_C superconductors, in contrast to the sharp transition width of a few tenths of a degree in conventional superconductors.

The jump in specific heat at T_C was determined by extrapolating the two branches of the C_P/T versus T curves around T_C. We have plotted C_P/T instead of C_P in the vicinity of the transition temperature because the jumps are best seen in such plots. For pure $EuBa_2Cu_3O_{7-\delta}$, the observed jump ΔC_P is about 2.1 J mol^{-1} K^{-1}, which is in good agreement with that observed by Plackowski *et al* [24]. With a small amount of Mn doping (0.5%), a nearly twofold decrease in the jump with $\Delta C_P \approx 1.2$ J mol^{-1} K^{-1} is observed. With further increase in Mn concentration to 1%, we could not detect any such obvious jump within the experimental errors. Nevertheless, we could notice a change in slope instead of a jump at the transition. The change in behaviour from 'clear peak' to 'change in slope' is perhaps correlated with the drastic change in resistivity. These values of ΔC_P suggest that either Mn induces gapless superconductivity for $x \geqslant 0.01$ or there are strong thermal fluctuations around T_C.

For describing the specific heat in the normal state, we use the following expression:

$$C_p = \gamma T + A\left(\frac{T}{\theta_D}\right)^3 \int_0^{\theta_D/T} \frac{u^4 e^u}{(e^u - 1)^2}\, du. \qquad (3)$$

This equation describes the electronic (first term) and phononic contributions (second term) to the specific heat. Here γ and A are independent of temperature. In particular, γ is proportional to the electronic density of states at the Fermi level (N_F). θ_D is the Debye temperature. We have fitted the experimental data of C_P with equation (3). Table 2 gives the parameters γ, A and θ_D for different Mn contents. It is noted that the Debye temperatures θ_D determined from equation (3) for the Eu-123 system are close to that obtained from thermal conductivity data (see tables 1 and 2), suggesting the validity of our present analyses.

We now consider the role of Mn in the specific heat in the superconducting state of the $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$ system. From table 2 we see that Mn does not have a strong effect on the Debye temperature θ_D. On the basis of the γ values we have found that the ratio $\Delta C_P/\gamma T_C$ is 0.0748 and 0.0474 for the $x = 0.0$ and 0.005 samples, respectively. These values are one to two orders of magnitude lower than the ratio of 1.43 in the Bardeen–Cooper–Schrieffer (BCS) theory. Such low values of $\Delta C_P/\gamma T_C$ for the $x = 0$ and $x > 0$ samples indicate strong effects of both the Eu^{3+} and Mn^{k+} ions in these systems.

Table 2. Values of parameters γ, A and θ_D according to the fitting equation with the specific heat data of $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$, for various x.

Sample	A (mJ g^{-1} K^{-1})	γ (mJ g^{-1} K^{-2})	θ_D (K)
$EuBa_2Cu_3O_{7-\delta}$	647.4	0.41	377.5
$EuBa_2(Cu_{0.995}Mn_{0.005})_3O_{7-\delta}$	623.3	0.38	370.5
$EuBa_2(Cu_{0.9925}Mn_{0.0075})_3O_{7-\delta}$	618.0	0.35	366.5
$EuBa_2(Cu_{0.99}Mn_{0.01})_3O_{7-\delta}$	610.3	0.37	371.5
$EuBa_2(Cu_{0.98}Mn_{0.02})_3O_{7-\delta}$	612.5	0.39	381.5

4. Conclusions

We have presented electrical resistivity, thermal conductivity, thermopower and specific heat studies of Mn doped $EuBa_2(Cu_{1-x}Mn_x)_3O_{7-\delta}$ samples prepared under identical conditions. From the thermal conductivity data, we have observed that all the samples show a hump below their respective transition temperatures. It is worth noting that the experimental results match quite well to the fittings through theoretical considerations. Thermopower measurements show that the pristine $EuBa_2Cu_3O_{7-\delta}$ sample exhibits an electron-like behaviour; in contrast, a hole-like behaviour is seen in the doped samples. Distinct features in the specific heat measurements are noticed in the Eu-123 system as compared to conventional superconductors, suggesting strong effects of both the Eu^{3+} ions and Mn^{k+} ions in the currently studied system.

Acknowledgments

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