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Thermoelectric power of single-phase samples of $Tl_2CaBa_2Cu_2O_y$ and $Ba_2CaSr_2Cu_2O_y$

V VIJAYASHREE, C K SUBRAMANIAM, R SRINIVASAN,
I K GOPALAKRISHNAN*, P V P S S SASTRY*, J V YAKHMI*
and R M IYER*

Department of Physics, Indian Institute of Technology, Madras 600036, India
*Chemical Group, Bhabha Atomic Research Centre, Trombay, Bombay 400085, India

Abstract. Single-phase 2122 samples of thallium and bismuth superconductors were made by the precursor matrix method. The thermopower of these samples was measured in the temperature range $250\,K - T_c$. The thermopower was positive and decreased linearly with increasing temperature above T_c (onset). The exponential enhancement of thermopower seen in the undoped and doped YBCO was not observed in these samples. The linear variation of thermopower can be explained on the basis of either a two-band model or a narrow band model.

Keywords. Thermopower; linear variation; exponential enhancement; two-band model.

1. Introduction

The thermopower of Tl–Ca–Ba–CuO and Bi–Ca–Sr–CuO has been reported in literature (Alcacer *et al* 1988; Jones *et al* 1988; Mandal *et al* 1988; Mitra *et al* 1988; Bhatnagar *et al* 1989; Pekala *et al* 1989). Most of these results are on multiphase samples and the thermopower has been measured in the temperature range 80–300 K. Thermopower is found to be small and positive, around 2 μV/K at room temperature, and increases linearly with decrease in temperature up to T_c(onset). Mitra *et al* (1988) fitted their data with good agreement to a narrow band Hubbard model proposed by Fisher *et al* (1988). We have measured thermopower on single-phase samples of $Tl_2CaBa_2Cu_2O_y$ and $Bi_2CaSr_2Cu_2O_y$ prepared by the precursor matrix method. The results are described below.

2. Experimental details

The details of the synthesis of these compounds have been described earlier (Gopalakrishnan *et al* 1988; Sastry *et al* 1988). The single-phase nature of these compounds has been confirmed by neutron diffraction analysis (Sequeira *et al* 1988). We have studied two Tl compounds and one Bi compound. The resistivity of these samples was measured by a four-probe method. Tl(a) compound which received additional annealing in flowing oxygen for 24 h at 950 °C had a room temperature resistivity, ρ_0 of 11·37 mΩ-cm and T_c(zero) of 104·33 K. Tl(b) had a ρ_0 of 1·609 and a T_c(zero) of 102·74 K. The Bi compound had a ρ_0 of 1·005 mΩ-cm and T_c(zero) at

*For correspondence

79·79 K. The thermopower of all these compounds was measured from 250 K down to T_c(zero) in an experimental set up described elsewhere (Vasudeva Rao *et al* 1984).

3. Results and discussions

In all these materials, the thermopower was positive at 250 K with a value of a few μV/K. The thermopower increased linearly as the temperature was decreased. Extrapolating this linear variation backwards, the value S_0 of the intercept on the thermopower axis at $T = 0$ K was obtained. We have plotted the reduced thermopower, S_T/S_0, of the different materials in figure 1. The linear decreases in thermopower with increasing temperature above the peak can be described by the relation,

$$S = S_0(1 - \alpha T), \tag{1}$$

where the values of S_0 and α are shown in table 1. We have studied well-oxygenated samples of $YBa_2Cu_3O_{7-y}$ and yttrium-substituted with rare earths (Srinivasan *et al* 1987) and copper-substituted with zinc (Vijayashree *et al* 1989). In these samples, the

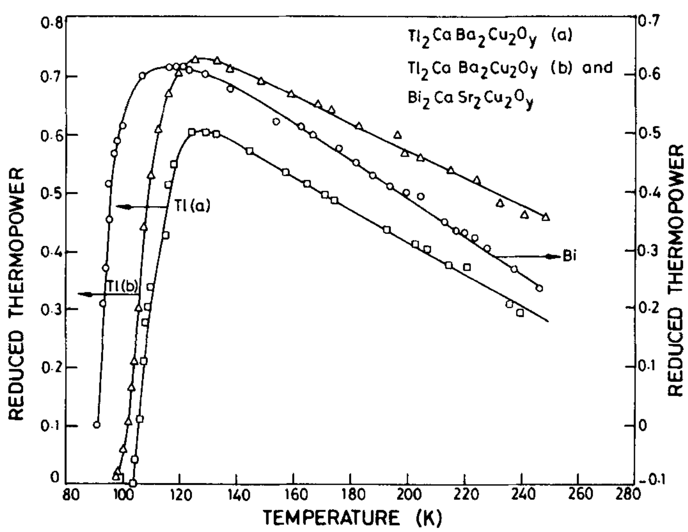

Figure 1. Reduced thermopower as a function of temperature for $Tl_2CaBa_2Cu_2O_y$ (**a** and **b**) and $Bi_2CaBa_2Cu_2O_y$.

Table 1. Parameters S_0 and α in the fit of thermopower to equation (1) in the range 130–250 K.

Sample	$S_0\mu$V/K	$\alpha(10^{-3})$ K^{-1}
$Tl_2CaBa_2Cu_2O_y$(a)	7·94	2·96
$Tl_2CaBa_2Cu_2O_y$(b)	15·76	2·15
$Bi_2CaSr_2Cu_2O_y$	10·33	3·04

temperature variation of thermopower was well represented by the formula,

$$S = aT[1 + b\exp(-T/T_0)]. \quad (2)$$

The samples showed an exponential enhancement of thermopower as T_c was approached. The behaviour of the Tl and Bi compounds is very different from the behaviour of the 123 compounds.

The resistivity of the Tl and Bi compounds shows a linear variation with temperature. The Hall coefficient of multiphase $TlCa_3BaCu_3O_x$ was measured as a function of temperature by Mandal *et al* (1989). The reciprocal of the Hall constant varies linearly with temperature. In this respect, these compounds show the same behaviour as the 123 compounds.

To account for the temperature dependence of resistivity and Hall effect in 123 compounds, Eagles (1989) proposed a two-band model. Here, electrons and holes are both responsible for the transport properties. The inverse mobility of electrons and holes are taken to have a linear temperature dependence. The temperature–independent term and the temperature coefficients of the mobilities are different for the electrons and holes. The carrier concentrations n_h and n_e are taken to be temperature-independent. Then, Eagles (1989) has shown that the resistivity can be written as

$$\rho = \rho_{-1} T^{-1} + \rho_0 + \rho_1 T. \quad (3)$$

In such a situation, the effective thermopower can be written as

$$S = \sigma_h S_h + \sigma_e S_e, \quad (4)$$

where σ_h and σ_e are the contributions to the electrical conductivity by holes and electrons respectively and S_h and S_e are the thermopower due to holes and electrons. Vijayashree *et al* (1990) have shown that taking S_h and S_e to vary linearly with temperature but with opposite signs, the thermopower will depend on temperature as

$$S = S_{-1} T^{-1} + S_0 + S_1 T. \quad (5)$$

The coefficient S_1 can be negative while S_0 can be positive depending on the relative magnitudes of the various coefficients. The temperature where the thermopower crosses the temperature axis depends on the ratio of the carrier concentrations of the electrons and holes and can vary from specimen to specimen.

There is a second explanation proposed by Bar-ad *et al* (1988). In this model, the conduction band is assumed to be narrow. A portion of the band-states is separated from the main part to form an upper sub-band of localized states. These localized states do not contribute to transport but participate in the statistics. At $T = 0$ K, the chemical potential lies above the middle of the conduction band. As the temperature rises, electrons are transferred from the conduction sub-band to the localized upper-band, thereby increasing the number of holes in the conduction band. This can account for the variation of the Hall coefficient with temperature and for *p*-type transport properties. On this basis, Bar-ad *et al* (1988) have calculated the temperature variation of thermopower, resistivity and Hall effect, assuming some values for the width of the localized band and its position relative to the middle of the conduction band. While one can reproduce the linear fall in thermopower with increasing temperature on the basis of this model, the carrier density required appears to be at least one order of magnitude higher than the observed value.

4. Conclusions

2122 single-phase samples of Tl and Bi were prepared by the precursor matrix method. The thermopower measurements carried out in the temperature range 250 K down to the superconducting transition, showed a behaviour different from those of the 123 compounds. The linear decrease in thermopower of these samples as the temperature is increased above the onset of superconducting transition, can be explained satisfactorily using the two-band model proposed by Eagles (1989). The narrow band model by Bar-ad *et al* (1988) can also reproduce the thermopower results.

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