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Scaling behaviour of the in-plane thermopower in $\text{Bi}_2\text{Sr}_2\text{RCu}_2\text{O}_8$ (R = Ca, Y, Pr, Dy and Er)

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Abstract. Single crystals of insulating $\text{Bi}_2\text{Sr}_2\text{RCu}_2\text{O}_8$ (R = Dy, Er, Y and $\text{Ca}_{0.5}\text{Er}_{0.5}$) and superconducting $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Pr}_x\text{Cu}_2\text{O}_8$ ($x = 0, 0.10, 0.25, 0.36$ and 0.42) were prepared, and the thermopower $S(T)$ along the CuO_2 plane was measured from 4.2 to 420 K. All of the thermopowers including those for the insulators are roughly expressed as $A - BT$ above a certain temperature T_{scale} , where A and B are positive constants. $S(T)/S(T_{\text{scale}})$ for all of the samples falls onto a single curve as a function of T/T_{scale} . Since T_{scale} is close to the pseudogap temperature for the superconductors, this scaling offers a rough estimate of the pseudogap temperature of high- T_c superconductors. The present study further suggests that the parent insulators have a pseudogap above room temperature.

1. Introduction

Recently the parent insulators of high- T_c superconductors (HTSC) have received renewed attention as a limit of underdoping [1]. There is increasing evidence that the pseudogap in underdoped HTSC is closely related to the superconducting gap [2], and how the pseudogap grows from undoped CuO_2 planes is a key issue. A significant finding is that a *d-wave*-like gap was observed in the parent insulator $\text{Ca}_2\text{CuO}_2\text{Cl}_2$ in a photoemission experiment [3]. In a previous paper, we measured the resistivity anisotropy of $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Er}_x\text{Cu}_2\text{O}_8$ with special interest taken in the parent insulator [4]. We have found that the carriers in the parent insulator ($x = 1$), just like those in the superconductor ($x = 0$) above T_c , are confined in the CuO_2 plane above the Néel temperature. These results possibly indicate that anomalous properties of HTSC are inherent to the parent insulator.

Thermopower studies can detect a slight change in the density of states in solids, because the thermopower is proportional to the logarithmic derivative of the density of states in the lowest order [5]. Tallon *et al* [6] have found that the thermopowers of $\text{YBa}_2\text{Cu}_{3-x}\text{Zn}_x\text{O}_y$ and $\text{YBa}_2\text{Cu}_{4-x}\text{Zn}_x\text{O}_8$ depend on the Zn content below the pseudogap temperature. According to their results, the thermopower can be a sensitive probe for the pseudogap, and should be measured over a whole range of doping including the parent–insulator region. Mandal *et al* [7] have found a good scaling relation for the thermopower of insulating $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Y}_x\text{Cu}_2\text{O}_8$ ($x = 0.5$ – 0.7). This study is, however, to be re-examined, because (1) the temperature range (77–300 K) was limited, (2) the samples were polycrystals and (3) their scaling method for the insulators was different from that for the superconductors.

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In this paper, we report on the systematic evolution of the in-plane thermopower for various $\text{Bi}_2\text{Sr}_2\text{RCu}_2\text{O}_8$ (Bi-2212) single crystals from parent insulators to slightly overdoped superconductors over a wide temperature range from 4.2 to 420 K. We have found that all the thermopowers are scaled to a *single* universal master curve. This is in good agreement with the scaling behaviour of the susceptibility [8] and the Hall coefficient [9] of HTSC, implying that the density of states is scaled by a certain characteristic temperature.

2. Experimental procedure

We prepared two sets of Bi-2212 single crystals. One was a set of the parent insulators expressed as $\text{Bi}_2\text{Sr}_2\text{RCu}_2\text{O}_8$ ($R = \text{Dy, Er, Y}$ and $\text{Ca}_{0.5}\text{Er}_{0.5}$) and the other was a set of the superconductors expressed as $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Pr}_x\text{Cu}_2\text{O}_8$ ($x = 0, 0.10, 0.25, 0.36$ and 0.42). All the crystals were prepared using a self-flux method, and the preparation conditions are given in reference [4] for the former set and in reference [10] for the latter set. The as-grown crystals with typical dimensions of $1 \times 1 \times 0.02 \text{ mm}^3$ were characterized through x-ray diffraction, energy-dispersive x-ray and resistivity measurements. The hole concentration per Cu was estimated from the empirical relation to the room-temperature thermopower [11].

The in-plane thermopower was measured using a steady-state technique from 4.2 to 420 K, where different measurement systems were used below and above room temperature. One edge of the sample was pasted on a metal plate working as a heat bath, while the other edge was pasted on a heater generating a temperature gradient. The temperature gradient was measured with a differential thermocouple attached between the metal plate and the heater, and the temperature was monitored by a resistance thermometer near the sample. For the measurement system below room temperature, a Cu sheet, a film-chip resistor and a Cu–constantan thermocouple were used as the metal plate, the sheet heater and the differential thermocouple respectively. For the measurement system above room temperature, a Pt sheet, a Nichrome film and a Pt–Ph thermocouple were used as the metal plate, the sheet heater and the differential thermocouple respectively. For both systems, the applied temperature gradient was typically $0.5\text{--}1.0 \text{ K mm}^{-1}$. The signal voltage was measured using a nanovoltmeter (HP 34420A), and a typical accuracy of $0.1 \mu\text{V K}^{-1}$ was realized, though the thermoelectric voltage was less accurate for a highly resistive sample. The contribution from the voltage leads was carefully subtracted.

3. Results and discussion

Figure 1(a) shows the in-plane resistivity ρ_{ab} of $\text{Bi}_2\text{Sr}_2\text{RCu}_2\text{O}_8$ ($R = \text{Dy, Er, Y}$ and $\text{Ca}_{0.5}\text{Er}_{0.5}$). All the resistivities increase with decreasing temperature (T), which indicates that the samples are in the parent–insulator region. The sample for $R = \text{Ca}_{0.5}\text{Er}_{0.5}$ is the most conductive, whereas the sample for $R = \text{Dy}$ is highly insulating. Note that the resistivity strongly depends on the rare-earth ion $R = \text{Dy, Er}$ and Y . The lattice parameters gradually decrease with R going from La to Lu owing to the lanthanoid contraction, which systematically changes the excess oxygen, the carrier concentration, the bandwidth or the interdiffusion with Sr. A similar case is seen for $\text{Pb}_2\text{Sr}_2\text{RCu}_3\text{O}_8$: the samples for $R = \text{Eu, Dy}$ and Y are superconducting, while the samples for $R = \text{Nd}$ and Sm are insulating [12].

Figure 1(b) shows ρ_{ab} for $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Pr}_x\text{Cu}_2\text{O}_8$ ($x = 0, 0.10, 0.25, 0.36$ and 0.42), which is in good agreement with ρ_{ab} for well-characterized crystals of $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Y}_x\text{Cu}_2\text{O}_8$ [13]. The $x = 0$ sample has a high T_c of 83 K, and is slightly overdoped. A small amount of Pr doping reduces the carrier concentration and improves the crystal quality. The $x = 0.25$ sample shows the ‘ T -linear’ resistivity without significant residual resistivity, and its T_c can

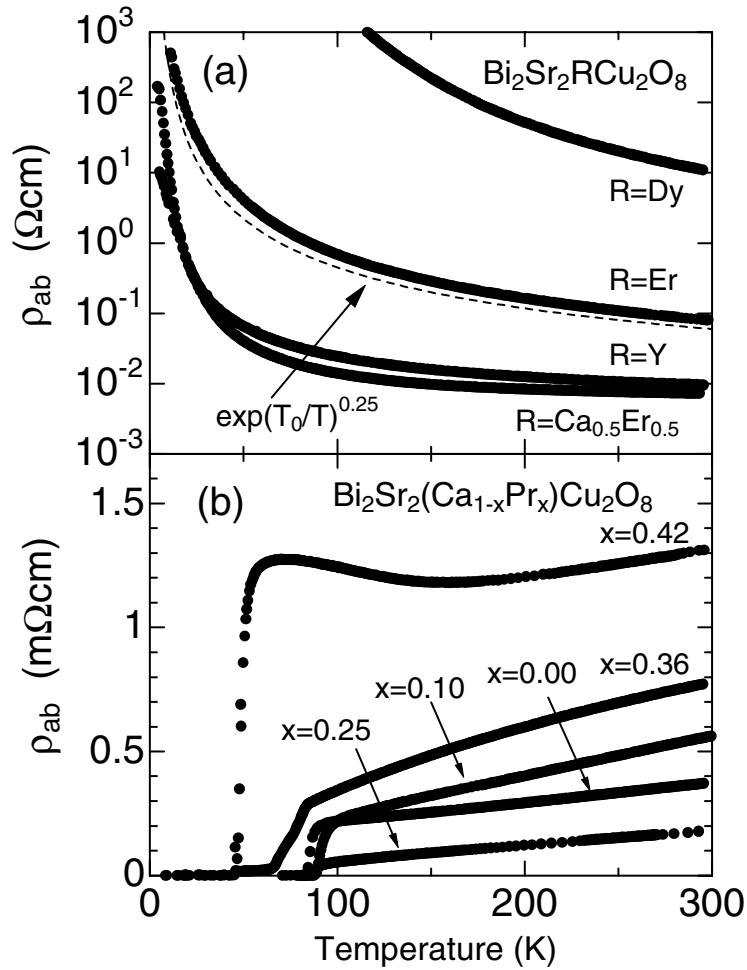


Figure 1. (a) The in-plane resistivity (ρ_{ab}) of insulating $\text{Bi}_2\text{Sr}_2\text{RCu}_2\text{O}_8$ ($R = \text{Dy}, \text{Er}, \text{Y}$ and $\text{Er}_{0.5}\text{Ca}_{0.5}$) single crystals. (b) ρ_{ab} for superconducting $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Pr}_x\text{Cu}_2\text{O}_8$. The dashed curve in (a) represents a resistivity in the variable-range-hopping regime $\rho \propto \exp[(T_0/T)^{1/4}]$ (see the text).

be as high as 88 K with a narrow transition width of 3 K. Further substitution of Pr for Ca causes an increase in resistivity accompanied by a decrease in T_c , and an upturn is seen near T_c for the $x = 0.42$ sample.

Figure 2(a) shows the in-plane thermopower $S(T)$ of $\text{Bi}_2\text{Sr}_2\text{RCu}_2\text{O}_8$ ($R = \text{Dy}, \text{Er}, \text{Y}$ and $\text{Er}_{0.5}\text{Ca}_{0.5}$). Similarly to the case for ρ_{ab} in figure 1(a), the magnitude of the thermopower increases from $R = \text{Er}_{0.5}\text{Ca}_{0.5}$ to Dy. (Note that the magnitude for $R = \text{Dy}$ ($350 \mu\text{V K}^{-1}$ at 300 K) is plotted with a 50% reduction.) As shown by the dashed curves, $S(T)$ is roughly proportional to \sqrt{T} below about 150 K, which can be understood from three-dimensional variable-range-hopping (3D-VRH) transport [14]. This is consistent with the fact that the low-temperature resistivity is also explained in terms of 3D-VRH where ρ_{ab} is proportional to $\exp[(T_0/T)^{1/4}]$ (T_0 is a characteristic temperature), as shown by the dashed curve in figure 1(a). It is worth noting that $S(T)$ decreases with T above 300 K, which is roughly expressed by $S(T) = A - BT$ (A and B are positive constants). The high-temperature thermopower

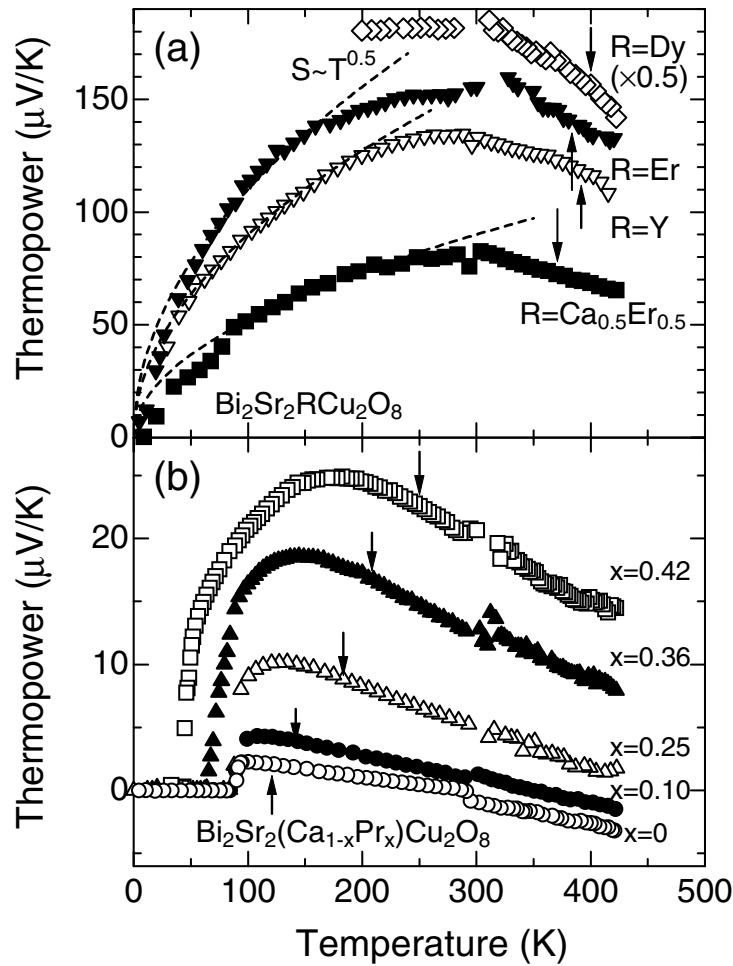


Figure 2. (a) The in-plane thermopower $S(T)$ of insulating $\text{Bi}_2\text{Sr}_2\text{RCu}_2\text{O}_8$ ($R = \text{Dy}$, Er , Y and $\text{Ca}_{0.5}\text{Er}_{0.5}$) single crystals. Note that the magnitude for $R = \text{Dy}$ ($350 \mu\text{V K}^{-1}$ at 300 K) is reduced by 50%. (b) $S(T)$ for superconducting $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Pr}_x\text{Cu}_2\text{O}_8$. The arrows indicate the scaling temperature T_{scale} . The dashed curves in (a) represent thermopowers in the variable-range-hopping regime $S \propto \sqrt{T}$ (see the text).

for strongly correlated systems is often described by the Heikes formula [15], where $S(T)$ is independent of T . For example, the cobalt oxide $\text{Bi}_2\text{Sr}_3\text{Co}_2\text{O}_9$ which is isomorphous to Bi-2212 shows a T -independent $S(T)$ above 200 K [16]. Mandal *et al* [7] attributed the decreasing $S(T)$ to an activation-type transport expressed by $S(T) = C/T + D$, where C and D are constants. Because of the limited range of the temperature measured, the data can be fitted by either $A - BT$ or $C/T + D$. We fitted our data with $C/T + D$ above 300 K, and found a small value of C (of the order of $k_B T$) and no systematic change in D . These results are seriously incompatible with the thermopower of semiconductors. We further note that ρ_{ab} of the VRH type shows weaker temperature dependence than activation-type resistivity ($\propto \exp(1/T)$), which is usually seen in other parent insulators [17]. We therefore think that there would be no reason to assume a finite activation energy in the parent insulators at high temperatures.

One can see that the thermopower of the insulating Bi-2212 shows a maximum, because it is proportional to \sqrt{T} at low temperatures and decreases with T at high temperatures. This is essentially the same as what is seen for the superconducting $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Pr}_x\text{Cu}_2\text{O}_8$, above T_c , as is shown in figure 2(b). The peak position shifts to lower temperature with increasing carrier concentration; this is accompanied by a decrease of the magnitude. This implies that S is scaled at the peak position. As is marked with an arrow in figure 2, we define the scaling temperature T_{scale} such that $S(T)/S(T_{\text{scale}})$ will be scaled to a master curve as a function of T/T_{scale} .

Figure 3 shows $S(T)/S(T_{\text{scale}})$ for all the samples as a function of T/T_{scale} , where all the data, for both the insulators and the superconductors, are scaled to a *single* curve. We emphasize that this scaling is not specific to Bi-2212, but seems to be common to all HTSC. As is shown in the inset of figure 3, the same scaling is successfully applied to the data for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ taken from reference [18]. Very recently, Yamamoto *et al* [19] have found that the same scaling is also valid for $\text{HgBa}_2\text{CuO}_{4+\delta}$. One may notice that the data for $R = \text{Dy}$ deviate from the master curve for $T < T_{\text{scale}}$. This arises from the nearly T -independent $S(T)$ below 300 K. We cannot answer at present the question of whether it is intrinsic or not. Since the $R = \text{Dy}$ compound is nearly undoped, the resistivity becomes extremely high with decreasing temperature, which makes the low-temperature thermopower highly unreliable.

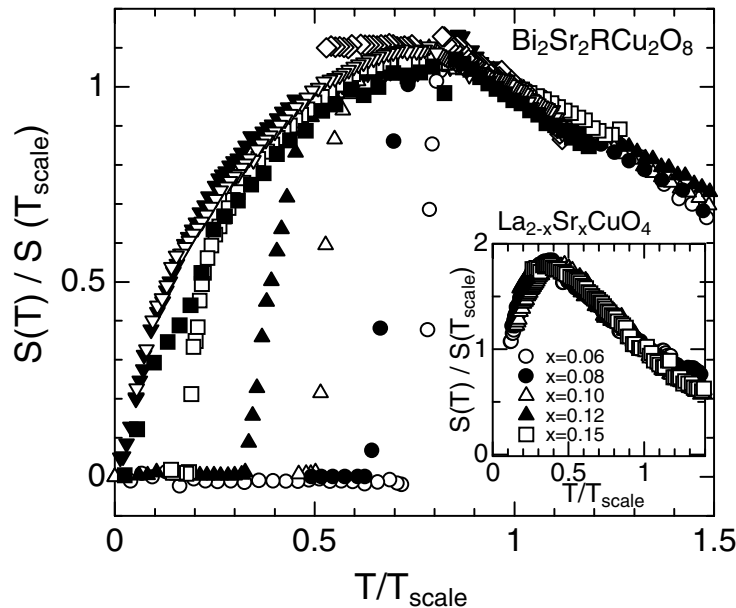


Figure 3. $S(T)/S(T_{\text{scale}})$ plotted as a function of T/T_{scale} . Note that the data are plotted using the same symbols as in figure 2. In the inset we show the same scaling for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ using the data from reference [18].

Figure 4 shows T_{scale} and T_c plotted as functions of hole concentration per Cu (p). T_{scale} decreases linearly with p , and the magnitude is very close to the pseudogap temperature T^* [20]. Mandal *et al* [7] obtained similar results, but they did not associate T_{scale} for the insulators with the pseudogap. In the inset of figure 4, T_{scale} for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ is plotted as a function of the Sr content, which is again consistent with the pseudogap temperature detected through other experiments [20]. We therefore propose that thermopower measurement is an easy way to estimate the pseudogap temperature from charge transport.

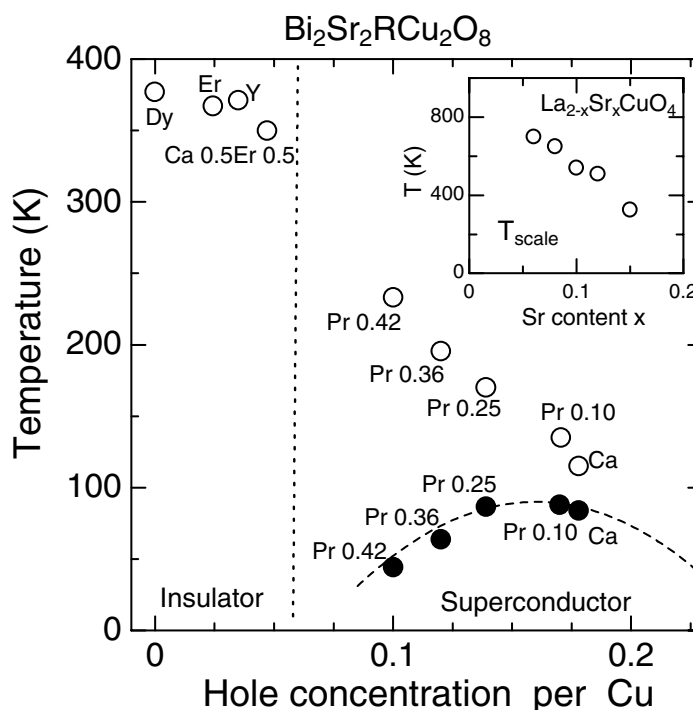


Figure 4. T_{scale} (open circles) and T_c (filled circles) plotted as functions of hole concentration per Cu estimated from the room-temperature thermopower. The data for $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Pr}_x\text{Cu}_2\text{O}_8$ are denoted as 'Pr x ', while the data for $\text{Bi}_2\text{Sr}_2\text{RCu}_2\text{O}_8$ are denoted as 'R'. In the inset we show T_{scale} for $\text{La}_{1-x}\text{Sr}_x\text{CuO}_4$ as a function of the Sr content x .

The present study further suggests that the parent insulator has a pseudogap above room temperature. One may think that an antiferromagnetic order cannot coexist with a pseudogap, because a magnon is a gapless excitation. This is not always true: the antiferromagnetic order does coexist with the gapped state in the disordered spin-Peierls compound $\text{Cu}_{1-x}\text{Zn}_x\text{GeO}_3$ [21]. Although the gapless magnon is well defined near the zone boundary, the spin-Peierls gap remains almost unchanged in the rest of the k -space. It should be further explored whether the same scenario can be applied to the parent insulator or not. At least we can say that the pseudogap evaluated here is consistent with the photoemission experiment of [3], in the sense that the ratio of T_{scale} to T_c is nearly the same as the ratio of the d-wave-like gap in the parent insulator to the superconducting gap of optimally doped HTSC.

Let us consider the physical meaning of the scaling. From the theoretical point of view, the pseudogap corresponds to spinon-spinon pairing [22], the preformed pair [23], the flux phase [24], or charge/spin-density-wave formation [25]. In these theories, the pseudogap temperature is the mean-field transition temperature T_{c0} for superconductivity or charge/spin-density waves. In either case, the density of states is scaled as a function of a reduced temperature T/T_{c0} , and the density of states of the HTSC is expected to be scaled as a function of T/T_{scale} . Then the normal-state parameters will also be scaled through the scaling relation of the density of states.

In the present paper we have not specified the origin of $S = A - BT$, which has been explained by several theories. Trodahl [26] has shown that $A - BT$ can be understood as a simple sum of the phonon drag term and the diffusive term. Forro *et al* [27] have assumed a

narrow band overlapping with a broad band as analogous to a heavy-fermion system, and have proposed the expression $S = C_1T + C_2T/(C_3^2 + T^2)$. By choosing an appropriate set of the parameters C_i , S is roughly reduced to $A - BT$ at high temperatures. Nagaosa and Lee [28] have calculated the thermopower in the spin-charge separation scenario. They have found $S = C_1 - C_2 \log T - C_3T$, which is reduced to $A - BT$ by neglecting the weak temperature dependence of $\log T$. The $C_1 - C_2 \log T$ term corresponds to a holon (charged boson) and the C_3T term corresponds to a spinon (neutral fermion). Coleman *et al* [29] have attributed $S = A - BT$ to two different scattering times in their charge-conjugation theory. Since our results reveal that the parent insulators also show $S = A - BT$, the theories of [28] and/or [29] are most likely; these seem still valid in the heavily underdoped region. Further investigation is of course necessary for a quantitative comparison with these theories.

4. Summary

We prepared a series of single crystals of $\text{Bi}_2\text{Sr}_2\text{RCu}_2\text{O}_8$ ($R = \text{Dy, Er, Y}$ and $\text{Ca}_{0.5}\text{Er}_{0.5}$) and $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Pr}_x\text{Cu}_2\text{O}_8$ ($x = 0, 0.10, 0.25, 0.36$ and 0.42), and measured the thermopower $S(T)$ along the CuO_2 direction. We have found that $S(T)/S(T_{\text{scale}})$ for all of the samples falls onto a single master curve as a function of T/T_{scale} , which strongly suggests that the density of states of the parent insulator is essentially similar to that of high- T_c superconductors. Since the scaling temperature is found to be close to the pseudogap temperature, we propose that thermopower measurement is an easy way to estimate the pseudogap temperature for high- T_c superconductors.

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