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Thermal conductivity and thermoelectric power of DyBaCuO bulk superconductors

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Abstract
The *ab*-plane thermal conductivity $\kappa(T)$ and thermoelectric power $S(T)$ have been investigated for DyBaCuO bulk superconductors for various contents of the Dy_2BaCuO_5 (Dy211) phase X. The absolute $\kappa(T)$ value is pretty small compared with that of other REBaCuO bulks (RE = Y and rare earth elements). The $S(T)$ value above the superconducting transition temperature T_c changes from negative to positive and $\kappa(T)$ slightly decreases with increasing oxygen deficiency in the $DyBa_2Cu_3O_{7-\delta}$ (Dy123) superconducting phase. The point-defect-type phonon scattering due to oxygen deficiency affects the $\kappa(T)$ value, but is not a main origin for the low $\kappa(T)$ even for the fully oxidized DyBaCuO bulk. Another type of powerful phonon scattering centre governs the thermal transport properties in the DyBaCuO bulk.

1. Introduction

For practical applications and fundamental research of melt-processed REBaCuO bulk superconductors (RE: Y and rare earth elements), thermal properties such as the thermal conductivity κ, thermal diffusivity α and thermal dilatation dL/L are valuable parameters, besides the electro-magnetic properties such as the superconducting transition temperature T_c and the critical current density J_c. The REBaCuO bulk is an anisotropic and complex material, which consists of the $REBa_2Cu_3O_{7-\delta}$ (RE123) superconducting phase, the RE_2BaCuO_5 (RE211) non-superconducting phase and Ag and Pt metals. The thermal properties depend on the quality and alignment of the RE123 phase, and the content, size and dispersion of the RE211 phase and added metals. Defects such as voids and cracks are also influencing factors.

We have measured the thermal and mechanical properties at low temperatures (4–300 K) and under a high magnetic field (0–10 T) for the REBaCuO bulks, which were fabricated by several companies and institutes. We have constructed a database of these properties, which is available online [1, 2]. The absolute value and temperature dependence of the thermal conductivity $\kappa(T)$ changes depending on the species of the RE ions; $\kappa(T)$ of RE = Y, Gd shows a high value and the $d\kappa/dT \leqslant 0$ behaviour above T_c [3, 4]. On the other hand, $\kappa(T)$ of RE = Sm, Nd shows a somewhat low value and the $d\kappa/dT \geqslant 0$ behaviour above T_c due to the light RE (LRE) element substitution for the Ba site [5]. The increased migration of LRE and Ba enhances the phonon scattering in the superconducting phase, working as strong phonon scattering centres. Recently, we reported that the DyBaCuO bulk showed a lower $\kappa(T)$ of about a half or one-third value than that of the YBaCuO bulk, but had a comparable high-J_c value [6]. In the RE = Y and Dy systems, however, it has been considered that the migration does not take place during the crystal growth due to the non-LRE element. The origin of the low $\kappa(T)$ in the DyBaCuO system is still an open question.

In this paper, we study the temperature dependence of the *ab*-plane thermal conductivity $\kappa(T)$ and thermoelectric power $S(T)$ of the DyBaCuO bulks with various Dy211 contents X. The samples with different oxygen content in the Dy123 phase are prepared and the relation between the $\kappa(T)$ and the oxygen deficiency is investigated. The $S(T)$ has proved to be a sensitive probe to estimate the oxygen deficiency in the REBaCuO superconductor. The origin of the low thermal conductivity for the DyBaCuO bulk is discussed from the viewpoint of the oxygen deficiency.

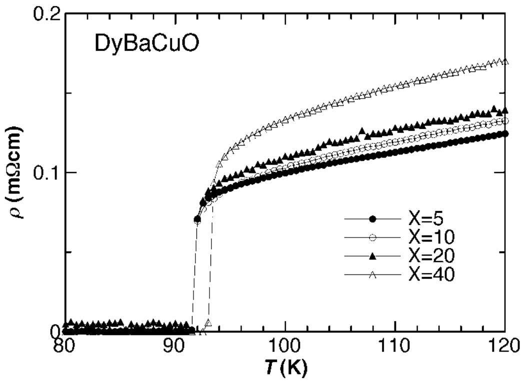

Figure 1. The temperature dependence of the *ab*-plane electrical resistivity $\rho(T)$ for the DyBaCuO bulk for various Dy211 contents X.

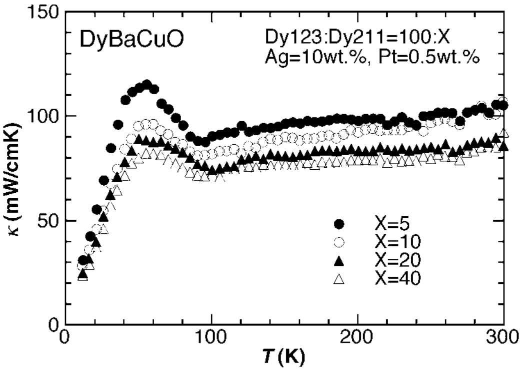

Figure 2. The *ab*-plane thermal conductivity $\kappa(T)$ for the DyBaCuO bulk for various Dy211 contents X.

2. Experimental details

The DyBaCuO bulk samples were fabricated at the International Superconductivity Technology Center, Superconductivity Research Laboratory (ISTEC-SRL). The content X of the Dy211 phase in the Dy123 phase was changed from 5 to 40 in the molar ratio of Dy123:Dy211 = 100:X. 10 and 0.5 wt% Ag_2O and Pt powders were added in all the samples [7, 8]. The mixed powders were uniaxially pressed into pellets 40 mm in diameter and 25 mm in thickness. Melt-processing was performed by the hot-seeding method in air. The melt-grown Nd123(100) crystal was used as a seed. The pellets were heated to 1100 °C and held for 0.5 h, cooled down to 1025 °C, then cooled at a rate of 0.15–0.5 °C h^{-1} to 950 °C and finally cooled down at a rate of 100 °C h^{-1} to room temperature. The grown bulks were heat treated in flowing oxygen gas at temperature T_{HT} = 400 °C for 100 h, which seems to be an optimum heat treatment temperature to obtain the highest T_c. For the measurements, rectangular pieces with dimensions of about 2.5 × 2.5 × 10.0 mm^3 were cut from the heat-treated bulks. In order to clarify the influence of the oxygen deficiency in the Dy123 phase on the $\kappa(T)$ and $S(T)$ behaviours, the as-grown bulks (X = 5) were heat treated in flowing oxygen at T_{HT} = 500 and 550 °C for 100 h. The heat treatment at higher T_{HT} is expected to increase the oxygen deficiency δ in the Dy123 phase. The *ab*-plane thermal conductivity $\kappa(T)$ and thermoelectric power $S(T)$ were simultaneously measured by a steady-state heat flow method using a Gifford–McMahon (GM) cycle helium refrigerator as a cryostat between 10 and 300 K [9]. The electrical resistivity $\rho(T)$ was measured by a four-terminal method.

3. Results and discussion

3.1. Dy211 content X dependence of $\kappa(T)$ and S(T)

Figure 1 shows the temperature dependence of the *ab*-plane electrical resistivity $\rho(T)$ for the DyBaCuO bulk for various Dy211 contents X. The ρ value increases with increasing X, which may result from the increase of the insulating Dy211 fraction. The superconducting transition temperature T_c is almost identical (=91.5 K) for X = 5–20 but T_c of the X = 40 sample is somewhat higher at T_c = 93 K.

Figure 2 presents the temperature dependence of the *ab*-plane thermal conductivity $\kappa(T)$ for the DyBaCuO bulk for various Dy211 content X. $\kappa(T)$ gradually decreases with decreasing T, shows a characteristic peak below T_c and then decreases to zero at low temperatures. The absolute $\kappa(T)$ value decreases with increasing X. For the X = 5 sample, which contains the maximum amount of Dy123 superconducting phase, the absolute κ value above T_c is larger and the $\kappa(T)$ enhancement below T_c is prominent. For the X = 10 sample, the $\kappa(T)$ enhancement decreases slightly and it is further reduced with further increase of X. However, it is to be noticed that absolute $\kappa(T)$ values of X = 20 and 40 samples almost coincide with each other over the entire temperature range. $\kappa(T)$ of high-T_c superconductors (HTSCs) is enhanced below T_c by both the electronic contribution κ_e and the phonon contribution κ_{ph} [10, 11]. In impure HTSCs with sizable residual electron scattering by impurities, the $\kappa(T)$ enhancement mainly comes from the phonon component κ_{ph}, which is caused by the reduction in the phonon scattering by quasi-particles with the progress of their condensation into Cooper pairs. Because the present DyBaCuO samples belong to the impure superconductor containing the Dy211 and Ag phases, the κ enhancement is due to the κ_{ph} contribution. The electronic thermal conductivity $\kappa_e(T)$ above T_c, estimated by the Wiedemann–Franz law using the ρ value, decreases with increasing X. The ratio of κ_e to the total κ at 200 K is 24% for all the X values. The decrease of $\kappa(T)$ with increasing X above T_c may come mainly from the reduction of κ_e, the increase of low thermal conductive Dy211 phase and the enhanced phonon scattering in the Dy123 phase. The κ_{ph} enhancement is masked out if the phonon scattering strength by other mechanisms becomes stronger. The slight suppression of the κ enhancement with increasing X may reflect the enhanced phonon scattering.

Figure 3 shows the temperature dependence of the *ab*-plane thermoelectric power $S(T)$ for the DyBaCuO bulk for various Dy211 contents X. In the normal state, the $S(T)$ value is negative and approaches zero with decreasing temperature T. The absolute S value decreases with increase of X. Quantitatively, the absolute S value is expected to increase with some kinds of defects because of increasing resistivity ρ values. It was reported that, in the $YBa_2Cu_3O_{7-\delta}$ polycrystals, the S value continuously changes from negative to positive with increasing oxygen deficiency δ, which also

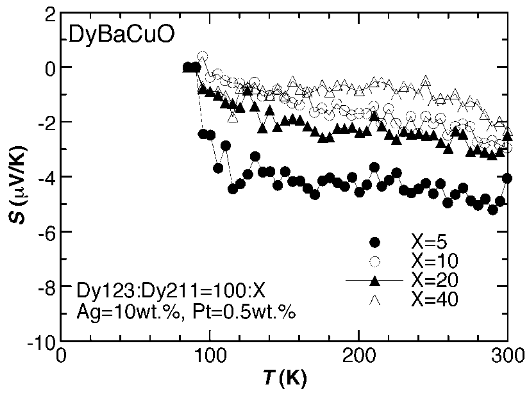

Figure 3. The *ab*-plane thermoelectric power $S(T)$ for the DyBaCuO bulk for various Dy211 contents X.

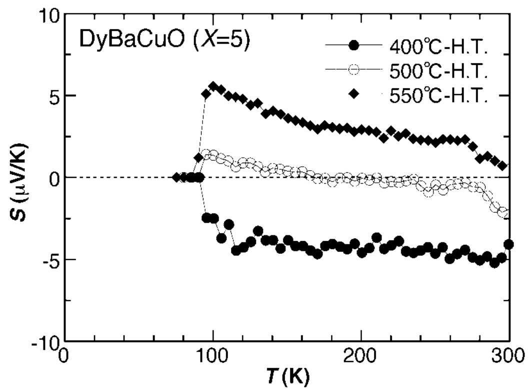

Figure 4. The temperature dependence of the $S(T)$ for the $X = 5$ samples with different heat treatment temperatures T_{HT}.

reflects the ρ values [12]. The observed $S(T)$ behaviour in the present DyBaCuO bulks may partly be attributable to the $\rho(T)$ increase in the Dy123 phase caused by the crystal defect and/or the increase of δ. The $S(T)$ of the SmBaCuO bulk system with Sm123:Sm211 = 100:X is positive and increases with increasing X, which may partly be attributable to the $\rho(T)$ increase in the Sm123 phase caused by the migration effect [5]. If the δ value is nearly identical for all the present DyBaCuO samples due to the identical heat treatment condition in flowing oxygen, the approach of absolute S value to zero with increasing X results from the crystal defect.

3.2. Influence of oxygen deficiency on $\kappa(T)$ and S(T)

The $X = 5$ samples which were heat treated in flowing oxygen at $T_{HT} = 500$ and 550 °C for 100 h were prepared beside the $X = 5$ sample heat treated at $T_{HT} = 400$ °C shown in section 3.1. Figure 4 shows the temperature dependence of the $S(T)$ for the $X = 5$ samples with different T_{HT}. The sign of $S(T)$ changes from negative to positive with increasing T_{HT}. Since the heat-treated bulk at higher T_{HT} seems to have the larger oxygen deficiency δ, the sign change in $S(T)$ comes from oxygen deficiency δ in the Dy123 phase, similarly to the YBaCuO [12]. The δ value for each T_{HT} cannot be estimated quantitatively because the bulk sample is a mixture of Dy123 and Dy211.

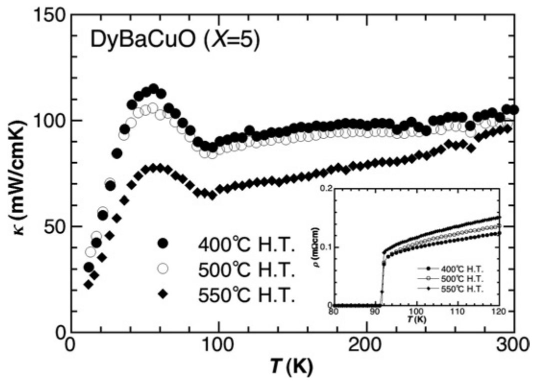

Figure 5. $\kappa(T)$ for the $X = 5$ samples with different T_{HT}. The inset shows the resistivity $\rho(T)$ for the same samples.

Figure 5 shows $\kappa(T)$ for the $X = 5$ samples with different T_{HT}. The inset shows the resistivity $\rho(T)$ for the same samples, which show the identical T_c value. For the heat-treated sample at $T_{HT} = 500$ °C, $\kappa(T)$ hardly changes compared with the sample commonly heat-treated at $T_{HT} = 400$ °C. On the other hand, for the heat-treated sample at $T_{HT} = 550$ °C, the $\kappa(T)$ enhancement is reduced and the magnitude of the κ value decreases. These results suggest that a small amount of oxygen deficiency has a fair influence on the magnitude of $\kappa(T)$, but is not the main origin for the low $\kappa(T)$ even in the fully oxidized DyBaCuO bulk. We have measured $\kappa(T)$ for $REBa_2Cu_3O_{7-\delta}$ polycrystals (RE = Y, Dy, Sm, Nd) and analysed the phonon scattering mechanisms using the phonon conduction model [13]. The estimated phonon mean free path l_b for the DyBaCuO system is pretty short compared with other REBaCuO systems, which suggests that a sizable crystal defect exists. A small amount of migration of Dy and Ba may take place, although there has not been the experimental obtained evidence to date for the migration of the Dy ion and the Ba ion.

4. Summary

The *ab*-plane thermal conductivity $\kappa(T)$ and thermoelectric power $S(T)$ have been investigated for the DyBaCuO bulk superconductors for various contents of the Dy_2BaCuO_5 (Dy211) phase X and for various oxygen contents 7-δ in the $DyBa_2Cu_3O_{7-\delta}$ (Dy123) phase. The absolute $\kappa(T)$ value is pretty small even for the $X = 5$ sample. The oxygen deficiency δ slightly affects the absolute $\kappa(T)$ value within the δ value where the superconducting transition temperature T_c is kept at 91 K. However, the oxygen deficiency is not the main origin of the low $\kappa(T)$ behaviour for the fully oxidized DyBaCuO bulk. Powerful phonon scattering centres, which drastically reduce the $\kappa(T)$ value, must exist in the DyBaCuO bulk, compared with other REBaCuO bulks (RE = Y, Gd). A crystal defect which is inherent for the DyBaCuO bulk may exist. A detailed investigation is in progress.

Acknowledgments

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