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Thermal conductivity of Er–Ba–Cu–O and Ho–Ba–Cu–O superconducting bulks

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Abstract
We have measured the temperature dependence of the in-plane thermal conductivity, $\kappa_{ab}(T)$, for both Er–Ba–Cu–O and Ho–Ba–Cu–O superconducting bulks. The absolute values of $\kappa_{ab}(T)$ for both bulks are smaller than that of Y–Ba–Cu–O bulk, but are larger than that of Dy–Ba–Cu–O bulk, which is known to show an anomalous small κ_{ab}. $\kappa_{ab}(T)$ for Ho–Ba–Cu–O shows a positive slope ($\mathrm{d}\kappa_{ab}/\mathrm{d}T > 0$) in the normal state, similarly to that for Dy–Ba–Cu–O. On the other hand, a slightly negative slope ($\mathrm{d}\kappa_{ab}/\mathrm{d}T < 0$) is found for Er–Ba–Cu–O. The out-of-plane thermal conductivity, κ_c, was also measured for Er–Ba–Cu–O bulk, and the anisotropy in the thermal conductivity, (κ_{ab}/κ_c), was estimated to be about 2.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Using a melt-texture growth method [1], we can obtain a large RE–Ba–Cu–O (REBCO; RE = rare earth elements) quasi-single crystal, which consists of a superconducting matrix phase of $REBa_2Cu_3O_{7-\delta}$(RE123) and a non-superconducting secondary phase of RE_2BaCuO_5(RE211). For designing a superconducting application such as a current lead, the thermal conductivity, κ, is an important physical property, because the estimation of the heat intrusion is necessary [2]. Therefore, we have measured the thermal conductivity for various REBCO bulk superconductors at low temperatures (below 300 K), and have constructed a database of the results obtained [3, 4]. It has been found that the temperature dependence of the thermal conductivity in the ab-plane, $\kappa_{ab}(T)$, of the REBCO bulk strongly depends on the kind of RE ion, except for the heavy RE ions such as Ho, Er, and so forth. There are few data for κ for REBCO containing heavy RE ions; in particular, the effect of the oxygen deficiency and the anisotropic behaviour have not been studied at all.

For YBCO bulk [5], $\kappa_{ab}(T)$ increases with decreasing temperature above the critical temperature T_c. Since the main heat carriers in the high-T_c cuprates are phonons, such temperature dependence is considered to originate from the phonon–phonon Umklapp process, which dominates the scattering mechanisms of the heat transport. In the superconducting state, $\kappa_{ab}(T)$ starts to increase rapidly on cooling below T_c and has a maximum at around 50–60 K, which is well known feature for the high-T_c cuprates [6]. In the case of SmBCO bulk [7], $\kappa_{ab}(T)$ shows a temperature-independent behaviour in the normal state and the enhancement below T_c is somewhat suppressed, which is interpreted to come from the substitution [8] between the Sm and Ba sites.

Recently, $\kappa_{ab}(T)$ of DyBCO bulk was found to show the following anomalous features [2, 9]. First, the absolute value of $\kappa_{ab}(T)$ is significantly smaller than that of other bulks: half or one-third of that in YBCO bulk. Second, the peak structure below T_c is strongly suppressed. Third, the slope of $\kappa_{ab}(T)$ in the normal state is positive, $\mathrm{d}\kappa_{ab}/\mathrm{d}T > 0$. A possible origin of these anomalies in DyBCO is the difference of ionic radius and/or mass of the RE. Although the ionic radius of Dy^{3+} is slightly larger than that of Y^{3+}, it is believed that the substitution between Dy and Ba sites hardly occurs, unlike in SmBCO bulk. On the other hand, the effect of the mass is not clear, and the origin of the anomalous $\kappa_{ab}(T)$ of DyBCO bulk

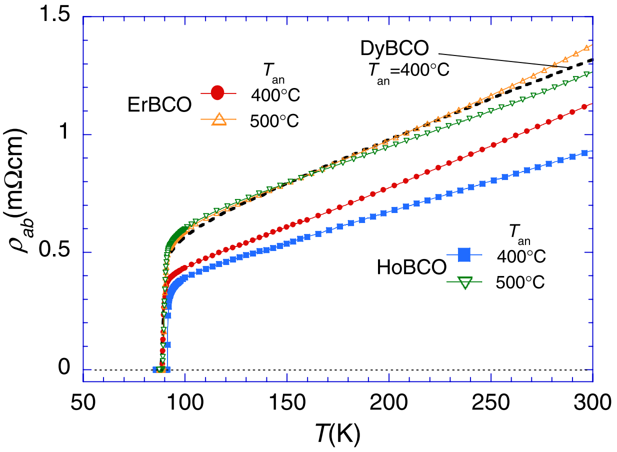

Figure 1. The temperature dependence of the in-plane resistivity, $\rho_{ab}(T)$, of RE–Ba–Cu–O (REBCO; RE = Er, Ho) bulk superconductors for different annealing temperatures, T_{an}s, 400 and 500 °C. The broken line shows the data for DyBCO bulk (T_{an} = 400 °C) as a reference.

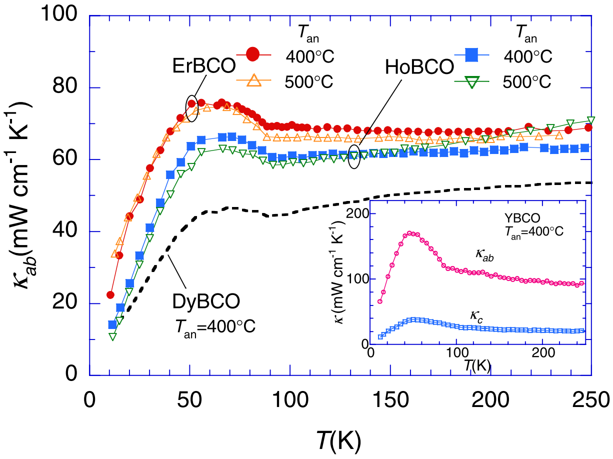

Figure 2. The temperature dependence of the in-plane thermal conductivity, $\kappa_{ab}(T)$, of RE–Ba–Cu–O (REBCO; RE = Er, Ho) bulk superconductors for the different annealing temperatures, T_{an}s, 400 and 500 °C. The broken line shows the data for the DyBCO bulk (T_{an} = 400 °C) as a reference. The inset shows $\kappa_{ab}(T)$ and the thermal conductivity along the c-axis, $\kappa_c(T)$, for YBCO bulk annealed at 400 °C [2].

Table 1. Specification of samples.

		T_c (K)		
Sample	RE123:RE211	T_{an} = 400 °C	450 °C	500 °C
Er–Ba–Cu–O	1:0.33	88.8	88.8	89.4
Ho–Ba–Cu–O	1:0.4	91.5	—	88.9
Dy–Ba–Cu–O	1:0.4	87.8	—	—

remains an unsolved problem. However, DyBCO has already been used for the current lead, utilizing its low κ [2]. Therefore it is interesting to examine the nature of $\kappa(T)$ of REBCO bulks containing the heavier RE elements in terms of the physics and practical applications.

In this paper, we study mainly the temperature dependence of the in-plane thermal conductivity, $\kappa_{ab}(T)$, as a function of annealing temperature, which is known to affect the oxygen deficiency, for both ErBCO and HoBCO bulks; Er and Ho are both heavier than Dy. We found that the absolute values of $\kappa_{ab}(T)$ of both bulks are larger than that of DyBCO bulk, and that the broad peak below T_c is also more visible. However, both the absolute value and peak height in $\kappa_{ab}(T)$ were still somewhat smaller than those of YBCO bulk. In addition, we estimated the anisotropy in the thermal conductivity for ErBCO bulk.

2. Experimental details

ErBCO and HoBCO bulks were grown by a melt-texture technique [10, 11]. For comparison, a DyBCO bulk was also prepared. The molar ratios of RE123 with RE211 powders were 1:0.33 (RE = Er), 1:0.4 (RE = Ho), and 1:0.4 (RE = Dy), respectively. 0.5 wt% Pt powder was added in all samples. The rectangular shaped sample was prepared by cutting an as-grown bulk near the seed crystal. The amount of oxygen deficiency, δ, in the RE123 phase of the sample was varied by controlling the annealing temperature [12]. In this study, the samples were annealed at 400–500 °C for 7 days in 0.1 MPa flowing oxygen gas. As shown in table 1, the superconducting transition temperatures, T_cs, of all samples were about 90 K, obtained by electrical resistivity measurement. The thermal conductivity was measured by a conventional steady-state heat flow method [5]. The temperature gradient of the sample was estimated by measuring the temperature difference along the sample using chromel–constantan thermocouples. The electrical resistivity was measured by a conventional four-probe method with a typical current density of about 0.1 A cm^{-2}. The temperature of the sample stage was controlled from 6 to 300 K using a Gifford–McMahon cycle helium refrigerator. To eliminate radiation loss, the thermal conductivity was measured below 250 K.

3. Results and discussion

3.1. Thermal conductivity in the ab-plane for Er–Ba–Cu–O and Ho–Ba–Cu–O bulks

Figure 1 shows the temperature dependence of the in-plane resistivity, $\rho_{ab}(T)$, for the ErBCO and HoBCO bulks with different annealing temperatures [12], T_{an}s, of 400 and 500 °C. The data of DyBCO bulk (T_{an} = 400 °C) are also shown as a reference. All samples show the sharp superconducting transition at around 90 K. The value of T_c for ErBCO is almost independent of T_{an}. On the other hand, T_c for HoBCO changes with T_{an}: T_c for the sample annealed at T_{an} = 400 °C is slightly higher than that for T_{an} = 500 °C. Although the T_c value does not systematically change with T_{an}, the absolute ρ_{ab} values increase with increasing T_{an}, meaning that the amount of the oxygen vacancies for the T_{an} = 400 °C sample is smaller than that for the T_{an} = 500 °C sample in both bulks.

Figure 2 shows the temperature dependence of the in-plane thermal conductivity, $\kappa_{ab}(T)$, for both ErBCO and

HoBCO bulks. The data for DyBCO bulk (T_{an} = 400 °C) are also shown as a reference. In the ErBCO bulk, κ_{ab} slightly increases with decreasing temperature in the normal state for samples annealed at both 400 and 500 °C. Note that the slope of the latter sample is somewhat small. In the superconducting state, a broad peak can be observed, and the κ_{ab} values at the peak temperature are almost the same in both samples. For HoBCO bulks, $\kappa_{ab}(T)$ of the sample annealed at 400 °C shows a slight positive slope ($d\kappa_{ab}/dT > 0$) in the normal state which becomes more discernible in the sample annealed at 500 °C. The κ_{ab} enhancement below T_c for HoBCO bulks is relatively smaller than that for ErBCO bulks.

The magnitude of $\kappa_{ab}(T)$ of ErBCO bulk is larger than that of HoBCO bulk. This result does not come from the different composition ratio between RE123 and RE211, because we previously found that the κ_{ab} values for the SmBCO and GdBCO bulks are almost independent of the composition ratio in the case of high RE211 content, RE211/RE123 $\geqslant$ 0.3 [4, 7].

The absolute κ_{ab} values for both ErBCO and HoBCO bulks studied here are larger than that for DyBCO bulk. However, they are still smaller than that of YBCO bulk. For comparison, the $\kappa_{ab}(T)$ data for YBCO bulk [2] are shown in the inset of figure 2. It is well known that a similar small magnitude of κ_{ab} was observed in SmBCO and NdBCO bulks [4, 7]. The origin of the reduction in κ_{ab} was interpreted as follows. With larger ionic radius of the RE, the substitution between the RE^{3+} and Ba^{2+} ions tends to occur more easily [8]. This substitution gives rise to the formation of a $RE_{1-x}Ba_{2+x}Cu_3O_y$-type solid solution, which works as a phonon scattering centre. As a result, the thermal conductivity is strongly suppressed. However, this scenario cannot be applicable to the results observed here. The ionic radii of Er^{3+} and Ho^{3+} are 0.1004 nm and 0.1015 nm, respectively, which are smaller than that of Y^{3+} (0.1019 nm); thus we conclude that the substitution-induced reduction of κ does not occur in both ErBCO and HoBCO bulks. Consequently, we can point out only that the REBCO bulk containing heavy RE tends to show a small magnitude of the thermal conductivity, which might come from the low phonon frequency due to the heavy mass of RE.

Let us discuss the change in the $\kappa_{ab}(T)$ slope owing to the different T_{an} in the normal state for HoBCO bulks. Note that the slope change is negligibly small for ErBCO bulk, so we do not discuss it here. In general, the thermal conductivity in a conducting solid is composed of the phonon and charge contributions, κ^{ph} and κ^{ch}. The latter contribution can be estimated from the resistivity data using the Wiedemann–Franz law, $\kappa_{ab}^{ch} = LT/\rho_{ab}$, where L is the Lorenz number. The obtained κ_{ab}^{ch} values are constant values of roughly 6 and 4 mW cm^{-1} K^{-1} above 90 K for T_{an} = 400 °C and 500 °C samples, respectively. For REBCO bulk, the oxygen deficiency, δ, in the RE123 phase, which increases with increasing T_{an}, is related to the amount of charge carrier and acts as a point defect itself at the same time. Therefore the slope change due to the different T_{an} is not brought about by the charge contribution but the phonon scattering due to the point defects. Similar behaviour was observed in Y123 polycrystals [13].

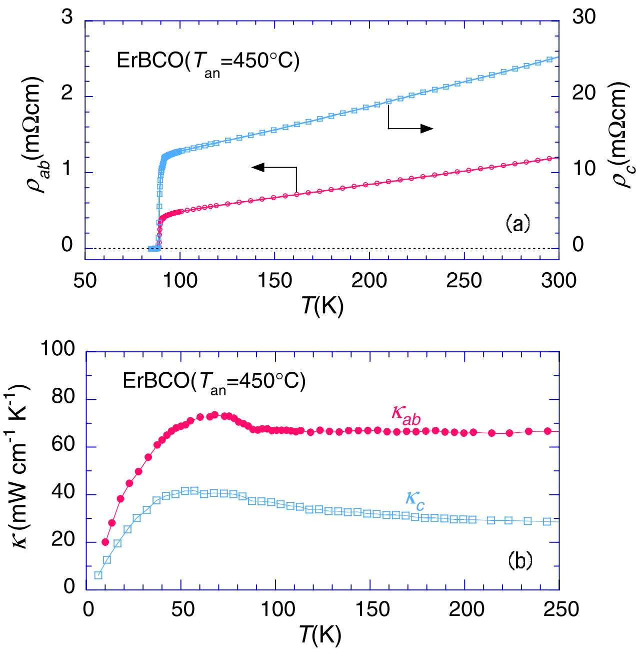

Figure 3. (a) The temperature dependence of the in-plane and out-of-plane electrical resistivity, $\rho_{ab}(T)$ and $\rho_c(T)$, respectively, for Er–Ba–Cu–O (ErBCO) bulks with T_{an} = 450 °C. (b) The temperature dependence of the in-plane and out-of-plane thermal conductivity, $\kappa_{ab}(T)$ and $\kappa_c(T)$, respectively, for the same sample as in the resistivity measurements.

Considering the different temperature dependence of κ_{ab} in the normal state between ErBCO and HoBCO bulks, the phonon scattering mechanism must differ considerably. Since κ^{ph} is generally described by the T^{-1}-dependence in the case that the phonon–phonon Umklapp process dominates the heat transport, the deviation from such behaviour for both bulks indicates the existence of a powerful phonon scattering centre in addition to the Umklapp process and point defects. To get a deeper understanding of the possible phonon scattering mechanism, we would need to measure the thermal diffusivity, α, and sound velocity, v, which would give the phonon mean free path ℓ_{ph} from the relation $\alpha = \frac{1}{3}v\ell_{ph}$.

3.2. Anisotropic thermal conductivity of Er–Ba–Cu–O bulks

Figure 3(a) shows the temperature dependence of the electrical resistivity in the ab-plane, $\rho_{ab}(T)$, and along the c-axis, $\rho_c(T)$, for ErBCO bulks annealed at T_{an} = 450 °C. The anisotropy parameter, γ, which is generally defined as the ratio of the in-plane and out-of-plane coherence length, is estimated to be of about 5.3 from $\sqrt{\rho_c/\rho_{ab}}$ at 100 K.

Figure 3(b) shows the temperature dependence of the thermal conductivity in the ab-plane, $\kappa_{ab}(T)$, and along the c-axis, $\kappa_c(T)$, for the same sample as in the resistivity measurements. The behaviours of $\kappa_{ab}(T)$ are quite similar to those demonstrated in figure 2. On the other hand, κ_c increases monotonically with decreasing temperature from 250 to around 50 K, with a rather broad peak at around 50 K, and decreases toward zero. The anisotropy of the thermal conductivity, γ_k (=κ_{ab}/κ_c), is estimated to be about 2 at around 100 K, which is somewhat smaller than γ = 5.3 described above or $\gamma_k = 5.7$ estimated from the data for Y123 single crystals [14]. Considering the reported values of $\gamma_k = 3$–5 at around 100 K for other REBCO bulks [3, 4], γ_k for ErBCO bulk is not so anomalous. As shown in our previous report [5], κ due to the Y211 phase is larger than κ_c of the Y123 phase. Moreover, the temperature dependence and absolute values of κ_c for ErBCO bulk are very similar to those for YBCO as shown in the inset of figure 2, although the former $\kappa_{ab}(T)$ is half of the latter. Therefore, the relatively small γ_k for ErBCO does not come from the content and/or distribution of the Er211 part, but the small magnitude of $\kappa_{ab}(T)$. The results obtained indicate that we have to use values of κ_{ab} and κ_c that have actually been measured for designing a practical application.

4. Summary

We have measured the temperature dependence of the in-plane thermal conductivity, $\kappa_{ab}(T)$, for both Er–Ba–Cu–O and Ho–Ba–Cu–O superconducting bulks as a function of the annealing temperature. To our knowledge, this is the first report of results for both bulks. We found that the absolute values of $\kappa_{ab}(T)$ of both bulks are larger than that of Dy–Ba–Cu–O bulk, which is well known to show an anomalous small κ_{ab}, and that the broad peak below T_c is also more visible. However their $\kappa_{ab}(T)$ values are still rather smaller than that of Y–Ba–Cu–O bulk. The behaviours of $\kappa_{ab}(T)$ in the normal state deviate from the T^{-1}-dependence for both bulks, which indicates the existence of powerful phonon scattering centres in addition to the crystal defects, unlike Y–Ba–Cu–O. As a result, both Er–Ba–Cu–O and Ho–Ba–Cu–O bulks show the relatively small magnitude of κ_{ab}. To understand this, we pointed out that only a RE–Ba–Cu–O bulk containing a heavy RE (rare earth elements) tends to show small κ_{ab}. The *c*-axis thermal conductivity was also measured for Er–Ba–Cu–O bulk, and the anisotropy of the thermal conductivity, (κ_{ab}/κ_c), was estimated to be about 2.

Acknowledgments

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