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Thermal conductivity of $REBa_2Cu_3O_7$ (RE = Y,Dy,Sm,Nd) superconducting oxides

M. Ikebe *, H. Fujishiro, K. Nakasato, K. Noto

Department of Materials Science and Technology, Faculty of Engineering, Iwate University, 4-3-5 Ueda, Morioka 020, Japan

Abstract

The thermal conductivity κ was measured for 90 K phase oxide RE123 (RE = Y,Dy,Sm,Nd) sintered samples. The shift of the κ maximum toward lower temperatures in Nd123 and Sm123 was observed in comparison with Y123 and Dy123. Based on the dominant phonon thermal carrier model, the shift of the $\kappa(T)$ maximum is explained as due to enhanced phonon scattering by point defects caused by the migration of Nd and Sm ions into the Ba sites of RE123 compounds.

1. Introduction

Thermal conduction is an important and fundamental transport phenomenon of solids. In dielectrics where electrical conduction is absent, the thermal conductivity κ offers valuable information concerning the state of the crystalline lattice through the phonon scattering. In metals and alloys, circumstances are a little bit complicated because the heat conduction is due to both electron (κ_e) and phonon (κ_{ph}) contributions. If the separation of κ_e and κ_{ph} can be suitably made, the information contained in the thermal conductivity should surpass, in principle, that contained in the electrical conductivity. In superconductors where the DC electrical conductivity diverges to infinity below T_c, the thermal conduction is almost a unique measure to study the transport properties. With regard to the thermal conduction of high-T_c superconductors, one recent problem in dispute is concerned with the origin of the observed characteristic enhancement below T_c: phonons or electrical carriers? In this paper we study the thermal conductivity of $REBa_2Cu_3O_7$ (RE123) superconductors with rare earth ions RE = Y, Dy, Sm and Nd. Recently, it was reported that the transition temperature T_c and the critical current J_c are enhanced in $NdBa_2Cu_3O_7$ (Nd123) and $SmBa_2Cu_3O_7$ (Sm123) in comparison with $YBa_2Cu_3O_7$ (Y123) [1]. The possibility of migration into a Ba site was suggested for Nd and Sm ions which have a relatively large ion radius. Nd and Sm ions on Ba sites might act as vortex pinning centers in RE123 compounds. We expected to get information on both the lattice disorder of the specimen and the phonon scattering by quasi-particles in this RE123 system.

2. Experimental procedure

$REBa_2Cu_3O_7$ samples were prepared from stoichiometric mixtures of $(RE)_2O_3$, $BaCO_3$ and CuO raw powders. The mixtures were calcined at 910°C for 24 h in air. They were pressed into pellets and sintered at between 945°C and 965°C for 30 h in

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* Corresponding author. Fax: +81 196 21 6373.

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flowing oxygen. The X-ray analyses confirmed that all samples are of single RE123 phase. The thermal conductivity measurement was made by a continuous heat flow method with Au(0.07 at.%Fe)–chromel thermocouples as thermometers. Measuring temperatures were varied between 10–150 K in a GM-cycle He refrigerator [2].

3. Experimental results

Fig. 1 shows the temperature dependence of the electrical resistivity ρ of RE123 compounds. The ρ value of each sample is 0.7–0.8 mΩ cm at room temperature and is almost independent of the RE ions. These values are rather small for sintered materials. If the sintering process is not optimized, these values easily become larger by 3–5 times. The inset in Fig. 1 shows the superconducting transition temperature in an expanded scale. T_c values of Nd123 and Sm123 are slightly higher than those of Y123 and Dy123. This tendency of T_c enhancement was pointed out by Yoo et al. [3,4] for melt-grown crystals.

Fig. 2 shows the thermal conductivity κ as a function of temperature T. We notice the characteristic enhancement below T_c common to all RE123 samples. However, κ shows some peculiar features which depend on RE ions: the maximum of κ below T_c occurs at lower temperatures for Nd and Sm than for Y and Dy. In the following, this shift of the maximum κ is analyzed based on the assumption that the thermal conduction is mainly due to phonons over the entire temperature range of the presented data.

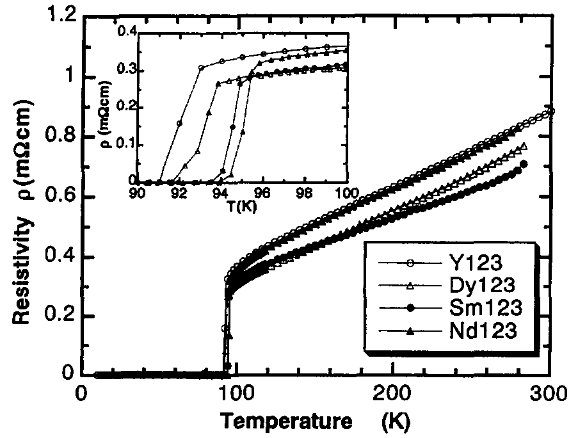

Fig. 1. The electrical resistivity $\rho(T)$ vs. temperature T. The inset shows the superconducting transition in an extended temperature scale. T_c is enhanced in Nd123 and Sm123 specimens.

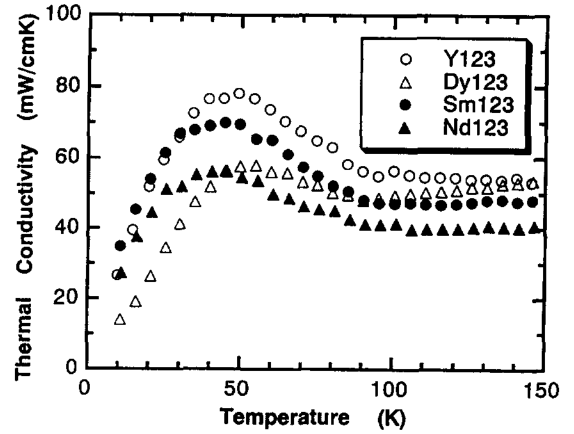

Fig. 2. Thermal conductivity κ as a function of T. The maximum of κ shifts toward lower temperatures in Nd123 and Sm123 and κ of these specimens is smaller in the high-temperature region ($T > T_c$). These facts suggest stronger phonon scattering by point defects in Nd123 and Sm123.

4. Discussion

The heat transport in conductors is due to both electrons and phonons. In the normal state the electron contribution κ_{en} can be estimated by using the Wiedemann-Franz law ($\approx$ 8 mW/cm K for the present RE123). In the superconducting state, we assume that κ_{es} follows the theory proposed by Kadanoff and Martin [5] which predicts the reduction of κ_e below T_c because of the formation of Cooper pairs. Taking account of the phonon scattering by various crystal defects and electrons, the phonon conductivity is given by [6]

$$\kappa_{ph} = \frac{3dnRT^3v^2}{M\,\Theta_D^3}\int_0^{\Theta_D/T}\frac{x^4 e^x}{(e^x-1)^2}\tau_{ph}\,dx, \qquad (1)$$

where d is the mass density, M the mass of 1 mole, $n(=13)$ the number of atoms composing RE123 compounds, R the gas constant, Θ_D the Debye temperature, v the average phonon velocity and x

the reduced phonon frequency. The phonon relaxation time τ_{ph} is given by

$$\frac{1}{\tau_{ph}} = \tau_b^{-1} + sT^2x^2 + pT^4x^4 + ETxg(x, y). \quad (2)$$

Here, τ_b is the phonon relaxation time due to grain boundaries and s, p and E refer to the strength of the phonon scattering by sheet-like faults, point defects and electrons, respectively. The function $g(x, y) = \tau_{phn}/\tau_{phs}$ stands for the ratio of the phonon–electron relaxation time in the normal and superconducting states as given by Bardeen, Rickayzen and Tewordt [7,8]. The values of $g(x, y)$ which depend on the energy gap through the parameter $y = \Delta(T)/k_BT$ become smaller than 1 below T_c in superconducting state because the Cooper pairs do not take part in the phonon scattering.

Fig. 3 shows examples of the fitting curves for the phonon thermal conductivity κ_{ph} $(= \kappa - \kappa_e)$. The parameters used and determined in the fitting process are summarized in Table 1. We note two important points. First, the shift of the maximum κ toward lower temperatures in Nd123 and Sm123 is caused by the enhanced point-defect scattering. If small amounts of Nd and Sm are located at Ba sites as reported [1], then they will work as point defects at Ba sites enhancing the point-defect scattering. The shift of maximum $\kappa(T)$ is consistent with the migration of Nd and Sm ions into Ba sites. Secondly, though the magnitude and the shape of the $\kappa(T)$ curve are somewhat different among RE123 compounds, the electron–phonon scattering term E is nearly constant irrespective of the RE ions. In other words, the electron–phonon coupling constant $\lambda (= 2a\langle t \rangle / \pi v$, where a is the lattice constant, $\langle t \rangle$ the effective hopping matrix element of electrons [6]) is not heavily influenced by RE ions, which seems physically reasonable. Thus, the phonon conduction model naturally explains the observed shift of the maximum κ temperature without introducing any special new assumption. If we try to explain this shift of the κ maximum on the basis of the electrical carrier model [9], we need a reasonable explanation for the change of the quasi-particle scattering rate which depends on RE ions in the superconducting state. The present experimental results seem to support our first assumption that phonons are the dominant thermal carriers over the entire temperature range. It will be interesting to investigate the possible correlation between phonon scattering by point defects and vortex pinning force F_p, because both quantities are possibly related to RE ions on Ba sites.

Table 1
Characteristic parameters of the $REBa_2Cu_3O_7$

Sample	Y123	Dy123	Nd123	Sm123
T_c(K)	91	92	94	94
d(g/cm^3)	5.68	6.12	5.89	6.21
Θ_D (K)	430	430	430	430
τ_b^{-1} (s^{-1})	3.0×10^8	1.6×10^8	3.0×10^8	3.1×10^8
l_b(μm)	11.2	2.1	11.1	10.6
p (K^{-4} s^{-1})	456	487	884	711
s (K^{-2} s^{-1})	2.7×10^6	3.2×10^6	2.7×10^6	2.1×10^6
E (K^{-1} s^{-1})	6.9×10^8	9.9×10^8	7.4×10^8	7.7×10^8
λ	0.26	0.38	0.28	0.29

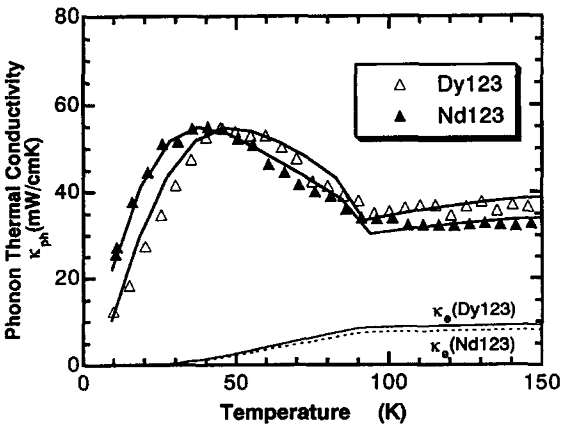

Fig. 3. Examples of the fitting curves of $\kappa(T)$ based on Eq. (1). The electrical component $\kappa_e(T)$ estimated by the Wiedemann–Franz law (in the normal state) and by the theory proposed by Kadanoff and Martin (in the superconducting state) is also shown.

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