Measurement of Out-of-Plane Thermal Conductivity of Epitaxial $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ Thin Films in the Temperature Range from 10 K to 300 K by Photothermal Reflectance

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Abstract We measured the out-of-plane (*c*-axis) thermal conductivity of epitaxially grown $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) thin films (250 nm, 500 nm and 1000 nm) in the temperature range from 10 K to 300 K using the photothermal reflectance technique. The technique enables us to determine the thermal conductivity perpendicular to a thin film on a substrate by curve fitting analysis of the phase lag between the thermoreflectance signal and modulated heating laser beam in the frequency range from 10^2 Hz to 10^6 Hz. The uncertainties of measured thermal conductivity of all samples were estimated to be within $\pm 9\%$ at 300 K, $\pm 12\%$ at 180 K, $\pm 16\%$ at 90 K and $\pm 20\%$ below 50 K. The experimental results show that the thermal conductivity is dependent on the thickness of the thin films across the entire temperature range. We also observed that the thermal conductivity of the present YBCO thin films showed $T^{1.4}$ to $T^{1.6}$ glass-like dependence below 50 K, even though the films are crystalline solids. In order to explain the reason for this temperature dependence, we attempted to analyze our results using phonon relaxation times for possible phonon scattering models, including stacking faults, grain boundary and tunneling states scattering models.

Keywords Low temperature · Photothermal reflectance · Thermal conductivity · Thin film · YBCO

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1 Introduction

Thirty years after the discovery of the first high-temperature copper oxide superconductor, high-temperature superconductor tapes have finally been realized as a result of several breakthroughs in their manufacturing processes and in the analysis of multiscale electromagnetic phenomena [1]. Coated superconductor tapes of an yttrium system ($\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$: YBCO) are already commercially available. In this system, the coated superconductor tape (typically about 10 mm wide and 100 μm thick) essentially consists of a metallic substrate (several tens of micrometers thick) with a thin buffer layer [an approximately one micrometer thick layer of *c*-axis-oriented yttria-stabilized zirconia (YSZ)] on which a YBCO film of several micrometer thickness is epitaxially deposited by pulsed laser deposition. On top of the YBCO film, a metallic layer of several micrometer thickness is deposited as a thermal stabilizer. Now that a coated high-temperature superconductor tape with the desired current performance has been realized, we must improve our understanding of the thermal stability of the system in order to meet the performance requirements for various applications [2,3]. To this end, we must precisely simulate the two-dimensional transient heat conduction in a superconducting tape that can be employed safely under all possible conditions, including low temperature and high magnetic field. Several experimental and analytical studies have attempted to quantitatively analyze the thermal behavior and quench propagation of a coated superconductor tape [4–6]. The thermal conductivity measurement of a coated conductor in the axial direction by the steady-state heat flow method [7] and in the radial (perpendicular to the tape surface) direction [8] are also available. However, no studies have taken into consideration both the anisotropic behavior and the thickness dependence of the thermal conductivity of thin YBCO films across a wide range of temperatures. In particular, in the case of a high-temperature superconducting magnet, the thermal conductivity of the radial direction (*c*-axis in YBCO) of the coil structure is critically important and has been reported to be unexpectedly very low in comparison with the estimated values [8].

Table 1 lists previous experimental studies on the thermal conductivity of YBCO in the *c*-axis direction [9–13,19]. It should be noted here that we have excluded some other studies on thermal conductivity because they measured thermal conductivity in the *ab*-axis direction [14,15] or because they used ceramic samples produced by the solid-state sintering process [16]. Hagen et al. [9] measured the thermal conductivity of YBCO single crystals for the first time in the temperature range of 10 K–330 K. They revealed that the anisotropy ratio of in-plane (*ab*-axis) to out-of-plane (*c*-axis) thermal conductivity was 4–5. However, because their samples were small, as summarized in Table 1, the conventional steady-state method using an experimental setup with two thermocouples and one heater may introduce a factor of two uncertainty in their absolute values, as mentioned by Wand et al. [15]. On the other hand, the samples employed by Fujishiro et al. [11], Gusakov et al. [12] and Shams et al. [13] were large enough (several hundred to several thousand times larger than those of Hagen et al. [9]) to apply the conventional steady- or quasi-steady-state heat flow methods to properly measure the thermal conductivity of the *c*-axis YBCO crystals. As for thin film samples, the literature contains only one report: Shaw-Klein et al. [10] used the thermal comparator technique [17]. In this technique, the effects of substrate and ther-

mal contact resistance between the probe tip and the thin film surface are supposed to be eliminated. However, this method is incapable of measuring the thickness dependence of the thermal conductivity of thin film. It should be noted that Marshall et al. [18] investigated the anisotropic thermal diffusion constants in thin films of YBCO (50 nm–350 nm) on an MgO substrate by surface-selective transient-grating experiments. Even though there are no explicit thermal conductivity values, they found that the *c*-axis thermal diffusion constant was about 10 times smaller than that of the *ab*-plane. In summary, as for previous experimental studies on the out-of-plane thermal conductivity of YBCO, the literature contains only three reports on bulk crystal and essentially no data on the *c*-axis thermal conductivity of thin film samples in a wide temperature range.

In our previous paper [19], we showed that the out-of-plane thermal conductivity of epitaxially grown YBCO thin films exhibited thickness dependence even at room temperature. This result may have been due to the quite broad spectral distribution of phonons responsible for the thermal transport perpendicular to the CuO₂ planes. In the present study, we measured the out-of-plane thermal conductivity λ_c of epitaxial thin films of YBCO with nominal thicknesses of 250 nm, 500 nm, and 1000 nm in a temperature range of 10 K–300 K using the photothermal reflectance technique. Our experiments showed that the thermal conductivity of the present crystalline thin films exhibited $T^{1.4}$ to $T^{1.6}$ glass-like dependence below 50 K. To determine the reason for this temperature dependence, we analyzed our experimental data by using the classical phonon relaxation times for possible phonon scattering models.

2 Sample Preparation and Characterization

Using reactive thermal co-evaporation, YBCO thin films were deposited on a polished (100) MgO single-crystal substrate, 10 mm × 10 mm and 500 μm in thickness (Ceraco Ceramic Coating, GmbH). The thicknesses of the films were 250 nm, 500 nm, and 1000 nm in nominal values as stated by the supplier. Since the uncertainty in sample thickness is one of the major factors contributing to the overall uncertainty in the thermal conductivity measurement technique used, it was important to accurately determine and verify the film thickness. We therefore measured the YBCO film thickness using a three-dimensional laser microscope (VK-9700, spatial resolution of 1.0 nm; Keyence). The thickness of the film was determined by the average of two steps chosen randomly. Table 2 summarizes the results of the film thickness measurements, together with their critical temperature and critical current density reported by the supplier.

The films were deposited with a *c*-axis orientation on the substrate surface. In order to verify the orientation, we investigated the crystalline structure of the films using XRD (Bruker-AXS, D8 Discover; Bruker). Based on the diffraction patterns obtained for all film samples, strong sharp peaks were derived from (001) to (007) from the *c*-axis crystal orientation. On the other hand, polycrystalline peaks such as (102) and (115) were not observed. The *c* lattice parameter calculated from the present diffraction pattern was 11.7 Å, which corresponded with a value of $\delta = 0.07$ in a previous report [20]. By using Raman spectroscopy, particularly the Raman frequency of the

Table 1 List of previous experimental studies on out-of-plane (*c*-axis) thermal conductivity of YBCO

Authors	Year	Sample	T_c (K)	Measurement method	Out-of-plane thermal conductivity at room temperature ($\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$)	Temperature range (K)
Hagen et al. [9]	1989	Single crystal Flux method D : $565 \times 590 \times 220 \mu\text{m}^3$ E : $720 \times 620 \times 320 \mu\text{m}^3$ F : $650 \times 420 \times 110 \mu\text{m}^3$ G : $650 \times 720 \times 220 \mu\text{m}^3$ Underlined: <i>c</i> -axis direction	90	Steady-state method (crystal is sandwiched between two sapphire chips with one corner exposed on both sides thermocouples are attached. Temperature difference is 0.2–0.8 K)	Sample F: ~ 0.9 Sample E: ~ 1.3 Sample D: ~ 1.7 Sample G: ~ 1.8 (taken from graphical presentation)	10–300
Shaw-Klein et al. [10]	1992	Thin film RF Magnetron sputtering film thickness: 250 nm, 500 nm, 750 nm and 1000 nm (MgO single-crystal substrate)	83	Thermal comparator technique (a plot of thickness/ λ_{eff} against thickness yields a slope whose inverse is the film thermal conductivity)	0.26 (no thickness dependence)	Room temperature
Fujishiro et al. [11]	1994	Mixed crystal Modified melt texture growth method $2.8 \times 3.9 \times 12 \text{ mm}^3$ underlined: <i>c</i> -axis direction	90	Steady-state heat flow method	~ 2.2 @ 200 K (taken from graphical presentation)	10–200
Gusakov et al. [12]	2000	Single crystal $3 \times 3 \times 2.4 \text{ mm}^3$	–	Steady-state method	~ 2.1 @ 300 K (taken from graphical presentation)	6–307

Table 1 continued

Authors	Year	Sample	T_c (K)	Measurement method	Out-of-plane thermal conductivity at room temperature ($\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$)	Temperature range (K)
Shams et al. [13]	2001	Single crystal $(7-\delta) = 6.93 \pm 0.02$ Self-flux technique $4 \times 4 \times 0.5 \text{ mm}^3$ Underlined: c -axis direction	92	Quasi-steady-state method	~ 2.2 @ 300 K (taken from graphical presentation)	20–300
Ikeda et al. [19]	2013	Epitaxial thin film Reactive thermal co-evaporation on MgO Film thickness: 250 nm, 500 nm, 1000 nm	88–90	Photothermal radiometry	250 nm: 0.905 500 nm: 1.31 1000 nm: 1.78	300

Table 2 Description of the YBCO thin film samples

Nominal thickness (nm)	Measured thickness (nm)	Critical temperature (K)	Critical current density (MA·cm ⁻²)
250	260.5 ± 2.9 %	87.9	3.3
500	503.5 ± 2.9 %	88.8	3.1
1000	1087 ± 0.3 %	90.2	2.7

The films were deposited using reactive thermal co-evaporation on a (100) MgO single-crystal substrate. The film thickness was accurately determined by a three-dimensional laser microscope. The critical temperatures and critical current densities were the values reported by the supplier

500 cm⁻¹ band, it is possible to evaluate the oxygen-deficient δ in YBa₂Cu₃O_{7- δ} [21]. We therefore obtained the Raman spectra for the YBCO thin film samples using a Raman spectrometer (inVia Raman Microscope, Reinshaw). It was found that the range of critical temperatures determined from the obtained oxygen content of 6.84–6.92 ($\delta = 0.16$ to 0.08) [22] was in sufficient agreement with the critical temperatures listed in Table 2. In conclusion, the present three epitaxially grown YBCO films have almost the same characteristics as the superconductor.

It is known that the reduction of thermal conductivity across or along a thin film occurs by boundary scattering when the mean free path of a phonon is on the order of or larger than the film thickness. Of course, we must also consider the possibility of phonon scattering by grain boundaries. For this purpose, we measured the surface crystal grain sizes on the surface images taken by AFM (SPM9500J3; Shimadzu). The measured grain sizes for each film were as follows: for the 250 nm film, 260 nm–720 nm; 500 nm film, 730 nm–980 nm; and 1000 nm film, 950 nm–1400 nm. Since the present epitaxial YBCO thin films exhibited grain sizes larger than the film thickness, the grain boundaries could be expected to have a very small effect on the phonon scattering in the *c*-axis direction.

3 Principle of Measurement

The principle of the photothermal reflectance technique used to measure the thermal conductivity of the YBCO thin film in the case of a two-layered film on substrate is schematically shown in Fig. 1. The temperature modulation of a sample surface caused by illumination with a heating laser beam leads to a modulation of a sample's optical reflectivity *R*. The relation between the change in reflectance ΔR and that of surface temperature ΔT can be written as follows:

$$\frac{\Delta R}{R_0} = \frac{1}{R_0} \frac{dR}{dT} \Delta T \quad (1)$$

where R_0 is the reflectance at reference temperature and $(1/R_0)dR/dT$ is the coefficient of thermal reflectance of the sample, which ranges from 10⁻⁶ to 10⁻⁴ per degree for most solids. Even though the heat conduction equation describes dissipative diffusion phenomena, the temperature response of a material with forced periodic heating

Fig. 1 Principle of photothermal reflectance to measure the out-of-plane thermal conductivity of a thin film (two-layered model)

can be represented by a strongly damped oscillation having wave-like properties. Note that phrases such as “temperature wave,” “thermal wave” or “AC temperature rise” are frequently used when applying photothermal techniques to determine the thermal properties of a given material, since they are easily comprehended [23]. The temperature change (amplitude of the AC temperature rise) at the sample surface in the coordinate system described in Fig. 1 can be expressed by the following heat conduction equations with respect to the complex temperature Φ at angular frequency ω [24]:

$$\frac{\partial^2 \Phi_g}{\partial x^2} = \frac{1}{a_g} \frac{\partial \Phi_g}{\partial t} \quad (0 \leq x), \quad (2)$$

$$\frac{\partial^2 \Phi_1}{\partial x^2} = \frac{1}{a_1} \frac{\partial \Phi_1}{\partial t} - \frac{\beta I_0}{2\lambda_1} (1 + \exp(j\omega t)) \exp(\beta x) \quad (-l_1 \leq x \leq 0), \quad (3)$$

$$\frac{\partial^2 \Phi_2}{\partial x^2} = \frac{1}{a_2} \frac{\partial \Phi_2}{\partial t} \quad (-l_1 - l_2 \leq x \leq l_1), \quad (4)$$

$$\frac{\partial^2 \Phi_b}{\partial x^2} = \frac{1}{a_b} \frac{\partial \Phi_b}{\partial t} \quad (x \leq -l_1 - l_2), \quad (5)$$

where a , λ , I_0 , β , l and j are the thermal diffusivity, the thermal conductivity, the intensity of heating laser light, the absorption coefficient of the sample, the thickness of a layer and the imaginary unit, respectively. The subscripts, g , b and i denote the gas, the backing and each layer of the sample, respectively. By solving the above heat conduction equations with respect to AC temperature variation on the basis of appropriate boundary conditions, the amplitude of the complex temperature θ at the surface of the sample ($x = 0$) is obtained. The amplitude of the complex temperature for a two-layered model θ is given by [24]:

$$\theta = \frac{\beta I_0}{2\lambda_1 (\beta^2 - \sigma_1^2)} [(\gamma - 1)(B_2 + 1) \exp(\sigma_1 l_1) - (\gamma + 1)(B_2 - 1) \exp(-\sigma_1 l_1) + 2(B_2 - \gamma) \exp(-\beta_1 l_1)] \times [(g + 1)(B_2 + 1) \exp(\sigma_1 l_1) - (g - 1)(B_2 - 1) \exp(-\sigma_1 l_1)]^{-1}, \quad (6)$$

$$B_2 = b_{12} \times \frac{b_{12}N + b_{1b}M}{b_{12}M + b_{1b}N}, \quad (7)$$

$$M = \exp(\sigma_2 l_2) + \exp(-\sigma_2 l_2), \quad (8)$$

$$N = \exp(\sigma_2 l_2) - \exp(-\sigma_2 l_2), \quad (9)$$

where $\sigma_i = (1 + j)k_i$ denotes the complex wave number of the temperature wave in the i th layer, $k_i = [\omega/(2a_i)]^{1/2}$ is the wave number in the i th layer, $\gamma = \beta_1/\sigma_1$, $g = e_g/e_1$, $b_{ij} = e_j/e_i$, $e_i = (\lambda_i \rho_i C_i)^{1/2}$ is the thermal effusivity of the i th layer, ρ_i is the density of the i th layer, and C_i is the specific heat of the i th layer. The relation between the complex temperature variation at the sample surface Φ_{AC} and θ can be written as

$$\Phi_{AC} = \Phi_g(x = 0) = \theta \exp(j\omega t). \quad (10)$$

The detected variation in temperature ΔT_{AC} is given by the real part of the complex temperature amplitude,

$$\Delta T_{AC} = \text{Re}[\theta \exp(j\omega t)] = q \cos(\omega t - \Delta\phi), \quad (11)$$

where

$$q = |\theta| = \left\{ [\text{Re}(\theta)]^2 + [\text{Im}(\theta)]^2 \right\}^{1/2}, \quad (12)$$

$$\Delta\phi = \arg(\theta) = \tan^{-1} \left[-\frac{\text{Im}(\theta)}{\text{Re}(\theta)} \right]. \quad (13)$$

Assuming l_1 , l_2 and β are measured in advance, we are able to determine the thermal conductivity by a curve fitting analysis of phase lag data in the frequency domain.

4 Experimental Apparatus

To measure the out-of-plane thermal conductivity, we used the photothermal reflectance technique in the temperature range of 10 K–300 K, as shown in Fig. 2. This technique essentially involves a frequency-modulated heating laser beam (Omicron, LDM445.500.TA.M; 445 nm, 500 mW and $1/e^2$ beam diameter of 2.53 mm with ellipticity of 1.07) and a probing laser beam (Omicron, LDM636.09.CWA.L.US; 636 nm, 9 mW and $1/e^2$ beam diameter of 1.25 mm with ellipticity of 1.05) to detect the induced thermorefectance signal from a reflective sample surface, and a lock-in amplifier (Signal Recovery, Model 7280) for the phase lag measurement between the detected thermorefectance signal and the modulated heating laser beam. The balanced detector is a Si/PIN photodiode (New Focus, Model 2107-FS) with a detector active area of 1.0 mm $\times$ 0.8 mm and a spectrally sensitive range of 300 nm–1070 nm. As described

Fig. 2 Experimental apparatus to measure the out-of-plane thermal conductivity of a thin film at low temperatures based on the photothermal reflectance technique

in the previous section, we were able to determine the thermal conductivity perpendicular to the sample film surface by a curve fitting analysis of the phase lag data in the appropriate frequency range from 10^2 Hz to 10^6 Hz so that the assumption of one-dimensional heat conduction at the largest thermal diffusion length (at the lowest frequency) under a given measurement condition is fulfilled. The two laser beams irradiated the sample through quartz optical windows of a cryogen-free cryostat (Oxford Instruments, Cryofree Spectromag CFSM7T-4), providing sample temperatures from 1.5 K to 300 K with a magnetic field up to 7 T in the horizontal direction. The temperature was measured using a CernoxTM thin-film resistance thermometer attached to a sample holder within an uncertainty of ± 40 mK. The entire measurement system is arranged on two optical benches with different height in order to avoid the effect of a leak magnetic field from the cryostat on the lasers and optical components such as the Faraday isolators and the balanced detector. The principle and working equations for the experimental technique were essentially consistent with those of photothermal radiometry [19, 24], for which we also discussed the uncertainty assessment of the measured thermal conductivity by using this technique.

In order to gain sufficiently strong thermoreflectance signals, the YBCO sample surfaces were coated with 159 ± 9 nm sputtering films of platinum as a reflecting layer against the probing laser light. Consequently, we adopted the two-layered heat conduction model (platinum + YBCO + MgO substrate) in a He gas atmosphere for the extraction of YBCO thermal conductivity from the phase lag versus frequency data. In this calculation, we used the thermal conductivity and diffusivity of bulk platinum, because we found that the effect on the measured phase lag due to the size effect on the thermophysical properties of platinum [25] was negligibly small. It was also assumed that the thermal contact resistance between the platinum and YBCO layer was less than 10^{-8} m² K/W, making the effect on the measured phase lag small enough to be ignored under all measurement conditions. The ΔT_{AC} [Eq. (11)] caused by the

irradiation of the frequency-modulated heating laser beam must be kept as small as possible in order to minimize systematic uncertainty owing to the temperature at which the measured thermal conductivity is allocated. This effect becomes significant in the lower temperature region where the temperature dependence of thermal conductivity increases, and at the same time the specific heat is significantly reduced. For the present measurement, we adjusted the heating laser power to maintain the temperature amplitude within 2 K.

5 Results and Discussion

5.1 Temperature and Thickness Dependencies of the Thermal Conductivity of YBCO Thin Films

Table 3 contains the experimental results for the out-of-plane thermal conductivity λ_c of epitaxial thin films of YBCO. In this table, we display one extra digit in order to avoid dropping digits that may be significant for future use. Measurements were repeated at the same temperature two to four times in order to check reproducibility. We carefully evaluated all the obtained phase lag versus frequency curves to determine the thermal conductivity. Our criterion was that the standard deviation between measured phase lag and fitting curve should be within one degree. For these thermal conductivity results, we evaluated the uncertainties based on the Guide to the Expression of Uncertainty in Measurement (GUM) [26]. Since the working equations of thermal conductivity cannot be expressed explicitly in the measurement principle [19], the uncertainty associated with each measurement component was numerically calculated. This was accomplished by taking the following sources of uncertainty into account: the platinum reflecting layer component (thermal conductivity, thermal diffusivity, and thickness); YBCO sample layer component (specific heat and thickness); thermal conductivity of MgO; heating beam radius; measured phase lag; and temperature. The relative combined standard uncertainties of the thermal conductivity of all samples were estimated to be within $\pm 9\%$ at 300 K, within $\pm 12\%$ at 180 K, within $\pm 16\%$ at 90 K and within $\pm 20\%$ below 50 K.

Figure 3 shows the experimental results for the out-of-plane thermal conductivity of *c*-axis-aligned epitaxially grown thin films λ_c of YBCO with nominal thicknesses of 250 nm, 500 nm and 1000 nm together with the empirical correlations, which will be described later in this section. The films exhibited thickness dependence not only near room temperature [19] but in the entire temperature range from 10 K to 300 K. The absolute values of thermal conductivity in the normal state for thin films are less than half of those for bulk single-crystal values at the smallest thickness. In the superconducting state, on the other hand, the reduction ratio (size effect) reaches less than one-tenth at the maximum. Concerning the peak thermal conductivity, λ_c for thin films increases gently as the temperature decreases from 300 K, exhibiting a broad maximum around 100 K–150 K and then decreasing sharply at lower temperatures. This maximum shifts toward higher temperatures as the film thickness is decreased. As can be seen from our experimental results, the out-of-plane thermal conductivity—unlike the *ab*-plane bulk thermal conductivity associated with the superconducting

Table 3 Experimental results of out-of-plane thermal conductivity of YBCO thin films in the temperature range from 10 K to 300 K measured by the photothermal reflectance technique

Temperature (K)	Thermal conductivity ($\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$)		
	Nominal thickness 250 (nm)	Nominal thickness 500 (nm)	Nominal thickness 1000 (nm)
10	0.036	0.083	0.168
20	0.139	0.265	0.431
30	0.258	0.476	0.679
40	0.427	0.665	1.108
50	0.628	0.931	1.442
60	0.781	1.171	1.779
70	0.954	1.401	1.946
80	1.081	1.461	1.939
90	1.044	1.413	2.016
100	1.135	1.471	1.960
110	1.110	1.547	2.010
120	1.124	1.544	1.970
150	1.143	1.534	1.993
180	1.134	1.556	1.911
210	1.080	1.515	1.813
240	1.055	1.399	1.769
270	1.034	1.333	1.725
300	1.044	1.372	1.656

Fig. 3 Out-of-plane thermal conductivity of YBCO thin films with empirical correlation using the best-fit parameters in Table 4

Table 4 Best-fit coefficients a_i and b_i in Eqs. (15)–(18)

	$i = 1$	$i = 2$	$i = 3$	$i = 4$
a_i	−0.018831	0.975	160.74	1.4205
b_i	−2.6802E−5	1.1703E−3	−3.1266E−2	1.5849E−4

transition—did not show any considerable peaks, because the heat conduction in the c -axis is almost governed by phonons not by electrons.

The experimental results were correlated empirically as a function of temperature and film thickness. We employed the following log-normal functional form for the correlation:

$$\lambda_c(T, l) = \lambda_0(l) + A_0(l) \exp \left[- \left\{ \frac{\ln(T/T_0(l))}{w(l)} \right\}^2 \right] \quad (14)$$

$$\lambda_0(l) = a_1 + b_1 l \quad (15)$$

$$A_0(l) = a_2 + b_2 l \quad (16)$$

$$T_0(l) = a_3 + b_3 l \quad (17)$$

$$w(l) = a_4 + b_4 l \quad (18)$$

where λ is the thermal conductivity in $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ and l is the thickness of the film in nm. The best-fit values of coefficients a_i and b_i are given in Table 4. Equations (14)–(18) reproduce the entire body of the experimental results (Table 3) within a standard deviation of $0.069 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$.

In order to compare the temperature dependence of the present results with that of other typical phonon heat conduction materials, Fig. 4 shows the present experimental results with corresponding bulk single-crystal ($3 \times 3 \times 2.4 \text{ mm}^3$) values measured by Gusakov et al. [13] compared to those of bulk crystalline Si and corresponding thin films (250 nm, 500 nm and 1000 nm in thickness) calculated by the models and parameters of Yang and Dames [27], together with the thermal conductivity of fused glass SiO_2 [28]. All of the heat is carried by phonons in the materials displayed in Fig. 4. In terms of thickness dependence, both YBCO and Si showed essentially the same tendency across the entire temperature range. If we compare the absolute values of thermal conductivity for the two crystalline materials, the λ_c of YBCO thin film was two to three orders of magnitude smaller than that of Si. Heat conduction across the CuO_2 plane in YBCO was very poor, and the thermal conductivity was as low as that of non-crystalline SiO_2 near room temperature. If we look closer at the temperature dependences in the low-temperature region, the thermal conductivity of Si thin films reflects the T^3 behavior of the specific heat, as predicted by theory, whereas non-crystalline SiO_2 shows a characteristic plateau with $\sim T^{1.8}$ dependence at lower temperatures due to structural and orientational disorder in the system. In contrast, the thermal conductivity of the present crystalline thin films of YBCO varied as $T^{1.4} \sim T^{1.6}$, resembling the non-crystalline behavior except for the lack of a plateau at higher temperature. In other words, the thermal transport in YBCO thin films perpendicular

Fig. 4 Thermal conductivity results of out-of-plane YBCO thin films and of crystalline Si and of non-crystalline SiO₂. Bulk single-crystal YBCO values are taken from Gusakov et al. [13]. The thermal conductivity of bulk crystalline Si and corresponding thin films were calculated using the models and parameters of Yang and Dames [27]. The dotted line shows the temperature dependence of the thermal conductivity of fused glass SiO₂ from Ref. [28]

to the surface was analogous to that in an amorphous system, even though they are crystalline solids.

In the next section, in order to determine the reason for this temperature dependence, we attempted to analyze our results using phonon relaxation times in order to identify possible phonon scattering models including stacking faults, grain boundary and tunneling states scattering.

5.2 Interpretation of Experimental Data Using Phonon Relaxation Times

It is well known that the total thermal conductivity of a high- T_c superconductor λ can be described as the sum of an electronic contribution λ^e and a phonon contribution λ^p as follows [29]:

$$\lambda = \lambda^e + \lambda^p. \quad (19)$$

In the case of the out-of-plane thermal conductivity (λ_c) of a single crystal of YBCO, the electronic contribution λ_c^e can be estimated by the Wiedemann–Franz–Lorenz law.

$$\frac{\lambda_c^e}{\sigma_c T} = \frac{\pi^2}{3} \left(\frac{k_B}{e} \right)^2 = 2.44 \times 10^{-8} \quad (\text{W} \cdot \Omega \cdot \text{K}^{-2}). \quad (20)$$

If we used the experimental results of out-of-plane electrical conductivity, σ_c , at 290 K [30], λ_c^e was calculated to be less than 2 % of the measured thermal conductivity for all thin film samples at 300 K. Therefore, it was a reasonable approximation that $\lambda_c \simeq \lambda_c^p$ for the present thin film samples.

Although, in principle, it is possible to obtain precise calculations of phonon thermal conductivity using molecular dynamics for Si [31], or through an approach based on the phonon Boltzmann equation for a test system of Lennard–Jones Ar [32], in the case of anisotropic and rather complex crystalline materials such as YBCO, these recently available first-principle methods are not applicable, due mainly to the difficulty of determining appropriate intermolecular potentials, and also because the number of atoms that must be taken into consideration in the simulations may become prohibitively large for sub-micrometer-order thicknesses with phonon MFP on the order of several tens of nanometers. On the other hand, for nonmetallic crystalline materials, theoretical frameworks for understanding certain aspects of phonon thermal conductivity at low temperatures, even in the case of complex polyatomic molecular systems, were essentially solved [33] by Klemens [34,35] and Callaway [36,37], who expanded the very early work of Debye and Peierls. In the present paper, we employed the Klemens–Callaway model [33,37] as appropriate for use in the analysis of our experimental results for the out-of-plane thermal conductivity of YBCO thin films. Our analysis incorporated the following assumptions. (1) The complexity arising from anisotropy did not affect the principles of the model [35]. (2) The sine-type phonon dispersion relation (Born–von Karman) was used for the acoustic modes while neglecting the optical modes [38]. (3) It was assumed that the total resistive phonon scattering rate can be described by the sum of the reciprocal relaxation times due to boundary scattering, point defects scattering caused by oxygen vacancies and phonon–phonon umklapp scattering.

Many previous studies that sought to calculate (or fit) the phonon thermal conductivity of YBCO in the wide range of temperature worked basically within the Debye framework of lattice dynamics [39–43]. In those cases, by neglecting the polarization dependence (allowing the phonon dispersion relation to be $\omega = v_s k$ for all three acoustic branches), the phonon thermal conductivity was given by Ref. [44] as follows:

$$\lambda_p(T) = \frac{k_B}{2\pi^2 v_s} \left(\frac{k_B}{\hbar} \right)^3 T^3 \int_0^{\Theta_D/T} \tau(x) \frac{x^4 e^x}{(e^x - 1)^2} dx. \quad (21)$$

Here, $x \equiv \hbar\omega/k_B T$ is the reduced phonon energy, τ is the total phonon relaxation time, Θ_D is the Debye temperature and v_s is the constant speed of sound. The phonon dispersion curves in YBCO were experimentally investigated by inelastic neutron scattering [45] and were also calculated based on the distributed-multipole model [46]. There were essentially 39 vibrational modes (phonon branches) because the unit cell of YBCO contains 13 constituent atoms. Although there are many phonon branches in the case of the complex crystal structure of YBCO, it is acceptable to consider the dispersion relation for only the three acoustic branches that pass through the origin of

the $\omega - k$ coordinate, because the optical phonons typically contribute little to the heat flux due to their low group velocity and high scattering rate. High-frequency phonons are usually scattered more significantly than low-frequency phonons [47]. Moreover, at the low temperatures we are interested in here, the optical modes will not be excited. As Dames and Chen pointed out [38], the sine-type phonon dispersion relation (Born–von Karman) [48] improves the rather poor approximation of Debye dispersion at higher frequencies. We used the following sine-type dispersion equation.

$$\omega = \omega_0 \sin\left(\frac{\pi}{2} \frac{k}{k_0}\right) \quad (22)$$

k_0 and ω_0 were determined using the following equations [47].

$$k_0 = (6\pi^2 N)^{1/3} \quad (23)$$

$$\omega_0 = \frac{2}{\pi} v_s (6\pi^2 N)^{1/3} \quad (24)$$

Here, N is the number of primitive cells per unit volume,

$$N = \frac{\rho N_A}{M} . \quad (25)$$

N_A is the Avogadro constant. For the material-dependent parameters of YBCO, we used $\rho = 6.37 \text{ g/cm}^3$ [49], $M = 666.194 \text{ g/mol}$, and $v_s = 4130 \text{ m/s}$ [49]. By substituting Eq. (22) into Eq. (21), thermal conductivity λ_c can be calculated by the following equation.

$$\lambda_c = \frac{\hbar^2}{2\pi^2 k_B T^2} \int_0^{\omega_0} \frac{\omega^2 e^{\hbar\omega/k_B T}}{(e^{\hbar\omega/k_B T} - 1)^2} \left[\frac{2k_0}{\pi} \sin^{-1}\left(\frac{\omega}{\omega_0}\right) \right]^2 v_g(\omega) \tau(\omega) d\omega \quad (26)$$

$$v_g(\omega) = \frac{\pi}{2k_0} \omega_0 \cos\left(\frac{\pi}{2} \frac{k}{k_0}\right) \quad (27)$$

The total relaxation time $\tau(\omega)$ due to multiple phonon scattering processes is expressed by Matthiessen's rule.

$$\frac{1}{\tau(\omega)} = \frac{1}{\tau_b} + \frac{1}{\tau_{pd}(\omega)} + \frac{1}{\tau_u(\omega)} + \dots . \quad (28)$$

We first considered the following phonon scattering mechanisms related to the out-of-plane thermal conductivity of YBCO.

5.2.1 Boundary Scattering

Boundary scattering plays an important role in limiting the MFP of small-wave number phonons at low temperatures, particularly in the case of thin films. The relaxation time

caused by the boundary scattering τ_b can be represented by a frequency-independent constant [36]:

$$\frac{1}{\tau_b} = \frac{v_g}{l}, \quad (29)$$

where v_g is the phonon group velocity and l is the thickness of the thin film (in the present calculation, 250 nm, 500 nm and 1000 nm).

5.2.2 Point Defects by Oxygen Vacancies

Here, we consider point defects due to oxygen vacancies among the phonon scattering by static imperfections. For scattering by point defects, Klemens [35] and Callaway [36] derived the following expression for relaxation time caused by the point defects τ_{pd} :

$$\frac{1}{\tau_{pd}} = A\omega^4 \quad (30)$$

and

$$A = \frac{V_0}{4\pi v_s^3} c \left(\frac{\Delta M}{M} \right)^2. \quad (31)$$

Here, V_0 is the volume of the unit cell, v_s is the velocity of sound and c is the concentration per atom for substitutional atoms with an atomic mass of $M + \Delta M$. For vacancies, the effective value of $\Delta M/M$ can be given as [50,51]

$$\Delta M/M = -M_a/M - 2, \quad (32)$$

where M is the average mass per atom and M_a is the mass of the missing atom. In the present YBCO samples, the concentration of vacancies can be calculated from the correlation between T_c and oxygen concentration in near-stoichiometric YBCO single crystals [22]. By employing the average oxygen concentration for these YBCO samples, and the sound velocity (4130 m/s) [49] and lattice constants ($a = 0.382$ nm, $b = 0.389$ nm, $c = 1.17$ nm) [52] of YBCO, we have $A = 5.44 \times 10^{-42} \text{ s}^3$, which was assumed to be independent of film thickness. It may be important to note that we also calculated A_{isotope} , which means the point defects due to naturally occurring isotopes given by Slack [53]. We found that $A_{\text{isotope}} = 2.86 \times 10^{-44} \text{ s}^3$ for the present YBCO samples, which was assessed as negligible in comparison with the effect of oxygen vacancies.

5.2.3 Phonon–Phonon Umklapp Scattering

At high temperatures, phonon–phonon umklapp processes are effectively the only cause of thermal resistance. Although several types of equations to describe the phonon–phonon umklapp scattering relaxation time have been proposed [27,54–56], these equations are essentially the same in expressing the $1/T$ behavior of thermal conductivity at relatively high temperatures. Here, we selected the following expression for the phonon–phonon umklapp scattering relaxation time τ_u , with just one adjustable

parameter given by Slack and Galginitis [55]:

$$\frac{1}{\tau_u} = B\omega^2 T e^{-\Theta_D/3T}, \quad (33)$$

where B is the adjustable parameter and $\Theta_D = 400$ K [49].

We first attempted to fit our experimental data for the out-of-plane thermal conductivity of YBCO thin films using the three-phonon scattering processes described above with just the single adjustable parameter B in Eq. (33). However, it was essentially not possible to reproduce the temperature dependence of the thermal conductivity results using any of the much larger values that we substituted for the adjustable parameter B or the point defects parameter A . Figure 5 compares one of the typical ill-fitting curves for the experimental data of 1000 nm thick films derived using “ $b + \text{pd} + u$ ” phonon scattering mechanisms with other fitting results described later. The major difficulty arose from the present rather gentle temperature dependences proportional to $T^{1.4 \sim 1.6}$ at lower temperatures. Consequently, we could not simply ascribe the temperature dependence in λ_c of YBCO thin films to three principal phonon scattering processes. We therefore needed to find another appropriate phonon scattering mechanism to describe the experimentally observed temperature dependence of the thermal conductivity that might be intrinsic to the cuprate superconductor.

5.2.4 Stacking Faults Scattering

If we take into account the orthorhombic crystal structure of YBCO with CuO_2 planes, stacking faults phonon scattering should be considered along with the three major phonon scattering mechanisms described thus far. In fact, Tewordt and Wölkhausen [39] considered the relaxation time for scattering of phonons by the strain field of sheet-like faults to analyze the in-plane thermal conductivity of bulk YBCO. As a fitting parameter, this term seems to be very insignificant (weak sensitivity to thermal conductivity). Morelli et al. [57] measured the thermal conductivity of a single crystal of $\text{La}_{1.96}\text{Sr}_{0.04}\text{CuO}_4$ in both the parallel and perpendicular directions to the CuO_2 planes. They fitted their thermal conductivity results by adding a stacking faults term, which was proportional to ω^2 , and concluded that the principal reason for the large anisotropy in the thermal conductivity was the extremely strong stacking faults spacing on the order of 10 nm. We consequently tried to analyze our experimental results by adding the relaxation time τ_{sf} [57]:

$$\frac{1}{\tau_{\text{sf}}} = C\omega^2. \quad (34)$$

Here, C is a fitting parameter. This addition of stacking faults did not work well for our thin film results, as shown under “ $b + \text{pd} + u + \text{sf}$ ” in Fig. 5. Moreover, the high density of stacking faults calculated by Morelli et al. [57] was physically unlikely to exist in the case of our epitaxially grown crystalline thin films of YBCO, as evidenced by the XRD analysis.

Fig. 5 Comparison between measured (*plots*) and fitted (*curves*) thermal conductivity for a 1000 nm thick YBCO film. The curves are fits using boundary scattering + point defects + umklapp scattering ($b + pd + u$) and an additional scattering mechanism of stacking faults ($b + pd + u + sf$), grain boundary ($b + pd + u + gb$) and tunneling states ($b + pd + u + ts$) together with the empirical correlation of Eq. (14)

5.2.5 Grain Boundary Scattering

The thermal conductivity reduction due to grain boundary scattering is generally interpreted to be interpreted using a scattering length equal to the grain size independent of the phonon frequency. Wang et al. [58] showed that the frequency-dependent MFP model for grain boundary scattering works well to explain the T^2 dependence of thermal conductivity of nanocrystalline silicon at low temperature. The frequency-dependent relaxation time τ_{gb} can be written as

$$\frac{1}{\tau_{gb}} = \frac{v_g}{D} \frac{\omega}{\omega_0}, \quad (35)$$

where v_g is the phonon group velocity of Eq. (27), ω_0 can be calculated by Eq. (24) and D is a fitting parameter. $D = \alpha D_{avg} \beta$ in Eq. (7) of Ref. [58], D_{avg} is the average grain size and α and β are constants smaller than 1. Figure 5 displays the curve fitting result of the inclusion of the grain boundary scattering model of Eq. (35) as “ $b + pd + u + gb$ ” with the fitting parameters listed in Table 5. Even though, in the case of the 1000 nm thick sample, the best-fit parameter D that roughly corresponds to the average grain size D_{avg} is comparable to the film thickness, the fitting curve did not adequately express (the level of expression was the same as that for “ $b + pd + u + sf$ ”) the temperature dependence of the thermal conductivity at low temperatures.

5.2.6 Tunneling States Scattering

Tunneling states (TS) are another possible supplementary phonon scattering mechanism for evaluating the present thermal conductivity results. In the past, a number of authors claimed that TS exist in cuprate superconductors based mainly on their experimental observations of the glass-like behavior of the thermal conductivity at low temperatures. For example, Regueiro et al. [59] measured the in-plane thermal

Table 5 Best-fit parameters for the adopted phonon scattering models to express temperature dependence of out-of-plane thermal conductivity of 1000 nm thick YBCO

Adopted phonon scattering mechanisms	Point defects A (s^3)	Umklapp B ($s \cdot K^{-1}$)	Stacking faults C (s)	Grain boundary D (m)	Tunneling states P ($m^2 \cdot s^{-2}$)	SD of experimental results from fitted curve ($W \cdot m^{-1} \cdot K^{-1}$)
$b + pd + u$	5.44×10^{-42}	24.8×10^{-18}	—	—	—	1.33
$b + pd + u + sf$		0.237×10^{-18}	0.727×10^{-15}	—	—	0.26
$b + pd + u + gb$		0.510×10^{-18}	—	0.0430×10^{-6}	—	0.22
$b + pd + u + ts$		4.76×10^{-18}	—	—	17.5×10^4	0.13

b boundary scattering, pd point defects scattering, u umklapp scattering, sf stacking faults scattering, gb grain boundary scattering, ts tunneling states scattering

conductivity of bulk La_2CuO_4 and $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ samples and showed $T^{1.8}$ and $T^{1.4}$ dependences, respectively, at low temperatures. From these thermal conductivity results, together with the behavior of the specific heat and sound attenuation, they concluded that two-level tunneling systems are characteristic of an amorphous state in the $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ system, probably caused by oxygen vacancies. Izbizky *et al.* [60] reached a similar conclusion that their low-temperature (below 10 K) thermal conductivity results supported the assumption of an interaction between phonon heat carriers and tunneling systems with a broad distribution of energies and relaxation times in polycrystalline $\text{EuBa}_2\text{Cu}_3\text{O}_{7-x}$. In the case of YBCO, Graebner *et al.* [61] first pointed out the possibility of the existence of TS from the observation that the thermal conductivity is proportional to $T^{1.8\sim 1.9}$ in the temperature range below 5 K. Even though this hypothesis remains far from certain and needs to be rigorously tested, here we simply attempted to use this model as one of the phenomenological fitting tools to account for our results.

Phillips [62] gave the explicit form of the phonon relaxation time due to TS τ_{ts} by experimentally accessible quantities:

$$\frac{1}{\tau_{\text{ts}}} = \frac{P\omega}{v_g^2} \tanh(\hbar\omega/2k_{\text{B}}T), \quad (36)$$

where P is the fitting parameter we used in order to bundle some experimentally inaccessible parameters into a single parameter, assuming they are all independent of frequency and temperature. By carrying out the same curve fitting procedure as in the case of stacking faults and grain boundary scattering, Fig. 5 compares the fitted thermal conductivity adopting the tunneling states scattering model of Eq. (36) with the three other fitting curves described in the previous sections. As can be seen from Fig. 5 and Table 5, we found that it was possible to adequately fit the low-temperature data by introducing the phonon relaxation time due to TS scattering with a smaller standard deviation compared to the three other fitted curves.

6 Conclusion

In order to contribute to precise simulation of the two-dimensional transient heat conduction for coated high-temperature superconductor tape, we measured the out-of-plane thermal conductivity of epitaxially grown YBCO thin films (250 nm, 500 nm and 1000 nm) in the temperature range from 10 K to 300 K by using the photothermal reflectance technique. Our experimental results reveal the thickness dependence of the thermal conductivity throughout the entire temperature range. In the superconducting state, the size effect ($\lambda_{\text{film}}/\lambda_{\text{bulk}}$) of the thermal conductivity reaches less than one-tenth at the maximum. We also found that the thermal conductivity of the present YBCO thin films shows $T^{1.4}$ to $T^{1.6}$ glass-like dependence below 50 K even though the YBCO thin films are crystalline solids. In order to determine the reason for this temperature dependence, we attempted to interpret our experimental data using possible phonon relaxation models, including stacking faults, grain boundary and tunneling states scattering models.

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