9th Cimtec-World Forum on New Materials
Symposium VI - Science and Engineering of HTC Superconductivity
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ULTRASONIC AND PHONON THERMAL TRANSPORT STUDIES ON $YBa_2Cu_3O_{7-\delta}$ OXIDE SUPERCONDUCTORS

Hiroyuki FUJISHIRO, Manabu IKEBE and Tetsuo FUKASE*

Faculty of Engineering, Iwate University, 4-3-5 Ueda, Morioka 020-8551, Japan.
*Institute for Materials Research, Tohoku University, 2-1-1 Katahira, Sendai 980-8577, Japan.

The thermal conductivity κ and sound velocity v_s were measured for 90K-phase (δ=0.08), 60K-phase ($\delta\approx$0.3) and non-superconductive ($\delta\approx$0.7) $YBa_2Cu_3O_{7-\delta}$ (YBCO) sintered crystals. The use of sintered crystals greatly reduced the electron component κ_e of heat conduction and allowed us to determine the phonon component κ_{ph} more precisely. The observed characteristic κ enhancement below T_c is consistent with κ_{ph} model under the energy gap with d-wave symmetry. The electron-phonon interaction was found to be pretty strong in 90K-phase YBCO. The sound velocity and ultrasonic attenuation showed anomalies at about 90K for 90K-phase YBCO and possible correlation with superconductivity was suggested.

1. INTRODUCTION

In superconductors the thermal conductivity is almost a unique transport phenomena measurable below the transition temperature T_c. However, both electron component κ_e and phonon component κ_{ph} contribute to the total conductivity κ ($\kappa=\kappa_e+\kappa_{ph}$) of a superconductor and the unique separation of κ_e and κ_{ph} always becomes a problem in dispute. Roughly speaking, κ_e is to be reduced in 'dirty' samples with high electrical resistivity ρ because of the Wiedemann-Franz law. In this paper, we present and discuss the thermal conduction in sintered $YBa_2Cu_3O_{7-\delta}$ (YBCO). It is well-known that 90K-phase YBCO exhibits a characteristic enhancement in κ below T_c.[1] By the use of sintered YBCO with large residual resistivity ρ_0, we can limit the contribution of the electronic component to the κ enhancement. The separated phonon thermal conductivity is analyzed on the basis of the classical theory of Bardeen, Rickayzen and Tewordt.[2] The observed enhancement in κ is satisfactorily explained for both 90K- and 60K-phase YBCO by the κ_{ph} model, only if we assume the superconducting energy gap Δ of d-wave symmetry. The estimated electron-phonon coupling parameter λ is larger for 90K-YBCO than for 60K-YBCO.

2. EXPERIMENTAL PROCEDURE

$YBa_2Cu_3O_{7-\delta}$ polycrystals were prepared by a standard solid state reaction method. The stoichiometric mixtures of Y_2O_3, $BaCO_3$ and CuO powders were calcined at 910°C for 24h in air. They were pulverized, pressed into pellets, sintered at 955°C for 30h in flowing oxygen and then slowly cooled to room temperature in about 24h. These samples were named as 90K-YBCO. The 60K-phase (60K-YBCO) and non-superconductive samples were obtained by quenching the 90K-YBCO into liquid nitrogen from 600°C and 900°C, respectively. The amount of the oxygen deficiency δ was estimated to be ~0.3 for 60K-YBCO and ~0.7 for non-superconductive YBCO from the c-axis lattice parameter. The thermal conductivity was measured by a standard continuous heat flow method between temperatures 15K and 170K.[3] The ultrasonic experiments were performed with ~7MHz longitudinal and transverse waves generated by the Z-cut $LiNbO_3$ transducers. The sound velocity v_s and the attenuation coefficient α were measured using the pulse superposition method and the pulse echo method, respectively.

3. EXPERIMENTAL RESULTS

Figure 1 shows the temperature dependence of the electrical resistivity $\rho(T)$ of the 90K-, 60K- and non-superconductive samples. The absolute values of $\rho(T)$ of these superconductive samples are not large as sintered polycrystals. The residual resistivities $\rho(0)$ estimated by the extrapolation from the linear $\rho(T)$ in the normal state are 0.11mΩcm for the 90K-phase and 0.14mΩcm for the 60K-phase YBCO.

Figure 2 shows the temperature dependence of the thermal conductivity $\kappa(T)$ of the same samples as in Fig. 1. With decreasing temperature down to T_c, $\kappa(T)$ of 90K-YBCO slightly increases and then shows a characteristic enhancement related to the onset of superconductivity. $\kappa(T)$ of 60K-YBCO follows a qualitatively similar curve to that of 90K-YBCO. The characteristic enhancement below T_c is, however, more conspicuous for the 90K-sample than for the 60K-sample. $\kappa(T)$ takes a maximum at $T_{max}\approx45K$ for 90K-YBCO and at $T_{max}\approx37K$ for 60K-YBCO. The absolute values of $\kappa(T)$ of 60K-YBCO are smaller than those of 90K-YBCO over the entire measured temperature range. $\kappa(T)$ of the non-superconductive sample is the smallest and monotonically decreases with decreasing temperature. The temperature dependence of $\kappa(T)$ of the non-superconductive sample is quite small above ~40K.

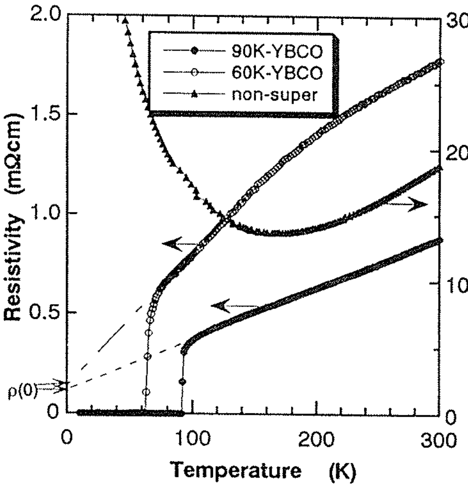

FIGURE 1
Temperature dependence of the electrical resistivity ρ(T) of the 90K-, 60K- and non-superconductive samples.

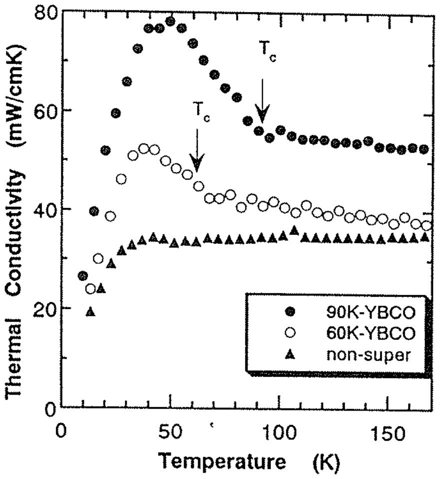

FIGURE 2
Temperature dependence of the thermal conductivity κ(T) of the same samples as in Fig. 1.

Figures 3(a) and (b) show the temperature dependence of the sound velocity $v_s(T)$ and the sound attenuation $\alpha(T)$ for 90K- and 60K-YBCO. In Fig. 3(a), $v_s(T)$ of 90K-phase YBCO takes a clear upturn at around 90K with decreasing temperature, which is accompanied with the maximum of attenuation $\alpha(T)$. In Fig. 3(b), at around 60K, the similar anomalies in $v_s(T)$ and $\alpha(T)$ are barely discernible for 60K-YBCO.

4. DISCUSSION

4.1 Separation of the electronic component κ_e from the total conductivity κ

As is well-known, the electronic thermal conductivity $\kappa_{en}(T)$ in the normal state can be estimated by the Wiedemann-Franz law. In the superconducting state, the thermal energy carried by quasi-particles (with the density n_{qp}) rapidly decreases with decreasing temperature. For cuprate superconductors, several experimental studies suggested a rapid suppression of the quasi-particle scattering rate τ_{qp}^{-1} relative to the normal electron scattering rate τ_e^{-1}.[4,5] Then there is a possibility that the electronic thermal conductivity κ_{es} in the superconducting state is enhanced in comparison to κ_{en}, the reduction in τ_{qp}^{-1} overwhelming the reduction in n_{qp}. Here we estimate $\kappa_{es}(T)$ following the formula derived by Kadanoff and Martin,[6]

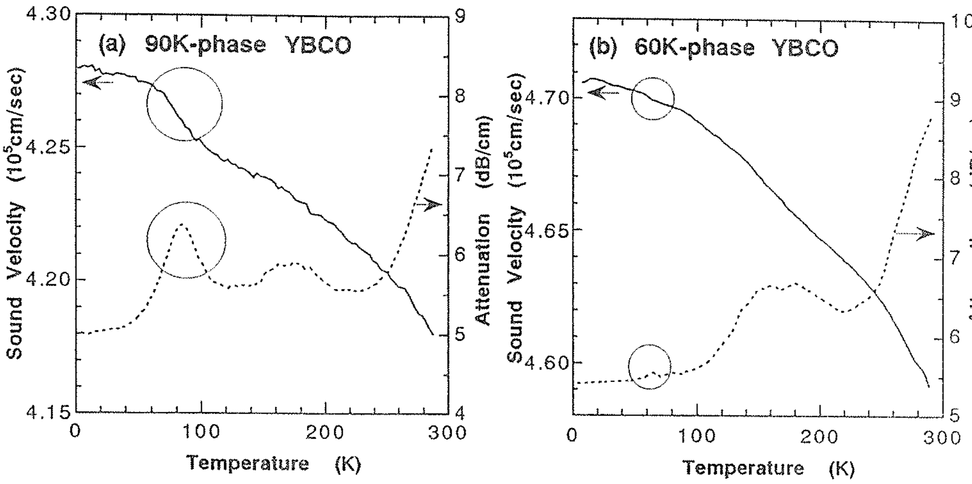

FIGURE 3 Temperature dependence of the sound velocity v_s(T) and the sound attenuation α(T) of (a) 90K-phase and (b) 60K-phase YBCO.

$$\frac{\kappa_{es}}{\kappa_{en}}=\frac{3}{4\pi^2}\int_0^{2\pi}d\phi\int_0^{\infty}d\varepsilon\varepsilon^2\mathrm{sech}^2\left\{\frac{1}{2}[\varepsilon^2+(\beta\Delta)^2]^{\frac{1}{2}}\right\}\frac{1+at^3}{\frac{\varepsilon}{[\varepsilon^2+(\beta\Delta)^2]^{\frac{1}{2}}}+at^3},\quad(1)$$

where ε is the electron energy relative to the chemical potential, β is $1/k_BT$ and t is the reduced temperature T/T$_c$. κ_{en} is equal to $(\pi^2/3)(n_e/m)k_BT\tau_e$, m and n_e being the mass and the density of the electron. The energy gap Δ is assumed to be $\Delta=\Delta_0$ for s-wave coupling, and $\Delta=\Delta_{max}\cos2\phi$ for d-wave coupling, φ being the azimuthal angle in the two-dimensional circular Fermi surface. We assume that the electrical resistivity (i.e., quasi-particle resistivity below T$_c$) follows t^n temperature dependence with the residual resistivity equal to ρ(T$_c$)/(a+1) at 0K,

$$\rho(T)=\rho(T_c)(\frac{1}{a+1}+\frac{a}{a+1}t^n),\quad(2)$$

where ρ(T$_c$) is the resistivity value at T$_c$. The a value in eqs. (1) and (2) is the ratio of the coefficient of the two terms at T$_c$. For the electrical resistivity above T$_c$ (t>1), n is set to be 1 in eq. (2) as is to be consistent with experimental results. Below T$_c$ we set the n=3, which corresponds to τ_{qp}^{-1} limited by the spin fluctuating scattering.[7] Following the results of the tunneling spectroscopy due to Nantoh et al.,[8] we take the maximum energy gap value $\Delta_{max}=\Delta_0=1.5\Delta_{BCS}$ and calculated eq. (1) for both d-wave and s-wave symmetry. The results of 90K-YBCO are presented in Figs. 4(a) and 4(b) for d-wave and s-wave symmetry and the results of 60K-YBCO are presented in Figs. 5(a) and 5(b). κ_{es} of both present 90K- and 60K-YBCO does not show any trend of enhancement below T_c. Because the present samples have sizable residual resistivity ($\rho(0) \geq 0.13$mΩcm), the absence of the κ_{es} enhancement remains almost intact even if we make more drastic assumption for τ_{qp}^{-1} such as $\tau_{qp}^{-1} \propto \exp(-2\Delta/k_BT)$.

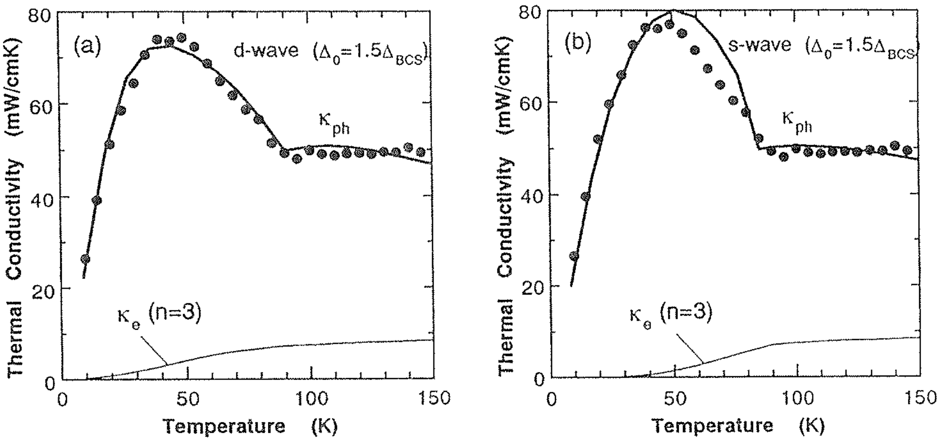

FIGURE 4 The calculated electronic thermal conductivity κ_{en} and the best fitting κ curves of 90K-YBCO for (a) d-wave and (b) s-wave symmetry.

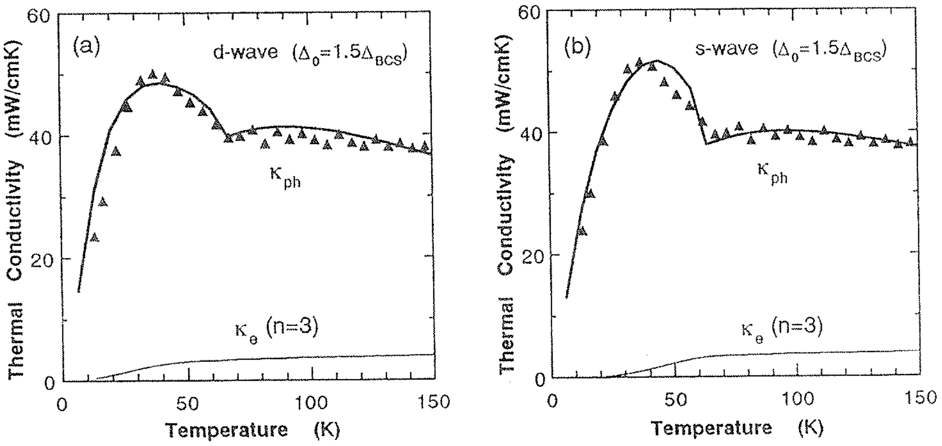

FIGURE 5 The calculated electronic thermal conductivity κ_{en} and the best fitting κ curves of 60K-YBCO for (a) d-wave and (b) s-wave symmetry.

4.2 Analyses of the phonon thermal conductivity κ_{ph} based on the BRT-TW theory

Assuming the Debye spectrum of phonons, the phonon thermal conductivity is generally given by the following formula;[9]

$$\kappa_{\mathrm{ph}} = \frac{3dn_0\mathrm{R} < v_s^{\,2} >}{2\pi\mathrm{M}} \left(\frac{T}{\Theta_\mathrm{D}}\right)^3 \int_0^{2\pi} d\phi \int_0^{\Theta_D/T} \frac{x^4 e^x}{(e^x - 1)^2} \tau_{\mathrm{ph}}(x) dx \quad , \qquad (3)$$

where x is the reduced phonon frequency, n_0 (=13-δ) the number of atoms composing $YBa_2Cu_3O_{7-d}$, R the gas constant, d the mass density and M is the molar weight. Eq. (3) contains integral as to ϕ to treat the d-wave anisotropy in the superconducting state. The phonon scattering rate τ_{ph}^{-1} is assumed to be given by the sum of contributions of various scattering centers,

$$\tau_{\mathrm{ph}}^{\;-1} = \tau_{\mathrm{b}}^{\;-1} + s(Tx)^2 + p(Tx)^4 + E(Tx)g(x,y) + UT^3x^2\exp(-\frac{\Theta_D}{bT}) \; . \qquad (4)$$

Here, parameters τ_b^{-1}, s, p, E and U represent the phonon scattering strength due to grain boundaries, sheet-like fault, point defects, conduction electrons and other phonons (umklapp process), respectively, and b(=2.1) is a constant which limits the cut-off frequency of the umklapp process. The function $g(x, y) = \tau^e_{phn}/\tau^e_{phs}$ stands for the ratio of the phonon scattering times by electrons in the superconducting and normal states, which was first calculated by Bardeen, Rickayzen and Tewordt (BRT)[2] on the basis of the BCS theory. With decreasing temperature in the superconducting state, $g(x, y)$ promptly decreases, depending on the magnitude of the energy gap Δ through the parameter $y=\Delta/k_B T$. The decrease of $g(x, y)$ results in the increase of κ_{phs} In the superconducting state as was observed in many conventional superconductors.[10] We calculated κ_{ph} on the basis of eqs. (3) and (4) for both the d-wave and s-wave energy gap symmetry with

Table I. Fitting parameters for the weak coupling of d-wave and s-wave symmetry with $\Delta_{max}=\Delta_0=1.5\Delta_{BCS}$.

	$\tau_b^{-1}(s^{-1})$	$s(K^{-2}s^{-1})$	$P(K^{-4}s^{-1})$	$E(K^{-1}s^{-1})$	$U(K^{-3}s^{-1})$	λ
d-wave						
90K-YBCO	4.7×10^8	3.2×10^6	2.3×10^2	1.2×10^9	3.6×10^3	0.42
60K-YBCO	4.9×10^8	3.3×10^6	1.2×10^3	4.3×10^8	3.4×10^3	0.16

$\Delta_{max}=\Delta_0=1.5\Delta_{BCS}$. The best fitting curves are presented in Figs. 4(a) and 4(b) for 90K-phase and Figs. 5(a) and 5(b) for 60K-phase YBCO. As one can see in these figures, only d-wave κ_{ph} model satisfactorily reproduces the experimental results. The parameter values used and determined in the d-wave κ_{ph} fitting are summarized in Table I.

Following the procedure due to Tewordt and Wölkhausen (TW),[9] the electron-phonon coupling constant λ is estimated by the following equation,

$$\lambda = \frac{2c < t >}{\pi v_s} E, \tag{5}$$

where E is the phonon scattering term by electrons in eq. (4), c is the average lattice constant and $< t >$ is the effective hopping matrix element in the CuO_2 plane. A rough estimation gives λ=0.42 for 90K-phase and λ =0.16 for 60K-phase YBCO. It is noteworthy that λ value of 90K-phase is pretty large and far larger than that of 60K-phase YBCO. It is also noteworthy that the v_s anomaly and the attenuation maximum at around 90K of 90K-phase YBCO is also far stronger than those of 60K-phase YBCO. It seems to be possible, though not conclusive, that the anomalous behavior of sound propagation, in especially 90K-phase YBCO, comes from the strong electron-phonon coupling related to the onset of the superconductivity.

5. SUMMARY

(1) The electronic component κ_{es} of the thermal conduction does not contribute to the observed enhancement κ below T_c in sintered 'dirty' 90K- and 60K-phase $YBa_2Cu_3O_{7-\delta}$ because of sizable residual resistivity $\rho(0)$.

(2) The κ enhancement can be satisfactorily explained by the dominant phonon conduction model under the energy gap of the d-wave symmetry and with the maximum energy gap $\Delta_{max}=1.5\,\Delta_{BCS}$.

(3) Electron-phonon coupling is pretty strong in 90K-phase YBCO and its contribution to superconductivity is not to be neglected.

(4) The observed anomalous behavior of the sound velocity v_s and ultrasonic attenuation α near T_c might be related to the onset of superconductivity in the $YBa_2Cu_3O_{7-\delta}$ system.

ACKNOWLEDGEMENT

The authors wish to thank K. Nakasato and T. Mikami of Iwate University in the fitting procedures. The sound velocity measurement was performed under the inter-university cooperative research program of the Institute for Material Research, Tohoku University.

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