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Thermal Conductivity and Phonon Scattering Mechanisms in $La_{2-X}M_XCuO_4$

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The thermal conductivity κ of high density $La_{2-X}M_XCuO_4$ (M=Ba, Sr) sintered materials was measured between 15K and 150K for the various concentrations of Ba and the phonon thermal conductivity was analyzed comparing with that of $Nd_{2-X}Ce_XCuO_4$. The pretty large κ value for pure La_2CuO_4 was drastically diminished by substituting La by a small amount of Ba atoms especially at low temperatures. It was found that a new type of the phonon scattering center such as a two-level tunneling must be taken account of in order to explain the observed κ reduction.

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1. INTRODUCTION

In several high-T_c cuprates such as YBCO, Bi2223 etc., the temperature dependence of the thermal conductivity κ(T) shows a characteristic enhancement below the transition temperature T_c. In contrast, so-called (214) family such as $La_{2-X}M_XCuO_4$ (M=Ba, Sr) with T-type and $Ln_{2-X}Ce_XCuO_4$ (Ln=Nd, Pr) with T'-type crystal structure do not clearly show the characteristic κ(T) enhancement. There is much argument about the origin of the κ(T) enhancement, whether it is electronic or phononic. We have measured the thermal conductivity κ of $YBa_2Cu_3O_7$[1], $(BiPb)_2Sr_2Ca_2Cu_3O_X$[2], $Nd_{2-X}Ce_XCuO_4$[3] etc. and have systematically analyzed the phonon thermal conductivity κ_{ph} based on the Tewordt and Wölkhausen (TW) theory[4]. We fabricated high density sintered $La_{2-X}Ba_XCuO_4$ and $La_{1.85}Sr_{0.15}CuO_4$ crystals and the thermal conductivity κ and the thermal diffusivity α were investigated as a function of Ba concentration X. In this paper, the phonon scattering mechanisms in La214 system are analyzed.

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2. EXPERIMENTAL

$La_{2-X}Ba_XCuO_4$ sintered materials ($0 \leq X \leq 0.2$) and $La_{1.85}Sr_{0.15}CuO_4$ were prepared from stoichiometric mixtures of La_2O_3, $BaCO_3$ ($SrCO_3$) and CuO raw powders. The mixtures were calcined twice at 880°C for 12h in air. They were pressed into pellets and sintered at 1100°C for 18h in air and then furnace-cooled. Finally, the samples were heat-treated at 800°C for 24h in flowing oxygen. The X-ray diffraction analyses confirmed a single T-phase structure for all the samples. The density of each sample was higher than 90% of the ideal density. The κ measurement was made between 10 ~ 150K by a continuous heat flow method and α was measured simultaneously with κ by an arbitrary heating method under an identical experimental setup[2].

3. RESULTS

Figure 1 shows the temperature dependence of the electrical resistivity ρ of $La_{2-X}Ba_XCuO_4$ ($0 < X \leq 0.2$) and $La_{1.85}Sr_{0.15}CuO_4$ samples. The ρ values of Ba-substituted samples decreased with increasing Ba concentration X. T_c increased with increasing X and reached a maximum of T_c=28K (X=0.15) and then decreased with further increase of X. $La_{1.85}Sr_{0.15}CuO_4$ showed T_c=38K, the highest T_c among the samples in this report. Figure 2 shows the thermal conductivity κ as a function of temperature T. The pretty large κ values for

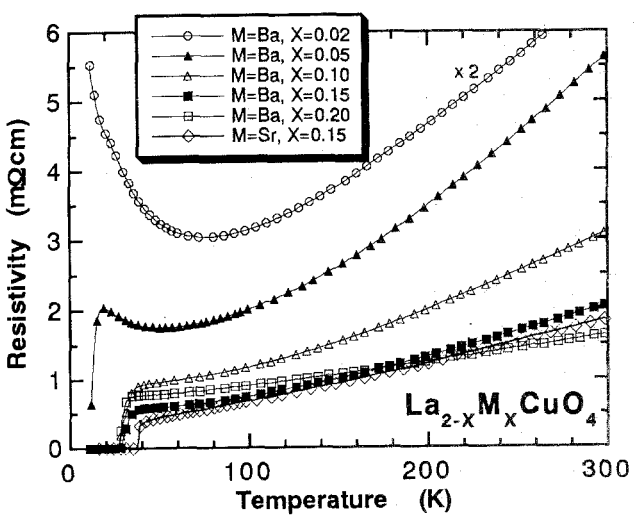

Fig. 1. The temperature dependence of the electrical resistivity ρ of $La_{2-X}Ba_XCuO_4$ ($0 \leq X \leq 0.2$) and $La_{1.85}Sr_{0.15}CuO_4$ samples.

the pure La_2CuO_4 sample with the maximum value about 80mW/cmK at about 37K were drastically diminished by substituting La by Ba atoms. This remarkable reduction in κ makes a marked contrast to Nd substitution by Ce in $Nd_{2-X}Ce_XCuO_4$, in which case κ was enhanced with X as shown in Fig. 3[3]. The specific heat C(T) was estimated from the relation $C=\kappa/\alpha$ and the Debye temperature Θ_D=400K was found to be roughly appropriate for all the present samples from Debye specific heat lines.

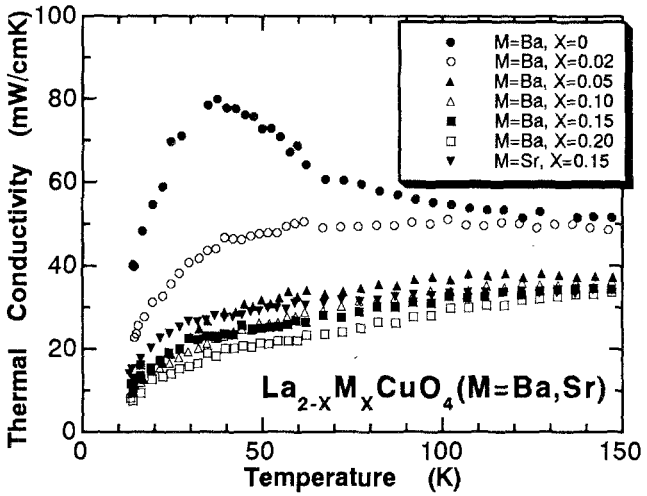

Fig. 2. The temperature dependence of the thermal conductivity κ of $La_{2-X}Ba_XCuO_4$ ($0 \leq X \leq 0.2$) and $La_{1.85}Sr_{0.15}CuO_4$ samples.

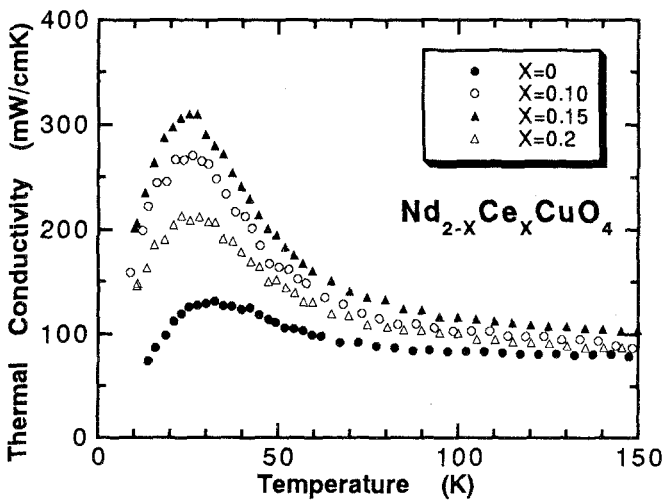

Fig. 3. The temperature dependence of the thermal conductivity κ of $Nd_{2-X}Ce_XCuO_4$ ($0 \leq X \leq 0.2$) samples of our previous study[3].

4. DISCUSSION

The heat transport in conductors is due to both electrons (κ_e) and phonons (κ_{ph}). We assume that the electron contribution in the superconducting state, κ_{es} follows the theory proposed by Kadanoff and Martin[5] with the d-wave energy gap $\Delta=2\Delta_{BCS}cos\,2\phi$[6]. Taking account of the phonon scattering by various crystal defects and electrons, the phonon thermal conductivity $\kappa_{ph}=\kappa-\kappa_e$ is given by[7]

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

where $n(=7)$ is the number of atoms composing these compounds, R the gas constant, $v(=5000\mathrm{m/s})$ the average phonon velocity and x is the reduced phonon frequency, respectively. The phonon relaxation time τ_{ph} is given by

$$\begin{aligned}\tau_{ph}{}^{-1} &= \tau_b{}^{-1} + \tau_p{}^{-1} + \tau_U{}^{-1} + \tau_e{}^{-1} + \tau_K{}^{-1} \\ &= \tau_b{}^{-1} + pT^4x^4 + Ux\exp(-\Theta_D/aT) + ET x g(x,\ y) + KTx.\end{aligned} \tag{2}$$

Here, τ_b is the phonon relaxation time due to grain boundaries and p and E refer to the strength of the phonon scattering by point defects and electrons, respectively. The function $U\exp(-\Theta_D/aT)$ $(a=1.8)$ is a standard form of phonon Umklapp processes at relatively low temperatures[8]. Let us notice that KTx term, which usually corresponds to the phonon scattering by strain fields of dislocation, may also be regarded as standing for the scattering by two-level tunneling states familiar in amorphous solids. The KTx term results in T^2 dependence of $\kappa(T)$ at very low temperatures and gives much the same results as $K'Tx\tanh(x/2)$ term which explicitly corresponds to the tunneling level scattering. The function $g(x,\ y)=\tau_{phn}/\tau_{phs}$ which depends on the energy gap through the parameter $y=\Delta(T)/k_BT$ stands for the ratio of the phonon-electron relaxation time in the normal and superconducting state[11]. $\tau_b=l_b/v$ was determined by the grain size of each sintered material l_b, which was measured by a scanning electron microscope (SEM).

Figure 4(a) shows the fitting curves for the phonon thermal conductivity κ_{ph} of the $La_{2-x}Ba_xCuO_4$ samples. The parameters used and determined in the fitting process are summarized in Table I. The calculated $\kappa_{ph}(T)$ could fully reproduce the experimental data. κ_{ph} was remarkably reduced even for Ba concentration of only x=0.02. The phonon scattering by point defects increased presumably by substituted Ba ions and the strength of Umklapp processes U decreased. The reduction in κ is not due to only the increase of

the point defect and the boundary scatterings. The strength of the phonon scattering represented by the KT^x term increased with increasing X. This increase of the K term is mainly responsible for the peculiar κ depression at low temperatures. We propose that the enhancement of the K term comes from local structural instability probably connected with the instability of the low temperature orthorhombic (LTO) lattice against the high temperature tetragonal (HTT) phase. The LTO phase is known to become more unstable

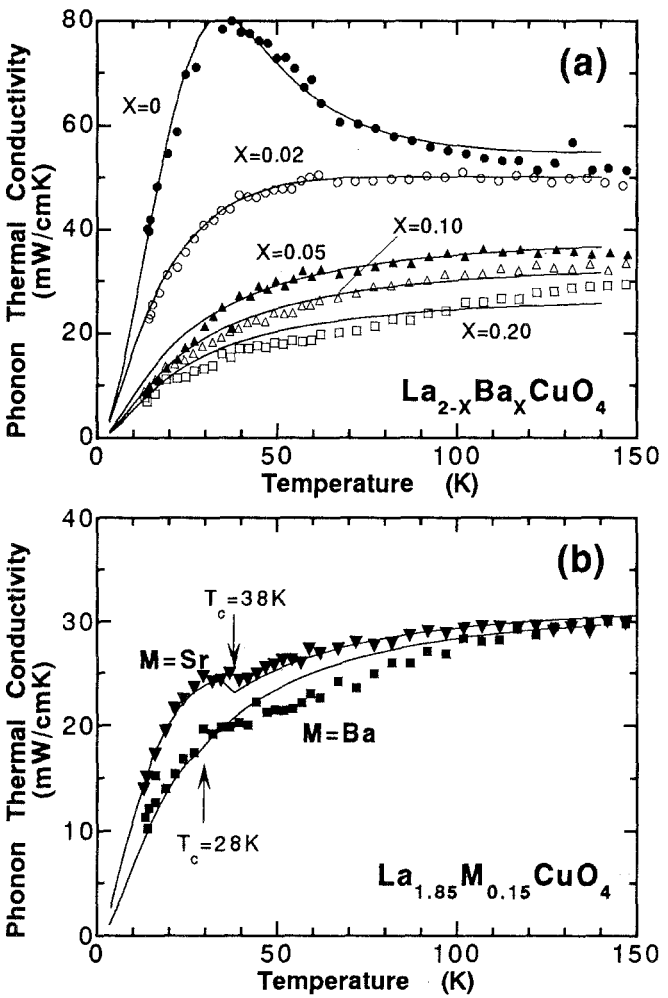

Fig. 4. (a) The fitting curves for the phonon thermal conductivity κ_{ph} of the $La_{2-X}Ba_XCuO_4$ samples. (b) The κ_{phs} fitting curves for $La_{1.85}Ba_{0.15}CuO_4$ and $La_{1.85}Sr_{0.15}CuO_4$ superconducting samples.

Table I. Fitting parameters determined by κ_{ph} analyses.

	LBCO (X=0)	LBCO (X=0.02)	LBCO (X=0.05)	LBCO (X=0.1)	LBCO (X=0.15)	LBCO (X=0.2)	LSCO (X=0.15)
$\tau_b^{-1}(s^{-1})$	1.6×10^8	2.9×10^8	5.7×10^8	8.1×10^8	8.1×10^8	1.0×10^9	6.7×10^8
$l_b(\mu m)$	31	17	9	6	6	5	7
$U(K^{-2}s^{-1})$	1.8×10^{12}	1.3×10^{11}	0	0	0	0	0
$p(K^{-4}s^{-1})$	1.1×10^3	4.8×10^3	6.8×10^3	7.3×10^3	8.1×10^3	9.3×10^3	1.1×10^4
$E(K^{-1}s^{-1})$	0	0	0	0	8.1×10^7	0	1.9×10^8
λ	0	0	0	0	0.02	0	0.05
$K(K^{-1}s^{-1})$	2.7×10^8	3.7×10^8	6.8×10^8	8.9×10^8	8.9×10^8	1.0×10^9	3.2×10^8
$T_c(K)$	-	-	10	26	28	25	38

by substitution of La by Ba and Sr atoms. The tunneling states related to apical oxygens may be the origin of the K term, because the apical oxygen plays a key role at LTO to HTT transition.

Figure 4(b) shows the κ_{phs} fitting curves for $La_{1.85}Ba_{0.15}CuO_4$ and $La_{1.85}Sr_{0.15}CuO_4$ superconducting samples. The characteristic enhancement associated with the onset of superconductivity is confirmed, though very weak, in this figure, which means that the electron-phonon coupling cannot be neglected in La214 system. The enhancement of κ of $La_{1.85}Sr_{0.15}CuO_4$ is clearer than that of $La_{1.85}Ba_{0.15}CuO_4$ and the electron phonon coupling constant λ of $La_{1.85}Sr_{0.15}CuO_4$ ($=2a<t>E/\pi v$, a is the lattice constant, $<t>$ the effective hopping matrix element of electrons[9]) is three-times larger than that of $La_{1.85}Ba_{0.15}CuO_4$, both of which, however, were about one order of magnitude smaller than that of the 90K-phase YBCO[1].

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