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Two-level-like anomalous phonon scattering in $La_{2-x}Sr_xCuO_4$ and $La_{1-x}Sr_xMnO_3$

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Abstract

Thermal conductivities κ of high-T_c cuprate $La_{2-x}Sr_xCuO_4$ ($0 \leqslant x \leqslant 0.30$) and manganese oxides $La_{1-x}Sr_xMnO_3$ ($0.11 \leqslant x \leqslant 0.30$) have been measured and phonon scattering mechanisms have been analyzed. In both systems phonon scattering is remarkably enhanced around a peculiar Sr concentration x, at which structural transformations occur. The tunneling motion of oxygen atoms related to the structural transformation is suggested to be the main origin of the enhancement.

Keywords: Thermal conductivity; Phonon scattering; Oxide superconductors; Manganese oxides

Among (2 1 4) cuprate superconductors, $La_{2-x}Sr_xCuO_4$ system with the so-called T-phase structure shows much smaller κ values in comparison to $Nd_{2-x}Ce_xCuO_4$ system with T′-phase structure. The existence of apical oxygen atoms at both sides of a unit Cu–O square and the structural instability between the high-temperature tetragonal (HTT) and the low-temperature orthorhombic (LTO) phase seem to be the origin of the smaller κ values of the T-phase [1]. As materials related to the high-T_c cuprates, manganese oxides with perovskite structure, $R_{1-x}A_xMnO_3$ (R is a rare-earth trivalent ion such as La and A a divalent ion such as Sr and Ca) have recently attracted renewed and extended interest because of several dramatic properties such as the colossal magnetoresistance. In $La_{1-x}Sr_xMnO_3$, MnO_6 octahedra network exist similar to CuO_6 octahedra in $La_{2-x}Sr_xCuO_4$ [2]. In this paper, we report the thermal conductivities κ of $La_{2-x}Sr_xCuO_4$ ($0 \leqslant x \leqslant 0.30$) oxide superconductors and $La_{1-x}Sr_xMnO_3$($0.11 \leqslant x \leqslant 0.30$) manganese oxides. The phonon scattering mechanisms operating in both systems are analyzed.

Specimens of $La_{2-x}Sr_xCuO_4$($0 \leqslant x \leqslant 0.30$) (LSCO) and $La_{1-x}Sr_xMnO_3$($0.11 \leqslant x \leqslant 0.30$) (LSMO) were fabricated by a standard solid-state reaction method. Detailed fabricating procedures were reported elsewhere [1,3]. The X-ray diffraction analysis confirmed that all samples were in a single phase except for $x = 0.30$ of LSCO which showed a trace of impurity phases. The density of the samples was higher than 90% and 85% of each ideal density for LSCO and LSMO, respectively. The thermal conductivity κ measurement was made between 10–300 K by a continuous heat flow method with an automatic measuring system [4].

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The heat conduction in conductors is due to both electrons (κ_e) and phonons (κ_{ph}). For the LSCO system, we assumed that the electron contribution in the superconducting state, κ_{es}, follows the theory proposed by Kadanoff and Martin with the d-wave energy gap anisotropy, $\Delta = \Delta_0 \cos 2\phi$ [5], and the electron contribution in the normal state, κ_{en}, follows the Wiedemann–Franz law. Fig. 1 shows the temperature dependence of the phonon thermal conductivity $\kappa_{ph}(=\kappa-\kappa_e)$ of LSCO for various Sr concentrations. The pretty large κ_{ph} values of the pure La_2CuO_4 sample ($\kappa_{ph}^{max} = 80$ mW/cm K) are drastically diminished by substituting La by Sr atoms up to $x = 0.20$ and then κ_{ph} again increases with further increase of x. The reduction of κ_{ph} is more pronounced in the low temperature region ($T < 40$ K). In order to analyze the phonon scattering mechanisms, we calculated $\kappa_{ph}(T)$ on the basis of the relaxation time approximation [6]. In this analyses, the phonon scattering rate τ_{ph}^{-1} is assumed to be given by the sum of contributions of various scattering centers,

$$\tau_{ph}^{-1} = \tau_b^{-1} + sT^2x^2 + pT^4x^4 + ETxg(x, y) + Ux\exp\left(-\frac{\Theta_D}{bT}\right) + KTx\tanh\frac{x}{2}. \quad (1)$$

Here, x is the reduced phonon frequency and parameters τ_b^{-1}, s, p and E represent the phonon scattering strength due to grain boundaries, sheet-like faults, point defects and electrons, respectively. The term $Ux\exp(-\Theta_D/bT)$ ($b = 2.0$, Θ_D : Debye temperature) is a standard form of the phonon umklapp processes. $g(x, y) = \tau_{phn}/\tau_{phs}$ is the ratio of the phonon–electron scattering rate in the normal and superconducting state, which depends on the energy gap through the parameter $y = \Delta(T)/k_BT$. The last term $KTx\tanh(x/2)$ stands for the scattering by two-level tunneling states [7], which becomes dominant at low temperatures. The fitting curves for $\kappa_{ph}(T)$ are also shown in Fig. 1 and the parameters determined in the fitting processes are summarized in Table 1. It is worthwhile to notice that the K value increases with increasing x up to $x = 0.20$ and then drastically decreases with the further increase of x. We propose that the enhancement of the K value comes from the local structural instability connected with the instability of the low temperature orthorhombic (LTO) phase against the high temperature tetragonal (HTT) phase. The structural phase boundary between LTO and HTT was reported to exist at $x \sim 0.21$ [8]. We infer that the dynamical tunneling motion of apical oxygens is the most important origin of the K term phonon scattering, because the apical oxygen plays a key role in the LTO to HTT transition.

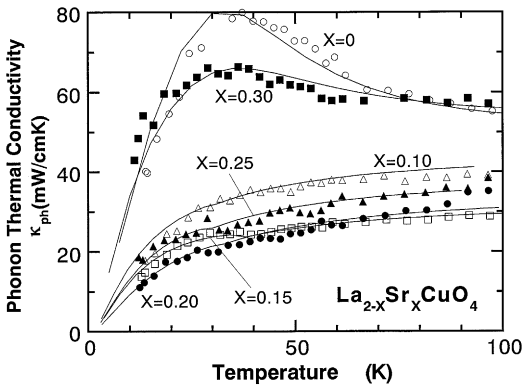

Fig. 1. The temperature dependence of the phonon thermal conductivity $\kappa_{ph}(=\kappa-\kappa_e)$ and the calculated fitting curves for $\kappa_{ph}(T)$ of LSCO for various Sr concentrations ($0 \leqslant x \leqslant 0.30$).

Fig. 2 shows the temperature dependence of the thermal conductivity $\kappa(T)$ of $La_{1-x}Sr_xMnO_3$ for various Sr concentrations x. The κ value for $x < 0.17$ samples is suppressed to low values and then drastically increases with the increase of the Sr concentration X from $x = 0.18$ to $x = 0.20$. In this LSMO system, the phase boundary of the structural transformation between high temperature rhombohedral and low temperature orthorhombic lattice exists at $X = 0.17$–0.18 [2]. The structural instabilities may suppress the κ values through a similar oxygen tunneling process as $La_{2-x}Sr_xCuO_4$ because the orthorhombic to rhombohedral transition at $x \sim 0.18$ is also related to the tilting of MnO_6 octahedra and shifts of oxygen sites play a major role. The local minima of κ at $T = 240$ K for $x = 0.155$ and $T = 270$ K for $x = 0.17$ result from the enhanced phonon scattering due to ferromagnetic transition and the dynamical nature of the phonon scattering around the ferromagnetic transition was pointed out in a previous paper [9].

Table 1
Summary of the parameters determined in the fitting processes in $La_{2-x}Sr_xCuO_4$ and $La_{1-x}Sr_xMnO_3$ systems

Sample	x	τ_b^{-1} (s^{-1})	s($K^{-2}s^{-1}$)	p($K^{-4}s^{-1}$)	E($K^{-1}s^{-1}$)	U(s^{-1})	K($K^{-1}s^{-1}$)
LSCO	0	9.9×10^{6}	1.9×10^{6}	2.0×10^{3}	0	9.9×10^{11}	1.1×10^{8}
	0.10	3.2×10^{8}	5.0×10^{6}	1.4×10^{4}	3.6×10^{7}	0	2.2×10^{8}
	0.15	6.4×10^{8}	5.5×10^{6}	1.5×10^{4}	2.5×10^{8}	0	2.5×10^{8}
	0.20	6.5×10^{8}	1.8×10^{7}	1.1×10^{4}	1.4×10^{8}	0	3.5×10^{8}
	0.25	7.0×10^{8}	9.0×10^{6}	1.3×10^{4}	1.2×10^{8}	0	2.2×10^{8}
	0.30	7.8×10^{8}	5.5×10^{6}	6.6×10^{3}	0	2.3×10^{11}	0
LSMO	0.135	1.0×10^{8}	1.1×10^{6}	2.8×10^{4}	0	1.1×10^{11}	2.1×10^{8}
	0.17	9.5×10^{7}	9.5×10^{5}	6.2×10^{3}	0	9.5×10^{10}	1.0×10^{9}
	0.18	9.1×10^{7}	9.1×10^{5}	5.5×10^{3}	0	9.1×10^{10}	1.8×10^{8}
	0.20	6.4×10^{7}	4.3×10^{6}	1.3×10^{3}	0	3.1×10^{11}	6.4×10^{6}
	0.30	4.0×10^{7}	4.4×10^{6}	1.1×10^{3}	0	1.9×10^{11}	4.0×10^{5}

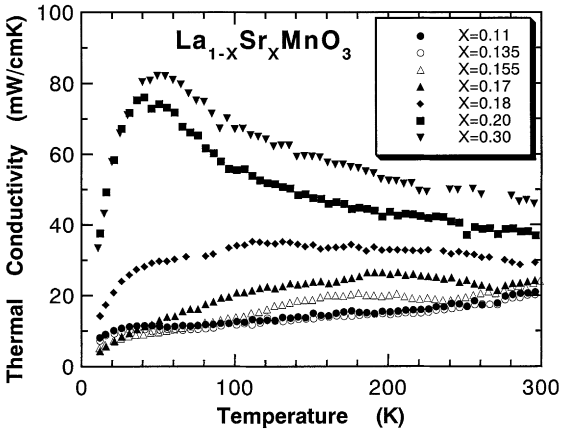

Fig. 2. The temperature dependence of the thermal conductivity $\kappa(T)$ of $La_{1-x}Sr_xMnO_3$ for various Sr concentrations ($0.11 \leqslant x \leqslant 0.30$).

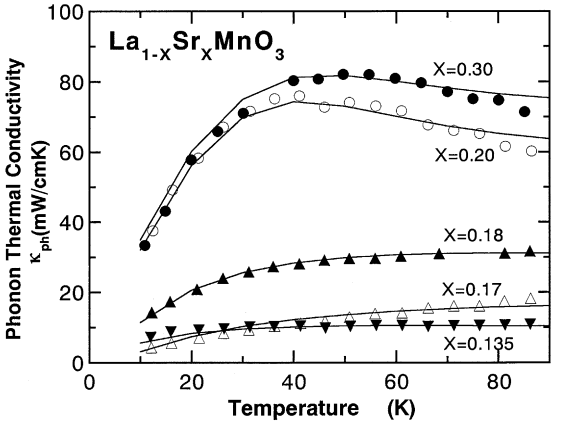

Fig. 3. The calculated fitting curves for $\kappa_{ph}(T)$ of typical $La_{1-x}Sr_xMnO_3$ samples.

The κ behavior of LSMO was analyzed in the same way as that of LSCO system. Since κ_e is very small for the LSMO samples as estimated from the electrical resistivity values [3], we regarded the measured κ as κ_{ph}. In this case the coefficient of the electron term E in the phonon scattering rate τ_{ph}^{-1} in Eq. (1) was assumed to be zero. The typical fitting curves for $\kappa_{ph}(T)$ are shown in Fig. 3 and the parameters determined in the fitting processes are also summarized in Table 1. For $x \leqslant 0.20$, the phonon scattering strength represented by the K term remarkably increases. Thus the main origin of the phonon scattering enhancement in LSMO at low temperatures can also be explained by the K term contribution. Thus the phonon scattering in both $La_{2-x}Sr_xCuO_4$ and $La_{1-x}Sr_xMnO_3$ systems is heavily enhanced near the structural instabilities and a dynamic phonon scattering mechanism such as two-level-like tunnelings of oxygen atoms is likely to be at work commonly in $La_{2-x}Sr_xCuO_4$ and $La_{1-x}Sr_xMnO_3$ systems.

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