# Anisotropic Thermal Conductivity of $RE_2CuO_4$ (RE = Pr, Nd, and Sm) Single Crystals Grown by TSFZ Method

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(Received April 12, 2010)

We have measured the thermal conductivity in the *ab*-plane and along the *c*-axis, $\kappa_{ab}(T)$ and $\kappa_c(T)$, for the large single crystallines of $RE_2CuO_4$ (RE = Pr, Nd, and Sm). A double-peak structure which originated from the phonon and magnon contributions was found in $\kappa_{ab}(T)$ for all the $RE_2CuO_4$ single crystals. The magnon peak in $\kappa_{ab}(T)$ strongly depends on the species of RE. We found a very large magnon contribution in $Sm_2CuO_4$. This can be explained by the interaction between the Sm and Cu moments. $\kappa_c(T)$ of $Sm_2CuO_4$ also showed an anomalous large peak around 25 K.

PACS numbers: 74.72.Cj, 65.40.G-

## I. INTRODUCTION

A double-peak structure of the thermal conductivity in the *ab*-plane, $\kappa_{ab}(T)$, was observed in $La_2CuO_4$ [1] and $Nd_2CuO_4$ [2] single crystals. The low-temperature peak was considered to be the conventional phonon peak. On the other hand, the high-temperature peak was interpreted to be originated from the magnetic excitations, i.e., the magnons, because $RE_2CuO_4$ (RE214; RE = rare earth elements) is an antiferromagnetic insulator. Later, Berggold *et al.* [3] measured both $\kappa_{ab}(T)$ and $\kappa_c(T)$ for Pr214 and Gd214 single crystals and only $\kappa_{ab}(T)$ for Sm214 and Eu214. They also observed the high-temperature peak in $\kappa_{ab}(T)$ of all the RE214 single crystals. Therefore, the high-temperature magnon peak in $\kappa_{ab}(T)$ seems to be established as a common feature in the RE214 system. However, the magnitude of the magnon contribution is still controversial as follows. For La214, the magnon peak is very large and comparable to the phonon peak. On the other hand, the high-temperature peak is rather smaller than the low-temperature peak for other RE214. The thermal conductivity along the *c*-axis, $\kappa_c(T)$, showed only the low-temperature peak and it is described by the conventional phononic thermal transport [1, 3]. The magnitude of the peak in $\kappa_c(T)$ is rather larger than that of the low-temperature peak in $\kappa_{ab}(T)$ for La214 [1]. On the other hand, the absolute value of $\kappa_c(T)$ is smaller than that of $\kappa_{ab}(T)$ for Pr214 and Gd214 [3].

The nature of thermal conductivity of the RE214 system strongly depends on the species of RE. To study deeply the mechanism of the phonon scattering in the RE214 system, we have measured both $\kappa_{ab}(T)$ and $\kappa_c(T)$ for large single crystals $RE_2CuO_4$ (RE = Pr, Nd, and Sm) grown by the traveling solvent floating zone (TSFZ) method.

 

## II. EXPERIMENT

RE214 single crystals were grown by a traveling solvent floating zone (TSFZ) method. RE214 powder was prepared by a solid state reaction method. Raw powders of $Pr_6O_{11}$ or $RE_2O_3$ (RE = Nd and Sm) and CuO were weighted with a proper molar ratio and ground. The mixed powder was calcined at 900°C for 24 h in air. A rod, which was prepared using the calcined material, was sintered at 1150–1300°C for 72 h in air. A solvent (RE : Cu = 78 : 22 in weight) was also prepared by the similar procedure. The calcination was performed at 850°C for 24 h in air, subsequently the rod was fired at 980°C for 24 h in air. The crystal growth was performed in a flowing 1–2 bar $O_2$ gas. To remove the excess oxygen, as-grown crystals were annealed for 1 week in a flowing 1 bar Ar gas.

Thermal conductivity, $\kappa$, was measured by a steady-state heat flow method. The experimental setup is schematically illustrated in the inset of Fig. 1(a). One end of the sample was adhered to the cold stage using silver epoxy. A small metal chip resistor (1 k$\Omega$) was adheard to the other end of the sample as a heater using GE7031 varnish. $\kappa$ is given as $\kappa = (Q/\Delta T) \cdot (L/S)$. $Q$ is the applied heat flow. $\Delta T$ is the temperature difference and $L$ is the distance between the two thermocouples. $S$ is the cross section of the sample. Au(0.07at%Fe)-chromel thermocouples (76 $\mu$m in diameter) were used. Susceptibility was measured using a commercial superconducting quantum interference device (SQUID) magnetometer (MPMS-XL, Quantum Design, Inc.).

## III. RESULTS AND DISCUSSION

Figure 1(a) shows the temperature dependence of the thermal conductivity in the *ab*-planes, $\kappa_{ab}(T)$, of the Pr214, Nd214, and Sm214 single crystals. $\kappa_{ab}(T)$ of Pr214 slightly increases with decreasing temperature from 300 K down to 50 K. It begins to increase rapidly below 50 K and takes a maximum of about 500 mW/cmK around 25 K. $\kappa_{ab}(T)$ of Nd214 increases moderately from 300 K down to around 250 K, subsequently decreases with the temperature. The low-temperature peak in $\kappa_{ab}(T)$ of Nd214 also appears, but its magnitude is quite smaller than that of Pr214. $\kappa_{ab}(T)$ of Sm214 is similar to that of Nd214. The high-temperature peak at 220 K for Sm214 is very pronounced and larger than the low-temperature peak. This feature is quite different from $\kappa_{ab}(T)$ reported by Berggold and co-workers [3]. Figure 1(b) shows the thermal conductivity along the *c*-axis as a function of the temperature, $\kappa_c(T)$, of the Pr214, Nd214, and Sm214 single crystals. $\kappa_c(T)$ of Pr214 and Nd214 increases moderately with decreasing temperature and shows the small peak around 20–25 K. For Sm214, the behavior of $\kappa_c(T)$ is similar to that of others above 50 K. However, $\kappa_c(T)$ begins to increase rapidly below 50 K and takes the extremely high peak of about 550 mW/cmK at 25 K.

Since the RE214 system is the antiferromagnetic insulator, the thermal carriers are generally the phonons and/or magnons. As already pointed out in previous reports [1–3], the high-temperature peak observed in $\kappa_{ab}(T)$ originates from the fact that the magnetic excitation, i.e., the magnon, contributes the thermal transport. On the other hand, the low-

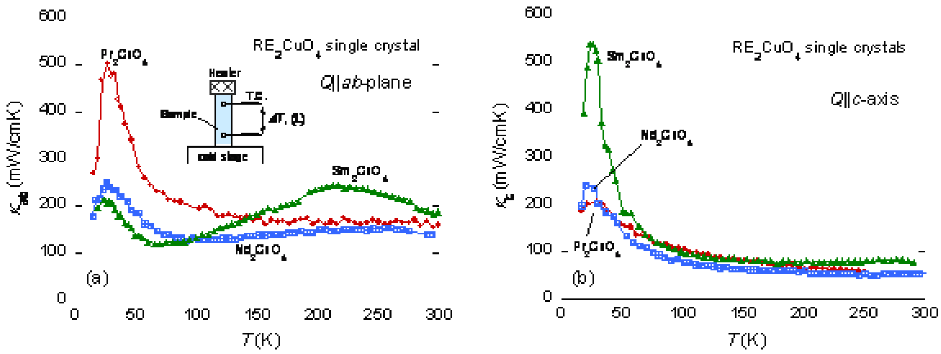


FIG. 1: Temperature dependence of the thermal conductivity (a) in the *ab*-plane and (b) along the *c*-axis, $\kappa_{ab}(T)$ and $\kappa_c(T)$, respectively, of the Pr214, Nd214, and Sm214 single crystals.

temperature peak found in both $\kappa_{ab}(T)$ and $\kappa_c(T)$ can be described by the conventional phononic thermal transport. The phonon peak is resulted from the competition between the decrease in the population of the phonons and the increase in their mean free path with decreasing temperature. In $\kappa_{ab}(T)$, the magnitude of the high-temperature peak increases, and at the same time, the low-temperature peak is suppressed. Therefore, the suppression of the low-temperature peak is caused by the fact that the magnons scatter the phonons.

Let us consider the magnitude of the magnon contribution in $\kappa_{ab}(T)$. Sachidanandam *et al.* [4] pointed out that the spin structure of Sm214 was different from that of Pr214 and Nd214 on the basis of the calculated results of the temperature dependence of the susceptibility in the *ab*-plane and along the *c*-axis, $\chi_{ab}(T)$ and $\chi_c(T)$. For Sm214, $\chi_c(T)$ is larger than $\chi_{ab}(T)$, indicating that Sm moments prefer to lie along the [001] direction. Moreover, the Sm and Cu sublattice in Sm214 are reported to be nearly decoupled [5]. On the other hand, the opposite anisotropy between $\chi_{ab}(T)$ and $\chi_c(T)$ was observed for Pr214 and Nd214, i.e., Pr and Nd moments prefer to lie along the [100] direction. Therefore, the antiferromagnetic order for Pr214 and Nd214 should be affected by the interaction between the RE and Cu moments, because the Cu spins lie in the *ab*-planes. This suppresses the magnon contribution in $\kappa_{ab}(T)$ of both Pr214 and Nd214. The fact that La is the nonmagnetic element and that Sm behaves like the nonmagnetic element in Sm214 can explain the large magnon peak in $\kappa_{ab}(T)$. Berggold *et al.* [3] pointed out that the magnon component tends to become suppressed with decreasing the Néel temperature, $T_N$. $T_N$ strongly relates with the excess oxygen. We estimated $T_N$ from the susceptibility measurement. The obtained $T_N$ values of Pr214, Nd214, and Sm214 are 325 K, 305 K, and 295 K, respectively. These $T_N$ values are larger than the reported values [3], meaning that the excess oxygen is fully removed in our RE214 single crystals. If we can ignore the difference in the species of RE, the magnitude of the magnon contribution in $\kappa_{ab}(T)$ does not relate with $T_N$. The crystallinity of our single crystals was evaluated by the full width at half maximum (FWHM) of the peak of the [004] plane by the X-ray rocking curve method. The FWHM values of Pr214, Nd214, and Sm214 are approximately 0.08°, 0.08°, and 0.06°, respectively. Therefore, the crystallinity also seems not to explain the magnitude of the high-temperature peak.

$\kappa_c(T)$ of Pr214 and Nd214 follows almost $1/T$ dependence which is typical phonon heat transport. On the other hand, $\kappa_c(T)$ of Sm214 starts to deviate the $1/T$ dependence below 50 K. Moreover, the quite large peak for Sm214 indicates that the phonon scattering centers rapidly decrease below 50 K or that the novel thermal carriers increase. Such the large peak in $\kappa_c(T)$, which is larger than the low-temperature peak of $\kappa_{ab}(T)$, was found also in La214. However, an origin of the anomalous $\kappa_c(T)$ of Sm214 remains unsolved problem.

## IV. CONCLUSIONS

We have measured the thermal conductivity in the *ab*-plane and along the *c*-axis, $\kappa_{ab}(T)$ and $\kappa_c(T)$, for large single crystals $RE_2CuO_4$ (RE214; RE = Pr, Nd, and Sm) grown by the TSFZ method. The double-peak structure was observed in $\kappa_{ab}(T)$ of all the crystals. The magnitude of high-temperature magnon peak strongly depends on the species of RE. It is very small for Pr214 and Nd214, but quite pronounced for Sm214. These behaviors can be explained by the spin structure suggested by Sachidanandam and co-workers [4]. We found an anomalous large $\kappa_c(T)$ peak for Sm214. To clarify its origin, a further study is now in progress.

### Acknowledgments

We are grateful to Dr. S. Kobayashi for his cooperation in magnetization measurements, which were performed at the Non-Destructive Evaluation and Science Research Center, Faculty of Engineering, Iwate University.

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