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Anomalies in low-temperature thermal conductivity of underdoped $La_{2-x}Sr_xCuO_4$ single crystals

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Abstract
We have measured the thermal conductivity in the *ab*-plane (κ_{ab}) as well as the *c*-axis (κ_c) of a series of underdoped $La_{2-x}Sr_xCuO_4$ (LSCO) single crystals grown by the travelling-solvent floating-zone (TSFZ) method. It was found that temperature dependence of thermal conductivity exhibits an interesting feature: there exists a steplike drop of thermal conductivity below ~10 K in both $\kappa_{ab}(T)$ and $\kappa_c(T)$, and this drop in thermal conductivity occurs at lower temperature with lower Sr concentration. Furthermore, it is found that the magnitude of this drop at $x = 0.11$ and 0.125 is larger than at other doping levels. We suspect that this phenomenon is due to an LTO (low-temperature orthorhombic) to short-range (local) or dynamical LTT-like (low-temperature tetragonal) structure transition.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Studying the structural dependence of physical properties in detail is an important approach to understand the mechanism of superconductivity in cuprates. The La-214 system is one of the simplest cuprate systems with well understood crystal structure (layered perovskite of K_2NiF_4 type), uncomplicated doping mechanisms, and simple electronic band structure. In particular, $La_{2-x}Sr_xCuO_4$ (LSCO) exhibits the full range of electronic properties that are universal for all high-T_c cuprates [1–5]. The anomalous suppression of the superconducting transition temperature T_c around 1/8 hole concentration (the so-called 1/8 problem) in La-214 is well known, but its origin has not been very clear to date. Some groups believe that it is related to the low-temperature structure phase transition from orthorhombic to tetragonal in the La-214 system [18, 19]. In fact, there are four structure phases [6] known in the La-214 series: (a) the tetragonal phase at high temperatures (the HTT phase space group $I4/mmm$), (b) the orthorhombic phase at mid-temperatures (the LTO_1 phase, $Bmab$), (c) the orthorhombic phase at low temperatures (the LTO_2 phase, $Pccn$), and (d) the tetragonal phase at low temperatures (the LTT phase, $P4_2/ncm$). All these phases can be classified with the order parameters ($Q1$, $Q2$) [7] which represent the tilt angles of the CuO_6 octahedra around [110] and [1$\bar{1}$0] axes of the HTT phase; for HTT ($Q1 = Q2 = 0$), LTO_1 ($Q1 \neq 0, Q2 = 0$), LTO_2 ($Q1 \neq Q2 \neq 0$) and LTT ($Q1 = Q2 \neq 0$). The compound of $La_{2-x}Ba_xCuO_4$ (LBCO) undergoes a two-step structure phase transition on lowering temperature [8]: from HTT to LTO_1 at the transition temperature T_{d1} and from LTO_1 to LTT at the transition temperature T_{d2}. In $La_{2-x-y}Nd_ySr_xCuO_4$ (LNSCO) the HTT, LTO_1 and LTT phases are observed; moreover, a second orthorhombic phase, LTO_2, that has never been found in LBCO, is definitely observed between the LTO_1 and LTT phases [6, 9]. Although much work has been done to investigate the low-temperature structure of LSCO, the details of the phase transition are not very clear yet. In LSCO the HTT to LTO1 transition temperature $T_{d1} \sim 250$ K ($x = 0.115$) can be determined by x-ray diffraction, but no diffraction peak of $(2h\ 0\ l)_{LTT}$ is observed, suggesting there exists no bulk

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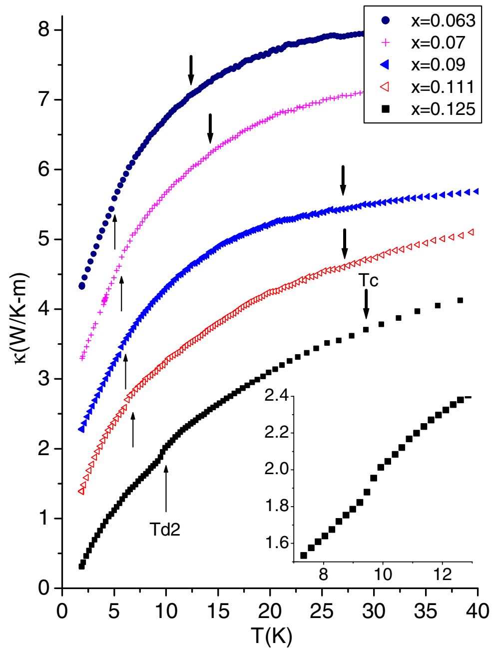

Figure 1. Temperature dependence of the ab-plane thermal conductivity $\kappa_{ab}(T)$ of underdoped $La_{2-x}Sr_xCuO_4$ ($x = 0.063, 0.07, 0.09, 0.11, 0.125$). The curves are shifted along the y axis. The up and down arrows denote the thermal conductivity steplike decrease temperature T_{d2} and the superconducting transition temperature T_c, respectively. Inset: details of $\kappa_{ab}(T)$ near T_{d2} for $x = 0.125$.

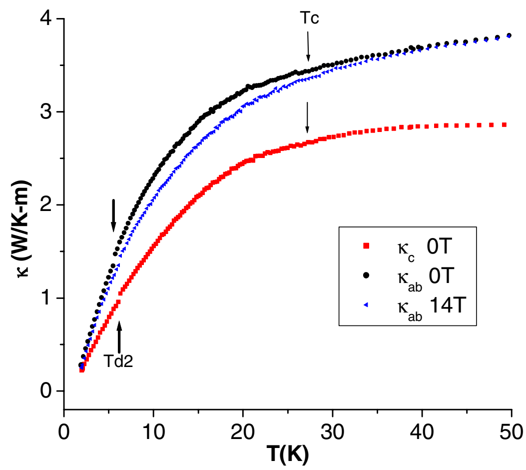

Figure 2. Temperature dependence of the thermal conductivity in $La_{2-x}Sr_xCuO_4$ of both ab-plane and c-axis for $x = 0.09$. The magnetic field are parallel to the c-axis up to 14 T. The thick and thin arrows denote T_{d2} and T_c respectively.

LTT phase down to 4.2 K. Instead, the peak positions of $(0\ 2h\ l)_{LTO1}$ and $(2h\ 0\ l)_{LTO1}$ were found to shift gradually to a higher and lower angle respectively [6]. Moodenbaugh *et al* [10] found through the measurement of high-resolution synchrotron x-ray diffraction that although there is no bulk LTT transformation in LSCO, that up to 10% LTT phase is observed at low temperatures. The results indicate that there is no long-range ordered LTT phase existing in LSCO. On the other hand, the ultrasonic studies [11] and the nuclear spin–lattice relaxation rate of ^{138}La in LSCO [12] show that there must be some kind of phase transition in LSCO in the low-temperature region below T_c.

Thermal transport measurements on cuprates can give important information about the various scattering processes of electrons and phonons, and they are sensitive to structural phase transition. Much work in heat transport has been done to investigate the structural phase transition in the La-214 system [7, 13, 14]. In $La_{1.88-x}R_xSr_{0.12}CuO_4$ (R is a rare earth ion), the structural phase transition temperature of LTO to LTT (T_{d2}) can be defined as a steplike enhancement of the thermal conductivity $\kappa(T)$ [13]. Other transport coefficients such as the dc electric conductivity and thermoelectric power cannot provide information about the scattering processes of transported carriers with the superconducting gap opening up, because they show a zero value in the superconducting state. By contrast, the thermal conductivity can still be observed even in such a state. In order to investigate the phase transition of LSCO below T_c, we have made much effort and succeeded in growing a series of large and high-quality $La_{2-x}Sr_xCuO_4$ single crystals with the travelling-solvent floating-zone (TSFZ) method. In this paper we will report the temperature dependence of both ab-plane and c-axis thermal conductivity of a series of underdoped LSCO single crystals.

2. Experiments

The single crystals of $La_{2-x}Sr_xCuO_4$ were grown by the travelling-solvent floating-zone technique. The details of the crystal growth were reported in our previous papers [5, 15]. The crystallographic orientation of the sample is determined by an x-ray Laue analysis. The crystals are cut into rectangular thin platelets of typical size $5.0 \times 1.0 \times 1.0$ mm^3, with the longitudinal direction parallel to the a-axis (in tetragonal notation) or c-axis within an accuracy of 1°. The thermal conductivity measurements were performed with the heat current flowing along the longitudinal direction of the platelets in the temperature range from 1.9 to 80 K using a physical property measurement system (PPMS, Quantum Design). Electrical contacts to the samples were made using silver epoxy cured at 450 °C in flowing O_2. The contact resistance is lower than 0.5 Ω.

3. Results and discussion

Figure 1 shows the temperature dependence of the thermal conductivity $\kappa_{ab}(T)$ in the ab-plane of $La_{2-x}Sr_xCuO_4$ ($x = 0.063, 0.07, 0.09, 0.11, 0.125$). As formerly reported by Nakamura *et al* [16] and Kudo *et al* [17], κ_{ab} decreases gradually with lowering temperature, and it exhibits a slight enhancement below the superconducting transition temperature T_c defined as the onset temperature of Meissner diamagnetism, which we have reported in our previous paper [15]. The enhancement below T_c can be observed in the $x = 0.09$, 0.11, and 0.125 samples, but it almost vanishes at lower Sr concentrations. Shown in figure 2 is the c-axis thermal conductivity $\kappa_c(T)$ in LSCO with $x = 0.09$. It exhibits almost the same magnitude of enhancement below T_c as $\kappa_{ab}(T)$. The thermal conductivity enhancement below

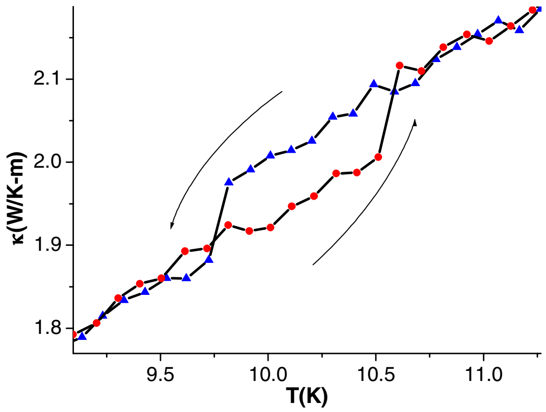

Figure 3. Thermal conductivity in *ab*-plane $\kappa_{ab}(T)$ for $x = 0.125$ near T_{d2} on cooling and heating.

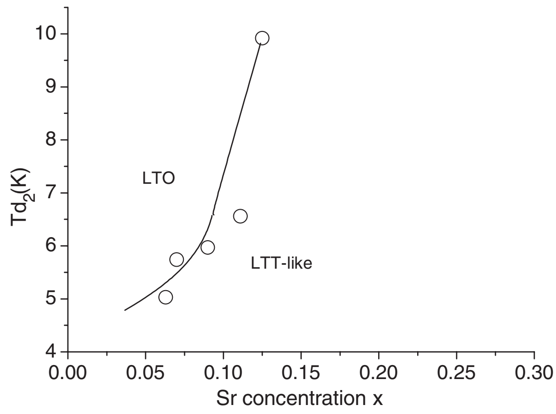

Figure 4. Phase transition temperature T_{d2} as a function of the hole concentration x. The data are derived from the thermal conductivity measurements.

T_c is a common feature of the high-T_c cuprates [16]. However, there is a long debate about its origin. One scenario attributes the slight enhancement to the phonon contribution, while in an alternative model the enhancement below T_c is ascribed to an electron contribution [16]. The measured thermal conductivity can be expressed as a sum of electron and phonon components, $\kappa = \kappa_e + \kappa_{ph}$. The electron contribution κ_e can be estimated from the electric conductivity data using the Wiedemann–Franz (WF) law, $\kappa_e = L\sigma T$, where L and σ are the Lorenz number and electrical conductivity, respectively. For our c-axis thermal conductivity data, the electron part of κ_c estimated from σ_c by the WF law is negligible (see below), and we have shown that κ_{ab} and κ_c have almost the same magnitude of enhancement below T_c. So we hold the opinion that the thermal conductivity enhancement below T_c of both $\kappa_{ab}(T)$ and $\kappa_c(T)$ in LSCO are due to the phonon contribution.

An interesting feature we found is that each underdoped sample exhibits a steplike drop in the $\kappa(T)$ curve at the temperature below $\sim$10 K, and this drop occurs at lower temperature with the lower Sr concentration, as shown in figure 1. In fact we have noted similar behaviour of $\kappa(T)$ in underdoped LSCO in the literature [7, 17]. Probably, this anomaly was so weak that no author focused on this problem. In order to investigate the details of the $\kappa(T)$ near the phase transition, we have measured the thermal conductivity very carefully using large ($5 \times 1 \times 1$ mm^3) and high-quality single crystals. The thermal conductivity is measured in a very quiet environment without any heat disturbance. The temperature is lowered steadily at a rate as slow as 0.1 K min^{-1}. The steplike anomaly of $\kappa(T)$ we found is an interesting behaviour, reminiscent of the ultrasonic studies of $La_{2-x}M_xCuO_4$ (M = Sr, Ba) by Fukase *et al* [11]. They found a low-temperature transformation from the orthorhombic phase to the tetragonal phase with lattice stiffening and an attenuation peak at T_{d2} in $La_{2-x}Ba_xCuO_4$ ($x = 0.10$–0.12). They also observed partial lattice stiffening and a small attenuation peak in LSCO ($x = 0.10$–0.12) at $\sim$10 K below T_c. Furthermore, in LSCO ($x = 0.12$) Goto *et al* [12] found the nuclear spin–lattice relaxation rate T_1 of ^{138}La-NQR to exhibit a significant maximum at the temperature below $\sim$10 K. These results indicate that there exists some kind of phase transition below $\sim$10 K in LSCO. Interestingly, all the above phenomena, including the drop in thermal conductivity, occur at exactly the same temperature region. We therefore reasonably think that they are of the same origin. Accordingly, the thermal conductivity drop position in $La_{2-x}Sr_xCuO_4$ may be taken as the LTO to LTT-like phase transition temperature T_{d2}. Shown in figure 4 is the low-temperature structure phase diagram thus derived from the thermal conductivity data. It is found that the phase diagram of LSCO is similar to LBCO but shifting to lower temperature region [21]. In LBCO, the LTO to LTT transition temperature T_{d2} is $\sim$60 K, which is higher than the superconducting transition temperature T_c, but in LSCO, T_{d2} is below $\sim$10 K, which is lower than T_c. This may be the reason why a number of drastic anomalies such as reduction of T_c, anomalous Seebeck coefficients and Hall coefficients appear obviously near $x = 0.125$ in LBCO but are weaker or absent in LSCO ($x = 0.11, 0.125$) [21].

In a further careful measurement of the cooling and heating curve of $\kappa_{ab}(T)$ in the area of the drop, a weak hysteresis is observed as shown in figure 3. This is consistent with the results of the ultrasonic experiments, where a narrow region of hysteresis was observed near the phase transition at the same temperature region as in figure 3. It was also reported that the nuclear spin lattice relaxation rate T_1 of ^{138}La-NQR in LSCO ($x = 0.12$) exhibits a diverging behaviour near the phase transition [12, 20].

Now let us discuss the possible mechanism of the steplike decrease of thermal conductivity. We note that both $\kappa_{ab}(T)$ and $\kappa_c(T)$ exhibit the steplike decrease at the same temperature ($\sim$5.7 K) in LSCO with $x = 0.09$ as shown in figure 2. The anisotropy ratio ρ_c/ρ_{ab} of LSCO is about 500–1000. Estimated by the WF law, the electron contribution κ_{abe} is about 500–1000 times larger than the κ_{ce}. So the electron contribution to the c-axis thermal conductivity $\kappa_c(T)$ is negligible. Considering that both $\kappa_{ab}(T)$ and $\kappa_c(T)$ exhibit almost the same magnitude of the decrease, we can conclude that the drop in thermal conductivity of both the ab-plane and the c-axis probably totally result from the phonon contribution. On the other hand, we have also measured $\kappa_{ab}(T)$ under magnetic field up to 14 T as shown in figure 2. Although the thermal conductivity is suppressed by application of the magnetic field, which is consistent with previous results of Kudo *et al* [17], it is found that the drop in $\kappa_{ab}(T)$ still exists and its magnitude does not change much, only its position shifts to a slightly higher temperature under the magnetic field up to 14 T. As we know, the electron contribution κ_e to the thermal conductivity is suppressed greatly by magnetic field as high as 14 T [17, 23]. Therefore this is further evidence for the phonon origin of the thermal conductivity drop. The phonon contribution to the measured thermal conductivity can be expressed as $\kappa_{ph} = C_{ph} v_s \lambda_{ph}/3$. Here C_{ph}, v_s, and λ_{ph} are the specific heat, the sound velocity, and the mean free path of phonons, respectively. Baberski *et al* [19] found that the phonon contribution κ_{ph} of thermal conductivity exhibits a step enhancement at the LTO to LTT phase transition only if the LTT phase is not superconducting in the LNSCO. The enhancement of κ_{ph} may be attributed to the increase of sound velocity v_s in the LTT phase. However, in the superconducting composition range of the LTT phase of LNSCO, no such enhancement was observed at the phase transition. They thought that in the superconducting LTT region the dynamics of charge order correlations might couple to the lattice causing a pronounced damping of the phonon heat transport [19]. In the case of LSCO without rare earth doping, no bulk LTO to LTT phase transition is observed but up to 10% LTT phase exists at low temperature. We suspect that, if the local (short-ranged) or dynamical LTT-like tilts of CuO_6 octahedra develop in LSCO, the dynamics of charge order correlations will couple to the lattice to result in a similar damping of the phonon heat transport just like the situation in the superconducting LNSCO. Furthermore, the phonons will be strongly scattered by the short-range or dynamical lattice distortion due to the development of local LTT-like tilts. As a result, the mean free path of the phonons will decrease abruptly near the phase transition. Note that κ_{ph} exhibits a steplike decrease although there is partial lattice stiffening (v_s increase) below ~10 K in the LTT-like phase of LSCO. We thus come to a conclusion that the observed drop in κ_{ph} originates from the abrupt reduction of the mean free path λ_{ph}, that carries more weight than the gradual increase of sound velocity v_s.

In addition, the thermal conductivity anomaly was also found at lower Sr concentrations of $x = 0.063$ and 0.07, but the drop is smaller than that near $x = 0.11$ and 0.125, as shown in figure 1. The superconducting transition temperature T_c as a function of the hole concentration x exhibits a plateau near the Sr concentration with $x = 0.11$ and 0.125, which we have reported in a previous paper [5]. We note that the thermal conductivity 'step' decrease $\delta\kappa$ increases with the emergence of the T_c plateau. The results strongly indicate the correlation between T_c reduction and the thermal conductivity anomaly around $x = 0.11$, 0.125 due to the LTO to LTT-like phase transition. $x = 0.11$, 0.125 may be the *magic* hole concentration [5, 15, 22] at which more of the crystal favours the LTT phase. We also found that it exhibits an anomaly with $x = 0.11$ in the $\delta\kappa/x$, $v_s x$ curve ('x' is Sr concentration). We suspect that this anomaly is related to the model of charge ordering with $x = 0.11$ [24, 5]. Further work on this problem is underway.

4. Conclusion

In summary, we have presented experimental results on the thermal conductivity of a series of underdoped LSCO single crystals. The temperature dependence of both *ab*-plane and *c*-axis thermal conductivity exhibits a steplike decrease at ~10 K below T_c. We suggest that some kind of phase transition exists at ~10 K. Although the origin of the phase transition is not very clear, comparison of the thermal conductivity data with the ultrasonic studies and ^{138}La-NQR of LSCO leads to the conjecture that a short-range (local) or dynamical LTT (LTT-like) phase develops in LSCO below ~10 K. Furthermore, it is found that the thermal conductivity steplike drop occurs at lower temperature with lower Sr concentration, and the magnitudes of the 'step' decrease for $x = 0.11$ and 0.125 are larger than the other Sr concentrations. Then it is easy to picture the structure phase diagram of underdoped LSCO derived from the thermal conductivity data.

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