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Anomalies in low-temperature thermal conductivity of underdoped La\(_{2-x}\)Sr\(_x\)CuO\(_4\) single crystals

Jun Zhao\(^1\), Cai-Xia Shen, Fan Zhou, Ji-Wu Xiong, Li-Hua Liu and Zhong-Xian Zhao

National Laboratory for Superconductivity, Institute of Physics, Chinese Academy of Sciences, PO Box 603, Beijing 100080, People’s Republic of China

E-mail: zj@ssc.iphy.ac.cn

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Abstract

We have measured the thermal conductivity in the \(ab\)-plane (\(\kappa_{ab}\)) as well as the \(c\)-axis (\(\kappa_c\)) of a series of underdoped La\(_{2-x}\)Sr\(_x\)CuO\(_4\) (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 \(\sim 10\) 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\(_2\)NiF\(_4\) type), uncomplicated doping mechanisms, and simple electronic band structure. In particular, La\(_{2-x}\)Sr\(_x\)CuO\(_4\) (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, \(P-42/nmc\)). All these phases can be classified with the order parameters \((Q_1, Q_2)\) \([7]\) which represent the tilt angles of the CuO\(_6\) octahedra around \([110]\) and \([1\overline{1}0]\) axes of the HTT phase; for HTT \((Q_1 = Q_2 = 0)\), LTO\(_1\) \((Q_1 \neq 0, Q_2 = 0)\), LTO\(_2\) \((Q_1 \neq Q_2 \neq 0)\) and LTT \((Q_1 = Q_2 = 0)\). The compound of La\(_{2-x}\)Ba\(_x\)CuO\(_4\) (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\(_y\)Sr\(_x\)CuO\(_4\) (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\(_2\) 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 LTO\(_1\) 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
LTT phase down to 4.2 K. Instead, the peak positions of (0 2h 1)LT0 and (2h 0 1)LT0 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 $^{139}$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$_{2-x}$Sr$_x$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$_x$CuO$_4$ 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$_x$CuO$_4$ 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$_x$CuO$_4$ (x = 0.063, 0.07, 0.09, 0.11, 0.125). As formerly reported by Nakamura et al. [16] and Kudo et al. [17], $\kappa_{ab}$ 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
\( T_c \) 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, \( \kappa = \kappa_e + \kappa_p \). The electron contribution \( \kappa_e \) can be estimated from the electric conductivity data using the Lorenz number and electrical conductivity, respectively.

\[ \kappa_e \text{ estimated from } \sigma_{ab} \text{ and } L \]

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 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\(_x\)CuO\(_4\) (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\(_x\)CuO\(_4\) (\( x = 0.10–0.12 \)) 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\(_x\)CuO\(_4\) 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 \( \rho_c/\rho_{ab} \) of LSCO is about 500–1000. Estimated by the WF law, the electron contribution \( \kappa_{abe} \) is about 500–1000 times larger than the \( \kappa_{c,e} \). 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 $\kappa_e$ 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 $\kappa_{ph} = C_{ph} v_s \rho_{ph}/3$. Here $C_{ph}$, $v_s$, and $\rho_{ph}$ are the specific heat, the sound velocity, and the mean free path of phonons, respectively. Baberski et al [19] found that the phonon contribution $\kappa_{ph}$ 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 $\kappa_{ph}$ 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 $\kappa_{ph}$ exhibits a steplike decrease although there is partial lattice stiffening ($v_s$ increase) below \( \sim 10 \) K in the LTT-like phase of LSCO. We thus come to a conclusion that the observed drop in $\kappa_{ph}$ originates from the abrupt reduction of the mean free path $\rho_{ph}$ 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 $\delta\kappa/x, v_s$ 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 \( \sim 10 \) K below $T_c$. We suggest that some kind of phase transition exists at \( \sim 10 \) K. Although the origin of the phase transition is not very clear, comparison of the thermal conductivity data with the ultrasonic studies and $^{139}$La-NQR of LSCO leads to the conjecture that a short-range (local) or dynamical LTT (LTT-like) phase develops in LSCO below \( \sim 10 \) 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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