Thermal conductivity of lightly Sr- and Zn-doped La_2CuO_4 single crystals

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Both ab-plane and c-axis thermal conductivities (κ_{ab} and κ_c) of lightly doped $La_{2-x}Sr_xCuO_4$ (LSCO) and $La_2Cu_{1-y}Zn_yO_4$ single crystals (x or $y=0$–0.04) are measured from 2 to 300 K. It is found that the low-temperature phonon peak (at 20–25 K) is significantly suppressed upon Sr or Zn doping even at very low doping, though its precise doping dependences show interesting differences between the Sr and Zn dopants, or between the ab plane and the c axis. Most notably, the phonon peak in κ_c decreases much more quickly with Sr doping than with Zn doping, while the phonon-peak suppression in κ_{ab} shows an opposite trend. It is discussed that the scattering of phonons by stripes is playing an important role in the damping of the phonon heat transport in lightly doped LSCO, in which static spin stripes have been observed by neutron scattering. We also show κ_{ab} and κ_c data of $La_{1.28}Nd_{0.6}Sr_{0.12}CuO_4$ and $La_{1.68}Eu_{0.2}Sr_{0.12}CuO_4$ single crystals to compare with the data of the lightly doped crystals for the discussion of the role of stripes. At high temperature, the magnon peak (i.e., the peak caused by the spin heat transport near the Néel temperature) in $\kappa_{ab}(T)$ is found to be rather robust against Zn doping, while it completely disappears with only 1% of Sr doping.

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I. INTRODUCTION

It has recently been discussed that holes in high-T_c cuprates self-organize into quasi-one-dimensional stripes.[1–17] The stripe phase is a periodic distribution of antiferromagnetically ordered spin regions separated by quasi-one-dimensional charged domain walls which act as magnetic antiphase boundaries. Although the relation between the stripe correlations and the mechanism of high-T_c superconductivity is not fully understood yet, it has become clear[10,17] that the charge stripes determine the charge transport behavior, at least in the lightly hole doped region: charges can move more easily along the stripes than across the stripes.[17] Given that the stripes indeed affect the basic physical properties such as charge transport, it is desirable to build a comprehensive picture of the roles of stripes in the cuprates. Since the nonuniform charge distribution is expected to induce variations of the local crystal structure,[1,4] which disturb phonons, the phonon heat transport is expected to be a good tool capable of detecting the influence of stripes even in the charge-localized region.

Thermal conductivity is one of the basic transport properties that provides a wealth of useful information on the charge carriers and phonons, as well as their scattering processes. It is known that the antiferromagnetic (AF) insulating compound La_2CuO_4 shows predominant phonon transport at low temperatures, which is manifested in a large phonon peak at 20–25 K in the temperature dependence of both ab-plane and c-axis thermal conductivities (κ_{ab} and κ_c);[18] such phonon peak disappears in Sr-doped $La_{2-x}Sr_xCuO_4$ (LSCO) with $x=0.10$–0.20.[18] The suppression of the phonon peak is normally caused by the defect scattering and the electron scattering of phonons in doped single crystals; however, it is also known that the phonon peak re-appears in both $\kappa_{ab}(T)$ and $\kappa_c(T)$ of overdoped $La_{1.7}Sr_{0.3}CuO_4$,[18] which cannot be explained in this scenario. Furthermore, it was found that in rare earth (RE) and Sr co-doped La_2CuO_4, such as $La_{1.28}Nd_{0.6}Sr_{0.12}CuO_4$, the phonon thermal conductivity is much more enhanced in the nonsuperconducting low-temperature tetragonal phase, compared to that in LSCO with the same Sr content.[19] This cannot happen if the defect scattering and electron scattering of phonons are the only source of the peak suppression. Based on the fact that in RE- and Sr-doped La_2CuO_4 systems the phononic thermal conductivity is always strongly suppressed in superconducting samples,[19] it was proposed that dynamical stripes cause a pronounced damping of phonon heat transport, while static stripes do not suppress the phonon transport so significantly.[19] It is indeed possible that the static stripes are not effective in scattering phonons, if they only induce *periodic* local distortions in the crystal structure. Since the spin stripes in lightly doped LSCO ($x=0.01$–0.05) are reported to be static,[5–8] one may naively expect that the phonon heat transport is not strongly suppressed in such lightly doped LSCO. However, there has been no measurement of the thermal conductivity of lightly doped LSCO single crystals.

In this paper, we report our study of the thermal conductivity of lightly doped $La_{2-x}Sr_xCuO_4$ ($x=0$–0.04) single crystals. Such low doping levels are intentionally selected for the reasons of both tracing the evolution of the phonon peak and avoiding significant modifications of the crystal structure and phonon mode, which may complicate interpretations of the data. It is found that the phonon peak is suppressed significantly even with very small doping concentration, especially for the c-axis heat transport. Since this result is contrary to the naive expectation mentioned in the previous paragraph, we also study the thermal conductivity of $La_2Cu_{1-y}Zn_yO_4$ (LCZO) ($y=0$–0.04) single crystals, which have similar amount of dopants as LSCO. The important difference between these two systems is that there cannot be stripes in LCZO (since there is no carrier), while there are static spin stripes in LSCO at low temperatures. By comparing the thermal conductivity behaviors in these two systems, we can elucidate whether the static stripes are responsible for strong scattering of phonons. The comparison indicates that the stripes, though they are static, indeed damp the c-axis

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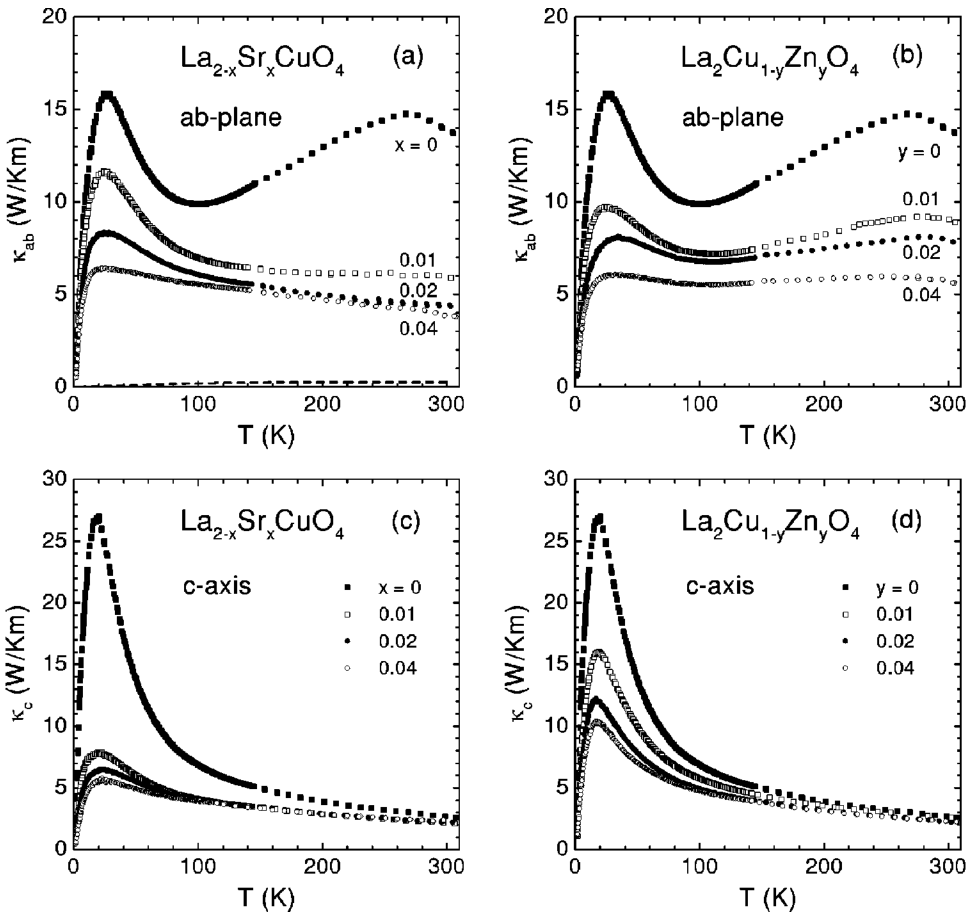

FIG. 1. Thermal conductivity of lightly doped $La_{2-x}Sr_xCuO_4$ and $La_2Cu_{1-y}Zn_yO_4$ single crystals along the ab plane and the c axis. The dashed line in panel (a) is $\kappa_{e,ab}$ of $La_{1.96}Sr_{0.04}CuO_4$ estimated from the Wiedemann-Franz law.

phonon transport significantly, while their role is minor in the in-plane phonon transport.

II. EXPERIMENTS

The single crystals of LSCO and LCZO are grown by the traveling-solvent floating-zone technique, and carefully annealed in flowing pure He gas to remove the excess oxygen.[20] After the crystallographic axes are determined by using an x-ray Laue analysis, the crystals are cut into rectangular thin platelets with the typical sizes of $2.5\times0.5\times0.15$ mm^3, where the c axis is perpendicular or parallel to the platelet with an accuracy of $1°$. The thermal conductivity κ is measured in the temperature range of 2–300 K using a steady-state technique;[21–23] above 150 K, a double thermal shielding is employed to minimize the heat loss due to radiation, and the residual radiation loss is corrected by using an elaborate measurement configuration. The temperature difference ΔT in the sample is measured by a differential Chromel-Constantan thermocouple, which is glued to the sample using GE vanish. The ΔT varies between 0.5% and 2% of the sample temperature. To improve the accuracy of the measurement at low temperatures, κ is also measured with "one heater, two thermometer" method from 2 to 20 K by using a chip heater and two Cernox chip sensors.[21] The errors in the thermal conductivity data are smaller than 10%, which are mainly caused by the uncertainties in the geometrical factors. Magnetization measurements are carried out using a Quantum Design superconducting quantum interference device magnetometer.

III. RESULTS

A. Anisotropic heat transport in La_2CuO_4

It is useful to first establish an understanding of the anisotropic heat transport in undoped crystals. The temperature dependences of the thermal conductivity measured along the ab plane and the c axis in pure La_2CuO_4 are shown in Fig. 1, which also contains data for LSCO and LCZO single crystals. For the undoped La_2CuO_4 sample, a sharp peak appears at low temperature, at $\sim$25 K in $\kappa_{ab}(T)$ and at $\sim$20 K in $\kappa_c(T)$, respectively. In the c direction, above the peak temperature, κ_c decreases with increasing T approximately following $1/T$ dependence, which is typical for phonon heat transport.[24] This phonon peak originates from the competition between the increase in the population of phonons and the decrease in their mean free path (due to the phonon-phonon umklapp scattering) with increasing temperature. In contrast to the mainly phononic heat transport in the c direction, in the ab plane another large and broad peak develops at higher temperature ($\sim$270 K), which has been

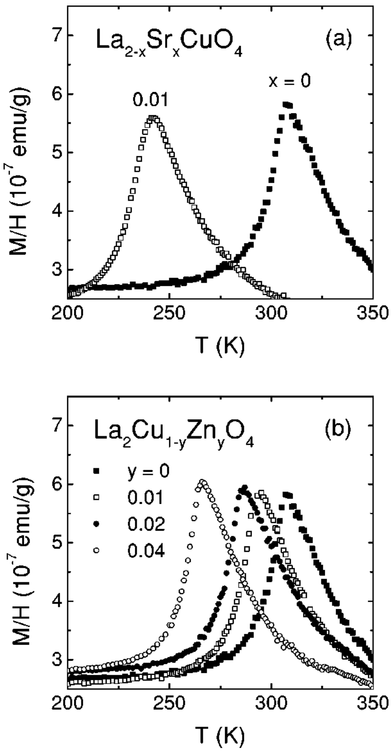

FIG. 2. Magnetic susceptibility of (a) $La_{2-x}Sr_xCuO_4$ and (b) $La_2Cu_{1-y}Zn_yO_4$ single crystals measured in a 0.5-T field applied along the c axis.

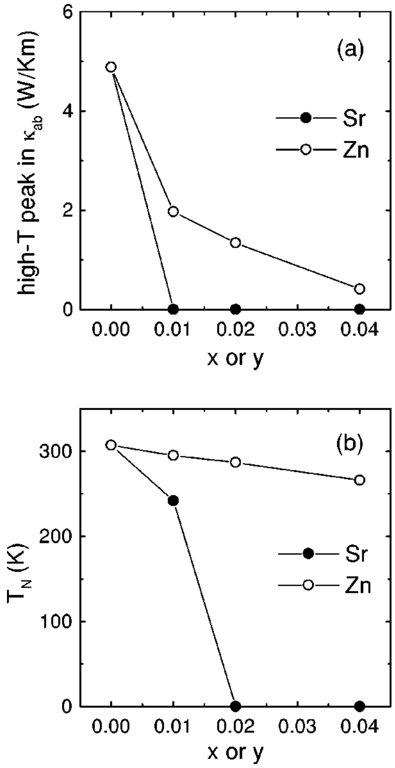

FIG. 3. (a) Doping dependences of the size of high-T peak in $\kappa_{ab}(T)$ [defined by the difference between the peak value and κ_{ab}(100 K)] of $La_{2-x}Sr_xCuO_4$ and $La_2Cu_{1-y}Zn_yO_4$ single crystals. (b) Doping dependences of the Néel temperature T_N for the two cases.

attributed[18] to the magnon transport in a long-range-ordered AF state. The heat conduction due to magnetic excitations has been observed in many low-dimensional quantum antiferromagnets, such as one-dimensional spin systems $CuGeO_3$,[21,23,25] Sr_2CuO_3,[26] $SrCuO_2$,[27] and $(Sr,Ca)_{14}Cu_{24}O_{41}$,[28] and also the two-dimensional antiferromagnet $K_2V_3O_8$.[29] The absence of this high-T peak in $\kappa_c(T)$ is obviously due to the much weaker magnetic correlations in the c direction compared to that in the CuO_2 plane. All these behaviors in undoped La_2CuO_4 are consistent with the previously reported data.[18] It is worthwhile to note that the phononic peak value of κ_c (27 W/Km) is considerably larger than that of κ_{ab} (16 W/Km). One possible reason for this difference is that the phonon heat transport is intrinsically easier along the c axis, which is not so easy to conceive in view of the layered crystal structure of La_2CuO_4. Another, more likely, possibility is that there exists phonon-magnon scattering in the ab plane that causes additional damping of the phonon peak. Such scattering may also happen in the c axis, however, it should be much weaker than in the ab plane because it is well known that the magnons are good excitations only for the in-plane physics in the La_2CuO_4 system.

B. Doping dependence of magnon peak and Néel transition

Upon Sr or Zn doping, there are strong doping dependences in both κ_{ab} and κ_c. Let us first discuss the change of the high-T magnon peak in $\kappa_{ab}(T)$ for different dopants. This peak is suppressed very quickly upon Sr-doping and in fact completely disappears at only 1% doping concentration. On the other hand, the suppression of this peak is much slower in the Zn-doped case, where the peak, though gradually suppressed, survives to $y=0.04$. To clarify the relation between the high-T peak and the Néel order, the magnetic susceptibility of LSCO and LCZO is measured from 5 to 350 K in the magnetic field of 0.5 T applied along the c axis. Figure 2 shows the temperature dependences of the magnetic susceptibility, where the peak corresponds to the Néel transition (no transition is observed in LSCO with $x \geqslant 0.02$). It is clear (as has been already reported[30,31]) that Sr doping is much stronger than Zn doping in destroying the AF long-range order, where the former decreases the Néel temperature T_N much more quickly. The reason is related to the fact that holes introduced by Sr are mobile and have a 1/2 spin, while Zn (zero spin) causes a static spin vacancy in the CuO_2 plane. Figure 3 summarizes the doping dependences of the size of the high-T peak in $\kappa_{ab}(T)$ [defined by the difference between the peak value and κ_{ab}(100 K)], as well as those of the Néel temperature T_N, for the two systems. This result confirms that the magnon peak is quickly diminished as T_N is reduced. It is useful to note that the disorder in the Néel state induced by Sr doping appears to be detrimental to the magnon heat transport, because the magnon peak completely disappears at $x=0.01$ even though the Néel order is still established at 240 K for this Sr doping.

C. Doping dependence of phonon peak

More interesting changes happen in the suppression of the phonon peak at low temperatures. One can see in Fig. 1 that

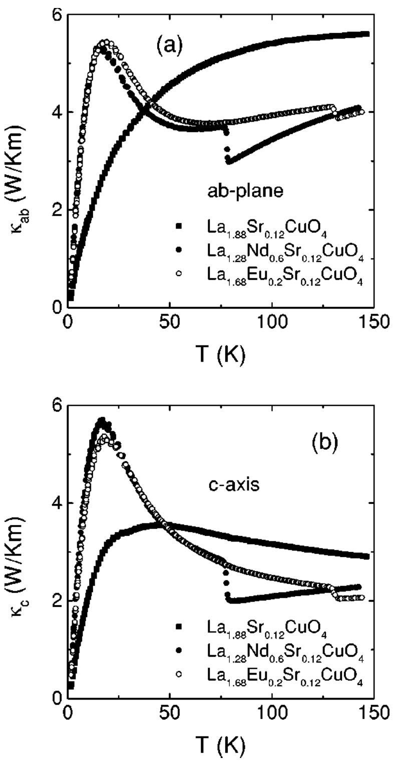

FIG. 4. Comparison of the low-temperature thermal conductivity of LSCO ($x=0.12$) single crystals to that of Nd- and Eu-doped crystals, in which the phonon peak reappears. The data for both (a) $\kappa_{ab}(T)$ and (b) $\kappa_c(T)$ are shown.

in $\kappa_{ab}(T)$ the peak magnitude decreases more quickly with Zn doping than with Sr doping; on the contrary, the peak value in κ_c decreases much more quickly with Sr doping than with Zn doping. It should be noted that those peculiar differences between Sr and Zn dopings cannot be due to the additional electronic thermal conductivity in the Sr-doped samples: The electronic thermal conductivity κ_e can roughly be estimated by the Wiedemann-Franz law $\kappa_e = L_0 T/\rho$, where ρ is the electrical resistivity and L_0 is the Lorenz number (which can be approximated by the Sommerfeld value, 2.44×10^{-8} WΩ/K^2). Using the resistivity data for our crystals,[32] the contribution $\kappa_{e,ab}$ for $La_{1.96}Sr_{0.04}CuO_4$ can be estimated to be smaller than 0.25 W/Km in the whole temperature region [dashed line in Fig. 1(a)], and samples with lower Sr doping should have smaller $\kappa_{e,ab}$ than this estimate for $x=0.04$; clearly, the electronic contribution can be safely neglected in the lightly Sr doped region in the discussion of the $\kappa_{ab}(T)$ behavior, not to mention the $\kappa_c(T)$ behavior. Thus, the changes in the low-temperature peak upon doping must be due to the changes in the phonon transport properties, that is, the scattering process that determines the phonon mean free path.

D. Reappearance of the phonon peak in RE-doped LSCO

Although both the phonon-impurity scattering and the phonon-carrier scattering can contribute to the suppression of the phonon peak, these scattering processes cannot be the only mechanisms to determine the Sr-doping dependence of the phonon heat transport, because the phonon peak re-appears in overdoped LSCO and in RE-doped LSCO.[18,19] Figure 4 shows our data for the re-appearance of the phonon peak in RE-doped samples (both Nd- and Eu-doped cases) for $x=0.12$; these data are taken on single crystals, and essentially confirms the polycrystalline data reported by Baberski *et al.*[19] The single crystal data of Fig. 4 allow us to compare the absolute values of κ_{ab} and κ_c of the RE-doped samples to those of the RE-free LSCO; such a comparison tells us that the low-T peak values of κ_{ab} and κ_c of the RE-doped crystals at $x=0.12$ are similar to those of the RE-free LSCO crystals at $x=0.04$, despite the factor of 3 difference in the Sr concentrations. This result clearly indicates that the phonon-impurity scattering and the phonon-carrier scattering are not the only scattering mechanisms to determine the phonon mean free path.

IV. DISCUSSION

A. Magnetic scattering of phonons in the *ab* plane of LCZO

Based on the apparent similarity of the doping dependence of the phonon peak in κ_{ab}, shown in Figs. 1(a) and 1(b), one might naively conclude that the lattice disorders induced by Zn and Sr in the *ab* plane are very similar and that the $\kappa_{ab}(T)$ behavior is essentially explained only by the lattice disorder; however, this is not likely to be the case, which can be understood by considering the nature of the impurity scattering and the additional charge carriers introduced in LSCO. First, it is important to notice that the low-T phonon peak in κ_{ab} is suppressed slightly more quickly by Zn doping than by Sr doping. In the impurity-scattering scenario,[24] it is difficult to conceive that the phonons are scattered more strongly in LCZO than in LSCO, because the atomic mass difference between Cu and Zn is much smaller than that between La and Sr. In addition, the contribution of the phonon-carrier scattering, which exists only in LSCO, would cause the phonon peak in LSCO to be suppressed more quickly than in LCZO. Thus the difference between the Sr-doping dependence and the Zn-doping dependence in $\kappa_{ab}(T)$ at low temperature, which is opposite to the naturally expected trend, suggests that there are additional scatterers of phonons in the Zn doped samples. It is most likely that the additional scatterers in LCZO are of magnetic origin; magnons, which cause the high-temperature peak in LCZO, are an obvious candidate, and the magnetic disordering around Zn atoms may also cause some scattering of phonons through magnetoelastic coupling.[33] It is useful to note that, as we mentioned in Sec. III A, magnons are likely to be already responsible for the relatively small phonon peak in κ_{ab} (compared to that in κ_c) in pure La_2CuO_4.

B. Scattering of phonons by static spin stripes in LSCO

Contrary to the relatively small difference in the phonon peak of κ_{ab} between LSCO and LCZO, the suppression of the phonon peak of κ_c is much quicker in LSCO than in LCZO; in particular, the sharp suppression of the phonon peak from $x=0$ to 0.01 is quite surprising and is the most striking observation of this work. Apart from the impurity scattering, two different scattering processes of phonons are possibly responsible for the suppression of the *c*-axis phonon heat transport. The first possibility is the magnon-phonon scattering. This may partly play a role (particularly in samples where the long-range Néel order is established) in the *c*-axis phonon heat transport, although their role is expected to be minor because of the essentially two-dimensional nature of the magnons in the LCO system, which means that the magnons cannot effectively change the wave vector of the *c*-axis phonons. The second possibility is the scattering of phonons caused by the static stripes that exist only in LSCO. In the following, we elaborate on this possibility.

It is known that static spin stripes are formed at low temperatures in lightly doped LSCO.[5–8] If the charges also form static stripes together with the spins, they certainly cause local lattice distortions. Even if the charges do not conform to the static stripe potentials set by the spins, the spin stripes themselves may well cause local lattice distortions due to the magnetoelastic coupling;[33] this direct coupling between the spin stripes and the local lattice distortions is in fact very likely, because recently Lavrov *et al.* found that the spin-lattice coupling is very strong in lightly doped LSCO.[34] It should be noted that local lattice distortions due to static stripes are *not* expected to scatter phonons when the stripes induce only static and *periodic* modulation of the lattice (which is in fact a kind of superlattice); however, possible disordering of stripes in the *c* direction can introduce rather strong scattering of phonons in this direction. In fact, because of the weak magnetic correlations along the *c* axis, the stripes in neighboring CuO_2 planes are only weakly correlated or even uncorrelated, which is best evidenced by the very short magnetic correlation length ξ_c in the stripe phase (usually smaller than the distance between neighboring CuO_2 planes) in the lightly doped LSCO.[6,7]

We can further discuss the possible role of static stripes in the suppression of the phonon peak in κ_{ab} of lightly doped LSCO. Although the stripes are much better ordered in the ab plane than in the *c* direction,[6,7] there are two reasons that phonons in the ab plane are possibly scattered by the stripes. First, the static spin stripes were reported to be established at about 30–17 K for $x=0.01$–0.04 by neutron measurements.[5,7] These temperatures are very close to the position of the phonon peak (20–25 K). Near the stripe freezing temperature, local lattice distortions are expected to be well developed and yet are slowly fluctuating (or disordered), which would tend to scatter phonons. Second, there is no evidence until today that well periodically ordered static charge stripes are formed in lightly doped LSCO; instead, our charge transport data strongly suggest[17,32,35] that charge stripes exist in a liquid (or nematic) state.[14] Such disordered state of charge stripes would tend to scatter phonons. Thus, it is possible that the static stripes also contribute to the scattering of phonons in the in-plane heat transport in LSCO, although their effect appears to be much weaker than that in the *c* direction.

V. SUMMARY

We have measured the ab-plane and *c*-axis thermal conductivities of lightly doped LSCO and LCZO single crystals. It is found that the low-temperature phonon peak is significantly suppressed upon Sr or Zn doping even at very low doping levels, and that the doping dependences show clear differences between the Sr and Zn dopants, and between κ_{ab} and κ_c. The experimental observations can be summarized as follows: (i) The phonon peak in κ_c decreases much more quickly with Sr doping than with Zn doping. (ii) On the other hand, the phonon peak in κ_{ab} is suppressed slightly more quickly with Zn doping than with Sr doping. (iii) At high temperature, the magnon peak in $\kappa_{ab}(T)$ decreases much more quickly with Sr doping than with Zn doping; in fact, the magnon peak completely disappears in LSCO with $x=0.01$, while it is still observable in LCZO with $y=0.04$. (iv) Rare-earth (Nd or Eu) doping for LSCO ($x=0.12$) enhances the phonon heat transport in both κ_{ab} and κ_c, and this is manifested in the reappearance of the low temperature phonon peak, whose height is similar to that of rare-earth-free LSCO with $x=0.04$.

Based on the peculiar doping dependences of the low-temperature phonon peak, we can deduce the following conclusions for the phonon heat transport at low temperatures: (i) In the in-plane direction, disordered static stripes are probably working as scatterers of phonon (in addition to the Sr impurities and the holes) in LSCO, while in LCZO magnetic scatterings cause strong damping of the phonon transport, which overcompensates the absence of the stripe scattering. (ii) In the *c*-axis heat transport, besides the possible contribution of the magnon-phonon scattering, the strong disorder of the stripe arrangement along the *c* axis is mainly responsible for the scattering of phonons. The latter scattering mechanism naturally explains the strong damping of the phonon peak in LSCO, while allowing a much slower suppression of the phonon peak in LCZO where there is no stripe. Therefore, the effect of static spin stripes, which are formed in the lightly doped LSCO, appears to be most dramatically observed in the *c*-axis phonon heat transport.

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