

# Thermal conductivity in the stripe-ordered phase of cuprates and nickelates

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The temperature dependence of the thermal conductivity  $\kappa$  and the resistivity  $\rho$  have been measured on well-characterized crystals  $\text{La}_{1.60-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  ( $x=0.12, 0.15$ , and  $0.20$ ),  $\text{La}_2\text{CuO}_{4+\delta}$ , and  $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$  ( $x=0, 0.33, 0.50$ ). In  $\text{La}_2\text{CuO}_{4+\delta}$ , the phonon component of  $\kappa(T)$  decreases sharply with increasing  $\delta$ . The charge ordering at  $T_{\text{CO}}$  in  $\text{La}_{1.67}\text{Sr}_{0.33}\text{NiO}_4$  enhances  $\kappa(T)$ . Similarly in the  $\text{La}_{1.60-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  system, ordering of the mobile holes into stripes within the  $\text{CuO}_2$  sheets on cooling through a low-temperature tetragonal/orthorhombic (LTT/LTO) transition at  $T_t$  gives a jump  $\Delta\kappa$  in  $\kappa(T)$ . In contrast to a previous report on ceramic samples, the superconductive crystals  $x=0.15$  and  $0.20$  of the  $\text{La}_{1.60-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  system show a clear low-temperature peak in the normal state  $\kappa(T)$  below  $T_t$  that shows phonons are the dominant carriers of heat in the LTT phase. Restoring phonons in the LTT phase is thus seen to be compatible with superconductivity in the copper oxides, albeit with a lower  $T_c$  than is found in the LTO phase where the phonon component of  $\kappa(T)$  is suppressed in the normal state by the bond-length fluctuations.

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## INTRODUCTION

High-temperature superconductivity has been found in the  $p$ -type cuprates in a narrow range of hole concentrations centered near  $p \approx 1/6$  at a crossover from localized to itinerant electronic behavior in the normal state. An electronic inhomogeneity characterized by dynamic, local structural distortions,<sup>1</sup> phase separation,<sup>2</sup> and a spatial distribution of the superconductive gap<sup>3</sup> has been interpreted to be an intrinsic feature of the crossover compositional range in narrow-band transition-metal oxides.<sup>4,5</sup> In mixed-valent cuprates and nickelates with the  $\text{K}_2\text{NiF}_4$  structure, phase segregation at lower temperatures is self-organized into hole-rich and hole-poor stripes; the hole-poor stripes retain localized-electron spins whereas the hole-rich stripes contain either itinerant electrons or hole-rich molecular-orbital clusters.<sup>6,7</sup> Although both static and dynamic stripes have been detected by several experimental techniques in some cuprate superconductors,<sup>8,9</sup> the relationship between superconductivity and static versus mobile stripes remains a topic of lively debate.

Thermal conductivity is a sensitive probe of heat transfer by phonons, and bond-length fluctuations associated with mobile stripes or other forms of dynamic phase segregation suppress heat transfer by phonons. As far as we know, all cuprate superconductors probed to date by thermal-conductivity measurements have exhibited a strong suppression of the phonon contribution<sup>10</sup>. This observation is clearly consistent with the conclusion from other techniques that the high-temperature superconductors are characterized by locally cooperative bond-length fluctuations characteristic of an inhomogeneous electronic character and/or distribution.

The low-temperature orthorhombic/high-temperature tetragonal (LTO/HTT) phase of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  ( $0 \leq x \leq 0.3$ ) has served as a prototype for the high- $T_c$  copper-oxide superconductors because of its relatively simple structure and the ability to access the entire range of superconductive compositions. However, substitution of Sr by Ba or of La by a smaller rare earth such as Nd or Eu can stabilize a low-temperature-tetragonal (LTT) phase that is distinguishable

from the HTT phase; cooperative rotations of the  $\text{CuO}_{6/1.5}$  octahedra are about  $[100]$  and  $[010]$  axes in alternate  $(001)$  planes in the LTT phase, but no long-range cooperative rotations are found in the HTT phase. The rotations in the LTT phase tend to stabilize itinerant hole-rich stripes in every fourth Cu-O-Cu row of a  $\text{CuO}_2$  sheet. At  $x=1/8$ , static stripes in the LTT phase suppress the superconductive critical temperature  $T_c$  to near zero.<sup>9,11</sup> The thermal conductivity  $\kappa(T)$  has been measured<sup>12</sup> on ceramic samples of the insulating LTT phase with  $x=1/8$  doping; it showed a typical phonon contribution, i.e., a low-temperature peak in  $\kappa(T)$  followed by a nearly  $1/T$  law with increasing temperature. Disappearance of the phonon peak in superconductive ceramic samples with the LTT structure and  $x \neq 1/8$  reinforced the argument that suppression of the phonon contribution to  $\kappa(T)$  is a characteristic feature of the high- $T_c$  copper-oxide superconductors. We report here  $\kappa(T)$  measurements on a series of superb-quality single crystals of  $\text{La}_{1.6-x}\text{Nd}_x\text{SrCuO}_4$  that transform to the LTT phase at lower temperatures;  $x$  was varied from the nonsuperconductive compound  $x=0.12$  to the slightly overdoped composition  $x=0.20$ . A well-defined phonon thermal conductivity was found in the superconductive compositions. This striking observation would seem to suggest that bond-length fluctuations have little to do with the superconductivity occurring in the LTT phase.

## EXPERIMENTS

All single crystals were grown by the traveling-solvent floating zone (TSFZ) technique. The feed and seed rods for the crystal growth of  $\text{La}_{1.60-x}\text{Nd}_{0.40}\text{Sr}_x\text{CuO}_4$  (Nd-LSCO) and  $\text{La}_2\text{CuO}_4$  (LCO) were prepared by solid-state reaction.  $\text{La}_2\text{O}_3$ ,  $\text{Nd}_2\text{O}_3$ ,  $\text{SrCO}_3$ , and  $\text{CuO}$  all having 99.99% purity were used to synthesize the feed rods, seed rods, and solvent.  $\text{La}_2\text{O}_3$  and  $\text{Nd}_2\text{O}_3$  were baked at  $1000^\circ\text{C}$  for 20 h before weighing. The starting materials were mixed in the mole ratio; an extra 2%  $\text{CuO}$  over the mole ratio was added to the starting mixture for the feed rod in order to enhance the rod density and to compensate the Cu evaporation during crystal

growth. The mixed starting materials were pre-fired at 900 °C for 12 h followed by sintering at 950 °C for 60 h with four intermediate grindings. The powder was pressed to a 6-mm-diameter $\times$ 100 mm rod under about 4000 atm hydrostatic pressure. The rods were finally sintered at 1200 °C for 20 h in oxygen. This procedure has been proven to be critical to obtain rods that have sufficiently high density. High-density rods are required in order to prevent solvent penetration near the melting zone. Because of incongruent melting, a solvent is needed to grow the crystals of  $\text{La}_{1.60-x}\text{Nd}_{0.40}\text{Sr}_x\text{CuO}_4$  and  $\text{La}_2\text{CuO}_4$ . The solvent was made from starting materials of  $\text{La}_2\text{O}_3$ ,  $\text{SrCO}_3$ , and  $\text{CuO}$  in the mole ratio  $(\text{La}_{2-x}\text{Sr}_x):\text{Cu}=22:78$  that were mixed and pre-fired at 900 °C for 12 h. The product was ground; the ground powder was pressed hydrostatically into a rod and sintered at 920 °C for 15 h. A disk with mass 80–100 mg cut from the rod was melted at the end of the feed rod. It has been found that the crystal quality is influenced by the mass of solvent used.

The crystal growth was carried out in an IR heating image furnace (NEC) with two halogen lamps and double ellipsoidal mirrors. The feed and the seed rotate at a speed of 30 rpm in opposite directions to ensure homogeneity of the melt. A ceramic rod, normally a short piece of the feed rod, was used to serve as the seed rod. However, the melting zone can be stabilized in a relatively short time if the seed rod contains a bit of melt from previous growths. High-quality crystals can be obtained with a growth rate as low as 0.50 mm/h. The growth of LSCO was performed in flowing air. The first 10 mm above the necking in the ingot always had some enclosed  $\text{La}_2\text{O}_3$ . This part decomposes into powder after exposing to air for 1–2 days. Local cavities or inhomogeneity in the feed rod alters the melting point of the solvent during crystal growth, which forces adjustment of the heating power in order to keep constant the volume of the melting zone. The ingot contains many domains of low quality crystal if the heating power was adjusted during growth. All crystals used for the measurements in this work were grown with constant heating power.

As-grown crystals were annealed in air at 900 °C for 20 h in order to release thermal stresses formed during crystal growth and to make uniform the oxygen stoichiometry. X-ray Laue diffraction was used to orient the crystals with an accuracy of 1°. A typical size of the bar used for thermal conductivity measurements was  $0.5\times 0.5\times 2.5\text{ mm}^3$  with the longest dimension parallel to either the  $a$ - $b$  plane or the  $c$  axis. The rectangular bars were carefully annealed in pure Ar, Ar+5%  $\text{H}_2$ , air, or oxygen to make the crystals oxygen stoichiometric. Thermoelectric power measurements were used to confirm the mobile charge density. The dc magnetization was obtained with a SQUID magnetometer (Quantum Design). The thermal conductivity  $\kappa(T)$  was measured in the temperature range 4–300 K with a steady-state heat-flow technique having a systematic error less than 20%.

## RESULTS

### Thermal conductivity of the parent compounds $\text{La}_2\text{CuO}_4$ and $\text{La}_2\text{NiO}_4$

More consistent measurements of the thermal conductivity  $\kappa(T)$  of the parent compound  $\text{La}_2\text{CuO}_4$  have been re-

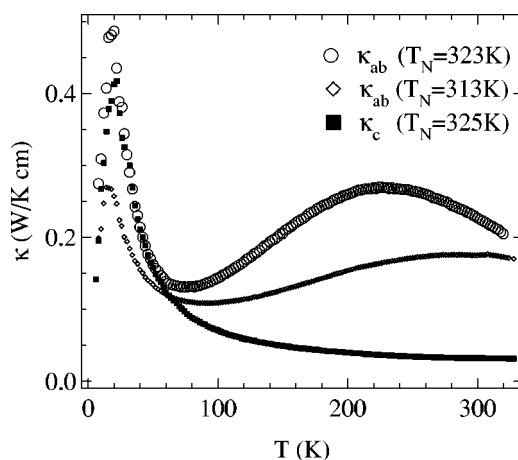


FIG. 1. Temperature dependence of the thermal conductivity in  $\text{La}_2\text{CuO}_{4+\delta}$  single crystals along the  $ab$  plane and the  $c$  axis.

ported in recent years due to improved crystal quality.<sup>13,14</sup> The in-plane  $\kappa_{ab}$  shows a low-temperature peak and a hump near 250 K whereas the  $c$ -axis conductivity  $\kappa_c$  shows only the low-temperature peak typical of the phonon component. Moreover, as shown in Fig. 1,  $\kappa_{ab}$  is highly sensitive to the oxygen stoichiometry of the crystal. The Néel temperature  $T_N$ , which is defined at the peak in  $\chi(T)$ , has also been found to be sensitive to the oxygen stoichiometry.<sup>15</sup> We have monitored the oxygen stoichiometry by a precise measurement of  $T_N$  in these crystals. The crystal with  $T_N=323$ –325 K shows a height of the low-temperature peak that is more than double that of the high-quality crystals previously reported. Both the low-temperature peak and the hump in  $\kappa_{ab}$  decrease in magnitude and the hump moves to a higher temperature in the crystal having  $T_N=313$  K, which corresponds roughly to  $\delta\approx 0.002$ .<sup>15</sup> We have observed a rapid decay of both  $\kappa$  and  $T_N$  under ambient conditions within 30 min after the Ar annealing in some crystals of

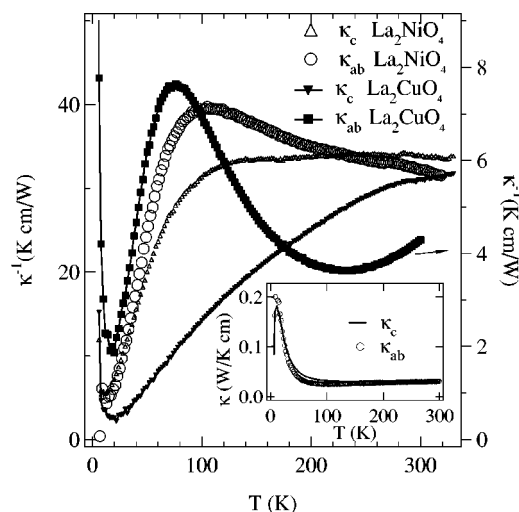


FIG. 2. Temperature dependence of  $1/\kappa$  in  $\text{La}_2\text{NiO}_4$  and  $\text{La}_2\text{CuO}_4$  single crystals along the  $ab$  plane and the  $c$  axis. Inset: temperature dependence of thermal conductivity of  $\text{La}_2\text{NiO}_4$  along the  $ab$  plane and the  $c$  axis.

$\text{La}_2\text{CuO}_4$ . It is clear that inconsistency of the  $\kappa(T)$  reported for  $\text{La}_2\text{CuO}_4$  is caused by the variation of oxygen content as well as different crystal quality.

For the sake of comparison, the  $\kappa(T)$  of an isostructural  $\text{La}_2\text{NiO}_4$  crystal was also measured; the result is shown Fig. 2. This crystal was annealed at 400 °C for 20 h in flowing Ar mixed with 5%  $\text{H}_2$  in order to remove the extra oxygen. A TGA measurement showed the oxygen content was 3.97(2) per formula unit. The dc magnetization is featureless below room temperature, which is below  $T_N$ . The  $\kappa_{ab}$  and  $\kappa_c$  in Fig. 2 show the pronounced low-temperature peak, but the peak height is lower than that of  $\text{La}_2\text{CuO}_4$ . No higher-temperature hump has been found in  $\kappa_{ab}(T)$  below room temperature. However, comparison of the  $\kappa_{ab}^{-1}$  versus  $T$  curves for  $\text{La}_2\text{CuO}_4$  and  $\text{La}_2\text{NiO}_4$ , both plotted in Fig. 2, suggests that a hump of  $\kappa_{ab}$  may occur above room temperature in  $\text{La}_2\text{NiO}_4$ , which is consistent with its higher  $T_N$ .

### Thermal conductivity of the stripe-ordered cuprates and nickelates

The low-temperature peak in  $\kappa_{ab}$  and  $\kappa_c$  and the hump in  $\kappa_{ab}$  give way to a glassy behavior of heat transport in the hole-doped LTO/HTT phase of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (LSCO). In contrast to LSCO, hole-doped crystals of  $\text{La}_{1.60-x}\text{Nd}_{0.40}\text{Sr}_x\text{CuO}_4$  undergo a LTT/LTO transition at  $T_t = 70$  to 80 K for  $0.12 \leq x \leq 0.20$ . Ordering of stripes in every fourth Cu-O-Cu row has been observed in all these compositions in the LTT phase.<sup>16</sup> The stripe ordering in the LTT phase revives the low-temperature peak from the phonon contribution. Moreover the jump at  $T_t$  in both  $\kappa_{ab}$  and  $\kappa_c$ , shown in Fig. 3, indicates a higher thermal conductivity in the LTT phase than that in the LTO phase. For the crystal  $x=0.12$ , the  $\kappa_{\text{LTO}}$  and the jump  $\Delta\kappa$  at  $T_t$  are precisely the same as that previously reported.<sup>14</sup> A much higher height of the low-temperature peak means our crystal had a better quality. The upper bound of the thermal conductivity from the electronic contribution  $\kappa_e(T)$  was calculated with the Wiedemann-Franz law from the temperature dependence of resistivity, see the lower part of Fig. 3. An enhanced electric conductivity, and therefore  $\kappa_e$ , with increasing  $x$  apparently accounts for a monotonic increase with  $x$  of  $\kappa_{ab}$  in the LTO phase and the LTT phase near  $T_t$  for  $x=0.12, 0.15$ , and  $0.20$ . The electronic contribution becomes negligible in  $\kappa_c$  because of quasi-2D conduction in these crystals. It is important to note that following the low-temperature peak,  $\kappa_c$  decays continuously as temperature rises up to  $T_t$  whereas  $\kappa_{ab}$  shows a minimum at  $T \approx 70$  K. This minimum in  $\kappa_{ab}(T)$  is not easily visible for the  $x=0.12$  crystal because the transition at  $T_t = 70$  K intercepts this evolution with temperature. It becomes more clear as  $T_t$  moves to higher temperature in the crystals  $x=0.15, 0.20$  and the crystal  $\text{La}_{1.48}\text{Nd}_{0.4}\text{Sr}_{0.12}\text{CuO}_4$  that has been reported to have a  $T_t = 125$  K.<sup>17</sup> The data provide clear evidence of an extra contribution to  $\kappa(T)$  at higher temperatures in the LTT phase as would occur for a “hump” in  $\kappa_{ab}(T)$  if the LTT phase extended to higher temperatures.

In  $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$ , a semiconductor-semiconductor transition takes place at  $T_{\text{CO}}$  where the charge orders into stripes.<sup>18</sup> For the crystal  $x=0.33$  in which a maximum  $T_{\text{CO}}$  was re-

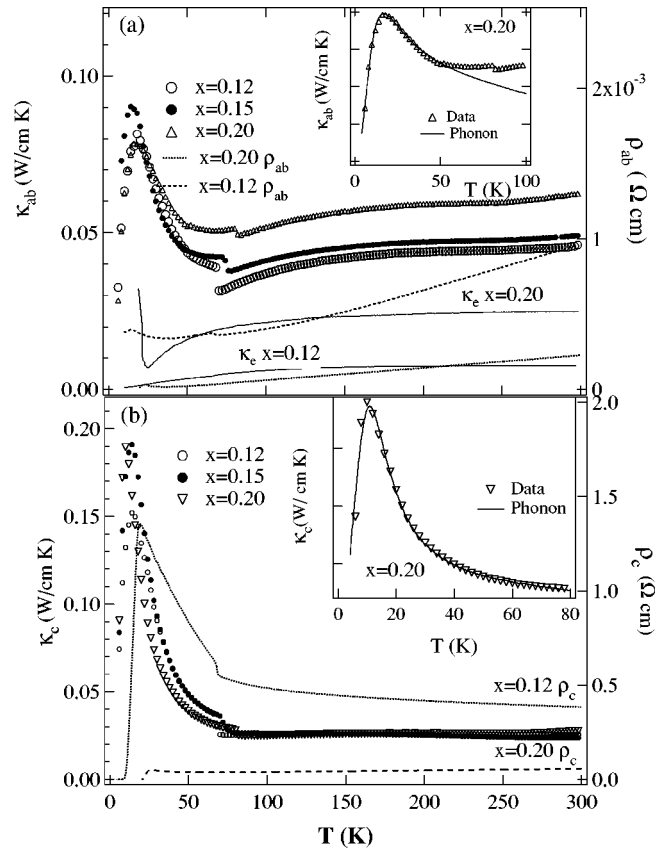


FIG. 3. Temperature dependence of the thermal conductivity in single crystal  $\text{La}_{1.60-x}\text{Nd}_{0.40}\text{Sr}_x\text{CuO}_4$  with  $x=0.12, 0.15$ , and  $0.20$  (a) in the  $a$ - $b$  plane and (b) along the  $c$  axis. The lower part of (a) shows the electronic contribution  $\kappa_e$  in the  $ab$  plane from our electrical resistivity data via the Wiedemann-Franz law. Inset: detail of the low-temperature peak in  $\kappa(T)$  for  $x=0.20$ ; the solid line is a fit to the phononic  $\kappa(T)$  of Debye formula.

ported in  $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$ ,  $T_{\text{CO}}$  also marks a transition from a glassy  $\kappa(T)$ , which is nearly temperature independent, to a phononlike  $\kappa(T)$ , see Fig. 4, though it is not as abrupt as the  $\Delta\kappa$  at  $T_t$  in  $\text{La}_{1.48}\text{Nd}_{0.4}\text{Sr}_{0.12}\text{CuO}_4$ . The  $\kappa(T)$  for the  $x=0.5$  crystal features the phonon component of the conductivity without an anomaly in  $\kappa(T)$  below room temperature.

### DISCUSSION

The highly anisotropic  $\kappa_c(T)$  versus  $\kappa_{ab}(T)$  at temperatures above 50 K in  $\text{La}_2\text{CuO}_4$  in Fig. 1 is in sharp contrast with the nearly isotropic phonon thermal conductivity  $\kappa_{\text{ph}}(T)$  shown as a low-temperature peak in  $\kappa(T)$  at 20 K. Given the isotropic  $\kappa_{\text{ph}}(T)$ , the extra contribution at 250 K in  $\kappa_{ab}$  is about 20 W/K m, which is much higher than  $\kappa \leq 5$  W/K m in most oxides having a perovskite or perovskite-related structure at this temperature. Therefore, heat-conducting channels other than phonons are needed to account for the extra thermal conductivity in the  $a$ - $b$  plane. In their report of  $\kappa(T)$  on single-crystal  $\text{La}_2\text{CuO}_4$ , Nakamura *et al.*<sup>13</sup> have attributed the hump centered in  $\kappa_{ab}$  at  $T \approx 250$  K to spin fluctuations below  $T_N$ . Allen *et al.*<sup>19</sup> have considered two ways magnetism can contribute to  $\kappa(T)$ : (1) spins serve as an additional

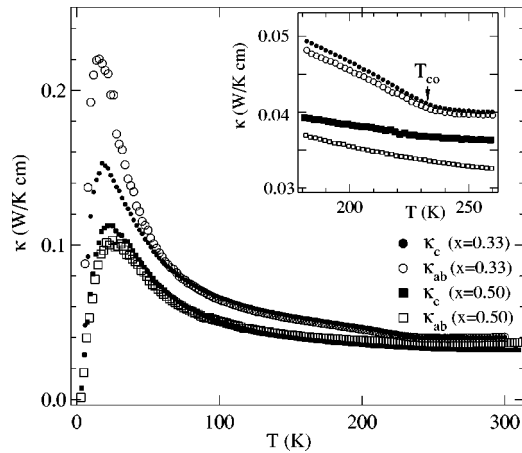


FIG. 4. Temperature dependence of the thermal conductivity in  $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$  ( $x=0.33$  and  $0.50$ ) in the  $a$ - $b$  plane and along the  $c$  axis. Inset: Enlarged representation of the data in the vicinity of the charge-ordering temperature  $T_{\text{CO}}$ .

carrier of heat and (2) spin disorder provides an extra scattering mechanism for the other heat carriers. These contributions have been shown to be either quite small compared to the phonon  $\kappa(T)$  or to make a minimum in  $\kappa(T)$  near  $T_N$  as a result of so-called critical scattering.<sup>20</sup> Low-dimensional, strongly coupled magnetic systems in which the quantum spin or spin-liquid state is formed were not considered in the early days. An additional contribution to  $\kappa(T)$  due to spin excitations have recently been demonstrated in many low-dimensional systems such as  $\text{SrCuO}_2$ ,  $\text{Sr}_2\text{CuO}_3$ ,  $(\text{Sr}, \text{Ca})_{14}\text{Cu}_{24}\text{O}_{41}$ , and  $\text{CuGeO}_3$ .<sup>21–23</sup> It is important to note that all crystals that exhibit a highly anisotropic  $\kappa(T)$  at higher temperatures due to low-dimensional spin excitations also show a well-defined, nearly isotropic low-temperature peak from phonons.

The interstitial oxygen in  $\text{La}_2\text{CuO}_{4+\delta}$  introduces not only a structural distortion, but also holes in the  $\text{CuO}_2$  sheets that perturb the magnetic coupling and lower  $T_N$ . As a result, both the low-temperature peak and the hump at 250 K in  $\kappa_{ab}(T)$  are reduced. Sun *et al.*<sup>17</sup> have tested the relation between the structural distortion and the hump in  $\kappa_{ab}(T)$  due to spin excitations by partially substituting Eu for La in the parent compound  $\text{La}_2\text{CuO}_4$ . Eu substitution essentially suppresses the low-temperature peak whereas the hump due to spin excitations is reduced only a little. The strong spin excitations, which are evidenced by a  $\kappa_{ab}(T) \gg \kappa_c(T)$  below room temperature, seem to have little to do with the phonons in their crystal.

The fluctuating stripe phase confirmed by XAS (Ref. 8) in underdoped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  is consistent with the coexistence of hole-rich and hole-poor regimes that are associated with local structural distortions. Neutron diffraction has shown that the stripes become static in the LTT phase of  $\text{La}_{1.60-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  over the entire range  $0 < x < 0.25$ , which covers the superconductive compositions and the insulating phase near  $x = 1/8$ .<sup>16</sup> The minimum hole doping  $x_{\text{min}}$  to induce superconductivity in the LTT phase is determined by the average size of the rare-earth ion Ln and the  $y$  in the double-rare-earth system  $\text{La}_{2-y-x}\text{Ln}_y\text{Sr}_x\text{CuO}_4$ . An  $x_{\text{min}}$

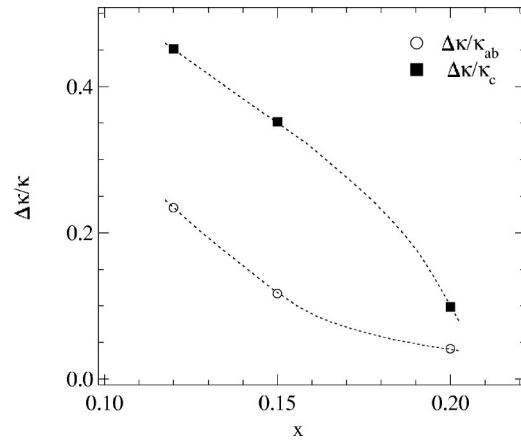


FIG. 5. Doping dependence of the thermal conductivity jump  $\Delta\kappa/\kappa$  at  $T_t$  in single crystals of  $\text{La}_{1.60-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$ . The dashed lines are guides to the eye.

$=0.17$  has been reported in the ceramic samples  $\text{La}_{1.84}\text{Eu}_{0.15}\text{Gd}_{0.01}\text{Sr}_x\text{CuO}_4$  (Ref. 12) whereas it is as low as 0.14 in  $\text{La}_{2-y-x}\text{Nd}_y\text{Sr}_x\text{CuO}_4$ .<sup>11</sup> The buckled  $\text{CuO}_2$  sheets and the size variation of the A site reduce  $T_c$  and narrow down the doping range of the superconductive compositions.<sup>24</sup> On the other hand, reduction of the average A-cation size stabilizes the LTT phase;  $T_t$  rises with increasing  $y$ . The mismatch between the mean A—O and B—O equilibrium bond lengths is relieved with increasing hole doping  $x$ . The LTT/LTO phase boundary located near  $x = 0.2$  varies little on changing  $y$ .

Structural distortions and ion-size variations also suppress  $\kappa(T)$ . A significant reduction in both  $\kappa_{ab}(T)$  and  $\kappa_c(T)$  relative to that of  $\text{La}_2\text{CuO}_4$  has been reported in  $\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4$ ,<sup>17</sup> which undergoes a transition on cooling through  $T_t$  from the LTO phase to an LTO2 phase that evolves with hole doping to the LTT phase. However, a step in  $\kappa(T)$  at  $T = T_t$  indicates that the LTO2 is more thermally conductive than the LTO phase in Eu-substituted  $\text{La}_2\text{CuO}_4$ . A jump  $\Delta\kappa$  of  $\kappa(T)$  at  $T_t$  found in our crystals of  $\text{La}_{1.60-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  ( $x=0.12, 0.15$ , and  $0.20$ ), see Fig. 5, can be used to signal stabilization of a single LTT phase below  $T_t$ . Comparison of  $\kappa(T)$  in  $\text{La}_2\text{CuO}_4$  and  $\text{La}_{1.48}\text{Nd}_{0.4}\text{Sr}_{0.12}\text{CuO}_4$  indicates that the insulator phase with static stripes in the LTT phase has a weaker thermal conductivity. A large and complicated unit cell or a superlattice structure normally results in a small and glassy  $\kappa(T)$ , a phenomenon that has been used to develop thermoelectric materials in which a low  $\kappa(T)$  is desired.<sup>25</sup>

The Meissner effect, Fig. 6, demonstrates the existence of bulk superconductivity in the crystals of  $\text{La}_{1.60-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$ ,  $x=0.15$  and  $0.20$ . Nevertheless, a low-temperature peak of  $\kappa(T)$  in the crystals manifests a phonon component. Moreover, the profile of the low-temperature peak can be fit to the Debye formula of thermal conductivity. The fitting parameters are in line with those of the parent crystal and the  $x=0.125$  insulator. From the documented data available to us, these are the first  $p$ -type cuprate superconductors to show a strong phonon contribution dominating  $\kappa(T)$  in the normal state. The phononic  $\kappa(T)$  found in



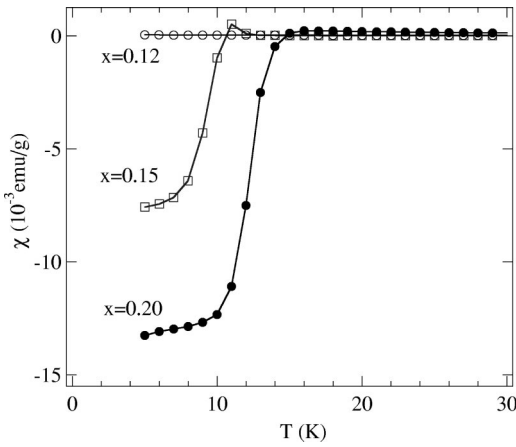


FIG. 6. Zero field cooling magnetization of  $\text{La}_{1.60-x}\text{Nd}_{0.40}\text{Sr}_x\text{CuO}_4$  single crystals with  $x = 0.12, 0.15$ , and  $0.20$  measured in a 35-Oe field applied along the  $c$  axis.

the superconductive crystals is surprising; it indicates that bond-length fluctuations, which severely suppress phonons, may not be involved in superconductive pairing in the LTT phase. Although it is not always required to have a low-temperature peak in  $\kappa(T)$  in order for conventional electron-phonon interactions to mediate formation of superconductive Cooper pairs,<sup>19</sup> the observation of a phonon component in  $\kappa(T)$  might revive the BCS pairing mechanism in this particular case. Recent results from angle-resolved photoemission have demonstrated unambiguously the existence of strong electron-phonon interactions in the normal state of the cuprates.<sup>26</sup> Moreover, an electron-phonon interaction appears to be responsible for the superconductivity found in  $\text{MgB}_2$ . A  $T_c$  as high as 39 K in this compound hints that a mechanism based on conventional electron-phonon interactions might be able to account for an even higher  $T_c$ .

An additional feature of our  $\kappa(T)$  results is the evidence for a contribution to  $\kappa(T)$  at higher temperatures from low-dimensional spin excitations in the LTT phase. This observation suggests that the hole-poor stripes in the  $\text{CuO}_2$  sheets of the LTT phase contain few holes; the hump centered at  $T \approx 250$  K in  $\text{La}_2\text{CuO}_4$  is suppressed by the introduction of holes in  $\text{La}_2\text{CuO}_{4+\delta}$ . We therefore conclude that for all  $x > 1/8$  in the LTT phase, the holes are located in the itinerant-electron stripes that order into every fourth Cu-O-Cu row. An  $x_{\min} > 1/8$  would then mean that the static metallic stripes carry the Cooper pairs in the LTT phase, which would require a coupling between metallic stripes across localized-electron stripes three Cu-O-Cu rows thick. It is too early to tell whether spin excitations in the localized-electron stripes support or compete against formation of Cooper pairs. However, we note that the maximum  $T_c$  in the LTT phase of  $\text{La}_{1.60-x}\text{Nd}_{0.40}\text{Sr}_x\text{CuO}_4$  is only half as high as the maximum  $T_c$  of the LTO phase in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ , which places it in the range of conventional BCS superconductors.

Baberski *et al.* have reached an almost opposite conclusion from their thermal conductivity measurements on polycrystalline samples  $\text{La}_{1.84-x}\text{Eu}_{0.15}\text{Gd}_{0.01}\text{Sr}_x\text{CuO}_4$ .<sup>12</sup> The  $\kappa_c(T)$  reported recently<sup>27</sup> from single crystals of  $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$  also seems to be consistent with their

argument. The experimental results from Baberski *et al.* could be summarized as follow: (1) The step  $\Delta\kappa$  at  $T_i$  reduces with increasing hole doping  $x$ ; the  $\Delta\kappa$  becomes invisible in the superconducting compositions  $x \geq 0.17$ . (2) The low-temperature peak, which is well recognized for the compositions near  $x = 1/8$  in the LTT phase, disappears in the compositions  $x \geq 0.17$ . These findings led them to conclude that high- $T_c$  superconductivity occurs in the LTT phase if the stripes become dynamic. Although the argument is very intriguing, it holds mostly for the measurements on Eu-substituted  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ . A large A-site cation-size variation may suppress the low-temperature peak. The disappearance of a  $\Delta\kappa$  at  $T_i$  in the superconductive compositions  $x \geq x_{\min}$  in this system means that the bulk LTT phase is not stabilized as a single phase at low temperatures. Moreover, there is no proof from neutron diffraction to show that the LTT phase exists in these compositions.

The concept of a quantum critical point has been put forward in recent years to explain the complicated phase diagram of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ .<sup>28</sup> The phase transition takes place at  $T = 0$  K due to a change of quantum-mechanical parameters. As the temperature rises, the critical point develops into a regime where there are phase fluctuations. Since high- $T_c$  superconductivity occurs at the crossover from localized to itinerant electronic behavior, phase fluctuations are inevitable in the normal state. The crossover has also been found in mixed-valent  $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$  and single valent  $\text{RNiO}_3$  ( $R = \text{La-Lu}$ ). The common feature in the  $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ ,  $\text{RNiO}_3$ , and the LTO/HTT phase of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  is a collapse of  $\kappa(T)$  in a narrow range of doping or of the overlap integral in the phase.<sup>13,29,30</sup> Ordered stripe phases appear to be an alternative solution to phase fluctuations at the crossover. Hall-effect measurements have confirmed the 1D conduction in the stripe ordered phase.<sup>31</sup> Phonons are restored in the ordered stripe phase, which makes possible conventional BCS pairing. On the other hand, the physical properties in the superconductive state, such as the isotope effect,<sup>32</sup> seem not to distinguish the superconductive mechanism in the LTT phase from that in other high- $T_c$  cuprate superconductors.

The nickelates  $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$  have been well-known for charge ordering and stripes. For  $x = 1/3$ , charge ordering on cooling through  $T_{\text{CO}}$  marks a transition from polaronic conduction at high temperature to an insulator.<sup>18</sup> Hess *et al.*<sup>33</sup> have reported a transition from a glassy  $\kappa(T)$  to a phononic  $\kappa(T)$  at  $T_{\text{CO}}$  in the  $x = 0.33$  crystal. An almost identical  $\kappa_c(T)$  has been found in our crystal of  $x = 0.33$ ; however, the  $\kappa_{ab}(T)$  is higher than  $\kappa_c(T)$  at low temperatures. Both  $\kappa_c(T)$  and  $\kappa_{ab}(T)$  are peaked at 20 K in our crystals whereas the peaks are 25 K in the report by Hess *et al.* The  $x = 0.5$  crystal undergoes a charge ordering at 340 K that is different from that found in the  $x = 0.33$  crystal.<sup>18</sup> The resistivity changes smoothly on crossing  $T_{\text{CO}} = 340$  K.<sup>18</sup> Even though our  $\kappa(T)$  measurement on this crystal could not be carried out above room temperature, the phononic  $\kappa(T)$  supports charge ordering in this composition.

In conclusion, measurements of thermal conductivity on crystals of the cuprates and nickelates reveal that spin excitations are responsible for an extra contribution to  $\kappa_{ab}(T)$  in

isostructural  $\text{La}_2\text{CuO}_4$  and  $\text{La}_2\text{NiO}_4$ . How well the spin excitations carry heat depends on the crystal quality. The low-temperature peak and the spin-excitation contribution have been found to be sensitive to extra oxygen in the structure. Second, a step of  $\Delta\kappa$  at  $T_i$  ensures the presence of the LTT phase, and the phononic  $\kappa(T)$  at  $T < T_i$  is consistent with the observation that the stripes are static in the LTT phase of  $\text{La}_{1.60-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  ( $x=0.15$  and  $0.20$ ). Third, the anisotropic  $\kappa(T)$  of  $\text{La}_{1.60-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$ , within the temperature

range  $30\text{ K} < T < T_i$ , which resembles that in the parent compound, suggests the existence of spin excitations in the LTT phase.

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