Stripe Correlations of Spins and Holes and Phonon Heat Transport in Doped La$_2$CuO$_4$

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Abstract

We present experimental evidence for a dramatic suppression of the phononic thermal conductivity of rare earth and Sr doped La$_2$CuO$_4$. Remarkably, this suppression correlates with the occurrence of superconductivity. Conventional models for the phonon heat transport fail to explain these results. In contrast, a straightforward explanation is possible in terms of static and dynamic stripe correlations of holes and spins.

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The structural phase transition from the orthorhombic (LTO) to the low temperature tetragonal (LTT) phase observed in rare earth (RE) doped La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) has a pronounced influence on the electronic properties of these materials. In particular, in a certain composition range the LTT phase is not superconducting, but antiferromagnetic order occurs at low temperature and at finite Sr, i.e. charge carrier concentration [1]-[7]. Various mechanisms have been suggested to explain the strong sensitivity of the electronic structure to the subtle structural changes associated with the phase transition as e.g. band structure effects [8], hole concentration dependent commensurability effects and charge density wave–like instabilities [9], as well as a novel coupling of the charge carrier motion to the tilt displacements of the CuO$_6$ octahedra via spin–orbit scattering [10]. More recently Tranquada et al. [7] have presented evidence for an important role of stripe correlations of spins and holes, i.e. an ordering in the CuO$_2$ planes into antiphase antiferromagnetic stripe domains separated by domain walls to which the holes segregate.

We show in this letter that in addition to its well established influence on the electronic properties the structural transition has also dramatic consequences for the lattice dynamics. Our main result is that the phononic contribution $\kappa_{ph}$ to the thermal conductivity increases strongly below the structural transition, but only if the LTT phase is not superconducting [11]. Remarkably, the behavior of the thermal conductivity of Sr doped La$_2$CuO$_4$ without additional RE doping suggests the even more general conclusion that $\kappa_{ph}$ is strongly suppressed for all superconducting compositions of RE and Sr doped La$_2$CuO$_4$. Our results have strong implications for the interpretation of heat transport in doped La$_2$CuO$_4$. In particular, the conventional models for phononic thermal conductivity based on enhanced phonon–defect scattering on alloying or conventional phonon–electron scattering fail to account for these observations. In contrast, a straightforward interpretation is possible in terms of static and dynamic stripe correlations of spins and holes. If this interpretation is correct our data in turn imply that stripe correlations are dynamic for superconducting compositions, whereas they are either static or absent for non–superconducting compositions [11].

The samples used in this study have been prepared by the usual solid state reaction [12]. They are well characterized with various physical properties as described in ref. [2,3,5,12]. The thermal conductivity $\kappa$ was measured by a standard method for compounds with a wide range of Sr und RE concentrations, $x$ and $y$, respectively. We note that the absolute values of the thermal conductivity in sintered materials differ from those in single crystals due to scattering from grain boundaries. In contrast, the temperature and doping dependencies of $\kappa$ found in single crystals and polycrystals of LSCO agree well with each other [13,14]. Since we shall analyse only the temperature and doping dependence of $\kappa$ we show normalized data throughout this paper. We mention that the absolute values of $\kappa$ at high temperatures do not change significantly as a function of RE doping within experimental errors.

As an example for the typical behavior of the thermal conductivity of RE doped LSCO we show in fig. 1 our results for a Pr and a Nd doped sample both with a Sr content of $x = 0.12$. The Pr doped sample does not show a low temperature structural transition [13]. Its thermal conductivity decreases monotonously with decreasing temperature, similar to what is found in LSCO at a comparable Sr content but without additional RE doping [13,14] (inset fig. 1). In the Nd doped sample the structural transition to the LTT phase at $T_{LT} \approx 80\text{K}$ has a dramatic influence on the thermal conductivity: $\kappa$ increases strongly below $T_{LT}$ reaching a
maximum around 25K. We note that this temperature dependence of $\kappa$ in the LTT phase is similar to that found for the purely phononic thermal conductivity of undoped insulating La$_2$CuO$_4$ (inset fig. 1).

In the high $T_c$ superconductors the thermal conductivity has an electronic and a phononic contribution, $\kappa_{el}$ and $\kappa_{ph}$, respectively \[10\]. Nevertheless, in the present case the increase of $\kappa$ below $T_{LT}$ is clearly due to an increase of $\kappa_{ph}$: Firstly, the electrical conductivity decreases below $T_{LT}$ \[1\]. According to the Wiedemann–Franz law $\kappa_{el}$ should then decrease also and the increase of $\kappa = \kappa_{ph} + \kappa_{el}$ must be due to an increase of $\kappa_{ph}$. Secondly, the pronounced increase of $\kappa$ below $T_{LT}$ occurs also for strongly underdoped samples (see fig. 2), where the charge carrier concentration is so low that $\kappa_{el} \ll \kappa_{ph}$ and any increase of $\kappa$ must be due to an increase of $\kappa_{ph}$.

From the data presented so far one may conclude that it is the structural phase transition which causes the strong increase of $\kappa_{ph}$ below $T_{LT}$. However, this is not true in general: We show in fig. 2 the thermal conductivity for Eu doped samples ($y = 0.15$) with Sr contents between 0.05 and 0.22. X–ray diffraction experiments reveal the occurrence of the low temperature structural transition to the LTT phase at $T_{LT} \approx 120$K in all these samples \[5\]. However, the low temperature increase of $\kappa$ occurs only in the limited Sr concentration range of $x \leq 0.17$. Remarkably, Eu doped LSCO with $y = 0.15$ is not superconducting for $x > 0.17$, whereas for $x > 0.17$ superconductivity occurs \[5\].

We find similar results in the Nd doped samples. As a measure of the increase of $\kappa$ at low temperatures one may use the magnitude $\Delta \kappa$ of the jump of $\kappa$ at $T_{LT}$, since it is apparent from the data of fig. 2 that $\Delta \kappa$ correlates with the size of the low temperature maximum. In fig. 3 we show $\Delta \kappa$ as a function of the Sr concentration for samples with Nd contents of $y = 0.3$ and $y = 0.6$. The low temperature increase of $\kappa$, i.e. $\Delta \kappa > 0$, occur for $x \leq 0.17$ for Nd doping of $y = 0.3$ and for $x \leq 0.23$ for Nd doping of $y = 0.6$. A comparison of these concentration pairs to the low temperature Nd/Sr phase diagram of ref. \[2\] shows that they lie on the boundary separating the superconducting and the non–superconducting region of the LTT phase. Thus, the low temperature maximum of $\kappa_{ph}$ occurs in the LTT phase only for compositions which are not superconducting.

Taking together these results we arrive at a remarkable correlation of the lattice dynamics with the electronic properties of the LTT-phase: The phononic thermal conductivity of RE doped La$_{2-x}$Sr$_x$CuO$_4$ increases strongly below the low temperature structural transition, but only if the LTT phase is not superconducting.

At this point it is instructive to reexamine the thermal conductivity of LSCO without additional RE doping (inset fig. 1) \[13\]. Insulating La$_2$CuO$_4$ has a purely phononic thermal conductivity with a pronounced maximum occurring around 25K. Notably, in strongly overdoped, non–superconducting samples the maximum of $\kappa$ at low temperatures occurs also. We shall come back to this below. For intermediate Sr doping of $x \leq 0.25$, when samples are superconducting, $\kappa$ is strongly suppressed and the low temperature maximum is absent. Obviously this Sr concentration dependence of $\kappa$ in LSCO fits very well into the correlation between the magnitude of $\kappa_{ph}$ and the occurrence of superconductivity suggested by our results on the RE doped samples. We are thus lead to the more general conclusion: Whenever Sr and RE doped La$_2$CuO$_4$ is superconducting, the phononic thermal conductivity is strongly suppressed; in particular, the low temperature maximum of $\kappa_{ph}$ is absent then.

We shall now discuss this result within the conventional models for phonon heat trans-
port in high $T_c$ materials. Since $\kappa_{ph}$ depends strongly on the RE and the Sr concentration, the scattering of the phonons should also depend on the doping. Two possible mechanisms are then apparent: 1. Doping induced phonon–impurity scattering and 2. phonon–electron scattering. We note first that the suppression of $\kappa_{ph}$ with increasing Sr doping in LSCO without additional RE doping (inset fig.1) is usually attributed to phonon–defect/disorder scattering which increases upon alloying [13,14,16]. However, our data immediately rule out such an explanation due to the reappearance of the maximum of $\kappa_{ph}$ at large Sr and RE doping in the non–superconducting LTT phase (fig.1). Regarding conventional phonon–electron scattering we note that, firstly, the suppression of $\kappa_{ph}$ does occur for Sr concentrations of $x \approx 0.05$. For such low charge carrier concentrations phonon–electron scattering is unimportant. Secondly, if phonon–electron scattering suppresses $\kappa_{ph}$ in the superconducting samples one expects that at least part of the phononic thermal conductivity reappears below the superconducting transition (which is in the same temperature range as $T_{LT}$), since the opening of the energy gap would strongly suppress phonon–electron scattering. However, no such enhancement of $\kappa_{ph}$ is found in LSCO or RE doped LSCO below $T_c$.

We should mention here as a further mechanism a possible suppression of $\kappa_{ph}$ due to scattering of phonons on anharmonic soft phonon vibrations associated with the structural transitions in doped $La_2CuO_4$. It is well known that the LTO phase is characterized by substantial anharmonicity of the tilting vibrations of the $CuO_6$-octahedra, which seems to increase with Sr-doping [17]. A possible reason for this anharmonicity is the instability of the LTO-phase towards the LTO $\rightarrow$ LTT phase transition. However, it is well known that this structural instability is mainly determined by sterical lattice properties which depend on the RE content. In particular, the low temperature transition does occur at comparable $T_{LT}$ for both Sr doped compounds and insulators with $x = 0$ [13,18]. It is apparent that the associated weak Sr concentration dependence of the structural instability does not correlate with the pronounced change of $\kappa$ as a function of $x$ in LSCO (inset of fig.4). Moreover, a scenario which explains the pronounced damping of $\kappa_{ph}$ in LSCO due to the structural instability becomes very unlikely when taking into account the findings for insulators with $x = 0$. We find essentially the same $\kappa_{ph}$ in the entire temperature range for $La_2CuO_4$ and for RE doped compounds regardless whether the structure is LTO or LTT. This means that neither the low temperature structure, i.e. the presence of the tilting instability at low temperatures in $La_2CuO_4$ and its absence in the LTT phase of the RE doped compounds, nor the very close proximity to the structural transition, which is apparently present close to $T_{LT}$ in the RE doped compounds, causes significant differences in the phonon heat transport of these antiferromagnetic insulators. Thus we conclude that anharmonicity due to the structural instability can not explain the damping of $\kappa_{ph}$ in LSCO ($x \neq 0$) and its reappearance in the non–superconducting LTT phase. Note that this conclusion is also supported by our findings for compounds with high Sr contents, since the strong low temperature increase of $\kappa$ is not linked to the phase transition alone but to the electronic properties of the LTT-phase (see fig. 2,3). In particular, in the superconducting composition range of the LTT-phase no low temperature enhancement of $\kappa_{ph}$ is observed, although anharmonicity due to the structural instability is absent.

We are thus led to the conclusion that conventional models for the phonon heat transport fail to explain the experimental results for RE and Sr doped $La_2CuO_4$. On the other hand, the correlation of the behavior of $\kappa_{ph}$ at low temperatures with the occurrence of
superconductivity suggests an electronic scattering channel for the phonons. If conventional phonon–electron scattering is inadequate, as shown above, one might imagine scattering of phonons on some ‘collective electronic excitation’. In the following we suggest a possible such mechanism based on the formation of so called stripe correlations of spins and holes and their dynamics.

The neutron scattering experiments of Tranquada et al. \[7\] give evidence for static stripe correlations, i.e. a static spatial modulation of the charge and spin density in the CuO$_2$ planes, in the non–superconducting low temperature tetragonal phase of Nd doped LSCO ($y = 0.4, x = 0.12$). Remarkably, the corresponding magnetic superstructure reflections occur exactly at the wave vector of the well known incommensurate peaks in inelastic neutron scattering on superconducting LSCO \[20\] (without additional RE doping) leading to the suggestion that dynamic stripe correlations are present in superconducting LSCO \[7\]. Within this scenario a rather natural explanation of our results is possible. We first note that via the well known relation between bondlengths and charge density a spatially inhomogeneous charge distribution implies corresponding variations of the lattice constants. Note that just these variations are observed in neutron scattering as superstructure reflections in the case of pinned hole stripes in the non–superconducting LTT phase. If the stripe correlations are dynamic one expects that dynamic lattice modulations are induced provided that the time scale of the dynamic stripe correlations is comparable to that of the lattice vibrations. We note that, being a collective excitation linked to the magnetic correlations, the time scale of the stripe correlations is not given by the Fermi energy but should be much smaller. In fact, the inelastic neutron results on superconducting Sr doped La$_2$CuO$_4$ \[20\] suggest an energy scale for the dynamic stripe correlations around 10meV, comparable with typical phonon energies. Then the dynamics of the stripe correlations will couple strongly to the lattice causing a pronounced damping of the phonons \[19\]. Accordingly, one expects a strong reduction of the lattice heat conductivity. In contrast, static stripe correlations will at most lead to some minor modifications of the phonon dispersion compared to pure La$_2$CuO$_4$, but they will not suppress the phonon heat conductivity significantly.

Note that, if we assume that this scenario is the correct explanation for the behavior of $\kappa_{ph}$, we may in turn conclude from the correlation between the suppression of $\kappa_{ph}$ and the occurrence of superconductivity that the dynamics of the stripe correlations is important for superconductivity.

Up to now we have discussed the behavior of $\kappa_{ph}$ at the structural transition and have attributed the increase of $\kappa_{ph}$ to the pinning of stripe correlations. However, we emphasize that the reappearance of the maximum of $\kappa$ found in strongly overdoped, non–superconducting LSCO without additional RE doping (inset fig. 1) finds a straightforward explanation within this picture also. At very high doping the magnetic correlations in the CuO$_2$ planes are weak or absent. Therefore the tendency to form stripe correlations vanishes or is at least strongly reduced. Accordingly, the strong suppression of phonon heat conductivity discussed above is absent and the phonon maximum should reappear. We should note here that the maximum of $\kappa$ at high Sr-doping is usually attributed to the electronic contribution to $\kappa$. However, at such high Sr-doping disorder scattering should suppress any low temperature maximum of the electronic contribution to the thermal conductivity, as is well known for conventional metals. Therefore, we believe that an electronic origin of the low temperature peak at high doping is possible in principle, but rather unlikely.
We mention at this point that as a further check for our interpretation we have measured the thermal conductivity of La$_{2-x}$Sr$_x$NiO$_4$ with $x = 1/3$. This material is an insulator, i.e. $\kappa = \kappa_{ph}$. Moreover, no structural transition occurs below 300K. On the other hand, the presence of charge and spin ordering below about 240K is well established in this compound from neutron and electron diffraction studies [22]. Remarkably, the behavior of $\kappa$ compares well with our findings for doped La$_2$CuO$_4$ in the non-superconducting LTT-phase [23]. Due to charge ordering $\kappa$ increases, the slope $\partial\kappa/\partial T$ changes sign from negative to positive, and at low temperatures a pronounced maximum of $\kappa$ occurs. These findings clearly confirm the interpretation for the cuprates presented here.

We finally mention that when assuming a relation between the dynamics of stripe correlations and superconductivity the influence of the buckling distortion on the superconducting properties of the LTT phase is qualitatively expected. We recall that it has been shown in ref. [2] that the LTT phase is not superconducting, if the buckling of the CuO$_2$ planes, i.e. the tilting of the CuO$_6$ octahedra, exceeds a critical value [11]. According to the results of Tranquada et al. the pinned stripes in a single CuO$_2$ layer are either parallel to the [100]– or to the [010]–direction (using the notation of an undistorted lattice). From the structure of the LTT phase it is apparent that there are differences between these directions, which increase with increasing buckling distortion. Therefore, assuming that the pinning of the stripe correlations requires a finite ‘pinning potential’ one expects that within the LTT phase stripe correlations are pinned only beyond a certain value of the buckling.

In conclusion, we have shown that in RE and Sr doped La$_2$CuO$_4$ the phononic thermal conductivity is strongly suppressed for all superconducting compositions. The conventional models of phonon heat transport based on phonon–defect scattering or conventional phonon–electron scattering fail to explain these results. In contrast, the recently suggested picture of dynamic and static stripe correlations of spins and holes allows for a straightforward explanation. If this explanation is correct the correlation between suppressed $\kappa_{ph}$ and the occurrence of superconductivity tells that the dynamics of stripe correlations is important for superconductivity in doped LSCO.

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REFERENCES

[1] B. Büchner et al., Physica C 185-189, 903 (1991); M.K. Crawford et al., Phys. Rev. B 44, 7749 (1991)
[2] B. Büchner et al., Phys. Rev. Lett. 73, 1841 (1994)
[3] B. Büchner et al., Europhys. Lett. 21, 953 (1993)
[4] Y. Nakamura and S. Uchida, Phys. Rev. B 46, 5841 (1992)
[5] B. Büchner et al., J. Low Temp. Phys. 95, 285 (1994); B. Büchner et al., Physica C 235-240, 235 (1994)
[6] M. Breuer et al., Z. Phys. B 92, 331 (1993)
[7] J.M. Tranquada et al., Nature 375, 561 (1995); J.M. Tranquada et al., Phys. Rev. B 52, 3581 (1995)
[8] W.E. Pickett et al., Phys. Rev. Lett. 67, 228 (1991)
[9] S. Barisic and J. Zelenko, Sol. State Comm. 74, 367 (1990)
[10] N. Bonesteel et al., Phys. Rev. Lett. 68, 2684 (1992)
[11] We mention that based on zero field cooled susceptibility (shielding) measurements J.M. Tranquada et al. (Phys. Rev. Lett. 73, 338 (1997)) have recently argued that long range antiferromagnetic order and superconductivity coexist in the LTT phase, i.e. they claim that pinning of stripes strongly reduces $T_c$ but does not suppress superconductivity completely. However, since at least for our samples bulk sensitive Meissner and specific heat measurements as well as shielding measurements (on powdered samples) signal a destruction of superconductivity (see the data and their discussion in ref. [2,3,5]) we follow ref. [2] and discriminate between a superconducting and a non-superconducting LTT phase.
[12] M. Breuer et al., Physica C 208, 217 (1993)
[13] Y. Nakamura et al., Physica C 185-189, 1409 (1991)
[14] M. Sera et al., Sol. State Comm. 74, 951 (1990)
[15] W. Schäfer et al., Phys. Rev. B 49, 9248 (1994)
[16] see e.g. C. Uher, p. 159, in Physical Properties of High Temperature Superconductors, Vol. III, D.M. Ginsberg (Ed.) (World Scientific, Singapore, 1992)
[17] M. Braden et al., Physica C 223, 396 (1994); Z. Phys. B 94, 29 (1994)
[18] B. Keimer et al., Z. Phys. B 91, 373 (1993)
[19] We note that a strong damping of phonons in the Bi based materials has been suggested by P.B. Allen et al., Phys. Rev. B 49, 9073 (1994) on the basis of thermal conductivity and neutron scattering results.
[20] T.E. Mason et al., Phys. Rev. Lett. 68, 1414 (1992)
[21] J.L. Cohn et al., Phys. Rev. B 46, 12053 (1992)
[22] see e.g. C.H. Chen et al., Phys. Rev. Lett. 76, 447 (1993); V. Sachan et al., Phys. Rev. B 51, 12742 (1995)
[23] C. Hess, M. Hücker, B. Büchner, and S.W.Cheong, to be published
FIGURES

FIG. 1. Thermal conductivity of Pr ($y = 0.85$) and Nd ($y = 0.6$) doped La$_{1.88-y}$RE$_y$Sr$_{0.12}$CuO$_4$ normalized to $\kappa(150\text{K})$ as a function of temperature (see text). Inset: In–plane thermal conductivity $\kappa_{ab}$ of La$_{2-x}$Sr$_x$CuO$_4$ single crystals as a function of temperature taken from ref. [13]. Samples with $x = 0.10, 0.15, 0.20$ are superconducting, those with $x = 0, 0.30$ are not.

FIG. 2. Thermal conductivity of Eu doped La$_{1.84-x}$Eu$_{0.15}$Gd$_{0.01}$Sr$_x$CuO$_4$ normalized to $\kappa(150\text{K})$ as a function of temperature for various Sr concentrations given in the figure. Curves are shifted for clarity. Only samples with $x > 0.17$ are superconducting. $T_{LT} \approx 120\text{K}$ is indicated by the dashed line. The small amount of Gd in the samples was used as an ESR-probe and is unimportant in the context of this paper.

FIG. 3. Anomaly $\Delta\kappa$ extracted from the jump–like anomalies of $\kappa$ at $T_{LT}$ (see fig.1) normalized to $\kappa(T_{LT})$ as a function of the Sr concentration for Nd doped La$_{2-x}$Sr$_x$CuO$_4$. $\triangle$: Nd content $y = 0.3$; $\bullet$: $y = 0.6$. 
Fig. 3

La$_{2-x-y}$Sr$^x$Nd$^y$CuO$_4$

$\Delta \kappa / \kappa$

Sr content $x$

$y=0.3$

$y=0.6$

$x=0.17$

$x=0.23$
Temperature (K)

La$_{1.84-x}$ Eu$_{0.15}$ Gd$_{0.01}$ Sr$_x$ CuO$_4$

$\kappa/\kappa(150\text{K})$

Fig. 2
La$_{1.88-y}$RE$_y$Sr$_{0.12}$CuO$_4$

Fig. 1