Low-temperature electronic heat transport in La$_{2-x}$Sr$_x$CuO$_4$ single crystals: Unusual low-energy physics in the normal and superconducting states

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The thermal conductivity $\kappa$ is measured in a series of La$_{2-x}$Sr$_x$CuO$_4$ ($0 \leq x \leq 0.22$) single crystals down to 90 mK to elucidate the evolution of the residual electronic thermal conductivity $\kappa_{res}$, which probes the extended quasiparticle states in the $d$-wave gap. We found that $\kappa_{res}/T$ grows smoothly, except for a 1/8 anomaly, above $x \approx 0.05$ and shows no discontinuity at optimum doping, indicating that the behavior of $\kappa_{res}/T$ is not governed by the metal-insulator crossover in the normal state; as a result, $\kappa_{res}/T$ is much larger than what the normal-state resistivity would suggest in the underdoped region, which highlights the peculiarities in the low-energy physics in the cuprates.

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It is now generally believed that the superconducting state of the high-$T_c$ cuprates can be more or less conventionally described by a BCS-like condensate with a $d$-wave symmetry and well-defined Fermi-liquid-like quasi-particle (QP) excitations from it, though the mechanism for the occurrence of the superconductivity is expected to be highly unconventional. Specifically, the $d$-wave phenomenology of the superconducting state predicts such phenomena as the “Volovik effect” and the “universal” heat conduction, both of which have been confirmed by experiments; namely, in optimally-doped YBa$_2$Cu$_3$O$_{7-\delta}$ (Y-123), the electronic specific heat has been shown to increase with the magnetic field $H$ as $\sqrt{H}$ and the electronic thermal conductivity for $T \rightarrow 0$ has been shown to be independent of impurity concentration. These effects are essentially caused by the QPs induced near the nodes of the $d$-wave gap either by vortices or by impurities. In particular, impurities are believed to induce an “impurity band” at the Fermi energy $E_F$ in the $d$-wave gap, and the extended QPs in this band are considered to form an ordinary Fermi liquid; in this sense, one can say that the low-energy physics of the cuprates in the superconducting state is governed by a Fermi liquid, while that in the normal state appears to be governed by a non-Fermi liquid.

Recently, there appeared several studies which suggest that the rather simple picture described above may not be the whole story. It was theoretically argued, for example, that the impurity band may actually develop a gap-like feature at very low energies near $E_F$; also, the quantum interference effect may lead to a localization of the extended QP states. Experimentally, it was recently reported that the QP contribution to the thermal conductivity is absent at low temperatures in an underdoped cuprate YBa$_2$Cu$_3$O$_8$ (Y-124), which indicates that there is no extended QP at low temperatures in this system; this behavior appears to be a result of some kind of localization of the QPs and thus is somewhat reminiscent of the unusual charge localization found in the normal state of the underdoped cuprates under high magnetic fields. If the disappearance of the extended QPs in the superconducting state and the “insulating” normal state are actually related, that might mean that the same mechanism dictates the charge localization in both the normal and the superconducting states. This issue becomes even more intriguing in view of the recent theoretical proposal that there may be a “superconducting thermal insulator” phase inside a superconductor. To address this issue, it is desirable to measure the thermal conductivity at very low temperatures in a series of La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) single crystals (where the behavior of the low-temperature normal state is known) and trace the behavior of the electronic thermal conductivity as $x$ is varied.

In this Letter, we report our measurements of the low-temperature thermal conductivity in a series of LSCO single crystals with $x = 0 - 0.22$; these $x$ values cover the whole range of the parent Mott insulator, the underdoped superconductor, and the overdoped superconductor. We find that the extended QPs appear to exist in the zero-temperature limit throughout the superconducting phase and that the transition to the “thermally insulating” state takes place concomitantly with the superconductor-to-insulator transition at $x = 0.05$. Therefore, the low-energy physics of the QPs in the superconducting state and that of the charge carriers in the normal state under 60 T appear to be fundamentally different in the underdoped region. Moreover, the evolution of the QP heat transport in the superconducting state clearly demonstrates the shortcomings of the simple $d$-wave phenomenology.

The single crystals of LSCO ($x = 0 - 0.22$) are grown by the traveling-solvent floating-zone technique. The underdoped (overdoped) crystals are annealed in reducing (oxygenating) atmosphere to minimize deviation from the stoichiometric oxygen content. The crystals are cut into rectangular platelets with the typical dimensions of $1.5 \times 0.5 \times 0.1$ mm$^3$, where the $c$ axis is perpendicular to
the platelets within an accuracy of 1°. The optimally-doped sample \((x=0.17)\) shows zero resistivity at 40.5 K, and the most underdoped superconducting sample \((x=0.06)\) shows zero resistivity at \(\sim 8\) K. The temperature dependence of the in-plane resistivity \(\rho_{ab}\) for most of the \(x\) values studied here are reported in Ref. [4].

The thermal conductivity \(\kappa\) is measured using the conventional steady-state “one heater, two thermometer” technique. A chip heater and two RuO\(_2\) chip sensors are attached to the sample with gold wires; on the sample, ohmic contacts with the contact resistance of less than 0.5 \(\Omega\) are made with gold contact pads and the gold wires are attached to these pads using silver epoxy. To minimize heat leak, superconducting NbTi wires with 15-\(\mu\)m diameter are used as the leads of the chip sensors. The lowest temperature of our thermal conductivity measurement is typically 90–100 mK, below which it becomes uncertain whether the true electron temperature is measured with our setup assembled on a cold finger of a dilution refrigerator. The temperature difference between the two thermometer contacts, which are separated by \(\sim 1\) mm, is controlled to be typically 3\% of the sample temperature. The error in \(\kappa\) due to geometrical factors is less than 10\%.

Figure 1(a) shows the plots of \(\kappa/T\) vs. \(T^2\) for a series of \(x\) from 0 to 0.22. If the electronic thermal conductivity \(\kappa_{el}\) and the phononic thermal conductivity \(\kappa_{ph}\) behave as \(aT\) and \(bT^3\), respectively, like in ordinary Fermi liquids at low temperatures, we can write \(\kappa/T = a + bT^2\); in this case, the zero-temperature intercept and the slope of a straight line in the \(\kappa/T\) vs. \(T^2\) plot respectively give \(a\) and \(b\). For cuprates, it has been demonstrated that \(\kappa(T)\) obeys the above temperature dependence for \(T^2 < 0.02\) K\(^2\) in typical crystalline samples [8,7], and the source of the \(aT\) term in \(\kappa\) of the superconducting cuprates has been discussed to be the Fermi-liquid-like impurity band created in the \(d\)-wave gap.

In Fig. 1(a), one can easily see the overall trend that \(\kappa/T\) is shifted up with increasing \(x\). A natural extrapolation of the data for \(T \to 0\) appears to give a finite intercept for superconducting samples \((x > 0.05)\), and this intercept grows rather rapidly with \(x\). Therefore, the data in Fig. 1(a) already tell us that there is some finite residual thermal conductivity \(\kappa_{res}\), which is of electronic origin, in the superconducting samples and this \(\kappa_{res}\) tends to grow as \(x\) is increased above 0.05.

To quantify the above observation, we have tried to draw a straight line that best fits the lowest-temperature part of the \(\kappa(T)/T\) data for each \(x\). The results are shown in Figs. 1(b)-1(m) for various \(x\); although for some of the \(x\) values it is not obvious whether the lowest-temperature data can be best described by a straight line \((x = 0.08\) and 0.14, for example), for many of the \(x\) values the data are actually well fitted with a straight line below \(T^2\) of \(\sim 0.02\) K\(^2\), which is in good correspondence with the previous studies of other cuprates [7,17]. The zero intercept of the straight-line fit gives our best estimate of \(\kappa_{res}/T\) for each \(x\). As is shown in Figs. 1(b) and 1(c), the data for the non-superconducting samples extrapolates to essentially zero, indicating that in these non-superconducting insulator samples the low-temperature \(\kappa\) is essentially phononic with negligible electronic contribution; this is consistent with the observation in insulating \(YBa_2Cu_3O_6.0\) [6]. Note that \(\kappa_{ph}\) becomes \(\sim bT^3\) when phonons are predominantly scattered by the crystal surfaces [13], in which case \(b\) is expressed as \(\beta^3 \langle v_{ph}\rangle/L_{ph}\), where, for LSCO, \(\beta \approx 3.9\) \(\mu\)J/cm\(^3\) is the phonon specific heat coefficient, \(\langle v_{ph}\rangle \approx 4 \times 10^5\) cm/s is the averaged sound velocity, and \(L_{ph}\) is the phonon mean-free path. In perfect crystals \(L_{ph}\) takes the maximum value \(1.12\bar{w}\) with \(\bar{w}\) the geometric mean width of the sample [21]. In our case, \(\bar{w} = 170 - 300\) \(\mu\)m and the fits in Figs. 1(b)-1(m) yield \(b = 2 - 6\) mW/cmK\(^2\), giving \(L_{ph}/1.12\bar{w}\) of 0.2 - 0.8. These ratios are a bit smaller than the ratios of 0.6 - 1.4 for the Y-based cuprates [4,10] (which may be due to the roughness of the polished surface or to the twins [18]), but they are comparable to the \(\beta_G\)Sr\(_2\)CaCu\(_2\)O\(_8\) (Bi-2212) case [22] and are still in the reasonable range [8].

Figure 2 shows \(\kappa_{res}/T\) for all the samples measured as a function of \(x\). One can clearly see that \(\kappa_{res}/T\) starts to grow only above \(x=0.05\) with increasing \(x\), and it is monotonically increasing except for an anomalous dip at \(x=1/8\). The anomaly at \(x=1/8\) is probably due to the charge ordering [23] which would tend to localize the carriers; it is useful to note that a similar anomaly is observed in the \(x\) dependence of the penetration depth [4]. The \(\kappa_{res}/T\) value at optimum doping \((x=0.17)\) is 0.20 mW/cmK\(^2\), which is comparable to the reported values for optimally-doped Y-123 [4] and optimally-doped Bi-2212 [4,7].

The most striking feature in Fig. 2 is probably the smooth evolution of \(\kappa_{res}/T\) across optimum doping \((x \approx 0.16)\). This is in sharp contrast to the behavior of the normal-state \(\rho_{ab}\) under 60 T in the zero-temperature limit [1,2], which shows an insulator-to-metal crossover at optimum doping; namely, \(\rho_{ab}\) diverges as \(T \to 0\) for \(x < 0.16\), while it stays small and finite for \(x > 0.16\) [2]. This contrast can be more quantitatively illustrated by calculating the expected normal-state electronic thermal conductivity in the zero-temperature limit, \(\kappa_{normal}^0\), using the Wiedemann-Franz law \(\kappa_{el}/T = L_0/\rho_{res}\), where \(L_0 = (\pi^2/3)(k_B/e)^2\) is the Sommerfeld value of the Lorentz number and \(\rho_{res}\) is the residual resistivity. (While the validity of this law is not clear in non-Fermi liquids, it should be satisfied in all Fermi liquids at low enough temperature [18].) Since \(\rho_{ab}\) diverges for \(x < 0.16\), \(\kappa_{normal}^0\) is zero in the underdoped region. The data for \(x=0.17\) and 0.22 given in Ref. [2] can be used to calculate \(\kappa_{normal}^0/T\) in the overdoped region, and the results are plotted in Fig. 2 with open squares; clearly, the normal-state \(\rho_{ab}\) suggests \(\kappa_{normal}^0/T\) to grow rapidly above \(x=0.16\), which is at odds with the behavior of \(\kappa_{res}/T\). It is intriguing
that $\kappa_{\text{normal}}/T$ is larger than $\kappa_{\text{res}}/T$ in the overdoped region but their relation switches in the underdoped region; in “conventional” BCS superconductors with a $d$-wave gap, $\kappa_{\text{el}}$ in the normal state should always be much larger than that in the superconducting state, reflecting the difference in the numbers of available heat carriers.

To corroborate the above comparison, we further measured both the normal-state $\rho_{ab}(T)$ under 18 T and $\kappa(T)$ in zero field in the identical sample for $x=0.06$. The main panel of Fig. 3 shows the $\rho_{ab}(T)$ data and the inset shows the comparison of $\kappa_{\text{res}}/T$ with the “expected” normal-state electronic thermal conductivity $\kappa_{\text{el}}^{\text{normal}}$ that would correspond to the $\rho_{ab}$ value; it is clear that $\kappa_{\text{el}}^{\text{normal}}$ for $T \to 0$ is much smaller than $\kappa_{\text{res}}/T$, which is very difficult to understand in the conventional picture of the superconducting condensate.

These comparisons suggest that either the scattering rate for $T \to 0$ in the superconducting state is much smaller than that in the normal state, or the Wiedemann-Franz law is strongly violated in the underdoped cuprates. If the former is the case, it is a highly unusual situation, because any inelastic scattering would normally vanish as $T \to 0$ and the same elastic impurity scattering would dominate the transport in both the normal and the superconducting state; if, on the other hand, the latter is the case, our result is a yet-another strong proof of the non-Fermi-liquid nature of the normal state of the cuprates. In any case, the above observation highlights an unusual contrast between the low-energy physics of the superconducting state and that of the normal state in underdoped LSCO, and this contrast is useful in examining the nature of the strongly-correlated electrons in cuprates. In fact, some of the existing theories offer intriguing possibilities to understand this unusual situation: (a) The nature of the charge carriers may be fundamentally different between the normal and the superconducting states. For example, it might be possible that the superconductivity occurs by avoiding a strong correlation effect that would otherwise localize the holes [23, 29]; in this case, the QPs excited from the superconducting condensate may not be bound to the correlation effect characteristic of the normal state. (b) When there is a quantum critical point (QCP), inelastic scatterings can survive down to very low temperatures because of the critical fluctuations [22]. In this case, inelastic scattering may survive as $T \to 0$ in the normal state, which causes the total scattering rate in the normal state to be significantly larger. (c) It was argued [24] that in a non-Fermi-liquid arbitrarily small concentration of impurities lead to a vanishing density of states at $E_F$ because of the strong electron correlations. In this case, the modification of the density of states may only be reflected in the heat carriers in the normal state. Of course, these three possibilities are just examples of the implication of the data, and our observation of the contrasting low-energy physics in the normal and the superconducting states would serve as a touchstone to test the theory of high-$T_c$ superconductivity.

In addition, the observed evolution of $\kappa_{\text{res}}/T$ clearly demonstrates that the QP transport in the superconducting state is much more complicated than the simple $d$-wave phenomenology suggests. The standard theory predicts [2] that $\kappa_{\text{res}}/T$ is proportional to the ratio $v_F/v_2$ ($v_F$ and $v_2$ are the energy dispersion normal and tangential to the Fermi surface at the node) once the $d$-wave superconductivity is established, and it has been reported that the measured values of $\kappa_{\text{res}}/T$ in Y-123 and Bi-2212 are in reasonable agreement with the theory at optimum doping [4, 5, 17]; therefore, we would expect that with increasing $x$ there is a discontinuous onset of $\kappa_{\text{res}}/T$ upon entering into the superconducting regime. However, what we actually observe is a continuous and gradual increase in $\kappa_{\text{res}}/T$ across $x=0.05$, which contradicts the theoretically expected behavior. We note that the first indication for the breakdown of the simple phenomenology came from the measurements on Y-124 [10], and the present result shows the breakdown of a different nature. Furthermore, a systematic photoemission study of Bi-2212 reported that $v_F/v_2$ decreases with carrier doping [22], which suggests that $\kappa_{\text{res}}/T$ as predicted by the theory should also decrease with increasing $x$. Hence, the results obtained here disagree with the theory in this respect as well, which suggests that either the agreement at optimum doping is accidental, or the doping dependence of the gap structure is not similar in different cuprates (which is improbable), or the standard theory breaks down in the underdoped regime. In any case, the continuity at $x=0.05$ appears to indicate that the QPs in the superconducting state are strongly influenced by the localizing tendency in the normal state [11]: namely, a part of the QPs somehow localize in the heavily-underdoped region and they cannot participate in the extended QP state. The mechanism of thisQP localization in the superconducting state is not clear at this stage; however, the charge stripe ordering should at least partly be responsible, because $\kappa_{\text{res}}/T$ is clearly suppressed at the 1/8 doping. Other mechanisms such as quantum interference effect [4] might also be responsible.

In summary, we find that the simple $d$-wave phenomenology for the QP transport in the superconducting state is not sufficient to explain the observed evolution of $\kappa_{\text{res}}/T$ with hole doping $x$; specifically, the smooth onset of $\kappa_{\text{res}}/T$ across $x=0.05$ suggests some additional mechanism that causes the QP states to localize. Moreover, we demonstrate that the low-energy physics shows a strong contrast between the normal and the superconducting states, which bears intriguing implications on the possible mechanism of superconductivity.

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FIG. 1. $\kappa/T$ of La$_{2-x}$Sr$_x$CuO$_4$ single crystals at low temperatures plotted vs. $T^2$. (a) Cumulative plot for selected $x$ values from 0 to 0.22. (b)-(m) Individual plots of $\kappa/T$ vs. $T^2$ for various $x$; dashed lines are linear fits to the low-$T$ part of the data. Zero-temperature intercept of the dashed line gives the estimate of $\kappa_{\text{res}}/T$.

FIG. 2. $\kappa_{\text{res}}/T$ of LSCO as a function of $x$ (filled circles). The expected normal-state electronic thermal conductivity for $T \to 0$, $\kappa_{\text{normal}}^0$, is estimated from the 60-T data [12] and is shown for comparison (open squares). Solid and dashed lines are guides to the eyes.

FIG. 3. $T$-dependence of $\rho_{ab}$ in 0 and 18 T for $x=0.06$. The same single crystal sample is used for the $\rho_{ab}(T)$ and $\kappa(T)$ [Fig. 1(d)] measurements. The magnetic field is applied parallel to the $c$ axis. Inset: The expected $\kappa_{\text{normal}}^0(T)/T$ for $x = 0.06$ calculated from the $\rho_{ab}(T)$ data in 18 T, plotted together with $\kappa_{\text{res}}/T$ estimated in Fig. 1(d).