Pitfalls in the analysis of low-temperature thermal conductivity of high-\(T_c\) cuprates

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Abstract. Recently, it was proposed that phonons are specularly reflected below \(~0.5\) K in ordinary single-crystal samples of high-\(T_c\) cuprates, and that the low-temperature thermal conductivity \(\kappa\) should be analyzed by fitting the data up to \(~0.5\) K with the formula \(\kappa = aT + bT^n\), where \(a\) is a fitting parameter. Such an analysis yields a result different from that obtained from the conventional analysis in which the fitting is restricted to a region below \(~0.15\) K, where \(\kappa\) has been found to obey \(\kappa = aT + bT^n\) very well, which means that phonons are in the boundary scattering regime without specular reflection. Here we show that the proposed new analysis is most likely flawed, because the specular phonon reflection means that the phonon mean free path \(\ell\) gets longer than the mean sample width, while the estimated \(\ell\) is actually much shorter than the mean sample width above \(~0.15\) K.

1. Introduction

Because high-\(T_c\) cuprate superconductors have a \(d\)-wave superconducting gap that has four “nodes” where the gap vanishes, there are low-energy quasiparticles (QPs) in the superconducting state of cuprates [1]. Those QPs, called nodal QPs, are extended and have well-defined wave vectors, and they can carry heat even at very low temperature. The thermal conductivity \(\kappa\) of cuprates below \(T_c\) has turned out to be a useful probe for studying the nature of the superconducting state, because the physics of the nodal QPs are directly reflected in \(\kappa\). In particular, at very low temperature where the QPs are in the residual elastic scattering regime, the electronic thermal conductivity \(\kappa_e\) is expected to be approximated by a “universal” thermal conductivity \(\kappa_0\), which is expressed as [2, 3]

\[
\frac{\kappa_0}{T} = \frac{k_B^2 n}{3h} \left( \frac{v_F}{v_2} + \frac{v_2}{v_F} \right) \approx \frac{k_B^2 n}{3h} \frac{v_F}{v_2},
\]

where \(n\) is the number of CuO\(_2\) planes per unit cell, \(c\) is the \(c\)-axis lattice constant, and \(v_F\) (\(v_2\)) is the QP velocity normal (tangential) to the Fermi surface at the gap node. This \(\kappa_0\) is called “universal” in the sense that it is expected to be independent of the QP scattering rate \(\gamma\); this happens because the effects of disorder to create QPs and to scatter them would exactly cancel each other in a certain range of \(\gamma\) [1]. Thus, the bulk measurement of \(\kappa_e\) at low enough temperature would give us microscopic information about the gap.

However, in metals both electrons and phonons carry heat, so \(\kappa\) is a sum of \(\kappa_e\) and the phonon thermal conductivity \(\kappa_p\). This causes a fundamental complication in the data analysis, that is,
one has to separate $\kappa_e$ from $\kappa_p$ in the measured $\kappa$. Such a separation can be accomplished if $\kappa_e$ and $\kappa_p$ have distinct and well-defined temperature dependences; actually, at low enough temperature, $\kappa_e \simeq aT$ is expected in the residual elastic scattering regime, and $\kappa_p \simeq bT^3$ is expected in the boundary scattering regime of phonons. Hence, experimentalists often employ dilution refrigerators to measure the thermal conductivity in the mK region, where the heat transport is usually found to be in the above-described regime. Indeed, in the case of cuprates, we have recently shown [4] that the description $\kappa = aT + bT^3$ holds very well in superconducting YBa$_2$Cu$_3$O$_y$ (YBCO) samples below $\sim 110$ mK.

Recently, there is a controversy in the high-$T_c$ community as to how best the $\kappa(T)$ data at low temperature are analyzed to separate $\kappa_e$ from $\kappa_p$. In a paper reporting the low-temperature $\kappa(T)$ behavior in YBCO and La$_{2-x}$Sr$_x$CuO$_4$ (LSCO), Sutherland et al. argued [5] that in single-crystal samples of those materials, the specular reflection of phonons becomes important at temperatures below $\sim 0.5$ K, and hence the analysis of the low-temperature data should take this effect into account. The new analysis proposed in Ref. [5] was adopted in subsequent papers from the same group, such as Refs. [6, 7, 8, 9, 10], and has been playing crucial roles in deducing the conclusions of those papers. In this article, we point out that the key assumption behind the new analysis, that the phonons are specularly reflected at $T$ up to $\sim 0.5$ K, is erroneous, by showing that the phonon mean free path is much shorter than the mean sample width in most of the proposed temperature range, which means that phonons are not even in the boundary scattering regime that is a prerequisite for the dominance of specular reflections.

2. Specular Phonon Reflection?

The standard way of analyzing the low-$T$ thermal conductivity data $\kappa(T)$ in terms of $\kappa/T = a + bT^2$, which was first employed in the present context by Taillefer et al. [11] for both insulating and superconducting YBCO, has proved to be reliable for all cuprate systems studied so far [12, 13, 14, 15, 16, 17]. The physical ground of this analysis is that the $T$ dependence of $\kappa$ does not change further at lower $T$ when the electrons are elastically scattered and the phonons are scattered by the boundary, and hence one can safely assume the formula $\kappa/T = a + bT^2$ to be valid down to $T = 0$ K, which gives confidence in the $T \rightarrow 0$ extrapolation; note that in the boundary scattering regime the phonons are usually diffusively scattered at the sample surface [18] and the phononic contribution $\kappa_p$ is simply proportional to the specific heat, yielding $\kappa_p = bT^3$. However, Sutherland et al. had advocated in their paper [5] that the specular reflection of phonons causes a $T$ dependence of $\kappa_p$ weaker than $T^3$ in the boundary scattering regime, and that a fitting of the data for a wider temperature range to $\kappa/T = a + bT^{\alpha-1}$ with $\alpha < 3$ is better than the standard one. In the following, since the analysis of the $\kappa(T)$ data of underdoped YBCO has been the most controversial [4, 8, 9] after the publication of Ref. [5], we focus our discussion on the case of YBCO.

When the phonons are specularly reflected at the surface, $\kappa_p$ becomes dependent on the averaged phonon wave length that changes with $T$, and empirically $\kappa_p$ is expressed as $bT^\alpha$ with $\alpha < 3$ [18, 19]. Past studies on insulating materials unanimously found [18, 19, 20] that the phonon mean free path $\ell$ becomes much longer than the mean width $W$ of the sample when the specular reflection becomes important, as is naturally expected ($W$ is usually taken to be $2/\sqrt{\pi}$ times the geometrical mean width $\bar{w}$, see Ref. [11]). Therefore, calculating $\ell$ in YBCO and comparing it with $W$ is a good way of judging whether the phonons are indeed specularly reflected. This can be rather easily done, because $\kappa_p$ is equal to $\frac{1}{2}C\bar{v}\ell$ where $C = 3\beta T^3$ is the phonon specific heat and $\bar{v}$ is the averaged sound velocity, and both $\beta$ and $\bar{v}$ are known for YBCO [11]. Figure 1 shows the temperature dependence of $\ell$ calculated with the data reported by Taillefer et al. [11] for $y = 0$ (where $W$ was specified) and with our own data [4] for $y = 6.45$ [21]. One can see that $\ell$ becomes saturating and comparable to $W$ only at the lowest temperature, and it stays smaller than $W$ in the temperature range where Sutherland et
al. argued [8, 5] that the specular reflection is taking place (0.07 – 0.5 K). Therefore, Fig. 1 demonstrates that the assertion of Sutherland et al. is dubious and the specular reflection is not important in YBCO. A natural conclusion is that in YBCO at temperatures higher than ~120 mK phonons are not in the boundary scattering regime [11, 15, 16] and the phenomenological $T^\alpha$ ($\alpha < 3$) dependence comes from some additional scattering (such as scatterings off point defects, dislocations, twin boundaries, etc.) that only reduces $\ell$ [22]; such additional scattering would eventually die away at lower $T$ as the boundary scattering regime is achieved, so the $T^\alpha$ fitting should not be used for the $T \to 0$ extrapolation.

3. Validity of Extrapolations
As a matter of fact, one must always bear in mind that, if an extrapolation is to be employed in the analysis, the formula used for the extrapolation should have a physical reasoning which assures that the same functional form holds throughout the range of the extrapolation. Otherwise, it is always possible that the true temperature dependence of the system changes below the experimental range (due, for example, to a change in the dominant scattering mechanism) and the extrapolation gives an inadequate estimate for the $T = 0$ value. In the present case, when the assumption of the specular phonon reflection fails, there is no physical ground to expect the phenomenological $T^\alpha$ dependence of $\kappa_p$ to continue to $T = 0$, and hence the conclusion about the electronic contribution at $T = 0$ loses its footing.

4. Conclusion
We have shown that in high-quality YBCO single crystals, the phonons are not likely to be specularly reflected at ~0.5 K because the estimated phonon mean free path is much shorter than the mean sample width at such relatively high temperatures. Hence, the data analysis assuming the specular phonon reflection lacks its physical ground. When analyzing the low-temperature thermal conductivity data, one should avoid falling into pitfalls of fitting the data using some accidental power law and extrapolating it to zero temperature, when there is no particular reason to expect to its validity.
Acknowledgment

Y.A. was supported by KAKENHI 16340112 and 19674002 provided by the Japan Society for the Promotion of Science, and X.F.S. acknowledges the support from the National Natural Science Foundation of China (10774137 and 50421201) and the National Basic Research Program of China (2006CB922005).

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