Doping dependence of the superconducting gap in $Tl_2Ba_2CuO_{6+\delta}$ from heat transport

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We present low-temperature thermal conductivity measurements on the cuprate $Tl_2Ba_2CuO_{6+\delta}$ throughout the overdoped regime. In the $T\rightarrow 0$ limit, the thermal conductivity due to d-wave nodal quasiparticles provides a bulk measurement of the superconducting gap Δ. We find Δ to decrease with increasing doping, with a magnitude consistent with spectroscopic measurements (photoemission and tunneling). This argues for a pure and simple d-wave superconducting state in the overdoped region of the phase diagram, which appears to extend into the underdoped regime down to a hole concentration of $\simeq 0.1$ hole/Cu. As hole concentration is decreased, the gap-to-T_c ratio increases, showing that the suppression of the superconducting transition temperature T_c (relative to the gap) begins in the overdoped regime.

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Over the past decade, low-temperature thermal conductivity has emerged as a powerful probe of low-energy quasiparticle excitations in the cuprates. Unlike conventional s-wave superconductors, the nodes in the d-wave superconducting order parameter of cuprates lead to a finite quasiparticle density of states at zero energy, resulting in "universal" transport in the low-temperature limit ($T\rightarrow 0$).[1,2] By performing measurements in the $T\rightarrow 0$ limit, the complicated physics of quasiparticle transport in these materials is greatly simplified and meaningful quantitative comparisons between mean-field theories in the self-consistent T-matrix approximation[3–5] (SCTMA) and experiments have been made.[6–8]

In the mean-field theory, the low-T thermal conductivity is proportional to the slope of the gap near the nodes, $\delta\Delta/\delta\phi|_{\phi\rightarrow\pi/4}$. By assuming a gap with a simple d-wave form $[\Delta=\Delta_0\cos(2\phi)]$, the gap maximum Δ_0 can be inferred from $\delta\Delta/\delta\phi$, making thermal conductivity a measure of the superconducting gap. This measure is notable for a number of reasons that make it an excellent complement to more conventional, spectroscopic measurements of the gap. First, it is a very-low-energy measurement of the gap, the energy scale being set by temperature (and impurity bandwidth γ). Second, it measures the gap near the nodes [in the $\Gamma\rightarrow(\pi,\pi)$ direction] as opposed to the antinodes $(\pi,0)$. Finally, thermal conductivity provides a bulk measure of the gap, unlike surface-sensitive techniques such as angle-resolved photoemission spectroscopy (ARPES) and tunneling. Together these properties allow one to impose important constraints on the nature of superconductivity in cuprates.

The cuprate $Tl_2Ba_2CuO_{6+\delta}$ (Tl-2201) is an ideal material for quantitative investigations of d-wave superconductivity in the cuprates. It is a single-layer material with $T_c^{max}\simeq 90$ K, comparable to the well-studied cuprates $Bi_2Ba_2CaCu_2O_{8+\delta}$ (Bi2212) and $YBa_2Cu_3O_y$ (YBCO). By varying the oxygen concentration, the material can be doped from optimal doping far into the overdoped region of the phase diagram. In this region, the metallic state is well characterized in terms of a single coherent Fermi surface and there are no indications of competing phases, such as encountered in $La_{2-x}Sr_xCuO_4$ (LSCO), for example. Moreover, Tl-2201 can be prepared with reasonably low levels of disorder because the dopant atoms are interstitial oxygen residing well away from the CuO_2 planes, instead of the more obtrusive cation doping located close to the CuO_2 planes (as in LSCO, for example).

In this paper, we present low-temperature thermal conductivity measurements of Tl-2201 as a function of doping, spanning the overdoped region of the phase diagram. Expanding upon previous work on cuprates,[1,2,6–8] we analyze our results using the mean-field SCTMA theory to provide a determination of the superconducting gap from thermal conductivity. We find agreement between thermal conductivity and spectroscopic measurements of the gap, confirming both the d-wave form of the superconducting gap and the validity of the mean-field description of low-temperature thermal conductivity in overdoped Tl2201. We track the gap as a function of doping throughout the overdoped region and find a gap-to-T_c ratio $\mu\equiv\Delta_0/k_BT_c$ that increases with decreasing doping, starting with a value close to the weak-coupling BCS value ($\mu=2.14$) at high doping and reaching a value twice as large near optimal doping ($\mu=5-6$). This shows that the suppression of the onset of coherent superconductivity (or T_c), so characteristic of the underdoped regime, actually begins above optimal doping.

Five flux-grown single crystals of $Tl_2Ba_2CuO_{6+\delta}$ are used in this study.[9,10] The platelet samples have approximate length and width of $\sim$0.5 mm in the ab plane and thickness $\times 20-65$ μm along the c axis. The heat current is applied in the ab plane and the magnetic field along the c axis. The concentration of holes in the CuO_2 plane, p, is set by tuning the excess oxygen content in the TlO_2 layer. Concentrations between optimal doping and heavily overdoped were achieved by annealing the samples in atmospheres ranging

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TABLE I. Residual linear term κ_0/T in the thermal conductivity of Tl-2201 from fits of κ/T vs T to Eq. (2) for five single crystals with hole concentration p estimated from measured T_c. The gap maximum Δ_0 is evaluated from κ_0/T using Eq. (5). The error applies to κ_0/T, Δ_0, and Δ_0/k_BT_c.

T_c (K)	p	κ_0/T (mW K^{-2} cm^{-1})	Δ_0 (meV)	Δ_0/k_BT_c	Error (%)
84[a]	0.188	0.08	40	5.6	17
76	0.203	0.15	22	3.4	16
72	0.209	0.14	23	3.8	15
68	0.214	0.12	28	4.8	25
27	0.252	0.34	9.5	4.1	24
26[a]	0.253	0.48	6.7	3.0	17

[a]Denotes the same sample measured at two dopings.

from vacuum ($\sim 5\times 10^{-7}$ torr) at 500 °C near optimal doping to 50–60 mbar oxygen at 400–480 °C for heavily overdoped samples. The value of p for each sample was estimated from the measured T_c (defined as the temperature at which the electrical resistivity has fallen to zero), using the well-known empirical formula[11]

$$\frac{T_c}{T_c^{max}} = 1 - 82.6(p-0.16)^2, \tag{1}$$

where T_c^{max}=90 K is taken as the nominal T_c of optimally doped Tl-2201. The values of T_c and p for each sample are listed in Table I.

Thermal conductivity κ was measured down to 50 mK in a dilution refrigerator using a standard four-wire steady-state technique described elsewhere.[8] Electrical contacts to the samples were made using evaporated gold pads, which were annealed to improve the contact conductance in the same anneal used to set the oxygen content. Silver wires were then attached to the contact pads using silver paint.

The largest error in the magnitude of κ, ranging from 15% to 25% (see Table I), comes from the measurement of sample dimensions for the appropriate geometric factor. In order to eliminate this dominant source of uncertainty when comparing two different hole concentrations, one of the samples was measured twice with the same contacts, at two different dopings, T_c=84 K and 26 K (see data in Fig. 1). This was achieved by first evaporating gold pads onto the sample and annealing the sample to set the doping, as described above. After measuring the sample, the wires and silver paint were removed and the sample was reannealed to a different doping and subsequently remeasured with the gold pads still in place.

The thermal conductivity measured below 1 K is shown in Fig. 1 for this sample and in Fig. 2 for four other samples, each at a different doping. To extract the electronic contribution to κ the data are fit to

$$\frac{\kappa}{T} = \frac{\kappa_0}{T} + AT, \tag{2}$$

where κ_0/T (intercept) is the electronic contribution to the total thermal conductivity and the second term (slope) is the phonon contribution, $\kappa_{ph}/T=AT$. The values of κ_0/T from fits to Eq. (2) are listed in Table I, along with the error bars for each sample, estimated from the statistical error in the fits ($<1.5\%$) and the error in the geometric factor (15%–25%). These results can be compared to a previous study on a strongly overdoped Tl-2201 crystal,[7] with T_c=15 K, which gave κ_0/T=1.4 mW K^{-2} cm^{-1} and an excellent fit to Eq. (2).

Note that in these overdoped samples $\kappa_{ph}\propto T^2$ (over a decade in temperature). This is in contrast to previous reports of the low-temperature thermal conductivity of cuprates, measured typically at or below optimal doping, where κ_{ph}

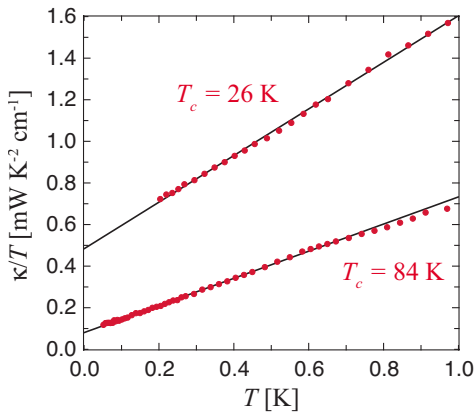

FIG. 1. (Color online) Thermal conductivity of one Tl-2201 sample, measured at two different hole concentrations, as indicated by the respective T_c values, plotted as κ/T vs T below 1 K. The lines are fits to Eq. (2). A threefold decrease in T_c corresponds to a sixfold increase in the residual linear term [which is inversely proportional to the superconducting gap, via Eq. (5)], hence a twofold decrease in the gap-to-T_c ratio with doping.

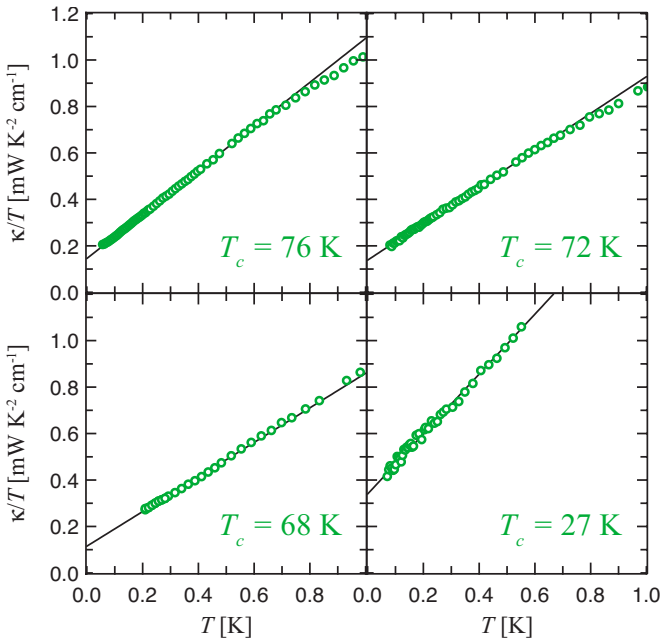

FIG. 2. (Color online) Thermal conductivity of four Tl-2201 samples, each with a different hole concentration, as indicated by their respective T_c values, plotted as κ/T vs T below 1 K. The lines are fits to Eq. (2).

$\propto T^{2.5-3}$, consistent with phonons being scattered from the boundaries of the sample.[1,2,6,8,12,13] In overdoped Tl-2201, with its higher carrier concentration, we attribute the T^2 dependence of κ_{ph} to dominant electron-phonon scattering, known to yield $\kappa_{ph} \propto T^2$ at low T in a metal.[14] Smith has shown that this is indeed the theoretically expected dependence of phonon transport in a d-wave superconductor[15] in the range 0.1–1.0 K. We emphasize that the main results of this paper, which stem from the doping dependence of κ_0/T, are unaffected by alternative methods to fit κ_{ph}, such as a power law with adjustable power ($\kappa/T = a + bT^{\gamma}$).

The T-linear electronic contribution to κ is due to transport from d-wave nodal quasiparticles in the limit of $k_B T \ll \gamma \ll \Delta_0$, where γ is the impurity bandwidth.[1,4,5] In this limit, the residual linear term in the thermal conductivity is *universal*, in the sense that it is independent of scattering rate and given by[5]

$$\frac{\kappa_0}{T} = \frac{k_B^2}{3\hbar}\frac{n}{c}\left(\frac{v_F}{v_\Delta} + \frac{v_\Delta}{v_F}\right), \tag{3}$$

where n is the number of CuO_2 planes per unit cell (=2 for Tl-2201) and c is the c-axis lattice constant (=23.2 Å for Tl-2201). The ratio v_F/v_Δ is the ratio of quasiparticle velocities normal and tangential to the Fermi surface at the node, respectively given by the Fermi velocity in the $(0,0)$-(π,π) direction, v_F, and

$$v_\Delta = \frac{1}{\hbar k_F}\left.\frac{d\Delta}{d\phi}\right|_{node}, \tag{4}$$

proportional to the slope of the gap at the node. Note that, unique among transport properties, κ_0/T was shown to be robust against both Fermi liquid and vertex corrections.[5]

The above formalism, however, is only valid in the limit $k_B T \ll \gamma \ll \Delta_0$. Before proceeding, it is important to determine whether this criterion is indeed satisfied for our samples, particularly since Δ_0 decreases with increasing doping and eventually goes to zero at the superconductor-metal phase transition.[16] In the Appendix, we provide two independent and consistent estimates of γ for each sample which show that in all cases $\gamma/k_B T > 10$ (for $T < 1$ K) and $\gamma/\Delta_0 < 0.16$, validating the use of Eq. (3) for our Tl-2201 samples.

The significance of Eq. (3) is that it provides a reliable way to measure the gap from a rather straightforward measurement of the thermal conductivity. Indeed, through Eqs. (3) and (4), the slope of the gap at the node, $\delta\Delta/\delta\phi|_{node}$, is immediately obtained from κ_0/T, provided v_F and k_F are known.[6,8] Making the assumption that Δ has the standard $d_{x^2-y^2}$ form $\Delta(\phi) = \Delta_0 \cos(2\phi)$, Eq. (3) can be rewritten (for $v_F \gg v_\Delta$) as

$$\frac{\kappa_0}{T} \simeq \frac{k_B^2}{6}\frac{n}{c}k_F\frac{v_F}{\Delta_0}. \tag{5}$$

κ_0/T can thus be related in a straightforward fashion to the gap maximum Δ_0 by making use of the fact that v_F and k_F at the nodes are approximately doping independent. ARPES measurements[17] have shown v_F to be independent of both doping and material and to have a value $v_F = 2.7 \times 10^7 \pm 20\%$ cm/s. Measurements of the Fermi surface by ARPES in LSCO,[18] Bi-2212,[19] and Tl-2201[22] show k_F to have a value ~ 0.7 Å^{-1} along the nodal direction $(0,0)$-(π,π), essentially independent of material or doping [k_F only has significant doping dependence along $(0,0)$-$(0,\pi)$]. In overdoped Tl-2201, this value of k_F was nicely confirmed by bulk-sensitive angular magnetoresistance oscillation (AMRO) measurements.[20] The values of Δ_0 obtained for our five Tl-2201 samples from Eq. (5) are listed in Table I and plotted vs p in the top panel of Fig. 3. Also shown is the value of Δ_0 for an ultrapure overdoped YBCO crystal,[8,21] obtained in the same way (with $\kappa_0/T = 0.16$ mW K^{-2} cm^{-1})—it is in excellent quantitative agreement with the Tl-2201 data.

Δ_0 decreases with increasing doping and follows the doping dependence of the gap measured by spectroscopic probes

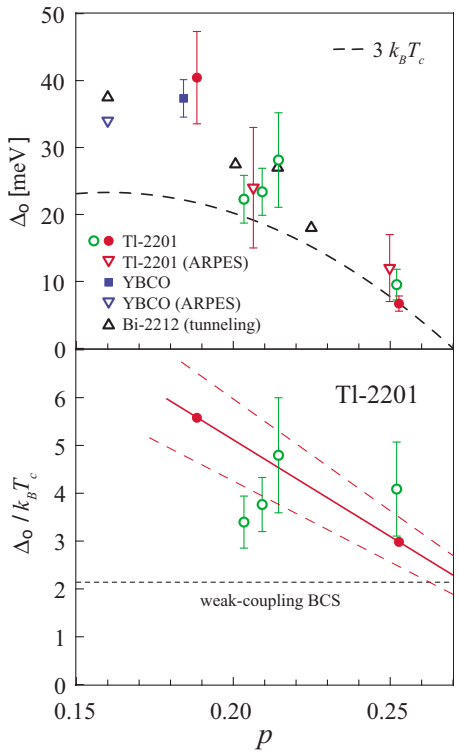

FIG. 3. (Color online) *Upper panel*: the gap maximum Δ_0 vs hole concentration p derived from thermal conductivity measurements on Tl-2201 (circles) and YBCO (square, Refs. 8 and 21). The solid (red) circles denote the same Tl-2201 sample measured at two different doping levels, as discussed in the text. Spectroscopic measurements of the gap from ARPES on Tl-2201 (red down-triangle, Refs. 9 and 22) and YBCO (blue down triangle, Ref. 23) and SIS tunneling on Bi-2212 (black up-triangles, Ref. 24) are also shown. The dashed line gives the BCS doping dependence of the gap based on the measured T_c—namely, $\Delta_0 = \mu k_B T_c$—using a strong-coupling value of $\mu = 3$ and $T_c(p)$ from Eq. (1). *Lower panel*: the gap-to-T_c ratio $\Delta_0/k_B T_c$ vs p for Tl-2201 from thermal conductivity data. The solid (red) line is a fit through the two data points (solid red circles) obtained on a single sample, for which the relative uncertainty is minimal ($\sim 3\%$), given the fixed geometric factor. The dashed (red) lines on either side reflect the uncertainty on the absolute value ($\pm 17\%$; see Table I). The horizontal (black) dashed line is the weak-coupling d-wave BCS prediction: namely, $\Delta_0/k_B T_c = 2.14$ (Ref. 25).

such as tunneling[24,26] and ARPES.[27] In Fig. 3 we also plot the doping dependence of Δ_0 in Bi-2212 measured using SIS tunneling[24] and find good quantitative agreement with Δ_0 measured by thermal conductivity,[28] similar to previous work on optimally doped Bi-2212.[6] Comparing directly to measurements on Tl-2201, recent ARPES measurements give $\Delta_0 \simeq 12\pm5$ meV and $\Delta_0 \simeq 24\pm9$ meV in samples with $T_c = 30$ K and 74 K, respectively,[9,22] which is also in favorable agreement with thermal conductivity. [Δ_0 here is taken as the average of two commonly used measures of Δ_0: the peak position and the leading-edge midpoint, which are 17 meV (33 meV) and 8 meV (15 meV), respectively, for $T_c = 30$ K (74 K).]

The agreement between the different measures of Δ has important implications. First, it provides further compelling evidence for coherent nodal quasiparticle excitations that are correctly described by mean-field *d*-wave quasiparticle theory throughout the overdoped region of the phase diagram. Second, the fact that the gap derived from thermal conductivity (i.e., from low-energy *nodal* quasiparticles) agrees well with the gap maximum derived from tunneling or ARPES (determined from *antinodal* quasiparticles at higher energies) suggests that the gap structure (as a function of angle) is not very different from the simple *d*-wave gap assumed in our analysis [$\Delta_0\cos(2\phi)$]. This places limits on any subdominant order parameter that may exist in the bulk. Subdominant order parameters that result in a fully gapped nodal region (such as *is* or *id*) are limited to having a magnitude much less than the impurity bandwidth γ,[29] which is estimated to be on the order of $\Delta_0/10$ in our Tl-2201 samples (see the Appendix). An even more stringent restriction is provided by the YBCO measurements, where, in crystals of the highest quality, γ can be orders of magnitude smaller than in Tl-2201.[8,21] These conclusions are in agreement with the phase-sensitive tricrystal experiments, which show $d_{x^2-y^2}$ symmetry of the order parameter and no signature of a subdominant *imaginary* contribution to the superconducting order parameter away from optimal doping.[30] (Note that a *real s* component[31] is allowed.)

The gap-to-T_c ratio $\mu = \Delta_0/k_B T_c$ is plotted vs p in Fig. 3. μ is seen to decrease with increasing doping from a strong-coupling BCS value of $\mu = 5-6$ near optimal doping towards the weak-coupling value of $\mu = 2.14$ at high doping. This trend is made compelling by comparing measurements on the same sample at two different dopings (solid red circles), for which the relative uncertainty is small. Indeed, for sample *a* (see Table I), μ drops by a factor of 1.9 (from 5.6 to 3.0) when the doping is increased from $p = 0.19$ ($T_c = 84$ K) to $p = 0.25$ ($T_c = 26$ K).

Having established how the superconducting gap grows with decreasing doping in the overdoped regime of the phase diagram, it is natural to ask how it continues to evolve below optimal doping into the enigmatic underdoped regime. Heat transport measurements on YBCO have shown that κ_0/T decreases monotonically, at least down to $p \simeq 0.1$.[8,32,33] Based on Eq. (5), this was interpreted in terms of a gap Δ that keeps on rising as p is reduced.[8] Note, however, that recent measurements of the in-plane resistivity in the (field-induced) normal state of YBCO reveal a "metal-insulator" crossover at $p \simeq 0.09$.[34] One should not expect the standard BCS analysis [leading to Eq. (3)] to apply below that crossover. (The same situation was encountered in LSCO,[8] setting in at a considerably higher doping since the "metal-insulator" crossover in that system is at $p \simeq 0.17$.[35]) The crossover to a nonmetallic normal state may account for the qualitative change observed in the behavior of κ_0/T when p is reduced to 5%–6%, where it becomes independent of both doping and magnetic field.[36] This overall evolution of κ_0/T vs p in YBCO is corroborated by recent ARPES measurements on Bi-2212, which show the gap near the nodes to have comparable magnitude and to rise with decreasing p until $p \simeq 0.08$, and then fall abruptly.[37]

In conclusion, we have measured the doping dependence of the low-temperature thermal conductivity in the overdoped cuprate Tl-2201. Analyzing the magnitude of κ_0/T using mean-field theory, we show quantitative agreement between the slope of the gap at the nodes from thermal conductivity and spectroscopic measurements of the gap maximum at the anti-nodes in overdoped cuprates. These measurements provide further confirmation of the applicability of mean-field SCTMA transport theory in the cuprates and, more generally, the basic description of overdoped cuprates as *d*-wave superconductors. As a function of reduced doping, the gap-to-T_c ratio increases by nearly a factor of 2 from $p = 0.25$ to $p = 0.19$. This reveals that the strong suppression of phase coherence (or T_c) characteristic of the underdoped regime, presumably caused by a combination of pair-breaking and phase fluctuations, in fact begins in the overdoped region, at least as high as $p \simeq 0.2$, perhaps higher.

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APPENDIX: ESTIMATES OF THE IMPURITY BANDWIDTH

In this appendix we estimate the impurity bandwidth γ using two approaches: from the normal-state transport and from the magnetic field dependence of κ_0/T in the superconducting state. Both approaches yield similar values of γ which in all samples satisfy the two criteria for the validity of a "universal" limit analysis: namely, $\gamma \ll \Delta_0$ and $T \ll \gamma$.

From normal-state transport

First, we estimate γ from the normal-state quasiparticle scattering rate Γ_N. Because the normal state of our samples could not be reached with the magnetic field available in our lab (15 T), we turn to a previous study[7] on a crystal with a similar quality but significantly lower T_c (higher doping): namely, 15 K. In that crystal, Γ_N was estimated from κ_N/T to be 0.52 meV.[7] Using this value as representative of Γ_N in our samples, we can estimate an upper bound on γ via the relation $\gamma \simeq 0.63\sqrt{\Gamma_N \Delta_0}$, valid in the unitarity limit (which yields

TABLE II. Estimates of the impurity bandwidth γ from the normal-state scattering rate Γ_N=0.52 meV in the unitary limit.

T_c (K)	p	γ (meV)	Δ_0 (meV)	γ/Δ_0
84	0.188	2.6	40	0.065
76	0.203	2.0	22	0.088
72	0.209	2.0	23	0.085
68	0.214	2.2	28	0.078
27	0.252	1.3	9.5	0.134
26	0.253	1.1	6.7	0.16

the largest γ for a given value of Γ_N).[38] These estimates are listed in Table II, based on the Δ_0 values of Table I. One can see that $\gamma/k_BT>13$ for $T<1$ K and $\gamma/\Delta_0\simeq 0.1$ in all cases, more precisely ranging from 0.07 to 0.16. When γ is not negligible, there is a known correction to Eq. (3) [and Eq. (5)], which for $\gamma/\Delta_0\simeq 0.1$ is about 20% (Ref. 39); i.e., the clean-limit gap value is 20% larger than estimated using Eq. (5). An error of this magnitude has little impact on the conclusions of this article.

From the magnetic field dependence of κ_0/T

As a crosscheck, we can also make use of the magnetic field dependence of the thermal conductivity in the $T\to 0$ limit to estimate γ in our samples. Unlike the zero-field thermal conductivity, which is independent of γ, the field dependence of the thermal conductivity is dependent upon the quasiparticle scattering rate.[40,41] In a d-wave superconductor κ_0/T increases with an applied magnetic field due to the Volovik effect[42] and in optimally doped cuprates[13,41,43] the thermal conductivity is well described by semiclassical calculations of the field dependence of κ_0/T based on the Volovik effect.[40,44]

An analytic model of $\kappa_0(H)/T$ for $H\ll H_{c2}$ and $\mathbf{J}\perp\mathbf{H}$ by Kübert and Hirschfeld[40] gives

$$\frac{\kappa(H)/T}{\kappa_0/T}=\frac{\rho^2}{\rho\sqrt{1+\rho^2}-\sinh^{-1}\rho}, \tag{A1}$$

where κ_0/T is the universal value in zero field [Eq. (3)] and (for $\gamma>E_H$)

$$\rho=\sqrt{6/\pi}\gamma/E_H. \tag{A2}$$

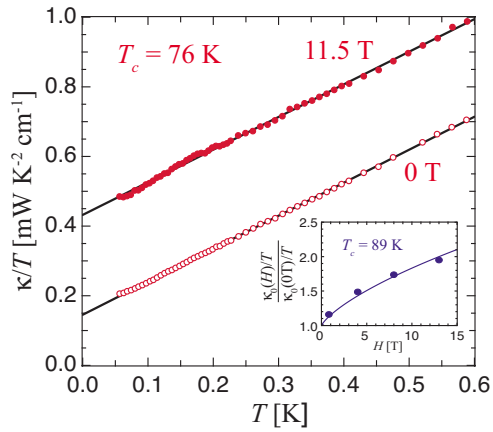

FIG. 4. (Color online) Thermal conductivity in an applied field ($H\|c$) for the Tl-2201 sample with T_c=76 K plotted as κ/T vs T. κ_0/T increases by a factor of ~3 in an applied field of 11.5 T. The lines are fits to Eq. (2). Inset: the in-field electronic thermal conductivity $\kappa_0(H)/T$ normalized to the zero-field value $\kappa_0(0)/T$ as a function of magnetic field in a Tl-2201 sample with T_c=89 K (not used in the main study). The solid line is a fit to the semiclassical model [Eq. (A1)], which yields $\gamma/\Delta_0\simeq 0.1$ (see text).

Here $E_H=a\hbar\sqrt{2/\pi}v_F\sqrt{H/\Phi_0}$ is the average energy shift experienced by a quasiparticle due to the Volovik effect, a is a constant dependent upon the geometry of the vortex lattice (=1/2 for a square lattice[45]), and Φ_0 is the quantum of flux. Note that aside from γ, Eq. (A1) is only dependent upon known constants. As such, we can estimate γ from a single-parameter fit to the field dependence of κ_0/T at $H\ll H_{c2}$.

In Fig. 4, we show κ/T vs T for the Tl-2201 sample with T_c=76 K in H=0 T and 11.5 T. We see that κ_0/T is enhanced by a factor of 3 or so. Similar field-induced enhancements of κ_0/T are observed in all samples by a factor which varies from 2 to 4.[46] In the inset, we show $\kappa_0(H)/T$ [normalized to $\kappa_0(0)/T$] vs H in a separate Tl-2201 sample (T_c =89 K).[47] Equation (A1) provides a reasonable fit (solid line) to the data, giving $\rho\sqrt{H}$=3.36 T$^{1/2}$, which results in γ =3.8 meV, so that $\gamma/\Delta_0\simeq 0.1$.This is similar in magnitude to our above estimate from normal-state transport, providing a nontrivial validation for both methods of estimating γ.

We conclude that within possible corrections on the order of 20% or so, it is legitimate to use Eqs. (3) and (5) to extract an estimate of Δ_0 from a measurement of κ_0/T in the Tl-2201 samples considered here.

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