Absence of residual quasiparticle conductivity in the underdoped cuprate YBa$_2$Cu$_4$O$_8$

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We report here measurements of the in-plane thermal conductivity $\kappa$ of the stoichiometric underdoped cuprate YBa$_2$Cu$_4$O$_8$ (Y124) below 1K. $\kappa(T)$ is shown to follow a simple, phononic $T^3$ dependence at the lowest $T$ for both current directions, with a negligible linear, quasiparticle contribution. This observation is in marked contrast with behavior reported in optimally doped cuprates, and implies that extended zero-energy (or low-energy) quasiparticles are absent in Y124.

Experimental evidence for $d$-wave superconductivity in high-$T_c$ cuprates is now well established. The presence of nodes in the gap is expected to produce a finite density of well-defined quasiparticle (QP) excitations at low energies that dominate the low-$T$ physics. For a pure $d$-wave superconductor with line nodes on the Fermi surface, for example, the density of states (DOS) is linear in the excitation energy, giving rise to a $T^2$ dependence of the low-$T$ specific heat and thermal conductivity (assuming a constant scattering rate). This excitation spectrum, however, is altered significantly in the presence of impurities. For an anisotropic two-dimensional (2D) superconductor with scattering in the unitary limit, a band of impurity states is expected to develop whose width $\gamma$ grows with increasing impurity concentration $n_{imp}$, leading to a finite zero-energy DOS. Lee showed that the residual conductivity is independent of $n_{imp}$, the result of compensation between the increased DOS and a reduction in the associated transport lifetime. This residual, or "universal" conductivity develops at low $T$ in the so-called "dirty" limit, $k_BT \leq \gamma$. Similarly, the low-$T$ $\kappa(T)$ will be dominated by QP states in the vicinity of the line nodes, and is given by the expression:

$$\frac{\kappa_{res}}{T} \approx \frac{(n/d)(k_B^2/3)(v_F/v_2)}{T}$$

where $n/d$ is the stacking density of CuO$_2$ planes and $v_F$ and $v_2$ are the energy dispersions (QP velocities) perpendicular and tangential to the Fermi surface respectively.

The issue of localization of electronic states in $d$-wave superconductors, however, is a complex problem and many competing viewpoints prevail. Balatsky and Salkola, for example, argue that the long-range nature of hopping between impurity states along the nodal directions leads to strong overlap of the impurity wave functions along the diagonals of the square lattice, and ultimately to "extended" impurity states. Senthil and co-workers, on the other hand, argue that quantum interference effects destabilize the extended QP states and lead to a vanishing DOS at zero energy (See also for a similar conclusion). They coined the term "superconducting insulator" to describe such a superconductor with localized states.

Experimentally, the observation of a finite linear term in $\kappa(T)$ in pure and Zn-doped YBa$_2$Cu$_3$O$_{7-\delta}$ (Y123) by Taillefer et al. appeared to confirm the existence of zero-energy quasiparticles. Moreover, the size of this term was indeed found to be "universal", i.e. independent of Zn concentration, in agreement with Lee's prediction. Similar behavior was later reported for Bi$_2$Sr$_2$CaCu$_2$O$_{8}$ (Bi2212) by Behnia et al., using irradiated crystals. More recently, the magnitude of $\kappa_{res}/T$ in Bi2212 ($\approx 0.15$ mW/cm.K$^2$) was shown to be consistent with absolute values of $v_F/v_2$ estimated from angle-resolved photoemission (ARPES).

Despite this apparent consistency between theory and experiment, $\kappa_{res}/T$ has only been reported for two compounds, both at their optimum doping level, and it is not immediately obvious how $\kappa_{res}/T$ will vary across the phase diagram. ARPES and penetration depth measurements support claims that $v_F/v_2$, and thereby $\kappa_{res}/T$, increase in the underdoped regime. On the other hand, in certain underdoped cuprates, where $T_c$ has been suppressed in high magnetic fields, there is a marked tendency towards localization below $T_c$, suggesting a vanishing QP contribution at low $T$. Clearly, low-$T$ $\kappa(T)$ measurements on underdoped cuprates are important to help clarify this seemingly contradictory behavior.
With this in mind, we have carried out the first low-
T $\kappa(T)$ measurements on the underdoped cuprate Y124
($T_c = 80$K), which is a self-doped, stoichiometric cuprate
and therefore relatively free of disorder. Below 0.25K,
$\kappa(T) \approx T^3$ for both $a$- and $b$-axis currents, consistent
with a phonon heat conduction in the ballistic regime.
The residual linear $\rho$ term, however, is either absent
or is negligibly small, with an upper bound of 0.02
mW/cm.K$^2$. This result reveals that the universal con-
ductivity scenario breaks down dramatically in under-
doped Y124. One compelling possibility is that the $\rho$ states in Y124 are localized at low $T$, due to the proxim-
itry to the superconductor/insulator (S/I) boundary, and
therefore do not contribute to the low-$T$ heat transport.

The Y124 crystals were grown by a flux method de-
scribed elsewhere [11]. For this particular study, three
plate-like crystals were selected, two with their longest
dimension along the $b$-axis, the other along the $a$-axis.
Approximate dimensions were 0.25 x 0.16 x 0.015 mm$^3$
for the $\kappa_a$ crystal (labelled hereafter as $a$1) and 1 x 0.09
x 0.05 mm$^3$ and 0.8 x 0.25 x 0.06 mm$^3$ for the two $\kappa_b$
crystals, $b_1$ and $b_2$. $T_c = 80$K for all crystals, with a
transition width, measured resistively, of less than 1K.

$\kappa(T)$ for each crystal was measured between 0.14K
and 1K using a conventional steady-state four-probe tech-
nique that allowed the electrical resistivity $\rho_{a,b}(T)$ of each
sample to be measured in situ without changing the con-
tact configuration. Gold wires were attached as electrical
contacts using Dupont 6838 silver paint. The $\rho_{a,b}(T)$ be-
havior was found to be in excellent agreement with pre-
nvious measurements [3], with room temperature values,
$\rho_a = 350 \mu\Omega$cm and $\rho_b = 90 \mu\Omega$cm (for both $b$-axis crys-
tals). This large in-plane anisotropy arises from the high
conductivity of carriers on the quasi-1D CuO chains that
run parallel to the $b$-axis (see schematic inset to Figure
1) and confirms not only the high quality of the crystals
used in this study, but also that current flow in each case is
uniaxial. The temperature gradient was measured by
two RuO$_2$ thermometers connected to the "voltage" con-
tacts through the gold wires, and the thermometers were
supported by long, thin superconducting Nb-Ti wires to
minimize heat losses. Uncertainties in the absolute mag-
nitudes of $\kappa(T)$ ($\rho(T)$), mainly due to the finite contact
dimensions on these small crystals, are estimated to be
around 15% for $\kappa_b$ ($\rho_b$) and around 25% for $\kappa_a$ ($\rho_a$).

The $\kappa(T)$ data for all crystals are shown on double-
logarithmic axes in Figure 1. The variation of $\kappa_b(T)$
for the two $b$-axis crystals is almost identical over the
whole temperature range studied, giving us confidence in the
reproducibility of our data. Below 0.25K, $\kappa_a$ and $\kappa_b$
both vary approximately as $T^3$, consistent with phonon
heat transport in the boundary-scattering limit. Above
0.25K, $\kappa_a(T)$ deviates more strongly from a $T^3$ de-
pendence. The origin of the enhancement of $\kappa_b$ over $\kappa_a$
above $T = 0.25$K is not understood at present, though we
assume it reflects an additional contribution to $\kappa$ from the
CuO chains; either QP conductivity on the chains devel-
ops swiftly above 0.25K (note that the CuO chains, being
quasi-1D, may be susceptible to charge ordering at very
low $T$), or there exists an additional channel for phonon
heat propagation [13] along the chains that reduces the
effects of phonon scattering beyond the ballistic regime.
Further measurements in a magnetic field are envisaged
to clarify the origin of this anisotropy.

In order to look for evidence of a linear $\kappa_{res}$, we have
re-plotted the low $T$ data in Figure 2 as $\kappa_{a,b}/T$ versus $T^2$
and fitted each data set below 0.25K to the expression
$\kappa_{a,b} = AT + BT^3$ [14]. The coefficients for each fit are $A$
$= 0.011, -0.007$ and $0.006 (\pm 0.02) \text{mW/cm.K}^2$ and $B$
$= 6.50, 7.00$ and $10.67 (\pm 0.50) \text{mW/cm.K}^4$ for $a_1, b_1$
and $b_2$ respectively.

In the boundary-scattering limit, $\kappa_{ph}$ is given by

$$\kappa_{ph} = 1/3\beta < v_{ph} > l_0 T^3$$

where $\beta$ is the phonon specific heat coefficient, $< v_{ph} >$
the average acoustic sound velocity and $l_0 = 2w/\sqrt{\pi}$ is
the maximum phonon mean free path. Here, $w$ represents
a mean width of the rectangular-shaped crystal. Taking
the dimensions of our crystals and suitable values for $\beta$
($= 0.5 \pm 0.1 \text{mJ/mol.K}^2$ [24] and $< v_{ph} > (= 5 \pm 1$
x $10^7 \text{cm/s})$ [22], we obtain estimates for $\kappa_{ph}/T^3 = 4.1$
$\pm 1.2, 5.25 \pm 1.5$ and $9.55 \pm 2.0 \text{mW/cm.K}^4$ for $a_1, b_1$
and $b_2$ respectively [2]. Given the uncertainties in
measuring dimensions and contact distances, we believe
these values compare favourably with the experimental
values. More importantly, the size of the $T^3$ term for the
two $b$-axis crystals scales well with $w$ and we conclude
that the $T^3$ contribution is indeed simply the phonon
contribution in the ballistic regime.

The most striking result here is the complete absence
(to within our experimental accuracy) of the residual lin-
erar term in the low-$T$ $\kappa(T)$ for both chain and plane cur-
cent directions. It should be emphasized, of course, that
a zero linear term in $\kappa_b$ also implies a negligible $\kappa_{res}$
within the planes, meaning we have effectively confirmed
the absence of the universal QP term in Y124 in all three
samples. Moreover, for there to be any finite zero-$T$
intercept in $\kappa/T$, it would require $\kappa_{ph}(T)$ below $0.15$K
to vary as $T^{3+n}$ with $n > 0$, which is simply not physical,
given that the lattice heat capacity is strictly cubic below
1K. Thus, we are confident that the main result of this
Letter, namely the absence of $\kappa_{res}$ in Y124, is robust.
For comparison, we also show in the inset to Figure 2,
$\kappa_{ab}/T$ for optimally doped Bi2212 [1] measured with the
same experimental set-up [2]. In Bi2212, we can clearly
distinguish a finite $\kappa_{res}/T \approx 0.15 \text{mW/cm.K}^2$ (shown by
a dotted line), that is an order of magnitude larger
than the upper limits for $\kappa_{res}/T$ in Y124.

Despite the overwhelming case for d-wave pairing in high-$T_c$
cuprates, there is still limited, direct evidence for a $d_{x^2-y^2}$
order parameter in Y124. Thus, before discussing our result in terms of nodal QP states, we should
first examine the possibility that there is a finite gap everywhere on the Fermi surface in Y124. First of all, the orthorhombic distortion in Y124, induced by the chains, introduces some \( d + s \) admixture in the gap function. As the \( s \)-component is increased from zero, the position of the nodal lines are first shifted away from \( (\pi, \pi) \), but as the \( s \)-component becomes comparable with the \( d \)-component, a nodeless gap may form. Secondly, magnetic impurities are thought to induce a local imaginary component that could also give rise to a fully gapped state and a suppression of low \( T \) thermal transport. This latter possibility, however, is not supported by specific heat data taken on crystals from similar batches to those studied here, which show no sign of a low \( T \) Schottky anomaly arising from such magnetic impurities. In addition, power law penetration depths have now been observed down to 2K for both the \( a \) - and \( b \)-axes in Y124, suggesting a simple s nodal gap picture is equally applicable to Y124. Of course, we cannot rule out completely the possibility of a finite gap in Y124, but if it does exist, it would have to be vanishingly small. In what follows, therefore, we assume that nodal lines are present in Y124 and turn to consider what might be happening to the low-energy QP states in their vicinity.

From (1), we see that \( \kappa_{\text{res}}/T \) is directly proportional to the ratio \( v_F/v_2 \) at the nodal positions, so a negligible \( \kappa_{\text{res}}/T \) may indicate a sharp gap feature at the nodes at very low energies, induced either by doping, impurities or structural modifications. However, as noted above, \( v_F/v_2 \) is expected to increase as we move into the underdoped regime, and indeed, independent estimates of \( v_F/v_2 \) from the slope of the low-\( T \) penetration depth in Y124 yield an estimate for \( \kappa_{\text{res}}/T \) that is larger than those for both optimally doped Bi2212 and Y123. Moreover, given that band structure estimates for \( v_F/v_2 \) are similar for Y123 and Y124, even with our upper bound estimate for \( \kappa_{\text{res}}/T \) \( \approx 0.02 \text{ mW/cm.K}^2 \), we obtain a physically unrealistic value \( \left( v_F/v_2 \approx 2 \right) \) for the gap slope within the nodes in Y124. It appears unlikely, therefore, that the gap structure can itself explain a value of \( \kappa_{\text{res}}/T \) one order of magnitude lower than in Y123.

Another important consideration is the size of the impurity band, \( \gamma \). We recall that in the unitary limit, the universal conductivity regime develops below \( k_B T \leq \gamma \). Thus, the observation of \( \kappa_{\text{res}} \) depends not only on the temperature range of the experiment, but also on the energy scale of the impurity band, imposed by the scattering phase shift. However, taking values for the \( b \)-axis residual resistivity \( \rho_0 = 0.5 \mu\Omega \text{cm} \), and plasma frequency \( \omega_p = 2.5eV \), we obtain a lower bound estimate of \( \gamma \) in the unitary limit of 14K, some two orders higher than the base temperature of our measurements. (Unfortunately, similar analysis for \( J/a \) cannot be performed due to the difficulties in estimating \( \rho_0 \) from \( \rho_a(T) \)). Even in the opposite (Born) limit, where \( \gamma \) becomes exponentially small and therefore, may fall below our measurement range, the product of the DOS and the lifetime is also energy independent above \( k_B T \geq \gamma \). Hence, \( \kappa_{\text{res}}/T \) should still be constant and finite and the same arguments still apply.

Given these simple yet rather compelling arguments, we are left to consider how QP localization might account for the absence of \( \kappa_{\text{res}}/T \) in underdoped Y124. As mentioned above, high magnetic field measurements on the underdoped cuprates \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) and \( \text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4 \) revealed that \( \rho_{ab}(T) \), though "metallic" at high \( T \), tends towards localization as \( T \to 0K \). The origin of this localization phenomenon is unknown at present. However, if the field-induced destruction of superconductivity leads directly to an insulating phase, then it is not unreasonable to assume that this transition is from a superconductor with already localized QP excitations. The crossover from metallic to insulating behavior in the normal state (i.e. above \( H_c2 \)) as we approach the Mott insulator, suggests an increasing role of long-range interactions on the mobility of low-energy quasiparticles. The superfluid condensate, suppressed in the vicinity of an impurity, becomes ineffective in screening completely the Coulomb repulsion between quasiparticles in the bound state. As we approach the parent insulator, we expect such interactions to grow, leading to insulating behavior of the quasiparticles above the superconducting condensate and a negligible QP contribution to the low-\( T \) heat transport. Such localization in a nominally clean superconductor is an exciting prospect, and measurements on Zn-doped or irradiated Y124 are envisaged to investigate this possibility further. We note here that most impurity models for \( d \)-wave (cuprate) superconductivity fail to take into account the developing role of long-range interactions between quasiparticles as the S/I boundary is approached. We hope, therefore, that this work stimulates renewed theoretical efforts to understand the nature of QP excitations in the CuO2 planes, deep inside the superconducting state, and in particular in stoichiometric crystals on the underdoped side of the phase diagram.

In conclusion, we have measured the low-\( T \) \( \kappa(T) \) of stoichiometric, underdoped Y124 and have found that, in marked contrast to optimally doped Y123 and Bi2212, the "universal" QP conductivity term is absent. We have considered several interpretations of this intriguing result, including localization of the quasiparticles themselves, due to enhanced long-range interactions as we approach the S/I boundary. Prior to this work, the observation of the universal conductivity in Y123 and Bi2212 had been widely regarded as solid support for the picture of long-lived quasiparticles above the superconducting ground state of high-\( T_c \) cuprates. Our surprising result offers important and timely counter evidence that the residual conductivity term is non-universal, and we hope it encourages further debate and investigation into this critical, and still controversial, issue.
We acknowledge enlightening discussions with A.S. Alexandrov, J.F. Annett, A.V. Balatsky, D.M. Broun, J.R. Cooper, P.J. Hirschfeld, F.V. Kusmartsev, A.P. Mackenzie, A.J. Schofield, L. Taillefer, I. Vekhter and V.W. Wittorff. S.N. acknowledges support from the National Science Foundation under Grant No.INT-9901436. This work was also partly supported by CREST, a Grant in Aid for Scientific Research from the Ministry of Education, Science and Culture, Japan and New Energy and Industrial Technology Department Organization (NEDO).

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In previous measurements on Y123 and Bi2212, the asymptotic $T^3$ regime was observed only at the lower end of the temperature range of our experiments, i.e. below 150mK. The range for boundary-scattering, however, is determined by the sample dimensions, as well as the finite $T$ excitations which might act to scatter the phonons (see P.D. Thacher, Phys. Rev. 156, 975 (1967)). Y124 crystals are appreciably smaller in cross-section than the Y123 or Bi2212 crystals studied previously, and therefore, it is natural to expect the $T^3$ behaviour to set in at a correspondingly higher $T$. Moreover, the use of a higher $T$ fit on optimally-doped systems leads to an overestimation, rather than a reduction or vanishing of the linear term.

Figure Captures

Fig.1. $\kappa_a(T)$ and $\kappa_b(T)$ of Y124, plotted on double-logarithmic axes. The dashed line represents the $T^3$ dependence expected for phonon heat transport in the ballistic regime. The inset shows a simple schematic of the crystal structure of Y124.

Fig.2. $\kappa/T$ versus $T^2$ below 0.4K for a$\uparrow$1 (closed circles), b$\uparrow$1 (open circles) and b$\uparrow$2 (open squares). Fits to the expression $\kappa = AT + BT^3$ below 0.25K are indicated by dashed lines for b$\uparrow$1 and b$\uparrow$2 and by a solid line for a$\uparrow$1. The dotted line represents the universal conductivity limit for Bi2212. Inset: Comparison of $\kappa_a/T$ for Y124 (closed circles) and $\kappa_{ab}/T$ of Bi2212 [10] (open diamonds), measured in the same apparatus.

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Figure 1

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Figure 2

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