Anisotropic in-plane thermal conductivity of single-crystal YBa$_2$Cu$_4$O$_8$

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We report measurements of the in-plane thermal conductivity ($\kappa$) of YBa$_2$Cu$_4$O$_8$ (Y-124) single crystals in the temperature range $4K \leq T \leq 300K$ and compare them with previous results on YBa$_2$Cu$_3$O$_{6+x}$ (Y-123). For transport perpendicular to the CuO chains, $\kappa_a(300K) \approx 10W/mK$, and along the chains, $\kappa_b(300K) \approx 40W/mK$. The temperature dependence of $\kappa$ for both transport directions is much stronger than in YBa$_2$Cu$_3$O$_{6+x}$ (Y-123), indicative of substantially superior lattice heat conduction in Y-124, and resulting in maximum values for $\kappa$ at $T \sim 20K$ exceeding 200W/mK. The data imply a surprisingly large anisotropy in the lattice conduction. $\kappa_a$ and $\kappa_b$ are enhanced in the superconducting state as in other cuprates. The magnitude and anisotropy of the enhancement are discussed and compared to those of Y-123.

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The in-plane thermal conductivity ($\kappa_{ab}$) of cuprate superconductors has been studied extensively over the past several years. Unresolved issues remain concerning the normal-state temperature dependence and the superconducting-state enhancement. Because of the low carrier density, heat conduction by the lattice accounts for more than half of the measured normal-state $\kappa_{ab}$ in these materials. One would expect $\kappa_{ab}(T)$ in high-quality crystals to reflect a dominant lattice contribution similar to that of crystalline insulators, i.e. $\kappa_{ab} \sim 1/T$ at high-$T$, rising sharply to a maximum at low $T$. This behavior is observed in Nd$_{2−x}$Ce$_x$CuO$_4$ (Nd-214) but for all other cuprates investigated $\kappa_{ab}$ is very weakly $T$ dependent for $T > T_c$. Whereas for Bi$_2$Sr$_2$CaCu$_2$O$_8$ structural disorder (e.g., the Bi-O layer modulation) might explain its nearly glass-like lattice contribution, this would not seem applicable to YBa$_2$Cu$_3$O$_{6+x}$ (Y-123) and La$_{2−x}$Sr$_x$CuO$_4$ (La-214). Furthermore, $\kappa_{ab}(T)$ for underdoped, insulating Y-123 and La-214 is unconventional, suggesting that the weak $T$-dependence of $\kappa_{ab}$ is generic to the latter materials and not directly related to the presence of free charge. An unidentified phonon damping mechanism appears to underlie this behavior, possibly due to local lattice distortions and/or magnetic excitations. Regarding the enhancement of the thermal Hall conductivity experiments on optimally-doped Y-123 imply that the phenomenon is largely electronic in origin, but less is known about underdoped compounds. The magnitude of the enhancement correlates with the superconducting pair density in Y-123 throughout the underdoped regime, and it is of interest to explore the generality of this behavior through studies of other cuprates.

Here we report measurements of the in-plane thermal conductivity of YBa$_2$Cu$_4$O$_8$ (Y-124). This material is of particular interest with regard to the issues mentioned above because of its structural similarity to Y-123, absence of oxygen vacancies on the CuO chains, and the naturally underdoped state of the CuO$_2$ planes. We find that the magnitude of $\kappa_a$ (transverse to the chains) at $T=300K$ is comparable to that of Y-123, but $\kappa_b$ is 3-4 times larger. A very large in-plane anisotropy in the lattice conduction is implied. Both $\kappa_a(T)$ and $\kappa_b(T)$ behave as in conventional crystalline insulators (like Nd-214); their maximum values (at $T \approx 20K$) are the highest reported for any cuprate and exceed those of Y-123 by an order of magnitude. These results imply a strong damping of phonons by static or dynamic structural distortions as the source for the much weaker $T$ dependence of $\kappa_{ab}$ in Y-123. The superconducting-state enhancement for Y-124 is comparable in magnitude to that of $T_c=60K$ Y-123, consistent with the underdoped state of the planes in the double-chain compound. We discuss the anisotropy of the enhancement and its implications for the Lorenz number.

Thermal conductivity measurements were performed on three single crystals grown by a high-pressure flux method as described previously. Two of these (189 and 259) were grown in Al$_2$O$_3$ crucibles yielding a slight Al contamination, and $T_c = 72K$. Recent analyses indicate that Al substitutes for 1-2% of the Cu(2) atoms in the CuO$_2$ planes. As we discuss below, this light doping has a substantial effect on the thermal conductivity. The third crystal (315), grown in Y$_2$O$_3$ and without Al contamination, had $T_c = 80K$. Typical crystal dimensions were $0.8 \times 0.5 \times 0.05mm^3$, with the shortest dimension along the crystalline $c$ axis. The $a$- and $b$-axis electrical resistivities of similarly-prepared crystals have been discussed extensively elsewhere. Typical values at $T = 300K$ are $\rho_a = 400\mu\Omega cm$ and $\rho_b = 130\mu\Omega cm$. The steady-state thermal conductivity measurements employed a fine-wire differential chromel-constantan thermocouple and miniature chip resistor as heater, both glued to the specimen with varnish or epoxy. The absolute accuracy of the measurements is $\pm 20\%$ due to uncertainty in the placement of the thermocouple junctions. No corrections for heat
The total thermal conductivity is a sum of electronic and lattice components, \( \kappa = \kappa_e + \kappa_L \). First let us consider the electronic contributions, \( \kappa_{e,a} \) and \( \kappa_{e,b} \). The Wiedemann-Franz law, \( \kappa_e(T) = L_0 T/\rho L_0 = 2.44 \times 10^{-8}\Omega\text{mK}^{-1} \), and electrical resistivity \( \rho \) provide upper-bound estimates: \( \kappa_{e,a}(300\text{K}) \approx 2\text{W/mK} \) and \( \kappa_{e,b}(300\text{K}) \approx 6\text{W/mK} \). These imply \( \kappa_{L,a} \approx 8\text{W/mK} \) and \( \kappa_{L,b} \approx 25-39\text{W/mK} \), and the very large in-plane lattice anisotropy, \( \kappa_{L,L}/\kappa_{L,a} \approx 3-4 \). This is to be contrasted with Y-123 where similar analyses suggest that \( \kappa_L \) is nearly isotropic in the planes. Since Y-124 and Y-123 differ only in their CuO-chain structures, we must conclude that the strong lattice anisotropy in Y-124 is directly connected with the chain-related vibrational spectrum.

Anisotropy in \( \kappa_L \) can arise from either anisotropy of the phonon group velocities, their relaxation times, or both. Though optic modes can contribute to heat transport and the anisotropy, their contribution should diminish with decreasing temperature as should any associated anisotropy. This is contrary to our observation that \( \kappa_b/\kappa_a \) in the normal-state is nearly constant or increases with decreasing \( T \) (Fig. 2) [the decrease in \( \kappa_b/\kappa_a \) below \( T_c \) is related to superconductivity, as discussed below]. Experimental dispersion curves are not available for Y-124, however the computed spectrum is quite similar to that of Y-123, and does not imply a substantial in-plane anisotropy in the acoustic-mode group velocities. These observations would suggest a large anisotropy in the phonon relaxation rates in Y-124, but further information about the low-energy vibrational spectrum is clearly needed to address this issue.

The rather strong influence of the light Al doping in the planes on \( \kappa \) provides insight into the phonon scattering in both Y-124 and Y-123. Nuclear quadrupole...
resonance studies of similar Y-124 crystals reveal substantially larger $^{63}\text{Cu}$ linewidths in the Al-doped crystals, attributed to static disorder in the Cu(2)-apical oxygen bondlengths (a typical Al-O bondlength for octahedral coordination is 1.94Å as compared with 2.28Å for Cu-O). Such local structural distortions evidently represent a much more effective phonon scattering mechanism than does the mass defect introduced by the substitution. The local structural modifications may also explain why the hole concentration and $T_c$ are so dramatically suppressed – the 2% Al substitution for planar Cu reduces the mobile planar hole concentration by about 20%, i.e. a ten-fold larger reduction than would be expected if each Al atom filled one hole. This observation suggests that the suppression of mobile holes, by the combined effects of charge compensation, disorder-induced localization, and changes in the local electronic structure, extends to next nearest neighbors of each Al atom in the planes.

In Y-123 a similar static disorder in the apical bondlengths is induced by oxygen vacancies on the chain[23] and there is evidence for static or dynamical structural distortions of the CuO$_2$ planes in the normal state[4,24] possibly related to oxygen vacancies or diffusion that are absent in Y-124. The implication is that these features give rise to substantial damping of in-plane heat-carrying phonons, and are the origin of the weaker $T$-dependence and in-plane anisotropy observed for $\kappa$ in the single-chain compound.

It is of interest to compare the superconducting-state enhancement of $\kappa$ for Y-124 with that of Y-123[5]. We define the enhancement as $\Gamma \equiv -d(\kappa^s/\kappa^n)/dt|_{t=1}$, the reduced temperature derivative of the superconducting to normal-state thermal conductivity ratio near $T_c$. $\kappa^n$ is determined by a polynomial extrapolation of the normal-state data just below $T_c$, as shown in Fig. 3 (a). Figure 3 (b) shows the normalized data and corresponding slopes.

The $\Gamma$ values agree with those found for twinned Y-123 at similar $p$ values indicating that for the regime investigated here ($p \leq 0.11$), $\Gamma$ is largely determined by the change in scattering that occurs in the CuO$_2$ planes. That $\Gamma_s$ is larger for Al-free specimen 315 than for specimen 259 is consistent with the higher mobile hole concentration in the planes of the former. Also of interest is the anisotropy: $\Gamma_a/\Gamma_b \approx 1.5$ for specimen 259. Evidently this anisotropy is responsible for the decrease in $\kappa_a/\kappa_b$ at $T \lesssim T_c$ (Fig. 3). A similar behavior was observed for untwinned Y-123 ($x \sim 0.9$) [solid and dashed lines in Fig. 3], where the enhancement is also anisotropic with a slightly smaller $\Gamma_a/\Gamma_b \approx 1.2 - 1.5$.

Consider $\Gamma$ in more detail. We may write,

$$\Gamma_i = (\kappa_{e,i}/\kappa_i)\Gamma_{e,i} + (\kappa_{L,i}/\kappa_i)\Gamma_{L,i},$$

(1)

where $i = a, b$, and $\Gamma_{e,i}$ and $\Gamma_{L,i}$ are the $i$-axis electronic and lattice slope changes, respectively. The thermal conductivities are evaluated at $T_c$. Theoretically, $\Gamma_e$ is a sum of two terms: one proportional to $d\Delta^2/dt|_{t=1}$ and the other proportional to $-d(\tau^s_{qp}/\tau^s_{qp})/dt|_{t=1}$ ($\Delta$ and $\tau_{qp}$ are the real part of the superconducting gap and quasiparticle (qp) lifetime, respectively). In the cuprates, the second term predominates due to the strong suppression of qp scattering at $T < T_c$, and $\Gamma_e$ is positive. Scattering of phonons by charge carriers always produces a positive lattice term ($\Gamma_L$), but the thermal Hall conductivity measurements of Krishna et al[26] indicate that the electronic contribution to the enhancement predominates in optimally-doped Y-123. Their data for twinned Y-123 ($x \sim 0.9$) yields: $\Gamma_{ab} \simeq 1.4$, $\Gamma_{e,ab} \simeq 12.5$, and $\kappa_{e,ab}/\kappa_{ab} \simeq 9 \times 10^{-2}$. Thus, $\Gamma_{L,ab} \simeq 0.3$, and about 80% of the slope change for optimally-doped Y-123 is electronic in origin. Recent measurements[22] suggest that

![Fig. 3](https://example.com/fig3.png)
the slope change in underdoped Y-123 is also largely electronic in origin, and thus it is likely that this conclusion also applies to Y-124.

Given these observations, one might expect that the \( \Gamma \) anisotropy simply follows that of the superfluid, with the larger qp lifetime enhancement occurring for transport along the crystallographic direction characterized by the largest superfluid density. However, this is not the case; for Y-123 and Y-124 the superfluid density is 2-3 times larger along the chains \( [\text{La}] \) whereas \( \Gamma \) is larger along the planes. To get further insight into the origin of the \( \Gamma \) anisotropy, we examine the limiting case, \( \Gamma_{L,i} = 0 \), for which \( \Gamma_i \) is determined entirely by the electronic term. The electronic superconducting-state slope change may be related to the low-frequency electrical conductivity via the Wiedemann-Franz law, \( \kappa_{i\alpha} = L_i \sigma_{i\alpha} T \), where \( L_i \) and \( \sigma_{i\alpha} \) are the Lorenz number and real part of the electrical conductivity, respectively. Substituting into Eq. 1 we have

\[
\Gamma_i = -\frac{L_i \sigma_{i\alpha} T^2}{\kappa_i} \left[ \frac{1}{L_i} \left( \frac{dL_i^b}{dT} - \frac{dL_i^n}{dT} \right) + \frac{1}{\sigma_{i\alpha}} \left( \frac{d\alpha_i^b}{dT} - \frac{d\alpha_i^n}{dT} \right) \right]_{T = T_c}
\]

(2)

Theoretically, the first term has magnitude and sign that are highly model dependent. We first examine the second term. For untwinned, optimally-doped Y-123, the microwave data of Zhang et al.\(^{12}\) yield, \( (dL_i^b/dT)_{T_c} \approx -0.8 \times 10^{13} (\Omega \text{ m K})^{-1} \) and \( (d\alpha_i^b/dT)_{T_c} \approx -1.8 \times 10^{13} (\Omega \text{ m K})^{-1} \). For the normal-state data we employ \( \sigma/dT = -(1/\rho^2)d\rho/dT \) and an average of \( \sigma(T) \) measurements by the Swiss National Science Foundation. Work at the University of Miami was supported, in part, by NSF Grant No. DMR-9631236, and at the ETH by the Swiss National Science Foundation.

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account for the suppressed low-\(T\) value of \(\kappa_a\) in specimen 259 as compared with that of Al-free specimen 315.

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