

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

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We report measurements of the in-plane thermal conductivity (κ) of $\text{YBa}_2\text{Cu}_4\text{O}_8$ (Y-124) single crystals in the temperature range $4 \leq T \leq 300$ K and compare them with previous results on $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ (Y-123). For transport perpendicular to the CuO chains, $\kappa_a(300 \text{ K}) \approx 10$ W/mK, and along the chains, $\kappa_b(300 \text{ K}) \approx 40$ W/mK. The temperature dependence of κ for both transport directions is much stronger than in $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ (Y-123), indicative of substantially superior lattice heat conduction in Y-124, and resulting in maximum values for κ at $T \sim 20$ K exceeding 200 W/mK. The data imply a surprisingly large anisotropy in the lattice conduction. κ_a and κ_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. [S0163-1829(98)06445-5]

The in-plane thermal conductivity (κ_{ab}) of cuprate superconductors has been studied extensively over the past several years.¹ 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 κ_{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 $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ (Nd-214),² but for all other cuprates investigated κ_{ab} is very weakly T dependent for $T > T_c$. Whereas for $\text{Bi}_2\text{Sr}_2\text{CaCuO}_8$ structural disorder (e.g., the Bi-O layer modulation³) might explain its nearly glasslike lattice contribution,⁴ this would not seem applicable to $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ (Y-123) and $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (La-214). Furthermore, $\kappa_{ab}(T)$ for undoped, insulating Y-123 and La-214 is unconventional,⁵ suggesting that the weak T dependence of κ_{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 distortions⁵ and/or magnetic excitations.⁶ Regarding the enhancement,^{4,7} thermal Hall conductivity experiments⁸ 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^{9,10} 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 $\text{YBa}_2\text{Cu}_4\text{O}_8$ (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 κ_a (transverse to the chains) at $T = 300$ K is comparable to that of Y-123, but κ_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 $T \approx 20$ K) 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 κ_{ab} in Y-123. The superconducting-state enhancement for Y-124 is comparable in magnitude to that of $T_c = 60$ K 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.¹¹ Two of these (189 and 259) were grown in Al_2O_3 crucibles yielding a slight Al contamination, and $T_c = 72$ K. Recent analyses indicate¹² 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_2O_3 and without Al contamination, had $T_c = 80$ K. Typical crystal dimensions were $0.8 \times 0.5 \times 0.05$ mm³, 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;¹³ typical values at $T = 300$ K 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 losses (via radiation and conduction through the leads) have been applied; these are estimated to be $\sim 10\%$ near room temperature and $\leq 2\%$ at $T \leq 120$ K. For specimens 259 and 315 gold contacts were vapor deposited and their a -axis thermoelectric powers measured, yielding $S(290 \text{ K}) = 36$ $\mu\text{V/K}$ and 16 $\mu\text{V/K}$, respectively. These values provide estimates of the hole concentration per planar Cu atom,¹⁴ $p \approx 0.09$ and 0.11 for the Al-doped and undoped specimens, respectively.

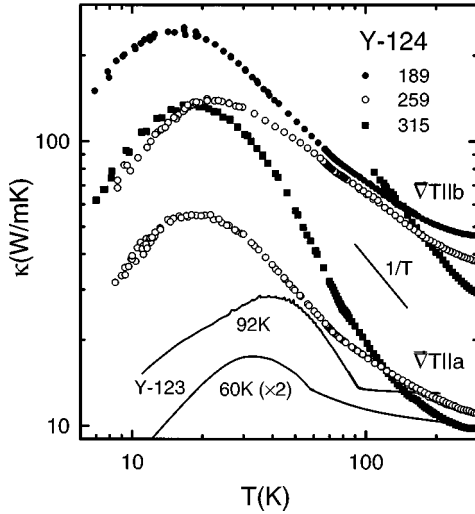


FIG. 1. $\kappa_a(T)$ and $\kappa_b(T)$ for three Y-124 crystals. Specimens 189 and 259 are lightly Al doped. Data for Y-123 crystals (solid lines) are from Ref. 16 ($T_c=92$ K) and Ref. 6 ($T_c=60$ K).

The results for κ_a and κ_b are shown in Fig. 1. The anisotropy in the normal state is substantial, with $\kappa_b/\kappa_a \sim 3-4$ (Fig. 2). The normal-state T dependence in both crystallographic directions is close to $1/T$ for specimen 315 and is significantly weaker ($\sim 1/T^{1/2}$) for the crystals containing Al. In all cases the T dependence is substantially stronger than that of both $T_c=92$ K and 60 K Y-123 (solid lines, Fig. 1). At low T the characteristic dielectric maxima are observed near 20 K. A change in slope is evident at T_c [Fig. 3 (a)] for all specimens, though not appearing as prominently as in Y-123 because of the substantially stronger normal-state T dependence. The low- T maximum values, $\kappa_b \approx 140$ and 245 W/mK for samples 189 and 259, respectively (and 500 W/mK for crystal 315 by extrapolation), are the largest reported for any cuprate, exceeding the 100 W/mK of insulating Nd-214,² and 25–40 W/mK observed in untwinned $T_c=92$ K Y-123.^{15–19} These observations indicate substantially superior and rather conventional lattice conduction in Y-124, confirming a similar conclusion based on previous measurements of polycrystals.²⁰

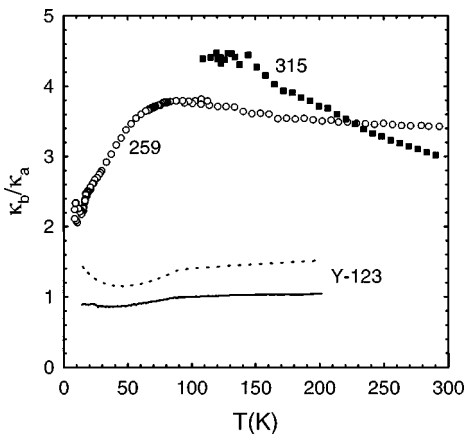


FIG. 2. Anisotropy κ_b/κ_a vs T for Y-124 crystals. Also shown are data for untwinned $T_c=92$ K Y-123 from Ref. 16 (solid curve) and Ref. 17 (dashed curve).

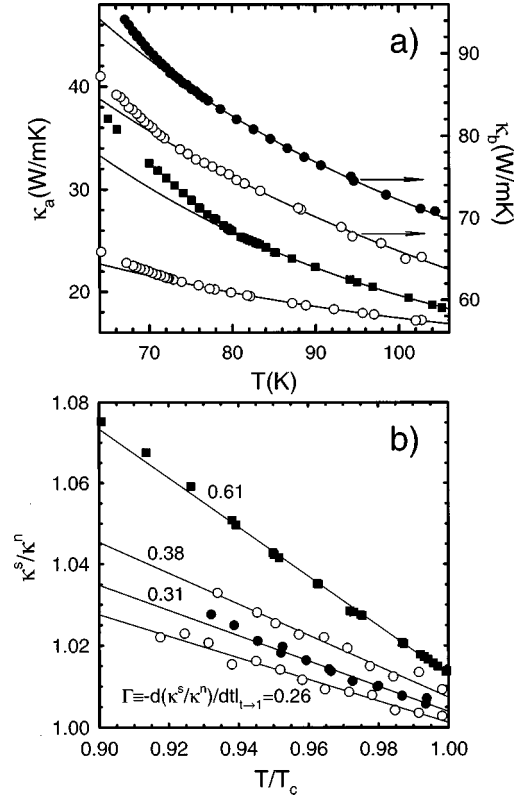


FIG. 3. (a) $\kappa_a(T)$ and $\kappa_b(T)$ showing the slope change near T_c on an expanded scale. Specimens are represented by the same symbols as in Fig. 1. Solid lines are polynomial fits to the normal-state data, defining κ^n . (b) Ratio of superconducting to normal-state thermal conductivity vs reduced temperature. Solid lines are linear least-squares fits to the data in the range $0.92 \leq t \leq 1$, defining $\Gamma \equiv -d(\kappa^s/\kappa^n)/dt|_{t \rightarrow 1}$. The data for specimen 315 (solid squares) are shifted vertically by 0.01 for clarity.

The total thermal conductivity is a sum of electronic and lattice components, $\kappa = \kappa_e + \kappa_L$. First let us consider the electronic contributions $\kappa_{a,e}$ and $\kappa_{b,e}$. The Wiedemann-Franz law, $\kappa_e(T) = L_0 T / \rho$ ($L_0 = 2.44 \times 10^{-8}$ W Ω / K^2), and electrical resistivities¹³ provide upper-bound estimates: $\kappa_{e,a}(300 \text{ K}) \approx 2$ W/mK and $\kappa_{e,b}(300 \text{ K}) \approx 6$ W/mK. These imply $\kappa_{L,a} \approx 8$ W/mK and $\kappa_{L,b} \approx 25-39$ W/mK, and the very large in-plane lattice anisotropy, $\kappa_{L,b}/\kappa_{L,a} \sim 3-4$. This is to be contrasted with Y-123 where similar analyses suggest that κ_L is nearly isotropic in the planes.^{16–19} 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 κ_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.²¹ This is contrary to our observation that κ_b/κ_a in the normal-state is nearly constant or increases with decreasing T (Fig. 2) (the decrease in κ_b/κ_a below T_c is related to superconductivity, as discussed below). Experimental dispersion curves are not available for Y-124; however, the computed spectrum²² 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 κ provides insight into the phonon scattering in both Y-124 and Y-123. Nuclear quadrupole resonance studies of similar Y-124 crystals¹² reveal substantially larger ⁶³Cu linewidths in the Al-doped crystals, attributed to static disorder in the Cu(2) apical oxygen bond lengths (a typical Al-O bond length for octahedral coordination is 1.94 Å as compared with 2.28 Å for Cu-O). Such local structural distortions evidently represent a much more effective phonon-scattering mechanism than does the mass defect^{23,24} 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 tenfold 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 bond lengths is induced by oxygen vacancies on the chains²⁵ and there is evidence for static or dynamical structural distortions of the CuO₂ planes in the normal state,²⁶ 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 κ in the single-chain compound.

It is of interest to compare the superconducting-state enhancement of κ for Y-124 with that of Y-123.^{9,10} We define the enhancement as¹⁰ $\Gamma \equiv -d(\kappa^s/\kappa^n)/dt|_{T \rightarrow T_c}$, the reduced temperature derivative of the superconducting to normal-state thermal conductivity ratio near T_c . κ^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 Γ values agree with those found for twinned Y-123 at similar p values,¹⁰ indicating that for the regime investigated here ($p \leq 0.11$), Γ is largely determined by the change in scattering that occurs in the CuO₂ planes. That Γ_a 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 κ_b/κ_a at $T \leq T_c$ (Fig. 2). A similar behavior was observed for untwinned Y-123 ($x \sim 0.9$) (solid and dashed lines in Fig. 2), where the enhancement is also anisotropic,¹⁶⁻¹⁹ with a slightly smaller $\Gamma_a/\Gamma_b \approx 1.2-1.5$.

Consider Γ in more detail. We may write

$$\Gamma_i = (\kappa_{e,i}/\kappa_i)\Gamma_{e,i} + (\kappa_{L,i}/\kappa_i)\Gamma_{L,i}, \quad (1)$$

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 . Theoretically²⁷ Γ_e is a sum of two terms: one proportional to $d\Delta^2/dt|_{t=1}$ and the other proportional to $-d(\tau_{qp}^s/\tau_{qp}^n)/dt|_{t=1}$ (Δ and τ_{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 scattering²⁸ at $T < T_c$, and Γ_e is positive. Scattering of phonons by charge carriers always produces a positive lattice term (Γ_L), but the thermal Hall conductivity measurements of Krishana *et al.*⁸ indicate that the electronic contribution to the enhancement predominates in optimally doped Y-123. Their data for twinned Y-123 ($x \sim 0.9$) yield $\Gamma_{ab} \approx 1.4$, $\Gamma_{e,ab} \approx 12.5$, and $\kappa_{e,ab}/\kappa_{ab} \approx 9 \times 10^{-2}$. Thus, $\Gamma_{L,ab} \approx 0.3$ and about 80% of the slope change for optimally doped Y-123 is electronic in origin. Recent measurements²⁹ suggest that 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 Γ 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,³⁰⁻³² whereas Γ is larger along the planes. To get further insight into the origin of the Γ anisotropy, we examine the limiting case $\Gamma_{L,i} = 0$ for which Γ_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 law³³ $\kappa_{e,i} = L_i \sigma_{1i} T$, where L_i and σ_{1i} are the Lorenz number and real part of the electrical conductivity, respectively. Substituting into Eq. (1) we have

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

Theoretically, the first term has magnitude and sign that are highly model dependent.³⁴ We first examine the second term. For untwinned, optimally doped Y-123, the microwave data of Zhang *et al.*³⁰ yield $(d\sigma_{1a}^s/dT)|_{T_c} \approx -0.8 \times 10^5$ ($\Omega \text{ m K}$)⁻¹ and $(d\sigma_{1b}^s/dT)|_{T_c} \approx -1.8 \times 10^5$ ($\Omega \text{ m K}$)⁻¹. For the normal-state data we employ $d\sigma/dT = -(1/\rho^2) d\rho/dT$ and an average of $\rho(T)$ measurements on high-quality untwinned crystals:³⁵ $\rho_a(T_c) \approx 75 \pm 25$ $\mu\Omega \text{ cm}$, $d\rho_a/dT|_{T_c} \approx 0.85 \pm 0.25$ $\mu\Omega \text{ cm/K}$, $\rho_b(T_c) \approx 38 \pm 12$ $\mu\Omega \text{ cm}$, $d\rho_b/dT|_{T_c} \approx 0.35 \pm 0.10$ $\mu\Omega \text{ cm/K}$. For the κ data we use¹⁷ $\kappa_a(T_c) = 11.5$ W/mK and $\kappa_b(T_c) = 16.0$ W/mK. With these values, the second terms in the expressions for Γ_a and Γ_b are $(1.2 \pm 0.2)L_a/L_0$ and $(2.0 \pm 0.2)L_b/L_0$, respectively. Experiments¹⁶⁻¹⁹ indicate $\Gamma_a = 1.5-1.6$ and $\Gamma_b = 1.0-1.3$. One possibility is that the first terms $(dL_i^s/dT - dL_i^n/dT)|_{T_c}$ are negligible, whence the Lorenz numbers at T_c are $L_a/L_0 \approx 1.3$ and $L_b/L_0 \approx 0.6$. Though $L^n > L_0$ has been predicted for strongly correlated systems,³⁶ these values for L_a and L_b are incompatible with the results of Ref. 8 that imply $L_{ab} \sim (0.3-0.5)L_0$ for twinned crystals. The latter estimate is consistent with the theoretical analysis of Hirschfeld and Putikka,⁷ which incorporates inelastic spin-fluctuation scattering and parameters

constrained to fit the normal-state NMR relaxation and electrical resistivity. We thus conclude that both L_a and L_b are less than L_0 and that the $(dL_i^s/dT - dL_i^n/dT)|_{T_c}$ are negative and of comparable magnitude to the second terms in Eq. (2). A considerable a/b anisotropy ($\sim 2-3$) in the L_i , $(dL_i^s/dT - dL_i^n/dT)|_{T_c}$, or both is then implied. Calculations

that incorporate inelastic as well as elastic scattering to yield the $L_i(T)$ near T_c would be useful to extract further information from the Γ_i .

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