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Anisotropy of the thermal conductivity of $YBa_2Cu_3O_{7-y}$

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The thermal conductivity, both in-plane (κ_{ab}) and out-of-plane (κ_c), has been measured in "90-K" crystals of $YBa_2Cu_3O_{7-y}$ from 10 to 330 K. κ_{ab} displays an 18 to 40% increase below the transition temperature T_c (as in ceramic data). κ_c, however, is four to five times smaller and shows no anomaly at T_c. Comparing these results to κ_{ab} of insulating crystals, we find evidence for very strong but highly anisotropic electron-phonon coupling in the normal state. Our results are consistent with a phonon origin for the peak in κ_{ab}.

A number of groups have reported measurements of the thermal conductivity κ in the high-temperature superconductors $YBa_2Cu_3O_{7-y}$ (1:2:3) (Refs. 1-5) and $La_{2-x}Sr_xCu$- O_{4-y}.[3,6,7] Studies on polycrystalline $YBa_2Cu_3O_{7-y}$ have shown a sharp increase of $\sim$25% in κ as T decreases below the transition temperature T_c, and then a T^2 behavior at low temperatures (T). Estimates[1-4] of the electronic contribution run as low as 10%. However, because of the large electronic anisotropy in 1:2:3, single crystals are necessary for reliable measurements. We have measured κ both in-plane and out-of-plane in single-crystal $YBa_2Cu_3O_{7-y}$ and tested its dependence on carrier concentration (by varying the oxygen content). We find that the electron-phonon scattering in the normal state is not only very strong but also highly anisotropic. We also find that the lattice conductivity κ^{ph} is more strongly anisotropic than would be expected simply from the crystal structure.

The superconducting crystals were grown by a flux method and slowly cooled in oxygen, giving $T_c \cong 90$-92 K and a transition width $\Delta T \cong 0.1$-0.5 K. To make the insulating samples, some crystals were annealed in Ar for 14 days at 600-670 °C to remove oxygen and carriers. Table I gives the dimensions of the samples. Thermal conductivity was measured by a low-frequency (0.015-0.025 Hz) pulsed-current technique using one heater and two thermometers. For the in-plane measurement, a miniature film resistor is varnished to one end of the crystal; the other end is epoxied to a copper heat sink. Two Chromel-Constantan thermocouples then measure the temperature gradient created across the sample by a heat pulse from the resistor. For the out-of-plane measurement, the crystal is sandwiched between two sapphire chips but with one corner exposed. The top sapphire holds a film resistor, the bottom sapphire is epoxied to a copper sink, and two thermocouples are epoxied to the exposed corner of the crystal. The temperature gradient produced across the sample is 0.2-0.8 K, and stray heat losses (radiation and conduction through leads) are found to be negligible.

TABLE I. Dimensions (a,b,c) and $YBa_2Cu_3O_{7-y}$ crystals studied.

Sample	T_c (K) nominal	Quantity measured	Dimensions $a\times b\times c$ (μm^3)
A	90	κ_{ab}	1000×550×50
B	90	κ_{ab}	1400×825×80
C	90	κ_{ab}	1340×430×65
D	90	κ_c	565×590×220
E	90	κ_c	720×620×320
F	90	κ_c	650×420×110
G	90	κ_c	650×720×220
H	<4	κ_{ab}	900×930×110
I	<4	κ_{ab}	700×600×50
J	<4	κ_{ab}	950×400×140
K	<4	κ_{ab}	950×700×120

Figure 1 shows the in-plane thermal conductivity κ_{ab} for three superconducting crystals of $YBa_2Cu_3O_{7-y}$. While the temperature dependence of κ_{ab} is very similar to that of polycrystalline 1:2:3,[1-4] its magnitude ($\approx$ 8-10 W/Km at 300 K) is twice as great. For 90 K $< T <$ 330 K, κ_{ab} is either very flat (samples *A*, *B*) or slowly increasing with temperature (*C*). Below T_c, κ_{ab} shows a sharp increase of 18 to 40% over its normal-state value, as in ceramic samples. The results for the out-of-plane thermal conductivity κ_c are shown in Fig. 2. κ_c is four to five times smaller than κ_{ab}. Although the experimental

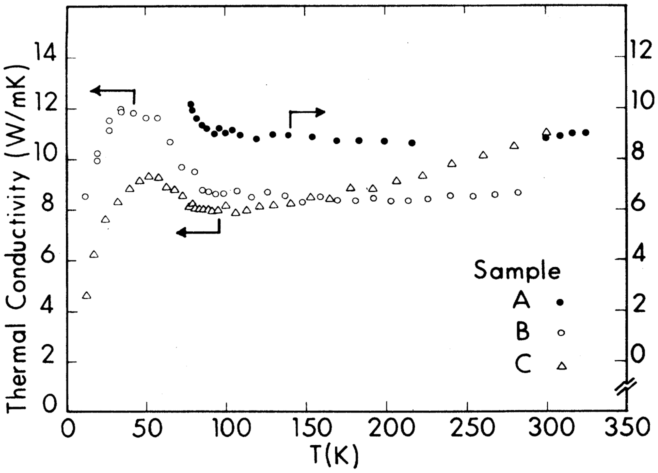

FIG. 1. The in-plane thermal conductivity κ_{ab} of 90-K crystals of $YBa_2Cu_3O_{7-y}$ vs temperature. The vertical scale for sample *C* has been shifted for clarity.

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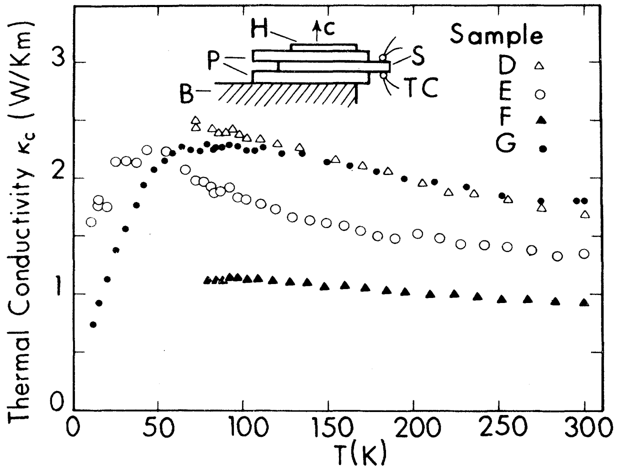

FIG. 2. The temperature dependence of the out-of-plane thermal conductivity κ_c in single crystals of $YBa_2Cu_3O_{7-y}$ with $T_c \approx 90$ K. The experimental configuration is shown in the inset. (H represents heater; P represents sapphire plates; B represents copper base; TC represents thermocouples; S represents crystal.)

geometry introduces some uncertainty into the absolute magnitude of κ_c, the T dependence is similar for all samples: κ_c increases gently as T decreases from high temperatures, exhibiting a broad maximum in the vicinity of 50–80 K and then decreasing sharply at low T. Significantly, in the vicinity of T_c, κ_c shows *no anomaly* associated with the superconducting transition.

The thermal conductivity may be written as[8]

$$\kappa_{ab} = \kappa_{ab}^{e} + \kappa_{ab}^{\mathrm{ph}} , \tag{1}$$

where κ_{ab}^{e} ($\kappa_{ab}^{\mathrm{ph}}$) is the electronic (phonon) contribution. A contribution from spin excitations is a distinct possibility in this oxide, but will not be discussed here. The lattice resistivity is the sum of terms due to scattering of phonons by electrons (W_{ab}^{e}) and by Umklapp processes (W_{ab}^{U}),

$$1/\kappa_{ab}^{\mathrm{ph}} = W_{ab}^{e} + W_{ab}^{U} . \tag{2}$$

An additional term W_{ab}^{D} (scattering by defects and domain boundaries) has been neglected in Eq. (2). Quantities for the **c** direction are defined in an analogous way with subscripts *ab* replaced by *c*. If we use the Wiedemann-Franz-Lorenz (WFL) law $[\kappa_{ab}^{e}/\sigma_{ab}T = 3 \times (k_B/e)^2/2]$ to estimate κ_{ab}^{e} from the measured in-plane electrical conductivity σ_{ab} $[\sim(150\ \mu\Omega\,\mathrm{cm})^{-1}$ at 290 K], we find that $\kappa_{ab}^{e} = 4.5$ W/Km, which is about 55% of the measured κ_{ab} above T_c. Thus, in contrast to the ceramic results, the electrons carry a substantial fraction of the normal-state *in-plane* heat current. By contrast, the WFL law estimates κ_c^{e} to be 0.02 W/Km above T_c, which is insignificant compared with κ_c (1–2 W/Km). However, more detailed analysis of the data casts serious doubts about the validity of applying Bloch-Boltzmann theory to the transport along **c** (see below).

An important question is the origin of the sharp increase in κ_{ab} at T_c and the pronounced peak near 50 K. The ceramic studies[1–4] suggested a sharp increase in the phonon mean free path below T_c as the cause. The size of the peak would require very strong electron-phonon scattering, at least in the plane. However, this conflicts with other studies[9] which argue that the electron-phonon coupling constant λ is very weak in the normal state. Ginzburg[10] has proposed that the peak may be caused by convection of the quasiparticles (QP) in the superconducting state. Comparing Figs. 1 and 2, we note that a phonon origin for the peak would require substantial anisotropy in κ^{ph} in the absence of electron-phonon scattering, since the phonon component has to account for most of the in-plane conductivity below T_c. (In sample B, κ_{ab} is ~ 12 W/Km at 50 K. Assuming that κ_{ab}^{e} decreases to approximately half its value at T_c in accordance with Geilikman's calculation[11] for the case of dominant inelastic scattering of the QP, we estimate that $\kappa_{ab}^{\mathrm{ph}}$ equals ~ 9 W/Km at 50 K. Thus, this interpretation requires $\kappa_{ab}^{\mathrm{ph}}$ to be four to five times κ_c in the limit when W_{ab}^{e} is negligible.)

To address this question, we have measured the in-plane thermal conductivity in insulating crystals (samples H–K). Here the absence of carriers should give $W_{ab}^{e} \approx 0$, so that the lattice resistivity at high temperatures should be limited only by W_{ab}^{U}. Since $\kappa_{ab}^{e} = 0$ as well, Eqs. (1) and (2) give $\kappa_{ab} \approx 1/W_{ab}^{U}$. At low temperatures, κ_{ab} is observed to rise sharply with T until $T \approx 100$ K, where it shows either a broad maximum or a gentle increase with further increase in T (Fig. 3). No trace of the peak in Fig. 1 remains in Fig. 3. The magnitude of κ_{ab} for $T > 100$ K is remarkable in the insulating samples. At 7–10 W/Km, it is fully as great as κ_{ab} in the "90-K" crystals, and four to five times larger than κ_c.[12] Thus, despite the loss of the substantial ($\sim 50\%$) electronic contribution, κ_{ab} in the insulating crystals is restored to close to the value in the 90-K crystals above 100 K. We conclude that $\kappa_{ab}^{\mathrm{ph}}$ roughly doubles in size when the charge carriers are removed, i.e., in the 90-K crystals, W_{ab}^{U} and W_{ab}^{e} are of the same order of magnitude [Eq. (2)]. This near equality is unusual in conventional metals, where W^{U} is ten times (or more) larger than W^{e} above 100 K.[8]

The results show that in the absence of carriers, $\kappa_{ab}^{\mathrm{ph}}$ is large (four to five times κ_c above 100 K).[12] When carriers are introduced, $\kappa_{ab}^{\mathrm{ph}}$ is strongly suppressed by a factor

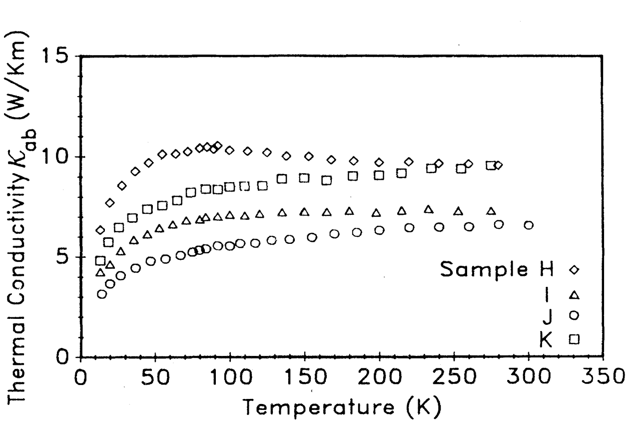

FIG. 3. The in-plane thermal conductivity κ_{ab} of insulating crystals of $YBa_2Cu_3O_{7-y}$. ac susceptibility measurements indicate that $T_c < 4$ K in these crystals.

of 2 for all $T > T_c$. The onset of superconductivity (with the concomitant reduction of W_{ab}^e) at T_c leads to a steady increase in $\kappa_{ab}^{\rm ph}$ with decreasing T until the phonon population is significantly reduced below 50 K. The overall magnitude of W_{ab}^U inferred from Fig. 3, together with the QP contribution at 50 K (which is still substantial), are thus sufficient to account for most of the observed peak in Fig. 1. The results here demonstrate very strong *in-plane* electron-phonon scattering in the normal state of the 90-K material, in disagreement with the conclusions of Jezowski *et al.*[13] and Gurvitch and Fiory.[9] The latter deduce a very weak $\lambda \leq 0.2$ from resistivity data up to 700 K. (The near equality of κ_{ab} above 100 K in the insulating and 90-K crystals also suggests the more radical viewpoint that κ_{ab}^e is negligible in *both* systems. However, we judge this to be unlikely.[14])

We turn next to the out-of-plane results. From Fig. 2, the absence of a peak or break in the slope at T_c imposes a bound on the magnitude of W_c^e relative to W_c^U. From the data we estimate that $W_c^e < 0.05 W_c^U$. As discussed above, the corresponding in-plane quantities, W_{ab}^e and W_{ab}^U, are approximately equal. Therefore, scattering by electrons affects less than 5% of the out-of-plane phonon flux, compared with 50% for the in-plane direction. This provides direct evidence for a very anisotropic electron-phonon coupling parameter.

It is also interesting to estimate the ratio of W_c^e with W_{ab}^e. From the preceding paragraph, $W_c^e/W_{ab}^e < 0.05\, W_c^U \times (2\kappa_{ab}^{\rm ph})$, using Eq. (2). However, since $\kappa_{ab}^{\rm ph} \approx \kappa_{ab}/2$ [applying the WFL estimate to Eq. (1)], and $W_c^U \approx \kappa_c$ in the 90-K system, we obtain the bound (near 100 K)

$$W_c^e/W_{ab}^e < 0.05(\kappa_{ab}/\kappa_c) \approx 0.2\,. \tag{3}$$

From this bound, we show next that Bloch-Boltzmann transport theory[8] leads to a serious contradiction if applied to the out-of-plane electronic transport. Since W^e is the limitation on the lattice conductivity imposed by electron scattering, it is directly related to the electrical resistivity ρ^L of the electrons due to scattering by phonons. Using the Bloch theory, Makinson[8,15] derives the relationship (for an isotropic metal),

$$W^e \approx (\rho^L/T)(ek_F S_F/12\pi^3)^2(3/C)^2\,, \tag{4}$$

where k_F (S_F) is the Fermi wave vector (surface area), and C the lattice specific heat, i.e., W^e scales with ρ^L. We argue that for an anisotropic solid with a simply connected Fermi surface, Eq. (4) can be applied to the two principal axes, so that the ratio W_c^e/W_{ab}^e is of the order of σ_{ab}/σ_c. In 90-K 1:2:3, the latter ratio is 200–300 near 100 K.[16] Comparing this "Bloch" estimate with the bound (<0.2) in Eq. (3), we find a disagreement of 3 orders of magnitude. The calculation leading to Eq. (4) may be improved by using the published band structure of 1:2:3. However, it is difficult to see how such a large discrepancy can be removed. The physical paradox is that despite the very poor electrical conductivity along **c** (due to the combined effect of short scattering times and large effective mass, within the Bloch theory), the lattice conduction along **c** is almost entirely unaffected by scattering from electrons, i.e., the electron-phonon coupling is very small, if not zero, along **c**. We believe that these results provide firm evidence that the very small σ_c is *not* due to strong phonon scattering, but arises from some other mechanism. Previously, Hagen *et al.*[16] argued that the observed difference in sign of $d\sigma_c/dT$ and $d\sigma_{ab}/dT$ below 150 K indicates qualitatively different transport mechanisms in the two directions. Recently, the same sign discrepancy was found for $T < 300$ K in 60-K 1:2:3, which also has a much larger anisotropy ($\sigma_{ab}/\sigma_c \approx 1000$).[17]

The extreme anisotropy of the electron-phonon coupling makes phonon-mediated superconductivity unlikely in this system, despite the large λ deduced for the in-plane transport. Previous attempts to derive a $T_c > 50$ K from phonon-mediated pairing had already encountered serious conceptual difficulties.[18] In light of the large anisotropy in λ found here, these difficulties are further compounded. Calculations based on the Eliashberg equations show that such an anisotropic electron-phonon coupling suppresses T_c compared with the isotropic case.[19] The very small shift in T_c observed when ^{16}O is replaced by ^{18}O is further evidence against a phonon-mediated mechanism for high-T_c superconductivity.[20]

In summary, we find that above T_c, the in-plane conductivity κ_{ab} is roughly 50% due to electrons and 50% due to phonons, while the out-of-plane conductivity κ_c is almost entirely due to phonons. By comparing κ_{ab} with κ_c, and between superconducting and insulating crystals, we deduce that the in-plane electron-phonon scattering is very strong, but weak, if not absent, along **c**. The large anisotropy in λ makes phonon-mediated superconductivity unlikely in this system. The large peak observed in κ_{ab} below T_c is attributed to an enhancement of the phonon component. No corresponding peak is observed in κ_c. Treating the anisotropic conductivity within the Bloch theory leads to serious conflict with these two observations. In insulating 1:2:3, we find that the lattice conductivity shows a large anisotropy of ~4–5.

Note added in proof. For recent calculations of κ^e in superconducting Pb and 1:2:3, we refer the reader to Refs. 21 and 22.

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