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Thermal conductivity in the *ab* plane of untwinned $YBa_2Cu_3O_{7-\delta}$

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We present measurements of thermal conductivity (κ) in the *ab* plane of untwinned $YBa_2Cu_3O_{7-\delta}$ ($\delta \approx 0.10$) for $T \le 200$ K. κ has been measured with heat flow along both the *a* and *b* axes and for two oxygen configurations of the same crystal. The conductivity anisotropy has its maximum value at the conductivity peak ($T \approx 40$ K), where we find $\kappa_a/\kappa_b \approx 1.15$ before and after an oxygen anneal. Very little *ab* anisotropy in the magnitude of κ is observed in the normal state, though near T_c the temperature dependences are noticeably different for the two directions: $d\kappa/dT > 0$ along the *b* axis and $d\kappa/dT < 0$ along the *a* axis. Near T_c ($T \le 110$ K) κ is enhanced slightly for both transport directions, features we attribute to the effect of superconducting fluctuations.

The normal-state transport properties of the cuprates have been the focus of considerable experimental and theoretical investigation for several years. In $YBa_2Cu_3O_{7-\delta}$ (YBCO) recent measurements of electrical resistivity[1] and thermopower[2,3] in untwinned crystals demonstrate that the in-plane electronic transport is highly anisotropic and sensitive to oxygen content due to the presence of the Cu-O chains. Little is known about the anisotropy of the lattice transport which should be manifested in thermal conduction.

In this paper we present measurements of in-plane thermal conductivity in untwinned YBCO. The *ab* anisotropy is found to be rather small, with a maximum $\kappa_a/\kappa_b \approx 1.15$ occurring near $T=40$ K. In the normal state we find qualitatively different temperature dependences for κ_a and κ_b. Precise measurements near T_c reveal a slight enhancement in κ_a and κ_b for the range $T_c \le T \le 110$ K. We attribute this feature to the effect of superconducting fluctuations on the electronic and lattice thermal conductivities.

The crystal used in this study had dimensions $1.2 \times 0.65 \times 0.04$ mm^3 and was grown in a gold crucible by a self-flux method[4] and detwinned by annealing under uniaxial stress.[5] Examination of the specimen by optical microscopy under crossed polarizers and by x-ray diffraction (XRD) before and after the oxygen anneal (100 h at 460 °C in flowing oxygen) confirm that the crystal is untwinned throughout $\ge 95\%$ of its volume, with residual twins occurring at the edges. The oxygen deficiencies have been estimated from measurements of the *c*-axis lattice parameter (from XRD), dc magnetization hysteresis loops,[6] and thermoelectric power[3] to be $\delta \approx 0.11$ and 0.08 before and after the anneal, respectively.

The thermal conductivity was measured by a steady-state method employing a differential Chromel-Constantan thermocouple and a small heater, glued to the specimen with varnish. A copper radiation shield was maintained at a temperature close to that of the specimen during measurement. The temperature gradient during measurement was typically 0.5–2.0 K/mm and linearity in the ΔT response was confirmed by varying the heater power. The absolute accuracy of the κ measurements is limited by the uncertainty in the specimen geometry and estimated to be $\pm 15\%$. We have previously estimated the combined heat losses via conduction through the leads and via radiation for a twinned YBCO crystal of comparable size by suspending the specimen from its measurement leads and measuring the heater power required to reproduce experimental conditions. We estimate the error in κ due to these heat losses to be $\le 2\%$ and nearly independent of temperature for $T < 200$ K. For $T > 200$ K the losses via radiation in our system increase rapidly with increasing temperature, and we thus limit our presentation to the data for $T < 200$ K. Four-probe, isothermal (dc) electrical resistivities (ρ) were measured in separate experiments, yielding $\rho_a(300\text{ K}) \approx 250$ $\mu\Omega$ cm and $\rho_b(300\text{ K}) \approx 175$ (140) $\mu\Omega$ cm before (after) the anneal, with ΔT_c (10%–90%) ≤ 0.3 K.

In Fig. 1 we plot κ_a and κ_b vs T for both oxygen

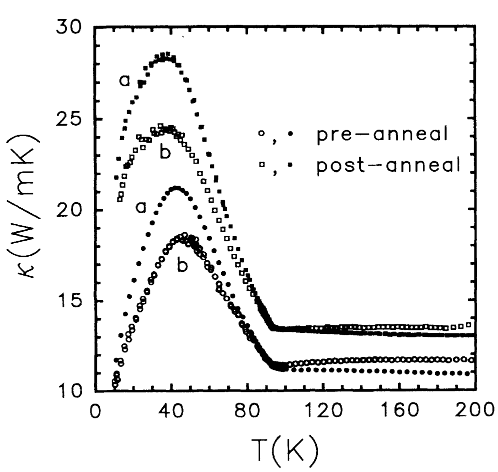

FIG. 1. Low-temperature thermal conductivity vs temperature for the untwinned YBCO crystal before and after oxygen anneal.

configurations, highlighting the low-temperature ($T < T_c$) data. The upturn in κ for temperatures below T_c is widely observed in the cuprates.[7] This feature is generally believed to reflect an enhancement of the lattice heat conduction, arising as a result of reduced phonon scattering by carriers as the latter condense into superconducting pairs.

In general, the total thermal conductivity can be viewed as a sum of lattice (L) and carrier (e) components. In order to assess the relative roles of κ_L and κ_e in determining the height of the conductivity maximum near $T = 40$ K, we make the following observations. We estimate κ_e using the measured electrical resistivities ρ and the Wiedemann-Franz law (WFL), which states that $\kappa_e \leq L_0 T/\rho$ ($L_0 = 2.45 \times 10^{-8}$ Ω W/K^2). This yields, at $T = 100$ K, an *upper-limit* estimate of the carrier component $\kappa_e/\kappa \approx 0.3$–$0.4$ for our specimen, in agreement with previous results for twinned crystals.[8] Inelastic scattering of the carriers (e.g., by phonons) tends to reduce the Lorenz number from its ideal value L_0, and hence κ_e could be substantially smaller than this estimate.

Theoretical studies of κ_e in the superconducting state of both weak- and strong-coupling superconductors[9] predict a peak in the quanitity $\kappa_e(T)/\kappa_e(T_c)$, with the height and position in temperature of this peak decreasing and increasing, respectively, as the elastic impurity scattering rate is increased. In the absence of defect scattering $\kappa_e(T)/\kappa_e(T_c)$ achieves its maximum value 2–2.5 at $T/T_c = 0.2$–0.3. Thus even if κ_e in YBCO is as large at T_c as $\kappa/3$ (the WFL upper limit), the phonon thermal conduction should still predominate near the maximum at $T \approx 40$ K. Furthermore, we expect the phonon-carrier and phonon-phonon scattering rates in YBCO to decrease rapidly with decreasing temperature for $T < 90$ K, and hence the value of κ at the maximum should largely reflect the degree of phonon-defect scattering. This conclusion is supported by recent lattice conductivity calculations fitting κ data for twinned YBCO.[10]

From the data in Fig. 1 we find $\Gamma_a \equiv \kappa_a(T_{\max})/\kappa_a(T_c) = 1.9$ (2.1) and $\Gamma_b = 1.6$ (1.8) before (after) the oxygen anneal. The temperature $T_{\max}$ at which the maximum occurs decreases from 42 K before the anneal to 38 K after the anneal for the a axis, and from 47 to 41 K for the b axis. These observations are consistent with a reduction in phonon-defect scattering after the oxygen anneal. Interestingly, the ratio $\Gamma_a/\Gamma_b \approx 1.19$, which represents a geometry-independent measure of the anisotropy, is relatively insensitive to the anneal. This suggests that the reduction in phonon-defect scattering is isotropic.

We now examine the data in the normal state (Fig. 2). There are two prominent features of the data which bear close examination: the different temperature dependences of κ for heat flow along the a and b axes, and the upturn evident in all of the data for $T \leq 110$ K.

We first note that, within the absolute accuracy of the measurements (15%), differences in the magnitudes of κ for all the data in Fig. 2 are not significant. The temperature dependences of κ for the two crystallographic directions are, however, clearly quite different. κ_a decreases with increasing temperature above T_c, whereas κ_b increases and tends to saturate or achieve a maximum near

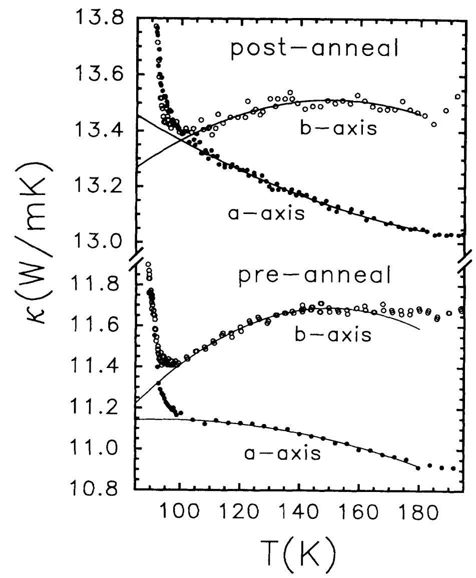

FIG. 2. High-temperature thermal conductivity vs temperature for the untwinned YBCO crystal before and after oxygen anneal. The solid lines represent second-order polynomial fits to the data for $108 \leq T \leq 170$ K.

150 K. After the anneal, κ_a exhibits a stronger and κ_b a weaker temperature dependence in the range $T_c \leq T \leq 150$ K. Note that the relative changes in κ with temperature are quite small (less than 4%) in all cases.

To gain insight into the difference in $\kappa_a(T)$ and $\kappa_b(T)$ and the changes induced by the anneal, we plot in the upper panel of Fig. 3 the differences $\Delta\kappa_1 \equiv \kappa$(post-anneal) $- \kappa$(preanneal), normalized to their values at 200

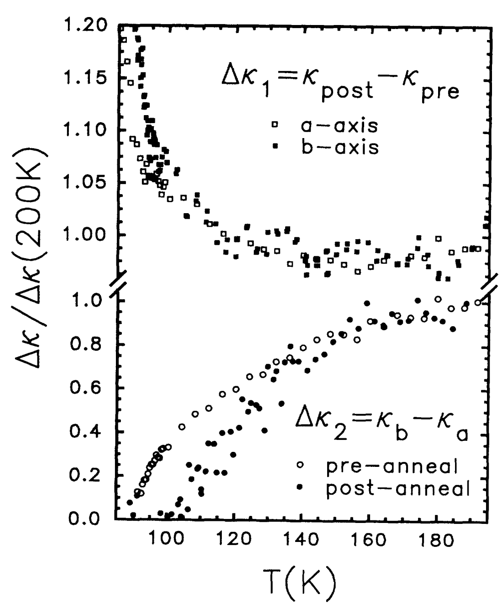

FIG. 3. Difference thermal conductivities vs temperature, calculated from the data in Fig. 3 and normalized to values at 200 K. The upper panel shows $\Delta\kappa_1 = \kappa$(postanneal) $- \kappa$(pre-anneal) for the a and b axis data. The lower panel shows $\Delta\kappa_2 = \kappa_b - \kappa_a$, before and after the oxygen anneal.

K. We note that ρ_a was essentially unchanged after the anneal whereas ρ_b decreased by $\approx 20\%$. Correspondingly, small changes are anticipated for κ_e and thus the quantities $\Delta\kappa_1$ predominantly reflect changes in κ_L. The apparent increase in the negative temperature coefficient of $\kappa(T)$ implies an increase in the relative weight of phonon-phonon scattering, as might occur if phonon-defect scattering were reduced after the anneal. Evidently the relative change in scattering induced by the anneal is reasonably isotropic, a conclusion which is consistent with our above observations regarding changes in the conductivity maximum.

Since YBCO is electronically anisotropic[1-3] we anticipate that the difference in the κ_a and κ_b temperature dependences may be associated, in part, with the anisotropy of the electronic thermal conduction. In the lower panel of Fig. 3 we plot, for both oxygen configurations, the differences $\Delta\kappa_2 = \kappa_b - \kappa_a$ also normalized to their 200-K values. If the lattice conductivity were isotropic, this difference would represent the difference in κ_e along the b and a axes. Though the temperature dependence of κ_L in YBCO is sensitive to the relative weights of phonon-defect, phonon-carrier, and phonon-phonon scattering,[10,11] the results for $\Delta\kappa_1$ indicate that this weighting is rather similar for the a and b axes. Thus it is unlikely that κ_L has a sufficiently different temperature dependence along the two directions to account for the qualitative difference in $\kappa_a(T)$ and $\kappa_b(T)$. The implication is that $\kappa_{e,b} - \kappa_{e,a}$ is an increasing function of temperature. The slightly stronger temperature dependence of $\Delta\kappa_2$ after the anneal suggests that this behavior is related to the Cu-O chain electronic heat conduction.

Returning to Fig. 2 we now examine the upturn in κ which is evident for all of the curves at $T < 110$ K. To quantify this enhancement in κ we fit the data for $108 \leq T \leq 170$ K to a second-order polynomial (solid lines in Fig. 2) thus defining κ^n. In Fig. 4 we plot the reduced fluctuation thermal conductivity $(\kappa - \kappa^n)/\kappa^n$ vs $(T - T_c)/T_c$. [We have also calculated κ^n with 104 and 112 K as cutoff temperatures and find no significant difference in $(\kappa - \kappa^n)/\kappa^n$.] Within the scatter of a factor of 2, $(\kappa - \kappa^n)/\kappa^n$ is isotropic and unchanged after the anneal.

The effect of superconducting fluctuations on electronic thermal conductivity has been studied theoretically.[12,13] The usual Aslamazov-Larkin[14] term, which describes the contribution of superfluid flow to the electrical conduction, does not contribute to the electronic thermal conduction because superconducting pairs carry no heat. In the dirty limit, Ref. 12 predicts an increase in κ_e due to enhancements in the quasiparticle density of states and lifetime. For two dimensions (2D) a weak logarithmic divergence is predicted near T_c, with no anomalous behavior expected in 3D.

Varlamov and Livanov[13] have recently calculated the fluctuation contribution to κ_e in a clean-limit model for layered superconductors such as the cuprates. The interlayer coupling is parameterized by an overlap integral ω which describes hopping between layers. In this model, $\kappa_e^{\rm fl}/\kappa_e^n \approx 0.3(\hbar/E_F\tau)[\epsilon(\epsilon+\delta_0^2)]^{-1/2}$, where E_F is the Fermi energy, τ is the scattering time, $\delta_0^2 \approx 0.11(\omega/T_c)^2$, and $\epsilon \equiv (T-T_c)/T_c$. This expression predicts a crossover from 2D behavior, $\kappa_e^{\rm fl}/\kappa_e^n \propto \epsilon^{-1}$, to 3D behavior, $\kappa_e^{\rm fl}/\kappa_e^n \propto \epsilon^{-1/2}$, for $\delta_0^2 \approx \epsilon$. The solid lines in Fig. 4 represent these 2D and 3D forms. The data imply $\delta_0^2 \approx 0.03$–0.04 and hence $\omega \approx 50$–60 K. This compares favorably with $\omega \approx 110$ K, estimated by Kresin, Wolf, and Deutscher[15] for plane-chain charge transfer.

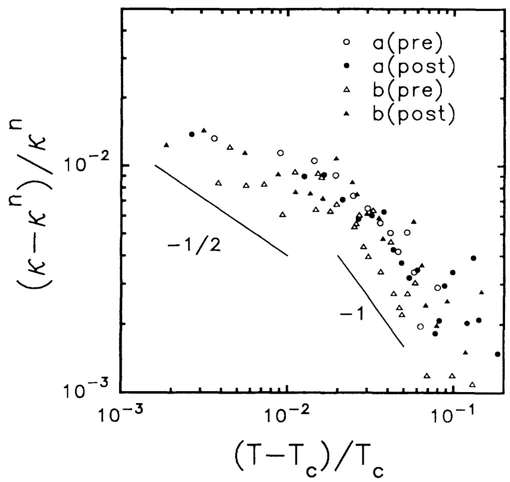

FIG. 4. Normalized fluctuation thermal conductivity, $(\kappa-\kappa^n)/\kappa^n$, vs $(T-T_c)/T_c$ (κ^n is defined by the solid lines in Fig. 2).

To check the magnitude of the effect against the theory we assume that all of the κ enhancement plotted in Fig. 4 is due to fluctuations in κ_e and use the WFL estimate, $\kappa_e/\kappa \leq 0.3$, to give $(\kappa-\kappa^n)/\kappa^n \leq 0.3\kappa_e^{\rm fl}/\kappa_e^n$. Using $(\kappa - \kappa^n)/\kappa^n \approx 0.0075$ at $\epsilon = 0.02$ then implies $E_F\tau \leq 2.4 \times 10^{-13}$ eV s. For $E_F = 0.3$ eV,[16] this yields the reasonable result $\tau \leq 8\times10^{-13}$ s. Evidently a fluctuation enhancement in κ_e alone can account for the effect we observe.

Given that the lattice conduction predominates at T_c in YBCO and that carriers are significant scatterers of phonons, we also anticipate that fluctuations will strongly influence κ_L as well. To our knowledge this effect has not been investigated theoretically or experimentally. The principal effect of fluctuations would be to reduce the effective number of carriers available to scatter phonons. A crude estimate of this effect could make use of the Bardeen-Rickayzen-Tewordt theory for lattice conduction[17,18] and employ a phonon-electron scattering rate proportional to $1-\langle|\Psi|^2\rangle$, where $\langle|\Psi|^2\rangle$ is the mean-square amplitude of the Ginzburg-Landau order parameter. This poses an interesting problem for future investigation.

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