Thermal Conductivity of an Untwinned $YBa_2Cu_3O_{7-\delta}$ Single Crystal and a New Interpretation of the Superconducting-State Thermal Transport

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We have measured the anisotropic thermal conductivity of an untwinned single crystal of $YBa_2Cu_3O_{7-\delta}$ in the most conducting **a**-**b** plane from 14 to 200 K. Unlike previous analyses, ours attributes the observed rapid rise in thermal conductivity in the superconducting state to the electronic contribution of the Cu-O plane. We propose that strong suppression of the quasiparticle scattering rate with decreasing temperature is responsible for the large enhancement of the superconducting-state thermal conductivity.

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Thermal conductivity has been used widely to study high-T_c superconductors [1], offering the advantage of probing transport properties in both normal and superconducting states.

In this Letter, we report a measurement of the thermal conductivity κ_a and κ_b of an untwinned $YBa_2Cu_3O_{7-\delta}$ (YBCO) single crystal in the most conducting **a**-**b** plane. We find that the temperature dependences of κ_a and κ_b are nearly identical in the normal state, except for a constant offset. We argue that the large enhancement of both κ_a and κ_b observed in the superconducting state is due to the electronic contribution of the Cu-O plane, which increases rapidly below T_c due to the strongly suppressed quasiparticle scattering rate in the superconducting state. This picture is much more consistent with the body of experimental data on YBCO than a scenario that ignores the electronic contribution in favor of a purely phononic mechanism.

The untwinned $YBa_2Cu_3O_{7-\delta}$ single crystals were grown by a flux method [2]. This technique consistently yields untwinned single crystals of high quality, characterized by ultralow impurity levels and low in-plane normal-state resistivity [3]. The untwinned crystal used for this study is of dimensions $0.6\times0.6\times0.025$ mm^3, with **a** and **b** axes parallel to the edges. Low-field susceptibility measurements show a superconducting transition temperature $T_c\sim90.5$ K, with transition width of ~1 K. The measurements of thermal conductivity were done by steady-state methods [4]. The main source of uncertainty ($<10\%$) is that involved in determining the separation of the differential thermocouple junctions which measure the temperature gradient.

In Fig. 1, we show the temperature dependence of κ_a and κ_b from 200 to 14 K. One can see that, from 200 to 90 K, both κ_a and κ_b increase monotonically with decreasing temperature. Below $T_c=90$ K, both κ_a and κ_b increase much more rapidly, peak at ~40 K, and then decrease rapidly at lower temperatures. We consider first the normal-state data κ_a^n, with the thermal gradient along **a**. The total thermal conductivity of a metal usually can be expressed as the sum of phonon and electron contributions, $\kappa_a^n=\kappa_{e,a}^n+\kappa_p$, with e and p denoting electron and phonon contributions, respectively; n denotes the normal state. From the Wiedemann-Franz law, we expect $\kappa_{e,a}^n=L_0\sigma_aT$, with $L_0=2.45\times10^{-8}$ WΩ/K^2 the Lorentz number. Since the electrical conductivity of $YBa_2Cu_3O_{7-\delta}$ is well known to follow $\sigma_{a(b)}\sim1/T$ [5], $\kappa_{e,a}^n$ should be temperature independent.

The phonon thermal conductivity can be expressed by the kinetic equation $\kappa_p=\frac{1}{3}cv^2\tau$, with c the specific heat, v the sound velocity, and τ^{-1} the total scattering rate. $\tau^{-1}=\sum_i\tau_i^{-1}$, with i denoting different scattering pro-

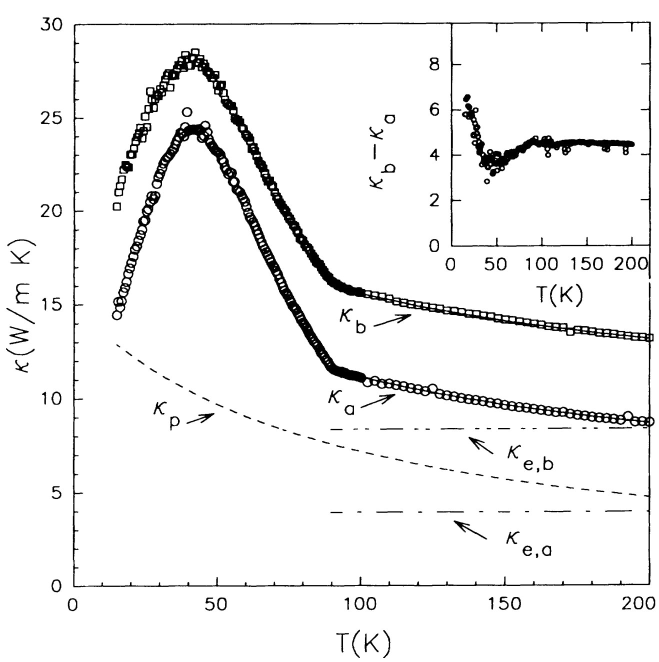

FIG. 1. Temperature dependence of thermal conductivity in the **a** (○) and the **b** (□) direction. Solid lines, Eqs. (1) and (2) fitted to κ_a and κ_b; they completely overlap with experimental data. Dashed line, the derived phonon thermal conductivity κ_p. Dot-dashed line and dot-dot-dashed line, the derived electronic thermal conductivity in the **a** and **b** directions, κ_a and κ_b, respectively. Inset: $\kappa_b-\kappa_a$ vs T.

cesses. The most prominent scattering mechanism for the phonon thermal conductivity at high temperatures is the umklapp process for which $\tau_U^{-1} \propto T$ [6]. Other scattering contributions, such as electron-phonon, phonon-defect, and phonon-boundary scatterings, which are weakly temperature dependent, can be approximated by a temperature-independent thermal resistivity W_0. The total thermal conductivity along **a** is then

$$\kappa_a^n = \kappa_{e,a}^n + 1/(W_0 + \alpha T) \tag{1}$$

with W_0 and α constants.

A least-squares fit of Eq. (1) to the κ_a^n data yielded $\kappa_{e,a}^n = 3.94 \pm 0.25$ W/m K, $W_0 = 0.0668 \pm 0.0014$ m K/W, and $\alpha = (7.25 \pm 0.61) \times 10^{-4}$ m/W. The resulting fit completely overlaps with the experimental data above T_c, as shown in Fig. 1.

Although the thermal conductivity is anisotropic, the temperature dependences of the normal-state κ_a and κ_b are nearly identical. Their difference is plotted in the inset of Fig. 1, from which one can see that $\kappa_b - \kappa_a$ is temperature independent above 90 K. We attribute this difference to the electronic contribution from the Cu-O chain $\kappa_{e,\text{chain}}^n$, which enters only the b-axis thermal conductivity, so that

$$\kappa_b^n = \kappa_{e,a}^n + \kappa_{e,\text{chain}}^n + 1/(W_0 + \alpha T) \,, \tag{2}$$

with $\kappa_{e,\text{chain}}^n = 4.45$ W/m K, and with W_0 and α the same as in Eq. (1). We plot Eq. (2) in Fig. 1, and the agreement with the experimental κ_b data is excellent.

To see whether the derived $\kappa_{e,a}^n$ and $\kappa_{e,b}^n$ are reasonable, we estimate the corresponding electrical resistivity from the Wiedemann-Franz law. Our result $\kappa_{e,a}^n \approx 3.94$ W/m K corresponds to a resistivity $\rho_a \approx 124$ $\mu\Omega$ cm and $\kappa_{e,b} = \kappa_{e,a} + \kappa_{e,\text{chain}} \approx 8.39$ W/m K to $\rho_b \approx 57.5$ $\mu\Omega$ cm at 200 K. These results are consistent with $\rho_a \approx 100$–147 $\mu\Omega$ cm and $\rho_b \approx 50$–61 $\mu\Omega$ cm obtained by a direct measurement on several untwinned single crystals of similar quality [3].

Now let us turn to the superconducting state. We can see that $\kappa_a(40\text{ K})/\kappa_a(90\text{ K}) \sim 2$ and $\kappa_b(40\text{ K})/\kappa_b(90\text{ K}) \sim 1.8$. This in-plane thermal conductivity enhancement in the superconducting state is a general property of high-T_c superconductors [1]. The common explanation to date [1,7-9] is the following: The normal-state thermal conductivity is assumed to be dominated by the lattice thermal conductivity, which is significantly limited by electron-phonon scattering. Below T_c, the number of electrons available to scatter phonons drops dramatically due to the superconducting condensation. The phonon mean free path therefore increases rapidly below T_c, thus giving rise to a large enhancement of the total thermal conductivity. While this scenario is able to produce a thermal conductivity enhancement below T_c, there are several weak points. First, it predicts a sizable enhancement of the thermal conductivity κ_c in the **c** direction below T_c [7], but various experiments consistently reported no observable κ_c anomaly below T_c [8,10]. Second, the behavior of the electronic thermal conductivity in the superconducting state has been either neglected or assumed to decrease rapidly with temperature as in the Bardeen-Rickayzen-Tewordt treatment [11], in which the quasiparticle scattering rate was assumed unaffected by superconductivity. These assumptions are not well justified in light of recent observations of a strongly suppressed quasiparticle scattering rate [12]. Third, the anomalous transport properties of the normal state are difficult to reconcile with predominant electron-phonon scattering [5]. Lastly, in order to account for the large enhancement, the phonon thermal conductivity in the absence of the electron-phonon scattering has to be at least as large as the peak value (~ 29 W/m K for this experiment). However, Hagen *et al.* showed that the thermal conductivities of several oxygen-deficient, nonsuperconducting $YBa_2Cu_3O_x$ single crystals are consistently smaller than 10 W/m K [8].

In the two-fluid picture, the superfluid does not carry any heat. The temperature dependence of the electronic thermal conductivity is dictated by the number density and the relaxation time of the normal-fluid carriers. From the Wiedemann-Franz law, the electronic thermal conductivity κ_e should scale with the normal-fluid conductivity σ_q as $\kappa_e \propto \sigma_q T$. Experiments on ac conductivity of the high-T_c superconductors showed that the quasiparticle scattering rate is much suppressed below T_c [12,13]. This rapid suppression indicates that the excitations that strongly damp the current-carrying quasiparticles freeze out rapidly below T_c, giving rise to a much enhanced electrical conductivity and hence electronic thermal conductivity. This scenario provides an alternative way to explain the observed temperature dependence of the thermal conductivity.

To derive the electronic thermal conductivity κ_e^s from the total thermal conductivity data, we assume that the temperature dependence of the phonon thermal conductivity κ_p is not affected by the superconducting transition. Therefore,

$$\kappa_{e,a}^s = \kappa_a^s - 1/(W_0 + \alpha T) \tag{3}$$

with W_0 and α the same as in Eq. (1). The derived $\kappa_{e,a}$ is shown in Fig. 2. We have to point out that at low temperature, the phonon thermal conductivity may deviate from the extrapolation, and hence affect the derived $\kappa_{e,a}^s$. We have compared the result of this analysis with the surface resistance data of Bonn *et al.* [13], using the Wiedemann-Franz law. The results agree quantitatively to within 20% [14].

To calculate the electronic thermal conductivity in the superconducting state rigorously, we adopt the formalism derived by Kadanoff and Martin and by Tewordt [15], in which

$$\kappa_e^s = \frac{1}{2k_B T^2 m} \int d^3p \frac{p_z^2 \epsilon_p^2}{\Gamma} \operatorname{sech}^2\left(\frac{E_p}{2k_B T}\right) \approx \frac{\varphi}{\Gamma} \tag{4}$$

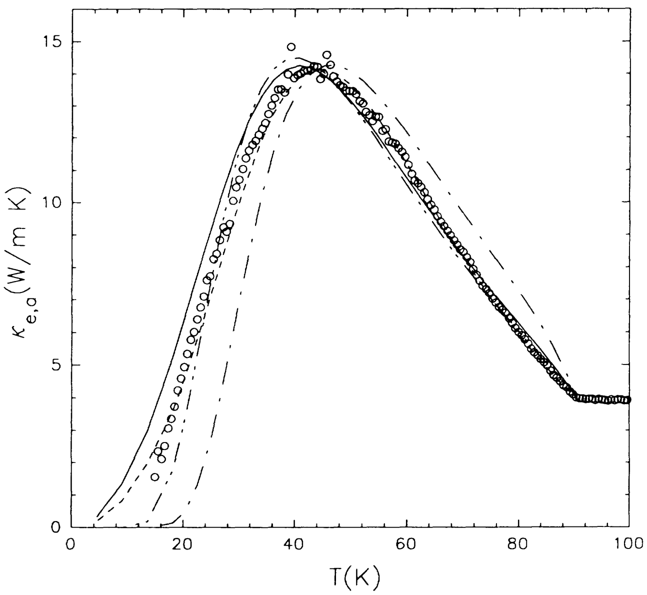

FIG. 2. Temperature dependence of the $\kappa_{e,a}$ derived from $\kappa_a-\kappa_p$ (○) and calculated fits. Dashed line, d wave, $g=1.75$, $w_i=0.09$; solid line, d wave, $g=3$, $w_i=0.03$; dot-dot-dashed line, s wave, $g=1.75$, $w_i=0.079$; dot-dashed line, s wave, $g=3$, $w_i=0.001$.

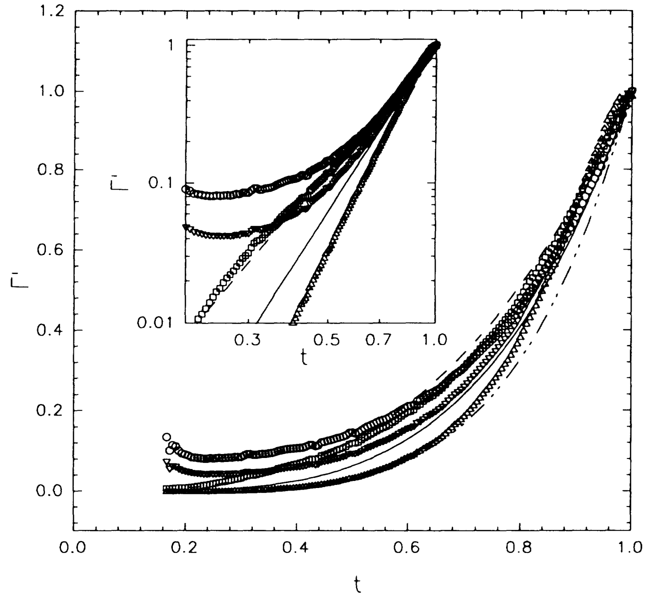

FIG. 3. Reduced scattering rate Γ' vs reduced temperature for different pairing states. ○, d-wave, $g=1.75$; ▽, d-wave, $g=3$; □, s-wave, $g=1.75$; △, s-wave, $g=3$. Dashed line, $y=t^3$; solid line, $y=t^4$; dot-dot-dashed line, $y=t^5$. Inset: plots on logarithmic scale.

with $E_p=(\epsilon_p^2+\Delta_p^2)^{1/2}$, p_z the momentum in the direction of the thermal gradient, ϵ_p the normal-state dispersion, and Δ_p the superconducting energy gap. Γ is the quasiparticle scattering rate. For the temperature dependence of the gap, we use $\Delta(T)/\Delta(0)=\tanh(\gamma\sqrt{1/t-1})$, with $\Delta(0)$ the energy gap at zero temperature, and $t=T/T_c$ the reduced temperature. For s-wave pairing, Δ is k independent; for d-wave pairing, we use $\Delta(k,T)=\Delta(T)[\cos(k_x)-\cos(k_y)]$ [16]. The gap-to-T_c ratio g is defined as $g=\max[\Delta(k,T=0)]/2k_BT_c$. The second equality in Eq. (4) holds when Γ is assumed to be energy independent.

In Fig. 3, we plot the reduced scattering rate

$$\Gamma'=\frac{\Gamma(T)}{\Gamma(T_c)}=\frac{\varphi(T)\kappa_{e,a}(T_c)}{\varphi(T_c)\kappa_{e,a}(T)},$$

where $\kappa_{e,a}$ is from experimental data derived from Eq. (3), and φ is calculated for s-wave and d-wave pairing states with two different values of the gap ratio $g=1.75$ and $g=3$, which correspond to weak and strong coupling, respectively. The calculations are not sensitive to γ in the range of 1.7–2.2, and $\gamma=2.2$ was used. Figure 3 suggests that the scattering rate roughly follows a power law $\Gamma'\approx t^n$, with $t=T/T_c$ the reduced temperature. For the d-wave pairing state, we have $n\sim4$, whereas for s-wave pairing, $3<n<5$ depending on the gap ratio g. It is evident that the derived scattering rates tend to nonzero values at low temperature, especially for the d-wave case. This is likely the effect of the residual scattering rate due to impurities. A more realistic scattering rate would be

$$\Gamma'\sim t^n+w_i\,, \tag{5}$$

with w_i a constant.

To fit the κ_e^s data, we calculate the thermal conductivity using Eqs. (4) and (5), and varying n and w_i to get the best fit. We find for d-wave pairing that $\Gamma'=t^4+w_i$ produces a good fit, independent of the gap ratio g. For s-wave pairing, a reasonable fit for $g=1.75$ can be obtained with $\Gamma'=t^{3.2}+w_i$. For s-wave pairing with $g=3$, $\Gamma'=t^5+w_i$ is required to fit the data over the full temperature range, but deviations are significant. These calculations are presented in Fig. 2.

A power-law scattering rate in the d-wave pairing state is expected when it is due to electronic interactions. In fact, a calculation based on the nearly antiferromagnetic-Fermi-liquid theory [17] shows that the quasiparticle scattering rate due to spin fluctuations in a d-wave pairing state follows $\Gamma\propto T^4$ [18]. A power-law scattering rate in a s-wave state is difficult to understand, because the uniform nonzero gap would give rise to an exponential decay of both the quasiparticle excitations and the scattering rate. Further study is required to determine uniquely whether the pairing state is d wave or s wave.

As noted earlier, the large enhancement below T_c has been observed in $\kappa_{a,b}$ in many high-T_c superconductors, but is absent in κ_c [8,10]. As the electrical resistivity ρ_c is several orders of magnitude larger than ρ_a and ρ_b [5], the electronic thermal conductivity $\kappa_{e,c}$ that follows from the Wiedemann-Franz law is negligibly small. Therefore, the lattice thermal conductivity $\kappa_{p,c}$ should dominate. The absence of an anomaly in κ_c is consistent therefore with our assertion that electron-phonon scattering does not dominate the temperature dependence of the total thermal conductivity.

We now turn to the superconducting-state thermal conductivity of the chain electrons. From Fig. 1, we see that the chain's thermal conductivity decreases slightly below T_c, then increases with temperature below 40 K. This behavior is in sharp contrast with the derived electronic thermal conductivity of the Cu-O plane. This different characteristic indicates that the quasiparticle properties on the Cu-O chain are very different from those of the Cu-O plane. NMR experiments reveal that the nuclear spin relaxation rate of Cu(1) decreases with temperature at a much slower rate than that of the Cu(2) sites [19]. This implies that excitations on the Cu-O chains are not as strongly reduced below T_c as those on the plane that affect the Cu(2) nuclear spin relaxation. The oxygen defects on the chain may further limit the quasiparticle relaxation time. Therefore, we expect the quasiparticle lifetime on the chain to remain short below T_c.

In the above analysis of thermal conductivity data, we neglected any change of phonon thermal conductivity due to the superconducting transition. In reality, the electron-phonon coupling is finite, and there will be a change in phonon thermal conductivity due to the decrease in electrons available to scatter phonons. However, electron-phonon scattering may not be significant in comparison with phonon-phonon or phonon-defect scattering. Hence electron-phonon scattering may not play an important role in the superconducting-state thermal transport in $YBa_2Cu_3O_{7-\delta}$. In our calculations, we assumed that the scattering rate Γ is energy independent, which may not be a good approximation. A more complete model should take energy dependence into account. In the comparison of s-wave and d-wave pairing, we should caution that this analysis depends on the accuracy of the phonon background subtraction. The determination of $\kappa^s_{e,\mathrm{chain}}$ involves the difference of two large and strongly temperature-dependent quantities, and hence the uncertainty may be significant. Note also that the observed large enhancement of κ below T_c in YBCO is very different from the behavior of some heavy-fermion superconductors [20,21].

In summary, we have measured the anisotropic thermal conductivity of an untwinned single crystal of $YBa_2Cu_3O_{7-\delta}$ in the most conducting **a-b** plane from 14 to 200 K. Unlike previous analyses, we attribute the observed rapid rise in thermal conductivity in the superconducting state to the electronic contribution from the Cu-O plane. We propose that the strong suppression of the quasiparticle scattering rate with decreasing temperature is responsible for the large enhancement of the superconducting-state thermal conductivity.

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