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Evidence for strong electron-phonon coupling in the thermal conductivity of $YBa_2Cu_3O_{7-\delta}$

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We have measured the *ab*-plane thermal conductivity (κ) on single-crystal $YBa_2Cu_3O_{7-\delta}$ ($\delta \approx 0.08$) in the temperature range $10 \leq T \leq 300$ K. Our main focus is an analysis of the slope change in $\kappa(T)$, which occurs just below the superconducting transition. The reduced-temperature ($t = T/T_c$) derivative of the normalized thermal conductivity, $-d(\kappa^s/\kappa^n)/dt|_{t=1}$, as determined from the data, is rather small, ≤ 1.1. From measurements of the electrical conductivity on the same specimens and application of the Wiedemann-Franz law, we determine the relative contributions to the heat conduction from the carriers and the lattice, and discuss the normal-state phonon-scattering mechanisms. Employing these results we calculate the slope of the lattice thermal conductivity at T_c and infer that the slope of the carrier conductivity must by very large, ≥ 6. This result implies strong coupling for some of the carriers.

The thermal conductivity κ in the high-T_c superconductors[1] is unique in providing information on the transport mechanisms occurring in both the normal and superconducting states. The dominance of the phonon heat transport over that of the carriers and the importance of phonon-carrier scattering in the cuprates are manifested as a sharp upturn in the *ab*-plane κ for $T < T_c$. An analysis of this feature offers the potential of revealing information about the superconducting energy gap Δ and the strength of the electron-phonon coupling, two fundamental parameters that are currently the focus of much experimental and theoretical research.

In this paper we analyze the slope change in the *ab*-plane thermal conductivity of two $YBa_2Cu_3O_{7-\delta}$ ($\delta \approx 0.08$) single crystals. Measurements of both the thermal and electrical conductivities on the same specimens enable us to estimate the carrier and lattice contributions to the total κ. The importance of phonon-phonon, phonon-carrier, and phonon-defect scattering are demonstrated. Employing these results we calculate the slope change in the lattice conductivity at the superconducting transition. We infer that the carrier thermal conductivity decreases sharply below T_c, consistent with strong coupling for some of the carriers.

The $YBa_2Cu_3O_{7-\delta}$ crystals were grown in zirconia crucibles by a self-decanting flux method described elsewhere,[2] and were subsequently annealed in oxygen at 450–500 °C. Both specimens were high-quality, mm-sized (80-μm-thick) single crystals with $\rho(295\ \text{K}) \approx 250$ $\mu\Omega$ cm, transition widths $\Delta T_c \leq 0.5$ K, and $R = 0$ at 92 K.

The oxygen deficiency of specimen 1 was estimated as $\delta \approx 0.08$ from measurements of thermoelectric power, performed simultaneously with the thermal conductivity and reported previously.[3] This value is consistent with the value inferred from the anomalous features in magnetization hysteresis loops, which have recently been shown[4] to correlate with the *c*-axis lattice parameter (a measure of oxygen content). The magnetization of specimen 2 implies an oxygen content similar to that of sample 1.

The thermal conductivity was measured using a steady-state technique, employing a differential chromel-constantan thermocouple and small resistive heater glued to the specimen with varnish. The temperature difference during measurement, ΔT, was typically 0.3–1.0 K. Linearity in the ΔT response was confirmed throughout the temperature range by varying the heater power. Errors in κ due to heat losses are estimated to be less than 2% except for $T > 200$ K where radiation losses become more significant. At room temperature we estimate an error in κ of $\approx 10\%$.

Figure 1 shows $\kappa(T)$ for both specimens. Near room temperature $\kappa \sim 8$–9 W/mK, which agrees with previous measurements on single-crystal $YBa_2Cu_3O_{7-\delta}$.[5] The most notable feature in these data is the sharp upturn and peak that occur for $T < T_c$. This behavior is widely ob-

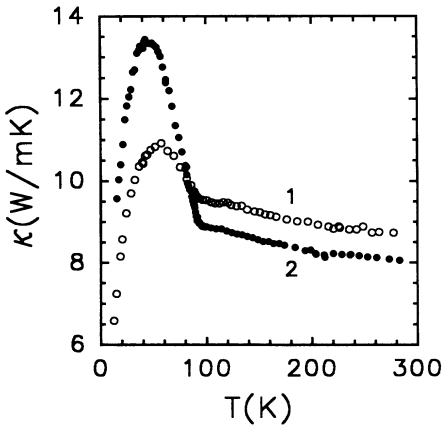

FIG. 1. Thermal conductivity vs temperature for two $YBa_2Cu_3O_{7-\delta}$ single crystals.

served in the cuprates,[1] and arises from an increase in the phonon mean free path due to the reduced scattering of phonons by carriers as the latter condense into superconducting pairs. An analysis of this slope change is our principal focus and is discussed in detail below. We first address the behavior of κ for $T > T_c$.

The contribution of the carriers to heat conduction may be calculated using the Wiedemann-Franz law (WFL), which states that $\kappa_e/\sigma T = L_0$, where κ_e is the electronic component of the thermal conductivity, σ is the electrical conductivity, and $L_0 = 2.45\times10^{-8}$ W Ω/K^2 is the Lorenz number. This yields an upper limit estimate of κ_e and we find at 100 K, $\kappa_e/\kappa \approx 0.31$ for both specimens.

In Fig. 2 we plot the lattice conductivity $\kappa_L = \kappa - \kappa_e$, where κ_e is determined, as above, from the measured electrical resistivity and the WFL. The slight upturn in κ_L at $T \leq 100$ K for specimen 2 is probably associated with superconducting fluctuations, an issue we address in more detail elsewhere.[6] In general, we anticipate three principal sources of scattering for phonons in a metal: defects, free carriers, and other phonons. Phonon-carrier and phonon-defect scattering yield a temperature independent or weakly decreasing κ_L with decreasing temperature at high temperatures.[7] The observed increase in κ_L with decreasing T is characteristic of phonon-phonon scattering. The overall change in κ_L from 300 K down to T_c is, for all specimens, relatively small ($\leq 20\%$) in comparison to that expected for a defect-free insulator (greater than or equal to a factor of 3). Thus with regard to lattice conduction, $YBa_2Cu_3O_{7-\delta}$ appears to be in the complicated regime where all three scattering mechanisms are significant. These observations are consistent with a more quantitative analysis discussed below.

We now address the change in heat transport at the superconducting transition and consider the ratio of the thermal conductivities in the superconducting (s) and normal (n) states, κ^s/κ^n, a quantity which is most readily compared with theory. We may write the temperature derivative of this ratio as

$$\frac{d}{dt}(\kappa^s/\kappa^n) = \eta\frac{d\alpha}{dt} + (1-\eta)\frac{d\beta}{dt} + \frac{d\ln\kappa^n}{dt}, \qquad (1)$$

where $t = T/T_c$ is the reduced temperature, $\alpha \equiv \kappa_e^s/\kappa_e^n$, $\beta \equiv \kappa_L^s/\kappa_L^n$, and $\eta = \kappa_e^n/\kappa^n$ is the relative contribution of carriers to the total heat conduction. The normal-state thermal conductivity κ^n is determined, for $T < T_c$, from the experimental data by smooth extrapolation of the normal-state data to several degrees below T_c.[8] In Fig. 3 we plot κ^s/κ^n vs t near $t = 1$. The slopes are $d(\kappa^s/\kappa^n)/dt|_{t=1} = -0.4$ (sample 1) and -1.1 (sample 2) as indicated by the solid lines. The third term on the right-hand side of (1) is determined directly from the data at $T \geq T_c$. This term is quite small and thus the slope is determined by a competition between the first two terms which have opposite sign.

The BCS theory of thermal conductivity for weak-coupling superconductors[9-11] predicts that, in the limiting cases of predominant electron-phonon and electron-defect scattering, respectively, α is a universal function of Δ/k_BT and for $\Delta = \Delta^{BCS}$, $d\alpha/dt|_{t=1} = 1.6$ (electron-phonon) and 0 (electron-defect). For strong coupling and when electron-defect scattering[12,13] predominates, $d\alpha/dt|_{t=1} = 0$ as in weak coupling. The case of strong coupling and dominant electron-phonon scattering is, however, much more complicated and depends on the form of the electron-phonon spectral function, $\alpha^2(\omega)F(\omega)$.[14] For materials such as lead and mercury where these parameters are known from tunneling measurements, the very rapid decrease in κ_e observed experimentally is in reasonable accord with theory. The difference from weak coupling arises both from the more rapid opening of the gap for strong coupling as well as the stronger frequency dependence of the quasiparticle relaxation rate.[14,15] The value of $d\alpha/dt|_{t=1} = 9$ for lead[16] is the largest known for any material. A slope $d\alpha/dt|_{t=1}$ substantially greater than 1.6 may thus be considered evidence for strong electron-phonon coupling.

The slope of the lattice conductivity for weak coupling

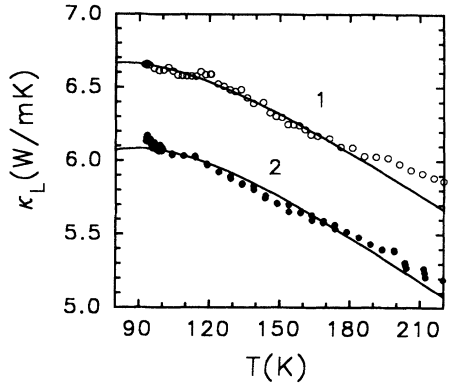

FIG. 2. Lattice conductivity (κ_L) vs temperature. κ_L was calculated by subtracting, from the data in Fig. 1, κ_e as determined from the measured electrical resistivity and the WFL. The solid lines are calculated from Eq. (2) with $\Delta = 0$ and parameters for sample 1 (2): $A_{ph\text{-}d} = 1.73\times10^3$ (2.80×10^3) s^{-1}K^{-4}, $A_{ph\text{-}e} = 6.0\times10^7$ (1.4×10^7) s^{-1}K^{-1}, $A_{ph\text{-}ph} = 40$ (19) s^{-1}K^{-4} (see text for details).

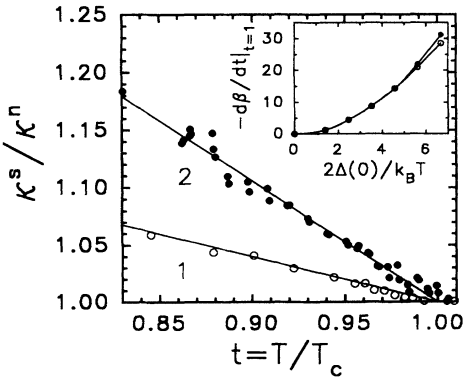

FIG. 3. The normalized thermal conductivity near T_c plotted vs reduced temperature. Inset: Values of the reduced-temperature derivative of the normalized lattice thermal conductivity $d\beta/dt|_{t=1}$ plotted vs gap size, as calculated from Eq. (2) using the same parameters as for the normal-state curves in Fig. 2.

and predominant phonon-electron scattering can be calculated from the Bardeen-Rickayzen-Tewordt (BRT) theory.[10] Tewordt and Wölkhausen have modified this theory to include the effects of other scatterers[17] and crystalline anisotropy[18] on the lattice conductivity. In addition to the dependence on Δ/k_BT, the slope $d\beta/dt|_{t=1}$ depends on the model phonon-scattering rate, in the case of $YBa_2Cu_3O_{7-\delta}$ a rate which must, as discussed above, incorporate the combined effects of scattering by carriers, phonons, and defects. For strong coupling, the lattice conductivity very close to T_c can be calculated from the weak-coupling theory, provided the BCS gap is scaled to the appropriate strong-coupling value.[18] Thus the temperature dependence of β in this regime is governed by $\Delta(T)$ which should follow the mean-field expression $\Delta(T)\approx a(1-t)^{1/2}$, independent of the coupling strength [a is a scaling constant $\approx 1.74\Delta(0)$ for the BCS gap].

It is important to note that the change in the lattice thermal conductivity at T_c generally reflects the coupling of carriers to longitudinal-acoustic phonons (presumed responsible for most of the lattice heat conduction) and these phonons may not be the most important for superconductivity. Recent calculations by Peacor *et al.*[19] suggest that the coupling for these modes in $YBa_2Cu_3O_{7-\delta}$ is rather weak. It is the value of $d\alpha/dt|_{t=1}$ which holds the most direct information regarding the strength of the carrier-phonon interaction relevant to superconductivity.

We now calculate the lattice conductivity slope from the BRT theory[10,17,18] and place limits on the slope of the electronic component for $YBa_2Cu_3O_{7-\delta}$ using our experimental results and Eq. (1). The lattice conductivity is written as[20]

$$\kappa_L=\frac{k_B}{2\pi^2 v}\left(\frac{k_BT}{\hbar}\right)^3\int_0^{\Theta/T}dx\frac{x^4e^x}{(e^x-1)^2}\times\int_0^1 d\zeta\,\tfrac{3}{2}(1-\zeta^2)\tau(x,\zeta,T)\,, \tag{2}$$

where $v=5000$ m/s is the sound velocity,[21] $\Theta=400$ K is the Debye temperature,[22] $x=\hbar\omega/k_BT$ is the reduced phonon frequency, and $\tau(x,\zeta,T)$ is the phonon relaxation time. The scattering of phonons by crystal boundaries, point defects, carriers, and other phonons is represented by

$$\tau^{-1}(x,T)=v/d+A_{\text{ph-}d}(xT)^4+A_{\text{ph-}e}xTg(x,T)(1-\zeta^2)^{3/2}+A_{\text{ph-ph}}x^2T^4\,,$$

where d is the crystallite thickness. Here, $g(x,T)$ is the ratio of the phonon-carrier scattering rates in the superconducting and normal states as defined by BRT, equal to unity for $T>T_c$ and a universal function of Δ/k_BT for $T<T_c$. The integration over the variable ζ describes the anisotropy of the electron-phonon interaction; it is assumed that only the in-plane component of phonon momentum is emitted or absorbed by the carriers.[18] Following Ref. 17 we take $\Delta=\chi\Delta^{\text{BCS}}$, where χ is a constant. The coefficients $A_{\text{ph-}d}$, $A_{\text{ph-}e}$, $A_{\text{ph-ph}}$ are determined self-consistently by numerically integrating Eq. (2) to fit the normal-state κ_L data and the data for $T\leq 20$ K where $\kappa\approx\kappa_L$. The normal-state curves for the two specimens are shown as solid lines in Fig. 2. The slope $d\beta/dt|_{t=1}$ is then determined from the calculated $\beta(t)$ curves. In the inset of Fig. 3 we plot $-d\beta/dt|_{t=1}$ versus the gap size $2\Delta(0)/k_BT_c$ for the two parameter sets. A number of experiments suggest the existence of two gaps in $YBa_2Cu_3O_{7-\delta}$, associated with the CuO_2 planes and Cu-O chains, with $2\Delta_{\text{pl}}(0)/k_BT_c\approx 5$–$6$ and $2\Delta_{\text{ch}}(0)/k_BT_c\approx 2.5$–$3.5$. The two-band expression[13] for κ_L is more complicated than Eq. (2), but we may place a conservative lower limit on $d\beta/dt|_{t=1}$ by using the smallest gap size, $2\Delta(0)/k_BT_c=2.5$. This implies $d\beta/dt|_{t=1}\approx -4.5$ which, from Eq. (1), yields $d\alpha/dt|_{t=1}\approx 6$. A more reasonable picture[23] that assigns strong coupling to the planes and weak coupling to the chains would imply a plane contribution, $d\alpha^{\text{pl}}/dt|_{t=1}\approx 20$. Our main conclusion, $d\alpha/dt|_{t=1}\geq 6$, indicates that strong coupling characterizes some of the carriers in $YBa_2Cu_3O_{7-\delta}$.

Since our analysis relies on the calculated value of $d\beta/dt|_{t=1}$, and specifically the choice of scattering parameters, we have examined the influence of the latter carefully. The coefficients employed for sample 1 (2) imply that at $T=100$ K defect and carrier scattering account, respectively, for approximately 75% (60%) and 18% (30%) of the lattice thermal resistivity. Hence $d\beta/dt|_{t=1}$ is determined principally by the relative weights of the carrier and defect scattering terms. We find that $d\beta/dt|_{t=1}$ varies by less than 20% when these terms are varied within acceptable ranges. We also find similar results when we allow for a substantial deviation from the WFL, as would be expected for $T<\Theta$ in a pure (electronically speaking) material[7] or a material with very strong electron-phonon coupling. Thus we believe that our overall conclusion regarding $d\alpha/dt|_{t=1}$ will survive a more detailed and specific model.

As mentioned above, in the context of the existing theory values of $d\alpha/dt|_{t=1}$ larger than 1.6 imply strong electron-phonon coupling *and* predominant electron-phonon scattering. Electron-phonon scattering predominates at T_c only in the purest of conventional materials and one may question whether it is feasible for this condition to hold in $YBa_2Cu_3O_{7-\delta}$. The most obvious reason why it may be reasonable is the higher transition temperature of $YBa_2Cu_3O_{7-\delta}$; the relative weight of phonons in the total scattering of carriers is substantially enhanced at higher temperatures. Second, the strength of the electron-phonon coupling may be substantially larger in $YBa_2Cu_3O_{7-\delta}$ than in any conventional material.

The slope $d\alpha/dt|_{t=1}$ for strong coupling and predominant electron-phonon scattering[14] is directly proportional to the product of the derivative $d(\Delta/k_B)^2/dt|_{t=1}$ and a frequency average of the quasiparticle relaxation rate $\Gamma(\omega)$. Employing the mean-field expression for the gap at T_c, a gap size of $2\Delta(0)/k_BT_c\approx 6$ would enhance $d(\Delta/k_B)^2/dt|_{t=1}$ by roughly a factor $(6/4.1)^2\approx 2$ over that of lead, for example. It is generally true that the more strongly $\Gamma(\omega)$ decreases with frequency, the larger is the slope $d\alpha/dt|_{t=1}$. A strong frequency dependence can arise from sharply peaked structure in the coupling function, $\alpha^2(\omega)F(\omega)$. In this regard we note that there is

substantial evidence from Raman scattering[24] that some carriers couple strongly to optical phonons, especially those modes yielding peaks in the phonon density of states $F(\omega)$ near energies of 15 and 45 meV. Recent tunneling results[25] on $Bi_2Sr_2CaCu_2O$ reveal such structure in the derived coupling spectrum (with $\lambda \approx 3.5$), and similar results for $YBa_2Cu_3O_{7-\delta}$ are anticipated.[26] Thus it is possible that the contribution to $d\alpha/dt|_{t=1}$ from $\Gamma(\omega)$ is also large. As more information about $\alpha^2(\omega)F(\omega)$ becomes available it will be possible to examine these hypotheses in a more quantitative fashion.

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