In-plane thermal conductivity of $Nd_{2-x}Ce_xCuO_{4-y}$

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We present measurements of *ab*-plane thermal conductivity (κ) for a series of $Nd_{2-x}Ce_xCuO_{4-y}$ single crystals ($x=0.025,0.15,0.22$) at $T \leq 300$ K. For all specimens κ increases with decreasing temperature, rises sharply for $T<100$ K, and reaches a maximum at $T=20$–30 K. Near room temperature $\kappa \approx 12$ W/mK for the insulator ($x=0.025$) and $\kappa \approx 14$–16 W/mK in superconducting ($x=0.15$) and metallic ($x=0.22$) specimens. These differences are attributed to differing electronic contributions to heat conduction. No anomaly in $\kappa(T)$ was observed at the superconducting transition of the $x=0.15$ sample. For $T<T_c$, κ is suppressed slightly by the application of a strong magnetic field, consistent with an increase in phonon-electron scattering. The κ data are compared to previous results for the other cuprates.

The normal-state transport properties of the cuprates have been the focus of considerable experimental and theoretical investigation in recent years. The lower-T_c cuprate $Nd_{2-x}Ce_xCuO_{4-y}$ (NCCO) is of particular interest because it is viewed as electron doped and because the normal-state properties may be investigated over a broader temperature range.

In this Brief Report, we present measurements of in-plane thermal conductivity on a series of NCCO crystals, spanning the insulating (underdoped), superconducting, and metallic (overdoped) regimes. Our general finding is that NCCO behaves like a conventional phonon-dominated heat conductor. The electronic contributions to heat conduction are estimated to be rather small (<20%) and can account for small differences in the room-temperature values of κ. The more heavily Ce-doped specimens have substantially reduced conductivity maxima at low temperatures, consistent with increased phonon-defect scattering. The effect of superconductivity on the heat transport is determined from direct measurements of the normal-state κ for $T<T_c$ by applying a strong magnetic field.

The crystals used in this study were grown in CuO flux using a directional solidification technique, as described in detail elsewhere.[1] The platelike crystals were 50–75 μm thick (c axis), with 1–2-mm dimensions in the ab plane. Superconductivity (ac susceptibility onset $T_c \approx 24$ K) was induced in the $x=0.15$ specimen by annealing in a reducing atmosphere. The electrical resistivity,[1,2] Hall coefficient,[2] and thermopower[3] of similarly prepared crystals have been reported previously.

The thermal conductivity was measured by a steady-state method employing a differential Chromel-Constantan thermocouple and a small heater, glued to the specimens with varnish. Samples were suspended in a radiation-shielded vacuum can. The temperature gradient during measurement was typically 0.5–2.0 K/mm. 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 is estimated to be $\pm 10\%$. For the measurements in a magnetic field, the magnetothermopower of the thermocouple was calibrated against carbon-glass sensors in a separate experiment.

The in-plane thermal conductivity (κ) versus temperature is plotted for three specimens in Fig. 1. The insulator ($x=0.025$) has κ (300 K)$\sim$12 W/mK, somewhat smaller than κ (300 K)$\approx$13.5 W/mK ($x=0.15$, superconducting), and 15.5 W/mK ($x=0.22$, metallic). These differences are undoubtedly due, in part, to a larger electronic contribution to heat conduction in the superconducting and metallic specimens. Generally, we consider the total thermal conductivity to be a sum of lattice (κ_L) and electronic (κ_e) components. We may estimate an

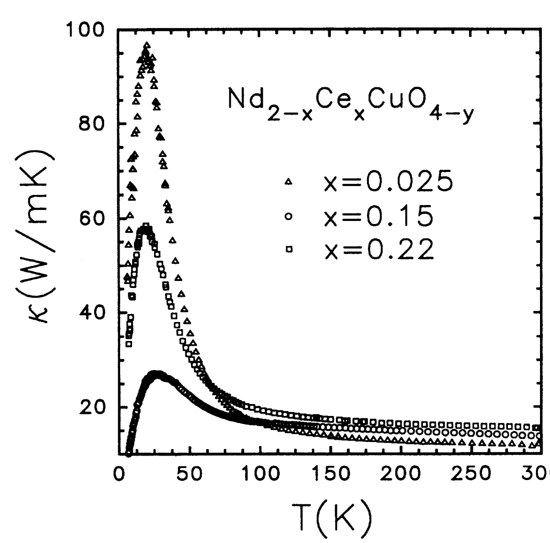

FIG. 1. In-plane thermal conductivity vs temperature for three NCCO crystals.

upper bound for κ_e by applying the Wiedemann-Franz law (WFL) $\kappa_e \leq L_0 T/\rho$ ($L_0 = 2.45 \times 10^{-8}$ Ω W/K^2). Using electrical resistivities[2,3] ρ(300 K)=450 $\mu\Omega$ cm ($x=0.15$) and 200 $\mu\Omega$ cm ($x=0.22$) implies $\kappa_e \leq 1.6$ and 3.7 W/mK, respectively. Thus, within the accuracy of the measurements, the differences in κ (300 K) for these three specimens are consistent with estimated differences in κ_e. From this, we conclude that κ_L (300 K) is comparable for the three specimens.

The increase in κ with decreasing temperature, the occurrence of a sharp maximum, and the rapid decrease in κ at the lowest temperatures are all characteristics of lattice heat conduction in insulating materials.[4] At temperatures approaching the Debeye temperature, phonon-phonon scattering tends to dominate the lattice heat conduction of nearly perfect insulators, giving rise to a $\kappa \propto 1/T$ behavior. The much weaker temperature dependencies evident for $T > 100$ K in Fig. 1 indicate that phonon-defect scattering is substantial in all specimens. Near the maximum (κ_{max}), defect scattering tends to dominate the lattice thermal resistance, with higher and sharper peaks expected in more perfect specimens. Generally, we anticipate that the cation disorder associated with the substitution of Ce for Nd will result in additional scattering of phonons at low temperature, leading to a suppression of κ_{max}. The $x=0.025$ specimen has the highest and sharpest maximum with $\kappa_{max} \approx 96$ W/mK at $T \approx 20$ K. However, the data for the $x=0.15$ and 0.22 specimens do not follow a simple trend. For $x=0.22$, $\kappa_{max} \approx 58$ W/mK at $T \approx 20$ K whereas the peak for $x=0.15$ is substantially smaller, $\kappa_{max} \approx 28$ W/mK, and occurs at $T \approx 25$ K.

One possibility we have considered is that the reducing anneal, to which the $x=0.15$ crystal was subjected to induce superconductivity, introduced defects that could account for its lower κ_{max}. To test this hypothesis, we annealed the $x=0.22$ specimen under similar conditions[2] and remeasured κ. These data are shown in the inset of Fig. 2. The decrease by ≈ 2 W/mK in the high-temperature value of κ for the annealed crystal can largely be accounted for by geometric uncertainties in the placement of the thermocouples, though a decrease in κ_e is also possible. The normalized data (Fig. 2) show clearly the reduction in the relative peak height, with little change in its temperature position. Also shown is the normalized data for the $x=0.15$ specimen.

The effects of the heat treatment were to reduce κ_{max}, though apparently not by as much as required to account for the observed difference in the data for the $x=0.15$ and 0.22 crystals. Chemical inhomogeneity (e.g., nonuniform Ce distribution) is another possible source of phonon scattering. Preliminary studies [5] of the nominally $x=0.15$ crystal using the color of polarization[6] and energy-dispersive x-ray analysis reveal distinct, phase-separated layers with differing Ce concentrations along the growth direction (c axis). This stratification undoubtedly introduces additional scattering at the interfaces between regions. Further measurements correlating κ with crystal chemistry and microstructure would be required to make more conclusive statements regarding phonon-defect scattering.

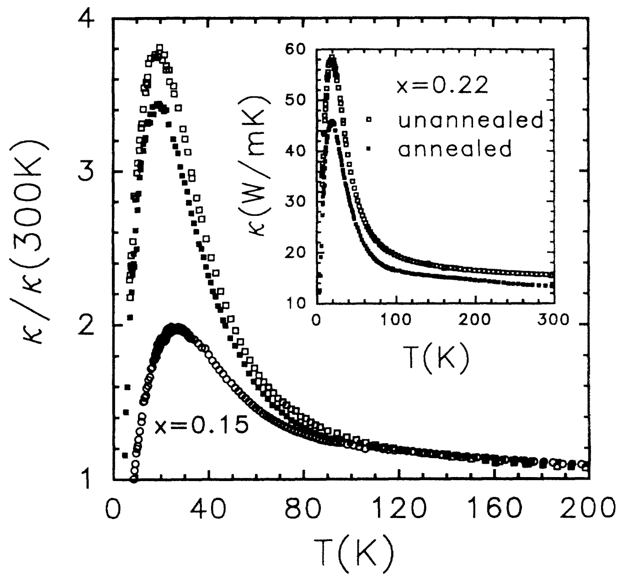

FIG. 2. In-plane thermal conductivity, normalized to values at 300 K, vs temperature for the $x=0.15$ and 0.22 (annealed and unannealed) crystals. The inset shows κ (T) for the $x=0.22$ crystal before and after the anneal.

Another issue we have explored is the effect of superconductivity on the heat flow in the $x=0.15$ crystal. In Fig. 3 we plot κ for the $x=0.15$ specimen, measured in zero magnetic field and in a field $H=9$ T parallel to the c axis. Note that the zero-field κ is slightly reduced (with T_{max} now ≈ 30 K) from the data of Fig. 1, the latter measured several months earlier. The origin of this aging effect is unknown. No deleterious effects of aging on the superconducting transition were observed.

For fields parallel to the c axis, magnetization measurements[7] on Sm-Ce-Cu-O crystals yield an upper critical-field slope $dH_{c2}/dT \approx -0.5$ T/K. We thus estimate that for a 9-T field our specimen is driven normal for $T \geq 6-7$ K. The data reveal a slight field-induced reduction in κ by ≈ 1 W/mK. In Fig. 4 we plot the normalized thermal

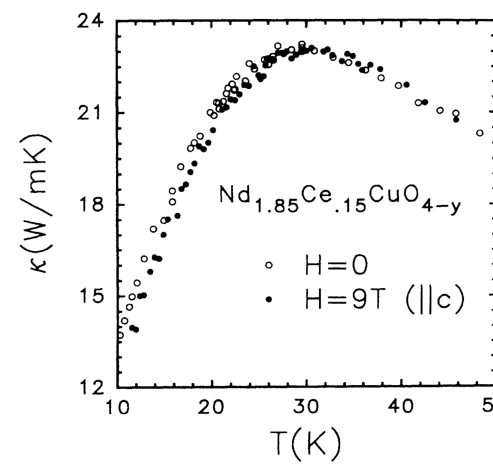

FIG. 3. In-plane thermal conductivity vs temperature for the superconducting crystal ($x=0.15$) with and without a 9-T magnetic field applied along the c axis.

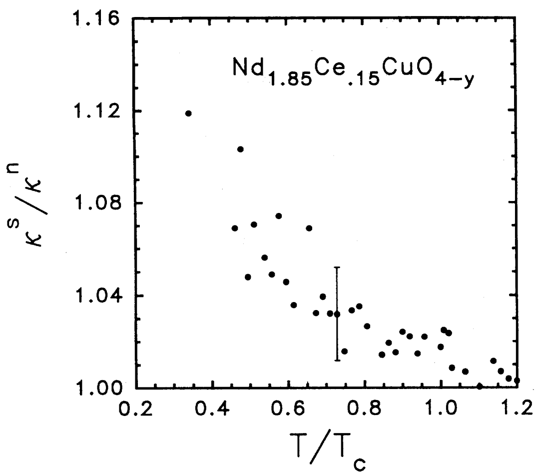

FIG. 4. Ratio of in-plane thermal conductivities in the superconducting and normal states, $\kappa^s/\kappa^n = \kappa\ (H=0)/\kappa\ (H=9\ \mathrm{T})$, plotted vs reduced temperature.

conductivity $\kappa^s/\kappa^n = \kappa\ (H=0)/\kappa\ (H=9\ \mathrm{T})$ versus reduced temperature. The error bar of $\pm 2\%$ represents the estimated uncertainty associated with the thermocouple calibration.

To interpret this result, we must consider the contributions from both κ_L and κ_e. At temperatures just above T_c, the electrical resistivities of nominally $x=0.15$ crystals[1-3] are nearly T independent (residual resistivity regime) and fall in the range 50–250 $\mu\Omega$ cm. Applying the WFL, these values correspond to κ_e in the range 0.3–1.2 W/mK. We see that, while κ_e represents only a small fraction ($<10\%$) of the total-heat transport for 10 K $\leq T \leq T_c$, it represents a substantial fraction of the *field-induced change* in κ.

Theoretically, κ_e decreases for $T < T_c$ when defects are the principal source of electron scattering (elastic-scattering limit).[8] The temperature independence of the electrical resistivity near T_c suggests that this is the appropriate description for NCCO. In contrast, κ_L tends to increase for $T < T_c$, as the formation of superconducting pairs reduces phonon-electron scattering. When we drive our specimen normal by turning on the field, we thus expect an increase in κ_e and a decrease in κ_L. The observed decrease in the total κ (Figs. 3 and 4) then implies that the reduction in κ_L is larger than the increase in κ_e.

The rather small field-induced change in κ indicates that κ_L is not strongly affected by scattering from carriers. This is not surprising, given that at these temperatures the long-wavelength acoustic phonons responsible for most of the heat conduction tend to scatter principally from defects and the crystal boundaries. Note that this conclusion is not necessarily inconsistent with a moderate or strong electron-phonon coupling parameter (λ), which represents an average over the entire phonon spectrum.

Finally, we compare the present results with measurements of in-plane κ in crystals of the hole-doped cuprates. All of the cuprates[9] (including NCCO) are similar in that phonons dominate the heat conduction. $YBa_2Cu_3O_{7-\delta}$ (YBCO) has the highest κ for the hole-doped materials. We find that the magnitude of κ for NCCO at $T=200$–300 K is larger than that of twinned[10] YBCO by a factor of 1–2, but comparable to that of untwinned YBCO.[11] YBCO crystals typically have electrical resistivities half that of NCCO, implying κ_e values twice as large. This suggests that κ_L is several W/mK larger in NCCO than in YBCO.

Another observation which suggests superior lattice conduction in NCCO is the temperature dependence of κ. From Fig. 2 we see that κ increases by $\approx 40\%$ as T decreases from 300 to 100 K. As noted above, this negative temperature coefficient of κ is the signature of phonon-phonon scattering. The substantially weaker temperature dependencies observed for the other materials in this regime suggest that phonon-defect scattering is relatively less important in NCCO. Analyses of κ_L in YBCO (Refs. 12 and 13) indicate that phonon-defect scattering accounts for more than half the thermal resistance at $T > T_c$. For $La_{2-x}Sr_xCuO_4$ ($x \leq 0.2$), the closest hole-doped analog of NCCO, κ is smaller than that of NCCO by a factor of 2 and decreases with decreasing temperature,[14] suggesting a more disordered lattice. The same can be said of κ in Bi 2:2:1:2 crystals.[15] Thus, with regard to lattice conduction, it appears that NCCO is "cleaner" than the other materials investigated to date.

To summarize, we have reported measurements of in-plane thermal conductivity on a series of NCCO crystals with various Ce concentrations. The data demonstrate that heat transport in NCCO is qualitatively similar to that of imperfect insulators, with a relatively small electronic contribution ($<20\%$). The low-temperature maximum in κ is suppressed with increasing Ce content and after a reducing heat treatment, consistent with increased phonon-defect scattering from cation and oxygen disorder. Generally, the lattice heat conductivity is larger and more strongly temperature dependent than in crystals of the other cuprates, implying relatively less phonon-defect scattering in NCCO. For the superconducting composition at $T < T_c$, κ is reduced in a magnetic field sufficient to drive the specimen normal, indicating an increase in phonon-electron scattering.

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