RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 43, NUMBER 10 1 APRIL 1991

Effect of magnetic field on thermal conductivity of $YBa_2Cu_3O_{7-\delta}$ single crystals

S. D. Peacor
Department of Physics, University of Michigan, Ann Arbor, Michigan 48109

J. L. Cohn
Materials Physics Branch, Naval Research Laboratory, Washington, D.C. 20375

C. Uher
Department of Physics, University of Michigan, Ann Arbor, Michigan 48109
(Received 26 December 1990; revised manuscript received 5 February 1991)

We report measurements of the thermal conductivity in a magnetic field on single crystals of $YBa_2Cu_3O_{7-\delta}$. Unlike in sintered samples, the zero-field thermal conductivity of these crystals above the superconducting transition temperature shows a marked decrease with increasing temperature, demonstrating the importance of phonon-phonon processes and the high quality of the crystals. We find that the thermal conductivity κ is very sensitive to the presence of fluxoids which are strong scatterers of phonons. The degradation of κ is particularly large when the magnetic field H is oriented perpendicular to the Cu-O planes. The anisotropic behavior of $\kappa(H)$ is in qualitative agreement with a geometric argument based on the anisotropy of the superconducting coherence length. We detect hysteresis at the level of several percent in the dependence of κ on the field.

The thermal conductivity κ of high-T_c superconductors has been an intensely studied subject[1] since the discovery of these remarkable materials. Unlike in conventional superconductors, lattice vibrations carry most of the heat in the normal state of the high-T_c materials, their dominance being more pronounced in sintered samples than in single crystals. As the temperature is lowered below the superconducting transition temperature T_c, a sharp increase in κ is observed. Although the charge-carrier thermal conductivity decreases in this situation due to the condensation of carriers into Cooper pairs, the large phonon contribution leads to an overall increase in κ due to the diminished scattering of phonons by the carriers. The rising thermal conductivity below T_c and a large peak observed near $T_c/2$ are distinct features of the heat transport in high-T_c superconductors. Theoretical modeling of the thermal conductivity has yielded an estimate on the carrier-phonon coupling strength[2] and insight[3] into the defect structure in the quasi-two-dimensional environment relevant to these perovskites.

In this paper we investigate the magnetic-field dependence of the *a-b* plane thermal conductivity of $YBa_2Cu_3O_{7-\delta}$ single crystals. In the normal state, a magnetic field is not expected to significantly alter the dynamics of the charge carriers (due to their low mobility) or the lattice; however, for $T < T_c$, such a field can have a dramatic effect on thermal transport. The way in which the field affects kinetic coefficients such as the thermal conductivity depends on how the flux enters a superconductor. In type-II superconductors, which encompass high-T_c perovskites, for fields above the lower critical field H_{c1}, the flux penetrates a homogeneous material in the form of single-quanta flux lines (Abrikosov's vortex filaments) arranged in a regular two-dimensional lattice. The density of fluxoids is controlled by the strength of the magnetic field and their average spacing decreases proportionally to $H^{-1/2}$. The presence of fluxoids may seriously impede the phonon heat current. The compact fluxoid cores in high-T_c materials should act as efficient scattering centers for phonons at temperatures not far below T_c where the phonon wavelength is comparable to the superconducting coherence length ξ. The thermal conductivity of high-T_c superconductors is thus expected to decrease significantly with increasing field in the mixed state.

An attempt to observe this effect has been made recently by Zhu *et al.*[4] in a study of sintered $YBa_2Cu_3O_{7-\delta}$ for fields of up to 3 T and temperatures $T < 25$ K. They detected a small decrease in the thermal conductivity amounting to 2–9% of the total conductivity. In their recent Letter describing transport entropy of vortex motion in $YBa_2Cu_3O_{7-\delta}$, Palstra *et al.*[5] indicated a significant decrease in the thermal conductivity of a single crystal in the field of 10 T directed perpendicular to the *a-b* plane. Similar changes in the thermal conductivity for fields of 3 T were seen by Florentiev *et al.*[6] In this Rapid Communication we describe a detailed study of the changes observed in the thermal conductivity of high-quality single crystals of $YBa_2Cu_3O_{7-\delta}$ arising from the effect of magnetic field directed both perpendicular and parallel to the Cu-O planes.

Measurements were performed on two crystals (designated sample 1 and sample 2, respectively) of typical size $2\times1\times0.2$ mm^3) grown by a self-flux method.[7] ac susceptibility measurements indicate superconducting transitions at 92 K with transition widths $\Delta T_c \sim 0.5$ K. The oxygen deficiency of these crystals is inferred from a study of similarly prepared crystals where the *c*-axis lattice parameter (a measure of oxygen content) is correlated with features in the dc magnetization hysteresis loops.[8] We estimate that $\delta \approx 0.1$ and 0.16 for crystals 1 and 2, respectively.

Thermal conductivity was measured using a steady-state technique with a temperature gradient applied along the *a*-*b* plane. The magnetic field was oriented perpendicular to the direction of the heat flow and could be rotated to achieve any desired angle θ with respect to the *c* axis of the crystal. One end of the sample was attached to a copper tip of the cryostat with Stycast epoxy. Two thin, copper strips, spaced about 0.7 mm apart and laid across the breadth of the sample, were affixed with Stycast to provide electrically insulated, isothermal platforms transverse to the temperature gradient. A chromel-constantan differential thermocouple, assembled from 0.001-in-diam wires, was soldered to these strips to monitor the temperature gradient along the sample. In addition, a simple thermocouple was soldered to a copper strip near the sink to establish the absolute sample temperature. Chromel-Constantan thermocouples were calibrated in a magnetic field and their sensitivity was found to vary with field in a similar way as reported in Ref. 9. For a heater we used a miniature 1 kΩ strain gauge which had a very thin piece of copper varnished to its back to ensure fast, uniform heating. The thermal response time of the experimental setup was relatively fast; in the entire temperature range investigated a steady state was reached within less than 30 s after applying heat. In all cases, magnetic field or angular sweeps at constant temperature were made on a zero-field-cooled sample to ensure that no initial flux was trapped in the sample. Temperature sweeps for fixed magnetic fields were performed by warming a zero-field-cooled sample. We observed no measurable difference in κ for field-cooled and zero-field-cooled configurations.

In Fig. 1 we show the temperature dependence of the thermal conductivity for both crystals in zero field and at several different magnetic fields oriented along the *c* axis of the crystals. Unfortunately, because crystal 1 broke when attempting measurements in a 3-T field, we have only one set of field measurements on this sample, the curve at 1.5 T. Crystal 2 shows a somewhat lower thermal conductivity than the first sample but we were able to measure the thermal conductivity up to fields of 6 T. We note that in these high-quality single crystals the thermal conductivity below T_c rises sharply and, at the maximum, the magnitude of κ is nearly twice the value at T_c. In sintered samples, for comparison, the corresponding increase in the thermal conductivity is typically only 10–15%. Clearly, the superior quality of the single-crystalline lattice allows for a greater enhancement in the phonon mean free path when the free carriers condense and this, in turn, leads to the larger increase in κ.

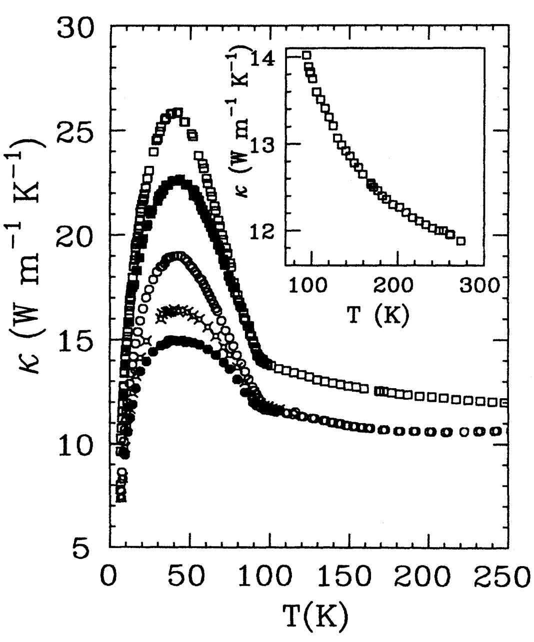

FIG. 1. Thermal conductivity for samples 1 and 2. Open and solid squares show sample 1 in zero field and at 1.5 T, respectively. Open circles, stars, and solid circles stand for sample 2 in zero field, 2, and 6 T, respectively. The inset presents a magnified plot of $\kappa(T)$ at $T > T_c$ for sample 1.

The high quality of the samples is also reflected in the behavior of the thermal conductivity above the superconducting transition temperature where we observe a distinctly decreasing thermal conductivity with increasing temperature. In this temperature range, the total thermal conductivity can be viewed as arising from two separate channels, free carriers and phonons. The nearly linear temperature dependence of the *a*-*b* plane electrical resistivity in $YBa_2Cu_3O_{7-\delta}$ implies, via an application of the Wiedemann-Franz law, a temperature-independent carrier thermal conductivity.[10] The temperature dependence of the measured thermal conductivity must then come from the phonon term. At these elevated temperatures the phonons will scatter on free carriers, defects, and other phonons. The phonon-carrier and phonon-defect scattering rates tend to constant values with increasing temperature,[11] thus leaving the phonon-phonon term to govern the temperature variation of κ. The dramatic decrease in κ with increasing temperature (displayed for better clarity in the inset of Fig. 1) attests to the importance of phonon-phonon scattering in our single crystals. In sintered specimens, the more pronounced defect scattering and higher radiation losses associated with a considerably lower thermal conductivity (a factor of 2–3 times in comparison to single crystals) can account for measurements which reveal temperature independent or increasing κ with temperature.

We now examine the magnetic-field dependence of κ at fixed temperature. As indicated in Fig. 1, the presence of fluxoids significantly diminishes the thermal conductivity, particularly in the range 20 K $< T <$ 60 K. As expected, for $T > T_c$ the magnetic field has no significant effect on κ. Figures 2(a) and 2(b) show the normalized magnetothermal conductivity $\kappa(H)/\kappa(0)$ at several temperatures for fields oriented perpendicular to the Cu-O planes. Note that the effect of magnetic field on the thermal conductivity increases as one cools below T_c, is greatest near the zero-field conductivity maximum at $T_{\max} \approx 40$ K, and then diminishes at lower temperatures. This behavior can be understood by the following qualitative arguments. As the temperature decreases below $T_{\max}$, the transport of heat is increasingly dominated by long-wavelength phonons. Because the phonon-carrier scattering rate for a given phonon mode is inversely proportional to the phonon

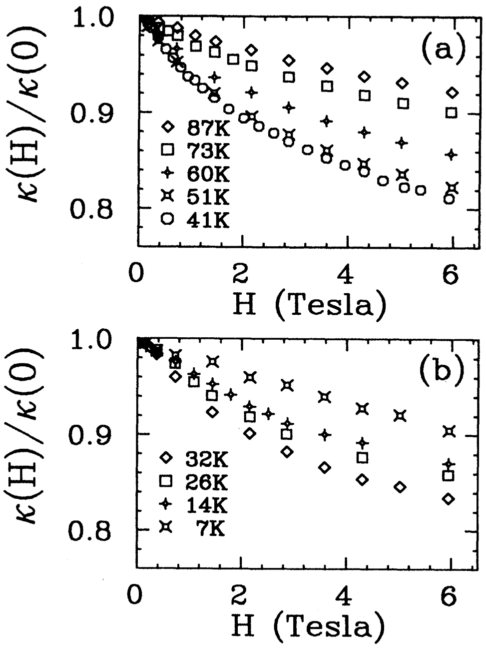

FIG. 2. (a),(b) Field dependence of the thermal conductivity at different temperatures for sample 2 (increasing field applied perpendicular to the *a*-*b* plane).

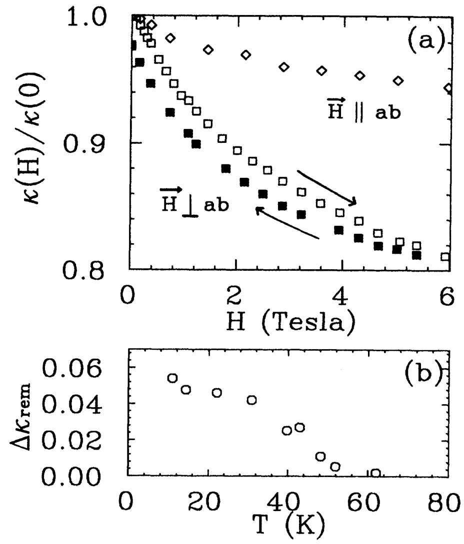

FIG. 3. (a) Field dependence of the thermal conductivity $\kappa(H)/\kappa(0)$ for sample 2 at 41 K. Diamonds indicate magnetic field oriented parallel to the *a*-*b* plane, squares represent a field perpendicular to the *a*-*b* plane. Open squares are for an increasing field, solid squares for a decreasing field. Note a significant hysteresis when the field is perpendicular to the *a*-*b* planes. There is virtually no hysteresis in the parallel-field orientation and $\kappa(H)$ retraces essentially the same points on the up and down field sweeps. (b) Temperature dependence of the normalized remnant thermal conductivity $\Delta\kappa_{rem}$ (defined in the text).

wavelength, the scattering of phonons by normal excitations in the vortex cores becomes less effective with decreasing temperature. Above T_{max}, as T tends to T_c the phonon scattering rate is increasingly determined by scattering from uncondensed carriers and other phonons. Thus, in the total phonon-scattering rate, the phonon-vortex interaction becomes less important. At its peak the zero-field heat conductivity is limited principally by defect scattering and thus "turning on" the phonon-carrier scattering with field has, at T_{max}, the largest relative effect on the overall scattering rate.

At all temperatures, $\kappa(H)$ continuously decreases with increasing field. If the thermal resistance were to increase in proportion to the number of flux lines, and hence to the average internal magnetic field B, we might expect κ to follow the relation $\kappa^{-1}=\alpha+\beta B$, where βB is the thermal resistance due to the flux lines and α is the thermal resistance due to all other scattering centers. Although this model predicts the general shape of $\kappa(H)$, it greatly overestimates the suppression of κ. The field is evidently less effective in reducing κ than this simple picture predicts. Clearly, further theoretical work is required to describe the field dependence of the thermal conductivity.

Two of the most interesting observations in the magnetic-field dependence of κ are hysteresis effects and strong anisotropy for fields oriented parallel and perpendicular to the Cu-O planes. Both of these are demonstrated in the $\kappa(H)$ data in Fig. 3(a) for sample 2 at 41 K. Hysteresis is evident for the field sweeps in the perpendicular orientation where, upon lowering the field (solid squares), a distinct change in the dependence of κ on H is measured as compared to that for increasing field (open squares). This hysteresis is evidently associated with trapped flux and is observed from 62 K down to the lowest temperatures measured. We define the normalized remnant thermal conductivity as $\Delta\kappa_{rem}=(\kappa_0-\kappa_{rem})/\kappa_0$, where κ_0 is the initial zero-field value of the thermal conductivity, and κ_{rem} is the value measured after applying and removing a field of 6 T. This quantity is plotted in Fig. 3(b) as a function of temperature. For $T>62$ K, $\Delta\kappa_{rem}$ is zero within the experimental resolution. In the range 40 K $< T <$ 62 K, $\Delta\kappa_{rem}$ increases rapidly with decreasing temperature, and for $T<40$ K the increase is more gradual. An overall increase in $\Delta\kappa_{rem}$ is consistent with an increasing remnant magnetic field with decreasing temperature as is widely observed in magnetization measurements. However, the particular temperature dependence of $\Delta\kappa_{rem}$ is modified by the relative importance of the fluxoid scattering which, as we have discussed above, changes with temperature. Magnetic hysteresis and time-dependent phenomena in the thermal conductivity of these specimens will be presented in more detail elsewhere.[12]

The most prominent feature of Fig. 3(a) is the significant anisotropy in $\kappa(H)$ for the two field orientations. Flux lines clearly appear far more effective in scattering phonons when they lie perpendicular to the planes rather than when they fit between them. This also follows from the angular dependence of the magnetothermal conductivity shown in Fig. 4(a). The anisotropy, defined by

$$\Delta\kappa_{\perp}/\Delta\kappa_{\parallel}\equiv[\kappa_{\perp}(T,H)-\kappa_{\perp}(T,0)]/[\kappa_{\parallel}(T,H)-\kappa_{\parallel}(T,0)]\,,$$

is approximately independent of field for $H>1.5$ T in the

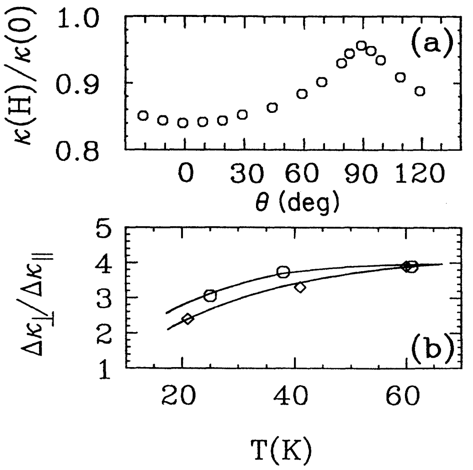

FIG. 4. (a) Angular dependence of the thermal conductivity for sample 1 at 41 K and $H = 1.5$ T. θ is the angle between the magnetic field and the c axis of the crystal. (b) Anisotropy $\Delta\kappa_\perp/\Delta\kappa_\parallel$ (defined in the text) for specimen 1 at 1.5 T (circles) and specimen 2 at 3 T (diamonds). The solid line is a guide to the eye.

range 20 K $< T <$ 60 K. In Fig. 4(b) we plot $\Delta\kappa_\perp/\Delta\kappa_\parallel$ versus temperature for specimen 1 at 1.5 T and specimen 2 at 3 T. We see that the anisotropy increases from a value of about 2 at 20 K and tends to saturate at a value near 4 at higher temperatures. For a uniform lattice of fluxoids penetrating the entire crystal, we should expect $\Delta\kappa_\perp/\Delta\kappa_\parallel \approx \xi_{ab}/\xi_c \sim 3$–5, the ratio of the superconducting coherence lengths. This follows if we assume that the field-induced phonon-carrier scattering scales with the cross-sectional area of vortices transverse to the heat flow. At higher temperatures the data are in reasonable agreement with this simple picture. Deviations of the anisotropy from the nominal value of 3–5 are probably associated with flux pinning and distortions in the field penetration profile. At lower temperatures and fields, where pinning is strong, we expect the simple picture to be less appropriate, in qualitative accord with the data.

In summary, we have shown that the thermal conductivity of high-quality single crystals of $YBa_2Cu_3O_{7-\delta}$ is very sensitive to the presence of magnetic vortices and the phonon-fluxoid scattering leads to a significant reduction in the phonon mean free path. The scattering of phonons on the vortices is highly anisotropic and is most effective when the flux lines are oriented perpendicular to the Cu-O planes. The anisotropy in $\kappa(H)$ is approximately given by the coherence length ratio ξ_{ab}/ξ_c, in agreement with a simple geometric argument. The great sensitivity of the phonon mean free path to the flux-line lattice density offers promise for using the thermal conductivity as a probe of the superconducting mixed state and associated magnetic relaxation processes. Such a novel application of thermal-conductivity measurements is now being investigated.

We are grateful for the efforts of T. A. Vanderah in preparing the single crystals of $YBa_2Cu_3O_{7-\delta}$ for this research and to M. S. Osofsky for measuring ac susceptibility. We wish to acknowledge useful discussions with F. Nori and R. A. Richardson. This work was supported by U.S. Army Research Office Contract No. DAAL-03-87-K-0007. One of the authors (J.L.C.) acknowledges financial support from the Office of Naval Technology.

[1]See, e.g., a review by C. Uher, J. Supercond. **3**, 337 (1990), and references therein.

[2]L. Tewordt and Th. Wölkhausen, Solid State Commun. **70**, 839 (1989).

[3]D. T. Morelli, G. L. Doll, J. Heremans, M. S. Dresselhaus, A. Cassanho, D. R. Gabbe, and H. P. Jenssen, Phys. Rev. B **41**, 2520 (1990).

[4]Da-Ming Zhu, A. C. Anderson, T. A. Friedmann, and D. M. Ginsberg, Phys. Rev. B **41**, 6605 (1990).

[5]T. T. M. Palstra, B. Batlogg, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. **64**, 3090 (1990).

[6]V. Florentiev, A. Inyushkin, A. Taldenkov, O. Melnikov, and A. Bykov, in *Progress in High Temperature Superconductivity*, edited by R. Nicolsky (World Scientific, Singapore, 1990), Vol. 25, p. 462.

[7]T. A. Vanderah, M. S. Osofsky, C. K. Lowe-Ma, E. A. Skelton, D. E. Bliss, and M. W. Decker (unpublished).

[8]M. S. Osofsky, J. L. Cohn, S. Qadri, E. A. Skelton, R. J. Soulen, and S. A. Wolf (unpublished).

[9]H. H. Sample, L. J. Neuringer, and L. G. Rubin, Rev. Sci. Instrum. **45**, 1 (1974).

[10]Here it is tacitly assumed that the scattering is strictly elastic. Although this is rather unlikely in view of the high crystalline perfection of the samples, any inelasticity in the carrier scattering would lead to a decrease in the Lorenz ratio with decreasing temperature, i.e., an increase in the carrier heat conductivity with increasing T.

[11]See, e.g., J. M. Ziman, *Electrons and Phonons* (Clarendon, Oxford, 1960).

[12]S. D. Peacor *et al.* (unpublished).