Thermal conductivity of polycrystalline $YBa_2Cu_3O_{7-\delta}$ in a magnetic field

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(Received 10 October 1989)

The thermal conductivity of high-purity sintered $YBa_2Cu_3O_{7-\delta}$ powder, in both zero magnetic field and a field of 3 T, has been measured at temperatures from 5 to 26 K. There is a reduction in the thermal conductivity when the magnetic field is applied perpendicular to the direction of heat flow. The results are compared with a crude model that assumes the scattering of phonons by magnetic fluxoids.

It is known that a magnetic field can strongly affect the thermal-transport properties of superconducting metals and alloys.[1-8] For type-II superconductors, it has been found that the thermal conductivity starts to decrease at the lower critical field H_{c1}, passes through a minimum, then increases with increasing field until reaching the normal-state value at the upper critical field H_{c2}. This behavior is caused by magnetic fluxoids penetrating into the metal or alloy. At low fields (much smaller than H_{c2}), electronic excitations within the fluxoids contribute a negligible amount to the total thermal conductivity, while the scattering of phonons and electrons by the fluxoids causes the net thermal conductivity to decrease. As the field approaches H_{c2}, an increasing density of electronic excitations within the fluxoids contributes to the thermal transport, causing the thermal conductivity to increase.

Little attention has been directed to the magnetic-field dependence of the thermal conductivities of the recently discovered high-temperature superconducting compounds,[9] although a number of experimental measurements of the thermal conductivities in zero field have been performed.[10,11] Here we report the results of a study of the effects of a magnetic field on the thermal conductivity of a polycrystalline sample of $YBa_2Cu_3O_{7-\delta}$. The results show that the thermal conductivity decreases when a magnetic field of 3 T is applied to the sample, and that the magnitude of the drop depends on the angle between the heat flow and the field direction.

The sample was prepared by thoroughly mixing and grinding with an agate mortar and pestle $BaCO_3$, Y_2O_3, and CuO powders (99.999% pure) and reacting the mixture in air at 950 °C for 24 h, with two intermediate grindings. After being treated in a stream of pure oxygen at 900 °C for 24 h, the powder was cooled to room temperature at a rate of 12 °C/h, and was ground again. The powder was then pressed into a pellet in a 2-cm-diam die using a pressure of 25 MPa applied for 5 min. Finally, the pellet was heated to 930 °C for 24 h and slowly cooled to room temperature at the same cooling rate in flowing oxygen. For this final heat treatment, the pressed pellet was placed on top of smaller pellets of $YBa_2Cu_3O_{7-\delta}$ that in turn rested on a platinum sheet.

A piece was cut from the pellet with a diamond saw to perform magnetization and x-ray powder-diffraction analysis. The results of magnetic susceptibility measurements, plotted in Fig. 1, show a sharp superconducting transition near 90 K, together with a small knee near 75 K in the zero-field-cooled (ZFC) data. This knee probably is due to intergrain coupling in the pelletized sample, which causes flux to be excluded from voids in the sample.[12] The powder x-ray diffraction measurements indicate impurity concentrations of Y_2BaCuO_5 of about 2% in weight, CuO of about 1% in weight, and no $BaCuO_2$.

A rectangular piece of dimensions $2.5 \times 3.5 \times 20$ mm^3 was cut from the pellet for thermal-conductivity measurements. The mass density of the sample, 5240 ± 140 kg/m^3, is about 83% of ideal crystal density.[13] The thermal conductivity was measured with a conventional steady-state heat flow method, with one end of the sample anchored to a copper block in a helium cryostat and a carbon thermometer attached to the opposite end of the sample. Two strain gauges,[14] anchored at two positions along the sample, were used as electrical heaters. The carbon thermometer was calibrated *in situ* against a calibrated carbon-glass commercial thermometer.[15] The magnetoresistance of the carbon-glass thermometer was further calibrated against a capacitance thermometer in magnetic fields, and the results of this calibration were found to be consistent with the general behavior of magnetoresistance in carbon-glass resistors.[16]

The magnetic field was provided by a superconducting

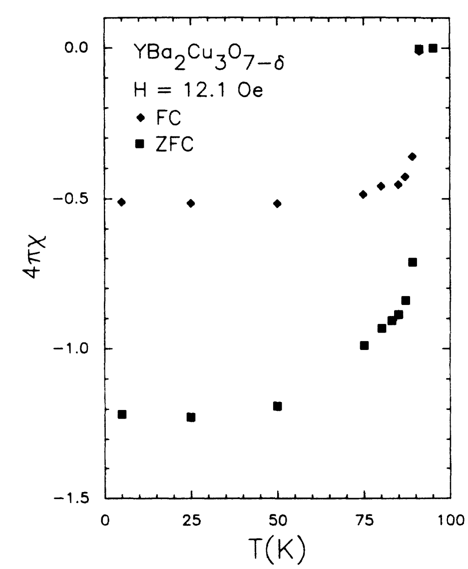

FIG. 1. The magnetic susceptibility of the polycrystalline $YBa_2Cu_3O_{7-\delta}$ sample, showing a sharp superconducting transition near 90 K, measured in both ZFC and FC conditions.

solenoid. The sample was first mounted on an insert oriented so that the heat flow was directed perpendicular to the field. After measuring its thermal conductivity, the sample was then remounted, with the heaters and thermometers untouched, so that the heat flow was directed parallel to the field. The thermal conductivity in zero field was measured with the sample oriented in both directions, and the results agreed. To determine whether the magnetic field would be distorted by the superconducting sample, we also measured the magnetic moment of the sample at temperatures from 5 to 27 K and in a field of 3 T with the field direction parallel to the long axis of the sample. The results indicated that more than 99% of the field penetrated into the sample, which agrees with the data of Reeves *et al.* for a similar sample.[12] Therefore, the magnetic field external to the sample can be assumed to be essentially undisturbed by the presence of the sample.

The thermal conductivities in zero field and in a magnetic field of 3 T, with the field both perpendicular and parallel to the heat flow, are shown in Fig. 2. The thermal conductivity in zero field shown here is about 20% higher than that of a $YBa_2Cu_3O_{7-\delta}$ sample with a density 80% of the ideal crystal density,[17] and about a factor of 3 larger than that of a sample with a density 70% of the ideal crystal density.[18] This trend is consistent with an inverse relationship between sample porosity and thermal conductivity.[18] As can be seen in Fig. 2, the thermal conductivity in a magnetic field is smaller than that in zero field, and the decrease is greater when the field direction is perpendicular to that of the heat flow. The fractional change in the thermal conductivity is displayed in Fig. 3.

The decrease in thermal conductivity is obviously caused by a magnetic-field-induced scattering of heat carriers. The field applied here is larger than H_{c1}, but much smaller than H_{c2} (less than 0.1 H_{c2}, based on a current estimate[19]). The material is therefore in the mixed state, with magnetic fluxoids penetrating the sample. As mentioned earlier, both phonons and electrons can be scattered by fluxoids, causing the thermal conductivity to decrease. It is usually difficult to distinguish between these two mechanisms except in some temperature range or in other special cases where either phonons or electrons are the dominant heat carriers. For $YBa_2Cu_3O_{7-\delta}$, several measurements have shown that electrons contribute only about 10% of the total thermal conductivity above T_c.[10,20] Below T_c, although a detailed knowledge of the ratio of the electronic contribution to the total thermal conductivity is still not available, there are indications that the phonons remain the dominant heat carriers to lower temperatures.[10,18] Thus, we will assume that phonons are the dominant source of thermal transport in our $YBa_2Cu_3O_{7-\delta}$ sample, and that the decrease in thermal conductivity in a magnetic field is caused primarily by the scattering of phonons at magnetic fluxoids.

The scattering of phonons by fluxoids in a superconductor depends on the angle between the phonon wave vector and the axis of a fluxoid. A simple calculation[21] of thermal conductivity limited by phonon scattering from cylindrical holes shows that the thermal conductivity along a direction parallel to the axis of the holes is a factor of 2 greater than that perpendicular to the axis of the holes, which qualitatively explains the angle dependence shown in Figs. 2 and 3. A similar angular dependence, also due to phonon scattering from fluxoids, is seen in conventional superconductors.[3,8]

We next attempt to compare quantitatively the mea-

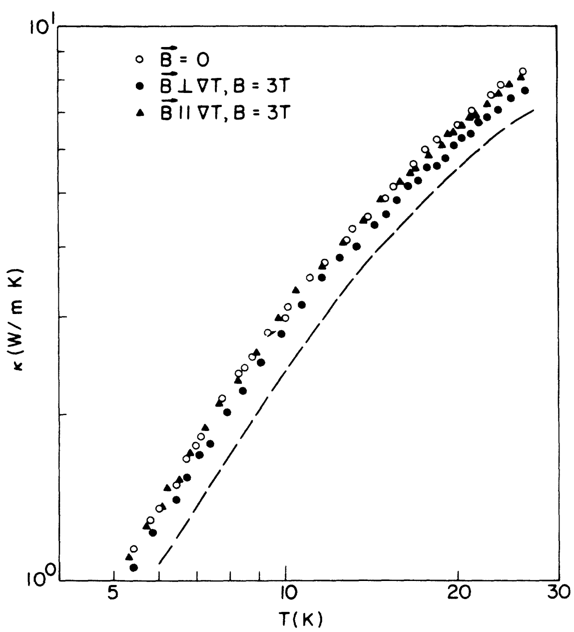

FIG. 2. The thermal conductivity of polycrystalline $YBa_2Cu_3O_{7-\delta}$ both in zero field and in a field of 3 T vs temperature. ○, in zero magnetic field; ●, in a 3-T field oriented perpendicular to the heat flow; ▲, in a 3-T field oriented parallel to the heat flow; dashed line, the zero-field data (Ref. 17) of a $YBa_2Cu_3O_{7-\delta}$ sample with 80% of the mass density of an ideal crystal.

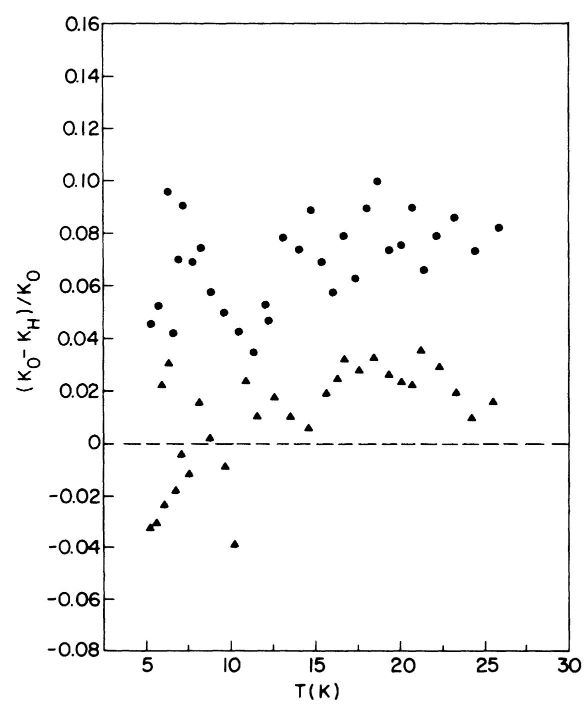

FIG. 3. Fractional changes of thermal conductivity of polycrystalline $YBa_2Cu_3O_{7-\delta}$ in a magnetic field of 3 T. ●, field perpendicular to the heat flow; ▲, field parallel to the heat flow.

sured decrease in the thermal conductivity of $YBa_2Cu_3O_{7-\delta}$ with the prediction of a model of phonon scattering by fluxoids. The phonon-scattering rate in a magnetic field can be expressed as a sum of the phonon-scattering rate from fluxoids and the phonon-scattering rate in zero field, provided that these are independent scattering mechanisms. Also, a recent measurement[22] of the sound velocity of $YBa_2Cu_3O_{7-\delta}$ as a function of magnetic field indicated no change to 8 T. Therefore, under the dominant phonon approximation, the thermal resistivity $\kappa_{\text{ph-}f}^{-1}$ caused by phonon scattering solely from fluxoids can be expressed in terms of the measured thermal conductivities in a magnetic field (κ_H) and in zero field (κ_0), $\kappa_{\text{ph-}f}^{-1}=\kappa_H^{-1}-\kappa_0^{-1}$. In a crude model,[3,4] which assumes that the electronic spectrum of energy levels in a fluxoid core is essentially the same as in the normal state while the remaining volume of the sample does not differ appreciably from Meissner state, the phonon mean free path $l_{\text{ph-}f}$ limited by scattering from fluxoids can be expressed as

$$l_{\text{ph-}f} \approx (\xi n \sigma)^{-1}, \tag{1}$$

where ξ is the coherence length (which is considered to be approximately equal to the diameter of the fluxoids) of the superconducting state, n is the density of fluxoids, and σ is the scattering probability. If the phonon mean free path in the normal state, as limited by scattering from electrons, is $l_{\text{ph-}e}$ the scattering probability can be expressed approximately as $\sigma \approx \xi / l_{\text{ph-}e}$ for $\xi < l_{\text{ph-}e}$. Substituting this into Eq. (2) gives $l_{\text{ph-}f} \approx (\xi^2 n)^{-1} l_{\text{ph-}e}$. Using the relations $n \approx B/\Phi_0$ and $\xi^2 \approx \Phi_0 / H_{c2}$, where B is the applied magnetic field and Φ_0 is the flux quantum, $\kappa_{\text{ph-}f}^{-1}$ can be related to the phonon thermal resistivity $\kappa_{\text{ph-}e}^{-1}$ caused by the scattering of phonons by electrons in the normal state of the material,[3,4] $\kappa_{\text{ph-}f}^{-1}/\kappa_{\text{ph-}e}^{-1}=l_{\text{ph-}f}^{-1}/l_{\text{ph-}e}^{-1} \approx B/H_{c2}$.

Thus $\kappa_{\text{ph-}f}^{-1}$ deduced from measurements, multiplied by a factor H_{c2}/B, should be approximately equal to the inverse phonon thermal conductivity in the normal state ($\kappa_{\text{ph-}e}^{-1}$). In Fig. 4 we plot $\kappa_{\text{ph-}f}^{-1}$ (H_{c2}/B) for $YBa_2Cu_3O_{7-\delta}$, as well as for $GdBa_2Cu_3O_{7-\delta}$ (from Ref. 9) and three conventional superconductors. The H_{c2} for $YBa_2Cu_3O_{7-\delta}$ was taken to be the average of the a, b, and c directions.[19] Note that $\kappa_{\text{ph-}f}^{-1}$ is proportional to the scattering rate of phonons by "normal state" electrons in the fluxoid cores. As can be seen in Fig. 4, this phonon scattering rate is consistently larger in conventional superconductors than in $YBa_2Cu_3O_{7-\delta}$ or $GdBa_2Cu_3O_{7-\delta}$. Perhaps this is because the carrier densities and electron mean free paths in the normal states of the high-T_c materials are about an order of magnitude smaller than those in conventional metals and alloys (see Table I). To test this possibility, we next estimate theoretically the magnitudes of $\kappa_{\text{ph-}e}$ for these two classes of superconductors.

Pippard[23] developed a theory of the phonon-electron interaction to describe acoustic attenuation in metals. From his theory the phonon conductivity, limited by scattering from free electrons, can be written as[24]

$$\kappa_{\text{ph-}e}=\frac{Dk^2T^2}{2(3\pi^2 N)^{4/3}\hbar^2}\int_0^\infty F(y)\frac{x^3 e^x}{(e^x-1)^2}dx, \tag{2}$$

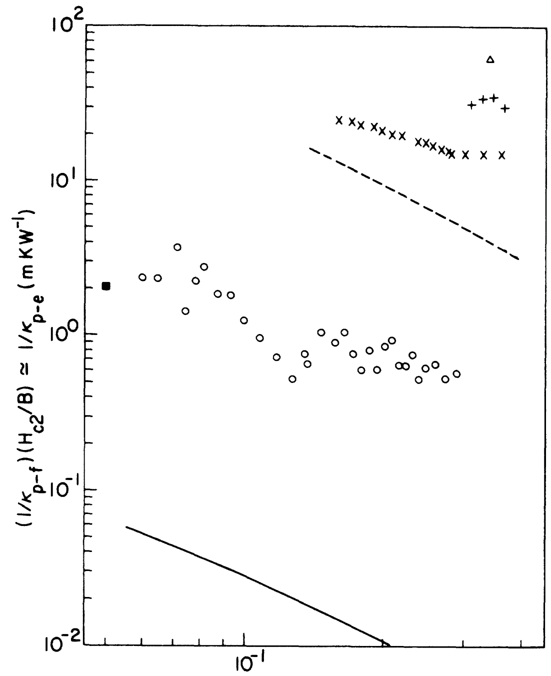

FIG. 4. Thermal resistivity $\kappa_{\text{ph-}e}^{-1}$, caused by phonon scattering from fluxoids vs reduced temperature T/T_c. $\kappa_{\text{ph-}e}^{-1}$ is roughly equal to $\kappa_{\text{ph-}f}^{-1}$ (H_{c2}/B) as explained in the text. ○, $YBa_2Cu_3O_{7-\delta}$ (this work); ■, $GdBa_2Cu_3O_{7-\delta}$ (Ref. 9, obtained by assuming that the decrease in thermal conductivity in a magnetic field is caused completely by the scattering of phonons from fluxoids, and that H_{c2} for $GdBa_2Cu_3O_{7-\delta}$ is the same as that for $YBa_2Cu_3O_{7-\delta}$); +, $Nb_{0.8}Mo_{0.2}$ (Ref. 4, $H=0.0258$ T); ×, $Nb_{0.98}Ta_{0.02}$ (Ref. 4, $H=0.0258$ T); △, vanadium (Ref. 8). The magnetic field was perpendicular to the heat flow in all cases, except possibly $GdBa_2Cu_3O_{7-\delta}$ for which the direction was not specified. The solid and dashed lines are the theoretical curves for $YBa_2Cu_3O_{7-\delta}$ and $Nb_{0.98}Ta_{0.02}$ assuming normal-state electrons in the fluxoids are responsible for the phonon scattering.

where D is the mass density and N is the free-electron density. The function $F(y)$ is related to the phonon-electron interaction. For the longitudinal phonon mode,

$$F(y)=\left[\frac{y\tan^{-1}y}{3(y-\tan^{-1}y)}-\frac{1}{y}\right]^{-1}, \tag{3}$$

where $y=ql_e$, q is the phonon wave number, and l_e the electron mean free path. It has been recognized that in order to account for the observed lattice thermal conductivity in many metals the attenuation of transverse modes should be approximately equal to that of the longitudinal mode.[25] Assuming that the attenuations for longitudinal and transverse modes are the same and the ratio between the longitudinal and transverse sound velocities is two, we have calculated the thermal resistivity $\kappa_{\text{ph-}e}^{-1}$ for $YBa_2Cu_3O_{7-\delta}$, $Nb_{0.98}Ta_{0.02}$, $Nb_{0.8}Mo_{0.2}$, and vanadium using Eqs. (2), (3), and the parameters listed in Table I. For conventional superconductors, the results of the calculation, shown in Fig. 4, agree reasonably well with the experimental data in magnitude (within a factor of about 3) and in temperature dependence. For the high-temperature superconductors, however, the calculation

TABLE I. Values of the parameters used in calculations. The last column is the ratio between measured and calculated values.

Materials	T_c (K)	l_e (Å)	$10^{-28}N$ (m^{-3})	D (kg/m^3)	$10^{-3}v_l$ (m/s)	$(\kappa_{ph\text{-}e})_{expt}/(\kappa_{ph\text{-}e})_{calc}$ at $T/T_c \approx 0.35$
$Nb_{0.98}Ta_{0.02}$	8.58[a]	143[a]	5.56[b]	8560[c]	5.1[c]	0.3
$Nb_{0.8}Mo_{0.2}$	4.23[a]	95[a]	5.56[b]	8760[c]	5.1[c]	0.3
V	5.06[d]	360[d]	14.4[b]	6100[c]	6.1[c]	0.8
$YBa_2Cu_3O_{7-\delta}$	90.	17[e]	0.9[e]	6310[f]	7.0[g]	0.014

[a]References 3 and 4. The electron mean free path in $Nb_{0.98}Ta_{0.02}$ is taken to be the same as that in $Nb_{0.98}Mo_{0.02}$.
[b]Reference 29. The carrier densities of $Nb_{0.98}Ta_{0.02}$ and $Nb_{0.8}Mo_{0.2}$ are taken to be the same as that of pure Nb. The carrier density of V was calculated by using Eq. (1.1) in Ref. 29 and assuming a valence of 2.
[c]Reference 30. The mass densities of $Nb_{0.98}Ta_{0.02}$ and $Nb_{0.8}Mo_{0.2}$ were determined by averaging over the density of each pure element according to their atomic percentage in the alloys. The longitudinal sound velocities of $Nb_{0.98}Ta_{0.02}$ and $Nb_{0.8}Mo_{0.2}$ were taken to be the same value as that of Nb. The values listed are the sound velocities averaged over all directions.
[d]Reference 8.
[e]Reference 26.
[f]Reference 13.
[g]References 18 and 28.

gives a value for $\kappa_{ph\text{-}e}^{-1}$ which is much smaller than the experimental values, although the temperature dependence of the calculated $\kappa_{ph\text{-}e}^{-1}$ roughly agrees with the experimental results. If our view of phonon scattering by fluxoids is correct, this discrepancy would indicate that a theory based on the free electron model is inadequate, perhaps because high-temperature superconductors are highly anisotropic and intrinsically inhomogeneous in structure.[27,31]

In summary, we have shown that the application of a magnetic field reduces the thermal conductivity of superconducting $YBa_2Cu_3O_{7-\delta}$. We interpret this decrease as being caused by phonon scattering from fluxoids. The magnitude of the decrease is similar to that caused by the scattering of phonons by fluxoids in conventional superconductors. We theoretically estimated the decrease using a very crude model, and obtained a smaller decrease than that indicated by the experimental results.

We thank M. B. Weissman for use of his cryostat, and G. Alers for assistance during the course of the experiment. This work was supported in part by Department of Energy, Division of Materials Science Contract No. DE-AC0276ER01198 through the University of Illinois Materials Research Laboratory (D.-M.Z. and A.C.A.), and in part by National Science Foundation Grant No. NSF-DMR-87-14555 (T.A.F. and D.M.G.).

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