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Thermal conductivity of superconductive Y-Ba-Cu-O

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We have measured the thermal conductivity of a superconducting Y-Ba-Cu-O compound as a function of temperature. We find that the thermal conductivity is nearly temperature independent as temperature is lowered below 140 K, but increases sharply at the transition temperature, which is about 85–90 K for our sample. These results can be accounted for by a reduction in phonon-electron scattering below T_c in this material.

INTRODUCTION

The area of high-temperature superconductors has experienced something of a revolution over the past few months. Within this time frame, the highest known transition temperature of any material has been raised from about 23 K (that of Nb_3Ge) to nearly 100 K. The groundwork for this extraordinary increase in T_c's was laid by Bednorz and Müller,[1] who discovered that a compound of lanthanum, barium, copper, and oxygen became superconducting at 36 K. Shortly thereafter a major breakthrough was achieved when Wu *et al.*[2] found superconductivity above liquid-nitrogen temperature in a compound of yttrium, barium, copper, and oxygen. These first experiments have given rise to a veritable flood of experimental and theoretical papers on these ceramic materials. On the theoretical side, attempts have been made to explain the high-transition temperature using, rather than a conventional BCS approach, more exotic theories such as electron-plasmon coupling[3] and a new electronic ground state.[4] While most of the experiments have concentrated on the electrical and magnetic properties of these fascinating materials, only a relatively few measurements of their thermal properties, such as specific heat [5] and thermoelectric power,[6] have been reported. Because the superconductive transition temperature is high, the phonon contribution completely dominates the specific heat, and the classical disappearance of the electronic specific heat when the sample becomes superconducting is very hard to observe.[5] This paper reveals that, on the contrary, there is a large effect in the thermal conductivity when the sample is cooled below the superconducting transition.

Whether the electron-phonon interaction in these materials can produce the high-transition temperatures observed to date is a critical question which has not yet been answered conclusively. In this regard, thermal conductivity measurements are an indispensable tool. Since heat is conducted by both charge carriers and phonons, a measurement of the thermal conductivity can yield valuable information not only about the spectra of electrons and phonons, but also the interactions between them. The results we report here do indeed shed some very important light on the nature of the electron-phonon interaction in this material.

EXPERIMENTAL TECHNIQUE

In order to obtain the superconducting phase $YBa_2Cu_3O_7$,[5] appropriate amounts of Y_2O_3, $BaCO_3$, and CuO powders were mixed together and heated at 875 °C in air in a quartz crucible for approximately 20 h. The resulting calcined material was ground using an Al_2O_3 mortar and pestle, and pressed at 1200 psi with a $\frac{3}{4}$ in. die. The pellet so produced was then annealed in air at 950 °C for 24 h, and, after cooling to ambient temperature , was reannealed in O_2.

A steady-state four-probe technique was used to measure the thermal conductivity of our sample. A parallelpiped of approximate dimensions $3\times5\times10$ mm^3 was cut from one pellet of $YBa_2Cu_3O_7$. On one end of the sample was placed a small metal film resistor embedded in a copper holder. The other end of the sample was placed in a copper clamp which was screwed to the cold tip of a closed-cycle helium refrigerator. By passing a known current through the resistor, a given amount of Joule heat can be passed through the sample and produce a temperature difference ΔT. If the rate of Joule heating in the resistor is P, the thermal conductivity is then given by

$$\kappa=(P/\Delta T)(L/S),$$

where L is the distance across which the temperature difference is measured, and S is the sample's cross-sectional area. In this experiment, we used a Chromel-Constantan thermocouple to monitor ΔT, with an uncertainty of about 2% above 50 K and about 5% at lower temperatures.

RESULTS AND DISCUSSION

Figure 1 shows our results for the electrical resistivity of our sample of $YBa_2Cu_3O_7$. In order to minimize self-heating and critical current effects, the measuring current was kept at 10 mA. At room temperature, $\rho\sim19\times10^{-3}$ Ω cm, and decreases roughly linearly as the temperature is lowered. At 100 K, the resistivity begins to drop precipitously, and by 86 K, a zero-resistance state is achieved. The width of the transition (90%–10%) is approximately 5 K, indicating that this sample may consist of a substan-

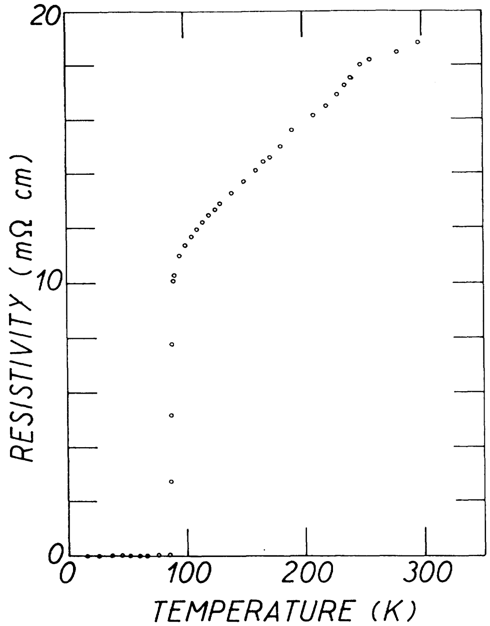

FIG. 1. Electrical resistivity of $YBa_2Cu_3O_7$ as a function of temperature.

tial amount of mixed phase. The superconductivity was verified independently by submerging the sample in liquid nitrogen and observing its repulsion of a small permanent magnet.

In Fig. 2 we present our results for the thermal conductivity of the same sample down to 15 K. We see that as the temperature is lowered below 140 K, the thermal conductivity is nearly constant. As T_c is approached, we observe a sudden increase in κ. This is clearly seen in the inset of Fig. 2, where the data are replotted on a linear scale. The thermal conductivity continues to rise as temperature is lowered below T_c, and reaches a peak at approximately 55 K, before falling off at lower temperatures.

As mentioned above, the thermal conductivity measured experimentally is the sum of conduction due to carriers (κ_c) and that due to phonons (κ_p); thus

$$\kappa = \kappa_c + \kappa_p \ .$$

In most metals, the thermal conductivity is dominated by the carrier contribution, which is large due to the large carrier densities found in most metallic materials. One can make an estimate of κ_c by using the Wiedemann-Franz law in conjunction with the electrical resistivity data. If the carriers are being scattered elastically, then one expects that

$$\kappa_c \rho = L_0 T \ ,$$

where $L_0 = 2.45 \times 10^{-8}$ W Ω K^{-1} is the Lorenz number. If there is significant inelastic scattering of the carriers, then the effective Lorenz number is always smaller than L_0. Thus we can obtain an upper limit to the carrier thermal conductivity by using our electrical resistivity data above the transition; we find $\kappa_c < 4 \times 10^{-4}$ W cm^{-1} K^{-1}. This is more than two orders of magnitude less than the measured room-temperature thermal conductivity. The implication is that in this Y-Ba-Cu-O compound, nearly all of the heat is transported by lattice vibrations.

One intuitively expects that the onset of superconductivity affects the way in which heat is conducted in two ways. First, those electrons which are condensed into Cooper pairs cannot carry entropy and therefore do not transport heat. Thus one expects an exponential decay of the carrier thermal conductivity below the transition.

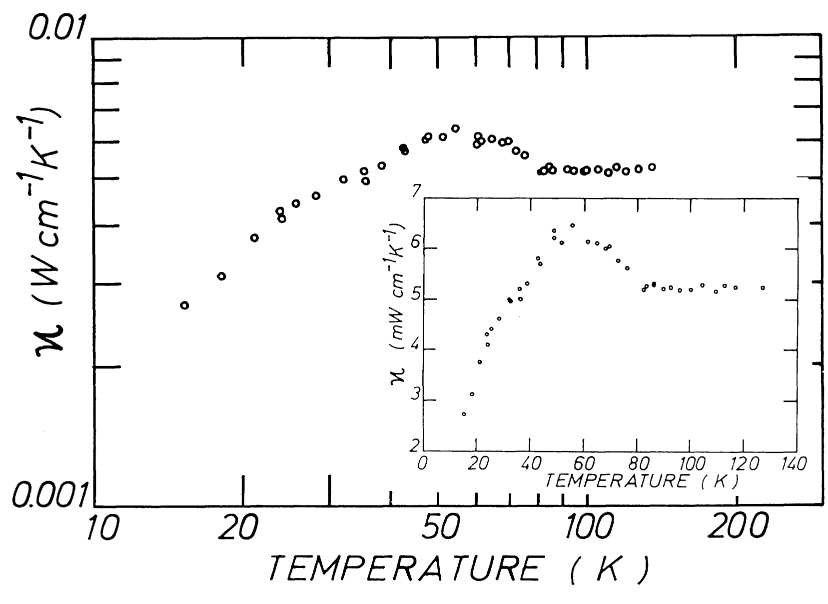

FIG. 2. Thermal conductivity of $YBa_2Cu_3O_7$ vs temperature. The inset shows the data replotted on a linear scale.

Since in our samples κ_c is almost negligible, this effect cannot be seen in our data. On the other hand, electrons which are bound into Cooper pairs can no longer scatter phonons. Thus, if the lattice thermal conductivity is being limited mainly by carrier scattering, one might expect an *enhancement* of κ_p as the temperature is lowered below T_c. This is precisely what we observe in Fig. 2. Below the transition temperature, the phonon mean free path increases as more and more carriers are condensed into the superconducting state. Finally, other scattering mechanisms (i.e., point defect and boundary) come into play which finally bring about a diminution in κ at lower temperatures.

Further evidence that the heat-carrying phonons are interacting strongly with carriers above the transition temperature is given by the nature of κ above T_c. When electrons are the dominant scatterers of phonons, Ziman[7] has shown that

$$\rho_1 \kappa_p = k_B^2 T/(n_a^2 e) ,$$

where ρ_1 is the electrical resistivity due to phonon scattering, n_a the number of free electrons per atom, and k_B the Boltzmann constant. Assuming that the electrical resistivity above T_c is due totally to phonon scattering, we find from this formula that there are about 0.13 free carriers per atom in our sample.

In summary, we have performed measurements of the thermal conductivity of superconducting $YBa_2Cu_3O_7$ from 15 K up to 140 K. Virtually all of the heat conduction in this system is via phonons. The nearly temperature-independent thermal conductivity above the transition temperature together with the sharp rise in κ below T_c both indicate a strong lattice-carrier interaction in the normal state. We therefore conclude that any theory addressing the superconducting properties of this compound must take into account this strong electron-phonon coupling.

Note added. After we had written this manuscript, we received a copy of unpublished work done by V. Bayot, F. Delannay, C. Dewitte, J-P. Erauw, X. Gonze, J-P. Issi, A. Jonas, M. Kinany-Alaoui, M. Lambricht, J-P. Michenaud, J-P. Minet, and L. Piraux. They report the thermal conductivity of a similar sample, and their results are in close agreement with ours.

ACKNOWLEDGMENTS

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