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Thermal conductivity of $YBa_2Cu_3O_7$ from 7 K to 260 K

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Abstract. Thermal conductivity of $YBa_2Cu_3O_7$ has been measured on samples of a few mm thickness. Above T_c thermal conductivity is found to decrease with increase in temperature, pointing towards a contribution to thermal resistivity from three-phonon Umklapp processes. Below T_c thermal conductivity increases rapidly before reaching a maximum at about 50 K and then falls towards zero at lower temperatures. The experimental set up is described and results discussed.

Keywords. Thermal conductivity; YBCO; three-phonon processes.

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1. Introduction

Studies on thermal conductivity of high temperature superconductors are important both from the basic and applied research points of view. The thermal conductivity will determine how rapidly heat will be removed from a local hot spot in the material and this is important in device applications. A study of the temperature variation of thermal conductivity in high T_c superconductors can provide information on the strength of the electron-phonon interaction and the dominant phonon scattering mechanisms in different ranges of temperature.

It is true that very detailed information on the anisotropy in thermal conductivity and its temperature variation can only come from a study of single crystals. However the difficulty in growing large single crystals has severely restricted such studies. The bulk of the measurements has been made on sintered polycrystalline samples. Uher (1990) and Nunez Regueiro and Dario Castello (preprint), among others, have reviewed the work.

In all the studies on superconducting samples of YBCO the thermal conductivity starts increasing as the sample is cooled below T_c. It reaches a maximum value around $T_c/2$ and decreases on further cooling. From the measured electrical resistivity in the normal state, one can estimate the electronic contribution to thermal conductivity by using the Wiedemann–Franz relation. It turns out that this contribution is less than 10% of the total thermal conductivity. So the phonon contribution is dominant.

In the normal state phonons are scattered by defects, electrons and phonons. If we neglect three-phonon Umklapp scattering, then defect and electron-phonon scattering lead to a slow increase in thermal conductivity with increase in temperature above T_c in the theoretical model of Tewordt and Wolkhausen (1989). Below T_c the charge carriers pair up to from the condensate and an energy gap $2\Delta(T)$ opens up in the quasiparticle excitation spectrum. Phonons are not scattered by the condensate but by the quasiparticle excitations which decrease rapidly in number as the temperature is reduced below T_c. Consequently the life time of the phonon due to electron-phonon scattering increases and its contribution to thermal conductivity increases. However, on further cooling the mean free path is limited by defect scattering and so the thermal conductivity drops after passing through a maximum. Tewordt and Wolkhausen have fitted the existing data on a model in which three-phonon U-processes are neglected.

Experimental measurements have been carried out in many laboratories around the world using steady state axial flow heat conduction in rod shaped samples as shown in figure 1a. This technique is well suited for samples of high thermal conductivity and low emissivity. The high temperature superconductors have a rather low thermal conductivity, roughly the same as that of stainless steel, and the thermal conductivity drops rapidly at low temperatures. The samples have a matted black surface which results in a large radiation loss. Above 150 K these radiation losses become large and the thermal conductivity values are not reliable. It is not certain from the measurements whether the thermal conductivity increases or decreases as the temperature is increased above 150 K. One careful measurement by Merisov *et al* (1989) in which care was taken to match the temperature gradient along the sample rod exactly with the gradient along the guard indicated that the conductivity decreased as the temperature was increased to 300 K. This suggests that three-phonon processes are effective in addition to defect scattering at room temperature.

Since the thermal conductivity of these materials is low, the steady state plate method shown in figure 1b is advantageous. Since the lateral surface area over which radiation occurs is a fraction of the cross-sectional area over which heat is conducted, the error arising from radiation loss is enormously reduced. However, the measured temperature drop ΔT between the hot and cold plate is

$$\Delta T = \Delta T_s + \Delta T_{I1} + \Delta T_{I2}$$

where ΔT_s is the actual temperature drop across the sample ΔT_{I1} and ΔT_{I2} are the temperature drops across the interfaces between the hot (cold) plate and the top (bottom) (top) surfaces of the sample. For a reasonable accuracy of measurement ($\cong 5\%$) the $\Delta T_{I1} + \Delta T_{I2}$ should be a small fraction of ΔT_s. This can be achieved by using a suitable high conducting interface layer between the sample and the hot and the cold plates.

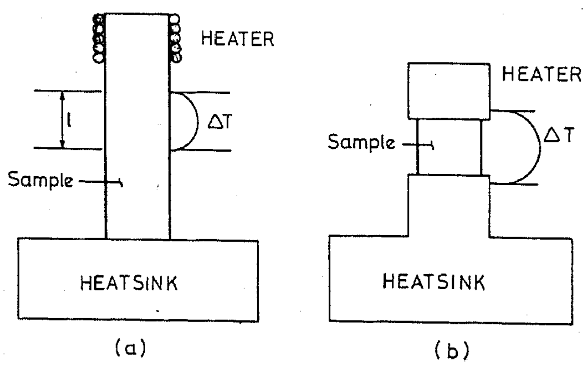

Figure 1. Schematic diagram of (**a**) Rod method and (**b**) Plate method of measuring thermal conductivity.

We have been able to achieve a sufficiently low thermal contact resistance by amalgamating the top (heat sink) and bottom (heater) plates of copper and squeezing the sample between the plates so that a thin layer of mercury spreads on the interface. The thermal conductivity of a sample of a single crystal of potassium dihydrogen phosphate cut perpendicular to the tetragonal axis was measured with the set up. A sample of YBCO was then studied. The experimental set up used for the study from 7 K to 260 K and the results obtained are described and discussed in this paper.

2. Experimental set-up

The sample holder is shown in figure 2. The cold plate C is made of copper and is suspended from the top copper flange F by three thin stainless steel rods (D). Liquid helium or nitrogen in the dewar stands several centimetres above F. A heater is wound around C. In the absence of a current through the heater, the plate C cools to about 7 K through conduction down the SS rods. The cold plate can be maintained at any temperature above the boiling point of the cryogen by passing a current through the heater. The diameter and length of the SS rods are so chosen that the cold plate can be maintained at a temperature much above the boiling point of the cryogen by giving a reasonably small heat input to the heater. For example, with liquid helium, a heat input of 0·25 W suffices to maintain the temperature of C at 80 K. The sample S in the form of

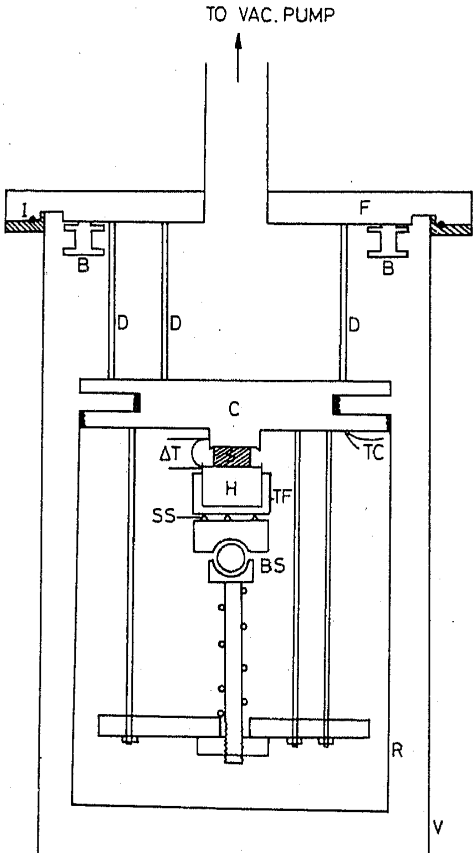

Figure 2. Schematic diagram of sample holder.

a disc or a plate of cross sectional area of approximately 60 mm^2 and thickness 2 to 4 mm is sandwiched between the cold plate C and the hot plate H. The hot plate is a copper cup containing a 120 ohm strain gauge heater stuck with GE varnish to the inner bottom of the heater cup. The heater cup is embedded in a teflon block TF and is pressed to the sample by three pointed SS tips on a spring-loaded ball and socket joint BS as shown in the figure. In order to improve the thermal contact between the hot and cold plates and the sample surfaces, the contact surfaces of these plates are amalgamated and a drop of mercury placed on them before they are pressed to the sample, as mentioned before. The temperature of the cold block is measured with a gold (0·07 at% Fe)-chromel thermocouple TC fixed to the cold plate with delta bond. The other junction of the thermocouple is fixed to the top flange F whose temperature is measured by a carbon resistance thermometer calibrated against a gold (0·07 at% Fe)-chromel thermocouple. The temperature difference between the hot and cold plates is determined by a differential thermocouple ΔT of gold (0·07 at.% Fe)-chromel again fixed by delta bond to the two plates. A platinum and a carbon resistance thermometer fixed to the heat sink by delta bond are used as temperature sensors to control the temperature of the heat sink through an intelligent temperature controller ITC4 supplied by Oxford Instruments. All the electrical leads are thermally anchored by winding them on two copper bobbins B fixed on the top flange F. A radiation shield R fixed to the heat sink surrounds the sample assembly. The whole arrangement is enclosed in a copper vacuum can V fixed to the top flange by screws and carrying an indium seal I. The entire assembly is suspended by a 70 cm long thin walled SS tube from the top flange of the helium dewar. Electrical leads are taken through this tube to feed throughs at the top of the cryostat. This tube also serves to evacuate the vacuum can V.

The current to the strain gauge is obtained from a programmable constant current source supplied by Advantest, Japan. The potential drop across the heater and the emf of the thermocouple measuring absolute temperature are measured by a solartron computing voltmeter. The ΔT thermo emf is measured by a Keithley model 181 nanovoltmeter. All the measuring instruments are interfaced to a microcomputer with IEEE 488 GPIB and RS 232.

To measure the thermal conductivity at any temperature the desired temperature of the heat sink is set on the temperature controller and the temperature T of the heat sink is measured at successive stated intervals of time. When the rate of change of temperature dT/dt is less than 1×10^{-5} K/s, the ΔT is similarly measured and when $d(\Delta T)/dt$ is also less than 1×10^{-5} K/s its initial value is noted down. Then a current is sent through the heater in the heater cup and the temperature ΔT and the heater current and voltage are measured at successive intervals of time till ΔT reaches a steady value with $d(\Delta T)/dt$ less than 1×10^{-5} K/s. Then the heater current is changed and at the same temperature T of the heat sink, ΔT is determined for three such heater powers $\dot{Q}$ such that the maximum $\Delta T \cong T/100$. The computer then calculates the slope of the heater power $\dot{Q}$ vs ΔT and computes the thermal conductivity using the gives area and thickness of the sample. The heat sink temperature is then changed and the experiment repeated at the new temperature. A suitable computer program was written in GWBASIC to perform the sequence of measurements.

Initially a similar set up was used with a continuous flow cryostat. It was verified that two samples of stainless steel of different thicknesses gave values of thermal conductivity which agree within 5% over a range of temperature from 300 K to 90 K. This indicates that the temperature drop across the mercury film does not cause an error larger than this value. The error due to the temperature drop across the film will become less important at low temperatures at which the thermal conductivity of high T_c superconductors drops below the value at room temperature.

In order to verify that this is true, the thermal conductivity of a KH_2PO_4 single crystal cut with the normal along the tetragonal axis was measured with the set up described here. The measured thermal conductivity is shown in figure 3.

The thermal conductivity values are in agreement with the values reported by Sievers and Pohl (1964). At about 120 K the slope of the thermal conductivity curve changes drastically indicating a transition to ferroelectric state below this temperature.

The pellets of $YBa_2Cu_3O_7$ used in the present measurements were prepared by the conventional solid state reaction technique described by Subba Rao *et al* (1987). The measured densities of the pellets are 78% of the ideal density for the pellet A and 84% for the pellet B. The thermal conductivity of the pellet A measured using the present set up is shown in figure 4. The thermal conductivity of the pellet B was measured with a similar set up but mounted on a continuous flow cryostat in which the lowest temperature attained was 35K. In the same figure the results of Merisov *et al* (1989) (mentioned earlier) obtained with the use of a radiation shield to minimize the heat loss by radiation is also shown for comparison. In contrast to the measurements of Uher and Kaiser (1987), Gottwick *et al* (1987) and others which show a thermal conductivity increasing with temperature above T_c, the present results show an opposite behaviour. One should expect the trend observed in the present results if the three-phonon scattering processes play an important role. It is also seen that at T_c there is an abrupt change of slope in the K vs T curve. This feature has also been observed by several investigators and has been attributed to the electron-phonon interaction which decreases abruptly as the material becomes superconducting.

The results of Uher and others have been theoretically interpretted in terms of the

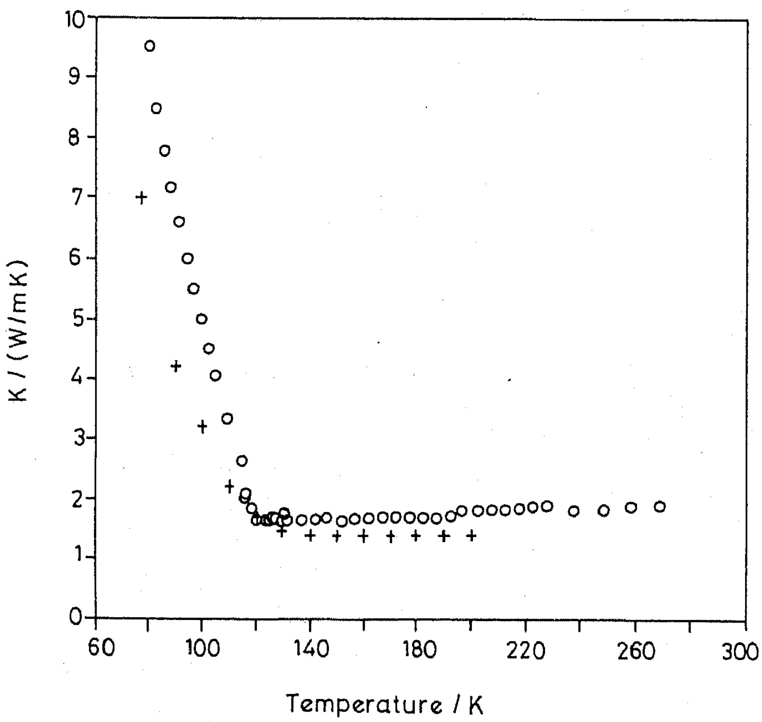

Figure 3. K vs T graph of KDP crystal: circles-present data, crosses-Sievers and Pohl (1964).

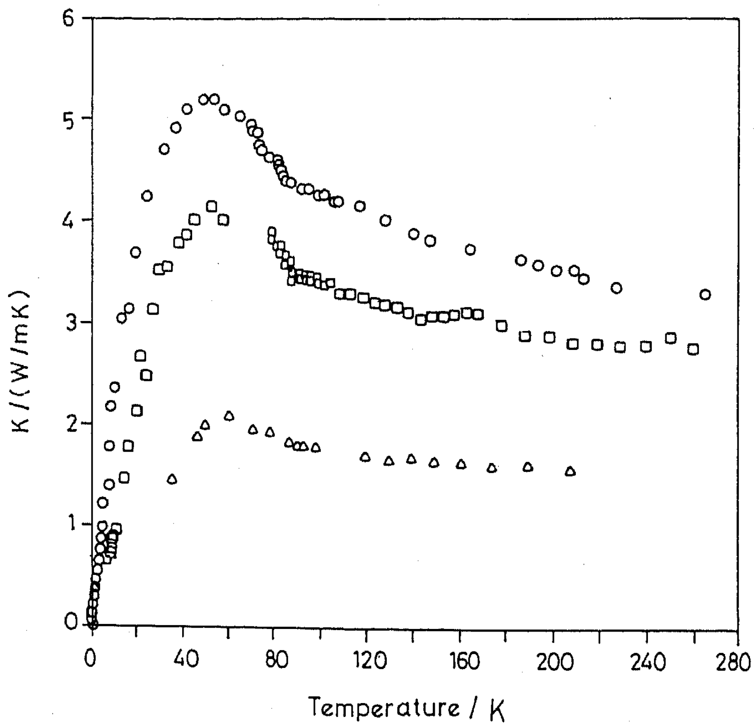

Figure 4. K vs T graph of YBCO samples: squares-sample A, triangle-sample B, circles-Cohn *et al.* (preprint).

Tewordt and Wolkhausen theory. In this theory they have neglected the three-phonon Umklapp processes and taken into account point defect scattering, scattering at two-dimensional defects, line defect scattering, boundary scattering and electron-phonon scattering. In this theory the $K(T)$ is given by

$$K = At^3 \int_0^\infty \frac{x^4 e^x}{(e^x - 1)^2} F(t, x)\, dx$$

where

$$F(t, x) = [1 + \alpha t^4 x^4 + \beta t^2 x^2 + \delta t x + \gamma t x g(x, y)]^{-1}.$$

Here A, α, β, δ and γ refer to the scattering strengths due to the boundary scattering, point-defect scattering, sheet like faults, dislocation scattering, and electron-phonon interaction. $x = \hbar\omega/kT$, $t = T/T_c$ and $g(x, y)$ is the BRT function defined by Bardeen *et al* (1959). Because of the non-inclusion of the Umklapp processes this expression will always give a $K(T)$ which is an increasing function of T at high temperatures. Since in the present studies we see evidence for U processes in the decreasing $K(T)$ with increase in temperature above T_c, this expression has to be modified. Cohn *et al* (preprint) have measured the lattice thermal conductivity of a single crystal of 123 in the ab plane and liquid phase processed specimens of 123 in the temperature range 10K to 300K. They also find the trend similar to the one reported in the present study and they have tried to include the three-phonon U-processes by adding a term $\varepsilon\, x^2 t^4$ to the terms in the brackets in the $F(x, t)$. We have therefore fitted our data using the modified expression

$$F(x, t) = [1 + \alpha x^4 t^4 + \beta x^2 t^2 + \gamma x t g(x, y) + \varepsilon x^2 t^4]^{-1}.$$

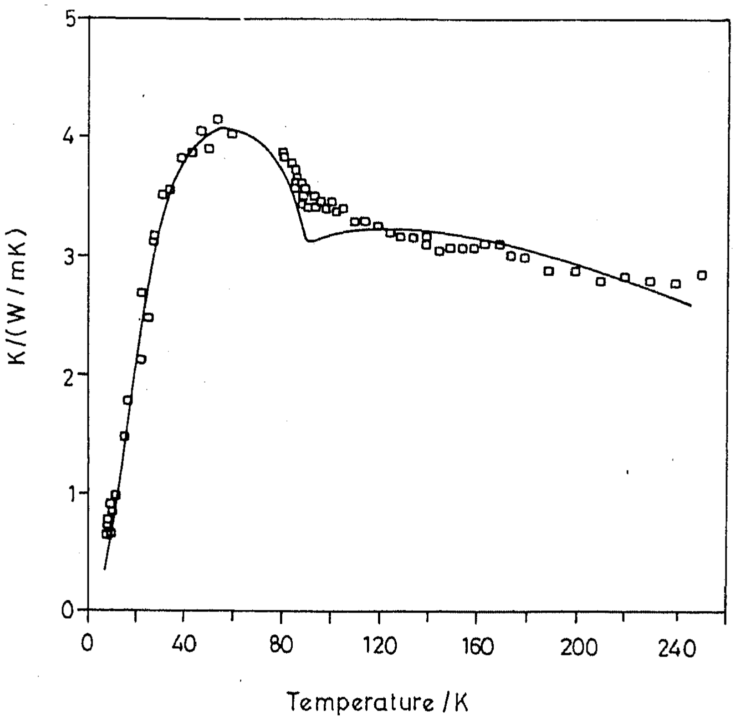

Figure 5. K vs T of 123, sample A, points-experimental, continuous line-theory.

Table 1. Parameters used to fit $K(T)$ of 123.

Ref.	A (W/cmK)	α	β	γ	ε
Uher and Kaiser (1987)	3·5	50	60	100	0
Gottwick *et al* (1987)	5·0	60	35	100	0
Present	2·0	20	35	50	30

The results of such a fit are shown as continuous curves in figure 5. The parameters in the fit are collected in table 1. In the same table the parameters of Uher and Kaiser, Gottwick *et al* are given.

It is seen from the table that the parameter values are comparable. It is especially noteworthy that the phonon–phonon interaction parameter ε is comparable to α, β and γ. The trend of $K(T)$ increasing with T above T_c which is seen in the results of Uher and Kaiser and Gottwick *et al* may be due to errors arising from radiation losses. The theoretical curve shown in figure 5 shows a very weak maximum around 130 K. The experimental results also show a weak maximum around 160 K. Such a weak maximum seen in earlier work was attributed to some structural change around this temperature (e.g He Yusheng *et al* 1987). However, it appears that this peak can be explained as due to the interplay of phonon-phonon scattering and other scattering mechanisms. The former will cause the $K(T)$ to decrease with increasing temperature while the latter will cause it to increase with T.

In conclusion, using the plate method which reduces radiation losses at high temperatures, we consistently see a $K(T)$ decreasing with increasing temperature at high temperatures. This highlights the importance of three-phonon processes. A theoretical fit to the $K(T)$ curve including the three phonon processes shows that the scattering due to three-phonon processes is comparable to the other scattering processes.

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