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LOW-TEMPERATURE THERMAL PROPERTIES OF La_2CuO_4 AND SUPERCONDUCTING $Ba_2YCu_3O_{7-x}$

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The thermal conductivity λ and the specific heat c_p of $Ba_2YCu_3O_{7-x}$ and La_2CuO_4 have been measured in the temperature range $0.03 < T < 30$ K. The thermal conductivity of sintered $Ba_2YCu_3O_{7-x}$ samples varies as T^2 between 0.7 and 8 K, while below 0.3 K $\lambda(T)$ is described as a sum of two terms where one is proportional to T and the other varies like T^3. For sintered La_2CuO_4, $\lambda(T)$ is distinctly smaller than in sintered $Ba_2YCu_3O_{7-x}$ in the whole temperature range. The specific heat of both compounds shows a non-vanishing linear term γT between 1.4 K and 15 K, where γ in sintered $Ba_2YCu_3O_{7-x}$ is nearly an order of magnitude larger than in crystalline La_2CuO_4.

1. INTRODUCTION

After the discovery (1) of high-T_c superconductivity in oxide compounds it was at once discussed what characteristics of the superconducting state might be anomalous. It was soon realized that in superconducting $Ba_2YCu_3O_{7-x}$ a linear-in-T contribution to c_p is observed at temperatures well below T_c (2), an uncommon feature in conventional superconductors.

In order to contribute to clarifying the situation we made a study of thermal properties of various different samples of $Ba_2YCu_3O_{7-x}$ and, for comparison, of insulating La_2CuO_4.

2. EXPERIMENTAL

In order to study the effect of the mass density and of the average grainsize on the thermal conductivity and the specific heat of sintered $Ba_2YCu_3O_{7-x}$ three different samples have been investigated. Sample 1 had a low mass-density (ρ = 75-80% of ρ_{theor}) and an average grainsize of 10 μm, samples 2 and 3 had a large mass density and average grainsizes of 150 and 20 μm, respectively. The thermal conductivity of La_2CuO_4 has been measured on a sintered sample, but the specific heat was obtained for a crystalline sample.

3. RESULTS

In Fig. 1 we show the results of the thermal conductivity experiments above 1.4 K by plotting log λ versus log T.

Above 4 K we may fit our data for La_2CuO_4 with $\lambda(T) = A_2T^2$, where $A_2 = 1.2 \times 10^{-2}$ W/mK^3. At temperatures below 0.3 K a two parameters fit $\lambda(T) = A_1T + A_3T^3$ yields the following values for A_i: $A_1 = 4.5 \times 10^{-5}$ W/mK^2 and $A_3 = 8.1 \times 10^{-3}$ W/mK^4. For $BaYCu_3O_{7-x}$, $\lambda(T) = A_2T^2$, with $A_2 = 2.24 \times 10^{-2}$ W/mK^3 for sample 1, $A_2 = 2.53 \times 10^{-2}$ W/mK^3 for sample 2, and

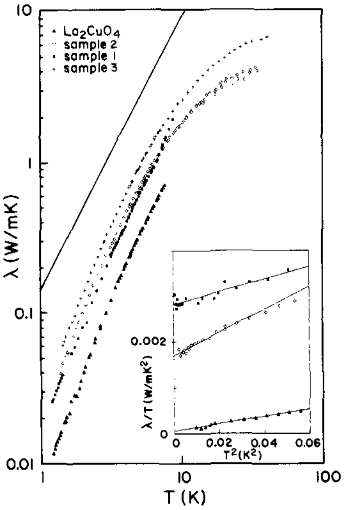

FIGURE 1
$\lambda(T)$ of sintered $Ba_2YCu_3O_{7-x}$ and La_2CuO_4 above 1 K. The solid line is the thermal conductivity of single-crystalline $Ba_2YCu_3O_{7-x}$ as given in ref. 3. The inset shows the behaviour of λ at the lowest temperatures.

$A_2 = 3.38 \times 10^{-2}$ W/mK^3 for sample 3 between 0.7 and 8 K.

We compare our results on sintered specimens of $Ba_2YCu_3O_{7-x}$ with previously published data (3) on a single crystalline sample of $Ba_2YCu_3O_{7-x}$. As it may be seen in Fig. 1, the thermal conductivity for a single-crystal in the a-b plane is nearly an order of magnitude larger than for our sintered samples.

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This big difference cannot be attributed to the enhanced scattering of the phonons by grain-boundaries of sintered samples alone. The thermal conductivity for the sample with large grains (150 μm) is lower than for the sample with small grains (20 μm). Above 1.4 K the thermal conductivity increases with increasing filling factor. A possible reason for the difference between sintered and crystalline material is an anisotropic thermal conductivity where λ is much larger in the a-b plane than perpendicular to it. For $T < 0.3$ K, we may fit our data by $\lambda(T) = A_1T+A_3T^3$ with $A_1 = 2.8 \times 10^{-3}$ W/mK^2 and $A_3 = 1.37 \times 10^{-2}$ W/mK^4 for sample 1 and $A_1 = 1.7 \times 10^{-3}$ W/mK^2 and $A_3 = 2.53 \times 10^{-2}$ W/mK^4 for sample 2. The prefactor A_3 increases with increasing grainsize and filling factor, while A_1 decreases. The mean free path related to boundary scattering in sample 2 is about 15 times larger than for sample 1. The prefactor A_3 is, however, only two times larger than for sample 1 because the cut-off frequency for sample 2 is much smaller than for sample 1.

The non vanishing linear terms in the specific heat of crystalline La_2CuO_4 and sintered $Ba_2YCu_3O_{7-x}$ (see below) suggest the existence of two-level tunneling states as was pointed out by Graebner and co-workers (3).

A non vanishing linear-in-T contribution to the specific heat $c_p(T)$ as $T \ll T_c$ may also be an indication for unconventional superconductivity. The resonating-valence-bond model (4) for superconductivity in these oxides, e.g., predicts a γT contribution to the specific-heat in the superconducting state.

The inset of Fig. 2 shows the result of our specific-heat measurements above 1.5 K. c_p of $Ba_2YCu_3O_{7-x}$ (sample 1) is described by a two parameter fit $c_p(T) = \gamma T + \beta T^3$, where $\gamma = 10^{-3}$ J/mole K^2 and $\beta = 5.89 \times 10^{-4}$ J/mole K^4. A three parameter fit $c_p(T) = \gamma T+\beta T^3 + \delta T^5$ with $\gamma = 1.3 \times 10^{-3}$ J/mole K^2, $\beta = 1.74 \times 10^{-4}$ J/mole K^4 and $\delta = 9.80 \times 10^{-7}$ J/mole K^6 is adequate for crystalline La_2CuO_4.

As shown in Fig. 2, the specific heat of both compounds increases with further decreasing temperature. In order to investigate this contribution to $c_p(T)$ we subtract from the low-temperature data the extrapolated fit of the data above 1 K. The resulting low-temperature contribution to c_p shows for both compounds a T^{-2} term, but the prefactor is much larger for La_2CuO_4 than for $Ba_2YCu_3O_{7-x}$. We ascribe this T^{-2} term to a nuclear specific heat c_N. It is well known that at high temperatures c_N can be expanded in a power series of 1/T. Assuming that the Cu nuclei (^{63}Cu and ^{65}Cu both have spin quantum number 3/2) are responsible for c_N, we obtain by omitting a term of higher order in the prefactors of the T^{-4} contribution:

$$c_N/R = (5/4a^2+p^2)T^{-2}-3a^2PT^{-3}-17/16a^4T^{-4}$$

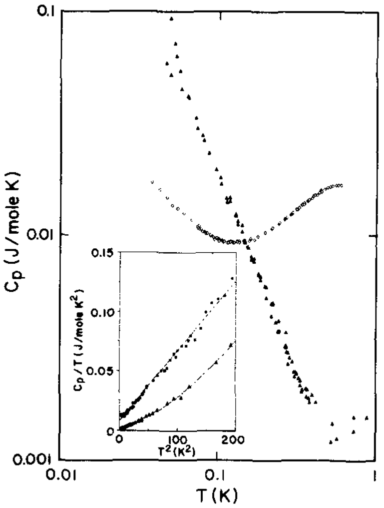

FIGURE 2
Specific heat of sintered $Ba_2YCu_3O_{7-x}$ and crystalline La_2CuO_4 below 1 K. The inset shows the specific heat above 1 K. Symbols for corresponding samples are the same as in Fig. 1.

where $a = 2/3\ \mu\ H_{eff}/k_B$ and $P = e^2Qq/4k_B$. Q is the nuclear quadrupolemoment, q the electric field gradient at the nucleus. H_{eff} is the effective magnetic field at the nucleus and μ is the nuclear magnetic moment.

For La_2CuO_4 we obtain $c_N(T) = 2.096 \times 10^{-4}T^{-2} - 9.05 \times 10^{-8}T^{-4}$ J/mole K. Since the T^{-3}-term is missing, we conclude that the observed nuclear specific heat in La_2CuO_4 is due to an effective magnetic field H_{eff}, as is expected from high-temperature antiferromagnetic order. From the prefactor of the T^{-2}-term we estimate $a = 4.49\ 10^{-3}$ K or $H_{eff} = 54$ kOe.

For $Ba_2YCu_3O_{7-x}$ $c_N(T) = 4.1 \times 10^{-5}T^{-2} - 9.3 \times 10^{-7}T^{-3}$ J/mole K. The need of a T^{-3}-term to fit our data suggests that the expected quadrupole interaction is responsible for the nuclear specific heat of orthorhombic superconducting $Ba_2YCu_3O_{7-x}$. Because of space limitations, we refrain from analysing the anomalous specific heat at intermediate temperatures above 0.2 K in $Ba_2YCu_3O_{7-x}$.

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