

# Thermal conductivity of the superconducting crystals $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+y}$ in the $ab$ plane

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The results of measurements of the thermal conductivity  $\kappa$  and the resistivity  $\rho$  of three different high- $T_c$   $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+y}$  crystals in the  $ab$  plane in the temperature range 10–140 K are presented. It was found that 1) the maximum thermal conductivity of the crystals is several times higher than that of previously studied samples; 2) in contrast to the computational results of the theory of electronic heat transport in anisotropic high- $T_c$  cuprate crystals, the maximum values of the ratio  $\kappa(T)/\kappa(T_c)$  in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+y}$  crystals can reach 2, and just as in the less anisotropic  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  crystals, as this ratio increases, the maximum point of the thermal conductivity shifts in the direction of lower temperatures  $T_m/T_c \leq 0.4$ ; and 3) the maxima on the curves of  $\kappa(T)$  are observed in samples with low resistivity  $\rho$  and metallic-type conductivity  $\rho(T) \sim T$ , in which the electronic component  $\kappa_e$  is comparable in order of magnitude to the total thermal conductivity  $\kappa$  at temperatures  $T \geq T_c$ , and in samples whose resistivity increases with decreasing temperature and therefore  $\kappa_e \ll \kappa$ . This shows that to explain the appearance of the maxima on the curves  $\kappa(T)$  for high- $T_c$  cuprate crystals in the superconducting state, it is necessary to take into account the increase in not only the electronic component of the thermal conductivity  $\kappa_e$ , as is standard in most modern theoretical calculations, but also in the phonon component  $\kappa_p$  below  $T_c$ . © 1995 American Institute of Physics.

1. It is well known<sup>1</sup> that the behavior of the thermal conductivity  $\kappa$  of a superconductor is determined by the sum of the contributions of the electronic  $\kappa_e$  and lattice  $\kappa_p$  components

$$\kappa = \kappa_e + \kappa_p. \quad (1)$$

In the normal state, at temperatures  $T \geq T_c$ ,  $\kappa_e$  can be estimated from the normal-state conductivity  $\rho^{-1}$  of the sample using the Wiedemann–Franz law

$$\kappa_e = LT/\rho, \quad (2)$$

where  $\rho$  is the resistivity. The numerical coefficient is  $L = L_0 = 2.4 \times 10^{-8} \text{ W}/\Omega \cdot \text{K}^2$  (Lorentz's number) if elastic scattering of carriers in the volume predominates and if  $L < L_0$  in the presence of inelastic scattering of the carriers. In contrast to the dc conduc-

tivity  $\rho^{-1}$ ,  $\kappa_e$  remains finite when the sample becomes superconducting. Consequently, the contributions of the electronic and lattice components of the thermal conductivity can, in principle, be separated in the measurements of  $\rho(T)$  and  $\kappa(T)$  in a wide temperature range above and below the critical temperature  $T_c$ , and hence an attempt can be made to infer from them the characteristic features of the interaction between quasiparticles and the role of different mechanisms for relaxation of excitations in the electronic and phonon systems. This approach has been found to be useful in discussing the transport coefficients of standard metallic superconductors whose properties are described well by the standard BCS model with weak or strong electron-phonon interaction.<sup>1-3</sup>

It would be especially interesting to clarify the behavior of  $\kappa_e(T)$  and  $\kappa_p(T)$  in high- $T_c$  superconductors. In the case of layered high- $T_c$  crystals, however, it has still not been unequivocally determined which component determines the behavior of the thermal conductivity below  $T_c$ . In particular, what causes the appearance of the maxima on the  $\kappa(T)$  curves which describe the temperature dependence of the thermal conductivity of crystals in the cuprate  $ab$  plane? For example, in Refs. 4 and 5 it is posited that below  $T_c$  the phonon component plays the main role and the increase in thermal conductivity below  $T_c$  is due to the decrease in the concentration of normal electronic excitations which limit the maximum phonon path lengths near  $T_c$ . Conversely, in most experimental and theoretical studies published after the experimental observation of the manyfold increase in the high-frequency conductivity  $\sigma_{HF}$  of a gas of normal electronic excitations below  $T_c$  (for example, in Refs. 6–8), the increase in  $\kappa(T)$  in the superconducting state is attributed to the manyfold increase in the thermal conductivity of the electronic component  $\kappa_e = \sigma_{HF} L_0 T$ , and the changes in  $\kappa_p$  are ignored. It is clear that to determine the suitability of different theoretical models, further experimental studies of the properties of more perfect high- $T_c$  crystals are in order.

2. In this letter we report the results of an experimental study of the thermal conductivity of superconducting  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+y}$  crystals in the cuprate  $ab$  plane.

A detailed analysis of previously published results of theoretical and experimental studies of the thermal conductivity of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+y}$  crystals in the normal and superconducting states and a list of publications are contained in Refs. 8 and 9. Comparing the measurements of high- $T_c$  crystal  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  and its nonconducting analog  $\text{Bi}_2\text{Sr}_2\text{YCu}_2\text{O}_8$ , the authors of Ref. 9 concluded that (first) the peak in the temperature dependence of the thermal conductivity  $\kappa(T)$  of the superconductor in the  $ab$  plane is due to the manyfold increase of the electronic component  $\kappa_e$  of the thermal conductivity below  $T_c$  and (second) heat is transported in the lattice of the  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  crystal by incoherently vibrating atoms (Einstein's model of the thermal conductivity) and not by traveling phonon waves (Peierls model), as in ordinary crystals.

Subsequent variational calculations of the electronic component of the thermal conductivity  $\kappa_e$  of high- $T_c$  crystals<sup>8</sup> showed that for the same concentration of ionized impurities in the bulk  $N=0.05-0.1$  the maximum values of the ratio  $\kappa_e(T)/\kappa_e(T_c)$  in the  $ab$  plane in the more strongly anisotropic  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  crystals  $\kappa_{e,\max}(T)/\kappa_e(T_c)=1.5-2.5$  should be much smaller than in the less anisotropic  $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$  crystals  $\kappa_{e,\max}(T)/\kappa_e(T_c)=5-7$ , and the position of the maximum point  $T_{\max}$  should shift in the direction of higher temperatures from  $T_{\max}=0.4T_c$  up to  $T_{\max}=(0.6-0.7)T_c$ . In Refs. 8 and 9 it is assumed that the temperature dependence of

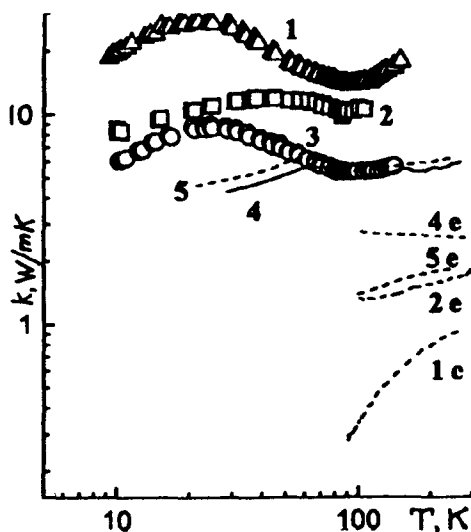


FIG. 1. Temperature dependences of the thermal conductivity of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$  crystals in the  $ab$  plane. Points 1, 2, and 3 are our measurements. Curves 4 and 5 were constructed on the basis of the data of Refs. 9 and 11. Dashed curves 1e–5e represent the estimates of the contribution of the electronic system  $\kappa_e$  for the same samples.

$\kappa_p(T)$  can be ignored in a wide temperature range above and below  $T_c$ . In other words,

$$\kappa(T) = \kappa_e(T) + \kappa_p. \quad (3)$$

Taking into account the contribution of the lattice component in the reduced coordinates  $K(t) = \kappa(T)/\kappa(T_c)$ , where  $t = T/T_c$  is the reduced temperature, the computed values of the peaks on the curves  $K(t)$  in Bi crystals should be much smaller  $K_{\max}(t) \approx 1.3$ , and they are shifted in the direction of higher temperatures  $t_{\max} \sim 0.6-0.7$  compared to the Y crystals with the same degree of perfection ( $K_{\max}(t) \approx 2$ ,  $t_{\max} \sim 0.4$ ).

As will be seen below (Figs. 1 and 2), the predictions of the theory in Ref. 8 do not agree with the results of our measurements: As the maximum thermal conductivity of Bi crystals increases, the ratio  $K(t)$  approaches 2 and the position of the maximum shifts in the direction of lower temperatures  $t < 0.4$ , as in the best of the known  $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$  crystals.<sup>6,10</sup>

3. The main parameters characterizing the properties of the experimental samples 1, 2, and 3 are given in Table I. The minimum dimensions correspond to the direction along the  $c$  axis.

We estimated the values of the resistivity  $\rho(120 \text{ K})$  and the thermal conductivity  $\kappa(120 \text{ K})$  for crystals 4 and 5, which were investigated in Refs. 9 and 11, from the plots of  $\rho(T)$  and  $\kappa(T)$  shown in Figs. 2a and 2b in Ref. 9 and Figs. 1a and 1b in Ref. 11.

Our method of measuring the thermal conductivity and resistivity is described in detail in Ref. 12. The measurements of  $\rho(T)$  were performed by a standard potentiometric method using dc current and  $\kappa(T)$  was measured by the stationary heat flux

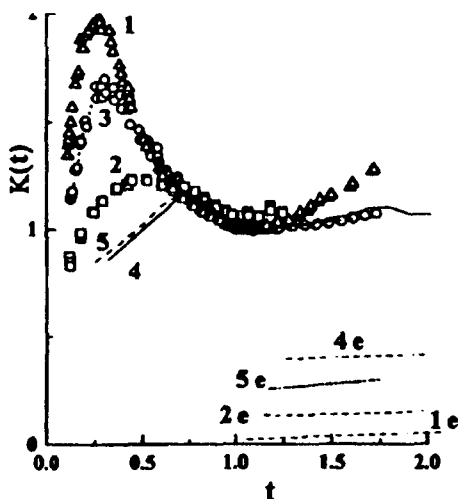


FIG. 2. Temperature dependences of the thermal conductivity of crystals 1-5 in reduced coordinates:  $K(t) = \kappa(T)/\kappa(T_c)$ ;  $t = T/T_c$ .

method. To decrease the error in estimating the ratios of  $\kappa_p$  and  $\kappa_e$  in an anisotropic crystal, the heat flux in the thermal conductivity measurements and the electric current in the resistivity measurements in the  $ab$  plane of the sample were directed in the same direction.

The temperature dependences of the thermal conductivity  $\kappa(T)$  of different Bi crystals in the  $ab$  plane are shown in Fig. 1 (points 1, 2, and 3). The experimental temperature dependences  $\kappa(T)$  are supplemented by the computed dependences  $\kappa_e(T)$ . The dashed curves 1e and 2e in Fig. 1 were constructed from the measurements of the resistivity  $\rho(T)$  of the same samples. Curves 4 and 4e describe the thermal conductivity  $\kappa(T)$  and  $\kappa_e(T)$  of the sample investigated in Ref. 9 (they were constructed from the plots 2a and 2b), and curves 5 and 5e describe the thermal conductivity and the contribution, calculated from relation (2), of the electronic component of the thermal conductivity for crystal 5 of Ref. 11 (plots 1a and 1b). We recall that the curves  $\kappa_e(T)$  correspond to upper estimates, disregarding the contribution of inelastic scattering of the charge carriers.

TABLE I.

Sample (numbers on the curves in the figures)	Dimensions, mm <sup>3</sup>	$T_c$ , K	$\Delta T_c$ , K	$\kappa$ (120 K) W/mK	$\rho$ (120 K) m $\Omega$ ·cm	$\kappa_e$ (120 K) W/mK
1	0.96×0.58×0.016	89	3.5	15	0.63	0.47
2	2.00×0.59×0.050	86	1.5	10.5	0.22	1.3
3	2.30×1.20×0.020	82	4	5.4	—	—
4	—	—	—	~5.1	~0.11	~2.6
5	2.1×0.01	89	—	~5.5	~0.18	~1.6

Figure 2 shows the temperature dependences of the thermal conductivity of the same samples in the reduced coordinates  $K(t) = \kappa(T)/\kappa(T_c)$ , where  $t = T/T_c$  is the reduced temperature. These coordinates are ordinarily used in theoretical calculations. They make it possible to eliminate the unavoidable errors in determining the absolute values of the transport coefficients  $\kappa$  and  $\rho$  for small samples and also the errors in calculating the numerical cofactors in the theoretical calculations of the transport coefficients of the crystals.

4. We see from Figs. 1 and 2 and Table I that in the normal state at  $T \geq T_c$  in crystals 1 and 2  $\kappa_e \leq 0.1\kappa$ ; i.e., the lattice component of the thermal conductivity predominates,  $\kappa_p \gg \kappa_e$ . At the same time, in the sample investigated in Ref. 9  $\kappa_e \sim 0.5\kappa$ ; i.e.,  $\kappa_p \approx \kappa_e$ . Moreover, the electric conductivity of samples 4 and 5 is several times higher than that of our samples 1 and 2. Therefore, the lattice component  $\kappa_p$  of these samples is much smaller than  $\kappa_p$  of crystals 1 and 2. In accordance with the calculations in Ref. 8, the increase in the electronic component  $\kappa_e$  which accompanies a transition to the superconducting state, should be strongest in these samples; i.e., in the reduced coordinates  $K(t) = \kappa(T)/\kappa(T_c)$  (Fig. 2) the maxima on curves 4 and 5 should be much higher and should occur at lower reduced temperatures than on curves 1 and 2. To attribute the maximum on curves 1 and 2 to an increase in only the electronic component  $\kappa_e$ , it was necessary to assume that the ratio  $\kappa_e(T)/\kappa_e(T_c)$  at the maximum increases by at least a factor of  $\sim 10$  in crystal 2 and by a factor of  $\sim 100$  in sample 1. This is obviously higher than the theoretical predictions in Ref. 8.

Therefore, it follows from a comparison of the ratios of the values of  $\kappa$  and  $\kappa_e$  in different samples that the appearance of maxima on the curves of  $\kappa(T)$  in the  $ab$  plane in the experimental crystals 1, 2 and 4, 5 cannot be attributed to an increase in only the electronic component  $\kappa_e(T)$  of the thermal conductivity below  $T_c$ , as assumed in Refs. 6–9. The increase in the thermal conductivity of samples 1 and 2, in which the lattice component near  $T_c$  predominates, just as the manifold increase in  $\kappa_p$  in the normal state in samples 1 and 2 as compared to samples 4 and 5, shows that heat is transported in the lattice of these crystals by phonons whose effective path lengths  $\lambda_p(T)$  are limited by scattering by normal electronic excitations, by one another, and by impurities and defects in the bulk of the sample. The lack of a correlation in the values of  $\kappa$  and  $\rho$  can be easily explained, since the path lengths  $\lambda_p$  and  $\lambda_e$  of the phonons and charge carriers, respectively, in different samples with close composition can be limited by different scattering mechanisms. For example, the electron path lengths can be limited by strong scattering by charged point defects, while the phonon path lengths in the case of scattering by the same point defects at low temperatures increase rapidly,  $\lambda_p \sim T^{-4}$  (Rayleigh scattering).

Our estimates of the effective phonon mean free path in the simple gas-kinetic approximation

$$\kappa_p \approx 1/3 C_p \nu \lambda_p \quad (4)$$

on the basis of the experimental values<sup>13</sup> of the thermal conductivity, the heat capacity  $C_p$ , and the sound velocity  $\nu$  show that at  $T \approx T_c$  the quantity  $\lambda_p(T)$  is several times larger than the characteristic lattice parameters in the  $ab$  plane; i.e., the description of the thermal conductivity by Peierls model (by the propagation of traveling waves — phonons) is completely correct. The Einstein model of vibrating oscillators in a solid,

studied in Ref. 9 as the main mechanism of heat transport in the lattice of a Bi crystal, could be applicable for describing the behavior of the thermal conductivity along the  $c$  axis, where according to estimates made in Ref. 13, the effective phonon mean free path reaches values of the order of several lattice constants only when the crystal is cooled below 30 K. It is well known that because of the high concentration of defects between the cuprate planes, the mechanism of charge-carrier motion in Bi crystals changes substantially: from band motion in the  $ab$  plane to hopping diffusion in the  $c$  direction.

The increase in  $\kappa_p(T)$  at a transition to the superconducting state is associated with the freezing out of normal electronic excitations which scatter phonons. We noted above that in Refs. 4 and 5 the computed curves  $\kappa_p(T)/\kappa_p(T_c)$  could be matched within a few percent with the experimental curves on the basis of the BCS model, assuming that for  $t \geq 0.3$  phonon-electron scattering plays the main role and the superconducting gap in the electronic spectrum  $\Delta = \chi \Delta_{\text{BCS}}$ , where the parameter  $\chi$  falls in the range  $\chi = 1.4 - 2$  for different high- $T_c$  samples. In principle, values  $\chi \leq 2$  are also sufficient for describing the behavior of curves 1-5 in Fig. 2. We recall that in metallic superconductors with strong electron-phonon coupling (for example, in lead<sup>3</sup>)  $\chi \leq 1.4$ ; i.e., the possibility of increasing  $\chi$  to 2 in the electron-phonon interaction approximation requires a separate study.

The experimental observation of a manyfold increase of the high-frequency conductivity  $\sigma_{HF}$  of a gas of normal electronic excitations below  $T_c$  in  $\text{YBa}_2\text{Cu}_3\text{O}_7$  crystals and the reasonable agreement between the computational relation  $\kappa_e = \sigma_{HF} \cdot L_0 T$  and the estimates of  $\kappa_e(T)$  according to the measured thermal conductivity in Ref. 6 indicate that the contribution of the electronic component to the thermal conductivity of high- $T_c$  superconducting crystals also cannot be ignored.

The resistance of the samples was not measured in Ref. 6, and for this reason the ratio of  $\kappa_e$  and  $\kappa_p$  at temperatures  $T$  above  $T_c$  in these samples is unknown. However, according to recently published data<sup>10</sup> from observations of the thermal Hall effect in such Y crystals, it can be estimated that in the normal state  $\kappa_e \sim 0.1 \kappa_p$  at  $T \sim T_c$  and in the superconducting state at  $T \approx 0.4 T_c$   $\kappa_{e,\text{max}}/\kappa_e(T_c) \approx 6$  and correspondingly  $\kappa_{p,\text{max}}/\kappa_p(T_c) \approx 1.3$ . Therefore, even though the electronic component increases much more rapidly than the phonon component, even at the point of the maximum the contribution of the phonon component is more than two times greater than  $\kappa_e$ . The fact that  $\kappa_e$  and  $\kappa_p$  are higher in the superconducting state indicates that at temperatures  $T \sim T_c$  the phonon and charge-carrier path lengths are limited mainly by mutual scattering of quasiparticles by one another. It is pertinent to note that under these conditions the increase occurring in the electronic component  $\kappa_e$  at the transition from the normal state to the superconducting state is also possible in the BCS model. According to the calculations in Ref. 2, in perfect metallic crystals with weak electron-phonon coupling the ratio  $\kappa_e(T)/\kappa_e(T_c)$  can increase by a factor of 2.4 on cooling to  $\sim 0.3 T_c$ , and only then does the exponential decrease in the carrier concentration cause  $\kappa_e$  to decrease with decreasing temperature. The distinguishing feature of high- $T_c$  crystals is that, in contrast to metals,  $\kappa_p \geq \kappa_e$  at  $T \sim T_c$ . In calculations of the behavior of perfect high- $T_c$  crystals it is therefore necessary to take into account the change in both components, just as it is necessary to take into account the possible influence of mutual phonon-electron drag.

We note that a new mechanism of heat transfer by carriers in high- $T_c$  crystals (acoustic bosons) was recently proposed<sup>7</sup> to explain the nature of the maxima on the

curves  $\kappa(T)$ . It is obvious that the difference in the values and the temperature dependence of the resistivity  $\rho(T)$  of samples 1, 2, 4, and 5 is associated with not only the differences in the effective carrier path length  $\lambda_e$ , but also the change in the mechanisms of motion and the concentration of charge carriers in each sample. To determine the suitability of the model of Ref. 7 for describing the phenomena considered here, it would be interesting to see how these changes affect the behavior of the thermal conductivity of acoustic bosons.

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