

## Thermal conductivity of $\text{SmBa}_2\text{Cu}_3\text{O}_{7-\delta}$ oxidized at 250 bar: A comparison of the phonon and electron models

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The thermal conductivity ( $\kappa$ ) of  $\text{SmBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , a high- $T_c$  superconductor (HTSC) with  $T_c \approx 95$  K, was measured in the temperature range 4–300 K. Two samples were examined, both were oxidized under 1 bar pressure, and the second one was additionally oxidized under a pressure of 250 bars. The latter sample exhibited a significant increase of  $\kappa$  (by  $\sim 80\%$  at room temperature) as well as a much higher maximum in  $\kappa(T)$  below  $T_c$  ( $\sim 50\%$  in comparison to  $\sim 20\%$  for the nonpressurized sample). A comparison with data of electrical resistivity for these samples indicates that the thermal conductivity of HTSC ceramics is not sensitive to the quality of grain boundaries. The thermal conductivity data for both samples were analyzed within two competing models explaining the origin of the superconducting maximum: the phonon model introduced by Tewordt and Wölkhausen and the electronic model proposed by Yu and co-workers. Some arguments in favor of the electronic model have been given. [S0163-1829(97)05241-7]

### I. INTRODUCTION

Measurements of thermal conductivity ( $\kappa$ ) give a unique opportunity to study the electronic transport phenomena in high- $T_c$  superconductors (HTSC's) in the superconducting state. The thermal conductivity coefficient of these solids is usually divided into phonon ( $\kappa_{\text{ph}}$ ) and electronic contributions ( $\kappa_e$ ), but, as in the case of specific heat, the electronic part of  $\kappa$  is often masked by the dominating phonon contribution. Moreover, the electron-phonon interactions impede the understanding of the  $\kappa(T)$  dependence below  $T_c$ . The origin of the usually observed increase of  $\kappa$  in the superconducting state still remains a subject of controversy. This maximum is frequently interpreted as a manifestation of the increasing phonon mean free path due to progressive condensation of charge carriers with cooling.<sup>1-4</sup> A new explanation was recently proposed<sup>5,6</sup> indicating the electronic contribution as being responsible for the maximum in  $\kappa(T)$ , due to strong suppression of the quasiparticle scattering rate below  $T_c$ . A discussion of literature data concerning both models was presented in Ref. 7.

In a previous paper<sup>8</sup> we presented the data of specific heat ( $C_p$ ), thermopower ( $S$ ), magnetoresistivity, ac magnetic susceptibility, as well as the results of structural, SEM, and EDAX analysis for the three 90-K samples of polycrystalline  $\text{SmBa}_2\text{Cu}_3\text{O}_{7-\delta}$  originating from the same batch. Two of these samples were processed differently under 250 bars oxygen pressure. This additional treatment resulted in a considerable enhancement of superconducting properties in comparison to the nonprocessed sample (much more pronounced specific heat jump at  $T_c$ , steeper fall in ac susceptibility at  $T_c$ , higher fraction of superconducting phase at 77 K) and in a reversal of the thermopower sign, indicating that the processed samples became overdoped. Despite the above, the electrical resistivity at room temperature (RT) increased six times and revealed a semiconductorlike instead of metal-like temperature dependence which was observed before the pressure oxidation (see Fig. 1). The superconducting transi-

tion was observed as a small but sharp drop in  $\rho(T)$  at  $T_c$ , followed by a long tail, strongly dependent on measuring current. These  $\rho(T)$  dependences indicate that the pressure oxidation of  $\text{SmBa}_2\text{Cu}_3\text{O}_{7-\delta}$  samples resulted in degradation of grain boundaries.

In this work we present the measurements of thermal conductivity for the pair of the above-described samples of well-

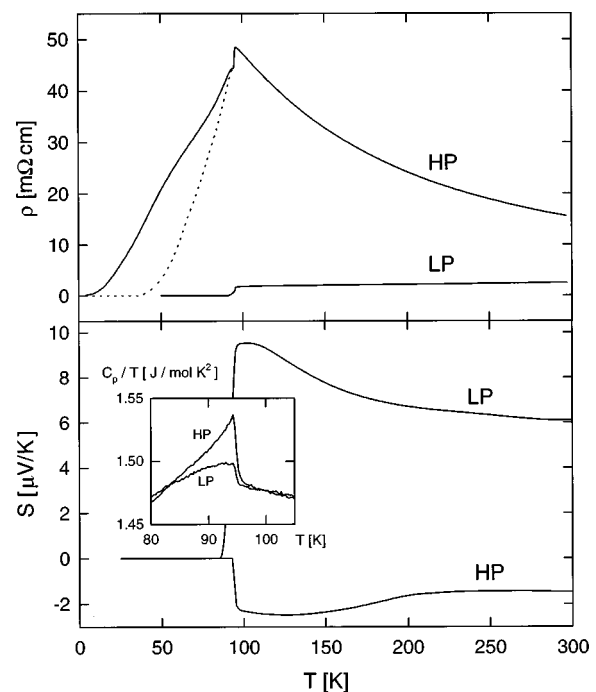


FIG. 1. Temperature dependences of several physical quantities measured for LP and HP samples of  $\text{SmBa}_2\text{Cu}_3\text{O}_{7-\delta}$  presented previously in Ref. 8. Upper panel: the electrical resistivity,  $\rho$  (the solid and dotted lines denote the  $\rho$  measurement with current of 1 and 0.1 mA, respectively). Lower panel: the thermopower,  $S$ . The inset shows the behavior of the specific heat,  $C_p/T$ , near the superconducting transition.

TABLE I. Basic data concerning the  $\text{SmBa}_2\text{Cu}_3\text{O}_{7-\delta}$  samples. The fourth column contains the oxygen content estimated using values of  $S_{300}$  and the data from Ref. 22.  $T_c$  values are assessed from maximum in  $d\rho(T)/dT$  curves. The height of the specific heat jump at  $T_c$  ( $\Delta C_p$ ) was estimated using the entropy-conserving construction.

Sample	$7-\delta$ , measured	$S_{300}$ ( $\mu\text{V/K}$ )	$7-\delta$ , assessed from $S_{300}$	$T_c$ , (K)	$\rho_{300}$ (m $\Omega$ cm)	$\Delta C_p$ (J/mol K)
LP	$6.95 \pm 0.05$	$+6.1 \pm 1$	6.81	95.25	2.5	1.6
HP	$7.00 \pm 0.05$	$-1.5 \pm 1$	6.98	95.25	15	4.6

defined similarities and differences. We have analyzed the collected data in terms of both contradicting models being used in literature to explain the behavior of thermal conductivity of high- $T_c$  superconductors: the phonon model (proposed by Tewordt and Wölkhausen<sup>1</sup>) and electronic model (proposed by Yu and co-workers<sup>5</sup>). The main aim of this paper was to find some arguments against or for these models.

## II. SAMPLES AND EXPERIMENT

For the thermal conductivity measurements we used two of the three  $\text{SmBa}_2\text{Cu}_3\text{O}_{7-\delta}$  samples described in Ref. 8. The pellets were prepared by the conventional solid-state reaction. Then they were heated at 800 °C in flowing oxygen at 1 bar pressure for 5 h, followed by slow (15 °C/h) cooling to room temperature (RT). Such samples were named LP- $\text{SmBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . A few pellets of LP- $\text{SmBa}_2\text{Cu}_3\text{O}_{7-\delta}$  were additionally annealed in pure oxygen under pressure of 250 bars (as measured at 450 °C) in a Morris Research HPS-5015E furnace. They were processed in the following procedure: 2 h at 500 °C, 18 h at 450 °C, 20 h at 400 °C, 24 h at 350 °C, and then furnace cooled. Such samples were named HP- $\text{SmBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (this is the sample HP2 described in Ref. 8).

More details concerning sample preparation procedures, iodometric oxygen content measurements, structural analysis, SEM observations, and other physical quantities as well are presented in Ref. 8. It was estimated from SEM pictures that the typical grain size is of the order of 10  $\mu\text{m}$  for both samples. Table I summarizes the most important data. Figure 1 presents the temperature dependences of the quantities which could help in the analysis of the thermal conductivity data.

The thermal conductivity was measured by a steady-state method employing a manganin-konstantan thermocouple and a small heater glued to the specimen. The temperature gradient along the sample was typically 0.3–0.4 K. Particular care was taken to avoid heat transfer between the sample and the environment. The temperature distribution on the monel radiation shield was maintained close to the thermal gradient on the sample. The maximum experimental systematic error was below 5% (caused mainly by uncertainty in the sample geometry). The samples dimensions varied around  $1 \times 1 \times 8 \text{ mm}^3$ .

## III. RESULTS

The thermal conductivity versus temperature of LP- and HP- $\text{SmBa}_2\text{Cu}_3\text{O}_{7-\delta}$  is presented in Fig. 2. In the normal state a weak temperature dependence was revealed for both investigated samples. The most striking feature is a consid-

erable increase of  $\kappa$  for the sample oxygenated under elevated oxygen pressure (HP), by almost 80% at RT. A slight difference in the slope of temperature dependences in the normal state was also observed: by cooling from 300 K down to 100 K the thermal conductivity decreased by 4% for the LP, but increased by 8% for the HP sample. Like for most of HTSC compounds, a distinct upturn was noticed at  $T_c$  for both samples. However, a wide maximum observed as usual below  $T_c$  is much more pronounced for the HP sample (increase by  $\sim 50\%$  relative to the  $\kappa$  value above  $T_c$ ) than for the LP one ( $\sim 20\%$ ). The temperature of the maximum for HP- $\text{SmBa}_2\text{Cu}_3\text{O}_{7-\delta}$  ( $T_{\text{max}} \approx 48 \text{ K}$ ) is shifted towards lower temperature by  $\sim 10 \text{ K}$  with respect to the initial LP sample ( $T_{\text{max}} \approx 58 \text{ K}$ ). The inset in Fig. 2 shows that at low temperature the dependences of  $\kappa$  for the LP and HP samples approach linear-in- $T$  and  $T^2$  relations, respectively. This difference may reflect different scattering mechanisms dominating the heat transport in the two samples.

The huge increase of  $\kappa$  due to the pressure oxidation is really surprising since it occurs with simultaneous substantial worsening of electrical properties of grain boundaries. This was manifested by a six-fold rise of the electrical resistivity at RT (see Table I) and conversion of its temperature characteristics into a semiconductorlike one (see upper panel in Fig. 1). Therefore, an unexpected conclusion may be formulated: the thermal conductivity of polycrystalline HTSC is not sensitive to the quality of the grain boundaries and re-

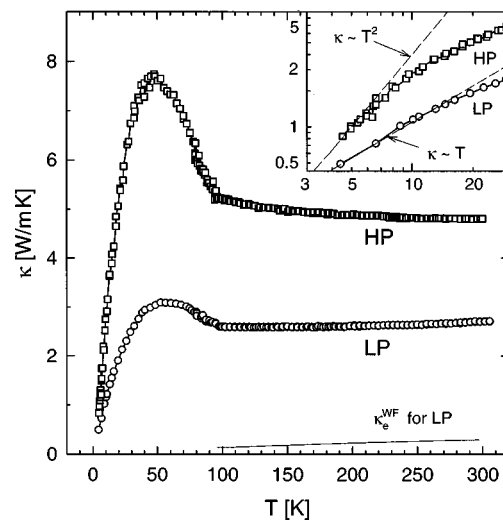


FIG. 2. Temperature dependences of the thermal conductivity of LP- and HP- $\text{SmBa}_2\text{Cu}_3\text{O}_{7-\delta}$  samples. The solid line represents the Wiedemann-Franz estimation of the electronic thermal conductivity ( $\kappa_e$ ) for the LP sample, the respective estimation for HP sample is a few times smaller. The inset shows the low temperature part of the data in the log-log scale.

fects rather the internal properties of grains. Similar insensitivity to grain boundaries was reported for the thermopower.<sup>9</sup> A small change of the  $\kappa(T)$  slope in normal state due to pressure oxidation suggests a slight decrease of phonon scattering due to enhancement of the intragrain quality. This conclusion may be inferred from some similarity of the  $\kappa(T)$  curve for the HP sample to that observed for pure single-crystal HTSC which follows  $1/T$  dependence, characteristic for dominating phonon-phonon scattering.<sup>2,3</sup>

#### IV. THEORETICAL MODELS

There is still a controversy on the origin of the maximum in  $\kappa(T)$  below  $T_c$ .<sup>7</sup> The phonon<sup>1</sup> and electronic<sup>5</sup> mechanisms are under debate. It seems that most authors are still convinced by the first model (see, e.g., Ref. 11). However, the second explanation was supported by several results, namely by a sharp rise of the carrier relaxation time below  $T_c$  inferred from absorption studies using ultrafast lasers<sup>12</sup> as well as by an observation of a rapid increase of the quasiparticle mean free path below  $T_c$  in surface resistance measurements<sup>13</sup> and, more recently, in thermal Hall conductivity experiments.<sup>14</sup> Moreover, the authors of Refs. 5 and 6 indicated that the absence of a clear superconducting anomaly in  $\kappa_c$ , the thermal conductivity along the  $c$  direction, seems to rule out the phonon explanation.

Despite the same  $T_c$  and similar oxygen concentration values, the LP and HP samples exhibit a significant difference in the values and temperature dependences of the total thermal conductivity as well as important dissimilarities in electronic properties. Therefore, they seem to be good candidates for testing the phonon and electronic models. The thermopower values locate them (see Table I) on opposite sides of the 90-K plateau, which is usually observed for 123 compounds on the dependence of  $T_c$  vs oxygen content. The height of the specific heat jump at  $T_c$ , which, according to the BCS theory, is a measure of the electronic density of states, is approximately three times higher for the HP sample. Doubling of the Hall charge carrier concentration ( $n_H$ ) was reported for the Y-123 thin film with  $\delta \approx 0$  with respect to that of  $\delta \approx 0.2$  (both samples exhibited  $T_c \approx 90$  K),<sup>15</sup> so similar differences in  $n_H$  could be expected for our pair of samples. Hence, taking into account the significant differences in electronic properties between both samples, most of the difference in thermal conductivity should be rather attributed to changes in  $\kappa_e$ .

On the other hand, the electronic contribution of thermal conductivity for polycrystalline HTSC was estimated by many authors based on Wiedemann-Franz (WF) law, as not exceeding 10–15% of the total  $\kappa$ .<sup>10</sup> This is why many authors connect the maximum in  $\kappa(T)$  with a lattice contribution. The WF estimation of  $\kappa_e$  for the LP sample is presented in Fig. 2 as a solid line, the estimation for the HP samples is considerable smaller. But, our HP sample is an expressive example that the measured electric resistivity of the polycrystalline material may be far away from the genuine intragrain value, which may be inferred from other measurements by comparison with literature data. Thus, due to the peculiar, nonhomogenous nature of this sample, the calculated  $\kappa_e$  values may be significantly misestimated.

#### V. PHONON MODEL

Hoping to find some conclusive arguments we have analyzed the  $\kappa$  data of the LP and HP samples using both concurring models. First, we tried out the phonon model introduced in the work by Tewordt and Wölkhausen.<sup>1</sup> The model is based on Callaway's expression for lattice thermal conductivity:

$$\kappa_{\text{ph}}(T) = \frac{k_B}{2\pi^2\nu} \left(\frac{k_B}{\hbar}\right)^3 T^3 \int_0^{\Theta_D/T} \frac{x^4 e^x}{(e^x - 1)^2} \tau(x, T) dx,$$

where  $\nu$  is the phonon velocity,  $\Theta_D$  is the Debye temperature, and  $\tau$  is the relaxation time given by

$$\tau^{-1}(x, T) = B + D_p T^4 x^4 + D_{sf} T^2 x^2 + D_d T x + U T^3 x^2 + E T x g(x, y),$$

where  $x = \hbar\omega/k_B T$  is the reduced phonon energy,  $y = \Delta(T)/k_B T$  denotes the reduced superconducting gap. The symbols  $B$ ,  $D_p$ ,  $D_{sf}$ ,  $D_d$ ,  $U$ , and  $E$  describe the phonon scattering by crystal boundaries, point defects, sheetlike faults, dislocations, phonons (Umklapp processes), and electrons, respectively. The Umklapp term was not included in the original TW theory. It was added by several authors<sup>2,3</sup> to delineate the increase of  $\kappa$  with decreasing temperature observed for high-quality HTSC samples in the normal state. The function  $g(x, y)$  was calculated in Ref. 16 and denotes the ratio of the relaxation times of phonons due to scattering on electrons in the normal and superconducting states. The function  $g(x, y)$  drops sharply down for phonons of energy smaller than  $2\Delta$ . According to the TW theory, this drop is responsible for the maximum observed in the  $\kappa(T)$  dependence in the superconducting state. In our analysis we neglected all effects connected with structural anisotropy, pairing symmetry (we assumed  $s$  pairing), and electron-phonon coupling strength (we assumed the ratio  $2\Delta/k_B T = 3.52$  for the weak coupling case), which were considered in Ref. 17. The reason is that these aspects do not result in such qualitative changes of  $\kappa(T)$  shape, which might be recognized in our experiment. Due to the very low value of the Wiedemann-Franz estimation, the electronic contribution was also neglected in this analysis.

During the work with the phonon model we have tried out several different combinations of scattering mechanisms (we used from three to five fitting parameters). In each trial, however, we employed the electron ( $E$ ) and Umklapp ( $U$ ) terms in order to reproduce the maximum in the superconducting state and follow the slope of the  $\kappa(T)$  dependence in normal state for the HP sample. For all fittings we assumed  $T_c = 95.2$  K (as measured for both samples in other experiments) and  $\Theta_D \approx 500$  K (as estimated in our previous paper<sup>18</sup> from the analysis of the specific heat of  $\text{SmBa}_2\text{Cu}_3\text{O}_{7-\delta}$  in the temperature range 200–300 K). Finally, we concluded that all models with only four parameters (i.e., with four independent scattering mechanisms, including  $E$  and  $U$ ) may be regarded as satisfactory. For some more complicated models it was even impossible to find out the optimal set of parameters values due to the effect of overparametrization. The exemplary results of the fitting by the phonon model comprising the most basic scattering mechanisms are pre-

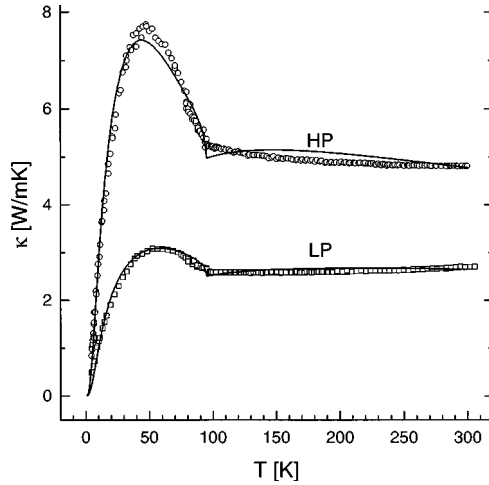


FIG. 3. A fit of the thermal conductivity for LP and HP samples by the phonon model of Tewordt and Wölkhausen (Ref. 1). The solid lines represent a model for lattice thermal conductivity including phonon scattering by boundaries, point defects, phonons (Umklapp processes), and electrons. The fitting parameters are shown in Table II. In this model the maximum in  $\kappa(T)$  is due to the rapid decrease of the phonon scattering on electrons below  $T_c$ .

sented in Fig. 3. The obtained optimal sets of parameters are presented in the upper part of Table II.

Before discussing the results of fitting some remarks about the fitting procedure would be in place. In the case of Callaway's expression the fitting is particularly inconvenient, since the errors of the fitting parameters are strongly correlated (we found correlation coefficients between pairs of parameters errors to be up to the value of  $|r|=0.97$ ). Moreover the topology of the multidimensional surface on which we are looking for the minimum (i.e., the sum of square deviations as function of parameters) is rather complicated—it includes saddle points and local minima. Therefore, we have put particular care on the error analysis and two types of errors have been calculated (see Table II). The smaller errors (“ $\delta$ ”) denote the confidence intervals for one parameter with

the other parameters being of optimal values. The larger errors (“ $\Delta$ ,” given in the parenthesis) denote the extensions of the full confidence ellipsoid for all parameters [which are proportional to  $(C_{ii})^{1/2}$ , where  $C_{ii}$  are the diagonal elements of the inverse Hessian matrix]. This method takes into account all correlations between the parameter errors. If  $\delta$  is higher than the parameter value then the respective term may be rejected from the model—e.g., this is the case for the  $D_{sf}$  term, which does not increase the goodness of the fit if added to the phonon model described in Table II. If the  $\Delta$  is higher than the parameter value, then the physics of the model is not ruled out, it means only that precise values of the parameters cannot be obtained from such an experiment. However, for a pair of closely related LP and HP samples the observed trends of parameter changes may be regarded as reliable. For both samples the estimated  $E$  parameters are the same (even within  $\delta$  accuracy). Therefore, in the framework of the phonon model the increase of the height of the superconducting maximum as well as the increase of  $\kappa$  in the normal state due to pressure oxidation could be regarded just as a result of the removal of most structural imperfections (see  $B$  and  $D_p$  parameters). However, in our opinion, this conclusion could hardly be reconciled with the observed strong increase of the electrical resistivity.

## VI. ELECTRONIC MODEL

The electronic model used here was constructed after Refs. 5 and 6. However, as in the case of the phonon model and for the same reason, we neglected the corrections due to the pairing symmetry and the strength of electron-phonon coupling. Therefore, to calculate the electronic thermal conductivity we applied the original isotropic formula from the work of Kadanoff and Martin:<sup>19</sup>

$$\kappa_e = \frac{1}{T} \frac{1}{k_B T} \frac{n_e}{2m_e} \int_0^\infty \epsilon^2 \operatorname{sech}^2\left(\frac{E}{2k_B T}\right) \Gamma(T)^{-1} d\epsilon,$$

where  $n_e$  is the electron concentration,  $m_e$  is the electron mass,  $\epsilon$  is the electron energy,  $E = (\epsilon^2 + \Delta^2)^{1/2}$ . The quasi-particle scattering rate  $\Gamma$  is assumed to be energy indepen-

TABLE II. The fitting parameters for the phonon and electronic models. The symbols  $B$ ,  $D_p$ ,  $U$ , and  $E$  denote the phonon scattering by crystal boundaries, point defects, phonons (Umklapp processes), and electrons, respectively;  $KM$  denotes the electron contribution to  $\kappa$ . The terms with  $E$  and  $KM$  parameters are responsible for the superconducting maximum in  $\kappa(T)$  within the framework of the respective model. The fitted curves are presented in Figs. 3 and 4. The errors ( $\delta$ ) denote the confidence region for respective parameter with other parameters being of optimal values. The errors in parenthesis ( $\Delta$ ) denote the range of the projection of the full confidence ellipsoid in the four-dimensional parameter space on the direction of the respective parameter.

Sample name	Fitted parameters $\pm \delta$ ( $\pm \Delta$ )			
	Phonon model			
	$B$ ( $\times 10^{-6}$ )	$D_p$	$U$ ( $\times 10^{-2}$ )	$E$ ( $\times 10^{-3}$ )
LP	$65 \pm 9$ ( $\pm 40$ )	$3.9 \pm 0.2$ ( $\pm 1.4$ )	$1.9 \pm 1.0$ ( $\pm 4$ )	$2.3 \pm 0.4$ ( $\pm 2$ )
HP	$22 \pm 5$ ( $\pm 20$ )	$1.3 \pm 0.1$ ( $\pm 1$ )	$4.3 \pm 1.0$ ( $\pm 6$ )	$2.2 \pm 0.5$ ( $\pm 2$ )
	Electronic model			
	$B$ ( $\times 10^{-6}$ )	$D_p$	$U$ ( $\times 10^{-2}$ )	$KM$ ( $\times 10^{-3}$ )
LP	$14 \pm 2$ ( $\pm 10$ )	$15.4 \pm 0.7$ ( $\pm 6$ )	$0.7 \pm 0.6$ ( $\pm 1.6$ )	$0.8 \pm 0.1$ ( $\pm 0.3$ )
HP	$0.9 \pm 0.3$ ( $\pm 4$ )	$21 \pm 2$ ( $\pm 33$ )	$1.7 \pm 0.4$ ( $\pm 3$ )	$3.5 \pm 0.2$ ( $\pm 1.2$ )

dent and composed of two parts. The first one originates from the inelastic scattering on phonons and for usual metals at low temperature it is proportional to  $T^3$ . For simplicity we neglected here the second term, the residual scattering rate due to impurities (temperature independent).

The physical reason for the superconducting maximum in  $\kappa(T)$  within this model is the steep increase of mean free path of the noncondensed charge carriers below  $T_c$ . This increase is high enough to override the effect of the diminishing number of entropy carrying carriers due to progressive superconducting condensation. The phenomenological models from Refs. 5 and 6 assumed that in the superconducting state the inelastic scattering rate follows a power law,  $\sim(T/T_c)^n$ , with  $3 < n < 5$ , depending on the pairing symmetry and the strength of electron-phonon coupling. In the Refs. 13 and 14 the steep fall of  $\Gamma$  just below  $T_c$  was observed experimentally. In this work we simply assumed that in the superconducting state  $\Gamma \sim (T/T_c)^4$ , however a change of the exponent  $n$  within borders estimated in Refs. 13 and 14 would not change qualitatively our results.

The transport properties of HTSC's in the normal state are unusual and not explained yet. However, one of the most universal observed features of high- $T_c$  superconductors, the  $T^{-2}$  dependence of the charge carrier mobility<sup>20</sup> (valid for both over- and underdoped materials), prompted us to assume  $\Gamma \sim T^2$ . We also have taken into account the temperature dependence of charge carrier concentration for optimal doped HTSC,  $n_e \sim T$ .<sup>20</sup> Thus, the resulting  $\kappa_e$  is temperature independent in a normal state, as in Ref. 6, where  $n_e \approx \text{const}$  and, based on the WF law and  $T$ -linear resistivity dependence,  $\Gamma \sim T$  were assumed.

To describe the total thermal conductivity we assumed that it is a sum of electronic and phonon contributions. The electronic term was expressed as  $KM * \kappa_e$ , where  $KM$  is a fitting parameter and  $KM \sim n_e / \Gamma_0$ ,  $\Gamma_0$  is the temperature independent part of the scattering rate (i.e., it is the electron-phonon interaction), and  $\kappa_e$  denotes here the temperature dependent part of the Kadanoff-Martin expression. The phonon term was described by Callaway's formula for phonon scattering on boundaries ( $B$ ), point defects ( $D_p$ ), and other phonons ( $U$ ). The errors of the fitting parameters were analyzed as for the phonon model. The fitting results are presented in the bottom of Table II and in Fig. 4. It may be noticed that within the framework of the electronic model the increase of the height of the superconducting maximum as well as the increase of  $\kappa$  in the normal state due to pressure oxidation should be regarded as a result of the significant increase of the electronic contribution (it also means that the WF law could not be applied at least for the HP sample). The phonon part of  $\kappa$  remained almost the same (see dashed lines in Fig. 4), whereas a four-fold increase of the  $KM$  parameter is evident (even in terms of  $\Delta$  errors). The change of the  $KM$  may be attributed to the decrease of  $\Gamma_0$  or the increase of  $n_e$ . The second explanation seems to be more convincing, since a strong increase of Hall carrier concentration due to the increase of oxygen content was indeed reported for 123-compounds (e.g., Ref. 15). Moreover, significant changes in other electronic properties (especially the correlation of  $\kappa_e$  and  $\Delta C_p$  values—see the inset in Fig. 4) also support this interpretation.

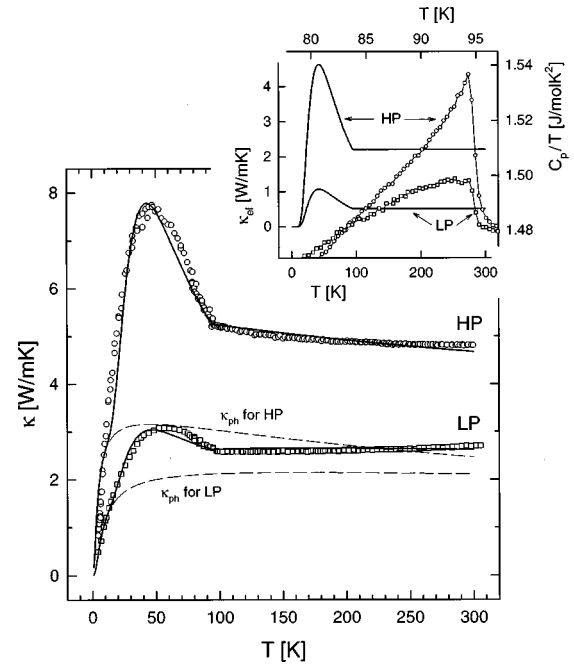


FIG. 4. A fit of the thermal conductivity for LP and HP samples by the electronic model of Yu and co-workers (Ref. 5). Solid lines represent a model composed of two contributions: electronic term (with inelastic electron scattering) and lattice term (taking into account the phonon scattering on: boundaries, point defects, and phonons). The dashed lines present the lattice term only. The fitting parameters are shown in Table II. The inset shows a correlation between the electronic part of  $\kappa$  and the jump of specific heat at  $T_c$ . In this model the maximum in  $\kappa(T)$  is due to a rapid decrease of electron scattering below  $T_c$ .

## VII. CONCLUSIONS

Few pellets of commonly oxidized  $\text{SmBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (LP) were additionally treated under an elevated oxygen pressure of 250 bars (HP). Pressure oxidation resulted in dramatic worsening of grain boundary quality, as observed by a multiple increase of electrical resistivity. Despite that, the HP sample exhibited a significant increase of the measured thermal conductivity (by  $\sim 80\%$  at RT) as well as a much more pronounced maximum in  $\kappa(T)$  below  $T_c$ . The unexpected conclusion was inferred that thermal conductivity of ceramic HTSC is not sensitive to the grain boundary quality.

The LP and HP samples are very similar in some aspects (the same batch in chemical synthesis, similar microstructure, the same  $T_c$ ), but significantly different in other aspects (opposite signs of thermopower, i.e., different doping level of  $\text{CuO}_2$  planes, different heights of  $\Delta C_p$ ). Therefore we analyzed the thermal conductivity results for this pair of well-defined samples within the two models still concurring for the explanation of the maximum in  $\kappa(T)$  in the superconducting state. It appeared that both models, the phonon<sup>1</sup> one and the electronic<sup>5</sup> one, may be satisfactorily fitted to the results for both samples. However, a careful error analysis of fitting procedures revealed high error correlations caused by numerical properties of the models. Therefore, the fitting parameters which are not responsible for the superconducting maximum in  $\kappa_e(T)$  (i.e., phonon scattering rates due to various defects and Umklapp processes) could be estimated with

only very limited accuracy. Nevertheless, we concluded that, in the framework of the phonon model, the changes in  $\kappa(T)$  due to pressure oxidation should be attributed rather to the removal of structural defects, and not to the increase of phonon-electron interactions. In the framework of the electronic model the changes in  $\kappa(T)$  seem to be connected with an  $\sim$ fourfold increase of the  $\kappa_e$  contribution for the HP sample with respect to that for the LP sample. This increase may be easily interpreted just as a result of the increase in charge carrier concentration.

The estimation of the validity of the two models is not obvious, but in our opinion the electronic model seems more reliable. First of all, it seems unlikely that the treatment at relatively low temperature (up to 500 °C) could result in such a significant removal of structural imperfections, what was inferred from the analysis within phonon model. The observed worsening of grain boundaries supports this doubt. On the other hand, the increase of  $n_e$ , necessary to explain changes in the  $\kappa(T)$  curve due to pressure oxidation, was actually observed in Hall effect measurements for 123 compounds.<sup>15</sup> Moreover, all main differences between LP and HP samples are of electronic origin (change of ther-

mopower sign,  $\sim$ threefold increase of  $\Delta C_p$  at  $T_c$ ), hence the electronic explanation of the differences in the thermal conductivity seems to complete the physical picture. It is noteworthy that the rise of  $\Delta C_p$  agrees well with the increase of  $\kappa_e$ . It agrees well with the observation of Cohn,<sup>21</sup> who indicated the correlation between heights of superconducting anomalies in thermal conductivity and specific heat. Thus, in the framework of the electronic model, it may be supposed that changes in the height of the superconducting maximum in  $\kappa(T)$  curves (or, equivalently, in  $\kappa_e$ ) observed for 123 compounds have the same physical origin as strong changes observed for other quantities: in the normal state,  $S$  (Refs. 22 and 23),  $n_e$  (Ref. 15) and in the superconducting state,  $\Delta C_p$  (Refs. 23 and 24) and critical currents,  $J_c$  (Ref. 15).

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