Quasiparticle contribution to heat carriers relaxation time in $DyBa_2Cu_3O_{7-x}$ from heat diffusivity measurements

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(Received 16 April 2001; revised manuscript received 18 June 2001; published 22 October 2001)

It is shown that the controversy on phonons or electrons being the most influenced heat carriers below the critical temperature of high-T_c superconductors can be resolved. Electrical and thermal properties of the same $DyBa_2Cu_3O_{7-x}$ monodomain have been measured for two highly different oxygenation levels. While the oxygenated sample $DyBa_2Cu_3O_7$ has very good superconducting properties ($T_c = 90$ K), the $DyBa_2Cu_3O_{6.3}$ sample exhibits an insulator behavior. A careful comparison between measurements of the thermal diffusivity of both samples allows us to extract the electronic contribution. This contribution to the relaxation time of heat carriers is shown to be large below T_c and more sensitive to the superconducting state than the phonon contribution.

DOI: 10.1103/PhysRevB.64.184521 PACS number(s): 74.25.Fy, 72.15.Jf, 74.72.Jt

I. INTRODUCTION

As far as the temperature dependence of the relaxation time of *heat* carriers in high-T_c superconductors (HTS) is concerned, different regions can be highlighted. At high temperature, the main process is due to phonon scattering.[1,2] At very low temperature, the different scattering processes are decoupled as shown by fast heat response measurements.[3] At intermediate temperatures, below the critical temperature T_c, a bump characterizes the thermal conductivity κ data. This bump is linked to an increase of the mean free path l of the heat carriers or in other words to an increase of their relaxation time τ. A major controversy subsists on the origin of such a behavior, thus about the thermal transport mechanisms themselves below the critical temperature T_c.[2,4,5] Two main models collide. The aim of this work is to determine the nature of the carrier responsible for the increase of the relaxation time τ of heat carriers below T_c and solve the puzzle.

On one hand, Wölkhausen *et al.*[6] have proposed a so-called phonon model in order to explain the bump in κ below the critical temperature T_c. This model which introduces seven parameters describes different relaxation time processes. Phonons are thought to be less scattered by electrons since these condense into Cooper pairs. If the main scattering process below T_c is taken to be the phonon-electron process, the calculation of κ does reproduce the bump observed. One can nevertheless wonder whether the origin of the bump in such a scheme is due to the reduced number of scatterers or to their mobility increase.

On the other hand, the electronic model[7,8] in which the main heat carriers are charges claims that the bump reflects the *mean free path* increase of electrons, below T_c, because the number of electron-electron scattering processes decreases. Indeed below T_c, normal electrons (quasiparticles) should be less often scattered by others because of the existence of Cooper pairs (which cannot carry entropy in the condensed state). This argument is especially interesting in HTS in which the gap symmetry is of *d*-wave type,[9] thus characterized by lines of nodes. Along these the gap is zero,[10] whence the normal electrons *density* always remaining finite even at very low temperature.

From the *electrical* transport point of view, a quasiparticle relaxation time τ_e can be deduced from microwave loss measurements, as done by Bonn *et al.*[11] on $YBa_2Cu_3O_{7-x}$ monocrystal. Another way to determine τ_e through the magneto-transport experiments was made by Krishana *et al.*[12] on a Zn-doped $YBa_2Cu_3O_7$. They measured the thermal Hall effect, also called Righi-Leduc effect, i.e., the temperature gradient orthogonal to the plane defined by the heat flux and an external magnetic field. In so doing, the *electronic contribution* is certainly selected since electrons are deviated by the magnetic field B. Interestingly, these Righi-Leduc effect measurements scale with the measurements of microwave losses[13] giving a correspondence between electrical and thermal measurements of the relaxation time in presence of a magnetic field.

However such a magnetothermal process implies high magnetic fields which disturb the quasiparticle (QP) system and blur interpretation at zero field. In fact, not only the distinction between QP and vortex contributions is far from obvious,[14] but also the existence or not of Landau levels has been argued upon and is still an open question.[15] Moreover in Ref. 12, the relaxation time is found by achieving extrapolation to zero-field; that is known to be very delicate in view of the complicated (B,T) phase diagram of HTS.[16]

II. METHOD

In contrast we propose a *direct comparative method* of electrical and thermal measurements in order to deduce whether phonons or electrons are responsible for the increase of the relaxation time of heat carriers below T_c. The measurement of the thermal diffusivity coefficient α allows one a *direct* access to the relaxation time of heat carriers indeed.

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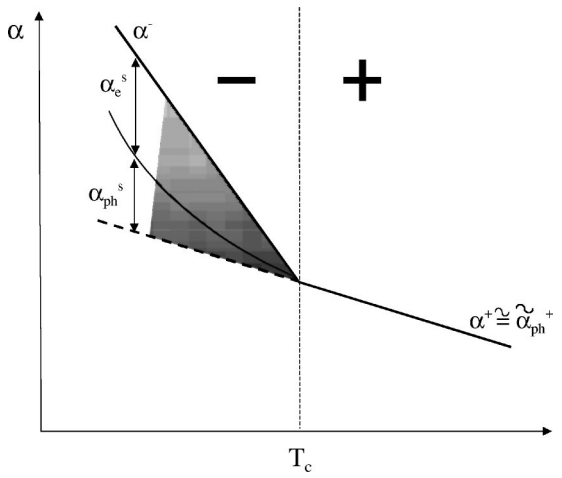

FIG. 1. Sketch of the behavior of the thermal diffusivity for a high-T_c superconductor (continuous line) as a function of temperature. The broken line ($\tilde{\alpha}^+$) is the extrapolated normal thermal diffusivity below T_c; the shadowed area represents the superconducting phase contribution made of α_e^s and $\alpha_{\rm ph}^s$ below T_c. The line separating α_e^s and $\alpha_{\rm ph}^s$ is merely indicative.

Let it be recalled that α reads[17]

$$\alpha = \frac{1}{3} v l = \frac{1}{3} v^2 \tau \tag{1}$$

if $2\pi\nu\tau \gg 1$, where ν is the phonon or electron change in frequency, and v is the average speed of the carriers. For electrons, the change of energy close to T_c is of the order of the gap energy; the change in frequency is $\nu \simeq 10^{13}$ Hz (for Dy-123) while $\tau \simeq 10^{-12}$ s.[11] Claiming without lack of much generality that v is constant in a 50 K range around T_c, the thermal diffusivity can be assimilated to the relaxation time. Moreover, α can be decomposed as

$$\alpha = \frac{\kappa}{dc} = \frac{\kappa_e + \kappa_{\rm ph}}{dc} = \frac{\kappa_e}{dc} + \frac{\kappa_{\rm ph}}{dc} = \alpha_e + \alpha_{\rm ph} \propto \tau_e + \tau_{\rm ph}, \tag{2}$$

where κ is the thermal conductivity, c the specific heat ($c_e \ll c_{\rm ph}$), and d the density. The subscripts e and ph represent the electronic and the phononic contribution in each quantity.

A home made apparatus has been constructed to measure α directly, together, and simultaneously with the thermoelectric power S and κ.[18,19] The same thermocouples are used to measure those three physical parameters. The thermal diffusivity was found to be described in a log-log plot by two straight lines crossing at T_c (Ref. 20) with the power law exponent larger below than above T_c. Figure 1 shows a sketch of the behavior of α versus temperature on a log-log plot for further reference. The normal state is designated by the + sign and the superconducting state by the − sign. The excess contribution due to the superconducting phase is found by extrapolating the normal state value below the critical temperature (gray area in Fig. 1). The thermal diffusivity below T_c can be written as

$$\alpha^- = \alpha_e^- + \alpha_{\rm ph}^- = \alpha_e^s + \tilde{\alpha}_e^+ + \alpha_{\rm ph}^s + \tilde{\alpha}_{\rm ph}^+, \tag{3}$$

where α_e^s and $\alpha_{\rm ph}^s$ are the true superconducting (s) contributions of electrons and phonons below T_c respectively and the tilde quantities are the normal state ones extrapolated below T_c. Our method consists in evaluating the extrapolated value of $\alpha_{\rm ph}^+$ below T_c, i.e., $\tilde{\alpha}_{\rm ph}^+$, in order to remove this term from Eq. (3), and to have only $\alpha_e^- + \alpha_{\rm ph}^s$ to analyze. This quantity is then *compared* to the quasiparticle relaxation time obtained by microwave losses measurements from Ref. 11 in order to *estimate* τ_e, in our notations τ_e^-, thus α_e^-. In such a method, no model is *a priori* assumed concerning the nature of heat transport carriers responsible for the increase of τ (in other words, α) below the critical temperature.

The various contributions to α are obtained by comparing the thermal properties of a Dy-123 monodomain before and after deoxygenation as in Refs. 21,22 for polycrystaline superconductors. In so doing, the thermal diffusivities of the sample should strictly behave as those of a superconductor in the well oxygenated case ($\alpha_{\rm ox}$) or as those of an insulator in the deoxygenized case ($\alpha_{\rm deox}$) because the density of holes is then largely decreased. A main argument is that the geometry is *conserved* and the phonon spectrum quasiundisturbed, such that any spurious effect can be easily taken into account by rescaling at high temperature. Is so doing, $\tilde{\alpha}_{\rm ph}^+$ can then be assimilated to the thermal diffusivity of the deoxygenated sample α_{deox}.

III. SYNTHESIS AND EXPERIMENTS

The simple domain synthesis is described in Ref. 23. A bar was cut out from the single grain. Its dimension was $15 \times 2 \times 2$ mm^3. The electrical resistance $R(T)$, thermal conductivity, thermoelectric power $S(T)$ (not shown here), thermal diffusivity have been measured and indicate the features expected from a good sample and allow for further work. The sample was then deoxygenized. This operation was controlled by TGA (thermogravimetric analysis). The weight of the sample was continuously recorded while it was heated in a furnace up to 850°C. The composition of the sample was thereby determined since the loss of weight was essentially due to the loss of oxygen. Starting from a fully oxygenated sample $DyBa_2Cu_3O_7$ as seen from $R(T)$, $S(T)$, and then T_c, a $DyBa_2Cu_3O_{6.3}$ stoichiometry was found after TGA in the present case.

The resistivity measurements have been achieved in a PPMS (physical properties measurement system from Quantum Design). The thermal conductivity and thermoelectric power on one hand, and the thermal diffusivity on the other hand, have been simultaneously measured following the steady state method and the pulse method Ref. 18 respectively, using the homemade setup described in Ref. 19.

IV. RESULTS

To begin with, the normalized resistance of the oxygenated $R_{\rm oxy}$ and deoxygenized samples $R_{\rm deox}$ are compared on Figs. 2(a) and 2(b), respectively. The electrical behaviors are quite opposite to each other. On one hand, the oxygenated sample, Fig. 2(a), is a very good superconductor with a very

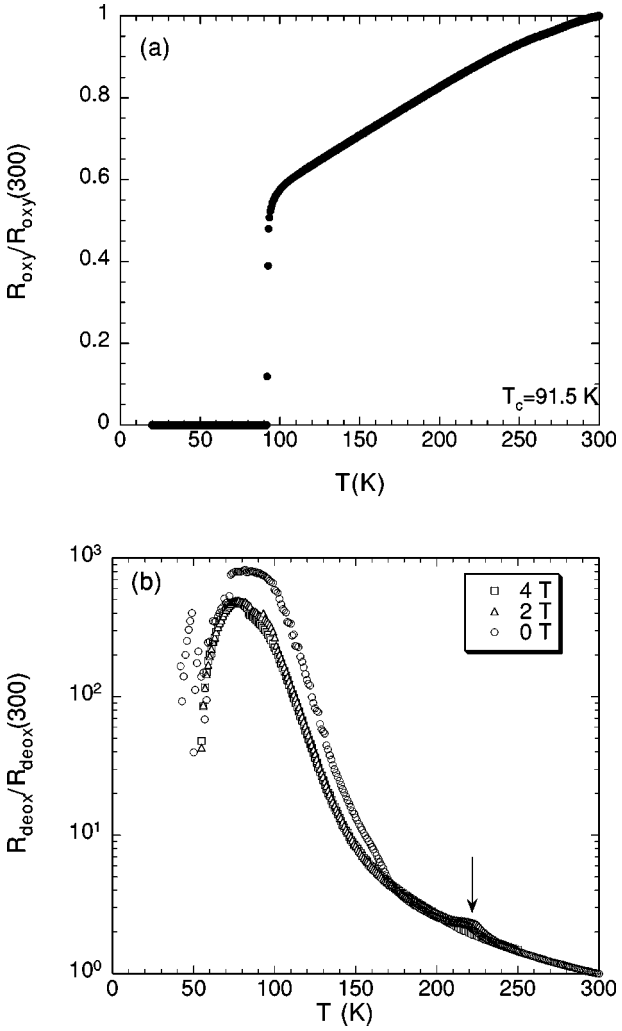

FIG. 2. (a) Normalized resistance of a $DyBa_2Cu_3O_7$ sample versus temperature. (b) Normalized resistance of the sample when it is deoxygenized to be $DyBa_2Cu_3O_{6.3}$. The data is shown when different magnetic fields are applied versus temperature in a semi log plot. The arrow indicates the antiferromagnetic transition.

sharp transition. On the other hand, the deoxygenized sample, Fig. 2(b), exhibits a semiconducting (nearly insulating) behavior. For the latter sample, its Néel temperature ($T_N = 230$ K) has been revealed by applying a small magnetic field. Notice that the maximum at $T \simeq 45$ K is slightly field dependent. The origin of such a maximum is unknown. It might be due to the existence of a spin glass or canted spin phase extending in the AF insulating phase. The curve shape reminds of that found in GMR materials.[24] This maximum limits the subsequent analysis to the above 50 K range. It seems therefore quite reasonable to claim that two different regions of the (T_c, x) phase diagram[25] are investigated, and that the electronic contribution to the electrical transport is wholly suppressed in the deoxygenated sample.

The thermal conductivity is shown in Fig. 3 for both cases. Since those results have been obtained for the same sample, the geometric factor does not influence the absolute values of the measurements. For the oxygenated sample (●), the bump below the critical temperature in the thermal

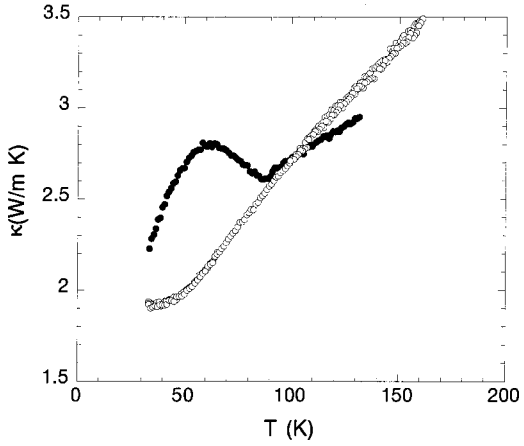

FIG. 3. Comparison of the thermal conductivity of $DyBa_2Cu_3O_7$ (●) and $DyBa_2Cu_3O_{6.3}$ (○) versus temperature.

conductivity κ_{oxy} is well defined. A sharp minimum occurs near T_c at 90 K. This feature is not found when the sample is deoxygenized (○): the bump disappears and the thermal conductivity κ_{deox} slope remains quite unchanged, smoothly increasing with T up to high temperatures. In the normal state, the thermal conductivity increases in both cases, thus behaving similarly to a glass. Notice the flattening of κ_{deox} curve below 50 K which might as for the resistivity due to some magnetic effects.

The total phonon contribution to the thermal conductivity $\kappa_{ph,t}$ can be written as[26]

$$\kappa_{ph,t}^{-1} = \kappa_{ph}^{-1} + \kappa_{ph,b}^{-1} + \kappa_{ph,i}^{-1} \tag{4}$$

where κ_{ph}, $\kappa_{ph,b}$ and $\kappa_{ph,i}$ are the contributions due to the scattering of phonons by phonons, of phonons by boundaries and of phonons by impurities, respectively. The slope of the heating conductivity of the deoxygenated sample is higher than in the oxygenated case. This can be explained by the thermal treatment used in order to decrease the oxygenation level by the TGA process. The terms $\kappa_{ph,b}$ and $\kappa_{ph,i}$ are then

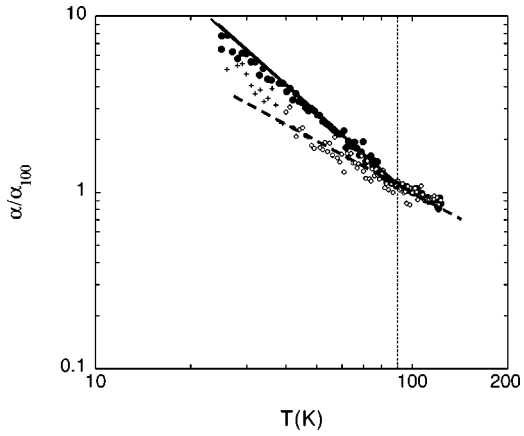

FIG. 4. Comparison of the thermal diffusivity of $DyBa_2Cu_3O_7$ (●) and $DyBa_2Cu_3O_{6.3}$ (○) versus the temperature presented in a log-log plot. Data below 50 K are represented by crosses. The lines drawn correspond to Fig. 1. All data are normalized at $T = 100$ K for better comparison with Fig. 1.

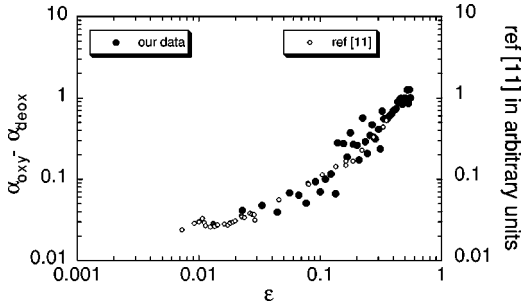

FIG. 5. Plot of $\alpha_{oxy}-\alpha_{deox}$ (●) obtained from Fig. 4 versus the reduced temperature $\epsilon=|T-T_c|/T_c$ for comparing with the microwave loss results obtained by Bonn *et al.* (Ref. 11) (○). These measurements have been rescaled by a multiplicative factor estimated at 100 K for comparison.

slightly modified in Eq. (4). As far as the term κ_{ph} is concerned, calculations for both oxygenated and deoxygenated samples show that their phonon spectra are very close to each other.[27] The authors of Ref. 27 also claim that just a few modes can be distinguished frequency spectra of the oxygenated and deoxygenated case. Consequently they are suitable as the reference phonons for the purpose of superstructure characterization, but the thermal transport by phonons should be only slightly affected. Therefore, $\tilde{\alpha}^{+}_{ph}$ can be interpreted as the thermal diffusivity of the deoxygenized sample α_{deox}: it is a purely phononic contribution. Nevertheless we stress that the analysis is valid only above 50 K up to T_c.

The measurements of both (normalized) thermal diffusivities are plotted in Fig. 4 in a log-log plot. The behavior of the thermal diffusivity of the oxygenated sample α_{oxy} is typical of high-T_c superconductors.[28–32] Straight lines are found in the normal state and in the superconductor state in the temperature range of interest. The characteristic power law exponents are found to be -0.77 and -1.5 in the normal and in the superconductor state. Those exponents can be explained along the lines of the electronic theory previously reported in Ref. 20. This behavior is not found in the thermal diffusivity α_{deox} of the deoxygenized sample. Note that both α's are superposed to each other in the normal state exactly like in the sketch of Fig. 1. In order to *numerically* substract α_{deox} from α_{oxy}, α_{deox} has been smoothened by filtering high frequencies in the Fast Fourier Transform spectrum of the signal. In Fig. 5, $\alpha_{oxy}-\alpha_{deox}$ (●) is shown versus the reduced temperature $\epsilon=|T-T_c|/T_c$. It is obvious from the graph that we can write

$$\alpha_{oxy}-\alpha_{deox}\equiv\alpha^{-}-\tilde{\alpha}^{+}_{ph}=\alpha^{-}_{e}+\alpha^{s}_{ph}. \tag{5}$$

For comparison, the QP relaxation time from Bonn *et al.*[11] is also shown as circles (○), which are rescaled by a multiplicative factor. The agreement is remarkable. The quantity of Eq. (5) is proportionnal to τ_e, the QP relaxation time found by microwave loss measurements. This shows that the *electronic* term α^{-}_{e} is unambiguously the dominant term in Eq. (3).

V. CONCLUSIONS

In conclusion, a comparison between electrical and thermal measurements of an oxygenated and desoxygenated sample allows one to determine the nature of thermal carriers in high-T_c superconductor ceramics. Direct measurements of the thermal diffusivity α on a $DyBa_2Cu_3O_{7-x}$ sample are reported for $x=0$ and $x=0.7$. Such a direct measurement method of α leads to an unambiguous result without the use of any magnetic field and without any *a priori* assumption about the theoretical model used for describing the scatterers: phonons or electrons. Electrons are found to be the most influenced heat carriers below the critical temperature. Phonons are only background carriers.

ACKNOWLEDGMENTS

Part of this work has been financially supported by the ARC 94-99/174 contract of the Ministry of Higher Education and Scientific Research through the University of Liège Research Council. S.D. has benefited from the FRIA. We also would like to thank Professor H.W. Vanderschueren for the use of MIEL equipment. We also want to thank Professor J.P. Maneval (ENS, Paris) and Professor E. Silva (U. Roma 3, Roma).

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