

# The Peak in the Thermal Conductivity of Cu-O Superconductors: Electronic or Phononic Origin?

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The thermal conductivity  $\kappa$  of hole-doped Cu-O plane high- $T_c$  perovskites exhibits a dramatic increase below  $T_c$  which results in a pronounced peak near  $T_c/2$ . The origin of this peak was initially thought to arise from an enhancement in the mean-free path of phonons as the charge carriers undergo condensation. Indeed, excellent fits to the data can be obtained with physically reasonable parameters using the conventional theory of lattice conduction in superconductors. In contrast, a recently observed sharp decrease in the quasiparticle scattering rate of YBCO single crystals below  $T_c$  has motivated proposals for an electronic origin of the thermal conductivity peak. We shall critically examine experimental evidence and highlight relative advantages and shortcomings of the two contrasting interpretations. Furthermore, we shall draw attention to recently available data on the relaxation time of out-of-equilibrium carriers in Cu-O superconductors obtained using pump-probe femtosecond laser studies and what new light they shed on the controversy.

**KEY WORDS:** High- $T_c$  superconductors; thermal conductivity; scattering rate.

## 1. INTRODUCTION

Rarely does one see as much attention paid to the behavior of the thermal conductivity,  $\kappa(T)$ , as we have witnessed in the case of high-temperature superconductors. An interesting temperature dependence coupled with a sensitivity to composition, microstructure, and external stimuli such as a magnetic field and neutron irradiation make high- $T_c$  perovskites a very attractive target for thermal conductivity studies [1]. The fact that the experimental data are available across a wide temperature range, uninterrupted by vanishing signals in the superconducting domain, adds to the appeal of the thermal conductivity as a rich source of information on the dynamics of carriers and phonons.

The most characteristic feature of the thermal conductivity for a vast majority of high- $T_c$  superconductors (although the electron-doped Cu-O

perovskites such as  $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$  seem to be an exception) is a sharp rise in the conductivity which sets in at  $T_c$  and which culminates in a peak near  $T_c/2$ . It is this feature, illustrated for a representative set of data on YBCO in Fig. 1, that is the focus of this paper.

A hint of an increase in  $\kappa(T)$  as the temperature of a superconductor falls below  $T_c$  was seen already in 1950 in a rare case where  $T_c$  of a conventional superconductor was high enough and the material had a large lattice thermal conductivity, i.e., Pb-10%Bi alloy [5]. The issue was pursued theoretically by Geilikman and Kresin [6] and by Bardeen, Rickayzen, and Tewordt in what is nowadays referred to as the BRT theory [7]. The essential point of this theory is the realization that as the electrons condense and the electronic thermal conductivity rapidly vanishes below  $T_c$ , the mean-free path of phonons may increase to such an extent that the lattice thermal conductivity more than compensates for the loss of the electronic contribution. A prerequisite for this to happen is a modest electron-phonon interaction and a lattice thermal conductivity which is non-negligible

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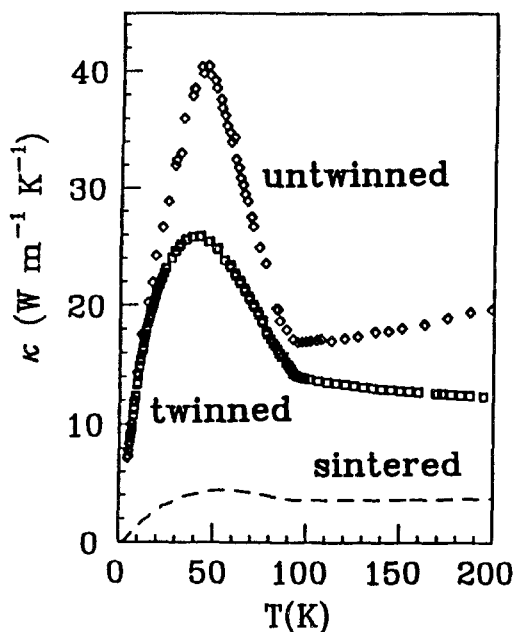


Fig. 1. Thermal conductivity of sintered [2], twinned [3], and untwinned (*a*-direction) [4] samples of YBCO.

in comparison to the electronic thermal conductivity in the normal state. Clean conventional superconductors do not satisfy the above criteria and one normally observes a rapidly diminishing thermal conductivity as the metal enters its superconducting domain. An extreme case where the relevant parameters are just right and the thermal conductivity of a conventional superconductor shows a dramatic enhancement below  $T_c$  is illustrated in Fig. 2. The data refer to measurements of Radosevich and Williams [8] made on NbC. This result, which has been unjustly underpublicized, demonstrates an enhancement in  $\kappa(T)$  for a conventional superconductor which stands unrivaled by even the most spectacular increases in  $\kappa(T)$  observed on the best untwinned single crystals of YBCO.

## 2. ARGUMENTS FOR PHONON ORIGIN OF THE PEAK

Numerous studies have confirmed that the rise in  $\kappa(T)$  below  $T_c$  and the pronounced maximum (Fig. 1) are features common to all hole-doped Cu-O superconductors. Resistivity measurements in conjunction with the Wiedemann-Franz law have clearly established that the lattice thermal conductivity is the dominant contribution, accounting for typically 90% of the normal-state total thermal conductivity in

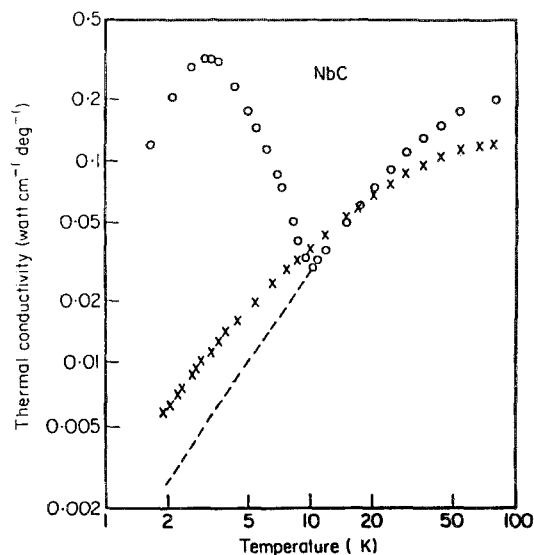


Fig. 2. Thermal conductivity of niobium carbide. Open circles refer to a superconducting NbC<sub>0.96</sub> with  $T_c \approx 10$  K. Crosses represent a nonsuperconducting NbC<sub>0.76</sub>. (Taken from [8].)

sintered samples and at least 50% of the heat conductivity in single crystals. It was thus natural to invoke the BRT theory and explain the rise in  $\kappa(T)$  as resulting from an enhancement of the mean-free path of phonons due to the diminishing influence of phonon-carrier scattering as the carriers undergo condensation. A formal theoretical treatment was carried out in a series of papers by Tewordt and Wölkhausen [9,10] with refinements appropriate to single crystals made by Peacor *et al.* [11]. In its latter form the expression for the total thermal conductivity  $\kappa(T) = \kappa_E(T) + \kappa_p(T)$  contains the lattice thermal conductivity term

$$\kappa_p(T) = \frac{k_B}{2\pi^2 v} \left( \frac{k_B}{\hbar} \right)^3 T^3 \int_0^{\Theta_D/T} dx \times \frac{x^4 e^x}{(e^x - 1)^2} \int_0^1 d\zeta \zeta^{\frac{3}{2}} (1 - \zeta^2) \tau(T, x, \zeta) \quad (1)$$

where

$$\tau^{-1}(T, x, \zeta) = B + D_p T^4 x^4 + D_{sf} T^2 x^2 + ETxg(x, y)(1 - \zeta^2)^{3/2} + UT^4 x^2 \quad (2)$$

In Eq. (2), coefficients  $B$ ,  $D_p$ ,  $D_{sf}$ ,  $E$ , and  $U$ , in that order, refer to the following scattering processes believed to be relevant to the Cu-O perovskite structure: phonon scattering by boundaries, point defects, sheetlike faults, electrons, and other phonons. The term of greatest interest and the one leading to a rise

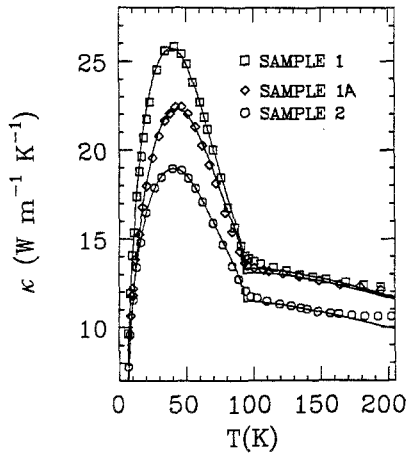


Fig. 3. Experimental data and theoretical curves showing  $\kappa_{ab}(T)$  for three single crystals of YBCO. (Taken from [11].)

in  $\kappa(T)$  below  $T_c$  is the second to the last term in Eq. (2). The function  $g(x, y)$  is the ratio of phonon-electron scattering times in the normal and superconducting states, and its exact form is given in [7]. The dependence of the energy gap enters via the parameter  $y = \Delta(T)/k_B T$ , and extensions of the theory which include the strong-coupling limit for both  $s$ - and  $d$ -wave pairing mechanisms have been derived [10]. Excellent fits with perfectly reasonable parameters, including a sensible value for the electron-phonon coupling constant, exist for both sintered and single-crystal YBCO data. In Fig. 3 we show the fits to the data on high-quality YBCO single crystals from [11]. A particularly noteworthy feature is the high sensitivity of the thermal conductivity to structural damage. Sample 1A is sample 1 after it broke in a cryostat when a large torque was applied in an attempt to rotate a magnetic field during measurements of the thermal conductivity in an intense field. The only change in the fitting procedure is a larger value of the parameter  $D_{sf}$  ( $3.2 \times 10^5 \text{ K}^{-2} \text{ sec}^{-1}$  vs.  $1.7 \times 10^5 \text{ K}^{-2} \text{ sec}^{-1}$ ) which, given the planar nature of the single crystal, reflects the tendency of the crystal to adjust to a large strain by introducing sheetlike faults throughout the structure. A high degree of consistency between the theoretical fit and the microstructure of the crystal extends also to considerations of the point-defect scattering in sample 2 where  $D_p$  is twice as large as for sample 1 ( $723 \text{ K}^{-4} \text{ sec}^{-1}$  vs.  $370 \text{ K}^{-4} \text{ sec}^{-1}$ ), corresponding to an oxygen content of  $\delta = 0.16$  as opposed to  $\delta = 0.1$  which characterizes sample 1. The overall high quality of the fits, the dominance of the phonon thermal conductivity in the normal state, and, above all, a striking similarity with

the behavior of those conventional superconductors which show a rise in  $\kappa(T)$  below  $T_c$  are the main reasons why the phononic origin of the peak has been widely accepted. Originally it was hoped that by carefully exploring the behavior of the thermal conductivity near its peak one might be able to distinguish between different pairing mechanisms. Although the quality of the crystals has improved tremendously and measurement techniques have been highly refined, a definitive claim as to the nature of pairing based on the measurements of the thermal conductivity is not yet possible. The problem resides chiefly in the interpretation of the data and the lack of detailed knowledge concerning several key parameters. For instance, in the phonon picture, the carrier contribution is very sensitive to the specific details of scattering and to the strength of coupling to bosonic excitations, yet neither is well known. Even if one neglects the possibility that the scattering rate of quasiparticles below  $T_c$  might be anomalously low (the key ingredient in explaining the peak as arising entirely due to the carriers), the exact form of how the electronic contribution varies has an influence on the initial rise of  $\kappa(T)$  below  $T_c$ . The assumed two-fluid picture with either phonon or defect limited carrier scattering may not be adequate for Cu-O perovskites.

Criticisms have been raised [12] concerning the phononic origin of the peak although most of these are ambiguous and at best inconclusive. For instance, it has been pointed out that the absence of the peak in the thermal conductivity along the  $c$ -axis is inconsistent with the peak in the basal plane being due to phonons. Since the electrical resistivity in the  $c$ -direction,  $\rho_c$ , is several orders of magnitude larger than the corresponding in-plane resistivity,  $\rho_{ab}$ , the Wiedemann-Franz law dictates that virtually all heat along the  $c$ -axis must be carried by phonons. Therefore, not only should the  $\kappa_c(T)$  be large but the peak in this direction should be quite prominent. The argument is a weak one. It not only ignores the underlying structural anisotropy which leads to the experimentally observed [13-15] range of heat anisotropies  $\kappa_{ab}/\kappa_c \approx 5-17$ , but it also disregards the likelihood that the carrier-phonon interaction is anisotropic, being much weaker in the  $c$ -direction than in the Cu-O plane. It is highly unlikely that in the  $c$ -direction, where the electrical resistivity is some three orders of magnitude larger than the  $ab$ -plane resistivity, the carriers are going to affect the lattice thermal conductivity the same way as they did in the basal plane. It should also be noted that a rise in  $\kappa_c(T)$  below  $T_c$  has been reported in [15], but the data do not extend

to low enough temperatures to see the actual peak in  $\kappa_c(T)$ .

The other objection centers on the oxygen dependence of the thermal conductivity and basically it states that if the phononic picture is right, the insulating YBCO specimens ( $O_6$  configuration), because of the absence of carrier-phonon interaction, should have much higher thermal conductivity than the metallic ( $O_7$  structure) samples. The early data of Hagen *et al.* [13] on single crystals with unspecified oxygen content contradict the above assertion, and this has been used as a “proof” that the phonon picture cannot be correct. The argument is of dubious value on several counts: adding carriers to an insulator has a profound effect not just on phonon scattering but also on the phonon spectrum. It is inconceivable to think that the  $O_6$  and  $O_7$  structures have identical phonon spectra, and therefore one can separate the lattice and the carrier thermal conductivities by simply subtracting the thermal conductivity measured on the oxygen-depleted sample from that of the fully oxygenated sample. Although the data of Hagen *et al.* represent the first measurements on single crystals, neither the quality of the crystals nor the accuracy of measurements are adequate to serve as the benchmark for the thermal conductivity of YBCO. In fact, well-oxygen-controlled studies [16,17] on sintered specimens show very convincingly that the thermal conductivity of oxygen-depleted ( $O_6$ ) samples is much higher than the conductivity of fully oxygenated ( $O_7$ ) specimens. Although it is more difficult to adjust oxygen content in single crystals than in sintered samples, measurements on high-quality crystals with well-controlled oxygen doping are needed to settle the issue once and for all.

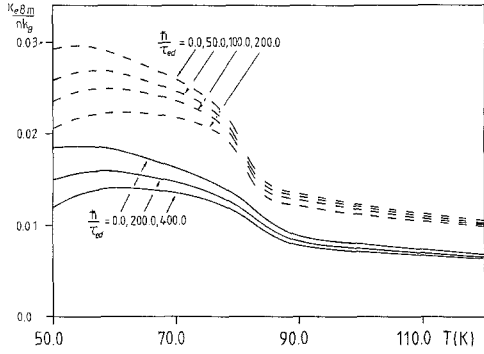
The point we are trying to make here is this: All reliable thermal conductivity investigations on hole-doped Cu-O perovskites, including studies of oxygen dependence, anisotropy, substitution at Y or Cu sites, neutron radiation damage, and the effects of magnetic field, can be explained consistently assuming that the peak below  $T_c$  is due to phonons. It should be recognized, however, that the existing treatment within the BRT theory has a weakness in that the carrier thermal conductivity contribution is described with insufficient accuracy. While an alternative explanation based on the charge-carrier origin of the peak is possible (see the next section), nobody as yet has offered concrete experimental evidence that the phonons are unaffected by the superconducting transition.

### 3. ARGUMENTS FOR THE CARRIER ORIGIN OF THE PEAK

Since the total heat flux consists of two distinct contributions, those due to charge carriers and those due to phonons, one should allow for the possibility that the rise in  $\kappa(T)$  and the peak could, in principle, be associated not with the phonons but with the charge carriers. The problem is that, until recently, there was no readily identifiable mechanism which could lead to an enhancement in the carrier thermal conductivity in a superconducting domain. On the contrary, experience with conventional superconductors led to the belief that the ratio  $\kappa_E^s/\kappa_E^n$  ought to vanish rapidly below  $T_c$  and, anyway, since the carrier contribution in Cu-O perovskites is only a minor fraction of the total heat conductivity, it would have no significant effect on  $\kappa(T)$  below  $T_c$ . This was a perfectly rational point of view at the time when only sintered samples were readily available, because in these structures the carrier contribution in the normal state does not exceed 10% of the total thermal conductivity. The argument was later extended to single crystals even though here the carrier contribution can reach close to 50% of the total thermal conductivity, particularly in untwinned crystals of YBCO.

Motivated by the highly unusual normal-state properties, Varma *et al.* [18] proposed that the high- $T_c$  cuprates might be viewed as marginal Fermi liquid systems where quasiparticles have an infinite lifetime but a logarithmically vanishing spectral weight. Although a detailed account of the thermal conductivity was not given nor was any attempt made to fit the existing data, the marginal Fermi liquid model does allow for a rise in the electronic thermal conductivity below  $T_c$ . This comes from  $\kappa_E(T) \propto C_v l$ , where the specific heat and the mean-free path of the quasiparticles vary as  $C_v \sim e^{-\Delta/T}$  and  $l \sim e^{2\Delta/T}$ . The divergence in  $\kappa_E(T)$  for  $T \rightarrow 0$  is rapidly cut off and the peak in  $\kappa_E(T)$  sets in when, eventually, the defect scattering starts to dominate the behavior of the mean-free path.

While making major contributions to a theoretical formulation of the phononic nature of the thermal conductivity of high- $T_c$  perovskites, Tewordt and coworkers also inquired [19] about the effect of spin and charge fluctuations (in addition to the influence of phonons) on the electronic thermal conductivity. They found an interesting result—a constant electronic contribution above  $T_c$ , and a rising  $\kappa_E(T)$  below  $T_c$ . The latter they associated with a strong pair-breaking effect of spin fluctuations. Furthermore, introduction of a finite amount of elastic electron



**Fig. 4.** Electronic thermal conductivity calculated assuming that the electrons are scattered inelastically by phonons, by spin fluctuations, and by charge fluctuations. The parameter  $\hbar/\tau_{e,d}$  indicates the elastic scattering rate of electrons by point defects. Solid curves correspond to the electron-phonon coupling constant  $\lambda_{ph} = 3.1$ ; the dashed curves are for  $\lambda_{ph} = 2.1$ . (Taken from [19].)

scattering (via point defects) led to a bending down and a peak on  $\kappa_E(T)$ ; see Fig. 4. Although the rise in  $\kappa_E(T)$  and the position of the peak depend on the combined effect of inelastic and elastic electron scattering, the enhancement is substantial ( $\approx 100\%$ ) and the location of the peak falls near  $T_c/2$ . The above work thus represents a viable alternative mechanism to a purely phononic picture of  $\kappa(T)$ , and both  $\kappa_E(T)$  and  $\kappa_p(T)$  should be taken into account when analyzing the temperature dependence of the thermal conductivity below  $T_c$ .

Over the past two years, reports on several key experiments have been published which inquire into the nature of quasiparticle scattering in YBCO. Among these, perhaps the most influential are measurements of microwave surface resistance by Bonn *et al.* [20,21]. The high precision of the experiments and careful analysis of the data indicate that the real part of the microwave conductivity,  $\sigma_1(T)$ , increases rapidly below  $T_c$  and peaks near 35 K with a value some 20 times its value at  $T_c$ . By reanalyzing the best experimental data for the temperature dependence of the penetration depth,  $\lambda(T)$ , Bonn and colleagues arrive at a surprisingly simple form for the normal fluid fraction,  $x_n(t) = t^2$  with  $t = T/T_c$ , which holds throughout the domain of superconductivity. The power-law dependence of  $x_n(t)$  at very low temperatures, differs sharply, of course, from the BCS-like activated behavior and points to the presence of nodes in the gap or to an excitation spectrum which is gapless. Since the frequencies used in the microwave experiments are low enough so that  $\omega\tau \ll 1$ , to a good approximation one can write  $\sigma_1(T) \propto x_n(T)\tau(T)$ . With  $x_n(T)$  given by a quadratic function, Bonn and

coworkers are led to the inescapable conclusion that large increases in the real part of the conductivity that they measure are the result of a dramatically enhanced relaxation time of quasiparticles below  $T_c$ . For instance, between  $T_c$  and 15 K they estimate a 400-fold increase in  $\tau(T)$ . As a likely agent responsible for a high scattering rate above  $T_c$  and its rapid demise below  $T_c$ , Bonn *et al.* favor antiferromagnetic spin fluctuations.

With such large enhancements in the electrical conductivity of the quasiparticles, one has a mechanism on hand which might yield a correspondingly large increase in the thermal conductivity purely because these same quasiparticles will participate in the transport of heat. This is essentially the approach taken by Yu *et al.* [12] in their recent evaluation of the thermal conductivity of untwinned YBCO crystals.

By subtracting the phonon contribution from the total thermal conductivity, they proceed to analyze the resulting carrier contribution in the superconducting state using the formula derived independently by Kadanoff and Martin and by Tewordt [22]

$$\kappa_E^s = \frac{1}{2k_B T^2 m} \int d^3p \frac{p_x^2 \varepsilon_p^2}{\Gamma} \operatorname{sech}^2 \frac{E_p}{2k_B T} \approx \frac{\phi}{\Gamma} \quad (3)$$

Here  $E_p = (\varepsilon_p^2 + \Delta_p^2)^{1/2}$ , where  $\varepsilon_p$  is the normal-state dispersion and  $\Delta_p$  the superconducting gap. The gap-to- $T_c$  ratio  $g$  is taken as  $g = \max[\Delta(k, T=0)]/2k_B T_c$  and  $\phi$  is evaluated for both  $s$ -wave and  $d$ -wave pairing states. The scattering rate  $\Gamma$  is written as  $\Gamma \sim (T/T_c)^n + w_i$ , implying a power law dependence augmented by a constant term  $w_i$  that stands for the residual scattering rate due to impurities. Parameters  $n$  and  $w_i$  are then varied to obtain the best fit to the data for  $\kappa_E^s$ . Yu *et al.* find that, independent of the gap ratio  $g$ , a  $d$ -wave pairing state fits best, and the relaxation rate varies as the fourth power of the temperature. Such a fast temperature dependence is in qualitative accord with the rapid decrease in the scattering rate observed by Bonn *et al.*, although the actual functional form differs from the exponential dependence ( $\Gamma \propto e^{T/T_0}$ ) observed in the microwave surface resistance. The obviously problematic issue in the work of Yu *et al.* is a total neglect of any change in the phonon thermal conductivity at  $T_c$ . In other words, the rise in  $\kappa(T)$  and the ensuing peak are viewed as arising solely due to the quasiparticles. This is probably unrealistic, and such an omission in turn influences conclusions regarding the dynamics of quasiparticles, including the temperature dependence of their relaxation rate.

#### 4. RELAXATION RATE FROM ULTRAFAST LASER STUDIES

Recent advances in the development of ultrafast lasers have paved the way for studies of out-of-equilibrium carrier distributions directly in the time domain. With laser pulses of 100 fs or less, one can, using a typical pump-probe configuration, monitor how out-of-equilibrium carriers relax and determine the characteristic time. This technique was initially found very successful in investigations of relaxation times of noble metals [23], while we were the first to adopt it in our temperature-dependent studies of the relaxation dynamics in films of high- $T_c$  superconductors [24]. The details concerning the experimental setup can be found in the original papers. Briefly, a laser beam is split into two components, a pump and a probe, the former (and more intense) generating an out-of-equilibrium carrier distribution as the photons are absorbed in the film. The probe, which can be variably delayed, tracks the evolution of the carrier temperature by monitoring changes in either the reflectivity or the transmissivity of the film as a function of the time delay. By mounting the film in an optical cryostat, one can study relaxation dynamics over a wide temperature range.

In Fig. 5 we show the temperature dependence of the relaxation time for films of YBCO and BISCO. A remarkable feature of the data is a sudden and dramatic rise in the relaxation time once the films become superconducting. For comparison, we also show that for a fully oxygen-depleted insulating  $O_6$  phase the relaxation time remains small and monotonic. These results convey a similar message as those from the surface-resistance studies of Bonn *et al.*

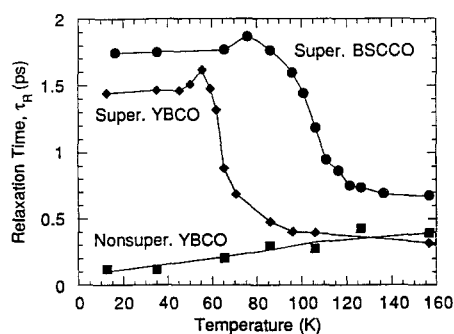


Fig. 5. Relaxation times from femtosecond pump-probe laser studies of a superconducting film of YBCO ( $\blacklozenge$ ), a nonsuperconducting film of YBCO ( $\blacksquare$ ), and a superconducting film of BISCO ( $\bullet$ ). (Taken from [24].)

Namely, the relaxation rate of quasiparticles dramatically decreases as the sample becomes superconducting. The results weigh in favor of the carrier thermal conductivity contribution as an important ingredient in the rise of  $\kappa(T)$  and the occurrence of the peak below  $T_c$ .

#### 5. CONCLUSIONS

A vast amount of data have been compiled on the behavior of the thermal conductivity in high- $T_c$  superconductors of various structural forms and under diverse experimental conditions. There is no dispute regarding the actual experimental results. On the contrary, the reports convey a remarkable unanimity concerning a “universal” behavior within the main family of high- $T_c$  superconductors, the hole-doped Cu-O perovskites. The issue of considerable contention deals with the origin of the characteristic features in the thermal conductivity—its rise below  $T_c$  and the peak near  $T_c/2$ . Similarity with the behavior of those conventional superconductors which display identical features, albeit at much lower temperatures, would suggest that phonons are the principal source behind the enhancement of the thermal conductivity at  $T_c$ . There is adequate and plausible theoretical description which supports this point of view. The difficulty with this interpretation arises because the dynamics of the charge carriers in high- $T_c$  perovskites may differ substantially from the assumed BCS-like behavior and the carrier contribution to the heat flux could thus be underestimated. The emerging experimental scene clearly demonstrates the importance of the quasiparticle transport below  $T_c$  and it is prudent to allow for the possibility that the enhancement and the peak in  $\kappa(T)$  are, at least partly, tied to the behavior of the quasiparticles.

The difficulty in making an unambiguous choice between the two competing interpretations rests in the fact that the charge carriers and phonons contribute roughly equally (in single crystals) to the heat transport in the normal state, and the physical processes which give rise to enhancements in either  $\kappa_p(T)$  or  $\kappa_E(T)$  below  $T_c$  have rather strong temperature dependences which lead to peaks in the thermal conductivity at virtually the same temperatures.

There are experiments which might help to resolve the issue, and a study of the thermal conductivity of neutron-damaged YBCO crystals accompanied by measurements of the relaxation rate

using ultrafast lasers might be one such experiment. It is well known from previous investigations [25] that the peak in  $\kappa(T)$  depends very sensitively on the neutron fluence, and a rather modest level of radiation damage which does not alter  $T_c$  significantly completely washes out the peak. It would be of interest to see whether the quasiparticle relaxation time determined by pump-probe laser studies on the same samples is also dramatically altered. Although an affirmative answer to the above would not be a conclusive outcome, the negative answer, i.e., a substantially unchanged quasiparticle relaxation rate, would signal that the peak in  $\kappa(T)$  has little to do with the quasiparticle transport.

The issue concerning the origin of the rise and the peak in the thermal conductivity of hole-doped Cu-O superconductors is important. While currently there is no definitive proof that either phonons or charge carriers are the sole reason for these characteristic features in  $\kappa(T)$ , there are clear indications that quasiparticle transport is highly unusual and the quasiparticles may play an important role in the energy flow below  $T_c$ . It is essential that we fully understand the role which phonons and charge carriers play in the heat transport of high- $T_c$  superconductors because this information bears on a viable mechanism of superconductivity.

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