Phonon thermal conductivity limited by electron scattering in amorphous $Zr_{70}Cu_{30}$

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The low-temperature thermal conductivity of amorphous $Zr_{70}Cu_{30}$ is investigated. The lattice thermal conductivity as limited by electrons in the superconducting state has been obtained at temperatures close to T_c. This has allowed for the first time the verification of the temperature dependence of the phonon conductivity as predicted by Bardeen, Rickayzen, and Tewordt. The analysis of the data in the normal and superconducting state shows that Matthiessen's rule can be used in amorphous metals to separate the different scattering processes.

Heat conduction in metals is due to the parallel contribution of electrons and phonons. For crystalline metals the electron contribution prevails over that of the phonon system and measurement of the lattice thermal conductivity is difficult. Amorphous metals are homogeneous systems with an extremely short elastic electron mean free path (EMFP) (of the order of interatomic distances) and, as a consequence, the electron contribution to heat conduction is reduced below that of phonons. Amorphous metals are thus convenient systems to study those interactions which determine the phonon mean free path (PMFP) in the heat-transport process. In particular, thermal conductivity measurements in amorphous superconductors at low reduced temperatures have shown that phonon interactions with low-energy excitations are similar to those in insulators.[1] The thermal conductivity shows structure at the superconducting transition, indicating that the electronic excitations contribute[1,2] to the thermal conduction either as heat carriers or as phonon scattering centers.

The thermal conductivity can be expressed quite generally as

$$\kappa=\kappa_g+\kappa_e=\left[\frac{1}{\kappa_g^e}+\frac{1}{\kappa_g^d}\right]^{-1}+\kappa_e \ , \qquad (1)$$

where κ_g and κ_e are the phonon and electron contribution, whereas κ_g^e and κ_g^d indicate the phonon conductivity as limited by electron and "disorder" scattering. When specifying the normal or superconducting conduction we will add the subscript n or s, i.e., κ_n, κ_{gs}.

Quite recently, Löhneysen *et al.* have measured[1] the thermal conductivity of amorphous $Zr_{70}Cu_{30}$ between 0.3 and 10 K. The measurements at temperatures well below $T_c \simeq 2.7$ K were used to determine $\kappa=\kappa_g^d$. By applying a magnetic field these authors were able to determine κ_n in the whole temperature range. The electronic contribution κ_e was obtained from the Wiedemann-Franz law $\kappa_e=(L/\rho_0)T$, where ρ_0 is the residual resistivity and $L=2.45\times10^{-8}$ W Ω K^{-2}. Using the measured κ_g^d they were able to determine κ_{gn}^e in the low-temperature range ($T<1$ K). In this way Löhneysen *et al.* showed that κ_{gn}^e is proportional to temperature as predicted by Pippard[3] in his analysis of the ultrasonic attenuation by conduction electrons when the EMFP is shorter than the phonon wavelength. Extrapolating this result to $T>1$ K they obtained κ_g^d in the whole temperature range.

The purpose of this work is to use a similar procedure to analyze our data in order to investigate the behavior of the thermal conductivity at temperatures close to T_c. Below T_c the density of electronic excitations changes exponentially with temperature, inducing a strong temperature dependence in κ_{gs}^e and κ_{es}. Bardeen, Rickayzen, and Tewordt (BRT) calculated[4] the temperature dependence of κ_{es}/κ_{en} and $\kappa_{gs}^e/\kappa_{gn}^e$ in the weak-coupling limit. This theory has been shown[2–5] to correctly describe the temperature dependence of κ_{es}/κ_{en} either in the impurity or lattice scattering limit. On the other hand, the experimental difficulties mentioned above have precluded a proper verification of the theoretical results when applied to $\kappa_{gs}^e/\kappa_{gn}^e$.

We have measured the thermal conductivity of $Zr_{70}Cu_{30}$ in the normal and superconducting state. Using these data and those of Ref. 1 for κ_{gn}^e, we obtained κ_{gs}^e. Our results show that the ratio $\kappa_{gs}^e/\kappa_{gn}^e$ follows the temperature dependence predicted by BRT.

The thermal conductivity data between 0.5 and 7 K are shown in Fig. 1. These results agree within 5% with those of Ref. 1. Details on the experimental technique and sample preparation can be seen in Ref. 6.

The residual electrical resistivity ρ_0 for this sample was 180 $\mu\Omega$ cm, 20% lower than that of Ref. 1. Considering the different origin of the alloys and a 10% error in the determination of the geometrical factor, the agreement between both results can be taken to be fair. The first step in our data analysis is to obtain κ_g. This is accomplished using the Wiedemann-Franz law together with the BRT expression for

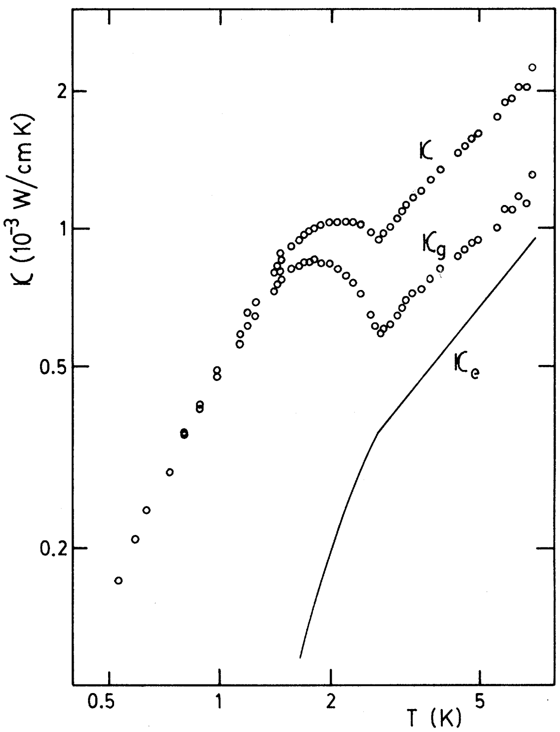

FIG. 1. Total, phonon, and electron thermal conductivity, κ, κ_g, and κ_e, respectively, as a function of temperature for $Zr_{70}Cu_{30}$ amorphous system.

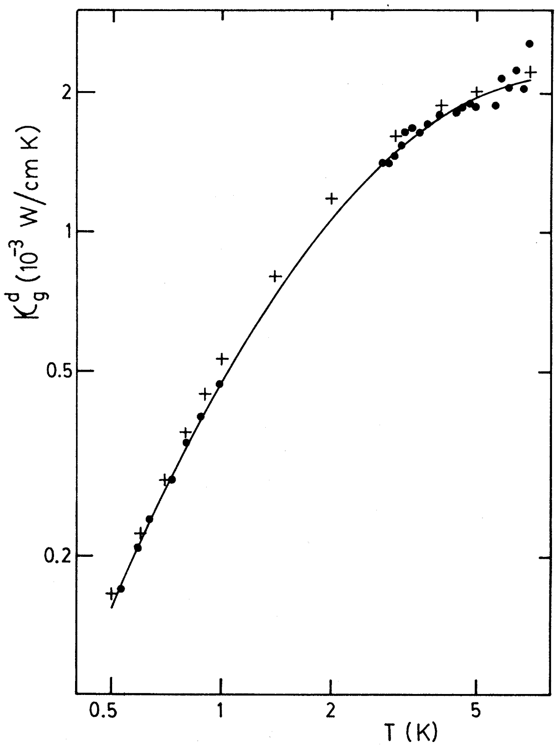

FIG. 2. Phonon thermal conductivity κ_g^d limited by "disorder," as a function of temperature. ● calculated points as explained in text; + points from Ref. 1; full curve is a smooth fitting.

κ_{es}/κ_{en}, generalized in the elastic scattering limit by Mrstik and Ginsberg[7] for strong-coupling superconductors and taking the BCS temperature dependence for $\Delta(T)/\Delta(0)$.

Since specific-heat measurements[8] in a similar system have shown that its superconducting state is characterized by a gap parameter $2\Delta(0)/kT_c = 3.8$, we have introduced this value in the BRT expression. The results for κ_e and κ_g are shown in Fig. 1. At temperatures below 1 K we have $\kappa = \kappa_g^d$ in agreement with Ref. 1. To obtain κ_g^d at temperatures higher than T_c we have used κ_g^e from Ref. 1, assuming the validity of Matthiessen's rule. Since in the short EMFP limit κ_g^e is proportional[3,9] to ρ_0, we have taken our lower electrical resistivity into account by decreasing the κ_{gn}^e given in Ref. 1 by 20%. The results are shown in Fig. 2 where the full curve is a smooth fitting of the experimental points above and below T_c. Data obtained from Ref. 1 for κ_g^d are included in the same figure. Using the κ_g and κ_g^d values, we obtained κ_g^e, as shown in Fig. 3.

The phonon conductivity limited by disorder can now be compared with results typical of amorphous insulators. We have used the Zaitlin and Anderson[10] simplified expression for the thermal conductivity due to phonons interacting with two level systems (TLS). In this theory the phonon scattering at low temperatures (in our case $T < 1$ K) is dominated by the resonant interaction between phonons and the TLS. The theoretical fitting[11] to our κ_g^d determines the coefficients $A = 1.95 \times 10^{-3}$ cm K and $\gamma = 0.08$ K^{-2}; the notation and the definition of the coefficients are those given in Ref. 10. While A is a parameter related with the sound velocity and the density of tunneling states, γ is a measure of its energy dependence. A nonzero value of γ is necessary to obtain the experimental slope ($\simeq 1.7$) in the low-temperature region. It is interesting to see that both coefficients have an absolute value quite similar to that for insulators. In Ref. 10 a relaxation term was introduced[10] (with β as a new parameter) to take into account the plateau region. It is interesting to note that to fit the theory to our data we need to use a coefficient $\beta \simeq 2 \times 10^{-2}$ K^{-2}. This value is 20 times larger than those typical for insulators. If we choose $\beta \leq 1 \times 10^{-2}$ K^{-2} the increase in the absolute value and the slope of κ_g^d is enough to observe clear deviation from the experimental data. It is important to remark that the theoretical fitting below 2 K is not sensitive to β. Nevertheless, the meaning of the high value here obtained is not thoroughly understood. It could be that the strong relaxation effect in the metal can be due to the fact that the relaxation time of the TLS is reduced[12] by the presence of the electrons. Another less profound reason can be that the β we have chosen is appropriate to fit the experimental data only in our available range of tempera-

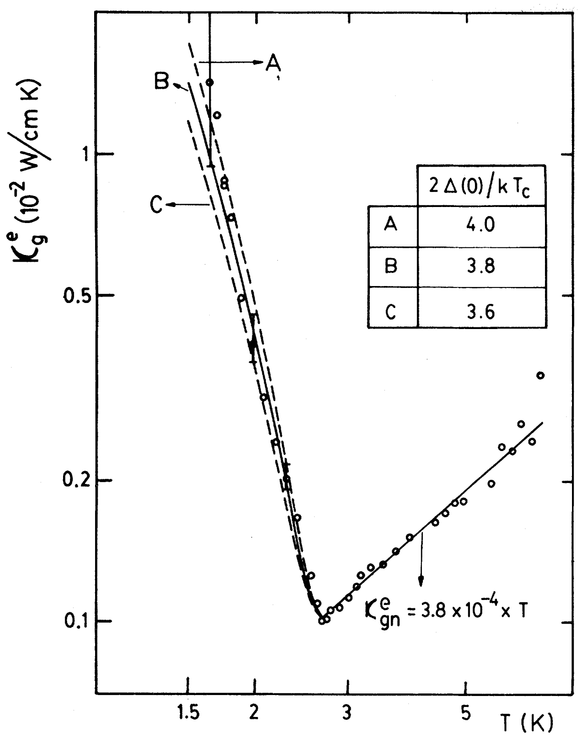

FIG. 3. Phonon thermal conductivity κ_g^e limited by electrons as a function of temperature T. Full and dashed lines ($T \leq T_c$) were calculated using the BRT formula for three different gap parameters, κ_{gn}^e obtained as explained in text, and the BCS temperature dependence for $\Delta(T)/\Delta(0)$. Bars indicate the maximum estimated error.

ture and is not the correct one for an extended temperature range. This effect should be investigated further since it could lend support to the idea that the plateau in the thermal conductivity is due to relaxation.

The theoretical fitting to the κ_{gs}^e data in Fig. 3 has been made calculating the ratio $\kappa_{gs}^e/\kappa_{gn}^e$ from the BRT theory (see below), where the gap at $T=0$, $\Delta(0)$ is left as a free parameter. It is quite clear that the best fit is for $2\Delta(0)/kT_c \simeq 3.8$, the same value used to calculate the electron contribution κ_{es}. It should be pointed out that if we change $2\Delta(0)/kT_c$ to 4.0 or 3.6 to obtain κ_{es}, the data points shift only one-third the error bars indicated in Fig. 3.

TABLE I. The theoretical ratio $\kappa_{gs}^e/\kappa_{gn}^e$ as a function of $\Delta(T)/kT$.

$\Delta(T)/kT$	$\kappa_{gs}^e/\kappa_{gn}^e$	$\Delta(T)/kT$	$\kappa_{gs}^e/\kappa_{gn}^e$
0	1.000	2.2	6.446
0.2	1.000	2.4	8.495
0.4	1.003	2.6	11.26
0.6	1.033	2.8	14.56
0.8	1.114	3.0	18.66
1.0	1.281	3.2	23.73
1.2	1.570	3.4	30.02
1.4	2.024	3.6	37.75
1.6	2.687	3.8	47.39
1.8	3.608	4.0	59.29
2.0	4.841	4.2	74.21

This result shows that the theoretical BRT expression for $\kappa_{gs}^e/\kappa_{gn}^e$ is verified when the EMFP is smaller than the phonon wavelength, and gives further support to the use of expression (1) to represent the different contributions to the thermal conduction in amorphous materials.

Since no tabulation[13] appears to be readily available for the BRT expression of $\kappa_{gs}^e/\kappa_{gn}^e$, in particular, for reduced temperature greater than 0.7, we present below, in Table I, this ratio as a function of $\Delta(T)/kT$. With the knowledge of $2\Delta(0)/kT_c$ and the temperature dependence of $\Delta(T)/\Delta(0)$, it is possible to interpolate in order to obtain $\kappa_{gs}^e/\kappa_{gn}^e$ as a function of T.

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[13]A. C. Anderson and S. O'Hara, J. Low Temp. Phys. 15, 323 (1974) have published a table with the ratio $\kappa_{gs}^e/\kappa_{gn}^e$, as a function of the reduced temperature t for different gap parameters. We found that those values differ from the exact values up to 30% for $t=0.8$, possibly due to use of the approximate BRT formula for the low-temperature region. For $t\leq0.5$, the differences are less than 1%.