Phonon scattering by electrons in noncrystalline alloys

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(Received 4 June 1979)

The low-temperature thermal conductivity of noncrystalline $Zr_x Be_{1-x}$ ($x \sim 0.5$) has been measured between 0.3 and 4 K. A pronounced decrease of the thermal conductivity near the superconducting transition temperature ($T_c \sim 2 \text{ K}$) has been observed. In the entire temperature range studied, the thermal conductivity can be explained with the model that the Debye phonon mean free path is determined by scattering through the electrons and the intrinsic states which appear to be characteristic for all amorphous solids. The observed scattering of the phonons by the electrons can be explained with the theories developed for highly disordered crystalline metals and for superconductors near T_c .

I. INTRODUCTION

Phonon scattering in amorphous dielectric solids has been a subject of intensive investigation since the first observation by Zeller and Pohl¹ that the low-temperature thermal conductivity of these materials is an almost universal function of temperature and is fairly independent of the material, a fact since then confirmed by many investigations.²⁻⁴ It is now generally believed that the phonons which carry the heat in these solids are scattered by some intrinsic defects which are characteristic for the amorphous state. It is also widely believed that these states are associated with some tunneling motion of atoms or groups of atoms in the amorphous structure.⁵ However, neither is the nature of these defects known nor is it understood why they scatter the phonons almost equally as strong in every amorphous dielectric. One remarkable point about amorphous dielectrics is that chemical impurities appear to have no effect on the thermal conductivity. This is in contrast to crystalline solids where certain kinds of impurities produce low-lying tunneling motions, and resonant phonon scattering from these levels can reduce the thermal conductivity below that of glasses.1

Matey and Anderson⁶ have studied the heat conduction below 100 K in a number of noncrystalline metals including one superconducting alloy. After subtracting the estimated electronic part of the thermal conductivity, they found that below 1 K the phonon heat conductivity is very similar to that of dielectric amorphous solids. The conclusion was that the intrinsic phonon-scattering centers present in other amorphous dielectrics are also present in metallic glasses. Matey and Anderson did not find any evidence of scattering of phonons by electrons. A study of the low-temperature thermal conductivity of superconducting noncrystalline $Zr_{0.7}Pd_{0.3}$ by Graebner *et al.*⁷ also showed the existence of the universal amorphous thermal conductivity below the superconducting transition temperature T_c . Near T_c , however, the thermal conductivity was depressed. This was explained as resulting from electron-phonon interaction and provided the first evidence of the occurrence of a phonon-scattering mechanism in the bulk of a glass *in addition* to that associated with the intrinsic defects. Previously, the only scattering mechanism by which the thermal conductivity of a glass could be altered had been internal and external surface scattering.⁴

In the present investigation we have studied the thermal conductivity in several superconducting noncrystalline alloys. We will show that the phonon scattering by the electrons can be explained at least semiquantitatively with the theory developed for highly disordered crystalline solids. The superconducting properties of the Zr-Be alloys chosen for this work have been studied in detail by Hasegawa etal.⁸ Their Debye temperatures are expected to be low. Furthermore, these alloys have a low average ionic mass, because of a large atomic percentage of beryllium. It will be shown in the discussion that due to the above two factors the influence of electrons on the phonon thermal conductivity should be relatively large, and hence more readily observable than in the previously studied alloys.6.7

II. EXPERIMENTAL TECHNIQUES

The alloys were made by quenching the melt into the form of long uniform strips about 30 μ m thick and 1-2 mm wide. The quenching rate was 10⁶ K/sec. The uncertainty in composition is ~1%. The low-temperature measurements were per-

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FIG. 1. Experimental arrangement used for the thermal conductivity measurements. The niobium titanium filaments were kept straight and were arranged in a fashion that would reduce vibrations of the sample. Ge 1 and 2 are the primary germanium thermometers. T_1 and T_2 are secondary carbon thermometers. They were calibrated in each run against Ge 1 and 2.

formed in a dilution refrigerator. The superconducting transition temperature T_c was measured by using a four-probe method. The temperature at which the resistance was 50% of the normal state was defined as T_c , and the transition width was taken between 90% and 10% of the normal state resistance. Measurements were obtained both during heating and cooling to ensure that the thermometer and sample were in good thermal contact.

The thermal conductivity was measured using a two-thermometer, one-heater technique, shown in Fig. 1. The samples were 2-3 cm long, the heater was a commercial metal film resistor. Thermometers T_1 and T_2 were carbon resistors made by slicing off pieces from a 220- Ω Speer carbon resistor. They were calibrated in each run against two germanium thermometers, one for T < 1 K and one for T > 1 K. Owing to the small thermal conductance of the samples, special care was taken to prevent heat drain through the electrical leads. A few filaments of $7-\mu$ m-diameter Nb-Ti wire, obtained by etching away the Cu-Ni matrix of a filamentary superconducting magnet wire were used for this purpose.⁹ The leads were cut to proper size so that they remained tight and did not produce vibrational heating. We estimated that the total heat loss through the leads was less than 2% of the total heat input.

The uncertainty as to the absolute value of the thermal conductivity resulted mainly from the nonuniformity of the thickness of samples. We estimate this uncertainty to be $\pm 10\%$.

III. RESULTS AND DISCUSSION

Figure 2 shows the superconducting transition of the three samples studied. For two of the samples the transition is quite sharp, with a width of ~0.02 °K indicating chemical homogeneity. The



FIG. 2. Superconducting transition of $\operatorname{Zr}_{x}\operatorname{Be}_{i-x}$. ρ_{s}/ρ_{n} is the resistivity ratio as a function of temperature. Samples as described in Table I.

transition of the third sample is not quite that sharp, ~0.1 K, implying the presence of some chemical inhomogeneity.¹⁰ Alloy Nos. 2 and 3 have the same nominal composition but different T_c . This can be explained by the uncertainty in composition. (The data are summarized in Table I.) Figure 3 shows the thermal conductivity K(T). Below 1 K, all samples have the same thermal conductivity within the experimental uncertainty. A fit to the relation $K = \alpha T^{\beta}$ results in the values of α and β listed in Table II. At T > 1 K the drop in the thermal conductivity shows that electrons are becoming involved in the scattering process.

To analyze our data we start from the fact that the thermal conductivity in metals generally contains an electronic part and a lattice part. For our alloys which have residual resistivity ratios ~1 (see Table I) one can get a fairly accurate estimate of the electronic thermal conductivity by using the Wiedemann-Franz law,¹¹ with the result that the electronic thermal conductivity contributes only 10–15% to the total observed thermal conductivity above T_c . Below T_c , the contribution decreases further with decreasing temperature. Hence, to a good approximation, we may consider the phonons as the sole carriers of heat in these alloys.

In the temperature range studied, the phonons are scattered by the lattice, by the electrons, and by the sample boundaries. From the measured thermal conductivity and the estimated speed of sound it can be estimated that boundary scattering has a negligible effect in this temperature range. Hence we may write

$$K^{-1}(T) = K^{-1}_{\text{el-ph}}(T) + K^{-1}_{\text{glass}}(T) , \qquad (1)$$

where $K_{elph}^{-1}(T)$ and $K_{glass}^{-1}(T)$ stand for the resistivity resulting from scattering of phonons by electrons and by the intrinsic glassy scatterers, re-

Sample no.	Nominal composition	Thickness (µm)	ρ(4.2 K) (μΩ cm)	$\frac{\rho(4.2 \text{ K})}{\rho(300 \text{ K})}$	Т _с (К)	Δ <i>T_c</i> (K)
1	$\operatorname{Zr}_{0,6}\operatorname{B}_{0,4}$	39.4	290	0.897	2.016	0.1
2	$Zr_{0.55}Be_{0.45}$	29.9	250	1.09	1.802	0.022
3	$\operatorname{Zr}_{0.55}\operatorname{Be}_{0.45}$	32.3	281	0.98	1.704	0.015

TABLE I. Summary of the data on superconducting transition temperatures and other properties of the alloys used (ρ : electrical resistivity).

spectively.

For crystalline superconductors it is known that phonon scattering and hence $K_{el-ph}^{-1}(T)$ decrease very rapidly with temperature below T_c , due to the condensation of electrons into the ground state.¹² Therefore even if $K_{el-ph}^{-1}(T)$ and $K_{glass}^{-1}(T)$ are comparable at or near T_c , we expect that at $T \ll T_c$, $K_{el-ph}^{-1}(T) \ll K_{glass}^{-1}(T)$ and hence, according to Eq. (1), $K(T) \approx K_{glass}(T)$.

From 0.3 to 0.8 K, i.e., $T \ll T_c$, all three samples have a thermal conductivity $K(T) = \alpha T^{\beta}$, where β lies in the range found for all dielectric glasses $(1.8 < \beta < 1.9)$. It is noteworthy that neither the very fast quenching rate (10⁶ K/sec) nor the chemical inhomogeneity suspected in one of the samples has any influence on the thermal conductivity in these noncrystalline metals. The values of α lie within



FIG. 3. Thermal conductivity of the Zr-Be alloys. The straight lines through the low-temperature part of the data are best fitted to the relation αT^{β} (see Table II). The numbered arrows indicate the superconducting transition temperatures as observed through electrical resistivity measurements.

the upper region of the range found in dielectric glasses.⁴

The drop of the thermal conductivity near T_c can be interpreted as the onset of electron-phonon scattering. In order to obtain quantitative information about the phonon scattering by the electrons, one has to extract $K_{el-ph}(T)$ from the experimentally measured K(T). This requires the knowledge of $K_{\text{glass}}(T)$.

We estimated $K_{\text{glass}}(T)$, for ZrBe alloys by comparison to K_{glass} for other glasses, we have plotted in Fig. 4 one representative curve from our data along with other data on amorphous solids. Below 1 K all glasses have a thermal conductivity varying almost as $T^{1.85}$. In order to see whether they have similar shapes above 1 K, we have scaled all the curves so that the low-temperature part of all of them match with our data, see Fig. 5. We find that $\text{Ti}_{0.5}\text{Zr}_{0.1}\text{Be}_{0.4}$, Ni-P, Fe-P, and also SiO₂ have very similar conductivities (all of them lie within the shaded band) even in the plateau region. The exception, Pd-Si-Cu, will be explained below as the result of electron-phonon interaction.

Inspection of Fig. 5 suggests a simple way of extrapolating K_{glass} (T) above 1 K: It seems likely that K_{glass} for our alloys has the same form as for all the other glasses and thus lies within the shaded band. With this assumption we calculated $K_{el-ph}(T)$ for our alloys using Eq. (1). The results are shown in Fig. 6. The bars indicate the uncertainty in $K_{el-ph}(T)$ due to the uncertainty in $K_{glass}(T)$, determined by the width of the shaded band in Fig. 5. Also shown in Fig. 6 is $K_{el-ph}(T)$ for $Zr_{0.7}Pd_{0.3}$, which we obtained by the same procedure.

In the following, we divide our discussion into two sections, one for the normal state $(T > T_c)$, the other for the superconducting state $(T < T_c)$.

(a) For the normal state, $K_{el-ph}(T)$ is found to be linear in T (Fig. 6). A fit of $K_{el-ph}(T)$ to the relation γT^{δ} has been obtained for several alloys. Values of γ and δ are listed in Table II. To explain this linear temperature dependence we refer to the work by Lindenfeld *et al.*¹³ and by Morton.¹⁴ The temperature dependence of the phonon thermal conductivity as limited by electron scattering $K_{el-ph}(T)$ in a normal metal depends critically on the relative magnitude of the electron mean free path l_e

House and a second second second	$K(T < 1 \text{ K}) = \alpha T^{\beta}$		$K_{\rm el-nh}(T > T_c) = \gamma T_c$	rδ		
Sample	α	β	$\gamma_{\rm el-ph}^{\rm a}$	δ	γ_{el-ph} [from Eq. (3)]	
1	$1.29 imes10^{-3}$	1.85	$(7.64 \pm 0.36) \times 10^{-4}$	0.94	1×10^{-3}	
2	$1.14 imes10^{-3}$	1.90				
3	$1.21 imes10^{-3}$	1.86	$(7.59 \pm 0.39) \times 10^{-4}$	1.09		
$\operatorname{Zr}_{0.7}\operatorname{Pd}_{0.3}$	Ref. 7	Ref.7	$(1.59\pm0.19) imes10$ $^{-3}$ ^b	1.3	$2.4 - 3.6 \times 10^{-3}$	
Pd-Si-Cu	Ref. 6	Ref.6	$\sim 6 imes 10^{-3 \text{ b}}$	~ 1	$9.5 imes10^{-3}$	
Ni-P	Ref. 6				$15.7 imes10^{-3}$	

TABLE II. Summary of the thermal conductivity data and expected phonon thermal conductivity in the normal state as limited by scattering from electrons. Values of K and K_{el-m} are given in the units $W \operatorname{cm}^{-1} K^{-1}$.

^a The uncertainty involved is due to the uncertainty in estimating K_{glass} (see text). ^b Calculated using data of Refs. 6 and 7.

and the phonon wavelength. We estimate the electron mean free path l_e from the electrical resistivity at 4.2 K by using a free-electron model.¹⁵ For our Zr-Be alloys, assuming 4 electrons per atom from zirconium and 2 electrons per atom from beryllium, we obtain $l_e \sim 1.4$ Å. For $Zr_{0.7}Pd_{0.3}$, the number of electrons per atom of palladium is uncertain. If we use all the 10-*d* electrons, we calculate $l_e \sim 1.2$ Å. If we assume, following a suggestion of Nagel and Tauc,¹⁶ that palladium contributes one electron per atom, we obtain $l_e \sim 1.8$ Å. If we take these two electron numbers as extreme



FIG. 4. Comparison of the thermal conductivities of various amorphous solids. O: our data on sample no. 1; $\bullet: Zr_{0.7}Pd_{0.3}$ after Ref. 7; Fe-P, Ni-P, Pd-Si-Cu after Ref. 6; SiO₂ after Ref. 1.

cases, we expect $l_e \sim 1.2-1.8$ Å. We estimate the phonon wave vector $q_{\rm ph}$ with the dominant phonon approximation $(q_{\rm ph} \approx 3kT/\hbar V_D)$, where V_D = Debye sound velocity). Taking $\theta_D = 180$ K (Ref. 7) for the $Zr_{0.7}Pd_{0.3}$ alloy, we calculate $q_{\rm ph} l_e \sim [(3-4) \times 10^{-2}/$ K]T, and for $Zr_{0.6}Be_{0.4}$ we obtain $q_{\rm ph} l_e \sim (4 \times 10^{-2}/\text{K})T$ with $\theta_D = 75$ K (Ref. 17). Thus in our temperature range of interest, $q_{\rm ph} l_e \ll 1$. Since $q_{\rm ph} l_e$ is very small, we expect that the strong electron-phonon interaction required for superconductivity will not affect the inequality $q_{\rm ph} l_e \ll 1$. In this limit, Morton¹⁴ gives the following expression for $K_{\rm el-ph}(T)$:

$$K_{\text{el-ph}} (T) = \frac{5}{3} \left(\frac{1}{24} + \frac{1}{9} \right) \frac{\hbar^2 k_B^2 M V_D \theta_D}{\chi^3 [m^{*3/2} (\frac{2}{3} E_F^*)^{5/2} \tau_e]} \left(\frac{T}{\theta_D} \right) J_2(\theta_D/T) ,$$
(2)



FIG. 5. Thermal conductivities of noncrystalline metal alloys (and of silica) plotted in such a way that they coincide below $\sim 1 \text{ K}$. Ti_{0.5}Zr_{0.1}Be_{0.4}, Ni-P, Fe-P, and Pd-Si-Cu after Ref. 6; Zr_{0.6}Be_{0.4}, this work (sample no. 1); SiO₂, after Ref. 1.



FIG. 6. Thermal conductivity limited by electronphonon scattering $K_{el-ph}(T)$ determined from the measured thermal conductivity K(T) as described in the text [see Eq. (1)]. The bars indicate the uncertainty involved in extracting K_{el-ph} from the K(T) due to the uncertainty K_{glass} . The data for alloy 1 and alloy 3 have similar uncertainty, therefore, bars are shown only in one. The data for alloy 2 is very close to that of alloy 3 so only alloy 3 has been shown. T_c indicate the superconducting transition temperatures. The thermal conductivity data for $Zr_{0.7}Pd_{0.3}$ used for this calculation were kindly provided by J. E. Graebner (Ref. 7).

where

$$J_2(y) = \int_0^y y^2 e^y (e^y - 1)^{-2} dy.$$

In the limit of low temperatures $(\theta_D/T \gg 1)$, $J_2(y) \rightarrow 3.25$ (*M* = ionic mass, x^3 = ionic volume, m^* = dressed electronic mass, E_F^* = Fermi energy, and τ_e = relation time of the electrons). Equation (2) contains contributions from both the longitudinal phonons and from the transverse phonons. By using a free-electron model, we can simplify Eq. (2):

$$K_{\text{el-ph}}(T) = \gamma_{\text{el-ph}} T$$

= (1.82×10⁻⁹)($\rho \theta_D$)AZ^{-2/3}T(W cm⁻¹ K⁻¹),
(3)

where ρ = electrical resistivity (at 4.2 K) expressed in units of $\mu \Omega$ cm, A = mass number of the alloy,

and

Z = average number of electrons per atom.

It should be mentioned that we are using the freeelectron model with some caution to obtain an estimate of the phonon-scattering rate. The limitations of the free-electron model arising from the strong electron-phonon interaction will be discussed below. The value of γ_{el-ph} is smaller for smaller θ_D and A. This explains the statement made earlier that a smaller θ_D and A will make $K_{el-ph}(T)$ more observable. The values of $\gamma_{\text{el-ph}}$, calculated by using Eq. (3), are listed in Table II. (Values of electrons per atom used in the calculation are Zr, 4; Br, 2; Ni, 1; P, 5; Si, 4; Cu, 1; for Pd we have used both one electron per atom and ten electrons per atom and this gives us a range of γ_{el-ph} which has been shown in Table II.) The agreement between the experimental and the theoretical γ_{el-ph} is considered to be adequate, considering the fact that we have used the simple free-electron model. It appears to be worth noting that the theoretical γ_{el-ph} is always somewhat higher than the experimental one. The reasons for this may be the enhanced density of states at the Fermi surface due to electron-phonon coupling in the zirconium alloys. This would mean more scattering centers and hence a lower γ_{el-ph} . It is certain that there is a fair amount of electron-phonon interaction in these alloys as indicated by the mass enhancement factor $\lambda \sim 0.7$ for the Zr-Be alloys and $\lambda \sim 0.5$ for $Zr_{0.7}Pd_{0.3}$, which one obtains from the Macmillan relation.¹⁸ It is not clear, however, how one can use the experimentally observed λ in calculating γ_{el-ph} in the extreme limit $q_{ph}l_e \ll 1.^{19}$ In any case, the perturbations caused by this coupling are small so that they do not affect $K_{el-ph}(T)$, and hence our conclusions, in a significant way.

For Ni-P we find that the estimated thermal resistivity $K_{el,ph}^{-1}$ due to scattering from electrons is much smaller than the observed total resistivity K^{-1} (~5% at 1 K). This may be the reason why it was not observed by Matey and Anderson.⁶

For Pd-Si-Cu, the predicted thermal resistivity K_{elph}^{-1} is expected to be somewhat larger, and should amount to ~10% of the total thermal resistivity K^{-1} between 1 and 5 K. The fact that the measured conductivity in this alloy bends away from the curve followed by the other alloys, see Fig. 5, might thus be explained by electron-phonon scattering. However, the percentage difference in thermal resistivity between the curves is larger than can be explained with Eq. (3), and is almost independent of temperature above 2 K.

We conclude that in the normal state a simple theory of the scattering of phonons by electrons provides a reasonable explanation of the experimental findings, with the exception of Pd-Si-Cu. This suggests that the basic scattering mechanism is the same as the scattering mechanism which occurs in highly disordered crystalline alloys with electron mean free paths much shorter than the phonon wavelength $(q_{\rm ph} l_e \ll 1)$. A detailed consideration of the departure from the free-electron model can be expected to improve the agreement.

(b) In order to investigate the scattering of phonons by electrons in the superconducting state, we plotted the ratio $K_{el-ph}^s(T)/K_{el-ph}^n(T)$ as a function of T/T_c in Fig. 7. $K_{el-ph}^s(T)$ is the value of $K_{el-ph}(T)$ for $T < T_c$ as shown in Fig. 6. $K_{el-ph}^n(T)$ is obtained by extrapolating $K_{el-ph}(T)$ estimated for the normal state by using the relation $K_{el-ph} = \gamma'T$, γ' was obtained by fitting the data above T_c with the linear relation. Considering the fact that we are already in a limit where $K_{el-ph}(T)$ in the normal state is linear in T we believe that this is a valid extrapolation. The values of $K_{el-ph}^s(T)/K_{el-ph}^n(T)$ as a function of T/T_c for two of the Zr-Be alloys and for $Zr_{0.7}Pd_{0.3}$ are very similar for $T/T_c > 0.7$. For



FIG. 7. $K_{\rm el-ph}^{s}/K_{\rm el-ph}^{n}$ as function of the reduced temperature $t = T/T_c$. The dashed lines are $K_{\rm el-ph}^{s}/K_{\rm el-ph}^{n}$ expected from the BRT theory (Ref. 12) for three values of Δ (0)/ kT_c obtained from Ref. 20. For clarity, error bars are shown on only one set of data points (alloy 1). They are similar for alloy 3 and somewhat larger for $\operatorname{Zr}_{0,7}\operatorname{Pd}_{0,3}$.

comparison, we have plotted in the same graph the values of $K_{el-ph}^{s}(T)/K_{el-ph}^{n}(T)$ as predicted from the Bardeen-Rickayzen-Tewordt (BRT) theory developed for crystals^{12,20} for three values of the ratio of gap to T_c , $\Delta(0)/kT_c$, for $T/T_c < 0.7$, the data points are close to the theoretical curves. Considering the large uncertainty in the data in this region (which arises mainly because two almost equal numbers are subtracted), we did not attempt to fit any theory to the data. It seems that for $Zr_{0.7}Pd_{0.3}$, $\Delta(0)/kT_c \approx 1.5$ and for the Zr-Be alloys, $\Delta(0)/kT_c$ lies between 1.6 and 1.8. Note that Graebner et al.⁷ obtained $\Delta(0)/kT_c = 1.47$ for $Zr_{0,7}Pd_{0,3}$ from specific-heat measurements. This observation agrees with the one by Tsuei et al.²¹ that in these noncrystalline superconductors $\Delta(0)/kT_c$ is not very different from the Bardeen-Cooper-Schrieffer (BCS) value, 1.76. For $T/T_c > 0.7$, the data in Fig. 7 show a deviation from the BRT theory, signifying a large drop in the phonon attenuation rate by the electrons near the transition-temperature region. A detailed discussion of this deviation is outside the scope of the paper, but a careful investigation of the phonon attenuation by electrons near the transition temperature is definitely worthwhile.

IV. SUMMARY AND CONCLUSION

The present investigation has shown that electrons act as scattering centers for phonons in noncrystalline solids. The scattering process can be semiquantitatively described with the picture that Debye phonons are carrying heat and that both longitudinal and transverse phonons are scattered by almost free electrons in addition to the instrinsic glassy states common to all amorphous solids. In the superconducting state the scattering is similar to the prediction based on the theory for crystalline superconductors with a gap-to-transition-temperature ratio $\Delta(0)/kT_c$ which is very close to the BCS value of 1.76. The sharp drop of phonon-scattering rate just below T_c requires further investigation.

It has been pointed out in the introduction that previously no bulk phonon scatterers has been found in amorphous dielectric solids which can scatter phonons strongly enough to become noticeable against the background of the mysterious intrinsic states. We can now state that in noncrystalline metals, at least, the free electrons represent such additional scattering centers. This finding suggests that there is no fundamental reason why the thermal conductivity of dielectric glasses should be entirely independent of defects or chemical dopants. It seems that we simply have not yet been able to find the dopants whose coupling to the lattice is strong enough to alter the conductivity of a glass. Thus we conclude that a continued search for extrinsic phonon scatterers in dielectric amorphous solids is justified. If such defects can indeed be identified, a study of their phonon scattering may shed some light on the nature of the intrinsic states in these solids.

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ACKNOWLEDGMENTS

The authors thank Professor R. O. Pohl for useful comments and for his help in preparing the manuscript. This work was supported in part by the U. S. Department of Energy, Contract No. EY-76-S-02-3151 and by the National Science Foundation Grant No. DMR 78-01560.

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