RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 51, NUMBER 1 1 JANUARY 1995-I

Eliashberg function of amorphous metals

D. Belitz and M. N. Wybourne
Department of Physics and Materials Science Institute, University of Oregon, Eugene, Oregon 97403
(Received 19 October 1994)

A connection is proposed between the anomalous thermal transport properties of amorphous solids and the low-frequency behavior of the Eliashberg function. By means of a model calculation we show that the size and frequency dependence of the phonon mean free path that has been extracted from measurements of the thermal conductivity in amorphous solids lead to a sizable linear region in the Eliashberg function at small frequencies. Quantitative comparison with recent experiments gives very good agreement.

The functional form of the Eliashberg function $\alpha^2F(\omega)$ in amorphous metals has been the subject of debate for a long time. Experimentally, there is uniform agreement that in amorphous simple metals the low-frequency part of the Eliashberg function is strongly enhanced over what is observed in crystalline materials.[1] The functional form at low frequencies, although difficult to determine experimentally, is usually found to be linear.[2] This experimental observation has given rise to a substantial theoretical debate. Bergmann[3] and many others[4] argued that a disorder enhancement of the electron-phonon coupling leads to a linear low-frequency behavior of the Eliashberg function, in accord with experiment. This was disputed by Schmid[5] and by Keck and Schmid,[6] who claimed that this disorder enhancement was spurious, and due to an incorrect application of the Fröhlich model to disordered materials. Schmid's calculation gave instead $\alpha^2F(\omega\to 0)\sim\omega^3$, in disagreement with experiment. The ω^3 result was also confirmed by others.[7] Since different models and different calculational methods had been used by these various authors, the theoretical problem was widely considered unsolved for a long time. This disagreement was finally settled by Reizer and Sergeyev,[8] who explicitly pointed out the errors in Ref. 4 and showed that a correct calculation leads to Schmid's result independent of the model and the method used. This led to the unsatisfying situation that a theoretically credible result, viz., Schmid's $\alpha^2F(\omega\to 0)\sim\omega^3$, disagreed with experiment, while incorrect arguments led to $\alpha^2F(\omega\to 0)\sim\omega$ in agreement with the experimental observations. This situation was summarized by one of us,[9] who also argued that no existing comparison between theory and experiment had been careful and accurate enough to really rule out Schmid's result. However, recent experiments by Watson and Naugle have shown that Schmid's result is not compatible with experiments on amorphous CuSn alloys.[10]

At this point it is important to distinguish between Schmid's general theory for the electron-phonon coupling in impure metals, and his and others' specific model calculations. Since the work of Reizer and Sergeyev showed that Schmid's general theory is physically correct, the search for reasons behind the discrepancy between his results and experiment should then turn to the model assumptions. On the electronic side the main assumption is that of nearly free electrons. It is hard to see how this could qualitatively fail in simple metals as long as the resistivities are moderate, and the electronic states are effectively three dimensional. On the phonon side, Schmid assumed undamped Debye phonons, an assumption that is necessary in order to obtain the ω^3 law as the asymptotic low-frequency behavior. As was shown in Ref. 9, the inclusion of phonon damping by electrons leads to a *linear* low-frequency asymptotic behavior (albeit with a prefactor that is too small by several orders of magnitude to explain the experimental results), which then crosses over to Schmid's ω^3 law. The prefactor of the linear term is proportional to the phonon damping. This raises the possibility that very strong phonon damping (which would have to be of other than electronic origin) might lead to a linear term in $\alpha^2F(\omega)$ whose prefactor is large enough to account for the experimental observations.

In order to pursue this last point, let us recall that besides the problems with the Eliashberg function mentioned above, amorphous materials have properties of entirely phononic origin that are hard to understand. In particular the thermal conductivity κ shows an enigmatic behavior. Even though it has been stressed that the thermal conductivity is not understood in any temperature region, the general phenomenology is clear, consistent, and well documented.[11,12] As a function of temperature T, the thermal conductivity behaves like $\kappa\sim T^2$ for $T/\Theta\lesssim 10^{-2}$, with Θ the Debye temperature. The origin for the phonon scattering in this region is not known for certain. The phenomenological two-level system concept has often been invoked in this context,[12] but no consensus has ever been reached. For $10^{-2}\lesssim T/\Theta\lesssim 10^{-1}$ the thermal conductivity is approximately independent of T. This is the so-called plateau region, which is characterized by strong, and strongly frequency-dependent, phonon scattering of uncertain origin. Finally, for $T/\Theta\gtrsim 10^{-1}$ the thermal conductivity becomes T dependent again, but it is not even clear whether the heat transport in this region is by phonons, much less what the scattering mechanisms are.

This poor state of physical understanding notwithstanding, the above phenomenology is remarkably universal, and seems to be characteristic of amorphous materials, both insulating[11] and metallic.[13] It has been used to deduce the following behavior of the phonon mean free path $l_{\rm ph}$ as a function of frequency. For frequencies $\omega\lesssim 10^{-2}k_B\Theta/\hbar$, $l_{\rm ph}$ is a linear function of frequency. For intermediate frequencies, $10^{-2}\lesssim\hbar\omega/k_B\Theta\lesssim 10^{-1}$, $l_{\rm ph}$ goes as a high power n of frequency. n has been reported to be at least 4, and possibly larger. This intermediate frequency regime corresponds to

the plateau region in the thermal conductivity. At still higher frequencies, $\omega \gtrsim 10^{-1} k_B \Theta/\hbar$, the phonon mean free path either becomes frequency independent,[14] or is a linear function of frequency again.[11]

In this paper we propose a connection between the thermal properties of amorphous materials as described above, and the low-frequency behavior of the Eliashberg function. In particular we show that Anderson's phenomenological functional form of the phonon mean free path, if used in Schmid's theory for the electron-phonon coupling, explains the observed behavior of the Eliashberg function as well as the observed behavior of the thermal transport. Let us start from the expression for the Eliashberg function, based on Schmid's general theory,[5] that was derived in Ref. 9,

$$\alpha^2 F(\omega) = \frac{1}{2\pi^2 N_F} \sum_{\mathbf{q},b} \alpha_b(\mathbf{q}) \frac{c_b}{\omega_b^2(\mathbf{q})} \operatorname{Im} D_b^R(\mathbf{q},\omega) \quad . \tag{1a}$$

Here $D_b^R(\mathbf{q},\omega)$ is the retarded phonon propagator, whose imaginary part reads,

$$\operatorname{Im} D_b^R(\mathbf{q},\omega) = \frac{4\omega\omega_b^2(\mathbf{q})\gamma_b(\mathbf{q})}{[\omega^2-\omega_b^2(\mathbf{q})]^2+4\omega^2\gamma_b^2(\mathbf{q})} \quad , \tag{1b}$$

where $\gamma(\mathbf{q})$ is the phonon damping coefficient. In writing Eqs. (1) we have assumed a free-electron model with N_F the electronic density of states per spin at the Fermi level. We have also assumed Debye phonons with one longitudinal and two transverse branches labeled by b ($b=L,T$), speed of sound c_b, and dispersion $\omega_b(\mathbf{q}) = c_b q$. $\alpha_b(\mathbf{q})$ in Eq. (1a) is the electronic contribution to the sound attenuation coefficient, for which we use the standard Pippard result,[15]

$$\alpha_b(\mathbf{q}) = \kappa_b f_b(ql) \quad , \tag{2a}$$

where $\kappa_b = (v_F/c_b)(\rho_e/\rho_{\text{ion}})/l$ with v_F the Fermi velocity, l the electronic mean free path, and ρ_e and ρ_{ion} the electronic and ionic mass density, respectively. The functions $f_{L,T}$ are given by,[15]

$$f_L(x) = \frac{1}{3} \frac{x^2 \arctan(x)}{x - \arctan(x)} - 1 \quad , \tag{2b}$$

$$f_T(x) = \frac{1}{2x^3} [2x^3 + 3x - 3(x^2+1)\arctan(x)] \quad . \tag{2c}$$

With phonon damping exclusively by electrons, as was assumed in Ref. 9, one has $\gamma_b(\mathbf{q}) = c_b \alpha_b(\mathbf{q})/2$. Here, however, we will consider the possibility of nonelectronic contributions to $\gamma_b(\mathbf{q})$. Accordingly, we write

$$\gamma_b(\mathbf{q}) = \tilde{\gamma} c_b q_D g(q/q_D) \quad , \tag{3}$$

where q_D is the Debye wave number, $\tilde{\gamma}$ is a number, and g is some function that determines the wave number or frequency dependence of the phonon damping. The latter we model after Anderson's proposal,[14] which has been extracted phenomenologically from thermal transport measurements in amorphous materials. Anderson's model consists of the three distinct regions mentioned above: (1) a low-frequency region where the damping is a linear function of frequency, (2) an intermediate region where the damping goes as a large power of the frequency, and (3) a high-frequency region where the damping is independent of frequency.[16] The intermediate region corresponds to the characteristic plateau that is observed in the T-dependent thermal conductivity. We thus model the function $g(x)$ in Eq. (3) as

$$g(x) = 10^n y \frac{x/y + (x/y)^n}{10^n + (x/y)^n} \quad . \tag{4}$$

Here y is the onset of the plateau region in units of the Debye wave number, the width of the plateau region has been assumed to be one decade, and n is the power that characterizes the frequency dependence of the phonon mean free path in the plateau region.

Before we turn to a numerical evaluation of the integral, Eq. (1a), that determines $\alpha^2 F(\omega)$, let us consider the low-frequency behavior analytically. Asymptotically, $\alpha^2 F(\omega) \sim \omega/\omega_\alpha$, with a slope ω_α^{-1}. The latter we estimate for a clean system, i.e., in the limit $l \to \infty$. In this limit only longitudinal phonons contribute, and we can use the asymptotic form of the function f_L in Eq. (2b), $f_L(x \to \infty) = \pi x/6$. Then we obtain,

$$\frac{\epsilon_F}{\omega_\alpha} = \frac{\tilde{\gamma}}{6\pi} \frac{q_D}{k_F} \left(\frac{v_F}{c_L}\right)^3 \frac{\rho_e}{\rho_{\text{ion}}} \int_0^1 \frac{dx}{x} g(x) \quad . \tag{5}$$

Typical parameter values are $q_D/k_F \approx 1$, $v_F/c_L \approx 10^3$, and $\rho_e/\rho_{\text{ion}} \approx 10^{-5}$. For the parameters y and n in Eq. (4) we take[14] $y \approx 0.02$ and $n \approx 4$. Finally, $\tilde{\gamma}$ determines the overall scale for the phonon mean free path l_{ph}. A typical value is $l_{\text{ph}} \approx 1$ cm at a frequency of 1 GHz. With $c_L \approx 2\times 10^5$ cm/s this corresponds to $\tilde{\gamma} \approx 2\times 10^{-4}$. This yields $\epsilon_F/\omega_\alpha \approx 1700$. With a Fermi energy $\epsilon_F \approx 10$ eV we obtain $\omega_\alpha \approx 6$ meV. This value for ω_α is of the same order of magnitude as the one typically obtained from tunneling experiments.[1,10]

Now that we have seen that we obtain promising results for $\alpha^2 F(\omega \to 0)$ with reasonable parameter values, let us calculate $\alpha^2 F(\omega)$ numerically, and compare quantitatively with experiments. Watson and Naugle[10] have performed a detailed study of amorphous SnCu. For the stoichiometry $Sn_{0.87}Cu_{0.13}$ they quote the following parameter values: $\epsilon_F = 1.54\times 10^{-11}$ erg, $k_F = 1.59\times 10^8$ cm^{-1}, $v_F = 1.84 \times 10^8$ cm/s, $l = 9.58\times 10^{-9}$ cm, $q_D = 1.31\times 10^8$ cm^{-1}, $c_L = 1.6 \times 10^5$ cm/s, $c_T = 8.1\times 10^4$ cm/s. Of these, the electronic parameters are much better known than the two sound velocities. Using these parameters, as well as $n=5$, $y=0.015$, and $\tilde{\gamma} = 8.0\times 10^{-5}$, we have calculated $\alpha^2 F(\omega)$ for frequencies up to 1.6 meV, which was the lower-frequency cutoff in the experiment of Ref. 10. The high-frequency behavior resulting from our calculation would not be realistic anyway due to our using a Debye model. The result was shown in Ref. 10, and was used as low-frequency input in a McMillan-Rowell inversion procedure to obtain $\alpha^2 F$ from tunneling data. It is also shown again as the curve labeled $n=5$ in Fig. 1. For the inversion procedure an overall factor multiplying the calculated $\alpha^2 F$ was used as a fit parameter. The need for such an overall scale factor is not surprising, given our free-electron model. The factor used for the best fit is equivalent to a deviation of the density of states in Eq. (1a) from its free-electron value by 14%. A compari-

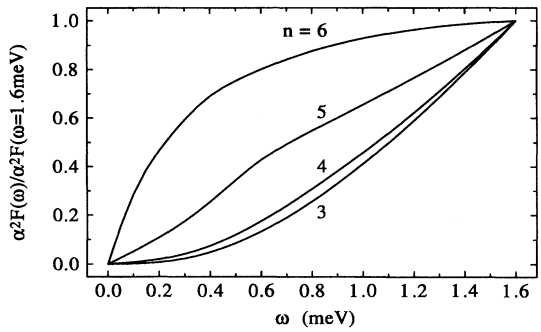

FIG. 1. Results for the Eliashberg function α^2F as a function of energy or frequency for different values of the exponent n in Eq. (4). All other parameters were held fixed at the values given in the text. α^2F has been normalized by its value at $\omega=1.6$ meV, which is 0.436, 0.279, 0.208, and 0.200 for $n=6$, 5, 4, and 3, respectively.

son between the calculated and the measured tunneling density of states then provides a measure of how well the low-frequency input describes the actual system. Watson and Naugle found that our calculated α^2F does very well, although not quite as well as if one assumes a strictly linear low-frequency behavior. It should be stressed that our calculation used the parameters as provided by the experimentalists, some of which are not known very accurately. Since the inversion procedure is quite involved no attempt was made to fine tune the parameters.

This result shows that Schmid's theory with a phonon damping that accounts for the thermal transport properties characteristic of amorphous metals gives good agreement between the calculated Eliashberg function and tunneling data. In contrast, the same theory with phonon damping by electrons only is not capable of explaining the experimental results.[10]

In addition to this comparison between theory and experiment, let us demonstrate the effects of some parameter changes on α^2F. We consider the four results for α^2F shown in Fig. 1. The curve labeled $n=5$ was obtained with the parameters as given above. The slight bulge in this curve results from the leveling off of the phonon mean free path at the high-frequency end of the plateau region. This moderates the rapid increase of α^2F at lower frequencies, which is due to the strong frequency dependence of the phonon mean free path. With a weaker frequency dependence of the phonon mean free path in the plateau region, i.e., a smaller exponent n in Eq. (4), the initial slope of α^2F is much smaller, and over the frequency range considered α^2F shows a purely positive curvature. Conversely, a still larger exponent n leads to a purely negative curvature of α^2F. This is demonstrated in Fig. 1. The curves with stronger curvature all led to substantially less good agreement with experiment than the one for $n=5$. We have also considered the sensitivity of the result to the ratio of the longitudinal and transverse speeds of sound, which is not known very accurately. We have found only a very weak dependence of the functional form of α^2F on this ratio in the region $1.8<c_L/c_T<2.5$. Finally, we have changed the damping parameter $\tilde{\gamma}$ with all other parameters held fixed. This was found to have a very similar effect to changing n, with α^2F changing from negative to positive curvature as $\tilde{\gamma}$ is increased, or the phonon mean free path at a reference frequency is decreased. The effect of changing $\tilde{\gamma}$ by a factor of 10 was roughly equivalent to changing n by 1. For instance, with $n=4$ and $\tilde{\gamma}=8\times10^{-4}$ we obtained a curve that was hardly distinguishable from the one for $n=5$ shown in Fig. 1. Generally, we found that with reasonable parameters for simple metals we need $4\lesssim n\lesssim 6$ in order for our explanation of the behavior of α^2F to be viable.

In conclusion, we have shown that Schmid's theory of electron-phonon coupling in impure metals can account for the observed low-frequency behavior of the Eliashberg function in amorphous simple metals if one assumes a strong phonon damping consistent with the one extracted from measurements of the thermal conductivity. While the physics underlying the strong damping is not known, this observation unifies two seemingly unconnected, and separately mysterious, properties of amorphous materials. It suggests that strong phonon scattering is a very fundamental feature of the amorphous state, and that understanding its origin would explain many different properties of amorphous materials at once.

We would like to thank Don Naugle for rekindling our interest in the present problem, and for numerous discussions. This work was supported by the NSF under Grants No. DMR-92-09879 and No. DMR-90-19525.

[1] See, e.g., G. Bergmann, Phys. Rep. **27C**, 159 (1976).

[2] It is worth mentioning that the low-frequency enhancement may be absent, and the low-frequency behavior of α^2F may be smaller than linear, in amorphous transition metals, D. B. Kimhi and T. H. Geballe, Phys. Rev. Lett. **45**, 1039 (1980). This difference between simple metals and transition metals is not well understood, and needs more experimental attention. For the purpose of our discussion we restrict ourselves to simple metals.

[3] G. Bergmann, Phys. Rev. B **3**, 3797 (1971).

[4] H. Takayama, Z. Phys. **263**, 329 (1973); S. G. Lisitin, Fiz. Nizk. Temp. **1**, 1516 (1975) [Sov. J. Low Temp. Phys. **1**, 728 (1975)]; S. J. Poon and T. H. Geballe, Phys. Rev. B **18**, 233 (1978).

[5] A. Schmid, Z. Phys. **259**, 421 (1973).

[6] B. Keck and A. Schmid, J. Low Temp. Phys. **24**, 611 (1976).

[7] E. Eisenriegler, Z. Phys. **258**, 185 (1973); G. Grünewald and K. Scharnberg, *ibid.* **268**, 197 (1974); Z. Phys. B **20**, 61 (1975).

[8] M. Yu. Reizer and A. V. Sergeyev, Zh. Eksp. Teor. Fiz. **90**, 1056 (1986) [Sov. Phys. JETP **63**, 616 (1986)].

[9] D. Belitz, Phys. Rev. B **36**, 2513 (1987).

[10] P. W. Watson III and D. G. Naugle, preceding paper, Phys. Rev. B **51**, 685 (1995); (private communication).

[11] J. J. Freeman and A. C. Anderson, Phys. Rev. B **34**, 5684 (1986).

[12] *Amorphous Solids*, edited by W. A. Phillips (Springer, New York, 1981).

[13] H. v. Löhneysen and F. Steglich, Phys. Rev. Lett. **39**, 1205 (1977); J. R. Matey and A. C. Anderson, J. Non-Cryst. Solids **23**, 129 (1977).

[14] A. C. Anderson, in *Amorphous Solids* (Ref. 12), Fig. 5.3.

[15] A. B. Pippard, Philos. Mag. **46**, 1104 (1955).

[16] As mentioned before, the behavior in the high-frequency region, above the plateau, is uncertain. We have also tried a linear frequency dependence for l_{ph} in this region (Ref. 11). In our results for $\alpha^2 F$ we have found no qualitative differences between this assumption and the one of a frequency-independent l_{ph}.