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Some effects of structural disorder on phonon lifetimes

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A calculation in the long-wavelength limit of the damping of a plane wave due to structural disorder is presented. Two contributions are found to be important: the first is incoherent and leads to a mean free path proportional to the fourth power of the phonon wave vector. The strength of the scattering, when estimated using available parameters, is sufficient to account for the thermal conductivity of glasses in the region of the plateau. The second contribution comes from coherent scattering and appears to be unique to strongly scattering systems such as amorphous substances. An estimate of its strength is very difficult because it depends on the magnitude of fourth-order elastic constants. However, it appears to be more than sufficient to account for a substantial part of the phonon mean free path at very long wavelengths.

INTRODUCTION

While the concept of a phonon is clear for crystalline materials, the problem of the normal modes of an amorphous substance has not yet been solved in a satisfactory way. Nevertheless, it has been shown that at long wavelengths a Debye-type description provides an adequate basis with which to account for the thermal conductivity at low temperatures.[1] Thus a Debye or continuum description should be adequate for long wavelengths. The question to be considered in this paper concerns the effect of the structural disorder on the lifetime of a propagating plane wave in this long-wavelength limit.

It will be found that fluctuations in density and force constant lead as might be expected to "Rayleigh" scattering, i.e., an inverse mean free path that varies as the phonon frequency to the fourth power. However, it will also be found that the strong scattering leads to another effect:

When summing the squared scattering amplitude there are two contributions,[2] one that arises from the squared amplitude for individual scatters, and another that consists of the products of the amplitudes from different sites. Normally, the second contribution can be neglected because it leads to a δ function in the forward direction. However, because the scattered wave is also attenuated, the volume over which the integration is performed should not exceed a dimension on the order of a mean free path. Restricting the volume broadens the δ function. When the mean free path (mfp) is very small the δ function is broadened sufficiently for a significant contribution to the total scattering to result. This contribution rapidly becomes important because it is proportional to the square of the number of scatters which outweighs the fact that it is small-angle scattering, confined essentially to angles less than $(\Lambda k)^{-1}$, where Λ is the mfp and k the phonon wave vector.

It has also been demonstrated experimentally[3] that local oscillators contribute to the lifetime at long wavelengths. It is likely that these are also a consequence of structural disorder, however, the present work makes no contribution towards a microscopic model for such oscillators.

The Hamiltonian for a structurally disordered monatomic solid insulator can be written in standard form:

$$
\begin{aligned}
H = {} & \sum_{\alpha}\sum_{i}\frac{P^2_{\alpha}(i)}{M} + \frac{1}{2}\sum_{\alpha}\sum_{\alpha'}\sum\sum_{i\neq j} B_{\alpha\alpha'}(\vec{r}_i-\vec{r}_j)[u_{\alpha}(i)-u_{\alpha}(j)][u_{\alpha'}(i)-u_{\alpha'}(j)] \\
& + \frac{1}{6}\sum_{\alpha}\sum_{\alpha'}\sum_{\alpha''}\sum\sum_{i\neq j} C_{\alpha\alpha'\alpha''}(\vec{r}_i-\vec{r}_j)[u_{\alpha}(i)-u_{\alpha}(j)][u_{\alpha'}(i)-u_{\alpha'}(j)][u_{\alpha''}(i)-u_{\alpha''}(j)] \\
& + \frac{1}{24}\sum_{\alpha}\sum_{\alpha'}\sum_{\alpha''}\sum_{\alpha'''}\sum\sum_{i\neq j} D_{\alpha\alpha'\alpha''\alpha'''}(\vec{r}_i-\vec{r}_j)[u_{\alpha}(i)-u_{\alpha}(j)][u_{\alpha'}(i)-u_{\alpha'}(j)][u_{\alpha''}(i)-u_{\alpha''}(j)][u_{\alpha'''}(i)-u_{\alpha'''}(j)], \qquad (1)
\end{aligned}
$$

where $u_{\alpha}(i)$ and $P_{\alpha}(i)$ are Cartesian components of the displacement and momentum of the atom at i. The $B_{\alpha\alpha'}$, $C_{\alpha\alpha'\alpha''}$ $D_{\alpha\alpha'\alpha\alpha''\alpha'''}$ are second, third-, and fourth-order force constants. The expansion is continued to quartic terms for reasons which will become clear below.

The conventional solution to the normal mode problem involves the transformation of the Hamiltonian using plane waves. This is difficult for structurally disordered systems because, as Morgan has discussed,[4] it is not clear how to choose an appropriate set of vectors. This difficulty may be circumvented if long wavelengths alone are of interest. In this case, a Debye approximation is adequate: writing

$$P_\alpha(i) = N^{-1/2}\sum_k P_\alpha(k) e^{-i\vec{k}\cdot\vec{r}_i} ,$$

$$u_\alpha(i) = N^{-1/2}\sum_k U_\alpha(k) e^{i\vec{k}\cdot\vec{r}_i} ,$$

and in turn expressing $P_\alpha(k)$ and $u_\alpha(k)$ in terms of the phonon annihilation and creation operators $a_p(k)$ and $a_p^\dagger(k)$, and only keeping the quadratic part, the Hamiltonian becomes

$$H = \sum_p \sum_k \hbar\omega_p(k)[a_p^\dagger(k) a_p(k) + \tfrac{1}{2}] - \sum_{p,p'} \sum_{k'\neq k} \hbar \tfrac{1}{2}\omega_p(k) \left[\left(\epsilon_p(k)\epsilon_{p'}(k') \sum_j e^{i(\vec{k}'-\vec{k})\cdot\vec{r}_j} \right) - \frac{B_{pp'}(-k,k')}{M\omega_p(k)\omega_{p'}(k')} \right] \times [a_p(-k)a_{p'}^\dagger(k') + a_p^\dagger(-k)a_{p'}(k')] . \tag{2}$$

In this expression, $\omega_p(k)$ is the frequency associated with wave vector k and polarization p, and ϵ is the polarization vector

$$B_{pp'}(-k,k') = \frac{1}{N} \sum_j e^{i(\vec{k}'-\vec{k})\cdot\vec{r}_j} \sum_\alpha \sum_{\alpha'} \sum_{l\neq j} \epsilon_\alpha(-k,p)\epsilon_{\alpha'}(k',p') B_{\alpha\alpha'}(\vec{r}_j - \vec{r}_l)(1 - e^{i\vec{k}\cdot(\vec{r}_j - \vec{r}_l)}) .$$

The first term in Eq. (2) is the familiar phonon Hamiltonian, the second contains the effect of density fluctuations. It cannot, however, include force-constant fluctuations because the force-constant B is harmonic, and, in this approximation, cannot depend on changes in interatomic distance. The effect of force-constant fluctuations is contained in the anharmonic terms C and D.

We choose to neglect density fluctuations because they should also scatter light, which many glasses do not to any great extent. In any case, the ratio of a fractional force-constant change to the associated fractional density change is on the order of a Grüneisen parameter. Therefore, scattering should be stronger by a factor on the order of the Grüneisen parameter squared, so force-constant changes should dominate for most amorphous substances.

A more important source of scattering lies in the atomic displacements themselves. The changes in elastic constant are determined by the individual displacements. The density fluctuations are an average of the displacements over some volumes and the force-constant fluctuations are seriously under estimated if they are obtained by multiplying the density fluctuations by the appropriate Grüneisen parameter as Jäckle[5] has done.

For these reasons, density fluctuations are probably an unimportant source of scattering and thus the physical basis for a simplified calculation by Walton[6] is probably incorrect.

Thus, the scattering is ascribed to the changes in force constant resulting from the departure of the nearest-neighbor distance from the average. There also exist large variations in bond angle,[7] but these are more difficult to estimate and will simply be ignored here. The Hamiltonian is now

$$H = H_0 + H' ,$$

where, including the polarization index in the wave vector

$$H_0 = \sum_k \hbar\omega(k)(a_k a_k^\dagger + \tfrac{1}{2}) ,$$

$$\begin{aligned} H' &= \sum_{k\neq k'}\sum \frac{\hbar\omega_k}{2N} \sum_{i\neq l}\sum e^{i(\vec{k}'-\vec{k})\cdot\vec{r}_i} \sum_{\alpha,\alpha'} \epsilon_\alpha(k)\epsilon_{\alpha'}(k')\Delta B_{\alpha\alpha'}(\vec{r}_i - \vec{r}_l)(1 - e^{i\vec{k}'\cdot(\vec{r}_i - \vec{r}_l)}) \\ &\qquad \times (1 - e^{-i\vec{k}'\cdot(\vec{r}_i - \vec{r}_l)})(a_k a_{k'}^\dagger + a_{k'} a_k^\dagger) \\ &= \sum_{i\neq l}\sum H'(\vec{r}_{il}) , \end{aligned} \tag{3}$$

where

$$\Delta B_{\alpha\alpha'}(\vec{r}_i - \vec{r}_l) \simeq \frac{1}{3}\sum_{\alpha''} C_{\alpha\alpha'\alpha''}(a)(\vec{r}_i - \vec{r}_l - \vec{a})_{\alpha''} + \frac{1}{12}\sum_{\alpha''}\sum_{\alpha'''} D_{\alpha\alpha'\alpha''\alpha'''}(\vec{r}_i - \vec{r}_l - \vec{a})_{\alpha''}(\vec{r}_i - \vec{r}_l - \vec{a})_{\alpha'''} , \tag{4}$$

$|a|$ is the average nearest-neighbor separation, and $\vec{a}$ is parallel to $\vec{r}_i - \vec{r}_l$.

The attenuation and energy shift of the phonons can be obtained from the imaginary and real parts of the self-energy

$$\pi = H' + H'GH' ,$$

where G is the propagator corresponding to H.

More explicitly, these quantities for a phonon of wave vector k are given by the real and imaginary parts of

$$\langle k|\pi|k\rangle = \langle k|H'GH'|k\rangle .$$

Since H' is quadratic in phonon operators, there is no diagonal contribution from H'.

In general, it is plausible that the Green's function G may be approximated by[8]

$$G(\vec{r}_i, \vec{r}_j; \omega) \simeq G_0(\vec{r}_i, \vec{r}_j; \omega) e^{-\Lambda^{-1}(\omega)|\vec{r}_i - \vec{r}_j|} , \tag{5}$$

where $\Lambda(\omega)$ is the mean free path of phonon ω, and G_0 is the unperturbed function, corresponding to H_0.

The physical meaning of this approximation is just that the phonon will be attenuated in propagating from i to j. Assuming that the attenuation is only a function of phonon frequency, and that the solid is isotropic, Eq. (5) is obtained.

Thus, a self-consistent equation can now be written for $\Lambda(\omega)$, since $\Lambda(\omega) = \tau(\omega)V(\omega)$, where $\tau(\omega)$ is the phonon lifetime and $V(\omega)$ the phonon group velocity. $\tau^{-1}(\omega) = (2\pi/\hbar)\mathrm{Im}\,\pi(\omega)$, and, therefore,

$$\Lambda^{-1}(\omega) = \frac{2\pi}{V(\omega)\hbar} \mathrm{Im} \sum_{k'} \sum_{ij} \sum_{l \neq i} \langle k|H'(R_{il})|k'\rangle G_0(k', \omega_{k'}) \sum_{m \neq j} \langle k'|H'(R_{jm})|k\rangle e^{-\Lambda^{-1}|R_{ij}|} . \tag{6}$$

When $i=j$ and $m=l$, or when $j=l$ and $m=i$, Eq. (6) becomes a direct relationship for $\Lambda^{-1}(\omega)$ yielding the incoherent contribution to the mean free path. This is just Rayleigh scattering $\Lambda^{-1} \alpha k^4$. For $i \neq j \Lambda^{-1}$ must be determined self-consistently. This is the coherent contribution[2] and yields quite a different behavior, $\Lambda^{-1} \alpha k$.

INCOHERENT SCATTERING

There are two contributions, the first comes from the terms in Eq. (6) for which $i=l$. In this case the exponential damping factor drops out. The second comes from terms in which i and l are interchanged, since $\Delta B_{il} \equiv \Delta B_{li}$. Again, the damping factor and the phase factor are unimportant since R_{il} is the nearest-neighbor distance.

The expression for Λ^{-1} becomes

$$\Lambda^{-1} = [\hbar V(\omega)(2\pi)^3]^{-1} \Omega \int d^3k' \sum\sum_{i \neq l} [\,|\langle k'|H'(r_{il})|k\rangle|^2 + 2\,|\langle k|H'(r_{il})|k'\rangle \langle k'|H'(r_{li})|k\rangle|\,] \frac{2\pi}{\hbar} \delta(\omega_k - \omega_{k'}) ,$$

where Ω is the volume of the solid.

From Eq. (3)

$$|\langle k'|H'(r_{il})|k\rangle| = \frac{\hbar}{2N} \sum_{\alpha,\alpha'} \epsilon_\alpha(k) \epsilon_{\alpha'}(k') \frac{\Delta B_{\alpha\alpha'}(\vec{r}_i - \vec{r}_l)}{M\omega_k} (1 - e^{i\vec{k}'\cdot\vec{R}_{il}})(1 - e^{-\vec{k}'\cdot\vec{R}_{il}}) . \tag{7}$$

Since

$$M\omega_k^2 = - \sum_{\alpha,\alpha'} \sum_{l \neq i} \epsilon_\alpha(k) \epsilon_{\alpha'}(-k) B_{\alpha\alpha'}(R_{il}) (1 - e^{i\vec{k}\cdot\vec{R}_{il}})(1 - e^{-i\vec{k}\cdot\vec{R}_{il}}) , \tag{8}$$

and ΔB and B are short ranged, for the case where k is a longitudinal phonon, straightforward algebra yields

$$\Lambda_{\mathrm{inc}}^{-1}(k) = \frac{3V_0k^4}{8\pi} \left[\frac{2}{5} \frac{\langle \Delta B_{\alpha\alpha}^2 \rangle}{\overline{B}_{\alpha\alpha'}^2} + \frac{4}{15} \frac{\langle \Delta B_{\alpha\alpha'}^2 \rangle}{\overline{B}_{\alpha\alpha'}^2} \left(\frac{V_t}{V_l} \right)^4 + \frac{16}{15} \frac{\langle \Delta B_{\alpha\alpha'}^2 \rangle}{\overline{B}_{\alpha\alpha'}^2} \left(\frac{V_l}{V_t} \right) + \frac{4}{15} \frac{\langle \Delta B_{\alpha\alpha}^2 \rangle}{\overline{B}_{\alpha\alpha}^2} \left(\frac{V_l}{V_t} \right)^5 \right] ,$$

where V_l and V_t are the longitudinal and transverse velocities of sound.

The first two factors in the brackets are due to scattering into a longitudinal mode, whereas the last two correspond to the scattered phonon being transverse. Taking $\langle \Delta B_{\alpha\alpha'}^2 \rangle / \overline{B}_{\alpha\alpha'}^2 \sim \langle \Delta B_{\alpha\alpha}^2 \rangle / \overline{B}_{\alpha\alpha}^2$ and ignoring terms in V_t/V_l,

$$\Lambda_{\mathrm{inc}}^{-1}(k_l) \sim \frac{3V_0}{8\pi} \frac{\langle \Delta B^2 \rangle}{\overline{B}^2} \left[\frac{2}{5} + \frac{16}{15} \left(\frac{V_l}{V_t} \right) + \frac{4}{15} \left(\frac{V_l}{V_t} \right)^5 \right] k^4 .$$

Similarly, if k is a transverse phonon

$$\Lambda_{\text{inc}}^{-1}(k_t)=\frac{3v_0k^4}{20\pi}\,\frac{\langle\Delta B_{\alpha\alpha}^2\rangle}{\overline{B}_{\alpha\alpha}^2}\left(\frac{V_l}{V_t}\right)^4 .$$

Now

$$\frac{\Delta B}{B}\sim\frac{\partial\ln B}{\partial\ln R}\left(\frac{\Delta R}{R}\right)=6\,\frac{\partial\ln\omega}{\partial\ln R}\left(\frac{\Delta R}{R}\right),$$

or

$$\Delta B/B\sim 6\gamma(\Delta R/R),$$

where γ in an average Grüneisen parameter obtained from thermal expansion. For vitreous silica $\gamma\sim-9$ at 4 °K, and[7] $\langle\Delta R^2\rangle/\overline{R}^2\sim10^{-3}$.

The mean-square fluctuation, calculated in this way is very large. The Grüneisen γ which is appropriate is not necessarily that obtained from thermal expansion. However, bearing these reservations in mind the following values are obtained:

$$\Lambda_{\text{inc}}^{-1}(k_l)\sim1.9v_0k^4$$

and

$$\Lambda_{\text{inc}}^{-1}(k_T)\sim1.0v_0k^4 .$$

v_0 is the volume of a molecule which for vitreous silica is 3.76×10^{-23} cm^3.

Using the resultant values for the mean free path in a standard calculation of the thermal conductivity, good agreement is obtained with experimental values in the plateau region.

A recent estimate of structural scattering by Zaitlin and Anderson[1] yields a mfp an order of magnitude too high. It is not quite clear why this is so because the expression used is one suggested by Zeller and Pohl,[9] and the latter authors obtain good agreement. In this model, the physical mechanism for the scattering was taken to be, in effect, the fluctuations in interatomic distance. The results obtained here, depend on quite a different mechanism, namely, the changes in force constant resulting from the fluctuating atomic displacements.

Jäckle[5] has calculated the scattering due to density fluctuations, and has included anharmonic effects. His scattering is roughly two orders of magnitude weaker than the result obtained here. The reason for this is that he uses the mean-square force-constant change over a small volume, obtained by multiplying the mean-square density fluctuation by a squared Grüneisen constant. This seriously underestimates the interaction, because the resultant average is not the mean-square force-constant change, but the mean-square fluctuation of the average force constant in the chosen volume.

COHERENT SCATTERING

The contribution from terms for which $i\neq l$ will now be estimated.

With Eqs. 7 and 8, Eq. (6) becomes, for scattering of a longitudinal phonon into another longitudinal phonon ,

$$\Lambda_{\text{coh}}^{-1}(\omega)=\frac{1}{\hbar V_l}\,\frac{\Omega}{(2\pi)^3}\int d^3k'\left(\frac{\hbar\omega}{2N}\right)^2\left(\sum\sum_{i\neq l}\frac{\Delta B_{\alpha\alpha}^{il}}{B_{\alpha\alpha}}\cos^2\theta\right)\left(\sum_{j\neq i}\sum_{m\neq j,\,i}\frac{\Delta B_{\alpha\alpha}^{jm}}{B_{\alpha\alpha}}\cos^2\theta\right)e^{i(\vec{k}-\vec{k}')\cdot\vec{R}_{ij}}e^{-\Lambda^{-1}|R_{ij}|}$$
$$\times\frac{2\pi}{\hbar}\delta(\omega_k-\omega_{k'}), \tag{9}$$

where θ is the angle between k and k'.

Replacing $\Delta B_{\alpha\alpha}^{il}$ and $\Delta B_{\alpha\alpha}^{jm}$ by their average values and converting the sum over i and j into an integral

$$\Lambda_{\text{coh}}^{-1}(\omega)=(V_l16\pi^2)^{-1}\int d^3k'(\omega)^2\left\langle\frac{\Delta B_{\alpha\alpha}}{B_{\alpha\alpha}}\right\rangle^2\delta(\omega_k-\omega_{k'})\cos^4\theta\int g(R)e^{i(\vec{k}-\vec{k}')\cdot\vec{R}}e^{-\Lambda^{-1}|R|}\,d^3R, \tag{10}$$

where $g(R)$ is the probability that an atom will be found at R if there is one at the origin. Performing the integration over R, letting $g(R)=1$,

$$\Lambda^{-1}=\frac{1}{V_l^2 2\pi}\int d^3k'(\omega)^2\left\langle\frac{\Delta B_{\alpha\alpha}}{B_{\alpha\alpha}}\right\rangle^2\cos^4\theta$$
$$\times\delta(\omega_k-\omega_{k'})\frac{\Lambda^{-1}}{(\Lambda^{-2}+Q^2)^2}, \tag{11}$$

where $Q=|\vec{k}-\vec{k}'|=k^2+k'^2-2kk'\cos\theta$.

Scattering involving mode conversion can probably be safely ignored since Q is never zero in that case and the denominator in (11) remains large.

Since $(\Lambda Q)^{-2}$ is small it is clear that the important values of k' are confined to the forward direction. In that case, letting $\cos\theta=1$, $\sin\theta=\theta$,

$$1=\frac{1}{8\pi}\left\langle\frac{\Delta B_{\alpha\alpha}}{B_{\alpha\alpha}}\right\rangle^2\int\frac{\theta\,d\theta\,d\phi}{(\frac{1}{2}k^{-2}\Lambda^{-2}+\theta^2)^2},$$

whence

$$1=\left\langle\frac{\overline{\Delta B}_{\alpha\alpha}}{\overline{B}_{\alpha\alpha}}\right\rangle^2\frac{1}{4}k^2\Lambda^2 ,$$

and

$$\Lambda^{-1}=\frac{1}{2}\,\frac{\overline{\Delta B}_{\alpha\alpha}}{\overline{B}_{\alpha\alpha}}\,k .$$

Again, the fluctuations in B are taken to be due to fluctuations in interatomic separation. The third-order term is linear in ΔR and yields zero. The first nonzero contribution comes from the quartic term. One way of estimating its magnitude can be obtained from the change in the γ with volume. D_{1111} for vitreous silica has been measured by Graham[10] at room temperature; he obtains $D_{1111} = 110 \times 10^3$ kbar.

Using

$$\frac{\Delta B}{B} = \frac{1}{12} \frac{D_{1111}}{C_{11}} \left\langle \frac{\Delta R^2}{R^2} \right\rangle ,$$

$$\Lambda^{-1}_{\text{coh}}(\omega) = 5 \times 10^{-3} k \; .$$

From Brillouin scattering[11] $\Lambda^{-1} \sim 10^{-3} k$, of which Golding[3] has shown that interaction with "two-level systems" accounts for roughly half the scattering, so the above overestimates the scattering by about an order of magnitude. This is not surprising considering the difficulty in estimating fourth-order constants.

Because of this uncertainty, an estimate of the mfp for transverse phonons is not presented here. It has been calculated and yields a similar result.

Naturally, the appropriate contribution due to a mechanism such as the one outlined above should be amenable to direct measurement. However, this is not a straightforward experimental question: If ultrasonic techniques are used, the coherent contribution will appear as a temperature-independent background. If this background is rejected, of course this coherent effect will most certainly be discarded too. Also, at low frequencies the mfp becomes larger than the sample size.[3] In this situation the above treatment is not appropriate, and in any case the coherent contribution would be difficult to untangle from the diffraction broadening.

At the present time the best estimate of the coherent contribution can come from Golding's measurements: he finds that when he extrapolates the mfp measured at 0.59 GHz to higher phonon frequencies the "two-level systems account for between 30% and 80% of the total." However, as he points out, this assumes an energy-independent density of states, and even a weak energy dependence would modify these figures considerably. It would be desirable to perform the same experiment at higher frequencies. Probably the best measurement of this sort could come from the temperature dependence of the Brillouin linewidth between 2 and 0.5 °K in backscattering.

SUMMARY AND CONCLUSION

Beginning with a microscopic Hamiltonian for an amorphous solid the effect of structural disorder on the lifetime of low-frequency phonons has been calculated. It is found that there are two contributions:

The usual incoherent contribution, $\Lambda^{1} \alpha k^4$. In contrast to other published estimates[1,5] it is found that the mfp is approximately equal to that obtained from the thermal conductivity.

In addition, coherent scattering must be considered. The reason it is important is that the mfp is short. Scattered waves should not be able to interfere if they come from sites separated by a distance greater than a mfp. If this distance is short, interference is no longer complete. Inclusion of what is essentially a multiple-scattering effect leads to $\Lambda^{1} \alpha k$. This dependence on k also accounts for the temperature dependence of the thermal conductivity at low temperatures. While a k dependence of this nature can be accounted for by interaction with two-level systems, it appears at present that these account for very roughly half the scattering.

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