

Calculation of phonon life time in amorphous solids

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Abstract. We present the calculation of phonon life time at low frequencies in an amorphous solid, which is assumed to be characterized by an elasticity that exhibits spatial fluctuation. Thermodynamic Green's function method is used to compute phonon self-energy, and an iterative method is devised to obtain an improvement upon the first order perturbation calculation. The elasticity correlation is taken to be an exponentially falling function of distance. We obtain an inverse life time that varies as the fourth power of phonon frequency for small values of the latter, and whose frequency-dependence becomes weaker and weaker as the frequency increases.

Keywords. Amorphous solids; elasticity fluctuation; phonon life time; thermal properties.

1. Introduction

Phonons in amorphous solids are known to be very short-lived and their life time is generally less by one or more orders than that of their counterparts in crystalline solids. This is because of the structurally disordered configuration of these solids. This disorder can affect the phonon life time in two ways: It is suggested that a small but finite number of atoms or groups of atoms can have two or more equilibrium configurations with different associated energies. They can alternate from one configuration to another exchanging energy with the excitations of the solid, and are called low-energy tunnelling systems (LTS) (Phillips 1972; Anderson *et al* 1972; Jäckle 1972). Power-dependent attenuation of ultrasonic waves at very low temperatures lends strong support to the existence of tunnelling systems (see, for example, Hunklinger and Arnold 1976). The approximately T^2 behaviour of thermal conductivity at very low temperatures can also be satisfactorily explained in terms of phonon scattering by LTS, assuming a uniform density of systems in energy space (see, for instance, Pohl 1976). The reciprocal of the corresponding phonon life time is roughly linear in phonon frequency. The plateau region of the conductivity *vs.* temperature curve, which is observed typically at a few degrees Kelvin and has a spread also of a few degrees, corresponds to a situation in which thermal phonons have a life time that falls rapidly with the increase in frequency. Conductivity in this region can be explained only if the density (in energy space) of LTS is assumed to be exceptionally high at energies that are of the same order as the energies of the phonons thermally important in this region. An alternative mechanism that has been suggested by some authors (Klemens 1965; Morgan and Smith 1974; Walton 1974; Choy 1977) to explain the anomalously low conductivity at low temperatures is the scattering of phonons by spatial fluctuations in density and elasticity in amorphous solids. The

local density and elasticity in a given region of an amorphous solid, which is generally regarded as a homogeneously disordered system on a macroscopic scale, can exhibit small departures from their respective average values. The characteristic length over which these mechanical quantities can be taken to remain practically constant would be of the order of a few tens of Ångström. It has been discussed (Joshi 1979) that phonons having very large wavelength would not be able to feel the existence of this microscopic inhomogeneity appreciably and their scattering would mainly be caused by LTS. With an increase in frequency, phonons are expected to grow more and more sensitive to the spatial variations in density and elasticity, and the corresponding scattering would become more important at relatively higher frequencies.

Here we study the contribution to phonon life time of scattering by spatial fluctuations in elasticity. Thermodynamic Green's function technique (Abrikosov *et al* 1964) is used to calculate the self-energy of phonons from which information on life-time is derived. An iterative method is used to obtain an improvement upon the first-order perturbation calculation. An exponentially decreasing function of distance is taken as a model description of the spatial correlation between local elasticities defined in different regions of the amorphous system.

2. Model Hamiltonian

The dynamical excitations in a solid continuum can be described by the following Hamiltonian (Ziman 1960)

$$H = \frac{1}{2} \int d^3r \left(\frac{1}{\rho} \mathbf{p} \cdot \mathbf{p} + \frac{\partial \mathbf{u}}{\partial \mathbf{r}} : \mathbf{g} : \frac{\partial \mathbf{u}}{\partial \mathbf{r}} \right), \quad (1)$$

where $\mathbf{u} \equiv \mathbf{u}(\mathbf{r})$ is the displacement vector of the medium at position \mathbf{r} , and $\mathbf{p} \equiv \mathbf{p}(\mathbf{r})$ is the corresponding momentum density. Integration is to be performed over the system volume V . Here ρ is the density of the material, which may be in general a function of position, and $\mathbf{g} \equiv \mathbf{g}(\mathbf{r})$ is the local elastic tensor of rank four. $\partial \mathbf{u} / \partial \mathbf{r}$ is in fact the strain tensor of rank two. To facilitate the calculations we assume that the density of the material would not be sensitively dependent on the disordered arrangement of the constituent atoms as the elasticity. Thus we ignore the spatial variation of ρ , treating it as a constant, and consider the variation of only \mathbf{g} , which would be more important.

If \mathbf{g}_0 denotes the elastic tensor averaged over the entire volume of the system, then we can write

$$\mathbf{g}(\mathbf{r}) = \mathbf{g}_0 + \Delta \mathbf{g}(\mathbf{r}), \quad (2)$$

and the Hamiltonian formally becomes

$$H = H_0 + \Delta H, \quad (3)$$

where ΔH contains the contributions of $\Delta \mathbf{g}$ and the contribution of other terms is included in H_0 . One can now define the normal Fourier modes of vibration of the

system such that H_0 is diagonalized. In second-quantized notation these modes can be defined by

$$\mathbf{u}(\mathbf{r}) = \sum_q \left(\frac{\hbar}{2\omega_q \rho V} \right)^{1/2} (a_q + a_{-q}^\dagger) \mathbf{e}_q \exp(i\mathbf{q} \cdot \mathbf{r}), \quad (4a)$$

and

$$\mathbf{p}(\mathbf{r}) = \sum_q \left(\frac{-\hbar \omega_q \rho}{2V} \right)^{1/2} (a_{-q}^\dagger - a_q) \mathbf{e}_q \exp(i\mathbf{q} \cdot \mathbf{r}), \quad (4b)$$

where \mathbf{q} is the wave vector of a mode with angular frequency ω_q and unit polarization vector \mathbf{e}_q . Here $q \equiv (\mathbf{q}, j)$ is an abbreviated notation for combined reference to the wave vector \mathbf{q} and polarization branch index j ; a_q and a_{-q}^\dagger are the usual destruction and creation operators for mode q . In terms of these operators we have

$$H_0 = \sum_q \hbar \omega_q (a_q^\dagger a_q + \frac{1}{2}), \quad (5)$$

which is the Hamiltonian that describes a phonon system in a perfectly homogeneous elastic continuum. The term ΔH can be now regarded as a perturbation acting on the excitations defined by H_0 . An exact analytical expression for ΔH cannot be obtained. However an approximate expression can be suggested, if we assume that all the elements of the tensor $\Delta g(\mathbf{r})$ differ from their spatially averaged values by the same proportion $\epsilon(\mathbf{r})$. Then a model representation of the perturbation can be given by

$$\Delta H = \sum_{q, q'} \frac{\hbar}{2V} (\omega_q \omega_{q'})^{1/2} \mathbf{e}_q \cdot \mathbf{e}_{q'} A_q A_{q'} \int d^3 r \epsilon(\mathbf{r}) \exp(i(\mathbf{q} + \mathbf{q}') \cdot \mathbf{r}), \quad (6)$$

where $A_q = 2^{-1/2} (a_q + a_{-q}^\dagger)$.

For the sake of brevity we define

$$U(q, q') = \frac{1}{V} (\omega_q \omega_{q'})^{1/2} \int d^3 r \epsilon(\mathbf{r}) \exp(i(\mathbf{q} + \mathbf{q}') \cdot \mathbf{r}), \quad (7a)$$

and $V(q, q') = U(q, q') \mathbf{e}_q \cdot \mathbf{e}_{q'}$, (7b)

so that $\Delta H = \frac{1}{2} \sum_{q, q'} V(q, q') A_q A_{q'}$. (8)

3. Green's function

We now define imaginary time thermodynamic Green's function in the usual way

$$\begin{aligned} G(q, q', \tau) &\equiv \langle \tilde{A}_q(i\tau); \tilde{A}_{q'}(0) \rangle \\ &\equiv - \langle W [\tilde{A}_q(i\tau) \tilde{A}_{q'}(0)] \rangle, \end{aligned} \quad (9)$$

where $\tau = i t$, measures the imaginary time, W is Wick's time ordering operator, and a tilde over the phonon field operator implies that the time development of the operator (in Heisenberg picture) is determined by the total Hamiltonian. The Fourier frequency representation for the Green's function can be defined by

$$G(q, q', \tau) = \frac{1}{\beta \hbar} \sum_n G(q, q', \omega_n) \exp(-i\omega_n \tau), \quad (10)$$

where $\beta = 1/k_B T$ is a measure of temperature on the inverse energy scale, and $\omega_n = 2\pi n/\beta \hbar$, n being an integral number.

A series representation of $G(\omega_n)$ in terms of the unperturbed Green's functions can be obtained using the S -matrix expansion method (Abrikosov *et al* 1964). In fact one can show that

$$\begin{aligned} G(q, q') &= G_0(q, q') + G_0(q, q_1) V(q_1, q_2) G_0(q_2, q') \\ &+ G_0(q, q_1) V(q_1, q_2) G_0(q_2, q_3) V(q_3, q_4) G_0(q_4, q') + \dots \end{aligned} \quad (11)$$

Here and in what follows it is implied that all the G 's are functions of ω_n , unless otherwise stated, and that summation is to be carried over the repeated variables q_1, q_2, \dots . The unperturbed Green's function is defined by

$$\begin{aligned} G_0(q, q') &= \delta_{q, -q'} G_0(q) \\ &= \delta_{q, -q'} \omega_q / [(i\omega_n)^2 - \omega_q^2]. \end{aligned} \quad (12)$$

One can rewrite (11) as

$$G(q, q') = G_0(q, q') + G_0(q) V(-q, q_1) G(q_1, q'). \quad (13)$$

We will be interested in $G(q, -q, \omega_n) \equiv G(q, \omega_n)$ which describes phonons in mode q . An iterative method can be now devised to obtain a series expansion for this quantity in terms of the other Green's functions. In fact (13) suggests that

$$G(q, q') = G_0(q) V(-q, q_1) G(q_1, q'), \text{ for } q' \neq -q. \quad (14)$$

By the repeated application of this relation to an infinite order, a series for $G(q)$ can be generated, each term of which has $G_0(q)$ as the last factor. This gives us

$$G(q) = G_0(q) + G_0(q) S(q) G(q), \quad (15)$$

which can also be written in the form

$$G^{-1}(q) = G_0^{-1}(q) - S(q). \quad (16)$$

Here we have neglected the term

$$\lim_{N' \rightarrow \infty} \sum_{q_1, q_2, \dots} G_0(q) V(-q, q_1) \left[\prod_{N=1}^{N'-1} G_0(q_N) V(-q_N, q_{N+1}) \right] G(q_{N'}, -q),$$

which would invariably remain there as the last term on the right hand side of (15). Note that in this term $q_N, q_{N'} \neq q$. Adopting this approximation is equivalent to truncating the series expansion of $G(q)$ to get rid of this last term. Alternatively, this amounts to evaluating this term, using the lowest order approximation for the non-diagonal element of the Green's function $G(q_{N'}, -q)$, which is zero. The advantage of taking this course is that the series left for $G(q)$ can be easily summed up to get a formal solution for $G(q)$ in terms of $S(q)$ defined by

$$S(q) = V(-q, q) + V(-q, q_1) G_0(q_1) V(-q_1, q) \\ + V(-q, q_1) G_0(q_1) V(-q_1, q_2) G_0(q_2) V(-q_2, q). \quad (17)$$

Here summation over q_1, q_2, \dots is implied and strictly speaking, these variables are not allowed to assume a value equal to q . But we shall ignore this restriction in view of the fact that in a real system a summation variable q_i will take an almost indefinitely large number of values. This permits us to write, using (11),

$$S(q) = V(-q, q) + V(-q, q_1) G(q_1, q_2) V(-q_2, q). \quad (18)$$

The first term on the right of this equation is zero, since the spatially averaged value of $\epsilon(\mathbf{r})$ is nil by definition.

The Green's function given above has been defined for frequencies on the imaginary axis of the complex frequency plane. The results can be extended to the real axis by some suitable limiting process to obtain the real-time Fourier representation (Abrikosov *et al* 1964). This can be done by letting $i\omega_n \rightarrow \omega$ where a positive imaginary part is implied to have been included in ω , which is therefore complex. $G(q, \omega)$ obtained in this manner will give the appropriate Green's function and correspondingly $S(q, \omega)$ obtained from (18) defines the phonon self-energy from which phonon life time can be calculated. The inverse life time can be approximately given by $\text{Im } S(q, \omega \approx \omega_q)$.

Using (7), the self-energy term can be written as

$$S(q, \omega) = \sum_{q_1, q_2} \omega_q (\omega_1 \omega_2)^{1/2} (\mathbf{e}_q \cdot \mathbf{e}_1) (\mathbf{e}_2 \cdot \mathbf{e}_q) G(q_1, q_2, \omega) I(q_1, q_2), \quad (19)$$

where
$$I(q_1, q_2) = \frac{1}{V} \int d^3r \int d^3r' \epsilon(\mathbf{r}) \epsilon(\mathbf{r}') \exp(i(\mathbf{q}_1 - \mathbf{q}) \cdot \mathbf{r}) \exp(i(\mathbf{q} - \mathbf{q}_2) \cdot \mathbf{r}). \quad (20)$$

Here we have written $\omega_{q_i} = \omega_i$ and $\mathbf{e}_{q_i} = \mathbf{e}_i$ for convenience.

We now regard $\epsilon(\mathbf{r})$ as a random variable of space coordinates. Then one can write (Ziman 1979)

$$\epsilon(\mathbf{r}) = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} \exp i(\mathbf{k} \cdot \mathbf{r} + \psi(\mathbf{k})), \quad (21)$$

where $\psi(\mathbf{k})$ is itself a random real function of \mathbf{k} , and $\epsilon_{\mathbf{k}}$ is a real positive number.

The random field $\epsilon(\mathbf{r})$ is real, therefore $\epsilon_{\mathbf{k}} = \epsilon_{-\mathbf{k}}$, and $\psi(\mathbf{k}) = -\psi(-\mathbf{k})$. Moreover, for a macroscopically isotropic system, we can take $\epsilon_{\mathbf{k}} = \epsilon_{|\mathbf{k}|}$. Consequently

$$I(\mathbf{q}_1, \mathbf{q}_2) = \epsilon_{|\mathbf{q}-\mathbf{q}_1|} \epsilon_{|\mathbf{q}-\mathbf{q}_2|} \exp i \Psi(\mathbf{q}, \mathbf{q}_1, \mathbf{q}_2), \quad (22)$$

where

$$\Psi(\mathbf{q}, \mathbf{q}_1, \mathbf{q}_2) \equiv \psi(\mathbf{q} - \mathbf{q}_1) - \psi(\mathbf{q} - \mathbf{q}_2).$$

It is obvious that the term I is a real positive number equal to $\epsilon_{|\mathbf{q}-\mathbf{q}_1|}^2$ for $\mathbf{q}_1 = \mathbf{q}_2$. Because of the random nature of the phase term ψ , $I(\mathbf{q}_1, \mathbf{q}_2)$ will exhibit random variation as a function of \mathbf{q}_1 and \mathbf{q}_2 , whenever $\mathbf{q}_1 \neq \mathbf{q}_2$. Therefore the statistically expected contribution to S of all those terms for which $\mathbf{q}_1 \neq \mathbf{q}_2$, can be neglected to get

$$S(\mathbf{q}, \omega) = \sum_{\mathbf{q}_1} \omega_{\mathbf{q}} \omega_1 G(\mathbf{q}_1, \mathbf{q}_2, \omega) \epsilon_{|\mathbf{q}-\mathbf{q}_1|}^2. \quad (23)$$

To arrive at this result, we have also replaced $(\mathbf{e}_q \cdot \mathbf{e}_1)^2$ by its angular average of $1/3$, and this factor gets cancelled by the factor 3 that would appear as a result of summation over the polarization branch index, ignoring the difference amongst the various branches. In order to justify neglecting the total contribution to S coming from terms with $\mathbf{q}_1 \neq \mathbf{q}_2$, we note that for a macroscopically homogeneous and isotropic system, $G(\mathbf{q}_1, \mathbf{q}_2)$ is expected to be dependent only on the angle between \mathbf{q}_1 and \mathbf{q}_2 . In other words, the absolute orientation of $(\mathbf{q}_1 - \mathbf{q}_2)$ would not appear in the function $G(\mathbf{q}_1, \mathbf{q}_2)$ except through a phase factor which would be physically unimportant. The summation notation in (19) essentially implies the process of summation over a set of six variables: $|\mathbf{q}_1|$, $|\mathbf{q}_2|$, θ , θ' , ϕ and ϕ' , where (θ, ϕ) and (θ', ϕ') define the orientation of \mathbf{q}_1 and \mathbf{q}_2 , respectively, with respect to an arbitrary spherical polar coordinate frame with polar axis along \mathbf{q} . Again the summation over ϕ and ϕ' is equivalent to that over $(\phi' - \phi)$ and ϕ . Now in the process of summation over ϕ , for a given set of values of all other variables (using $(\phi' - \phi)$ in place of ϕ'), the relative orientations of \mathbf{q} , \mathbf{q}_1 and \mathbf{q}_2 are not affected, and consequently factors like $\mathbf{e}_q \cdot \mathbf{e}_1$ appearing in a given term on the right side of (19) remain unaltered. Thus the summation over ϕ is operative essentially on the phase factor contained in $I(\mathbf{q}_1, \mathbf{q}_2)$ modified by a phase factor, if any arising from the functions $G(\mathbf{q}_1, \mathbf{q}_2)$. This sum is likely to reduce to zero in the limiting case when the number of terms to be summed becomes indefinitely large. Terms with $\mathbf{q}_1 = \mathbf{q}_2$ have ψ necessarily equal to zero, and hence their contribution is significant. The validity of (23) thus depends on our assumption that the phase terms ψ 's defined by (21) are totally uncorrelated.

The quantity $\epsilon_{|\mathbf{q}-\mathbf{q}_1|}$ can be obtained from the knowledge of the spatial correlation in elasticity fluctuation. Let us define a spatial correlation function $\Gamma(\mathbf{R})$ by

$$\epsilon_0^2 \Gamma(\mathbf{R}) = \int d^3 r \epsilon(\mathbf{r}) \epsilon(\mathbf{r} + \mathbf{R}), \quad (24)$$

$$\text{where } \epsilon_0^2 = \int d^3 r \epsilon^2(\mathbf{r}), \quad (25)$$

is the mean square value of $\epsilon(\mathbf{r})$. Then using (21) we get

$$\epsilon_{|\mathbf{k}|}^2 = \frac{1}{V} \Gamma(\mathbf{k}) \epsilon_0^2 \quad (26)$$

where $\Gamma(\mathbf{k})$ is the Fourier component of $\Gamma(\mathbf{R})$. It is easy to understand that $\Gamma(\mathbf{R})$ is unity for $\mathbf{R} = 0$ and it is expected to tend to zero as $\mathbf{R} \rightarrow \infty$. A model correlation that is simple to handle mathematically and which can describe a random system reasonably well can be given by (Joshi 1979; Ziman 1979)

$$\Gamma(\mathbf{R}) = \exp(-\lambda R), \quad (27)$$

where λ is a constant parameter that can be regarded as a measure of the ordering in the system. Roughly speaking the relevant property of the system can be taken to remain almost constant over a spatial extension of order $1/\lambda$. Using this expression for $\Gamma(\mathbf{R})$, one can show that

$$\epsilon_{|\mathbf{k}|}^2 = \frac{8\pi \lambda \epsilon_0^2}{V(\lambda^2 + k^2)^2}. \quad (28)$$

Replacing the summation over q_1 in (23) by an integration in the wave vector space, and using (28), we obtain

$$S(q, \omega) = \frac{2}{\pi} \epsilon_0^2 \lambda \omega_q \int dq_1 \int_0^\pi \sin \theta d\theta \frac{q_1^2 \omega_1 G(q_1)}{(\lambda^2 + |\mathbf{q} - \mathbf{q}_1|^2)^2} \quad (29)$$

where θ as defined earlier is the angle between \mathbf{q} and \mathbf{q}_1 . The lower limit of integration for \mathbf{q}_1 is zero and some suitable value has to be fixed for the upper limit. The system is macroscopically homogeneous and isotropic; we therefore take $G(q_1)$ to be angle independent. Integration over θ can easily be performed to obtain

$$S(q, \omega) = \frac{4}{\pi} \epsilon_0^2 \omega_q \lambda \int dq_1 \frac{q_1^2 \omega_1 G(q_1)}{(\lambda^2 + \mathbf{q}^2 + \mathbf{q}_1^2) - 4\mathbf{q}^2 \mathbf{q}_1^2}. \quad (30)$$

The upper limit of integration in this relation is not clearly definable. In the case of crystalline systems, one can define a Brillouin zone, and the upper limit of integration in their case essentially corresponds to the zone boundary. In fact no acoustic wave can exist with a wave vector larger than some sharply-defined upper limit, and consequently there is an upper limit of the associated phonon frequency. The situation is, however, not simple in the case of amorphous systems. In an ideal continuum there is no upper limit for the wave vector and therefore for the phonon frequency. In a real amorphous system vibrational modes with small wave length as compared to the separations between neighbouring atoms cannot really exist, yet there is no sharp cut-off. In other words, the density of the phonon states can be taken to be roughly a quadratic function of the wave vector up to some frequency ω_m and thereafter there is a rapid drop to an asymptotic value equal to zero. Here, ω_m is something analogous

to the mobility edge in the case of electronic states in disordered systems. Hence while writing the self-energy in terms of an integral in frequency space some factor has also to be included to take this nature of density of states into account. We will be interested in phonons having frequency sufficiently small compared with ω_m , and in that case one could ignore the trailing part of the density of states above ω_m . Thus we write

$$S(q, \omega) \approx \frac{4}{\pi} \epsilon^2 \omega_0 \omega_q \int d\omega_1 \frac{\omega_1^3 G(q_1, \omega)}{(\omega_0^2 + \omega_q^2 + \omega_1^2)^2 - 4\omega_q^2 \omega_1^2}, \quad (31)$$

where a linear relation between frequency and wave vector has been assumed, that is $\omega_q = vq$, v being the phonon velocity; and $\omega_0 = v\lambda$. We now apply an iterative method to calculate S . To the lowest order, S can be obtained by using G_0 for G in the above equation, remembering that an infinitesimal imaginary part is implied to be contained in ω . To refer to this approximation we use a naught as a subscript. The result is

$$\begin{aligned} \text{Re } S_0 = \frac{\epsilon_0^2 \omega_0}{2\pi \mathcal{D}} \left\{ B \ln \left[\frac{\omega_m + \omega}{\omega_m - \omega} \right] + C \ln \left[\frac{\omega_0^2 + (\omega_m - \omega_q)^2}{\omega_0^2 + (\omega_m + \omega_q)^2} \right] \right. \\ \left. + \frac{2C\omega_q + D}{\omega_0} \left[\tan^{-1} \frac{\omega_m - \omega_q}{\omega_0} + \tan^{-1} \frac{\omega_m + \omega_q}{\omega_0} \right] \right\}, \quad (32) \end{aligned}$$

$$\text{and } \text{Im } S_0 = -2\epsilon_0^2 \omega^3 \omega_0 \omega_q / \mathcal{D}, \quad (33)$$

$$\text{where } B = 4\omega^3 \omega_q, \quad (34a)$$

$$C = 4\omega^2 \omega_q^2 - (\omega_0^2 + \omega_q^2)(\omega^2 + \omega_0^2 + \omega_q^2), \quad (34b)$$

$$D = 4\omega^2 \omega_q (\omega_0^2 + \omega_q^2), \quad (34c)$$

$$\mathcal{D} = (\omega^2 + \omega_0^2 + \omega_q^2)^2 - 4\omega^2 \omega_q^2. \quad (34d)$$

4. Life time

To the lowest order, the phonon life time τ_0 can be given by $\text{Im } S_0$ evaluated at $\omega \approx \omega_q$. One obtains

$$\tau_0^{-1} = 2\epsilon_0^2 \omega_q^4 / \omega_0 (\omega_0^2 + 4\omega_q^2). \quad (35)$$

The real and imaginary parts of S can be now used in (16) to obtain an improved expression for the phonon Green's function, which would offer a better approximation for use in (31). We will use a subscript 1 to denote the resulting self-energy and other quantities. In calculating $\text{Im } S_1(q, \omega \approx \omega_q)$ analytically, we note that an important contribution to the integral in (31) comes from the values of ω_1 in the neighbourhood of ω_q . Therefore a convenient and reasonable approximation is to write ω_q for ω_1 in the expression for $S_0(q_1, \omega)$ which is then used in $G(q_1, \omega)$. For this purpose we define dimensionless quantities f_R and f_I :

$$\omega_q (f_R - if_I) = S_0(q_1, \omega_1) |_{\omega_1 = \omega_q}. \quad (36)$$

If ϵ_0^2 be assumed to be sufficiently small then f_I and f_R would also be small compared with unity for phonon frequencies of our interest. With a little algebra the corrected life time τ_1 can be given by

$$\tau_1^{-1} = \frac{\epsilon_0^2 \omega_0}{2\pi F^{3/2} (1 + f^2)} \sum_{j=1} \text{Im } \mathcal{J}_j, \quad (37)$$

where
$$\mathcal{J}_1 = \int_0^{\omega_m} d\omega_1 \frac{\omega_1^2 (1 + if)}{[\omega_0^2 + (\omega_1 - \omega_q)^2] [\omega_1 - \omega_q - if \omega_1]}, \quad (38)$$

and the integral expression for \mathcal{J}_2 can be obtained from the above by replacing $(\omega_1 - \omega_q - if \omega_1)$ with $(\omega_1 + \omega_q - if \omega_1)$. The integrals \mathcal{J}_3 and \mathcal{J}_4 differ from \mathcal{J}_1 and \mathcal{J}_2 , respectively, in having a negative sign and also the factor $[\omega_0^2 + (\omega_1 - \omega_q)^2]$ replaced by $[\omega_0^2 + (\omega_1 + \omega_q)^2]$. Here we have defined

$$F = 1 + f_R, \quad (39a)$$

and
$$f = f_I / 2F. \quad (39b)$$

In integrals \mathcal{J} 's a factor $F^{1/2}$ has been absorbed in the frequency terms ω_0 , ω_q and ω_1 . This implies that the effective relation between ω_q and \mathbf{q} is now $\omega_q \approx |\mathbf{q}| v F^{1/2}$. $v F^{1/2}$ can thus be interpreted as the effective velocity in the presence of perturbation, and would be slightly different from v that was defined for the starting unperturbed phonon system. This step does not introduce significant error in computing τ_1 .

The integrals \mathcal{J}_j can be evaluated analytically without much difficulty. If one defines

$$x = \omega_q / \omega_0, \quad (40a)$$

and
$$x_m = \omega_m / \omega_0, \quad (40b)$$

then the final result can be obtained as

$$\tau_1^{-1} = \frac{\epsilon_0^2 \omega_0 f (T_1 - T_2)}{2 F^{3/2} (1 + f^2)}, \quad (41)$$

where

$$\begin{aligned} T_1 = & A_1 \ln \left[\frac{1 + (x_m - x)^2}{1 + (x_m + x)^2} \right] + 2(B_1 + A_1 x) \left[\pi - \tan^{-1} \frac{2x_m}{x^2 - x^2 - 1} \right] \\ & + \frac{C_1}{1 + f^2} \ln \left[\frac{(x_m - x)^2 + f^2 x_m^2}{(x_m + x) + f^2 x_m} \right] \\ & + \frac{2}{f} \left(D_1 + \frac{C_1}{1 + f^2} \right) \left[\pi - \tan^{-1} \frac{2fx x_m}{(1 + f^2) x_m^2 - x^2} \right], \end{aligned} \quad (42a)$$

and

$$\begin{aligned}
 T_2 = & \frac{A_2}{2} \ln \left[\frac{[(x_m - x)^2 + 1] [(x_m + x)^2 + 1]}{(x^2 + 1)^2} \right] \\
 & + (B_2 + A_2 x) \left[2 \tan^{-1} x - \tan^{-1} \frac{2x}{(x_m^2 - x^2 + 1)} \right] \\
 & + \frac{C_2}{2(1 + f^2)} \ln \{ x^{-4} [(x_m - x)^2 + f^2 x_m^2] [(x_m + x)^2 + f^2 x_m^2] \} \\
 & + \frac{1}{f} \left(D_2 + \frac{C_2}{1 + f^2} \right) \left[2 \tan^{-1} \frac{1}{f} - \tan^{-1} \frac{2fx^2}{(1 + f^2)^2 x_m^2 - x^2(1 - f^2)} \right] \quad (42b)
 \end{aligned}$$

Here A_1, B_1 , etc. are given by

$$A_1 = \frac{1}{\mathcal{D}_1} [1 - 3x^2 + f^2(1 + x^2)] + \frac{1}{\mathcal{D}_2} [1 + 5x^2 + 8x^4 + f^2(1 + x^2)^2],$$

$$B_1 = 2x(1 + x^2) \left[\frac{1}{\mathcal{D}_1} - \frac{1 + 2x^2}{\mathcal{D}_2} \right],$$

$$C_1 = x^2 [3 - f^2(1 + x^2)] \left[\frac{1}{\mathcal{D}_1} - \frac{1}{\mathcal{D}_2} \right] - \frac{8x^5}{\mathcal{D}_2},$$

$$D_1 = 2x^2 \left[\frac{1 + 2x^2}{\mathcal{D}_2} - \frac{1}{\mathcal{D}_1} \right],$$

$$A_2 = D_1,$$

$$B_2 = x(1 + x^2) \left[\frac{1 + f^2(1 + x^2)}{\mathcal{D}_1} + \frac{1}{\mathcal{D}_2} \right],$$

$$C_2 = 2x^2 (1 + f^2) \left[\frac{1}{\mathcal{D}_1} - \frac{1 + 2x^2}{\mathcal{D}_2} \right],$$

$$D_2 = x^2 [1 + f^2(1 + x^2)] \left[\frac{1}{\mathcal{D}_2} - \frac{1}{\mathcal{D}_1} \right],$$

where $\mathcal{D}_1 = [1 + f^2(1 + x^2)]^2 - 4f^2 x^2$,

$$\mathcal{D}_2 = (1 + 4x^2)^2 + 2f^2(1 + x^2)(1 + 2x^2) + f^4(1 + x^2).$$

In the limiting case when $f \gg 1$, τ_1 reduces to τ_0 .

5. Discussion

The expression for τ_0 given by (35) is essentially the same as obtained earlier in connection with the thermal conductivity analysis of vitreous silica (Joshi 1979). The inverse life time τ_0^{-1} is observed to vary as the fourth power of frequency for values of the latter which are small compared with ω_0 . With the increase in frequency, there is a weakening in the dependence of τ_0 on frequency, such that for $\omega_q \gg \omega_0$, τ_0^{-1} is practically a quadratic function of ω_q . This nature of τ_0 was found to give a satisfactory explanation of the temperature-dependence of conductivity in and around the plateau region where thermal phonons have frequencies of order ω_0 . It is also noted that the parameter ϵ_0^2 can be of the order of 5% and the parameter $1/\lambda$ can be of the order of 20 Å. This has the implication that the elasticity of the medium can be treated to remain almost constant over a region of about 20 Å in dimension. The corresponding value of ω_0 is of the order of 3.0×10^{12} rad. sec⁻¹ in a typical glassy solid. In figure 1 we present some numerical calculations of the phonon life time as given by (41), using $\omega_0 = 3.0 \times 10^{12}$ rad. sec⁻¹ and $x_m = 20$, which are the typical values for a solid like vitreous silica. It is observed that the frequency dependence of τ_1 and τ_0 are essentially the same for $\omega_q \lesssim \omega_0$. There is a slight difference in magnitude between these two depending on the value of ϵ_0^2 . The difference increases with increasing ϵ_0^2 and is not noticeable for $\epsilon_0^2 = 0.01$. We also note that, depending on the value of ϵ_0^2 , there is some frequency larger than ω_0 above which τ_1 shows a much weaker dependence on ω_q than τ_0 . This in fact defines

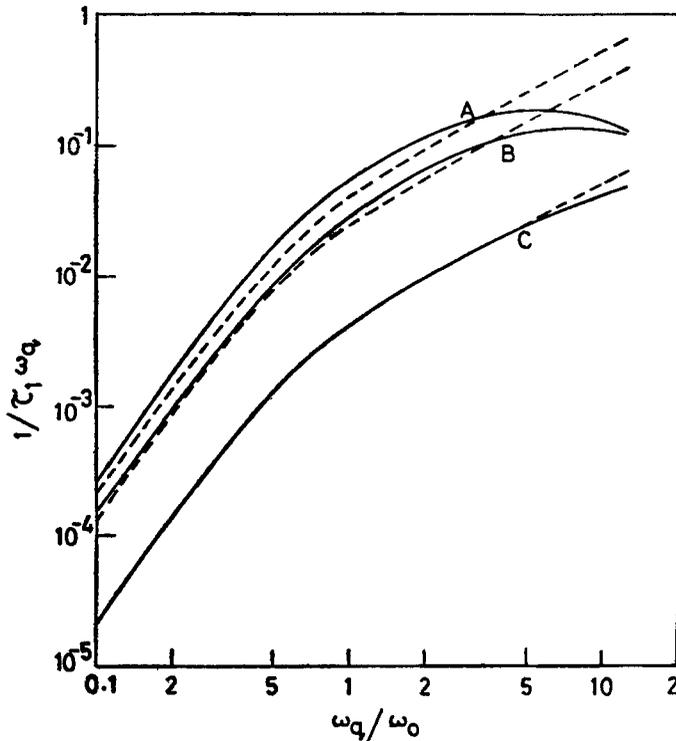


Figure 1. Typical variation of $1/\tau_1\omega_q$ (solid line) with phonon frequency. Parameters used are $\omega_0 = 3.0 \times 10^{12}$ sec⁻¹, $x_m = 20$ and $\epsilon_0^2 = 0.10$ (A), 0.06 (B), and 0.01 (C). Broken curve represents $1/\tau_0\omega_q$.

the frequency above which the corrected life time would be appreciably different from the one obtained by first-order perturbation calculation, that is τ_0 . In this region the life time tends to a practically constant value, with the rise in ω_q . This result is perhaps valid qualitatively (Ziman 1960). We cannot, however, be sure of the quantitative validity of this calculation as the present continuum model would not offer a good description of the dynamics at higher frequencies.

For frequencies in the neighbourhood of ω_0 , the life time can be approximately represented by a function of the type ω_q^n , where n is very nearly equal to 3. Recent ultrasonic attenuation studies by Dietsche and Kinder (1979) in vitreous silica at frequencies in the vicinity of 200 GHz have also indicated this kind of frequency dependence of the life time. In fact, they have suggested that the phonon mean free path can be satisfactorily given by

$$l = (10 \pm 1) \nu^{-2.9 \pm 0.3} \mu\text{m}$$

where ν is the frequency in units of 100 GHz. Thus the mean free path at 200 GHz is something between 0.98 and 1.81 μm and similarly at 500 GHz it lies between 0.052 and 0.168 μm . Present calculations give us 0.63 μm and 0.055 μm for the mean free path at these frequencies, respectively, when we use $\nu = 4.1 \times 10^5 \text{ cm sec}^{-1}$, $1/\lambda = 15 \text{ \AA}$ and $\epsilon_0^2 = 0.053$. These are the values of parameters that have yielded good results in the case of conductivity analysis (Joshi 1979). The difference between the experimental results and the theory is tolerable.

It is possible to perform analytic calculations for τ_0 , using a more complex model expression for the correlation function $\Gamma(\mathbf{R})$. However, for a correlation that is unity for small \mathbf{R} and falls rapidly to a zero value with increasing \mathbf{R} , the frequency dependence of the life time would not be very different from what is obtained above. In conclusion, we may mention that for physically reasonable values of the parameter ϵ_0^2 , the corrected expression τ_1 for the inverse life time does not differ appreciably from τ_0 for frequencies comparable to or less than ω_0 . Further, other scattering mechanisms (such as LTS) with comparatively weaker frequency dependence would generally be more important in the case of very low frequency phonons, since in that case the present inverse life time exhibits a quartic dependence on frequency.

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