Spectral diffusion of phonons in glasses

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Because of a spectral diffusion, phonons generated in a glass by a monochromatic signal spread out over the spectrum. As a result, a wider hole is burnt out, and the phonon bottleneck is greatly modified.

The resonant absorption of sound or of microwaves in an insulating glass at low temperatures is known to be a consequence of so-called two-level systems.[1] After a two-level system absorbs a quantum of sound or of the microwave field, $\hbar\omega$, it generally emits a phonon of the same or of nearly the same energy. The subsequent fate of these resonant phonons depends on the relation between the two times τ_r and τ_{nr}. The first time is the lifetime of a phonon with respect to its resonant absorption by a two-level system of the same energy: $\tau_r^{-1} = (\pi PM^2\Omega/\rho v^2)\tanh(\hbar\Omega/2T)$. Here Ω is the phonon frequency, T is the temperature, P is the state density of the two-level systems in the glass, M is the transition matrix element, ρ is the density of the glass, and v is the sound velocity. The second time is the lifetime of a phonon with respect to its nonresonant absorption by thermal two-level systems with an energy on the order of[1] $T(T\gg\hbar\Omega):\tau_{nr}^{-1} = \pi^3PD^2M^2T^3/32\rho^2\hbar^4v^7$, where D is the strain-energy constant.

The ratio of these times is $\tau_{nr}/\tau_r \simeq E_c^2(\hbar\Omega)^2/T^4$, where $E_c = (\rho v^5\hbar^3)^{1/2}/D$ is a characteristic energy, on the order of 10–30 K in glasses.[2] Under the conditions $T\ll E_c$ and $\hbar\Omega\gg T^2/E_c$, the relation $\tau_{nr}\gg\tau_r$ holds even at frequencies in the classical region[1)] $(\hbar\Omega\ll T)$. In this case, the resonant phonons with frequency $\Omega\approx\omega$ have a tendency to accumulate. In the case of a strong pulsed excitation, the results will be the appearance of a phonon bottleneck in the nonlinear resonant absorption.[3]

As we will show below, however, in the case of a weak steady-state excitation the accumulation of phonons in the narrow resonant region is hindered by spectral diffusion.[4] Although the resonant phonons are reabsorbed many times by resonant two-level states during the lifetime τ_{nr}, changes which occur randomly in time in the distance between levels in the two-level systems [because of transitions (jumps) in the thermal two-level systems] lead to the emission of a phonon at the same frequency as that of the phonon that is absorbed; i.e., the nonequilibrium phonon distribution spreads out over the spectrum. This effect causes changes in both the threshold for the appearance of a nonlinearity in the resonant absorption and the width of the hole which is burnt out.[5]

The diagonal part (n_s) of the density matrix of the s-th resonant two-level system, the nondiagonal part $[if_s\exp(i\omega t)]$, and the phonon distribution function N_k are determined by the following system of equations:

$$\frac{\partial n_s}{\partial t} = \frac{2\pi}{\hbar}\sum_{\mathbf{k}}\Lambda_k\,[N_k - n_s(2N_k+1)]\,\delta(e_s(t) - \hbar\Omega_k) - F\,\mathrm{Re}f_s - \frac{n_s - n_s^0}{\tau'}, \quad (1a)$$

$$\frac{\partial f_s}{\partial t} + i\left(\omega - \frac{e_s(t)}{\hbar}\right) f_s + f_s \frac{\pi}{\hbar} \sum_k \Lambda_k (2N_k + 1)\delta(e_s(t) - \hbar\Omega_k) = F\left(n_s - \frac{1}{2}\right), \quad (1b)$$

$$\frac{\partial N_k}{\partial t} + \frac{N_k - N_k^0}{\tau_{nr}} + \frac{2\pi}{\hbar} \sum_s \Lambda_k [N_k (1 - 2n_s) - n_s]\delta(e_s(t) - \hbar\Omega_k) = 0. \quad (1c)$$

Here $\hbar F/2$ is the transition matrix element which characterizes the interaction of the signal with the resonant two-level system, N_k^0 is the equilibrium phonon distribution function, Ω_k is the frequency of a phonon with a wave vector **k**, $\Lambda_k = \hbar k^2 M^2 / 2\rho V \Omega_k$ is the square of the matrix element of the interaction of the two-level systems with the phonons, V is the volume, n_s^0 is the equilibrium filling of the upper level by the resonant two-level system, and τ' is the time scale of the relaxation of the population of the resonant two-level system due to two-phonon processes.[6] The time-dependent distance between the levels of the s-th resonant two-level system is $e_s(t) = e_{0s} + \hbar\Delta\omega_s(t)$, where $\hbar\Delta\omega_s(t) = \hbar\Sigma_l J_l \xi_l(t)$ is the increment in the "seed" energy e_{0s} which results from the interaction of the resonant two-level system with the thermal two-level systems surrounding it (the summation is carried out over all of the thermal two-level systems). In addition, we have $J_l = D^2/\hbar\rho v^2 r_l^3$, where r_l is the distance from the l-th thermal two-level system to the given resonant system. The function $\xi_l(t)$ is a "telegraph" process.[7] It takes on the values 1 and -1 alternately at random times; the average frequency of these switches is Γ_l. The various functions ξ_l are assumed to be uncorrelated.

Assuming that the pump amplitude F is small, we find from (1c) a relationship between the correction to the phonon distribution function, ΔN_k, and the average change in the population of the upper level of the resonant two-level systems, $\Delta n(\Omega)$:

$$\Delta N_k = \frac{\tau_{nr}}{\tau_r + \tau_{nr}} \Delta n(\Omega_k) \coth^2 \frac{\hbar\omega}{2T}, \quad (2)$$

where

$$\Delta n(\Omega) = P^{-1} V^{-1} \hbar^{-1} \sum_s \Delta n_s \delta(e_s(t)/\hbar - \Omega). \quad (3)$$

The function $\Delta n(\Omega)$ describes the burnt-out hole, i.e., the change in the absorption coefficient for a weak signal at the frequency Ω in the presence of a pump signal at the frequency ω (Ref. 5). If the pump is weak, the distribution function of the nonequilibrium thus reproduces the shape of the burnt-out hole.

From (1) we find the following integral equation for $\Delta n(\Omega)$:

$$\Delta n(\Omega) = \int_{-\infty}^{+\infty} d\Omega' R(\Omega - \Omega') \Delta n(\Omega') + I(\Omega - \omega), \quad (4)$$

where

$$R(x) = \frac{\nu}{\pi} \int_0^{\infty} d\tau \cos x\tau \int_0^{\infty} d\tau' e^{-\gamma'\tau'} \langle B(\tau, \tau') \rangle_c \quad (5)$$

and

$$I(x) = \frac{1}{2}F^2 \tanh \frac{\hbar\omega}{2T}\int_0^\infty d\tau e^{-(\gamma/2)\tau}\cos x\tau \int_0^\infty d\tau' e^{-\gamma\tau'}\langle L(\tau,\tau')\rangle_c . \quad (6)$$

Here γ^{-1} is the equilibrium relaxation time of the population of the resonant two-level system for the relaxation caused by single-phonon processes; $\gamma' = \gamma + 1/\tau'$, $\nu = \gamma\tau_{nr}/(\tau_r + \tau_{nr})$; the angle brackets, $\langle\cdots\rangle_c$, mean an average over the parameters and position of the thermal two-level systems; and $B(\tau,\tau')$ is the product of independent averages, each corresponding to one thermal two-level system and is given by

$$B(\tau,\tau') = \langle \exp[iJ\tau(\xi(\tau') - (0))]\rangle_{\xi,\,\xi(0)} . \quad (7)$$

The average is taken over all relations of the random process $\xi(t)$ at a given value of $\xi(0)$ at an arbitrary time $t = 0$ and over all $\xi(0)$. The function $I(x)$ describes the shape of the burnt-out hole in the case in which phonons do not accumulate ($\tau_{nr} \ll \tau_r$). That function and also the function $L(\tau,\tau')$ were calculated in Ref. 5.

Carrying out the averaging in (7), with the help of the procedure used in Ref. 7, we find

$$B(\tau,\tau') = 1 - (1 - e^{-2\Gamma\tau'})\sin^2 J\tau . \quad (8)$$

Now taking an average over all possible positions of the thermal two-level systems[8] and over the values of their tunneling transparency, on which the transition frequencies Γ depend, we find

$$\langle B(\tau,\tau')\rangle_c = \exp\left[-\frac{\pi\tau}{2\tau_d}\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}\left(1 - e^{-2\Gamma\tau'}\right)\right]. \quad (9)$$

Here Γ_0 is the characteristic frequency of transitions of thermal two-level systems occurring as a result of interactions with thermal phonons; this frequency is equal in order of magnitude to the quantity $\Gamma_0 \simeq M^2T^3/\rho\hbar^4v^5$. The quantity $\hbar/\tau_d = D^2PT/\rho v^2$ is the characteristic energy of the interaction of the thermal two-level systems when they are separated by an average distance $\bar{r} = (PT)^{-1/3}$.

Equation (4) has been solved by a Fourier method. Here we will present the results of the calculation in one of the limiting cases—that of low temperatures ($T \ll T_D \simeq (P\hbar^3v^3)^{1/2} \simeq 1K$, i.e., $\Gamma_0\tau_d \ll 1$), in which we have $\hbar\omega \ll T$. For the increment to the phonon distribution function we find

$$\Delta N_k = \frac{2\nu\tau_d F^2}{\gamma^2\ln\Gamma_0/\gamma}\tan\frac{\hbar\omega}{2T}\int_0^\infty dx\cos\left(\frac{\omega - \Omega_k}{\Delta_0}x\right)\frac{e^{-x}}{1 - \frac{\nu}{\gamma'}e^{-x}} , \quad (10)$$

where $\Delta_0 = [\pi\ln(\Gamma_0/\gamma)]/2\tau_d$. In the case $\nu \ll \gamma'$ ($\tau_{nr} \ll \tau_r$), phonons do not accumulate, and ΔN_k is described as a function of the frequency deviation $\omega - \Omega_k$ by a Lorentzian curve with a half-width Δ_0. If we have $1 - \nu/\gamma' \ll 1$ ($\tau_{nr} \gg \tau_r$), on the other hand, the phonons are reabsorbed many times by resonant two-level systems. The dependence of ΔN_k on the frequency deviation is very non-Lorentzian in this case. The effective width of the distribution is $\Delta_0(\tau_{nr}/\tau_r)$—larger by a factor of τ_{nr}/τ_r than in the absence of an accumulation of phonons. The phonons spread out markedly over the spectrum as a result of spectral diffusion.

At small values of F, the nonlinear absorption coefficient α can be written $\alpha = \alpha_0(1 - F^2/F_c^2)$, where α_0 is its linear value. The crystal value of the signal amplitude, F_c, was calculated by the same method. Under the condition $\tau_{nr} \gg \tau_r$, we find $F_c^2 = (\pi\gamma \ln\Gamma_0/\gamma)/2\tau_d \ln(\tau_{nr}/\tau_r)$ in our case; i.e., this quantity decreases with increasing τ_{nr} only in proportion to the reciprocal of a logarithm (not in proportion to $1/\tau_{nr}$, as in the case of the phonon bottleneck[9]). Again, this result is a consequence of the spreading out of the phonon distribution along the energy scale. It follows from (10) that the number of phonons in the resonance region ($|\omega - \Omega_k| < \Delta_0$) increases with increasing τ_{nr}/τ_r only in proportion to $\ln(\tau_{nr}/\tau_r)$.

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[1)]In the quantum-mechanical case, with $\hbar\Omega \gtrsim T$ (but $\hbar\Omega \ll E_c$), the relation $\tau_{nr} \gg \tau_r$ wil always hold.[2]

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