Revised theory of relaxation ultrasound attenuation in glasses

B.D. Laikhtman

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The effect of phonon disturbance by two-level systems on ultrasound attenuation and velocity is considered. It is shown that the phonon distribution function is substantially disturbed when the ultrasound frequency is much less than the inverse two-level system and phonon relaxation times. In such a case, the ultrasound attenuation appears to be much greater than Jäckle's theory predicts. The transition to the phenomenological theory of ultrasound propagation is discussed. A new mechanism of phonon energy relaxation due to spectral diffusion is shown.

I. INTRODUCTION

Nearly all the thermodynamic and kinetic properties of glasses at low temperatures are rather well described by the two-level-system model.^{1–3} According to this model, glass contains a great number of localized two-level systems (TLS's) with a very broad distribution of energy splitting.^{4,5} TLS's form the main contribution to the specific heat of glass at low temperatures and such kinetic properties as ultrasonic and microwave attenuation as well. The only relaxation mechanism of TLS's is the emission and absorption of phonons, which are considered to be in thermodynamic equilibrium. The last assumption is the least well-founded one. The relaxation process connects TLS's and phonons, so that a nonequilibrium ensemble of TLS's would disturb an equilibrium phonon distribution function. The disturbance might be neglected if there were another effective relaxation mechanism to reduce the phonon distribution function to its equilibrium form. But it is well known (e.g., from ultrasonic experiments) that the main relaxation mechanism of the phonon distribution function is connected with the absorption and emission of phonons by TLS's. So TLS's and phonons should be considered as a united system, and in general any external force disturbs the system as a whole. Such a situation is known in the theory of paramagnetic $6-8$ and paraelectric⁹ resonance as a phonon bottleneck. The purpose of this paper is to explore how a deviation of the phonon distribution function from its equilibrium form affects the attenuation and velocity of small-amplitude ultrasound in glasses.

A TLS can be imagined as an atom or a group of atoms tunneling through a potential barrier between two potential energy minima, though its microscopic model is of no importance to the thermodynamic and kinetic properties of glasses. It is enough to consider the TLS model as a phenomenological one and to characterize a TLS by an energy spacing Δ of levels in isolated potential wells and a tunneling coupling energy $\Delta_0 = \hbar \omega_0 e^{-\lambda}$ where $\hbar \omega_0$ is a typical zero-point energy in either well and λ is a tunneling integral, so that the Hamiltonian of a TLS has the form

$$
\mathscr{H}_0 = \frac{1}{2} \begin{bmatrix} \Delta & \Delta_0 \\ \Delta_0 & -\Delta \end{bmatrix} . \tag{1}
$$

Diagonalizing \mathcal{H}_0 , one can describe a TLS by the energy splitting of its levels $E = (\Delta^2 + \Delta_0^2)^{1/2}$ and a parameter ϑ : $\Delta = E \cos\vartheta$, $\Delta_0 = E \sin\vartheta$. A state of a TLS can be described by the populations n of the upper level and $1-n$ of the lower level. In thermodynamic equilibrium the mean upper-level population is

$$
n_0 = (e^{E/T} + 1)^{-1}, \tag{2}
$$

where T is the temperature. Interaction of an ultrasound wave and phonons with a TLS is usually described by the Hamiltonian

$$
\mathcal{H}_1 = \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix} \gamma_{ij} u_{ij} , \qquad (3)
$$

where γ_{ij} is a deformation potential tensor, and u_{ij} is a local strain.

The theory of ultrasound propagation in glasses was formulated by Jäckle¹⁰ (see also Refs. $11-13$). He found two contributions from TLS's to ultrasound attenuation and velocity. The first is the resonance contribution. It results from the absorption of ultrasound quanta by the TLS's whose energy splitting E is equal to $\hbar\omega$ (ω is the sound frequency). The absorption probability depends on a mean TLS population n . Thus for resonance attenuation, at least in a linear approximation, the deviation of a phonon distribution function from thermodynamic equilibrium can be neglected.

The second contribution to ultrasound attenuation is a nonresonant or relaxation one. It results from the fact that an ultrasound wave periodically modulates the energy splittings of TLS's and so shifts their equilibrium state. Due to TLS—phonon relaxation, ^a phonon distribution function becomes periodically modulated too. Naturally, a considerable disturbance of the phonon distribution function can be expected if the average phonon relaxation time $\bar{\tau}$ is much smaller than the period of ultrasound wave $2\pi/\omega$. In such a case, phonon occupation numbers relax to the values corresponding to the current TLS population $n(E,\vartheta)$ before $n(E,\vartheta)$ changes its value. That is, if $\omega \bar{\tau} \ll 1$, the phonon system is in a quasiequilibrium state and deviation of the phonon occupation numbers from their thermodynamic equilibrium values can be expected to have the same order of magnitude as the deviation n_1 of n from its equilibrium form (2). To be more precise, one should recall that phonons with the same wave number k can be emitted and absorbed by TLS's with different parameters ϑ . Thus, the phonon distribution function feels only some sort of an average of the TLS population over ϑ , \bar{n} , and its deviation from the equilibrium form (2) is proportional to $\overline{n}_1(E)$.

On the other hand, TLS's relax only by emitting and absorbing phonons, and if the phonons are not in thermodynamic equilibrium, the TLS's relaxation brings their population not to the equilibrium value (2), but to some other value corresponding to the current phonon distribution function. In other words, the deviation $n_1(E,\vartheta)$ relaxes not to zero but to $\overline{n}_1(E)$. Such a result is due to the fact that the TLS—phonon relaxation takes place for each energy E separately and does not allow for any change in the energy distribution. For low frequencies a value of $\overline{n}_1(E)$ is limited from above only by the minimum of the values $(\omega \overline{\tau})^{-1}$ and $(\omega \tau_0)^{-1}$, where τ_0 is a characteristic TLS relaxation time. The nonequilibrium population $n_1(E,\vartheta)$ and therefore ultrasound attenuation coefficients acquire the same factor. Hence, for small $\omega\tau_0$ and $\omega\bar{\tau}$ ultrasound absorption is much greater than Jäckle's theory predicts.

II. EVALUATION OF ULTRASOUND ATTENUATION AND VELOCITY

The exact calculation of the relaxation ultrasound attenuation and the velocity shift is based on solving a coupled system of kinetic equations for the TLS mean population n ,

$$
\frac{\partial n}{\partial t} = \frac{\pi E^3 \sin^2 \vartheta}{\rho \hbar^4} \sum_{s} \frac{\gamma_s^2}{w_s^5} \int \frac{d\Omega_k}{(2\pi)^3} [(1-n)N_s - n(N_s+1)]_{k=E_V/\hbar w_s}
$$
(4)

and the phonon occupation numbers N_s ,

$$
\frac{\partial N_s}{\partial t} + w_s \cos \theta \frac{\partial N_s}{\partial x} = \frac{\pi P \gamma_s^2}{\rho w_s} k \int_0^{\pi/2} \left[n (N_s + 1) - (1 - n) N_s \right]_{E_V = \hbar w_s} k \sin \theta \, d\theta \quad . \tag{5}
$$

Here $E_V = [(\Delta + 2\gamma V)^2 + \Delta_0^2]^{1/2}$ is the TLS energy splitting in the presence of ultrasound wave strain V ; w_s is a sound velocity ($s = l$ for a longitudinal mode, and $s = t$ for a transverse mode); γ_s are effective deformation constants:

$$
\gamma_l^2 = \frac{1}{15} \left[2 \, \text{tr} \gamma^2 + (\text{tr} \gamma)^2 \right] \,,
$$

$$
\gamma_l^2 = \frac{1}{30} \left[3 \, \text{tr} \gamma^2 - (\text{tr} \gamma)^2 \right]
$$
 (6)

(see Ref. 1); ρ is a mass density and θ is the angle between the directions of the phonon wave vector \vec{k} and the ultrasound wave propagation $(x \text{ axis})$. The integration on the right-hand side of Eq. (4) is performed over all directions of \vec{k} . The TLS parameters Δ and λ are considered to be uniformly distributed over intervals much greater than those important for low-temperature kinetics, $^{10, 1, 14, 15}$ so the TLS density per unit energy splitting $(0 \le E < \infty)$ interval and per unit parameter ϑ ($0 \le \vartheta \le \pi/2$) interval is taken as $P/\sin\theta$, P=const.

In the linear approximation $V \propto e^{i(qx - \omega t)}$, $\omega = wq$, and one can put $E_V \simeq E + 2\gamma \cos\theta \text{Re}V$,

$$
n = n_0 + n_1 e^{i(qx - \omega t)}, \tag{7}
$$

$$
N_s = (e^{\hbar w_s k/T} - 1)^{-1} + N_{s1} e^{i(qx - \omega t)}, \qquad (8)
$$

so that Eqs. (4) and (5) become

 \mathbf{r}

$$
-i\omega n_1 = \frac{1}{\tau} \left[2\frac{dn_0}{dE} \gamma V \cos\theta - n_1 + 2\pi^2 \tanh^2\left(\frac{E}{2T}\right) \sum_s \xi_s \int N_{s1} \frac{d\Omega_k}{(2\pi)^3} \right], \quad (9)
$$

 $i(\omega - q w_s \cos \theta) N_{s_1}$ $-\left|\coth^2\left|\frac{E}{2T}\right|\int_0^{\pi/2} n_1 \sin\vartheta d\vartheta - N_{s1}\right|$ (10)

Here

$$
\tau_s = \frac{\rho w_s}{\pi P \gamma_s^2 k} \coth\left(\frac{\hbar w_s k}{2T}\right) \tag{11}
$$

is the relaxation time of the s-phonon mode,

$$
\tau = \frac{\tau_0}{\sin^2 \theta} \left(\frac{T}{E} \right)^3 \tanh \frac{E}{2T},
$$

\n
$$
\tau_0 = \frac{2\pi \rho \hbar^4}{\sigma^3} \left[\sum_{s} \frac{\gamma_s^2}{s} \right]^{-1},
$$
\n(12)

$$
\xi_s = \frac{\gamma_s^2}{w_s^5} \left[\frac{\gamma_l^2}{w_l^5} + 2 \frac{\gamma_l^2}{w_t^5} \right]^{-1} . \tag{13}
$$

The elastic equation for an ultrasound wave in glass has the form

$$
\rho \frac{\partial^2 u}{\partial t^2} = \frac{\partial}{\partial x} \left[c \frac{\partial u}{\partial x} + \sigma' \right],
$$
 (14)

where u is an elastic displacement ($V = \partial u / \partial x$), $c = \rho w_0^2$ is an elastic modulus, and

$$
\sigma' = P \int_0^\infty dE \int_0^{\pi/2} \frac{d\vartheta}{\sin \vartheta} \langle (2n-1)\gamma \rangle \cos \vartheta \tag{15}
$$

is the contribution of TLS's to the elastic stress, where angular brackets represent an averaging over all orientations of a TLS relative to the ultrasound direction and polarization. In the linear approximation

$$
\sigma' = 2P \int_0^\infty dE \int_0^{\pi/2} \langle \gamma n_1 \rangle \cot \vartheta \, d\vartheta \; . \tag{16}
$$

Substituting Eq. (7) in (15), the first term leading to a constant stress has been omitted.

Using Eqs. (14) and (16) the ultrasound attenuation coefficient α and the contribution of the TLS to the ultrasound velocity $\Delta w = w - w_0$ can be written as

$$
\alpha = 2 \operatorname{Im} \Delta q, \quad \frac{\Delta w}{w} = -\frac{\operatorname{Re} \Delta q}{q} \tag{17}
$$

Here

$$
\Delta q = -\frac{P(\gamma^2)}{\rho w^2} q \int_0^\infty \frac{dn_0}{dE} dE \int_0^{\pi/2} g(E, \vartheta) \cot \vartheta d\vartheta , \quad (18)
$$

$$
n_1 = \gamma V \frac{dn_0}{dE} g(E, \vartheta) , \qquad (19) \qquad \tau_m = \tau_0 \left[\frac{T}{F} \right]^3 \tanh \left[\frac{E}{2T} \right]
$$

where $\langle \gamma^2 \rangle$ is equal to γ_l^2 for longitudinal and equal to γ_l^2 for transverse ultrasound.

Equations (9) and (10) form a system of linear integral equations with degenerated nuclei which can be solved analytically. The exact analytical solution contains rather cumbersome expressions useful only for' numerical calculations. To realize the physical contents of the results and to obtain simple expressions and estimations, it is more convenient to consider different limiting cases. Such a program is realized in the following.

From Eq. (10) one has

$$
N_{s1} = \frac{\coth^2(E/2T)}{1 - i\omega\tau_s + iqw_s\tau_s\cos\theta} \gamma V \frac{dn_0}{dE} \bar{g}(E) ,
$$
 (20)

$$
\bar{g}(E) = \int_0^{\pi/2} g(E, \vartheta) \sin \vartheta \, d\vartheta \quad . \tag{21}
$$

For $\omega \tau_s \gg 1$ Eq. (20) leads to $N_{s1} \ll (1/\omega \tau_s)n_1$, so that the last term on the right-hand side of Eq. (9) can be neglected. It means the deviation of the phonons from their equilibrium state does not matter, and

$$
g(E,\vartheta) = \frac{2\cos\vartheta}{1-i\omega\tau} \tag{22}
$$

For $\omega \tau_s \ll 1$ expression (20) can be expanded in $\omega \tau_s$, and its substitution in Eq. (9) gives

$$
-i\omega g = \frac{1}{\tau} \left[\overline{g} - g - i\omega \overline{\tau} \frac{T}{E} \coth\left(\frac{E}{2T} \right) \overline{g} \right] + \frac{2\cos\vartheta}{\tau} \ . \quad (23)
$$

Here

$$
\overline{\tau} = \frac{\overline{\rho}\hbar}{\pi PT} \sum_{s} \xi_{s} \frac{w_{s}^{2}}{\gamma_{s}^{2}}
$$

=
$$
\frac{\rho\hbar}{\pi PT} \left[\frac{\gamma_{I}^{2}}{w_{I}^{5}} + 2 \frac{\gamma_{I}^{2}}{w_{t}^{5}} \right]^{-1} \sum_{s} \frac{1}{w_{s}^{3}}
$$
(24)

is an average phonon relaxation time.

If $\omega\tau_0 \gg 1$ then relaxation terms on the right-hand side of Eq. (23), containing g and \bar{g} , are small, and

$$
g(E,\vartheta) = \frac{2i\cos\vartheta}{\omega\tau} + \frac{2}{(\omega\tau)^2} \left[\cos\vartheta - \frac{1}{4\sin^2\vartheta} \right].
$$
 (25)

If two of the inequalities $\omega \bar{\tau} \ll 1$ and $\omega \tau_0 \ll 1$ are satisfied, the main contribution in Eq. (23) is from the relaxation terms. In such a case, to solve Eq. (23) one should try to obtain an equation for \bar{g} from it at first. It has the form

$$
\left\{ -\frac{i\omega\tau_m}{2}\left[\ln\left(\frac{4}{\omega\tau_0}\right)+i\frac{\pi}{2}\right]-i\omega\overline{\tau}\frac{T}{E}\coth\left(\frac{E}{2T}\right)\right]\overline{\mathcal{g}}=1,
$$

where

$$
\tau_m = \tau_0 \left(\frac{T}{E}\right)^3 \tanh\left(\frac{E}{2T}\right) \tag{27}
$$

is the value of τ at $\vartheta = \pi/2$. Apparently there are two limiting cases: $\tau_0 \ln(1/\omega \tau_0) \ll \overline{\tau}$, when the value of \overline{g} is controlled by phonon relaxation, and the opposite case, when TLS relaxation plays this part. In both cases the deviation of the TLS population from its equilibrium form (2) is mainly due to ^a shift of the entire TLS—phonon systern from the thermodynamic equilibrium state rather than some violation of an equilibrium between TLS's and phonons. This fact is revealed when the last term on the right-hand side of Eq. (23) is neglected after $\bar{g}(E)$ has been calculated, so that

$$
g(E,\vartheta) = \frac{\overline{g}(E)}{1 - i\omega\tau} \tag{28}
$$

Now $g(E, \vartheta)$ is known for all limiting cases, and one should make use of Eq. (18).

The case of $\omega \bar{\tau} >> 1$ [see Eq. (22)] corresponds to Jäckle's theory:

$$
\alpha = \frac{\pi^4}{12} \frac{P(\gamma^2)}{\rho w^2} \frac{1}{w \tau_0},\tag{29}
$$

$$
\frac{\Delta w}{w} = -\frac{64\pi^6}{315} \frac{P(\gamma^2)}{\rho w^2} \frac{1}{(\omega \tau_0)^2}
$$
(30)

for $\omega\tau_0\gg1$, and

$$
\alpha = \frac{\pi}{2} \frac{P(\gamma^2)}{\rho w^2} q \tag{31}
$$

$$
\frac{\Delta w}{w} = -\frac{1}{2} \frac{P(\gamma^2)}{\rho w^2} \ln \frac{1}{\omega \tau_0}
$$
 (32)

for $\omega \tau_0 \ll 1$.

For the case of $\omega \bar{\tau} \ll 1$, $\omega \tau_0 \gg 1$ [see Eq. (25)] α has the form (29), and

$$
\frac{\Delta w}{w} = -\frac{4\pi^6}{315} \frac{P(\gamma^2)}{\rho w^2} \frac{1}{(\omega \tau_0)^2} \ . \tag{33}
$$

For the case of $\omega \overline{\tau} \ll 1$, $\omega \tau_0 \ll 1$ [see Eqs. (26) and (28)],

$$
\alpha = \frac{\pi^4}{4} \frac{P\langle \gamma^2 \rangle}{\rho w^2} \frac{1}{w\tau_0} \,, \tag{34}
$$

(26)

$$
\frac{\Delta w}{w} = -\frac{\pi^5 \ln 2}{8} \frac{P(\gamma^2)}{\rho w^2} \frac{1}{\omega \tau_0 [\ln(1/\omega \tau_0)]^2}
$$
(35)

if $\bar{\tau} \ll \tau_0 \ln(1/\omega \tau_0)$, and

$$
\alpha = \frac{1}{2} \frac{P(\gamma^2)}{\rho w^2} \frac{\ln(1/\omega \tau_0)}{w \overline{\tau}} , \qquad (36)
$$

$$
\frac{\Delta w}{w} = \frac{\pi}{8} \frac{P(\gamma^2)}{\rho w^2} \frac{1}{\omega \overline{\tau}}
$$
 (37)

if $\tau_0 \ln(1/\omega \tau_0) \ll \overline{\tau}$.

The temperature T_1 , when $\bar{\tau}=\tau_0$, can be expressed as

$$
T_1 = \pi \left[P \hbar^3 / \sum_s w_s^{-3} \right]^{1/2} .
$$
 (38)

Using the values $P \approx 0.4 \times 10^{32}$ erg⁻¹ cm⁻³, $w_l \approx 6 \times 10^5$ cm/sec. $w_s \approx 4 \times 10^5$ cm/sec for fused silica and borosilicate glass,^{2,16} one has $T_1 \approx 1$ K. The boundary temperature between cases (34) and (35) and (36) and (37) is close to T_1 and also depends on $\omega \tau_0$. If $\gamma_t \approx \gamma_l \approx 1.5$ eV and $\rho \approx 2$ g/cm³ (see Refs. 2 and 16–18) then $\tau_0 = \bar{\tau} = 10^{-8}$ sec at $T=T_1$.

111. DISCUSSION OF THE LOW-FREQUENCY LIMIT

Equations (34) and (36) show that ultrasound attenuation does not decrease with ω tending to zero. Naturally the question arises as to how to match this result to the phenomenological theory which predicts $\alpha \propto \omega^2$ if ω tends to zero.¹⁹ The answer is that the applicability conditions of the phenomenological theory are violated in the present theory as well as in Jäckle's theory.¹⁰ One can make use of the phenomenological theory only if the condition $\omega \tau \ll 1$ is satisfied for any relevant relaxation time τ .

First of all, there is a very broad distribution of TLS relaxation times τ in glasses. When $\omega \tau_0 \ll 1$ the main contribution in the integral with respect to parameter ϑ in Eq. (18) arises from the region of ϑ where $\omega \tau \sim 1$. It results from the assumption that a tunneling coupling energy Δ_0 , and so the parameter ϑ , can attain arbitrarily small values. If there is a maximum in the tunneling integral and so there are minimum values of Δ_0 and ϑ , say ϑ_m , then there is a maximum value of the TLS relaxation time τ : $\tau_{\text{max}} \sim \tau_0 / \vartheta_m^2$ (apparently only $E \sim T$ is essential). If an ultrasound frequency is so small that $\omega\tau_{\text{max}} \ll 1$, then Jäckle's theory leads to $\alpha \propto \omega^2$ in accordance with the phenomenological theory, but the present theory gives only $\alpha \propto \omega$. If the tunneling integral λ can really acquire arbitrarily large values, then there are arbitrarily large relaxation times τ_1 and indeed there is some physical reason for the inapplicability of the usual phenomenological theory in glasses.

The second reason for the discrepancy between the result of the present theory and that of the phenomenological theory is connected with the deviation of the phonon distribution function from its equilibrium form. The point is, the TLS—phonon relaxation is shown not to reduce the average nonequilibrium TLS population $\overline{n}_1(E)$. So $\bar{n}_1 \propto \omega^{-1}$ when ω tends to zero. In Jäckle's theory

phonons are considered to be in equilibrium, and $n_1 \propto \omega^0$ when ω tends to zero. That is, the discrepancy arises from neglecting any relaxation mechanism bringing the TLS—phonon system into the thermodynamic equilibrium state. The most rapid relaxation of this kind is very likely connected with the spectral diffusion. It can be described in the following way. Let some TLS with an energy splitting E be in the initial state with the upper-level occupation number $n=0$. Absorbing a phonon with the energy $\hbar\omega=E$, it passes into the state with $n=1$. Then let some adjacent TLS with an energy splitting E_1 also absorb a phonon with energy $\hbar\omega_1 = E_1$ and pass from the initial state with $n_1 = 0$ to the state with $n_1 = 1$. Due to the interaction of the TLS's by virtual phonons, the energy of the first TLS becomes $E' = E + \Delta E$ after this second transition. Here $\Delta E \sim \gamma^2/(\rho w^2 r^3)$, $r \sim (PT)^{-1/3}$ is the distance between the TLS's, and PT is the concentration of the TLS's whose energy splittings have the order of magnitude lower or equal to \hat{T} . ^{20,21} Now if the first TLS emits a phonon, its energy will be $\hbar \omega' = \hbar \omega + \Delta E$. After this, the second TLS can emit a phonon with the energy $\hbar\omega'_1 = \hbar\omega_1 - \Delta E$. The result is equivalent to some fourphonon interaction. As Black and Halperin¹⁴ have shown, the mean TLS energy shift due to spectra diffusion for a time interval $t \leq \tau_0$ has the order of magnitude $\Delta E(t/\tau_0)$. Hence, the corresponding energy relaxation time is

$$
\tau_E \sim \tau \frac{T}{\Delta E} \sim \tau \frac{\rho w^2}{P\gamma^2} \gg \tau \,. \tag{39}
$$

The described relaxation process may be neglected only if $\omega\tau_E \gg 1$. When ω becomes so small that $\omega\tau_E \ll 1$, n₁ t ends to some large but finite frequency-independent value, ²² and α becomes proportional to ω^2 in accordance with the phenomenological theory.

So, an observation of low-frequency ultrasound relaxation attenuation could give some information concerning properties of TLS's and their microscopic nature. Comparison of Eqs. (33) and (35) with the resonant attenuation coefficient

$$
\alpha_{\text{res}} = \tau_1 \frac{P(\gamma^2)}{\rho w^2} \frac{\omega}{w} \tanh\left[\frac{\hbar \omega}{2T}\right]
$$

shows that the relaxation attenuation dominates for $\omega < (T/\hbar\tau_1)^{1/2}$ where τ_1 is the maximum value of $\bar{\tau}$ and τ_0 . For $T \sim T_1 \sim 1$ K it means $\tau_1 \sim 10^{-8}$ sec and $\omega/2\pi$ < 600 MHz. For lower temperature the condition is stronger. So for $T \sim 0.4$ K it means $\omega/2\pi < 100$ MHz.

To conclude, it is worth noting that the threshold for nonlinear relaxation attenuation $\gamma V \sim T$ is rather high^{23,24} and does not depend on the existence of a phonon bottleneck. Another situation takes place for resonance attenuation observed in many experiments (see, e.g., Refs. 2, 15, and 25). In this case the deviation of the phonon distribution function from its equilibrium form can play a very important role, but discussion of the question goes beyond the limits of the present paper.

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