Multiphonon theory of kinetic processes in amorphous dielectrics

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(Submitted 18 November 1982)
Zh. Eksp. Teor. Fiz. **85**, 251–267 (July 1983)

The interaction between phonons and two-level systems in amorphous dielectrics is investigated with account taken of the multiphonon mechanism of the fluctuational preparation of the barrier. The analysis is carried out at an arbitrary ratio of the asymmetry parameter ε_a of the two-level system with the probability amplitude Δ of the coherent tunnel transition. It is shown that the previously obtained results for ultrasound damping and thermal conductivity in the $\varepsilon_a \gg \Delta$ approximation remain valid in the general case. The temperature dependence of the renormalization of the speed of sound in glasses is obtained. The causes of the large negative Grüneisen coefficient are explained.

PACS numbers: 63.20.Hp, 66.70. + f, 65.70. + y, 43.35.Cg

1. INTRODUCTION

According to the prevailing viewpoint, the principal role in kinetic processes that take place in dielectric glasses are played by the so-called two-level systems (TLS) first introduced by Phillips[1] and by Anderson *et al.*[2] for the description of anomalies in the low-temperature behavior of the heat capacity and thermal conductivity of amorphous dielectrics. It became possible subsequently to use this mechanism to explain a large group of phenomena observed in glasses not only at low (helium and lower) but also at relatively high temperatures. The main results obtained by now on this subject are described in detail in a collection of review articles edited by Phillips,[3] and also in a survey by Smolyakov and Khaimovich.[4] However, notwithstanding the considerable progress, the general status of the theory can still not be regarded as fully satisfactory. This pertains in particular to low temperatures. A number of difficulties encountered in the investigation of high-temperature thermal conductivity were pointed out, for example, by Anderson.[5] It is likewise difficult to explain within the framework of this scheme the negative value of the Grüneisen coefficient and its temperature dependence.[6]

In recent papers[7,8] we pointed out that the one-phonon approximation, which is customarily used in the theory of phonon interaction with TLS and of relaxation processes in TLS,[1–3,9,11] does not provide an adequate description. The point is that in tunneling of a heavy particle in a TLS a significant role is played by the mechanism of fluctuational preparation of the barrier,[12–14] a mechanism that leads at low temperatures to a strong (by several orders) renormalization of the rate constant of the tunneling transition, and at comparatively high temperatures (above those of liquid helium) it leads to the appearance of a strong nonmonotonic dependence.

In Refs. 7 and 8, two substantial simplifying assumptions were made, capable in principle of strongly restricting the applicability of the results. First, it was assumed that the amplitude Δ of the coherent tunnel transition is always small compared with the bare distance ε_a frequently called in the literature the TLS asymmetry parameter. Second, no account was taken of the diagonal party of the phonon interaction with the TLS, corresponding to the change of ε_a on passage of the phonon wave. As a result, the theory in the form expounded in Refs. 7 and 8 does not allow us to go in the limit to the universally accepted scheme[1–3,9–11] according to which the principal role is played by TLS with $\Delta \gg \varepsilon_a$. In addition, by disregarding the diagonal part of the interaction we discard by the same token the incoherent transitions,[12,15] which in the case $\Delta \gg \varepsilon_a$ can play the decisive role. As will be shown below, the latter is valid if one deals with single-phonon transitions.

In the present paper we forgo the two aforementioned assumptions and construct a theory in which account is taken of both the diagonal and nondiagonal process, and which holds at an arbitrary ratio of the parameters ε_a and Δ. The arguments concerning the role of the mechanism of the fluctuational preparation of the barrier in kinetic processes in glasses, which were set forth in Refs. 7 and 8, will play as before the principal role in the reasoning. In addition, we do not confine ourselves here to the imaginary part of the polarization operator for the phonons in an amorphous medium, the part responsible for the ultrasound damping, but analyze also its real part. This makes it possible to find the temperature dependence of the renormalization of the sound velocity in a wide range of temperatures, and also calculate the temperature dependence of the Grüneisen coefficient, which is an important experimentally observable characteristic of the elastic properties of the medium.

2. MULTIPHONON TRANSITIONS IN TWO LEVEL SYSTEMS

When speaking of two-level systems we shall bear in mind, as usual, that in an amorphous substance certain atoms or molecules can tunnel between states that are close in energy. Just as in Refs. 7 and 8, we shall assume that the characteristic frequency ν of the motion of the tunneling particle exceeds the characteristic phonon frequencies ω_p. We consider an individual TLS interacting with the vibrations of the medium. It can be described by the Hamiltonian

$$H=E_1(\{\xi_\mu\})a_1^+a_1+E_2(\{\xi_\mu\})a_2^+a_2 +\Delta(\{\xi_\mu\})(a_2^+a_1+a_1^+a_2)+H_{ph}(\{\xi_\mu\}). \tag{1}$$

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Here $H_{\mathrm{ph}}(\{\xi_\mu\})$ is the Hamiltonian of the phonon subsystem, and ξ_μ are the normal coordinates of the phonon oscillations; a_i^+ and a_i are quasifermion creation and annihilation operators. The matrix element of the nondiagonal tunnel transitions between two states 1 and 2 has the following structure:

$$\Delta(\{\xi_\mu\})=\hbar\nu\exp[-{}^1/_2J(\{\xi_\mu\})]. \qquad (2)$$

Since the tunneling particle must be located either in well 1 or in well 2, some quasifermion state, 1 or 2, is always excited, and consequently we have the relation

$$a_1^+a_1+a_2^+a_2=1.$$

We can use this relation to transform the Hamiltonian (1) into

$$H={}^1/_2[E_1(\{\xi_\mu\})+E_2(\{\xi_\mu\})]-{}^1/_2\varepsilon_a a_1^+a_1+{}^1/_2\varepsilon_a a_2^+a_2 +\sum_\mu g_\mu(c_\mu^++c_\mu)(a_2^+a_2-a_1^+a_1)+\Delta(\{\xi_\mu\})(a_2^+a_1+a_1^+a_2) +H_{ph}(\{\xi_\mu\}), \qquad (3)$$

where c_μ^+ and c_μ are the phonon creation and annihilation operators. We confine ourselves here right to terms linear in ξ_μ in the expansion of the function $E_2(\{\xi_\mu\})-E_1(\{\xi_\mu\})$. Then

$$g_\mu=iB_\varepsilon(\hbar/2MN\omega_\mu)^{1/2}(\mathbf{e}_\mu\mathbf{q}_\mu), \qquad (4)$$

where $\mathbf{e}_\mu$, $\mathbf{q}_\mu$, and ω_μ are repectively the polarization vector, the wave vector, and the frequency, all corresponding to the phonon mode μ, while B_ε is a constant that characterizes the coupling of the TLS with the phonons for an interaction that is diagonal in the number of the well. The first term in the Hamiltonian (3) can lead only to an insignificant renormalization of the phonon spectrum, which we shall disregard hereafter.

The analysis of the nondiagonal processes accompanied by tunneling of the particle from one well to another must be carried out with allowance for the multiphonon effects.[7,8] When calculating the amplitude of a coherent transition between the wells it is necessary to sum the series corresponding to the diagram of Fig. 1. The indices α and β indicate the number of the well in which the particle is located, and in the present case it is assumed that $\alpha\neq\beta$. The unshaded circle of Fig. 1 corresponds to the amplitude

$$\Delta_0=\hbar\nu\exp(-{}^1/_2J_0),$$

which is obtained from (2) at equilibrium values of the phonon coordinates, i.e., $\xi_\mu=0$. As a result of the summation of the diagrams of Fig. 1, the argument J_0 of the exponential becomes renormalized and we obtain the amplitude of the coherent tunnel transitions

$$\Delta_1(T)=\hbar\nu\exp[-{}^1/_2J^*(T)], \qquad (5)$$

where $J^*(T)=J_0-J_1(T)$. The temperature dependent function $J_1(T)$ reflects the circumstance that real tunneling takes place not when the phonon subsystem is at equlibrium, but only at those instants of time when the system fluctuations lower the potential barrier substantially. It is this mechanism which is known as the mechanism of fluctuational preparation of the barriers.[12] Approximate equations for the function $J_1(T)$ can be found in Ref. 8 (see also Eqs. (22) and (33)).

We shall be interested below in processes at an arbitrary ratio of the asymmetry parameter ε_a and the coherent-transition amplitude $\Delta_1(T)$. It is convenient to carry out the calculations in this case using the Keldysh diagram technique.[16]

The bare retarded Green function

$$G_{\alpha\beta}^{r(0)}=(\varepsilon-\varepsilon_\alpha+i\delta)^{-1}\delta_{\alpha\beta},\quad \varepsilon_\alpha=\mp\varepsilon_a/2 \qquad (6)$$

is diagonal in the well indices α and β. But on account of the coherent processes that are nondiagonal in this index (see Fig. 1) this function is renormalized into a nondiagonal matrix of Green functions $\tilde{G}_{\alpha\beta}{}^r$. Dyson's equations for the elements of this matrix are written in the form

$$\tilde{G}_{\alpha\alpha}^r=G_{\alpha\alpha}^{r(0)}+G_{\alpha\alpha}^{r(0)}\Delta_1(T)G_{\beta\beta}^{r(0)}\Delta_1(T)\tilde{G}_{\alpha\alpha}^r,$$

$$\tilde{G}_{\alpha\beta}^r=G_{\alpha\alpha}^{r(0)}\Delta_1(T)\tilde{G}_{\beta\beta}^r,\quad \alpha\neq\beta.$$

Solving these equations, we obtain

$$\tilde{G}_{\alpha\alpha}{}^r=[\varepsilon-\varepsilon_\alpha-\Delta_1^2(T)/(\varepsilon+\varepsilon_\alpha)+i\delta]^{-1}, \qquad (7)$$

$$\tilde{G}_{\alpha\beta}{}^r=\tilde{G}_{\beta\alpha}{}^r=\Delta_1(T)\{(\varepsilon+i\delta)^2-[\varepsilon_a^2/4+\Delta_1^2(T)]\}^{-1}. \qquad (8)$$

The matrtix (8) is diagonalized with the aid of the transformation

$$\hat{T}=\frac{1}{(2\Delta E)^{1/2}}\begin{vmatrix}(\Delta E+\varepsilon_a)^{1/2} & (\Delta E-\varepsilon_a)^{1/2}\\ (\Delta E-\varepsilon_a)^{1/2} & (\Delta E+\varepsilon_a)^{1/2}\end{vmatrix}. \qquad (9)$$

As a result we obtain a matrix of Green functions

$$G_{\alpha\beta}{}^r=(\varepsilon-\tilde{\varepsilon}_\alpha+i\delta)^{-1}\delta_{\alpha\beta}, \qquad (10)$$

that depend explicitly on the renormalized particle energy in the TLS

$$\tilde{\varepsilon}_\alpha=\mp\Delta E=\mp[\varepsilon_a^2+4\Delta_1^2(T)]^{1/2}. \qquad (11)$$

We note that a canonical transformation similar in form to the transformation (9) is used to diagonalize a Hamiltonian of the type (3) in the one-phonon approximation,[10] but $\Delta_1(T)$, must then be replaced by Δ_0. To obtain in the new representation the interaction amplitude it is necessary to apply the transformation (9) to the amplitude matrix

$$\begin{vmatrix}\sum_\mu g_\mu(c_\mu^++c_\mu) & \Delta(\{\xi_\mu\})\\ \Delta(\{\xi_\mu\}) & -\sum_\mu g_\mu(c_\mu^++c_\mu)\end{vmatrix}.$$

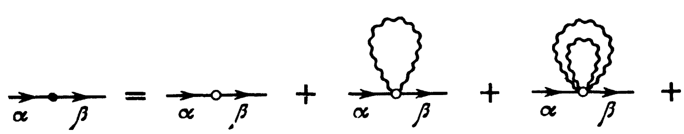


FIG. 1. Amplitude of coherent transition in a TLS. The wavy lines denote phonon Green functions, the straight lines the Green functions of quasifermion TLS excitations.

The result can be written in the following form: the diagonal amplitudes without change of the index α:

$$\hat{M}_d=\sum_\mu g_\mu(c_\mu^+ + c_\mu)\frac{\varepsilon_a}{\Delta E}+\frac{2\Delta_1(T)}{\Delta E}\Delta(\{\xi_\mu\}), \qquad (12)$$

and the nondiagonal with change of the index α:

$$\hat{M}_n=-\sum_\mu g_\mu(c_\mu^+ + c_\mu)\frac{2\Delta_1(T)}{\Delta E}+\frac{\varepsilon_a}{\Delta E}\Delta(\{\xi_\mu\}). \qquad (13)$$

If we confine ourselves in (12) and (13) to the one-phonon approximation, we obtain the expressions used in the traditional TLS theory (see, e.g., Eqs. (32) and (34) of Ref. 11). At the same time, the results of Refs 7 and 8 were obtained under the assumptions $g_\mu=0$ and $\Delta_1(T)\ll\varepsilon_a$ when the principal role is played by the second term in (13).

3. RELAXATION IN TWO LEVEL SYSTEMS AND PHONON DAMPING

In this section we consider the central questions of the theory: the relaxation of the excited state of a TLD and the damping of phonons as a result of their scattering by the TLS with account taken of the multiphonon character of these processes. As we shall show below, the results of the analysis in Ref. 8 remain in the main valid also in the general case considered here. We shall dwell therefore only on the changes due to disregard of the approximations $g_\mu=0$, $\Delta_1(T)\ll\varepsilon_a$.

We consider first the temperature dependence of the reciprocal TLS relaxation time. To this end we must find the expression corresponding to the diagrams shown in Fig. 2. When calculating the mass operator corresponding to a diagram with n phonon lines, we must substitute as the vertices the nth derivative of the amplitude (12) or (13) with respect to the phonon variables. The first terms, proportional to g_μ make no contribution in this case to the vertices of order higher than the first ($n\geqslant 2$) In addition, it can be easily seen that in each concrete diagram both vertices should be simultaneously either diagonal or, conversely, nondiagonal in the indices α and β. The physically observable quantities, as a rule, contain the value of the mass operator of Fig. 2, calculated on the mass shell. Therefore, by virtue of the energy conservation law

$$\hbar\omega_\mu=E_\alpha-E_\beta=\begin{cases}\pm\Delta E, & \alpha\neq\beta,\\ 0, & \alpha=\beta,\end{cases} \qquad (14)$$

it is necessary to confine oneself in the calculation of the corresponding diagram for the process in Fig. 2a to the contribution of the nondiagonal processes. No such restriction applies to the remaining diagrams of Fig. 2, for now the left-hand side of (14) contains a sum of several phonon particles.

To calculate the reciprocal relaxation time it is necessary to find the imaginary part of the mass operator Σ_α (Fig. 2). In the Keldysh diagram technique[16] the damping is directly connected with one of the elements of the mass-operator matrix elements:

$$\Sigma_\alpha^+=-if(\varepsilon)\hbar/\tau, \qquad (15)$$

where

$$f(\varepsilon)=(1+e^{\beta\varepsilon})^{-1},\quad \beta^{-1}=k_BT.$$

in accord with the form of the diagrams of Fig. 2, out of the entire set of the Green functions, in the Keldysh technique the expression for Σ_α^+ will contain only one quasifermion function

$$G_\alpha^+(\varepsilon)=-if(\varepsilon)\,\mathrm{Im}\,G_\alpha^r(\varepsilon) \qquad (16)$$

and one phonon function

$$D_\mu^+(\omega)=-{}^1/_2 iF(\omega)\,[\delta(\omega-\omega_\mu)-\delta(\omega+\omega_\mu)], \qquad (17)$$

where

$$F(\omega)=(e^{\beta\omega}-1)^{-1}.$$

As shown in Ref. 8, the need for taking into account the multiphonon processes arises only starting with a certain characteristic temperature T_1, which is usually of the order of the helium temperature. At lower temperature the one-phonon approximation is sufficient (diagam 2a). Then, if it is assumed that $\Delta_1(T)\gg\varepsilon_a$, the main contribution to the TLS relaxation is due to the first term in (13), which in this case does not depend on $\Delta_1(T)$ at all. Expressing in standard fashion Σ_α^+ in terms of the Green functions (16) and (17) and the vertex (13), we obtain for the reciprocal damping time an expression that coincides with the corresponding expressions in Refs. 1–3 and 9–11. In the opposite case $\Delta_1(T)\ll\varepsilon_a, g_\mu=0$ we obtain the result of Ref. 8.

The situation is more complicated at $T>T_1$, when the multiphonon processes Figs. 2b, c, etc., become important. Taking into account the adiabatic approximation in the parameter $\omega_D/\nu\ll 1$, the general expression for the sum of the contributions of the nondiagonal processes is of the form

$$1/\tau_n=K_{12}+K_{21}, \qquad (18)$$

where

$$K_{\alpha\beta}=\frac{2\pi}{\hbar}\mathrm{Av}_i\sum|\langle\{\nu_i\}|\hat{M}_n|\{\nu_f\}\rangle|^2\delta(E_{\alpha\beta}+E_i-E_f),$$

$$E_{\alpha\beta}=E_\alpha-E_\beta, \qquad (19)$$

i and f are the indices of the initial and final states of the phonon substem, the symbol Av_i stands for averaging over the initial states of the phonon subsystem. An expression such as (19) in the limit $g_\mu=0$ and $\Delta_1(T)\ll\varepsilon_a$ was discussed in Ref. 8. To obtain the contribution $1/\tau_d$ of the diagonal process it is necessary to replace $\hat{M}_n$ by $\hat{M}_d$ in (19) and put $E_{\alpha\beta}=0$.

The bare diagonal processes (4) contribute only to the one-phonon diagram 2a. At $T>T_1$, however, the damping is due mainly to the multiphonon diagrams that do not contain

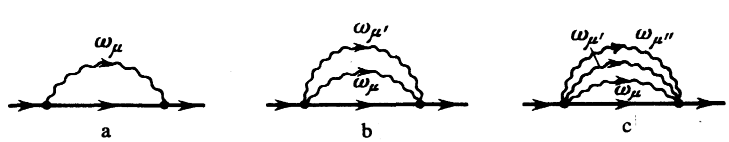


FIG. 2. Diagrams for the mass operator of quasifermion excitations of TLS. The thick straight lines denote the renormalized Green functions (10).

g. We can therefore put right away $g = 0$ in (12) and (13) and calculate $1/\tau_n$ and $1/\tau_d$ exactly as we calculated the reciprocal ultrasound damping length in the multiphonon limit $T > T_1$.[7,8] The expressions for $1/\tau_n$ and $1/\tau_d$ will then differ from the expression for the reciprocal ultrasound damping length $l_\lambda^{-1}(\Delta E)$ (prior to averaging over the parameter ΔE) only in the absence of the factor 2 $\sinh(\beta\hbar\omega_\lambda/2)$ and in the presence of factors due to the more complicated forms of matrix elements (12) and (13) of the TLS interaction with the phonons (ω_λ is the ultrasound frequency).

For the nondiagonal processes

$$K_{12}=K_{21}e^{-\beta\Delta E}=\frac{1}{\tau^{(0)}}\left(\frac{\varepsilon_a}{\Delta E}\right)^2 e^{\beta\Delta E/2},$$

and for the diagonal processes

$$K_{11}=K_{22}=\frac{4\Delta_1^2(T)}{\Delta E^2}\frac{1}{\tau^{(0)}}.$$

As a result we have

$$\frac{1}{\tau}=\frac{1}{\tau_n}+\frac{1}{\tau_d}=\frac{1}{\tau^{(0)}}\left[\mathrm{ch}\frac{\beta\Delta E}{2}-8\frac{\Delta_1^2(T)}{\Delta E^2}\mathrm{sh}^2\frac{\beta\Delta E}{4}\right]. \quad (20)$$

The expression for $1/\tau^{(0)}$ can be found here with the aid of (18) and (19) in which $\hat{M}_n$ is replaced by $\Delta(\{\xi_\mu\})$ and $\Delta E = 0$.

In Ref. 8 we carried out the calculations in two approximations. In one case we retained in $\Delta(\{\xi_\mu\})$ only the terms linear in ξ_0 in the expansion $J(\{\xi_\mu\})$; then (cf. Eq. (25) in Ref. 8)

$$1/\tau^{(0)}=\hbar v^2[2\pi/F(T)]^{1/2}\exp[-J_0+J_2(T)], \quad (21)$$

where

$$F(T)=\frac{1}{8}\sum_\mu |A_\mu|^2(\hbar\omega_\mu)^2\,\mathrm{sh}^{-1}\frac{\beta\hbar\omega_\mu}{2},$$

$$J_2(T)=\frac{1}{8}\sum_\mu |A_\mu|^2\,\mathrm{cth}\frac{\beta\hbar\omega_\mu}{4}, \qquad A_\mu=\frac{\partial J(\{\xi_\mu\})}{\partial\xi_\mu}.$$

In the second case one can retain also the derivatives of the function $J(\{\xi_\mu\})$, but it is necessary to confine oneself to the Einstein model for the phonon spectrum with characteristic frequency Ω; the (cf. Eq. (7) in Ref. 8)

$$\frac{1}{\tau^{(0)}}=\frac{v^2}{\Omega\varphi_3^{1/2}}\exp[-J_0+J_1(T)]I_0(\varphi_1\varphi_2), \quad (22)$$

where

$$\varphi_1={}^1/_4R[\mathrm{th}(\beta\hbar\Omega/4)+{}^1/_2R_1]^{-1}, \quad J_1(T)=\varphi_1(1-\varphi_2),$$

$$\varphi_2=\mathrm{sh}\frac{\beta\hbar\Omega}{4}\left[2\,\mathrm{ch}^3\frac{\beta\hbar\Omega}{4}\left(\mathrm{th}\frac{\beta\hbar\Omega}{4}+\frac{1}{2}R_1\right)\right]^{-1},$$

$$\varphi_3=\left(1+\frac{1}{2}R_1\,\mathrm{cth}\frac{\beta\hbar\Omega}{4}\right)\left(1+\frac{1}{2}R_1\,\mathrm{th}\frac{\beta\hbar\Omega}{4}\right),$$

$$J'=\frac{\partial J}{\partial\Delta R}, \quad J''=\frac{\partial^2 J}{\partial\Delta R^2}, \quad R=\frac{\hbar J'^2}{M\Omega}, \quad R_1=\frac{\hbar J''}{M\Omega},$$

M is the characteristic mass that determines the phonon spectrum of the glass, ΔR is the width of the potential barrier in the TLS, $I_0(x)$ is a Bessel function of imaginary argument. We shall return to a more detailed discussion of the calculations that lead to (21) and (22) later in the analysis of the renormalization of the speed of sound on account of its scattering by the TLS.

We shall not dwell here in detail on a discussion of the features of the $1/\tau(T)$ dependence that follows from Eqs. (20) and (21), since an analysis of similar equations for the reciprocal length was carried out in Ref. 8. It was shown there that functions such as (20) and (21) go through a minimum at a relatively low temperatures ($T_2 \approx 10$ K), and then increase exponentially like $1/\tau(T) \propto \exp(T/T_0)$. It must be noted that the depth of this minimum decreases with decreasing parameter R. Therefore at relatively low values of R the maximum at $T_1 < T < T_2$ and the minimum at $T \approx T_2$ may not exist at all. This is probably the situation in B_2O_2 and $Zn(PO_3)_2$ glasses, in which only a slightly observable step is seen at $T \approx 5$ K in the temperature dependence of the reciprocal untrasound-damping length.[17]

The high-temperature behavior of $1/\tau(T)$ is poorly described by Eq. (20), and it is necessary to use (21), according to which $1/\tau(T)$ goes through a maximum at a temperature $T_3 \gtrsim 50$ K whose value depends strongly on the characteristics of the system. As already noted, the exponential growth $1/\tau(T) \propto \exp(T/T_0)$, apparently manifest itself in the temperature dependence of the luminescence intensity in the glass g-As_2S_3.[18]

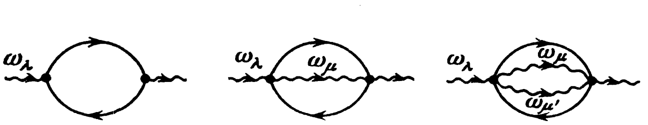
We proceed now to a discussion of the temperature dependence of the reciprocal ultrasound damping length l_λ^{-1}. In this case the calculation is perfectly analogous to the calculation of the temperature dependence of the relaxation time. We must calculate the imaginary part of the polarization operator represented by the diagram of Fig. 3. Usually one singles out in the ultrasound damping two mechanisms: resonant and relaxational. Among the diagrams of Fig. 3, one diagram, 3a, corresponds to these processes. To verify this it suffices to find the polarization operator

$$\Pi_\lambda^+(\omega)=-iF(\omega)\hbar/\tau_\lambda(\omega). \quad (23)$$

Knowing the lifetime $\tau_\lambda(\omega)$ of a phonon with frequency ω_λ, we obtain the reciprocal ultrasound damping length with the aid of the formula

$$l_\lambda^{-1}=(\tau_\lambda(\omega)s)^{-1},$$

where s is the speed of sound. Then, if it is assumed that the vertices in the diagram 3a are the nondiagonal (13), we obtain for l_λ^{-1} an expression corresponding to the resonant mechanism. If, however, these vertices are diagonal, we obtain an expression corresponding to the relaxational mechanism. In the limit $\varepsilon_a \ll \Delta_1(T)$ we obtain Eqs. (65) and (66) of Ref. 11. We note that in this case it is necessary to take into account the finite lifetime of the particle at the given TLS level, substituting in (6) and (16) the corresponding imaginary part.



a b c

FIG. 3. Polarization operator of phonons in glass.

It is known[11] that at $\varepsilon_a \ll \Delta_1(T)$ the relaxation mechanism makes in the low temperature limit a contribution proportional to T_3 to the ultrasound damping. A similar dependence at $\varepsilon_a \gg \Delta_1(T)$ results also from the two-phonon processes of Fig. 3b (Refs. 7 and 8). The difference between these results can be experimentally deduced from the frequency dependence of l_λ^{-1}.

In the multiphonon regime at $T > T_1$ the main contribution to the ultrasound damping is connected with those diagrams of Fig. 3 which have a large number of phonon lines. We can therefore neglect the bare diagonal processes and put $g_\mu = 0$. The contribution of the nondiagonal processes (13) to the ultrasound damping for TLS with given ΔE is then described by the formula

$$l_\lambda^{-1}(\Delta E) = \frac{2\pi}{\hbar s} \mathrm{Av} \sum_f \left| \left\langle \{\nu_i\} \middle| \xi_\mu \frac{\partial \hat{M}_n}{\partial \xi_\mu} \middle| \{\nu_f\} \right\rangle \right|^2 \times \delta(\pm\Delta E + E_i - E_f \pm \hbar\omega_\lambda)\, n(0)\, \Delta E_0, \tag{24}$$

where $n(0)$ is the TLS density and ΔE_0 is the upper limit of the ΔE distribution. The symbol Av denotes thermodynamic averaging over the initial state of the TLS and of the phonon subsystem. The plus sign in front of $\hbar\omega_\lambda$ is placed then if the ultrasound phonon $\hbar\omega_\lambda$ is absorbed, and the minus sign if it is emitted. The plus sign in front of ΔE corresponds to an increase of the TLS energy as a result of the interaction with the phonon, and the minus sign corresponds to a lowering of the TLS energy(see Fig. 2 in Ref. 7). The contribution of the diagonal processes (12) is obtained from (24) by substituting the operator $\hat{M}_d$ for $\hat{M}_n$ and $\Delta E = 0$.

Equation (24) was analyzed in detail in Refs. 7 and 8 for the case $\Delta_1(T) \ll \varepsilon_a$. Now, however, we can find l_λ^{-1} in the general case with the aid of (12), (13), and (14). The ultrasound damping due to the scatteirng by TLS with a definte value of ΔE is described by the expression (cf. the derivation of Eq. (20))

$$l_\lambda^{-1}(\Delta E) = \left[1 + 8\left(\frac{\Delta_1(T)}{\Delta E}\right)^2 \mathrm{sh}^2 \frac{\beta\Delta E}{4}\right] \mathrm{ch}^{-1} \frac{\beta\Delta E}{2} (l_\lambda^{(0)})^{-1}. \tag{25}$$

Here $(l_\lambda^{(0)})^{-1}$ is the contribution made to the ultrasound damping by the scattering from the TLS with $\Delta E = 0$ and obtained in the multiphonon regime in the approximation $\Delta_1(T) \ll \varepsilon_a$ and $g_\mu = 0$:

$$(l_\lambda^{(0)})^{-1} = \frac{2\gamma\omega_\lambda}{\hbar^2 \tau^{(0)} s} \mathrm{sh} \frac{\beta\hbar\omega_\lambda}{2}, \tag{26}$$

where $\gamma = \pi^2\hbar^2 R / 20\omega_D$ (see Ref. 8). We obtain the final result by averaging (25) over ΔE, obtaining

$$l_\lambda^{-1} = 2g(T) \frac{n(0)\gamma\omega_\lambda}{\hbar^2 s \tau^{(0)}} \mathrm{sh} \frac{\beta\hbar\omega_\lambda}{2}, \tag{27}$$

where

$$g(T) = \int_{2\Delta_1(T)}^{\Delta E_0(T)} d\Delta E \left[1 + 8\left(\frac{\Delta_1(T)}{\Delta E}\right)^2 \mathrm{sh}^2 \frac{\beta\Delta E}{4}\right] \mathrm{ch}^{-1} \frac{\beta\Delta E}{2}.$$

$$\Delta E_0(T) = [\varepsilon_{a\,max}^2 + 4\Delta_1^2(T)]^{1/2}, \tag{28}$$

Here $\varepsilon_{a\,\max}$ is the upper limit of the distribution of the asymmetry parameter. The question of the distribution of the parameter Δ_0, and hence $\Delta_1(T)$, was already discussed by us in Ref. 8, where we adhered to the viewpoint that Δ_0 is bounded from below. This assumption is partially confirmed by the experimental results of the measurements of the time dependence of the heat capacity of a number of glasses at low temperatures.[19] It follows from the results of the latter paper that the distribution of Δ_0 is at any rate not uniform, although it can apparently not be regarded as δ-like.

The integral (28) can be evaluated analytically in three limiting cases. First, at $\beta\Delta_1(T) < 1$ and $\beta\Delta E_0(T) > 1$ the integration limits in (28) can be replaced by 0 and ∞. We then obtain

$$g(T) = \pi k_B T + \Delta_1^2(T)/2k_B T, \tag{29}$$

which is valid apparently in the region of helium temperatures. Second, at $\beta\Delta E_0(T) < 1$ and $\varepsilon_{a\,\max} > 2\Delta_1(T)$ we have

$$g(T) \approx \Delta E_0(T) \approx \varepsilon_{a\,max}. \tag{30}$$

Finally, the third limiting case is realized also at high temperatures, when $\varepsilon_{a\,\max}/2\Delta_1(T) \ll 1$. Expanding (28) in powers of this small parameter we obtain the formula

$$g(T) = \frac{\varepsilon_{a\,max}^2}{4\Delta_1(T)} \left(1 + 8\,\mathrm{sh}^2 \frac{\beta\Delta_1(T)}{4}\right) \mathrm{ch}^{-1}(\beta\Delta_1(T)), \tag{31}$$

which gives $g(T) = \varepsilon_{a\,\max}^2/\Delta_1(T)$, at $k_B T < \Delta_1(T)$ and $g(T) = \varepsilon_{a\,\max}^2/4\Delta_1(T)$ at $k_B T > \Delta_1(T)$. The question of just which situation, (30) or (31), is realized in the high-temperature experiments cannot be answered in general. In both cases the reciprocal ultrasound-damping length (as well as the sound-velocity renormalization, See the next section reaches a maximum when the function $J_1(T)$ saturates [Eq. (22)]. After that, l_λ^{-1} begins to decrease because of the factor sinh $(\beta\hbar\omega_\lambda/2)$ connected with the statistics of the level occupation.

4. REAL PART OF THE POLARIZATION OPERATOR

The sound–velocity corrections δs due to the sound scattering by the TLS are connected with the damping by a Kronig-Kramers relation. It would therefore be possible to express δs with the aid of the corresponding integral of the functions (27) with respect to frequency. Such a method was used in Ref. 20 at $T < T_1$. At $T > T_1$, however, the frequency dependence of the reciprocal ultrasound-damping length l_λ^{-1} contains a factor sinh$(\beta\hbar\omega_\lambda/2)$, and as a result this integral diverges at $\beta\hbar\omega_\lambda > 1$. The region of such high frequencies has not yet been sufficiently well investigated (preliminary results for the resonant and relaxational processes were published recently,[21] and we shall therefore calculate the real part of the polarization operator directly from the diagrams of Fig. 3.

As shown in the preceding section, it is sufficient for us to know the polarization operator in the limit $g_\mu = 0$, $\Delta_1(T) \ll \varepsilon_a$. The transition to the general case entails no difficulty. In this approximation the retarded polarization operator corresponding to the sum of the diagram of Fig. 3 can be written in the form

$$\Pi_\lambda^{r}(\omega)|_{\omega=\omega_\lambda}=\mathrm{Av}\sum_f\left|\left\langle\{v_i\}\left|\xi_\lambda\frac{\partial\Delta(\{\xi_\mu\})}{\partial\xi_\lambda}\right|\{v_f\}\right\rangle\right|^2$$
$$\times[\pm\Delta E+E_i-E_f\pm(\hbar\omega_\lambda+i\delta)]^{-1}. \qquad (32)$$

It is easy to verify that Im $\Pi_\lambda{}^r = \hbar l_\lambda/2s$, and that Re $\Pi_\lambda{}^r$ is connected with Im $\Pi_\lambda{}^r$ by the Kramers-Kronig relations and determines the renormalization of the phonon spectrum on account of their interaction with the TLS.

To calculate the explicit form of (32) we shall use the same procedure as for the calculation of Im $\Pi_\lambda{}^r$ in Ref. 8. We use here the integral representation

$$[\pm\Delta E+E_i-E_f\pm(\hbar\omega_\lambda+i\delta)]^{-1}$$
$$=\pm i\int_{-\infty}^{0}dv\exp\{-iv[\hbar\omega_\lambda\mp(E_i-E_f)\pm\Delta E+i\delta]\}. \qquad (33)$$

The total expression for $\Pi_\lambda{}^r$ contains four terms that differ in the signs of ΔE and $\hbar\omega_\lambda$ (see the discussion following Eq. (24)). Any of these terms can be calculated in accord with the scheme of Ref. 8. Taking for the sake of argument both signs positive, we obtain

$$\Pi^{(1)'}=i\gamma\omega_\lambda v^2 e^{-J_0}\int_{-\infty}^{0}dv\{\det\Phi\}^{-1/2}$$
$$\times\exp\left\{i(\Delta E+\hbar\omega_\lambda)v+\frac{1}{4}\sum_{\mu\mu'}A_\mu B_{\mu\mu'}^{-1}A_{\mu'}\right\}. \qquad (34)$$

Here

$$\tilde B_{\mu\mu'}=\partial^2 J(\{\xi_\mu\})/\partial\xi_\mu\partial\xi_{\mu'},$$
$$B_{\mu\mu'}={}^1/_2\tilde B_{\mu\mu'}+2\delta_{\mu\mu'}\,\mathrm{sh}(\beta\hbar\omega_\mu/2)$$
$$\times\{\mathrm{ch}(\beta\hbar\omega_\mu/2)+\cos[\hbar\omega_\mu(v+i\beta/2)]\}^{-1},$$
$$\Phi_{\mu\mu'}=\{\delta_{\mu\mu'}+{}^1/_4\tilde B_{\mu\mu'}\{\mathrm{ch}(\beta\hbar\omega_\mu/2)$$
$$+\cos[\hbar\omega_\mu(v+i\beta/2)]\}\mathrm{sh}^{-1}(\beta\hbar\omega_\mu/2)\}$$
$$\times\{\delta_{\mu\mu'}+{}^1/_4\tilde B_{\mu\mu'}\{\mathrm{ch}(\beta\hbar\omega_\mu/2)$$
$$-\cos[\hbar\omega_\mu(v+i\beta/2)]\}\mathrm{sh}^{-1}(\beta\hbar\omega_\mu/2)\}.$$

It is easy to verify that the imaginary part of (34) reduces to the corresponding expression in Ref. 8, for in this case the integration can be carried out from $-\infty$ to $+\infty$ and one can make the change of variable $v+i\beta/2\to v$.

We shall analyze (34), as is customary, in two aproximations. We assume first that $\tilde B_{\mu\mu'}=0$. Then

$$\Pi^{(1)'}=i\gamma\omega_\lambda v^2\exp\{-J_0+J_1(T)\}\int_{-\infty}^{0}dv\exp\left\{i(\Delta E+\hbar\omega_\lambda)v\right.$$
$$\left.+\frac{1}{8}\sum_\mu|A_\mu|^2\cos\left[\hbar\omega_\mu\left(v+\frac{i\beta}{2}\right)\right]\mathrm{sh}^{-1}\frac{\beta\hbar\omega_\mu}{2}\right\},$$
$$J_1(T)=\frac{1}{8}\sum_\mu|A_\mu|^2\,\mathrm{cth}\frac{\beta\hbar\omega_\mu}{2}. \qquad (35)$$

The exponential factor in front of the integral determines the renormalization of the tunneling probability on account of the fluctuational preparation of the barrier (see Fig. 1). At low temperatures $T<T_1$, despite the large value of the parameter $|A_\mu|^2\sim R$, the integrand can be expanded in terms of R. The zeroth order term corresponds to resonant interaction (Fig. 3a). In this case the integral with respect to v can be calculated. Summing next the four contributions of type (33) with the corresponding statistical weights, we obtain for the sound-velocity renormalization the expression

$$\frac{\delta s}{s}=\frac{n(0)}{\hbar}\gamma v^2\exp[-J_0+J_1(T)]\Psi(T), \qquad (36)$$

where

$$\Psi(T)=\int_0^{\Delta E_0}\frac{\Delta E\,d\Delta E}{\Delta E^2-(\hbar\omega_\lambda)^2}\,\mathrm{th}\frac{\beta\Delta E}{2}.$$

At $\hbar\omega_\lambda\ll\Delta E_0$ and $\beta\hbar\omega_\lambda\ll1$ for $\Psi(T)$ the expression

$$\Psi(T)\approx\ln(\beta\Delta E_0).$$

At $T<T_1$ the sound-velocity renormalization has thus a logarithmic dependence on temperature. The transition to the case $\Delta_1(T)\gg\varepsilon_a$ can be effected with the aid of expression (13) for the amplitude of the nondiagonal interaction. As a result we arrive at the expressions previously obtained within the framework of the one-phonon theory[20] (see also Ref. 17).

5. RENORMALIZATION OF THE SOUND VELOCITY IN THE MULTIPHONON REGIME

Since the parameter $R\approx200$ is large,[8] the one-phonon approximation becomes inapplicable for the calculation of the integral with respect to v in (35) even at relatively low temperatures ($T_1<10$ K). In Ref. 8 we calculated the imaginary part of this integral by the saddle-point method. The saddle point is obtained from the equation

$$i(\Delta E+\hbar\omega_\lambda)=\frac{1}{8}\sum_\mu|A_\mu|^2\hbar\omega_\mu\sin\left[\hbar\omega_\mu\left(v+\frac{i\beta}{2}\right)\right]\mathrm{sh}^{-1}\frac{\beta\hbar\omega_\mu}{2}. \qquad (37)$$

This method can be used upon satisfaction of the inequality

$$\beta F(T)\gg\Delta E+\hbar\omega_\lambda,$$

where

$$F(T)=\frac{1}{16}\sum_\mu|A_\mu|^2(\hbar\omega_\mu)^2\,\mathrm{sh}^{-1}\frac{\beta\hbar\omega_\mu}{2}=2\pi^8R\left(\frac{T}{\Theta}\right)^4(k_BT)^2, \qquad (38)$$

and Θ is the Debye temperature. The solution of (37) is then

$$v_0=-i\beta/2. \qquad (39)$$

The direction of the path of steepest descent passes through the point v_0 parallel to the real axis. It is easy to verify that in the saddle-point approximation the integral (33) is pure imaginary. To go outside the framework of this approximation and find the real part of (33) we proceed as follows.

We replace in (33) the integration along the real negative axis $(-\infty,0)$ by integration along the contour shown in Fig. 4 by the thick line. The imaginary part of the integral over the section $(-\infty+v_0,v_0)$, in the lowest order in the parameter $R^{-1/2}$, corresponds to the saddle point approximation. The contribution to the real part appears in the next

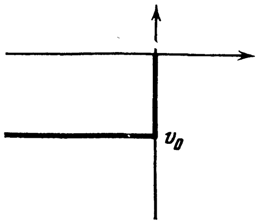


FIG. 4. Integration contour for the calculation of the integrals (35) and (45); $v_0 = -i\beta/2$ is the saddle point.

order in terms of the parameter, and is proportional to R^{-1}. Making the change of variable $z = v + i\beta/2$, we obtain

$$\operatorname{Re}\Pi^{(1)} = -\gamma\omega_\lambda v^2 e^{-J_0}\int_{-\infty}^{0} dz\,\sin[(\Delta E+\hbar\omega_\lambda)z]\exp\left\{\frac{1}{2}\beta(\Delta E+\hbar\omega_\lambda) + \frac{1}{8}\sum_\mu |A_\mu|^2\cos(\hbar\omega_\mu z)\,\mathrm{sh}^{-1}(\beta\hbar\omega_\mu)\right\}. \quad (40)$$

If $T > T_1$, the integral (40) converges at values of z satisfying the conditions

$$|\hbar\omega_\mu z| \ll 1, \quad |(\Delta E+\hbar\omega_\lambda)z| \ll 1.$$

The trigonometric functions can therefore be expanded accurate to terms quadratic in z. The integral (40) is now readily evaluated.

$$\operatorname{Re}\Pi^{(1)} = -\frac{\gamma\omega_\lambda v^2}{2F(T)}(\Delta E+\hbar\omega_\lambda)\exp\left[-J_0+J_2(T)+\frac{1}{2}\beta(\Delta E+\hbar\omega_\lambda)\right], \quad (41)$$

where

$$J_2(T) = \frac{1}{8}\sum_\mu |A_\mu|^2\,\mathrm{cth}\frac{\beta\hbar\omega_\mu}{4}.$$

To find the complete expression for the real part of the polarization operator it is necessary to sum four terms of the type (41), which differ from one another in the signs of ΔE and $\hbar\omega_\lambda$, with appropriate statistical weights. As a result we arrive at the following expression for the renormalization of the sound velocity, connected with the TLS with the given value of ΔE:

$$\frac{\delta s}{s} = \frac{\operatorname{Re}\Pi^r n(0)\Delta E_0}{2\hbar\omega_\lambda} = -\frac{\gamma\omega_\lambda v^2}{F(T)}\exp[-J_0+J_2(T)]\frac{\mathrm{sh}(\beta\hbar\omega_\lambda/2)}{\mathrm{ch}(\beta\Delta E/2)} \times n(0)\Delta E_0. \quad (42)$$

The quantity $\delta s/s$ has an order of smallness R^{-1}. Corrections of the same order arise in $\operatorname{Re}\Pi^{(1)}$ because of the integral on the section $(v_0,0)$ of the contour (see Fig. 4). However, averaging over the initial states of the TLS, which makes it possible to go over from (41) to (42), cancels these corrections and their only contribution to the sound-velocity renormalization is small of order R^{-2}.

If we remain in the same approximation as in Refs. 7 and 8 ($g_\mu = 0$, $\Delta_1(T) \ll \varepsilon_a$), to obtain the final answer it suffices to average (42) over ΔE. The factor $\Delta E_0 \cosh^{-1}(\beta\Delta E/2)$ in (42) is then replaced by $\pi k_B T$. In the general case it is necessary to proceed as in the derivation of (27). As a result we have

$$\delta s/s = g(T)\,\delta s(0)/s. \quad (43)$$

The approximation used in the derivative of (43) restricts its validity to temperatures lower than the characteristic temperature T_3 at which the reciprocal ultrasound-damping length goes through the second maximum. We proceed therefore to derive for the sound-velocity renormalization an expression that would be valid also in this temperature region.

At high temperatures ($T \sim T_3$) the oscillations of the barrier width increase so much that Eq. (42) derived under the assumption $\tilde{B}_{\mu\mu'} = 0$, no longer holds. We must therefore return to Eq. (34) and analyze it now without this assumption. We must, however make instead other simplifications. First, we assume that the phonon spectrum of the system is described by the Einstein model. At sufficiently high temperatures, when the restrictions connected with the occupation of the phonon states are relaxed, the main contribution to the fluctuational preparation of the barrier is made by the high-frequency part of the spectrum, which is quite satisfactorily described by this model. The Einstein model is best suited for the case of optical or local oscillation modes.

Second, we assume that the function $J(\{\xi_\mu\})$ really depends only on the two coordinates that determine the positions of the minima of the TLS potential curve. We assume also that these two coordinates are simultaneously the normal coordinates of the phonons. The index μ in (34) runs now through only two values, 1 and 2, and

$$A_\mu = R^{1/2}\begin{vmatrix} 1 \\ -1 \end{vmatrix}, \quad \tilde{B}_{\mu\mu'} = R_1\begin{vmatrix} 1 & -1 \\ -1 & 1 \end{vmatrix}. \quad (44)$$

Substituting (44) in (34) we obtain

$$\Pi^{(1)} = i\gamma\omega_\lambda v^2 e^{-J_0}\int_{-\infty}^{0} dv\,\{\det\Phi\}^{-1/2} \times\exp\left\{i(\Delta E+\hbar\omega_\lambda)v + \frac{R}{2}\left[R_1+\frac{2\,\mathrm{sh}(\beta\hbar\Omega/2)}{\mathrm{ch}(\beta\hbar\Omega/2)+\cos[\hbar\Omega(v+i\beta/2)]}\right]^{-1}\right\}. \quad (45)$$

The saddle point equation for the integral (45)

$$\sin[\hbar\Omega(v+i\beta/2)] = 0 \quad (46)$$

allows us to conclude that the difference $\cos[\hbar\Omega(v+i\beta/2)] - 1$ can be regarded as small. We can therefore transform (45) into

$$\Pi^{(1)} = \frac{i\gamma\omega_\lambda v^2 e^{-J_0}}{\varphi_3^{1/2}}\int_{-\infty}^{0} dv\,\exp\left\{i(\Delta E+\hbar\omega_\lambda)v+\varphi_1(1-\varphi_2) + \varphi_1\varphi_2\cos\left[\hbar\Omega\left(v+\frac{i\beta}{2}\right)\right]\right\}. \quad (47)$$

The imaginary part of the integral (47) yields, after averaging over the initial states of the TLS and ΔE, an expression for the reciprocal ultrasound damping length[7,8] (cf. also Eq. (22) for the reciprocal TLS relaxation time). To find the real part of $\Pi^{(1)}$ we transform, just as in the analysis of (33), to integration along the contour shown in Fig. 4, and then confine ourselves to calculation of the integral along the semiaxis $(-\infty+v_0, v_0)$. From among all the roots of Eq. (46) we need retain only one, $v_0=-i\beta/2$, and carry out the integration near this point. The remaining roots are unphysical and drop out as soon as we recognize that the phonon spectrum has in fact a nonzero width. As a result we arrive at the expression

$$\Pi^{(1)}=-\frac{\gamma\omega_\lambda v^2}{2\varphi_1\varphi_2\varphi_3^{1/2}(\hbar\Omega)^2}(\Delta E+\hbar\omega_\lambda)\exp\left[-J_0+\varphi_1+\frac{1}{2}\beta(\Delta E+\hbar\omega_\lambda)\right]. \tag{48}$$

We now average (48) over the initial TLS states and sum the contributions of the diagonal (12) and nondiagonal (13) processes, and the average over ΔE. As a result we have

$$\frac{\delta s}{s}=-g(T)\frac{\gamma\omega_\lambda v^2 n(0)}{2(\hbar\Omega)^2\varphi_1\varphi_2\varphi_3^{1/2}}\exp[-J_0+\varphi_1(T)]\,\mathrm{sh}\frac{\beta\hbar\omega_\lambda}{2}. \tag{49}$$

The further analysis of (40), (41), and (47) differs only in insignificant details from the analysis of the corresponding equations for the ultrasound-damping length in Refs. 7 and 8. The general picture of the temperature dependence of the sound-velocity renormalization due to its scattering by the TLS is the following (see Fig. 5). At low temperatures $(T<T_1)$ the absolute value $|\delta s|$ of the renormalization decreases with increasing temperature logarithmically [Eq. (34)] (see also Refs. 20 and 17). In this region, the main contribution is made by resonant scattering from TLS with $\Delta E>k_B T$, in which the populations of the two levels still differ markedly. At $T>T_1$ the multiphonon processes come into play and s is described by Eqs. (42), (43), and (49). As a result of the detuning from resonance (cf. Refs. 7 and 8) the absolute value $|\delta s|$ continues to decrease, but now in accordance with a faster temperature dependence (T^{-6} in the approximation (42) and $\exp(\beta\hbar\omega_\lambda/4)$ in the approximation (49). The $|\delta s|$ goes through a minimum at $T\approx T_2$ and begins to increase exponentially because of the increase in the amplitude of the oscillations of the width of the potential barrier. At still higher temperatures, $T\approx T_3|\delta s|$ reaches a maximum. In this region become probable barrier fluctuations such that the barrier is lowered so much that the particle passes in fact above it. Further decrease with increasing temperature is ensured by the factor $\sinh(\beta\hbar\omega_\lambda/2)$ (see the discussion at the end of Sec. 3). This picture agrees on the whole with the experimental data cited in Ref. 17, except for the region of fast decrease $|\delta s|$ with temperature at $T_1<T<T_2$. This section is difficult to observe in experiment, possibly because it is very small.

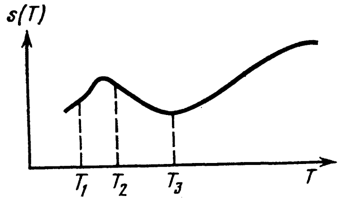


FIG. 5. Temperature dependence of the speed of sound.

6. THE GRÜNEISEN COEFFICIENT

To conclude this section we shall discuss the anomalous properties of the Grüneisen coefficnet γ_G, which are due to the contribution of the TLS. It is known that this coefficient characterizes the thermal expansion of matter and is connected with the derivative of the speed of sound with respect to the volume by the relation

$$\gamma_G={}^1/_3-\partial\ln s/\partial\ln V. \tag{50}$$

When the substance is compressed, the barrier width in the TLS increases, and this alters greatly the renormalization of the speed of sound. The interaction between the sound wave and the TLS becomes stronger and the speed of sound decreases, so that this mechanism makes a negative contribution to the Grüneisen coefficient.

We substitute (49) in (50). We take it into account here that $R\gg R_1$ and neglect the third derivatives $\partial^3 J/\partial\Delta R^3$. Then

$$\gamma_G=\frac{1}{3}\left[1-a\frac{\partial(\delta s/s)}{\partial\Delta R}\right]=\frac{1}{3}\left\{1-\pi\left(\frac{RMs^2}{\hbar\omega_D}\right)^{1/2}\left(1+\frac{R_1}{2}\mathrm{cth}\frac{\beta\hbar\Omega}{4}\right)^{-1}\left|\frac{\delta s}{s}\right|\right\}, \tag{51}$$

where a is the characteristic interatomic distance. If we use (43) in place of (49), we obtain an expression similar to (49) but with $R_1=0$. Although the sound-velocity renormalization is by itself small: $|\delta s/s|\sim 10^{-2}$ (see, e.g., Ref. 6), it is preceded in (51) by a large factor, on the order of several hundred. As a result, the Grüneisen coefficient turns out to be negative and can noticeably exceed unity in absolute value, as is indeed observed in experiment. Equation (51) also describes correctly the low-temperature behavior of the coefficient $\gamma_G(T)$, where it decreases, just as $|\delta s/s|$, and approaches zero asymptotically. This, unfortunately, cannot be said concerning the region of lower temperatures (≈ 10 K), so that an additional analysis is needed here.

7. CONCLUSION

The results obtained in the present paper, as well as in the two preceding ones,[7,8] make it possible to explain a large number of experimental facts involving the temperature dependence of a number of kinetic coefficients in amorphous dielectrics in practically the entire range of temperatures in which glass can exist. Despite the rather detailed exposition of the calculation results, it is necessary to discuss separately the basic assumptions made in the calculations.

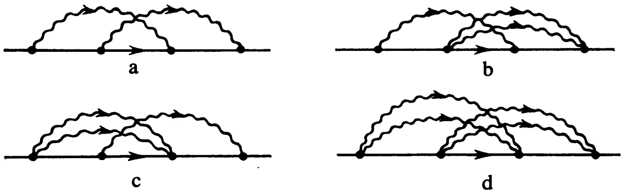


FIG. 6.

It is assumed throughout that the frequency of the tunneling particle is higher than the frequency of variation of the shape of the TLS potential. Strictly speaking, this assumption pertains not only to the multiphonon theory developed above, but also to the standard one-phonon theory.[1-3,9-11] In the opposite case it would be difficult to justify the separation of the phonon and intrawell degrees of freedom. In our case this assumption is necessary also to make the time of passage of the particle below the barrier shorter than the time of change of the barrier shape. The latter can in this case be regarded as static.

It is difficult to assess at present the degree to which this is justified for actually existing glasses, since we do not know which of the numerous presently existing microscopic models actually describe a two-level system. The adiabatic approximation should apparently be well satisfied in the TLS model proposed by Phillips.[22] In this case the change of the form of the TLS potential is due mainly to the relatively low-frequency oscillations of heavy clusters. If, on the other hand, some other TLS models are valid, the situation can be more complicated. In this case, however, a distinction must be made between the contribution of the zero-point oscillations and the thermal excitations. The former determine the renormalizations of constants which after all are taken from experiment. On the other hand, all the temperature dependences discussed above are connected with the contribution of thermal excitations, for which it suffices to require satisfaction of the weaker condition $k_B T < \hbar v$.

Besides the multiphonon processes considered above (see Figs. 2 and 3), there are also possible processes of higher order in the amplitudes (12) and (13), of the type shown in Fig. 6a. However, when separating processes of the type b, c, etc. in Fig. 3, we took into account the presence of a large parameter R, so that we cannot restrict ourselves to the first terms of the expansion of the exponential in $\Delta(\{\xi_\mu\})$ in power of ξ_μ even when the amplitudes $\hat{M}_d$ and $\hat{M}_n$ themselves are small. It is therefore necessary to consider not only the processes a in Fig. 6, but also processes of type b, c, d, etc. In this case we deal not simply with a two-phonon process (of type a in Fig. 6), but with a process in which two (or more) multiphonon packets participate. The duration of the emisson of one such a packet, and hence the time of its interaction with the TLS

$$\tau_p \sim (\hbar^2/F(T))^{1/2}$$

is shorter at $T > T_1$ than the characteristic phonon times

$$\tau_p \omega_D \ll 1. \qquad (52)$$

The contribution of a process with emisssion of one multiphonon packet (see Figs. 2 and 3) is proportional to $(\tau_p \omega_D)^{1/2}$ in the case of TLS relaxation or ultrasound damping, and to $\tau_p \omega_D$ in the case of sound-velocity renormalization. It is clear that processes with emission of a larger number of packets will contain the small parameter $\tau_p \omega_D$ to a higher power. This circumstance made it possible to neglect such processes in the calculations above.

With rising temperature, the reciprocal TLS relaxation time increases, i.e., the uncertainty ΔE increases. In this case the inequality

$$\hbar/\tau > k_B T. \qquad (53)$$

may hold still in the region of relatively low temperatures. Since the main contribution is always due to a TLS with $\Delta E \sim k_B T$, the inequality (53) means that the uncertainty of the energy levels of a TLS exceeds the distance between them. This by itself, however, may still not mean validity of the calculations above in the multiphonon regime. It is easy to verify that allowance for the imaginary part of ΔE in the calculations becomes necessary only when the TLS relaxation takes place in times shorter than the packet emission time, i.e., $\tau < \tau_p$. In the opposite case the relaxation can be neglected.

Estimates show that all the approximations discussed above can become invalid only at temperatures higher than T_3. This, however, leads apparently only to quantitative corrections, without changing the qualitative picture even at such high temperatures.

The authors are indebted to the late I. M. Lifshitz, to M. I. Kaganov, and to D. E. Khmel'nitsiĭ for helpful discussions of the results.

[1]W. A. Phillips, J. Low Temp. Phys. **7**, 351 (1972).
[2]P. W. Anderson, B. I. Halperin, and C. M. Varma, Phil. Mag. **25**, 1 (1972).
[3]Amorphous Solids, W. A. Phillips, ed., Springer, 1981.
[4]B. P. Smolyakov and E. P. Khaimovich, Usp. Fiz. Nauk **136**, 317 (1982) [Sov. Phys. Usp. **25**, 102 (1982)].
[5]A. C. Anderson, in Ref. 3, p. 65.
[6]W. A. Phillips, in Ref. 3, p. 53.
[7]V. N. Felurov and L. I. Trakhtenberg, Sol. St. Commun. **44**, 187 (1982).
[8]L. I. Trakhtenberg and V. N. Flerov, Zh. Eksp. Teor. Fiz. **83**, 1908 (1982) [Sov. Phys. JETP **56**, 1103 (1982)].
[9]J. A. Sussman, Phys. kond. Mat. **2**, 146 (1964). J. Phys. Chem. Sol. **28**, 1643 (1967).
[10]J. Jäckle, Z. Phys. **257**, 212 (1972).
[11]J. Jäckle, L. Piché, W. Rnold, and S. J. Nunklinger, J. Non-Cryst. Sol. **20**, 365 (1976).
[12]Yu. Kagan and M. I. Klinger, Zh. Eksp. Teor. Fiz. **70**, 255 (1976) [Sov. Phys. JETP **43**, 132 (1976)].
[13]V. L. Klochikhin, S. Ya. Pshezhetskiĭ, and L. I. Trakhtenberg, Dokl. Akad. Nauk SSSR **239**, 879 (1978).
[14]L. I. Trakhtenberg, V. L. Klochikhin, and S. Ya. Pshezhetsky, Chem. Phys. **69**, 121 (1982).
[15]Yu. Kagan and L. A. Maksimov, Zh. Eksp. Teor. Fiz. **65**, 622 (1973) [Sov. Phys. JETP **38**, 307 (1974)].
[16]L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1515 (1962) [Sov. Phys. JETP **20**, 1018 (1963)]
[17]S. Unklinger and M. von Schikfus, in : Amorphous Solids, **24**, 81 (1981), Ed. by W. A. Phillips, Springer, Berlin (Ref. 3).
[18]R. A. Street, T. M. Searle, and I. G. Austin, Amorphous and Liquid Semiconductors, W. E. Spear, ed., Univ. of Endinburgh, 1977, p. 392.
[19]M. T. Looponen, R. C. Dynes, V. Narayanamurti, and J. P. Garno, Phys. Rev. **B25**, 1161 (1981).
[20]L. Piché, R. Maynard, S. Hunklinger, and J. Jäckle, Phys. Rev. Lett. **32**, 1426 (1974).
[21]V. L. Gurevich and D. A. Parshin, Fiz. Tverd. Tela (Leningrad) **24**, 1372 (1982) [Sov. Phys. Solid State **24**, 777 (1982)].
[22]J. C. Phillips, Phys. Rev. **B24**, 1744 (1981).

Translated by J. G. Adashko