Theory of nonlinear relaxation absorption of ultrasound and electromagnetic waves in dielectric glasses

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The nonlinear relaxation absorption of ultrasound and electromagnetic microwaves in dielectric glasses is considered at low temperatures, when two-level systems (tunnel states) are responsible for the absorption. It is shown that in the nonlinear regime there exist three characteristic frequency ranges in which the dependences of the absorption on the frequency are different. With decrease in intensity, the high and low frequency regimes go over into the respective ranges of the linear theory, whereas the intermediate range disappears. For each of the frequency ranges mentioned, the dependences of the absorption on the wave intensity and on the frequency, and also on the temperature, are determined.

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1. INTRODUCTION

Many papers have appeared during the past several years on the study of the acoustic and electromagnetic properties of glasses (see, for example, Refs. 1 and 2). Interest in this field is due to the fact that many properties of glasses at low temperatures differ essentially from the corresponding properties of crystals and are to a large degree universal in this situation. That is, the properties do not depend on the specific glass composition. Such properties include the linear temperature dependences of the specific heat and the quadratic dependence of the thermal conductivity, and also the very unusual anomalies of the acoustic and electromagnetic properties, of which we shall speak later. Pracitcally all these anomalies find their explanation within the framework of a two-level-system model (TLS), which was proposed independently by Anderson *et al.* and by Phillips (Refs. 3–5). According to this model, the kinetic and thermodynamic properties of the glasses are explained by the presence in them of a TLS, the separation 2ε between the levels of which is smoothly distributed over a wide range of values. The lower bound of this interval, as contemporary experiments show, is less than 0.01 K, while the upper bound is determined from the condition that the separation between the levels be greater than their blurring due to relaxation.[6] This condition gives the value $\varepsilon_c \sim 10$–30 K for the upper boundary of the interval.

One of the most powerful methods of study of the TLS is the study of absorption of ultrasound and electromagnetic waves of the microwave band in glasses. These experiments make it possible to assess the TLS distribution in energy, and also their relaxation properties. Two different absorption mechanisms are connected with the TLS. The first is resonance absorption, which is connected with the direct absorption of a relaxational absorption due to relaxation of the level populations of the TLS upon change in the level separation under the action of the electromagnetic or sound waves. The relation between the corresponding contributions depends on the frequency ω and the temperature T. In the case $\hbar\omega \ll T$, in which we shall be interested, the coefficient of resonance absorption $\Gamma^{(\mathrm{res})}$ (for definiteness, we shall be speaking of (sound) is determined by the equation[1]

$$\Gamma^{(res)} = (\alpha/s)\,(\hbar\omega^2/T), \tag{1}$$

where α is a dimensionless coupling constant and s is the speed of sound. The coefficient of relaxation absorption is equal to[1,7]

$$\Gamma^{(rel)} = (\alpha/s)\begin{cases}\tau^{-1}, & \omega\tau \gg 1;\\ \omega, & \omega\tau \ll 1,\end{cases} \tag{2}$$

where τ is the minimum time of relaxation of the populations of the TLS for which $\varepsilon \approx T$. At $\omega\tau \ll 1$, the relaxation absorption always predominates. If $\omega\tau \ll 1$, the ratio of the contributions of the given mechanisms is determined by the dimensionless parameter $\hbar\omega^2\tau/T$, which can be either greater than or less than unity under experimentally achievable conditions.

As experiments show, the absorption becomes essentially nonlinear, even at very low intensities. As a rule, the nonlinearity of the absorption is connected with the equalizing of the level populations of the TLS in resonance transitions. Such an equalization appears even at sound intensities $\gtrsim 10^{-7}$ W/cm^2; at higher intensities the resonance mechanism is completely disengaged. Thus, even at relatively low intensities, the relaxation absorption becomes the principal form even in those cases in which the resonance absorption predominates in the linear regime.

We shall show that the relaxation absorption can also be nonlinear, and the investigation of the nonlinear relaxation absorption can give many important data on the relaxation properties of the TLS. As an example, we cite the recently interesting experiments of Elbaum *et al.*[8] on the observation of nonlinear absorption of ultrasound in metallic glasses, the acoustic properties of which are similar in many ways to the properties of dielectric glasses.

The purpose of the present work is the calculation of the nonlinear coefficient of relaxation absorption and the analysis of the conditions under which it can be observed.

In the next section, we write out the basic equations and

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formulate the setup of the problem. Before proceeding to the nonlinear theory, we shall show how to obtain qualitative estimates of the linear absorption under different limiting cases. We shall then construct the nonlinear theory of relaxation absorption, and in the last section, we shall discuss the physical meaning of the results and make appropriate estimates.[1)]

2. CALCULATION OF THE ABSORPTION COEFFICIENT

We write the Hamiltonian of the TLS in an external field in the form[7]

$$H=(\Delta+d\cos\omega t)\sigma_3+\Delta_0\sigma_1. \tag{3}$$

Here Δ is the energy gap between the levels of isolated potential wells, $\Delta_0=\hbar\omega_0\exp(-\lambda)$ is the tunnel matrix element (ω_0 is the characteristic frequency of oscillation of an atom in a single well, λ is the tunnel integral), σ_i are Pauli matrices; $d=\Lambda_{ik}u_{ik}^{(0)}$ in the case of a sound wave (Λ_{ik} is the deformation potential tensor of the TLS, $u_{ik}^{(0)}$ is the amplitude value of the deformation tensor) in the case of an electromagnetic wave $d=\beta\mathbf{E}_0$ (β is the dipole moment of the TLS, $\mathbf{E}_0$ is the amplitude of the electric field); as is usually done, we shall assume Δ and λ to be independent random quantities, uniformly distributed over a range of values much broader than that which makes the basic contribution to the absorption; we shall assume Λ_{ik} to be a random quantity, independent of Δ and λ, the distribution of which has a maximum near some value of the order of unity (in eV; a similar assumption can be introduced relative to β).

As has already been noted, we shall be interested in the case of rather low frequencies. On the other hand, the characteristic separation between the levels of the TLS, which makes a contribution to the absorption, is not less than T, as we shall see. Therefore the energy of a quantum of the external field turns out to be small in comparison with the characteristic separation between the levels. The slow variation of the external field makes it possible to use the adiabatic approximation for the solution of the quantum mechanical problem, i.e., in the solution of the Schrödinger equation we neglect derivatives of the external field with respect to time. The sufficient condition for the applicability of the adiabatic approximation, as analysis shows, is the inequality.

$$\hbar\omega\ll T^2/d,\ T. \tag{4}$$

Upon satisfaction of the condition (4), the TLS is characterized by the spacing 2ε between the levels, which depends on the instantaneous values of the field

$$\varepsilon=[(\Delta+d\cos\omega t)^2+\Delta_0^2]^{1/2}, \tag{5}$$

and the occupation numbers of the upper (n) and lower ($1-n$) levels. We can obtain for the occupation numbers the balance equation

$$\frac{dn}{dt}=\frac{n_0-n}{\tau}, \tag{6}$$

where

$$n_0=(e^{2\varepsilon/T}+1)^{-1}, \tag{7}$$

and the relaxation time τ is determined by the same expression as in linear theory[6,7]:

$$\frac{1}{\tau}=\frac{\Delta_0^2}{\tau_0T^2}\frac{\varepsilon}{T}\operatorname{cth}\frac{\varepsilon}{T}, \tag{8}$$

$$\tau_0=\frac{\varepsilon_c^2\hbar}{T^3},\quad \varepsilon_c\sim(\rho\hbar^3s^5/\Lambda^2)^{1/2}, \tag{9}$$

ρ is the glass density. It is important that Eq. (8) contains the time-dependent energy ε. We note that in the derivation of Eq. 6 we have assumed the phonons to be at equilibrium.

The absorbed power is determined by the expression

$$P=\left\langle 2N\int_0^\infty d\Delta\int_0^\infty\frac{d\Delta_0}{\Delta_0}\overline{n\dot\varepsilon}\right\rangle, \tag{10}$$

where N is the density of states of the TLS, and the angle brackets indicate averaging of the various values of Λ_{ik} or β_i. The bar above denotes averaging over the period:

$$\overline{n\dot\varepsilon}=\frac{\omega}{2\pi}\int_0^{2\pi/\omega}n\frac{d\varepsilon}{dt}dt. \tag{11}$$

In a number of limiting cases, it is convenient to use for $n\dot\varepsilon$ the following expression

$$\overline{n\dot\varepsilon}=\frac{\omega}{2\pi}\int_0^{2\pi/\omega}\frac{(n-n_0)}{\tau}(\varepsilon-\bar\varepsilon)\,dt, \tag{12}$$

which is obtained from (11) by integration by parts and using Eq. (6). In experiments on microwave absorption, measured quantity is usually P, while in acoustical experiments it is the coefficient of sound absorption

$$\Gamma=P/I, \tag{13}$$

where

$$I=\rho s^3(u_{ik}^{(0)})^2 \tag{14}$$

is the flux density of the sound energy.

3. LINEAR THEORY

In the linear approximation

$$\varepsilon=\varepsilon_0+\frac{\Delta}{\varepsilon_0}d\cos\omega t,\quad \varepsilon_0=(\Delta^2+\Delta_0^2)^{1/2} \tag{15}$$

$$n=n_0(\varepsilon_0)+n_1, \tag{16}$$

$$\dot n_1=-\frac{n_1}{\tau}+\frac{\Delta}{\varepsilon_0}\frac{\partial n_0}{\partial\varepsilon_0}\omega d\sin\omega t, \tag{17}$$

where τ is calculated at $\varepsilon=\varepsilon_0$. The basic contribution to the absorption is made by the TLS for which $n_0\approx 1$, i.e., $\varepsilon_0\lesssim T$.

We now analyze the contribution to the absorption of a single TLS, equal to $\overline{n\dot\varepsilon}$. If $\omega\tau\gg 1$, the TLS cannot relax within the period of the sound wave. The principal part of its population n does not depend on the time and, in the linear approximation, it can be assumed to be equal to $n_0(\varepsilon_0)\sim 1$. The time-dependent part of n is equal to $d/\omega\tau T$ in order to magnitude. According to (11), the contribution of this system to the absorption is of the order of $d^2/T\tau$. Systems for which $\omega\tau\ll 1$ relax within a time much shorter than the period, and their populations differ little from $n_0(\varepsilon)$. The nonequilibrium increment, which makes a contribution to the absorption, is $n-n_0(\varepsilon)\sim\omega\tau d/T$. To sum up, the absorbed power, as is seen from (11), is of the order of $\omega^2\tau v d^2/T$. We now recall that the relaxation time τ depends essentially on Δ_0 [see (8)]. If $\omega\tau_0 \gg 1$, the condition $\omega\tau \gg 1$ is satisfied for all TLS making a contribution to the absorption, since $\Delta_0 \lesssim T$ for them. Upon decrease in Δ_0, the power absorbed by such systems falls off as Δ_0^2 so that the important condition is $\Delta_0 \simeq T$. As a result, the absorbed power [see (17)] is

$$P \approx NT\frac{d^2}{T\tau_0} \approx \frac{Nd^2}{\tau_0}. \tag{18}$$

If $\omega\tau_0 \ll 1$, then systems with $\tau \gtrsim \tau_0$ can make a contribution to the absorption. For TLS with $\omega\tau \ll 1$, the aborbed power is proportional to Δ_0^{-2}. With decrease in Δ_0, this power increase, until $\omega\tau$ does not become of order unity (at $\Delta_0 \approx T(\omega\tau_0)^{1/2} \ll T$). Systems with $\omega\tau \approx 1$ also make a fundamental contribution to the absorption, so that

$$P \approx NT\omega d^2/T \approx N\omega d^2. \tag{19}$$

As has already been noted in Ref. 7, the absorption in this case does not depend on the relaxation time, and consequently does not depend on the temperature. This is a consequence of the exponentially broad distribution of the relaxation times.

4. NONLINEAR THEORY

Significant nonlinear effects can be expected when, in the case of a change in the separation between levels by an amount of the order d, a significant change can take place in the equilibrium population n_0, or in the relaxation time τ determined by the equilibrium number of phonons taking part in the transitions between levels. In either case we mean by the nonlinearity parameter the ratio d/T.

Generally speaking, Eq. (6) can be solved exactly and the absorbed power P can be expressed in the form of quadratures:

$$P = \left\langle \frac{N\omega}{2\pi T}\int_0^\infty d\Delta \int_0^\infty \frac{d\Delta_0}{\Delta_0}\int_0^{2\pi/\omega} dt \int_0^{2\pi/\omega} dt' \frac{\dot\varepsilon(t)\dot\varepsilon(t-t')}{\mathrm{ch}^2[\varepsilon(t-t')/T]} \times \exp\left[-\int_0^{t'}\frac{dt_1}{\tau(t-t_1)}\right]\left(1-\exp\left[-\int_0^{2\pi/\omega}\frac{dt_1}{\tau(t_1)}\right]\right)^{-1}\right\rangle,$$

where τ depends on t through $\varepsilon(t)$. However, the expression obtained is so complicated that it can be analyzed only under certain limiting cases. For this reason, we shall consider from the very beginning only the limiting cases which admit of clear physical interpretation. In particular, we shall consider only the case of a strong nonlinearity, when

$$d/T \gg 1. \tag{20}$$

We now consider the dependence of the energy of the TLS on the time, shown in Fig. 1. The maximum value of the energy is $\Delta_{\max} = [(\Sigma + |d|)^2 + \Delta_0^2]^{1/2}$, and the minimum $\varepsilon_{\min} = \Delta_0$. For TLS for which $\Delta_0 \gg T$ the condition $\varepsilon \gg T$ is satisfied throughout the entire period. Therefore such systems are unpopulated and do not make a contribution to the absorption. The same applies to systems for which $\Delta - |d| \gg T$. Thus, we shall be interested only in systems $\Delta_0 \lesssim T$ and $\Delta \lesssim d$. On the other hand, $\varepsilon_{\max} \gg T$ by virtue of the condition (20). We shall say that the system is in region I when its energy $\varepsilon \lesssim T$ and in region II when $\varepsilon \gg T$. The system is located in region I for a time interval $t_\mathrm{I} \sim T/d\omega$ that is much shorter than $2\pi/\omega$.

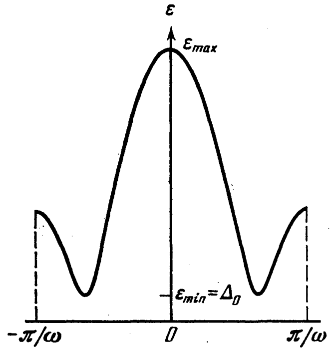

Fig. 1. Characteristic dependence of one half of the level spacing on the time.

In region II, the equilibrium population density is exponentially small. Therefore, are increase in the occupation numbers n is possible only in region I, while in region II the relaxation leads to a decrease in the occupation numbers. The character of the absorption depends significantly on how rapidly the relaxation of the system takes place in comparison with the time t_I of its being in region I, and also with the time $\approx 2\pi/\omega$ of its being in region II between two successive entries into region I.

In region I we have $\varepsilon \approx T$ and the characteristic relaxation time $\tau_\mathrm{I} \simeq \tau_0(T/\Delta_0)^2$. In region II, $\varepsilon \sim d$ and the characteristic relaxation time $\tau_\mathrm{II} \simeq \tau_\mathrm{I} T/d \ll \tau_\mathrm{II}$. As has been seen from consideration of the linear theory, the character of the absorption can depend significantly on how systems with $\Delta_0 \approx T$, for which $\tau_\mathrm{I} \simeq \tau_0$, relax. There are three limiting cases: $\omega\tau_\mathrm{II} \gg 1$, $\omega\tau_\mathrm{II} \ll 1$, and $t_\mathrm{I} \ll \tau_0$, $\tau_0 \ll t_\mathrm{I}$.

A. Case $\omega\tau_\mathrm{II} \gg 1$.

This inequality means that in region II the relaxation takes place slowly. A tthe same time, it follows from this that $t_\mathrm{I}/\tau_\mathrm{I} \approx (\omega\tau_\mathrm{II})^{-1}(T/d)^2 \ll (\omega\tau_\mathrm{II})^{-1} \ll 1$, the relaxation in region I is also slow. This means that the occupation numbers differ little from their average value $\bar n$ over the period, which values can be determined from the balance equation

$$\frac{n_0}{\tau_\mathrm{I}}t_\mathrm{I} \approx \frac{\bar n}{\tau_\mathrm{II}}\frac{1}{\omega}. \tag{21}$$

The left-hand side of this equation contains the increase in the occupation numbers in region I, and the right side their decrease in region II. From the relation (21) we have

$$\bar n \approx \frac{\tau_\mathrm{II}}{\tau_\mathrm{I}}\omega t_\mathrm{I} \approx \left(\frac{T}{d}\right)^2 \ll 1. \tag{22}$$

The absorption of a single TLS takes place principally in region II, so that Eq. (12) gives

$$\overline{n\dot\varepsilon} \approx \frac{\bar n}{\tau_\mathrm{II}}d \approx \frac{\Delta_0^2}{T\tau_0}. \tag{23}$$

It is then seen that the principal contribution to the absorption is made by systems for which $\Delta_0 \approx T$ and $\Delta \approx d$, meaning that

$$P \approx NdT/\tau_0. \tag{24}$$

In order to determine the numerical coefficient in this formula, we obtain $\bar{n}$ by averaging Eq. (6) over a period:

$$\bar{n} = [\overline{\varepsilon \operatorname{cth}(\varepsilon/T)}]^{-1} \left[\overline{\frac{\varepsilon}{\exp(2\varepsilon/T)-1}}\right]. \tag{25}$$

In the second factor of this expression (arrival) the principal contribution is made by region I, so that we can set

$$\omega t = x + {}^1\!/_2\pi + u, \quad |u| \ll 1, \tag{26}$$

where $x + \pi/2$ is that value of ωt where the energy has a minimum

$$\sin x = \Delta/d, \quad 0 \leqslant x \leqslant \pi/2. \tag{27}$$

Here the energy has the form

$$\varepsilon = [d^2u^2\cos^2 x + \Delta_0^2]^{1/2} \tag{28}$$

and integration over t reduces to integration over u in the limits from $-\infty$ to $+\infty$. In the first factor (departure the principal contribution is made by regionII, so that we can set $\varepsilon = d\,|\sin x + \cos \omega t\,|, \coth(\varepsilon/T) \approx 1$ in it. As a result,

$$\bar{n} = \frac{(T/d)^2}{2(\cos x + x\sin x)\cos x} \int_{-\infty}^{\infty} \frac{d\xi[\xi^2 + (\Delta_0/T)^2]^{1/2}}{\exp\{2[\xi^2+(\Delta_0/T)^2]^{1/2}\}-1}. \tag{29}$$

It is taken into account here that region I reached twice in one period. According to (12), the power absorbed by a single system is

$$\overline{n\dot{\varepsilon}} = \frac{\bar{n}\omega}{\pi}\int_0^{\pi/\omega} dt\varepsilon/\tau = \frac{\Delta_0^2 d^2}{T^3\tau_0}\left(\frac{1}{2} + \sin^2 x\right)\bar{n}. \tag{30}$$

Substituting this expression in (10) and integrating over Δ_0 and ξ, we obtain

$$P = \frac{\pi^4 a}{120}\frac{N\langle|d|\rangle T}{\tau_0}, \quad a = \frac{1}{2}\int_0^{\pi/2}\frac{1+2\sin^2 x}{\cos x + x\sin x}dx \approx 1.2. \tag{31}$$

B. Case $\omega\tau_{\mathrm{II}} < 1$, $t_{\mathrm{I}} < \tau_0$

In this case, within the time of passage through region I, the system with $\Delta_0 \approx T$ does not manage to relax to the equilibrium value, while in region II, it relaxes within a time that is much shorter than the period. Thus, such systems go from region II to region I with negligibly small values of the occupation numbers n. In passing through region I, n increases to the value

$$n_m \approx n_0\frac{t_{\mathrm{I}}}{\tau_{\mathrm{I}}} \approx \frac{1}{\omega\tau_0}\frac{\Delta_0^2}{Td}. \tag{32}$$

On going into region II, the systems relax in a small neighborhood of region I. In this neighborhood, $\varepsilon < d$. Therefore the relaxation time there is $t_{\mathrm{II}} \approx (\Delta_0^2 \varepsilon/\tau_0 T^3)^{-1} \gg \tau_{\mathrm{II}}$. On the other hand, in this neighborhood we can use the substitution (26) and the expression for the energy (28), where $u \sim \omega t_{\mathrm{II}}$. By virtue of the conditions $\varepsilon \gg T$ (i.e., the region of relaxation is much greater than the region I, $t_{\mathrm{II}} \gg t_{\mathrm{I}}$) and $\Delta_0 \lesssim T$, the expression (28) is simplified: $\varepsilon \approx d\omega t_{\mathrm{II}}$. As a result we obtain for t_{II} the following estimate:

$$t_{\mathrm{II}} \approx \left(\frac{T^3\tau_0}{\Delta_0^2 d\omega}\right)^{1/2} \approx t_{\mathrm{I}}\left(\omega\tau_{\mathrm{I}}\frac{d}{T}\right)^{1/2} \gg t_{\mathrm{I}}. \tag{33}$$

The power absorbed by a single system is

$$\overline{n\dot{\varepsilon}} \approx n_m d\omega \cdot \omega t_{\mathrm{II}} \approx \Delta_0(T\omega/d\tau_0)^{1/2}. \tag{34}$$

It is then seen that values $\Delta_0 \approx T$ are significant, and since $\Delta \lesssim d$ the total absorbed power is

$$P \approx Nd\overline{n\dot{\varepsilon}} \approx NT^{3/2}(d\omega/\tau_0)^{1/2}. \tag{35}$$

For the determination of the numerical coefficient, we first calculate the maximum value n_m achieved by the occupation numbers in the passage through region I. We recognize that in region I we can neglect the relaxation term $-n/\tau$ in Eq. (6), make use of the substitution (26), and set $n = 0$ as $u \to -\infty$. Then

$$n_m = n|_{u\to+\infty} = \frac{1}{\omega\tau_0}\frac{\Delta_0^2}{Td\cos x}\int_{-\infty}^{\infty}\frac{d\xi[\xi^2+(\Delta_0/T)^2]^{1/2}}{\exp\{2[\xi^2+(\Delta_0/T)^2]^{1/2}\}-1}. \tag{36}$$

In region II, we can neglect the source n_0/τ in Eq. (6) and neglect the quantity Δ_0^2 in the expression for the energy (28). Selecting $n|_{u=0} = n_m$ as the initial value in this region, we obtain

$$n(u) = n_m \exp\left(-\frac{\Delta_0^2 d\cos x}{2T^3\omega\tau_0}u^2\right). \tag{37}$$

Substituting this expression in (11), taking it into account that $\dot{\varepsilon} = d\omega$ in this region, integrating over u from 0 to ∞, keeping it in mind that there are two regions I in the period, we obtain

$$\overline{n\dot{\varepsilon}} = n_m\left(\frac{dT^3\omega^3\tau_0}{2\pi\Delta_0^2}\cos x\right)^{1/2}. \tag{38}$$

Substituting (38) in (10), we have with account taken of (36)

$$P = \frac{\pi^2\zeta(3)}{\Gamma^2({}^1\!/_4)}NT^{3/2}(\omega/\tau_0)^{1/2}\langle|d|^{1/2}\rangle. \tag{39}$$

C. Case $\tau_0 < t_{\mathrm{I}}$

In this case, for systems with $\Delta_0 \approx T$, the relaxation should occur in region I; in region II, the occupation numbers are negligibly small and we do not have to consider this region. For systems with $\tau_{\mathrm{I}} < t_{\mathrm{I}}$, all the arguments advanced above for the case of the linear theory at $\omega t < 1$ are valid, except that the role of the period for them is played by the time t_{I}. As a result, it turns out that the principal contribution to the absorption is made by those systems for which $\tau_{\mathrm{I}} \approx t_{\mathrm{I}}$, i.e., $\Delta_0 \approx T(\tau_0/t_{\mathrm{I}})^{1/2}$. Obviously, for systems with $\tau_{\mathrm{I}} \approx t_{\mathrm{I}}$, the occupation numbers are of the order of unity, while $\dot{\varepsilon}$ in region I is of the order of $d\omega$ for these systems, by virtue of the condition $\Delta_0 < T$.

Thus, the power absorbed by a single system is

$$\overline{n\dot{\varepsilon}} \approx d\omega \cdot \omega t_{\mathrm{I}} \approx \omega T, \tag{40}$$

and the total absorbed power is

$$P \approx Nd\overline{n\dot{\varepsilon}} \approx Nd\omega T. \tag{41}$$

For the determination of the numerical factor, we again use the substitution (26), and neglect the quantity Δ_0 in the expression for the energy (28). Equation (6) is solved with the initial condition $n = 0$ as $u \rightarrow -\infty$, while in the calculation of n by Eq. (11), the integration over u is carried out in the limits from $-\infty$ to $+\infty$. As a result, $P = bN\omega T\langle |d|\rangle$, where

$$b = \frac{1}{2}\int_0^\infty d\eta \int_{-\infty}^{\infty} d\xi \,\mathrm{sgn}\,\xi \int_{-\infty}^{\xi} d\xi' \frac{|\xi'|}{e^{2|\xi'|}-1} \times \exp\left(-\eta \int_{\xi'}^{\xi} d\xi''\,\xi''\,\mathrm{cth}\,\xi''\right) = 0.89. \quad (42)$$

5. DISCUSSION OF THE RESULTS

In considering the limits of applicability of cases A, B, and C, if it is not difficult to verify that in the nonlinear regime, in contrast to the linear, there are not two but three characteristic frequency intervals. Case A corresponds to the high frequencies:

$$\omega\tau_0 \gg d/T, \quad (43)$$

case C, to the low frequencies:

$$\omega\tau_0 \ll T/d, \quad (44)$$

while case B corresponds to the intermediate frequencies:

$$T/d \ll \omega\tau_0 \ll d/T. \quad (45)$$

It is not difficult to show that at $d/T \approx 1$ the results obtained for cases A and C go over to the results of the linear theory in order of magnitude, for the limiting cases $\omega\tau_0 \gg 1$ and $\omega\tau \ll 1$, respectively. As for the frequency interval B, it vanishes in the case $d/T \approx 1$, as is seen from (45).

The frequency dependence of the absorption coefficient (for definiteness, we shall now speak of the case of sound) is characterized by the fact that in the low-frequency interval C we have $\Gamma \propto \omega$, while $\Gamma \propto \omega^{1/2}$ in the intermediate interval B and Γ does not depend on ω in the high-frequency interval A.

The dependence of Γ on the intensity has the same character for all values of $\omega\tau_0$ (although the absolute value of Γ depends on $\omega\tau_0$) and is shown schematically in Fig. 2. The intensity I_1 at which the nonlinear effects set in is determined by the condition $d \approx T$. On going through I_1, we find ourselves either in region A at $\omega\tau_0 \gg 1$, or in region C at $\omega\tau_0 \ll 1$.

We note that the $\Gamma(I)$ dependence in the region $I_1 \ll I \ll I_2$ is the same as the asymptotic dependence for the resonance absorption.[1] The threshold value of I_1 is proportional to T^2 and does not depend on ω. We note that for resonance absorption, the theory of Ref. 9 predicts the following dependences of the critical intensity: $I_c \infty \omega^2 T^2$. In the experiment of Ref. 10, a quadratic dependence of I_c was observed. However, I_c is practically independent of ω. The reason for this divergence is apparently not clear at the present time.

We estimate the possibility of realization of the nonlinear regime as applied to sound. Setting $\rho \approx 5$ g/cm³, $s \approx 2 \times 10^5$ cm/s and ≈ 2 eV we obtain

$$d\,[\mathrm{erg}] \approx (4 \cdot 10^{-17} I\ [\mathrm{W/cm^2}])^{1/2}.$$

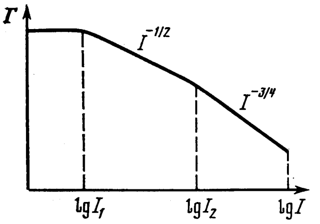

Fig. 2. Schematic form of the dependence of the coefficient Γ of the relaxation absorption of the ultrasound on its intensity.

Thus, at $I = 1$ W/cm² and $T = 0.1$ K we obtain $d/T \simeq 3$.

The quantity $\omega\tau_0$ can vary over wide limits because of the strong dependence of τ_0 on the temperature. At $T \approx 0.1$ K and $\varepsilon_c \approx 20$ K we obtain $\tau_0 \approx 4 \times 10^{-6}$ s. Thus, the low-frequency region is reached at frequencies below 1 MHz.

In conclusion, we shall discuss what information on the TLS can be obtained from experiments on low-temperature relaxation absorption. The frequency and temperature dependence of Γ allow us to assess how well the usually employed TLS model describes the real situation in glasses. However, from this point of view, the nonlinear absorption is not much greater than the linear. What is essentially new in the nonlinear absorption is its dependence on the intensity. In the first place, we can estimate directly the value of the deformation potential (or the dipole moment in the case of microwave absorption) from the threshold value I_1. Furthermore, the data on the linear absorption coefficents of longitudinal and transverse sound allow us in principle to determine $\langle(\mathrm{Sp}\,\mathcal{A})^2\rangle$ and $\langle \mathrm{Sp}\,\mathcal{A}^2\rangle$. The nonlinear absorption contains more complicated functions of the tensor $\mathcal{A}_{ik}$ and thus permits us to assess not only the order of magnitude of its characteristic values, but also on their distribution.

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[1)] It has been made known to the writer that similar results have been obtained independently by B. D. Laĭkhtman.

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