## Nonlinear relaxation absorption of ultrasound in amorphous metals

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A theory of nonlinear relaxation absorption of sound by two-level systems (TLS) in amorphous metals (metallic glasses) is developed. Absorption nonlinearity arises when the amplitude of the modulation of the separation between the levels of the TLS by a sound wave exceeds the temperature T. The existence is established of two frequency ranges in which the behavior of the relaxation absorption has different dependences on the sound intensity J, on the frequency  $\omega$ , and on the temperature. Nonlinear effects become significant at  $J > J_{c2}$ , where the critical intensity  $J_{c2}$  is proportional to  $T^2$  and to the cube of the sound velocity. At  $J > J_{c2}$ , the absorption coefficient is  $\Gamma \approx \Gamma_0 [J/J_{c2})^{-1/2}$  in the high-frequency region and  $\Gamma \approx \Gamma_0 [a_1(J_{c2}/J)^{1/2} + a_2/\ln(J/J_{c2}(\omega\tau_0)^2)]$  in the low-frequency region. Here  $\Gamma_0$  is the linear absorption coefficient in the corresponding frequency range,  $\tau_0$  is the characteristic relaxation time of the population density of the TLS;  $a_1$  and  $a_2$  are numerical coefficients of the order of unity. The features of nonlinear relaxation absorption in amorphous superconductors are discussed briefly.

#### **1. INTRODUCTION**

A new type of nonlinear behavior of sound absorption has recently been observed in metallic glasses at low temperatures.<sup>1-4</sup> That is, two steps were seen on the curve of the dependence of the absorption coefficient on the intensity.<sup>4</sup> The first step is connected with the nonlinearity of the well known resonance absorption. So far as the other is concerned, it can be connected, as has been shown by us,<sup>5</sup> with the nonlinearity of the relaxation absorption. In our brief note,<sup>5</sup> qualitative considerations were expressed and orderof-magnitude estimates given. Here we wish to present a more complete quantitative theory and to analyze the various limiting cases. At the same time, we would wish to correct certain inaccuracies present in Ref. 5.

The relaxation absorption is due to the modulation of the population density f, of two-level systems (TLS) by the alternating deformation field of the sound wave. The modulation is due to the time-periodic change in the level spacing E, which is assumed to depend on time t and is given by the following expression:

$$E(t) = [(\Delta + d\cos\omega t)^{2} + \Delta_{0}^{2}]^{\frac{1}{2}}, \qquad (1)$$

Here  $\Delta$  and  $\Delta_0$  are parameters that characterize the Hamiltonian of the TLS in the absence of deformation, which has the form

$$^{i}/_{2}\Delta\sigma_{3}-^{i}/_{2}\Delta_{0}\sigma_{1}$$

where  $\sigma_i$  are Pauli matrices, d is the amplitude of the change of the parameter  $\Delta$  under the action of the deformation created by the sound wave:  $d = 2\gamma_{ik} u_{ik}^0$ , where  $\gamma_{ik}$  is the tensor of the deformation potential and  $u_{ik}^0$  is the amplitude of the deformation in a sound field with frequency  $\omega$ . The corresponding change in the tunnel transmissivity  $\Delta_0$  we shall, following Jackle<sup>6</sup> and Black,<sup>7</sup> assume to be small and neglect.

Such a modulation of the spacing E produces in the population density f of the upper level a change (in comparison with its equilibrium value  $f_0$ ) which lags in phase the

splitting. This also leads to sound-wave energy dissipation. The power absorbed by a single TLS is

$$p = \langle j \vec{E} \rangle = \frac{\omega}{2\pi} \int_{0}^{2\pi/\alpha} dt f(t) \frac{dE}{dt}, \qquad (2)$$

where the angular brackets indicate averaging over the period. The derivation of this formula is given in Sec. 4. It is obtained in the adiabatic approximation. In the general case, the conditions of its applicability  $are^{1}$  (see Ref. 8 and the literature cited there):

$$\hbar |dE/dt| \ll E^2, \quad \hbar \omega \ll E. \tag{3}$$

They represent the requirement of sufficient slowness of the change in the perturbation, which should not by itself produce quantum transitions.

The occupation numbers f are determined from the equation:

$$\frac{\partial f}{\partial t} = -\frac{f - f_0}{\tau} \quad , \tag{4}$$

where

 $f_0 = (1 + e^{E/T})^{-1}$ 

is the equilibrium population density of the upper level,  $\tau$  is the relaxation time of the TLS. As is known (see the review of Black<sup>7</sup>), in metallic glasses it is due primarily to the interaction of the TLS with the conduction electrons; the expression for it has the form

$$\frac{1}{\tau} = \frac{\pi}{4\hbar} \chi \frac{\Delta_0^2}{E} \operatorname{cth} \frac{E}{2T}.$$
(5)

Here

$$\chi = \overline{V}^2 / V_0^2, \tag{6}$$

the matrix element V characterizes the change in the parameter  $\Delta$  due to interaction of the TLS with the conduction electrons; the bar indicates averaging over the states of the electrons with which the TLS interacts (see below, Sec. 4); 1/  $V_0$  is the density of electron states at the Fermi level per electron.<sup>2)</sup> Thus, generally speaking, the relaxation time  $\tau$  entering into (4) should be regarded as dependent on the time t according to Eqs. (1) and (5)—by virtue of the  $\tau(E)$  dependence.

In order to obtain the total absorption, the absorption  $p(\Delta, \Delta_0)$  from the individual must be summed over all TLS, viz., over the parameters  $\Delta$  and  $\Delta_0$  that characterize them and are assumed to be random:

$$P = \int_{0}^{\infty} d\Delta \int_{0}^{\infty} \frac{d\Delta_{0}}{\Delta_{0}} N(\Delta, \Delta_{0}) p(\Delta, \Delta_{0}).$$
 (7)

Here  $N(\Delta, \Delta_0)$  is the corresponding distribution function, which at its presently known accuracy is regarded as a constant,  $N_0$ .<sup>3)</sup> The final expression for *P* should have included averaging also over all possible values of the deformation potential  $\gamma_{ik}$ . We shall not write down this procedure nor give it in explicit form. The fact is that at the present time there are no reliable data on the width of this distribution and especially to the degree of which it correlates with the distributions of the parameters  $\Delta$  and  $\Delta_0$ . We can only state that a typical absolute value of  $\gamma$  is of the order of 1–3 eV.

The results that will be given below were obtained under the assumption of the absence of a correlation between the parameters  $\gamma_{ik}$  and  $\Delta_0$ ,  $\Delta$  for some definite value of the same quantity  $\chi$  for all TLS. It is necessary to note, however, that the frequency and temperature dependences of the absorption coefficient in limiting cases remain the same even under more general assumptions concerning the character of the distribution of these random quantities (cf. for example, the averaging method used in Ref. 9).

At low sound amplitudes (the so-called linear region), Eq. (4) must be solved in the linear approximation in d; here  $\tau$ can be assumed to be independent of the time t, and for the contribution (2) from a single TLS to the absorption we obtain a formula of the Mandel'shtam-Leontovich type:

$$p = \frac{1}{2} d^2 \frac{\Delta^2}{E^2} \omega \frac{\omega \tau}{1 + (\omega \tau)^2} \frac{1}{4T \operatorname{ch}^2(E/2T)}.$$
 (8)

It is clear from this expression that the fundamental contribution to the absorption is made by the TLS with  $E \leq T$ . On the other hand, the relaxation time  $\tau$  depends on  $\Delta_0$  as well as on E. Correspondingly, for each characteristic value of E we can indicate a minimum relaxation time  $\tau_{\min}$  corresponding to  $E = \Delta_0$ , to wit,

$$\frac{1}{\tau_{min}(E)} = \frac{\pi \chi}{4\hbar} E \operatorname{cth} \frac{E}{2T}.$$
(9)

It is seen from (9) that the minimum relaxation time  $\tau_{\min}(E)$  has the same value in metals both at  $E \approx T$  and at  $E \ll T$ :

$$1/\tau_{min} \approx \pi \chi T/2\hbar = 1/\tau_0. \tag{10}$$

At  $E \ll T$  this equality becomes exact.

We begin with a discussion of the results of linear theory,<sup>7</sup> in order to compare it later with the nonlinear case. The frequency and temperature dependence of the linear absorption coefficient is determined by the parameter  $\omega \tau_0$ .

If  $\omega \tau_0 \ge 1$ , then the condition  $\omega \tau \ge 1$  is satisfied for all TLS with  $E \le T$ . This means that we can neglect the term 1 in the denominator of (8) in comparison with  $(\omega \tau)^3$ . In the up-

shot, the absorption coefficient turns out to be independent of the frequency and proportional to  $\tau_0^{-1}$ , i.e., to the temperature. According to (7) and (8), it is equal to<sup>10</sup>

$$\Gamma_{0j} = \frac{\pi^3}{24} \frac{N_0 \gamma_j^2 \chi}{\rho \hbar v_j^3} T, \qquad (11)$$

Here j = l, t is the index of polarization (longitudinal and transverse),  $\rho$  is the density of the glass,  $v_j$  is the velocity of sound,

$$\gamma_t^2 = \frac{1}{15} \left\langle \left( \operatorname{Sp} \gamma \right)^2 + 2 \operatorname{Sp} \gamma^2 \right\rangle,$$
  

$$\gamma_t^2 = \frac{1}{30} \left\langle \left( 3 \operatorname{Sp} \gamma^2 - (\operatorname{Sp} \gamma)^2 \right) \right\rangle.$$
(12)

The averaging in (12), which is denoted by  $\langle ... \rangle$ , is carried out over all TLS. As follows from (12), the inequality

$$\gamma_t^2 > 4/_3 \gamma_t^2 \tag{13}$$

holds. Experimentally, the independence of the sound absorption coefficient of frequency and its proportionality to the temperature were observed in Ref. 10 in amorphous Ni– P in the range of frequencies of the order of hundreds of MHz and at temperatures in the range of seveal tens of degrees.

In the opposite limiting case,  $\omega \tau_0 \ll 1$ , the physical picture is as follows. the TLS with  $\tau \ll 1/\omega$  relax rapidly, their population densities differ little from equilibrium, and their contribution to the absorption is small. The TLS with  $\tau \gg 1/\omega$ "feel" only the average field during the time  $\tau$ , and therefore their population densities are almost time-independent, so that their contribution to the absorption is also small. This means that the fundamental contribution to the absorption is made by TLS with

$$\tau(E, \Delta_0) \approx 1/\omega.$$

There are always TLS with such  $\tau$ , therefore the distribution of  $\tau$  over the parameter  $\Delta_0$  is assumed to be very broad. The absorption in this case does not depend on T and is proportional to  $\omega$  (Refs. 7, 11, 12):

$$\Gamma_{oj} = \frac{\pi}{2} \frac{N_0 \gamma_j^2}{\rho v_j^3} \omega.$$
(14)

Such a dependence of the absorption coefficient has been observed experimentally in several glasses.<sup>12,13</sup>

The range of frequencies and temperatures at which the condition  $\omega \tau_0 \ll 1$  is satisfied in metallic glasses is achieved in most experiments, and the sound absorption under these conditions permits experimental observation. On the other hand, for the satisfaction of the condition  $\omega \tau_0 \gg 1$  [with  $\tau_0$  determined by (10)], either very high frequencies or very low temperatures are required. Therefore, the dependence (11) has been observed only in Ni–P glass, which, at the experimental temperature ( $\approx 0.3$  K) is still in the normal state.

However, at low temperatures, many metallic glasses undergo a transition to the superconducting state. Here, as Black and Fulde have shown,<sup>14</sup> the relaxation time of the TLS due to electron excitations increases exponentially with decrease in the temperature. Therefore, at sufficiently low temperatures, the relaxation time  $\tau_0$  is determined by the interaction of the TLS with the phonons and the situation turns out to be the same as in dielectric glasses, where  $\tau_0$  (at  $T \approx 1 \text{ K}$ ) is three orders of magnitude greater than in metallic glasses and, as a rule, the situation with  $\omega \tau_0 \gg 1$  is realized experimentally.<sup>15</sup>

All these considerations are valid in the absence of a magnetic field. A magnetic field exceeding the critical value destroys the superconductivity and we are returned to the picture of a normal amorphous metal (see Ref. 16).

It is important to note that in obtaining the result (14), the decisive circumstance leading to such frequency and temperature dependences is the assumption that the random quantity  $\ln \Delta_0$  has an equilibrium or near-equilibrium distribution. A consequence of this assumption is the presence of a broad spectrum of relaxation times  $\tau$  at a given energy *E*. The experimental verification of the presence of such a broad spectrum of relaxation times in metallic glasses has been given in Refs. 12 and 13.

### 2. NONLINEAR ABSORPTION. QUALITATIVE PICTURE

We begin with the remark that in the linear region the relaxation absorption is determined in all cases by TLS with  $E \approx T$ . The boundaries of the nonlinear region are determined by the inequality  $d \gtrsim T$ . We shall limit ourselves in this analysis to the case of strong nonlinearity [the general picture will be given in Sec. 3; its consideration requires numerical calculations according to Eqs. (25) and (26)];

 $d\gg T$ .

For this case we analyze the dependence of the level separation E on the time t. If  $\Delta_0 \gg T$ , this separation is also much greater than T at all instants of time, so that such TLS cannot be excited in the interaction with the equilibrium electron system and make a contribution to the absorption. The same circumstance holds for systems in which  $\Delta \gg d$ . Thus, the effective contribution to the absorption can be made only by systems for which  $\Delta \leq d$  and  $\Delta_0 \leq T$ . The separation between levels of such TLS reaches a minimum value  $\Delta_0$  at the instant of time  $t_0$  determined by the condition

 $\Delta + d \cos \omega t_0 = 0.$ 

In the immediate vicinity of these instants of time, the TLS can be excited by the electrons (see Fig. 1).

The interval of times in which such excitation is possi-



FIG. 1. The time dependence of the spacing E between levels of the TLS under the conditions  $\Delta_0 < T$ , d > T. The interval  $t_T \approx T/d\omega$  is the time interval within which E(t) < T.

ble is of the order of

 $t_T \equiv T/d\omega \ll \omega^{-1}$ .

The characteristic relaxation time in the course of this interval is  $\tau_0$ . Therefore, two additional limiting cases are possible.

1) High-frequency:  $\omega \tau_0 \gg T/d$   $(t_T/\tau_0 \ll 1)$ —the relaxation does not take place in the thermal layer in the course of a single period of the sound wave.

2) Low-frequency:  $\omega \tau_0 \ll T/d$   $(t_T/t_0 \gg 1)$ —the relaxation takes place (by relaxation, we mean both excitation of the TLS by electrons and their de-excitation; in the thermal layer, E < T, the times of both these processes are of the same order).

In order to analyze qualitatively the relaxation absorption in both limiting cases, we introduce the quantity  $\lambda(E) = t_E/\tau$ , where  $t_E \approx E/d\omega$  is the time during which the energy *E* changes by a quantity of the order of itself. The physical meaning of  $\lambda(E)$  is that it characterizes the probability of relaxation of the TLS, W(E), during the time  $t_E$ :

$$W(E) \approx 1 - e^{-\lambda/(E)}.$$
(15)

Since this expression is required by us only for qualitative analysis, we shall not trouble ourselves with the determination of the numerical coefficients in the exponent of Eq. (15). All that is important for us is only that at  $\lambda(E) \leq 1$  the probability  $W(E) \approx \lambda(E)$  and at case  $\lambda(E) \geq 1$  the probability W(E) = 1. Using Eq. (5) for  $\tau$  we obtain for  $\lambda(E)$ :

$$\lambda(E) \approx \frac{\Delta_0^2}{dT \omega \tau_0} \operatorname{cth} \frac{E}{2T} \approx \begin{cases} \Delta_0^2 / dE \omega \tau_0 & \text{at} \quad E < T, \\ \Delta_0^2 / dT \omega \tau_0 & \text{at} \quad E > T. \end{cases}$$
(16)

This quantity reaches its maximum value  $\lambda_{\max} = \Delta_0 / \Delta_{01}$ , where  $\Delta_{01} = d\omega\tau_0$  at the instant of time  $t_0$  when  $E \approx \Delta_0$ . The minimum value  $\lambda_{\min} (\Delta_0 / \Delta_{02})^2$  where  $\Delta_{02} = (dT\omega\tau_0)^{1/2}$ , is attained when  $E \approx T$ . It is important that  $\lambda$  (E) does not depend on E at  $E \gtrsim T$ .

The high-frequency case  $\omega \tau_0 \gg T/d$  permits the simplest qualitative analysis, and we shall therefore begin with it. In this case,  $T \ll \Delta_{02} \ll \Delta_{01}$ . Taking it into account that we are interested in systems with  $\Delta_0 \leq T$ , we obtain the result that  $\lambda_{\max} \ll 1$ . This means that the TLS, passing the thermal layer in energy (E < T) can be excited in it only with low probability,  $W \approx \lambda_{\max}$ . This probability is maximal for systems with  $\Delta_0 \approx T$ ; therefore, the disequilibrium of their distribution is very large and they will make the main contribution to the absorption.

Such TLS, excited in the thermal layer, i.e., drawing an energy of order T from the electron subsystem, will remain in an excited state for the duration of many periods (the number of which is of order  $\tau_0/t_T$ ). Since the probability of deexcitation of such a TLS does not depend on the energy, upon de-excitation, it will return an average energy of order  $d \ge T$  to the electron subsystem. Thus, in the mean and over a time of order  $(2\pi/\omega)(\tau_0/t_T)$  the considered TLS will be excited once. Therefore, the power absorbed by it will be

$$p \approx d/(2\pi/\omega) (\tau_0/t_T) \approx T/\tau_0. \tag{17}$$

The power absorbed in a unit volume by such systems is obtained by the multiplication of (17) by the concentration of

such TLS, which is of the order of  $N_0 d$ . Dividing the obtained quantity by the sound-energy flux density (the intensity)  $J \approx (d\gamma)^2 \rho v^3$ , where  $\gamma$  is the characteristic value of the deformation potential tensor  $\gamma_{ik}$ , we find the estimate for the absorption coefficient:

$$\Gamma \approx \Gamma_0(T/d), \tag{18}$$

where  $\Gamma_0$  is determined by Eq. (11) [for a detailed derivation of (18), see Sec. 3, Eq. (31)].

Thus, in the high-frequency case, the relation between the nonlinear and linear coefficients of absorption is the same as in dielectric glasses (see Ref. 8 and the literature cited there). However, since  $\Gamma_0$  depends on the temperature, the temperature dependence of the absorption in the linear regime is  $\Gamma \approx T^2 J^{-1/2}$ .

The low-frequency case,  $\omega \tau_0 \ll T/d$ , is much more complicated. Here  $\Delta_{01} \ll \Delta_{02} \ll T$ . This means that TLS with  $\Delta_0 \approx T$  passing through the thermal region E < T, is repeatedly excited and de-excited by the electrons  $[\lambda(E) \gg 1]$ . On leaving the thermal layer, they discard the excitation almost instantly, and in the superthermal region E > T they are always in the ground state. Thus, their distribution relaxes very rapidly to the equilibrium value  $f_0$  and hence the contribution of such systems to the absorption is small (similar to the low-frequency case in linear theory). In this situation, the principal contribution is made by TLS for which  $\Delta_0 \ll T$ . By virtue of the smallness of the tunnel transmissivity  $\Delta_0$ , such TLS relax to equilibrium more slowly and their population density can be significantly far from the equilibrium one.

It is easy to understand which TLS makes the principal contribution to the absorption. This is the TLS with  $\Delta_0$  determined from the condition  $\lambda_{\min} \approx 1$ , i.e., with  $\Delta_0 \approx \Delta_{02}$ . We shall call such TLS intermediate. For them,  $\tau(\Delta_0,$  $E = T \sim t_T$ . The unusual physical phenomenon of "protraction of relaxation," which is absent in dielectric glasses, is connected with the intermediate TLS. Leaving the thermal layer, such a TLS can turn out to be in an excited state with probability of almost unity. Since the quanity  $\lambda$  (E) does not depend on the energy at  $E \gtrsim T$ , this TLS can, with probability of the order of unity, be de-excited, transferring energy of the order of T to the electrons, but it can also keep its relaxation, reach an energy  $E \approx d$ , and only then give it off to the electron subsystem. Correspondingly, the power absorbed per unit volume by an intermediate TLS can represent the sum of two terms:  $N_0 d\omega T$  and  $N_0 d\omega d$ . Allowance for the first term against the background of the second may seem unjustified, since we assume that the condition  $d \ge T$  is satisfied. Actually, the absolute value of the absorbed power is determined by the second term, and the sound absorption coefficient turns out to be of the same order of magnitude as in the linear theory [see Eq. (14)]. The meaning of the separation of the small first term is that frequently it is not the absolute value of the sound absorption coefficient that is measured experimentally, but only the part that is dependent on the intensity. This value is determined by just this term. The quantitative analysis carried out in Sec. 3 gives the following dependence of the absorption coefficient on the intensity in the low-frequency case:

$$\Gamma = \Gamma_0 \left[ a_1 \frac{T}{d} + a_2 \frac{\mathbf{1}}{\ln \left( d/T \omega \tau_0 \right)} \right]. \tag{19}$$

where  $a_1$  and  $a_2$  are numerical coefficients of order unity, while  $\Gamma_0$  is determined by Eq. (14). Equation (19) is consistent with our qualitative considerations. It is seen from it that at  $d\omega\tau_0 \ll T$  the dependence  $\Gamma(J)$  represents the sum of the relatively rapidly decreasing term  $J^{-1/2}$  and the relatively slowly decreasing part  $\ln^{-1}J^{.4}$  Apparently the first part was measured in Refs. 1–4, while the slowly changing part played the role of the background. For a comparison of this theory with experiment, it would be interesting to measure the absolute value of the absorption coefficient.

We shall now discuss briefly the contribution to the absorption from the "slow" TLS with  $\Delta_0 \ll \Delta_{02}$  and from the "fast" with  $(\Delta_0 \gg \Delta_{02})$ . For rapid TLS,  $\lambda(E) \gg 1$  for all E; this means that all the relaxation processes with these TLS take place in the thermal layer. Outside the thermal layer (E > T), they are all in the ground state, since they rapidly discard the excitation, reaching the boundary layer region  $E \approx T$ . The energy  $\delta E$  which the fast TLS deliver to the electron system within a single period is the same in order of magnitude as the increase in the energy of the TLS within the time between two successive relaxation acts, i.e.,  $d\omega\tau$ . This quantity reaches its greatest value in the thermal layer at  $E \approx T$ .

$$\delta E \approx d\omega \tau (\Delta_0, T) \approx d\omega \tau_0 T^2 / \Delta_0^2 \approx T (\Delta_{02} / \Delta_0)^2.$$
(20)

The power absorbed per unit volume by the fast TLS is of the order of  $N_0 d\omega \delta E$  and correspondingly, the contribution from them to the absorption coefficient turns out to be  $(\Delta_0/\Delta_{02})^2$  times smaller than the value of the first term in Eq. (19).

The slow TLS can be divided into two groups: with  $\Delta_0 < \Delta_{01}$  and with  $\Delta_0 > \Delta_{02}$  (but  $\Delta_0 < \Delta_{02}$ ). The first group possesses such small tunnel transmissivity that it is excited in the thermal layer only with the small probability  $\lambda_{max} \ll 1$ . Becoming excited, it remains excited over the duration of several periods. The greatest probability  $\lambda_{max}$  of discarding the excitation is achieved at the instant of time close to  $t_0$  in which  $E \approx \Delta_0$ . However, the power absorbed by such TLS is determined by the very rare acts of de-excitation in the superthermal region (E > T) with the probability  $\lambda_{min} \ll 1$ , when a large energy of the order of d is transferred to the electron system. The power absorbed by such TLS per unit volume is determined by the expression

$$N_0 d\omega d\lambda_{min} \approx N_0 d\omega d \left(\Delta_0 / \Delta_{02}\right)^2. \tag{21}$$

The same result turns out to be valid also for slow TLS with  $\Delta_0 > \Delta_{01}$ . Although they relax rapidly in the thermal region, they give little energy to the electrons in this region. The absorption is determined by the relatively rare de-excitation processes, which take place over several periods in the superthermal layer with transfer of energy of the order of d. Thus, the contribution from the slow TLS to the absorbed power is determined by Eq. (21). The absorption coefficient here turns out to be  $(\Delta_{02}/\Delta_0)$  times smaller than the second term in Eq. (19).

We now see that the qualitative picture of the absorption in the low-frequency region is much more complicated than that in the high-frequency region. This is due, in final analysis, to the already mentioned protraction of the relaxation, the reason for which being that the relaxation time in metallic glasses increases with increase in the energy E (at a fixed tunnel transmissivity  $\Delta_0$ ).

We not discuss briefly, from the point of view of the theory developed in the present work, the results of the experiments of Ref. 4, where two steps were observed in the amplitude dependence of the ultrasonic absorption coefficient. The first step is connected with the nonlinear resonance absorption. At first glance, resonance absorption should not be observed in the investigated situation, since in resonance absorption those TLS with  $E = \hbar \omega$  are important, and the condition that is satisfied here is  $\omega \tau_0 \ll 1$ . But, as we have seen, the time  $\tau_0$  is the minimum relaxation time of the TLS with energies less than T. Therefore there are TLS with larger values of  $\tau$  (having a small transmissivity  $\Delta_0$ ). Moreover, as a consequence of the broad distribution of the TLS over  $\Delta_0$ , the concentration of such systems is of the same order (with logarithmic accuracy) as the concentration of systems with  $\tau \approx \tau_0$ . Therefore, there are always systems available to take part in the nonlinear resonance absorption. These are systems for which  $\omega \tau(\Delta_0, \hbar \omega) \gg 1$ , i.e.,  $\Delta_0$  $\langle \hbar \omega (\omega \tau_0)^{1/2} \langle \hbar \omega \rangle$ . In other words, the contribution to the resonance absorption is made by a strongly asymetric double-well potential in contrast with what happens in dielectric glasses, where, as a rule, the condition  $\omega \tau_0 \ge 1$  is realized.

In addition, a plateau is observed on the plot of  $\Gamma(J)$ , and after that, a new falloff. We shall assume that this falloff is due to the nonlinear relaxation absorption considered above. Unfortunately, there remains an open question as to how to interpret the results in accord with (19), since the absolute sound intensities were not given in Refs. 1–4 and the "background" part of the absorption was not measured. Moreover, Eq. (19) has an asymptotic character; it is valid at  $d \ge T$ ,  $d\omega \tau_0 \lt T$ . The first inequality here is satisfied without great margin.

The critical intensity  $J_{c2}$ , beginning with which a significant decrease in the relaxation absorption coefficient takes place, is determined from the condition  $d \approx T$ . We then have

$$J_{c2} \approx (T/\gamma)^2 \rho v^3. \tag{22}$$

It is thus proportional to the square of the temperature, which agrees with the results of experiment.<sup>1,3</sup> Moreover, it follows from (22) that, at a given intensity, the nonlinear effects should be more clearly expressed for transverse sound than for longitudinal, since in the latter the critical intensity is  $(v_l/v_t)^3$  times greater. This is in agreement with the results of Ref. 1.

We note that while the nonlinear relaxation absorption has been observed in metallic glasses, the corresponding nonlinearity has not been seen in dielectric glasses, so far as we know.<sup>1</sup> It is possible that the point is that the coefficients of proportionality in the amplitude-dependent parts of the absorption are different: in metallic glasses, at  $\omega \tau_0^{(m)} \ll 1$ , the coefficient is  $A\omega$ , where A is some constant, and in dielectrics at  $\omega \tau_0^{(d)} \ge 1$  in order of magnitude, it is  $A / \tau_0^{(d)}$ , where  $\tau_0^{(d)}$  is the analog of  $\tau_0$  in dielectric glass. Thus, the order of the ratio of the corresponding coefficients is  $\omega \tau_0^{(d)} \ge 1$ . To conclude this section, we shall make a few numerical estimates. Assuming  $\gamma \approx 2 \text{ eV}$ ,  $\rho \approx 10 \text{ g/cm}^2$  and  $v \approx 2 \times 10^5 \text{ cm/s}$ , we obtain  $d = (\gamma^2 J \rho v^3)^{1/2} \approx 3 \cdot 10^{-17} [\text{J}(\text{W/cm}^2)]^{1/2} \text{ erg.}$ . Thus, at  $J \approx 10 \text{ W/cm}^2$ , the quantity d can rach  $10^{-14}$  erg and at  $T \approx 0.1$  K, the ratio of  $d/T \approx 10$ . A typical value of  $\tau_0$  at  $T \approx 0.3$  K amounts to  $10^{-9}$  s in metallic glasses. Therefore, the quantity  $\omega \tau_0$  can, in experiment, be both less than and greater than unity.

It must be noted that Arnold, Doussineau, and Levelut, in their paper,<sup>17</sup> with the aim of explaining the experiments of Refs. 1–4, also took the point of view that the sound relaxation absorption in amorphous metals can fall off with increase in the intensity. From their theory, an estimate for  $J_{c2}$ is obtained that agrees with ours [Eq. (22)].

However, a number of results of our theory differ from the conclusions of Ref. 17. We shall consider the most important of these: 1) the dependence on the intensity of the parameter  $\omega \tau_0(T)d/T$ , which determines the boundary between the low- and the high-frequency nonlinear absorptions, and 2) the phenomenon, predicted by our theory, of protraction of the relaxation, which makes a contribution to the coefficient of nonlinear absorption that is inversely proportional to  $\ln[d/T\omega\tau_0(T)]$ .

The reason for this difference lies in the different character of the approximations used in our research and in Ref. 17. In our opinion, under the condition of satisfaction of the inequality (3), it is necessary, for the calculation of the nonequilibrium distribution function of the TLS, to use the adiabatic approximation, while the resonance approximation is applicable upon satisfaction of opposite inequalities. Since the basic contribution is made by TLS with  $E \approx T$  in the experiments of Refs. 1–4, the conditions (3) were satisfied.

It should be kept in mind that attempts to calculate the contribution of direct transitions with the absorption of a sound quantum in the interaction of the TLS with the sound wave under the conditions  $\omega \tau_{\min}(T) \leq 1$  run into serious difficulties, as is clear from the work of Maleev.<sup>18</sup> These are connected with the fact that those TLS with  $E < \hbar \tau_{\min}^{-1}(T)$  are important in the expression for the corresponding contribution to the absorption, and their interaction with the electrons cannot be described by perturbation theory.

# 3. NONLINEAR ABSORPTION. QUANTITATIVE CONSIDERATION

For quantitative analysis, it is convenient to introduce the following dimensionless variables:

$$\begin{split} \xi = \Delta/d, \quad \eta = \Delta_0/2T, \quad w = d/2T, \quad \varepsilon = E/2T, \quad x = \omega t, \\ \nu(x) = \nu_0 \text{ cth } \varepsilon(x)/\varepsilon(x), \quad \nu_0 = \eta^2/\omega\tau_0. \end{split}$$
(23)

In these variables,

$$\varepsilon(x) = [w^2(\xi + \cos x)^2 + \eta^2]^{\frac{1}{2}}, \qquad (24)$$

and the power absorbed per unit volume is

$$P = \frac{1}{\pi} N_{0} \omega T^{2} w \int_{0}^{\infty} d\xi \int_{0}^{\infty} \frac{d\eta}{\eta} q(\xi, \eta).$$
 (25)

Here  $(\omega T/2\pi)q(\xi,\eta)$  is the power absorbed by a single TLS with the parameters  $\xi$ ,  $\eta$ , i.e.,  $p(\Delta, \Delta_0)$ —see Eq. (7). The

expression for  $q(\xi, \eta)$  is obtained after substitution of the periodic solution of Eq. (4) in Eqs. (2). We shall use two equivalent expressions for q, obtained one after the other by integration by parts:

$$q = \int_{0}^{2\pi} dx \int_{0}^{2\pi} dy \frac{\varepsilon'(x)\varepsilon'(x-y)}{\operatorname{ch}^{2}\varepsilon(x-y)} \frac{\exp\left(-\int_{0}^{2} v(x-z) dz\right)}{1-\exp\left(-2\pi v\right)}, \quad (26)$$

or

$$q = 2\pi v_0 \left[ 1 - \frac{v_0}{2\pi (1 - \exp(-2\pi v))} \times \int_{0}^{2\pi} dx \int_{0}^{2\pi} dy \frac{\operatorname{cth} \varepsilon(x)}{\varepsilon(x-y)} \exp\left\{ -\int_{0}^{\pi} v(x-z) dz \right\} \right], (27)$$

where  $\varepsilon'(x) \equiv d\varepsilon/dx$  and the angle brackets denote averaging over the period.

It can be seen from (26) and (27) that the principal contribution is made by TLS for which  $\xi \leq 1$ ,  $\eta \leq 1$ ; we shall carry out all the estimates only for such systems. In the quantity

$$\langle 2\pi\nu\rangle = v_0 \int_{0}^{2\pi} dx \frac{\operatorname{cth} \varepsilon(x)}{\varepsilon(x)}$$

at  $\xi < 1$  the principal contribution is made by the neighborhood of the points  $\pm x_0 = \pm \arccos(-\xi)$ . The estimate of this contribution is

$$\langle 2\pi\nu\rangle \approx \frac{\nu_0}{\eta w |\sin x_0|} \left(1 + \eta \ln \frac{w}{\eta}\right).$$
 (28)

It is convenient to represent this expression in the form

$$\langle 2\pi\nu\rangle \approx \frac{\eta}{\eta_1 |\sin x_0|} \left( 1 + \eta \ln \frac{w}{\eta} \right),$$
 (29)

where

$$\eta_{i} \equiv w_{\omega} \tau_{0} \approx \Delta_{0i} / T. \tag{30}$$

It is seen that at  $w\omega\tau_0 \ge 1$ , (the high-frequency case) for all characteristic  $\eta$  we can assume that  $(2\pi\nu) \le 1$ . Using (27), we then obtain the following equation for the power absorbed per unit volume:

$$P = c_1 (N_0 dT / \tau_0), \tag{31}$$

where

$$c_{i} = \int_{0}^{\infty} d\xi \int_{0}^{\infty} d\eta \eta \left[ 1 - \frac{\langle \varepsilon^{-i} \rangle \langle \operatorname{cth} \varepsilon \rangle}{\langle \varepsilon^{-i} \operatorname{cth} \varepsilon \rangle} \right].$$

The coefficient  $c_1 \approx 1$  and can be determined by numerical integration. Equation (31) has a clear physical meaning. As we have already noted in Sec. 2, in the case  $d\omega\tau_0 \gg T$ , the processes of relaxation of the distribution function f rarely takes place: no more than once during several periods (the number of which is of the order of  $d\omega\tau_0/T$ ). This menas that the distribution function f differs little from its mean value  $\langle f \rangle$  over the period, which can be determined by averaging the kinetic equation (4) over the period:

$$\langle f \rangle \langle \tau^{-1} \rangle = \langle f_0 / \tau \rangle. \tag{32}$$

The power absorbed by a single TLS,  $p(\Delta, \Delta_0)$ , can be determined from (2) (in this expression, it is convenient first to

carry out integration by parts). With account of the explicit forms of  $f_0$  and  $\tau$  [(4),(5)], we have

$$p = \left\langle E \frac{f - f_0}{\tau} \right\rangle = \frac{T}{\tau_0} \left( \frac{\Delta_0}{2T} \right)^2 \left[ 1 - \frac{\langle E^{-1} \rangle \langle \operatorname{cth} (E/2T) \rangle}{\langle E^{-1} \operatorname{cth} (E/2T) \rangle} \right].$$
(33)

This result, after substitution in (7), leads to (31). The estimate (18) for the absorption coefficient follows directly from (31).

Analysis of the case  $d\omega \tau_0 \ll T$  is carried out conveniently on the basis of Eq. (27). Making use of the identity

$$1 - e^{\langle -2\pi \varkappa \rangle} = v_0 \int_0^{2\pi} \frac{dy}{\varepsilon (x-y)} \exp\left\{-\int_0^y \varkappa (x-z) dz\right\}, \qquad (34)$$

where  $\kappa(x) \equiv v_0/\varepsilon(x)$  are functions identical with  $\nu(x)$  outside the region where  $\varepsilon \leq 1$ , we can reduce (27) to the form

$$q(\xi, \eta) = q_1 + q_2 + q_3, \tag{35}$$

$$q_1 = 2\pi v_0 \exp \langle -2\pi \varkappa \rangle, \qquad (36a)$$

$$q_2 = -2\pi\nu_0 \frac{1 - \exp\langle -2\pi\nu\rangle}{1 - \exp\langle -2\pi\nu\rangle} \exp\langle -2\pi\nu\rangle, \qquad (36b)$$

$$q_{3} = 2\pi\nu_{0} (1 - \exp\langle -2\pi\nu\rangle)^{-1} \left[ \langle \operatorname{cth} \varepsilon -1 \rangle (\exp\langle -2\pi\varkappa\rangle -1) + \frac{\nu_{0}}{2\pi} \int_{0}^{2\pi} dx \operatorname{cth} \varepsilon(x) \int_{0}^{2\pi} dy \frac{1}{\varepsilon(x-y)} \left\{ \exp\left(-\int_{0}^{y} \varkappa(x-z) dz\right) - \exp\left(-\int_{0}^{y} \nu(x-z) dz\right) \right\} \right].$$
(36c)

To avoid any misunderstanding, we emphasize that the representation of the quantity q in the form of a sum of three terms has no relation to the corresponding contributions from fast, intermediate and slow TLS.

The estimated order of magnitude of  $\langle 2\pi \varkappa \rangle$  is  $\eta^2/w\omega \tau_0$ [see (38)]. Therefore at

 $\eta > \eta_2 \equiv (w \omega \tau_0)^{\frac{1}{2}} \approx \Delta_{02}/T$ 

the quantity  $q_1$  falls off rapidly with increase in  $\eta$ . This term corresponds to the principal part of the contribution from the intermediate TLS. As a result, we obtain the following contribution to the power absorbed per unit volume:

$$P^{(1)} = \frac{2N_0 \omega T^2 w}{\omega \tau_0} \int_0^1 d\xi \int_0^\infty d\eta \eta \exp(-2\pi \varkappa).$$
(37)

Here we have the estimate

$$\langle 2\pi\kappa \rangle \approx \frac{2\nu_0}{w|\sin x_0|} \ln \frac{w|\sin x_0|}{\eta}$$
(38)

(the factor 2 is connected with the presence of two points of minimum  $\varepsilon$ ). Therefore,

$$P^{(1)} \approx N_0 \omega T^2 w^2 \int_0^{\pi/2} \sin^2 \varkappa_0 dx_0 \int_0^{\infty} dz \exp\left\{-z \ln \frac{w |\sin x_0|}{z \omega \tau_0}\right\}.$$
 (39)

The asymptote of the integral over z at  $\ln(w/\omega\tau_0) \ge 1$  is

$$\ln^{-1}\left(\frac{w|\sin x_0|}{\omega\tau_0}\right).$$

Since the logarithm depends weakly on its argument, a good

approximation is the expression

$$P^{(1)} \approx \frac{\pi}{4} N_0 \omega T^2 w^2 \ln^{-1} \left(\frac{w}{\omega \tau_0}\right).$$
(40)

It leads results in the second term in Eq. (19). We note that the corresponding contribution is determined by the entire period of the wave and not by the small vicinities of the instants  $t_0$ .

The term  $q_2$  is negative and falls off rapidly with increase in  $\eta$  at  $\eta > \eta_1$ . Taking it into account that  $\eta_1 \ll \eta_2$  in the low-frequency case, and using (38), we arrive at the following estimate for the contribution of this term:

$$P^{(2)} \approx -N_0 \omega T^2 \frac{w}{\omega \tau_0} \int_0^{\pi} d\eta \eta \left(\frac{\eta}{\eta_2}\right)^2 \ln\left(\frac{w}{\eta}\right)$$
$$\approx -N_0 \omega T^2 w^2 (w_0 \tau_0)^2 \ln\left(\frac{1}{\omega \tau_0}\right). \tag{41}$$

Numerically, this contribution is obviously very small in comparison with the contribution of (40) and the contribution from  $q_3$ , which we shall consider below, since we have actually required the statisfaction of the condition  $\eta_2 = (w\omega\tau_0)^{1/2} \ll 1$ , and the fourth power of a small quantity enters into the estimate (41). Therefore, we have neglected this term in Eq. (19).

The value of the term  $q_3$  in (35) is determined by the neighborhoods of the points  $x_0$ . At  $\eta_1 \ll \eta_2$  this term is small in comparison with the considered quantity relative to the parameter  $w^{-1}(\eta/\eta_2)^2$  (the factor  $w^{-1}$  is connected with the fact that the integration in this expression is taken over the neighborhoods of the points x of width  $w^{-1}$ ). Therefore, the basic contribution is made by the region  $\eta \gtrsim \eta_2$  and at the same time both  $q_1$  and  $q_2$  are determined by the region  $\eta \le \eta_2$ . For the determination of the dependence of the contribution of  $q_3$  on the amplitude d, it is convenient to make use of Eq. (26) for the total absorbed power. Setting

$$\int_{0}^{\infty} v(x-z) dz = yv(x), \quad \exp\langle -2\pi v \rangle = 0$$

in this expression, and cutting off the integration at the lower limit  $\eta_2$  we obtain Eq. (19) of Sec. 2.

# 4. KINETIC EQUATION AND THE DERIVATION OF THE EXPRESSION FOR THE ABSORPTION COEFFICIENT

We shall write the Hamiltonian of the TLS interacting with electrons and with the deformation field in the sound wave in the form

$$H = \frac{1}{2} \Delta \sigma_{\mathfrak{s}} - \frac{1}{2} \Delta_{\mathfrak{o}} \sigma_{\mathfrak{s}} + \gamma_{ik} u_{ik} \sigma_{\mathfrak{s}} + \frac{1}{2} \sum_{\alpha\beta} V_{\alpha\beta} \sigma_{\mathfrak{s}} c_{\alpha}^{+} c_{\beta}, \qquad (42)$$

where  $u_{ik} = u_{ik}^0 \cos \omega t$  is the deformation tensor in the sound field at the location of the TLS,  $u_{ik}^0$  is the amplitude of the deformation,  $\omega$  is the frequency of the sound wave,  $V_{\alpha\beta}$  is the diagonal matrix element describing the change of  $\Delta$  in the transition in the electron subsystem, and  $c_{\alpha}^+(c_{\alpha})$  are the creation (annihilation) operators of the electron in the state  $\alpha$ . Further, we shall not assume that the electron states are plane waves, keeping it in mind that such an assumption is not strictly valid in the amorphous metal. Assuming the validity of the adiabatic approximation, the condition of which (3) was given above, we make a canonical transformation, diagonalizing the three terms in (42). As a result, we find

$$\widehat{H} = \frac{1}{2} E \sigma_{3} + \sum_{\alpha\beta} W_{\alpha\beta} \sigma_{1} c_{\alpha} + c_{\beta} + \sum_{\alpha\beta} W_{\alpha\beta} \sigma_{3} c_{\alpha} + c_{\beta}, \qquad (43)$$

where

$$W_{\alpha\beta}^{\perp} = \frac{1}{2} \frac{\Delta_0}{E} V_{\alpha\beta}, \quad W_{\alpha\beta}^{\parallel} = \frac{1}{2} \frac{\Delta}{E} V_{\alpha\beta}.$$
(44)

The quantity E depends on the time and is determined by Eq. (1).

We have the following set of equations for the population density  $f^s$  of the upper level of the sth TLS and for the distribution function  $n_{\alpha}$  of the electrons over the states  $\alpha$ :

$$\frac{\partial f^{s}}{\partial t} = \frac{2\pi}{\hbar} \sum_{\alpha\beta} |W_{\alpha\beta}^{\perp s}|^{2} [-f^{s} n_{\alpha} (1-n_{\beta}) + (1-f^{s}) n_{\beta} (1-n_{\alpha})] \\ \times \delta(E_{s} + \varepsilon_{\alpha} - \varepsilon_{\beta}); \qquad (45)$$

$$\frac{\partial n_{\alpha}}{\partial t} = \frac{2\pi}{\hbar} \sum_{s} \sum_{\beta} |W_{\alpha\beta}^{\perp s}|^{2} \{ [-n_{\alpha}(1-n_{\beta})f^{s} + n_{\beta}(1-n_{\alpha})(1-f^{s})] \delta(E_{s} + \varepsilon_{\alpha} - \varepsilon_{\epsilon}) + [-n_{\alpha}(1-n_{\beta})(1-f^{s}) + n_{\beta}(1-n_{\alpha})f^{s}] \delta(E_{s} + \varepsilon_{\beta} - \varepsilon_{\alpha}) \} + I^{e-e} \{n_{\alpha}\} \equiv I^{e-TLS} + I^{e-e}.$$
(46)

Here  $\varepsilon_{\alpha}$  is the energy of the electron in the state  $\alpha$  and  $T^{e^{-e}}$  is the integral of the electron-electron collisions.<sup>5)</sup> We shall assume that the electron-electron collisions take place much more frequently than the collisions of electrons with the TLS. In this case, the electron distribution function  $n_{\alpha}$  can, with sufficient accuracy, be regarded as equal to the equilibrium Fermi function  $n_{\alpha}^{0}$ . More precisely,

$$n_{\alpha} = n_{\alpha}^{0} + \delta n_{\alpha}, \tag{47}$$

where the nonequilibrium part  $\delta n$  is small in comparison with the equilibrium part in the parameter  $I^{e^{-TLS}}/I^{e^{-e}} \ll 1$ .

Taking into account what has been said above, we can substitute  $n_{\alpha}$  in the kinetic equations (45) and (46) in the collision integrals of electrons with TLS in place of  $n_{\alpha}$ , while the integral of the electron-electron collisions  $I^{e-e}$  is linearized with respect to  $\delta n_{\alpha}$ . As a result Eq. (45) transforms into (40), while the relaxation time  $\tau$  is determined by the relation

$$\frac{1}{\tau(E)} = \frac{2\pi}{\hbar} \sum_{\alpha\beta} |W_{\alpha\beta}^{\perp}|^2 [n_{\alpha}^{0}(1-n_{\beta}^{0}) + n_{\beta}^{0}(1-n_{\alpha}^{0})] \delta(E+\varepsilon_{\alpha}-\varepsilon_{\beta}).$$
(48)

We shall carry out the summation over  $\alpha$  and  $\beta$  in (38) in two stages. First, we sum over the states  $\alpha$  and  $\beta$  that correspond to certain energies  $\varepsilon_{\alpha} = \varepsilon$  and  $\varepsilon_{\beta} = \varepsilon'$ , and then integrate over  $\varepsilon$  and  $\varepsilon'$  from zero to infinity. As a result, we obtain the following expression, with account of the  $\delta$  function

$$\frac{1}{\tau(E)} = \frac{\pi}{4\hbar} \left(\frac{\Delta_0}{E}\right)^2 (1 + e^{E/T}) \int_0^\infty d\varepsilon e^{(\varepsilon - \mu)/T} (1 + e^{(\varepsilon - \mu)/T})^{-1} \times (1 + e^{(E + \varepsilon - \mu)/T})^{-1} \chi(\varepsilon, \varepsilon + E),$$
(49)

where

$$\chi(\varepsilon,\varepsilon') = 2 \sum_{\alpha\beta} |V_{\alpha\beta}|^2 \delta(\varepsilon - \varepsilon_{\alpha}) \delta(\varepsilon' - \varepsilon_{\beta}).$$
 (50)

It is natural to assume the function  $\chi(\varepsilon,\varepsilon')$  which characterizes the interaction of the electrons with the TLS as slowly varying function of  $\varepsilon$ ,  $\varepsilon'$  at  $\varepsilon \sim \varepsilon' \sim \mu$  in the scale of energies T,  $E \ll \mu$ . This allows us simply to replace it in the integral (49) by the constant  $\chi \equiv \chi(\mu,\mu)$ . Calculating the integral (49), we arrive at the formula (5).

We can calculate the absorbed power if we take it into account that in final analysis it is transferred to the electron subsystem.<sup>6)</sup> The rate of increase of the energy of the electron subsystem is equal to

$$\dot{Q} = \sum_{\alpha} \varepsilon_{\alpha} \frac{\partial n_{\alpha}}{\partial t} = \sum_{\alpha} \varepsilon_{\alpha} \frac{\partial}{\partial t} \delta n_{\alpha}.$$
 (51)

The, with account of (46) and after uncomplicated transformations, we obtain

$$\dot{Q} = \sum_{\bullet} E_{\bullet} (f^{\bullet} - f_{0}^{\bullet}) / \tau(E_{\bullet}).$$
(52)

With account of (4) and subsequent averaging over the period of the sound wave, we come to the conclusion that this formula is equivalent to (2).

In order to obtain (7) from (2), it is necessary to find the solution of Eq. (4), where  $\tau$  and  $f_0$  depend on the time through the dependence E(t), which is determined by (1). Substitution of the periodic solution (4) in Eq. (2) leads to the expressions (26) and (27).

We shall now discuss the limits of applicability of the perturbation theory, on the use of which the calculation of the collision operator of the kinetic equation is based. In order that perturbation theory be applicable, satisfaction of the condition

$$\hbar/E\tau_{min}(E) \ll 1 \tag{53}$$

is necessary in every case.

In the study of nonlinear relaxation absorption, we have been interested in an energy E of the order of or greater than T. In this region, the quantity on the left side of the inequality is a constant of the operator of  $\chi$ , the experimental value of which varies for different metallic glasses in the range from several hundredths to several tenths of degrees. However, the relaxation time of the TLS on phonons, which depends on E as<sup>15</sup>

$$\frac{1}{\tau^{ph}_{min}} \propto E^{3} \operatorname{cth} \frac{E}{2T}$$
 (54)

increases rapidly with increase in E. Therefore, at some value  $E = E_1$ , the relaxation time of the TLS on phonons becomes comparable with the relaxation time on electrons. The value of E is determined by the estimate

$$E_1 \approx E_c \chi^{1/2}, \quad E_c \approx \gamma^{-1} (\rho \hbar^3 v^5)^{1/2}$$

where  ${}^9E_c \approx 10-20$  K. Numerically, the value of  $E_1$  amounts to several degrees. Since we have not taken into account the relaxation of the TLS on phonons, the obtained results are valid at  $d \ll E_1$ . Upon violation of this inequality, it is generally necessary to consider (along with the electron mechanism) the phonon mechanism of absorption.

#### 5. CONCLUSION

The results indicate that the nonlinear relaxation absorption of sound (especially in the case of low frequencies) is very sensitive to the energy dependence of the relaxation of the TLS. Therefore, it seems to us that the experimental value of the nonlinear absorption should allow us to obtain a whole series of data on the mechanism of TLS relaxation. An especially interesting picture should be observed in superconducting glasses, where it is easy to create conditions in which the various relaxation processes compete with one another. Actually, the number of normal excitations in the superconducting state decreases exponentially with the temperature. The same is true for the reciprocal of the relaxation time of those TLS for which  $E < 2\Delta_0$ ,  $(2\Delta_s)$  is the width of the superconducting gap). The relaxation time of the TLS with  $E > 2\Delta_s$  is much less and depends very weakly on the temperature, since such TLS can relax, separating the Cooper pair. Therefore, at  $T < 2\Delta_s$ , a peculiar situation is possible, in which the TLS, excited in the vicinity of  $t_0$  remains in an excited state until the time at which E becomes equal to the width of the superconducting gap, after which de-excitation takes place very rapidly.

A second interesting aspect of the nonlinear absorption in superconducting glasses is that at sufficiently low temperatures, the relaxation time of the TLS on the quasiparticles becomes equal to the corresponding relaxation time on the phonons. Since these two times depend differently on E, a rather variegated picture arises of the behavior of the nonlinear absorption, the study of which by experiment would be, it would seem, very interesting. However, a detailed consideration of the nonlinear absorption in amorphous superconductors goes beyond the range of the present research.

- <sup>1)</sup>It should be kept in mind that the very possibility of introducing the concept of a time-dependent level spacing E is a consequence of the adiabatic approximation.
- <sup>2)</sup>This is the quantity that Black<sup>7</sup> denotes by  $\tilde{\rho}_0$ .
- <sup>3)</sup>This corresponds to an assumption that the distribution functions of the random quantities  $\Delta$  and  $\ln \Delta_0$  are constant.<sup>7</sup>
- <sup>4</sup>)It was this contribution which was not taken into account by us in Ref. 5. <sup>5</sup>)Generally speaking, in the writing down and in the analysis of this integral of the collisions, we must bear it in mind that in dirty metals, as Schmidt<sup>9</sup> and Al'tshuler, Aronov<sup>20</sup> have shown, the expression for the operator of the inelastic interelectron collisions must be modified in comparison with those for pure metals.
- <sup>6)</sup>This derivation is easily generalized to the case in which we must take the electron-phonon collisions into consideration. In that case, it would be necessary to consider also the transfer of energy to the phonons while the expression (2) would remain unchanged.

<sup>3</sup>G. Park, A. Hikata and C. Elbaum, J. Non-Cryst. Sol. 45, 93 (1981).

<sup>&</sup>lt;sup>1</sup>H. Araki, G. Park, A. Hikata and C. Elbaum, Phys. Rev. **21B**, 4470 (1980).

<sup>&</sup>lt;sup>2</sup>G. Park, A. Hikata and C. Elbaum, Phys. Rev. 24B, 7389 (1981).

<sup>&</sup>lt;sup>4</sup>A. Hikata, G. Cibuzar and C. Elbaum, J. Low Temp. Phys. **49**, 399 (1982).

<sup>&</sup>lt;sup>5</sup>Yu. M. Gal'perin, V. L. Gurevich and D. A. Parshin, Pis'ma Zh. Eksp. Teor Fiz. **36**, 386 (1982) [JETP Lett. **36**, 466 (1982)].

<sup>&</sup>lt;sup>6</sup>J. Jackie, Z. Phys. 257, 212 (1972).

<sup>&</sup>lt;sup>7</sup>J. L. Black, in Metallic Glasses (H. J. Güntherodt, ed.) Springer-Verlag, New York, 1981, p. 167.

<sup>&</sup>lt;sup>8</sup>Yu. M. Gal'perin, Zh. Eksp. Teor. Fiz. **84**, 1386 (1983) (Translator's note: there is no article in volume 84 by Gal'perin by himself. This is one

by Yu. M. Gal'perin, A. A. Dobrovolskii, V. I. Kozub and A. B.Sherman, 84, 1391 (1983) [Sov. Phys. JETP 57, 809 (1983)].

<sup>9</sup>V. L. Gurevich and D. A. Parshin, Zh. Eksp. Teor. Fiz. **83**, 1301 (1982) [Sov. Phys. JETP **56**, 1334 (1982)].

- <sup>10</sup>P. Doussineau and A. Robin, in Phonon Scattering in Condensed Matter (H. J. Maris, ed.) Plenum Press, New York, London, 1980, p. 165.
- <sup>11</sup>B. Golding, J. E. Graebner, A. B. Kane and J. L. Black, Phys. Rev. Lett. 41, 1487 (1978).
- <sup>12</sup>W. Arnold, P. Doussineau, Ch. Frenois and A. J. Levelut, Phys. Lett. **42**, L-289 (1981).
- <sup>13</sup>W. Arnold, A. Billmann, P. Doussineau and A. J. Levelut, Physica 110B, 2036 (1982).
- <sup>14</sup>J. L. Black and P. Fulde, Phys. Rev. Lett. 43, 453 (1979).

- <sup>15</sup>S. Hunklinger and W. Arnold, in Physical Acoustics (W. P. Mason and R.N. Thurston, eds.) Academic Press, New York, 1976, Vol. 12, p. 155.
- <sup>16</sup>G. Weiss, W. Arnold, K. Dransfeld and H. J. Güntherodt, Sol. St. Commun. 33, 111 (1980).
- <sup>17</sup>W. Arnold, P. Doussineau and A. J. Levelut, Phys. Lett. 43, L-695 (1982).
- <sup>18</sup>S. V. Maleev, Zh. Eksp. Teor. Fiz. 84, 260 (1983) [Sov. Phys. JETP 56, 482 (1979)]; Sol. St. Commun. 38, 11 (1981).
- <sup>19</sup>A. Schmid, Z. Phys. 271, 251 (1974).
- <sup>20</sup>B. L. Al'tshuler and A. G. Aronov, Pis'ma Zh. Eksp. Teor. Fiz. **30**, 514 (1979) [JETP Lett. **30**, 482 (1979)]; Sol. St. Commun. **38**, 11 (1981).

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