

Nonlinear resonant absorption in glasses and spectral diffusion. Absorption by small particles

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A theory is developed of nonlinear resonant absorption of an RF electric field by two-level systems (TLS) in powdered glass. It is assumed that the RF-field frequency satisfies the condition $\hbar\omega \ll kT$. It is shown that in this case the spectral diffusion mechanism plays a significant role in the formation of the nonlinear-absorption spectrum. The presence of two TLS in a powder particle is considered, one resonant (level spacing $e = \hbar\omega$) and the other "thermal" (level spacing $E \approx kT$). Phonon-induced transitions in the thermal TLS produce random changes of the level spacing in the resonant TLS, which alternately goes off and into resonance. A diagram technique is used to describe the linear absorption in such a situation and leads to results that agree with a model in which the transitions of the thermal TLS from one state to another are instantaneous (jumpwise). The problem of nonlinear resonant absorption is solved in a model of thermal-TLS jump transitions described by the telegraph process. The solution is by the method of stochastic differential equations. It is shown that the plot of the absorption vs the intensity in powdered glass has two steps. The first is due to the saturation of the resonant TLS in particles that have no thermal TLS, or in particles where the frequency of the thermal-TLS transitions is low. The second step is due to saturation of those TLS which have a fast enough thermal neighbor. It is noted that the results can be used directly to describe nonlinear absorption by a system of paramagnetic noncentral ions and the NMR of molecules having a chemical shift.

1. INTRODUCTION

Our purpose is to consider nonlinear resonant absorption of RF electromagnetic radiation by two-level systems (TLS) in dielectric glasses. A dependence of this absorption on the intensity was observed in Refs. 1–3 (see also Ref. 4). This dependence can be interpreted by positing saturation of the TLS by an intense wave.^{5,6}

The RF radiation frequency is assumed to satisfy the condition ($\hbar = 1$)

$$\omega \ll T, \quad (1.1)$$

where T is the temperature (in energy units). The resonant absorption is due to those TLS in which the level spacing e satisfies with sufficient accuracy the condition

$$\omega = e. \quad (1.2)$$

The electromagnetic-energy absorption by a resonant TLS is determined by the off-diagonal component of its density matrix. This component is usually found by solving the Bloch equations.^{5,6} These equations contain two characteristic times. These are the time τ_1 that describes the population relaxation of the resonant width of the resonant-TLS and the time τ_2 that characterizes the resonant-TLS absorption linewidth in the linear approximation.

The time τ_1 is usually determined by the interaction of the resonant TLS with phonons. As for the time τ_2 , at sufficiently high frequencies $\omega \gtrsim T$ it is also determined by the interaction with the phonons. In this (simplest) case $\tau_2 = 2\tau_1$.¹⁾

The frequencies dealt with in contemporary experiments, however, are shorter, so that this condition is not satisfied and $\tau_2 \ll \tau_1$. In this case, as indicated by Hunklinger

and Arnold^{6,7} and by Joffrin and Levelut,⁸ the time τ_2 is determined by the interaction of the resonant TLS with the neighboring thermal levels, i.e., with TLS having E of the order of T .

The point is that each TLS is surrounded by a strain field. This field depends on the state—upper or lower—of the given TLS and decreases in inverse proportion to the cube of the distance from the system.^{6,8}

On the other hand, the distance e between the levels of a resonant TLS depends on the strain u_{ij} at its location. Thus, the neighboring-TLS population fluctuations due to interaction with thermal phonons cause the TLS in question to alternate between off-resonance and resonance. This decreases the time during which the given TLS is resonant. The variation of the level spacing of the resonant TLS is random. This phenomenon was designated spectral diffusion. Although this designation is not quite appropriate, we retain it in this article.

We shall consider a simplified approach to the role of spectral diffusion in nonlinear absorption. We shall assume in fact that adjacent to the resonant TLS there is only one thermal TLS capable of effecting transitions. Such a problem can have a direct bearing on a glass powder, in which the individual particles are interspersed in a crystal matrix. On the one hand, this problem admits of exact solution, and on the other it can track the basic physics of spectral diffusion in glasses.

In addition, the results of Sec. 3 can be directly used to describe the saturation of the paramagnetic resonance of the so-called noncentral ions, which can occupy several equivalent positions in a unit cell.^{9,10} Transitions between these

positions are either by tunneling or by surmounting the barrier, with phonon participation. Owing to the anisotropy of the g factor, however, the value the spectroscopic splitting depends on the location of the noncentral ion. A similar situation is realized in NMR spectra of molecules subject to the so-called chemical shift.

Getting ahead of ourselves, we note that our analysis leads to the following important conclusion. It can be seen from the concrete form of the expression for the nonlinear-absorption line shape of a two-resonant TLS [Eq. (3.11)], this form is not described by a solution of the Bloch equations and by some constant value of the time τ_2 , since these equations can yield only a Lorentz absorption line. Only in the limiting case (3.17) of frequent transitions in the thermal TLS is the line shape Lorentzian and can be described by introducing a suitable time τ_2 .

The plan of the article is the following. We derive first, using a diagram technique, an expression for the linear absorption coefficient. We compare the results of this derivation with the earlier results based on the concept of quantum jumps. We verify that, at the accuracy of interest to us (and determined by the ratio of the absorption linewidth to its frequency ω) both methods lead in the linear theory to equivalent results. By the same token, the diagram-based derivation can thus serve as confirmation of the quantum-jump concept.

The jump method is used next to solve the main problem of interest, the nonlinear one.

2. FORMULATION OF PROBLEM AND CALCULATION OF LINEAR ABSORPTION

We write the Hamiltonian of the system considered in the form

$$\mathcal{H} = \frac{1}{2}eS_3 + \frac{1}{2}ES_3 + \frac{1}{2}JS_3S_3 + m_{ii}\hat{u}_{ii}S_1 + M_{ii}\hat{u}_{ii}S_1 + \mathcal{H}_{ph} + F \cos \omega t s_1. \quad (2.1)$$

Here e is the distance between the resonant TLS levels, E is the distance between the thermal TLS level, s_i and S_i are Pauli matrices acting respectively in the spaces of the resonant and thermal TLS, and J is the energy of their interaction. This is the interaction referred to in the Introduction. The corresponding expression for the Interaction Hamiltonian were obtained by Aminov and Kochelaev,¹² Joffrin and Levelut,⁸ and by Black and Halperin (Ref. 13).²⁾ The energy J is inversely proportional to r^3 , where r is the distance between the interacting TLS. We assume next that

$$|J| \ll \omega \ll T. \quad (2.2)$$

Only in this case can allowance for the interaction between the resonant and thermal TLS be reconciled with the resonance approximation [see Eq. (2.9) below] in which the entire calculation that follows is carried out. The next two terms of the Hamiltonian (2.1) describe respectively the phonon interactions of the resonant and thermal TLS, which cause the transition in these systems; m_{ii} and M_{ii} are the corresponding tensors of the strain potential, and \mathcal{H}_{ph} is the Hamiltonian of the phonon system. Finally, the last term describes the interaction between the resonant TLS and the alternating field of frequency ω .

We shall regard the first three terms of (2.1) as the unperturbed Hamiltonian which has the four eigenvalues $h_1, h_2, h_3,$ and h_4 , where

$$\begin{aligned} 2h_1 &= e + E + J, & 2h_2 &= e - E - J, \\ 2h_3 &= -e + E - J, & 2h_4 &= -e - E + J \end{aligned} \quad (2.3)$$

(a similar approach was used by Gauthier and Walker⁹).

The scheme of these levels is shown in Fig. 1. For simplicity, we shall disregard hereafter the interaction of the resonant TLS with the phonons, and confine ourselves only to allowance for such an interaction for the thermal TLS (we shall indicate below how the results change when this interaction is taken into account). The transitions that can be effected in this case by the phonons are shown in the figure by dashed lines. Transitions induced by the resonant field are shown by wavy lines.

The interaction between the resultant four-level system and the phonons will be described by perturbation theory. We shall sum the series that appear by the diagram technique proposed by Abrikosov¹⁴ for a spin interacting with electrons.³⁾ This technique was used by Maleev¹⁶ to describe interactions of two-level systems with phonons. It can be verified that the non-uniform level spacing does not prevent the use of such a technique to describe also the interaction of our four-level system with phonons.

According to this technique, a four-level can be set in correspondence with a system of four fermions, to which correspond the renormalized propagators

$$g_i(\varepsilon) = (\varepsilon - h_i + i\Gamma_i/2)^{-1}.$$

Here ε is the energy variable, $\Gamma_i/2$ is the imaginary part of the self-energy and is due to the interaction of the quasifermions with the phonons (the real part of the self-energy is regarded as included in h_i via renormalization of the bare energies).

For Γ_i we obtain, in full analogy with Ref. 16,

$$\frac{\Gamma_i}{2} = -\frac{1}{\pi} \text{Im} \int_{-\infty}^{+\infty} dx \Phi(x) g_i(\varepsilon + x) \Big|_{\varepsilon=h_i} = \Phi(h_i - h_i). \quad (2.4)$$

The subscripts i and \bar{i} correspond to states coupled by a phonon transition: $\bar{1} = 2, \bar{3} = 4$ and the converse. Here $\Phi(x)$ is the imaginary part of the phonon propagator and is equal to

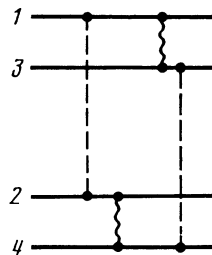


FIG. 1. Level scheme for a dipole-dipole interacting pair comprising a resonant TLS and a thermal TLS [see (2.3)]. The dashed lines show transitions, with phonon participation, in the thermal TLS, while the wavy lines show transitions induced in the resonant TLS by the RF field.

$$\Phi(x) = \frac{1}{4\pi\rho} x^3 N(x) \left(\frac{M_l^2}{v_l^5} + 2 \frac{M_t^2}{v_t^5} \right), \quad (2.5)$$

$$N(x) = [\exp(x/T) - 1]^{-1}.$$

Here N is the Planck function, ρ the density of the glass, M_l and M_t the longitudinal and transverse constants of the TLS strain potential^{17,18}:

$$M_l^2 = \frac{1}{15} [(Sp M)^2 + 2 Sp M^2],$$

$$M_t^2 = \frac{1}{30} [3 Sp M^2 - (Sp M)^2],$$

and v_l (v_t) the corresponding sound velocities.

Proceeding in the spirit of Refs. 16, we obtain the following expression for the retarded susceptibility $\chi(\omega)$, whose imaginary part describes the absorption in the linear theory:

$$\chi(\omega) = \frac{1}{\mathcal{N}} \int_{-\infty}^{+\infty} \frac{dx}{2\pi i} e^{-x/T} \sum_{i,t} \{g_i(x+\omega) \mathcal{T}_{it}(x+\omega, x) g_i(x) - g_i(x) \mathcal{T}_{it}(x, x-\omega) g_i(x-\omega) + g_i(x) \mathcal{T}_{it}(x, x-\omega) g_i(x-\omega)\}, \quad (2.6)$$

where \mathcal{T}_{it} are the vertex parts describing the interaction of the quasifermions with the resonant field F . As shown in Fig. 1, only the vertices \mathcal{T}_{it} for the transitions $1 \leftrightarrow 3$ and $2 \leftrightarrow 4$ differ from zero. The bare vertices for these transitions are assumed equal to unity. It suffices to calculate the normalization factor \mathcal{N} for this case in the zeroth approximation in the phonons. It is equal to

$$\mathcal{N} = Sp \exp(-\hat{\mathcal{H}}_0(T)) = \sum_i \exp(-h_i/T), \quad (2.7)$$

where $\hat{\mathcal{H}}_0$ is that part of the Hamiltonian (2.1) which does not contain interactions with the resonant field and with the phonons. Taking the inequalities (1.1) and (2.2) into account we have

$$\mathcal{N} = 4 \operatorname{ch}(E/2T). \quad (2.8)$$

It is easy to verify that in the resonant approximation

$$|\omega - e| \ll \omega, \quad (2.9)$$

to which we confine ourselves, only the second and fourth terms of (2.6) are significant.

Being interested in the absorption line shape, we consider only the imaginary part of the susceptibility. It can be represented, taking the inequalities (1.1) and (1.2) into account, in the form (the so-called pole approximation)

$$\operatorname{Im} \chi(\omega) = -\frac{\omega}{T} \sum_{i,k} P_k \operatorname{Im} \Pi_{ik}(\omega), \quad (2.10)$$

where

$$P_k = \mathcal{N}^{-1} e^{-h_k/T}, \quad (2.11)$$

$$\Pi_{ik}(\omega) = \int_{-\infty}^{+\infty} \frac{dx}{2\pi i} g_i(x+\omega) \mathcal{T}_{ik}(x+\omega, x) g_k(x). \quad (2.12)$$

The ladder approximation is sufficient for the calculation of the vertices \mathcal{T}_{ik} (cf. Ref. 16). The small parameter that en-

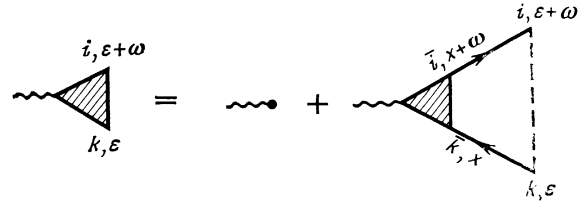


FIG. 2. Equation for the vertex $\mathcal{T}_{ik}(\epsilon + \omega, \epsilon)$ that describes the interaction of the four-level system (2.3) with the resonant RF field. $(i, k) = (1, 3)$ or $(2, 4)$; $\bar{1} = 2, \bar{3} = 4$. The imaginary part of the phonon propagator $\Phi(x)$ is shown dashed.

sures validity of this approximation is the ratio

$$\Gamma_0/T \approx (T/E_c)^2 \ll 1, \quad (2.13)$$

where Γ_0 is the characteristic energy-level width of the thermal TLS and is defined by Eq. (2.4), while $E_c \approx (\rho \hbar^3 v_t^5)^{1/2}/M$ is the characteristic energy introduced by Gurevich and Parshin¹⁸; it is of the order of 30 K.

The result is the integral equation (Fig. 2)

$$\mathcal{T}_{ik}(\epsilon + \omega, \epsilon) = 1 + \frac{1}{\pi} \int_{-\infty}^{+\infty} dx \Phi(x - \epsilon) g_i(x + \omega) \times \mathcal{T}_{\bar{i}\bar{k}}(x + \omega, x) g_{\bar{k}}(x). \quad (2.14)$$

Since the function $\Phi(x)$ is smooth, it can be reduced to the form

$$\mathcal{T}_{ik}(\epsilon + \omega, \epsilon) = 1 + 2i\Phi(h_{\bar{k}} - \epsilon) \Pi_{\bar{i}\bar{k}}(\omega). \quad (2.15)$$

Multiplying this expression by $g_i(\epsilon + \omega) g_k^*(\epsilon)/2\pi i$ and integrating with respect to ϵ from $-\infty$ to $+\infty$, we obtain for Π the system of equations

$$[\Pi_{ik}^{(0)}(\omega)]^{-1} \Pi_{ik}(\omega) - 2i\Phi(h_{\bar{k}} - h_k) \Pi_{\bar{i}\bar{k}} = 1, \quad (2.16)$$

where

$$\Pi_{ik}^{(0)}(\omega) = \int_{-\infty}^{+\infty} \frac{d\epsilon}{2\pi i} g_i(\epsilon + \omega) g_k^*(\epsilon) = \left[\omega - (h_i - h_k) + \frac{i}{2}(\Gamma_i + \Gamma_k) \right]^{-1}. \quad (2.17)$$

In the resonant approximation of interest to us, (2.16) at $\omega > 0$ is a system of two equations, with two unknowns, which relate the quantities Π_{13} and Π_{24} .

Taking into account expressions (2.3) for h_i and introducing the notation $\Gamma_{\pm} = 2\Phi(\pm E)$, $z = \omega - e$, we obtain

$$\Pi_{13} = \frac{z + J + i(\Gamma_+ + \Gamma_-)}{(z - J + i\Gamma_-)(z + J + i\Gamma_+) + \Gamma_+ \Gamma_-}. \quad (2.18)$$

The expression for $\Pi_{13} = \Pi_{24}$ differs from (2.18) by the substitution $J \rightarrow -J$ in the numerator.

The result is the following expression for the susceptibility:

$$\chi(\omega) = \frac{\omega}{4TD^{1/2}} \left[\frac{z_2 + J \operatorname{th}(E/2T)}{z - z_1} - \frac{z_1 + J \operatorname{th}(E/2T)}{z - z_2} \right], \quad (2.19)$$

where

$$D = \left[J + \frac{i}{2}(\Gamma_+ - \Gamma_-) \right]^2 - \Gamma_+ \Gamma_-, \quad (2.20)$$

$$z_{1,2} = -\frac{i}{2}(\Gamma_+ + \Gamma_-) \pm D^{1/2}. \quad (2.21)$$

Expression (2.19) admits of a simple qualitative interpretation (see Refs. 19 and 20). It describes the absorption spectrum of a two-level system in which the level spacing fluctuates on account to transitions that occur in a thermal TLS and changes jumpwise from a value $e + J$ to a value $e - J$. The average lifetime in these states is respectively $1/\Gamma_-$ and $1/\Gamma_+$.

Let us analyze (2.19) in the limiting cases of strong and weak interaction between the TLS. It can be verified that the measure of the interaction force is the ratio of the interaction energy J to the characteristic smearing $\Gamma = \Gamma_+ + \Gamma_-$ of the quasi-fermion levels due to transitions in the thermal TLS.

1. At

$$J \gg \Gamma \quad (2.22)$$

expression (2.19) is transformed into

$$\chi(\omega) = -\frac{\omega}{2T} \left[\frac{n_0(E)}{\omega - e - J + i\Gamma_-} + \frac{1 - n_0(E)}{\omega - e + J + i\Gamma_+} \right], \quad (2.23)$$

$$n_0(E) = [\exp(E/T) + 1]^{-1},$$

where $n_0(E)$ is the Fermi function. Actually this is none other than the average occupation number of the upper level of the thermal TLS. Expression (2.23) has a lucid physical meaning. It describes the splitting of the single absorption peak at $\omega = e$, which is present in the absence of the interaction, into two peaks corresponding to the resonant frequencies $\omega = e \pm J$. The relative powers of each of these peaks are proportional to the fractions of the time that the thermal TLS remains in the upper and lower states, respectively, i.e., to equilibrium populations $n_0(E)$ and $1 - n_0(E)$.

As for the resonance widths, in this situation (see below) they are in no way determined by the real transitions of the resonant TLS, but by the loss of phase coherence of its wave function when transitions occur in the thermal TLS. These transitions are due either to emission of thermal phonons [first term of (2.23)] or to their absorption (second term). By virtue of inequality (2.22) each such transition produces complete dephasing, so that the peak widths are determined by the probabilities of departure from the states of the doublets (2.3) and (2.4) corresponding to transition energies $e \pm J$ (see Fig. 1). The total absorbed power remains the same as before.

2. At

$$J \ll \Gamma \quad (2.24)$$

expression (2.19) reduces to

$$\chi(\omega) = -\frac{\omega}{2T} \left[\omega - e + J \operatorname{th} \frac{E}{2T} + 4i \frac{J^2 \Gamma_+ \Gamma_-}{(\Gamma_+ + \Gamma_-)^3} \right]^{-1}. \quad (2.25)$$

This result describes the line narrowing known from paramagnetic and nuclear magnetic resonance,^{19,20} and ad-

mits of a simple interpretation. Indeed, when the level spacing of the resonant TLS is changed very frequently by transitions in the thermal TLS, the external alternating field "feels" the average value of this spacing: $e + J [n_0(E) - (1 - n_0(E))] = e - J \operatorname{tanh}(E/2T)$. As for the damping, in this case it is also determined by the dephasing of the resonant-TLS wave function. The dephasing proceeds now, by virtue of (2.24), in a diffuse manner, in small steps. Therefore the characteristic broadening must be equal to D_φ , where D_φ is the phase diffusion coefficient. This quantity can be easily expressed in term of the change of the energy J in each transition and in terms of the transition frequency Γ :

$$D_\varphi \approx J^2/\Gamma.$$

We obtain thus an estimate that agrees qualitatively with (2.25)—the higher the transition frequency in the thermal TLS, the narrower the absorption line of the resonant TLS (line narrowing). We note that it is precisely in the second limiting case (2.24) that the absorbing TLS goes off resonance via diffusion.

In the foregoing analysis of the linear absorption we disregarded the intrinsic damping γ , due to the fourth term in Hamiltonian (2.1), of the resonant TLS. It can be shown that this is justified if $\gamma \ll \min(\Gamma, J^2/\Gamma)$ [see (3.15), (3.18), (3.21) and (3.23)].

The approach developed above makes it possible to consider relatively simply also the case when the resonant TLS has several (N) thermal neighbors. The result is a level scheme of the type shown in Fig. 1, consisting of 2^N doublets. The system of linear equations for the 2^N unknown II_{ik} is also similar in form to (2.16), with each equation having $N + 1$ terms (each doublet is connected with configurations that differ from it by a transition to one thermal TLS). Such a system of equations is simple to analyze in the limiting cases (2.22) and (2.24) (cf. Ref. 20).

3. NONLINEAR ABSORPTION. METHOD OF STOCHASTIC DIFFERENTIAL EQUATIONS

The results of the preceding section reduce thus briefly to the following physical picture. The thermal TLS executes, under the influence of the interaction with the phonons, quantum transitions (jumps) that influence the state of the absorbing resonant TLS. This picture will be used in the present section to describe nonlinear absorption. We use for this purpose the method of stochastic differential equations, since it permits the nonlinearity to be taken into account in a relatively simple manner.

This method is relatively simple because the jumps are assumed in it to be instantaneous. In other words, the time of the jump is not contained at all in the formulation of the problem. This corresponds fully to the results of the preceding section. Its final formulas likewise did not involve the behavior of the system at short times of the order of $1/e$ or $1/E$. Moreover, we go below to the limit of the linear case, compare directly the results with the final expressions of the preceding section, and ultimately verify that they agree.

We shall assume that the duration of the RF field pulse is much longer than the time interval $1/\Gamma$ between the

jumps. In other words, we consider absorption in the stationary regime (the same stationary condition was in fact used by us in the preceding section, where the Green's functions were assumed to be independent of the total time).

We consider a resonant TLS in a classical random strain field. The fluctuations of this field are due to jumplike transitions of the thermal TLS. As a result, the level spacing of the resonant TLS is a random function of the time:

$$e + J\xi(t), \quad (3.1)$$

where $\xi(t)$ is described by the so-called telegraph process²¹: it takes on alternately values $+1$ and -1 at random instants of the time; the random frequency of the jumps is Γ . For the sake of simplicity we shall not distinguish here between the lifetimes of the thermal TLS on the upper and lower levels. In terms of the preceding section this corresponds to the approximation

$$\Gamma_+ = \Gamma_- = \Gamma. \quad (3.2)$$

The absorption of the resonant TLS is determined by its density matrix:

$$\begin{pmatrix} n & -ife^{i\omega t} \\ if^*e^{-i\omega t} & 1-n \end{pmatrix}.$$

In the resonance approximation (2.9), the equations for n and f take the form (cf. Ref. 22)

$$\frac{\partial n}{\partial t} = -\gamma(n-n_0) - F \operatorname{Re} f, \quad (3.3)$$

$$\frac{\partial f}{\partial t} + isf + \frac{1}{2}\gamma f = \frac{1}{2}F(2n-1), \quad (3.4)$$

where $\gamma = \gamma_+ + \gamma_-$ is the total resonant-TLS level width due to emission and absorption of phonons of energy e :

$$\begin{aligned} s &= z - J\xi(t), \quad z = \omega - e, \\ n_0 &= [\exp(e/T) + 1]^{-1}, \end{aligned} \quad (3.5)$$

n_0 is the equilibrium population of the upper level of the resonant TLS.

It must be noted that in the nonlinear-absorption problem allowance for the natural damping γ of the resonant TLS is essential wherever it describes the population relaxation of the resonant TLS [see (3.3)]. In addition, in this section we take γ into account also where it describes the relaxation of the resonant-TLS wave-function phase [third term in the left-hand side of (3.4)], in order to understand its contribution in comparison with the spectral-diffusion mechanism.

The absorption of the resonant-TLS alternating-field energy is determined by the imaginary part of the susceptibility, $\operatorname{Im} \chi(\omega)$, which is connected with the off-diagonal component of the density matrix f by the relation

$$\operatorname{Im} \chi(\omega) = 2 \operatorname{Re} \langle f \rangle / F, \quad (3.6)$$

where the angle brackets denote averaging over the realization of the random process $\xi(t)$.

The value of $\langle f \rangle$ is determined from Eqs. (3.3) and (3.4) averaged over the realizations of the random process $\xi(t)$

$$\frac{\partial \langle n \rangle}{\partial t} = -\gamma(\langle n \rangle - n_0) - F \operatorname{Re} \langle f \rangle, \quad (3.7)$$

$$\frac{\partial \langle f \rangle}{\partial t} + iz \langle f \rangle - J \langle \xi f \rangle + \frac{1}{2}\gamma \langle f \rangle = \frac{1}{2}F(2\langle n \rangle - 1). \quad (3.8)$$

The set of equations obtained for the mean values is not closed, since it contains the quantity $\langle \xi f \rangle$. Since the telegraph process $\xi(t)$ is a Markov random process, the following equation, as shown in Ref. 21, holds for $\langle \xi f \rangle$:

$$\frac{\partial \langle \xi f \rangle}{\partial t} + 2\Gamma \langle \xi f \rangle - \left\langle \xi \frac{\partial f}{\partial t} \right\rangle = 0. \quad (3.9)$$

A similar equation holds also for $\langle \xi n \rangle$. Using now Eqs. (3.3) and (3.4) and the fact that $\xi^2(t) = 1$ is a determined quantity, we obtain in the stationary case the following system of four equations for the quantities $\langle f \rangle$, $\langle n \rangle$, $\langle \xi f \rangle$, and $\langle \xi n \rangle$:

$$\begin{aligned} \gamma(\langle n \rangle - n_0) + F \operatorname{Re} \langle f \rangle &= 0, \\ (\gamma/2 + iz) \langle f \rangle - iJ \langle \xi f \rangle &= F(\langle n \rangle - 1/2), \\ (2\Gamma + \gamma) \langle \xi n \rangle + F \operatorname{Re} \langle \xi f \rangle &= 0, \\ (2\Gamma + \gamma/2 + iz) \langle \xi f \rangle - iJ \langle f \rangle &= F \langle \xi n \rangle. \end{aligned} \quad (3.10)$$

The solution of this system of linear equations for $\operatorname{Re} \langle f \rangle$ is

$$\operatorname{Re} \langle f \rangle = \frac{1}{2}F \operatorname{th} \frac{e}{2T} \frac{az^2 + d}{z^4 + 2bz^2 + c}, \quad (3.11)$$

where

$$\begin{aligned} a &= \frac{\gamma}{2}, \quad d = \frac{1}{2}(\gamma + 4\Gamma) \left(J^2 + \frac{\gamma^2}{4} + \gamma\Gamma + \frac{F^2}{2} \right), \\ b &= \frac{\gamma^2}{4} + \gamma\Gamma + 2\Gamma^2 - J^2 + \frac{1}{2}F^2 \frac{\gamma + 3\Gamma}{\gamma + 2\Gamma}, \\ c &= \left(J^2 + \frac{\gamma^2}{4} + \gamma\Gamma \right)^2 + 2F^2 \left(\Gamma + \frac{\gamma}{4} \right)^2 + \frac{\gamma^2 F^2}{8} \frac{\gamma + 4\Gamma}{\gamma + 2\Gamma} \\ &\quad + \frac{1}{2}J^2 F^2 \left(\frac{\gamma + 4\Gamma}{\gamma} + \frac{\gamma}{\gamma + 2\Gamma} \right) + \frac{F^4}{4} \frac{\gamma + 4\Gamma}{\gamma + 2\Gamma}. \end{aligned} \quad (3.12)$$

As $F \rightarrow 0$ and at $\gamma \ll \min(\Gamma, J^2/\Gamma)$ we obtain from (3.11), taking (3.6) into account, the same results that expression (2.19) gives for $\operatorname{Im} \chi(\omega)$, provided we put in (2.19) $\Gamma_+ = \Gamma_- = \Gamma$ and $\tanh(E/2T) = 0$. These two equations are interrelated and stem from the fact that they are quite well satisfied at $E \ll T$.

We start the analysis of Eq. (3.11), which describes the nonlinear absorption, with a typical experimental situation, when

$$\Gamma \gg \gamma. \quad (3.13)$$

Three limiting cases are possible here: a) $J \gg \Gamma$, b) $\Gamma \gg J \gg (\gamma\Gamma)^{1/2}$, c) $(\gamma\Gamma)^{1/2} \gg J$. We consider the first:

$$\text{a) } J \gg \Gamma \gg \gamma. \quad (3.14)$$

In this case we have from (3.11) in the principal pole approximation

$$\begin{aligned} \operatorname{Im} \chi(\omega) &= \frac{\omega}{4T} \left[\frac{\Gamma}{(z-J)^2 + \Gamma^2 (1 + F^2/2\gamma\Gamma)} \right. \\ &\quad \left. + \frac{\Gamma}{(z+J)^2 + \Gamma^2 (1 + F^2/2\gamma\Gamma)} \right]. \end{aligned} \quad (3.15)$$

It follows from this that the nonlinear effects in the absorption of one resonant TLS become substantial at an amplitude F of the order of and larger than a certain critical value

$$F \approx F_{c1} = (2\gamma\Gamma)^{1/2}. \quad (3.16)$$

In the second limiting case

$$b) \Gamma \gg J \gg (\gamma\Gamma)^{1/2}. \quad (3.17)$$

In the principal pole approximation we have from (3.11)

$$\text{Im } \chi(\omega) = \frac{\omega}{2T} \frac{J^2}{2\Gamma} \left[z^2 + \frac{J^2}{2\Gamma} \left(1 + \frac{2\Gamma F^2}{\gamma J^2} \right) \right]^{-1}. \quad (3.18)$$

In this case the critical amplitude F_{c2} is given by

$$F_{c2} = (J^2\gamma/2\Gamma)^{1/2}. \quad (3.19)$$

Finally, in the third case

$$c) \Gamma \gg (\gamma\Gamma)^{1/2} \gg J \quad (3.20)$$

we have

$$\text{Im } \chi(\omega) = \frac{\omega}{2T} \frac{\gamma}{2} \left[z^2 + \frac{\gamma^2}{4} \left(1 + \frac{2F^2}{\gamma^2} \right) \right]^{-1}, \quad (3.21)$$

and

$$F_{c3} = \gamma/\sqrt{2}. \quad (3.22)$$

These results have a rather simple qualitative interpretation. The critical intensity F_c^2 is determined by the balance between the processes of the transition of the resonant TLS with absorption of a quantum, on the one hand, and processes of relaxation of their population on account of emission of phonons of frequency close to ω , on the other. The rate of the transitions with absorption of a quantum (at resonance) at $F = F_c$ is (in order of magnitude) F_c^2/δ , where δ is the resonant-TLS absorption linewidth in the linear regime; the population relaxation rate is γ . Hence $F_c \approx (\gamma\delta)^{1/2}$. In case (a), $\delta \approx \Gamma$ and we have the estimate (3.16); in case (b) $\delta \approx J^2/\Gamma$ and we arrive at (3.19); finally, in case (c) the principal role in the dephasing of the wave function of the resonant TLS is played by emission and absorption of phonons of the resonant TLS itself, with energy ω , while the spectral diffusion is inessential. We obtain therefore for the critical amplitude the same result (3.21) as in the absence of spectral diffusion.

We consider now the case $\gamma \gg \Gamma$. From (3.11) we obtain

$$\text{Im } \chi(\omega) = \frac{\omega}{4T} \left[\frac{\gamma/2}{(z-J)^2 + \gamma^2/4 + 2F^2/\gamma^2} + \frac{\gamma/2}{(z+J)^2 + \gamma^2/4 + 2F^2/\gamma^2} \right]. \quad (3.23)$$

In this case the processes of transition to the thermal TLS do not influence the resonant-TLS effective linewidth, since the characteristic time that forms the line profile is too short for such transition to take place. The answer is therefore a sum of peaks centered at $\omega = e \pm J$ and having a width determined by the natural damping γ of the resonant TLS.

The nonlinear-absorption problem is thus reduced to a determination of the effective dephasing time of the resonant-TLS wave function, i.e., of its absorption linewidth δ .

4. NONLINEAR ABSORPTION. AVERAGING OVER A TLS ENSEMBLE

We have determined absorption by one pair consisting of a TLS with energy e close to ω , and a thermal TLS with energy $E \approx T$. It is known, however, that the energy spectrum of TLS in glasses is quite broad.⁵ Therefore there is practically equal probability of encountering in different glass-powder particles low-energy TLS with different values of e , as well as their neighbors with different values of E , including E of the order of T . Moreover, even TLS having the same energy make different contributions to the absorption. The point is that the squared matrix element F^2 of the interaction with the wave is proportional to the square of the ratio of the tunnel level splitting λ to the energy e (Ref. 23):

$$F^2 = F_0^2 p, \quad p = (\lambda/e)^2 \leq 1. \quad (4.1)$$

The same holds for the quantities γ and Γ :

$$\gamma = \gamma_0 p, \quad \Gamma = \Gamma_0 P, \quad P = (A/E)^2 \leq 1, \quad (4.2)$$

where A is the energy of the tunnel splitting of the thermal TLS, and $1/\gamma_0$ and $1/\Gamma_0$ are the minimum relaxation times, respectively, of resonant and thermal TLS with specified level spacing. The interaction energies J of the resonant and thermal TLS in two different powder particles are also generally speaking different, since $J \propto r^{-3}$, where r is the distance between the TLS. It must be noted that J depends also on the difference between p and P ($J \propto [(1-p) \cdot (1-P)]^{1/2}$ (Refs. 6 and 13). We, however, will disregard this dependence in the estimates that follow, since allowance for it leads to no qualitative changes.

To determine the total absorption it is necessary to sum over all the glass-powder particles in the volume. We assume that the characteristics of the resonant and thermal TLS that constitute the interacting pair do not correlate. We sum accordingly over all the glass-powder particles that contain resonant TLS, and average the result over the characteristics of their environment, i.e., over the parameters E and P of the neighboring thermal TLS and over the distance r to it.

To this end we take into account the fact that the distribution function of the resonant TLS in the parameters e and p can be represented in the form²³

$$N_0/p(1-p)^{1/2}, \quad (4.3)$$

where N_0 is the TLS density of states in glasses and is assumed to be independent of the energy e . The summation over the resonant TLS reduces therefore to integration with respect to de and dp with weight (4.3). It can be seen from this that the nonlinear absorption is not by the position of the absorption-peak center, but only by the effective width of this peak at a given amplitude F of the resonant wave.

The density of the thermal TLS in glass is of the order $N_0 T$, so that the average distance between them is $r_T \approx (N_0 T)^{-1/3}$. More accurately, $r_T \approx (N_0 T \mathcal{L})^{-1/3}$, where $\mathcal{L} = \ln \Gamma_0/\gamma_0$, i.e., of importance to us are those thermal TLS in which the transitions between levels are fast enough compared with the resonant ones. Since we are interested in a situation in which a powder particle having a characteristic dimension r_0 contains not more than one thermal TLS, we

assume that the condition $r_0 < r_T$ is satisfied.

Each powder particle containing a resonant TLS has then a maximum probability, $W_0 \approx 1$, of having no fast thermal TLS. The probability W_1 that a resonant TLS has one neighboring fast thermal one is of the order of

$$W_1 \approx (r_0/r_T)^3 = N_0 T r_0^3 \mathcal{L} < 1. \quad (4.4)$$

The probability of configurations with large numbers of fast thermal neighbors is accordingly much less than W_1 , and we shall disregard these configurations. Spectral-diffusion processes are important only for particles containing thermal TLS. As for particles that contain only resonant TLS, their absorption coefficient saturates already at small amplitudes $F_c \sim \gamma_0$. This coefficient is equal in the linear regime to $(F \ll \gamma_0) c_0 \alpha_0 (1 - W_1)$, where c_0 is the relative density of the glass particles and α_0 is the absorption coefficient in bulky glass. At $F \gg \gamma_0$ it is determined by those powder particles which contain both a resonant and a thermal TLS.

The nonlinear absorption coefficient depends on the ratio of the characteristic parameter J_{\min} and Γ , where J_{\min} is the energy of the interaction of the resonant and thermal TLS separated by a characteristic distance of the order of the size r_0 of the amorphous particle.

We begin with an analysis of the case

$$J_{\min} \gg \Gamma_0. \quad (4.5)$$

The inequality $J_{\min} \gg (\Gamma_0 \gamma_0)^{1/2}$ is then satisfied automatically, since $\gamma_0 \ll \Gamma$ because $\omega \ll T$. For the absorption coefficient we obtain in this case

$$\alpha = c_0 \alpha_0 W_1 \frac{1}{\mathcal{L}} \left\langle \Phi_1 \left(\frac{2\gamma_0 \Gamma_0}{F^2} \right) \right\rangle_p, \quad (4.6)$$

where

$$\Phi_1(z) = \ln [1 + 2z + 2(z(1+z))^{1/2}], \quad (4.7)$$

α_0 is the linear absorption coefficient, and the angle brackets with subscript p denote averaging over the parameter p of the resonant TLS with distribution function (4.3) and weight p . This averaging does not alter the asymptotic form of the dependence. Since $W_1 \propto \mathcal{L}$, the large logarithm \mathcal{L} in this equation is canceled out and does not enter in the final answer. Inasmuch as we assume here that $F \gg \gamma_0$, the absorption in the nonlinear regime (4.6) in this range of F turns out to be smaller than the linear absorption with $\alpha_0 W_1$.

It can be seen from (4.6) that the critical value of the wave amplitude is

$$F_{c1} = (2\gamma_0 \Gamma_0)^{1/2}. \quad (4.8)$$

At $F \gg_{c1}$ we have

$$\alpha \approx \alpha_0 (W_1/\mathcal{L}) (F_{c1}/F),$$

and at $F \ll_{c1}$

$$\alpha \approx \alpha_0 (W_1/\mathcal{L}) \ln (4F_{c1}^2/F^2),$$

i.e., the dependence on the intensity is weakly logarithmic. The main contribution to the absorption are made then by powder particles with thermal TLS, for which $\Gamma \approx F^2/$

$\gamma_0 \ll \Gamma_0$, i.e., with a strongly asymmetric two-well potential.

There are thus two stages of nonlinear behavior of α : during the first stage are saturated the resonant TLS that have no neighbors (or have a slow neighbor with $\Gamma < \gamma_0$); during the second state are saturated the remaining TLS, and the critical amplitude is determined by the characteristic frequency of the transitions to the thermal TLS.

The case

$$\gamma_0 \ll J_{\min} \ll \Gamma_0 \quad (4.9)$$

is more complicated, since the powder particle can contain a thermal TLS with $\Gamma \ll J_{\min}$. We are therefore unable to use the asymptotic expression for α at $J \ll \Gamma$.

To estimate the $\alpha(F)$ dependence we subdivide the plane of the parameters J and Γ into regions where $J > \Gamma$ and $J < \Gamma$ respectively. In each of these regions we use the corresponding asymptotic expression [(3.15) or (3.18)] and assume that the quantities are matched, in order of magnitude, on the boundary between the regions. It is found as a result that the critical amplitude is

$$F_{c2} \approx (\gamma_0 J_{\min})^{1/2}. \quad (4.10)$$

At $F \gg_{c2}$ we have

$$\alpha \approx \alpha_0 (W_1/\mathcal{L}) (F_{c2}/F), \quad (4.11)$$

at $F \ll_{c2}$

$$\alpha \approx \alpha_0 (W_1/\mathcal{L}) \ln (F_{c2}/F) \quad (4.12)$$

and the main contribution to the absorption is made by powder particles whose thermal TLS has $\Gamma \approx J_{\min}$.

The nonlinear behavior goes thus in this case through two stages: the logarithmic plateau (4.12) and the fall-off section (4.11). The critical intensity F_{c2} however, is determined in this case by expression (4.10).

In the case $\Gamma_0 \gg \gamma_0 \gg J_{\min}$, finally, the spectral diffusion is negligible and does not affect the nonlinear behavior of the absorption.

5. DISCUSSION OF RESULTS

Thus, the general character of the dependence of the absorption on the intensity, except in the last case $\Gamma_0 \gg \gamma_0 \gg J_{\min}$, is of the following form. The first section of the relatively steep (like $I^{-1/2}$) decrease is due to the contribution of both the resonant TLS that have no thermal neighbor and of the ones having a "slow" neighbor (with transition frequency $\Gamma \lesssim \gamma_0$). This is followed, as the intensity is increased, by a "plateau" section, where the dependence on the intensity is logarithmic. It extends to the critical intensity determined by the smaller of the two expressions (4.8) and (4.10). Next comes the second section of the steep decrease, again like $1/F$. The position of the start of this second section depends thus on the ratio of J_{\min} and Γ_0 , i.e., on the relation between the particle dimensions and the temperature.

In the estimates that follow we introduce a temperature T_0 connected with the particle size by the relation

$$r_T|_{T=r_0} = r_0, \quad T_0 = 1/N_0 r_0^3 \mathcal{L}. \quad (5.1)$$

In this notation we have $W_1 = T/T_0$, so that the condition

for the validity of our theory takes the form

$$T < T_0. \quad (5.2)$$

According to Ref. 6, we have for Γ_0 , γ_0 , and $J(r)$

$$\Gamma_0 \approx \frac{T^3}{E_c^2}; \quad \gamma_0 \approx \frac{\omega^2 T}{E_c^2}; \quad J(r) \approx \frac{\Lambda_0^2}{\rho v_i^2 r^3}, \quad (5.3)$$

where Λ_0 is the deformation-potential constant (of the order of 1 eV), and $E_c = (\rho \hbar^3 v_i^5)^{1/2} / \Lambda_0 \approx 30$ K. Therefore $J_{\min} \approx \beta \mathcal{L} T_0$, where $\beta = \Lambda_0^2 N_0 / \rho v^2$ is a dimensionless parameter. The condition $J_{\min} \approx \Gamma_0$ is thus equivalent to the condition

$$T \approx (T_0 T_D^2 \mathcal{L})^{1/2}, \quad (5.4)$$

where $T_D = (N_0 v^3 \hbar^3)^{1/2}$ is the characteristic temperature, which plays an important role in the theory of spectral diffusion. The quantity \mathcal{L} is determined by the formula

$$\mathcal{L} \approx \ln(\Gamma_0 / \gamma_0) \approx \ln(T / \omega)^2. \quad (5.5)$$

The case $J_{\min} > \Gamma_0$ (4.5), with allowance for (5.2), is thus realized at

$$\omega \ll T < T_0, \quad (T_0 T_D^2 \mathcal{L})^{1/2}. \quad (5.6)$$

The opposite case $J_{\min} < \Gamma_0$ occurs when

$$T < T_0 < T^3 / T_D^2. \quad (5.7)$$

These conditions are compatible only when

$$T > T_D. \quad (5.8)$$

Finally, the case $J_{\min} < \gamma_0 \ll \Gamma$, when the spectral diffusion is not important, is realized under the condition

$$\mathcal{L} T_0 T_D^2 / \omega^2 < T < T_0, \quad (5.9)$$

i.e., it calls for rather high frequencies $\omega > \mathcal{L}^{1/2} T_D$.

We obtain numerical estimates of the quantities that enter in the theory. Putting $N_0 \approx 10^{23} \text{ erg}^{-1} \text{ cm}^{-3}$ (Ref. 6) and $v_i = 2 \cdot 10^5 \text{ cm/s}$ we obtain $T_D \approx 1$ K.

For $T \approx 1$ K and at $2\pi/\omega \approx 1$ GHz the characteristic distance between the significant thermal pairs is

$$r_T \approx 10^{-6} \text{ cm} \approx 100 \text{ \AA},$$

i.e., the characteristic particle size should not exceed 100 \AA in this case.

The quantity usually measured is the nonlinear-absorption critical intensity and its dependence on frequency and temperature. It follows from (4.8) and (4.10) that

$$F_{c1}^2 \sim \omega^2 T^4 / E_c^4; \quad F_{c2}^2 \sim \omega^2 T J_{\min} / E_c^2. \quad (5.10)$$

6. CONCLUSION

Let us summarize our results. The behavior of the nonlinear absorption of a wave by a resonant TLS + thermal TLS system is determined by the relation between three quantities, the energy of interaction of the two TLS, the frequency Γ of the transitions in the thermal TLS, and the natural absorption linewidth γ of the resonant TLS.

At $J \gg \max(\Gamma, \gamma)$ the absorption spectrum takes in the linear case the form of two relatively narrow peaks (doublet at $\omega \pm J$, with width $\sim \max(\Gamma, \gamma)$). In the case $\Gamma \gg \max(J, (\Gamma\gamma)^{1/2})$ one peak of width $\max(J^2/\Gamma, \gamma)$ is produced. The quantity J^2/Γ is none other than the coefficient of the resonant-TLS wave-function phase diffusion due to frequent transitions in the thermal TLS. At $J \ll \gamma$, finally, the thermal TLS has no significant effect on the absorption of the resonant TLS.

The described structure of the absorption spectrum remains the same also in the nonlinear situation, but the heights and widths of the peaks become dependent on the wave amplitude F . The nonlinearity becomes substantial when the quantity F^2/γ becomes comparable with absorption linewidth in the linear regime. This condition determines the critical amplitude F_c of the nonlinear absorption. At $F \gg F_c$ the height of the peak decreases in proportion to F^{-2} , and its width increases in proportion to F .

Owing to the broad distribution of the energies ϵ of the resonant TLS in glasses, the wave absorption coefficient is determined only by the absorption intensity integrated over the spectrum. The spectral diffusion processes therefore do not affect the linear absorption. Their influence in the nonlinear regime is extremely important.

Nonlinear absorption in a system of amorphous particles (glass powder) is determined to a considerable degree by the dimension of the particles r_0 , as well as by the frequency ω of the wave and by the temperature T . The first of these quantities determines the minimum energy $J_{\min} \propto r_0^{-3}$ of the TLS interaction in the particle, and the last two determine the minimum relaxation times of the resonant and thermal TLS, i.e., $1/\gamma_0$ and $1/\Gamma_0$.

The dependence of the absorption α on the intensity F^2 in powdered glass at $\omega \ll T$ takes the form of two steps (except for the case $\Gamma_0 \gg \gamma_0 \gg J_{\min}$, when the spectral diffusion is of no importance at all). The first step is due to the saturation of the resonant TLS in particles that have no thermal TLS, or else in particles where the transition frequency of the thermal TLS is low. The corresponding critical amplitude is $F_c \propto \gamma_0$. The second step is due to saturation of those TLS which have a sufficiently "fast" thermal neighbor. In view of the large scatter of the transition frequencies Γ of the thermal TLS in glass, it turns out that the critical intensity for the second step is of the order of $\gamma_0 \min(\Gamma_0, J_{\min})$.

By varying the particle size r_0 and the temperature it is possible, as we have seen, to change the ratio of J_{\min} and Γ_0 . By the same token, a study of the nonlinear absorption in glass powders can yield much additional important information on the properties of TLS in glasses.

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¹ Validity of the Bloch equations can be corroborated only in this case.

² We disregard here the off-diagonal Hamiltonian terms that describe the TLS interaction, such as $s_2 S_1, s_3 S_1$, etc.⁸ It can be shown that at $|J| \ll \omega$ their contribution can be neglected compared with the contribution of the term $s_3 S_3$.

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