Nonlinear resonant absorption in glasses in the two-dimensional case; spectral diffusion

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A theory is developed to describe nonlinear low-temperature phenomena in two-dimensional amorphous dielectrics. Nonlinear resonant absorption of microwaves or surface sound, and hole-burning in the spectrum of a weak signal due to the presence of a strong signal at a slightly different frequency are considered. Both of these phenomena are caused by the interaction of an external signal with two-level systems present in amorphous materials. The interaction among the two-level systems results in spectral diffusion and its influence on the absorption is analyzed. A two-dimensional problem is considered for a thin amorphous film on a crystal substrate and for a crystal surface with a high degree of disorder. Techniques developed previously for the three-dimensional case are applied to the two-dimensional situation to derive general expressions for the critical intensity for nonlinear absorption, and for the shape of the burnt hole. Analytic expressions are derived for several limiting cases.

1. INTRODUCTION AND STATEMENT OF THE PROBLEM

Like microwave absorption, low-temperature absorption of sound in amorphous dielectrics is known to be due to the interaction of the absorbed signal with two-level systems present in the material[1,2] (see also the review in Ref. 3). The two-level systems consist of an atom or group of atoms capable of tunneling between two equilibrium positions.

The purpose of the present paper is to develop a two-dimensional theory of resonant absorption which treats the spectral diffusion. The importance of the latter in numerous physical systems was first pointed out by Klauder and Anderson[4] in their study of magnetic resonance. Its role in the physics of glasses was discussed by Joffrin and Levelut[5] and by Hunklinger and Arnold.[3] A quantitative theory of nonlinear phenomena in the three-dimensional case has been developed by Gal'perin, Parshin, and the author.[6,7]

Nevertheless, numerous two-dimensional nonlinear effects are encountered in current experimental work. Two cases are of particular interest to us. a) The case of a perfect crystal whose surface, however, is highly disordered, due both to imperfections in the surface itself and to adsorption of foreign atoms. The surface influences the absorption of surface acoustic waves, the nonlinear losses upon reflection of a volume acoustic wave, and the absorption of microwaves. The latter absorption is most easily observed in systems with a large surface area, such as crystal powders.[1)] b) The case of a thin amorphous film on a crystal substrate. Because the methods applied here are the same as in Refs. 6 and 7, we will describe them rather briefly, emphasizing those features special to the two-dimensional case.

Spectral diffusion can be described as follows. Consider a resonant two-level system with levels separated by $\hbar\omega$, where ω is the frequency of the ultrasound or microwave radiation. The resonant two-level system interacts with neighboring thermal centers, whose energy levels are spaced by an amount comparable to the temperature T expressed in energy units. This interaction is due to the elastic stresses and electric fields generated by the thermal two-level centers. In our application the dynamic part of the interaction, i.e., the component that depends on whether the thermal two-level system is in the ground or an excited state, is important. For these thermal two-level systems, quantum transitions (hops) modulate the distance between the levels of the resonant two-level system, thereby broadening the resonance.

In this paper we consider two nonlinear phenomena in which spectral diffusion can play a dominant role: nonlinear absorption (characterized by the critical intensity F_c), and hole burning. We will consider the stationary case, although the techniques developed also apply in principle to the time-dependent case. Moreover, we assume that the phonon system relaxes fast enough so that the phonons may be assumed to be in equilibrium. Nonlinear absorption of resonant radiation in three-dimensional amorphous dielectrics was analyzed in Refs. 8–10 for the case when the departure of the phonon system from equilibrium is significant. It was shown there that phonon nonequilibrium can change the qualitative behavior. Nevertheless, in the two-dimensional case one can usually choose the experimental conditions so as to eliminate this factor by ensuring that the phonons rapidly leave the two-dimensional region in which they are excited.

The governing parameters are as follows. The interaction between the external ac field and the resonant two-level system is characterized by the matrix element for the transition, $\hbar F/2$. The explicit expression for F will of course depend on whether the system is excited by a sound wave or by a microwave field. The quantity F is just the Rabi frequency for the resonant two-level system, i.e., the frequency of the coherent oscillations in the populations of the upper and lower levels during the resonant excitation.

If the interaction among the two-level systems (i.e., the spectral diffusion) can be neglected, the resonant absorption coefficient can be expressed in terms of F and the damping coefficient γ of the resonant two-level systems (here γ is the intrinsic damping due to the interaction with the phonons). The absorption coefficient is proportional to the population difference for the upper and lower levels, multiplied by the spectral width of the absorption line. For $F \ll \gamma$, the popula-

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tion difference is equal to its equilibrium value and independent of F to lowest order. The absorption line is Lorentzian with width γ. The absorption coefficient is described by the linear theory in this approximation. To next higher order, one finds a negative correction to the absorption coefficient which for $F \ll \gamma$ is of relative order $(F/\gamma)^2$.

For $F \gg \gamma$ the population difference falls off as F^{-2}, i.e., it is inversely proportional to the intensity. On the other hand, because of the Rabi oscillations the width of the resonant region is directly proportional to F. The absorption coefficient is thus proportional to F^{-1}, and the critical amplitude F_c determining the magnitude of the nonlinear effects is equal to γ. It is also clear from this discussion that the width of the burnt hole is $\sim \gamma$ for $F \ll F_c$ and $\sim F$ for $F \gg F_c$.

The above picture changes radically when spectral diffusion is present. Hopping of neighboring two-level systems in this case causes the energy (the spacing between the levels) of a resonant two-level system to vary randomly with time. The scale of these fluctuations, denoted by $\hbar/\tau_d$, is proportional to the interaction energy for two adjacent thermal two-level systems, i.e., it is determined by $\bar{r}$, the average distance between them.

The distance $\bar{r}$ is given by a simple expression in each of the cases mentioned above. In case b), we have the order-of-magnitude estimate

$$\bar{r} \approx (PaT)^{-1/2}, \tag{1.1}$$

where a is the width of the amorphous layer and the constant P characterizes the volume density of the states in the two-level system. Expression (1.1) is valid as long as $\bar{r} \gg a$, i.e., $T \ll 1/Pa^3$; this is precisely the condition under which the problem may be regarded as two-dimensional.

In case a), $\bar{r}$ depends significantly on the energy dependence of the density Q of surface states for the two-level system. As systematic experimental data are lacking, we will assume for simplicity that Q is constant. In this case, $\bar{r} \approx 1/(QT)^{1/2}$.

The interaction energy for two two-level systems in an unbounded continuous medium can be expressed in the form A/r^3, where the angle-dependent quantity A determines the relative "orientation" of the two-level systems, i.e., the alignment of the dipole moments characterizing their electrical interaction, and also the orientation of the principal axes of the deformation potential tensors, characterizing the elastic interaction (see Ref. 11). We are interested in bounded continuous media; in this case, the interaction depends in a more complicated way on the spatial coordinates because of the additional terms needed to treat the image forces. However, as already noted we are interested in the case when the mean distance between the two-level systems is much greater than the width of the amorphous region. The decay in the interaction strength with distance is then the same as for an unbounded continuous medium. Only the explicit expression for A changes; we will not write it out, since it will not be required below. However, we will need the order-of-magnitude estimate

$$A \approx D^2/\rho w^2 \tag{1.2}$$

for the case of an elastic interaction; here D is the deformation potential, ρ is the density, and w is a suitably defined average speed of sound.[2)] Bearing in mind that D is typically of the order of 1 eV, we see that this numerical estimate also holds for the electrical interaction of the two-level systems. We will therefore treat only the elastic interaction below, with the understanding that the electrical interaction of the two-level systems in glasses leaves the order of magnitude of the estimates unchanged.

In view of the foregoing, for case a) we obtain the following estimate for the interaction energy of two-level systems separated by the mean distance $\bar{r}$:

$$\hbar/\tau_d \approx A(QT)^{3/2}. \tag{1.3}$$

For case b), we must replace Q here by Pa. Thus in contrast to the three-dimensional case for which $1/\tau_d \sim T$, here we have $1/\tau_d \propto T^{3/2}$. This is because the dependence of the interaction energy on $\bar{r}$ is of the same type as in the three-dimensional case, but the mean distance between the thermally excited two-level systems is proportional to $T^{-1/2}$ rather than to $T^{-1/3}$.

Even in the two-dimensional case, however, there are many situations of interest in which $1/\tau_d$ turns out to be larger than γ and F. This indicates that broadening of the resonant spectral interval by spectral diffusion may be important. On the other hand, it is clear that the parameter $1/\tau_d$ cannot describe the spectral diffusion completely, because it measures only the static interaction among the two-level systems. An additional parameter Γ giving the hopping frequency for the thermal two-level systems is also needed to describe the dynamics.

These hops are a consequence of one-phonon processes, and the characteristic hopping frequency Γ_0 can be estimated as follows[3]:

$$\Gamma_0 \approx T^3/\hbar E_c^2. \tag{1.4}$$

Here $E_c \approx (\rho\hbar^3 w^5)^{1/2} D \sim 10$–$30$ K is the characteristic energy introduced in Ref. 12.

The product $\Gamma_0 \tau_d$ plays a very important role in determining the qualitative nature of the spectral diffusion. This may be seen as follows. Consider a disk of radius r_t with a resonant two-level system at its center. The disk contains QTr_t^2 thermal two-level systems. Since the characteristic hopping frequency is Γ_0, the condition for at least one two-level system in the disk to undergo a transition during the time t is

$$QTr_t^2\Gamma_0 t \approx 1. \tag{1.5}$$

The corresponding characteristic change in the distance between the levels of a resonant two-level system is

$$\frac{A}{r_t^3} = A(QT\Gamma_0 t)^{3/2} = \frac{\hbar}{\tau_d}(\Gamma_0 t)^{3/2}. \tag{1.6}$$

The energy dependence here differs completely from the relation $A/r_t^3 \propto t$ found in the three-dimensional case. This also accounts for the different expression derived below for the phase relaxation time. Formula (1.6) holds if the number of thermal two-level systems in the disk is large but the transition probability for any one of them is small, so that $\Gamma_0 t \ll 1$.

The random jumps in the energy of a resonant two-level system give rise to relaxation of the phase of the wave function for the system (more precisely, of the off-diagonal elements of the density matrix). The characteristic phase relaxation time τ_φ is given by

$$\tau_d^{-1}\Gamma_0^{3/2}\tau_\varphi^{5/2} \approx 1, \tag{1.7}$$

so that

$$\tau_\varphi \approx \Gamma_0^{-3/5}\tau_d^{2/5}. \tag{1.8}$$

This expression is valid if the $t^{3/2}$ law still holds for times on the order of τ_φ, i.e., if $\tau_\varphi\Gamma_0 \ll 1$. Since $\tau_\varphi\Gamma_0 = (\tau_d\Gamma_0)^{2/5}$, the first possible limiting case corresponds to the inequality

$$\tau_d\Gamma_0 \ll 1. \tag{1.9}$$

Using (1.3) and (1.4), we can rewrite this as

$$T \ll T_d, \tag{1.10}$$

where the characteristic temperature T_d satisfies the following simple estimate:

$$T_d \approx \hbar^2 w^2 Q, \tag{1.11}$$

i.e., in the two-dimensional case Td is proportional to the surface density of states for the two-level systems.

In the opposite limit $\tau_d\Gamma_0 \gg 1$, the phase does not change significantly over times shorter than Γ_0^{-1}. In other words, we have $\tau_\varphi \gg \Gamma_0^{-1}$. On the other hand, for large times $t \gg \Gamma_0^{-1}$ the characteristic deviation of the energy for a resonant two-level system becomes independent of time because it cannot exceed $\hbar/\tau_d$ in order of magnitude. The energy spacing of the levels in this case thus varies randomly over an interval of width $\hbar/\tau_d$. The spectral width of this interval determines the phase relaxation time, which is equal to τ_d.

Studies of nonlinear resonant absorption in two-dimensional amorphous systems are of interest primarily for the light they can shed on the two-dimensional density of the two-level systems and hence, for example, on the density of surface defects on a crystal. Experimental data on spectral diffusion in this case would yield information on the nature of the interaction among the two-level systems and on their spatial correlation.

2. ESTIMATES; QUALITATIVE ANALYSIS

Before giving a quantitative description, we will use simple physical arguments to estimate the critical amplitude F_c and the width $\Delta\nu$ of the burnt hole.

We begin with the case of high temperatures $T \gg T_d$, for which $\Gamma_0\tau_d \gg 1$. Spectral diffusion determines the nonlinear absorption if

$$\gamma \ll 1/\tau_d, \tag{2.1}$$

which we will assume. In this case, all the two-level systems in a spectral interval of width $1/\tau_d$ about the resonant energy will undergo transitions induced by the external field and the populations will not be in equilibrium. The field-induced rate of population increase is $F^2\tau_d$, and the relaxation rate due to interactions with thermal phonons is γ. Comparison of these quantities leads to the estimate

$$F_c \approx (\gamma/\tau_d)^{1/2}. \tag{2.2}$$

The width of the burnt hole in this case is clearly $\sim 1/\tau_d$, i.e., it is much greater than F_c. Equation (2.2) has the same form as in the three-dimensional case; however, one should bear in mind that the expression for τ_d is different.

In the opposite limit $T \ll T_d$ we must distinguish two limiting cases in which spectral diffusion plays a role, namely

$$\Gamma_0 \ll \gamma \ll 1/\tau_\varphi \tag{2.3}$$

and

$$\gamma \ll \Gamma_0 \ll 1/\tau_\varphi. \tag{2.4}$$

In the first case the critical intensity can be found from the condition that the energy of a resonant two-level system fluctuates randomly by $\hbar F$ during a time F^{-1} equal to the Rabi oscillation period. We thus obtain

$$F_c \approx \Gamma_0^{3/5}\tau_d^{-2/5}, \tag{2.5}$$

The width of the burnt hole in this case may be estimated as follows. For $F \gtrsim F_c$, a resonant two-level system passing through the resonance interval during the time τ_φ will be excited with a probability close to 1. It remains excited for a much longer time $\approx \gamma^{-1}$ before emitting a phonon. Inserting this time into (1.6), we get

$$\Delta\nu \approx \frac{1}{\tau_d}\left(\frac{\Gamma_0}{\gamma}\right)^{3/2}. \tag{2.6}$$

(In the three-dimensional case the expression for $\Delta\nu$ contains the ratio Γ_0/γ to the first power.)

The estimate for F_c in the case (2.4) is based on the following qualitative arguments.[6] As in the previous case, the magnitude $\hbar/\tau_d$ of the random fluctuations in the energy of the resonant two-level systems greatly exceeds the width $\hbar/\tau_\varphi$ of the resonance itself. During the "lifetime" $\gamma^{-1} \gg \tau_\varphi$ the energy of a resonant two-level system returns to the resonance interval many times, the population each time increasing by $F^2\tau_\varphi^2 \ll 1$, i.e., while in the resonance interval the population grows at the rate $F^2\tau_\varphi$. However, the energy of the resonant two-level system lies within the resonance interval only during a time proportional to the width of the interval, so that

$$(1/\tau_\varphi)/(1/\tau_d) = \tau_d/\tau_\varphi.$$

The average population growth rate is thus

$$F^2\tau_\varphi(\tau_d/\tau_\varphi) = F^2\tau_d.$$

The critical intensity is found by equating this rate to the relaxation rate associated with phonon emission; one finds that

$$F_c^2 \approx \gamma/\tau_d. \tag{2.7}$$

The width of the burnt hole in this case is clearly equal to τ_d.

3. BASIC EQUATIONS. AVERAGING PROCEDURE

In the rest of this paper we present our quantitative theory for nonlinear absorption and hole burning with allowance for spectral diffusion. The treatment will be confined to lowest order in the intensity F^2.

The contribution to the absorption coefficient from a single resonant two-level system with level spacing $e \approx \hbar\omega$ is determined by the density matrix

$$\begin{pmatrix} n & -ife^{i\omega t} \\ if^* e^{-i\omega t} & 1-n \end{pmatrix}. \tag{3.1}$$

In the resonance approximation to which we limit ourselves, the matrix elements obey the system of equations

$$\frac{\partial n}{\partial t} = -\gamma(n-n_0) - F\,\mathrm{Re}\,f, \tag{3.2}$$

$$\frac{\partial f}{\partial t} + iSf + {}^1/_2\gamma f = {}^1/_2 F(2n-1), \tag{3.3}$$

where $n_0 = [\exp(e/T) + 1]^{-1}$ is the equilibrium population of the upper level of the resonant two-level system, and

$$S = \omega - e/\hbar - \Delta\omega(t) = z - \Delta\omega(t), \tag{3.4}$$

$$\Delta\omega(t) = \sum_l J_l \xi_l(t). \tag{3.5}$$

Here $\hbar J_l = |A|/r_l^3$ is the interaction energy for the resonant and the lth thermal two-level systems, separated by the distance r_l; the random time variables $\xi_l(t)$ determine a "telegrapher's" stochastic process (Ref. 13). They switch back and forth between the values 1 and -1 at random instants with an average frequency equal to Γ_l. The various functions $\xi_l(t)$ are assumed to be uncorrelated. The resonance approximation is valid when $\omega\tau_d \gg 1$.

The resonant absorption is proportional to the imaginary part of the susceptibility, for which the following expression can be derived:

$$\mathrm{Im}\,\chi(\omega) = \frac{2Q}{F}\int_0^\infty de\,\mathrm{Re}\langle\langle f\rangle_\xi\rangle_c. \tag{3.6}$$

Here $\langle\ldots\rangle_\xi$ denotes expectation with respect to the telegrapher's process, while $\langle\ldots\rangle_c$ denotes a configuration average over the spatial distribution of the thermal two-level systems.

The configuration averaging is necessary because $\mathrm{Im}\,\chi(\omega)$ is a sum of contributions from various resonant two-level systems, each with a unique thermal environment. Summation over the resonant two-level systems is thus equivalent to averaging of the thermal environments, which we denote by $\langle\ldots\rangle_c$.

Strictly speaking, the expression for $\mathrm{Im}\,\chi(\omega)$ should involve a further averaging over the tunneling parameter for the resonant two-level systems, which depends on F and γ. However, we have omitted it for simplicity since it does not significantly change the expression for $\mathrm{Im}\,\chi$.

Rewriting the differential equation (3.2) as an integral equation, substituting into (3.3), and integrating as in Ref. 7, we obtain the following result for $\mathrm{Im}\,\chi$ (including the first nonlinear correction):

$$\mathrm{Im}\,\chi(\omega) = 2\pi\hbar Q(n_0 - {}^1/_2)(1 - F^2/F_c^2), \tag{3.7}$$

where we have

$$F_c^{-2} = \int_0^\infty d\tau \int_0^\infty d\tau' \exp[-\gamma(\tau+\tau')]\langle K(\tau,\tau')\rangle_c \tag{3.8}$$

for the critical amplitude F_c, and

$$K(\tau,\tau') = \mathrm{Re}\left\langle \exp\left[i\int_0^{2\tau+\tau'} dt\,\Delta\omega(t)\vartheta(t)\right]\right\rangle, \tag{3.9}$$

$$\vartheta(t) = \begin{cases} 1, & 0 \leqslant t < \tau \\ 0, & \tau \leqslant t < \tau+\tau'. \\ -1, & \tau+\tau' \leqslant t < \tau+2\tau' \end{cases} \tag{3.10}$$

Since the functions $\xi_l(t)$ are uncorrelated, we may average over them independently to get

$$K(\tau,\tau') = \prod_l k_l(\tau,\tau'). \tag{3.11}$$

According to Refs. 6 and 7, the functions $k_l(\tau,\tau')$ are given by

$$k(\tau,\tau') = e^{-2\Gamma\tau}\left(\mathrm{ch}(\Gamma^2-J^2)^{1/2}\tau + \frac{\Gamma}{(\Gamma^2-J^2)^{1/2}}\mathrm{sh}(\Gamma^2-J^2)^{1/2}\tau\right)^2 + \frac{J^2}{\Gamma^2-J^2}\exp[-2\Gamma(\tau+\tau')]\,\mathrm{sh}^2(\Gamma^2-J^2)^{1/2}\tau. \tag{3.12}$$

This expression is valid both for $\Gamma > J$ and for $\Gamma < J$; in the latter case the hyperbolic functions are to be replaced by trigonometric ones.

It remains to average $K(\tau,\tau')$ over the parameters of the thermal two-level systems. $K(\tau,\tau')$ is a product of many expectations with respect to the telegrapher's process (3.12). Each of these expectations k_l depends on the distance r_l and the transition frequency Γ_l. We assume that there is no correlation among these parameters for distinct thermal two-level systems. This allows us to carry out the averaging independently for each thermal two-level system. If the total number of the latter is equal to N, the result is $\langle k\rangle_c^N$.

Although for most values of Γ and J, $\langle k\rangle_c$ is close to unity, $\langle k\rangle_c^N$ may differ greatly from 1 because N is large. We may use the Holtsmark method (see, e.g., Ref. 14) to evaluate $\langle k\rangle_c^N$; in essence, this reduces to the following approximation:

$$\begin{aligned} K &= \langle k\rangle_c^N = \exp N\ln\langle k\rangle_c \\ &= \exp N\ln[1 + (\langle k\rangle_c - 1)] = \exp[-N(1-\langle k\rangle_c)]. \end{aligned} \tag{3.13}$$

In evaluating $\langle k\rangle_c$ we will assume that each thermal two-level system is completely characterized by two parameters: its distance r from a resonant two-level system, and its tunneling parameter Λ. Averaging over the characteristics of the thermal two-level systems thus amounts to averaging over r and Λ. In averaging over r, we assume that all spatial configurations of the two-level systems are equally probable. The average over the tunneling parameter will be carried out by integrating with respect to $\Gamma \propto \Lambda^2$ rather than Λ. The corresponding distribution function is[15]

$$\Gamma^{-1}(1-\Gamma/\Gamma_0)^{-1/2},$$

where Γ_0 is the maximum hopping frequency for the thermal two-level systems. We may approximate the square root by unity; this simplifies the formulas somewhat without greatly affecting the results.

The method used to derive the expression for F_c^{-2} by integrating (3.8) with respect to the times τ and τ' suggests a natural physical interpretation for the latter. The τ dependence characterizes the time evolution of the off-diagonal components of the density matrix for a resonant two-level system, while the τ' dependence gives the evolution of the diagonal components.

Using (3.13) we obtain

$$\langle K(\tau,\tau')\rangle_c=\exp\left\{-QT\int d^2r\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}[1-k(\tau,\tau')]\right\}. \tag{3.14}$$

Using the fact that $d^2r=rdrd\varphi$ and making the change of variable $r\to J=|A|/r^3$ in the integral, we can rewrite (3.14) in the form

$$\langle K(\tau,\ \tau')\rangle_c=\exp[-(\tau/\tau_d)^{2/3}s(\tau,\ \tau')], \tag{3.15}$$

where

$$\tau^{2/3}s(\tau,\tau')=\int_0^\infty\frac{dJ}{J^{5/3}}\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}[1-k(\tau,\tau')], \tag{3.16}$$

$$\tau_d^{-2/3}=\frac{2\pi}{3}QT\frac{|A|}{\hbar}. \tag{3.17}$$

Although not indicated explicitly by our notation, the quantity $|A|$ in the last formula is actually an average over all angles. Up to a numerical factor ~ 1, expression (3.17) coincides with the result obtained above from order-of-magnitude estimates.

4. NONLINEAR ABSORPTION; ANALYSIS OF SOME LIMITING CASES

In terms of the new variables of integration $x=J/\Gamma$ and $y=\Gamma\tau$, (3.16) becomes

$$s=\int_0^{y_0}\frac{dy}{y^{5/3}}\int_0^\infty\frac{dx}{x^{5/3}}[1-k(\tau,\tau')], \tag{4.1}$$

where $y_0=\Gamma_0\tau$. At high temperatures $T\gg T_d$, we have $\Gamma_0\tau_d\gg 1$ and the integration over Γ in (4.1) can be extended to ∞. We conclude that the function s depends only on the ratio $\beta=\tau'/\tau$.

Recalling (3.15), we have

$$F_c^{-2}=\int_0^\infty d\tau\,\tau\int_0^\infty d\beta\exp[-(\tau/\tau_d)^{4/3}s(\beta)]\exp[-\gamma\tau(1+\beta)]. \tag{4.2}$$

We are interested in the case when the "intrinsic damping" of the resonant two-level system is small:

$$\gamma\tau_d\ll 1. \tag{4.3}$$

One expects that the regions $\tau\approx\tau_d$ and $\tau'\approx 1/\gamma$ will be significant in the integral in (4.2). That is, β values of the order of $1/\gamma\tau_d\gg 1$ should be important in the integration over β (this estimate will be made more precise below). It is shown in the Appendix that for $\beta\gg 1$

$$s(\beta)=b\ln\beta,\qquad b=\frac{3}{2^{8/3}}\Gamma\left(\frac{1}{3}\right). \tag{4.4}$$

Substitution in (4.2) and integration over β yields

$$F_c^{-2}=\frac{1}{\gamma}\int_0^\infty d\tau\exp\left[-\left(\frac{\tau}{\tau_d}\right)^{2/3}b\ln\frac{1}{\gamma\tau}\right].$$

Here we have approximated the slowly (logarithmically) varying factor $s(\beta)$ in the argument of the first exponential by its value at $\beta=1/\gamma\tau$ and taken it outside the integral. The integration over τ can be performed using the same approximation to obtain the result

$$F_c^2=\frac{4}{3\pi^{1/2}}\frac{\gamma}{\tilde{\tau}_d}, \tag{4.5}$$

where

$$\tilde{\tau}_d=\frac{\tau_d}{(b\ln(\gamma\tau_d)^{-1})^{3/2}}. \tag{4.6}$$

The condition for (4.5) to be valid is that

$$\gamma\ll 1/\tilde{\tau}_d, \tag{4.7}$$

which is a refinement of relation (2.1).

At low temperatures $T\ll T_d$ we have $\Gamma_0\ll 1/\tau_d$ and the function (3.15) decays with a characteristic time of the order of τ_φ, and for these times we have $y_0=\Gamma_0\tau_\varphi$. The function s for this case is calculated in the Appendix and found to be

$$s=b\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}(1-e^{-2\Gamma\tau'}). \tag{4.8}$$

Expression (4.2) now depends on the three characteristic frequencies γ, Γ_0, and τ_d^{-1}; the latter is more conveniently replaced by the combination $\tau_d^{-2/5}\Gamma_0^{3/5}=1/\tau_\varphi\gg\Gamma_0$. There are thus three limiting cases. If $\gamma\gg 1/\tau_\varphi$, most of the contribution to the integral (4.2) is from $\tau,\tau'\sim 1/\gamma$, and $F_c=\gamma$. There is little spectral diffusion in this case.

If

$$\Gamma_0\ll\gamma\ll 1/\tau_\varphi,$$

the integral is readily found by expanding the first exponential; to within a factor of the order of unity this gives the estimate (2.5) $F_c=1/\tau_\varphi$. If however

$$\gamma\ll\Gamma_0\ll 1/\tau_\varphi,$$

then in our approximation

$$\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}(1-e^{-2\Gamma\tau'})\approx\ln\Gamma_0\tau'$$

and

$$F_c^2=\frac{4b^{3/2}}{3\pi^{1/2}}\frac{\gamma}{\tau_d}\left(\ln\frac{\Gamma_0}{\gamma}\right)^{3/2}. \tag{4.9}$$

Here the argument of the logarithm differs from that in (4.5).

5. THEORY OF HOLE BURNING

We consider the situation when a signal of amplitude F and frequency ω propagates in glass and it is desired to measure the absorption of a second, weak signal of amplitude $F_1\ll F$ and frequency ω_1, where $\nu=\omega_1-\omega$ satisfies the inequality $|\nu|\ll\omega$. To first order in F^2, we obtain the following expression for the power attenuation of the weak signal per unit volume due to absorption[6,7]:

$$-\Delta q=B\int_0^\infty d\tau'\, e^{-\gamma\tau'}\int_0^\infty d\tau\, e^{-\gamma\tau/2}\cos\nu\tau\langle L(\tau,\tau')\rangle_c, \qquad (5.1)$$

where

$$L(\tau,\tau')=\left\langle \exp\left[i\Delta\omega|_{\tau=0}\tau-i\int_{\tau'}^{\tau+\tau'} dt\,\Delta\omega(t)\right]\right\rangle_\xi, \qquad (5.2)$$

$$B=\frac{\pi}{2}\omega F_1^2F^2Q\,\mathrm{th}\frac{\hbar\omega}{2T}. \qquad (5.3)$$

The same method used above to evaluate (3.9) can be used to calculate the function (5.3). We omit the details and state the result:

$$\langle L(\tau,\tau')\rangle_c=\exp\left\{-QT\int d^2r\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}[1-l(\tau,\tau')]\right\}, \qquad (5.4)$$

$$l(\tau,\tau')=\cos J\tau\, e^{-\Gamma\tau}\left[\mathrm{ch}(\Gamma^2-J^2)^{1/2}\tau + \frac{\Gamma}{(\Gamma^2-J^2)^{1/2}}\mathrm{sh}(\Gamma^2-J^2)^{1/2}\tau\right] + \sin J\tau\, e^{-2\Gamma\tau'-\Gamma\tau}\frac{J}{(\Gamma^2-J^2)^{1/2}}\mathrm{sh}(\Gamma^2-J^2)^{1/2}\tau. \qquad (5.5)$$

Switching to J as the variable of integration, we can rewrite (5.4) in the form

$$\langle L(\tau,\ \tau')\rangle_c=\exp[-(\tau/\tau_d)^{2/3}v(\tau,\ \tau')], \qquad (5.6)$$

where

$$v=\int_0^{y_0}\frac{dy}{y^{5/3}}\int_0^\infty\frac{dx}{x^{5/3}}[1-l(\tau,\tau')]. \qquad (5.7)$$

The rest of the analysis is essentially as in Sec. 3, but slightly more complicated because the function to be determined now depends on the frequency ν.

We begin once again with the high-temperature case, $T\gg T_d$. Using the integration variable $\beta=\tau'/\tau$ in (5.1), we see that for $\beta\gg 1$ we have

$$v(\beta)=b\ln\beta \qquad (5.8)$$

in complete analogy with the function $s(\beta)$. The values $\beta\approx 1/\gamma\tau$ give the primary contribution to the integral over β. On the other hand for small ν, τ is typically $\sim\tau_d$ and these values can only increase with ν; the representative values of β are thus indeed much greater than unity. In evaluating the integral (5.1) we may therefore replace the $v(\beta)$ by its asymptotic expression (5.8).

To within our approximation we then obtain

$$-\Delta q=\frac{B}{\gamma}\int_0^\infty d\tau\cos\nu\tau\exp\left[-\left(\frac{\tau}{\tau_d}\right)^{2/3} b\ln\frac{1}{\gamma\tau}\right] \qquad (5.9)$$

for the shape of the hole. To the same accuracy, we have the asymptotic expressions

$$-\Delta q=\frac{B}{\gamma}\begin{cases}\dfrac{3\pi^{1/2}}{4}\tilde{\tau}, & \nu\ll 1/\tilde{\tau}\\ \dfrac{5}{6}\Gamma\left(\dfrac{2}{3}\right)\tau_\nu(\nu\tau_\nu)^{-5/3}, & \nu\gg 1/\tilde{\tau}\end{cases}, \qquad (5.10)$$

where $\tau_\nu=\tau_d\ln(\nu/\gamma)$. The hole width is thus found to be $\sim\tau_d$ (up to a logarithmic factor), in agreement with the order-of-magnitude estimates in Sec. 2.

The asymptotic relation $-\Delta q\propto\nu^{-5/3}$ can be deduced on simple physical grounds. The profile near the edges of the hole is determined by the contribution from a relatively small number of the resonant two-level systems, those whose distance r from the thermal systems is less than the average value. Specifically, the shape of the edge at the frequency ν is determined by the two-level systems with $J=\nu$, i.e., for which $r\propto\nu^{-1/3}$. Their number is proportional to

$$r\,dr\propto\nu^{-1/3}d\nu^{-1/3}\propto\nu^{-5/3}d\nu.$$

This result is completely general and independent of the relative magnitudes of the parameters γ, Γ_0, and $1/\tau_\varphi$. Thus in the two-dimensional case, unlike the case of three dimensions, the hole cannot be Lorentzian.

For $T\ll T_d$, a method similar to that described in the Appendix leads to the expression

$$-\Delta q=B\int_0^\infty d\beta\int_0^\infty d\tau\,\tau e^{-\gamma\tau(\beta+1/2)}\cos\nu\tau\exp[-(\tau/\tau_d)^{2/3}v], \qquad (5.11)$$

where

$$v=b\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}(1-e^{-2\Gamma\tau'}). \qquad (5.12)$$

There are two limiting cases. For

$$1/\tau_\varphi\gg\gamma\gg\Gamma_0,\ \text{where}\ \tau_\varphi=\tau_d^{2/5}\Gamma_0^{-3/5}, \qquad (5.13)$$

the shape of the hole is given by the expression

$$\Delta q=-B\tau_\varphi^2\mathscr{F};\quad \mathscr{F}=\int_0^\infty d\xi\frac{\cos\nu\tau_\varphi\xi}{\gamma\tau_\varphi+2\xi^{2/3}}e^{-\xi^{5/3}}. \qquad (5.14)$$

Up to coefficients of order 1, the following limiting expressions for the integral $\mathscr{F}$ are valid:

$$\mathscr{F}=\begin{cases}1, & \nu\tau_\varphi\ll 1\\ (\nu\tau_\varphi)^{-1/3} & 1\ll\nu\tau_\varphi\ll(\gamma\tau_\varphi)^{-3/2}.\\ (\gamma\tau_\varphi)^{-2}(\nu\tau_\varphi)^{-5/3}, & \nu\tau_\varphi\gg(\gamma\tau_\varphi)^{-3/2}\end{cases} \qquad (5.15)$$

In this case the shape of the "hole" is quite complicated and it is thus necessary to agree on what is meant by its width $\Delta\nu$. It is natural to take $\Delta\nu$ to be the interval of ν values for which (5.14) is appreciable when the latter is integrated over ν. It turns out that these values coincide with the estimate (2.6), and also with the region of transitional ν for which the second asymptotic expression in (5.15) is replaced by the third.

If

$$1/\tau_\varphi\gg\Gamma_0\gg\gamma \qquad (5.16)$$

we evaluate the integral (5.12) using the following approximation:

$$\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}(1-e^{-2\Gamma\tau'})=\ln\frac{1}{2\Gamma_0\tau'}.$$

The profile of the "hole" is given by

$$-\Delta q=\frac{B}{\gamma}\int_0^\infty d\tau\cos\nu\tau\exp\left[-\left(\frac{\tau}{\tau_d}\right)^{2/3}b\ln\frac{1}{\Gamma_0\tau}\right]. \quad (5.17)$$

This expression differs from (5.9) in that γ in the logarithm is replaced by Γ_0; the asymptotic expressions (5.10) must clearly be modified in the same way.

6. THE NONLINEAR LIMIT

We will content ourselves with a qualitative description of the strong nonlinearity at high intensities $F \gg F_c$. The power absorbed per unit volume is given by (cf. Refs. 6, 7)

$$q=\hbar Q\omega\gamma\int_0^\infty de\langle n-n_0\rangle_t, \quad (6.1)$$

where $\langle ... \rangle_t$ denotes a time average.

We consider the case of large spectral diffusion (the opposite limit of negligible spectral diffusion has been analyzed in detail using the Bloch equations; see Ref. 3). The qualitative analysis is quite similar to the treatment in Refs. 6, 7.

The nonlinear behavior of q is determined by the length of the interval of e values over which the integration is performed (it is equal to the width of the burnt hole) and by the value $\langle n-n_0\rangle_t$ averaged over this interval. The above heuristic arguments regarding the width of the hole are easily seen to hold for arbitrary intensities. In other words, the hole width (although not, in general, its shape) is independent of the intensity whenever spectral diffusion is important. In fact, this result remains valid as long as the Rabi oscillation frequency F is less than the value $\Delta\nu$ calculated in the linear theory.[16–18]

Furthermore, the difference $\langle n-n_0\rangle_t$ for e in the interval corresponding to the burnt hole must also be independent of the intensity at high intensities. For $\gamma \ll \Gamma_0 \ll 1/\tau_\varphi$ we have

$$\langle n-n_0\rangle_t \approx {}^1/_2-n_0 \quad (6.2)$$

for both high ($T \gg T_d$) and low temperatures ($T \ll T_d$). By contrast, for $\Gamma_0 \ll \gamma \ll 1/\tau_\varphi$ the population of a resonant two-level system within a resonance band of width $\hbar/\tau_\varphi$ changes during a time of the order of τ_φ. The system remains excited for a time $\gamma^{-1} \gg \tau_\varphi$ and then emits a phonon. The resonant two-level system spends the remainder of the time in a region of width $\hbar/\tau_d$, but its population is equal to n_0. The system returns to the resonance interval after a time of the order of $\Gamma_0^{-1} \gg \gamma^{-1}$. The fraction of the time during which the resonant two-level system is excited (i.e., $n = 1/2$) is thus of the order of $\Gamma_0/\gamma \ll 1$. We therefore have

$$\langle n-n_0\rangle_t = ({}^1/_2-n_0)\,(\Gamma_0/\gamma) \quad (6.3)$$

in the present case. In all cases $\langle n-n_0\rangle_t$ is thus found to be independent of the intensity, and the same is therefore true of the absorbed power (6.1). This in turn implies that the absorption coefficient is inversely proportional to the intensity at high intensities:

$$\alpha \approx \alpha_0 (F_c/F)^2. \quad (6.4)$$

This conclusion holds universally, provided one uses the expression for F_c appropriate to any given limiting case.

On the other hand, if spectral diffusion is negligible it follows immediately from an analysis of the equations for the density matrix[3] that the hole width is proportional to the amplitude, while the absorption coefficient is inversely proportional to it.

We stress once again that all the conclusions reached above are valid only if the phonon distribution function at the resonance frequency can be approximated by an equilibrium distribution. We have already pointed out that this condition is nearly always satisfied for the cases mentioned in the Introduction, because most of the phonons must leave the two-dimensional region in which they are generated. An exception may occur for free-standing films,[19] for which there is no place for the phonons to escape. The theory for such films requires modification in any event, because the interaction among the two-level systems must differ in the purely two-dimensional case.

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APPENDIX

The integral for the function $s(y_0, \beta)$ can be recast in a form better suited for calculation. We have $s = s_1 + s_2$:

$$s_1=\int_0^{y_0}\frac{dy}{y^{5/3}}\int_0^1\frac{dx}{x^{5/3}}(1-\varkappa_1); \qquad s_2=\int_0^{y_0}\frac{dy}{y^{5/3}}\int_1^\infty\frac{dx}{x^{5/3}}(1-\varkappa_2), \quad (A1)$$

$$\varkappa_1(x, y; \beta)=e^{-2y}\left(\operatorname{ch} y(1-x^2)^{1/2}+\frac{1}{(1-x^2)^{1/2}}\operatorname{sh} y(1-x^2)^{1/2}\right)^2 + \frac{x^2}{1-x^2}e^{-2y(1+\beta)}\operatorname{sh}^2 y(1-x^2)^{1/2}, \quad (A2)$$

$$\varkappa_2(x, y; \beta)=e^{-2y}\left(\cos y(x^2-1)^{1/2}+\frac{1}{(x^2-1)^{1/2}}\sin y(x^2-1)^{1/2}\right)^2 + \frac{x^2}{x^2-1}e^{-2y(1+\beta)}\sin^2 y(x^2-1)^{1/2}. \quad (A3)$$

At high temperatures $T \gg T_d$ the upper limit y_0 may be replaced by ∞. Since, as noted in Sec. 4, large β give the dominant contribution to the integral (4.2), the behavior of $s(\beta)$ for $\beta \gg 1$ is of interest. It is easily seen that the expression for s_1 remains finite and ~ 1 as $\beta \to \infty$, but that s_2 diverges logarithmically. The term s_2 thus gives the main contribution for $\beta \gg 1$.

We may write

$$s_2(\beta)=s_2(0)+\Delta s_2(\beta).$$

Again, $s_2(0)$ turns out to be ~ 1, and

$$\Delta s_2(\beta)=\int_0^\infty\frac{dy}{y^{5/3}}e^{-2y}(1-e^{-2\beta y})\mathscr{G}, \quad (A4)$$

$$\mathscr{G}=\int_1^\infty\frac{dx}{x^{5/3}}\frac{x^2}{x^2-1}\sin^2 y(x^2-1)^{1/2}. \quad (A5)$$

The integral (A5) is conveniently evaluated by making the change of variable $z = y(x^2-1)^{1/2}$:

$$\mathscr{G}=\int_0^\infty\frac{dz\sin^2 z}{z(1+z^2/y^2)^{1/3}}; \quad (A6)$$

for $y \ll 1$ we have $\mathscr{G} = by^{2/3}$. Insertion in (A4) gives

$$s=b\ln\beta. \tag{A7}$$

For $T \ll T_d$ the characteristic values of y_0 are $\ll 1$. In this case we evaluate s_1 by expanding the integrand as a series in powers of x and y:

$$\varkappa_1=1-2x^2y^3(\beta+{}^2/_3),\quad s_1={}^9/_{14}y_0{}^{7/3}(\beta+{}^2/_3).$$

Thus once again, s_1 is found to be small compared to s_2. The latter quantity can be calculated by the same change of variable as above, and we find

$$s=s_2=b\int\limits_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}(1-e^{-2\Gamma\tau'}). \tag{A8}$$

Essentially the same method can be used to calculate the function $v(\beta)$ in the various limiting cases.

[1)] This idea was suggested by S.Hunklinger (Heidelberg), to whom the author is deeply grateful.

[2)] Given the crudeness of these order-or-magnitude estimates, we do not distinguish between the densities and speeds of sound in the amorphous material and in the crystal substrate.

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