Spectral diffusion and phonon bottleneck in glasses

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A theory is derived for the nonlinear resonant absorption of sound and of a microwave field in dielectric glasses at low temperatures and in the steady state. The nonequilibrium nature of the phonon subsystem is taken into account. Spectral diffusion changes the phonon bottleneck substantially from that in the case without diffusion because the nonequilibrium phonon distribution spreads out over the spectrum. The width of the phonon spectral distribution is proportional to the phonon lifetime τ_{nr} with respect to the nonresonant absorption by two-level systems. The critical intensity characterizing the onset of the nonlinear behavior of the absorption coefficient depends weakly (logarithmically) on τ_{nr} as $\tau_{nr} \to \infty$. The shape of the burned-out hole is analyzed. In the case of weak pumping, this shape is the same as that of the distribution of nonequilibrium phonons.

1. INTRODUCTION

Many low-temperature properties of glasses stem from the existence of so-called two-level systems in the glasses.[1] In the present paper we are interested in the nonlinear resonant absorption and the burned-out hole in dielectric glasses in the case in which the nonequilibrium nature of the phonon subsystem (the phonon bottleneck) is important. Gal'perin *et al.*[2] have derived a theory for these effects in the steady state, ignoring the accumulation of resonant phonons.

One of the most interesting features of the low-temperature kinetics in dielectric glasses is that spectral diffusion plays an important role. This effect was first discussed by Klauder and Anderson[4] in a theory of magnetic resonances. Related ideas have been used in theoretical work on the low-temperature properties of glasses by Joffrin and Levelut,[5] Hunklinger and Arnold,[6] Black and Halperin,[7] and Laikhtman.[8,9] An important point to be noted is that spectral diffusion is manifested only in nonlinear effects. Included among these effects are two- and three-pulse echos,[7,10,11,8] the nonlinear resonant absorption of sound and microwave power,[6,8,12] and the burned-out hole.[6,8]

Spectral diffusion can be summarized as follows: Each two-level systems creates a strain field around itself. The magnitude of this strain depends on the particular energy state (upper or lower) of the given two-level system. As we will see below, the most important two-level systems are the so-called thermal two-level systems, with an energy (a distance between levels) $E \lesssim T$. Under the influence of thermal phonons, these systems are continually undergoing transitions from one state to the other. The strain field which they create in their vicinity thus fluctuates over time.

In turn, the energy of any two-level system changes in a strain field. Transitions in thermal two-level systems near a given two-level system thus cause this energy to also fluctuate over time. The scale of these fluctuations can be estimated quite easily. We are not interested in the static part of the strain, since in a glass there is a wide spread in the energies of the two-level systems, with a roughly uniform state density.

A two-level system may be thought of as an elastic dipole. The strain field which it creates in its vicinity is given by

$$u_{ik} \approx D/\rho v^2 r^3.$$

Here D is the strain energy, ρ is the density of the glass, v is the sound velocity, and r is the distance from the two-level system. The sign of this strain depends on the sign of the strain-energy constant D and also on the particular state—upper or lower—of the given two-level system. In a transition, the sign of the strain reverses (the orientation of the elastic dipole changes).

A strain which fluctuates in time is caused by thermal two-level systems. Their concentration is on the order of PT, where P is the constant, energy-independent, state density of the two-level systems in the glass. The scale of the fluctuations, Δu_{ik}, can be found if we take r to be the average distance between thermal two-level systems. As a result we find $\Delta u_{ik} \approx DPT/\rho v^2$. The characteristic change in the energy of the two-level system, E_d, upon a characteristic change in the strain, Δu_{ik}, is

$$E_d \approx D\Delta u_{ik} \approx D^2 PT/\rho v^2. \quad (1)$$

In typical glasses, the dimensionless parameter $D^2P/\rho v^2$ has values on the order of 1/200 to 1/300 (Refs. 1 and 6). It is this random change with time in the energy of a two-level system due to interactions with other two-level systems that is called "spectral diffusion."

In this paper we analyze the effect of spectral diffusion on the distribution of nonequilibrium acoustic phonons generated by a coherent monochromatic signal in a glass. We also examine the effect of these phonons on the nonlinear resonant absorption and the shape of the burned-out hole. Some of the results of this study were summarized in Ref. 13.

Gurevich and Rzaev[14] have analyzed this problem without spectral diffusion.

2. QUALITATIVE PICTURE

At low temperatures, resonant two-level systems with an energy $e = \hbar\omega$, where ω is the signal frequency, are responsible for the resonant absorption of a sound or microwave field in a dielectric glass. As a rule, when a resonant two-level system absorbs a quantum of the sound or the mi-

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crowave field, $\hbar\omega$, it then emits a phonon of the same, or nearly the same, energy. The subsequent fate of these resonant phonons depends on the relation between two times: τ_r and τ_{nr}. The first of these times is the lifetime of a phonon with respect to its resonant absorption by a two-level system with the same energy[1]:

$$\frac{1}{\tau_r}=\frac{\pi P M^2}{\rho v^2}\Omega \operatorname{th}\frac{\hbar\Omega}{2T}. \tag{2}$$

Here Ω is the phonon frequency, and M the transition matrix element.

The second time is the phonon lifetime with respect to its nonresonant absorption by two-level systems with an energy on the order of[15] $\max(T,\hbar\Omega)$:

$$\frac{1}{\tau_{nr}}=\frac{\pi^3 P M^2 D^2 T^4}{16\rho^2\hbar^5 v^7\Omega}\left[1+\frac{16}{3\pi^2}\left(\frac{\hbar\Omega}{2T}\right)^2+\frac{16}{3\pi^4}\left(\frac{\hbar\Omega}{2T}\right)^4\right]\operatorname{th}\frac{\hbar\Omega}{2T}. \tag{3}$$

At frequencies in the classical region, $\Omega \ll T$, this expression is the same as that found by Jäckle.[16]

The times t_r and t_{nr} play different roles in the kinetics of resonant phonons. After being absorbed in a resonant process, a phonon excites a two-level system with the same energy. After a certain time, this two-level system reverts to the ground state, emitting a phonon of the same, or nearly the same, energy. This process does not change the number of resonant phonons.

In contrast, after being absorbed in a nonresonant way by a two-level system, with an energy significantly different from the phonon energy, a resonant phonon irreversibly leaves the resonant group. To complete the picture we should add that a phonon may also leave the resonant group after its resonant absorption by a two-level system. This event occurs if the two-level system excited by a resonant phonon goes to the ground state by a two-phonon process (emitting two phonons or emitting one and absorbing another). Although the probability for such a process is substantially lower than that for the one-phonon process, it plays an important role, along with nonresonant absorption, in establishing the phonon temperature in glasses at low temperatures.[17] We will show that in the problem of the present paper this process can be ignored in the classical frequency region, $\hbar\omega \ll T$. In the quantum frequency region, $\hbar\omega \gg T$, it makes a contribution on the same order of magnitude as that of nonresonant absorption to the rate at which phonons leave the resonant region.

The ratio of the two relaxation times, τ_{nr}/τ_r, is

$$\frac{\tau_{nr}}{\tau_r}=\frac{16}{\pi^2}\frac{(\hbar\Omega)^2 E_c^2}{T^4}\left[1+\frac{16}{3\pi^2}\left(\frac{\hbar\Omega}{2T}\right)^2+\frac{16}{3\pi^4}\left(\frac{\hbar\Omega}{2T}\right)^4\right]^{-1}, \tag{4}$$

where $E_c=(\rho v^5\hbar^3)^{1/2}/D$ is a characteristic energy on the order of 10–30 K in glasses.[15] Under the conditions $T \ll E_c$ and $\hbar\Omega \gg T^2/E_c$, the relation $\tau_{nr} \gg \tau_r$ holds even in the classical frequency region. The ratio in (4) reaches its maximum value $\approx 6.4(E_c/T)^2$ in the quantum region, at $\hbar\Omega \approx 11T$. As Ω is increased further, the ratio $\tau_{nr}\tau_r$ decreases, remaining much greater than unity as long as the concept of a two-level system remains applicable.[15,18]

Under the condition $\tau_{nr} \gg \tau_r$, resonant phonons with a frequency $\Omega \approx \omega$ are reabsorbed by resonant two-level systems many times before they leave the resonant group. In this case, there is the possibility that these phonons will accumulate. During intense pulsed excitation, this effect gives rise to the phenomenon of a bottleneck in nonlinear resonant absorption.[9]

As we will show below, however, in the case of weak steady-state excitation the accumulation of phonons in a narrow resonant region is reduced by spectral diffusion. Although resonant phonons are reabsorbed many times by resonant two-level systems during the lifetime τ_{nr}, the changes in the energy of two-level systems which result from transitions (jumps) in thermal two-level systems and which occur randomly in time lead to the result that the emitted phonon does not have the same frequency as that which was absorbed. This effect in turn causes a nonequilibrium phonon distribution to undergo spectral spreading. It changes the phonon-bottleneck phenomenon greatly from that in the case without spectral diffusion.

Under the condition $\tau_{nr} \gg \tau_r$, and in the absence of spectral diffusion, phonons accumulate in a narrow resonant region with a width on the order of the spectral width of the pump signal. We will assume below that this width is zero. The threshold for the nonlinearity in the resonant absorption (i.e., the intensity at which a saturation is manifested in the absorption) turns out to be lower in this case by a factor of τ_{nr}/τ_r than in the case without an accumulation of phonons.[14,19–22]

In a situation with spectral diffusion, the phonons spread out over the spectrum to a great extent in most of the cases which have been considered. As a result, we find that under the condition $\tau_{nr} \gg \tau_r$ the threshold (critical) intensity is inversely proportional not to the ratio τ_{nr}/τ_r but only to its logarithm.

The spreading of phonons over the spectrum due to spectral diffusion also has a great effect on the width of the burned-out hole. The "burned-out hole" is the decrease in the absorption coefficient for a weak test signal at the frequency ω_1 in the presence of a pump signal (usually strong) at the frequency ω (Ref. 23). Here we show that in the case of weak steady-state excitation the shape of the burned-out hole (as a function of the frequency difference $\omega_1-\omega$) reproduces the shape of the nonequilibrium phonon distribution function (which depends on $\omega-\Omega$, where Ω is the phonon frequency). If the phonons are able to accumulate, the width of the burned-out hole is increased by the spectral diffusion of phonons to a magnitude τ_{nr}/τ_r greater than that in the case without phonon accumulation.[13]

This "nondiffusion" result is explained on the basis that the width of the phonon distribution is determined primarily by comparatively rare collisions of resonant phonons with so-called quasiresonant two-level systems, near which, within a distance $r \ll (PT)^{-1/3}$, is a thermal two-level system. A quasiresonant two-level system spends only half its time in resonance, until the neighboring thermal two-level system goes into the other state. Because of the proximity of the thermal two-level system, the energy of the quasiresonant two-level system changes at once by a large amount—significantly greater than E_d—upon such a transition. Accordingly, after absorbing a resonant phonon a quasiresonant two-level system can emit a phonon which is far from resonance.

Let us examine the most important parameters which arise in the solution of the problem. As we have already men-

tioned, jumps (transitions) in neighboring thermal two-level systems cause the energy e of a resonant two-level system to vary in a random way over time. The time scale of this variation is on the order of E_d as given by (1), i.e., on the order of the interaction energy of two thermal two-level systems separated by an average distance $\bar{r} = (PT)^{-1/3}$. We introduce the quantity $\tau_d = \hbar/E_d$, with the dimensions of time, which we will need below. As we will see, the relation between $1/\tau_d$ and the characteristic frequency of the jumps of thermal two-level systems will play an important role throughout the phenomenon of spectral diffusion. This characteristic jump frequency Γ_0 is given by[6]

$$\Gamma_0 \approx M^2 T^3/\rho\hbar^4 v^5. \tag{5}$$

The appearance of a dimensionless parameter $\Gamma_0\tau_d$ in the theory can be explained in the following way. At early times, $t \ll \Gamma_0^{-1}$, the energy of a resonant two-level system deviates from resonance linearly with time[4,7]:

$$|e(t)-e(0)| \approx \hbar\Gamma_0 t/\tau_d. \tag{6}$$

This formula has the following origin. We consider a volume with linear dimensions of order r_t around a resonant two-level system. This volume contains $\approx PTr_t^3$ thermal two-level systems with characteristic transition frequencies on the order of Γ_0. A transition of at least one thermal two-level system in this volume by the time t occurs with a probability of order unity if r_t satisfies the condition $\Gamma_0 tPTr_t^3 \approx 1$. The characteristic change in the energy of a resonant two-level system corresponding to this jump is

$$D^2/\rho v^2 r_t^3 \approx \hbar\Gamma_0 t/\tau_d.$$

We thus arrive at (6).

It follows from (6) that the time scale for the phase relaxation of the wave function of a resonant two-level system, τ_φ, is[9]

$$\tau_\varphi \approx (\tau_d/\Gamma_0)^{1/2}. \tag{7}$$

Expression (7) holds if this time is much shorter than the characteristic time between jumps, $1/\Gamma_0$, i.e., if

$$\Gamma_0\tau_d \ll 1. \tag{8}$$

Under the condition $\Gamma_0\tau_d \gg 1$, the phase of the wave function of a resonant two-level system can change by no more than a small amount over a time $t \approx 1/\Gamma_0$; i.e., there is not enough time for a disruption of this phase. The time scale of the phase disruption thus satisfies $\tau_\varphi \gg \Gamma_0^{-1}$. After a long time $t \gg \Gamma_0^{-1}$, on the other hand, the characteristic value of the deviation will cease to depend on the time, since the difference $|e(t) - e(0)|$ cannot exceed the characteristic energy $\hbar/\tau_d$ in order of magnitude. In other words, the deviation in this case undergoes a random walk over an interval of $\hbar/\tau_d$. Correspondingly, the phase disruption time τ_φ is determined by the spectral width of this interval and is given in order of magnitude by $\tau_\varphi \approx \tau_d \gg \Gamma_0^{-1}$.

It is clear from this discussion that there exist two regions, of high and low temperatures in comparison with the characteristic temperature T_d. The latter is found by equating the characteristic parameter $\Gamma_0\tau_0$ to unity. We have

$$T_d = (P\hbar^3 v^3)^{1/2}. \tag{9}$$

This temperature was introduced in Refs. 9 and 24. Its typical value for dielectric glasses is about 1 K. This value is found if we take P to be the state density determined from data on the specific heat.[1] If we instead experimental data on the absorption of sound,[1] we find the value $T_d \approx 0.1$ K.

3. BASIC EQUATIONS

In the resonant approximation, the system of equations for the components of the density matrix of the sth resonant two-level system,

$$\begin{pmatrix} n_s & -if_s e^{i\omega t} \\ if_s^* e^{-i\omega t} & 1-n_s \end{pmatrix}, \tag{10}$$

and for the phonon distribution function N_k ($\mathbf{k}$ is the phonon wave vector) is[1)]

$$\frac{\partial n_s}{\partial t} = \frac{2\pi}{\hbar}\sum_{\mathbf{k}} \Lambda_k [N_k - n_s(2N_k+1)]\,\delta(e_s(t) - \hbar\Omega_k) - F\,\mathrm{Re}\, f_s - \frac{n_s - n_s^0}{\tau'}, \tag{11a}$$

$$\frac{\partial f_s}{\partial t} + i\left(\omega - \frac{e_s(t)}{\hbar}\right) f_s + f_s \frac{\pi}{\hbar}\sum_{\mathbf{k}} \Lambda_k (2N_k+1)\,\delta(e_s(t) - \hbar\Omega_k) = F\left(n_s - \frac{1}{2}\right), \tag{11b}$$

$$\frac{\partial N_k}{\partial t} + \frac{2\pi}{\hbar}\sum_s \Lambda_k [N_k(1-2n_s) - n_s]\,\delta(e_s(t) - \hbar\Omega_k) = -\frac{N_k - N_k^0}{\tau_{nr}}. \tag{11c}$$

Here $\hbar F/2$ is the transition matrix element which characterizes the interaction of the (acoustic or microwave) signal of frequency ω with the resonant two-level system;

$$N_k^0 = [\exp(\hbar\Omega_k/T) - 1]^{-1}$$

is the equilibrium distribution function of the phonons; Ω_k is the phonon frequency;

$$\Lambda_k = \hbar k^2 M^2/2\rho V\Omega_k$$

is the square of the matrix element of the interaction of the two-level system with the phonons; V is the volume;

$$n_s^0 = [\exp(e_s/T) + 1]^{-1}$$

is the equilibrium filling of the upper level of the resonant two-level system; and τ' is the time scale of the relaxation of the population of the resonant two-level system as a result of two-phonon processes. An expression for this time is given in the Appendix [expression (104)]. The time-dependence between the levels of the sth resonant two-level system is

$$e_s(t) = e_s + \hbar\Delta\omega_s(t), \tag{12}$$

where

$$\Delta\omega_s(t) = \sum_l J_l \xi_l(t) \tag{13}$$

is that increment in the bare energy e_s which is caused by the interaction of the resonant two-level system with the nearby

thermal two-level systems (the summation is over all the thermal two-level systems). We can then write

$$J_l = D^2/\hbar\rho v^2 r_l^3,$$

where r_l is the distance from the lth thermal two-level system to the given resonant two-level system. Here we have a characteristic energy $E_d \sim J(\bar{r})$. The function $\xi_l(t)$ is a random function of the time, which is described by a "telegraphic" process. This function alternately takes on the values $+1$ and -1 at random times; the average frequency of these jumps is Γ_l. We assume that the different functions ξ_l are uncorrelated.

The absorption of an alternating field by a resonant two-level system is determined by the imaginary part of the susceptibility, $\mathrm{Im}\,\chi_s(\omega)$, which is related to the nondiagonal component f_s of the density matrix by

$$\mathrm{Im}\,\chi_s(\omega) = -\frac{2}{F}\,\mathrm{Re}\langle f_s\rangle_\xi. \tag{14}$$

Here the angle brackets mean an average over realizations of all the telegraph processes $\xi_l(t)$. This average is equivalent to an average over the time. The total susceptibility is determined by the sum of the contributions of the type (14) from all the resonant two-level systems in a unit volume:

$$\mathrm{Im}\,\chi(\omega) = -\frac{2}{VF}\sum_s \mathrm{Re}\langle f_s\rangle_\xi. \tag{15}$$

We assume that the arrangement of the thermal two-level systems and their transition frequencies Γ_l are uncorrelated with the parameters of the resonant two-level system. In this case we can write

$$\mathrm{Im}\,\chi(\omega) = -\frac{2P}{F}\int_0^\infty de_s\,\mathrm{Re}\langle\langle f_s\rangle_\xi\rangle_c. \tag{16}$$

Here $\langle\ldots\rangle_c$ means a configurational average over the positions of the thermal two-level systems and the values of their tunneling transparency, on which the transition frequencies Γ_l depend.

Actually, we should have also taken an average over the tunnel transparencies of the resonant two-level systems, on which the quantities F and M depend.[25] However, one can verify that this averaging contributes nothing which is fundamentally new, and we will omit it in order to keep the equations from becoming too complicated.

4. SUCCESSIVE ITERATIONS IN THE AMPLITUDE OF THE PUMP FIELD, F

Assuming that the pump amplitude is small, we solve the system of equations (11) by taking successive iterations in F:

$$n_s = n_s^0 + \Delta n_s,\ \Delta n_s \sim F^2,\ N_k = N_k^0 + \Delta N_k,\ \Delta N_k \sim F^2,$$
$$f_s = f_s^{(1)} + f_s^{(3)},\quad f_s^{(1)} \sim F,\quad f_s^{(3)} \sim F^3. \tag{17}$$

From the last equation of system (11), for example, we find the relationship between the correction to the phonon distribution function, ΔN_k, and the average change in the population of the upper level of resonant two-level systems, $\Delta N(\Omega)$:

$$\Delta N_k = \frac{\tau_{nr}}{\tau_r + \tau_{nr}}\,\Delta n(\Omega_k)\,\mathrm{cth}^2\frac{\hbar\omega}{2T}, \tag{18}$$

where

$$\Delta n(\Omega) = P^{-1}V^{-1}\hbar^{-1}\sum_s \Delta n_s\,\delta(e_s(t)/\hbar - \Omega). \tag{19}$$

The function $\Delta N(\Omega)$ describes the burned-out hole. Specifically, the change ΔQ in the absorption of a weak test signal of frquency ω_1 in the presence of a pump signal which causes a change in population Δn_s is

$$\Delta Q = -\frac{\pi\omega_1}{\hbar}F_1^2\sum_s \Delta n_s\,\delta(e_s(t)/\hbar - \omega_1) = -\pi PV\omega_1 F_1^2\,\Delta n(\omega_1), \tag{20}$$

where F_1 is the amplitude of the test signal. In the case of weak pump, the shape of the distribution function of the nonequilibrium phonons thus reproduces the shape of the burned-out hole.

Substituting $n_s = n_s^0$ and $N_k = N_k^0$, into (11b), we find an equation for $f_s^{(1)}$. Its solution is

$$f_s^{(1)} = -\frac{F}{2}\,\mathrm{th}\frac{\hbar\omega}{2T}\int_0^\infty d\tau\exp\Big\{-\frac{\gamma}{2}\tau - i\Big[z_s\tau - \int_{t-\tau}^{t}\Delta\omega_s(t')\,dt'\Big]\Big\}, \tag{21}$$

where $z_s = \omega - e_s/\hbar$, and γ^{-1} is the equilibrium relaxation time of the population of the resonant two-level system due to single-phonon processes:

$$\gamma = \frac{M^2\omega^3}{2\pi\rho\hbar v^5}\,\mathrm{cth}\frac{\hbar\omega}{2T}. \tag{22}$$

Now substituting (18) into (11a), we find the following equation for Δn_s:

$$\partial\Delta n_s/\partial t + \gamma'\Delta n_s = \nu\Delta n(e_s(t)/\hbar) - F\,\mathrm{Re}\,f_s^{(1)}(t). \tag{23}$$

Here $\gamma' = \gamma + 1/\tau'$ and $\nu = \gamma\tau_{nr}/(\tau_{nr} + \tau_r)$. We can write a formal solution of Eq. (23):

$$\Delta n_s(t) = \nu\int_0^\infty d\tau\, e^{-\gamma'\tau}\Delta n\Big[\frac{e_s(t-\tau)}{\hbar}\Big] - F\int_0^\infty d\tau\, e^{-\gamma\tau}\,\mathrm{Re}\,f_s^{(1)}(t-\tau). \tag{24}$$

In the second term in (24) we can ignore the difference between γ' and γ by virtue of the relation $\gamma \gg 1/\tau'$ [see (104), (99), and (100)]. Multiplying both sides of (24) by $P^{-1}V^{-1}\hbar^{-1}\delta(e_s(t)/\hbar - \varepsilon)$, and summing over s, we find an integral equation for the function $\Delta n(\varepsilon)$:

$$\Delta n(\varepsilon) = \int_{-\infty}^{+\infty} d\varepsilon'\,\Delta n(\varepsilon')R(\varepsilon - \varepsilon') + I(\varepsilon - \omega), \tag{25}$$

where the kernel is

$$R(\varepsilon - \varepsilon') = \frac{\nu}{PV\hbar}\int_0^\infty d\tau\, e^{-\gamma'\tau}\sum_s \delta\Big(\frac{e_s(t)}{\hbar} - \varepsilon\Big)\cdot\delta\Big(\frac{e_s(t-\tau)}{\hbar} - \varepsilon'\Big), \tag{26}$$

and the source is

$I(\varepsilon-\omega)$

$$-\frac{F^2}{2VP\hbar}\operatorname{th}\frac{\hbar\omega}{2T}\int_0^\infty d\tau\, e^{-\gamma\tau}\int_0^\infty d\tau'\, e^{-\gamma\tau'/2}\sum_s \delta\left(\frac{e_s(t)}{\hbar}-\varepsilon\right)$$
$$\cdot\cos\left(z_s\tau'-\int_{t-\tau-\tau'}^{t-\tau}\Delta\omega_s(t')\,dt'\right). \tag{27}$$

The sums over the resonant two-level systems which appear in expression (26) and (27) are macroscopic quantities. To within small fluctuations, which are of no importance for our purposes, these sums are equal to their average values, which we will calculate [cf. (16)]:

$$\sum_c \ldots = PV\int_0^\infty de_s \langle\langle \ldots\rangle_\xi\rangle_c. \tag{28}$$

Using (28), we can transform expressions (26) and (27) to the following form:

$$R(x)=\frac{\nu}{\pi}\int_0^\infty d\tau\cos x\tau\beta(\tau), \tag{29}$$

where

$$\beta(\tau)=\int_0^\infty d\tau'\, e^{-\gamma\tau'}\langle B(\tau,\tau')\rangle_c, \tag{30}$$

and

$$B(\tau,\tau')=\langle\exp\{i\tau[\Delta\omega_s(t-\tau')-\Delta\omega_s(t)]\}\rangle_\xi. \tag{31}$$

Likewise, we write

$$I(x)=\frac{1}{2}F^2\operatorname{th}\frac{\hbar\omega}{2T}\int_0^\infty d\tau\, e^{-\gamma\tau/2}\cos x\tau\mathscr{L}(\tau), \tag{32}$$

where

$$\mathscr{L}(\tau)=\int_0^\infty d\tau'\, e^{-\gamma\tau'}\langle L(\tau,\tau')\rangle_c, \tag{33}$$

and

$$L(\tau,\tau')=\left\langle\exp\left\{i\left[\tau\Delta\omega_s(t)-\int_{t-\tau-\tau'}^{t-\tau'}\Delta\omega_s(t')\,dt'\right]\right\}\right\rangle_\xi. \tag{34}$$

Here we have made use of the circumstance that when we take an average over ξ of a functional which is an odd function of the telegraphic process $\xi(t)$ we get zero.

Equation (25) can be solved by the Fourier method. As a result we find

$$\Delta n(\varepsilon)=\frac{1}{2}F^2\operatorname{th}\frac{\hbar\omega}{2T}\int_0^\infty d\tau\cos((\omega-\varepsilon)\tau)\,e^{-\frac{1}{2}\gamma\tau}\frac{\mathscr{L}(\tau)}{1-\nu\beta(\tau)}. \tag{35}$$

To determine the nonlinear increment in the absorption coefficient, i.e., in $\operatorname{Im}X(\omega)$, we need to evaluate the quantity $f_s^{(3)}$. For this quantity we find the following equation from (11b):

$$\frac{\partial f_s^{(3)}}{\partial t}+i\left(\omega-\frac{e_s(t)}{\hbar}\right)f_s^{(3)}+\frac{\gamma}{2}f_s^{(3)}$$
$$+\nu f_s^{(1)}(t)\,\Delta n\left(\frac{e_s(t)}{\hbar}\right)\operatorname{cth}\frac{\hbar\omega}{2T}=F\Delta n_s(t). \tag{36}$$

The last term on the left side of Eq. (36) actually describes the change in the relaxation time of the nondiagonal component of the density matrix due to the appearance of nonequilibrium phonons in the system. A detailed calculation shows that in cases in which spectral diffusion is important this change can be ignored if the deviation from equilibrium is small. Spectral diffusion leads to either a more rapid relaxation of the nondiagonal component of the density matrix, f_s, or a pronounced smearing of the phonon distribution, so the resonant region ends up with few phonons. We will accordingly ignore this term below.

Using (24) and (35), we find the function $f_s^{(3)}(t)$ from (36). Substituting it, summed with (21), into (16), and integrating, we find

$$\operatorname{Im}\chi(\omega)=\pi\hbar P(1-F^2/F_c^2)\operatorname{th}\frac{\hbar\omega}{2T}. \tag{37}$$

The characteristic amplitude F_c—the saturation of the resonant absorption—is determined in this case from the expression

$$F_c^{-2}=\int_0^\infty d\tau\, e^{-\gamma\tau}\left[\nu\frac{\mathscr{L}^2(\tau)}{1-\nu\beta(\tau)}+\mathscr{K}(\tau)\right], \tag{38}$$

where

$$\mathscr{K}(\tau)=\int_0^\infty d\tau'\, e^{-\gamma\tau'}\langle K(\tau,\tau')\rangle_c, \tag{39}$$

and

$$K(\tau,\tau')=\left\langle\exp\left\{-i\left[\int_{t-\tau}^{t}\Delta\omega_s(t')\,dt'-\int_{t-\tau-\tau'}^{t-\tau-\tau'}\Delta\omega_s(t')\,dt'\right]\right\}\right\rangle_\xi. \tag{40}$$

The first term in brackets in (38) describes the effect of the nonequilibrium phonons on the characteristic saturation amplitude F_c of the resonant absorption (the effect of the phonon bottleneck). In a case without an accumulation of phonons ($\tau_{nr}=v=0$) the amplitude F_c is determined by the second term, and we obtain the result derived in Refs. 2 and 3. As would be expected, the accumulation of resonant phonons results in a lowering of the threshold for the nonlinearity in the resonant absorption.

5. AVERAGING PROCEDURE

The quantities in which we are interested can be expressed in terms of averages of the type $\langle\langle\exp(iA)\rangle_\xi\rangle_c$, where A is some linear functional of $\Delta\omega_s(t)$. An important property of this functional is that it vanishes, and the corresponding average becomes unity, if $\Delta\omega_s$ is independent of t. These results reflect the obvious fact that the two-level systems which contribute to the spectral diffusion are those which can undergo transitions.

We will carry out the averaging procedure in two steps. We first take an average over the telegraphic processes in the thermal two-level systems. We then take a configurational average over the parameters and positions of the thermal two-level systems. Since the functional A is linear, the average $\langle\exp(iA)\rangle_\xi$ breaks up into a product of independent averages, each corresponding to some thermal two-level system. We assume that the telegraphic processes in them are uncorrelated with each other.

We begin the calculations with the simplest case: the function $B(\tau,\tau')$ in (31). This function breaks up into the product of the functions

$$b(\tau,\ \tau')=\langle\exp\{iJ\tau[\xi(\tau')-\xi(0)]\}\rangle_{\xi,\,\xi(0)},\qquad \tau,\ \tau'>0, \tag{41}$$

where J and $\xi(t)$ correspond to some thermal two-level system. The average in (41) is carried out over all realizations of the random process $\xi(t)$ for the given value of $\xi(0)$ and over all $\xi(0)$. In deriving (41) we set $t=0$ in (31), since in the steady state this average does not depend on the particular time t. We also made use of the circumstance that there is no preferred direction for time in the telegraphic process $\xi(t)$, so we can take this process to be an even function of t when we take the average. Noting that the modulus of $\xi(t)$ is always unity, we can transform $b(\tau,\tau')$ to

$$b(\tau,\ \tau')=\langle(\cos J\tau-i\xi(0)\sin J\tau)\cdot(\cos J\tau+i\xi(\tau')\sin J\tau)\rangle_{\xi,\,\xi(0)}. \tag{42}$$

With a fixed $\xi(0)$, the average is[26] $\langle\xi(\tau)\rangle=\xi(0)\exp(-2\Gamma\tau')$, where Γ is the frequency of the jumps of the given thermal two-level system. The quantity $\xi(0)$ itself takes on the values ± 1 with probabilities of 1/2, so averages over this quantity vanish. As a result we find

$$b(\tau,\ \tau')=1-(1-e^{-2\Gamma\tau'})\sin^2 J\tau. \tag{43}$$

Our problem now reduces to one of calculating the average value of the quantity $B(\tau,\tau')$ over the configurations of the thermal two-level systems. The effect of a thermal two-level system on a resonant two-level system is determined by two factors: the distance from the thermal two-level system, r (on which the quantity J depends), and the frequency of its jumps, Γ. A configurational average thus actually reduces to an average over these two quantities. With regard to the average r, we assume that all spatial positions of the thermal two-level system are equally probable. The distribution in Γ (i.e., actually the distribution in the tunneling transparency) is (Ref. 25, for example)

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

where Γ_0 is the maximum frequency of the jumps of a thermal two-level system, given by (5). For simplicity we replace the expression in the radical by unity; this simplification has no substantial effect on the results.

Using the Holtsmark method[26] to calculate the average, we find

$$\langle B(\tau,\tau')\rangle_c=\exp\left\{-PT\int_0^\infty d^3r\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}[1-b(\tau,\tau')]\right\}. \tag{44}$$

Using $J=D^2/\rho v^2 r^3\hbar$, we can put (44) in the form

$$\langle B(\tau,\ \tau')\rangle_c=\exp(-Q(\tau,\ \tau')/\tau_d), \tag{45}$$

where

$$Q(\tau,\tau')=\int_0^\infty\frac{dJ}{J^2}\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}[1-b(\tau,\tau')], \tag{46}$$

and

$$\frac{\hbar}{\tau_d}=\frac{4\pi}{3}\frac{D^2PT}{\rho v^2}\equiv E_d \tag{47}$$

is the characteristic width of the region of spectral diffusion, introduced by Hunklinger and Arnold.[6] This width is equal in order of magnitude with the characteristic change in the energy of the resonant two-level system, e, due to a transition in a thermal two-level system at an average distance $(PT)^{-1/3}$.

Substituting (43) into (46), and integrating over J, we find

$$Q(\tau,\tau')=\frac{\pi}{2}\tau\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}(1-e^{-2\Gamma\tau'}). \tag{48}$$

We turn now to an evaluation of the quantity $L(\tau,\tau')$ in (34). The average over ξ breaks up into a product of averages:

$$l(\tau,\tau')=\left\langle\exp\left\{iJ\left[\tau\xi(0)-\int_{\tau'}^{\tau'+\tau}\xi(t')\,dt'\right]\right\}\right\rangle_{\xi,\xi(0)} \tag{49}$$

[we set $t=0$ in (34) and replaced $\xi(t)$ by $\xi(-t)$]. We first take the average over ξ at a fixed value of $\xi(0)$. For this purpose we introduce the auxiliary function

$$\Psi(t)=\left\langle\exp\left\{-iJ\int_0^t\xi(t')v(t')\,dt'\right\}\right\rangle_\xi\Bigg|_{\xi(0)} \tag{50}$$

As was shown by Klyatskin,[27] the function $\Psi(t)$ satisfies the integrodifferential equation

$$\frac{d\Psi}{dt}=-iJ\xi(0)e^{-2\Gamma(t)}v(t)-J^2v(t)\int_0^t dt'\,e^{-2\Gamma(t-t')}v(t')\Psi(t'). \tag{51}$$

It is equivalent to the second-order differential equation

$$\frac{d^2\Psi}{dt^2}+\left[2\Gamma-\frac{d\ln v(t)}{dt}\right]\frac{d\Psi}{dt}+J^2v^2(t)\Psi=0 \tag{52}$$

with the initial condition

$$\Psi(0)=1,\qquad \frac{d\Psi}{dt}\Bigg|_{t=0}=-iJ\xi(0)v(0). \tag{53}$$

To evaluate (49), we adopt as $v(t)$ in (50) a piecewise-constant function equal to $+1$ at $\tau'\leqslant t\leqslant\tau+\tau'$ and 0 in the interval $0\leqslant t<\tau'$. At the point $t=\tau'$, this function is discontinuous; the function $\Psi(t)$ is continuous; and its derivative is found from Eq. (51):

$$\frac{d\Psi}{dt}\Bigg|_{t=\tau'+0}=-iJ\xi(0)e^{-2\Gamma\tau'},\qquad \Psi(\tau')=1. \tag{54}$$

The quantity $l(\tau,\tau')$ is related to the function Ψ determined in this manner by

$$l(\tau,\ \tau')=\langle[\cos J\tau+i\xi(0)\sin J\tau]\Psi(\tau+\tau')\rangle_{\xi(0)}. \tag{55}$$

Solving Eq. (52) on the interval $\tau'\leqslant t\leqslant\tau+\tau'$ under the boundary conditions (54), we find $\Psi(\tau+\tau')$. Substituting it into (55), and taking the average over $\xi(0)$, we find

$$l(\tau,\tau')=e^{-\Gamma\tau}\left[\cos J\tau\left(\operatorname{ch}(\Gamma^2-J^2)^{1/2}\tau+\frac{\Gamma}{(\Gamma^2-J^2)^{1/2}}\operatorname{sh}(\Gamma^2-J^2)^{1/2}\tau\right)+e^{-2\Gamma\tau'}\frac{J}{(\Gamma^2-J^2)^{1/2}}\sin J\tau\operatorname{sh}(\Gamma^2-J^2)^{1/2}\tau\right]. \tag{56}$$

We wish to stress that this expression holds for both $\Gamma>J$ and $\Gamma<J$. Taking a configuration average by the Holtsmark method, we find

$$\langle L(\tau,\ \tau')\rangle_c=\exp(-V(\tau,\ \tau')/\tau_d), \tag{57}$$

where

$$V(\tau,\tau')=\int_0^\infty \frac{dJ}{J^2}\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}\,[1-l(\tau,\tau')]. \tag{58}$$

It remains to calculate the function $K(\tau,t')$, determined by (40), and the configurational average of this function. For this purpose we set $t=\tau'+2\tau$ in (40). The average over the telegraphic process at one thermal two-level system, $k(\tau,\tau')$, can be written in the form in (50). In this case the function $v(t)$ is -1 in the interval $0\leqslant t\leqslant\tau$, at $\tau<t<\tau+\tau'$, and $+1$ in the interval $\tau+\tau'\leqslant t\leqslant 2\tau+\tau'$. Then calculating $\Psi(\tau'+2\tau)$ with the help of (52), (53), and (51), and taking an average over $\xi(0)$, we find

$$k(\tau,\tau')=e^{-2\Gamma\tau}\left(\operatorname{ch}(\Gamma^2-J^2)^{1/2}\tau+\frac{\Gamma}{(\Gamma^2-J^2)^{1/2}}\operatorname{sh}(\Gamma^2-J^2)^{1/2}\tau\right)^2 + \frac{J^2}{\Gamma^2-J^2}e^{-2\Gamma(\tau+\tau')}\operatorname{sh}^2(\Gamma^2-J^2)^{1/2}\tau. \tag{59}$$

Then taking a configurational average, we find

$$\langle K(\tau\,\tau')\rangle_c=\exp(-S(\tau,\tau')/\tau_d), \tag{60}$$

where

$$S(\tau,\tau')=\int_0^\infty \frac{dJ}{J^2}\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}\,[1-k(\tau,\tau')]. \tag{61}$$

6. STUDY OF LIMITING CASES

The functions $\langle L(\tau,\tau')\rangle_c$ and $\langle K(\tau,\tau')\rangle_c$, given by (57) and (60) depend on the relation between the characteristic value of Γ (which is equal to Γ_0) and that of J, (which is equal to $1/\tau_d$). It is thus natural to consider two limiting cases. Which is actually realized depends on the relation between the temperature T and its characteristic value T_d given by (9).

a) Case of low temperatures, $T\ll T_d$ ($\Gamma_0\tau_d\ll 1$). In this case, calculations yield (see the Appendix)

$$V(\tau,\tau')=\frac{\pi}{2}\tau\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}\,[1-e^{-\Gamma(\tau+2\tau')}], \tag{62}$$

$$S(\tau,\tau')=\frac{\pi}{2}\tau\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}\,[1-e^{-2\Gamma(\tau+\tau')}]. \tag{63}$$

In this region there are three characteristic frequencies: γ, Γ_0, and $(\Gamma_0/\tau_d)^{1/2}\gg\Gamma_0$. Correspondingly, there are three limiting cases.

Under the conditions[2)]

$$\gamma\ll\Gamma_0\ll(\Gamma_0/\tau_d)^{1/2}\quad(\hbar\omega\ll T) \tag{64}$$

the integrals (30), (33), (35), (38) and (39) are dominated by $\tau\lesssim\tau_d$, and we have $\tau'\leqslant\gamma^{-1}$, i.e., the characteristic values of τ' are much greater than τ. In this case the integrals in (48), (62), and (63) depend logarithmically on τ'. Carrying out the integration, we find the following expression for F_c^2, which holds to within logarithmic corrections

$$F_c^2=\frac{\pi\nu\ln(\Gamma_0/\gamma)}{2\tau_d|\ln(1-\nu/\gamma')|}. \tag{65}$$

Under the condition $\nu\ll\gamma'$ ($\tau_{nr}\ll\tau_r$), without an accumulation of phonons, we find from (65) the result[2]

$$F_c^2=(\pi\gamma\ln\Gamma_0/\gamma)/2\tau_d.$$

This result is interpreted in the following way[9]: We apply the label "resonant" to those two-level systems whose frequency deviation $|e_s(t)/\hbar-\omega|$, does not exceed $1/\tau_d$. Because of jumps in neighboring thermal two-level systems, this deviation sometimes falls in an interval (near zero) with a width on the order of $(\Gamma_0/\tau_d)^{1/2}$, which we call the "resonant region." The time spent by a resonant two-level system in this region is on the order of $\tau_\varphi=(\tau_d/\Gamma_0)^{1/2}$ [see (7)].

The population of a resonant two-level system can be changed by the pump field F only if the system is in the resonant region. The average rate of change of the population in this case would be $-\dot n\approx F^2\tau_\varphi$. Since we have $\gamma\ll\Gamma_0$, a resonant two-level system finds itself in the resonant region many times over its lifetime γ^{-1}. The total time it spends there is $\gamma^{-1}(1/\tau_\varphi)/(1/\tau_d)$. This time is smaller than the time γ^{-1} by a factor equal to the ratio of the width of the region of spectral diffusion, $1/\tau_d$, to the width of the resonant region, $1/\tau_\varphi$. The critical intensity is found from the condition that over this time the resonant two-level system is excited with a probability of order unity:

$$F_c^2\tau_\varphi\gamma^{-1}\tau_d/\tau_\varphi\approx 1.$$

We thus find $F_c^2\approx\gamma/\tau_d$, which agrees with the calculated result within a large logarithm. The change in the population during a single crossing of the resonant region is small, $\Delta n\approx F_c^2\tau_\varphi^2\ll 1$, and a resonant two-level system must reach this region many times for any significant change in its population.

Under the condition $1-\nu/\gamma'\ll 1$ ($\tau_{nr}\gg\tau_r$), that phonons do accumulate, we find from (65)

$$F_c^2=\pi\gamma\ln(\Gamma_0/\gamma)/2\tau_d\ln(\tau_{nr}/\tau_r).$$

In this case F_c^2 turns out to be inversely proportional to only the logarithm of the large ratio τ_{nr}/τ_r, not to its first power, as in the ordinary phonon-bottleneck effect (Refs. 19–22; see also Ref. 14). Consequently, F_c^2 changes only slightly from the case without phonon accumulation. The reason for this result is the spreading of the nonequilibrium phonons over the spectrum caused by spectral diffusion.

To illustrate this point, we note that the shape of the burned-out hole in this case is

$$\Delta n(\varepsilon)=B_1\int_0^\infty dx\,\frac{\cos\Delta_1 x}{e^x-1+\delta_1}, \tag{66}$$

where

$$B_1=F^2\tau_d\hbar\omega/2\pi T\gamma\ln(\Gamma_0/\gamma),$$
$$\Delta_1=2\tau_d(\omega-\varepsilon)/\pi\ln(\Gamma_0/\gamma),\quad \delta_1=1-\nu/\gamma'. \tag{67}$$

It follows from (66) that in the case without phonon accumulation, i.e., under the condition $\nu/\gamma'\ll 1$ ($\tau_{nr}\ll\tau_r$), the hole has Lorentzian shape,[2]

$$\Delta n(\varepsilon)=B_1/(1+\Delta_1^2) \tag{68}$$

with a width $\Delta_\varepsilon\approx(\pi\ln\Gamma_0/\gamma)/2\tau_d$. In other words, this width is on the order of the width of the region of spectral diffusion. This result is understandable. After being excited in the resonant region, a resonant two-level system undergoes a random walk over the entire region of spectral diffusion, $_1/\tau_d$, over a time $\gamma^{-1}\gg\Gamma_0^{-1}$. It can lose its excitation, i.e., emit a phonon, while at any point in this interval.

Under the condition $\delta_1 \ll 1$ ($\tau_{nr} \gg \tau_r$), i.e., in the case with phonon accumulation, the shape of the hole is quite different from Lorentzian:

$$\Delta n(\varepsilon) = B_1 \begin{cases} \ln(1/\delta_1), & |\Delta_1| \ll 1, \\ \ln(1/|\Delta_1|\delta_1), & 1 \ll |\Delta_1| \ll 1/\delta_1, \\ 1/\Delta_1^2\delta_1^2, & |\Delta_1| \gg 1/\delta_1. \end{cases} \quad (69)$$

The effective width of the hole in this case is greater by a factor of $1/\delta_1 = \tau_{nr}/\tau_r$ than the width of the region of spectral diffusion, $1/\tau_d$. The linear time dependence of τ_{nr} (instead of a square-root diffusion dependence) of the width of the nonequilibrium distribution results from infrequent collisions of phonons with quasiresonant two-level systems close to a thermal two-level system [within a distance r substantially smaller than the average value $(PT)^{1/3}$]. For roughly half the time, a quasiresonant two-level system has a frequency deviation $|e_s(t)/\hbar - \omega| \lesssim 1/\tau_d$. It sometimes enters the resonant region as a result of transitions in thermal two-level systems in its vicinity (other than the nearest). The nearest thermal two-level system is in one of its quantum states at this time. When it goes into the other state, the energy of the quasiresonant two-level system changes abruptly by a large amount $\hbar J \gg \hbar/\tau_d$. Consequently, the quasiresonant two-level system spends the other half of its time far from resonance.

Since we have $J \sim r^{-3}$, the concentration of quasiresonant two-level systems with

$$J \approx (1/\tau_d)(\tau_{nr}/\tau_r)$$

is smaller by a factor of τ_{nr}/τ_r than the concentration of resonant systems. Accordingly, over a time τ_{nr} a resonant phonon can be absorbed only once by a two-level system of this sort, on the average. An excited quasiresonant two-level system may, on the other hand, emit a phonon while far from resonance, after a transition of the nearest thermal two-level system to the corresponding state. The frequency of the phonon emitted in the process will differ from that of the absorbed phonon by an amount of order J.

Because of this spreading of the nonequilibrium phonons over the spectrum, the number of resonant phonons [i.e., the number of phonons which can be absorbed by resonant two-level system) as follows from (69)] is proportional to just the large logarithm $\ln(\tau_{nr}/\tau_r)$, while in the absence of a spectral diffusion the number of such phonons would be proportional to τ_{nr}/τ_r.

We turn now to the other limiting case. Under the conditions

$$\Gamma_0 \ll \gamma \ll (\Gamma_0/\tau_d)^{1/2} \quad (T \ll \hbar\omega \ll T^{2/3}T_d^{1/3}) \quad (70)$$

the characteristic values of τ and τ' in the integrals in (30), (33), (35) and (38) satisfy the conditions $\Gamma_0\tau$, $\Gamma_0\tau' \ll 1$. In this case, the exponential functions in (48), (62), and (63) can be expanded in series, and the integration over Γ can be carried out. The integrals over τ' which appear as a result can be evaluated quite easily; for F_c^2 we find

$$F_c^2 = \frac{2\pi\Gamma_0}{\tau_d \ln[\Gamma_0/\gamma^2\tau_d(1-\nu/\gamma')]}. \quad (71)$$

Under the condition $\nu/\gamma' \ll 1$, without phonon accumulation, we have

$$F_c^2 = 2\pi\Gamma_0/\tau_d \ln(\Gamma_0/\gamma^2\tau_d).$$

To within a large logarithm, the value of F_c in this case is the same in order of magnitude as $1/\tau_\varphi$ [see (7)]. A resonant two-level system undergoes a significant change in population at $F \approx F_c$ during even a single crossing of the resonant region, over a time τ_φ. The accumulation of phonons because of spectral spreading changes only the argument of the logarithm, as in the preceding case.

For the burned-out hole in this case we have (cf. Ref. 2)

$$\Delta n(\varepsilon) = B_2 \int_0^\infty dx \cos\Delta_2 x \frac{e^{-x}}{x+\delta_2}, \quad (72)$$

where

$$B_2 = F^2\tau_d/2\pi\Gamma_0, \quad \Delta_2 = (2\tau_d/\pi\Gamma_0)^{1/2}(\omega-\varepsilon),$$
$$\delta_2 = \gamma(\pi\tau_d/2\Gamma_0)^{1/2}(1-\nu/\gamma') \ll 1. \quad (73)$$

Its shape is non-Lorentzian. The asymptotic behavior is the same as in (69), where the subscript 1 should be replaced by 2 on all quantities. The effective width Δ_ε is

$$\Delta_\varepsilon \approx \frac{\Gamma_0/\gamma\tau_d}{1-\nu/\gamma'} \approx \frac{\Gamma_0}{\gamma\tau_d}\left(1+\frac{\tau_{nr}}{\tau_r}\right). \quad (74)$$

This result has the following physical meaning: The quantity $(\Gamma_0/\tau_d)^{1/2}$ is the width of the resonant region. In this region, the population of the resonant two-level system is changed significantly by the pump field. According to (6), the time scale for the crossing of this region is $(\tau_d/\Gamma_0)^{1/2} = \tau_\varphi$. In the case at hand, (70), however, this time is far shorter than the relaxation time of the resonant two-level system, γ^{-1}. Consequently, after crossing the resonant region the two-level system remains excited for a time γ^{-1}. Over this time, its frequency (the energy of the resonant two-level system divided by $\hbar$) takes on the value $\Gamma_0/\gamma\tau_d$, according to (6). This value is the effective width of the burned-out hole in the case without phonon accumulation ($\tau_{nr} \ll \tau_r$). If $\tau_{nr} \gg \tau_r$, on the other hand, as in the case under consideration here, (70), the width of the hole turns out to be larger by a factor of τ_{nr}/τ_r because of the scattering of nonequilibrium phonons by quasiresonant two-level systems.

We turn now to the case

$$\Gamma_0 \ll (\Gamma_0/\tau_d)^{1/2} \ll \gamma \quad (\hbar\omega \gg T^{2/3}T_d^{1/3}). \quad (75)$$

From (38) we find the following expression for F_c^2:

$$F_c^{-2} = \frac{\tau_d}{\pi\Gamma_0}\int_0^\infty dx \frac{e^{-x}}{x+2\delta_3}, \quad (76)$$

where

$$\delta_3 = \frac{\gamma^2\tau_d}{2\pi\Gamma_0}\left(1-\frac{\nu}{\gamma'}\right). \quad (77)$$

The shape of the burned-out hole in this case is described by

$$\Delta n(\varepsilon) = B_3 \int_0^\infty dx \cos\Delta_3 x \frac{e^{-x}}{x+\delta_3}, \quad (78)$$

where

$$B_3 = F^2\tau_d/2\pi\Gamma_0, \quad \Delta_3 = 2(\omega-\varepsilon)/\gamma. \quad (79)$$

If $\delta_3 \gg 1$, we find from (76)

$$F_c^2 = \gamma^2(1-\nu/\gamma') \approx \gamma^2\tau_r/\tau_{nr}. \quad (80)$$

This result agrees with the result of Ref. 14, where spectral diffusion was ignored. The condition $\delta_3 \gg 1$ thus determines the range of applicability of the theory of Ref. 14. This condition can be rewritten in the form

$$\hbar\omega \gg T^{1/3}(T_d E_c)^{1/3}. \tag{81}$$

For the burned-out hole in this case we find from (78)

$$\Delta n(\varepsilon) = \frac{B_3}{\delta_3}\frac{1}{1+\Delta_3^2}. \tag{82}$$

In other words, the hole has a Lorentzian shape with a half-width $\Delta\varepsilon = \gamma/2$. However, an important reservation must be made here.[1)]

It follows from (18) and (82) that the phonon distribution which is found is quite narrow, with a width on the order of the uncertainty in the energy of the resonant two-level system. It is thus incorrect to find it from a kinetic equation in this case. Kinetic equations (11) do not apply to such narrow distributions. Since resonant absorption in the absence of spectral diffusion is essentially resonant scattering, the nonequilibrium phonons should have this same frequency as the pump field, ω (Ref. 13). Consequently, although the width of the burned-out hole is on the order of γ in the case, the shape of the distribution function of nonequilibrium phonons does not reproduce that of the burned-out hole. The width of the nonequilibrium phonon distribution is $\Gamma_0/\gamma\tau_d$ in this case; it is substantially smaller than γ and is determined by the spectral diffusion. Experimentally, it could be measured by studying Brillouin scattering of light.

In the opposite limit, with $\delta_3 \ll 1$ or $\hbar\omega \ll T^{1/2}(T_d E_c)^{1/4}$, [but with $\hbar\omega \gg T^{2/3}T_d^{1/3}$; see (75)], we find the following expression, to within logarithmic corrections, for F_c^2 from (76):

$$F_c^2 = \frac{\pi\Gamma_0}{\tau_d}\ln^{-1}\left(\frac{1}{2\delta_3}\right) \ll \gamma^2. \tag{83}$$

In this case the burned-out hole, (78), has a definitely non-Lorentzian shape. Its asymptotic behavior is described by (69), with a subscript 1 must be replaced by 3 on all quantities. The effective width is

$$\Delta_e \approx (\Gamma_0/\gamma\tau)(\tau_{nr}/\tau_r) \gg \gamma.$$

b) Case of high temperatures, $T \gg T_d$ ($\Gamma_0\tau_d \gg 1$). In this region there are three characteristic frequencies: Γ_0, $1/\tau_d$, and γ. Correspondingly, there are three limiting cases. In the case

$$\Gamma_0 \gg \frac{1}{\tau_d} \gg \gamma \qquad (\hbar\omega \ll T_d) \tag{84}$$

the integral over Γ can be extended to ∞ in (58) and (61). As a result, we find the following result, which holds to within logarithmic corrections under the condition $\tau' \gg \tau$, as is shown in the Appendix:

$$V(\tau,\tau') = S(\tau,\tau') = \frac{\pi}{2}\tau\ln\frac{\tau'}{\tau}. \tag{85}$$

The ranges $\tau \lesssim \tau_d$ and $\tau' \lesssim \gamma^{-1}$ contribute to the integrals in (30), (33), (35), (38) and (39); i.e., the characteristic values of τ' are much greater than τ. In this case, expression (48) for $Q(\tau,\tau')$ takes the form

$$Q(\tau,\tau') = \frac{\pi}{2}\tau\ln\Gamma_0\tau'. \tag{86}$$

Going through the calculations, we find F_c^2,

$$F_c^2 = \frac{\pi\gamma\ln(1/\gamma\tau_d)}{2\tau_d}\left(1+\frac{\nu}{2\gamma}+\frac{\nu}{\gamma}\frac{\ln(1/\gamma\tau_d)}{\ln(\Gamma_0/\gamma)}\left|\ln(1-\nu/\gamma')\right|\right)^{-1}, \tag{87}$$

and the width of the burned-out hole,

$$\Delta n(\varepsilon) = B_4\int_0^\infty dx\cos(\Delta_4 x)\frac{\gamma' e^{-x}}{\gamma' - \nu e^{-\lambda x}}, \tag{88}$$

where

$$B_4 = \frac{F^2\tau_d\hbar\omega}{2\pi\gamma T\ln(1/\gamma\tau_d)}, \qquad \Delta_4 = \frac{2\tau_d(\omega-\varepsilon)}{\pi\ln(1/\gamma\tau_d)},$$
$$\lambda = \frac{\ln(\Gamma_0/\gamma)}{\ln(1/\gamma\tau_d)} \gg 1.$$

If there is no phonon accumulation, i.e., under the condition $\tau_{nr} \ll \tau_r$ ($\nu \ll \gamma$), we find

$$F_c^2 = (\pi\gamma\ln(1/\gamma\tau_d))/2\tau_d.$$

In this case the burned-out hole has a Lorentzian shape[2]:

$$\Delta n(\varepsilon) = B_4/(1+\Delta_4^2). \tag{89}$$

The width of the hole is on the order of the width of the region of spectral diffusion, $1/\tau_d$.

In the opposite case, with $\tau_{nr} \gg \tau_r$ ($1 - \nu/\gamma' \ll 1$) and with phonon accumulation, the critical intensity is changed only insignificantly by the spreading of the phonons over the spectrum. The shape of the burned-out hole, in contrast, changes radically. The hole becomes non-Lorentzian, and from (87) we find

$$\Delta n(\varepsilon) = B_4\begin{cases} \dfrac{1}{1+\Delta_4^2} - \dfrac{1}{\lambda}\ln(1-\nu/\gamma'), & |\Delta_4| \ll \lambda, \\ \dfrac{1}{\lambda}\ln\dfrac{\lambda}{|\Delta_4|(1-\nu/\gamma')}, & \lambda \ll |\Delta_4| \ll \dfrac{\lambda}{1-\nu/\gamma'}, \\ \dfrac{\lambda^2}{\Delta_4^2(1-\nu/\gamma')^2}, & |\Delta_4| \gg \dfrac{\lambda}{1-\nu/\gamma'}. \end{cases} \tag{90}$$

The effective width of the hole is

$$\Delta_e \approx (\ln(\Gamma_0/\gamma)/\tau_d)(\tau_{nr}/\tau_r),$$

larger than the width of the region of spectral diffusion by a factor of τ_{nr}/τ_r.

We now consider the case

$$\Gamma_0 \gg \gamma \gg \frac{1}{\tau_d} \qquad (T \gg \hbar\omega \gg T_d). \tag{91}$$

In this case, the integrals (30), (33), (35), (38), and (39) are dominated by τ, $\tau' \leqslant \gamma^{-1}$. The quantities $V(\tau,\tau')$ and $S(\tau,\tau')$ can be ignored in the arguments of the exponential functions in (57) and (60) in this case. Expression (86) remains valid for $Q(\tau,\tau')$. For F_c^2 we find [cf. (76)]

$$F_c^{-2} = \frac{2\tau_d}{\pi\nu\ln(\Gamma_0/\gamma)}\int_0^\infty dx\frac{e^{-x}}{x+2\delta_5}, \tag{92}$$

where

$$\delta_5 = \frac{\gamma^2\tau_d}{\pi\nu\ln(\Gamma_0/\gamma)}\left(1-\frac{\nu}{\gamma'}\right). \tag{93}$$

For the burned-out hole we have [cf. (78)]

$$\Delta n(\varepsilon) = B_5\int_0^\infty dx\cos(\Delta_5 x)\frac{e^{-x}}{x+\delta_5}, \tag{94}$$

where

$$B_5 = F^2\tau_d\hbar\omega/2\pi\nu T\ln(\Gamma_0/\gamma), \quad \Delta_5 = 2(\omega-\varepsilon)/\gamma. \tag{95}$$

Under the condition $\delta_5 \gg 1$ we find [if there is an accu-

mulation of phonons, i.e., if $\tau_{nr} \gg \tau_\gamma$, the condition $T \gg (T_d E_c)^{1/2}$) as well as (91) would also have to be satisfied in this case]

$$F_c^2 = \gamma^2(1-\nu/\gamma'),$$

as in the ordinary phonon bottleneck (see also Ref. 14). In this case the burned-out hole has a Lorentzian shape with a half-width $\gamma/2$ [see the comment following Eq. (82) in the connection]. In the opposite case, $\delta_5 \ll 1$ ($T \ll (T_d E_c)^{1/2}$), we find

$$F_c^2 = \pi\gamma \ln(\Gamma_0/\gamma)/2\tau_d \ln(1/2\delta_5), \qquad (96)$$

which agrees to within logarithmic factors with (87). In this case the burned-out hole has a non-Lorentzian shape. The expressions for this shape in the various limiting cases are the same as (69), where the subscript 1 must be replaced by 5 on all quantities.

Finally, we consider the limiting case

$$\gamma \gg \Gamma_0 \gg 1/\tau_d \quad (\hbar\omega \gg T \gg T_d). \qquad (97)$$

The distinction from the preceding case is that now the characteristic values satisfy $\Gamma\tau' \approx \Gamma/\gamma \ll 1$. We can thus write (48) in the form

$$Q(\tau, \tau') = \pi\Gamma_0\tau\tau'.$$

The results turn out to be the same as in case (75). The critical intensity is given by (76), and the burned-out hole is described by (78). The condition $\delta_3 \gg 1$ ($\hbar\omega \ll T^{1/2}(T_d E_c)^{1/4}$) is compatible with (97) only if $T \ll (E_c T_d)^{1/2}$. At $T \gg (E_c T_d)^{1/2}$, only the case $\delta_3 \gg 1$ is realized.

7. DISCUSSION OF RESULTS; NUMERICAL ESTIMATES

Spectral diffusion in dielectric glasses thus gives rise to rather different pictures of the burned-out hole and the nonlinear resonant absorption, depending on the signal frequency ω and the temperature T. When the accumulation of nonequilibrium resonant phonons is taken into consideration, we find that spectral diffusion also plays a role in cases in which it was previously negligible [cases (75), (91), and (97)].[2,3] The reason for this result is that the nonequilibrium phonons spread out markedly over the spectrum as a result of spectral diffusion, thereby modifying the phonon-bottleneck phenomenon. The width of the phonon distribution increases linearly with increasing nonresonant lifetime τ_{nr}. The functional dependence of the width of the phonon distribution on the time τ_{nr} is not of a "diffusive" nature because of collisions of nonequilibrium phonons with quasiresonant two-level systems in the vicinity of a thermal two-level system.

The theory derived here refers to the steady state. Consequently, the length of the excitation pulse must exceed $\max(\tau_{nr}, \tau_{nr}/\gamma\tau_r)$. This condition reflects the circumstance that under the condition $\gamma\tau_r \ll 1$ nonequilibrium phonons are captured by resonant two-level systems.[17] A long time is required for a phonon to collide with a quasiresonant two-level system.

If the accumulation of resonant phonons is to be manifested, it is further necessary that the dimensions of the excitation region exceed the diffusion length

$$L = v(\tau_{nr}\tau_r)^{1/2}.$$

Otherwise, the phonons will escape from the excitation region before they collide with quasiresonant two-level systems. In amorphous quartz (SiO_2), for example, at a temperature $T = 0.5$ K and an excitation frequency $f = \omega/2\pi = 1$ GHz, these parameters would have the following numerical values (for transverse phonons)[1]: $\tau_r \approx 2.5$ μs, $\tau_{nr} \approx 20$ μs, $\gamma^{-1} \approx 0.5$ μs, and $L = 3$ cm.

Let us examine the effect of two-phonon relaxation processes of resonant two-level systems [described by the time τ' in (11a)] on the accumulation of resonant phonons. It follows from the analysis in Sec. 6 that nonequilibrium phonons affect the resonant absorption under the condition $1 - \nu/\gamma' \ll 1$. Since we have

$$\nu = \gamma\tau_{nr}/(\tau_{nr}+\tau_r), \quad \gamma' = \gamma + 1/\tau',$$

and $\gamma\tau' \gg 1$ a necessary condition here is $\tau_{nr} \gg \tau_\gamma$. In this case we have

$$1 - \nu/\gamma' = 1/\gamma\tau' + \tau_r/\tau_{nr}. \qquad (98)$$

Using expression (104) for τ', given in the Appendix, along with (22) and (4), we find that under the condition $\hbar\omega \ll T$ we have

$$\frac{1}{\gamma\tau'} = \frac{1}{12}\left(\frac{T}{E_c}\right)^2, \quad \frac{\tau_r}{\tau_{nr}} = \frac{\pi^2}{16}\left(\frac{T}{\hbar\omega}\right)^2\left(\frac{T}{E_c}\right)^2 \gg \frac{1}{\gamma\tau'}; \qquad (99)$$

i.e., the first term in (98) can be ignored in this case. In the classical frequency region, two-phonon relaxation processes of resonant two-level systems contribute much less to the rate at which excitations leave the resonant region than does the nonresonant absorption of phonons by thermal two-level systems.

In the quantum frequency region, $\hbar\omega \gg T$, the situation is different:

$$\frac{1}{\gamma\tau'} = \frac{1}{24\pi^2}\left(\frac{\hbar\omega}{E_c}\right)^2, \quad \frac{\tau_r}{\tau_{nr}} = \frac{1}{48\pi^2}\left(\frac{\hbar\omega}{E_c}\right)^2; \qquad (100)$$

i.e., the first term in (98) is twice the second. In this case, the rate at which excitations leave the resonant region is thus determined by both the nonresonant scattering of phonons and two-phonon relaxation of two-level systems.

We conclude with a few words regarding the behavior of the absorption coefficient as a function of the intensity at high intensities, under the condition $F \gg F_c$. In this case it follows from qualitative considerations similar to those in Ref. 3 that the absorbed power will not depend on the intensity. The absorption coefficient will therefore fall off in inverse proportion to the intensity. The ultimate reason for this result is that in a situation with spectral diffusion the number of two-level systems which are involved in the absorption is far greater than that in the absence of spectral diffusion. The spectral width of the interval in which the energies of these two-level systems fall is significantly greater than the width of the resonant region. Accordingly, for $F > F_c$ the number of two-level systems which are involved in the absorption does not increase with increasing intensity, as it would in the absence of spectral diffusion. Instead it remains constant.

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APPENDIX

A) Two-phonon relaxation time of resonant two-level systems. The rate of change of the population of the upper level of a two-level system with an energy e as a result of two-phonon processes can be written

$$\frac{\partial n}{\partial t}=\sum_{qq'}\{[-n(1+N_q^0)(1+N_{q'}^0)W_{qq'}^{(1)}+(1-n)N_q^0N_{q'}^0W_{qq'}^{(2)}]\delta(e-\hbar\omega_q-\hbar\omega_{q'})+[-nN_q^0(1+N_{q'}^0)W_{qq'}^{(3)}+(1-n)N_{q'}^0(1+N_q^0)W_{qq'}^{(4)}]\delta(e+\hbar\omega_q-\hbar\omega_{q'})\}\equiv-\frac{n-n^0}{\tau'}, \tag{101}$$

where n^0 is the equilibrium population of the upper level of the two-level system, N_q^0 is the equilibrium distribution function of the nonresonant phonons, and $W_{qq'}^{(i)}$ are the probabilities for the corresponding transitions. The first expression in square brackets in (101) describe decay-emission (or absorption) processes involving two-level systems and two phonons. The second expression describes association processes: One phonon is absorbed by a two-level system, and another is emitted.

The transition probabilities $W_{qq'}^{(i)}$ satisfy relations which follow from the principle of detailed balance:

$$W_{qq'}^{(1)}=W_{qq'}^{(2)},\quad W_{qq'}^{(3)}=W_{qq'}^{(4)}. \tag{102}$$

In the simple isotropic model, a calculation by second-order perturbation theory yields

$$W_{qq'}^{(1)}=W_{qq'}^{(3)}=\pi M^2D^2e^2/2V^2\hbar\rho^2v^2\hbar\omega_q\hbar\omega_{q'}. \tag{103}$$

Using (101), (102), and (103), and going through the calculations, we find the following expression for the relaxation time τ' of a resonant two-level system with $e=\hbar\omega$:

$$\frac{1}{\tau'}=\frac{D^2M^2\omega^2T^3}{8\pi^3\hbar^5\rho^2v^{10}}\left[\frac{\pi^2}{3}\frac{\hbar\omega}{T}+\frac{\hbar\omega}{T}I_1\left(\frac{\hbar\omega}{T}\right)-I_2\left(\frac{\hbar\omega}{T}\right)\right]\operatorname{cth}\frac{\hbar\omega}{2T}, \tag{104}$$

where

$$I_n(y)=\int_0^y dx\frac{x^n}{1-e^{-x}}=\begin{cases}y^n/n, & y\ll 1,\\ y^{n+1}/(n+1), & y\gg 1.\end{cases} \tag{105}$$

Under the condition $\hbar\omega\ll T$, connection processes dominate the value of $1/\tau'$. In the opposite case, with $\hbar\omega\gg T$, the rate of two-phonon relaxation is dominated by decay processes.

B) Calculation of the function $V(\tau,\tau')$. Introducing the new variables $x=J/\Gamma$, $y=\Gamma\tau$, and partitioning the range of integration over x and 0 to ∞ into the two subranges from 0 to 1 and from 1 to ∞, we rewrite (58) as

$$V(\tau,\tau')=V_1(\tau,\tau')+V_2(\tau,\tau'), \tag{106}$$

where

$$V_1(\tau,\tau')=\tau\int_0^{\Gamma_0\tau}\frac{dy}{y^2}\int_0^1\frac{dx}{x^2}[1-l_1(x,y)], \tag{107}$$

$$l_1(x,y)=e^{-y}\cos xy\left(\operatorname{ch}y(1-x^2)^{1/2}+\frac{1}{(1-x^2)^{1/2}}\operatorname{sh}y(1-x^2)^{1/2}\right)+\frac{x}{(1-x^2)^{1/2}}e^{-y(1+2\beta)}\sin xy\operatorname{sh}y(1-x^2)^{1/2} \tag{108}$$

and

$$V_2(\tau,\tau')=\tau\int_0^{\Gamma_0\tau}\frac{dy}{y^2}\left[1-\int_1^\infty\frac{dx}{x^2}l_2(x,y)\right], \tag{109}$$

where

$$l_2(x,y)=e^{-y}\cos xy\left(\cos y(x^2-1)^{1/2}+\frac{1}{(x^2-1)^{1/2}}\sin y(x^2-1)^{1/2}\right)+\frac{x}{(x^2-1)^{1/2}}e^{-y(1+2\beta)}\sin xy\sin y(x^2-1)^{1/2}, \tag{110}$$

and $\beta=\tau'/\tau$.

At low temperatures, $T\ll T_d$ ($\Gamma_0\tau_d\ll 1$), the characteristic values satisfy $\Gamma_0\tau\ll 1$. The integrand (in the integrals over y) can thus be expanded in a series in y, and only the first nonvanishing term need be retained. In the calculation of $V_1(\tau,\tau')$, we can expand the function $l_1(x,y)$ directly in y and then integrate over x. As a result we find

$$V_1(\tau,\tau')=\tau\int_0^{\Gamma_0\tau}dy[1-e^{-y(1+2\beta)}]. \tag{111}$$

In the calculation of $V_2(\tau,\tau')$ we can carry out the expansion in y only after the integration over x, since the integral over x is dominated at values $y\ll 1$ by $x\approx 1/y$. As a result we find, after some calculations,

$$V_2(\tau,\tau')=\frac{\pi}{2}\tau\int_0^{\Gamma_0\tau}\frac{dy}{y}[1-e^{-y(1+2\beta)}]. \tag{112}$$

Comparison with $V_1(\tau,\tau')$ shows that in the regions of values of τ and τ' in which we are interested here the following conditions holds:

$$V_1(\tau,\tau')\ll V_2(\tau,\tau').$$

In other words, we have

$$V(\tau,\tau')=V_2(\tau,\tau'),$$

and we find (62).

At high temperatures, $T\gg T_d$ ($\Gamma_0\tau_d\gg 1$), the characteristic values satisfy $\Gamma_0\tau\gg 1$. The upper limit on the integral over y can thus be replaced by ∞. As a result we find

$$V(\tau,\tau')=\tau v(\beta)\equiv\tau[v_1(\beta)+v_2(\beta)], \tag{113}$$

where

$$v_1(\beta)=\int_0^\infty\frac{dy}{y^2}\int_0^1\frac{dx}{x^2}[1-l_1(x,y)], \tag{114}$$

$$v_2(\beta)=\int_0^\infty\frac{dy}{y^2}\left[1-\int_1^\infty\frac{dx}{x^2}l_2(x,y)\right]. \tag{115}$$

For values $\beta\ll 1$, the quantity $v(\beta)=v(0)$ is some number on the order of unity. We are interested in the behavior of the function $v(\beta)$ as $\beta\to\infty$:

$$v_1(\beta)=v_1(0)+\int_0^\infty\frac{dy}{y^2}\int_0^1\frac{dx}{x^2}\Delta l_1(x,y), \tag{116}$$

$$v_2(\beta)=v_2(0)+\int_0^\infty \frac{dy}{y^2}\int_1^\infty \frac{dx}{x^2}\,\Delta l_2(x,y), \tag{117}$$

where

$$\Delta l_1(x,y)=\frac{x}{(1-x^2)^{1/2}}e^{-y}(1-e^{-2\beta y})\sin xy\ \mathrm{sh}\ y(1-x^2)^{1/2}, \tag{118}$$

$$\Delta l_2(x,y)=\frac{x}{(x^2-1)^{1/2}}e^{-y}(1-e^{-2\beta y})\sin xy\sin y(x^2-1)^{1/2}. \tag{119}$$

Using (118), we find from (116) that $v_1(\infty)$ is some number, since the integral in (116) converges if we set $\beta=\infty$ in (118).

With regard to the function $v_2(\beta)$ we have a different situation. This function can be written in the form

$$v^2(\beta)=v_2(0)+\int_0^\infty \frac{dy}{y^2}e^{-y}(1-e^{-2\beta y})\cdot\int_1^\infty \frac{dx}{x(x^2-1)^{1/2}}\sin xy\sin y(x^2-1)^{1/2}. \tag{120}$$

In the case $y\ll 1$ we have

$$\int_1^\infty \frac{dx}{x(x^2-1)^{1/2}}\sin xy\sin y(x^2-1)^{1/2}=\frac{\pi}{2}y, \tag{121}$$

so that as $\beta\to\infty$ the integral over y in (120) diverges logarithmically at its lower limit. As a result we find, for $\beta\to\infty$,

$$v(\beta)=\frac{\pi}{2}\ln\beta+\mathrm{const} \tag{122}$$

and thus Eq. (85). The function $S(\tau,\tau')$ is calculated in a similar way.[3]

[1)]This system of equations holds if the width of the nonequilibrium distributions n_s and N_k is greater than the uncertainty in the energy of a resonant two-level system, $\hbar\gamma$ [see (22)]. Because of spectral diffusion, this condition holds in most of the cases considered (Sec. 6).

[2)]In general, this inequality would also contain some logarithmic factors. We have omitted them to avoid making the calculations overly complicated. These factors can be reconstructed quite easily by requiring that the results found for the different limiting cases join at the boundary between their ranges of applicability.

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