Nonlinear rf hopping conductivity of semiconductors in connection with spectral diffusion

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A theory is derived for the nonlinear resonant rf hopping conductivity of a semiconductor. The region of fairly high frequencies, where the so-called two-site approximation can be used to describe the rf conductivity, is studied. Cases in which spectral diffusion plays an important role are examined. This diffusion results from dipole-dipole interactions between resonant pairs, which are responsible for absorption, and so-called thermal pairs, in which quantum transitions (jumps) cause resonant pairs to leave resonance. Expressions are derived for the shape of the burnt-out hole in cases in which spectral diffusion is important.

1. INTRODUCTION

As Pollak and Geballe have shown,[1] the rf hopping conductivity of a doped semiconductor is determined by hops of electrons within close pairs. Each such pair consists of a filled impurity center and a vacant impurity center, separated by a distance r far smaller than the mean separation $\bar{r} = (4\pi N/3)^{-1/3}$, where N is the density of impurities. The two-site model is valid under the condition $\omega \gg \sigma(0)$, where ω is the field frequency, and $\sigma(0)$ is the static conductivity of the sample (Refs. 2 and 3, for example). A study of the rf hopping conductivity yields information which supplements that found from research on the static conductivity. It can lead to conclusions regarding the state density corresponding to close pairs and regarding relaxation processes of importance to localized electrons.

In this paper we examine the nonlinear resonant absorption which is caused by transitions of electrons between levels of a pair of centers, accompanied by the absorption of a photon of an electromagnetic field. A similar situation arises in the resonant absorption of ultrasound, where the transition is accompanied by the absorption of a phonon.

A study of nonlinear absorption appears worthwhile for the following reason. In the impurity band of a doped semiconductor there are wide distributions of both the difference between single-site energies, $\Delta = \varphi_1 - \varphi_2$, and the tunneling overlap integrals $I(r)$ of the wave functions of the components of the pair. A similar situation apparently prevails in several amorphous semiconductors. Accordingly, for any frequency ω there exists a fairly large number of pairs for which the separation E between levels, given by

$$E=(\Delta^2+\Lambda^2)^{1/2}, \quad \Lambda=2I(r)=2I_0 e^{-r/a} \tag{1.1}$$

(I_0 is an energy on the order of the binding energy, and a is the localization radius of the state), is quite accurately equal to $\hbar\omega$ (we will call such pairs "resonant" pairs).

Among these pairs, the only ones which contribute to the absorption are those which contain a single electron. Shklovskiĭ and Éfros have shown (Ref. 4; see also Ref. 2) that in order to calculate the number of such pairs we need to consider the Coulomb repulsion of the electrons (if two electrons are localized at the sites of a pair separated by a distance r, an additional repulsive energy $e^2/\varepsilon r$ arises, where ε is the dielectric constant). We consider only the case in which

this energy, for a pair with a characteristic distance r_c between sites, is much greater than $\hbar\omega$ and also much greater than the temperature T:

$$e^2/\varepsilon r_c \gg \hbar\omega, T. \tag{1.2}$$

In this case the real part of the rf hopping conductivity, which determines the linear absorption, is[4]

$$\sigma_0(\omega) = \frac{\pi^2}{3} \th \frac{\hbar\omega}{2T} ag^2 \frac{e^4}{\varepsilon} \omega r_\omega^3, \tag{1.3}$$

where g is the density of one-electron states, and

$$r_\omega = a \ln (2I_0/\hbar\omega)$$

is the minimum "arm" of a pair with a distance $\hbar\omega$ between levels [it is this quantity which plays the role of r_c in the estimate (1.2)].[1] The functional dependence $\sigma_0(\omega)$ is thus smooth and carries no information on the relaxation time of the nonequilibrium pairs. The factor $\tanh(\hbar\omega/2T)$ is the difference between the populations of the lower and upper levels of the resonant pair.

If the amplitude of the oscillating field, $\mathscr{E}_0$, is sufficiently high, this difference decreases, and the absorption falls off (i.e., there is a so-called saturation of the resonant absorption). Nonlinear experiments can thus yield information on the nonequilibrium population and relaxation of pairs in a narrow energy interval (unfortunately, nonlinear experiments have so far been rather rare[5-7]).

We will discuss two types of such experiments: nonlinear resonant absorption proper (i.e., the dependence of the absorption coefficient α for a wave on the amplitude of this wave, $\mathscr{E}_0$)[2] and "hole burning." In the latter case we will examine the frequency dependence of the absorption of a weak test signal in the presence of a pump signal with a fixed frequency ω. The pump wave leads to saturation of the populations of the resonant pairs; this saturation is in turn reflected in both the absorption of the wave itself and the transparency of the material for a test signal whose frequency ω_1 is approximately but not exactly the same. A hole is burnt out of the energy distribution of the pairs which are responsible for the resonant absorption. In both of these phenomena, a group of nonequilibrium pairs with energies close to $\hbar\omega$ appears, and their state determines the magnitude of the effect.

Estimates show that direct relaxation processes in reso-

nant pairs as a result of the emission and absorption of phonons can explain neither the order of magnitude of the nonlinear resonant effects nor their characteristic temperature and frequency dependences in most cases of interest. The reason is that these processes are usually complicated by spectral diffusion. Spectral diffusion was first discussed in Klauder and Anderson's magnetic-resonant theory.[10] Corresponding ideas were pursued in a theory of the low-temperature properties of glasses derived by Joffrin and Levelut[11] and Hunklinger and Arnold.[12] This phenomenon can be outlined as follows: We consider some resonant pair, i.e., a pair for which the value of $E/\hbar$, where E is the distance between levels, is approximately equal to the frequency of the external field, ω. A resonant pair of this sort interacts with neighboring thermal pairs, i.e., pairs for which the distance between levels is less than or on the order of the temperature T. The distance between levels in a resonant pair depends on the strength of the interaction with the thermal pairs and (the most important point) on the particular state of the given thermal pair: ground state or excited state. In turn, the interaction with the phonons causes the thermal pairs to undergo transitions to the excited state and then back to the ground state at random times. As a result, the distance between the levels of a resonant pair turns out to be a random function of the time. Transitions between the levels of thermal pairs lead to a change in the distance between the levels of a resonant pair and thus disrupt the resonance.

We pointed out in Ref. 13 that spectral diffusion can play an important role in nonlinear resonant absorption in semiconductors. However, we do not yet have a theory for this phenomenon, because the derivation runs into serious difficulties. These difficulties stem primarily from the fact that the random field created by quantum transitions of thermal pairs is a very complicated stochastic function with statistical properties which are definitely non-Gaussian.

Our purpose in the present paper is to derive a theory for spectral diffusion under conditions of nonlinear resonant absorption in a semiconductor.

To draw a clearer physical picture of the situation, let us examine which parameters will characterize this phenomenon. The interaction of an alternating field with a resonant pair is characterized by the matrix element $\hbar F/2$, where the frequency F is related to the electric field amplitude $\mathscr{E}_0$ by

$$\hbar F = e\mathscr{E}_0 \mathbf{r} 2I(r)/E, \qquad (1.4)$$

where $\mathbf{r}$ is the radius vector connecting the components of the pair (the "arm" of the pair). The quantity F is none other than the Rabi frequency for a resonant pair. This quantity characterizes the frequency of coherent oscillations of the population of a pair driven by a resonant perturbation.[14] If the interaction between pairs and the associated spectral diffusion are ignored, the resonant absorption coefficient α is determined by the relation between F and the intrinsic damping γ of a resonant pair, which results from its interaction with phonons.

The quantity γ is none other than the reciprocal of the population relaxation time τ^{-1}, which we discussed in Ref. 13. The explicit expression for γ depends on the particular mechanism by which the resonant pair interacts with phonons, and we will postpone moving to a more specific level in this regard.

The absorption coefficient of a single resonant pair is proportional to the product of the difference between the populations of the lower and upper levels of the resonant pair, on the one hand, and the spectral width of its absorption line, on the other. For $F \ll \gamma$, the population difference is independent of F in the zeroth approximation and is determined by its equilibrium value $\tanh(\hbar\omega/2T)$. The absorption line of a single resonant pair is Lorentzian with a width γ in this case. The total absorption coefficient, in the same approximation, is given by the linear theory [see (1.3)]. A correction on the order of the parameter $(F/\gamma)^2$ arises in the next approximation.

If, on the other hand, we have $F \gg \gamma$, then the difference between populations falls off in inverse proportion to F^2, i.e., the wave intensity; the width of the resonance region increases in proportion to F because of Rabi oscillations. As a result, the absorption coefficient turns out to be inversely proportional to F. The critical amplitude F_c, which determines the nonlinear effects, is thus equal to γ in this case. Correspondingly, the width of the burnt-out hole is on the order of γ for $F \ll F_c$ or on the order of F for $F \gg F_c$.

Spectral diffusion changes the picture fundamentally. Let us find the typical change in the energy of a resonant pair, E, due to transitions in a thermal pair positioned a distance R from the resonant pair. This change is equal to the energy of the interaction of two electric dipoles with moments er_ω and er_T, where r_T is the arm of the thermal pair which is separated by a distance R: $e^2 r_\omega r_T/\varepsilon R^3 \equiv A/R^3$. The total energy of the interaction with all thermal pairs is on the order of $A/\bar{R}^3$, where $\bar{R}$ is the mean distance between thermal pairs. As we will see below, the only pairs which contribute to the spectral diffusion are those which can undergo transitions. As was shown in Ref. 13 [see also (9.2)], the relaxation time of the pairs increases rapidly with increasing energy E if the latter is greater than a characteristic value $E_a = 2\hbar w/a$, where w is the sound velocity. If the temperature satisfies $T < E_a$, the density of excited thermal pairs increases in proportion to the temperature:

$$1/\bar{R}^3 = P_T T. \qquad (1.5)$$

The quantity $P_T = 4\pi g^2(e^2/\varepsilon r_T)ar_T^2$ plays the role of a state density of the thermal pairs. It depends weakly (logarithmically) on the temperature.

If, on the other hand, we have $T > E_a$, we can assume that the transitions of thermal pairs with $E > E_a$ occur so rarely that they have no effect on spectral diffusion. In this case we would need to replace the temperature T by E_a in the expression for P_T.

The quantity $A/\bar{R}^3$ is an important characteristic of spectral diffusion. It tells us the width of the interval in which the deviation of the resonant pair from resonance, $E - \hbar\omega$, undergoes its random walk. To denote it we introduce a quantity with the dimensionality of a time, τ_d: $A/\bar{R}^3 \approx \hbar/\tau_d$ [see (4.5)]. For $T < E_a$ we have

$$\hbar/\tau_d \approx \eta_T T, \qquad (1.6)$$

where

$$\eta_T \approx g^2 e^4 \varepsilon^{-2} r_\omega r_T^2 a. \qquad (1.7)$$

In the case of a lightly doped semiconductor with an intermediate degree of compensation, we can use a more spe-

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cific expression for the electron state density g (Refs. 2 and 13), and we find

$$\eta_T \approx N^{4/3} r_\omega r_T^2 a. \qquad (1.8)$$

To find the dimensionless parameter η_T, we note that when the two-site model is applicable the quantities r_T and r_ω should be substantially smaller than the average distance between impurities $N^{-1/3}$. It thus follows from explicit expression (1.8) that where the two-site approximation is applicable we have

$$\eta_T \ll 1. \qquad (1.9)$$

There is also another mechanism which operates to change the distance between the energy levels of a resonant pair: a mechanism involving the elastic stress fields set up by the thermal pairs. Estimates show[13] that the contribution of this mechanism is smaller than (1.6) and (1.7) in order of magnitude.

In several interesting experimental situations the value of $1/\tau_d$ turns out to be significantly larger than γ or F, so the dynamic broadening of the spectral line of a resonant pair is inconsequential, and spectral diffusion plays the leading role.

As we will see, an important role is played throughout the phenomenon of spectral diffusion by the relation between $1/\tau_d$ and the characteristic frequency of transitions of the thermal pairs, Γ_0. The frequency of transitions in a pair depends on the distance between the levels of the pair, so γ and Γ_0 are different quantities, and the product $\Gamma_0 \tau_d$ is a function of the temperature. Expressions for Γ_0 for various interaction mechanisms are given in Ref. 13 and also in the final section of the present paper.

The appearance of the 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 excursion of a resonant pair from resonance increases linearly with the time[3]:

$$|E(t) - E(0)| \approx \hbar \Gamma_0 t / \tau_d. \qquad (1.10)$$

To see the origin of this relation[10] we consider a volume with linear dimensions on the order of R_t around a resonant pair. This volume contains $\approx (R_t/\bar{R})^3$ thermal pairs with characteristic transition frequencies of order Γ_0. In this volume, by the time t, a jump of at least one thermal pair occurs with a probability of order unity if R_t satisfies the condition $(R_t/\bar{R})^3 \Gamma_0 t \approx 1$. The characteristic change in the energy of the resonant pair which corresponds to this jump is

$$A/R_t^3 \approx \hbar \Gamma_0 t / \tau_d. \qquad (1.11)$$

Relation (1.10) follows immediately. The time scale τ_φ for the phase relaxation of the wave function of a resonant pair is thus given by

$$\tau_\varphi = (\tau_d/\Gamma_0)^{1/2}. \qquad (1.12)$$

This expression is valid if the time τ_φ is shorter than the time scale between jumps of thermal pairs, Γ_0^{-1}:

$$\Gamma_0 \tau_d \ll 1. \qquad (1.13)$$

If, on the other hand, the relation $\Gamma_0 \tau_d \gg 1$ holds, the phase of the wave function of a resonant pair will have time to change by only a small amount over the time interval $t \lesssim \Gamma_0^{-1}$; i.e., there will be essentially no phase relaxation. In this case the time scale of the phase relaxation is therefore $\tau_\varphi \gg \Gamma_0^{-1}$. On the other hand, after a long time $t \gg \Gamma_0^{-1}$ the typical value of the difference $|E(t) - E(0)|$ ceases to depend on the time, since this difference cannot exceed $\hbar/\tau_d$ in order of magnitude. In other words, the difference undergoes a random walk over an interval $\hbar/\tau_d$ in this case. Correspondingly, the phase relaxation time 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 apparent from this discussion that there are two regions—of high and low temperatures—in comparison with the characteristic temperature T_d. The latter is determined by equating the characteristic parameter $\Gamma_0 \tau_d$ to unity. Its value depends on the particular parameter values of the semiconductor. Ordinarily, $\Gamma_0 \tau_d$ is an increasing function of the temperature, and the high-temperature case $T \gg T_d$ corresponds to the condition $\Gamma_0 \tau_d \gg 1$, and the low-temperature case $T \ll T_d$ to the condition $\Gamma_0 \tau_d \ll 1$.

Let us find the characteristic (critical) amplitude (F_c) for the saturation of the resonant absorption, and also the width of the burnt-out hole, in cases in which spectral diffusion is important. Under the conditions $T > T_d$ and $1/\tau_d \gg \gamma$, because of the frequent jumps of thermal pairs, all the resonant pairs from a spectral interval of width $\hbar/\tau_d$ are away from equilibrium. The typical rate of change of the population is $F^2 \tau_d$, and the rate of relaxation due to thermal phonons is γ. A comparison of these quantities yields the estimate

$$F_c \approx (\gamma/\tau_d)^{1/2} \gg \gamma, \qquad (1.14)$$

while the width of the burnt-out hole is evidently $1/\tau_d$, which is in turn much larger than F_c.

At $T \ll T_d$ we can distinguish two limiting cases in which spectral diffusion is important:

$$\Gamma_0 \ll \gamma \ll (\Gamma_0/\tau_d)^{1/2}, \qquad (1.15)$$

$$\gamma \ll \Gamma_0 \ll (\Gamma_0/\tau_d)^{1/2}. \qquad (1.16)$$

In the first of these cases the critical amplitude is found from the condition that over a time $1/F$, on the order of the period of the Rabi oscillations, the energy E of a resonant pair deviates by an amount of order $\hbar F$ as the result of spectral diffusion, according to (1.10). We then find the estimate

$$F_c \approx (\Gamma_0/\tau_d)^{1/2}. \qquad (1.17)$$

An estimate of the width of the burnt-out hole is then found on the basis of the following considerations. As a resonant pair crosses a resonance region with a width on the order of $\hbar(\Gamma_0/\tau_d)^{1/2}$, it is excited at $F \gtrsim F_c$ with a probability of order unity. It then leaves the resonance region in accordance with (1.10), remaining in an excited state for yet another time interval $t \approx \gamma^{-1} \ll \Gamma_0^{-1}$. Substituting this time into (1.10), we reach the conclusion that the width of the burnt-out hole is of order $\Gamma_0/\gamma \tau_d$.

In the second case—(1.16)—the critical intensity can be estimated on the basis of the following qualitative picture.[15] The region of the random changes in the energy of a resonant pair, $\hbar/\tau_d$ is much greater than the width of the resonance, $\hbar(\Gamma_0/\tau_d)^{1/2}$, in this case. During random

changes in the energy E, a resonant pair returns repeatedly to the resonance region. On each occasion, its population increases by a small amount

$$F^2\tau_\varphi^2=F^2\tau_d/\Gamma_0\ll1.$$

The total number of such returns over the lifetime $1/\gamma$ is $\Gamma_0/\gamma\gg1$, so the total change in the population over this time is $(F^2\tau_d/\Gamma_0)(\Gamma_0/\gamma)$. Equating this quantity to unity, we find an estimate of F_c,

$$F_c\approx(\gamma/\tau_d)^{1/2},\tag{1.18}$$

which is the same as the estimate at high temperatures. The width of the burnt-out hole is on the order of $1/\tau_d$.

The detailed calculation carried out below shows that the exact expressions for F_c actually differ from estimates (1.14), (1.17), and (1.18) by large logarithmic factors, which depend on the temperature T, the frequency ω, and the parameters of the semiconductor.

At high intensities, $F\gg F_c$, the power Q absorbed by the resonant pairs in a unit volume does not depend on F in any case in which spectral diffusion is important, as is shown below in Sec. 8. It follows that the absorption coefficient α falls off with increasing intensity in proportion to F^{-2}, i.e., in inverse proportion to the intensity. In the absence of a spectral diffusion we would have $\alpha\sim F^{-1}$; i.e., the absorption coefficient would be inversely proportional to the square root of the intensity.

2. BASIC EQUATIONS AND LIMITING CASES

The coefficient of the absorption by resonant pairs with a level separation E close to $\hbar\omega$ is given by the density matrix

$$\begin{pmatrix} n & -ife^{i\omega t} \\ if^*e^{-i\omega t} & 1-n \end{pmatrix}.\tag{2.1}$$

In the resonant approximation, the equations for the elements of this matrix are

$$\partial n/\partial t=-\gamma(n-n_0)-F\,\mathrm{Re}\,f,\tag{2.2}$$

$$\partial f/\partial t+i[\omega-\hbar^{-1}E(t)]f+\tfrac{1}{2}\gamma f=\tfrac{1}{2}F(2n-1).\tag{2.3}$$

Here

$$n_0=[\exp(E/T)+1]^{-1}\tag{2.4}$$

is the equilibrium population of the upper level of the resonant pair, $\hbar F/2$ is the matrix element for a transition of this pair driven by a periodic external field of an electromagnetic wave, γ is the total damping of the resonant pair due to the emission and absorption of phonons of energy E (the phonons are assumed to be in equilibrium),

$$E(t)=E+\hbar\Delta\omega(t),\tag{2.5}$$

$$\Delta\omega(t)=\sum_l J_l\xi_l(t),\tag{2.6}$$

$\hbar J_l$ is the energy of the interaction of a resonant pair with the lth thermal pair, and ξ_l is a random function of the time, described by a so-called telegraphic process. It 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, so that we can carry out independent averaging steps over these functions. Here we can directly see the validity of our earlier assertion that the

random process described by the function $\Delta\omega(t)$ is non-Gaussian.

The absorption of the oscillating field by a resonant pair is determined by the imaginary part of the susceptibility, $\mathrm{Im}\,\chi_E(\omega)$, which is related to a nondiagonal component f of the density matrix by

$$\mathrm{Im}\,\chi_E(\omega)=-2F^{-1}\,\mathrm{Re}\langle f\rangle_\xi.\tag{2.7}$$

Here the angle brackets denote an average over realizations of all the telegraphic processes $\xi_l(t)$. The total susceptibility is determined by the sum of the contributions (2.7) of all the resonant pairs in a unit volume (V is the volume):

$$\mathrm{Im}\,\chi(\omega)=-\frac{2}{FV}\sum_{\mathrm{res}}\mathrm{Re}\langle f\rangle_\xi.\tag{2.8}$$

We assume that the arrangement of the thermal pairs and also the transition frequencies of these pairs, Γ_l, are uncorrelated with the parameters of the resonant pairs. In this case expression (2.8) becomes

$$\mathrm{Im}\,\chi(\omega)=-\frac{2P_\omega}{F}\int_0^\infty dE\,\mathrm{Re}\,\langle\langle f\rangle_\xi\rangle_c.\tag{2.9}$$

where

$$P_\omega=g^2\,\frac{e^2}{\varepsilon r_\omega}\,4\pi a r_\omega^2\tag{2.10}$$

plays the role of an effective state density of the resonant pairs, and $\langle\ldots\rangle_c$ means a configurational average over the parameters and arrangement of the thermal pairs.

Strictly speaking, we should have also taken an average over the tunnel transparencies $I(r)$ of the resonant pairs, on which the quantities F and γ depend. However, it can be verified directly that this average leads to nothing fundamentally new, and we have omitted it to avoid making the equations overly complicated.

3. ITERATIONS IN THE FIELD AMPLITUDE

The solution of Eq. (2.2) for the filling of the upper level n of the resonant pair can be written formally as

$$n=n_0-\int_0^\infty dt'\,e^{-\gamma t'}\,\mathrm{Re}\,f(t-t').\tag{3.1}$$

Substitution of this solution into (2.3) yields the following integral equation for

$$\varphi(t)=\frac{2}{F(2n_0-1)}\,\mathrm{Re}\,f(t):$$

$$\varphi(t)=\int_0^\infty dt'\,R(t,t')-F^2\int_0^\infty dt'\,R(t,t')\int_0^\infty dt''\,e^{-\gamma t''}\varphi(t-t'-t''),\tag{3.2}$$

where

$$R(t,t')=\exp(-\gamma t'/2)\cos\left[zt'-\int_{t-t'}^t\Delta\omega(t'')dt''\right],\tag{3.3}$$

and

$$Z=\omega-E/\hbar$$

is the deviation from resonance.

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Taking iterations of (3.2) in powers of F^2, we find

$$\varphi = \varphi^{(0)} + \varphi^{(1)},\qquad (3.4)$$

$$\varphi^{(0)} = \int_0^\infty dt'\, R(t,t'),\qquad (3.5)$$

$$\varphi^{(1)}(t) = -F^2 \int_0^\infty dt'\, R(t,t') \int_0^\infty dt''\, e^{-\gamma t''} \int_0^\infty dt'''\, R(t-t'-t''-t''').\qquad (3.6)$$

In addition to the nonlinear absorption, we are interested in the problem of the shape of the burnt-out hole. We can write a correction to the population of the resonant pairs, which we need in order to solve this problem:

$$\Delta n^{(1)} = -F^2\left(n_0 - \frac{1}{2}\right)\int_0^\infty dt'\, e^{-\gamma t'}\int_0^\infty dt''\, R(t-t',t'').\qquad (3.7)$$

Substituting (3.4) into (2.9), and integrating over E, we find the following result in the resonant approximation (which allows us to carry out the integration over z from $-\infty$ to $+\infty$):

$$\mathrm{Im}\,\chi(\omega) = \pi\hbar P_\omega\left(1 - \frac{F^2}{F_c^2}\right)\mathrm{th}\frac{\hbar\omega}{2T},\qquad (3.8)$$

where

$$F_c^{-2} = \int_0^\infty d\tau \int_0^\infty d\tau'\, \exp\{-\gamma(\tau+\tau')\}\langle K(\tau,\tau')\rangle_c,\qquad (3.9)$$

$$K(\tau,\tau') = \left\langle\cos\left[-\int_{t-\tau}^t \Delta\omega(t')dt' + \int_{t-2\tau-\tau'}^{t-\tau-\tau'} \Delta\omega(t')dt'\right]\right\rangle_\xi.\qquad (3.10)$$

The quantity K is now independent of the time t. In calculating the average over the telegraphic processes, it is convenient to set $t = 2\tau + \tau'$ and to write the function $K(\tau,\tau')$ in the form

$$K(\tau,\tau') = \left\langle\cos\left[\int_0^{2\tau+\tau'} \Delta\omega(t)v(t)dt\right]\right\rangle,\qquad (3.11)$$

where

$$v(t) = \begin{cases} 1 & 0\leqslant t\leqslant\tau, \\ 0 & \tau < t\leqslant\tau+\tau', \\ -1 & \tau+\tau'<t\leqslant 2\tau+\tau'. \end{cases}\qquad (3.12)$$

The function $K(\tau,\tau')$ becomes equal to unity, and we have $F_c^2 = \gamma^2$, if $\Delta\omega(t)$ does not depend on the time. This result reflects the fact (which we mentioned back in the Introduction) that only those thermal pairs which are capable of undergoing transitions contribute to the spectral diffusion.

Let us examine the function

$$\psi(t) = \left\langle\exp\left[i\int_0^t \Delta\omega(t')v(t')dt'\right]\right\rangle_\xi,\qquad (3.13)$$

which is related to (3.11) by

$$K(\tau,\tau') = \mathrm{Re}\,\psi(2\tau+\tau').\qquad (3.14)$$

Since the contributions of the thermal pairs to $\Delta\omega(t)$ are additive, the average value (3.13) can be broken up into a

product of averages over independent telegraphic processes in each of the thermal pairs:

$$\psi(t) = \prod_l k_l(t).\qquad (3.15)$$

We will thus first calculate the average value in the case of a single telegraphic process: $\Delta\omega(t) = J\xi(t)$ (we omit the index l).

As was shown in the book by Klyatskin,[16] the function $k(t)$ satisfies the integrodifferential equation

$$\frac{dk}{dt} = -J^2 v(t)\int_0^t dt'\, \exp\{-2\Gamma(t-t')\}v(t')k(t').\qquad (3.16)$$

This equation is equivalent to the differential equation

$$\frac{d^2 k}{dt^2} + \left[2\Gamma - \frac{d\ln v(t)}{dt}\right]\frac{dk}{dt} + J^2 v^2(t)k = 0\qquad (3.17)$$

with the initial conditions

$$k(0) = 1,\qquad dk/dt|_{t=0} = 0.\qquad (3.18)$$

According to (3.12), $v(t)$ is a piecewise-constant function. At those points where it is discontinuous, the function $k(t)$ is continuous, and its derivative is found from integral equation (3.16). We have

$$k(2\tau+\tau') = \exp(-2\Gamma\tau)\left\{\mathrm{ch}[(\Gamma^2-J^2)^{1/2}\tau]\right.$$
$$\left. + \frac{\Gamma}{(\Gamma^2-J^2)^{1/2}}\mathrm{sh}[(\Gamma^2-J^2)^{1/2}\tau]\right\}^2$$
$$+ \frac{J^2}{\Gamma^2-J^2}\exp\{-2\Gamma(\tau+\tau')\}\mathrm{sh}^2[(\Gamma^2-J^2)^{1/2}\tau].\qquad (3.19)$$

We wish to stress that expression (3.19) is valid for both $\Gamma > J$ and $\Gamma < J$. It is real in both cases, so the sign specifying the real part can be omitted from (3.14). The reason for this result is that the average value of a functional which is an odd function on the telegraphic process $\xi(t)$ is zero.

4. CONFIGURATIONAL AVERAGE

Our problem thus reduces to one of calculating the average of the quantity $K(\tau,\tau')$ over the configurations of the thermal pairs. This quantity is in turn the product of average values over telegraphic processes characterizing different thermal pairs.

The effect of a thermal pair on a resonant pair is determined by two factors: the distance to the thermal pair, R, and the frequency of its jumps, Γ. Finding the configurational average thus actually reduces to taking an average over these two quantities. With regard to the average over R, we assume that all the spatial positions of a thermal pair are equally probable. The distribution in Γ [i.e., actually the distribution in the tunnel transparency $I(r)$] is of the form $1/\Gamma(1 - \Gamma/\Gamma_0)^{1/2}$, where Γ_0 is the maximum frequency of the jumps of the thermal pair. For simplicity, we replace the expression in parentheses by unity; this simplification has no substantial effect on the results.

Using Holtmark's method to calculate the average (see, for example, the review of Chandrasekhar[17]), we find

$$\langle K(\tau, \tau')\rangle_c = \exp\left\{-\frac{1}{\bar{R}^3}\int_0^\infty d^3R\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}[1-k(2\tau+\tau')]\right\}. \tag{4.1}$$

Using

$$\hbar J = A/R^3, \tag{4.2}$$

we can put (4.1) in the form

$$\langle K(\tau, \tau')\rangle_c = \exp[-S(\tau, \tau')/\tau_d], \tag{4.3}$$

$$S(\tau, \tau') = \int_0^\infty\frac{dJ}{J^2}\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}[1-k(2\tau+\tau')], \tag{4.4}$$

and

$$\hbar/\tau_d = 4\pi/3\bar{R}^3 \tag{4.5}$$

is the characteristic width of the region of spectral diffusion. This width is equal in order of magnitude to the typical change in the energy E due to the effect of a thermal pair at a characteristic distance $\bar{R}$.

5. STUDY OF LIMITING CASES

The quantity $\langle K(\tau,\tau')\rangle_c$ given by (4.3) depends on the ratio between the characteristic value of Γ (equal to Γ_0) and the characteristic value of J (equal to $1/\tau_d$). It is thus natural to consider the two limiting cases $\Gamma_0\tau_d\gg 1$ and $\Gamma_0\tau_d\ll 1$. Just which of these cases is realized depends on the relation between the temperature T and its characteristic value T_d, at which we have $\Gamma_0\tau_d = 1$.

At high temperatures $(T\gg T_d)$ we have $\Gamma_0\tau_d\gg 1$. In this case, the integration over Γ in (4.4) can be extended to infinity. As a result we find

$$S(\tau, \tau') = \tau\sigma(\tau'/\tau). \tag{5.1}$$

We will not need the explicit functional dependence $\sigma(\beta)$ below; all that we will need are the following properties of this dependence. In the limit $\beta\to 0$, the function $\sigma(\beta)$ tends toward a constant limit on the order of unity, while at $\beta\gg 1$ we find the following result from (4.4), to logarithmic accuracy:

$$\sigma(\beta) = \frac{\pi}{2}\ln\beta. \tag{5.2}$$

Using (5.1), and substituting (4.3) into (3.9), we find

$$F_c^{-2} = \int_0^\infty d\tau\int_0^\infty d\tau'\exp\left[-\gamma(\tau+\tau')-\frac{\tau}{\tau_d}\sigma\left(\frac{\tau'}{\tau}\right)\right]. \tag{5.3}$$

Introducing the new integration variable $\beta = \tau'/\tau$, and integrating over τ, we find

$$F_c^{-2} = \int_0^\infty\frac{d\beta}{[\gamma(1+\beta)+\sigma(\beta)/\tau_d]^2}. \tag{5.4}$$

The integral (5.4) can be analyzed easily in two limiting cases. For $\gamma\gg 1/\tau_d$, spectral diffusion is unimportant, and we have $F_c = \gamma$. In the opposite limit,

$$\gamma\ll 1/\tau_d, \tag{5.5}$$

values $\beta\approx(\gamma\tau_d)^{-1}\ln(\gamma\tau_d)^{-1}$ contribute to integral (5.4), and we have, at a logarithmic accuracy level,

$$F_c^2 = \frac{\pi\gamma}{2\tau_d}\ln\frac{1}{\gamma\tau_d}. \tag{5.6}$$

The origin of the factor of $1/\tau_d$ in the argument of the logarithm can be illustrated by the following discussion. Thermal pairs close to a resonant pair can be classified as "fast," "intermediate," or "slow," depending on the frequency of their jumps, Γ. We classify as "fast" pairs for which we have $\Gamma\gg 1/\tau_d$. As is clear from the calculations above, these pairs play only a minor role in spectral diffusion. What is happening here is that their effect is being averaged out (in a way similar to the effect which occurs during dynamic line contraction in magnetic resonance theory[18]).

The very slowest thermal pairs, for which the jump frequency is much smaller than γ, are also unimportant, since over the decay time of a resonant two-level system they do not have time to undergo even a single jump. In other words, they represent a frozen configuration from the standpoint of the resonant pair.

We thus see that the "intermediate" pairs, for which the jump frequency lies between γ and $1/\tau_d$, are the effective ones. Accordingly, there is a large logarithm in expression (5.6).

We now turn to low temperatures $T\ll T_d$ $(\Gamma_0\tau_d\ll 1)$. Calculations yield

$$S(\tau, \tau') = \frac{\pi}{2}\tau\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}[1-\exp\{-2\Gamma(\tau+\tau')\}]. \tag{5.7}$$

Hence

$$F_c^{-2} = \int_0^\infty d\tau\int_0^\infty d\tau'\exp\left\{-\gamma(\tau+\tau')\right.$$
$$\left.-\frac{\pi\tau}{2\tau_d}\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}[1-\exp\{-2\Gamma(\tau+\tau')\}]\right\}. \tag{5.8}$$

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. For $\gamma\gg(\Gamma_0/\tau_d)^{1/2}$, τ and τ' values on the order of $1/\gamma$ play a role in integral (5.8), and the second term in braces can be ignored in comparison with the first. This statement means that spectral diffusion plays no role, and we have $F_c = \gamma$.

In the interval

$$\Gamma_0\ll\gamma\ll(\Gamma_0/\tau_d)^{1/2} \tag{5.9}$$

the exponential function in the brackets can be expanded. The double integral can then be evaluated easily; we find

$$F_c^2 = 2\pi\Gamma_0/\tau_d\ln(\Gamma_0/\gamma^2\tau_d). \tag{5.10}$$

This result agrees to within the large logarithm with the qualitative estimate (1.17).

In the case

$$\gamma\ll\Gamma_0\ll(\Gamma_0/\tau_d)^{1/2} \tag{5.11}$$

the integral (5.8) is dominated by values $\tau\lesssim\tau_d$, and we have $\tau'\lesssim\gamma^{-1}$. In other words, the characteristic values of τ' are much greater than τ. The integral over Γ depends logarithmically on τ'. Carrying out the integration, we find the following expression for F_c, with logarithmic accuracy:

$$F_c^{-2} = (\tau_d/2\pi\Gamma_0)\ln(\Gamma_0\tau_d)^{-1} + 2\tau_d/\pi\gamma\ln(\Gamma_0/\gamma). \quad (5.12)$$

If

$$\gamma < 4\Gamma_0/\ln(1/\Gamma_0\tau_d)\ln(\Gamma_0/\gamma), \quad (5.13)$$

the second term in (5.12) is greater than the first, and for F_c we find

$$F_c^2 = (\pi\gamma/2\tau_d)\ln(\Gamma_0/\gamma). \quad (5.14)$$

This result agrees to within the large logarithm with the qualitative estimate (1.18)

6. THEORY OF THE BURNT-OUT HOLE

The change ΔQ in the absorption of a weak test signal of frequency ω_1 in the presence of a strong signal, which causes a population change $\Delta n^{(1)}$ is

$$\Delta Q = -\pi\omega_1 V^{-1}\sum_{\text{res}}\hbar^2 F_1^2 \Delta n^{(1)}(t)\delta(\hbar\omega_1 - E(t)), \quad (6.1)$$

where the summation is over all the resonant pairs, $E(t) = E + \hbar\Delta\omega(t)$, and ΔQ is a macroscopic quantity. To within small fluctuations which are of no importance here, this quantity is equal to its mean value, which we will also calculate (the average is taken over the thermal surroundings of each resonant pair and over all possible realizations of the telegraphic processes describing transitions in thermal pairs). We thus have

$$\Delta Q = -\pi\omega\hbar^2 F_1^2 P_\omega \int_c dE \,\langle\langle\Delta n^{(1)}(t)\delta[\hbar\omega_1 - E(t)]\rangle_\xi\rangle_c. \quad (6.2)$$

In the resonant approximation, the frequency deviation $\nu = \omega - \omega_1$ is assumed to be small in comparison with the fundamental frequency ω, so we have replaced ω_1 by ω in front of the summation. Substituting in $\Delta n^{(1)}$ from (3.7), we find

$$\Delta Q = -B \int_0^\infty d\tau\, e^{-\Gamma\tau/2} \int_0^\infty d\tau'\, e^{-\Gamma\tau'} \left\langle\left\langle \cos\left[\nu\tau + \tau\Delta\omega(t) - \int_{t-\tau-\tau'}^{t-\tau'} \Delta\omega(t')dt'\right]\right\rangle_\xi\right\rangle_c, \quad (6.3)$$

where B is shorthand for the quantity

$$B = {}^1/_2\pi\omega\hbar^2 F_1^2 F^2 P_\omega\,\text{th}(\hbar\omega/2T). \quad (6.4)$$

Since the average over ξ of a functional which is an odd function of the telegraphic process $\xi(t)$ vanishes, we can write ΔQ as

$$\Delta Q = -B \int_0^\infty d\tau\, e^{-\Gamma\tau/2}\cos\nu\tau\int_0^\infty d\tau'\, e^{-\Gamma\tau'}\langle L(\tau,\tau')\rangle_c, \quad (6.5)$$

where

$$L(\tau,\tau') = \left\langle\exp\left[i\tau\Delta\omega(t) - i\int_{t-\tau-\tau'}^{t-\tau}\Delta\omega(t')dt'\right]\right\rangle_\xi. \quad (6.6)$$

Since the results of the averaging do not depend on t, we set $t = 0$. We then proceed as in the derivation of (3.19). Specifically, we make use of the additivity of the contributions of the thermal pairs to $\Delta\omega(t)$, and we first calculate the mean value (6.6) in the case of a single telegraphic process:

$$\varphi(\tau,\tau') = \left\langle\exp\left[iJ\tau\xi(0) - iJ\int_{\tau'}^{\tau+\tau'}\xi(t)dt\right]\right\rangle_\xi. \quad (6.7)$$

Here we have made use of the circumstance that for the telegraphic process $\xi(t)$ there is no preferred direction for time, so it can be assumed to be an even function of t during the averaging. We introduce the auxiliary function

$$M(t) = \left\langle\exp\left[-iJ\int_0^t\xi(t')v_1(t')dt'\right]\right\rangle_\xi\bigg|_{\xi(0)}, \quad (6.8)$$

where the function $v_1(t)$ is equal to 1 at $\tau' \leqslant t \leqslant \tau + \tau'$ and 0 in the interval $0 \leqslant t < \tau'$. The symbol $|_{\xi(0)}$ means that the mean value is calculated at a fixed value of $\xi(0)$. For a single telegraphic process, the expression for $\varphi(\tau,\tau')$ takes the form

$$\varphi(\tau,\tau') = \langle\exp[iJ\tau\xi(0)]M(\tau+\tau')\rangle_{\xi(0)}, \quad (6.9)$$

where the average is taken over the value of $\xi(0)$, which has the values ± 1 with probabilities of $1/2$.

The equation for the function $M(t)$ is the same as (3.17), and the conditions at the point $t = \tau'$ are

$$M(\tau') = 1, \quad dM/dt|_{t=\tau'+0} = -iJ\xi(0)\exp(-2\Gamma\tau'). \quad (6.10)$$

Solving this equation, we find

$$M(\tau+\tau') = e^{-\Gamma\tau}\{\text{ch}[(\Gamma^2-J^2)^{1/2}\tau] + \Gamma(\Gamma^2-J^2)^{-1/2}\text{sh}[(\Gamma^2-J^2)^{1/2}\tau] - i\xi(0)J(\Gamma^2-J^2)^{-1/2}\exp(-2\Gamma\tau')\text{sh}[(\Gamma^2-J^2)^{1/2}\tau]\}. \quad (6.11)$$

Substituting this expression into (6.9), and then taking an average over the initial value $\xi(0)$, we find

$$\varphi(\tau,\tau') = e^{-\Gamma\tau}\cos J\tau\,\{\text{ch}[(\Gamma^2-J^2)^{1/2}\tau]$$
$$+ \Gamma(\Gamma^2-J^2)^{-1/2}\text{sh}[(\Gamma^2-J^2)^{1/2}\tau]\}$$
$$+ \exp\{-\Gamma(\tau+2\tau')\}J(\Gamma^2-J^2)^{-1/2}\sin J\tau\,\text{sh}[(\Gamma^2-J^2)^{1/2}\tau]. \quad (6.12)$$

Taking the configurational average by the procedure described in the derivation of (4.3), we find

$$\langle L(\tau,\tau')\rangle_c = \exp[-V(\tau,\tau')/\tau_d], \quad (6.13)$$

where

$$V(\tau,\tau') = \int_0^\infty \frac{dJ}{J^2}\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}[1-\varphi(\tau,\tau')]. \quad (6.14)$$

7. SHAPE OF THE BURNT-OUT HOLE: STUDY OF LIMITING CASES

At high temperatures $T \gg T_d$ ($\Gamma_0\tau_d \gg 1$), the upper limit on the integral over Γ on (6.14) can be replaced by ∞:

$$V(\tau,\tau') = \tau v(\tau'/\tau). \quad (7.1)$$

We will not need the explicit functional dependence $v(\beta)$. The only results of importance to an analysis of limiting cases are

$$v(\beta) = \begin{cases} v(0) \approx 1 & \beta \to 0 \\ (\pi/2)\ln\beta & \beta \to \infty \end{cases} \tag{7.2}$$

Substituting (6.13) into (6.5), we find

$$\Delta Q = -B \int_0^\infty d\tau \exp(-\gamma\tau/2)\cos\nu\tau$$

$$\times \int_0^\infty d\tau' \exp[-\gamma\tau' - \tau v(\tau'/\tau)/\tau_d]. \tag{7.3}$$

We introduce the new integration variable $\beta = \tau'/\tau$, and we integrate over τ:

$$\Delta Q = -4B \int_0^\infty d\beta \frac{[(1+2\beta)\gamma + v(\beta)/\tau_d]^2 - 4\nu^2}{\{[(1+2\beta)\gamma + 2v(\beta)/\tau_d]^2 + 4\nu^2\}^2}. \tag{7.4}$$

For $\gamma \gg 1/\tau_d$, the spectral diffusion is unimportant, and we have

$$\Delta Q = -B/2(\nu^2 + \gamma^2/4). \tag{7.5}$$

If, on the other hand, we have $\gamma \ll 1/\tau_d$, then large values of β are important in integral (7.4), and we have

$$\Delta Q = -\frac{B}{\gamma}\frac{L/\tau_d}{\nu^2 + (L/\tau_d)^2}, \tag{7.6}$$

where

$$L = \frac{\pi}{2}\ln\frac{1}{\gamma\tau_d}. \tag{7.7}$$

At low temperatures $T \ll T_d$ ($\Gamma_0\tau_d \ll 1$), calculations yield

$$V(\tau,\tau') = \frac{\pi}{2}\tau \int_0^{\Gamma_0} \frac{d\Gamma}{\Gamma}[1 - \exp\{-\Gamma(\tau+2\tau')\}] \tag{7.8}$$

and

$$\Delta Q = -B \int_0^\infty d\tau \int_0^\infty d\tau' \cos\nu\tau \exp\left\{-\frac{\gamma}{2}(\tau+\tau')\right.$$

$$\left. -\frac{\pi\tau}{2\tau_d}\int_0^{\Gamma_0}\frac{d\Gamma}{\Gamma}[1-\exp\{-\Gamma(\tau+2\tau')\}]\right\}. \tag{7.9}$$

For $\gamma \gg (\Gamma_0/\tau_d)^{1/2}$, the quantity $\tau \approx \tau' \approx \gamma^{-1}$ plays a role in integral (7.9), and the second term in braces can be ignored. This result means that spectral diffusion plays no role, and we find the result (7.5).

Under the conditions

$$\Gamma_0 \ll \gamma \ll (\Gamma_0/\tau_d)^{1/2} \tag{7.10}$$

we can expand the exponential function in brackets in (7.9); it then becomes a simple matter to evaluate the integral over τ'. We have

$$\Delta Q = -B \int_0^\infty d\tau \cos\nu\tau \frac{\exp(-\pi\Gamma_0\tau^2/2\tau_d)}{\gamma + (\pi\tau\Gamma_0/\tau_d)}$$

$$= -\frac{B\tau_d}{\pi\Gamma_0}\begin{cases} \ln[(\Gamma_0/\tau_d)^{1/2}/\gamma] \gg 1, & |\nu| \ll (\Gamma_0/\tau_d)^{1/2}, \\ \ln(\Gamma_0/|\nu|\gamma\tau_d) \gg 1, & (\Gamma_0/\tau_d)^{1/2} \ll |\nu| \ll \Gamma_0/\gamma\tau_d, \\ (\Gamma_0/\gamma\tau_d\nu)^2 \ll 1, & |\nu| \gg \Gamma_0/\gamma\tau_d. \end{cases} \tag{7.11}$$
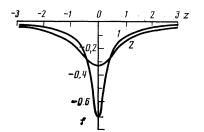

FIG. 1. 1—Shape of the burnt-out hole under the conditions $T \ll T_d$, $\Gamma_0 \ll \gamma \ll (\Gamma_0/\tau_d)^{1/2}$ [$\gamma^{-1}(2\pi\Gamma_0/\tau_d)^{1/2} = 10$]; 2—Lorentzian shape with the same asymptotic behavior and the same area:

$$f(z) = \frac{1}{\pi}\int_0^\infty dx \cos xz \frac{\exp[-(x/10)^2]}{1+x}; \quad (1)$$

$$f(z) = -\frac{1}{\pi(z^2+1)}. \quad (2)$$

It follows from (7.11) that the shape of the burnt-out hole is characterized by two energy scales (Fig. 1). In the region $|\nu| < (\Gamma_0/\tau_d)^{1/2}$ we have a plateau at a level

$$-\frac{\tau_d}{\pi\Gamma_0}B\ln\left[\frac{1}{\gamma}\left(\frac{\Gamma_0}{\tau_d}\right)^{1/2}\right] \ll -B\frac{\tau_d}{\Gamma_0}.$$

As $|\nu|$ is then increased, the quantity ΔQ increases logarithmically, reaching a value on the order of $-B\tau_d/\Gamma_0$ at $|\nu| \approx \Gamma_0/\gamma\tau_d$. As the deviation $|\nu|$ is increased further, ΔQ tends toward zero $\sim \nu^{-2}$. The characteristic size of the Lorentzian wings is again $\Gamma_0/\gamma\tau_d$. The effective width of the burnt-out hole is therefore $\Gamma_0/\gamma\tau_d$ (we are ignoring logarithmic factors). The physical meaning of these characteristic sizes can be summarized as follows: The first, $\hbar(\Gamma_0/\tau_d)^{1/2}$, is the width of the resonance region in which the pump field causes a significant change in the population of a resonant pair. According to (1.10), the time scale for the crossing of this region by a resonant pair is $(\tau_d/\Gamma_0)^{1/2}$ [i.e., τ_φ; see (1.12)]. In the case at hand, this transit time is far shorter than the lifetime γ^{-1}. Accordingly, after traversing the resonance region the pair remains excited for another time interval γ^{-1}. Over this time, the energy of the pair reaches the value $\hbar\Gamma_0/\gamma\tau_d \gg \hbar(\Gamma_0/\tau_d)^{1/2}$, according to (1.10). This is the second characteristic size, and we call it the "width of the burnt-out hole."

Finally, for

$$\gamma \ll \Gamma_0 \ll (\Gamma_0/\tau_d)^{1/2} \tag{7.12}$$

values $\tau \approx \tau_d/\ln(\Gamma_0/\gamma)$ dominate integral (7.9). These values are far smaller than the characteristic values $\tau' \approx \gamma^{-1}$. We can thus ignore τ in comparison with τ' in the argument of the exponential function, and we have

$$\Delta Q = -\frac{B}{\gamma}\frac{L_1/\tau_d}{\nu^2 + (L_1/\tau_d)^2}. \tag{7.13}$$

where

$$L_1 = \frac{\pi}{2}\ln\frac{\Gamma_0}{\gamma}. \tag{7.14}$$

Let us compare the width of the burnt-out hole with the size of the corresponding critical amplitude F_c. We see that in all cases in which spectral diffusion is important the width of the hole is far greater than F_c. At high temperatures, with $\gamma \ll 1/\tau_d$, we have

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

[see (5.6)], while the width of the burnt-out hole is

$$\Delta v \approx 1/\tau_d \gg F_c$$

[see (7.6)]. At low temperatures, with

$$\Gamma_0 \ll \gamma \ll (\Gamma_0/\tau_d)^{1/2},$$

we have, in order of magnitude,

$$F_c \approx (\Gamma_0/\tau_d)^{1/2},$$

[see (5.10)], and we also have $\Delta v \approx \Gamma_0/\gamma \tau_d \gg F_c$ [see (7.11)]. In the case $\gamma \ll \Gamma_0 \ll (\tau_0/\tau_d)^{1/2}$ we have $F_c \approx (\gamma/\tau_d)^{1/2}$ [see (5.14)] and $\Delta v \approx 1/\tau_d \gg F_c$. In all of the cases listed above, the width of the burnt-out hole is related to the critical amplitude F_c by

$$F_c \approx (\gamma \Delta v)^{1/2}. \qquad (7.15)$$

The physical picture of the nonlinear resonant absorption and of the burnt-out hole in a situation with spectral diffusion is thus much more diverse and complicated than would follow from the theory based on the Bloch equations and the assumption that the relaxation time Θ_2 of the nondiagonal component of the density matrix is on the order of τ_d.

8. REGION OF A STRONG NONLINEARITY ($F \gg F_c$)

We can now draw a qualitative picture of the behavior of the absorption at high intensities ($F \gg F_c$). The time average of the power absorbed per unit volume by resonant pairs is

$$Q = -\frac{\omega}{V} \sum_{res} \hbar F \langle \text{Ref} \rangle_t. \qquad (8.1)$$

Using (2.2), we can put this expression in the form

$$Q = \hbar \omega \gamma P_\omega \int_0^\infty dE \langle (n - n_0) \rangle_t. \qquad (8.2)$$

This expression is actually a consequence of energy conservation. The nonlinear behavior of Q is therefore determined by the characteristic width of the region of the integration over E and by the value of the mean $\langle n - n_0 \rangle_t$ in this region.

We will consider only those limiting cases in which spectral diffusion is important. In other cases, the nonlinear behavior is described by solutions of the Bloch equations with longitudinal and transverse relaxation times Θ_1 and Θ_2, respectively, with $\Theta_2 = 2\Theta_1 = 2/\gamma$.

At high temperatures ($\Gamma_0 \tau_d \gg 1$) the case $\gamma \ll 1/\tau_d$ is interesting. As we have seen, the characteristic size of the region of the integration over E (the region of the burnt-out hole) is of order $\hbar/\tau_d$, and the mean value $\langle n - n_0 \rangle$ for $F \gg F_c$ is equal to $1/2 - n_0$. We then have

$$Q \approx \hbar^2 \omega \gamma P_\omega \tau_d^{-1} \, \text{th}(\hbar\omega/2T) \approx \hbar^2 P_\omega \omega F_c^2 \, \text{th}(\hbar\omega/2T), \qquad (8.3)$$

where we have $F_c \approx (\gamma/\tau_d)^{1/2}$, according to (5.6). The absorption coefficient α is found by dividing (8.3) by the energy flux density, which is proportional to F^2. Correspondingly, for $F \gg F_c$, we have

$$\alpha \approx \alpha_0 (F_c/F)^2, \qquad (8.4)$$

where α_0 is the absorption coefficient of the linear theory, given by

$$\alpha_0 = 4\pi\sigma_0(\omega)/c\varepsilon^{1/2}, \qquad (8.5)$$

and c is the velocity of light. At high intensities the absorption coefficient is therefore inversely proportional to the intensity.

As we will see, the same comment holds in other limiting cases in which spectral diffusion plays a role, which we will discuss below. On the other hand, in those cases in which spectral diffusion does not play a role we have $\alpha \approx \alpha_0(F_c/F)$; i.e., the absorption coefficient is inversely proportional to the square root of the intensity.

At low temperatures in the case $\Gamma_0 \ll \gamma \ll (\Gamma_0/\tau_d)^{1/2}$, the width of the region of the integration over E is on the order of the characteristic width of the region of spectral diffusion, $\hbar/\tau_d$. The value of $\langle n - n_0 \rangle_t$ can be estimated on the basis of the following considerations. We have seen that a resonant pair undergoes a change in population in a resonance region of width $\hbar(\Gamma_0/\tau_d)^{1/2}$, which is crossed in a time $(\tau_d/\Gamma_0)^{1/2}$. It exists in an excited state for a time $\approx \gamma^{-1} \gg (\tau_d/\Gamma_0)^{1/2}$; it then reverts to the ground state, emitting a phonon. For all the rest of the time (while it is traversing the region of spectral diffusion, of width $\approx \hbar/\tau_d$) we have $n = n_0$ for this pair. It returns to the resonance region after a time $\approx \Gamma_0^{-1} \gg \gamma^{-1}$. The fraction of the time which a resonant pair spends in the excited state (i.e., with $n = 1/2$) is therefore $\Gamma_0/\gamma \ll 1$ in order of magnitude. Hence

$$\langle n - n_0 \rangle_t \approx (1/2 - n_0) \Gamma_0/\gamma.$$

Using this result and the expression (5.10) for F_c in the case of interest here, we find the estimate (8.3).

In the case $\gamma \ll \Gamma_0 \ll (\Gamma_0/\tau_d)^{1/2}$ a resonant pair returns to the resonance region many times during the lifetime γ^{-1}. Each time it returns, its population increases by a small amount $F^2 \tau_\varphi^2 = F^2 \tau_d/\Gamma_0$. Consequently, the number of returns must be high (on the order of $1/F^2 \tau_\varphi^2$) if the level populations are to become equal. This situation is reached in a time of order $1/\Gamma_0 F^2 \tau_\varphi^2$. On the other hand, we have

$$1/(\Gamma_0 F^2 \tau_\varphi^2) = 1/\tau_d F^2 \approx \gamma^{-1}(F_c/F)^2,$$

where $F_c \approx (\gamma/\tau_d)^{1/2}$ [see (5.14)]. For $F \gg F_c$, the pumping time is thus significantly shorter than the lifetime of the resonant pair in its excited state, γ^{-1}. In other words, the resonant pair is excited over the greater part of its lifetime. As a result, we have $\langle n - n_0 \rangle_t \approx 1/2 - n_0$. The characteristic interval of the integration over E is again on the order of $\hbar/\tau_d$. As a result, we again find the estimate (8.3).

9. DISCUSSION OF RESULTS

To see which of these cases is realized at a given frequency ω and a given temperature T in a specific semiconductor, we should compare the values of Γ_0, γ, and τ_d, which figure in the theory. The first two of these quantities are expressed in terms of the relaxation time of the corresponding pairs, $\tau(E,r)$ (E is the distance between the energy levels, and r is the arm of the pair), as follows:

$$\gamma = \frac{1}{\tau(\hbar\omega, r_\omega)}, \quad \Gamma_0 = \begin{cases} 1/\tau(T, r_T), & T < E_a, \\ 1/\tau(E_a, r_a), & T > E_a, \end{cases} \qquad (9.1)$$

where

$$r_T = a \ln(2I_0/T), \quad r_a = a \ln(2I_0/E_a).$$

TABLE I.

Interaction mechanism	n	β_n	$\Phi_n(x)$ $x \ll 1$	$x \gg 1$
DA $\begin{cases} D_1 \neq D_2 \end{cases}$	1	$\dfrac{D_1{}^2 + D_2{}^2}{8\pi\rho\hbar^4 w^5}$	$\dfrac{(D_1 - D_2)^2}{2(D_1{}^2 + D_2{}^2)}$	1
$\quad\ \ D_1 = D_2 = D$	1	$\dfrac{D^2}{4\pi\rho\hbar^4 w^5}$	$\dfrac{x^2}{6}$	1
PA	-1	$\dfrac{3}{35\pi}\left(\dfrac{4\pi l\beta}{\varepsilon}\right)^2 \dfrac{1}{\rho\hbar^2}\left(\dfrac{1}{w_l{}^3} + \dfrac{4}{3w_t{}^3}\right)$	$\dfrac{x^2}{6}$	1

The relaxation time $\tau(E,r)$ is given by[13]

$$\frac{1}{\tau(E,r)} = \beta_n E^n \Phi_n\left(\frac{E}{E_r}\right)\frac{4I^2(r)\operatorname{cth}(E/2T)}{[1 + (E/E_a)^2]^4}. \qquad (9.2)$$

Here $E_a = 2\hbar w/a$ and $E_r = \hbar w/r$. The exponent n, the coefficient β_n, and the function Φ_n depend on the particular mechanism by which the pairs interact with phonons; these quantities are listed in Table I for a deformation interaction (for two cases: that in which the deformation potentials of the components of the pair are different, and that in which they are the same) and for a piezoelectric interaction with acoustic phonons. Table I uses the following notation: $D_{1,2}$ are the deformation potentials of the components of the pair, ρ is the density of the semiconductor, β is the piezoelectric modulus (calculations were carried out for the case of cubic symmetry T or T_d), ε is the dielectric constant, w_l (w_t) is the longitudinal (transverse) sound velocity, and DA and PA represent the deformation acoustic and piezoacoustic interactions, respectively.

We see that there are two characteristic values of E which determine the relaxation of pairs through interaction with phonons. The first of these values, E_a, corresponds to the condition that the wavelength of a phonon with an energy E_a be equal to the localization radius of the state, a. For $E > E_a$, the deformation field which results from phonons with an energy E oscillates rapidly over a length scale a, and the probability for a transition of a pair from one state to the other falls off sharply with increasing E. The second value, $E_r \ll E_a$, corresponds to the condition that the wavelength of a phonon with an energy E_r is of order the arm of the pair, r. For $E < E_r$, the deformation field varies smoothly over a distance corresponding to the size of a pair; this circumstance has a major influence on the results if the deformation potentials of the components of the pair are approximately equal. In this case an interaction arises only to the extent to which the values of the phonon-induced deformation at the different sites making up the pair are different. The situation is precisely the same in the case of the piezoelectric interaction.

The reason for the difference between the expressions for Γ_0 in (9.1) in the cases $T < E_a$ and $T > E_a$ is that pairs with energies greater than E_a interact extremely weakly with phonons; transitions occur only rarely; and these pairs accordingly play almost no role in the spectral diffusion. Correspondingly, we have

$$\frac{\hbar}{\tau_d} \approx \begin{cases} \eta_T T & T < E_a \\ \eta_a E_a & T > E_a \end{cases}, \qquad (9.3)$$

where η_T is given by (1.7), and η_a differs from η_T in that r_T is replaced by r_a.

We have thus presented all the information required in order to understand which of the limiting cases we have discussed will actually be realized in some particular semiconductor or other.

All the discussion above has been based on the idea that the pairs of levels are well-determined, i.e., that the following relation holds for the characteristic energies:

$$\hbar/E\tau(E, r_E) \ll 1. \qquad (9.4)$$

For the case of a deformation interaction with $D_1 \neq D_2$, this condition is violated if we have $E_a > E_c = \beta_1^{-1/2}$, at temperatures between E_c and E_a. A corresponding region also exists in certain piezoelectric semiconductors, as can be verified. In regions with $\Gamma_0 > T$ or $\gamma > \hbar\omega$, the theory constructed here does not apply. The boundaries of the corresponding regions can be found on the basis of the expressions given above for the relaxation time of pairs and the particular parameter values of the materials.

It can be seen from the expressions given here that the critical intensity and the width of the burnt-out hole depend strongly on the frequency ω, the temperature T, and the parameter values of the semiconductor. These values can span an extremely wide interval. For example, with $T \approx 1$ K and a frequency $\omega/2\pi \approx 1$ the critical amplitude $\mathscr{E}_{0c}$ can range from 10^{-7} to 10^{-2} V/cm, depending on the particular material.

An experimental study of nonlinear resonant defects at low temperatures yields the important characteristics of semiconductors in the regime of hopping conductivity.

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[1] Necessary conditions for the applicability of the two-site model and expression (1.3) are the inequalities $a \ll r_\omega \ll \bar{r}$.

[2] The nonlinear hopping conductivity in a semiconductor was first studied by Böttger and Bryksin.[8] The primary distinctions between our own results and those of Ref. 8 stem from the circumstance that we are incorporating spectral diffusion and the Coulomb correlation in the pair occupation numbers. Nonlinear effects in rf hopping conductivity have also been studied by Zvyagin.[9]

[3] The reason for the linear time dependence in this expression (instead of the square-root diffusion dependence) is the strong correlation between the magnitude of a jump and its probability: The probabilities for small jumps are far greater than those for large jumps. From this standpoint, the term "spectral diffusion" itself is something less than perfect, but we retain it here for the sake of continuity.

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