High-frequency hopping conductivity of semiconductors: theory of nonlinear and quantum effects

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A theory is derived for the nonlinear high-frequency hopping conductivity of a semiconductor far from the percolation threshold, i.e., from the Anderson transition. As was shown by Pollak and Geballe, the conductivity is determined by the transitions of an electron between a pair of centers separated by less than the average distance. The real part of the conductivity is studied as a function of the temperature and of the frequency and amplitude of the high-frequency field. The nonlinear behavior of both the resonant (phononless) and nonresonant (relaxation) components of the conductivity is analyzed. The role played by spectral diffusion is discussed in a description of the nonlinear behavior of the resonant component. The nonlinear decay of the resonant conductivity is shown to begin in comparatively weak high-frequency fields, so that special measures would have to be taken to observe the linear region. The nonlinear behavior of the relaxation component of the conductivity becomes substantial in far stronger fields, and this is a typical low-temperature effect that becomes easier to observe as the temperature is lowered. An expression is derived for the nonresonant linear conductivity in the quantum frequency range. It is found that in this frequency range the conductivity depends only slightly on the temperature. Its frequency dependence can take different forms, depending on the nature of the interaction with the phonons responsible for the transitions between donors. The close analogy between electromagnetic absorption in semiconductors and glasses is discussed. In glasses, the absorption results from an interaction of the high-frequency field with two-level tunnel systems.

PACS numbers: 72.20.Ht, 72.30. + q

I. INTRODUCTION

After two-level tunnel systems were found responsible for the low-temperature properties of glasses, in research influenced by the insight of Anderson et al.[1] and Phillips,[2] attention was attracted to the analogy between the properties of glasses and the properties of systems in another field of study: semiconductors in the region of the hopping conductivity. This analogy concerns primarily such phenomena as the absorption of electromagnetic radiation.[1] As was first shown by Pollak and Geballe,[3] above certain frequencies ω the high-frequency conductivity of doped semiconductors (and thus their absorption) is determined by hops of electrons between pairs of adjacent donors separated by less than the average distance (the two-site model). The analogy between these pairs and the two-level systems in glasses has proved quite close. The two-site model has since been used to describe the high-frequency hopping conductivity of amorphous semiconductors (amorphous germanium and silicon, for example).[4-6]

Comparison of the situations in glasses and semiconductors has stimulated a transfer of several ideas from the physics of glasses to the physics of semiconductors. On the other hand, several ideas which are only hypothetical for glasses (dealing with, for example, the details of the interaction of phonons with the two-level systems) can be formulated quite clearly and studied in the theory of semiconductors. We see thus that a systematic comparison can enrich the physics of both fields.

Our purpose in the present paper is to make this comparison for the particular example of electromagnetic absorption. By taking this approach we can go beyond the comparison to derive several new results for semiconductors, concerning both the nonlinear absorption and the linear absorption in the quantum region.

For definiteness we will discuss a model of a disordered system similar to that used by Shklovskiĭ and Éfros.[7] This model is closest in meaning to the case of an amorphous semiconductor. Nevertheless, since the results found for the two cases do not differ greatly (see the Conclusion), we will briefly discuss in appropriate places the situation with regard to doped semiconductors. We assume thus that there are N sites per unit volume, distributed at random. The state at each site may be either occupied by an electron (in which case the site is negatively charged) or vacant (and the site neutral). Overall electrical neutrality results from positively charged centers which are distributed randomly over space. The one-electron energy levels φ_i at each site are also distributed randomly, and they have a continuous energy distribution over a broad energy range. The characteristic width A of the energy distribution is assumed to be not only far higher than the average energy of the Coulomb interaction between adjacent sites but also higher than the energy of the interaction between the centers forming the pairs responsible for the absorption. This scatter of energies may result from random disruptions of the short-range order.

II. RESONANT (PHONONLESS) ABSORPTION

1. General relations

The high-frequency conductivity is the sum of two components: a phononless (or resonant) component and a relaxa-

tion (or nonresonant) component. The physics underlying the first component can be outlined as follows: The alternating field "selects" pairs which have an energy (a level separation) $E = \hbar\omega$, and these are the pairs which are responsible for the absorption.[4-6] Only those pairs which contain one electron can of course contribute to the absorption. As Shklovskiǐ and Éfros showed,[7] a computation of the number of such pairs must incorporate the Coulomb repulsion of electrons: It must be kept in mind that when there are two electrons in a pair there is an additional energy $e^2/\varepsilon r$, where r is the "arm" of the pair, and ε is the dielectric constant. We are interested in the case in which the characteristic Coulomb energy is far higher than both $\hbar\omega$ and T:

$$e^2/\varepsilon r \gg \hbar\omega, \; T, \tag{1}$$

where ω is the frequency of the alternating field, and T is the temperature in energy units. For the real part of the high-frequency conductivity, $\sigma(\omega)$, the linear approximation yields the following expression, which is similar to that derived by Shklovskiǐ and Éfros[7]:

$$\sigma_0(\omega) = \frac{\pi^2}{3} \operatorname{th}\left(\frac{\hbar\omega}{2T}\right) a g^2 \frac{e^4}{\varepsilon} \omega r_\omega{}^3. \tag{2}$$

Here g is the density of one-electron states, a is the localization radius of the state, and r_ω is the smallest pair arm for which the distance between levels is $\hbar\omega$.

Since r_ω is a weak (logarithmic) function of ω, expression (2) has essentially the same frequency and temperature dependence as that of the coefficient of the resonant electromagnetic absorption (or sound absorption) in glasses. On the other hand, the resonant absorption in glasses characteristically reaches saturation very rapidly as the intensity is raised.[8] To determine the nature of this nonlinearity we consider expression (2). The factor $\tanh(\hbar\omega/2T)$ in this expression is none other than the difference between the equilibrium populations of the lower and upper levels of the resonant pair. A strong alternating field equalizes the populations, i.e., causes a nonlinear effect: a decrease in σ with increasing field amplitude.[2] For the nonlinear resonant absorption we find by a method similar to that used in Ref. 8 the following expression:

$$\sigma(\omega) = \alpha\sigma_0(\omega)\,(\mathscr{E}_{c1}/\mathscr{E}_0), \quad \mathscr{E}_0 \gg \mathscr{E}_{c1}. \tag{3}$$

Here $\alpha \approx 1$ [$\alpha = 3\pi/8$ when the interaction with phonons is responsible for the relaxation; see Eq. (25) and the text which follows it][3]; $\mathscr{E}_0$ is the electric field amplitude; and $\mathscr{E}_{c1}$ is its critical value, given by

$$\mathscr{E}_{c1} = \hbar/er_\omega\,(\tau_1\tau_2)^{1/2}, \tag{4}$$

where τ_1 is the relaxation time of the populations of pairs with an arm r_ω and a level separation $\hbar\omega$, and τ_2 is the phase relaxation time of the wave function of an electron in the upper level. This time and also τ_1 may be determined by the interaction of the resonant pair with thermal phonons. In this case, we would have $\tau_2 = 2\tau_1$.

2. Role of spectral diffusion. Estimate of the critical field $\mathscr{E}_{c1}$

It may turn out, however, that the time τ_2 is much less than τ_1 and is determined by the interaction of the resonant

pair with the thermally excited pairs around it. This interaction causes the pair of interest to depart from the resonance; specifically, the departure is caused by the time-varying random fields which are produced in transitions in adjacent pairs with an energy-level separation on the order of T. This effect is called "spectral diffusion."[8] The random fields may be of either electrical or elastic origin; we will discuss the first case first.

Let us examine the change ΔE which is caused in the energy of a resonant pair by transitions in the nearest thermal pair. The characteristic distance between the components of such a pair is r_T; this is the minimum arm[4] of a pair with a level separation T.

The energy change ΔE is the energy of the interaction of two dipoles, with moments er_ω and er_T, separated by a distance R: $e^2 r_\omega r_T/\varepsilon R^3$. The total energy of the interaction with all significant pairs is determined in order of magnitude by the nearest thermal pair. The characteristic distance to this pair, R, is on the order of the average distance between thermal pairs. The concentration of such pairs is[7]

$$n_T \approx g^2\,(e^2/\varepsilon r_T)\,T r_T{}^2 a. \tag{5}$$

We thus find

$$\Delta E \approx \gamma T, \tag{6}$$

where

$$\gamma \approx g^2\,(e^4/\varepsilon^2)\,r_\omega r_T{}^2 a. \tag{7}$$

For a weakly doped semiconductor of an intermediate degree of compensation, expression (7) would be replaced by (see the Conclusion)

$$\gamma \approx N^{4/3} r_\omega r_T{}^2 a, \tag{8}$$

where N is the impurity concentration.

The energy ΔE is a measure of the rate at which the pair is driven from resonance by the fluctuational transitions in the thermal pairs. At present there is no quantitative theory which relates ΔE and τ_2. The assumption[11] $\tau_2 \approx \hbar/\Delta E$ is used (in reasonable agreement with several experiments).

We turn now to the strain contribution to spectral diffusion. We assume that the random energy of an electron at a site is shifted by strain by an amount proportional to the strain. The corresponding proportionality factor, Λ (the strain potential), is usually on the order of a few electron volts. We should apparently discuss two physically distinct cases.

A) The strain potentials of the components of the pair are so different that the difference is on the order of the strain potentials themselves, a few electron volts. This situation should arise in semiconductor glasses, although there may be cases in which it occurs in crystalline semiconductors. An order-of-magnitude estimate of the interaction is reminiscent of that of the Coulomb case[8]:

$$\Delta E \approx \Lambda^2/\rho s^2 R^3, \tag{9}$$

where ρ is the mass density of the semiconductor, s is the sound velocity, and the distance R should be determined from (5): $R^{-3} \approx n_T$. Estimates show that the strain contribution to ΔE can be comparable to the Coulomb contribution.

To estimate τ_2 we use (8). Adopting $N = 10^{16}$ cm^{-3}, $a = 10^{-7}$ cm, and $T = 1$ K, we find $\tau_2 \approx \hbar/\Delta E \approx 10^{-7} - 10^{-8}$ s. We must emphasize that this is a crude estimate. It can be refined by resorting to experimental data on the frequency and temperature dependences of the nonlinear high-frequency conductivity or by pursuing the theory of spectral diffusion.

B) The strain potentials of the two components of the pair differ only slightly; correspondingly, the strain interaction is weaker than the Coulomb interaction. This situation can occur in doped crystalline semiconductors.

3. Population relaxation mechanisms; the time τ_1

It remains to derive an expression for τ_1 and to estimate this time. It depends on the energy separation E of the levels of the resonant pair:

$$E = [(\varphi_1 - \varphi_2)^2 + 4I^2(r)]^{1/2}, \tag{10}$$

where φ_i are the one-site energies, which are determined by the neighboring centers (not in the pair under consideration), and $I(r)$ is the energy overlap integral of the wave functions of the components of the pair. This integral is conveniently written

$$I(r) = I_0 e^{-r/a}, \tag{11}$$

where a is the localization radius of the state, and the quantity I_0 has the dimension of energy. Its order of magnitude depends on the nature of the medium. Furthermore, $I(r)$ may fall off over r in a manner different from that described by (11). In this case the frequency and temperature dependences remain basically the same as before, as does the intensity dependence, but the power of the logarithm changes [we are assuming that the basic $I(r)$ dependence nevertheless remains exponential at large distances]. For an $\exp(-r^2/a^2)$ decay law, for example, the powers of the logarithms turn out to be half as large. The logarithmic behavior which follows from both the nonlinear and linear theories thus depends strongly on the particular model used for the disordered system.

The time τ_1 is determined by transitions between the levels of the pair, accompanied by the absorption or emission of a phonon with an energy E. The corresponding expression can be written easily for case A (cf. Ref. 9):

$$\frac{1}{\tau_1} = \frac{1}{\tau_0} \frac{I^2(r)E}{T^3} \operatorname{cth} \frac{E}{2T}, \tag{12}$$

where τ_0 has the physical meaning of a relaxation time for a pair with $I(r) = E = T$. This time is inversely proportional to T^3, and it can be approximated by

$$\hbar/\tau_0 \approx T^3/E_c^2, \tag{13}$$

where E_c is the characteristic energy at which the uncertainty $\hbar/\tau_1$ in the energy of the upper level of the pair [at $T = 0$ and $I(r) = E$] reaches a value on the order of the level separation E. This energy is given in order of magnitude by[12]

$$E_c \sim (\rho \hbar^3 s^5)^{1/2}/\Lambda. \tag{14}$$

Assuming $\Lambda \approx 1-2$ eV, we find E_c to be 10–20 K. The energy E_c is thus that characteristic value of the energy E above

which the relaxation of the level pair under consideration can no longer be described by the one-phonon approximation, and the coupling with phonons becomes strong. In our opinion, this situation is one of the most interesting problems awaiting solution in the theory of disordered semiconductors.

Expression (12) was derived under the assumption that the wave vector $q_E = E/\hbar s$ of the phonon which is emitted or absorbed is far smaller than a^{-1}. With increasing E, this assumption breaks down, and (12) must be replaced by an expression containing the additional factor $[1 + (E/T_a)^2]^{-4}$:

$$\frac{1}{\tau_1} = \frac{1}{\tau_0} \frac{I^2(r)E}{T^3} \operatorname{cth}\left(\frac{E}{2T}\right) \frac{1}{[1 + (E/T_a)^2]^4}, \tag{15}$$

where $T_a = 2\hbar s/a$. The condition written above for the applicability of the one-phonon approximation is valid for $E_c < T_a$. In the opposite case, this approximation can be used over the entire energy interval. The value of T_a depends on the particular semiconductor and can vary over a broad range, 10–100 K.

To reach an understanding of case B we consider transitions caused between levels by a phonon with an energy E and thus with a wave vector q_E. If the parameter $q_E r$ is small, the displacements of the levels φ_1 and φ_2 occur essentially in phase, and the distance between these levels remains constant. Transitions become possible only to the extent that the corresponding phase difference becomes different from zero. Calculations show that this circumstance is taken into account (in the isotropic case) by the additional factor

$$1 - J_0(E/T_r); \quad T_r = T_a a/2r = \hbar s/r, \tag{16}$$

where $J_0(x)$ is a Bessel function of order 0. The final result is (cf. Refs. 6 and 13)

$$\frac{1}{\tau_1} = \frac{1}{\tau_0} \frac{I^2(r)E}{T^3} \operatorname{cth}\left(\frac{E}{2T}\right) \frac{1 - J_0(E/T_r)}{[1 + (E/T_a)^2]^4}. \tag{17}$$

We recall that expression (4) contains the times τ_1 and τ_2 at the argument values $E = \hbar\omega$ and $r = r_\omega$ (in the absence of spectral diffusion).

There is yet another important case in which the piezoelectric interaction is predominant in the energy range of interest. This case can arise in essentially all crystalline semiconductors which lack a center of inversion. The piezoelectric interaction leads to the following expression for the time τ_1:

$$\frac{1}{\tau_1} = \frac{1}{\tau_0^{(p)}} \left(\frac{T}{E}\right) \frac{I^2(r)}{T^2} F\left(\frac{E}{T_r}\right) \operatorname{cth}\left(\frac{E}{2T}\right) \frac{1}{[1 + (E/T_a)^2]^4}. \tag{18}$$

Here $F(x) = 1$ at $x \gg 1$, and $F(x) \propto x^2$ at $x \ll 1$; $\tau_0^{(p)}$ is a relaxation time determined by the piezoelectric interaction. In order of magnitude, this time is given by

$$1/\tau_0^{(p)} = \chi 4\pi e^2 T/\varepsilon \hbar^2 s, \tag{19}$$

where χ is the square of the electromechanical coupling constant averaged over the directions.

We now estimate the critical amplitude $\mathscr{E}_{c1}$. To the best of our knowledge, no experimental results are available for a direct determination of this amplitude. For an estimate we

will thus work from the data of Ref. 14, where acoustic effects in doped p-type germanium were studied.[5] This experiment yielded the value $\tau_2 = 5 \cdot 10^{-8}$ s, while τ_1 varied in inverse proportion to T, having the value $4 \cdot 10^{-7}$ s at $T = 2$ K and $\omega = 2\pi \cdot 1.022$ GHz [the order of magnitude of τ_1 and its temperature dependence agree with the predictions of expression (12) for case A, if the difference between strain potentials is assumed to be 1–2 eV].[6] We then find

$$\mathscr{E}_{c1} \sim 10^{-2} - 10^{-3} \text{ V/cm};$$

i.e., the nonlinearity of the resonant absorption sets in at a vanishingly low intensity (on the order of 10^{-7} W/cm^2). It would therefore be difficult to observe the linear, phononless conductivity.

The situation which we have found here is thus the opposite of that to which we are accustomed in dielectric glasses.[15] In the latter the critical electromagnetic intensity for resonant absorption is far higher than the critical acoustic intensity (by four or five orders of magnitude). The opposite situation prevails in semiconductors, where the corresponding acoustic intensity is about 10^{-4} W/cm^2 (Ref. 16). The physical reason for this marked difference in critical electromagnetic absorption intensities in glasses and semiconductors is the large arm of the resonant pair in a semiconductor.

4. Nonlinear high-frequency conductivity when the time τ_2 is determined by an interaction with phonons

For a quantitative calculation of the nonlinear resonant absorption we consider the case $\tau_2 = 2\tau_1$. We must take into account the circumstance that the absorption is due to pairs whose average energies $(\varphi_1 + \varphi_2)/2$ fall in a band of width $e^2/\varepsilon r$ below the Fermi level.[7] Under conditions (1), the contributions of most such pairs are identical, equal to

$$P = \frac{2\pi}{\hbar} \hbar\omega \frac{(e\vec{\mathscr{E}}_0 \mathbf{r})^2}{4} \frac{I^2(r)}{E^2} D(\hbar\omega - E) \text{ th} \frac{E}{2T}, \qquad (20)$$

where

$$D(\hbar\omega - E) = \frac{1}{\pi} \frac{\hbar}{\tau_2} \Big\{ (\hbar\omega - E)^2 + \Big(\frac{\hbar}{\tau_2}\Big)^2$$
$$\times \Big[1 + 2e^2(\vec{\mathscr{E}}_0\mathbf{r})^2 I^2(r) \frac{\tau_1\tau_2}{4\hbar^2 E^2} \Big] \Big\}^{-1}. \qquad (21)$$

This expression is derived by analogy with Ref. 8; the following condition is assumed in its derivation:

$$\omega\tau_\omega \gg 1 + \hbar^{-2}(e\mathscr{E}_0 r_\omega \tau_\omega)^2, \qquad (22)$$

where τ_ω is the minimum value of τ_1 at $E = \hbar\omega$. By virtue of this condition, the function $D(\hbar\omega - E)$ can be replaced by $G\delta(\hbar\omega - E)$, where the coefficient G is determined from the normalization condition. Taking this circumstance into account, and summing over all the pairs contributing to the absorption, we find

$$\sigma(\omega) = \frac{2\pi^2 g^2 e^4}{\varepsilon} \omega \text{ th}\Big(\frac{\hbar\omega}{2T}\Big)$$

$$\times \int_0^\pi d\theta \sin\theta \int_{r_\omega}^\infty dr\, r^3 \cos^2\theta \exp\Big[-\frac{2(r_\omega - r)}{a} \Big]$$

$$\times \Big[1 + \Big(\frac{\mathscr{E}_0}{\mathscr{E}_{c1}}\Big)^2 \cos^2\theta \Big(\frac{r}{r_\omega}\Big)^2 \exp\frac{2(r - r_\omega)}{a} \Big]^{-1/2}$$
$$\qquad (23)$$

$$\times \int_0^{\hbar\omega} d\Delta\delta \Big\{ \hbar\omega - \Big[\Delta^2 + (\hbar\omega)^2 \exp\frac{2(r_\omega - r)}{a} \Big]^{1/2} \Big\},$$

where θ is the angle between $\vec{\mathscr{E}}_0$ and $\mathbf{r}$, $\Delta = \varphi_2 - \varphi_1$, and $\mathscr{E}_{c1}$ is given by (4). Integrating over r, θ, and Δ, we find

$$\sigma(\omega) = \sigma_0(\omega) F(\mathscr{E}_0/\mathscr{E}_{c1}), \qquad (24)$$

where $\sigma_0(\omega)$ is given by (2), and

$$F(x) = \frac{3x^2}{4} \int_0^{x^{-2}} \frac{z^{1/2} dz}{(1 - x^2 z)^{1/2}} \Big[(1+z)^{1/2} - z \ln\frac{1 + (1+z)^{1/2}}{z^{1/2}} \Big]. \qquad (25)$$

In the limit $x \to 1$ we have $F(x) \to 1$, and in the limit $x \to \infty$ we have $F(x) = 3\pi/8x$.

III. RELAXATION (NONRESONANT) ABSORPTION ($\hbar\omega < T$)

1. The qualitative picture and the initial equations

The relaxation absorption results from a modulation caused by the alternating electric field in the separation E between the levels of the pair. In the external alternating electric field $\vec{\mathscr{E}}$ the difference between the one-site energies, $\varphi_1 - \varphi_2$, acquires an increment $e\vec{\mathscr{E}}\mathbf{r}$, where $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$. This energy modulation changes the population (f) of the upper level from its equilibrium value; this population change lags in phase behind the change in the energy. As a result, energy of the alternating field is dissipated. The power P absorbed by one pair of levels is

$$P = \langle f\dot{E}\rangle = \frac{\omega}{2\pi} \int_0^{2\pi/\omega} dt f(t) \frac{dE}{dt}. \qquad (26)$$

This expression can be derived, for example, by noting that all the energy from the electric field is ultimately transferred to the phonon system of the semiconductor. The separation $E(t)$ is given by

$$E(t) = [(\varphi_1 - \varphi_2 + e\vec{\mathscr{E}}(t)\mathbf{r})^2 + 4I^2(r)]^{1/2}. \qquad (27)$$

Expression (26) was derived in the adiabatic approximation. In general, the conditions for its applicability are

$$\hbar |dE/dt| \ll E^2, \qquad (28)$$
$$\hbar\omega \ll E. \qquad (29)$$

These conditions mean that the changes in the perturbation must be slow enough not to cause quantum transitions.

The occupation numbers f are determined from the equation

$$\frac{\partial f}{\partial t} = -\frac{f - f_0}{\tau_1}; \quad f_0 = (1 + e^{E/T})^{-1}. \qquad (30)$$

The linear approximation is valid for

$$\mathscr{E}_0 \ll \mathscr{E}_{c2} \equiv T/er_c. \qquad (31)$$

Here r_c is the characteristic arm of the pairs dominating the absorption (the values of this arm are different in the different limiting cases, and we will determine them below). In this approximation, the power P absorbed by one pair is given by an expression of the Debye type, according to (26) and (30)

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(cf. Ref. 8, where an analogous quantity was calculated for glasses):

$$P = \frac{1}{2}|e\vec{\mathscr{E}}_0\mathbf{r}|^2 \frac{\omega^2\tau_1}{1+(\omega\tau_1)^2}\left(-\frac{\partial f_0}{\partial E}\right),\qquad(32)$$

where $\tau_1(r,E)$ is determined by one of the expressions (12), (15), (17), (18).

Expression (32) should be summed over all pairs. It is clear that the predominant pairs are those with $E \approx T$ and that r_c, the characteristic value of r, depends on the ratio of the field period $2\pi/\omega$ and the time

$$\tau_{min}(T) = \tau_1(r_T, T),$$

which has the physical meaning of being the minimum relaxation time of the occupation numbers of pairs with a level separation T. If $\omega\tau_{min}(T) \gg 1$, the predominant pairs are those with τ_1 on the order of τ_{min}, and r_c is correspondingly on the order of r_T. Since the only pairs which absorb are those which have a single electron, we find the known result[7]

$$\sigma_0(\omega) = \frac{\pi^5}{288}\frac{e^4}{\varepsilon}ag^2r_T^3\tau_{min}^{-4}(T).\qquad(33)$$

A study of the temperature dependence $\sigma(\omega)$ under these conditions would reveal which case (A or B) holds in the given material. This is also an important consideration for interpreting experiments[14,16] on the acoustic properties of semiconductors.

Under the condition $\omega\tau_{min}(T) \ll 1$, the predominant pairs are those with $r = r_c$, where

$$r_c = \frac{a}{2}\ln\frac{1}{\omega\tau_{min}},\qquad(34)$$

for which $\omega\tau_1(r_c,T) = 1$ (i.e, $r_c > r_T$). As a result we find[7]

$$\sigma_0(\omega) \approx (e^4/\varepsilon)ag^2\omega r_c^3.\qquad(35)$$

This expression is completely analogous to the Jäckle expression[17] for the coefficient of the absorption of sound and electromagnetic radiation caused by two-level systems in glasses.

In the nonlinear region, to which we now turn, a qualitative analysis reveals the dependence of the absorption on the amplitude, frequency, and temperature in limiting cases. The quantitative theory is required only for determining the numerical coefficients in the corresponding expressions and for analyzing intermediate cases.

2. Nonlinear relaxation absorption; analysis for case A [expression (12)]

If the electric field amplitude $\mathscr{E}_0$ is so high that it causes the energy levels in the pair to separate by an amount exceeding T, i.e., if

$$d \equiv e\mathscr{E}_0 r_c \gg T,\qquad(36)$$

then the relaxation absorption depends on the amplitude $\mathscr{E}_0$; specifically, it decreases as this amplitude increases. In the nonlinear regime the quantity r_c in this inequality can, in general, depend on the wave amplitude; we will derive this quantity below.

To describe the qualitative picture we begin with case A. We will discuss the general situation below. Further-
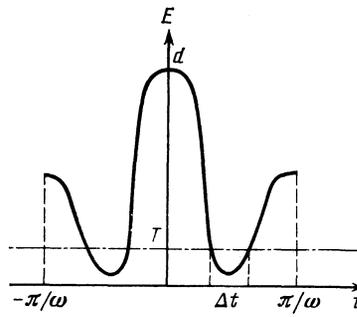

FIG. 1.

more, in analyzing the nonlinear relaxation absorption we will restrict the discussion to the case $d \ll T_a$ (the opposite case would seem to be extremely difficult to realize in experiment).

Figure 1 helps explain the physics involved. This is a schematic diagram of the time (t) dependence of the level separation E. We see that, under condition (36), the level separation E is of the order of T only during the short time intervals

$$\Delta t \approx T/d\omega \ll \omega^{-1}.\qquad(37)$$

It is during these time intervals that thermal phonons can excite the pair (if it is initially in the lower energy state). The characteristic relaxation time with respect to these processes is $\tau_{min}(T)$. At other times, the pairs can only relax by emitting phonons with energies $E(t)$. The characteristic relaxation time of the pairs with respect to such processes is $\tau_{min}(d) \ll \tau_{min}(T)$. We can thus distinguish three characteristic limiting cases: a) $\Delta t \gg \tau_{min}(T)$; b) $\Delta t \ll \tau_{min}(T)$ but $\omega\tau_{min}(d) \ll 1$; c) $\omega\tau_{min}(d) \gg 1$.

a) *Low frequencies*: $\omega\tau_{min} \ll T/d$. If

$$\tau_{min}(T) \ll \Delta t\qquad(38)$$

[or, equivalently, if $\omega\tau_{min}(T) \ll T/d \ll 1$], then there exist pairs with $\tau_1(r,T) \lesssim \Delta t$ which manage to relax after crossing the T layer. The energy of the emitted (and absorbed) phonons, averaged over the period, is on the order of T. The conductivity is dominated by pairs with arm r_c determined from the condition $\tau_1(r_c,T) \approx \Delta t$:

$$r_c = \frac{a}{2}\ln\frac{I_0^2}{\omega\tau_{min}(T)dT}.\qquad(39)$$

The functional dependence of the density of such pairs on the distance $\Delta = |\varphi_1 - \varphi_2| \lesssim d$ between levels is

$$g^2(e^2/\varepsilon r_c)r_c^2ad\qquad(40)$$

[we are assuming $e^2/\varepsilon r_c \gg d$; pairs with a large initial splitting Δ have at all times an energy gap exceeding T and do not contribute to $\sigma(\omega)$].

On the average over a period, such a pair transfers an energy on the order of T to the phonon subsystem. Calculating the energy absorbed by such pairs per unit time per unit volume of the semiconductor, and dividing it by $\mathscr{E}_0^2$, we find the conductivity to be

$$\sigma(\omega) = 0.68g^2\frac{e^3}{\varepsilon}r_c^2a\frac{T\omega}{\mathscr{E}_0}.\qquad(41)$$

If $\tau_{min} > \Delta t$, the picture is considerably more complicated. In this case an unexcited pair which has traversed the T layer has a small probability $[\Delta t / \tau_{min}(T)]$ to be excited after capturing a thermal phonon. It emits a phonon whose energy is, in contrast, much higher than the thermal level. Just how large this energy is depends on how rapidly the relaxation time $\tau_1(E)$ falls off with increasing E. In the case at hand, i.e., case A with $T < E < T_a$, we have from (12)

$$\tau_1(E) = \tau_{min}(T)(T/E). \tag{42}$$

The characteristic time in which the pair emits a phonon is given by

$$t^* = \tau_1[E(t^*)]. \tag{43}$$

As an estimate we may assume that the difference between the level energies is proportional to t, $E(t) = d\omega t$, in the relaxation region. We then find

$$t^* = [\tau_{min}(T)\Delta t]^{1/2} = [\tau_{min}(d)\omega^{-1}]^{1/2}. \tag{44}$$

Limiting cases b) and c) correspond to different relations between t^* and ω^{-1}.

b) *High frequencies*, $\omega\tau_{min} > d/T$. If $t^* > \omega^{-1}$ or

$$\omega\tau_{min}(d) \gg 1 \tag{45}$$

(case b), the excited pair does not manage to emit a phonon in one period and instead emits it after several periods, on the average after a time interval $\tau_{min}(d) \gg 1/\omega$. The average energy of the emitted phonon is on the order of d, and the power absorbed by one pair is

$$P \approx \omega d\Delta t / \tau_1(r, T). \tag{46}$$

The absorption is dominated by pairs with $\tau_1(r,T) \approx \tau_{min}$, i.e., with $r = r_T$. As a result, after summing over all pairs, we find

$$\sigma(\omega) = 0.76 g^2 \frac{e^3}{\varepsilon} a r_T^2 \frac{T}{\tau_{min}(T)\mathscr{E}_0} \approx \frac{\sigma_0(\omega)T}{\varepsilon\mathscr{E}_0 r_T}, \tag{47}$$

where $\sigma_0(\omega)$ is given by (33). The numerical factor in (47) can be evaluated by working from the exact equation (53) below.

c) *Intermediate frequencies*, $T/d < \omega\tau_{min} < d/T$. In this case, which corresponds to the condition $t^* < \omega^{-1}$, a pair excited in the T layer manages to emit a phonon in a time t^* far shorter than the period. The characteristic energy of the emitted phonons is on the order of $d\omega t^*$, and the power absorbed by one pair is

$$P \approx \frac{\Delta t}{\tau(r, T)} \frac{d\omega t^*}{\omega^{-1}} \approx \omega T \left[\frac{\Delta t}{\tau_1(r, T)}\right]^{1/2}. \tag{48}$$

The absorption is dominated by pairs with $r = r_T$ [i.e., with $\tau_1 = \tau_{min}(T)$]. As a result we find

$$\sigma(\omega) = 1.99 g^2 \frac{e^{5/2}}{\varepsilon} a r_T^{5/2} \omega^{1/2} T^{1/2} \frac{1}{\mathscr{E}_0^{3/2}\tau_{min}^{3/4}(T)}, \tag{49}$$

where the numerical coefficient is found from (53).

Let us estimate the characteristic values $\mathscr{E}_{c2} = T/er_c$ of the field amplitude $\mathscr{E}_0$ which are required for observing the nonlinearity of the relaxation absorption. With $T = 0.3$ K and $r_T = 2\cdot10^{-6}$ cm we have $\mathscr{E}_{c2} = 10$ V/cm. There is the hope that this value will be below the critical field for

impurity breakdown, but in general $\mathscr{E}_{c2}$ falls off in proportion to T as the temperature is lowered.

3. Nonlinear relaxation absorption; the general case

In general, the nonlinear behavior of the conductivity is determined by the particular dependence of the time τ_1 on the energy E. In several important limiting cases this functional dependence can be described by

$$\tau_1(E) = \tau(T)(T/E)^\nu \text{th}(E/2T). \tag{50}$$

We have $\nu = 1$ in case A at $E < T_a$, in case B at $T_r < E < T_a$, and also in the case in which the piezoelectric interaction is predominant, at $E < T_r$. In addition, we have $\nu = 3$ in case B at $E < T_r$ and $\nu = -1$ in the case of the piezoelectric interaction at $T_r < E < T_a$. The case $\nu = 1$ is analogous to that of dielectric glasses; the case $\nu = -1$ is analogous to that of metallic glasses; and the case $\nu = 3$ so far has no analog in the physics of glasses.

In the case $\nu > 0$ we can replace (44) by
$$t^* = [\tau_{min}(T)(\Delta t)^\nu]^{1/(1+\nu)}.$$
Correspondingly, we can replace (49) by

$$\sigma(\omega) \approx g^2 \frac{e^{(2\nu+3)/(\nu+1)}}{\varepsilon} a r_T^{(\nu+2)/(\nu+1)} \frac{\omega^{1/(1+\nu)} T^{(2\nu+1)/(\nu+1)}}{\mathscr{E}_0^{(2\nu+1)/(\nu+1)}\tau_{min}^{\nu/(\nu+1)}}. \tag{51}$$

With $\nu = 3$ we have

$$\sigma(\omega) \approx g^2 \frac{e^{9/4}}{\varepsilon} a r_T^{5/4} \frac{\omega^{1/4} T^{7/4}}{\mathscr{E}_0^{7/4}\tau_{min}^{3/4}(T)}. \tag{52}$$

The numerical coefficients in the corresponding expressions can be determined from the following exact expression for the power absorbed by one pair[7]:

$$P = \frac{\omega}{8\pi T} \int_0^{2\pi/\omega} \int_0^{2\pi/\omega} dt\, dt' \frac{\dot{E}(t)\dot{E}(t-t')}{\text{ch}^2[E(t-t')/2T]}$$

$$\times \exp\left[-\int_0^{t'} \frac{dt''}{\tau_1(t-t'')}\right] \left\{1 - \exp\left[-\int_0^{2\pi/\omega} \frac{dt''}{\tau_1(t'')}\right]\right\}^{-1}. \tag{53}$$

This expression follows from (26) and (30); τ_1 depends on the time t through the functional dependence $E(t)$. The integrals in (53) can be simplified in various limiting cases, making it possible to evaluate the coefficients in the expressions for $\sigma(\omega)$. This was the procedure which we used to determine the coefficients in expressions (41), (47), and (49).

We are left with the case of the piezoelectric interaction, $\nu = -1$. This case is more complicated than the preceding cases, since low energies, $E < T$, are important under the condition $\omega\tau_{min} < T/d$. We will not go through a detailed analysis for this case, since the functional dependences are similar to those in the case of metallic glasses. We proceed immediately to the results.

In the region $\omega\tau_{min} > T/d$, the result differs from (47) by a numerical factor. At $\omega\tau_{min} < T/d$ the conductivity is on the order of, but lower than, the conductivity in the linear regime, given by (35). The dependence $\sigma(\mathscr{E}_0)$ can be determined only through numerical calculations from Eq. (53). The physical reason for the important difference between the results in this case and those in the case of the deformation interaction is the prolongation of the relaxation of the pair

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populations into the region with $E \gg T$ because of the increase in the relaxation time with increasing E.

In summary, from the standpoint of the nonlinear effects the presence of two stages in the intensity dependence of the absorption is a distinguishing feature of semiconductors and in general of entities which contain two-level systems with a broad distribution of relaxation times. We have in mind the decay of the resonant absorption and, at high intensities, the decay of the relaxation absorption. These stages have been observed experimentally in metallic glasses,[18] and we have explained them elsewhere[19] by similar arguments. Semiconductors do have the distinguishing feature that the critical amplitudes $\mathscr{E}_{c1}$ and $\mathscr{E}_{c2}$ are sharply different (they differ by a factor of only a few units in metallic glasses).

IV. NONRESONANT ABSORPTION IN THE QUANTUM REGION ($\hbar\omega \gg T$)

We have been discussing the relaxation absorption in the classical frequency range, $\hbar\omega \ll T$. Because of the extremely small critical fields required for saturation of the resonant absorption, on the other hand, we should also examine the conductivity in the quantum frequency range, $\hbar\omega \gg T$, at amplitudes $\mathscr{E} \gg \mathscr{E}_{c1}$ given by (4). In this case, we cannot use expression (33) for the nonresonant absorption; instead we must resort to quantum- mechanical theory to calculate $\sigma(\omega)$. The absorption (and the conductivity) is dominated by the pairs with $E \lesssim \hbar\omega$ and $r \approx r_\omega$. For such pairs we have $\omega\tau_1(r_\omega, \hbar\omega) \gg 1$, and the conductivity can be calculated by perturbation theory, by a method analogous to that used in calculating the acoustic and electromagnetic absorption of dielectric glasses.[12,20] The results for the case $\hbar\omega \approx T$ are not easily interpreted, but at $\hbar\omega \gg T$ the results can be described in order of magnitude by the simple general expression

$$\sigma_0(\omega) \approx (e^4/\varepsilon)ag^2r_\omega{}^3\tau_{min}^{-1}(\hbar\omega). \tag{54}$$

This expression differs from (33) in the replacement $T \to \hbar\omega$.

A special stipulation is required in the case

$$\tau_{min}^{-1}(\hbar\omega) \infty \omega$$

(corresponding to piezoelectric semiconductors, with $\hbar\omega \gg T_r$). In this case, expression (54) describes a linear frequency dependence, as in relaxation expression (35) with $\omega\tau_{min} \ll 1$. In the quantum case, however, $\sigma_0(\omega)$ differs from (35) by a factor $1/\omega\tau_{min}(\hbar\omega)$, which does not depend on ω. This factor is small if perturbation theory can be used to describe the electron-phonon interaction.

V. CONCLUSION

In deriving the basic results we have been working primarily from the same model as in Ref. 7. It was assumed there that the energy spread of the levels is of non-Coulomb origin and is much greater than the Coulomb interaction of the carriers over distances on the order of $\bar{r}$, the average distance between centers. A model of this type can correctly describe the situation in amorphous semiconductors, but it is generally unjustified for describing the hopping conductivity in doped semiconductors, where the level spread results from the interaction of electrons with charged centers.

However it turns out that, although the detailed picture of the high-frequency conductivity is different in doped semiconductors, in many cases the frequency, temperature, and amplitude dependences in which we are interested are the same as predicted by the model used above.

In the case of a weakly doped, weakly compensated semiconductor, with $N_A \ll N_D$ (N_A and N_D are the concentrations of acceptors and donors, respectively), there is a charged donor near essentially every negatively charged acceptor, at an average distance on the order of $N_D^{-1/3}$ (a so-called 1-complex[13]). Pairs consisting of a charged donor and a neutral donor separated by a distance less than the average contribute to the high-frequency conductivity. The concentration of such pairs, with arms between r and $r + dr$, is given in order of magnitude by

$$4\pi N_A N_D r^2 dr. \tag{55}$$

The spread of the levels φ_1 and φ_2 in such a pair is determined by the Coulomb interaction of the dipole of the pair with the charged acceptor. In order of magnitude, the difference is

$$|\varphi_1 - \varphi_2| \sim (e^2/\varepsilon)N_D^{1/3}r; \tag{56}$$

this difference is determined by the orientation of the dipole with respect to the acceptor. The distribution function of the concentration of such pairs, with a spread $|\varphi_1 - \varphi_2|$ between Δ and $\Delta + d\Delta$, is

$$g(\Delta, r) \approx (\varepsilon/e^2)N_A N_D^{1/3}r. \tag{57}$$

Working from this distribution function we can easily show that all the results derived above remain valid when g is taken to be

$$g = (\varepsilon/e^2)N_A^{1/3}N_D^{1/3} \tag{58}$$

within a factor on the order of unity.

The case of an intermediate compensation, with $N_A \lesssim N_D$, is treated in the same way. In this case, g must be understood as being a quantity on the order of $(\varepsilon/e^2)N_D^{2/3}$ in all the equations. This quantity is equal to the state density in the impurity band.

The case of strong compensation, with $n \equiv N_D - N_A \ll N_D$, is more complicated. Here the concentration of neutral donors, n, is low, so that it is preferable from the energy standpoint for a neutral donor to lie within a distance less than the average distance from a charged donor. The concentration of pairs of donors whose components are separated by a distance r less than the average distance is

$$N_D(N_D r^3). \tag{59}$$

The electrons preferentially occupy pairs with $r < r_m$, where the characteristic arm r_m is determined from the equality of the density of electrons and the density of such pairs:

$$N_D{}^2r_m{}^3 \approx n, \quad r_m \approx (n/N_D{}^2)^{1/3}. \tag{60}$$

The distribution function of the density of pairs consisting of a neutral donor and a charged donor with $r < r_m$ is[21]

$$n(r^2/r_m{}^3) \approx N_D{}^2r^2. \tag{61}$$

For the level spread $|\varphi_1 - \varphi_2|$ in such pairs we again have (56). Comparing (61) with (55), we conclude that again in this case we should use the state density given by (58) instead of g in all the equations. In other words, we should use the same state density as in the case of an intermediate compensation. This assertion is correct as long as the characteristic values of the arm of the pair, r_c $(r_\omega, r_T$, etc.), are less than or of the order of r_m. The result in this case does not depend on the degree of compensation.

If, on the other hand, the characteristic values of the arm exceed r_m, the probability to find such a pair falls off rapidly with increasing r in accordance with[21] $(r_m/r_c)^l$, where $l > 6$. The result thus differs from that in the case $r_c \ll r_m$ by a factor on the order of $(r_m/r_c)^l \ll 1$, i.e., by the power of the logarithm of the frequency ω or the temperature T.

In conclusion we wish to repeat that many of the results and conclusions of this paper also apply to the absorption of sound in semiconductors in the hopping-conduction regime. In particular, the thermal conductivity of such semiconductors can be proportional to T^2, as in glasses, at sufficiently low temperatures.

We sincerely thank V. V. Bryskin, E. L. Ivchenko, G. E. Pikus, B. I. Shklovskiĭ, and A. L. Éfros for interesting discussions.

[1] And also the absorption of sound.

[2] Böttger and Bryksin[9] were the first to point out this possible mechanism for the nonlinearity of the hopping conductivity in semiconductors. The primary reason for the difference between the results of Ref. 9 and those of the present paper is that we are incorporating the Coulomb correlation, as in Ref. 7. Zvyagin[10] has also studied nonlinear high-frequency effects in semiconductors in the hopping-conductivity region.

[3] In our opinion, it is of limited value to determine the numerical factors in this expression and in the corresponding expressions of the nonlinear theory (discussed below). The results usually contain a comparatively high power of a logarithm, whose argument contains functions of ω, T, and, in the nonlinear case, $\mathscr{E}_0$. The numerical coefficient in the argument of the logarithm remains unknown.

[4] In the range of applicability of the two-site model, we have $r_T N^{1/3} \ll 1$.

[5] We wish to point out that the physical picture of electromagnetic absorption which we have analyzed here applies in many regards to acoustic absorption also, at least under the condition $gr_\omega \ll 1$ (where q is the wave vector of the sound), i.e., when the field of the acoustic wave varies only slightly over the dimensions of the pair.

[6] An evaluation of τ_1 from (17) for case B would lead to a result two orders of magnitude larger.

[7] This expression was derived by one of the present authors (Yu. G.) and also by B. D. Laĭkhtman.

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Translated by Dave Parsons