

Absorption of one of two interacting strong optical waves in a semiconductor

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An analytic study is made of the interband absorption by a semiconductor of a strong optical wave of frequency ω_1 and electric field amplitude F_1 in the presence of a second strong wave of frequency ω_0 and with an electric field F_0 . An interband transition is induced by the alternating electric field of the absorbed wave, whereas intraband states are formed by the simultaneous action of the fields of both waves. Explicit expressions are obtained for the probabilities of the transitions as a result of the absorption of a large number s' of photons of frequency ω_1 in the presence of a large number of photons s of frequency ω_0 . It is shown that the interaction between the waves is manifested most strongly when they are polarized parallel to one another. The absorption spectrum depends strongly on the parity of the total number of photons $s + s'$ participating in the investigated transition. When the frequency detuning $\omega_1 - \omega_0 \equiv \Omega \ll \omega_{0,1}$ is small, a subradiation structure appears in the absorption spectrum in the range $\Omega < \Omega_R = e^2 F_0 F_1 (2\mu\hbar \omega_0 \omega_1)^{-1}$ (μ is the reduced effective band mass). This structure is in the form of high-frequency oscillations with maxima at $\Omega_s = \Omega_R/s$ modulated by the dependences $\Omega^{1/2}$ or $\Omega^{3/2}$. At low values of Ω it is shown that the absorption could change severalfold as a result of a small (within the limits of 10%) change in the ratio of the field amplitudes F_0/F_1 . When the polarizations of the waves are perpendicular (crossed), the interaction between them is weaker. The absorption spectrum depends also on the parity of each of the numbers s and s' of photons participating in the investigated transition.

1. INTRODUCTION

The continuing growth of the physics and technology of semiconductors and widening of the range of applications of strong laser radiation sources has resulted in a permanent interest of both theoreticians and experimentalists in the behavior of semiconductors in the fields of strong optical waves.

Theoretical studies of the influence of a strong monochromatic wave on electron states in a semiconductor were started by the work of Keldysh,¹ who considered many-photon transitions of electrons from the valence to the conduction band. Keldysh's theory of many-photon interband absorption¹ was generalized subsequently²⁻⁴ to the case of action of external static electric² and magnetic^{3,4} fields. A quasi-energy spectrum of carriers in bands linked by the interband resonant part of the interaction between the radiation and a semiconductor was calculated in Refs. 5 and 6; the influence of radiation on intraband motion of particles was allowed, in particular, in Refs. 7 and 8. The coupling between the bands is important in the case of narrow-gap semiconductors and it gives rise to a number of characteristic features in the spectra of many-photon interband magnetoabsorption.^{9,10}

The attention of theoreticians has been attracted recently to the processes of nonlinear interaction of two electromagnetic waves with a semiconductor. One of the waves may be regarded as a probe of the state of a semiconductor in the field of another strong optical wave or both waves may play an equal role, as is true in two-mode lasers. Some problems relating to electron states of a semiconductor subjected to a bichromatic optical field have already been solved.

The coefficient of intraband¹¹ and interband.^{3,12,13} absorption of a probe wave has been calculated on the assumption

that this wave is weak compared with the other wave. The case of adjacent frequencies was considered in Ref. 12. The magnetoabsorption of a weak probe wave in a semiconductor with a wide band gap was studied in Ref. 3, whereas a narrow-gap semiconductor was considered in Ref. 13. Calculations of a quasienergy spectrum of carriers in the field of two strong waves with equal¹⁴ or very different¹⁵ frequencies were reported. The ratio of the wave frequencies assumed in Ref. 15 was such as to exclude the interaction between them: one wave at the resonance frequency was assumed to cause interband transitions and to leave the intraband motion of particles unaffected, whereas the other of much lower frequency was postulated to form intraband states without contributing to transitions between them.

The experimental conditions do not necessarily correspond to the available theoretical results. In the case of a laser with a fixed output frequency the contribution of one photon of a weak probe wave with a fairly narrow frequency tuning range may be insufficient to achieve a resonance between an interband transition and a given number of laser wave photons. In the case of two-mode and also multimode lasers the interaction is between strong radiations of comparable intensity and with such frequency ratios that each of the waves can both influence intraband states of carriers and cause interband transitions. Finally, the experimentally measured quantity is the interband absorption coefficient. It is governed not only by the quasienergy spectra of the bands, the calculation of which is of intrinsic interest,^{14,15} but also by the probability of interband transitions. It would therefore be desirable to consider the general problem of interband absorption (in a semiconductor) of strong interacting waves with arbitrary intensities and frequencies.

We shall report an analytic study of the interband ab-

sorption in a semiconductor of a strong optical wave of frequency ω_1 and with an amplitude of the electric field F_1 in the presence of a second strong wave of frequency ω_0 and with an electric field F_0 . Transitions between isolated hole and electron bands, separated by an energy gap \mathcal{E}_g , are induced by the alternating electric field of the absorbed wave, whereas intraband transient states appear as a result of the simultaneous action of the fields of both waves. We shall obtain expressions for the probabilities $W_{s'}^{(s)}(\omega_1)$ of transitions due to the absorption of a large number s' of photons of frequency ω_1 accompanied by a large number s of photons of frequency ω_0 . These expressions remain qualitatively valid also in the case of small numbers $s, s' = 1, 2, \dots$. The formulas for $W_{s'}^{(s)}(\omega_1)$ are compact in the many-photon limit corresponding to the experimental situation when the wave frequencies $\omega_{0,1}$ are considerably higher than the frequencies of interband tunneling of carriers in the corresponding electric fields $F_{0,1}$. We shall consider two characteristic relative polarizations of the waves: parallel and mutually perpendicular.

The interaction between the waves is manifested most clearly when they are polarized parallel to one another. The absorption spectrum governed by the energy deficit of the optical fields compared with the energy gap $\Delta \approx s\hbar\omega_1 + s'\hbar\omega_0 - \mathcal{E}_g$, is found to depend on the parity of the total number of photons $s + s'$ participating in a transition. If the sum $s + s'$ is odd, then near $\Delta = 0$ resonances we find that $W_{s'}^{(s)} \propto \Delta^{1/2}$, whereas if $s + s'$ is even, then $W_{s'}^{(s)} \propto \Delta^{3/2}$. Consequently, the absorption spectrum for a fixed number s' of photons of frequency ω_1 has a structure with the period ω_0 due to different absorption near resonances differing in respect of the parity of $s + s'$. The complete spectrum is governed also by its dependence on the electric field, which in the case of different frequencies is directly proportional to the quantity $F_0^{2s} F_1^{2s'}$.

The interaction of parallel-polarized waves creates an additional structure in the many-photon absorption spectrum. This structure appears for a small frequency detuning $\Omega \equiv \omega_1 - \omega_0$ in the range

$$\Omega < \Omega_R, \quad \Omega_R = e^2 F_0 F_1 (2\mu\hbar(\omega_0\omega_1)) \ll \omega_{0,1},$$

where μ is the reduced effective band mass. It represents high-frequency oscillations with maxima at $\Omega_s = \Omega_R/s$, where $s = 1, 2, \dots$, modulated by smooth dependences of the $\Delta^{1/2}$ or $\Delta^{3/2}$ type. The appearance of such oscillations is an indication that the frequency of a probe wave ω_1 approaches the frequency ω_0 and may be used to determine the latter frequency or to measure it more accurately. The separations between the maxima contain information on the characteristics of the optical waves ($F_0, F_1, \omega_0, \omega_1$) and on the reduced carrier mass μ . Such a subradiation structure may be used as the basis of a semiconductor device with a high selectivity in respect of the frequency of the absorbed radiation. Moreover, the possibility of a large change in the absorption as a result of a slight change in the ratio of the fields F_0/F_1 , established below, may also be of practical importance; a change in this ratio within 10% can alter the absorption severalfold.

The structure in question was observed experimentally in the absorption of a component of a bichromatic field in two-level atomic systems.¹⁶ A theoretical interpretation of the structure of Ref. 16 and the associated effects were considered in Ref. 17.

If the waves are polarized perpendicularly to one another, their interaction is weak. The absorption spectrum of one of them now depends on the parity of each of the numbers s and s' of the photons participating in an interband transition. Near the $\Delta = 0$ resonances the frequency spectrum of the absorption is governed by the functions $\Delta^{1/2}$, $\Delta^{3/2}$, and $\Delta^{5/2}$ depending on the parity of s and s' . The influence of the electric fields of the waves on the absorption intensity is then characterized by the factor $F_0^{2s} F_1^{2s'}$ for any ratio of the frequencies.

2. PROBABILITY OF MANY-PHOTON INTERBAND ABSORPTION. GENERAL THEORY

Electron-hole states in semiconductors subjected to external fields varying slowly in space compared with internal crystal fields are described by the effective mass method, generalized to the case of time-dependent external fields in Ref. 3. In our case the external field is an alternating electric field of two optical waves:

$$\mathbf{F}(t) = \xi_0 F_0 \cos \omega_0 t + \xi_1 F_1 \cos \omega_1 t, \quad (1)$$

where ω_j, F_j and ξ_j ($j = 0$ or 1) are the frequencies, amplitudes of the electric fields, and polarization unit vectors of the waves. Spatial dispersion is ignored in the optical frequency range and this corresponds to the dipole approximation.

We shall be interested in the absorption of just one of the waves so that we shall assume that the transient states of carriers in the electron and hole bands appear under the action of the total field (1), whereas interband transitions are caused only by the field $\xi_1 F_1 \cos \omega_1 t$. This field causes a crystal to undergo a transition from the ground state to an excited one and this is accompanied by the formation of an electron-hole pair.¹⁸ Since in the dipole approximation the total momentum of such a pair and the photon momenta are ignored, we shall consider only the relative motion of an electron and a hole characterized by the coordinate \mathbf{r} .

In the effective mass method³ the probability of such an interband transition per unit time and per unit volume is $W(\omega_1)$ and it is described by the following sum over the final states:

$$W(\omega_1) = \frac{1}{t} \sum_f |a(t)|^2, \quad (2)$$

where

$$a(t) = \frac{1}{i\hbar} \int_0^t \mathcal{H}'(\tau) \int \psi^*(\mathbf{r}, \tau) \psi_0(\mathbf{r}, \tau) d\mathbf{r} d\tau, \quad (3)$$

ψ_0 and ψ are the functions of the initial and final states, and \mathcal{H}' is the operator of the dipole-allowed interband transition.

We shall consider the case of orbitally nondegenerate parabolic electron (e) and hole (h) bands with effective carrier

masses m_e and m_h , separated by an energy gap \mathcal{E}_g sufficiently wide for us to ignore the action of an electric field on the coupling between the bands in the zeroth approximation. Then, the function $\psi(\mathbf{r})$ describing the newly formed electron-hole pair satisfies the equation

$$-\frac{\hbar^2}{2\mu} \nabla^2 \psi(\mathbf{r}, t) - e\mathbf{F}(t) \mathbf{r} \psi(\mathbf{r}, t) = i\hbar \frac{\partial \psi(\mathbf{r}, t)}{\partial t}, \quad (4)$$

$$\mu^{-1} = m_e^{-1} + m_h^{-1},$$

and the operator \mathcal{H}' has the form³

$$\mathcal{H}' = (ie\hbar F_1(\xi_1 \mathbf{p}_{eh}) / m_0 \mathcal{E}_g) \cos \omega_1 t, \quad (5)$$

where m_0 is the mass of a free electron and \mathbf{p}_{eh} is the matrix element of the momentum calculated between the Bloch amplitudes of the electron and hole bands.

A solution of Eq. (4) is

$$\psi(\mathbf{r}, t) = (2\pi)^{-3/2} \exp[i(\mathbf{k}(t) \mathbf{r} + \varphi(t))], \quad (6)$$

where

$$\mathbf{k}(t) = \frac{e}{\hbar} \int_0^t \mathbf{F}(\tau) d\tau + \mathbf{q} \quad (\mathbf{q} = \text{const}), \quad (7)$$

$$\varphi(t) = -\frac{\hbar}{2\mu_0} \int_0^t \mathbf{k}^2(\tau) d\tau. \quad (8)$$

The initial state of a crystal before its interaction with the radiation is described by the function $\psi_0 = \delta(\mathbf{r})$ (Ref. 18). Substituting this function together with the function (6), calculated for the field $\mathbf{F}(t)$ of Eq. (1), and together with Eq. (5) into Eq. (3), we find that integration with respect to \mathbf{r} gives

$$a(t) = \frac{eF_1(\xi_1 \mathbf{p}_{eh})}{(2\pi)^3 m_0 \mathcal{E}_g} \int_0^t \exp(i\mathcal{E}\tau/\hbar) M_0(\tau) M_1(\tau) \times M_+(\tau) \overline{M_-(\tau)} d\tau, \quad (9)$$

where

$$\mathcal{E} = \mathcal{E}_g + \frac{\hbar^2 q^2}{2\mu} + \frac{e^2}{4\mu} \left(\frac{F_0^2}{\omega_0^2} + \frac{F_1^2}{\omega_1^2} \right) \quad (10)$$

is the quasienergy with allowance for the band gap \mathcal{E}_g ,

$$M_j(t) = \exp \left\{ \frac{i\hbar}{2\mu} \left[-\frac{e^2 F_j^2}{4\hbar^2 \omega_j^3} \sin 2\omega_j t + \frac{2eF_j}{\hbar \omega_j^2} \mathbf{q} \xi_j (1 - \cos \omega_j t) \right] \right\} \times (\delta_{j0} + \delta_{j1} \cos \omega_1 t), \quad (11)$$

$$M_\alpha(t) = \exp \left[\pm i \frac{\xi_0 \xi_1 e^2 F_0 F_1 \sin \omega_\alpha t}{2\mu \hbar \omega_0 \omega_1 \omega_\alpha} \right]. \quad (12)$$

In Eq. (11) and (12) we have $j = 0$ or 1 and $\alpha = +$ or $-$; for $\alpha = (-)$, we take the upper sign in the argument of the exponential function, whereas $\alpha = (+)$ we take the lower sign; $\omega_\pm = \omega_1 \pm \omega_0$.

The function M_β periodic in time ($\beta = 0, 1, +, -$)

can be represented by the series

$$M_\beta(t) = \sum_{\nu_\beta = -\infty}^{+\infty} A_{\nu_\beta}^{(\beta)}(\omega_\beta) \exp(-i\nu_\beta \omega_\beta t), \quad \beta = 0, 1, +, -; \quad (13)$$

$$A_{\nu_\beta}^{(\beta)}(\omega_\beta) = \frac{\omega_\beta}{2\pi} \int_{-\pi/\omega_\beta}^{+\pi/\omega_\beta} M_\beta(t) \exp(i\nu_\beta \omega_\beta t) dt. \quad (14)$$

Substituting these series into Eq. (9), we shall group in the argument of the exponential function in the integral all the terms containing the frequencies ω_0 and ω_1 . We shall relabel the coefficients in front of the frequencies by the application of the rules $\nu_0 + \nu_+ - \nu_- = s$, and $\nu_1 + \nu_+ - \nu_- = s'$. Then, integrating in Eq. (9) and substituting the result into Eq. (2), we obtain in the usual way (in the limit of large values of t) an expression for the interband transition probability in the form

$$W(\omega_1) = \sum_{s'} W_{s'}(\omega_1), \quad W_{s'}(\omega_1) = \sum_s W_{s'}^{(s)}(\omega_1),$$

$$W_{s'}^{(s)}(\omega_1) = \frac{|\xi_1 \mathbf{p}_{eh}|^2 e^2 F_1^2}{(2\pi)^3 m_0^2 \mathcal{E}_g^2} 2\pi \hbar \times \int d\mathbf{q} |G_{s'}^{(s)}(\omega_1, \mathbf{q})|^2 \delta(\mathcal{E} - s\hbar\omega_0 - s'\hbar\omega_1), \quad (15)$$

where

$$G_{s'}^{(s)}(\omega_1, \mathbf{q}) = \sum_{\nu_+, \nu_-, \nu_0 = -\infty}^{+\infty} A_{s-\nu_++\nu_-}^{(0)} A_{s'-\nu_+-\nu_-}^{(1)} A_{\nu_+}^{(+)} A_{\nu_-}^{(-)}. \quad (16)$$

The quantity $W_{s'}^{(s)}(\omega_1)$ is the probability of an interband transition due to the absorption of s' photons of frequency ω_1 in the presence of s photons of frequency ω_0 . In the derivation of Eq. (15) an allowance is made for the fact that the summation in Eq. (2) represents integration with respect to \mathbf{q} . Then, $W_{s'}(\omega_1)$ is the probability of a transition in which the absorption of these photons is accompanied by all possible contributions of photons from the field of the other wave.

The coefficients (14) with the indices $(+)$ and $(-)$ can be calculated explicitly. Substituting Eq. (12) into Eq. (14), we obtain

$$A_{\nu_+}^{(+)} = J_{\nu_+}(|\xi_0 \xi_1 \gamma_+^{-2}|), \quad \gamma_\pm^{-2} = \frac{e^2 F_0 F_1}{2\mu \hbar \omega_0 \omega_1 \omega_\pm}; \quad (17)$$

$$A_{\nu_-}^{(-)} = \left(-\frac{\omega_-}{|\omega_-|} \right)^{\nu_-} J_{\nu_-}(|\xi_0 \xi_1 \gamma_-^{-2}|) \quad (18)$$

(J_ν are Bessel functions of real argument).

If we substitute the coefficients (17) and (18) together with the integral representations of the coefficients $A_{\nu'}^{(0)}$ and $A_{\nu'}^{(1)}$ of Eq. (14) into Eq. (16), the sum in the latter equation becomes a product of generating functions for Bessel functions and is then calculated explicitly. In the resultant expression for $G_{s'}^{(s)}$, following Ref. 3, it is convenient to represent the arguments of the exponential functions in Eq. (11) in

integral form such as in Eqs. (6) and (8). Adopting then new variables $\sin \omega_0 t = u$, and $\sin \omega_1 t' = u'$, we finally obtain

$$G_s^{(s')}(\omega_1, \mathbf{q}) = \frac{1}{(2\pi)^2} \oint du \oint du' (1-u^2)^{-1/2} \\ \times \exp \left\{ i \left[\lambda_0 \Phi_0(u) + \lambda_1 \Phi_1(u') \right. \right. \\ \left. \left. - (\xi_0 \xi_1) \frac{1}{\gamma_+^2} [u(1-u'^2)^{1/2} + (1-u^2)^{1/2} u'] \right. \right. \\ \left. \left. + (\xi_0 \xi_1) \frac{1}{\gamma_-^2} [u(1-u'^2)^{1/2} - (1-u^2)^{1/2} u'] \right] \right\}, \quad (19)$$

where

$$\Phi_j(u) = \int_0^u (1-v^2)^{-1/2} \left(1 + 2 \frac{\eta_j \xi_j}{\gamma_j} v + \frac{1}{\gamma_j^2} v^2 \right) dv, \\ \eta_j = \frac{\hbar \mathbf{q}}{(2\mu E_j)^{1/2}}, \quad E_j = s_j \hbar \omega_j - \frac{e^2 F_j^2}{4\mu \omega_j^2} \quad (s_0 \equiv s, \quad s_1 \equiv s'), \\ \gamma_j^2 = \frac{2\mu E_j \omega_j^2}{e^2 F_j^2}, \quad \lambda_j = \frac{E_j}{\hbar \omega_j} \quad (j=0, 1).$$

The integration in Eq. (19) is carried out over closed contours in complex planes u and u' , including cuts drawn from -1 to $+1$.

We shall calculate the integrals in Eq. (19) in analytic form by assuming, following Refs. 1, 3, 9, 10, and 13, that the parameters $\lambda_j \gg 1$ are large ($j = 0$ or 1 ; adiabatic approximation). The expressions obtained in this way will be given later in the many-photon limit $\gamma_j^2 \gg 1$, when the frequency of an alternating electric field ω_j is much higher than the frequency of interband tunneling $eF_j (2\mu E_j)^{-1/2}$. The large parameters of Eq. (19) then become the photon numbers $\lambda_j = s_j (E_j = s_j \hbar \omega_j)$. Moreover, the case of isolated parabolic electron and hole energy bands implies that the vectors η_j are small. This corresponds to the proximity of the radiation energy of the absorption edge. Therefore, the range of validity of the results given below is set by the conditions

$$s_j \gg 1, \quad \gamma_j^2 = \frac{2\mu s_j \hbar \omega_j^3}{e^2 F_j^2} \gg 1, \quad \gamma_+^2 \gg 1, \quad \eta_j \ll 1. \quad (20)$$

In a crystal characterized by an effective mass $\mu = 0.1m_0$ and a band gap $\mathcal{E}_g = 0.6$ eV the absorption of $s_j = 3$ phonons of an optical field with an amplitude $F_j = 10^5$ V/cm and a frequency $\omega_j = 2 \times 10^{14}$ sec $^{-1}$ corresponds to the parameter $\gamma_j^2 \approx 150$.

We shall now consider different relative polarizations of the two waves.

3. PARALLEL POLARIZATIONS OF THE WAVES ($\zeta_0 \zeta_1 = 1$). DIFFERENT FREQUENCIES

The existence of large parameters $\lambda_j = s_j \gg 1$ makes it possible to calculate the integrals in Eq. (19) by the saddle-point method.^{1,3} The saddle points, which are determined by the roots of the equations $\Phi_j' = 0$, are $\pm i\gamma_j - \gamma_j \eta_{j\parallel}$, where

$\eta_{j\parallel}$ and q_{\parallel} are the projections of the vectors η_j and \mathbf{q} along the directions of the wave polarizations. In view of the inequality $\gamma_j \gg 1$, the saddle points are located at high values of u and u' . We can show that in the range of such values when the frequency detuning $\Omega = \omega_1 - \omega_0$ satisfies the condition

$$\Omega \approx \omega_- \gg e^2 F_0 F_1 / 2\mu \hbar \omega_0 \omega_1, \quad (21)$$

the argument of the exponential function of Eq. (19) containing γ_+^{-2} is small and does not affect the integral. The coefficient in front of γ_+^{-2} is allowed for in the vicinity of a saddle point and it is represented by a linear function with a suitable variable. Omitting cumbersome transformations, we shall give the final result for the frequencies differing in the sense of Eq. (21):

$$G_s^{(s')}(\omega_1, q_{\parallel}) = \frac{\gamma_1 \exp[-i\pi(s+s'-1)/2]}{2\pi(ss')^{1/2}(1-g)^{1/2}} \\ \times \exp \left\{ -s \ln 2\gamma_0 - s' \ln 2\gamma_1 \right. \\ \left. + \frac{(s+s')(1-3g)}{2(1-g)} \right\} \{ e^{-Q} \sin[2s_+ \eta_{0\parallel} - 2s_+' \eta_{1\parallel} - (s-s')\pi/2] \\ - e^Q \sin[2s_- \eta_{0\parallel} + 2s_-' \eta_{1\parallel} - (s+s')\pi/2] \}; \quad (22)$$

here,

$$0 < g = \frac{\omega_1 \omega_0}{(\omega_1 + \omega_0)^2} \leq \frac{1}{4}, \quad Q = \frac{2(gss')^{1/2}}{1-g}, \\ s_{j,\pm} = \frac{s_j}{1-g} \pm \frac{1}{2} Q.$$

It follows from Eq. (22) that the absorption depends on the parity of the total number of photons $s + s'$. If the sum $s + s'$ is even, then $G \propto q_{\parallel}$, whereas if it is odd, then $G \propto \text{const}$ in the limit $q_{\parallel} \rightarrow 0$. This gives the following results for the transition probability (15):

$$W_s^{(s')}(\omega_1) = \frac{|\xi_1 \mathbf{p}_{eh}|^2 (\hbar \omega_1)^3 \mu}{2\pi^2 \mathcal{E}_g^{3/2} m_0^2 \hbar s (1-g)} \left(\frac{2\mu}{\hbar^2} \right)^{3/2} \\ \times \exp \left[\frac{(s+s')(1-3g)}{1-g} \right] \\ \times (4\gamma_0^2)^{-s} (4\gamma_1^2)^{-s'} f_{\parallel s}^{(s')}(\omega_1), \quad (23)$$

for the odd total number of photons $s + s' = 2m + 1$ or $s - s' = 2n + 1$ ($m, n = 0, 1, 2, \dots$; s and s' have different parities), we have

$$f_{\parallel s}^{(s')}(\omega_1) = [(-1)^n e^{-Q} - (-1)^m e^Q]^2 (\Delta/\mathcal{E}_g)^{1/2}, \quad (24)$$

whereas for the even total number of photons $s + s' = 2m$ or $s - s' = 2n$ ($m, n = 1, 2, \dots$; s and s' have the same parity),

we find that

$$f_{\parallel s'}^{(s)}(\omega_1) = \frac{4}{3} \left\{ (-1)^n e^{-Q} \left[\left(\frac{s_+^2 \mathcal{E}_g}{s \hbar \omega_0} \right)^{1/2} - \left(\frac{s_+'^2 \mathcal{E}_g}{s' \hbar \omega_1} \right)^{1/2} \right] - (-1)^m e^Q \left[\left(\frac{s_-^2 \mathcal{E}_g}{s \hbar \omega_0} \right)^{1/2} + \left(\frac{s_-'^2 \mathcal{E}_g}{s' \hbar \omega_1} \right)^{1/2} \right] \right\}^2 \left(\frac{\Delta}{\mathcal{E}_g} \right)^{1/2}. \quad (25)$$

In Eqs. (24) and (25), we have used the quantity

$$\Delta = s \hbar \omega_0 + s' \hbar \omega_1 - \mathcal{E}_g - \frac{e^2}{4\mu} \left(\frac{F_0^2}{\omega_0^2} + \frac{F_1^2}{\omega_1^2} \right)$$

and the parameters γ_j are defined in Eq. (20).

If we consider s' -photon absorption as a function of the frequency ω_1 [i.e., $W_{s'}(\omega_1)$], we find that it has a structure with the period ω_0 . This structure is due to the fact that the step-like dependence of the $\Delta^{1/2}$ type near a resonance $\Delta = 0$ corresponding to a certain value of s changes to a steeper dependence $\Delta^{3/2}$ for the neighboring value of s .

The absorption coefficient of different numbers of photons s and s' is also governed by the factor $F_0^{2s} F_1^{2s'}$ containing electric fields.

In the case of very different frequencies when $g \ll 1$, $s_{\pm} = s$, $s'_{\pm} = s'$, and $Q \ll 1$, an additional dependence on the parity appears for each of the types of absorption. If the total number of photons is odd, then the functions f_{\parallel} of Eq. (24) differ by the factor $Q^2 \ll 1$ when n and m have the same and different parities, respectively. When the total number of photons is even, the ratio of the corresponding functions f_{\parallel} of Eq. (25) is $s' \omega_0 / s \omega_1$.

The expression for the transition probability in the case of a weak absorbed wave with an amplitude of an electric field $F_1 \ll F_0$ can be obtained from Eqs. (23)–(25) where we have to substitute $s' = 1$. The same qualitative result is obtained also by self-consistent calculation if right from the beginning we ignore the influence of a weak field on intraband carrier states. It follows that the use of the saddle method gives results which are qualitatively correct not only in the case of large, but also in the case of arbitrary numbers of photons s and s' .

If the strong optical field is monochromatic ($F_0 = 0$), then $\gamma_+^{-1} = \gamma_-^{-1} = \gamma_0^{-1} = 0$, and the integral with respect to u in Eq. (19) differs from zero for $s = 0$. Further integration yields the familiar results of Ref. 1:

$$W_{s'}(\omega_1) \propto (\gamma_1^2)^{-s'} \left(s' \hbar \omega_1 - \mathcal{E}_g - \frac{e^2 F_1^2}{4\mu \omega_1^2} \right)^{\nu},$$

where $\nu = 1/2$ applies in the case of odd values of s' and $\nu = 3/2$ in the case of even values of s' .

4. PARALLEL POLARIZATIONS OF THE WAVES ($\zeta_0 \zeta_1 = 1$). ADJACENT FREQUENCIES

If the frequency detuning is small $\Omega \equiv \omega_- \ll \omega_j$ ($j = 0$ or 1), then in the integrals of Eq. (19) expressed in terms of the variables t and t' we can make the substitutions $\Omega t_1 = \omega_0 t$ and $\Omega t_2 = \omega_1 t'$. In the limit of small values of Ω Eq. (19) is a

product of two integrals, the first of which contains the variable $t_1 - t_2$ and the second, for example, the variable t_2 . In the first integral there is only the exponential function $t_1 - t_2$ and it is calculated in an elementary manner. The second integral is similar to that which is encountered in Sec. 3. It contains a large parameter in the form of a factor of the function Φ [see Eq. (19)] and depends on the total field $F_0 + F_1$. Integration is again carried out by the saddle-point method. After substitution of the above expression for G from Eq. (19) to Eq. (15), we obtain the transition probability in the form

$$W_i^{(s)}(\omega_1) = \frac{2 |\zeta_1 \mathbf{p}_{eh}|^2 (\hbar \omega_0)^3 \mu e^N}{\pi^2 m_0^2 \mathcal{E}_g^{3/2} \hbar} \left(\frac{2\mu}{\hbar^2} \right)^{1/2} \left(\frac{F_1}{F_1 + F_0} \right)^2 \left(\frac{1}{4\gamma^2} \right)^N \times \Lambda_s(\Omega) \left\{ \frac{4}{3} N \frac{\mathcal{E}_g}{\hbar \omega_0} \left(\frac{\Delta}{\mathcal{E}_g} \right)^{1/2} \cos^2 \frac{N\pi}{2} + \left(\frac{\Delta}{\mathcal{E}_g} \right)^{1/2} \sin^2 \frac{N\pi}{2} \right\}. \quad (26)$$

Here,

$$\Lambda_s(\Omega) = \frac{\sin^2 \pi (\Omega_R / \Omega - s)}{\pi^2 (\Omega_R / \Omega - s)^2};$$

$$\Delta = l \hbar \omega_0 + \hbar \Omega N - \mathcal{E}_g - \frac{e^2}{4\mu \omega_0^2} (F_0 + F_1)^2;$$

$$\gamma^2 = \frac{2\mu N \hbar \omega_0^3}{e^2 (F_0 + F_1)^2}; \quad N = l + \left(\frac{\Omega_R}{\Omega} - s \right); \quad (27)$$

l is the total number of photons.

We shall now consider the total probability of an l -photon transition

$$W_l(\omega_1) = \sum_s W_i^{(s)}(\omega_1) \quad (28)$$

as a function of the detuning $\Omega = \omega_1 - \omega_0$, assuming that the frequency ω_0 is fixed and also that the fields F_0 and F_1 are constant.

When the frequencies coincide exactly, so that $\Omega \rightarrow 0$, we find from

$$\sum_s \Lambda_s \rightarrow \int d \left(\frac{\Omega_R}{\Omega} - s \right) \delta \left(\frac{\Omega_R}{\Omega} - s \right)$$

that the earlier expression¹ is obtained for the probability of absorption of $N = l$ photons of an optical wave with an electric field amplitude $F_0 + F_1$.

The main dependence of the transition probability (28) on the detuning is given by the functions $\Lambda_s(\Omega)$ of Eq. (27). Each of them represents a rapidly oscillating function with a period which is small ($\Omega_R \ll \omega_j$), and decreases on reduction in Ω . The function Λ_s reaches a single maximum at resonance values of the detuning $\Omega_s = \Omega_R / s$ and we then have $N = l$. Oscillations of the function $\Lambda_s(\Omega)$ are modulated by a smooth dependence on Ω contained in the functions $\Delta^{1/2}$ or $\Delta^{3/2}$.

Obviously, the absorption in the case of resonant detuning is possible if $\Delta(\Omega_s) > 0$. Therefore, if the frequency ω_0 is such that

$$-\frac{\hbar\Omega_R l}{s_0} = l\hbar\omega_0 - \mathcal{E}_g - \frac{e^2(F_0 + F_1)^2}{4\mu\omega_0^2} \ll 0, \quad (29)$$

then s_0 resonances will be observed for detunings $\Omega_s \geq \Omega_R/s_0$ ($s = 1, 2, \dots, s_0$). For smaller detuning the absorption is impossible because the energy of a given number of photons is insufficient to excite an interband transition. The dependence of the absorption probability on the detuning, subject to the condition (29), is described by the expression

$$W_l(\Omega) \propto \sum_s \Lambda_s(\Omega) \cdot \begin{cases} (N\Omega - l\Omega_R/s_0)^{1/2} \\ (N\Omega - l\Omega_R/s_0)^{3/2} \end{cases}, \quad (30)$$

where the upper row applies when l is odd and the lower when l is even. The function $W_l(\Omega)$ of Eq. (30) is plotted in Fig. 1.

If the frequency ω_0 corresponds to the absorption edge and satisfies the condition (29) in the limit $s_0 \rightarrow \infty$, then an unlimited number of resonances is possible right down to $\Omega = 0$. The curves in the figure then begin from the value $\Omega = 0$.

The half-width $\Delta\Omega_s$ of a subradiation resonance maximum is easily estimated to be $\Delta\Omega_s \approx \Omega_s/2s$. For typical parameters $\mu = 0.1m_0$, $\omega_0 \approx \omega_1 = 2 \times 10^{14} \text{ sec}^{-1}$, $F_0 \approx F_1 \approx 10^5 \text{ V/cm}$, $\Omega_R \approx 4 \times 10^{12} \text{ sec}^{-1}$, we find that $\Delta\Omega_3 \approx 2.2 \times 10^{11} \text{ sec}^{-1}$. The relative narrowness of the maxima and their tendency to become narrower on reduction in the detuning can also be used in developing a semiconductor device with a fairly high selectivity in respect of the absorbed radiation frequency.

In the adjacent frequency case discussed here it is possible to influence significantly many-photon absorption by a slight change in the optical wave intensity. An analysis of Eq. (26) shows that in the vicinity of a resonance $\Omega = \Omega_s$ a change in the amplitude of the absorbed field $F_1 \approx F_0$ by 10% reduces the transition probability W_l by a factor exceeding 3.

The subradiation structure $W_l(\Omega)$ analogous to that discussed above has been observed in experiments on the ab-

sorption of a component of a bichromatic field in a two-level atomic system (^{113}Cd vapor).¹⁶ This structure is justified theoretically in Ref. 17. We shall conclude this section by noting that the very appearance of a subradiation structure in the absorption of one of the two waves is a qualitative sign of the proximity of their frequencies. This circumstance can be used to determine the frequency of a strong optical wave using a different strong wave with known characteristics as the probe.

5. MUTUALLY PERPENDICULAR POLARIZATIONS OF THE WAVES ($\zeta_0 \zeta_1 = 0$)

In this case Eq. (19) is a product of two integrals of one variable u or u' , each of which corresponds to a transition associated with the absorption of a monochromatic wave. Therefore, the interaction between the waves is weakened in the selected polarization.

The integrals in Eq. (19) are calculated by the saddle-point method in which the large parameters are the numbers of photons s and s' . Substituting the function $G_s^{(s)}$ found for an arbitrary ratio of the frequencies ω_0 and ω_1 into Eq. (15), we obtain the following expression for the transition probability:

$$W_s^{(s)}(\omega_1) = \frac{2|\zeta_1 \mathbf{p}_{eh}|^2 (\hbar\omega_1)^3 \mu}{\pi^3 m_0^2 \mathcal{E}_g^{3/2} \hbar s} \left(\frac{2\mu}{\hbar^2} \right)^{1/2} \times e^{s+s'} \left(\frac{1}{4\gamma_0^2} \right)^s \left(\frac{1}{4\gamma_1^2} \right)^{s'} f_{\perp s'}^{(s)}(\omega_1), \quad (31)$$

where in the case of odd s' and even s , we have

$$f_{\perp s'}^{(s)}(\omega_1) = (\Delta/\mathcal{E}_g)^{1/2}; \quad (32)$$

when both s' and s are even, we find that

$$f_{\perp s'}^{(s)}(\omega_1) = \frac{4s'}{3} \left(\frac{\mathcal{E}_g}{\hbar\omega_1} \right) \left(\frac{\Delta}{\mathcal{E}_g} \right)^{3/2}; \quad (33)$$

when s' is even and s is odd, we obtain

$$f_{\perp s'}^{(s)}(\omega_1) = \frac{16ss'}{15} \left(\frac{\mathcal{E}_g^2}{\hbar^2 \omega_1 \omega_0} \right) \left(\frac{\Delta}{\mathcal{E}_g} \right)^{5/2}, \quad (34)$$

where Δ is defined just after Eqs. (24) and (25).

The function $f_{\perp s'}^{(s)}$ corresponding to odd both s' and s can be obtained from Eq. (33) by the substitutions $s' \rightarrow s$ and $\omega_1 \rightarrow \omega_0$.

In contrast to the parallel polarization case [Eqs. (24) and (25)], the absorption now depends on the parity of each of the photon numbers s and s' separately. Another difference is that in the perpendicular polarization the frequency dependence can vary over a wider range: $\Delta^{1/2}$, $\Delta^{3/2}$, and $\Delta^{5/2}$, whereas in the parallel polarization case we have two possibilities: $\Delta^{1/2}$ and $\Delta^{3/2}$. This results in a stronger structure in the case of s' -photon absorption [$W_{s'}(\omega_1)$] regarded as a function of the frequency ω_1 of the absorbed wave.

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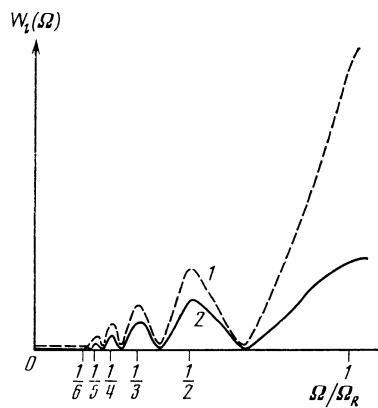


FIG. 1. Qualitative dependence of the probability $W_l(\Omega)$ [Eq. (30)] of an l -photon transition on the relative detuning Ω/Ω_R for $\Omega \geq \Omega_R/6$: 1) even values of l ; 2) odd values of l . The rising branches are described by the law $(\Omega - \Omega_R/6)^\alpha$, where $\alpha = 3/2$ for even values of l and $\alpha = 1/2$ for odd values of l .

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