# The optical Stark effect in a transient double resonance in semiconductors

E. Yu. Perlin

*S. I. Vavilov State Optical Institute, 199034 St. Petersburg, Russia*

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The optical Stark effect in bulk semiconductors and in materials with quantum wells, wires, and dots is considered. The effect is due to the interaction of an electronic system with intense light whose frequency $\omega$ is small in comparison with the forbidden zone, but falls on the resonance between two conduction bands $c$ and $c'$ or between two sub-bands of dimensional quantizations $cn'$ and $cn$ of the $c$ band. Probing of states mixed by this light is effected by two femtosecond pulses, one following the other with delay time $\tau_D$, of light of a different frequency $\Omega$, which is in resonance with the adjacent transition between the valence band $v$ and the $c$ band. The polarization of the medium induced by the first probing light pulse influences the absorption of the second pulse, the dependence of which on $\tau_D$ is calculated by the density matrix method. In the presence of pumping $\omega$, depending on the type of band structure, one observes either decay of the induced polarization with characteristic time $\sim\omega_1^{-1}$ ($\omega_1$ is the Rabi frequency), or oscillations with frequency $\sim\omega_1$. Such types of time dependence correspond to various spectral manifestations of the Stark effect in the steady-state regime—that is, the appearance of a break or a gap in the energy band spectrum in the field of a strong light wave.

## 1. INTRODUCTION

Beginning with Refs. 1–3, phenomena associated with rearrangement of the energy spectra of crystals in the field of a strong electromagnetic wave have become the subject of theoretical, and later experimental, studies.[4–10] Already in Refs. 1–3 it was shown that in a resonance at frequency $\omega$ between an intense light beam and an interband transition, either a break or a gap arises in the energy spectrum of a crystal, brought about by the optical Stark effect. The interaction $V$ of the electronic subsystem of the crystal with the field of the wave under resonance conditions removes the degeneracy in the electron–photon system. Accordingly, the Stark effect is linear in the field intensity of the light wave. The peculiarities of the spectrum which arise can be manifested in various ways—in particular, in the absorption of sound or of probing light. In the latter case, under certain conditions a region of transparency can appear against the background of the fundamental absorption band. The width of the absorption region (the width of the gap in the spectrum) is linear in the field of the light wave at light intensities such that the interaction energy $V \ll \hbar\omega$. At very high light intensities and/or at transitions in the far infrared, the magnitude of $V$ ceases to be small in comparison with $\hbar\omega$, and in this case the dependence of the gap width on the field becomes very complicated and even nonmonotonic.[11]

Most papers dedicated to the Stark effect in crystals in an intense light field have examined the situation in which the intense light is in resonance with the transition between states of the upper valence band and the lower conduction band or between the ground state of the crystal and an exciton level. In this case, there are a number of optical processes which take place against the background of the optical Stark effect. States in bands near the extrema become filled, with a corresponding decrease of the effective width of the forbidden band $E_g$, thanks to numerous correlation effects. On the other hand, the dynamic Moss-Burstein effect leads to a shortwave shift of the absorption region.

Thanks to progress in the fabrication technology of semiconductor quantum wells and other low-dimensional structures, with relatively easily varied parameters and large-amplitude optical nonlinearities, and also thanks to the development of the technique of ultrashort light pulses, it has become possible in recent years to investigate nonlinearities of such kinds not only theoretically, but also experimentally.[8–10,12–14] The data that have been obtained as a result of these efforts indicate that the dynamics of coherent processes are substantially determined by the concentration of free electrons and holes generated by the intense light. It was shown, in particular, that coherent optical effects in excitons at high excitation levels differ radically from analogous effects in two-level systems.[15]

In connection with the above it is clear that, in principle, it would be desirable to study the dynamic Stark effect in crystals in the field of an intense, resonant light wave under conditions in which the radiation does not cause a marked change in the populations of the electronic system. Such conditions are realized by a double optical resonance in which the frequency $\omega$ of the intense radiation falls in resonance between two excited states $|1\rangle$ and $|2\rangle$ (band states, excitonic states, impurity states), and transitions between the ground state $|0\rangle$ and one of the excited states $|1\rangle$ are caused by weak radiation with frequency $\Omega$. Such a situation has been considered theoretically for cases of double optical resonance in interband transitions,[2,3] excitonic transitions,[16] exciton–band transitions where the intense light induces a Fano resonance,[17] in transitions be-

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tween the Landau levels in a quantizing magnetic field,[18] and in impurity transitions.[16]

The many references cited above consider the dynamic Stark effect for the case of stationary double optical resonance. In this case, to observe the effects of rearrangement of the spectrum, it is necessary that the light intensity $J_\omega \gtrsim 10^8$ W/cm$^2$. Sufficiently long light pulses of such power, even in the absence of a single-photon resonance between the ground state and excited states of the electronic subsystem, give rise to an appreciable number of free carriers, including—as a result of multiphoton absorption—ionization of impurities, etc. Since this complicates the actual physical picture of double optical resonance, it makes sense to investigate the dynamic Stark effect under nonstationary conditions in which the pumping $\omega$ and weak probing light $\Omega$ appear in the form of picosecond or shorter pulses. In this case, when speaking of "weak" light (the probing beam $\Omega$), we have in mind the smallness of the "area" under a pulse of such light. Instead of the spectral picture of the dynamic Stark effect characteristic of stationary double optical resonance, the transient approach provides a time sweep with distinguishing features corresponding to this or that type of band spectrum in the field of the strong electromagnetic wave.

## 2. THE DENSITY MATRIX OF A THREE-LEVEL SYSTEM DURING A TRANSIENT DOUBLE OPTICAL RESONANCE

Let us consider an intrinsic semiconductor (or dielectric) in the field of strong pump radiation with frequency $\omega$, which is small in comparison with $E_g$ (or with $\Omega_{cn',cn}$—the spacing between the levels of the dimensional quantization in the conduction band $c$ and the valence band $v$ in quantum-confined systems), but falls at the resonance between states of two conduction bands or two valence bands. For definiteness, in what follows we will speak of conduction bands $|1\rangle$ and $|2\rangle$. For the sake of brevity, we will drop for now the subscript $\mathbf{k}$ (the wave vector), which characterizes the states in the band. The field $\omega$ is assumed to be quasistationary. This means here that the duration $\tau_\omega$ of a pulse of light $\omega$ exceeds the relaxation time $\tau_m$ of a pulse of carriers in the band, i.e., $\tau_\omega \gtrsim 10^{-12}$ s. Two ultrashort ($\tau_\Omega \sim 10$–$100$ fs) pulses of weak radiation with frequency $\Omega$, resonant with the transition between the ground state of the crystal $|0\rangle$ (the vacuum of the electron–hole pairs) and the $|1\rangle$ state, are incident upon the crystal in succession, with delay time $\tau_D$.

Let light with frequency $\Omega$ propagate in the medium in the direction of the $z$ axis between its front ($z=0$) and back ($z=L$) surfaces. We denote by $E_\Omega^{(i)}$ the electric field strength of the light wave $\Omega$ of the $i$th pulse ($i=1, 2$) in the medium near $z=0$. We assume that the change in $E_\Omega^{(i)}$ due to passage through the medium, $\Delta E_\Omega^{(i)}$, is small in comparison with $E_\Omega^{(i)}$. Then the absorptivity per unit area is given by[19]

$$\Delta J_i = \Omega L E_\Omega^{(i)} P_s \quad [z=L, t-t_1(L)], \tag{1}$$

where $P_s(z,t)$ is the amplitude of the reactive component of the polarization of the medium, and $t_1(z)$ is the time at which the field begins to act on the medium at the point $z$.

To calculate $P_s$ we will make use of the density matrix formalism in the approximation which represents the electronic system of the crystal in the form of a set of noninteracting three-level systems, each of which is characterized by its own wave vector $\mathbf{k}$ and the corresponding energy $\varepsilon_n(\mathbf{k})$. Obviously, this is a very rough approximation, but the factors which detract from its faithfulness turn out in the situation under consideration not to be very important. These factors include the uncertainty in the quasimomentum of the electron–hole pairs which arise in an interband transition induced by an ultrashort light pulse: $\Delta k \sim (c\tau_\Omega)^{-1}$. However, taking account of the actual indirect transitions in the given case leads to the same results as are obtained for a system with direct transitions connecting states with the same $k$ to different bands (see, for example, Ref. 20). At the same time, the adopted model allows for spreading of a packet of states with uncertainty $\Delta k$ in the quasimomentum (and this plays a substantial role here). In the case in which generation takes place due to the weak light $\Omega$, the effects of filling of the bands and of the Coulomb interaction[1)] are also negligible. Outside of the adopted approximation, the solution of the problem would have required the successive consideration of the relaxation of the photoexcited carriers. However, as will become clear in what follows, the dynamics of the processes under consideration here is usually determined not so much by relaxation as by the peculiarities of the electronic spectra which are being rearranged in the field of the wave $\omega$, and also by the aforementioned spreading of states in the continuous spectrum. Therefore we will limit ourselves to a description of relaxation with the help of the phenomenologically introduced decay parameter $\gamma$ (the reciprocal transverse relaxation time). The applicability of a two-level model to interband nonlinearities with allowance for saturation and relaxation effects is discussed in detail in Ref. 21.

We denote by $\tilde{\rho}_{m,n}$ the elements of the density matrix of a three-level system in the field of the light waves $\Omega$ and $\omega$. In place of the matrix elements $\tilde{\rho}_{10}$ and $\tilde{\rho}_{20}$ it is convenient to introduce the quantities

$$\rho_{10} = \tilde{\rho}_{10} \exp[-i(\Omega - \kappa_\Omega z)],$$

$$\rho_{20} = \tilde{\rho}_{20} \exp[-i(\Omega+\omega)t - i(\kappa_\Omega + \kappa_\omega)z], \tag{2}$$

where $\kappa_{\Omega,\omega}$ are the wave vectors of the light waves.

Assuming that the interaction with the light $\Omega$ connects only the states $|0\rangle$ and $|1\rangle$, we represent the polarization in the form

$$P_s = -(N_0/V_0) \cdot \mathrm{Im}(d_{01}\rho_{10}), \tag{3}$$

where $d_{mn}$ are the matrix elements of the dipole moment operator, and $N_0/V_0$ is the number of three-level systems per unit volume.[2)]

We introduce the notation

$$a_{10}=\Omega_{10}-\Omega-i\gamma\equiv\Delta_{10}-i\gamma,$$

$$a_{20}=\Omega_{20}-\Omega-\omega-i\gamma\equiv\Delta_{20}-i\gamma,$$

$$V_{mn}^{\Omega(\omega)}=\hbar^{-1}d_{mn}E_{\Omega(\omega)},\quad \omega_1^2=|V_{21}^{\omega}|^2. \tag{4}$$

Discarding terms oscillating with frequencies $2\Omega$ and $2\omega$, we obtain

$$i\dot{\rho}_{10}=a_{10}\rho_{10}+V_{12}^{\omega}\rho_{20}+V_{10},\quad i\dot{\rho}_{20}=a_{20}\rho_{20}+V_{21}^{\omega}\rho_{10}, \tag{5}$$

where $V_{10}^{\Omega}$ depends on time while $V_{21}^{\omega}$ can be taken as constant in the time interval of interest. We consider only processes associated with the induced polarization, which is linear in $V_{10}^{\Omega}$. Variation of the populations in the electronic subsystem is described by the terms of higher order in $V_{10}^{\Omega}$ and is not taken into account in Eqs. (5).

The system (5) reduces to a single equation:

$$i\ddot{\rho}_{10}+i(a_{10}+a_{20})\dot{\rho}_{10}+(\omega_1^2-a_{10}a_{20})\rho_{10}=a_{20}V_{10}^{\Omega}-i\dot{V}_{10}^{\Omega}. \tag{6}$$

As the initial condition we take $\rho_{10}(t=0)=0$. We represent the solution of Eq. (6) in the form

$$\rho_{10}=-i\int_0^t dt'\,V_{10}^{\Omega}(t')\Phi\{(a_{20}+\lambda_{1,2})\exp[i\lambda_{1,2}(t-t')]\}, \tag{7}$$

where

$$\lambda_{1,2}=\tfrac{1}{2}[-(a_{10}+a_{20})\pm\sqrt{(a_{10}-a_{20})^2+4\omega_1^2}]. \tag{8}$$

Here and below we use the notation

$$\Phi\{f(\lambda_{1,2})\}\equiv(\lambda_1-\lambda_2)[f(\lambda_1)-f(\lambda_2)]. \tag{9}$$

The quantities $\lambda_{1,2}$ describe the energy spectrum of the excited states of the electronic system in the optical Stark effect.

## 3. ABSORPTION OF WEAK LIGHT

We will calculate the energy $\Delta J_1$ absorbed from the first pulse of light $\Omega$ per unit area of beam cross section. We assume that $\tau_\Omega\gamma\ll 1$. We introduce the notation

$$V_{10}^{\Omega}(t)\equiv V_{10,0}^{\Omega}\varphi(t). \tag{10}$$

In what follows we assume that the light pulse $\Omega$ has a Gaussian envelope:

$$\varphi(t)=A\exp[-(t-t_0)^2/\sigma]. \tag{11}$$

The normalizing factor $A$ can be conveniently expressed in terms of the energies $\mathscr{E}_i$ $(i=1,2)$ per unit area of transverse beam cross section transported into the medium by each of the two pulses $\Omega$:

$$A^2|E_\Omega^{(i)}|^2=\mathscr{E}_i\frac{8}{c}\left(\frac{2\pi}{\varepsilon\sigma}\right)^{1/2}. \tag{12}$$

The quantity $\varepsilon$ in Eq. (12) differs from the true dielectric constant $\varepsilon(\Omega)$ by the fact that it does not include the contribution of the transitions, explicitly considered here, in the isolated two- and three-level systems.

To calculate the energy absorbed during the pulse, we substitute Eqs. (7)–(12) into Eqs. (3) and (1) and integrate over time from zero to the end of the pulse. We obtain

$$\Delta J_1^{(0)}=-(N_0/V_0)\theta_{11}\Phi\{F(\lambda_{1,2})\}, \tag{13}$$

$$\theta_{pq}=\frac{\Omega L}{\hbar c}|d_{10}|^2\left(\frac{2\pi\sigma}{\varepsilon}\mathscr{E}_p\mathscr{E}_q\right)^{1/2}, \tag{14}$$

$$F(\lambda,\tau)=(a_{20}+\lambda)\exp(-\sigma\lambda^2/2+i\tau\lambda). \tag{15}$$

In the absence of pumping

$$\Delta J_1^{(0)}(\omega=0)=-(N_0/V_0)\theta_{11}\exp(-\sigma\Delta^2/2). \tag{16}$$

It is clear that for fixed energy in the short light pulse the energy absorbed by the two- or three-level systems is proportional to the duration of the pulse $(\sim\sigma^{1/2})$.

Let us now calculate the energy absorbed from the second light pulse $\Omega$, following the first after the time delay $\tau_D$. For simplicity, we take the shape of both light pulses $\Omega$ to be identical. We have

$$\Delta J_1^{(0)}=\Delta J_{21}^{(0)}+\Delta J_{22}^{(0)}, \tag{17}$$

where $\Delta J_{22}^{(0)}$ differs from the quantity $\Delta J_1^{(0)}$, given by formula (13), only by the substitution $\theta_{11}\rightarrow\theta_{22}$. The term $\Delta J_{21}^{(0)}$ on the right-hand side of formula (17) describes the absorption from the second pulse, due to the polarization induced by the first pulse:

$$\Delta J_{21}^{(0)}=-2(N_0/V_0)\theta_{21}\Phi\{F(\lambda_{1,2},\tau_D)\}. \tag{18}$$

In the absence of pumping Eq. (18) reduces to

$$\Delta J_{21}^{(0)}=-2\left(\frac{N_0}{V_0}\right)\theta_{21}^{(0)}\frac{\exp(-\gamma\tau_D)\cos(\Delta_{10}\tau_D+\varphi d/2)}{(1+\zeta^{-2})^{d/4}(\tau_D\nu)^{d/2}}, \tag{19}$$

$$\varphi=\operatorname{arctg}\zeta,\quad \zeta=\tau_D/(\sigma\Delta_{10}). \tag{20}$$

It is useful to consider the absorption of two ultrashort light pulses $\Omega$ on the interband transitions in bulk materials, and also in systems with quantum wells and wires, starting from the case in which pumping $\omega$ is absent. In this case we have to deal with a coherent, linear optical process in which absorption of the light pulse is modulated as a result of the polarization induced by the preceding pulse. In the parabolic bands model we have for this case

$$\lambda_1=\nu k^2-\Delta,\quad \lambda_2=0,\quad \nu=\hbar(2m_r)^{-1},$$

$$m_r^{-1}=m_c^{-1}+m_v^{-1},\quad \Delta=\Omega-E_g/\hbar\quad (d=3),$$

$$\Delta=\Omega-\Omega_{cn',vn}\quad (d=1,2), \tag{21}$$

where $m_c$ and $m_v$ are the effective masses of the electron in the conduction band $c$ and the valence band $v$, $k^2$ is the square of the three-, two-, and one-dimensional wave vector for systems with dimensionality $d=3$, 2, 1, respectively, and $\Omega_{cn',cn}$ are the frequencies of the transitions between the levels with dimensional quantization in the conduction band and in the valence band.

We now integrate over **k**-space. For the case $\sigma\Delta^2/4 \ll 1$, i.e., for short pulses and a modest offset of the frequency $\Omega$ from the absorption band edge, we find that the energy absorbed from the first light pulse $\Omega$ is

$$\Delta J_1^{(d)} \approx \alpha_d \kappa_d \theta_{11} (\nu\sqrt{\sigma})^{-d/2} \propto \tau_\Omega^{(2-d)/2}, \tag{22}$$

where

$$\kappa_0 = N_0/V_0, \quad \kappa_1 = N_1/S, \quad \kappa_2 = N_2/L_1, \tag{23}$$

$$\alpha_0 = 1, \quad \alpha_1 = 2^{-11/4}\pi/\Gamma(3/4) \approx 0.381,$$

$$\alpha_2 = 2^{-5/2}\sqrt{\pi} \approx 0.313,$$

$$\alpha_3 = 2^{1/4}/\Gamma(3/4) \approx 0.970, \tag{24}$$

$\Gamma(x)$ is the gamma function, $N_1/S$ is the number of quantum wires per unit area of cross section perpendicular to them, and $N_2/L_1$ is the number of quantum wells per unit length in the direction perpendicular to their plane.

It is clear that for $\sigma\Delta^2/4 \ll 1$, the dependence on the pulse duration $\tau_\Omega$ of the energy absorbed from the first light pulse in an interband transition is determined largely by the dimensionality of the system.

Let us carry out a rough numerical estimate of the absorption of a light pulse $\Omega$. We assume band structure parameters corresponding to GaAs: $E_g \approx 1.43$ eV, $\Delta_{s0} \approx 0.33$ eV, $m_c \approx 0.05 m_0$, the matrix element of the optical interband transition $Z_{cv} \approx 0.5 \cdot 10^{-7}$ cm. Then

$$\left.\begin{matrix} \Delta J_1^{(0)} \\ \Delta J_1^{(1)} \\ \Delta J_1^{(2)} \\ \Delta J_1^{(3)} \end{matrix}\right\} \times J_1^{-1} \approx \left.\begin{matrix} 0.9 \cdot 10^{-19}\ (N_0/V_0) \\ 0.35 \cdot 10^{-13}\ (N_1/S) \\ 0.3 \cdot 10^{-7}\ (N_2/L_1) \\ 0.95 \cdot 10^{-1} \end{matrix}\right\} \times \frac{L}{1\ \mu\text{m}}.$$

If we take the linear distance between the quantum-dimensional objects to be 200 Å, then we have

$$N_2/L_1 = 0.5 \cdot 10^6\ \text{cm}^{-1},$$

$$N_1/S = 2.5 \cdot 10^{11}\ \text{cm}^{-2},$$

$$N_0/V_0 = 1.25 \cdot 10^{17}\ \text{cm}^{-3}.$$

These estimates show that in order to observe the above effects, the light should propagate distances of several microns in bulk materials, and tens of microns in materials with quantum wells, wires, and dots. Therefore, for $d=1$ and $d=2$, the light should obviously propagate in the direction of the wire or parallel to the planes of the quantum wells.

Let us consider one more case, with a large offset from the absorption edge: $\sigma\Delta^2/4 \gg 1$. Then

$$\Delta J_1^{(d)} \approx \bar{\alpha}_d \kappa_d \theta_{11} (\Delta\sqrt{\sigma})^{-1} (\Delta/\nu)^{d/2}, \tag{25}$$

where

$$\bar{\alpha}_1 = \sqrt{2\pi}/4, \quad \bar{\alpha}_2 = \sqrt{2\pi}/4, \quad \bar{\alpha}_3 = 1/\sqrt{8\pi}. \tag{26}$$

For the energy absorbed from the second pulse, for delay times $\tau_D \gtrsim \sigma\Delta$ we have

$$\Delta J_2^{(d)}(\omega_1=0) = \Delta J_{22}^{(d)} (1 + \chi_0^{(d)}), \tag{27}$$

where $\Delta J_{22}^{(d)}$ is given by the right-hand side of Eq. (22) with the substitution $\theta_{11} \to \theta_{22}$,

$$\chi_0^{(d)} = \beta_d \left(\frac{\mathscr{E}_1}{\mathscr{E}_2}\right)^{1/2} \left[\frac{\sigma}{\tau_D^2(1+\xi^{-2})}\right]^{d/4} \times \exp\left(-\frac{\sigma\Delta^2}{4} - \gamma\tau_D\right) \cos\left(\Delta\tau_D + \frac{\varphi d}{2}\right), \tag{28}$$

where

$$\beta_0 = 2, \quad \beta_1 = \pi^{-1/2} \cdot 2^{5/4}\Gamma(3/4) \approx 1.16,$$

$$\beta_2 = 2^{3/2}\pi^{-1/2} \approx 1.596,$$

$$\beta_3 = 2^{-5/4}\pi^{-1/2}\Gamma(3/4) \approx 0.344. \tag{29}$$

The time-dependent factor in brackets on the right-hand side of Eq. (28) is associated with the spreading in the continuous spectrum of a packet containing states with different energies and momenta created by a light pulse of finite duration ($\sim \sqrt{\sigma}$). For small $\sigma$ this factor reduces to $(\sqrt{\sigma}/\tau_D)^{d/2}$, and for $\sigma \gg \tau_D/\Delta$ it ceases to depend on $\tau_D$.

From Eqs. (27) and (28), it is obvious that coherent modulation of the second light pulse $\Omega$ can be observed at delay times $\tau_D$ such that $\sqrt{\sigma} < \tau_D \lesssim \gamma^{-1}$. If the pulse duration $\tau_\Omega$ is $\sim 30$ fs, then, as follows from Eqs. (27) and (28), it is possible to observe the effect by varying $\tau_D$ within these limits while detuning the resonance by $\Delta \lesssim 2/\sqrt{\sigma}$ (i.e., $\Delta \lesssim 30$–$40$ meV). All the same, if the detuning $\Delta$ is less than the frequency of the optical phonon $\Omega_0$, then in pure materials at low temperatures the relaxation time $\gamma^{-1}$ can exceed the above value by 1–2 orders of magnitude.

## 4. NONSTATIONARY DOUBLE OPTICAL RESONANCE IN INTERBAND TRANSITIONS

We introduce in addition to (4) the notation

$$\Delta' = \Omega + \omega - E_g'/\hbar \quad (d=3),$$

$$\Delta' = \Omega + \omega - \Omega_{c'n',vn} \quad (d=1,2),$$

$$\nu' = \hbar/(2\mu'), \quad \mu'^{-1} = m_{c'}^{-1} + m_v^{-1},$$

$$\nu_\pm = \nu \pm \nu', \quad \delta_\pm = \Delta \pm \Delta', \quad \rho_- = \nu\Delta' - \nu'\Delta. \tag{30}$$

The quantities marked with a prime in definitions (30) belong to the conduction band $c'$, which is mixed with the $c$ band by the intense light $\omega$. In this new notation

$$a_{20} = \nu' k^2 - \Delta', \tag{31}$$

$$\lambda_{1,2}(k) = \tfrac{1}{2}[-\nu_+ k^2 + \delta_+^2 \pm \sqrt{(\nu_- k^2 - \delta_-)^2 + 4\omega_1^2}]. \tag{32}$$

So for the case of a $d$-dimensional system ($d=1,2,3$), according to Eqs. (15) and (18), the integral

$$W_d = \int_0^\infty dk (k^2)^{(d-1)/2} \Phi\{F[\lambda_{1,2}(k), \tau_D]\} \tag{33}$$

enters into the expression for $\Delta J_{21}^{(d)}$ [see Eqs. (9) and (15)].

In order to obtain an approximate value for $W_d$, we will use the method of steepest descent. Its application here requires a certain amount of care, especially in the case of identical signs of $\nu$ and $\nu'$ [see definitions (21) and (30)]. The integration contour in the complex plane passing through the saddle points in the right ($r$) and left ($l$) half-planes

$$k_{S(r,l)}=k'_{S(r,l)}+ik''_{S(r,l)},$$

$$k'_{S(r)}=-k'_{S(l)}=(2\nu_-)^{-1/2}(\chi+\delta_-)^{1/2},$$

$$k''_{S(r)}=-k''_{S(l)}=-(2\nu_-)^{-1/2}(\chi-\delta_-)^{1/2},$$

$$\chi=\left(\delta_-^2+\frac{\omega_1^2\nu_-^2}{\nu\nu'}\right)^{1/2} \tag{34}$$

should bypass the branch points associated with the presence of the root in Eqs. (32), (33).

Omitting the details of the calculations, we simply present the expression for the energy absorbed from the second light pulse $\Omega$ thanks to the polarization induced by the first light pulse:

$$\Delta J_2^{(d)}=\Delta\tilde{J}_{22}^{(d)}(1+\tilde{\chi}^{(d)}), \tag{35}$$

where $\Delta J_{22}^{(d)}$, the energy absorbed from the second light pulse $\Omega$ in the absence of the first light pulse, is given by the right-hand side of Eq. (22) with the substitution $\theta_{11}\rightarrow\theta_{22}$,

$$\tilde{\chi}^{(d)}\approx\left(\frac{\mathscr{E}_1}{\mathscr{E}_2}\right)^{1/2}\left(\frac{\omega_1\sigma}{\tau_D}\right)^{1/2}Q_D(\nu,\nu',\Delta,\Delta')$$
$$\times\exp\left[-\frac{(\rho_-^2-4\omega_1^2\nu\nu')\sigma}{2\nu_-^2}-\tau_D\left(\gamma+\frac{\omega_1\sqrt{\nu\nu'}}{\nu_-}\right)\right]$$
$$\times\cos\left[\left(\tau_D-\frac{2\sigma\omega_1\sqrt{\nu\nu'}}{\nu_-}\right)\frac{\rho_-}{\nu_-}+\varphi_d\right], \tag{36}$$

$$Q_d=\beta_d\left(\frac{\nu'}{\nu}\right)^{1/4}\left(\frac{\nu}{\nu_-}\right)^{(d-1)/2}\left(\delta_-^2+\frac{\omega_1^2\nu_+^2}{\nu\nu'}\right)^{(d-2)/4}, \tag{37}$$

$$\beta_1=4\sqrt{2},\quad \beta_2=\sqrt{2}/\pi,\quad \beta_3=\sqrt{2}/\pi. \tag{38}$$

The phase $\varphi_d$ depends weakly on the delay time $\tau_d$. The quantity $\rho_-$ in Eq. (36) vanishes at the double resonance when there is a point $\mathbf{k}_R$ in $\mathbf{k}$-space at which

$$\hbar^{-1}E_g+\nu k_R^2=\Omega,\quad \hbar^{-1}E'_g+\nu' k_R^2=\Omega+\omega \tag{39}$$

simultaneously.

It is clear from Eq. (36) that the described effect takes place only in a small neighborhood ($\sim\hbar/\sqrt{\sigma}$) of the double resonance point, while in the absence of a pump field $\omega$, the first of the light pulses $\Omega$ has a marked influence on the absorption of the second pulse only at small offsets $\Delta$ of the frequency $\Omega$ from the absorption edge.

Let us turn our attention to the fact that formula (36) is valid only when the effective masses in the $c$ and $c'$ bands are somewhat different, such that $\nu_- > 2\omega_1\sigma\sqrt{\nu\nu'}/\tau_D$. The case in which $m_c$ and $m_{c'}$ are identical is considered in the following section. Furthermore, Eq. (36) does not describe the situation in which the double resonance is realized extremely close ($\Delta\lesssim\sigma^{1/2}$) to the absorption edge. The calculation for this case leads to extraordinarily cumbersome expressions. The case of identical signs of the reduced masses ($\nu,\nu'>0$) corresponds, as was shown in Refs. 2 and 3, to the presence of a break in the band energy spectrum, which is altered by the interaction with the pump radiation field $\omega$. The dispersion law for the two branches of the spectrum (see Fig. 1a) is given by Eq. (32). It follows from Eq. (36) that in such a situation interference of states in the two branches of the spectrum leads to an exponential decay of the induced polarization (with time constant $\nu_-/\sqrt{\nu\nu'}\omega_1\propto E_\omega^{-1}$), which is modulated by oscillations with a frequency that vanishes when the double optical resonance conditions are fulfilled exactly.

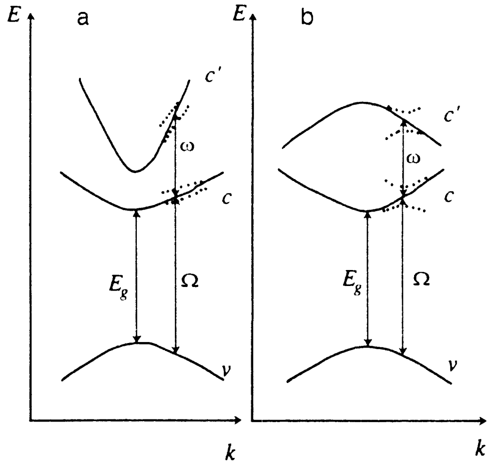

FIG. 1. Band diagram of a crystal at double interband optical resonance: a) same, b) different signs of the effective masses in the $c$ and $c'$ bands. The dotted curves represent the bands in the presence of intense pumping $\omega$.

Consideration of the problem in terms of the dynamic Stark effect is adequate at high enough pump intensities, when $\omega_1\gtrsim\gamma$. In this case, as is clear from Eq. (36), field-induced decay of the polarization dominates over the usual relaxation as a result of collisions with phonons, electron–electron collisions, or scattering from defects. If the light $\omega$ mixes two different conduction bands, then the typical value of the matrix element $d_{12}\sim10^{-18}$ cgs units. To obtain the Rabi frequency $\omega_1\sim10^{13}$ s$^{-1}$ requires the light intensities $J_\omega\sim10^9$ W/cm$^2$. If the light $\omega$ propagates in a direct-band material with quantum wells of width $l$, then

$$\langle cn|\hat{d}|cn'\rangle=-\frac{4l}{\pi^2}e_z[1-(-1)^{n+n'}]nn'(n^2-n'^2)^{-2}.$$

In this case, for $l\sim100$ Å, $n=1$, $n'=2$, the matrix element $d_{12}$ takes values of the order of $\sim10^{-17}$ cgs units. Accordingly, to obtain $\omega_1\sim10^{13}$ s$^{-1}$ here requires intensities two orders smaller: $J_\omega\sim10^7$ W/cm$^2$ (see also Sec. 5).

For different signs of $\nu$ and $\nu'$ in the band spectrum renormalized by the interaction with the field $\omega$, there appears now not a break, but a gap (see Fig. 1b). Under stationary conditions when the frequency of the probing light $\Omega$ falls within the gap, absorption disappears (at least, if relaxation $\gamma$ is neglected), and against the background of the fundamental absorption band there appears a spectral region in which the medium is transparent.[2,3] For the case of a pulsed probe, we obtain (35), where now

$$\tilde{\chi}^{(d)}=\left(\frac{\mathscr{E}_1}{\mathscr{E}_2}\right)^{1/2}\left(\frac{\omega_1\sigma}{\tau_D}\right)^{1/2} Q'_d(\nu,\nu')\sum_{p=1}^{2}\Lambda_p^{(d-2)/2}$$

$$\times\exp\left[\frac{(-1)^p 2\omega_1\rho_-\sigma\sqrt{-\nu\nu'}}{\nu_-^2}-\gamma\tau_D\right]$$

$$\times\cos(\Omega_p\tau_D+\varphi'_d), \tag{40}$$

$$Q'_d(\nu,\nu')=\beta_d\left(-\frac{\nu'}{\nu}\right)^{1/4}\left(\frac{\nu}{\nu_-}\right)^{(d-1)/2}, \tag{41}$$

$$\Lambda_{1,2}=\frac{\delta_-\pm\omega_1\nu_+/\sqrt{-\nu\nu'}}{\Delta},\quad \Omega_{1,2}=\frac{\rho_-\pm 2\omega_1\sqrt{-\nu\nu'}}{\nu_-}. \tag{42}$$

It should be kept in mind that now $-\nu\nu'>0$, $\nu_-=\nu+|\nu'|$, $\Delta'<0$, and $\delta=\Delta+|\Delta'|$. The phase $\varphi'_d$, as before, depends weakly on $\tau_d$.

From Eq. (40) it is clear that field-induced decay is absent at $\rho_-=0$, and the induced absorption oscillates with frequency $2\omega_1\sqrt{|\nu\nu'|}/(\nu+|\nu'|)$, proportional to the field strength of the pump wave. Decay of the oscillations in the actual range of values of the parameters is determined to an equal degree by two factors: the "relaxation" exponential $\exp(-\gamma\tau_d)$ and the pre-exponential factor $(\omega_1\sigma/\tau_d)^{1/2}$ associated with the spreading of the packet of states created by the first light pulse $\Omega$.

## 5. TRANSIENT DOUBLE OPTICAL RESONANCE FOR IDENTICAL EFFECTIVE MASSES OF CARRIERS IN EXCITED STATES

Let us return to the case of identical signs of the reduced masses in the $c$ and $c'$ bands, mixed by the intense light field $\omega$. In Section 4 we already noted that Eq. (36) is valid only for somewhat different values of the effective masses in the $c$ and $c'$ bands and $\nu_-$ correspondingly not too small. However, when the roles of the $c$ and $c'$ bands (states $|1\rangle$ and $|2\rangle$) are played by states in quantum wells or quantum wires, obtained as a result of dimensional quantization of one nondegenerate band (e.g., $\Gamma_6$ in $A_3B_5$ materials), but the effective masses in layers forming quantum wells and barriers differ only slightly, then the values of $\nu_-$ turn out to correspondingly close to zero. Since the situation at small $\nu$ differs only slightly from that at $\nu_-=0$, we limit ourselves to the latter case. The calculations for $\nu_-=0$ are significantly simpler than for different $\nu$ and $\nu'$. As a result, for one- and two-dimensional materials we obtain

$$\tilde{\chi}^{(d)}_{\nu_-=0}=\left(\frac{\mathscr{E}_1}{\mathscr{E}_2}\right)^{1/2}\kappa^{(d)}\sum_{p=1}^{2}\left\{\left[\frac{\tau_d^2}{\sigma}+(\delta_++q_p)^2\sigma\right]^{-d/4}\right.$$

$$\times\left(1+\frac{\delta_+}{q_p}\right)^{d-1}\frac{(\delta_-+q_p)}{|q_p|}$$

$$\times\exp\left[\frac{-(\delta_++q_p)^2\sigma}{8}-\gamma\tau_D\right]$$

$$\left.\times\cos\left[\frac{(\delta_++q_p)\tau_D}{2}+\varphi_p^{(d)}\right]\right\}, \tag{43}$$

$$q_{1,2}=\sqrt{\delta_-^2+4\omega_1^2},$$

$$\varphi_p^{(d)}=\tfrac{1}{2}(-1)^d\operatorname{arctg}\frac{\tau_D}{\sigma(\delta_++q_p)},$$

$$\kappa^{(1)}=0.29,\quad \kappa^{(2)}=-1.13. \tag{44}$$

For exact resonance at the absorption edge and for long enough delay times $\tau_D\gg 2\omega_1\sigma$, (43) takes an especially simple form:

$$\tilde{\chi}^{(d)}_{\nu_-,\Delta,\Delta'=0}=2\kappa^{(d)}(\mathscr{E}_1/\mathscr{E}_2)^{1/2}(\sqrt{\sigma}/\tau_D)^{d/2}$$

$$\times\exp(-\tau_D\gamma)\cos(\omega_1\tau_D). \tag{45}$$

Clearly, even in the case of "parallel" bands, decay of oscillations is determined by both the time $\gamma^{-1}$ which characterizes (in some sense) homogeneous broadening of dimensional quantization levels and by the power-law factor $(\sqrt{\sigma}/\tau_D)^{d/2}$ associated with coherence spreading in the continuous spectrum.

[1] We do not deal here with excitonic effects. The optical Stark effect in excitons during transient double optical resonance will be considered separately.

[2] Equations (3)–(23) can be tied directly to the case of 0-dimensional systems (quantum dots) or to the case of impurity centers, described by the three-dimensional model.

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