

OPTICAL PROPERTIES OF SEMICOMDUCTOR IN EXCITON RANGE OF SPECTRUM IN PRESENCE OF STRONG PUMP PULSE IN M-BAND OF LUMINESCENCE

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Abstract

The behavior of the semiconductor dielectric susceptibility in the exciton range of spectrum due to the interaction with photons of a weak test pulse under the stationary action of a strong pump pulse in the range of M-band of luminescence is investigated. It is shown that the well pronounced Autler–Townes effect occurs at the exciton resonance. The position of the absorption peaks is determined by the amplitude and frequency of the pump pulse.

1.Introduction

The optical Stark–effect in semiconductors in the exciton range of spectrum is one of the brightest nonlinear optical effects [1-3]. This effect is manifested by the spectral shift of the exciton and biexciton levels under the action of a strong ultrashort pulse of resonant laser radiation and by it returning to the initial position after finishing of the pulse action. The interpretation of this effect, based on the idea of Bose–Einstein condensation of excitons induced by an external coherent laser radiation, was proposed in [2,3]. The absorption and gain of a weak test signal in the presence of Bose–condensed excitons that occur in the nonequilibrium conditions due to the field of coherent laser radiation were investigated in [1,4,5]. It was shown that instabilities occur due to the real emergence of two laser photons and their conversion into two noncondensed particles, which affect essentially the test signal absorption. In connection with the experimental investigations of the optical properties of semiconductors in the exciton range of spectrum at high level of excitation the pump–probe approach has acquired a special significance. This approach is based on the use of two beams of laser radiation, namely the strong pump and the weak test beams. The weak beam tests the changes of the optical properties of crystal, which occurred due to the action of the strong pump beam. This approach was used for the experimental investigation of the radiative recombination and nonlinear response of high density system of excitons and biexcitons [6–8], the red and blue shifts of the exciton resonance in the condition of picosecond pumping [9–12] and the analogy of the Autler–Townes effect on the biexcitons in CuCl [13]. The theory of this effect was proposed in [14, 15]. The well pronounced splitting of the biexciton absorption band in CuCl into two separated lines at high level of excitation was observed experimentally [13]. Testing of the biexciton state was carried out by use of two–photon absorption with the excitation of biexcitons from the ground state of the crystal in the presence of the strong pump pulse tuned to the M–band of luminescence. From the obtained value of the splitting the authors of [13] determined the value of the relevant dipole momentum in the range of M–band of luminescence due to the optical exciton–biexciton conversion. The idea about the observation of this splitting was firstly proposed in [16].

The different aspects of the pump–probe approach for the high density system of excitons and biexcitons were considered in [1, 4, 5, 14–18]. It was shown in [18] that the susceptibilities of semiconductors in the exciton range of spectrum taking into account the elastic exciton–exciton interaction undergo the bistable behavior depending on the frequency and intensity of the pump pulse and on the frequency of a test pulse too. The dielectric susceptibilities of the CuCl–crystal were investigated in [14, 15] for the case when the strong pump pulse acted in the range of M–band of luminescence and the testing was carried out by two–photon absorption followed by the generation of biexcitons.

In this connection the questions arises about the possibility of testing of crystals like CuCl in the exciton range of spectrum using the simple mechanism of one–photon generation of excitons from the ground state of the crystal, keeping the pump in the M–band. In this case the model of the energy spectrum of semiconductor with the exciton and biexciton levels is substantially the nonequidistant one (Fig. 1). The photons of the pump pulse can proceed only the optical exciton–biexciton conversion and cannot take place in the test procedure because of a great detuning at the excitonic self–frequency.

2. Statement of the problem and main equations

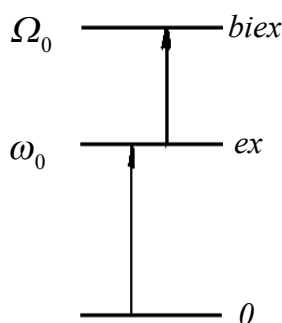


Fig.1. Schematic three–level diagram of the exciton–biexciton system. The exciton and biexciton states are coherently mixed by the resonant pump field and the resulting dressed states are tested by the one photon exciton absorption.

Let the strong monochromatic wave (pump pulse) of coherent laser radiation with the amplitude E_0 and frequency $\omega_l \approx \Omega_0 - \omega_0$ tuned to the transition between the exciton and biexciton states and the weak wave (test pulse) with the amplitude E and frequency $\omega \approx \omega_0$ incident on the semiconductor like CuCl. Here the Ω_0 and ω_0 are the self–frequencies of exciton and biexciton states respectively. As far as the pump pulse frequency is tuned to the transition between the exciton and biexciton states, the two excited states are mixed to create the dressed states as shown in Fig. 1.

This corresponds to the Autler–Townes effect. In the scheme of Fig. 1 the pump pulse is set below the exciton absorption band and we can observe the coherent effect in the steady state regime. The photons of pump pulse change the semiconductor energy spectrum essentially and the photons of the weak pulse probe these changes in the exciton range of spectrum.

The Hamiltonian of interaction of excitons and biexcitons with light in the resonant approximation we can write in the form

$$H = -\hbar g(E^+ a^+ e^{-i\omega t} + E^- a e^{i\omega t}) - \hbar \sigma(E_0^- a^+ b e^{i\omega_l t} + E_0^+ a b^+ e^{-i\omega_l t}), \quad (1)$$

where a and b are the exciton and biexciton polarization waves of the medium respectively, g is the constant of exciton–photon interaction, σ is the constant of optical exciton–

biexciton conversion [19], $E_0^+(E^+)$ and $E_0^-(E^-)$ are the positive and negative frequency components of the fields of pump (probe) pulse.

Assuming the exciton, biexciton and photon states to be macrofilled we can derive the following material equations for the amplitudes a and b :

$$i\dot{a} = (\omega_0 - i\gamma_1)a - gE^+e^{-i\omega t} - \sigma E_0^-be^{i\omega_l t}, \quad (2)$$

$$i\dot{b} = (\Omega_0 - i\gamma_2)b - \sigma E_0^+ae^{-i\omega_l t}, \quad (3)$$

where γ_1 and γ_2 are the phenomenological relaxation parameters, which take into account the damping of exciton and biexciton states respectively due to the scattering processes from the coherent states to the noncoherent ones.

We investigate the response of the system in all orders of the perturbation theory for the pump amplitude E_0 and the first order for the test amplitude E in the steady state. From (2) and (3) we can obtain the expression for the stationary amplitudes a and b , and then for the polarization of medium and finally we derive the following expression for the susceptibility χ of medium:

$$\chi = -\frac{\hbar g^2}{\Delta + i\gamma_1 - \frac{\sigma^2 E_0^2}{\Delta + \Delta_l + i\gamma_2}}, \quad (4)$$

where $\Delta = \omega - \omega_0$, $\Delta_l = \omega_l - \Omega_0 + \omega_0$ are the detunings.

In the limit of evanescent small values for the dampings ($\gamma_1 \rightarrow 0$, $\gamma_2 \rightarrow 0$) the susceptibility of medium is the real function of the form

$$\chi = -\frac{\hbar g^2 (\Delta + \Delta_l)}{\Delta(\Delta + \Delta_l) - \sigma^2 E_0^2}. \quad (5)$$

The susceptibility χ in this case diverges for two values of the resonance detuning Δ for the field of a weak pulse, which are determined by the expressions

$$\Delta_{\pm} = (-\Delta_l \pm \sqrt{\Delta_l^2 + 4\sigma^2 E_0^2})/2. \quad (6)$$

The detunings $\Delta_{\pm}$ determined the renormalized eigenfrequencies of the dressed exciton states $\omega_{\pm} = \omega_0 + \Delta_{\pm}$, which appear under the action of the strong pump pulse. The difference between these eigenfrequencies (Autler–Townes splitting) $\Omega = \omega_+ - \omega_- = \sqrt{\Delta_l^2 + 4\sigma^2 E_0^2}$ determine the frequency of optical nutation in the system of excitons and biexcitons in the range of M–band [19].

From (6) and Fig. 2 we can see, that the more the field amplitude E_0 of pump pulse and the more the absolute value of the pump detuning Δ_l , the more the Autler–Townes splitting. In the limit when the Rabi–frequency σE_0 is much more than the resonance detuning $|\Delta_l|$, the splitting increases with the increase of the amplitude E_0 . In the reverse limit, when $|\Delta_l| \gg \sigma E_0$, we obtain $\Delta_+ \rightarrow \sigma^2 E_0^2 / \Delta_l$, $\Delta_- \rightarrow -\Delta_l$ for $\Delta_l > 0$ and $\Delta_+ \rightarrow -\Delta_l$, $\Delta_- \rightarrow \sigma^2 E_0^2 / \Delta_l$ for $\Delta_l < 0$.

As we can see from (4), the susceptibility χ is the nonlinear function of the pump amplitude E_0 and depends on the frequencies ω_l and ω of the both pulses. We point out that the absorption band of the weak pulse, when $\gamma_1 \rightarrow 0$ and $\gamma_2 \rightarrow 0$, consists of two δ –shaped

peaks at the frequencies $\omega = \omega_+$ and $\omega = \omega_-$. Therefore we can interpret the curves in Fig. 2 as curves of the spectral position of the peaks of absorption bands of weak pulse depending on the detuning Δ_l and field amplitude E_0 of the pump pulse.

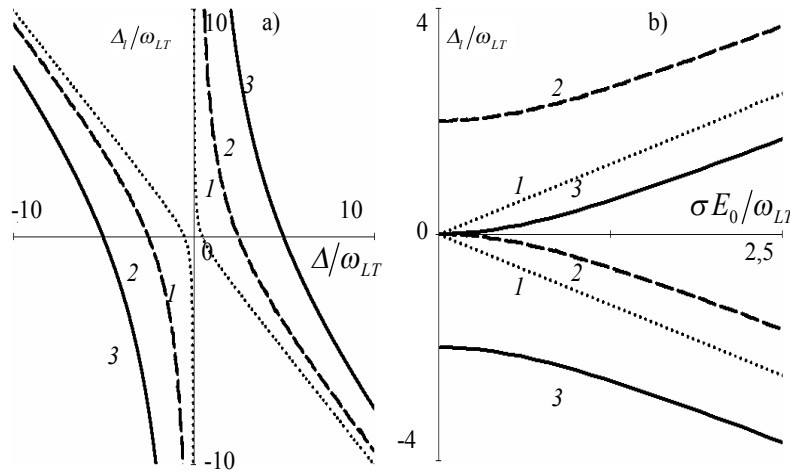


Fig. 2. The renormalized frequencies $\omega_{\pm} - \omega_0 = \Delta_{\pm}$ of the dressed exciton quasilevels depending a) on the pump field detuning Δ_l for the fixed values of the pump amplitudes $\sigma E_0 / \omega_{LT}$, which equals 0,5 (1), 2,5 (2), 5 (3) and b) on the normalized field amplitude $\sigma E_0 / \omega_{LT}$ for the different values of the pump detuning Δ_l / ω_{LT} , which equals 0 (1), -2 (2), 2 (3).

Using (5) we can obtain the dispersion law $\omega(k)$ for the photons of the test pulse in the presence of the strong pump pulse in the form

$$\frac{c^2 k^2}{\omega^2} = \varepsilon_{\infty} \left(1 - \omega_{LT} \left(\Delta - \frac{\sigma^2 E_0^2}{\Delta + \Delta_l} \right)^{-1} \right), \quad (7)$$

where ε_{∞} is the background dielectric constant and ω_{LT} is the longitudinal-transversal splitting in the exciton range of spectrum. All the significant peculiarities of the dispersion law depending on the pump amplitude E_0 take place in the vicinity of the exciton frequency (Fig. 3). The dispersion law

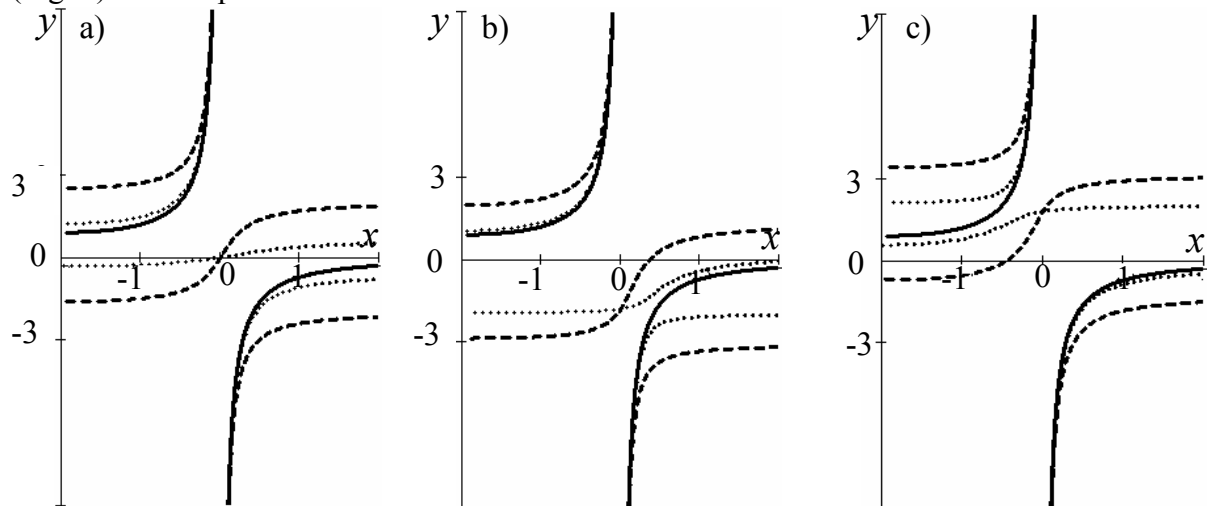


Fig. 3. Polariton-like dispersion curves for the photons of the weak pulse around the exciton state for the different values of the pump detuning Δ_l / ω_{LT} , which equals a) 0, b) 2 and c) -2 and for the different values of the normalized pump field $\sigma E_0 / \omega_{LT}$, which equals 0,5 (dotted lines) and 5 (dashed lines). The linear exciton-polaritons in the absence of the pump field are depicted by the solid lines. Here $x = ck / \sqrt{\varepsilon_{\infty}} \omega_{LT}$, $y = \sigma E_0 / \omega_{LT}$.

consists of three branches. The spectral region of the third branch is bounded by the frequencies ω_+ and ω_- and it becomes broader when the value of the field amplitude increases. The lower and upper dispersion branches approach the values of detunings Δ_+ and Δ_- at high wave vectors k . The branches of the dispersion law shift toward the longwave (shortwave) side with the increasing of $|\Delta_l|$ when $\Delta_l > 0$ ($\Delta_l < 0$) (Fig. 3).

Introducing the normalized variables $\delta = \Delta/\gamma_1$, $\delta_l = \Delta_l/\gamma_1$, $s = \gamma_2/\gamma_1$, $f_0 = \sigma E_0/\gamma_1$ and $\chi_0 = \hbar g^2/\gamma_1$ we obtain from (4) the following expressions for the dispersive (real) χ' and absorptive (imaginary) χ'' components of the susceptibility

$$\frac{\chi'}{\chi_0} = - \frac{\left(\delta - (\delta + \delta_l) \frac{f_0^2}{(\delta + \delta_l)^2 + s^2} \right)}{\left(\delta - (\delta + \delta_l) \frac{f_0^2}{(\delta + \delta_l)^2 + s^2} \right)^2 + \left(1 + s \frac{f_0^2}{(\delta + \delta_l)^2 + s^2} \right)^2}, \quad (8)$$

$$\frac{\chi''}{\chi_0} = \frac{\left(1 + s \frac{f_0^2}{(\delta + \delta_l)^2 + s^2} \right)}{\left(\delta - (\delta + \delta_l) \frac{f_0^2}{(\delta + \delta_l)^2 + s^2} \right)^2 + \left(1 + s \frac{f_0^2}{(\delta + \delta_l)^2 + s^2} \right)^2}. \quad (9)$$

3. Discussion

It is seen from (9), that the absorptive component χ'' is the positive defined quantity for any values of f_0 , δ and δ_l . This means that it is possible only the absorption of the weak signal. The gain effect is impossible.

It follows from (8) and (9), that the susceptibilities χ' and χ'' are the nonlinear functions of the pump amplitude and depend on the frequencies ω and ω_l of the both pulses. In the case of exact resonance between the frequencies of pulses and the self-frequencies of the corresponding transitions we obtain $\chi' = 0$ and

$$\chi''/\chi_0 = (1 + f_0^2/s)^{-1}. \quad (10)$$

It is clear, that the absorptive component of the susceptibility χ'' in the conditions of exact resonance quickly saturates, when the pump intensity increases. The absorption of the weak pulse at the frequency of the exciton transition is suppressed at high level of excitation by the pump pulse in M-band region.

Now we will discuss the behavior of the absorptive component of susceptibility (absorption band of the weak pulse in the exciton range of spectrum) depending on the resonance detuning δ in the conditions, when the photons of the pump pulse are in the exact resonance at the frequency of M-band ($\delta_l = 0$, $\omega_l = \Omega_0 - \omega_0$). At low level of excitation the absorption band has the Lorentz-like shape $\chi''/\chi_0 = (1 + \delta^2)^{-1}$ with the maximum at $\delta = 0$. The amplitude of the absorption peak rapidly decreases and the half-width of the absorption band increases, when the pump amplitude f_0 increases. The radical transformation of the spectral shape of the absorption band takes place when intensity of pump pulse approaches

the value $f_0^2 = s^3/(1+2s)$ (Fig. 4a). The central peak at $\delta = 0$ in this case converts into the minimum and two new symmetrical absorptive peaks appear at the detunings

$$\delta_{\pm} = \pm \left[(1+s)f_0 \sqrt{f_0^2 + s} - s(f_0^2 + s) \right]^{1/2}. \quad (11)$$

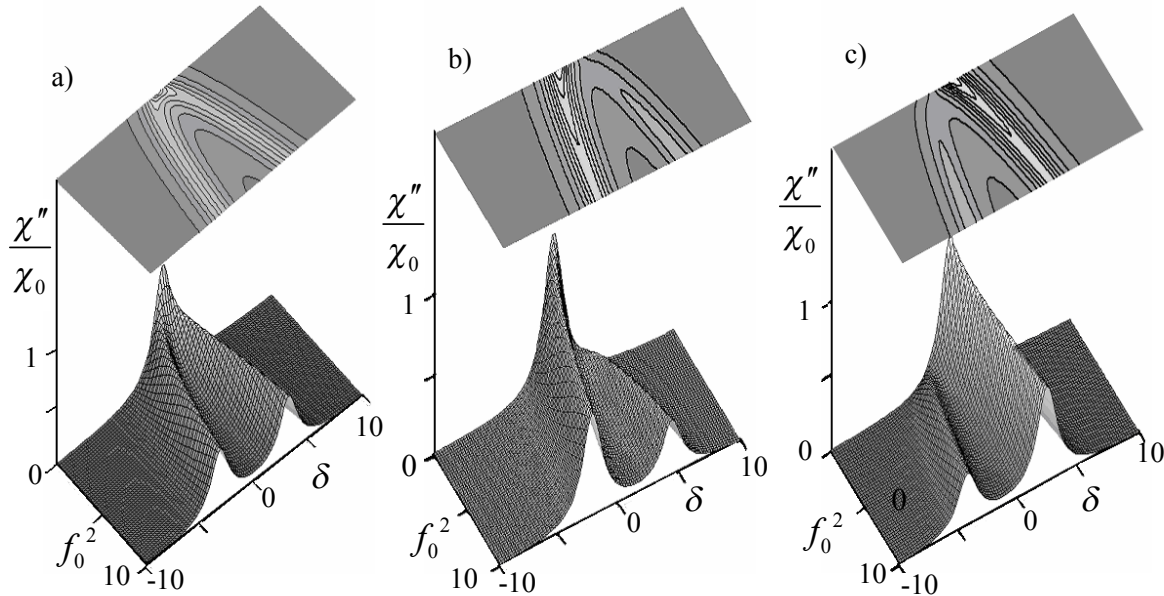


Fig. 4. The absorptive component of the susceptibility χ'' depending on the resonance detuning δ and pump field intensity f_0^2 for the different fixed pump detuning δ_l , which equals a) 0, b) -2 and c) 2.

The new absorptive peaks move apart and their amplitudes decrease monotonously, when the pump amplitude f_0 increases (Fig. 4a). The appearance of two new maxima and conversion of the former maximum into the minimum is due to the renormalization of the energy spectrum of the strong pump pulse. The exciton level splits into two quasilevels, which more and more move apart from the former position of the exciton level with the increasing of f_0 . From (11) we obtain $\delta_{\pm} = \pm f_0$ for the case $f_0^2 \gg s$, which coincides with (6) at $\Delta_l = 0$. From (7) it is seen that for $\delta_l = 0$ the upper and lower polariton branches of the dispersion law symmetrically shift relatively to the frequency ω_0 when the pump amplitude f_0 increases.

If the photons of the pump pulse have the nonzero resonance detuning ($\delta_l \neq 0$), then it takes place the appreciable nonsymmetrical (relatively to $\delta = 0$) renormalization of the absorptive component of the susceptibility χ'' (Fig. 4b, c) due to the peculiarities of the dispersion law. In the absence of the pump field ($f_0 = 0$) the absorptive band as before has the symmetrical Lorenz-like shape. The spectral shape of the absorption band is changed with increasing of f_0 at $\delta_l < 0$. Its maximum rapidly decreases and shifts to the longwave side and the half-width increases (Fig. 4b). At some value of the pump field f_0 the weak shortwavelength peak splits from the shortwave tail of the intense absorption band, the height of which slowly increases with the increase of the excitation level and shifts to the shortwave side. The further increase of the pump amplitude f_0 leads to the shifting of the longwave

absorption peak and to decreasing of its amplitude, whereas the amplitude of the weak shortwave peak slowly increases and shifts to the shortwave side. For the high level of excitation the absorptive susceptibility consists of the well pronounced longwave and weak pronounced shortwave peaks, every of which has a Lorenz shape (Fig. 4b). Analogous phenomena take place in the case $\delta_l > 0$ (Fig. 4c).

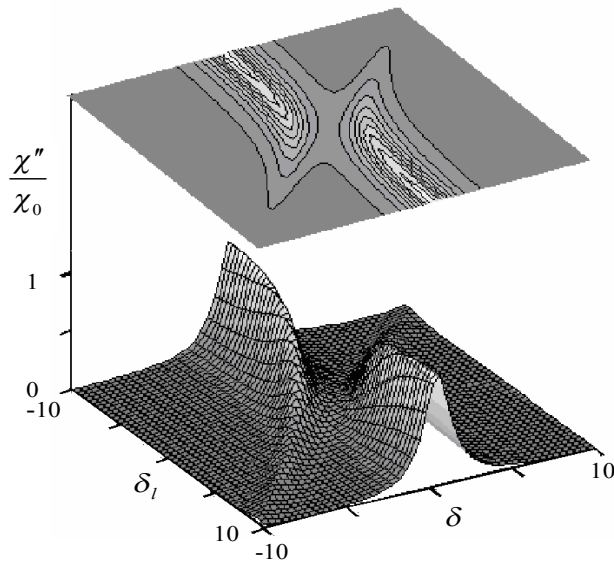


Fig. 5. The absorptive component of the susceptibility χ'' depending on the resonance detuning δ and δ_l of the test and pump pulses for the fixed value of the pump field intensity f_0^2 equals 5.

It is of interest also the dependence of the absorptive component of the susceptibility χ'' on the pump field amplitude f_0 for the fixed values of the resonance detuning δ and δ_l . It follows from (9) that the function $\chi''(f_0)$ slowly decreases with increasing of f_0 for $\delta_- < \delta < \delta_+$, but in the case, when $\delta \geq \delta_+$ and $\delta \leq \delta_-$ the function $\chi''(f_0)$ firstly increases with increasing of f_0 , approaches its maximum at $f_0^2 = \sqrt{s^2 + (\delta + \delta_l)^2} \left[|(s+1)\delta + \delta_l| - \sqrt{s^2 + (\delta + \delta_l)^2} \right] / s$, then monotonously increases (Fig. 4a–c). Here $\delta_+ = \left(\sqrt{\delta_l^2 + s(2+s)} - \delta_l \right) / (2+s)$, $\delta_- = -\left(\sqrt{\delta_l^2 + s(2+s)} + \delta_l \right) / (2+s)$. It is due to the fact that the function χ'' has the resonance character not only in the dependence on the resonance detuning of the weak pulse, but also in the dependence on the pump field amplitude f_0 for the fixed δ . Therefore it takes place the saturation effect of the weak pulse as before, but for the case of low level of excitation it is possible the increase of the absorption with the increase of the level of excitation (Fig. 4a–c).

The evolution of the absorption bands in the plane of variables (δ, δ_l) for fixed values of pump field amplitudes f_0 is presented in Fig. 5. As is seen from Fig. 5 the peak of the longwave (shortwave) absorption band gradually shifts to the longwave side and its height decreases (increases) with the increase of the resonance detuning δ_l of the pump pulse. In the case of large negative (positive) detuning δ_l there exists only longwave (shortwave) peak of the absorption band.

4. Conclusion.

We point out the qualitative similarity of the expressions for the susceptibilities obtained by us in this paper and in [8, 9] in the frame of one and the same model of the energy spectrum of semiconductor, the type of pump, but for two different methods of observation by the weak pulses: in [8, 9] the optical properties are observed by two-photon excitation of biexcitons from the ground state of the crystal and in this paper by the one-photon excitation of excitons.

The conceptual result namely the appearance of the Autler–Townes splitting on the excitons and biexcitons is due to the pump acting and therefore it takes place in both papers. As for the spectral shapes of the absorption bands we can point out that they are qualitatively similar, but they differ in details. Besides in [8, 9] the susceptibility $\chi^{(3)}$ was calculated unlike this paper. In both papers it was proposed that the intensity of the test pump is very small, so that it does not change the energy spectrum of the semiconductor. This test pulse serves for the registration of the changes of optical properties of the medium, which appear under the action of the strong pump pulse in the range of M–band of luminescence.

We carried out the comparison of our results with the experimental ones devoted to the investigation of the optical properties of semiconductors in the exciton range of spectrum in the presence of a strong pump pulse. We pointed out above that the results of our paper qualitatively coincide with the results of the investigation of the absorption spectra in [7]. We can point out also that it takes place the qualitative coincidence of our results with the results of the investigation of the luminescence spectra in quantum dots in InGaAs [20] along the following points: 1) the Autler–Townes splitting appearance and the doublet formation; 2) alteration of the splitting depending on the pump pulse intensity and detuning; 3) coincidence of the peaks of the absorption (luminescence) bands; 4) the linear dependence of the Rabi–frequency of the pump field amplitude. The variation of the absorption bands were observed in [13,20] depending on the resonance detunings δ and δ_l for the fixed pump amplitude f_0 , presented in Fig. 5. In [21] the Rabi–oscillations were observed in semiconductor quantum dots and it was proved that the Rabi–frequency was proportional to the pump field amplitude.

We estimated the dipole momentum $\mu = \hbar\sigma$ of the excitonic transition, using (6) and the experimental data from [20], where it was stated that for the intensity of the excitation $J_0 = 18 \text{ kW cm}^{-2}$ the Autler–Townes $\hbar\Omega$ was equal to $94 \text{ } \mu\text{eV}$. As $J_0 = cE_0^2/4\pi$ we obtain $\mu = \frac{1}{4} \hbar\Omega \sqrt{c/\pi J_0} \approx 3 \cdot 10^{-17} \text{ CGSE} = 30 \text{ D}$. The nutation frequency in this case is equal to $1.5 \cdot 10^{11} \text{ c}^{-1}$.

Thus the obtained expressions for the dispersive and absorptive components of the dielectric susceptibility depending on the level of pump excitation allow us to carry out the detail investigation of the absorptive and dispersive properties of semiconductors in the exciton range of spectrum.

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