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Triexcitons and their effect on absorption in the exciton region of the spectrum

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The effect of the triexciton state on the absorption of exciton-polaritons is studied under conditions when two high-power laser radiation pulses interacting with biexcitons and triexcitons and a probe pulse at the frequency of the exciton transition are incident on the medium. It is shown that even at low triexciton binding energies under the action of two high-power pulses, the exciton state splits into three quasi-levels, and the Autler–Townes effect (optical Stark effect) is observed. It turned out that the position of the quasi-levels depends on the detuning of the resonance of the pump pulses and their intensities, which makes it possible to identify them. These circumstances make it possible to diagnose the triexciton state in semiconductors with a higher degree of probability not by studying the absorption spectral line of the biexciton–triexciton transition, but by the effect of the triexciton state on absorption in the region of the exciton transition.

Keywords: excitons, biexcitons, triexcitons, Autler–Townes effect.

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1. Introduction

The presence of exciton, biexciton, triexciton, and multiexciton states [1,2] and the possibility to selectively control their properties under coherent laser radiation determine to a significant extent optical characteristics of semiconductor structures and, as a consequence, stimulate the development of advanced research activities in many fields: quantum information processing, creation of energy-efficient electronic devices, confocal microscopy [3], polariton generation, Bose-Einstein condensation of exciton-polaritons [4], polariton superfluidity. The discovery of excitons with large quantum numbers [5], that are analogues of the Rydberg atoms, allows studying new effects, that were not observed in the physics of atoms, because bond energies of the Rydberg states of the exciton are small and excitons are more sensitive to the impact of external electric and magnetic fields, therefore with changing intensity of external field a variety of shifts and crossings of absorption levels can be observed.

A number of works study the potential possibilities to increase the generation of quantum-correlated photons radiated by quantum dots using high order excitons, i.e., triexcitons [6,7], and the thin structure of triexciton states in quantum dots [8–11]. One of the most studied issues in quantum optics is the interaction of multiexcitons with electromagnetic field in the mode of strong bond at changing the Rabi frequencies [12]. In [13] the electron structure of multiexcitons with limited quantization in colloidal quantum dots of CdSe is studied experimentally. It is shown that depending on the excitation energy and intensity, a biexciton and triexciton can be observed that demonstrate bright bound states, which are a result of exciton-exciton interaction. Studying of such effects is important for many applications of quantum information in quantum networks [14], implementation of quantum memory cells [15,16], quantum operations with qubits using cold atoms of Rb in quantum informatics [17].

These effects are studied in different modes of excitation by electromagnetic field of polaritons in semiconductor and atomic structures, i.e., taking into account one-photon [18], two-photon [19–22], three-photon [23] transitions or simultaneously taking into account one-photon and multi-photon transitions and their impact on the absorption. The possibility to control the absorption spectrum by changing intensities of fields and detuning resonance in multilevel systems is described in [24–27]. From the analysis of experimental studies a conclusion can be made that observation of triexciton and multiexciton luminescence lines requires special conditions and is a complex task. In addition, since the bond energy of a triexciton is small as compared with bond energies of exciton and biexciton states, the absorption is small as well. On the other hand, as it follows from [28,29], we know that absorption in the exciton region of spectrum changes if biexciton recombination processes are taken into account, the Autler-Townes type splitting of exciton state is observed [30], the presence of power-, polarization- and detuning- dependent resonance of the Autler-Townes type splitting of autolocalized exciton state is shown [31]. In the system of InAs/GaAs AlAs/GaAs the effect of frequency difference of the resonator and natural frequencies, as well as field intensity on the position of absorption levels is studied [32,33]. It is reasonable to expect that taking into account triexciton state will result in additional changes in the reconstruction of the energy spectrum in the exciton region due to successive recombination of triexcitons into the exciton state.

2. Problem formulation

Let a semiconductor is exposed to three ultrashort pulses of resonance laser radiation with frequencies of ω_1, ω_2, and ω_3, respectively (Fig. 1). The first pulse with a frequency of ω_1 excites excitons from the ground state of the crystal, the second pulse with a frequency of ω_2 converts these excitons to biexcitons, the third pulse transforms biexcitons to triexcitons. Studying a system in the pump-probe mode, that was theoretically studied for the system of excitons and biexcitons in [21–23,29], we assume, that pulses acting in the region of exciton–biexciton and biexciton–triexciton transitions are powerful as compared with the pulses that excite excitons from the ground state of the crystal.

Also, another problem formulation is possible. A semiconductor is placed in a microresonator with a natural frequency coinciding with the bond energy of a biexciton or triexciton, with the third pulse sounding the exciton state. The media placed in the microresonator can be semiconductor structures of InAs/GaAs AlAs/GaAs, that have proved themselves to be good for the studying of biexciton states impact on the position of the exciton level, or CuCl-type crystals that are characterized by relatively high bond energy of the biexciton 32 meV [34,35].

At the same time, in the energy diagram the constructive two-photon transitions are taken into account. We assume that pulse length is much less than the relaxation times of particles. In this case the relaxation processes can be ignored, because they have no time to act during the pulse duration (relaxation times for the above-mentioned semiconductors are about 1–10 ps).

The Hamiltonian of pulse interaction with medium can be written as follows

$$H_{\text{int}} = -\hbar g(\hat{a}^+\hat{c}_1 + \hat{c}_1^+\hat{a}) - \hbar\sigma(\hat{a}^+\hat{c}_2^+\hat{b} + \hat{b}^+\hat{c}_2\hat{a}) - \hbar\kappa(\hat{b}^+\hat{c}_3^+\hat{p} + \hat{p}^+\hat{c}_3\hat{b}) - \hbar\mu_{12}(\hat{b}^+\hat{c}_1\hat{c}_2 + \hat{b}\hat{c}_1^+\hat{c}_2^+) - \hbar\mu_{23}(\hat{p}^+\hat{c}_2\hat{c}_3\hat{a} + \hat{p}\hat{c}_2^+\hat{c}_3^+\hat{a}^+), \quad (1)$$

where g — constant of exciton–photon interaction, σ — constant of optical exciton-biexciton conversion, κ — constant of biexciton-triexciton conversion, μ_{12}, μ_{23} — constants of two-photon generation of biexcitons from the ground state of the crystal and triexcitons from the exciton state. It is worth noting, that processes of optical exciton-biexciton conversion and two-photon excitation of biexcitons described by σ and μ_{12} constants are characterized by giant oscillator forces [18,19]. $\hat{a}, \hat{b}, \hat{p}, \hat{c}_1, \hat{c}_2, \hat{c}_3$, ($\hat{a}^+, \hat{b}^+, \hat{p}^+, \hat{c}_1^+, \hat{c}_2^+, \hat{c}_3^+$) operators are annihilation (creation) operators for excitons, biexcitons, triexcitons, and photons, respectively. Using Hamiltonian (1), it is easy to obtain a system of the Heisenberg material equations for $\hat{a}$, $\hat{b}$, $\hat{p}$, $\hat{c}_1$, $\hat{c}_2$, $\hat{c}_3$ operators. In the mean field approximation, after averaging the Heisenberg equations for operators we obtain a system of non-linear differential equations for amplitudes: $a = \langle\hat{a}\rangle$, $b = \langle\hat{b}\rangle$, $p = \langle\hat{p}\rangle$, $c_1 = \langle\hat{c_1}\rangle$, $c_2 = \langle\hat{c_2}\rangle$, $c_3 = \langle\hat{c_3}\rangle$:

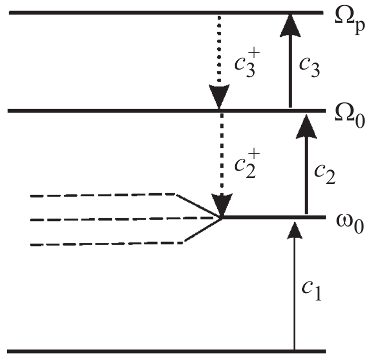

Figure 1. Energy diagram of semiconductor.

$$\begin{aligned} i\dot{a} &= \omega_0 a - g c_1 - \sigma b c_2^* - \mu_{23} p c_2^* c_3^*, \\ i\dot{b} &= \Omega_0 b - \sigma a c_2 - \mu_{12} c_1 c_2 - \kappa p c_3^*, \\ i\dot{p} &= \Omega_p p - \kappa b c_3 - \mu_{23} c_2 c_3 a, \\ i\dot{c}_1 &= \omega_1 c_1 - g a - \mu_{12} b c_2^*, \\ i\dot{c}_2 &= \omega_2 c_2 - \sigma a^* b - \mu_{12} b c_1^*, \\ i\dot{c}_3 &= \omega_3 c_3 - \kappa b^* p - \mu_{23} p c_2^*, \end{aligned} \quad (2)$$

where $\omega_0, \Omega_0, \Omega_p$ — natural frequencies of excitons, biexcitons, and triexcitons, respectively. At the same time, photon densities of power pump pulses are much higher than the density of quasi-particles and photons of the weak pulse acting in the region of exciton transition: $c_2, c_3 \gg c_1, a, b, p$. It can be seen from (2), that equations for a and c_1 include terms proportional to bc_2^* and $pc_2^*c_3^*$. Their respective $\hat{b}\hat{c}_2^+$ and $pc_2^+c_3^+$ operators describe the states with energies of $\hbar(\Omega_0 - \omega_2)$ and $\hbar(\Omega_p - \omega_2 - \omega_3)$, that coincide with the energy of exciton state formation, $\hbar\omega_0$, and are a consequence of the double Autler–Townes effect. Hence, the exciton level and replicas of biexciton and triexciton states, shifted downwards by an energy of photons, $\hbar\omega_2$ and $\hbar(\omega_2 + \omega_3)$, are energy degenerated. Using (2), it is easy to obtain equations of motion for bc_2^* and $pc_2^*c_3^*$ expressions:

$$\begin{aligned} i(bc_2^*)^\bullet &= (\Omega_0 - \omega_2)bc_2^* - f_{20}(\sigma a + \mu_{12}c_1) - \kappa p c_2^* c_3^*, \\ i(pc_2^*c_3^*)^\bullet &= (\Omega_p - \omega_2 - \omega_3)pc_2^*c_3^* \\ &\quad - \kappa c_2^* b f_{30} - \mu_{23} a f_{20} f_{30}, \end{aligned} \quad (3)$$

where f_{20} and f_{30} — photon densities of the second and the third pulses, respectively.

Within the limit of pre-defined fields of the second and the third pulses $c_2 = c_{20} = \text{const}$, $c_3 = c_{30} = \text{const}$, system (3) can be written as follows

$$i(bc_{20}^*)^\bullet = (\Omega_0 - \omega_2)bc_{20}^* - f_{20}(\sigma a + \mu_{12}c_1) - \kappa pc_{20}^*c_{30}^*,$$
$$i(pc_{20}^*c_{30}^*)^\bullet = (\Omega_p - \omega_2 - \omega_3)pc_{20}^*c_{30}^* - \kappa c_{20}^* bf_{30} - \mu_{23}af_{20}f_{30}. \quad (4)$$

Equations (4), that control the time evolution of bc_{20}^* and $pc_2^*c_3^*$ relationships, are linear in the above-mentioned approximations. In combination with equations for a and c_1 from (2) they form a system of four linear equations for amplitudes of quasi-particles with the same energy $\hbar\omega \approx \hbar\omega_1 \approx \hbar(\Omega_0 - \omega_2) \approx \hbar(\Omega_p - \omega_2 - \omega_3)$:

$$i\dot{a} = \omega_0 a - gc_1 - \sigma(bc_{20}^*) - \mu_{23}(pc_{20}^*c_{30}^*),$$
$$i\dot{c}_1 = \omega_1 c_1 - ga - \mu_{12}(bc_{20}^*),$$
$$i(bc_{20}^*)^\bullet = (\Omega_0 - \omega_2)(bc_{20}^*) - f_{20}(\sigma a + \mu_{12}c_1) - \kappa(pc_2^*c_3^*),$$
$$i(pc_{20}^*c_{30}^*)^\bullet = (\Omega_p - \omega_2 - \omega_3)(pc_{20}^*c_{30}^*) - \kappa(bc_{20}^*)f_{30} - \mu_{23}af_{20}f_{30}. \quad (5)$$

We shall search for a solution to system (5) in the form of a, c_1, bc_{20}^*, $pc_{20}^*c_{30}^* \sim \exp(-i\omega t)$, where ω is natural frequency of new polaritons in the exciton region of the spectrum. In steady-state mode we get the following determinant:

$$\begin{vmatrix} \omega - \omega_0 & g & \sigma & \mu_{23} \\ g & \omega - \omega_1 & \mu_{12} & 0 \\ \sigma f_{20} & \mu_{12}f_{20} & \omega - (\Omega_0 - \omega_2) & \kappa \\ \mu_{23}f_{20}f_{30} & 0 & \kappa f_{30} & \omega - (\Omega_p - \omega_2 - \omega_3) \end{vmatrix}$$

By equating the determinant to zero we obtain a quartic equation.

If we introduce all Rabi frequencies

$$\Omega_\sigma^2 = \sigma^2 f_{20},\ \Omega_{\mu 12}^2 = \mu_{12}^2 f_{20},\ \Omega_{ex}^2 = g^2,$$
$$\Omega_\kappa^2 = \kappa^2 f_{30},\ \Omega_{\mu 23}^2 = \mu_{23}^2 f_{20} f_{30}, \quad (6)$$

then this equation can be written as follows

$$(\omega - \omega_0)(\omega - \omega_1)(\omega - \bar{\omega}_2)(\omega - \bar{\omega}_3) + I_1 + I_2 = 0, \quad (7)$$

$$I_1 = -(\omega - \bar{\omega}_2)(\omega - \bar{\omega}_3)\Omega_{ex}^2 - (\omega - \omega_1)(\omega - \bar{\omega}_3)\Omega_\sigma^2 - (\omega - \omega_1)(\omega - \omega_0)\Omega_\kappa^2 - \Omega_{\mu 12}^2(\omega - \omega_0)(\omega - \bar{\omega}_3) - \Omega_{\mu 23}^2(\omega - \omega_1)(\omega - \bar{\omega}_2),$$

$$I_2 = 2(\omega - \omega_1)\Omega_\kappa\Omega_\sigma\Omega_{\mu 23} + 2(\omega - \bar{\omega}_3)\Omega_{ex}\Omega_{\mu 12}\Omega_\sigma + \Omega_{ex}^2\Omega_\kappa^2 + \Omega_{\mu 23}^2\Omega_{\mu 12}^2 - 2\Omega_{ex}\Omega_\kappa\Omega_{\mu 12}\Omega_{\mu 23}.$$

In (7) there are six terms of sum, each containing a square of appropriate constant of interaction, i.e. these terms describe the contribution of each individual process. In addition, in (7) there are interference terms of sum of different degrees.

By expressing from (7) the wave vector k_1 of weak pulse photon, we can represent in an explicit form its dependence on ω:

$$ck_1 \equiv \omega_1 = \omega + ch/zn, \quad (8)$$

where

$$ch = -(\omega - \bar{\omega}_2)(\omega - \bar{\omega}_3)\Omega_{ex}^2 - \Omega_{\mu 12}^2(\omega - \omega_0)(\omega - \bar{\omega}_3) + \Omega_{ex}^2\Omega_\kappa^2 + \Omega_{\mu 23}^2\Omega_{\mu 12}^2 + 2(\omega - \bar{\omega}_3)\Omega_{ex}\Omega_{\mu 12}\Omega_\sigma + 2\Omega_{ex}\Omega_\kappa\Omega_{\mu 12}\Omega_{\mu 23}, \quad (9)$$

$$zn = (\omega - \omega_0)(\omega - \bar{\omega}_2)(\omega - \bar{\omega}_3) - \Omega_\sigma^2(\omega - \bar{\omega}_3) - \Omega_\kappa^2(\omega - \omega_0) - \Omega_{\mu 23}^2(\omega - \bar{\omega}_2) + 2\Omega_\kappa\Omega_\sigma\Omega_{\mu 23}. \quad (10)$$

Roots of equation (10) determine frequencies of quasi-levels that depend on the Rabi frequencies, that is on densities of pump pulses.

Thus, the conclusion can be made that action of two pump pulses in the regions of exciton-biexciton and biexciton-triexciton conversions results in a strongly pronounced double Autler–Townes effect. The exciton level splits into three quasi-levels, which positions are determined by resonance detunings.

By equating (10) to zero, we study the obtained equation. Let us assume, that the Rabi frequencies are infinitely low $\Omega_\kappa, \Omega_\sigma, \Omega_{\mu 23} \ll 1$. In this case (10) can be written as $(\omega - \omega_0)(\omega - \bar{\omega}_2)(\omega - \bar{\omega}_3) = 0$. Hence, the position of quasi-levels is determined by frequencies of $\omega_0, \bar{\omega}_2, \bar{\omega}_3$, i.e., natural frequency of the exciton transition and two replicas of biexciton and triexciton states. For this reason, in the following discussion we define quasi-levels as triexciton, biexciton, and exciton levels in accordance with the mechanism resulted in the emergence of the quasi-level. Since frequencies of $\bar{\omega}_2 = \Omega_0 - \omega_2$ and $\bar{\omega}_3 = \Omega_p - \omega_2 - \omega_3$ are determined by pulse frequencies, then, by varying their intensity, the position of quasi-levels can be controlled.

Let us consider the conditions of equality for frequencies: $\bar{\omega}_2 = \omega_0$, $\bar{\omega}_3 = \omega_0$, $\bar{\omega}_3 = \bar{\omega}_2$. In this case a degeneration of quasi-energy states can be observed. Frequencies of incident pulses in this case should be equal to each other: $\omega_2 = \Omega_0 - \omega_0$, $\omega_2 + \omega_3 = \Omega_p - \omega_0$, $\omega_3 = \Omega_p - \Omega_0$. Hence, the degeneration of quasi-energy states at low Rabi frequencies $\Omega_\kappa, \Omega_\sigma, \Omega_{\mu 23} \ll 1$ is observed at the condition when power pump pulses act in conditions of precise resonance with a frequency of exciton transition or when pump pulse resonance detunings compensate each other.

Lett us consider successively the impact of $\Omega_\kappa, \Omega_\sigma, \Omega_{\mu 23}$ Rabi frequencies on positions of quasi-levels. Let us assume that Rabi frequency $\Omega_\sigma \gg \Omega_\kappa, \Omega_{\mu 23}$, then (10) can be transformed as follows

$$(\omega - \omega_0)(\omega - \bar{\omega}_2)(\omega - \bar{\omega}_3) - \Omega_\sigma^2(\omega - \bar{\omega}_3) = 0. \quad (11)$$

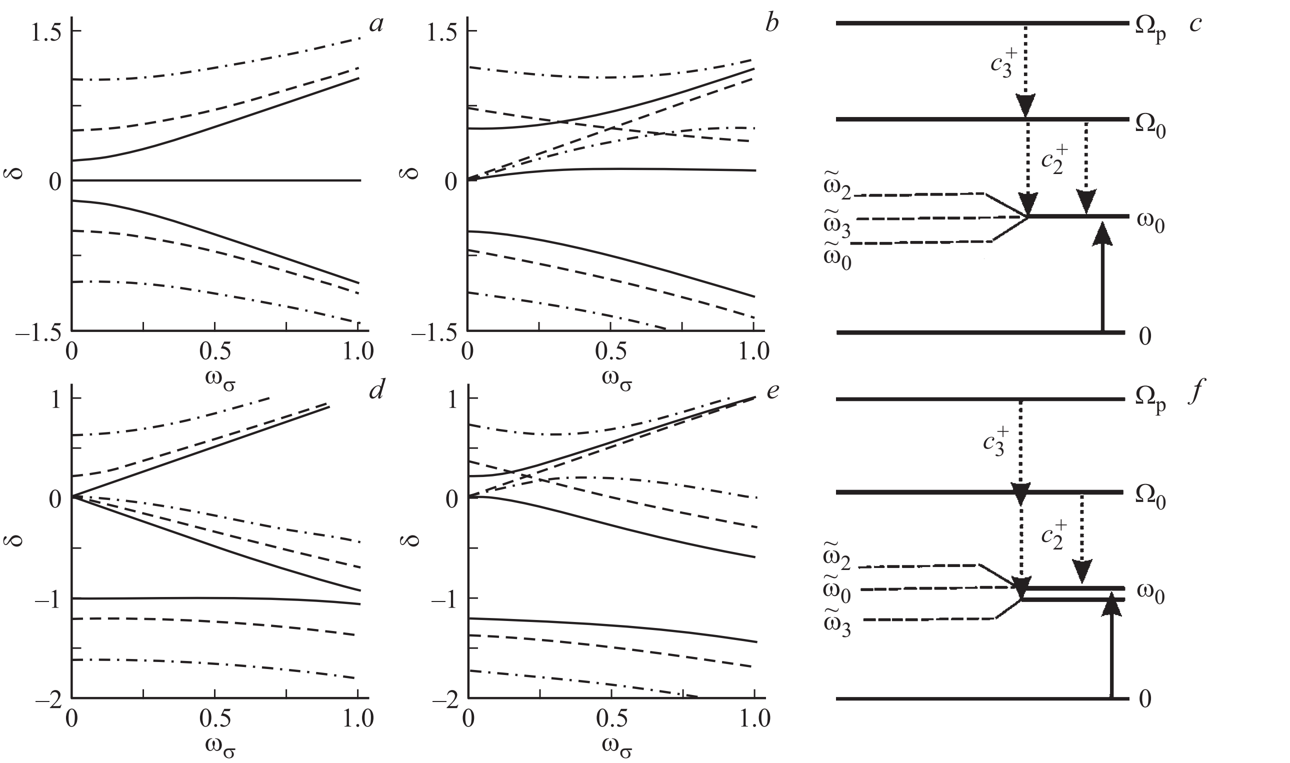

Figure 2. Dependencies $\delta(\omega_\sigma)$, that determine the position of normalized frequencies of absorption for quasi-levels of (a, b, d, e) and energy diagrams (c,f) at different parameters: a — $\delta_2 = 0$, $\delta_3 = 0$, $\omega_{\mu 23} = 0$; b — $\delta_2 = 0$, $\delta_3 = 0$, $\omega_{\mu 23} = 0.5$; d — $\delta_2 = 0$, $\delta_3 = 1$, $\omega_{\mu 23} = 0$; e — $\delta_2 = 0$, $\delta_3 = 1$, $\omega_{\mu 23} = 0.5$ and μ_κ equal to 0.1 (solid lines), 0.5 (dashed lines), 1.0 (dash-dotted lines).

Solution to equation (11) will be as follows

$$\tilde{\omega}_3 = \bar{\omega}_3,\ \ \tilde{\omega}_{0,2} = \left(\omega_0 + \bar{\omega}_2 \pm \sqrt{(\omega_0 - \bar{\omega}_3)^2 + 4\Omega_\sigma^2}\right)/2, \tag{12}$$

where $\tilde{\omega}_{0,2,3}$ are new frequencies of shifted quasi-levels subject to $\Omega_\sigma \gg \Omega_\kappa, \Omega_{\mu 23}$. With $\Omega_\kappa \gg \Omega_\sigma, \Omega_{\mu 23}$ frequencies of quasi-levels are determined by the following relationship

$$\tilde{\omega}_0 = \omega_0,\ \ \tilde{\omega}_{2,3} = \left(\bar{\omega}_3 + \bar{\omega}_2 \pm \sqrt{(\bar{\omega}_2 - \bar{\omega}_3)^2 + 4\Omega_\kappa^2}\right)/2. \tag{13}$$

With $\Omega_{\mu 23} \gg \Omega_\sigma, \Omega_\kappa$ frequencies of quasi-levels are determined by the following relationships

$$\tilde{\omega}_2 = \bar{\omega}_3,\ \ \tilde{\omega}_{0,3} = \left(\omega_0 + \bar{\omega}_3 \pm \sqrt{(\omega_0 - \bar{\omega}_3)^2 + 4\Omega_{\mu 23}^2}\right)/2. \tag{14}$$

In addition to the above-mentioned terms, (10) includes the term of $2\Omega_\kappa\Omega_\sigma\Omega_{\mu 23}$, which is a consequence of quantum interference of the Rabi frequencies and taking which into account affects the position of quasi-levels as well.

The analysis of (12) shows that when measuring Ω_σ, the frequency of triexciton quasi-level remains unchanged as compared with frequencies of two other quasi-levels. It can be seen from (13)–(14), that the position of exciton and biexciton quasi-levels with changing Ω_κ and $\Omega_{\mu 23}$ remains unchanged. These circumstances make it possible to conditionally identify quasi-levels in the case when detunings of resonance of the incident pump pulses equal to zero. Thus, the position of quasi-levels depends on the intensity and frequencies of ω_2, ω_3 of pump pulses.

Let us introduce detunings of resonance, $\omega - \omega_0 = \Delta$, $\bar{\omega}_2 - \omega_0 = \Delta_2$, $\bar{\omega}_3 - \omega_0 = \Delta_3 + \Delta_2$, of incident pulses and normalize the detunings of resonance and Rabi frequencies at the exciton-photon interaction frequency Ω_g. In this case equation (10) can be written as follows

$$\delta(\delta + \delta_2)(\delta + \delta_2 + \delta_3) - \omega_\sigma^2(\delta + \delta_2 + \delta_3) - \omega_\kappa^2\delta - \omega_{\mu 23}^2(\delta + \delta_2) + 2\omega_\kappa\omega_\sigma\omega_{\mu 23} = 0. \tag{15}$$

3. Discussion of results

Solutions to equation (15) describe the position of absorption frequencies of quasi-levels depending on detunings of resonance and Rabi frequencies. Fig. 2–4 shows dependencies $\delta(\omega_\sigma)$, that determine the position of quasi-levels depending on the Rabi frequency ω_σ at different sets of parameters and energy diagrams that demonstrate the processes that take place. $\tilde{\delta}_0, \tilde{\delta}_2, \tilde{\delta}_3$ symbols used below define the values of absorption frequencies of exciton, biexciton, and triexciton quasi-levels.

Fig. 2, a–c shows the case of precise resonance of pump pulses $\delta_2 = \delta_3 = 0$. In this case equation (15) can be

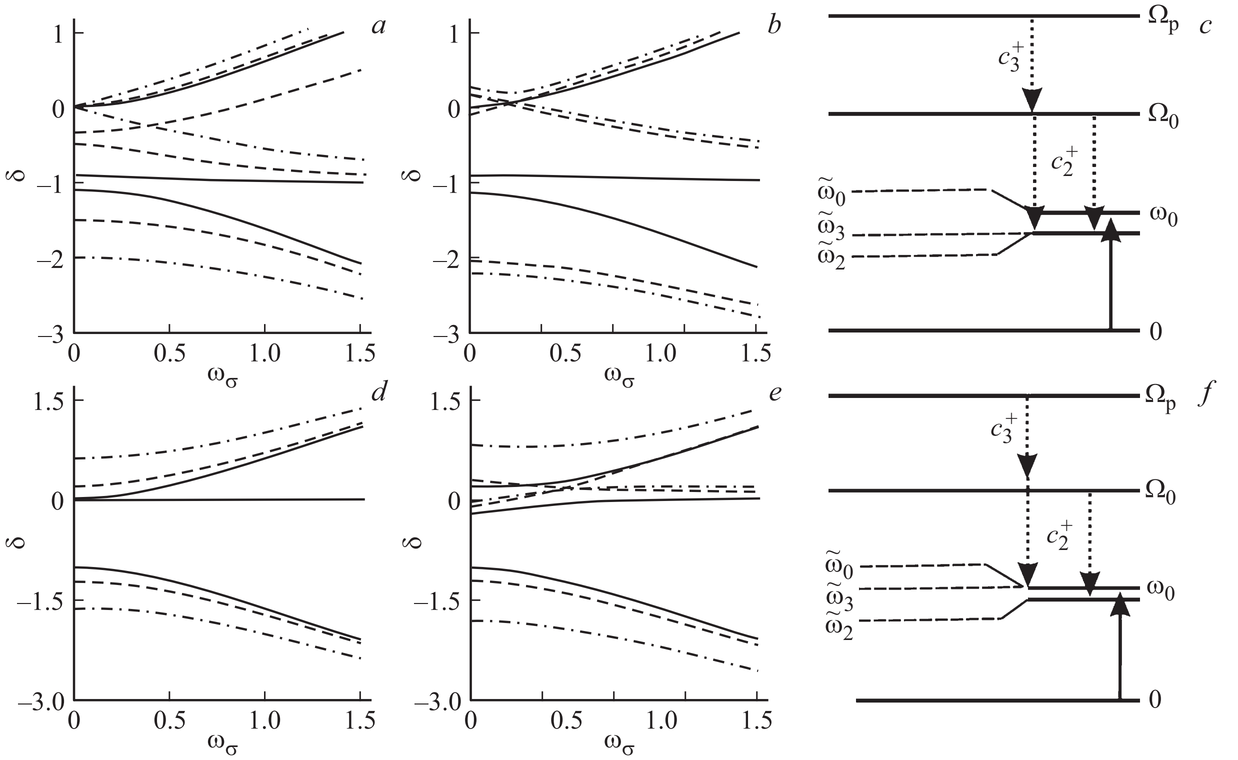

Figure 3. Dependencies $\delta(\omega_\sigma)$, that determine the position of normalized frequencies of absorption for quasi-levels of (a, b, d, e) and energy diagrams (c, f) at different parameters: a — $\delta_2 = 0$, $\delta_3 = 0$, $\omega_{\mu 23} = 0$; b — $\delta_2 = 0$, $\delta_3 = 0$, $\omega_{\mu 23} = 0.5$; d — $\delta_2 = 0$, $\delta_3 = 1$, $\omega_{\mu 23} = 0$; e — $\delta_2 = 0$, $\delta_3 = 1$, $\omega_{\mu 23} = 0.5$ and μ_κ equal to 0.1 (solid lines), 0.5 (dashed lines), 1.0 (dash-dotted lines).

written as follows:

$$\delta^3 - (\omega_\sigma^2 + \omega_\kappa^2 + \omega_{\mu 23}^2)\delta + 2\omega_\kappa \omega_\sigma \omega_{\mu 23} = 0. \quad (16)$$

As it was shown before, in this case $\bar{\omega}_3 = \bar{\omega}_2 = \omega_0$, therefore, the impact of one or another transition on the position of quasi-levels can not be identified unambiguously. The triexciton quasi-level can be identified conditionally since its position does not change with a change in ω_σ and is determined by (12).

Let us consider the case of weak impact of two-photon interaction with triexcitons $\omega_{\mu 23} \ll 1$ (Fig. 2, a). In this case the position of absorption bands is determined by relationships for normalized absorption frequencies of quasi-levels $\tilde{\delta}_3 = 0$, (frequency of triexciton quasi-level coincides with the frequency of exciton state) and $\tilde{\delta}_{0,2} = \pm\sqrt{\omega_\sigma^2 + \omega_\kappa^2}$. With these conditions three absorption frequencies are observed, two of which linearly move off the central band as ω_0 grows. With $\omega_\sigma \ll 1$ position of absorption frequencies will be determined by the following relationships: $\tilde{\delta}_3 = 0$, $\tilde{\delta}_{0,2} \approx \pm\omega_\kappa^2$. Solid lines shows the case of $\omega_\kappa = 0.1$. As ω_κ increases, the distance between frequencies becomes larger.

Fig. 2, b shows the case of $\omega_{\mu 23} \neq 0$. With $\omega_\sigma \ll 1$ position of absorption frequencies will be determined by the following relationships: $\tilde{\delta}_3 = 0$, $\tilde{\delta}_{0,2} \approx \pm\sqrt{(\omega_{\mu 23}^2 + \omega_\kappa^2)}$. An increase in ω_κ and $\omega_{\mu 23}$ results in approaching of absorption frequencies of quasi-levels to each other in the short-wave region of the spectrum. The energy state degenerates despite the non-zero Rabi frequencies. With certain values of parameters there are only two absorption frequencies (dashed lines in Fig. 2, b), frequency coincidence takes place when $(\omega_\sigma^2 + \omega_\kappa^2 + \omega_{\mu 23}^2)^2/27 = (\omega_\kappa \omega_\sigma \omega_{\mu 23})^2$, which is only met when $\omega_\kappa = \omega_\sigma = \omega_{\mu 23}$. In this case frequencies of quasi-levels are determined by the following relationships: $\tilde{\delta}_{0,3} = \omega_\sigma$, $\tilde{\delta}_2 = -2\omega_\sigma$. Further increase in ω_κ results in the repeated emergence of three quasi-levels and an increase in distance between them. Thus, taking into account the term $\omega_\kappa \omega_\sigma \omega_{\mu 23}$ in (15), that is responsible for quantum interference, results in approaching of absorption frequencies of quasi-levels to each other and degeneration of energy states at equal Rabi frequencies.

Fig. 2, d–f shows the case of $\delta_2 = 0$, $\delta_3 > 0$. With these conditions equation (15) can be written as follows: $\delta^2(\delta + \delta_3) - (\omega_\kappa^2 + \omega_{\mu 23}^2)\delta - (\delta + \delta_3)\omega_\sigma^2 + 2\omega_\kappa \omega_\sigma \omega_{\mu 23} = 0$. Solid lines in Fig. 2, d show the case of $\omega_\kappa \approx 0$. Position of shifted absorption frequencies of quasi-levels is determined by the following relationships: $\tilde{\delta}_3 = \delta_3$, $\tilde{\delta}_{0,2} = \pm\omega_\sigma$. Quasi-level that emerges in the region of negative resonance detunings, is a triexciton level, because its position does not change with an increase in ω_σ. Taking into account that with an increase in ω_κ at $\omega_\sigma = 0$ (Fig. 2, d) the absorption frequency of the middle quasi-level is almost unchanged (13), therefore, we shall consider it as exciton level.

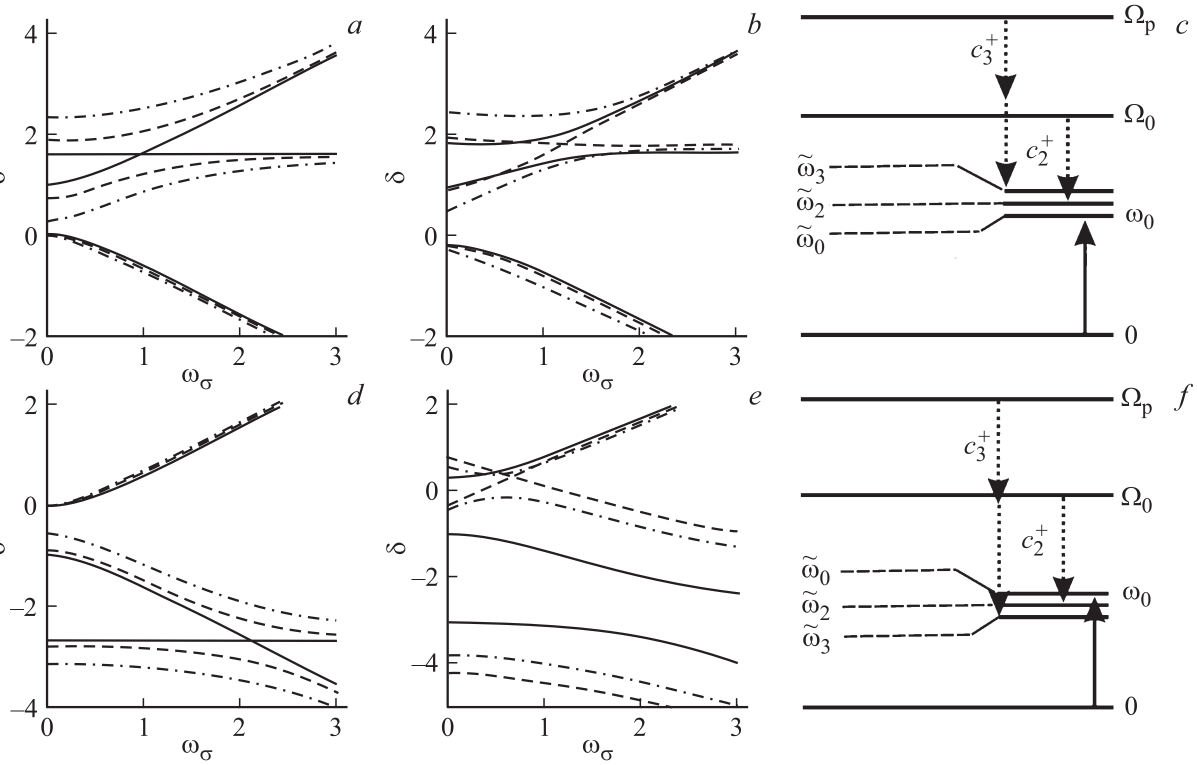

Figure 4. Dependencies $\delta(\omega_\sigma)$, that determine position of normalized frequencies of absorption for quasi-levels of (a, b, d, e) and energy diagrams (c, f) at different parameters: a — $\delta_2 = 1$, $\delta_3 = 0$, $\omega_{\mu 23} = 0$ and μ_κ equal to 0.1 (solid lines), 0.5 (dashed lines), 1.0 (dash-dotted lines); b — $\delta_2 = 1$, $\delta_3 = 0$, $\omega_{\mu 23} = 0.2$ and μ_κ equal to 0.1 (solid lines), 0.5 (dashed lines), 1.2 (dash-dotted lines); d — $\delta_2 = 1$, $\delta_3 = -1$, $\omega_{\mu 23} = 0$ and μ_κ equal to 0.1 (solid lines), 0.5 (dashed lines), 1.0 (dash-dotted lines); e — $\delta_2 = 1$, $\delta_3 = -1$, $\omega_{\mu 23} = 0.2$ and μ_κ equal to 0.1 (solid lines), 0.5 (dashed lines), 1.2 (dash-dotted lines).

At low ω_σ the position of absorption frequencies of quasi-levels is determined by the following relationships $\tilde{\delta}_2 = \delta_2 = 0$, $\tilde{\delta}_{0,3} = \left(\delta_3 \pm \sqrt{\delta_3^2 + 4(\omega_{\mu 23}^2 + \omega_\kappa^2)}\right)/2$. With $\omega_\kappa, \omega_{\mu 23} \ll 1$ shifted frequencies are determined by the following relationships: $\tilde{\delta}_{2,0} = 0$, $\tilde{\delta}_3 = \delta_3$ (Fig. 2, d). It can be seen from the comparison of these results with the case shown in Fig. 2, a, that frequencies of exciton and biexciton quasi-levels are nearly the same at low values of the Rabi frequency ω_σ. The described cases are important from the experimental point of view. Let us assume that we have a semiconductor with a low bond energy of triexciton ($\omega_\kappa \approx 0$, $\omega_{\mu 23} \approx 0$). ω_2 and ω_3 pulses act in conditions of precise resonance. In this case the researcher will observe three quasi-levels spaced at an insignificant distance apart from each other (Fig. 2, a). With abrupt change in ω_3, frequency of the quasi-level emerged as a result of cascade recombination of triexcitons shifts by a frequency equal to resonance detuning of the pulse with a frequency of ω_3 (Fig. 2, d, f). In addition, a change in ω_3 results in approaching of two other quasi-levels to each other at low Rabi frequencies.

In the case when all Rabi frequencies are different from zero, the position of the quasi-level is significantly affected by the interference term in (15), which is responsible for the approaching of exciton and biexciton quasi-level frequencies to each other (Fig. 2, e). In case of coincidence of the Rabi frequencies, $\omega_{\mu 23} = \omega_\kappa$ and $\omega_\sigma = -1/2\delta_3 + 1/2\sqrt{\delta_3^2 + 4\omega_\kappa^2}$, frequencies of exciton and biexciton quasi-levels coincide with each other and are determined by the following relationships: $\tilde{\delta}_3 = -\delta_3 - 2\omega_\sigma$, $\tilde{\delta}_{0,2} = \omega_\sigma$.

Fig. 3, a–c shows the case of $\delta_2 > 0$, $\delta_3 = 0$. With these conditions (15) can be written as follows: $\delta(\delta + \delta_2)^2 - (\omega_\sigma^2 + \omega_{\mu 23}^2)(\delta + \delta_2) - \delta\omega_\kappa^2 + 2\omega_\kappa\omega_\sigma\omega_{\mu 23} = 0$. In the case shown in Fig. 3, a, b solid lines represent $\omega_\kappa \ll 1$, the position of absorption frequencies of quasi-levels will be determined by the following relationships: $\tilde{\delta}_3 \approx \delta_2$, $\tilde{\delta}_{0,2} \approx 1/2\delta_2 \pm 1/2\sqrt{\delta_2^2 + 4(\omega_\sigma^2 + \omega_{\mu 23}^2)}$. With an increase in ω_σ and $\omega_{\mu 23}$, the position of the triexciton quasi-level remains almost unchanged, at the same time the absorption frequency of the biexciton quasi-level decreases with growing ω_σ and $\omega_{\mu 23}$, and the frequency of the exciton quasi-level increases (Fig. 3, a).

With low, but non-zero, ω_κ and $\omega_{\mu 23}$ three absorption bands are observed with the position of one of them fully determined by the detuning of resonance δ_2, and other two dependent on the δ_2 detuning of resonance and Ω_σ Rabi frequency. The case that takes into account ω_κ

is shown in Fig. 3, *a, b* by dashed and dash-dotted lines. The frequency of the triexciton quasi-level decreases at ω_κ growing, while the frequency of the biexciton quasi-level grows. the frequency of the exciton quasi-level remains nearly unchanged with an increase in ω_κ. The impact of interference term results in approaching of triexciton and biexciton quasi-level frequencies to each other (Fig. 3, *b*). With $\omega_\sigma = \Omega_{\mu 23} = \sqrt{-\delta_2\omega_\kappa + \omega_\kappa^2}$ frequencies of triexciton and biexciton quasi-levels coincide with each other and position of the absorption frequencies in this case is determined by the following relationships: $\tilde{\delta}_0 = -2\omega_\kappa$ and $\tilde{\delta}_{2,3} = -\delta_2 + \omega_\kappa$.

Fig. 3, *d–f* shows the case of $\delta_3 = -\delta_2$. With these values of parameters (15) can be written as follows: $\delta^2(\delta + \delta_2) - (\omega_\sigma^2 + \omega_\kappa^2)\delta - \omega_{\mu 23}^2(\delta + \delta_2) + 2\omega_\kappa\omega_\sigma\omega_{\mu 23} = 0$. In the case shown in Fig 3, *d* at $\omega_{\mu 23} = 0$ position of absorption frequencies of quasi-levels will be determined by the following relationships: $\tilde{\delta}_3 = 0$, $\tilde{\delta} = 1/2 \times \left(\delta_2 \mp \sqrt{\delta_2^2 + 4(\omega_\sigma^2 + \omega_\kappa^2)}\right)$. An increase in ω_κ, ω_σ results in the growth of the frequency of the exciton quasi-level and decrease in the frequency of the biexciton quasi-level. With low values of $\omega_{\mu 23} \approx 0$ the position of the triexciton quasi-level coinciding with the frequency of the exciton level does not change with an increase in ω_σ (Fig. 3, *d*). With an increase in $\omega_{\mu 23}$ the frequency of the exciton quasi-level grows, while the frequency of the triexciton quasi-level decreases with an increase in ω_σ and ω_κ. Taking into account the interference results in approaching of exciton and triexciton quasi-level frequencies to each other. The degeneration of quasi-energy states is observed at $\omega_\kappa = \omega_\sigma = \sqrt{\delta_2\omega_{\mu 23} + \omega_{\mu 23}^2}$, at the same time positions of quasi-levels are determined by the following relationships: $\tilde{\delta}_2 = -\delta_2 - 2\omega_{\mu 23}$, $\tilde{\delta}_{0,3} = \omega_{\mu 23}$.

Up to now, we have considered cases of resonance values of pump pulses: $\bar{\omega}_3 = \bar{\omega}_2$, $\bar{\omega}_2 = \omega_0$, $\bar{\omega}_3 = \omega_0$. As it was shown above, the degeneration of quasi-energy states is observed, if at least two of the Rabi frequencies are equal to each other. Generally, with arbitrary non-zero detunings of resonance, a change takes place in the restrictions imposed on Rabi frequencies at which quasi-energy states coincide with each other.

Let us consider some of these cases. The case of $\delta_3 = 3\delta_2/5$ ($\delta_2, \delta_3 < 0$) is shown in Fig. 4, *a–c*. In the case shown in Fig. 4, *a* solid lines correspond to $\omega_\kappa = 0$. In this case it is possible to unambiguously identify quasi-energy states, since $\omega_0 \neq \bar{\omega}_2 \neq \bar{\omega}_3$. From (15) the Rabi frequencies $\omega_{\mu 23} = 0$, $\omega_\kappa = 0$, $\omega_\sigma = 2|\delta_2|/5\sqrt{6}$ were derived at which biexciton and triexciton quasi-levels coincide with each other. With an increase in ω_κ (Fig. 4, *a, b*) the distance between absorption frequencies of biexciton and triexciton quasi-levels increases and an increase in $\omega_{\mu 23}$ results in the shift of absorption frequencies of quasi-levels (Fig. 4, *b*). The coincidence of absorption frequencies of exciton and triexciton quasi-levels takes place at $\omega_{\mu 23} = \omega_\sigma/2$, $\omega_\kappa = \omega_\sigma/3$, $\omega_\sigma = 6|\delta_2|/5$. Absorption frequencies of quasi-levels in this case are determined by the following relationships: $\tilde{\delta}_0 = \delta_2$, $\tilde{\delta}_{2,3} = -9\delta_2/5$.

Fig. 4, *d–f* shows the case of $\delta_3 = 5\delta_2/3$ ($\delta_2, \delta_3 > 0$). Solid lines (Fig. 4, *d*) show the case of $\omega_{\mu 23} = 0$, $\omega_\kappa = 0$ and $\omega_\sigma = 2\delta_2/3\sqrt{10}$, and in this case triexciton and biexciton quasi-levels coincide with each other. With an increase in ω_κ (Fig. 4, *d*) the distance between absorption frequencies of biexciton and exciton quasi-levels increases. An increase in $\omega_{\mu 23}$ results in the shift of absorption frequencies of quasi-levels (Fig. 4, *e*). At the Rabi frequencies of $\omega_{\mu 23} = 3\omega_\sigma/2$, $\omega_\kappa = 3\omega_\sigma$, $\omega_\sigma = -2\delta_2/3$ (Fig. 4, *f*) absorption frequencies of exciton and triexciton quasi-levels coincide with each other. In this case absorption frequencies of quasi-levels are determined by the following relationships: $\tilde{\delta}_3 = -13\delta_2/3$, $\tilde{\delta}_{0,2} = \delta_2/3$.

Results shown in Figs. 2–4 are indicative of a considerable impact of the Rabi frequencies of $\omega_{\mu 23}$, ω_κ, ω_σ on the absorption in the exciton region of the spectrum. The Rabi frequencies, in turn, are determined by field intensities of incident pulses.

Currently, there is no experimental data for the estimate of field intensities at which splitting of the exciton level is observed when triexciton states are taken into account. Here we present experimental data for the estimate of the field intensity of the exciton state splitting due to taking into account the impact of the mechanism of exciton-biexciton conversion. According to [36], in quantum dots on InGaAs the intensity of pumping where splitting of quasi-levels is observed is $18\,\mathrm{kW/cm^2}$ and the value of the splitting in this case is $94\,\mu\mathrm{eV}$. With an increase in intensity up to $50\,\mathrm{kW/cm^2}$, the splitting increases up to $150\,\mu\mathrm{eV}$. Since the value of the splitting does not depend on the pulse intensity acting in the region of exciton transition, it can be arbitrary, but sufficient to ensure necessary concentration of excitons, and its intensity can vary from $2-5\,\mathrm{kW/cm^2}$. Even if the bond energy of the triexciton is by an order of magnitude less than the bond energy of the biexciton, these effects can be observed at a pump intensity not greater than $1\,\mathrm{MW/cm^2}$.

4. Conclusion

Thus, under the action of two strong pumpings the splitting of exciton level takes place and three quasi-energy states arise. Frequencies of new quasi-levels are determined by detuning of resonance of the strong pump pulses, that allows for their identification. In addition, the position of absorption bands is affected by $\omega_{\mu 23}$, ω_κ, and ω_σ Rabi frequencies that are determined by intensities of the strong pump pulses, and their values can vary in a wide range. Taking into account the constructive two-photon transition attributable to the value of $\omega_{\mu 23}$ does not result in the emergence of a new quasi-level, however it leads to the Stark shifts of quasi-levels. It is worth noting, that (15) includes the $2\omega_\kappa\omega_\sigma\omega_{\mu 23}$ term attributable to quantum interference of all taken into account mechanisms of non-linearity, and with certain parameters the quantum interference results in the possibility of coincidence of the absorption frequencies of quasi-levels. Thus, despite the low bond energy of biexciton-triexciton conversion, at a low field acting in the region of biexciton-triexciton transition a shift of quasi-levels arising under the action of two other fields will be observed. An increase in the field acting in the region of biexciton-triexciton transition will result in the emergence of the third quasi-level in the region of exciton transition.

Conflict of interest

The authors declare that they have no conflict of interest.

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