Possibility of a parametrical resonance when investigating optical properties of the semiconductors in exciton range of spectrum

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***Abstract* — The results of the theoretical investigation of the semiconductor susceptibilities in the frame of pump–probe approach are presented using the exciton–photon and exciton– exciton interaction. The strong pump beam excitates excitons.The excitons interact with each other and the weak laser pulse with the frequency tuned to the exciton transition tests the optical properties of a semiconductor.**

***Index Terms* —** exciton, pump-probe, non-stationary, exponential coefficient**.**

I. INTRODUCTION

Pump-probe method of theoretical and experimental investigation of optical properties of semiconductors in the exciton range of spectrum at high level of pump excitation acquired the great significance. This method is based on the use of two beams of laser radiation, namely the strong pump beam and the weak probe beam. The probe beam tests changing of optical properties of the crystal due to the action of the strong pump pulse. The kinetics of the radiative recombination of biexcitons, the nonlinear response of the system of excitons and biexcitons [1-3], the red and blue shifts of exciton bands at the picosecond pumping [4-6] , the Autler-Townes effect on the biexcitons in the crystals CuCl [7-10] were investigated by the use the pump-probe method. In theoretical investigations [8-14] the different aspects of the pump-probe method were considered for the high density system of excitons and biexcitons. The dielectric susceptibilities were investigated in the crystal like CuCl in the case, when the strong pump pulse acts in the region of M-band of luminescence and testing was made by two-photon light absorption with the biexciton excitation [8,9]. It was investigated in [14] the mechanism, when both pulses (pump and probe) act in the exciton region of spectrum taking into account the elastic exciton-exciton interaction. It was shown, that the density of excitons and the susceptibility of the semiconductor discover the bistable behavior depending on the intensity and frequency of pump pulse. Besides it for the definite values of the frequency and intensity of pump pulse the imaginary part of the susceptibility has the negative values, what evidenced about the possibility of the amplification of a weak pulse. Here we present the results of investigation of the optical properties of semiconductor taking into account the elastic exciton-exciton interaction in the case when the strong pump and weak probe pulses act in the exciton range of spectrum.

II. THE MAIN EQUATIONS

We suppose that the monochromatic wave (pump pulse) of coherent laser radiation with the amplitude E_0 and frequency ω_l incidents on the front end of the crystal, where $\omega_l \approx \omega_0$. Here ω_0 is the exciton selffrequency. Just the weak (probe) wave with amplitude E and frequency $\omega \approx \omega_0$ incident on that point of the crystal (Fig. 1).

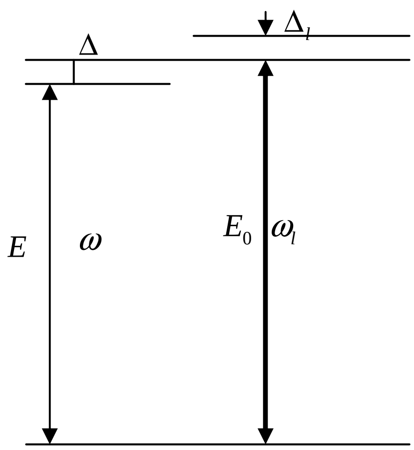

Figure 1. The scheme of levels and optical transitions

The photons of the first pulse excite the excitons and essentially change the energy spectrum of the semiconductor and the photons of the second pulse test these changes in the exciton range of spectrum. At high level of excitation the processes of exciton–exciton interaction begin to manifest itself. The Hamiltonian of interaction of excitons between each other and excitons with photons in the resonant approximation has the form:

$$H_{\text{int}} = \frac{1}{2}\hbar\nu a^+ a^+ aa - \frac{1}{2}\hbar g(a^+ E_0^+ e^{-i\omega_l t} + E_0^- e^{i\omega_l t} a) - \frac{1}{2}\hbar g(a^+ E^+ e^{-i\omega t} + E^- e^{i\omega t} a) \quad (1)$$

where a is the amplitude of the exciton wave of polarization, g is the exciton-field coupling, ν is the constant of the elastic exciton–exciton interaction, $E_0^+(E_0^-)$ and $E^+(E^-)$ is the positive (negative)-frequency part of the fields.

Using (1) we obtain the Heisenberg (material) equations for the amplitude a of the exciton wave:

$$i\dot{a} = (\omega_0 - i\gamma)a + \nu a^+ aa - \frac{1}{2}gE_0^+ e^{-i\omega_l t} - \frac{1}{2}gE^+ e^{-i\omega t}, \quad (2)$$

where γ is the phenomenological dumping parameter, which describe the dumping of coherent excitons. We can define the response of the system in all orders of the perturbation theory on E_0 and in the first order on the amplitude E of the probe field. We look for the solution of eq. (2) in the form

$$a = \frac{1}{2}\left(a_0 e^{-i\omega_l t} + Ae^{-i\omega t} + Be^{-i(\omega - 2\omega_l)t} + c.c\right), \quad (3)$$

where a_0, A, B are the amplitudes of the respective waves. Inserting (3) in (2) in the lowest orders of the perturbation theory on the amplitudes A and B we obtain the system of equations:

$$\begin{aligned} i\dot{a}_0 &= -gE_0^+ - \left(\omega_l - \omega_0 + i\gamma - \frac{3}{4}\nu|a_0|^2\right)a_0, \\ i\dot{A} &= -gE - \left(\omega - \omega_0 + i\gamma - \frac{3}{4}\nu|a_0|^2\right)A + \frac{3}{4}a_0^2 B, \quad (4) \\ i\dot{B} &= -\left(\omega - 2\omega_l - \omega_0 + i\gamma - \frac{3}{4}\nu|a_0|^2\right)B + \frac{3}{4}a_0^2 A. \end{aligned}$$

Further we introduce the resonance detuning Δ and Δ_l for the probe and pump fields and the expression for the exciton density

$$\Delta = \omega - \omega_0, \Delta_l = \omega_l - \omega_0, n_0 = |a_0|^2 \quad (5)$$

and normalized quantities:

$$z = \frac{\nu n_0}{\gamma}, \delta = \frac{\Delta}{\gamma}, \delta_l = \frac{\Delta_l}{\gamma}, \chi_0 = \frac{\hbar g^2}{\gamma}, \tau = \frac{t}{\gamma}. \quad (6)$$

Using (6) we obtain the following system of equations:

$$\begin{aligned} i\dot{a} &= -F_0 - (\delta_l + i - \frac{3}{4}|a|^2)a, \\ i\dot{A} &= -F - (\delta + i - \frac{3}{2}|a|^2)A + \frac{3}{4}a^2 B^*, \\ i\dot{B} &= -(2\delta_l - \delta + i - \frac{3}{2}|a|^2)B + \frac{3}{4}a^2 A^*. \end{aligned}$$

The solution of the first equation in (7) gives us the time evolution of the density of coherent excitons under the action of the pump field $F_0 = \frac{E_0}{E_s}$, where $E_s^2 = \gamma^3 / \nu g^2$. The polarisation of the semiconductor on the frequency of the probe pulse we can obtain in the first order of perturburbation theory in the form: $P^+ = \frac{1}{2}\hbar ga$ .Using this expression we can derive the formal expression, which describes the time evolution of the complex dielectric susceptibility in form $\chi(\tau) = A(\tau)/F(\tau)$, where $F(\tau) = \frac{E(\tau)}{E_s}$ is the field of probe pulse.

III. DISCUSSION OF RESULTS

Let's discuss the results of numerical integration of system of the equations (7) when the pump pulse incidents at the initial moment of time. The first equation in (7) describes dependence on time of concentration of excitons and does not linked on two other equations. Concentration of particles essentially depends on the resonance detuning of pump pulse δ_l and the value of the field f_0 of the pump pulse. We have obtained the multivalued dependence between concentration of excitons and amplitude of radiation of a pump pulse in a crystal in a stationary regime. In a non-stationary regime the behavior becomes complicated. On Fig. 2a the dependences of concentration of excitons z on amplitude of radiation f_0 and τ is presented for the rectangular pulse, at the critical value detuning $\delta_l = \sqrt{3}$. In behavior of concentration of excitons the oscillatory regime exists, which is more strongly for more intensive of incident radiation. The oscillatory regime weakens when time increase.

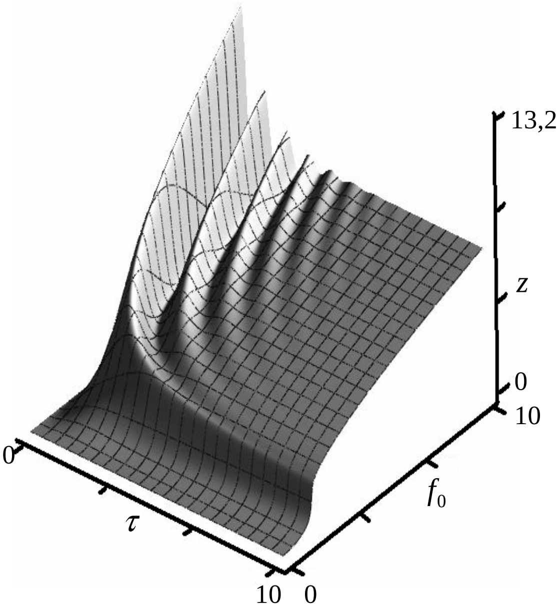

Figure 2. Time evolution of the normalized exciton density z for the different values of the field to of rectangular pump pulse for $\delta_l = \sqrt{3}$

On Fig. 3 the dependences on time τ and amplitude of radiation f_0 imaginary components of susceptibility χ'' are presented value from detuning resonance of a probe pulse $\delta = -5$. In behavior χ'' the oscillatory regime is observed at increase of f_0. Thus χ'' during the certain time intervals accepts negative values that testifies the possibility of strengthening of a probe pulse. In a non-stationary regime the interval of values f_0 extends, at which χ'' accepts the negative values.

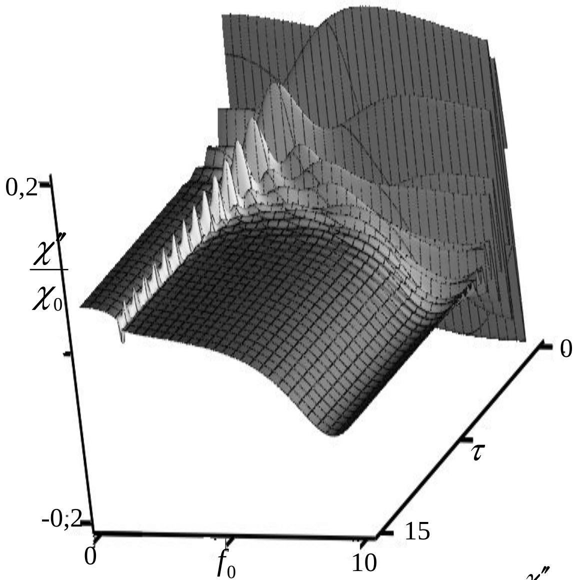

Figure 3. Time evolution of the susceptibility $\dfrac{\chi''}{\chi_0}$ for different values of the field to of pump pulse for $\delta_l = \sqrt{3}$ and $\delta = -5$

It is necessary to note that there is a small interval of values f_0, at which oscillatory regime observes. In a stationary limit the system (7) passes in following system of the equations.

$$
\begin{aligned}
i\dot{A} &= -F - (\delta + i - \frac{3}{2}|a_s|^2)A + \frac{3}{4}a_s{}^2 B^*, \\
i\dot{B} &= -(2\delta_l - \delta + i - \frac{3}{2}|a_s|^2)B + \frac{3}{4}a_s{}^2 A^*.
\end{aligned}
\tag{8}
$$

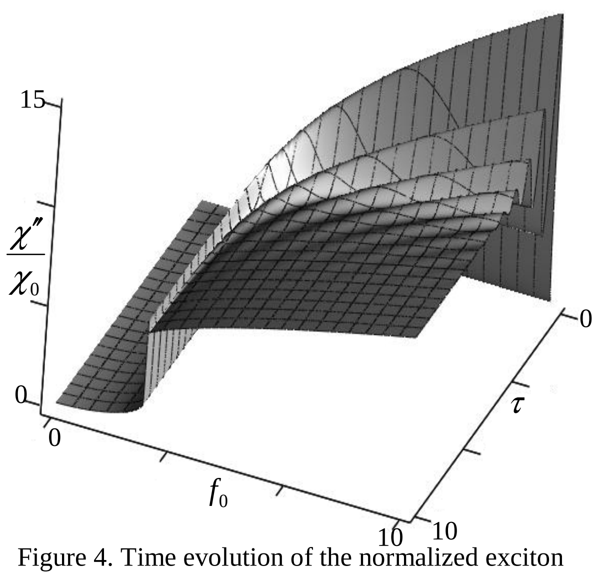

Figure 4. Time evolution of the normalized exciton density z for the different values of the field to of rectangular pump pulse for $\delta_l = 3$

The increase of detuning of the resonance (Fig. 4-5) leads to the jump increase in behavior of concentration on excitons for a long times.

Amplitude of oscillations increases for the growth f_0. As for the absorbing components of a susceptibility that detuning increase has led to strengthening of an oscillatory regime on the resonant values of f_0.

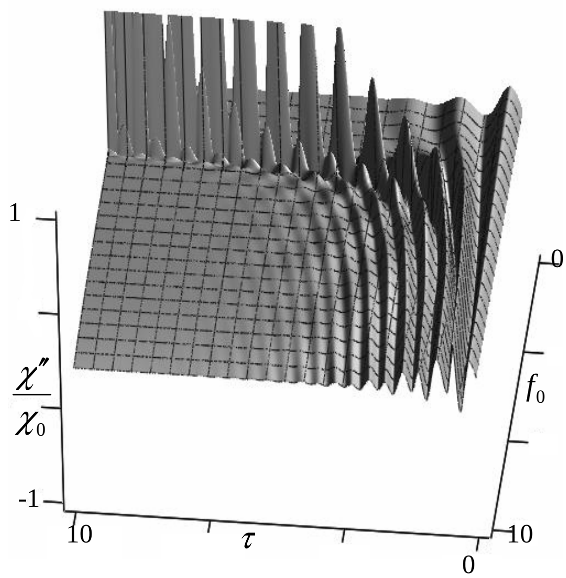

Figure 5. Time evolution of the susceptibility $\dfrac{\chi''}{\chi_0}$ for different values of the field to of pump pulse for $\delta_l = 3$ and $\delta = -5$

We have investigated also the stability of the obtained solution.

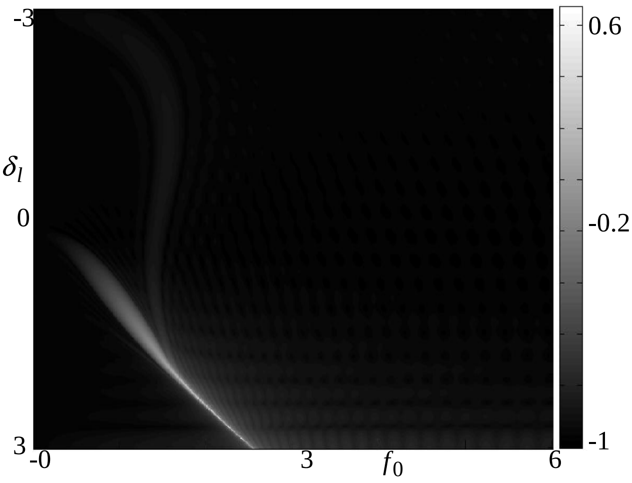

Figure 6. Exponential coefficient of Lyapunov for the different values resonance detuning and values of the field to of pump pulse.

On Fig 6. the exponential coefficient of Lyapunov for the different values resonance detuning and values of the field to of pump pulse is presented. It is visible that the exponential coefficient accepts positive values in narrow area of change of values of a field and resonance detuning. Just at these values the strong oscillatory mode in behavior absorbing susceptibility components was also observed.

Thus the arising resonant oscillation are a consequence of non-stability of system at these values of a field pump and resonance detuning.

On Fig. 7 a the dependences of concentration of excitons on z amplitude of radiation f_0 and τ is presented for the Gaussian pulse $f_0(\tau) = e^{-\frac{(\tau-\tau_0)^2}{T^2}}$. In behavior of concentration of excitons the oscillatory regime exists, which is more strongly on the forward front of a Gaussian pulse.

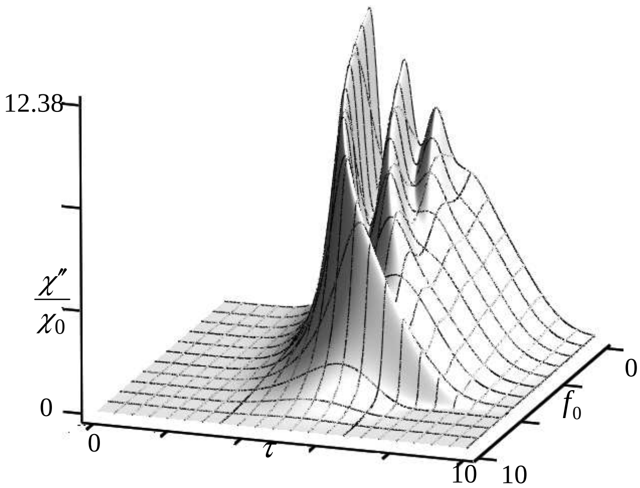

Figure 7. Time evolution of the normalized exciton density z for the different values of the field to of rectangular pump pulse for $\delta_l = 3$ in case of a Gaussian impulse $\tau_0 = 5$, $T = 2$

On the back front of a Gaussian impulse the oscillatory regime is absent.

On Fig. 8 the dependences on time τ and amplitude of radiation f_0 imaginary components of susceptibility χ'' from Gaussian pulse are presented.

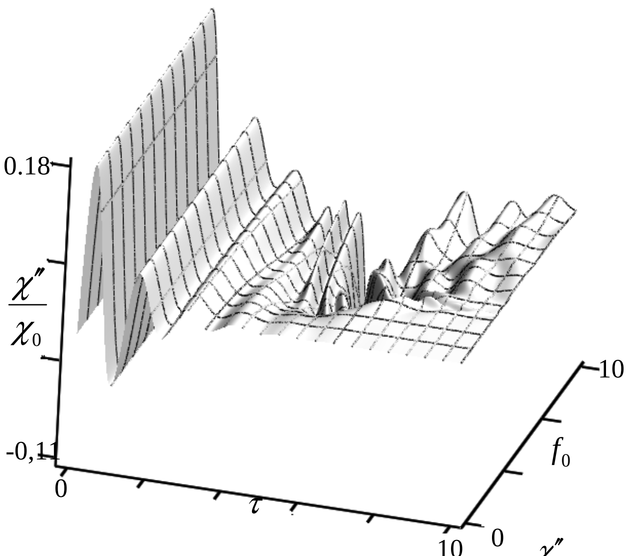

Figure 8. Time evolution of the susceptibility $\frac{\chi''}{\chi_0}$ for different values of the field to of pump pulse for $\delta_l = 3$ and $\delta = -5$ in case of a Gaussian impulse $\tau_0 = 5$, $T = 2$

In behavior χ'' the difficult oscillatory regime is observed on the forward front of a Gaussian pulse. The resonant values of f_0 a rating are shown and in this case.

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