INTERACTION OF SUPERSHORT LIGHT PULSES WITH THIN SEMICONDUCTOR FILMS IN EXCITON RANGE OF SPECTRUM

P.I. Khadzhi[a], I.V. Belousov[a*], D.A. Markov[b], and A.V. Corovai[b]

[a]*Institute of Applied Physics, Academy of Sciences of Moldova, 5, Academiei str., MD-2028, Chisinau, Republic of Moldova*
[2]*Dniester State University, 128, 25 October str., MD-3300, Tiraspol, Republic of Moldova*
[*]*E-mail: igor.beloussov@phys.asm.md*
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Abstract

Taking into account the exciton-photon and elastic exciton-exciton interactions we investigated peculiarities of transmission of supershort light pulses by thin semiconductor films. We predict the appearance of time dependent phase modulation and dynamical red and blue shifts of transmitted pulse.

1. Introduction

Studies of the unique optical properties of thin semiconductor films (TSF) induce a raised interest by virtue of great prospects of the practical applications. The nonlinear relation between the field of an electromagnetic wave propagating through a TSF and polarization of the medium gives rise to a number of interesting physical effects both under the stationary and nonstationary excitation. It is very important that the TSF has the property of the optical bistability in the transmitted and reflected light wave without any additional device. There were studied the peculiarities of the nonstationary interaction of the supershort pulses (SSP) of laser radiation with TSF, which consists of two- and three-level atoms [1-3]. A number of new results was obtained in the investigation of the TSF transmission taking into account the effects of exciton-photon interaction, the phenomenon of exciton dipole momentum saturation, the optical exciton-biexciton conversion, the single-pulse and two-pulse two-photon excitation of biexciton from the ground state of the crystal [4-8]. The new possibilities of ultraspeed control by the transmission (reflection) of TSF were predicted, which can have the great prospects of their utilization in the optical information processing systems. Therefore the further investigation of the peculiarities of the TSF transmission (reflection) in the exciton range of spectrum at high level of excitation is the problem of high priority.

2. Basic equations

Below we present the main results of the theoretical investigation of nonlinear optical properties of the TSF in the conditions of the generation of high density excitons by the supershort pulse of the resonant laser radiation with the amplitude of the electric field E_i and the frequency ω being in resonance with the exciton self-frequency ω_0, normally incidents on the TSF with the thickness L, which is much smaller than the light wave length λ, but much larger than the exciton radius a_0. In the frame of these conditions the elastic exciton-exciton interaction is the main mechanism of nonlinearity. We consider the process of pulse transmission through the TSF taking into account the exciton-photon and elastic exciton-exciton interactions. We suppose that only one macrofilled mode of the coherent excitons and photons exists. We handle the problem using the Maxwell equations for the field and the Heisenberg (material) equation for the amplitude of the exciton wave of polarization. We proceed from the Keldysh equation [9] for the amplitude a of the exciton wave

$$i\hbar\frac{\partial a}{\partial t}=-\hbar(\Delta+i\gamma)a+g|a|^2a-\frac{d_{ex}}{\sqrt{v_0}}E^+, \quad (1)$$

where $\Delta=\omega-\omega_0$ is the resonance detuning, γ is the damping constant of the excitons, g is the constant of the elastic exciton-exciton interaction, d_{ex} is the exciton dipole momentum transition from the ground state of the crystal, v_0 is the volume of the unit cell of the crystal, E^+ is the positive-frequency component of the electric field of the wave propagating through the TSF. The parameters d_{ex} and g contained in (1) are defined by the expressions

$$d_{ex}=(\varepsilon_b v_0\hbar\omega_{LT}/4\pi)^{1/2}, \qquad g=(26/3)\pi I_{ex}a_0^3, \quad (2)$$

where ε_b is the background dielectric constant, a_0, ω_{LT} and I_{ex} are the radius, longitudinal-transversal frequency and the binding energy of the exciton correspondingly. Equation (1) for the bulk crystal corresponds to the mean field approximation, the applicability of which was considered in [9, 10]. Following [1-8] the boundary conditions for the tangential components of the fields of the propagating pulses, we can obtain the electromagnetic relationship between the fields of incident E_i, transmitted E_t, and reflected E_r waves and the amplitude a of the exciton wave of polarization in the form:

$$E_t^+=E_i+i\frac{2\pi\omega L}{c}\frac{d_{ex}}{\sqrt{v_0}}a, \qquad E_t^+=E_i+E_r^+, \quad (3)$$

where L is the film thickness, c is the light velocity in vacuum. We present the macroscopic amplitudes as the products of slowly varying in time envelopes and fast oscillating exponential functions with the frequency ω. By introducing the normalized quantities

$$A=\sqrt{n_0}\,a,\ F_t^+=E_t^+/E_{eff},\ F_i=E_i/E_{eff},\ \tau=t/\tau_0,\ \delta=\Delta\tau_0,\ \Gamma=\tau_0\gamma, \quad (4)$$

we can rewrite equations (1) and (3) in the form

$$i\frac{\partial A}{\partial\tau}=-(\delta+i\Gamma)A+|A|^2A-iA-F_i, \quad (5)$$

$$F_t^+=F_i+iA,\ F_r^+=iA, \quad (6)$$

where the characteristic time parameter τ_0 of the TSF response, the effective field strength E_{eff}, and the exciton density n_0 are defined by the expressions

$$\tau_0^{-1}=2\pi\omega L d_{ex}^2/(c\hbar v_0),\ n_0=\hbar/(g\tau_0),\ E_{eff}=\hbar\sqrt{n_0v_0}/(d_{ex}\tau_0). \quad (7)$$

Let us estimate the values of parameters for CdS-like TSF with the thickness $L=10^{-6}cm$, using $\varepsilon_b=9{,}3$, $v_0=1{,}25\cdot10^{-22}cm^{-3}$, $a_0=2{,}8nm$, $\hbar\omega_{LT}=1{,}9meV$, $I_{ex}=29meV$, $\gamma=10^{11}s^{-1}$ [11-15]. We have obtained $\Gamma=5{,}7\cdot10^{-2}$, $d_{ex}=0{,}53\cdot10^{-18}erg^{1/2}cm^{3/2}$, $g=2{,}8\cdot10^{-32}erg\cdot cm^3$, $\tau_0=5{,}7\cdot10^{-13}s$, $n_0=6{,}3\cdot10^{16}cm^{-3}$, $E_{eff}=2{,}8\cdot10^3\,V/cm$, which corresponds to the intensity $10^3\,W/cm^2$.

Presenting the functions $A(\tau)$ and $F_t^+(\tau)$ as a sum of the real and imaginary parts: $A=x+iy$, $F_t^+=F+iG$ and supposing that the envelope of the incident pulse F_i is a real function of time, we can write equations (5)-(6) in the form

$$\dot{x}=-(1+\Gamma)x-(\delta-z)y, \quad (8)$$

$$\dot{y}=-(1+\Gamma)y+(\delta-z)x+F_i, \quad (9)$$

$$F = F_i - y\,,\;\; G = x\,, \tag{10}$$

where $z = x^2 + y^2$ is the normalized density of excitons, for which we can obtain the equation too

$$\dot{z} = -2(1+\Gamma)z + 2F_i y\,. \tag{11}$$

We see that the amplitude of transmitted wave through TSF (as well as the amplitude of exciton polarization) is the phase-modulated function of time. In the linear approximation the solution for the function $A(\tau)$ is the damped function of time

$$A = \frac{iF_0}{1+\Gamma-i\delta}\left(1-e^{-(1+\Gamma-i\delta)\tau}\right), \tag{12}$$

where the damping constant is equal to $1+\Gamma$. In (12) the exponentially damping factor is conserved even though $\Gamma = 0$, i.e., if the exciton state does not decay. This is due to the fact that the film is the open system and the emergence of the radiation presents the additional "dissipative" mechanism in the system of excitons. In the limit $\Gamma << 1$ ($\gamma << \tau_0^{-1}$) the real dissipation due to the outflow of excitons from the coherent macrofilled mode to the incoherent one is not accomplished.

From (8)-(11) we obtain the dependence of the stationary density of excitons z_s on the field amplitude $F_i = F_0 = const$ of the rectangular incident pulse. The dependence is defined by the solution of the cubic equation

$$z_s\left[(z_s-\delta)^2 + (1+\Gamma)^2\right] = F_0^2\,. \tag{13}$$

For the detuning $\delta < \delta_c = \sqrt{3}(1+\Gamma)$ the density of excitons monotonously increases when the amplitude F_0 of the incident pulse increases. The dependence $z_s(F_0)$ in this case is the nonlinear, but single-valued one. At high level of excitations the dependence $z_s(F_0)$ is the nonlinear, multivalued, and characterized by the hysteresis when $\delta > \delta_c$. For the values of the field amplitude $F_\pm$ the jumps appear in the behavior of the function $z_s(F_0)$ at $z = z_\pm$, where

$$z_\pm = \frac{1}{3}\left[2\delta \pm \sqrt{\delta^2 - 3(1+\Gamma)^2}\right], \tag{14}$$

$$F_\pm^2 = \frac{2}{27}\left[\delta\left(\delta^2 + 9(1+\Gamma)^2\right) \mp \left(\delta^2 - 3(1+\Gamma)^2\right)^{3/2}\right]. \tag{15}$$

For the critical resonance detuning $\delta = \delta_c = \sqrt{3}(1+\Gamma)$ we obtain $z_c = 2(1+\Gamma)/\sqrt{3}$ and $F_c = z_c^{3/2}$. The cyclic change of the amplitude F_i of incident pulse results in the jump-like change of the exciton density in film forming the hysteretic behavior for $F_+ < F_0 < F_-$. The shift of the exciton level for the increasing excitation intensity is the main physical reason of the hysteretic dependence $z(F_0)$. An investigation of these solutions concerning the stability upon the small deviation from the stationary values leads to a conclusion that the instability of the stationary state for z_s takes place in the range $z_- < z < z_+$, which corresponds to the middle part of the hysteretic curve $z_s(F_0)$. The system of equations (8)-(11) has three stationary solutions, two of which are the specific points such as stable node (for $\delta/3 < z < \delta$) or stable focus (for $z < \delta/3$ and $z > \delta$) and the third point is a saddle. The phase portrait of the dynamic system (8)-(9) in the region of the threevaluedness is presented in Fig. 1.

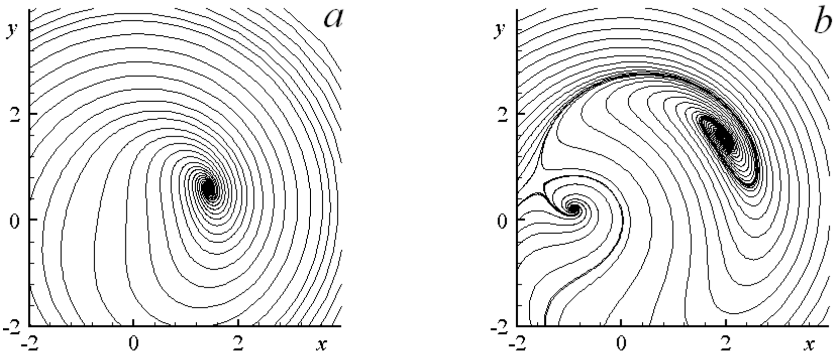

Fig. 1. The phase portrait of the dynamical system under the investigation for the detunings $\delta = 0$ (a) and $\delta = 5$ (b).

3. Discussion of results of numerical solutions

We now discuss the peculiarities of the nonstationary behavior of the system supposing that at the initial moment of time ($\tau = 0$) the crystal was in the ground state, i.e. $x|_{\tau=0} = y|_{\tau=0} = z|_{\tau=0} = 0$. The behavior of the system is defined by the level of excitation and by the detuning. Let consider at first the case of exact resonance: $\delta = 0$. Let the rectangular pulse with the amplitude $F_i = F_0 = const$ incident on the TSF. In Fig. 2 we presented the time evolution of the exciton density in the TSF and the amplitude of transmitted pulse $|F_t|$ for the different values of the field amplitude F_0 of the incident pulse. At low level of excitation the exciton density monotonously increases in time: $z = F_0^2/(1+\Gamma)^2 \times \left(1 - e^{-(1+\Gamma)\tau}\right)^2$ and at long times ($\tau >> 1$) it reaches the stationary value $z_s \approx F_0^2$. Exciton density at the initial stage of evolution accelerates when the level of excitation increases, the small-amplitude oscillations of the density with a long period appear. The further increase of the amplitude F_0 of the incident pulse leads to a sharp increase of the exciton density at the initial stage of the evolution and to the pronounced oscillations of the exciton density. The amplitude of oscillations increases with F_0 and monotonously decreases in time for fixed F_0. As for the period of oscillations of exciton density, it monotonously decreases when the level of excitation increases. Then the oscillations of exciton density gradually decay and the steady state is reached with the exciton density z_s (Fig. 2a).

The obtained peculiarities of the behavior of the function $z(t)$ determine the peculiarities of the time evolution of the field amplitude $|F_t|$ of transmitted radiation. At low level of excitation the field amplitude $|F_t|$ monotonously decreases in time and approaches the stationary transmission. When $\Gamma = 0$ the TSF is closed in transmission and all incident radiation is reflected. In this limit the TSF can play role of an ideal mirror for SSP with the duration $\tau_{pulse} << \Gamma^{-1}$. When the level of excitation increases the week pronounced oscillations with a long period appear after initial decrease of the function $|F_t(\tau)|$. The further increase of the incident amplitude F_0 leads to the increase of the rate of a change of the transmitted pulse at the initial evolution stage and to appearance of a well-pronounced oscillations of the film transmittance. In this case the period of oscillations decreases and the amplitude of oscillations increases with the increase of F_0 and even can happen that $|F_t| > F_0$ in the peak of the first maximum (Fig. 2d). It is due to the fact that the field of transmitted pulse is the sum of the field of incident pulse and the secondary field, which is generated by the exciton polarization. If both fields are changed in phase then the amplitude of transmitted pulse appears to be more than the incident amplitude. For the fixed F_0 the amplitude of oscillations monotonously decreases in time and the steady state transmission with the amplitude $|F_t| \approx F_0$ is set.

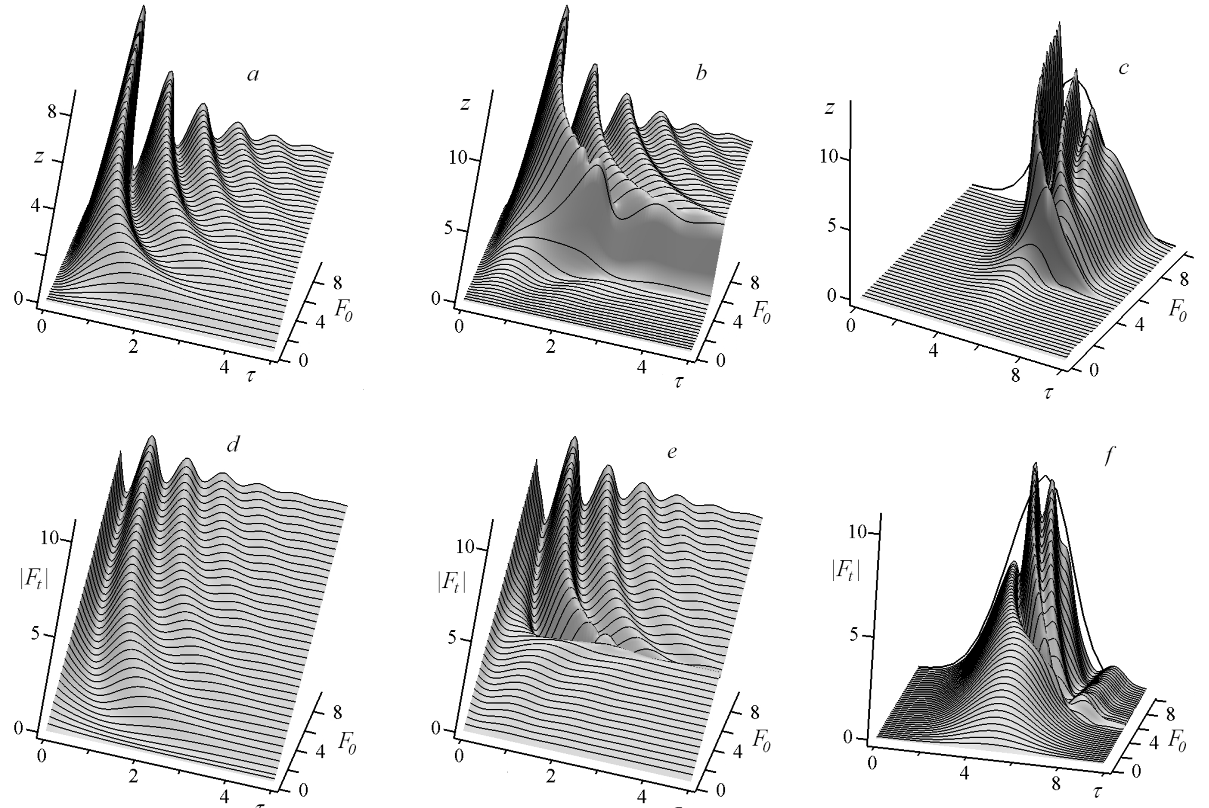

Fig. 2. The time evolution of the exciton density z (a, b, c) and amplitude $|F_t|$ of transmitted pulse (d, e, f) for the different values of the amplitude F_0 of incident rectangular (a, b, d, e) and Gauss (c, f) pulses and for the resonance detuning $\delta = 0$ (a, c, d, f) and $\delta = 5$ (b, e).

Let us discuss now the peculiarities of the time evolution of the system for the detunings $\delta > \delta_c$ (Figs. 2b and 2e). In this case at low level of excitation the exciton density very slowly increases depending both on F_0 and on time. At the initial stage of the time evolution we observe a small increase of the exciton density. The same takes place in transmission too: the amplitude of transmitted pulse at first decreases in time, one or two slightly appreciable oscillations appear and then the system transits into the steady state. But this takes place up to the moment when the amplitude of incident pulse F_0 will be equal to F_-, which corresponds to the jump from one branch to another of the hysteretic dependence of exciton density on F_0 in the stationary state (Figs. 2b and 2e). Immediately after the bifurcation the time evolution of the system at the initial stage exhibits a very speed increase of the exciton density, which then is transformed into a pronounced oscillatory regime of evolution with the great amplitudes and small periods of oscillations. The amplitude of oscillations decreases in time and the system gradually reaches its steady state. The same phenomena take place in the transmission too: at once after bifurcation the regime with pronounced oscillations of the transmitted pulse appears. From the Figs. 2b and 2e we can see that for $\delta > \delta_c$ the transmittance with the amplitude of the first peak much more than the amplitude of incident pulse is possible.

We point out that the change of the amplitude of incident rectangular pulse and the resonance detuning leads to the generation of the complex forms of the transmitted pulses.

Let discuss now the peculiarities of the nonstationary transmission of thin film for the case of normal incidence of Gauss-like pulse $F_i(\tau)=F_0\exp\left(-t^2/T^2\right)$, where F_0 and T are the amplitude and half-width of pulse. In Fig. 2c,f we presented the time evolution of the exciton density z and the amplitude $|F_t|$ of transmitted pulse for different values of the amplitude F_0 of incident pulse. We can see that the shape of the transmitted pulse is changed sufficiently with the change of amplitude F_0. The small peak of the exciton density and transmitted pulse appear at small values F_0. The maximum of transmitted pulse falls at the early stage with respect to the maximum of the incident pulse. When the value of F_0 increases the oscillations of the peak of exciton density appear, which synchronously change the behavior of the transmitted pulse. The peak of this pulse gradually becomes narrow and it shifts to the front tail of the incident pulse, whereas at the moments, which correspond to the centre or to the rear tail of the incident pulse, the new peaks appear one after another, the amplitude and the quantity of which increase with the increase of F_0. Far from the rear tail the weak pulse appears, which is due to the emission of a secondary radiation. The half-widths of new peaks are significantly small in comparison with the half-width of the incident pulse.

In Fig. 3 we presented the time evolution of the amplitude of transmitted pulse for the Gauss-like incident pulse and the transmission function, i.e., dependence of the amplitude of transmitted pulse on the amplitude of incident one. We can point the difference between the stationary and non-stationary transmittances. The incident pulses with a large amplitude lead to the strong

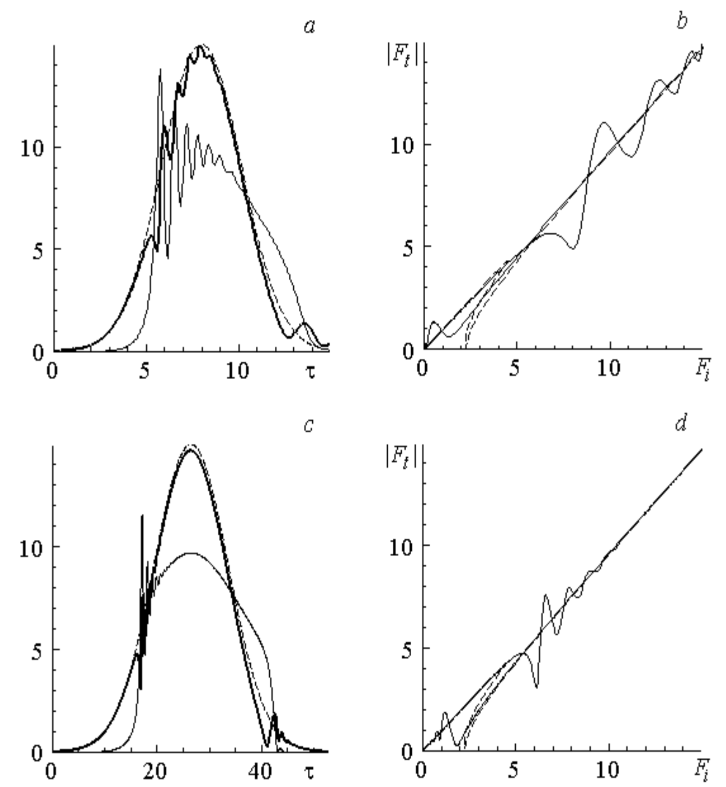

Fig. 3. Time evolution of the exciton density (thin curve) and amplitude of transmitted pulse $|F_t|$ (thick curve) (*a, b*) and the transmittance function $F_t(F_0)$ in comparison with the steady-state transmittance for the Gauss pulses with half-widths T, equal 3 (*a, b*) and 10 (*c, d*).

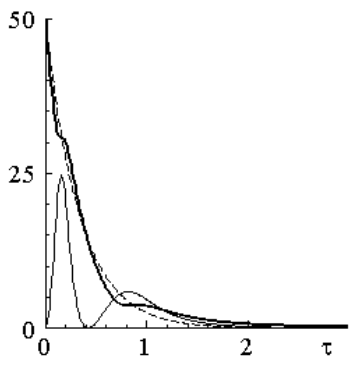

Fig. 4. Time evolution of the exciton density *z* and amplitude F_t (thin curve) of transmitted pulse (thick curve) for the incident pulse of the form $F_i=F_0\exp(-3\tau)$.

oscillations of the transmitted pulse, which represents the significant deviation from the steady state transmittance. Therefore, we can affirm that the steady state transmittance is obtained using the values of the amplitudes of transmitted pulses, which are set after the transient stage, when all oscillations were terminated. The result of the experimental investigations of the transmission function using the supershort Gauss pulses will significantly differ from the theoretical results, which were obtained in the assumption of the stationary transmission. The multivaluedness of the transmittance will be rather due to the transient effects than due to the bistability in the steady-state regime.

As we pointed out we have not succeeded in obtaining the nonlinear solution of eqs. (8)-(11). But for one unique case we have obtained the solution for the exciton density $z(\tau)$ under the action of the incident pulse with the shape $F_i = F_0 \exp(-3\tau)$, which has the form

$$z = \frac{F_0^{2/3}\left(\sqrt{3}-1\right)}{2^{1/3}} e^{-2\tau}\left[1-\mathrm{cn}\left(\sqrt[4]{3}\left(\frac{F_0}{2}\right)^{2/3}\left(1-e^{-2\tau}\right)\right)\right]\cdot\left[1+\left(2-\sqrt{3}\right)\mathrm{cn}\left(\sqrt[4]{3}\left(\frac{F_0}{2}\right)^{2/3}\left(1-e^{-2\tau}\right)\right)\right]^{-1}, \quad (16)$$

where $\mathrm{cn}(x)$ is the elliptic Jacoby function [16] with the modulus $k = \sqrt{2-\sqrt{3}}/2 = \sin(\pi/12)$. We presented the exciton density z and the amplitude F_t of transmitted pulse in Fig. 4. We can see the appearance of the weak oscillations of these functions for the case of a strong pump.

4. Conclusions

In conclusion, we studied the transmittance of thin semiconductor films under the action the supershort pulses of resonant laser radiation taking into account the exciton-photon and the elastic exciton-exciton interaction. We pointed out the differences between stationary and nonstationary transmission and possible experimental difficulties of its observation.

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