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Nonlinear interaction of ultrashort light pulses with a thin semiconductor film under conditions of two-photon exciton-biexciton conversion

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Abstract. Specific features of the transmission of ultrashort resonance laser pulses by a thin semiconductor film are studied taking into account the two-photon exciton-biexciton conversion. It is shown that ultrashort pulses are transformed into shorter solitary pulses or a train of pulses whose duration is determined to a large extent by parameters of a nonlinear medium.

Considerable recent attention has been drawn to the study of physical properties of thin-film structures containing two-level atoms [1 – 7]. Within the framework of simple models of a medium, some new physical phenomena were predicted. The study of the dynamics of propagation of ultrashort laser pulses through resonance thin-film structures is of considerable interest for the search for promising elements for ultra-high-speed information processing. In Refs [8, 9], specific features of transmission and reflection of ultrashort laser pulses by thin semiconductor films (TSFs) in the excitonic region of the spectrum were studied. Note that the interaction of TSFs with light has also been studied in the case of two-photon excitation of a system of two- and three-level atoms [5] and biexcitons from the ground state of a crystal [9].

Here, we theoretically study the nonstationary nonlinear transmission of ultrashort resonance laser pulses by a TSF under conditions of two-photon optical exciton-biexciton conversion in the region of the M band of radiative biexciton recombination [10]. It is known [10] that the two-photon biexciton excitation and the optical exciton-biexciton conversion are characterised by giant oscillator strengths, which can favour a more pronounced manifestation of nonlinear optical effects.

Let an ultrashort laser pulse with the electric field envelope $E_{\mathrm{i}}(t)$, which slowly varies in time, and frequency ω be normally incident on a TSF with thickness L, which is of the order of or smaller than the wavelength of light λ. The pulse duration T (measured at half maximum) is assumed to be much smaller than the exciton and biexciton relaxation time, but much greater than the oscillation period of the wave field. The portion of a pulse transmitted through the TSF changes the optical properties of the film and has a certain effect on the passage of the remaining portion of the incident pulse, which results in the substantial deformation of the pulse shape.

The problem consists in determining the shape of the transmitted (reflected) pulse for the given shape of the incident pulse. The simplest way to solve it is to use the semiclassical approach. The photons of the pulse propagating through the film convert excitons into biexcitons or induce radiative biexciton recombination with the formation of free excitons due to the two-photon exciton-biexciton conversion.

The interaction of excitons and biexcitons with light is described by the Hamiltonian [10, 11]

$$\hat{H} = -\hbar\Psi\left(a^+bE^-E^- + b^+aE^+E^+\right), \tag{1}$$

where Ψ is the two-photon interaction constant; $a(b)$ is the amplitude of the exciton (biexciton) wave of polarisation of the medium; and E^+ (E^-) is the positive (negative) frequency component of the wave field (the sign + at a and b means Hermitian conjugation). Note that Hamiltonian (1) has been earlier used in Ref. [11] to study the specific features of two-photon lasing in the region of the M band, which has been predicted by Haken [12]. Below, we will assume that the exciton, biexciton, and photon states are macroscopically filled. Because the optical exciton-biexciton conversion is characterised by a giant oscillation strength, one can observe nonlinear optical effects at moderate intensities of exciting radiation. Because of this, we may ignore the Stark shift of excitons and biexcitons.

Using (1), we can easily derive the Heisenberg equations for the diference of populations $\rho = N - n$ and the transition amplitude $Q^+ = a^+b$, where N and n are the exciton and biexciton concentrations, respectively:

$$\mathrm{i}\dot{\rho} = 2\Psi\left(Q^+E^-E^- - Q^-E^+E^+\right), \tag{2}$$

$$\mathrm{i}\dot{Q}^+ = \Psi\rho E^+E^+. \tag{3}$$

Equations (2) and (3) were obtained for the exact resonance, i.e., $2\omega = \Omega_0 - \omega_0$, where Ω_0 and ω_0 are the natural frequencies of biexciton and exciton states, respectively. Following Refs [6, 7], we can easily obtain from the conditions of conservation of tangential field components at the crystal-vacuum interface, taking into account the polarisation of the medium, the electrodynamic relation

$$t_0\,\frac{\partial E^+}{\partial t} + E^+ = \mathscr{E}_{\mathrm{i}} + \mathrm{i}\alpha Q^+E^+, \tag{4}$$

where $\alpha = 8\pi\hbar\omega\Psi L/c$; $t_0 = \bar{n}L/c$; and $\bar{n}$ is the linear refractive index of the medium.

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Let us assume that the nonzero exciton concentration n_0 was produced in the film before the arrival of the pulse. Assuming that the field envelope $\mathscr{E}_{\rm i}(t)$ of the incident pulse is a real function of time, we represent the transition amplitude Q^+ and the field E^+ in the film (equal to the envelope of the transmitted field) as a sum of real and imaginary parts: $Q^+ = u + {\rm i}v$ and $E^+(t) = \mathscr{E}(t) + {\rm i}F(t)$. Then, in the case of the exact resonance, it follows from (2)–(4) that $u(t) = 0$ and $F(t) = 0$ at an arbitrary moment of time. Therefore, the envelope $\mathscr{E}(t)$ of the transmitted pulse contains no phase modulation.

As a result, Eqs (2)–(4) are simplified and take the form

$$\dot{v} = -\Psi \rho \mathscr{E}^2, \tag{5}$$

$$\dot{\rho} = 4\Psi v \mathscr{E}^2, \tag{6}$$

$$t_0\dot{\mathscr{E}} + \mathscr{E} = \mathscr{E}_{\rm i} - \alpha v \mathscr{E}. \tag{7}$$

By introducing the normalised quantities

$$y = \frac{v}{n_0},\ r = \frac{\rho}{n_0},\ \mathscr{E}_{\rm i}(t) = \mathscr{E}_0 F(t),\ f = \frac{\mathscr{E}}{\mathscr{E}_0},$$
$$\tau = \frac{t}{\tau_0},\ \tau_0^{-1} = \Psi \mathscr{E}_0^2,\ s = \frac{t_0}{\tau_0}, \tag{8}$$

where $\mathscr{E}_0$ is the amplitude of the rectangular incident pulse or the peak value of the envelope of the incident Gaussian pulse, we obtain the system of nonlinear equations

$$\frac{{\rm d}y}{{\rm d}\tau} = -2rf^2, \tag{9}$$

$$\frac{{\rm d}r}{{\rm d}\tau} = 2yf^2, \tag{10}$$

$$s\frac{{\rm d}f}{{\rm d}\tau} = F(\tau) - (1 + \beta y)f \tag{11}$$

(where $\beta = \alpha n_0$) with the initial conditions

$$y|_{\tau=-\infty} = f|_{\tau=-\infty} = 0,\quad r|_{\tau=-\infty} = -1. \tag{12}$$

It follows from (8) that the time τ_0 characterising the film response to external emission is inversely proportional to the square of the incident-pulse amplitude $\mathscr{E}_0$. This result was earlier obtained in [9] in the study of nonstationary transmission of TSFs upon two-photon excitation of biexcitons from the ground state of a crystal.

One can easily obtain from (9), (10) the integral of motion

$$r^2 + y^2 = 1. \tag{13}$$

It is reasonable to use the relations

$$y = \sin\varphi,\quad r = -\cos\varphi \tag{14}$$

and introduce a new function φ with the initial condition $\varphi|_{\tau=-\infty} = 0$. As a result, system (9), (10) is reduced to the simpler form

$$s\dot{f} = F(\tau) - (1 + \beta\sin\varphi)f, \tag{15}$$

$$\dot{\varphi} = 2f^2. \tag{16}$$

One can see from (15), (16) that the change in φ by $2\pi k$ ($k = 1, 2, \ldots$) does not change the form of equations. Therefore, the phase trajectory of system (15), (16) in the plane f, φ, is a periodic function of φ, with the period 2π for a rectangular pulse (Fig. 1). In turn, system (15), (16) can also be represented in the form of the integro-differential equation

$$s\dot{f} = F(\tau) - f\left\{1 + \beta\sin\left[2\int_0^\tau f^2(\tau'){\rm d}\tau'\right]\right\}. \tag{17}$$

Thus, the rate of variation of the amplitude f of the transmitted pulse is determined by the shape of the envelope of the incident pulse and the transmitted-pulse amplitude itself.

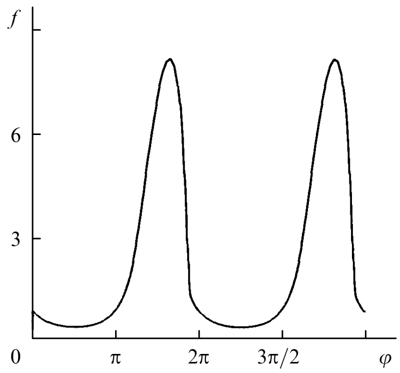

Figure 1. Phase trajectory of the system of equations (15), (16) for $\beta = 1$ and $s = 0.002$.

Consider the specific features of the TSF transmission for a rectangular incident pulse with the amplitude $\mathscr{E}_0$, i.e., $F(\tau) = 1$. After a certain transient stage, the projection of phase trajectories of system (9)–(11) onto the plane f, r passes to the limiting cycle whose parameters are determined by the constants s and β (Fig. 2). Thus, it is expected that the time evolution of the transmitted pulse in the case of a rectangular incident pulse represents a self-pulsation regime.

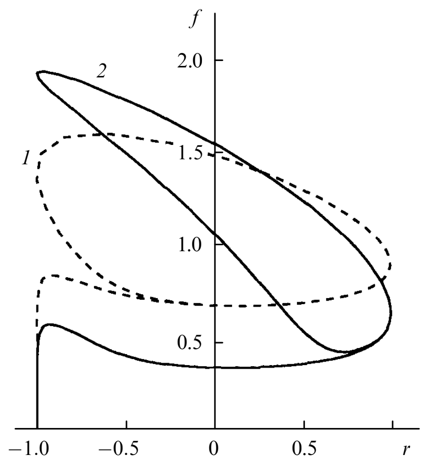

Figure 2. Limiting cycle of the system of Eqns (9)–(11) for $s = 0.2$, $\beta = 0.5$ (*1*) and $s = 1$, $\beta = 2$ (*2*).

Fig. 3 presents the results of the numerical integration of system of equations (9)–(11) for different values of parameters s and β and $F(\tau) = 1$. One can see that after a certain transient stage whose duration is determined by the parameters s, β, and $\mathscr{E}_0$, the system passes to a stable spike transmission regime. The modulation depth of the transmitted signal, the amplitudes and durations of spikes, and the duty factor of the train are determined by the parameters s and β. In the case of a vanishingly small parameter s (Fig. 3c), we obtain the 'lethargic' evolution of the transmitted pulse at the transient stage and, then, an ultrashort spike of large amplitude is formed at a certain moment of time. The duration of spikes is smaller by one or two orders of magnitude than their period.

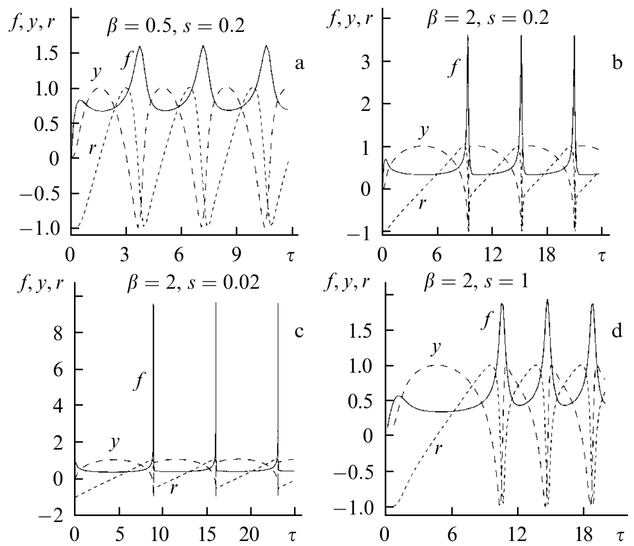

Figure 3. Time evolution of the amplitude f of the pulse transmitted through the TSF (*1*), the polarisation y of the medium (*2*), and the difference r of populations (*3*) for a rectangular incident pulse with $F(\tau) = 1$ for different β and s.

As the parameter s is increased (at a fixed β), the spikes are broadened, their amplitudes and period decrease, and the background intensity increases. An increase in the parameter β at a fixed s also leads to a decrease in the duration of resulting spikes. Each spike arises when inversion reaches a maximum. As a result, inversion rapidly decreases to a minimum and an ultrashort spike is formed. At the moment when the transmitted pulse has the maximum amplitude, inversion vanishes. The polarisation of the medium periodically varies as well. Thus, a TSF can operate as an efficient converter of a short rectangular pulse into a train of shorter pulses.

Fig. 4 presents results of the numerical integration of system (9)–(11) for the case of a short Gaussian pulse with the envelope $F(\tau) = \exp(-\tau^2/T^2)$ incident on the TSF. If the duration T of the incident pulse is sufficiently small, the TSF almost totally reflects it. At a certain critical pulse duration, a solitary ultrashort pulse (a spike) can be formed. The transmitted pulse is shorter by an order of magnitude than the incident pulse and its amplitude is several times greater. One can see from Fig. 4 that the narrow spike is formed at the moment of maximum inversion. The number of generated spikes transmitted through the TSF may increase with increasing duration of the incident pulse (Fig. 4b). This once again suggests that ultrashort pulses incident on a TSF can be transformed into shorter solitary pulses or a train of pulses.

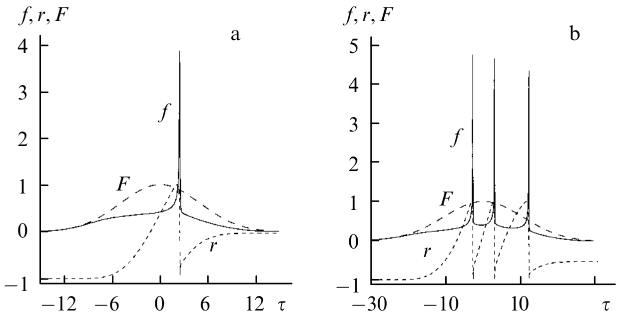

Figure 4. Time evolution of the amplitude f of the pulse transmitted through the TSF (*1*), and the difference r of populations (*2*) for a Gaussian incident pulse with $F(\tau) = \exp(-\tau^2/T^2)$ (*3*) and $T = 7$ (a) and 15 (b) for $\beta = 3$ and $s = 0.05$.

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