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Features of the interaction of ultrashort laser pulses with a thin semiconductor film caused by the generation of excitons and biexcitons

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***Abstract*.** **Nonstationary transmission (reflection) of two time-separated ultrashort laser pulses by thin semiconductor films is studied. Photons of one of these pulses generate biexcitons in the films due to two-photon absorption from the crystal ground state, while photons of the other pulse induce an optical exciton – biexciton conversion. Several basically new effects in nonstationary light transmission (reflection) are found and interpreted.**

Keywords: *exciton, biexciton, ultrashort pulse, transmission.*

1. Introduction

Nonlinear optical effects involving coherent excitons and biexcitons in semiconductors were usually studied using the model of an infinitely extended medium (e.g., [1, 2] and references therein). However, nonlinear optical properties of low-dimensional semiconductor structures, such as thin films, currently evoke an ever increasing scientific and practical interest. A specific relation between the field of an electromagnetic wave passing through a thin semiconductor film (TSF) and the medium polarisation results in a number of interesting physical effects [3 – 11].

Specific features of nonstationary interaction of ultrashort laser pulses with thin films consisting of two-level and three-level atoms [10], and with TSFs, in which excitons and biexcitons are excited [11 – 14], were studied theoretically taking into account coherent quantum transitions and interactions (exciton – photon interaction, optical exciton – biexciton conversion, saturation of the dipole moment of the exciton transition, and two-photon excitation of biexcitons from the crystal ground state). It turned out that an ultrashort laser pulse incident onto a TSF is appreciably modified during its passage through (reflection by) the film: during the characteristic response time, the thin film may transfer to a regime of total internal reflection or become fully transparent; as a result, the transmitted pulse may be split into a sequence of shorter subpulses, and its amplitude, half-width, and other parameters can be changed.

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The functional capabilities of such a film are determined by the amplitude, half-width, and shape of the incident pulse. They are numerous and diversified, as well as the characteristics of steady-state laser-radiation transmission and reflection by TSFs. The results obtained in [1 – 14] concern the study of the interaction of single pulses with TSFs. At the same time, the study of the response of a TSF to two (and more) incident pulses of different amplitudes, frequencies, and half-widths (envelopes), which correspond to different quantum transitions and different types of light interaction with elementary excitations in a medium, is also of significant interest. It is also interesting to investigate the effects of the photon echo type involving two time-separated ultrashort laser pulses at different frequencies.

Below, we present the results of studies of the interaction of two ultrashort laser pulses with TSFs. The frequency of one of the pulses is resonant to the transition frequency in the M-band (in the region of optical exciton – biexciton conversion), while the frequency of the other pulse provides a resonant two-photon excitation of biexcitons from the crystal ground state (Fig. 1). In this case, the first pulse coherently mixes the exciton and biexciton states. This changes the energy spectrum of the semiconductor and substantially affects the character of the interaction of the second pulse with the medium. The results of an experimental study of the so-called Autler – Townes effect on biexcitons are presented in [15]. This effect consists in the splitting of the biexciton (exciton) level under the action of the field of a high-power laser pulse. In this case, photons of the high-power pulse were in resonance with the transition

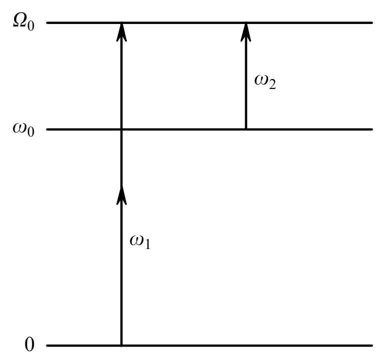

Figure 1. Energy level diagram for an exciton and a biexciton and quantum transitions in the fields of two different pulses with frequencies ω_1 and ω_2.

frequency in the region of the M-band, and a weak probe pulse excited biexcitons from the crystal ground state due to two-photon absorption. A theory of the stationary Autler–Townes effect on biexcitons was developed in [16]. This work uses the same formulation of the problem as in [15, 16], but it is assumed that both pulses are ultrashort and interact with a TSF.

2. Formulation of the problem and basic equations

Let two ultrashort laser pulses be incident normally onto a thin film of a CuCl crystal of thickness L, which is much less than the light wavelength λ. The pulse durations are assumed to be much shorter than the relaxation time of excitons (biexcitons) in the semiconductor. Under such conditions, the exciton (biexciton) relaxation processes are insignificant and the light interaction with excitons and biexcitons is coherent. The Hamiltonian of the interaction of excitons and biexcitons with the fields of both pulses has the form

$$H = -\hbar\sigma\left(a^+ bE_2^- + b^+ aE_2^+\right) - \hbar\mu\left(b^+ E_1^+ E_1^+ + bE_1^- E_1^-\right), \quad (1)$$

where $E_2^+(E_2^-)$ is the positive- (negative-) frequency component of the field of a pulse with a photon frequency ω_2 acting in the region of the M-band; $\omega_2 \simeq \omega_M = \Omega_0 - \omega_0$; ω_M is the frequency of the transition in the region of the M-band; Ω_0 and ω_0 are the eigenfrequencies of the biexciton and exciton transitions, respectively; $E_1^+(E_1^-)$ is the positive- (negative-) frequency component of the field of a pulse with a photon frequency $\omega_1 \simeq \Omega_0/2$ that excites biexcitons from the crystal ground state due to two-photon light absorption; $a(b)$ is the amplitude of the exciton (biexciton) polarisation wave; σ is the optical exciton–biexciton conversion constant [1, 2]; and μ is the two-photon biexciton excitation constant [1, 2]. Since the biexciton binding energy in the CuCl crystal is sufficiently high ($\sim$ 30–40 meV), the photons with the frequencies ω_1 and ω_2 cannot excite excitons and biexcitons from the crystal ground state because of a large resonance detuning. For this reason, Hamiltonian (1) takes into account only the two strongest quantum transitions, as in [15, 16].

Using (1), it is easy to derive Heisenberg (material) equations describing the time-dependent amplitudes of the exciton and biexciton waves of the medium polarisation. Under conditions of an exact resonance where $2\omega_1 = \Omega_0$ and $\omega_2 = \omega_M = \Omega_0 - \omega_0$, these equations take the form

$$\mathrm{i}\dot{a} = -\sigma b E_2^-, \quad (2)$$

$$\mathrm{i}\dot{b} = -\sigma a E_2^+ - \mu E_1^+ E_1^+. \quad (3)$$

Following [11–14], we obtain two equations for the field amplitudes E_1^+ and E_2^+ in the film (the amplitudes of the pulses transmitted through the TSF) as the boundary conditions:

$$E_1^+ = E_{\mathrm{i}1} + \mathrm{i}\alpha_1 E_1^- b, \quad (4)$$

$$E_2^+ = E_{\mathrm{i}2} + \mathrm{i}\alpha_2 a^+ b, \quad (5)$$

where $E_{\mathrm{i}1}$ and $E_{\mathrm{i}2}$ are the field amplitudes (envelopes) of the pulses incident on the TSF, which are considered to be real values; $\alpha_1 = 2\pi\hbar\omega_1\mu L/c$; and $\alpha_2 = 2\hbar\omega_2\sigma L/c$. In addition, we assume that, at the initial moment, the crystal was in the ground state, i.e., excitons and biexcitons were absent in it.

By representing the macroscopic amplitudes as the sum of the real and imaginary components

$$a = u + \mathrm{i}v, \;\; b = z + \mathrm{i}w, \;\; E_1^+ = E_1 + \mathrm{i}F_1, \;\; E_2^+ = E_2 + \mathrm{i}F_2, \quad (6)$$

we can easily obtain that $v(t) = 0$, $z(t) = 0$, $F_1(t) = 0$ and $F_2(t) = 0$ at the exact resonance and zero initial conditions for the material variables ($a|_{t=0} = b|_{t=0} = 0$). Therefore, system of Eqns (2)–(5) is transformed as

$$\dot{u} = -\sigma E_{\mathrm{i}2} w + \alpha_2 \sigma w^2 u, \quad (7)$$

$$\dot{w} = \sigma E_{\mathrm{i}2} u - \alpha_2 \sigma u^2 w + \mu E_{\mathrm{i}1}^2 (1 + \alpha_1 w)^{-2}, \quad (8)$$

$$E_1 = E_{\mathrm{i}1}(1 + \alpha_1 w)^{-1}, \quad (9)$$

$$E_2 = E_{\mathrm{i}2} - \alpha_2 u w. \quad (10)$$

Consequently, the imaginary field components of the transmitted pulses are equal to zero, and, thus, no phase modulation of these pulses takes place.

We failed to derive exact analytical solutions to system of Eqns (7)–(10) at arbitrary $E_{\mathrm{i}1}$ and $E_{\mathrm{i}2}$. From (7) and (8), it is easy to find the relation showing that the sum of the concentrations of excitons $n = u^2$ and biexcitons $N = w^2$ at any moment is determined only by the pump pulse $E_{\mathrm{i}1}(t)$ and is independent of the pump pulse $E_{\mathrm{i}2}(t)$, because the latter transforms excitons into biexcitons and vice versa with their total number maintained at a constant level:

$$u^2 + w^2 = 2\mu \int_{-\infty}^{t} E_1^2(t') w(t') \mathrm{d}t' = 2\mu \int_{-\infty}^{t} E_{\mathrm{i}1}^2(t') [1 + \alpha_1 w(t')]^{-2} w(t') \mathrm{d}t'. \quad (11)$$

3. Discussion

3.1 Transmission of delta- and step-shaped pulses

Consider a number of particular cases of the solution to the system of Eqns (7) and (8) for various shapes of the envelopes $E_{\mathrm{i}1}(t)$ and $E_{\mathrm{i}2}(t)$ of the incident pulses. Suppose that, at the instant $t = 0$, a delta-shaped pump pulse $E_{\mathrm{i}1}(t) = E_{\mathrm{i}1}\delta(t/T)$ and a rectangular pulse $E_{\mathrm{i}2}(t) = E_{\mathrm{i}2}\vartheta(t)$ are applied to the system in the ground state. Here, T is a parameter characterising the half-width of the spread delta-shaped pulse, and $\vartheta(t)$ is the Heaviside function. One can see from (7) and (8) that the role of the delta-shaped pulse is to produce a system of biexcitons with an amplitude $w_0 = \mu E_{\mathrm{i}1}^2 T$ at the instant of time $t = 0$. The exciton density is zero in this case. The system then undergoes an evolution at $t > 0$ only under the action of the pulse $E_{\mathrm{i}2}(t)$. Generally speaking, we may also consider that the pulse $E_{\mathrm{i}2}(t)$ is switched on at an arbitrary instant $t > 0$ after the termination of the pulse $E_{\mathrm{i}1}(t)$, which generated a certain concentration of biexcitons. This situation corresponds to separate and independent actions of two time-separated pulses. Equations (7) and (8) show that, in this case, the time evolution is described by the coupled equations

$$\dot{u} = -\sigma E_2 w, \ \dot{w} = \sigma E_2 u \,. \tag{12}$$

At the initial conditions $w|_{t=0} = w_0 = \mu E_{i1}^2 T$ and $u|_{t=0} = 0$, these equations have the solutions

$$w = w_0 \cos \vartheta_2, \quad u = -w_0 \sin \vartheta_2, \tag{13}$$

where the pulse area ϑ_2 is expressed by the formula

$$\vartheta_2 = \sigma \int_{-\infty}^{t} E_2(t')\mathrm{d}t'. \tag{14}$$

The equation

$$\frac{\partial \psi}{\partial \tau} = F_{i2} + \sin \psi, \tag{15}$$

where

$$\tau = \frac{t}{\tau_0}; \ \psi = 2\vartheta_2; \ F_{i2} = \frac{2E_{i2}}{\alpha_2 \mu^2 T^2 E_{i1}^4}; \ \tau_0^{-1} = \alpha_2 \sigma w_0^2 \tag{16}$$

follows from (10) and (13). Solving Eqn (15) and using relations (13) and (14), one can easily determine the expression for the envelope of the TSF-transmitted pulse $E_2(\tau)$. The system's evolution is determined by the parameter F_{i2}. At $F_{i2} < 1$, we have

$$E_2(\tau) = \frac{\left(1 - F_{i2}^2\right)/(2\sigma\tau_0)}{\cosh\left[\left(1 - F_{i2}^2\right)^{1/2}\tau - \operatorname{artanh}\left(1 - F_{i2}^2\right)^{1/2}\right] - F_{i2}}. \tag{17}$$

The latter relation shows that the envelope of the pulse $E_2(\tau)$ at the instant

$$\tau = \frac{1}{\left(1 - F_{i2}^2\right)^{1/2}} \operatorname{artanh}\left(1 - F_{i2}^2\right)^{1/2} \tag{18}$$

reaches a maximum $E_2^{\max} = (1 + F_{i2})/(2\sigma\tau_0)$. The ratio $E_2^{\max}/E_2(0) = 1 + F_{i2}^{-1}$ increases, as F_{i2} decreases. For $F_{i2} > 1$, the solution for the envelope of the transmitted pulse has the form

$$E_2(\tau) = \frac{F_{i2}^2 - 1}{2\sigma\tau_0} \times \left\{F_{i2} - \sin\left[\left(F_{i2}^2 - 1\right)^{1/2}\tau + \arctan\left(F_{i2}^2 - 1\right)^{-1/2}\right]\right\}^{-1}. \tag{19}$$

Finally, for $F_{i2} = 1$, using (17) and (19), we derive

$$E_2(\tau) = (\sigma\tau_0)^{-1}\left[1 + (\tau - 1)^2\right]^{-1}. \tag{20}$$

From solutions (17)–(20) and Fig. 2, it follows that, for $F_{i2} < 1$, the field amplitude $E_2(\tau)$ of the TSF-transmitted pulse at first rapidly rises, reaches a maximum, and then rapidly decreases. At $\tau \gg 1$, the film transforms into an ideal mirror for rectangular ultrashort pulses. If $F_{i2} > 1$, the film transforms an ultrashort pulse into a train of pulses with shorter durations; i.e., the film transmission has an oscillatory character. The higher the amplitude F_{i2} of the arriving pulse, the shorter the oscillation period $\tau_1 = 2\pi \times (F_{i2}^2 - 1)^{-1/2}$. In this case, the expressions for the minimum and maximum amplitudes of the TSF-transmitted pulse have the form

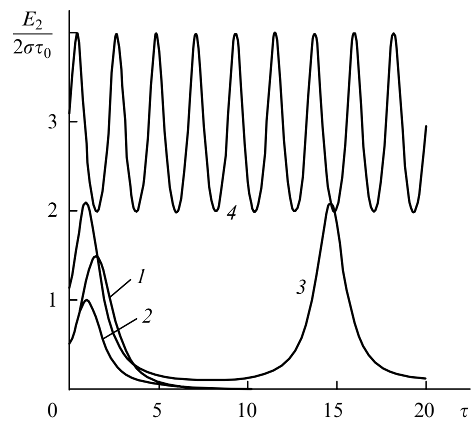

Figure 2. Shapes of the envelopes of the transmitted pulse $E_2(\tau)/(2\sigma\tau_0)$ for a delta-shaped pulse at a frequency in the exciton spectrum region and a step-shaped pulse in the region of the M-band incident onto a TSF. The normalised amplitude of the second pulse is $F_{i2} = 0.5$ (*1*), 1 (*2*), 1.1 (*3*), and 3 (*4*).

$$E_2^{\min} = \frac{F_{i2} - 1}{2\sigma\tau_0}, \ E_2^{\max} = \frac{F_{i2} + 1}{2\sigma\tau_0}. \tag{21}$$

Hence, the closer F_{i2} to unity, the larger the modulation depth of the transmitted pulse. Oscillations of $E_2(\tau)$ in this case are shaped as sharp peaks with large spacings between them. At large F_{i2} ($F_{i2} \gg 1$), oscillations of $E_2(\tau)$ look like a slight ripple on a high background:

$$E_2(\tau) = \frac{F_{i2} + \sin(F_{i2}\tau)}{2\sigma\tau_0}. \tag{22}$$

The ratio of the half-width $\Delta\tau$ of the arising pulses to the period τ_1 is $\Delta\tau/\tau_1 = {}^1/_2 - \pi^{-1} \arcsin F_{i2}^{-1}$.

3.2 Transmission of Gaussian and rectangular pulses

Similar results are obtained in the case where the envelopes of the incident pulses $E_{i1}(t)$ and $E_{i2}(t)$ are not rectangular. Fig. 3 shows the results of a numerical solution of the system of Eqns (7)–(10) for time-separated pulses: a Gaussian pulse $E_{i1}(t) = E_{i1}\exp(-t^2/T^2)$ and a short rectangular pulse $E_{i2}(t) = E_{i2}\vartheta(t - \Delta t)$. Note that the time delay Δt of the pulse $E_{i2}(t)$ with respect to the peak of the pulse $E_{i1}(t)$ exceeds the half-width T. In this case, the expression for the envelope of a TSF-transmitted pulse $E_1(t)$ has the form

$$E_1(t) = E_{i1} \exp\left(-\frac{t^2}{T^2}\right) \times \left\{\frac{3\sqrt{\pi}\alpha_1 \mu E_{i1}^2 T}{2\sqrt{2}}\left[1 - \Phi\left(\sqrt{2}\,\frac{t}{T}\right)\right]\right\}^{-1/3}, \tag{23}$$

where $\Phi(x)$ is the error function [17, 18]. Fig. 3 shows that the amplitude of the transmitted pulse $E_1(\tau)$ is smaller than the amplitude of the incident pulse E_{i1}, and the peak of the transmitted pulse is ahead of the peak of E_{i1}. This is due to the fact that a certain fraction of the energy of the first pulse $E_{i1}(\tau)$ is expended on the generation of a system of biexcitons in the film, whose density increases with the amplitude E_{i1} and half-width T. Since excitons are absent in the system, there exists a population inversion of the exciton and biexciton levels at the transition frequency ω_M (in the region of the M-band). The photons of the second pulse with the envelope $E_{i2}(\tau)$ and frequency $\omega_2 = \omega_M$ hit the film and cause a rapid induced decrease in the density of biexcitons, which recombine with the formation of excitons and photons with a frequency $\omega_2 = \omega_M$. This results in the formation of an ultrashort laser pulse with the envelope $E_2(\tau)$. One can see from Figs 3a – 3c that its width decreases and amplitude rises with an increase in the amplitude (width) of the incident pulse $E_{i1}(\tau)$, and the biexciton concentration is virtually independent of time until the second pulse is incident onto the TSF.

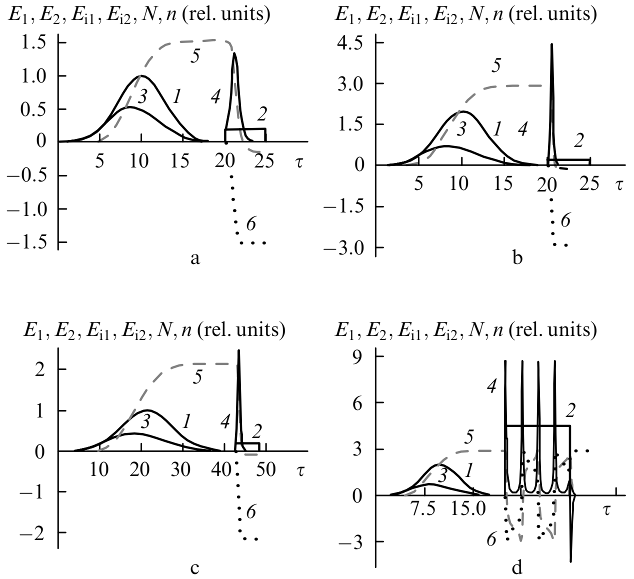

Figure 3. Shapes of the envelopes of the incident pulses: (*1*) a Gaussian pulse $E_{i1}(\tau)$ in the exciton spectrum region and (*2*) a rectangular pulse $E_{i2}(\tau)$ in the region of the M-band; shapes of the envelopes of the corresponding transmitted pulses $E_1(\tau)$ (*3*) and $E_2(\tau)$ (*4*); the time evolution of the concentrations of (*5*) biexcitons N and (*6*) excitons n for (a) $E_{i1} = 1$, $E_{i2} = 0.2$, $T/\tau_0 = 4$; (b) $E_{i1} = 2$, $E_{i2} = 0.2$, $T/\tau_0 = 4$; (c) $E_{i1} = 1$, $E_{i2} = 0.2$, $T/\tau_0 = 8$; (d) $E_{i1} = 2$, $E_{i2} = 4.5$, $T/\tau_0 = 4$.

As the duration and amplitude of the second pulse increase, the generation of a train of ultrashort pulses (spikes) at a frequency $\omega_2 = \omega_M$ becomes possible; thus, the TSF efficiently transforms the incident pulse $E_{i2}(\tau)$ into a train of shorter pulses with a width 10 – 100 times less than that of the incident pulse (Fig. 3d). Note that, the higher the rectangular pulse E_{i2}, the smaller its width and off – duty ratio and the higher the amplitude of each subpulse in the train and the background level about which subpulses oscillate. The situation presented in Fig. 3d corresponds to a low-level background. It should be also noted that the time succession in which pulses are transmitted corresponds to the sequence of their incidence onto the film.

3.3 Transmission of two time-separated Gaussian pulses

Fig. 4 presents the results of the numerical integration of Eqns (7) – (10) for the case where two time-separated Gaussian pulses with different amplitudes and half-widths are successively incident onto a TSF [$E_{i1}(\tau)$ is followed by $E_{i2}(\tau)$]. One can see from Fig. 4a that the pulse $E_{i1}(\tau)$ generates biexcitons and the transmitted pulse $E_1(\tau)$ is, to a certain extent, similar to the incident pulse $E_{i2}(\tau)$. It is of interest that a sharp ultrashort radiation pulse $E_2(\tau)$ at a frequency $\omega_2 = \omega_M$ suddenly arises at the end of the pulse $E_{i1}(\tau)$ trailing edge and at the vanishingly small leading edge of the pulse $E_{i2}(\tau)$. There is an impression that the consequence [the appearance of $E_2(\tau)$] occurs earlier than the cause [the incidence of $E_{i2}(\tau)$ on the film]. The point is that the leading edge with the vanishingly small amplitude of the incident pulse $E_{i2}(\tau)$ causes the rapid induced dumping of the inversion, which was produced by the first pulse, and this process is speeded up as it develops.

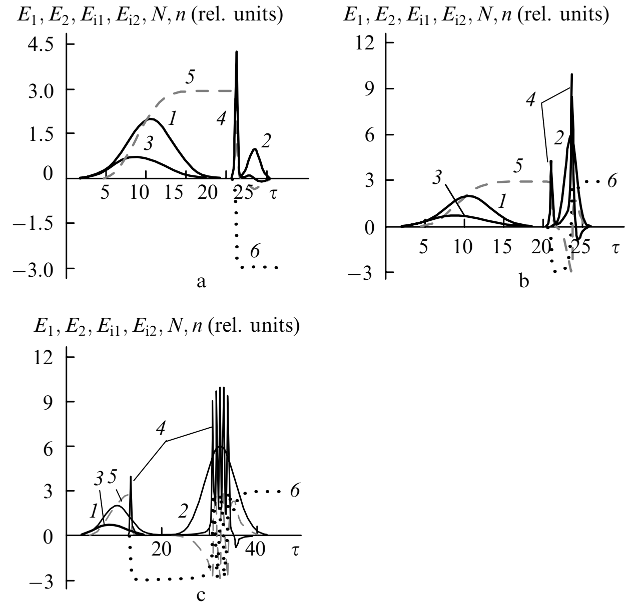

Figure 4. Shapes of the envelopes of the incident Gaussian pulses: (*1*) a pulse $E_{i1}(\tau)$ with a half-width T_1/τ_0 in the exciton spectrum region and (*2*) a pulse $E_{i2}(\tau)$ with a half-width T_2/τ_0 in the region of the M-band; shapes of the envelopes of the corresponding transmitted pulses (*3*) $E_1(\tau)$ and (*4*) $E_2(\tau)$; the time evolution of the concentrations of (*5*) biexcitons N and (*6*) excitons n for (a) $E_{i1} = 2, E_{i2} = 1, T_1/\tau_0 = 4, T_2/\tau_0 = 1$; (b) $E_{i1} = 2$, $E_{i2} = 6$, $T_1/\tau_0 = 4, T_2/\tau_0 = 1$; (c) $E_{i1} = 2$, $E_{i2} = 6, T_1/\tau_0 = 4, T_2/\tau_0 = 4.5$.

Fig. 4b presents similar results for the case where the amplitude of the second incident pulse is several times larger than that in Fig. 4a. Fig. 4b shows that, immediately after the transmission of the first pulse $E_1(\tau)$, biexcitons disappear from the crystal, but excitons appear instead of them. Subsequently, as the amplitude of the incident pulse $E_{i2}(\tau)$ rises with time, it gradually converts excitons into biexcitons. At a certain instant of time, the second radiation peak $E_2(\tau)$ is generated in the vicinity of the $E_{i2}(\tau)$ peak, and the former is shorter than $E_{i2}(\tau)$. After that, the trailing edge of the incident pulse $E_{i2}(\tau)$ passes through the TSF and produces a certain exciton concentration.

Fig. 4c shows similar results for the case where the peak amplitude of $E_{i2}(\tau)$ is the same as in Fig. 4b but the half-width is larger. One can see that the response of the film is somewhat different: as in the previous case, a solitary ultrashort pulse $E_2(\tau)$ is first generated at the leading edge of the incident pulse $E_{i2}(\tau)$; then, five successive ultrashort pulses $E_2(\tau)$ are rapidly generated in the vicinity of the $E_{i2}(\tau)$ peak; and, finally, the trailing edge of the incident pulse produces an exciton polarisation of the medium and is totally reflected by the film. Thus, the response of the film to the incidence of a Gaussian pulse $E_{i2}(\tau)$ in the region of the M-band is determined by the amplitude and half-width of this pulse. Note that, at a smaller amplitude of the pulse $E_{i2}(\tau)$, the radiation at the frequency of this pulse is not transmitted through the film but is totally reflected by it.

3.4 Transmission of rectangular and Gaussian pulses

A very interesting effect, the transmission of pulses in the reverse sequence, is illustrated in Fig. 5. A rectangular pulse $E_{i2}(\tau)$ is incident first, and a Gaussian pulse $E_{i1}(\tau)$ arrives with a certain time delay (of the order of their half-widths). Note that these pulses slightly overlap, although this overlap is very or even vanishingly small. In this case, the pulse $E_{i2}(\tau)$ immediately passes through the TSF, as if it is an absolutely transparent medium, because almost no excitons and biexcitons are present in it. Nevertheless, the small-amplitude initial part of the leading edge of the Gaussian pulse $E_{i1}(\tau)$ induces a very low concentration of biexcitons, which are immediately converted into excitons by photons of the pulse $E_{i2}(\tau)$. After the termination of $E_{i2}(\tau)$, the pulse $E_{i1}(\tau)$ continues to generate biexcitons, whose concentration rapidly rises. In the absence of the field at a frequency ω_2 and at a very low concentration of excitons, biexcitons cannot totally recombine. Therefore, the pulse $E_{i1}(\tau)$ is gradually transmitted through the film, simultaneously continuing to increase the biexciton density. However, as the process develops with time, conditions for dumping the inversion formed in the region of the M-band appear due to the exciton – biexciton inducing in the absence of both photons with ω_2 and ultrashort pulses at the frequency ω_2 at the far trailing edge of the pulse $E_{i1}(\tau)$.

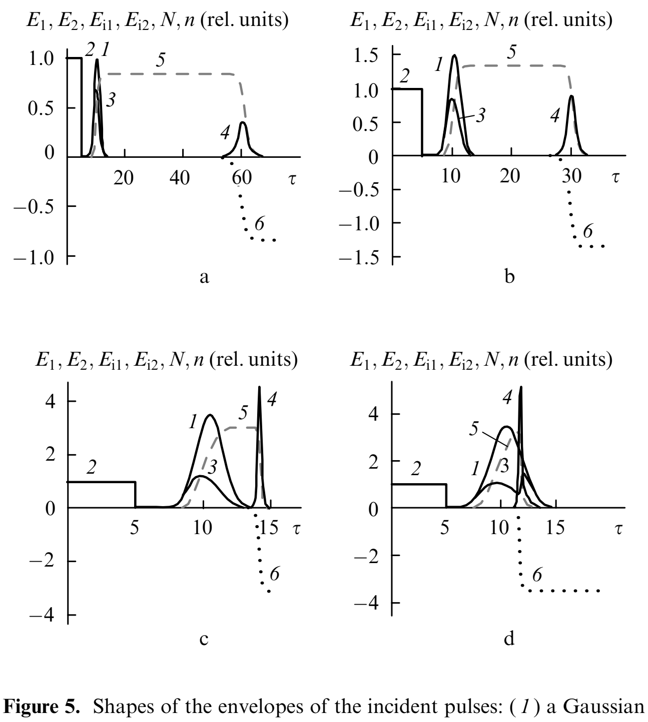

Figure 5. Shapes of the envelopes of the incident pulses: (*1*) a Gaussian pulse $E_{i1}(\tau)$ in the exciton spectrum region and (*2*) a rectangular pulse $E_{i2}(\tau)$ in the region of the M-band; shapes of the envelopes of the corresponding transmitted pulses $E_1(\tau)$ (*3*) and $E_2(\tau)$ (*4*); the time evolution of the concentrations of (*5*) biexcitons N and (*6*) excitons n for (a) $E_{i1} = 1$, $E_{i2} = 1$, $T/\tau_0 = 1.5$; (b) $E_{i1} = 1.5$, $E_{i2} = 1$, $T/\tau_0 = 1.5$; (c) $E_{i1} = 3.5$, $E_{i2} = 1$, $T/\tau_0 = 1.5$; (d) $E_{i1} = 3.5$, $E_{i2} = 1$, $T/\tau_0 = 2$.

Fig. 5a shows that, if the incident $E_{i1}(\tau)$ amplitude is small, then the second pulse $E_2(\tau)$ generated at a frequency ω_2 and transmitted through the film has a large delay with respect to the peak of the incident pulse $E_{i2}(\tau)$, a small amplitude, and a significant half-width. The $E_2(\tau)$ formation process and the evolution of the medium polarisation are of 'lethargic' character. For a long time interval, only the biexciton density is nonzero and remains almost constant. Subsequently, dramatically rapid changes in the biexciton and exciton densities and in the amplitude of the secondary emission generated by the medium polarisation are observed.

Figs 5a, 5b allow us to conclude that, as the amplitude of the incident pulse $E_{i1}(\tau)$ rises, the duration of the lethargic evolution of the medium polarisation shortens; i.e., the delay of the generated pulse $E_2(\tau)$ decreases, its amplitude increases, and the duration shortens. If the incident pulse $E_{i1}(\tau)$ has a sufficient amplitude, the pulse generated at ω_2 may have an amplitude that exceeds the amplitude of the incident pulse at this frequency, and the generated pulse becomes noticeably narrower. An increase in the half-width of the incident pulse $E_{i1}(\tau)$ has the same effect: the amplitude of the generated pulse increases, and its half-width and delay relative to the $E_{i1}(\tau)$ peak decrease. As to the amplitude and half-width of the incident pulse $E_{i2}(\tau)$, their changes have no effect on the parameters of the pulse $E_2(\tau)$ generated at the frequency ω_2.

Therefore, two time-separated pulses at the frequency ω_2 appear after the passage of the film. Note that only one film-reflected pulse at ω_2 is observed: at the instant when the first pulse is transmitted, the reflection is absent, while, at the instant of the second pulse generation, a reflected pulse of the same profile arises. This effect is determined by the fact that the second pulse is produced by the time-varying medium polarisation, which generates identical secondary radiation pulses in two directions from the film. Consequently, it can be asserted that the film 'reflects' a pulse, when no pulse is incident on it, or, more precisely, the film-reflected pulse arises with a significant delay relative to the incidence of the pulse $E_{i2}(\tau)$.

4. Conclusions

The cases of the interaction of time-separated ultrashort laser pulses considered above lead to a conclusion that, when a delta-shaped or Gaussian pulse in the region of two-photon excitation of biexcitons from the crystal ground state and a short rectangular or Gaussian pulse in the region of the M-band of a TSF are incident onto this film, the TSF may lose its transparency at the M-band frequency or convert the incident pulses into delta-shaped peaks or spike trains in the region of exciton – biexciton conversion. A significant delay of the generation of the transmitted pulse relative to the incident one is predicted. An interpretation of the apparent effects of pulses' transmission before their incidence onto a TSF and effects of pulse generation and reflection in the absence of an incident pulse is proposed.

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