Interaction between ultrashort laser pulses and a thin semiconductor film due to two-photon creation of biexcitons

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The transmission and reflection of ultrashort laser pulses by a semiconductor thin film under the conditions corresponding to two-photon, two-pulse creation of biexcitons from the ground state have been studied using both analytic and numerical techniques. Specific examples are given to demonstrate various possibilities of controlling the transmission of both incident pulses by changing the amplitude of one of them.

1. INTRODUCTION

In recent years several theoretical studies[1–5] were dedicated to the resonant nonlinear optics of thin films. Like the authors of these papers, we define a thin film as an object whose thickness, L, is much smaller than the wavelength of incident light. Within this approach, the system of partial differential equations for the field and material can be reduced to a relatively simple system of ordinary differential and algebraic equations, and in many cases exact analytic solutions can be derived. Note than until now, the effects of nonlinear time-dependent transmission and reflection of ultra-short laser pulses have been studied largely in thin films of two-level atoms under the conditions of both one-photon[1,2] and two-photon[3,4] resonance. Undisputed progress in recent years in fabricating semiconductor structures with limited dimensions has, however, stimulated studies of nonlinear optical properties of semiconductor thin films. These studies are also of practical interest because thin films can be used in optical data-processing devices.

This paper presents a theoretical investigation of time-dependent nonlinear transmission and reflection of ultra-short laser pulses at a resonant wavelength by a semiconductor thin film under the conditions of resonant two-photon generation of biexcitons from the ground state. It is well known that two-photon biexciton generation is characterized by a giant oscillator strength.[6] Since typical times of interaction between optical pulses and materials are determined by oscillator strengths of specific optical transitions, it is feasible that two-photon generation of biexcitons from the ground state may facilitate our understanding of time-dependent nonlinear transmission of ultra-short laser pulses through a thin film. In contrast to systems investigated by Zakharov and Manykin,[3,4] specific effects related to biexcitons, in principle, depend on the relationship between the density of film atoms and that of incident photons.[7] Furthermore, owing to the giant oscillator strength of the biexciton transition, nonresonant components are negligible in comparison to resonant ones, so the Stark effect can be ignored in analyzing the biexciton transition.

2. BASIC EQUATIONS

Suppose that two monochromatic laser pulses with electric field envelopes $E_{10}(t)$ and $E_{20}(t)$ and frequencies ω_1 and ω_2, respectively, are incident on a semiconductor thin film in vacuum. The widths of the pulses are much smaller than the biexciton relaxation time in the film, and the envelopes are slow functions of time. Suppose that the frequency of each pulse is resonant with neither the excitonic nor M-band exciton–biexciton transitions,[7] but that the sum of the photon energies equals to that of the ground state to biexciton transition. In the general case, when $\omega_1 \neq \omega_2$, only two-photon generation of biexcitons by photons from the two pulses is allowed, but not by photons of each pulse individually. Pairs of coherent photons generate coherent biexcitons in the film, whose radiative recombination is responsible for luminescence generated in the semiconductor.

The Hamiltonian for a definite photon–biexciton mode includes the sum of the free biexciton and field Hamiltonians and the interaction Hamiltonian:[7,8]

$$H_{\text{int}} = -\hbar\mu(b^+ E_1^+ E_2^+ + b E_1^- E_2^-), \tag{1}$$

where μ is the constant of the two-photon generation of biexcitons from the ground state,[6] b is the amplitude of the biexciton wave, E_i^+ (E_i^-) is the component of the transmitted pulse amplitude ($i=1,2$) with positive (negative) frequency. From Eq. (1) we derive the Heisenberg equation of motion for the biexciton wave amplitude

$$i\dot{b} = \Omega_{bi} b - \mu E_1^+ E_2^+ , \tag{2}$$

where Ω_{bi} is the frequency of the ground state to biexciton transition. Assuming that the resonance condition $\omega_1 + \omega_2 = \Omega_{bi}$ is fulfilled and taking the wave amplitudes in the form

$$E_1^+ = E_1 e^{-i\omega_1 t}, \quad E_2^+ = E_2 e^{-i\omega_2 t},$$

$$b = (u + iv) e^{-i(\omega_1 + \omega_2)t}, \tag{3}$$

one can easily prove that if the crystal is initially in the ground state, then the real component u of the biexciton wave is exactly zero, and its imaginary component is described by the equation

$$\dot{v} = \mu E_1 E_2 . \tag{4}$$

As a consequence, the envelopes of the transmitted and reflected pulses are not phase modulated.

Using the techniques described in Refs. 1–4, we derive from the continuity of the tangential field component at the

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vacuum–film interface the following electrodynamic relation for amplitudes of incident, (E_{01}, E_{02}), transmitted, (E_1, E_2), and reflected, (E_{r1}, E_{r2}), pulses:

$$E_1 = E_{01} - \alpha_1 v E_2, \quad E_2 = E_{02} - \alpha_2 v E_1, \tag{5}$$

$$E_{r1} = E_1 - E_{01}, \quad E_{r2} = E_2 - E_{02}, \tag{6}$$

where

$$\alpha_1 = \frac{2\pi\hbar\omega_1 \mu L}{c}, \quad \alpha_2 = \frac{2\pi\hbar\omega_2 \mu L}{c}.$$

From Eqs. (4) and (5) we obtain

$$\frac{dv}{dt} = \mu \frac{(E_{01} - \alpha_1 v E_{02})(E_{02} - \alpha_2 v E_{01})}{(1 - \alpha_1 \alpha_2 v^2)^2}. \tag{7}$$

Equations (5)–(7) yield a complete solution to the problem in terms of the shapes of transmitted and reflected pulses derived from the envelopes of incident pulses $E_{01}(t)$ and $E_{02}(t)$ under the conditions of two-photon resonant biexciton generation from the ground state.

3. INTERACTION WITH TWO IDENTICAL PULSES

Consider the degenerate case, in which the two incident pulses have identical shapes, $E_{01}(t) = E_{02}(t) = E_0(t)$, and frequencies, $\alpha_1 = \alpha_2 = \alpha$. Then Eqs. (5)–(7) can be substantially simplified:

$$E(t) = \frac{E_0(t)}{1 + \alpha v}, \quad E_r(t) = E(t) - E_0(t), \tag{8}$$

$$\frac{dv}{dt} = \frac{\mu E_0^2(t)}{(1 + \alpha v)^2}. \tag{9}$$

Given an arbitrary shape of the incident pulse $E_0(t)$, we can derive the general solution for the fields of the incident and transmitted pulses:

$$E(t) = E_0(t) \left(1 + 3\alpha\mu \int_{-\infty}^{t} E_0^2(t') dt' \right)^{-1/3}, \tag{10}$$

$$E_r(t) = E_0(t) \left[\left(1 + 3\alpha\mu \int_{-\infty}^{t} E_0^2(t') dt' \right)^{-1/3} - 1 \right]. \tag{11}$$

Consider as an example a solution for a soliton-like incident pulse, $E_0(t) = E_0/\cosh(t/T)$ with amplitude E_0 and width T. We obtain from Eqs. (10) and (11)

$$E(t) = E_0 \frac{1}{\cosh(t/T)} \left[1 + 3\frac{T}{\tau_0} e^{t/T} \frac{1}{\cosh(t/T)} \right]^{-1/3}, \tag{12}$$

$$E_r(t) = E(t) - E_0 \frac{1}{\cosh(t/T)}, \tag{13}$$

where

$$\tau_0^{-1} = \alpha\mu E_0^2. \tag{14}$$

The typical time τ_0 over which the amplitude of the transmitted (reflected) pulse changes is determined by the square of the incident pulse amplitude, i.e., its intensity.

It follows from Eq. (12) that when the field of the incident pulse increases (curve *1* in Fig. 1), the amplitude of the transmitted pulse also rises (curve *2* in Fig. 1) and peaks at $t = t_{max}$ where

$$t_{max} = -\frac{1}{2} T \ln\left(\sqrt{\frac{T^2}{\tau_0^2} + 6\frac{T}{\tau_0} + 1} - \frac{T}{\tau_0} \right), \tag{15}$$

i.e., ahead of the incident pulse maximum $(t=0)$, and then drops rapidly.

The reflected pulse (curve *3* in Fig. 1) is generated at a considerable delay and peaks when most of the incident pulse has traversed the film. The shape of the transmitted pulse and position of its maximum with respect to that of the incident pulse is controlled by the FWHA of the incident pulse, T, and the parameter τ_0, i.e., its amplitude E_0. At $T \ll \tau_0$ the transmitted pulse maximum $(t_{max} \approx -2T^2/\tau_0)$ almost coincides with that of the incident pulse, and, as follows from Eq. (12), its shape, $E(t)$, is similar to that of the incident pulse, $E_0(t)$. At $T \gg \tau_0$ the maximum position, $t_{max} \approx -T \ln\sqrt{3}$, is totally determined by the FWHM of the incident pulse. In this case the film transmits only the leading edge of the incident pulse, and reflects essentially the entire peak—especially the trailing edge.

Similar behavior is observed in transmission and reflection of pulses with different shapes. Thus, under the conditions of single-pulse, two-photon excitation of biexcitons from the ground state, a semiconductor thin film efficiently discriminates pulses with respect to their intensity and width, transmitting short pulses of low intensity without distortion and reflecting longer and more intense ones.

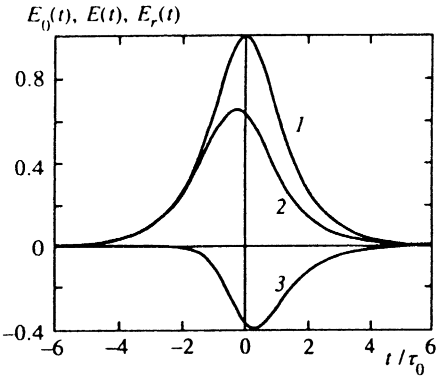

FIG. 1. Shapes of (*1*) incident, (*2*) transmitted, and (*3*) reflected pulses. The half-width of the incident soliton-like pulse equals τ_0. The pulse amplitudes are normalized to E_0.

4. INTERACTION WITH TWO DIFFERENT PULSES

We now consider a nondegenerate case. It follows from Eq. (7) that pump pulses E_{01} and E_{02} can transfer the system to a steady state. Taking $dv/dt = 0$ in Eq. (7), we find the steady state values of v: $\alpha_1 v = E_{01}/E_{02}$, $\alpha_2 v = E_{02}/E_{01}$. Assuming that one pump field, for example E_{02}, is constant, and the other is adiabatically slow, one can derive v, E_1, and E_2 as functions of E_{01} at $E_{02} = \text{const}$. For $\sqrt{\alpha_2} E_{01} < \sqrt{\alpha_1} E_{02}$, the stable solutions are

$$v = \frac{E_{01}}{\alpha_1 E_{02}}, \quad E_1 = 0, \quad E_2 = E_{02}, \tag{16}$$

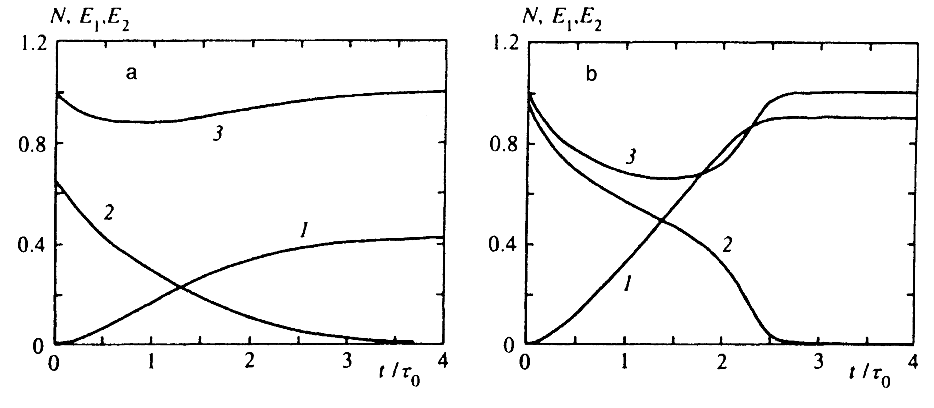

FIG. 2. (*1*) Biexciton concentration N, and amplitudes (*2*) E_1 and (*3*) E_2 of transmitted pulses at constant amplitudes E_{01} and E_{02} of incident pulses versus time. The biexciton density is normalized to $\alpha_1\alpha_2$, the light amplitudes to E_{02}. The parameter s equals (a) 0.65 and (b) 0.95.

and for $\sqrt{\alpha_2}E_{01} > \sqrt{\alpha_1}E_{02}$

$$v=\frac{E_{02}}{\alpha_2 E_{01}}, \quad E_1=E_{01}, \quad E_2=0. \tag{17}$$

These equations indicate that with increasing E_{01} the function v first grows linearly to $\sqrt{\alpha_2}E_{01}=\sqrt{\alpha_1}E_{02}$, at which point $\sqrt{\alpha_1\alpha_2}v=1$, and then drops monotonically as a hyperbolic function. Hence the steady-state amplitudes of the transmitted pulses depend on those of the incident pulses, so that the film transmits a pulse with a high amplitude, whereas a less intense pulse is totally reflected.

We next consider time-dependent transmission when incident pulses are different. If only their amplitudes are different, but their shapes are similar, i.e.,

$$E_{01}(t)=E_{01}f(t), \quad E_{02}(t)=E_{02}f(t),$$

then the solution of Eq. (7) in terms of $v(t)$ is given by the transcendental equation

$$Av+Bv^2+Cv^3+D\ \ln|1-s^{-1}\sqrt{\alpha_1\alpha_2}v|-F\ \ln|1-s\sqrt{\alpha_1\alpha_2}v|=\sqrt{\alpha_1\alpha_2}\mu E_{01}E_{02}\int_{-\infty}^{t} f^2(t')dt', \tag{18}$$

where

$$A=(s^2+s^{-2}-1)\sqrt{\alpha_1\alpha_2}, \quad D=s^3-s,$$

$$B=(s+s^{-1})\frac{\alpha_1\alpha_2}{2}, \quad F=s^{-3}-s^{-1},$$

$$C=\frac{(\alpha_1\alpha_2)^{3/2}}{3}, \quad s=\frac{\sqrt{\alpha_2}E_{01}}{\sqrt{\alpha_1}E_{02}}.$$

Let the incident pulses have constant amplitudes E_{01} and E_{02}, i.e., $f(t)=1$, and let us assume for definiteness that $s<1$. It follows from Eq. (7) and the solution of Eq. (18) that the function $v(t)$, which is numerically equal to the square root of the biexciton concentration N, monotonically rises with time and asymptotically approaches the lesser of the parameters $E_{01}/\alpha_1 E_{02}$ and $E_{02}/\alpha_2 E_{01}$, in this particular case $E_{01}/\alpha_1 E_{02}$. Given $v(t)$, one can derive from Eq. (5) the shape of the transmitted pulse and biexciton concentration, $N=v^2(t)$. The parameter τ_0, which is a characteristic of the time derivative of the transmitted and reflected pulse amplitudes, is controlled by the product of the incident pulse amplitudes:

$$\tau_0^{-1}=\sqrt{\alpha_1\alpha_2}\mu E_{01}E_{02}. \tag{19}$$

Figure 2 shows the amplitudes of transmitted pulses versus time when the pump pulses are rectangular with amplitudes E_{01} and E_{02}. The transmission of the pulse with the lesser amplitude, in this case $E_1(t)$, monotonically drops to zero. As for the second pulse (with the greater amplitude), its transmission initially drops and reaches a local minimum $E_{2\,\min}$ at time $t=t_1$, and then its transmission returns to essentially the initial value. In this case

$$\frac{t_1}{\tau_0}=\frac{1-\sqrt{1-s^2}}{6s^2}\left(13-5s^2+6s^4-(7+3s^2)\sqrt{1-s^2}\right)+\frac{1-s^2}{s^2}\ln\sqrt{1-s^2}-s^2(1-s^2)\ln\frac{\sqrt{1-s^2}}{1-\sqrt{1-s^2}}, \tag{20}$$

$$E_{2\,\min}=\frac{1}{2}E_{02}(1+\sqrt{1-s^2}). \tag{21}$$

The concentration of biexcitons monotonically rises from zero to the steady-state value $N_s=s^2/\alpha_1\alpha_2$. Figure 2(a) indicates that whereas the difference between the amplitudes of the incident pulses. E_{01} and E_{02}, is fairly large, the time-dependent amplitudes $E_1(t)$ and $E_2(t)$ change gradually. If the system evolution is treated in the approximation of $E_{02}\gg E_{01}$ (the approximation of a prespecified field for the second pulse) the solution of Eqs. (5) and (7) yields $E_2(t)=E_{02}=\text{const}$ and $E_1(t)=E_{01}\exp(-\alpha_1\mu E_{02}^2 t)$. The higher the amplitude E_{02} of the second pulse, the faster the amplitude of the transmitted pulse $E_1(t)$ drops, and the faster the film switches to total reflection mode, in which the amplitude of the weaker pulse is completely reflected while the more intense pulse is transmitted essentially unchanged. If the amplitudes of the initial pulses are similar (Fig. 2b), then after some interval during which the transmitted pulse amplitudes change gradually, their amplitudes change abruptly—in essentially step-like fashion, after which the film transmission is constant. The behavior of $E_2(t)$ changes

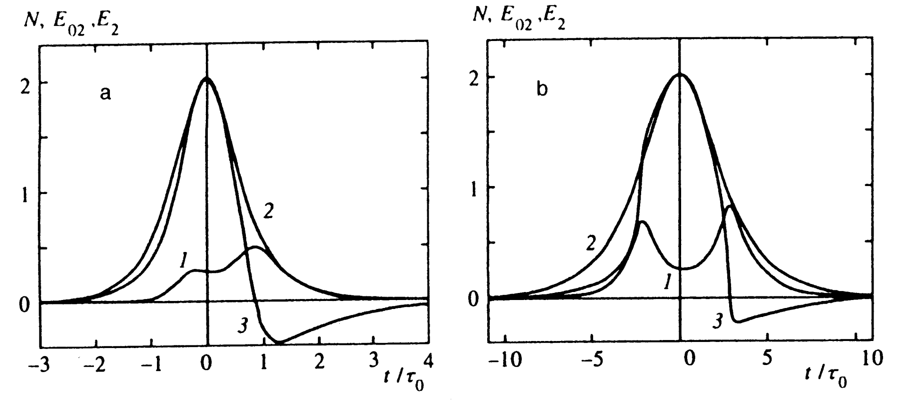

FIG. 3. (*1*) Biexciton concentration N, and amplitudes of (*2*) incident, E_{02}, and (*3*) transmitted, E_2, pulses at $E_{01}(t) = E_0 = \text{const}$ and $E_{02}(t) = 2E_0/\cosh(t/T)$. The biexciton concentration is normalized to $\alpha_1\alpha_2$, light amplitudes to E_0. The half-width of the incident pulse $E_{02}(t)$ equals (a) $0.5\tau_0$ and (b) $2.0\tau_0$.

so radically because the original pulse E_{01} is essentially totally reflected, and photons of the pulse with the greater amplitude are transmitted through the film as if through a transparent material, because as originally assumed, biexcitons cannot be generated without photons of the first pulse.

If at some moment one of the incident pulses, for example E_{02}, is cut off, system evolution continues according to

$$\frac{dv}{dt} = -\frac{\mu E_{01}^2 \alpha^2 v}{(1-\alpha_1\alpha_2 v^2)^2}. \tag{22}$$

The solution of this equation has the form

$$\ln\frac{v}{v_0} + \alpha_1\alpha_2(v_0^2 - v^2) + \frac{1}{4}\alpha_1^2\alpha_2^2(v^4 - v_0^4) = -\alpha_2\mu\int_0^t E_{01}^2(t')dt', \tag{23}$$

where v_0 is the value of $v(t)$ when the second pulse is cut off. If follows from Eq. (23) that $v(t)$ drops with time monotonically. The higher the amplitude of the incident pulse E_{01}, the faster it decreases. The amplitudes of the pulses transmitted through the film are

$$E_1(t) = \frac{E_{01}}{1-\alpha_1\alpha_2 v^2}, \quad E_2(t) = -\frac{E_{01}\alpha_2 v}{1-\alpha_1\alpha_2 v^2}. \tag{24}$$

Clearly, although one of incident pulses is cut off, radiation can be detected at both frequencies because of the induced recombination of biexcitons previously created in the film. The shapes of the transmitted pulse $E_2(t)$ and the corresponding reflected pulse $E_{r2}(t)$ are similar because they both are generated by the secondary emission due to induced recombination of biexcitons. The pulse $E_1(t)$ is partly due to the incident pulse $E_{01}(t)$, and partly due to the secondary emission.

Equations (5)–(7) can be solved in the case of arbitrary pulse shapes only numerically. Below we consider a fairly simple case in which one of the pulses, e.g., E_{01}, is rectangular, and the amplitude of the other pulse varies with time as $E_{02}(t) = E_{02}/\cosh(t/T)$. The system evolution is largely determined by the ratio $\sqrt{\alpha_1}E_{02}/\sqrt{\alpha_2}E_{01}$. If the peak value of the second pulse amplitude is such that $\sqrt{\alpha_1}E_{02}/\sqrt{\alpha_2}E_{01} < 1$, the density of biexcitons will initially rise peak, and then drop rapidly. The transmitted pulse $E_2(t)$ has a distorted bell-like shape. The larger the FWHM of the incident pulse $E_{02}(t)$, the steeper the trailing edge of the transmitted pulse, and the sooner its peak is observed with respect to that of the incident pulse.

If, on the contrary, $\sqrt{\alpha_1}E_{02}/\sqrt{\alpha_2}E_{01} > 1$, new features of the process become evident. As follows from Eq. (7), the time derivative of $v(t)$ vanishes twice, and its sign changes at the leading and trailing edges of the incident pulse. As a result, the density of biexcitons (curve 1 in Fig. 3) has two maxima and one minimum. The normalized density of biexcitons $\alpha_1\alpha_2 N$ is at most unity, and the amplitude of the negative peak drops with increasing ratio $\sqrt{\alpha_1}E_{02}/\sqrt{\alpha_2}E_{01}$ and increasing FWHM of the pulse $E_{02}(t)$. While $\sqrt{\alpha_1}E_{02}(t) < \sqrt{\alpha_2}E_{01}$, the energies of both transmitted pulses are largely derived from the incident pulses, and the luminescence intensity is small. But when $\sqrt{\alpha_1}E_{02}(t) \approx \sqrt{\alpha_2}E_{01}$, the radiative recombination of biexcitons induced by the external field is the dominant process. Therefore the transmitted pulse $E_2(t)$ consists of two subpulses, the first of which has an FWHM smaller than that of $E_{02}(t)$. As in the case of two identical pulses, the transmission of a soliton-like incident pulse is controlled by the relationship between its FWHM (T) and parameter τ_0 determined by Eq. (19). If $T < \tau_0$ (Fig. 3a), the film easily transmits its leading edge and maximum, and at the trailing edge of the incident pulse, where the density of biexcitons in the film is a maximum, the transmission drops to zero. At a lower amplitude of $E_{02}(t)$, radiative annihilation of biexcitons is induced, and a second subpulse with negative amplitude is generated.

A very short pulse ($T \ll \tau_0$) cannot generate a sufficient density of biexcitons, so the shape of the transmitted pulse is essentially unchanged. As the FWHM $E_{02}(t)$ of the incident pulse increases, the shape of the transmitted pulse approaches the quasistable solution defined by Eqs. (16), (17), i.e., as long as $\sqrt{\alpha_1}E_{02}(t) < \sqrt{\alpha_2}E_{01}$ the amplitude $E_2(t)$ is very small, otherwise the shape of the transmitted pulse is similar to that of the incident pulse. Therefore for $T > \tau_0$ (Fig. 3b), the transmitted pulse is shorter than the incident one. Thus, the thin film easily transmits short pulses and shortens longer ones. Since $\tau_0 \sim E_0^{-2}$, the film also easily transmits weak pulses and shortens those of higher intensity.

It has not been possible to derive exact analytic solutions of Eqs. (5)–(7) for arbitrary incident pulse shapes. Nonetheless, fairly simple solutions of the inverse problem can be derived, i.e., the shapes of incident pulses can be derived from predetermined envelope functions of transmitted pulses. Indeed, if $E_1(t)$ and $E_2(t)$ are given functions of time, Eqs. (5) and (7) yield

$$E_{01}(t)=E_1(t)+\alpha_1\mu E_2(t)\int_{-\infty}^{t} E_1(t')E_2(t')dt', \qquad (25)$$

$$E_{02}(t)=E_2(t)+\alpha_2\mu E_1(t)\int_{-\infty}^{t} E_1(t')E_2(t')dt'. \qquad (26)$$

Specifically, if one transmitted pulse is required to have constant amplitude, $E_2(\tau)=E_0=\text{const}$, while the amplitude of the other rises linearly, $E_2(\tau)=E_0\tau$, where $\tau=t/\tau_0$, $\tau_0^{-1} = \sqrt{\alpha_1\alpha_2\mu E_0^2}$, then the amplitudes of incident pulses versus time must be

$$E_{01}(t)=E_0\left(1+\frac{1}{2}\tau^3\right), \quad E_{02}(t)=E_0\tau\left(1+\frac{1}{2}\tau\right). \qquad (27)$$

Clearly, although the amplitudes of both incident pulses grow with time, one of the transmitted pulses has constant amplitude. This again suggests that incident pulses can be discriminated with respect to their intensities.

5. CONCLUSIONS

We have obtained basic equations and studied features of time-dependent transmission and reflection of ultrashort resonant laser pulses by a semiconductor thin film under the conditions of two-pulse, two-photon generation of biexcitons from the ground state. We predict that this technique can be widely used to control the transmission of both pulses by varying the amplitude of one pulse, and therefore can be employed in fast optical integrated circuits.

Note that our results may be altered if some of the conditions of our approximation are not fulfilled. In particular, at higher pulse intensities, elastic biexciton–biexciton scattering may lead to phase modulation of the transmitted pulses, and the function relating output and input amplitudes may be multivalued.

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