Specific features of nonlinear interaction between femtosecond laser pulses and molecular atmosphere

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Peculiarities of nonlinear interaction between high-power femtosecond laser pulses and molecular atmosphere are analyzed. The peculiarities are connected with combined action of elementary nonlinear effects such as multiphoton or tunnel ionization of molecules, Kerr nonlinearity, and nonlinear resonance absorption by gas components of the atmosphere. Particular emphasis is placed on the possible mechanisms of nonlinear effects of the lowest orders affecting the femtosecond laser pulse propagation along atmospheric paths.

Introduction

The advent of high-power lasers generating short pulses of tens femtoseconds duration has yielded a new technology of analyzing the atmospheric gas composition by laser absorption spectroscopy with a new broadband light source — the supercontinuum. The supercontinuum is generated in a chosen atmospheric region by terawatt femtosecond-duration laser pulses. As was demonstrated by P. Rairoux et al. (Ref. 1), the supercontinuum, like a laser beam, keeps well collimated along the entire propagation path.

Propagation of terawatt femtosecond pulses (FSP) through molecular atmosphere differs from the well-studied mode of propagation of nanosecond pulses. Interaction of the FSP with vibrational-rotational transitions of the atmospheric molecules is nonstationary. The FSP are shorter than the vibrational or rotational relaxation time, and the spectral width of the FSP delivered by a Ti:Sapphire laser covers several vibrational-rotational bands of H_2O and O_2 molecules. In this situation, one should expect a complicated absorption of the FSP radiation by air.

Indeed, measurements (Matvienko et al., Ref. 2) of the atmospheric absorption of nano- and femtosecond pulses having comparable widths of the generated spectrum (23.4 nm for 9-ns pulse and 17.7 nm for FSP of 80 fs) show that for the pulse energy from 0.1 to 10 mJ dependence of the air-absorbed energy is close to a quadratic for the FSP while being linear for nanosecond pulses. Later, with the same setup the result indicating that with a further increase of energy, the dependence of air absorption follows a power law: $E_{\text{abs.}} \sim E_{\text{las}}^n$, where $n > 2$ was obtained.[3] The high FSP energy concentrated in a spatially limited volume results in the peculiarities of nonlinear interaction of laser pulses with molecular atmosphere connected with the combined action of elementary nonlinear effects such as multiphoton or tunnel ionization of molecules, Kerr nonlinearity, and nonlinear resonance absorption by the atmospheric gas components, etc. Appearance of new channels for the transfer of laser energy to the ambient medium makes its interaction with the medium much more complicated giving rise to nonlinear effects, which are not observed within the traditional nonlinear optics.

In this paper, we review and analyze manifestations of the quadratic and cubic nonlinear effects in the interaction of femtosecond laser pulses with molecular atmosphere.

Quadratic nonlinearity like other even-order nonlinearities is typical only of the media with no symmetry centers like, for example, anisotropic crystals (see, e.g., Ref. 4). In gas media, because of their isotropy, the third order is the lowest order of nonlinearity possible. It governs the self-action effects, third harmonic generation, etc. Under conditions of self-action in such media, the second order nonlinear effects are impossible in principle. In the isotropic media, these effects occur only under an additional static electric field.[4]

1. Air ionization

Oxygen molecules have the lowest ionization threshold among air molecules (12.1 eV), which several times exceeds the energy of a visible or an IR photon. The multiphoton ionization process is determined by the Keldysh parameter[5]:

$$\gamma = \omega \frac{\sqrt{2m_e U(0)}}{eE},$$

where E and ω are the laser field strength and the frequency; m_e and e are the electron mass and charge, respectively; $U(0)$ is the field free ionization potential. If $\gamma \gg 1$, then the multiphoton ionization of atoms prevails. At $\gamma \ll 1$, the Coulomb potential of an ion is strongly distorted by the external field, and the mechanism of tunnel ionization becomes

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prevailing. In the case of especially strong optical fields, the Coulomb barrier is suppressed by the external field and the overbarrier ionization becomes possible.[6]

In describing the ionization, we use kinetic models, where ionization rates are determined by the type of the process. The rate of multiphoton ionization is connected with the laser radiation intensity in the following way[5]:

$$A = \sigma^{(k)} I^k(r,t), \qquad (1)$$

where A is the ionization rate; $k = \mathrm{mod}(U(\omega)/\hbar\omega + 1)$ is the number of photons required for ionization (in ionization of oxygen molecules exposed a Ti:Sapphire laser radiation $k = 8$, Ref. 7); $\sigma^{(k)}$ is the multiphoton ionization cross section. Here

$$U(\omega) = U(0) + e^2E^2/(4m_e\omega^2).$$

A typical model of generation of free electrons in air under the influence of femtosecond laser pulses is determined by the following equation[8–11]:

$$\frac{\partial \rho}{\partial t} = \sigma^{(k)} I^k(r,t)(\rho_{at} - \rho), \qquad (2)$$

where ρ is the charge concentration; ρ_{at} is the concentration of free electrons in the atmosphere.

The electron knocked out from an atom undergoes the action of electromagnetic field of laser radiation, which acts as an additional barrier[5] determined by the ponderomotive energy, which equals $U_p(I) = 9.33 \cdot 10^{-14} I\lambda$, eV, where λ is the laser radiation wavelength, μm; I is the intensity, W/cm^2. Note that the threshold of multiphoton ionization does not depend on barrier height. This is indicative of that the ionization is a two-stage process: first, the electron is excited to the barrier-defined energy of continuum states being still near the nucleus. Then, it starts absorbing more light energy and escapes from the atom.

Strictly speaking, the Keldysh theory is applicable to hydrogen-like atoms.[12] To calculate molecular ionization rate, one needs for an empirical correction. The comparison of the theory and experiment shows that Keldysh theory provides quite good agreement up to the electron density of ~ 10^{16} cm^{-3}.

One of the important consequences of plasma formation in air is the change of the air refractive index Δn_p, which can be calculated by the Drude model[13]:

$$\Delta n_p = -2\pi e^2 N_e/(m_e\omega^2). \qquad (3)$$

From Eqs. (2) and (3) it follows that

$$\Delta n_p = -2\pi e^2 \rho_{at} \exp\left(\sigma^{(k)} \int_{-\infty}^{t} I^k(t')\,dt'\right)/(m_e\omega^2).$$

For the radiation of a Ti:Sapphire laser, taking into account that $\rho_{at} = 2.7 \cdot 10^{19}$ cm^{-3} (Ref. 11) we obtain:

$$\Delta n_p = 5.7 \cdot 10^{-7} \exp\left(\sigma^{(k)} \int_{-\infty}^{t} I^k(t')\,dt'\right).$$

A usual value for air is $\sigma^{(k)} = 3.7 \cdot 10^{-96}$ s$^{-1} \cdot$ cm^{16} W^{-8} (Ref. 11); therefore, a change in the air refractive index caused by its multiphoton ionization by femtosecond pulses of a Ti:Sapphire laser is important only for the intensities greater than 10^{13} W/cm^2.

2. Generation of the second harmonic in air

Generation of the second harmonic belongs to the second order nonlinear effects, when nonlinear polarization of a medium is quadratically dependent on the optical field intensity:

$$P_i^{(2)}(r,t) = \sum_{jk} \chi_{ijk}^{(2)} E_j(r,t) E_k(r,t). \qquad (4)$$

Here $P_i^{(2)}(r,t)$ and $E_j(r,t)$ are the components of nonlinear polarization of a medium and optical field intensity, respectively.

Generation of the second harmonic (and other even-order harmonics) in a centrosymmetric medium is possible only if high-order multipoles, namely, the electric quadrupole or magnetic dipole moments of a molecule or an atom exist. Since the electric quadrupole component is related to the electric dipole component as a characteristic of the order[4]

$$ka = 2\pi a/\lambda,$$

where a is the size of an atom or a molecule, then for the optical range, the electric quadrupole polarization is three orders of magnitude weaker than the dipole polarization.

If propagation of a laser pulse is accompanied by generation of inhomogeneous plasma, then the medium is no more centrosymmetrical, and generation of the second harmonic is also possible with the third order nonlinearity, when the effect of laser radiation field on the medium is combined with that of the electric field of the inhomogeneous plasma:

$$P_{2\omega} = \chi^{(3)} \langle E_s \rangle EE, \qquad (5)$$

where $\langle E_s \rangle$ is the field intensity of inhomogeneous plasma averaged over the optical period; $\chi^{(3)}$ is the medium nonlinear susceptibility tensor of the third order, and $P_{2\omega}$ is the medium double-frequency polarization component.

Theoretical and experimental analyses of the process of generation of the second harmonic of the type presented by Eq. (5), when plasma inhomogeneity is due to field inhomogeneity and due to ponderomotive forces, were performed by K. Miyazaki et al. (Ref. 14). Their experiments were carried out with the use of a Q-switched Nd:YAG laser and atomic sodium vapor. The laser delivered 50 mJ per 28-ps long pulse; the beam was focused to

0.3 mm. The experiments showed that the signal of the second harmonic is roughly proportional to the fourth power of the laser radiation intensity.

H.S. Brandi et al. (Ref. 15) have suggested the method of calculating the field of the second harmonic based on paraxial approximation and formalism of the Green's functions. Their calculations show that the total field power of the second harmonic is proportional to the squared laser field intensity.

Experimental observations of the signal of the second harmonic of the femtosecond laser pulses of a Ti:Sapphire laser in air for the intensities below the optical breakdown threshold show that the signal is proportional to pulse energy to the power 1.9 ± 0.1 (Ref. 16). Here it is noted that ionization of air by femtosecond laser pulses starts from intensities of $\sim 10^{11}$ W/cm^2 and becomes much stronger at the breakdown. This value can be considered a threshold for this effect.

3. Generation of the third harmonic in air

For an effective generation of the third harmonic one has to fulfill the requirement of phase synchronism at the frequencies of the fundamental wave and the third harmonic:

$$n(\omega) = n(3\omega).$$

Beyond the regions of the resonance absorption, air has a normal dispersion. Hence, fulfillment of this requirement is impossible. However, it is easy to meet it if a nonlinear change of the refractive index at the fundamental frequency ω occurs due to the Kerr effect and plasma generation[17]:

$$n_0(\omega) + n_2(\omega) I(\omega) - \Delta n_p(\omega) = n_0(3\omega) - \Delta n_p(3\omega),$$

where n_0 is the linear air refractive index; n_2 is a nonlinear (Kerr) contribution to the refractive index. For a free electron concentration of $\sim 10^{16}$–10^{17} cm^{-3} the laser radiation intensity that provides phase synchronism is 10^{13}–10^{14} W/cm^2 (Ref. 17). In this case, the intensity of the third harmonic is determined by the following relation

$$I_{3\omega}(z) = \frac{(3\omega)^2}{n_{3\omega} n_\omega^2 \varepsilon_0 c^4} I_\omega^3 l_{\text{eff}}^2,$$

where $l_{\text{eff}}^2 = \chi_{\text{eff}} z$ is the effective interaction length, within which the phase synchronism requirement is fulfilled.

In the experiments, generation of the third harmonic of a Ti:Sapphire laser in air started from $I(\omega) \sim 2.5 \cdot 10^{13}$ W/cm^2 (Ref. 11), which can be regarded as a threshold for this effect.

4. Optical rectification effect

With generation of free electrons in air under the action of femtosecond laser pulses according to model (2), the charges with a time dependent density create in the ambient medium an electromagnetic wave, whose field strength E_e can be described by the wave equation

$$\nabla^2 E_e - \frac{1}{c^2} \frac{\partial^2 E_e}{\partial t^2} = \frac{4\pi}{c^2} \frac{\partial j_e}{\partial t} - 4\pi e \nabla \rho. \quad (6)$$

Here j_e is the current density of free charges; c is the speed of light. To determine this quantity, let us use the following equation[10]:

$$\frac{\partial j_e}{\partial t} = \frac{e^2}{m} \rho E_e - \frac{j_e}{\tau_c}, \quad (7)$$

where τ_c is the time of collisional relaxation of plasma. For the air under normal conditions $\tau_c = 3.5 \cdot 10^{-13}$ s (Ref. 9).

Equation (6) does not allow for the secondary effects connected with the interaction of the field E_e with the medium and determined by the material equations of the form $P = f(E_e)$, where P is specific medium polarization.

The presence of the additional field E_e in the medium in combination with the laser field E can produce nonlinear air polarization, which, in particular, will have the following component:

$$P^{(3)}(r,t) = \chi^{(3)} \left| E(r,t) \right|^2 E_e(r,t) = n_2 I(r,t) E_e(r,t). \quad (8)$$

If the field has a quasi-stationary character (if it does not change at least with the frequency close to that of the laser field), then nonlinear medium polarization (8) can result in the effect similar to that of optical rectification.

Let us analyze the properties of free charge field E_e, appearing due to multiphoton air ionization by a high-power laser field. For this purpose, we will reduce the order of Eq. (6). Usually, to reduce the order of a wave equation that describes propagation of optical pulses, including femtosecond pulses, one uses the method of slowly varying amplitudes.[6,18] However, since we cannot assert it *a priori* that a free charge field has a high carrier frequency, the method of slowly varying amplitudes is inapplicable here. So, to reduce the equation order, we will use the approximation of a unidirectional propagation (Ref. 19). Equation (6) in this and in the quasi-optical approximations will take the following form

$$\frac{\partial E_e}{\partial z} + \frac{1}{c} \frac{\partial E_e}{\partial t} = \frac{c}{2} \int_0^t \nabla_\perp^2 E_e dt' - \frac{2\pi}{c} j_e - 2\pi c \int_0^t \nabla \rho dt'. \quad (9)$$

For simplicity, assume the laser field having the shape of a slit-beam. Then, in a scalar approximation, the transverse field component $E_e = (E_e)_x$ will be described by the following equation

$$\frac{\partial E_e}{\partial z} + \frac{1}{c} \frac{\partial E_e}{\partial t} = \frac{c}{2} \frac{\partial^2}{\partial x^2} \int_0^t E_e dt' - \frac{2\pi}{c} j_e - 2\pi c \int_0^t \frac{\partial \rho}{\partial x} dt'. \quad (10)$$

The problem of propagation of the free charge field (Eqs. (2), (7), and (10)) was analyzed

numerically. In this case, the following circumstance was taken into account:

$$\rho / \rho_{at} \ll 1, \quad \tau_p / \tau_c \ll 1 . \qquad (11)$$

The calculations were performed for the following values of the problem parameters: a Ti:Sapphire laser delivers a pulse of 50-fs duration, 10 mJ energy, and 3-mm diameter; $\sigma^{(k)} = 3.7 \cdot 10^{-96}\ s^{-1} \cdot cm^{16} \cdot W^{-8}$, $\rho_{at} = 2.7 \cdot 10^{19}\ cm^{-3}$ (Ref. 5). Note that the data on nonlinear response of a medium that can be found in literature correspond to the pulsed radiation with a high carrier frequency that lies within the range of the visible or UV radiation. Strictly speaking, polarization field (10) is not the one of this type. However, the dependence of n_2 on the frequency is not very strong. So, in our estimates, we used the following value for the UV region: $n_2 = 8 \cdot 10^{-19}\ s^2 \cdot W^{-1}$ (Ref. 11).

The laser pulse field as it enters the medium has the form

$$E(x,z,t)\,|_{z=0} =$$

$$= \begin{cases} E_0 \sin(\pi t / \tau_p) \exp(-x^2 / x_0^2) \exp(i\omega t), & 0 \le t \le \tau_p, \\ 0, \quad t > \tau_p, \quad t < 0, \end{cases} \qquad (12)$$

where x_0 is the initial beam radius. For simplicity, we used the approximation of a preset pump field typical of the problems of nonlinear parametric interaction, i.e., we assumed that the laser pulse field in the medium would take the form of Eq. (12).

In Fig. 1 we illustrate the free charge field strength in the medium. It is seen that this field does not possess a high-frequency carrier, i.e., it is a quasi-stationary field indeed. Note that if we take into account a collisional relaxation the free charge field starts to decay with time at τ_c time after the start of the interaction between the laser pulse and medium.

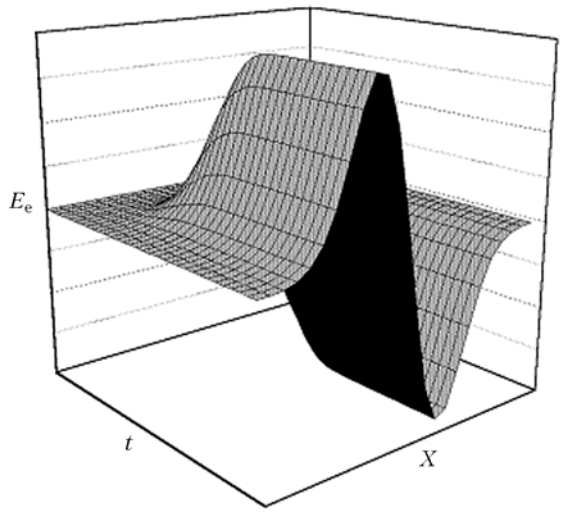

Fig. 1. Spatiotemporal structure of the field of free charges generated by a femtosecond pulse of a Ti:Sapphire laser in air.

Let us now analyze the field created by the cubic nonlinearity of the type (8). In the calculations, we used the following form of the wave equation describing the strength distribution of this field E_d in the medium following the approximation of a unidirectional propagation.

$$\frac{\partial E_d}{\partial z} + \frac{1}{c}\frac{\partial E_d}{\partial t} = -\frac{2\pi}{c}\frac{\partial P^{(3)}}{\partial t}, \qquad (13)$$

where the cubic medium polarization $P^{(3)}$ is determined by Eq. (7). Note that here, for simplicity, we neglect the diffraction-induced blurring, because the field length along the propagation path $c\tau_p \ll x_0$.

Figure 2 shows the field strength E_d in the medium calculated by Eq. (13).

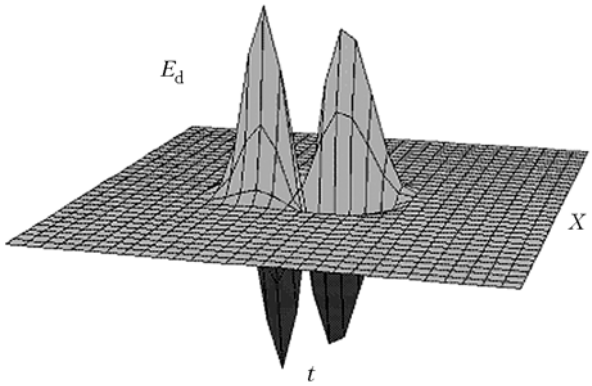

Fig. 2. Spatiotemporal structure of the field of an optical videopulse.

Figure 3 shows the lines of the equal field strength E_d in the relative units E_d/E_m, where E_m is the maximum intensity of the laser field after it has passed a distance of 0.4 cm in the medium.

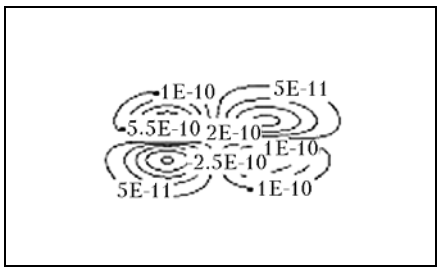

Fig. 3. The lines of the equal strength of the field of an optical videopulse.

From Figs. 2 and 3 we can see that unlike the optical rectification effect in the constant electric field,[4] in this case the structure of the videopulse formed is determined by the derivative of the spatiotemporal shape of the laser field envelope. Since the mechanism of this phenomenon is very similar to the generation of the second harmonic in air, then to estimate the threshold intensity of the optical rectification effect in air we can use the intensity of $10^{11}\ W/cm^2$.

Note that within the approximations accepted, the rate of the laser energy transfer to the videopulse wave is close to an exponential (Fig. 4).

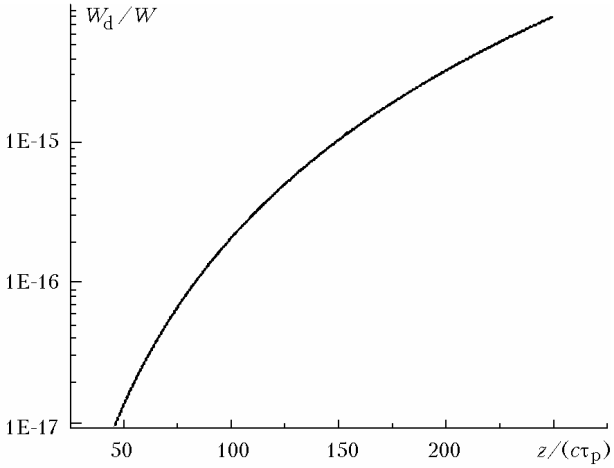

Fig. 4. Dynamics of the relative energy growth of the optical videopulse W_d/W at propagation of the femtosecond pulse of a Ti:Sapphire laser in air. Here, W_d stands for the optical videopulse energy, W is the laser pulse energy.

5. Resonance absorption effect

Let us analyze the resonance absorption effect using the quasi-optical approximation and the method of slowly varying amplitudes. The main nonlinear effects for the femtosecond pulses with the intensity $I < 10^{14}$ W/cm^2 in a gaseous medium are[13]:

1) the electron Kerr effect connected with the nonlinear variation of the medium refractive index, which in the first approximation is described by the expression

$$\Delta n_k = n_2 \mid E \mid^2 . \qquad (14)$$

Here, the nonlinear modulation of the medium refractive index is caused, first of all, by the anharmonicity of the molecular electronic response and Raman scattering at rotational transitions of the medium molecules (see Refs. 13 and 20);

2) material dispersion of the medium, connected with the presence of the gaseous medium absorption lines within the laser emission spectrum. Dispersion and its accompanying absorption by medium can be taken into account in the approximation of rotating wave and slowly varying amplitudes as follows (Ref. 18):

$$\frac{dP}{dt} = i\Delta\omega P + i\varsigma w E, \qquad (15)$$

$$\frac{dw}{dt} = -\varsigma \mathrm{Im}(P^* E), \qquad (15a)$$

where P^* is the complex molecular polarization; w is the population difference; $\Delta\omega$ is detuning from the absorption line center; $\varsigma = 2d/\hbar$, d is the transition dipole moment.

With the allowance made for these mechanisms, the propagation of pulsed radiation can be described by the following model:

$$2i\frac{\partial E(z,\tilde{\rho},\eta)}{\partial z} + \frac{1}{k}\Delta_\perp E(z,\tilde{\rho},\eta) + kn_2 \mid E \mid^2 E = \\ = -i4\pi kT_2 \sum_i d_i N_i \varsigma_i P_i, \qquad (16)$$

the beam in this model is assumed axially symmetric; the inhomogeneous broadening is neglected; T_2 is the time of the medium phase relaxation; N_i is the concentration of molecules; $\eta = t - z/c$; $\tilde{\rho}$ stands for the radial coordinate.

The propagation problems similar to the model (13)–(16) are analyzed in Refs. 6–13. With no resonance absorption, Eq. (16) corresponds to the known model of nonlinear Schrödinger equation, which at $n_2 > 0$ gives the soliton solutions, which are due to the balance between the diffraction-induced beam blurring and its nonlinear focusing-induced self-contraction. Without any distorting factors, this equation can be integrated by the inverse scattering method.[21]

Without the Kerr nonlinearity and in the case of single resonance transition at the exact resonance, Eq. (16) describes the solitons of a self-induced transparency (Ref. 18). With two resonance transitions and $d_1/d_2 = 2$, radiation propagation is described by the model of a double sin-Gordon, which cannot be integrated, but has the soliton-like solutions.[22]

The numerical scheme of solving Eq. (16) was based on the method of a differential pass along the radial coordinate in combination with the iterative method of successive approximations with the allowance for the nonlinear component, resonance absorption, and anomalous dispersion.[23] Simulation was run for the spectrum comprising equidistant absorption lines.

The initial pulse shape had a radial Gaussian distribution. The time behavior of the field amplitude was described by the following function:

$$E(t) = \begin{cases} E_0 \sin^q(\pi t/t_p), & t \in [0, t_p], \\ 0, & t \notin [0, t_p], \end{cases}$$

where t_p is the pulse duration; the parameter q determines the steepness of the pulse fronts. In calculations, we took the value $q = 0.25$. The coordinate z was normalized to the linear stationary resonance absorption coefficient at the line center.

The results calculated for pulse shape at its propagation in the medium with no regard for the resonance medium component are presented in Fig. 5.

We can see that the pulse generally keeps its original shape unchanged, which corresponds to the conditions for existence of nonlinear Schrödinger equation solitons.

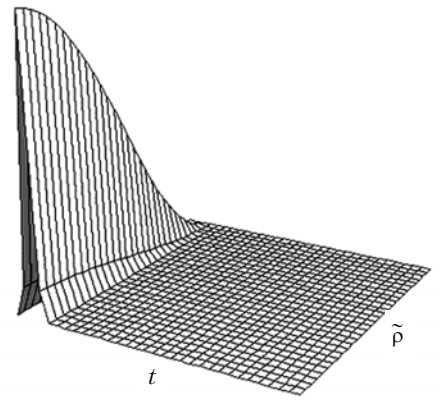

Fig. 5. Calculated pulse shape in the medium. The optical thickness is taken to be $\tau = 30$.

Figure 6 illustrates the influence of two absorption lines on the pulse shape. In this case, we observe a significant distortion of the pulse shape, which is due to the combined action of the Kerr and resonance nonlinearities. Note that the pulse amplitude is sufficient for exciting the self-induced transparency solitons, but the presence of two resonance transitions with different dipole moments leads to formation of the sequence of pulses with the invariable total area and variable amplitudes of each of them (Ref. 24).

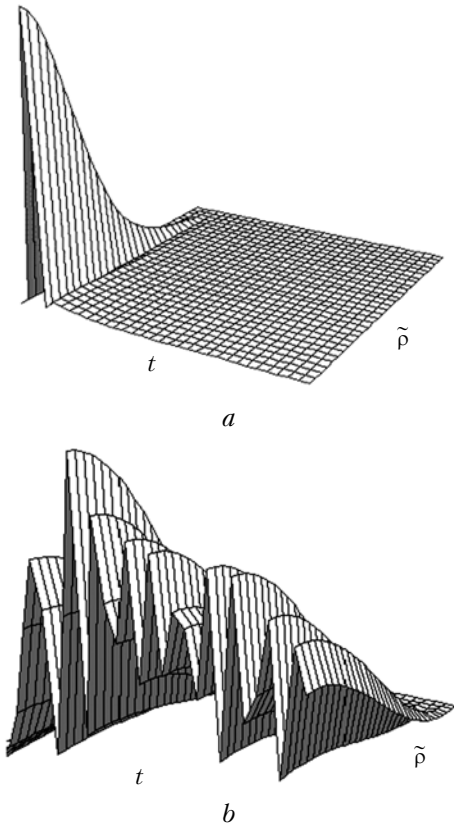

Fig. 6. Calculation of pulse shape in the medium. Calculation parameters: $d_1 = 3d_2$, $N_1 = N_2$, $\Delta\omega_1 T_2 = \Delta\omega_2 T_2 = 1$, $\varsigma E_0 t_p = 1$, the optical thickness $\tau = 1$ (*a*); $\tau = 30$ (*b*).

Calculated results on the pulse shape transformation in the medium whose absorption spectrum involves a set of ten equidistant lines of equal intensity spaced by a line half-width are shown in Fig. 7. We see that with a significant optical thickness, the resonance absorption can play an important role in transformation of laser pulse characteristics.

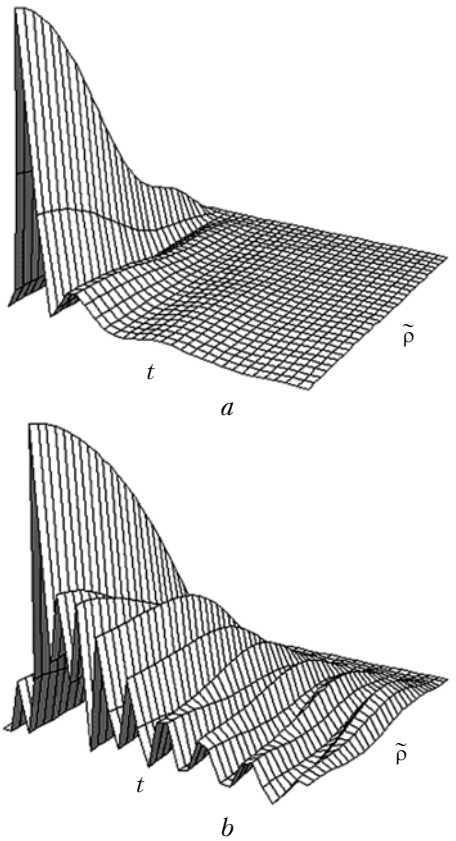

Fig. 7. Calculation of the pulse shape in the medium. Calculation parameters: $\varsigma E_0 t_p = 1$, the optical thickness $\tau = 1$ (*a*); $\tau = 30$ (*b*).

Acknowledgments

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