Contribution to the theory of coherent interaction of light pulses with resonant multilevel media

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The interaction of coherent radiation pulses in resonant multilevel media is considered. Analytic solutions that describe the propagation of multifrequency pulses and their collisions under self-induced transparency conditions are obtained via the inverse scattering method. The conditions for the appearance of soliton solutions for multifrequency pulses are obtained. The interaction between coherent pulses is considered in the case of parametric mixing of the frequencies, and the transformation length in a resonant medium is obtained. The results are generalized to include multilevel media.

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1. INTRODUCTION

Coherent interaction of laser radiation with resonant media is being intensively investigated over a number of years. The overwhelming majority of studies of this subject deal with the interaction of monochromatic radiation with media that are satisfactorily described by the two-level approximation.[1–3] In addition to detailed numerical investigations, effective analytic methods were developed and make it possible to describe in sufficient detail the evolution of the longitudinal structure of the pulses in both absorbing[4,5] and amplifying two-level media.[6,7] It became clear in the last few years that substantial role in the propagation of coherent pulses in two-level media is played by a small-scale instability that gives rise to a transverse structure of the pulses.[8–10] The evolution of this instability limits the propagation length of the coherent pulses in resonant media. The one-dimensional description is thus valid at not too large interaction lengths, and its applicability region should be determined in each concrete case by a two-dimensional analysis of the transverse structure of the pulse.

Ever increasing interest is now attracting by the interaction of short multifrequency laser pulses with resonant multilevel medium. It is shown in Ref. 11 that when resonant multifrequency radiation interacts with cascade transition it is possible to control, in a wide range, the parameters of short pulses in the self-induced transparency (SIT) regime. The use of multifrequency pulses can lower substantially the SIT thresholds and permits measurements of the dipole moments for weakly allowed transitions. Joint propagation of multifrequency pulses in multilevel absorbing and amplifying medium was investigated in Refs. 12–14 by analytic and numerical methods.

Stationary solutions that describe the propagation of pulsed multifrequency radiation with a common envelope in a multilevel resonant medium were obtained in Refs. 12 and 13 (the authors called these pulses "simultons"). A stationary propagation regime corresponds to a definite choice of the amplitudes of the individual frequency components and populations that participate in the level interaction. The last requirement means that a definite relation must be established between the level until the start of the interaction with the radiation. The solutions obtained in Ref. 14 describe stationary propagation of a multifrequency pulse in a medium, a pulse causing transitions to several upper levels from one common lower level. Experimental observation of the stationary propagation regime in this case does not call for a special "preparation" of the initial state of the multilevel system, and requires only selection of definite ratios of the amplitudes and durations of the individual frequency components of the pulse. Of course, a set of stationary solutions cannot describe the evolution of a multifrequency pulse having an arbitrary shape and entering a resonant medium.

Of particular interest is the frequency conversion of the pulses in multilevel system in the coherent regime. i.e., coherent parametric interaction of pulses with different frequencies. The point is that in the SIT regime high-power pulses pass through resonant medium without loss to absorption, scattering, etc. At the same time, the resonant character of the interaction gives grounds for hoping for a high effectiveness of parametric conversion of the frequency of light. To solve the posed questions it is necessary to trace the evolution of multifrequency radiation in resonant multilevel systems. The present paper is devoted to an exposition of exact results obtained because we succeeded in a number of cases to apply the method of the inverse problem of scattering theory to the systems described above. To simplify the exposition we consider first three-level system, and then present a generalization to the multilevel case.

2. BASIC EQUATIONS

The propagation of multifrequency radiation

$$E=\sum_j E_j \exp(ik_jx-i\omega_jt)$$

in a resonant multilevel system is described by the equations

$$\frac{\partial E_j}{\partial x}+\frac{n}{c}\frac{\partial E_j}{\partial t}=\frac{2\pi\omega_j}{nc}P_j, \tag{1}$$

where P_j is the corresponding component of the resonant polarization of the medium, and n is the nonresonant refrac-

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tive index whose dispersion we neglect. The polarization of the medium is expressed in terms of the density-matrix elements

$$P_j=N\mu_{km}\rho_{mk}, \tag{2}$$

where N is the density of the resonant multilevel particles, μ_{km} is the dipole moment of the transition between the levels k and m, which are at resonance with the frequency ω_j (i.e., $\omega_j\approx\omega_{km}$); the evolution of the density matrix is usually described by the Bloch equation.[2,3] In the coherent approximation, when the relaxation times can be regarded as infinitely long, the material equations of the multilevel medium are much simplified if they are written directly for the amplitudes a_m of the level-filling probability. In the case of a three-level system the equations for the fields and for the level amplitudes take the form

$$\begin{aligned}\dot a_1&=i(\varepsilon_1a_2+\varepsilon_3a_3), & \varepsilon_1'&=ia_1a_2^*;\\ \dot a_2&=i(\varepsilon_1^*a_1+\varepsilon_2a_3), & \varepsilon_2'&=i\varkappa_2^2a_2a_3^*;\\ \dot a_3&=i(\varepsilon_3^*a_1+\varepsilon_2^*a_2), & \varepsilon_3'&=i\varkappa_3^2a_1a_3^*,\end{aligned} \tag{3}$$

where the dimensionless variables $\varepsilon_j=\mu_jE_j/2\hbar\Omega$ are introduced; a dot denotes differentiation with respect to the dimensionless proper time $(t-xn/c)\Omega$, and a prime denotes differentiation with respect to the dimensionless coordinate $xn\Omega/c$. Here

$$\Omega^2=2\pi N\mu_1^2\omega_1/\hbar n^2,$$

$$\varkappa_{2,3}^2=\mu_{2,3}^2\omega_{2,3}/\mu_1^2\omega_1,$$

and the numbering of the levels and transitions is shown in Fig. 1.

Equations (3) were written for a general case when all three transitions are allowed and have commensurate dipole moments μ_i.

In Sec. 3 we consider a case typical for gaseous media, when the total-angular-momentum selection rules allow only two transitions, while the third transition between the upper levels is forbidden ($\mu_2=0$). This situation is of interest from viewpoint of joint propagation of pulses in the SIT regime. We shall describe the form, structure, and interaction of the simultons, and also obtain conditions for their formation.

In Sec. 4 we consider the case when all the μ_i differ from zero, and present solutions that describe the effective parametric interaction of the light pulses.

Both situations can be investigated numerically, but in a number of cases it is possible to obtain analytic solutions of the system (3) by the method of the inverse scattering problem. This method[15] sets in correspondence with the initial nonlinear problem a system of two linear equations for matrices φ whose elements depend on x and t:

$$-i\dot\varphi=A\varphi, \tag{4a}$$

$$i\varphi'=B\varphi. \tag{4b}$$

The operators A and B depend on x and t and are chosen such that the condition for compatibility of (4a) and 4(b)

$$A'+\dot B=i[A,B], \tag{5}$$

be equivalent to the initial system. The construction of the matrices A and B is the principal step for the analytic solution of the nonlinear problem.

Assume that the matrix A can be presented in the form $A=\lambda J+U$, where J is a diagonal matrix with constant elements, λ is a spectral parameter, and U is a matrix with zero diagonal, the so-called potential, with $|U|\to0$ as $|t|\to\infty$, so that

$$\int_{-\infty}^{\infty}|U_{ij}|dt<\infty.$$

It is then possible, using (4a), to construct the scattering matrix $S(\lambda,x_0)$ given a potential $U(t,x)$ specified at the point x_0. Equation (4b) determines the dependence of S on x, and when $U(t,x)$ is reconstructed from $S(\lambda,x)$ the variable x serves as a parameter. The solution of the inverse problem is based on the methods of solving the classical problem of the theory of functions of complex variable, known as the Riemann problem,[15] and is given by a system of integro-differential equation analogous to the Gel'fand-Levitan-Marchenko equation.

It is easy to verify by direct calculation that the matrices A and B can be chosen for the system (3) in the form

$$A=\lambda\begin{pmatrix}-1-\varkappa_2^2 & 0 & 0\\ 0 & 1-\varkappa_2^2 & 0\\ 0 & 0 & 1+\varkappa_2^2\end{pmatrix}+\begin{pmatrix}0 & \varepsilon_1 & \varepsilon_3\\ \varepsilon_1^* & 0 & \varepsilon_2\\ \varepsilon_3^* & \varepsilon_2^* & 0\end{pmatrix},$$

$$B=(1/2\lambda)\rho,\quad \rho_{ij}=a_ia_j^* \tag{6}$$

where ρ is the density matrix of the resonant particles. Equations (5) and (6) are equivalent to Eqs. (3) if the parameters $\varkappa_2$ and $\varkappa_3$ are connected by the relation

$$\varkappa_3^2=1+\varkappa_2^2, \tag{7}$$

the meaning of which will be clarified below.

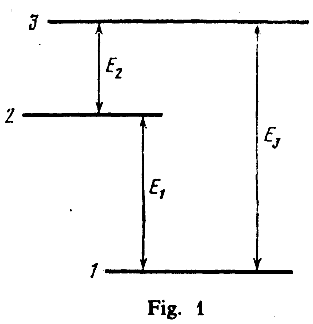

Fig. 1

3. SELF-INDUCED TRANSPARENCY

After writing the equations in the form (4)–(6), the inverse-problem method makes it possible in principle to solve the problem of the evolution of arbitrary pulses that enter into the medium. It is obvious, however, that in a resonantly absorbing medium the entering pulse, at sufficiently large distances, either attenuates as a result of dispersion spreading, or is transformed into one or several pulses of stationary form (solitons). Therefore, in our opinion, the following questions are of interest. First, solitons of what type can propagate (including simultaneous propagation) in the given three-level system; second, how do they interact with one another; third, what are the threshold conditions for the formation of multifrequency solitons.

The construction of soliton solutions[15] reduces to the problem of the construction of definite matrix functions ψ^+ and $\tilde{\psi}^-$ that are respectively analytic in the upper and lower half-plane of the variable λ and have in the region of their analyticity equal numbers of zeros λ_n and $\mu_n = \lambda^*_n$ (with coinciding degrees of degeneracy of $\psi^+(\lambda_n)$ and $\tilde{\psi}^-(\mu_n)$), satisfying on the real axis the condition

$$\tilde{\psi}^-\psi^+=1 \tag{8}$$

and having a unity asymptote at infinity. The matrix U is expressed in terms ψ^+ via the formula

$$U=\lim_{\lambda\to\infty}\lambda[J,\psi^+]. \tag{9}$$

The soliton parameters are determined by the zeros of the Riemann function (8). A detailed exposition of the solution would take too much space. Using the fact that the differences of the solution from the general analysis of the inverse-problem variant considered in detail in Ref. 15 are negligible, we proceed directly to an exposition of the results. The details of the solutions are given in a separate paper.[16]

The simplest single-pole solution describes simultaneous propagation of pulses that are at resonance with different transitions:

$$\varepsilon_{1,3}=\frac{C_{1,3}}{\tau}\operatorname{sech}\left(\frac{t-x/v}{\tau}+\alpha\right). \tag{10}$$

The constants $C_{1,3}$ are connected by the condition $C_1^2+C_2^2=1$, so that the areas $\theta=\int\varepsilon dt$ under the pulses satisfy the relation $(\theta_1^2+\theta_3^2)^{1/2}=2\pi$. In this sense the solution (10) can be regarded as a two-frequency analog of the known 2π-pulse in a two-level system. The duration and velocity of such a pulse are connected by the condition

$$c/nv-1=\Omega^2\tau^2. \tag{11}$$

The solution (10) was obtained earlier[12-14] as a stationary solution of Eqs. (3). It can be added that (10) includes as a particular case the propagation of a single 2π-pulse that is at resonance with one or the other transition ($C_1=0$ or $C_2=0$).

The inverse-problem method makes it possible to study the interaction of simultons. We begin with a two-soliton solution. It describes the propagation and collision of two pulses of the type (19) moving with different velocities. The general form of this solution is quite unwieldy and is not presented here (see Ref. 16), but for the interesting particular case when one pulse moves like a 2π-pulse at resonance with one transition and the second moves (with a different velocity) as a 2π-pulse that is at resonance with the other the solution takes the form

$$\varepsilon_1=\frac{1}{D}\left\{\tau_3\operatorname{ch}\left(\frac{t-x/v_3}{\tau_3}\right)-\left(\frac{1}{\tau_1}+\frac{1}{\tau_3}\right)^{-1}\exp\left(-\frac{t-x/v_3}{\tau_3}\right)\right\},$$

$$\varepsilon_3=\frac{1}{D}\left\{\tau_1\operatorname{ch}\left(\frac{t-x/v_1}{\tau_1}\right)-\left(\frac{1}{\tau_1}+\frac{1}{\tau_3}\right)^{-1}\exp\left(-\frac{t-x/v_1}{\tau_1}\right)\right\}, \tag{12}$$

where

$$D=\tau_1\tau_3\operatorname{ch}\left(\frac{t-x/v_1}{\tau_1}\right)\operatorname{ch}\left(\frac{t-x/v_3}{\tau_3}\right)-\left(\frac{1}{\tau_1}+\frac{1}{\tau_3}\right)^{-2}\times\exp\left(-\frac{t-x/v_1}{\tau_1}\right)\exp\left(-\frac{t-x/v_3}{\tau_3}\right).$$

For brevity, we have left out of these formulas the phase constants that determine the initial position of the solitons.

One can trace the interaction between the pulses by assuming that the faster one, say ε_3, overtakes the pulse ε_1. Equation (12) show that as $t\to-\infty$ the fields are separated single-frequency 2π-pulses:

$$\varepsilon_1=\frac{1}{\tau_1}\operatorname{sech}\left(\frac{t-x/v_1}{\tau_1}\right),\qquad \varepsilon_3=\frac{1}{\tau_3}\operatorname{sech}\left(\frac{t-x/v_3}{\tau_3}\right). \tag{13}$$

After the collision, when the pulses move far enough from one another ($t\to+\infty$) they take the form

$$\varepsilon_1=-\frac{1}{\tau_1}\operatorname{sech}\left(\frac{t-x/v_1}{\tau_1}-\chi\right),\qquad \varepsilon_3=\frac{1}{\tau_3}\operatorname{sech}\left(\frac{t-x/v_3}{\tau_3}+\chi\right). \tag{14}$$

It can be seen, however, that upon collision the amplitude of the slow pulse reverses sign. During the time of the collison the slow pulse slows down and the fast one is accelerated, and the relative shifts are determined by one and the same quantity

$$\chi=\ln[(\tau_1+\tau_3)/(\tau_1-\tau_3)].$$

An analysis of simulton collisions in the general case shows that despite the larger number of degrees of freedom, the character of the interaction of a two-frequency field with a three-level system does not differ qualitatively from the interaction of 2π-pulses (of solitons in a two-level system). Just as in pair collision of solitons in any fully conservative system investigated with the aid of the inverse-problem method, the collisions change only the phases of the simultons, and forward and backward shifts take place of the fast and slow pulses, respectively. The ratio of the field amplitudes, however, is not altered by the collision at both frequencies in each simulton.

More and more complicated multisoliton solutions can be constructed just as the two-pole solution. Of course, they assume an ever more complicated form. We proceed now to the conditions for the appearance of solitons.

Assume that two pulses $\varepsilon_{1,3}(t)$ of known form enter the medium at the boundary $x=x_0$, and that $|\varepsilon_i|\to 0$ as $|t|\to\infty$. We consider two different fundamental matrices of the solutions of Eqs. (4a) at the point x_0, namely φ^+, which has the asymptotic form

$$\varphi^+=\exp(i\lambda Jt),\qquad t\to+\infty, \tag{15}$$

and φ^-, which has the asymptotic form

$$\varphi^-=\exp(i\lambda Jt),\qquad t\to-\infty. \tag{16}$$

Since both matrices $\varphi^\pm$ are fundamental, i.e., made up of linearly independent columes, which are solutions of Eqs. (4a), there exists a nondegenerate matrix $S(\lambda)$, called the scattering matrix, such that

$$\varphi^-=\varphi^+S. \tag{17}$$

The dependence of S on x is obtained from Eq. (4b) on the asymptote $|t|\to\infty$, where we know the matrix B. In the case of a resonantly absorbing medium, all the elements of the density matrix vanish as $|t|\to\infty$, with the exception of $\rho_{11}=1$ (the medium is in the ground state). The zeros of the Riemann problem (8), which determine the parameters of the solitons, are the zeros of the principal minors of the matrix S (Ref. 15).

We consider the case when rectangular pulses ε_1 and ε_3 enter the medium in succession without overlapping and have the frequencies of the corresponding transitions. An analysis of the scattering matrix shows in this case that for the first zero to appear it is necessary that the area of at least one of the pulse exceed π. If this condition is satisfied for only one pulse, then the usual 2π pulse is produced in the corresponding transition. If, however, the areas of both initial pulses exceed π, then two 2π-pulses are produced in the medium in different transitions; this solution is described by Eq. (12), and the second pulse does not necessarily have a larger velocity than the first. These results do not differ from those obtained for a two-level medium.[4,5]

It is easy to obtain the scattering matrix for identical rectangular entering pulses. In this case the first soliton appears at $(\theta_1^2+\theta_3^2)^{1/2}>\pi$, where $\theta_{1,3}$ are the areas of the entering pulses. The solution is then described by Eq. (10) and corresponds to a stationary joint two-frequency soliton.

All the foregoing results are easily generalized to multilevel systems in which pulses interacting with a common lower level propagate. The inverse-problem can be used in the case when the oscillator strengths of all the transitions are equal, $\mu_i^2\omega_i=\text{const}$. Violation of this condition changes the situation substantially. Numerical calculations show that in this case the soliton collison leads to their breakup, to excitation of the medium, etc.

4. PARAMETRIC INTERACTION

We consider now a three-level system in which all three types of transition are possible. The condition (7) relates the oscillator strengths of the transitions

$$\mu_1^2\omega_1+\mu_2^2\omega_2=\mu_3^2\omega_3, \tag{18}$$

or, equivalently, $\Omega_1^2+\Omega_2^2=\Omega_3^2$, where $\Omega_i=2\pi/N\mu_i^2\omega_i/\hbar$. The simplest one-pole solution is of the form

$$\begin{aligned}
\varepsilon_1&=\frac{1}{\tau_1}\left\{\mathrm{ch}\left(\frac{t-x/v_1}{\tau_1}+\alpha_1\right)+A_1\exp\left[\frac{t-x/v_1}{\tau_1}+\frac{x}{L_1}\right]\right\}^{-1},\\
\varepsilon_2&=\frac{1}{\tau_2}\left\{\mathrm{ch}\left(\frac{t-x/v_2}{\tau_2}+\alpha_2\right)\right.\\
&\qquad\left.+A_2\exp\left[-\left(t-\frac{x}{v_2}\right)\left(\frac{1}{\tau_1}+\frac{1}{\tau_3}\right)+\frac{x}{L_2}\right]\right\}^{-1},\\
\varepsilon_3&=\frac{1}{\tau_3}\left\{\mathrm{ch}\left(\frac{t-x/v_3}{\tau_3}+\alpha_3\right)+A_3\exp\left[\frac{t-x/v_3}{\tau_3}-\frac{x}{L_3}\right]\right\}^{-1}
\end{aligned}\tag{19}$$

and describes the decay of a 2π-pulse ε_3 into two 2π-pulses ε_1 and ε_2 or the inverse process of the mixing of the frequencies of the pulses ε_1 and ε_2 and generation of a pulse ε_3. The constants α_i and A_i in (19) determine the initial positions of the solitons. The pulse durations are connected by the condition

$$\Omega_i^2\tau_i=\text{const}, \tag{20}$$

and their velocities are given by

$$c/nv_{1,3}-1=\Omega_{1,3}^2\tau_{1,3}^2,\quad v_2=c/n. \tag{21}$$

The propagation velocity v_2 of the pulse ε_2 turns out to be equal to the velocity of light in the medium. In fact, since all the particles are on the lower level prior to the arrival of the pulses ε_1 and ε_2, the medium is transparent to the field of ε_2. The pulse decay lengths are equal to

$$L_i=\frac{c}{2n}(\Omega_2^2\tau_2)^{-1}\frac{\Omega_i^2}{\Omega_2^2} \tag{22}$$

and turn out to be of the same order as the lengths of the 2π-pulses in the corresponding resonant media. Since the decay takes place at sufficiently short pulses, the condition (18) should be satisfied only with accuracy $\delta\Omega^2/\Omega^2<1$. If (18) is rigorously satisfied, the conversion efficiency is 100%.

An analysis of the coefficients α_i and A_i shows that the phase of the pulse ε_3 is reckoned from the point where the pulses ε_1 and ε_2 collide, i.e., form the point where the maxima of the fields of ε_1 and ε_2 coincide. The condition (18) is none other than the Manley-Rowe relation for the pulses as a whole, i.e., the law of conservation of the total number of photons in a coherent interaction. Indeed, in any three-wave mixing process in a nondissipative medium one photon of frequency ω_3 is produced for each pair of photons ω_1 and ω_2. If the radiation intensities in a transparent nonresonant medium are not optimal from the point of view of the Manley-Rowe relation, the strong pulse is mixed only partially with the weaker one. The remaining radiation leaves the medium freely. In coherent interaction with a resonantly absorbing medium the stable formations are solitons. If we require that the system, be fully conservative, a requirement common to problems solved by the inverse-problem method, only solitons can be produced in processes of decay or mixing of radiation at different frequencies. Otherwise part of the radiation is retained in the medium as a result of dispersion spreading. This reasoning, of course, is not a proof, but explains qualitatively why in this system the relation between the oscillator strengths of three transition, a relation equivalent to the law of conservation of the total number of photons in the interacting 2π pulses, appears as the condition of the applicability of the inverse-problem method.

Let us discuss the possibility of realizing this three-level system for a three-frequency interaction of light pulses. One of the variants is to place the resonant particles in a crystal matrix, where the hindrances are lifted because of the interaction with the environment and all three transitions can become allowed. It is important here to satisfy the condition that the phase velocities of all waves be equal, i.e., the usual condition of spatial synchronism. Thus, introduction of specially chosen impurities into the nonlinear crystal can increase strongly the efficiency of conversion of ultrashort pulses. Moreover, at sufficiently large oscillator strengths and a high density of the resonant particles in the crystal matrix, the dispersion of the refractive index of the matrix

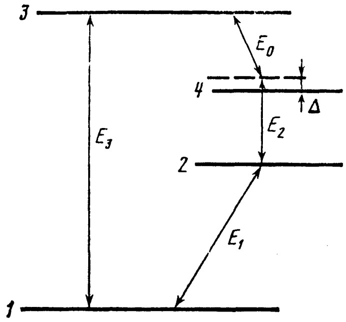

Fig. 2

can become negligible. Since the interaction length $L_{int} \sim c/\Omega^2\tau$ of the pulses in the medium should be less than the phase-synchronism length $2\pi/k\delta n$, and the pulse durations should not exceed the polarization transverse-relaxation time T_2, the dispersion of the crystal-lattice matrix can be neglected under the condition $2\mu^2 T_2 N/\hbar > \delta n$.

In a gaseous media, where the momentum selection rules are satisfied, the indicated interaction scheme cannot be realized without resorting to supplementary fields. However, a three-frequency parametric interaction of the pulses can be effected in a four-level gas medium in the presence of a stationary electromagnetic field. One such scheme is shown in Fig. 2. If the field E_0 is strong enough, i.e., $\mu_{24}E_0\tau_{1,2}/\hbar \gg 1$, and the resonance condition $\omega_3 = \omega_1 + \omega_2 + \omega_0$ is satisfied, the results obtained above are valid. In place of μ_2 it is necessary to substitute in (18) the effective value of the dipole moment of the transition between the levels 2 and 3

$$\mu_{23\,\mathrm{eff}} = \mu_{34}\mu_{42}E_0/\hbar\Delta,$$

where Δ is the deviation of the frequency of the field E_2 from the frequency of the 2–4 transition. By varying the frequency deviation and the amplitude of the field E_0 it is possible to satisfy the condition (18) exactly.

5. CONCLUSION

Within the framework of the inverse-problem method it is possible to obtain also more complicated solutions that describe different variants of soliton and simulton interactions in a three-level system. In the present paper are cited only the simplest of them, which demonstrate the qualitative features of such an interaction. It should be noted that it is possible to obtain in similar fashion analytic solutions that describe nonlinear interaction of coherent pulses in multilevel resonant media with a number of k of levels larger than three. For the use of the method to be valid, the oscillator strengths of the resonant transitions must in this case be connected by $(k-1)(k-2)/2$ conditions similar to (18).

By writing Eqs. (3) in the form (5) and (6) it is possible also to investigate the amplification of multifrequency pulses in an inverted three-level medium. This problem differs in its formulation from the one considered here only by the initial conditions for the density matrix. The amplification of pulses in two-level media was considered in this manner in Refs. 6 and 7. The amplification in a three-level system is the subject of a separate analysis.

We point out in conclusion that all the results were considered on the basis of a one-dimensional treatment. Allowance for the transverse structure of the pulses limits the applicability region because of the development of small-scale transverse perturbations.[8–10] The characteristic lengths of the resonant media at which development of a transverse structure can be neglected amount to $l_p \ln(E_0/E_1)$, where l_p is the length of the pulse in the medium, and E_0 and E_1 are the amplitudes of the field and of the initial transverse perturbations.

The results obtained here are thus applicable for a sufficiently smooth transverse profile of the pulses at the entrance into the resonant medium. The region of one-dimensional treatment is much larger for coherent pulses with off-resonance frequencies, since the growth rate of the transverse instability decreases upon detuning.[9]

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