

Asymptotic behavior of ultrashort light pulses in resonant interactions with a medium

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We investigate the behavior, at large distances, of ultrashort pulses (USP) of radiation with frequencies ω_j satisfying one or several resonance conditions of the type $\sum_j r_j \omega_j = \omega_{m_1}$, where ω_{m_1} is the transition frequency of the molecule of the medium, r_j is the multiplicity of the degeneracy in the frequency ω_j . It is observed that the resonant interactions (RI) are divided into two groups: in one of them production of self-narrowing pulses of self-induced transparency (SIT) is possible, and in the other not. It is shown that parametric RI (when several resonances of the indicated type are present) are characterized by establishment, everywhere except at some individual points, of finite amplitudes at the frequencies of all fields whose USP participate in the process. The use of RI for the conversion of the USP frequencies with simultaneous shortening of their duration is discussed. It is shown that four-photon parametric RI suitable for this purpose make it possible to tune the frequency of the converted radiation in the range from 0 to $2\omega_{m_1}$. It is noted that the energy of the SIT subpulses is calibrated and depends only on the system parameters; this makes measurement of the polarizability of the resonant transition possible. We consider the effect exerted on the possible asymptotic behavior of USP by the following factors: the dispersion of the group velocities, the dispersion spreading of the wave packets, the wave mismatch, the dispersion of the polarizabilities of the energy levels of the resonant transition, the frequency detuning of resonance, the inhomogeneous line broadening, and the inhomogeneity of the light beams in the transverse direction.

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Resonant interactions (RI) of ultrashort pulses (USP) of light with a medium, compared with RI of quasistationary waves, have a number of characteristic features due to the coherence of the interaction. These include first of all the phenomenon of self-induced transparency (SIT), discovered in 1967 by McCall and Hahn for one-photon resonance.¹ Among the other types of RI, the conditions for the onset of SIT (i. e., the conditions for the formation of USP propagating hereafter without change of energy) were investigated for two-photon absorption, stimulated Raman scattering, and third-harmonic generation under conditions of two-photon resonance at the pump frequency (see, e. g., Refs. 2-6).

Self-induced transparency can find a variety of applications, one of the most attractive of which is, in our opinion, its use for substantially shortening the duration of radiation pulses with practically no change in their energy.⁷ In this connection, and recognizing that the foregoing list of resonant interactions is far from complete, it seems appropriate to formulate the conditions for the onset of SIT in RI of general form, as well as to find the asymptotic (steady-state at large distances) distribution of the amplitudes of the fields in the pulse. Such an investigation would clarify the extent to which parametric bleaching (see the book⁸ and the references therein) remains typical of resonant parametric interactions (RPI) on going from quasistationary light wave to USP.

One more remark is in order. An exact analytic description of RI is as a rule impossible. It is quite frequently possible to obtain an approximate analytic solution for the initial stage of the interaction, when the amplitudes of some of the fields are still small.^{6,9} In the experiments, at the same time, the interaction region is by no means small enough to be able to confine

oneself to the initial stage. To determine the character of the subsequent behavior of the fields in similar situations the problem is usually solved with a computer.¹⁰⁻¹² This, however, does not tell us whether the distances at which the solution is already close to the steady state has been reached, or whether the calculation must be continued and the form of the solution will change qualitatively in the succeeding stages. Knowledge of the asymptotic form of the solutions permits a correct determination of the instant when the calculation can be stopped. A study of the asymptotic form is therefore important also for obtaining the complete picture of the interaction by numerical methods. In this respect, (as well as possibly in several others), the investigation of the asymptotic behavior of USP in RI is just as useful as, e. g., the investigation of the solutions near the equilibrium states in limit cycles in the theory of oscillations of a system with lumped parameters.¹³

1. INITIAL RELATIONS

We consider the interaction of an electromagnetic radiation of the form

$$E = \sum_j E_j = \sum_j C_j \exp[i(\omega_j t - k_j z)] + \text{c.c.} \quad (1)$$

with a medium characterized by transition frequencies ω_{mn} between the energy states m and n . Assume that the resonance conditions

$$\sum_j r_{js} \omega_j = \omega_{m_1} + \nu_s \quad (2)$$

are satisfied for one of the transitions from the ground states; we assume that the levels 1 and m are not degenerate. In Eqs. (1) and (2), ω_j and k_j are the frequency and wave number of the j th wave, r_{js} is the multiplicity of the degeneracy of the s th resonance with re-

spect to the frequency ω_j , and ν_s is the frequency detuning from the s th resonance (we put henceforth $\nu_s = \nu$). The order of the s th resonance is $q_s = \sum_j r_{js}$.

To describe such an interaction we use the equations

$$\left[\frac{\partial}{\partial \mathbf{z}} + \frac{1}{v_j} \frac{\partial}{\partial t} + \frac{i}{2k_j} \left(\Delta - \frac{1}{v_j} \frac{\partial^2}{\partial t^2} - k_j \frac{\partial^2 k}{\partial \omega^2}(\omega_j) \frac{\partial^2}{\partial t^2} \right) \right] C_j = -\frac{2\pi i \omega_j N}{n_j c} \left[\frac{1}{2} (\kappa_j^{mm} - \kappa_j^{11}) C_j \int \eta g(\nu) d\nu + \left(\exp \sum_i \tilde{r}_{i, k_i} \right) r_{j, \kappa_s} (C_j^*)^{r_{j, \kappa_s} - 1} \prod_{i \neq j} (C_i^*)^{r_{i, \kappa_s}} \int \sigma g(\nu) d\nu \right], \quad (3)$$

which are obtained when the material-atom polarization obtained in Refs. 14 and 15 for resonant processes is substituted in the abbreviated equations for the amplitudes C_j .¹⁶ Here t and z are the time and longitudinal coordinate; n_j is the refractive index at the frequency ω_j ; κ_j^{mm} and κ_j^{11} are the polarizabilities, at the frequency ω_j of the molecule (atom) in the m th and first energy states, respectively; κ_s is the polarizability of the transition $1 \rightarrow m$ for the s th resonance; N is the number of molecules (atoms) per unit volume; v_j is the group velocity at the frequency ω_j ; Δ is the three-dimensional Laplace operator; $g(\nu)$ is the distribution function, normalized to unity, which characterizes the inhomogeneous line broadening of the resonant transitions ($\int g(\nu) d\nu = 1$); the tilde over the summation or product sign means that these operations must be carried out over the indices of the frequencies contained in the s th resonance (the one corresponding to r_{j, κ_s}). Equations (3) take into account the dispersion spreading of the momenta and the diffraction of the wave beams. If we make the substitution $\omega_j \rightarrow -\omega_j$ in the resonance condition (2), then $C_j \rightarrow C_j^*$ in (3). The slow components of the population difference η and of the off-diagonal element of the density matrix σ satisfy the equations of the generalized two-level system,¹⁷ which for pulses with duration τ_p much shorter than the longitudinal (τ) and transverse (T) relaxation times take the form

$$\begin{aligned} \partial \sigma / \partial t - i(\Omega + \nu) \sigma &= i \hbar^{-1} W \eta, \\ \partial \eta / \partial t &= -4 \hbar^{-1} \text{Im}(\sigma W^*), \end{aligned} \quad (4)$$

where the matrix element of the Hamiltonian of the average motion W and the Stark shift Ω are given by

$$W = - \sum_s \kappa_s \prod_j C_j^{r_{j, \kappa_s}}, \quad \Omega = \hbar^{-1} \sum_{j > 0} (\kappa_j^{mm} - \kappa_j^{11}) |C_j|^2.$$

If some frequency ω_j is present in several of resonance conditions (2), then it is necessary to sum in the right-hand side of Eqs. (3) the terms pertaining to these resonances, with allowance for the sign of ω_j in (2).

We neglect in (3) the terms with the second derivatives and put $g(\nu) = \delta(\nu)$ and $v_j = v$ (the role of the dispersion of the group velocities, of the dispersion and diffraction spreading of the pulses, as well as of the inhomogeneous broadening will be dealt with later on). Then, changing over to equations for the real amplitudes A_j and phases φ_j ($C_j = A_j \exp(-i\varphi_j)$), we get

$$\frac{\partial A_j}{\partial \xi} = -\frac{\pi \omega_j N \kappa_s}{n_j c} A_j^{r_{j, \kappa_s} - 1} r_{j, \kappa_s} \prod_{i \neq j} A_i^{r_{i, \kappa_s}} (R \cos \psi_s + I \sin \psi_s), \quad (5)$$

$$\begin{aligned} \frac{\partial \psi_s}{\partial \xi} &= \delta k_s + \frac{\pi N}{c} \left\{ \eta \left[\sum_j \tilde{r}_{j, \kappa_s} \frac{\kappa_j^{mm} - \kappa_j^{11}}{n_j} \omega_j - \sum_j r_{j, \kappa_s} \frac{\kappa_j^{mm} - \kappa_j^{11}}{n} \omega_j \right] + I \kappa_s \sum_j \frac{r_{j, \kappa_s} \omega_j}{n} A_j^{r_{j, \kappa_s} - 2} \prod_{i \neq j} A_i^{r_{i, \kappa_s}} - \kappa_s \sum_j \frac{r_{j, \kappa_s} \omega_j}{n_j} A_j^{r_{j, \kappa_s} - 2} \prod_{i \neq j} A_i^{r_{i, \kappa_s}} (I \cos \psi_s - R \sin \psi_s) \right\}, \end{aligned} \quad (6)$$

$$\frac{\partial R}{\partial \xi} - \left(\frac{\Omega}{v} - \frac{\partial \Phi_1}{\partial \xi} \right) I = -\frac{2}{v \hbar} \sum_s \kappa_s \prod_j A_j^{r_{j, \kappa_s}} \cos \psi_s \eta, \quad (7)$$

$$\frac{\partial I}{\partial \xi} + \left(\frac{\Omega}{v} - \frac{\partial \Phi_1}{\partial \xi} \right) R = -\frac{2}{v \hbar} \sum_s \kappa_s \prod_j A_j^{r_{j, \kappa_s}} \sin \psi_s \eta, \quad (8)$$

$$\eta^2 + R^2 + I^2 = 1, \quad (9)$$

where

$$\xi = z, \quad \zeta = z - vt, \quad \psi_s = \Phi_1 - \Phi_s, \quad \Phi_s = \sum_j r_{j, \kappa_s} (\varphi_j + k_j z),$$

$$\delta k_s = \sum_j r_{j, \kappa_s} k_j - \sum_j r_{j, \kappa_s} k_j; \quad R, I = \text{Re}, \text{Im}[2i\sigma \exp(i\Phi_1)].$$

We begin the investigation of these equations with the simplest case, when the wave mismatch $\delta k_s = 0$, and the difference between the polarizabilities κ_j^{mm} and κ_j^{11} can be neglected.

In typical experiments dealing with frequency conversion upon entering a medium, the pump fields whose frequencies participate in one of the resonances (pump channel, $s=1$) are different from zero, and one of the fields A_j pertaining to the remaining resonances (the conversion channels) is absent. Then in the case when $r_{j, \kappa_s} = 1$ and there is no phase modulation of the pump at the entrance into the medium, the phase difference ψ_s assumes practically instantaneously, according to (6), a value zero or π , depending on the sign of $\sin \psi_s$ in (6). Thus, just as in quasistationary resonant parametric interactions,^{8, 18} phase locking of the interacting waves takes place here. The differences of the phases ψ_s subsequently remain unchanged until the amplitude of one of the fields A_j again vanishes. At that instant, the phase φ_j can change jumpwise by π . We see therefore that the amplitudes C_j always remain real (if the phases are reckoned from the input phases of the pump); only their sign can change. Taking this into account, we can write,¹⁾ after integrating (7)–(9) and substituting σ and η in (3), the following equation for the amplitude C_j

$$\frac{\partial C_j}{\partial \xi} = \frac{\pi N \kappa_s r_{j, \kappa_s} \omega_j}{n_j c} C_j^{r_{j, \kappa_s} - 1} \prod_{i \neq j} C_i^{r_{i, \kappa_s}} \sin \Theta(\xi), \quad (10)$$

where we have introduced the area under the pulse

$$\Theta(\xi) = \frac{2}{v \hbar} \int_{\xi}^{\cdot} W d\xi.$$

It can be verified that if frequencies with $r_{j, \kappa_s} \geq 2$ are present in a certain conversion channel, and $A_j = 0$ at the input, then no field of frequency ω_j will be produced, and the processes that take place in the remaining channels will proceed as if the channel in question were absent. We therefore exclude parametric processes of this type from consideration.

Equations (10) are applicable to multiphoton processes in which there is only one resonance.

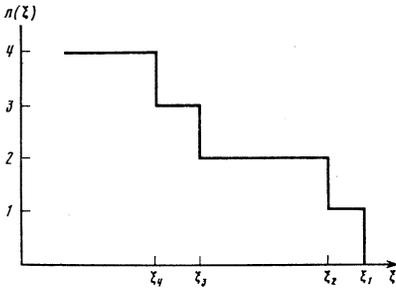


FIG. 1. Example of steady-state distribution of the pulse area $\Theta(\xi)$ (accurate to a factor 2π).

For systems with an arbitrary number of resonances it is easy to obtain from (10)

$$\frac{\partial}{\partial \xi} \left(\sum_j \frac{n_j |C_j|^2}{\omega_j r_j} d\xi \right) = \frac{\pi N v \hbar}{c} [\cos \Theta(\xi) - 1], \quad (11)$$

$$\frac{n_j |C_j(\xi, \xi)|^2}{r_j \omega_j} - \frac{n_i |C_i(\xi, \xi)|^2}{r_i \omega_i} = \frac{n_j |C_j(\xi=0, \xi)|^2}{r_j \omega_j} - \frac{n_i |C_i(\xi=0, \xi)|^2}{r_i \omega_i}. \quad (12)$$

From (11), using (12) and (2), we easily obtain the expression

$$\frac{\partial}{\partial \xi} \sum_j \int_0^{\xi} n_j |C_j|^2 d\xi = \frac{\pi v N \omega_m \hbar}{c} [\cos \Theta(\xi) - 1], \quad (13)$$

from which it follows, first, that the energy in the pulse does not increase and, second, that in any interval $\Delta \xi = \xi_2 - \xi_1$ a constant field-energy flux density is established sooner or later. A stationary distribution of the energy in a pulse is established if we have for any ξ

$$\Theta(\xi) = \frac{2}{v \hbar} \int_0^{\xi} W d\xi = 2\pi n(\xi), \quad (14)$$

where $n(\xi)$ is a staircase function that assumes only integer values (one of the examples is shown in Fig. 1). With the aid of (10) and (14) it can be shown that establishment of the distribution of the total energy within the limits of the pulse leads to establishment of a stationary distribution of the amplitudes of each of the fields. Obviously, this distribution must be such that

$$W = \pi v \hbar \sum_i \Delta n_i [\delta(\xi - \xi_i)]^p \quad (15)$$

[Δn_i is an integer equal to the change of the function $n(\xi)$ at the point $\xi = \xi_i$; $\delta(\xi - \xi_i)$ is a δ -function; $0 \leq p_i \leq 1$]. The requirement that the pulse duration at the input be limited excludes the possibility of the appearance of pulses moving with velocity less than v (similar to the pulses considered in Ref. 2), if among the resonances (2) there are no first-order resonances, and the spontaneous transitions can be neglected. The interactions that occur under conditions when resonances with $q=1$ are present will be considered in Sec. 6.

2. MULTIPHOTON INTERACTIONS

We consider now processes that occur if only one of the resonances of type (2) is present. If some of the frequencies in (2) are negative, one refers to a Raman process; on the other hand if all the frequencies in (2) are positive, then we deal with multiphoton absorption. In the case of Raman interactions the amplitudes of the

fields C_j are bounded by the initial conditions [see (12)], i. e., $|W|$ is a bounded quantity and therefore the only value of W for steady-state fields is $W=0$. From this, using (12), we find that at large distances ($\xi \rightarrow \infty$) there are established the following flux densities of the photon numbers $M_j = n_j C_j^2 / r_j |\omega_j|$:

$$M_j(\xi) = M_{j0}(\xi) \mp M_{i0} r_j / r_i, \quad (16)$$

where the upper sign corresponds to $\omega_j > 0$ (radiation of the pump) and the lower to $\omega_j < 0$ (scattered radiation); $M_{i0} = \min\{n_j C_{j0}^2 / r_j \omega_j\}$ [in the last relation, $\omega_j > 0$ and $C_{j0} = C_j(\xi=0)$]. In particular, for ordinary stimulated Raman scattering (SRS) we have as $\xi \rightarrow \infty$

$$M_p(\xi) = 0, \quad M_s(\xi) = M_{p0}(\xi) + M_{s0}(\xi),$$

just as in stationary SRS (the subscripts "p" and "s" pertain to the pump and to the Stokes components).

In systems with multiphoton absorption, the amplitudes C_j are not restricted by the conditions (12). The steady-state values of M_j are

$$M_j = M_{j0} - M_{i0} r_j / r_i \quad (17)$$

everywhere except at the points ξ_i where a burst of W is possible [see (15)], and consequently also of C_j . Obviously, since $|C_j| \gg |C_{j0}|$ in this case, the fields at these points satisfy the proportionality condition

$$n_j C_j^2 / r_j \omega_j = n_i C_i^2 / r_i \omega_i. \quad (18)$$

On the basis of numerical solutions of the equations that describe two-photon absorption (TPA) and Raman interaction of ultrashort pulses, the authors of Refs. 10 and 11 have concluded that sufficiently intense pulses, such that $\Theta \geq 2\pi n$ (n is an integer) break up into n individual subpulses, which decrease in duration and increase in power as they move in the medium. Our asymptotic solution (16) for SRS does not contain short subpulses, and is determined only by the input values of C_{p0} and C_{s0} . Consequently, in the case of SRS the breakup into subpulses and their narrowing takes place only up to definite distances, beyond which the subpulses begin to be smoothed out.

In the case of multiphoton absorption, on the contrary, the narrowing and the growth of the amplitude of the subpulses can proceed without limit within the framework of the given model. Starting from the definition of W , from the condition of proportionality of the fields (18), and from expression (15), it is easy to see that the limiting energy of the i th subpulse at the frequency of each of the fields is proportional to $[\delta(\xi - \xi_i)]^{2p/q} d\xi$, and consequently, as the subpulse evolves the SIT tends to zero in systems with resonances of order higher than the second. Therefore the use of resonances with $q > 2$ to obtain intense and short pulses is ineffective, since the energy of such a pulse decreases substantially when the pulse becomes sharper. This conclusion is valid also for resonant parametric interactions, in which the order of the resonance in the conversion channel is larger than two. In systems with $q=2$, the energy of the subpulses is damped at any $p \neq 1$. We shall therefore be interested in Secs. 3-5 only in subpulses with $p=1$. Systems containing resonances with $q=1$ will be considered in Sec. 6.

The results obtained here can be used to determine the asymptotic behavior of nonsynchronous resonant parametric frequency conversion (of the type considered in Ref. 4). Namely, regardless of the type of resonance in which the pump-pulse fields participate, it is impossible to realize SIT in the second pulse, if the frequencies of the fields in this pulse satisfy a resonance condition of the Raman type; on the other hand if all the frequencies of the fields in the second pulse enter in the resonance relation (2) with equal signs, then δ -shaped subpulses W should be produced under certain conditions imposed on the input values of the fields and on the frequency.

3. RESONANT FOUR-PHOTON PARAMETRIC INTERACTIONS; ALL $\omega_j > 0$

We proceed to resonant parametric interactions. We confine ourselves here to excitation of resonant four-photon parametric interactions (RFPI), which have been the subject of the largest number of experimental studies. Resonant four-photon parametric interactions can be divided into two groups. In the first of them are satisfied two resonance conditions of the TPA type: all $\omega_j > 0$. In the second group, one of the resonances corresponds to Raman interaction, and the other to two-photon absorption (one of the frequencies ω_j is negative and the remaining ones positive). The third group consists of RFPI, in which both resonances are of the Raman type.

If all $\omega_j > 0$, then we easily obtain from (10) the first integral

$$\frac{a_1 + a_2}{a_{10} + a_{20}} = \left(\frac{a_3 + a_4}{a_{30} + a_{40}} \right)^\gamma, \quad (19)$$

where

$$a_j = C_j (n_j / \omega_j)^{1/2}, \quad a_{j0} = C_{j0} (n_{j0} / \omega_j)^{1/2}, \\ \gamma = \frac{\kappa_p}{\kappa_c} \left(\frac{\omega_1 \omega_2 n_3 n_4}{\omega_3 \omega_4 n_1 n_2} \right)^{1/2},$$

κ_p and κ_c are respectively the polarizabilities of the transitions between the levels 1 and m in the pump and conversion channels. For RFPI of general type, if the resonances are not degenerate in frequency, the corresponding first integral is of the form

$$\rho_1 e^{i f_1 t} = \rho_2 e^{i f_2 t}, \quad (20)$$

where

$$\rho_1 = \left| \frac{a_1 + a_2}{a_{10} + a_{20}} \right|, \quad \rho_2 = \left| \frac{a_3 + a_4}{a_{30} + a_{40}} \right|, \\ f_1 = \arg \frac{a_1 + a_2}{a_{10} + a_{20}}, \quad f_2 = \arg \frac{a_3 + a_4}{a_{30} + a_{40}}. \quad (21)$$

Inasmuch as in the course of establishment of the stationary distribution of W there should take place an unlimited growth of $W(\xi_j)$, the amplitude of at least one of the fields will increase without limit at the points ξ_j . It follows from (12) and (19) that at these points the amplitudes of all the interaction fields should tend to infinity simultaneously. The pulses in both the conversion channel and in the pump channel then become proportional ($a_1 = a_2$; $a_3 = a_4$), since it turns out that $a_j(\xi_j) \gg a_{j0}(\xi_j)$. Using this fact as well as (12), (15), and (19), we can write down near the point ξ_j (we assume

that $\Delta n_j = -1$)

$$-W = \beta \left[\gamma \left(\frac{a_{10} + a_{20}}{2} \right)^2 \left(\frac{2}{a_{30} + a_{40}} \right)^{2\gamma} a_1^{2\gamma} a_2^{2\gamma} \right] \\ = \beta \left[\gamma a_1^2 + \left(\frac{a_{30} + a_{40}}{2} \right)^2 \left(\frac{2}{a_{10} + a_{20}} \right)^{2/\gamma} a_1^{2/\gamma} \right] = \pi v \hbar \delta(\xi - \xi_j), \quad (22)$$

where $\beta = \kappa_c (\omega_3 \omega_4 / n_3 n_4)^{1/2}$. From this and from the requirement that the pulse energy be finite it follows that if $\gamma > 1$, then near ξ_j ;

$$a_1^2 = a_2^2 \sim \delta(\xi - \xi_j), \quad a_3^2 = a_4^2 \sim [\delta(\xi - \xi_j)]^{1/\gamma},$$

the energies of the pump fields passing through a unit area during the i th SIT pulse are equal to

$$U_{1,2}(\xi_i) = \frac{\omega_{1,2c}}{2\pi v_{1,2}} \int_{\xi_i - \epsilon}^{\xi_i + \epsilon} a_{1,2}^2 d\xi \approx \frac{v \hbar n_{1,2}}{2\kappa_p} \left(n_1 n_2 \frac{\omega_{1,2}}{\omega_{2,1}} \right)^{1/2}, \quad (23)$$

and the corresponding energies at the frequencies of the triggering and generated fields are equal to zero.

On the other hand if $\gamma < 1$, then the energies $U_{1,2}$ of the pump pulses turn out to be zero, and

$$U_{3,4}(\xi_i) \approx \frac{v \hbar n_{3,4}}{2\kappa_c} \left(n_3 n_4 \frac{\omega_{3,4}}{\omega_{4,3}} \right)^{1/2}. \quad (24)$$

[In this case $a_1^2 = a_2^2 \sim [\delta(\xi - \xi_j)]^\gamma$ and $a_3^2 = a_4^2 \sim \delta(\xi - \xi_j)$. When $\gamma = 1$ all $a_j^2 \sim \delta(\xi - \xi_j)$, and the relations between $U_{1,2}(\xi_j)$ and $U_{3,4}(\xi_j)$ depend on the values of the fields as they enter into the medium. For example,

$$U_i(\xi_i) = \frac{n_4 \omega_4}{n_1 \omega_1} \left[\frac{a_{30} + a_{40}}{a_{10} + a_{20}} \right]_{\xi_i}^2 U_1(\xi_i) \\ = \frac{v \hbar}{2\kappa_c} \left(\frac{\omega_4}{\omega_3} n_3^3 n_4 \right)^{1/2} \left[\frac{(a_{30} + a_{40})^2}{(a_{10} + a_{20})^2 + (a_{30} + a_{40})^2} \right]_{\xi_i}.$$

If two proportional pulses ($a_{10} = a_{20}$, $a_{30} = a_{40}$) enter a medium with $\gamma = 1$, then their behavior is described by the solution

$$a_1 = a_2 = \frac{a_{10}}{a_{30}} a_3 = \frac{a_{10}}{a_{30}} a_4 = a_{10} \left\{ \sin^2 \frac{\Theta_0}{2} \left[1 + \left(\text{ctg} \frac{\Theta_0}{2} + \Gamma \xi \right)^2 \right] \right\}^{-1/2} \\ \left(\Theta_0 = \frac{2\alpha}{v \hbar} \int_{\xi}^0 (a_{10}^2 + a_{30}^2) d\xi, \quad \Gamma = \frac{2\pi N \alpha}{c}, \quad \alpha = \beta \gamma \right). \quad (25)$$

It follows from (25) that δ -like pulses with energies U_j that coincide with those obtained from the asymptotic expressions are produced at the frequencies of all the fields.

In all the remaining points ($\xi \neq \xi_j$), parametric bleaching is established [$W(\xi) = 0$, see (15)]. To find the steady-state values of the fields it is necessary here to use, besides the condition $W = 0$, also the first integrals (12) and (19). These values turn out to be the same as in quasistationary interaction,¹⁹ since the sets of relations for their determination are perfectly identical in both cases.

It is of interest to consider also the initial stage of conversion, when $\gamma a_1 a_2 \gg a_3 a_4$. We consider the situation most frequently encountered in experiments, when the pump frequency is equal to half the transition frequency (we note that if the frequencies ω_1 and ω_2 are different, and the pump pulse at the input is proportional, i. e., $a_{10} = a_{20}$, then the results will be the same). In this case it is easy to obtain for the problem of frequency conversion ($a_{40} = 0$) from (10)

$$a_{3,4} = \frac{a_{30}}{2} \left(\left| \sin \frac{\Theta}{2} / \sin \frac{\Theta_0}{2} \right|^{1/\gamma} \pm \left| \sin \frac{\Theta_0}{2} / \sin \frac{\Theta}{2} \right|^{1/\gamma} \right), \quad (26)$$

$$a_1 = a_2 = a_{10} \left| \sin \frac{\vartheta}{2} / \sin \frac{\vartheta_0}{2} \right|. \quad (27)$$

In (26), the plus sign pertains to a_3 and the minus sign to a_4 . The area under the pump pulse

$$\vartheta(\xi) = \frac{2\alpha}{v\hbar} \int_0^{\xi} a_1 a_2 d\xi$$

varies in accord with the law⁷

$$\operatorname{ctg} \frac{\vartheta}{2} = \operatorname{ctg} \frac{\vartheta_0}{2} + \Gamma \zeta, \quad (28)$$

and $\vartheta_0(\xi) = \vartheta(\xi, \zeta = 0)$. Equations (26) differ from the corresponding expressions in Ref. 9 because in the latter it was assumed that $a_{30} = a_{40} \neq 0$.

It is obvious from (26) that the energies of the fields a_3 and a_4 first increase independently of the value of γ and of the coordinate ξ (i. e., both in the region where parametric bleaching should become established, and at the points where SIT pulses can be produced). It is interesting to note that at $\gamma > 1$ formulas (26) and (27) describe quite well the interaction up to its asymptotic behavior. In particular, it is easy to find with the aid of (26) that, as expected, the energy of the SIT pulses at the frequencies ω_3 and ω_4 decrease to zero at infinity, whereas the energies of the pump subpulses assume constant values. At $\gamma < 1$ the amplitudes of the subpulses of fields a_3 and a_4 increase more rapidly than the amplitudes of the pump subpulses, as a result of which the condition $\gamma a_1 a_2 \gg a_3 a_4$ is violated.

4. RESONANT FOUR-PHOTON PARAMETRIC INTERACTIONS; ONE OF THE FREQUENCIES ω_j IS NEGATIVE

We assume for the sake of argument that the negative frequency is ω_2 , regardless of the resonance to which the pump channel corresponds. In this case γ in (20) is imaginary, $\rho_1 = 1$, $f_1 = |\gamma| \ln \rho_2$, and $f_2 = -1 |\gamma|^{-1} \ln \rho_1 = 0$. The fields a_j take in terms of the variables $\rho_{1,2}$ and $f_{1,2}$ the form

$$a_1 = a_{10} \cos f_1 + a_{20} \sin f_1, \quad a_2 = a_{20} \cos f_1 - a_{10} \sin f_1; \quad (29)$$

$$a_3 = \frac{1}{2} [a_{30} (\rho_2 + \rho_2^{-1}) + a_{40} (\rho_2 - \rho_2^{-1})], \quad (30)$$

$$a_4 = \frac{1}{2} [a_{30} (\rho_2 - \rho_2^{-1}) + a_{40} (\rho_2 + \rho_2^{-1})].$$

Using the relations written out here and the requirement $W(\xi) = 0$, it is easy to obtain the expression that determines the values of the field a_j for those points where parametric bleaching is established:

$$(a_{30}^2 + a_{40}^2) (\rho_2^2 - \rho_2^{-2}) + 2a_{30} a_{40} (\rho_2^2 + \rho_2^{-2}) = 4|\gamma| \left[\frac{a_{10}^2 - a_{20}^2}{2} \sin(2|\gamma| \ln \rho_2) - a_{10} a_{20} \cos(2|\gamma| \ln \rho_2) \right].$$

As follows from (15) and from the conservation laws (12), the SIT regime can be realized only in that channel in which both frequencies are positive. In analogy with the preceding section, we can determine the energy of the SIT subpulses at the frequencies ω_3 and ω_4 :

$$U_{3,4}(\xi_i) = \frac{v\hbar n_{3,4}}{2\kappa} \left(n_3 n_4 \frac{\omega_{3,4}}{\omega_{1,2}} \right)^{1/2},$$

where κ is the polarizability of the two-photon transition between the levels 1 and m under the influence of the fields C_3 and C_4 . Then $\rho_2(\xi_i)$ increases without

limit in the course of formation of the δ -like subpulse, and this leads, as seen from (29), to a continuous energy exchange between the frequencies ω_1 and ω_2 .

We see thus that generation of intense short (in the limit, δ -like) pulses in the course of upward frequency conversion on the basis of TPA of the pump is impossible in frequency-nondegenerate RFPI. As we shall see below, this is possible when one and the same frequency takes part in both resonances but with opposite signs:

$$\omega_1 + \omega_2 = \omega_3 - \omega_4 = \omega_{m1}. \quad (31)$$

The changes of the amplitudes of the interacting fields $a_j = C_j(n_j/\omega_j)^{1/2}$ is described by the equations

$$\begin{aligned} \partial a_1 / \partial \zeta &= \pi N c^{-1} \alpha a_2 \sin \Theta(\xi), \\ \partial a_2 / \partial \zeta &= \pi N c^{-1} (\alpha a_1 - \beta a_3) \sin \Theta(\xi), \\ \partial a_3 / \partial \zeta &= \pi N c^{-1} \beta a_2 \sin \Theta(\xi), \end{aligned} \quad (32)$$

from which follow the integrals of motion

$$a_3^2 + a_4^2 - a_1^2 = a_{30}^2 + a_{40}^2 - a_{10}^2, \quad (33)$$

$$\alpha a_1 - \beta a_3 = \alpha a_{10} - \beta a_{30}. \quad (34)$$

Here

$$\begin{aligned} \Theta(\xi) &= \frac{2}{v\hbar} \int_0^{\xi} W d\xi, \quad W = -a_2 (\alpha a_1 + \beta a_3), \\ \alpha &= \kappa_{\text{TPA}} \left(\frac{\omega_1 \omega_2}{n_1 n_2} \right)^{1/2}, \quad \beta = \kappa_{\text{SRS}} \left(\frac{\omega_2 \omega_3}{n_2 n_3} \right)^{1/2}, \end{aligned}$$

$\gamma = \alpha/\beta$, and κ_{TPA} and κ_{SRS} are the polarizabilities corresponding to the $1 \leftrightarrow m$ transition in TPA and SRS. Just as in the processes considered above, in this case a parametric bleaching regime is also established (everywhere except at the individual points ξ_i). Using (33) and (34) and the fact that W is zero, we obtain for the stationary values of $a_j(\xi)$

$$a_1 = \frac{\beta(\beta a_{10} - \alpha a_{30})}{\alpha^2 + \beta^2}, \quad a_3 = \frac{\alpha(\alpha a_{30} - \beta a_{10})}{\alpha^2 + \beta^2}$$

$$a_2 = [a_{20}^2 + a_{30}^2 - a_{10}^2 + (\beta^2 - \alpha^2)(\alpha^2 + \beta^2)^{-2} (\alpha a_{30} - \beta a_{10})^2]^{1/2}.$$

The SIT regime is realized at the points ξ_i where $|\Theta(\xi_i + \varepsilon) - \Theta(\xi_i - \varepsilon)| = 2\pi$. At these points all $a_j^2 \sim \delta(\xi - \xi_i)$. The energies passing through a unit area during the SIT subpulse are equal to

$$\begin{aligned} U_1 &= \gamma^2 \frac{\omega_1 n_1}{\omega_3 n_3} U_3, \quad U_2 = (\gamma^2 - 1) \frac{\omega_2 n_2}{\omega_3 n_3} U_3, \\ U_3 &= \frac{v\hbar (n_2 n_3^2 \omega_3)^{1/2}}{2\kappa_{\text{SRS}} (\gamma^2 + 1) [\omega_2 (\gamma^2 - 1)]^{1/2}}. \end{aligned} \quad (35)$$

It follows therefore that the considered process is of interest from the point of view of obtaining near- δ -function pulses of radiation that is tunable in frequency, not only in the case of upward conversion of the frequency on the basis of the TPA of the pump, but also for downward conversion on the basis of SRS of the pump (ω_3 is the pump frequency and ω_2 is the frequency of the Stokes component). We recall that no such pulses can be produced in all other processes with SRS in the conversion channel.

It is easy to deduce from (33) and (34) that if $a_{10} = \gamma a_{30}$ and $a_{20} = a_{30}(\gamma^2 - 1)^{1/2}$, then also subsequently $a_1 = \gamma a_3$ and $a_2 = a_3(\gamma^2 - 1)^{1/2}$. In this case Eqs. (32) have an exact solution

$$a_1 = a_{10} \left\{ \sin^2 \frac{\Theta_0}{2} \left[1 + \left(\operatorname{ctg} \frac{\Theta_0}{2} + K \zeta \right)^2 \right] \right\}^{-1/2}, \quad (36)$$

where

$$\Theta_0 = \frac{2\alpha}{v\hbar} (1+\gamma^{-2})(1-\gamma^{-2})^{1/2} \int_0^{\xi} a_{10}^2 d\xi, \quad K = \frac{2\pi N\alpha}{c} (1-\gamma^{-2})^{1/2}.$$

The solution (36) coincides in form with the law of the change of the fields in TPA of proportional pulses,^{3,7} but the conditions for the proportionality of the pulse in the solution obtained here differ from the corresponding conditions in TPA. In the presence of third-harmonic generation, for which the resonant conditions are a particular case of (31) at $\omega_2 = \omega_1$, there also exists a proportional regime described by a formula of the type (36), where the proportionality condition and the expressions for Θ_0 and K differ from those given above [$a_3 = \gamma(1 - \sqrt{1 - \gamma^{-2}})a_1$, Ref. 6]:

$$\Theta_0 = \frac{2\alpha}{v\hbar} (2 - \sqrt{1 - \gamma^{-2}}) \int_0^{\xi} a_{10}^2 d\xi, \quad K = \frac{2\pi N\alpha}{c} (1 - \sqrt{1 - \gamma^{-2}}).$$

An analysis of formulas (35) and (36) shows that realization of a proportional regime and the onset of δ -function subpulses are possible only at $\gamma > 1$.

For the initial stage of frequency conversion on the basis of TPA of proportional pump pulses ($a_{10} = a_{20}$, $a_{30} = 0$) we can obtain the solutions

$$\begin{aligned} a_{1,2} &= a_{10} \left| \sin \frac{\Phi}{2} / \sin \frac{\Phi_0}{2} \right|, \\ a_3 &= a_{10} \gamma^{-1} \left(\left| \sin \frac{\Phi}{2} / \sin \frac{\Phi_0}{2} \right| - 1 \right) \end{aligned} \quad (37)$$

[$\Phi(\xi)$ is the same quantity as in (26)–(28)]. These solutions are valid when

$$\gamma a_{1,2} \gg a_3, \quad \left| \frac{2\beta}{v\hbar} \int_0^{\xi} a_2 a_3 d\xi \right| \ll 1.$$

It is seen from (37) that establishment of a proportional regime begins with establishment of a constant ratio a_1/a_3 .

To conclude this section, we present for the initial stage of RFPI solutions that are not degenerate in frequency. In two-photon pumping by a proportional pulse [the pump fields $a_{1,2}$ vary in accord with (37)] we have for the fields in the frequency-conversion channel

$$\begin{aligned} a_3 &= a_{30} \cos f - a_{40} \sin f, \quad a_4 = a_{40} \cos f + a_{30} \sin f, \\ f &= \frac{1}{2|\gamma|} \ln \left\{ \sin^2 \frac{\Phi_0}{2} \left[1 + \left(\operatorname{ctg} \frac{\Phi_0}{2} + \frac{2\pi N\alpha}{c} \zeta \right)^2 \right] \right\}. \end{aligned}$$

In SRS pumping, the initial stage of the interaction of the fields a_j is described by the equations

$$\begin{aligned} a_1 &= a_{10} \left[\frac{a_{10}^2 + a_{20}^2}{a_{10}^2 + a_{20}^2 \exp(-2K_c \zeta \xi)} \right]^{1/2}, \\ a_2 &= a_{20} \left[\frac{a_{10}^2 + a_{20}^2}{a_{10}^2 + a_{20}^2 \exp(-2K_c \zeta \xi)} \right]^{1/2} \exp(-K_c \zeta \xi), \\ a_{3,4} &= \frac{a_{30} + a_{40}}{2} \exp \left\{ -\gamma^{-1} \left[\operatorname{arc} \operatorname{tg} \left(\frac{a_{20}}{a_{10}} \exp(-K_c \zeta \xi) \right) - \operatorname{arc} \operatorname{tg} \frac{a_{20}}{a_{10}} \right] \right\} \\ &\quad \pm \frac{a_{30} - a_{40}}{2} \exp \left\{ \gamma^{-1} \left[\operatorname{arc} \operatorname{tg} \left(\frac{a_{20}}{a_{10}} \exp(-K_c \zeta \xi) \right) - \operatorname{arc} \operatorname{tg} \frac{a_{20}}{a_{10}} \right] \right\}, \end{aligned} \quad (38)$$

where

$$K_c = 2\pi N\alpha^2 (a_{10}^2 + a_{20}^2) / chv.$$

5. RESONANT FOUR-PHOTON PARAMETRIC INTERACTIONS WITH ONE NEGATIVE FREQUENCY IN EACH OF THE RESONANCES

In this section we consider stimulated Raman scattering of biharmonic pumping and the process of raising the radiation frequency on the basis of SRS of the pump (the corresponding processes in quasistationary interactions are the subject of Refs. 20 and 21). In addition, there exist two RFPI that are degenerate in frequency, for which one of the frequencies in each resonance is negative, viz., generation of the second Stokes and anti-Stokes SRS components.

For nondegenerate RFPI of the type considered, γ is real and the quantities ρ_1 , ρ_2 , f_1 , and f_2 in (20) and (21) satisfy the relations

$$\rho_1 = \rho_2 = 1, \quad f_1 = \gamma f_2. \quad (39)$$

The normalized amplitudes a_j are connected with f_1 and f_2 as follows:

$$\begin{aligned} a_1 &= a_{10} \cos f_1 + a_{20} \sin f_1, \quad a_2 = a_{20} \cos f_1 - a_{10} \sin f_1, \\ a_3 &= a_{30} \cos f_2 + a_{40} \sin f_2, \quad a_4 = a_{40} \cos f_2 - a_{30} \sin f_2. \end{aligned} \quad (40)$$

From (15) and from the conservation laws (12) it is easily seen that the only possible steady-state regime is parametric bleaching. Using (39), (40), and the condition $W(\xi) = 0$, we obtain for the stationary distribution of $f_1(\xi)$

$$\begin{aligned} \sin \left(2f_1 + \operatorname{arc} \sin \frac{2a_{10}a_{20}}{a_{10}^2 + a_{20}^2} \right) &= -\gamma^{-1} \frac{a_{30}^2 + a_{40}^2}{a_{10}^2 + a_{20}^2} \\ &\times \sin \left(2f_1 \gamma^{-1} + \operatorname{arc} \sin \frac{2a_{30}a_{40}}{a_{30}^2 + a_{40}^2} \right). \end{aligned} \quad (41)$$

An analysis of (41) shows that if $\gamma < 1$ and the quantity $\gamma^{-1}(a_{30}^2 + a_{40}^2)/(a_{10}^2 + a_{20}^2)$ is sufficiently small compared with unity, then the stationary values of f_1 and f_2 are approximately $f_1 \approx \pi/2$ and $f_2 \approx \pi/2\gamma$. In quasistationary interaction it follows therefore that while the strong pump a_1 goes over practically completely into its Stokes component a_2 , there will take place γ^{-1} cycles of total conversion of the energy of the weak pump a_3 into its Stokes component a_4 , and vice versa; parametric bleaching takes place at a low level of the pump a_1 . In the case of RFPI of UPS, the dependence of f_1 on ξ for certain points ξ inside the pulse can be nonmonotonic. For these points, therefore, γ^{-1} turns out to be only the minimum number of the cycles of the total conversion $a_3 \rightarrow a_4$.

The initial stage of the interaction of the considered type lends itself to calculation under conditions $|\alpha a_1 a_2| \gg |\beta a_3 a_4|$ and $\Theta(\xi) \ll 1$. The solutions that describe the behavior of the weak pump and of its Stokes component during the initial stage agree with the corresponding solution in the quasistationary case²⁰ if we substitute in it $T \rightarrow -\xi/v$.

We consider now RFPI that are degenerate in frequency. For the generation of the second Stokes component the resonance conditions take the form $\omega_p - \omega_s = \omega_{m1}$ (the subscripts p , s , and $2s$ pertain respectively to the pump radiation and to the first and second Stokes components). The behavior of the fields

C_j is described with the aid of the equations

$$\begin{aligned} \partial a_p / \partial \xi &= \pi N c^{-1} \alpha a_s \sin \Theta(\xi), \\ \partial a_s / \partial \xi &= \pi N c^{-1} (\beta a_{2s} - \alpha a_p) \sin \Theta(\xi), \\ \partial a_{2s} / \partial \xi &= \pi N c^{-1} \beta a_s \sin \Theta(\xi), \end{aligned} \quad (42)$$

where

$$\alpha = \kappa_p (\omega_p \omega_s / n_p n_s)^{1/2}, \quad \beta = \kappa_s (\omega_s \omega_{2s} / n_s n_{2s})^{1/2},$$

κ_p and κ_s are the polarizabilities of the transition between the levels 1 and m in the course of the SRS of the pump and of the first Stokes component, and correspondingly for this system $W = -a_s (\alpha a_p + \beta a_{2s})$. Equations (42) have two integrals of motion

$$\begin{aligned} a_p^2 + a_s^2 + a_{2s}^2 &= a_{p0}^2 + a_{s0}^2 + a_{2s0}^2, \\ \beta a_p + \alpha a_{2s} &= \beta a_{p0} + \alpha a_{2s0}. \end{aligned} \quad (43)$$

Since these integrals limit the possible values of the field, the matrix element of the average-motion Hamiltonian W cannot become infinite and, as follows from (15), the asymptotic value of W is zero everywhere; parametric bleaching sets in; the SIT regime is impossible. From (43) and from the requirement $W = 0$ we easily obtain the steady-state values of the fields

$$\begin{aligned} a_{2s} &= \alpha (\beta a_{p0} + \alpha a_{2s0}) (\alpha^2 - \beta^2)^{-1}, \quad a_p = -\beta (\beta a_{p0} + \alpha a_{2s0}) (\alpha^2 - \beta^2)^{-1}, \\ a_s &= [a_{p0}^2 + a_{s0}^2 + a_{2s0}^2 - (\alpha^2 + \beta^2) (\beta a_{p0} + \alpha a_{2s0})^2]^{1/2}. \end{aligned} \quad (44)$$

During the initial stage of the conversion, when $|\beta a_{2s}| \ll |\alpha a_p|$ and $\Theta(\xi) \ll 1$ the behavior of the field of the second Stokes component is described by the formula

$$a_{2s} = a_{2s0} + a_{p0} \gamma^{-1} \left\{ 1 - \left[\frac{a_{p0}^2 + a_{s0}^2}{a_{p0}^2 + a_{s0}^2 \exp(-2K_c \xi \xi_0)} \right]^{1/2} \right\}$$

[K_c is the same as in (38) and $\gamma = \alpha/\beta$], while the solutions for the pump fields a_p and for the Stokes component a_s are obtained from (38) by making the substitutions $a_1 \rightarrow a_p$ and $a_2 \rightarrow a_s$.

The system (42) describes also the behavior of the fields in the case of anti-Stokes stimulated Raman scattering. It is necessary only to make the substitutions

$$\begin{aligned} \omega_p \rightarrow \omega_s, \quad \omega_s \rightarrow \omega_p, \quad \omega_{2s} \rightarrow \omega_s, \quad a_p \rightarrow a_s, \\ a_s \rightarrow a_p, \quad a_{2s} \rightarrow a_s, \quad \kappa_s \rightarrow \kappa_p, \quad \kappa_p \rightarrow \kappa_s \end{aligned} \quad (45)$$

($a_p a_s$ and a_s are respectively the fields of the pump and of the Stokes and anti-Stokes components). Obviously here, as in the case of the generation of the second Stokes component, the asymptotic value of W is zero everywhere (the parametric bleaching regime), and the SIT regime is impossible. The steady-state values of the fields a_j are obtained from (44) by using the substitutions (45). If $|\alpha a_s| \ll |\beta a_p|$ and $\Theta(\xi) \ll 1$, then the dependence of the pump and Stokes-component fields a_p and a_s on the coordinates ζ and ξ is the same as in the case of generation of the second Stokes component during the initial conversion stage. The anti-Stokes component increases during this stage with increasing ζ like

$$a_s = a_{s0} G \left\{ \left[\frac{a_{p0}^2 + a_{s0}^2}{a_{p0}^2 \exp(2K_c \xi \xi_0) + a_{s0}^2} \right]^{1/2} - 1 \right\}, \quad G = \frac{\kappa_p}{\kappa_s} \left(\frac{\omega_s n_s}{\omega_p n_p} \right)^{1/2}.$$

6. RPI WITH PARTICIPATION OF A FIELD OF FREQUENCY RESONANT TO THE TRANSITION FREQUENCY

By way of example of such interactions we consider three-photon RPI: resonant generation of the sum fre-

quency (SF) in TPA of the pump ($\omega_1 + \omega_2 = \omega_{m1} = \omega_3$) and generation of the difference frequency (DF) in SRS ($\omega_1 - \omega_2 = \omega_{m1} = \omega_3$).²²⁻²⁴ A particular case of generation of the SF is second-harmonic (SH) generation in TPI ($2\omega_p = \omega_{m1} = \omega_{SH}$).^{18,25} It is easy to obtain for the description of these processes the equations

$$\begin{aligned} \partial a_1 / \partial \xi &= -\pi N c^{-1} \alpha a_2 \sin \Theta(\xi), \\ \partial a_2 / \partial \xi &= \mp \pi N c^{-1} \alpha a_1 \sin \Theta(\xi), \\ \partial a_3 / \partial \xi &= -\pi N c^{-1} \beta \sin \Theta(\xi). \end{aligned} \quad (46)$$

In the second equation of (46) one must use the minus sign for the SF generation problem and the plus sign for the DF generation problem; $\alpha = \kappa (\omega_1 \omega_2 / n_1 n_2)^{1/2}$, $\beta = d(\omega_3 / n_3)^{1/2}$; $W = \alpha a_1 a_2 + \beta a_3$; d is the matrix element of the dipole moment for the one-photon transition $m = 1$; κ is the polarizability of this transition and determines the pump TPA or the SRS; $a_j = C_j (n_j / \omega_j)^{1/2}$.

For SF generation we can obtain integrals of motion of the type

$$a_1^2 - a_2^2 = a_{10}^2 - a_{20}^2. \quad (47)$$

$$\frac{\alpha}{\beta} a_3 = \ln \frac{a_1 + a_2}{a_{10} + a_{20}}. \quad (48)$$

It follows from (48) that outside the limits of a pump pulse moving with velocity v [where $a_{10}(\xi) + a_{20}(\xi) = 0$], no fields at frequencies ω_1 and ω_2 are produced; relations (47) and (48) impose no limitations whatever on the amplitude of the field a_3 . The stationary distribution of a_3 is determined here by the third equation of (46), where $W = \beta a_3$. In other words, the problem of determining the stationary distribution of a_3 in this region reduces to the problem of finding SIT pulses in one-photon absorption. Among these one can separate pulses moving with velocity $V < v$ (Ref. 26) and having a finite duration (these pulses lag the pump), as well as pulses moving with velocity v . For the latter, the form of W is similar to (15). The requirement that the energy be finite limits the possible values of p , namely $p \leq 1/2$. At $p < 1/2$ the energy is zero; a finite energy is carried by SIT pulses with $C_3 \sim [\delta(\xi - \xi_0)]^{1/2}$ (these are $0 \cdot \pi$ pulses, since their area is equal to zero). This raises the question: can such resonant-field pulses be situated within the limits of the pump pulse?

A steady-state distribution of the fields within the pump pulse should satisfy the condition $\Theta(\xi) = 2\pi m(\xi)$. From this, just as for the remaining RPI, it follows that everywhere with the exception of a finite number of points there is established the regime of parametric bleaching: $W(\xi) = 0$. As for these points, in some of them there can be produced δ -like proportional subpulses of the fields a_1 and a_2 , similar to solitons in TPA; together with them there are produced at the frequency ω_3 pulses of infinite amplitude but zero energy, as follows from (48). The production of pulses with $a_3 \sim [\delta(\xi - \xi_0)]^{1/2}$, moving with velocity v within the limits of the pump pulse, should be accompanied by the appearance of pulses a_1 and $a_2 \sim \exp(\alpha_3/\beta)$ [see (48)] with infinite energy. It is obvious therefore that the production of solitons $a_3 \sim [\delta(\xi - \xi_0)]^{1/2}$ within the pump pulse is impossible.

In the case of DF generation we have in place of (47)

and (48)

$$a_1^2 + a_2^2 = a_{10}^2 + a_{20}^2, \quad (49)$$

$$a_3 |\alpha/\beta| = \arg[(a_1 + a_2)/(a_{10} + a_{20})]. \quad (50)$$

The behavior of the fields outside the pump pulse is analogous to the preceding case. Just as in the SF generation, parametric bleaching is likewise established within the limits of the pump pulse everywhere except for individual points. In the latter there can be produced $0 \cdot \pi$ pulses of SIT with $a_3 \sim [\delta(\xi - \xi_j)]^{1/2}$, moving with velocity v (in contrast to the SF generation process); no solitons of self-induced transparency can be produced at the frequencies ω_1 and ω_2 [by virtue of (49)].

These results, as well as the results of an investigation of other processes, lead to the conclusion that self-narrowing radiation pulses can be produced in RPI with participation of a field at a frequency resonant to the transition frequency.

7. ALLOWANCE FOR INHOMOGENEOUS BROADENING AND FOR FREQUENCY DETUNING

Thus, in the simplified model considered above only three types of asymptotic behavior of USP are possible in resonant interactions: 1) damping of the fields of one or several frequencies to zero; this is typical of one- and multiphoton absorption and of Raman interaction; 2) parametric bleaching wherein stationary finite amplitudes are established for all waves; this bleaching takes place in resonant parametric interactions; 3) formation of δ -function self-induced transparency pulses propagating subsequently without change of energy. We now clarify the role of the factors previously excluded from consideration.

In this section we neglect the diffraction and dispersion spreading of the wave beams and assume the v_j to be all equal. Changing from Eq. (3) to the equations for $|C_j|^2$, multiplying them by n_j , and integrating with respect to ξ , we obtain

$$\frac{\partial}{\partial \xi} \sum_j n_j \int_{\xi_1}^{\xi_2} |C_j|^2 d\xi = \frac{\pi \nu N \hbar}{c} \omega_{m1}^{(0)} \left(\int \eta g(\nu) d\nu - 1 \right), \quad (51)$$

where $\omega_{m1}^{(0)}$ is the frequency of the transition that is at exact resonance ($\omega_{m1}^{(0)} = \sum_j r_{js} \omega_j$). It follows from (4) that the total energy decreases everywhere with ξ , and that in any interval $\Delta \xi = \xi_2 - \xi_1$ a constant field energy flux density is established sooner or later. The condition for the formation of a stationary distribution of the fields independently of the form of $g(\nu)$ is

$$\int \eta g(\nu) d\nu = \int g(\nu) d\nu = 1.$$

This is possible only when $\eta(\xi) = 1$ for molecules with arbitrary detuning ν . Taking the integral of motion (9) into account, we obtain from this $\text{Im} \sigma = \text{Re} \sigma = 0$. Substituting $\eta = 1$ and $\sigma = 0$ in (3), we easily verify that the steady-state field distribution satisfies the same equation as the steady-state distribution in a system without inhomogeneous broadening [when $g(\nu) = \delta(\nu)$]. This means that the presence of inhomogeneous broadening exerts no influence on the form of the asymptotic solutions for the USP (i. e., it does not disrupt either the

parametric bleaching or the self-induced transparency); however, the inhomogeneous broadening can apparently change the threshold at which the SIT sets in. It is easy to verify that frequency detuning away from the center of the homogeneous line likewise does not lead to a qualitative change in the asymptotic behavior.

8. INFLUENCE OF THE WAVE MISMATCH AND OF THE STARK EFFECT

We shall use Eqs. (5)–(9). When the field distribution is close to the steady-state, $\eta \approx 1$, $R \approx I \approx 0$, and the phases

$$\psi_s \sim z \left\{ \delta k_s + \frac{\pi N}{c} \left[\sum_j r_{j1} \omega_j \frac{\kappa_j^{mm} - \kappa_j^{11}}{n_j} - \sum_j r_{js} \omega_j \frac{\kappa_j^{mm} - \kappa_j^{11}}{n_j} \right] \right\}$$

vary continuously relative to one another in the general case with further motion [see (6)]. When (7) and (8) are taken into account, it follows therefore that R and I can be kept equal to zero only when all the $\Pi_j A_j^{r_{js}} = 0$, i. e., at least one of the fields attenuates to zero in each of the resonances (we exclude SIT from consideration for the time being). On the other hand if

$$\delta k_s = - \frac{\pi N}{c} \left[\sum_j r_{j1} \omega_j \frac{\kappa_j^{mm} - \kappa_j^{11}}{n_j} - \sum_j r_{js} \omega_j \frac{\kappa_j^{mm} - \kappa_j^{11}}{n_j} \right]$$

(the wave mismatch is offset by the dispersion of the polarizations κ^{mm} and κ^{11} , which determine the Stark shift), and $R = I = 0$ under the condition $W = 0$, then the ψ_s are constant and R , I , and $\partial A_j / \partial \xi$ remain equal to zero, i. e., the regime of parametric bleaching is realized. In these cases the integral curves for the amplitudes of the fields participating in the RPI [see, e. g., (20)] remain the same as in the absence of wave detuning and of the Stark effect. Therefore the latter, just as the inhomogeneous broadening, do not change the conservation laws (12); the connection between the values of the field in the parametric bleaching regime and their values at the entrance into the medium also remains unchanged.

We consider now self-induced transparency. Assume that in the course of establishment of stationary field distribution there was produced in the vicinity of the point ξ_1 a sharpened subpulse of the fields at frequencies pertaining to one of the resonances. We assume also that near these points the changes of the phases are much slower than the changes of the amplitudes. It is then easy to find from (7)–(9) that in the vicinity of ξ_1

$$R \approx \cos \psi_s \sin \left(\frac{2\kappa_s}{v\hbar} \int_{\xi_1}^{\xi_2} \Pi_s d\xi \right) \approx I \text{ctg} \psi_s, \quad (52)$$

$$\eta \approx \cos \left(\frac{2\kappa_s}{v\hbar} \int_{\xi_1}^{\xi_2} \Pi_s d\xi \right), \quad \Pi_s = \prod_j A_j^{r_{js}}.$$

Substituting these expressions in (5), we obtain an equation that coincides with (10), whose steady-state solution should have an averaged Hamiltonian in the form (15). Since, as can be easily verified, the assumptions made above become more and more valid as the steady state is approached, it can be concluded that the SIT can be realized at least in one of the resonances, independently of the presence of wave mismatch and of the

Stark effect. If the SIT subpulses exist at one and the same point ξ_j at frequencies of fields pertaining to several resonances, then energy can be transferred from the fields of certain resonances into others and back, with the total energy of the pulse conserved. For example, if there are two resonances, then the coordinate dependence of the population difference takes the form

$$\eta = \cos \left\{ \frac{2\kappa_1}{v\hbar} \sin \psi_2 \left[1 + \left(\operatorname{ctg} \psi_2 + \frac{F}{\sin \psi_2} \right)^2 \right]^{1/2} \int_{\xi_1 - \zeta}^{\xi_1 + \zeta} \Pi_1 d\xi \right\} \quad (53)$$

(κ_1 pertains to the resonance with $\psi_1 = 0$, ψ_2 pertains to the second resonance, $F = \kappa_1 \Pi_1 / \kappa_2 \Pi_2$). When $\Pi_1 \sim \delta(\xi - \xi_j)$ and the argument of the cosine in (53) is equal to $2\pi n$, the population difference is $\eta = 1$. If

$$F = -\cos \psi_2 \pm [\cos^2 \psi_2 + B - 1 - B \cos^2 \psi_2]^{1/2}$$

(B is an arbitrary constant), then η remains equal to unity at all ζ , thereby ensuring energy conservation in accordance with (51). Since $\cos \psi_2$ is an oscillating function of z [see (6)], the ratio $F = \kappa_1 \Pi_1 / \kappa_2 \Pi_2$ also oscillates.

9. ROLE OF DISPERSION OF GROUP VELOCITIES

Assume that despite the difference in v_j there can exist a stationary wave distribution moving with velocity V . Then, changing over to the coordinates $\xi = z$ and $\zeta = z - Vt$ and neglecting the terms with Δ and $\partial^2/\partial t^2$, we easily obtain from (3)

$$\frac{\partial}{\partial \xi} \int_s \sum_j \frac{n_j |C_j|^2}{r_{j,\omega_j}} d\xi - \sum_j \left(1 - \frac{V}{v_j} \right) \frac{n_j |C_j|^2}{r_{j,\omega_j}} = \frac{\pi N v \hbar}{c} \left(\int \eta g(v) dv - 1 \right) \quad (54)$$

(we assume that at $t = -\infty$ the amplitudes of all the fields were equal to zero). In the steady state, the first term in (54) vanishes, and we obtain the relations that must be satisfied by the amplitude of the steady-state distributions of C_j :

$$\sum_j \left(1 - \frac{V}{v_j} \right) \frac{n_j |C_j|^2}{r_{j,\omega_j}} = \frac{\pi N v \hbar}{c} \left(1 - \int \eta g(v) dv \right). \quad (55)$$

Since the right-hand side of (55) is non-negative, V should be less than the smallest of the v_j in the case of multiphoton absorption ($s=1$, $q \geq 2$, $\omega_j > 0$).²⁾ Since the durations of the pulses of all the fields at the input are bounded ($C_j = 0$ outside them), it is obvious that the solutions with $V < \min\{v_j\}$ cannot occur; thus, in the case of multiphoton absorption the dispersion of the group velocities disrupts the self-induced transparency. A similar analysis for the Raman processes shows the possibility of formation of SIT pulses of finite amplitude if $\max\{v_{sc}\} < \min\{v_p\}$ ($\{v_p\}$ are the velocities of the pump components, $\omega_j > 0$; $\{v_{sc}\}$ are the velocities of the scattering components, $\omega_j < 0$), with $\max\{v_{sc}\} < V < \min\{v_p\}$. For the case of stimulated Raman scattering (SRS) the corresponding solutions were obtained in Refs. 5 and 27.

In RPI, when there are several resonances, the possibility of formation of SIT pulses are greater. Thus, in a system with two TPA resonances there can be produced SIT pulses with finite amplitude, moving with velocity V that satisfies the condition $v_1, v_2 < V < v_3, v_4$

[analogous relations for RPI can be easily obtained from (55)], something possible at a definite law of dispersion of the refractive index (we recall that SIT is impossible in each of the resonances taken separately). If at the same time $\delta k = 0$ and there is no dispersion of the Stark shift, and $\gamma = \alpha/\beta = 1$,

$$\left(\alpha = \kappa_p \left(\frac{\omega_1 \omega_2 v_1 v_2}{n_1 n_2 (v_1 - V)(v_2 - V)} \right)^{1/2}, \quad \beta = \kappa_c \left(\frac{\omega_3 \omega_4 v_3 v_4}{n_3 n_4 (v_3 - V)(v_4 - V)} \right)^{1/2} \right)$$

then the steady-state field distribution takes the form

$$a_1^2 = a_2^2 = D^2 a_3^2 = D^2 a_4^2 = \frac{2\pi V \hbar N}{c[1 + 4\pi^2 N^2 \alpha^2 c^{-2} (\xi - \xi_0)^2]} \frac{D^2}{1 + D^2}, \quad (56)$$

where $a_j = C_j [n_j |v_j - V| / \omega_j v_j]^{1/2}$, ξ_0 is determined by the instant of time t_0 at which the maximum of the pulse should pass through the point $z = 0$, and

$$D^2 = \lim_{t \rightarrow -\infty} (a_i^2 / a_j^2).$$

Steady-state solutions with nonzero values of all the fields that participate in the RPI can be called parametric solitons.

We shall show that the dispersion of the group velocities destroys the parametric bleaching that should be realized when $W(\xi) \equiv 0$ and all the $C_j(\xi) \neq 0$. If $W(\xi) = 0$, then we should have [see (4)] $\eta(\xi) \equiv 1$. We then find from (55) that all the amplitudes $C_j = 0$ with the possible exception of only one (for which $v_j = V$), and this contradicts the definition of parametric bleaching.

We note that all the qualitative conclusions concerning the influence of the dispersion of v_j were made without any assumptions concerning the magnitude of the wave mismatch and of the Stark shift of the levels, are concerning the presence or absence of inhomogeneous broadening.

10. ALLOWANCE FOR DISPERSION AND DIFFRACTION SPREADING OF WAVE PACKETS

We consider first the influence of the dispersion spreading of USP, confining ourselves to an example of multiphoton absorption, and neglecting the inhomogeneous broadening and the Stark effect. For this case, in the coordinate system with $\zeta = z$ and $\xi = z - Vt$, we have in place of (3) the equation

$$\frac{\partial C}{\partial \xi} + \left(1 - \frac{V}{v} \right) \frac{\partial C}{\partial \xi} + \frac{i}{2k} \left(1 - \frac{V^2}{v^2} - V^2 k \frac{\partial^2 k}{\partial \omega^2} \right) \frac{\partial^2 C}{\partial \xi^2} = -i \frac{2\pi N \omega}{nc} \kappa C^* \sigma; \quad (57)$$

σ and η are described by Eqs. (4) in which $\nu = \Omega = 0$. From (57) we easily find that so long as the pulse duration is not too small, and the area is large enough, a shortening of the pulse takes place, and this would lead to a δ -shaped pulse W if the term with $\partial^2 C / \partial \xi^2$ would not grow. On the other hand, a solution with $W \sim \delta(\xi - \xi_j)$ cannot simultaneously satisfy the equations for σ and η and Eq. (57). This means that the dispersion spreading of the pulses is a substantial factor that determines the shape of the solitons and imposes a lower bound on their duration.

We consider now the influence of the diffraction of light beams. Since the qualitative arguments that explain the diffraction-induced instability of one-dimen-

sional waves²⁸ remain valid also for multiphoton resonances, it is obvious that this instability should take place also in this case. However, a more important factor in the character of the asymptotic behavior at large distances is that the beam has a limited cross section.

From (3), neglecting dispersion factors, inhomogeneous broadening, and the Stark effect, it is easy to obtain an equation for $|C_j|^2$, from which, after integrating first over the cross section and then with respect to ξ , we get

$$\frac{\partial}{\partial \xi} \sum_j n_j \int \int_{-\infty}^{\infty} |C_j|^2 dx dy d\xi = \frac{\pi N v \hbar \omega_m}{c} \int \int_{-\infty}^{\infty} [\cos \Theta(\xi) - 1] dx dy$$

(we have used the radiation conditions at infinity). The steady-state solution must satisfy the relation

$$-\frac{\partial}{\partial \xi} \sum_j n_j \int \int_{-\infty}^{\infty} |C_j|^2 dx dy d\xi = 0,$$

and this is possible only if the area Θ is everywhere equal to an integer number of 2π . Since all the $C_j \rightarrow 0$ as $x^2 + y^2 \rightarrow \infty$, it follows that $Q(x, y, \xi)$ should be everywhere equal to zero. From the equations for C_j , written down with allowance for diffraction, it is easy to find that the equality $W(x, y, \xi) = 0$ will not be preserved when ξ is varied if it is recognized that the terms proportional to $(\partial^2/\partial x^2 + \partial^2/\partial y^2)C_j$ are different because the k_j are different. Thus, the limited character of the dimensions of the light beams participating in the resonant interaction disrupts in final analysis both the SIT and the parametric bleaching (the latter is true also in the quasistationary case). A similar result is produced also by nonresonant energy loss of the interacting waves, due e.g., to scattering.

CONCLUSION

We present the basic results of the study and make a few estimates (using the data of Ref. 29 for Na and of Refs. 6 and 30 for Li). The first group of conclusions (items 1–7) pertain to systems to which the simplified model used in Secs. 2–6 is applicable.

1. The formation of intense and very short subpulses of radiation (corresponding to δ -like subpulses of SIT in our model) is possible at frequencies that enter in the resonant conditions of the type of multiphoton absorption [all $\omega_j > 0$ in (2)].

2. No such subpulses can be produced as a rule at the frequencies that enter in the resonance conditions of the Raman type [some of the frequencies negative in (2)]. In this case the inhomogeneities of the radiation, which occur during the initial stage, become smoothed out ultimately. Exceptions are RPI, in which one of the resonances corresponds to multiphoton absorption and the other to Raman scattering, with the frequency of any one of the fields entering in both resonance conditions.

3. Parametric bleaching, which consists of establishment of finite amplitudes, independent of the distance covered by the pulse, at the frequencies of all the fields that take part in the process, is also a characteristic

of resonant parametric interactions of USP. The connection between the amplitudes of the field at the entrance and their steady-state values is the same as in the corresponding quasistationary RPI. The parametric bleaching regime is disturbed at those points inside the pulse, where subpulses of self-induced transparency are produced.

4. In the case of formation of δ -like subpulses, their energy turns out to have the standard value, depending only on the order of the resonance, on the values of the frequencies, of the refractive indices, and of the polarizability (connected with the corresponding resonance) of the transition between the levels. This makes it possible to determine in simple fashion, by measuring the energy, the polarizabilities of both transitions with two-photon absorption and of SRS transitions, if the frequency-degenerate RPI considered in Sec. 4 are used.

5. The energies of the SIT subpulses at frequencies that enter only in the resonances with $q > 2$ are equal to zero. Therefore among the set of RI in centrosymmetric media (gases, liquids), from the point of view of generation of frequency tunable subpulses of SIT, greatest interest attaches to RFPI, in which at least one of the resonances corresponds to two-photon absorption.

6. If both resonances correspond to the TPA, the formation of SIT subpulses calls for satisfaction of the condition $\omega_c(\omega_3\omega_4/n_3n_4)^{1/2} \geq \omega_p(\omega_1\omega_2/n_1n_2)^{1/2}$ ($1 \geq \gamma$). In such systems it is possible to achieve considerable narrowing of the duration and increase of the power of the fields even in the case when the initial energies of these fields are insufficient to obtain the corresponding effect by using two-photon resonance alone. For this purpose it is sufficient to have the intensity of the fields in the pump channel exceed the threshold of formation of the subpulses in their two-photon absorption (without RFPI).

By way of example we consider sodium vapor with concentration $N = 2 \times 10^{16} \text{ cm}^{-3}$. The working transition is $3s-4d$. The pump frequencies are $\omega_{1,2} = 17275.5 \text{ cm}^{-1}$, the frequencies in the conversion channel are $\omega_3 = 4226 \text{ cm}^{-1}$ and $\omega_4 = 30272 \text{ cm}^{-1}$, the polarizabilities are $\kappa_p = 2 \times 10^{-22} \text{ cm}^3$ and $\kappa_c = 3 \times 10^{-22} \text{ cm}^3$. In this case $\gamma = 1$, the pump-power threshold is 750 MW/cm^2 at an input pulse duration $\tau_{\text{pulse}} = 10^{-11} \text{ sec}$. Applying to the input, simultaneously with the threshold pump pulse, pulses with intensities $I_{30} = 1.8 \text{ MW/cm}^2$ and $I_{40} = 12.5 \text{ MW/cm}^2$ ($\tau_{\text{pulse}} = 10^{-11} \text{ sec}$), we can obtain at the output pulses that are 100–1000 times more intense and shorter at the frequencies ω_3 and ω_4 (both photoionizations⁷ and the dispersion of v , limit the duration of the SIT pulses in TPA to the level 10^{-13} – 10^{-14} sec). We note that by using simply the TPA of the frequencies ω_3 and ω_4 , no such contraction of the pulses is possible so long as the initial intensities I_{30} and I_{40} are lower than 180 MW/cm^2 and 1.25 GW/cm^2 , respectively. Using (25), we can easily determine the characteristic distance at which an SIT regime is established in RFPI, namely, $L \sim \Gamma^{-1} \approx 2 \text{ cm}$. If $\gamma < 1$ ($\omega_{1,2} = 17275.5 \text{ cm}^{-1}$, $\omega_3 = 4276 \text{ cm}^{-1}$, $\omega_4 = 30276 \text{ cm}^{-1}$, $\kappa_p = 2 \times 10^{-22} \text{ cm}^3$, and $\kappa_c = 5 \times 10^{-21} \text{ cm}^3$), the steady-state energies of the sub-

pulses of the field in the conversion channel do not depend on the initial values ($U_3 \sim 1.2 \times 10^{-4} \text{ J/cm}^2$; $E_4 \sim 0.75 \times 10^{-3} \text{ J/cm}^2$).

7. The resonant four-photon parametric interaction discussed in item 6 (all $\omega_j > 0$), can cause both a decrease of the frequency and an increase (to ω_{m1}). The frequency can be lowered also by using RFPI in which one of the ω_j is negative—generation of a difference frequency in SRS pumping. To increase the frequency to more than ω_{m1} with formation of SIT subpulses, the only suitable RI is RFPI, which takes place under conditions of a resonance of the type (31).

A similar interaction can be effected in lithium vapor with working levels $2s-5s$, $\kappa_{\text{TPA}} = 3.8 \times 10^{-24} \text{ cm}^3$, $\kappa_{\text{SRS}} = 7 \times 10^{-25} \text{ cm}^3$, $N = 2 \times 10^{16} \text{ cm}^{-3}$, $\omega_1 = 18\,868 \text{ cm}^{-1}$, $\omega_2 = 19\,668 \text{ cm}^{-1}$, $\omega_3 = 58\,204 \text{ cm}^{-1}$ ($\omega_{1,2}$ are the pump frequencies and ω_3 is the frequency of the generated field). The energies of the SIT subpulses turn out here to be $U_1 \approx 0.4 \text{ J/cm}^2$, $U_2 = 0.38 \text{ J/cm}^2$ and $U_3 = 0.14 \text{ J/cm}^2$. In the proportional interaction regime, the characteristic distance over which SIT subpulses are produced [a distance equal to K^{-1} , see Eq. (36)] is $\sim 170 \text{ cm}$; the threshold pump power needed to realize the SIT regime is $\sim 40 \text{ GW/cm}^2$ at a pulse duration 10^{-11} sec .

8. The inhomogeneous level broadening and the deviation from resonance do not influence the character of the stationary field distributions, which can set in at large distances (although they undoubtedly affect the transient process).

9. When account is taken of the remaining factors that make the theoretical model closer to reality, a formulation similar to the Le Chatelier principle remains valid: the evolution of USP in resonant interaction with a medium leads to changes such that the action of the medium on the pulses is stopped. Their influence, however, leads to a qualitative change of the possible forms of the asymptotic solutions.

The wave mismatch disrupts the parametric bleaching. Self-induced transparency remains possible also in the absence of wave synchronism. The wave mismatch can lead to a new type of parametric soliton, in which periodic energy exchange can take place between the subpulses of fields belonging to one of the resonances and subpulses of fields at frequencies entering in the other resonance condition.

10. The presence of a dynamic Stark shift of the level does not change the character of the possible asymptotic solutions, either by itself or in conjunction with inhomogeneous broadening. The solutions are changed by the frequency dispersion of the polarizabilities that determine the Stark effect; the influence of the polarizability is analogous to the influence of the wave mismatch.³⁾

11. The dispersion of the group velocities of the interacting waves makes parametric bleaching impossible. Its action should lead also to the appearance of SIT solitons with finite duration and amplitude. The soliton velocity should lie between the highest and lowest of the group velocities of these waves. Impossible-

ity of existence of δ -like solitons should result also from the dispersion spreading of the wave packets.

The requirement that the light pulses entering the medium be of limited duration excludes the possibility of soliton formation in processes of the multiphoton absorption type. The possibility of their onset in Raman processes and in RPI is not affected by this factor. If the SIT regime is not realized, very short pulses can nevertheless appear also at various group velocities; in this case pulses with different frequencies are separated in space (in the longitudinal direction).

12. The limited transverse dimensions of the light beams, as well as nonresonant losses, can cause only the intensities of all the fields to be attenuated to zero.

13. Estimates performed using typical values ($\kappa_j^{11} \sim 10^{-24} \text{ cm}^3$, $\kappa_j^{nm} \sim 10^{-23} \text{ cm}^3$, $\partial \kappa / \partial \omega \sim 10^{-40} \text{ cm}^3 \text{ sec}$) show that the dispersion of the Stark shift of the level is equivalent to the influence of wave mismatch with a synchronism length exceeding by many orders the distances over which the SIT pulses are produced. Ordinary wave mismatch can be made small enough by a suitably chosen buffer gas.³¹ If the phase velocities are made equal in this manner, the quantity $(v_j - v_l)/v_j < 10^{-3}$ ($j \neq l$) and the duration of the produced solitons, estimated with the aid of (56), does not exceed $10^{-13} - 10^{-14} \text{ sec}$. Therefore the dispersion factors do not change the estimates, made in the first part of the paper, of the limiting parameters of the self-narrowing pulses.

It is obvious that in cases when a distinct hierarchy of the parameter values characterizing the effectiveness of the action of each factors can be established, the real distribution of the fields in time and in space takes on alternately a form close to one and to the other of the asymptotic solutions obtained above. This makes it possible to track in greater detail the evolution of the pulses from the initial stage to the end. Thus, in the case of frequency-degenerate two-photon absorption of an intense beam of limited cross section but of sufficient width, with a small spatial modulation (due, for example, to the structure of the transverse mode of the master laser), the following should take place in a medium with a dispersion of the refractive index: an SIT pulse is first produced practically over the entire beam cross section; the pulse narrows down to a definite duration, after which, owing to an instability of the type considered in Ref. 28, the inhomogeneity in the transverse direction is strengthened. After that, diffraction causes a gradual destruction of the SIT, and the field ultimately vanishes everywhere.

¹⁾If the temperature of the medium $\neq 0$, then the equilibrium population difference $\eta_0 \neq 1$. Generalization to the case of nonzero temperature is easy by multiplying by η_0 the right-hand sides of the expressions for R and η and by replacing N by $N\eta_0$ everywhere except in Eqs. (51), (54), and (55), where $\eta - 1$ must be replaced by $\eta - \eta_0$.

²⁾We are interested here in a nontrivial solution in which all $C_j \neq 0$. The case when V coincides with one of the v_j corresponds to spatial separation of waves with different frequencies, inasmuch as in this case the remaining C_j at a given value of ξ are equal to zero.

- ³The question of which of the possible stationary solutions (zero or SIT) is established, and under what conditions, remains open in the general case. Numerical calculations performed for a number of specific systems [K. N. Drabovich and L. M. Kocharin, *Sov. J. Quantum Electron.* **10**, 1386 (1980)] have shown that in these systems the frequency detuning and the Stark effect lead to damping of the USP, and not to establishment of SIT.
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