Interaction of ultrashort light pulses with an excited two-photon resonant medium

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The resonant interaction of ultrashort light pulses with a two-photon-absorbing (amplifying) medium preexcited by an additional field has been studied. The area (energy) theorem for an arbitrary type of excitation has been obtained in a general form. A detailed study of the problem has been made by using the example of an exciting field with a frequency resonant with the frequency of the working transition. It is shown that the presence of the exciting field gives rise to a new type of pulses of self-induced transparency (SIT), among which the most interesting properties are exhibited by $2|\mathscr{S}|$ pulses. In contrast to a 2π pulse, a $2|\mathscr{S}|$ pulse is limited in the cross section (for finite transverse dimensions of the exciting field) and the SIT effect of a $2|\mathscr{S}|$ pulse can be observed over its entire cross section. The transverse distribution of intensity in a $2|\mathscr{S}|$ pulse turns out to be similar to that of the exciting radiation intensity. The stability of SIT pulses to the relaxation of the medium and to transverse inhomogeneity of the light fields in the presence of the exciting field is investigated.

1. INTRODUCTION

The last few years have seen the publication of a large number of both theoretical and experimental studies dealing with aspects of resonant interactions (RI) of ultrashort pulses (USP) of light with a substance or of coherent resonant interactions (CRI) (see for example Refs. 1–21). The simplest of these, i.e., single-photon CRI, such as photon echo,[1,2] self-induced transparency (SIT),[3] optical nutation,[4] attenuation of free polarization,[5] and adiabatic transmission,[6] have been studied fairly closely. Interest in coherent processes is due primarily to the large volume of spectroscopic information that can be obtained on a substance when they are used (see Ref. 7 and the literature cited there). Even greater possibilities are offered by two-photon coherent processes. First, they can be used to obtain new information on a substance (for example, coherent spectroscopy free from Doppler broadening,[8] or three-level stimulated echo[9]). Second, two-photon RI of USP can be used in problems of conversion of the frequency of short pulses as well as their shortening.[10–15] Nevertheless, two-photon coherent interactions remain little-studied experimentally (let us note at this point Refs. 16–19, which investigated two-photon SIT and Raman beats[20,21]). This is apparently due to the fact that for two-photon coherent processes to be observed the laser pulse must meet more exacting requirements. In particular, the pulse power must be much higher than in single-photon processes, and the phase modulation in it must be low. Frequently, sources of tunable USP available in practice satisfy neither the first nor the second requirement. Furthermore, if one deals with CRI (such as two-photon SIT) at large distances, great importance is assumed by the inhomogeneity in the cross section of the USP[22] and by the finiteness of the relaxation times of the medium. Both of these factors (as well as the phase modulation[23]) lead to loss of the coherence of propagation of the USP and to pulse attentuation.[15] On the other hand, if an additional field parametrically coupled with the USP fields is applied to the medium, then in addition to the pulse energy losses (due to diffraction, relaxation, etc.), amplification of USP may occur as a result of energy transfer from the additional field. Self-sustaining pulses can thus be expected to appear in such systems.

This paper discusses the coherent interaction of USP with a two-photo-absorbing (amplifying) medium preexcited by a field having a frequency resonant to the transition frequency. The object of the work was to study the possibilities of coherent lossless propagation of USP, inhomogeneous in the cross section, over large distances in media with finite relaxation times, to study the self-contraction of USP in such media, and to observe two-photon SIT and other CRI related to it in the system under consideration.

2. INITIAL EQUATIONS

Let us consider the interaction of a short pulse of two fields

$$E_{1,2}=C_{1,2}\exp\left[i(\omega_{1,2}t-k_{1,2}z)\right]+\text{c.c.},\quad C_{1,2}=A_{1,2}\exp\left(-i\varphi_{1,2}\right)$$

with a noncentrally symmetric medium in which prior to the arrival of USP a nonzero polarization is induced by means of an exciting resonance field

$$E_0=\left[C_{01}\exp\left(-ik_0z\right)+C_{02}\exp\left(ik_0z\right)\right]\exp\left(i\omega_0t\right)+\text{c.c.},$$
$$C_{01,02}=A_{01,02}\exp\left(-i\varphi_{01,02}\right),$$

where C_{01}, C_{02} are the amplitudes of the forward and backward resonance waves. Initially, the medium is excited by a resonant field of frequency ω_0, starting at time $t=t_0$. Then at time $t=0$, the medium is subjected to USP of fields of duration τ_p (the resonance field can also act when $t>0$). The resonance conditions are

$$\omega_1+\omega_2=\omega_{m1}+\nu_1,\qquad \omega_0=\omega_{m1}+\nu_0,$$

where ω_{m1} is the frequency of the working transition between the levels 1 and m, and ν_0 and ν_1 are small detunings from resonance. The changes in $C_{1}^{-}{}_{2}$ of the pulse (which will hereinafter be referred to as converted fields) and resonant fields C_{01}, C_{02} are described by the equations

$$\left(\frac{\partial}{\partial z}+\frac{1}{v}\frac{\partial}{\partial t}\right)C_{1,2}+\frac{i}{2k_{1,2}}\Delta_\perp C_{1,2}=-\frac{2\pi i\omega_{1,2}N}{n_{1,2}c}\left[\frac{1}{2}C_{1,2}\langle\eta\rangle\right.$$
$$\left.\times(\varkappa_{1,2}^{mm}-\varkappa_{1,2}^{11})+\varkappa_{12}\exp\{i(k_1+k_2)z\}C_{2,1}^{*}\langle\sigma\rangle\right],\qquad(1)$$

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$$\left(\frac{\partial}{\partial z}\pm\frac{1}{v_0}\frac{\partial}{\partial t}\right)C_{01,02}\pm\frac{i}{2k_0}\Delta_\perp C_{01,02}=-\frac{2\pi i\omega_0 N}{n_0 c}\left[\frac{1}{2}C_{01,02}\langle\eta\rangle\right.$$
$$\left.\times(\varkappa_0^{mm}-\varkappa_0^{11})+d\exp(\pm ik_0 z)\langle\sigma\rangle\right], \tag{2}$$

obtained by substituting in the Maxwell equation in the parabolic approximation[25] the atomic polarization found by Butylkin *et al.*[24] for resonant processes. In Eqs. (1) and (2), n_j is the refractive index at frequency ω_j; κ_j^{mn} and κ_j^{11} are the polarizabilities at the frequency ω_j of the atom (molecule) in the energy states m and 1, respectively, κ_{12} is the polarizability of the two-photon transition $1\leftrightarrow m$, d is the dipole moment of the $1\leftrightarrow m$ transition, and N is the density of the atoms (molecules) of the medium; v_0, v are group velocities of the resonant wave and converted waves

$$\Delta_\perp=\partial^2/\partial x^2+\partial^2/\partial y^2,\qquad \langle(\ )\rangle=\int(\ )g(\nu)d\nu,$$

$g(\nu)$ being the distribution function of the atoms over ν, normalized to unity and characterizing the inhomogeneous line broadening. We will postulate that $g(\nu)$ is an even function of ν and assume that $\omega_1+\omega_2$ coincides with the central transition frequency ω_{ml}^0. The population difference η and the nondiagonal element of the density matrix σ satisfy the equations of a generalized two-level system[26]

$$\partial\sigma/\partial t+[T^{-1}-i(\Omega+\nu)]\sigma=i\hbar^{-1}\mathscr{V}\eta, \tag{3}$$
$$\partial\eta/\partial t+(\eta-\eta_e)\tau^{-1}=-4\hbar^{-1}\,\mathrm{Im}\,(\sigma\mathscr{V}^*), \tag{4}$$

where

$$\mathscr{V}=-\varkappa_{12}C_1C_2[-i(k_1+k_2)z]$$
$$-dC_{01}\exp(-ik_0z)-dC_{02}\exp(ik_0z);$$
$$\Omega=\hbar^{-1}\sum_j(\varkappa_j^{mm}-\varkappa_j^{11})|E_j|^2\qquad(j=0,1,2);$$

τ and T being the time of longitudinal and transverse relaxation, and η_e the equilibrium difference of the populations. If there exists a pumping source by means of which an inverted population can be produced between the levels 1 and m, then τ and η_e are determined in terms of the probabilities W_{ij} of transitions from the i state to the j state. For a three-level system, the expressions for τ and η_e are given in Ref. 26.

3. THE PLANE-WAVE APPROXIMATION

We consider first the case in which the fields C_j change fairly slowly along the transverse coordinates x and y, and the terms proportional to $\Delta_\perp C_j$ may be neglected in Eqs. (1) and (2). Changing to the coordinate system $t\to t-z/v$, $z\to z$, we obtain from Eq. (1) the equations for amplitudes $A_{1,2}$ and phase $\varphi_{1,2}$:

$$\partial A_{1,2}/\partial z=-\pi N\varkappa_{12}\omega_{1,2}(n_{1,2}c)^{-1}A_{2,1}\langle R\rangle, \tag{5}$$
$$\partial(\varphi_1+\varphi_2)/\partial z=\pi Nc^{-1}\{\varkappa_{12}(A_2\omega_1/A_1n_1+A_1\omega_2/A_2n_2)\langle I\rangle$$
$$+\langle\eta\rangle[(\varkappa_1^{mm}-\varkappa_1^{11})\omega_1n_1^{-1}+(\varkappa_2^{mm}-\varkappa_2^{11})\omega_2n_2^{-1}]\}, \tag{6}$$

where $R=iI=2i\sigma\exp(i\Phi)$, $\Phi=\varphi_1+\varphi_2$. We will assume[1)] that one of the fields, the triggering field $C_1(z=0)=C_{10}\neq0$, is applied to the medium, whereas $C_2(z=0)=0$. It can be shown that for the excitation cases most interesting from an applied point of view, the presence of a resonant field in the interval $0<t<\tau$ does not affect the evolution of USP fields. Hence it may be assumed that $C_{01,02}$ $(0<t<\tau_{\rm pul})=0$. The complete system of equations for fields $A_{1,2}$ in this case will consist of Eqs. (5), (6) and the equations for the medium (3), (4) in the approximation $\tau_{\rm pul}\ll\tau$, T, i.e.,

$$\dot R+I(\dot\Phi+\nu+\Omega_p)=2\varkappa_{12}\hbar^{-1}A_1A_2\eta, \tag{7}$$
$$\dot I=(\dot\Phi+\nu+\Omega_p)R, \tag{8}$$
$$\dot\eta=-2\varkappa_{12}\hbar^{-1}A_1A_2R, \tag{9}$$

where $\Omega_p=\Omega(C_{01,02}=0)$. We find the solution for $A_{1,2}$ in general form, assuming that the initial values $R_0(z)$, $I_0(z)$, $\eta_0(z)$ at $t=0$, determined by the exciting field, are specified functions of z. From Eqs. (5), (6) and (12), one can readily ascertain that in the initial stage, when A_2 is small and $R\approx R_0(z)$ as a result of phase capture, A_1/A_2 is independent of t, and as z increases, $a_2\to a_1$, where $a_j\equiv A_j(n_j/\omega_j)^{1/2}$. If $a_2\approx a_1$ has been established at some point, then, as is evident from the Manley-Rowe relation ($a_1^2-a_2^2=a_{10}^2$), as $a_{1,2}$ increases further, $a_2\approx a_1$ all the more. In the latter case, one speaks of a proportional interaction regime. Let the proportional regime begin before the initial stage of the interaction ends. As shown in Ref. 14 (for an analogous problem), this is always the case when the triggering field a_{10} is sufficiently low. In this case a_1/a_2 may be considered always independent of t. Using this fact, after some simple transformations we obtain from Eqs. (5), (6), (7)–(9) the following expression for $A_{1,2}$:

$$\frac{\partial A_{1,2}}{\partial z}=-\frac{\pi N\varkappa_{12}\omega_{1,2}}{n_{1,2}c}A_{2,1}aq\sin\left[\mathscr{P}(z)+\frac{2\varkappa_{12}a}{\hbar}\int_0^t A_1A_2\,dt\right], \tag{10}$$

where

$$\tan\mathscr{P}(z)=a\langle R_0\rangle/(\langle\eta_0\rangle-\langle I_0\rangle F),\ a=(1+F^2)^{1/2},\ q=(\rho/a^2-b)^{1/2},$$
$$b=F(\langle I_0\rangle+F\langle\eta_0\rangle)/a^2,\quad \rho=\langle R_0\rangle^2+\langle I_0\rangle^2+\langle\eta_0\rangle^2,$$

with $F(z)$ changing from zero at $a_2=0$ to

$$r/2=2\pi N(\hbar\gamma c)^{-1}[(\varkappa_1^{mm}-\varkappa_1^{11})\omega_1/n_1+(\varkappa_2^{mm}-\varkappa_2^{11})\omega_2/n_2]$$

in the proportional regime ($a_1=a_2$);

$$\gamma=2\pi Nc^{-1}(\omega_1\omega_2/n_1n_2)^{1/2}.$$

We introduce the notation

$$\psi=\mathrm{ctn}\frac{1}{2}[\mathscr{P}(z)+\vartheta_t],\qquad \vartheta_t=\frac{2\varkappa_{12}a}{\hbar}\int_0^t A_1A_2\,dt,\qquad s(z)=\gamma aq.$$

Taking the proportionality condition into account, we then obtain from Eq. (10) a Riccati type equation:

$$2\partial\psi/\partial z+(\psi^2+1)\partial\mathscr{P}/\partial z+s(z)[\psi^2-1-(\psi^2+1)\cos\mathscr{P}(z)]=0, \tag{11}$$

which has a partial solution $\psi=\cot(\varphi/2)$. The general solution (11) is

$$\psi=\mathrm{ctn}(\mathscr{P}/2)+P(z)\left\{P(z_0)[\psi(z_0)-\mathrm{ctn}(\mathscr{P}(z_0)/2)]^{-1}\right.$$
$$\left.+{}^1/_2\int_{z_0}^{z}[d\mathscr{P}/dz+s(1-\cos\mathscr{P})]P(z)\,dz\right\}^{-1}, \tag{12}$$

where

$$P(z)=\exp\left\{-\int \operatorname{ctn}(\mathscr{P}/2)\,[d\mathscr{P}/dz+s(1-\cos\mathscr{P})\,]dz\right\}.$$

The solution (12) is essentially a generalized area theorem describing the interaction of USP with a two-photon-absorbing (amplifying) medium excited in an arbitrary manner (not necessarily by a resonant field).

4. STATIONARY EXCITATION

In this section, we will consider the case in which the amplitudes of the resonant field C_{01}, C_{02} are independent of t (stationary approximation). Equations (3), (4), in which $C_{1,2}=0$, give simple solutions (see for example Ref. 24). As shown by the estimates in Ref. 26, for single-photon-allowed transitions usually $|\Omega T|\ll 1$. In this case, from Eq. (2), averaging both sides of the equation over a segment equal to the wavelength, we obtain the equations for the amplitudes and phases of the resonant field:[2)]

$$\partial A_{01,02}/\partial z=\mp\pi N\omega_0 d(2n_0c)^{-1}\langle G_{1,2}\rangle, \tag{13}$$

$$\frac{\partial\varphi_{01,02}}{\partial z}=\frac{\pi N\omega_0}{n_0c}\left[(\varkappa_0^{mm}-\varkappa_0^{11})\langle G_0\rangle\mp\frac{\nu Td}{2}\frac{\langle G_{1,2}\rangle}{A_{01,02}}\right], \tag{14}$$

where

$$G_{1,2}=\hbar\eta_p(2\tau d)^{-1}$$
$$[8\tau Td^2\hbar^{-2}A_{01,02}+A_{01,02}^{-1}((C^2-4B^2)^{1/2}-C)\,](C^2-4B^2)^{-1/2},$$
$$G_0=\eta_e(1+\nu^2T^2)(C^2-4B^2)^{-1/2},$$
$$B=4\tau T\hbar^{-2}A_{01}A_{02},\quad C=1+\nu^2T^2+4\tau T\hbar^{-2}d^2(A_{01}^2+A_{02}^2).$$

The initial values R_0, I_0, η_0, averaged over the wavelength, are

$$R_0=G_1(\cos\Delta\psi_0+\nu T\sin\Delta\psi_0),$$
$$I_0=G_1(\sin\Delta\psi_0-\nu T\cos\Delta\psi_0),\qquad \eta_0=G_0, \tag{15}$$

where $\Delta\psi_0=\Delta\psi\quad(t=0)\quad(\Delta\psi=\Phi-\varphi_0+\delta kz,\delta k=k_1+k_2-k_0)$, in accordance with Eqs. (6) and (14), satisfies the equation

$$\partial\Delta\psi_0/\partial z=\delta k+\pi Nc^{-1}\Big\{\varkappa_{12}(A_2\omega_1/A_1n_1+A_1\omega_2/A_2n_2)\,|_{t=0}$$
$$\times\langle G_1\rangle\sin\Delta\psi_0+\Big[(\varkappa_1^{mm}-\varkappa_1^{11})\frac{\omega_1}{n_1}+(\varkappa_2^{mm}-\varkappa_2^{11})\frac{\omega_2}{n_2}$$
$$-(\varkappa_0^{mm}-\varkappa_0^{11})\frac{\omega}{n_0}\Big]\langle G_0\rangle\Big\}. \tag{16}$$

At the initial stage of the transformation, we obtain from Eqs. (5), (15)

$$\partial A_{1,2}/\partial z=-\pi N\varkappa_{12}\omega_{1,2}(n_{1,2}c)^{-1}A_{2,1}\langle G_1\rangle\cos\Delta\psi_0. \tag{17}$$

We will consider the case in which the resonant excitation is distributed uniformly along the medium: G_0, $G_{1,2}=\text{const}$. Such a situation exists, for example, in the following two types of media:

1. Media with an inverted population, placed in a cavity and generating a resonant field. As shown in Ref. 27, if the reflectances of the mirrors are close to 100%, the distribution of amplitudes A_{01} and A_{02} along the sample is nearly constant.

2. Media placed in an electrostatic field. Let $A_{20}=0$, and the electrostatic field $\mathscr{E}_0$ increases along z according to the law

$$d=d_{1m}+\varkappa_{20}\mathscr{E}_0=d(z=0)\,(1-KZ)^{-1/2},$$
$$K=\pi N\omega_0\eta_p\hbar[n_0c\tau A_{01}^2(z=0)\,]^{-1}[c(z=0)-1\,]/C(z=0), \tag{18}$$

where d_{1m} and $\kappa_{20}\mathscr{E}_0$ are the intrinsic and induced dipole moments of the $1\leftrightarrow m$ transition.

Considering that $G_{1,2}$, G_0 are constant and integrating Eqs. (16), (17), we obtain the following solutions in the initial stage of the interaction:

1) for $|\delta|<\gamma\langle G_1\rangle$

$$a_2=a_{10}(1-\delta^2/\gamma^2\langle G_1\rangle^2)^{-1/2}\,\mathrm{sh}\,[{}^1/_2(\gamma^2\langle G_1\rangle^2-\delta^2)^{1/2}z], \tag{19}$$

(2) for $|\delta|>\gamma\langle G_1\rangle$

$$a_2=a_{10}(\delta^2/\gamma^2\langle G_1\rangle^2-1)^{-1/2}\sin\,[{}^1/_2(\delta^2-\gamma^2\langle G_1\rangle^2)^{1/2}z],$$
$$a_1=(a_{10}^2+a_2^2)^{1/2}, \tag{20}$$

$$\delta=\delta k+\pi Nc^{-1}[\,(\varkappa_1^{mm}-\varkappa_1^{11})\,\omega_1/n_1$$
$$+(\varkappa_2^{mm}-\varkappa_2^{11})\,\omega_2/n_2-(\varkappa_0^{mm}-\varkappa_0^{11})\,\omega_0/n_0\,].$$

It is evident from Eqs. (19), (20) that if the wave detuning δ is small, the increase of the fields is monotonic, whereas for $|\delta|>\gamma\langle G_1\rangle$ the fields $a_{1,2}$ oscillate sinusoidally. For $|\delta|<\gamma\langle G_1\rangle$, because of unlimited increase, the condition of smallness of a_2 will break down sooner or later, and Eq. (19) will cease to apply. As shown in Ref. 14 (for an analogous problem), for a sufficiently small a_{10}, even before the condition of smallness of a_2 breaks down, a proportional regime of interaction is established ($a_1\approx a_2$). Hence, starting at some points z_0, where Eq. (19) still applies, we will assume that $a_1=a_2$. In this case one can readily obtain from Eq. (12) the area theorem for $z\geqslant z_0$:

$$\psi=\operatorname{ctn}\frac{\mathscr{P}}{2}\{1+\zeta\exp[D(z-z_0)\,]\}/\{1-\zeta\exp[D(z-z_0)\,]\}, \tag{21}$$

where

$$\zeta=\left(\psi_0-\operatorname{ctn}\frac{\mathscr{P}}{2}\right)\Big/\left(\psi_0+\operatorname{ctn}\frac{\mathscr{P}}{2}\right),$$
$$\psi_0=\psi(z=0),\quad D=[\gamma^2\langle G_1\rangle^2-\delta^2]^{1/2}.$$

In accordance with Eq. (21), the dependence of USP fields $A_{1,2}$ on the coordinates and time is

$$A_{1,2}^2(z,t)$$
$$=\frac{A_{1,2}^2(z=0)\,4\operatorname{ctn}^2(\mathscr{P}/2)\,(1+\psi_0^2)\,e^{D(z-z_0)}}{(\psi_0+\operatorname{ctg}(\mathscr{P}/2))^2[\operatorname{ctn}^2(\mathscr{P}/2)\,(1+\zeta e^{D(z-z_0)})^2+(1-\zeta e^{D(z-z_0)})^2]} \tag{22}$$

It is easy to see that in the limit, when $A_{01,02}\to 0$, Eq. (21) changes into the well-known theorem of cotangents for two-photon transitions of USP.[10] It follows from the area theorem (21) that pulses having an area

$$\theta=2\varkappa_{12}\hbar^{-1}(1+r^2/4)^{1/2}\int_0^\infty A_1A_2\,dt=\begin{cases}2|\mathscr{P}|+2\pi n & (\eta_e>0)\\ 2\pi(n+1)-2|\mathscr{P}| & (\eta_e<0)\\ 2\pi n\end{cases}$$

(n being an integer) propagate in the medium without energy

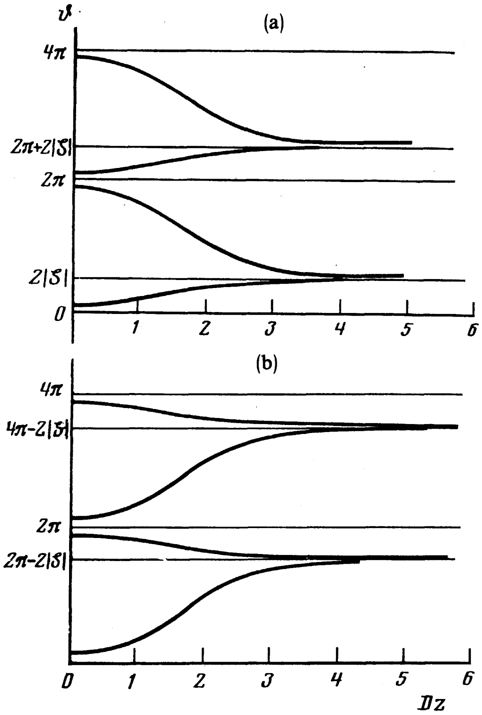

FIG. 1. Dependence of pulse area ϑ on distance, calculated from Eq. (21): (a) in a noninverted medium ($\eta_e > 0$) (b) in an inverted medium ($\eta_e < 0$); $|\mathscr{S}| = 0.25\pi$.

change, i.e., are SIT pulses. It is evident from Fig. 1 that the areas of SIT pulses $2|\mathscr{S}| + 2\pi n$ ($n_e > 0$) and $2\pi(n+1) - 2|\mathscr{S}|$ ($\eta_e < 0$) are stable, in contrast to $2\pi n$ pulses.

Somewhat unexpected was found to be the pattern of propagation of a 2π pulse in the system under consideration. As is evident from Fig. 2 [plotted in accord with Eq. (22)], the pulse breaks up into two subpulses. The first subpulse propagates at a velocity greater than $v \approx c/n$, and its area approaches $2|\mathscr{S}|$ (when $\eta_e < 0$, it approaches $2\pi - 2|\mathscr{S}|$). The second subpulse has a velocity less than v. When $\vartheta_0 < 2\pi$ [see Fig. 2(c)], the second subpulse attenuates. If $\vartheta_0 = 2\pi$, the area of the second subpulse tends to $2\pi - 2|\mathscr{S}|$ [see Fig. 1; 2(a)]. When $\vartheta_0 > 2\pi$ (but $\vartheta_0 < 2\pi - 2|\mathscr{S}|$), the area of the second subpulse approaches 2π [Fig. 1; 2(b)]. The duration of both subpulses decreases upon the propagation, whereas their energies (areas) remain practically unchanged. As can readily be seen from Eq. (22), the contraction of USP takes place much faster than in the absence of the resonant field (in the first case, the law of decrease in duration is exponential, and in the second case, quadratic; see Ref. 10).

Let us find the possible stationary (self-similar) solutions of Eq. (10). Let V be the group velocity of a stationary pulse. We change to a coordinate system which moves with the pulse ($z = Vt \to \xi$, $z \to z$). Assuming that $A_{1,2}$ and with it R, I, and η depends only on ξ, and performing elementary calculations, we obtain from Eqs. (5)–(9)

$$A_1A_2 = 4\langle R_0\rangle^2 \exp\left(\frac{\langle R_0\rangle\gamma v}{v-V}(\xi-\xi_0)\right)\frac{1}{(b_1^2-4\langle R_0\rangle^2 b_2)^{1/2}}$$
$$\times\left\{\left[1+\exp 2\left(\frac{\langle R_0\rangle\gamma v}{v-V}(\xi-\xi_0)\right)\right]\right.$$
$$\left.-2b_1\exp\left(\frac{\langle R_0\rangle\gamma v}{v-V}(\xi-\xi_0)\right)\right\}^{-1}, \quad (23)$$

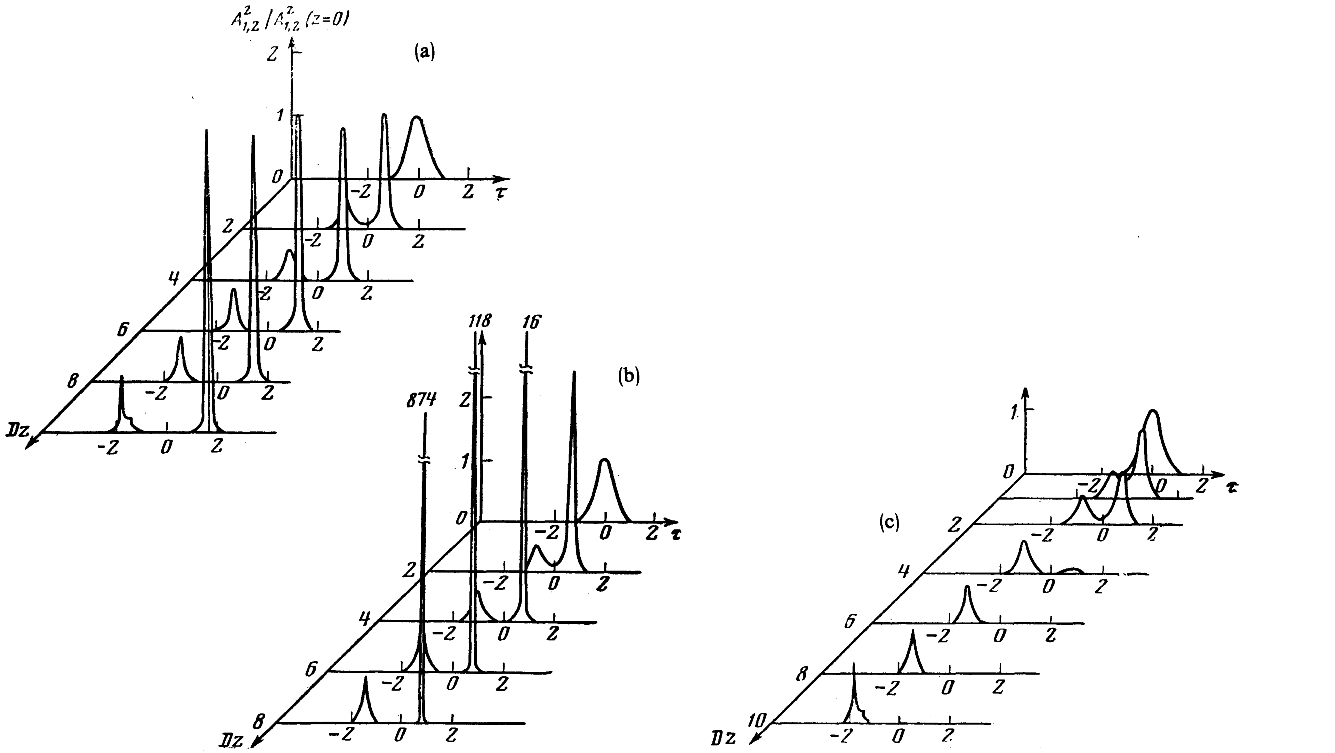

FIG. 2. Evolution in space and time of a pulse with a Gaussian envelope in the absorbing medium ($\eta_e > 0$); $\mathscr{S} = -0.25\pi$; initial pulse area: (a) $\vartheta_0 = 2\pi$, (b) $\vartheta_0 = 2.05\pi$, (c) $\vartheta_0 = 1.95\pi$.

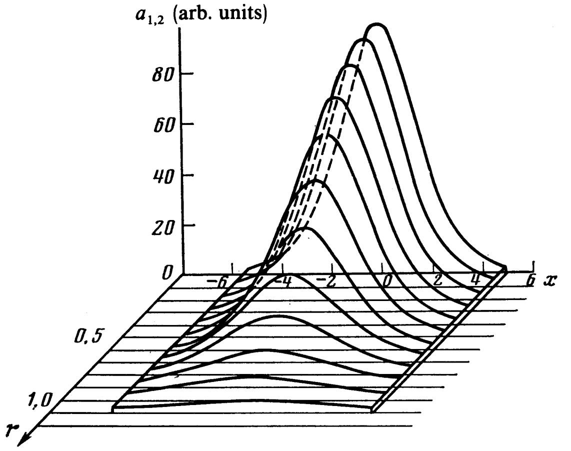

FIG. 3. Distribution of fields $a_{1,2}$ in a $2|\mathscr{S}|$ pulse with respect to the transverse coordinate r and time $[x = \gamma v(\xi - \xi_0)/(v - V)]$, when the beam of the exciting field has a Gaussian distribution of intensity over the cross section $[E_0 = E_0^m \exp(-\pi r^2/2)]$.

where

$$b_1=2g(\langle\eta_0\rangle-\langle I_0\rangle F), \quad b_2=-g^2(1+F^2),$$
$$g=2\varkappa_{12}(v-V)/\gamma\hbar V v.$$

It is easy to see that in an absorbing medium ($\eta_0 > 0$), the area of the "fast" subpulse ($V > v$) does indeed become equal to $2|\mathscr{S}|$, and the area of the "slow" subpulse ($V < v$) becomes equal to $\vartheta = 2\pi - 2|\mathscr{S}|$; conversely, in an amplifying medium ($\eta_0 < 0$), the area of the "fast" subpulse is $\vartheta = 2\pi - 2|\mathscr{S}|$, and the area of the "slow" subpulse is $\vartheta = 2|\mathscr{S}|$. Let us note one important feature of $2|\mathscr{S}|$ pulses. If the beam of the exciting field is limited in cross section, $\mathscr{S}\to 0$ as the distance from the center of the beam increases [see Eqs. (23), (15)]. Then the $2|\mathscr{S}|$ SIT pulse is also limited in cross section. Figure 3 shows the field distribution in the $2|\mathscr{S}|$ pulse as a function of the transverse coordinate r and of time, when the exciting field E_0 in the cross section has a Gaussian profile. Let us recall that the SIT pulses found in Refs. 3 and 28 in one- and two-photon resonances are in principle pulses of plane waves with infinite transverse dimensions. In contrast to the latter, the $2|\mathscr{S}|$ pulse can be realized in practice, and the SIT effect for it can be observed along the entire cross section of the pulse.

This section discusses the case in which $\mathscr{S}$ = constant. It follows from the generalized theorem of areas (12) that if $\mathscr{S}$ changes fairly slowly along z (near its mean value), the pulse area ϑ will be able to follow the change in $\mathscr{S}$ adiabatically. In this case, all the results obtained above for $\mathscr{S}$ = const. will remain valid. As can be readily seen from Eq. (12), the adiabaticity condition is

$$|d\mathscr{S}/dz| \ll S(1-\cos\mathscr{S}).$$

5. ALLOWANCE FOR THE FINITENESS OF RELAXATION TIME. STABILITY TO RELAXATION

As can readily be ascertained from Fig. 1, the possibility of stability of $2|\mathscr{S}|$ ($\eta_e > 0$) and $2\pi - 2|\mathscr{S}|$ ($\eta_e < 0$) SIT pulses to external disturbances is implied in the area theorem itself. As such an external disturbance, we will consider the relaxation of the medium and study its effect on the coherent propagation of USP, assuming $\tau_{\rm pul}\tau^{-1}$, $\tau_{\rm pul}T^{-1}$ to be small parameters. We will seek the solution of the system of Eqs. (1), (3), (4) (in the approximation $\Delta_\perp C_j = 0$) in the form

$$C_{1,2}=C_{1,2}^{(0)}+\Delta C_{1,2}, \quad \sigma=\sigma^{(0)}+\Delta\sigma, \quad \eta=\eta^{(0)}+\Delta\eta, \qquad (24)$$

where $C_{1,2}^{(0)}$, $\sigma^{(0)}$, $\eta^{(0)}$ are solutions for the fields and density matrix elements, obtained above on the assumption that $\tau = T = \infty$, and $\Delta C_{1,2}$, $\Delta\sigma$, $\Delta\eta$ are small corrections ($|\Delta C_{1,2}| \ll |C_{1,2}^{(0)}|$, $|\Delta\sigma| \ll |\sigma^{(0)}|$, $|\Delta\eta| \ll |\eta^{(0)}|$). We integrate both parts of Eq. (5) and substitute η from Eq. (24) into the right side of the expression obtained, keeping only first-order terms. We obtain the equation for the pulse area in the first approximation:

$$\partial\vartheta/\partial z = s[\cos(\mathscr{S}+\vartheta)-\cos\mathscr{S}+\delta\eta], \qquad (25)$$

where

$$\delta\eta=\langle\Delta\eta\rangle+\tau^{-1}\int_0^t\langle\eta^{(0)}-\eta_0\rangle dt.$$

The correction $\Delta\eta$ is easy to determine by substituting the solutions in the zeroth approximation into the right sides of Eqs. (3) and (4), and integrating. We will study the obtained equation (25). As is evident from the latter, the area ϑ is stable to perturbations of $\delta\eta$ related to relaxation. Small changes in $\delta\eta$ lead to only to small changes in ϑ. The stability region of the pulse area with respect to $\delta\eta$ is limited by the inequalities $\cos\mathscr{S} - 1 < \delta\eta < \cos\mathscr{S} + 1$. The solution for ϑ from Eq. (25) will differ little from $\vartheta^{(0)}$, and correspondingly, $A_{1,2}$ will differ little from $A_{1,2}^{(0)}$ [and hence, Eq. (24) will hold] if

$$|\delta\eta| \ll |\cos(\vartheta+\mathscr{S})-\cos\mathscr{S}|. \qquad (26)$$

When $z\to\infty$ the steady-state value of the pulse area is $\vartheta_{\rm st} \to \vartheta^{(0)}(z=\infty) + \Delta\vartheta$. As can readily be seen from Eq. (25), the correction $\Delta\vartheta$ is given by

$$\delta\eta=\cos(\mathscr{S}+\vartheta_{\rm st})-\cos\mathscr{S}, \qquad (27)$$

whence

$$|\Delta\vartheta| \approx \delta\eta/\sin\mathscr{S}.$$

The stability mechanism consists in the following. Thanks to the parametric coupling between the resonance field and the transformed USP fields, energy transfer takes place from $A_{01,02}$ to $A_{1,2}$ and vice versa. If $\delta\eta < 0$ (energy decrease due to relaxation), the pulse area will be such that $\vartheta_{\rm st} > 2\pi n + 2|\mathscr{S}|$ [or $\vartheta_{\rm st} < 2\pi(n+1) - 2|\mathscr{S}|$ when $\eta_e < 0$]. If however $\delta\eta > 0$, then, conversely, we will have $\vartheta_{\rm st} < 2\pi n + 2|\mathscr{S}|$ [or $\vartheta_{\rm st} > 2\pi(n+1) - 2|\mathscr{S}|$ when $\eta_e < 0$]. In both cases $\vartheta_{\rm st}$ is given by Eq. (27). In other words, the decrease or increase of pulse energy due to relaxation is compensated by an increase or decrease of energy, respectively, as a result of mutual energy transfer between the fields being transformed and the resonance field. Ultimately, the formation of a single pulse propagating in the medium without energy change and stable to relaxation is possible.

The condition (26) will necessarily be satisfied if

$$\Delta_m \ll |\cos(\mathscr{S}+\vartheta)-\cos\mathscr{S}|, \qquad (28)$$

where, as indicated by estimates of the correction $\delta\eta$,

$$\Delta_m = t\tau^{-1}q|\cos(\mathscr{S}+\vartheta^{(0)}) - \cos\mathscr{S}| + 2\vartheta^{(0)}tT^{-1}(1+T/\tau)^{1/2} > |\delta\eta|.$$

Thus (28) is the stability condition for coherent propagation of USP.

To conclude this section, we will consider for comparison the case of "pure" two-photon absorption (amplification), when the resonance field corresponds to ($\mathscr{S}=0$). Integrating both parts of Eq. (5) and substituting $\eta^{(0)}$ into the right side of the expression obtained, we have

$$\partial\vartheta/\partial z = \gamma\eta_0 a^{-1}(\cos\vartheta - 1 + \Delta), \tag{29}$$

where

$$\Delta = \tau^{-1}\int_0^t (\cos\vartheta - 1)\,dt < 0.$$

It is evident from Eq. (29) that the presence of the nonzero correction Δ, caused by relaxation, leads to loss of the coherence of propagation of USP. In an absorbing medium, the pulse ultimately damps out completely. In an amplifying medium, an unlimited growth of the pulse area (energy) should be observed. In other words, in the absence of the resonance field, instability of SIT pulses to relaxation takes place.

6. STABILITY TO TRANSVERSE INHOMOGENEITY OF LIGHT FIELDS

We will study the influence of diffraction on the coherent propagation of USP in an excited medium. Drabovich *et al.*[22] and Bol'shov *et al.*[29] showed that the coherent propagation of USP in both one-photon and two-photon-absorbing media involves an instability to the transverse inhomogeneity of light beams. Nonstationary self-focusing develops in the course of the propagation. Diffraction ultimately causes the pulse to damp out completely.[15,29] It will be shown that in the presence of excitation of the medium by a resonant field, SIT pulses are stable to transverse inhomogeneity, and the propagation of USP over large distances without energy change is possible. The situation in this case is in many ways similar to the one discussed above. As in the preceding section, we will seek the solution of Eq. (1) in the form

$$C_{1,2} = C_{1,2}^{p} + \Delta C_{1,2}, \tag{30}$$

where $C_{1,2}^{p}$ is the solution of Eq. (1) in the plane-wave approximation (with $\tau = T = \infty$). $\Delta C_{1,2}$ is a small addition $|\Delta C_{1,2}| \ll |C_{1,2}^{p}|$. Using Eq. (30) and integrating Eq. (1), we obtain to a first approximation

$$\partial\vartheta/\partial z = s[\cos(\vartheta+\mathscr{S}) - \cos\mathscr{S} - \mathscr{D}], \tag{31}$$

where

$$\mathscr{D} = \frac{1}{sk_1}\int_0^t \left(\frac{\partial\varphi_1}{\partial x}\frac{\partial\dot\vartheta_p}{\partial x} + \dot\vartheta_p\frac{\partial^2\varphi_1}{\partial x^2} + \frac{\partial\varphi_1}{\partial y}\frac{\partial\dot\vartheta_p}{\partial y} + \dot\vartheta_p\frac{\partial^2\varphi_1}{\partial y^2}\right)dt, \tag{32}$$

$$\vartheta_p = \vartheta(A_{1,2} = A_{1,2}^{p}).$$

We will study the obtained equation (31). The solution for ϑ from Eq. (31) will differ little from ϑ_p (correspondingly, $A_{1,2}$ will differ little from $A_{1,2}^{p}$) if

$$|\mathscr{D}| \ll |\cos(\vartheta+\mathscr{S}) - \cos\mathscr{S}|. \tag{33}$$

When $z\to\infty$, the steady-state pulse area is $\vartheta_{st} = \vartheta_p(z\to\infty) + \Delta\vartheta$, where, as can readily be seen from Eq. (31), ϑ_{st} is given by

$$\mathscr{D} - \cos(\vartheta_{st} + \mathscr{S}) + \cos\mathscr{S} = 0, \tag{34}$$

whence $|\vartheta| \approx |\mathscr{D}(z=\infty)/\sin\mathscr{S}|$. Physically, the condition (34) means that the pulse energy change due to diffraction is compensated by the energy transfer between the USP fields $A_{1,2}$ and the resonance field.

Since the light pulse in the cross section is always limited, of greatest practical interest is the question of stability of the aperture-limited $2|\mathscr{S}|$ SIT pulses obtained above. Let us estimate the diffraction term $\mathscr{D}$. We will assume for simplicity that $|\langle R_0\rangle|$, $|\langle I_0\rangle| \ll 1$. As shown by the calculations, to estimate the phase derivative $\partial\varphi/\partial r$ in this case, use may be made of the geometrical optics approximation ($k\to\infty$) and the eikonal equation.[30,31] We thus obtain

$$|\mathscr{D}| < \mathscr{D}_m = s^{-1}(N|\varkappa^{11} - \varkappa^{mm}|/2\pi)^{1/2}(1/2|\partial\mathscr{S}/\partial r| + 2|\mathscr{S}|)r_0^{-1}, \tag{35}$$

where r_0 is the characteristic radius of the beam of the exciting resonant field. It can be shown that in order to satisfy the condition of stability of the plane-wave approximation (33), it suffices to require that

$$\mathscr{D}_m \ll 1 - \cos\mathscr{S}. \tag{36}$$

Some numerical estimates are in order. We assume the transverse intensity distribution in the beam of field A_{01} to be Gaussian. For $2\hbar^{-1}(\tau T)^{1/2}A_{01}(r=0) = 1$ [assuming for simplicity that $\delta = 0$, $g(\nu) = \delta(\nu)$], $\tau \sim 10^{-7}$ *s*, $T \sim 10^{-9}$ s (times characteristic of gaseous media), we obtain from Eqs. (35), (36).

$$(r_0\gamma)^{-1}(N|\varkappa^{11} - \varkappa^{mm}|/2\pi)^{1/2}(\pi r + 4)\,\mathrm{ch}\,(\pi r^2/2) \ll 0{,}1. \tag{37}$$

It is evident from (37) that for large r, the conditions (37) as well as (36) will break down. Physically this means the following. Far from the axis, the resonance field is low. It can no longer compensate for the diffraction, so that the distributions of the fields remains close to (23). Thus inequality (37) determines the region around the axis of the beam of radius r_b where the pulse envelope will be close to (23). Beyond the confines of this region ($r > r_b$) there will apparently be established a steady-state field distribution [satisfying a condition of the type of Eq. (34)] that can differ appreciably from (23). If one takes the following typical parameters: $r_0 = 3$ *mm*, $\kappa_{12} \sim 10^{-22}$ cm^3, $|\kappa^{11} - \kappa^{mm}| \sim 10^{-22}$ cm^3, $\gamma \approx 5$ cm^{-1}, then $r_b \approx \sqrt{2}$, i.e., the diffraction being taken into account, the steady-state distribution of the fields will be close to (23) practically for the entire pulse, with the exception of peripheral areas, where the intensity is hundreds of times less than at the maximum.

As was done in Secs. 5 and 6, one can apparently prove the stability of SIT pulses to inhomogeneous line broadening as well.

7. DESCRIPTION OF PROPOSED EXPERIMENT

Let us take cesium vapor as the resonant medium; the working levels are $6^2S_{1/2}$–$9^2D_{3/2}$. The frequency of the working transition $\omega_{ml} = 28818.90$ cm^{-1} coincides with twice the frequency of a ruby laser if the ruby sample is cooled to a temperature of ≈ 148 K. Thus the second harmonic of the ruby laser may be used as the exciting resonance field. Picosecond laser pulses may be used as the source of trigger pulses. To increase the polarizability of the two-photon transition, κ_{12}, the frequency ω_1 of the trigger field may be chosen close to the frequency of the intermediate transition $6^2S_{1/2}$–$6^2P_{3/2}$ (or $6^2P_{3/2}$–$9^2D_{3/2}$). Phase synchronism in the system can be achieved by introducing a buffer gas along with the vapor into the cell.[32] The cell containing the cesium vapor is placed in an electrostatic field; when $\kappa_{20}\approx 10^{-23}$ cm^3 and $\mathscr{E}_0\approx 100$ KV/cm, the induced dipole moment is $d=\kappa_{20}\mathscr{E}_0\approx 10^{-21}$ cgs. Assuming $\kappa_{12}\approx 10^{-22}$ cm^3 at frequencies $\omega_1\approx 17{,}000$ cm^{-1}, $\omega_2\approx 11{,}819$ cm^{-1}, with relaxation times $\tau\sim 10^{-7}$ s, $T\sim 10^{-8}$ s and vapor density $N=10^{16}$ cm^{-3}, we obtain $D\approx 1.6$ cm^{-1}, $\gamma\approx 5.4$ cm^{-1}. When the power of the exciting resonance field is 0.31 MW/cm^2 [$A_{01}^2(z=0)\approx 625$ cgs] $\mathscr{S}=-0.25$. Let a trigger pulse at frequency ω_1 of power 155 kW/cm^2 ($A_{10}^2\approx 300$ cgs) and duration $\tau_p\approx 30$ ps be supplied to the input of the cell. In this case, a proportional field pulse at frequencies ω_1, ω_2 is formed on a length of ≈ 3.3 cm The area of such a pulse ϑ approaches $2|\mathscr{S}|(\vartheta\approx 0.95\cdot 2|\mathscr{S}|)$ at a length ≈ 5.3 cm, i.e., an interaction regime close to SIT is established. As the propagation continues, the pulse becomes compressed: at a length of 13.3 cm, the pulse duration decreases by a factor of approximately 5. It is evident from the estimates that appreciable changes in pulse energy and duration occur at lengths at which the resonant field undergoes little change. In this case, use may be made of an electrostatic field $\mathscr{E}_0$ constant along z. At greater lengths, $\mathscr{E}_0$ should increase along z in accordance with (18) ($K\approx 0.06$ cm^{-1}). To compensate for the decrease of the resonance field, focusing of the exciting radiation inside the cell may also be used (at constant $\mathscr{E}_0$). Estimates show that for a pulse with Gaussian intensity distribution in the cross section and characteristic radius $r_0\approx 3$ mm [$|\kappa^{11}-\kappa^{mm}|\sim 10^{-22}$ cm^3, the remaining parameters are given above in this section], the stability condition of the plane-wave approximation (36) is satisfied for practically any pulse with the exception of the areas distant from the beam axis ($r\geqslant\sqrt{3}$), where the intensity is four orders of magnitude less than at the maximum [see (23)]. It is easy to see that (when $\tau\sim 10^{-7}$ s, $T\sim 10^{-8}$ s, $\tau_p\approx 30$ ps) in the same region ($r\leqslant\sqrt{3}$), the stability condition for SIT $2|\mathscr{S}|$ pulses to the relaxation of the medium is satisfied.

Let us note in conclusion that the medium can also be excited by other means, for example, with the aid of SRS or two-photon absorption in the 1, m levels. In this case, it is no longer necessary to use an electrostatic field, and there is no qualitative change in the interaction.

8. CONCLUSION

This paper studied the resonant interaction of ultrashort light pulses in a medium with two-photon transitions in the case of pure excitation of the system with a field of a frequency at resonance with the transition frequency.

1. It is shown that in such systems, the pulse area (energy) of selfinduced transparency is $\vartheta=2\pi n+2|\mathscr{S}|$[or $\vartheta=2\pi(n+1)-2|\mathscr{S}|$ in an amplifying medium; $n=0, 1, ...$)], where $\mathscr{S}$ is determined by the polarization and difference of the populations induced in the medium by the exciting field.

2. In the course of propagation in the medium, a 2π pulse splits into two subpulses, the first of which (fast) travels at a group velocity $V>v\approx c/n$ and has an area equal to $2|\mathscr{S}|$, and the second (slow) has a group velocity $V\leqslant v\approx c/n$ and area $2\pi-|2|\mathscr{S}|$. In the general case, the $2\pi n$ pulse splits into $n+1$ subpulses whose duration decreases, and the power increases with the distance. The rate of compression of such subpulses, determined by the dependence of pulse duration and of maximum intensity on distance, is higher in the interaction considered than the rate of compression of analogous subpulses during two-photon absorption (TPA) in the abscence of the resonance field. For example, for rectangular input pulses, the subpulses are compressed exponentially with distance, in the first case and quadratically in the second.

3. It is shown that in the presence of the exciting field, SIT pulses can be stable to relaxation of the medium and to the transverse inhomogeneity of the light fields. The stability regions were determined.

4. It was found that SIT $2|\mathscr{S}|$ pulses can be generated by a weak "priming" at the frequency of the trigger field, and independently of the phase modulation of the trigger field. The energy of a $2|\mathscr{S}|$ pulse can change continuously over a wide range as the exciting field changes. The transverse distribution of the field in a $2|\mathscr{S}|$ pulse is completely determined by that for the exciting field. In contrast to 2π pulses, the SIT effect can here be observed along the entire cross section of aperture-limited $2|\mathscr{S}|$ pulses. As the propagation goes on, the $2|\mathscr{S}|$ pulse is compressed. It is shown that for typical radiation characteristics and parameters of the medium, SIT pulses $2|\mathscr{S}|$ [in the form (28)] are stable to relaxation and to the transverse inhomogeneity of the light fields.

Since the area of the $2|\mathscr{S}|$ pulse or the total angle of rotation of the Bloch vector is determined by the angle of rotation of the Bloch vector taken with the opposite sign for the exciting field (in the same system of levels), degeneracy of the levels apparently will not affect the existence of SIT $2|\mathscr{S}|$ pulses.

Thus the interaction considered may be used for contraction of ultrashort pulses and also for observing SIT in two-photon absorption (and other coherent phenomena related to this transparency) due to pulses with low power and an appreciable phase modulation, for which coherent phenomena are not observed in the usual case of TPA.

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[1)] A situation typical of the majority of frequency conversion problems.
[2)] Equations (3) and (14) differ from the equations derived by Mikaelyan *et al.*[27] for lasers only in the term proportional to $\kappa_0^{mm}-\kappa_0^{11}$.

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