Soliton regimes of stimulated Raman scattering

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It is shown that in strong ultrashort pulse fields, specific SRS (stimulated Raman scattering) regimes arise which are not accompanied by amplification (SRS solitons). The existence of any of the regimes depends on the initial conditions and the dispersive properties of the medium. The stability of a number of soliton regimes with respect to weak perturbations of various kinds is demonstrated. The results are applied to the case of two-photon resonance absorption of pulses of different frequencies.

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1. INTRODUCTION

The effect of self-induced transparency, which arises in the passage of powerful ultrashort light pulses through a medium, has been widely studied in recent years.[1] Interest in this phenomenon is connected both with the fact that one can extract additional information on the optical characteristics of matter (the determination of the constants of radiative transitions, the longitudinal and transverse relaxation times), and with the possibility of obtaining supershort pulses. The theory of the effect of coherent illumination for a medium with single-photon resonance absorption was first given by McCall and Hahn.[2] It was shown in Refs. 1, 3, 4 that this effect takes place also in media with two-photon resonance and non-resonance absorption.

In our previous researches,[5] we established the possibility of the existence of the effect of self-induced transmission in the case of stimulated Raman scattering (SRS) under the conditions of the interaction of ultrashort pulses of exciting and Stokes radiation in a lossless medium (SRS solitons). Similar transmission arises only when the length of both pulses is much less than the transverse relaxation time T_2. The reason for the effect is that the energy absorbed by matter from the pulses is then coherently returned to the field as a result of the stimulated scattering. As a consequence, a stationary scattering regime becomes possible, in which the form of both pulses and their amplitudes do not change. It was found in Ref. 5 that for definite form of the initial conditions, the SRS solitons should be pulses of Lorentzian shape, and the value of the overlap integral is

$$\theta=\frac{\lambda}{\hbar}\int_{-\infty}^{\infty}E_i(\xi')E_s(\xi')\,d\xi'=2\pi, \tag{1}$$

where E_l, E_s are the real amplitudes of the pulses of the exciting and first Stokes radiation, λ is the scattering matrix element. The condition (1) shows the threshold character of the effect. Estimates show (see Ref. 5) that the upper boundaries of the threshold values of the fields E_l and E_s amount to $\sim(10-100)T_2^{-1/2}\approx 10^5-10^6$ V/cm at normal pressures in gases and $\sim 10^7$ V/cm in liquids. Under such conditions, a strong change in the level populations takes place during the scattering, which sharply distinguishes the situation from SRS in the field of quasistatic or ultrashort pulses with an intensity less than the given threshold (see Ref. 6). However, even if the threshold conditions are satisfied at the input to the medium, to make clear whether the SRS pulses go over to the soliton regime, it is necessary to solve the complete set of nonstationary equations which describe their temporal evolution. This can be done only in the special case of "proportional" input pulses, which are considered in Ref. 5. The numerical solution of the nonstationary equations cannot give a final answer to the problem. It is therefore of fundamental importance to consider the problem of the stability of the motion of the SRS solitons relative to small perturbations.

In the present work, we have found all the possible soliton regimes which can exist in the case of combination interaction of ultrashort pulses in a lossless medium (Secs. 2 and 3). Whether one obtains this or that type of soliton depends on the initial conditions and the dispersive properties of the medium. The stability of several soliton regimes is studied in Sec. 4, relative to the different types of small perturbations. In particular, the existence of perturbations is shown in which the solitons change their duration and velocity without change in the shape of the pulse.

2. BASIC EQUATIONS

We shall assume that the scattering takes place on one pair of levels of the scattering molecules, and consider the case of the one-dimensional problem for simplicity. In the envelope approximation, the set of equations which describes the evolution of the pulses of the exciting and first Stokes radiations of the SRS consists of the abbreviated Maxwell's equations for the amplitudes of the fields and the equations of motion for the averaged values of the polarization. In the case of resonance ($\omega_l - \omega_s = \omega_v$, where ω_l and ω_s are the frequencies of the scattering and Stokes fields, ω_v is the frequency of the operating transition) this set has the form1) [5]:

$$\begin{gathered}
\frac{\partial E_s}{\partial z}+\frac{1}{c_s}\frac{\partial E_s}{\partial t}=-\frac{1}{2k_s}\lambda\mu_0\omega_s^2N_v vE_i,\\
\frac{\partial E_i}{\partial z}+\frac{1}{c_i}\frac{\partial E_i}{\partial t}=\frac{1}{2k_i}\lambda\mu_0\omega_i^2N_v vE_s,\\
\frac{du}{dt}=-\frac{u}{T_2},\quad \frac{dv}{dt}=-\frac{v}{T_2}+\frac{\lambda}{\hbar}E_iE_sW,\\
\frac{dW}{dt}=-\frac{\lambda}{\hbar}E_iE_sv-\frac{W-W^{eq}}{T_1},
\end{gathered} \tag{2}$$

where u, v are the amplitudes of the transverse polarization, W is half the difference between the populations of the upper and lower levels, W^{eq} is the equilibrium value of W; T_1 and T_2 are the times of longitudinal and transverse relaxations; $\mu_0 = 4\pi/c^2$; N_V is the density of the scattering molecules; $c_{i,s} = (\partial\omega/\partial k)_{\omega=\omega_{i,s}}$ are the group velocities of the pump wave and the Stokes wave, respectively, which can be close to the phase velocities $c/\sqrt{\varepsilon(\omega_{i,s})}$ under the condition $\partial\varepsilon/\partial\omega \ll \varepsilon/\omega$ ($k_{i,s}$ are the moduli of the wave vector, $\varepsilon(\omega)$ is the dielectric constant of the medium). For the consideration of ultrashort pulses with durations much less than T_2 and T_1, we set $T_1 = T_2 = \infty$, as usual (account will be taken below of the effect of the finiteness of relaxation times). Then it is not difficult to reduce the set (2) to the form

$$\frac{\partial E_s}{\partial z} + \frac{1}{c_s}\frac{\partial E_s}{\partial t} = -\varkappa_s E_i \sin\varphi, \quad \frac{\partial E_i}{\partial z} + \frac{1}{c_i}\frac{\partial E_i}{\partial t} = \varkappa_i E_s \sin\varphi,$$
$$\varphi = \frac{\lambda}{\hbar}\int E_i(z,t) E_s(z,t)\,dt; \quad \varkappa_{i,s} = \lambda\mu_0 c\omega_{i,s} N_V W^{eq}/2\eta_{i,s}. \tag{3}$$

The quantity $\varphi(z,t)$ has the meaning of the turning angle of the polarization vector.

We now find the general soliton solution of the set (3), corresponding to stationary pulses E_i and E_s propagating with the same group velocity V. It has the form

$$\varphi = \varphi_0(t - z/V), \quad E_i = E_{0i}(t - z/V), \quad E_s = E_{0s}(t - z/V). \tag{4}$$

We note that the equality of the group velocities should guarantee the maximum degree of coherence of the interaction of the pulses. Introducing the variable $\xi = t - z/V$, we get, after substitution of (4) in (3),

$$\frac{dE_{0s}}{d\xi} = -\varkappa_s \frac{Vc_s}{V - c_s} E_{0i}\sin\varphi_0, \quad \frac{dE_{0i}}{d\xi} = \varkappa_i \frac{Vc_i}{V - c_i} E_{0s}\sin\varphi_0, \tag{5}$$

whence

$$E_{0s}^2 = a_s(C_1 - \cos\varphi_0), \quad E_{0i}^2 = a_i(C_2 - \cos\varphi_0), \tag{6}$$

where C_1, C_2 are constants of integration,

$$a_s = -\frac{2\hbar}{\lambda}\frac{Vc_s}{V - c_s}\varkappa_s, \quad a_i = \frac{2\hbar}{\lambda}\frac{Vc_i}{V - c_i}\varkappa_i. \tag{7}$$

From (5) and (6), we have the equation

$$d\varphi_0/d\xi = \{b(C_1 - \cos\varphi_0)(C_2 - \cos\varphi_0)\}^{1/2}, \tag{8}$$

where $b = \lambda^2/\hbar^2 a_i a_s$. Introducing the new variable $x = \tan(\varphi_0/2)$, we obtain the solution (8) in the form

$$\int_{x_0}^{x} \frac{dx}{\{b[C_1 - 1 + (C_2 + 1)x^2][C_2 - 1 + [C_2 + 1)x^2]\}^{1/2}} = \frac{1}{2}(\xi - \xi_0). \tag{9}$$

The integral (9) contains all the soliton solutions of the set (3). By specifying different C_1 and C_2, we obtain different classes of SRS solitons. Limitations on the dispersive characteristics of the medium, at which the existence of solitons of the given class is possible, follow here from the requirement of the positiveness of (6) and the form of Eqs. (7) for a_s, a_i ($W^{eq} < 0$). We note that the quantities C_1 and C_2 are connected with the initial values of the fields and the polarizations.

Thus, in the formation of solitons, an important role is played by the effects of group retardation. In this connection, it must be noted that account of dispersion in first approximation, to which Eqs. (2) correspond, is completely adequate for the problems of the optics of ultrashort pulses (see Ref. 6).

3. SOLITON SOLUTIONS OF THE SRS EQUATIONS

We now consider soliton regimes which correspond to various relations between the group velocities of the interacting waves (normal and anomalous dispersion) and different initial populations of the working levels of the molecules of the medium.

1. We find the solitons corresponding to the cases $C_1 = C_2 = \pm 1$.

A. Let $C_1 = C_2 = 1$. It then follows from (6) and (7) that the velocity V should lie in the interval $c_s < V < c_i$ (i.e., $a_i, a_s > 0$). The condition which arises here corresponds to the case of anomalous dispersion.[7] We integrate (9) with the initial conditions $x_0 = 0$, $\xi_0 = -\infty$, which, for given choice of constants C_1 and C_2, corresponds to $\varphi_0(-\infty) = 0$, $E_{0s}(-\infty) = 0$, $E_{0i}(-\infty) = 0$, i.e., at the initial moment of the interaction of the fields, the medium is in equilibrium. Then

$$\varphi_0(\xi) = 2[{}^1/_2\pi + \operatorname{arctg}(\xi/\tau)], \quad \theta = \varphi_0(\infty) - \varphi_0(-\infty) = 2\pi, \tag{10}$$

$$E_{0s}^2 = \frac{2a_s}{1 + (\xi/\tau)^2}, \quad E_{0i}^2 = \frac{2a_i}{1 + (\xi/\tau)^2}, \quad \tau^{-1} = \sqrt{b}, \tag{11}$$

which agrees with solutions of (1) obtained[5] by another method.

B. Let $C_1 = C_2 = -1$. The soliton solutions are possible upon satisfaction of the condition $c_i < V < c_s$ ($a_i, a_s < 0$), which corresponds to the region of normal dispersion. Integrating (9) in the case $x_0 = 0$, $\xi_0 = 0$, we obtain $x = \xi\sqrt{b}$. From (6), we have

$$E_{0s}^2 = -\frac{2a_s}{1 + (\xi\sqrt{b})^2}, \quad E_{0i}^2 = -\frac{2a_i}{1 + (\xi\sqrt{b})^2}, \tag{12}$$

and $\varphi_0(-\infty) = -\pi$, $\varphi_0(\infty) = \pi$ and $\theta = 2\pi$. It then follows that in the case $t = -\infty$, the considered pair of levels should have an inverted population ($W(-\infty) = W^{eq} \times \cos\varphi_0(-\infty)$). Thus, the propagation of 2π-pulses of SRS of Lorentzian shape in a medium with normal dispersion is possible only under the condition of inverted initial population.

2. We now consider the cases $C_1 = C_2 = C > 1$, $C_1 = C_2 = C < -1$.

A. Let $C > 1$. As in the class 1A, we obtain the condition $c_s < V < c_i$ ($a_{i,s} > 0$), which is satisfied in the region of anomalous dispersion. According to (9),

$$\varphi_0(\xi) = 2\operatorname{arctg}\left[\left(\frac{C-1}{C+1}\right)^{1/2}\operatorname{tg}\left(\frac{1}{2}\xi\sqrt{b(C^2-1)}\right)\right], \tag{13}$$

whence

$$E_{0s}^2 = \frac{a_s(C^2-1)}{C + \cos(\xi\sqrt{b(C^2-1)})}, \quad E_{0i}^2 = \frac{a_i(C^2-1)}{C + \cos(\xi\sqrt{b(C^2-1)})}. \tag{14}$$

The solitons (14) represent an unbounded periodic succession of pulses with period $T=2\pi/\sqrt{b(C^2-1)}$. The value of the overlap integral over one period is equal to

$$\theta_T=\frac{\lambda}{\hbar}\int_0^T E_{0i}E_{0s}\,d\xi=2\pi,$$

i.e., the solitons (14) are an infinite train of 2π pulses (we shall call them solitons of the trigonometric type). We note that in the given case, absence of relaxation processes is assumed ($T_1=T_2=\infty$). Therefore, in the case of 2A, it is necessary that $T\ll T_2$. In the real situation, the effect of finite T_2 should lead to a gradual incoherent dephasing of the scatterers and consequently to a cutoff of the train. Thus the number of pulses in the train is $\sim T_2/T$.

B. Let $C<-1$. Here the condition $c_i<V<c_s$ ($a_{i,s}<0$) follows from (6), (7). This condition is satisfied in the case of normal dispersion. The solution (9) leads to the expressions (13), (14). The solitons form an infinite train of 2π pulses of the trigonometric type.

3. We consider the case $C_1>1$, $C_2>1$, where $C_1\neq C_2$. It follows from (6) that $c_s<V<c_i$ is the case of anomalous dispersion.

A. Let $C_1>C_2$. We introduce the parameters $\alpha^2=(C_1-1)/(C_1+1)$, $\beta^2=(C_2-1)/(C_2+1)$ and make the change of variables $x=\beta\tan\psi$. Integration of (9) with the boundary conditions $x_0=0$ ($\psi=0$), $\xi_0=0$ gives

$$F\left(\psi,\frac{(\alpha^2-\beta^2)^{1/2}}{\alpha}\right)=\frac{1}{2}\tau^{-1}\xi,$$

where $V(\psi,q)$ is the elliptic integral of the first kind. Then

$$\varphi_0(\xi)=2\,\mathrm{arctg}\left\{\beta\,\mathrm{tn}\left[\frac{1}{2}\tau^{-1}\xi\right]\right\}; \tag{15}$$

$$E_{0s}^2=a_s\frac{C_1C_2-1+(C_1-C_2)\,\mathrm{cn}(\xi/\tau)}{C_2+\mathrm{cn}(\xi/\tau)},$$
$$E_{0i}^2=\frac{a_i(C_2^2-1)}{C_2+\mathrm{cn}(\xi/\tau)},\quad \tau^{-1}=[b(C_1-1)(C_2+1)]^{1/2}, \tag{16}$$

where cn and tn are the elliptic functions. The solutions of (16) represent an infinite periodic train with period $T=4K(q)\tau$, where $q^2=2(C_1-C_2)/(C_1-1)(C_2+1)$; $K(q)$ is the complete elliptic integral of the first kind. It is not difficult to obtain the result that $\theta_T=\varphi_0(2K\tau)-\varphi_0(-2K\tau)=2\pi$, i.e., the solitons (16) are infinite trains of 2π pulses (we shall call them solitons of the elliptic type).

B. Let $C_1<C_2$. In this case we obtain solutions similar to 3A:

$$\varphi_0(\xi)=2\,\mathrm{arctg}\,\{\alpha\,\mathrm{tn}\,(\xi/2\tau)\}; \tag{17}$$

$$E_{0s}^2=\frac{a_s(C_1^2-1)}{C_1+\mathrm{cn}(\xi/\tau)},$$
$$E_{0i}^2=a_i\frac{C_1C_2-1+(C_2-C_1)\,\mathrm{cn}(\xi/\tau)}{C_1+\mathrm{cn}(\xi/\tau)},\quad \tau^{-1}=\sqrt{b(C_1+1)(C_2-1)}; \tag{18}$$

they are infinite trains of 2π pulses of the elliptic type with period $T=4K(q)\tau(q^2=2(C_2-C_1)/(C_1+1)(C_2-1))$.

4. We consider the case $C_1<-1$, $C_2<-1$, $C_1\neq C_2$. In this case, $c_i<V<c_s$ is the region of normal dispersion. It is not difficult to see that the solutions represent infinite trains of 2π pulses, which in the case $C_1>C_2$ have the form (15), (16) and in the case $C_1<C_2$—(17), (18).[2)]

5. The following cases are considered that are similar to classes 3 and 4.

A. The case $C_1>1$, $C_2<-1$, in which the conditions $V>c_s$, $V>c_i$ ($a_s>0$, $a_i<0$).

B. The case $C_1<-1$, $C_2>1$, in which the conditions $V<c_s$, $V<c_i$ ($a_s<0$, $a_i>0$).

Limitations on the dispersive properties of the medium do not arise in either case. Solitons are infinite periodic trains of 2π pulses, described in the case 5A by the expressions (17), (18), and in case 5B by the expressions (15), (16).

6. The study of the solutions (9) shows that in the cases $C_1\neq\pm1$, $C_2=1$; $C_1=1$, $C_2\neq\pm1$ there are no soliton solutions having physical meaning in the case $\xi>0$.

7. We now consider the case in which one of the constants is equal to -1 and the second is greater than 1 in modulus.

A. Let $C_1<-1$, $C_2=-1$ ($c_i<V<c_s$ is the region of normal dispersion). In this case, integration of (9) at $x_0=0$, $\xi_0=0$ gives

$$\varphi_0(\xi)=2\,\mathrm{arctg}\left[\left(\frac{C_1-1}{C_1+1}\right)^{1/2}\mathrm{sh}\left(\frac{\xi}{2\tau}\right)\right],\quad \tau^{-1}=[-2b(C_1+1)]^{1/2} \tag{19}$$

We obtain the following expressions for the fields (we shall call them solitons of the hyperbolic type):

$$E_{0s}^2=\frac{a_s(C_1-1)\,\mathrm{ch}^2(\xi/\tau)}{1+[(C_1-1)/(C_1+1)]\,\mathrm{sh}^2(\xi/\tau)},$$
$$E_{0i}^2=-\frac{2a_i}{1+[(C_1-1)/(C_1+1)]\,\mathrm{sh}^2(\xi/\tau)}. \tag{20}$$

Here $\varphi_0(-\infty)=-\pi$, $\varphi_0(\infty)=\pi$, and $\theta=2\pi$. The fields (20) represent 2π pulses, and for the case $t=-\infty$ the medium is inverted ($\varphi_0(-\infty)=-\pi$).

B. In the case $C_1=-1$, $C_2<-1$ ($c_i<V<c_s$) we have

$$\varphi_0(\xi)=2\,\mathrm{arctg}\left[\left(\frac{C_2-1}{C_2+1}\right)^{1/2}\mathrm{sh}\left(\frac{\xi}{2\tau}\right)\right],\quad \tau^{-1}=[-2b(C_2+1)]^{1/2}$$
$$E_{0s}^2=-\frac{2a_s}{1+[(C_2-1)/(C_2+1)]\,\mathrm{sh}^2(\xi/\tau)}, \tag{21}$$
$$E_{0i}^2=\frac{a_i(C_2-1)\,\mathrm{ch}^2(\xi/\tau)}{1+[(C_2-1)/(C_2+1)]\,\mathrm{sh}^2(\xi/\tau)}. \tag{22}$$

Just as in case A, $\varphi_0(-\infty)=-\pi$, $\varphi_0(\infty)=\pi$, $\theta=2\pi$, i.e., the solutions (22) are 2π pulses.

C. In the cases $C_1>1$, $C_2=-1$ ($V>c_s$, $V>c_i$—arbitrary dispersion) and $C_1=-1$, $C_2>1$ ($V<c_s$, $V<c_i$—arbitrary dispersion), the soliton solutions are described by Eqs. (19), (20) and (21), (22), respectively.

8. We consider the following cases.

A. $C_1=1$, $C_2=-1$ ($V>c_s$, $V>c_i$—arbitrary dispersion). In this case,

$$\varphi_0(\xi)=2\,\mathrm{arctg}\,[e^{\xi/\tau}],\quad \tau^{-1}=\sqrt{b},$$
$$E_{0s}^2=\frac{2a_se^{2\xi/\tau}}{1+e^{2\xi/\tau}},\quad E_{0i}^2=-\frac{2a_i}{1+e^{2\xi/\tau}}. \tag{23}$$

B. $C_1=-1$, $C_2=1$ ($V<c_s$, $V<c_i$—arbitrary dispersion). In this case, the solutions are obtained from (23) by the substitution $a_s\to -a_s$, $a_i\to -a_i$, $E_{0s}\to E_{0i}$. It follows from (23) that $\varphi_0(-\infty)$, $\varphi_0(\infty)=\pi$, $\theta=\pi$. Thus, the fields (23) are π pulses.

9. In cases in which one of the constants is smaller than unity in modulus and the other is not equal to unity, the arising soliton regimes correspond to turning angles of the polarization vector of the medium that are restricted to separated parts of the interval $(-\pi,\pi)$. For example, in the case $C_1=C_2=C$ $(0\le C<1)$, when $c_i<V<c_s$ is the case of normal dispersion, we have

$$\varphi_0(\xi)=-2\,\mathrm{arctg}\left[\frac{1}{a}\,\mathrm{th}\frac{\xi}{2\tau}\right],\quad |\varphi_0|\le 2\,\mathrm{arctg}\frac{1}{a},\quad a^2=\frac{1+C}{1-C}$$

$$E_{0s}^2=-\frac{a_s(1-C^2)}{C+\mathrm{ch}(\xi/\tau)},\quad E_{0i}^2=-\frac{a_i(1-C^2)}{C+\mathrm{ch}(\xi/\tau)};\quad \tau^{-1}=[b(1-C^2)]^{1/2}.$$

If, on the other hand, $c_s<V<c_i$, then

$$\varphi_0(\xi)=2\,\mathrm{arctg}\left[\frac{1}{a}\,\mathrm{cth}\left(\frac{1}{2}\left(\frac{\xi}{\tau}+\sigma\right)\right)\right],\quad \sigma=\ln\left|\frac{1+a}{1-a}\right|$$

$$E_{0s}^2=\frac{a_s(1-C^2)}{\mathrm{ch}(\xi/\tau+\sigma)-C},\quad E_{0i}^2=\frac{a_i(1-C^2)}{\mathrm{ch}(\xi/\tau+\sigma)-C}.$$

In the first case $\theta=4\arctan(1/a)$, in the second $\theta=2\pi-4\arctan(1/a)$. In the case $0\le C_1<1$, $C_2>1$, we have, at $V<c_i$, $V<c_s$,

$$\varphi_0(\xi)=2\,\mathrm{arctg}\left[\frac{a_1a_2}{(a_1^2+a_2^2)^{1/2}}\frac{\mathrm{sh}(\xi/\tau)}{\mathrm{dn}(\xi/\tau)}\right],\quad |\varphi_0|\le 2\,\mathrm{arctg}\,a_1;$$

at $c_s<V<c_i$,

$$\varphi_0(\xi)=2\,\mathrm{arctg}\left[\frac{a_1}{\mathrm{cn}(\xi/\tau)}\right],$$

$$a_1^2=\frac{1-C_1}{1+C_1},\quad a_2^2=\frac{C_2-1}{C_2+1};\quad \tau^{-1}=[2|b|(C_2-C_1)]^{1/2}.$$

The solitons represent an infinite periodic train of the elliptic type with period $4\tau K(a_1/\sqrt{a_1^2+a_2^2})$ and areas $\theta_T=4\arctan a_1$ in the case of arbitrary dispersion and $\theta_T=2\pi-4\arctan a_1$ in the case of anomalous dispersion.

We note that the analysis given here can be extended to the case of two-phonon resonance absorption of the pulses with unequal frequencies. For this process, the virtual level is found between the working levels of the transition of the molecule, i.e., $\omega_i+\omega_s=\omega_v$. It is not difficult to see that the description of the two-photon absorption can be obtained formally by replacing ω_s by $-\omega_s$ in (2) and (5). Thus, all the obtained classes of soliton solutions also take place in the case of two-photon absorption with the obvious change in the conditions on the dispersive properties of the medium. We shall also show that the subsequent investigation of the stability is applicable also to the case of two-photon absorption.

The results obtained above were applied to the case of total absence of relaxation ($T_1=T_2=\infty$). We shall show that account of the finiteness of T_1 and T_2 leads, as in the case of the usual effect of self-induced transparency,[2] to a gradual small change in the soliton pulses in the process of their propagation. The considerations given below are valid both for solitary pulses and for each of the T_2/T periods of the train pulses.

With the aid of (2), it is easy to obtain the result that the amounts of energy of the pump pulses and the Stokes wave passing through a unit area of cross section,

$$\Omega_i=\frac{c\sqrt{\varepsilon(\omega_i)}}{8\pi}\int_{\delta_1}^{\delta_2}E_i^2(z,t)\,dt,\quad \Omega_s=\frac{c\sqrt{\varepsilon(\omega_s)}}{8\pi}\int_{\delta_1}^{\delta_2}E_s^2(z,t)\,dt$$

obey the equations

$$\frac{1}{\omega_i\varepsilon(\omega_i)}\frac{d\Omega_i}{dz}=-\frac{1}{\omega_s\varepsilon(\omega_s)}\frac{d\Omega_s}{dz}=-c^2\frac{\hbar\mu_0N_V}{8\pi}\int_{\delta_1}^{\delta_2}\frac{W(z,t)-W^{eq}}{T_1}\,dt,$$

where $\delta_1=-\infty$, $\delta_2=\infty$ in the case of solitary pulses, $\delta_1=-\delta_2=-T/2$ in the case of trains (T is the period of the train). These equations show that an additional transformation of the energy from one wave to another takes place as a result of the relaxation processes. We make use here of the relation

$$\int_{\delta_1}^{\delta_2}\left\{\frac{u^2(z,t)+v^2(z,t)}{T_2}+\frac{W(z,t)[W(z,t)-W^{eq}]}{T_1}\right\}dt=0,$$

which is easily obtained from (2) by considering the change in the length of the "Bloch" vector with components (u,v,W) because of the relaxation. Then

$$\frac{1}{\omega_i\varepsilon(\omega_i)}\frac{d\Omega_i}{dz}=-\frac{1}{\omega_s\varepsilon(\omega_s)}\frac{d\Omega_s}{dz}$$

$$=c^2\frac{\hbar\mu_0N_V}{8\pi W^{eq}}\int_{\delta_1}^{\delta_2}\left\{\frac{u^2(z,t)+v^2(z,t)}{T_2}+\frac{[W(z,t)-W^{eq}]^2}{T_1}\right\}dt.$$

This formula is valid for pulses of arbitrary shape (not necessarily solitons) and for any relation between their durations and relaxation times. We now turn to the solutions of soliton type and use the smallness of the ratios τ/T_1, τ/T_2, where the quantities τ corresponding to each type of solitons defined above have the meaning of a characteristic scale of their duration (in the case of trains, the same role is played by the ratios T/T_1, T/T_2). Then we have

$$\frac{1}{\omega_i\varepsilon(\omega_i)}\frac{d\Omega_i}{dz}=-\frac{1}{\omega_s\varepsilon(\omega_s)}\frac{d\Omega_s}{dz}=c^2\frac{\hbar\mu_0N_VW^{eq}}{4}\left(\frac{\tau}{T_1}+\frac{\tau}{T_2}\right).$$

The expression thus obtained shows that the rate of change of the energy of the solitons is a small quantity, $\sim\tau(T_1^{-1}+T_2^{-1})$, i.e., account of the small relaxation terms leads to a weak effect of them on the passage of the solitons (in this sense, the given problem does not differ from other problems of the theory of ultrashort pulses[9]). The relaxation effects lead to a gradual attenuation of the jump and a growth in the intensity of the soliton wave. This takes place both from the growth of the population of the lower level, with characteristic time T_1 and from the damping of the polarization of the medium with characteristic time T_2. The transformation of one wave into the other takes place much more slowly, however, than in the case of the exponential regime of nonstationary SRS.[6] It is easy to show that the critical length at which the relaxation effects disrupt the soliton regime is given by the expression

$$z_c \sim \frac{2a_i}{c\hbar\mu_0 N_\nu \omega_i \sqrt{\varepsilon(\omega_i)}\,|W^{eq}|}\left(\frac{1}{T_1}+\frac{1}{T_2}\right)^{-1}.$$

It should be noted that, in the absence of relaxation ($T_1 = T_2 = \infty$) $z_c \to \infty$. By virtue of the specifics of the combination interaction, the relaxation leads not to a gradual absorption of the solitons, as in the case of resonance approach, but to a gradual increase in one wave at the expense of the other. As in the case of the ordinary effect of self-induced transparency,[2] the effect of the relaxation processes can be compensated with the help of weak focusing of the beams in the medium.

Thus, the SRS solitons, as all other waves of such a type,[10] are intermediate self-similar asymptotic forms that exist until relaxation effects begin to appear. For a demonstration of their physical realizability, we must verify that the soliton waves are stable to small perturbations, the regions for which always exist experimentally (inhomogeneity of the composition of the medium, fluctuations of the thermodynamic quantities and so forth).

4. STABILITY OF THE SRS SOLITONS

We shall call the SRS solitons stable if small perturbations of the fields do not grow with time, thus violating their stationarity. Introducing the quantities

$$\varphi_{i,s}(z,t)=\frac{\lambda}{\hbar}\int E_{i,s}^2(z,t)\,dt,\quad \varphi(z,t)=\frac{\lambda}{\hbar}\int E_i(z,t)E_s(z,t)\,dt, \tag{24}$$

we get with the help of (3)

$$\begin{gathered}\frac{\partial\varphi_s}{\partial z}+\frac{1}{c_s}\frac{\partial\varphi_s}{\partial t}=2\varkappa_s(\cos\varphi-C_1),\\ \frac{\partial\varphi_i}{\partial z}+\frac{1}{c_i}\frac{\partial\varphi_i}{\partial t}=-2\varkappa_i(\cos\varphi-C_2),\quad \left(\frac{\partial\varphi}{\partial t}\right)^2=\frac{\partial\varphi_s}{\partial t}\frac{\partial\varphi_i}{\partial t}.\end{gathered} \tag{25}$$

The stationary pulses found in Sec. 3 correspond to stationary solutions of the set (25)

$$\varphi_{0s}=\frac{\lambda}{\hbar}\int_{\xi_0}^{\xi}E_{0s}^2(\xi')\,d\xi',\quad \varphi_{0i}=\frac{\lambda}{\hbar}\int_{\xi_0}^{\xi}E_{0i}^2(\xi')\,d\xi',$$
$$\varphi_0=\frac{\lambda}{\hbar}\int_{\xi_0}^{\xi}E_{0s}(\xi')E_{0i}(\xi')\,d\xi'.$$

The investigation of the stability of the solitons E_{0s}, E_{0i} is equivalent to the investigation of the stability of the quantities φ_0, φ_{0s}, φ_{0i}. We consider the behavior of the small perturbations near $\varphi_0(\xi)$, $\varphi_{0s}(\xi)$, $\varphi_{0i}(\xi)$:

$$\begin{gathered}\varphi_s(z,t)=\varphi_{0s}(\xi)+f_s(z,t),\quad \varphi_i(z,t)=\varphi_{0i}(\xi)+f_i(z,t),\\ \varphi(z,t)=\varphi_0(\xi)+f(z,t),\end{gathered} \tag{26}$$

where $|f_s| \ll |f_{0s}|$, $|f_i| \ll |\varphi_{0i}|$, $|f| \ll |\varphi_0|$. According to (25), the set of equations which describes the evolution of the small perturbations has the form

$$\begin{gathered}\frac{\partial f_s}{\partial z}+\frac{1}{c_s}\frac{\partial f_s}{\partial t}=-2\varkappa_s f\sin\varphi_0,\quad \frac{\partial f_i}{\partial z}+\frac{1}{c_i}\frac{\partial f_i}{\partial t}=2\varkappa_i f\sin\varphi_0,\\ \frac{\partial f}{\partial t}=\frac{1}{2}\frac{E_{0i}}{E_{0s}}\frac{\partial f_s}{\partial t}+\frac{1}{2}\frac{E_{0s}}{E_{0i}}\frac{\partial f_i}{\partial t}.\end{gathered} \tag{27}$$

In the general case, the study of such a system presents great difficulties. However, it is simplified for solitons of classes 1, 2, 9 ($C_1 = C_2 = C$), for which we can directly connect the perturbations f, f_s, f_i with the help of (24):

$$f=\frac{1}{2}\left(\frac{a_i}{a_s}f_s+\frac{a_s}{a_i}f_i\right).$$

Using this relation in place of the last equation in (27), and transforming to the variables $\xi = t - z/V$ and $p = t$, we obtain

$$\begin{gathered}\frac{\partial^2 f}{\partial\xi^2}+(\gamma_1+\gamma_2)\frac{\partial^2 f}{\partial\xi\,\partial p}+\gamma_1\gamma_2\frac{\partial^2 f}{\partial p^2}\\ -\sqrt{b}\left[\frac{\partial f}{\partial\xi}+\frac{1}{2}(\gamma_1+\gamma_2)\frac{\partial f}{\partial p}\right]\sin\varphi_0-f\sqrt{b}\frac{\partial\varphi_0}{\partial\xi}\cos\varphi_0=0,\end{gathered} \tag{28}$$

where $\gamma_1 = V/(V-c_s)$, $\gamma_2 = V/(V-c_i)$, $b = \lambda^2 a_i a_s/\hbar^2 > 0$.

1. We shall seek a solution of (28) in the form

$$f(\xi,p)=\sum_{l=0}^{\infty}f_l(\xi)\exp(\alpha_l p), \tag{29}$$

where $f_l(\xi)$ are assumed to be bounded functions in the case $|\xi| < \infty$, and

$$f(\xi,0)=\sum_{l=0}^{\infty}f_l(\xi)$$

gives the distribution of the perturbation at the initial instant of time $p = 0$. If the real part of one of the exponents α_l turns out to be positive, then, by virtue of (29), the soliton regime is unstable. If $\mathrm{Re}\,\alpha_l < 0$ for all l, then the soliton regime is asymptotically stable. In this case, the perturbation tends to damp out with increasing p, and the system is returned to the soliton regime. If $\mathrm{Re}\,\alpha_l \leq 0$, then the soliton regime is stable, but it does not possess asymptotic stability, since the system does not return to its initial state and is close to it under the action of a small perturbation, even in the case $p \to \infty$. Substituting (29) in (28) and introducing the new function $K_l(\xi)$ according to

$$f_l(\xi)=K_l(\xi)\exp\left\{-{}^1\!/_2\int[\alpha_l(\gamma_1+\gamma_2)-\sqrt{b}\sin\varphi_0]\,d\xi\right\},$$

we obtain the "Schrödinger equation"

$$\begin{gathered}-\frac{d^2K_l}{d\xi^2}+\left[\frac{1}{4}b\sin^2\varphi_0+\frac{1}{2}\sqrt{b}\frac{d\varphi_0}{d\xi}\cos\varphi_0-\lambda_l\right]K_l=0,\\ \lambda_l=-{}^1\!/_4(\gamma_1-\gamma_2)^2\alpha_l^2,\end{gathered} \tag{30}$$

with the "potential"

$$U(\xi)=\frac{1}{4}b\sin\varphi_0+\frac{1}{2}\sqrt{b}\frac{d\varphi_0}{d\xi}\cos\varphi_0$$

with "energy" eigenvalues λ_l. The spectrum of λ_l of Eq. (30) is bounded from below, beginning with some λ_0 corresponding to the ground state. According to the properties of the potential $U(\xi)$, the initial part of the spectrum can be discrete. We number the λ_l in the discrete portion in increasing order of $\lambda_0 < \lambda_1 < \lambda_2 < \ldots$. Then the index of the given λ_l will simultaneously give the number of zeroes of the corresponding eigenfunction K_l

of Eq. (30). The eigenfunction of the ground state K_0 should not have zeroes. We shall show that in the considered case the ground state corresponds to $\lambda_0 = 0$. The bounded solution of Eq. (30) with $\lambda_0 = 0$ is of the form

$$K_0(\xi) = \exp\left\{\frac{1}{2}\sqrt{b}\int \sin\varphi_0\, d\xi\right\}.$$

Using (8), we easily obtain the result

$$K_0 = (D\, d\varphi_0/d\xi)^{1/2}, \quad D = \text{const}. \tag{31}$$

Substituting the specific expressions $\varphi_0(\xi)$ for class 1 (see (10), (12)), class 2 (see (13)) and class 9, it is not difficult to establish the fact that the derivative $d\varphi_0/d\xi$ never vanishes. Consequently, $\lambda_0 = 0$ corresponds to the ground state, and all the remaining eigenvalues are positive, i.e., $\lambda_l > 0$, $l = 1, 2, \ldots$. Now, taking into account the connection between λ_l and α_l, we finally obtain the result that the indices α_l in the case $l \geq 1$ are purely imaginary, and $\alpha_0 = 0$. Thus, Eq. (28) for the perturbation of the overlap integral does not have exponentially increasing solutions, i.e., solitons of classes 1, 2, and 9 are stable, although they do not possess asymptotic stability.

2. The absence of asymptotic stability can demonstrate the existence of weakly increasing perturbations. Therefore, it is important to investigate the effect of such a type of perturbation on the soliton regime. We shall show that there exists a perturbation that is linearly increasing with the time p, of the form

$$f(\xi, p) = A(\xi) + pB(\xi). \tag{32}$$

Substituting (32) in (28), we obtain

$$\frac{d^2A}{d\xi^2} - \sqrt{b}\frac{dA}{d\xi}\sin\varphi_0 - \sqrt{b}A\frac{d\varphi_0}{d\xi}\cos\varphi_0 + (\gamma_1+\gamma_2)\frac{dB}{d\xi} - \frac{1}{2}\sqrt{b}(\gamma_1+\gamma_2)B\sin\varphi_0 = 0, \tag{33a}$$

$$\frac{d^2B}{d\xi^2} - \sqrt{b}\frac{dB}{d\xi}\sin\varphi_0 - \sqrt{b}B\frac{d\varphi_0}{d\xi}\cos\varphi_0 = 0. \tag{33b}$$

Differentiating Eq. (8) twice with respect to ξ and equating the resultant expression to (33b), we find that the bounded solution (33b) has the form $B(\xi) = d\varphi_0/d\xi$. It is not difficult, by direct substitution in (33a) to show that

$$A(\xi) = B(\xi)\left[-{}^1/{}_2(\gamma_1+\gamma_2)\xi\right].$$

Thus, the perturbation that is increasing linearly with time has the form

$$f(\xi, p) = \left[p - \frac{1}{2}(\gamma_1+\gamma_2)\xi\right]\frac{d\varphi_0}{d\xi}. \tag{34}$$

The result obtained can be interpreted formally as the presence of an instability of solitons relative to the perturbation (32). We shall prove, however, by using the approach applied in Ref. 11 in the case of a study of the stability of solitons in a medium with single-photon absorption, that such perturbations do not destroy the SRS solitons. For this purpose, we write down the expression (26) for the perturbed overlap integral $\varphi(z,t)$ near the soliton solution:

$$\varphi(z, t) = \varphi_0(\xi) + \nu[p - \delta\xi]\frac{d\varphi_0}{d\xi}, \tag{35}$$

where the small amplitude of the perturbation ν is introduced, and $\delta = \frac{1}{2}(\gamma_1 + \gamma_2)$. The expression (35) represents the expansion in a series in the linear approximation in ν. Therefore, we have, with the same accuracy,

$$\varphi(z, t) = \varphi_0[(\xi + \nu(p - \delta\xi))/\tau], \tag{36}$$

where the duration τ for the unperturbed pulses has been introduced (or the period in the case of a train):

$$\tau^{-1} = \begin{cases} \sqrt{b}, & C = \pm 1 \\ \sqrt{b(C^2-1)}, & C > 1, \quad C < -1. \\ \sqrt{b(1-C^2)}, & -1 < C < 1 \end{cases}$$

The possibility of the representation of (35) in the form (36) shows that the perturbations of the field again represent solitons, but now with a different velocity and duration relative to the initial values. It is not difficult to establish this by transforming the argument in (36) to the form $\varphi_0(z,t) = \varphi_0[(t - x/V')/\tau']$, where τ' is the duration and V' the velocity of the perturbed pulses:

$$\tau'^{-1} = \tau^{-1}[1 + \nu(1-\delta)], \quad V' = V[1 + \nu/(1 - \delta\nu)].$$

It can be shown that in the first order of smallness in ν the quantities τ' and V' are connected with one another by the same relations as the quantities τ and V:

$$\tau'^{-1} = \begin{cases} \sqrt{b'}, & C = \pm 1 \\ \sqrt{b'(C^2-1)}, & C > 1, \quad C < -1, \\ \sqrt{b'(1-C^2)}, & -1 < C < 1 \end{cases}$$

where $b' = 4\varkappa_l\varkappa_s c_l c_s V'^2/(V' - c_s)(V' - c_l)$. This result means that the perturbed solitons relate to the same type of solitary pulses or trains as the unperturbed. Consequently, the perturbations (32) although they increase with time, do not lead to a destruction of the solitons, but only to a change in their length and their velocity, keeping in this case the shape of the pulse envelope. Under the action of the perturbations (32), one 2π-pulse transforms in continuous fashion into another, while in the case considered in Item 1 of Sec. 4, no return to the initial regime takes place.

5. CONCLUSION

The results show that, in addition to the well-studied quasistatic and nonstationary regimes of SRS, specific soliton regimes of scattering are generated in strong fields of ultrashort pulses. They are not accompanied by amplification. Depending on the initial conditions and the dispersion characteristics of the scattering medium, soliton regimes of different classes can exist. In particular, in the region of normal dispersion, for an initially equilibrium medium, SRS solitons can represent only periodic trains of 2π pulses. Solitons in the form of isolated 2π pulses of finite duration at these same initial conditions can be realized only in the region of anomalous dispersion. Similar soliton regimes in the region of normal dispersion are realized only in the case of inversion of the initial population of the levels of the working transition, and represent solitons of Lorentzian shape or 2π pulses of hyperbolic type. Under arbitrary dispersion conditions, soliton regimes are also possible in the form of periodic trains of 2π pulses of elliptical and trigonometric types. The solitons of Lorentzian shape, trains of a trigonometric type and special types of isolated solitons with $\theta < 2\pi$ (class 9) are stable, which indicates the possibility of their practical realization (see the estimates in Sec. 1).

We note in conclusion that the soliton regimes of SRS can have great value in the analysis of the detailed temporal structure of the radiation of combination lasers, since the fields arising in such systems have intense fluctuation discharges of short duration. The observation of soliton regimes in "pure form" is advantageously carried out in gases at low pressure ~ 0.1 atm in the case of durations of the initial laser pulses of ~ 1–10 nanosec.

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[1)]We note that here and below we neglect the pumping of energy into the antistokes and higher Stokes components, since the intense fields of the exciting and first Stokes radiations are given at the input to the medium, and the fields of the other components are generated from noise nuclei.

[2)]We note that for the special case of the absence of dispersion $\eta_i = \eta_s$, this type of soliton solutions was found also in a recent publication.[8]

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Crossing of quasistationary levels[1)]

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The crossing of discrete energy levels, each of which interacts with the continuous spectrum, is discussed. The problem is reduced to the consideration of only two, but quasistationary, levels with suitably modified interaction. A formula for the amplitude of the nonadiabatic transition in this problem is derived for a sufficiently general dependence of the terms on the interatomic distance. The behavior of the populations of such states is investigated, and it is shown that the interaction between the levels through the continuum has an important effect both on the nonadiabatic transition amplitude and on the population of states. It is noted that both the formulation of the problem and the method of solution given by Karas' *et al.* (1974) and by Bazylev and Zhevago (1975) are subject to error.

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1. It is necessary to introduce the concept of quasistationary energy terms when different atomic-collision processes are investigated. In contrast to the usual discrete states, quasistationary terms are characterized not only by the energy E but also by the width Γ which describes the possibility of decay, i. e., the possibility of a transition of the system from a given electronic state to the continuum with the emission of an electron or photon. In general, both E and Γ are functions of the distance between the colliding particles. The interaction and crossing of such terms play a fundamental role in collisions leading to the formation of vacancies in the inner electron shells of atoms.[2,3] Such states are commonly referred to as the autoionization states. Another example is charge transfer on negative ions. Thus, analysis of experimental data shows[4] that the crossing of quasistationary molecular terms corresponding to different charge-transfer channels must be taken into account in these reactions. Multiple crossing of quasistationary terms is also expected in collision processes involving the participation of atoms in highly excited states, for example, in Penning ionization processes of