Localized Dissipative Unipolar Objects under the Condition of Stimulated Raman Scattering

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The possibility of the formation of dissipative unipolar soliton pulses in an amplifying medium of Raman-active molecules has been analyzed. It has been shown that the formation of such pulses is possible under the mutual compensation of Raman enhancement and irreversible losses caused by fast relaxation in the system of electronic optical transitions. Since Raman enhancement is nonlinear, the threshold duration and energy of a soliton-like object being formed are determined by the parameters of the medium.

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1. INTRODUCTION

Dissipative optical solitons now attract great attention in studies of nonlinear processes [1–10]. They are of both fundamental and applied interest. The mutual compensation of the energy entering a nonlinear medium and irreversible energy losses can result in the formation of dissipative solitons under certain conditions. The energy can be fed by various methods, e.g., from a continuous external source or an external pulse source, which can excite the nonlinear medium to a nonequilibrium state. In the latter case, the stored energy can further ensure its supply for the formation of dissipative solitons. If the duration \( \tau_p \) of a dissipative soliton and the observation time \( \Delta t \) exceed the characteristic relaxation times \( T_2 \) and \( T_1 \) of the dipole moment and populations of stationary states, the nonequilibrium medium after the passage of the soliton becomes thermodynamically equilibrium.

The condition \( T_2 \ll T_1 \) is usually valid in solids. The ratio \( T_2/T_1 \) can vary from \( 10^{-2} \) to \( 10^{-5} \) [11]. Consequently, it is possible to ensure the interesting condition

\[
T_2 < \tau_p \ll \Delta t \ll T_1. \tag{1}
\]

Under this condition, the pulse entering the medium can induce the transition of the initially nonequilibrium medium to another nonequilibrium metastable state. This process is accompanied by the formation of localized soliton-like objects [9, 12, 13]. These objects were called incoherent solitons in [14, 15] and soliton-like structures and soliton-like objects in nonequilibrium dissipative media in [12, 13]. These localized objects are short-lived: their lifetime is always shorter than the relaxation time \( T_1 \) according to condition (1). Therefore, these objects can be observed in media and at quantum transitions where the relaxation time \( T_1 \) is long. The relaxation time \( T_1 \) at low temperatures is: \( T_1 \sim \omega_{tr}^{-3} \), where \( \omega_{tr} \) is the transition frequency. Hence, it is necessary to use quantum transitions with low frequencies \( \omega_{tr} \), e.g., electron vibrational (Raman) transitions corresponding to normal vibrational modes of molecules. The relaxation time \( T_1 \) for these transitions is fairly long. In particular, the relaxation time of the populations of Raman sublevels in liquid nitrogen is 56 s [16]. This giant \( T_1 \) value with a great margin satisfies inequalities (1). It is also important that Raman transitions are forbidden in the electron dipole approximation and are fundamentally two-photon.

When the spectrum of an optical pulse covers the forbidden electron vibrational transition at the frequency \( \omega_{tr} \), stimulated Raman self-scattering occurs [17, 18]. In this case, the carrier frequency of the pulse is continuously redshifted in proportion to the traveled distance. The authors of [19, 20] showed that this mechanism at sufficiently long distances can generate a single-cycle or even unipolar signal.

The nonlinear optics of unipolar signals in the 1970s was exclusively of theoretical interest [21, 22]. Many theoretical studies performed in this field from 1988 to 1995 and in the 2000s, (see, e.g., [23–31]) were stimulated by experimental achievements in the generation of nearly single-cycle pulses [32–34].
The nonlinear optics of unipolar pulses is currently under rapid development [35–45]. In particular, the propagation of both conservative and dissipative solitons is studied.

Stimulated Raman self-scattering processes, which promote the formation of unipolar signals, can be accompanied by irreversible losses of the pulse energy at other, e.g., electron optical, quantum transitions. This can lead to the formation of localized unipolar soliton-like objects at the initial nonequilibrium populations of vibrational sublevels of molecules under the dominance of stimulated Raman scattering processes. This work is devoted to the study of this problem.

2. BASIC EQUATIONS

The electric field $E$ of an unipolar pulse with the duration $\tau_p$ propagating in the $z$ direction in an isotropic insulator containing Raman-active molecules satisfies the wave equation

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} (P_e + P_R).$$

(2)

Here, $c$ is the speed of light in vacuum and $P_e$ and $P_R$ are the polarization electron optical and Raman responses, respectively.

The duration $\tau_p \approx 10^{-13}$ s certainly satisfies the inequality

$$\delta_1 = (\omega_0 \tau_p)^{-1} \ll 1,$$

(3)

where $\omega_0 \approx 10^{15}$ s$^{-1}$ is the characteristic frequency of electron optical transitions.

If the left inequality in Eq. (1) is satisfied, the polarization response of electron optical transition can be treated as linear in the electric field strength of the pulse and [13]

$$P_e = \chi E - \eta \frac{\partial E}{\partial t}.$$

(4)

Here, $\chi$ and $\eta$ are the noninertial and inertial components of the electric susceptibility of the medium, the latter being due to the phase relaxation of electron optical transitions. They can be estimated as [12, 13] $\chi \sim 2d^2 n/\hbar \omega_0$ and $\eta \sim 8d^2 n/(\hbar T_0 \omega_0^3) \sim \chi/(T_0 \omega_0^2)$, where $d$ is the characteristic dipole moment of allowed transitions, $n$ is the concentration of molecules responsible for the linear susceptibility $\chi$, and $\hbar$ is the reduced Planck constant.

Under the described conditions, the populations of quantum levels involved in electron optical transitions hardly change and correspond to thermodynamic equilibrium of the electron optical subsystem.

The Raman polarization response is given by the standard expression [46]

$$P_R = n_R \alpha' q E,$$

(5)

where $\alpha' = (\partial \alpha/\partial q)_{q=0}$, $\alpha$ is the electric polarizability of a Raman-active molecule, $q$ is the displacement of atoms in the molecule from the equilibrium position, and $n_R$ is the concentration of Raman-active molecules.

The dynamic parameters of a Raman transition at the frequency $\omega_0$ satisfy the well-known equations [46]

$$\frac{\partial^2 q}{\partial t^2} + \omega_0^2 q = \frac{\alpha'}{2M} E^2 w, \quad \frac{\partial w}{\partial t} = \frac{\alpha'}{\hbar \omega_0} E^2 \frac{\partial q}{\partial t},$$

(6)

where $w$ is the population difference between the excited and ground Raman sublevels ($w = 1$ and $-1$ if the excited and ground sublevels are populated, respectively), $M$ is the reduced mass of the molecule, and the relaxation terms are neglected because the duration of the pulse is much shorter than the relaxation times $\tau_{1R}$ and $\tau_{2R}$ for the Raman transition. It is noteworthy that the ground Raman sublevel coincides with the ground electron level in the molecule.

The frequency $\omega_0 \sim 10^{13}$ s$^{-1}$ and the characteristic durations $\tau_p$ taken above satisfy the inequality

$$\delta_2 = (\omega_0 \tau_p)^2 \ll 1.$$

(7)

The spectral width of the unipolar pulse is $\delta \omega \sim 1/\tau_p$. Consequently, the inequality (7) should be considered as the condition mentioned above that the spectrum of the signal includes the Raman frequency. As a result, this inequality ensures the most favorable conditions for stimulated Raman self-scattering.

The second term on the left-hand side of the first equation in Eqs. (6) can be neglected under the condition (7). In this case, the solution of system (5) has the form [19, 20]

$$\frac{\partial q}{\partial t} = -\text{sgn}(\alpha') w_{\infty} \frac{\hbar \omega_0}{\sqrt{2M}} \sin \theta, \quad w = w_{\infty} \cos \theta,$$

(8)

where

$$\theta = \kappa \int_{-\infty}^{t} E^2 dt,$$

(9)

where $w_{\infty}$ is the initial (at $t = -\infty$) population difference between Raman sublevels, and $\kappa = \frac{|\alpha'|}{\sqrt{2M \hbar \omega_0}}$.

Under the condition (7), the displacement $q$ is negligibly small; i.e., $q = 0$ can be set with a high accuracy [19]. Then, according to Eqs. (5) and (8),

$$\frac{\partial P_R}{\partial t} \approx n_R \alpha' \frac{\partial q}{\partial t} E = -w_{\infty} n_R |\alpha'| \frac{\hbar \omega_0}{\sqrt{2M}} E \sin \theta.$$

(10)
The substitution of Eqs. (4) and (10) into Eq. (2) yields the equation
\[
\frac{\partial^2 E}{\partial z^2} - \frac{n_0^2 \partial^2 E}{c^2 \partial t^2} = -4\pi \frac{\partial}{c \partial t} \left( \eta \frac{\partial^2 E}{\partial t^2} + w_{\text{inc}} |\alpha'| \frac{\hbar \omega}{2M} \sin \theta \right),
\]
(11)
where  is the noninertial part of the refractive index of the medium.

The two terms on the right-hand side of Eq. (11) are proportional to small parameters  and . Under these conditions, the unidirectional propagation approximation for the pulse along the  axis at a velocity close to  is applicable [22, 24, 25, 27, 30]. In this approximation, Eq. (11) takes the form
\[
\frac{\partial E}{\partial z} + \frac{n_0 \partial E}{c \partial t} = \mu E \sin \theta + D \frac{\partial^2 E}{\partial t^2},
\]
(12)
where
\[
D = \frac{2\pi}{\eta n_0}, \quad \mu = \frac{2\pi}{\eta n_0} w_{\text{inc}} |\alpha'| \frac{\hbar \omega}{2M}.
\]

The last, diffusion, term on the right-hand side of Eq. (12) describes irreversible losses caused by phase relaxation on electron optical transitions. The first term describes a nonlocal nonlinear source caused by stimulated Raman self-scattering at the inverted initial populations  of Raman quantum sublevels.

Because of this term, Eq. (12) does not meet the general electric area conservation rule:  = const [47]. However, this nonlinear diffusion term is relatively small in the approximation (7). Thus, the violation of the  rule is a consequence of the taken approximation and does not mean a physical contradiction.

Multiplying Eq. (12) by , using Eq. (9), and integrating with respect to , we obtain
\[
\frac{\partial \theta}{\partial z} + \frac{n_0 \partial \theta}{c \partial t} = 4\mu \sin^2 \theta \frac{\partial^2 \theta}{2 \partial t^2} + D \frac{\partial^2 \theta}{\partial t^2} - 2D \int_{-\infty}^{t} \left( \frac{\partial \theta}{\partial t'} \sqrt{\frac{\partial \theta}{\partial t'}} \right)^2 dt'.
\]
(14)

Nonlinear integro-differential equations (12) and (14) describe the propagation of the pulse in the medium of Raman-active molecules in the presence of irreversible losses to electron optical quantum transitions.

3. SOLITON SOLUTION AND ITS ANALYSIS

An exact nontrivial analytical solution of Eq. (14), as well as Eq. (12), can hardly be found. For this reason, an approximate soliton-like solution is sought. To this end, it is desirable to approximate  in Eq. (14) by an appropriate polynomial. A Taylor series near  is inappropriate for this purpose because the angular range  is of interest, as will be shown below. In this range, the approximation  can be used with the constants  and  to be determined from the two conditions: (i) the polynomial reaches a maximum at the point  and (ii) this maximum is equal to unity.

As a result,  and . The resulting approximation in the angular range  has the form
\[
\sin^2 \frac{\theta}{2} = F(\theta) = \frac{3}{\pi^2} \theta^2 - \frac{2}{\pi^3} \theta^3.
\]
(15)

It is seen in Fig. 1 that the approximation (15) in the angular range  is quite satisfactory.

The substitution of Eq. (15) into Eq. (14) yields the equation
\[
\frac{\partial \theta}{\partial z} + \frac{n_0 \partial \theta}{c \partial t} = a \theta^2 - b \theta^3 + D \frac{\partial^2 \theta}{\partial t^2} - 2D \int_{-\infty}^{t} \left( \frac{\partial \theta}{\partial t'} \sqrt{\frac{\partial \theta}{\partial t'}} \right)^2 dt',
\]
(16)
where  and .

In the absence of the last, integral, term, Eq. (16) is a reaction–diffusion equation. However, this term is important and cannot be omitted.

With the method considered in [9], the stationary solution of Eq. (16) can be found in the form of a propagating pulse
\[
\theta = \frac{A_0}{2} (1 + \tanh \xi),
\]
(17)
where  and  for the soliton profile.

Fig. 1. (Solid line)  and (dashed line)  curves given by Eq. (15) in the angular interval  0 ≤ θ ≤ 4π/3.
In Eqs. (17)–(20), the above expressions for \( a \) and \( b \) are used.

The electric field of the unipolar soliton-like pulse is expressed from Eqs. (17), (18), and (9) in the form

\[
E = \pm \frac{2\pi}{\sqrt{3\kappa \tau_p}} \text{sech} \xi,
\]

Here, the constants \( \tau_p \) and \( v \) are the duration and velocity of the pulse, respectively.

The substitution of Eq. (21) into the expression \( I = cE^2/(4\pi n_0) \) for the signal intensity gives

\[
I = I_m \text{sech}^2 \xi,
\]

where

\[
I_m = \frac{c}{6\tau_p \kappa \xi}.
\]

From Eqs. (8), (17), and (18), we obtain the expressions for the translational velocity \( V \) of atoms in molecules and the population difference between the Raman sublevels in the form

\[
V = \frac{\partial q}{\partial t} = -\text{sgn}(\alpha') V_m \sin \left[ \frac{2\pi}{3} \left( 1 + \tanh \xi \right) \right],
\]

\[
w = w_\infty \cos \left[ \frac{2\pi}{3} \left( 1 + \tanh \xi \right) \right],
\]

where \( V_m = w_\infty \sqrt{\hbar \alpha_x / 2M} \).

Figure 2 shows (a) the profile of the intensity (22) of the field of the unipolar pulse, as well as (b) the profile of the translational velocity of atoms (24) and (c) the profile of the population difference between the Raman sublevels (25), which are induced by this pulse.

A horizontal plateau appears in the velocity profile immediately after the passage of the pulse; it is easily explained in terms of the first equation of the system (6). Indeed, the term \( \omega_q^2 q \) (restoring force) is neglected in this equation under the condition (7).

Then, this equation has the form \( \partial^2 q/\partial t^2 = 0 \), which gives the mentioned plateau \( V = \text{const} \). The lifetime of the plateau can be estimated as \( \omega_q^{-1} \). The term \( \omega_q^2 q \) in the first equation of the system (6) becomes significant beginning with times \( \sim \omega_q^{-1} \) after the passage of the pulse; this term is responsible for free oscillations corresponding to optical molecular modes.

In the absence of irreversible losses (for conservative solitons), the population difference \( w \) after the passage of the pulse returns to its initial value \( w_\infty \) [25, 30]. As seen in Fig. 2, under the effect of irreversible phase relaxation in the system of electron optical transitions, the population difference \( w \) no longer returns to the initial value. In the central part of the pulse, Raman-excited molecules transit to the ground state. Immediately after the passage of the pulse in the medium, only about one-fourth of molecules on
average return to the excited vibrational state. As a
result, the average population difference becomes
\( w_{+0} = -0.5w_{0-} \), which corresponds to another, meta-
stable, state of molecules with the lifetime \( \sim T_{1R} \). Thus,
the pulse, transferring the most part of nonequilib-
rium Raman-active molecules to the ground Raman
sublevel takes the energy \( 0.5\hbar \omega \eta_R (w_{0-} - w_{0+}) = 0.75\hbar \omega \eta_R w_{0-} \) from the unit volume of the medium.
This energy income is compensated by losses caused
by irreversible phase relaxation in the equilibrium sys-
tem of electron optical transitions.

The energy of the pulse proportional to

\[
A \equiv \theta_{t \rightarrow +\infty} = \kappa \int_{-\infty}^{+\infty} E^2 dt
\]

satisfies the equation (see Eq. (14) at \( t \rightarrow +\infty \))

\[
\frac{dA}{dz} = 4\mu \sin^2 \frac{A}{2} - 2D \int_{-\infty}^{+\infty} \left( \frac{\partial \theta}{\partial t} \right)^2 dt.
\]

Here, the first/second term corresponds to the energy
supply to/loss from the pulse. The soliton-like object
described above is due to the balance of these two pro-
cesses.

The further analysis is performed with the approx-
imation given by Eq. (15) and under the assumption
that \( \theta \) is given by Eq. (17) with the substitution
\( A_0 \rightarrow A(z), \xi \rightarrow \xi = \frac{t - f(z)}{\tau_0} \), where \( f(z) \) is a certain
function and \( \tau_0 \) is the duration of the pulse generally
different from \( \tau_p \) (see Eq. (19)). Summarizing,
Eq. (27) is reduced to

\[
\frac{dA}{dz} = \frac{8\mu}{\pi^2} Q(A),
\]

where

\[
Q(A) = -\frac{9\pi^2 \tau_p^2 A}{16\tau_0^2} + \frac{3\pi}{2} A^2 - A^3,
\]

\[
\tau_c = \frac{2}{3 \sqrt{\frac{\pi D}{3\mu}}}
\]

Figure 3 shows the plots of \( Q(A) \) at \( \tau_0 > \tau_c \) and \( \tau_0 < \tau_c \). In the former case, the point \( A = A_2 \) is an attractor
if \( A > A_1 \) at the entrance to the medium. Here,

\[
A_{1,2} = \frac{3\pi}{4} \left( 1 \mp \sqrt{1 - \frac{\tau_c^2}{\tau_0^2}} \right).
\]

At \( A < A_1 \), the point \( A = 0 \) is an attractor.

In the latter case \( \tau_0 < \tau_c \), the point \( A = 0 \) is the
only attractor. Thus, the found dissipative unipolar
soliton-like object can be formed under two threshold
conditions

\[
\tau_0 > \tau_c, \quad A > A_1,
\]

These conditions are consistent with Eqs. (19) and
(18), respectively, because \( \tau_0 > \tau_c \) and \( A_\infty > A_1 \).

The substitution of \( \tau_0 = \tau_p \) and Eq. (19) into
Eq. (31) gives \( A_1 = \pi/6 \) and \( A_2 = 4\pi/3 \). Consequent-
ly, the point \( A_\infty \) given by Eq. (18) is an attractor
in this case at \( A > A_1 \). This property is important evidence
of the stability of the dissipative unipolar object
under consideration.

Thus, the unipolar pulse satisfying the condi-
tions (32) should be formed at the entrance to the
Raman-active medium. Then, it can be transformed
in the medium to the pulse with the duration, electric
field strength, and intensities given by Eqs. (19)–(21),
respectively. Numerous methods to generate unipolar
electromagnetic signals are known (see, e.g., reviews
[45, 48]). In our case, a significant part of the spec-
trum of the pulse with the duration \( \tau_p \sim 10^{-13} \) s lies in
the terahertz range. Consequently, this pulse can be
referred to as a terahertz unipolar pulse. Such a pulse
can be generated, e.g., by splitting of a bipolar tera-
hertz signal into two unipolar pulses with opposite
polarities [49]. One of these pulses under the condi-
tions (32) can be used as an initial pulse at the entrance
to the Raman-active medium.

Some numerical estimates are given below. For media
with fast phase relaxation with the typical parameters
[50] \( T_2 \sim 10^{-13} \) s, \( \omega_0 \sim 10^{15} \) s\(^{-1}\), and \( \chi \sim 0.1 \), we
obtain \( \eta \sim \chi/(T_2\omega_0^3) \sim 10^{-18} \) s. Then, \( D = 2\pi\eta/cn_0 \sim 10^{-27} \) s\(^2\)/cm. Taking in addition \( |\alpha'| \sim 10^{-15} \) cm\(^2\), \( \omega_0 \sim 10^{12} \) s\(^{-1}\), \( n_R \sim 10^{21} \) cm\(^{-3}\), and \( M \sim 10^{-22} \) g [20],
we find \( \mu = (2\pi/c)n_0|\alpha'\sqrt{\hbar/\omega_0}/M \sim 0.1 \) cm\(^{-1}\). As a
result, the duration of the dissipative soliton-like
object is \( \tau_p \sim \sqrt{D/\mu} \sim 10^{-13} \) s, as expected above. The
duration $\tau_p$ should be several times longer than the relaxation time $T_2$ (see Eq. (1)). Note also that this duration of the pulse is much shorter than the phase relaxation time $T_{\text{fr}} \sim 10^{-8}$ s [46] on the Raman transition in agreement with expectation.

The characteristic length of the pulse with the found duration $\tau_p$ in the propagation direction is $l_\parallel \sim c\tau_p \sim 10^{-3}$ cm. The relative difference of the velocity of the pulse from $c/n_0$ can be estimated as $|c/n_0 - 1| \sim (c/n_0)\sqrt{\mu D} \sim 10^{-4}$ (see Eq. (20)). Thus, the velocity $v$ of the soliton-like pulse only slightly differs from the linear velocity $c/n_0$. With the above parameters, we have $\kappa = |\alpha'|/\sqrt{2M\hbar\omega} \sim 10^4$ cm$^3$/erg s. Then, Eq. (23) gives the maximum intensity $I_m \sim 10^{12}$ W/cm$^2$. Such high intensities are quite achievable in real experiments. The power of the pulse with the aperture $d_p \sim 1$ mm is $N \sim I_m d_p^2 \sim 10^5$ W, and its energy is $W \sim N\tau_p \sim 1$ mJ.

The above parameters satisfy the inequality $l_\parallel \gg d_p$. The diffraction length under the considered conditions is estimated as $l_d \sim d_p^2/l_\parallel \sim 10$ cm. The one-dimensional approximation under consideration can be used at these distances. Then, the observation time of the process under study is estimated as $\Delta t \sim l_d/c \sim 10^{-10}$ s. This estimate, as well as the estimates presented above for $\tau_p$ and $T_2$, certainly satisfies the condition (1).

4. CONCLUSIONS

To summarize, it has been shown that dissipative unipolar soliton pulses can be formed at the inverted initial populations of Raman sublevels. It is important that irreversible losses in the system of Raman sublevels are negligibly low at the observed observation time. These losses caused by phase relaxation are significant on other quantum transitions, e.g., electron optical transitions with the equilibrium populations of quantum states. The mutual compensation of these losses and the energy transferred from the nonequilibrium Raman subsystem makes it possible to form soliton-like pulses.

Since Raman enhancement is nonlinear, the dissipative soliton-like object is formed under the threshold conditions (32). At the same time, the initial state of the medium and its final metastable state to which it transits after the passage of the pulse are relatively stable because the expected observation time of the process of propagation of pulses is much shorter than the irreversible relaxation times in the system of Raman sublevels.

In agreement with expectation, such parameters of the considered dissipative object such as the amplitude, duration, and velocity are unambiguously determined by the characteristics of the medium. At the same time, these parameters are independent of the characteristics of the pulse at the entrance to the medium. This property is common for dissipative solitons and is confirmed by the fact that the pulse seemingly “forgets” its parameters at the entrance to the medium because of irreversible losses. This situation is similar to the limit cycle in the theory of self-oscillations.

The approximate solution Eq. (14) has been obtained in the approximation (15), which is satisfied with a high accuracy in the interval $0 < \theta < 4\pi/3$. In this case, $A_\omega = 4\pi/3$. As a result, other solutions for which $\theta$ values leave this interval most probably remain unfound. The amplitude, duration, and velocity of such pulses will likely have another set of values. In any case, this set should be fixed for a soliton-like pulse with a certain parameter $A_\omega$. This is consistent with the property of the parameters of dissipative solitons, in contrast to conservative solitons, to take discrete sets of values [1]. I am going to study this issue separately in application to solutions of Eq. (14) (see also Eq. (12)).

CONFLICT OF INTEREST

The author declares that he has no conflicts of interest.

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