Spatiotemporal dynamics of electromagnetic pulses in saturating nonlinear optical media with normal group velocity dispersion

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The nonlinear dynamics of ultrashort optical pulses in nonlinear saturating media with normal group velocity dispersion is examined. The studied saturating nonlinearity changes the sign at the peak intensity of the laser pulses. In the bulk media and the planar wave guides the temporal collapse of the pulse is arrested by its splitting in spatial domain leading to rings formation. The wave collapse in one dimensional geometry cannot be arrested; the field singularity develops for a finite propagation distance. [S1063-651X(99)10812-2]

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Self-focusing and self-guiding of light beams have received much attention in recent years in connection with their important applications, such as soliton propagation, all-optical switching, and logic [1]. The stable spatial solitons with two transverse dimensions can exist in materials characterized by saturable nonlinearity that exactly compensates the diffraction. The requirement to have a high-power beam implies the use of lasers in pulsed regime. Recent achievements in short-pulse generation have motivated the studies of short-pulse propagation in nonlinear media. In such a case the pulse dispersion plays an important role. The spatiotemporal solitons called light bullets can be generated whenever a saturating nonlinearity compensates the anomalous group-velocity dispersion together with the diffraction [2]. However, most transparent bulk materials exhibit normal group-velocity dispersion (NGVD) that prevents solitons formation. The spatiotemporal dynamics of a light pulse in NGVD case has been comprehensively treated for Kerr media [3]. Analyzing three-dimensional nonlinear Schrödinger equation it appears that NGVD prevents collapse, by splitting the pulse in time domain into several smaller-scale structures.

In this paper we study analytically and numerically the spatiotemporal dynamics of a light pulse propagating in NGVD media with saturating nonlinearities. As a model nonlinearity we consider a particular type of saturating nonlinearity that, with increasing pulse intensity, changes the sign from the positive to the negative one. Recent measurements show that the polydiacetylene *para*-toluene sulfonate (PTS) exhibits this kind of saturation nonlinearities [4]. Indeed the nonlinear index of refraction corresponding to PTS is established to be $\delta n = n_2 I + n_4 I^2$, where I is intensity of the electromagnetic (EM) radiation. For the 1.6 μm laser radiation the measured values of second and fourth-order optical indices are respectively $n_2 = 2.2\times10^{-3}$ cm^2/GW and $n_4 = -0.8\times10^{-3}$ cm^4/GW^2. The critical intensity at the peak of the pulse profile giving $\delta n = 0$ is $I_0 = |n_2/n_4| = 2.75$ GW/cm^2. Such a nonlinearity can be considered even for the intensities above the critical one (I_0). In this case the nonlinear index of refraction becomes negative at the peak while remaining positive in the wings of the pulse intensity profile. A phenomenon of spatial ring formation has been observed in PTS due to the nonlinearity sign changes at the beam center when the two-dimensional beam intensity $I(\sim 8$ GW/cm$^2)$ is above the critical one [5].

Although PTS exhibits the largest saturating nonlinearity known in any material one may not be remiss in speculating that the same kind of saturating nonlinearities may be obtained by cascading in noncentrosymmetric media with large effective nonlinear coefficients n_2^{eff} and n_4^{eff}. The appropriate sign of these coefficients can be obtained choosing the occurrence sign of interacting waves. These coefficients can be increased by extending the length of nonlinear medium [6]. Dynamics of ultrashort laser pulses will be strongly affected by NGVD of media. Since diffraction and dispersion operators are of opposite sign, the intense laser pulse evolution in Kerr media results from the competition between two main tendencies, the pulse compression in the transverse spatial direction (the self focusing), and the pulse stretching along the time axis (the temporal dispersion). In media with saturating nonlinearity that changes the sign above a critical intensity, the pulse near its peak value undergoes spatial diffraction while compressing in time, contrary to the behavior in Kerr materials. In what follows we demonstrate that under certain conditions NGVD leads to temporal wave collapse in one dimensional geometry, while in bulk media catastrophic temporal blow-up is arrested by spatial splitting of pulse into smaller cells.

The dynamics of EM pulse propagating in nonlinear materials is based on the analysis of NSE

$$2ik\left(\frac{\partial \mathcal{E}}{\partial z} + \frac{1}{v_g}\frac{\partial \mathcal{E}}{\partial t}\right) + \Delta_\perp \mathcal{E} - kD\frac{\partial^2 \mathcal{E}}{\partial t^2} + 2k^2\frac{\delta n(|\mathcal{E}|^2)}{n_0}\mathcal{E} = 0, \tag{1}$$

where $\mathcal{E}$ is a slowly varying field envelope, v_g is the group velocity of the pulse propagating along the z axis, n_0 and $\delta n(|\mathcal{E}|^2)$ are respectively linear and nonlinear optical indices, and $\Delta_\perp = \partial^2/\partial x^2 + \partial^2/\partial y^2$ is the two-dimensional Laplacian describing beam diffraction, k is wave vector, and $D = d^2k/d\omega^2$ is group velocity dispersion (GVD). The EM pulse propagation in media with normal GVD is considered.

The dimensionless NSE reads

$$i\frac{\partial E}{\partial z} - \frac{\partial^2 E}{\partial \tau^2} + \Delta_\perp E + f(|E|^2)E = 0, \tag{2}$$

where the amplitude of the field envelope and all coordinates are appropriately renormalized (see Skarka *et al.* in Ref. [2]). A retarded time variable $\tau = t - z/v_g$ is used. In the case of cylindrically symmetric pulses, the following Lagrangian density is associated with Eq. (2):

$$L = r\left|\frac{\partial E}{\partial r}\right|^2 - r\left|\frac{\partial E}{\partial \tau}\right|^2 + \frac{i}{2}r\left(E\frac{\partial E^*}{\partial z} - E^*\frac{\partial E}{\partial z}\right) - rF(|E|^2), \tag{3}$$

where the radius $r = (x^2+y^2)^{1/2}$ and the nonlinear term $F(u) = \int_0^u f(u')du'$.

General dynamical properties of nonstationary solutions of Eq. (2) are rather complex. One has to resort to computer simulation in order to investigate the solutions of such an equation. However, the obtained simulation data can be qualitatively understood using analytical approach. We study the pulse dynamics governed by NSE using the variational approach [7]. The localized solution is approximated by a Gaussian trial function

$$E = A(z)\exp\left(-\frac{r^2}{2R^2(z)} - \frac{\tau^2}{2T^2(z)} + i(r^2 b(z) + \tau^2 c(z) + \phi(z))\right). \tag{4}$$

The self-similar pulse evolution in the z direction is parametrized by amplitude A, phase ϕ, transverse width R, and temporal duration T. The parameter b is wave front curvature and c the ''temporal curvature'' corresponding to the chirp.

Substituting trial function into Eq. (3) and integrating over r and τ, the average Lagrangian is obtained. It depends only on optimizing z-dependent parameters of this trial function. The condition that the variation of average Lagrangian with respect to each of these parameters is zero gives corresponding Euler-Lagrange equations. The equations for effective forces following respectively R and T ''directions'' are

$$\frac{d^2R}{dz^2} = F_R = -\frac{\partial}{\partial R}V(R,T), \tag{5}$$

$$\frac{d^2T}{dz^2} = F_T = 2\frac{\partial}{\partial T}V(R,T), \tag{6}$$

where V is the effective potential

$$V(R,T) = \frac{2}{R^2} - \frac{1}{T^2} - \frac{K(A^2)}{A^2} \tag{7}$$

with the nonlinearity function

$$K(u) = \frac{8}{\sqrt{\pi}}\int_0^\infty dp\, p^2 F(ue^{-p^2}). \tag{8}$$

The ''energy,'' $N = A^2R^2T$ is conserved during the pulse evolution. The wave front curvature is given by the equation $b = (1/4R)dR/dz$, while the chirp parameter is $c = -(1/4T)dT/dz$.

The dynamics that follows from Eqs. (5)–(8) is analogous to the behavior of a ''particle'' in a two-dimensional potential. Pushing analogy further gives us better physical understanding of the problem. Notice that in T direction the effective particle has a negative mass. Since the net force acting on the particle is always nonvanishing $[F = (F_T^2 + F_R^2)^{1/2} \neq 0]$, the localized steady solution corresponding to the equilibrium does not exist. One can readily show that $d^2(R^2+T^2)/dz^2 > 0$. Consequently the pulse spreads at least in one of two dimensions, R and T. Nevertheless, a catastrophic growth of the field amplitude, so-called collapse, may occur in the remaining dimension, i.e., either R or T may tend to zero for finite z.

In the general formalism we did not use an explicit form for the nonlinear function $f(|E|^2)$. In order to investigate the dynamical properties of nonsteady solutions, in the subsequent analysis we will consider a nonlinear term of the following form:

$$f(|E|^2) = |E|^2 - |E|^4. \tag{9}$$

Such a saturating nonlinearity has been widely applied in different domains of research [8]. However, in what follows we do not limit ourselves to the case when the second term in Eq. (9) is smaller than the first one. The nonlinearity function K in Eq. (8) is now $K(A^2) = \alpha A^4 - \beta A^6$, where $\alpha = 2^{-3/2}$ and $\beta = 2\times 3^{-5/2}$. The forces acting on the effective particles respectively in R and T direction are written as

$$F_R = \frac{4}{R^3} - \frac{2}{R}Q(A^2) \tag{10}$$

and

$$F_T = \frac{4}{T^3} + \frac{2}{T}Q(A^2) \tag{11}$$

with the nonlinearity

$$Q = \alpha A^2 - 2\beta A^4 = \frac{\alpha N}{R^2T} - \frac{2\beta N^2}{R^4T^2}. \tag{12}$$

In the Kerr media ($\beta = 0$) the nonlinearity Q is always positive and consequently the force $F_T > 0$ pushes the particle towards higher T. Assuming that the initial ''velocity'' of particles, i.e., the chirp is zero, the pulse always spreads in temporal domain. If the force F_R in spatial direction is initially positive the pulse also spreads in spatial domain. In the opposite case ($F_R < 0$), the initially negative force can lead either to the collapse ($R \to 0$) or to the final spreading. This behavior is related to the competition between the increase of duration T and decrease of radius R. If the duration T reaches the value $T = \alpha N/2$ corresponding to the force inflection

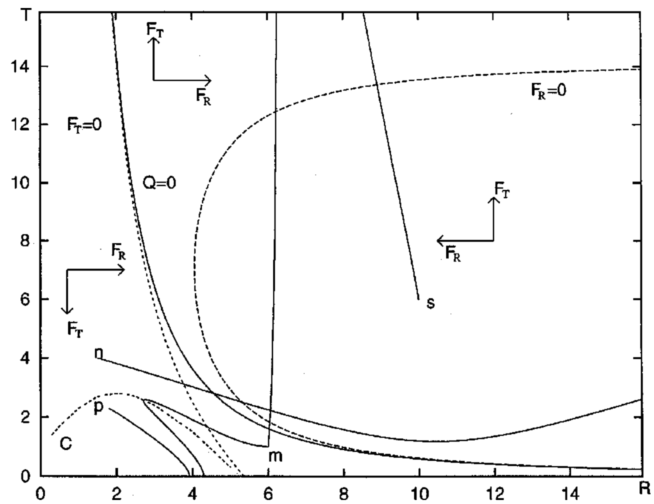

FIG. 1. The time and space components of the force in the domains with the positive $(Q>0)$ and with the negative nonlinearity $(Q<0)$. The arrow indicates the direction of the force components.

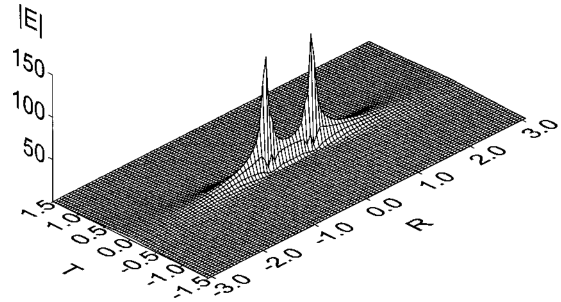

FIG. 2. The Gaussian pulse with the parameters in the collapse region, $R=4$, $T=1$, and $N=80$, splits in spatial domain.

point before the radius shrink to zero, the collapse is prevented since the force changes the sign and the radius now starts spreading [3].

In general, the saturating nonlinearity prevents the collapse. However, the nonlinearity (9) changes the sign for higher field intensities and interesting phenomena may occur. For instance, the pulse may undergo the collapse in time dimension. The pulse dynamics is considered in both the positive and the negative nonlinearity domains that are separated by the line $Q=0$ in Fig. 1, illustrating the case when the energy is $N=80$. At the right hand side of this line the duration T is always increasing but the radius R spreads only outside of the dashed curve corresponding to the zero force in radial direction $(F_R=0)$. The force F_R is negative inside the curve producing the initial shrinking of radius which, due to the saturation, leads during evolution to the inflection point $(F_R=0)$ but never to the collapse. Consequently, in the positive domain $Q>0$, the final stage of evolution corresponds to the spreading in both dimensions. In the negative domain $(Q<0)$, only at the right hand side of the dotted curve $(F_T=0)$ both widths, R and T, always spread. Since the force F_T is negative on the left of this curve, the initial T-shrinking takes place. In the zone below the line C the collapse in T dimension occurs although R is always spreading. In the remaining part of the domain $F_T<0$, passing the force inflection point the duration T starts to increase. In order to test the behavior of the pulse having different initial conditions, we plot the trajectories of corresponding particles. The particles s and n having zero initial velocities go to infinity. The spatial width R, corresponding to the particle s initially decreases, however, whenever this particle enters the region above the curve $F_R=0$, the force F_R changes the sign and pushs the particle towards $R\to\infty$. As a consequence, the trajectory of the particle will bend to the right side (this part of trajectory lies out of the plot in Fig. 1). Thus the pulses corresponding to the particles s and n, ultimately spread in space and in time. All pulses with initial parameters below the line C collapse in time (see, for instance, the trajectory of the particle p). A particle initially out of the concerning domain can reach it having an appropriate initial velocity oriented towards this region. The position of the particle m with initial zero velocity predetermine its evolution towards infinity (see upper curve starting at m), but judicious choice of its initial velocity indeed brings it in the collapse domain. The pulse destinated to spread, can however collapse if it is initially prefocused. The second, lower curve starting at point m corresponds to the trace of the particle having an initial velocity $(v_R=-10,\ v_T=0)$. Thus, using variational approach we showed that the short laser pulse above the critical intensity may undergo the temporal collapse in saturating media.

The main and well known shortcoming of the variational approach is that it is not able to account for structural changes of the pulse shape. Consequently the result of variational approach can be used to predict dynamics of the pulse before the considerable changes of its shape take place. To see the realistic behavior of the pulse we carried out numerical simulations of Eq. (2) for different initial parameters of the pulse. The results of simulations qualitatively agree with the predictions of the variational approach in the domain of parameters out of the collapse region. A pulse with such initial parameters undergoes initial temporal or spatial shrinking and after spreads out in both directions. The structural changes of the pulse at later stages of its dynamics do not alter such evolution significantly.

Our primary goal is to verify the prediction of variation approach concerning the possibility of the temporal collapse of the pulse. During the propagation in the collapse region, $R=4$, $T=1$ and $N=80$, the Gaussian pulse considerably contracts in temporal domain as predicted by analytical approach. Though amplitude of the field increases significantly, the unlimited amplitude growth is prevented by pulse splitting in spatial domain (see Fig. 2). This ultimate stage of pulse evolution is beyond the reach of the variational approach. The splitting presented in space only, as a function of x and y variables, corresponds to the ring formation (see Fig. 3) reported in a recent experimental study [5]. The pulse center initially situated in origin undergo spatial spreading and the sharpening of the ring edge occurs. Ulterior evolution leads to the further splitting into the new concentrical rings with the tendency of asymptotical spreading. Thus, the variational approach predicts existency of a domain of parameters where the pulse has a tendency to increase its amplitude significantly. However, only numerical simulations can show the real end up of the evolution: the temporal collapse, i.e., the development of a mathematical singularity is prevented by splitting in the spatial domain.

For the same saturating nonlinearity, essentially the same

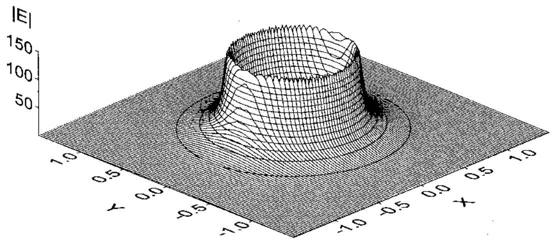

FIG. 3. The ring is obtained by plotting the pulse amplitude versus the space coordinates X and Y.

behavior as in bulk media can be obtained in the two-dimensional wave guide geometry.

In one-dimensional wave guides (like fibers) the variational approach also predicts the occurrence of the temporal collapse. Due to the field confinement in transverse directions the spatial splitting of the field is prevented. Consequently, the temporal collapse takes place as can be seen by an exact analysis. Indeed, for one-dimensional wave guides the Eq. (2) can be written as

$$i\frac{\partial E}{\partial z}-\frac{\partial^2 E}{\partial \tau^2}+|E|^2E-|E|^4E=0. \qquad (13)$$

The momentum approach gives us the following virial type relation:

$$\frac{d^2}{dz^2}\int d\tau \tau^2|E|^2=8H-2\int d\tau |E|^4, \qquad (14)$$

where

$$H=\int d\tau\left[\left|\frac{\partial E}{\partial \tau}\right|^2+\frac{1}{2}|E|^4-\frac{1}{3}|E|^6\right] \qquad (15)$$

is the integral of motion of Eq. (13) called Hamiltonian. The wave collapse takes place if $H<0$, since the second term at the right hand side of Eq. (14) is negative. The unlimited growth of the field amplitude occurs at the finite propagation distance. The condition $H<0$ for an unchirped Gaussian pulse leads to the following inequality:

$$(3\beta A_0^2-2\alpha)A_0^2>\frac{1}{T^2}. \qquad (16)$$

Thus necessary condition for the appearance of collapse is $A_0^2>2\alpha/3\beta=1.84$.

Therefore, the occurrence of the one-dimensional temporal collapse follows directly from the momentum theory without any approximation. Such a collapse cannot be arrested.

In conclusion, the laser pulse dynamics in NGVD media with sign changing saturating nonlinearity is considered. It is shown that the pulse above critical intensity can undergo temporal collapse. In bulk and planar media the collapse is arrested by spatial splitting while in the one-dimensional geometry the blow up of the field imminently takes place. The tendency of temporal collapse can be used to produce intense ultrashort pulses choosing the appropriate length of the guide.

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