

Nonlinear wave equation to describe propagation of ultrashort high-power laser pulses in matter

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Using numerical techniques, we have solved the nonlinear wave equation modeling time-dependent spatially two-dimensional propagation of ultrashort high-power laser pulses in matter with a nonlinear relativistic ponderomotive term. Auxiliary dynamical modulation of the pulse is established and the process by which a precursor forms is described. It is shown that for a picosecond pulse there is quantitative agreement between the solution of the nonlinear wave equation and the simpler nonlinear Schrödinger equation only for times prior to the appearance of the first focus. At times between the first and second focus the solutions agree only qualitatively. Thereafter they disagree entirely.

1. INTRODUCTION

1.1. Limitations on the nonlinear Schrödinger equation in the nonlinear optics of ultrashort radiation pulses

The propagation of an electromagnetic wave in free space is described by a linear wave equation, which is a consequence of the Maxwell equations. Electromagnetic wave propagation at low intensities in matter is also described by a wave equation with constant coefficients (linear optics). As the intensity of the electromagnetic wave increases, its propagation in matter is often described by a nonlinear wave equation whose coefficients depend on the intensity of the radiation. The direct solution of the nonlinear wave equation is difficult, so simplifying approaches are often used.

To describe the transport of intense light beams and highly collimated laser pulses in matter, a technique has come into wide use which involves reducing the problem to the solution of the nonlinear Schrödinger equation (NSE) for the slowly varying complex amplitude of the electromagnetic field. The use of the NSE is justified if the complex amplitude of the electromagnetic field varies little over distances on the order of the wavelength in the direction of beam propagation, and over times of the same order as the optical oscillations of the field.

When the radiation intensity varies smoothly over the propagation distance, the applicability of the NSE is not in question, although the problem of estimating quantitatively the differences between the NSE and the nonlinear wave equation as a function of distance is of interest in its own right. However, in many studies the NSE is used to describe self-focusing, and not only of steady beams, but also short or even ultrashort ($\tau_0 < 1$ ns) laser pulses. As is well known, in these problems time-dependent wave structures develop, consisting of powerful foci propagating in the medium with sharply varying field amplitudes at their fronts. Close to the foci, the conditions for the applicability of the NSE break down. On the other hand, it is known that the solution of the

NSE is unstable, i.e., small perturbations can grow. Consequently, the small deviations in the solutions of the nonlinear wave equation and the NSE which accumulate when the radiation passes through the first focus result in additional discrepancies between the two methods. Thus, the applicability of the NSE for describing the nonlinear propagation of ultrashort laser pulses after formation of the first focus requires particular study.

1.2. Properties of the nonlinear propagation of ultrashort pulses in matter

Let L be the nonlinear propagation distance for laser radiation in matter, $L_d = d^2/\lambda$ be the diffraction length associated with the transverse dimension d of the pulse in matter, λ be the wavelength of the laser radiation, τ_0 be the length of the laser pulse, and τ_{nl} the time needed to develop the nonlinear response of the medium. At present we can recognize the following fundamental regimes of nonlinear laser radiation propagation in matter:

- 1) steady state, including waveguide propagation¹⁻⁵
 $L \gg L_d$;
- 2) the quasisteady regime with a fast nonlinearity^{6,7}
 $\tau_0 \gg \tau_{nl}$, $\tau_0 \gg L/c$;
- 3) the time-dependent regime with a fast nonlinearity⁸
 $\tau_0 \gg \tau_{nl}$, $\tau_0 \ll L/c$;
- 4) the time-dependent regime with a slow nonlinearity⁹
 $\tau_0 < \tau_{nl}$.

The first regime arises when radiation from CW lasers propagates in matter. The second regime usually arises for Q -switched lasers. The third regime, as a rule, is achieved for lasers with ultrashort pulse lengths $\tau_0 < 1$ ps. The fourth regime occurs when the nonlinear response of the medium reacts back on the radiation with a lag of the order of the pulse length itself.

The present work is devoted to justifying the theoretical description of the ultrashort lasers most frequently encountered in practice and to the less thoroughly studied regime 3.

Regimes 1, 2, and 4 have been studied in reviews¹⁰⁻¹² and in the monograph of Ref. 13.

This nonlinear propagation regime for laser radiation was referred to in Ref. 14 as self-channeling of an ultrashort pulse. Essentially it amounts to saying that a burst of electromagnetic field propagates in a medium with variable dielectric properties over a distance L many times the length $L_i = c\tau_0$ of the pulse itself and the diffraction length, $L \gg L_i, L_d$. Over the propagation length L of the pulse there is no physical channel in the medium. The filament, otherwise known as the channel, is nothing but the trajectory or track along which the burst of electromagnetic field with dimensions small in the longitudinal and transverse directions propagates.

The effect of relativistic ponderomotive self-focusing of an ultrashort high-power laser pulse was predicted theoretically¹⁵ and subsequently observed experimentally.¹⁶

Borisov *et al.*¹⁷ observed self-channeled propagation of the pulse from a KrF* excimer laser ($\lambda = 0.248 \mu\text{m}$, $\tau \approx 600$ fs, $P \approx 3 \cdot 10^{11}$ W) over a distance of up to 2 mm, which amounts to $L \approx 100L_d$. The channel diameter was $d \approx 1 \mu\text{m}$. The length L of self-channeled propagation was determined by the rate of dissipation of the pulse energy in the material.

The self-channeling of an ultrashort high-power laser pulse leads to strong self-concentration of the optical energy in a small region which is displaced in the material with a velocity close to that of light (the intensities can reach magnitudes $I > 10^{20}$ W/cm²). This opens up interesting prospects for various new fundamental and practical applications of laser radiation, and also essentially changes the form of the conditions for familiar physical processes to occur.^{18,19} For these reasons it is of interest to study this phenomenon.

Until recently the theory of the self-channeling effect was based on the use of the NSE.^{14,15,20-22} In Ref. 8 a theory was constructed based on the modified nonlinear Schrödinger equation, which took into account, in addition to the NSE, the second derivatives in the longitudinal coordinate. In the general case the process is described by the nonlinear wave equation.²³

In the present work we use numerical techniques for the first time to solve the three-dimensional problem of the time-dependent spatially two-dimensional (one time and two space) propagation of an ultrashort high-power laser pulse in a medium with a relativistic ponderomotive electron nonlinearity. The process by which the laser pulse is stretched out in the longitudinal direction (precursor formation) is described, and additional spatial modulation of the pulse is also established. A detailed comparison is made of the solutions obtained using the NSE and the nonlinear wave equation.

2. FORMULATION OF THE PROBLEM

References 8, 14-19, and 21, 22 are devoted to the theoretical analysis of self-channeling of ultrashort high-power laser pulses in matter. The nonlinear wave equation which describes the effect takes the form⁸

$$\left(\frac{1}{v_g} \partial_t + \partial_z \right) a + \frac{i}{2k} \left\{ \square + k_p^2 \left[1 - \frac{(1 + k_p^{-2} \Delta \gamma)}{\gamma} \right] \right\} a = 0,$$

$$\gamma = (1 + |a|^2)^{1/2}. \quad (1)$$

Here $a(r, z, t)$ is the complex amplitude of the vector potential of the electromagnetic field, v_g is the group velocity, and we have written $k^2 = k_0^2 - k_p^2$, where $k_0 = \omega/c$ is the wave number and $k_p = \omega_{p,0}/c$ and $\omega_{p,0} = (4\pi e^2 n_{e,0}/m_{e,0})^{1/2}$ are the Posner wave number and the unperturbed plasma frequency, and $\square = \Delta_{\perp} + \partial_{zz}^2 - c^{-2} \partial_{tt}^2$ is the wave operator.

The first factor (enclosed in parentheses) is the linear transport operator for the complex amplitude of the vector potential with group velocity v_g in the z direction. The wave operator in braces describes diffraction of the radiation. The second term describes refraction; the γ in the denominator accounts for relativistic increase in the mass of the free electron oscillating in the superintense electromagnetic field; the term $k_p^{-2} \Delta \gamma$ is the ponderomotive term, which describes the force acting to push electrons into the region where the field is weaker.

If we specify an infinite medium $-\infty < z < \infty$ and at time $t=0$ a pulse of known form is introduced, then it is convenient to change variables so as to localize the solution:

$$\xi = v_g t - z, \quad \tau = t. \quad (2)$$

Then Eq. (1) assumes the form

$$\frac{1}{v_g} \partial_{\tau} a + \frac{i}{2k} \left\{ \Delta_{\perp} + \left(1 - \frac{v_g^2}{c^2} \right) \partial_{\xi\xi}^2 - \frac{2v_g}{c^2} \partial_{\xi\tau}^2 - \frac{1}{c^2} \partial_{\tau\tau}^2 + k_p^2 \left[1 - \frac{1 + k_p^{-2} (\Delta_{\perp} \gamma + \partial_{\xi\xi}^2 \gamma)}{\gamma} \right] \right\} a = 0. \quad (3)$$

The transformation (2) performs a rotation in z, t space so as to make the direction of propagation parallel to the time axis.

To pose the problem for the nonlinear wave equation (3), we must include the initial conditions

$$a(r, \xi, \tau=0) = a_0(r, \xi), \quad \partial_{\tau} a(r, \xi, \tau=0) = a_1(r, \xi), \quad (4)$$

and the boundary conditions

$$a(r=\infty, \xi, \tau) = 0, \quad \partial_r a(r=0, \xi, \tau) = 0, \\ a(r, \xi = \pm \infty, \tau) = 0. \quad (5)$$

If we assume that the complex amplitude varies slowly over distances on the order of the wavelength in the direction of propagation and over times on the order of the oscillation period of the field $|\partial_z a|, c^{-1} |\partial_t a| \ll k|a|$, then the wave operator and the Laplacian in the nonlinear term should be replaced by transverse Laplacians: $\square, \Delta \rightarrow \Delta_{\perp}$. The resulting equation is called a nonlinear Schrödinger equation and has been the subject of a great deal of work.¹⁴⁻²²

In view of the complexity of the nonlinear wave equation (3), it is usual in the literature⁸ to introduce an equation in which the cross-term $\partial_{\xi\tau}^2$ and the second derivative with respect to time $\partial_{\tau\tau}^2$ are omitted, but which takes into account the second derivatives with respect to the longitudinal coordinate, $\partial_{\xi\xi}^2$. This equation is called the modified nonlinear Schrödinger equation.

3. NUMERICAL ANALYSIS OF SELF-CHANNELING IN THE NONLINEAR WAVE EQUATION MODEL

3.1. The numerical method

We will assume that at time $\tau=0$ the pulse has a hyper-Gaussian intensity profile in the radial and longitudinal coordinates:

$$|a_0(r, \xi)|^2 = a_0^2 \exp\left(-\left(\frac{r}{r_0}\right)^{N_1} - \left(\frac{\xi}{\xi_0}\right)^{N_2}\right), \quad \xi_0 = \tau_0 v_g. \quad (6)$$

We specify the phase distribution in the form of a uniform plane front:

$$a_0(r, \xi) = |a_0(r, \xi)| e^{i\varphi_0(r, \xi)}, \quad \varphi_0(r, \xi) = 0. \quad (7)$$

We set the first derivative with respect to time equal to zero initially:

$$a_1(r, \xi) = 0, \quad (8)$$

which corresponds to the analogous condition in linear optics.

The problem specified by Eqs. (3)–(5) has been solved using a spectral–finite-difference method on an infinite domain: semi-infinite in r and extending to infinity in both directions in ξ . In terms of the variable ξ the problem can be treated as two semi-infinite regions. From the leading edge $\xi=0$ of the pulse the continuation beyond the trailing edge is in the direction $+\infty$. From $\xi=0$ the continuation beyond the leading edge is in the direction $-\infty$. The spectral–finite-difference method we used is an outgrowth of techniques for solving nonlinear problems involving partial differential equations developed in Refs. 24–26. The numerical algorithm for the problem defined by Eqs. (3)–(5) is implemented using the collocation method based on representing the desired functions as a finite sum of Chebyshev polynomials of the first kind. In each region we arrive at a system of second-order ordinary differential equations in time. The conditions for the continuity of the solution and its derivative with respect to ξ at the point $\xi=0$ yield the requisite equations for simultaneous solution of these two systems. This technique can be regarded as a version of the finite-element method. The points of the collocation grid in this version are bunched up in both directions near the point $\xi=0$, which enables us to get a detailed description of the process by which the precursor forms. By means of a change of variables we arrive at a system of nonlinear ordinary differential equations with twice as many dependent variables, but now of first order. To integrate the latter in time we have used a predictor-corrector scheme²⁶ with a second-order predictor.

3.2. Solution of the 2D+1 problem

The main purpose of this work is to simulate the propagation of an ultrashort high-power laser pulse in matter by numerically solving the nonlinear wave equation. We have treated the propagation of a Gaussian $N_1=2, N_2=2$ pulse from an excimer laser with wavelength $\lambda=0.248 \mu\text{m}$ and aperture $2r_0=6 \mu\text{m}$, of length $2\tau_0=800$ fs with energy $E=6.0$ J in a plasma with electron density $N_e=7.5 \cdot 10^{20} \text{cm}^{-3}$. Figures 1a–d show the solutions of the nonlinear

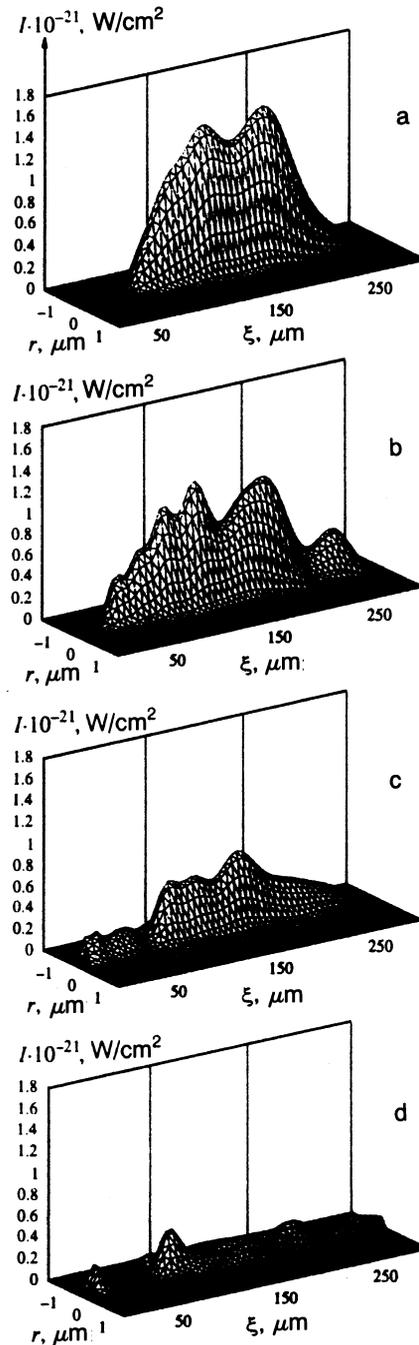


FIG. 1. Two-dimensional surface plots of the intensity profile for an initially Gaussian laser pulse with wavelength $\lambda=0.248 \mu\text{m}$, aperture $2r_0=6 \mu\text{m}$, length $2\tau_0=800$ fs, and energy $E=6.0$ J, propagating in a plasma with electron density $N_e=7.5 \cdot 10^{20} \text{cm}^{-3}$. The calculations were carried out using the nonlinear wave equation. The figures are shown at times τ (in fs): a) 150.0; b) 187.5; c) 262.5; d) 281.25.

wave equation (3) at times $\tau=150.0, 187.5, 262.5$ and 281.25 fs respectively.

The initial Gaussian profile of the laser pulse is not shown in Fig. 1. Its intensity is lower than those shown in Fig. 1 by more than a factor of 100; it is 25 times as wide in the radial direction. On the scale of Fig. 1 the initial profile would look like a nearly zero uniform background. As it propagates the pulse converges toward the axis in the trans-

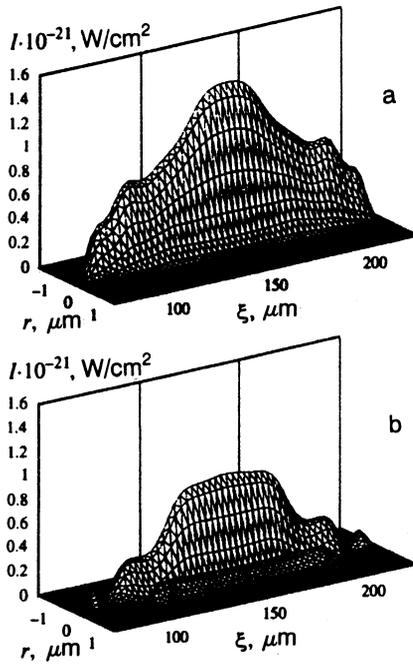


FIG. 2. Two-dimensional surface plots of the intensity profile for an initially Gaussian laser pulse with wavelength $\lambda=0.248 \mu\text{m}$, aperture $2r_0=6 \mu\text{m}$, length $2\tau_0=800 \text{ fs}$, and energy $E=6.0 \text{ J}$, propagating in a plasma with electron density $N_e=7.5 \cdot 10^{20} \text{ cm}^{-3}$. The calculations were carried out using the modified NSE. The figures are shown at times τ (in fs): a) 187.5; b) 262.5.

verse direction. Figures 1a, b illustrate the pulse passing through the first focus; Fig. 1c shows the second focus. Figure 1d illustrates the intensity profile that develops beyond the second focus.

In order to analyze the conditions for the applicability of the simpler models shown in Figs. 2a,b we have presented the solution of the modified NSE for exactly the same problem in which we solved the nonlinear wave equation (3) at times $\tau=187.5$ and 262.5 fs respectively. This solution is illustrated in more detail in Ref. 8. From the physical standpoint, Fig. 2a illustrates the two-dimensional intensity profile of the laser pulse at the time when the first focus forms in the center; Fig. 2b is close to the second focus.

The most important thing to note is the asymmetry of the solutions of the nonlinear wave equation in the ξ direction. This is due to the inclusion of the mixed derivative term $\partial_{\xi\tau}^2$ in the complete model. The solutions of the modified NSE are symmetric in ξ , as can be clearly seen in Fig. 2. Thus, taking the mixed derivative into account is a very important consideration for the correct solution of the problem.

Qualitative agreement in the development of the structure of the foci using the NSE and nonlinear wave equation models is only seen in the approach to the first focus; thereafter the solution in the nonlinear wave equation model qualitatively and quantitatively differs from that obtained with the modified NSE. The qualitative discrepancy results mainly from the effect of the second derivative in time, $c^{-2}\partial_{\tau\tau}^2$. The self-modulation of the pulse in the ξ direction in the nonlinear wave equation model begins to develop earlier than in the modified NSE, which is especially clear when Figs. 1b and 2a are compared. Thus, including the second

derivative with respect to time is also an important consideration.

Comparison of Figs. 1c and 2b shows that part of the radiation in the nonlinear wave equation model proceeds ahead, outstripping the main pulse. That is to say, a precursor forms. Note that the leading edge of the pulse corresponds to a smaller value of ξ in Fig. 1. In the nonlinear wave equation model the pulse is stretched out in the direction of propagation; this is clearly seen in Fig. 1d.

The calculations shown in Figs. 1 and 2 were carried out for a peak value $P_0/P_{cr}>10$. In the present work we have also considered cases with a small ratio $P_0/P_{cr}\approx 1.4$. They exhibited retardation of the self-focusing in the nonlinear wave equation model. The first and last foci developed at somewhat larger distances than in the NSE model. This effect is due to spreading of the pulse in the longitudinal direction in the nonlinear wave model, which naturally causes the conditions for self-focusing to deteriorate.

4. CONCLUSIONS

In the present work we have continued the theoretical study of the nonlinear wave equation (1) describing the propagation of ultrashort high-power laser pulses in matter, including the effects of diffraction and refraction in a medium with a nonuniform radial profile of the index of refraction resulting from electron relativistic and ponderomotive nonlinearities.

Two problems arise when the nonlinear wave equation (1) is studied. The first is related to the analysis of the effect of nonlinearity on the propagation of radiation. The relativistic-ponderomotive nonlinearity is a combination of the two, and it is therefore of interest to clarify the role of each effect individually. This can be done by considering a hierarchy of increasingly complicated nonlinearities. In the weakly relativistic approximation we have a Kerr-type nonlinearity; at higher intensities, a relativistic nonlinearity, and then the relativistic-ponderomotive nonlinearity. Some aspects of this problem have been considered in Refs. 14 and 21.

The second problem is related to the proper treatment of the wave properties of Eq. (1). When we go to the comoving variable in order to localize the solution of Eq. (1) spatially it takes the form, e.g., of Eq. (3). For Eq. (3) it is traditional (beginning with the work of V. I. Talanov, V. I. Lugovoi, and A. M. Prokhorov; see the reviews in Refs. 10–12) to use the approximation in which the complex amplitude of the electromagnetic field is slowly varying. In this approximation all second-order derivatives $\partial_{\xi\xi}^2$, $\partial_{\xi\tau}^2$, $\partial_{\tau\tau}^2$ which enter into the NSE should be omitted. Other approximations are also possible. For example, the equation in which the derivative $\partial_{\xi\xi}^2$ is retained but the $\partial_{\xi\tau}^2$, $\partial_{\tau\tau}^2$ derivatives are omitted is naturally called the modified NSE. We can treat the form of the equation in which only the second derivative with respect to time $\partial_{\tau\tau}^2$ is omitted, and all the others are retained. The resulting equation is classified as a modified NSE with a mixed derivative. It would be of the greatest interest to treat the exact nonlinear wave equation (3), including all the derivatives. Thus, including the wave properties of Eq. (1) leads us to the study of a hierarchy of four models: the NSE, the modified

NSE, the modified nonlinear wave equation, and the nonlinear wave equation itself. In each model, in principle, we can treat different nonlinearities.

Early work analyzed only the eigenmodes of the two-dimensional NSE. Most studies of Eq. (1) for different forms of nonlinearity were carried out using the NSE model, but only in the two-dimensional problem which describes the evolution of a specified thin transverse slice of the laser pulse. Among these one should note the studies carried out with the participation of A. B. Borisov, in which the corresponding numerical techniques were developed.^{14,21} Recently Ya. M. Zhileikin has written a fast-running code that solves the three-dimensional (one temporal, two spatial) problem in the nonlinear wave equation model.²⁹ In a recent paper this three-dimensional problem was solved for the modified NSE model with a relativistic-ponderomotive nonlinearity.

In the present work the study of the wave properties of Eq. (3) led to the development of the basic ideas.⁸ The numerical calculations were carried out for the relativistic-ponderomotive nonlinearity, which when certain conditions are imposed leads to the self-channeling of an ultrashort high-power laser pulse in matter (in the plasma produced by the pulse itself²¹). In carrying out this work we wrote a code specifically designed for a 486/66 MHz IT computer, intended to solve two- and three-dimensional problems using the four models listed from the NSE to the nonlinear wave equation. It is based on spectral-finite-difference methods with expansion of the dependent variables in both spatial variables in finite series of Chebyshev polynomials of the first kind. The three-dimensional (one temporal, two spatial) problems were solved for the propagation of an initial pulse which was Gaussian in both directions in a nonlinear medium. Special attention was given to solving the nonlinear wave equation, since this is the more accurate model.

In solving the 2D time-dependent problem with the nonlinear wave equation we found the following physical effects:

First, the existence of an asymmetry in the longitudinal (ξ) direction due to the retention in the model of the second-order mixed derivative term.

Second, strong self-modulation of the laser pulse in the longitudinal direction due to the retention in the model of the second time derivative. Such self-modulation occurs when the pulse passes the first focus. In models using the NSE it is less pronounced and occurs considerably later. The pulsating nature of the modulation in the nonlinear wave equation model should be noted. The effect first appears and then disappears (it is smoothed out due to overlapping of the peaks).

Third, in the nonlinear wave equation a precursor develops over the course of time: part of the radiation moves on ahead, outstripping the main pulse. The laser pulse has a tendency to spread in the propagation direction. Note that this effect is decidedly wavelike and is not described by the NSE model.

Fourth, near the threshold the self-focusing of the pulse is observed to lag in time in the nonlinear wave equation model in comparison with calculations using the NSE model;

this is due to the longitudinal wave spreading of the pulse, which causes the focusing conditions to deteriorate.

We note an important fact revealed by the nonlinear wave equation calculations for picosecond pulses ($2\tau_0=800$ fs). It was found that the results of solving the problem using the NSE model agree qualitatively with the nonlinear wave equation solutions only over times prior to the appearance of the first focus. Then the agreement is only qualitative prior to the appearance of the second focus. Thereafter the solutions disagree greatly.

Thus, in order to develop a reliable picture of the nonlinear propagation of ultrashort laser pulses in a nonlinear medium we should use the nonlinear wave equation. The NSE model, which has attained general respectability in nonlinear optics, is probably valid in a very restricted region in problems involving the nonlinear propagation of ultrashort pulses.

Note that in the present work we have not taken into account a number of physical effects, such as ionization of the medium by the radiation, generation of plasma waves,²⁸ the Kerr nonlinearity due to the background ions, etc. There are prospects for studying these by including them in the nonlinear wave equation model.

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