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An improved nonlinear optical pulse propagation equation

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Abstract

A new equation for self-focusing of extremely focused short-duration intense pulses is derived using a method that treats diffraction and dispersion to all orders with nonlinearity present, and self-consistently determines the nonlinear derivative terms present in the propagation equation. It generalizes both the previous formulation of linear optical pulse propagation to the nonlinear regime, and the cw nonlinear regime propagation to the pulsed regime by including temporal characteristics of the pulse. We apply the new equation and propagate a tightly focused picosecond pulse in silica and explicitly show the effects of spatial-derivative nonlinear coupling terms. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

A formulation for propagating optical pulses and cw beams, valid to *all orders* in the dimensionless variables $\eta \equiv (\omega_0 \tau_p)^{-1}$ and $\epsilon \equiv (k_0 \sigma_p)^{-1}$, has been developed for the linear pulse propagation regime [1–3] and experimentally verified [4]. Here τ_p and ω_0 are the pulse duration and central frequency of the pulse, $k_0 = n(\omega_0)\omega_0/c$ is central wavevector magnitude in the medium, σ_p is the transverse pulse width and $n(\omega_0)$ is the refractive index at the central frequency. The method can be applied to propagate extremely short pulses and extremely focused pulses and cw beams without approximation. Consequently, linear near-field diffraction effects can be exactly calculated in dispersive media (even nonisotropic dispersive media [3]). Here we extend the approach of [3], which is based on a consistent and mathematically rigorous expansion of the linear dispersion relation, to include a nonlinear optical response of the medium. Previous studies have not treated nonlinearity to arbitrary orders [5a–11], or not treated temporal

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[12a–d] or spatial [13–15b] aspects of the expansion to arbitrary order. To go beyond these lowest order approximations in the nonlinearity and yet retain all orders in η and ϵ, we generalize our method by incorporating a systematic perturbation analysis used before for cw beams [15a,15b] into the formulation previously used for linear pulse propagation [3]. As a first example, we present here the results for propagation in an isotropic Kerr-type nonlinear media. Our treatment is limited only by our assumption that the coupling of backscattering modes can be neglected; we consider only a one-way propagation of the field and do not do a two-way propagation boundary matching [16a–d].

2. Propagation equation: linear susceptibility

We begin by considering the wave equation for the electric field

$$\left(\nabla_\perp^2 + \frac{\partial^2}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\vec{E}(x,y,z,t) = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\left(\vec{P}^{\mathrm{L}}(x,y,z,t) + \vec{P}^{\mathrm{NL}}(x,y,z,t)\right), \tag{1}$$

where $\vec{P}^{\mathrm{L}}$ and $\vec{P}^{\mathrm{NL}}$ are the linear and nonlinear polarization vectors, respectively, or in Fourier space

$$\left(k_x^2 + k_y^2 + k_z^2 - \frac{1}{c^2}\omega^2\right)\vec{E}(\vec{k},\omega) = \frac{4\pi}{c^2}\omega^2\left(\vec{P}^{\mathrm{L}}(\vec{k},\omega) + \vec{P}^{\mathrm{NL}}(\vec{k},\omega)\right). \tag{2}$$

We first consider the case of a linear isotropic medium where

$$\vec{P}^{\mathrm{L}} = \frac{1}{(2\pi)^2}\cdot\int \mathrm{d}^3k\,\mathrm{d}\omega\chi^{(1)}(\vec{k},\omega)\vec{E}(\vec{k},\omega) \times \exp(\mathrm{i}\vec{k}\cdot\vec{x} - \mathrm{i}\omega t)$$

and

$$\chi^{(1)}(\vec{k},\omega) = \frac{\epsilon(\vec{k},\omega)-1}{4\pi} = \frac{n^2(\omega)-1}{4\pi}.$$

The last equality holds for ordinary isotropic media. For the time being (in this section) we set the nonlinear polarization term to zero.

Let us rewrite the propagation equation in terms of the slowly varying envelope (SVE) $\vec{A}$ of the electric field for a light pulse, $\vec{E}(\vec{x},t) = \vec{A}(\vec{x},t)\,\exp(\mathrm{i}\vec{k}_0\cdot\vec{x} - \mathrm{i}\omega_0 t)$, or in Fourier space, $E(\omega,\vec{k}) = A(\omega-\omega_0,\vec{k}-\vec{k}_0)$. The SVE multiplies the quickly oscillating temporal and spatial terms associated with central frequency ω_0 and central wavevector $\vec{k}_0 = k_0\hat{z} = n(\omega_0)(\omega_0/c)\hat{z}$ in the expression for the electric field. Note that we have chosen the z-axis to be along the central wavevector. Upon substituting the SVE into Eq. (2), changing variables $\omega\to\omega+\omega_0$, $\vec{k}\to\vec{k}+\vec{k}_0$ so as to remove the central frequency and central wavevector from the SVE, Eq. (2) yields

$$\left(k_x^2 + k_y^2 + (k_z+k_0)^2 - \left[\frac{n(\omega+\omega_0)(\omega+\omega_0)}{c^2}\right]^2\right)\vec{A}(\vec{k},\omega) = 0. \tag{3}$$

This is a linear homogeneous equation in the SVE that can be solved for k_z as a function of k_x, k_y and ω, independent of the field. By rearranging Eq. (3) and expanding the term

$$k_z = \sqrt{(n(\omega+\omega_0)(\omega+\omega_0)/c)^2 - k_x^2 - k_y^2} - \omega_0 n \times (\omega_0)/c$$

appearing in the resultant equation in the quantities ω, k_x and k_y, and then Fourier transforming back to configuration space, the propagation equation for the SVE for linearly polarized light in an isotropic medium is obtained [3]:

$$\frac{\partial A}{\partial z} = -\beta_1\frac{\partial A}{\partial t} - \frac{\mathrm{i}}{2}\beta_2\frac{\partial^2 A}{\partial t^2} + \frac{1}{6}\beta_3\frac{\partial^3 A}{\partial t^3} + \frac{\mathrm{i}}{2}\gamma_{xx}\left(\frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2}\right) + \frac{1}{2}\gamma_{txx}\left(\frac{\partial^3 A}{\partial x^2\partial t} + \frac{\partial^3 A}{\partial y^2\partial t}\right) + \cdots.$$

Here β_1 is an inverse of group velocity, β_2 is the group velocity dispersion, β_3 the third-order dispersion, γ_{xx} is the Fresnel diffraction coefficient and γ_{txx} is the coefficient of the mixed space–time third-order term that accounts for the spherical nature of the wavefront surface of a pulse originating from a point source [3]. This equation takes on a particularly enlightening form in *dimensionless units* in which time is in units of the temporal pulse duration $\tau_p = (\omega_0\eta)^{-1}$, length in the transverse directions is in units of the characteristic transverse pulse width $\sigma_p = (k_0\epsilon)^{-1}$, and length in the propagation direction z (along the central wavevector of the optical pulse) is in units of the diffraction length $L_{DF} = k_0\sigma_p^2$ [10,11]

$$\begin{aligned}L_{DF}^{-1}\frac{\partial A}{\partial z} &= -\beta_1\omega_0\eta\frac{\partial A}{\partial t} - \frac{i\beta_2\omega_0^2\eta^2}{2}\frac{\partial^2 A}{\partial t^2} + \frac{\beta_3\omega_0^3\eta^3}{6} \\ &\times \frac{\partial^3 A}{\partial t^3} + \frac{i\beta_4\omega_0^4\eta^4}{24}\frac{\partial^4 A}{\partial t^4} \\ &+ i\frac{\gamma_{xx}k_0^2\epsilon^2}{2}\left(\frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2}\right) \\ &+ \frac{\gamma_{txx}\omega_0 k_0^2\epsilon^2\eta}{2}\left(\frac{\partial^3 A}{\partial x^2\,\partial t} + \frac{\partial^3 A}{\partial y^2\,\partial t}\right) \\ &+ i\frac{\gamma_{xxxx}k_0^4\epsilon^4}{8}\left(\frac{\partial^4 A}{\partial x^4} + 2\frac{\partial^4 A}{\partial x^2\,\partial y^2}\right. \\ &\left. + \frac{\partial^4 A}{\partial y^4}\right) + \cdots, \end{aligned} \tag{4}$$

where $\gamma_{xx} = c/(n(\omega_0)\omega_0)$, $\gamma_{txx} = c\beta_1/(n(\omega_0)\omega_0)^2$. The result is expressed in terms of the two dimensionless parameters ϵ and η which are crucial in developing our expansion procedure described below since the expansion will be in these variables. Note that the right-hand side (RHS) of Eq. (4) is expressed as a power series in ϵ and η. The effect of each of the terms on the RHS of Eq. (4) has been extensively discussed previously [3] and the values of the coefficients on the RHS of the equation have been specified.

It is difficult to use this method to develop a self-consistent treatment including nonlinearity [17]. Therefore, we develop a different method for treating the linear propagation problem that will allow generalization to inclusion of nonlinear terms to all orders in perturbation theory.

Eq. (1) can be rewritten in terms of the SVE, thereby eliminating quickly oscillating phases

$$\begin{aligned}&\left(\nabla_\perp^2 + 2ik_0\frac{\partial}{\partial z} + \frac{\partial^2}{\partial z^2}\right)A \\ &= \frac{-1}{(2\pi)^2c^2}\int d^3k\, d\omega\Big[(\omega+\omega_0)^2 n^2(\omega+\omega_0) \\ &\quad - \omega_0^2 n^2(\omega_0)\Big]A(\vec{k},\omega)e^{(i\vec{k}\cdot\vec{x}-i\omega t)}. \end{aligned} \tag{5}$$

Eq. (5) can be rearranged to take the form

$$\frac{\partial}{\partial z}A = \frac{1}{2ik_0}\left(-\nabla_\perp^2 - k_0^2\mathscr{F} - \frac{\partial^2}{\partial z^2}\right)A,$$

where the RHS of Eq. (5) has been written as $-k_0^2\mathscr{F}A$ thereby defining the linear differential operator $\mathscr{F}$. This operator is a polynomial of $\partial/\partial t$, which can be obtained as a power series by expanding integrand of the RHS of Eq. (5) in ω about ω_0. We now write the equation using dimensionless units of x, y, z and t

$$\left(L_{DF}^{-1}\frac{\partial}{\partial z}\right)A = \frac{ik_0}{2}\left(\mathscr{L}_0 + \frac{1}{k_0^2}\left(L_{DF}^{-1}\frac{\partial}{\partial z}\right)^2\right)A, \tag{6}$$

where the operator $\mathscr{L}_0$ is defined as

$$\mathscr{L}_0 \equiv \left(\epsilon^2\nabla_\perp^2 + \mathscr{F}\left(\omega_0\eta\frac{\partial}{\partial t}\right)\right).$$

By differentiating both sides of Eq. (6) with respect to z and using the resulting expression in the last term of the right-hand side of Eq. (6), we see that Eq. (6) can be written in terms of a hierarchy of recurrence equations. This hierarchy of equations can be rewritten using continuous fractions,

$$\mathscr{O}A \equiv \left(L_{DF}^{-1}\frac{\partial}{\partial z}\right)A = \frac{ik_0}{2}\mathscr{L}_0\frac{1}{1+\frac{\mathscr{L}_0/4}{1+\frac{\mathscr{L}_0/4}{1+\cdots}}}A. \tag{7}$$

This equation can be solved exactly to give

$$\mathscr{O}A = ik_0\left(\sqrt{1+\mathscr{L}_0} - 1\right)A. \tag{8}$$

Operator $\mathscr{O} = ik_0(\sqrt{1+\mathscr{L}_0} - 1)$ in Eq. (8) can be expanded in the Taylor series in the spatial and temporal derivatives and consecutive terms can be compared with our previous approach. Using a symbolic algebra program, we expanded the expression on the RHS of Eq. (8) in the small parameter ϵ and found perfect agreement with the coefficients in Eq. (4). It can be easily verified that Eq. (8) yields the original wave equation by adding ik_0A to both sides of (8) and squaring the resulting equation.

3. Propagation equation: nonlinear susceptibility

For specificity and simplicity, let us consider a nonlinear medium with a Kerr-type nonlinear susceptibility of the form $\chi^{(3)}_{ijkl}(-\omega;\omega_1,-\omega_2,\omega_3)$.

The nonlinear source term in the wave equation can be written in terms of the SVE as follows:

$$\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}P_i^{\rm NL}(\vec{x},t)=\frac{-4\pi}{c^2(2\pi)^8}\times\int {\rm d}^3k\,{\rm d}\omega\,{\rm d}^3k_1\,{\rm d}\omega_1\,{\rm d}^3k_2\,{\rm d}\omega_2\times\,{\rm d}^3k_3\,{\rm d}\omega_3\cdot\delta(\vec{k}-\vec{k}_1+\vec{k}_2-\vec{k}_3)\times\delta(\omega-\omega_1+\omega_2-\omega_3)\cdot\exp\left[{\rm i}(\vec{k}+\vec{k}_0)\cdot\vec{x}-{\rm i}(\omega+\omega_0)t\right]\times(\omega_0+\omega)^2\cdot\chi^{(3)}_{ijkl}(-(\omega_0+\omega);\omega_0+\omega_1,-(\omega_0+\omega_2),\omega_0+\omega_3)\cdot A_j(\vec{k}_1,\omega_1)A_k^*(\vec{k}_2,\omega_2)A_l(\vec{k}_3,\omega_3). \tag{9}$$

Here we have assumed that the wavevector dependence of the nonlinear susceptibility is unimportant. If one neglects the frequency dependence of $\chi^{(3)}_{ijkl}$, then in coordinate space one finds

$$\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}P_i^{\rm NL}(\vec{x},t)=-\frac{4\pi\chi^{(3)}}{c^2}\left(\omega_0^2|A|^2A+2\omega_0\frac{\partial(|A|^2A)}{\partial t}+\frac{\partial^2(|A|^2A)}{\partial t^2}\right){\rm e}^{{\rm i}(k_0z-\omega_0t)}. \tag{10}$$

Note the presence of the two time derivative terms on the RHS of the equation. The second term on the RHS gives rise to self-steepening and the third is a higher-order term.

In order to develop an expansion to all orders of the equations of motion for a light pulse that is valid for nonlinear pulse propagation, we now introduce (in dimensionless units) a new operator $\mathscr{L}$ to replace $\mathscr{L}_0$

$$\mathscr{L}A\equiv\mathscr{L}_0A+\frac{4\pi\chi^{(3)}}{n^2(\omega_0)}\left(|A|^2A+2\eta\frac{\partial(|A|^2A)}{\partial t}+\eta^2\frac{\partial^2(|A|^2A)}{\partial t^2}\right). \tag{11}$$

$\mathscr{L}$ is z dependent through $A(x,y,z,t)$, and does not commute with $\partial/\partial z$.

With the new $\mathscr{L}A$ we cannot obtain as simple a result as Eq. (8), valid for the linear propagation of optical pulses, but we still obtain a perturbation expansion in the parameters ϵ and η. Eq. (6) is now modified and becomes

$$\mathscr{O}A\equiv\left(L_{\rm DF}^{-1}\frac{\partial}{\partial z}\right)A=\frac{{\rm i}k_0}{2}\left(\mathscr{L}+\frac{1}{k_0^2}\left(L_{\rm DF}^{-1}\frac{\partial}{\partial z}\right)^2\right)A=\frac{{\rm i}k_0}{2}\left(\mathscr{L}+\frac{1}{k_0^2}\mathscr{O}^2\right)A. \tag{12}$$

If we operate on Eq. (12) with $\mathscr{O}$ and decide the resulting equation by k_0^2 we find

$$\frac{1}{k_0^2}\mathscr{O}^2A=\left(\frac{{\rm i}}{2}\epsilon^2\frac{\partial[\mathscr{L}A(z)]}{\partial z}-{\rm O}(\epsilon^4)\right). \tag{13}$$

The RHS of Eq. (13) contains both $\partial A/\partial z$ and $\partial A^*/\partial z$ terms. To carry out the expansion we simply replace $\partial A/\partial z$ with the RHS of Eq. (12) and $\partial A^*/\partial z$ with the RHS of the complex conjugate of Eq. (12). Next, we isolate terms of different order in ϵ. This procedure is tedious due to the large number of terms that must be included, but it is mathematically rigorous. Applied to the linear case it gives results equivalent to our final result of the previous section, Eq. (8). This expansion procedure can be carried out using a symbolic mathematics program, even for anisotropic media, where the algebra can become extremely tedious. After carrying out the algebra for our case of an isotropic medium we finally obtain

$$L_{\rm DF}^{-1}\frac{\partial A}{\partial z}={\rm RHS}({\rm Eq.\,(4)})+\frac{2\pi\chi^{(3)}\omega_0}{nc}\left\{{\rm i}|A|^2A-{\rm i}\frac{\pi\chi^{(3)}}{n^2}|A|^4A+\eta\left[\left(\frac{c\beta_1}{n}-2\right)\frac{\partial(|A|^2A)}{\partial t}\right]-{\rm i}\epsilon^2\frac{1}{n^2}\left[A\frac{\partial A^*}{\partial x}\frac{\partial A}{\partial x}+A\frac{\partial A^*}{\partial y}\frac{\partial A}{\partial y}+\frac{1}{2}A^*\left(\frac{\partial A}{\partial x}\right)^2+\frac{1}{2}A^*\left(\frac{\partial A}{\partial y}\right)^2+|A|^2\frac{\partial^2A}{\partial x^2}+|A|^2\frac{\partial^2A}{\partial y^2}\right]-\epsilon^2\eta\left[\left(\frac{c\beta_1}{2n^3}-\frac{8}{n^2}\right)\frac{\partial}{\partial t}\left(\frac{\partial^2(|A|^2A)}{\partial x^2}+2|A|^2\frac{\partial^2A}{\partial x^2}-A^2\frac{\partial^2A^*}{\partial x^2}\right)+\frac{c\beta_1}{n^3}\left(2|A|^2\frac{\partial^3A}{\partial x^2\partial t}+A^2\frac{\partial^3A^*}{\partial x^2\partial t}\right)+(x\leftrightarrow y)\right]\right\}. \tag{14}$$

The first term on the RHS of this equation is the Kerr nonlinearity, the second is a higher-order Kerr nonlinear term, the third gives rise to self-steepening and is first-order in η (note that its coefficient has been modified due to the dispersion affecting the Kerr nonlinearity), the fourth is a nonlinear spatial-derivative term that is first-order in ϵ^2 and modifies the self-focusing even of beams and long duration pulses, the fifth term is a nonlinear term that is first-order in both η and ϵ^2 and affects both dispersion and diffraction. Thus, our expansion method introduces corrections leading both to temporal and spatial modifications of the pulse shape and phase. In the coordinate system moving with the group velocity of the pulse, obtained by making the transformation $t \rightarrow t - z/\beta_1$, one need only eliminate the β_1 term on the right-hand side of Eqs. (4) and (14). We have checked that the resulting equation is equivalent to the one derived by Fibich and Papanicolaou in [18].

It is trivial to rewrite Eq. (14) from dimensionless units to Gaussian units for time and space, and to rewrite the equation in SI units one simply writes time and space variables is SI units and makes the substitution

$$\chi^{(3)}(\text{Gaussian}) = \frac{(3 \times 10^4)^2}{4\pi} \chi^{(3)}\ (\text{SI})$$

or

$$\chi^{(3)}(\text{Gaussian}) = \frac{(3 \times 10^4)^2}{4\pi\epsilon_0} \chi^{(3)}\ (\text{SI}),$$

depending upon which SI convention is preferred. We have only included the first-order time-derivative nonlinear terms and we have not included higher-order terms in ϵ in Eq. (14). We can, however, easily obtain higher-order time-derivative and spatial-derivative nonlinear terms and higher-order terms to any desired order in ϵ. When all time-derivative terms are set to zero, we recover the results of Fibich and Ilan [15a,15b] for linearly polarized cw beams, when vectorial effects are neglected. When the nonlinear coupling terms are set to zero, we trivially recover the linear pulse propagation results [3]. Extensive studies of these results will be presented elsewhere.

4. Numerical example

The numerical case study presented here shows a particular example of the influence of the new terms in the wave equation (14), and is designed to demonstrate the effects resulting due to tight focusing (rather than short pulse duration). We consider a tightly focused short-duration pulse propagating in silica (SiO_2). The central wavelength is taken to be $\lambda_0 = 800$ nm, the temporal pulse duration $\tau_p = 1000$ fs, the initial spot size $\sigma_p = \lambda_0$ in x but very large stop size in y, and the

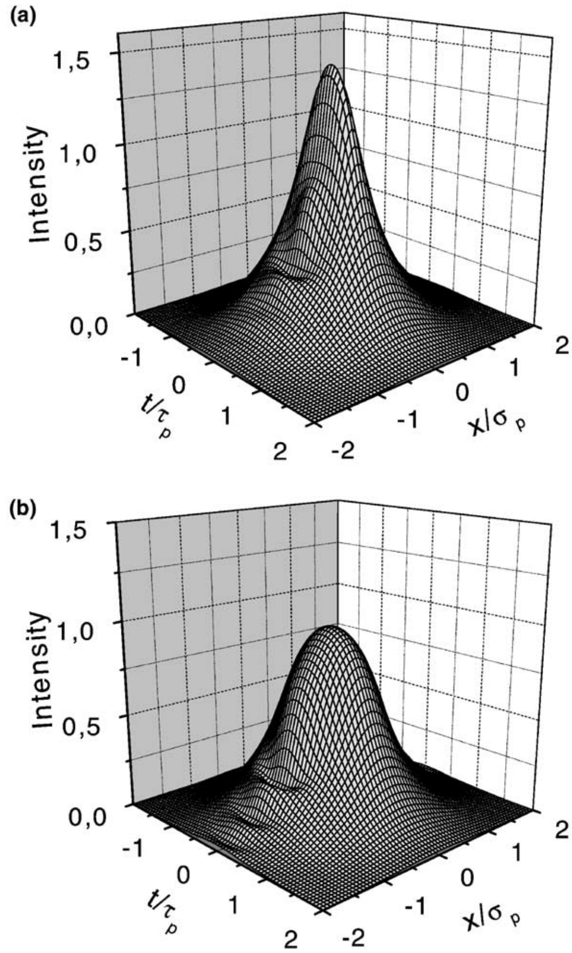

Fig. 1. $|A(x,t)|^2$ versus x/σ_p and t/τ_p after propagating a distance $z = 0.3L_{DF} = 2.2$ μm in the frame travelling with the z component of the group velocity. (a) Full nonlinearity, and (b) only the Kerr term included.

ratio of the nonlinear length to the diffraction length in x is 0.037 (so the effects of the nonlinearity are stronger than those of diffraction). Fig. 1 shows $|A(x,t)|^2$ versus x/σ_p and t/τ_p after propagating a distance $z = 0.3 \times L_{DF} = 2.2$ μm and Fig. 2 shows $|A(k_x,\omega)|^2$ versus $k_x\sigma_p$ and $\omega\tau_p$. We compare the results obtained via numerical propagation with *only* the self-phase modulation nonlinear term $((2\pi\chi^{(3)}\omega_0)/nc)\mathrm{i}|A|^2A$, with those obtained using the full nonlinearity presented in Eq. (14). We used a split-step method, with adaptive grid and step size to increase the numerical stability. The most prominent effect of the new terms shown in Fig. 1 is a spatial flattening in the region of the maximum of the pulse and the self-focusing process is slowed down. This is an effect of phase generated by the terms containing spatial derivatives of the nonlinearity in Eq. (14). Their effect in the Fourier domain is shown in Fig. 2, where the asymmetry of the ring surrounding central maximum is smooth out by the new terms. In Fig. 3 we show the instantaneous local frequency shift [11] $\delta\omega = -\partial\phi(x,t)/\partial t$. With new terms included a more vigorous phase buildup occurs at the edge of the region where the variation of the amplitude is large. These effects will be even more evident as the

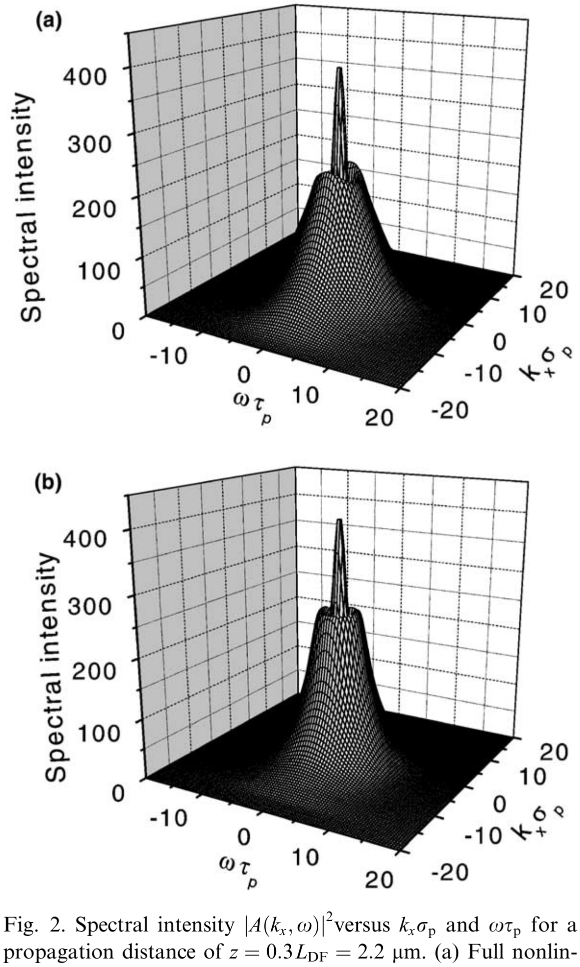

Fig. 2. Spectral intensity $|A(k_x,\omega)|^2$versus $k_x\sigma_p$ and $\omega\tau_p$ for a propagation distance of $z = 0.3L_{DF} = 2.2$ μm. (a) Full nonlinearity, and (b) only the Kerr term included.

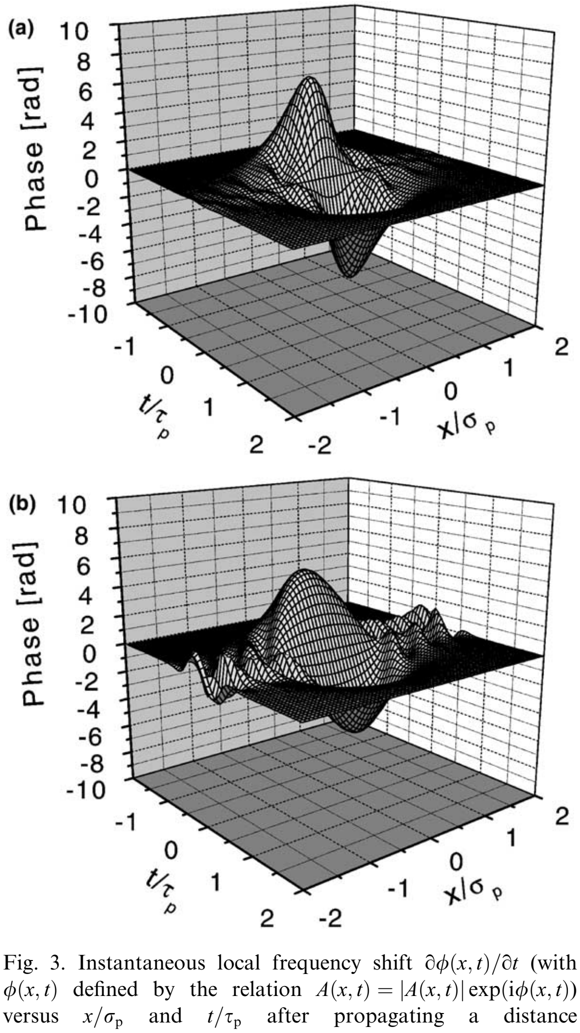

Fig. 3. Instantaneous local frequency shift $\partial\phi(x,t)/\partial t$ (with $\phi(x,t)$ defined by the relation $A(x,t) = |A(x,t)|\exp(\mathrm{i}\phi(x,t))$ versus x/σ_p and t/τ_p after propagating a distance $z = 0.3L_{DF} = 2.2$ μm in the frame travelling with the z component of the group velocity. (a) Full nonlinearity, and (b) only the Kerr term included.

pulse duration increases, since the self-focusing process becomes stronger.

Notice that since we consider a relatively long temporal duration pulse of 1000 fs, the effects of the terms including derivatives with respect time of the nonlinearity do not significantly influence the dynamics. They become important for shorter pulses. The most important modification of the temporal derivative nonlinear terms introduced by our expansion is the modification of the coefficient in front of the self-steepening term $\partial(|A|^2A)/\partial t$. The coefficient obtained from the Kerr nonlinearity (equal 2 in our dimensionless units) is replaced by $((c\beta_1/n) - 2)$ [12a–d].

5. Conclusions

We have presented a mathematically rigorous derivation of the nonlinear pulse propagation equation, free from slowly varying and paraxial approximations. This generalizes both the previous formulation of linear optical pulse propagation to the nonlinear propagation regime, and the cw nonlinear regime to the pulsed regime. A numerical example that shows the effects of the of spatial-derivative nonlinear coupling terms for propagation of a tightly focused picosecond pulse propagating in silica was presented. The symbolic mathematics program to calculate the coefficients in the nonlinear propagation equation is available upon request.

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