Simplified field wave equations for the nonlinear propagation of extremely short light pulses

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(Received 1 December 2001; published 25 July 2002)

We suggest a method to derive the nonlinear wave equations suitable for describing the propagation of light pulses as short as two optical cycles in transparent nonlinear optical media. The equations are suitable for efficient numerical simulation of the propagation of extremely short pulses without the need to resort to any type of envelope approximation, although they contain these as a limiting case. We demonstrate the power of this approach by modeling some recent experiments in which ultrabroadband radiation was generated upon propagating 150 fs duration 390 nm pulses in a deuterium-gas-filled hollow-core waveguide. The calculated spectrum agrees well with the measured spectrum.

DOI: 10.1103/PhysRevA.66.013811

PACS number(s): 42.65.Re

I. INTRODUCTION

Since the initial demonstrations of pulsed lasers, significant effort has been focused on generating shorter and shorter pulses of light from them. Recently, pulses of 4.5 fs duration have been created in the laboratory, corresponding to about 2-3 periods of oscillation of the electromagnetic field in the near-ir region of the spectrum [1-3]. The measurement of the pulse shape and even the pulse duration in this regime is itself a difficult technical problem [4]. For the purposes of this work we will label pulses containing approximately ten or fewer cycles extremely short pulses. We distinguish them from pico- and femtosecond optical pulses containing many field oscillations, which are classified as ultrashort. The defining characteristic of extreme brevity is the number of oscillations of the field, not the pulse duration per se. Using our definition, both subpicosecond ir pulses [5,6] and attosecond uv pulses [7,8] are also extremely short.

The usual approximations made in deriving equations to describe the interaction of such brief pulses with matter must be reexamined. This is because of both the inadequacy of the standard approaches to the slowly varying envelope approximation [9,10] and the important role played by space-time coupling even in linear propagation problems. Although it is mathematically possible to introduce the concept of an envelope for a pulse consisting of one or two field oscillations, the notion of a slowly varying pulse envelope becomes less fruitful. For this reason a great deal of attention has been paid in the past few years to modifying the envelope approximation (see, for example, [11-15]) and to developing another theoretical methods for analysis of laser pulse field propagation and dynamics in optical media [16-25].

In this paper we formulate principles for constructing nonlinear wave equations describing directly the dynamics of the electric field (but not the envelope) of extremely short pulses in transparent optical media. We show that our equations automatically include the known equations for the envelope as a special case. The resulting system is used to model experimental effects that have been demonstrated to occur when extremely short pulses propagate in transparent nonlinear media. Some outstanding theoretical problems in the optics of extremely short pulses are discussed.

II. WAVE EQUATIONS FOR EXTREMELY SHORT PULSES

Extremely short light pulses are usually generated by means of nonlinear optics [1-3,7,8]. Such pulses are amenable to description within the framework of classical electromagnetism. Classical theory can also be used for the description of extremely short pulses that are generated by a laser interacting with free electrons [6]. For the important case of pulse propagation in a nonmagnetic dielectric medium, Maxwell's equations are easily reduced to the form of [26]

$$\nabla \times \nabla \times \mathbf{E} + \mu_0 \frac{\partial^2 \mathbf{D}}{\partial t^2} = \mathbf{0},\tag{1}$$

where **E** is the electric field of the light wave, the electric induction **D** characterizes the response of the medium, which is usually nonlinear in the strong field of the extremely short pulse, and μ_0 is the magnetic permeability of vacuum. The first term on the left hand side of Eq. (1) accounts for diffraction and the second for dispersion and the nonlinear character of the light-matter interaction.

Even though it is well known that the theoretical analysis of the nonlinear dynamics of light fields extremely localized in time and space is fully contained in the solutions of Eq. (1) it is important to note that such solutions have not been investigated fully even in the particular case of the propagation of a monochromatic wave through a transparent isotropic medium with a cubic nonlinearity. This problem has been considered only within the paraxial approximation, where Eq. (1) becomes a cubic Schrödinger equation [27]. But in this approximation the theory predicts that the beam will collapse, in which case the propagation becomes highly nonparaxial. Theoretical research into nonlinear light field dynamics is still a very active area for the case when the beam diameter is comparable to the wavelength of the radiation even for monochromatic radiation [27,28]. The problem of self-focusing of extremely short pulse in the highly nonparaxial regime remains to be solved.

At present, theoretical studies of extremely short pulse self-action are most fully developed for plane waves, i.e., in the (1+1)-dimensional approximation. The approximation is adequate for the fundamental mode in guiding structures, such as hollow-core waveguides, where it is possible to neglect the existence of a longitudinal field component, and when the input power is below the critical power for selffocusing. In this approximation the wave equation (1) can be transformed to

$$\frac{\partial^2 \mathbf{E}}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}_1}{\partial t^2} + \mu_0 \frac{\partial^2 \mathbf{P}_{nl}}{\partial t^2}, \qquad (2)$$

where z is the spatial coordinate in the direction of propagation, \mathbf{P}_{l} is the linear part of the medium polarization, and \mathbf{P}_{nl} is the nonlinear component.

A. Principles of constructing equations for extremely short pulses

Any mathematical model of the self-action of an extremely short pulse in an optical medium must properly account for the linear and nonlinear dispersion of the medium over a wide spectral range. (For example, in supercontinuum generation, the spectral width of the generated light covers more than one octave.) We begin by modifying Eq. (2) to describe the propagation of ultrabroadband spectrum pulses in a linear medium ($\mathbf{P}_{nl}=\mathbf{0}$) [29,30]. The dependence of the linear index of refraction *n* of an isotropic optical medium on the optical frequency ω within the transparency region can be described by the following equation [31]:

$$n^{2}(\omega) = N_{0}^{2} + 2cN_{0}a\omega^{2} + 2cN_{0}a_{1}\omega^{4} + \dots - 2cN_{0}b\omega^{-2}$$
$$-2cN_{0}b_{1}\omega^{-4}\cdots, \qquad (3)$$

where N_0 , a, a_1 , ..., b, b_1 , ... are the empirical dispersion constants of the medium. This provides the required precision in the range of transparency of the medium. The dispersion relation (3) produces a wave equation of the form

$$\frac{\partial^{2}\mathbf{E}}{\partial z^{2}} - \frac{N_{0}^{2}}{c^{2}} \frac{\partial^{2}\mathbf{E}}{\partial t^{2}} = -\frac{2N_{0}}{c} a\frac{\partial^{4}\mathbf{E}}{\partial t^{4}} + \frac{2N_{0}}{c} a_{1}\frac{\partial^{6}\mathbf{E}}{\partial t^{6}} - \cdots + \frac{2N_{0}}{c} b\mathbf{E} - \frac{2N_{0}}{c} b_{1}\int_{-\infty}^{t} dt' \int_{-\infty}^{t'} \mathbf{E} dt'' + \cdots$$

$$(4)$$

This statement is satisfied by finding the particular solution to Eq. (4) for a monochromatic wave

$$\mathbf{E} = \frac{1}{2} \boldsymbol{\mathcal{E}}_{\boldsymbol{\omega}} e^{i(kz - \omega t)} + \text{c.c.}, \qquad (5)$$

where \mathcal{E}_{ω} is the amplitude of a spectral component of the radiation and $k(\omega)$ is the wave number. It can be easily shown that Eq. (5) is a solution of Eq. (4) if the refractive index $n(\omega) = k(\omega)c/\omega$ depends on frequency according to relation (3).

Equation (4) describes the propagation of pulses along the z axis in both forward and backward directions. If we are

interested in the evolution of the electric field of light pulses propagating in one (for example positive) direction of the z axis, it is expedient to transform the equation to the new variables z'=z and $\tau=t-(N_0/c)z$, leading to the form

$$\frac{\partial^{2}\mathbf{E}}{\partial z'^{2}} - \frac{2N_{0}}{c} \frac{\partial^{2}\mathbf{E}}{\partial z' \partial \tau} = -\frac{2N_{0}}{c} a \frac{\partial^{4}\mathbf{E}}{\partial \tau^{4}} + \frac{2N_{0}}{c} a_{1} \frac{\partial^{6}\mathbf{E}}{\partial \tau^{6}} - \cdots + \frac{2N_{0}}{c} b\mathbf{E} - \frac{2N_{0}}{c} \times b_{1} \int_{-\infty}^{\tau} d\tau' \int_{-\infty}^{\tau'} \mathbf{E} d\tau'' + \cdots$$
(6)

Using the approximation of a slowly varying temporal profile of the pulse field [26], which is often used in the theory of acoustic waves (i.e., assuming that the variations of the pulse field profile are small at a distance commensurate with the central wavelength), we can neglect the second derivative term $\partial^2 E/\partial z'^2$. Then, integrating Eq. (6) over the time τ , we obtain the wave equation in the form

$$\frac{\partial \mathbf{E}}{\partial z'} - a \frac{\partial^3 \mathbf{E}}{\partial \tau^3} + a_1 \frac{\partial^5 \mathbf{E}}{\partial \tau^5} - \dots + b \int_{-\infty}^{\tau} \mathbf{E} d\tau'$$
$$- b_1 \int_{-\infty}^{\tau} d\tau' \int_{-\infty}^{\tau'} d\tau'' \int_{-\infty}^{\tau''} \mathbf{E} d\tau''' + \dots = \mathbf{0}.$$
(7)

The wave equation (7), which is truncated with respect to the first z' derivative, is equivalent to the one derived using the dispersion relation

$$n(\omega) = N_0 + ca\omega^2 + ca_1\omega^4 + \dots - cb\omega^{-2} - cb_1\omega^{-4} - \dots$$
(8)

Thus, in our case the approximation of a slowly varying profile is associated with replacement of $n^2 - N_0^2$ in the dispersion relation (3) by $2N_0(n-N_0)$. Since Eq. (8) also describes the dispersion of the refractive index of optical materials in their transparency range with good accuracy [31] the approximation of a slowly varying wave profile, which is fruitful, for example, in acoustics, is also fully justified in the optics of transparent media.

It should be noted that Eq. (7) could be used to describe not only the propagation of a transversely homogeneous plane wave, but also that of a spatially transversely inhomogeneous mode in a hollow waveguide. The waveguide contribution can be introduced into Eq. (7) by addition of a new coefficient $b' = cu^2/(2N_0r^2)$ [24] to the coefficient *b* associated with the matter dispersion. Here *r* is the radius of the hollow core and *u* is a mode-related constant (one of the roots of the Bessel function). For the minimum loss TE₀₁ mode u = 2.405.

The waveguide dispersion can significantly modify the dispersion compared to that of the dilute gas in the core. For instance, if the capillary is filled with a noble gas with normal dispersion in the relevant spectral region, then the waveguide structure can exhibit anomalous dispersion in some range of the spectrum. Comparing Eqs. (2) and (4) we can

easily see that the generalization of Eq. (4) to the case of a nonlinear medium is an equation of the form

$$\frac{\partial^{2}\mathbf{E}}{\partial z^{2}} - \frac{N_{0}^{2}}{c^{2}} \frac{\partial^{2}\mathbf{E}}{\partial t^{2}} = -\frac{2N_{0}}{c} a\frac{\partial^{4}\mathbf{E}}{\partial t^{4}} + \frac{2N_{0}}{c} a_{1}\frac{\partial^{6}\mathbf{E}}{\partial t^{6}} - \cdots + \frac{2N_{0}}{c} b\mathbf{E} - \frac{2N_{0}}{c} b_{1}\int_{-\infty}^{t} dt' \int_{-\infty}^{t'} \mathbf{E} dt'' + \cdots + \mu_{0}\frac{\partial^{2}\mathbf{P}_{\mathrm{nl}}}{\partial t^{2}}.$$
(9)

If the nonlinearity of the polarization response does not lead to the generation of radiation propagating in the backward (negative z') direction, then applying the approximation of a slowly varying profile (which physically means the approximation of forward propagation, see Sec. II D) to Eq. (9) we obtain the truncated nonlinear wave equation

$$\frac{\partial \mathbf{E}}{\partial z'} - a \frac{\partial^3 \mathbf{E}}{\partial \tau^3} + a_1 \frac{\partial^5 \mathbf{E}}{\partial \tau^5} - \dots + b \int_{-\infty}^{\tau} \mathbf{E} d\tau'$$
$$- b_1 \int_{-\infty}^{\tau} d\tau' \int_{-\infty}^{\tau'} d\tau'' \int_{-\infty}^{\tau''} \mathbf{E} d\tau''' + \dots + \frac{\mu_0 c}{2N_0} \frac{\partial \mathbf{P}_{\mathrm{nl}}}{\partial \tau} = \mathbf{0}.$$
(10)

An analysis of the solutions of the system of the wave equation (10) simultaneously with the equations for the nonlinear polarization response \mathbf{P}_{nl} determines the features of the propagation of extremely short pulses as a function of their initial energy, polarization, and temporal and spectral profiles.

B. Nonlinear equations for interaction of extremely short pulses with matter

The next requirement for a description of the nonlinear part of the polarization response is an account of its dispersive properties. The material equations for isotropic dielectric media were derived from a density matrix description of the matter in [32,33]. The major mechanisms leading to an optical nonlinearity \mathbf{P}_{nl} for an extremely short pulse are the electronic and electronic-vibrational polarizabilities. Each of these rapidly responding mechanisms was described by a three-level nonresonant approximation for the interaction of the optical field with a molecule. It was demonstrated in [34] that this approximation is the minimum necessary for an adequate description of the dispersion of a nonlinear index of refraction of a dielectric medium in the range of transparency. This model was based on the idea of matter as two parametrically excited bound nonlinear oscillators. In this case, as in the Lorentz model for linear dispersion, the dependence of the nonlinear refraction index of an optical medium on the frequency of a light wave will have the same form as in quantum theory. It was shown that the nonlinear polarization response of a dielectric medium \mathbf{P}_{nl} can be described by a system of matter equations:

$$\frac{\partial^2 \mathbf{P}_{nl}}{\partial t^2} + \frac{2}{T_{e1}} \frac{\partial \mathbf{P}_{nl}}{\partial t} + \omega_{e1}^2 \mathbf{P}_{nl} = \varepsilon_0 (R_e + R_\nu) \mathbf{E},$$
$$\frac{\partial^2 R_e}{\partial t^2} + \frac{2}{T_{e2}} \frac{\partial R_e}{\partial t} + \omega_{e2}^2 R_e = \gamma_e E^2, \tag{11}$$
$$\frac{\partial^2 R_\nu}{\partial t^2} + \frac{2}{T_\nu} \frac{\partial R_\nu}{\partial t} + \omega_\nu^2 R_\nu = \gamma_\nu E^2,$$

where R_e and R_v describe the nonlinear parametric connection between the pulse electric field and the polarization of the medium. The dynamic parameter R_e is responsible for the electronic nonlinearity, and R_v for the Raman or electronic-vibrational contribution. The phenomenological parameters of the medium T_{e1} , ω_{e1} , T_{e2} , ω_{e2} , γ_e and T_v , ω_v , γ_v characterize the respective dispersions of the nonlinear polarization responses, and ε_0 is the dielectric permeability of vacuum.

Thus the theoretical problem of propagation of extremely short optical pulses, whose spectrum is in the transparency range of the dielectric medium, can be reduced to a study of solutions of the wave equation (10) with the matter equations (11). The model (10),(11) describes phase self- and cross modulation, stimulated Raman scattering, high-harmonic generation, and other nonlinear phenomena, which for extremely short pulses cannot be observed separately.

In conclusion we remark that even in gaseous media the field of an extremely short pulse in experiments can be so strong that a plasma of free electrons may be created. The resulting plasma nonlinearity will also influence the pulse evolution [8,35]. The system of equations in (11) does not include this effect and in this case they should be supplemented by the appropriate equations.

C. Primary equation for extremely short pulses and its modifications

In many practical situations the model of the self-action of extremely short pulses in an optical medium (10),(11) can be simplified into a single equation. For the analysis of linear propagation of extremely short pulses by Eqs. (7) it is necessary to describe the refractive index using only the first two and the fourth terms of Eq. (8). For example, for fused silica, Eq. (8) with these three terms describes the dependence of the linear refraction index to an accuracy of three decimal places in the spectral range from 460 nm to 2500 nm (with $N_0 = 1.4508$, $a = 2.7401 \times 10^{-42}$ s³ m⁻¹, b = 3.9437 $\times 10^{19}$ s⁻¹ m⁻¹). Thus Eq. (8) describes a considerable part of the region of normal dispersion of this material as well as the anomalous group dispersion in the near-ir region. Note that the high-frequency end of the dispersion range is limited by the onset of two-photon absorption. Due to the nonresonant character of the electronic nonlinearity in fused silica in this spectral range, it is possible, as a first approximation, to neglect the dispersion of the nonlinear refractive index [33]. For extremely short pulses we can also ignore Raman effects [20,33], since the response time of the vibrational motion, the phonon oscillation period, is rarely less than 100 fs or so. Hence, for the propagation of linearly polarized extremely short pulses with spectra in the transparency range of a wideband-gap isotropic dielectric medium, the matter equations (11) can usually be simplified to the relation $P_{\rm nl}$ = $(\varepsilon_0 \gamma_e / \omega_{e1}^2 \omega_{e2}^2) E^3$, and the set of equations (10),(11) to

$$\frac{\partial E}{\partial z} - a \frac{\partial^3 E}{\partial \tau^3} + b \int_{-\infty}^{\tau} E d\tau' + g E^2 \frac{\partial E}{\partial \tau} = 0, \qquad (12)$$

where $g = (3/2cN_0)(\gamma_e/\omega_{e1}^2\omega_{e2}^2)$. Equation (12) was derived earlier in [20] by direct reduction of the wave equation and matter oscillator equations accounting for the linear electronic and vibration and nonlinear electronic polarizations.

This equation plays the same role for extremely short pulses as the cubic Schrödinger equation [9,10] does in the nonlinear optics of ultrashort pulses (pico- and femtosecond pulses with many field oscillations) and describes all major physical phenomena relevant to the field dynamics in a dispersive dielectric medium with an instantaneous nonlinearity.

The Schrödinger equation describing the self-action of ultrashort pulses has been modified in different ways for various media, and to include the polarization, harmonic generation, and other features of light pulse propagation [9,10]. The wide range of experimentally attainable parameters of extremely short pulses makes it necessary to discuss similar modifications of the nonlinear wave equation (12).

In the case of extremely short pulses with arbitrary polarization, Eq. (12) becomes [36,37]

$$\frac{\partial \mathbf{E}}{\partial z} - a \frac{\partial^3 \mathbf{E}}{\partial \tau^3} + b \int_{-\infty}^{\tau} \mathbf{E} d\tau' + g(\mathbf{E}, \mathbf{E}) \frac{\partial \mathbf{E}}{\partial \tau} + h \mathbf{E} \times \left(\mathbf{E} \times \frac{\partial \mathbf{E}}{\partial \tau} \right) = \mathbf{0},$$
(13)

where *h* and *g* characterize the magnitude of the instantaneous nonlinearity of the medium polarization response. In [25,38] a similar vector equation is deduced for the field in the approximation of a two-level medium with b=0 and g < 0. However, for the case of a transparent medium being considered here, we have g > 0 [33] (at least if the spectrum of the extremely short pulse does not cover a two-photon absorptive resonance of the medium [39]), and for the analysis of polarization field dynamics it is necessary to solve Eq. (13).

If it is necessary to take into account the effect of electronic-vibrational nonlinearity on the pulse evolution, then Eqs. (10) and (11) can be reduced to the form

$$\frac{\partial E}{\partial z} - a \frac{\partial^3 E}{\partial \tau^3} + b \int_{-\infty}^{\tau} E d\tau' + g E^2 \frac{\partial E}{\partial \tau} + q \frac{\partial}{\partial \tau} (R_{\nu} E) = 0,$$
$$\frac{\partial^2 R_{\nu}}{\partial \tau^2} + \frac{2}{T_{\nu}} \frac{\partial R_{\nu}}{\partial \tau} + \omega_{\nu}^2 R_{\nu} = \gamma_{\nu} E^2, \qquad (14)$$

where $q = 1/2cN_0\omega_{e1}^2$.

In addition to the features described by Eqs. (12), Eqs. (14) also incorporate the stimulated Raman effect. The selfaction of extremely short pulses in Raman-active media was analyzed in Refs. [40,41] by including additional terms describing the population dynamics of the vibration levels. However, the linear dispersion of the medium due to electronic resonances was not taken into account.

In the presence of a high-frequency spectral component in the spectrum of an extremely short pulse which is resonant with a two-photon electronic transition, Eqs. (12) can be modified and written in the following form:

$$\frac{\partial E}{\partial z} - a \frac{\partial^3 E}{\partial \tau^3} + b \int_{-\infty}^{\tau} E d\tau' + g' E^2 \frac{\partial E}{\partial \tau} + q \frac{\partial}{\partial \tau} (R_e E) = 0,$$
$$\frac{\partial^2 R_e}{\partial \tau^2} + \frac{2}{T_{e2}} \frac{\partial R_e}{\partial \tau} + \omega_{e2}^2 R_e = \gamma_e E^2, \tag{15}$$

where g' describes the instantaneous contribution of excited electronic states that do not have a two-photon resonance.

It is straightforward to take into account linear absorption of the medium in Eq. (12). In this case the equation contains additional terms describing the damping of the electric field, quantified by the parameters Γ_0 , Γ_1 , and Γ_2 :

$$\frac{\partial E}{\partial z} + \Gamma_0 E - a \frac{\partial^3 E}{\partial \tau^3} + b \int_{-\infty}^{\tau} E d\tau' + g E^2 \frac{\partial E}{\partial \tau} - \Gamma_1 \frac{\partial^2 E}{\partial \tau^2} - \Gamma_2 \int_{-\infty}^{\tau} d\tau' \int_{-\infty}^{\tau'} E d\tau'' = 0.$$
(16)

By making a comparison with the usual formalism for the complex index of refraction $n' = n + i\kappa$ [considering *E* in the form of Eq. (5)] these terms can be identified with the spectral dependence of the linear absorption of the medium via the equation $\kappa(\omega) = c(\Gamma_0/\omega + \Gamma_1\omega + \Gamma_2/\omega^3)$.

The diffraction of extremely short pulses can be described in the paraxial limit by a modified version of Eq. (12) [20]:

$$\frac{\partial E}{\partial z} - a \frac{\partial^3 E}{\partial \tau^3} + b \int_{-\infty}^{\tau} E d\tau' + g E^2 \frac{\partial E}{\partial \tau} = \frac{c}{2N_0} \Delta_{\perp} \int_{-\infty}^{\tau} E d\tau',$$
(17)

where $\Delta_{\perp} = \partial^2 / \partial x^2 + \partial^2 / \partial y^2$ is the transverse Laplacian. As far as we are aware, Eq. (17) was first suggested in the context of space-time coupling in free space (a=b=g=0) in Ref. [42].

If we suppose that the parameters N_0 , a, b, and g in Eq. (17) are dependent on the spatial coordinate then weak inhomogeneity of a medium can be taken into account. This would encompass, for example, waveguiding in a glass fiber.

We emphasize that in the limit of "long" pulses with durations of more than about ten cycles Eq. (12) can be transformed into a modified form of the cubic nonlinear Schrödinger equation when higher dispersion orders are accounted for [20]. That is, Eq. (12) includes the commonly accepted fundamental equation describing the nonlinear optics of ultrashort pulses as a special case. In [36] the corresponding envelope equations are derived from Eqs. (13).

In fact, any of the above equations (12)-(17) can be transformed to the equivalent equations for \mathcal{E} by using the substi-

tution (5). For example, by taking a specific frequency ω in Eq. (5) as the mean frequency of the input pulse spectrum it is possible to label \mathcal{E} as an "envelope" even for an extremely short pulse [11]. This makes a direct connection to the statement of Ref. [11] that the dynamics of extremely short pulses can be considered using traditional analytical methods for the propagation of the pulse envelope. However, the equations for the "envelope" \mathcal{E} in the case of extremely short pulses become much more cumbersome than the equation for the field *E*. For example, the self-action of extremely short pulses in media with a normal group dispersion and instant cubic nonlinearity (in inertial gases with nonresonant electronic interactions) is described compactly by a modified Korteweg–de Vries equation [43]:

$$\frac{\partial E}{\partial z} - a \frac{\partial^3 E}{\partial \tau^3} + g E^2 \frac{\partial E}{\partial \tau} = 0.$$
(18)

However, the equation for the "envelope" \mathcal{E} obtained by substituting Eq. (5) in Eq. (18) is a cumbersome modified nonlinear Schrödinger equation with additional terms describing the generation of new harmonics:

$$\frac{\partial \mathcal{E}}{\partial z} + \frac{1}{V} \frac{\partial \mathcal{E}}{\partial t} + i \frac{\alpha_2}{2} \frac{\partial^2 \mathcal{E}}{\partial t^2} - \frac{\alpha_3}{6} \frac{\partial^3 \mathcal{E}}{\partial t^3} - i\beta_1 |\mathcal{E}|^2 \mathcal{E} + \beta_2 \frac{\partial}{\partial t} (|\mathcal{E}|^2 \mathcal{E}) - i\beta_1 \mathcal{E}^3 \exp[2i(k_0 z - \omega_0 t)] + \beta_2 \mathcal{E}^2 \frac{\partial \mathcal{E}}{\partial t} \times \exp[2i(k_0 z - \omega_0 t)] = 0,$$
(19)

where $V = (\partial k/\partial \omega)_{\omega_0}^{-1}$, $\alpha_2 = (\partial^2 k/\partial \omega^2)_{\omega_0}$, $\alpha_3 = (\partial^3 k/\partial \omega^3)_{\omega_0}$, $k = (N_0/c)\omega + a\omega^3$, $k_0 = \omega_0 N_0/c$, $\beta_1 = 3g\omega_0/4$, $\beta_2 = 3g/4$, and ω_0 is the central frequency of input radiation.

Equation (18) "automatically" describes generation of new harmonics via self-phase- and cross-phase modulation. To describe the phenomena using the envelope concept, one should generalize Eq. (19) [omitting the exponential terms in Eq. (19)] and write down additional equations characterizing the evolution and interactions of the harmonics.

The concept of the pulse field envelope can be inconvenient not only for extremely short pulses but also for longer pulses. For instance, it is not useful for the description of self-action in Raman-active media when propagation is accompanied by the generation of a nonuniform spectral supercontinuum. In Sec. III B we will show that Eqs. (14) adequately describe these nonlinear effects.

D. Spectral equations for extremely short pulses

Strong changes in the spectral structure of extremely short pulses can occur when they propagate in nonlinear media (see, for example, the effects described in Sec. III B). It therefore seems natural to present spectral analogs of the time-domain partial differential equations. In addition, by analyzing the dynamics of the pulse spectrum $\mathbf{G}(\omega) = \int_{-\infty}^{+\infty} \mathbf{E}(t) e^{-i\omega t} dt$, we will gain further insight into the approximations made in deriving the temporal equations. For simplicity we restrict ourselves to consideration of the spectral dynamics of linearly polarized radiation in the (1+1)-dimensional approximation.

The linear equation (4) has a simple spectral analog

$$\frac{\omega^2 G}{\partial z^2} + \frac{\omega^2 n^2}{c^2} G = 0,$$
 (20)

where n^2 is defined by Eq. (3). It is important to note that almost any dispersion formula for a specific medium can be used in Eq. (20) instead of the expansion of $n^2(\omega)$ given in Eq. (3).

The solution to Eq. (20) is

$$G(\omega,z) = C_1(\omega)e^{-i[\omega n(\omega)/c]z} + C_2(\omega)e^{i[\omega n(\omega)/c]z}.$$
 (21)

The first term in Eq. (21) describes radiation propagating in the positive z direction; the second term is for the opposite direction. The unidirectional propagation of radiation (i.e., for $C_2=0$) is described by the equation

$$\frac{\partial G}{\partial z} + i \frac{\omega n}{c} G = 0, \qquad (22)$$

which is an analog of the field equation (7) but written in a fixed rather than a moving frame. Here $n(\omega)$ is given by Eq. (8).

It is clear that the solution to the reduced equation (22) is simultaneously an exact particular solution to Eq. (20). Hence, the physical meaning of the reduction of Eq. (20) to Eq. (22) retaining only the lowest order derivatives in z is that we are dealing only with unidirectional propagation. The slowness of the variation of the field profile is not a necessary condition of the formulation.

The spectral analog of Eq. (9) with the simplest nonlinear polarization response $P_{nl} = \varepsilon_0 (\gamma_e / \omega_{e1}^2 \omega_{e2}^2) E^3 = \varepsilon_0 \chi E^3$, where χ is a cubic nonlinear susceptibility, is

$$\frac{\partial^2 G}{\partial z^2} + \frac{\omega^2 n(\omega)^2}{c^2} G + \frac{\chi \omega^2}{4\pi^2 c^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} G(\omega - \alpha) \times G(\alpha - \beta) G(\beta) d\alpha d\beta = 0.$$
(23)

We can derive a reduced analog of Eq. (23) in a similar manner to the transition from Eq. (20) to Eq. (22) using the technique suggested in [28]. This yields

$$\frac{\partial G}{\partial z} + i \frac{\omega n(\omega)}{c} G + i \frac{\chi \omega^2}{4\pi^2 c} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{G(\omega - \alpha)G(\alpha - \beta)G(\beta)}{\omega n(\omega) + (\omega - \alpha)n(\omega - \alpha) + (\alpha - \beta)n(\alpha - \beta) + \beta n(\beta)} d\alpha d\beta = 0.$$
(24)

Unidirectional solutions to Eq. (24) are the particular solutions to Eq. (23). According to the method of Ref. [28] the reduction is based only on the assumption of the smallness of the third term of Eq. (23) compared to the second one. That is, we assume that the nonlinear addition to the refractive index is much less than the total linear refractive index. In practice, this condition is usually met.

It is easy to derive generalizations of the spectral equations (23) and (24) for more complex polarizations.

For a linear refractive index of the form of Eq. (8) using the N_0 , a, and b coefficients only and under the assumption of weak linear dispersion, Eq. (24) becomes

$$\frac{\partial G}{\partial z} + i \left(N_0 \frac{\omega}{c} + a \,\omega^3 - \frac{b}{\omega} \right) G + i \frac{g \,\omega}{12 \pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} G(\omega - \alpha) \\ \times G(\alpha - \beta) G(\beta) d\alpha d\beta = 0, \qquad (25)$$

where $g = 3\chi/2cN_0$. Equation (25) is the spectral analog of Eq. (12).

We emphasize that Eq. (24) is more general than Eqs. (12) and (25) because it does not impose a restriction on media with weak linear dispersion.

Finally, we note that the spectral approach has some wellknown advantages and disadvantages in comparison with the temporal approach. The nonlinear convolution in Eq. (25) is more difficult to compute than the nonlinear term in Eq. (12). But the description of the dispersion of the refraction index is easier in the spectral approach. The field analog of Eq. (24) is simple if the dispersion dependence n(w) is a series whose zero-order term is the largest. If this dependence is more complicated, for example, in the form of the Sellmeier formula, the temporal analog of Eq. (24) is either a complex integro-differential equation which can be derived from Eq. (24) by inverse Fourier transform or a coupled set of field and oscillation equations for the medium polarization [17,33,44]. However, the conclusion of Oughstun and Xiao [45] that the Sellmeier formulas are incorrectly approximated by an expansion into a Taylor series in the vicinity of some fixed reference frequency does not imply that they cannot be fitted by series of the form of Eq. (8) with empirically chosen factors (see the description of the linear dispersion of fused silica in series form in Sec. II C).

In Sec. III B we simulate the generation of supercontinuum radiation in a gas-filled hollow-core fiber. We use the field rather than the more general spectral equations, because the dispersion of the refractive index is weak and the series (8) required only a few teams.

E. Numerical modeling

At present there is no known analytical procedure to solve Eq. (12) and its variants. These equations must therefore be solved numerically, but are already cast in a form that is appropriate for standard methods. In our computations we use the split-step Fourier transform technique [9] because it combines the advantages of both field and spectral propagation methods. We calculate the effect of linear terms in the frequency domain, which allows us to incorporate explicitly any dispersion relation for the propagation constant $k(\omega)$ that satisfies the unidirectional approximation. A Crank-Nicholson method is used to calculate the effects of the non-linearities in the time domain. The use of a fast Fourier transform makes the method computationally efficient.

The similarity of these methods to those used in the conventional envelope approaches belies the fact that the reduced field equations lead to a five- to ten-fold increase in accuracy for the same computational effort (comparing the results for both approaches for supercontinuum generation in hollow-core fibers). Even so, the (1+1)-dimensional modeling of spectral supercontinuum generation by a few-cycle optical pulse takes from several minutes to half an hour on a standard microcomputer.

F. Experimental measurements

The electric field is the fundamental entity that enters the propagation equations derived above. Even in the case of linear propagation, knowledge of the temporal intensity of two individual pulses is not sufficient to allow any inference of the temporal intensity of their combination. Moreover, the presence of space-time coupling demands that measurements are available for the field rather than the temporal or spectral intensity. Because the equations are nonlinear, this becomes critical, since even small changes in the input pulse shape may affect the nonlinear propagation dramatically. For this reason it is important to employ methods for complete pulse characterization to compare with the output of the numerical models. There are now several methods that have been demonstrated to be effective in measuring the electric fields of extremely short optical pulses, in the regime of two cycles or so. One of these, frequency-resolved optical gating (FROG) [46] measures the spectrum of the intensity autocorrelation function of the pulse, and uses an iterative deconvolution algorithm to extract the field [47]. Geometries for use with very broadband pulses have been developed by several groups [48,49].

Another method that provides the same information is spectral interferometry for direct electric field reconstruction (SPIDER) [50]. This technique measures the spectral interferogram of the input pulse and its frequency-shifted replica (generated by means of nonlinear optics). The spectral phase can be extracted from this interferogram using standard noniterative algorithms that enable rapid reconstruction of the pulse shape [51,52]. The apparatus is easily adapted for use with extremely short duration pulses [53].

To date there have been very few experimental studies of the propagation of short pulses in nonlinear media that provide the required detail needed for comparison with simulation [54-56], and this remains an area ripe for study.

III. THE ELECTRICAL FIELD DYNAMICS OF EXTREMELY SHORT PULSES

Using the wave equations derived above we consider in this section some important features of the propagation dynamics of extremely short pulses in optical media under both linear and nonlinear conditions.

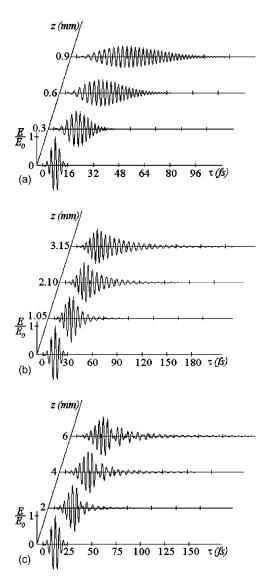


FIG. 1. The linear temporal broadening of an extremely short pulse with the spectrum in the range of (a) normal; (b) anomalous; (c) zero dispersion of fused silica. *E* is the pulse electrical field and E_0 is its maximum at the medium input (z=0).

A. Dispersive broadening of extremely short pulses

The nonlinear term in the wave equation (12) can be neglected for weak optical fields. The equation reduces to

$$\frac{\partial E}{\partial z} - a \frac{\partial^3 E}{\partial \tau^3} + b \int_{-\infty}^{\tau} E d\tau' = 0.$$
 (26)

Solutions to Eq. (26) demonstrating dispersive broadening of extremely short input pulses are shown in Fig. 1. The input pulse profile is given by

$$E(0,\tau) = E_0 e^{-(\ln 2/2)(2\tau/\tau_p)^2} \cos(\omega_0 \tau), \qquad (27)$$

where E_0 is the maximum value of the input electric field; τ_p is the full width at half maximum pulse duration; and ω_0 is the mean frequency of the pulse. The propagation of a two-cycle input pulse ($\tau_p = 2T$ and $T = 2\pi/\omega_0$) was analyzed by

numerical simulation using Eq. (26). The input pulse (27) was numerically truncated outside the region where the field was less than 10^{-6} of the peak and the dc spectral component was set to zero.

Figure 1(a) depicts the dispersive broadening of this pulse when its spectrum is largely in a range of normal dispersion. The carrier frequency $\omega_0 = 1.63 \ \omega_{th}$, where $\omega_{th} = \sqrt[4]{b/3a}$ is the frequency of zero group dispersion [i.e., the frequency at which $\partial^2 k / \partial \omega^2 = 0$ in the dispersion relation (8)]. In fused silica, for example, these correspond to wavelengths λ_0 $= 2\pi c / \omega_0 = 0.78 \ \mu m$ and $\lambda_{th} = 2\pi / \omega_{th} = 1.27 \ \mu m$ [9,10]. The changes of shape of extremely short pulses during linear propagation in fused silica are illustrated in Fig. 1. The zeros of the electric field at the front of the pulse are spaced farther apart in time than in the input pulse, and those at the rear are closer together, as shown in Fig. 1(a). As expected, temporal broadening is accompanied by a chirp of the pulse due to positive group velocity dispersion.

When the spectrum of the extremely short pulse is predominantly in a region of anomalous dispersion the opposite chirp is seen, as in Fig. 1(b). For this case $\omega_0 = 0.85 \ \omega_{th}$ $(\lambda_0 = 1.5 \ \mu\text{m})$ and $\lambda_{th} = 1.27 \ \mu\text{m}$. The "period" is shorter at the front of an output pulse and increases toward its tail.

The evolution of an input pulse with spectrum in the range of zero group dispersion is shown in Fig. 1(c). The dynamics differs qualitatively from those shown in Fig. 1(a) and Fig. 1(b), because in this case the higher-order dispersive terms dominate. First, the dispersive pulse broadening occurs much more slowly. Second, as the main part of the pulse broadens, rapid oscillations appear at the pulse tail, and begin to extend over quite a long duration. These subpulse oscillations have a complicated structure, and adjacent subpulses differ from one another by a π phase jump.

The self-action of extremely short pulses in nonlinear optical media differs qualitatively from the examples discussed above. It is also different from that of longer pulses characterized by an envelope [9,10]. In particular, the pulse spectral density does not vary during dispersive propagation in linear media, while spectral broadening may occur in nonlinear media. Using the equations derived in Sec. II we may study the spectral superbroadening of intense femtosecond pulses in a nonlinear waveguide.

B. Generation of spectral supercontinua

In this section we analyze the development of the spectral supercontinuum when a femtosecond pulse propagates in a hollow waveguide filled with a pressurized gas. The gas considered here is deuterium, which has a significant Raman nonlinearity in addition to the electronic nonlinearity.

If the spectrum during superbroadening contains frequencies significantly smaller than the frequencies of electronic transitions in gas then the nonlinear part of the electronic polarization response can be considered dispersionless in the first approximation. Taking into consideration that the population of excited vibrational states in Raman scattering can usually be neglected in moderately intense femtosecond pulses, we can use the system of wave equations in the form of (14).

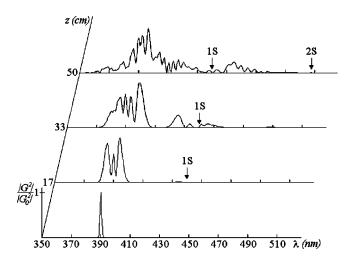


FIG. 2. Spectral evolution of a 150 fs second harmonic pulse of a Ti:sapphire laser with input intensity of 9×10^{12} W/cm² during propagation in a hollow waveguide filled with deuterium under pressure of 45 atm. *G* is the pulse spectral density, and $|G_0|$ is its maximum at the medium input (z=0). Arrows indicate first (1*S*) and second (2*S*) Stokes frequencies.

For the purposes of computation the system (14) can be written in normalized units as

$$\frac{\partial \tilde{E}}{\partial \tilde{z}} - \frac{\partial^3 \tilde{E}}{\partial \tilde{\tau}^3} + B \int_{-\infty}^{\tilde{\tau}} \tilde{E} d\tilde{\tau}' + G \tilde{E}^2 \frac{\partial \tilde{E}}{\partial \tilde{\tau}} + H \frac{\partial \tilde{Q} \tilde{E}}{\partial \tilde{z}} = 0,$$
$$\frac{\partial^2 \tilde{Q}}{\partial \tilde{\tau}^2} + \frac{1}{T_\nu \omega_0} \frac{\partial \tilde{Q}}{\partial \tilde{\tau}} + \left(\frac{\omega_\nu}{\omega_0}\right)^2 \tilde{Q} = \left(\frac{\omega_\nu}{\omega_0}\right)^2 \tilde{E}, \qquad (28)$$

where $\tilde{E} = E/E_0$, $\tilde{Q} = Q/E_0^2$, $\tilde{z} = a\omega_0^3 z$, and $\tilde{\tau} = \omega_0 \tau$. In Eq. (28) $B = 3(\omega_{th}/\omega_0)^4$, $G = 4\Delta n_{NL}^e/a\omega_0^2 c$, $H = 2\Delta n_{NL}^R/a\omega_0^2 c$, where $\omega_{th} = \sqrt[4]{b'/3a}$ is the frequency of zero group dispersion; *a* and *b'* characterize the material and waveguide dispersion and *b'* corresponds to the TE₀₁ mode (see Sec. II A); $\Delta n_{NL}^e = gcE_0^{2/4}$ and $\Delta n_{NL}^R = q\gamma cE_0^2/2\omega_{\nu}^2$ are actually the non-linear additions to the index of refraction, i.e., $\Delta n_{NL}^{e,R} = n_2^{e,R}I$, where *I* is the radiation intensity and $n_2^e = gc/2$ and $n_2^R = q\gamma c/\omega_{\nu}^2$ are the coefficients characterizing the electronic- and Raman-nature contributions to the nonlinear refractive index of the medium. The input pulse in the numerical calculations is given in the form (27).

Figures 2 and 3 (solid line) present the results of a simulation corresponding to the conditions of an experiment described in Ref. [57] where supercontinuum generation was observed. A pulse generated from the second harmonic of a Ti:sapphire laser with central frequency $\omega_0 = 4.83 \times 10^{15} \text{ s}^{-1}$, intensity $I=9\times10^{12} \text{ W/cm}^2$, and input pulse duration $\tau_p = 150$ fs was input to hollow capillaries with a diameter of 0.18 mm and lengths of 17, 33, and 50 cm filled with deuterium under pressure *P* of 45 atm. Experimental settings corresponding to the parameters of the mathematical model (28) are B=0.22, $G=8\times10^{-2}$, $H=5\times10^{-3}$, $\tau_p\omega_0 = 7\times10^2$, $1/T_v\omega_0 = 3.4\times10^{-6}$, and $\omega_v/\omega_0 = 0.12$.

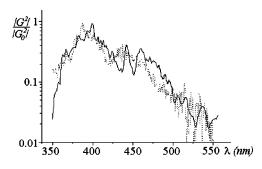


FIG. 3. Spectral supercontinuum generated in 50 cm section of hollow waveguide filled with deuterium under pressure of 45 atm. Input pulse parameters are the same as in Fig. 2. The solid line represents the numerical simulation, and the dotted one is for the experimental data.

Figure 2 shows the evolution of the spectrum of the pulse as it propagates in capillaries. According to both experimental and simulation data, nonuniform broadening of the pump spectrum (by a factor of 5-7) dominates due to phase modulation in the initial stage of supercontinuum generation at the section z=17 cm in the capillary. After that the spectrum broadens nonuniformly with an intensity of 3-5% of the maximum arising in the broadened pulse close to the Stokesshifted frequencies. For a capillary length of 33 cm, the input spectrum broadens 10-12 times. Spectral components comparable in intensity with the central pulse frequency appear near the first Stokes frequency. The most intense components in turn generate Raman-shifted frequencies (see, for example, the component near 490 nm). At the output of a 50 cm capillary the pump spectrum and the first Stokes and anti-Stokes components merge, forming a spectral supercontinuum extending from 350 to 470 nm (10000 cm⁻¹) at an intensity of 3% of maximum. This numerical simulation corresponds to the experimentally observed supercontinuum spectra [58].

For instance, Fig. 3 (dotted line) presents the supercontinuum spectrum experimentally observed at the output of a 50 cm capillary. Note that the spectrum was registered by averaging over 1000 laser pulses with fluctuations of the pulse energy. The result of the numerical simulation using similar averaging is presented in Fig. 3 by the solid line. The comparison of the numerical and experimental data demonstrates not only a qualitative but also a reasonable quantitative fit of the calculated and measured supercontinuum spectra. This provides preliminary evidence supporting the validity of the mathematical model.

It is evident that in the presence of Raman resonances the emission spectrum is broadened via sequential generation of Stokes and anti-Stokes components. Each spectral component is broadened by self-phase and cross-phase modulations, which cause partially overlapped spectra of neighboring Raman components and thereby form a quasidiscrete spectrum. An analysis of a similar problem using a slowly varying envelope approach would require the simultaneous solution of about ten coupled nonlinear equations for envelopes of each of the individual sidebands.

IV. CONCLUSION

The nonlinear dynamics of pulses consisting of only a few oscillations of an optical field may not be correctly described by the formalism of the slowly varying envelope approximation. This approximation applies primarily to quasimonochromatic radiation which is not applicable to extremely short pulses with a corresponding broad spectrum. Although modifications to the envelope approach are possible that provide good agreement with experimental data, the derivation of these equations from Maxwell's equations, and their range of applicability, has yet to be established.

We have outlined in this paper the derivation of wave equations describing the evolution of femtosecond electric fields directly. Techniques for constructing nonlinear evolution equations for the electric field of extremely short light pulses propagating in bulk and waveguide optical media with nonlinearities of various natures are formulated. A system of equations which takes into account the material and waveguide dispersions of the medium that are linear with respect to the field in the transparency range, as well as nonlinearities of electronic and vibronic nature, is obtained. Several effects arising in the propagation of extremely short pulses and the characteristic features of these effects have been analyzed on the basis of these equations, among them dispersive broadening of extremely short pulses and the generation of a spectral supercontinuum in a hollow fiber filled with gaseous deuterium.

There are a number of unsolved problems in the theory of the nonlinear propagation of extremely short light pulses. It is a subject in which it is difficult to gain insight because of the complex interplay of dispersion and nonlinearities. Nonetheless, the area is rich with new physics, and is likely to be technologically important as the boundaries of ultrashort pulse generation and spectroscopy are pushed back.

The solution of similar problems is of great importance for applications in technology. We can expect that femto- and subfemtosecond pulses may be efficiently used in information systems, in fabricating materials with new properties, in diagnostics of superfast processes, and in other areas of physics, chemistry, and optics.

ACKNOWLEDGMENTS

This work was supported by Grant Nos. RP1-2249 of the U.S. Civilian Research and Development Foundation, Nos. 01-02-17841 and 02-02-06885 of the Russian Foundation of Basic Research, and No. UR.01.01.030 of the Program of the Russian Education Ministry "Universities of Russia." I.A.W. acknowledges the support of the National Science Foundation, and informative conversations with Alex Gaeta.

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