Nonlinear Optical Pulse Propagation in the Single-Cycle Regime

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(Received 27 September 1996)

A general three-dimensional wave equation first order in the propagation coordinate is derived covering a broad range of phenomena in nonlinear optics. This equation provides an accurate description of the evolution of the wave packet envelope down to pulse durations as short as one carrier oscillation cycle. The concept of envelope equations is found to be applicable to the single-cycle regime of nonlinear optics. [S0031-9007(97)02995-5]

PACS numbers: 42.65.−k, 31.15.−p

The propagation equation governing the evolution of the complex envelope of optical pulses plays a key role in nonlinear optics. Owing to the assumption of a slowly varying envelope, this partial differential equation contains only the first derivative with respect to the spatial coordinate along the propagation direction. Hence, it can be solved with substantially smaller computational effort than Maxwell's wave equation, which is of second order in the propagation coordinate. This benefit has been exploited in the investigation of a vast number of nonlinear optical phenomena [1–10].

The evolution of ultrashort pulse optics has now arrived at a point where light pulses with durations comparable to the carrier oscillation cycle have become available [11]. This progress opens up new prospects in nonlinear optics as well as strong-field physics and prompts the question of whether the first-order propagation equation is valid in this new and important regime of optics. The purpose of this Letter is to show that (i) the powerful concept of the envelope can be extended to pulse durations equal to the carrier oscillation period T_0, and (ii) the validity of the first-order envelope equation, commonly used to model one-dimensional nonlinear pulse evolution, extends down to pulse durations as short as T_0. Furthermore, (iii) our approach leads, in a natural way, to a generalized, scalar, three-dimensional, first-order propagation equation, which obviates the need for the Maxwell equations in tackling a number of problems, where transverse effects cannot be neglected in the interaction of ultrashort light pulses with matter.

A prerequisite for introducing an envelope equation is the unambiguous definition of the envelope. To this end, the electric field $E(t) = \tilde{E}(t) + \text{c.c.}$ is represented by the complex electric field which is written as $\tilde{E}(t) = A(t)\exp(-i\omega_0 t + i\psi)$. Here, $\omega_0 = \int_0^\infty \omega |E(\omega)|^2 d\omega / \int_0^\infty |E(\omega)|^2 d\omega$ is the carrier frequency, $E(\omega)$ is the Fourier transform of $E(t)$, and ψ is defined such that the imaginary part of the complex envelope $A(t)$ is zero at $t = 0$. The above definition of $A(t)$ is only physically meaningful as long as the envelope remains *invariant* under a change of ψ. This condition is equivalent to the requirement that a phase shift of the electric field $\tilde{E}'(t) = \tilde{E}(t)\exp(i\Delta\psi)$ does not change the center frequency $\omega_0' = \omega_0$. That indeed applies to a high accuracy for pulse durations down to $\tau_p \approx T_0$, as revealed by Fig. 1. The pulse width τ_p is defined as the full width at half maximum (FWHM) of $|A(t)|^2$. For shorter wave packets, the variation of ψ gives rise to non-negligible changes of the spectral intensity at low frequencies and results in the increased sensitivity of ω_0 to ψ. The invariance of ω_0 under a change of ψ can be checked for arbitrary wave forms following the procedure presented above. In conclusion, a physically meaningful envelope can be assigned to ultrashort light wave packets that contain at least one carrier cycle within the FWHM of their intensity envelope. At the 780 nm center wavelength of the Ti:sapphire laser this implies $\tau_p = 2.6$ fs.

Our derivation of the envelope equation starts with Maxwell's equations. For small transverse inhomogeneities of the medium polarization, the three-

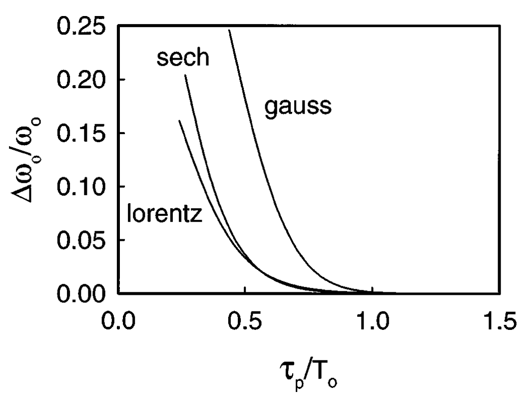

FIG. 1. Phase dependence of the carrier frequency $\Delta\omega_0/\omega_0 = |\omega_0(\psi = 0) - \omega_0(\psi = \pi/2)|/\omega_0$ versus the normalized pulse duration for various analytic pulse shapes, $E(t) = A(t)\sin(\omega_c t + \psi)$; the envelopes are described by a Gaussian, $A_g(t) = \exp[-(1.67t/\tau_p)^2]$, a hyperbolic secant, $A_s(t) = \text{sech}(1.76t/\tau_p)$, and a Lorentzian, $A_l(t) = 1/[1 + (1.29t/\tau_p)^2]$ pulse. The optical cycle $T_0 = 2\pi/\omega_c = 2.67$ fs at a wavelength of 800 nm. Here, ω_c is the carrier frequency of the analytical pulse shape and ω_0 is the center of gravity of the spectrum, as defined in the text. For $\tau_p \geq T_0$ the phase sensitivity is negligible and $\omega_c \approx \omega_0$.

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dimensional wave equation can be written as [1–7]

$$(\partial_z^2 + \nabla_\perp^2)E(\mathbf{r},t) - \frac{1}{c^2}\partial_t^2\int_{-\infty}^{t} dt'\varepsilon(t-t')E(\mathbf{r},t') = \frac{4\pi}{c^2}\partial_t^2 P_{nl}(\mathbf{r},t), \tag{1}$$

where $\nabla_\perp^2 = \partial_x^2 + \partial_y^2$ is the transversal Laplace operator, $\partial_{i=x,y,z,t}$ stands for the respective partial derivatives, $\varepsilon(t) = (2\pi)^{-1}\int_{-\infty}^{\infty} d\omega\,\varepsilon(\omega)e^{-i\omega t}$, $\varepsilon(\omega) = 1 + 4\pi\chi(\omega)$, and $\chi(\omega)$ is the linear electric susceptibility. The electric field E propagates along the z direction. Both E and the nonlinear polarization P_{nl} are polarized parallel to the x axis.

In order to introduce a first-order propagation equation for the wave packet envelope defined above, we use the ansatz $\mathbf{E}(\mathbf{r},t) = A(\mathbf{r}_\perp,z,t)e^{i(\beta_0 z - \omega_0 t + \psi_0)}$ + c.c., where ω_0 and ψ_0 are determined at the input reference plane $z = 0$ on the beam axis $\mathbf{r}_\perp = 0$ as prescribed above. Further, $\beta_0 = \mathrm{Re}[k(\omega_0)] = (\omega_0/c)n_0$, where $k(\omega) = (\omega/c)\sqrt{\varepsilon(\omega)}$ is the complex propagation constant and n_0 is the refractive index of the medium at ω_0. The induced nonlinear polarization is written as $P_{\mathrm{nl}}(\mathbf{r},t) = B(\mathbf{r}_\perp,z,t,A)e^{i(\beta_0 z - \omega_0 t + \psi_0)}$ + c.c., where the complex amplitude B depends nonlinearly on the amplitude of the electric field. The neglect of backward propagating waves is consistent with the approximations that will be made in the following derivation of the envelope equation and will be commented on later.

The substitution of the above expressions of $\mathbf{E}(\mathbf{r},t)$ and $P_{\mathrm{nl}}(\mathbf{r},t)$ in Eq. (1), Fourier transform of the integral term with respect to the time coordinate and the subsequent Taylor-expansion of $k(\omega)$ about ω_0 followed by an inverse Fourier transform yields

$$(-\beta_0^2 + 2i\beta_0\partial_z + \partial_z^2 + \nabla_\perp^2)A + \left(\beta_0 + i\frac{\alpha_0}{2} + i\beta_1\partial_t + \hat{D}\right)^2 A = \frac{4\pi\omega_0^2}{c^2}\left(1 + \frac{i}{\omega_0}\partial_t\right)^2 B. \tag{2}$$

Here we assumed P_{nl} to be a small perturbation to the linear polarization, again, in conformance with later approximations, the dispersion operator $\hat{D}$ is given by

$$\hat{D} = -\frac{\alpha_1}{2}\partial_t + \sum_{m=2}^{\infty}\frac{\beta_m + i\alpha_m/2}{m!}(i\partial_t)^m, \tag{3}$$

and $\beta_m = \mathrm{Re}[(\partial^m k/\partial\omega^m)_{\omega_0}]$ and $\alpha_m = \partial\,\mathrm{Im}[(\partial^m k/\partial\omega^m)_{\omega_0}]$. In the moving reference frame $\tau = t - \beta_1 z$, $\xi = z$, Eq. (2) can be written as

$$\left(1 + \frac{i}{\omega_0}\partial_\tau\right)\left[\left(\partial_\xi + \frac{\alpha_0}{2} - i\hat{D}\right)A + \frac{2\pi\beta_0}{in_0^2}\left(1 + \frac{i}{\omega_0}\partial_\tau\right)B\right] + \frac{1}{2i\beta_0}\nabla_\perp^2 A$$
$$\simeq \left(\frac{\beta_0 - \omega_0\beta_1}{\beta_0}\right)\frac{i}{\omega_0}\partial_\tau\left(\partial_\xi + \frac{\alpha_0}{2} - i\hat{D}\right)A - \frac{1}{2i\beta_0}\left(\partial_\xi^2 + \hat{D}^2 - \frac{\alpha_0^2}{4} - \alpha_0\beta_1\partial_\tau\right)A, \tag{4}$$

where $\hat{D}$ is obtained by replacing ∂_t with ∂_τ in (3). The terms on the right side are small as compared with the left side if

$$|\partial_\xi A| \ll \beta_0|A| \tag{5a}$$

and

$$|\partial_\tau A| \ll \omega_0|A| \tag{5b}$$

or

$$\left|\frac{\beta_0 - \omega_0\beta_1}{\beta_0}\right| \ll 1. \tag{5c}$$

If either (5a) and (5b) or (5a) and (5c) are satisfied, (4) simplifies to

$$\partial_\xi A = -\frac{\alpha_0}{2}A + i\hat{D}A + \frac{i}{2\beta_0}\left(1 + \frac{i}{\omega_0}\partial_\tau\right)^{-1}\nabla_\perp^2 A + i\frac{2\pi\beta_0}{n_0^2}\left(1 + \frac{i}{\omega_0}\partial_\tau\right)B, \tag{6}$$

where $[1 + (i/\omega_0)\partial_\tau]^{-1}$ can be evaluated in the frequency domain. This generic nonlinear envelope equation (NEE) *first-order in the propagation coordinate* ξ provides a powerful means of describing light pulse propagation in dispersive nonlinear media. In the specific case of one-dimensional propagation (i.e., with the diffraction term discarded) with $\alpha_0 \approx 0$ and $\hat{D} \approx -(\beta_2/2)\partial_\tau^2$ Eq. (6) reduces to a *nonlinear Schrödinger equation*, which has been widely used to describe ultrashort pulse propagation in Kerr media ($B \propto |E|^2$) [1,3,5].

From the solution $A(\mathbf{r}_\perp,\xi,\tau)$ of Eq. (6) the electric field can be reconstructed as

$$E(\mathbf{r}_\perp,\xi,\tau) = Ae^{-i\omega_0\tau + i\psi(\xi)} + \text{c.c.},$$
$$\psi(\xi) = \psi_0 + (\beta_0 - \omega_0\beta_1)\xi. \tag{7}$$

These equations describe the evolution of light wave packets in terms of a *fixed* carrier frequency ω_0 defined at $\xi = 0$, the entrance of the propagation medium, and an *evolving* complex envelope $A(\mathbf{r}_\perp,\xi,\tau)$ and phase $\psi(\xi)$, which determines the "position" of the carrier wave relative to the envelope. The complex envelope $A(\mathbf{r}_\perp,\xi,\tau)$ evolves due to absorption (or gain), dispersion, diffraction, and nonlinearities, whereas $\psi(\xi)$ evolves due to a difference between group velocity (β_1^{-1}) and phase velocity in the propagation medium.

Traditionally, specific forms of the NEE such as the nonlinear Schrödinger equation have been derived by making use of the *slowly-varying-envelope approximation* (SVEA), $|\partial_z A| = |\partial_\xi A - \beta_1 \partial_\tau A| \ll \beta_0 |A|$. This condition can be decomposed into (5a) and (5b) by utilizing $\beta_0/\beta_1 \approx \omega_0$, which impose distinctly different requirements on the physical system. The former, "nonlocal" part of the SVEA requires that the complex amplitude does not excessively change *during propagation*, a requirement whose violation was previously shown to result in the emergence of a backward propagating wave [2]. The latter "local" contribution demands that the pulse duration must be much longer than the carrier oscillation period. The essential new finding from our derivation [12] of the NEE is that this latter requirement may be dropped if (5c) is satisfied. As a matter of fact, condition (5c) is met if the difference between group and phase velocity relative to the latter is small compared to one, which is fulfilled in a wide range of propagation phenomena. Drawing on (7), condition (5c) can be reexpressed as $|\partial\psi/\partial\xi| \ll \beta_0$. Consequently, (5a) and (5c) can be merged into a single mathematical requirement

$$|\partial_\xi E| \ll \beta_0 |E|, \tag{8}$$

which we refer to as the *slowly-evolving-wave approximation* (SEWA). The SEWA requires more from the propagation medium than the SVEA: not only the envelope A but also the relative carrier phase ψ must not significantly vary as the pulse covers a distance equal to the wavelength $\lambda_0 = 2\pi c/\omega_0$. In return, it does not explicitly impose a limitation on the pulse width. Therefore, *in the frame of the SEWA the nonlinear envelope equation accurately describes light pulse propagation down to the single cycle regime*. The region of validity of the SEWA can be easily assessed by introducing the characteristic propagation lengths $L_\psi = (2\pi)^{-1} |dn/d\lambda|_{\lambda_0}^{-1}$, over which ψ is changed by 1, further $L_{\alpha,m} = \tau_p^m / |\alpha_m| (m = 0, 1, \ldots)$, $L_{\beta,m} = \tau_p^m / |\beta_m| (m = 2, 3, \ldots)$, and $L_\perp = \beta_0 w_0^2$, and $L_{\mathrm{nl}} = |A/B| (n_0^2 / 2\pi\beta_0)$, over which the envelope is significantly modified due to absorption (α_0) dispersion ($\alpha_m, m \geq 1$; $\beta_m, m \geq 2$), diffraction, and nonlinearities, respectively. Here τ_p is the pulse duration, w_0 is the beam radius at the beam waist for a hypothetical linear propagation, whereas A and B are the respective field amplitudes at an arbitrary instant and position. The SEWA is applicable as long as each of these characteristic length scales meets the condition $\beta_0 L_{\mathrm{char}} \gg 1$. Careful inspection of the parameters of various nonlinear media yields the remarkable finding that these conditions are well satisfied for a wide range of phenomena in the parametric regime (i.e., ω_0 is far off resonances) below the ionization threshold. Strong resonant coupling may, however, in specific cases give rise to extremely short $L_{\psi,\alpha,\beta,\mathrm{nl}}$, violating the condition for the SEWA [13].

The validity of the NEE down to the single-cycle regime has been tested by solving (6) and the Maxwell equations numerically for a Kerr medium characterized by the constitutive law $B = (2\pi)^{-1} n_0 n_2 |A|^2 A$, where n_2 is the nonlinear index of refraction [1] and $|A|^2$ is normalized to give the intensity. For simplicity, we considered one-dimensional propagation, assumed the Kerr response to be instantaneous [1], and neglected the emergence of harmonic radiation [10]. A sech-shaped pulse with an FWHM duration of $\tau_p = 2.67$ fs, which corresponds to one optical cycle at the carrier wavelength of 0.8 μm, was propagated through a hypothetical dielectric medium with $\beta_2 = 0.0385$ fs$^2/\mu$m ($\beta_m = 0$ for $m > 2$) and $n_2 = 3 \times 10^{-16}$ W/cm^2 ($dn_2/d\omega = 0$), which correspond to the respective parameters of fused silica. In order to simulate an extremely strong parametric interaction the peak intensity of the incident pulse was chosen as $|A(\xi = 0, \tau = 0)|^2 = 4 \times 10^{13}$ W/cm^2, which is close to the expected critical intensity level for optical damage in fused silica and other dielectric materials. In advancing the coupled Maxwell's equations, from which (1) has been derived, in space the leap frog method [14] was used. The time derivatives were evaluated in the ω space using a fast Fourier transform. The NEE was integrated by applying the split-step Fourier method [1].

In Fig. 2 the solutions of the Maxwell's equations and Eq. (6) as given by (7) are depicted for the case of $\beta_2 = 0$. In the absence of dispersion, self steepening due to the time derivative of the nonlinearity creates an optical shock at the trailing edge of the pulse. The critical distance for self steepening [1] is given by $z_s = 0.43 L_{\mathrm{nl}} \omega_0 \tau = 23.36$ μm, which is close to the chosen propagation distance of $z = 20$ μm. Even along the shock front, where the change of the envelope is comparable to the change of the carrier frequency, the two solutions are virtually identical. Figure 3 shows the electric field

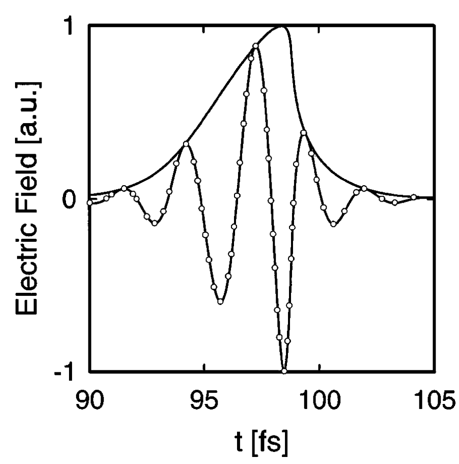

FIG. 2. Shows the electric field strength and the field envelope in arbitrary units versus propagation time. The electric fields obtained by the solution of the Maxwell equation and of Eq. (4) are depicted by the full line and by the open circles, respectively; the initial pulse has a sech-shape and the parameters are, $I = 4 \times 10^{13}$ W/cm^2, $n_2 = 3 \times 10^{-16}$ cm^2 W^{-1}, $\omega_0 = 2.35$ fs^{-1} (0.8 μm), $\tau_p = 2.67$ fs, $n_0 = 1.45$, $dn/d\omega \mid_{\omega_0} = 0$ fs (no dispersion), and a propagation distance $z, \xi = 20$ μm.

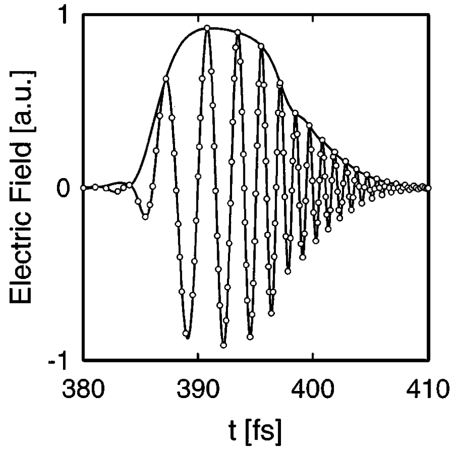

FIG. 3. The same as in Fig. 1, with a propagation distance $z = 80\ \mu\text{m}$ and $dn/d\omega \mid_{\omega_0} = 5.776 \times 10^{-3}$ fs, which gives $\beta_1 = 4.882\ \text{fs}/\mu\text{m}$ and $\beta_2 = 3.853 \times 10^{-2}\ \text{fs}^2/\mu\text{m}$.

for the case of $\beta_2 > 0$ and $z = 80\ \mu\text{m}$. Dispersion suppresses the formation of a shock front in this case. Again, an excellent agreement between the solution of Maxwell's equations and that of the NEE is found, which has also been verified for longer propagation distances.

The NEE incorporates the "correction" operator $[1 + (i/\omega_0)\partial_\tau]$ in the terms accounting for diffraction and nonlinear effects when compared with the paraxial wave equation describing the propagation of stationary or long-pulse radiation [2–6]. Physically, the appearance of the correction operator in the nonlinear term is responsible for introducing a phase modulation if the nonlinearity modulates primarily the amplitude in the long-pulse limit (as is the case in a near-resonant interaction), and vice versa (e.g., self steepening versus self phase modulation in the case of a Kerr nonlinearity). Mathematically, the correction operator becomes zero at $\omega = 0$ in the frequency domain, frustrating the emergence of a dc component of the wave packet during propagation. The same operator in the denominator of the diffraction term in (6) accounts for an enhanced beam divergence at lower-frequency components. Since this generalized diffraction operator [15] has a singularity at $\omega = 0$ in the frequency domain, the use of Eq. (6) is restricted to initial pulses having a negligible dc spectral component, a requirement which is consistent with the definition of the envelope.

So far, our discussion was confined to studying the evolution of the incident wave packet carried at ω_0. The nonlinear response of the medium may arise to the emergence of waves at new (e.g., harmonic) frequencies. The concept of the SEWA can be extended to nonlinear frequency mixing processes, such as harmonic generation and parametric amplification, as long as the difference between phase and group velocities of the waves involved in the interaction is small compared to the phase (or group) velocity of the fundamental pulse [13]. Far from resonances, this requirement is fulfilled in the majority of parametric processes in which all waves propagate in the same direction. The condition is clearly violated in processes such as Brillouin scattering [2,3], where the pulses propagate in opposite directions. Therefore Brillouin scattering can be addressed using the NEE only in the frame of the SVEA, i.e., for pulse durations much longer than the optical cycle.

In conclusion, the concept of the envelope in the description of nonlinear light wave propagation has been extended to a regime, where the pulse duration is comparable to the carrier oscillation cycle. Drawing on this concept, a three-dimensional first-order envelope propagation equation has been derived, which, in the perturbative limit of small population transfer, provides an accurate description of the propagation of ultrashort light transients in nonlinear media.

The authors gratefully acknowledge illuminating discussions with G. A. Mourou and P. B. Corkum. This work was supported by the ONB grants No. 5124 and No. 5335 and by FWF Grant Y44-PHY. T. Brabec is supported by the Österreichische Akademie der Wissenschaften, APART Grant.

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