

Ultra-short Pulse Propagation in Nonlinear Optics

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Abstract

In this work, we perform a brief review about the theoretical modeling of the electromagnetic propagation of ultra-short pulses (\sim femtoseconds) in different scenarios of nonlinear optics.

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1 Introduction

The propagation of electromagnetic waves can be modeled by using Maxwell's equations, the essence of classical electromagnetism. They are able to describe the structure of the electromagnetic field and the fundamental interactions between the electric field strength (\mathcal{E}) and the magnetic induction (\mathcal{B}). In their fundamental form, the *macroscopic* Maxwell equations read as follows (we use the notation $\partial_t = \partial/\partial t$):

$$\nabla \times \mathcal{E}(\mathbf{r}, t) = -\partial_t \mathcal{B}(\mathbf{r}, t); \quad (1)$$

$$c_0^2 \nabla \times \mathcal{B}(\mathbf{r}, t) = \frac{1}{\varepsilon_0} \mathcal{J}(\mathbf{r}, t) + \partial_t \mathcal{E}(\mathbf{r}, t); \quad (2)$$

$$\varepsilon_0 \nabla \cdot \mathcal{E}(\mathbf{r}, t) = \rho(\mathbf{r}, t); \quad (3)$$

$$\nabla \cdot \mathcal{B}(\mathbf{r}, t) = 0. \quad (4)$$

where ρ and \mathcal{J} are the total charge and current densities, respectively^{1,2}. Both terms include the free and bound charges and currents, i.e., $\rho = \rho_f + \rho_b$ and $\mathcal{J} = \mathcal{J}_f + \mathcal{J}_b$ [1].

As is well known, the fields \mathcal{E} and \mathcal{B} are able to modify the charge distribution of a medium and induce currents in it. Such alterations act as new sources that generate additional electromagnetic fields. Thus, it is necessary to know the electromagnetic response of the medium to model theoretically the different electromagnetic wave propagation phenomena. These field-matter interactions can be adequately described by defining two new auxiliary macroscopic fields, namely \mathcal{D} (electric displacement) and \mathcal{H} (magnetic field strength), which account for the macroscopic response of the medium charges and currents to the applied fields. The connection between the fundamental and auxiliary fields is given through the so-called constitutive relations:

$$\mathcal{D}(\mathbf{r}, t) := \varepsilon_0 \mathcal{E}(\mathbf{r}, t) + \mathcal{P}(\mathbf{r}, t); \quad \mathcal{H}(\mathbf{r}, t) := \frac{1}{\mu_0} \mathcal{B}(\mathbf{r}, t) - \mathcal{M}(\mathbf{r}, t), \quad (5)$$

where \mathcal{P} and \mathcal{M} are the polarization and magnetization fields, and basically express the density of electric and magnetic dipole moments, respectively.

In a dielectric medium (e.g. an optical fiber), the free charges and currents are found to be null, $\rho = \rho_b = -\nabla \cdot \mathcal{P}$, $\mathcal{J} = \mathcal{J}_b = \partial_t \mathcal{P}$, $\mathcal{M} = \mathbf{0}$ and Eqs. (1)-(4) are reduced to [1]:³

$$\nabla \times \mathcal{E}(\mathbf{r}, t) = -\partial_t \mathcal{B}(\mathbf{r}, t); \quad (6)$$

$$\nabla \times \mathcal{H}(\mathbf{r}, t) = \partial_t \mathcal{D}(\mathbf{r}, t); \quad (7)$$

$$\nabla \cdot \mathcal{D}(\mathbf{r}, t) = 0; \quad (8)$$

$$\nabla \cdot \mathcal{B}(\mathbf{r}, t) = 0. \quad (9)$$

A rigorous electromagnetic analysis of the linear and nonlinear pulse propagation through an optical fiber should be performed by solving Eqs. (6)-(9) in the temporal and longitudinal orders of reference of \mathcal{E} and \mathcal{B} , i.e., $\delta t \sim 2\pi/\omega_0$ and $\delta z \sim \lambda$, where ω_0 and λ are the values of the angular frequency and the wavelength of the laser in the core of the fiber.

However, the numerical calculation of these equations requires a high computational time and memory when several meters ($z > 10^6 \lambda$) or kilometers ($z > 10^9 \lambda$) are involved. In such a case, it is more suitable to consider the temporal (T_P) and longitudinal width (Λ_P) of the optical pulses as the temporal and longitudinal orders of reference ($\delta t \sim T_P$ and $\delta z \sim \Lambda_P$). In other

¹All the fields appearing in Eqs. (1)-(4) are volume-averaged quantities, expressed in S.I. units.

² $c_0 = 1/\sqrt{\varepsilon_0 \mu_0}$ is the speed of light in vacuum. ε_0 and μ_0 are respectively the electric permittivity and the magnetic permeability in vacuum.

³Eqs. (6)-(9) are also valid to describe the structure of the electromagnetic field in vacuum by taking $\mathcal{P} = \mathbf{0}$.

words, the complexity of the numerical calculations can be alleviated if we are able to simulate only the slowly-varying temporal and longitudinal evolution of \mathcal{E} and \mathcal{B} , the so-called *complex envelope* (\mathcal{A} , see Fig. 1).

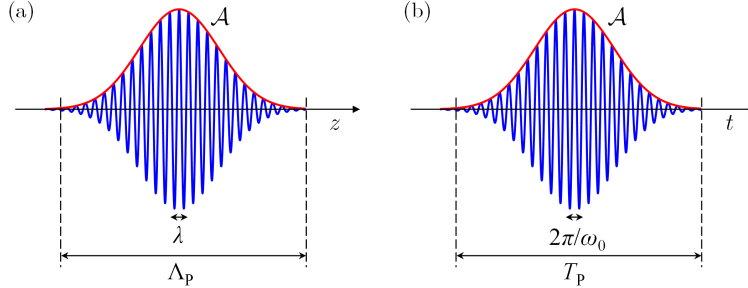


Fig. 1. Optical pulse: (a) longitudinal and (b) temporal changes. (Blue line: rapidly-varying fluctuations. Red line: slowly-varying fluctuations, complex envelope.)

The partial differential equation modeling the linear and nonlinear propagation of \mathcal{A} in guided and unguided optical media will be referred to as the pulse propagation equation (PPE) in this work. In the photonics literature, this equation is usually termed as the generalized paraxial wave equation or the pulse propagation equation [2, 3, 4]. Specifically, in guided media, it can also be referred to as the nonlinear Schrödinger equation (NLSE) due to the intimate relation between the PPE and the NLSE when the space and time are normalized [5]. In the following, we first review the mathematical derivation of the PPE from Maxwell's equations when propagating femtosecond optical pulses through a single-core single-mode optical fiber, later, we discuss the propagation in uniform (\equiv unguided) nonlinear dielectric media, and finally, we focus our attention in gases for white light continuum generation applications.

2 Pulse propagation in optical fibers

In general, the derivation of the PPE is simpler than initially foreseen. Figure 2 shows a flowchart of the steps required during our mathematical discussion. Let us remember that our goal is to derive a partial differential equation of the complex envelope (the slowly-varying longitudinal and temporal evolution of the electromagnetic field) starting from Maxwell's equations.

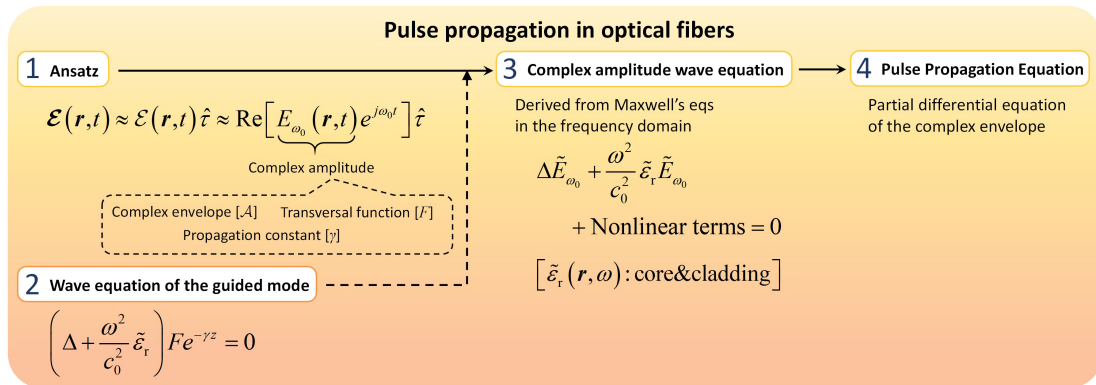


Fig. 2. Flowchart of transformations to derive the PPE from Maxwell's equations in optical fibers.

First step: Ansatz of the electric field strength of the fiber

In the first step, we should propose the ansatz of the global electric field strength (\mathcal{E}) of the fiber⁴. In this work, we will consider an ultra-short optical pulse (femtosecond regime) generated from a laser with optical carrier of angular frequency ω_0 and launched into a single-core single-mode fiber. For the sake of simplicity, we will assume the weakly-guiding approximation (also referred to as the paraxial approximation in the photonics literature) and omit the polarization and birefringent effects of the optical medium. Consequently, the longitudinal component of \mathcal{E} can be neglected and we can approximate $\mathcal{E} \simeq \mathcal{E}\hat{\tau}$, where $\hat{\tau}$ is a transverse unitary vector ($\hat{\tau} \perp \hat{z}$). Without loss of generality, $\hat{\tau}$ can be related to the x or y axis.

At this point, we should decouple the rapidly and the slowly-varying temporal and longitudinal changes of \mathcal{E} . The rapidly-varying *temporal* changes are decoupled by using the *slowly-varying complex amplitude approximation* (SVAA):

$$\mathcal{E}(\mathbf{r}, t) \simeq \text{Re} \{ E_{\omega_0}(\mathbf{r}, t) \exp(j\omega_0 t) \} = \frac{1}{2} [E_{\omega_0}(\mathbf{r}, t) \exp(j\omega_0 t) + E_{-\omega_0}(\mathbf{r}, t) \exp(-j\omega_0 t)], \quad (10)$$

where E_{ω_0} is the complex amplitude of the electric field strength of the fundamental mode LP₀₁ (polarized along the τ axis) satisfying that $|\delta_t E_{\omega_0}| \ll |E_{\omega_0}|$ in $\delta t \sim 2\pi/\omega_0$, where $\delta_t E_{\omega_0} := E_{\omega_0}(\mathbf{r}, t + \delta t) - E_{\omega_0}(\mathbf{r}, t)$. It should be noted that the SVAA allows us to decouple the rapid temporal oscillation of the optical carrier from the slow temporal evolution of the complex amplitudes of the optical pulses [Fig. 1(b), red line]. Therefore, the herein proposed model is valid *if and only if* the Maxwell equations are approximately satisfied when using Eq. (10). However, this assumption is not fulfilled if the pulse is too narrow, namely around the order of the period of the optical carrier or shorter. In such a case, the decomposition performed in Eq. (10) is no longer useful and the concept of the complex amplitude itself becomes unclear. Specifically, the validity of Eq. (10) can be easily tested by verifying that the pulse bandwidth is much lower than $\omega_0/2\pi$.

Now, we should also decouple the rapidly- and slowly-varying *longitudinal* variations of the electric field [Fig. 1(a), red line] by introducing the complex envelope in Eq. (10). Let us describe this point in detail:

1. First, for simplicity, we can start by assuming a laser radiating a continuous-wave (CW) signal. In this case, we can write the complex amplitude E_{ω_0} as follows:

$$E_{\omega_0}(\mathbf{r}, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \tilde{\mathbf{A}} F(x, y, \omega) \exp(-\gamma(\omega)z) \exp(j(\omega - \omega_0)t) d\omega, \quad (11)$$

where $\tilde{\mathbf{A}}$ is a complex constant related to the optical power of the CW signal ($P \propto |\tilde{\mathbf{A}}|^2$), F is the transverse Bessel eigenfunction of the LP₀₁ mode [6] and $\gamma = \alpha/2 + j\beta$ is the propagation constant involving the power attenuation constant α and the phase constant β . Note that in Eq. (11) we have only considered the forward propagation by using the sign $-$ by convention. We can omit the backward propagation because we are not considering fiber perturbations along the z axis and the optical fiber is assumed with a low nonlinear nature. In such a case, we can neglect the reflected power [7]. Thus, the

⁴Since the electric field strength and the magnetic induction can be expressed in identical form, we will use the former field for our theoretical discussions with straightforward extrapolation of the results to the magnetic induction by using the intrinsic impedance of each dielectric region.

Fourier transform⁵ of Eq. (10) is found to be:

$$\tilde{\mathcal{E}}(\mathbf{r}, \omega) = \mathcal{F}[\mathcal{E}(\mathbf{r}, t)] \simeq \text{Re} \left\{ \tilde{E}_{\omega_0}(\mathbf{r}, \omega - \omega_0) \right\} \hat{\tau}, \quad (12)$$

where:

$$\tilde{E}_{\omega_0}(\mathbf{r}, \omega - \omega_0) = \mathcal{F}[E_{\omega_0}(\mathbf{r}, t) \exp(j\omega_0 t)] = \tilde{\mathbf{A}} F(x, y, \omega) \exp(-\gamma(\omega) z). \quad (13)$$

Equation (13) is the well-known solution of an LP mode in the classical modal analysis of a single-core fiber [6].

2. Now, assume a pulsed-wave laser. In this case, the constant $\tilde{\mathbf{A}}$ is found to be frequency-dependent accounting for the amplitude of each spectral component of the pulse propagated by the LP₀₁ mode. That is, Eq. (13) should be rewritten as:

$$\tilde{E}_{\omega_0}(\mathbf{r}, \omega - \omega_0) = \tilde{\mathbf{A}}(\omega - \omega_0) F(x, y, \omega) \exp(-\gamma(\omega) z), \quad (14)$$

with the complex function $\tilde{\mathbf{A}}$ modeling the Fourier transform of the optical pulse in *baseband*. Concretely, in Eq. (14), $\tilde{\mathbf{A}}$ is shifted to the angular frequency of the optical carrier ω_0 because \tilde{E}_{ω_0} (the slowly-varying temporal evolution of the electric field) is shifted to ω_0 .

3. In the pulsed-wave scenario of the previous point, $\tilde{\mathbf{A}}$ is the complex envelope of the optical pulse in the frequency domain. Unfortunately, $\tilde{\mathbf{A}}$ is only able to describe the slowly-varying temporal evolution of the electric field because is the shape of the optical pulse at $z = 0$. To include the slowly-varying *spatial* evolution of the electric field at any z point we should multiply this function by an exponential of the form $\exp(-j\beta(\omega) z)$ but subtracting the rapidly-varying longitudinal variations of the optical carrier $\exp(-j\beta(\omega_0) z)$. That is, we should perform the transformation:

$$\tilde{A}(z, \omega - \omega_0) := \tilde{\mathbf{A}}(\omega - \omega_0) \exp[-j(\beta(\omega) - \beta(\omega_0)) z]. \quad (15)$$

Note that the subtraction $\beta(\omega) - \beta(\omega_0)$ in the spatial exponential is analog to the subtraction $\omega - \omega_0$ in the temporal exponential of Eq. (11). Remarkably, the new complex envelope \tilde{A} describes, in the frequency domain, the slowly-varying *longitudinal* and *temporal* evolution of the global electric field of the fiber (\mathcal{E}).

All in all, the ansatz of Maxwell's equations (first step of Fig. 2) is described by the following expressions in the time domain [$\beta(\omega_0) \equiv \beta_0$]:

$$\mathcal{E}(\mathbf{r}, t) \simeq \text{Re} \{ E_{\omega_0}(\mathbf{r}, t) \exp(j\omega_0 t) \}; \quad (16)$$

$$E_{\omega_0}(\mathbf{r}, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \tilde{A}(z, \omega - \omega_0) F(x, y, \omega) \exp\left(-j\beta_0 z - \frac{1}{2}\alpha(\omega) z\right) \exp(j(\omega - \omega_0) t) d\omega, \quad (17)$$

and in the frequency domain:

$$\tilde{\mathcal{E}}(\mathbf{r}, \omega) \simeq \text{Re} \left\{ \tilde{E}_{\omega_0}(\mathbf{r}, \omega - \omega_0) \right\} \hat{\tau}; \quad (18)$$

$$\tilde{E}_{\omega_0}(\mathbf{r}, \omega - \omega_0) = \tilde{A}(z, \omega - \omega_0) F(x, y, \omega) \exp\left(-j\beta_0 z - \frac{1}{2}\alpha(\omega) z\right), \quad (19)$$

⁵The direct and inverse Fourier transforms employed in this work are defined as:

$$\tilde{X}(\omega) = \mathcal{F}[X(t)] := \int_{-\infty}^{\infty} X(t) \exp(-j\omega t) dt; \quad X(t) = \mathcal{F}^{-1}[\tilde{X}(\omega)] := \frac{1}{2\pi} \int_{-\infty}^{\infty} \tilde{X}(\omega) \exp(j\omega t) d\omega.$$

with the complex envelope in the time domain (A) given by the inverse Fourier transform of \tilde{A} :

$$A(z, t) = \mathcal{F}^{-1} \left[\tilde{A}(z, \Omega) \right] = \frac{1}{2\pi} \int_{-\infty}^{\infty} \tilde{A}(z, \Omega) \exp(j\Omega t) d\Omega, \quad (20)$$

and satisfying the slowly-varying envelope approximation (SVEA) in the space and time domain. Mathematically, the SVEA can be expressed as $|\delta_z A| \ll |A|$ in $\delta z \sim \lambda = 2\pi/\beta_0$ and $|\delta_t A| \ll |A|$ in $\delta t \sim 2\pi/\omega_0$, where $\delta_z A := A(z + \delta z, t) - A(z, t)$ and $\delta_t A := A(z, t + \delta t) - A(z, t)$. From the above statement, we found the order criteria of the SVEA:

$$|\partial_z^2 A| \ll \beta_0 |\partial_z A| \ll \beta_0^2 |A|; \quad |\partial_t^2 A| \ll \omega_0 |\partial_t A| \ll \omega_0^2 |A|. \quad (21)$$

It should be noted that the temporal order criterion of the SVEA is fulfilled thanks to the SVAA, performed in Eq. (10). Moreover, it is noticeable that we are using a different typography to describe the complex envelope (A) from the typography depicted in Figs. 1 and 2 (\mathcal{A}). Later, a new complex envelope transformation $\mathcal{A} := A \exp(-\alpha z/2)$ will be performed to include the attenuation constant α in the left-hand side (LHS) of the PPE and isolating the nonlinear terms (sources) in the right-hand side (RHS).

Second step: Wave equation of the guided mode

From the classical modal analysis of a single-core fiber [6], it is worthy to note that the guided mode $F \exp(-\gamma(\omega)z)$ should satisfy the Helmholtz equation in each spectral component ω :

$$\left(\Delta + \frac{\omega^2}{c_0^2} \tilde{\epsilon}_r(\mathbf{r}, \omega) \right) F(x, y, \omega) \exp(-\gamma(\omega)z) = 0, \quad (22)$$

where $\tilde{\epsilon}_r$ is the Fourier transform of the relative electric permittivity of the fiber, including the core and cladding regions [see below Eq. (33) for more details]. In particular, the above equation is the step 2 of Fig. 2.

Third step: Nonlinear complex amplitude wave equation

In the third step, we should derive the nonlinear wave equation of the optical medium as a function of the complex amplitude in the frequency domain (\tilde{E}_{ω_0}). We start by combining the Faraday's and Ampère's laws [Eqs. (6) and (7)]. Using the constitutive relations given by Eq. (5), the nonlinear wave equation of the electric field strength is found to be:

$$\Delta \mathcal{E}(\mathbf{r}, t) - \frac{1}{c_0^2} \partial_t^2 \mathcal{E}(\mathbf{r}, t) = \mu_0 \partial_t^2 \mathcal{P}^{(1)}(\mathbf{r}, t) + \mu_0 \partial_t^2 \mathcal{P}^{(3)}(\mathbf{r}, t), \quad (23)$$

where $\mathcal{P}^{(1)}$ and $\mathcal{P}^{(3)}$ are the linear and nonlinear polarization of the fiber, respectively. Note that we have neglected the term $\nabla(\nabla \cdot (\mathcal{P}^{(1)} + \mathcal{P}^{(3)}))$ in Eq. (23) considering slowly-varying refractive index profiles⁶ and the low birefringent and low nonlinear nature of silica fibers. Unfortunately, this approximation has not been rigorously verified in the literature [2, 3, 4, 5]. For the above reason, we include in an appendix of [7] a numerical verification of this point.

Now, applying the Fourier transform to Eq. (23) we obtain:

$$\left(\Delta + \frac{\omega^2}{c_0^2} \right) \tilde{\mathcal{E}}(\mathbf{r}, \omega) = -\omega^2 \mu_0 \tilde{\mathcal{P}}^{(1)}(\mathbf{r}, \omega) - \omega^2 \mu_0 \tilde{\mathcal{P}}^{(3)}(\mathbf{r}, \omega). \quad (24)$$

⁶That is, $\delta_r n \ll n$ in $\delta r \sim \lambda$, where $\delta_r n := |n(r + \delta r) - n(r)|$.

Remember that the field $\tilde{\mathcal{E}}$ can be expressed in terms of \tilde{E}_{ω_0} by using Eq. (18). In a similar way, $\tilde{\mathcal{P}}^{(1)}$ and $\tilde{\mathcal{P}}^{(3)}$ can be written in terms of their complex amplitude in the frequency domain as:

$$\begin{aligned}\tilde{\mathcal{P}}^{(k)}(\mathbf{r}, \omega) &= \mathcal{F} \left[\mathcal{P}^{(k)}(\mathbf{r}, t) \right] \simeq \mathcal{F} \left[\operatorname{Re} \left\{ P_{\omega_0}^{(k)}(\mathbf{r}, t) \exp(j\omega_0 t) \right\} \hat{\tau} \right] = \operatorname{Re} \left\{ \tilde{P}_{\omega_0}^{(k)}(\mathbf{r}, \omega - \omega_0) \right\} \hat{\tau} \\ &= \frac{1}{2} \left[\tilde{P}_{\omega_0}^{(k)}(\mathbf{r}, \omega - \omega_0) + \tilde{P}_{-\omega_0}^{(k)}(\mathbf{r}, \omega + \omega_0) \right] \hat{\tau}; \quad k \in \{1, 3\}.\end{aligned}\quad (25)$$

where the nonlinear polarization term in $3\omega_0$ was omitted taking into account that the phase-matching condition in this nonlinear term is not satisfied in silica fibers [8]. Thus, Eq. (24) becomes:

$$\left(\Delta + \frac{\omega^2}{c_0^2} \right) \tilde{E}_{\omega_0}(\mathbf{r}, \omega - \omega_0) = -\omega^2 \mu_0 \tilde{P}_{\omega_0}^{(1)}(\mathbf{r}, \omega - \omega_0) - \omega^2 \mu_0 \tilde{P}_{\omega_0}^{(3)}(\mathbf{r}, \omega - \omega_0). \quad (26)$$

Now, let us discuss the constitutive relation between $\tilde{P}_{\omega_0}^{(1)}$ and \tilde{E}_{ω_0} . Modeling the linear response of the fiber ($\mathcal{P}^{(1)}$) to an incident electric field (\mathcal{E}) as a system: isotropic, spatially heterogeneous nondispersive and temporally invariant and dispersive, the linear polarization in the time domain $\mathcal{P}^{(1)}$ can be expressed as:

$$\mathcal{P}^{(1)}(\mathbf{r}, t) = \varepsilon_0 \int_{-\infty}^{\infty} \chi^{(1)}(\mathbf{r}, t - \zeta) \mathcal{E}(\mathbf{r}, \zeta) d\zeta, \quad (27)$$

with ε_0 the electric permittivity in vacuum and $\chi^{(1)}$ the first-order electric susceptibility. Hence, the linear polarization can be regarded as a linear and time-invariant system, with the constitutive relation of the linear polarization expressed in the frequency domain as:

$$\tilde{\mathcal{P}}^{(1)}(\mathbf{r}, \omega) = \varepsilon_0 \tilde{\chi}^{(1)}(\mathbf{r}, \omega) \tilde{\mathcal{E}}(\mathbf{r}, \omega), \quad (28)$$

where:

$$\tilde{\chi}^{(1)}(\mathbf{r}, \omega) := \mathcal{F} \left[\chi^{(1)}(\mathbf{r}, t) \right] = \int_{-\infty}^{\infty} \chi^{(1)}(\mathbf{r}, t) \exp(-j\omega t) dt. \quad (29)$$

It should be taken into account that the real part of $\tilde{\chi}^{(1)}$ gives information about the material dispersion and the imaginary part accounts for the optical absorption induced by the resonant frequencies of fused silica, located in the ultraviolet (68.4 nm and 116.2 nm) and infrared (9896.2 nm) bands [2]. In the third transmission window, the optical absorption is mainly induced by the Rayleigh scattering (modeled by α) and the imaginary part of $\tilde{\chi}^{(1)}$ can be neglected. In any case, the complex amplitude of the linear polarization in the frequency domain is found to be:

$$\tilde{P}_{\omega_0}^{(1)}(\mathbf{r}, \omega - \omega_0) = \varepsilon_0 \tilde{\chi}^{(1)}(\mathbf{r}, \omega) \tilde{E}_{\omega_0}(\mathbf{r}, \omega - \omega_0). \quad (30)$$

Now, we still need to investigate the nonlinear polarization term $\tilde{P}_{\omega_0}^{(3)}$ of Eq. (26). Nonetheless, the constitutive relation of the nonlinear polarization with the electric field strength involves a convolution operation in the frequency domain, increasing the complexity of the mathematical discussion to derive the PPE. Hence, for simplicity, we will maintain the term $\tilde{P}_{\omega_0}^{(3)}$ in Eq. (26), which becomes:

$$\left(\Delta + \frac{\omega^2}{c_0^2} \right) \tilde{E}_{\omega_0}(\mathbf{r}, \omega - \omega_0) + \frac{\omega^2}{c_0^2} \tilde{\chi}^{(1)}(\mathbf{r}, \omega) \tilde{E}_{\omega_0}(\mathbf{r}, \omega - \omega_0) = -\omega^2 \mu_0 \tilde{P}_{\omega_0}^{(3)}(\mathbf{r}, \omega - \omega_0). \quad (31)$$

Furthermore, defining $\tilde{\varepsilon}_r(\mathbf{r}, \omega) := 1 + \tilde{\chi}^{(1)}(\mathbf{r}, \omega)$ and omitting the imaginary part of $\tilde{\chi}^{(1)}$ as indicated before, the nonlinear complex amplitude wave equation is finally derived:

$$\left(\Delta + \frac{\omega^2}{c_0^2} \tilde{\varepsilon}_r(\mathbf{r}, \omega) \right) \tilde{E}_{\omega_0}(\mathbf{r}, \omega - \omega_0) = -\omega^2 \mu_0 \tilde{P}_{\omega_0}^{(3)}(\mathbf{r}, \omega - \omega_0). \quad (32)$$

Fourth step: Complex envelope propagation equation

In the fourth step, we should finally derive the PPE by using the results of step 1 [Eq. (19)] and step 2 [Eq. (22)] in step 3 [Eq. (32)]. However, the fourth step is not straightforward. We should previously indicate some relevant points about $\tilde{\varepsilon}_r(\mathbf{r}, \omega)$ and $\beta(\omega)$.

In particular, $\tilde{\varepsilon}_r(\mathbf{r}, \omega)$ gives information about the relative electric permittivity distribution in the core and cladding regions. It is related with the refractive index of the core (n_{co}) and cladding (n_{cl}) as indicated below:

$$\begin{aligned} \tilde{\varepsilon}_r(\mathbf{r}, \omega) &= \tilde{\varepsilon}_{r,\text{cl}}(\mathbf{r}, \omega) + \Delta\tilde{\varepsilon}_{r,\text{co}}(\mathbf{r}, \omega) \\ &= \begin{cases} \mathbf{r} \equiv \text{core} & \tilde{\varepsilon}_{r,\text{co}}(\mathbf{r}, \omega) = \tilde{\varepsilon}_{r,\text{cl}}(\mathbf{r}, \omega) + \Delta\tilde{\varepsilon}_{r,\text{co}}(\mathbf{r}, \omega) \equiv n_{\text{co}}^2(\mathbf{r}, \omega), \\ \mathbf{r} \equiv \text{cladding} & \tilde{\varepsilon}_{r,\text{cl}}(\mathbf{r}, \omega) \equiv n_{\text{cl}}^2(\mathbf{r}, \omega) \end{cases}, \end{aligned} \quad (33)$$

where $\tilde{\varepsilon}_{r,\text{co}}$ and $\tilde{\varepsilon}_{r,\text{cl}}$ are respectively the relative electric permittivity in the core and in the cladding, and $\Delta\tilde{\varepsilon}_{r,\text{co}}$ is the difference between $\tilde{\varepsilon}_{r,\text{co}}$ and $\tilde{\varepsilon}_{r,\text{cl}}$. Along this line note that we have assumed these functions spatial dependent to describe step and gradual-index fibers⁷. In addition, remember that $\tilde{\varepsilon}_r$ is assumed to be a real function taking into account that we have neglected the imaginary part of $\tilde{\chi}^{(1)}$ when operating in the third transmission window.

On the other hand, it is worthy to note that Eq. (22) allows us to obtain a fundamental relation to describe the chromatic dispersion. It is straightforward to derive this relation performing the next two steps. First, the phase constant β should be written as:

$$\beta(\omega) = \sum_{k=0}^{\infty} \frac{1}{k!} (\omega - \omega_0)^k \beta_k, \quad (34)$$

with $\beta_k := d^k \beta(\omega = \omega_0) / d\omega^k$. Second, we should approximate:

$$\begin{aligned} \beta^2(\omega) - \beta_0^2 &= (\beta(\omega) + \beta_0)(\beta(\omega) - \beta_0) \\ &\simeq 2\beta_0(\beta(\omega) - \beta_0) = 2\beta_0 \sum_{k=1}^{\infty} \frac{1}{k!} (\omega - \omega_0)^k \beta_k \equiv -j2\beta_0 \mathfrak{D}(\omega), \end{aligned} \quad (35)$$

where \mathfrak{D} is the complex function defined as:

$$\mathfrak{D}(\omega) := \sum_{k=1}^{\infty} \frac{j}{k!} (\omega - \omega_0)^k \beta_k. \quad (36)$$

Thus, using Eq. (35) we directly obtain from Eq. (22) [step 2] the expression that allows us to describe the chromatic dispersion during the propagation of the pulses:

$$\left(\Delta_{\text{T}} + \frac{\omega^2}{c_0^2} \tilde{\varepsilon}_r(\mathbf{r}, \omega) \right) F(x, y, \omega) = [\beta_0^2 - j2\beta_0 \mathfrak{D}(\omega) - j\alpha(\omega) \beta(\omega)] F(x, y, \omega), \quad (37)$$

with $\Delta_{\text{T}} = \partial_x^2 + \partial_y^2$ the transverse Laplacian operator.

Now, at this point we can start with the derivation of the PPE by inserting our ansatz [Eq. (19)] in the nonlinear complex amplitude wave equation [Eq. (32)] and using Eq. (37) during the algebraic work. This is the solid line connecting the steps 1 and 2 with step 3 which gives rise to step 4 in Fig. 2. As a result, after some algebraic work and retaining the second-order longitudinal derivatives of the complex envelopes considering that $\partial_z^2 \tilde{A} \neq 0$ in $\delta z \sim \lambda$

⁷The cladding can also be designed with a gradual-index profile (e.g. hollow fibers)

for femtosecond optical pulses (Appendix B4 of [7]), we obtain (the independent variables are omitted for simplicity):

$$F \exp(-j\beta_0 z) [\partial_z^2 - (j2\beta_0 + \alpha)(\partial_z + \mathfrak{D})] \tilde{A} = -\omega^2 \mu_0 \tilde{P}_{\omega_0}^{(3)} \exp\left(\frac{1}{2}\alpha z\right). \quad (38)$$

As can be seen, the LHS of Eq. (38) describes the linear propagation and the RHS accounts for the nonlinear effects (sources). In this vein, the derivation of the PPE requires to investigate the constitutive relation between the complex amplitude of the nonlinear polarization and the electric field strength in the frequency domain.

To this end, we should start from the constitutive relation in the time domain. Therefore, at this point let us remember the expressions of the nonlinear polarization in the time and frequency domain as a function of the corresponding complex amplitudes:

$$\mathcal{P}^{(3)}(\mathbf{r}, t) = \mathcal{P}^{(3)}(\mathbf{r}, t) \hat{\tau} \simeq \text{Re} \left\{ P_{\omega_0}^{(3)}(\mathbf{r}, t) \exp(j\omega_0 t) \right\} \hat{\tau}; \quad (39)$$

$$\tilde{\mathcal{P}}^{(3)}(\mathbf{r}, \omega) = \mathcal{F} \left[\mathcal{P}^{(3)}(\mathbf{r}, t) \right] \simeq \text{Re} \left\{ \tilde{P}_{\omega_0}^{(3)}(\mathbf{r}, \omega - \omega_0) \right\} \hat{\tau}, \quad (40)$$

with:

$$\tilde{P}_{\omega_0}^{(3)}(\mathbf{r}, \omega - \omega_0) = \mathcal{F} \left[P_{\omega_0}^{(3)}(\mathbf{r}, t) \exp(j\omega_0 t) \right], \quad (41)$$

and omitting the nonlinear polarization in $3\omega_0$, as indicated before. Hence, the nonlinear terms of Eq. (38) can be found by analyzing the constitutive relation $\mathcal{P}^{(3)}$ - \mathcal{E} in the time domain and performing the Fourier transform of the complex amplitudes.

In ultra-short optical pulses, the constitutive relation between the nonlinear polarization and the electric field strength should include the delay response of the electronic and nuclei structure of silica atoms when an electric field stimulates the optical medium. The most general expression to describe the third-order nonlinear response in silica media is to consider an output of a system: nonlinear, anisotropic, spatially homogeneous⁸ nondispersive, and temporally varying and dispersive. In such circumstances, the constitutive relation can be written as [Einstein summation convention, $(i, j, k, l) \in \{x, y, z\}^4$]:

$$\mathcal{P}_i^{(3)}(\mathbf{r}, t) = \varepsilon_0 \iiint_{-\infty}^{\infty} \chi_{ijkl}^{(3)}(t, \zeta_1, \zeta_2, \zeta_3) \mathcal{E}_j(\mathbf{r}, \zeta_1) \mathcal{E}_k(\mathbf{r}, \zeta_2) \mathcal{E}_l(\mathbf{r}, \zeta_3) d\zeta_1 d\zeta_2 d\zeta_3. \quad (42)$$

The time dispersive nature allows us to describe the exact time delay induced by the electronic and nuclei response of silica atoms to the incident electric field. For optical frequencies well below the electronic transitions, the electronic contribution to the nonlinear polarization can be considered instantaneous. However, since nucleons (protons and neutrons) are considerably heavier than electrons, the nuclei motions have resonant frequencies much lower than the electronic transitions and, consequently, they should be retained in the constitutive relation. Specifically, *Raman scattering* is a well-known effect arising from the nuclear contribution to the nonlinear polarization. Therefore, considering ultra-short optical pulses wider than 1 fs, the electronic response can be assumed instantaneous and Eq. (42) can be reduced to [3]:

$$\begin{aligned} \mathcal{P}_i^{(3)}(\mathbf{r}, t) &\simeq \mathcal{P}_i^{(3I)}(\mathbf{r}, t) + \mathcal{P}_i^{(3R)}(\mathbf{r}, t) \\ &= \varepsilon_0 \chi_{ijkl}^{(3I)}(t) \mathcal{E}_j(\mathbf{r}, t) \mathcal{E}_k(\mathbf{r}, t) \mathcal{E}_l(\mathbf{r}, t) + \varepsilon_0 \mathcal{E}_j(\mathbf{r}, t) \int_{-\infty}^{\infty} \chi_{ijkl}^{(3R)}(t - \zeta) \mathcal{E}_k(\mathbf{r}, \zeta) \mathcal{E}_l(\mathbf{r}, \zeta) d\zeta, \end{aligned} \quad (43)$$

⁸Although the nonlinear electric susceptibility can be found slightly different in each dielectric region of an optical fiber, we assume spatial homogeneity in $\chi_{ijkl}^{(3)}$ due to the low nonlinear nature of such optical media. In particular, we found that $\chi_{ijkl}^{(3)} \sim 10^{-22} \text{ m}^2/\text{V}^2$ in the core and cladding regions.

where the first term of the RHS describes the instantaneous response (3I) accounting for the electronic resonances, and the second term describes the nuclei motions inducing the intrapulse stimulated Raman scattering effect (3R). The third-order electric susceptibility tensors of the above equation can be expressed in silica fibers as follows [9]:

$$\chi_{ijkl}^{(3I)}(t) = \chi_{\text{NL}} \left(\frac{1 - f_{\text{R}}}{3} \right) (\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) \delta(t); \quad (44)$$

$$\chi_{ijkl}^{(3R)}(t) = \chi_{\text{NL}} f_{\text{R}} \left[h(t) \delta_{ij}\delta_{kl} + \frac{1}{2} u(t) (\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) \right], \quad (45)$$

where δ_{ij} is the Kronecker delta function; $\chi_{\text{NL}} = 2.6 \cdot 10^{-22} \text{ m}^2/\text{V}^2$ at the wavelength of 1550 nm; $f_{\text{R}} = 0.245$ represents the fractional contribution of the delayed Raman response to the nonlinear polarization; and h and u functions describe the isotropic and anisotropic Raman response, respectively [9]:

$$h(t) = f_1 \tau_1 (\tau_1^{-2} + \tau_2^{-2}) \exp(-t/\tau_2) \sin(t/\tau_1); \quad (46)$$

$$u(t) = f_2 \left(\frac{2\tau_3 - t}{\tau_3^2} \right) \exp(-t/\tau_3) + \frac{f_3}{f_1} h(t), \quad (47)$$

with $f_1, f_2, f_3, \tau_1, \tau_2$ and τ_3 constants of the nonlinear medium satisfying the following relations:

$$\chi_{\text{NL}} (1 - f_{\text{R}}) = \frac{4}{3} \varepsilon_0 c_0 n_{\text{NL}} \bar{n}^2; \quad \sum_{i=1}^3 f_i = 1; \quad \int_{-\infty}^{\infty} (\chi_{ijkl}^{(3I)}(t) + \chi_{ijkl}^{(3R)}(t)) dt = \chi_{\text{NL}}, \quad (48)$$

with \bar{n} the average value of the material refractive index of the fiber and n_{NL} the nonlinear refractive index modeling the changes induced in \bar{n} by the nonlinear polarization. In our case, we assume $\bar{n} \simeq 1.45$ and $n_{\text{NL}} = 2.6 \cdot 10^{-20} \text{ m}^2/\text{W}$. From Eq. (48) and Ref. [9] we found: $f_1 = 0.75$, $f_2 = 0.21$, $f_3 = 0.04$, $\tau_1 = 12.2 \text{ fs}$, $\tau_2 = 32 \text{ fs}$, $\tau_3 = 96 \text{ fs}$. As shown in the Supplemental Material of [10], we should note that the isotropic Raman response predominates over the anisotropic response due to the molecular symmetry of the SiO_2 .

However, our scenario omits the polarization effects. Therefore, taking into account this point and using Eqs. (39), (43) and (44), it is straightforward to obtain the complex amplitude of the instantaneous nonlinear polarization in the time domain:

$$P_{\omega_0}^{(3I)} = \varepsilon_0 \gamma_{\text{I}} |E_{\omega_0}|^2 E_{x, \omega_0}, \quad (49)$$

where $\gamma_{\text{I}} := (3/4) \chi_{\text{NL}} (1 - f_{\text{R}}) = 1.5 \cdot 10^{-22} \text{ m}^2/\text{V}^2$ at the wavelength of 1550 nm. The term $3/4$ is inferred from Eq. (43) by using the intrinsic permutation symmetry of $\chi_{ijkl}^{(3)}$. A more detailed description of this point can be found in [8].

Furthermore, the complex amplitude of the nonlinear polarization modeling the nuclei motions (Raman response) can also be found by using Eqs. (39), (43) and (45):

$$P_{\omega_0}^{(3R)}(\mathbf{r}, t) = \frac{1}{4} \varepsilon_0 \chi_{\text{NL}} f_{\text{R}} \left\{ 2E_{\omega_0}(\mathbf{r}, t) \int_{-\infty}^{\infty} (h + u)(t - \zeta) |E_{\omega_0}(\mathbf{r}, \zeta)|^2 d\zeta \right. \\ \left. + E_{-\omega_0}(\mathbf{r}, t) \exp(-j2\omega_0 t) \int_{-\infty}^{\infty} (h + u)(t - \zeta) E_{\omega_0}^2(\mathbf{r}, \zeta) \exp(j2\omega_0 \zeta) d\zeta \right\}. \quad (50)$$

including in this case the independent variables to clarify the mathematical discussion. In order to simplify Eq. (50), we can use the commutation property of the convolution. In this way, the last term of the RHS can be expressed as ($f := h + u$):

$$\begin{aligned}
& E_{-\omega_0}(\mathbf{r}, t) \exp(-j2\omega_0 t) \int_{-\infty}^{\infty} f(t - \zeta) E_{\omega_0}^2(\mathbf{r}, \zeta) \exp(j2\omega_0 \zeta) d\zeta \\
&= E_{-\omega_0}(\mathbf{r}, t) \exp(-j2\omega_0 t) \int_{-\infty}^{\infty} f(\zeta) E_{\omega_0}^2(\mathbf{r}, t - \zeta) \exp(j2\omega_0(t - \zeta)) d\zeta \\
&= E_{-\omega_0}(\mathbf{r}, t) \int_{-\infty}^{\infty} f(\zeta) \exp(-j2\omega_0 \zeta) E_{\omega_0}^2(\mathbf{r}, t - \zeta) d\zeta \\
&= E_{-\omega_0}(\mathbf{r}, t) [(f(t) \exp(-j2\omega_0 t)) * E_{\omega_0}^2(\mathbf{r}, t)]. \tag{51}
\end{aligned}$$

As can be noted, Eq. (51) involves a convolution of the modulated Raman response with the complex amplitudes of the electric field strength. Considering that the bandwidth of the Raman response (given by the f function) is around 15 THz [9], centered at $-\omega_0 \approx -380$ THz, and the bandwidth of the complex amplitudes is lower than 100 THz for ultra-short pulses wider than 10 fs (in baseband $\Omega := \omega - \omega_0$), the Fourier transform of the convolution is found to be null:

$$\begin{aligned}
& \mathcal{F} [(f(t) \exp(-j2\omega_0 t)) * E_{\omega_0}^2(\mathbf{r}, t)] = \\
&= \frac{1}{2\pi} \tilde{F}(\Omega + 2\omega_0) [\tilde{E}_{\omega_0}(\mathbf{r}, \Omega) * \tilde{E}_{\omega_0}(\mathbf{r}, \Omega)] = 0, \tag{52}
\end{aligned}$$

and therefore, the last term of the RHS of Eq. (50) can be neglected. Finally, defining the nonlinear constant $\gamma_R := 0.5\chi_{NL}f_R = 3.2 \cdot 10^{-23} \text{ m}^2/\text{V}^2$ at the wavelength of 1550 nm, the complex amplitude of the nonlinear polarization modeling both electronic and nuclei responses is found to be:

$$P_{\omega_0}^{(3)}(\mathbf{r}, t) = \varepsilon_0 \gamma_I |E_{\omega_0}(\mathbf{r}, t)|^2 E_{\omega_0}(\mathbf{r}, t) + \varepsilon_0 \gamma_R (f(t) * |E_{\omega_0}(\mathbf{r}, t)|^2) E_{\omega_0}(\mathbf{r}, t). \tag{53}$$

Once we have derived the complex amplitude of the nonlinear polarization in the time domain, Eq. (38) can be completed using Eqs. (41) and (53). At this point, note that the nonlinear constants γ_I and γ_R are also frequency dependent when operating with ultra-short optical pulses in the femtosecond regime. However, we assume that the frequency variation of these parameters is much lower than their average value in the pulse bandwidth, as same as the frequency changes of F . Hence, Eq. (38) becomes (for the sake of simplicity, the independent variables are only included in the convolution operations of the nonlinear terms):

$$\begin{aligned}
& F \exp(-j\beta_0 z) \left[\partial_z^2 \tilde{A} - (j2\beta_0 + \alpha) (\partial_z \tilde{A} + \mathfrak{D} \tilde{A}) \right] \\
&+ \frac{\omega^2}{c_0^2} \gamma_I \exp(-\alpha z) \exp(-j\beta_0 z) F^3 \mathcal{F} \left\{ A |A|^2 \exp(j\omega_0 t) \right\} \\
&+ \frac{\omega^2}{c_0^2} \gamma_R \exp(-\alpha z) \exp(-j\beta_0 z) F^3 \mathcal{F} \left\{ A [f(t) * |A(z, t)|^2] \exp(j\omega_0 t) \right\} = 0. \tag{54}
\end{aligned}$$

From Eq. (54), the PPE can be found by multiplying by $F \exp(j\beta_0 z)$ and integrating in an infinite cross-sectional area of the fiber:

$$\begin{aligned}
j \left(\frac{j}{2\beta_0} \partial_z^2 + \partial_z + \mathfrak{D} \right) \tilde{A} = \exp(-\alpha z) \left\{ \tilde{q}^{(I)} \mathcal{F} \left\{ A |A|^2 \exp(j\omega_0 t) \right\} \right. \\
\left. + \tilde{q}^{(R)} \mathcal{F} \left\{ A [f(t) * |A(z, t)|^2] \exp(j\omega_0 t) \right\} \right\}, \tag{55}
\end{aligned}$$

where $\tilde{q}^{(I)}$ and $\tilde{q}^{(R)}$ are the nonlinear mode-coupling coefficients (MCCs) defined as:

$$\tilde{q}^{(S)}(\omega) := \frac{\omega^2}{2c_0^2\beta_0} \frac{\iint_{\infty} \gamma_S(\omega) F^4(x, y, \omega) dx dy}{\iint_{\infty} F^2(x, y, \omega) dx dy}; \quad S \in \{I, R\}. \quad (56)$$

Note that the frequency dependence of the nonlinear MCCs also involves the nonlinear dispersion of γ_I and γ_R , induced by the frequency dependence of n_{NL} . In such a scenario, the function n_{NL} can be approximated by a first-order Taylor series expansion as $n_{NL}(\omega) \simeq n_{NL,0} + (\omega - \omega_0) n_{NL,1}$, where $n_{NL,0} = 2.6 \cdot 10^{-20} \text{ m}^2/\text{W}$ and $n_{NL,1} = 8.3 \cdot 10^{-24} \text{ ps}\cdot\text{m}^2/\text{W}$ in silica fibers [10]. Now, taking into account that $(1/2\beta_0) \left| \partial_z^2 \tilde{A} \right| \ll \left| \partial_z \tilde{A} \right|$ as demonstrated in [7], redefining the complex envelopes in the frequency domain as $\tilde{A} := \tilde{\mathcal{A}} \exp(\alpha z/2)$ and assuming the attenuation coefficient α with low frequency dependence satisfying $\alpha(\omega_0) \gg d\alpha(\omega = \omega_0)/d\omega$, Eq. (55) can be reduced to:

$$j \left(\partial_z + \mathfrak{D} + \frac{\alpha}{2} \right) \tilde{\mathcal{A}} = \tilde{q}^{(I)} \mathcal{F} \left\{ \mathcal{A} |\mathcal{A}|^2 \exp(j\omega_0 t) \right\} + \tilde{q}^{(R)} \mathcal{F} \left\{ \mathcal{A} \left[f(t) * |\mathcal{A}(z, t)|^2 \right] \exp(j\omega_0 t) \right\}. \quad (57)$$

In order to derive the final expression of the PPE in the time domain, we should perform the following Taylor series expansion at $\omega = \omega_0$ of the holomorphic functions (we denote the instantaneous and Raman nonlinear MCCs by the superindex $S \in \{I, R\}$):

$$\tilde{q}^{(S)}(\omega) = \sum_{n=0}^{\infty} \frac{1}{n!} (\omega - \omega_0)^n \left. \frac{d^n \tilde{q}^{(S)}(\omega)}{d\omega^n} \right|_{\omega_0} \equiv \sum_{n=0}^{\infty} \frac{1}{n!} (\omega - \omega_0)^n \tilde{q}_n^{(S)}; \quad (58)$$

$$\alpha(\omega) \simeq \alpha(\omega_0) + (\omega - \omega_0) \left. \frac{d\alpha(\omega)}{d\omega} \right|_{\omega_0} \equiv \alpha_0 + (\omega - \omega_0) \alpha_1, \quad (59)$$

which can be expressed in the time domain by the following linear operators:

$$\hat{q}^{(S)} := \sum_{n=0}^{\infty} \frac{(-j)^n}{n!} \tilde{q}_n^{(S)} \partial_t^n \quad \hat{\alpha} := \alpha_0 - j\alpha_1 \partial_t, \quad (60)$$

and the complex function \mathfrak{D} is expressed in the time domain by the dispersion operator:

$$\hat{\mathfrak{D}} := \sum_{n=1}^{\infty} \frac{(-j)^{n-1}}{n!} \beta_n \partial_t^n. \quad (61)$$

Finally, applying the inverse Fourier transform to Eq. (57) the final expression of the PPE is derived in the time domain:

$$j \left(\partial_z + \hat{\mathfrak{D}} + \frac{1}{2} \hat{\alpha} \right) \mathcal{A}(z, t) = \hat{q}^{(I)} \left(|\mathcal{A}(z, t)|^2 \mathcal{A}(z, t) \right) + \hat{q}^{(R)} \left[\left(f(t) * |\mathcal{A}(z, t)|^2 \right) \mathcal{A}(z, t) \right]. \quad (62)$$

From this equation, the following remarks are in order:

- In order to reduce the computational complexity of the PPE, $\widehat{\mathbf{D}}$ and $\hat{q}^{(1,R)}$ can be calculated in optical fibers by using a third- and first-order Taylor series expansion, respectively [10].
- The units of $\mathcal{A}(z, t)$ are V/m. If we are interested in the optical power propagated by the optical pulse in the LP₀₁ mode, we must integrate the time average of the Poynting vector ($\mathbf{S} = \mathbf{E} \times \mathbf{H}$) in a period of the optical carrier ($T_0 = 2\pi/\omega_0$) and in an infinite cross-sectional area of the fiber, which can be expressed as a function of the complex envelope as follows:⁹

$$P(z, t) = \iint_{\infty} \langle \mathbf{E} \times \mathbf{H} \rangle \cdot \hat{z} dx dy = \mathcal{C}^{(P)} |\mathcal{A}(z, t)|^2, \quad (63)$$

where:¹⁰

$$\mathcal{C}^{(P)} := \iint_{\infty} \frac{1}{2\eta(x, y, \omega_0)} F^2(x, y, \omega_0) dx dy, \quad (64)$$

with η the intrinsic impedance of the medium, which can be calculated as a function of the intrinsic impedance in vacuum $\eta_0 = 120\pi$ (Ω) as $\eta(x, y, \omega_0) = \eta_0/n_{co}(x, y, \omega_0)$ in the core and $\eta(x, y, \omega_0) = \eta_0/n_{cl}(x, y, \omega_0)$ in the cladding.

3 Pulse propagation in uniform dielectric media

Let us now consider other basic scenario of nonlinear optics: a pulsed plane wave propagating along the z axis through an uniform (or unguided¹¹) nonlinear dielectric medium. The mathematical derivation of the PPE is similar to the previous section. However, the following points should be taken into account in each step depicted in Fig. 2.

First step: Ansatz of the electric field strength of the medium

The ansatz of Maxwell's equations of a pulsed plane wave propagating in an uniform medium is similar to an optical pulse propagating through an optical fiber. Nevertheless, the weakly-guiding approximation is not required in this case. Without loss of generality, we can consider only a single component of the electric field strength when assuming a pulsed plane wave propagated through an uniform (\equiv homogeneous) isotropic optical medium. In this way, the ansatz of step 1 is described by the following equations in the time domain:

$$\mathcal{E}(\mathbf{r}, t) = \mathcal{E}(\mathbf{r}, t) \hat{\tau} \simeq \text{Re} \{ E_{\omega_0}(\mathbf{r}, t) \exp(j\omega_0 t) \} \hat{\tau}; \quad (65)$$

$$E_{\omega_0}(\mathbf{r}, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \tilde{A}(\mathbf{r}, \omega - \omega_0) \exp\left(-j\beta_0 z - \frac{1}{2}\alpha(\omega) z\right) \exp(j(\omega - \omega_0) t) d\omega, \quad (66)$$

and in the frequency domain:

$$\tilde{\mathcal{E}}(\mathbf{r}, \omega) \simeq \text{Re} \left\{ \tilde{E}_{\omega_0}(\mathbf{r}, \omega - \omega_0) \right\} \hat{\tau}; \quad (67)$$

$$\tilde{E}_{\omega_0}(\mathbf{r}, \omega - \omega_0) = \tilde{A}(\mathbf{r}, \omega - \omega_0) \exp\left(-j\beta_0 z - \frac{1}{2}\alpha(\omega) z\right). \quad (68)$$

⁹Note that the complex envelope is assumed to be constant in a period of the optical carrier. Thus, in this case, the time average operator only applies over the rapidly-varying exponential terms of \mathcal{E} and \mathcal{H} .

¹⁰In the single-mode regime F is a real function, that is, $F = F^*$.

¹¹The terminology *unguided* medium can also be found in the literature as *uniform* medium (homogeneous refractive index). This concept is intimately related in optical communications with a *wireless* propagation.

It should be noted that we have included the spatial dependence of \tilde{A} not only in the z axis, but also in the transverse coordinates (x, y) to include the diffraction of the optical pulse during the propagation. In addition, note that $F \equiv 1$ and $\beta_0 \equiv k_z(\omega_0)$ in unguided media, where k_z is the z -component of the wavevector.

Second step: Wave equation of the guided mode

There is not a guided mode (\equiv bound state). Thus, the second step [Eq. (22)] is irrelevant in this case. We do not need to replace a term of the form $\Delta_T F$ [Eq. (37)] in the fourth step.

Third step: Nonlinear complex amplitude wave equation

The nonlinear complex amplitude wave equation is found to be the same as Eq. (32) given that the linear constitutive relation $\mathcal{P}^{(1)}\text{-}\mathcal{E}$ is the same as in an optical fiber. Nonetheless, in this case it is more convenient to retain the imaginary part of $\chi^{(1)}$. In silica fibers, $\text{Im}\{\chi^{(1)}\}$ can be neglected in the third transmission window because we operate far from the resonance frequencies of the medium. Unfortunately, this assumption is not general and should be discarded in this scenario. In addition, accordingly with the unguided nature of the optical propagation, we should also consider a homogeneous $\chi^{(1)}$ distribution. As a result, the nonlinear complex amplitude wave equation is found to be:

$$\left(\Delta + \frac{\omega^2}{c_0^2} \tilde{\varepsilon}_r(\omega)\right) \tilde{E}_{\omega_0}(\mathbf{r}, \omega - \omega_0) = -\omega^2 \mu_0 \tilde{P}_{\omega_0}^{(3)}(\mathbf{r}, \omega - \omega_0), \quad (69)$$

with:

$$\frac{\omega^2}{c_0^2} \tilde{\varepsilon}_r(\omega) = \frac{\omega^2}{c_0^2} \left(1 + \tilde{\chi}^{(1)}(\omega)\right) \equiv \left(\beta(\omega) - j\frac{1}{2}\alpha(\omega)\right)^2, \quad (70)$$

and $\beta(\omega) = \beta_0 - j\mathfrak{D}(\omega)$.

Fourth step: Complex envelope propagation equation

Following the same procedure as in the fourth step in optical fibers, i.e., replacing the results of step 1 [Eq. (68)] in step 3 [Eq. (69)], we will be able to derive the PPE in unguided media. More specifically, Eq. (38) becomes:

$$\exp(-j\beta_0 z) \left[\Delta_T + \partial_z^2 - (j2\beta_0 + \alpha)(\partial_z + \mathfrak{D})\right] \tilde{A} = -\omega^2 \mu_0 \tilde{P}_{\omega_0}^{(3)} \exp\left(\frac{1}{2}\alpha z\right), \quad (71)$$

with a new term $\Delta_T \tilde{A}$ accounting for the diffraction of the complex envelope. Once again, if we assume that the pulse is propagated through a dielectric media with electronic absorption resonances well above the frequencies of the optical fields, $\tilde{P}_{\omega_0}^{(3)}$ can be written as in optical fibers [Eq. (43)]: an instantaneous response accounting for the electronic and a delayed response induced by the nuclei motions [3]. In addition, note that the term $\tilde{P}_{3\omega_0}^{(3)}$ can also be neglected if we consider that $|\text{Re}\{\tilde{\chi}^{(1)}(3\omega) - \tilde{\chi}^{(1)}(\omega)\}| \gg 0$ due to the temporal dispersive nature of the optical medium [8]. All in all, the PPE is finally derived of the form:

$$j \left(\frac{j}{2\beta_0} \Delta_T + \partial_z + \hat{\mathbf{D}} + \frac{1}{2}\hat{\alpha}\right) \mathcal{A}(\mathbf{r}, t) = \hat{\mathbf{q}}^{(I)} \left(|\mathcal{A}(\mathbf{r}, t)|^2 \mathcal{A}(\mathbf{r}, t)\right) + \hat{\mathbf{q}}^{(R)} \left[\left(f(t) * |\mathcal{A}(\mathbf{r}, t)|^2\right) \mathcal{A}(\mathbf{r}, t)\right]. \quad (72)$$

Along this line, the following points should be taken into account:

- The linear operators $\hat{q}^{(I)}$ and $\hat{q}^{(R)}$ describe the frequency dependence of the nonlinear MCCs $\tilde{q}^{(I)}(\omega)$ and $\tilde{q}^{(R)}(\omega)$ in the time domain, respectively. However, given that $F \equiv 1$, Eq. (56) becomes:

$$\tilde{q}^{(S)}(\omega) = \frac{\omega^2 \gamma_S(\omega)}{2c_0^2 \beta_0}; \quad S \in \{I, R\}. \quad (73)$$

- The term $(j/2\beta_0)\partial_z^2 \mathcal{A}$ has been neglected in the LHS of the PPE as in the previous section because $(1/2\beta_0)|\partial_z^2 \mathcal{A}| \ll |\partial_z \mathcal{A}|$. Nevertheless, the term $(j/2\beta_0)\Delta_T \mathcal{A}$ cannot be neglected because, in general, $(1/2\beta_0)|\Delta_T \mathcal{A}|$ can be found of the same order (or higher) as $|\partial_z \mathcal{A}|$. Remarkably, this term accounts for the pulse diffraction, as commented before.
- The linear operators \hat{D} and $\hat{q}^{(I,R)}$ can also be calculated by using a *finite* Taylor series expansion. Unfortunately, the required order of the Taylor series in each term is not general. It depends on the particular dielectric medium simulated in the numerical calculations (see [11] for more details).
- The units of $\mathcal{A}(\mathbf{r}, t)$ are V/m. If we are interested in the optical *intensity* (W/m²) propagated by the pulsed plane wave, we must calculate the module of the time average of the Poynting vector in a period of the optical carrier ($T_0 = 2\pi/\omega_0$):¹²

$$I(\mathbf{r}, t) = \|\langle \mathcal{S} \rangle\| = \langle \mathcal{E} \times \mathcal{H} \rangle \cdot \hat{z} = \frac{1}{2\eta(\omega_0)} |\mathcal{A}(\mathbf{r}, t)|^2, \quad (74)$$

with $\eta(\omega_0) = \eta_0/n(\omega_0)$ and $n(\omega_0) := \sqrt{1 + \text{Re}\{\tilde{\chi}^{(1)}(\omega_0)\}}$. Of course, we do not calculate the optical power of a pulsed plane wave because is infinite (note that a plane wave impinges an infinite transverse plane).

4 White Light Continuum (WLC) in gases

Following [12] as a guideline, it should be noted that the ideal phase constant $\beta \equiv k_z$ of Section 3 is *perturbed* by the presence of the plasma. Therefore, we can use the same approach as in [7, 10], i.e., modeling the plasma as a *perturbation* of the optical medium. To clarify this approach, let us perform once again a detailed review of the derivation of the PPE from Maxwell's equations. The steps are similar to Fig. 2.

First step: Ansatz of the electric field strength of the medium

The ansatz of Maxwell's equations of a pulsed plane wave propagating in this scenario is similar to the ansatz of the previous section. Nevertheless, we should include the perturbation of the plasma in the propagation constant $\gamma = \alpha/2 + j\beta$. To this end, we should assume the electric field strength in the time domain of the form:

$$\mathcal{E}(\mathbf{r}, t) = \mathcal{E}(\mathbf{r}, t) \hat{\tau} \simeq \text{Re}\{E_{\omega_0}(\mathbf{r}, t) \exp(j\omega_0 t)\} \hat{\tau}; \quad (75)$$

$$E_{\omega_0}(\mathbf{r}, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \tilde{A}(\mathbf{r}, \omega - \omega_0) \exp\left(-j\beta_0^{(\text{eq})} z - \frac{1}{2}\alpha(\omega) z\right) \exp(j(\omega - \omega_0)t) d\omega, \quad (76)$$

¹²As in the previous section, the complex envelope is assumed to be constant in a period of the optical carrier. Thus, the time average operator only applies over the rapidly-varying exponential terms of \mathcal{E} and \mathcal{H} .

and in the frequency domain:

$$\tilde{\mathcal{E}}(\mathbf{r}, \omega) \simeq \text{Re} \left\{ \tilde{E}_{\omega_0}(\mathbf{r}, \omega - \omega_0) \right\} \hat{\tau}; \quad (77)$$

$$\tilde{E}_{\omega_0}(\mathbf{r}, \omega - \omega_0) = \tilde{A}(\mathbf{r}, \omega - \omega_0) \exp \left(-j\beta_0^{(\text{eq})} z - \frac{1}{2}\alpha(\omega) z \right). \quad (78)$$

with $\beta_0^{(\text{eq})} := \beta^{(\text{eq})}(\omega = \omega_0)$, where $\beta^{(\text{eq})}(\omega)$ is the *equivalent* phase constant at each spectral component ω accounting for the ideal phase constant $\beta(\omega)$ and the perturbation induced by the plasma $\beta^{(\text{PL})}(\omega)$:

$$\beta^{(\text{eq})}(\omega) := \beta(\omega) + \beta^{(\text{PL})}(\omega). \quad (79)$$

Second step: Wave equation of the guided mode

There is not a guided mode (\equiv bound state). Therefore, the second step [Eq. (22)] is irrelevant in this case. We do not need to replace a term of the form $\Delta_{\text{T}} F$ [Eq. (37)] in the fourth step. Here, we have the same conclusions as in the second step of Section 3.

Third step: Nonlinear complex amplitude wave equation

The nonlinear complex amplitude wave equation is found to be the same as Eq. (69) given that the linear constitutive relation $\mathcal{P}^{(1)}\text{-}\mathcal{E}$ is the same as in the previous section. We repeat this equation for clarity:

$$\left(\Delta + \frac{\omega^2}{c_0^2} \tilde{\varepsilon}_{\text{r}}(\omega) \right) \tilde{E}_{\omega_0}(\mathbf{r}, \omega - \omega_0) = -\omega^2 \mu_0 \tilde{P}_{\omega_0}^{(3)}(\mathbf{r}, \omega - \omega_0), \quad (80)$$

with:

$$\frac{\omega^2}{c_0^2} \tilde{\varepsilon}_{\text{r}}(\omega) = \frac{\omega^2}{c_0^2} \left(1 + \tilde{\chi}^{(1)}(\omega) \right) \equiv \left(\beta^{(\text{eq})}(\omega) - j\frac{1}{2}\alpha(\omega) \right)^2. \quad (81)$$

It is worthy to note that $\tilde{\varepsilon}_{\text{r}}$ involves now the contribution of the plasma via $\beta^{(\text{eq})}$.

Fourth step: Complex envelope propagation equation

Following the same procedure as in the fourth step of previous examples, we should replace the results of step 1 [Eq. (78)] in step 3 [Eq. (80)]. Thus, in this case Eq. (71) becomes:

$$\exp(-j\beta_0 z) \left[\Delta_{\text{T}} + \partial_z^2 - (j2\beta_0 + \alpha) \left(\partial_z + j\beta_0^{(\text{PL})} + \mathfrak{D}^{(\text{eq})} \right) \right] \tilde{A} = -\omega^2 \mu_0 \tilde{P}_{\omega_0}^{(3)} \exp \left(\frac{1}{2}\alpha z \right), \quad (82)$$

with:

$$\mathfrak{D}^{(\text{eq})}(\omega) := \sum_{k=1}^{\infty} \frac{j}{k!} (\omega - \omega_0)^k \beta_k^{(\text{eq})}, \quad (83)$$

and $\beta_k^{(\text{eq})} := d^k \beta^{(\text{eq})}(\omega = \omega_0) / d\omega^k$. According to [12], the term $\beta_0^{(\text{PL})} := \beta^{(\text{PL})}(\omega_0)$ will be the predominant perturbation of the plasma during the propagation of the optical pulse. The other terms $\beta_{k>0}^{(\text{PL})}$, included in $\mathfrak{D}^{(\text{eq})}$, describe the higher-order dispersion terms of the plasma. Concretely, the holomorphic complex function $\mathfrak{D}^{(\text{eq})}$ can be modeled in the time domain by the equivalent dispersion operator $\hat{\text{D}}^{(\text{eq})}$ defined as:

$$\hat{\text{D}}^{(\text{eq})} := \sum_{k=1}^{\infty} \frac{(-j)^{k-1}}{k!} \beta_k^{(\text{eq})} \partial_t^k. \quad (84)$$

Taking into account that $\beta_k^{(\text{eq})} = \beta_k + \beta_k^{(\text{PL})}$, we can rewrite $\widehat{\mathbf{D}}^{(\text{eq})}$ as the linear contribution of the ideal dispersion operator $\widehat{\mathbf{D}}$ and the higher-order dispersion terms of the plasma, modeled by $\widehat{\mathbf{D}}^{(\text{PL})}$, defined as:

$$\widehat{\mathbf{D}}^{(\text{PL})} := \sum_{k=1}^{\infty} \frac{(-j)^{k-1}}{k!} \beta_k^{(\text{PL})} \partial_t^k. \quad (85)$$

That is, we separate the dispersion induced by the frequency dependence of the ideal phase constant and the plasma phase constant: $\widehat{\mathbf{D}}^{(\text{eq})} = \widehat{\mathbf{D}} + \widehat{\mathbf{D}}^{(\text{PL})}$.

Now, if we assume that $\widetilde{P}_{\omega_0}^{(3)}$ can be written as in previous sections, i.e., with an instantaneous response accounting for the electronic and a delayed response induced by the nuclei motions, the PPE is derived in the time domain of the form:

$$\begin{aligned} j \left(\frac{j}{2\beta_0} \Delta_{\text{T}} + \partial_z + \widehat{\mathbf{D}} + \frac{1}{2} \widehat{\alpha} \right) \mathcal{A}(\mathbf{r}, t) &= \beta_0^{(\text{PL})} \mathcal{A}(\mathbf{r}, t) - j \widehat{\mathbf{D}}^{(\text{PL})} \mathcal{A}(\mathbf{r}, t) \\ &+ \hat{q}^{(\text{I})} \left(|\mathcal{A}(\mathbf{r}, t)|^2 \mathcal{A}(\mathbf{r}, t) \right) \\ &+ \hat{q}^{(\text{R})} \left[\left(f(t) * |\mathcal{A}(\mathbf{r}, t)|^2 \right) \mathcal{A}(\mathbf{r}, t) \right]. \end{aligned} \quad (86)$$

Finally, from the above equation the following points are in order:

- The linear operators $\hat{q}^{(\text{I})}$ and $\hat{q}^{(\text{R})}$ describe the frequency dependence of the nonlinear MCCs $\tilde{q}^{(\text{I})}(\omega)$ and $\tilde{q}^{(\text{R})}(\omega)$ in the time domain, respectively. These nonlinear MCCs should be calculated as indicated in Eq. (73).
- The linear operators can be calculated by using a *finite* Taylor series expansion. The minimum order required to describe accurately the optical medium and the plasma should be investigated.
- The units of $\mathcal{A}(\mathbf{r}, t)$ are V/m. If we are interested in the optical *intensity* (W/m²) propagated by the pulsed plane wave, we can use Eq. (74) to this end. However, note that the intrinsic impedance $\eta(\omega_0)$ must be calculated taking into account the plasma contribution. Specifically, the optical intensity is found to be:

$$I(\mathbf{r}, t) = \frac{1}{2\eta(\omega_0)} |\mathcal{A}(\mathbf{r}, t)|^2; \quad (87)$$

$$\eta(\omega_0) = \frac{\eta_0}{\sqrt{1 + \text{Re}\{\tilde{\chi}^{(1)}(\omega_0)\}}} = \frac{\eta_0}{n(\omega_0) + \Delta n^{(\text{PL})}(\omega_0)}. \quad (88)$$

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