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Theory and mathematics modeling of ultrashort pulse propagation

Computational Methods for Nonlinear PDEs describing Ultrashort Optical Pulse Propagation

A. Couairon[1,a] and M. Kolesik[2,3,b]

[1] Centre de Physique Théorique, CNRS, Ecole Polytechnique, F-91128 Palaiseau, France
[2] College of Optical Sciences, Tucson, Arizona 85721, USA
[3] Department of Physics, Constantine the Philosopher University, Nitra, Slovakia

Abstract. The purpose of these notes is to provide practical introduction into numerical modeling of ultrashort optical pulses in extreme nonlinear regimes. The theoretic background section covers derivation of modern pulse propagation models starting from Maxwell's equations, and includes both envelope-based models and carrier-resolving propagation equations. We then continue with a detailed description of implementation in software of Nonlinear Envelope Equations as an example of a mixed approach which combines finite-difference and spectral techniques. Fully spectral numerical solution methods for the Unidirectional Pulse Propagation Equation are discussed next. The modeling part of this text concludes with a brief introduction into efficient implementations of nonlinear medium responses.

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[a] e-mail: couairon@cpht.polytechnique.fr
[b] e-mail: kolesik@optics.arizona.edu

1 Introduction

As femtosecond lasers become available to many groups, simulation of propagation of optical pulses of ultra-short duration and their interaction with media gains new importance. Modern experiments can barely exist without the support from modeling and simulation in order to understand and interpret measured data. This is especially true as nonlinear optics continues to explore ever more extreme regimes [1–3].

As a result of this development, the need to perform sophisticated simulations widens considerably. However, the development of software for numerical experiments requires specific knowledge, time and means that too often constitute a barrier between practitioners of real experiments and their modeling needs.

Fortunately, simulation in *ultrafast nonlinear optics* has reached a degree of maturity at which it makes sense for the community to have certain *standard tools* that can be used in support of state of the art experiments. Alongside researchers mainly involved in computer simulation, informed non-specialists have been more and more engaged in numerical modeling [4, 5]. In line with these trends, these notes target those working in the field of ultrafast nonlinear optics, who need to perform numerical simulation with proper understanding of modeling, implementation, and numerical issues. This is therefore a didactic- and instruction-motivated text which combines a detailed overview including necessary theoretical background, and a number of opportunities to hone practical skills through a set of examples presented during the lectures.

The physics governing the effects we aim to understand and model in ultrafast nonlinear optics belong to several broader fields: classical and quantum optics, electromagnetism, plasma physics, solid state physics. This rich physics results from laser-matter interaction in the *ultrashort* pulse regime, i.e. with sub-picosecond durations ($T \leq 10^{-12}$ s), typically produced by the Chirped Pulse Amplification technique [6]. Nowadays, lasers that produce extremely powerful (1 PW $\equiv 10^{15}$ W) ultrashort pulses are developed. Very high light intensities in these pulses induce extreme *nonlinear effects* in any condensed material or gaseous medium. In this text, we concentrate on the regime of intensities up to 10^{15} W/cm^2 obtained e.g. by focusing a pulse with power in the MW-TW range in a transparent dielectric medium (gas, liquid or solid). Our rationale is that in this regime, the *propagation* effects are as significant as the *interaction* effects. Laser-matter interaction at these intensities can induce nonlinear refraction index change or lead to partial ionization of the dielectric medium, however we do not consider the regime of interaction with fully ionized plasmas. Another boundary in nonlinear optics is given by the definition of *ultrashort*, or sub-picosecond times; they typically correspond to time scales above which the response of matter to laser excitation starts to involve heat transfer, relaxation processes and hydrodynamic phenomena. The pulse durations we consider are shorter than the time scales for these phenomena.

Ultrashort laser pulse filamentation constitutes an example of physical phenomena in ultrafast nonlinear optics where approaches described in these notes apply particularly well. It denotes a specific regime of nonlinear propagation of intense laser pulses

$\mathcal{I} \sim 10^{13}$–$10^{14}\mathrm{W/cm}^2$ with narrow beam widths, over distances much larger than a typical diffraction length. The reader is referred to Ref. [7] for the discovery of this phenomenon and to Ref. [3] for a detailed review of the rich physics it involves. Here we describe methods for the modeling of nonlinear laser pulse propagation that apply in a broader context, and we will use ultrashort laser pulse filamentation as a concrete context for illustrating a specific medium model *plug-in* for general pulse-propagation models.

The notes are organized into two main sections covering (i) the theoretic background (Sec. 2) and (ii) the description of model implementation (Sec. 3). Section 2 starts from Maxwell's equations and presents the derivation of several families of propagation models suitable for nonlinear optics in regimes where Maxwell's equations are intractable. It introduces envelope models describing the propagation of laser pulses with many optical cycles as well as carrier-resolving pulse propagation models suited for few-cycle pulses. Section 3 deals with translation of theoretical models into simulation software. It shows how to implement propagation models by means of numerical algorithms that apply to a broad class of physical problems, namely those which exhibit a well-defined propagation direction. In particular, we present methods valid for envelope as well as carrier-resolving pulse propagation models, and discuss their advantages when the distance a laser pulse travels along the propagation direction is much larger than the wavelength and the dimensions of the pulse. Numerical implementation of medium-response models is treated in this section too, in particular for nonlinearities playing a role in the physics of ultrashort laser pulse filamentation.

As a concluding word for this introduction, we would like to comment on the spirit of the presentation to help readers navigate the notes. This text concentrates on propagation equations that take a canonical form, namely (i) Nonlinear Envelope Propagation Equations (listed in table 2) solved by combination of finite-difference and spectral methods, and (ii) Carrier Resolving Propagation Equations (listed in table 1) solved by purely spectral methods. [1] Our goal is to provide a self-contained overview of the state of the art in numerical simulation of femtosecond optical pulses, and, first and foremost, a practical way to embark on practical simulation. That is why Section 3 was written in the way allowing for a step-by-step building of general tools for numerical resolution of propagation equations, from the simplest to the most elaborate models. We show how to decompose a problem at hand, i.e. construction of a simulation engine into basic building blocks (e.g. linear propagation, nonlinear source terms, etc). Then we progressively include specifics describing various linear or nonlinear physical effects. Since partial differential equations of the same type as the considered propagation equations are encountered in several fields of physics (e.g., heat equation, advection-diffusion equations, amplitude equations in pattern formation problems, soliton propagation equations), the techniques taught here are naturally applicable in many other fields.

These notes are part of the 2013 edition of the Cork School on theory and mathematics modeling of ultrashort pulse propagation, and extensively relies on several texts written for earlier summer schools [8,5,4].

[1] Solution methods presented for paraxial Nonlinear Envelope Propagation Equations also apply to paraxial Carrier Resolving Propagation Equations.

Table 1. List of angular and chromatic dispersion functions for Carrier-Resolved Propagation Equations of canonical form (1). $k(\omega)$ denotes the chromatic dispersion for the medium and v_g, the pulse group velocity at its central frequency. UPPE: Unidirectional Pulse Propagation Equation. FME: Forward Maxwell Equation. FWE: Forward Wave Equation: FOP: First-Order Propagation equation. UA: Unidirectional Approximation. MA: Minimal Approximation. SEWA: Slowly Evolving Wave Approximation. P: Paraxial. ND: No Dispersion

Eq.	Ref.	Approximation	$K_z(\omega, \mathbf{k}_\perp)$	$Q(\omega, \mathbf{k}_\perp)$
UPPE	[9,4]	UA	$\sqrt{k^2(\omega) - k_\perp^2}$	$\frac{\omega^2}{c^2\sqrt{k^2(\omega) - k_\perp^2}}$
FME	[10]	SEWA, P	$k(\omega) - \frac{k_\perp^2}{2k(\omega)}$	$\frac{1}{n(\omega)}\frac{\omega}{c}$
FWE	[11,12]	MA, P	$k(\omega) + \frac{v_g}{2\omega}[(k(\omega) - \frac{\omega}{v_g})^2 - k_\perp^2]$	$\frac{v_g}{c}\frac{\omega}{c}$
FOP	[13]	SEWA, P, ND	$\frac{\omega}{c} - \frac{ck_\perp^2}{2\omega}$	$\frac{\omega}{c}$

1.1 List of Propagation Equations discussed in this text

All propagation equations considered in this text can be expressed in Fourier space in a canonical form for unidirectional equations, which read:

$$\frac{\partial \tilde{E}}{\partial z} = iK_z(\omega, \mathbf{k}_\perp)\tilde{E} + iQ(\omega, \mathbf{k}_\perp)\frac{\tilde{P}}{2\epsilon_0}, \tag{1}$$

where z denotes the propagation coordinate (or evolution variable), $\tilde{E}(\omega, \mathbf{k}_\perp, z)$ denotes the spectral components of the (scalar) electric field $E(t, \mathbf{r}_\perp, z)$ for the laser pulse, $\tilde{E}(\omega, \mathbf{k}_\perp, z)$ denotes the spectral components of the nonlinear polarization representing the response medium, and $K_z(\omega, \mathbf{k}_\perp)$ and $Q(\omega, \mathbf{k}_\perp)$ represent frequency and transverse wavenumber dependent functions, the specific form of which depends on the assumptions and model. Note that $\tilde{E}$ and $\tilde{P}$ are complex quantities even though the electric field $E(t, \mathbf{r}_\perp, z)$ for the laser pulse and the nonlinear polarization $P(t, \mathbf{r}_\perp, z)$ are real fields. Propagation equations are furthermore subdivided into carrier-resolving propagation equations (see table 1) and envelope propagation equations (see table 2). All propagation equations follow the canonical form but we will use curly notations for quantities related to envelopes, hence, the canonical equation for nonlinear envelope equations reads:

$$\frac{\partial \tilde{\mathcal{E}}}{\partial z} = i\mathcal{K}(\Omega, \mathbf{k}_\perp)\tilde{\mathcal{E}} + i\mathcal{Q}(\Omega, \mathbf{k}_\perp)\frac{\tilde{\mathcal{P}}}{2\epsilon_0}. \tag{2}$$

The numerical methods we present are valid to solve all propagation equations listed in tables 1 and 2. Independently of the distinction between carrier-resolving and envelope equations, we distinguished non-paraxial propagation equations, which can be solved more easily in the spectral domain for space-and-time by the method presented in section 3.2, from paraxial propagation equations which can also be solved by various combinations of finite-difference and spectral methods presented in section 3.1. The method presented for solving non-paraxial equations applies to paraxial equations, but the opposite is not true: Not all the methods of section 3.1 can solve non-paraxial carrier resolving propagation equations. Tables 1 and 2 list only scalar propagation equations but vectorial propagation equations can also be derived in the canonical form, thus our resolution methods extend to vectorial propagation equations.

Table 2. List of angular and chromatic dispersion functions for the Envelope Propagation Equations with canonical form (2). A moving frame co-propagating with the pulse under examination at velocity v_g is assumed and $\kappa(\omega) \equiv k_0 + (\omega - \omega_0)/v_g$, where $k_0 = k(\omega_0)$, $\omega \equiv \omega_0 + \Omega$. FEE: Forward Envelope Equation. NEE: Nonlinear Envelope Equation. LEE: Linear Envelope Equation. NLS: Nonlinear Schrödinger Equation. PC-NLS: Partially Corrected Nonlinear Schrödinger Equation. P: Paraxial. MA: Minimal Approximation. GFEA: Generalized Few-cycle Envelope Approximation. SEEA: Slowly Evolving Envelope Approximation. SEWA: Slowly Evolving Wave Approximation. SVEA: Slowly Varying Envelope Approximation.

Eq. name	Ref.	Approximation	$\mathcal{K}(\Omega, \mathbf{k}_\perp)$	$\mathcal{Q}(\Omega, \mathbf{k}_\perp)$
FEE		P	$k(\omega) - \kappa(\omega) - \frac{k_\perp^2}{2k(\omega)}$	$\frac{\omega^2}{c^2 k(\omega)}$
NEE	[3]	MA	$\frac{k^2(\omega) - \kappa^2(\omega)}{2\kappa(\omega)} - \frac{k_\perp^2}{2\kappa(\omega)}$	$\frac{\omega^2}{c^2\kappa(\omega)}$
NEE	[14]	GFEA	$k(\omega) - \kappa(\omega) - \frac{k_\perp^2}{2\kappa(\omega)}$	$\frac{\omega^2}{c^2\kappa(\omega)}$
LEE	[15]	SEEA	$k(\omega) - \kappa(\omega) - \frac{k_\perp^2}{2\kappa(\omega)}$	0
NEE	[16]	SEWA	$k(\omega) - \kappa(\omega) - \frac{ck_\perp^2}{2n_0\omega}$	$\frac{\omega}{cn_0}$
NLS	[17]	SVEA	$\frac{k_0''\Omega^2}{2} - \frac{k_\perp^2}{2k_0}$	$\frac{\omega_0}{cn_0}$
PC-NLS			$k(\omega) - \frac{ck_\perp^2}{2n_0\omega_0}(2 - \frac{\omega}{\omega_0})$	$\frac{\omega}{cn_0}$

2 Theory

When viewed through the eyes of a computational physicist, Maxwell's equations appear to consist of three coupled components. Divergence equations express initial conditions or constraints. They are *automatically* satisfied by any *good* numerical method (for example, direct Maxwell solvers will preserve $\nabla \cdot \mathbf{D} = 0$ as long as this was the case for the initial condition). Then we have the curl, or *propagation* equations, which give us the wave equation. These must be implemented, in some form, by the simulator. This part will be referred to as propagation models. The third component is embodied in material constitutive relations, which express properties of light-matter interactions. The latter part will be referred to as medium response models.

A good implementation in software should treat Maxwell system components as distinct. In other words, propagation models and medium responses should be separated as much as possible. In particular, realistic pulse evolution equations must not build on specific assumptions about the light-matter interactions. Organization of this theory Section reflects this divide and conquer approach. We will first discuss two classes of propagation models, namely envelope based and carrier-resolving in a general frame where all light-matter interactions is described by a nonlinear polarization. Then we deal with nonlinear medium properties that typically play a role in optical filamentation.

2.1 Derivation of unidirectional propagation models from the wave equation

2.1.1 From Maxwell's to the wave equation

Maxwell-Faraday and Maxwell-Ampere equations in a nonmagnetic dielectric medium read:

$$\nabla \times \mathbf{E} \quad = -\frac{\partial \mathbf{B}}{\partial t} \tag{3}$$

$$\nabla \times \mathbf{B} = \mu_0(\mathbf{J} + \frac{\partial \mathbf{D}}{\partial t}) \tag{4}$$

where $\mathbf{E}$ and $\mathbf{B}$ denote the electric and magnetic fields, $\mathbf{D}$ denotes the electric displacement field, $\mathbf{J}$ is the current density of free charges. All fields (amplitude and phase) depend on space variables $\mathbf{r} \equiv (x, y)$, time t and the propagation variable z, where we implicitly assumed existence of a well defined propagation direction. The constant μ_0 is the permeability of free space. The vector wave equation is derived from Eqs (3,4) and the relation between the electric displacement field, the electric field, and the polarization which models the response of bound electrons in the medium to the electric field. The polarization itself usually depends on the electric field via a model forming a material constitutive relation. Without entering into the details of constitutive relations at this stage, it is useful to decompose the polarization into a linear (or first order) part $\mathbf{P}^{(1)}$ describing the response of the medium for weak electric fields, and a nonlinear part $\mathbf{P}$ that is a nonlinear function of the electric field components and becomes relevant for stronger fields. The validity limit of this decomposition is expressed mathematically by the condition $\mathbf{P} \sim \mathbf{P}^{(1)}$ and corresponds physically to the range of electric fields where most electrons are still bound to the nucleus. Laser intensities up to $\sim 10^{15}$ W/cm^2 belong to this regime when ionization induced by the optical field leads to a plasma of smaller density than that of the neutral medium. In this text, we consider gases or dielectrics that are isotropic and homogeneous media, for which the components of the first-order polarization are linear functions of the components of the electric field in the frequency domain. The first order polarization follows the linear relation:

$$\hat{\mathbf{P}}^{(1)}(\mathbf{r}, \omega, z) = \epsilon_0 \chi^{(1)}(\omega) \hat{\mathbf{E}}(\mathbf{r}, \omega, z), \tag{5}$$

where ϵ_0 denotes the permittivity of free space and $\chi^{(1)}(\omega)$ is the linear susceptibility of the medium. The expression for the electric displacement reads:

$$\hat{\mathbf{D}}(\mathbf{r}, \omega, z) = \epsilon_0 \epsilon(\omega) \hat{\mathbf{E}}(\mathbf{r}, \omega, z) + \hat{\mathbf{P}}(\mathbf{r}, \omega, z), \tag{6}$$

where $\epsilon(\omega) \equiv 1 + \chi^{(1)}(\omega)$ denotes the relative permittivity of the medium [18]. By insertion of Eq. (6) in the Maxwell-Ampere equation, derivation in time of the resulting equation and by combining it with the curl of Maxwell-Gauss equation, we obtain the vectorial wave equation governing the evolution of the laser pulse in a transparent nonlinear medium. In the space-time domain with linear terms gathered on the left hand side and nonlinear material response on the right hand side, it reads:

$$\nabla^2 \mathbf{E} - \nabla(\nabla \cdot \mathbf{E}) - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \int_{-\infty}^{t} \epsilon(t - t') \mathbf{E}(\mathbf{r}, t', z) dt' = \mu_0 \left(\frac{\partial \mathbf{J}}{\partial t} + \frac{\partial^2 \mathbf{P}}{\partial t^2} \right) \tag{7}$$

where $\mathbf{E}$, $\mathbf{J}$ and $\mathbf{P}$ depend on $(\mathbf{r}, t, z)$ and we use indifferently the same notation for the frequency dependent material permittivity $\epsilon(\omega) \equiv n^2(\omega)$, which defines the complex refraction index $n(\omega)$ of the material (including effects of linear absorption), and its

time representation $\epsilon(t)$. Note that in the general case, the wave equation (7) involves a time-convolution of the permittivity with the electric field. In the following, it is useful to work with the space-frequency representation of Eq. (7):

$$\nabla^2\hat{\mathbf{E}} - \nabla(\nabla\cdot\hat{\mathbf{E}}) + \frac{\omega^2 n^2(\omega)}{c^2}\hat{\mathbf{E}} = \mu_0\left(-i\omega\hat{\mathbf{J}} - \omega^2\hat{\mathbf{P}}\right) \tag{8}$$

where $\hat{\mathbf{E}}$, $\hat{\mathbf{J}}$ and $\hat{\mathbf{P}}$ depend on $(\mathbf{r},\omega,z)$. Resolution of Eq. (7) or Eq. (8) requires constitutive equations for the medium $\mathbf{P}(\mathbf{E})$, $\mathbf{J}(\mathbf{E})$ which define a model for the medium response (free and bound electrons). Examples are given in section 2.5.

Several successive approximations can be made to derive from Eq. (7) a pulse propagation equation that is suitable for numerical implementation when the processes to simulate occur over long propagation distances along a dominant direction z. We specify these approximations and the associated simplifications of Eq. (7) in the following sections.

2.1.2 Scalar wave equation

This section details approximations to reduce the vectorial wave equation to the scalar wave equation.

First, the electric field is assumed to remain linearly polarized along a direction $\mathbf{e}_s$ transverse to the propagation axis. Thus, $\mathbf{E} = E\mathbf{e}_s$, $\mathbf{J} = J\mathbf{e}_s$, $\mathbf{P} = P\mathbf{e}_s$. There are actually two assumptions in one: First the electric field and the medium response (current $\mathbf{J}$, nonlinear polarization $\mathbf{P}$) are transverse, i.e., perpendicular to the propagation direction determined by the wave number $\mathbf{k}$. This standard assumption in propagation of electromagnetic fields means that the term $\nabla(\nabla\cdot\mathbf{E})$ in Eq. (7) can be neglected. This remains valid as long as beams are not too strongly focused. When the beam numerical aperture[2] exceeds a few percent, a small longitudinal component E_z may develop close to the focus and makes this approximation invalid. The reader interested by this case is refereed to Ref. [47], dealing with vectorial corrections to scalar wave equations. Second, there is the assumption that the electric field polarization is *linear*. The latter is not too restrictive as it essentially means that Eq. (7) can be rewritten in a scalar form by projection along the polarization direction $\mathbf{e}_s$:

$$(\partial_z^2 + \nabla_\perp^2)E(\mathbf{r},t,z) - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\int_{-\infty}^{t}\epsilon(t-t')\mathrm{E}(\mathbf{r},t',z)dt' = \mu_0\left(\frac{\partial^2 P}{\partial t^2} + \frac{\partial J}{\partial t}\right) \tag{9}$$

In the case of more than one direction for the electric field polarization, an equation of the same type as that of Eq. (9) would be obtained for each polarization direction, with coupling encoded in the material response (see section 2.3.3).

Without loss of generality, we note that the free charge current on the right hand side of Eq. (9) is formally equivalent to a time derivative of the nonlinear polarization: in the time domain, $J(\mathbf{r},t,z) \leftrightarrow \partial_t P(\mathbf{r},t,z)$. This has a counterpart in the frequency domain, $\hat{J}(\mathbf{r},\omega,z) \leftrightarrow -i\omega\hat{P}(\mathbf{r},\omega,z)$ where $\hat{P}$ and $\hat{J}$ are the Fourier transformed nonlinear polarization and current. Therefore, up to the point where we will need to specify the material nonlinear response and separate explicitly the current from the nonlinear polarization, we will consider a single term (nonlinear polarization) in propagation models. The current may then be reintroduced in any propagation equation by changing $\hat{P}$ into $\hat{P} + i\hat{J}/\omega$. Equation (9) then becomes:

$$(\partial_z^2 + \nabla_\perp^2)E(\mathbf{r},t,z) - \frac{1}{c^2}\partial_t^2\int_{-\infty}^{t} n^2(\mathbf{r},t-t',z)E(\mathbf{r},t',z)\,dt' = \mu_0\partial_t^2 P(\mathbf{r},t,z) \tag{10}$$

[2] the numerical aperture is defined as the ratio between beam diameter and focal distance

which is formally easier to handle in the Fourier domain:

$$(\partial_z^2 + \nabla_\perp^2)\hat{E}(\mathbf{r},\omega,z) + k^2(\omega)\hat{E}(\mathbf{r},\omega,z) = -\mu_0\omega^2\hat{P}(\mathbf{r},\omega,z) \tag{11}$$

where $k(\omega) \equiv n(\omega)\omega/c$.

2.1.3 Forward Maxwell Equation by factorization of the scalar wave equation

A standard way to derive a propagation equation, starting from Eq. (11), is to use a factorization method proposed by Feit and Fleck [19], which consists in separating the forward and backward propagators as:

$$(\partial_z + ik(\omega))\,(\partial_z - ik(\omega))\,\hat{E} = -\Delta_\perp\hat{E} - \mu_0\omega^2\hat{P}(\mathbf{r},\omega,z) \tag{12}$$

In the absence of the right hand side, which represents diffraction in the transverse plane and the nonlinear polarization term, Eq. (12) would admit a superposition of two solutions:

$$\hat{E}(\omega,z) = \hat{A}_+(\omega)\exp[ik(\omega)z] + \hat{A}_-(\omega)\exp[-ik(\omega)z] \tag{13}$$

which represent waves propagating in the forward or in the backward direction. Equation (12) is transformed into a unidirectional propagation equation by assuming that the backward propagating component can be neglected with respect to the forward propagating component: $|\hat{A}_-| \ll |\hat{A}_+|$, leading to the approximation: $\partial_z + ik(\omega) \simeq 2ik(\omega)$, and to the Forward Maxwell Equation (FME) [10]:

$$\frac{\partial\hat{E}}{\partial z} = ik(\omega)\hat{E} + \frac{i}{2k(\omega)}\Delta_\perp\hat{E} + \frac{i}{2n(\omega)}\frac{\omega}{c}\frac{\hat{P}}{\epsilon_0}, \tag{14}$$

where μ_0 was replaced by $1/\epsilon_0 c^2$ so as to make apparent the quantity P/ϵ_0 which has the same unit as the electric field E. As shown in section 2.1.6, the FME (14) belongs to the class of carrier resolving paraxial propagation equations, which assumes that the extent of the angular spectrum of the propagated beams remains significantly smaller than the pulse central wave number in the propagation direction. This is usually the case except for beams that are tightly focused by means of microscope objectives. The FME is therefore correctly models the propagation of beams with numerical aperture smaller than ~ 0.1 or conical beams with cone angle smaller than a few degrees [20].

We will see that in the spectral domain for both space and time, all unidirectional propagation equations take a canonical form:

$$\frac{\partial\tilde{E}}{\partial z} = iK_z(\omega,\mathbf{k}_\perp)\tilde{E} + iQ(\omega,\mathbf{k}_\perp)\frac{\tilde{P}}{2\epsilon_0} \tag{15}$$

where $K_z(\omega,\mathbf{k}_\perp)$ and $Q(\omega,\mathbf{k}_\perp)$ specifically depend on the approximations made to derive the equation. For the FME (14), we find:

$$K_z^{(\mathrm{FME})}(\omega,\mathbf{k}_\perp) \equiv k(\omega) - \frac{k_\perp^2}{2k(\omega)}, \qquad Q^{(\mathrm{FME})}(\omega,\mathbf{k}_\perp) \equiv \frac{\omega}{cn(\omega)} \tag{16}$$

which are reported in Table 1 to facilitate comparison with other models.

2.1.4 From the laboratory to the pulse local frame

In numerical simulations, it is convenient to follow a propagating pulse in its motion when (i) it travels along large distances, (ii) the main phenomena under investigation are determined by the interaction with the medium over durations of the same order of magnitude as the pulse itself, and (iii) this interaction does not lead to a strong reflected component in the backward direction so as to keep valid the unidirectional approximation $|\hat{A}_-| \ll |\hat{A}_+|$ assumed in the previous section. The latter condition does not hold e.g. in a multiple layer mirror but conditions (i)–(iii) are valid for many experimental situations where propagation effects prevail over interaction with the medium. Following the pulse in its motion is then usually performed by a change of reference frame from the laboratory to the pulse local frame $(z,t) \to (\zeta,\tau)$, where τ denotes the retarded time in the pulse frame:

$$\zeta = z, \qquad \tau = t - z/v_g \tag{17}$$

$$\partial_z = \partial_\zeta - (1/v_g)\partial_\tau, \qquad \partial_t = \partial_\tau \tag{18}$$

Note that v_g denotes a constant velocity corresponding to the change of reference frame. It is possible to chose v_g arbitrarily but a convenient choice is $v_g = 1/k_0'$, i.e. v_g coincides with the pulse group velocity obtained from the derivative $k_0' \equiv (\partial k/\partial\omega)|_{\omega_0}$ of the dispersion relation in the medium $k = k(\omega)$ at the central frequency of the pulse ω_0.

The spectral counterparts of the first equation of Eqs. (18), i.e. $\partial_z = \partial_\zeta + i(\omega/v_g)$,[3] can be directly introduced into Equation (14) to obtain:

$$\frac{\partial\hat{E}}{\partial\zeta} = i[k(\omega) - \omega/v_g]\hat{E} + \frac{i}{2k(\omega)}\Delta_\perp\hat{E} + \frac{i}{2n(\omega)}\frac{\omega}{c}\frac{\hat{P}}{\epsilon_0} \tag{19}$$

Equation (19) is still a paraxial propagation equation.

We will see through various examples that all unidirectional propagation equations originally expressed in the laboratory frame have a counterpart in the pulse frame given by a canonical form similar to Eq. (15) with the same Q and modified $K_z \to K_z - \omega/v_g$:

$$\frac{\partial\tilde{E}}{\partial\zeta} = i[K_z(\omega,\mathbf{k}_\perp) - \omega/v_g]\tilde{E} + iQ(\omega,\mathbf{k}_\perp)\frac{\tilde{P}}{2\epsilon_0} \tag{20}$$

It can be readily seen that Equation (19) indeed takes the canonical form of Eq. (20) with K_z and Q defined by Eqs. (16). This means that the different ingredients in Eq. (20) constitute the basic building blocks for the numerical implementation of a flexible numerical tool that applies to all other carrier resolving propagation equations having the canonical form, with a simple change of (i) the frequency and wave number dependence of operators K_z and Q, and (ii) constitutive relations, i.e. $P(E)$.

2.1.5 Slowly Evolving Wave Approximation

The change of reference frame made in section 2.1.4 may be applied directly to the scalar wave equation by introducing $\partial_z = \partial_\zeta + i(\omega/v_g)$ into Eq. (11). This yields:

$$\frac{\partial^2\hat{E}}{\partial\zeta^2} + 2i\frac{\omega}{v_g}\frac{\partial\hat{E}}{\partial\zeta} = -\Delta_\perp\hat{E} - [k^2(\omega) - \frac{\omega^2}{v_g^2}]\hat{E} - \frac{\omega^2}{c^2}\frac{\hat{P}}{\epsilon_0} \tag{21}$$

[3] It is found by introducing the spectral counterpart of the second equation $\partial_\tau = -i\omega$ into the first equation of Eqs. (18)

From the still exact Eq. (21), the slowly evolving wave approximation (SEWA) consists in neglecting the second order derivative in ζ with respect to the second term: $|\partial_\zeta^2 \hat{E}| \ll 2(\omega/v_g)|\partial_\zeta \hat{E}|$, or equivalently:

$$\left|\partial_\zeta \hat{E}\right| \ll \frac{\omega}{v_g}|\hat{E}|. \tag{22}$$

Physically, this approximation means that the field amplitude *and* phase are evolving sufficiently slowly along the propagation direction ζ, i.e., the typical length scale to observe a variation of $\hat{E}(\mathbf{r},\omega)$ is much larger than v_g/ω. Note that this approximation does not impose that the electric field be free of a fast oscillating carrier. The SEWA remains valid for carrier-resolving models with electric fields $E(\mathbf{r},\tau) \propto \exp(-i\omega\tau)$. Originally proposed in the context of nonlinear envelope equations [16], the SEWA does not only consist in neglecting ∂_ζ^2 with respect to $(\omega/v_g)\partial_\zeta$ in Eq. (21), called here the Minimal approximation (MA), but also in another correction presented below. We start with the MA that corresponds to Eq. (22). Under the MA, Equation (21) becomes a unidirectional propagation equation in the form of Eq. (15), called the Forward Wave Equation (FWE), which reads:

$$\frac{\partial \hat{E}}{\partial \zeta} = \frac{i}{2(\omega/v_g)}\Delta_\perp \hat{E} + i\frac{[k^2(\omega)-(\omega/v_g)^2]}{2(\omega/v_g)}\hat{E} + \frac{iv_g}{2c}\frac{\omega}{c}\frac{\hat{P}}{\epsilon_0}. \tag{23}$$

It is readily seen[4] that Eq. (23) takes the same form as Eq. (20) with

$$K_z^{(\mathrm{FWE})}(\omega,\mathbf{k}_\perp) \equiv k(\omega) + \frac{v_g}{2\omega}\left(k(\omega)-\frac{\omega}{v_g}\right)^2 - \frac{v_g k_\perp^2}{2\omega}, \qquad Q^{(\mathrm{FWE})}(\omega,\mathbf{k}_\perp) \equiv \frac{v_g\omega}{c^2}. \tag{24}$$

The FWE (23) is a carrier resolving paraxial propagation equation, that allowed for simulations of filamentation and few-cyle pulse formation in argon, coupled with a model for high harmonic generation [11,21,12,22]. In order to specify the physical content of the MA, we consider a pulse with carrier frequency ω_0: $E(z,t) \propto \exp(-i\omega_0 t + ik_0 z)$. In the pulse frame defined by Eq. (17), the field becomes $E(\zeta,\tau) \propto \exp[-i\omega_0\tau + i(k_0-\omega_0/v_g)\zeta]$. Thus, the field is oscillating at the same carrier frequency but the wave number seen in the pulse frame is $k_0-\omega_0/v_g$, justifying the assumption that the field *evolves* along ζ sufficiently slowly with respect to the central wavelength, but does not necessarily vary slowly in time. The MA as well as other approximations of the SEWA class (see Table 1) only concern the evolution variable ζ, but do not impose any restriction on the variation in time τ of the pulse. Taking the ζ derivative of the field yields $\partial_\zeta E \propto (k_0-\omega_0/v_g)E$. Inserting the latter expression in Eq. (22) leads to the condition:

$$\left|\frac{v_g k_0}{\omega_0} - 1\right| \ll 1 \quad \leftrightarrow \quad \left|\frac{v_g n_0}{c} - 1\right| \ll 1. \tag{25}$$

In other words, taking v_g as the pulse group velocity, the MA and the SEWA are justified if the relative difference between phase and group velocities is sufficiently small. More generally, the MA is valid for all frequencies of the pulse which have a phase velocity close enough to the velocity of the moving frame: $|v_g n(\omega)/c - 1| \ll 1$.

Comparison of Equation (23) with Eq. (19) shows that differences appear in the linear and nonlinear dispersion operators, i.e., the frequency-dependence in K_z and

[4] $K_z^{(\mathrm{FWE})}(\omega,\mathbf{k}_\perp) \equiv (v_g/2\omega)[k^2(\omega)-(\omega/v_g)^2] + \omega/v_g$

Q:

$$K_z^{(\mathrm{FME})} - K_z^{(\mathrm{FWE})} = -\frac{\omega}{2v_g}\left(1 - \frac{n(\omega)v_g}{c}\right)^2 - \left(1 - \frac{n(\omega)v_g}{c}\right)\frac{\mathbf{k}_\perp^2}{2k(\omega)} \tag{26}$$

$$Q^{(\mathrm{FME})} - Q^{(\mathrm{FWE})} = \frac{\omega^2}{c^2}\left(1 - \frac{n(\omega)v_g}{c}\right). \tag{27}$$

All terms on the right hand side of Eqs. (26) and (27) are first or second order terms with respect to the quantity $(1 - n(\omega)v_g/c)$. In addition to Eq. (22), the SEWA consists in also neglecting the right hand sides of Eqs. (26) and (27), i.e. in neglecting considering the FWE and the FME as identical.

2.1.6 Non paraxiality

The FME (19) and FWE (23) are paraxial propagation equations. We will see in this section how to account for nonparaxiality in unidirectional propagation equations. In Fourier space, Eq. (14) is expressed in the canonical form Eq. (15) with $K_z(\omega, \mathbf{k}_\perp) = K_z^{(\mathrm{FME})}(\omega, \mathbf{k}_\perp)$ given by Eq. (16). This expression is exactly the first order, small $k_\perp/k(\omega)$-expansion [5] of the dispersion relation obtained from the left hand side of the wave equation (11):

$$K_z(\omega, k_\perp) = \sqrt{k^2(\omega) - k_\perp^2}. \tag{28}$$

In order to account for nonparaxial effects while keeping the advantage of the unidirectional propagation, the forward and backward propagators in Eq. (11) must be rewritten in the Fourier space as :

$$\left[\partial_z + iK_z(\omega, k_\perp)\right]\left[\partial_z - iK_z(\omega, k_\perp)\right]\tilde{E} = -\mu_0\omega^2\tilde{P} \tag{29}$$

As in Eq. (12), we now consider only the forward propagating component, i.e., we make the approximation $\partial_z + iK_z \sim 2iK_z$. This leads to the nonparaxial version of the FME, i.e. the Unidirectional Pulse Propagation Equation (UPPE):

$$\frac{\partial\tilde{E}}{\partial z} = iK_z(\omega, k_\perp)\tilde{E} + \frac{i}{2K_z(\omega, k_\perp)}\frac{\omega^2}{c^2}\frac{\tilde{P}}{\epsilon_0} \tag{30}$$

which follows the canonical form with $K_z^{(\mathrm{UPPE})}$ given by Eq. (28) and $Q^{(\mathrm{UPPE})} \equiv \omega^2/K_z^{(\mathrm{UPPE})}c^2$. The UPPE (30) was obtained here by the factorization method. Sections 2.3 and 2.3.3 will present a rigorous derivation from which the physical meaning of the approximation made in the factorization will appear.

Comparing the UPPE (30) with the FME (14), we note two important differences: (i) The FME involves differential operators in transverse spatial variables that describe diffraction in the transverse plane and space-time focusing due to the frequency dependence in the diffraction coefficient. Its nonparaxial version Eq. (30) is naturally expressed in the spectral domain due to the factorization method. Therefore, it cannot be translated easily in terms of differential operators in spatial variables, apart from a formal writing which becomes rather intractable when numerical implementation with finite difference is concerned. The effects of diffraction and space-time focusing are still described via the wave number ($k_\perp$) and frequency (ω) dependence in

[5] $|k_\perp| \ll |k(\omega)|$ means that the validity limit of the expansion is the paraxial propagation regime

$K_z(\omega, k_\perp)$ [Eq. (28)], respectively, but nonparaxiality is now accounted for. (ii) Since $K_z^{(\mathrm{UPPE})}(\omega, k_\perp)$ depends on $k_\perp$ and $Q^{(\mathrm{UPPE})}$ is obtained as a function of $K_z^{(\mathrm{UPPE})}$, wave numbers appear as new Fourier variables in the operator Q acting on nonlinear polarization in the UPPE (30), whereas Eqs (16) show that the quantity $Q^{(\mathrm{FME})}$ is independant of $k_\perp$. This means that Eq. (30) is potentially much easier to implement numerically than equations in the form of the FME. Evaluation of the r.h.s. of Eq. (30) simply requires multiplications in the spectral domain $K_z(\omega, k_\perp) \times \tilde{E}(\omega, k_\perp)$ to describe linear propagation effects instead of dealing with the transverse Laplacian in the spatial domain as in Eq. (19) and paraxial equations in general. However, the nonlinear polarization $\tilde{P}(\omega, k_\perp)$ is often most easily evaluated in the space-time domain $P(\tau, r)$. Thus, the gain due to the simplicity in linear propagation in Eq. (30) is partly compensated by the need to perform three-dimensional Fourier transforms back and forth each time the nonlinear polarization $\tilde{P}(\omega, k_\perp)$ will be evaluated from $P(\tau, r)$.

The axial wave number for the FME departs from that of the UPPE for large transverse wave numbers, which corresponds to the paraxiality assumption made in the FME. The axial wave number for the FWE exhibits a small departure from that of the FME even for small transverse wave numbers, which corresponds to a systematic distortion in the dispersion relation introduced when only the MA is assumed. The SEWA corrects this distortion. However, this deviation is really small for common focusing geometries and pulse durations. More limiting form the practical point of view is the uncertainty in medium parameters, and availability of dispersion relations accurate over wide frequency ranges [23].

2.2 Derivation of envelope propagation equations

Envelope models are useful when there exist separate scales for the evolution of the pulse, a fast scale typically of the order of the wavelength, and a slow scale much larger than the wavelength. The pulse propagation can then be advantageously described by considering the electric field as a superposition of the pulse envelope $\mathcal{E}$ with a carrier wave of frequency ω_0: $E(\mathbf{r}, t, z) = \mathcal{E}(\mathbf{r}, t, z) \exp(ik_0 z - i\omega_0 t)$. In the local pulse frame: $E(\mathbf{r}, \tau, \zeta) = \mathcal{E}(\mathbf{r}, \tau, \zeta) \exp[i(k_0 - \omega_0/v_g)\zeta - i\omega_0\tau]$. A similar decomposition holds for the nonlinear polarization and the free charge current: $\{P, J\}(\mathbf{r}, \tau, \zeta) = \{\mathcal{P}, \mathcal{J}\}(\mathbf{r}, \zeta, \tau) \exp[i(k_0 - \omega_0/v_g)\zeta - i\omega_0\tau]$. Propagation equations for the envelope can be obtained by introducing this decomposition in any carrier resolving propagation equation or in the wave equation itself. We will examine both techniques. Formally, one simply needs to apply transformations to the evolution operator and time derivatives which read:

In the laboratory frame:

$$\partial_z E = \exp(ik_0 z - i\omega_0 t)[\partial_z + ik_0]\mathcal{E} \tag{31}$$

$$\partial_t E = \exp(ik_0 z - i\omega_0 t)[\partial_t - i\omega_0]\mathcal{E} \tag{32}$$

In the pulse frame:

$$\partial_\zeta E = \exp(i(k_0 - \omega_0/v_g)\zeta - i\omega_0\tau)[\partial_\zeta + i(k_0 - \omega_0/v_g)]\mathcal{E}, \tag{33}$$

$$\partial_\tau E = \exp(i(k_0 - \omega_0/v_g)\zeta - i\omega_0\tau)[\partial_\tau - i\omega_0]\mathcal{E} \tag{34}$$

We will see in several examples that these transformations lead to envelope propagation equations that also take the canonical form:

$$\frac{\partial \tilde{\mathcal{E}}}{\partial \zeta} = i\mathcal{K}(\Omega, \mathbf{k}_\perp)\tilde{\mathcal{E}} + i\mathcal{Q}(\Omega, \mathbf{k}_\perp)\frac{\tilde{\mathcal{P}}}{2\epsilon_0} \tag{35}$$

where $\Omega \equiv \omega - \omega_0$.

2.2.1 Nonlinear Envelope Equations from Carrier-Resolving Propagation equations

In this section, we derive nonlinear envelope equations obtained when the carrier and envelope decomposition is introduced in a carrier resolving propagation equation. Introducing Equations (33) and $\partial_z = \partial_\zeta + \omega/v_g$ into the canonical carrier resolving propagation equation (15) and removing the carrier wave $\exp[i(k_0 - \omega_0/v_g)\zeta - i\omega_0\tau]$ transforms it into Eq. (35) with

$$\mathcal{K}(\Omega, \mathbf{k}_\perp) \equiv K_z(\omega = \omega_0 + \Omega, \mathbf{k}_\perp) - \kappa(\omega = \omega_0 + \Omega) \tag{36}$$

$$\mathcal{Q}(\Omega, \mathbf{k}_\perp) \equiv Q(\omega = \omega_0 + \Omega, \mathbf{k}_\perp) \tag{37}$$

where $\kappa(\omega) \equiv k_0 + (\omega - \omega_0)/v_g$.

Using the Forward Maxwell Equation (19) as a starting point leads to the nonlinear envelope equation written in the spectral domain:

$$\frac{\partial \hat{\mathcal{E}}}{\partial \zeta} = \frac{i}{2k(\omega)} \Delta_\perp \hat{\mathcal{E}} + i[k(\omega) - \kappa(\omega)]\hat{\mathcal{E}} + \frac{i}{2k(\omega)} \frac{\omega^2}{c^2} \frac{\hat{\mathcal{P}}}{\epsilon_0} \tag{38}$$

One notes that Eq. (38), as the original FME (19), is a paraxial equation. The main difference is that the fields in Eq. (19) have to be treated as real quantities with high temporal resolution of the order of the optical period, whereas the envelopes in Eq. (38) are complex quantities that require lower temporal resolution of the order of the pulse duration.

2.2.2 Nonlinear Envelope Equation from the wave equation

In this section, we derive Nonlinear Envelope Equations (NEE) directly from the wave equation. Since we aim at finding a nonlinear envelope equation in the pulse frame, we start from the wave equation (21) in the pulse frame, we introduce the carrier-envelope decomposition (33), remove the carrier and recombine the ω/v_g terms with $k_0 - \omega_0/v_g$ to form $\kappa(\omega) = k_0 + (\omega - \omega_0)/v_g$. This yields:

$$\frac{\partial^2 \hat{\mathcal{E}}}{\partial \zeta^2} + 2i\kappa(\omega) \frac{\partial \hat{\mathcal{E}}}{\partial \zeta} = -\Delta_\perp \hat{\mathcal{E}} - [k^2(\omega) - \kappa^2(\omega)]\hat{\mathcal{E}} - \frac{\omega^2}{c^2} \frac{\hat{\mathcal{P}}}{\epsilon_0}. \tag{39}$$

Equation (39) is exact and did not require any approximation except when vectorial effects in the original wave equation were neglected. We discuss below different Nonlinear Envelope Equations obtained by performing various approximations.

Minimal Approximation (MA) From Equation (39), the only required approximation to obtain a propagation equation in the canonical form for nonlinear envelope equations is that of neglecting ∂_ζ^2. This yields a NEE that has been extensively used in simulations of ultrashort laser pulse filamentation [3]:

$$\frac{\partial \hat{\mathcal{E}}}{\partial \zeta} = \frac{i}{2\kappa(\omega)} \Delta_\perp \hat{\mathcal{E}} + i \frac{[k^2(\omega) - \kappa^2(\omega)]}{2\kappa(\omega)} \hat{\mathcal{E}} + \frac{i}{2\kappa(\omega)} \frac{\omega^2}{c^2} \frac{\hat{\mathcal{P}}}{\epsilon_0} \tag{40}$$

Equation (40) contains all terms found in various types of NEEs found in the literature, derived under various approximations. To discuss these approximations, we rewrite the second term on the right hand side of Eq. (39) as:

$$k^2(\omega) - \kappa^2(\omega) = 2\kappa(\omega)[k(\omega) - \kappa(\omega)] + [k(\omega) - \kappa(\omega)]^2. \tag{41}$$

The rationale behind this rewriting is understood from a small $\Omega \equiv \omega - \omega_0$ Taylor expansion of the difference $k(\omega) - \kappa(\omega)$ around ω_0, which introduces the high-order dispersive terms

$$k(\omega_0 + \Omega) - \kappa(\omega_0 + \Omega) = \sum_{l=2}^{+\infty} \frac{k_0^{(l)}}{l!} \Omega^l, \tag{42}$$

where $k_0^{(l)} \equiv \partial_\omega^l k|_{\omega_0}$. Note that the lowest order term in the second term on the rhs of Eq. (41) is $O(\Omega^4)$, and thus appears as a small correction with respect to the first term on the rhs of Eq. (41) that is $O(\Omega^2)$. Equation (40) is thus rewritten as

$$\frac{\partial \hat{\mathcal{E}}}{\partial \zeta} = \frac{i}{2\kappa(\omega)} \Delta_\perp \hat{\mathcal{E}} + i(k(\omega) - \kappa(\omega))\hat{\mathcal{E}} + i\frac{[k(\omega) - \kappa(\omega)]^2}{2\kappa(\omega)} \hat{\mathcal{E}} + \frac{i}{2\kappa(\omega)} \frac{\omega^2}{c^2} \frac{\hat{\mathcal{P}}}{\epsilon_0} \tag{43}$$

which is convenient to review the various type of NEEs and compare them to the NEE-MA, since all of them neglect the $O(\Omega^4)$ third term on the rhs of Eq (43). We will follow the terminology introduced by Kinsler et al. [14] that classifies various approximations in the literature on nonlinear envelope equations.

Generalized Few-cycle Envelope Approximation (GFEA) This approximation proposed by Kinsler et al [14] consists in first rewriting the envelope equation (39) by using Eq. (41) as for the NEE-MA:

$$2i\kappa(\omega)\frac{\partial \hat{\mathcal{E}}}{\partial \zeta} = -\Delta_\perp \hat{\mathcal{E}} - 2\kappa(\omega)[k(\omega) - \kappa(\omega)]\hat{\mathcal{E}} + \{[k(\omega) - \kappa(\omega)]^2 - \partial_\zeta^2\}\hat{\mathcal{E}} - \frac{\omega^2}{c^2}\frac{\hat{\mathcal{P}}}{\epsilon_0}. \tag{44}$$

Equation (44) is still exact. Then the third term on the r.h.s. of Equation (44) is neglected. This yields:

$$\frac{\partial \hat{\mathcal{E}}}{\partial \zeta} = i[k(\omega) - \kappa(\omega)]\hat{\mathcal{E}} + \frac{i}{2\kappa(\omega)}\Delta_\perp \hat{\mathcal{E}} + \frac{i}{2\kappa(\omega)}\frac{\omega^2}{c^2}\frac{\hat{\mathcal{P}}}{\epsilon_0}, \tag{45}$$

which has a counterpart in the space-time domain:

$$\frac{\partial \mathcal{E}}{\partial \zeta} = i\mathcal{D}(i\partial_\tau)\mathcal{E} + \frac{i}{2k_0}\left(1 + i\frac{k_0'}{k_0}\partial_\tau\right)^{-1}\Delta_\perp \mathcal{E} + \frac{i}{2}\frac{\omega_0}{n_0 c}\left(1 + i\frac{k_0'}{k_0}\partial_\tau\right)^{-1}\left(1 + \frac{i}{\omega_0}\partial_\tau\right)^2 \frac{\mathcal{P}}{\epsilon_0}, \tag{46}$$

where $\mathcal{D}(i\partial_\tau) \equiv \sum_{l=2}^{+\infty} \frac{k_0^{(l)}}{l!}(i\partial_\tau)^l$.

Thus there are two approximations in the GFEA: (i) An implicit small Ω-Taylor expansion of the dispersion operator is assumed and $O(\Omega^4)$ terms are neglected. The neglected terms precisely make the remaining dispersive terms of the NEE-GFEA (45) identical to those of the Forward Envelope Equation (38). (ii) The envelope is evolving slowly with respect to the propagation variable ζ: $|\partial_\zeta \mathcal{E}| \ll |k_0 \mathcal{E}|$. In the original terminology introduced for the linear version of the NEE-GFEA (45), i.e. in the absence of a nonlinear polarization, the latter approximation was called the Slowly Evolving Envelope Approximation (SEEA) [15]. The operator $(1 + i(k_0'/k_0)\partial_\tau)^{-1}$ in front of the diffraction term describe space-time focusing, i.e, a frequency dependence of diffraction carried by the effective propagation constant $\kappa(\omega) = k_0 + k_0'(\omega - \omega_0)$. In medium with normal dispersion, red frequencies propagate faster than blue frequencies and a pulse with flat phase front would normally broaden. For a beam, due to space-time focusing, red frequencies are diffracted at larger angles than blue frequencies making diffraction equivalent to an anomalous dispersive phenomenon that can compensate normal dispersion [15].

Slowly Evolving Wave Approximation (SEWA) The SEWA was explained in the context of carrier-resolving equations. In the context of Nonlinear Envelope Equations, the SEWA was introduced by Brabec and Krausz [16] and consists in the same approximation as the GFEA with an additional approximation on the propagation constant $\kappa(\omega) \sim n_0\omega/c$ appearing in the second and third terms[6] on the r.h.s. of Eq. (45). This leads to the spectral version of the Nonlinear Envelope Equation under the SEWA (NEE-SEWA):

$$\frac{\partial\hat{\mathcal{E}}}{\partial\zeta} = \frac{ic}{2n_0\omega}\Delta_\perp\hat{\mathcal{E}} + i[k(\omega)-\kappa(\omega)]\hat{\mathcal{E}} + \frac{i}{2n_0}\frac{\omega}{c}\frac{\hat{\mathcal{P}}}{\epsilon_0}, \tag{47}$$

which was initially derived in the space-time domain [16]. From the NEE-GFEA, the approximation made to derive the NEE-SEWA are equivalently expressed in the temporal domain as $(1+i(k_0'/k_0)\partial_\tau) \sim \left(1+i/{\omega_0}^{-1}\partial_\tau\right)$, thus the space-time counterpart of the NEE-SEWA reads:

$$\frac{\partial\mathcal{E}}{\partial\zeta} = i\mathcal{D}(i\partial_\tau)\mathcal{E} + \frac{i}{2k_0}\left(1+\frac{i}{\omega_0}\partial_\tau\right)^{-1}\Delta_\perp\mathcal{E} + \frac{i}{2}\frac{\omega_0}{n_0c}\left(1+\frac{i}{\omega_0}\partial_\tau\right)\frac{\mathcal{P}}{\epsilon_0}. \tag{48}$$

The SEWA introduces a slight distortion in the space-time focusing operator $(1+(i/\omega_0)\partial_\tau)^{-1}$, obtained from its original version $(1+i(k_0'/k_0)\partial_\tau)$ by the change $k_0'/k_0 \to {\omega_0}^{-1}$ which amounts to neglecting the difference between phase and group velocities $c/n_0 = \omega_0/k_0 \sim 1/k_0'$, as seen from the frequency dependence of effective propagation constant for the NEE-GFEA and NEE-SEWA $k_0+k_0'(\omega-\omega_0) \sim k_0\omega/\omega_0$.

Slowly Varying Envelope Approximation (SVEA) At the lowest order, we may retain only the second order dispersive term in Equation (42), or the second and third orders:

$$k(\omega_0+\Omega)-\kappa(\omega_0+\Omega) = \frac{k_0^{(2)}}{2}\Omega^2 + O(\Omega^3), \tag{49}$$

and by keeping the lowest order in $\kappa(\omega_0+\Omega) \sim k_0$ and $\omega \sim \omega_0$, we obtain from Eq. (40) an equation of the Nonlinear Schrödinger (NLS) type that is valid for pulses with a narrow spectrum:

$$\frac{\partial\hat{\mathcal{E}}}{\partial\zeta} = \frac{i}{2k_0}\Delta_\perp\hat{\mathcal{E}} + i\frac{k_0^{(2)}}{2}\Omega^2\hat{\mathcal{E}} + \frac{i}{2n_0}\frac{\omega_0}{c}\frac{\hat{\mathcal{P}}}{\epsilon_0}. \tag{50}$$

Strictly speaking, the Nonlinear Schrödinger Equation involves a cubic dependence of the nonlinear polarization $\mathcal{P} \propto |\mathcal{E}|^2\mathcal{E}$ as obtained with a Kerr nonlinearity (see section on the medium response) and is usually derived by introducing the carrier and envelope decomposition directly into the wave equation written in the space-time domain.

$$\frac{\partial\mathcal{E}}{\partial\zeta} = \frac{i}{2k_0}\Delta_\perp\mathcal{E} - i\frac{k_0^{(2)}}{2}\frac{\partial^2\mathcal{E}}{\partial\tau^2} + \frac{i}{2n_0}\frac{\omega_0}{c}\frac{\mathcal{P}}{\epsilon_0}. \tag{51}$$

As written in Eq. (50), the NLS takes the canonical form of envelope equations. All nonlinear envelope equations derived in the previous sections can be considered as extended NLS equations in the sense that they can be obtained from the NLS equation by introducing the proper frequency dependence in the propagation constant, dispersion operator, and nonlinear dispersion.

[6] It does not effectively appear in the first term. This can be viewed from the Taylor expansion (42).

2.3 Derivation of carrier-resolving pulse propagation models

The following section is devoted to rigorous derivations of Unidirectional Pulse Propagation Equations (UPPE). The idea here is to start from Maxwell's equations and avoid any approximation in the *propagation part of the problem*. As the reader will see, approximations become only necessary at the point when nonlinear interactions are considered, and then they only have to do with splitting an exact system of pulse propagation equations into two decoupled *unidirectional* equations.

The UPPE approach has been previously derived in two flavors, depending on direction of the numerically simulated evolution [9,4]. The latter can proceed either along the time coordinate, or it can follow a wave packet along one of the spatial coordinates (usually chosen as z) in the direction of the laser beam.

In the first case, termed *time-propagated evolution*, one has an initial condition (i.e. a given description of the electric and magnetic fields) specified in all space for a given initial time. The evolution is calculated along the time axis, and it naturally reflects the structure of Maxwell's equations.

In the second case, termed *z-propagated evolution*, the initial condition is given as a function of the local pulse time and of two transverse (with respect to the propagation direction) coordinates. Numerical evolution then proceeds along the propagation axis. From the mathematical point of view, this case is an initial value problem very much the same way the t-propagated case is. However, from the physical point of view this is a rather subtle issue because the true *initial* condition requires knowledge of the *total* field in the past and in the future. This includes the light which may be nonlinearly reflected from the focal region of an experiment. Only if we can assume that this is sufficiently weak, we can practically solve the corresponding initial value problem.

The time-propagated approach is common for solvers based on direct integration of Maxwell's equations. Accordingly, the time-propagated versions of UPPE are more suitable for tight-focusing scenarios when the non-paraxial effects start to play a role.

On the other hand, the z-propagated equations are easier to use in situations which allow to neglect longitudinal field components as contributing sources of nonlinear material responses. That is the main reason the z-propagated approach is more common, especially in nonlinear optics. In this text, we will restrict our attention to the z-propagated case. However, numerical techniques described here are directly applicable to any t-propagated simulator implementation.

In relation to the previous section, the following material will reveal one interesting fact. We have seen that some of the equations derived from the wave equations seem to require certain approximations. Now it will be shown that despite of this, the resulting equations are in a certain sense *exact*, and that assumptions needed for the Laplacian factorization are in fact not necessary but only sufficient conditions. This apparent contradiction originates in the fact that a solution to a unidirectional propagation equation must not satisfy the wave equation. Only two coupled unidirectional solutions (propagating in opposite directions) must, and indeed obey the wave equation.

2.3.1 A simplified case first: One-dimensional Maxwell's equations

The most important ingredient in unidirectional evolution equations is the separation of those wave-form portions which propagate in opposite direction. To emphasize this, and also to make the subsequent full derivation easier to digest, we start with a simplified case of one-dimensional Maxwell's equations. This reduced case is free of notational complications while it illustrates all important steps of the fully vectorial

treatment. In particular, it shows very clearly that a pair of unidirectional equations is exact.

The one dimensional Maxwell equations reduced to linearly polarized electric field can be written as (for convenience and notational simplicity, appropriate scaled units are used in this subsection):

$$\begin{aligned} -\partial_z H &= \partial_t E + \partial_t P \\ -\partial_z E &= \partial_t H \end{aligned} \tag{52}$$

where z is the optical axis and E and H are implicitly understood to be orthogonal to each other and to z. P represents nonlinear polarization that itself is a functional of the electric field history at a given spatial point. Section 2.5 provides several examples the reader can keep in mind, but a concrete form of this nonlinearity is completely unimportant for the purpose of the following derivation.

This simplified Maxwell system has harmonic waves as solutions in the linear regime when $P = 0$:

$$\begin{aligned} \mathcal{E}_\lambda(\omega, z, t) &= \mathcal{E}_0 \exp\left[-i\omega t + i\lambda k(\omega) z\right] \\ \mathcal{H}_\lambda(\omega, z, t) &= \lambda \mathcal{H}_0 \exp\left[-i\omega t + i\lambda k(\omega) z\right] \quad \omega > 0 \quad \lambda = \pm 1 \end{aligned} \tag{53}$$

The direction indicator λ selects forward and backward (or left and right) propagating waves. We can use these plane-waves as a basis in which to express a full, nonlinear solution as

$$E = \sum_{\mu=\pm 1} \int d\Omega A_\mu(\Omega, z) \mathcal{E}_\mu(\Omega, z, t) \qquad H = \sum_{\mu=\pm 1} \int d\Omega A_\mu(\Omega, z) \mathcal{H}_\mu(\Omega, z, t)$$

Here, $A_\mu(\Omega, z)$ are spectral amplitudes for which we have to find an evolution equation. Taking (52), and multiplying with the above linear basis solutions we get

$$\begin{aligned} \mathcal{E}_\lambda \partial_z H &= -\mathcal{E}_\lambda \partial_t E - \mathcal{E}_\lambda \partial_t P \\ \mathcal{H}_\lambda \partial_z E &= -\mathcal{H}_\lambda \partial_t H \end{aligned} \tag{54}$$

In these equations and in following formulas, we assume that the arguments of $\mathcal{E}_\lambda$ and $\mathcal{H}_\lambda$ are Ω, z, t. We now add these two equations, and collect terms that constitute full derivatives while using the fact that $\mathcal{H}_\lambda$, $\mathcal{E}_\lambda$ solve the linear Maxwell system:

$$\partial_z[\mathcal{E}_\lambda H + \mathcal{H}_\lambda E] = -\partial_t[\mathcal{E}_\lambda H + \mathcal{H}_\lambda E] - \mathcal{E}_\lambda \partial_t P \tag{55}$$

The next step is to integrate over the whole domain perpendicular to the direction of propagation. In this simplified case it means the t domain alone. After integration over t, the middle term gives rise to boundary terms at past and future temporal infinities. To get rid of these, we will restrict our solution space to those functions which satisfy

$$\lim_{t \to \pm\infty} [\mathcal{E}_\lambda H(z, t) + \mathcal{H}_\lambda E(z, t)] = 0 \tag{56}$$

What this condition requires is that for every fixed z along the laser beam axis, the field will vanish if we wait for a sufficiently long time. In other words, light energy will dissipate into positive and negative z-infinities. This is certainly a very benign assumption in the context of pulse propagation, because this is exactly what happens to localized pulsed wave packets - they eventually disappear from our sight.

The above condition eliminates the middle term in (55), and the rest can be transformed as follows. First, in the left-hand side we use the fact that the basis

solutions are orthogonal, and after time integration they eliminate the sum over Ω and the modal index μ:

$$\begin{aligned}
&\int dt\partial_z[\mathcal{E}_\lambda(\omega,z,t)H+\mathcal{H}_\lambda(\omega,z,t)E]=\\
&=\int dt\partial_z\mathcal{E}_\lambda(\omega,z,t)\sum_{\mu=\pm1}\int d\Omega A_\mu(\Omega,z)\mathcal{H}_\mu(\Omega,z,t)+\\
&+\int dt\partial_z\mathcal{H}_\lambda(\omega,z,t)\sum_{\mu=\pm1}\int d\Omega A_\mu(\Omega,z)\mathcal{E}_\mu(\Omega,z,t)=\\
&=2\lambda\mathcal{E}_0\mathcal{H}_0\partial_z A_\lambda(\omega,z)
\end{aligned}\tag{57}$$

On the right-hand side of Eq. (55), the polarization term yields essentially a Fourier transform

$$-\int dt\mathcal{E}_\lambda(\omega,z,t)\partial_t P=i\omega\exp\left[-i\lambda k(\omega)z\right]\hat{P}(\omega,z)$$

Collecting both sides, we arrive at an evolution equation for spectral amplitudes:

$$\partial_z A_\lambda(\omega,z)=\frac{i\omega}{2\lambda\mathcal{E}_0\mathcal{H}_0}\exp\left[-i\lambda k(\omega)z\right]\hat{P}(\omega,z)\tag{58}$$

To obtain a corresponding equation for the electric field, we recall that

$$\hat{E}_\lambda(\omega,z)=A_\lambda(\omega,z)\exp\left[i\lambda k(\omega)z\right]$$

and expressing its z-derivatives using the evolution equation (58) for the spectral amplitudes, we have:

$$\partial_z\hat{E}_\lambda(\omega,z)=i\lambda k(\omega)\hat{E}_\lambda(\omega,z)+i\lambda\frac{\omega}{2\mathcal{E}_0\mathcal{H}_0}\hat{P}(\omega,z)$$

This is a pair of equations for forward and backward ($\lambda=\pm1$) propagating fields. The two are coupled through the polarization which depends on their sum. Explicitly,

$$P(z,t)=P[E_+(z,t)+E_-(z,t)]\ .$$

where the concrete functional form of this dependence is not important for the present purpose, but as an example one can consider the instantaneous Kerr nonlinearity for which the polarization is simply proportional to the cube of the electric field (for more examples, see section 2.5):

$$P(z,t)=P[E_+(z,t)+E_-(z,t)]\propto[E_+(z,t)+E_-(z,t)]^3$$

The above derivation illustrates the scheme we will use in the next Section to derive the general, fully vectorial Unidirectional Pulse Propagation Equation. The important point to note here is that within the admissible subspace specified by condition (56), the pair of unidirectional equations is *exact*.

2.3.2 Maxwell's equations as a boundary value problem for pulsed beam propagation

As a first step in derivation of various versions of UPPE, we derive an exact coupled-modes system of equations. Electromagnetic fields of a light pulse propagating along

the z-axis can be expanded into modal contributions that reflect the geometry of the waveguide (we can consider a homogeneous medium as a special case of the latter).

$$\begin{aligned}\mathbf{E}(x,y,z,t) &= \sum_{m,\omega} A_m(\omega,z)\mathcal{E}_m(\omega,x,y)e^{i\beta_{\mathrm{m}}(\omega)z-i\omega t}\\ \mathbf{H}(x,y,z,t) &= \sum_{m,\omega} A_m(\omega,z)\mathcal{H}_m(\omega,x,y)e^{i\beta_{\mathrm{m}}(\omega)z-i\omega t}\end{aligned} \tag{59}$$

Here, m labels all transverse modes, and an initial condition $A_m(\omega, z = 0)$ is supposed to be given or calculated from the known field values at $z = 0$. Note that the above expansion is valid for the transverse components only, and that the modal index m is a short hand for all quantities which are required to specify a unique propagation mode. For example, in a homogeneous bulk medium, the eigen modes are the well known plane waves, and the index m represents polarization, two transverse wave numbers, and a binary value selecting the forward or backward direction of propagation.

To save space and reduce clutter, the following short-hand notation will be used below

$$\begin{aligned}\mathcal{E}_m &\equiv \mathcal{E}_m(\omega,x,y)e^{i\beta_{\mathrm{m}}(\omega)z-i\omega t}\\ \mathcal{H}_m &\equiv \mathcal{H}_m(\omega,x,y)e^{i\beta_{\mathrm{m}}(\omega)z-i\omega t}\ .\end{aligned} \tag{60}$$

We consider a non-magnetic medium ($\mu = \mu_0$) with a linear permittivity $\epsilon(\omega, x, y)$ that does not depend on the propagation coordinate z which coincides with what we consider forward and backward propagation direction. Note that the permittivity or, equivalently, the index of refraction may depend on the transverse coordinates x, y. That would be the case for example in a micro-structured waveguide, or in a hollow-core fiber or capillary; at this first stage, we want to treat bulk media and fiber-like geometries together. Later we can *branch* and derive separate, specialized equations for waveguides and for bulk media.

The starting point of our derivations is the Maxwell equations:

$$\begin{aligned}\mathbf{J} + \partial_t\mathbf{P} + \epsilon_0\partial_t\epsilon * \mathbf{E} &= \nabla\times\mathbf{H}\\ -\mu_0\partial_t\mathbf{H} &= \nabla\times\mathbf{E}\end{aligned} \tag{61}$$

where the star represents a convolution so that the term is a short hand for

$$\epsilon_0\partial_t\epsilon * \mathbf{E} = \epsilon_0\partial_t\int_0^\infty \mathrm{d}\tau\epsilon(\tau)\mathbf{E}(t-\tau)$$

Here $\epsilon(\tau)$ is the temporal representation of frequency-dependent permittivity $\epsilon(\omega)$. The same notation will be used for both quantities, and their arguments will serve to distinguish them where needed.
As a first step, we scalar-multiply Maxwell's equations by complex conjugate modal fields

$$\begin{aligned}\mathcal{E}_m^*.(\mathbf{J} + \partial_t\mathbf{P}) + \epsilon_0\mathcal{E}_m^*.\partial_t\epsilon * \mathbf{E} &= \mathcal{E}_m^*.\nabla\times\mathbf{H}\\ -\mu_0\mathcal{H}_m^*.\partial_t\mathbf{H} &= \mathcal{H}_m^*.\nabla\times\mathbf{E}\ .\end{aligned} \tag{62}$$

Using the formula $\mathbf{b}.(\nabla\times\mathbf{a}) = \nabla.(\mathbf{a}\times\mathbf{b}) + \mathbf{a}.(\nabla\times\mathbf{b})$, we transform both right-hand sides to obtain

$$\begin{aligned}\mathcal{E}_m^*.(\mathbf{J} + \partial_t\mathbf{P}) + \epsilon_0\mathcal{E}_m^*.\partial_t\epsilon * \mathbf{E} &= \nabla.[\mathbf{H}\times\mathcal{E}_m^*] + \mathbf{H}.[\nabla\times\mathcal{E}_m^*]\\ -\mu_0\mathcal{H}_m^*.\partial_t\mathbf{H} &= \nabla.[\mathbf{E}\times\mathcal{H}_m^*] + \mathbf{E}.[\nabla\times\mathcal{H}_m^*]\ .\end{aligned} \tag{63}$$

Now we can take advantage of the fact the modal fields themselves satisfy Maxwell's equations

$$\begin{aligned} \nabla \times \mathcal{E}_m^* &= -\mu_0 \partial_t \mathcal{H}_m^* \\ \nabla \times \mathcal{H}_m^* &= \epsilon_0 \partial_t \epsilon * \mathcal{E}_m^* \ , \end{aligned} \tag{64}$$

and therefore the previous equations can be written as

$$\begin{aligned} \mathcal{E}_m^*.(\mathbf{J} + \partial_t \mathbf{P}) + \epsilon_0 \mathcal{E}_m^*.\partial_t \epsilon * \mathbf{E} &= \nabla.[\mathbf{H} \times \mathcal{E}_m^*] - \mu_0 \mathbf{H}.\partial_t \mathcal{H}_m^* \\ -\mu_0 \mathcal{H}_m^*.\partial_t \mathbf{H} &= \nabla.[\mathbf{E} \times \mathcal{H}_m^*] + \epsilon_0 \mathbf{E}.\partial_t \epsilon * \mathcal{E}_m^* \end{aligned} \tag{65}$$

Next, we subtract the two equations and collect terms that constitute full time derivatives

$$\mathcal{E}_m^*.(\mathbf{J} + \partial_t \mathbf{P}) + \partial_t[\epsilon_0 \mathcal{E}_m^*.\epsilon * \mathbf{E}] = \nabla.[\mathbf{H} \times \mathcal{E}_m^*] - \partial_t[\mu_0 \mathcal{H}_m^*.\mathbf{H}] - \nabla.[\mathbf{E} \times \mathcal{H}_m^*] \ . \tag{66}$$

Now we integrate over the whole xyt domain. Note that all terms except the first and ∂_z, which is implicit in the $\nabla.$ operator, are derivatives that give rise to *surface terms* after integration over x, y, t. These surface terms are supposed to vanish far from the axis of the laser beam, as well as in past and future temporal infinities. Intuitively, admissible solutions include spatially and temporally localized pulse-like solutions. As a consequence, the only surviving derivatives will be ∂_z:

$$\int \mathcal{E}_m^*.(\mathbf{J} + \partial_t \mathbf{P}) \, \mathrm{d}x\mathrm{d}y\mathrm{d}t = \partial_z \int \mathbf{z}.[\mathbf{H} \times \mathcal{E}_m^*] \, \mathrm{d}x\mathrm{d}y\mathrm{d}t - \partial_z \int \mathbf{z}.[\mathbf{E} \times \mathcal{H}_m^*] \, \mathrm{d}x\mathrm{d}y\mathrm{d}t \tag{67}$$

Here and in what follows, t integrations are understood as: $\int \mathrm{d}t \equiv \frac{1}{T} \int_{-T/2}^{+T/2} \mathrm{d}t$ where T is a large normalization volume, and integrals over x, y are understood in a similar way. This will give us a convenient way to obtain the correct normalization and *translate* it into implementation which will be in terms of numerical Fourier transforms.
Because only transverse field components enter the above equation, we can use our modal expansion here (recall that those are only valid for transverse vector components):

$$\begin{aligned} &\int \mathcal{E}_m^*.(\mathbf{J} + \partial_t \mathbf{P}) \, \mathrm{d}x\mathrm{d}y\mathrm{d}t = \\ &\partial_z \int \mathbf{z}.[\textstyle\sum_{n,\Omega} A_n(\Omega, z) \mathcal{H}_n(\Omega) \times \mathcal{E}_m^*(\omega)] e^{i\beta_\mathrm{n}(\Omega)z - i\Omega t} e^{-i\beta_\mathrm{m}(\omega)z + i\omega t} \, \mathrm{d}x\mathrm{d}y\mathrm{d}t \\ &-\partial_z \int \mathbf{z}.[\textstyle\sum_{n,\Omega} A_n(\Omega, z) \mathcal{E}_n(\Omega) \times \mathcal{H}_m^*(\omega)] e^{i\beta_\mathrm{n}(\Omega)z - i\Omega t} e^{-i\beta_\mathrm{m}(\omega)z + i\omega t} \, \mathrm{d}x\mathrm{d}y\mathrm{d}t \ . \end{aligned} \tag{68}$$

Integration over time gives a Kronecker delta between angular frequencies, $\delta_{\Omega\omega}$, which in turn reduces the sum over Ω:

$$\begin{aligned} &\int \mathcal{E}_m^*.(\mathbf{J} + \partial_t \mathbf{P}) \, \mathrm{d}x\mathrm{d}y\mathrm{d}t = \\ &\partial_z \int \mathbf{z}.[\textstyle\sum_n A_n(\omega, z) \mathcal{H}_n(\omega, x, y) \times \mathcal{E}_m^*(\omega, x, y)] e^{i\beta_\mathrm{n}(\omega)z} e^{-i\beta_\mathrm{m}(\omega)z} \, \mathrm{d}x\mathrm{d}y \\ &-\partial_z \int \mathbf{z}.[\textstyle\sum_n A_n(\omega, z) \mathcal{E}_n(\omega, x, y) \times \mathcal{H}_m^*(\omega, x, y)] e^{i\beta_\mathrm{n}(\omega)z} e^{-i\beta_\mathrm{m}(\omega)z} \, \mathrm{d}x\mathrm{d}y \ . \end{aligned} \tag{69}$$

Collecting like terms results in an equation

$$\begin{aligned} \int \mathcal{E}_m^*.(\mathbf{J} + \partial_t \mathbf{P}) \mathrm{d}x\mathrm{d}y\mathrm{d}t = \partial_z \sum_n A_n(\omega, z) e^{i\beta_\mathrm{n}(\omega)z} e^{-i\beta_\mathrm{m}(\omega)z} \times \\ \int \mathbf{z}.[\mathcal{H}_n(\omega, x, y) \times \mathcal{E}_m^*(\omega, x, y) - \mathcal{E}_n(\omega, x, y) \times \mathcal{H}_m^*(\omega, x, y)] \, \mathrm{d}x\mathrm{d}y \ . \end{aligned} \tag{70}$$

At this point we are going to use a general property of electromagnetic modal fields which constitute an orthogonal basis: all radiative waveforms can be expressed as their linear combinations. To calculate such expansions, one can utilize the following orthogonality relation

$$\int \mathbf{z}.[\mathcal{E}_m \times \mathcal{H}_n^* - \mathcal{H}_m \times \mathcal{E}_n^*] \, \mathrm{d}x\mathrm{d}y = 2\delta_{m,n} N_m(\omega) \tag{71}$$

Here $N_m(\omega)$ is a normalization constant, whose explicit functional form has to be derived for each concrete set of modes.

Orthogonality of modes is used to reduce the sum over n in (70)

$$\int \mathcal{E}_m^*.(\mathbf{J} + \partial_t \mathbf{P}) \, \mathrm{d}x\mathrm{d}y\mathrm{d}t = -\partial_z \sum_n A_n(\omega, z) e^{i\beta_{\mathrm{n}}(\omega)z} e^{-i\beta_{\mathrm{m}}(\omega)z} 2\delta_{m,n} N_m(\omega) \ , \tag{72}$$

and we finally obtain an evolution equation for our expansion coefficients:

$$\partial_z A_m(\omega, z) = -\frac{1}{2N_m(\omega) \ XYT} \int_{-T/2}^{+T/2} \mathrm{d}t \int_{-Y/2}^{+Y/2} \mathrm{d}y \int_{-X/2}^{+X/2} \mathrm{d}x \times \\ e^{-i\beta_m(\omega)z + i\omega t} \mathcal{E}_m^*(\omega, \mathbf{r}).[\mathbf{J}(\mathbf{r}, t) + \partial_t \mathbf{P}(\mathbf{r}, t)] \tag{73}$$

This is the common representation for various z-propagated unidirectional equations, and in fact the form in which numerical solutions should be implemented. In the following sections, we will specialize this to the case of bulk media.

2.3.3 z-propagated UPPE for homogeneous media: General case

In this section, Eq. (73) is specialized for the case of a homogeneous medium. This is done by inserting explicit expressions for a given family of modal fields. In a bulk medium, these field modes are the well-known plane waves. They can be labeled by transverse wave numbers k_x, k_y, by a polarization index $s = 1, 2$, and by a $\pm$ sign signifying the direction of propagation along the z direction. Thus the index m, which we used to label modes in the preceding Section, is actually a list:

$$m \equiv k_x, k_y, s, \pm \ . \tag{74}$$

The following notation will be used for the frequency- and wave number-dependent propagation constant of a plane wave characterized by its angular frequency ω:

$$\beta_{k_x,k_y,s,\pm}(\omega) \equiv k_z(\omega, k_x, k_y) = \sqrt{k^2(\omega) - k_x^2 - k_y^2} \ , \tag{75}$$

where $k^2(\omega) \equiv \epsilon(\omega)\omega^2/c^2$ depends on the dispersive properties of the medium through the permittivity ϵ.

Electric and magnetic amplitudes in plane waves are mutually determined by Maxwell's equations. We can choose them as

$$\mathcal{E}_{k_x,k_y,s,\pm} = \mathbf{e}_s \exp\left[ik_x x + ik_y y \pm ik_z(\omega, k_x, k_y)\right] \tag{76}$$

$$\mathcal{H}_{k_x,k_y,s,\pm} = \frac{1}{\mu_0 \omega} \mathbf{k} \times \mathcal{E}_{k_x,k_y,\omega,s,\pm} \ . \tag{77}$$

The polarization of modal fields is determined by polarization vectors $\mathbf{e}_{s=1,2}$ which are of unit length and are normal to the wave-vector

$$\mathbf{k} = \{k_x, k_y, k_z \equiv \sqrt{k^2(\omega) - k_x^2 - k_y^2}\} \ . \tag{78}$$

Using the above formulas, it is straightforward to calculate the modal normalization constant

$$2N_{k_x,k_y,s,\pm}(\omega) = \int \mathbf{z}.[\mathcal{E}_m \times \mathcal{H}_m^* - \mathcal{H}_m \times \mathcal{E}_m^*]\ \mathrm{d}x\mathrm{d}y = 2\mathbf{z}\cdot[\mathbf{e}_s \times (\mathbf{k}\times\mathbf{e}_s)]\frac{1}{\mu_0\omega} = \pm 2k_z(\omega,k_x,k_z)\frac{1}{\mu_0\omega} \tag{79}$$

$$N_{k_x,k_y,s,\pm}(\omega) = \pm\frac{k_z(\omega,k_x,k_z)}{\mu_0\omega}\ . \tag{80}$$

Now we can insert expressions for modal fields and the corresponding normalization constant into coupled mode equation Eq. (73) to obtain

$$\partial_z A_{k_x,k_y,s,\pm}(\omega,z) = \mp\frac{\omega\mu_0}{2k_z}e^{\mp ik_z z}\int\frac{\mathrm{d}x\mathrm{d}y\mathrm{d}t}{L_xL_yT}e^{i(\omega t-\mathbf{k}\cdot\mathbf{r})}\times\mathbf{e}_s\cdot[\mathbf{J}(\mathbf{r},t,z)+\partial_t\mathbf{P}(\mathbf{r},t,z)] \tag{81}$$

The above integral is nothing but a spatial and temporal Fourier transform, and one can write the equations in the spectral domain

$$\partial_z A_{k_x,k_y,s,+}(\omega,z) = \frac{\omega}{2\epsilon_0c^2k_z}e^{-ik_zz}\mathbf{e}_s\cdot[i\omega\mathbf{P}_{k_x,k_y}(\omega,z)-\mathbf{J}_{k_x,k_y}(\omega,z)]\ . \tag{82}$$

This is the propagation equation that will actually be solved numerically because it is cast in terms of the slowest variables our propagation problem has. We can see that the only source of evolution in spectral amplitudes is nonlinearity.

For those who prefer to see evolution equations for electric fields proper, we express the above in terms of the electric field rather than in terms of modal expansion coefficients. From a modal expansion, the transverse part of the electric field is

$$\mathbf{E}^{\perp}_{k_x,k_y,+}(\omega,z) = \sum_{s=1,2}\mathbf{e}_s^{\perp}\mathbf{A}_{k_x,k_y,s,+}(\omega,z)e^{ik_z(\omega,\mathbf{k}_\perp)z}\ , \tag{83}$$

and therefore its z derivative reads

$$\partial_z\mathbf{E}^{\perp}_{k_x,k_y,+}(\omega,z) = ik_z(\omega,\mathbf{k}_\perp)\mathbf{E}^{\perp}_{k_x,k_y,+}(\omega,z)+\sum_{s=1,2}\mathbf{e}_s^{\perp}\partial_z\mathbf{A}_{k_x,k_y,s,+}(\omega,z)e^{ik_z(\omega,\mathbf{k}_\perp)z} \tag{84}$$

Using Eq. (82), we obtain the vectorial UPPE for a homogeneous medium:

$$\partial_z\mathbf{E}^{\perp}_{k_x,k_y,+}(\omega,z) = +ik_z\mathbf{E}^{\perp}_{k_x,k_y,+}(\omega,z)+\sum_{s=1,2}\mathbf{e}_s^{\perp}\mathbf{e}_s\ \cdot[\frac{i\omega^2}{2\epsilon_0c^2k_z}\mathbf{P}_{k_x,k_y}(\omega,z)-\frac{\omega}{2\epsilon_0c^2k_z}\mathbf{J}_{k_x,k_y}(\omega,z)] \tag{85}$$

Of course, an analogous equation holds for the backward propagating component:

$$\partial_z\mathbf{E}^{\perp}_{k_x,k_y,-}(\omega,z) = -ik_z\mathbf{E}^{\perp}_{k_x,k_y,+}(\omega,z)-\sum_{s=1,2}\mathbf{e}_s^{\perp}\mathbf{e}_s\ \cdot[\frac{i\omega^2}{2\epsilon_0c^2k_z}\mathbf{P}_{k_x,k_y}(\omega,z)-\frac{\omega}{2\epsilon_0c^2k_z}\mathbf{J}_{k_x,k_y}(\omega,z)] \tag{86}$$

This pair of equation is exact and completely analogous to the pair of z-propagated equations discussed in the previous section. Because the nonlinear polarization in these equations results as a response to the *complete* electric field, they cannot be used to calculate the forward field in isolation (i.e. without its backward propagating

counterpart). The equation becomes unidirectional only when the following approximation can be adopted:

$$\mathbf{P}(\mathbf{E}), \mathbf{J}(\mathbf{E}) \to \mathbf{P}(\mathbf{E}_f), \mathbf{J}(\mathbf{E}_f) \tag{87}$$

In other words, to obtain a closed system *which is restricted to a single direction*, we must require that nonlinear polarization can be calculated accurately from only the forward propagating field. This means that UPPE is only applicable when the back-reflected portion of the field is so small that its contribution to the nonlinearity can be neglected. Note that this assumption is inherent in any one-way propagating pulse evolution equation. Since it is the only approximation required for the UPPE, it should not be surprising that all other pulse propagation models can be derived from the UPPE by adopting further approximations.

2.3.4 z-propagated UPPE: Simplified, practical version

Equation (85), with nonlinear polarization approximated by Eq. (87) can easily become a rather large system to solve numerically. This is especially true for experiments with wide-beam, multi-TW lasers. Fortunately, in most cases transverse dimensions of resulting structures remain relatively large in comparison to wavelength, and further approximations are possible. For example in femtosecond filamentation in gases, the typical diameter of the filament core is about hundred microns which dimension is large in comparison with the laser wavelength. Consequently, the longitudinal vector component of the electric field is much smaller than the transverse (x, y) components, and can be neglected in calculation of the nonlinear medium response. It thus makes sense to take advantage of this fact to obtain a simpler equation.
Concretely, one can neglect the z components of the field and polarization vectors. In such a situation the sum over polarization vectors reduces approximately to unity

$$\sum_{s=1,2} \mathbf{e}_s^{\perp} \mathbf{e}_s \approx 1 \ . \tag{88}$$

To see this, it is enough to recall that the left-hand side constitutes a projector onto the wave-vector (recall that these vectors are mutually orthogonal). As the wave-vector is pointing in the direction almost parallel to the beam axis, it is also approximately a unity *operator* in the vector subspace spanned by x, y.

Replacing the transverse projection by unity, the full UPPE simplifies into an equation for transverse component(s)

$$\begin{aligned} \partial_z E_{k_x,k_y}(\omega, z) &= ik_z E_{k_x,k_y}(\omega, z) + \frac{i\omega^2}{2\epsilon_0 c^2 k_z} P_{k_x,k_y}(\omega, z) - \frac{\omega}{2\epsilon_0 c^2 k_z} J_{k_x,k_y}(\omega, z) \ , \\ k_z &= \sqrt{k^2(\omega) - k_x^2 - k_y^2} \ . \end{aligned} \tag{89}$$

This is the most useful form for practical calculation, and is therefore called simply UPPE. While we write it as a scalar equation, it should be understood that it is in general coupled to its counterpart governing the other polarization. The two polarization components of the electric field both contribute to the nonlinear polarization and this is how they become mutually coupled.

2.3.5 Other propagation models as approximations of UPPE

The previous section showed that Unidirectional Pulse Propagation Equations can be rigorously derived under a very general assumption that nonlinear interaction between

light and matter occurs in a regime which makes it possible to calculate the nonlinear medium response with sufficient accuracy only from the forward-propagating field component. Because this is how *all* one-way pulse propagation equation treat the nonlinearity, one can expect that other types of equations can be derived from UPPE. In fact, a universal scheme to derive all other propagation models can be given.

Several types of unidirectional propagation equation appear frequently in the literature on nonlinear optics. The most important examples are Non-Linear Schrödinger (NLS) equation [17], Nonlinear Envelope Equation [16] (NEE), the First-Order Propagation equation [13] (FOP), Forward Maxwell's equation [10] (FME), and several other equations that are closely related to these. All of these can be understood as approximations to the UPPE.

The unified derivation procedure, described in detail in Ref.[4], brings various propagation equations under one roof, and elucidates exactly what approximations must be assumed to justify their original derivations. This allows us to compare physical assumptions and approximation underlying different equations. It also reveals relations between equations which may not be obvious either because of their apparently different form, or because of different methods used in the original derivations.

It is instructive to break the derivation procedure into several steps. As a first step, we adopt a scalar, one-component approximation and write the Unidirectional Pulse Propagation Equation in the canonical form:

$$\partial_z E_{k_x,k_y}(\omega,z) = iK_z E_{k_x,k_y}(\omega,z) + iQ\frac{P_{k_x,k_y}(\omega,z)}{2\epsilon_0} \tag{90}$$

where

$$K_z(k_x,k_y,\omega) = \sqrt{k^2(\omega)-k_x^2-k_y^2} \tag{91}$$

and

$$Q(k_x,k_y,\omega) = \frac{\omega^2}{c^2 K_z(k_x,k_y,\omega)} \tag{92}$$

will be called nonlinear coupling. In most cases, the concrete form of the nonlinear polarization P is unimportant, and we will assume that it can be specified in terms of an algorithm which accepts the electric field (in general as a function of time at a given spatial location) as its input.

Let us note that this one-component, or scalar representation can be still understood as a description of a single polarization in a coupled system describing two transverse vector components of an optical field. While each equation appears scalar, the two become coupled through the polarization term, for example due to the nonlinear birefringence. These coupling effects can play a role even if a laser beam is much wider than the light wavelength, and can lead to a rich polarization dynamics within femtosecond filaments. What *is* neglected at this step is the longitudinal part of the electric field. That only becomes important when the beam focuses to a size comparable with wavelength. However, in the naturally occurring filaments, i.e, without focusing by short focal-length optical elements, such extreme focusing is never achieved, because self-focusing collapse is always arrested either by chromatic dispersion [28] or by the free-electron induced de-focusing [7,29]. Thus, for many practical purposes, the above representation is sufficiently rich and accurate.

In the second derivation step, we replace couplings K_z and Q by suitable approximations. In most cases, they are closely related to Taylor expansions in frequency and in transverse wave numbers. It is at this stage that artificial parameters are introduced into a propagation model (a typical example is the reference frequency). It is important to keep in mind that information extracted from simulations should

not depend on such degrees of freedom. In this respect, the improvements introduced into various pulse evolution equations can be viewed as corrections which (partially) restore the invariance of the model with respect to these choices.

Having specified an approximations for the linear and nonlinear coupling, we are still in the real-field representation. However, most of the published models are written using envelopes. Thus, in the next step, we obtain *envelope* equations. To do this, one can expresses the field in terms of an envelope by factoring out the carrier wave at a chosen reference angular frequency ω_r with the corresponding wave-vector $k_\mathrm{r} = K_z(0, 0, \omega_\mathrm{r})$:

$$E(\mathbf{r}, t, z) = \mathcal{A}(\mathbf{r}, t, z) e^{i(k_\mathrm{r} z - \omega_\mathrm{r} t)} \tag{93}$$

A similar factorization is of course introduced for the nonlinear polarization $P(\mathbf{r}, t, z)$ as well.

The final step consist in transforming the equation from the spectral- to the real-space representation. Mathematically, this is nothing but a Fourier transform, and the following standard replacement rules for differential operators provide quick and easy way to do this transformation:

$$ik_x \to \partial_x \qquad ik_y \to \partial_y \qquad (\omega - \omega_\mathrm{R}) \to i\partial_t \qquad \partial_z \to ik(\omega_\mathrm{R}) + \partial_z \tag{94}$$

Finally, in most cases we also transform to a frame moving with a suitable group velocity such that the pulse remains close to the center of the computational domain. We invite the reader to consult reference [4] which shows details of application of this method to several examples of propagation equations. Also note that the above procedure is closely related to that described in Section 2.2, where the Nonlinear Envelope Equation was derived from the Forward Maxwell Equation.

2.4 Pulse propagation in confined geometries

Most formulations of the nonlinear, long-distance pulse propagation problem are restricted to bulk media. A notable exception is the case of a wave-guiding structure, for example a capillary or a hollow waveguide, in which the refractive index only depends on coordinates perpendicular to the direction of propagation. Spectral unidirectional propagation equations have been derived for such structures in which the refractive index is z-independent [4]. However, knowledge of the complete system of propagating modal solutions is required if they have to be applied in practice. This is a show-stopper even in the case that exact solutions can be found. One source of difficulty is that the propagation properties of modal fields depend on solutions to an eigenvalue equation for the propagation constant, and that has to be solved numerically. Moreover, if the optical field is interacting with the medium, say one enclosed within the waveguide, the nonlinear response must be calculated for each spatial point. This in turn means that a sufficiently fast algorithm must be available to convert modal expansion representation into the real-space representation. While for the bulk media this is easily achieved with the help of fast Fourier transforms, such spectral transforms are in general not available for wave-guiding structures. This prevents us from writing down the evolution equations for the modal field amplitudes in a sufficiently closed form amenable to practical calculations.

2.4.1 Approximate methods using modal expansions

Fortunately, sometimes we can calculate a few important modes, and if the nonlinear propagation regime is such that the energy transferred to higher-order modes can be neglected, simulations are possible [48,49]. An extreme example is of course pulse propagation in fibers which have been treated in the single-transverse mode approximation with great success. However, more complex systems become also of great interest more recently. Because they are often connected with the optical filamentation regime, we will refer to this as *filamentation in restricted geometries*.

An important example is a hollow-core photonic crystal fiber. As a rule such fibers are simulated with the same technology as the traditional fiber propagation problems; Namely, the optical field is approximated by a single mode propagating in the hollow core, and its (frequency-dependent) propagation properties are usually obtained from numerical mode solvers. It is also possible to utilize a higher number of guided modes when they can be reasonably approximated. We describe such an approach next, using an example of *planar leaky waveguide*.

The planar leaky waveguide is in fact an important system in extreme nonlinear optics. It attracted attention as a device with a potential for control of femtosecond filaments, pulse self-compression, and for scaling-up the energy of high-intensity pulses. The method described here applies equally well to modeling intense pulses propagating in capillaries (important for High-Harmonic Generation), and we mention the required modifications later. Propagation effects in such waveguiding systems are important, especially in the recently explored regimes that involve a filament created within the capillary core.

Let us first consider an implementation of a spectral, unidirectional pulse propagation solver for a three-dimensional, homogeneous bulk medium. It is usually implemented in terms of plane waves, playing the role of the complete system of modes. With the help of this modal system, all quantities are expressed, and subsequently stored in the form of spectral Fourier amplitudes. What are the relevant modes of a planar hollow waveguide formed between two infinite slabs of glass? Obviously the dimensions parallel to the slabs retain plane waves as their eigenmodes. The eigenmodes in the direction perpendicular to the air-glass interfaces are of several kinds;

including those confined to the glass, and those propagating predominantly within the hollow core. The latter can form superpositions that exhibit maxima in the core and are thus interesting for us. However, these are radiation modes, and constitute a continuous family, so it is rather difficult to work with them directly. Fortunately, it is the special superpositions of the radiation modes called leaky modes that are important here.

In the first approximation, the leaky modes centered in the core can be approximated by Fourier modes of the core gap which have zeros at the air-glass boundary. Then, the bulk medium implementation of the pulse propagation solver only needs to be modified in two simple ways. First, instead of periodic basis functions, the space must be restricted such that zeros at the boundary are preserved at all times - this amounts to replacing FFT transforms with their sine-transform versions. Second, the leaky modes exhibit substantial propagation losses, and this must be properly reflected in the model. In the spectral representation this is also achieved very simply by adding an appropriate amount of imaginary part to the modal propagation constants. This is on the "background," or in addition to the whatever chromatic dispersion is caused by the medium filling in the waveguide core. Note that the geometric, or waveguide, component of dispersion is automatically accounted for through the transverse wavenumber exactly the same way as in the bulk case. The real part β_p and the imaginary part α_p of the mode-dependent wave vector $k_p = \beta_p + i\alpha_p$ for propagation within a waveguide of radius a is then given by

$$\beta_p = k(\omega)\sqrt{1 - \left(\frac{p\pi}{2k(\omega)a}\right)^2} \quad , \quad \alpha_p = \left(\frac{p\pi}{2k(\omega)a}\right)^2 \frac{n_g^2}{a\sqrt{n_g^2 - 1}} \tag{95}$$

The modification of the solver for a capillary geometry is even simpler. From the modeling point of view, the only technical difference between the planar and axially symmetric geometries is the use of the Fourier and Hankel spectral transform, respectively, which are needed for the implementation of the spectral propagator. The modes used in a radially symmetric solvers are Bessel functions $J_0(k_\perp r)$ that have zeros at the boundary of the computational domain, and they are already approximations of the leaky modes in cylindrical waveguides (capillaries). For example, the lowest-order mode is $J_0(2.405r/a)$. Once the free-space, radially symmetric propagator is implemented, the only modification required is an addition of the absorption dependent on the transverse wavenumber of each mode. The corresponding formulas can be found in the Marcatili's paper[50]. The loss-related modification of the modal propagation constant is analogous to that above:

$$\alpha_m = \left(\frac{u_m}{2\pi}\right)^2 \frac{\lambda^2}{a^3} \frac{1}{\sqrt{n_g^2 - 1}} \tag{96}$$

with u_m standing for the zero of the Bessel function.

One practically important aspect of simulations in planar hollow waveguides and capillaries is that only a few transverse modes are usually necessary. This is because the propagation loss of a mode scales with the square of its order as can be seen from the formulas above. Higher-order modes are effectively damped and their energy is "evacuated" into the glass cladding. Depending on the ratio between the wavelength and the waveguide core radius, as few as six to ten modes may be sufficient. Simulations thus become effectively close to one-dimensional, and two-dimensional in capillaries and slabs, respectively. Including only relevant modes with the lowest losses brings therefore substantial saving in compute times.

The above-described approaches have been used with success in recent investigation into extreme filamentation regimes. Midorikawa's [51] and Mysyrowicz's [52–55] groups in particular investigated planar slab waveguide arrangements as means to scale up the power in femtosecond filamentation while improving control and partially suppressing unwanted break-up into multiple filaments. It was also shown that efficient pulse compression can be achieved in the interplay between filamentation and effects induced by the leaky wave-guiding structure. The computational methods are described in detail in Ref. [53,55]

Another application example is high-harmonic generation in capillaries pressurized with inert gases [56]. As demonstrated recently, an interesting new regime occurs which exhibits signatures of filamentation confined within the capillary bore, and where the efficiency of extremely-high (up to 5-thousand) harmonic order radiation generation is controlled by the dynamics of the mid-infrared excitation pulse.

2.4.2 Nonlinear pulse propagation in waveguides: generalized UPPE.

Most pulse propagation models are designed for bulk media, and can not deal with situations in which material interfaces are present that affect the propagation of the laser pulse. This subsection discusses a generalized version of pulse propagation equations which overcomes this limitation: Derivation of the Generalized Unidirectional Pulse Propagation Equation (gUPPE) is sketched here for waveguiding structures with z-invariant refractive index.

Consider a geometry given by a frequency-dependent relative permittivity which only depends of the coordinates transverse to the direction of propagation. The latter is taken to be the z-axis, and $\epsilon(\mathbf{r}_\perp, \omega)$ will stand for the susceptibility at the angular frequency ω, at the transverse location $\mathbf{r}_\perp$. The idea is to separate, without any approximations, the effects related to the chromatic properties and the geometry of the waveguide, and the nonlinear interaction of light with the materials that constitute the structure.

The linear propagation regime is controlled by the linear operator $\hat{L}$, closely related to the Helmholtz equation. It reflects the geometry and material of the problem, and acts on the transverse electric vector field as follows:

$$\hat{L}\mathbf{E}_\perp \equiv \frac{\omega^2}{c^2}\epsilon(r_\perp, \omega)\mathbf{E}_\perp + \Delta_\perp \mathbf{E}_\perp + \nabla\frac{1}{\epsilon}\mathbf{E}_\perp.\nabla_\perp\epsilon. \tag{97}$$

All nonlinear interactions are included in the operator $\hat{N}$ that acts on $\mathbf{E}_\perp$, and in general also depends on the E_z component:

$$\hat{N}[\mathbf{E}] \equiv \frac{\omega^2}{\epsilon_0 c^2}\mathbf{P}(\mathbf{E}) + \nabla\frac{1}{\epsilon_0\epsilon}\nabla\cdot\mathbf{P}(\mathbf{E}). \tag{98}$$

Depending on the concrete form of the polarization model, the above expression may depend of E_z. If it does, the longitudinal component of the electric field can be obtained from the divergence equations. For simplicity, the nonlinear current density is omitted as all is straightforward to generalize.

To construct the propagation equations for $\mathbf{E}_\perp(z, x, y, \omega)$, auxiliary field amplitudes are introduced first, doubling the number of variables utilized to describe the electric field:

$$E_i(z, x, y, \omega) = E_i^+(z, x, y, \omega) + E_i^-(z, x, y, \omega), \tag{99}$$

where

$$\begin{aligned} E_i^+ &= A_i^+(z, x, y, \omega)e^{+i\zeta z} \\ E_i^- &= A_i^-(z, x, y, \omega)e^{-i\zeta z}, \end{aligned} \tag{100}$$

and $i = x, y$ and ζ stands for a parameter to be chosen freely. Because no ζ appears in Maxwell's equations, no physical observables can depend on it. This "gauge invariance" becomes manifest in the final result. ζ is called a reference wavenumber to indicate that it has no physical meaning by itself.

Next we need a projection operation that selects from an arbitrary pulsed waveform the portion that propagates in the positive (forward) z direction. Similar to Ref. [9], two projector operators can be constructed from the Helmholtz operator $\hat{L}$ and its square root $\hat{L}^{\frac{1}{2}}$:

$$\begin{aligned}\mathcal{P}_F &\equiv \frac{\hat{L}^{-\frac{1}{2}}}{4\zeta}\begin{pmatrix} +(\zeta+\hat{L}^{\frac{1}{2}})^2 & +(\hat{L}-\zeta^2) \\ -(\hat{L}-\zeta^2) & -(\zeta-\hat{L}^{\frac{1}{2}})^2 \end{pmatrix} \\ \mathcal{P}_B &\equiv \frac{\hat{L}^{-\frac{1}{2}}}{4\zeta}\begin{pmatrix} -(\zeta-\hat{L}^{\frac{1}{2}})^2 & -(\hat{L}-\zeta^2) \\ +(\hat{L}-\zeta^2) & +(\zeta+\hat{L}^{\frac{1}{2}})^2 \end{pmatrix}\end{aligned} \tag{101}$$

It is straightforward to show that these operators have the usual properties of projectors,

$$\mathcal{P}_F^2 = \mathcal{P}_F \quad \mathcal{P}_B^2 = \mathcal{P}_B \ , \tag{102}$$

and

$$\mathcal{P}_F + \mathcal{P}_B = 1 \ , \ \mathcal{P}_F\mathcal{P}_B = \mathcal{P}_F\mathcal{P}_B = 0 \ . \tag{103}$$

It can also be shown that eigenvectors of $\mathcal{P}_F$ and $\mathcal{P}_B$ have propagation constants equal to those of the linear forward and backward propagating modes, respectively. This means that these operators are the right pair to define the split into forward and backward going waves in the linear regime. To do this, unidirectional amplitudes are produced with $\mathcal{P}_{F,B}$ such that they only exhibit evolution in the presence of nonlinearity:

$$E_\perp^F = e^{+i\sqrt{\hat{L}}z}A_\perp^F(z) \ , \ E_\perp^B = e^{-i\sqrt{\hat{L}}z}A_\perp^B(z) \ . \tag{104}$$

Note that $A_\perp^{F,B}(z)$ are analogous to the spectral, i.e. plane-wave, representation of the electric field in bulk media. Their evolution equation is

$$\begin{aligned}\partial_z A_\perp^F &= \frac{+i}{2\sqrt{\hat{L}}}e^{-i\sqrt{\hat{L}}z}\hat{N}_\perp[e^{+i\sqrt{\hat{L}}z}A^F + e^{-i\sqrt{\hat{L}}z}A^B] \\ \partial_z A_\perp^B &= \frac{-i}{2\sqrt{\hat{L}}}e^{+i\sqrt{\hat{L}}z}\hat{N}_\perp[e^{+i\sqrt{\hat{L}}z}A^F + e^{-i\sqrt{\hat{L}}z}A^B]\end{aligned} \tag{105}$$

This system shows that the coupling of the forward and backward propagating is mediated by the nonlinear terms. This is the point at which the unidirectional approximation is invoked. Namely, it is assumed that the backward propagating fields are negligible in the sense that they do not contribute to the nonlinearity. Under such conditions, the nonlinear term reads

$$\hat{N}_\perp[e^{+i\sqrt{\hat{L}}z}A^F + e^{-i\sqrt{\hat{L}}z}A^B] \approx \hat{N}_\perp[e^{+i\sqrt{\hat{L}}z}A^F] \ , \tag{106}$$

and the system is reduced to only the forward-propagating field:

$$\partial_z A_\perp^F(r_\perp, \omega, z) = +\frac{i}{2\sqrt{\hat{L}}}e^{-i\sqrt{\hat{L}}z}\hat{N}_\perp[e^{+i\sqrt{\hat{L}}z}A^F]. \tag{107}$$

This is the generalization of the Unidirectional Pulse Propagation Equation. It is analogous to the bulk UPPEs of Eq. (214), with the linear and nonlinear effects

cleanly separated into linear pulse propagator $\exp(i\hat{L}^{1/2}z)$, and nonlinear coupling between linear modes.

As one should expect, the above propagation equation reduces to a full vector version of UPPE for bulk media, showing that the latter is just a special case of Eq.(107). One practically important property is that once an implementation for the linear propagator is available, the numerical technique to solve gUPPE is nearly identical to that described earlier for UPPE.

Since the linear propagator is diagonal in angular frequency, its numerical application is equivalent to a set of independent beam-propagation problems. The action of $\exp(i\hat{L}^{1/2}z)\psi$ only requires one independent BPM-like update for each ω resolved in the simulation. Many wide-angle BPM methods available, and any of them can be employed in gUPPE. However, the optimal choice depends on the geometry and material properties in the given pulse-propagation problem.

2.5 Medium response and nonlinear interactions

This section describes typical components of medium response models in nonlinear optics of ultrashort pulses. They all can be viewed as contributions to the polarization or to the induced current density, both of which appear in Maxwell's equations or in the wave equation as source terms.

2.5.1 Optical Kerr effect

We will use the optical Kerr effect as a prototypical example of nonlinear source term that can be included in the polarization. At the dominant third order for a centro-symmetric medium, the nonlinear polarization $\mathbf{P}$ reads :

$$\mathbf{P} \equiv \epsilon_0 \chi^{(3)} \mathbf{E}^3 \tag{108}$$

By expressing the scalar components of the electric field as an envelope and a carrier: $E = \frac{1}{2}[\mathcal{E}\exp(ik_0 z - i\omega_0 t) + \mathcal{E}^* \exp(-ik_0 z + i\omega_0 t)]$, we obtain an expression for E^3:

$$E^3 = \frac{1}{8}[\mathcal{E}^3 \exp(i3k_0 z - i3\omega_0 t) + 3|\mathcal{E}|^2 \mathcal{E}\exp(ik_0 z - i\omega_0 t) + \text{c.c.}], \tag{109}$$

where c.c. denotes complex conjugation. By introducing Eq. (109) into Eq. (108), we identify the nonlinear polarization envelope $\mathcal{P}$ from its carrier envelope decomposition $P = \frac{1}{2}[\mathcal{P}\exp(ik_0 z - i\omega_0 t) + \mathcal{P}^* \exp(-ik_0 z + i\omega_0 t)]$:

$$\mathcal{P} \equiv \epsilon_0 \chi^{(3)} \frac{3}{4} |\mathcal{E}|^2 \mathcal{E}. \tag{110}$$

It is readily seen that Equation (109) contains a component oscillating at the third harmonic ($3\omega_0$) that has been discarded in the nonlinear polarization, as we only identified the fundamental components oscillating at ω_0 to derive the nonlinear polarization envelope (110). Thus, introduction of Eq. (110) in an envelope propagation model describes the effects called self-focusing (for a positive $\chi^{(3)}$) and self-phase modulation but not third-harmonic generation. Next section will show how to describe the latter effect either with an envelope or a carrier resolving propagation equation.

Up to now, we have written our models with *hidden* material coefficients such as, e.g., the linear susceptibility $\chi^{(1)}(\omega)$ which is included in the dispersion relation $k(\omega)$ of the medium, in turn involving specific resonance frequencies and amplitudes. For practical situations, it is needed to supply numerical codes with parameter values for the material coefficients. For the nonlinear third-order susceptibility, values may be obtained from ab-initio calculations but measurements of the nonlinear index coefficient $n_2 \equiv 3\chi^{(3)}/4\epsilon_0 c n_0^2$ in units of m^2/W may also be found in the literature. For instance, see Refs. [25] for optical crystals. By introducing the above definition for the nonlinear index coefficient n_2 in Eq. (110), and by using the definition of the intensity $\mathcal{I} \equiv \epsilon_0 c n_0 |\mathcal{E}|^2/2$, an expression is obtained for the nonlinear polarization envelope modeling an instantaneous Kerr response of the medium due to the electronic contribution to the polarization:

$$\frac{\mathcal{P}}{\epsilon_0} \equiv 2 n_0 n_2 \mathcal{I} \mathcal{E}, \tag{111}$$

where both the linear refraction index n_0 and the nonlinear index $n_2\mathcal{I}$ are dimensionless, since $\mathcal{I}$ is expressed in W/m^2 and n_2 in m^2/W. Eq.(111) can then be introduced

into all nonlinear envelope propagation equations. Using for instance Eq. (51) to describe the propagation of a monochromatic beam, which allows us to neglect dispersion ($k_0^{(2)} = 0$), we obtain the NLS equation with its standard cubic nonlinearity:

$$\frac{\partial \mathcal{E}}{\partial z} = \frac{i}{2k_0}\Delta_\perp \mathcal{E} + i\frac{\omega_0}{c} n_2 \mathcal{I}\mathcal{E} \tag{112}$$

Equation (112) models beam propagation under the effects of diffraction and the optical Kerr effect, leading to beam self-focusing (for a positive n_2), i.e. to the cumulative lens effect bending the phase fronts due to the higher refraction index in the most intense part of the beam. In a planar geometry, beam self-focusing and diffraction alternatively prevail, resulting in beam width oscillations without energy losses [26]. In cylindrical geometry, it is known that Eq. (112) is mathematically singular: Beams with power above a certain threshold P_{cr} undergo catastrophic collapse at a finite distance [27]. For Gaussian beams, this critical threshold is given by:

$$P_{cr} = \frac{3.77\pi n_0}{2k_0^2 n_2}. \tag{113}$$

For a collimated Gaussian beam $\mathcal{E} = \mathcal{E}_0 \exp(-r^2/w_0^2)$ of power $P_{in} \equiv \pi w_0^2 \mathcal{I}_0/2$, where $\mathcal{I}_0 \equiv \epsilon_0 c n_0 \mathcal{E}_0^2/2$, the self-focusing (collapse) distance follows [27]:

$$z_c = \frac{0.367 z_R}{\sqrt{[(P_{\rm in}/P_{\rm cr})^{1/2} - 0.852]^2 - 0.0219}} \tag{114}$$

where $z_R \equiv k_0 w_0^2/2$ denotes the Rayleigh (typical diffraction) length. Section 3.1.2 shows how to implement model (112) and Eqs. (113) and (114) provide analytical scaling laws for the collapse distance and the critical power threshold that can be used as test cases to check the correct implementation of the model.

2.5.2 Optical Kerr effect and third harmonic generation

Closely related to the optical Kerr effect caused by electrons in bound states is third harmonic generation which is often observed in femtosecond filaments. In fact, from a certain point of view the two effects are actually one.

Consider an instantaneous (electronic) Kerr effect in an isotropic medium. Also consider a propagation model which simulates directly the physical field rather than its envelope. A polarization response that is third-order in the electric field E must be constructed solely from $\mathbf{E}$ taken at the given instant in time, so there is a single vector to *work with*. That is why, the only possible form of an *instantaneous* third-order nonlinearity is as given in Eq. (108).

Note that as soon as there is memory, two frequency dependent components of third-order susceptibility tensor are needed for full description. The important point is that the frequency content of Eq. (108) consists of both the fundamental frequency of E and of its third harmonic. This is the main source of third harmonic radiation observed in femtosecond filaments (although it is not the only one).

It is worthwhile to remark that Readers will find in the literature also a very different approach to modeling third-harmonic generation. In works and simulations based on application of envelope pulse propagation equations, several authors used two envelope functions, one for the fundamental frequency and one for the third harmonic generation [30]. The electric field decomposition into carriers and envelopes with these two components reads: $E = \frac{1}{2}[\mathcal{E}_{\omega_0} \exp(ik_{\omega_0} z - i\omega_0 t) + \mathcal{E}_{3\omega_0} \exp(ik_{3\omega_0} z - i3\omega_0 t) + \text{c.c.}]$,

and the nonlinear polarization can be decomposed similarly. Introduction of this decomposition into Eq. (108) yields expressions for the fundamental and third harmonic envelope components of the nonlinear polarization:

$$\mathcal{P}_{\omega_0} = \epsilon_0 \chi^{(3)} \frac{3}{4} [(|\mathcal{E}_{\omega_0}|^2 + 2|\mathcal{E}_{3\omega_0}|^2)\mathcal{E}_{\omega_0} + \mathcal{E}_{\omega_0}^{*2}\mathcal{E}_{3\omega_0}], \tag{115}$$

$$\mathcal{P}_{3\omega_0} = \epsilon_0 \chi^{(3)} \frac{3}{4} [(|\mathcal{E}_{3\omega_0}|^2 + 2|\mathcal{E}_{\omega_0}|^2)\mathcal{E}_{3\omega_0} + \mathcal{E}_{\omega_0}^{3}/3]. \tag{116}$$

For each component $\mathcal{P}_{\omega_0}$ and $\mathcal{P}_{3\omega_0}$, the first two terms represent self- and cross- phase modulation. The third terms are responsible for energy exchange between fundamental and third harmonics fields, namely third harmonic generation and back conversion. Envelope propagation equation for each component $\mathcal{E}_{\omega_0}$ and $\mathcal{E}_{3\omega_0}$ are derived following the methods presented in section 2.2 and follow the canonical form valid for envelopes with broad spectra.

It has to be emphasized that the two-envelope method can only be justified as long as spectral components centered around the fundamental and third harmonic frequency are well separated. However, spectra can become extremely broad in femtosecond filamentation. Then, distinction between fundamental and third harmonic is impossible, and any two-envelope parametrization is therefore non-unique leading to a fundamentally inconsistent model. Thus, if harmonic frequencies are expected to appear in the numerical experiment and if the spectrum for the fundamental component broadens sufficiently to overlap the third harmonic spectrum, a carrier resolving model implementing the optical Kerr effect as in (108) should be used. This will naturally capture also generation of other odd harmonics in a cascade process which may become important for longer-wavelength filamentation.

2.5.3 Nonlinear absorption

The NLS Equation (112) leads to optical beam collapse in cylindrical geometry but in a real experiment, saturation mechanisms prevent collapse to occur [29]. This means that the mathematical singularity of Eq. (112) must be cured by extending the model to a more realistic case. Close to the collapse, the beam intensity is sufficient to ionize the medium after absorption of several photons. The process is associated with nonlinear absorption of energy, which is the main physical effect playing a saturation role in preventing the collapse to occur[7].

Nonlinear absorption is described by an effective current $\mathbf{J}$ such that the averaged dissipated power corresponds to that necessary for optical field ionization of the medium with density of neutral atoms ρ_{nt}, ionization potential (or gap for a solid medium) U_i and intensity-dependent ionization rate $W(\mathcal{I})$ [32]. Different ionization regimes exist and correspond to different ionization rates. For example for intensities smaller than a certain threshold, ionization occurs in the multiphoton regime and requires absorption of several (K) photons to liberate an electron. In this text, without loss of generality, we will consider only the multiphoton regime for which a simple law for ionization rates reads $W(\mathcal{I}) = \sigma_K \mathcal{I}^K$, where σ_K denotes the cross section for multiphoton ionization. The above condition for power dissipation is expressed as

$$\frac{1}{2}\mathbf{J} \cdot \mathbf{E}^* = W(\mathcal{I}) K \hbar \omega_0 \rho_{nt} \tag{117}$$

[7] Note that plasma induced defocusing can also prevent collapse to occur, however, nonlinear absorption of energy is necessary to prevent subsequent catastrophic behavior [31]

The current is therefore obtained as

$$\frac{\mathbf{J}}{\epsilon_0 c} = n_0 \frac{W(\mathcal{I}) K \hbar \omega_0 \rho_{nt}}{\mathcal{I}} \mathbf{E}, \tag{118}$$

and its envelope counterpart, in the multiphoton regime, reads as

$$\frac{\mathcal{J}}{\epsilon_0 c} = n_0 \beta_K \mathcal{I}^{K-1} \mathcal{E}. \tag{119}$$

where $\beta_K \equiv K\hbar\omega_0 \sigma_K \rho_{nt}$ denotes the cross section for multiphoton absorption.

By introducing Eq. (119) into the nonlinear envelope equations derived in section 2.2, the effect of multiphoton absorption is accounted for in propagation models. For instance, with the change $\mathcal{P} \to \mathcal{P} + i\mathcal{J}/\omega_0$ the NLS Equation (51) for a monochromatic beam undergoing diffraction, optical Kerr effect and multiphoton absorption becomes:

$$\frac{\partial \mathcal{E}}{\partial z} = \frac{i}{2k_0} \Delta_\perp \mathcal{E} + i \frac{\omega_0}{c} n_2 \mathcal{I} \mathcal{E} - \frac{\beta_K}{2} \mathcal{I}^{K-1} \mathcal{E} \tag{120}$$

Equation (120) is an extended NLS equation that is also valid to describe the propagation of pulses provided they keep a narrow spectrum during their nonlinear propagation.

An evolution equation for the pulse intensity is obtained by multiplying Equation (120) by $\mathcal{E}^*$ and by adding the result to its complex conjugate:

$$\frac{\partial |\mathcal{E}|^2}{\partial z} = \frac{i}{2k_0} [\mathcal{E}^* \Delta_\perp \mathcal{E} - \mathcal{E} \Delta_\perp \mathcal{E}^*] - \beta_K \mathcal{I}^{K-1} |\mathcal{E}|^2 \tag{121}$$

Note that the Kerr term is no longer present in Eq. (121), reflecting the fact that the optical Kerr effect does not directly modify the laser beam intensity but leads to nonlinear phase accumulation. Spatial phase gradients mediated by the first term on the r.h.s. of Eq. (121) then lead to an energy flux toward the beam center, which is responsible for an increase of the beam intensity. For a very large beam, the term $[\mathcal{E}^* \Delta_\perp \mathcal{E} - \mathcal{E} \Delta_\perp \mathcal{E}^*]$ can be neglected in the central part of the beam[8], the intensity of which is governed by:

$$\frac{\partial \mathcal{I}}{\partial z} = -\beta_K \mathcal{I}^K, \tag{122}$$

An analytical solution of Eq. (122) reads

$$\mathcal{I}(z) = \frac{\mathcal{I}_0}{(1 + (K-1)\beta_K \mathcal{I}_0^{K-1} z)^{1/(K-1)}} \tag{123}$$

Tests of the numerical implementation of nonlinear energy losses can be easily made by comparison of simulation results with their analytical counterpart from Equation (123).

2.5.4 Plasma generation and plasma defocusing

When the pulse is so intense that it ionizes the medium, the contribution of the plasma follows from the evolution equation for the plasma current density:

$$\frac{\partial \mathbf{J}}{\partial t} + \frac{\mathbf{J}}{\tau_c} = \frac{q_e^2}{m_e} \rho \mathbf{E}, \tag{124}$$

[8] By denoting ϕ the phase of the beam, a straightforward calculation shows that $[\mathcal{E}^* \Delta_\perp \mathcal{E} - \mathcal{E} \Delta_\perp \mathcal{E}^*] = 2i \nabla_\perp \cdot (|\mathcal{E}|^2 \nabla_\perp \phi)$. For a large beam, this term is small close to the beam center where the phase is flat.

where ρ denotes the electron density and τ_c is the electron collision time. Equation (124) can be solved in the Fourier domain and its solution introduced in propagation equations with a source term $(\omega/c)(\hat{J}/\epsilon_0 c)$ involving spectral components of the free charge current. From Eq. (124), we obtain:

$$\frac{\omega}{c}\frac{\hat{\mathbf{J}}}{\epsilon_0 c} = \frac{q_e^2 \omega \tau_c}{\epsilon_0 c^2 m_e}\frac{1+i\omega\tau_c}{1+\omega^2\tau_c^2}\widehat{\rho\mathbf{E}}. \tag{125}$$

We rewrite Eq. (125) as:

$$\frac{\omega}{c}\frac{\hat{\mathbf{J}}}{\epsilon_0 c} = k(\omega)\sigma(\omega)\widehat{\rho\mathbf{E}}, \tag{126}$$

where

$$\sigma(\omega) = \frac{\omega_0}{n(\omega)c\rho_c}\frac{\omega_0\tau_c(1+i\omega\tau_c)}{(1+\omega^2\tau_c^2)} \tag{127}$$

is a complex frequency-dependent coefficient with real part equal to the cross section for inverse Bremsstrahlung [33] and $\rho_c \equiv \epsilon_0 m_e \omega_0^2/q_e^2$, the critical plasma density above which the plasma becomes opaque to the laser beam at frequency ω_0. This is known as the Drude model. Note that $\sigma(\omega)$ is not effectively depending on the central frequency of the laser pulse ω_0 since the quantity ω_0^2/ρ_c in Eq. (127) is a constant, the critical density being only a reference chosen for the pulse central frequency ω_0.

The current in Equation (126) accounts for plasma absorption (real part) and plasma defocusing (imaginary part). Both effects are frequency dependent, for example defocusing is stronger for longer wavelengths. It can be important that our models capture such dispersive behavior, for example in pump probe experiments with different pulse wavelengths.

To close the model, one needs to know the evolution of the electron density $\rho(\mathbf{r}, t)$ entering in Eq. (126). It is is governed by a rate equation in the form:

$$\frac{\partial\rho}{\partial t} = W_{\text{ofi}}(\mathcal{I})(\rho_{\text{nt}} - \rho) + W_{\text{ava}}(\mathcal{I})\rho \tag{128}$$

where the first term on the r.h.s. of Eq. (128) represents optical field ionization and the second term represents avalanche ionization. As stated in section 2.5.3, optical field ionization in the multiphoton regime occurs with a rate $W_{\text{ofi}}(\mathcal{I}) = \sigma_K \mathcal{I}^K$. The rate for avalanche ionization can be considered as proportional to the pulse intensity $W_{\text{ava}}(\mathcal{I}) = \sigma(\omega_0)\mathcal{I}/U_i$, where $\sigma(\omega_0)$ is the inverse Bremsstrahlung coefficient given by Eq. (127) evaluated at the central frequency ω_0 of the laser pulse and U_i the ionization potential.

An example of nonlinear envelope equation with nonlinear terms accounting for the optical Kerr effect, nonlinear absorption and plasma effects is obtained by introducing the corresponding source terms in the FEE (38):

$$\frac{\partial\hat{\mathcal{E}}}{\partial z} = \frac{i}{2k(\omega)}\Delta_\perp\hat{\mathcal{E}} + i[k(\omega) - \kappa(\omega)]\hat{\mathcal{E}} + i\frac{\omega}{c}\frac{n_0}{n(\omega)}n_2\widehat{\mathcal{I}\mathcal{E}} - \frac{\beta_K}{2}\widehat{\mathcal{I}^{K-1}\mathcal{E}} - \frac{\sigma(\omega)}{2}\widehat{\rho\mathcal{E}} \tag{129}$$

2.5.5 Raman-Kerr effect

The optical Kerr effect includes in general the electronic contribution which is nearly instantaneous and a delayed component of fraction α, due to stimulated molecular Raman scattering.

Air is by a large part made of two-atomic molecules with different polarizabilities parallel and perpendicular to their symmetry axes. This leads to a nonlinear effect

which is referred to as stimulated Raman effect, although the recent nomenclature acknowledges the fact that dynamic reorientation of molecules plays a central role in it.

The interaction energy of a molecule in an external field is such that it prefers to align with the direction of the field. When a femtosecond pulse hits such a molecule, it excites rotational motion; this is a stimulated Raman effect. Molecular rotation then changes the effective linear polarizability of the molecule as projected on the direction of the field. Because the interaction Hamiltonian is quadratic in field, the effect is of third-order. It is therefore often considered a companion of the electronic Raman effect. Taken a very different microscopic origins, this may seem arbitrary, but one has to keep in mind that manifestations of the two effects (i.e. self-focusing) are very difficult to distinguish in longer pulses.

A proper, first-principles model would need to integrate quantum mechanical equations of motion for a density matrix describing the rotational state of an ensemble of molecules. Note that such a system is to be solved at each spatial grid location, and at each propagation step! Instead of this (relatively) difficult calculation, the Raman effect is approximately parametrized as described next.

Let us denote Q_i a generalized coordinate of an effective oscillator *embedded* in the medium which responds to a force that is quadratic in external field,

$$\frac{\partial^2 Q_i}{\partial t^2} + 2\Gamma\frac{\partial Q_i}{\partial t} + (\omega_R^2 + \Gamma^2)Q_i = (\omega_R^2 + \Gamma^2)|\mathcal{E}(r,t,z)|^2, \tag{130}$$

for Q_i with boundary conditions $\partial Q_i/\partial t(-\infty) = 0$ and $Q_i(-\infty) = 0$. The solution to Eq. (130) reads:

$$Q_i(r,t,z) = \int_{-\infty}^{t} \mathcal{R}_0 \exp[-\Gamma(t-\tau)] \sin[\omega_R(t-\tau)]|\mathcal{E}(r,\tau,z)|^2 d\tau, \tag{131}$$

where $\mathcal{R}_0 = (\Gamma^2 + \omega_R^2)/\omega_R$.

The nonlinear polarization for the Kerr term with its Raman contribution therefore reads as

$$\frac{\mathcal{P}}{\epsilon_0} = 2n_0 n_2 \left((1-\alpha)\mathcal{I}(r,t,z) + \alpha \int_{-\infty}^{t} \mathcal{R}(t-\tau)\mathcal{I}(r,\tau,z)\, d\tau \right) \mathcal{E}\,, \tag{132}$$

where $\mathcal{I} = \epsilon_0 c n_0 |\mathcal{E}(r,t,z)|^2/2$. The function $\mathcal{R}(t)$ mimics the molecular response with a characteristic time Γ^{-1} and frequency ω_R:

$$\mathcal{R}(t) = \mathcal{R}_0 \exp(-\Gamma t) \sin \omega_R t \tag{133}$$

One advantage of the above phenomenological approach is that it can be applied also to other types of stimulated Raman scattering. For example in glasses, this single-oscillator model can serve as an acceptable approximation of what is actually a significantly more complex process. It can also be generalized to include multiple effective oscillators, which in turn can mimic the true medium response quite accurately. In water, the single-oscillator model could accurately reproduce measured signatures of stimulated Raman scattering in angularly resolved spectra [34,35]. We will discuss efficient numerical implementation of this and similar models in Section 3.3.

2.6 Light-matter interaction model extensions

The model for light-matter interaction described in the previous section consists of more or less independent components that were conceived when typical laser pulses were longer, and less powerful. It is thus not too surprising that recently it becomes obvious that its useful lifetime is limited. The following are just few of the conceptual problems in the current approach:

- electronic states are considered either bound or completely free
- weakly bound and free, but correlated electronic states are neglected
- ionization is modeled as a rate, and as such neglects the history of the system subject to the time-dependent external field
- the ionization model has no natural extension to multicolor, broad-bandwidth excitation pulses
- there is no refractive interaction that is causally coupled to the ionization
- the ionization-related energy losses are accounted for through a purely phenomenological current arbitrarily added to the propagation equation
- Kerr-related and plasma-related components of the model are independent, thus not reflecting the fact that these effects have a common origin and are therefore intimately related

The phenomenological nature of the "standard" light-matter interaction model used in pulse propagation studies of femtosecond filamentation and also in high-harmonic generation modeling has been recently recognized. Efforts aimed at qualitative improvement are underway in several groups and it is to be expected that a lot of work will be devoted to these issues in the near future. One possible approach is to couple TDSE simulations directly to pulse propagation solvers (see [57] for one of the first works), and apply massive computing power. Such truly first principle approaches have been pursued e.g. by the Bandrauk's group [58–60]. However, so far direct attacks are rather restricted in their applicability due to the extreme computational resources they require even for relatively "small" modeling problems. Research will therefore continue to explore other approaches which acknowledge that all the effects that comprise the standard model originate from a complex electronic system response.

The goal of the following section is to review the various options from this viewpoint. Because of the enormous breadth of the light-matter interaction field, these notes only concentrate on those models and theories that have been, at least in some form, designed to study *spatially resolved pulse propagation simulation* coupled to first-principle light-matter interactions.

2.6.1 Quintic nonlinearity and Kerr effect saturation

While the collapse arrest by the combined effect of multi-photon ionization losses and de-focusing by freed electrons have been considered to be the two most important mechanisms, researchers have been looking for alternatives, too. It is fair to say that in the early years of the filamentation research and modeling, part of the motivation also came from the difficulties related to numerical resolution of the often violent arrest of the self-focusing collapse. It was quite natural to ask if the numerical difficulties indicated that additional mechanisms also contributed to the collapse-arresting processes.

For applications to filaments, the fifth-order susceptibility was often considered in conjunction with plasma effects as a way to soften the collapse, and modify the intensities occurring in the filament cores [61,62,29,63]. The argument is based on the

relative comparison of nonlinear effect of different orders. In the perturbative regime, it is expected that the ratio of the higher-order nonlinear polarization orders scales as [64]

$$\frac{P^{(k+2)}}{P^{(k)}} \approx \frac{|E|^2}{|E_{atomic}|^2} \ .$$

With the atomic field strength of $E_{atomic} \approx 10^{10}\mathrm{Vm}^{-1}$, the higher-order nonlinearity contribution is expected to be small. Nevertheless, filamentation can not be viewed as a strictly perturbative regime despite the fact that the intensities involved are still a couple of orders of magnitude lower than atomic fields; In fact, optical filamentation is a regime on the boundary between perturbative and strong-fields. It is thus not unreasonable to expect that, for example, quintic nonlinearity could saturate the Kerr-induced self-focusing (see e.g. [67,66,65]).

2.6.2 Higher-order Kerr nonlinearity

In 2009, Loriot and co-authors performed a pump-probe experiment [68] in which a probe beam polarized at 45 degrees with respect to the strong pump pulse polarization direction detected a time-resolved birefringence induced by the interaction within the femtosecond pulse. Authors interpreted their findings as evidence for the instantaneous, Kerr-type nonlinearity of a very high order. In particular, it was proposed that already for light intensities that are readily available in femtosecond optical filaments, the usual Kerr effect alone is insufficient to describe the nonlinear response due to bound electrons, and must be replaced by the so-called higher-order Kerr effect (later termed HOKE) for which the intensity dependent nonlinear index becomes a highly nonlinear function of instantaneous light intensity:

$$\Delta n(I) = n_2 I + n_4 I^2 + n_6 I^3 + n_8 I^4 + n_{10} I^5 \tag{134}$$

An important feature of this nonlinear modification of refractive index was that it not only saturates, but turns negative with a steep slope at a cross-over intensity which depends on the gas species. Not long after the original experiment, this particular feature was invoked to question the filamentation paradigm that had been accepted for more than a decade, and it was suggested that plasma was not necessary as an effect crucial for the self-focusing collapse arrest. The debate that followed (see e.g. [69,70,73,74,71,72,79,75,76,78,80–87,77,88]) has not been completely settled at time of this writing, and it is not our goal to add to the particular discussion. Rather, we concentrate on the numerical modeling issues related to this higher-order nonlinearity and the consequences that the potential acceptance of this kind of model implies.

From the numerical implementation point of view the HOKE model does not require anything special beyond a simple replacement of the nonlinear index coefficient already present in the standard filamentation model with the prescription shown above. This is indeed how the modification was implemented in various simulation tests of the theory. However, physical questions concerning the precise meaning of the higher-order nonlinear index immediately arise. If the cycle averaged intensity is used to calculate $\Delta n(I)$, the resulting nonlinear polarization does not contain any higher-order harmonics, including the third and fifth, both of which have been experimentally observed numerous times in femtosecond filaments. This is a drawback that must be dealt with by adding a dedicated nonlinear term that generates the third and fifth harmonic. Alternatively, assuming truly instantaneous response, one could replace I in $\Delta n(I)$ with:

$$\Delta P(t) \approx \Delta n(E^2)E = \left[\tilde{n}_2(E^2) + \tilde{n}_4(E^2)^2 + \tilde{n}_6(E^2)^3 + \tilde{n}_8(E^2)^4 + \tilde{n}_{10}(E^2)^5\right] E(t) \tag{135}$$

The above expression would be in line with the conclusion of the original experiment that the new effect was instantaneous. However, it would automatically generate harmonics of up to order eleven, and this would happen directly, without the need of a cascaded process that is normally responsible for the appearance of the fifth harmonic from the third harmonic radiation. Clearly, neither of the two possible formulations feels very satisfactory.

It thus becomes obvious that if the HOKE effect is indeed operational in some femtosecond filamentation regimes, its modeling will require more accurate specification of its memory or delay time, and how it can be accounted for in the numerical model.

2.6.3 Molecular reorientation in strong fields

As a laser pulse propagates through a molecular gas medium, it causes the molecules to rotate and align, at least partially, with the electric field. The origin of this effect is in the different polarizibility of an asymmetric molecule; it is larger along the molecule axis (say, in an O_2 or N_2) than in directions perpendicular to it. A simple picture involves a model of such a molecule in the form of a rigid rotor. The polarizibility of the medium is then given by an ensemble average of molecular contributions. The latter can be written as

$$\mathbf{P} = \alpha_\perp \mathbf{E} + \Delta\alpha \mathbf{n}(\mathbf{n}.\mathbf{E})$$

where $\mathbf{n}$ is a unit vector along the molecule axis. To account for the chaotic, or partially ordered orientation of molecules in the gaseous ensemble, one can write the resulting polarization response as a function of the mean orientation angle θ between the molecular axis and polarization direction of the field:

$$\mathbf{P} = (\alpha_\perp + \Delta\alpha \cos^2\theta)\mathbf{E}$$

The resulting polarization direction aligns with that of the electric field as the perpendicular contributions cancel out. Of course, the average value of the above expression is normally included in the linear chromatic dispersion of the model gas. The deviation is the polarization due to increased order in molecular orientation. The effective susceptibility arising from such a partial order is

$$\chi_{rot} = \rho_N \Delta\alpha [< \cos^2\theta > -1/3] \quad ,$$

where the fraction $1/3$ stands for the equilibrium value of the alignment factor. The alignment is dynamic and therefore in general different for different species constituents of the gas. Different species contributions are additive, but in general exhibit alignment revivals at different times after impulsive excitation.

The basic task in modeling the interaction with femto-second pulses on the microscopic level consists in evaluation of the mean alignment factor. At the first principle level, this is a quantum-mechanical problem which requires significant numerical effort. The method of calculation is nicely summarized in Ref. [89], and we sketch the main points next.

The evolution of the density matrix describing the state of a rotating molecule reads

$$\frac{d}{dt}\rho_{IJ} = -i\omega_{IJ}\rho_{IJ} + \frac{i}{\hbar}\sum_K (\rho_{IK}V_{KJ} - V_{IK}\rho_{KJ}) \tag{136}$$

Here, $I = (i, m)$ and other double indices denote the total and projected angular momentum quantum numbers, and the eigenfrequencies are

$$\omega_{IJ} = \frac{B}{\hbar}[i(i+1) - j(j+1)] - \frac{D}{\hbar}[i^2(i+1)^2 - j^2(j+1)^2] \tag{137}$$

with D, B rotational constants characterizing the molecule. The interaction matrix element V_{IJ} between the molecule and the external electric field includes the induced dipole moment

$$V_{IJ} = - < I|\frac{1}{2}\mathbf{E}.\bar{\alpha}.\mathbf{E}|J > \tag{138}$$

For an axially symmetric molecule this interaction can be expressed in terms of the alignment factor as follows:

$$V_{IJ} = -\frac{1}{4}E_0^2(\alpha_\perp \delta_{IJ} + \Delta\alpha < I|\cos^2\theta|J >) \tag{139}$$

where θ is the angle between the molecular and z axes. Having solved for the density matrix elements, the physical observable measuring the degree of molecular alignment is

$$\sum_{IJ} \rho_{IJ} < I|\cos^2\theta|J > \tag{140}$$

This is evaluated as a function of time for a given driving electric field $E(t)$. The solution provides a first-principle based input for the effective susceptibility of the molecular gas exposed to a strong lase pulse. However, computational requirements of the full density-matrix method are too large for effective integration with the pulse propagation simulation. This is because at every point in space the quantum mechanical problem complexity scales with the fourth power of the maximal rotational quantum number excited. The latter can be quite high if excitation is due to broadband laser pulses. Fortunately, the dynamics of the alignment can be quite accurately described by semi-classical means, which turn out to work especially well for short-duration pulses. Ref [90] derives a Green's function method and gives analytic expressions for the memory function that determines the response to a general time-dependent field in the second-order approximation in its intensity. Application in the context of ultrashort laser pulses is described in detail in [91].

In general, the response due to molecular alignment can be cast in the convolution form

$$< \cos^2\theta >= \int_{-\infty}^{t} G(t-\tau)|\mathbf{E}(\tau)|^2 d\tau \tag{141}$$

where the Green's, or memory function represents the response to an impulse excitation. Note that this theory neglects higher-order nonlinear effects in the laser-field amplitude. Several models for G have been presented, with various degrees of computational complexity (that in general increases for models with least approximations).

For molecules treated as classical rotors the Green's function is obtained as

$$G(x) = \frac{\Delta\alpha}{15k_T M}\left[x + \frac{1}{2}\pi^{1/2}(1-2x^2)e^{-x^2}erfi(x)\right] \tag{142}$$

where $x = k_T\tau$, $k_T = c^{-1}(2T/M)^{1/2}$ with M the moment of inertia, and T standing for the temperature (in energy units). This approach requires convolution evaluation, and can be further simplified by fitting the memory function by a combined response of effective oscillators. The suitable parametrization for oxygen and nitrogen is provided in [91].

One physical limitation of the above method is that the response calculation must be restricted to times shorter than a few hundred femtosecond after the initial laser pulse. At later times, the discreteness of the quantum mechanical rotor spectrum can not be ignored, and manifests itself in the well-known revivals of molecular alignment. This effect is not captured here, but the model can be generalized for situations in which probe pulses explore the excitation left in the wake of a pump laser pulse by

means of delayed-excitation of effective oscillators. The oscillator equation is driven by a delayed force originating in the first laser pulse:

$$\left[\frac{d^2}{dx^2} \pm 2\gamma\frac{d}{dx} + \omega^2\right]\chi(x) = \pm\frac{1}{2\pi}\omega^2\bar{n}_2 I_0 |E_1(x-\tau_0)|^2 \tag{143}$$

The delay τ_0 in the forcing term correspond to the molecular revival time of the given molecular species. This equation generates the susceptibility experienced by the second, probe pulse. Of course, the direct interaction of the probe with the molecular ensemble must still be included if the probe is sufficiently intense. Overall this approach provides a viable alternative to the full quantum mechanical approach at a computational expense that is reasonable for practical simulations.

2.6.4 Semi-analytic quantum model for strong-field ionization

One particularly interesting family of exactly solvable models is constructed by replacing the atomic potential function with a separable interaction. Such systems have been often used especially in nuclear physics (pioneered by Yamaguchi [92]). It was also shown that non-local separable potentials may be used to study bound states of particles in multicenter potentials [93], and hydrogenic states in quantum dots [94]. For the specific context of light-matter interaction, Refs. [95,97,96] show details and many explicit expressions that can be obtained for these exactly solvable models. The time-dependent Schrödinger equation is written in the momentum representation as

$$\left[i\partial_t - p^2/2 - \mathbf{A}(t).\mathbf{p}\right]\Psi(p,t) - \int \frac{d\mathbf{k}}{(2\pi)^3}V(\mathbf{p},\mathbf{k})\Psi(\mathbf{k},t) = 0 \quad , \quad V(\mathbf{p},\mathbf{k}) = -\sum_n^N v_n(\mathbf{p})v_n(\mathbf{k})^* \tag{144}$$

where the term on the right shows the general structure of a separable potential consisting of N components. For example, $V(\mathbf{p},\mathbf{k}) = -16\pi/(p^2+1)(k^2+1)$ results in a system that has a single bound state identical to the ground state $\psi_g(\mathbf{p}) = 8\sqrt{\pi}/(p^2+1)^2$ of the hydrogen atom. More functions $v(p)$ can be chosen as to reproduce several of the exact bound states [97]. Naturally besides the discrete spectrum, all these systems exhibit a three-dimensional energetic continuum of "free" states.

The general form of the wavefunction is $\Psi = \Psi_F + \Psi_S$, with a free-propagating and "scattering" terms. For a system with single bound state, the solution can be written explicitly as follows:

$$\Psi(\mathbf{p},t) = K_V(t,\mathbf{p})\Psi(\mathbf{p},0) + \frac{16i\pi}{(p^2+1)}\int_0^t K_V(\mathbf{p},\tau,x_{cl}(\tau))A(\tau)d\tau \tag{145}$$

with

$$K_V(\mathbf{p},t,x) = \exp\left[-ip^2t/2 - p_z x\right] \tag{146}$$

and $A(\tau)$ satisfies the integral equation

$$A(t) = A_0(t) + \int_0^t W(t,s)A(s)ds \tag{147}$$

with the right hand side A_0

$$A_0(t) = \int \frac{d\mathbf{p}}{(2\pi)^3}\frac{\Psi(\mathbf{p},0)}{p^2+1}K_V(\mathbf{p},t,x_{cl}(t)) \ , \tag{148}$$

and the kernel

$$W(t,s) = 16i\pi \int \frac{d\mathbf{p}}{(2\pi)^3} \frac{1}{(p^2+1)^2} K_V(\mathbf{p}, t-s, x_{cl}(t) - x_{cl}(s)) \tag{149}$$

As shown in [97], these expressions can be evaluated analytically. Similar calculations are possible for systems with multiple bound states [97].

Important for the application in pulse propagation simulations is that the ionization probability can be exactly evaluated for an arbitrary time-dependent excitation field, once the solution to the integral equation is known. The total ionization yield

$$P(t) = 1 - |c_0(t)|^2 \quad , \quad c_0(t) = < \psi_g | \psi(t) > \tag{150}$$

is obtained from the projection of the total wavefunction onto the ground-state. The distinct advantage is that the calculation circumvents the need for explicit wavefunction calculation.

The method based on separable potentials can, naturally, yield also the energetic spectrum of ionized electrons. Such results could serve as suitable input data for treatment of plasma-equilibration effects [99,98].

2.6.5 Integrated Maxwell-Schrödinger systems

Lorin an co-authors recently presented an ab-initio approach to Maxwell-Schrödinger integration for simulation of filamentation with intense ultrashort pulses propagating in a molecular gas [58–60]. The electromagnetic field of the laser pulse is modeled using Maxwells equations coupled with many time-dependent quantum Schrödinger systems modeling the molecular gas. The nonlinear response of model atoms thus contributes an ab initio description of the light-matter interaction. This is termed Maxwell-Schrödinger-Plasma, or MASP, model. It allows one to unify the description high harmonic generation with the nonlinear effects that govern the driving pulse propagation, namely self-focusing and de-defocusing nonlinearities. The high intensities cause the gas to become partially ionized and create a free electron plasma which also affects the pulse defocusing and losses.

It is fair to say that at time of this writing these works represent the most sophisticated first-principle based pulse propagation simulation coupled to the quantum-mechanical description of light-medium interaction. There are several interesting ingredients in this work worth of mention in some detail.

Let us start with the treatment of plasma, or free electron contribution and its effect on laser pulse propagation. A major difficulty in application of TDSE simulation to pulse propagation problems, but also in general, is that at high enough intensities the free part of the electronic wavefunction spreads rapidly, and before long it reaches the boundary of the computational domain. In order to circumvent this issue authors of Ref. [59] decided to add an evolution equation for free electrons, allowing one to model the current density which is included in the Maxwell equations [60]. To this end, absorbing boundary conditions are implemented in the TDSE simulation, resulting in a decreasing norm of the simulated electronic wavefunction. If one can assume that that the portion of the wave-function that has been annihilated at the computational box boundary is such that it would in future never interact with the parent atom again, the corresponding "leaking" probability density can be reinterpreted as representing free electrons. These free electrons are subsequently modeled as usual, along the lines of the standard filamentation model described in the previous sections. The surviving part of the electronic wavefunction continues to contribute to both self-focusing and de-focusing nonlinearity and thus affects the laser pulse propagation. For

this approximation to be consistent, the absorbing boundary must be sufficiently far from the atom or molecule being ionized, otherwise one could not neglect later interaction with the ionic potential. This means that the effective ionization is delayed, and that also the core wavefunction partially exhibits free electron properties. This approach therefore can be viewed as an alternative definition of what free and bound electronic states are. It also serves as an effective way to decrease computational requirements of the simulation.

The electromagnetic portion of the simulated system is based on Maxwell equations, with polarization and current density terms sourcing the interaction with the quantum medium of model atoms. In [60], the latter represent 1D one-electron H_2^+ molecules, and one quantum system is assigned to each macroscopic sub-domain in which the polarization is calculated. The one-dimensional TDSE is solved with the Crank-Nicolson scheme, while 3-D Maxwell equations are computed with a modified Yee scheme. Clearly, this represent a truly large scale computation and requires parallelization and significant hardware resources.

The method was illustrated for laser pulses with five to six cycles at a 800 nm wavelength with typical pulse duration of 150 fs. The total propagation length of the computational domain was up to 0.05 mm with the transverse cross section of the domain of one hundred square microns. The Maxwell-Schrödinger-Plasma system captures the basic physical ingredients that govern femtosecond filaments: As the intensity increases, longer filament forms, kept together by self-focusing action of the quantum system exposed to the optical field of the pulse. At the same time, freed electron effect show up as the intensity of the pulse remains clamped. Although these calculations assumed what is a rather small interaction zone in the context of optical filamentation, they have demonstrated all necessary physical effects that govern filaments, here unified into a single self-consistent and first-principle based description of light-matter interaction.

2.6.6 An exactly solvable model for intense light-matter interaction

Ionization in strong fields

Tolstikhin, Morishita, and Watanabe considered a one-dimensional zero-range-potential model [100], and developed an adiabatic theory of ionization by an intense laser pulse. The results are discussed here as a model suitable for large-scale simulations with spatial resolution. The system is perhaps the simplest possible one that has both the discrete and continuous energy spectrum. These are indeed the minimal necessary ingredients to model interactions that characterize femtosecond filaments with the light intensities that drive the atom or a molecule in the "no-mans-land" regime in which neither bound and/or free electronic states dominate, and both contribute to the rich dynamics. Simple models that allows for detailed analysis are precious tools in this context. The one-dimensional system studied by Tolstikhin et al. [100] has a single bound state realized by a short-range, or contact potential in the form of Dirac delta function, and it is exposed to an external electric field $F(t)$ with an arbitrary time dependence. The time-dependent Schrödinger equation can be symbolically written as

$$\left[i\partial_t + \frac{1}{2}\partial_x^2 + B\delta(x) - xF(t)\right]\psi(x,t) = 0 \quad . \tag{151}$$

The theory is developed for a small dimensionless parameter $\epsilon = 1/(B^2 T_0)$ where T_0 stands for characteristic time-scale of the external field. Authors construct an

asymptotic solution for $\epsilon \to 0$ and calculate the photo-ionization probability and freed-electron momentum spectra. Such results can be in principle combined with the pulse propagation solvers, and serve as a first-principle based source of freed electrons.

It has to be noted that this particular model has been studied in many other contexts, and utilized as a testbed for various physical systems. Recently, the exact solvability of this system was utilized to gain insight into possible manifestation of higher-order nonlinearity in strong optical fields [101]. The same authors also show that an exact solution for the complete nonlinear response can be obtained for an arbitrary time dependent external field [102]. That solution provides a useful tool for simulations of laser pulse interactions with atomic gases — it is described next.

Nonlinear response for arbitrary time-dependent field

The structure of the exact solution is quite similar to one that occurs in the exactly solvable system with separable interaction discussed earlier [95,97]. Namely, the wave function consists of the free-propagating and re-scattered components:

$$\psi(x,t) = \psi_F(x,t) + \psi_S(x,t) \tag{152}$$

$$\psi_F(x,t) = \int dx'\, K_F(x,t|x',0)\psi_G(x') \tag{153}$$

$$\psi_S(x,t) = iB \int_0^t dt'\, K_F(x,t|0,t')\psi(0,t') \tag{154}$$

$$\psi_G(x) = B^{1/2} e^{-B|x|}\ . \tag{155}$$

where the central quantity is the Volkov propagator that describes the system's evolution in an arbitrary time dependent field, but in absence of the atomic potential:

$$K_F(x,t|x',t') = e^{i\phi(x,t,x',t')} K_0(x - x_{cl}(t), t|x' - x_{cl}(t'), t') \tag{156}$$

$$K_0(x,t|x',t') = \frac{1}{\sqrt{2\pi i(t-t')}} e^{-\frac{(x-x')^2}{2i(t-t')}} \tag{157}$$

$$\phi(x,t,x',t') = xp_{cl}(t) - x'p_{cl}(t') - [S_{cl}(t) - S_{cl}(t')] \tag{158}$$

here, the quantities $p_{cl}(t), x_{cl}(t), S_{cl}(t)$ denote the classical particle momentum, position and kinetic energy as imposed by the external field.

To integrate this system with a laser pulse simulator, the solution for either nonlinear polarization or current density is desired, and both can be obtained in semi-closed forms. The current density, for example, reads

$$J(x,t) = \sum_{i,j=S,F} J_{ij} = \mathrm{Im}\left\{\psi_F^* \nabla\psi_F + \psi_F^* \nabla\psi_S + \psi_S^* \nabla\psi_F + \psi_S^* \nabla\psi_S\right\} \tag{159}$$

where the nonlinear components of the four contributions are

$$J_{SS}^{(nl)} = 2\mathrm{Im}\{\int_0^t dt_1 \int_0^{t_1} dt_2 \frac{(-i)^{\frac{3}{2}} B^3 W(t_1,t_2)}{\sqrt{2\pi}} \times \left[e^{\frac{i[x_{cl}(t_1) - x_{cl}(t_2)]^2}{2(t_1-t_2)}} A^*(t_1)A(t_2) - 1\right] \frac{x_{cl}(t_1) - x_{cl}(t_2)}{t_1 - t_2}\ \}$$

$$J_{FS} = \mathrm{Im}\{iB^3 \int_0^t dt_1 A^*(t_1)[e^{+Bx_{cl}(t_1)} \mathrm{erfc}\left(\frac{(1+i)(Bt_1 - ix_{cl}(t_1))}{2\sqrt{t_1}}\right) - e^{-Bx_{cl}(t_1)} \mathrm{erfc}\left(\frac{(1+i)(Bt_1 + ix_{cl}(t_1))}{2\sqrt{t_1}}\right)]\}$$

$$J_{FS}^{(nl)} = J_{FS} - \mathrm{Im}\left\{2B^3 \int_0^t dt_1 x_{cl}(t_1) \left(iB\ \mathrm{erfc}\left(\frac{(1+i)B\sqrt{t_1}}{2}\right) - \frac{1+i}{\sqrt{\pi t_1}} e^{-i\frac{B^2}{2}t_1}\right)\right\} \tag{160}$$

and are expressed in terms of a central quantity $A(t)$ which solves the following integral equation

$$A(t) = \psi_R(-x_{cl}(t), t) + \frac{iB}{\sqrt{2\pi i}} \int_0^t dt' \frac{e^{+i\frac{B^2}{2}(t'-t)}}{\sqrt{t-t'}} \exp\left[\frac{i(x_{cl}(t) - x_{cl}(t'))^2}{2(t-t')}\right] A(t') \quad (161)$$

with the first term on the right-hand-side being the free-propagated initial condition

$$\psi_R(x,t) \equiv \frac{e^{+Bx}}{2} \mathrm{erfc}\left(\frac{iBt+x}{\sqrt{2it}}\right) + \frac{e^{-Bx}}{2} \mathrm{erfc}\left(\frac{iBt-x}{\sqrt{2it}}\right). \quad (162)$$

A numerical algorithm with a complexity that scales with the square of the number of sampling points on the temporal axis is described in detail in ref. [102]. The numerical evaluation for the driving wave-forms $F(t)$ typical of femtosecond filamentation and/or harmonic generation in hollow waveguides is sufficiently efficient to allow fully spatially resolved experiment modeling — an illustration is shown next.

The above described model has been used for investigations into the dynamics of light-matter interaction in various regimes. Despite the simplicity if the model quantum system, it serves as a useful laboratory and a tool to understand the physics in highly dynamic regimes that do not lend themselves to easy intuitive insights. The most important advantage of the above solution is that it unifies several aspects that have been so far modeled as different and independent facets of the standard light-matter interaction model; The exact nonlinear response of this system, when exposed to a few-cycle infrared wavelength laser pulse exhibits the self-focusing Kerr effect, third-harmonic generation, high-harmonic generation, resonant state excitation, and freed-electron induced current, all in a single quantity, the polarization or current, that serves as a source in the Maxwell equations.

2.7 High-harmonic generation and propagation effects

Propagation effects are in general of great importance in modeling high-harmonic generation. This is easy to understand intuitively, once we look at the polarization response of the medium that enters Maxwell equations, and realize how broad the frequency bandwidth is, and how large is the dynamic range of the spectrum. It is the propagation mechanisms that *select* [103] which components of the polarization response spectrum that will actually be converted into radiation. Because of complex phase-matching issues, the spectrum of high-harmonic radiation in general only represents a "subset" of the spectral content found in the induced dipole moment spectrum we describe next. For detailed computing and modeling aspects of the following section, a very nice overview is given in Ref. [103] by Gaarde and co-authors.

The usual scheme utilized in a comprehensive HHG model is based on separation between the driver pulse field and the high-harmonic radiation generated by the former. Moreover, while the former is in general strongly affected by the underlying nonlinearities, the latter can be safely considered a linear problem with a source. Typically, field propagation equations are written in the mixed representation, and treat diffractive effects in real space, while the temporal domain is handled in the Fourier space (we write these equations in the lab frame)

$$\begin{aligned}\partial_z E_D(x,y,\omega,z) = {} & ik(\omega) E_D(x,y,\omega,z) + \frac{i}{2k(\omega)} \Delta_\perp E_D(x,y,\omega,z) \\ & + \frac{i\mu_0 \omega c}{2n_b(\omega)} P_{Driver}(x,y,\omega,z) \end{aligned} \quad (163)$$

$$\partial_z E_H(x,y,\omega,z) = ik(\omega)E_H(x,y,\omega,z) + \frac{i}{2k(\omega)}\Delta_\perp E_H(x,y,\omega,z) + \frac{i\mu_0\omega c}{2n_b(\omega)}P_{Dipole}(\{E_D\},\omega,z) \quad (164)$$

Here E_D and E_H are the infrared driver pulse and harmonic fields, respectively. The first equation is essentially equivalent to what is normally utilized in the filamentation modeling, with the quantitative difference being that in most regimes relevant for HHG, it is the ionization and freed-electron de-focusing that plays the most important role in the dynamics of the driver pulse. In the second equation, the P_{Dipole} stands for the dipole moment density (polarization) that originates in the atoms driven by the strong field E_D. E_H does not enter here, because the intensity of harmonic radiations in orders of magnitude lower than that of the driver pulse. Thus, the evolution equation for harmonics is linear in nature, which simplifies its solution. In particular, frequency-space sampling can be independent in the two equations, and the latter may only be solved for the frequency range of interest. However, we will see shortly, that this does eliminate the need for very fine resolution of the temporal grid at which the atomic response is calculated. This calculation we describe next.

2.7.1 Strong field approximation

Strong field approximation has been a workhorse in the computational HHG modeling field. The origin of the algorithm discussed in this section goes back to the classical three-step model proposed by Corkum. The mathematical basis for the quantum (actually semi-classical) treatment was put forward by M. Lewenstein in [104].

The dipole term sourcing the propagation equation for the harmonic field E_H is given as

$$P_H(\omega) = FT[N_{atom}(t)X_{nl}(t)] \quad (165)$$

where FT denotes a Fourier transform from t to ω, and $X_{nl}(t)$ is the expectation value for the nonlinearly induced dipole moment of an atom exposed to the driver field $E_D(t)$. $N_{atom}(t)$ is the time-dependent density of remaining (i.e. so far not ionized) neutrals. The latter quantity is normally obtained in the course of solution for the driver field $E_D(t)$ which is coupled to the freed-electron density equation in the way described in the previous Section.

The expression for the dipole moment $X_{nl}(t)$ is given in the form of an integral which can be interpreted as a sum of contributions from different electron trajectories that start with the atom ionization, continue as free particles driven by the field $E_D(t)$, and return to the parent ion where they finally recombine. This motion contributes to the total dipole moment source seen by the propagation equation for the harmonic field. In atomic units, the contribution to the dipole moment from these trajectories is

$$X_{nl}(t) = 2Re\{i\int_{-\infty}^{t} dt' \left(\frac{\pi}{\epsilon + i(t-t')/2}\right)^{3/2} e^{-iS_{st}(t',t)} d^*[p_{st}(t',t) + A(t)]d[p_{st}(t',t) + A(t')]E_D(t')\} \quad (166)$$

Here $A(t)$ is the vector potential of the driving field, and S_{st} and p_{st} are stationary values for the classical electron momentum

$$p_{st}(t',t) = \frac{1}{t'-t}\int_t^{\prime t} A(\tau)d\tau \quad (167)$$

and classical action (which is just the kinetic energy as the electron is considered free)

$$S_{st}(t',t) = (t-t')(I_p - p_{st}^2/2) + \frac{1}{2}\int_t^{'t} A^2(\tau)d\tau \tag{168}$$

with I_p representing the atom ionization potential. The properties of the atom enter through the function $d[k]$ which is the dipole matrix element between the ground state and a plane wave with momentum k.

The above equations represent the most computationally intensive part of a HHG simulation. Various tricks and approximation are used in practice to lessen this burden. The most important one addresses the fact that the computational complexity of the above scales as the square of number of grid point in the time axis. Note that the latter must span the duration of the infrared driver pulse which is potentially many hundreds of atomic units of time. This extent must be sampled with a grid resolving a fraction of the atomic time unit, with longer driver wavelength requiring finer resolutions. The cost of evaluation in (166) can thus easily become prohibitive, especially when coupled with a spatially resolved driver simulation which in turn require to solve the above system at each spatial grid point. This may be addressed by restricting the double integral to $t - t'$ being shorter then a couple of optical cycles of the driver pulse. The rationale is that the main contribution to the integral comes from the electron trajectories that originate and end in the near past.

The following sections illustrate application of these techniques to practical modeling of experiments in the field of high-harmonic generation.

2.7.2 High-harmonic generation in a filamentation regime

I this subsection, we concentrate on the propagation aspect in generation of high harmonic radiation in high-intensity optical pulses. The interest from the modeling standpoint is in the role that the propagation effects play in the situations that lead to extreme frequencies being generated "spontaneously" in natural filamentary structures that occur as a result of spectral and spatio-temporal reshaping of the driving optical pulse. This is a field in which simulation plays a crucial role. Not only it serves as a tool to understand and interpret experiments, but it can predict and identify potentially interesting regimes and thus motivate further experimental investigations.

High-harmonic generation can be realized in many different ways, depending mainly on the employed geometry. Two most popular arrangements are utilizing gas jets and hollow waveguides. In the former, the nonlinear interaction with a noble gas occurs within a a short propagation distance that is restricted by the thickness of the jet. Yet, propagation effects manifest in phase-matching effects. On the other hand, reshaping of the driving pulse is relatively unimportant because of the short propagation distance within the gas. In the other arrangement, laser pulses interact with the gas inside a capillary waveguide over several centimeters, and the propagation effects are naturally more important. A novel regime of high-harmonic generation was discovered recently in highly pressurized capillary waveguide [56] filled with various gases. Computer simulations (based on the Nonlinear Envelope Equation [16]) suggest that the unusual regime is very much similar to that of femtosecond filamentation in bulk media and especially in gases. Naturally, under such condition propagation effects are crucial, and computer modeling provides important insights.

Simulations show that as the pressure increases to a few tens of atmospheres, the intensity profile inside capillary becomes significantly smoother, which in turn indicates that a relatively stable propagation regime is achieved. It is believed that

the latter is what underlines high-harmonic generation. Parameters of the dynamics appear very much resembling a free-space filamentation regime as it is known at shorter wavelengths.

Perhaps most important are propagation effects when high harmonic radiation is generated directly from a femtosecond filament [105]. That such a regime can indeed result in strong HH radiation as has been demonstrated in Ref. [106].

Numerical methods employed in modeling this kind of experiment were described in [105,107]. For each propagation distance, time-dependent laser electric field is calculated by Fourier transform from its native spectral representation, and is subsequently used to calculate the (time-dependent) nonlinear response terms. Then, Fourier transform takes the source terms back to the frequency domain where they are used to propagate the laser and harmonic frequencies to the next step in the propagation direction. For the driving laser field, the nonlinear terms include the third-order response, and the ionization terms that are evaluated using intensity-dependent ionization rates calculated, as described in [108]. This technique is essentially that described in the section on spectral pulse propagation solvers. The ionization model includes nonlinear absorption and concomitant losses to the optical field due to multiphoton ionization, and the ionization-driven plasma refractive index. For the harmonic radiation field the driving (source) term is given by the time-dependent dipole moment, calculated using the strong field approximation [104], multiplied by the atomic density. Absorption (for frequencies above the ionization threshold) and linear dispersion are treated with frequency-dependent coefficients adopted from [109].

Generation of high harmonic radiation directly form the filament, as the simulations showed, is mediated by the occurrence of high-intensity spikes. Intensity of these features can be significantly higher that that of the typical filament core. In a sense they can be regarded as extreme events that temporarily invalidate the notion of intensity clamping [110] that is believed to underlie much of the optical filament physics.

This behavior, predicted in an earlier work [107], reveals that the maximal intensity vs distance exhibits localized spikes during which the clamping argument does not apply because of their very short temporal duration. Indeed, as the inset illustrates, these spikes are carried by nearly single-cycle waveforms that form dynamically in the trailing portion of the pulse.

It was for the first time in this work that a coupled computer simulation was done simultaneously for both the driving pulse and the high-harmonic radiation. The simulations showed that very intense, and extremely short sub-pulses arise with peak intensities exceeding the intra-filament clamping intensity by a factor of three. These sub-pulses form in the laser pulse when its trailing end is refocused onto the axis. It is these intensity spikes that give rise to intense, isolated attosecond pulses which could be coupled out of the filament.

3 Implementation of propagation models

This section is devoted to a presentation of the numerical implementation of propagation equations. As shown in the previous section, a canonical form exists for all unidirectional propagation equations. This form has the same structure for envelope and for carrier-resolving equations, opening the question of the possibility for a universal scheme and solver. A numerical scheme valid for the UPPE should indeed apply for solving all other equations. However, there are a few distinguishing features in the propagation equations in the canonical form making worth presenting different schemes. First, carrier-resolving propagation equations involve real fields as in Maxwell equations, whereas envelope equations deal with complex envelopes. This implies slight differences in treating the spectra of real fields compared to complex envelope spectra. Second, Nonparaxiality is one of the distinguishing features of the UPPE, making necessary to solve that equation in the three-dimensional spectral domain for frequency and wave numbers, whereas alternatives exist for paraxial equations, be they propagating envelopes or fields. At the price of a loss of universality, paraxial equations can indeed be solved by finite difference methods in the spatial domain, by space marching each frequency component after a one-dimensional temporal to spectral Fourier transform. We adopt a presentation covering both options with a first section devoted, but not restricted to paraxial envelope equations. We will start by the simplest propagation equation which describes diffraction of a laser beam as this provides the basic building block of the general scheme valid for all paraxial propagation equations. We then consider resolution of the UPPE in Fourier space, which apply in general to nonparaxial, carrier-resolving or envelope, propagation equations.

The generic form of the considered propagation equations is retrieved in models for different physical problems, therefore the methods we describe apply as well in different fields. The reader is refferred to [36] and reference therein for a review of numerical schemes we apply to propagation equations.

3.1 Envelope propagation models

In this section, we consider paraxial envelope equations. The proposed implementation methods extend in a straightforward way to paraxial carrier-resolving equations, thus without loss of generality, we restrict the presentation to complex envelopes.

3.1.1 Diffraction

Diffraction occurs in all media and even in vacuum. Not surprisingly, the structure of the diffraction operator as a product of a frequency dependent coefficient and a transverse Laplacian acting only on transverse coordinates is the common feature of all paraxial envelope or carrier-resolving propagation equations. Implementing diffraction therefore serves as a basic building block for all paraxial equations. We first consider monochromatic beams, i.e. a sufficiently long laser pulse with central wave number k_0 and a narrow spectrum so that all frequency dependence can be neglected. Diffraction of the monochromatic beam is described by the paraxial equation

$$\frac{\partial \mathcal{E}}{\partial z} = \frac{i}{2k_0}\Delta_\perp \mathcal{E} \tag{169}$$

We start by describing beam propagation in 1+1 dimensions, i.e, one transverse dimension and one evolution (or propagation) variable z. We will consider either a

planar geometry with transverse direction x, or a cylindrical geometry with revolution symmetry, with a radial transverse variable $r \equiv \sqrt{x^2+y^2}$. From the point of view of the computational cost, the revolution symmetry is an interesting geometry for propagating a laser beam when it does not break-up into multiple beamlets since its cost corresponds to that of a 1-dimensional numerical method. In this section $\mathcal{E}$ is therefore assumed to depend only on the transverse variable x or r, and the evolution variable z. The transverse Laplacian operator is therefore reduced to $\Delta_\perp \equiv \partial^2/\partial x^2$ in the planar geometry, or to $\Delta_\perp \equiv \partial^2/\partial r^2 + (1/r)\partial/\partial r$ in cylindrical geometry. To avoid redundancy, we will use r as a transverse variable but switching from cylindrical to planar geometry can be implemented in a single generic tool: Unless otherwise stated, all formulas are the same for both geometries with $r \leftrightarrow x$. We will specify minor differences between planar and cylindrical geometry by using x instead of r only where it is necessary.

Initial condition The propagation starts at $z = 0$ where the beam amplitude and phase profiles are known, e.g., the beam has a Gaussian shape with quadratic spatial phase modeling a flat-phase beam having passed through a lens of focal length f:

$$\mathcal{E}(r, z=0) = \mathcal{E}_0 \exp\left(-\frac{r^2}{w_0^2} - i\frac{k_0 r^2}{2f}\right). \tag{170}$$

The quantities w_0 and $\mathcal{E}_0$ are the beam width and the initial amplitude.

Boundary conditions Boundary conditions must also be specified to solve Equation (169). In free space, one usually requires that the field vanish far from the peak. The highest order derivative in Equation (169) is second order, thus two boundary conditions must be specified at the boundaries of the numerical grid $r = r_{\min}$ and $r = r_{\max}$. These depend on the type of beam one wants to model.

In a cylindrical geometry, standard beams have intensity with a zero-slope at the origin and vanishes far from the origin, which gives the boundary conditions:

$$\left.\frac{\partial \mathcal{E}(r,z)}{\partial r}\right|_{r=0} = 0, \tag{171}$$

$$\mathcal{E}(r = r_{\max}, z) = 0 \tag{172}$$

In a planar geometry, the boundary conditions for a standard beam vanishing far from the peak read:

$$\mathcal{E}(x = x_{min}, z) = 0, \tag{173}$$

$$\mathcal{E}(x = x_{max}, z) = 0 \tag{174}$$

Boundary conditions (173) and (174) in a cylindrical grid would be suitable for the propagation of vortex beams (having a zero field and phase singularity at the origin).

On finite difference schemes Finite difference scheme consist in representing the solution to the original PDE by its values at discrete set of points and replacing the PDE by a set of coupled equations for these discrete quantities. In practice, we discretize the transverse variable r to form a numerical grid of finite size. For a uniform grid with $N_\perp + 2$ grid points, i.e., $N_\perp$ inner points and 2 boundaries. We define $r_j = r_{\min} + j\Delta r$, $j = 0 \ldots N_\perp + 1$ and the step-size $\Delta r = (r_{\max} - r_{\min})/(N_\perp + 1)$.

For a cylindrical geometry $r_{\min} = 0$. Similarly to the discretization of the transverse variable, we will describe the propagation over a distance z_{max} by making steps $z_n = n\Delta z$, $n = 0 \dots N_z$ of constant size Δz. Let E_j^n denote $\mathcal{E}(r = r_j, z = z_n)$.

A standard task of numerical analysis is to design robust algorithms, which have desirable properties including good *numerical stability*, *accuracy*, and *efficiency* with respect to computational time.

Discretization of PDEs is associated with local and global truncation errors due to the approximation of partial derivatives by their discretized versions. For example in Eq. (169), the evolution operator is discretized as:

$$\frac{\partial \mathcal{E}}{\partial z}(r = r_j, z = z_n) \simeq \frac{E_j^{n+1} - E_j^n}{\Delta z} + O(\Delta z), \tag{175}$$

which is first-order accurate in Δz. A second-order accurate discretization of the second-order space-derivative appearing in the transverse Laplacian reads:

$$\frac{\partial^2 \mathcal{E}}{\partial r^2}(r = r_j, z = z_n) \simeq \frac{E_{j+1}^n - 2E_j^n + E_{j-1}^n}{\Delta r^2} + O(\Delta r^2). \tag{176}$$

The *local truncation error* in the numerical solution is the error generated at a particular step, when the solution at the previous step is considered as exact (in practice, it is not exact, except if it corresponds to the initial condition). For example Eq. (176) gives a second order local truncation error. The cumulative error in the numerical solution to a PDE on an interval in the evolution variable is called the global truncation error and the order of accuracy is the order of the global truncation error.

Numerical stability refers to the fact that a numerical calculation does not amplify truncation or approximation errors. If approximation errors decay as the computation is carried forward, the numerical scheme is stable. If the errors grow, the numerical solution departs from the correct physical behavior of the modeled system and the numerical scheme is said to be unstable. The stability of finite difference schemes applied to linear partial differential equations can be commonly determined by a von Neumann stability analysis [37,38] which is based on the decomposition of the solution including the instability waves representing errors into Fourier series and a linear stability analysis of these waves. Depending on the numerical scheme, stability can require restrictive conditions on the step-sizes to be fulfilled. Scheme stability is in general difficult to investigate when the partial differential equations under consideration are nonlinear or nonuniform. Therefore, stability conditions on step-sizes associated with the linear-part of our canonical propagation equation may not be sufficient to ensure the stability of the complete scheme but constitute a good starting guess of the restrictions on the step sizes used in the scheme.

Implicit vs *explicit* schemes: Finite difference schemes are furthermore classified into *explicit* and *implicit* schemes: An explicit scheme allows for the calculation of quantities at each position J on the grid for the evolution variable $N + 1$, say E_J^{N+1}, explicitly from the previously known quantities E_j^n, $n = 0 \dots N$, $j = 0 \dots N_\perp + 1$. An implicit scheme requires the numerical resolution of implicit equations coupling E_j^{N+1} for several j with the already known quantities in order to find E_j^{N+1} for all j, and are computationally more expensive. However, the efficiency of a calculation must be evaluated with stability constraints in mind. Explicit finite difference schemes for solving Eq. (169) are associated with very restrictive stability conditions in the form $\Delta z \ll k_0 \Delta r^2$, ruining the advantage of an explicit scheme [38]. We leave it to the reader to analyze the details in Ref. [38] and we will directly present a resolution method based on an implicit method, namely the Crank-Nicolson method [39], the computational cost of which is balanced by the fact that it is unconditionally stable

and allows us to replace many small explicit steps by a single large implicit step to advance the solution.

Crank-Nicolson method A standard and efficient scheme to perform numerical simulations of Eq. (169) and equations having the same structure, as e.g. Eq. (23), is the Crank-Nicolson scheme [39]. It is an implicit, unconditionally stable numerical scheme that is second order accurate in both Δr and Δz.

Below, Δ_j denotes the discretized diffraction operator defined as

$$\Delta_j E_j^n \equiv E_{j-1}^n - 2E_j^n + E_{j+1}^n + \frac{\nu}{2j}(E_{j+1}^n - E_{j-1}^n), \tag{177}$$

where $\nu = 0$ for planar geometry and $\nu = 1$ for cylindrical geometry. Δ_j can be formally represented as a tridiagonal matrix acting on the vector E_j^n, with $j = 0 \dots N_\perp + 1$.

$$\Delta_j E_j^n = \begin{pmatrix} -2 & v_0 & 0 & \cdots & \cdots & 0 & 0 \\ u_1 & -2 & v_1 & 0 & \cdots & 0 & 0 \\ 0 & \ddots & \ddots & \ddots & 0 & 0 & 0 \\ 0 & 0 & u_j & -2 & v_j & 0 & 0 \\ 0 & 0 & 0 & \ddots & \ddots & \ddots & 0 \\ 0 & \cdots & \cdots & 0 & u_{N_\perp} & -2 & v_{N_\perp} \\ 0 & \cdots & \cdots & \cdots & 0 & u_{N_\perp+1} & -2 \end{pmatrix} \begin{pmatrix} E_0^n \\ E_1^n \\ \vdots \\ E_j^n \\ \vdots \\ E_{N_\perp}^n \\ E_{N_\perp+1}^n \end{pmatrix} \tag{178}$$

where $u_j = 1 - \nu/2j$ and $v_j = 1 + \nu/2j$. At this stage, note that u_0 and v_0 are ill-defined for a cylindrical geometry. In fact, the coefficients in the first and last lines of the matrix Δ_j will be replaced later to take boundary conditions into account. We therefore assume $v_0 = 1$ for both planar and cylindrical geometries.

The Crank-Nicolson scheme consists in discretizing the evolution operator in Eq. (169) as $\partial_z \mathcal{E}(r = r_j, z = z_n) = (E_j^{n+1} - E_j^n)/\Delta z$, thus it is centered at step $n + 1/2$. On the right hand side of Eq. (169), the diffraction operator is discretized and applied to the average of the field at distance n and $n + 1$ so as to center the scheme at step $n + 1/2$. This yields the implicit equation:

$$E_j^{n+1} - E_j^n = i\delta(\Delta_j E_j^{n+1} + \Delta_j E_j^n) \tag{179}$$

where $\delta = \Delta z/4k_0(\Delta r)^2$. The solution of Eq. (179) formally reads as

$$E_j^{n+1} = (1 - i\delta\Delta_j)^{-1}(1 + i\delta\Delta_j)E_j^n, \tag{180}$$

or equivalently:

$$L_- E_j^{n+1} = L_+ E_j^n \tag{181}$$

and requires multiplication of the tridiagonal complex matrix $L_+ \equiv 1 + i\delta\Delta_j$ by the vector E_j^n, inversion of the tridiagonal complex matrix $L_- \equiv 1 - i\delta\Delta_j$ and multiplication of L_-^{-1} by $L_+ E_j^n$. The above definition of matrix L_+ and L_- must be modified to take into account the boundary conditions. Equation (180) allows for space marching the field over one propagation step. Applying it repeatedly for N_z steps will thus propagate the input field $E_j^{n=0}$ over a distance $N_z \Delta z$, where $E_j^{n=N_z}$ is obtained. Without modifications, the first line ($j = 0$) of Equation (180) reads as $(1+2i\delta)E_0^1 - i\delta v_0 E_1^1 = (1-2i\delta)E_0^0 + i\delta v_0 E_1^0$, which does not match the discretized version of the boundary condition (171): $E_0^1 = 0$, or that of (171). Boundary conditions

can be simply enforced by rewriting this first line of L^- as $(1, 0, 0, \ldots, 0)$ for the planar geometry, or as $(1, -1, 0, \ldots, 0)$ for the circular geometry, and the first line of L^+ as $(0, \ldots, 0)$ in both cases. In circular geometry, this ensures that a first-order-accurate discretized version of Eq. (171) is satisfied: $(E_1^1 - E_0^1)/\Delta r = 0$. An implementation of the boundary condition should at least match the order of the inner scheme. Introducing a ghost value E_{-1} at r=$-\Delta r$ and a second-order-accurate version of Eq. (171): $(E_1^1 - E_{-1}^1)/2\Delta r = 0$ together with the discretization introduced in the scheme for inner grid points (180) allows us to eliminate the ghost value and preserve the overall second order accuracy of the scheme. The first lines of $L^{\pm}$ are obtained by identifying the coefficients of: $(1 + 4i\delta)E_0^1 - 4i\delta E_1^1 = (1 - 4i\delta)E_0^0 + 4i\delta E_1^0$. The last lines of L^- and L^+ are treated similarly to enforce boundary condition (172). Thus, the complex matrix L^- and L^+ including proper treatment of boundary conditions reads:

$$L_+E_j^n = \begin{pmatrix} d_{0,+}^{(o)} & d_{1,+}^{(o)} & 0 & 0 & 0 & 0 & 0 \\ i\delta u_1 & 1-2i\delta & i\delta v_1 & 0 & 0 & 0 & 0 \\ 0 & \ddots & \ddots & \ddots & 0 & 0 & 0 \\ 0 & 0 & i\delta u_j & 1-2i\delta & i\delta v_j & 0 & 0 \\ 0 & 0 & 0 & \ddots & \ddots & \ddots & 0 \\ 0 & 0 & 0 & 0 & i\delta u_{N_\perp} & 1-2i\delta & i\delta v_{N_\perp} \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} E_0^n \\ E_1^n \\ \vdots \\ E_j^n \\ \vdots \\ E_{N_\perp}^n \\ E_{N_\perp+1}^n \end{pmatrix} \tag{182}$$

$$L_-E_j^n = \begin{pmatrix} d_{0,-}^{(o)} & d_{0,-}^{(o)} & 0 & 0 & 0 & 0 & 0 \\ -i\delta u_1 & 1+2i\delta & -i\delta v_1 & 0 & 0 & 0 & 0 \\ 0 & \ddots & \ddots & \ddots & 0 & 0 & 0 \\ 0 & 0 & -i\delta u_j & 1+2i\delta & -i\delta v_j & 0 & 0 \\ 0 & 0 & 0 & \ddots & \ddots & \ddots & 0 \\ 0 & 0 & 0 & 0 & -i\delta u_{N_\perp} & 1+2i\delta & -i\delta v_{N_\perp} \\ 0 & 0 & 0 & 0 & 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} E_0^n \\ E_1^n \\ \vdots \\ E_j^n \\ \vdots \\ E_{N_\perp}^n \\ E_{N_\perp+1}^n \end{pmatrix} \tag{183}$$

where first order accurate boundary conditions ($o = 1$) at the origin are enforced by:

$$d_{0,+}^{(1)} = 0, \qquad d_{1,+}^{(1)} = 0, \tag{184}$$

$$d_{0,-}^{(1)} = 1, \qquad d_{1,-}^{(1)} = -\nu, \tag{185}$$

and ν is unity for cylindrical geometry or zero for the planar case. For second order accurate boundary conditions ($o = 2$) at the origin:

$$d_{0,+}^{(2)} = 1 - 4i\delta, \qquad d_{1,+}^{(2)} = 4i\delta, \tag{186}$$

$$d_{0,-}^{(2)} = 1 + 4i\delta, \qquad d_{1,-}^{(1)} = -4i\delta. \tag{187}$$

Different type of boundary conditions can be implemented in a similar way, and potentially involve more than a single line of the complex matrix L^+ and L^- for each boundary. For example, it may be needed to add boundary layers on one or several edges of the numerical box so as to avoid spurious reflection on the boundary in cases where free-space propagation is desired. The goal of the boundary layer is to mimic a physical process over a limited domain close to the boundary, that will prevent as much as possible incoming waves to be reflected. This can be achieved with absorption of the incoming waves, or diffusion. With a careful selection of the boundary layer

Table 3. Crank-Nicolson Algorithm

- 1. definition of useful data, e.g., beam width w_0, focusing length f, laser wavelength λ_0, index of refraction n_0 and central wave number $k_0 = n_0 2\pi/\lambda_0$.
- 2. definition of grids and z-invariant quantities for space marching the field over one step Eq. (180).
  - number of inner grid points $N_\perp$
  - r-grid: $r_j = r_{\min} + j\Delta r$, for $j = 0, \cdots, N_\perp + 1$
  - matrix L_+ and L_-: $L_\pm = 1 \pm i\delta\Delta_j$
  - boundary conditions: replace first and last lines of $L^\pm$ as indicated in Eqs. (182) and (183)
  - calculation of the inverse L_-^{-1}
  - calculation of the product $L = L_-^{-1} L_+$
- 3. definition of the initial field $E_j^0 = \mathcal{E}(r_j, z = 0)$, $j = 0 \ldots N_\perp + 1$, e.g., as in Eq. (170) for a Gaussian beam
- 4. perform a loop for $N_z = M \times K_{max}$ propagation steps, with an outer loop including computationally expensive diagnostics each M steps and an inner loop including costless diagnostics performed each step:

```
outer loop: k = 1, ..., K_max
    inner loop: m = 1, ..., M
        n = (k − 1)M + m
        E_j^n = L E_j^{n−1}
        perform costless diagnostic n
    end inner loop
    perform expensive diagnostic k
end outer loop
```

features, the latter choice was shown to lead theoretically to no reflection and was called *perfectly matched layers* boundary conditions [40].

We can now detail in table 3 the different steps to build a simple propagation code for simulations of Eq. (169):

Steps 1 to 3 correspond to the initialization of the propagation. Step 4 constitutes the bulk of the scheme; it is centered around one propagation step following Eq. (180). By diagnostics, we mean the selection of a given set of computed data and their storage in external files for post-processing. In order to save computational time and memory, diagnostics can be separated into computationally expensive diagnostics and costless diagnostics. Costless diagnostics do not require long CPU time or large disk-memory and can thus be performed at each step without significant performance degradation. Typically, these concern sub-dimensional diagnostics such as, e.g., monitoring of the maximum intensity vs propagation distance. In contrast, expensive diagnostics require more computer resources, time or memory, such as monitoring the full spatial, temporal or spectral- beam or pulse dynamics and not only subdimensional slices. A proper balance between expensive and costless diagnostics must be ensured for a good efficiency of the whole simulation.

Spectral technique We present in this section one of the most straightforward way to implement a numerical scheme for solving Eq. (169). It consists in a spectral method relying on a Fourier decomposition of the laser beam into its spectral components. We assume here a planar geometry and note that the method extends easily to the

cylindrical geometry by replacing Fourier by Hankel transforms:

$$\tilde{\mathcal{E}}(k_x, z) = \int_{-\infty}^{+\infty} \mathcal{E}(x, z) \exp(-ik_x x) dx \tag{188}$$

Applying this transformation to Eq. (169) leads to a simple set of ordinary differential equations for the spectral components of the beam envelope $\tilde{\mathcal{E}}(k_x, z)$:

$$\frac{\partial \tilde{\mathcal{E}}(k_x, z)}{\partial z} = -i \frac{k_x^2}{2k_0} \tilde{\mathcal{E}}(k_x, z) \tag{189}$$

which has a formal solution:

$$\tilde{\mathcal{E}}(k_x, z) = \tilde{\mathcal{E}}(k_x, z = 0) \times \exp\left(-i \frac{k_x^2}{2k_0} z\right) \tag{190}$$

From Eq. (190), the solution obtained by back transforming the spectral components into the spatial domain reads

$$\mathcal{E}(x, z) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \tilde{\mathcal{E}}(k_x, z = 0) \times \exp\left(-i \frac{k_x^2}{2k_0} z + ik_x x\right) dk_x \tag{191}$$

The numerical implementation of this is made straightforward by the availability of libraries including fast Fourier transform modules. We can now detail in Table 4 the different steps to simulate propagation governed by Eq. (169) with a Spectral Decomposition Algorithm: Boundary conditions are assumed to be periodic and automatically enforced by the Fourier decomposition, i.e., by the Fast Fourier Transforms performed at each step. In practice, this means that a sufficiently large spatial box must be chosen if this method is used to simulate free space propagation with fields exponentially decaying far from the peak. In a too small box, a beam would sooner or later hit a boundary and be artificially reintroduced at the opposite boundary.

Naturally Eq. (191) shows that the solution at an arbitrarily large propagation distance can be obtained in a single step from the input spectral components $\tilde{\mathcal{E}}(k_x, z = 0)$ to the final far-field $\tilde{\mathcal{E}}(k_x, z)$, without iteratively space marching the solution over N_z propagation steps of size z/N_z. This option is however restricted to linear propagation equations. Multiple steps as indicated in table 4 become necessary when nonlinear terms are added on the right hand side of Eq. (169), which is the reason for having specified them in the scheme. Due to the availability of Fast Fourier Transform routines in computational libraries and in spite of the imposed periodic boundary conditions, this scheme is fully explicit and might thus appear as more efficient than the Crank-Nicolson scheme. This advantage is lost when the scheme is extended to cylindrical geometries. In this case, FFT must be replaced by Hankel transforms. Although Fast Hankel Transform algorithms have been developed, these are not so fast as FFTs and usually require a specific grid point distribution. These constraints must be kept in mind in the design of efficient extensions of the above schemes.

Test of Diffraction Any practical implementation of Eq. (169) must reproduce properly existing analytical solutions. Equation 169) allows for simulations of the propagation of Gaussian beams. The laws of Gaussian optics must therefore be reproduced by a numerical simulation of Eq. (169). In appendix A, we remind the reader of the laws for Gaussian optics for direct test of the practical implementaion of Eq. (169)

Table 4. Spectral Decomposition Algorithm

- 1. definition of useful data, e.g., beam width w_0, focusing length f, laser wavelength λ_0, index of refraction n_0 and central wave number $k_0 = n_0 2\pi/\lambda_0$.
- 2. definition of grids and z-invariant quantities for space marching the field over one step Eq. (190).
  - number of grid points N_x
  - x-grid: $x_j = x_{min} + j\Delta x$, for $j = 0, \cdots, N_x - 1$
  - k_x-grid: $k_{xj} = j\Delta k_x$, for $j = 0, \cdots, N_x/2 - 1$; $k_{xj} = -\pi/\Delta x + (j - N_x/2)\Delta k_x$, for $j = N_x/2 \cdots N_x - 1$ with $\Delta k_x = 2\pi/(N_x \Delta x)$.
  - precalculation of the vector $A_j \equiv \exp[-2i\delta(k_{xj}\Delta x)^2]$ for $j = 0, \cdots, N_x - 1$, with $\delta \equiv \Delta z/4k_0(\Delta x)^2$ and $k_{xj}\Delta x = 2\pi j/N_x$ for $j = 0, \cdots, N_x/2 - 1$, $k_{xj}\Delta x = 2\pi(-1 + j/N_x)$ for $j = N_x/2 \cdots N_x - 1$.
- 3. definition of the initial field $E_j^0 = \mathcal{E}(x_j, z = 0)$, $j = 0 \ldots N_\perp + 1$, and its spectrum $\tilde{E}_j^0 = \tilde{\mathcal{E}}(k_{xj}, z = 0) = FFT(E_j^0)$, $j = 0 \ldots N_\perp + 1$.
- 4. space march the solution by performing a double-loop for $N_z = M \times K_{max}$ propagation steps, with expensive and costless diagnostics:

```
outer loop: k = 1, ..., K_max
    inner loop: m = 1, ..., M
        n = (k − 1)M + m
        Ẽ_j^{n−1} = FFT(E_j^{n−1})
        Ẽ_j^n = Ẽ_j^{n−1} × A_j for all j
        E_j^n = FFT(Ẽ_j^n)
        perform costless diagnostic n
    end inner loop
    perform expensive diagnostic k
end outer loop
```

3.1.2 Diffraction and nonlinear effects

Extended Crank-Nicolson scheme The Crank-Nicolson scheme extends to propagation equations of the NLS type such as Eq. (112) or Eq. (120). In this aim, we will treat nonlinearity by the second order Adams-Bashforth scheme which is an explicit scheme working in general for all type of nonlinear terms. Thus, we define a prototypical equation

$$\frac{\partial \mathcal{E}}{\partial z} = \frac{i}{2k_0}\Delta_\perp \mathcal{E} + \mathcal{N}(\mathcal{E}), \tag{192}$$

where $\mathcal{N}(\mathcal{E})$ models the nonlinearity under investigation, e.g. $\mathcal{N}(\mathcal{E}) \equiv i\frac{\omega_0}{c} n_2 \mathcal{I}\mathcal{E}$ for the optical Kerr effect as in Eq. (112) or $\mathcal{N}(\mathcal{E}) \equiv i\frac{\omega_0}{c} n_2 \mathcal{I}\mathcal{E} - \frac{\beta_K}{2}\mathcal{I}^{K-1}\mathcal{E}$ for multiphoton absorption and Kerr effect as in Eq. (120). The advantage of the Adams-Bashforth scheme with respect to a completely implicit sheme lies in the fact that it preserves the second-order accuracy of the Crank-Nicolson sheme and allows fast calculations of the right hand side in Eq. (192). The implementation of a fully implicit scheme with nonlinear terms would indeed require the resolution of nonlinear implicit equations, which cannot be done as easily as solving linear implicit equations. It usually requires predictor-corrector routines and of a large number of matrix inversions, thereby increasing the computational cost.

The proposed alternative can be viewed as applying a second order Adams-Bashforth time integrator to nonlinear term in the Crank-Nicolson scheme which reads:

$$E_j^{n+1} - E_j^n = i\delta(\Delta_j E_j^{n+1} + \Delta_j E_j^n) + \left\{\frac{3}{2}N_j^n - \frac{1}{2}N_j^{n-1}\right\}, \tag{193}$$

Table 5. Crank-Nicolson Algorithm with explicit nonlinearity treated by the second order Adams-Bashforth scheme

- 1. same as in table 3 and definition of data for nonlinearity, e.g., n_2, β_K.
- 2. definition of grids and matrix storage of L_+ and L_-^{-1}: $L_\pm = 1 \pm i\delta\Delta_j$ as in table 3. Introduction of boundary conditions in L_+ and L_-.
- 3. definition of the initial field $E_j^0 = \mathcal{E}(r_j, z = 0)$, $j = 0 \dots N_\perp + 1$.
- 4. double-loop for $N_z = M \times K_{max}$ propagation steps with two types of diagnostics at each step and each M steps:

```
outer loop: k = 1, ..., K_max
    inner loop: m = 1, ..., M
        n = (k − 1)M + m
        calculate and store vector N_j^{n−1}          (loop j, ··· , N_⊥)
        calculate V_j^{n−1} = L_+ E_j^{n−1}           (product matrix-vector)
        add S_j^{n−1} = V_j^{n−1} + (3N_j^{n−1} − N_j^{n−2})/2 (sum of vectors)
        E_j^n = L_−^{−1} S_j^{n−1}                    (product matrix-vector)
        perform costless diagnostic n
    end inner loop
    perform expensive diagnostic k
end outer loop
```

where

$$N_j^n \equiv \Delta z \mathcal{N}(E_j^n) = \Delta z \left\{ i\frac{\omega_0}{c} n_2 |E_j^n|^2 E_j^n - \frac{\beta_K}{2} |E_j^n|^{2K-2} E_j^n \right\}. \tag{194}$$

Note that the nonlinear terms on the rhs of Eq. (193) only involve previously obtained fields E_j^n and E_j^{n-1}. The coefficients 3/2 and -1/2 ensure the second order accuracy. A scheme where the nonlinear terms are simply written as N_j^n on the rhs of Eq. (193) would work as well, however, the second order accuracy of the Crank-Nicolson scheme would be lost. Equation (193) is still an implicit equation but it allows us to express the vector E_j^{n+1} without doing more effort than in the absence of nonlinearities:

$$E_j^{n+1} = (L_-)^{-1}[L_+ E_j^n + \frac{3}{2} N_j^n - \frac{1}{2} N_j^{n-1}] \tag{195}$$

Equation (195) extends Eq. (181) to the case of nonlinear propagation over a single step and constitutes the core of the scheme, which must be repeated to cover the entire propagation domain. The numerical scheme will therefore be similar to the Crank-Nicolson scheme in table 3, with the following differences: It is no longer necessary to compute and store the product $L_-^{-1}L_+$ in the initialization step 2 since each propagation step use independently L_+ and L_-^{-1}, which should thus be stored in different tables. Only the 4th step in the numerical scheme of table 3 must be modified as indicated in table 5.

The overall stability of the scheme depends on the nonlinear terms, so that a control of the step size Δz may be necessary in contrast to the unconditionally stable Crank-Nicolson scheme of table 3; however, the stability constraint is often found to be not so drastic as that for the description of diffraction with an explicit scheme ($\Delta z \leq k_0(\Delta r)^2$) [38], which justifies the explicit treatment of nonlinearities.

Split-step technique Propagation equations and more generally PDEs including several source terms as in Eq. (192) can be solved by an alternative scheme, the split-step

Table 6. Split-step algorithm with nonlinearity treated by the explicit second order Adams-Bashforth scheme and linear term treated by the Crank-Nicolson scheme

- 4- double-loop for $N_z = M \times K_{max}$ propagation steps with two types of diagnostics at each step and each M steps:

outer loop: $k = 1, \ldots, K_{max}$
  inner loop: $m = 1, \ldots, M$
    $n = (k-1)M + m$
    calculate and store vector $N_j^{n-1} = \Delta z \mathcal{N}(E_j^{n-1})$ (loop $j, \cdots, N_\perp$)
    first *half*-step:$E_j^{n-1/2} = E_j^{n-1}(3N_j^{n-1} - N_j^{n-2})/2$
    second *half*-step:$E_j^n = LE_j^{n-1/2}$ (product matrix-vector)
    perform costless diagnostic n
  end inner loop
  perform expensive diagnostic k
end outer loop

method, that we introduce in the following. The idea of the split-step technique is to perform a fractional step for each source term by means of a suitable scheme that applies to each part. For example the propagation equation (192) takes the form $\partial_z \mathcal{E} = \mathcal{L}\mathcal{E} + \mathcal{N}(\mathcal{E})$, where $\mathcal{L} \equiv (i/2k_0)\Delta_\perp$. Previously presented algorithms allows us to treat each source term as in the set of equations:

$$\partial_z \mathcal{E} = \mathcal{L}\mathcal{E}, \tag{196}$$
$$\partial_z \mathcal{E} = \mathcal{N}(\mathcal{E}). \tag{197}$$

Equation (196) representing the linear part of Eq. (192) can be solved by e.g. the Crank-Nicolson algorithm (Eq. (180) and table 3) or by means of the spectral decomposition algorithm (Eq. (190) and table 4). Equation (197) representing the nonlinear part of Eq. (192) can be solved by e.g. the second order Adams-Bashforth method presented in previous section which reads:

$$E_j^{n+1} = E_j^n + \frac{3}{2}N_j^n - \frac{1}{2}N_j^{n-1} \tag{198}$$

A split step scheme consists in sequentially propagating the envelope over a *fractional-step* of size Δz by each of the algorithms used for Eqs. (196) and (197). Here, *fractional-step* is not related to the step size but means that only part of source terms are considered. By denoting $E_j^{n+1/2}$ the discretized envelope after the first split-step, we obtain the scheme

$$E_j^{n+1/2} = E_j^n + \frac{3}{2}N_j^n - \frac{1}{2}N_j^{n-1}, \tag{199}$$
$$E_j^{n+1} = (1 - i\delta\Delta_j)^{-1}(1 + i\delta\Delta_j)E_j^{n+1/2}. \tag{200}$$

Hence, for the split-step scheme applied to Eq. (192), stages 1 to 3 of table 5 are identical with additional storage of matrix $L = L_-^{-1}L_+$ as in table 3, whereas stage 4 is modified as indicated in table 6. Note that the split-step scheme can be generalized to more than two *half-steps* when there are more than two source terms. The separation between linear and nonlinear effects is also convenient but not mandatory to implement a split-step scheme.

3.1.3 Diffraction and Dispersion

In Sections 3.1.1 and 3.1.2, we have considered beam propagation with $\mathcal{E} = \mathcal{E}(r, z)$ depending on the transverse r and evolution z variables only. The methods we have presented also apply to pulses with one or more additional dimensions. For example with a temporal dimension $\mathcal{E} = \mathcal{E}(r, t, z)$, the new coordinate is discretized as $t_l = t_{min} + l\Delta t$ and pulse propagation is described by applying one of the presented schemes to each time t_l. This is achieved by including each propagation step of previous schemes in a loop on the time index l. The discretized pulse envelope at a given propagation distance z_n must be defined over a two dimensional grid as $E^n_{j,l} = \mathcal{E}(r = r_j, t = t_l, z = z_n)$ for $j = 0, \cdots, N_\perp$ and $l = 0, \cdots, N_t$. The input pulse, e.g., a Gaussian pulse with spatial and temporal quadratic phases modeling lens-focusing (focal length f) and chirp C can be defined as:

$$\mathcal{E}(r, t, z = 0) = \mathcal{E}_0 \exp\left(-\frac{r^2}{w_0^2} - i\frac{k_0 r^2}{2f} - (1 + iC)\frac{t^2}{t_p^2}\right), \tag{201}$$

and its discretized counterpart as $E^0_{j,l} = \mathcal{E}(r = r_j, t = t_l, z = z_0)$. As long as the propagation equation does not involve differential operators with time derivatives, these are the only additional features extending previous schemes from (1+1) to (2+1) dimensions. However, higher dimensionality is usually associated with couplings between the different slices in the additional dimension. In the time direction, dispersive effects plays this role. We will consider the lowest dispersive order, namely second order dispersion to present a standard way to extend the Crank-Nicolson scheme to (2+1)D simulations. We thus start from the diffraction-dispersion equation:

$$\frac{\partial \mathcal{E}}{\partial z} = \frac{i}{2k_0}\Delta_\perp \mathcal{E} - i\frac{k_0^{(2)}}{2}\frac{\partial^2 \mathcal{E}}{\partial t^2} \tag{202}$$

The split-step technique naturally applies to design an algorithm for Eq. (202) with each fractional step relying on either a Crank-Nicolson algorithm, a Fourier decomposition, or a combination of both. The efficient way to implement the first option leads to the Alternate direction implicit scheme. The second option is straightforward and the third will be presented in section 3.1.4.

Gaussian optics for dispersion Starting from a numerical code where diffraction described by Eq. (169) was successfully implemented and checked with respect to the laws of Gaussian optics, any implementation of dispersive terms as in Eq. (202) requires new tests ensuring that (i) diffraction still works properly, (ii) dispersion is correctly implemented and (iii) the combination of both effects is correct.

Test (i) can be easily performed by setting $k_0^{(2)} = 0$ as an additional input condition and checking again the laws for Gaussian optics. Similarly, test (ii) can be performed by setting the diffraction coefficient to zero (parameter δ in the schemes) and by comparing the results to the laws for Gaussian pulse optics. See appendix B for a reminder of the laws of Gaussian optics to be used for a direct test of the implementation of Eq. (169).

3.1.4 Diffraction, dispersion and nonlinear effect - spectral extended Crank-Nicolson scheme

This section presents an extension of the Crank-Nicolson scheme for (2+1)D simulations of paraxial propagation equations presented in the theory section, namely those

which in the canonical form read as:

$$\frac{\partial\hat{\mathcal{E}}}{\partial z} = \frac{i}{2\mathcal{K}_0(\Omega)}\Delta_\perp\hat{\mathcal{E}} + i\mathcal{D}(\Omega)\hat{\mathcal{E}} + \frac{i}{2\mathcal{K}_0(\Omega)}\frac{\omega^2}{c^2}\frac{\hat{\mathcal{P}}}{\epsilon_0}, \tag{203}$$

where $\hat{\mathcal{E}} = \hat{\mathcal{E}}(r,\Omega,z)$, $\Omega = \omega - \omega_0$, $\mathcal{K}_0(\Omega) \equiv \mathcal{K}(\Omega, \mathbf{k}_\perp = 0)$ denotes any of the $\mathcal{K}$ functions listed in the Table 2, and the subscript zero in $\mathcal{K}_0$ will be omitted for simplicity. The proposed extended Crank-Nicolson scheme is simply applied frequency component by frequency component and therefore includes all effects that are naturally included in frequency dependent terms of Eq. (203), namely dispersion, space-time focusing and self-steepening.

The temporal coordinate is discretized with N_t equally spaced steps of size Δt: $t_l = t_0 + l\Delta t$ for $l = 0 \cdots N_t - 1$, we have a corresponding discretization of the spectral domain $\omega_l = \omega_0 + l\Delta\omega$ for $l = 0 \cdots N_t/2 - 1$, $\omega_l = \omega_0 - \pi/\Delta t + l\Delta\omega$ for $l = N_t/2 \cdots N_t - 1$, with $\Delta\omega = 2\pi/[N_t\Delta t]$. The natural variable for the envelope spectra is $\Omega_l = \omega_l - \omega_0$. Note that in this section, the index l will refer to either discrete times or discrete frequencies, depending on whether the quantity we consider belongs to the temporal or the spectral domain. For completeness, we reintroduce a current in the nonlinear terms of Equation (203):

$$\frac{\partial\hat{\mathcal{E}}}{\partial z} = \frac{i}{2\mathcal{K}(\Omega)}\Delta_\perp\hat{\mathcal{E}} + i\mathcal{D}\hat{\mathcal{E}} + \frac{i}{2\mathcal{K}(\Omega)}\frac{\omega^2}{c^2}\frac{\hat{\mathcal{P}}}{\epsilon_0} - \frac{1}{2\mathcal{K}(\Omega)}\frac{\omega}{c}\frac{\hat{\mathcal{J}}}{\epsilon_0 c} \tag{204}$$

Let $\mathcal{K}_l$ and $\mathcal{D}_l$ denote $\mathcal{K}(\Omega_l)$ and $\mathcal{D}(\Omega_l)$, respectively. The numerical scheme extending Eq. (193) and corresponding to Eq. (204) reads:

$$\hat{E}^{n+1}_{j,l} - \hat{E}^n_{j,l} = i\delta_l(\Delta_j\hat{E}^{n+1}_{j,l} + \Delta_j\hat{E}^n_{j,l}) + id_l(\hat{E}^{n+1}_{j,l} + \hat{E}^n_{j,l}) + \frac{3}{2}\hat{N}^n_{j,l} - \frac{1}{2}\hat{N}^{n-1}_{j,l} \tag{205}$$

where

$$\delta_l = \delta\frac{k_0}{\mathcal{K}_l} = \frac{\Delta z}{4(\Delta r)^2\mathcal{K}_l} \tag{206}$$

$$d_l = \frac{\Delta z\mathcal{D}_l}{2} \tag{207}$$

$$\hat{N}^n_{j,l} \equiv \frac{i\Delta z}{2\mathcal{K}_l}\frac{\omega_l^2}{c^2}\frac{\hat{\mathcal{P}}^n_{j,l}}{\epsilon_0} - \frac{\Delta z}{2\mathcal{K}_l}\frac{\omega_l}{c}\frac{\hat{\mathcal{J}}^n_{j,l}}{\epsilon_0 c} \tag{208}$$

The solution to equation (205) represents one step along the propagation direction:

$$\hat{E}^{n+1}_{j,l} = (L_{-,l})^{-1}[L_{+,l}\hat{E}^n_{j,l} + \frac{3}{2}\hat{N}^n_{j,l} - \frac{1}{2}\hat{N}^{n-1}_{j,l}] \tag{209}$$

where $L_{-,l} \equiv 1 - id_l - i\delta_l\Delta_j$, $L_{+,l} \equiv 1 + id_l + i\delta_l\Delta_j$. With respect to previous expressions of L_- and L_+, the matrices $L_{-,l}$ and $L_{+,l}$ are different only by an additional frequency dependent term, i.e., l-dependent, on the diagonal. As for L_- and L_+, the matrices $L_{-,l}$ and $L_{+,l}$ operate on vectors representing transverse profiles (described by index j) of the Fourier components $\hat{E}_{j,l}$ for the envelope, corresponding to the fixed frequency Ω_l; in other words, Eq. (209) allows us to apply the standard Crank-Nicolson scheme to each frequency component l of the envelope spectrum, thus performing one step from $E^n_{j,l}$ at distance n to $E^{n+1}_{j,l}$ at distance $n+1$. This step must be inserted within a loop on frequencies (l).

Depending on the need to save either memory or simulation time, matrices $L_{-,l}$ and $L_{+,l}$ may be either precomputed (to minimize simulation time, in which case the

Table 7. Spectrally extended Crank-Nicolson algorithm

- 1. definition of useful data: laser and medium parameter; in particular everything needed to properly define the dispersion relation in the medium and nonlinearity, e.g., Kerr parameters, ionization rates, etc.
- 2. definition of grids and z invariant quantities:
  - r-grid: $r_j = j\Delta r$, for $j = 0, \cdots, N_\perp + 1$
  - t-grid: $t_l = t_{min} + l\Delta t$, for $l = 0, \cdots, N_t - 1$
  - ω-grid: $\omega_l = \omega_0 + l\Delta\omega$ for $l = 0, \cdots, N_\omega/2 - 1$
    $\omega_l = \omega_0 - \pi/\Delta t + l\Delta\omega$ for $l = N_\omega/2, \cdots N_\omega - 1$, with $\Delta\omega = 2\pi/[N_\omega \Delta t]$ and $N_\omega = N_t$
  - calculation and storage of the one dimensional tables: $\mathcal{K}_l$, $\mathcal{D}_l$, d_l, δ_l, $p_l \equiv \frac{i\Delta z}{2\mathcal{K}_l}\frac{\omega_l^2}{\epsilon_0 c^2}$, $u_l \equiv -\frac{\Delta z}{2\mathcal{K}_l}\frac{\omega_l}{\epsilon_0 c^2}$
- 3. definition of the initial field, $E^0_{j,l} = \mathcal{E}(r_j, t_l, z = 0)$ by e.g., Eq. (201) for Gaussian profiles of the beam and pulse. Input spectral components by FFT: $\hat{E}^0_{j,l} = FFT(E^0_{j,l})$
- 4. double-loop on propagation steps with diagnostics each step and each M steps:

outer loop: $k = 1, \ldots, K_{max}$	
inner loop: $m = 1, \ldots, M$	
$n = (k-1)M + m$	
calculate quantities for nonlinearity, e.g.:	
electron density $\rho^{n-1}_{j,l} = \rho(r_j, t_l, z_{n-1})$	(solve ODE (128) for $j = 1, \cdots, N_\perp$)
Raman-Kerr term $Q^{n-1}_{j,l} = Q_i(r_j, t_l, z_{n-1})$	(solve ODE (130) for $j = 1, \cdots, N_\perp$)
calculate and store $P^{n-1}_{j,l}, J^{n-1}_{j,l}$	($j = 1, \cdots, N_\perp$; $l = 0, \cdots, N_\omega - 1$)
$\{\hat{P}^{n-1}_{j,l}, \hat{J}^{n-1}_{j,l}\} = \mathrm{FFT}(\{P^{n-1}_{j,l}, J^{n-1}_{j,l}\})$	(FFT) $j = 1, \cdots, N_\perp$
$\hat{E}^{n-1}_{j,l} = \mathrm{FFT}(E^{n-1}_{j,l})$	(FFT) $j = 1, \cdots, N_\perp$
calculate and store $N^{n-1}_{j,l}$ from Eq. (208)	(multiply $p_l \hat{P}^{n-1}_{j,l}$, $u_l \hat{J}^{n-1}_{j,l}$ and sum)
loop on frequencies	
$l = 0, \cdots, N_\omega - 1$	
calculate $L_{+,l}, L_{-,l}$	(tridiagonal complex matrices)
calculate$L^{-1}_{-,l}$	(matrix inversion)
calculate $V^{n-1}_{j,l} = L_{+,l}\hat{E}^{n-1}_{j,l}$	(product matrix-vector)
add $S^{n-1}_{j,l} = V^{n-1}_{j,l} + (3\hat{N}^{n-1}_{j,l} - \hat{N}^{n-2}_{j,l})/2$	(sum of vectors)
$\hat{E}^n_{j,l} = L^{-1}_{-,l} S^{n-1}_{j,l}$	(product matrix-vector)
end loop on frequencies (l)	
inverse Fourier transform $\hat{E}^n_{j,l} \to E^n_{j,l}$	(FFT^{-1}) $j = 1, \cdots, N_\perp$
store $E^n_{j,l}$	
perform costless diagnostic n	
end inner loop (m)	
perform expensive diagnostic k	
end outer loop (k)	

additional amount of memory used corresponds to the size of a $14 \times N_\perp \times N_\omega$ table of real numbers, seven diagonals of complex numbers being needed to describe $L_{-,l}$ and $L_{+,l}$) or recomputed at each step (to minimize memory usage). In the second option, step 2 of the scheme is simplified since only a few frequency dependent tables must be precomputed in order to reconstruct $L_{-,l}$ and $L_{+,l}$ in step 4 with a minimum number of operations. The changes in steps 1 to 4 of the Crank-Nicolson scheme are indicated in table 7.

Step 4 still constitutes the bulk of the scheme. It is evident that efficiency of the code is enhanced if all unnecessarily repetitive calculations are avoided. These concern loops including multiplications by factors which do not vary with the loop index. For example it is clear that the quantities p_l and u_l in front of the nonlinear polarization and current must be precalculated and stored in step 2 so that the calculation of $N_{j,l}^{n-1}$ needs only two multiplications and an addition per element (j, l). Constant factors can also usually be removed via renormalization of the discretized equations. As a general rule, the efficiency of a code must be optimized by a careful count of all operations appearing in nested loops and an attempt to minimize them. For example, all matrix-vector operations performed on with tridiagonal matrices must be implemented so as to avoid unnecessary multiplication and sum of zeros. This can be achieved by using compact matrix storage for the non-nil diagonals only, as discussed in [38].

3.2 Numerical Methods for UPPE Solution

Having formulated our pulse propagation models in Section 2, we have seen a number of numerical simulation techniques valid for paraxial equations and we now address the question of how to solve non-paraxial equations numerically. To keep the notation simple, and equations readable, we will restrict ourselves to carrier-resolving equations in the form of the simplest version of UPPE, namely a one-component propagation equation in a bulk medium. We also suppose that all nonlinear interactions are expressed in the nonlinear polarization P and for simplicity omit from our equations the current density term. Note that the numerical approach described next translates directly to a general vectorial case, and its practical implementation in software is essentially the same.

3.2.1 UPPE as a large system of ordinary differential equations

We have written the UPPE equation in a form which resembles the usual structure of pulse propagation equations and to which simulation practitioners in the field are most used to. The UPPE expresses evolution of the spectral (both temporal and spatial) transform of the electric field, and its right-hand-side contains linear and nonlinear terms:

$$\partial_z E_{k_x,k_y}(\omega,z) = ik_z E_{k_x,k_y}(\omega,z) + \frac{i\omega^2}{2\epsilon_0 c^2 k_z} P_{k_x,k_y}(\omega,z) \text{ where } k_z = \sqrt{k^2(\omega) - k_x^2 - k_y^2}\,. \tag{210}$$

Let us point out a few important points before going into details of a solver implementation. First, unlike many propagation models, this is in a spectral representation. It describes evolution of a Fourier spectrum, rather than that of a real physical field. One important consequence is that what we have is not a partial differential equation anymore. Rather, it is a system of ordinary differential equations for spectral amplitudes, albeit a very large system. Thus, there are no partial derivatives to approximate, which makes numerical solution conceptually very simple: One can utilize *any* available library for ODE systems, and the *only* remaining thing to do is to define a right-hand-side calculation subroutine which will be fed to the chosen ODE solver.

Second, the above representation, which is in terms of spectral amplitudes for electric field, is not exactly the one a numerical solver should work with. This is because the slowest-evolving variables in this problem only change in response to nonlinearity, and these are actually the *native variables* A_{k_x,k_y} which appeared in the course of UPPE derivation:

$$E_{k_x,k_y}(\omega,z) = A_{k_x,k_y}(\omega,z)\exp\left[ik_z z\right] \equiv A_{k_x,k_y}(\omega,z)\exp\left[i\sqrt{k^2(\omega) - k_x^2 - k_y^2}\,z\right] \tag{211}$$

Their evolution equation (see Section 2.3) only contains nonlinear terms:

$$\partial_z A_{k_x,k_y}(\omega,z) = +\frac{i\omega^2}{2\epsilon_0 c^2 k_z} e^{-ik_z z} P_{k_x,k_y}(\omega,z) \quad \text{with} \quad k_z = \sqrt{k^2(\omega) - k_x^2 - k_y^2}\ . \tag{212}$$

It is obvious that spectral amplitudes A are the slowest variable in the pulse evolution problem, because they do not change at all in a linear regime. This also means that UPPE equations exactly solve the linear part of a problem, which is an extremely desirable property (reader is encouraged to review various propagation equations specifically to recall how much *effort* often goes even into design of the linear part of all these equations). The most important advantage is the ability to model an arbitrary medium with frequency dependent index of refraction *and* frequency dependent losses.

Alternatively, one can view Eq. (212) as Eq. (210) to which integrating factor $\exp[ik_z z]$ has been applied. This cancels oscillations in the spectral amplitudes of the electric field which are due to linear propagation. Nonlinearity alone contributes to the evolution of the native UPPE variables A_{k_x,k_y}, and implementation based on them thus yields to faster numerical integration. This point of view makes it evident that there is a degree of freedom in the relation between E and A amplitudes. Namely, one could also use an integrating factor $\exp[ik_z(z-z_0)]$, thus moving the point at which $A = E$ from $z = 0$ to z_0. Lacking a better term, we say that E and A amplitudes are *aligned* at z_0.

It is imperative that a simulator utilizes this degree of freedom. At $z = z_0$ spectral amplitudes of E and native variables A coincide, but as z increases, E and A diverge from each other. Although always connected by a simple complex phase change $\Delta\phi = k_z(\omega,k_x,k_y)(z-z_0)$, one must realize that the latter can attain very large values. To avoid numerical difficulties in handling the corresponding exponentials, the relation between E and A has to be re-aligned after every integration step by moving z_0 to the current propagation distance. After the new array of A is produced in the ODE solver taking a step Δz, re-alignment with E is achieved by

$$A^{\text{new}}_{k_x,k_y}(\omega,\Delta z) = \exp[ik_z(\omega,k_x,k_y)\Delta z] A^{\text{old}}_{k_x,k_y}(\omega,\Delta z) \tag{213}$$

After this operation, E and A amplitudes coincide once again when the next integration step is to be executed. Note that the above procedure corresponds to nothing but to the free, linear propagation of the field, and re-alignment therefore amounts to applying a linear propagator. In what follows it is assumed that amplitudes are aligned before each step, which means that $z = 0$ in Eq. (212) should be understood as a beginning of the current ODE solver step. It also means that all z values are small, restricted to $z < \Delta z$.

Equation (212) is not completely explicit, because it hides the fact that the nonlinear polarization P must be calculated from the current value of the electric field. Indeed, polarization is a functional of $E(x,y,t,z)$ taken at a fixed z value. As a rule, medium models are formulated in real space. For example Raman-effect contribution to the change of refractive index can be expressed as convolution (in time) calculated at a given spatial point $(x,y,z) \equiv (\mathbf{r},z)$. While the concrete relation between electric and polarization fields is unimportant for how the UPPE solver is designed, the fact that medium response is calculated in real space is crucial. We will therefore assume that a function implementation $P(x,y,t,z) = P_{\text{NL}}(\{E(x,y,t,z)\})$ is given, and that it calculates nonlinear polarization as a function of time from a history of the electric field at a fixed spatial point.

Let us incorporate this into our notation and write down an explicit definition of the UPPE ODE system. The unknown functions are $A_{k_x,k_y}(\omega,z)$ and they obey

$$\partial_z A_{k_x,k_y}(\omega,z) = +\frac{i\omega^2}{2\epsilon_0 c^2 k_z} e^{-ik_z z} P_{k_x,k_y}(\omega,z,\{E(x,y,z,t)\}) \ , \ \ k_z = \sqrt{k^2(\omega)-k_x^2-k_y^2} \tag{214}$$

where

$$P_{k_x,k_y}(\omega,z,\{E(x,y,t)\}) = (2\pi)^{-3/2} \int e^{+i\omega t - ik_x x - ik_y y} P_{\mathrm{NL}}(\{E(x,y,t,z)\})\, dtdxdy \tag{215}$$

is a Fourier (or in general spectral) transform of $P_{\mathrm{NL}}(\{E(x,y,t,z)\})$ which in turn encapsulates nonlinear medium properties. (Note that a solver implementation does not need to, and in fact should not know about its concrete functional form!) To evaluate the above, one first needs to calculate the real-space field from the native computational variables through another (inverse) Fourier transform

$$E(x,y,t,z) = (2\pi)^{-3/2} \int e^{-i\omega t + ik_x x + ik_y y} A_{k_x,k_y}(\omega,z) e^{ik_z z}\, d\omega dk_x dk_y \tag{216}$$

Now it should be clear that when an ODE solver requests evaluation of its right-hand-side function for a given value of z, and for a given array $A_{k_x,k_y}(\omega,z)$, spectral transforms will be invoked in both directions. One can view this as a price to pay for the elimination of partial derivatives from propagation equations, and for the ability to solve the linear problem exactly.

There is one more issue to clarify before transition to discretization, and that is that of a moving frame. It is of course advantageous, and in fact necessary, to follow pulse evolution in a frame of reference which moves with a velocity v_f (with respect to the laboratory frame) chosen such that at each location z in a lab, the pulse arrives at $t \approx 0$, and thus stays located around the center of the temporal computational domain. This is achieved by expressing time t through $t = \tau + z/v_f$ where τ is our new temporal variable. Inspection of Eqs. (214-216) reveals that this amounts to a simple modification of the linear propagator:

$$\exp\left[ik_z(\omega,k_x,k_y)\Delta z\right] \to \exp\left[ik_z(\omega,k_x,k_y)\Delta z - \Delta z\omega/v_f\right] \tag{217}$$

We emphasize that v_f is an arbitrary parameter which does not reflect any physics of the model. It is merely an expression of what one deems to be the best reference frame. Quite often it is reasonable to choose

$$\frac{1}{v_f} = \frac{1}{v_g} = \frac{\partial k_z(\omega_{\mathrm{pulse}}, k_x = 0, k_y = 0)}{\partial \omega} \tag{218}$$

which means that the computational frame of reference moves with the group velocity of the pulse. Readers familiar with Nonlinear Schrödinger Equation should realize that it is exactly the choice that makes a pulse described by NLS to stay localized in the vicinity of $\tau \approx 0$.

3.2.2 Discretization and spectral transforms

Because any UPPE solver is spectral in all dimensions, grid representation of both, real-space and spectral space fields is determined by properties of discrete spectral transforms. Depending on the symmetry of the problem, these are either variants of Fourier or discrete Hankel transforms.

Spatial (linear) axis

For a spatial dimension, say x, that spans one side of a computational domain box, the values of coordinates (in real space) and transverse wave numbers k_x (in spectral space) are those of ordinary Fourier transform sampling points. Both sets are equidistant and equal in size.

Temporal axis

A computational domain axis in time direction has its corresponding Fourier transform which is slightly modified due to the fact that physical fields are real-valued. It is sufficient to sample spectral amplitudes $A_{k_x,k_y}(\omega)$ only for positive angular frequencies ω. Moreover, one can restrict discrete sampling points to $\omega \in (\omega_{\min}, \omega_{\max})$ if one only knows the medium susceptibility $\chi^{(1)}(\omega)$ in this interval. Only these discrete frequencies become active in the simulations in the sense that they carry corresponding spectral amplitudes. When spectral-to-real transform is invoked, the active set of frequency-samples is padded by zeros before a standard Fourier transform is executed. This has the effect that the resulting real-space amplitude becomes the so-called analytic signal. While its real part corresponds to physical electric field, its modulus squared can be interpreted (in suitable units) as the time-averaged light intensity. Both quantities are often needed in the calculation of various nonlinear medium responses. Note that in this arrangement the total number of discrete samples in the time dimension is more than twice the number of active samples in the frequency dimensions. However, the number of ODEs to solve is given by the active frequency samples.

Radial axis

For problems with axial symmetry, it is advantageous to utilize the radial discrete Hankel transform instead of a two-dimensional Fourier transform. Because the discrete Hankel transform is represented by a full matrix, it is not *fast* in the sense fast Fourier transforms are. Still, the main computational savings are related to the reduced dimensionality of variable-arrays representing physical fields. Sampling points in both real and spectral space are the same and namely given by scaled zeros of Bessel function J_0. For example N samples in real space are $r_i = u_i/u_N R$ where $J_0(u_i) = 0$. Note that there is no radial sample located directly on the axis.

Readers concerned about the usage of *slow* spectral transform, should note that there are fast discrete Hankel transforms. However, UPPE solver requires a truly orthogonal transform implementation, because forward and inverse transformations are executed many times over the same array. Only the proper Hankel transform is orthogonal (and in addition equal to its own inverse) and should be preferred on grounds of numerical accuracy.

3.2.3 Integration of evolution equations for spectral amplitudes

The core of a UPPE solver can be based on essentially arbitrary ODE-solver library. A good library should have the capability to choose between different algorithms, and it will also take care of allocating auxiliary arrays based on the selected method. The advantage of using a canned library over hard-coding a concrete algorithm into the solver implementation is the flexibility in the choice of method, and also the fact that everything concerning auxiliary variables involved in ODE solution remains hidden. However, the UPPE system can contain several million variables, and that an ODE solver will, depending on the requested method, allocate several auxiliary arrays of the same size. As a consequence, the bulk of the memory allocated for the whole simulation will actually be requested by the solver. Not only the memory needs will

be several times larger than those for one copy of all fields, but also that the method performance can be affected by the size of the system solved. In practice simpler methods tend to perform better than the more sophisticated ones. In particular, all methods that require calculation of a Jacobian are utterly unsuitable for UPPE solution. Fortunately, the standard ODE solver work-horses such as various Runge-Kutta methods work well.

Now, suppose we have calculated an array $A_{k_x,k_y}(\omega, z)$ representing the solution at propagation distance z. To keep the notation simple, discrete wave numbers k_x, k_y and active angular frequencies ω now represent array indices. To calculate $A_{k_x,k_y}(\omega, z + \Delta z)$, an ODE solver is invoked to produce it. Because we synchronize A and E representations after each step, we can understand $z = 0$ as if the currently executed step was the very first one. A pseudo-code for the integration loop reads:

Repeat for each step:
A) Invoke ODE Solver:
$$A_{k_x,k_y}(\omega, 0) \to A_{k_x,k_y}(\omega, \Delta z)$$
B) Re-align native and field variables:
$$A_{k_x,k_y}(\omega, \Delta z) = \exp\left[i(k_z(\omega, k_x, k_y) - \omega/v_f)\Delta z\right] A_{k_x,k_y}(\omega, \Delta z)$$

Readers may note that the above integration may seem like an operator splitting method. It does looks as if nonlinear and linear *propagators* were applied in turns the same way as in the split-step approach. However, this is where the similarity ends. The second sub-step is nothing but a *shift* of our reference frame. If our numerics did not suffer from rounding, and could evaluate exponentials with arbitrarily large arguments, this addition would not be necessary.

Behind the scenes, while executing A), ODE solver will invoke calculation of the right-hand-side of our ODE system, i.e. it asks to evaluate

$$\frac{i\omega^2}{2\epsilon_0 c^2 k_z} e^{-i(k_z - \omega/v_f)\delta z} P_{k_x,k_y}(\omega, z, \{E(x, y, \Delta z, t)\})$$

for given spectral amplitudes $A_{k_x,k_y}(\omega)$. Depending on the ODE algorithm, call of this function occurs several times during a single integration step, each time with a different value of Δz. UPPE solver implements the function call in the following steps:

1. Apply linear propagator to shift from $z = 0$ to Δz:

$$A_{k_x,k_y}(\omega) \mapsto A_{k_x,k_y}(\omega) \exp\left[+i(k_z(\omega, k_x, k_y) - \omega/v_f)\Delta z\right]$$

2. Perform the spectral transform from spectral to real space:

$$E(x, y, t) = \mathrm{FFT}\{A_{k_x,k_y}(\omega)\}$$

3. In real-space representation, calculate nonlinear medium response for a given field $E(x, y, t)$, using a user-supplied medium-response implementing algorithm P_{NL}:

$$P(x, y, t) = P_{\mathrm{NL}}\{E(x, y, t)\}$$

4. Perform spectral transform from the real to spectral space:

$$P_{k_x,k_y}(\omega) = \mathrm{FFT}^{-1}\{P(x, y, t)\}$$

5. Apply linear propagator to *undo* the previous shift in z:

$$P_{k_x,k_y}(\omega) \mapsto P_{k_x,k_y}(\omega) \exp\left[-i(k_z(\omega, k_x, k_y) - \omega/v_f)\Delta z\right]$$

6. Finally, multiply by the coupling factor and return result to the ODE solver:

$$P_{k_x,k_y}(\omega) \mapsto \frac{i\omega^2}{2\epsilon_0 c^2 k_z} P_{k_x,k_y}(\omega)$$

The above is the most computation-intensive part of UPPE solution and thus merits attention with respect to efficient parallel implementation.

3.2.4 Parallelization

Even if a problem has axial symmetry, a typical UPPE ODE system contains several million variables. It is therefore more or less necessary that computations are parallelized. Let us briefly point out facts that may influence our parallelization design decisions.

First, we have to take into account the fact that the UPPE framework is inherently spectral. This means that each processor or a thread of execution will, at some point, require access to *distant* locations in allocated arrays. The shared memory paradigm is therefore a natural way to go, and the current UPPEcore implementation uses Pthreads. OpenMP (i.e. pragma based loop parallelization) would probably work equally well for our purposes.

Second, one has to decide which parts of the code will actually execute in parallel. Of course, ideally all of them, but the question is if it is worth of the trouble. It turns out that most of the computational effort in UPPE is spent within the ODE system right-hand-side calculations and in performing spectral transforms. This invites a parallel work crew strategy: A master thread creates a family of workers or slaves and dispatches these to perform work as needed. The master thread executes all work outside of the ODE solver loop which includes all initialization, analysis or diagnostics of results, input, and output. Master also runs the main ODE loop without help from its slaves. (This is of course a compromise between between the achievable parallelization efficiency and complexity of the solution!) This means that almost any serial ODE solver library can be used as a plug-in for the UPPE solver.

The parallel working crew enters a synchronization barrier immediately after their creation. Here, they wait for commands from their master who specifies which is the next function to execute in parallel. For example, this may be a spectral transform. Within the parallel section, each of the workers takes over a proportional part of the load. When the parallel section is done, workers meet again at the synchronization barrier, awaiting further commands from the master.

Obviously the biggest drawback of this design is that the ODE part of the code remains serial. Although it is relatively small, it would limit possible parallel speed up with a large number of threads. An alternative solution is akin to the domain decomposition strategy; the set of ODE equations is evenly distributed between multiple instances of ODE solvers, each executed by an independent thread. However, this solution requires a mild modification of the ODE routines that control adaptive integration step. Some earlier versions of the UPPEcore were based on this approach. Since the performance penalty with the working crew method is mostly negligible in practice, it is preferred because it does not require open access to the code of the ODE solver.

3.3 Numerical methods for nonlinear medium response models

This section is devoted to the numerical implementation of nonlinear response models by using a method similar to the so-called exponential time differencing method [42].

3.3.1 Numerical implementation of plasma related terms

A formal solution to Eq. (124) may be written as:

$$J(\mathbf{r},t,z) = \frac{q_e^2}{m_e}\int_{-\infty}^{t} \exp\left(-\frac{t-t'}{\tau_c}\right)\rho(\mathbf{r},t',z)E(\mathbf{r},t',z)\,dt' \tag{219}$$

From the knowledge of the field and electron density for all grid points, it is therefore possible to determine the current step by step by expressing for each fixed spatial position $(\mathbf{r},z)$ the current at time $t+\Delta t$ as a function of the current at previous time t:

$$J(\mathbf{r},t+\Delta t,z) = \frac{q_e^2}{m_e}\int_{-\infty}^{t+\Delta t} e^{-t+\Delta t-t'/\tau_c}\rho(\mathbf{r},t',z)E(\mathbf{r},t',z)\,dt' \tag{220}$$

Omitting the $(\mathbf{r},z)$ dependence for simplicity, we obtain:

$$J(t+\Delta t) = \frac{q_e^2}{m_e}e^{-\Delta t/\tau_c}\left\{\int_{-\infty}^{t} e^{-(t-t')/\tau_c}\rho' E'\,dt' + \int_{t}^{t+\Delta t} e^{-(t-t')/\tau_c}\rho' E'\,dt'\right\} \tag{221}$$

where $\rho' E' \equiv \rho(\mathbf{r},t',z)E(\mathbf{r},t',z)$. This is rewritten by using a trapezoidal integration rule for the second term on the right hand side:

$$J(t+\Delta t) = e^{-\Delta t/\tau_c}\left\{J(t) + \frac{\Delta t}{2}\frac{q_e^2}{m_e}\rho(t)E(t)\right\} + \frac{q_e^2\Delta t}{2m_e}\rho(t+\Delta t)E(t+\Delta t) \tag{222}$$

Similarly, a formal solution to Eq. (128) reads:

$$\rho(t) = \rho_{\mathrm{nt}}\int_{-\infty}^{t}\exp\left(-\int_{t'}^{t}[W''_{\mathrm{ofi}} - W''_{\mathrm{ava}}]\,dt''\right)W'_{\mathrm{ofi}}\,dt' \tag{223}$$

where e.g. $W'_{\mathrm{ofi}} \equiv W_{\mathrm{ofi}}[\mathcal{I}(t')]$. Eq. (223) is similar to Eq. (219) , thus its solution is similar to Eq. (222) and reads:

$$\rho(t+\Delta t) = e^{-\int_t^{t+\Delta t}[W'_{\mathrm{ofi}}-W'_{\mathrm{ava}}]\,dt'}\left\{\rho(t) + \frac{\Delta t}{2}\rho_{nt}W_{\mathrm{ofi}}[\mathcal{I}(t)]\right\} + \frac{\rho_{\mathrm{nt}}\Delta t}{2}W_{\mathrm{ofi}}[\mathcal{I}(t+\Delta t)] \tag{224}$$

The solutions (224) and (222) take the generic form:

$$A(t+\Delta t) = a[A(t)+\eta Q(t)] + \eta Q(t+\Delta t), \tag{225}$$

where a and η are step-dependent constants and $Q(t)$ is a known function over the whole integration domain. With the (a,η) couples indicated in table 8, the discretized version of Eq. (225) which read as

$$A_{l+1} = a[A_l + \eta Q_l] + \eta Q_{l+1}, \tag{226}$$

allows for the determination of the electron density and current densities over the entire time window by a loop over the time index l.

Table 8.

$A(t)$	$Q(t)$	a	η
$J(t)$	$\rho(t)E(t)$	$e^{-\Delta t/\tau_c}$	$\frac{\Delta t}{2}\frac{q_e^2}{m_e}$
$\rho(t)$	$W_{\text{ofi}}[\mathcal{I}(t)]$	$e^{-\int_t^{t+\Delta t}[W'_{\text{ofi}}-W'_{\text{ava}}]\,dt'}$	$\frac{\Delta t}{2}\rho_{nt}$

3.3.2 Numerical implementation of the Raman-Kerr response

The inclusion in the numerical scheme of the Raman-Kerr contribution is formally equivalent to that of the plasma. We indicate two possibilities to achieve this task.

- 1. Resolution of an ordinary differential equation: Assuming that the envelope $\mathcal{E}(r,t,z)$ is known at a given propagation distance z, both the electron density $\rho(r,t,z)$ and the Raman-Kerr contribution $Q_i(r,t,z) = \int_{-\infty}^{t} \mathcal{R}_0 \exp[-\Gamma(t-\tau)]\sin[\omega_R(t-\tau)]|\mathcal{E}(r,\tau,z)|^2 d\tau$ are solutions to a non-homogeneous ODE that involves $\mathcal{E}(r,t,z)$ as a source term. The temporal profiles $\rho(r,t,z)$ and $Q_i(r,t,z)$ are indeed obtained for each fixed spatial coordinate (r,z) by solving Eq. (128) for ρ with boundary condition $\rho(-\infty) = \rho_0 \ll \rho_{at}$ and Eq. (130):
  These tasks can be done by any ODE solver based on, e.g., the Runge-Kutta scheme.
- 2. Direct resolution. An explicit formulation satisfying Eq. (130) exists for the Raman-Kerr response (the electron density, solution to Eq (128) admits a similar and simpler explicit formulation):

$$Q_i(r,t,z) = \int_{-\infty}^{t} \mathcal{R}_0 \exp[-\Gamma(t-\tau)]\sin[\omega_R(t-\tau)]|\mathcal{E}(r,\tau,z)|^2 d\tau \tag{227}$$

  which can be rewritten as the imaginary part of

$$Q(r,t,z) = \mathcal{R}_0 \left\{ \mathrm{e}^{-\Gamma t + i\omega_R t} \int_{-\infty}^{t} \mathrm{e}^{\Gamma\tau - i\omega_R\tau} |\mathcal{E}(r,\tau,z)|^2 d\tau \right\} \tag{228}$$

  A numerical scheme to compute Eq. (228) is obtained by using a trapezoidal evaluation of the integral term:

$$Q(r,t+\Delta t,z) = \mathrm{e}^{(-\Gamma+i\omega_R)\Delta t} Q(r,t,z) + \frac{\Delta t}{2}\left[|\mathcal{E}(r,t+\Delta t,z)|^2 + \mathrm{e}^{(-\Gamma+i\omega_R)\Delta t}|\mathcal{E}(r,t,z)|^2 \right] \tag{229}$$

  Discretization of this scheme leads to an expression allowing the calculation of the temporal profiles for the complex Raman-Kerr response $Q(r,t,z)$ at each fixed spatial position (r,z).

$$Q^n_{j,l+1} = \left\{ \mathrm{e}^{(-\Gamma+i\omega_R)dt} Q^n_{j,l} + \frac{\Delta t}{2}[|E^n_{j,l+1}|^2 + \mathrm{e}^{(-\Gamma+i\omega_R)dt}|E^n_{j,l}|^2] \right\} \tag{230}$$

  from which $Q_i(r,t,z) \equiv \mathrm{Im}(Q^n_{j,l})$ is obtained. Scheme (230) must be inserted within an outer loop on j (transverse coordinate) and an inner loop on l (time).

4 Conclusion

The last decade has brought lot of progress in ulrafast nonlinear optics, especially in the area of generation and control of femtosecond pulses with extreme intensities. This advancement would not be possible without the contribution of numerical

simulations. Computer models have not only been instrumental in interpretation of experimental results, but represent the most important component of the theoretical picture. As a result, a growing number of researchers in the area wear two hats, one of an experimentalist and one of a computational physicist. Ranks of those who need to utilize computing as a component of their experimental work are wider still.

This motivated the inclusion of a simulation and modeling course and writing of this text. Our main goal was to provide a self-contained overview of theoretical approaches and practical computer simulation methods relevant in the general area of nonlinear optics. Most of the material is presented at a level of detail sufficient to serve as an introduction into computer simulation for the broadest possible audience of researchers and students.

A Gaussian Optics

Using the input Gaussian beam defined by Eq. (170), analytical formulas for Gaussian beam propagation read as:

$$\mathcal{E}(r,z) = \mathcal{E}_0 \frac{w_0}{w(z)} \exp\left(-\frac{r^2}{w^2(z)} + i\frac{k_0 r^2}{2R(z)} - i\Psi(z)\right), \tag{231}$$

where the beam parameters evolution is defined as:

$$\begin{bmatrix} w(z) \\ R(z) \\ \Psi(z) \end{bmatrix} = \begin{bmatrix} w_0\left[(1-\frac{z}{f})^2 + \frac{z^2}{z_R^2}\right]^{1/2} \\ z - d_f + \frac{d_f(f-d_f)}{z-d_f} \\ \arctan\left(\frac{z-d_f}{(fd_f - d_f^2)^{1/2}}\right) \end{bmatrix} \quad \text{or} \quad \begin{bmatrix} w(z) \\ R(z) \\ \Psi(z) \end{bmatrix} = \begin{bmatrix} w_f\left[1 + \frac{(z-d_f)^2}{z_f^2}\right]^{1/2} \\ z - d_f + \frac{z_f^2}{z-d_f} \\ \arctan\left(\frac{z-d_f}{z_f}\right) \end{bmatrix} \tag{232}$$

where the first set in Eq. (232) involves only the input beam parameters and the focal distance $d_f = f/(1 + f^2/z_R^2)$, where $z_R = k_0 w_0^2/2$ denotes the Rayleigh length associated with the input beam width. The second set in Eq. (232) refers to the standard laws for which the origin of coordinate along the propagation axis is the waist position $z = d_f$. Other quantities are the beam waist $w_f = w_0 f/\sqrt{f^2 + z_R^2}$ and the Rayleigh length relative to the beam waist $z_f \equiv k_0 w_f^2/2$. Both sets of equations are identical as consistency is ensured by the relation $d_f(f - d_f) = z_f^2$.

B Gaussian Optics for dispersion

Using Eq. (201) as an input condition, the law for Gaussian pulse propagation reads:

$$\mathcal{E}(t,z) = \mathcal{E}_0 \frac{t_p}{T(z)} \exp\left(-\frac{t^2}{T^2(z)}\left\{1 + i[C + (1+C^2)\frac{z}{z_{\rm ds}}]\right\} - i\Phi(z)\right), \tag{233}$$

where the beam parameters evolution is defined as:

$$\begin{bmatrix} T(z) \\ \Phi(z) \end{bmatrix} = \begin{bmatrix} t_p\left[(1 + C\frac{z}{z_{\rm ds}})^2 + \frac{z^2}{z_{\rm ds}^2}\right]^{1/2} \\ \arctan\left(\frac{(1+C^2)z + C}{z_{\rm ds}}\right) \end{bmatrix} \tag{234}$$

where $z_{\rm ds} = t_p^2/2k_0^{(2)}$ denotes the dispersion length. For normal dispersion ($k_0^{(2)} > 0$) and a positive chirp coefficient, the pulse duration increases with distance whereas

Table 9. List of Abbreviations

PDE	Partial Differential Equation
ODE	Ordinary Differential Equation
UPPE	Unidirectional Pulse Propagation Equation
FME	Forward Maxwell Equation
FWE	Forward Wave Equation
FOP	First-Order Propagation equation
SPE	Short Pulse Equation
NLS	Nonlinear Schrödinger Equation
NEE	Nonlinear Envelope Equation
LEE	Linear Envelope Equation
FEE	Forward Envelope Equation
PC-NLS	Partially Corrected Nonlinear Schrodinger Equation
SVEA	Slowly Varying Envelope Approximation
SEWA	Slowly Evolving Wave Approximation
SEEA	Slowly Evolving Envelope Approximation
GFEA	Generalized Few-cycle Envelope Approximation
MA	minimal approximation
PA	Paraxial Approximation
FFT	Fast Fourier Transform
FHT	Fast Hankel Transform
MPI	Multiphoton Ionization
MPA	Multiphoton Absorption
GVD	Group Velocity dispersion
THG	Third Harmonic Generation
SPM	Self Phase Modulation
SCG	Super Continuum Generation
CEP	Carrier Envelope Phase
FWHM	Full Width at Half Maximum

for a negative chirp coefficient, the pulse duration first decreases until it reaches $T_m = t_p/(1 + C^2)$ at distance $z_m = -Cz_{\mathrm{ds}}/(1 + C^2)$, and then increases while the Gaussian pulse shape is preserved. All these properties must be reproduced in a correct implementation of dispersion.

Since the linear propagation of the pulse preserves the separation of time and space variables, test (iii) consists again in comparing numerical results with the laws for Gaussian optics for the beam and the pulse when both diffraction and dispersion have non nil coefficients.

C Useful definitions

In order to facilitate navigation throughout this text, we provide tables with a list of abbreviations (see Table 9) and a list of symbols used in Equations (see Table 10).

C.1 Abbreviations

C.2 Symbols

Table 10: List of symbols

Symbol	Unit	Physical quantity or constant
$\mathbf{E}$, E, $\mathcal{E}$	V/m	electric field (vector, scalar, envelope)
$\mathbf{B}$, B, $\mathcal{B}$	H/m	magnetic field (vector, scalar, envelope)
$\mathbf{H}$, H, $\mathcal{H}$	H/m	magnetic induction (vector, scalar, envelope)
$\mathbf{D}$, D, $\mathcal{D}$	C/m^2	displacement field (vector, scalar, envelope)
$\mathbf{J}$, J, $\mathcal{J}$	A/m^2	free charge current (vector, scalar, envelope)
$\mathbf{P}$, P, $\mathcal{P}$	C/m^2	nonlinear polarization (vector, scalar, envelope)
$\mathbf{P}^{(1)}$, $P^{(1)}$, $\mathcal{P}^{(1)}$	C/m^2	Linear polarization (vector, scalar, envelope)
$\mathbf{e}_s$		unit vector in the polarization plane (direction)
$\mathbf{z}$	-	unit vector in the z-direction
A_+, A_-, A_μ	units of A	amplitudes of the forward (+) or backward (−) propagating components
$\lambda = \pm 1$, $\pm$	-	indicator for forward or backward propagation
$\mathcal{E}_\lambda$, $\mathcal{H}_\lambda$		harmonic wave solutions to Maxwell equations
$\mathcal{E}_0$, $\mathcal{H}_0$		amplitudes of harmonic waves
$\mathcal{E}_m$, $\mathcal{H}_m$		amplitudes of transverse modes
$N_m(\omega)$		normalisation constant
X, Y, T	m, m, s	normalization volumes or window sizes
$\delta_{m,n}$, $\delta_{\Omega,\omega}$	-	Kroenecker symbols
$\epsilon_0 = 8.85 \times 10^{-12}$	F/m	permittivity of free space
$\mu_0 = 4\pi \times 10^{-7}$	H/m	permeability of free space
$c = 2.998 \times 10^8$	m/s	light velocity in vacuum
$m_e = 9.11 \times 10^{-31}$	kg	electron mass
$q_e = 1.60 \times 10^{-19}$	C	electron charge
$\hbar = 1.05 \times 10^{-34}$	m^{2}kg/s	Planck constant
ω	s^{-1}	frequency coordinate
ω_0, ω_r	s^{-1}	pulse central and reference frequencies
$\Omega \equiv \omega - \omega_0$	s^{-1}	frequency departure from ω_0
t, τ, t'	s	(laboratory, local) time coordinate
z,ζ	m	propagation coordinate
x,y	m	transverse coordinates
$\epsilon(t)$, $\epsilon(\omega)$	-	permittivity of the medium
$\chi^{(1)}(\omega)$	-	material linear susceptibility
$n(\omega)$	-	linear refraction index
$n_0 \equiv n(\omega_0)$	-	linear refraction index at ω_0
$\mathcal{I}$	W/m^2	$\mathcal{I} = \epsilon_0 c n_0 \lvert\mathcal{E}\rvert^2/2$, Pulse intensity
$\mathcal{E}_{\omega_0}$, $\mathcal{E}_{3\omega_0}$	V/m	Electric field envelopes for fundamental and third harmonic pulses
$k(\omega) \equiv n(\omega)\omega/c$	m^{-1}	material dispersion relation
$k_0 \equiv k(\omega_0)$, $k_r \equiv k(\omega_r)$	m^{-1}	wavenumbers at frequency ω_0, ω_r
$v_g \equiv 1/k_0^{(1)}$, v_f	m/s	pulse group and moving frame velocities
$k_0^{(l)} \equiv d^l k/d\omega^l\lvert_{\omega_0}$	s^l/m	dispersion coefficients
$\kappa(\omega) \equiv k_0 + k_0^{(1)}(\omega - \omega_0)$	m^{-1}	propagation constant in NEEs with MA
$\mathcal{D}(i\partial_\tau) \equiv \sum_{l=2}^{+\infty} \frac{k_0^{(l)}}{l!}(i\partial_\tau)^l$	m^{-1}	high-order dispersion
K_z, K_ζ	m^{-1}	propagation constants for the canonical equation
Q, $\mathcal{Q}$	m^{-1}	nonlinear dispersion operators for the canonical equation
$\mathcal{K}_0(\Omega) \equiv \mathcal{K}(\Omega, \mathbf{k}_\perp = 0)$	m^{-1}	linear frequency dispersion
$\partial_z \equiv \partial/\partial z$	m^{-1}	partial derivative with respect to z

$\mathbf{r} \equiv (x, y)$, $r \equiv \sqrt{x^2 + y^2}$	m	transverse coordinate vector, modulus
k_x, k_y, k_z	m^{-1}	transverse (x, y) and longitudinal (z) wave numbers
$\mathbf{k}_\perp \equiv (k_x, k_y)$	m^{-1}	transverse wave vector
$k_\perp \equiv \sqrt{k_x^2 + k_y^2}$	m^{-1}	modulus of the transverse wave vector
$\Delta_\perp$, $\nabla_\perp^2$	m^{-2}	transverse Laplacian
$\chi^{(3)}$	$\mathrm{m}^2/\mathrm{V}^2$	material third-order susceptibility
n_2	m^2/W	nonlinear index coefficient
α	-	fraction of delayed contribution to the Kerr effect
ω_R, Γ	s^{-1}	characteristic Raman frequencies
Q_i	$\mathrm{V}^2/\mathrm{m}^2$	effective oscillator amplitude (Raman response)
$\mathcal{R}_0 \equiv (\Gamma^2 + \omega_R^2)/\omega_R$, $\mathcal{R}(t)$	s^{-1}	Raman response (amplitude and functional form)
K	-	number of photons involved in MPA
β_K	$\mathrm{W}^{1-K}\mathrm{m}^{2K-3}$	cross section for multiphoton absorption
σ_K	$\mathrm{s}^{-1}\mathrm{W}^{-K}\mathrm{m}^{2K}$	cross section for multiphoton ionization
U_i	J	ionization potential or gap
$W(\mathcal{I})$, W_{ofi}, W_{ava}	s^{-1}	ionization rate, optical field ionization, avalanche
τ_c	s	collision time
ρ, ρ_{nt}	m^{-3}	density of electrons and neutral atoms
ρ_c	m^{-3}	critical plasma density
ρ_f	m^{-3}	density of free space charge
$\sigma(\omega)$	m^2	cross section for inverse Bremsstrahlung
P_{cr}, P_{in}, P_0	W	critical, input and peak powers
$p = P_{in}/P_{cr}$	-	ratio of input to critical powers
f, d_f	m	beam curvature (focal length) and focal distance
w_0, w_f	m	beam radius ($1/e^2$), waist
$z_R \equiv k_0 w_0^2/2$, $z_f \equiv k_0 w_f^2/2$, $z_{\mathrm{ds}} \equiv t_p^2/2k_0^{(2)}$	m	Rayleigh lengths for input beam width or waist, Dispersion length
C	-	Chirp coefficient
t_p, τ_{FWHM}	s	pulse duration (half duration at $1/e^2$, FWHM)
$w(z)$, $R(z)$, $T(z)$, $\Psi(z)$, $\Phi(z)$	m, m, s, - , -	Beam width, curvature, pulse duration, axial phases in laws for Gaussian optics
$T_m \equiv t_p/(1 + C^2)$, $z_m \equiv -Cz_{\mathrm{ds}}/(1 + C^2)$	s, m	Minimal pulse duration (chirped pulse) and pulse shortening distance
Δr, Δx, Δz	m	radial, transverse and longitudinal step-sizes
Δt, $\Delta\omega$	s, s^{-1}	time and frequency step-sizes
$x_{\min}$, $x_{\max}$	m	boundaries of the transverse numerical grid
$r_{\min}$, $r_{\max}$	m	boundaries of the radial numerical grid
$\omega_{\min}$, $\omega_{\max}$	s^{-1}	boundaries of the spectral grid
$N_\perp$, N_x, N_t, N_z	-	Numbers of steps in radial, transverse, time and longitudinal directions
M, $K_{\max}$	-	Numbers of propagation steps between numerical diagnostics
Δ_j, Δ_t		Discretized Laplacian operators
$\nu = 0$ or 1	-	switch between planar and cylindrical geometries
$\delta = \Delta z/4k_0(\Delta r)^2$	-	normalized diffraction coefficient
$d = -\Delta z k_0^{(2)}/4(\Delta t)^2$	-	normalized dispersion coefficient
$L_\pm(\delta) \equiv 1 \pm i\delta\Delta_j$, $L \equiv L_-^{-1}L_+$	-	Tridiagonal matrices in the Crank-Nicolson scheme
$L_{+,l}$, $L_{-,l}$	-	frequency dependent tridiagonal matrices
$\mathcal{K}_l$, $\mathcal{D}_l$	m^{-1}	discretized values for $\mathcal{K}$ and $\mathcal{D}$ at ω_l
$\delta_l = \frac{\Delta z}{4(\Delta r)^2\mathcal{K}_l}$		frequency dependent diffraction coefficient

$d_l = \frac{\Delta z \mathcal{D}_l}{2}$		discretized dispersion values
$\mathcal{L}$, $\mathcal{N}$		linear and nonlinear source terms
N_j^n, $N_{l,j}^n$	-	discretized nonlinearity at z_n, r_j, ω_l
A^* (or c.c.), A^T	-	complex conjugate of A, transpose of A
$A * B$	-	convolution
$\hat{A}(x, y, \omega)$		1D Fourier transform of $A(x, y, t)$, $(t \to \omega)$
$\tilde{A}(k_x, k_y, \omega)$		3D Fourier transform of $A(x, y, t)$

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