Theory and Simulation of Ultrafast Intense Pulse Propagation in Extended Media

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(Invited Paper)

***Abstract*—The theory of femtosecond pulse propagation in dispersive nonlinear media is reviewed with emphasis on modeling light–matter interactions in femtosecond optical filaments. Discussion of the principles underlying the pulse propagation models is followed by the description of the "standard" light–medium interaction model utilized in the ultrafast nonlinear optics, and open problems are identified across the field.**

***Index Terms*—Femtosecond filamentation, nonlinear optics, ultrafast optics, wave propagation.**

I. Introduction

THE propagation of intense ultrashort laser pulses in gases and condensed media [1] (see, e.g., [2], [3] for a review) involves strong coupling between temporal and spatial degrees of freedom. Consequently, a rigorous propagation model must allow in principle for ultrabroadband spectrum generation in the presence of realistic linear and nonlinear material dispersion and absorption, and vectorial and potentially nonparaxial effects. Progress over the past decade in experiments utilizing multiterawatt laser pulses in the atmosphere, somewhat lower power pulses in gases (in a cell or extended hollow waveguide) and with milli-Joule or nano-Joule pulses in condensed media, has prompted the derivation of a number of improved nonlinear envelope propagation models (see, e.g., [4]–[12], [14]–[21]). Ultrafast pulse experiments and applications are now entering a phase that challenge the validity of physical models utilized for longer pulses in nonlinear optics. As mentioned earlier, these experiments require resolution of the optical response to the exciting field over huge spectral bandwidths. Additionally, when nonlinearity and strong self-focusing effects dominate, pulses can undergo strong compression down to a few cycles, potentially develop optical shock waves, and generate strongly anisotropic nonequilibrium distributions of photoionized electrons and ions. Currently, there are two major challenges facing the modeler when it comes down to capturing the correct physics: 1) a correct electromagnetic (EM) propagator derived from the vector Maxwell equations and 2) a quantitatively correct physical description of all physical processes occurring within the strongly interacting nonlinear core. The latter material models currently utilized in pulse propagation studies are phenomenological in nature describing situations more relevant to near-monochromatic or long pulse regimes. When propagation over extended distances is not an issue, such as higher harmonic generation in a gas cell, for example, more rigorous quantum mechanical models describing intense field photoionization are being developed. Even such time domain models are computationally intensive and are restricted typically to simple atomic or diatomic gases. Marrying these two rigorous approaches and scaling them to large scale to represent potentially hundreds of interacting light filaments or smart beams (Bessel [22]–[24], Airy [25]–[28], etc.) with transversely extended wings remains an open challenge.

In this paper, we highlight a unidirectional pulse propagation equation (UPPE) [29], [30] optical carrier resolved propagation model that has proved remarkably successful in yielding almost quantitative agreement with experiments in air and condensed media. In fact, we step back and first introduce a full counterpropagating spectral propagator derived directly from Maxwell's equations and discuss its broad applicability. We then specialize to the unidirectional scalar version and show how the many nonlinear envelope models, including nonlinear Shrödinger equation (NLSE) [31], the extensively used nonlinear envelope equation (NEE), and other nonlinear envelope equations, can be seamlessly derived from the former. Following this, we illustrate the applicability of the UPPE model and contrast it with the well-known NEE [12], [13] model.

II. Pulse Propagation Modeling

The main focus of this section is to present a state-of-the-art carrier-resolved ultrashort pulse propagator and connect it to existing nonlinear envelope models. Such a model is suitable for the extremely nonlinear regimes when the temporal, spatial, and spectral shapes of the pulsed waveform undergo significant changes during its interaction with the medium. Much of our discussion is directed toward the so-called UPPEs. The derivation of the UPPE has been discussed in a relatively detailed fashion in [29]. Here, for convenience, we recap the key steps in its derivation and further elucidate its broad applicability to EM propagation problems.

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A. Time-Propagated and Space-Propagated Equations

Pulse propagation models in nonlinear optics are derived from the underlying vector Maxwell equations and can be formulated in two possible ways: 1) the initial condition, which means both the electric and magnetic fields, is specified throughout the space for a given initial time, and the evolution is then calculated along the time axis. This is the natural setting for evolving the original vector Maxwell equations. 2) The initial condition is given as a function of the local time (in the frame moving with the pulse) and of the two transverse (w.r.t. propagation direction) coordinates. Then, the numerical evolution proceeds along the propagation, e.g., z-axis. We refer to these cases as time- and z-propagated equations. The z-propagated approach is much more common in nonlinear optics simulations, while the time-propagated method is common for solvers based on direct integration of Maxwell's equations.

The versions of the UPPE which are numerically evolved in the spatial and temporal directions are in fact quite similar to each other. Not only is the mathematical form of these equations formally the same, but, most importantly, also the approximation which makes it possible to separate the two counterpropagating waves is expressed in the same physical assumptions which concerns the nature and the strength of the nonlinearity. However, the respective derivations are rather different, and so far there is no proof that the two approaches are in some sense equivalent. A point worthwhile to make is that it can be explicitly showed that while the pair of forward and backward evolution equation is formally exact as long as they are solved together, the set of all UPPE solutions is in fact smaller than the set of all Maxwell's equation solutions. It is interesting to note that such distinction is not required for the t-propagation, and this is one of the reason we believe that the two approaches are in fact not equivalent. From the practical point of view, the z-propagated approach has more than one advantage over simulation in the t-propagated picture, and these will be discussed in the following.

B. Bidirectional and Unidirectional Pulse Propagation Equations

We now review the formal derivation of the "bidirectional pulse propagation equation (BPPE)" and then discuss its reduction to propagation in one direction. In the following section, for simplicity, we connect its solutions to those of the 1-D wave equation as a means of elucidating the solution structure and a connection to other nonlinear envelope models.

In order to derive the fully vectorial, carrier-resolved BPPE, we start from Maxwell's equations

$$\begin{aligned} \vec{J} + \partial_t \vec{P} + \epsilon_0 \partial_t \epsilon * \vec{E} &= \nabla \times \vec{H} \\ -\mu_0 \partial_t \vec{H} &= \nabla \times \vec{E} \end{aligned} \tag{1}$$

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

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

Here, we have separated the linear and nonlinear medium responses. This separation is not unique, and indeed, it may be appropriate to do a different splitting if we pursue formal mathematical methods using asymptotics to extract the dominant component of the nonlinear response. The linear properties of the medium (or, in a waveguide, also the geometric contribution to the dispersion) are accounted for exactly through the frequency-dependent permittivity. The nonlinear response is represented by the polarization and current density

$$\vec{P} = \vec{P}(\{\vec{E}\}) \quad \vec{J} = \vec{J}(\{\vec{E}\})$$

where we emphasize that both are functionals of the electric field. It will become evident later that while this formal separation of linear and nonlinear contributions in the light–matter interaction admits an arbitrary split, it is important to include the full linear chromatic dispersion and absorption, both as functions of frequency, into the medium permittivity, and only leave truly nonlinear interactions contribute to $\vec{P}$ and $\vec{J}$.

EM fields of a light pulse propagating along the z-axis can be expanded into modes that reflect the geometry of the problem. Naturally, in the homogeneous bulk medium, these modes are the well-known plane wave solutions. Another possible set of basis functions in bulk would be so-called conical waves, i.e., Bessel, Airy, etc. However, at this point, we only require that the studied structure, or geometry, only depends on the coordinate z. The electric and magnetic field expansions are then expressed through the common spectral amplitudes A

$$\begin{aligned} \vec{E}(x,y,z,t) &= \sum_{m,\omega} A_m(\omega,z) \vec{\mathcal{E}}_m(\omega,x,y) e^{i\beta_m(\omega)z - i\omega t} \\ \vec{H}(x,y,z,t) &= \sum_{m,\omega} A_m(\omega,z) \vec{\mathcal{H}}_m(\omega,x,y) e^{i\beta_m(\omega)z - i\omega t}. \end{aligned} \tag{2}$$

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

The procedure, outlined in detail in [29], is based on using the modal fields, and their orthogonality properties to project from the full Maxwell's system the evolution equations for the spectral decomposition amplitudes A. These in turn can be used straightforwardly to obtain the two coupled unidirectional (bidirectional) pulse propagation equations in the following form:

$$\begin{aligned} \partial_z \vec{E}^{\perp}_{k_x,k_y,+}(\omega,z) &= +ik_z \vec{E}^{\perp}_{k_x,k_y,+}(\omega,z) \\ &+ \sum_{s=1,2} \vec{e}^{\perp}_s \vec{e}_s \cdot \left[\frac{i\omega^2}{2\epsilon_0 c^2 k_z} \vec{P}_{k_x,k_y}(\omega,z) - \frac{\omega}{2\epsilon_0 c^2 k_z} \vec{J}_{k_x,k_y}(\omega,z) \right] \\ \partial_z \vec{E}^{\perp}_{k_x,k_y,-}(\omega,z) &= -ik_z \vec{E}^{\perp}_{k_x,k_y,-}(\omega,z) \\ &- \sum_{s=1,2} \vec{e}^{\perp}_s \vec{e}_s \cdot \left[\frac{i\omega^2}{2\epsilon_0 c^2 k_z} \vec{P}_{k_x,k_y}(\omega,z) - \frac{\omega}{2\epsilon_0 c^2 k_z} \vec{J}_{k_x,k_y}(\omega,z) \right] \end{aligned} \tag{3}$$

where $\vec{k}$ denotes the wavevector whose z-component is the frequency- and transverse wavenumber-dependent plane-wave propagation constant

$$\vec{k} = \{k_x, k_y, k_z \equiv \sqrt{\omega^2 \epsilon(\omega)/c^2 - k_x^2 - k_y^2}\} \tag{4}$$

and $\vec{e}_s$ is a pair of (polarization) unit vectors orthogonal to $\vec{k}$ and to each other. The forward and backward electric fields that appear in these equations are the Fourier transforms in time and in the transverse coordinates. For example, to obtain the forward-propagating $(+)$ field in the real-space representation, we compute

$$\vec{E}_+(x,y,z,t) = \int E_{k_x,k_y,+}(\omega,z) e^{-i\omega t + i\vec{k}.\vec{r}} dk_x dk_y d\omega.$$

The forward and backward equations are mutually coupled through the polarization and current density terms, because these are medium responses to the *total* electric field of the optical pulse. Explicitly,

$$\vec{P}_{k_x,k_y}(\omega,z) = \vec{P}_{k_x,k_y}(\omega,z,\{\vec{E}_+(x,y,z,t) + \vec{E}_-(x,y,z,t)\})$$

depends on the total electric field, and similarly does the current density. The concrete functional dependence of P and J on the electric field is unimportant for establishing the validity of these equations (see the next section devoted to the medium models for examples). Moreover, as we resolve the full optical field, all higher harmonics are automatically included in contrast to envelope models where one generates spectral features relative to a reference carrier frequency. It has to be emphasized that the two coupled unidirectional (bidirectional) equations are exact. Additionally, the BPPE model includes nonparaxial effects that would be relevant in realistic settings involving strong focusing in a vacuum prior to entering a nonlinear medium or in simulating petawatt pulses.

The UPPE reduction arises when we specialize to a laser pulse propagating in a single direction under conditions where there are no significantly generated backward propagating fields. In the absence of natural reflecting boundaries, this could be violated if strong gradients from shock waves are established downstream or a mechanism for phase-matched back propagation via a generated plasma, for example, was operative. The UPPE assumption is that the nature of the nonlinearity is such that we can calculate the nonlinear response *solely* from the forward propagating field, e.g.,

$$\begin{aligned}\vec{P}_{k_x,k_y}(\omega,z) &\approx \vec{P}_{k_x,k_y}(\omega,z,\{\vec{E}_+(x,y,z,t)\}) \\ \vec{J}_{k_x,k_y}(\omega,z) &\approx \vec{J}_{k_x,k_y}(\omega,z,\{\vec{E}_+(x,y,z,t)\}).\end{aligned} \tag{5}$$

With this approximation, the forward-going UPPE can be solved alone, isolated from its backward propagating counterpart. This then represents a very large, nonlinear system of ordinary differential equations for the field Fourier components that can be practically solved in many important applications. In general, all three components of the field are needed when the medium response is calculated. In such a case, the longitudinal component E_z is obtained from the divergence equation $\nabla.\vec{D} = 0$ using the medium constitutive relation.

The characteristic feature of the UPPE is its spectral character. This is what gives us the capability to include, essentially without any approximations, both the chromatic dispersion (i.e., the wavelength dependence of the refractive index) and the frequency-dependent linear losses. On the other hand, the fact that the native representation of the UPPE is in the spectral space spanned by frequency and transverse wave numbers make it necessary to use spectral transforms to go forth and back between the real and spectral representations. This is because the models of the light–medium interactions are invariably expressed in the real space, and in general require knowledge of the whole history of the electric field at a given point in space.

C. Alternative Path to UPPE: The Wave Equation

The aforementioned derivation and the resulting evolution model appear rather complicated, partly due to their generality. It is, therefore, useful to narrow our attention to a simplified case intended to elucidate certain issues. The following section is meant to show a simplified derivation corresponding to the scalar, 1-D wave equation. It will allow us to make connection to usual way forward propagating evolution equations have been derived in the literature. One may assume that it makes no difference whether we start derivations from Maxwell or wave equations, but it will soon become clear that even such important issues as conditions for validity of the model strongly depend on its mode of derivation.

For the moment, let us ignore any transverse variations in the fields, and assume propagation in the positive z-direction. Furthermore, assume that the electric and nonlinear polarization fields are polarized in the same direction, i.e., we consider a dispersive but isotropic medium. Then, $\mathbf{E}(\mathbf{x},t) \to \widehat{\mathbf{e}} E(z,t)$ and $\mathbf{P}_{\mathrm{NL}}(\mathbf{x},t) \to \widehat{\mathbf{e}} P_{\mathrm{NL}}(z,t)$ for some field direction $\widehat{\mathbf{e}}$ which is transverse to the propagation direction. The nonlinear wave equation can then be written in scalar form as

$$\left(\partial_z^2 - \frac{n^2(\omega)}{c^2}\partial_t^2\right) E(z,t) = \mu_0 \partial_t^2 P_{\mathrm{NL}}(z,t) \tag{6}$$

with the polarization term implicitly depending on the electric field in some nonlinear way, details of which are irrelevant for now. We next assume that the electric field takes the form of

$$E(z,t) = \int_{-\infty}^{\infty} A(z,\omega) e^{i[k(\omega)z - \omega t]} \frac{d\omega}{2\pi} \tag{7}$$

where the electric field is a real quantity, so $E(z,t) = E^*(z,t)$. This implies $A(z,-\omega)e^{ik(-\omega)z} = A^*(z,\omega)e^{-ik(\omega)z}$ and we can write (7) in terms of two integrals over positive frequencies

$$\begin{aligned}E(z,t) = &\int_0^{\infty} A(z,\omega) e^{i[k(\omega)z - \omega t]} \frac{d\omega}{2\pi} \\ &+ \int_0^{\infty} A^*(z,\omega) e^{-i[k(\omega)z - \omega t]} \frac{d\omega}{2\pi}.\end{aligned}$$

By substituting into the driven wave equation (6), we obtain

$$\int_0^\infty \left[\partial_z^2 A + 2ik(\omega)\partial_z A - k^2(\omega)A\right] e^{i[k(\omega)z-\omega t]}\frac{d\omega}{2\pi} + \int_0^\infty \left[\partial_z^2 A^* - 2ik(\omega)\partial_z A^* - k^2(\omega)A^*\right] e^{-i[k(\omega)z-\omega t]}\frac{d\omega}{2\pi} - \frac{1}{c^2}\partial_t^2\int_{-\infty}^t n^2(t-\tau)E(z,\tau)d\tau = -\mu_0\int_{-\infty}^\infty \omega^2 P_{\rm NL}(z,\omega)e^{-i\omega t}\frac{d\omega}{2\pi}.$$

Since the convolution of the index of refraction with the electric field is a real quantity and the right side involving the nonlinear polarization is also real, we have $n^2(-\omega) = n^2(\omega)$ and $P_{\rm NL}(z,-\omega) = P_{\rm NL}^*(z,\omega)$. After some manipulations, the following equality holds:

$$\int_0^\infty \left[\partial_z^2 A + 2ik(\omega)\partial_z A\right] e^{i[k(\omega)z-\omega t]}\frac{d\omega}{2\pi} + \text{c.c.} = -\mu_0\int_0^\infty \omega^2 P_{\rm NL}(z,\omega)e^{-i\omega t}\frac{d\omega}{2\pi} + \text{c.c.}. \quad (8)$$

The aforementioned equality is satisfied when the condition

$$\left[\partial_z^2 A(z,\omega) + 2ik(\omega)\partial_z A(z,\omega)\right] e^{ik(\omega)z} = -\mu_0\omega^2 P_{\rm NL}(z,\omega) \quad (9)$$

is met for positive frequencies. *If in addition to this $A(z,\omega)$ satisfies the slowly varying spectral amplitude approximation $|\partial_z A(z,\omega)| \ll |k(\omega)A(z,\omega)|$*, then the evolution equation becomes

$$\partial_z A(z,\omega) = \frac{i}{2k(\omega)}\mu_0\omega^2 P_{\rm NL}(z,\omega)e^{-ik(\omega)z}. \quad (10)$$

We stress that this is not an envelope equation. Using (8), we obtain that the electric field satisfies the forward UPPE equation

$$\partial_z E(z,\omega) = ik(\omega)E(z,\omega) + \frac{i}{2k(\omega)}\mu_0\omega^2 P_{\rm NL}(z,\omega). \quad (11)$$

Let us emphasize again that this is nothing but the scalar case of the UPPE equation which we derived in the previous section. It may, therefore, come as a surprise that we arrive at this point using a slowly varying amplitude approximation—something we clearly did not have to assume in our derivation based on Maxwell's equations in [29]. Next, we show that while it was a sufficient condition for the validity of the resulting propagation model, it is certainly not necessary. Indeed, a complete description of the fields will require a backward field component; if this field assumes the form

$$E(z,t) = \int_{-\infty}^\infty B(z,\omega)e^{-i[k(\omega)z+\omega t]}\frac{d\omega}{2\pi} \quad (12)$$

then the driven wave equation implies

$$\left[\partial_z^2 B(z,\omega) - 2ik(\omega)\partial_z B(z,\omega)\right] e^{-ik(\omega)z} = -\mu_0\omega^2 P_{\rm NL}(z,\omega). \quad (13)$$

In particular, if the electric field is the sum of a forward traveling piece and a backward traveling piece, then we can satisfy the driven wave equation if

$$\left[\partial_z^2 A(z,\omega) + 2ik(\omega)\partial_z A(z,\omega)\right] e^{+ik(\omega)z} + \left[\partial_z^2 B(z,\omega) - 2ik(\omega)\partial_z B(z,\omega)\right] e^{-ik(\omega)z} = -\mu_0\omega^2 P_{\rm NL}(z,\omega). \quad (14)$$

We can rewrite (14) as

$$\partial_z\left[(\partial_z A)e^{ik(\omega)z} + (\partial_z B)e^{-ik(\omega)z}\right] + ik(\omega)\left[(\partial_z A)e^{ik(\omega)z} - (\partial_z B)e^{-ik(\omega)z}\right] = -\mu_0\omega^2 P_{\rm NL}(z,\omega)$$

from where it is easy to see that the wave equation is satisfied *exactly* if the forward and backward amplitudes satisfy the following first-order evolution equations:

$$\partial_z A(z,\omega) = +\frac{i}{2k(\omega)}\mu_0\omega^2 P_{\rm NL}(z,\omega)e^{-ik(\omega)z}$$
$$\partial_z B(z,\omega) = -\frac{i}{2k(\omega)}\mu_0\omega^2 P_{\rm NL}(z,\omega)e^{+ik(\omega)z}. \quad (15)$$

This is of course equivalent to the pair of UPPEs (or Bidirectional equation) in the scalar, 1-D approximation. Thus, here we can see what was the origin of the seeming inconsistency in derivation of (11): if the wave equation is considered as the source for a one-way evolution equation (which it has been numerous times in the literature), then it is necessary to utilize *both* the forward and backward field components. A single, directed field component *does not satisfy the nonlinear wave equation*. This means that such a successful model as the NEE was originally derived from an equation that should not be satisfied by the sought waveform in the first place.

At this stage, we can connect to other NEEs derived in the literature ([4]–[12], [14]– [21]). These can be seamlessly derived from the scalar UPPE by simply choosing an (ω_r, k_r) frequency–wavenumber reference pair and Taylor expanding in (ω, k) relative to this reference pair. This is the basis for all nonlinear envelope models and clearly distinguishes them from UPPE in that the latter involves absolute frequencies and wavevectors. Details of the derivation of these models from UPPE are given elsewhere, and here, we simply write down the well-known NEE model due to Brabec and Krausz [12] as this is the most extensively used in modeling and we will use this in the next section to contrast with UPPE.

The NEE is an envelope equation in which the complex envelope amplitude $\mathcal{A}$ describes the optical field, and evolves according to

$$\partial_\xi\mathcal{A} = \frac{i}{2k_{\rm R}}\left(1+\frac{i}{\omega_{\rm R}}\partial_t\right)^{-1}\Delta_\perp\mathcal{A} + i\hat{D}\mathcal{A} + \frac{ik_{\rm R}}{2\epsilon_0 n_b^2(\omega_{\rm R})}\left(1+\frac{i}{\omega_{\rm R}}\partial_t\right)\mathcal{P} \quad (16)$$

where

$$\hat{D} = -\frac{\alpha_1}{2}\partial_t + \sum_{n=2}^\infty\left(\frac{\partial^n k}{\partial\omega^n}\right)_{\omega=\omega_{\rm R}}\frac{(i\partial_t)^n}{n!} \quad (17)$$

is the dispersion operator whose coefficients can be related to the real and imaginary parts of the complex propagation constant $k(\omega)$ and thus to the index of refraction.

This equation has been derived under the assumption of the slowly evolving wave approximation which, in contrast to the usual slowly varying envelope approximation [32], requires the field not to change significantly over a wavelength distance along the propagation characteristic. This approximation includes an assumption that there is little difference between the phase and group velocities. Note that the latter requirement only concerns the linear chromatic properties of the medium, which is rather unexpected, because in a linear Maxwell problem there is no difficulty in separating the forward and backward field components and their corresponding evolution equations. It is *solely* the effect of nonlinearity that makes such a separation nontrivial. Thus, in the light of the previous section, one should ask if the assumptions put forward by the authors of the NEE are truly necessary conditions? Recall that they were invoked in order to reconcile the one-way propagating NEE solution with the full wave equation, but the latter is not the equation which a nonlinear pulse should satisfy. Indeed, it is possible to show [29], starting from UPPE and adopting a paraxial approximation followed by neglecting chromatic dispersion in the wavelength-dependent diffraction term and expressing the field in terms of an envelope, that the NEE is in fact more robust than it may seem based on the original assumptions. This emphasizes the importance and advantage of consistent Maxwell equations-based approach in treatment of pulse propagation models.

III. Light-matter Interaction Models in Femtosecond Nonlinear Optics

A. Linear Material Response

The total wavevector $\vec{k}(\omega)$ appearing in the UPPE (11) describes all linear optical properties of the host medium. This includes linear dispersion and absorption bands of the real material. The latter could be extracted from experimental data, for example. In particular, for all applications in the nonlinear regime that lead to significant spectral broadening, it is essential to employ models that can treat the linear chromatic properties of the medium with sufficient accuracy.

Historically, the NLSE remains the canonical description of ultrashort pulse propagation in extended media. Formally, this propagation model can be derived consistently via a singular perturbation approach starting from Maxwell's equations [31]. Traditionally, higher order corrections to NLSE arise as higher order terms in an asymptotic expansion. The standard NLSE only describes the chromatic dispersion "landscape" of the modeled material up to second order in the dispersion. At this level, in 1-D we observe (in certain regimes) solitons, for which inclusion of the group velocity dispersion (GVD) is crucial. However, such an equation cannot properly describe radiation of dispersive waves from solitons—higher order dispersion terms are necessary to capture such effects. In 2-D and 3-D, the standard NLSE has singular solutions that lead to blow up over finite distances. These are termed "critical self-focusing" or "critical collapse" in 2-D and supercritical collapse in 3-D. A more ad hoc approach to adding correction terms to NLSE is to augment the second derivative w.r.t. time in the NLSE by corresponding higher order terms, with their coefficients chosen to mimic the proper frequency dependence of the refractive index (and/or of the geometric dispersion in waveguides). An example of such an approach is the well-known NEE by Brabec and Krausz. This was formulated originally as a formal Taylor expansion of a dispersion operator that can, in principle, account for arbitrarily high-dispersion orders. However, such a formal treatment does not recognize the fact that Taylor series expansions in general exhibit a finite radius of convergence. As a consequence, while the NEE has been intended for extremely broad spectra situations, the dispersion operator expansion may limit its utility in certain regimes. Let us assume that we want to investigate ultrafast nonlinear dynamics in a medium with a narrow absorption band located within the spectral window of interest. Such an absorption feature will result in a pole (in the complex frequency plane) that restricts the radius of convergence of the dispersion operator. Then, no matter how many terms are included in the operator expansion, the resulting chromatic dispersion as "seen" by the numerical model would not converge for frequencies generated beyond the absorption band. In other words, the way the NEE has been formulated, its applications are restricted to the relatively simple chromatic landscapes it can capture. This may be fully sufficient for applications in bulk media confined to the "center" of a transparency window, but it may not suffice for structured materials such as photonic crystal fibers in which the effective refractive index and the accompanying frequency-dependent loss can exhibit a very rich behavior which includes multiple "transparency windows" alternating with "absorption frequency bands."

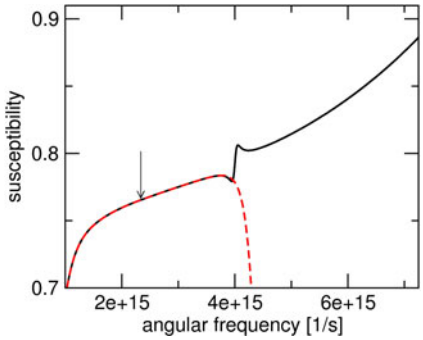

Fig. 1. Susceptibility of water with an added "agent" that exhibits a narrow absorption band around 500-nm wavelength (black line). The red dashed line depicts the numerical susceptibility as "experienced" by an ideal model implementation of the dispersion operator used in the NEE. The order of the dispersion operator expansion is 20 in this picture, and the center wavelength is 800 nm (marked by an arrow). Inclusion of more higher order terms cannot provide any approximation beyond the absorption band.

This issue is illustrated in Fig. 1, which shows the susceptibility of a hypothetical water sample with addition of a dilute absorber. The added agent has an absorption band located at around 500-nm wavelength. To illustrate what a perfect implementation of the NEE equation would result in, we assume a central wavelength of 800 nm and include 20 orders in the expansion of the dispersion operator in the NEE. The picture

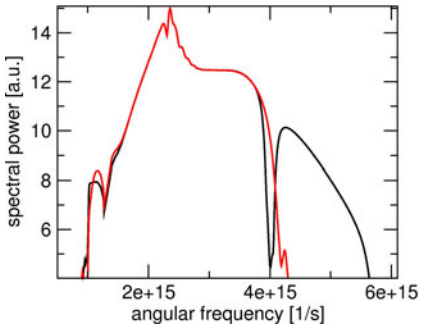

Fig. 2 Effect of the finite radius of convergence in the dispersion operator of the NEE. The simulated spectra for a water sample with an absorption band. The black curve shows the UPPE-based results that take into account index and loss over the whole spectra range. The red curve corresponds to an ideal implementation of the Brabec and Krausz NEE—this model fails at frequencies near and above the absorption band.

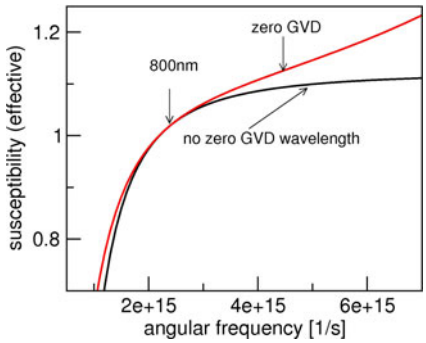

Fig. 3. Submicrometer diameter silica strand effective susceptibility (red) and its SPE-type approximation for the wavelength of 800 nm. Note that while the real system exhibits a zero-GVD wavelength, the approximation does not.

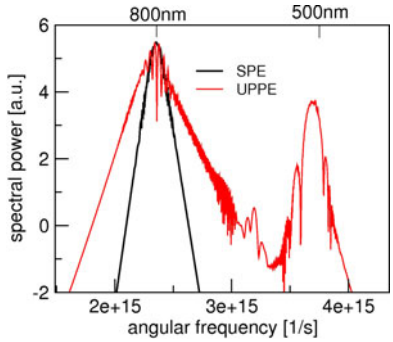

Fig. 4 SC generation in the silica strand. The UPPE-based simulation (red) correctly generates a strong dispersion wave component that shows up as a peak at short wavelength. Due to the lack of zero-GVD wavelength in the SPE model (black), the simulation fails to reproduce basic features of the SC generation process in microstructure fibers.

illustrates the fact that such an expansion is unable to provide any reasonable approximation at frequencies higher than the absorption band. This is a principal problem caused by the fact that the radius of convergence of the operator is finite.

The restricted frequency interval in which the NEE dispersion operator can approximate the properties of the medium becomes very evident in the simulation. This is shown in Fig. 2, which compares a UPPE-based supercontinuum (SC) generation (black) in the previously described "sample," with that calculated using the dispersion operator approach (red). The UPPE spectral solver correctly exhibits a gap in the SC spectrum caused by the absorber, but the rest of the SC radiation remains unaffected. The dispersion operator cuts off the spectrum close to the absorption band, and thus restricts the applicability of the model to a single "transparency window."

On the other hand, UPPE as a Maxwell-like carrier-resolved propagator does not suffer from this restriction. The chromatic properties of the medium enter through a complex-valued function, say $\epsilon(\omega)$ which captures both the index of refraction and losses over an arbitrarily broad spectral range. The "only" real restriction here is in fact the availability of reliable and sufficiently accurate experimental data that span the relevant spectral window to serve as input to the model $\epsilon(\omega)$. Once we have a sufficiently accurate, smooth (i.e., free of numerical interpolation artifacts) representation of the permittivity, this is "applied" directly during the solution of the UPPE. This is of course made possible by the fact that the native computational variables are actually the spectral amplitudes of the electric field, and the propagator is a diagonal operator in the spectral space.

To emphasize the importance of the accurate treatment of the linear medium properties, we offer yet another illustration which concerns the so-called 1-D short pulse equation (SPE). Derived by Alterman and Rauch [33] as an asymptotic expansion in a small parameter [31] and further analyzed by Schäfer and Wayne [34], this equation is designed to capture the ultrabroadband spectrum of the propagating pulse.

Here, we want to point out one property of this equation that is intimately connected to the way it was derived. The authors chose to restrict the model to those media which can be described by the wavelength-dependent susceptibility of the form

$$\chi \approx \chi_0 + \chi_2 \lambda^2.$$

This choice results in the wave equation that subsequently lends itself to multiple scales analysis, and leads to the SPE. As pointed out by the authors, different functional dependence of the linear dispersion on wavelength would lead to different SPEs.

We want to emphasize that the fact that the medium chromatic properties are described in terms of only two parameters drastically restricts the range of applicability of this propagation model.

This is demonstrated in Figs. 3 and 4, which relate to an important regime of short-pulse propagation in silica wires and SC radiation in fibers in general. Fig. 3 compares the effective susceptibility of the silica waveguide to its best SPE-type approximation for the wavelength of 800 nm. While the approximation is locally good, and one could argue that the shape of the susceptibility function is also captured, the simulated spectra reveal a principal drawback of the SPE. Fig. 4 compares the UPPE spectrum to that generated in the corresponding SPE-type approximation, and shows that the SPE is completely missing the so-called dispersive wave component of the SC. The reason for this is the absence of the zero-GVD wavelength in the SPE model. What is worse, it can be shown that the SPE has encoded

the medium properties in such a way that no zero-GVD point can exist in this model. One consequence of this restriction is that the equation is inappropriate to simulate this very important regime of pulse propagation in fibers.

With respect to previous examples, it has to be emphasized that accurate treatment of chromatic dispersion is possible, and indeed necessary, both in the normal and anomalous group velocity dispersion regimes. While equivalent from the mathematical or model-implementation point of view, there is a significant difference between the anomalous and normal GVD frequency domains (see, e.g., [35], [36]). In particular, while normal GVD results in pulse splitting [37], the anomalous dispersion regime exhibits different pulse dynamics, which in turn can be measured and visualized in angularly resolved spectra. Because of the typical shape of the chromatic dispersion landscape, anomalous GVD regime is often affected by the generation of dispersive waves that actually propagate with normal group velocity dispersion. As a consequence, integration of pulse evolution equations tends to be somewhat more demanding from the numerical point of view. Nevertheless, spectral-based methods such as UPPE can handle both mixed dispersion regimes very well.

B. Nonlinear Material Response

Most of extreme nonlinear optics is currently studied in nonmagnetic materials and in the nonrelativistic intensity regimes in which the effects due to magnetic fields are negligible. In the following, we describe what can be considered as the standard model in the general areas of femtosecond optical filamentation and high-harmonic generation.

The most important physical effects that influence propagation of ultrashort, high-power light pulses in nonlinear dispersive media include the optical Kerr [38], [39] and stimulated Raman effects [40], free-electron generation, defocusing by the generated plasma, and losses caused by avalanche and multiphoton ionization (MPI). With minor modifications, models including these effects can be used for description of ultrashort optical pulses propagation in gases [1], [41]–[56], in condensed bulk media [57]–[61], and in conventional, microstructured, and tapered fibers [62]–[64] as well as in ultrathin waveguides [65]. Over the last 15 years, the community developed an accepted model that has been instrumental in understanding many filamentation-related phenomena. However, recent experimental improvements and some unexpected results from them have provided a strong indication that the light–medium interaction models need revision. We are entering a stage in which experiments are about to achieve the fidelity that will require much more accurate modeling. This motivates the discussion in this section: we review the current state of the art, expose its weaknesses, and identify ways to qualitatively improve the models.

Nonlinear effects are usually described in terms of nonlinearly induced polarization and current density. The polarization P enters through the material constitutive relation

$$\vec{D} = \epsilon_0 \epsilon * \vec{E} + \vec{P}(\{\vec{E}\}. \tag{18}$$

The star in this formula represents a convolution integral with ϵ being the linear response function corresponding to the frequency-dependent $\epsilon(\omega, x, y)$. The nonlinear polarization can be an "arbitrary" function of the electric field $\vec{P} = \vec{P}(\vec{E})$. In the present context, it is related to the optical Kerr effect and to the "delayed Kerr nonlinearity" which is due to anisotropic polarizibility of molecules and their alignment in strong optical fields.

We will also discuss the current density that is driven by the optical field $\vec{J} = \vec{J}(\vec{E})$ to describe interactions with plasma generated by the high-intensity optical pulse. Often, defocusing effects are treated within the polarization term. It has to be emphasized that while formally equivalent, the way such a model is usually implemented results in different dispersion properties. We will discuss this approximation as a special case of the current–density-based model, and will provide an example of an experiment where the distinction between the two approaches becomes important.

1) Optical Kerr and Stimulated Raman Effects: Although very different in microscopic origin, the optical Kerr and the stimulated Raman effects appear similar from a modeling point of view. The local contribution to the nonlinear polarization can be written, and conveniently implemented, in terms of the time-dependent modification of the medium susceptibility

$$\vec{P} = \epsilon_0 \Delta\chi \vec{E}. \tag{19}$$

The susceptibility change itself then responds to the history of the light intensity I at a given spatial location

$$\Delta\chi = 2n_b n_2 \left[(1-f)I + f\int_0^\infty \mathcal{R}(\tau) I(t-\tau)\mathrm{d}\tau\right]. \tag{20}$$

Here, f is the fraction of the delayed nonlinear response and $\mathcal{R}$ is the memory function of the stimulated Raman effect. It has to be emphasized that, here, the intensity I should be understood as the cycle-averaged quantity. Therefore, in both terms, we have nonlinear sources that probe the spectrum (very) roughly centered around the wavelength of the driving pulse. The stimulated Raman effect memory function of course depends on the medium. In gases, it originates from the molecular reorientation induced by the high-intensity field and exhibits "revivals" on a picosecond time scale. This is typically a much longer scale than that of the individual pulse generated by the femtosecond laser system. It is, therefore, often sufficient to include the initial portion of $\mathcal{R}(\tau)$. Most often this is parametrized in the form $\mathcal{R}(\tau) \sim \sin(\Omega\tau)e^{-\Gamma\tau}$ in filament simulations (see, e.g., [2]) in gases. An advantage of this particular response form is that it can be very efficiently implemented without performing the convolution explicitly. A considerably more complex function may be necessary to use in condensed media, this, again, depending on the pulse duration. For example, in silica more than ten "effective" oscillators must be used to capture the Raman memory accurately [66]. Then, it may be more effective to apply the Fourier transform-based evaluation of the convolution integral.

The typical ratio f between the instantaneous and delayed Kerr effect contribution is about one half. However, it has to be noted that some recent experiments employing a pump–probe

approach can measure the respective contribution more precisely than the previous experiments performed more than a decade ago. At the time of this writing, this is a subject of debate with strong indication that in longer duration pulses it is actually the delayed nonlinearity that is the major effect.

Now, let us turn our attention to the instantaneous part of the Kerr effect. This is due to the response of the tightly bound electrons to the optical field. As such, electrons can follow the driving field without much of a delay, which in turn results in an instantaneous response. If we treat the Kerr effect as strictly instantaneous, then the polarization is simply proportional to the cube of the real electric field (it has to be emphasized that this is only true for *an instantaneous* effect)

$$P(\vec{r},t) \sim E(\vec{r},t)^3. \tag{21}$$

Note that both quantities here are real physical fields, not their envelopes. This model can be used with the propagation equations that do not rely on the use of envelopes. The physical difference from the aforementioned envelope-based treatment is that (21) contributes not only to the frequency range of the driving pulse (i.e., to the so-called fundamental), but also generates the third harmonic [67] and higher harmonics. Third harmonic generation is a well-characterized effect in optical filaments and it is, therefore, important to include this real-field-based model in simulations designed to capture higher harmonics. The first attempt to include third harmonic generation in the filamentation models used coupled envelopes, each centered at the fundamental and higher harmonics. We emphasize that such an approach lacks proper justification in the context of ultrafast optics and should be avoided. This is because the generated SC typically spans many decades in frequency encompassing these harmonics, making a unique separation into separate envelope components impossible.

2) Ionization in Strong Optical Fields: Now we turn to the next major player affecting dynamics in femtosecond filaments and in the high-harmonic generation. This is ionization in intense optical fields. Somewhat imprecisely, this is often referred to as the MPI, although it has to be kept in mind that tunneling ionization can also occur. In filaments at atmospheric pressure, the actual regime of ionization is in the crossover between the MPI and tunneling.

Because of the potentially high intensities occurring in femtosecond pulses, free electrons are generated by MPI and by the avalanche mechanisms. Consequently, it is then also necessary to account for the response of the optical field to the presence of a dilute plasma. Since the relevant time scales are very short, plasma diffusion and ion motion can be neglected. In the standard model, the free-electron density ρ is usually obtained as a solution to an equation of the following form [45], [50], [51]:

$$\partial_t \rho = aI\rho + b(I) - c\rho^2. \tag{22}$$

Here, I is the light intensity, a parametrizes the avalanche free-electron generation, and $b(I)$ represents the MPI rate that is a highly nonlinear function of the intensity. The last term describes plasma recombination.

Another important issue related to ionization is, naturally, that it induces losses in the optical fields. Within the standard model, this effect is included simply as a loss term in the propagation equation which is designed to impose a loss of energy equal to that needed to liberate the electrons from their parent neutrals. Also possible is to model the losses caused by MPI as either an equivalent current (see, e.g., [16], [68]) or an imaginary susceptibility contribution (which itself is proportional to the free electron density) that extracts from the field the energy needed for the free-electron generation.

IV. Free-Electron Induced Defocusing and Losses

It is usually assumed that the collective electron velocity $\vec{v}$ responds to the optical field and the total current density is governed by the following simple equation (see, e.g., [68]):

$$\frac{d}{dt}\vec{J}(t) = \frac{e^2}{m_e}\rho(t)\vec{E}(t) - \vec{J}(t)/\tau_c \tag{23}$$

where τ_c is the mean time between collisions experienced by electrons. This equation is solved together with (22) to capture effects of the plasma on the propagation of the optical field, namely defocusing due to plasma and plasma-induced losses. Effectively, the aforementioned equations are mathematically equivalent to the so-called Drude plasma model.

As an alternative to treating the electrons freed from the neutrals (atoms or molecules constituting the gas) via the current density, a simplified model is often used due to its ease of implementation. Given the density of free electrons, one can treat the plasma-induced effects as a susceptibility modification, and lump them with the rest of $\vec{P}$ which simplifies numerical calculation. Then, $\partial_t \vec{P} = \vec{J}$ is interpreted as a part of nonlinear polarization time derivative, and $\vec{P}$ is approximated by

$$\vec{P} = \epsilon_0 \Delta\chi_{\mathrm{pla}}(\rho)\vec{E} = \rho\frac{ie^2}{m_e\omega_{\mathrm{R}}(1/\tau_c - i\omega_{\mathrm{R}})}\vec{E} \tag{24}$$

with ω_{R} being a chosen reference angular frequency. It needs to be emphasized that this approximation completely neglects the plasma-induced chromatic dispersion. In most situations related to femtosecond filaments this may not be a problem, especially if we take into account how crude the model is to start with. However, to model certain pump–probe experiments in which the plasma generated by the pump pulse is detected by the probe pulse centered at a sufficiently different frequency, the model based on the current density equation must be used. The susceptibility-based approach would fail to reflect that the induced index of refraction depends on the wavelength. For example, if the probe is derived from the second harmonic of the fundamental, the simplified treatment would overestimate the effect on the probe by a factor of four!

V. Open Problems in Modeling Light-Matter Interactions in Ultrashort Pulses

There is no doubt that the standard model as described in previous section is a crude, essentially phenomenological way to describe the dynamics in the optical filaments in air and other gases. The only reason, we believe, that it has "survived" for more than a decade is actually related to the fact that also the experiments in the filamentation field have been a far cry from

being quantitative and reproducible. While this situation has been improving recently, it is fair to say that filamentation experiments are in general not reproduced by other groups, many of them are qualitative in nature, and the few quantities that experimentalists attempt to characterize quantitatively are very difficult to access in real (as opposed to simulated) experiments. The impact on the simulation efforts is that even the parameters that characterize the present models are not known with sufficient accuracy. Historically, the standard model provided an important simulation tool, and was effective in bringing us to the current level of understanding of the femtosecond pulse propagation dynamics. However, such qualitative phenomenology is no longer sufficient, and one now has to address the issue of more quantitative first principles model development. We address in the following sections what we see as the most important related problems.

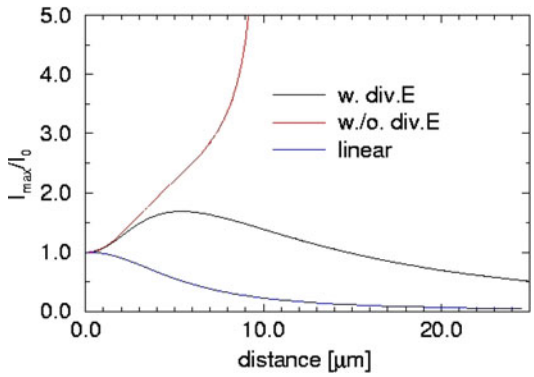

Fig. 5. Collapse regularization via the nonlinear $\nabla.E \neq 0$ related terms. A tightly focused pulse in a pure Kerr medium.

A. Open Problems Related to Extreme Propagation Regimes

So far, the filamentation community has concentrated its efforts on understanding of "naturally" occurring filaments. While a great deal of interest has been devoted to the control of single and multiple filamentation regimes, the aim has been mostly on manipulating the location of the resulting hot spots. This is very much in line with current understanding that the individual filament properties are to a certain degree universal. Because the onset of filamentation is driven by the self-focusing, which in turn is controlled by the optical Kerr effect responding to the local light intensity, the inner structure of the filaments is mostly independent of how they are created. In particular, it is assumed that due to the dynamic balance between the self-focusing and collapse arrest mechanism, the latter being dispersion and/or plasma based, the typical thickness of filaments is significantly larger than the light wavelength. Consequently, vectorial effects are not likely to play a role in natural filaments. While the issues related to the possibility of the collapse arrest due to nonparaxial diffraction [14], [69], [70] regimes as well as effects caused by the vector nature of the optical field [7] have been studied theoretically, there are so far no experiments which would reveal, say, vectorial effects in filamentation in gases. This is why the great majority of the pulse propagation models used in this field are based on paraxial approximations, and the few approaches which go "beyond paraxial" can be viewed as an "overkill" in the context of filamentation. One of the effects held responsible for preventing filaments to "enter" nonparaxial and vectorial regimes is so-called intensity clamping [71], [72]. Even when the energy carried by the pulse increases, the collapse cannot reach finer spatial scales and instead the volume and the transverse cross section of the filament core actually increase, and the paraxial approximation remains well justified.

However, the aforementioned argument may not be valid for future experiments with "artificial" filaments. For example, preparation of the hot spot in a medium with low (e.g., helium) or zero nonlinearity (vacuum) and launching the beam into a medium with a wavelength-sized waist may potentially induce novel dynamics. To illustrate the potential effect that the modeling would have to deal with, we present Fig. 5 which shows a comparison of scalar and vectorial approaches for a tightly focused pulse suddenly entering a Kerr-type nonlinear medium. For a spot size comparable with the wavelength, the longitudinal field component, usually disregarded in filament modeling, increases. Together with the gradient of intensity, it gives rise to a $\nabla.P$ term that appears in the vectorial UPPE model. It turns out that for moderate power that leads to collapse within several micrometers of propagation, this correction is sufficient to arrest critical self-focusing.

The gap between the filament size and nonparaxial regimes is significantly smaller in condensed media. So, it is conceivable that truly nanoscale nonlinear optics could be studied in future experiments with filaments. Then, we have to address practical simulation issues related to the implementation of fully vectorial models, as these require significantly larger computation volumes in comparison with the models restricted to the paraxial approximation.

An open theoretical question related to this is whether one-way propagation can be sustained when nonlinear interactions occur within a volume of a few cubic wavelengths. A possible testbed for such problems could be efficient coupling of the UPPE-type solvers with the direct Maxwell equations' solvers. While the unidirectional propagator would handle the pulse approach toward the tight interaction "zone," the Maxwell solver could take over within the small volume where strong vector–nonlinear effects take place.

B. Open Problems Related to the Kerr Effect

Of course, (19) neglects the dependence of the Kerr effect on wavelength. Although $\Delta\chi$ may exhibit a finite memory due to the Raman contribution, it acts on the instantaneous value of $\vec{E}$ only. This is in part due to only rather limited data available on frequency dependence of the nonlinear coefficients n_2 (see [73] for silica), but it also simplifies practical calculations considerably. Consequently, the "background" index of refraction n_b can be approximated by a constant value taken at the central frequency of the initial pulse. Physically, this approximation is equivalent to the assumption that the Kerr effect is strictly instantaneous. However, with the ever shorter and more intense

pulse lasers available, the fact that the Kerr effect has a finite, albeit short memory will have to be addressed in the near future.

Another important issue which generated a heated debate recently is the role of higher order nonlinearity saturation versus plasma limiting. In a recent experiment, Loriot *et al*. have presented measurements for major gases present in the atmosphere [74], [75]. They proposed that the Kerr effect strongly depends on the intensity of light, and published values for the corresponding nonlinear coefficients are up to an order of 10. This is currently an open, very exciting issue. While it is beyond the scope of this paper, we mention it as an example of a development that clearly indicates limitations of the standard filamentation model.

C. Open Problems Related to Ionization and Defocusing

The part of the model dealing with free electrons is even more problematic than the Kerr nonlinearity model. First, let us consider ionization. This is characterized in terms of rates, be it in the filamentation modeling or in the fundamental, theoretical, and experimental studies of ionization in strong fields. In this respect, one has to recognize that the notion of the rate is an ill-defined quantity in the present context. Indeed, if we characterize the ionization probability as a rate, we silently assume that the ionization yield at the given moment of time does not depend on the previous history of the atom or molecule subjected to the time-dependent waveform of the femtosecond pulse. Intuition alone states that this cannot possibly be true in sufficiently strong fields. It has been argued that the notion of rate can be applied to the driving pulses that contain several optical cycles under their envelopes. At the same time, simulations have indicated that much shorter subpulses can be created in the process of self-compression. The problem is made worse by the fact that such ultrafast features are extremely broad in frequency. Because the ionization models and measurements that provide input to pulse propagators are invariably related to a specific wavelength, one has to ask, what is the carrier frequency that determines the ionization when the spectrum is very broad? Clearly, for such situations what is desirable is a model that incorporates correctly the *history of the driven electronic system*, and "translates" it into the polarization and current density contributions in the light-propagation equations.

To illustrate the need for a wavelength-sensitive model of ionization in strong optical fields, we have performed a simulation of multiple filament formation in an ultrashort "pancake" pulse. Namely, we consider a 7 fs duration pulse with an initial intensity of 4×10^{16} W/m^2. We concentrate on a 5 mm $\times$ 5 mm patch of what we assume to be a very wide beam that suffers from a random phase modulation in both transverse dimensions. The width to length "aspect ratio" of such a pulse is enormous, hence the characterization as a "pancake." This geometry is very different from what is usually employed in filamentation experiments. In most cases, the initial duration of the pulse is significantly longer than that of the self-compressed "subpulses" that occur inside of optical filaments. In this case, the initial duration is so short that dispersion immediately competes with the self-focusing and random phase front perturbations. We are, thus, in a regime in which all important effects "kick in" simultaneously. This is different from naturally occurring filaments in which the main players contribute more or less accumulatively. It has to be emphasized that while not at all common, this regime is in principle possible to achieve in experiments. For example, one could launch a widely collimated beam from vacuum through an aerodynamic window. The observation we want to emphasize for the purpose of this review is related to an extreme spectral behavior that can potentially invalidate the MPI model based on the notion of a central pulse wavelength.

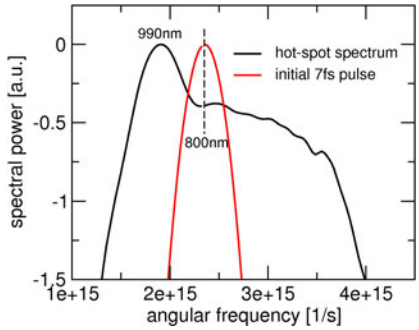

Fig. 6. Extreme frequency shift in a hot spot occurring within a wide "pancake" femtosecond pulsed beam propagating in air. The initial propagation was a pulse with the duration of 7 fs, and the beam passed through a random phase screen before impinging on a vacuum–air "interface" where the perturbed phase front initiated random filament formation.

Fig. 6 shows a spectrum of a selected hot spot on the cross section of a wide beam after the initial propagation over 2.5 m. The most violent dynamics occur in the initial few meters of propagation due to the extreme nature of the launch pulse. It is interesting to compare the initial spectrum, itself rather broad due to the very short pulse duration, to that of a hot spot (i.e., randomly generated filament). The latter appears to be red-shifted to such a degree that the notion of the central wavelength spanning the "entire life" of a pulse completely loses justification. This is, therefore, an example of a regime when a first-principle ionization model independent of a prescribed pulse wavelength would be much more appropriate. Although the overall spectrum of the beam may still be assigned a roughly 800-nm wavelength, it is the high-intensity spots where essentially all ionization occurs, and such a frequency shift should be taken into account. We believe that construction of such models and their integration with the pulse propagation simulators are one of the most interesting and important emerging problems in this field.

Besides the reference wavelength, another fundamental problem in the standard model is related to ionization losses, which are included as "correlated" with the ionization yield, but are otherwise independent. In particular, they affect all spectral components of the pulse equally. This is of course in conflict with the fact that shorter wavelengths are more efficient in overcoming the ionization potential of the medium. In reality, the ionization losses of the optical energy are intimately connected with the induced phase changes, and are in fact two aspects of the same process. (The good analogy here is the real and imaginary parts of the linear optical susceptibility that are related

to each other through causality.) The current standard model completely disregards this issue.

Next, let us consider the free-electron-induced defocusing. This is described in terms of a Drude model in which all electrons are characterized by the same, sharply defined velocity, and by the typical time between collisions. It is further envisioned that the electrons "lose" any memory of their kinetic state preceding the collision. This model is also strictly speaking unjustified in the context of femtosecond pulses. Indeed, within a few optical cycles, there is no time for the electron liberated from an atom to collide with other molecules or atoms, or other free electrons. What we have is essentially a single-atom, single-electron process, and it is questionable whether this can be phenomenologically modeled by equations mathematically equivalent to the Drude model. It is conceivable that the Drude-like model can be justified in some sense, in particular if the collisional "equilibration" is replaced by the scattering off the parent ion. However, at present a microscopically based treatment of this problem is still missing.

VI. Conclusion

Theory and simulation of ultrashort, high-power optical pulse propagation in dispersive, nonlinear media have contributed crucially to progress in the ultrafast science, and especially in the field of optical filamentation. We have reviewed the main ideas that support various pulse propagation models with an emphasis on identifying the open challenging problems.

Recent experiments provide more and better insight into the dynamics governing intense light–matter interactions, and this means that the theoretical basis and practical implementations of the computer models for extreme nonlinear optics require significant improvement.

Among the most important issues to address, at least for the simulation community, is a departure from the phenomenology based models to ones that are rigorously justified on the microscopic level. This represents a major challenge for both theory and practical computing.

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