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Mini-Review

Linear and Nonlinear Optics of Broad-Band Laser Pulses: Diffraction

Ekaterina Iordanova, Pavlina Miteva, Diana Dakova, Hassan Chamati,* Georgi Yankov, Daniela A. Georgieva, and Lubomir M. Kovachev*

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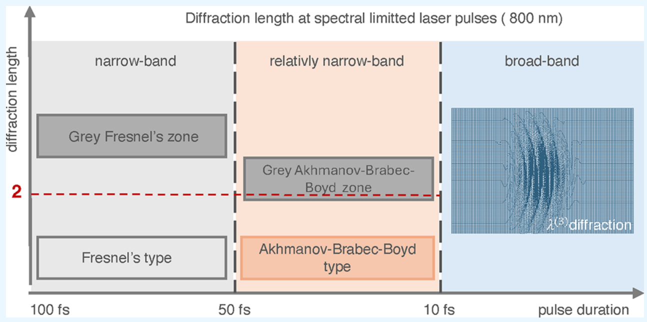

ABSTRACT: The typical spectrally limited laser pulse in the near-infrared region is narrow-band up to 40−50 fs. Its spectral width Δk is much smaller than the carrying wavenumber k_0 ($\Delta k \ll k_0$). For such kinds of pulses, on distances of a few diffraction lengths, the diffraction is of a Fresnel's type and their evolution can be described correctly in the frame of the well-known paraxial evolution equation. The technology established in 1985 of amplification through chirping of laser pulses triggered remarkable progress in laser optics along with the construction of femtosecond (fs) laser facilities producing high intensity fields of the order of 10^{15}−10^{21} W/cm^2. However, the duration of the pulse was quickly shortened from picoseconds down to 5−6 fs, which have a broad-band nature ($\Delta k \sim k_0$). The linear and nonlinear propagation dynamics of broad-band pulses is quite different form their narrow-band counterparts. Here, we review the appropriate theoretical approach to study the evolution of the pulse. Moreover, we shed light on the different diffraction regimes inherent to both narrow-band and broad-band laser pulses and compare them to unveil the main differences. Using this very method, in subsequent papers, we will investigate the influence of the dispersion and nonlinearity on the laser pulse propagation in isotropic media.

1. INTRODUCTION

The technology established in 1985 of amplification through chirping of laser pulses[1] promoted fast progress and construction of femtosecond (fs) laser facilities producing high intensity electric fields on the order of 10^{17}−10^{21} W/cm^2. However, the duration of the pulse was quickly reduced from a few picoseconds to 5−6 fs. This allowed the unfolding of unexpected novel nonlinear effects due to multiphoton ionization, high harmonics emission, plasma defocusing, tunnel ionization, and many other mechanisms. Experiments investigating the propagation of fs pulses in air, in a gas medium, and quartz glass unveiled other nonlinear phenomena such as filamentation,[2] GHz and THz emissions,[3,4] rotation of the polarization plane,[5] merging, and energy exchange between filaments.[6] During the early experiments in air on filamentation,[3] the typical experimental set up already produced nonlinear focus by self-focusing, where the field intensity reached values as high as 10^{14}−10^{15} W/cm^2 to allow for plasma generation and defocusing processes. Therefore, the first theoretical models[3,7] relate the waveguide propagation with a balance between self-focusing and plasma defocusing. The standard theory of filamentation applies under the assumption that the intensity is about $I \sim 10^{14}$ W/cm^2 and that the plasma density is on the order of 10^{16} cm^{-3}. The theoretical models obeying these requirements[8] are based on a balance between self-focusing, diffraction, and ionization defocusing. However, "this balance was never identified in numerical simulations" as it was claimed. Recently, it was determined[9] that a stable ionization-free filament with power in the range $P \sim 2$−10 of the critical for self-focusing, propagates with an intensity significantly lower than I ~ 10^{14} W/cm^2. Moreover, a stable filament with a weak plasma string of a few centimeters shows up a few meters away from the source, with power in the range $P \sim 10$−19 of the critical for self-focusing, with a stable spot (without self-focusing) and intensity in the range $I \sim 10^{10}$−10^{11} W/cm^2. So, why does the plasma string appear at such a low intensity of the laser pulse? The answer to this question is provided in ref 9, where it is suggested that this effect could be traced back to the emergence of a new type of collision ionization.

Despite the overwhelming number of papers on the standard classical model, the equations of paraxial nonlinear optics include terms accounting for tunnelling and multiphoton ionization, higher orders of nonlinearities ($\chi^{(5)}$, $\chi^{(7)}$, ...), Raman effects, and other effects. In general, the problem is not solvable analytically and thus the equations should be solved

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numerically with the aid of very powerful computers. Through parameters' tuning, the main aim is to obtain a relatively stable waveguide propagation of the filament. Let us point out that the first inconsistencies in these models are related to the fact that the paraxial equations assume narrow-band pulses, while the filament becomes broadband over a few diffraction lengths. This is one of the main reasons this approach has not demonstrated waveguide propagation, while in the experiments on a vertical trace the stable filament reaches a few kilometers. *Seemingly, there are some shortcomings manifested in the linear component of the model's amplitude equations. Additionally, there are discrepancies between the plasma-like interpretation of filamentation and the outcome of real experiments*. For instance, in experiments employing long-focus distance lenses aiming to avoid the nonlinear focus[10] and then in almost all leading laboratories a filamentation without ionization of the medium is observed. Another fundamental contradiction between experiments and the standard theory is that the measured intensity in the stable filament is on the order of 10^{11}−10^{12} W/cm^2, which is two to three orders lower than the intensity needed for defocusing by ionization. In all experiments on filamentation this effect is being observed when the power of the laser pulse is a bit higher than the critical one for self-focusing 5−10 GW. The critical power value for self-focusing is given by $P_{cr} = \pi(0.\,61\lambda_0)^2/(8n_0\, n_2)$, where n_0 and n_2 are the linear and nonlinear refractive indices, respectively, and λ_0 is the carrying wavelength of the laser pulse. Let us point out that this quantity also defines the intensity of the electric field for laser pulses. For pulses with spot diameters in the range of 100−200 μm, the intensity $I \approx 10^{12}$ W/cm^2. Recently,[9] experiments on the filamentation process of a 35 fs laser pulse showed that when the power reaches the value of 15 P_{cr}, a weak plasma column is observed with an intensity of the electric field below the critical one needed to trigger ionization. Thus, a new nonparaxial model of filamentation and a new type of collision ionization obtained from single broadband filaments in air were developed. The ensuing collision ionization may be traced back to trapping by polarization forces of neutral particles into the pulse envelope. As the density of trapped particles grows gradually, and as it reaches a critical value, their collision with the free ones leads to ionization of the medium.[11] The stable propagation of the laser pulse with a power in the range $P_{cr} < P_{pulse} < 20P_{cr}$ and the observation of very weak ionization patterns into the pulse during this process, lead to the following basic questions:

1 *What kinds of equations are suitable to describe both diffraction and dispersion of broad-band (few and phase-modulated 20 − 40) femtosecond pulses?*

2 *What kinds of nonlinear processes of narrow and broad-band pulses lead to asymmetrical spectrum broadening from the infrared spectral region to the visible one and the mechanism of filamentation?*

3 *What kinds of mechanisms are involved in merging and energy exchange between the filaments?*

This review attempts to correctly solve the diffraction problem raised in the first question, based on the accumulated theoretical and experimental results obtained over the last few decades.

2. LIMITS OF APPLICABILITY OF THE AMPLITUDE APPROXIMATION IN THE LINEAR AND NONLINEAR OPTICS

Typically, the electric field vector of a laser source is linearly polarized, i.e., ($\vec{E} = E_x\, \vec{x}$), and the nonlinear propagation of ultrashort optical pulses in isotropic materials is described generally by the scalar integro-differential nonlinear wave equation[12,13]

$$\Delta E - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\left\{ \int_0^\infty R^{(1)}(\tau)E(t-\tau, r)d\tau \right. \\ + \iiint_0^\infty R^{(3)}(\tau_1, \tau_2, \tau_3)\{[E(t-\tau_1, r)E^*(t-\tau_2, r)]E(t-\tau_3, r)\} \\ \left. d\tau_1 d\tau_2 d\tau_3 \right\}, \tag{1}$$

where $E(\mathrm{t},r) \equiv E(x, y, z, t)$ and $E^*(\mathrm{t},r) \equiv E^*(x, y, z, t)$ are the E_x component and its complex conjugate of the linearly polarized electrical field, $\Delta = \frac{\partial^2}{\partial^2 x^2} + \frac{\partial^2}{\partial^2 y^2} + \frac{\partial^2}{\partial^2 z^2}$ is the Laplace operator, $R^{(1)}$ and $R^{(3)}$ are, respectively, the linear and nonlinear response functions of the isotropic medium, and c is the vacuum light velocity. The methods for solving this complex integro-differential equation are based on the fact that the convolution integrals are performed only over time. Representing the electrical field as an amplitude envelope and a plane wave, i.e., $E(r,\, t) = A(r,\, t)\ e^{-i(k_0 z - \omega_0 t)} + c.c$, where k_0 and ω_0 are the carrying wavenumber and frequency of the laser electric field, equation (1) transforms into

$$\Delta A(r, t) - 2ik_0\frac{\partial A(r, t)}{\partial z} - k_0^2 A(r, t) \\ = -\int_{-\infty}^{\infty} k^2(\omega)\hat{A}(r, \omega - \omega_0)\mathrm{e}^{-i(\omega-\omega_0)t} d(\omega - \omega_0), \tag{2}$$

where $\hat{A}$ is the Fourier transform in time of the amplitude A. In the integrand in the Fourier-like integral on the right-hand side of equation (2) only the linear wave vector function $k_{\mathrm{lin}}^2(\omega) = \omega^2\, \varepsilon(\omega)\,/c^2$ [$\varepsilon(\omega)$ is the dispersion of the media] depends on the frequency ω. All other functions depend on the frequency difference $\omega - \omega_0$. The transformation of this linear integro-differential equation to the usual differential one can be obtained after performing a series expansions of the square of the linear wavenumber $k^2(\omega)$ around the carrying frequency of the laser pulses ω_0 yielding

$$\Delta A(r, t) - 2ik_0\frac{\partial A(r, t)}{\partial z} - k_0^2 A(r, t) \\ = -\left[k_0^2 A - i\left(\frac{\partial k^2}{\partial \omega}\right)_{\omega_0}\frac{\partial A}{\partial t} - \frac{1}{2}\left(\frac{\partial^2 k^2}{\partial \omega^2}\right)_{\omega_0}\frac{\partial^2 A}{\partial t^2} \right. \\ \left. + \sum_{m=3}^{\infty}\frac{(-i)^m}{m!}\left(\frac{\partial^m k^2}{\partial \omega^m}\right)_{\omega_0}\frac{\partial^m A(r, t)}{\partial t^m} \right]. \tag{3}$$

This is the so-called approximation of slowly varying amplitude, which assumes that equation (3) is valid for narrow-band pulses with a significant number of cycles under the envelope.[12] Hereafter, we will show that actually, such an expansion is correct up to single-cycle pulses (broad-band), if the differential operator series in the brackets of the right-hand side of equation (3) is *strongly convergent*. The convergence of the series gives the possibility to approximate it and thus for solving the differential equation it may be usually cut off to the second order of the linear dispersion.

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Table 1. Results for the Dispersion Expansion up to the 5th Order for $N = t_0/T_0 = 100$[a]

β_0	β_1	β_2	β_3	β_4	β_5
6.1×10^9	9.8×10^6	3.9×10^3	6.38×10^{-5}	1.4×10^{-8}	9.2×10^{-13}

[a]That is, for a 266 fs pulse (t_0 = 266 fs) at wavelength λ = 800 nm and optical period $T_0 \approx 2.66 \times 10^{-15}$ s.

Table 2. Values of the 1st to the 5th Components of the Taylor Series (11) for Spectrally-Broad 5.3 fs Pulse (n = 2)[a]

β_0	β_1	β_2	β_3	β_4	β_5
6.17×10^9	4.9×10^{8}	9.7×10^{6}	7.9	9.01×10^{-2}	2.8×10^{-4}

[a]The series is also strongly convergent. The ratio of the third to the second orders term of dispersion β_3/β_2 is of the order of 10^{-6}.

To make a quantitative analysis of the series convergence, we normalize in time the amplitude function and its derivatives in equation (3). The normalized amplitude of a localized in time pulse can be written as

$$A = A_0 A'(x, y, z, t/t_0), \tag{4}$$

where A_0 is the normalizing amplitude, t_0 denotes the initial temporal duration of the pulse, $A' \to 0$ at the limits $t \to \pm\infty$, and $\max A' = 1$. Substituting expression (4) in the Taylor series on the right-hand side of equation (3), we obtain a product of normalized functional series and numerical Taylor series of the linear polarization operator

$$k_0^2 A' - i\left(\frac{\partial k^2}{\partial \omega}\right)_{\omega_0} \frac{1}{t_0}\frac{\partial A'}{\partial t} - \frac{1}{2}\left(\frac{\partial^2 k^2}{\partial \omega^2}\right)_{\omega_0} \frac{1}{t_0^2}\frac{\partial^2 A'}{\partial t^2} + \sum_{m=3}^{\infty} \frac{(-i)^m}{m!}\left(\frac{\partial^m k^2}{\partial \omega^m}\right)_{\omega_0} \frac{1}{t_0^m}\frac{\partial^m A'}{\partial t^m}. \tag{5}$$

The normalization of the amplitude transforms the functional series (5) into a series of the derivatives of the normalized localized function (distribution)

$$f'(A') = A', \frac{\partial A'}{\partial t}, \frac{\partial^2 A'}{\partial t^2}, ..., \frac{\partial^m A'}{\partial t^m}, \propto f'(A)'^0, f'(A)'^1, f'(A)'^2, ..., f'(A)'^m \,, \tag{6}$$

where superscripts designate the order of the derivatives. Alternatively, the normalized numerical Taylor series of the wavenumber square in equation (5) takes the form

$$g'(k^2) = k_0^2; \left(\frac{\partial k_0^2}{\partial \omega}\right)_{\omega_0} \frac{1}{t_0}; \frac{1}{2}\left(\frac{\partial^2 k_0^2}{\partial \omega^2}\right)_{\omega_0} \frac{1}{t_0^2}; \frac{1}{3!}\left(\frac{\partial^3 k_0^2}{\partial \omega^3}\right)_{\omega_0} \frac{1}{t_0^3}; ...; \frac{1}{m!}\left(\frac{\partial^m k^2}{\partial \omega^m}\right)_{\omega_0} \frac{1}{t_0^m} + \cdots, \propto g'(k^2)^0; g'(k^2)^1; g'(k^2)^2; g'(k^2)^3; ...; g'(k^2)^m; \cdots, \tag{7}$$

where the superscripts denote the corresponding derivatives. The Taylor series of the wavenumber square expansion (7) around the carrier frequency turns into a series, for which the dimensionality of each term is equal to the dimensionality of the first term—the square of the wavenumber (cm^{-2}). If expressed in terms of the functional (6) and the numerical series (7), then the right-hand side of equation (5) becomes

$$R(A, k^2) = A_0(g'^0 f'^0 - ig'^1 f'^1 - g'^2 f'^2 + \cdots + (-i)^m g'^m f'^m + \cdots). \tag{8}$$

The maximal value of the function f'^0 in the first term of the function series (8) is 1, and the maximal values of the derivatives f'^m are always less than one. This is a typical property of the normalized distribution functions. Each term of the series (8) is a product of a numerical Taylor series (7) and a normalized *majorant* series of a distribution function and its derivatives (6). For the series (8), we choose the *maximal values* of the function terms in equation (6). Thus, the series (8) turns into a numerical series of the type

$$P = A_0\left(g'^0 - \frac{i}{2}g'^1 - \frac{1}{2^2}g'^2 + \cdots + \frac{(-i)^m}{2^m}g'^m + \cdots\right). \tag{9}$$

Thus, we apply the following lemma: *a sufficient condition for the convergence of the series of the linear polarization operator (5) is that of the convergence of the numerical series*

$$P_m(k^2, t_0) = \frac{(-i)^m}{m!}\frac{\partial^m k^2}{\partial \omega^m}\bigg|_{\omega=\omega_0} \frac{1}{t_0^m}\frac{1}{2^m}, \; m = 0, \cdots, \infty. \tag{10}$$

The properties of the series $P_m(k^2, t_0)$ will be analyzed for optical pulses of wavelength λ = 800 nm propagating in air and having distinct temporal durations t_0. More specifically, we will express the different pulse widths t_0 in terms of the number of oscillations of the carrier frequency at a level e^{-1} from the pulse maximum $t_0 = N\, T_0$ with optical period $T_0 = n_0\lambda_0/c$. For a wavelength λ = 800 nm, we obtain $T_0 \approx 2.6 \times 10^{-15}$ s. Moreover, the Ciddor formula[14] has been applied to compute the dielectric constant $\varepsilon(\omega)$, the square of the wavenumber and its derivatives in equation (10), whose convergence will be studied by varying the number N from N = 100 (the case of slowly varying amplitudes) to $N \sim 2$ (two optical cycle pulse) for the dispersion expansion up to the 5th order. Let us use the following notation for the mth component of the Taylor series:

$$\beta_0 = k_0^2; \beta_m = P_m = \frac{1}{m!}\left(\frac{\partial^m k^2}{\partial \omega^m}\right)_{\omega_0} \frac{1}{t_0^m}\frac{1}{2^m}. \tag{11}$$

The values of the numerical expansion (11) for a spectrally narrow pulse (t_0 = 266 fs and N = 100) are presented in Table 1.

The value of the ratio β_3/β_2, i.e., the third to the second series term (11) is of the order of 10^{-7}. Thus, truncating the expansion in equation (3) at the second order term of the dispersion is sufficient for describing the dynamics of a narrow-band wave packet propagating in air.

Now, we turn our attention to the convergence of the series (11) for broad-band pulses. Table 2 displays the results for the series truncated to the 5th order term for a spectrally broad pulse with two cycles (N = 2 and t_0 = 5.3 fs). These estimates show that for a gas medium with weak dispersion, truncation of the expansion in equation (3) at the second order term is sufficient for the description of pulses with even one or two optical cycles under the envelope. It is worth mentioning that it can be easily seen from the data in Tables 1 and 2 that the approximation used for the dispersion curves is close to a quadratic one for the amplitude function.

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In addition, we perform similar calculations for fused silica, where the dispersion $\varepsilon(\omega)$ was calculated using the Sellmeier dispersion formula. The series is also strongly convergent for spectrally broad 5.3 fs, but the ratio β_3/β_2 of the third to the second dispersion term is now 10^{-3}. As it can be seen, for fused silica, it is also possible to truncate the expansion in equation (3) to the second order, but the third order dispersion can play the role of a small parameter, even in the spectral region where $\beta_2 \neq 0$.

To sum up, the above analysis of the series expansion of the linear polarization (3) suggests that it is possible to employ a linear amplitude approximation to describe the dynamics of broad-band optical pulses with durations up to one optical cycle, using the expansion (3) to the second order in air, and including the third dispersion order as a small parameter in fused silica.

In general, the nonlinear response time function $R^{(3)}$ is shorter than the linear one $R^{(1)}$. As it is shown in ref 13, this corresponds to a nonlinear response time $\tau_{\text{nlin}} \approx 100$, which is much shorter than the linear response time $\tau_{\text{lin}} \approx 2.5$ fs. This is yet an additional reason to use the nonlinear amplitude approximation to describe the dynamics of broad-band optical pulses with durations down to one optical cycle, using the second-order amplitude approximation in air or the third dispersion order in fused silica.

3. LINEAR AMPLITUDE EQUATION UP TO FIRST ORDER OF THE DISPERSION: DIFFRACTION

The linear amplitude equation (3) up to the second order of the group-velocity dispersion can be written as

$$2ik_0\left[\frac{\partial A}{\partial z} + \frac{1}{v_{\text{gr}}}\frac{\partial A}{\partial t}\right] = \Delta_\perp A - \frac{\beta}{v_{\text{gr}}^2}\frac{\partial^2 A}{\partial t^2} + \frac{\partial^2 A}{\partial z^2} - \frac{1}{v_{\text{gr}}^2}\frac{\partial^2 A}{\partial t^2}, \tag{12}$$

where $\Delta_\perp = \partial^2/\partial x^2 + \partial^2/\partial y^2$ designates the transverse Laplace operator, $A(x, y, z, t)$ is the amplitude envelope, $\beta = k_0 k'' v_{\text{gr}}^2$ is the dimensionless group velocity dispersion parameter, k_0 is the carrying wavenumber, v_{gr} is the group velocity, and k'' is the group velocity dispersion. In some applications, such as generation of high harmonics with fs pulses, construction of petawatt laser systems etc., when vacuum tubes are used, due to the significant reduction of atmospheric gases, dispersion effects are absent. For this type of investigation, the group velocity dispersion may be neglected and the amplitude equation (12) to the first order of the dispersion, group velocity, takes the form

$$2ik_0\left(\frac{\partial A}{\partial z} + \frac{\partial A}{\partial t}\right) = \Delta_\perp A + \frac{\partial^2 A}{\partial z^2} - \frac{1}{v_{\text{gr}}^2}\frac{\partial^2 A}{\partial t^2}. \tag{13}$$

Equation (13) is actually equivalent to the wave equation

$$\Delta_\perp E + \frac{\partial^2 E}{\partial z^2} - \frac{1}{v_{\text{gr}}^2}\frac{\partial^2 E}{\partial t^2} = 0, \tag{14}$$

when the electrical field is represented as

$$E(x, y, z, t) = A(x, y, z, t)\exp[ik_0(z - v_{\text{gr}}t)], \tag{15}$$

So, by solving the problem of dispersion-free propagation of an optical pulse, we simultaneously solve the wave equation for propagation in a medium with group velocity equal to the phase velocity, without accounting for the dispersion. The solution of the wave equation (14) will be obtained from the solution of the amplitude equation (13) after multiplication by the carrier frequency and wavenumber (15). Using the method of the amplitude envelope (15), a number of analytical solutions, with finite energy, of the linear wave equation (14) were obtained in ref 15.

The theory in the standard paraxial optics is written in local time coordinates, $z = z$ and $\tau = t - z/v_{\text{gr}}$, thus we may apply the analysis in the same coordinate system. The amplitude equation (13) in local time coordinates takes the form

$$2ik_0\frac{\partial A}{\partial z} = \Delta A_\perp + \frac{\partial^2 A}{\partial z^2} - \frac{2}{v_{\text{gr}}}\frac{\partial^2 A}{\partial \tau \partial z}. \tag{16}$$

We will apply a three-dimensional Fourier transform to the amplitude function (16) of the kind: $\hat{A}(k_x, k_y, \Delta\omega/v_{\text{gr}}, \text{z}) = \text{F}[A(x, y, \tau, z)]$, where the function F stands for the three-dimensional Fourier transform in the (x, y, τ) coordinates. In the momentum $(k_x, k_y, \Delta\omega/v_{\text{gr}})$ Fourier space, equation (16) transforms into the one-dimensional second order ordinary differential equation of the kind:

$$2i\left(k_0 - \frac{\Delta\omega}{v_{\text{gr}}}\right)\frac{\partial \hat{A}}{\partial z} = -(k_x^2 + k_y^2)\hat{A} + \frac{\partial^2 \hat{A}}{\partial z^2}, \tag{17}$$

where $\Delta\omega = \omega - \omega_0$; $\Delta k_z = \Delta\omega/v_{\text{gr}}$ are the frequency and wavenumber spectral components of the laser pulse, respectively. The fundamental solution of equation (17) reads

$$\hat{A}(k_x, k_y, \Delta\omega, z) = \hat{A}(k_x, k_y, \Delta\omega, 0) \exp\left\{i\left[\left(k_0 - \frac{\Delta\omega}{v_{\text{gr}}}\right) \pm \sqrt{\left(k_0 - \frac{\Delta\omega}{v_{\text{gr}}}\right)^2 - (k_x^2 + k_y^2)}\right]z\right\}. \tag{18}$$

The solution of the amplitude equation in real space can be obtained by analytical or numerical solution of the inverse Fourier transform

$$A(x, y, \tau, z) = FFF^{-1}\left\langle \hat{A}(k_x, k_y, \Delta\omega, 0) \exp\left\{i\left[\left(k_0 - \frac{\Delta\omega}{v_{\text{gr}}}\right) \pm \sqrt{\left(k_0 - \frac{\Delta\omega}{v_{\text{gr}}}\right)^2 - (k_x^2 + k_y^2)}\right]z\right\}\right\rangle, \tag{19}$$

where $v_{\text{gr}} = c$ in vacuum. Depending on the relation between the wavenumber k_0 and spectral width on the level of the e^{-1} from the maximum of the initial pulse $\Delta k_z^0 = \Delta\omega_0/v_{\text{gr}}$, we determine three different domains and kind of the pulses:

(a) Narrow-band pulses when the spectral width is much smaller than the carrying wavenumber.

$$\Delta k_z^0 = \Delta\omega_0/v_{\text{gr}} \ll k_0$$

(b) Relatively narrow-band pulses with spectral width smaller than the carrying wavenumber.

$$\Delta k_z^0 = \Delta\omega_0/v_{\text{gr}} < k_0$$

(c) Broad-band laser pulses having a spectral width of the order of the carrying wavenumber.

$$\Delta k_z^0 = \Delta\omega_0/v_{\text{gr}} \approx k_0$$

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Hereafter, we will perform the analysis of the fundamental solution (18) in these three cases.

4. NARROW-BAND LASER PULSES ($\Delta K_Z^0 = \Delta\omega_0/V_{GR} \ll K_0$): PROPAGATION DISTANCES, WHERE THE FRESNEL'S TYPE DIFFRACTION WORKS

Spectrally limited laser pulses with time duration from nanoseconds up to $\Delta t > 50$−100 fs, in the near-infrared region (800 nm) satisfies the relation for narrow- band pulses $\Delta k_z^0 = \Delta\omega_0/v_{gr} \ll k_0$.

In this case, as the wavenumber k_0 is significantly larger than the spectral width Δk_z^0, thus we can use the approximation $k_0 - \frac{\Delta\omega}{v_{gr}} \approx k_0$ in equation (18) to obtain

$$\hat{A}(k_x, k_y, z) = \hat{A}(k_x, k_y, 0)\exp\left\{i\left[k_0 \pm k_0\sqrt{1 - \frac{(k_x^2 + k_y^2)}{k_0^2}}\right]z\right\}. \tag{20}$$

Alternatively, the square of the wavenumber k_0^2 is significantly larger than the square of the transverse wave numbers or the laser pulse k_x^2, k_y^2, $(k_x^2 + k_y^2)/k_0^2 \ll 1$. Whence, we can use correctly the Tailor series of the square root as the series is rapidly converging

$$\begin{aligned}&\left[k_0 - k_0\left(1 - \frac{k_x^2 + k_y^2}{k_0^2}\right)^{1/2}\right]z\\ &= \left[k_0 - k_0\left(1 + \frac{1}{2}\frac{k_x^2 + k_y^2}{k_0^2} - \frac{1}{8}\left(\frac{k_x^2 + k_y^2}{k_0^2}\right)^2 + \cdots\right)\right]z\\ &= -\frac{1}{2}\left(\frac{k_x^2 + k_y^2}{k_0}\right)z + \frac{1}{8}\left(\frac{k_x^2 + k_y^2}{k_0^{3/2}}\right)^2 z + \cdots.\end{aligned} \tag{21}$$

In the case of narrow-band pulses, having in mind the linear order term from equation (21), we obtain

$$\hat{A}(k_x, k_y, z) = \hat{A}(k_x, k_y, 0)\exp\left\{-i\left[\frac{k_x^2 + k_y^2}{2k_0}\right]z\right\}. \tag{22}$$

This is the typical Fresnel's spectral kernel and it is equal to the spectral kernel of a laser in CW regime. Equation (22) is solution in the Fourier space of the well-known Leontovich equation:[16]

$$2ik_0\frac{\partial A}{\partial z} = \Delta A_\perp. \tag{23}$$

As it can be seen, for narrow-band pulses in the diffraction regime, that the transverse wave numbers increase rapidly with the propagation distance $(k_x^2 + k_y^2)\,z/2k_0$, where k_0 is a constant. *It must be pointed out that the Fresnel's diffraction is valid in a domain, where the transverse spectra $k_\perp(z) = (k_x^2 + k_y^2)\,z$ is much smaller than the square of the wavenumber k_0,* i.e.

$$\frac{1}{2k_0}k_\perp(z) = \frac{z}{2k_0}(k_x^2 + k_y^2) \ll 1. \tag{24}$$

As it can be seen from equation (24), the transverse spectra $k_\perp(z)$ grows linearly with the propagation distance z. Thus, at some fixed distance from the source z_0 the series (21) will not converge faster and thus will turn into a domain, where the higher order terms of the Tailor series should be accounted for. From the mathematical point of view, a series is still rapidly converging when the second term is $o^{(-1)} = 0.1$ and the third term is $o^{(-2)} = 0.01$ with respect to unity, i.e

$$\begin{aligned}&1 > (o^{-1})\frac{1}{2}\frac{k_x^2 + k_y^2}{k_0}z > (o^{-2})\frac{1}{8}\left(\frac{k_x^2 + k_y^2}{k_0}\right)^2 z \text{ or } 1 > 0.1 \times \frac{1}{2}\frac{k_x^2 + k_y^2}{k_0}z\\ &> 0.01 \times \frac{1}{8}\left(\frac{k_x^2 + k_y^2}{k_0}\right)^2 z.\end{aligned} \tag{25}$$

Thus, we have the following definition: *The paraxial approximation for laser pulses is valid over a distance, where the Taylor series (21) is strongly converging. This condition is fulfilled from the mathematical point of view when the relations (25) are satisfied.*

This definition, along with the relations (25) give us one *quantitative criteria* for determining the propagation distance, where we can apply the Fresnel's approximation. From equation (25) it follows that this distance must obey

$$z_0^{\text{Fresnel}} < \frac{1}{0.05}\frac{k_0}{k_x^2 + k_y^2}. \tag{26}$$

To determine the correct Fresnel's propagation distance z_0^{Fresnel} of a laser pulse, we will analyze equation (26) in two basic cases depending on the initial length of the laser pulse compared to e^{-1} from the maximum, namely (i) $\Delta x_0 = \Delta y_0 = 10^{-1}$ cm and (ii) $\Delta x_0 = \Delta y_0 = 10^{-2}$ cm. The calculation of the transverse spectra in both cases is based on the relations between the spectral width and the spatial dimension of a spectrally limited Gaussian pulse, i.e., $\left(k_x = k_y = \frac{2}{\Delta x_0} = \frac{2}{\Delta y_0}\right)$. Thus, in both cases the initial pulse spectra satisfies the condition $(k_x^2 + k_y^2)/k_0^2 \ll 1$, that is using only the first term in the Taylor series (Fresnel's approximation) is correct.

The Fresnel's propagation distance z_0^{Fresnel} obtained from equation (26) for both cases takes the values $z_0^{\text{Fresnel}} = 20$ m and $z_0^{\text{Fresnel}} = 20$ cm for (i) and (ii), respectively. In the first case, the diffraction length is $z_{\text{diffr}} = k_0(\Delta x^2 + \Delta y^2) \approx 15.7$ m and nearly up to one and half of diffraction length, we can expect Fresnel's type diffraction without influence of higher terms in the Taylor series (21). While, in the second case, the diffraction length is $z_{\text{diffr}} = k_0(\Delta x^2 + \Delta y^2) \approx 15.7$ cm and again up to one and half of diffraction lengths we can expect the appearance of a Fresnel's type diffraction.

There is a so-called "grey zone" at distances higher than a few diffraction lengths, where again the diffraction is still in a plane, orthogonal of the direction of propagation, but the higher order term in the Taylor series (21) must be kept in mind. This is due to the fact that the series (21) in these domains become slowly convergent.

Contemporary petawatt laser facilities use vacuum tubes for laser pulses propagation up to hundred meters to reach the investigated objects. In vacuum, due to the negligible non-linearity, the main physical mechanism leading to the transformation of femtosecond laser pulses is diffraction. Thus, for correct engineering of the tubes one must bear in mind the results of this paragraph, stating that Fresnel diffraction takes place even at narrow band pulses of one/two diffraction length(s) only.

5. RELATIVELY NARROW-BAND PULSES $\Delta K_Z^0 = \Delta\omega_0/V_{GR} < K_0$: REDUCTION OF THE DIFFRACTION LENGTH AND THE FRESNEL'S ZONE

Spectrally limited laser pulses in the near-infrared region, with time duration in the range $10 < \Delta t < 50$ fs, as well as phase modulated fs pulses, satisfy the condition $\Delta k_z^0 = \Delta\omega_0/v_{gr} < k_0$. In

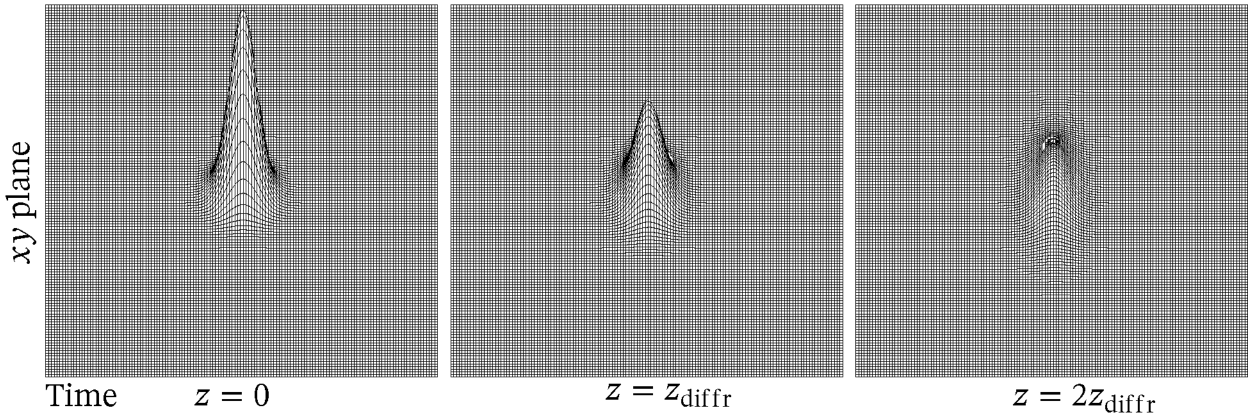

Figure 1. Side projection of the Intensity profile of a Gaussian pulse with time duration 20 fs, as solution of equation (32), at a distance of two reduced diffraction lengths $z_{\mathrm{diffr}} = k\tilde{\sigma}_0^2 = (k_0 - \Delta\omega_0/v_{\mathrm{gr}})\,\sigma_0^2$ at $z = 0$ (left panel), $z = z_{\mathrm{diffr}}$(middle panel) and $z = 2z_{\mathrm{diffr}}$(right panel). As it can be expected, the diffraction picture is of a Fresnel's type, with enlargement of the pulse in the plane, orthogonal to the propagation direction.

this case in the spectral kernel of equation (18), the difference $k_0 - \frac{\Delta\omega}{v_{\mathrm{gr}}} = k_0 - \Delta k_z$ cannot be replaced by k_0 and we will look for a solution of equation (18) for the translated wavenumber's $\tilde{k}_z = k_0 - \frac{\Delta\omega}{v_{\mathrm{gr}}}$. With respect to the transverse spectra, we still have the condition

$$(k_x^2 + k_y^2)/(k_0 - \Delta\omega/v_{\mathrm{gr}})^2 = (k_x^2 + k_y^2)/\tilde{k}^2 \ll 1. \tag{27}$$

Therefore, we can expand the spectral kernel in equation (18) in a Taylor series and restrict ourselves to the first term in the expansion

$$\left[(k_0 - \Delta\omega/v_{\mathrm{gr}}) - (k_0 - \Delta\omega/v_{\mathrm{gr}})\left(1 + \frac{k_x^2 + k_y^2}{(k_0 - \Delta\omega/v_{\mathrm{gr}})^2}\right)^{1/2}\right]z = \left(\frac{1}{2}\frac{k_x^2 + k_y^2}{(k_0 - \Delta\omega/v_{\mathrm{gr}})} - \cdots\right)z. \tag{28}$$

The solution (18), keeping in mind the Taylor expansion (28), for such kind of pulses transforms into

$$\hat{A}(k_x, k_y, \Delta\omega/v_{\mathrm{gr}}, z) = \hat{A}(k_x, k_y, \Delta\omega/v_{\mathrm{gr}}, 0)\exp\left\{-i\left[\frac{k_x^2 + k_y^2}{2k_0(1 - \Delta k/k_0)}\right]z\right\}. \tag{29}$$

As $\Delta k/k_0 < 1$, we can use the series $(1 - \Delta k/k_0)^{-1} \approx 1 + \Delta k/k_0 + (\Delta k/k_0)^2 + \cdots$, and the solution (29) becomes

$$\hat{A}(k_x, k_y, \Delta\omega/v_{\mathrm{gr}}, z) = \hat{A}(k_x, k_y, \Delta\omega/v_{\mathrm{gr}}, 0)\exp\left\{-i\left[\frac{k_x^2 + k_y^2}{2k_0}((1 + \Delta k_z/k_0 + \ldots))\right]z\right\}. \tag{30}$$

As it can be seen from equation (30) in the first approximation again we have a Fresnel's spectral kernel. In the second approximation, as there is a linear dependence on $\Delta\omega/v_{\mathrm{gr}} = \Delta k_z$, the pulse grows very weakly in the z direction, but the deformation is still in the plane orthogonal to the direction of propagation. This weak enhancement in the z direction leads to a weak decrease of the diffraction length

$$z_{\mathrm{diffr}} = \tilde{k}\sigma_0^2 = \left(k_0 - \frac{\Delta\omega_0}{v_{\mathrm{gr}}}\right)\sigma_0^2. \tag{31}$$

It is not difficult to show that the spectral kernel in equation (29), corresponds to the Akhmanov-Brabec-Boyd (ABB) approximation[13,17] of the amplitude equation (16) in dispesionless media after removal of the second derivative in the z direction from equation (16). Then, we end up with

$$2ik_0\frac{\partial A}{\partial z} = \Delta A_{\perp} - \frac{2}{v_{\mathrm{gr}}}\frac{\partial^2 A}{\partial\tau\partial z}. \tag{32}$$

The main difference of (32) from the Leontovich one (23) is that in the solution (29) of equation (32) the spectral width of the laser pulse is taken into account via the translated wavenumber $\tilde{k} = k_0 - \Delta k_z$. However, the reduction of the wavenumber leads to a reduction of the length of the Fresnel's zone too. For instance, the Fresnel's domain shrinks from 20 to 18.5 m for a 20 fs pulse. In the case of pulses with time duration greater than 100 fs, the reduction of the Fresnel's zone is negligible. In Figure 1, we present the side projection of the evolution of a Gaussian pulse with time duration 20 fs, as a solution of equation (32), with spectral kernel (29) at a distance of two reduced diffraction lengths. As it can be expected, the diffraction picture again is of a Fresnel's type, with enlargement of the pulse in the plane, orthogonal to the propagation direction.

The typical petawat laser system produces femtosecond laser pulses solely in the time region $10 < \Delta t < 50$ fs. That is why in vacuum tubes the ABB equation (32) determines the Fresnel zone more accurately. Thus, for engineering these tubes, instead the Leontovich equation (23), calculations must be performed with the aid of ABB (32). In this regime a limitation up to one/two reduced diffraction length(s) holds also, where a similar to the Fresnel diffraction enlarging of the laser pulse in a plane, orthogonal to its propagation direction, can be observed.

As mentioned in the above two paragraphs, for engineering laser facilities with spectrally limited laser pulses with time durations from nanoseconds up to 10 fs, as well as for the transportation of these pulses by vacuum tubes, the calculation of Fresnel type diffraction by equations (23) or (32) must be restricted up to one/two diffraction length(s), where the Taylor expansions (21) and (28) are strongly convergent.

It is natural that the following main question raises: What equation describes pulse propagation on arbitrary distances up to a hundred diffraction lengths? The answer will be given hereafter.

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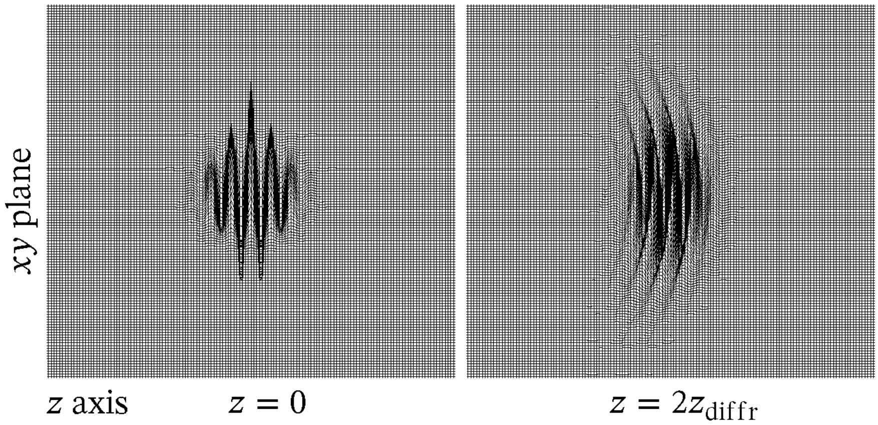

Figure 2. Side projection of the evolution in vacuum of the electrical field of a broad-band Gaussian pulse with time duration 6 fs (λ_0 = 800 nm), as numerical solution of equation (33). Left panel ($z = 0$) and right panel ($z = 2z_{\text{diffr}}$).

6. DIFFRACTION OF BROAD-BAND PULSES ($\Delta K_Z^0 = \Delta\omega_0/V_{GR} \approx K_0$): SOLVING THE AMPLITUDE EQ (13) IN GALILEAN FRAME WITHOUT APPROXIMATIONS: $\lambda^{(3)}$ TYPE DIFFRACTION

The analysis provided above shows that paraxial optics is applicable only at a few diffraction lengths for narrow-band optical pulses. The ABB modified equation (32) determines the paraxial diffraction in a plane, orthogonal of the direction of propagation, on a reduced distance and with reduced diffraction length. Diffraction of broad-band pulses is quite different from narrow band ones and it is not in a plane, orthogonal to the propagation direction. For the first time, laser pulses with time durations of 4–6 fs in the frame of a wave equation were investigated numerically by Christov,[18] who obtained an unexpected parabolic deformation of the intensity profile, outside of the plane, which characterizes the Fresnel diffraction. Later, by numerically solving the Maxwell's equation in a vacuum for attosecond pulses, (which is equal to analyzing the same wave equation), such parabolic deformation of the pulse was obtained by other authors.[19] This type of diffraction with parabolic deformation of the intensity profile was dubbed $\lambda^{(3)}$ diffraction. A few years later the authors of ref 20 analytically solved the wave equation (13) for an initial Gaussian pulse. It was shown that depending on the number of cycles inside the pulse, there are different kinds of diffractions. For pulses with 2–4 cycles inside the pulse, as well as for phase modulated broadband ones,[21] at two-three diffraction lengths, typical $\lambda^{(3)}$ diffraction was obtained. While the solution for narrowband ones (with more than 5–10 cycles inside the pulse), at the same distances, only Fresnel's diffraction can be seen. We determine one important dependence: with decreasing of the spectral width of the pulse from broad-band to narrow-band, the $\lambda^{(3)}$ diffraction shows up at long distances from the source, while at a few diffraction lengths the solution is typical to Fresnel's type one.

To see the actual three-dimensional deformation of a laser pulse due to diffraction, as well as to create a Fourier code for numerically or analytically solving the amplitude equation (13), it is more convenient to transform it in Galilean coordinates. The linear amplitude equation (13) in Galilean coordinates ($z = z - v_{\text{gr}}t$; $t = t$), when the second order of the group velocity dispersion is neglected $\beta = k_0 v_{\text{gr}}^2 k'' \ll 1$, takes the form

$$\frac{2ik_0}{v_{\text{gr}}}\frac{\partial A}{\partial t} = \Delta_\perp A - \frac{1}{v_{\text{gr}}^2}\left(\frac{\partial^2 A}{\partial t^2} - 2v_{\text{gr}}\frac{\partial^2 A}{\partial t \partial \zeta}\right). \tag{33}$$

The amplitude equation (33) is equal to the wave equation but this time written in Galilean coordinates

$$\Delta_\perp E - \frac{1}{v_{\text{gr}}^2}\left(\frac{\partial^2 E}{\partial t^2} - 2v_{\text{gr}}\frac{\partial^2 E}{\partial t \partial z}\right) = 0. \tag{34}$$

when the electric field is presented as

$$E(x, y, z, t) = A(x, y, z, t)\exp[-ik_0 z]. \tag{35}$$

As mentioned in Section 5, the Leontovich and ABB equations are valid up to one/two diffraction length(s), while the amplitude equations (13) and (33) are obtained using the convergence of the series equation (9) and are correct over a range from single cycle pulses up to ns regime. Therefore, these equations are valid on the propagation distance of the laser pulse.[20]

To solve equation (33), we proceed by applying a three-dimensional Fourier transform to the amplitude function of the kind: $\hat{A}(k_x, k_y, k_z - k_0, t) = F[A(x, y, z, t)]$, where F is the three-dimensional Fourier transform in the (x, y, z) space coordinates. In the $(k_x, k_y, k_z - k_0, t)$ Fourier space equation (33) transforms into the one-dimensional second order ordinary differential equation of the kind

$$2i\frac{(k_0 - \Delta k_z)}{v_{\text{gr}}}\frac{\partial \hat{A}}{\partial t} + \frac{1}{v_{\text{gr}}^2}\frac{\partial^2 \hat{A}}{\partial t^2} + (k_x^2 + k_y^2)\hat{A} = 0. \tag{36}$$

where $\Delta k_z = k_z - k_0 = \Delta\omega/v_{\text{gr}}$ is wavenumber's spectral variable of the laser pulse. The fundamental solution of equation (36) reads

$$\hat{A}(k_x, k_y, \Delta k_z, t) = \hat{A}(k_x, k_y, \Delta k_z, 0) \exp\left\{i(k_0 - \Delta k_z)\left[1 - \sqrt{1 + \frac{(k_x^2 + k_y^2)}{(k_0 - \Delta k_z)^2}}\right]v_{\text{gr}}t\right\}. \tag{37}$$

When we investigate broad band pulses, as $\Delta k_z^{\text{falf-max}} \approx k_0$, the relation $(k_x^2 + k_y^2)/(k_0 - \Delta k_z)^2$ is no longer a small parameter and a Taylor expansion, as it was performed in the previous cases of narrow band pulses, is not possible. Thus, we solve equation (37) without the use of approximations for the initial Gaussian pulse. The solution of the amplitude equation in the real space

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here as well can be obtained by analytical or numerical solution of the inverse Fourier transform

$$A(x, y, z, t) = FFF^{-1}\left\langle \hat{A}(k_x, k_y, \Delta k_z, 0) \exp\left\{ i(k_0 - \Delta k_z)\left[1 - \sqrt{1 + \frac{(k_x^2 + k_y^2\}}{(k_0 - \Delta k_z)^2}}\right] v_{gr}t) \right\} \right\rangle. \quad (38)$$

The expression under the square root is always positive. Thus, the numerical inverse Fourier transform in real space may be obtained always.

Using equation (38) we can solve equation (33) numerically. The ensuing results showing the behavior of the amplitude of electrical field in vacuum are depicted on Figure 2. Notice that at a distance of two diffraction lengths, a typical $\lambda^{(3)}$ diffraction shows up. By increasing the time duration of the pulse (decreasing its spectral width), the $\lambda^{(3)}$ diffraction is shifted in the faraway domain with respect to the source. It is worth mentioning that the numerical result repoduces exactly the analytical solution obtained in ref 20.

7. CONCLUSIONS

The popular models describing laser pulse propagation in transparent media usually use a paraxial approximation in the linear component and do not take into account that the named approximation is limited on the propagation distance. Alternatively, as it can be seen from the above analysis, in the femtosecond domain the pulse can be easily transformed from narrow-band to broad-band.

The evolution dynamics and the diffraction regime of these two types of pulses are quite different. While the narrow-band pulses diffract following Fresnel's type diffraction law, at a distance up to few diffraction lengths, broad band ones diffract in the $\lambda^{(3)}$ regime at the same distance. The analytical and numerical investigations of laser pulse propagation governed by the amplitude equations (13)−(33) show that by increasing the time duration of the broad-band pulses (a decrease of its spectral width), and then transforming them to narrow-band ones, the $\lambda^{(3)}$ diffraction is shifted in the faraway domain with respect to the laser source. To conclude, the *nonparaxial* amplitude equation (13) and its representation in Galilean frame (33) correctly describe the pulse diffraction without restriction on the distance and the time duration of the pulses. Thus, in future papers we foresee to investigate nonparaxial type equations taking the dispersion and nonlinear effects into consideration.

It is important to point out that the $\lambda^{(3)}$ type diffraction plays a key role in the production of petawatt laser systems, as well as for creating electron mirrors for these systems. When the spot size of the pulse is on the order of a few optical periods and the duration is less than 10 fs, the pulses with millijoule energy can produce intensities above 10^{18} W/cm^2.[22] These types of lasers were dubbed $\lambda^{(3)}$ type lasers since at one/two diffraction length(s) the pulses diffract in the $\lambda^{(3)}$ regime. The standard optics fails at such intensities and the $\lambda^{(3)}$ type electron e-bunches can be used for electron mirror manufacturing.[19] Thus, the following question arises: *Is it possible for this process to be managed and for electron mirrors with different focal planes to be produced?* In ref 21, we answer positively to this question, suggesting that by using chirped pulses and changing the sign and the value of the chirp parameter this process can be managed and it is possible to obtain parabolic intensity profiles with different curvatures and sign with respect to the z axis. By properly tuning the chirp parameters of the 5−25 fs pulses, the process of generation of electron bundles with arbitrary forms can be controlled. Thus, different kinds of converging or diverging electronic mirrors can be produced.

AUTHOR INFORMATION

Corresponding Authors

Hassan Chamati − *Institute of Solid State Physics, Bulgarian Academy of Sciences, 1784 Sofia, Bulgaria*; orcid.org/0000-0002-0831-6945; Email: chamati@issp.bas.bg

Lubomir M. Kovachev − *Institute of Electronics, Bulgarian Academy of Sciences, 1784 Sofia, Bulgaria*; Email: lubomirkovach@yahoo.com

Authors

Ekaterina Iordanova − *Institute of Solid State Physics, Bulgarian Academy of Sciences, 1784 Sofia, Bulgaria*

Pavlina Miteva − *Institute of Electronics, Bulgarian Academy of Sciences, 1784 Sofia, Bulgaria*

Diana Dakova − *Physics and Technology Faculty, University of Plovdiv "Paisii Hilendarski", 4000 Plovdiv, Bulgaria*

Georgi Yankov − *Institute of Solid State Physics, Bulgarian Academy of Sciences, 1784 Sofia, Bulgaria*

Daniela A. Georgieva − *Faculty of Applied Mathematics and Computer Science, Technical University of Sofia, 1000 Sofia, Bulgaria*

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.4c02996

Notes

The authors declare no competing financial interest.

Biographies

Ekaterina Iordanova received her PhD degree in Applied Physics from the Technical University Eindhoven. She is an Associate Professor in the Department of Laser, Atomic, Molecular and Plasma Physics and currently the Director of the Institute of Solid State Physics at the Bulgarian Academy of Sciences. The main research interest and established experience are in the field of laser and plasma physics and nonlinear effects by ultrashort laser pulses.

Pavlina Miteva graduated from Plovdiv University "Paisii Hilendarski" with Bachelor's degree in Engineering Physics in 2009 and with Master's degree in Photonics and modern optical technology in 2011. Additionally Pavlina Miteva completed courses from SoftUni on Python programming in 2021, Data Science and Machine learning in 2022. Currently, she is a physicist at the Institute of Electronics of Bulgarian Academy of Science. Her research interests are in the fields of theoretical physics, photonics, linear and nonlinear optics, data science, and machine learning.

Diana Dakova graduated in Engineering Physics from Plovdiv University "Paisii Hilendarski" in 1987. She received her PhD in Physics from the Institute of Electronics at the Bulgarian Academy of Sciences in 2009. Currently, Dr. Dakova is Associate professor in the Department of Physics at the Faculty of Physics and Technology of Plovdiv University "Paisii Hilendarski". Her research interests are in the fields of theoretical physics, physics of wave processes, photonics, linear, and nonlinear optics.

Hassan Chamati got his Ph.D. and the research degree Doctor of Sciences in physics from the Institute of Solid State Physics, Bulgarian Academy of Sciences, where he has been acting as a Director since December 2015 until December 2023. He has been a Postdoctoral

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Fellow in many universities across Europe. He is a Full Professor and Head of the Theoretical Department at the same Institute. His main research interests are in the theory of Condensed Matter, including Soft Matter. He has published numerous papers in renowned international journals on quantum magnetism and computer modeling of materials and biomembranes. He established international collaborations with researchers from different countries. He served as Principal Investigator or team member in several research projects funded by national and international funding agencies.

Georgi Yankov received his PhD degree from the Institute of Solid State Physics at the Bulgarian Academy of Sciences. At the moment he is an Associate Professor at the Institute of Solid State Physics at the Bulgarian Academy of Sciences in the Department of Laser, Atomic, Molecular, and Plasma Physics. His main research interests are in the field of ultrashort laser physics, laser matter interactions, and nonlinear optics.

Daniela Georgieva got her Ph.D. in mathematics from the Technical University of Sofia, Bulgaria, in 2014. She has been an associate professor at the same university since June 2020. She is a member of the Faculty of Applied Mathematics and Informatics, Department of Mathematical Modeling and Numerical Methods. Her research interests are in the field of mathematical modeling of nonlinear physical processes. She has published papers in renowned international journals on nonlinear optics. The main results include: obtaining of localized solutions with finite energy of the wave equation, investigation of ionization-free filamentation processes and coherent THz generation, merging and energy exchange between filaments, asymmetric spectrum broadening of femtosecond laser pulses, and generation of supercontinuum.

Lubomir M. Kovachev got his PhD from Institute of General Physics (Prokhorov Institute), Moscow in 1991. The research degree Doctor of Sciences in physics he obtain from Institute of Electronics, Bulgarian Academy of Sciences. He was for many years Head of Nonlinear and Fiber Optics Laboratory in Institute of Electronics and now is Head of the Bulgarian Consortium "Extreme light" ELI-ERIC-BG. His scientific interests are in the fields of nonlinear optics, optical solitons, and the nonlinear field theory. The latest scientific investigations of Dr. Kovachev are on new nonlinear mechanism for generation coherent GHz and THz radiation from femtosecond pulses in isotropic materials, as well as on longitudinal polarization force, leading to a new type of collision ionization. He served as principal investigator or member in several research project funded by national and international funding agencies. Dr. Kovachev is author of 113 papers in scientific journals and Conference Proceedings, and guest editor and reviewer in journals of optics.

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