Spatial Evolution of a Strong Field of Few-cycle Light Beam in Dielectric Media with Induced Plasma Nonlinearity.

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Abstract. The paper reports results of computer simulation of strong light beam propagation in dielectric media in case of plasma generation. We investigate an extra-broadening of radiation spectrum to a ‘violet’ wing of visible range. We show that the resulting pulse spectrum is represented by sequence of well-separated maximums, broadening as propagation distance increases. Experimental data are compared with simulation results, showing a good mutual correspondence of spectral representations.

1. Introduction.

A propagation of high-intense femtosecond pulses through dielectric media without its optical breakdown allows observing of nonlinear optical phenomena more shaped and illustrative than for longer pulses. Nowadays spectral supercontinuum generation is investigated deeply and complexly for self-action of intense single pulse in dielectric media. So a significant attention is paid to examination of simultaneous propagation of two or more intense femtosecond pulses with different spectra, because spectral extra-broadening of such complex beam demonstrates new peculiarities and gives a notably richer output spectrum.

A number of papers [1-5] investigate a propagation of intense two-color beam through air in conditions when inharmonic oscillations of valence electrons convert to a plasma nonlinearity. Effective generation of wide-range IR-radiation (wavelength up to 1 mm) is experimentally demonstrated. This method allows acquiring the most intense THz-radiation [2], widely applicable [3-5]. So an actual problem is optimal conditions for such generation. This problem requires a theory of two femtosecond pulse interaction propagating in optical media upon plasma excitation, which was derived by us in paper [6]. This theory gives an excellent correlation between computed data and experimental results of observation of quazi-periodic dependence of generated THz radiation power on interacting pulses spatial shift.

Recent years studies examine experimentally an amplification of spectral short-wave wing caused by plasma nonlinearity and free electron motion [7-10]. The main source of a generation of light quants with higher energy (and, though, shorter wavelength) is assumed to be a conversion of kinetic energy accumulated by free electron in an oscillating electrical field to an electromagnetic wave at its recombination with an originating ion. Our dynamic model contains all necessary components to describe such an interaction: a wave equation describing self-action of light wave; a description of plasma excitation by a strong light beam and opposite process – recombination as the wave passes by, an equation of ponderomotive dynamics of free electron in an oscillating electric field.
The present paper reports main results of application of the model to an investigation of spectral short-wave wing amplification of high-intense femtosecond pulse propagating through dielectric media in case of with plasma excitation. We show the resulting spectrum, present its spatial dynamics and compare it with experimental data obtained in [11].

2. A strong few-cycle pulse evolution equation for isotropic dielectric media with plasma nonlinearity.

We have deduced an equation for strong few-cycle pulse dynamics in transparent isotropic dielectric media in [6], on a basis of density matrix formalism in three-band optical media approach. The higher conductivity subband is assumed to be a free electron motion state for a describing of plasma nonlinearity. We assumed that a field of radiation is essentially smaller than electric field of unitary ionized atom on a distance equal to Bohr radius \( E_\text{B} = 10^5 \text{ V/m} \), i.e. we are considering a radiation with intensity \( I \ll 10^{17} \text{ W/cm}^2 \). This allows use of habitual energetic model of a dielectric with quasi-discrete energy state structure. We consider a ESP interaction with dielectric medium, assuming that the spectrum of pulse, though it is very wide, is bounded by a material transparency range. This assumption is correct up to a several femtoseconds duration of optical pulse. In circumstances of non-resonant (in single-photon approximation) interaction we assume that linear part of dielectric polarization principally exceeds the nonlinear part, depending on a higher field orders. This assumption, for example, in fused silica, is met for pulse intensities up to \( I \sim 5 \times 10^{14} \text{ W/cm}^2 \). So this makes possible to limit ourselves to analyzing of third-order nonlinearity of media polarization. Thus we analyze only electronic nonlinearity, neglecting other mechanisms in few-cycle field because of their inertia characteristic times cardinally exceed pulse length [12, 13].

The result has a form of system of waveform evolution equation and dynamic material ones, considering inertial part of field-cube electron nonlinearity as well as inertial plasma nonlinearity induced in pulse strong field. Neglecting a self-reflected wave, as grounded in [14, 15], we combine this system to united evolution equation (1):

\[
\frac{\partial E}{\partial z} - \frac{\partial^3 E}{\partial \tau^3} + g \frac{\partial E^3}{\partial \tau} + \frac{\partial}{\partial \tau} \left( g^{(1)} E \left( \frac{\partial E}{\partial \tau} \right)^2 + g^{(2)} E^2 \frac{\partial^2 E}{\partial \tau^2} \right) + \frac{2\pi}{c n_0} j = 0
\]

\[
\frac{\partial \rho}{\partial \tau} + \frac{\rho}{\tau_\rho} = \alpha E^2
\]

\[
\frac{\partial}{\partial \tau} j + \frac{j}{\tau_e} = \beta \rho E^3,
\]

where \( E \) is pulse field electrical component, \( z \) is a spatial coordinate collinear to pulse propagation direction, \( \tau = t - \frac{n_0}{c} z \), \( c \) - vacuum light speed, \( n_0 \) and \( \alpha \) characterize a dependence of linear refraction index \( n \) on a frequency \( \omega \)

\[
n(\omega) = n_0 + c a \omega^2,
\]

which can be easily refined, when necessary, with higher order additives, by expanding (3) with higher order field time derivatives [15]; \( j \) stands for density of ionized electrons current, \( \rho \) – for their density; factors \( g \), \( g^{(1)} \), \( g^{(2)} \) describe nonlinearity refraction index of dielectric media

\[
n^2(\omega) = n^2_0 + A \omega^2,
\]
where \( n_2^0 = \frac{3}{2} c g + \frac{1}{2} \tau_{12} c g^{(1)}, \quad A = \frac{1}{2} c (g^{(1)} - 3 g^{(2)}), \quad \alpha = \frac{1}{2 n_0} \frac{|p_{23}|^2}{T_{23} (h \omega_{32})^2}, \quad \beta = \frac{e^2}{m_e c} n_0^2 - 1 \) characterizes inertial plasma nonlinearity of dielectric media (see [6]), \( p_{ij} \) are scalar values of dipole moments of interstate transitions \( i \rightarrow j \), \( \omega_{ij} \) – frequencies according to these transition energy, \( T_{ij} \) are interstate relaxation times and \( \tau_{ij} \) are state excitation relaxation times, \( m_e^* \) is an effective electron mass in conductivity band, \( \sigma = 2 \tau_e^{-1} \) is defined by average collision relaxation time \( \tau_e \) of free electrons, which has an order of tens femtoseconds for most dielectrics.

3. A broadening of high-intense few-cycle pulse spectrum to ‘violet’ wing while propagating in SF6.

Pictures 1-3 illustrate results of numerical simulations of few-cycle pulse represented as

\[
E(t) = E_0 \exp \left(-\left(\frac{t}{\tau_0}\right)^2\right) \sin(\omega_0 t),
\]

where \( E_0 \) – electric field amplitude, \( \tau_0 \) – input pulse duration, \( \omega_0 \) – central wavelength, propagating in dielectric media. Modeled media is chosen to be sulfur hexafluoride (SF6) having an electrical resistance index several times higher than atmosphere air. We use the following values for media properties: a linear refraction index \( n_0 = 1.0021 \) ([16]), nonlinear refraction index \( n_2 = 1.6 \cdot 10^{-19} \text{cm}^2 / \text{W} \) ([17, 18]).

Figure 1 shows the extrabroadening of pulse spectrum in a wavelength range of \( 350 - 950 \text{nm} \), at pulse properties: duration \( \tau_0 = 30 \text{fs} \), central wavelength \( \lambda_0 = 2 \pi c / \omega_0 = 800 \text{nm} \), input peak intensity \( I = 1 \cdot 10^{14} \text{W} / \text{cm}^2 \), propagation distance \( z = 6.4 \text{mm} \) (additional picture gives an overview of spectrum in wider range \( 350 \text{nm} - 8 \mu \text{m} \)). For better illustration, this spectrum is compared with the one obtained by modeling the same circumstances and propagation regime, except plasma nonlinearity, that is supposed to be absent in some model media (dashed line). It is notable that plasma nonlinearity causes a drastic broadening of pulse spectrum into a short-wave range, having a form of separated spectral peak sequence. And when considering only inertia-less nonlinearity, we obtain some generation of longer wave components.

![Figure 1. Extrabroadening of pulse spectrum caused by plasma nonlinearity (solid line), compared with the one in case of plasma neglecting (dashed) and initial spectrum (dotted).](image-url)
Figure 2 illustrates this effect in dynamics, as it increases and evolves as the pulse propagates for a longer distance. This leads us to a conclusion that a propagation of a separated femtosecond pulse through a dielectric media with notable plasma nonlinearity may be accompanied with generation of high-frequency spectral components, dominating frequency ‘green shift’, in this case – to approximately 500 nm. It is obvious, that this phenomena differs from well-known harmonic generation, caused by inertia-less nonlinearity of third and higher orders.

![Figure 2](image)

**Figure 2.** Spatial dynamic of broadening pulse spectrum, amplifying itself as distance increases.

![Figure 3](image)

**Figure 3.** A combination of modelled (solid line) and experimental (shaded area) data on examined spectral broadening in sulphur hexafluoride.
Figure 3 combines a spectrum of modeled output pulse and experimental data on femtosecond pulse propagation in sulfur hexafluoride, published in [11]. Experimentally detected spectrum broadening, forming several high-frequency maximums (shaded area) correlates well with numerically simulated effect, reflecting, as well, appearance of several contrast maximums (solid line). We see the good accordance between quantity and positions of spectral peaks for experimental and simulated data. Higher contrast for modeled picture may be grounded by ideality of input pulse spectra and spatial profile, neglecting of diffraction and boundary effects. This allows to conclude that the approach derived in the present report is practically consistent and may be applied for investigations of interaction between high-intense laser pulses with dielectric media, considering media ionization and pre-breakdown phenomena.

4. Conclusion.
The study reports an application of dynamic interaction model to a simulation of high-intense femtosecond pulse propagation through dielectric media, in case pulse properties cause the initiation of optical breakdown, generation of free electrons. It is shown that the model demonstrates a generation of several isolated spectral maximums between main and second harmonics of initiative pulse, and besides the quantity and intensity of these maximums is dependent on propagation distance. Simulation results are compared with experimental data of investigation of femtosecond pulse propagation in sulfur hexafluoride, showing a notably high level of data correspondence. That enables concluding the definite practical value of derived approach and prospective application of the model to planning experiments, processing experimental data and explaining a physical nature of observed phenomena.

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