Generation of multiplexed harmonics at interaction of multi-harmonic light beams in dielectric media with induced plasma.

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Abstract. A significant number of papers investigate a propagation of intense two-color beam through air in conditions when inharmonic oscillations of valence electrons convert to a plasma nonlinearity. This paper extends the scope of analysis, examining more complex combinations of synchronized beams propagating through dielectric media: triple beam and a beam combined with quazi-static field. It is shown that adding a third harmonic to a mix of first and second ones does not show principal changes in interaction picture: all phenomena appear to be described well before. But adding a static field to an interaction scene shows new peculiarities in redistribution of pulse spectrum energy to infra-red wing of optical range.

1. Introduction.

A generation of spectral supercontinuum by a pair of synchronized few-cycle pulses, including a generation of radiation in terahertz range (THz), is an object of complex and thorough studies this decade [1-5]. Fine tuning of generation procedure, opening a way to cost-effective and efficient generation of long-wave radiation (up to 1 mm), requires an accurate and applicable theory, combining a description of optical processes in dielectric media with analysis of high-energetic processes such as plasma induction. We derived the theory several years ago [6]. This theory gives an excellent correlation between computed data and experimental results of observation of quaizi-periodic dependence of generated THz radiation power on interacting pulses spatial shift.

Noticing a fast progress of experimental set-ups, allowing generation and combination of multiple synchronized optic pulses, we found out that theoretically it is possible to increase an efficiency of THz generation by adding pulses of tripled basic wavelength to an interaction scene. So the present paper shows some results of examining such multiwave high-intense femtosecond pulse propagation through dielectric media with plasma excitation. The additional feature of this study is an investigation of high-intense few-cycle pulse interaction with media at presence of quasi-static electric field, causing a separate ponderomotive dynamics of free electrons induced by input pulse.

2. A model for few-cycle pulse evolution in isotropic dielectric media with excited free electrons.

The model for strong few-cycle pulse dynamics in transparent isotropic dielectric media we deduced in [6] based on density matrix for three-band approximation of media energy structure. We assume the higher conductivity subband as a quaizi-free motion state for valence electrons, thus it describes an induced photo-plasma. Other assumptions are: an electric field of input wave is essentially smaller than
a field of ionized atom on a Bohr radius \( E_s = 10^9 V/m \), i.e. we consider pulse intensities \( I << 10^{17} W/cm^2 \). This allows use of habitual energetic model of a dielectric with quasi-discrete energy state structure. We consider dielectric medium as transparent in a wide range, assuming that the spectrum of input and propagating pulse is very wide but does not exceed the media transparency range. This assumption limits pulse duration to several femtoseconds that stands for only few oscillations of optical field. We assume also linear component of polarization response notably exceeding the nonlinear one, that is met, for example, in fused silica at pulse intensities up to \( I \sim 5 \cdot 10^{14} W/cm^2 \). Thus we analyze only electronic nonlinearity, neglecting other mechanisms in few-cycle field because of their inertia characteristic times cardinally exceed pulse length [7, 8].

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 [9, 10], we combine this system to united evolution equation (1):

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

\[
\frac{\partial \rho}{\partial \tau} + \frac{\rho}{\tau_p} = aE^2
\]

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

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 \( a \) characterize a dependence of linear refraction index \( n \) on a frequency \( \omega \)

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

which can be easily refined, when necessary, with higher order additives, by expanding (3) with higher order field time derivatives [10], \( j \) is induced electric circuit density, \( \rho \) is a density of excited (to lower conductivity band) dipoles; factors \( g , g^{(1)} , g^{(2)} \) describe nonlinearity refraction index of dielectric media

\[
n_2(\omega) = n_2^0 + A\omega^2,
\]

where \( n_2^0 = \frac{3}{2} c g + \frac{1}{2} \gamma_2 c g^{(1)}, \quad A = \frac{1}{2} c (g^{(1)} - 3 g^{(2)}), \quad \alpha = \frac{1}{2 n_0 T_{21}} \left( \frac{p_{23}}{h \omega_{21}} \right)^2, \quad \beta = \frac{e^2}{m_e^* c} n_2^0 - 1 \)

characterizes inertial plasma nonlinearity of dielectric media (see [6]), \( p_{ik} \) are scalar values of dipole moments of interstate transitions \( i \rightarrow k \), \( \omega_{ik} \) – frequencies according to these transition energy, \( T_{ik} \) are interstate relaxation times and \( \tau_{ik} \) are state excitation relaxation times, \( m_e^* \) is an effective electron mass in conductivity band, \( \sigma = 2\tau_c^{-1} \) is defined by average collision relaxation time \( \tau_c \) of free electrons, which has an order of tens femtoseconds for most dielectrics.

This model was applied to an investigation of field structure of generated IR and THz radiation in an experiment of bi-harmonic beams propagation in air held in ITMO University [6]. A dependence of generation efficiency on filament (generated by pulses interaction in air) length is acquired and examined. It is shown that the dependence is periodical by filament lengths. We gave some recommendations on optimization of experimental setup for acquiring a most intensive THz radiation: an output power can be significantly increased by controlling a filament length (by focusing system
parameters). It was also shown that integral intensity of generated pulse in range 25.5 μm – 0.2 mm depends periodically on filament length, and this THz pulse appears as few-cycle.

3. An interaction of multi-harmonic input pulse with dielectric media at photoplasma induction conditions.

Figures 1-2 illustrate results of numerical simulations of multi-harmonic few-cycle pulse represented as

\[ E(t) = E_0 \exp\left(-\left(\frac{t}{\tau_u}\right)^2\right) \sin(\omega_0 t) + E_1 \exp\left(-\left(\frac{t}{\tau_u}\right)^2\right) \sin(2\omega_0 t + \varphi_1) + E_2 \exp\left(-\left(\frac{t}{\tau_u}\right)^2\right) \sin(3\omega_0 t + \varphi_2), \]

where \( E_0 \) – initial electric field amplitude, \( \tau_u \) – input pulse duration, \( \omega_0 \) – central wavelength, \( E_1, E_2 \) are amplitudes of synchronized derivative harmonics (they are functions of \( E_0 \), determined by a scheme of experimental set-up), \( \varphi_1, \varphi_2 \) are relative phase shifts accordingly. Modeled media is chosen to be an atmosphere air, so we use the following values for media properties: a linear refraction index \( n_0 = 1.00027 \), nonlinear refraction index \( n_2 = 5.6 \times 10^{-19} \text{cm}^2/\text{W} \), \( \rho = 1.225 \times 10^{-3} \text{g/cm}^3 \), \( \tau_p = 65 \text{fs} \), \( \tau_p = 150 \text{fs} \). Figure 1 shows the extrabroadening of input pulse spectrum (dotted line) in a wavelength range of 350 – 950 nm, at pulse properties: duration \( \tau_u = 10 \text{fs} \), central wavelength \( \lambda_0 = 2\pi c / \omega_0 = 800 \text{nm} \), input peak intensity \( I = 1 \times 10^{14} \text{W/cm}^2 \), propagation distance \( z = 0.32 \text{mm} \), \( E_1 = 0.5 E_0 \), \( E_2 = 0 \). To illustrate the role and importance of ionization mechanisms, we compare the modelled spectrum (solid line) with the one obtained by modeling the same circumstances and propagation regime, except plasma nonlinearity (dashed line). It is notable that plasma nonlinearity causes a drastic broadening of pulse spectrum into a long-wave range, that was examined in a set of previous our publications [14, 15, 16]. We see that this effect is mostly caused by plasma nonlinearity, as reference dashed line (effect without plasma) shows almost no generation in long-wave wing.

Figure 2 illustrates adding the third harmonic to the interaction scheme: \( E_3 = 0.5 E_0 \), all other properties are the same as above. The most noticeable thing is that no principal addition is caused to an interaction scheme by such addition: because a generation of third harmonic is generic and well-known.
effect of inertia-less nonlinearity mechanisms, the photo-induced plasma makes no principal contribution in the whole process. All differences between examined (solid) and reference (dotted) plots are included into Figure 1 and caused by second harmonic participation in interaction process.

![Figure 2](image1.png)

**Figure 2.** Spectral density of energy of electric field for pulse (4) with $E_2 = 0.5E_0$ propagating with plasma nonlinearity (solid line), compared with the one in case of plasma neglecting (dashed) and initial spectrum (dotted). $G_0$ stands for maximum of spectral density.

![Figure 3](image2.png)

**Figure 3.** Spectral density of energy of electric field for pulse (4) with $E_2 = 0.5E_0$ propagating with plasma nonlinearity (solid line), compared with the one in case of plasma neglecting (dashed) and initial spectrum (dotted). $G_0$ stands for maximum of spectral density.

Figure 3 shows a result of a propagation of high-intense few-cycle pulse in dielectric media at presence of quazi-static electric field:

$$E(t) = E_0 \exp(-t/t_0^2) \sin(\omega_0 t) + E_\perp \exp(-(t/t_\perp)^2),$$

(5)
It is noticeable that the most visible effect of such propagation is a notable redistribution of pulse energy to low-wave wing of optical spectrum (for better view, exponential abscissa is presented on a plot for wavelength). In experiment, the situation can be modelled by adding an electric condenser with charging contour to a focusing point of input pulse. It is expectable to obtain wide-band radiation in IR-spectrum, caused by an interaction of quasi-free electrons induced by optical pulse, static and oscillating electric field.

4. Conclusion.
The study reports an application of dynamic interaction model to simulate a propagation of complex multi-harmonic high-intense femtosecond pulse through dielectric media, under circumstances initiating an excitation of free electrons in media volume. We show that a propagation of plasma-inducing pulse through dielectric at presence of quasi-static field is accompanied by a notable redistribution of pulse energy to low-wave wing of optical spectrum. That gives a recommendation how to set up an experiment for obtaining wide-band radiation in IR-spectrum, caused by an interaction of quasi-free electrons induced by optical pulse, static and oscillating electric field. The model shows its definite practical value in prospective application to experiment planning and experimental data processing.

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