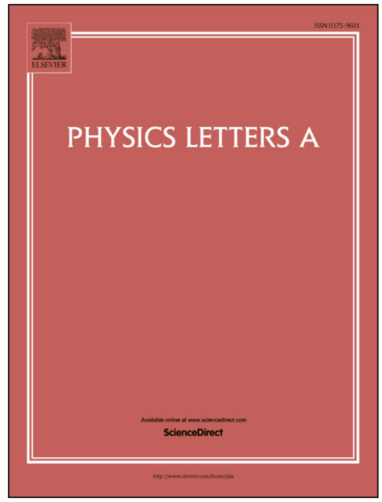
Accepted Manuscript



Harmonic Generation Enhancement due to Interaction of Few-Cycle Light Pulses in Nonlinear Dielectric Coating on a Mirror

Elizaveta M. Buyanovskaya, Mikhail A. Kniazev, Sergei A. Kozlov, Andrey A. Sukhorukov

PII: S0375-9601(17)30864-2
DOI: http://dx.doi.org/10.1016/j.physleta.2017.09.010
Reference: PLA 24723

To appear in: *Physics Letters A*

Received date: 9 July 2017
Revised date: 7 September 2017
Accepted date: 8 September 2017

Please cite this article in press as: E.M. Buyanovskaya et al., Harmonic Generation Enhancement due to Interaction of Few-Cycle Light Pulses in Nonlinear Dielectric Coating on a Mirror, *Phys. Lett. A* (2017), http://dx.doi.org/10.1016/j.physleta.2017.09.010

Highlights

- Problem of few-cycle pulse reflection from a mirror with a nonlinear layer was solved
- Interaction of input and reflected pulse modify THG efficiency
- Counter-directional interactions increase spectral broadening for ultra-short pulses.

Harmonic Generation Enhancement due to Interaction of Few-Cycle Light Pulses in Nonlinear Dielectric Coating on a Mirror

Elizaveta M. Buyanovskaya[a,*], Mikhail A. Kniazev[a], Sergei A. Kozlov[a], Andrey A. Sukhorukov[b]

[a]*ITMO University, 49 Kronverskiy pr., Saint-Petersburg, Russia, 197101, (812)232-1467*
[b]*Nonlinear Physics Centre, Research School of Physics and Engineering, Australian National University, Canberra, ACT 2601, Australia*

Abstract

We theoretically investigate reflection of a few-cycle light pulse from a mirror with nonlinear dielectric coating. We employ a nonlinear equation that describes spatiotemporal evolution of a few-cycle light pulse with a broad spectrum that lies in the transparency range of nonlinear dielectric media. This model is formulated directly for the electric field without slowly varying amplitude approximation. Analytical and numerical analysis shows that counter-propagating wave interactions in thin films can strongly enhance or suppress third harmonic generation of the central frequency, whereas this effect is neglected in the framework of slowly varying amplitude approximation.

Keywords: nonlinear optics; few-cycle pulses; harmonics generation
2016 MSC: 78A40, 78A60

1. Introduction

Nonlinear optics of waves consisting from only few cycles of electromagnetic field is an active research field at the forefront of optics and laser physics of ultra short intense pulses [1, 2, 3]. Various nonlinear effects associated with the

*Corresponding author
Email addresses: `lee.buyanovskaya@niuitmo.ru` (Elizaveta M. Buyanovskaya), `knyazev.michael@gmail.com` (Mikhail A. Kniazev), `kozlov@mail.ifmo.ru` (Sergei A. Kozlov), `Andrey.Sukhorukov@anu.edu.au` (Andrey A. Sukhorukov)

few-cycle pulses have been investigated, including their temporal and spectral broadening, self-focusing, nonlinear reflection, generation of single-cycle solitons, co- and counter-directional interactions [4, 5, 6, 7, 8, 9, 10, 11]. Due to the lack of destruction of optical media in the field of intense extremely short pulses, the nonlinear effects can be significant, being beneficial to potential practical applications ranging from all-optical signal processing in integrated circuits to sum-frequency generation spectroscopy [12, 13].

In this paper, we investigate the generation of high frequency radiation in a thin layer of nonlinear dielectric media on top of a perfect metal mirror, and address the influence of interaction between the counter-propagating incident and reflected pulses, considering Kerr-type of nonlinearity [14]. Such geometries, as well as multilayered structures and micro-cavities, are usually analyzed using the transfer matrix method [15, 16, 17]. Here, we develop a flexible theoretical approach based on the wave equation formulated directly for the electric field of a few-cycle optical pulse [18, 19, 20, 21, 22, 23, 24, 25], thus overcoming the limitations of the traditional method of slowly varying envelope. For extremely short pulses, we can neglect laser damage in thin dielectric films, which was extensively investigated in series of papers [26, 27]. We predict that due to the nonlinear interaction of a few-cycle light pulse with the wave reflected from the mirror, there appears an increase in nonlinear phase modulation and spectral broadening accompanied by a modification of third-harmonic generation (THG) efficiency due to interferences between its components.

The paper is organized as follows. In section 2 we introduce a nonlinear equation describing the field dynamics of few-cycle pulses interacting in a dielectric medium with cubic nonlinearity, and illustrate that for the quasi-monochromatic incident wave the governing equation reduces to a system of four coupled equations for slowly-varying envelopes of interacting harmonics. We show that slowly varying amplitude approximation does not account for this effect. In the following section 3, we normalize the field equation and provide physical estimates of the strength of diffraction, dispersion or nonlinearity for few-cycle pulses in terahertz and near IR spectral ranges.

We then analyze in detail nonlinear harmonic generation of high-power pulses in thin layers, when dispersion and diffraction are relatively weak. We obtain a solution of the field equation for pulse reflection from the perfect metal mirror using the method of successive approximations and visualize results with numerical simulations. We find that the efficiency of the third harmonics generation is significantly affected by the interaction of counter-propagating pulses in the nonlinear layer and depends on layer thickness and duration of the incident pulse. Finally, we present conclusions in Sec. 4.

2. Nonlinear equation for few-cycle pulses

We aim to investigate the evolution of few-cycle light pulses in a nonlinear dielectric layer. The traditional method of slowly varying envelope is not suitable to describe propagation of few-cycle pulses in optical media, since the concept of wave envelope becomes physically meaningless. Therefore the dynamics of either the radiation field itself or its spatial-temporal spectra need to be considered for modeling of nonlinear optical effects for few-cycle waves.

For a linearly polarized paraxial wave normally incident on a dielectric layer, and assuming that the pulse spectrum is in the transparency range of the nonlinear dielectric, the wave equation can be written as [19, 20, 18, 25]:

$$\frac{\partial^2 E}{\partial z^2} + \Delta_\perp E - \frac{{N_0}^2}{c^2}\frac{\partial^2 E}{\partial t^2} + \frac{2N_0}{c}a\frac{\partial^4 E}{\partial t^4} - \frac{2N_0}{c}g\frac{\partial^2 E^3}{\partial t^2} = 0. \tag{1}$$

Here E is the amplitude of the electrical field, z is the propagation direction, $\Delta_\perp = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}$ is the transverse Laplace operator, t is time, $g = 2n_2/c$, n_2 is the nonlinear coefficient, and c is the light velocity in vacuum. Parameters N_0 and a characterize the typical nonresonant dependence of linear dielectric refractive index within its transparency range,

$$n^2 = {N_0}^2 + 2cN_0 a\omega^2, \tag{2}$$

where ω is optical frequency.

The wave equation (1) describes propagation of transversely bounded optical waves with broad spectra in both positive and opposite directions of z

axis, including the case of counter-propagating wave interaction in nonlinear media. The model can be extended to account for arbitrary polarization and cases of more complex dispersion of linear refractive index and nonlinear media response [28].

It is instructive to illustrate that this equation reduces to coupled envelope equation in case of quasi-monochromatic input wave, taking into account the effect of third-harmonic generation:

$$
\begin{aligned}
E(x,y,z,t) = \frac{1}{2}\Big(& \epsilon_{+}(x,y,z,t)\exp\left[i\left(k_0 z-\omega_0 t\right)\right] \\
& + \epsilon_{+}^{(3)}(x,y,z,t)\exp\left[i(k(3\omega_0)z-3\omega_0 t)\right] \\
& + \epsilon_{-}(x,y,z,t)\exp\left[-i(k_0 z+\omega_0 t)\right] \\
& + \epsilon_{-}^{(3)}(x,y,z,t)\exp\left[-i\left(k(3\omega_0)z+3\omega_0 t\right)\right] + \text{c.c.}\Big)
\end{aligned} \tag{3}
$$

where $\epsilon_{\pm}$, $\epsilon_{\pm}^{(3)}$ are the slowly varying envelopes of counter-propagating quasi-monochromatic pulses with the carrier frequencies ω_0 and $3\omega_0$, respectively. Considering low conversion efficiency, when $|\epsilon_{\pm}^{(3)}| \ll |\epsilon_{\pm}|$, under typical assumptions of slowly varying envelope approximation the wave equation (1) can be reduced to a system of four coupled equations for envelopes of interacting quasi-monochromatic pulses at the fundamental and third harmonic [29]:

$$
\begin{aligned}
&\frac{\partial \epsilon_{+}}{\partial z}+\frac{1}{v_g}\frac{\partial \epsilon_{+}}{\partial t}+i\frac{\beta_2}{2}\frac{\partial^2 \epsilon_{+}}{\partial t^2}-\frac{\beta_3}{6}\frac{\partial^3 \epsilon_{+}}{\partial t^3}-i\gamma\left(|\epsilon_{+}|^2+2|\epsilon_{-}|^2\right)\epsilon_{+}=\frac{i}{2k_0}\Delta_{\perp}\epsilon_{+}\,,\\
&\frac{\partial \epsilon_{-}}{\partial z}-\frac{1}{v_g}\frac{\partial \epsilon_{-}}{\partial t}-i\frac{\beta_2}{2}\frac{\partial^2 \epsilon_{-}}{\partial t^2}+\frac{\beta_3}{6}\frac{\partial^3 \epsilon_{-}}{\partial t^3}+i\gamma\left(|\epsilon_{-}|^2+2|\epsilon_{+}|^2\right)\epsilon_{-}=-\frac{i}{2k_0}\Delta_{\perp}\epsilon_{-}\,,\\
&\frac{\partial \epsilon_{+}^{(3)}}{\partial z}+\frac{1}{v_g^{(3)}}\frac{\partial \epsilon_{+}^{(3)}}{\partial t}+i\frac{\beta_2^{(3)}}{2}\frac{\partial^2 \epsilon_{+}^{(3)}}{\partial t^2}-\frac{\beta_3^{(3)}}{6}\frac{\partial^3 \epsilon_{+}^{(3)}}{\partial t^3}-3i\gamma\epsilon_{+}^3 e^{i\Delta k z}=\frac{i}{2k(3\omega_0)}\Delta_{\perp}\epsilon_{+}^{(3)},\\
&\frac{\partial \epsilon_{-}^{(3)}}{\partial z}-\frac{1}{v_g^{(3)}}\frac{\partial \epsilon_{-}^{(3)}}{\partial t}-i\frac{\beta_2^{(3)}}{2}\frac{\partial^2 \epsilon_{-}^{(3)}}{\partial t^2}+\frac{\beta_3^{(3)}}{6}\frac{\partial^3 \epsilon_{-}^{(3)}}{\partial t^3}+3i\gamma\epsilon_{-}^3 e^{i\Delta k z}=-\frac{i}{2k(3\omega_0)}\Delta_{\perp}\epsilon_{-}^{(3)}\,,
\end{aligned} \tag{4}
$$

where $v_g, v_g^{(3)} = (\partial k/\partial\omega)^{-1}_{\omega_0,3\omega_0}$ are the group velocities of fundamental wave and the third harmonic, respectively, $\beta_n, \beta_n^{(3)} = (\partial^n k/\partial\omega^n)_{\omega_0,3\omega_0}$, $n = 2,3$; $\gamma = 3g\omega_0/4$, $\Delta k = 3k(\omega_0) - k(3\omega_0)$, $k(\omega) = \omega n(\omega)/c$.

The coupled-mode equations (4) are derived using typical simplifications of slowly varying envelopes and therefore do not include fast oscillating terms such as $\pm 9i\gamma|\epsilon_\pm|^2\epsilon^*_\mp \exp\{3i\,[k(\omega_0) - k(3\omega_0)]\,z\}$ in the third and the forth equations, respectively. Therefore THG for quasi-monochromatic pulses is produced only by co-directional pump wave, whereas a counter-directional pump wave leads just to an additional phase modulation of generated wave [14]. However in several studies of optical harmonics generation in thin films, multilayers, and micro-resonators [12, 15, 30], it was shown that counter-directional wave can significantly enhance the harmonic generation. Accordingly, in this work we employ the general theoretical model Eq. (1), rather then the coupled-mode Eqs. (4). Indeed, we show in the following that interactions of counter-propagating waves can significantly affect THG in thin nonlinear films on top of a metal mirror.

3. Third harmonic generation

3.1. Solution for few-cycle pulses

In order to solve equation (1), we first perform normalization by introducing new dimensionless variables:

$$E' = \frac{E}{E_{+0}},\, t' = \frac{4t}{T_{+c}},\, z' = \frac{z}{L_0}, \Delta'_\perp = \frac{\Delta_\perp}{\rho^2} \tag{5}$$

where E_{+0} is the maximum of the incident forward wave E_+ and ρ is its transverse size at the boundary of nonlinear media, T_{+c} is its central oscillation period, L_0 is the distance of the order of wavelength. Using new variables, the model equation is written in the following form:

$$\frac{\partial^2 E}{\partial z^2} + \frac{L_0^2}{L_{\mathrm{dif}} L_w}\Delta_\perp E - \frac{L_0^2}{L_w^2}\frac{\partial^2 E}{\partial t^2} + 2\frac{L_0^2}{L_{\mathrm{disp}} L_w}\frac{\partial^4 E}{\partial t^4} - 2\frac{L_0^2}{L_{\mathrm{nl}} L_w}\frac{\partial^2 E^3}{\partial t^2} = 0, \tag{6}$$

where

$$L_w = \frac{cT_{+c}}{4N_0},\, L_{\mathrm{disp}} = \frac{T_{+c}^3}{64a}, L_{\mathrm{dif}} = \frac{\rho^2}{L_w},\, L_{\mathrm{nl}} = \frac{T_{+c}c}{8n_2E_0^2} = \frac{T_{+c}c}{4n_2 I}, \tag{7}$$

and I is the intensity of light. In Eq. (6) and in the following we omit the symbol ,, $'$ " for the sake of brevity and take $L_0 = L_w$.

The chosen normalization equalizes the scales of fields and their derivatives for the few-cycle pulses. The relations between coefficients L_w^{-1}, L_{disp}^{-1}, L_{dif}^{-1}, L_{nl}^{-1} depend on the dielectric medium characteristics and initial radiation parameters (pulse duration τ_0, width ρ and its amplitude E_{+0} defining the pulse energy), which in turn indicate if dispersion, diffraction or nonlinearity has a dominant effect on the pulse evolution.

Let us estimate the normalization coefficients for dielectric materials in visible and far IR spectral regions. First we consider a fused silica with parameters $N_0 = 1.45$, $a = 2.74 \cdot 10^{-44}\ s^3/cm$, $n_2 = 2.9 \cdot 10^{-16}\ cm^2/W$ [18]. We note that electron-hole plasma was registered for pulses with duration $\sim 100\ fs$ at 800 nm wavelength and for intensity $1.3 \cdot 10^{13} W/cm^2$ in fused silica glass [31], and other experimental studies support this observation [32]. In Ref. [33], it was shown that critical intensity that leads to plasma formation in fused silica reaches $\sim 3 \cdot 10^{13}\ W/cm^2$ for pulses with a duration of 10 fs. However in our current study, we consider fs pulses with duration down to one cycle (∼3 fs), and in this regime only instantaneous cubic nonlinearity of dielectric media should be dominant for peak intensities $I = 3 \cdot 10^{13}\ W/cm^2$, because for such short pulses with accordingly lower energy the plasma formation would not occur. For such a peak intensity, central wavelength $\lambda_{+c} = 780\ nm$, $T_{+c} = 3\ fs$, and pulse transverse size $\rho = 10 \cdot \lambda_{+c}$, we obtain $L_w = 0.14\ \mu m$, $L_{\mathrm{disp}} = 108\ \mu m$, $L_{\mathrm{dif}} = 464\ \mu m$, and $L_{\mathrm{nl}} = 23\ \mu m$.

We now perform estimates for terahertz pulses. We consider a crystalline quartz with parameters $N_0 = 2.105$, $a = 3.3 \cdot 10^{-39}\ s^3/cm$ (obtained by interpolation of data from [34] using equation (2)) and $n_2 = 4.4 \cdot 10^{-12}\ cm^2/W$ [35]. Then, for a THz pulse with peak intensity $I = 5 \cdot 10^9\ W/cm^2$, central wavelength $\lambda_{+c} = 300\ \mu m$, $T_{+c} = 1\ ps$, and transverse size $\rho = 10 \cdot \lambda_{+c}$, we obtain $L_w = 36\ \mu m$, $L_{\mathrm{disp}} = 47\ mm$, $L_{\mathrm{dif}} = 253\ mm$, and $L_{\mathrm{nl}} = 3.4\ mm$.

We see that under the physical conditions considered above, the nonlinear effect dominates at the initial stage of wave propagation in the given conditions. Therefore, we can neglect dispersion and diffraction at small propagation lengths. The intensity of counter-propagating waves which does not lead to destruction of the media can be quite high [3] because of their extremely short durations. However the interaction between counter-propagating pulses can still be relatively weak due to the transience of pulse collision. Accordingly, we choose the Picard's method of successive approximations [36], in which $L_w/L_{\rm nl}$ is a small parameter, as the analytical approach to approximately solve Eq. (6). In Ref. [7] it was shown that effects for the third harmonic generation predicted analytically by the perturbation theory are in good agreement with results obtained numerically. Then, we seek a solution in the following form:

$$E = E^{(0)} + \frac{L_w}{L_{\rm nl}} E^{(1)}. \tag{8}$$

In the following we consider propagation lengths of the order of $L_w \ll L_{\rm disp}, L_{\rm dif}$. Accordingly, we neglect both diffraction and dispersion of linear refractive index. This approximation is valid due to the small non-resonant dispersion of dielectrics refractive index [18]. The presence of weak dispersion can be accounted for if necessary by including them as a perturbation [7], to account for the pulse reshaping involving asymmetry of the electric field profile and their effect in the third harmonics generation efficiency. We do not perform such analysis in the current paper to concentrate on the key nonlinear effects in the regime of negligible dispersion. Then we consider the limit $L_{\rm disp} = \infty$ (dispersionless media) and $L_{\rm dif} = \infty$ (plane wave), substitute Eq. (8) into Eq. (6), and obtain

$$\frac{\partial^2 E^{(0)}}{\partial z^2} - \frac{\partial^2 E^{(0)}}{\partial t^2} = 0, \tag{9}$$

$$\frac{\partial^2 E^{(1)}}{\partial z^2} - \frac{\partial^2 E^{(1)}}{\partial t^2} - 2\frac{\partial^2 \left(E^{(0)}\right)^3}{\partial t^2} = 0. \tag{10}$$

The zero-order Eq. (9) is linear and its solution can be written as a superposition of forward and backward waves,

$$E^{(0)} = E_+^{(0)}\left(\tau_-\right) + E_-^{(0)}\left(\tau_+\right), \tau_- = t - z,\, \tau_+ = t + z. \tag{11}$$

In this paper we model the pulse reflection from the perfect metal mirror located at the boundary $z = \tilde{L}$ with nonlinear dielectric layer (thickness of the layer is

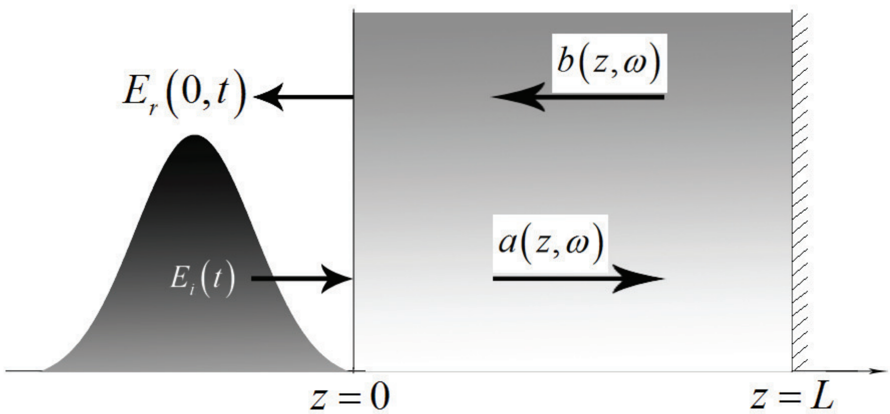


Figure 1: Schematic of few-cycle pulse reflection from a metal mirror (located at $z = \tilde{L}$) coated with a nonlinear dielectric layer (extending through $0 \leq z \leq \tilde{L}$).

normalized to L_w), as illustrated in Fig. 1. In this case there appears a counter-directional interaction between the front and the rear sections of the same pulse. Based on the relations between plasmonic frequencies of the common reflective materials [37] and frequency range considered in the following we assume perfect reflection from the mirror with zero accumulated phase, although it can be included if needed in the formulas below. We neglect linear and nonlinear reflections from the dielectric interface at $z = 0$ as well, assuming a low linear refractive index contrast. More general analysis accounting for reflections, when the layer forms a nonlinear Fabry-Perot interferometer, can also be performed using the developed analytical techniques, however this is outside the scope of the present paper.

The reflected wave at the dielectric boundary in the linear regime, corresponding to the zero-order approximation, is found as:

$$E_r^{(0)}(0,t) = -E_i\left(t - 2\tilde{L}\right), \tag{12}$$

where $E_i(t)$ is the incident pulse amplitude profile.

We use Fourier method to solve the model equations. In order to solve Eq. (10) for the first-order nonlinear correction, we reformulate it in the spectral domain,

$$\frac{\partial^2 \hat{E}^{(1)}}{\partial z^2} + \omega^2 \hat{E}^{(1)} = f\left(z, \omega\right), \tag{13}$$

where $\hat{E}^{(1)} = \mathcal{F}\left(E^{(1)}\right)$, hereinafter $\mathcal{F}\left(\ \right)$ denotes Fourier transform, and

$$f\left(z, \omega\right) = -2\omega^2 \mathcal{F}\left(\ \left[E^{(0)}\right]^3\right). \tag{14}$$

We seek solution of Eq. (13) in the form

$$\hat{E}^{(1)} = a(z)e^{i\omega z} + b(z)e^{-i\omega z}. \tag{15}$$

Here $a(z)$ and $b(z)$ are the complex amplitudes of the forward and backward waves, respectively. According to this definition,

$$\frac{\partial \hat{E}^{(1)}(z)}{\partial z} = i\omega\left[a(z)e^{i\omega z} - b(z)e^{-i\omega z}\right], \tag{16}$$

and therefore

$$\frac{\partial a(z)}{\partial z} e^{i\omega z} + \frac{\partial b(z)}{\partial z} e^{-i\omega z} = 0. \tag{17}$$

Now by substituting Eq. (15) and the first derivative of Eq. (16) into Eq. (13), and taking Eq. (17) into account, we obtain explicit form of the function $f\left(z, \omega\right)$:

$$f\left(z, \omega\right) = 2i\omega \frac{\partial a(z)}{\partial z} e^{i\omega z}. \tag{18}$$

The boundary conditions at the dielectric (assuming no reflections) and mirror (complete reflection) interfaces are given as:

$$a(z=0) = 0,\ \hat{E}^{(1)}(z=\tilde{L}) = 0. \tag{19}$$

The expressions for forward and backward waves in the general form follow from Eqs. (17) and (18):

$$\begin{aligned} a(z) &= \int_0^z f\left(z', \omega\right) \frac{1}{2i\omega} e^{-i\omega z'}\, dz' + c_a, \\ b(z) &= -\int_0^z f\left(z', \omega\right) \frac{1}{2i\omega} e^{i\omega z'}\, dz' + c_b, \end{aligned} \tag{20}$$

where the constants c_a and c_b are determined from the boundary conditions (19):

$$c_a = 0,\ c_b = a(\tilde{L}) \exp(2i\omega\tilde{L}) - \int_0^{\tilde{L}} f\left(z', \omega\right) \frac{1}{2i\omega} e^{i\omega z'}\, dz', \tag{21}$$

It can be checked by direct substitution that Eqs. (15) and (20) are exact solutions of Eq. (13).

Now we can determine the nonlinear contribution to the reflected wave at the dielectric boundary $z = 0$:

$$\begin{aligned} E_r^{(1)}(0,t) &= \mathcal{F}(b(z=0)) = \mathcal{F}(c_b) = \\ &= \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{-i\omega t}\, d\omega \frac{1}{2i\omega}\int_0^{\tilde{L}} f(z',\omega) \times \left[e^{i\omega(2\tilde{L}-z')} - e^{i\omega z'}\right] dz' = \\ &= -\frac{1}{2}\int_0^{\tilde{L}} dz' \int_{-\infty}^{t} \left[f\left(z', t' - 2\tilde{L} + z'\right) - f(z', t' - z')\right] dt' . \end{aligned} \tag{22}$$

By substituting the explicit form of the source $f(z,t)$ (that follows from the initial Eq. (10)) into Eq. (22), we derive the final expression that defines the addition to the reflected wave due to the nonlinearity of the medium, which together with Eq. (12) and according to Eq. (8) provides a general asymptotic expression for the field of the wave reflected from metal mirror with nonlinear dielectric layer:

$$\begin{aligned} E_r(0,t) = -E_i\left(t - 2\tilde{L}\right) - \frac{L_w}{L_{\text{nl}}}\left(\int_0^{\tilde{L}} \frac{\partial}{\partial t}\left[\left(E^{(0)}\left(z', t - 2\tilde{L} + z'\right)\right)^3 - \right.\right. \\ \left.\left. -\left(E^{(0)}(z', t - z')\right)^3\right] dz'\right), \end{aligned} \tag{23}$$

where

$$E^{(0)}(z,t) = E_i(t - z) - E_i\left(t + z - 2\tilde{L}\right). \tag{24}$$

To evaluate the changes due to the interaction with the counter-propagating wave reflected from the mirror, we perform a comparison with the case of one-directional pulse propagation through a nonlinear dielectric layer of thickness $2\tilde{L}$, such that the total propagation distance through dielectric is the same as in the reflection geometry considered above. We use the same analytical approach as for the reflected pulse to derive the expression for the transmitted pulse. In this case boundary conditions are:

$$a(z=0) = 0, b\left(z = 2\tilde{L}\right) = 0. \tag{25}$$

and therefore the constants $c_a = 0$ and $c_b = \int_0^{2\tilde{L}} f(z', \omega) \frac{1}{2i\omega} e^{i\omega z'} dz'$. Then we can find the nonlinear contribution to the transmitted wave at the boundary $z = 2\tilde{L}$:

$$E_t^{(1)}\left(2\tilde{L}, t\right) = \mathcal{F}\left(a\left(z = 2\tilde{L}\right) e^{i\omega 2\tilde{L}}\right) = \\ = -\frac{1}{2}\int_0^{2\tilde{L}} dz' \int_{-\infty}^{t} \left[f\left(z', t' - 2\tilde{L} + z'\right)\right] dt' . \tag{26}$$

Finally, by following the procedure described above we obtain the asymptotic expression for the transmitted pulse:

$$E_t\left(z = 2\tilde{L}, t\right) = E_t^{(0)}(z, t) + E_t^{(1)}(z, t) = E_i\left(t - 2\tilde{L}\right) - \\ -\frac{L_w}{L_{\mathrm{nl}}}\left(\int_0^{2\tilde{L}} \frac{\partial}{\partial t}\left(E^{(0)}\left(z', t + z' - 2\tilde{L}\right)\right)^3 dz'\right), \tag{27}$$

where

$$E^{(0)}(z, t) = E_i(t - z) . \tag{28}$$

In the following, we compare solutions in Eqs. (23) and (27) to distinguish the effects due to counter-directional interactions.

3.2. Third harmonic generation by long pulses

For quasi-monochromatic incident pulses, the primary effects in thin layer of nonlinear dielectric media are self-phase modulation and third-harmonic generation. For long pulses we can consider the limiting case of continuous waves (CW), with the incident monochromatic electric field $E_i(t) = E^{(0)}(t) = \sin(\omega t)$. For such a wave the spectrum of the second term on the right-hand side of Eqs. (23) and (27), that describe harmonic generation, can be rearranged as follows:

$$E_t^{(1)}\left(\tilde{L}, t\right) = E_t^{(\omega)} + E_t^{(3\omega)} + c.c. = \\ = -\frac{3}{4} e^{i\omega t} e^{-2i\omega\tilde{L}} \tilde{L}\omega + \frac{3}{4} e^{3i\omega t} e^{-6i\omega\tilde{L}} \tilde{L}\omega + c.c. \tag{29}$$

and

$$
\begin{aligned}
E_r^{(1)}\left(\tilde{L},t\right) &= E_r^{(\omega)} + E_r^{(3\omega)} + c.c. \\
&= -\frac{3}{32} i e^{i\omega t}\left(8 - e^{2i\tilde{L}\omega} - 8e^{-4i\tilde{L}\omega} + e^{-6i\tilde{L}\omega} + 24e^{-2i\tilde{L}\omega}\tilde{L}\omega\right) \\
&+ \frac{1}{32} i e^{3i\omega t}\left(2 - 9e^{-2i\tilde{L}\omega} + 18e^{-4i\tilde{L}\omega} - 18e^{-8i\tilde{L}\omega} + 9e^{-10i\tilde{L}\omega}\right. \\
&\left. -2e^{-12i\tilde{L}\omega} + 24e^{-6i\tilde{L}\omega}\tilde{L}\omega\right) + c.c.
\end{aligned}
\tag{30}
$$

We now analyze the amplitudes of third-harmonic components defined through the Fourier transform as:

$$
\hat{E}_{t,r}^{(3\omega)}\left(\tilde{L}\right) = \int_{-\infty}^{+\infty} E_{t,r}\left(\tilde{L},t\right) e^{-i3\omega t}\, dt. \tag{31}
$$

The calculated third harmonic dependencies on the layer thickness $\tilde{L}$ are presented in Fig. 2. We observe an oscillatory dependence for wave reflected from a mirror with nonlinear dielectric coating (solid line), with zero at $\tilde{L} \simeq 0.86292$ and first enhanced resonance at $\tilde{L} = 4/3$. For comparison, the dashed line indicates the harmonic which would be generated in a layer of width $2\tilde{L}$ with no reflections, i.e. when counter-propagating interactions are excluded. We see that counter-propagating wave interactions can significantly (by two times) enhance THG or reduce it to zero. This shows that even in CW regime, counter-propagating interactions cannot be neglected in thin layers, and the full wave Eq. (1) should be used instead of the conventional coupled-mode Eqs. (4). Such investigation is useful to predict and understand effects for few cycle pulses. However we note that CW regime is not practical from experimental point of view, since high average power can lead to material damage even for pulses with duration 120 fs in visible and IR spectral range [38], but in terahertz range where large nonlinearities were predicted [35], we can expect the regime described above for lower intensities without medium breakdown.

3.3. Spectral transformations of few-cycle pulses

We now study the reflection of an input Gaussian pulses of arbitrary duration:

$$
E_i(t) = E^{(0)}(t) = E_0 \exp\left(-\frac{t^2}{\tau_0^2}\right)\sin\left(\frac{\pi t}{2}\right), \tag{32}
$$

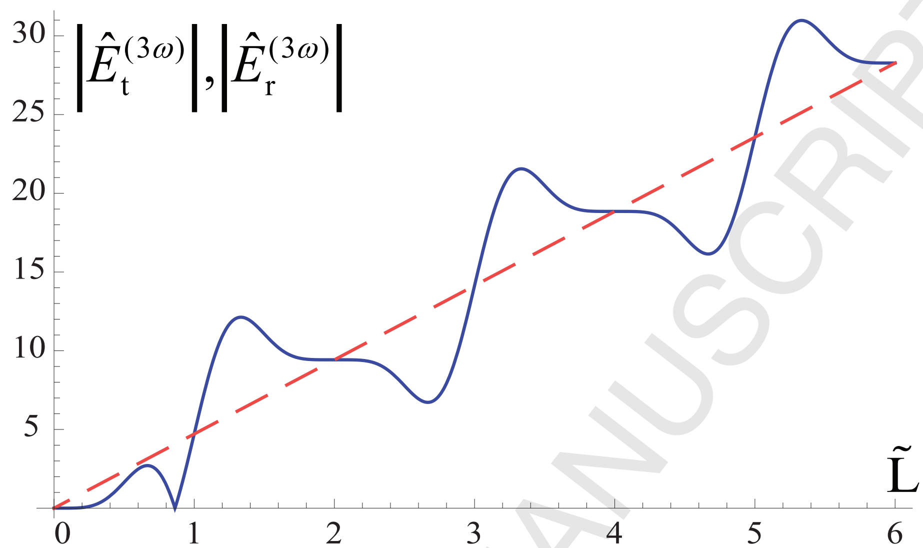


Figure 2: Third-harmonic component of wave reflected from a mirror with a nonlinear dielectric layer of thickness $\tilde{L}$ (blue solid line) compared with transmission through a layer of thickness $2\tilde{L}$ without counter-directional interactions (dashed red line).

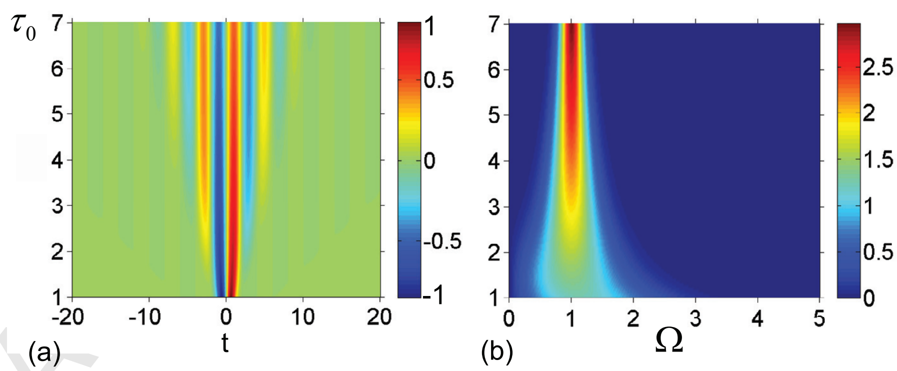


Figure 3: The incident pulse (a) amplitude of the temporal electric field profile $E_i\,(t)$ and (b) modulus of the spectrum $|\mathcal{F}\,(E_i\,(t))|$ vs. the pulse duration (energy of the pulse for each duration is constant). t and Ω are normalized time and frequency, respectively.

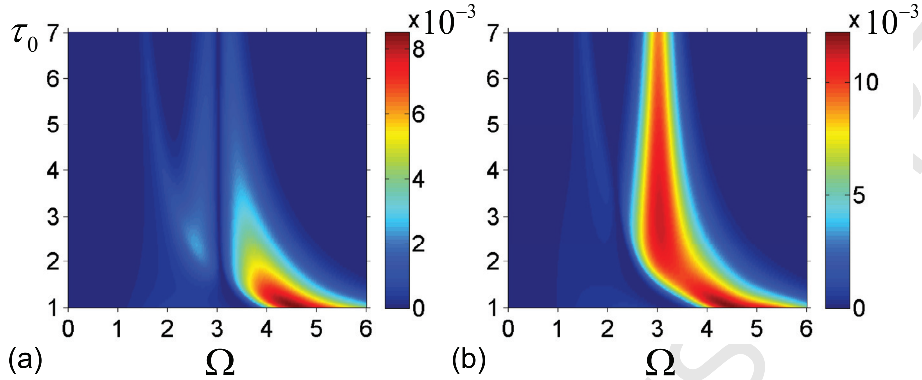


Figure 4: Spectrum changes of the reflected pulse δF due to the effect of cubic nonlinearity vs. the pulse duration for different dielectric layer thickness (a) $\tilde{L} = 0.86292$ and (b) $\tilde{L} = 4/3$.

where $\tau_0 = 4t_0/T_c$ is the normalized pulse duration, t_0 is the pulse duration in fs, T_c is its central period. In numerical simulations, we keep the pulse energy,

$$W = \int_{-\infty}^{+\infty} E_i^2\left(t'\right)\, dt', \tag{33}$$

fixed (normalized to unity in dimensionless variables) by accordingly adjusting initial pulse amplitude E_0, to provide a comparison between pulses of different duration. We consider the nonlinear coefficient value of $L_w/L_{\rm nl} = 0.006$, which satisfies the validity of Picard's method of successive approximations used to obtain the analytical solution.

First, we study the effect of varying the pulse duration. The incident electric field profile vs. the pulse duration is shown in Fig. 3(a), and modulus of the corresponding pulse spectrum, $F_i(\omega) = \mathcal{F}\left(E_i\left(t\right)\right)$, is illustrated in Fig. 3(b). We consider the range of pulse duration down to just one optical cycle duration ($\tau_0 = 1$ in normalized units).

To characterize the pulse transformation due to the effect of cubic nonlinearity, we plot in Fig. 4(a,b) the differences between modulus of the spectra of the reflected and input radiation $\delta F = ||F_r| - |F_i||$, where $F_r\left(\omega\right) = \mathcal{F}\left(E_r\left(0,t\right)\right)$. This spectral transformation can be directly measured experimentally, whereas a reconstruction of the electric field profile is a challenging problem. In Fig. 4(a) the dimensionless layer thickness corresponds to the zero reflection on triple

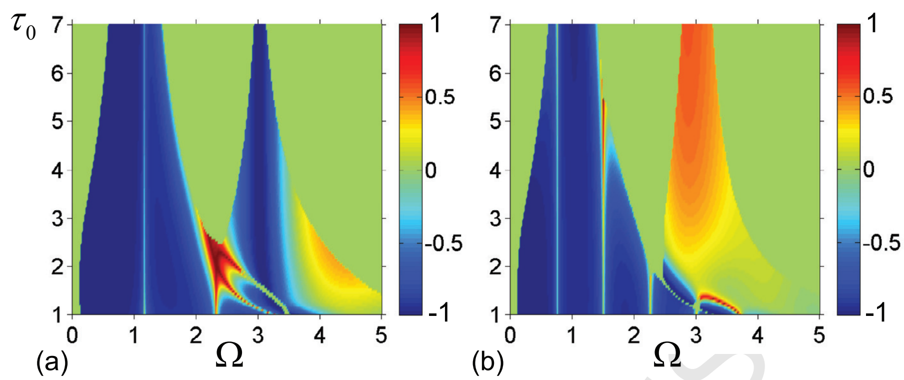


Figure 5: Contributions due to counter-directional interactions to the spectrum changes of the reflected pulse δS vs. the pulse duration for different dielectric layer thickness (a) $\tilde{L} = 0.86292$ and (b) $\tilde{L} = 4/3$.

frequency for long pulses ($\tilde{L} = 0.86292$). Interestingly, in this case the third harmonic of the central frequency gets suppressed even for ultra-short pulses. In Fig. 4(b) the thickness corresponds to the maximum of reflected third harmonic for long input pulse ($\tilde{L} = 4/3$). In this regime strong third harmonic is observed for long and intermediate pulse durations, whereas it gets shifted to the higher frequency region for ultra-short pulses. The latter effect has a general nature for short pulses [7].

We perform further analysis to distinguish the effects due to counter-directional interactions by comparing pulse reflection from a mirror coated with a dielectric layer with thickness $\tilde{L}$ and one-directional pulse transmission through a layer of thickness $2\tilde{L}$. Specifically, we determine the relative spectrum changes as

$$\delta S = \frac{S_r(\omega) - S_t(\omega)}{S_r(\omega) + S_t(\omega)}, \tag{34}$$

where

$$S_r(\omega) = \left| |F_r(\omega)|^2 - |F_i(\omega)|^2 \right|,$$
$$S_t(\omega) = \left| |F_t(\omega)|^2 - |F_i(\omega)|^2 \right|,$$

$F_t(\omega) = \mathcal{F}\left(E_t\left(2\tilde{L}, t\right)\right)$ is the spectrum of the wave that propagated through a layer of thickness $2\tilde{L}$ without counter-directional interactions determined by

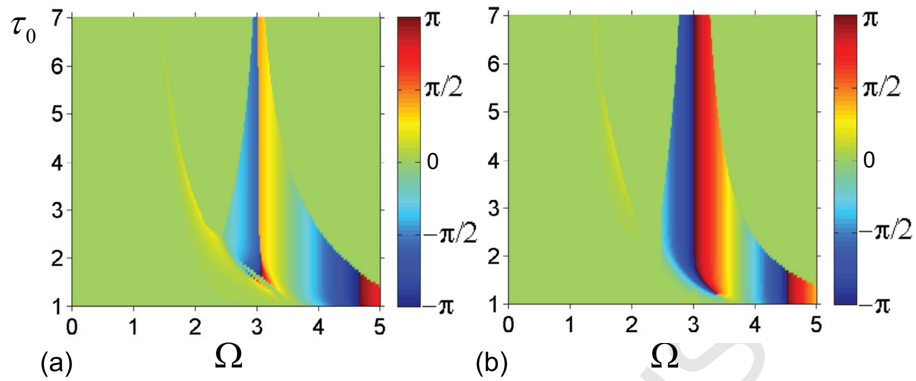


Figure 6: Contributions due to counter-directional interactions to the nonlinear phase shift $dF_{\rm cd}$ vs. the pulse duration for different dielectric layer thickness (a) $\tilde{L} = 0.86292$ and (b) $\tilde{L} = 4/3$.

Eq. (27). We also identify a nonlinear phase shift due to the counter-directional interactions,

$$dF_{\rm cd} = \arg\left(\frac{F_r}{F_t}\right). \tag{35}$$

The results are presented in Figs. 5, 6. We see that there is a strong effect of counter-propagating interactions on third-harmonic generation, which results in its suppression [Fig. 5(a)] or enhancement [Fig. 5(b)] depending on the layer thickness. There are also noticeable difference in higher-harmonic amplitudes for ultra-short pulses, and associated differences in the nonlinear phase shifts [Fig. 6].

We note however that nonlinear phase shifts are not visible at the fundamental frequency in Figs. 6(c,d). To investigate this further, it is instructive to consider the limit of long pulses. Using expressions (29) and (30), we obtain the nonlinear phase shift on fundamental frequency due to effects of self- and cross-phase modulation:

$$\Delta\phi_{\rm nl,t} = \frac{3}{4}\tilde{L}\omega\frac{L_w}{L_{\rm nl}} \tag{36}$$

$$\Delta\phi_{\rm nl,r} = \frac{9}{4}\tilde{L}\omega\frac{L_w}{L_{\rm nl}} \tag{37}$$

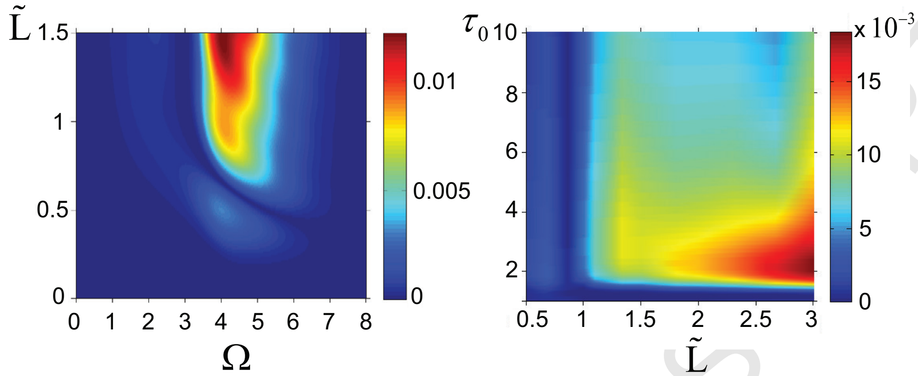


Figure 7: (a) Spectrum changes of the reflected pulse (δF) vs. the nonlinear layer thickness $\tilde{L}$, for the incident pulse duration $\tau_0 = 1.2$. (b) Reflected third harmonic vs. the thickness $\tilde{L}$ for different pulse durations.

We see that $\Delta\phi_{\rm nl,r}$ is three times that of $\Delta\phi_{\rm nl,t}$, properly recovering the well-established result for quasi-CW regime [29]. Using these expressions, we estimate the phase shift due to counter-directional interactions as approximately 10^{-2} for $L_w/L_{\rm nl} = 0.006$. Such value can be detected in experiment, although it is not visible in Figs. 6(c,d) due to the color scale extending over large resonant phase shifts at higher harmonics.

We further consider in more detail the high frequency generation dependence on layer thickness. In Fig. 7(a) we plot the spectrum changes due to the effect of cubic nonlinearity (δF) for the pulse duration of $\tau_0 = 1.2$. For such ultrashort pulse, the generation of the third harmonics reaches its minimum as the maximum of spectral density is moved to the higher frequency region [7]. We observe that such harmonic generation increases for larger (but still of the order of wavelength) layer thickness. Fig. 7(b) illustrates the dependence of third-harmonic on the pulse duration and layer thickness. We see that the third harmonic reaches maximum for short pulses with optimal two-cycle duration.

4. Conclusions

We investigated theoretically few-cycle optical pulse reflection from a perfect metal mirror covered with a nonlinear dielectric layer, and developed a general asymptotic analytical solution in the limit of negligible dispersion and diffraction for small layer thickness. We identified nonlinear transformations of the electric field profiles and optical spectra of the reflected pulses, and performed comparison with the regime of one-directional propagation. We demonstrated that nonlinear counter-directional interactions between the incident and reflected pulse can enhance or fully suppress third-harmonic generation of the central frequency, and also lead to increased spectral broadening for ultra-short pulses. These results suggest a possibility to experimentally detect a fundamentally important effect of counter-propagating interactions between the leading and trailing sections of the pulse.

Acknowledgements

This work was financially supported by Government of Russian Federation, Grant 074-U01 and by the Australian Research Council (Discovery Project DP130100135 and DP160100619).

References

[1] M. Wegener, Extreme Nonlinear Optics: An Introduction, Springer-Verlag, Berlin, 2005. `doi:10.1007/b137953`.
URL `http://dx.doi.org/10.1007/b137953`

[2] A. Nazarkin, Nonlinear optics of intense attosecond light pulses, Phys. Rev. Lett. 97 (16) (2006) 163904–4. `doi:10.1103/PhysRevLett.97.163904`.
URL `http://dx.doi.org/10.1103/PhysRevLett.97.163904`

[3] T. Brabec, F. Krausz, Nonlinear optical pulse propagation in the single-cycle regime, Phys. Rev. Lett. 78 (17) (1997) 3282–3285. `doi:10.1103/`

PhysRevLett.78.3282.
URL http://dx.doi.org/10.1103/PhysRevLett.78.3282

[4] P. Kinsler, Limits of the unidirectional pulse propagation approximation, J. Opt. Soc. Am. B 24 (9) (2007) 2363–2368. doi:10.1364/JOSAB.24.002363.
URL http://dx.doi.org/10.1364/JOSAB.24.002363

[5] A. N. Berkovsky, S. A. Kozlov, Y. A. Shpolyanskiy, Self-focusing of few-cycle light pulses in dielectric media, Phys. Rev. A 72 (4) (2005) 043821–9. doi:10.1103/PhysRevA.72.043821.
URL http://dx.doi.org/10.1103/PhysRevA.72.043821

[6] O. A. Mokhnatova, S. A. Kozlov, Nonlinear reflection of a femtosecond spectral supercontinuum, JETP 106 (2) (2008) 218–227. doi:10.1134/S1063776108020027.
URL http://dx.doi.org/10.1134/S1063776108020027

[7] A. A. Drozdov, S. A. Kozlov, A. A. Sukhorukov, Y. S. Kivshar, Self-phase modulation and frequency generation with few-cycle optical pulses in nonlinear dispersive media, Phys. Rev. A 86 (5) (2012) 053822–10. doi:10.1103/PhysRevA.86.053822.
URL http://dx.doi.org/10.1103/PhysRevA.86.053822

[8] A. A. Drozdov, A. A. Sukhorukov, S. A. Kozlov, Spatio-temporal dynamics of single-cycle optical pulses and nonlinear frequency conversion, Int. J. Mod. Phys. B 28 (12) (2014) 1442007–10. doi:10.1142/S0217979214420077.
URL http://dx.doi.org/10.1142/S0217979214420077

[9] A. V. Kim, S. A. Skobelev, D. Anderson, T. Hansson, M. Lisak, Extreme nonlinear optics in a kerr medium: Exact soliton solutions for a few cycles, Phys. Rev. A 77 (4) (2008) 043823–6. doi:10.1103/PhysRevA.77.043823.
URL http://dx.doi.org/10.1103/PhysRevA.77.043823

[10] M. A. Bakhtin, S. A. Kozlov, Formation of a sequence of ultrashort signals in a collision of pulses consisting of a small number of oscillations of the light field in nonlinear optical media, Opt. Spectrosc. 98 (3) (2005) 425–430. doi:10.1134/1.1890523.
URL http://dx.doi.org/10.1134/1.1890523

[11] E. M. Buyanovskaya, S. A. Kozlov, Dynamics of the fields of counterpropagating light pulses of a few oscillations in nonlinear dielectric media, JETP Lett. 86 (5) (2007) 297–301. doi:10.1134/S0021364007170031.
URL http://dx.doi.org/10.1134/S0021364007170031

[12] J. Renger, R. Quidant, L. Novotny, Enhanced nonlinear response from metal surfaces, Opt. Express 19 (3) (2011) 1777–1785. doi:10.1364/OE.19.001777.
URL http://dx.doi.org/10.1364/OE.19.001777

[13] A. A. Mani, L. Dreesen, C. Humbert, P. Hollander, Y. Caudano, P. A. Thiry, A. Peremans, Development of a two-color picosecond optical parametric oscillator, pumped by a nd : Yag laser mode locked using a nonlinear mirror, for doubly-resonant sum frequency generation spectroscopy, Surf. Sci. 502 (2002) 261–7. doi:10.1016/S0039-6028(01)01954-9.
URL http://dx.doi.org/10.1016/S0039-6028(01)01954-9

[14] R. W. Boyd, Nonlinear Optics, 3rd Edition, Academic Press, San Diego, 2008.
URL http://www.sciencedirect.com/science/book/9780123694706

[15] G. Klemens, Y. Fainman, Optimization-based calculation of optical nonlinear processes in a micro-resonator, Opt. Express 14 (21) (2006) 9864–9872. doi:10.1364/OE.14.009864.
URL http://dx.doi.org/10.1364/OE.14.009864

[16] D. S. Bethune, Optical harmonic-generation and mixing in multilayer media - analysis using optical transfer-matrix techniques, J. Opt. Soc. Am. B 6 (5)

(1989) 910–916. doi:10.1364/JOSAB.6.000910.
URL http://dx.doi.org/10.1364/JOSAB.6.000910

[17] M. G. Martemyanov, T. V. Dolgova, A. A. Fedyanin, Optical third-harmonic generation in one-dimensional photonic crystals and microcavities, JETP 98 (3) (2004) 463–477. doi:10.1134/1.1705697.
URL http://dx.doi.org/10.1134/1.1705697

[18] V. G. Bespalov, S. A. Kozlov, Y. A. Shpolyanskiy, I. A. Walmsley, Simplified field wave equations for the nonlinear propagation of extremely short light pulses, Phys. Rev. A 66 (1) (2002) 013811–10. doi:10.1103/PhysRevA.66.013811.
URL http://dx.doi.org/10.1103/PhysRevA.66.013811

[19] P. Kinsler, G. H. C. New, Few-cycle pulse propagation, Phys. Rev. A 67 (2) (2003) 023813–8. doi:10.1103/PhysRevA.67.023813.
URL http://dx.doi.org/10.1103/PhysRevA.67.023813

[20] M. Kolesik, J. V. Moloney, Nonlinear optical pulse propagation simulation: From Maxwell's to unidirectional equations, Phys. Rev. E 70 (3) (2004) 036604–11. doi:10.1103/PhysRevE.70.036604.
URL http://dx.doi.org/10.1103/PhysRevE.70.036604

[21] T. Schafer, C. E. Wayne, Propagation of ultra-short optical pulses in cubic nonlinear media, Physica D 196 (1-2) (2004) 90–105. doi:10.1016/j.physd.2004.04.007.
URL http://dx.doi.org/10.1016/j.physd.2004.04.007

[22] H. Leblond, S. V. Sazonov, I. V. Mel'nikov, D. Mihalache, F. Sanchez, Few-cycle nonlinear optics of multicomponent media, Phys. Rev. A 74 (6) (2006) 063815–8. doi:10.1103/PhysRevA.74.063815.
URL http://dx.doi.org/10.1103/PhysRevA.74.063815

[23] H. Leblond, D. Mihalache, Optical solitons in the few-cycle regime: recent theoretical results, Rom. Rep. Phys. 63 (2011) 1254–1266.

[24] K. Glasner, M. Kolesik, J. V. Moloney, A. C. Newell, Canonical and singular propagation of ultrashort pulses in a nonlinear medium, Int. J. Optics 2012 (2012) 898. doi:doi:10.1155/2012/868274.
URL http://www.hindawi.com/journals/ijo/2012/868274/

[25] S. A. Kozlov, V. V. Samartsev, Fundamentals of femtosecond optics, Woodhead Publishing, Cambridge, 2013.
URL http://www.sciencedirect.com/science/book/9781782421283

[26] M. Mero, B. R. Clapp, J. C. Jasapara, W. G. Rudolph, D. Ristau, K. Starke, J. Krger, S. Martin, W. Kautek, On the damage behavior of dielectric films when illuminated with multiple femtosecond laser pulses, Opt. Eng. 44 (2005) 44 – 44 – 7. doi:10.1117/1.1905343.
URL http://dx.doi.org/10.1117/1.1905343

[27] D. N. Nguyen, L. A. Emmert, D. Patel, C. S. Menoni, W. Rudolph, Transient phenomena in the dielectric breakdown of hfo2 optical films probed by ultrafast laser pulse pairs, Appl. Phys. Lett. 97 (19) (2010) 191909. arXiv:http://dx.doi.org/10.1063/1.3511286, doi:10.1063/1.3511286.
URL http://dx.doi.org/10.1063/1.3511286

[28] S. A. Kozlov, Polarization self-action of pulses consisting of a few oscillations of optical field in dielectric media, Opt. Spectrosc. 84 (1998) 887–889.

[29] G. P. Agrawal, Nonlinear Fiber Optics, 5th Edition, Academic Press, New York, 2013.
URL http://www.sciencedirect.com/science/book/9780123970237

[30] W. S. Kolthammer, D. Barnard, N. Carlson, A. D. Edens, N. A. Miller, P. N. Saeta, Harmonic generation in thin films and multilayers, Phys. Rev. B 72 (4) (2005) 045446–15. doi:10.1103/PhysRevB.72.045446.
URL http://dx.doi.org/10.1103/PhysRevB.72.045446

[31] S. Mao, F. Quéré, S. Guizard, X. Mao, R. Russo, G. Petite, P. Martin, Dynamics of femtosecond laser interactions with dielectrics, Appl. Phys. A

79 (7) (2004) 1695–1709. doi:10.1007/s00339-004-2684-0.
URL http://dx.doi.org/10.1007/s00339-004-2684-0

[32] S.-H. Cho, H. Kumagai, K. Midorikawa, In situ observation of dynamics of plasma formation and refractive index modification in silica glasses excited by a femtosecond laser, Opt. Commun. 207 (1-6) (2002) 243–253. doi:doi:10.1016/S0030-4018(02)01410-4.
URL http://www.sciencedirect.com/science/article/pii/S0030401802014104

[33] S. A. Shtumpf, A. A. Korolev, S. A. Kozlov, Dynamics of the strong field of a few-cycle optical pulse in a dielectric medium, Bulletin of the Russian Academy of Sciences: Physics 71 (2) (2007) 147–150. doi:10.3103/S1062873807020013.
URL http://link.springer.com/article/10.3103%2FS1062873807020013

[34] Synthetic crystal quartz.
URL http://www.tydexoptics.com/materials1/for_transmission_optics/crystal_quartz/

[35] K. Dolgaleva, D. V. Materikina, R. W. Boyd, S. A. Kozlov, Prediction of an extremely large nonlinear refractive index for crystals at terahertz frequencies, Phys. Rev. A 92 (2015) 023809–8. doi:10.1103/PhysRevA.92.023809.
URL http://link.aps.org/doi/10.1103/PhysRevA.92.023809

[36] G. A. Korn, T. M. Korn, Mathematical Handbook for Scientists and Engineers, 2nd Edition, Mcgraw-Hill, New York, 1968.
URL http://store.doverpublications.com/0486411478.html

[37] P. R. West, S. Ishii, G. V. Naik, N. K. Emani, V. M. Shalaev, A. Boltasseva, Searching for better plasmonic materials, Laser Photon. Rev. 4 (6) (2010) 795–808. doi:10.1002/lpor.200900055.

URL http://onlinelibrary.wiley.com/doi/10.1002/lpor.200900055/abstract

[38] D. von der Linde, H. Schüler, Breakdown threshold and plasma formation in femtosecond laser–solid interaction 13 (1) (1996) 216–222. doi:10.1364/JOSAB.13.000216.
URL http://josab.osa.org/abstract.cfm?URI=josab-13-1-216