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Control of the third-harmonic generation efficiency upon interaction of few-cycle waves in nonlinear media

M.A. Knyazev, S.A. Kozlov

Abstract. **It is shown that the interaction of few-cycle waves propagating at an angle to each other in media with instantaneous cubic nonlinearity may lead to a considerable increase in the third-harmonic generation efficiency without energy redistribution between the intersecting beams with a wide spectrum.**

Keywords: *nonlinear optics, few-cycle waves, third-harmonic generation, optical transistor.*

1. Introduction

Analysis of the interaction of intense light waves propagating in an optical media at an angle to each other is a classical problem of nonlinear optics [1]. This problem has been already thoroughly studied in the last century for quasi-monochromatic waves. Diffraction of light beams on a dynamic refractive index grating induced in the region of their overlap in the nonlinear medium may cause radiation energy redistribution over directions, including energy redistribution between the beams [2]. This phenomenon is interesting for some applications, for example, for enhancing the time contrast of high-power femtosecond laser pulses [3] or for designing ultrafast all-optical switches and transistors [4, 5]. However, it is important to note that the energy exchange between intersecting quasi-monochromatic beams does not occur in media with instantaneous refractive index nonlinearity [6].

In recent decades, the development of efficient sources of high-intensity few-cycle optical waves [7–9] required a fresh approach to the traditional problems of nonlinear optics [10–12]. This occurred because, first, the spectrum of such wave packets with extremely small number of oscillations becomes very broad and, second, they may propagate through a material without its breakdown at considerably higher intensities than quasi-monochromatic radiation. Nonlinear effects, which are weak in the field of quasi-monochromatic radiation up to the breakdown intensity, can be very strong in the field of extremely short pulses due to increasing breakdown threshold. In particular, as the number of oscillations in a pulse decreases to three and smaller, the self-focusing efficiency at a given power excess over the critical value may noticeably decrease [13] with a considerable increase in the third-harmonic generation efficiency [14, 15] up to the energy conversion efficiency of several percent.

The reflection of co- and counterpropagating pulses from a refractive index inhomogeneity, which is induced in the medium by a high-intensity ultrashort pulse and propagates with a pulse velocity, was considered in works [16, 17]. It was theoretically demonstrated that the radiation frequency considerably increases due to the Doppler shift on an induced high-velocity inhomogeneity of the medium. It was shown in [18, 19] that the interaction of collinear counterpropagating few-cycle waves may change the third-harmonic generation efficiency in a nonlinear medium. The specific features of reflection and refraction of ultrashort signal pulses, which propagate at an angle to a high-intensity pulse, from a refractive index inhomogeneity induced by this pulse were considered in [20–22]. It was shown that noncollinear interaction may cause a frequency shift, a change in the velocity, and a deviation from the initial trajectory of the signal light beam.

Let us emphasise that the analysis of the strong changes in the character of propagation of one of the intersecting ultrashort wave packets was performed in [16, 17, 20–22] in the approximation of a given refractive index inhomogeneity induced in the nonlinear medium by the other wave. The estimates in these works were obtained using typical femtosecond radiation parameters and characteristics of transparent dielectric media, i.e., under the conditions at which the dominant nonlinearity of the polarisation response has an electronic nature and this response can be with good accuracy considered as instantaneous.

The aim of the present work was to find the key regularities of the interaction between intense few-cycle waves propagating at an angle to each other in transparent dielectric media with an instantaneous nonlinearity. We analysed this interaction strictly as a self-consistent wave problem with boundary conditions at the entrance of two femtosecond optical beams into a nonlinear medium without introduction of the artificial concept of induced nonlinear refractive index in the medium. The analysis showed that, both for quasi-monochromatic radiation and ultrashort wave packets (i.e., wave packets with a very broad spectrum), energy redistribution between colliding wave packets in isotropic dielectric media with instantaneous cubic nonlinearity does not occur. In this case, the interaction between these packets may lead to a significant (by several times) increase in the third-harmonic generation efficiency. By the third-harmonic generation efficiency we mean the energy ratio of the generated triple-frequency radiation to the initial radiation at the entrance to the nonlinear medium.

The enhancement of generation of new frequencies in a primary wave due to the intersection with another intense

M.A. Knyazev, S.A. Kozlov ITMO University, Kronverkskii prosp. 49, 197101 St. Petersburg, Russia;
e-mail: knyazev.michael@gmail.com, kozlov@mail.ifmo.ru

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wave packet in a nonlinear medium corresponds to the main property of the optical transistor, i.e., to amplification of light by another light [23]. Thus, at radiation intensities that do not cause irreversible changes of the material, the considered light-control system makes it possible to achieve the optical transistor effect with a response time of several periods of optical field oscillations.

2. Mathematical model of the interaction of few-cycle waves in a medium with instantaneous nonlinearity

The dynamics of spectral density g of a TE-polarised two-dimensional nonparaxial radiation, including the case of two waves propagating at an angle to each other, in a homogeneous isotropic dielectric medium with an instantaneous cubic nonlinearity can be described by the equation [24]

$$\frac{\partial^2 g}{\partial z^2} = [k^2(\omega) - k_x^2]g = -\frac{\omega^2\varepsilon_{\rm nl}}{c^2}\frac{1}{(2\pi)^4} \times \iiiint_{-\infty}^{\infty} g(\omega-\omega',\, k_x-k_x',\, z)g(\omega'-\omega'',\, k_x'-k_x'',\, z) \times g(\omega'',\, k_x'',z)\,{\rm d}\omega'{\rm d}k_x'{\rm d}\omega''{\rm d}k_x'', \qquad (1)$$

where

$$g(\omega,k_x,z) = \iint_{-\infty}^{\infty} E(t,x,z)\exp[-{\rm i}(\omega t + k_x x)]\,{\rm d}t{\rm d}x, \qquad (2)$$

ω and k_x are the temporal and spatial frequencies, $k(\omega) = \omega n(\omega)/c$ is the wave number, $n(\omega)$ is the frequency dependence of the refractive index of the medium, $\varepsilon_{\rm nl}$ is its nonlinear permittivity, c is the speed of light in vacuum, t is the time, x and z are the spatial Cartesian coordinates, and $E(t,x,z)$ is the strength of the electric field of radiation polarised perpendicular to the xz plane. The axis z in (1) is the direction along which the nonparaxial radiation field is $E \to 0$ at any finite distance z at $x \to \pm\infty$ [24], i.e., this direction, for example, for wave packets intersecting at an angle to each to other, may coincide with the axis of one of the beams, which we will call the primary beam.

Note that the field analogue of Eqn (1) that describes the slit diffraction of waves in a nonlinear medium has the form [25]

$$\frac{\partial^2 E}{\partial z^2} + \frac{\partial^2 E}{\partial x^2} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\int_0^{\infty}\varepsilon(t')E(t-t')\,{\rm d}t' = \frac{\varepsilon_{\rm nl}}{c^2}\frac{\partial^2 E^3}{\partial t^2}, \qquad (3)$$

and the dispersion characteristics of the medium are related by the expression

$$n^2(\omega) = \int_0^{\infty}\varepsilon(t)\exp({\rm i}\omega t)\,{\rm d}t.$$

The spectral density of two waves propagating at an angle α to each other at the boundary of a nonlinear medium (at $z = 0$) is written as

$$g(\omega,k_x,0) = g_1(\omega,k_x) + g_2(\omega,k_x),$$

$$g_1(\omega,k_x) = {\rm i}g_{01}\exp\Big[-\Big(\frac{k_x}{\Delta k_x}\Big)^2\Big] \times\Big\{\exp\Big[-\Big(\frac{\omega+\omega_0}{\Delta\omega}\Big)^2\Big] - \exp\Big[-\Big(\frac{\omega-\omega_0}{\Delta\omega}\Big)^2\Big]\Big\}, \qquad (4)$$

$$g_2(\omega,k_x) = \frac{{\rm i}g_{02}}{\cos\alpha}\exp\Big\{-\Big[\frac{(k_x+\omega\sin\alpha/V)^2}{\Delta k_x^2\cos^2\alpha}\Big]\Big\} \times\Big\{\exp\Big[-\Big(\frac{\omega+\omega_0}{\Delta\omega}\Big)^2\Big] - \exp\Big[-\Big(\frac{\omega-\omega_0}{\Delta\omega}\Big)^2\Big]\Big\}\exp[-{\rm i}(\omega\tau + k_x h)],$$

where $g_1(\omega,k_x)$ is the spectrum of the primary wave packet, $g_2(\omega,k_x)$ is the spectrum of the wave acting on the primary wave, g_{01} and g_{02} are the maxima of their spectral densities, Δk_x and $\Delta\omega$ are the widths of the spatial and temporal spectra of the wave packets at the entrance to the medium, ω_0 is the centre wavelength of the radiation, $V = c/n(\omega_0)$ is the phase velocity of the incident wave (the difference between its phase and group velocities and their dispersion at the entrance to the medium are neglected), α is the angle between the z axis (which coincides with the primary wave propagation direction) and the axis of the light beam acting on the primary wave, τ is the delay time of the incident beam, and h is the transverse shift of the incident beam along the x axis.

The field analogues of boundary conditions (4) takes the form

$$E(t,x,0) = E_1(t,x) + E_2(t,x),$$

$$E_1(t,x) = E_0\exp\Big[-\Big(\frac{x}{\Delta x}\Big)^2\Big]\exp\Big[-\Big(\frac{t}{\Delta t}\Big)^2\Big]\sin(\omega_0 t), \qquad (5)$$

$$E_2(t,x) = E_{02}\exp\Big\{-\Big[\frac{(x-h)\cos\alpha}{\Delta x}\Big]^2\Big\} \times\exp\Big\{-\Big[\frac{t-\tau-(x-h)\sin\alpha/V}{\Delta t}\Big]^2\Big\} \times\sin\Big\{\omega_0\Big[t-\tau-\frac{(x-h)\sin\alpha}{V}\Big]\Big\},$$

where field amplitudes are $E_{01} = \Delta\omega\Delta k_x g_{01}/2\pi$ and $E_{02} = \Delta\omega\Delta k_x g_{02}/2\pi$, $\Delta x = 2/\Delta k_x$ is the transverse size of wave packets at the entrance to the medium, and $\Delta t = 2/\Delta\omega$ is the initial duration of the wave packets. Another boundary condition that determines the propagation directions of the waves will be considered in the linearised form [25]

$$\frac{\partial g(\omega,k_x,z)}{\partial z}\Big|_{z=0} = -{\rm i}g(\omega,k_x,0)\sqrt{k^2(\omega)-k_x^2}. \qquad (6)$$

For convenience and simplicity of calculations, we normalise Eqn (1) and boundary conditions (4) and (6) by introducing dimensionless variables

$$\tilde g = \frac{g}{g_{01}},\quad \tilde\omega = \frac{\omega}{\omega_0},\quad \tilde k_x = \frac{k_x}{\Delta k_x},\quad \tilde z = zk_0, \qquad (7)$$

where $k_0 = \omega_0 n(\omega_0)/c$.

Then, Eqn (1) and boundary conditions (4) and (6) in variables (7) take the form

$$\frac{\partial^2\tilde g}{\partial\tilde z^2} + [(1+\mu_{\rm dsp}\Delta\tilde n(\tilde\omega)\tilde\omega^2 - \mu_{\rm dfr}\tilde k_x^2]\tilde g = -\mu_{\rm nl}\frac{\tilde\omega^2}{\Delta\tilde\omega^2}\times$$

$$\times \iiiint_{-\infty}^{\infty} \tilde{g}(\tilde{\omega}-\tilde{\omega}', \tilde{k}_x-\tilde{k}_x', \tilde{z})\tilde{g}(\tilde{\omega}'-\tilde{\omega}'', \tilde{k}_x'', z)$$

$$\times \tilde{g}(\tilde{\omega}'', \tilde{k}_x'', \tilde{z})\,\mathrm{d}\tilde{\omega}'\mathrm{d}\tilde{k}_x'\mathrm{d}\tilde{\omega}''\mathrm{d}\tilde{k}_x'' \qquad (8)$$

$$\tilde{g}_1(\tilde{\omega},\tilde{k}_x) = \mathrm{i}\exp(-\tilde{k}_x^2)$$

$$\times\left\{\exp\left[-\left(\frac{\tilde{\omega}+1}{\Delta\tilde{\omega}}\right)^2\right] - \exp\left[-\left(\frac{\tilde{\omega}-1}{\Delta\tilde{\omega}}\right)^2\right]\right\},$$

$$\tilde{g}_2(\tilde{\omega},\tilde{k}_x) = \mathrm{i}\frac{\eta}{\cos\alpha}\exp\left[-\left(\tilde{k}_x + \frac{\tilde{\omega}\sin\alpha}{\sqrt{\mu_{\mathrm{dfr}}}}\right)^2\right]$$

$$\times\left\{\exp\left[-\left(\frac{\tilde{\omega}+1}{\Delta\tilde{\omega}}\right)^2\right] - \exp\left[-\left(\frac{\tilde{\omega}-1}{\Delta\tilde{\omega}}\right)^2\right]\right\} \qquad (9)$$

$$\times\exp\{-2\pi\mathrm{i}[\tilde{\omega}\tilde{\tau} + n(\omega_0)\tilde{k}_x\tilde{h}]\},$$

$$\frac{\partial\tilde{g}}{\partial\tilde{z}}\Big|_{\tilde{z}=0} = -\mathrm{i}\tilde{g}(\tilde{\omega},\tilde{k}_x,0)\sqrt{[1+\mu_{\mathrm{dsp}}\Delta\tilde{n}(\tilde{\omega})]\tilde{\omega}^2 - \mu_{\mathrm{dfr}}\tilde{k}_x^2}, \qquad (10)$$

where

$$\Delta\tilde{\omega} = \frac{\Delta\omega}{\omega_0},\ \ \Delta\tilde{n}(\tilde{\omega}) = \frac{n(\tilde{\omega}) - n(\omega_0)}{\Delta n_{\mathrm{dsp}}},\ \ \Delta n_{dsp} = n\left(\omega_0 + \frac{\Delta\omega}{2}\right)$$

$$-n\left(\omega_0 - \frac{\Delta\omega}{2}\right),\ \ \eta = \frac{g_{02}}{g_{01}},\ \ \tilde{h} = \frac{h}{\lambda_0},\ \ \tilde{\tau} = \frac{\tau}{T_0},$$

$\lambda_0 = 2\pi c/\omega_0$ is the centre wavelength, and $T_0 = 2\pi/\omega_0$ is the wave oscillation period.

All products of normalised variables, functions, and functionals in Eqn (8) are on the order of unity at least at the initial stage of the spectral density dynamics of radiation in the nonlinear medium. Therefore, the dimensionless parameters

$$\mu_{\mathrm{dfr}} = \left(\frac{\Delta k_x}{k_0}\right)^2,\ \ \mu_{\mathrm{dsp}} = 2\frac{\Delta n_{\mathrm{dsp}}}{n(\omega_0)},\ \ \mu_{\mathrm{nl}} = \frac{8}{3}\pi^2\frac{\Delta n_{\mathrm{nl}}}{n(\omega_0)}$$

(where

$$\Delta n_{\mathrm{nl}} = \frac{3}{8}\frac{\varepsilon_{\mathrm{nl}}}{n(\omega_0)}E_{01}^2 = n_2 I_{01},$$

n_2 is the nonlinear refractive index of the medium, and I_{01} is the primary wave intensity at the entrance to the medium) allow one to estimate the relative influence of diffraction, dispersion, and nonlinear response of the medium, respectively, on the radiation propagation character.

Let us estimate these parameters, e.g., for femtosecond pulses propagating in quartz glass. Assuming that the transverse size of the slit beam (at the e^{-1} level) at the entrance to the nonlinear medium contains 20 wavelengths ($L = 2\Delta x/\lambda_0 = 20$), the number of complete oscillations in the pulse (at the e^{-1} level) is equal to seven ($N = 2\Delta t/T_0 = 7$), centre wavelength is λ_0 = 800 nm, and the radiation intensity is $I = 1.3 \times 10^{12}$ W cm^{-2}, we obtain for quartz glass ($n_2 = 2.9 \times 10^{-16}$ cm^2 W^{-1}) $\mu_{\mathrm{dfr}} = 4 \times 10^{-4}$, $\mu_{\mathrm{dsp}} = 1.7 \times 10^{-3}$, and $\mu_{\mathrm{nl}} = 7.2 \times 10^{-3}$ (Δn_{dsp} and $n(\omega_0)$ were calculated using the data for quartz glass dispersion from [26]). As is seen from the estimates, the dynamics of the spectrum of a beam propagating under these conditions is determined by the medium nonlinearity, because of which below we will neglect the beam dispersion. However, of course, it is clear that, with decreasing number of field oscillations in a pulse (with respect to the considered case), dispersion may become the strongest effect. The diffraction term, despite its smallness, will be taken into account because we will consider the interaction of two beams propagating at an angle to each other rather than a single pulse propagation. The diffraction broadening of beams at the given parameters is expected to be insignificant.

To solve Eqn (8), we used the Crank–Nicolson numerical scheme with an adaptive step [27]. The convolution in the right-hand side of Eqn (8) was calculated using the fast Fourier transform algorithm. The calculation was performed using a software module, the Fortran language, and the OpenMP parallelism [28].

3. Regularities of the interaction of few-cycle waves in a medium with instantaneous nonlinearity

Figure 1 shows the results of calculation of the interaction of a wave packet (with width $\Delta x = 14\lambda_0$, duration $\Delta t = 3.5T_0$, and intensity ensuring $\mu_{\mathrm{nl}} = 7.2 \times 10^{-3}$) propagating along the z axis with a wave packet (with width $\Delta x = 14\lambda_0$, duration $\Delta t = 3.5T_0$, relative intensity $\eta^2 = 0.75$, transverse shift $\tilde{h} = 57$, and delay time $\tilde{\tau} = -7$) propagating at the angle $\alpha = 0.2$ to the primary wave. This figure presents the plane images of the space–time distribution of the electric field strength of wave packets. The light-grey and dark-grey bands correspond to the positive and negative field strengths, respectively (we used normalised spatial coordinate $\tilde{x} = x/\lambda_0$ and time $\tilde{t} = t/T_0$).

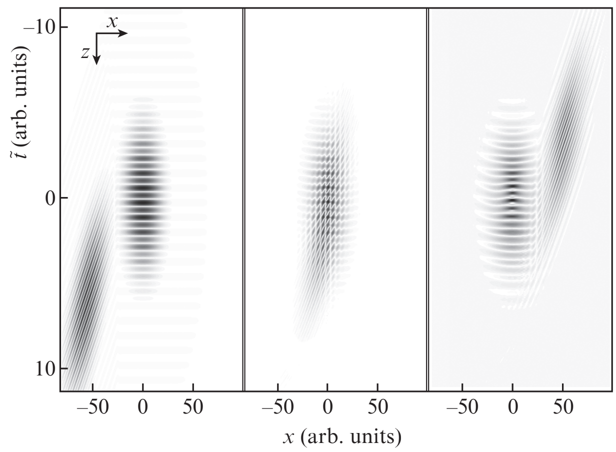

Figure 1. Space–time distribution of the electric field of waves (a) before (at $\tilde{z} = 0$), (b) during ($\tilde{z} = 2.5\times10^3$), and (c) after ($\tilde{z} = 5.0\times10^3$) their interaction.

One can see from Fig. 1 that the waves at the entrance to the nonlinear medium (at $\tilde{z} = 0$) are spatially separated and can be considered as noninteracting (Fig. 1a). The waves intersect at the distance $\tilde{z} = 2.5\times10^3$ (Fig 1b), pass through each other, and become spatially separated again, after which they propagate independently (Fig. 1c). Self-phase modulation of wave packets becomes noticeable at the distance $\tilde{z} = 5.0\times10^3$. The nonlinear phase incursion in the intense part of the wave reaches $\pi/4$.

As the duration of optical pulses decreases to only several periods of the electric field oscillations, there occurs an interchange of dominant nonlinear effects. The third-harmonic generation becomes important. Its efficiency may reach several percent [14, 15]. The change in this efficiency due to the interaction of the ultrashort optical pulse with another pulse is shown in Fig. 2, which presents the dependences of the wave-packet energies at the fundamental and the generated triple frequencies on the relative intensity of the incident wave η^2 at the distance $\tilde{z} = 5.0 \times 10^3$. One can see that, at the considered few-cycle wave intensity in the absence of an incident wave ($\eta = 0$), about 2.5% of energy is spent on the triple frequency radiation. However, the action of an intersecting wave leads to a considerable increase in the generated third-harmonic energy. For example, this energy increases twofold at $\eta^2 = 0.85$. It is important that the total energy of the primary wave in this case does not change. This means that energy transfer from the incident wave does not occur, and the energy of radiation at the triple frequency increases due to an increase in the third-harmonic generation rate during the interaction of waves.

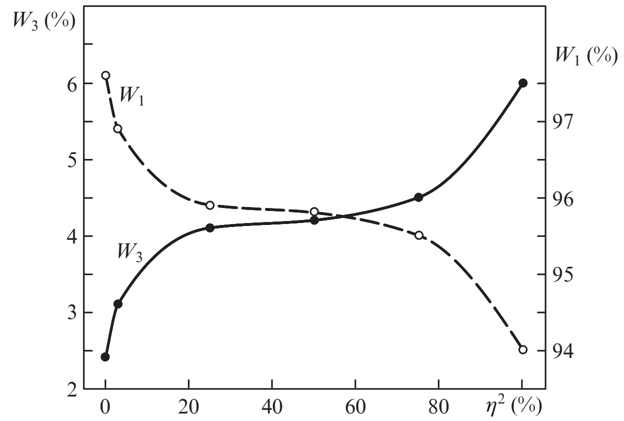

Figure 2. Dependences of the primary wave energy at the triple (W_3) and fundamental (W_1) frequencies on the relative intensity of the incident wave η^2. The points on the curves correspond to the numerical experiments performed in this work.

Figures 3 and 4 show the changes in the time (at $k_x = 0$) and space–time spectra of the third harmonic of the primary wave at the distance $\tilde{z} = 5.0 \times 10^3$ at different relative intensities η^2 of the incident wave. The solid curves in Fig. 3 correspond to the calculated points in Fig. 2. Figure 3 demonstrates that an increase in the third-harmonic energy with increasing intensity of the incident wave is accompanied by broadening and reshaping of the time spectrum in this frequency range. As follows from Fig. 4, the spatial spectrum of the third harmonic also broadens.

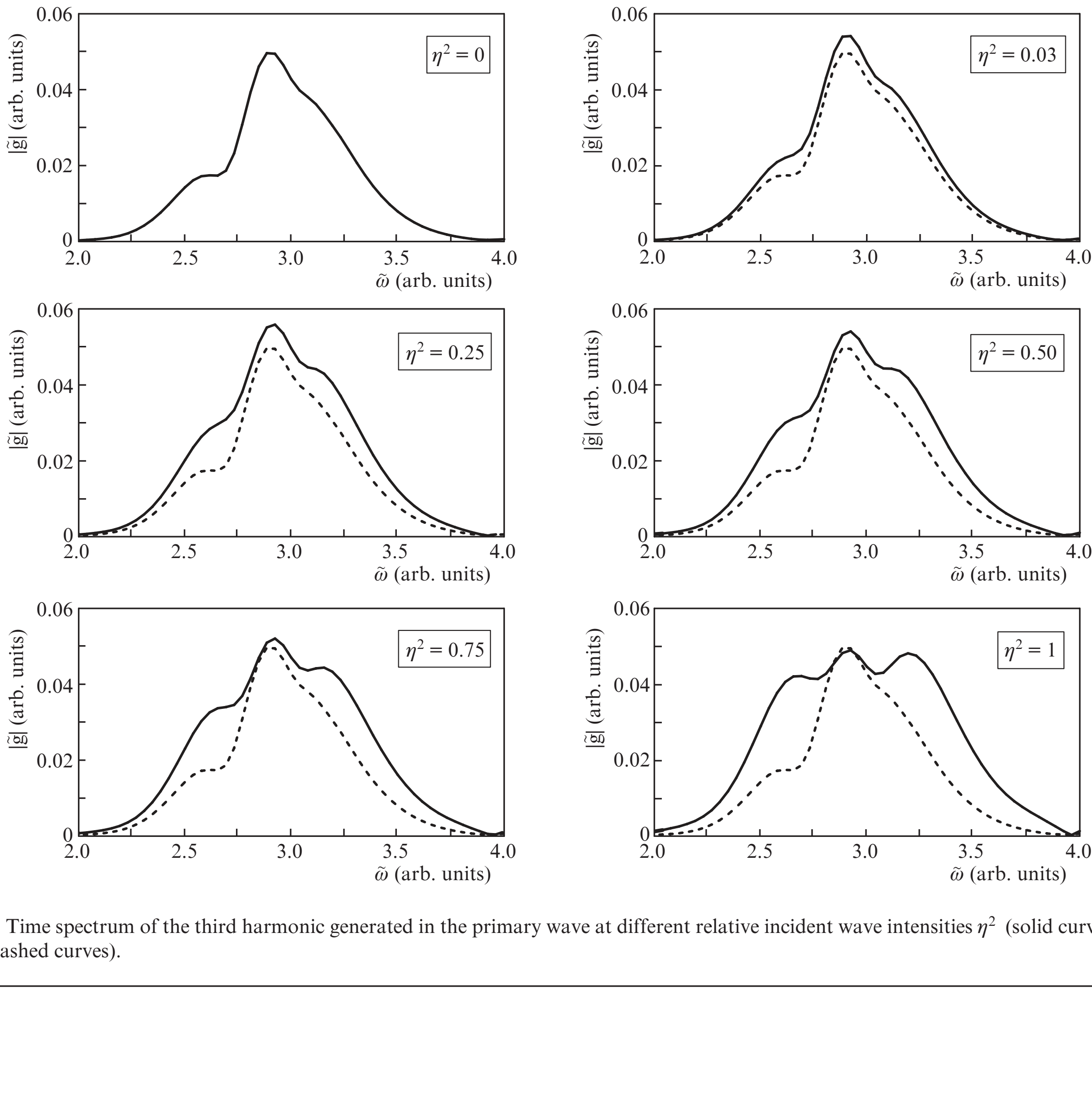

Figure 3. Time spectrum of the third harmonic generated in the primary wave at different relative incident wave intensities η^2 (solid curves) and at $\eta^2 = 0$ (dashed curves).

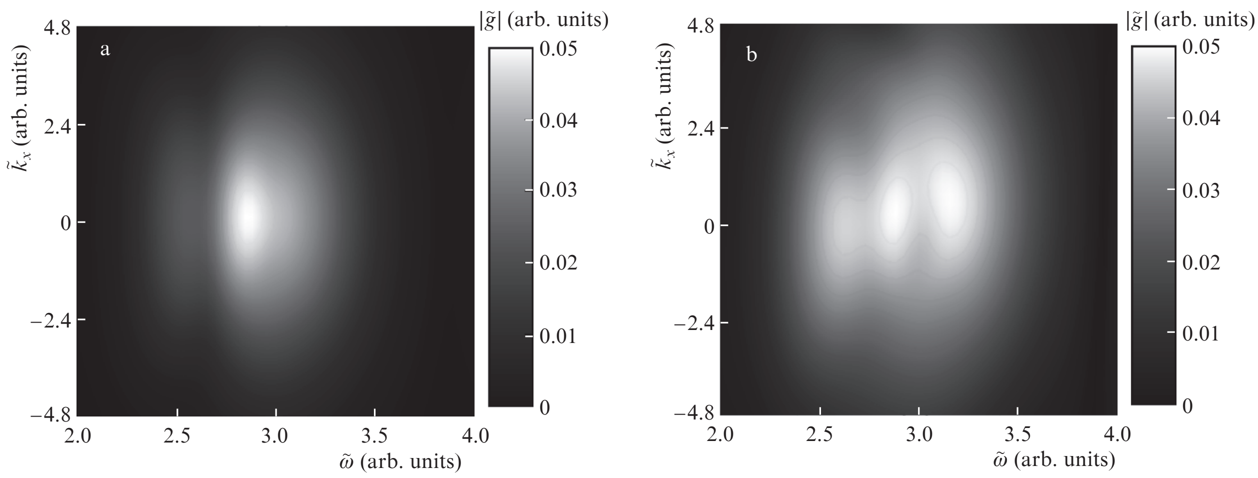

Figure 4. Space–time spectrum of the third harmonic of the primary wavelength at relative incident wave intensities η^2 = (a) 0 and (b) 1.

4. Conclusions

In this work, we showed that, similar to the case of quasi-monochromatic radiation, the cross-interaction of few-cycle waves with a broad spectrum in a medium with an instantaneous cubic nonlinearity causes no energy redistribution between intersecting optical beams. However, control of light by light in this case is possible, i.e., the third-harmonic generation efficiency in the primary wave can be considerably (by several times) increased due to the action of another intense wave propagating at an angle to the primary wave. Thus, using a simple scheme, one can create an optical transistor with a response time of only several periods of light field oscillations.

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