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Harmonic Generation with Single-Cycle Light Pulses

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Abstract. We study theoretically spatiotemporal pulse dynamics in cubic nonlinear media with instant response, nonresonant absorption and normal group velocity dispersion and reveal new features of harmonic generation when the pulse duration is reduced, including the suppression of third-harmonic generation for single-cycle light pulses.

1 Introduction

Rapid progress over the past two decades in the area of ultrafast optics has led to the production, manipulation, and control of optical pulses with durations down to a few optical cycles [1]. The generation of intense short pulses has opened a door to the study of novel effects in extreme nonlinear optics [2] and attosecond physics [3]. The experimental advances have motivated extensive theoretical studies of the propagation of few-cycle pulses in nonlinear media, including the analysis of spatiotemporal dynamics and self-focusing [4–7]. However, many aspects of harmonic generation with few-cycle optical pulses remain largely unexplored. In this work, we report on a systematic theoretical analysis of spatiotemporal pulse dynamics in cubic nonlinear media with normal group dispersion and reveal new features of harmonic generation when the pulse duration is reduced, including the suppression of third-harmonic generation for single-cycle light pulses.

2 Propagation of single-cycle light pulses in nonlinear media

We analyse theoretically the propagation of intense single-cycle light pulses in isotropic dielectric media with instant cubic (Kerr) nonlinearity and nonresonant absorption. We consider unidirectional paraxial propagation corresponding to a beam width much larger than the optical wavelength, and assume that the wavelength spectrum is within the region of normal group-velocity-dispersion. Under such conditions, the evolution of the electric field $E(z, x, y, t)$ of an optical wave can be modelled by the following equation [4]:

$$\frac{\partial E}{\partial z} + K_0 E + \frac{N_0}{c}\frac{\partial E}{\partial t} - K_1 \frac{\partial^2 E}{\partial t^2} - a\frac{\partial^3 E}{\partial t^3} + gE^2 \frac{\partial E}{\partial t} = \frac{c}{2N_0}\Delta_\perp \int_{-\infty}^{t} E dt', \quad (1)$$

where z is the distance along the propagation direction, $\Delta_\perp = \partial^2/\partial x^2 + \partial^2/\partial y^2$ is the transverse Laplace operator, t is time, c is the speed of light in vacuum. Parameters N_0, a and K_0, K_1 characterize the dependence of linear refractive index n_0 and absorption coefficient κ of the medium on the radiation frequency ω:

$$n_0(\omega) = N_0 + ac\omega^2, \quad (2)$$

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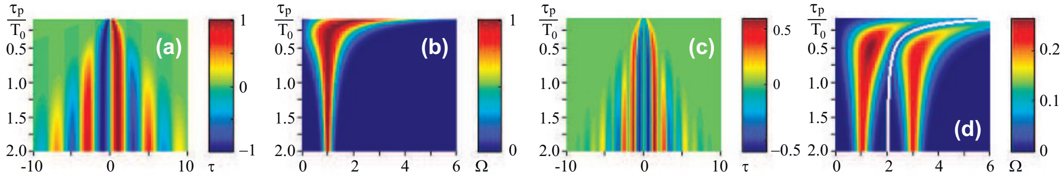

Fig. 1. (a,b) Input Gaussian pulse (a) field and (b) spectrum vs. normalized duration. (c,d) Nonlinearly-induced corrections to the pulse (c) field and (d) spectrum vs. normalized duration. The solid line in (d) marks the minimum of the spectral correction. τ, Ω are normalized time and frequency, respectively.

$$\kappa(\omega) = \frac{K_0 c}{\omega} + K_1 c\omega. \tag{3}$$

Coefficient g characterizes the Kerr-type nonlinear response, and it is related to the cubic nonlinear susceptibility n_2 as $g = 2n_2/c$. Absorption (3) will be neglected in our simulations. We underline that equation (1) is formulated for the electric field E of the optical wave, and it is suitable for theoretical modeling of ultra-short pulse evolution with very broad spectrum, including the case of light pulses with single-cycle field oscillation.

The regimes of pulse dynamics can be classified according to the relative values of the dispersion $L_{\text{disp.}} = \pi^2\lambda_0 N_0 \big/ 16\Delta n_{\text{disp.}}$, diffraction $L_{\text{dif.}} = 8r_0^2\big/\lambda_0$, and nonlinear $L_{\text{nl.}} = \lambda_0 N_0/16\Delta n_{\text{nl.}}$ lengths, where $\lambda_0 = cT_0/N_0$ is the central wavelength, $\Delta n_{\text{disp.}} = ac\omega_0^2$ is the modification of the refractive index due to dispersion, $\Delta n_{\text{nl.}} = (1/2)n_2 E_0^2$ is the nonlinearly-induced change of the optical refractive index, E_0 is the characteristic input field amplitude, $\omega_0 = 2\pi/T_0$ is the central frequency.

We study the process of harmonic generation, considering the incident Gaussian pulse of the form

$$E^{(0)}(0,t) = E_0 \exp\left(-t^2\big/\tau_p^2\right)\sin(\omega_0 t), \tag{4}$$

where E_0 is characteristic field amplitude and τ_p is the input pulse duration.

We first identify the general features of harmonic generation depending on the pulse duration by considering a case when the effects of dispersion and diffraction can be neglected. In this case, we obtain an approximate analytical solution of equation (1) using Picard's method of successive approximations over a small parameter $L_{\text{wave}}/L_{\text{nl.}}$, where $L_{\text{wave}} = \lambda_0/4$. Our results are summarized in Fig. 1. We show the input pulse profile in Fig. 1(a) and the corresponding input spectrum in Fig. 1(b). The nonlinearly-induced corrections to the pulse profile and spectrum at the output are shown in Figs. 1(c) and (d), respectively. We see that the spectral correction has two maxima due to combined action of self-phase modulation and high frequency generation, and we show the frequency where the correction completely vanishes with solid line in Fig. 1(d). For a long pulse duration (e.g. $\tau_p\big/T_0 = 2$) there is no second-harmonic generation while the third harmonic is generated efficiently. It fully agrees with the well-known results for quasi-monochromatic pulses. However as the pulse duration is reduced, the spectral maximum and minimum get shifted to higher frequencies. For the pulse duration $\tau_p\big/T_0 = 0.3$, when the input pulse is single-cycle containing just one electric field oscillation [see Fig. 1(a)], the third-harmonic generation vanishes. This is a remarkably surprising result, since the third-harmonic generation is one of the fundamental effects in optical media with Kerr-type nonlinearity.

We then investigate the effect of temporal dispersion through extensive numerical simulations, and find that the nonlinear spectral correction also vanishes for second-harmonic for long pulses. For short pulses, there appear series of minima in nonlinear spectral corrections when dispersion is taken into account, and in particular the third-harmonic generation may be suppressed for certain durations of single-cycle light pulses (e.g. $\tau_p/T_0 \approx 0.4$, $\tau_p/T_0 \approx 0.7$).

Finally, we analyze the self-action features of axisymmetric paraxial single-cycle light pulses under the combined effects of cubic nonlinearity, temporal dispersion and spatial diffraction by numerical simulations of Eq. (1). In Fig. 2 we present two-dimensional contour plots of the electric field and spectral distributions during pulse propagation in a medium with $L_{\text{nl.}} = 7$ mm, $L_{\text{disp.}} = L_{\text{difr.}} = 35$ mm for the transverse beam width at the input of the nonlinear medium $r_0 = 8\lambda_0$. Diffraction induces a

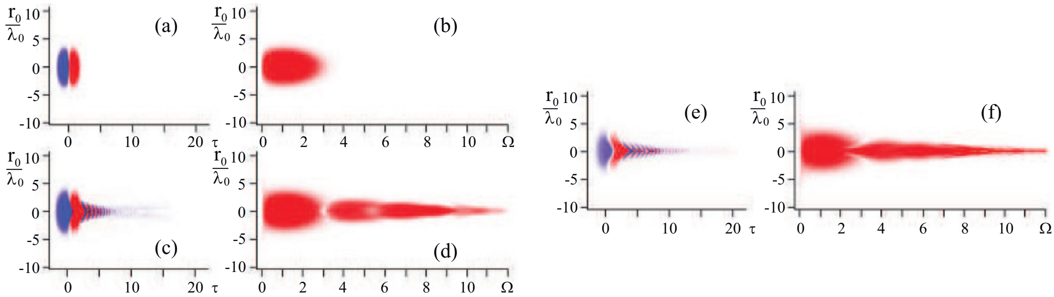

Fig. 2. Evolution of a single-cycle wave with Gaussian transverse distribution. (a,c,e) Spatiotemporal electric field profiles and (b,d,f) modulus of their spectra at distances: (a,b) 0 mm, (c,d) 4 mm and (e,f) 8 mm. Characteristic lengths are $L_{\mathrm{nl.}} = 7$ mm, $L_{\mathrm{disp.}} = L_{\mathrm{difr.}} = 35$ mm.

strong increase in the transverse spectrum size at low frequencies. Combined action of nonlinearity and dispersion lead to the formation of the high frequency narrow wave tail up to twelve central frequencies, and in the presented example at $z = 8$ mm the third-harmonic generation is strongly suppressed [Fig. 2(d)]. This illustrates the generality of the third-harmonic generation suppression effect for single-cycle light pulses. Interestingly, for longer propagation length (e.g. $z = 8$ mm) the third harmonic appears again [Fig. 2(f)].

3 Conclusion

We have studied the spatiotemporal pulse dynamics in nonlinear media based on theoretical analysis and numerical solutions of the model equation for the electric field, which can describe evolution of few-cycle light pulses in Kerr nonlinear media taking into account temporal normal group dispersion and spatial diffraction. We have predicted novel features for the harmonic generation with few-cycle pulses compared to the well-known case of long quasi-monochromatic pulses. In particular, we have predicted analytically and confirmed through spatiotemporal numerical simulations that the generation of the third light harmonic can be suppressed for single-cycle pulses at certain propagation distances. These effects shall be taken into account for the development of nonlinear optical systems operating with ultra-short light pulses.

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