# New nonlinear parametric conversion mechanism for coherent THz generation

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# New Nonlinear Parametric Conversion Mechanism for Coherent THz Generation

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**Abstract.** We propose a new nonlinear mechanism for coherent THz generation in cubic media. We show that this phenomenon can be presented only by $\chi^{(3)}$ parametric process without inclusion of $\chi^{(2)}$ processes. In this work we consider the particular case of generation of THz signal from femtosecond laser pulses propagating in optical fibers (fused silica). On the base of proposed theoretical model we derive a system of nonlinear partial differential equations investigated numerically by means of the split-step Fourier method. The numerical results show a significant coherent THz generation when the dispersion is negligible.

## INTRODUCTION

The process of filamentation is connected with propagation of ultrahigh-intensity femtosecond ($10^{-12}$-$10^{-14}$ sec.) laser pulses through gases and dielectrics. For the first time self-channeling of ultra-short pulses in air with optical field intensities of the order of $5\times10^{13}$ W/cm$^2$ was reported by A. Braun et al. [1]. The single filament is characterized by stable propagation up to 8-10 kilometers in the high atmosphere and by asymmetrical spectrum transformation from the infrared (invisible) region – 800 nm – to the visible region which leads to white spectrum generation [2]. During the filamentation process coherent and non-coherent GHz/THz generation is observed [3,4]. Channel-type propagation of the femtosecond pulse has been obtained also in glasses [5-8]. The first theoretical models for explanation of the filamentation process were focused on the balance between self-focusing and plasma defocusing [1, 9]. In recent experiments, the observation of stable post-filamentation regime at several meters from the laser source with intensity of the order of $10^{11}$ – $10^{12}$ W/cm$^2$ is reported. Such intensity is close to the critical one for self-focusing, but is not enough for plasma grid generation [10-12].

Recently, coherent structures and discrete lines in the spectrum of single filament propagating in different types of glass plates were observed [13]. These discrete strictures cannot be connected with ionization of the medium since the plasma spectrum is continuous. The existence of the discrete lines is explained by avalanche generation of signal waves with THz frequency shift leading to asymmetrical spectral broadening of the filament towards the higher frequencies.

The purpose of our investigations in the present work is to answer the question about the generation of coherent THz wave in cubic media, in particular optical fibers (fused silica). In the first studies on coherent THz generation from femtosecond laser pulses in $\chi^{(3)}$ media the process is explained by optical rectification mechanism which requests strong Second Harmonics (SHs) – $\chi^{(2)}$ mechanisms – with energies at least 10-20 percents of the main wave [2]. However, such strong SHs are not observed in the experiments. In this paper we propose a new nonlinear $\chi^{(3)}$ mechanism for generation of coherent THz emission.

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## THEORETICAL MODEL

### Generation of Signal Waves with THz Spectral Shift

Since the optical fibers are with waveguide structure, the propagation dynamics of laser pulses is described by 1+1 dimensional approximation of the Slowly Varying Nonlinear Amplitude Equation (SVNAE).

The nonlinear evolution of broad-band femtosecond pulses can be described correctly in the frames of cubic nonlinearity $P_i = \chi^{(3)}_{ijkl} E_j E_k E_l$ which includes the third harmonics term. The scalar approximation $\vec{E} = E_x \vec{x}$ of the electrical field on one carrying frequency $\omega_0$ leads to only two components of the tensor of cubic nonlinearity:

$$\chi^{(3)}_{xxxx}(3\omega_0 = \omega_0 + \omega_0 + \omega_0) \text{ and } \chi^{(3)}_{xxxx}(\omega_0 = \omega_0 + \omega_0 - \omega_0), \tag{1}$$

which present the third harmonics $3\omega_0 = 3k_0 v_{ph}$ ($k_0$ is the carrying wave number, $v_{ph}$ is the phase velocity) and the self-action correspondingly.

The scalar equation describing the pulse propagation written in Galilean frame $(z' = z - v_{gr}t;\quad t' = t)$ – moving with the group velocity $v_{gr}$– has the form (the primes are removed for clarity):

$$-2i\frac{k_0}{v_{gr}}\frac{\partial A_x}{\partial t} = -\beta\frac{\partial^2 A_x}{\partial z^2} + k_0^2 n_2\left[\frac{1}{3}A^3{}_x \exp\left(2ik_0\left(z - \left(v_{ph} - v_{gr}\right)t\right)\right) + \left|A_x\right|^2 A_x\right], \tag{2}$$

where $\beta = k_0 v_{gr}^2 k_0''$ is the dimensionless dispersion parameter, $A_x$ is the amplitude envelope and $n_2$ is the nonlinear refractive index. The analysis of Eq. (2) shows that the new generated frequency shift $\omega_{nl}$ is not equal to the third harmonics $3\omega_0 = 3k_0 v_{ph}$, but to $\omega_{nl} = 3\omega_{CEF} = 3k_0\left(v_{ph} - v_{gr}\right)$, with $\omega_{CEF}$ being the Carrier-to-Envelope Frequency (CEF). This frequency $\omega_{nl}$ is in the THz region for solids [13].

Due to the generation of new frequency $\omega_1$ spectrally shifted with THz delay in respect to the main wave $\omega_1 = \omega_0 + \omega_{nl} = \omega_0 + 3\omega_{CEF}$, new components of the nonlinear tensor $\chi^{(3)}$ arise:

$$\chi^{(3)}_{xxxx}(3\omega_0 = \omega_0 + \omega_0 + \omega_0);\ \chi^{(3)}_{xxxx}(\omega_0 = \omega_0 + \omega_1 - \omega_1);\ \chi^{(3)}_{xxxx}(\omega_0 = \omega_0 + \omega_0 - \omega_0),$$
$$\chi^{(3)}_{xxxx}(3\omega_1 = \omega_1 + \omega_1 + \omega_1);\ \chi^{(3)}_{xxxx}(\omega_1 = \omega_1 + \omega_0 - \omega_0);\ \chi^{(3)}_{xxxx}(\omega_1 = \omega_1 + \omega_1 - \omega_1). \tag{3}$$

The system of equations written in Galilean frame in normalized coordinates becomes:

$$\begin{aligned} &i\frac{\partial A_0}{\partial t} = -\beta_0\frac{\partial^2 A_0}{\partial z^2} + \gamma_0\left(\left|A_0\right|^2 A_0 + 2\left|A_1\right|^2 A_0 + A^{*2}_0 A_1 \exp(-i\delta kz)\right) \\ &i\left(\frac{\partial A_1}{\partial t} + \Delta v\frac{\partial A_1}{\partial z}\right) = -\beta_1\frac{\partial^2 A_1}{\partial z^2} + \gamma_2\left(\left|A_1\right|^2 A_1 + 2\left|A_0\right|^2 A_1 + A_0^3 \exp(i\delta kz) + A_1^3 \exp(-i\delta Kz)\right) \end{aligned}, \tag{4}$$

where $\delta k = (k_1 - 3k_0)/2k_0$ and $\delta K = (k_2 - 3k_1)/2k_1$ denote the normalized wave mismatch vectors, $\Delta v = \left(v_{1,gr} - v_{0,gr}\right)/v_{0,gr}$ is the normalized group velocity delay, $\beta_i = k_i'' v_{i,gr}^2 / z_0$ are the dimensionless dispersion parameters and $\gamma_i = n_2\left|A_0\right|^2 k_i z_0^2$ are the nonlinear coefficients with longitudinal spatial dimension $z_0 = v_{gr} t_0$. As the waves are spectrally close, the group velocity delay and the mismatch wave numbers are practically negligible: $\Delta v \ll 1$ and $\delta k \cong \delta K \approx 10^{-2} - 10^{-3}$. The system of equations (4) is not conservative. The nonlinear term $A_1^3 \exp(-i\delta Kz)$ generates a new higher frequency $\omega_2$ with THz spectral shift in respect to $\omega_1$.

In order to interrupt the continuous process of frequency generation we neglect this term. The restricted system, where the nonlinear term $A_1^3 \exp(-i\delta Kz)$ and the group velocity delay $\Delta v$ are neglected, has the form:

$$i\frac{\partial A_0}{\partial t}=-\beta_0\frac{\partial^2 A_0}{\partial z^2}+\gamma_0\left(\left|A_0\right|^2 A_0+2\left|A_1\right|^2 A_0+A^{*2}_0 A_1\exp(-i\delta kz)\right)$$
$$i\frac{\partial A_1}{\partial t}=-\beta_1\frac{\partial^2 A_1}{\partial z^2}+\gamma_1\left(\left|A_1\right|^2 A_1+2\left|A_0\right|^2 A_1+A_0^3\exp(i\delta kz)\right) \quad (5)$$

and obeys the Manley-Raw energy conservation lows.

The process of cascade generation of signal waves with a THz frequency shift leads to energy transfer towards the higher frequencies (*i.e.*, shorter wavelengths) from the infrared region to the visible region. This energy flow is connected with the asymmetric broadening of the spectrum and formation of white continuum. The parametric conversion mechanism working simultaneously with the four-photon parametric wave mixing is proposed for first time in [13]. The developed theoretical model shows very good agreement with the experimental data. The next natural step is ***to find out a mechanism for generation of coherent THz wave***.

## Generation of coherent THz wave

Let us denote the waves and their frequencies in the following manner:

$$\omega_s \xleftarrow{\Delta} \omega_p \xleftarrow{\Delta=\omega_{nl}=\omega_{THz}} \omega_i \ldots\ldots\ldots\ldots\ldots \omega_{THz} \quad (6)$$

with $\omega_p=\omega_0$, $\omega_s,\omega_i$ and $\omega_{THz}$ being the frequencies of the main (pump), signal, idler and THz waves correspondingly. The energy flow is towards the signal wave; *i.e.*, $\omega_s$ is the highest frequency. The spectral distance between the pump and idler wave, and also between the signal wave and pump, is equal to $\omega_{nl}=\omega_{THz}=\Delta$. In this case the following additional components of the nonlinear tensor $\chi^{(3)}$ arise:

$$\chi^{(3)}_{xxxx}\left(\omega_s=\omega_i+\omega_{THz}+\omega_{THz}\right),\ \chi^{(3)}_{xxxx}\left(\omega_i=\omega_s-\omega_{THz}-\omega_{THz}\right),\ \chi^{(3)}_{xxxx}\left(\omega_{THz}=\omega_s-\omega_i-\omega_{THz}\right). \quad (7)$$

The wave synchronisms which the pulses satisfy are

a) Two conditions for energy transfer between THz shifted waves:

$$\omega_s=\omega_p+\Delta=\omega_p+\omega_{THz}, \quad (8)$$
$$\omega_p=\omega_i+\Delta=\omega_i+\omega_{THz}.$$

b) One four-photon parametric mixing (FPPM) condition between three waves

$$2\omega_p=\omega_s+\omega_i. \quad (9)$$

For the generation of coherent THz wave we propose the following **new parametric wave synchronism** (see the scheme (6)):

$$2\omega_{THz}=\omega_s-\omega_i. \quad (10)$$

Thus the system governing the generation of THz wave and the evolution and energy exchange between all the waves takes the form:

$$
\begin{aligned}
i\frac{\partial A_p}{\partial t} &= \beta_p \frac{\partial^2 A_p}{\partial z^2} + \gamma_p \Big[ \left|A_p\right|^2 A_p + 2\left( \left|A_s\right|^2 + \left|A_i\right|^2 + \left|A_{THz}\right|^2 \right) A_p + 2A_p^* A_s A_i \exp(-i\delta_k z) \\
&\qquad + A^{*2}_p A_s \exp(-i\delta_k z) + {}_i^3 \exp(\ \delta_k \ ) \Big] \\
i\frac{\partial A_s}{\partial t} &= \beta_s \frac{\partial^2 A_s}{\partial z^2} + \gamma_s \Big[ \left|A_s\right|^2 A_s + 2\left( \left|A_p\right|^2 + \left|A_i\right|^2 + \left|A_{THz}\right|^2 \right) A_s + A_p^2 A_i^* \exp(i\delta_k z) \\
&\qquad + A_p^3 \exp(i\delta_k z) + A_i A_{THz}^2 \exp\left(i\Delta_k z\right) \Big] \\
i\frac{\partial A_i}{\partial t} &= \beta_i \frac{\partial^2 A_i}{\partial z^2} + \gamma_i \Big[ \left|A_i\right|^2 A_i + 2\left( \left|A_p\right|^2 + \left|A_s\right|^2 + \left|A_{THz}\right|^2 \right) A_i + A_p^2 A_s^* \exp(i\delta_k z) \\
&\qquad + A_p A_i^{*2} \exp(-i\delta_k z) - A_s A_{THz}^{*2} \exp\left(-i\Delta_k z\right) \Big] \\
i\frac{\partial A_{THz}}{\partial t} &= \beta_{THz} \frac{\partial^2 A_{THz}}{\partial z^2} + \gamma_{THz} \Big[ \left|A_{THz}\right|^2 A_{THz} + 2\left( \left|A_p\right|^2 + \left|A_s\right|^2 + \left|A_i\right|^2 \right) A_{THz} + \\
&\qquad + 2A_p^* A_s A_i \exp(-i\Delta_k z) \Big]
\end{aligned}
\quad . \qquad (11)
$$

Here the wave mismatch vector $\delta_k = \left(2k_p - k_s - k_i\right)/k_p$ corresponds to the four-photon parametric wave mixing and to parametric generation of waves with THz spectral delay. The second wave mismatch vector $\Delta_k = \left(2k_{THz} - k_s + k_i\right)/k_{THz}$ is related to the coherent THz generation.

## NUMERICAL SIMULATIONS

The nonlinear system of equations (11) is treated numerically by using the split-step Fourier method. We use localized initial conditions in the form:

$$A_j = A_{0j} \exp\left(i\varphi_j\right) \exp\left(i\Delta k_j z\right) \mathrm{sec}h\left(z / z_{0j}\right), \qquad j = p, s, i, THz, \qquad (12)$$

where $A_{0j}$ are the initial amplitude constants, $\varphi_j$ are the initial phases, $\Delta k_j = 2\pi/\Delta\lambda_j$ are the spectral shifts between the carrying wave numbers of waves, and $z_{0j}$ are the initial widths of pulses. The numerical simulations performed in the present work show significant THz generation when the dispersion is negligible. In our opinion this result is due to the fact that the theoretical model is developed on the base of $\chi^{(3)}$ nonlinear parametric processes – generation of signal waves with THz spectral shift (8), FPPM processes (9) and generation of THz wave (10). As is shown in the experiments, the FPPM processes are effective in optical fibers when $\beta \approx 0$ [14,15,16].

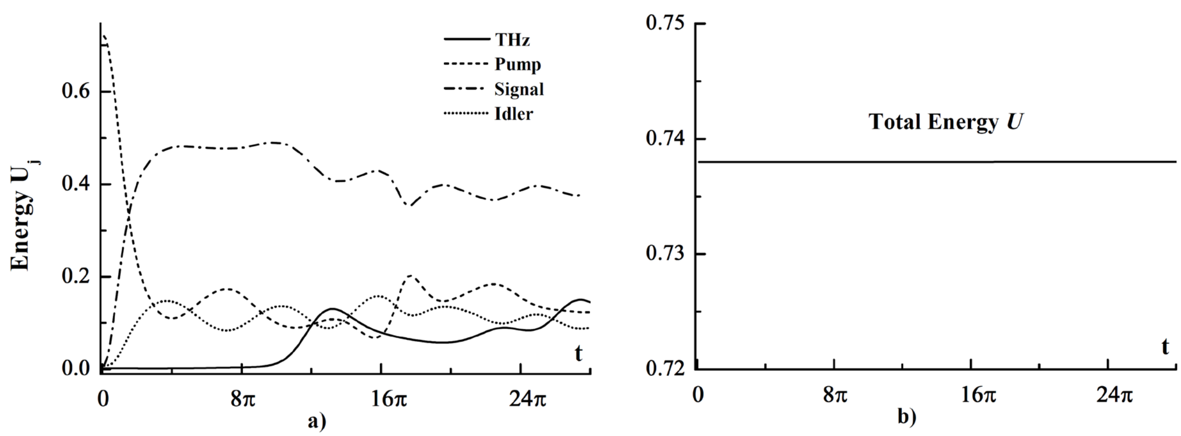


**FIGURE 1.** a) The calculated energies of each generated wave. The difference between the amplitudes of signal and idler waves generates at next stage coherent THz signal (solid line). (b) Conservation of the total energy of system (11) during the process of energy exchange between the components

Figure 1a presents the calculated energies $U_j = \int |A_j|^2 dz$ for the following parameters: $\gamma = 0.8, \beta_j = 0.01$ and initial constants:

$$\begin{aligned} &A_{0p} = 0.6, A_{0s} = A_{0i} = 0.02, A_{0THz} = 0.01, \\ &z_{0p} = 1, z_{0s} = z_{0i} = z_{0THz} = 10, \delta_k = 0.01, \Delta_k = 0.2, \\ &\Delta k_p = 0, \quad \Delta k_s = 1, \quad \Delta k_i = -1; \ \Delta k_{THz} = -10. \end{aligned} \tag{13}$$

As can be seen, initially the parametric generation with THz shift $\omega_s = \omega_p + \omega_{nl}$ and the FPPM process $2\omega_p = \omega_s + \omega_i$ generate the signal and idler waves. Afterwards the process $\omega_p = \omega_i + \omega_{nl}$ causes difference between the signal and idler amplitudes. The obtained difference through the proposed nonlinear mechanism $2\omega_{THz} = \omega_s - \omega_i$ generates coherent THz wave. This is the reason for observation of the phenomenon at a later stage.

The system of nonlinear equations (11) is conservative and obeys the energy conservation lows. The calculated total energy $U = U_p + U_s + U_i + U_{THz}$ in the process of periodical energy exchange between the optical pulses is presented in Figure 1b and confirms the Manley-Raw conditions.

Figure 2 shows the evolution and gradual amplification of coherent THz signal. The THz wave is generated later because a difference between the amplitudes of signal and idler pulses at first has to be obtained.

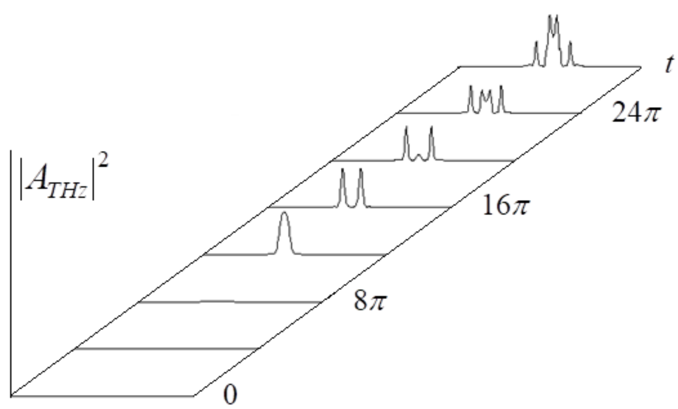


**FIGURE 2.** Generation and evolution of coherent THz signal propagating in optical fiber with weak dispersion

## CONCLUSIONS

We present a new $\chi^{(3)}$ parametric conversion mechanism for generation of coherent THz signal in optical fibers. Our mechanism does not request second harmonics generation ($\chi^{(2)}$ processes) and optical rectification mechanism. We found new components of the cubic tensor which satisfy wave synchronism between the signal, idler and THz waves. This synchronism exists when the signal and idler waves are shifted at THz frequency delay from each other. The THz generation is effective in the regions with weak dispersion of the optical fiber.

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