# FOURIER METHOD USED TO STUDY THE SECOND HARMONIC GENERATION SPECTRUM IN METAMATERIALS

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The nonlinear optical process of second harmonic generation (SHG) resulting from frequency doubling of high-intensity laser radiation in metamaterials has been investigated, with particular attention given to the role of cubic nonlinearity. In analyzing the nonstationary SHG process, a conventional spectral approach was employed. The governing differential equations for the field amplitudes were reformulated in terms of their spectral counterparts, providing a clearer and more accurate representation of the physical mechanisms underlying the frequency-doubling process. Metamaterials characterized by engineered dispersion and a negative refractive index offer exceptional conditions for enhancing nonlinear interactions and achieving controllable harmonic generation [1]. In this work, the effects of self- and cross-phase modulation arising from the third-order nonlinearity of the medium on the spectral and temporal behavior of SHG are examined in detail. It is shown that fine adjustment of the pump intensity or the phase mismatch enables precise tuning of the harmonic spectral peak, thereby facilitating smooth and continuous frequency control of the second harmonic in metamaterials. The controllability waves is the way for the development of compact and tunable nonlinear optical devices with advanced photonic functionalities.

**Keywords:** nonstationary generation spectrum, self- and cross-interaction, negative refraction, Fourier method.
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## INTRODUCTION

The metamaterial can be created from a composite material forming a dielectric matrix, with inclusions providing the resonant properties of the material. A similar approach was used in the development of solid-state lasers, when activator ions were introduced into the matrix, for example, of a crystal, which ultimately determined the physical properties of the laser medium. The electromagnetic properties of the metamaterial are also dictated by the characteristics of the microstructures that are embedded in the composite material. This opens up new prospects for their use [2–4]. For example, the modern development of photonics is associated, in particular, with the technology of developing metamaterials. As is known, in optical information processing systems photons are used as information carriers, and here the problem arises of controlling this data carrier. The discovery of metamaterials contributed to the possibility of controlling light by changing the optical properties of such meta-atoms. In optical switching systems using the phenomenon of bistability, it is proposed to use a sandwich structure. The use of thin films of material opens up new possibilities for the design of photonic crystals [5].

A broad range of nonlinear phenomena in metamaterials has been extensively studied, including harmonic generation, parametric interactions under counter-propagating wave configurations [6-7], self-focusing effects, and other related nonlinear processes [4,8]. In this context, the nonstationary regime of harmonic generation in metamaterials is of particular practical interest, especially due to the unique wave interaction geometries achievable in these engineered media, such as counter-propagating nonlinear waves.

The propagation of a high-intensity electromagnetic wave in a nonlinear medium induces interactions between the pump and harmonic fields, arising from the intrinsic nonlinearity of the medium. Specifically, the effective refractive index of the pump wave depends not only on its own intensity (self-action effect) but also on the intensity of the co-propagating second harmonic. The self-action effect modifies the spatial profile of the laser beam, leading to spectral broadening of the generated radiation. Energy flux analyses for self-focusing and self-defocusing regimes have been reported in [9], while [10] examined single-negative and Kerr-type double-negative metamaterials. Moreover, both theoretical and experimental studies of the harmonic spectrum of ultrashort Gaussian pulses in conventional media have been presented in [11–13], revealing measurable frequency shifts in the harmonic spectra.

Although numerical simulations of nonstationary processes provide the resulting interaction patterns for specific parameter sets, they offer limited insight into the role of individual parameters. In contrast, analytical approaches enable a systematic evaluation of each contributing factor, allowing the identification of dominant mechanisms in nonlinear optical processes and facilitating controlled manipulation of the system dynamics.

In the present work, an analytical analysis of the simultaneous influence of quadratic and cubic nonlinearities on the spectrum of second harmonic generation of an ultrashort pulse in a metamaterial is carried out.

We investigate SHG in a metamaterial exhibiting both quadratic and cubic nonlinearities under the oo-e scalar synchronism condition. It is assumed that the medium behaves as a left-handed material only at the pump-wave frequency $\omega_1$, that is, it possesses simultaneously negative values of both the dielectric permittivity and magnetic permeability at the pump frequency $\omega_1$ ($\varepsilon_1 < 0,\ \mu_1 < 0$), while at the harmonic frequency ($\varepsilon_2 > 0,\ \mu_2 > 0$) these parameters are positive. Consequently, the energy flux of the pump wave and that of the generated harmonic are directed



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oppositely, resulting in a counter-propagating interaction of the waves [12-13].

Ultrashort laser pulses find widespread applications in medicine [4], biology, terahertz radiation generation [14], nonlinear atmospheric probing [15], and numerous other areas. A key characteristic of ultrashort pulses, particularly femtosecond pulses, is their ability to concentrate extremely high energy within very narrow spectral, temporal, and spatial domains, with record intensities at the laser focus reaching approximately $10^{21}$–$10^{22}$ W/cm². As is well known, for ultrashort pulses, the nature of wave interactions is strongly influenced by the dispersive properties of the medium. In a medium with a positive refractive index, during the interaction of ordinary and extraordinary waves, the wave with a higher group velocity propagates ahead of the slower wave. Consequently, after a certain propagation distance, the wave packets become spatially separated, and their interaction effectively ceases. In contrast, in left-handed (negative-index) media, the group velocities of the interacting waves are directed toward each other. Under such conditions, wave packets can fully overlap at specific regions within the material, leading to enhanced nonlinear interactions. Subsequently, the waves separate and propagate independently, terminating their interaction. The location and extent of this overlap depend on the ratio of the group velocities. Additionally, the presence of different phase velocities for the frequency components of the pulse in a dispersive medium must be considered, as this leads to dispersive pulse broadening. Therefore, accounting for group velocity mismatch in counter-propagating waves is essential when studying nonlinear parametric interactions of ultrashort pulses in metamaterials.

The investigation of nonlinear effects in artificial media and the engineering of their nonlinear response are of critical importance for advancing the field of metamaterials [16]. Research in this area is proceeding along multiple directions. For example, composite metamaterials formed from a mixture of two isotropic dielectric materials have been studied in [17]. Ultrathin active chiral metamaterials with highly tunable chiroptical responses, designed for applications such as ultrasensitive sensors, have been investigated in [18]. Structural metamaterials employed for spectral deformation analysis have been explored in [19]. To study nonlinear optical processes, reduced forms of the governing equations are often used, which are solved using approximate methods such as the constant-field approximation and the Tagiev-Chirkin approximation [20–21].

## THEORY

The study is carried out within the first-order dispersion approximation, i.e., without accounting for the effect of pulse broadening due to dispersion during wave propagation in the metamaterial and neglecting losses. For the considered wave-propagation geometry, the well-known reduced equations describing SHG in a medium with quadratic and cubic nonlinearities [21] and exhibiting negative refraction at the pump frequency are transformed into the form [22]:

$$\frac{\partial A_1}{\partial z} - \frac{1}{|u_1|}\frac{\partial A_1}{\partial t} = i|\gamma_1|A_1^* A_2 exp(z) + i|\gamma_{11}|I_{1l}A_1$$

$$\frac{\partial A_2}{\partial z} + \frac{1}{u_2}\frac{\partial A_{12}}{\partial t} = -i\gamma_2 A_1^2 exp(i\Delta z) + i\gamma_{21}I_{1l}A_2 \qquad (1)$$

Here $A_{1,2}$ denote the complex amplitudes of the pump and SH waves at the frequencies $\omega_1$ and $\omega_2$ ($\omega_2 = 2\omega_1$), respectively. The parameters $I_{1l} = A_{1l} \cdot A_{1l}^*$, $\gamma_{1,2}$ are the coefficients of the quadratic nonlinear coupling between the waves in the metamaterial at the respective frequencies $\omega_{1,2}$ , i.e. The coefficients $\gamma_j \sim \chi^{(2)}$ and $\gamma_1 = -|\gamma_1|$, $\gamma_{11}$ , $\gamma_{21}$ are related to the cubic nonlinearity of the medium, i.e., $\gamma_{nj} \sim \chi^{(3)}$ and $\gamma_{11} = -|\gamma_{11}|$. The term with $\gamma_{11}$ corresponds to the self-action of the waves, while that with $\gamma_{21}$ describes cross-interaction effects. $u_{1,2}$ denote the group velocities of the respective waves $u_1 = -|u_1|$ , $\Delta = k_2 - 2k_1$ represents the linear phase mismatch relative to the central frequency of the pump wave.

The analysis of the nonstationary regime of harmonic generation assumes that the wave amplitudes are temporally modulated. The spectrum of the SH in the nonstationary generation regime, taking into account cubic nonlinearity, is studied in the constant approximation for $\gamma_1 = 0$ . In the analysis of nonstationary SHG, a spectral approach is typically employed: instead of solving the differential equation for the field amplitudes, the corresponding equation for the spectral amplitudes is solved, and the field amplitude of the SH is then obtained from the spectral amplitude. This frequency-domain approach makes it possible to reveal the underlying physical mechanisms of the conversion process more clearly.

An optical pulse can be represented as a superposition of monochromatic waves with frequencies $\omega$ and wave vectors $k(\omega')$ . The frequencies $\omega'$ occupy a certain interval $\Delta\omega'$ centered around the carrier frequency $\omega$ . All these monochromatic components propagate in a dispersive medium, and therefore their phase velocities $\frac{c}{n(\omega')}$ differ. As a result, the optical pulse becomes distorted during propagation (the effect of dispersion broadening).

Ultrashort pulses are characterized by a relatively wide frequency spectrum. The spectral width $\Delta\omega$ is related to the pulse duration $\tau$ through the well-known time–bandwidth relation

$$\Delta\omega \geq 1/\tau$$

Using the spectral approach, the amplitude of the second-harmonic field $A_2(z,\mu)$ can be represented in the form of a Fourier integral:

$$A_2(z,\mu) = \int_{-\infty}^{\infty} \Phi_2(z,\Omega)e^{i\Omega\mu}d\Omega, \Omega = \omega - 2\omega_1. \quad (2)$$

By analogy for $A_1(\mu)$

$$A_1(\mu) = \int_{-\infty}^{\infty} \Phi_1(\xi)e^{i\xi\mu}d\xi\,. \quad (3)$$

Introducing the auxiliary function

$$\Lambda_2(\mathrm{z},\Omega) = \Phi_2(z,\Omega)e^{-i(\nu\Omega-\gamma_{21}I_{1l})z+i(\nu'\Omega+\gamma_{21}I_{1l})z} \quad (4)$$

and the generalized phase mismatch

$$\varphi = \Delta - \nu\Omega + \gamma_{21}I_{1l},\ \varphi' = \Delta + \nu'\Omega + \gamma_{21}I_{1l},$$

the equation (3) is transformed into the form

$$\frac{\partial\Lambda_2}{\partial z} = \left[\frac{d\Phi}{dz} - \mathrm{i}(\nu\Omega + i\gamma_{21}I_{1l})\Phi_2\right]e^{-i(\nu\Omega-\gamma_{21}I_{1l})z-i(\nu'\Omega+\gamma_{21}I_{1l})z} \quad (5)$$

$$\frac{\partial\Lambda_2}{\partial z} = -i\gamma_2 e^{i(\Delta z+\nu'\Omega+\gamma_{21}I_{1l})z} \quad (6)$$

By substituting expression (6) into (3), we obtain

$$\int_{-\infty}^{\infty}\left(\frac{d\Phi}{dz} + \nu' + i\Omega + \mathrm{i}\omega - \mathrm{i}\nu\Omega\Phi_2\right)e^{i\Omega\mu}d\Omega = -i\gamma_2 e^{i\Delta z}\int\int_{-\infty}^{\infty}\Phi_1(\xi)\Phi_1(\xi')e^{i(\xi+\xi')\mu}d\xi\,d\xi' -$$

$$i\gamma_{21}I_{1l}A_2\int_{-\infty}^{\infty}\Phi_2 e^{i\Omega\mu}d\Omega. \quad (7)$$

Then,

$$\oint_0^z lnA_1(z) = i|\gamma_{11}|I_{1l}\int_0^z dz. \quad (8)$$

Let us assume that,

$$A_1(z=0) = A_{1l},\ A_1(z) = A_{1l}e^{-i|\gamma_{11}|I_{1l}z} \quad (9)$$

Taking into account the boundary conditions (9) in equation (6), integration over $z$ yields the spectral amplitude for the second harmonic generation process:

$$\frac{\partial\Lambda_2}{\partial z} = -i\gamma_2 e^{i[\Delta+\nu'\Omega+\gamma_{21}I_{1l}]z-2i|\gamma_{11}|I_{1l}z}\int\Phi_1 l(\xi)\xi\int\Phi_1 l(\xi)\xi \quad (10)$$

Here

$$\varphi' = \Delta + \nu'\Omega + (\gamma_{21} - 2|\gamma_{11}|)I_{1l}. \quad (11)$$

And

$$\Delta^{NL} = (2|\gamma_{11}| + \gamma_{21})I_{1l} \quad (12)$$

$$\int_{-\infty}^{\infty} e^{-\frac{\tau_1^2}{4}\Omega^2}\cdot e^{-\frac{\tau_1^2}{2}\xi^2+\frac{\tau_1^2}{4}2\Omega\xi}d\xi = \frac{\sqrt{2\pi}}{\tau_1}e^{-\frac{\tau_1^2}{4}\Omega^2}\cdot e^{\frac{\Omega^2}{8}\tau_1^2} = \frac{\sqrt{2\pi}}{\tau_1}e^{-\frac{\Omega^2\tau_1^2}{8}} \quad (13)$$

Let us substitute equation (13) into equation (10):

$$\frac{\partial\Lambda_2}{\partial z} = -i\gamma_2 e^{i\varphi z}\left(\frac{\tau_1 A_0}{2\sqrt{\Omega}}\right)^2\frac{\sqrt{2\pi}}{\tau_1}e^{-\frac{\Omega^2\tau_1^2}{8}} == -i\gamma_2 e^{i\varphi z}\frac{\tau_1 A_0^2}{\sqrt{8\Omega}}e^{-\frac{\Omega^2\tau_1^2}{8}} \quad (14)$$

If we integrate with respect to $z$ from 0 to $l$, it follows that

$$\Lambda_2(z,\Omega) = -i\frac{\gamma_2\tau_1 A_0^2}{\sqrt{8\Omega}} e^{-\frac{\Omega^2\tau_1^2}{8}} \int_0^z e^{i\varphi z} dz = -\frac{\gamma_2\tau_1 A_0^2}{\sqrt{8\Omega}} e^{-\frac{\Omega^2\tau_1^2}{8}} \cdot \frac{e^{i\varphi l}-1}{\varphi}. \quad (15)$$

From expression (15), we obtain the amplitude spectrum of the second-harmonic pulse:

$$\Phi_2(z,\Omega) = \Lambda_2 e^{-i(\nu'\Omega+\gamma_{21}I_{1l})z} = -i\frac{\gamma_2\tau_1 A_0^2}{\sqrt{8\Omega}} z \cdot e^{-\frac{\Omega^2\tau_1^2}{8}+i(\nu'\Omega+\gamma_{21}I_{1l})z} \quad (16)$$

Similarly, the output second-harmonic spectrum of the crystal can be represented in the form of

$$|\Phi_2(z,\Omega)|^2 = \frac{(\gamma_2 A_{1l}^2\,\tau_1 z)^2}{8\pi} e^{-\frac{\Omega^2\tau_1^2}{4}} \mathrm{sinc}^2\frac{(\nu\Omega+\Delta+\Delta^{NL})}{2} z, \quad (17)$$

Here $\Phi_2(z,\Omega)$ represents the amplitude

$$A_1(z=l,t) = A_{1l}\, exp(-\frac{t^2}{\tau_1^2})$$

of the second-harmonic spectrum, $\tau_1$ is the pulse duration of the Gaussian pump radiation at the input of the medium, and the relation between the group velocities in the medium with negative refraction at the frequency

$$\nu = u_2^{-1} + |u_1|^{-1} - \omega_1$$

is taken into account. $\Omega = \omega - \omega_2$ denotes the central frequency of the fundamental radiation.

Thus, the analysis of the second harmonic generation process via frequency spectra provides a clear and precise understanding of the underlying interaction dynamics.

## CONCLUSION

In the study of non-stationary second-harmonic generation, a conventional spectral methodology was employed. The differential equations governing the field amplitudes were substituted with their counterparts expressed in terms of spectral amplitudes. Examining the second-harmonic conversion process through the lens of frequency spectra allows for a more transparent and precise understanding of the underlying physical mechanisms.


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