On theory of approximation methods in the study of second harmonic generation of ultrashort laser pulses in periodic crystals

Cite as: AIP Conference Proceedings **2432**, 020008 (2022); https://doi.org/10.1063/5.0089970
Published Online: 16 June 2022

Z. J. Ruziev, B. Sapaev, I. B. Sapaev, et al.

On Theory of Approximation Methods in the Study of Second Harmonic Generation of Ultrashort Laser Pulses in Periodic Crystals

Z.J. Ruziev[1], B. Sapaev[2], I.B. Sapaev[3], U.K. Sapaev[1, a)], R. Amanov[2], D. Nazaraliev[3]

[1]*Tashkent State Technical University, 2, University str., Olmazor district, 100095, Tashkent, Uzbekistan*
[2]*Tashkent State Agrarian University, 2, University str., 100140, Tashkent, Uzbekistan*
[3]*Tashkent Institute of Irrigation and Agricultural Mechanization Engineers, 39, Kari Niyaziy str., 100000, Uzbekistan*

[a)]*Corresponding author: usapaev@gmail.com*

Abstract. Frequency doubling of ultrashort laser pulses in nonlinear photonic (periodic) crystals is investigated by numerical methods by using the approximation of the slowly varying amplitudes and a unidirectional approximation, applicable to simplify the wave equation with nonlinear polarization in a dispersive medium. Under the same experimental conditions, results of these approximations are compared. Comparative analysis shows that, up to 10 fs of the main pulse duration, both approximate methods describe this frequency conversion in almost the same way, but below 10 fs, there is a discrepancy between their results. Mainly, the formation of the temporal profile of the second harmonic pulse and its efficiency are compared. A technique for obtaining time profiles of the second harmonic pulse, when one uses a unidirectional approximation is also shown.

Keywords: Maxwell equations, nonlinear optics, slowly varying amplitude approximation, unidirectional approximation, unidirectional pulse propagation equation, second harmonic generation, lithium niobite, femtosecond pulses.

INTRODUCTION

As known, that in laser technology, researchers always strive to reduce the duration of laser pulses and accordingly their power. If at the initial stage of the development of lasers, coherent pulses from them had only nano-picosecond ranges, then, at present, thanks to the improvement of the laser system itself and the amplification and generation system of the so-called of "chirped" laser pulses, obtaining high-power femtosecond (fs) pulses is practically not difficult [1]. It is known that short and ultrashort laser pulses are widely used to solve various fundamental and applied problems of the interaction of laser radiation with matter. For example, fs pulses are widely used in studies of various fast phenomena, in studies of the fundamental properties of substances at ultrahigh temperatures, as well as in various fields of medicine and diagnostics etc. [2,3].

Interest in various frequency conversion processes, including second harmonic generation (SHG), is due, in particular, to the production of coherent radiation in the short-wavelength range of the spectrum, where direct laser radiation can be ineffective or completely absent. To solve this problem, nonlinear optical crystals with quadratic nonlinearity are successfully accepted. Of these, the most common are crystals with a regular domain structure or their other terminology is nonlinear photonic crystals (see for example in the last review [4]). Since they have a number of advantages over other nonlinear optical crystals; they do not require the fulfillment of the phase matching condition, and it is realized by changing the sign of the second-order nonlinear susceptibility from layer to layer, while the size of the domain must be equal to the coherent interaction length [5, 6]. Moreover, they can be

The 1st International Conference on Problems and Perspectives of Modern Science
AIP Conf. Proc. 2432, 020008-1–020008-6; https://doi.org/10.1063/5.0089970
Published by AIP Publishing. 978-0-7354-4345-7/$30.00

oriented towards the incident radiation in such a way that the largest value of the second-order nonlinearity contributes to the frequency conversion process, ensuring efficient energy exchange between optical waves. The efficient implementation of SHG also requires high-intensity laser sources, which can usually be achieved using fs laser pulses.

The use of short and ultrashort laser pulses in frequency conversion processes in nonlinear photonic crystals causes a number of difficulties that severely limit their efficiency. Such effects during SHG include, for example, the detuning of the group velocities of the fundamental radiation and the second harmonic, the dispersion of the group velocities, as well as the third higher orders of dispersion [7]. These unwanted effects also lead to a rapid increase in the second harmonic pulse width. In the above case, for the theoretical analysis of SHG, the so-called truncated Maxwell equations in the slowly varying amplitude (SVA) approximation, taking into account the nonlinear polarization of the medium, are usually taken [7-9]. The applicability of this approximation is limited only by the duration of the incident pulse (usually up to approximately~10 fs). It is interesting to note that relatively recently, another method has been developed, the so-called unidirectional pulse propagation equation (UPPE) in a dispersive medium [10-12]. As the authors of these works show, this method is premium for analyzing the propagation and interaction of optical waves in dispersive media up to one light period. That is, it follows that this approximation is not limited by the duration of the incident pulses. At the same time, the SVA approximation is applicable up to approximately~10 fs duration of the main pulse [7].

Thus, this work was given the task of comparing the results of SVA and UPPE under the same initial conditions in order to determine the limit of applicability of these approximations when using ultrashort laser pulses. For this, the process of SHG of short and ultrashort laser pulses in a periodically polarized crystal of lithium niobate ($LiNBO_3$) was chosen. Since this crystal is widely used as a nonlinear photonic crystal, and its linear and nonlinear optical characteristics are well studied. Below we will analyze the results of these approximations using the example of SHG in a nonstationary regime with an interaction of the ee-e type [8].

THEORETICAL PART

The SHG process taking into account dispersion up to the third order in a moving coordinate system in the SVA approximation has the following system of partial differential equations [7, 8]:

$$\begin{aligned} &\frac{\partial A_1}{\partial z} - i\frac{g_1}{2}\frac{\partial^2 A_1}{\partial t^2} + \frac{h_1}{6}\frac{\partial^3 A_1}{\partial t^3} = -i\gamma_1\sigma(z)A_1^* A_2 e^{i\Delta kz} \\ &\frac{\partial A_2}{\partial z} + v\frac{\partial A_2}{\partial t} - i\frac{g_2}{2}\frac{\partial^2 A_2}{\partial t^2} + \frac{h_2}{6}\frac{\partial^3 A_2}{\partial t^3} = -i\gamma_2\sigma(z)A_1^2 e^{-i\Delta kz} \end{aligned} \qquad \textbf{(1.1)}$$

here, A_1 and A_2 are the complex amplitudes of the fundamental and second harmonics, respectively $v = 1/V_1 - 1/V_2$, where V_1 and V_2 are the group velocities of the fundamental and second harmonics respectively; $g_1 = \left.\frac{\partial^2 k}{\partial \omega^2}\right|_{\omega=\omega_0}$, $g_2 = \left.\frac{\partial^2 k}{\partial \omega^2}\right|_{\omega=2\omega_0}$, $h_1 = \frac{1}{2k_0}\left.\frac{\partial^3 k}{\partial \omega^3}\right|_{\omega=\omega_0}$ and $h_2 = \frac{1}{2k_0}\left.\frac{\partial^3 k}{\partial \omega^3}\right|_{\omega=2\omega_0}$ where g_i and h_i - coefficients taking into account the dispersion of group velocities of the second and third orders, respectively (i = 1, 2); $k_0 = \frac{2\pi}{\lambda_0}$, $\omega_0 = \frac{2\pi c}{\lambda_0}$, λ_0 - incident wavelength in the center of its spectrum and c is the speed of light in vacuum; $\Delta k = 2k(\omega) - k(2\omega)$ - wave detuning; γ_i - coefficients of nonlinear connection, which are equal to $\gamma_1 = \gamma_2 = \pi d_{eff}/n\lambda_0$ where $d_{eff} = \chi^{(2)}/2$ effective second-order nonlinear susceptibility coefficient ($\chi^{(2)}$ - nonlinear susceptibility of the second order) [Boyd, 2019; Sidick et al., 1996]; $\sigma(z)$ - sign-changing function with period $d_0 = \pi/|\Delta k|$, that is, the thickness of one domain periodic crystal.

To numerically solve the system of the equations (1.1), we used the Runge – Kutta method for the nonlinear part, and for the linear one, the fast Fourier transform using a symmetric scheme on dz [13, 14].

Let us now consider the technique of UPPE taking into account the second-order nonlinear polarization [10-12]. In this case, the SHG equation in the spectral region and in scalar form can be written as follows:

$$\frac{\partial E(\omega,z)}{\partial z} = ik(\omega)E(\omega,z) + \frac{i\omega^2}{2\in_0 c^2 k(\omega)} P_{NL}(\omega,z) \quad \textbf{(1.2)}$$

where, $P_{NL}(t,z) = \chi^{(2)}E^2(t,z)$, $\in_0$ - electrical constant. If we take into account the field as, then we can obtain the following equation for its amplitude: $E(\omega,z) = A(\omega,z)\exp(ik(\omega)z)$

$$\frac{\partial A(\omega,z)}{\partial z} = i\frac{\mu_0\omega^2}{2k(\omega)} e^{-ik(\omega)z} P_{NL}(\omega,z) \quad (1.3)$$

equation (1.2) can be solved using the following algorithm (see, also in [11]):

- Move the field amplitude from $z=0$ to Δz then $A(\omega,\Delta z) \to A(\omega)\exp[i(k(\omega)-\omega_0/V_\omega)\Delta z]$
- Using the inverse Fourier transform, we transform it from the spectral domain to the time domain; $A(t) = FFT\{A(\omega)\}$;
- Having the field amplitude in the time domain, we calculate the nonlinear polarization as $P_{NL}(t) = \chi^{(2)}E^2(t)$;
- After calculation, using the inverse Fourier transform, transform it back to the spectral region $P_{NL}(\omega) = FFT^{-1}\{P(t)\}$;
- We apply a linear propagator to cancelprevious shift by $P_{NL}(\omega) \to P_{NL}(\omega)\exp\left[-i\left(k(\omega)-\frac{\omega_0}{V_\omega}\right)\Delta z\right]$

and finally we solve (1.3) ordinary differential equation using the Runge Kutta method of its fourth order.

The boundary conditions of equations (1.1) and (1.3) are chosen as follows: $A_1(z=0,t) = A_0\exp\left(-2\ln 2\left(\frac{t}{\tau}\right)^2\right)$, $A_2(z=0,t)=0$, $E(z=0,t) = \frac{A_0}{\sqrt{\pi}}\exp\left(-2\ln 2\left(\frac{t}{\tau}\right)^2\right)\cos(\omega_0 t)$ respectively. Note that, choosing the boundary conditions in this way, the impulses fall on crystal with the same energy and duration.

RESULTS AND DISCUSSION

As noted above, lithium niobate was chosen as a crystal with a regular domain structure. Its transparency area from 400 nm to 5500 nm and $\chi^{(2)}$= 41 pm^2 /V^2 [15]. Therefore, with increasing (1.2), only this region of the spectrum was taken into account, the rest and the negative region of the spectrum were set to zero. As a coherent source of the main radiation, we chose radiation from a Ti: Sapphire laser, whose central wavelength is λ_0 = 1050 nm. As far as this laser can generate ultrashort laser pulses with high intensity in the femtosecond range [16]. For radiation with a wavelength λ_0 = 1050 nm, the period of the nonlinear grating will be approximately equal to $d_0 \approx 3.23$ μm. The pulse width is selected at FWHM. The Sellmeier equation for calculating the refractive index versus the wavelength of lithium niobate was chosen from [17]. For all calculations, we chose the peak intensity of the main pulse of 0.5 GW / cm^2. (The value of the latter was chosen in order to avoid the influence of third-order nonlinearity [18]).

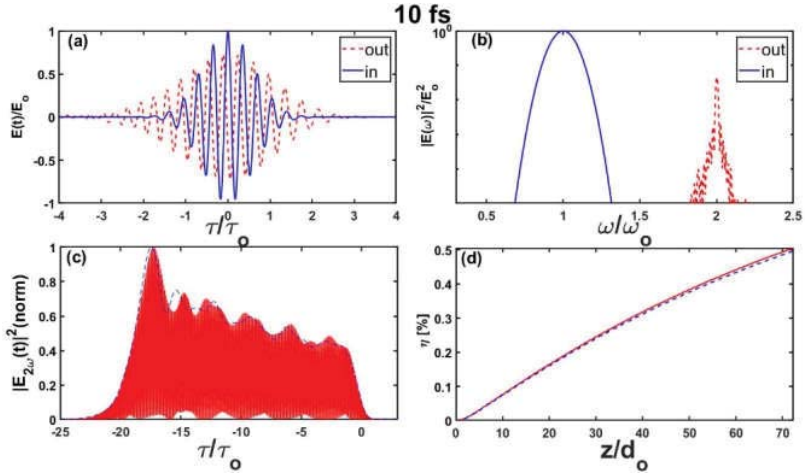

FIGURE 1. Results of numerical calculations for 10 fs: (a) - time profile of the field itself at the input (solid curve) and at the output (dashed curve) from the crystal; (b) - its spectrum at the input (solid line) and at the output (dashed line); (c) Comparative analysis results showing the temporal profile of the second harmonic intensity at the exit from the crystal. Here the dashed line is the results of the MMA approximation, and the solid line is the unidirectional approximation; (d) - dependence of the SHG efficiency on the number of domains. Here, the solid curve shows the result of the MMA approximation, the dashed line shows the unidirectional approximation of the wave equation.

Before presenting the results of a comparative analysis of the approximate methods, we would like to note that both approximate methods gave practically the same results up to 10 fs, as we expected. For comparison, we chose the SHG efficiency and the time profile of the second harmonic intensity at the exit from the periodic crystal. When calculating UPPE, the time profile of the second harmonic amplitude was obtained from the spectrum of the field itself from 1.5 to 2.5 normalized frequencies using the inverse Fourier transform. Figure 1 shows the results of numerical calculations for the case of 10 fs, where the dynamics of the second harmonic was studied during the passage of a photonic crystal with approximately 70 domains: (a) the time profile of the field itself at the input (solid curve) and at the output (dashed curve) from the crystal ; (b) - its spectrum at the input (solid line) and at the output (dashed line); (c) Comparative analysis results showing the temporal profile of the second harmonic intensity at the exit from the crystal. Here the dotted line is the results of the SVA approximation, and the solid line is the unidirectional approximation; (d) - dependence of the SHG efficiency on the number of domains. Here the solid curve shows the result of the SVA approximation, the dashed line shows the UPPE.

In this figure, almost both approximate methods give the same results. But only a slight difference is observed when comparing the time profile of the second harmonic intensity (see part (c)). Although here in two different calculations a small deviation of the second harmonic profiles from each other is observed, when comparing the SHG efficiency, they coincide with each other (see part (d)).

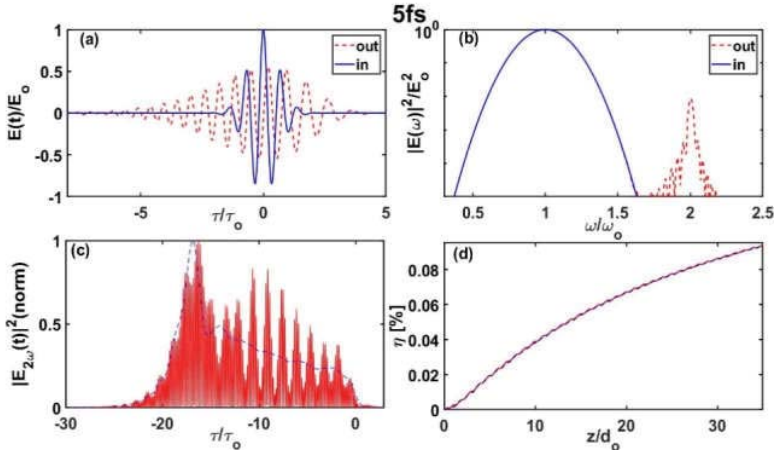

FIGURE 2. Results of approximate methods for the case of 5 fs. The order of figures and curves are defined as in Figure 1

The comparative results of the two approximate methods for the 5 fs case are shown in Figure 2. Here the curves and the order of the figures are defined as in Figure 1. In this figure, there is a noticeable deviation in the results when comparing the time profile of the second harmonic intensity, which is shown in part (c). In calculations using the unidirectional approximation, the second harmonic pulse is divided into several parts, but according to the SVA approximation, it is not. Despite this, the results on the second harmonic efficiency in both calculations coincide. Comparing the results for the 5 fs and 10 fs cases, we observe that the results for the two approximations differ insignificantly from each other, if the dispersion up to the third order of smallness is taken into account in the SVA approximation. From this it follows that both approximations can be used in calculations. However, it should be noted that here we used a fixed value $\chi^{(2)}$ of the quadratic nonlinearity of polarization. However, this parameter can be overestimated by the frequency of the incident wave radiation. In this case, if the incident pulse on the crystal has a sufficiently wide spectrum, then it is necessary to take into account the dependence of $\chi^{(2)}$ on ω as $\chi^{(2)}(\omega)$. It is in this case that one can successfully apply a unidirectional approximation rather than the SVA approximation.

CONCLUSION

In conclusion, SHG of ultrashort laser pulses (up to 5 fs) in a crystal with a regular domain structure has been studied numerically. A lithium niobate crystal was chosen as a periodically polarized crystal. For numerical calculations, the SVA approximation and the unidirectional wave equation approximation were used. Taking into account the smallness of the dispersion to the third order in the SVA approximation, the results of these approximations are compared. The results of calculations show that both approximate methods give the same results up to 10 fs of the duration of the fundamental pulse when comparing the temporal profile of the pulse and the efficiency of the second harmonic. But below 10 fs, there is a discrepancy in the results.

A technique was shown for obtaining the temporal profile of the second harmonic pulse from the main field of study using a unidirectional approximation of the wave equation. Here we limited ourselves to the second harmonic only, but the technique can be applied to other higher order harmonics.

This work was partially supported by the grant UZB-Ind-2021-96 and UZB-Ind-2021-83 of the Ministry of Innovative Development of the Republic of Uzbekistan

REFERENCES

1. Morgner, U., Kärtner, FX, Cho, SH, Chen, Y., Haus, HA, Fujimoto, JG, Tschudi, T. Sub-two-cycle pulses from a Kerr-lens mode-locked Ti: *sapphire la-ser ... Optics letters,* 1999, 24, no. 6, pp. 411-413.
2. Daido, H., Nishiuchi, M., & Pirozhkov, AS Review of laser-driven ion sources and their applications. *Reports on progress in physics,* 2012, 75, no. 5, pp. 056401.

3. Rizvi, NH Femtosecond laser micromachining: Current status and applications. *Riken review,* 2003, pp. 107-112.
4. Jankowski, M., Langrock, C., Desiatov, B., Marandi, A., Wang, C., Zhang, M., Fejer, MM Ultrabroadband nonlinear optics in nanophotonic periodically poled lithium niobate waveguides. *Optica,* 2020, 7, No. 1, pp. 40-46.
5. Tanzilli, S., De Riedmatten, H., Tittel, W., Zbinden, H., Baldi, P., De Micheli, M., Gisin, N. Highly efficient photon-pair source using periodically poled lithium niobate waveguide. *Electronics Letters,* 2001, 37, No. 1, pp. 26-28.
6. Miller, GD, Batchko, RG, Tulloch, WM, Weise, DR, Fejer, MM, & Byer, RL 42% -efficient single-pass cw second-harmonic generation in periodically poled lithium niobate. *Optics letters,* 1997, 22, no. 24, pp. 1834-1836.
7. Akhmanov SA, Vysloukh, VA, Chirkin, AS Optics of femtosecond laser pulses. Moscow, *Nauka,* 1988, pp. 308.
8. Dmitriev, VG, Tarasov, LV Applied nonlinear optics. Moscow, Fizmatlit 2004, 512c.
9. Boyd, Robert W. Nonlinear optics. *Academic press,* 2019, pp. 613.
10. Husakou AV, Herrmann J. Supercontinuum generation of higher-order soli-tons by fission in photonic crystal fibers, *Phys. Rev. Lett.* 2001. V. 87. No. 20.P. 203901.
11. Couairon, A., Brambilla, E., Corti, T., Majus, D., Ramírez-Góngora, ODJ, Kolesik, M. Practitioner's guide to laser pulse propagation models and simulation, The European Phys. J. *Special Topics.* 2011. V. 199. No. 1.pp. 5-76.
12. Mlejnek, M., Wright, EM, Moloney, JV Femtosecond pulse propagation in argon: A pressure dependence study, *Phys. Rev. E.* 1998. V. 58. no. 4.pp. 4903.
13. Olimov, AN, Ruziev, ZJ, Yusupov, DB, Sapaev, UK Frequency doubling of femtosecond laser pulses in nonlinear photonic crystals with account of high-order dispersion, Journal of *Russian Laser Research.* 2019. V. 40. No. 3.pp. 280.
14. Agrawal, GP, Mamyshev, PV, Chernikov, SV Nonlinear fiber optics. Mosvka, *Mir,* 1996, pp. 323.
15. Nikogosyan, DN Nonlinear optical crystals: a complete survey. *Springer Sci-ence & Business Media*, 2006, 350p.
16. https://www.rp-photonics.com/titanium_sapphire_lasers.html
17. Dalgarno, A., Kingston, AE The refractive indices and Verdet constants of the inert gases. Proceedings of the Royal Society of London. Series A. *Mathematical and Physical Sciences*, 1960, 259, 1298, pp. 424-431.
18. Kulagin, IA, Sapaev, UK, Usmanov, T., Uzakov A., A., Yusupov DB Non-stationary frequency doubling in periodically poled nonlinear crystals in the presence of self-action effects. J. *Russ. Laser. Res*. 2007, v. 28, pp. 279-287.