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Quasi-Phase-Matched Backward wave and THG in PPNC via the Constant Intensity Approximation Method

Nazaket V. Kerimli*

Department of Medical and Biological Physics, Faculty of General Medicine,
Azerbaijan Medical University, Baku, Azerbaijan

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Abstract

In the framework of the constant intensity approximation, we analyze a nonlinear quasi-phase-matched process of frequency tripling, where the second harmonic wave propagates in the backward direction. The efficiency of third harmonic generation and second harmonic backward wave are calculated analytically for various orders of quasi-phase matching. Studies of quasi-phase-matched harmonic generation in PPLN crystals, conducted under the constant intensity approximation, have shown that increasing the order of quasi-phase matching leads to an increase in the optimal domain thickness. According to the analysis based on the constant intensity approximation, it has been found that the accuracy in determining the quasi-phase-matching periods of an optical superlattice depends on several factors: the precision of the refractive indices at the frequencies of the interacting waves for the specific sample, the second-order nonlinear susceptibility of the domains, and the intensities of the interacting waves. It is shown that the initial phases of the interacting waves have a significant impact on the efficiency of frequency conversion. Optimal phase values are identified, demonstrating that proper phase selection can substantially enhance the conversion efficiency. Based on this analysis, design recommendations are provided for optimizing the structure of the optical superlattice to achieve maximum efficiency. These results are of practical interest for the development of devices based on third harmonic generation when backward second harmonic is simultaneously generated.

Keywords: constant intensity approximation, quasi-phase-matching, periodically poled crystal (PPLN)
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*Corresponding author. Tel.: (+994) 70 670 96 16.
e-mail: nazaketkerimli3@gmail.com; ORCID ID: 0000-0002-9768-8248.

1. Introduction

With the global population both aging and expanding, conventional centralized pathology laboratories are becoming less affordable and increasingly unsustainable. By 2035, developments in integrated photonics are expected to make it possible to perform point-of-care testing directly in general practitioners' offices or at patients' bedsides using compact chips. Presently, the point-of-care testing market is estimated to reach a value of $31 billion by 2025, which could reduce the reliance on traditional pathology labs by up to 25%.

The application of lasers in medicine holds vast potential for the future. For instance, techniques like spectroscopy for blood glucose monitoring and precisely targeted irradiation for light-activated cancer therapies performed through minimally invasive (keyhole) surgery are just a few examples. These, along with many other diagnostic and therapeutic innovations, are either already in use or currently undergoing active clinical trials.

Non-linear optical (NLO) crystals offer a highly versatile method for producing new wavelengths from standard, readily available laser sources. While a broad range of commercial lasers spans much of the optical spectrum, direct or cost-effective light sources are not always accessible for every application. In such cases, where a suitable direct source is lacking, wavelength conversion through efficient NLO crystals becomes a valuable and effective alternative.

When it comes to non-linear optical crystal materials, lithium niobate ($LiNbO_3$) stands out due to its exceptionally high non-linear coefficient. In particular, magnesium oxide-doped, periodically poled lithium niobate (MgO:PPLN) offers a highly efficient and versatile solution. Its ability to be periodically poled, combined with broad optical transparency, makes it ideal for generating wavelengths across a wide spectral range—from 400 nm to 5 µm.

Visible wavelengths within the optical spectrum can be produced through processes such as Second Harmonic Generation (SHG) or Sum Frequency Generation (SFG). By selecting suitable pump lasers—whether with fixed or tunable wavelengths—it is possible to generate a desired output across the visible range. High-efficiency Second Harmonic Generation (SHG) of 1064 nm light using periodically poled lithium niobate (PPLN) can produce 532 nm green light at power levels in the watt range. This output is well-suited for dermatological applications, including the treatment of port-wine stains, birthmarks, melanomas, as well as for tattoo and hair removal.

A pulsed laser in the near-infrared range (700–900 nm) can be used as a surrogate radiation source for diode-pumped alkali lasers (DPAL). Currently, such sources include alexandrite lasers, titanium-sapphire lasers, and dye lasers [1]. Nonlinear optics offers an alternative approach to generating the desired wavelength. The use of quasi-phase matching in crystals with periodically polarized structures is one of the most effective methods for creating optical parametric generators (OPGs). Periodically poled lithium niobate doped with MgO (MgO:PPLN), which has a high second-order nonlinear coefficient (d33d_{33}d33), is a typical example of a crystal that provides quasi-phase matching [1]. t has a broad transparency range (from 0.35 to 5 μm) and a high degree of homogeneity. An OPG based on MgO:PPLN, pumped by a neodymium laser (1064 nm), can efficiently generate mid-infrared radiation with wavelengths ranging from 2.4 to 4.2 μm [5, 6]. By sum-frequency mixing of the idler and the pump laser radiation, a tunable wavelength in the range of 737 to 845 nm can be obtained.

Recently, periodically poled crystals—also known as optical superlattices—have gained widespread use as efficient frequency converters through the quasi-phase-matching (QPM) technique. As depicted on the picture 1. reversing the direction of the z-axis or the domain alignment alters the sign of the nonlinear coupling, thereby reversing the direction of energy flow. Flipping the sign of the nonlinear susceptibility at each coherence length causes a π phase shift in the polarization wave, effectively resetting the phase mismatch and enabling a continuous transfer of power into the harmonic wave.

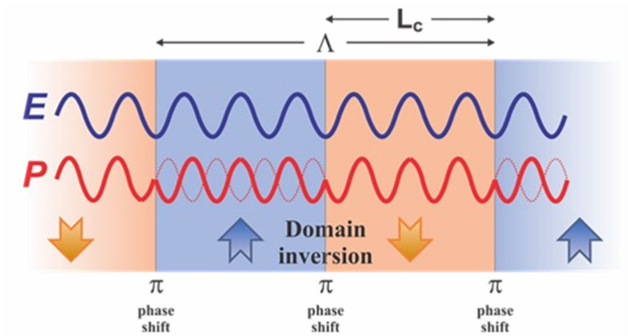

Fig 1. Quasi-phase matching (QPM)

The range of practical applications significantly expands with the implementation of sequential quasi-phase-matched interactions using a common pump wave. In media with quadratic nonlinearity, the number of possible parametric processes increases when a common pump wave is used. Studies conducted by the authors of works [2–6, 9–12] have shown that, through sequential interactions, by selecting the modulation period of the nonlinear susceptibility and the quasi-phase-matching orders for different harmonics, it is possible to achieve their simultaneous generation. This means the possibility of obtaining, in practice, a source of coherent radiation using a single laser and a periodically polarized crystal, which simultaneously generates at several optical harmonics. In the approximation of a given intensity [10], we conducted an analysis of the interaction of co-propagating waves in an PPLN crystal, which leads to the simultaneous generation of the second and third harmonics from a common pump wave. The present work continues this research, focusing on the case of sequential interaction in a counter-propagating wave geometry, which holds great potential for applications.

2. Theory

Behavior of interacted waves are presented by well-known shortened equations for negligible small wave loss [10]:

$$\frac{dA_1}{dz} = -i\beta_3 g(z) A_3 A_2^* \exp(-i\Delta_3 z) - i\beta_2 g(z) A_2 A_1^* \exp(-i\Delta_2 z),$$

$$\frac{dA_2}{dz} = +i2\beta_3 g(z) A_3 A_1^* \exp(-i\Delta_3 z) + i\beta_2 g(z) A_1^2 \exp(i\Delta_2 z), \tag{1}$$

$$\frac{dA_3}{dz} = -i3\beta_3 g(z) A_1 A_2 \exp(i\Delta_3 z),$$

It is consider that two waves with frequencies ω_1 and ω_3 are in the left entrance along the positive z direction of the PPLN crystal, ω_2 is backward wave with doubled frequency.$A_{1,3}(z=0) = A_{10,30} \cdot \exp\left(i\varphi_{10,30}\right), A_2(z=L) = A_{2l} \cdot \exp(i\varphi_{2l})$. A_j, $j = 1 \div 3$ complex amplitudes of all three interacted waves at frequencies $\omega_j = j\omega$. $A_{10,30}$, $\varphi_{10,30}$ the initial amplitudes and phases of the waves entering the nonlinear medium from the left, and A_{2l}, φ_{2l} are the initial amplitude and phase of the wave at the doubled frequency entering the nonlinear medium (a crystal of length L). The nonlinear coupling coefficients $\beta_{2,3}$ are related to the generation of the second harmonic and to the frequency mixing process ($\omega + 2\omega = 3\omega$), respectively. n_2 and n_3 are the refractive indices at frequencies ω_2 and ω_3, respectively, and χ^2 is the second-order nonlinear susceptibility of each domain. In order to find complex amplitude of third harmonic wave (1) shortened equations have been solved by CFA and $\lambda^{CFA} = \sqrt{6} \cdot |g_3|\Gamma_{13}$ consideration was taken into account. Then complex amplitude of third harmonic will be:

$$A_3^{CFA}(z) = \left[\left(\frac{A_{2l}}{A_{10}} \cdot \frac{sinh\lambda^{CFA} z}{cosh\lambda^{CFA} L}\right) + \left(\frac{2g_3^*\beta_3 A_{30} + g_2\beta_3 A_{30}}{\sqrt{6}|g_3|\Gamma_{13}}\right) \times\right.$$

$$\left.\times\left(tanh\lambda^{CFA} L \cdot sin\lambda^{CFA} z - cos\lambda^{CFA} z + 1\right)\right]\left(\frac{3\beta_3|g_3|(A_{10})^2}{\sqrt{6}|g_3|\Gamma_{13}}\right).$$

But for second harmonic backward wave complex amplitude has been found with CIA when phase altering of all waves are considering to be and ($I_{1,3}(z) = I_{1,3}(z=0) = I_{10,30} = const$. [10] is taken into account. Then equation which describes behavior of second harmonic backward wave will be:

$$A_2^{CIA}(z) = A_{2l}\frac{cos\lambda z}{cos\lambda l}\exp(i\varphi_{2l}) - \frac{i}{\lambda}A_{10}[2g_3^*\Gamma_3\exp\,[i(\varphi_{30}-\varphi_{10})] +$$

$$g_2\Gamma_{12}\exp(i2\varphi_{10})](sin\lambda z - tan\lambda l \cdot cos\lambda z),$$

where

$$\lambda = \sqrt{2[|g_3|^2(\Gamma_3^2 - 3\Gamma_{13}^2) - |g_2|^2\Gamma_{13}^2]},$$

$$\Gamma_{12} = \beta_2\sqrt{I_{10}}, \Gamma_{13} = \beta_3\sqrt{I_{10}},\ \ \Gamma_3 = \beta_3\sqrt{I_{30}}.$$

Efficiency of the process of generation of third harmonic will be:

$$\eta_3^{CFA} = \left[\left(A_{2l}\cdot\frac{sinh\lambda^{CFA}z}{cosh\lambda^{CFA}L}\right)^2 + \left(\frac{2g_3^*\beta_3A_{30} + g_2\beta_3A_{30}}{\sqrt{6}|g_3|\Gamma_{13}}\right)^2 \times\right.$$

$$\left.\times\left(tanh\lambda^{CFA}L\cdot sin\lambda^{CFA}z - cos\lambda^{CFA}z + 1\right)^2\right]^2\left(\frac{3\beta_3|g_3|I_{10}}{\sqrt{6}|g_3|\Gamma_{13}}\right)^2.$$

3. Results and Discussion

The influence of the ratio of nonlinear coupling coefficients β_3/β_2 of the waves on the dynamics of the second harmonic gain coefficient $I_2(z)/I_{2l}$ is shown in Fig. 2. It can be seen that the gain coefficient decreases with increasing β_3/β_2 (compare curves 1, 2 and 3).

At $M_2 = M_3$=1 (curves 3), the maximum value of η_3^{max} is reached at $(\beta_3/\beta_2)_{opt}$=0.5 in the approximation of a given field and 0.667 in the approximation of a given intensity, the numerical calculation of system (1) for the associated geometry gave the following values 0.67 [6] and 1.128 [7]. With increasing orders of quasi-synchronism, large values of $((\beta_3/\beta_2)_{opt})$ opt are required to obtain a maximum of η_3^{max}.

As it is depicted on the picture 3. $\eta_2 = I_2(z)/I_{10}$ and $\eta_3 = I_3(z)/I_{10}$, efficiencies depend on $\Gamma_{12}z$ at $M_2 = M_3 = 1$ for various relationship between $\beta_{2,3}$,nonlinear coefficient of interaction which is found at constant field approximation (for second harmonic (2 curves) and for third harmonic 3 curves) and constant intensity approximation (1 curves). In the case of counter-propagating interaction, the intensity of the backward second harmonic wave increases more rapidly at low input intensities (curves 1–3). This can be explained by competing processes between the second harmonic (SH) and third harmonic (TH) waves. As β_3/β_2 decreases, the conversion efficiency to the second harmonic increases, while conversion efficiency to the third

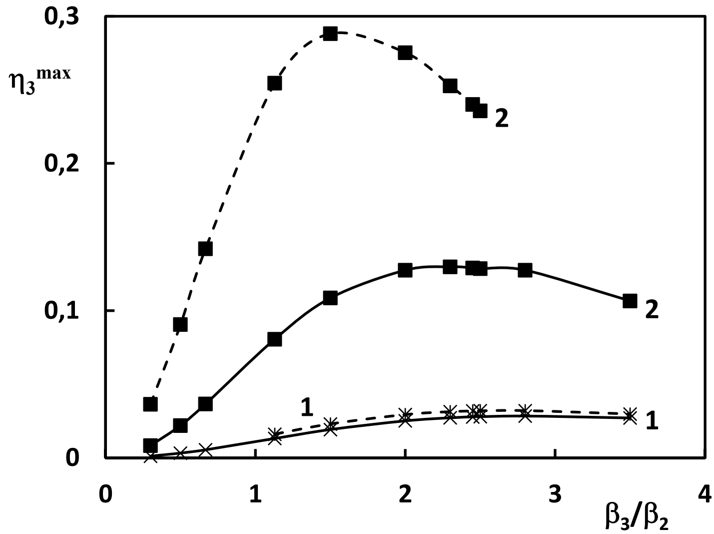

Fig. 2. Maximum conversion efficiency of the pump wave energy into the third harmonic η_3^{max} as a function of parameter β_3/β_2, calculated using the constant-field approximation (dashed lines) and the constant-intensity approximation (solid lines), for I_{2l}=I_{30}=0 at various values of M_2 and M_3: 1 and 3 (curves 2), 3 and 5 (curves 1)

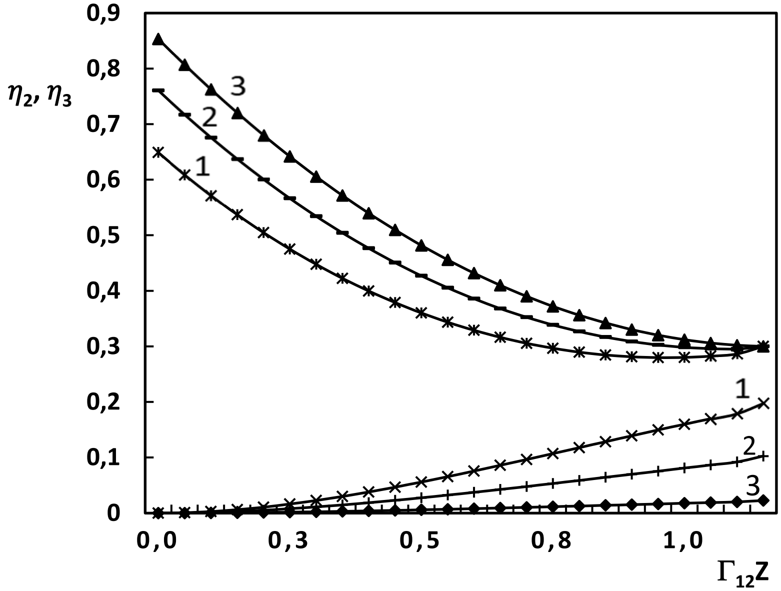

Fig. 3. Dependences of the conversion efficiency $\eta_2 = I_2(z)/I_{10}$ and $\eta_3 = I_3(z)/I_{10}$ on the parameter $\Gamma_{12}z$ for $M_2 = M_3 = 1$ and I_{30}=0.001 at the different I_{2l}: 0.3 (curves 1-3); β_3/β_2: 0.3 (curves 3), 0.67 (curves 2) and 1 (curves 1); All the curves calculated in the CFA and the CIA.

harmonic η$_3$ decreases. For comparison, the results of calculations using the constant-field approximation are also presented. The observed reduction in η$_2$ efficiency under the constant-intensity approximation is due to the consideration of changes in the complex amplitude of the pump wave through phase variation, where as in the constant-field approximation, the amplitude is assumed to be constant [13]. That is, the following condition is satisfied: $A_1(z) = A_{10} = const$, which implies $\varphi_1(z) = \varphi_{10} = const$ (compare 1,2 curves with 3). For $M_2 = M_3 = 1$ (curve 3), the maximum value of η$_3$ is reached at $(\beta_3/\beta_2)_{opt}$*=0.5* in the constant-field approximation and at 0.667 in the constant-intensity approximation. The numerical solution of the system (1) for the co-propagating geometry yielded values of 0.67 [4] and 1.128 [12]. As the orders of quasi-phase matching increase, larger values of $(\beta_3/\beta_2)_{opt}$ are required to achieve the maximum η$_3$.

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