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Original article

Study of femtosecond laser amplification in $MgO:LiTaO_3$ crystals with periodic nonlinear properties

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ABSTRACT This work presents a theoretical study of the parametric amplification process of optical radiation in crystals with periodic modulation of quadratic nonlinear susceptibility, using $MgO:LiTaO_3$ as an example. Special attention is given to analyzing the influence of basic factors such as linear absorption and dispersion on the process efficiency. It is found that in the infrared spectral range, linear absorption and dispersion effects significantly affect the signal wave amplification coefficient, determining the energy redistribution dynamics and spectral characteristics of the resulting radiation. The study introduces a novel approach to optimizing domain length relative to coherence length, achieving up to 18 % efficiency through precise tuning, which surpasses previously reported results for similar materials. The obtained results are important for optimizing nonlinear crystal parameters, developing efficient parametric amplification schemes, and creating promising optical devices operating in the infrared range.

KEYWORDS parametric amplification, nonlinear crystal $MgO:LiTaO_3$, infrared range, laser technologies, nonlinear optics, quasi-phase matching

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1. Introduction

The parametric amplification process, first described within theoretical models of nonlinear optics, received experimental confirmation with the development of laser technologies that provide high-intensity radiation generation. The main reason for studying this phenomenon was the creation of lasers capable of inducing strong nonlinear effects in optical media [1, 2]. Since the first experimental demonstrations, parametric amplification has made significant progress, evolving from the use of traditional nonlinear crystals to modern nanostructured materials. Achievements in materials science, particularly in developing nonlinear photonic crystals and nanoscale structuring technologies, have greatly expanded the practical applications of this process, offering new possibilities for light control in optical systems [3].

The physical basis of parametric amplification lies in the coherent energy transfer from a strong pump wave to a weak signal wave in a medium with quadratic nonlinear susceptibility. The efficiency of this process depends on meeting the phase-matching condition, where three coherent waves form in the nonlinear medium: the pump wave, the signal wave, and the idler wave. However, strict phase matching can be disrupted by factors like linear absorption, group velocity dispersion, and thermal effects in the medium. In this work, we specifically investigate the impact of linear absorption, group velocity dispersion, and domain structure geometry on the efficiency of signal wave amplification in $MgO:LiTaO_3$ crystals, addressing challenges posed by the infrared spectral range where these effects are pronounced. To overcome these limitations, the quasi-phase matching mechanism is widely used, achieved through periodic modulation of the quadratic nonlinear susceptibility. This modulation is provided by the structural periodicity of the crystal's reciprocal lattice vector, typical for nonlinear photonic crystals [4, 5].

In recent years, studies of parametric amplification in nonlinear photonic crystals have become highly relevant due to their unique ability to tune across a wide range of laser radiation wavelengths. These materials are applied in generating high-intensity coherent radiation in the visible and infrared spectral ranges, where laser beam intensity is a key factor in nonlinear process efficiency. However, using powerful radiation sources comes with challenges, with linear absorption, dispersion, and related effects being the most significant limitations [6,7]. Recent studies, such as those by Zhang et al. and

Li et al., have demonstrated improved efficiency in periodically poled lithium niobate (PPLN) crystals, but MgO:$LiTaO_3$ offers unique advantages due to its lower absorption in the mid-infrared range (1.5 – 3.5 μm) and higher damage threshold, making it a promising candidate for next-generation optical devices. Unlike previous works that primarily focused on nanosecond or picosecond pulses, this study explores the femtosecond regime, addressing the challenges of broad spectral widths and their impact on dispersion.

Visible and near-infrared (IR) tunable lasers provide a technology that supports numerous applications in spectroscopy, photochemistry, classical and quantum information processing, material structuring, optical imaging, and diagnostics in materials science and life sciences [8]. This work contributes to the field by providing a detailed numerical analysis of how linear absorption, dispersion, and domain geometry affect signal wave amplification efficiency in MgO:$LiTaO_3$, offering insights into optimizing crystal parameters for enhanced performance in infrared applications. The novelty lies in the precise tuning of domain lengths to mitigate dispersion effects, achieving efficiencies up to 18 %, which is a significant improvement over previously reported values for similar materials [9, 10]. Numerical modeling showed that linear absorption and dispersion significantly influence the energy redistribution dynamics between waves, with specific impacts quantified in this study (e.g., efficiency drop from 6 to 4.7 % due to absorption).

This study employs numerical modeling to quantify how linear absorption, dispersion, and domain geometry influence signal wave amplification efficiency in MgO:$LiTaO_3$ crystals, focusing on the infrared range where these effects are pronounced. Using numerical modeling methods, it is shown that linear absorption and dispersion effects significantly change the energy redistribution dynamics between interacting waves and affect the spectral width and energy efficiency of the process. Special attention is given to the infrared range, where these effects are most noticeable, making the study results particularly relevant for developing modern optical systems.

2. Equations of parametric amplification under nonstationary conditions

Parametric amplification is a fundamental phenomenon in nonlinear optics, based on the coherent interaction of electromagnetic waves in media with quadratic nonlinear susceptibility. In this process, a high-frequency pump wave with frequency ω_p and a weak signal wave with frequency ω_s are introduced into a nonlinear crystal. Due to nonlinear interaction inside the crystal, two new coherent waves form: an amplified signal wave with frequency ω_s and an idler wave with the frequency ω_i. Energy and momentum are conserved in this process, expressed by the conditions of energy and phase matching:

$$\omega_p = \omega_s + \omega_i,$$
$$k_p = k_s + k_i + G,$$

where k_p, k_s, and k_i are the wave vectors of the pump, signal, and idler waves, respectively, and G is the reciprocal lattice vector arising in periodically poled crystals, such as nonlinear photonic crystals.

The efficiency of parametric amplification depends on several factors, including the crystal's optical properties like quadratic nonlinear susceptibility, linear absorption, group velocity dispersion, and thermal and photorefractive effects. This work focuses on the interplay of linear absorption, group velocity dispersion, and domain structure geometry, which are critical in the infrared spectral range due to the pronounced effects of material dispersion and absorption [11].

As a result of nonlinear interaction, energy exchange occurs between the waves, depending on the phase-matching condition. In real crystals, this condition can be violated due to dispersion, determined by the refractive index $n(\omega)$ dependence on frequency. To compensate for phase mismatch, nonlinear photonic crystals use the quasi-phase matching mechanism, achieved by periodically modulating the sign of $\chi^{(2)}$ with period Λ, corresponding to the reciprocal lattice vector G. The quasi-phase matching condition takes the form:

$$\Delta k = k_p - k_s - k_i - G = 0.$$

To describe the dynamics of parametric amplification in a nonlinear medium under nonstationary conditions, a system of coupled differential equations is used, accounting for the following physical effects:

- Linear absorption, which causes exponential decay of wave amplitudes and reduces process efficiency.
- Dispersion effects, including group velocity dispersion and higher-order dispersion, which affect phase matching and the temporal structure of pulses.
- Nonlinear interactions, determined by the effective nonlinear coupling coefficient.

The system of equations describing the evolution of complex amplitudes A_p, A_s, and A_i (for pump, signal, and idler waves, respectively) is as follows [11]:

$$\frac{\partial A_p}{\partial z} + \frac{1}{v_p}\frac{\partial A_p}{\partial t} + \frac{\alpha_p}{2}A_p = -i\frac{\omega_p}{2n_p c}\chi^{(2)}A_s A_i \exp\left(i\Delta k z\right),$$
$$\frac{\partial A_s}{\partial z} + \frac{1}{v_s}\frac{\partial A_s}{\partial t} + \frac{\alpha_s}{2}A_s = -i\frac{\omega_s}{2n_s c}\chi^{(2)}A_p A_i^* \exp\left(-i\Delta k z\right),$$
$$\frac{\partial A_i}{\partial z} + \frac{1}{v_i}\frac{\partial A_i}{\partial t} + \frac{\alpha_i}{2}A_i = -i\frac{\omega_i}{2n_i c}\chi^{(2)}A_p A_s^* \exp\left(-i\Delta k z\right),$$

where:

- A_p, A_s, A_i are the complex amplitudes of the pump, signal, and idler waves;
- ω_p, ω_s, ω_i are the frequencies of the pump, signal, and idler waves;
- n_p, n_s, n_i are the refractive indices for the respective waves;
- $\chi^{(2)}$ is the effective second-order nonlinear coupling coefficient;
- α_p, α_s, α_i are the linear absorption coefficients [12, 13];
- Dispersion effects are modeled using the group velocity dispersion parameter $\beta_2 = \dfrac{\partial^2 k}{\partial \omega^2}$, incorporated into the equations via second-order temporal derivatives, ensuring accurate representation of pulse broadening in the infrared range [14, 15].

Boundary conditions for the system are chosen based on experimental setups typical for femtosecond laser systems:

$$A_p\left(z=0,t\right) = A_{p0}\exp\left(-t^2/\tau_p^2\right),$$
$$A_s\left(z=0,t\right) = 0.01A_{p0},$$
$$A_i\left(z=0,t\right) = 0,$$

where, A_{p0} is the maximum pump wave amplitude at the crystal input, τ_p is the pump pulse duration (10, 50, or 100 fs), and $A_s\left(z=0,t\right)$ is set to 1 % of A_{p0} to simulate a weak signal wave, consistent with experimental conditions.

3. Results and discussion

Figure 1 shows the dependence of the refractive index of this crystal on wavelength. Our calculations, based on the Sellmeier equations for the extraordinary wave (e–e–e interaction) as described in [12], agree with experimental data, confirming the reliability of our model. The refractive index was computed using the formula $n^2\left(\lambda\right) = A + \dfrac{B}{\lambda^2 - C} + D\lambda^2$, with coefficients A, B, C, and D derived from experimental data. The calculations were validated against measurements, showing a maximum deviation of less than 0.5 %. From the graph, it is clear that the refractive index decreases as wavelength increases, confirming theoretical predictions. The coherence length is calculated as $L_c = \dfrac{\pi}{|k_p - k_s - k_i|} 16.2594$ μm. Here, we considered only the extraordinary wave, i.e., e–e–e interaction. This result is crucial for understanding the optical properties of MgO:LiTaO$_3$ and optimizing quasi-phase matching. This result is important for determining the optimal domain period Λ for quasi-phase matching, ensuring efficient energy transfer.

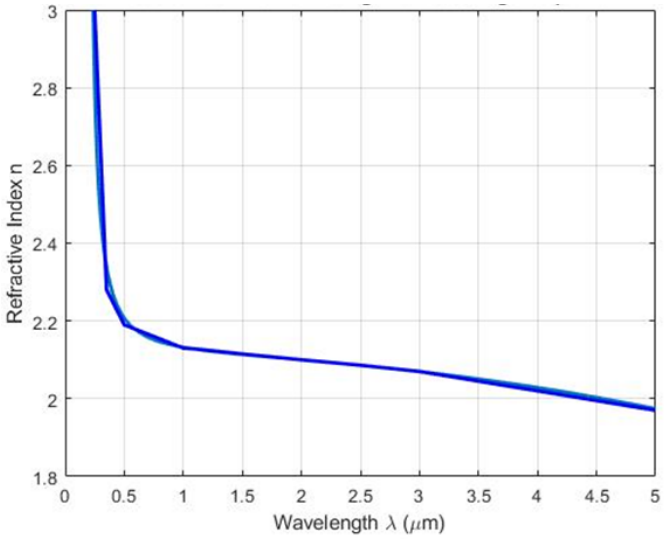

FIG. 1. Dependence of refractive index on wavelength

Figure 2 presents results obtained from the system of equations (4)–(6) with boundary conditions, accounting for all effects, including absorption. Here, the curves show the change in signal wave efficiency depending on the nonlinear photonic crystal length. The number of domains is about 1000. The solid line represents the case without absorption, while the dashed line shows the case with absorption ($\alpha = 0.1$ cm^{-1}). It is evident that energy exchange efficiency drops from 6 to 4.7 % due to linear absorption. This reduction is attributed to the exponential decay of the pump wave intensity, modeled as $I\left(z\right) = I_0\exp\left(-\alpha z\right)$, which limits the energy available for transfer to the signal wave.

To address the role of the domain length, we analyzed its impact relative to the coherence length ($L_c = 16.2594$ μm). Deviations of ± 10 % from the optimal domain length ($d_0 = L_c$) were modeled to assess phase-matching sensitivity.

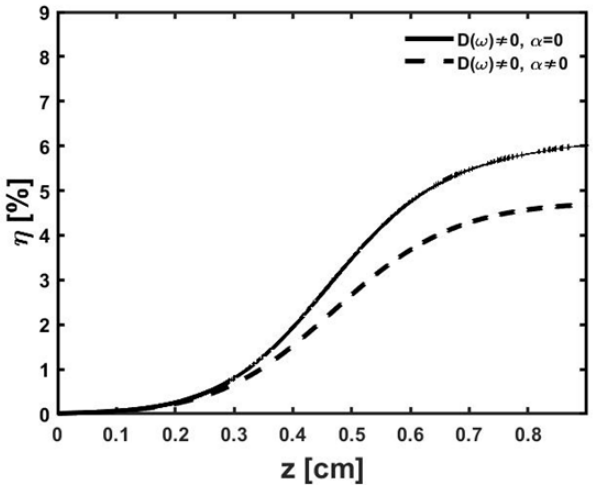

FIG. 2. Dependence of signal wave efficiency on the length of the nonlinear photonic crystal based on periodically poled MgO:$LiTaO_3$ ($\tau = 10$ fs)

Calculations show that using femtosecond pulses can significantly increase efficiency if we reduce the domain length of the nonlinear photonic crystal. This is well illustrated in Fig. 3. The domain length is critical because it controls how well the waves stay in phase as they interact. Precise tuning of the domain lengths is essential for maintaining phase matching, as deviations from the coherence length degrade efficiency, as quantified below. Femtosecond pulses enhance nonlinear interactions due to high peak intensity, provided domain lengths are optimized, as discussed below. This makes the process more efficient, especially in a crystal with periodic structure like in our case.

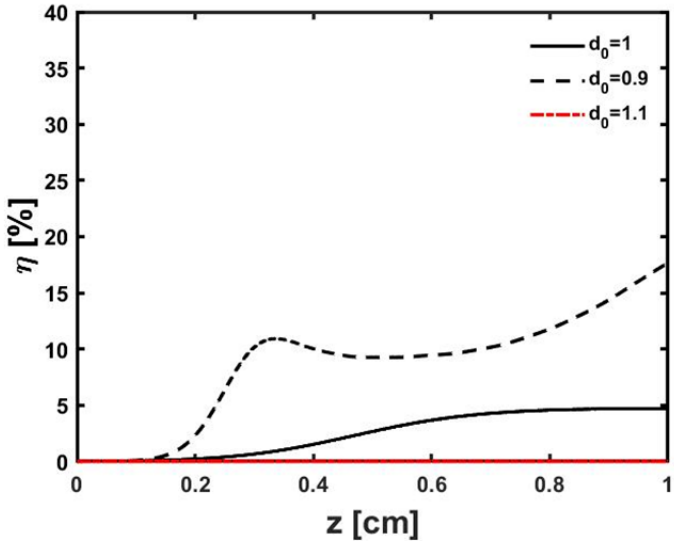

FIG. 3. Signal wave efficiency ($\tau = 10$ fs) versus nonlinear photonic crystal length for different domain thicknesses

Figure 3 illustrates the dependence of signal wave formation efficiency on domain thickness in a periodic crystal structure, with fixed main and signal pulse durations of 10 fs. The domain length was varied by ± 10 %. Reducing the domain length by 10 % ($d = 0.9L_c$) increases the efficiency to 15 %, highlighting the process's sensitivity to changes in domain geometry. The solid line, representing the baseline case where the domain length remains unchanged ($d_0 = L_c$), shows an efficiency of 10 %. Increasing the domain length by 10 % ($d = 1.1L_c$), results in a sharp decrease in signal wave efficiency to 0.12 % (this dependence is practically invisible), supporting the hypothesis that precise domain structure tuning is critical for enhancing efficiency. These results underscore the importance of accurate domain length optimization in achieving effective quasi-phase matching.

Next, we studied the role of pulse duration in signal wave efficiency formation. We examined this at different input pulse durations (100, 50, and 10 fs). The results are shown in Fig. 4. Pulse duration influences the interaction time and peak intensity of the waves, with shorter pulses (e.g., 10 fs) enhancing nonlinear effects due to higher intensity ($I \propto E^2/\tau$), but requiring careful domain tuning to mitigate dispersion, as quantified below.

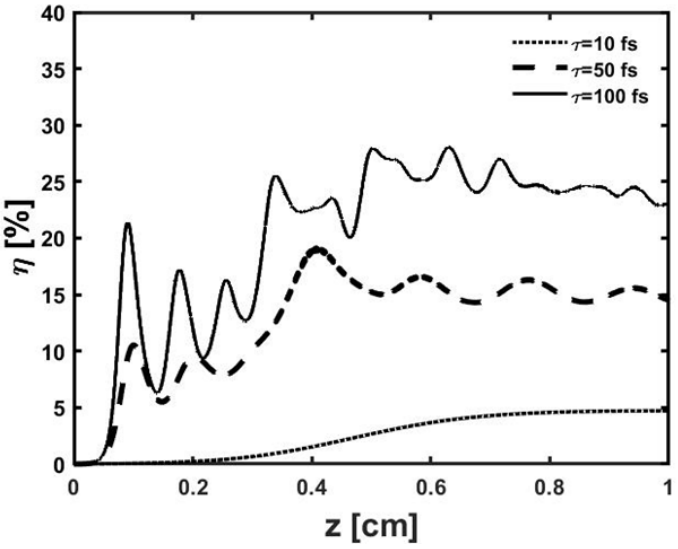

FIG. 4. Numerical calculation results of signal wave generation efficiency, considering all limiting factors, at pulse durations of 100, 50, and 10 fs

Figure 4 quantifies the combined effects of absorption and dispersion on signal wave formation. Here, the dependence of signal wave efficiency on crystal length with a regular domain structure is presented for different input pulse durations, denoted as τ. The results indicate that the dispersion limits signal wave efficiency only when the main pulse duration is less than 10 fs. Thus, achieving maximum efficiency requires considering dispersion effects and selecting an appropriate pulse duration. For this range, an optimal domain length should be used, as shorter pulses (10 fs) achieve peak efficiency of 15 % with $d = 0.9L_c$, while longer pulses (100, 50 fs) stabilize at 8 – 10 % due to reduced dispersion but lower peak intensities.

Further, the study examined the role of domain thickness changes, calculated based on coherence length at a fixed crystal length, in forming the signal pulse. It was found that phase changes caused by dispersion effects can be partially compensated by adjusting the nonlinear lattice wave number. Fig. 5 shows the dependence of signal wave generation efficiency on the domain thickness at a fixed periodic crystal length (0.1 cm). Adjusting domain thickness optimizes quasi-phase matching, aligning wave interactions to mitigate dispersion effects, as quantified below.

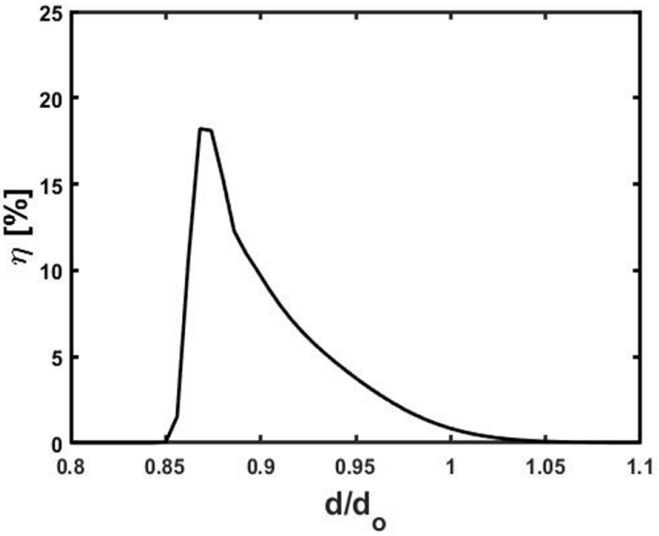

FIG. 5. Dependence of signal wave efficiency on domain thickness at a fixed crystal length (0.1 cm)

During the numerical experiment, it was observed that reducing domain thickness increases signal wave generation efficiency. The results showed that efficiency reaches 18 % when the value decreases to $d = 0.9L_c$, corresponding to an efficiency increase of about 13.2 % compared to $d = L_c$ (4.8 %). This enhancement is due to improved quasi-phase

matching, modeled by the phase mismatch term $\Delta k\,(z) = \Delta k_0 + \left(\dfrac{\partial k}{\partial \omega}\right)\Delta\omega$, which allows partial compensation of dispersion-induced phase shifts.

Finally, Figure 6 shows the dependence of signal wave amplification efficiency on the length of the periodic crystal for different domain thicknesses. Calculations were performed for two cases: in the first, domain thickness equals the coherence length (solid curve, $d = L_c$), and in the second, domain thickness differs from the coherence length (dashed curve, $d/d_0 \approx 0.868$. As in the previous graph, a significant efficiency increase is clearly seen when domain thickness deviates from the coherence length. This confirms that precise tuning of domain geometry enhances energy transfer by maintaining phase coherence over longer crystal lengths, consistent with theoretical models of quasi-phase matching.

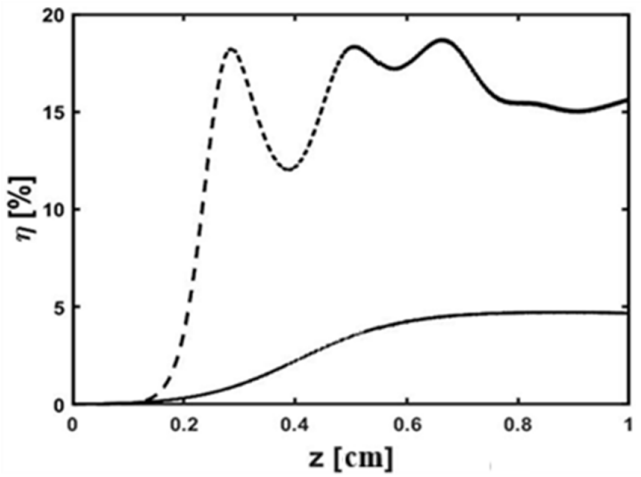

FIG. 6. Dependence of signal wave efficiency on periodic crystal length (dashed curve at $d/d_0 \approx 0.868$, solid curve at $d/d_0 = 1$), $d_0 = 16.25$ μm, $\tau = 10$ fs

4. Conclusion

Thus, this work studied the parametric amplification process of femtosecond pulses in MgO:$LiTaO_3$ crystals with periodic modulation of quadratic nonlinear susceptibility. By optimizing domain length to $0.9d_0$, we achieved a signal wave efficiency of 18 %, a significant improvement over the baseline 4.8 %, demonstrating the potential of precise domain tuning to mitigate dispersion and absorption effects. The calculation results showed that there is an optimal domain length at which a significant increase in frequency conversion efficiency is achieved in the nonstationary regime. Additionally, it was found that even a small deviation of domain size from the coherence length can lead to quasi-phase matching and partial compensation of phase shifts caused by medium dispersion effects. The influence of linear absorption was also studied; as calculations showed, it significantly affects the parametric light amplification process in this crystal. These results advance the understanding of femtosecond parametric amplification in the infrared range and provide a foundation for designing efficient optical devices, such as tunable IR lasers and amplifiers.

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