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On the generation of optical third-harmonic through quasi-phase-matching in quadratic nonlinear crystals

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ABSTRACT

We investigate the simultaneous processes of optical frequency doubling and sum frequency generation in quasi-phase-matched quadratically nonlinear crystals. Specifically, we focus on lattices with domain thicknesses corresponding to the coherence lengths of the two cascaded parametric processes and explore the realistic scenario of a single crystal containing sequential domain segments, each independently satisfying quasi-phase-matching for optical frequency doubling and summing, respectively. We demonstrate that high conversion efficiencies to the third harmonic can be obtained from an arbitrary fundamental wavelength.

1. Introduction

Cascaded parametric interactions have been demonstrated capable of mimicking and yielding four-wave mixing effects with high conversion efficiencies [1–11]. One recent example is the combination of frequency doubling (SHG) with sum frequency generation (SFG) to yield frequency tripling, i.e., third-harmonic generation (THG) (see e.g., [12,13] and references therein). This is a remarkable result inasmuch as such parametric conversion cannot take place in homogeneous bulk crystals owing to dispersion of the phase velocity with wavelength, and therefore, the refractive index mismatch due to $n(\omega) \neq n(2\omega) \neq n(3\omega)$, being ω, 2ω and 3ω the fundamental frequency, its second harmonic and its third harmonic, respectively [14,15].

With the discovery and implementation of domain engineering in ferroelectric quadratically nonlinear crystals, the implementation of the quasi-phase-matching (QPM) technique for SHG and SFG has greatly increased the conversion efficiency via a mean velocity match along the propagation distance, effectively compensating the detrimental effect of Maker fringes [16,17]. In crystals with periodic nonlinear domains, QPM at first [18] or higher orders [19] allows for the cascading of second-harmonic and sum frequency generation to mimic third-harmonic generation in crystals for parametric conversion [20]. It has been proven that the efficient implementation of such an effective THG would require either an optimum amplitude ratio of fundamental to double frequency waves at the input of the sample [19,21], or coupled nonlinearities [21–24], or a nonlinear grating with spectrum optimized in Fourier space [25,26], etc. Notably, non-periodic lattices with domains in a Fibonacci sequence [27], randomized domains [28], spatial inhomogeneities [29] could also entail the possibility of frequency tripling via combined SHG and SFG.

In this work, we illustrate third-harmonic generation from an arbitrary input frequency by employing an optimized sequence of domain lengths for cascaded second-harmonic and sum frequency generation in a parametric crystal. We utilize the QPM approach in a non-homogeneous crystal with domains of opposite nonlinear susceptibilities and two unequal thicknesses, corresponding to the coherence lengths of the two conversion processes, respectively frequency doubling and frequency summing. We monitor the

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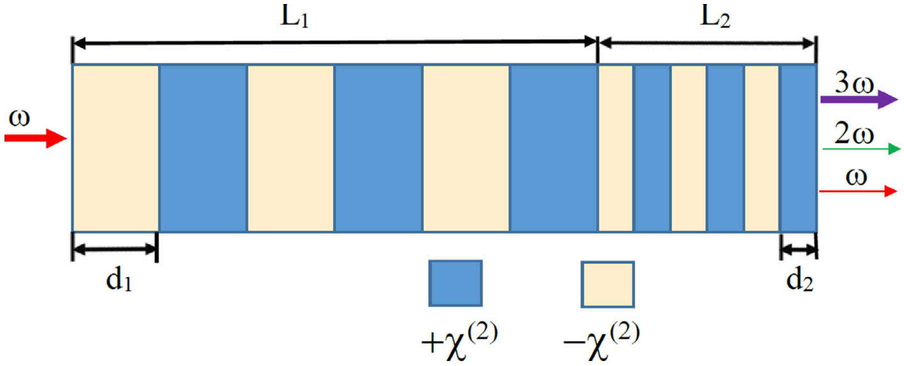

Fig. 1. Artist's rendering of a QPM lattice with domain thicknesses d_1 and d_2 of alternating signs of the quadratic susceptibility $\chi^{(2)}$. Here, d_1 and d_2 are the coherence lengths for the interactions $2\omega = \omega + \omega$ and $3\omega = 2\omega + \omega$, respectively. The QPM sections L_1 and L_2 separately phase-match second-harmonic and sum-frequency generation.

simultaneous generation of double and triple frequencies. The results of numerical experiments show that optical tripling can be achieved with high conversion efficiency from input at an arbitrary fundamental wavelength. The presented design-based ratio of domain segments is an effective and convenient approach for optimizing cascaded parametric interactions in quadratic crystals and third-harmonic generation.

2. Model

At variance with previously reported methods, the approach presented here requires neither a specific QPM-order for the simultaneous generation of the 2nd and 3rd harmonics of a fundamental input, nor the simultaneous input of two (or three) frequencies at the excitation boundary. Along propagation, we consider a nonlinear susceptibility grating with two sections: the first exclusively phase-matched to produce the second-harmonic, the second phase-matched to generate the third-harmonic via SFG. Fig. 1 is a sketch of the examined case: a QPM lattice encompasses domain thicknesses d_1 and d_2 of alternating signs, i.e., opposite nonlinear second-order response $\chi^{(2)}$. d_1 and d_2 are equal to the coherence lengths of the interactions $2\omega = \omega + \omega$ and $3\omega = 2\omega + \omega$, respectively.

In order to keep the complexity of the model and the design effort at minimum levels, we focus hereby on the parametric interaction of plane waves using first-order perturbation theory and the slowly varying phase and envelope approximation, assuming Kleinmann symmetry for the crystal and wavelength involved, and Type 0 (ee-e) three-photon mixing between triplets of extraordinarily-polarized wave components [12] :

$$\begin{aligned} \frac{dA_1}{dz} &= \delta(z)A_1^* A_2 e^{-i\Delta k_2 z} + \delta(z)A_2^* A_3 e^{-i\Delta k_3 z} \\ \frac{dA_2}{dz} &= 2\delta(z)A_1^* A_3 e^{-i\Delta k_3 z} + \delta(z)A_1^2 e^{i\Delta k_2 z} \\ \frac{dA_3}{dz} &= 3\delta(z)A_1 A_2 e^{i\Delta k_3 z} \end{aligned} \tag{1}$$

where $\delta(z) = -i(4\pi^2\chi^{(2)})/(n(\lambda)\lambda) * f(z)$ with the function $f(z)$ being the spatially changing polarity; $A_j (j = 1, 2, 3)$ are the electric field amplitudes and $k_j (j = 1, 2, 3)$ are the wavenumbers at the involved frequencies ω, 2ω, and 3ω, respectively; $\Delta k_2 = k_2 - 2k_1$ is the phase mismatch for frequency doubling, $\Delta k_3 = k_3 - k_2 - k_1$ is the phase mismatch for SFG towards frequency tripling. To calculate the coupling coefficients in system (1) we assumed $1/n(\lambda) \approx 1/n(\lambda/2) \approx 1/n(\lambda/3)$ for simplicity. Nevertheless, in realistic cases such as periodically poled Lithium Niobate, the results would be essentially unaffected.

3. Results and discussion

First, we focus on the dynamics of three harmonics that are mutually coupled in an ideal and dimensionless nonlinear scenario with no losses. To this extent, we assume $|\delta(z)| = 1$, $d_1 = 0.03$, $d_2 = 0.01$ and $N = 500$, where N is the total number of domains. For the boundary conditions, since we wish to realize a frequency tripler, we set $A_1(z = 0) = 1$, $A_2(z = 0) = 0$, $A_3(z = 0) = 0$. We analyze three distinct cases: $L_1/L_2 = 1$, $N_1/N_2 = 1$, and $N_1/N_2 = \alpha$, where N_1 is the total number of domains of the first section and N_2 is the total number of domains of the second section, with $N_1 + N_2 = N$. Here, α is the optimum ratio between domain numbers, allowing for complete energy conversion to the third harmonic wave. One can easily find $L_1/L_2 = \alpha d_1/d_2$.

Numerical results of this analysis are plotted in Fig. 2 for three examples. All numerical calculations were checked by ensuring a constant sum of energy fluxes of the interacting harmonics. Each domain was divided in 10 portions to maintain accuracy. The graphs reveal that, for the specified configuration and the chosen parameters, α approximately equals 0.15 (corresponding to $N_1 = 66$ and

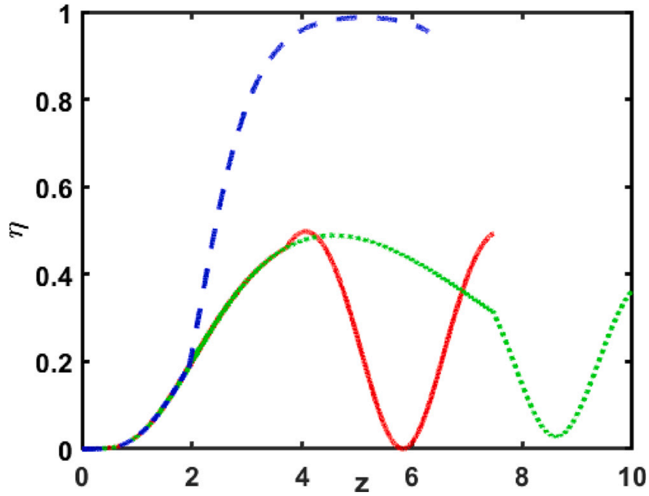

Fig. 2. Dynamics of energy conversion to a frequency triple of the input fundamental after numerical experiments in a dimensionless sample with $|\delta(z)| = 1$, $d_1 = 0.03$, $d_2 = 0.01$, and $N = 500$. Here, the solid (red), dotted (green), and dashed (blue) lines correspond to $L_1/L_2 = 1$, $N_1/N_2 = 1$, and $N_1/N_2 \approx \alpha$, respectively, with a chosen $\alpha \approx 0.15$. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

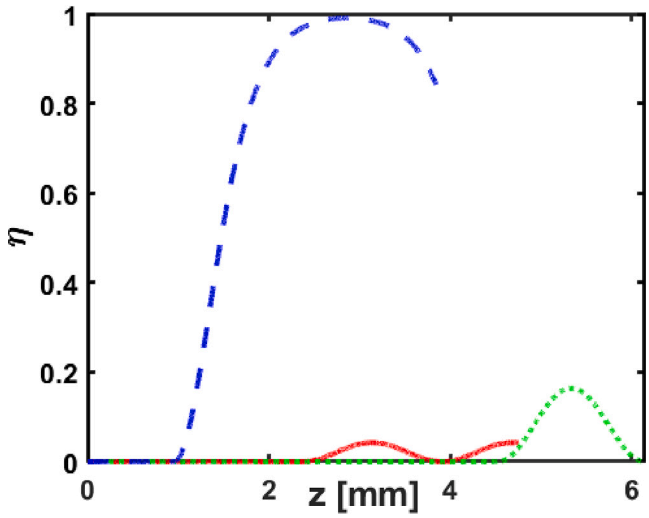

Fig. 3. Results of numerical experiment in a realistic PP-LN sample ∼6 mm long; the input fundamental wavelength is $\lambda = 1.52$ μm and the intensity 0.1 GW/cm^2; the total domain number is set to N = 1000. Here $\alpha \approx 0.12$. The line types are as in Fig. 2.

$N_2 = 434$). The blue dashed line refers to the latter value. The solid and dotted lines indicate that neither $L_1/L_2 = 1$, nor $N_1/N_2 = 1$ support full energy conversion to the third harmonic.

The results presented in Fig. 2 highlight the existence of an optimal ratio of domain numbers (section length), which significantly affects the energy growth/evolution of the third harmonic versus propagation distance. Additionally, it is paramount that the outcome is highly sensitive to the value α, as even small deviations from the best (resonant) value can greatly reduce the conversion efficiency η.

Next, we investigated the same case under realistic material conditions. In order to do this, we selected a periodically-poled z-cut bulk crystal of Lithium Niobate. Both linear and nonlinear optical characteristics of this crystal have been extensively studied [30]. In such material, quadratic interactions can benefit of the highest second-order nonlinear susceptibility for the copolarized extraordinary waves in a Type 0 interaction. We calculate the energy exchange versus z between the three harmonics in a ∼ 6 mm long sample of periodically-poled Lithium Niobate (PP-LN) crystal, which consists of two sections: in the first quasi-phase matching is satisfied for SHG; in the second for SFG of fundamental and second-harmonic. For th enumerical experiment we took a fundamental wavelength $\lambda = 1.52$ μm, an intensity of 0.1 GW/cm^2, N = 1000, $d_{eff} = d_{33} = \chi^{(2)}_{33}/2 = 26$ pm/V, $d_1 \approx 9.07$ μm and $d_2 \approx 3.23$ μm [12].

Fig. 3 indicates that an optimal value $\alpha \approx 0.12$ (corresponding to $N_1 = 106$ and $N_2 = 894$). practically yields complete energy conversion of energy from the fundamental to the third harmonic waves (blue dashed line); conversely, the length ratio $L_1/L_2 = 1$ or the domain number ratio $N_1/N_2 = 1$ does not allow a substantial growth of energy transfer from the fundamental to the triple harmonic. It is worth to pinpoint that the difference between the resonant values of α in dimensionless (Fig. 2) and realistic (Fig. 3) cases stems from the input intensity values in the two examples, as this parameter gets affected by both the second-order nonlinear coefficient and the optical intensity. The precise determination of α appears to require an analytical approach and has to be tailored to a specific (cascaded) frequency conversion process. We aim to tackle this analytical task in forthcoming research activities.

4. Conclusions

In conclusion, our numerical study on THG in parametric crystals delved into cascaded process involving optical sum-frequency generation within quasi-phase-matched structures, concurrently with frequency-doubling. We dealt with single-crystal quasi-phase-matched gratings featuring two sequential sections, each independently satisfying phase-matching for the two parametric interactions, namely SHG and SFG. Our results underscore the potentials of QPM design for large conversion efficiencies in parametric cascading, namely frequency tripling, for any fundamental wavelength input. The required realization technologies and fabrication tolerances, based upon, e.g., textured growth of semiconductors or periodic poling of ferroelectric crystals, are well within reach.

IRB statement

This study adheres to ethical principles approved by the Tashkent State Technical University Review Board (TSTURB). Prior to commencement, it underwent review and approval by the IRB, ensuring participant safety and ethical standards.

Informed consent

Participants received comprehensive information about study objectives, methods, and associated risks. They had the opportunity to ask questions before providing voluntary, written consent.

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CRediT authorship contribution statement

Usman Sapaev: Writing – original draft, Methodology, Investigation, Formal analysis, Conceptualization. **Gaetano Assanto:** Writing – review & editing, Supervision, Conceptualization.

Declaration of competing interest

The authors declare no conflicts of interests

Data availability

Data will be made available on request.

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