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Direct Third Harmonic Generation in Single Quadratic Crystal in Quasi Phase Matched Regime

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Abstract. We present a theoretical investigation of the process of phase matched direct third harmonic (TH) generation in single quadratic crystal with periodical change of the sign of the second-order nonlinearity. The phase matching is achieved with quasi phase matching by appropriate choice of the period of the periodical change of the sign of the crystal second-order nonlinearity. The peculiarity in our case is that the phase matched direct TH radiation is generated as a result of the cascading of two *non phase matched* second-order processes: (i) second harmonic generation and (ii) frequency mixing of the second harmonic field and the fundamental field. In contrast to the process of TH generation in bulk birefringence second-order nonlinear crystals, where the phase matched TH wave is a sum of the contributions of both intrinsic cubic nonlinearity and cascaded $[\chi^{(2)} : \chi^{(2)}]$ cubic nonlinearity, in the case considered here only the cascaded $[\chi^{(2)} : \chi^{(2)}]$ cubic nonlinearity contributes to the phase matched TH signal. The developed model enables the optimization of the design parameters (period, duty factor) of periodically poled quadratic crystals for efficient TH generation at a given laser wavelength. As an example we analyzed the direct frequency tripling of 1500 nm radiation in periodically poled $LiNbO_3$ crystal with period 18 μm and found that conversion efficiency up to 25% could be achieved with moderate pump intensities.

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1 Introduction

The third harmonic generation (THG) is well known nonlinear process used for frequency tripling of laser light sources. For best efficiency the process has to

be phase matched, *i.e.*, the phase velocity of the fundamental wave should be equal to the phase velocity of the third harmonic wave. The birefringence phase matched THG governed by the intrinsic cubic nonlinearity $\chi^{(3)}_{\rm int}$ of the media has been demonstrated in centrosymmetric crystals [1–4]. This method however is not suitable for practical applications due to the low conversion efficiency as a result of the very small magnitude of the involved in the process components of the tensor $\chi^{(3)}_{\rm int}$. Instead, two consecutive second-order nonlinear crystals in which, the first second-harmonic generation (SHG) and then the sum-frequency generation (SFG) of the second harmonic (SH) and the fundamental wave (FW) have been commonly used for efficient frequency tripling. As the processes of SHG and SFG are performed in two separate crystals both of them can be easily phase matched. The drawback of two-crystal scheme is that it is not compact and requires refocusing of the output from the first crystal. More recently it has been suggested that the processes of SHG and SFG can be simultaneously phase matched in a single second-order nonlinear crystal and high TH conversion efficiencies have been demonstrated [5-9]. It is important to emphasize that in the last case the efficiency of the THG process is proportional to the squared effective cascaded cubic nonlinearity $\chi^{(3)}_{\rm casc} \propto \left[\chi^{(2)}\chi^{(2)}\right]$. If both steps SHG and SFG are phase matched or even only one of them is phase matched, the cascaded cubic nonlinearity $\chi^{(3)}_{\rm casc}$ is higher in value than $\chi^{(3)}_{\rm int}$ by several order of magnitudes and the generated TH is a result only of the cascaded processes.

Generally, the phase matched cascaded THG can be generated in single quadratic crystal in four different situations (see, *e.g.* [10,11]): (a) when only the process of SHG is phase matched; (b) when only the process of sum frequency mixing (SFM) is phase matched; (c) when both processes are not phase matched, but the mismatches for both processes SHG and SFM are equal in magnitude but opposite in sign; and (d) when the two processes SHG and SFM are phase matched simultaneously. The case (d) offers the most efficient way for THG in single crystal. The maximum efficiency achieved so far is 20–25% [5-7]. The drawback of method (d) is the necessity of providing simultaneous phase matching of the two second-order processes, which is not an easy task [9]. For this reason the case (c), that we call *direct* THG process could be a promising alternative for efficient frequency tripling. We use the terminology direct in order to stress that neither SHG step $\omega+\omega=2\omega$, nor SFM step $\omega+2\omega=3\omega$ is phase matched, but in the same time the overall process $\omega+\omega+\omega=3\omega$ is phase patched. To the best of our knowledge this case of direct THG in quadratic media has been investigated only in bulk homogeneous crystals [12–14]. The phase matching conditions $\Delta k_3 = k_3 - 3k_1 \to 0$ (where k_3 and k_1 are the wave vectors of the fundamental and the third harmonic waves respectively) were satisfied with utilization of the birefringence and efficiencies up to 6% have been reported [12]. In a recent paper [15] we have presented what we believe to be the first experimental demonstration of direct THG in quadratic crystals with periodical change of $\chi^{(2)}$ by frequency tripling of 1.064 μm radiation in PP-LNB

with period 7 μm. We have shown that in this case the phase matching condition for direct THG has to be modified to $\Delta k_3 = k_3 - 3k_1 - G_m \to 0$, where G_m is one of the reciprocal vectors of the QPM periodic structure. The obtained efficiency was estimated to be about 10^{-4}% for relatively low input intensity (below 400 KW/cm^2) and for duty factor $D = 0.6$ and fifth QPM order. Finally in [15], a simplified analytical model applicable for low conversion efficiencies, when the depletion of fundamental wave could be neglected, was developed.

The purpose of the present paper is the development of a numerical model that enables the optimization of the design parameters (period, duty factor) of periodically poled quadratic crystals for efficient TH generation at a given laser wavelength. By direct solution of the respective slowly varying envelope equations we show that using the 4$^{\text{th}}$ QPM order one can achieve 25% intensity conversion from fundamental to third harmonic with a periodically poled-$LiNbO_3$ crystal with period of 18 μm and duty factor 0.5. Furthermore, in part 4 we propose a way for further improvement of the analytical model published in [15] that extends its applicability towards higher conversion efficiencies. The results of our numerical simulations are compared with those of the improved analytical model and an excellent agreement is observed with intensities for which the conversion is below 15%.

2 Phase Matching Conditions

First we will calculate the optimal period of sign reversal of the second-order nonlinearity, necessary for achieving phase matching for direct THG. For taking advantage of the highest second-order component of $LiNbO_3$ $d_{33} = d_{zzz}$, we chose all three waves: the fundamental one, the second harmonic and the third harmonic to be polarized along "z" axis. The bulk phase mismatches for the processes of direct THG, SHG and SFM are $\Delta k_3 = k_3 - 3k_1$, $\Delta k_1 = k_2 - 2k_1$ and $\Delta k_2 = k_3 - k_2 - k_1$, respectively. Note that $\Delta k_3 = \Delta k_2 + \Delta k_1$. As already mentioned the phase matching for the direct THG could be achieved by compensation of the bulk phase mismatch with one of the reciprocal wave vectors of the grating created by the periodical change of the sign of the nonlinearity in the crystal: $\Delta k_3 + \dfrac{2\pi}{\Lambda} H \approx 0$. Here Λ is the period of the grating and H is an integer called QPM order. This PM condition can be presented as

$$\Delta k_3 + \frac{2\pi}{\Lambda} H = \left(\Delta k_1 + \frac{2\pi}{\Lambda} m\right) + \left(\Delta k_2 + \frac{2\pi}{\Lambda} l\right), \qquad (1)$$

where m and l are integers with $m + l = H$.

The two terms in RHS of (1) we denote as $\Delta k_{1,m}$ and $\Delta k_{2,l}$. The LHS of (1) we denote as $\Delta k_{3,H}$. The necessary periods of the second-order nonlinearity sign reversal for achieving phase matched direct third harmonic (*i.e.* satisfying Eq. (1)) are shown as a function of the fundamental wavelength in Figure 1a for

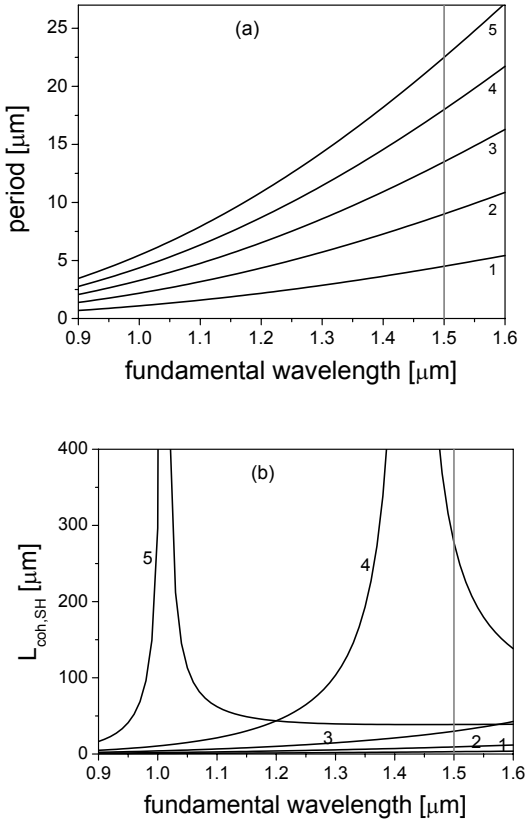

Figure 1. The necessary periods of the second-order nonlinearity sign reversal for achieving phase matched direct third harmonic (a), and the second harmonic generation coherence length (b) as a function of the fundamental wavelength for the first five QPM orders ($H = 1 \dots 5$). The vertical lines indicate the design fundamental wavelength $\lambda_1 = 1,5\ \mu$m.

the first five QPM orders ($H = 1 \dots 5$). They are calculated using Sellmeier equations [16] according to the formula $\Lambda_H = 2\pi H/\Delta k_3$. For fundamental wavelength $1.5\ \mu$m and temperature $80°$C the design periods for the first 5 QPM orders are $\Lambda_H = \{4.5;\ 9;\ 13.5;\ 18;\ 22.5\}\ \mu$m, respectively.

For bulk quadratic crystals the efficiency of the generated direct third harmonic depends on the effective cubic nonlinearity that has the form [10,11,17]

$$\chi^{(3)}_{\rm casc} \propto \frac{d^{(2)}_{\rm SHG} d^{(2)}_{\rm SFM}}{\Delta k_1} = \frac{d_{zzz} d_{zzz}}{\Delta k_1}, \qquad (2)$$

where we remind that $\Delta k_1 = k_2 - 2k_1$ is the bulk mismatch of the SHG process. The effective cubic nonlinearity expressed by the coherence length $L_{\rm coh,SH} =$

$\pi/\Delta k_1$ of the SHG process is: $\chi^{(3)}_{\rm casc} \propto d^2_{zzz} L_{\rm coh,SH}$. The bigger $L_{\rm coh,SH}$, the stronger cascaded direct TH signal. For QPM nonlinear media for the effective cubic nonlinearity instead of (2) we will have [15]

$$\chi^{(3)}_{\rm casc,m} \propto \frac{d^{(2)}_{\rm SHG} d^{(2)}_{\rm SFM}}{\Delta k_{1,m}} = \frac{\left(\frac{2}{\pi m} d_{zzz}\right)\left(\frac{2}{\pi\, l} d_{zzz}\right)}{\Delta k_{1,m}}, \qquad (3)$$

where $\Delta k_{1,m} = k_2 - 2k_1 - 2\pi m/\Lambda_H$ as we denoted in (1).

The second-order nonlinearities responsible for the two separate cascading steps are $\frac{2}{\pi m} d_{zzz}$ for the SHG step and $\frac{2}{\pi\, l} d_{zzz}$ for the SFM step. We recall that m and l can be arbitrary integers with the restriction that $m + l = H$. This restriction is consequence of the phase matching condition for the direct THG process: $\Delta k_{3,H} = 0$.

In Figure 1b we plot $L_{\rm coh,SH} = \pi/|\Delta k_{1,1}|$ for $m = 1$ as a function of the wavelength for the first five QPM orders $H = 1 \ldots 5$. As can be seen from this figure for fundamental wavelength $\lambda_1 = 1.5$ μm 4$^{\rm th}$ order QPM gives the biggest SH coherence length and therefore we can expect the highest TH efficiency, exceeding with several orders of magnitude the TH efficiency for the other QPM orders. Similar calculations for $m = 2$ and $m = 3$ give order of magnitudes at smaller $L_{\rm coh,SH}$, that is why the essential contribution to the process of direct TH generation is expected from cascading steps with $m = 1$ and $l = 3$. Indeed, the coherence length for the second step SFM is defined as $L_{\rm coh,SF} = \pi/|\Delta k_{2,l}|$ (with $\Delta k_{2,l} = k_3 - k_1 - k_2 - 2\pi l/\Lambda_H$) and since $\Delta k_{2,l} = -\Delta k_{1,m}$, in condition of phase matched direct THG, the SFM process will have the same coherence length as SHG process and it will be maximized for $H = 4$ and $l = 3$. In contrast at wavelength $\lambda_1 = 1.064$ μm the value of $H = 5$ and $\{m = 1;\ l = 4\}$ gives the longest SH coherence length and the highest TH efficiency compared with the other QPM orders. It is interesting to notice at this point that the wavelengths for which $L_{\rm coh,SH} \Rightarrow \infty$ in Figure 1b correspond to the points of double phase matching discussed in the introduction as case (d).

The identification of the steps indices m and l that give the most effective contribution to the process of direct TH generation is important from the point of view of development of analytical model as it has been done in [15] and for understanding the physics of the process. In the numerical analysis described in the next section the contributions of all possible combinations $\{m;\ l\}$ are included.

Summarizing, in this part we have calculated the period of periodical modulation of the second-order nonlinearity and the QPM order for achieving efficient direct THG at fundamental wavelength $\lambda_1 = 1.5$ μm. The obtained for temperature $80°$C of the $LiNbO_3$ crystal period of periodical modulation of the second-order nonlinearity is $\Lambda_H = 18$ μm and the QPM order is $H = 4$. The value of the period of modulation of the second-order nonlinearity is needed for running the simulations described in the next section.

3 Slow Varying Envelope Equations

Starting from Maxwell's equations, assuming slowly varying amplitude approximation and neglecting the losses for all three waves as well as the possible effects due to group velocity dispersion, we end up with the coupled amplitude equations for fundamental, second and third harmonic radiation as follows:

$$\frac{dA_1}{dx} = -i\sigma_1 A_1^* A_2 \exp(-i\Delta k_1 x) - i\sigma_3 A_2^* A_3 \exp(-i\Delta k_2 x) \quad (4a)$$

$$\frac{dA_2}{dx} = -i\sigma_2 A_1^2 \exp(i\Delta k_1 x) - i\sigma_4 A_1^* A_3 \exp(-i\Delta k_2 x) \quad (4b)$$

$$\frac{dA_3}{dx} = -i\sigma_5 A_1 A_2 \exp(i\Delta k_2 x) - i\gamma A_1^3 \exp(i\Delta k_3 x), \quad (4c)$$

where $\sigma_{1,2} = 2\pi d_0/(\lambda_1 n_{1,2}) f(x)$ and $\sigma_j = (\omega_{j-2}/\omega_1)[2\pi d_0/\lambda_1 n_{j-2})] f(x)$ $(j = 3, 4, 5)$. d_0 is the bulk second-order nonlinearity and in our case $d_0 = d_{zzz}$. The third order coupling coefficient is $\gamma = 6\pi\chi^{(3)}_{zzzz}/(8\lambda_1 n_3)$. The periodical function $f(x)$ describes the periodical change of the sign of the nonlinearity with period, Λ_H and duty factor D and is defined as

$$\begin{cases} f(x) = 1 & 0 < z < \Lambda_H D \\ f(x) = -1 & \Lambda_H D < z < \Lambda_H \\ f(x + j\Lambda_H) = f(x) & j = 1, 2, 3, \ldots \end{cases} \quad (5)$$

In [15] we have shown analytically that for even orders QPM for direct THG the optimal duty factor is D=0.5 and for odd orders QPM the optimum is $D = 0.62$. The direct numerical solution of equations (4) confirmed that for even orders QPM for direct THG the optimal duty factor is $D = 0.5$ and for odd orders QPM the optimum is D=0.62. Therefore, as we have already chosen to work with $H = 4$ for direct THG at $\lambda = 1.5$ μm in $LiNbO_3$ we will start with a numerical solution of the system (4) for periodical change of the sign of the nonlinearity with duty factor $D = 0.5$. In what follows we are interested to solve several problems: a) to verify if periodical change of the sign of the second-order nonlinearity leads to periodical sign change of the effective cascaded cubic nonlinearity; b) what is the maximum efficiency that can be achieved with the process of direct THG and c) to verify the area of applicability of the analytical approach described in [15].

4 Results and Discussion

Intuitively one may expect that periodical poling of the second-order nonlinearity should not lead to phase matched THG since the effective cubic nonlinearity $\chi^{(3)}_{\text{casc}} \propto \dfrac{d_{zzz} d_{zzz}}{\Delta k_1}$ responsible for this process consists of the product of two

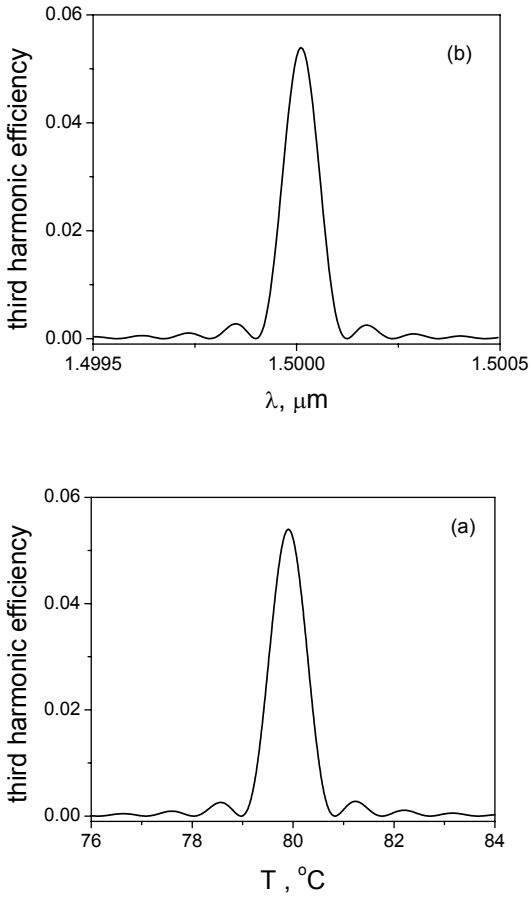

Figure 2. Calculated third harmonic conversion efficiency in 20 mm long periodically poled-$LiNbO_3$ with period $\Lambda_H = 18$ μm, duty factor $D = 0.5$, and normalized pump amplitude $\sigma A_{10} L = 12$ (intensity 30 MW/cm^2): (a) as a function of temperature for fixed input wavelength $\lambda_1 = 1.5$ μm; (b) as a function of the input wavelength for fixed temperature $T = 80^\circ$C.

third rank tensors $\langle d_{zzz} d_{zzz} \rangle$ that could be considered as fourth rank tensor and it should not change the sign when axis z changes its sign. So it is interesting to verify if the process of direct THG is phase matched and also if the efficiency curve has the typical for QPM processes step-like dependence. In Figure 2 the temperature and wavelength tuned phase matched curves are shown, while in Figure 3a the efficiency curve for sample length $L = 500\Lambda$ is presented. The existence of phase matching with respect to the wavelength and the temperature and the step-like behaviour of the efficiency curve clearly prove that the effective cubic nonlinearity changes periodically its sign with respect to axis x. Such

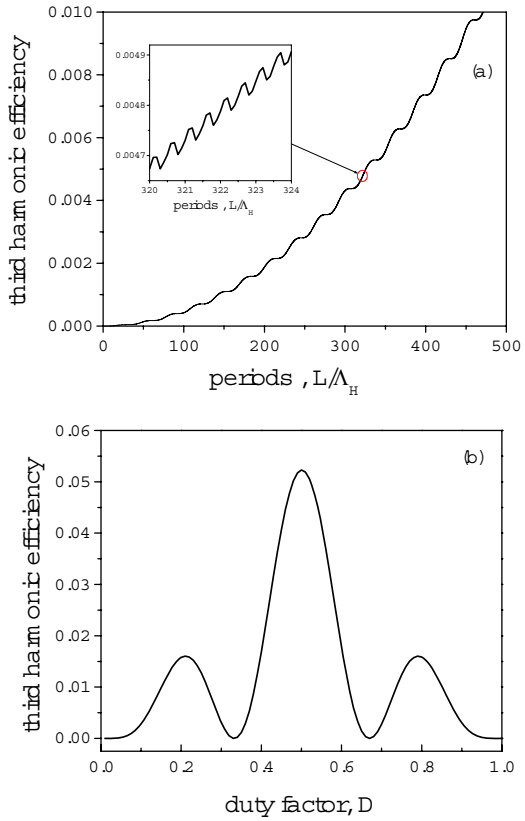

Figure 3. Calculated third harmonic conversion efficiency in periodically poled $LiNbO_3$ with $\Lambda_H = 18\,\mu$m: (a) as a function of the number of periods of nonlinearity sign reversal for fixed duty factor $D = 0.5$ and fundamental amplitude $\sigma A_{10} = 0.6$ mm^{-1}; (b) as a function of the duty factor D for sample length 20 mm and normalized pump amplitude $\sigma A_{10} L = 12$.

step-like behaviour is typical for efficiency curves for second-order processes [18]. Figure 3b demonstrates that for even order QPM vector ($H = 4$) used for phase matching of the direct TH generation the optimal duty factor is really $D = 0.5$ as predicted by the analytical model developed in [15].

Another very important question when considering the use of direct TH generation in quadratic crystals for actual applications is the maximum conversion efficiency that can be achieved. In Figure 4 we show the intensity conversion as a function of the input intensity calculated for 10 ps pulses in $LiNbO_3$ crystal. This numerical test demonstrates that efficiency of THG exceeding 25% can be achieved with single phase matching condition for quite moderate input intensity. The periodical character of the efficiency as a function of input intensity is

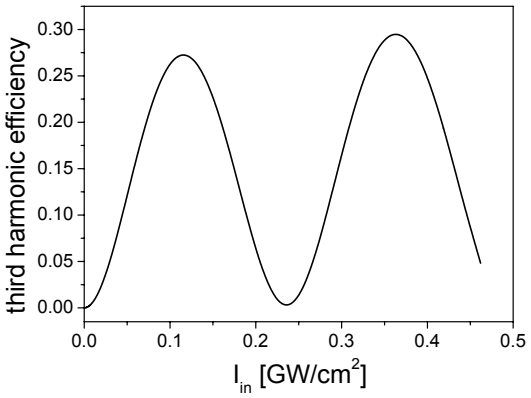

Figure 4. Calculated third harmonic conversion efficiency in 20 mm long periodically poled $LiNbO_3$ with period $\Lambda_H = 18$ μm and duty factor $D = 0.5$ as a function of the input intensity.

an indication of collection of nonlinear phase shift by the interacting wave that leads to destructive interference at certain power levels. In these calculations for efficiency we took the intensity ratio $\eta^{(\mathrm{TH})} = \dfrac{n_3|A_3|^2}{n_1|A_1|^2}$, that can be considered as energy conversion for rectangular shape in space and time. For Gaussian shape in space and time and not very big conversions (no depletion for the fundamental) the energy conversion is three times less than the intensity conversion [19].

Finally, we tested the theoretical model presented in [15]. In Figure 5 we compare the numerical solution of system (4) and the approximate analytical solution from [15] that is supposed to work in non depleted regime only

$$\eta_{\mathrm{nd}}^{(\mathrm{TH})} = \frac{n_3|A_3|^2}{n_1|A_1|^2} \approx \frac{n_3}{n_1}\left(\sigma_5\sigma_2 L^2|A_1|^3\right)^2 K_{D,H}\left|\frac{\sin(\Delta k_{3,H}L/2)}{(\Delta k_{3,H}L/2)}\right|^2, \quad (6)$$

where

$$K_{D,H} = \left|\sum_m \frac{g_m g_{H-m}}{\Delta k_{1,m}L}\right|^2 = \left|\frac{4}{\pi^2 L}\sum_{m\neq H}\frac{\sin(|m|D\pi)\sin(|H-m|D\pi)}{|m(H-m)|\Delta k_{1,m}}\right|^2. \quad (7)$$

Our aim is to verify the range of applicability of the analytical approach. Indeed, as can be seen from the figure there is a very good agreement between the numerical and analytical solutions Eqs. (6–7) at efficiencies below 5%, but significant discrepancy at higher input intensities. The analytical solution, however, can be corrected for the depletion by introducing here the **depletion correction factor** (DCF) applicable to TH generation process. Such kind DCF was proposed for

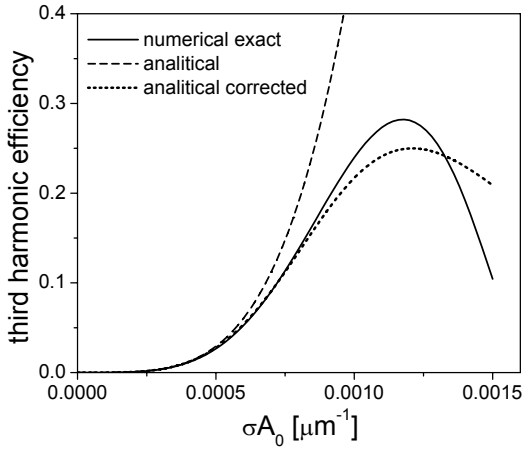

Figure 5. Third harmonic conversion efficiency in 20 mm long PP-$LiNbO_3$ with period $\Lambda_H = 18$ μm and duty factor $D = 0.5$ as a function of the normalized input amplitude as calculated with the numerical model (solid line), analytical model (dashed line) and corrected analytical model (dotted line).

correction of the process of SHG [20]. To derive the DCF for the case of TH generation we note that $P_3 = KP_0^3$ and $\eta_{\rm nd}^{(\rm TH)} = KP_0^2$. At high conversion into TH (noting that total power is conserved) the fundamental power responsible for TH generation has to be reduced by the amount of generated TH power and we will have $\eta_d^{(\rm TH)} = KP_1^2 = K(P_0 - P_3)^2$. Then we finally obtain the following relation between the nondepleted conversion $\eta_{\rm nd}^{(\rm TH)}$ and depleted conversion $\eta_{\rm d}^{(\rm TH)}$:

$$\eta_{\rm d}^{(\rm TH)} = \frac{\eta_{\rm nd}^{(\rm TH)}}{\left[1 + \eta_{\rm nd}^{(\rm TH)}\right]^2}. \tag{8}$$

It is clear from Figure 5 that the corrected analytical model with DCF given by Eq. (8) describes very well the process of THG in single periodically poled quadratic crystal up to 15% conversion efficiencies. To the best of our knowledge, such a correction factor that accounts for the depletion of third order processes has not been proposed in the literature. We are convinced that the presented here approach for description of direct TH generation in nonlinear media with periodical change of the sign of quadratic nonlinearity will be a useful design tool for researchers and engineers.

5 Conclusions

In conclusion, we theoretically investigated the process of direct third harmonic generation in QPM quadratic media. It was shown that in periodical structures with Duty factor 0.5 only even QPM orders can phase match the process of direct third harmonic generation . This is in big contrast to the result for the quasi phase matching of second-order processes when in periodical structures with Duty factor 0.5 only odd QPM orders can be used. In addition we show that the process of direct third harmonic generation can be very efficient. For the case of periodically poled $LiNbO_3$ and fundamental wavelength 1.5 μm we show that efficient TH can be generated for input intensities ~ 100 MW/cm^2 that corresponds to pulse energy less than 1 μJ !! if pulse duration is 10 ps and beam diameter 200 μm. Such parameters are easily accessible. In this way the direct third harmonic generation with requirement for **single** phase matching condition can be very attractive alternative to the schemes that require simultaneously phase matching of both cascading steps.

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