

# Direct third harmonic generation due to quadratic cascaded processes in periodically poled crystals

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## Abstract

We report the first, according to our knowledge, experiment for direct third harmonic (TH) generation in periodically poled quadratic crystals. TH radiation of a diode array pumped and acousto-optically Q-switched Nd:YVO<sub>4</sub> laser is generated in a 25 mm long periodically poled lithium niobate (PPLN) crystal of period 7.0 μm. A theoretical model is developed that explains the experimental results. It predicts that the efficient phase-matched direct TH generation in periodically poled structures with duty factor 0.5 requires even order quasi-phase matching. In contrast to the similar experiments in bulk birefringence crystals, where direct phase matched TH signal is the sum of the contribution of both intrinsic and cascaded cubic nonlinearities, in periodically poled quadratic crystals only cascaded cubic nonlinearity contributes to the direct phase-matched TH signal.

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## 1. Introduction

The quasi-phase matching (QPM) [1] has opened up a new area in nonlinear optics because of its several advantages over conventional birefringence phase matching in frequency conversion process, such as: phase matching in materials having high nonlinear optical coefficient but low or no birefringence, utilization of the highest element of nonlinear susceptibility tensor, suppression of walk-off, and great flexibility in the choice of the wavelengths involved in nonlinear optical (NLO) frequency mixing.

Third-harmonic generation (THG) is an important method to implement compact coherent light sources at

new and shorter wavelengths. Direct phase-matched third harmonic generation has been done utilizing the large birefringence of the nonlinear crystals, such as beta barium borate [2]; but such a process is not suitable, as most of the NLO materials do not have enough birefringence to support it. Moreover, one cannot utilize this method for THG of any desired wavelength. Two-step processes [3,4] involving successive second harmonic generation (SHG) and sum frequency generation (SFG) in two consecutive crystals, is another approach for efficient THG. Nowadays, this is one of the most conventional methods for THG; but it suffers from the problem of utilization of two crystals. On the other hand, to exploit the inherent advantages of QPM, THG by cascaded SHG and SFG in a single QPM crystal has been reported by several groups [5–7] and the review [8]. Quasi-phase matched direct THG using  $\chi^{(3)}$  has also

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been demonstrated in silica, but one requires multilayer structure of the materials to realize it [9]. Single crystal THG can be phase matched in four different situations (see, e.g. [10,11]): (i) when only the process of SHG is phase matched; (ii) when only the process of SFG is phase matched; (iii) when both processes are not phase matched, but the mismatches for both the processes of SHG and SFG are equal in magnitude but opposite in sign; and (iv) when the two processes of SHG and SFG are phase matched simultaneously. The case (iii), that we call direct THG process has phase matching condition  $\Delta k_3 = k_3 - 3k_1 \rightarrow 0$ , for bulk crystals or  $\Delta k_3 = k_3 - 3k_1 - G_m \rightarrow 0$  for QPM media ( $G_m$  is one of the reciprocal vectors). This case has been investigated only in bulk homogeneous crystals [12–14] with utilization of birefringence phase matching and efficiencies up to 6% have been achieved [12] in BBO crystal. The TH signal in bulk quadratic crystals for the case (iii) direct phase matching is result of simultaneous contribution of direct and cascaded processes. To the best of our knowledge, the direct THG with phase matching of case (iii) has never been investigated experimentally in periodically poled crystals and this is one of the goals of this work.

During periodic polling the dipoles of the ferroelectric crystal reorient according to the external electric field. This orientation of the dipole yields a periodic variation in second order nonlinear coefficient,  $\chi^{(2)}$  of the material. Grating wave vectors thus obtained, serves for achieving phase matching by compensation of the bulk mismatch [15]. But periodic poling can not modulate intrinsic  $\chi^{(3)}$  in anisotropic material because during this process each suffix of optical susceptibility tensor transforms like a vector. The second order susceptibility has 3 suffixes, as such it changes sign in alternate domains of periodically poled crystal, whereas the bulk third order susceptibility  $\chi_{\text{dir}}^{(3)}$  does not change its sign in alternate domains because of even number of suffixes. However there are some report that reorientation of the dipoles also induce some changes in the value of the third order nonlinear coefficient,  $\chi^{(3)}$  [16–18].

Here we show that in crystals with periodical change of the sign of the quadratic nonlinearity, the effective (cascaded) cubic nonlinearity also varies periodically with respect to the propagation coordinate and we propose this behavior of the cascaded cubic nonlinearity to be used for direct third harmonic generation in a periodically poled ferroelectric. In an initial experiment we demonstrate direct third harmonic generation in periodically poled lithium niobate (PPLN), where the mismatch between the phase velocities of the fundamental and third harmonic waves is compensated by the effective fifth order reciprocal vector associated with the periodical modulation of cascaded cubic nonlinearity. To our knowledge, there is no report of this type of quasi-phase-matching, although technique of periodical poling of ferroelectrics has very extensively studied and utilized in the last decade. In a PPLN with grating period of 7.0  $\mu\text{m}$ , we achieve 355 nm radiation by direct third harmonic generation of 1064 nm radiation available from a laboratory developed acousto-optic

Q-switched Nd:YVO<sub>4</sub> laser system [19]. The conversion, estimated to be in the order of  $10^{-4}\%$  corresponding to the input intensity of 0.4 MW/cm<sup>2</sup>.

## 2. Theory

In a noncentrosymmetric crystal, the cascading of second order processes can mimic practically all third order processes such as of nonlinear phase shift, four wave mixing, third harmonic generation, self-focusing and defocusing etc. The nonlinear polarization in such media is represented as

$$\tilde{P}_{\text{NL}} = \tilde{P}_{\text{NL}}^{(2)} + \tilde{P}_{\text{NL,dir}}^{(3)} = \chi^{(2)} \tilde{E}_\omega \tilde{E}_\omega + \chi_{\text{dir}}^{(3)} \tilde{E}_\omega \tilde{E}_\omega \tilde{E}_\omega + \dots \quad (1)$$

where  $\chi^{(2)}$  and  $\chi_{\text{dir}}^{(3)}$  are the second and third order nonlinear coefficients of the material and  $\tilde{E}_\omega$  is electric field of the fundamental beam having frequency  $\omega$ . The term  $\tilde{P}_{\text{NL,dir}}^{(3)}$  is responsible for direct third harmonic generation. To account for cascading of second order effects, an additional cubic term  $\tilde{P}_{\text{NL,casc}}^{(3)} = \chi_{\text{casc}}^{(3)} \tilde{E}_\omega \tilde{E}_\omega \tilde{E}_\omega$  can be symbolically introduced. This cascaded third order nonlinear coefficient is proportional to the product  $\chi^{(2)} \chi^{(2)}$  [20], which is responsible for third harmonic generation and other third order effects. Here we consider the specific case when all the fundamental, the second harmonic and the third harmonic waves are polarized along  $z$ -axis while propagation direction is along  $x$ -axis of the crystal (Fig. 1).

Inserting the nonlinear polarization (Eq. (1)) in Maxwell's equations and assuming slowly varying amplitude approximation as well as considering uniform periodically poled structure with period,  $\Lambda$ , and duty factor,  $D$ , we end up with the coupled amplitude equations for fundamental, second and third harmonic radiation as follows:

$$\frac{dA_1}{dz} = -i\sigma_1 A_1^* A_2 \sum g_m \exp(-i\Delta k_{1,m} z) - i\sigma_3 A_2^* A_3 \sum g_m \exp(-i\Delta k_{2,m} z) \quad (2a)$$

$$\frac{dA_2}{dz} = -i\sigma_2 A_1^2 \sum g_m \exp(i\Delta k_{1,m} z) - i\sigma_4 A_1^* A_3 \sum g_m \exp(-i\Delta k_{2,m} z) \quad (2b)$$

$$\frac{dA_3}{dz} = -i\sigma_5 A_1 A_2 \sum g_m \exp(i\Delta k_{2,m} z) - i\gamma A_1^3 \exp(i\Delta k_3 z) \quad (2c)$$

where  $\Delta k_{1,m} = k_2 - 2k_1 - G_m$ ,  $\Delta k_{2,m} = k_3 - k_2 - k_1 - G_m$ ,  $\Delta k_3 = k_3 - 3k_1$ ,  $\sigma_{1,2} = \frac{2\pi d_o}{\lambda_1 n_{1,2}}$  and  $\sigma_j = \frac{(\omega_j - 2)}{\omega_1} \frac{2\pi d_o}{\lambda_1 n_{j-2}}$  ( $j = 3, 4, 5$ ).  $d_o$  is the bulk second order nonlinearity and in our case  $d_o = d_{zzz}$ . Third order coupling coefficient,  $\gamma = \frac{6\pi\chi_{\text{casc}}^{(3)}}{8\lambda_1 n_3}$ . The

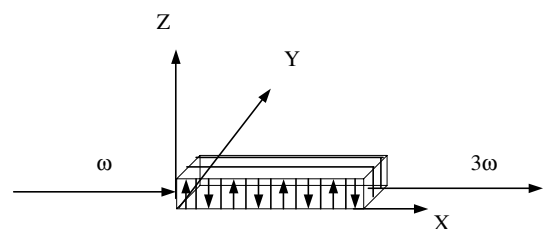


Fig. 1. Schematic of the THG process in periodically poled crystal.

reciprocal vectors are  $G_m = \frac{2\pi}{\Lambda}m$  and the coefficients,  $g_m = \frac{2}{\pi|m|} \sin(|m|D\pi)$ . The summation reflects the contribution of all high order QPM terms and it is for all  $m$  except  $m = 0$ . There is no summation in the last term of Eq. (2c) because the volume cubic nonlinearity is not affected by the periodical poling. A solution of these equations (see, e.g., [10,11]) gives maximum for THG in three different single phase matching conditions, when (1)  $\Delta k_{1,m} \rightarrow 0$ ; (2)  $\Delta k_{2,m} \rightarrow 0$ ; (3)  $\Delta k_{3,m,l} = \Delta k_{1,m} + \Delta k_{2,l} \rightarrow 0$ . We solve Eq. (2) for the last condition with  $\Delta k_{3,m,l} \rightarrow 0$ .

We consider situation of phase matched third harmonic generation with efficiency small enough to be in non-depleted regime for the fundamental and second harmonic wave. More precisely we consider the case when both  $\Delta k_{1,m}L \gg 1$  and  $\Delta k_{2,m}L \gg 1$  but for certain combination of the integers  $m$  and  $l$ ,  $\Delta k_{1,m}L + \Delta k_{2,l}L \ll 1$ . Thus we can approximate the RHS of Eq. (2a) equal to zero and neglect second term of RHS of Eq. (2b). Then Eq. (2) is simplified to

$$\frac{dA_1}{dz} = 0 \quad (3a)$$

$$\frac{dA_2}{dz} = -i\sigma_2 A_1^2 \sum g_m \exp(i\Delta k_{1,m}z) \quad (3b)$$

$$\frac{dA_3}{dz} = -i\sigma_5 A_1 A_2 \sum g_m \exp(i\Delta k_{2,m}z) - i\gamma A_1^3 \exp(i\Delta k_3 z) \quad (3c)$$

To find the TH amplitude we use the solution of Eq. (3b) for second harmonic amplitude,  $A_2(z) = \sigma_2 A_1^2 \sum g_m \frac{[1 - \exp(i\Delta k_{1,m}z)]}{\Delta k_{1,m}}$ . Substituting this in Eq. (3c) we have

$$\frac{dA_3}{dz} = -i\sigma_5 \sigma_2 \left[ \sum_{m,l} \frac{g_m g_l}{\Delta k_{1,m}} e^{i\Delta k_{2,l}z} - \sum_{m,l} \frac{g_m g_l}{\Delta k_{1,m}} e^{i(\Delta k_{1,m} + \Delta k_{2,l})z} + \frac{\gamma}{\sigma_5 \sigma_2} e^{i\Delta k_3 z} \right] A_1^3 \quad (4)$$

The first and third terms in Eq. (4) have small contribution to efficiency of the THG process, since they give non-phase matched TH signal ( $\Delta k_3 L \gg 1$  and all  $\Delta k_{2,l}L \gg 1$ ) and can be neglected. The only term that contributes to phase matched TH generation is the middle one, because we pointed out before that for some pairs of  $m$  and  $l$ , the mismatch that controls the direct TH generation process,  $\Delta k_{1,m}L + \Delta k_{2,l}L \ll 1$ . Then Eq. (4) can be rewritten in a way to illustrate that the effective cascaded cubic nonlinearity is a periodic function of  $z$ .

$$\frac{dA_3}{dz} = i \left\{ \sigma_5 \sigma_2 \sum_{m,l} \frac{g_m g_l}{\Delta k_{1,m}} \exp[-i(G_m + G_l)z] \right\} \exp(i\Delta k_3 z) A_1^3 \quad (5)$$

The expression in the curled brackets is the cascaded nonlinear cubic coupling coefficient  $\gamma_{\text{casc}} = 6\pi\chi_{\text{casc}}^{(3)}/(8\lambda_1 n_3)$  with

$$\chi_{\text{casc}}^{(3)} = 16 \frac{\pi d_{zzz}^2}{\lambda_1 n_2} \sum_{m,l} \frac{g_m g_l}{\Delta k_{1,m}} \exp \left[ -i \frac{2\pi}{\Lambda} (m+l)z \right] \quad (6)$$

It is clear from the Fourier transform presentation in Eqs. (4) and (6) that  $\gamma_{\text{casc}}$  and  $\chi_{\text{casc}}^{(3)}$  are periodic functions of  $z$ .

This periodic variation with  $z$  of the cascaded cubic nonlinearity compensates the bulk mismatch  $\Delta k_3$ , that appears in Eq. (5). Integrating Eq. (5) and neglecting the non-phase matched terms we obtain for the square of the normalized TH amplitude:

$$|A_3|^2 \approx (\sigma_5 \sigma_2 L^2 |A_1|^3)^2 K_{D,H} \left| \frac{\sin(\Delta k_{3,H}L/2)}{(\Delta k_{3,H}L/2)} \right|^2 \quad (7)$$

In Eq. (7)  $\Delta k_{3,H} = \Delta k_{1,m} + \Delta k_{2,l} = k_3 - 3k_1 - \frac{2\pi}{\Lambda}H$ , where  $H = m + l$  is a design constant integer for which  $\Delta k_{3,H} \approx 0$ . In fact  $H$  is the order of the QPM reciprocal vector used to phase match the direct TH process.

The efficiency of the process depends on the dimensionless design coefficient

$$K_{D,H} = \left| \sum_m \frac{g_m g_{H-m}}{\Delta k_{1,m}L} \right|^2 = \left| \frac{4}{\pi^2 L} \sum_{m \neq H} \frac{\sin(|m|D\pi) \sin(|H-m|D\pi)}{|m(H-m)|\Delta k_{1,m}} \right|^2 \quad (8)$$

The dependence of the constant,  $K_{D,H}$  on the filling factor ( $D$ ) for different orders ( $H$ ) is shown in Fig. 2. Analyzing Eq. (8) for THG of  $\lambda = 1.064 \mu\text{m}$  in LiNbO<sub>3</sub> of grating period  $7 \mu\text{m}$  ( $H = 5$ ), we reach to the conclusion that main contribution of the terms in the sum (8) is from the term with  $m = 1$ , since  $\Delta k_{1,m=1} = -\Delta k_{2,l=4} = 0.033 \mu\text{m}^{-1}$  is much smaller than  $|\Delta k_{1,m}|$  for  $m = 2, 3, 4$  and  $|\Delta k_{2,l}|$  for  $l = 1, 2, 3$ . If we consider only the term  $m = 1$  we have  $K_{D,H}(\text{max}) \approx 1/(0.033\pi^2 L)^2 \approx 1.5 \times 10^{-8}$ , where as considering all  $m$  terms  $K_{D,H}(\text{max}) \approx 1.2 \times 10^{-8}$  as evident from Fig. 2 (curve for  $H = 5$ ) for appropriate value of  $D$ .

It is obvious from Eq. (8) that for QPM structures with duty factor  $D = 0.5$ , the coefficient  $K_{0.5,H} = 0$ , if  $H$  is odd integer and hence there will be no phase-matched THG signal. To obtain efficient THG in a periodically poled structure of duty cycle 0.5, both  $m$  and  $l$  should be odd integer. Consequently  $H$ , the order of the QPM vector that compensates the bulk mismatch  $\Delta k_3$  for direct THG has to

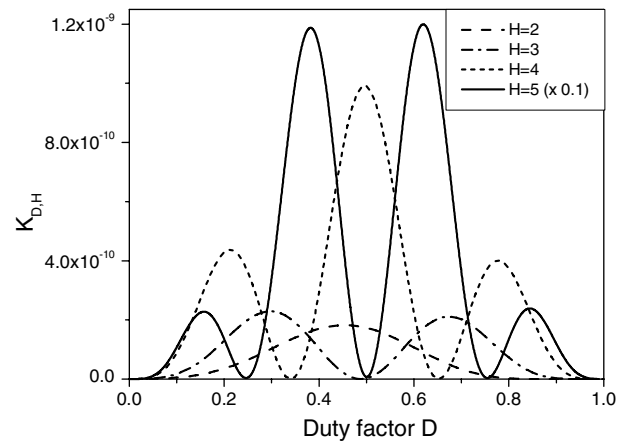


Fig. 2. Dependence of the constant  $K_{D,H}$  on the duty factor  $D$  for different orders  $H$  (respectively, different poling period  $\Lambda = 1.4 \text{ H}$ ), calculated using mismatches for LiNbO<sub>3</sub> sample:  $k_3 - 3k_1 = 4.488 \mu\text{m}^{-1}$ ;  $k_2 - 2k_1 = 0.931 \mu\text{m}^{-1}$ ;  $k_3 - k_2 - k_1 = 3.557 \mu\text{m}^{-1}$ . Sample length  $L = 2.5 \text{ cm}$ .

be even order. We see in Fig. 2, that the maximum value for  $K_{D,H}$  is for  $H = 5$  with duty cycle  $\neq 0.5$ .

In contrast to the direct phase-matched THG in bulk birefringence crystals, where TH signal is governed by both direct and cascaded cubic nonlinearities [12–14], the direct phase matched TH generation in periodically poled crystals, as we proved above, is only due to second order cascaded processes.

Fig. 3 shows the variation of THG conversion efficiency of PPLN of length 25 mm with temperature for  $\lambda_1 = 1064$  nm,  $\Lambda = 7.0$   $\mu\text{m}$ ,  $D = 0.6$  and  $m + l = 5$ . For the refractive index values at different wavelengths and temperatures, we use the Sellmeier equations given in Ref. [21]. The phase matching temperature is calculated to be 58.6 °C. Similar to the analysis for the temperature bandwidth of SHG [15], the temperature bandwidth for third harmonic generation can be described as

$$dT = \frac{5.57\lambda_1}{6\pi L} \left| \frac{\partial \Delta n}{\partial T} + \alpha \Delta n \right|^{-1} \quad (9)$$

where  $\Delta n = n_3^e - n_1^e$  and  $\alpha$  is the linear thermal expansion co-efficient of LiNbO<sub>3</sub>. The temperature bandwidth is calculated to be 0.39 °C.

### 3. Experiment and result

The schematic diagram of the experimental setup is shown in Fig. 4. The pump beam used for this experiment is an acousto-optically Q-switched Nd:YVO<sub>4</sub> laser [19] of wavelength 1064 nm. The laser emits Q-switched pulses of width 120 ns at repetition rate 45 kHz with average output power of 1.34 W. The collimated beam of 400  $\mu\text{m}$  diameter is passed through the congruent melt grown PPLN crystal with QPM grating period of 7  $\mu\text{m}$  (HC Photonics, USA) having length 25 mm and thickness 0.5 mm placed inside an oven. Two KG1 filters F<sub>1</sub> and F<sub>2</sub> (each having transmittance 10<sup>-4</sup>% and 90% at 1064 nm and 355 nm, respectively) are placed in front of the monochromator (77250, Oriel, USA). The generated third harmonic signal (355 nm) is detected using a photo-multiplier tube (PMT) (IP21, EMI Electronics Ltd., UK) having quantum efficiency nearly 90% at 355 nm. The temperature of the oven, measured using the Cu-Constantine thermocouple, can be varied from room temperature to 400 °C with sensitivity of 0.25 °C. The experimental variation of the generated TH signal as a function of the temperature is plotted in Fig. 5. The maximum THG signal is obtained

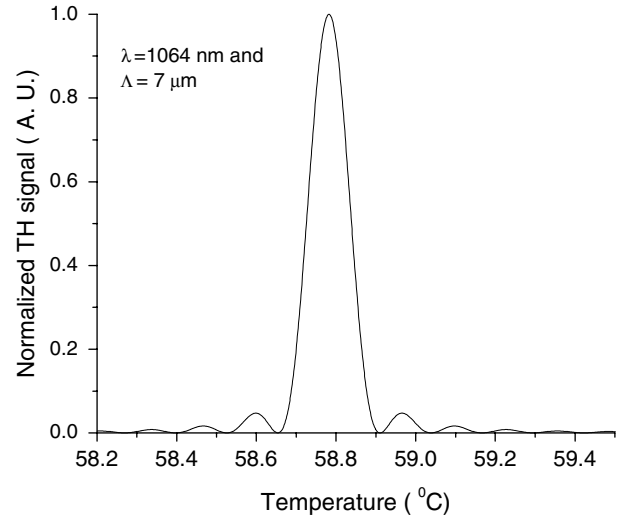


Fig. 3. Calculated normalized third harmonic intensity in PPLN with  $D = 0.6$  and fifth order QPM ( $H = 5$ ) as a function of temperature.

at 57.5 °C with temperature bandwidth nearly 1 °C. The calculated value of the temperature (58.6 °C) corresponding to maximum TH signal for  $H = 5$ , is very close to the experimentally observed value. The fact, phase matched third harmonic is recorded with fifth order QPM, proves that the duty factor is different from 0.5. We carefully inspected the sample used in the experiment under high-resolution polarization microscope. The period was quite constant through the sample, but the duty factor was varying from 0.5 to 0.67. This is illustrated on the microphotograph (Fig. 6) of  $220 \times 300$   $\mu\text{m}^2$  area of the periodically poled structure, where we notice the change of the duty factor in the range of 0.54–0.67. The averaged value of the real duty factor different from the 0.5 explains the observation of direct THG with fifth order QPM in our experiment with PPLN sample. The several times broader experimental phase matching curve compared to the theoretical one can be explained by varying duty factor that reduces the effective length of the sample. The efficiency of the generated third harmonic signal can be estimated on the basis of the developed model. In our experimental conditions, with  $d_{zzz} = 30$  pm/V for LiNbO<sub>3</sub>, we derive  $\eta_{\text{THG}} = 7.5 \times 10^{-6} I^2$  (where  $I$  is intensity in MW/cm<sup>2</sup>) from Eq. (7). Substituting the intensity of our fundamental radiation  $I \approx 0.4$  MW/cm<sup>2</sup> we obtain the THG efficiency  $\eta_{\text{THG}} = 1.2 \times 10^{-4}\%$ .

The input power of radiation at 1064 nm is measured to be 1.34 W by power meter (30-A-SH & Nova, Ophir Optronics Ltd., Israel). But the generated TH radiation is

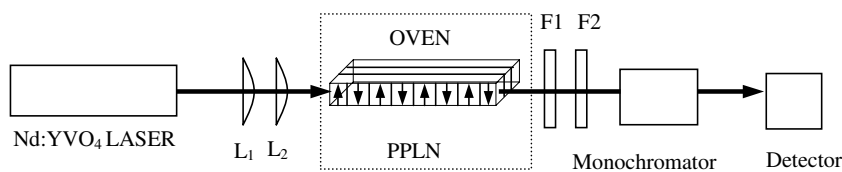


Fig. 4. Schematic of the experimental setup for THG process in PPLN; L<sub>1</sub> and L<sub>2</sub> lenses; F<sub>1</sub> and F<sub>2</sub> colour glass filter (KG1); the temperature controlled oven is shown in the dotted box.

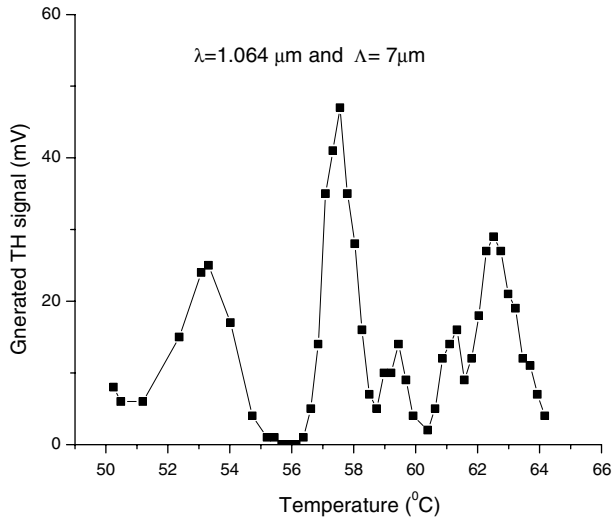


Fig. 5. Variation of generated third harmonic intensity with temperature as measured by PMT.

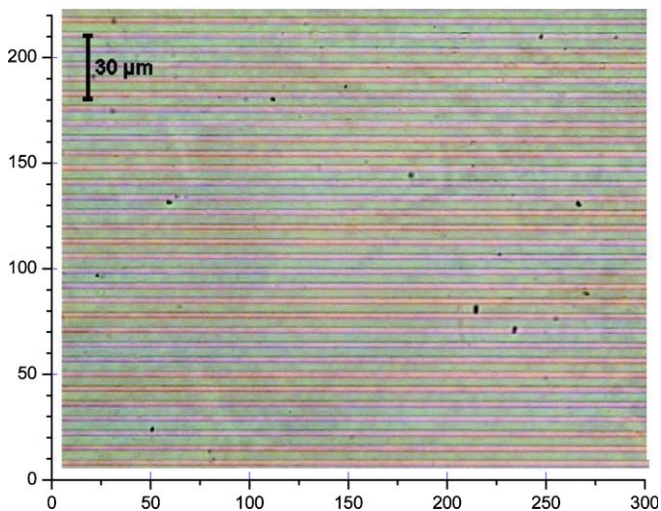


Fig. 6. Microphotograph showing the domain boundaries of the PPLN crystal.

too low to be measured by the power meter. To increase the dynamic range of measurement, we calibrated the photo multiplier tube with the power meter at the second harmonic radiation at 532 nm. The necessary quantum efficiency correction for 355 nm is taken into consideration in the calculation of experimental conversion efficiency. An average power of 48 mW at 532 nm as measured by power meter corresponds to 1.2 V of peak voltage as measured by PMT with neutral density filter of  $D = 3$  for pulses of repetition rate 45 kHz and pulse width 120 ns. The relative responsivity of the used PMT at 532 nm and 355 nm are 65% and 100%, respectively. However the power meter has same responsivity at these wavelengths. With these calibration data the observed PMT peak voltage of 47 mV at 355 nm corresponds to 1.22  $\mu$ W of average power. This gives the experimental efficiency of  $0.9 \times 10^{-4}\%$ .

In Ref. [6], authors have used pico second pulse of intensity 36 MW/cm<sup>2</sup> for their experiment in a partly poled and partly bulk crystal and have got efficiency of 3%. From the formula given above it is clear that we can have an efficiency of 1% with this intensity level. But unlike our case, in the above work, both the SHG and SFG processes are phase matched. In Ref. [13], a very high input intensity of 28 GW/cm<sup>2</sup> was used to achieve conversion efficiency of 2.4% in bulk KTP crystal in phase matched THG process due to intrinsic  $\chi^{(3)}$ . It is expected that the conversion efficiency with such input, will be very large in our case.

It is clear that the optimization of the duty cycle, in a sense to be constant, equal to the optimal value and using more powerful laser source, would allow obtaining efficiencies in third harmonic of the order of 10–30%.

#### 4. Conclusion

In summary we have demonstrated experimentally direct third harmonic generation in periodically poled LiNbO<sub>3</sub>. The estimations show that using picosecond and femtosecond laser sources with intensity several hundreds MW/cm<sup>2</sup> one may expect very efficient generation of direct third harmonic in quadratic periodically poled crystals.

The approach developed here can be extended to describe other direct nonlinear optical processes based on cascading of lower order processes in periodically poled nonlinear crystals.

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