

# Type I quasi-phase-matched blue second harmonic generation with different polarizations in periodically poled LiNbO<sub>3</sub>

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## Abstract

First-order type I quasi-phase-matched (QPM)  $E_Y^\omega + E_Y^\omega \rightarrow E_Z^{2\omega}$  blue second-harmonic generation was demonstrated in periodically poled LiNbO<sub>3</sub> with period of 14.5 μm using  $d_{31}$ . 52 μJ of harmonic blue light at 0.473 μm was generated pumped by 114 μJ 35 ps pulse laser at 0.946 μm at 150 °C with a conversion efficiency of 45.6%. The average conversion efficiencies of 41.3% and 19% were also obtained at 150 °C, respectively, in the conventional first- and third-order QPM  $E_Z^\omega + E_Z^\omega \rightarrow E_Z^{2\omega}$  blue second-harmonic generation at 0.473 μm. The temperature acceptance bandwidths of 20 mm length periodically poled LiNbO<sub>3</sub> with first-order grating periods of 14.5 and 4.5 μm are 2.0 and 0.9 °C, respectively. The larger acceptance bandwidths and grating period for  $E_Y^\omega + E_Y^\omega \rightarrow E_Z^{2\omega}$  than those for  $E_Z^\omega + E_Z^\omega \rightarrow E_Z^{2\omega}$  enhance the frequency conversion efficiency, which shows the polarization dependence of quasi-phase matching.

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## 1. Introduction

Second-harmonic generation (SHG) in nonlinear optics materials is an effective approach to obtain blue, green, or UV coherent light sources. In the recent decade, Quasi-phase matching (QPM) was used to realize high conversion efficiency SHG in periodic poling of ferroelectric crystals [1–7]. Periodically poled lithium niobate (PPLN) is widely used owing to its wide transparency range down to 0.35 μm, its high effective nonlinear coefficient, and excellent mechanical stability [1–6]. In the application of PPLN, people always focus on how to improve the frequency conversion efficiency. In spite of the large pump light power, the high

uniformity of the domain conversion grating and large acceptance bandwidths are very useful for obtaining high conversion efficiency.

Recently, in *z*-cut periodically poled LiNbO<sub>3</sub> crystal with a QPM period of 6.5 μm [8], we demonstrated first-order QPM  $E_Z^\omega + E_Z^\omega \rightarrow E_Z^{2\omega}$  (e + e → e) (which is called QPM(e) hereinafter) SHG at 0.532 μm green light with its largest nonlinear coefficient  $d_{33}$ . However, 0.532 μm CW green light was obtained at 5 °C, which corresponds to 1.5% cm<sup>-1</sup> W<sup>-1</sup> of normalized conversion efficiency, instead of 190 °C as we designed.

The main reason of low conversion efficiency and QPM temperature shift in our experiment is due to the departure from ideal QPM in periodicity, i.e. the fabrication errors of grating period. In the short wavelength region, it is particularly difficult to maintain the uniformity of the QPM grating. To alleviate those fabrication constraints, the larger period of higher-order

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QPM(e) is used. However, the effective nonlinear coefficient is reduced by a factor of  $m$ , where  $m$  is an odd number of the QPM order (for a 50% duty-cycle grating). So the conversion efficiency cannot be improved effectively. However, by advanced technique, some papers reported fabrication of uniform first-order QPM grating in the short-wavelength region [3,4,6,7], and high conversion efficiency of  $7.1\% \text{ cm}^{-1} \text{ W}^{-1}$  had been realized [3]. So the fabrication constraints are not the main difficulty. Another important resolution to improve the nonlinear performance of the QPM device is to enhance acceptance bandwidths of the QPM device.

In Ref. [9], we suggested a new QPM method-type I QPM  $E_Y^\omega + E_Y^\omega \rightarrow E_Z^{2\omega}(\text{o} + \text{o} \rightarrow \text{e})$  (which is called QPM(o) hereinafter), with the input waves as ordinary wave instead of extraordinary wave. The larger acceptance bandwidths and grating periods for QPM(o) SHG process compared with QPM(e) SHG lower the fabrication constraints especially in the short-wavelength SHG region, and also enhance the frequency conversion efficiency compared to the higher order in QPM(e) SHG.

In this letter we report QPM(o) blue SHG in PPLN with the length of 20-mm and with the first-order period of  $14.5 \mu\text{m}$  using nonlinear coefficient  $d_{31}$ . In a single-pass configuration,  $52 \mu\text{J}$  of harmonic blue light at  $0.473 \mu\text{m}$  was generated from an input of  $114 \mu\text{J}$  at  $150^\circ\text{C}$  with the average conversion efficiency of 45.6% in our experiment.

## 2. Theoretical comparison between QPM(o) and QPM(e)

For the QPM(o) SHG process, from Eq. (1), we can see that the nonlinear coefficient  $d_{33}$  is utilized in QPM(e) SHG and  $d_{31}$  is utilized in QPM(o) SHG in bulk  $z$ -cut PPLN, respectively [9]:

$$P_z(2\omega, t) = d_{31}E_y^2(\omega, t) + d_{33}E_z^2(\omega, t). \quad (1)$$

For the QPM collinear interaction in PPLN, the conversion efficiency  $\eta$  of QPM SHG is proportional to  $(d_Q)^2$  and  $\sin^2(\Delta\kappa_Q L/2)$ , as shown in Eq. (2):

$$\eta \propto (d_Q)^2 \sin^2(\Delta\kappa_Q L/2), \quad (2)$$

where  $\sin^2(\Delta\kappa_Q L/2) \equiv \sin^2(\Delta\kappa_Q L/2)/(\Delta\kappa_Q L/2)^2$ ,

$$\text{and } d_Q = d_{\text{eff}} \frac{2}{\pi m}, \quad (3)$$

where  $d_Q$  is the amplitude of the periodically modulated nonlinear coefficient, and  $d_{\text{eff}}$  is the effective nonlinear coefficient of SHG in single-domain bulk material;  $\Delta\kappa_Q$  is the total wave vector mismatch.  $L$  is the total length of PPLN.

The QPM grating period is given by  $\Lambda = 1/2 \times \lambda_1/(n_1 - n_2)$ . For QPM(e) SHG,  $n_1$  and  $n_2$  are the indices

of fundamental extraordinary light and second harmonic extraordinary light. For QPM(o) SHG,  $n_1$  and  $n_2$  represent the indices of fundamental ordinary light and second harmonic extraordinary light. In Ref. [9], we compared the grating periods of the two different order QPM SHG with different polarization at the same temperature. It is shown that the domain period for the first-order QPM(o) SHG is larger than that for the third-order QPM(e) SHG, which facilitates the fabrication of QPM devices, decreases the possibility of phase mismatching and improves the conversion efficiency partially.

From Eq. (3), the corresponding effective nonlinear coefficients are  $d_{Q3}(\text{e}) = (2d_{33}/\pi/3)^2$  and  $d_{Q1}(\text{o}) = (2d_{31}/\pi)^2$  for third-order QPM(e) and first-order QPM(o), respectively. The theoretical ratio of the conversion efficiencies of the two processes is  $[d_{Q3}(\text{e})/d_{Q1}(\text{o})]^2 = 1.26$  (assuming  $d_{33} = 27 \text{ pm/v}$  and  $d_{31} = 8 \text{ pm/v}$ ). However, the larger grating period and full-width at half-maximum (FWHM) acceptance bandwidth of QPM(o) SHG ensures better uniformity of domain inversion and higher quality of the QPM SHG performance, which makes higher conversion efficiency possible [9,10].

## 3. Experiment

An ideal uniform periodic domain structure in PPLN crystal is very important to obtain high frequency conversion efficiency in nonlinear interaction. In our experiment, we used  $z$ -cut, standard optical-grade congruent LiNbO<sub>3</sub> samples with 20-mm length, 10-mm width and 0.5-mm thickness to fabricate PPLN with grating periods of 4.5, 13.5 and  $14.5 \mu\text{m}$ , respectively. The  $+z$  face of LiNbO<sub>3</sub> flake was lithographically patterned with a periodic array of sputtered nickel stripes with the grating vector parallel to the crystallographic  $x$ -axis.  $1\text{-}\mu\text{m}$ -thick photoresist was patterned on the  $+z$  face of the sample by the standard lithography technique and then the sample was coated by a  $0.1\text{-}\mu\text{m}$ -thick nickel film forming periodic array nickel electrodes. The connection between the positive electrode and the high voltage source is made by contacting with liquid electrolyte lithium chloride, which was confined by an O ring. The negative electrode, a metal plate connecting the ground, is contacted with the  $-z$  surface directly.  $12 \text{ kV}$  high pulsed voltage was applied on the LiNbO<sub>3</sub> sample. After poling, the metal was etched away by vitriol and the insulator was washed by acetone. To check the uniformity of the domain pattern, we etched the sample by hydrofluoric acid and nitric acid (1:2). Both the faces of the samples showed good domain pattern uniformity, which was observed by a BX50WI Olympus Fixed Stage Upright Microscope as shown in Fig. 1a–c. The duty cycles for grating

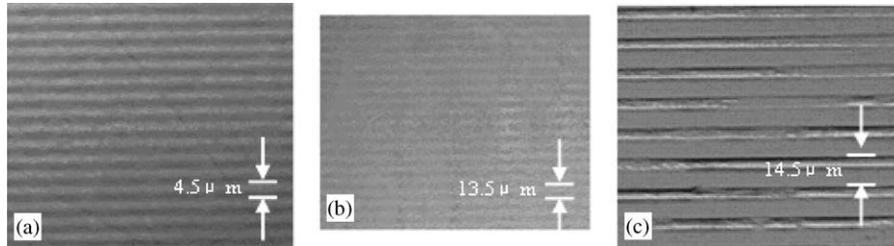


Fig. 1. (a), (b) and (c) are the etching figures for  $-z$  face, grating periods of 4.5, 13.5 and 14.5  $\mu\text{m}$  show the duty cycles of about 45%, 50% and 50%, respectively.

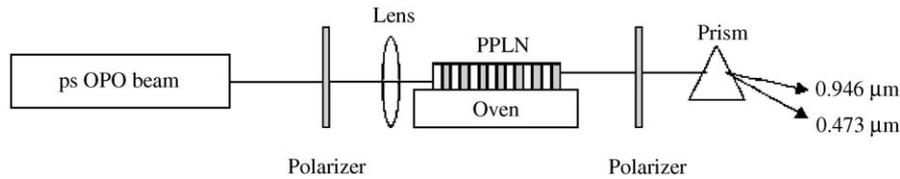


Fig. 2. The measuring setup for the blue light QPM SHG experiment.

periods of 13.5 and 14.5  $\mu\text{m}$  are all close to 50% in which the conversion efficiency is highest by theoretical prediction.

The schematic diagram of the measuring setup of QPM blue SHG conversion efficiency in PPLN with 4.5, 13.5 and 14.5  $\mu\text{m}$  periods is shown in Fig. 2. We used an optical parametric oscillator (OPO) with a  $\beta$ -barium borate crystal pumped by the frequency tripled beam (the wavelength is 0.355  $\mu\text{m}$ ) of a Q-switched Nd:YAG laser, which provided 35 ps pulses at a repetition rate of 10 Hz. In QPM(o) blue SHG, the OPO beam was polarized along the  $y$ -axis, loosely focused on the PPLN sample with the beam waist diameter of about 40  $\mu\text{m}$ . The end faces of samples were all polished but without anti-reflection coating. The PPLN was put in an oven, in which the temperature can be controlled with an accuracy of 0.1  $^{\circ}\text{C}$ . The other polarizer was used to analyze the polarization of the output blue SHG light. We found the output blue beam was polarized along the  $z$ -axis, which was extraordinary light. At 150  $^{\circ}\text{C}$ , with the end coupling, corresponding to the highest average conversion efficiency of 45.6%, we obtained 52  $\mu\text{J}$  of harmonic blue light pumped by 114  $\mu\text{J}$  of 0.946  $\mu\text{m}$  input light.

Using the setup shown in Fig. 2, we also demonstrated the QPM(e) blue SHG in PPLN with the first-order grating period of 4.5  $\mu\text{m}$  and the third order of 13.5  $\mu\text{m}$  from the input beam of 0.946  $\mu\text{m}$  with the polarization parallel to  $z$ -axis at 150  $^{\circ}\text{C}$ . The highest average conversion efficiencies of 41.3% and 19% were obtained, which are both lower than that for QPM(o) blue SHG in PPLN at the same temperature.

For the first-order type I QPM (o) and QPM(e) SHG in PPLN with the length of 20 mm, the theoretical temperature acceptance bandwidths are about 2.4 and 1.0  $^{\circ}\text{C}$ , respectively as shown as in Fig. 3, and the

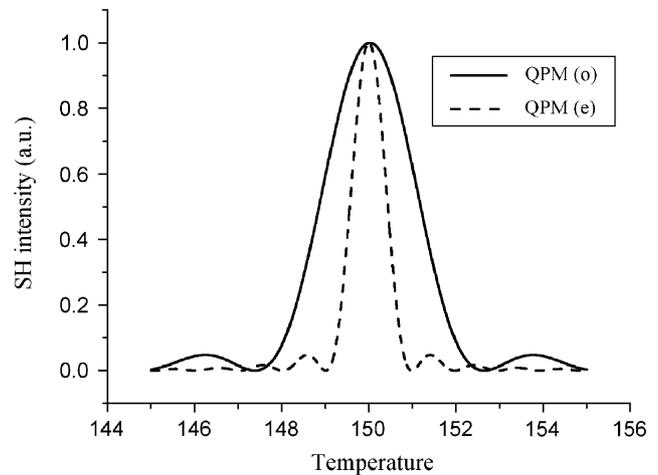


Fig. 3. Theoretical temperature tuning curves for SHG from 0.946  $\mu\text{m}$  centered at 150  $^{\circ}\text{C}$  in a 20 mm-long PPLN, for first-order QPM(o) and first-order QPM(e). The FWHM acceptance bandwidths are 2.4 and 1.0  $^{\circ}\text{C}$ .

experimental temperature acceptance bandwidths were about 2.0 and 0.9  $^{\circ}\text{C}$ , respectively, as shown in Fig. 4. The experimental results agree well with the calculated results, which show the polarization dependence of QPM in PPLN.

#### 4. Conclusion and discussion

In conclusion, we have demonstrated blue harmonic generation of 52  $\mu\text{J}$  by employing the type I QPM (o) SHG in PPLN at 150  $^{\circ}\text{C}$ . The  $d_{31}$  nonlinear coefficient was utilized for the QPM (o) interaction, and the grating was made for the first-order quasi-phase matching. Pumped by the same laser, the blue generation at

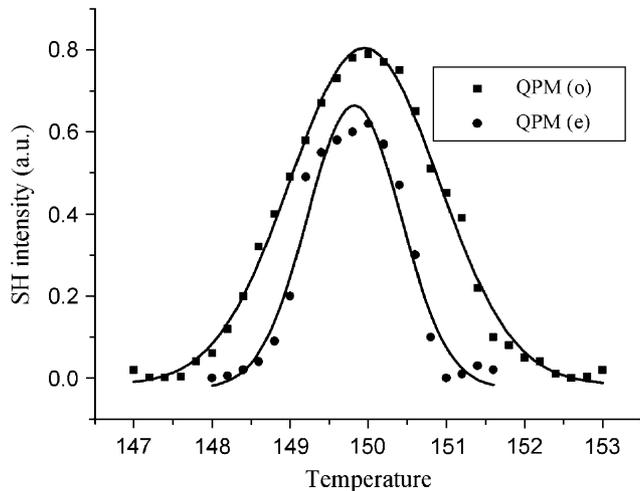


Fig. 4. Measured temperature tuning curves for SHG from  $0.946\ \mu\text{m}$  in a 20 mm-long PPLN, for first-order QPM(o) and first-order QPM(e). The FWHM acceptance bandwidths are  $2.0$  and  $0.9\ ^\circ\text{C}$ .

$0.473\ \mu\text{m}$  by employing the conventional type I first-order and third-order QPM(e) SHG in PPLN were also demonstrated. The highest average conversion efficiency of  $41.3\%$  and that of  $19\%$  were obtained at  $150\ ^\circ\text{C}$ . The larger experimental temperature acceptance bandwidth of  $2.0\ ^\circ\text{C}$  for QPM(o) than that of  $0.9\ ^\circ\text{C}$  for QPM(e) enhances the QPM(o) SHG phase matching and conversion efficiency.

The QPM(o) scheme makes it possible to use a larger period grating than those available with conventional type I QPM(e), which considerably eases the fabrication of the periodically domain-inverted structure. Otherwise the larger acceptance bandwidths of type I QPM(o) process also assure better optical nonlinear performances. This technique can be useful for other nonlinear optical parametric generations such as QPM OPO, sum frequency generation and different frequency genera-

tion, etc., and can also be used to broaden phase-matching wavelength bandwidth at communication band.

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