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Polarization dependence of quasi-phase-matched second-harmonic generation in bulk periodically poled LiNbO₃

Yuping Chen, Xianfeng Chen, Shengwu Xie, Xianglong Zeng, Yuxing Xia and Yingli Chen

Institute of Optics and Photonics, Department of Applied Physics, Shanghai Jiao Tong University, Shanghai 200240, People's Republic of China

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Abstract

Numerical comparisons of the acceptance bandwidths for quasi-phase-matched $E_Z^{\omega} E_Z^{\omega} - E_Z^{2\omega}$ (ee–e) and $E_Y^{\omega} E_Y^{\omega} - E_Z^{2\omega}$ (oo–e) second-harmonic generation processes in bulk periodically poled LiNbO₃ (PPLN) are given. The larger acceptance bandwidths and grating periods for the latter process lower the fabrication constraints, especially in the short-wavelength region, and also enhance the frequency conversion efficiency. The corresponding (oo–e) quasi-phase-matching conditions in bulk PPLN are discussed for different fundamental wavelength regions.

Keywords: LiNbO₃, quasi-phase-matching, second-harmonic generation, polarization

1. Introduction

Quasi-phase-matching (QPM) [1-6] permits noncritical phase matching in nonlinear optical interactions through compensation of the relative phase mismatch by regular domain inversion, which is based on a periodic domain grating built into the nonlinear medium by periodically changing the spontaneous polarization of the crystal. The main advantage of QPM is that noncritical phase matching can be used at any wavelength within the transparency range of the nonlinear material by appropriate choice of the period for the domain inversion, which can greatly broaden the range of application. Additionally, QPM permits noncritical phase matching with the largest nonlinear coefficient of the nonlinear optical crystal, without the need for strictly orthogonal polarizations. A great number of experiments have already been performed with QPM [2–6]. LiNbO₃ is an ideal nonlinear material for QPM, with a transparency range from 0.35 to 5.0 μ m, covering the visible, near-infrared and mid-infrared regions. We have successfully realized domain inversion [7] with a 6.5 μ m period by periodic electric poling in Z-cut LiNbO₃ at room temperature, where the three interacting beams propagate along the x-axis of the crystal, with each beam polarized parallel to the *z*-axis of the crystal (extraordinary rays). About 18 mW of cw 0.532 μ m green light was obtained at 5 °C, instead of 190 °C, with 1.1 W of cw 1.064 μ m pump power from an Nd:YAG laser and with end coupling. This corresponds to a normalized conversion efficiency of 1.5% cm⁻¹ W⁻¹.

The main reason for the relatively low conversion efficiency in our experiment is due to the departures from ideal QPM periodicity. The QPM temperature shift in our experiment is a result of a fabrication error in the grating period [8]. It is particularly difficult to maintain the uniformity of the QPM grating, especially in the short-wavelength region. The shorter the period is, the narrower the acceptance bandwidth for QPM second-harmonic generation (SHG). Higher-order QPM $E_Z^{\infty}E_Z^{-}-E_Z^{2\omega}$ (ee–e) SHG (which is called QPM(e) SHG hereinafter) is always used to alleviate this fabrication constraint since larger periods can be used. However, in this case, 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).

In this paper we suggest a new QPM method using input waves with perpendicular polarizations, instead of

using the same polarization but higher-order QPM waves. The influence of polarization on the full-width-at-halfmaximum (FWHM) acceptance bandwidth of QPM SHG performance will be investigated. The advantages of this new QPM approach is revealed through numerical comparison of the acceptance bandwidths for QPM(e) SHG and QPM $E_{y}^{\omega}E_{y}^{\omega}-E_{z}^{2\omega}$ (oo-e) SHG (called QPM(o) SHG hereinafter) in periodically poled lithium niobate (PPLN) at a temperature of 150 °C, and the same result is also suitable for QPM SHG at other temperatures at which photorefractive damage can be avoided in LiNbO₃. In [9], QPM of perpendicular polarization waves with the d_{24} coefficient was used to increase the acceptance bandwidth of SHG in periodically poled KTP; i.e., the process was $E_Y^{\omega} E_Z^{\omega} - E_Y^{2\omega}$ instead of $E_Z^{\omega} E_Z^{\omega} - E_Z^{2\omega}$, which is used for QPM SHG with the d_{33} coefficient. In periodically poled LiNbO₃, the highest nonlinear coefficient d_{33} is frequently used in QPM(e) technology. However the lower acceptance bandwidth and the short grating period reduce the performance of the QPM parametric process. To solve this problem, QPM(o) with d_{31} has been employed for LiNbO₃ [10].

The organization of this paper is as follows. In section 2, we introduce the parametric FWHM acceptance bandwidth theory of QPM SHGs in bulk PPLN. In section 3, we give results based on the numerical comparison of acceptance bandwidths and grating periods for collinear SHG interactions with two kinds of QPM method. We also discuss different phase matching conditions for QPM(o) SHG in different fundamental wavelength regions. The periods and effective nonlinear coefficients of third-order QPM(e) SHG and first-order QPM(o) SHG are then discussed. In section 4, the whole work is summarized.

2. FWHM acceptance bandwidth theory of QPM SHG in bulk PPLN

Assuming low conversion efficiency, loose focusing of the pump laser in the sample, cw interaction and no loss of the fundamental and second-harmonic waves, the periodically modulated nonlinear coefficient can be expressed [11, 12] by the Fourier expansion

$$d(x) = d_{eff} \sum_{m=-\infty}^{\infty} \frac{2}{m\pi} \sin\left(\frac{m\pi l}{\Lambda}\right) \exp\left(-i\frac{m\pi}{\Lambda}z\right)$$
(1)

where d_{eff} is the effective nonlinear coefficient of SHG in single-domain bulk material, Λ is the period of the modulated structure, l the length of the reversed domain and integer m is the order of the QPM interaction. For *m*th-order QPM (m is an odd number) and a perfect duty cycle $D = 1/\Lambda = 1/2$, the *m*th-order grating wavevector is $k_m = 2\pi m/\Lambda$, so from the theory of conventional birefringent phase matching the QPM can be described with the substitutions

$$d_Q = d_{eff} \frac{2}{\pi m} \tag{2}$$

and

$$\Delta k_Q \equiv \Delta k - k_m = k_2 - 2k_1 - k_m = \frac{\pi}{l_c} - k_m$$
$$= \frac{4\pi (n_2 - n_1)}{\lambda} - \frac{2\pi m}{\Lambda}$$
(3)

where d_Q is the amplitude of the relevant harmonic of d(x), Δk_Q is the total wavevector mismatch while Δk is the wavevector mismatch due to material dispersion, k_1 and k_2 are the fundamental and second-harmonic wavevectors, respectively, and $l_c = \lambda/4(n_2 - n_1)$ is the coherent length. Here n_1 and n_2 are the indices of refraction for the fundamental and second-harmonic waves at the given QPM temperature, respectively, and are determined by the Sellmeier equations [8, 13].

In Z-cut PPLN, assuming that the three interacting beams propagate along the x-axis of the crystal, the nonlinear polarization leading to second-harmonic generation in terms of d can be described by equation (4) below, where the polarization components of the fundamental wave along the yand z-axes will contribute to the SHG with different nonlinear coefficients:

$$P_z(2\omega, t) = d_{31}E_v^2(\omega, t) + d_{33}E_z^2(\omega, t).$$
(4)

In addition, the conversion efficiency η of QPM SHG is proportional to $(d_Q)^2$ and sinc²($\Delta k_Q L/2$), as shown in equation (5):

$$\eta \propto (d_Q)^2 * \sin c^2 (\Delta k_Q L/2) \tag{5}$$

where $\sin c(\Delta k_Q L/2) \equiv \sin(\Delta k_Q L/2)/(\Delta k_Q L/2)$.

For QPM collinear interaction in a PPLN crystal of length L containing uniform domain inversion grating periods, the phase-matching condition can be altered by changing the input wavelength or the operating temperature of the sample according to equation (3). It is important when designing QPM devices to understand how sensitive the phase-matching condition (and thus the conversion efficiency) is to these changes.

2.1. Wavelength acceptance bandwidth

The phase-matching factor [11, 12] in the expression for the power conversion efficiency is $\operatorname{sinc}^2(\Delta k_0 L/2)$. То calculate the wavelength acceptance bandwidth, we consider SHG interaction at a fixed temperature (a similar procedure is followed to calculate wavelength acceptance bandwidths for difference frequency generation (DFG), sum frequency generation (SFG) and optical parametric oscillation (OPO)). A desired pump wavelength (i.e. fundamental wavelength), $\lambda_{1, peak}$ is chosen. The QPM period which satisfies the phasematching equation at that wavelength is designed and fixed for the QPM device. The value of λ_1 is then increased slightly from $\lambda_{1, peak}$ and the second-harmonic wavelength λ_2 is equal to half the fundamental wavelength λ_1 . The phase mismatch $\Delta k(\lambda_1)$ at this new wavelength is calculated according to the following equation:

$$\Delta k_{\mathcal{Q}} = \frac{4\pi (n_2 - n_1)}{\lambda_1} - \frac{2\pi}{\Lambda}.$$
 (6)

The value $\lambda_{1,HM}$ at which the conversion efficiency λ_1 falls to half of its maximum value is calculated by solving the equation

$$\frac{\sin^2 \left[\Delta k_Q(\lambda_{1,HM})L/2\right]}{\left[\Delta k_Q(\lambda_{1,HM})L/2\right]^2} = \frac{1}{2}.$$
 (7)

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Figure 1. Period of PPLN versus fundamental wavelength at 150 °C, for first-order QPM(e) and QPM(o) SHG.

The pump wavelength acceptance bandwidth $(\delta \lambda)_{1,FWHM}$ for the interaction is given by

$$(\delta\lambda)_{1,FWHM} = 2(\lambda_{1,HM} - \lambda_{1,peak}). \tag{8}$$

Equation (7) shows that the bandwidth is inversely proportional to crystal length. In the acceptance bandwidth calculation of section 3, we shall assume a standard crystal length of 1 cm.

2.2. Temperature acceptance bandwidth

In contrast to the wavelength acceptance bandwidth, deviations in the temperature acceptance bandwidth result from $(\Delta k_Q L)$, where Δk_Q is changed by the index dependence on operating temperature, while the thermal expansion can also change the period Λ and length L of PPLN devices. The temperature acceptance bandwidth is calculated as follows: the fundamental and second-harmonic wavelengths, λ_1 and λ_2 , are chosen, and the QPM period required for phase matching at the desired operating temperature is calculated. The temperature is then allowed to deviate from this optimum value. The phase mismatch as a function of temperature is calculated from the following equation:

$$\Delta k_{Q} = \frac{4\pi [n_{2}(T) - n_{1}(T)]}{\lambda_{1}} - \frac{2\pi}{\Lambda(T)}.$$
(9)

The indices and dispersion of the fundamental and secondharmonic ordinary beam and extraordinary beam at any wavelength in the transparency range of LiNbO₃ can be obtained numerically from the Sellmeier equation [8, 13] for a given QPM temperature. The temperature dependence of the QPM period is then calculated from the thermal expansion equation

$$\delta l = \alpha \cdot \Lambda \cdot \delta T \tag{10}$$

where α is the linear coefficient of thermal expansion. In LiNbO₃, α is equal to 1.54×10^{-5} [13]. The temperature acceptance bandwidth is then calculated just as the wavelength acceptance bandwidth was calculated above. This acceptance bandwidth is also, to a good approximation, inversely proportional to crystal length *L*.



Figure 2. The wavelength acceptance bandwidths of PPLN versus fundamental wavelength, 150 °C, for the first-order QPM(e) SHG and for the first-order QPM(o) SHG.

3. Numerical calculation of acceptance bandwidths and grating periods for interactions with two different polarizations

To investigate the polarization dependence of the QPM SHG collinear interaction in bulk PPLN, we calculate the grating periods for first-order QPM(o) SHG and first-order QPM(e) SHG, and give a numerical comparison of their wavelength and temperature acceptance. For QPM(e) SHG n_1 and n_2 are the indices of the fundamental extraordinary and second-harmonic extraordinary light, while for QPM(o) SHG n_1 and n_2 represent the indices of the fundamental ordinary and second-harmonic extraordinary light, respectively.

From the equation of the grating period, $\Lambda = \lambda/2$ $(n_2 - n_1)$, we can plot the grating period versus fundamental wavelength from 0.76 μ m to 1.09 μ m, as shown in figure 1. Here we see that, for the same order QPM SHG, the longer the fundamental wavelength, the larger the QPM period. Moreover, the domain period for first-order QPM(o) SHG is longer than that for first-order QPM(e) SHG, which facilitates the fabrication of QPM devices, decreases the possibility of phase mismatching and thus can improve the conversion efficiency.

From equations (6)–(8), the wavelength acceptance bandwidths for first-order QPM(o) and QPM(e) SHG are shown in figure 2.

In figure 2, we find that the wavelength acceptance bandwidths for the first-order QPM(o) SHG are greater than those for the first-order QPM(e) SHG at the same fundamental wavelength, λ , especially at longer wavelengths; wavelength bandwidths increase at the long wavelengths in which LiNbO₃ has the lower dispersion, which improves the SHG performance.

When the QPM temperature is given at $150 \,^{\circ}$ C, calculated by equation (9) and (10) and the same method as the calculation of wavelength acceptance bandwidth, the temperature acceptance bandwidths are obtained as shown in figure 3.

From figure 3, we can obtain results similar to those shown in figure 2: at the same fundamental wavelength λ and the same temperature 150 °C, the tunable temperature acceptance bandwidth for first-order QPM(o) SHG is larger than that for



Figure 3. The temperature acceptance bandwidths of PPLN versus fundamental wavelength, 150 °C, for the first-order QPM(e) SHG and for the first-order QPM(o) SHG.



Figure 4. Refractive indices of PPLN for second-harmonic extraordinary light and fundamental ordinary light versus fundamental wavelength at 150 °C.

first-order QPM(e) SHG, while for the same type of QPM SHG, the temperature acceptance bandwidth increases as the wavelength becomes longer.

From the above analysis, we see that the tolerance of QPM(o) SHG is larger than that of the same order QPM(e) SHG. Moreover, the phase matching condition for QPM(e) SHG in bulk PPLN is related to the fundamental wavelength, as shown in figure 4.

When $n_2(e) = n_1(o)$, where $n_2(e)$ and $n_1(o)$ represent the refractive indices of the second-harmonic extraordinary light and the fundamental ordinary light, respectively, the fundamental wavelength λ is equal to about 1.13 μ m, corresponding to critical phase matching. When $n_2(e) >$ $n_1(o)$, then $\lambda < 1.13 \ \mu$ m and the fundamental wave is in the shorter-wavelength region, so the QPM period takes positive values. In the longer wavelengths where $\lambda > 1.13 \ \mu$ m, the period is negative, which implies that $n_2(e) < n_1(o)$. In this paper we only investigate the performance of QPM(o) SHG in the visible region.

It is also important to compare higher-order QPM(e) SHG, such as the third order, with the first-order QPM(o)



Figure 5. The periods of PPLN versus fundamental wavelength, 150 °C, for the first-order QPM(o) SHG and for the third-order QPM(e) SHG.

SHG. From equations (4) and (1), we know that QPM(e) SHG and QPM(o) SHG utilize the nonlinear coefficients d_{33} and d_{31} , respectively. The corresponding *m*th-order effective nonlinear coefficients are $d_Q(e) = (2d_{33}/\pi m)^2$ and $d_Q(o) = (2d_{31}/\pi m)^2$, respectively. As shown in figure 1, it is clear that the periods are larger for the QPM(o) SHG than for the QPM(e) SHG of the same order at a given temperature. We also find that, at the same temperature, the periods for the first-order QPM(o) SHG are almost the same as, or larger than, those for the third-order QPM(e) SHG. The comparison between them is shown in figure 5.

For example, at 150 °C, for blue SHG pumped by a laser with a wavelength of 0.946 μ m, the period for firstorder QPM(o) SHG is about 14.5 μ m, which is larger than the 13.5 μ m period for the third-order QPM(e) SHG. The theoretical ratio of the conversion efficiencies of the two processes is $d_{Q3}(e)/d_{Q1}(o) = (2d_{33}/\pi \cdot 3)^2/(2d_{31}/\pi \cdot 1)^2 = (27/3)^2/8^2 = 1.26$ (assuming $d_{33} = 27$, $d_{31} = 8$) [14]. However, the larger grating period and 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.

4. Conclusion

In this paper a new QPM SHG method with perpendicular polarization waves rather than parallel polarization waves is proposed. A comparison of the tuning properties of QPM(o) SHG and QPM(e) SHG in bulk PPLN is given. The dependence of phase-matching FWHM acceptance bandwidth on temperature and wavelength and the relevant tuning curves are calculated, which manifests the polarization dependence of QPM SHG. The results show that the various acceptance bandwidths can be significantly enhanced for QPM interactions through use of the proper polarization rather than by simply using the higher-order traditional QPM(e) SHG with the same polarization waves.

At a given temperature, the domain grating period for the first-order QPM(o) SHG is larger than that for QPM(e) SHG of not only the first order, but also the third order, which allows much better uniformity of the domain-inversion grating and lowers the device fabrication constraints. The polarization dependence of QPM SHG can also be applied to other QPM nonlinear parametric processes, such as optical parametric oscillation (OPO), sum frequency generation (SFG) and different frequency generation (DFG).

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