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High-efficiency single-pass cw quasi-phase-matched frequency doubling based on PP-MgO:SLT*

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We demonstrate a single-pass cw quasi-phase-matched (QPM) frequency doubling based on periodically poled MgO-doped stoichiometric lithium tantalate (PP-MgO:SLT) with crystal length of 20mm. For the maximum 1064nm fundamental power of 7.69W, the maximum conversion efficiency and the maximum output second-harmonic power are 11.8% and 905mW, respectively. In the experiment we found that the optimum temperature of the oven for PP-MgO:SLT should be adjusted with the change of the fundamental power and the focal length. In addition, angular acceptance bandwidth and temperature acceptance bandwidth are studied. Their experimental results are in good agreement with the theoretical calculations.

Keywords: quasi-phase-matching, periodically poled MgO-doped stoichiometric lithium tantalate (PP-MgO:SLT), second-harmonic generation, angular acceptance bandwidth
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1. Introduction

Quasi-phase-matched (QPM) frequency doubling[1−15] is an effective method for expanding the outputs of various laser sources to short wavelengths. For the QPM technique,[16−19] a periodical modulation of the sign of the second-order nonlinear polarization is used to compensate for the difference of propagation vectors by a grating vector in a nonlinear crystal. Because the phase matching condition for QPM can be satisfied by the grating period irrelative to the inherent properties of the crystal, there is no restriction imposed on material and polarization. As a result, QPM can use the largest tensor component which allows high conversion efficiency for any wavelength in the transparency range of the crystal. In addition, noncritical QPM can be easily realized, which means that the demands of the fundamental beam angle of divergence and the adjusting angle of crystal can decrease due to its larger angular acceptance bandwidth.

Although the nonlinear grating can be fabricated to realize QPM second-harmonic generation (SHG) in germanosilicate fibres,[7] the most commonly used QPM materials are periodically poled lithium niobate (PPLN),[2,5,13] periodically poled $KTiOPO_4$ (PPKTP),[6,9,14,15] periodically poled lithium tantalate (PPLT)[3,4,11,12,20] and other periodically poled ferroelectric crystals. Because lithium tantalate (LT) has the advantages of higher transmission at ultraviolet band and higher resistance to photorefractive damage, it is more suitable for QPM SHG.[12] Recently, stoichiometric LT (SLT) has appeared, whose coercive field is ∼1 order of magnitude lower than congruent-melt LT (CLT), and consequently it is more

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convenient for the method of applying an external electric field.[20,21] In addition, SLT has the stronger nonlinear-optic effects.[10,21] It has been reported that slight MgO doping in the SLT crystals almost eliminated the photorefractive damage.[21] In our experiment, we used periodically poled MgO-doped SLT (PP-MgO:SLT). As far as we know, this is the first report of QPM SHG by using PP-MgO:SLT crystal.

There are three SHG configurations available, i.e. the single-pass extracavity frequency doubling, the resonantly enhanced extracavity frequency doubling and the intracavity frequency doubling. The disadvantages of resonantly enhanced extracavity frequency doubling and intracavity frequency doubling result from the rather complex configuration and high sensitivity to the thermal effect.[14] For the single-pass extracavity frequency doubling, it has a simple configuration, where there is no direct relation between lasing and frequency doubling processes, which means that the green problem can be eliminated.[22] Though it is generally accepted that single-pass cw extracavity frequency doubling has a lower conversion efficiency due to the limitation of the interaction length, high efficiency single-pass cw QPM SHG has been demonstrated. The highest conversion efficiency of the single-pass extracavity frequency doubling for cw lasers in the bulk periodically poled crystal is 42%, which was reported by Miller *et al.*[23] The frequency-doubling crystal they used was PPLN with a length of 53mm and a grating period of 6.5μm. From this experimental result, it can be seen that if the periodically poled crystal is long enough, high-efficiency single-pass cw QPM SHG could be realized. However, up to now, high-efficiency (>10%) single-pass cw QPM SHG based on LT has not been reported. In this paper, we present the single-pass cw QPM SHG based on PP-MgO:SLT with a conversion efficiency ($\eta_{\rm Q}$) of 11.8%. 905mW of second-harmonic power was generated with 7.69W of 1064nm fundamental power (P_λ). In fact, the length (L) of PP-MgO:SLT we used was only 20mm, and the normalized conversion efficiency $\eta_{\rm Q}/(P_\lambda \cdot L^2)$ reached 0.39%/W·cm^2 which was higher than 0.23%/W·cm^2 obtained by Miller *et al.*[23] Moreover, we have made theoretical analysis and experimental research on angular acceptance bandwidth and temperature acceptance bandwidth, whose experimental values were respectively 4.6° and 1.6°C, showing good agreement with our theoretical predictions.

2. Theoretical analysis

We consider the QPM configuration conditions as that PP-MgO:SLT is periodically poled along the crystal z-axis and all interacting waves are also polarized parallel to the crystal z-axis with the propagation direction paraxially along the crystal x-axis. The phase mismatch $\Delta \boldsymbol{k}_{\rm Q}$ is given by

$$\Delta \boldsymbol{k}_{\rm Q} = \boldsymbol{k}_{\lambda/2} - \boldsymbol{k}_\lambda - \boldsymbol{k}_\lambda - \boldsymbol{k}_m, \tag{1}$$

where $\boldsymbol{k}_\lambda$, $\boldsymbol{k}_{\lambda/2}$ and $\boldsymbol{k}_m$ are the vectors of the fundamental wave, the second-harmonic wave and the QPM grating in the crystal, respectively. λ and $\lambda/2$ represent respectively the wavelengths of the fundamental wave and the second-harmonic wave. m is the QPM order. In general, m is taken to be unity in order to obtain the largest effective nonlinear coefficient for QPM. In the case of collinear configuration, we have $k_m = 2\pi m/\Lambda(T)$. $\Lambda(T)$ is the grating period at crystal temperature T, expressed as

$$\Lambda(T) = \Lambda(25^\circ{\rm C})[1 + a(T - 25^\circ{\rm C}) + b(T - 25^\circ{\rm C})^2], \tag{2}$$

where a and b are the thermal expansion coefficients of MgO:SLT. Since we have not found the thermal expansion coefficients of MgO:SLT, we will use the coefficients of CLT as Bruner *et al*[21] did, a=1.6×10^{-5} and b=7.0×10^{-9}. If the crystal is rotated about its z-axis for a certain angle α, which results in the oblique incidence of the fundamental wave on the end face of the crystal, $\boldsymbol{k}_\lambda$ and $\boldsymbol{k}_{\lambda/2}$ may be noncollinear in the crystal and noncollinear QPM SHG would be formed. The crystal rotation angle, α, is just the incident angle of the fundamental wave outside the PP-MgO:SLT crystal. We take θ as the refraction angle of the fundamental wave inside the crystal, and it should satisfy the law of refraction (the refractive index in air is taken to be 1)

$$\sin\alpha = n\lambda, T \cdot \sin\theta, \tag{3}$$

where $n(\lambda, T)$ is the refractive index of the periodically poled crystal which is determined by its temperature-dependent Sellmeier equations. Nakamura *et al*[20] have reported the Sellmeier equations for the refractive index of MgO:SLT at room temperature, and pointed out that the refractive indices of SLT were found to be almost independent of MgO concentration at the doping level of 0.5 and 1.0 mol%. At present, however, there is no report of the temperature-dependent Sellmeier equations for the refractive index of MgO:SLT crystal. But we can theoretically analyze the PP-MgO:SLT SHG by using the temperature-dependent Sellmeier equation for

the refractive index of undoped SLT crystal[21] to obtain a qualitative conclusion on the assumption that there is not a significant difference between the two crystals. In view of the symmetry, the plus–minus sign of θ (or α) can be defined arbitrarily. When $|a|$ is small, the refraction angle of the second-harmonic wave in the PP-MgO:SLT crystal approximates to θ. Consequently, the value of the grating period in the direction of $\boldsymbol{k}_\lambda$ can be taken as the effective grating period $\Lambda_{\rm eff}(T)$.[17,24] In this approximation, the relation between $\Lambda_{\rm eff}(T)$ and θ can be described as

$$\Lambda_{\rm eff}(T) = \Lambda(T)/\cos\theta, \tag{4}$$

and the phase mismatch can be approximated by the following expression using Eq.(1):

$$\begin{aligned}\Delta k_{\rm Q} =& k_{\lambda/2} - 2k_\lambda - k_m \\ =& 4\pi[n(\lambda/2,T) - n(\lambda,T)]/\lambda - 2\pi m/\Lambda_{\rm eff}(T). \end{aligned}\tag{5}$$

If the Fresnel reflections on the input and output end faces of the crystal are ignored, the conversion efficiency of single-pass extracavity frequency doubling (i.e. $\eta_{\rm Q}$) in the case of low-conversion can be described as follows:

$$\eta_{\rm Q} = K \cdot P_\lambda \cdot L^2 \cdot {\rm sinc}^2(\Delta k_{\rm Q} \cdot L/2), \tag{6}$$

where L is the interaction length and P_λ is the power of the fundamental wave. When the wavelength of the fundamental wave is fixed, K is a constant. If the change of L with temperature or other parameters is ignored and the wavelength of the fundamental wave is fixed, from Eq.(6) we can find that the condition to maximize the conversion efficiency of QPM SHG (i.e. the phase matching condition) is $\Delta k_{\rm Q}$=0.

When the crystal is rotated a certain angle α about its z-axis, it can be seen from Eqs.(2)–(6) that $\eta_{\rm Q}$ would change correspondingly. The angular acceptance bandwidth of QPM SHG is defined as the full width at half-maximum of α in $\eta_{\rm Q}$. In view of the small size (such as $\leq$1mm) of the periodically poled crystal along its z-axis, we only analyse the angular acceptance bandwidth with the crystal rotating about the z-axis (i.e. the angular acceptance bandwidth with the fundamental wave rotating about the crystal z-axis in the x–y plane) because the fundamental wave commonly propagates paraxially along the x-axis and enters the crystal at the centre of its end face.

As an example for PP-MgO:SLT SHG, if λ=1064nm and Λ (25°C)=7.93μm, the crystal temperature should be 74.1°C in order to satisfy the phase matching condition in the case of collinear configuration. Figure 1 shows the dependence of the conversion efficiency on the crystal temperature of collinear QPM on the assumption that the crystal length is 20mm (this same value is used below) by using Eqs.(2)–(6). The theoretical temperature acceptance bandwidth is 1.3°C. Figure 2 shows the dependence of the conversion efficiency on the corresponding incident angle of the fundamental wave outside the PP-MgO:SLT crystal for T=74.1°C. The theoretical angular acceptance bandwidth is 4.6°. It can be seen from Fig.2, that $\eta_{\rm Q}$ reaches its maximum value when α is equal to zero, and $\eta_{\rm Q}$ falls when α deviates from zero. That case is shown in Fig.2 as a near-rectangular single-peak structure for the angle-tuning curve of $\eta_{\rm Q}$ when α is around zero.

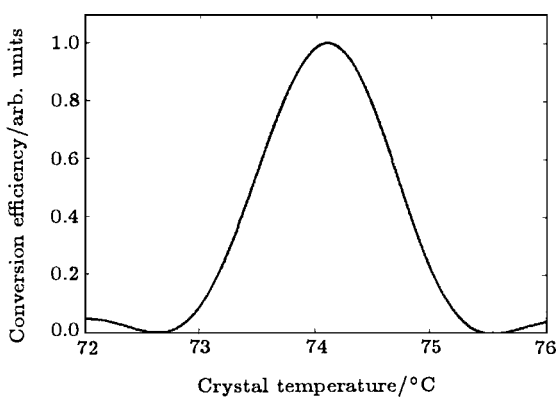


Fig.1. Theoretical conversion efficiency as a function of the crystal temperature for collinear QPM SHG (λ=1064nm, Λ (25°C)=7.93μm, L=20mm).

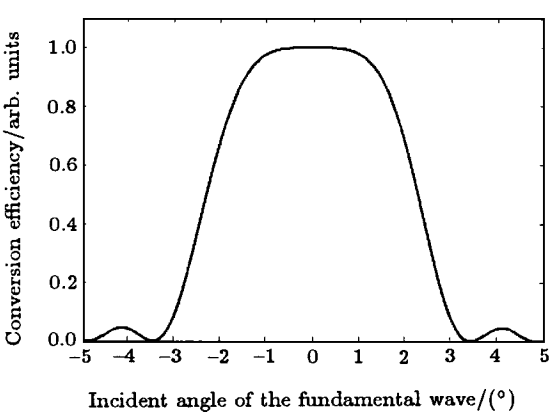


Fig.2. Theoretical conversion efficiency as a function of the incident angle of the fundamental wave (λ=1064nm, Λ (25°C)=7.93μm, L=20mm, T=74.1°C).

It is particularly indicated that in the experiment the temperature of the oven for PP-MgO:SLT crystal is not set at the theoretically calculated value of

74.1°C. One reason is that we use the thermal expansion coefficients of CLT and temperature-independent Sellmeier equation of the refractive index of undoped SLT crystal in our calculations. Furthermore, though the periodically poled crystal is thin in the z direction, there is a certain deviation between the oven temperature and the actual temperature of the crystal. In order to obtain the maximum harmonic output, the oven temperature should be adjusted with the change of the power, the waist size or the location of the input fundamental wave. However, the calculated results provide an important theoretical guide to the setting of the oven temperature which should be about 74°C.

3. Results of single-pass cw QPM SHG based on PP-MgO:SLT

The experimental setup of the single-pass extracavity frequency doubling in PP-MgO:SLT crystal is shown in Fig.3. The fundamental source used in our experiment was a 1064nm cw diode-pumped Nd:YVO_4 laser. The laser mode was TEM_{00} with beam quality factor $M^2 < 1.3$. This beam was focused into PP-MgO:SLT crystal by a focusing lens and propagated paraxially along the x-axis of the PP-MgO:SLT crystal. The PP-MgO:SLT crystal had a grating period of 7.93μm (at room temperature), which was periodically poled along the crystal z-axis. This grating period was satisfied to achieve first-order QPM SHG of 1064nm at a temperature of ~74°C. The PP-MgO:SLT crystal had a length of 20mm (at room temperature) and a thickness of 1mm. Both its end faces were antireflection coated at 532nm and 1064nm. PP-MgO:SLT crystal was mounted in a temperature-controlled oven with the operating range from 25 to 250°C which permitted accurate temperature tuning for the phase matching condition. A dielectric filter was used to block the fundamental wave so as to let the harmonic wave pass through with high transmission. A Molectron EPM1000 power meter was used to measure the powers of the input fundamental wave and the output harmonic wave.

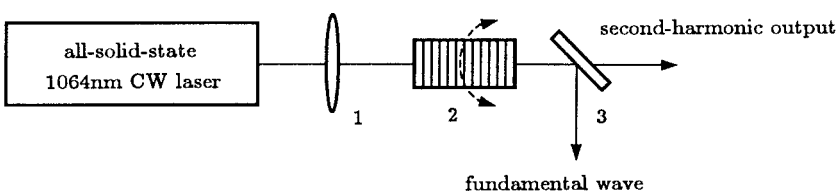


Fig.3. Experimental setup of the single-pass cw extracavity frequency doubling. 1 is focusing lens; 2 is the PP-MgO:SLT crystal; 3 is dielectric filter.

We fixed the input power of the fundamental wave at a lower level and measured the conversion efficiencies when the oven temperature changed around 74°C in the case of collinear configuration. Two focusing lenses were used in this experiment in sequence. Firstly, the fundamental beam was loosely focused into the PP-MgO:SLT crystal in a spot size of ~100μm radius by using a focusing lens (referred to as "F1" in this paper) with a focal length of 100mm. The distance between F1 and PP-MgO:SLT was kept at 114mm, and the power of the fundamental wave after F1 was kept at 3.14W. Figure 4 shows the conversion efficiency achieved as a function of the oven temperature for collinear QPM SHG. As we can see, the optimum oven temperature was 73.2°C under the conditions mentioned above, and the experimental temperature acceptance bandwidth was 1.6°C which is basically in accordance with our calculation. Then, a focusing lens (referred to as "F2" in this paper) with a focal length of 50mm was used to substitute for F1. The distance between F2 and PP-MgO:SLT was kept at 53mm, and the power of the fundamental wave after F2 was kept at 3.18W. In this case, the optimum oven temperature obtained from this experiment was not 73.2°C, but 72.8°C. These results indicate that the thermal field distribution in the crystal is affected by space focusing conditions. Figure 5 is a plot of conversion efficiency versus fundamental power while using F2. The second-harmonic power achieved while using F2 was much more than that while using F1. From Fig.5 we can see that along with the increase of the fundamental power, the conversion efficiency is increased linearly. In other words, the SHG power is proportional to the square of the fundamental power. This result is in excellent agreement with Eq.(6).

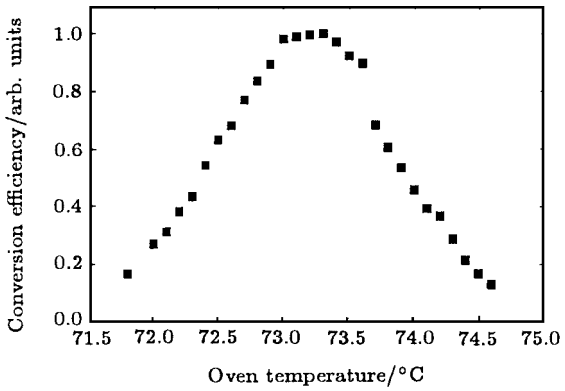


Fig.4. Experimental conversion efficiency as a function of the oven temperature for collinear QPM SHG while using F1.

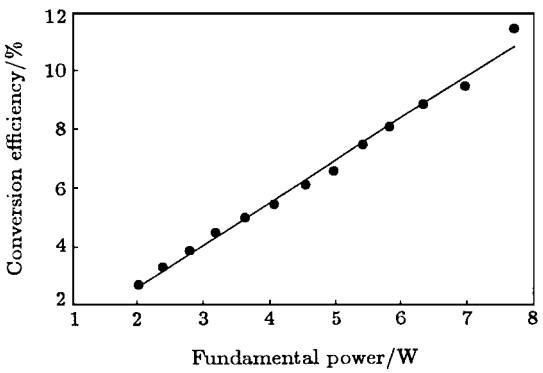


Fig.5. Dependence of experimental conversion efficiency on the fundamental power for collinear QPM SHG at the oven temperature of 72.8°C.

We fixed the power of the input fundamental wave at the highest level (7.69W) and measured the conversion efficiencies when the oven temperature changed around 72.8°C in the case of collinear configuration while using F2. Figure 6 shows the second-harmonic power achieved as a function of the oven temperature for collinear QPM SHG while using F2 with 7.69W of 1064nm fundamental power. Obviously, the optimum oven temperature is 72.7°C, which is lower than 72.8°C obtained in the case of lower fundamental power. In our experimental results, the oven temperature should be reduced with the increase of the fundamental power. The maximum harmonic output is 905mW, and the maximum conversion efficiency is 11.8%.

We also measured the dependence of the conversion efficiency on the incident angle of the fundamental wave to obtain the experimental angular acceptance bandwidth. In this experiment, the fundamental beam was only loosely focused into PP-MgO:SLT crystal by using F1 in order to reduce the influence of the divergence of the fundamental wave upon the angular acceptance bandwidth. The oven temperature was 73.2°C correspondingly. Figure 7 shows the angle-tuning curve of η_Q. In view of symmetry, we only measured the dependence when crystal rotated about its z-axis in one direction. It could be found that the angular acceptance bandwidth of PP-MgO:SLT SHG was 4.6°. The experimental result agrees completely with the theoretical analysis.

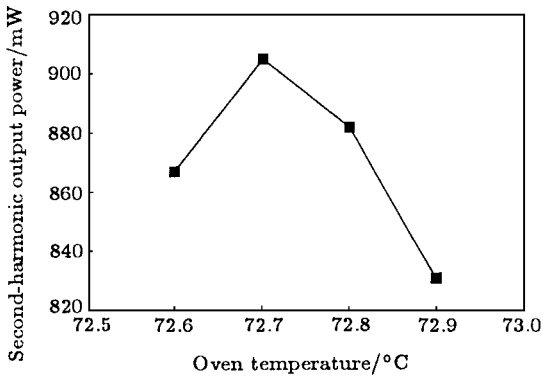


Fig.6. Second-harmonic output power versus the oven temperature for collinear QPM SHG with the maximum input fundamental power.

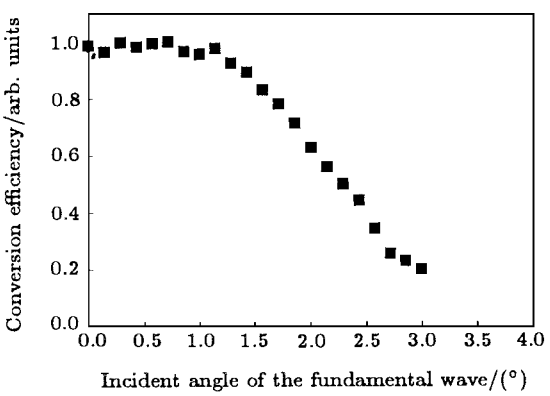


Fig.7. Experimental conversion efficiency as a function of the incident angle of the fundamental wave while using F1.

4. Conclusion

In summary, we have demonstrated a single-pass cw QPM SHG based on PP-MgO:SLT. For the maximum 1064nm fundamental power of 7.69W, the maximum conversion efficiency was 11.8% with the output second-harmonic power of 905mW. To our knowledge, no such discussion as high-efficiency (>10%) single-pass cw QPM SHG based on LT has been reported. There was a temperature difference between the oven temperature and the actual temperature of the crystal, and the temperature difference had a relation with

space focusing conditions and the input power of the fundamental wave. This result could be expressed as that the optimum oven temperature should be adjusted with the change of the fundamental power or the focal length in the experiment. Moreover, angular acceptance bandwidth and temperature acceptance bandwidth were studied. Their experimental values were respectively 4.6°and 1.6°C, which are in good agreement with our theoretical calculations.

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