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Enhanced Third Harmonic Generation by Introducing Quasi-Phase Mismatches Due to Electro-Optic Effect[1]

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Abstract—Third harmonic can be realized by the coupled second-harmonic ($\omega_1 + \omega_1 \longrightarrow \omega_2$) and sum-frequency ($\omega_1 + \omega_2 \longrightarrow \omega_3$) generation. However, in quasi-phase matching conditions, the efficiency of third harmonic is usually low due to the larger coupling coefficient ratio. In this paper, the third-harmonic generation in a periodically poled $LiTaO_3$ has been experimentally studied, where the electro-optic effect was employed to manipulate the phase mismatches and energy distribution of the coupled waves. With the DC electric field varying from 0 to 140 V/mm, the output of third harmonic can be enhanced from about 95 to 312 mW. The method provides a simple and convenient way to control the efficiencies of frequency conversion.

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1. INTRODUCTION

In the field of artificial microstructures, advances in quasi-phase-matching (QPM) technology based on periodically poled nonlinear crystals have inspired great interest [1]. As one excellent nonlinear material, $LiTaO_3$ (LT) is often used for various devices, especially in the field of frequency conversion. Periodically or quasi-periodically poled LT have been widely employed to realize Second harmonic generation (SHG) and Third harmonic generation (THG). For example, THG by use of coupled SHG and sum-frequency generation (SFG) processes has been demonstrated [2–6]. In the past few years, THG in a periodic optical superlattice with a 1342 nm pump light has been achieved [7]. THG of a 1064 nm pump light with a dual-periodic or a quasi-periodic optical superlattice (OSL) has also been investigated [8]. THG can also be realized in a $LiNbO_3$ channel waveguide [9]. In the above mentioned THG processes, the THG conversion efficiency turns out to be strongly dependent on the sample length, coupling coefficient ratio, and the input power of the fundamental wave [10]. In addition, LT is an interesting electro-optic material and many devices have been investigated employing its electro-optic (EO) effect, such as spectral filter [11], EO switch [12]. The influence of EO effect on second harmonic has been researched by Chen et al. [13], where simultaneous SHG and amplitude modulation in a periodic OSL were demonstrated using a DC electric field applied along the optical axis. Moreover, with the DC electric field normal to the optical axis, simultaneous SHG and polarization coupling have been proposed and realized in our group [14, 15].

Previous study showed that, under the QPM conditions, an efficient THG depends not only on the magnitude of the coupling coefficients but also on their ratio t. There exists a critical ratio τ ($\tau = 0.8858$), for which the efficiency of third harmonic could be maximal [10]. In many cases, the ratio t is larger than

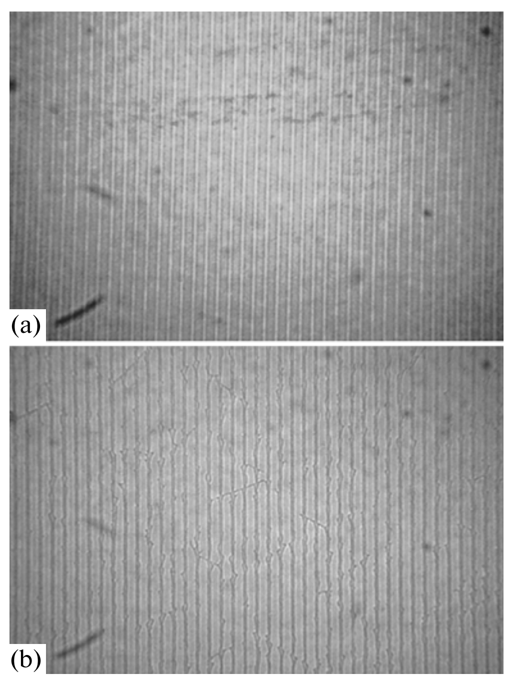

Fig. 1. Optical micrograph of etched domain-inverted patterns on the (a) +C and (b) –C surface of the OSL LT sample.

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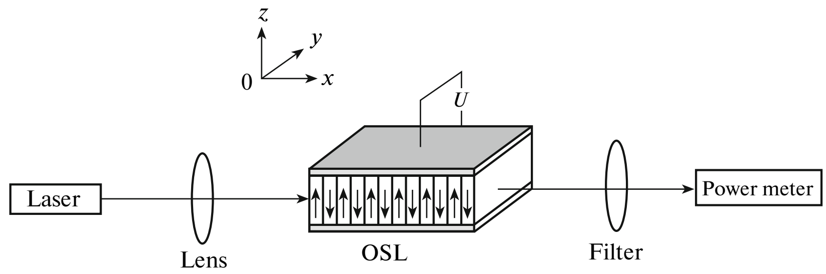

Fig. 2. The schematic view of experimental setup: The input source is a LD-pumped, Q-switched Nd:YVO_4 laser; the OSL LT sample is heated in an oven.

the critical value τ; as a result, the THG is greatly suppressed when the pump intensity increases. Actually, the above result is based on the quasi-phase-matching conditions; if the phase is mismatched, the result will be different (the nonlinear frequency conversion is sensitive to the phase which is related to the refractive index). In the case of electro-optic material, the refractive index can be modulated by an external DC electric field. In [16], the influence of the EO effect on coupled QPM process has been analyzed, which predicted that the conversion efficiency of TH can be enhanced compared with that obtained under the exact QPM conditions. In this paper, we investigate experimentally the THG based on the EO effect in the coupled QPM processes. With the application of DC electric field, a significant enhancement of TH output has been realized.

2. EXPERIMENT SETUP

In the experiment, a periodic OSL LT has been specially designed with the first-order reciprocal vector for compensating the phase mismatch of SHG and the third-order reciprocal vector for the SFG. According to the temperature-dependent Sellmeier equation of $LiTaO_3$ [17], the period of the sample is determined to be 14.78 μm, where the QPM conditions can be satisfied for the fundamental wavelength of 1342 nm at the working temperature 74.1°C. In order to enhance the effect of EO modulation on frequency conversion, we take an asymmetric-duty-cycle $D_c = 0.25$ [the corresponding Fourier coefficients are calculated to be $f_1 = 0.45$ and $f_3 = 0.15$ with $f_m = (2/m\pi)\sin(mDc\pi)$]. The periodic OSL LT sample with 20 mm in length and 0.5 mm in thickness was fabricated using the conventional electrical poling technique. Figures 1a and 1b are the optical micrograph of +C and –C surface of the PPLT sample. One can see from the sample that the inverted domain distribution is uniform on the +C/–C surface and the duty cycle is uniform. Here the end face of the LT sample was polished but no antireflection coating was used.

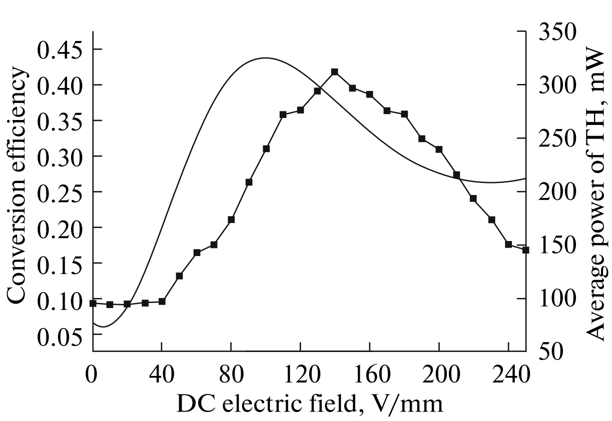

Fig. 3. Measured DC electric field tuning curves for THG. The black solid line represents the theoretical result and the blue square represents the experimental result.

The schematic view of experiment setup is shown in Fig. 2. The input source is a laser-diode-pumped (LD-pumped), Q-switched, 1342 nm Nd:YVO_4 laser. The resonant cavity of the laser is composed of two mirrors, one mirror with high reflection at 1342 nm and high transmission at 809 nm is coated on the gain crystal's front surface, the other with reflectivity R = 92% at 1342 nm is coated on the output coupler. To suppress the loss, the output end face of the Nd:YVO_4 crystal has a high transmission at 1342 nm. In this system, an acousto-optical Q-switch is laid in the cavity, which generated pulse with duration of 90 ns at a repetition rate of 5 kHz. The fundamental light was polarized and focused on the LT sample, which propagates in the x direction with its polarization along the z-axis. The focus length of the lens is 25 mm, and the diameter of beam waist inside the sample is estimated to be about 50 μm. The average fundamental power incident on the end face of the sample is 1.72 W, with the corresponding peak intensity about 194 MW/cm^2 at the beam waist. A heater (model CNi1633-C24, Omega Engineering Inc.) was used to heat the sample to the corresponding phase-matching temperature with an accuracy of 0.1°C. The efficiency of third harmonic was improved with a DC electric field E, which was applied along the z axis of the sample. The output blue light was separated with a filter and detected with a power meter (model EPM1000, Coherent Inc.).

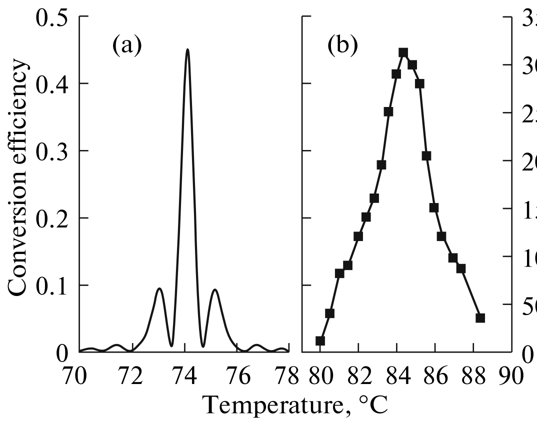

Fig. 4. (a) The theoretical temperature tuning curve of TH at phase-matching temperature. (b) The average output power of TH as a function of the temperature.

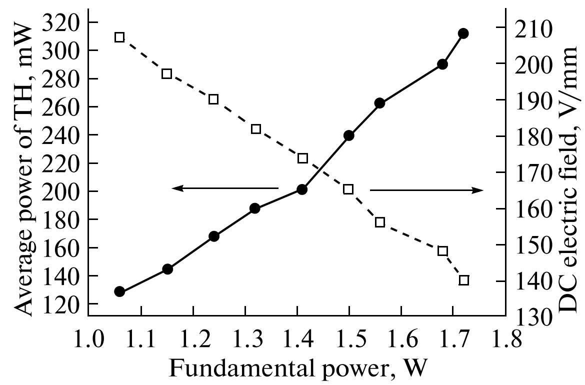

Fig. 5. Dependence of the improved average output of TH and the corresponding DC electric field on the fundamental intensity. Here, the solid circles represent the average output of TH, and the solid squares correspond to the required DC electric field.

3. EXPERIMENTAL RESULTS AND DISCUSSIONS

The coupling coefficient ratio of the designed sample $t = (f_a/f_b)(n_{30}/3n_{10})^{1/2}$ [10] is 1.781, which is larger than the theoretical critical ratio 0.8858 (the coupling coefficient ratio t is also 1.781 for the symmetric-duty-cycle $D_c = 0.5$). This means that in the absence of external electric field the SHG process is dominant and no efficient TH can be obtained with the pump intensity increasing. When an external electric field is applied to the sample, an additional phase mismatch will be introduced in the coupling process. In this case, the SHG process will be suppressed and the TH may be enhanced. Figure 3 shows the experimental results of the average power output of TH as a function of the DC electric field (where the temperature is set as 84.4°C). At the beginning, when the DC electric field is zero, the average output power of TH is only 95 mW, corresponding to a conversion efficiency 5.5% of the TH. This suggests that the QPM conditions satisfied at the predesigned working point are not most beneficial for the THG. With the electric field increasing, it can be seen that the efficiency of TH is significantly enhanced. When the DC field $E = 140$ V/mm, the efficiency of TH reaches its maximum 312 mW, which corresponds to a conversion efficiency of 18%. Considering the Fresnel reflection of fundamental and harmonic waves (~13%) on the front and end faces of the sample, the actual internal conversion efficiency of TH will be even higher than the above value. In addition, as the electric field continues to increase, the TH output is then decreased.

For comparison, we have also calculated the field-tuning curve for the TH using the coupling equations [16]. The result is shown as the solid line in Fig. 3. One can see that the theoretical result has a similar line-shape as that of experiment, thus revealing the underlying coupling behavior. According to the theory, two quasi-phase mismatches will be induced, due to the electro-optic effect, in the SHG and SFG processes. In contrast to the case of QPM conditions where the SHG is dominant (an efficient energy retransfer from the TH to SH exists), the introduction of phase-mismatches will greatly affect the coupling behavior. That is, the energy retransfers from the TH to SH will be suppressed and thus high efficient TH can be obtained instead. Since the quasi-phase mismatches are proportional to the DC electric field, the phase mismatches, the coupling process, and the efficiency of TH can be manipulated freely. It should be noticed that there are some discrepancies, such as the TH conversion efficiency and electric field drift, exist between the theory and experiment. These discrepancies may be caused by the following reasons. First, the theory is based on the plane-wave approximation. Nonetheless, the laser beam used in the experiment is actually a tightly focused Gaussian wave, where the Rayleigh length $z = \pi w^2/\lambda_1 \approx 24$ mm is comparable with the length of the OSL. In this case, the plane-wave model gives an overestimate of the conversion efficiency (the peak efficiency is up to 43%). Second, in the electric-poling process, the actual domain period and duty cycle may be slightly different from the initial setting values, thus resulting in a deviation of DC electric field from that predicted.

We also investigated the temperature-tuning curve of the third harmonic power near the phase-matching temperature. Figure 4 shows the experimental result (see the solid squares). By tuning the crystal temperature carefully, the temperature for the maximal third-harmonic generation was found to be 84.4°C ($E = 140$ V/mm), which deviates obviously from the expected working temperature 74.1°C. Note that the Sellmeier equation in [17] may not match the QPM wavelengths near 671 and 1342 nm with the same high precision as it does in the range 532–325 nm. This may cause some error in the design of domain period and the deviation of phase-matching temperature. The measured full width at half maximum is 3.2°C for TH, wider than the theoretical value of 0.6°C. The extension of phase-matching temperature range is related to the strongly focused fundamental beam used in the experiment.

In addition, by varying the fundamental intensity, we have studied the maximal output of TH and the required DC electric field. The results are shown in Fig. 5, where the solid circles represent the improved average output of TH, and solid squares correspond to the required DC electric field. Experimentally, when the fundamental power increases from 1.06 to 1.72 W, the maximal output of TH is improved from 126 to 312 mW (corresponding to a conversion efficiency from 12 to 18%. And correspondingly, the required DC electric field decreases from 205 to 140 V/mm.

4. CONCLUSIONS

Third-harmonic generation in a periodically poled LT has been experimentally studied, incorporating the electro-optic effect of OSL. By varying the external DC electric field, the quasi-phase mismatches, the coupling behavior, and the efficiency of TH can be manipulated. The experimental results demonstrated that, with a DC electric field of 140 V/mm, the average power output of TH can be enhanced from 95 to 312 mW, corresponding to a conversion efficiency from 5.5 to 18%). The method provides a simple and fast way for improving or tuning the high harmonic efficiency.

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