Simultaneously efficient blue and red light generations in a periodically poled LiTaO₃

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Generations of efficient blue light at 447 nm and red light at 671 nm were achieved by frequency doubling and tripling of a diode-pumped, Q-switched 1342 nm Nd:YVO₄ laser with a periodically poled LiTaO₃ (PPLT). The blue light at 447 nm was generated by sum-frequency mixing of the fundamental at 1342 nm with the generated second harmonic at 671 nm. The first-order and third-order reciprocals of the PPLT compensated the phase mismatches of second-harmonic and sum-frequency processes, respectively, making them quasiphase matched. The resulting averaged blue light power of 51 mW and red light power of 207 mW under the averaged fundamental power of 500 mW indicate that the PPLT may be used to construct an all-solid-state blue and red dual wavelength laser. © 2001 American Institute of Physics. [DOI: 10.1063/1.1371245]

The conventional approach to generate visible light from diode-pumped solid-state lasers is by frequency doubling of a near-infrared laser source with a laser host such as Nd:YVO₄ or Nd:yttrium–aluminum–garnet (YAG) using a nonlinear optical crystal by birefringence phase matching. Recently, optical superlattices, such as periodically poled LiNbO₃, periodically poled LiTaO₃ (PPLT), and periodically poled KTiOPO₄, have become attractive materials for second-harmonic generation (SHG) by quasiphase-matched (QPM) scheme. It was reported that second-harmonic green and blue generations from the diode pumped 1064 nm and 946 nm Nd:YAG lasers had been realized using optical superlattices, respectively.

Nd:YVO₄ is of great laser performance and can be produced into single crystal with adequate size. The spectrum of Nd:YVO₄ demonstrates a strong emission of the 4F₃/₂ → 4I₁₃/₂ transition of Nd³⁺ active ions at 1342 nm with polarization along the z axis. The wavelength has been frequency doubled to 671 nm to generate red light by using nonlinear optical crystal such as KTiOPO₄ and LiB₃O₅. As a matter of fact, it is also highly estimated for the generation of blue light at 447 nm by frequency tripling. However, one has not found a proper nonlinear crystal available for this aim up to now. Here we report, an efficient blue light generation by frequency tripling of a Nd:YVO₄ laser with a PPLT crystal.

Conventionally, an efficient third-harmonic generation (THG) is achieved by two steps in two nonlinear crystals, respectively, the first one for SHG and the second one for sum frequency (SFG). The first efficient THG from a single nonlinear crystal was experimentally demonstrated in our early work by using a Fibonacci superlattice. The basic principle behind the demonstration is well understood, that is, two reciprocals provided by a quasiperiodic superlattice compensated the wave vector mismatches of SHG and SFG, respectively, which led to a continuous energy transfer from fundamental to second and third harmonic by cascaded QPM processes. The basic building blocks and sequence in a quasiperiodic structure are fairly flexible in design. This permits one to predesign two reciprocals required in any QPM frequency tripling process. Yet the periodic structure generally fails to simultaneously provide such two reciprocals because its reciprocals all equal integral multiples of 2π/λ. There exist, however, some exceptions. For some nonlinear crystals, there is some particular fundamental wavelength at which the wave vector mismatch of SFG coincidentally equals integral multiples of that of SHG. In this case, a periodic superlattice can provide two proper reciprocals participating in the QPM THG process. For example, Ref. 13 reported that a PPLN was employed for QPM frequency tripling a cw 3.54–3.62 μm CO laser, where the first-order reciprocal of the periodic structure simultaneously phase matches the SHG and SFG processes. In this letter we give another example: efficient blue and red lights were simultaneously generated in a PPLT by frequency doubling and frequency tripling of a 1342 nm Nd:YVO₄ fundamental source. The first- and third-order reciprocals of the periodic structure compensated the wave vector mismatches of SHG and SFG processes, respectively. Our results offers a possible route to simultaneous generations of high average powers at the blue (447 nm) and red (671 nm) using all-solid-state technology, therefore, is of importance in particular.

We first consider the SHG process in the PPLT. We choose the first-order reciprocal for the most efficient nonlinear conversion. The QPM condition in a collinear interaction is

\[
k_f - 2k_s - \frac{2\pi}{\Lambda} = 0,
\]

where \(k_f, k_s\) are the wave vectors of the fundamental and the second harmonic, respectively, and \(\Lambda\) is the period of the superlattice. Similarly, the QPM condition for the SFG process is...
The pump beam was focused into the Nd:YVO$_4$ wavelength of 1342 nm, which is the $4\text{th}$ line of Nd$^{3+}$. The fundamental source for the measurement was a Nd:YVO$_4$ crystal besides Nd:YVO$_4$, such as Nd:YAP in which the gain crystal was prepared by standard electric field poling technique. The faces of the sample were polished but no antireflection coating was provided. One can change matching wavelength within the range by adjusting the operating temperature and the period of superlattice. Figure 1 shows the effect of operating temperature on matching fundamental wavelength and corresponding period vs temperature.

$$k_i - k_s - k_f - m \frac{2\pi}{\Lambda} = 0,$$

where $k_i$ is the wave vector of the third harmonic. Because the first-order reciprocal has been used for SHG process, and the duty cycle of 0.5 forbids the choice of second-order reciprocal, we choose the third-order reciprocal ($m = 3$) for SFG process. Combining Eqs. (1) and (2), we have

$$3(k_i - 2k_f) = k_i - k_s - k_f,$$

where $k_{(f,s,t)} = 2\pi n_{(f,s,t)}/\lambda_{(f,s,t)}$, hence, we get

$$3(2n_s - 2n_f) = 3n_f - 2n_s - n_f,$$

where $n_s$, $n_t$, $n_f$ are the refractive indices of the third harmonic, second harmonic, fundamental, respectively, and they are dependent upon wavelength and temperature in terms of Sellmeier equation. The QPM conditions of SHG and SFG can be simultaneously satisfied at the fundamental wavelength of 1342 nm and the temperature of 74.1 °C by numerically evaluating Eq. (4). Substituting these values into Eq. (2) or Eq. (1), we get the period $\Lambda = 14.778 \mu$m.

The earlier calculation is based on the fundamental wavelength of 1342 nm, which is the $4F_{3/2} - 4I_{13/2}$ emission line of Nd$^{3+}$ ions in Nd:YVO$_4$ crystal. The calculation also indicates that phase matching wavelength for THG in a PPLT does not lie at a single point, but within a narrow range of wavelength. In fact, three parameters, phase matching wavelength, temperature, and period are closely related to each other. One can change matching wavelength within the range by adjusting the operating temperature and the period of superlattice. Figure 1 shows the effect of operating temperature on matching fundamental wavelength and corresponding period. The figure illustrates that the PPLT may be used for QPM blue light generation from other Nd-doped gain crystal besides Nd:YVO$_4$, such as Nd:YAP in which the $4F_{3/2} - 4I_{13/2}$ emission line of Nd$^{3+}$ ions is 1341 nm.

A PPLT sample with the nominal period of 14.778 $\mu$m was prepared by standard electric field poling technique. The sample was 0.5 mm thick and 12 mm long. The two end faces of the sample were polished but no antireflection coating was used. Figure 2 is the schematic experiment setup. The fundamental source for the measurement was a $Q$-switched, 1342 nm Nd:YVO$_4$ laser, pumped by a continuous laser diode (OPC-DO15-809) emitting near-infrared at 809 nm. The pump beam was focused into the Nd:YVO$_4$ crystal with a 35 mm focal length lens $F_1$. One mirror with high reflection at 1342 nm and high transmission at 809 nm was coated directly onto the input end-face of the Nd:YVO$_4$, and the other with reflectivity $R = 92\%$ at 1342 nm was coated on the output coupler as shown in Fig. 2. To suppress the loss, the output end face of the Nd:YVO$_4$ crystal had a high transmission at 1342 nm. The acousto-optical $Q$ switch in the cavity turned the output into quasicontinuous pulses, with the duration of $\sim 90$ ns and a repetition rate of 10 kHz. The focal length of the lens $F_2$ was 25 mm, and a waist spot approximate 50 $\mu$m in diameter inside the sample was estimated. The average fundamental power incident on the end face of sample was 500 mW, and the corresponding peak power was 0.56 kW. In view of the Fresnel reflection of about 13% on the front surface of the sample, the actual fundamental average power and peak power transmitted into the PPLT sample was about 430 mW and 0.48 kW, respectively, with the peak intensity of $\sim 24.4$ MW/cm$^2$ at the waist. The sample was heated in an oven (model OTC-PPLN-20, Super Optronics Lt.) to the corresponding phase-matching temperature with accuracy of 0.1 °C. The red and blue lights produced from the sample were filtered at output end with appropriate filters, respectively, and were detected with a power meter (model EPM1000, Moletron Lt.).

Figure 3 shows the experimental results of SHG and THG output powers as a function of temperature. The measured phase-matching temperatures for SHG and THG are 92.5 and 85.6 °C, respectively. The peak of the blue light lies on the shoulder of the curve of the red light. They do not overlap, and both of them deviate from the expected value of 74.1 °C. There are probably two causes leading to the discrepancies. First, the phase matching conditions involved in this work were derived in terms of the temperature-dependent Sellmeier equation in Ref. 14. The equation may not match the QPM wavelengths near 671 and 1342 nm with...
the same high precision as it matches in the range 532–325 nm. This may give rise to the design error of domain period, thus the peak shift and peak separation of blue and red. Second, there may be the fabrication error of domain pattern. Owing to the limit of photographic precision, the actual domain period may be different from the nominal one. Since the matching temperature is sensitive to the period, a tiny deviation from preset value will cause great shifts of the phase matching temperatures of SHG and SFG processes resulting in the separation. The measured bandwidth in Fig. 3 are approximate 10 °C for SHG and 3.5 °C for THG, wider than the theoretical values of 6.0 and 1.5 °C, respectively. The widening of the curves may result from the strong focusing of fundamental wave and the decrease of the effective length such as missing domains or unpoled regions.

The conversion efficiency is defined as \( \eta = P_s / P_f \), where \( P_s \) is the average power of red or blue lights, and \( P_f \) is the average power of fundamental of 1342 nm incident on the crystal. As shown in Fig. 3, the measured maximum average powers are 51 mW for blue and 207 mW for red light with the efficiencies of 10.2% and 41.4%, respectively. Taking into account the Fresnel reflections from the front and rear faces of the crystal, the maximum internal conversion efficiencies are greater than above values. At the temperature of 85.6 °C where THG peak locates, SHG output power is 48 mW. At this point, blue and red lights are on the same power level. The fluctuation of power of blue light at its peak was less than 2% throughout the measurement period. No obvious degradation of the output power or beam quality is observed during the period, indicating that photorefractive effect is negligible under the experimental conditions.

The experiment results are fairly satisfactory, and there lies great potential in the increase of blue light power. Theoretically, a ~40% conversion efficiency of THG can be realized in a PPLT. Two methods are under way. The first is to coat end faces with high reflection at 671 nm to let the second harmonic come and go in the sample, which can increase the effective interaction length of sum-frequency process, generating brighter blue light. The second is to rectify the parameters of the Sellmeier equation and to improve the precision of lithograph to adjust the phase-matching temperatures of SHG and SFG, making them close up. In this case, the output of THG will be expected to increase greatly because of the participation of more photons of 671 nm in SFG process.

In summary, utilizing the coincidence that phase mismatch of the SFG process equals three times that of the SHG process in LiTaO₃ crystal, a PPLT with a nominal period of 14.778 μm was designed and prepared to realize the frequency doubling and frequency tripling of the 1342 nm output of Nd:YVO₄ laser. Efficient blue light at 447 nm and red light at 671 nm were obtained simultaneously. The highest average output powers were 51 mW for blue and 207 mW for red, corresponding efficiencies were 10.2% and 41.4%, respectively. Compared with the quasiperiodic structure, the periodic structure has larger effective nonlinear coefficient, and the fabrication of sample is more convenient. The con- ciseness of structure and high conversion efficiency make this scheme very attractive to obtain compact dual wavelength laser of red and blue. Further work to obtain higher power and efficiency of blue laser is under way.

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