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Quasi-Cw Ultraviolet Generation in a Dual-periodic LiTaO$_3$ Superlattice by Frequency Tripling

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We propose a dual-periodically domain-inverted structure for realizing an efficient third-harmonic generation at an arbitrary fundamental wavelength. The structure allows to couple two separate optical parametric processes, frequency doubling and frequency adding, through a cascaded quasi-phase-matched interaction, therefore, generate third-harmonic directly from a superlattice sample. As an example, we designed and fabricated such a structure in a LiTaO$_3$ crystal. Using a LD pumped 1.064 μm quasi-cw Nd:YVO$_4$ laser as a fundamental source, considerable ultraviolet at 355 nm was generated from this superlattice by frequency tripling. And green at 532 nm was simultaneously obtained by frequency doubling.

KEYWORDS: QPM; frequency conversion, Third-harmonic generation LiTaO$_3$ crystal UV laser

Optical parametric processes are of importance for creating laser sources at new frequency bands and have been developed into a fascinating, widely applied field in nonlinear optics. With the development of domain-inversion techniques in LiNbO$_3$, LiTaO$_3$ and other ferroelectric crystals,[1–3] the quasi-phase-matching (QPM) optical-frequency-conversion has been very successful in recent years.[4–6] The superlattice with periodically domain-inverted structure has been widely used to achieve a single optical parametric process such as a second-harmonic generation (SHG),[7] a difference-frequency generation (DFG)[8] and a sum-frequency generation (SFG)[9] in a QPM scheme. The creation of the third harmonic directly from the low third-order optical nonlinearity of crystals is of little practical value. Conventionally, an efficient third-harmonic generation (THG) can be realized using two periodic superlattices in series, the first for SHG, and the second for SFG.[10] The QPM conditions for SHG and SFG are fulfilled in two separate superlattices, respectively. Recently some papers have confirmed THG can also be realized in a single superlattice. Zhu and his co-workers reported this kind of work in a Fibonacci superlattice in LiTaO$_3$ crystal.[5] Since then, several other sequences, such as Thue-Morse sequence,[11] intergrowth sequence,[12] and aperiodic sequence[13] etc have also been proposed to construct superlattices to obtain the required QPM conditions in a multi-wavelength SHG or a cascade THG process.

Compact solid-state laser sources in the ultraviolet (UV) spectral region are currently finding a burgeoning range of applications in, such as, spectroscopy, medical diagnostics, biomolecular and biomedicine research, and optical storage. Frequency doubling or tripling of a solid-state laser pumped by a near-infrared laser diode in QPM scheme is a feasible approach to obtain cost-effective UV sources with salable power. The advantage of QPM approach is that noncritical phase matching can be used at any wavelength with the transparency range of the nonlinear material by appropriate choice of the period for the domain inversion. Besides, the largest nonlinear coefficient of the material $d33$ can also be utilized in the scheme. In this paper we report a novel dual-periodic optical superlattice, which can realize THG directly by a coupled parametric process at any fundamental wavelength. Using this kind of superlattice, a feasible scheme to construct an all solid-state UV 355 nm laser is provided.

QPM process can be well understood in wave vector space. The reciprocal vectors provided by a kind of superlattice structure may be used to compensate the phase mismatching of optical parametric process, thus make the process quasi-phase-matched. The third harmonic is generated by coupling SHG process and SFG process in a quadric nonlinear medium, therefore, the QPM condition of THG in a superlattice for a collinear interaction is

$$\Delta k_1 = k_2 - 2k_1 - G_{m,n} = 0 \tag{1}$$

for SHG and

$$\Delta k_2 = k_3 - k_2 - k_1 - G_{m',n'} = 0 \tag{2}$$

for SFG, respectively, where k_1, k_2, and k_3 are the wave vectors of the fundamental, second-, and third-harmonic fields, and $G_{m,n}$ and $G_{m',n'}$ are two pre-designed reciprocal vectors of the superlattice.

The dual-periodic structure can be described with a periodic phase-reversal sequence superimposed upon another smaller periodic structure,[14] as shown in Fig. 1. If we define the two periodic modulated sequences as $f_1(x)$ and $f_2(x)$, respectively, they can be described by the Fourier series respectively as

$$f_1(x) = \sum_{m=-\infty}^{\infty} g_m e^{-iG_m x}$$

$$f_2(x) = \sum_{n=-\infty}^{\infty} g_n e^{-iG_n x}. \tag{3}$$

Therefore, the dual-periodic structure can be given as

$$f(x) = f_1(x)\, f_2(x) = \sum_{m,n=-\infty}^{\infty} g_m g_n e^{-i(G_m+G_n)x}$$
$$= \sum_{m,n=-\infty}^{\infty} g_{m,n} e^{-iG_{m,n}x} \tag{4}$$

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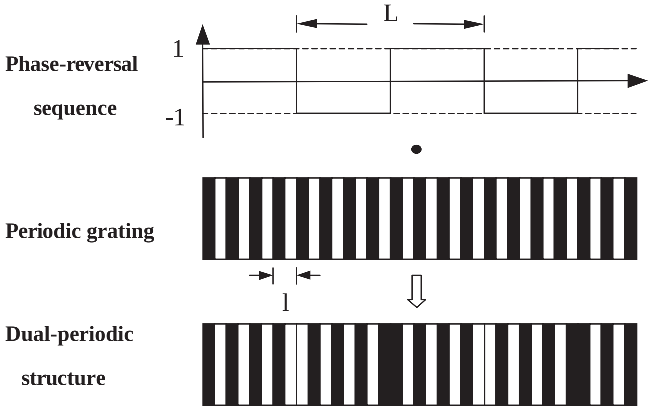

Fig. 1. Dual-periodic QPM structure formed by twice periodical phase-reversal modulating.

where $G_{m,n} = G_m + G_n$, and $g_{m,n} = g_m g_n$. The reciprocal vectors of this dual-periodic structure can be expressed as

$$G_{m,n} = G_m + G_n = mG_l + nG_L \tag{5}$$

where $G_l = 2\pi/l$ and $G_L = 2\pi/L$ are the first order reciprocal vectors of the periodic structure and the modulating sequence, and their periods are l and L, respectively (see Fig. 1). Integers m and n label the order of reciprocal vectors.

Using two reciprocal vectors of this structure to compensate the mismatching of SHG and SFG in a THG process, respectively, we can get two equations according to the equations mentioned above

$$\begin{cases} G_{m,n} = mG_l + nG_L = \Delta k_1 = \dfrac{4\pi}{\lambda}(n_2 - n_1) = \dfrac{2\pi m}{l} + \dfrac{2\pi n}{L} \\ G_{m',n'} = m'G_l + n'G_L = \Delta k_2 = \dfrac{2\pi}{\lambda}(3n_3 - 2n_2 - n_1) = \dfrac{2\pi m'}{l} + \dfrac{2\pi n'}{L} \end{cases} \tag{6}$$

here, λ is the fundamental wavelength, n_1, n_2, n_3 are the refractive indexes at fundamental, second-harmonic and third-harmonic of nonlinear crystal, respectively. If the fundamental wavelength is given at a specific wavelength, select appropriate parameters of m, n, m', and n', the two main parameters of the dual-periodic structure (l and L) can be determined from the eqs. (6).

For THG of the fundamental wavelength $\lambda = 1.064$ m, we selected $LiTaO_3$ as nonlinear crystal for it is transparent to 280 nm. Indexes $m = 1$, $n = -1$, $m' = 3$, $n' = 1$, respectively. According to eqs. (6) and the Sellmeier equation of $LiTaO_3$ crystal in the reference 15, we had $l = 6.77\,\mu$m and $L = 50.86\,\mu$m, respectively. In this design, the reciprocal vector $G_{1,-1}$ was used for QPM SHG from the fundamental wavelength at 1064 nm, whereas $G_{3,1}$ was used for QPM SFG through adding1064 nm and the second harmonic of 532 nm. The Fourier spectrum of this dual-periodic superlattice is shown in Fig. 2.

We fabricated the superlattice with the dual-periodic domain reversal structure in a z-cut wafer of $LiTaO_3$ crystal through electric field poling at room temperature.[1] The sample's thickness was about 0.5 mm with a total length of 12 mm approximately. The dual-periodic domain patterns were observed on the etched $+c$, $-c$, and y surfaces of the sample using an optical microscope.

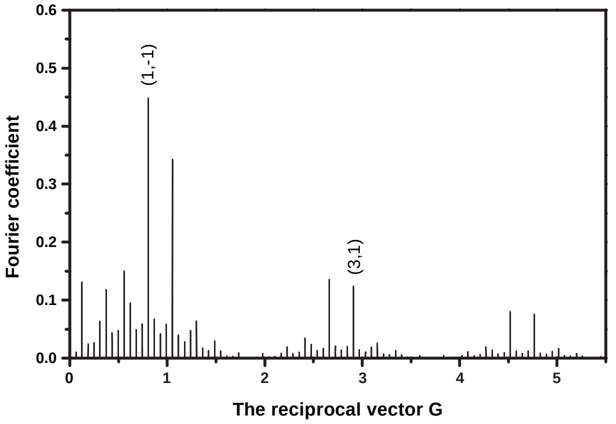

Fig. 2. The Fourier spectrum of the dual-periodic structure with the structural parameters $m = 1$, $n = -1$, $m' = 3$, $n' = 1$, $l = 6.77\,\mu$m, $L = 50.86\,\mu$m. The marked two reciprocals are used for SHG and SFG, respectively.

The schematic experimental setup used in our experiment is shown in Fig. 3. An infrared 1064 nm laser was generated from a Nd:YVO_4 crystal pumped by a 808 nm laser diode. Modulated by an acoustic-optical Q-switch, a quasi-cw laser with pulse duration of 150 ns and repetition rate of 13 kHz was achieved (see it in Fig. 4.). The fundamental wave was focused into a beam whose radius of waist was 0.2 mm around, coupled into the polished but uncoated end face of the sample by a focusing lens. The foci of the lens is about 50 mm. Both fundamental and harmonics were polarized along the z axis, transmitted along the x axis of the $LiTaO_3$ wafer. The sample was heated in an oven (Model OTC-PPLN-20, Super Optronics Ltd.) for tuning it to corresponding phase-matched temperature. The accuracy of the temperature controller is 0.1°C. The nonlinear optical features of the sample were characterized by measuring the powers of second harmonic and third harmonic versus phase-matched temperature and input power of fundamental.

Figure 5(a) shows the output of SHG and THG as a function of temperature. The phase-matched temperatures was located at 50.5°C for SHG and at 49.0°C for THG, respectively. Under the fundamental average power of 1.1 W, the maximal output power of SHG and THG are 95 mW and 3.6 mW, respectively. Owing to the Fresnel reflection of about 13% on

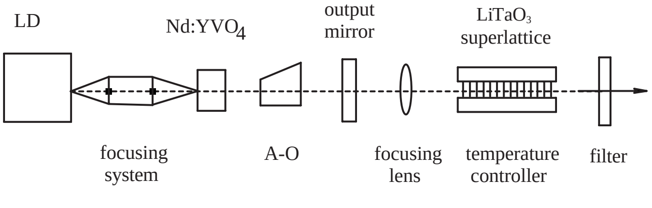

Fig. 3. Schematic setup for an all solid-state quasi-cw UV 355 nm laser.

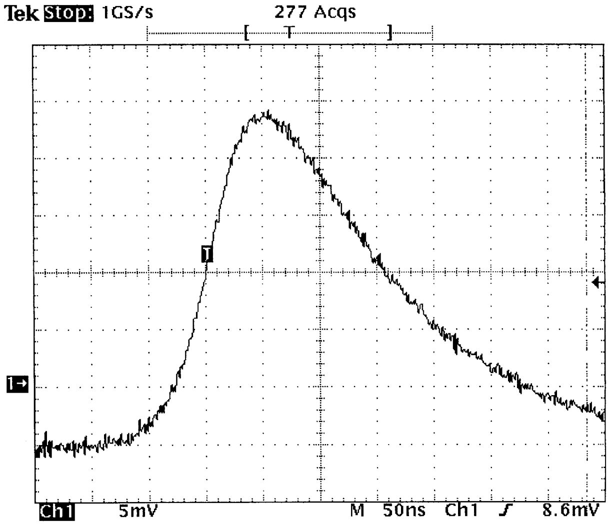

Fig. 4. The infrared 1064 nm pulses generated from the laser cavity measured with a fast photodiode. The pulse duration is about 150 ns with repetition rate 13 kHz.

the front surface of the sample, the actual fundamental average power transmitted into the sample was about 0.96 W. The fact that the peaks of SHG and THG do not overlap each other may originate from the pattern precision of lithographic mask or the deviation of the Sellmeier equation of the $LiTaO_3$ crystal or the both. Figure 5(b) shows that the SHG and THG power increase gradually with the increasing of fundamental power at the operating temperature of 49°C. When the fundamental average power was tuned to 1.56 W, about 10 mW UV was achieved. Because this temperature is not a perfect phase matching one for SHG, the SHG power at this fundamental power level is just 120 mW, lower than 300 mW at 50.5°C.

The measured efficiencies of SHG and THG are far below the theoretical value estimated by solving the coupled equations with fundamental depletion in Fig. 5(c). The reasons may be as follows:

The first, the peak temperatures of SHG and THG did not overlap each other, showing that the QPM conditions of frequency doubling and frequency adding are not perfectly satisfied at the same time in the measured sample. At the peak temperature of THG, the efficiency of SHG was around 8% for the measurement condition. Because the efficiency of sum-frequency is in proportion to the intensity of second harmonic, which leads to sharp decrease of THG efficiency from expected one. Secondly, the phase matched temperature for the superlattice is 50°C. The photorefractive effect in short wavelength region was still strong for $LiTaO_3$ crystal at this temperature, which resulted in the changes of refractive indexes and the degradation of laser beam quality in crystal directly, therefore, the phase mismatching and the decrease of conversion efficiency. In order to avoid the influence of photorefractive effect, it is necessary to redesign the structure parameters and make the phase-matched temperature of the new structure higher than 150°C. In addition, the non-perfect poling, including uneven, deviation duty cycle from designation, may result in the rapid reduction in conversion efficiency.

We have proved that the conversion efficiency of harmonic waves might be increased sharply with the fundamental power in a dual-periodic superlattice. Theoretically, the average output power of THG may reach 200 mW, with 1.5 W average fundamental power, for a 1.2 cm length dual-periodic superlattice. Therefore, this scheme to realize an all-solid-state UV laser, especially UV and green bicolor laser, is considerably valuable and promising. More accurate parameter design, perfect sample preparation and elevated operating temperature would be very important to improve efficiency up to a usable and salable level.

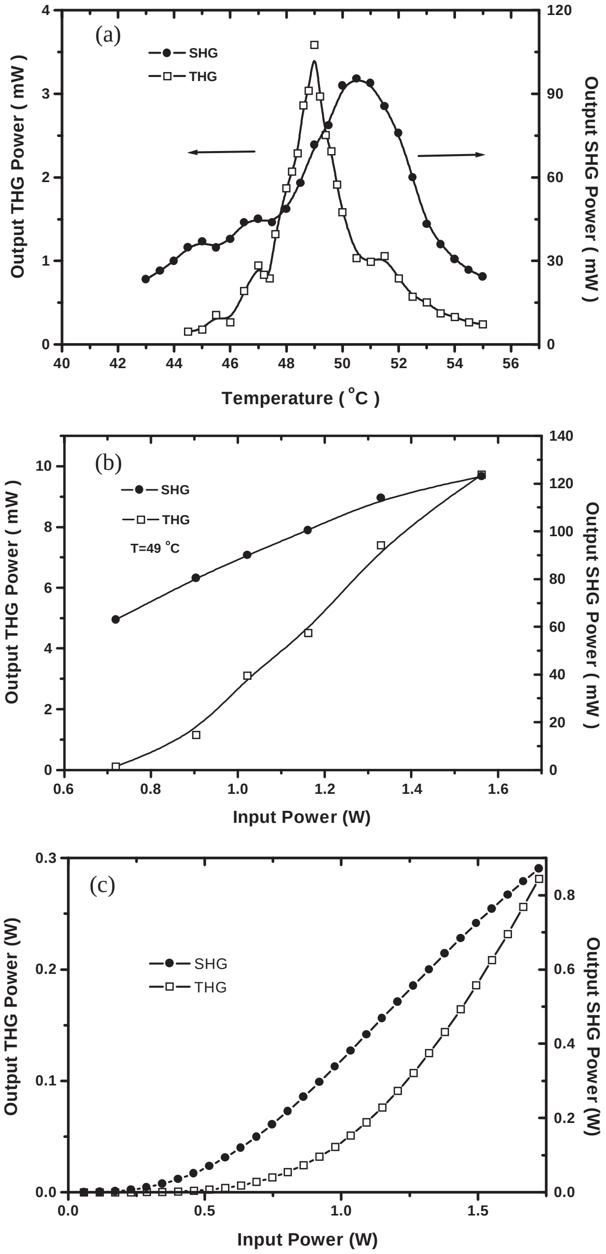

Fig. 5. (a) The average powers of second- and third-harmonic wave versus the tuned temperature. The average power of fundamental wave is 1.1 W. (b) The average power of second- and third-harmonic wave versus the average power of fundamental wave. The operating temperature is constant at 49°C. (c) Theoretical average SHG and THG output power versus average fundamental wave power in our experimental circumstance. Length of the superlattice is 12 mm. The duty cycle is 0.5.

In conclusion, a novel dual-periodic superlattice used in coupling optical parametric process is presented. We have designed and fabricated one kind of this dual-periodic domain reversal structure in $LiTaO_3$. The structure realized a third harmonic generation at 355 nm by tripling an all solid-state quasi-cw 1064 nm Nd:YVO$_4$ laser in a cascaded QPM scheme. Theoretically, the scheme has a higher efficiency and a simpler light path for frequency tripling than a traditional one using two bulk crystals or periodic superlattices in series. Owing to its wider feasibility for material design, the dual-periodic superlattice may have potential applications on the UV lasers and other frequency conversion devices.

Acknowledgements

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