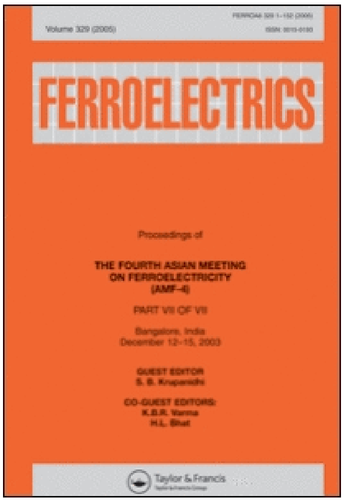

Ferroelectrics
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713617887

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Online Publication Date: 01 February 2001

To cite this Article Liu, Zhaowei, Qin, Yiqiang, Zhu, Yongyuan, Wang, Huitian, Zhang, Chao, Zhu, Shining and Ming, Naiben(2001)'Ultraviolet generation in a dual-periodic domain inverted structure in $LiTaO_3$ crystal by frequency tripling a 1.064 m laser',Ferroelectrics,253:1,263 — 270

To link to this Article: DOI: 10.1080/00150190108008467

URL: http://dx.doi.org/10.1080/00150190108008467

Ferroelectrics, 2001, Vol. 253, pp. 263-270
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Ultraviolet Generation in a Dual-Periodic Domain Inverted Structure in $LiTaO_3$ Crystal by Frequency Tripling a 1.064 μM Laser

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(Received June 2, 2000)

We demonstrate a coupled optical parametric process that is able to realize in a dual-period domain inverted structure in complex quasi-phase-matched scheme. As an example, we designed and fabricated a dual-periodic domain inverted structure in $LiTaO_3$ for the first time. This structure couples second-harmonic generation and sum-frequency generation through two cascade quasi-phase-matched processes, therefore, a direct third-harmonic generation can be realized. The test verified that ultraviolet was generated in the optical superlattice through frequency tripling a 1.064 μm laser.

Keywords: dual-periodic domain inverted structure; QPM; $LiTaO_3$

INTRODUCTION

With the development of domain-inversion techniques in lithium niobate ($LiNbO_3$), lithium tantalate ($LiTaO_3$) and other ferroelectic crystal,[1,2,3] the optical-frequency-conversion has been very successful in recent years.[4,5,6]

The periodic domain inverted superlattice has been widely used to single optical parametric process such as second-harmonic generation (SHG), difference-frequency generation (DFG) and sum-frequency generation (SFG) in quasi-phase-matching (QPM) scheme. QPM process can be well understood in wave vector pace. The reciprocal provided by the structure may be used to compensate the phase mismatching of optical parametric process, thus make the process quasi-phase-matched. The creation of the third harmonic directly from the low third-order optical nonlinearity is of little practical value. Conventionally, an efficient THG can be realized using two periodic superlattices in series, the first for SHG, and the second for SFG. The QPM conditions of two different processes are fulfilled in two superlattices, respectively. In 1997, Zhu et al. reported a direct THG in the quasi-periodic Fibonacci superlattice in $LiTaO_3$ crystal. [5] Some more complex sequences, such as Thue-Morse sequences, [7] intergrowth sequences, [8] and aperiodic sequences [9] etc. can also be chosen to construct superlattices to obtain the required QPM conditions in a single parametric optical parametric process or in a coupled parametric process.

$LiTaO_3$ crystal is transparent to 280 nm and is therefore more suitable for nonlinear optical interactions in mid UV than $LiNbO_3$ crystal which is transparent to 330 nm. A Nd:YAG laser is now commercially available and its output of 1064 nm is the most popular laser source. Theoretical analysis verifies a direct tripling 1.064 nm cannot be realized with high efficiency in a standard Fibonacci superlattice as Zhu has done in Reference [5]. In order to solve this problem, we design a novel dual-periodic domain reversal structure. The structure can satisfy the QPM conditions of SHG and SFG simultaneously at any wavelength, therefore, it can realize the QPM third harmonic generation at any given wavelength. It has wider feasibility for material design than a standard Fibonacci sequences.

In this paper we report THG down to 355 nm through tripling a 1064 nm laser using the dual-periodic superlattice in $LiTaO_3$ crystals. The superlattice sample was fabricated using the electric field poling technique. Experimental, the second harmonic of 532 nm and the third harmonic of 355 nm were proved with an yttrium-aluminum-garner

(Nd:YAG) laser as fundamental optical source.

FUNDAMENTAL THEORY

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 predesigned reciprocal vectors of the superlattice.

One question is naturally raised how can we pinpoint the structure that is able to provide the two precise reciprocal vectors simultaneously if the fundamental wavelength was given in advance. Here we give a solution through using a dual-periodic superlattice.

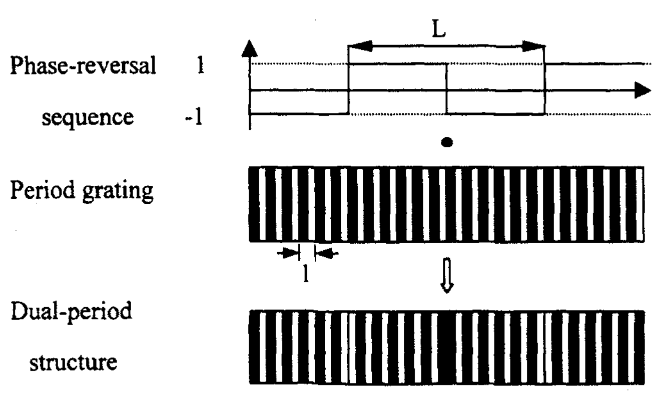

FIGURE 1 Dual-period QPM structure formed by superimposition of a phase-reversal grating upon a periodic grating.

The dual-periodic structure can be described with a periodic phase-reversal sequence superimposed upon another small periodic structure, [10] as shown in Fig.1. The Fourier transform of the new modulated structure can be viewed as the convolution of a series of δ function (a FT of the period structure) and a comb function (a FT of the modulating sequence wave). Then the reciprocal vectors of this dual-periodic structure can be expressed as:

$$G_{m,n} = mG_1 + nG_2 \tag{3}$$

where $G_1 = 2\pi / l$ and $G_2 = 2\pi / L$ 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.). m and n are integers, which 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 as:

$$\begin{aligned} G_{m,n} &= mG_1 + nG_2 = \Delta k_1' = \frac{4\pi}{\lambda}(n_2 - n_1) \\ G_{m',n'} &= m'G_1 - n'G_2 = \Delta k_2' = \frac{2\pi}{\lambda}(3n_3 - 2n_2 - n_1) \end{aligned} \tag{4}$$

here, λ is the fundamental wavelength, n_1, n_2, n_3 are the refractive indexes of at fundamental, second-harmonic and third-harmonic of $LiTaO_3$ crystal, respectively. If the fundamental wavelength is given at a specific wavelength, the two main parameters of the dual-periodic structure (l and L) can be determinate from the equations given above. The detail discussion on the design of the dual-periodic structure in terms of a complex QPM theory will be presented in another paper.

For Nd:YAG laser, the output wavelength λ=1.064 μm, we select indexes m=1, n= -1, m'=3, n'=1, respectively. According to Eq.(4) and the Sellmeier equation of $LiTaO_3$ crystal, [11] the reciprocal vectors of two periodic structure are $G_1 = 9.28\times10^5$/m, $G_2 = -1.23\times10^5$ /m, respectively, thus we have l =6.77 μm and L=51.08 μm, respectively. In this design, the reciprocal vector [see Fig. 2.] $G_{1,1}$ was used for QPM SHG from the fundamental wave of 1064 nm, whereas $G_{3,-1}$ was used for QPM SFG through adding1064 nm to the second harmonic of 532 nm. In this case, the two processes no longer proceed alone but couple each other. This

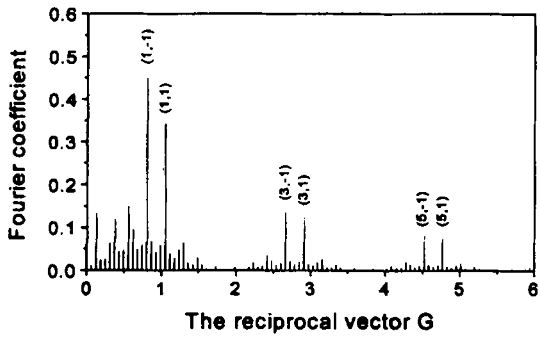

FIGURE 2 The fourier spectrum of the dual-periodic structure with the structural parameters: l=6.77 μm, L=51.08 μm. The main peaks are indexed.

coupling is able to led to a continuous energy transfer from fundamental to second- to third-harmonic fields, and thus a direct THG was generated from the dual-periodic superlattice.

EXPERIMENT

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 two periods, the basic period *l* and phase-reversal period *L* is 6.77 μm and 51.08 μm, respectively. The sample's thickness was about 0.5 mm with a total length of 8 mm approximately. The dual-periodic domain pattern can be directly confirmed by observing the etched *y* surface of the sample. The nonlinear optical features of the sample were characterized by measuring second harmonic generation and third harmonic generation versus phase-match temperature. The fundamental source is a Nd:YAG laser with the output wavelength of 1.064 μm, the line width of ~ nm and the pulse duration of 43 picosecond(ps), respectively. Both fundamental and harmonics were polarized along the z axis of the $LiTaO_3$ wafer. The sample was heated in an oven (Model OTC-PPLN-20, Super Optronics Lt.) for tuning it to

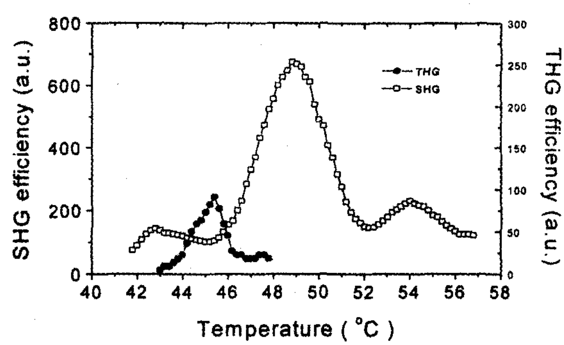

FIGURE 3 The average powers of second- and third-harmonic fields versus the tuned temperature

corresponding phase-matching temperature. The oven was monitored by a temperature controller with an accuracy of 0.1°C. The fundamental wave was weakly focused into a beam whose radius of waist was 0.1 mm around, coupled into the polished end face of the sample and transmitted along the x axis of sample. The output power of second harmonic and third harmonic of the sample were measured in the temperature range from 42°C to 57°C [see Fig. 3.] respectively.

RESULTS AND DISCUSSION

Figure 3 shows the output of SHG and THG as a function of temperature. The phase-matching temperatures locate respectively at 48.9°C for SHG and at 45.4°C for THG, respectively, shifting off the designed value of 40°C. We obtained SHG green and THG ultraviolet output with the maximum conversion efficiencies up to 33% and 0.54% under the fundamental power of ~ mW at these two temperatures, respectively. The efficiency of THG is far below the theoretical estimation by solving the coupled equations with fundamental depletion. The fact that the peaks of SHG and THG do not overlap each other shows that the QPM conditions, frequency doubling and frequency adding, are not satisfied at the same time in the measured sample, which lead to the decrease of THG efficiency. Figure 3 shows that the peak of third harmonic locates the shoulder of peak of SHG, where the practical conversion efficiency of SHG is only 1.6%. As there is not enough second harmonic participating in the sum frequency, the corresponding THG can not reach a considerable level. The discrepancy of phase-matching temperature between the calculated and measured values might originate the deviation of Sellmeier equation or the fabrication error of electrode pattern. In Fig 3, the bandwidths of the phase-matching curves are respectively 3.6°C for SHG and 1.6 °C for THG that are close to the values of theoretical prediction. Like in a periodic superlattice, the bandwidth of phase matching in a dual-periodic superlattice decreases with the increase of the effective length of superlattice. Longer sample, narrower temperature bandwidth and higher frequency conversion efficiency.

According to Eq.(4), the position of efficiency peak critically depends on the refractive indexes and their temperature coefficients at three wavelengths. The temperature asynchronous explains the dissatisfactory precision of the refractive index equation mainly because of the poignant variance of refractive index in ultraviolet and possible photorefractive effect of the measured crystal. If we can rectify the constant of the refractive index equation exactly, the matched temperature of SHG and THG may be modulated to the same point, then the output of THG should be considerable.

CONCLUSION

We have designed and fabricated a novel optical superlattice with a dual-periodic domain reversal structure. The structure can realize a third harmonic generation, through the quadric nonlinearity of crystal, at any given wavelength in complex QPM scheme. It has a higher efficiency for frequency tripling than the traditional scheme using two periodic superlattices in series. Along with its wider feasibility for material design, the dual-periodic superlattice shows a potential application on the high-order harmonic generations of laser.

Acknowledgements

This work is supported be a grant for the state Key Program for Basic Research of China, by the National Advanced Materials Committee of China, and by the National Natural Science Foundation of China (69938010). S.N.Zhu also thanks the support from a mono-grant RFPP.

References

[1] S.N.Zhu, Y.Y.Zhu, Z.Y.Zhang, H.Shu, H.F.Wang, J.F.Hong, C.Z.Ge, and N.B.Ming, *J. Appl. Phys.* **77**,5481 (1995).
[2] V.Ya.Shur, E.L.Rumyantsev, E.V.Nikolaeva, E.I.Shishkin, D.V.Fursov, R.G.Batchko, L.A.Eyres, M.M.Fejer, and R.L.Byer, *Appl. Phys. Lett.* **76**, 143 (2000).
[3] G.Rosenman, A.Skliar, D.Eger, M.Oron, and M.Katz, *Appl. Phys. Lett.* **73**, 3650 (1998).
[4] Xinghua Gu, Maxim Makarov, Yujie J. Ding, Jacob B. Khurgin, and William P.Risk, *Opt. Lett.* **24**, 127 (1999).
[5] S.N.Zhu, Y.Y.Zhu, and N.B.Ming, *Science.* **278**, 843 (1997).
[6] S.N.Zhu, Y.Y.Zhu, Y.Q.Qin, H.F.Wang, C.Z.Ge and N.B.Ming, *Phys. Rev. Lett.***78**, 2752 (1997).
[7] Zheming Cheng, Robert Savit, and R.Merlin, *Phys. Rev. B* **37**, 4375 (1988).
[8] Xianjie Liu, Zhenlin Wang, Xiaoshu Jiang, Jun Wu, and Naiben Ming, *J. Phys. D:Appl. Phys,* **31**, 2502 (1998).
[9] Ben-Yuan Gu, Bi-Zhen Dong, Yan Zhang, and Guo-Zhen Yang, *Appl. Phys. Lett.* **75**, 2175 (1999).
[10] M. H. Chou, K. R. Parameswaran, M. M. Fejer, and I. Brener, *Opt. Lett.* **24**, 1157 (1999).
[11] J. P. Meyn and M. M. Fejer, *Opt. Lett.* **22**, 1214 (1997).