Periodically domain-inverted LiNbO₃ for waveguide quasi-phase matched nonlinear optic wavelength conversion devices

M FUJIMURA*, T SUHARA and H NISHIHARA

Department of Electronic Engineering, Graduate School of Engineering, Osaka University, 2-1 Yamada-Oka Suita, Osaka 565-0871, Japan

Abstract. The authors' work on LiNbO₃ waveguide quasi-phase matched nonlinear optic wavelength conversion devices is reviewed. The requirements of domain-inverted gratings for efficient wavelength conversions are shown by theoretical analysis of the device performances. The fabrication apparatus of the domain-inverted gratings is described, and it is shown that the fabricated gratings satisfy the requirements. A variety of LiNbO₃ waveguide quasi-phase matched (QPM) wavelength conversion devices were fabricated and examined. Second-harmonic green/blue/ultraviolet light generation with normalized conversion efficiencies of ~100%/W, third-harmonic generation by cascading second-harmonic generation and sum-frequency generation, and difference-frequency generation for wavelength conversion in infrared wavelength range are demonstrated.

Keywords. Lithium niobate; domain inversion; nonlinear optics; optical waveguides; quasi-phase matching; wavelength conversion.

1. Introduction

Compact coherent light sources are required in various wavelength ranges for applications in many fields, for example in high-density optical data storage, high-precision optical measurement, optical sensing, and so on. A waveguide nonlinear optic wavelength conversion device pumped by a laser diode is promising for such light sources. The device can be compact and efficient because high optical intensity is maintained for long interaction length in the waveguide. Phase-velocity matching between the interacting waves is required for high conversion efficiency. This is feasible by compensation of the mismatch using a periodic structure (Armstrong *et al* 1962; Suhara and Nishihara 1990). Such phase matching technique is called quasi-phase matching (QPM). The QPM technique offers the wavelength conversion through the largest second-order nonlinear optic tensor component at arbitrary wavelengths in the transparent range by appropriate design of the period. Efficient QPM wavelength conversion requires periodic modulation of the second-order nonlinear optic coefficient, which can be obtained by periodic inversion of the ferroelectric domain.

There are many nonlinear optic materials studied for the waveguide QPM wavelength conversion devices. Lithium niobate (LiNbO₃) is studied most extensively because of the large nonlinear optic coefficient ($d_{33} = 34$ pm/V) and well-established waveguide fabrication techniques. LiNbO₃ waveguide QPM second-harmonic generation (SHG) devices with high conversion efficiency have been

reported so far (Yamada *et al* 1993; Kintaka *et al* 1996a; Webjorn *et al* 1997). Efficient devices have been implemented also in lithium tantalate (LiTaO₃) (Mizuuchi *et al* 1994; Yi *et al* 1996) and in potassium titanil phosphate (KTiOPO₄) (Eger *et al* 1994). Preliminary experiments have been carried out in organic materials (Suhara *et al* 1993), semiconductor materials (Yoo *et al* 1996), and glass materials (Kazansky *et al* 1997).

In this paper, our work on LiNbO₃ waveguide QPM wavelength conversion devices is reviewed. In § 2, the coupled mode analysis shows the requirement of the ferroelectric domain-inverted grating for efficient QPM wavelength conversion. In § 3, a domain inversion technique which provides such gratings is described. In § 4, a variety of waveguide wavelength conversion devices are demonstrated, and in § 5, conclusions are given.

2. Description and analysis of waveguide QPM wavelength conversion devices

Second-harmonic generation (SHG) device is a basic device in nonlinear optic wavelength conversion devices. Figure 1 shows a LiNbO₃ waveguide QPM-SHG device. On a z-cut LiNbO₃ single crystal, an optical channel waveguide with a cross-section of ~10 µm² and a ferroelectric-domain-inverted grating are integrated. The sign of the second-order nonlinear optic coefficient, d coefficient, inverts in the domain-inverted regions, and therefore the periodic sign-modulation of d coefficient is obtained in the device by the domain-inverted grating. A pump wave with wavelength of λ_0 is coupled with the waveguide at one facet, and a second-harmonic wave

*Author for correspondence

with wavelength of $\lambda_{2\omega}$ is obtained from the other facet. The pump and the SH waves are both TM-polarized. The SHG interaction takes place through the largest d coefficient, d_{33} .

The QPM-SHG in the waveguide along x axis is described by the following coupled mode equations (Suhara and Nishihara 1990):

$$\frac{d}{dx}A(x) = -j\kappa^* B(x)A(x)^* \exp(-j2\Delta x), \quad (1a)$$

$$\frac{d}{dx}B(x) = -j\kappa [A(x)]^2 \exp(+j2\Delta x), \quad (1b)$$

$$2\Delta = \beta_{2\omega} - 2\beta_{\omega} - 2\pi/\Lambda, \quad (1c)$$

$$\kappa = \frac{2\omega}{4} \iint [E_{2\omega}]^* d_1(y, z) [E_{\omega}]^2 dy dz, \quad (1d)$$

and

$$d_1(y, z) = \frac{2d_{33}}{\pi} \sin[a(y, z)\pi]. \quad (1e)$$

In the above equations, A and B represent the complex amplitudes of the pump and the SH waves, respectively. Δ is a phase mismatch parameter, κ the nonlinear coupling coefficient, and E_{ω} and $E_{2\omega}$ represent normalized mode profiles of the pump and the SH waves, respectively. The QPM grating is described by the period Λ , the fundamental Fourier component d_1 , and the duty cycle $a(y, z)$, which is a ratio of the grating line width to the period Λ . When the conversion efficiency is so low that the pump depletion is small, the SHG efficiency η is given by

$$\eta = \kappa^2 P_0 L^2 [\sin(\Delta L)/\Delta L]^2, \quad (2)$$

where P_0 is the incident pump power. Since η is pro-

portional to P_0 , a normalized conversion efficiency $\eta_{\text{norm}} = \eta/P_0$ is used to represent the device performance.

The requirement of the QPM grating structure for efficient SHG is discussed using (1) and (2). The expression for the grating period Λ_{QPM} can be derived by setting $2\Delta = 0$ in (1c) as

$$\Lambda_{\text{QPM}} = \lambda_{\omega}/2 (N_{2\omega} - N_{\omega}), \quad (3)$$

where N_{ω} and $N_{2\omega}$ represent effective indices of the waveguide. The dependence of Λ_{QPM} on the pump and the SH wavelengths was calculated and is shown in figure 2. The refractive indices for extraordinary waves in bulk LiNbO₃ were used as approximations for N_{ω} and $N_{2\omega}$ in the calculation. The QPM period is $\sim 6 \mu\text{m}$ for green light generation and $\sim 3 \mu\text{m}$ for blue light generation.

The SHG efficiency reduces when the grating period deviates from the phase-matched value. The dependence of relative SHG efficiency on the period error was calculated using (1c) and (2). The acceptance full-width half-maximum $\delta\Lambda$ is inversely proportional to the interaction length L and is about 11 nm for a green light generation device with $L = 3 \text{ mm}$. The optimum duty cycle is $a(y, z) = 0.5$ as seen from (1e). This is satisfied when each grating line has rectangular cross-sectional structure with the width of $\Lambda/2$. The dependence of relative SHG efficiency on the depth of the inverted region was calculated using (1d) for the case of green light generation devices and is shown in figure 3. The guided mode profiles were measured in the experiments

Figure 1. A LiNbO₃ waveguide quasi-phase matched second-harmonic generation device.

Figure 2. The QPM grating period dependent on the pump and second-harmonic wavelengths.

and used in the calculations. Figure 3 shows that the depth should be deeper than the waveguide mode depth to obtain good overlap with the guided modes and high SHG efficiency. The above discussion is applicable to other wavelength conversion devices.

3. Fabrication of LiNbO_3 domain-inverted gratings

Fabrication of ferroelectric-domain-inverted grating is a crucial process for fabrication of efficient QPM wavelength conversion devices. The grating period is as fine as a few micron, and therefore requires accurate period control. Further the inverted region should be deep enough and have rectangular cross-section with duty cycle of 0.5.

Various inversion methods have been examined for fabrication of the domain-inverted gratings. (i) Ti diffusion method (Lim *et al* 1989, Ishigame *et al* 1991): Thermal diffusion of Ti into the +z surface of LiNbO_3 at $\sim 1000^\circ\text{C}$ causes domain inversion. The domain-inverted grating is obtained by diffusion of Ti stripes. (ii) Li_2O outdiffusion method (Webjorn *et al* 1989): Li_2O outdiffusion from LiNbO_3 crystal takes place when the crystal is heated to $\sim 1100^\circ\text{C}$. Then the domain inversion is induced on the +z surface. Since the SiO_2 thin film prevents the outdiffusion, the domain-inverted grating is obtained by heating the crystal with patterned SiO_2 mask.

Figure 3. Relative SHG efficiency dependent on D/T , where D is the depth of domain-inverted region and T the waveguide depth.

(iii) SiO_2 clad and heating method (Fujimura *et al* 1991): Heat treatment at $\sim 1000^\circ\text{C}$ of LiNbO_3 crystal with SiO_2 clad on the +z surface causes domain inversion underneath the clad. The domain-inverted grating is obtained by using a grating-patterned SiO_2 layer. (iv) Electron beam writing (Ito *et al* 1991; Fujimura *et al* 1993): Electron beam irradiation on the -z surface of LiNbO_3 crystal with +z surface grounded causes domain inversion. The inverted region continues from the -z surface to the +z surface. Scanning the electron beam on the crystal provides the domain-inverted grating. (v) External voltage application (Yamada *et al* 1993; Myers *et al* 1995; Kintaka *et al* 1996a). High voltage pulse application between a periodic electrode on the +z surface and a uniform electrode on the -z provides the domain-inverted grating. Cross-sectional structures of the domain-inverted gratings fabricated by the above methods are illustrated in figure 4. The inverted regions obtained by (i), (ii) or (iii) are shallow and triangular. The region obtained by (iv) is needle-like near the top surface. Despite several efforts efficient QPM wavelength conversion devices have not been implemented using the above-mentioned methods. Currently, external voltage application method is being used as the most effective method to fabricate efficient devices.

Figure 5 shows the external voltage application apparatus established by authors for fabrication of LiNbO_3 domain-inverted gratings (Kintaka *et al* 1996a). A uniform gold electrode on -z and a corrugation electrode on the +z surfaces of LiNbO_3 crystal are present. The crystal thickness is as thin as $0.15\ \mu\text{m}$ to obtain the grating with period less than $3\ \mu\text{m}$. The corrugation

method	cross section
Ti diffusion	
Li_2O out-diffusion	
SiO_2 clad & heat	
Electron Beam irradiation	
External voltage application	

Figure 4. Cross-sectional structures of the domain-inverted gratings fabricated by various inversion techniques.

electrode consists of an insulator layer, with the thickness periodically modulated, and an aluminium film covering the layer. The aluminium film corrugates and parts of the film contact the crystal directly. The external voltage pulse is applied between the two electrodes, and the strongest electric field is induced at the crystal surface contacting the aluminium film. The electric field can be larger than the coercive field, and domain inversion takes place. The applied voltage is about -4 kV. During the domain inversion, the domain-inverted regions grow toward the $-z$ surface, inversion current runs around the circuit and is accumulated by the charge monitor. When the total inversion charge increases to the double of the product of aimed inversion area and the spontaneous

Figure 5. Apparatus for fabrication of LiNbO_3 domain-inverted gratings by external voltage application.

polarization, the controller generates a signal to halt the voltage application.

Typical waveforms of the voltage pulse and the inversion charge are shown in figure 6. The pulse duration is ~ 300 μsec . The DC voltage of -2 kV is necessary to prevent the inverted domains reversing at the trailing edge of the pulse. Figure 7 is the SEM photographs of the fabricated domain-inverted grating of 2 μm period. The crystal was etched by a $\text{HF}:\text{HNO}_3$ mixture to visualize the domain structure. Figure 7a is the structure on the top surface and 7b is the cross-sectional structure. It is seen that the grating structure suitable for efficient QPM wavelength conversion is obtained: the duty cycle is successfully controlled to be 0.5 and it continues to the bottom. We obtained the domain-inverted grating with period as fine as 2 μm and with area of ~ 10 mm^2 (Kintaka *et al* 1996b).

4. Demonstration of the devices

4.1 Device design and fabrication

We fabricated the LiNbO_3 waveguide QPM wavelength conversion devices using the external voltage application technique. The actual device configuration is shown in figure 8. It consists of a fan-out domain-inverted grating and channel waveguide array (Ishigame *et al* 1991). The period is slightly different for each waveguide channel and ranges from $\Lambda_{\min}$ to $\Lambda_{\max}$. One can find a channel with the optimum grating period from the array despite

Figure 6. Typical waveforms of the applied voltage and the inversion charge.

the uncertainty of the waveguide effective indices and/or fabrication error. The number of the channel M is determined to make the period difference between the adjacent channels, $(\Lambda_{\max} - \Lambda_{\min})/M$, smaller than the acceptance period error $\delta\Lambda$.

The channel waveguides were fabricated by annealed proton exchange technique. An aluminum film with channel openings is defined on the periodically domain-inverted LiNbO₃ crystal by electron beam writing and lift-off. The crystal is soaked in pure benzoic acid melt at 200°C for 20–90 min and thermally annealed at 350°C for 1–6 h. The input and the output facets of the waveguide are made by polishing the crystal.

4.2 Green/blue/ultraviolet generation by QPM-SHG

We fabricated and examined the LiNbO₃ waveguide QPM-SHG devices for green, blue, or ultraviolet light generation (Kintaka *et al* 1996a, b). The device parameters are listed in table 1. A CW-Nd:YAG laser was used as a pump source in SH green light generation experiments, and a CW-Ti:Al₂O₃ laser for blue and ultraviolet light generation experiments. Figure 9 shows the results of the green and blue light generation

experiments. The SHG efficiency was proportional to the pump power and reached about 5% in both the devices. The normalized SHG efficiencies of 130%/W for green and 150%/W for blue were obtained in the devices with $L = 3$ mm. The normalized efficiencies are comparable to the theoretical prediction, that is 110%/W for green and 440%/W for blue. For more precise comparison more characterization of the device performances are required.

Figure 10 shows the experimental results of ultraviolet light generation. An ultraviolet of 393 nm wavelength was obtained with a normalized efficiency of 70%/W in the device with $\Lambda = 2.2$ μm and $L = 3$ mm. The maximum output power was as high as 1.9 mW. Generation of a wave with 383 nm wavelength was also obtained for a device with $\Lambda = 2.0$ μm , although the normalized efficiency was not high because of the short interaction length $L = 1$ mm.

We developed a resonant waveguide QPM-SHG device for enhancement of the efficiency (Fujimura *et al* 1996a, b). It is shown in figure 11. Two dielectric pump mirrors were attached on the waveguide facets to construct a waveguide resonator for the pump wave. The pump resonance was achieved by tuning the electrooptic phase modulator. In resonance the pump power is accumulated in the waveguide cavity and higher SHG efficiency is obtained compared with the travelling wave-type device. A fabricated device was tested by using a laser diode emitting at 865 nm wavelength. The phase modulator was driven by a ramp voltage wave to tune the resonance condition. Then the SH output was modulated as shown in figure 12. The peaks represent the enhanced resonant SHG output. In figure 13 the SHG efficiency dependent on the pump power is plotted and compared with the results measured before constructing the cavity. A normalized efficiency of 400%/W was obtained in the resonant device. Eight-fold enhancement of the efficiency was achieved by the resonance effect.

Figure 7. SEM photographs of a top view (a), and a cross-section (b), of a domain-inverted grating after etching by HF:HNO₃. The grating period was 2 μm . The thickness of the crystal was 0.15 mm.

Figure 8. Schematic illustration of fabricated LiNbO₃ waveguide QPM second-harmonic generation devices.

Table 1. Device parameters for SH green/blue/ultraviolet light generation devices.

	Green	Blue	UV
Λ [μm]	5.8–7.0	2.9–3.4	2.0, 2.2
M	166	168	–
L [mm]	3	3	1, 3
Pump	Nd : YAG	Ti : Al_2O_3	Ti : Al_2O_3

Figure 9. Dependence of SHG efficiency on the pump power in green light generation (circles; dashed line) and in blue light generation (squares; solid line) experiments. **Figure 10.** Dependence of SHG efficiency on the pump power in 393 nm light generation (open circles) and in 383 nm light generation (closed circles).

4.3 Third harmonic generation by cascading SHG and sum frequency generation

Third harmonic generation (THG) is another method to generate short-wavelength light. We proposed a waveguide THG device (Kintaka *et al* 1997), in which a QPM-SHG part and a QPM sum-frequency generation (SFG) part were cascaded. A prototype device configuration is shown in figure 14. Pumped by a CW-Nd : YAG laser light the first SHG part generates SH green light. The green light and the transmitted residual pump wave are coupled into the second SFG part and mixed to generate sum-frequency wave, i.e. TH wave of 335 nm wavelength. We fabricated the SHG and the SFG parts separately and butt-coupled for preliminary experiments. The interaction lengths were 3 mm for both parts. We obtained the TH power of 0.3 μW for 30 mW pumping. Higher output power will be obtained in one-chip THG devices.

Figure 11. The resonant waveguide QPM-SHG device with an electrooptic phase modulator for tuning. **Figure 12.** The SH output power when the phase modulator was driven by a ramp wave with peak-to-peak amplitude of 30 V.

4.4 Wavelength conversion in IR region by difference frequency generation

We also examined difference frequency generation (DFG) devices for wavelength conversion in wavelength range from 1 to 2 μm (Fujimura *et al* 1998). A pump wave with frequency ω_p and a signal wave with ω_s are coupled in the waveguides simultaneously. The waves are mixed and DF wave of $\omega_d = \omega_p - \omega_s$ is generated. The pump wavelength and the QPM grating period for the conversion were calculated and are shown in figure 15. We designed and fabricated a DFG device for the wavelength conversion from 1.54 μm to 1.67 μm with pumping at 0.8 μm . A CW-Ti:Al₂O₃ laser was used for a pump source and a laser diode for a signal source. Figure 16 shows the experimental result observed by an optical spectrum analyzer. A conversion efficiency of about

– 30 dB was obtained. The normalized conversion efficiency was estimated to be 10%/W. Experimental work is continuing to improve the device performances.

5. Conclusions

Our work on LiNbO₃ waveguide quasi-phase matched (QPM) nonlinear optic wavelength conversion devices

Figure 13. The SHG efficiency dependent on the pump power for the resonant SHG device (solid line) and that for the SHG device without cavity (dashed line).

Figure 14. LiNbO₃ waveguide third-harmonic generation device for preliminary experiments. The SHG part and SFG part are butt-coupled.

Figure 15. Pump wavelength and QPM grating period required for the wavelength conversion by LiNbO₃ QPM difference frequency generation in wavelength range from 1 to 2 μm .

Figure 16. Experimental result of wavelength conversion from 1.54 μm to 1.67 μm in a waveguide QPM difference frequency generation device.

was reviewed. The requirements to the domain-inverted grating for efficient wavelength conversions were shown by theoretical analysis of the device performances. The fabrication apparatus of ferroelectric-domain-inverted gratings was described, and it was shown that the resultant grating satisfy the requirements. A variety of LiNbO_3 waveguide QPM wavelength conversion devices were fabricated using the apparatus and demonstrated. We are pursuing research work to expand the available wavelength range through not only second-harmonic generation but also sum-frequency generation and difference frequency generation.

Some research groups are studying new research topics related to the QPM wavelength conversion device, e.g. pulse compression (Arbore 1997), optical signal processing utilizing nonlinear phase shift (Baek *et al* 1996), and squeezed light generation (Suhara *et al* 1996; Serkland *et al* 1997).

References

- Arbore M A, Marco O and Fejer M M 1997 *Opt. Lett.* **22** 865
- Armstrong J A, Bloembergen N, Ducuing J and Pershan P S 1962 *Phys. Rev.* **127** 1918
- Baek Y, Schiek R, Stegeman G I, Krijnen G, Baumann I and Sohler W 1996 *Appl. Phys. Lett.* **68** 2055
- Eger D, Oron M, Katz M and Zussman Z 1994 *Appl. Phys. Lett.* **64** 3208
- Fujimura M, Suhara T and Nishihara H 1991 *Electron. Lett.* **27** 1207
- Fujimura M, Kintaka K, Suhara T and Nishihara H 1993 *J. Lightwave Technol.* **11** 1360
- Fujimura M, Suhara T and Nishihara H 1996a *J. Lightwave Technol.* **14** 1899
- Fujimura M, Sudoh M, Kintaka K, Suhara T and Nishihara H 1996b *IEEE J. Selected Topics in Quantum Electron.* **2** 396
- Fujimura M, Shiratsuki A, Suhara T and Nishihara H 1998 *Jpn J. Appl. Phys.* **37** L659
- Ishigame Y, Suhara T and Nishihara H 1991 *Opt. Lett.* **16** 375
- Ito H, Takyu C and Inaba H 1991 *Electron. Lett.* **27** 1221
- Kazansky P G, Russel P, St J and Takabe H 1997 *J. Lightwave Technol.* **15** 1484
- Kintaka K, Fujimura M, Suhara T and Nishihara H 1996a *J. Lightwave Technol.* **14** 462
- Kintaka K, Fujimura M, Suhara T and Nishihara H 1996b *Electron. Lett.* **32** 2237
- Kintaka K, Fujimura M, Suhara T and Nishihara H 1997 *Electron. Lett.* **33** 1459
- Lim E J, Fejer M M, Byer R L and Kozlovsky W J 1989 *Electron. Lett.* **25** 731
- Mizuuchi K, Yamamoto K and Sato H 1994 *J. Appl. Phys.* **75** 1311
- Myers L E, Eckardt R C, Fejer M M, Byer R L, Bosenberg W R and Pierce J W 1995 *J. Opt. Sci. Am.* **B12** 2102
- Serkland D K, Kumar P, Arbore M A and Fejer M M 1997 *Opt. Lett.* **22** 1497
- Suhara T and Nishihara H 1990 *IEEE J. Quantum Electron.* **26** 1265
- Suhara T, Morimoto T and Nishihara H 1993 *IEEE Photon Technol. Lett.* **5** 934
- Suhara T, Fujimura M, Kintaka K, Nishihara H, Kurz P and Mukai T 1996 *IEEE J. Quantum Electron.* **3** 690
- Webjorn J, Laurell F and Arvidsson G 1989 *IEEE Photon Technol. Lett.* **1** 316
- Webjorn J, Siala S, Nam D W, Waarts R G and Lang R J 1997 *IEEE J. Quantum Electron.* **33** 1673
- Yamada M, Nada N, Saitoh M and Watanabe K 1993 *Appl. Phys. Lett.* **62** 435
- Yi S, Shin Y J and Son Y 1996 *Appl. Phys. Lett.* **68** 2493
- Yoo S J B, Caneau C, Bhat R, Koza M A, Rajhel A and Antoniadis N 1996 *Appl. Phys. Lett.* **68** 2609