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Second-harmonic generation in a LiTaO₃ waveguide domain-inverted by proton exchange and masked heat treatment

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Second-harmonic generation in the proton-exchanged LiTaO₃ waveguide is greatly enhanced by reducing the amount of proton exchange in fabricating the domain grating and the waveguide. To reduce the amount of proton exchange, a Ta-SiO₂ mask is used for the periodic domain inversion and a SiO₂ mask for the proton-exchanged waveguide. The fabricated device generates second-harmonic blue light of 1 mW at 429 nm with the fundamental wave power of 10.3 mW. Its normalized efficiency is 1500 %/Wcm², which is the highest value for LiTaO₃ waveguide devices reported to date. © 1996 American Institute of Physics. [S0003-6951(96)02518-1]

The quasi-phase-matched second-harmonic generation (QPM-SHG) in a periodically domain-inverted waveguide is a promising technique to get a compact blue light source. It has been already demonstrated in LiNbO₃ and LiTaO₃ waveguides.^{1,2} LiTaO₃ is a favorable nonlinear optic crystal owing to its high optical damage resistance and high nonlinearity ($d_{33}=26$ pm/V).³ LiTaO₃ waveguides fabricated by proton exchange (PE) and a short-time annealing² may have a tight mode confinement that is required to achieve a high conversion efficiency. The conversion efficiency over 1000%/Wcm² is expected by a theoretical calculation.⁴ However, the reported efficiency is about 230%/Wcm². A possible explanation for this difference is a reduced nonlinearity due to the PE process for domain inversion (DI) and waveguide formation.⁵ It is reported that the nonlinearity of LiTaO₃ is severely degraded near the crystal surface by a high PE rate, and the damaged nonlinearity cannot be recovered in the whole waveguide cross section by the conventional short-time annealing.⁵ For example, if a device has zero nonlinearity from surface to 1 μm depth due to the damaged nonlinearity,⁵ the calculated efficiency is lower than 200%/Wcm². This is similar to the reported values. Therefore, it is desirable to reduce the amount of PE to fully utilize the original nonlinearity in LiTaO₃.

In this letter, we have shown that the SHG efficiency in the LiTaO₃ QPM waveguide can be greatly enhanced by reducing the amount of PE. To reduce the amount of PE, we use a Ta-SiO₂ mask for the domain grating and a SiO₂ mask for the waveguide. The second-harmonic blue light of 1 mW at 429 nm is generated with the fundamental wave of 10.3 mW. Its normalized efficiency is 1500%/Wcm² which is the highest value for LiTaO₃ waveguide devices reported to date.

Previously, we proposed proton diffusion (PD) with a SiO₂ mask to fabricate good quality waveguides.^{6,7} This pro-

cess uses a small amount of initial PE owing to the proton-conserving property of the SiO₂ mask. PD is carried out at a high temperature using the SiO₂ mask above the waveguide channel. During PD the SiO₂ mask promotes proton indiffusion by inhibiting proton outdiffusion, which facilitates the appropriate index increase for guided modes in the depth direction. By using this process, good waveguides in Y-cut LiNbO₃ were demonstrated^{7,8} without surface damage and degradation of electrooptic effect. On the other hand, the annealed proton exchange (APE) method has some difficulty in fabricating a Y-cut LiNbO₃ single-mode waveguide because of the surface damage resulting from PE.

Recently, we also reported DI in LiTaO₃ formed by PE and subsequent heat treatment with a mask.^{9,10} The heat treatment with the mask was found effective to form a deep DI with a reduced amount of PE. In this process a Ta-SiO₂ mask is used, instead of a thick SiO₂ film, to enhance its effect.¹⁰ The SiO₂ film thicker than 1 μm on the surface of LiTaO₃ often cracks during the heat treatment above 500 °C due to the difference of thermal expansion between LiTaO₃ substrate and SiO₂ film. Instead of SiO₂ film, Ta film can be used as a mask. However, the discoloration at the crystal surface occurs due to thermal oxidation of the metal film at high temperature. To prevent these problems, we inserted a thin SiO₂ film with about 250 nm thickness between Ta film and the top surface of the crystal. The combination of Ta and SiO₂ film is better than a thick SiO₂ film for suppressing the proton outdiffusion.

During the heat treatment the mask promotes proton diffusion into the substrate by suppressing proton diffusion out of the substrate. According to the DI mechanism,¹¹ the enhanced indiffusion causes a strong poling field inside the substrate, and it results in a deep DI. For the same DI depth the heat treatment with the mask requires less amount of initial PE than that without the mask. Typically, the amount of PE is reduced to about 50% of that used in a conventional DI process without the mask.

Therefore, by reducing the amount of PE in fabricating the domain grating and the waveguide, the original nonlin-

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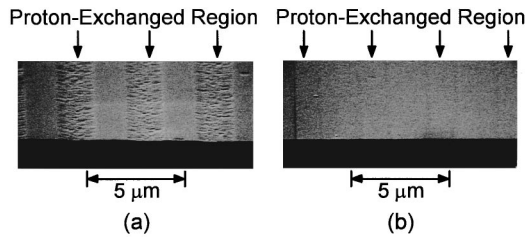


FIG. 1. Etched surface photographs of heat-treated samples (a) without mask and (b) with mask. Both samples were proton-exchanged in pure benzoic acid at 220 °C for 2 h, subsequently heat-treated in dry O₂ flow at 550 °C for 30 s, and then etched in 1HF+2HNO₃ at 25 °C for 30 min.

erarity in LiTaO₃ can be maintained. Thus the second-harmonic blue light is generated efficiently.

In addition, the heat treatment with the mask gives an important advantage. That is, the mask prevents a formation of the crystal defect on the proton-exchanged surface during the heat treatment for the domain grating. After a conventional heat treatment without the mask in dry O₂ flow an opalescent compound is usually observed in the proton-exchanged region by naked-eye or microscope inspection. However, if the heat treatment is done with the mask, this compound is not found. To confirm that this compound is different from bulk crystal, we may etch the heat-treated samples in a mixture of HF and HNO₃ and detect crystal defects.^{5,12} Figure 1 shows the etched surface of the samples heat-treated without and with the mask, respectively. The proton-exchanged regions are severely etched in the sample without the mask, but they are hardly etched in the sample with the mask. The formation of this compound is obviously related to the proton outdiffusion. This compound will be a Li-deficient phase of LiTaO₃ induced by proton outdiffusion. During the heat treatment of Li_(1-x)H_xTaO₃, lattice hydrogen combines with lattice oxygen to give rise to water molecules, which may evaporate near the crystal surface as proton outdiffusion. This dehydration reaction results in the Li-deficient layer of Li_(1-x)TaO_(3-0.5x) near the crystal surface. From the surface observation of the proton-exchanged LiNbO₃ specimens heat-treated at 550 °C for 2 h by using the transmission electron microscopy (TEM), it was reported that the Li-deficient phase of LiNbO₃ is formed in the

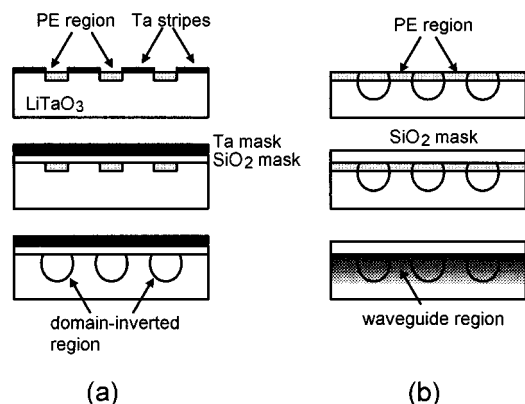


FIG. 2. Fabrication procedures of a SHG device. (a) domain grating and (b) waveguide.

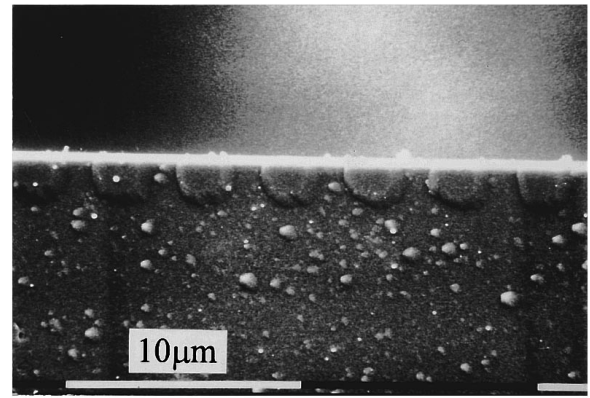


FIG. 3. Revealed domain-inversion structure.

proton-exchanged region due to the dehydration reaction.¹³

The fabrication procedures of a LiTaO₃ QPM waveguide device are shown in Fig. 2. After depositing periodic Ta film stripes with a 3.5 μm period and a 2.0 μm stripe width on *-c* face of LiTaO₃, we put the sample for PE in pure benzoic acid melt at 220 °C for 40 min. After removing Ta stripes, a Ta-SiO₂ mask is deposited on the proton-exchanged surface to get a deep DI. The sample is heat-treated at 570 °C for 30 s using a rapid thermal annealing (RTA) furnace. We use a rapid heat-up rate of 80 °C/s. Both DI depth and width are equal to 1.8 μm as shown in Fig. 3. We form channel waveguides with a pattern width of 4 μm, perpendicular to the domain grating. PE is performed in pure benzoic acid melt at 220 °C for 40 min. After PE, a SiO₂ mask is deposited in the waveguide region to get a tight mode confinement⁶ by suppressing the proton outdiffusion. PD with the SiO₂ mask is performed for 5 min in a furnace at 400 °C. Both end faces of the sample are polished for the end-fire light coupling. Our sample length is 8 mm long after polishing.

Table I shows estimated PE depths in the conventional procedure and in our procedure. The estimated amount of PE in our procedure is 50–60% of that in the conventional procedure. After the PD is done, the exchange factor *x* of Li_(1-x)H_xTaO₃ in the whole region of our waveguide is estimated to be lower than the critical value⁵ of 0.16. Below this critical value, the crystal phase is maintained in the single original α phase, and the crystal nonlinearity is completely restored.⁵

The characteristics of our SHG device are measured by

TABLE I. Comparison of the amount of proton exchange.

| | Conventional procedure | Our procedure |
|------------------|--|--|
| Domain inversion | PE for 20 min at 260 °C, $d_e=0.55 \mu\text{m}^{\text{a,c}}$ | PE for 40 min at 220 °C, $d_e=0.28 \mu\text{m}^{\text{b}}$ |
| PE waveguide | PE for 14 min at 260 °C, $d_e=0.46 \mu\text{m}^{\text{a,d}}$ | PE for 40 min at 220 °C, $d_e=0.28 \mu\text{m}^{\text{b}}$ |

^aPE depth for pyrophosphoric acid,¹⁴ $d_e=6.81 \times 10^3 \sqrt{t} \exp(-4.73 \times 10^3/T)$.

^bPE depth for pure benzoic acid,¹⁵ $d_e=1.15 \times 10^5 \sqrt{t} \exp(-6.27 \times 10^3/T)$.

^cPE conditions in Ref. 16.

^dPE conditions in Ref. 2.

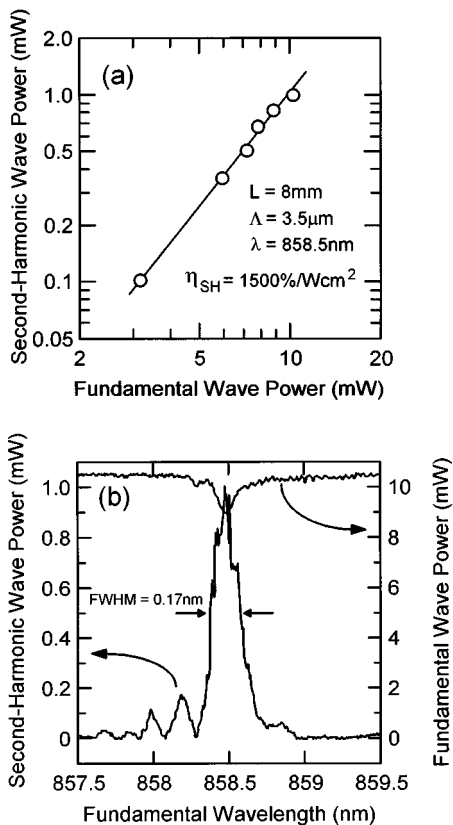


FIG. 4. Measured characteristics of SHG device (a) second-harmonic wave power vs fundamental wave power and (b) second-harmonic wave power vs fundamental wavelength.

using a widely tunable $\text{Ti:Al}_2\text{O}_3$ laser as fundamental wave source. The measured results are shown in Fig. 4. The conversion efficiency is calculated by measuring the powers of the fundamental wave and the second-harmonic blue light through the waveguide end face. The SHG device stably generates blue light of 1 mW at 429 nm when the fundamental wave power is 10.3 mW. The conversion efficiency is almost 10% and the normalized efficiency is $1500\%/W\text{cm}^2$. The dependence of SHG on the fundamental wave power is shown in Fig. 4(a). The second-harmonic wave power is proportional to the square of the fundamental wave power. The

dependence of the fundamental and the second-harmonic wave power is shown in Fig. 4(b), which also demonstrates the maximum conversion of the fundamental wave at 858.5 nm into the second-harmonic wave. The measured FWHM bandwidth is 0.17 nm that agrees reasonably with theoretical bandwidth of 0.12 nm.

In conclusion, we demonstrated that the SHG efficiency of LiTaO_3 waveguide is significantly enhanced by reducing the amount of PE in fabricating the SHG device. To reduce the amount of PE we used a Ta- SiO_2 mask in the heat-treatment for the domain grating and a SiO_2 mask in the proton diffusion process for the waveguide. In the fabricated device, the second-harmonic blue light of 1 mW at 429 nm was generated with the fundamental wave power of 10.3 mW. Its normalized efficiency is $1500\%/W\text{cm}^2$ which is the highest value for LiTaO_3 devices reported to date.

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