

Quasi-phasematched frequency conversion in lithium niobate and lithium tantalate waveguides

E. J. Lim, S. Matsumoto,[†] M. L. Bortz, and M. M. Fejer

Edward L. Ginzton Laboratory,
Stanford University, Stanford, California, 94305

[†]Corporate Research Laboratories, Sony Corporation,
6-7-35, Kitashinagawa, Shinagawa-ku, Tokyo, 141 Japan

ABSTRACT

We discuss second harmonic generation of green and blue light in periodically-poled LiNbO₃ and LiTaO₃ waveguides, and difference frequency generation of infrared radiation in periodically-poled LiNbO₃ waveguides.

1. INTRODUCTION

There currently exist numerous uses for miniature sources of blue-green and infrared radiation. In the blue-green spectral range of 400–500 nm, applications exist in the fields of optical storage, reprographics, color displays, and biomedical instrumentation. For these applications the minimum usable power is about 1 mW. In the mid-infrared region of 2–4 μm, applications exist in the fields of chemical sensors, infrared fibers, and remote sensing, where the minimum useful power for these applications is about 100 μW. There are diode lasers operating in these two wavelength regions, but they currently work at cryogenic temperatures, and many potential users desire room temperature operation. Guided-wave nonlinear frequency conversion of near-infrared laser diodes, however, is a potential solution to the problem of efficiently generating coherent radiation at these wavelengths at room temperature. Waveguides enhance the efficiency of nonlinear optical interactions because they enable large optical intensities to be maintained over considerable interaction lengths.¹ AlGaAs and InGaAs lasers generate radiation in the 0.75–1 μm range, while InGaAsP lasers provide wavelengths in the 1.3–1.5 μm range. Blue-green radiation can thus be obtained through second harmonic generation (SHG) by directly doubling the output of AlGaAs and InGaAs diode lasers, while mid-infrared radiation can be obtained through difference-frequency generation (DFG) by mixing the radiation from AlGaAs/InGaAs and InGaAsP diode lasers.

2. QUASI-PHASE-MATCHING

Quasi-phasematching (QPM) was an early proposal for compensating refractive index dispersion in nonlinear optics, and involves modulating the nonlinear coefficient of the medium with a spatial frequency equal to the wavevector mismatch of the nonlinear interaction.² In its simplest implementation, called first-order QPM, the sign of the nonlinear coefficient is changed after every coherence length of the interaction. For SHG of blue-green radiation in LiNbO₃ and LiTaO₃ for example, coherence lengths are about 1–2 μm, while for DFG the coherence lengths are longer, ~10 μm. Third-order QPM, where the sign of the nonlinear coefficient is changed after every three coherence lengths, can be used to simplify the fabrication of the device at the expense of efficiency. QPM is a useful technique because it allows phasematching of any nonlinear coefficient at any temperature regardless of the medium's birefringence, thus permitting a nonlinear medium to be used across its entire transparency range. This is important for SHG of blue light in LiNbO₃ and LiTaO₃ because these crystals lack enough birefringence to phasematch at these wavelengths. For DFG in LiNbO₃, QPM allows operation at room temperature instead of the ~550 °C temperature that would be required for birefringent phasematching.³

In ferroelectrics, the signs of the nonlinear coefficients are linked to the direction of the spontaneous electric polarization, and thus QPM can be accomplished through periodic ferroelectric domain reversal. We now discuss our devices in LiNbO₃ and LiTaO₃.

3. PERIODICALLY-POLED LiNbO₃ DOUBLER

During the development of the Ti indiffusion process for waveguide fabrication in LiNbO₃, it was found that Ti diffusion on the +Z face of the crystal, under certain diffusion conditions, caused ferroelectric domain reversal at the surface of the crystal.⁴ Selective Ti diffusion provides a way of patterning the domain reversal, and periodically-reversed domains fabricated in this manner have been applied to surface-acoustic wave (SAW) devices.⁵ We have combined the Ti-diffusion method of patterning the ferroelectric domains with an annealed proton exchange waveguide process⁶ to fabricate waveguide doublers^{7,8} and a difference-frequency mixer.⁹

For a doubler reported recently,¹⁰ a periodically-poled substrate was created by diffusing a Ti grating with a period of 8.5 μm , appropriate for third-order QPM of SHG of blue light. Lift-off lithography was used to define the grating of 1.5 μm -wide lines on the +Z surface of a 1 mm-thick integrated optics-grade, Z-cut LiNbO₃ wafer. The Ti lines had a thickness of 5 nm and were parallel to the Y-axis of the crystal. The Ti grating was then diffused into the substrate with a 2 h ramp-up from room temperature to 1100 °C and a 15 min soak at 1100 °C, after which the oven was turned off and allowed to cool to room temperature. To prevent outdiffusion of lithium oxide from the sample during the poling process, the substrate was placed in a closed alumina boat filled with congruent lithium niobate powder.

Following the poling process, annealed proton exchange (APE) channel waveguides were fabricated in the substrate. Channels parallel to the X-axis, with widths in the range 2–10 μm were defined in a 100 nm-thick sputtered silicon dioxide film with lithography followed by wet etching in 6:1 buffered oxide etch. The masked sample was soaked for 1.5 h in pure benzoic acid at 160 °C, after which the mask was etched away and the sample was annealed in air for 4 h at 333 °C. After the endfaces were polished, the waveguides were about 1 cm long.

The sample was characterized using a tunable cw Ti:Al₂O₃ laser operating in the range 0.8–0.9 μm as the source of fundamental radiation. Endfire coupling was accomplished with microscope objectives. In a 5.5 μm -wide waveguide, we found three blue modes with significant conversion efficiency, a TM₀₀ mode for fundamental wavelength $\lambda_\omega = 839$ nm, a TM₂₀ mode for $\lambda_\omega = 828$ nm, and a superposition of modes for $\lambda_\omega = 820$ nm. The output power level in the blue, corrected for Fresnel reflections, was 1.1 mW for the TM₀₀ mode with a fundamental power of 232 mW in the waveguide. The most efficient interaction in this waveguide involved the superposition of blue modes. Up to 200 mW power levels of the fundamental, the second harmonic power depended quadratically on the fundamental power. The maximum amount of blue light we observed was 2.8 mW at 415 nm, generated from 300 mW of 830 nm radiation in an 8 μm -wide waveguide.

4. PERIODICALLY-POLED LiNbO₃ DIFFERENCE-FREQUENCY MIXER

The fabrication of the difference-frequency mixer required steps similar to those discussed above for the doubler.⁹ A Ti grating with 5 μm -wide lines and a period of 21 μm was diffused to enable first-order QPM of a DFG interaction where $\lambda_p = 0.81$ μm , $\lambda_s = 1.32$ μm , and $\lambda_i = 2.1$ μm , where p , s , and i identify the pump, signal, and idler wavelengths, and where the respective frequencies are related by $\omega_p = \omega_s + \omega_i$. The diffusion required a 2 h soak at 1100 °C. Fig 1(a) shows a polished and etched Y-face of a Z-cut LiNbO₃ substrate following the indiffusion of the Ti grating, illustrating the resulting triangular, reversed domains. Preferential etching in HF was used to reveal the reversed domains. For the proton exchange mask, channels parallel to the X-axis, with widths in the range 2–15 μm , were defined in a 200 nm-thick layer of aluminum on the substrate using lift-off lithography. Following a 2 h soak in pure benzoic acid at 200 °C, the Al mask was removed in a solution of NaOH, and the sample was annealed in air for 12 h at 333 °C. Fig. 1(b) shows the polished and etched cross section of a periodically-poled sample after an unmasked APE process, where we see that the process has changed the shape of the triangular domains, rounding them but not significantly reducing their depth.

For the optical experiment, pump radiation in the 0.8–0.9 μm range from a cw tunable Ti:Al₂O₃ laser and signal radiation at 1.32 μm from a cw Nd:YAG laser were endfire coupled into a 5 μm -wide waveguide on a 7 mm-long sample. For a pump wavelength of $\lambda_p = 0.81$ μm , difference-frequency output at $\lambda_i = 2.1$ μm was detected after a germanium filter with a lead sulfide detector and a lock-in amplifier. Several modes at 0.81 μm and the lowest-order mode at 1.3 μm were observed, and the waveguide was assumed to be single-moded at 2.1 μm . All modes were TM, and thus were coupled by the d_{33} coefficient. Fig. 2 shows the idler power increasing linearly with pump power, as

expected in a low-gain interaction. A maximum of 1.8 μW of 2.1 μm idler radiation was measured with 160 mW of 0.81 μm and 1 mW of 1.32 μm radiation coupled into the waveguide. Fig. 3 shows the difference-frequency power as a function of pump wavelength. The observed full width at half maximum (FWHM) pump wavelength tuning bandwidth, 0.86 nm, corresponds to a tuning bandwidth of 5.8 nm for the idler wavelength. From the bulk refractive indices for LiNbO₃, the theoretical FWHM pump bandwidth is calculated to be 4.5 nm-mm. The observed FWHM in Fig. 3 thus implies an effective interaction length of about 5 mm, in reasonable agreement with the 7 mm device length. Together with the power levels given earlier, this results in an experimental normalized conversion efficiency of about 4%/W·cm².

5. PERIODICALLY-POLED LiTaO₃ DOUBLER

LiTaO₃, like LiNbO₃, is a uniaxial, ferroelectric crystal that is produced in large quantities for SAW devices. It is reported to be more resistant to photorefractive damage than LiNbO₃, and has relatively large nonlinear coefficients ($d_{33}(\text{LiTaO}_3) = 26 \text{ pm/V} \approx 0.75d_{33}(\text{LiNbO}_3)$).¹¹ Despite these qualities, LiTaO₃ has not found wide use in frequency conversion because its small birefringence prevents phasematching of many interactions. Recently, however, QPM techniques have been applied to LiTaO₃ for SHG of visible radiation. Periodically-poled LiTaO₃ has been fabricated in bulk form using Czochralski growth,¹² and in wafer form using proton exchange followed by heat treatment,^{13,14,15,16} and periodic electric fields.^{17,18} We discuss here electrically periodically-poled LiTaO₃ channel waveguides for SHG of blue light.

To pole the surface of a substrate, a spatially periodic electric field was applied with interdigital electrodes while heating the LiTaO₃ sample to just below the Curie temperature, T_C , ~ 610 °C. A poling period of 14 μm was chosen to accomplish third order QPM of SHG of 450 nm radiation, working from a Sellmeier equation fit to the bulk refractive index data for LiTaO₃.¹⁹ Interdigital electrodes were fabricated on the +Z face of a Z-cut, SAW-grade LiTaO₃ wafer²⁰ using liftoff lithography. Each interdigital electrode was made up of 3.5 μm -wide fingers, parallel to the Y axis, spaced every 14 μm , and consisting of a 200 nm-thick Au layer with a 5 nm-thick Ti adhesion layer. The patterned region extended 1 mm in the X direction and 2 mm in the Y direction. For poling, the sample was heated to 600 °C and a voltage of about 1.4 V was applied between the electrodes for a 10 minute period as the sample was cooled approximately 5 K. During this time the poling current decreased from about 8 μA /finger to 3.5 μA /finger. Fig. 4 shows the temperature profile of a typical poling process. After poling, the Au electrodes were removed in an iodine solution. The surface corrugation on the Z face caused by the electrodes during the poling process was measured with a surface profilometer to be about 20 nm. Fig. 5(a) shows a Y-face of a sample poled below T_C that was polished and then etched in HF to reveal the periodic ferroelectric domain structure. Deeper domains can be obtained by applying the poling fields at temperatures above T_C , as shown in Fig. 5(b), but the surface corrugation arising from the electrodes is more significant.

After poling the substrates, we fabricated channel waveguides using the annealed proton exchange process.²¹ The mask for the channel waveguides was formed by first sputtering 200 nm of SiO₂ onto the poled surface of the sample. Channels parallel to the X-axis with widths in the range 3–10 μm were then formed in the SiO₂ film by photolithography and wet etching. The sample was exchanged in pure benzoic acid at 200 °C for 4 h. Following the exchange, the SiO₂ mask was etched away and the sample was annealed in a flowing oxygen atmosphere at 333 °C for 10 h.

For the SHG experiments, a cw tunable Ti:Al₂O₃ laser was used as the source for the fundamental radiation at $\lambda \approx 0.9$ μm . Channels with a width of 10 μm were found to have the highest conversion efficiency. Frequency doubling into the TM₀₀ and TM₁₀ blue modes occurred, where the TM₀₀ interaction had the higher efficiency. Fig. 6 displays the measured wavelength tuning curve for doubling into both modes. With 41 mW of cw power at 916 nm measured at the output, we observed the generation of 1.3 μW of 458 nm radiation.

Using the bulk index data for LiTaO₃, the theoretical wavelength tuning bandwidth for frequency doubling of 0.9 μm radiation is calculated to be 1.2 nm-mm. Thus the measured wavelength tuning curves shown in Fig. 6 imply effective interaction lengths of 1 mm, in agreement with the 1 mm-long gratings used in the device. Together

with the measured powers presented above, the 1 mm interaction length results in a normalized conversion efficiency of about 8%/W-cm².

6. DISCUSSION

The experimentally observed conversion efficiencies for our LiNbO₃ and LiTaO₃ devices are roughly 3–20 times less than theoretically estimated. We suspect the discrepancy between the measured and calculated conversion efficiencies results mainly from a reduction in the nonlinear coefficient due to the proton exchange waveguide process. In LiNbO₃, the value of d_{33} has been estimated to be 40–60% of its bulk value after proton exchange.^{22,23,24} Post-exchange annealing has been reported to restore the nonlinear coefficient.^{23,24} We believe a similar situation exists for LiTaO₃ because we observed no SHG in unannealed guides. We also suspect that our annealed waveguides were not annealed enough to recover the full nonlinear coefficient, and we are currently studying the APE process to correct this.

7. CONCLUSIONS

Quasi-phasematching is a useful technique for phasematching any nonlinear interaction within a material's transparency range, at any temperature, using any nonlinear coefficient. We have demonstrated quasi-phasematched second harmonic generation of blue light in periodically-poled LiNbO₃ and LiTaO₃ waveguides, interactions which are impossible to birefringently phasematch. We have also demonstrated room temperature difference-frequency generation of 2.1 μm radiation in a periodically-poled LiNbO₃ waveguide, an interaction that would otherwise require a birefringent phasematching temperature of ~550 °C. All of these interactions used the largest nonlinear coefficient, d_{33} .

8. ACKNOWLEDGEMENTS

We are grateful to Crystal Technology, Inc. for providing the LiNbO₃ substrates, to Amoco Laser Company for the use of a 1.32 μm Nd:YAG, to Coherent, Inc. for providing mirrors for a dye laser, to T. Carver, L. Goddard, J. Vrhel, and C. Remen for assistance in fabrication, and to W. J. Kozlovsky and P. Wysocki for assistance in characterization. This work was supported by the IBM Corporation, the Sony Corporation, the Joint Services Electronics Program, and the DARPA Optoelectronic Materials Center.

9. REFERENCES

- ¹ G. I. Stegeman, J. J. Burke, and C. T. Seaton, "Nonlinear integrated optics," in *Integrated optical circuits and components*, edited by L. D. Hutcheson, Marcel Dekker, New York, 1987.
- ² J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, "Interaction between lightwaves in a nonlinear dielectric," *Phys. Rev.*, vol. 127, pp. 1918–1939, 1962.
- ³ H. Herrmann and W. Sohler, *J. Opt. Soc. Am. B*, vol. 5, pp. 278– , 1988.
- ⁴ S. Miyazawa, "Ferroelectric domain inversion in Ti-diffused LiNbO₃ optical waveguide," *J. Appl. Phys.*, vol. 50, 4599–4603, 1979.
- ⁵ K. Nakamura, H. Ando, and H. Shimizu, "Partial domain inversion in LiNbO₃ plates and its applications to piezoelectric devices," *Proc. 1986 Ultrasonics Symposium*, pp. 719–722.
- ⁶ P. G. Suchoski, T. K. Findakly, and F. J. Leonberger, "Stable low-loss proton-exchanged LiNbO₃ waveguide devices with no electro-optic degradation," *Opt. Lett.*, vol. 13, pp. 1050–1052, 1988.
- ⁷ E. J. Lim, M. M. Fejer, and R. L. Byer, "Second-harmonic generation of green light in periodically poled planar lithium niobate waveguide," *Electron. Lett.*, vol. 25, pp. 174–175, 1989.
- ⁸ E. J. Lim, M. M. Fejer, R. L. Byer, and W. J. Kozlovsky, "Blue light generation by frequency doubling in periodically poled lithium niobate channel waveguide," *Electron. Lett.*, vol. 25, pp. 731–732, 1989.
- ⁹ E. J. Lim, H. M. Hertz, M. L. Bortz, and M. M. Fejer, "Infrared radiation generated by quasi-phasematched difference-frequency mixing in a periodically-poled lithium niobate waveguide," *Appl. Phys. Lett.* , accepted for publication.

- 10 E. J. Lim, M. L. Bortz, and M. M. Fejer, "Milliwatt levels of blue light generated by quasi-phases-matched second harmonic generation in a periodically-poled lithium niobate waveguide," in *Integrated Photonics Research, 1991*, Technical Digest Series, (Optical Society of America, Washington, DC 1991) pp. 45-46.
- 11 S. K. Kurtz, J. Jerphagnon, and M. M. Choy, "Nonlinear dielectric susceptibilities," in Landolt-Börnstein Numerical Data and Functional Relationships in Science and Technology, Group III: Crystals and Solid State Physics, vol. 11, edited by K.-H. Hellwege, and A. M. Hellwege, (Springer Verlag, Berlin, 1979) pp. 671-743.
- 12 W. Wang, Q. Zou, Z. Geng, and D. Feng, "Study of LiTaO₃ crystals grown with a modulated structure," *J. Cryst. Growth*, vol. 79, pp. 706-709, 1986.
- 13 H. Ahlfeldt, J. Webjorn, and G. Arvidsson, "Periodic domain inversion in lithium tantalate," in *Integrated Photonics Research, 1991*, Technical Digest Series, (Optical Society of America, Washington, DC 1991) p. 60.
- 14 I. Sawaki, and S. Kurimura, "Second-harmonic generation in periodically domain-inverted lithium tantalate channel waveguides," Conference on Lasers and Electro-Optics, May 12-17, 1991, Baltimore, Maryland, paper CTuV4.
- 15 K. Mizuuchi, K. Yamamoto, and T. Taniuchi, "Second-harmonic generation of blue light in a LiTaO₃ waveguide," *Appl. Phys. Lett.*, vol. 58, pp. 2732-2734, 1991.
- 16 K. Yamamoto, K. Mizuuchi, and T. Taniuchi, "High power 12 mW blue light generation in periodically domain-inverted LiTaO₃ waveguide," Conference on Lasers and Electro-Optics, May 12-17, 1991, Baltimore, Maryland, postdeadline paper CPDP23
- 17 K. Nakamura and H. Shimizu, "Poling of ferroelectric crystals by using interdigital electrodes and its application to bulk-wave transducers," *Proc. 1983 Ultrasonics Symposium*, pp. 529-532.
- 18 S. Matsumoto, E. J. Lim, M. M. Fejer, H. M. Hertz, "Second-harmonic generation of blue light in a periodically poled LiTaO₃ waveguide," in *Integrated Photonics Research, 1991*, Technical Digest Series, (Optical Society of America, Washington, DC 1991) p. 97.
- 19 W. L. Bond, "Measurement of the refractive indices of several crystals," *J. Appl. Phys.*, vol. 36, pp. 1674-1677, 1965.
- 20 Shin-Etsu Chemical Co., Ltd., Tokyo, Japan.
- 21 T. Findakly, P. Suchoski, and F. Leonberger, "High-quality LiTaO₃ integrated-optical waveguides and devices fabricated by the annealed-proton-exchange technique," *Opt. Lett.*, vol. 13, pp. 797-799, 1988.
- 22 T. Suhara, H. Tazaki, and H. Nishihara, "Measurement of reduction in SHG coefficient of LiNbO₃ by proton exchanging," *Electron. Lett.*, vol. 25, pp. 1326-1328, 1989.
- 23 R. W. Keys, A. Loni, and R. M. De La Rue, "Measurement of the increase in the SHG coefficient of proton exchanged LiNbO₃ after annealing using a grating diffraction technique," *Electron. Lett.*, vol. 26, pp. 624-626, 1990.
- 24 X. Cao, R. Srivastava, R. V. Ramaswamy, and J. Natour, "Recovery of second-order optical nonlinearity in annealed proton-exchanged LiNbO₃," *IEEE Photon. Technol. Lett.*, vol. 3, pp. 25-27, 1991.

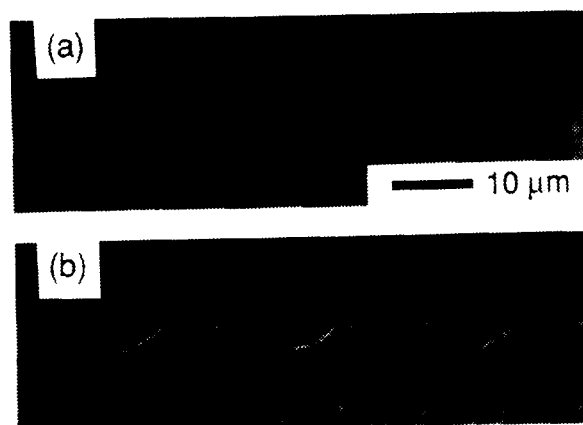


Fig. 1. Photomicrographs of polished and etched Y-faces of planar, periodically-poled Z-cut LiNbO₃ substrates (a) after indiffusion of the Ti grating, showing the triangular shape of the reversed domains, and (b) after the annealed proton exchange process, showing the rounding of the domains. In both photographs the period of the pattern is 21 μm.

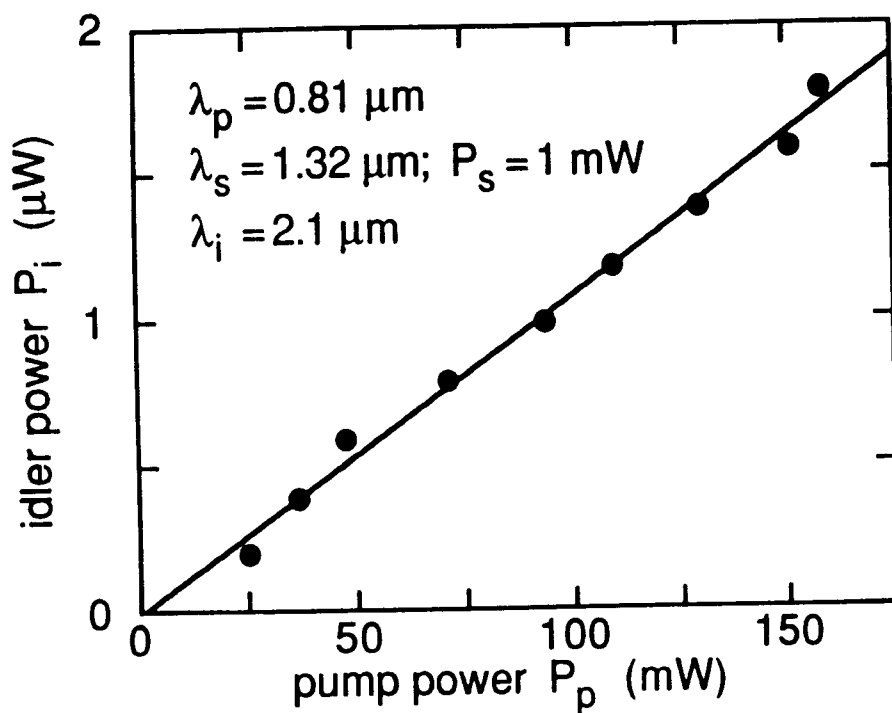


Fig. 2. Linear dependence of the measured difference-frequency power at 2.1 μm on pump power.

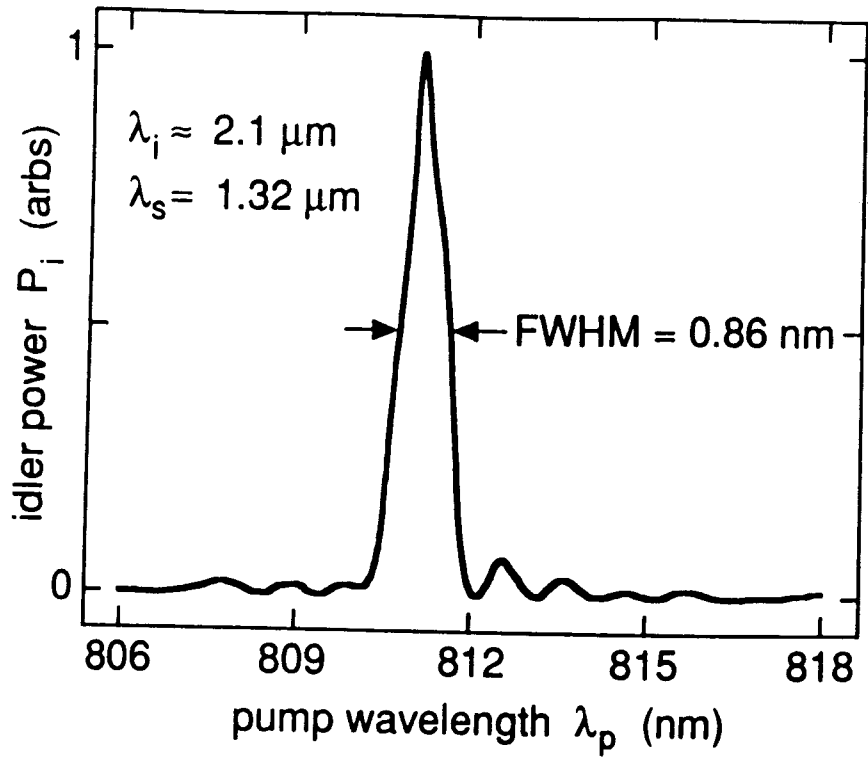


Fig. 3. Measured difference-frequency power at about 2.1 μm versus pump wavelength. The 0.86 nm FWHM bandwidth of the pump wavelength corresponds to a tuning bandwidth in the idler wavelength of 5.8 nm.

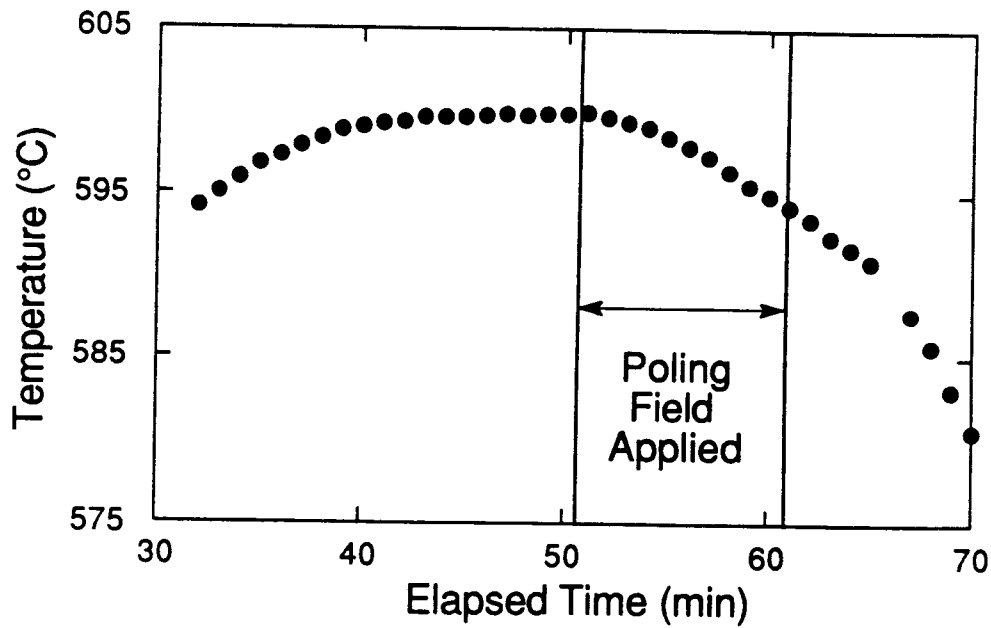


Fig. 4. Temperature profile of a typical electrical poling procedure for LiTaO_3 with interdigital electrodes.

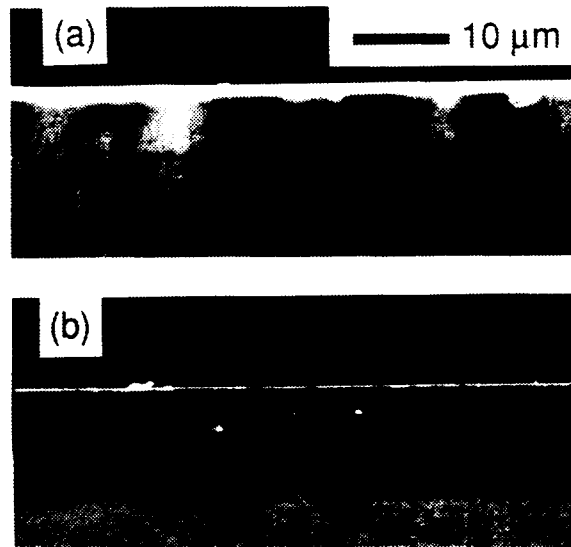


Fig. 5. Scanning electron micrographs of the cross sections of periodically reversed domains in LiTaO₃ produced by electrical poling at (a) ~600 °C (below T_C) and (b) ~650 °C (above T_C).

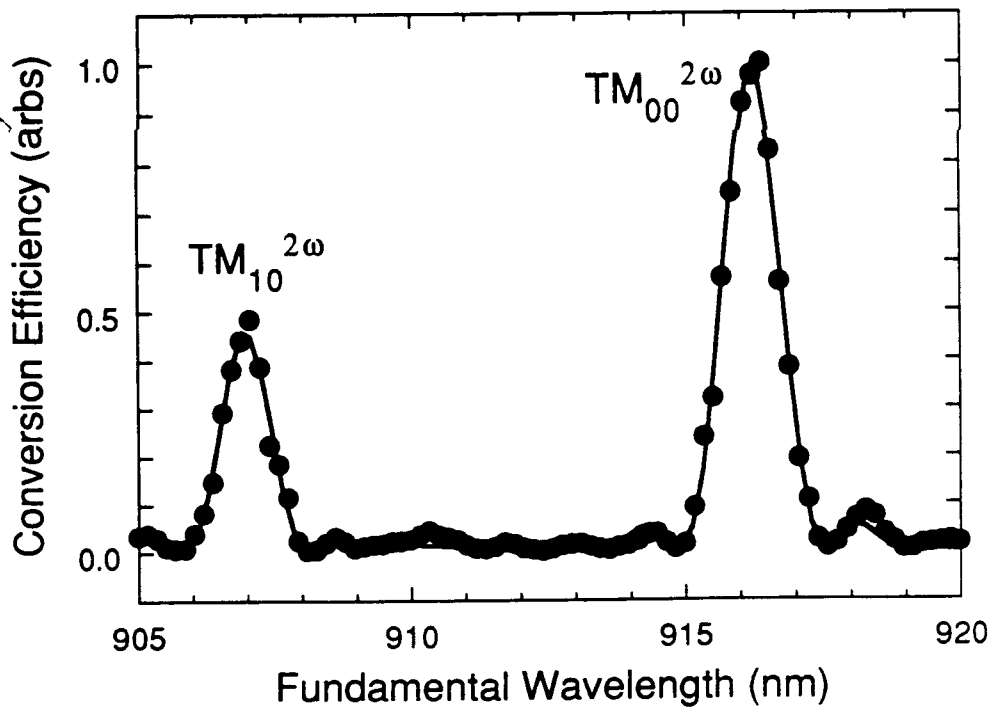


Fig. 6. Measured SHG conversion efficiency as a function of fundamental wavelength, with fitted sinc² curves, for periodically-poled LiTaO₃ channel waveguides, where the length of the QPM grating is 1 mm.