

# NONLINEAR FREQUENCY CONVERSION IN PERIODICALLY-POLED FERROELECTRIC WAVEGUIDES

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ABSTRACT. By patterning the domains in ferroelectric crystals, it is possible to introduce periodic changes in the sign of the nonlinear susceptibility in waveguide frequency conversion devices. Appropriate choice of the spatial frequency of the modulation allows quasi-phases matching of any nonlinear interaction involving radiation in the transparency range of the crystal. Room temperature second harmonic generation of visible radiation and difference frequency generation of infrared radiation have been demonstrated.

## 1. Introduction

Nonlinear frequency conversion in waveguides, studied since the first days of integrated optics, has recently received renewed attention. The advent of diode lasers with 100 mW single-mode output powers has made efficient solid-state sources available at a range of near-infrared wavelengths. It has not yet been possible to extend room-temperature operation of diode lasers to wavelengths shorter than 600 nm or longer than 2  $\mu$ m, so coherent sources for important applications such as high density optical storage based on single-mode blue light or sensors based on narrow-band infrared radiation are not available. Nonlinear frequency conversion of the output of near infrared diode lasers to extend the range of wavelengths available from all-solid-state sources is a potential solution to this problem, but efficient interactions at 100 mW input powers in bulk media can be accomplished only in relatively complex resonant devices. Frequency conversion in waveguides, which can be several orders of magnitude more efficient than comparable single-pass bulk interactions, is an attractive alternative. [Stegeman (1985)]

While the promise of waveguide frequency conversion was recognized early in the history of integrated optics, practical application has up to the present been limited. Media suited to these interactions must have reasonably large nonlinear susceptibilities, adequate transparency and damage threshold, a viable waveguide technology, and means for phasematching. Phasematching, defined as a means for maintaining the phase relationship between the waves involved in a nonlinear interaction, is essential for efficient frequency conversion devices. In birefringently phasematched interactions the waves are polarized orthogonally, so that the birefringence can be used to compensate for the phase velocity mismatch due to the dispersion of the refractive indices. While birefringent phasematching has been used in the vast majority of practical frequency conversion devices, it limits the choice of nonlinear crystal to those having an appropriate balance of dispersion and birefringence, and restricts interactions to those involving components of the

nonlinear susceptibility coupling orthogonally polarized waves. These requirements are rather restrictive, and have limited the choice of materials for birefringently phase-matched waveguide interactions essentially to lithium niobate and, recently, potassium titanyl phosphate (KTP). [Stegeman (1985)] The range of wavelengths accessible to birefringent phase-matching in these materials is limited, and operation at inconvenient temperatures is often required. Alternative methods for phase-matching, such as modal dispersion [Stegeman (1985)] and the Cerenkov effect [Taniuchi (1987)] are often accompanied by reduced conversion efficiency and nonoptimal output modes.

Another approach to phase-matching, involving periodic modulation of the material properties to compensate for the mismatch in the phase-velocity, quasi-phase-matching (QPM), has recently emerged as a practical method. Taking advantage of modern planar processing techniques, QPM is applicable in a variety of material systems and can be used in media with inadequate birefringence for conventional phase-matching, in interactions involving components of the nonlinear susceptibility tensor coupling modes of the same polarization, and over the entire transparency range of the medium.

In this chapter we discuss the basic theory of quasi-phase-matched interactions, processes for patterning domains in three important ferroelectric crystals, lithium niobate, lithium tantalate, and KTP, and implementations of second harmonic generators and difference frequency mixers in these crystals

## 2. Quasi-Phase-matching

QPM was one of the first techniques proposed for compensating for the effects of dispersion in nonlinear interactions. [Armstrong (1962)] In a quasi-phase-matched interaction, the waves propagate with different phase velocities, but accumulated phase mismatch is prevented by appropriate periodic modulation of the material properties. Consider second harmonic generation (SHG) for specificity. The direction of power flow in a phase-matched interaction is always from the fundamental to the second harmonic, and the second harmonic power grows quadratically with distance. In nonphase-matched SHG the direction of power flow reverses periodically as the relative phase of the fundamental and the second harmonic change, so that the power in the second harmonic wave oscillates, initially growing while the fundamental and second harmonic are in phase, then decreasing after the accumulated phase difference reaches  $\pi$  (at a distance  $l_c$ ), ultimately returning to zero after a distance  $2l_c$ . The coherence length  $l_c$  is defined as the distance over which the fundamental and the second harmonic accumulate a  $\pi$  relative phase shift, i.e. for propagation constant mismatch  $\Delta\beta$ ,  $l_c = \pi/\Delta\beta$ . In ideal quasi-phase-matched SHG, the sign of the nonlinear susceptibility  $d_{eff}$  is reversed every coherence length, so that the relative phase of the driving polarization and the free second harmonic field is "reset", and power continues to flow monotonically from the fundamental to the second harmonic. The second harmonic power increases in a complicated fashion, but with an average that remains quadratic with distance. If the sign of  $d_{eff}$  is reversed every three coherence lengths (third order QPM), the contributions to the second harmonic field from the first two coherence lengths of each half period cancel, leaving only one third of the crystal effectively contributing to the interaction. The average growth is again quadratic, but slower than for first order QPM. One can also view QPM in reciprocal space. In this case, the Fourier component  $K$  of the periodic modulation can compensate for the momentum mismatch between the fundamental and the second harmonic, if  $K = \Delta\beta$ .

The advantages of QPM stem from its decoupling of the phasematching from birefringence. With QPM, an appropriate choice of the modulation period allows use of a nonlinear medium over its entire transparency range at any convenient operating temperature, taking advantage of any of the components of the nonlinear susceptibility tensor (including those coupling waves of the same polarization), and utilizing waveguides that support only a single polarization. In the case of lithium niobate, interactions involving wavelengths between 350 nm and 4  $\mu\text{m}$  are possible using the nonlinear coefficient  $d_{33}$  ( $= 34 \text{ pm/V}$ ) which is 7 times larger than the birefringently phasematchable  $d_{31}$ . Proton exchanged waveguides, which are highly resistant to photorefractive damage but guide waves only of the extraordinary polarization, are used to advantage in lithium niobate and lithium tantalate quasi-phasematched nonlinear devices.

The barrier to widespread use of QPM has been the difficulty in reversing the sign of the nonlinear coefficient every coherence length. For typical visible interactions,  $l_c$  is on the order of several microns, precluding the conceptually simplest implementation of QPM, i.e. slicing the crystal into wafers of thickness  $l_c$ , rotating every other wafer by  $180^\circ$ , and reassembling the stack to obtain the desired periodic modulation of  $d_{eff}$ . In many ferroelectric crystals, reversal of the orientation of a domain of spontaneous electric polarization is equivalent to a rotation by  $180^\circ$  around one of the crystallographic axes. Thus, recently developed techniques for lithographically patterning the orientation of ferroelectric domains allow monolithic implementations of such a periodically reversed stack of nonlinear plates. These techniques are discussed in section 4.

### 3. Theory of Waveguide QPM Devices

Consider second harmonic generation (SHG) involving a single mode at both frequencies in a waveguide extending over the range  $0 \leq z \leq L$ . The output of the device ideally scales with the square of the length and the input power, so it is convenient to define a normalized conversion efficiency  $\eta_{nor}$  that is independent of these parameters. The output power at the second harmonic, in the undepleted pump limit, is given by

$$P_{2\omega} = \eta_{nor} P_{\omega}^2 L^2 \text{sinc}^2(\Delta\beta L/2) \quad (1)$$

where the propagation constant mismatch is  $\Delta\beta = \beta_{2\omega} - 2\beta_{\omega} = 2\pi/l_c$  and  $l_c$  is the coherence length. In a conventionally phasematched device,  $\eta_{nor}$  is given by [Stegeman (1985)]

$$\eta_{nor} = \frac{8\pi^2 d_{eff}^2}{n^2 n_{2\omega} c \epsilon_0 \lambda^2} \left| \int_{A_{\infty}} E_{2\omega}^* E_{\omega}^2 dx dy \right|^2 \quad (2)$$

where  $d_{eff}$  is the appropriate component of the nonlinear coefficient tensor,  $n$  is the index of refraction at the frequency specified by the substrate,  $\lambda$  is the free space wavelength, and  $A_{\infty}$  is an infinite cross section normal to  $z$ . The modal electric field profiles  $E$  of the assumed weakly guiding waveguide are normalized so that  $\langle |E|^2 \rangle = 1$ , where  $\langle f \rangle$  is defined as the integral of  $f$  over  $A_{\infty}$ .

In ideal periodically-poled ferroelectrics, the linear properties and the magnitude of the nonlinear susceptibility of adjacent domains are the same, but the sign of the nonlinear susceptibility is reversed. We can describe the spatial dependence of the nonlinear susceptibility in such a medium as  $d_{eff}(x, y, z) = |d_{eff}|g(x, y, z)$ , where  $g(x, y, z)$  takes the values  $\pm 1$ . Ideally  $g(x, y, z)$  is periodic in  $z$ , in which case the output power is given by [Jaskorzynska (1986)], [Fejer (1989)]

$$P_{2\omega} = \eta_{nor} P_{\omega}^2 L^2 \text{sinc}^2[(\Delta\beta - K_m)L/2] \quad (3)$$

where

$$\eta_{\text{ov}} = \frac{8\pi^2 d_{\text{eff}}^2}{n_x^2 n_{z_0} c \epsilon_0 \lambda^2} \left| \int_{-\Lambda}^{\Lambda} G_m E_{2m}^* E_m^2 dx dy \right|^2 \quad (4)$$

the  $K_m = 2\pi m/\Lambda$ ,  $\Lambda$  is the period of  $g(x, y, z)$ , and  $G_m(x, y)$  is a Fourier component of  $g(x, y, z)$  defined by

$$G_m = \frac{1}{\Lambda} \int_0^{\Lambda} g(x, y, z) \exp(-iK_m z) dz \quad (5)$$

From Eq. (3) we see that if one of the spatial harmonics of the modulation is equal the propagation constant mismatch, the second harmonic power grows quadratically with the length.

From Eq. (4) we see that the overlap integral involves the appropriate Fourier component of the modulation. If  $g$  is a 50% duty cycle square wave independent of  $x$  and  $y$ ,  $G_m$  is simply  $2/m\pi$  for  $m$  odd and vanishes for  $m$  even, so the conversion efficiency of the QPM interaction is the same as that of a conventional interaction with a nonlinear coefficient  $d_Q$  where

$$d_Q = \begin{cases} 2/m\pi & m \text{ odd} \\ 0 & m \text{ even} \end{cases} \quad (6)$$

More generally, defining a duty cycle  $D = l/\Lambda$  where  $l$  is the length of one of the positive domains,  $G_m$  is given by

$$G_m = \frac{2}{m\pi} \sin(m\pi D) \quad (7)$$

Note that even order QPM is possible for  $D \neq 1/2$ . For both both odd and even order QPM,  $d_Q = 2d_{\text{eff}}/m\pi$  for the optimum  $D$ .

When the domains are not homogenous throughout the cross section of the waveguide, so that  $D$  is a function of  $x$  and  $y$ , the overlap integral in Eq. (4) can be seriously reduced, as will be seen in the discussion of actual domain structures, especially in lithium niobate.

## 4. Ferroelectric Crystals for QPM Devices

### 4.1. GENERAL REQUIREMENTS

The essential step in the fabrication of a quasi-phaseshifted frequency conversion device, patterning of the nonlinear susceptibility, has been demonstrated in several types of media. Among these are stacks of discrete plates, [Thompson (1976)] centrosymmetric media with periodic electric fields applied to break the symmetry and induce an effective nonlinear susceptibility (electric field induced second harmonic generation), [Levine (1975)] III-V semiconductor films with alternating high and low nonlinearity for surface emitting devices, [Normandin (1990)] modulation of the free carrier density by proton implantation damage in media with carrier-induced nonlinearities, [Yoo (1991)], and polymers poled with periodic fields [Khanarian (1990)].

The approach that is emphasized in this paper involves the creation of periodic arrays of domains of spontaneous electric polarization in ferroelectric crystals. The sign of the nonlinear susceptibility depends on the domain orientation, so such an array of domains of sufficiently small period can be used for QPM. This technique adds two further practical considerations to the list of material requirements: A waveguide fabrication technique compatible with the domain reversal process must

be available, as should be substrates adequate for carrying out the lithographic patterning of the domains and the waveguides. At present, only three crystals meet these requirements, lithium niobate, lithium tantalate, and potassium titanyl phosphate (KTP).

Of these three crystals, lithium niobate has the largest nonlinear coefficient,  $d_{33} = 34$  pm/V, while lithium tantalate and KTP have 25 pm/V and 14 pm/V, respectively. On the other hand, KTP has the lowest susceptibility to photorefractive damage, while lithium niobate has the highest. Both lithium niobate and lithium tantalate are widely used for applications such as surface acoustic wave devices, and so are available commercially in 75 mm wafers, while KTP, applied primarily as a bulk second harmonic convertor, is available in wafers up to 20 mm.

The discussion in this paper emphasizes lithium niobate as it is thus far the best characterized of these materials. Excellent results have been obtained in lithium tantalate and KTP, which will be discussed briefly here for comparison with the lithium niobate results. Quasi-phasematched interactions in KTP are discussed in more detail in the paper by Bierlein in these proceedings.

#### 4.2. LITHIUM NIOBATE

Lithium niobate is a well known ferroelectric crystal, with useful piezoelectric, electrooptic, and nonlinear optical properties. A large body of work on waveguide technology in lithium niobate is available in the literature. [Korotky (1987)]

4.2.1. *Domain Reversal.* As-grown lithium niobate crystals are multi-domain, and must be poled through application of fields of several V/cm as the crystal is cooled through its Curie temperature ( $\approx 1150^\circ\text{C}$ ). The field necessary to reverse the orientation of the domains (the coercive field) increases with decreasing temperature, until at room temperature, the domains are essentially frozen, and cannot be reversed without the application of extremely large fields. [Haycock (1987)] Attempts to pattern the domains at elevated temperatures with periodic electric fields applied with interdigital electrodes have proved unsuccessful due to diffusion of the electrodes during the poling process.

Several techniques have been used successfully to controllably induce domain reversal. Patterned in- or out-diffusion of certain dopants leads to a corresponding distribution of reversed domains. One approach was suggested by the work of Miyazawa, who observed that inconsistent performance of electrooptic devices based on Ti-indiffused waveguides in +Z cut substrates could be explained by irregular domain reversals in the Ti-rich regions. [Miyazawa (1979)] We found that reproducible periodic domain reorientations could be induced by indiffusion of a Ti film patterned into a grating of the desired period by standard lithographic techniques on a +Z substrate at temperatures close to the Curie temperature. [Lim (1989)] The penetration depth of the domains is limited to typically one third of their period by lateral diffusion of the dopant. A similar process, based on out-diffusion of Li from +Z substrates through a periodic  $\text{SiO}_2$  mask, has also been successfully applied, though interaction of the mask with the substrate at the elevated processing temperatures has been problematic. [Webjörn (1989a)] The microscopic mechanism responsible for the domain reorientation is not yet clear. One explanation consistent with the experimental observations is that Li and Ti, which both reduce the Curie temperature of lithium niobate, reduce the coercive field at a given process temperature sufficiently that the domains can be reoriented by the thermoelectric fields generated by the temperature gradients present in the substrate during cooling from the process temperature. [Fejer 1989] The triangular shape of the domains, also unexplained to date, has a significant effect on the device performance, as is discussed in section 4.2.3.

Another class of domain reversal process is based on irradiation of the substrate by an electron beam. In the work of [Keys (1990)], periodic domains were generated by exposing a lithium niobate substrate with a 10 kV e-beam through a gold mask at a temperature of 580 °C. Recently, domain reversal through an entire 1 mm thick substrate exposed to a focused, scanned 25 kV electron beam at room temperature was demonstrated. [Yamada (1991)] Device applications of these materials has not yet been reported.

Pyroelectric fields generated by cooling substrates patterned with a periodic electrode structure have been used to produce domains with periods as small as 6.5  $\mu\text{m}$ . [Seibert (1990)] This technique can be applied to Y-cut substrates, and has the additional advantage of a low process temperature,  $\approx 400^\circ\text{C}$ .

**4.2.2. Waveguide Fabrication.** A suitable waveguide technology for quasi-phasematched devices must be compatible with the domain reversal process and be capable of supporting the high optical powers typical of nonlinear interactions. Of the ions commonly used to form lithium niobate waveguides, i.e.  $\text{Ti}^{4+}$  and  $\text{H}^+$  indiffusion and  $\text{Li}^+$  outdiffusion,  $\text{H}^+$  appears best suited to QPM applications, as proton exchange is a low temperature process ( $< 400^\circ\text{C}$ ) and produces waveguides resistant to photorefractive damage, and thus have been used for most devices fabricated to date. Proton exchanged (PE) waveguides are formed by immersing a lithium niobate substrate in a molten acid, often benzoic though a number of other acids have been used, at a temperature generally below  $300^\circ\text{C}$ . The PE waveguides, produced by the exchange of Li in the substrate for H from the acid, have step-like refractive index profiles and large changes in the extraordinary refractive index ( $\Delta n = 0.125$  at 633 nm). [Jackel (1982)] Though only the z-polarized modes are guided (the ordinary index of refraction is reduced by PE) the waveguides are suitable for quasi-phasematched interactions, which generally take advantage of the largest nonlinear coefficient,  $d_{33}$ . Channel waveguides are formed by carrying out the exchange through a mask, often Al,  $\text{SiO}_2$ , or, for some of the stronger acids, Ta. While the tight confinement obtained with this large  $\Delta n$  is attractive for maintaining a small effective area for nonlinear interactions, a significant reduction in the nonlinear susceptibility accompanying PE (discussed in more detail in section 4.2.3) necessitates a post-exchange annealing step to reduce the proton concentration. [Suchoski (1988)] The fabrication and modeling of these annealed proton exchange (APE) waveguides is complicated by concentration dependent diffusion coefficients and a nonlinear dependence of the index of refraction on the proton concentration, and remains a topic of current research. [Howerton (1991)]

**4.2.3. Devices.** Two approaches have been taken to the fabrication of QPM SHG devices. In one, the domain reversal pattern is formed first, typically by a high temperature process like patterned Ti-indiffusion, then a waveguide is formed, typically by a low temperature process like APE. [Lim (1989b)] In the other, the domain reversal and the waveguide fabrication are accomplished with a single process. [Webjörn (1989b)] For example a periodically patterned Ti strip can simultaneously block Li-outdiffusion to achieve the patterned domain inversion, and can itself indiffuse to produce a waveguide. While the latter type of process is in principle simpler, the flexibility of the former facilitates optimization of the entire device, and has generally led to better performance. This discussion therefore emphasizes devices based on two-step processes.

The first quasi-phasematched interaction demonstrated in periodically-poled waveguides was SHG, including planar devices for 532 nm generation [Lim (1989)] and channel devices for blue light generation. [Lim (1989b)] [Webjörn (1989a)] The process for a typical blue light device involves first patterning a 5 nm thick Ti film into a grating with the desired domain period (6 – 8  $\mu\text{m}$  for third order QPM), then indiffusing the Ti at  $1100^\circ\text{C}$  for 15 min in a closed crucible filled

with lithium niobate powder to suppress Li out-diffusion, to produce a substrate with periodically reversed domains. A channel mask of Al or SiO<sub>2</sub> is then patterned on the substrate, and a 1.5 h exchange at 160 °C is carried out in a benzoic acid bath. The mask is then stripped off and a 4 hour anneal at 333°C is the final process step.

We find that the typical efficiency of such a device generating 410 nm radiation is 40 %/W-cm<sup>2</sup>. In 1 mm long devices, the full width half-maximum (FWHM) of the wavelength tuning curves is close to the theoretical 4 Å-mm, but for 10 mm long devices, the bandwidth narrows only to ≈ 1.5 Å, indicating an effective length ≈ 3 mm. The output power increased quadratically with pump power up to the maximum power that we have observed, 1.1 mW in a TM<sub>00</sub> mode. Photorefractive effects were not observed in this device or in multimode outputs as large as 3 mW.

The efficiency of these devices is substantially smaller than the 350%/W-cm<sup>2</sup> calculated from the overlap integral for an ideal device. Several factors contribute to the lowered efficiency. Approximately a factor of two comes from the nonideal, triangular shape of the domains, which reduces the overlap integral as described in Eqs. (4) and (7). Another major factor is the effect of the proton exchange on the nonlinear susceptibility of the lithium niobate. Several studies indicate a reduction of 40% – 60% in the nonlinear susceptibility of PE waveguides, with recovery of the nonlinearity after adequate annealing. [Suhara (1989)] [Keys (1990b)] [Cao, X.(1991)] We suspect that the nonlinear susceptibility may be almost completely eliminated prior to annealing. In earlier work, we attempted to fabricate quasi-phaseshifted devices in PE waveguides, and found no visible second harmonic output even with several hundred watt peak powers coupled into the waveguide, suggesting a very small nonlinear susceptibility. This conclusion is supported by reflected second harmonic generation measurements of 266 nm radiation, which also indicate a greater than tenfold reduction. Clarification of the discrepancy between these measurements, and further characterization of the annealed proton exchange process are important for optimizing the tradeoff between high proton concentrations for tight confinement and good overlap with the shallow domain reversed region, and low proton concentrations to recover the nonlinear susceptibility.

The limited effective length presumably is a result of inhomogeneities in the width or depth of the waveguides, which affect the phase velocities of the fundamental and the second harmonic modes differently, and hence lead to axial variations in the wave vector mismatch. Tolerances for such inhomogeneities are in general rather strict, but recent work in "noncritical" waveguide designs with no first order dependence of the mismatch on small variations in the waveguide dimensions indicate that this problem is manageable if adequate information on the waveguide fabrication process are available. [Lim (1990)]

The largest reported output power for quasi-phaseshifted SHG in lithium niobate waveguides is 4 mW, at a fundamental input power of 115 mW in a 9 mm long device, for a normalized conversion efficiency of 40%/W-cm<sup>2</sup>, [Nada (1991)] typical of the efficiencies reported for third order quasi-phaseshifted devices in lithium niobate. [Lim (1989b)] The highest efficiency reported for a quasi-phaseshifted interaction in lithium niobate is 60%/W-cm<sup>2</sup>, in a second order device. [Srivastava (1991)] The efficiency of first order devices has been rather low, largely because of poor overlap of the waveguide modes with the shallow domains. [Webjörn (1989a)]

Some interesting variations on the basic periodic domain and channel waveguide device have been reported. A fan shaped array of domains has been designed to make available a continuously varying periodicity to facilitate optimization of the period. [Ishigame (1991)] A device in which the Ti grating served both to create the domain reversals and as a Bragg mirror for an external cavity laser has also been reported. [Shinozaki (1991)]

In addition to SHG devices for visible light applications, difference frequency mixers have been fabricated for the generation of infrared radiation from two near infrared sources. Such devices using birefringent phasematching have been reported previously, but generally have required operating temperatures of several hundred degrees. With QPM, it has been possible to produce 2.1  $\mu\text{m}$  radiation at room temperature by differencing a 0.81  $\mu\text{m}$  pump and a 1.3  $\mu\text{m}$  signal. [Lim (1991)] The fabrication process is similar to that described for doublers, except that the weaker dispersion in the infrared allows a first order device with a 21  $\mu\text{m}$  domain period. With 160 mW at 0.81  $\mu\text{m}$  and 1 mW at 1.3  $\mu\text{m}$  in a 7 mm long device, an output of 1.8  $\mu\text{W}$  at 2.1  $\mu\text{m}$  was observed, for a normalized conversion efficiency of  $\sim 4\%/W\text{-cm}^2$ . This efficiency is again substantially smaller than the  $\sim 100\%/W\text{-cm}^2$  that could be expected in an ideal device. The long domain periods lead to deep domain reversals, leading us to again suspect the effects of the reduced nonlinear susceptibility.

#### 4.3. LITHIUM TANTALATE

Lithium tantalate is a ferroelectric crystal isostructural with lithium niobate, with a lower Curie temperature ( $= 610^\circ\text{C}$ ), somewhat smaller nonlinear susceptibilities, and markedly less sensitivity to photorefractive damage. It is widely applied in surface acoustic wave devices, leading to ready availability of 75 mm substrates.

**4.3.1. Domain Reversal.** Similar to lithium niobate crystals, as-grown lithium tantalate is polydomain and must be poled by application of an electric field as the crystal is cooled through  $T_c$ . Unlike lithium niobate, lithium tantalate has a sufficiently low  $T_c$  that successful periodic poling with interdigital electrodes has been possible, for both surface acoustic wave [Nakamura (1983)] and quasi-phasematched optical devices. [Matsumoto (1991)] Choice of electrode material and poling conditions is important for minimization of electrode indiffusion. We have found that application of 1.4 V to Au/Ti electrodes separated by 3.5  $\mu\text{m}$  at temperatures close to  $T_c$  yields well-reversed domains with minimal surface damage. [Matsumoto (1991)] The cross section of these domains more closely approaches the ideal rectangular shape than do lithium niobate domains.

A patterned dopant technique based on proton exchange through a periodic mask followed by annealing has also yielded well-formed domains in lithium tantalate. [Nakamura (1990)] [Mizuuchi (1991)] [Åhlfeldt (1991)] In a typical process, proton exchange is carried out on the -Z face through a periodic mask at  $220^\circ\text{C}$  for several hours, followed by a soak at  $550^\circ\text{C}$  to reverse the domains and diffuse the protons. Subsequent fabrication of an annealed proton exchange waveguide does not appear to disturb the domain pattern. The shape of the domains produced by this technique is nearly semicircular and typically allows somewhat better overlap with the waveguide modes than do the triangular domains produced by Ti-doping in lithium niobate.

**4.3.2. Waveguide Fabrication.** The low  $T_c$  of lithium tantalate precludes most metal indiffusion techniques for waveguide fabrication in quasi-phasematched devices. All quasi-phasematched devices reported to date have been based on annealed proton exchange waveguides. The basic process for these waveguides is similar to that described for lithium niobate, except for a slower diffusion rate and a smaller change in the refractive index. [Tada (1987)] [Findakly (1988)] Proton exchange in lithium tantalate is less well characterized than in lithium niobate, but appears to share much the same complexity. We have found that, as in PE lithium niobate, no SHG was observed in PE lithium tantalate waveguides, even with Q-switched laser pulses. It appears that there is again



a reduction in the nonlinear susceptibility accompanying PE that must be corrected with a post-exchange annealing step, though no quantitative measurements of the nonlinear susceptibility have been reported to date.

#### 4.3.3. Devices

In the past year, several groups have demonstrated quasi-phasematched SHG of blue light in lithium tantalate waveguides periodically-poled by patterned proton exchange. [Mizuuchi (1991)] [Ählfeldt (1991)] Of these, the highest output power reported to date was 12 mW of blue output in the TM<sub>00</sub> mode for an input of ~180 mW in a 1 cm long waveguide, for a conversion efficiency of 40%/W-cm<sup>2</sup>. [Yamamoto (1991)]. No evidence of photorefractive damage at these power levels was reported.

SHG of blue light has also been reported in waveguides poled by application of periodic fields with interdigital electrodes. [Matsumoto (1991)] The normalized efficiency of these devices was 8 %/W-cm<sup>2</sup>, smaller than the 22%/W-cm<sup>2</sup> predicted from the overlap integral. It is likely that the discrepancy arose from a reduced nonlinear susceptibility due to inadequate annealing of the waveguide. Further characterization of the properties of APE waveguides in lithium tantalate will be important for systematic optimization of these devices. One interesting feature of this poling process is that it works on X-cut substrates as well as on Z-cut, unlike the doping techniques. X-cut substrates allow propagation of TE modes in APE waveguides, an advantage for efficient coupling to the TE output modes typical of diode lasers.

#### 4.4. KTP

KTP is a ferroelectric crystal ( $T_c = 950^\circ\text{C}$ ) developed primarily for nonlinear optical applications. [Bierlein (1989)] It is grown by both flux and hydrothermal techniques, with substrates as large as 20 mm available commercially. While the nonlinear susceptibility  $d_{33} = 14$  pm/V of KTP is smaller than that of both lithium niobate and lithium tantalate, KTP has the advantage of a very small sensitivity to photorefractive effects. Waveguide fabrication is generally by exchange of potassium with alkali metal ions from molten salts. [Bierlein (1987)] The diffusion of these ions is highly anisotropic, allowing the fabrication of deep structures along the Z-axis with little lateral diffusion. The only domain reversal technique reported to date is a patterned dopant method, involving exchange with Ba ions. The anisotropy of the diffusion process allows the creation of domains with a nearly ideal rectangular cross-section. In such devices, the waveguide generally is defined by the same ion exchange that creates the domain reversal. The "segmented" waveguide formed in this way apparently has little excess loss, particularly if the dimensions are chosen such that the waveguide is multimode at the pump wavelength. [van der Poel (1990)]

Efficiencies as large as 200%/W-cm<sup>2</sup> in 5 mm long waveguides have been reported, as have output powers in the blue of 22 mW for > 400 mW of fundamental power in the waveguide. [Bierlein (1991)] Efficient sum frequency generation of visible radiation has also been demonstrated in periodically-poled KTP segmented waveguides. [Laurell (1991)]

### 5. Summary and Prospects

Since the first demonstration of quasi-phasematched SHG in periodically-poled ferroelectric waveguides in 1989, rapid progress in the field has been driven by the demand for compact

sources of coherent radiation at wavelengths currently inaccessible to room temperature diode laser technology. Over the past two years, output powers in the visible have increased from tens of nanowatts to 22 mW, and operation has been extended to the infrared by difference frequency generation.

In the lithium niobate and lithium tantalate systems, device performance remains well below the theoretical potential. Difficulties arising from the effects of proton exchange on the material properties must be addressed either through more detailed characterization of the PE and APE processes, or the development of new waveguide technologies compatible with the domain reversal process and capable of supporting the high intensities involved in these interactions. The limitations on the effective length due to inhomogeneities in the dimensions of the waveguides appear to be manageable with careful processing and the use of noncritical waveguide designs. The triangular domain shapes produced in lithium niobate by periodic Ti doping remain a difficulty, especially for first order devices, but new poling techniques are emerging that promise better aspect ratios and more rectangular domains. Photorefractivity remains an open question, but powers in the blue in excess of 4 mW in lithium niobate and 12 mW in lithium tantalate without evidence of photorefractive damage have been reported. It is likely that the domain structure is affecting the photorefractive sensitivity, perhaps through the periodic changes in the sign of the electrooptic coefficients and the Glass constant.

Another important issue for eventual practical applications is coupling of the pump laser to the frequency conversion device. Both alignment tolerances and the effects of feedback from the waveguide to the laser must be addressed. Solutions based on incorporating one of the laser mirrors in or on the frequency convertor appear promising. More desirable, but requiring considerable progress in materials technology, is the growth of the frequency convertor directly on the same substrate as the pump laser.

## 6. References

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