

Quasi-phase-matched second-harmonic generation of blue light in periodically poled LiNbO₃

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LiNbO₃ crystals with periodically alternating ferroelectric domains have been produced using laser-heated pedestal growth. Domain thicknesses as small as 1 μm have been achieved. This material was applied to room-temperature, quasi-phase-matched frequency doubling to generate light at wavelengths as short as 407 nm, using the d_{33} and d_{22} nonlinear coefficients. The measured conversion efficiencies and wavelength and temperature tuning bandwidths are consistent with an effective interaction length of $\approx 320 \mu\text{m}$ (> 230 domains). An initial test with high-intensity focused blue beams showed that the periodically poled material exhibits no discernible photorefractive damage effect.

Materials with high nonlinearity, good optical quality, and the ability to phase match for direct frequency doubling of GaAlAs diode lasers are in demand for the development of compact sources of short-wavelength light to be used in optical storage and other applications. Lithium niobate (LiNbO₃) has insufficient birefringence to use conventional phase-matching techniques at these wavelengths. We have demonstrated the capability of growing periodic ferroelectric domain structures of high spatial frequency in LiNbO₃. Using quasi-phase-matching in this material, we have accomplished collinear cw second-harmonic generation (SHG) with wavelengths and nonlinear coefficients which would have been impossible to phase match using only the material birefringence.

Quasi-phase-matching (QPM) was the first technique to be suggested for compensating refractive-index dispersion to obtain efficient SHG.¹ In this technique, the nonlinear coefficient is modulated spatially with a period $\Lambda = 2ml_c$, where $m = 1, 3, 5, \dots$ is the "order" of QPM and l_c is the coherence length, given by

$$l_c = \lambda / 4(n_2 - n_1), \quad (1)$$

where λ is the fundamental wavelength, and n_1 and n_2 are the refractive indices at the fundamental and second-harmonic wavelengths, respectively. The condition for QPM is

$$\mathbf{k}_2 - 2\mathbf{k}_1 - \mathbf{K} = 0, \quad (2)$$

where \mathbf{K} is the wave vector of the nonlinear coefficient grating, having the magnitude $K = 2m\pi/\Lambda$, and \mathbf{k}_1 and \mathbf{k}_2 are the wave vectors for the fundamental and second-harmonic waves. The effective nonlinear coefficient is determined by the amplitude of the pertinent spatial-frequency component of the periodic structure. If the modulation consists of alternating the sign of the nonlinear coefficient d_{ij} at equal intervals, then

$$d_{\text{eff}} = (2/m\pi)d_{ij}. \quad (3)$$

It is thus desirable to use the lowest-order QPM possible, where $m = 1$ and $\Lambda = 2l_c$, in order to obtain the highest conversion efficiency, which is proportional to d_{eff}^2 . In LiNbO₃, the sign of the nonlinear coefficient can be reversed either by rotating a slice of the crystal through 180° around the x axis, or by creating an oppositely polarized ferroelectric domain, which is crystallographically equivalent to this

rotation. Quasi-phase-matched SHG in LiNbO₃ has been previously demonstrated in Czochralski-grown periodically poled LiNbO₃ at $\lambda = 1.06 \mu\text{m}$, for which $l_c = 3.4 \mu\text{m}$,² and, more recently, using periodically reversed domains in an integrated-optic geometry.³

Small-diameter single crystals of LiNbO₃ are produced by laser-heated pedestal growth using the apparatus described in Ref. 4. In this technique, 10.6 μm radiation from a CO₂ laser is focused on the tip of a typically 500-μm-diam single-crystal source rod of LiNbO₃ to make a small molten droplet into which a seed crystal is dipped. The seed, which determines the growth orientation, is then withdrawn from the droplet as fresh source material is supplied; the ratio of the pull rate to the feed rate determines the average diameter of the growing crystal, which in these experiments was approximately 250 μm. Crystals are grown either in air or in a 4:1 mixture of helium and oxygen. Periodically reversed ferroelectric domains with boundaries following the growth interface shape, as shown in Fig. 1, are created during growth by modulating the freezing interface position, either by ro-

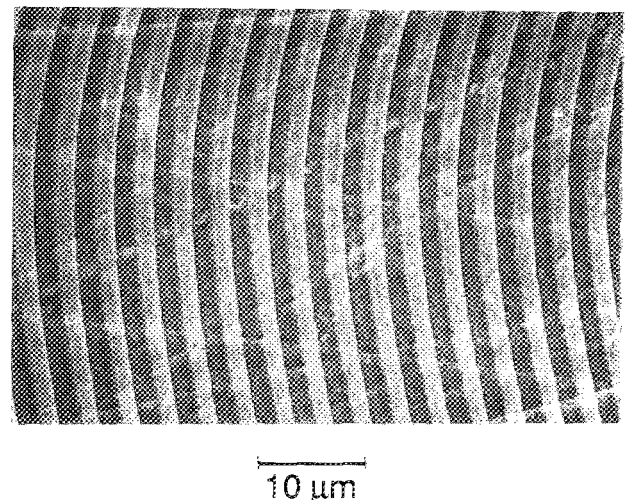


FIG. 1. SEM photograph showing periodically alternating domains with period $\sim 3.5 \mu\text{m}$ as revealed by polishing and etching the y face of a pedestal-grown crystal. The crystal pull direction (along the x axis) is to the right in the photograph. The curvature of the domains is due to the convex shape of the growth interface. Straight lines are scratches from polishing.

tating an asymmetric heat input produced by a rotating mask or by periodically modulating the heating power. The period of the domain structure in μm is given by $\Lambda = v_{\text{pull}}/f_{\text{mod}}$, where v_{pull} is the crystal pull rate in $\mu\text{m/s}$ and f_{mod} is the frequency of modulation in Hz. Using these techniques, we have grown domain structures with periods as small as $2\ \mu\text{m}$, in either nominally 5% Mg-doped or undoped congruent material, and in crystals grown along either the x or z axis.

Modulation of the position of the freezing interface corresponds to a modulation of the microscopic growth rate at the interface. Components or dopants with an effective segregation coefficient $k_{\text{eff}} \neq 1$ are, as a result, incorporated into the growing crystal with a periodically varying concentration.⁵ Sections of the length of the growing crystal with composition gradients of the correct sign and sufficient magnitude to overcome steady-state self-generated poling fields⁶ form reversed ferroelectric domains. The mechanisms through which composition gradients influence the domain formation is a topic of current research.⁷ The large axial temperature gradients and high growth rates present during laser-heated pedestal growth, together with the high Curie temperature of the material, aid in the formation of well-defined domain walls near the freezing interface.

The sample used in the optical experiments was a Mg-doped⁸ x -axis LiNbO_3 crystal approximately $250\ \mu\text{m}$ in diameter, grown at a pull rate of $v_{\text{pull}} = 2\ \text{mm/min}$. The heat input was modulated using a periodically triggered electro-mechanical shutter with $f_{\text{mod}} = 12.35\ \text{Hz}$, resulting in a domain structure of period $\Lambda = 2.7\ \mu\text{m}$. This period was picked so that SHG using either the d_{33} or d_{22} nonlinear coefficient could be first-order quasi-phase-matched within the wavelength tuning range of a cw Styryl-9 dye laser. After annealing in flowing dry O_2 for 3 h at $725\ ^\circ\text{C}$ to increase the crystal transparency without erasing the periodic domain structure, the sample was glued into a 2 mm length of glass capillary tubing and polished on the x faces to a length of 1.77 mm.

Figure 2 shows phase-matching curves obtained by plotting the second-harmonic power while scanning the dye laser wavelength λ . The d_{33} nonlinear coefficient was addressed by polarizing the x -propagating fundamental wave linearly along the crystal z axis; then, without moving the sample, the d_{22} coefficient was addressed by rotating the polarization 90° so that it was parallel to the y axis. The similarity of the shapes of these two curves suggests that phase matching occurs in the same region of the sample, and that both n_o (d_{33}) and n_o (d_{22}) have similar optical quality, in spite of the many domain walls and the modulated composition. We confirmed that the generated second-harmonic wave has the same polarization as the fundamental wave for both these coefficients. QPM occurred at $\lambda = 814.5\ \text{nm}$ for d_{33} , and at $850.5\ \text{nm}$ for d_{22} . The effective interaction length l_{eff} inferred from the width of these tuning curves⁹ is limited by the depth of focus of the $5\times$ microscope objective (focal length $\approx 30\ \text{mm}$) used to obtain them.

To determine l_{eff} as limited by the uniformity of the periodicity of the domain structure, the $5\times$ objective was replaced by a 65 mm focal length lens. With this lens, the

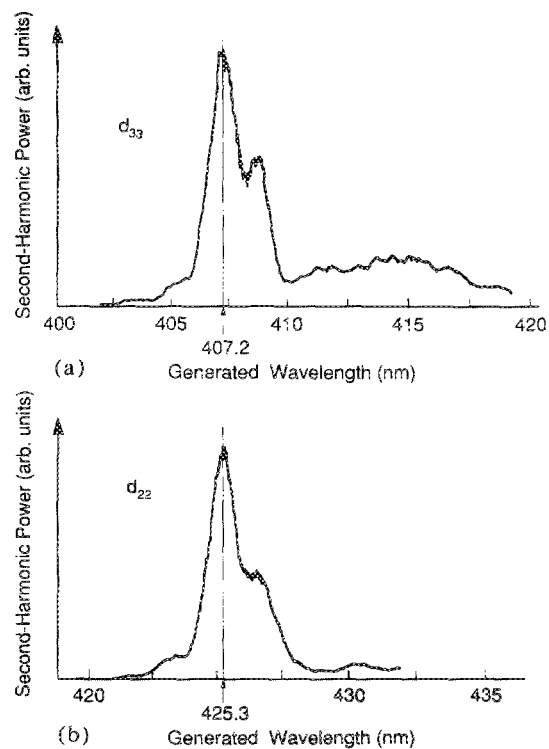


FIG. 2. Second-harmonic power vs wavelength tuning curves obtained with $5\times$ microscope objective for (a) d_{33} and (b) d_{22} .

confocal parameter of the fundamental beam within the crystal was 1.3 mm. The width of the d_{33} and d_{22} wavelength-tuning curves indicated an $l_{\text{eff}} \approx 320\ \mu\text{m}$. Temperature-tuned phase-matching curves for both the nonlinear coefficients were also obtained by placing the sample in a heated mount. The temperature-tuned widths were found to be consistent with the above l_{eff} when values for dn/dT from Ref. 10 were used. We conclude that in this sample the effective length is due to the nonuniformity of the domain periods, since it is significantly shorter than both the overall length of the sample and the confocal parameter of the fundamental beam.

The theoretical conversion efficiency for d_{33} interaction was calculated using $l_{\text{eff}} = 320\ \mu\text{m}$ and $d_{\text{eff}} = 22\ \text{pm/V}$, obtained by inserting $m = 1$ and the value for d_{33} tabulated by Kurtz *et al.*¹¹ into Eq. (3). The conversion efficiency measured using the 65 mm lens agreed with this calculation within 20%. Using the $5\times$ lens, which produced nearly optimal focusing, we measured a maximum conversion efficiency, corrected for Fresnel reflection at the crystal faces, of 8×10^{-5} . This efficiency was obtained with 83 mW of input fundamental power and $6.7\ \mu\text{W}$ of blue light generated within the crystal. The second-harmonic power was quadratic with input power over the entire range measured. The normalized theoretical conversion efficiency using d_{33} and QPM with optimal focusing is calculated to be $0.52\% \text{ W}^{-1} \text{ mm}^{-1}$, or 13.5 times that which would be obtained were it possible to birefringently phase match a d_{31} interaction at this wavelength.

It was surmised that the photorefractive properties of the periodically poled material might be altered by the rapid-

ly alternating sign of charge transport or by fields present at the many domain walls. This question is of great importance in applications of LiNbO₃ at visible wavelengths. No photorefractive effect was observed in periodically poled Mg-doped or undoped LiNbO₃ with the relatively low levels of blue light generated in the SHG experiments. Therefore, a separate experiment was performed to compare an undoped periodically poled sample with $\Lambda = 3 \mu\text{m}$ against an identically processed undoped uniformly poled sample. In this experiment, extraordinarily polarized 488 nm light from an argon-ion laser, propagating along the x axis, was focused to a beam waist of radius $w = 6 \mu\text{m}$ in the sample. A slit was centered in the transmitted beam and adjusted at very low power ($\sim 3 \mu\text{W}$) to block approximately half of the light. The slit was oriented so that beam spreading along the z axis, characteristic of the photorefractive effect,¹² would reduce the proportion of light transmitted by the slit. When the incident power was stepped up to 100 mW in the uniformly poled material, the fraction passed by the slit dropped to 37% of the low-power value in < 100 ms. In contrast, at the same power level in the periodically poled material, the transmitted fraction stayed at 100% of the low-power value, with no sign of change for measurement intervals of several minutes. After the test, the uniformly poled sample exhibited spots of persistently changed refractive index where it had been exposed to the blue beam, whereas there were no such marks in the periodically poled sample. These results indicate that periodically poled LiNbO₃ grown and processed by our methods should be useful for interactions involving hundreds of milliwatts of visible light.

Periodically poled LiNbO₃ can be engineered to quasi-phase-match nonlinear interactions at any desired temperature, using any nonlinear coefficient, and involving any wavelengths within the transparency range of the crystal. This control over its phase-matching properties makes the already important material LiNbO₃ potentially applicable to a much wider variety of interactions. The material is easy to grow and the domain structure is stable to at least several hundred degrees Celsius.

In future work, the uniformity of the domain-structure periodicity may be improved, and hence the effective interaction length increased, by upgrading the translators in the crystal puller to achieve better velocity stability. Other nonlinear interactions utilizing QPM will be investigated, as well as the use of resonantly enhanced SHG¹³ to obtain milliwatts of blue light at input power levels available from laser diodes.

In summary, we have grown periodically poled LiNbO₃ with domains as thin as $1 \mu\text{m}$ using the laser-heated pedestal growth method. This material was used to demonstrate room-temperature, cw, quasi-phase-matched SHG using either the d_{33} or d_{22} nonlinear coefficients, at wavelengths as short as 407 nm. The measured conversion efficiencies and wavelength and temperature tuning bandwidths are all consistent with an effective phase-matched interaction length of $\approx 320 \mu\text{m}$ (> 230 domains). An initial test with high-intensity focused blue beams showed that the periodically poled material exhibits no discernible photorefractive effect.

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