

Depth profiling of the d_{33} nonlinear coefficient in annealed proton exchanged LiNbO₃ waveguides

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We report depth profiling of the d_{33} nonlinear coefficient in annealed proton exchanged LiNbO₃ waveguides using reflected second-harmonic generation from angle-lapped samples. At depths greater than the initial proton exchange depth the d_{33} coefficient retains nearly its bulk LiNbO₃ value, but within the initial proton exchange region the value of d_{33} varies with annealing time. These results reconcile previous conflicting measurements of the d_{33} coefficient and explain the variation in the efficiency of guided wave frequency conversion devices with annealing.

Quasi-phase matched (QPM) frequency conversion in LiNbO₃ waveguides is an efficient method for the generation of short wavelength radiation from infrared laser diodes. While different techniques have been used to form the ferroelectric domain grating necessary for QPM, the annealed proton exchange (APE) process has been used exclusively to provide waveguide confinement. Contradictory measurements of the nonlinear optical properties of proton exchange (PE) and APE-LiNbO₃ waveguides have been previously reported.¹⁻⁶ The d_{33} coefficient of PE-LiNbO₃ has been measured to be between 0 (Refs. 4 and 5) and 0.63 (Ref. 3) of the bulk LiNbO₃ value. The effect of annealing is similarly controversial, with measurements of d_{33} in APE-LiNbO₃ varying from 0% (Ref. 4) to 90% (Ref. 3) of the bulk value. Recently demonstrated^{7,8} guided-wave QPM second-harmonic generation (SHG) devices with normalized conversion efficiencies $> 600\%/W\text{ cm}^2$ indicate that APE-LiNbO₃ waveguides must have a large nonlinear coefficient.

In this letter we report measurements of the depth dependence of the d_{33} nonlinear coefficient in APE-LiNbO₃ waveguides using reflected SHG from angle-lapped samples. With a 532 nm fundamental wavelength, the 266 nm second-harmonic (SH) wavelength is above the APE-LiNbO₃ band edge so that only the SH generated within a 0.05 μm absorption depth⁴ of the surface is observed. The samples were angle lapped so that lateral position could be mapped onto depth into the sample, allowing depth profiling via lateral translation. This technique circumvented complications caused by the graded refractive index profile and the unknown spatial variation in the d_{33} coefficient in the APE-LiNbO₃. The depth profiles presented here explain the variation in the d_{33} values reported in the literature. Normalized conversion efficiencies calculated with our results are consistent with those observed from guided-wave QPM SHG devices.

APE waveguides were fabricated on *x*-cut LiNbO₃ by proton exchange in pure benzoic acid at 173 °C for 66 min to a depth of 0.42 μm , calculated using the results of Ref. 9, and annealed in air at 333 °C for varying times. The samples were then polished at a wedge angle < 2 mrad, with the actual relationship between lateral position and depth determined using surface profilometry. 532 nm radiation from an injection seeded Q-switched frequency-

doubled Nd-YAG laser was focused onto the sample at normal incidence with a 30 μm FWHM spot and a peak intensity of $< 150\text{ MW/cm}^2$. The samples were scanned under the focused spot using a motorized translation stage and the reflected 266 nm SH was detected using a dichroic mirror, a solid blind photomultiplier, and a gated integrator. The fundamental and SH fields were polarized parallel to each other and the *z* axis of LiNbO₃ and thus were coupled by the d_{33} nonlinear coefficient. To discriminate against polishing artifacts, each sample had an internal, unexchanged reference portion formed by masking half the sample with Al prior to proton exchange. The mask was removed by etching in NaOH prior to annealing. A portion of the sample remained unwedged, so that reflected SH could be obtained from the original surface. SHG and profilometry measurements were performed over the whole surface to eliminate errors associated with polishing skew. Figure 1 shows the sample orientation and geometry, with the dashed line indicating the beginning of the wedged portion of the sample and the origin for measurements of lateral position; negative values of lateral position represent data from the unwedged portion of the sample, i.e., the original surface.

Figure 2 shows the reflected SH power at 266 nm, normalized to the signal from bulk LiNbO₃ versus depth and lateral position for APE-LiNbO₃ waveguides annealed for 3, 5, 9, and 63 h. The error bars represent the variation in the depth measurement for 5 different scans. The figure inset shows the spatial step response of the detection system projected onto depth, measured by scanning the focused spot off the end of a bulk LiNbO₃ sample. This

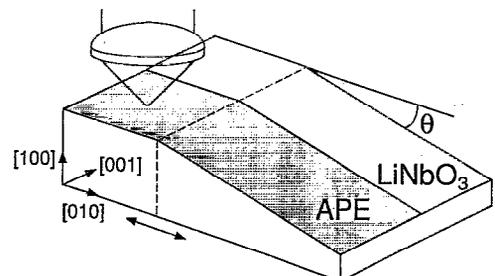


FIG. 1. Geometry of the wedged sample used for reflected SHG measurements.

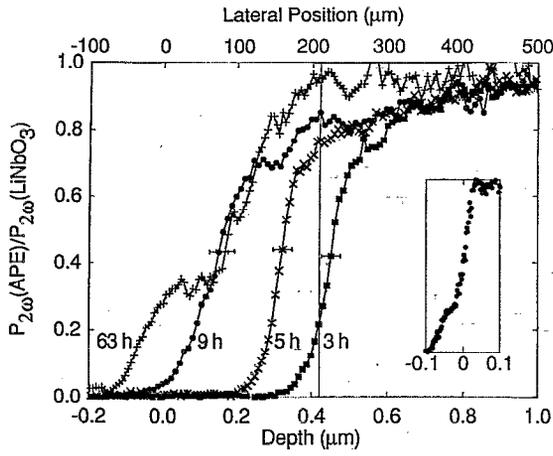


FIG. 2. Reflected 266 nm SH intensity, normalized to that from bulk LiNbO₃, vs lateral position (top scale) or depth into the waveguide (bottom scale) for samples proton exchanged to 0.42 μm and annealed for 3, 5, 9, and 63 h. The inset shows the step response of the detection system.

demonstrates a depth resolution of <0.05 μm. The samples displayed constant reflected SH over the entire unexchanged side with intensities equal to that from bulk LiNbO₃, with the exception of the sample annealed for 63 h, which had a polishing artifact between lateral positions -75 and +50 μm.

For the APE-LiNbO₃ sample annealed for 3 h no reflected SH signal was observed from the original surface or from the portion of the wedged surface corresponding to the original PE layer. However, there is an abrupt increase from 0% to 80% of the bulk LiNbO₃ value at a position corresponding to the interface between the original PE film and the LiNbO₃ substrate. At lateral positions corresponding to depths greater than the initial PE depth, the reflected SH power approaches that measured from bulk LiNbO₃. At depths >1 μm the measured reflected SH power is indistinguishable from that of bulk LiNbO₃.

With further annealing, the position of the abrupt increase in the reflected SH power moves toward the original surface, and the reflected power approaches that from bulk LiNbO₃ at shallower positions. No reflected signal was ever observed from the original surface regardless of annealing time, except from the sample annealed for 63 h at lateral positions where polishing damage was observed. However, no signal was observed over the rest of the unwedged portion of this sample.

The two previous measurements^{4,6} of the d_{33} coefficient of APE-LiNbO₃ waveguides performed using reflected SHG with a 532 nm fundamental source were made on unwedged samples, only probing a 0.05-μm-thick region at the original surface. Reference 2 reported no reflected SH regardless of annealing time at annealing temperatures of 350 °C, similar to our results when the probe is confined to the unwedged part of the sample. Reference 4 reported that APE-LiNbO₃ proton exchanged for 0.5 h and annealed for 1.5 h at 310 °C or for 1 h at 350 °C exhibited reflected SH with an intensity of 1/2 or 1/4 that obtained from bulk

LiNbO₃. For samples proton exchanged longer than 0.5 h, no reflected SH was observed, regardless of annealing time or temperature.

The absence of any reflected SH from the unannealed PE region could be explained by the formation of a PE-LiNbO₃ film with either inversion symmetry or a different $\chi^{(2)}$ tensor that did not result in any reflected SH. Similarly, the progression of the abrupt steplike recovery to the surface region with increasing annealing could be explained by the solid state epitaxial regrowth of the LiNbO₃ phase from the original interface between the PE film and the LiNbO₃ substrate. After the abrupt increase in the reflected SH from 0% to 80% of the bulk value there is a slower approach to the bulk value occurring over the next 0.5 μm in depth. In this region the measured d_{33} value correlated with concentration, independent of annealing time, so we conclude that for low concentrations the d_{33} coefficient is slightly reduced from its bulk value. While the details of the progression of the d_{33} recovery to the original surface may be dependent on the orientation of the LiNbO₃ substrate and annealing temperature, it is unlikely that the d_{33} coefficient of APE-LiNbO₃ waveguides fabricated on z- and x-cut substrates differ substantially at depths greater than the initial PE depth. Understanding the absence of reflected SH from the original surface of APE-LiNbO₃ may require microstructural and x-ray diffraction experiments to elucidate the crystal structure within the top 0.05 μm of the surface. This is of little practical importance since the modal amplitudes are very small at the surface. To design guided wave frequency conversion devices it is adequate to assume that at depths greater than the initial PE depth the d_{33} coefficient nearly retains its bulk value throughout annealing, while variations occur within the original PE depth.

The measurements of the d_{33} coefficient shown in Fig. 2 can be used to determine the normalized conversion efficiencies (η) for any guided wave nonlinear optical interaction. As an example, we compute η for SHG of $\lambda_{\omega}=850$ nm radiation in z-cut, x-propagating APE-LiNbO₃ waveguides fabricated like those shown in Fig. 2. We assume first-order QPM over an interaction length l with a uniform, depth-independent ferroelectric domain grating, which can be fabricated using an electric-field⁷ or electron-beam⁸ poling technique. With the output SH power given by $P_{2\omega}=\eta P_{\omega}^2 l^2$, η is given by

$$\eta = \frac{8\pi^2 (d_{\text{QPM}})^2}{n_{\omega}^2 n_{2\omega} c \epsilon_0 \lambda_{\omega}^2} \times \left| \int_{-\infty}^{+\infty} \int_0^{+\infty} \bar{d}_{33}^{\text{APE}}(z) E_{\omega}^2(y,z) E_{2\omega}(y,z) dy dz \right|^2,$$

where $d_{\text{QPM}} = 2d_{33}^{\text{LiNbO}_3}/\pi$, $\bar{d}_{33}^{\text{APE}}(z) = d_{33}^{\text{APE}}(z)/d_{33}^{\text{LiNbO}_3}$, and the fields are normalized to carry unity power. The depth dependence of the refractive index profile was determined using a one-dimensional nonlinear diffusion model for the APE process,¹⁰ and we assume a 4-μm-wide top-hat dependence for the lateral refractive index profile. We assume that the two-dimensional modes are separable, $E(y,z) = E_y(y)E_z(z)$, and compute the mode profiles at

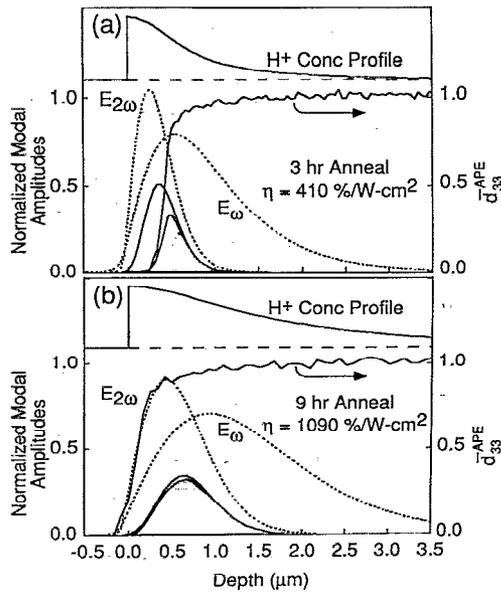


FIG. 3. Fundamental ($\lambda_\omega=850$ nm) and SH ($\lambda_{2\omega}=425$ nm) modes (dotted lines), modal overlap (solid line), measured normalized nonlinear coefficient $\bar{d}_{33}^{\text{APE}}$, the modal overlap with $\bar{d}_{33}^{\text{APE}}$ (shaded region), and the H^+ concentration profile vs depth in APE-LiNbO₃ waveguides annealed for (a) 3 h and (b) 9 h. The abscissa is the same for (a) and (b).

$\lambda_\omega=850$ nm and $\lambda_{2\omega}=425$ nm using the effective index method. The depth overlap integral was evaluated using the depth dependence of the d_{33} coefficient derived by taking the square root of the normalized reflected SH power given in Fig. 2. Shown in Fig. 3 versus depth are the optical modes, the measured normalized nonlinear coefficient $\bar{d}_{33}^{\text{APE}}(z)$, the modal overlap $E_\omega^2(z)E_{2\omega}(z)$; and as the shaded region, the modal overlap with the measured value of the nonlinear coefficient, $\bar{d}_{33}^{\text{APE}}(z)E_\omega^2(z)E_{2\omega}(z)$, for waveguides annealed for 3 and 9 h; the data for a 5 h anneal are not shown for brevity. Also shown is the H^+ concentration profile, proportional to the index profile. At λ_ω the surface index changes are 0.0503, 0.0364, and 0.0230, for 3, 5, and 9 h anneals, respectively. The overlap using the measured value of d_{33} yields η versus annealing of 410, 950, and 1090%/W cm². For comparison, the modal overlap assuming the d_3 coefficient to be independent of depth yields η versus annealing of 2010, 1640, and 1280%/W cm².¹¹ For unannealed waveguides $\eta=0$ since

$d_{33}=0$ for unannealed PE-LiNbO₃. For anneal times longer than 9 h the modal overlap is removed from the region where $d_{33}=0$ and η decreases because of reduced confinement. Figure 3 shows that the variation in η with annealing is not due to a restoration in the d_{33} coefficient of the APE-LiNbO₃ waveguide as much as a variation in the overlap integral due to a spatial redistribution of the modal fields away from the initial PE region.

There are two recent reports of guided-wave QPM SHG using depth-independent ferroelectric domain gratings, with η of 600 (Ref. 7) and 700%/W cm² (Ref. 8) for devices with APE waveguides fabricated similarly to the ones modeled above. These values fall within the range modeled, indicating that the general form of the d_{33} depth profile presented here allows reasonably accurate device modeling. However, waveguide loss, laser longitudinal mode structure, QPM grating and waveguide inhomogeneities, and the value of the d_{33} coefficient of bulk LiNbO₃ (recently measured to be 23.7 pm/V¹² rather than the accepted value of 34.5 pm/V) render the modeling of the absolute η of real devices difficult and questionable. Measurements of η as a function of waveguide processing are ongoing to refine the models for the linear and nonlinear optical properties of APE-LiNbO₃.

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