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## Ultraviolet generation in periodically poled lithium tantalate waveguides

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We demonstrate ultraviolet generation in lithium tantalate channel waveguides for frequency doubling via quasi-phase-matching. The samples, proton exchanged and nanostructured by electric-field assisted surface periodic poling with domains as deep as 40  $\mu\text{m}$ , yield continuous wave light at 365.4 nm with conversion efficiencies larger than 7.5%  $\text{W}^{-1} \text{cm}^{-2}$ . © 2008 American Institute of Physics. [DOI: 10.1063/1.2992201]

Coherent optical sources in the ultraviolet (UV) region of the optical spectrum are needed or recommended in several applications.<sup>1–3</sup> Nowadays, such a demand is only partially satisfied by semiconductor or gas devices, which typically exhibit a pulsed behavior or a low continuous wave power. In the last decades, substantial improvements in nonlinear optical technology and materials have allowed obtaining high frequency conversion efficiencies using quasi-phase-matching (QPM), particularly by electric-field poling in noncentrosymmetric crystals such as lithium niobate and lithium tantalate (LT).<sup>4</sup>

LT is a positive uniaxial crystal with ferroelectric properties; it exhibits excellent linear and nonlinear optical properties, with high threshold to photorefractive damage, extended transparency in the UV [down to 280 nm (Ref. 5)], large electro-optic and quadratic responses [ $d_{33} = 15.1 \text{ pm/V}$  at 852 nm (Ref. 6)]. The technique of proton exchange (PE) permits the realization of good quality channel waveguides because of the low Curie temperature of LT, while frequency conversion in the whole transparency region can be achieved via QPM, thanks to the controlled periodic inversion of ferroelectric domains.<sup>7–11</sup> These features make LT the best dielectric candidate for efficient frequency doublers into the UV (Refs. 12–15) and eventually, pulse shaping of femtosecond Ti:sapphire sources.<sup>16</sup> In this letter we report the demonstration of guided-wave UV generation at 365 nm by frequency doubling a continuous-wave source in LT waveguides realized by PE.

The samples were prepared from standard optical grade wafer substrates of congruent LT with thickness of 500  $\mu\text{m}$ . The doublers were realized by first poling the crystal and later defining the channel waveguides. For surface periodic poling (SPP) we used high voltage pulses applied across the LT thickness in order to achieve overpoling with domains as deep as 40  $\mu\text{m}$ .<sup>17</sup> We spin coated the  $-Z$  face of the substrate with a 1.3- $\mu\text{m}$ -thick photoresist film (Shipley S1813) and defined a 2.0  $\mu\text{m}$  periodic pattern by standard photolithography. After development, the pattern was soft baked overnight at 90 °C and hard baked at 130 °C for 3 h. The temperature was raised gradually in order to avoid problems due to pyroelectric effects, the formation of poling dots and the insurgenence of mechanical stress in the bulk. The baked photolithographic mask was the insulating layer during the high voltage poling for producing domain inversion. The sample was placed between gel electrolyte layers to ensure an ad-

equate electric contact. The high voltage was supplied by an amplified (Trek 662) waveform generator (Agilent 3220A). In order to exceed the LT coercive field and obtain a controlled periodic inversion, we applied single 1.3 kV pulses over a 10 kV bias for appropriate time intervals, the latter adjusted to fulfil the poling condition  $Q < 1.5AP_s$  with  $Q$  the flowing charge,  $A$  the poled area, and  $P_s$  the spontaneous polarization of the ferroelectric crystal. The waveforms of both the poling voltage and the displacement current were monitored with a digital oscilloscope. Using this approach, the inverted domains enucleated from the  $-Z$  facet in the region under the electrodes and extended towards the  $+Z$  facet. By halting the process before domain merging at the sample bottom, we obtained domains as deep as 40  $\mu\text{m}$ , as visible in the microphotographs in Fig. 1 subsequent to etching with  $\text{H}_2\text{O}$ -diluted hydrofluoric acid (50%). The mark-to-space ratio of the insulating mask was varied in order to obtain a QPM duty cycle as close as possible to the ideal value of 50:50 for efficient second harmonic generation (SHG).

We prepared channel waveguides of widths ranging from 2 to 7  $\mu\text{m}$  using PE in a “sealed ampoule.” We employed a 400 Å radio-frequency sputtered  $\text{SiO}_2$  mask and a melted mixture of benzoic acid and 3.6% lithium benzoate; this ratio was chosen in order to preserve the nonlinear optical properties and domain orientation in the crystal and yielded soft PE waveguides in the  $\alpha$ -phase.<sup>18</sup> We performed PE at about 300 °C for various times, up to 6 days. The silica mask was then removed and the planar waveguides formed on the  $+Z$  face were characterized at 632.8 nm by distributed (prism) coupling. The waveguides exchanged for 6 days supported a single TM mode at the pump wavelength with an effective index ( $N_{\text{eff}}$ ) of 2.1826 (substrate index 2.1816) and several modes at the second harmonic. An example of a 3  $\mu\text{m}$  wide

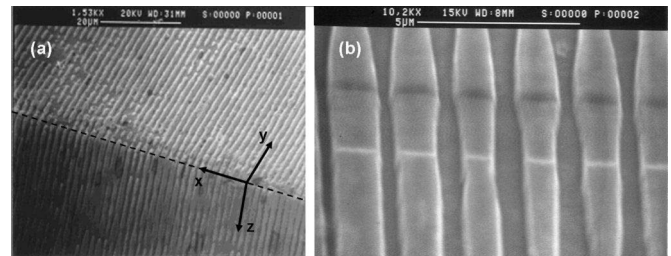


FIG. 1. (a) Photograph of the periodic domain pattern with 2  $\mu\text{m}$  periodicity as revealed by chemical etching in hydrofluoric acid; the dashed line indicates the edge of the sample with the  $-Z$  facet above it. (b) Detail of a PE channel waveguide (white trace) on the etched sample.

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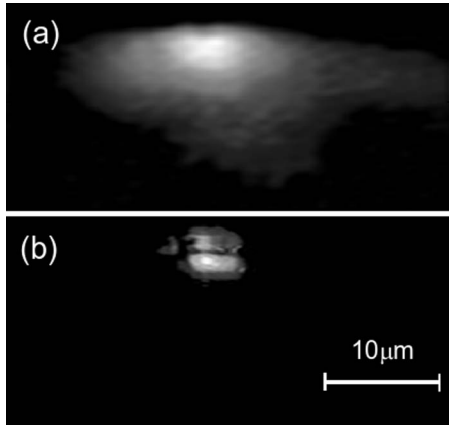


FIG. 2. (a) FF and (b) SH intensities profiles at the output of a  $7 \mu\text{m}$  wide LT channel. The stray light visible above the  $\text{TM}_{00}$  mode profile in (b) is due to diffraction from the edge of the channel.

channel on the  $-Z$  facet of the sample is shown in Fig. 1(b) after chemical etching to reveal the poled domain structure.

For the nonlinear characterization, an Ar-ion pumped Ti-sapphire laser, tunable from 700 to 980 nm and with a 40 GHz linewidth, was end fire coupled by a microscope objective into the channel waveguides, keeping the sample at a constant temperature of  $250 \pm 0.1 \text{ }^\circ\text{C}$  in order to reduce or eliminate the chances of photorefractive damage.

A filter at the waveguide output helped eliminating the fundamental frequency (FF) pump in the near infrared and another filter suppressed the remaining argon ion light. The generated second-harmonic (SH) power in the UV was measured with a calibrated (silicon) photodiode equipped with a chopper and a lock-in amplifier to reduce the noise level, while both FF and SH modal profile were imaged by a charge coupled device camera, as shown in Fig. 2 for a  $7 \mu\text{m}$  wide channel.

Figure 3(a) graphs the generated SH power versus FF wavelength in  $7 \mu\text{m}$  wide waveguides: for a QPM period of  $2.0 \mu\text{m}$ . The SHG resonance occurred between fundamental-order TM modes for an input wavelength close to 730.7 nm. Although the PE channel was multimode in the UV and the

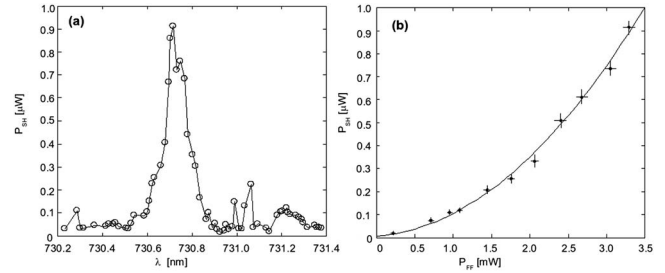


FIG. 3. (a) SH generated power versus wavelength of the FF injected in a  $7 \mu\text{m}$  wide channel. The resonance is at  $\lambda_{\text{FF}} \approx 730.7 \text{ nm}$ ; (b) parabolic trend of the generated UV light versus FF input power at the QPM resonant wavelength.

$N_{\text{eff}}$  of some higher-order SH modes were rather close to the  $\text{TM}_{00}$ , the latter dominated the process owing to a significantly larger overlap integral. Features in Fig. 3(a) are due to slight inhomogeneities in the QPM sample. The generated corresponding mode in the UV exhibited the expected quadratic growth versus FF power, as displayed in Fig. 3(b) for a 2 cm long channel with a 1.0 cm QPM (SPP) region in the middle, equidistant from input and output facets. The values graphed in Fig. 3(b) are purged of the external (Fresnel reflections) losses. From the SHG resonance in Fig. 3(a), with a FWHM  $\delta\lambda = 0.152 \text{ nm}$ , we could estimate the effective length of the surface-poled region to exceed 9.5 mm, the latter value indicating that almost the whole poled region (10 mm) contributed to the parametric process, i.e., proving that the SPP process produced a uniform QPM grating.

By fitting the experimental data in Fig. 3(b) we got normalized peak conversion efficiency  $\eta = P_{\text{SH}}/P_{\text{FF}}^2 L_{\text{eff}}^2$  close to  $7.5\% \text{ W}^{-1} \text{ cm}^{-2}$ . In order to estimate the actual quadratic nonlinearity of the LT waveguides, we compared these experimental data with the conversion efficiency for first-order quasi-phase-matched SHG,

$$\eta = \frac{4}{\pi^2} d_{33}^2 \frac{8\pi^2 \eta_0 f_{\text{SHG}}}{\lambda_{\text{FF}}^2 N_{\text{FF}}^2 N_{\text{SH}}}, \quad (1)$$

where  $\eta_0$  is the vacuum impedance,  $N_{\text{FF}}$  and  $N_{\text{SH}}$  are the effective refractive indices at FF and SH wavelengths, respectively, and  $f_{\text{SHG}}$  is the SHG overlap integral

$$f_{\text{SHG}} \equiv \frac{\left[ \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} e_{\text{SH}}^*(x,y) e_{\text{FF}}^2(x,y) dx dy \right]^2}{\left[ \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} |e_{\text{FF}}(x,y)|^2 dx dy \right]^2 \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} |e_{\text{SH}}(x,y)|^2 dx dy}, \quad (2)$$

with  $e_{\text{FF}}(x,y)$  and  $e_{\text{SH}}(x,y)$  the dimensionless transverse (modal) profiles at the two wavelengths. From the output profiles of Fig. 2 we calculated an effective area  $1/f_{\text{SHG}} = 130 \mu\text{m}^2$  and using Eq. (1), we obtained an effective nonlinearity  $d_{\text{eff}} = 4.4 \text{ pm/V}$  and a material nonlinearity  $d_{33} = 6.9 \text{ pm/V}$ . In spite of the neglected absorption at SH and FF, the latter value is consistent with what ( $d_{\text{eff}} = 4.7 \text{ pm/V}$ ) was reported by Meyn *et al.* for bulk poled crystals.<sup>15</sup>

In conclusion, we have realized and tested (soft) PE and surface periodically poled LT guided-wave parametric generators. These LT waveguides, featuring short period ( $2 \mu\text{m}$ ) QPM with  $40 \mu\text{m}$  deep domains, are the first coherent integrated optics UV sources at  $365.4 \text{ nm}$ , with remarkable conversion efficiencies exceeding  $7.5\% \text{ W}^{-1} \text{ cm}^{-2}$ . We are currently improving the fabrication parameters in order to optimize the channel profile, improve the input coupling efficiency and perform experiments with a picosecond source.

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