Continuous-wave quasi-phase-matched generation of 60 mW at 465 nm by single-pass frequency doubling of a laser diode in backswitch-poled lithium niobate

Robert G. Batchko, Martin M. Fejer, and Robert L. Byer
Ginzton Laboratory, Stanford University, Stanford, California 94305-4085

Dirk Woll and Richard Wallenstein
Fachbereich Physik, Universität Kaiserslautern, Erwin-Schroedinger-Strasse 46, D-67663 Kaiserslautern, Germany

Vladimir Y. Shur
Institute of Physics and Applied Mathematics, Ural State University, Ekaterinburg 620083, Russia

Levent Erman
Coherent Laser Group, 5100 Patrick Henry Drive, Santa Clara, California 95054

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We report continuous-wave single-pass second-harmonic generation (SHG) in 4-μm-period 0.5-mm-thick backswitch-poled lithium niobate. Pump sources at 920–930 nm include both Ti:sapphire and diode-oscillator–amplifier lasers. SHG of a Ti:sapphire laser at 6.1%/W efficiency, producing 61 mW of power at 460 nm, is demonstrated in 50-mm-long periodically poled lithium niobate samples with a nonlinear coefficient $d_{33} \approx 9 \text{ pm/V}$, and 60 mW at 465 nm and 2.8%/W efficiency is obtained by SHG of a laser-diode source.

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Quasi-phase-matching (QPM) nonlinear optical processes involving visible and ultraviolet radiation typically require short-period domain gratings. In LiNbO$_3$, domain periods of approximately 6 μm or shorter are useful for first-order QPM second-harmonic generation (SHG) of green, blue, and ultraviolet wavelengths. A conventional electric-field-poling technique for the fabrication of periodically poled LiNbO$_3$ (PPLN) with periods as short as 6.5 μm is well established and has been applied to other ferroelectric materials, including LiTaO$_3$ (Ref. 2) and KTiOPO$_4$ (KTP; Ref. 3). Because of the large effective nonlinear optical coefficient of PPLN, $d_{33} \approx 18 \text{ pm/V}$, it continues to attract interest as a QPM material for the generation of blue and ultraviolet wavelengths. Previous demonstrations of blue-light SHG in PPLN included 5 mW of power from a 102%/W-efficient waveguide pumped at 967 nm by a tunable diode laser and 450 mW at 40% efficiency pumped by a pulsed 946-nm Nd:YAG laser. The PPLN described in Ref. 6 was fabricated by use of the conventional poling technique and demonstrated a $d_{33}$ of 10 pm/V in 15-mm-long samples. However, utilization of the conventional technique with longer devices and QPM periods for the generation of shorter wavelengths in PPLN has been limited by a reduced $d_{33}$ owing to uncontrolled spreading and merging of domains in the bulk material.

Recently we introduced an improved electric-field-poling technique that incorporates spontaneous back-switching as the means for achieving increased domain-pattern resolution in 0.5-mm-thick LiNbO$_3$ substrates. In this Letter we report first-order single-pass cw SHG at ~460 nm from 4-μm-period 0.5-mm-thick 50-mm-long backswitched PPLN. Pump sources include Ti:sapphire and diode-oscillator–amplifier lasers.

Spontaneous flip-back, or backswitching, of inverted domains is well known in the field of ferroelectric materials. For many ferroelectrics, if the applied voltage is abruptly removed during poling, the poled domains tend to switch back to their original orientation. As shown in Fig. 1, this behavior is also demonstrated in the fabrication of domain-patterned ferroelectrics such as PPLN and periodically poled LiTaO$_3$. In these cases in which the voltage was abruptly removed once the duty cycle of the domains reached approximately 50%, backswitched domains nucleated along the electrode edges. The high density and submicrometer spacing of these backswitched nuclei led us to investigate the utilization of backswitching as a means for achieving high-fidelity ferroelectric-domain patterning.

In backswitch poling the sample is photolithographically patterned with stripe electrodes overcoated with a dielectric insulator, similar to the conventional poling method. An external electric field (i.e., the applied voltage divided by the sample thickness) is then applied to the sample by means of a typical poling apparatus containing a liquid electrolyte for electrical contact with the patterned electrodes and the unpatterned wafer surface, a waveform generator, and...
During this forward-growth stage, the domains are most sensitive to the average field in the crystal. The optimum field for which the domain-wall velocity is identical to the conventional waveform from the start through to the forward-growth stage. Unlike conventional poling, however, the forward-growth stage is sustained until the domains have fully spread beneath the insulator and merged in the bulk, approaching duty cycles of ~100%. On completion of forward growth, the external field is lowered [see Fig. 2(b); time, ~210 ms]. During this backswitching stage, the external field is decreased before the internal field10 (i.e., the field attributed to the orientation of defect dipoles) has relaxed. Depending on the screening rates of the defects, the value of the internal field can be sufficient to cause the forward-switched domains to backswitch. Increasing the external field back up to a value higher than the remaining internal field terminates backswitching [see Fig. 2(b); time, ~250 ms]. The ability to initiate, maintain, and terminate backswitching selectively, together with the high nucleation density, allows for the formation of domain patterns with small feature sizes and high uniformity through large volumes of material.

To demonstrate backswitch poling we patterned by lift-off three 76.2-mm-diameter 0.5-mm-thick congruent-composition z-cut LiNbO₃ wafers with 4-μm-period 0.8-μm-wide 75-mm-thick sputtered NiCr stripe electrodes on the +z face. A 750-μm-wide gap separated consecutive 1-cm-wide NiCr grating sections. A 1-μm-thick photoresist layer (AZ 4620) overcoated the NiCr. Patterning the photoresist with an array of 250-μm-wide stripe openings allowed electrical contact with the NiCr. A stripe opening in the photoresist (oriented orthogonally to the NiCr stripes) ran along the middle of each grating section. The photoresist was postbaked for 1 h on a hotplate at 150 °C. The patterned wafers were mounted in a conventional poling fixture incorporating a liquid electrolyte of lithium chloride in saturated solution. The wafers were forward poled for 145 ms and then backswitched for 39.2 ms by use of a synthesized function generator (Stanford Research Systems Model DS345) and a high-voltage amplifier (Trek Model 20/20). The current and the delivered charge for the backswitch-poling process are shown in Fig. 2(c).

Uncoated 50-mm-length samples were obtained from the backswitched wafers and characterized by cw single-pass SHG of blue light by use of both Ti:sapphire and laser-diode sources. Both lasers were tuned to wavelengths in the range 920−930 nm and

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**Fig. 1.** Backswitched domain nucleation along the electrode edges of periodically poled ferroelectrics: (a) top (+z) surface photograph of 10-μm-period PPLN; (b) +z surface photograph of 26-μm-period periodically poled LiTaO₃; (c) illustration showing the direction of spontaneous polarization (arrows), the electrodes on the +z surface (shaded regions), and the backswitched nuclei (cones).

**Fig. 2.** Poling waveforms, showing (a) programmed external field versus time for the conventional method, (b) programmed external field (solid line) and monitored external field (dotted curve) versus time for the backswitch method, and (c) current (solid line) and delivered charge (dotted curve) versus time for the backswitch method. In (b) the discrepancy between the programmed and the monitored fields is due to voltage regulation by the ~20-mA-current-limited high-voltage amplifier.
focused to 44-μm waist radii in the center of the PPLN. We chose a high phase-matching temperature range of 225–300 °C to avoid photorefractive effects and reduce blue-light-induced infrared absorption in the PPLN. Figure 3 shows plots of SHG powers as functions of fundamental power. Both the SHG and the fundamental powers are given internally to the PPLN. For the 920-nm single-frequency Ti:sapphire pump, 60 mW of blue SHG was produced at 1 W of pump power, giving a normalized efficiency of 6.1%/W. The Ti:sapphire beam had an $M^2$ of <1.2, and the Ti:sapphire SHG beam had an $M^2$ of ≈2.6 owing to thermal lensing in the PPLN crystal. The effective nonlinear coefficient, $d_{\text{eff}}$, is 9 pm/V (approximately 50% of ideal) for the 50-mm-length samples, was limited by variation in the positions of the domain walls. A SHG temperature-tuning curve for a 50-mm-length sample with a 1-W Ti:sapphire pump is shown in the inset of Fig. 3.

Laser-diode-pumped SHG was performed with an antireflection-coated InGaAs single-stripe diode master oscillator (SDL prototype) in a Littman external-cavity configuration. The oscillator was single frequency and tunable from 874 to 936 nm, with a maximum output of 20 mW at 930 nm. The master oscillator was tuned to 930 nm and followed by a first pair of optical isolators, providing more than 60 dB of total isolation. Following the first isolator pair, the laser-diode beam was passed through a laser-diode tapered amplifier (SDL prototype) and followed by a second 60-dB isolation stage. The pump power after the second isolation stage was more than 2 W, with $M^2$ values of 2.2 and 1.5 along the PPLN crystal na and nb axes, respectively. With 1.5 W of laser-diode power, 60 mW of SHG at 465 nm was produced, giving a normalized conversion efficiency of 2.8%/W. As a result of the amplified diode's high $M^2$ value and large percentage of amplified spontaneous emission, an improved efficiency of 3.4%/W was obtained by expansion and spatial filtering of the diode beam before it was focused into the PPLN.

In summary, backswitch poling has been investigated as an effective solution for achieving wafer-scale 4-μm-period domain patterning of 0.5-mm-thick LiNbO3. With 50-mm-length period samples, first-order single-pass cw SHG at 460–465 nm produced 61 mW of power at 6.1%/W efficiency from a Ti:sapphire laser source and 60 mW at 2.8%/W efficiency for a laser-diode source. In the future we plan to improve backswitch poling to obtain a higher effective nonlinear coefficient over large sample areas, and we plan to apply the technique reported here to other ferroelectric materials, such as LiTaO3, with lower optical absorption.

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