Periodically poled potassium niobate for second-harmonic generation at 463 nm

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We report on the fabrication and characterization of quasi-phase-matched potassium niobate crystals for second-harmonic generation. Periodic 30-μm-pitch antiparallel ferroelectric domains are fabricated by means of poling in an electrical field. Both birefringence and periodic phase shift of the generated second harmonic contribute to phase matching when the \( d_{31} \) nonlinear optical tensor element is used. 3.8 mW of second-harmonic radiation at 463 nm is generated by frequency doubling of the output of master-oscillator power-amplifier diode laser in a 5-mm-long crystal. The measured effective nonlinear coefficient is 3.7 pm/V. The measured spectral acceptance bandwidth of 0.25 nm corresponds to the theoretical value. © 1999 Optical Society of America

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The development of periodically oriented crystals for quasi-phase-matched (QPM) nonlinear optical frequency conversion has been very successful in recent years. Periodical poling of ferroelectric crystals is now a mature technology for fabricating QPM devices. Different materials have been investigated and compared. Periodically poled lithium niobate is widely used at present, owing to its high effective nonlinear coefficient of \( d_{eff} = 17 \) pm/V, an available crystal length as great as 60 mm, and excellent mechanical stability. Periodically poled lithium tantalate (PPLT) is attractive because of its wide transparency range, down to 280 nm. Both PPLN and PPLT are photorefractive and therefore have to be operated at elevated temperatures. Periodically poled potassium titanate phosphate and its isomorph rubidium titanyl arsenate can both be operated at room temperature, and poling of crystals as thick as 3 mm for high-power optical parametric oscillator applications has been shown. Magnesium oxide–doped lithium niobate does not exhibit photorefractive effects, either; however, this material requires the use of an advanced poling technique. Recently periodically poled barium titanate was presented as a new QPM material. None of these materials can be considered ideal for all applications. Therefore the search for additional QPM materials continues.

In this Letter we report what is to the best of our knowledge the first fabrication of periodically poled potassium niobate (PPKN). Periodically poled potassium niobate (PPKN) has drawn attention because of its large birefringence and nonlinearity, which allow efficient second-harmonic and sum-frequency generation to the blue spectral range. One can obtain noncritical birefringent phase matching of second-harmonic generation (SHG) in the range 840–1070 nm by tuning the crystal temperature from −40 to 210 °C. Inconvenient crystal temperatures either close to a phase transition at 225 °C or below the dew point are avoided by use of quasi phase matching, as demonstrated here. Moreover, noncritical quasi phase matching, is possible for the entire transparency range (0.4–5 μm), thus making PPKN an interesting material for mid-infrared OPO's.

Potassium niobate is an orthorhombic crystal with the point group \( mm2 \). Different axes have been reported in the literature for this crystal class: An overview is given in Ref. 14. The lattice constants are \( a_0 = 0.569 \) nm, \( b_0 = 0.397 \) nm, and \( c_0 = 0.573 \) nm. The principal tensor axes \( X, Y, \) and \( Z \) coincide with \( a, b, \) and \( c \), respectively. Potassium niobate crystals are grown by the top-seeded-solution growth method. The crystal goes through two phase transitions, at 435 °C (cubic to tetragonal 4\( mm \)) and at 225 °C (tetragonal to orthorhombic \( mm2 \)), when it is cooled from growth temperature at 1040 °C to room temperature. The orthorhombic phase is both ferroelectric and ferroelastic. Crystals usually exhibit domain walls in 60°, 90°, 120°, and 180° orientations. Poling at elevated temperature is required if one wishes to obtain a single-domain crystal.

The QPM periods required for SHG of near-infrared lasers have been calculated with the Sellmeier equations of Ref. 19. Five different cases of QPM that correspond to five nonvanishing nonlinear tensor elements are possible. The QPM grating periods as a function of the second-harmonic wavelength are plotted in Fig. 1. Table 1 summarizes the nonlinear tensor elements \( d_{ij} \) (as reported in Ref. 14), the magnitude of the effective nonlinear coefficients \( d_{eff} = 2\pi d_{ij} \), the QPM grating period \( \Lambda \), and the fundamental wavelength-acceptance bandwidth \( \Delta \Lambda \) for a 1-cm-long crystal and a laser wavelength of 925.8 nm as used in our experiment. When the \( d_{31} \) tensor element is used, a large QPM period of \( \Lambda = 30 \) μm is sufficient, whereas the QPM period of \( \Lambda = 3.9 \) μm that is required for QPM when the \( d_{33} \) tensor element is used is beyond our present technical capabilities. Interactions using \( d_{24} \) or \( d_{15} \) require even shorter periods, less than \( \Lambda = 2.4 \) μm.

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Table 1. Quasi-Phase-Matching Parameters for SHG at 925.8 nm for Various Polarizations of Fundamental and Second-Harmonic Radiation at Room Temperature

<table>
<thead>
<tr>
<th>Polarization</th>
<th>Temperature (°C)</th>
<th>$d_{ij}$ (pm/V)</th>
<th>$d_{eff}$ (pm/V)</th>
<th>$\Lambda$ (µm)</th>
<th>$\Delta\lambda_1$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$cc \rightarrow c$</td>
<td>20</td>
<td>$d_{33} = 19.5$</td>
<td>12.4</td>
<td>3.94</td>
<td>0.090</td>
</tr>
<tr>
<td>$aa \rightarrow c$</td>
<td>20</td>
<td>$d_{33} = 11.3$</td>
<td>7.2</td>
<td>30.03</td>
<td>0.125</td>
</tr>
<tr>
<td>$ab \rightarrow c$</td>
<td>20</td>
<td>$d_{33} = 12.8$</td>
<td>8.2</td>
<td>18.87</td>
<td>0.153</td>
</tr>
<tr>
<td>$bc \rightarrow b$</td>
<td>20</td>
<td>$d_{32} = 12.8$</td>
<td>8.2</td>
<td>1.87</td>
<td>0.050</td>
</tr>
<tr>
<td>$bb \rightarrow c$</td>
<td>150</td>
<td>$d_{32} = 12.8$</td>
<td>12.8</td>
<td>0.139</td>
<td></td>
</tr>
</tbody>
</table>

*The parameters for birefringent noncritical phase matching at 150°C are given in the bottom line for comparison. For further details see text.

Nevertheless this type of quasi phase matching can be interesting for applications in the infrared, in which the QPM periods are generally larger. For example, a 1064-nm-pumped OPO using $d_{15}$ would require QPM periods of the order of $\Lambda = 17.8$ µm to $\Lambda = 20.2$ µm for idler wavelengths from 2.1 to 5 µm.

For our investigations several crystal plates of 10 mm × 9 mm × 1 mm (the lengths along the $a$, $b$, and $c$ axes, respectively) were cut from a single potassium niobate crystal and polished on the $c$ and $c$ faces. The crystal-growth procedure was similar to the one described in Ref. 17. Periodically oriented antiparallel (180°) domains were fabricated with the electric-field method, similarly to the method developed for PPLN. The crystal’s $c$-face surface was coated with a photoresist layer (Shipley Microposit 1805). We created openings with a width of 12 µm and a period of 30 µm by exposing the resist and developing the sample in Shipley 321 developer. We chose the electrode width to be 25% smaller than the desired domain width to allow spreading of domains under the insulating resist, similarly to the process used for PPLN and PPLT. The grating lines are parallel to the $a$ axis; i.e., the $k$ vector of the grating is parallel to the $b$ axis.

The setup for poling is shown in Ref. 4. The crystal was held in an insulating fixture by silicone rubber o rings. Electrical contact was made by a solution of lithium chloride in water. The poling process was conducted at room temperature. A 1-MΩ series resistor was used for current limitation. The poling current was 1.5 mA for an external voltage of 2000 V. Hence the voltage across the crystal surfaces was 500 V, corresponding to a coercive field of 500 V/mm.

Two rectangular pulses with a duration of 5.0 ms were applied. After both pulses we observed that approximately 50% of the charge flowed back through the crystal at zero external field. This phenomenon is caused by backswitching of domains and is known from both congruent lithium niobate and lithium tantalate. The amount of backswitching varied from sample to sample. A net charge of 6.5 µC was delivered to a poled region of 7-mm diameter. This charge is only 40% of the value calculated from the spontaneous polarization of 0.41 C/m².19,20 The origin of this discrepancy needs to be investigated in more detail. A diode in the circuit allowed us to avoid backflipping; however, the domains were less uniform than the domains that were obtained without a diode in the circuit.

After they were poled, the crystals were etched in hydrofluoric acid (48%) at room temperature for 2 min so that the domains were revealed. The domains were straight and uniform on the $c$+ face and extended through the entire crystal, as indicated by the periodic structure of the $c$− face shown in Fig. 2. A similar result was obtained when only the $c$− face was patterned. In several cases additional 60° domains were observed, mainly with the $b$ axis in plane and outside the periodically poled region. The domains have a very strong tendency to propagate along the $a$ axis. We observed domains growing under the insulating resist layer for several hundred micrometers, which led to scrambled domains when channels of different periods were separated by small insulating regions of 100 µm. When the $k$ vector of the electrode structure was chosen to be parallel to the crystal’s $b$ axis, the domains did not grow in straight lines in either the $b$ or the $c$ direction. The domain structures that have been obtained in this orientation have not been sufficient for optical experiments.

A 5-mm-long piece of PPKN with a 30-µm poling period ($k || b$) was used for SHG of a single-frequency master-oscillator power-amplifier diode laser.21 The master-oscillator power-amplifier emitted 2.3 W of single-frequency 926-nm radiation in an almost-diffraction-limited beam. The laser beam was focused confocally; i.e., the minimum beam spot size was $w_0 = 18$ µm. For efficiency measurements the laser power that was incident upon the crystal and the power of the second-harmonic radiation were measured with a calibrated powermeter (Newport Model 835).
frequency-doubled radiation was separated from the laser light with dielectric filters. Quadratic growth of the second harmonic with the fundamental power was confirmed experimentally. The maximum output power of 3.8 mW at 463 nm was obtained with 1.9-W fundamental power incident upon the crystal. The conversion efficiency was calculated with the assumption of diffraction-limited beam propagation and confocal focusing. Since the crystals were uncoated and wedged, the Fresnel reflections at the surfaces were taken into account for calculating the internal conversion efficiency. For single-longitudinal-mode laser light with dielectric filters. Quadratic growth of the second harmonic (QPM) and the birefringence was compensated for by a combination of periodic phase shift of the second harmonic (QPM) and the birefringence of the crystal. 3.8 mW of blue 463-nm radiation was generated in a single pass of 1.9-W fundamental power from a master-oscillator power-amplifier diode laser at 926 nm. The effective nonlinear coefficient was 3.7 pm/V. No photorefractive or other degradation was observed at room temperature. We plan to study optimization of domain quality, power scaling of blue-light generation, poling of crystals with larger dimensions, and applications in OPO's.

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