5 mm Thick Periodically Poled Rb:KTiOPO₄ for High Power Optical Frequency Conversion

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Abstract: A periodically poled bulk Rb-doped KTiOPO₄ crystal with 5 mm aperture was fabricated at room temperature. The ferroelectric domain structure is shown to be homogeneous across the whole aperture with a dₑeff of 11 pm/V.

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1. Introduction

High-energy mid-infrared sources are required for a number of applications including spectroscopy [1], remote sensing [2], biology and medicine [3], material processing [4], standoff defense systems [5] and others. Recent advances in laser technology made efficient and robust, high-energy and good beam-quality diode-pumped Q-switched mode-locked lasers at 976 nm available. Optical parametric oscillators (OPOs), pumped by such lasers, are capable of providing high-power tunable radiation in spectral regions not covered by commercially available lasers.

The nonlinear materials used as gain medium in such OPOs must meet certain requirements: they must be transparent for the three interacting waves, optically homogeneous, chemically stable, and relatively easy to produce. For high power applications high optical damage thresholds and the availability of large apertures are necessary.

Materials of the KTiOPO₄ (KTP) family are excellent nonlinear gain media which meet all above requirements. They have a wide transparency range covering 0.35-4.5 μm [6] and can be employed in cascaded OPO schemes reaching 6-8 μm [7]. Furthermore, the ferroelectric nature of those materials allows using the highest nonlinear coefficient the material is capable to provide by involving domain engineering and quasi-phase matching (QPM) techniques.

Standard commercially available periodically poled crystals are obtainable only up to a thickness of 1 mm along the polar direction, limited by the reproducibility of the commercial fabrication process. Therefore, only relatively low energy output can be generated in parametric devices employing such crystals. Elliptical pumping beams [8] are not a particularly good option in OPOs. A larger thickness of up to 5 mm along the polar axis in periodically poled MgO:LiNbO₃ (PPMgLN) has been demonstrated recently [9], but from the domain broadening and the duty cycle variation one can appreciate the difficulty of scaling up the aperture in PPMgLN. However, periodically poled KTP (PPKTP) and periodically poled RbTiOAsO₄ (PPRTA) crystals with an aperture thickness of up to 3 mm have been demonstrated previously [10].

There are two main difficulties arising from scaling up the aperture of PPKTP: the high voltage needed to overcome the coercive field of the material, and the ferroelectric domain broadening during the growth of large aspect ratio domain structures. To address those problems the bulk Rb-doped KTP (RKTP) was chosen as the best candidate for large aperture ferroelectric domain engineering. The RKTP crystals were flux-grown with 1.4 mol % Rb added to the flux solution resulting in a final Rb-concentration of less than 1%. This material has similar transmission and nonlinear properties as flux-grown KTP, however orders of magnitude lower ionic conductivity makes it an excellent choice for fabrication of high aspect ratio domain gratings [11]. Furthermore, this material shows improved optical performance compared to that of flux-grown KTP [12].

In this work we present periodic poling of 5 mm thick RKTP at room temperature involving the short electric field pulse poling technique [13]. The ferroelectric domain grating quality in the fabricated crystals was evaluated employing OPO experiments. The results attest to remarkably small domain broadening and duty cycle variation in this material and homogeneous performance over the whole aperture.

2. Experiments

For our experiments we used commercial, single domain, c-cut, flux-grown RKTP crystals with a dopant concentration close to 1%. The crystals were of dimensions 12 mm, 7 mm, 5 mm in the x-,y-,z- directions of the dielectric tensor. The c faces of the samples were patterned with a Λ = 38.86 μm aluminum grating with a duty cycle of 50% using standard photolithography techniques, and the photoresist was left as an insulator. The poling area was 8 x 4 mm² and it was only limited in size by the available photolithography mask and the need of proper crystal
surface isolation due to the high poling fields. The crystal was contacted to the external electric circuit using saturated KCl liquid electrodes.

Due to the relatively long grating period and the large sample aperture we chose 5 ms long pulses of square shape to pole our crystals. The best samples were obtained applying two electric field pulses of magnitude 3.2 kV/mm. We checked the quality of the fabricated QPM grating by chemically etching the periodically poled RKTP (PPRKTP) crystals in a KOH and KNO₃ solution. Fig. 1 shows the domain structure at (a) the patterned face and (b) the non patterned face of a 5 mm thick periodically poled RKTP crystal. The duty cycle of fabricated periodic structure was 51% on the former c⁻ face and 57% on the former c⁺ face. Hence, no significant domain broadening was observed, which can be attributed to a much lower ionic conductivity in RKTP compared to that of flux-grown KTP. Furthermore, no significant material homogeneity variations across the wafer were observed, which guaranteed good crystal-to-crystal poling reproducibility.

The performance and homogeneity of the large aperture PPRKTP crystals was further assessed in an OPO setup. The QPM period was originally designed for close to degeneracy parametric frequency conversion of a 1064 nm pump, hence giving signal and idler wavelengths close to 2.1 μm. The OPO was pumped by a Q-switched Nd:YAG laser emitting 1064 nm pulses of 6.5 ns (FWHM) pulse length at a repetition rate of 20 Hz. The pump power was controlled by an attenuator consisting of a wave-plate and a polarizer. The pump light, polarized parallel to the crystal z-axis, was collimated to a beam waist radius of ~350 μm (1/e² intensity) and launched along the x-axis of the PPRKTP crystal while the crystal temperature was stabilized to 44 °C by a Peltier element. As an input coupler we used a flat dielectric mirror transmitting the pump light while being highly reflective (R>99%) at the signal and idler wavelengths, respectively. A flat dielectric mirror with reflectivity of 50% at signal and idler wavelengths was used as output coupling mirror. The cavity length was 30 mm. The pump radiation and the signal and idler wavelengths were separated by two dielectric mirrors to allow measurements of the pump depletion and the conversion efficiency. Fig. 2 shows the pump depletion and combined signal and idler efficiency for different pump energies.
A maximum OPO output efficiency of 40% was reached in this configuration at maximum pump energy of 6.55 mJ. The difference between pump depletion and conversion efficiency arises from linear losses for the signal and idler at the uncoated crystal surfaces. The OPO threshold was 1.6 mJ, corresponding to a value of $d_{\text{eff}} = 11 \text{ pm/V}$ calculated using the theory given by Brosnan and Byer [14], while the maximum output energy reached was 2.65 mJ.

To evaluate the quality and homogeneity of the fabricated periodic ferroelectric domain structure we have scanned our PPRKTP crystal inside the OPO cavity in two directions perpendicular to the pump beam. The position of the crystal was changed in steps of 500 μm. For each crystal position the OPO output energy was measured at a constant pump energy of 6.25 mJ, which is about 4 times above threshold. Fig. 3 shows the measured OPO output energy distribution across the PPRKTP crystal aperture. The OPO output energy was nearly constant across the whole crystal aperture. It is important to note that the drop of output energy towards the edges of the ferroelectric grating aperture arises because parts of the pump beam are outside of the crystal aperture in this case. However, we estimate that the poled aperture filled the whole 5 x 4 mm². This is a good indication that the fabricated ferroelectric domain grating is homogeneous and of high quality.

3. Conclusions

We have shown that high-quality periodic poled with very little domain broadening can be achieved in 5 mm thick KTP crystals. To the best of our knowledge these are the thickest crystals periodically poled from the KTP family to date. The PPRKTP samples with period $\Lambda = 38.86 \text{ μm}$ were used in a Nd:YAG-pumped OPO setup to demonstrate close to degenerate optical parametric generation at 2.1 μm. The periodic ferroelectric domain grating was of high quality, making the fabricated QPM devices suitable for high power optical frequency conversion.

4. References