5 mm thick periodically poled Rb-doped KTP for high energy optical parametric frequency conversion

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Abstract: A 5 mm thick periodically poled bulk Rb-doped $KTiOPO_4$ (KTP) crystal with a period of 38.86 µm was fabricated by electric field poling. Chemical etching and optical evaluation show a high quality of the periodic ferroelectric domain structure through the whole crystal aperture. The fabricated quasi-phase matching (QPM) device was used in an optical parametric oscillator pumped at 1064 nm with 12 ns pulses at 100 Hz repetition rate to generate 60 mJ parametric radiation with a conversion efficiency of 50%.

OCIS codes: (160.2260) Ferroelectrics; (190.4410) Nonlinear optics, parametric processes.

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1. Introduction

Nanosecond pulses with energies on the order of 100 mJ and tunable in the 1.5 – 3.4 µm region are of interest for applications such as spectroscopy [1], remote sensing [2], biology and medicine [3], and material processing [4]. Optical parametric oscillators (OPOs) pumped by well-established, efficient lasers operating at 1 µm probably offer the simplest and most reliable way to generate such pulses. Quasi-phase matched (QPM) nonlinear materials in high energy OPOs provide definite advantages, such as noncritical phase matching for any chosen signal-idler pair, a large acceptance angle and the possibility to use the highest nonlinear coefficient available in the material. These advantages of QPM can be achieved in the ferroelectric oxide crystals $KTiOPO_4$ (KTP), $LiNbO_3$ (LN) and $LiTaO_3$ (LT) structured by using the electric field poling technique. Due to the noncritical interaction in QPM media the parametric gain scales simply as a square of the crystal length, so that the relative differences in effective nonlinearities in different QPM materials can be easily compensated for by choosing an appropriate crystal length. Therefore, for obtaining high energies from nanosecond OPOs there are only two characteristics which are important, namely, the area of the optical aperture and the homogeneity of the QPM grating in the whole crystal volume.

Fabricating large aperture periodically poled crystals in any of the above-mentioned materials is challenging due to, first, the increasingly high electric field required for polarization-switching; and second, the difficulty of propagating the domain grating through the whole crystal thickness. Furthermore, in thick samples even a small domain broadening can lead to merging of the domains, overall resulting in a spatially inhomogeneous QPM grating. Periodically poled Mg-doped congruent $LiNbO_3$ (PPMgLN) and Mg-doped congruent $LiTaO_3$ (PPMgLT) with thicknesses of up to 5 mm along the polar axis have been recently reported [5,6]. In these materials the decrease of the coercive field with increasing temperature was employed in order to limit the electric-field magnitude required for the ferroelectric domain inversion. However, the periodic domain structures obtained, especially those demonstrated in PPMgLT, are still limited in homogeneity due to duty cycle variations, domain broadening and domain merging throughout the crystal thickness.

KTP is considered to be one of the best materials for frequency conversion in the visible and near-infrared spectral regions because of its good mechanical and thermal properties (the thermal conductivity is about twice as high as in $LiNbO_3$), as well as a high nonlinearity and a high damage threshold. Those properties together with a wide transparency range (0.35-4.5 µm) [7], make it a good material choice for 1-µm-pumped high energy and high average power QPM-OPO applications. High output energy and good beam quality OPOs can also be employed in cascaded schemes reaching further into the mid-infrared [8]. Electric field periodically poled KTP (PPKTP) crystals with a thickness up to 1 mm [9], can be obtained

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with good reproducibility, making this the standard thickness for commercially available PPKTP crystals [10]. Unfortunately, such limited apertures can ensure only relatively low energy output.

Periodically poled structures in KTP and isomorphic $RbTiOAsO_4$ (RTA) crystals with a thickness of up to 3 mm have been reported previously [11,12]. RTA has a substantially lower ionic conductivity than KTP which is beneficial for large aperture poling, however this material is not readily available from commercial vendors. In flux-grown KTP the high ionic conductivity, the inhomogeneous stoichiometry over a single wafer and a poor wafer-to-wafer consistency makes large-aperture poling a complicated process even for 3-mm thick samples.

In this work we demonstrate that the above-mentioned limitations of KTP can be overcome by employing Rb-doped KTP. Consistent periodic poling of this material over a crystal thickness of 5 mm has been achieved. These crystals were employed in a 1064 nm pumped OPO to demonstrate pump-power limited generation of 2.1 µm pulses with an energy of 60 mJ and an average power of 6 W. It was confirmed that the ferroelectric domain grating is uniform throughout the whole crystal aperture which guarantees an efficient OPO output with negligible beam distortion or spectral variations.

2. Experiments and results

A substantially reduced ionic conductivity and an improved material homogeneity due to doping with Rb allow domain inversion with higher precision in large bulk Rb-doped KTP (RKTP) crystals [13]. The RKTP crystals are grown from a flux melt containing 1.4 mol% Rb. The resulting Rb concentration in the crystals is less than 1% [14], suggesting that the linear and nonlinear optical properties of this material are very similar to those of undoped flux-grown KTP. However, the ionic conductivity of RKTP is 2-orders of magnitude lower than that of KTP [15], while its coercive field is 3.7 kV/mm. In addition, substantially lower induced absorption [16] makes RKTP an ideal candidate for high-energy OPO applications.

Periodic poling was done on 5 mm thick commercially supplied single domain c-cut flux-grown RKTP crystals of dimensions 12 x 7 x 5 mm^3 (a, b and c crystallographic axes respectively). A periodic metal-photoresist pattern with a duty cycle of 50% was created on the c- faces of the crystals using standard photolithography. A period of $\Lambda = 38.86$ µm was chosen for frequency conversion of 1064 nm to 2128 nm at room temperature. The crystals were contacted to an external electric circuit using liquid electrodes. Due to the relatively long grating period and large crystal thickness we chose to work in the low poling-field regime [17]. The crystals were periodically poled at room temperature by applying 5 ms long square electric field pulses of magnitude 3.2 kV/mm. The best samples were obtained applying two pulses. The size of the created ferroelectric domain grating in our periodically poled crystals was 8 x 4 x 5 mm^3 along a, b and c crystallographic axes respectively, set by the available photolithographic mask.

The obtained domain structure, revealed on the polar faces by selective chemical etching, for a typical periodically poled RKTP (PPRKTP) crystal is shown in Fig. 1. The duty cycle is close to 50% on the patterned face and approximately 57% on the unpatterned face of the crystal. This indicates that the created ferroelectric domain grating is of high quality and homogeneous throughout the whole crystal aperture. The low domain broadening obtained with these samples is attributed to the approximately two orders of magnitude lower ionic conductivity compared to ordinary flux-grown KTP. Furthermore, RKTP was found to be more homogeneous in material quality than KTP, and we believe this is a reason for the better yield and more consistent poling result obtained.

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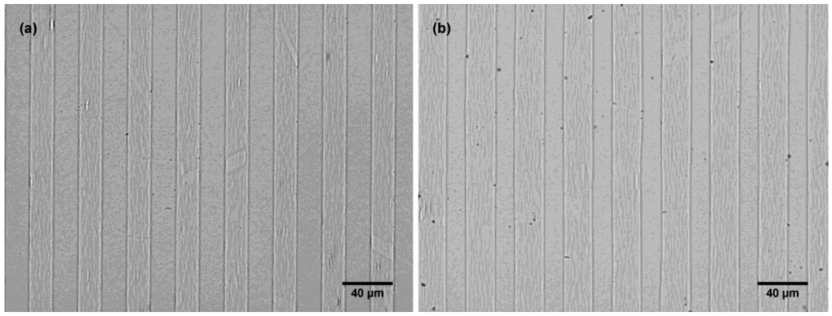

Fig. 1. Ferroelectric domain structure after chemical etching on the former patterned (a) and unpatterned (b) faces of 5 mm thick PPRKTP crystal.

In order to optically evaluate the domain grating quality, we placed one of our uncoated PPRKTP crystals in a linear OPO cavity pumped by Nd:YAG laser emitting 6.5 ns pulses at 20 Hz repetition rate. The domain grating period was designed for close-to-degeneracy parametric frequency conversion of 1064 nm radiation at room temperature, giving signal and idler wavelengths close to 2.1 µm. The fundamental beam polarized parallel to the crystal z axis was collimated to 250 µm radius ($1/e^2$ intensity) spot size and launched along the x axis of the crystal. We used two plane dielectric mirrors. The input coupler was highly reflective (R>99%) at 2.1 µm, while the output coupler had a 50% reflectivity at the same wavelength. Both mirrors transmitted the pump light. The signal and idler radiation was separated from the fundamental wavelength by dielectric mirrors, highly reflective at 2.1 µm and transmitting the pump light. The crystal temperature was stabilized to 44 °C using a thermoelectric element. The OPO cavity length was 30 mm and the OPO pump threshold was reached at pump energy of 1.4 mJ. The OPO was pumped at energy 4 times above threshold while the crystal was translated in the OPO cavity in two directions perpendicular to the pump beam. The position of the crystal was changed in steps of 500 µm and the OPO output was recorded at each point. In total the scan consisted of 80 measurements and the whole aperture area of 4 x 5 mm^2 was recorded. The OPO output energy distribution is more or less constant over the whole aperture, as shown in Fig. 2.

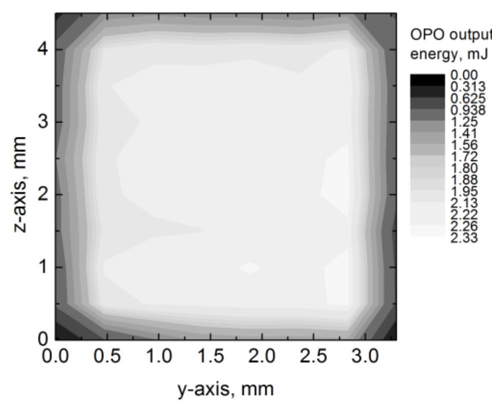

Fig. 2. OPO output energy distribution in *yz* plane of the PPRKTP crystal.

The drop in output energy towards the edges of the scanned area is an artifact due to parts of the pump beam being outside the ferroelectric domain grating. From this measurement we conclude that we have a uniform, high quality ferroelectric domain grating of 4 x 5 mm^2 in the bulk of the crystal.

To evaluate the high-energy performance of our large aperture PPRKTP sample the OPO setup was then pumped by a seeded diode-pumped, single-longitudinal mode Nd:YAG laser

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and amplifier system providing 1064 nm pulses of 12 ns pulse length at 100 Hz repetition rate with a maximum output energy of 130 mJ. To extract more energy the OPO output coupler was changed to one with 30% reflectivity at 2.1 µm. The OPO cavity length was also reduced to 20 mm, while the temperature was set and controlled just above room temperature. The z-polarized pump was expanded and collimated into a beam with a ~2 mm radius and then launched along the crystal x-axis. A wave-plate/polarizer combination was used to adjust the pump power. Figure 3 shows the pump depletion and the combined signal and idler efficiency as a function of pump energy.

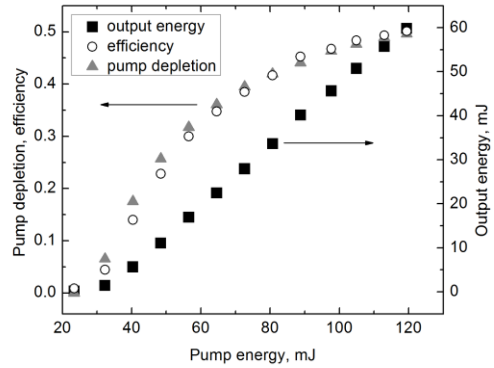

Fig. 3. OPO output energy, conversion efficiency and pump depletion for different pump energies inside the crystal.

A combined OPO signal and idler output energy of 60 mJ was reached at the maximum pump energy in the crystal of 120 mJ, corresponding to incident pump energy of 130 mJ (1.03 J/cm^2). The OPO output energy was limited by the maximum available pump energy. Nanosecond PPKTP OPOs generating at 2.1 µm can be operated reliably at higher fluencies of at least up to 2 J/cm^2 [18]. In our case this fluence would correspond to a peak pump intensity of about 170 MW/cm^2 and hence the pump energy could be safely increased up to 250 mJ. Accordingly we expect that with higher pump energy and appropriate coatings on the PPRKTP OPO output energies exceeding 100 mJ can be achieved.

We studied the temperature tuning properties of the OPO by changing the crystal temperature between 10 – 70 °C. The signal and idler wavelengths were deduced by measuring the wavelength of the generated sum-frequency waves between pump and signal or idler with a spectrum analyzer (ANDO AQ-6315A).

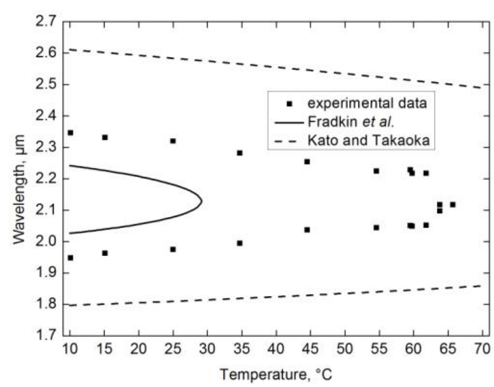

Fig. 4. Temperature tuning of signal and idler waves. The squares represent the experimental data, with error bars as large as the square size. The tuning curves based on Sellmeier equations by Fradkin et al. are shown as solid lines, and by Kato and Takaoka – as dashed lines.

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Figure 4 shows the experimental data together with the theoretical tuning curves based on the Sellmeier equations for KTP derived by Fradkin *et al.* [19] including temperature correction proposed by Emanueli and Arie [20], and temperature-corrected Sellmeier equations by Kato and Takaoka [21]. The discrepancy between measured and theoretical temperature tuning curves may be explained by the fact that neither of the Sellmeier equations were derived at this wavelength span.

To investigate the spectral properties of the OPO at degeneracy the crystal temperature was set to 50 °C and an OPO output spectrum was recorded, as shown in Fig. 5.

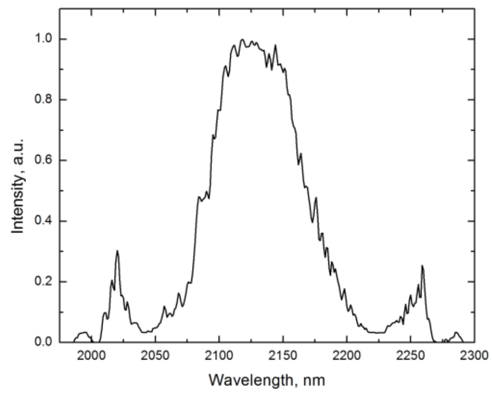

Fig. 5. OPO output spectrum at degeneracy, the crystal temperature set to 50 °C.

The measurement was done using a spectrometer (Jobin Yvon iHR 550) with a PbSe detector. The output is centered at ~2.1 µm with a FWHM of 80 nm. Two side peaks with a distance in frequency of ~16 THz to each other can be seen too. The frequency of 8 THz corresponds to the lowest-frequency infrared-active TO phonon mode in KTP [22]. Therefore we attribute these peaks to four-wave mixing, assisted by intra-cavity stimulated Raman scattering [23], and followed by amplification of the sidebands by the second order parametric process. Narrowing down the intra-cavity gain, e.g. using a volume Bragg grating [18], will effectively remove the gain from this process.

3. Conclusions

In conclusion we have demonstrated a high-energy QPM-OPO based on an uncoated PPRKTP crystal with a QPM grating volume of 8 x 4 x 5 mm^3 (a, b and c axis, respectively) and a period of Λ = 38.86 µm. Chemical etching on the polar faces and optical evaluation showed that the domain grating in PPRKTP is homogeneous and of high quality. The OPO generated 60 mJ of combined signal and idler output energy, corresponding to 6 W of average power with a conversion efficiency of 50% at room temperature. We did not observe any kind of optical damage during the experiments, and the OPO output was only limited by the maximum available pump energy. With higher pump energy and appropriate anti-reflective coatings considerably higher OPO output energy can be expected.

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