Periodically Poled $KTiOAsO_4$ For Mid-Infrared Light Generation

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Abstract: A periodically poled $KTiOAsO_4$ crystal was fabricated at room temperature. The poled crystal shows a d_{eff} of 10.1 pm/V and gives a parametric conversion efficiency of 45%.

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1. Introduction

The mid-infrared spectral region between 2 µm and 5 µm is important for spectroscopy and sensing [1], directed countermeasures and few-cycle femtosecond pulse generation [2], with subsequent efficient production of high-harmonics and attosecond pulses. Optical parametric oscillators are efficient and reliable devices to obtain tunable coherent radiation in this spectral region. Furthermore, they can be used in cascaded schemes reaching into the mid-infrared [3], e.g. 6 µm - 12 µm spectral region, covering the second atmospheric transmission window and also the vibrational absorption bands of biologically-important organic molecules.

Ferroelectric domain gratings in materials such as $KTiOPO_4$ (KTP) isomorphs, MgO-doped $LiNbO_3$, MgO-doped $LiTaO_3$ are appealing as a gain media in 3 - 5 µm parametric devices since they allow quasi-phase-matching of the interacting waves giving access to the largest second order nonlinearity and noncritical interactions. Moreover, these materials can be pumped by well-established and reliable lasers operating around 1 µm.

The arsenate KTP isomorphs, $RbTiOAsO_4$ (RTA) and $KTiOAsO_4$ (KTA), are one of the most promising candidates for mid-infrared parametric devices due to their high optical damage threshold and extended range of transmission [4]. Although, the ionic conductivity of RTA is 3 orders of magnitude lower than that of KTA, which simplifies the poling process, the lack of commercial suppliers of high-quality large single-domain crystals limits the usability of this material. On the other hand, KTA is readily available and widely used in birefringence phase-matched mid-infrared parametric devices [3]. Periodic poling of 0.5 mm thick KTA crystals was previously done by poling at temperature below 170 K [5], where it is known that the KTP isomorphs have a transition from super-ionic conductors to insulators [6]. This method not only adds complexity in instrumentation and processing but also limits the total aperture of the poled device since the coercive field of the material increases substantially at lower temperatures. Moreover, it might introduce internal stresses that can have a negative effect in the crystal's optical performance.

In this work we present periodic poling of KTA at room temperature by taking advantage of relatively short electric field pulses. The poled crystal, which presents a d_{eff} of 10.1 pm/V, is used is used in a Nd:YAG – pumped optical parametric oscillator pumped at 1064 nm to generate radiation at 1538 nm and 3452 nm with a conversion efficiency of 45%.

2. Experiments

In this work we have used a commercial, single domain, c-cut, flux-grown KTA crystal of dimensions 11 mm, 6 mm, 1 mm in x-,y-,z- directions of the dielectric tensor respectively, which correspond to the *a*, *b*, *c* crystallographic axes. The conductivity of the crystals varied from 1.5×10^{-7} S/m to 4×10^{-7} S/m. To fabricate the periodic domain structure, an aluminum grating with a period of 39.5 µm was deposited on the photolithographically patterned c^- face of the crystal and the photoresist was left as an electric insulator. Liquid electrodes were used to contact the crystal to the external electric circuit. The poled area had dimensions of 8 x 3 mm^2. Due to the large ionic conductivity of KTA, the switching current is not suitable to monitor the poling process , therefore we have used the method developed by Karlsson *et al* [7], based on the transverse electro-optic effect together with *in-situ* 9th order second harmonic generation (SHG) [8].

We have previously observed that poling with relatively short pulses prevents domain broadening in KTP [9]. Due to the relatively long grating period we chose a pulse length of 5 ms for poling KTA. The magnitude of the electric field was set between 2.3 kV/mm to 2.6 kV/mm depending on the specific ionic conductivity of each sample, while the number of pulses was adjusted to maximize the SHG output. Fig 1 shows the resulting domain structure at (a) the patterned face and (b) the non patterned face of a sample poled with a magnitude of 2.3 kV/mm. When pumped with 330 mW of a CW Ti:Sapphire lasing at 880 nm this sample gave 11 µW of blue radiation, which corresponds to a first order normalized conversion efficiency of slightly more than 1%/Wcm.

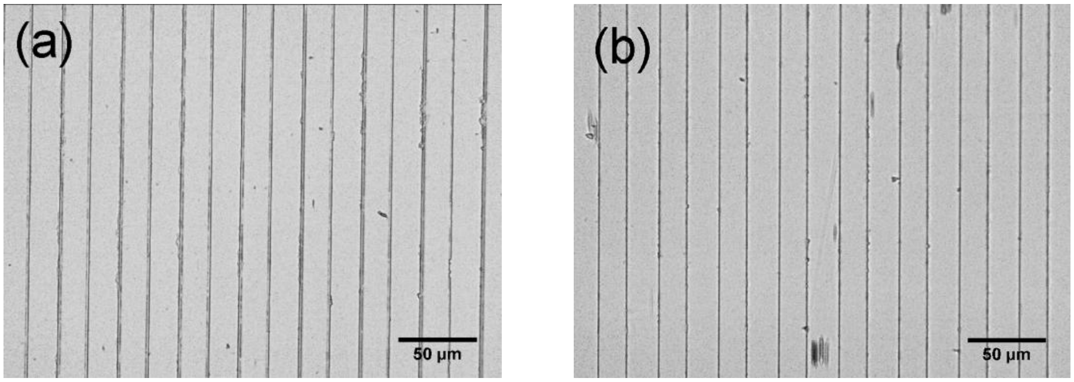

Fig. 1. Photographs of the domain structure revealed by chemical etching on (a) the patterned face and (b) the non patterned face of a PPKTA sample poled with 2.3 kV/mm.

The uncoated periodically poled KTA (PPKTA) crystal was used in a linear optical parametric oscillator (OPO) cavity to show the conversion of 1064 nm pump light to 1.538 µm and 3.452 µm signal and idler output, respectively. It was pumped by a flash lamp pumped Q-switched Nd:YAG laser producing pulses of 6.5 ns (FWHM) pulse length at a repetition rate of 20 Hz and a wavelength of 1064 nm. The pump power was controlled by a waveplate-polarizer arrangement. The z-polarized pump light was launched along the x-axis of the crystal and focused by a 200 mm focal length lens into the crystal to a beam waist of ~300 µm radius ($1/e^2$ intensity). The crystal temperature was stabilized by a Peltier element to 25 °C if not stated otherwise. Flat dielectric mirrors, both transmitting the pump light, were used as input and output couplers with the input coupler being highly reflective (R=99%) for the signal. The effective nonlinear coefficient d_{eff} of the PPKTA crystal was evaluated by measuring the threshold energy for different cavity lengths while using a 90% reflectivity output coupler. In order to avoid parasitic oscillation, the crystal was rotated by about 5° and Fresnel reflections on the crystal surfaces were taken into account during the theoretical calculations. The threshold energies were measured for cavity lengths of 30-50 mm. Theoretical calculations were done in SNLO [10], assuming that threshold was reached when the signal output energy was at 1% of the pump energy. It was found that a d_{eff} of 10.1 pm/V is in good agreement with the experimental results. This value is very close to the expected 10.3 pm/V obtained by $d_{eff}=2d_{33}/\pi$, where d_{33}=16.2 pm/V [11] for a perfect 50% duty-cycle QPM grating.

For pump depletion and efficiency measurements we used a 50% signal-reflective output coupler and a cavity length of 30 mm. Fig 2 shows the pump depletion, the signal efficiency and the combined signal and idler efficiency for different pump energies. The signal wavelength was 1538.2 nm with a bandwidth (FWHM) of 2.5 nm while the maximal signal output was 0.6 mJ (130 MW/cm^2) for a pump energy of 2 mJ (440 MW/cm^2). The depletion was measured by comparing the transmitted pump above and below OPO threshold and scaling appropriately. The signal efficiency was determined from the signal output and the incident pump energy and the combined signal and idler efficiency was calculated from the signal efficiency using the Manley-Rowe relation. Depletion and combined signal and idler efficiency reached 45% for 2 mJ pump energy and were approximately the same over the whole pump energy range. This supports the expected low absorption of PPKTA at the signal wavelength.

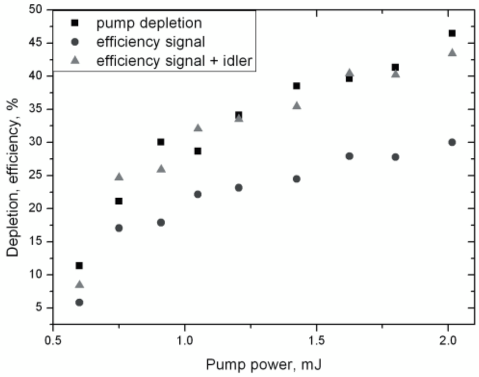

Fig. 2. OPO pump depletion and efficiency as a function of pump power. Squares – pump depletion, circles – signal efficiency, triangles – signal and idler efficiency

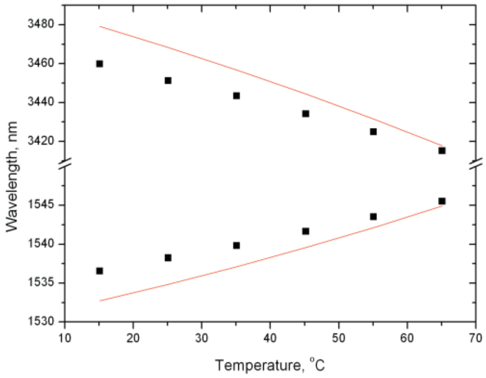

Fig. 3. Temperature tuning of the signal and idler waves. Squares – experimental data, solid lines – calculated theoretical dependence

Finally, we tuned the wavelength of the parametric waves by varying the crystal temperature between 15-65 °C. Fig 3 shows the measured signal wavelength and calculated idler wavelength together with the theoretical tuning curves based on the Sellmeier equations derived by Fradkin-Kashi *et al* [12], and the temperature correction proposed by Emanueli and Arie [13]. The discrepancy between the theoretical and the experimental results might be caused by differences in the used KTA material, for instance, impurities concentration. Further studies of the refractive index temperature dependence for KTA should be done for this spectral region.

3. Conclusions

We have shown high-quality periodic poling of KTA at room temperature. The PPKTA sample has a d_{eff} of 10.1 pm/V, in good agreement with the expected value for a 50% duty-cycle QPM grating. The PPKTA sample was used in a Nd:YAG–pumped OPO to generate parametric radiation at 1538 nm and 3452 nm. The maximum signal energy was 0.6 mJ for a pump energy of 2 mJ, while the pump depletion and combined signal and idler efficiency reached 45%.

4. References

[1] A. K. Y. Ngai, S. T. Persijn, M. M. J. W. Van Herpen, S. M. Cristescu, and F. J. M. Harren, "Photoaccoustic spectroscopy using continuous wave optical parametric oscillators", in Mid-Infrared Coherent Sources and Applications, Eds. M. Ebrahim-Zadeh, I. T. Sorokina, (Springer, 2008)
[2] O. Chalus, Ph. K. Bates, M. Smolarski, and J. Biegert, "Mid-IR short-pulse OPCPA with micro-Joule energy at 100kHz," Opt. Express **17**, 3587-3594 (2009)
[3] M. W. Haakestad, G. Arisholm, E. Lippert, S. Nicolas, G. Rustad, and K. Stenersen, "High-Pulse-Energy 8 μm Laser Source Based on Optical Parametric Amplification in ZnGeP$_2$," SPIE **6998**, 699812-1 – 699812-7, (2008)
[4] G. Hansson, H. Karlsson, S. Wang, and F. Laurell, "Transmission Measurements in KTP and Isomorphic Compounds," Appl. Opt. 39, 5058-5069 (2000)
[5] G. Rosenman, A. Skliar, Y. Findling, P. Urenski, A. Englander, P. A. Thomas and Z. W. Hu, "Periodically poled KTiOAsO$_4$ crystals for optical parametric oscillation," J. Phys. D. **32**, L49-L52 (1999)
[6] G. Rosenman, A. Skliar, D. Eger, M. Oron, and M. Katz, "Low temperature periodic electrical poling of flux-grown KTiOPO$_4$ and isomorphic crystals," Appl. Phys. Lett. **73**, 3650 (1998)
[7] H. Karlsson, F. Laurell, and L. K. Cheng, "Periodic poling of RbTiOPO$_4$ for quasi-phase matched blue light generation," Appl. Phys. Lett. **74**, 1519–1521 (1999)
[8] S. Wang, V. Pasiskevicius and F. Laurell, "High-efficiency frequency converters with periodically-poled Rb-doped KTiOPO$_4$," Opt. Mat. **30**, 594-599 (2007)
[9] C. Canalias, M. Nordlöf, V. Pasiskevicius, F. Laurell, "A KTiOPO$_4$ nonlinear photonic crystal for blue second harmonic generation," Appl. Phys. Lett. **94**, 081121-1 - 081121-3 (2009)
[10] Arlee V. Smith, "How to use SNLO nonlinear optics software to select nonlinear crystals and model their performance" SPIE **4972**, 50-57 (2003)
[11] L. K. Cheng, L. T. Cheng, J. D. Bierlein and F. C. Zumsteg, "Properties of doped and undoped crystals of single domain KTiOAsO$_4$," Appl. Phys. Lett. **62,** 346-348 (1993)
[12] K. Fradkin-Kashi, A. Arie, P. Urenski, and G. Rosenman, "Mid-infrared difference–frequency generation in periodically poled KTiOAsO$_4$ and application to gas sensing," Opt. Lett. **25**, 743-745 (2000)
[13] S. Emanueli and A. Arie, "Temperature-Dependent Dispersion Equations for KTiOPO$_4$ and KTiOAsO$_4$," Appl. Opt. **42**, 6661-6665 (2003)