Periodically poled $KTiOAsO_4$ for highly efficient midinfrared optical parametric devices

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We demonstrate high pattern-fidelity periodic poling of $KTiOAsO_4$ at room temperature. The periodically poled crystal shows a d_{eff} of 10.1 pm/V and is used in an optical parametric oscillator pumped at 1064 nm to generate parametric radiation at 1538 and 3452 nm with a conversion efficiency of 45%. © *2009 American Institute of Physics*. [doi:10.1063/1.3258001]

Optical parametric devices in the mid-infrared spectral region between 2 and 5 μm are 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. Moreover, these devices are employed in cascaded schemes reaching further into the mid-infrared,[3] e.g., 6–12 μm spectral region, covering the second atmospheric transmission window and also the vibrational absorption bands of biologically important organic molecules. All these parametric devices are preferentially pumped by well-established and reliable lasers operating around 1 μm. Unfortunately, highly nonlinear semiconductors cannot be used with these pump sources either due to their high absorption as in the case of GaAs, $ZnGeP_2$ or, due to the limited phase-matching possibilities as in the case of $CdSiP_2$.[4] Ferroelectric oxides, such as $KTiOPO_4$ (KTP) isomorphs, MgO-doped $LiNbO_3$ (MgLN), and MgO-doped $LiTaO_3$ (MgLT), are the preferred choice as a gain media in 3–5 μm parametric devices since they can be electric-field-poled to produce quasi-phasematched (QPM) structures, exploiting the largest second order nonlinearity and noncritical interactions. Owing to the high-peak power requirements that are rather common in midinfrared devices, the possibility to fabricate large-aperture QPM structures becomes an important consideration. Such capability has been demonstrated in MgLN,[5] MgLT,[6] KTP,[7] and $RbTiOAsO_4$ (RTA).[8] Due to their high optical damage threshold and extended range of transmission the arsenate KTP isomorphs, RTA and $KTiOAsO_4$ (KTA), are very promising for midinfrared parametric devices pumped at 1 μm.[9] Although RTA presents ionic conductivity three orders of magnitude lower than that of KTA, which simplifies the poling process, its use is severely limited by the lack of commercial suppliers of high-quality large single-domain crystals. 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,[10] where it is known that the KTP isomorphs have a transition from superionic conductors to insulators.[11] 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 paper, we demonstrate that periodic poling of KTA with high pattern fidelity can be achieved at room temperature by taking advantage of relatively short electric field pulses. The resulting PPKTA samples present a d_{eff} of 10.1 pm/V and give a parametric conversion efficiency of 45%.

For the present studies, we have used commercial, single domain *c*-cut, flux-grown KTA crystals,[12] of dimensions 11, 6, and 1 mm in x, y, and z directions of the dielectric tensor, respectively, which correspond to the *a*, *b*, and *c* crystallographic axes. The conductivity of the crystals varied from 1.5×10^{-7} 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 c^- face of the crystals by standard photolithographic techniques 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 8×3 mm^2. Due to the large ionic conductivity of KTA, the switching current is not suitable to monitor the poling process since it is usually difficult to discriminate it from the ionic current. Therefore we have used the method developed by Karlsson *et al.*[13] based on the transverse electro-optic effect together with *in situ* ninth order second harmonic generation (SHG).[14]

We have previously observed that poling with relatively short pulses prevents domain broadening in KTP.[15] For KTA, due to the relatively long grating period, we chose a pulse length of 5 ms. The magnitude of the electric field was set between 2.3 and 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. Figure 1 shows the resulting domain structure at (a) the patterned face and (b) the nonpatterned 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 focused to a beam waist of 23 μm radius ($1/e^2$ intensity) by a 50 mm focal length lens, this sample gave 11 μW of blue radiation (ninth order SHG). This corresponds to a first order normalized conversion efficiency of slightly more than 1%/W/cm and to an effective nonlinear coefficient d_{eff} of 9.8 pm/V. The duty cycle of this sample varies from 54% on the patterned face to 56% on the nonpatterned face. This indicates that KTA presents a domain dynamics dependence on the electric field magnitude similar

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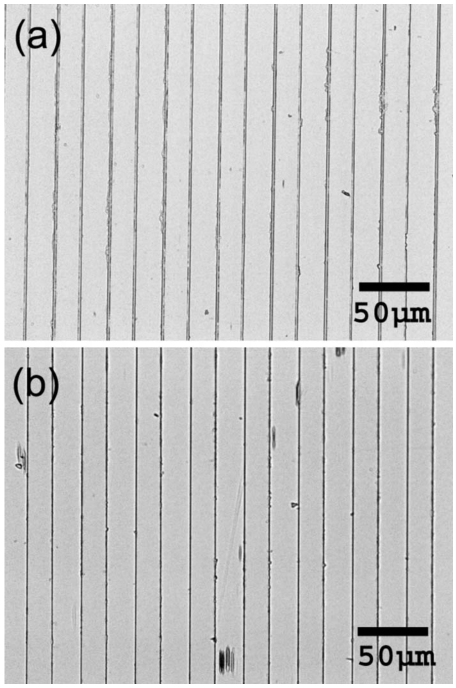

FIG. 1. Photographs of the domain structure revealed by chemical etching on (a) the patterned face and (b) the nonpatterned face of a PPKTA sample poled with 2.3 kV/mm.

to that of KTP (Ref. 16) and therefore a slightly lower field magnitude would have resulted in a 50% duty cycle all the way through the crystal. Similar results were obtained for all the samples, and therefore from now on we will refer to the results obtained with the sample shown in Fig. 1.

We used the uncoated periodically poled KTA (PPKTA) crystal 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. The pump source was a flash lamp pumped Q-switched neodymium doped yttrium aluminum garnet (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. We evaluated the effective nonlinear coefficient d_{eff} of the PPKTA crystal 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 where taken into account in the calculations below. Figure 2 shows the measured threshold energies for cavity lengths of 30–50 mm together with calculated thresholds for different values of d_{eff}. The calculations were done in SNLO (Ref. 17) 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 slightly larger than the one obtained with the ninth

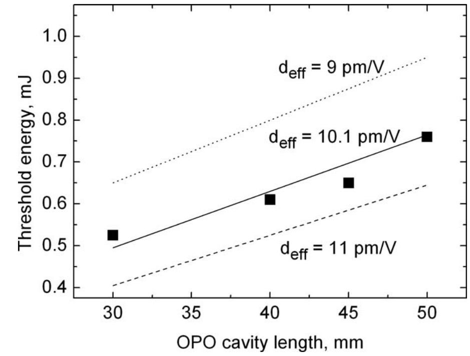

FIG. 2. OPO threshold as a function of cavity length. Squares—experimental data, dotted, solid, and dashed lines—theoretical OPO threshold corresponding to a d_{eff}=9 pm/V, d_{eff}=10.1 pm/V, and d_{eff}=11 pm/V, respectively.

order SHG. This is expected since the QPM ninth order is more sensitive to small duty-cycle variations. It should be noted that 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 (Ref. 18) for a perfect 50% duty-cycle QPM grating.

To obtain a highly efficient OPO, we used a 50% signal-reflective output coupler and a cavity length of 30 mm. Figure 3 shows the pump depletion, the signal efficiency and the combined signal and idler efficiency for different pump energies. Two dielectric mirrors, transmitting 1064 nm and reflecting 1.5 μm, were used to separate the signal and the pump light. 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 although the combined signal and idler efficiency was calculated from the signal efficiency after 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.

Finally, we tuned the wavelength of the parametric waves by varying the crystal temperature between 15–65 °C. In this measurement, the beam was propagating parallel to the grating vector, and the wavelength was mea-

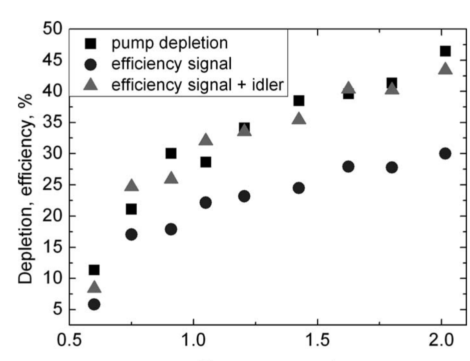

FIG. 3. OPO pump depletion and efficiency as a function of pump power. Squares—pump depletion, circles—signal efficiency, triangles—signal and idler efficiency.

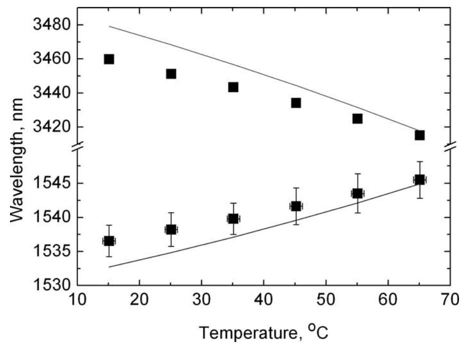

FIG. 4. Temperature tuning of the signal and idler waves. Squares—experimental data, solid lines—calculated theoretical dependence. The vertical error bars show FWHM of the measured signal spectrum and the horizontal ones show the uncertainty in temperature.

sured with an Ando spectrometer (AQ-6315A). Figure 4 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.*[19] and the temperature correction proposed by Emanueli and Arie.[20] There is a discrepancy between the calculated and the experimental tuning curves, which might be caused by differences in the used KTA material, for instance, impurities concentration. At the same time the measurement uncertainty is increased because the parametric gain is broadening as the parametric wavelengths approach the zero group velocity dispersion point. Further studies of the refractive index temperature dependence should be done for this spectral region.

In summary, we demonstrated high-quality periodic poling of KTA at room temperature. The PPKTA sample has a d_{eff} of 10.1 p.m./V. The sample was used in a Nd:YAG-pumped OPO to generate a signal wavelength of 1538 nm and an idler wavelength of 3452 nm, with an efficiency of 45%. The maximum signal energy was 0.6 mJ for a pump energy of 2 mJ while the pump depletion reached 45%.

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