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Green and ultraviolet quasi-phase-matched second harmonic generation in bulk periodically-poled $KTiOPO_4$

A. Arie [a], G. Rosenman [a], V. Mahal [a], A. Skliar [a], M. Oron [b], M. Katz [b], D. Eger [b]

[a] *Department of Electrical Engineering – Physical Electronics, Faculty of Engineering, Tel Aviv University, 69978 Tel Aviv, Israel*
[b] *NRC, 81800 Soreq, Israel*

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Abstract

A periodically-poled flux-grown KTP crystal was used for first order quasi-phase-matched (QPM) second harmonic generation of a Nd:YAG laser. The normalized conversion efficiency of the 1 cm long, 0.5 mm thick sample was 1.28% W^{-1}. A lower limit on the nonlinear coefficient of KTP, $d_{33} \geq 14.9 \pm 1.5$ pm/V was derived from these measurements. The temperature and wavelength tuning coefficients near 1064 nm were determined. The same device was also used for third order QPM frequency doubling of 784 nm light, and the normalized conversion efficiency in this case was 0.12% W^{-1}.

Quasi-phase-matched second harmonic generation is achieved by periodic reversal of the sign of the nonlinear coefficient to compensate for the phase velocity mismatch between the fundamental and second harmonic waves [1]. The main advantages of QPM nonlinear processes are higher efficiency obtained by choosing the larger, diagonal elements of the $\chi^{(2)}$ tensor which are not accessible to birefringent phase-matching and the possibility to noncritically phase-match at a chosen temperature any interaction within the transparency range of the nonlinear material. Until recently, QPM was mainly used with waveguide devices. However, the relatively small guiding area in waveguides is a drawback for high power nonlinear interactions. This problem can be solved by using bulk materials. Unfortunately, in most of the early methods, the poling occurred only in a small area close to the surface of the device. Recently, poling over 0.5 mm thick $LiNbO_3$ was achieved by electric field poling, which enabled the demonstration of bulk nonlinear interactions [2]. Electric field poling was also demonstrated with waveguide hydrothermally-grown KTP devices [3], with bulk hydrothermally-grown KTP [4] and with bulk $RbTiOAsO_4$ [5]. Recently, electric field poling has been achieved in relatively long and thick (0.5 mm) flux-grown KTP crystals [6,7] and was used for the generation of blue light. KTP crystals are easier to grow by the flux technique than by the hydrothermal technique. As a result, hydrothermally-grown KTP crystals of large size are rare and more expensive. Furthermore, the different growing techniques result in different macroscopic properties, such as the electrical resistivity and the refractive index. Finally, the Sellmeier equations, which describe the wavelength dependence of the refractive index, are different for flux-grown and hydrothermally-grown KTP crystals. In this paper we report detailed studies of the nonlinear conversion efficiency to the green and ultra-violet as well as the temperature and wavelength tuning parameters of flux-grown periodically-poled KTP (PP-KTP).

Our device was a 10 mm long, 0.5 mm thick flux-grown KTP. Titanium gratings with periods of 8.98, 8.99, 9.00 and 9.01 μm were deposited on the C + side of the crystal. The titanium strips were electrically separated by an isolating photoresist layer. A homogeneous layer of titanium was deposited on the C − side of the crystal. Ferroelectric domain inversion was achieved under appropriate conditions [6,7] by applying a single 4.5 kV pulse on the crystal. Monitoring of the switching current and subsequent control of the domain pattern by SEM and optical microscopy methods [8,9] revealed a nearly homogeneous periodic structure with vertical domain walls. The ob-

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served gratings are suitable for second harmonic generation of the 1064 nm Nd:YAG laser near room temperature.

To test the device we used as a pump source a diode-pumped CW 1064 nm Nd:YAG laser (Lightwave Electronics model 122-300). A pulsed Nd:YAG laser was used for efficient doubling with another sample having the same grating periods [10]. The laser light was focused using a 150 mm lens to a slightly elliptic spot size of 19 × 23 micrometers, as measured by the knife edge method. The PP-KTP crystal itself was then placed at the focal point. The crystal was mounted inside a copper housing and its temperature was controlled by a thermo-electric cooler, a thermistor and a temperature controller. The output light, which included both the fundamental and the second harmonic was then separated using a dichroic beamsplitter. The fundamental and second harmonic power were measured by a power meter with germanium and silicon detectors, respectively.

Fig. 1 shows the second harmonic power versus the fundamental power squared, when the $\Lambda = 9.01$ μm grating was used. The PP-KTP crystal was held at the optimal temperature for second harmonic generation of 28.6°C. The only correction applied to the measured power levels was to divide them by the calculated Fresnel transmission (91.4% at 1064 nm and 90.5% at 532 nm) of the crystal output facet. The normalized conversion efficiency is 1.28% $\mathrm{W}^{-1}\ \mathrm{cm}^{-1}$. It should be noted that this value is already larger by a factor of 6 with respect to the highest normalized doubling efficiency attainable with bulk, birefringently phase-matched MgO:$\mathrm{LiNbO_3}$ or KTP and is about 60% of the attainable efficiency with periodically-poled $\mathrm{LiNbO_3}$.

The theoretical frequency conversion efficiency is [11]:

$$\eta_{\mathrm{SH}} = \frac{P_{2\omega}}{P_\omega} = \left(\frac{2\omega^2 d_{\mathrm{eff}}^2 k_\omega P_\omega}{\pi n_\omega^2 n_{2\omega} \epsilon_0 c^3} \right) L h(B,\xi). \qquad (1)$$

Here ω and k_ω are the laser fundamental frequency and wavevector, respectively, n_ω and $n_{2\omega}$ are the refractive indices, c is the speed of light, ϵ_0 the vacuum permittivity and L is the crystal length. $h(B,\xi)$ is the Boyd-Kleinman focusing factor. In our case there is noncritical phase-matching and $B = 0$. The focusing parameter is $\xi = L/b$ where b is the confocal parameter. With an average waist size of 21 μm we obtain $\xi \approx 2.1$ and $h(0,2.1) \approx 1$. For the optimal focusing parameter of $\xi = 2.84$, the Boyd-Kleinman factor reaches its maximum value of 1.07.

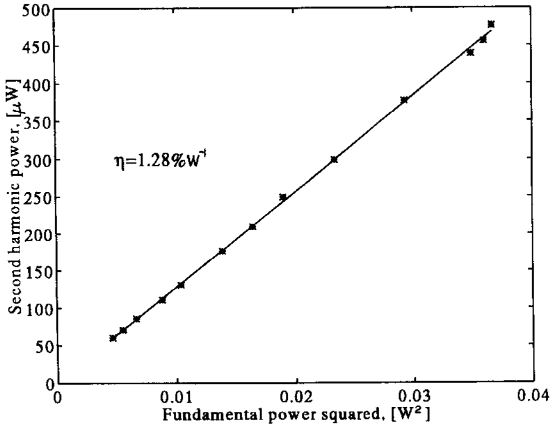

Fig. 1. Measurement of 1064 nm internal doubling efficiency in a 10 mm long PP-KTP crystal.

The effective nonlinear coefficient for QPM doubling of order m is given by [12]

$$d_{\mathrm{eff}} = \frac{2}{\pi m} \sin(\pi m D) d_{33}, \qquad (2)$$

where D is the duty cycle, i.e. the ratio between the size of the inverted ferroelectric domain and the QPM period. In this experiment $m = 1$ and the optimal effective nonlinear coefficient of $2d_{33}/\pi$ is obtained for $D = 0.5$. We can fit the experimental data by assuming $d_{33} = 14.9$ pm/V. This is a lower limit value of d_{33}, because any imperfection in the poling process or deviations in the duty cycle and period will reduce the doubling efficiency. The main uncertainty in our measurements is the systematic error in the measurements of the power levels. By comparing measurements of our power meter with an additional power meter at 532 nm and 1064 nm we concluded that the inaccuracy is less than 10%. We note that our measurement barely agrees with the commonly accepted value, $d_{33} = 13.7$ pm/V [13]. Measurements of doubling efficiency in KTP waveguides [14] indicate that the value of d_{33} could be much higher.

The second harmonic power as a function of the crystal temperature is shown in Fig. 2. The measurements agree well with the theoretical curve and the full temperature width at half maximum is ≈ 6.5 degrees. Several different Sellmeier equations for flux grown KTP [15–17] and hydrothermally grown KTP [13] appeared in the literature. At the operating (vacuum) wavelength of 1064.46 μm, the predicted QPM grating is 9.003 μm according to Ref. [15], 9.103 μm according to Ref. [16], 9.304 according to Ref. [17] and 9.317 μm according to Ref. [13]. Hence, the Sellmeier equations of Fan et al. [15] give the closest estimate to the measured data. We note that the Sellmeier equations we use do not include the temperature dependence of the refractive index.

The peak second harmonic power for gratings with periods $\Lambda = 9.01$, 9.00, 8.99 and 8.98 μm were obtained at temperatures of 28.6°C, 35.4°C, 42°C and 47.6°C, respectively. Hence there is an average shift of 6.3 degrees between successive gratings. The QPM grating itself is related to the refractive indices by

$$\Lambda = \frac{\lambda_\omega}{2(n_{2\omega} - n_\omega)}. \qquad (3)$$

The refractive index difference $(n_{2\omega} - n_\omega)$ changes by 6.57×10^{-5} between successive gratings, hence we can deduce that $\mathrm{d}(n_{2\omega} - n_\omega)/\mathrm{d}T \approx 1.04 \times 10^{-5}$/K. The small

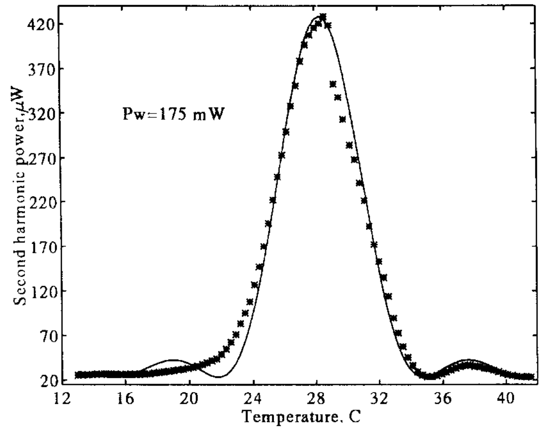

Fig. 2. Second harmonic power at 532 nm as a function of the temperature of the PP-KTP crystal. Solid curve is the theoretical fit to the measured power.

change in Λ owing to thermal expansion [12] is ignored. A direct calculation of the change in the refractive index temperature derivative [18] gives 1.0007×10^{-5}/K, in good agreement with our measurement. We can now use this number together with the measured temperature bandwidth (see Fig. 2) to calculate the effective second harmonic interaction length L_{eff}: The temperature change from the peak to the first minimum was ≈ 6.5 degrees, which means that $(\Delta k L_{\mathrm{eff}}/2) = \pi$, with $\Delta k = k_{2\omega} - 2k_{\omega} - 2\pi/\Lambda = 4\pi(n_{\omega} - n_{2\omega})/\lambda_{\omega} - 2\pi/\Lambda$. The refractive index difference $(n_{2\omega} - n_{\omega})$ changes by $6.5 \times 1.04 \times 10^{-5}$. This implies that $L_{\mathrm{eff}} \approx 8$ mm, in reasonable agreement with the 10 mm physical size of the sample.

We have also measured the wavelength tuning of the second harmonic peak within the gain bandwidth of the Nd:YAG laser. This was done by changing the laser temperature from 22.7°C to 49.5°C. The fundamental wavelength, which was measured using a Burleigh WA-20 wavemeter, shifted from 1.064462 μm to 1.064575 μm. The temperature of the KTP crystal had to be readjusted for achieving maximum second harmonic generation, from 28.6°C (35.4°C) to 30.4°C (37.3°C) for the $\Lambda = 9.01$ μm ($\Lambda = 9.00$ μm) period. The average temperature shift is therefore ≈ 60 pm/K. The temperature shift of hydrothermally-grown KTP is 55 pm/K [19].

To test the homogeneity of the device we have scanned it in the Z (vertical) direction. The beam waist in this case was 30 μm. The second harmonic power, shown in Fig. 3, was nearly constant over the scan length of 0.35 mm. The scan length was limited by beam divergence in the 0.5 mm thick sample. This is an indication that poling occurred over the entire thickness of the device. In the horizontal (Y) direction the second harmonic power was nearly constant for a width of ~ 0.5 mm out of the 1 mm stripe of each QPM grating.

The periodically-poled KTP we used is also suitable for third order QPM doubling of 784 nm light. Although the doubling efficiency is a factor of 9 lower than that of first order doubling, the effective nonlinear coefficient, $2d_{33}/3\pi \approx 3.2$ pm/V is still large compared to that of nonlinear materials used in the ultraviolet range, e.g. BBO and LBO. To test the doubling efficiency we used an Ar-Ion pumped Ti:Al$_2$O$_3$ laser, Coherent model 890. The laser was focused using a 150 mm lens to a (nearly circular) waist of 20 μm. The fundamental and second harmonic were then separated using a heat reflecting mirror. The fundamental beam was directly measured using a power meter with a silicon detector whereas the second harmonic beam passed through another short-wave pass filter and measured by a second power meter with a silicon detector. Most of our measurements were performed with the 8.98 μm grating, whose peak second harmonic power was at a wavelength of 783.5 nm. The generated UV light as a function of the crystal temperature exhibited a narrow central peak having full width at half maximum bandwidth of 2.2°C. The normalized conversion efficiency, see Fig. 4, was 0.12% W^{-1}. The highest internal UV power of 75.3 μW was generated with internal pump power of 259 mW. With the focusing parameters we used, the normalized conversion efficiency for ideal third order QPM doubling should be 0.72% W^{-1} (taking here into account the multimode spectrum of the Ti:Al$_2$O$_3$ pump laser). This lower than expected conversion efficiency may be due to the duty cycle being different from 50%. The third order QPM process is much more sensitive than the first order QPM to this effect. As can be calculated from Eq. (2) the highest efficiency for $m = 1$ and $m = 3$ is achieved with $D = 0.5$, however, if $D = 0.4$ (or $D = 0.6$) for example, the efficiency of the first order process will be reduced by only 10%, whereas for $m = 3$ the second harmonic power is reduced by 65%. Moreover, for $D \approx 0.33$ the first order doubling efficiency is reduced by 14%, but the third order doubling efficiency becomes zero. We should note that our

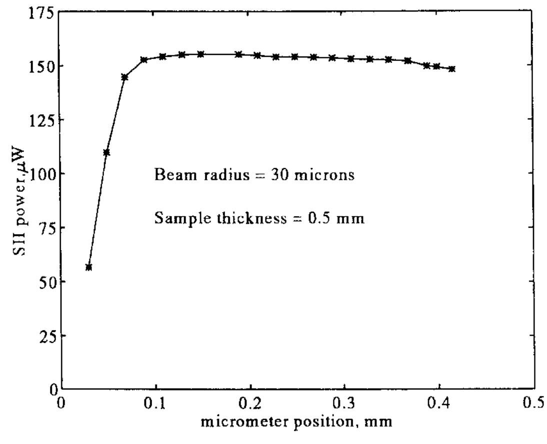

Fig. 3. Second harmonic power at 532 nm as the 30 μm waist beam was vertically scanned along to 0.5 mm thick PP-KTP sample.

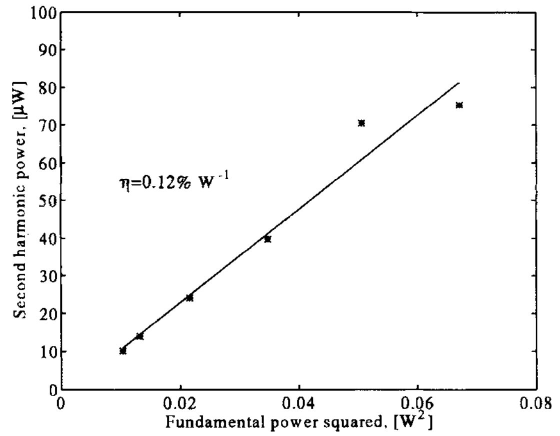

Fig. 4. Measurement of 784 nm internal doubling efficiency in a 10 mm long PP-KTP crystal.

very preliminary result in the UV is already comparable with the highest attainable normalized doubling efficiency for bulk LBO and BBO. Furthermore, the doubling process in PP-KTP is noncritically phase-matched.

We have also tested the device with an SDL 5401 diode laser, operating in an external cavity Littrow configuration and emitting 30 mW at 784 nm. The second harmonic power was visible on a screen but was too weak compared to the fundamental power leaking through the filter to be measured in a reliable fashion. We used this setup to measure using a wavemeter the optimal doubling wavelength for the different QPM periods. For gratings with periods $\Lambda = 9.01$, 9.00, 8.99 and 8.98 μm the corresponding optimal wavelengths are 784.1 nm, 783.9 nm, 783.7 nm and 783.5 nm.

The device was operated for more than 50 hours under Nd:YAG radiation and more than 10 hours under Ti:Al_2O_3 radiation. After that we rechecked the 1064 nm doubling efficiency and found it to be the same as at the beginning of the experiments. We did not notice any other visible degradation, e.g. in the quality of the beam.

In summary, PP-KTP provides high nonlinear efficiency, as well as room temperature operation, and was used here to generate green and UV light. The 1064 nm doubling efficiency is already close to the theoretical limit. The UV radiation, however, which was generated by a third order QPM process, exhibited lower efficiency and the main cause seems to be that the QPM duty cycle was not 50%. Further improvements in the doubling efficiency to the UV are expected by tight control of the duty cycle and by poling the device for first or second order QPM doubling.

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