

Efficient doubling of femtosecond pulses in aperiodically and periodically poled KTP crystals

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Abstract: Efficient doubling of femtosecond pulses in periodically and aperiodically poled KTP crystals is demonstrated by employing the quasi-phasematched frequency conversion technique. Conversion efficiencies as high as 60% were achieved using either aperiodically or periodically poled KTP nonlinear crystals in an extracavity, single-pass configuration using a diode-pumped femtosecond Yb:KYW laser as the pump source. The temporal characteristics of the frequency-doubled pulses as a function of focusing conditions in a “thick” nonlinear crystal regime have been investigated experimentally and pulses as short as 177 fs have been generated at around 520 nm under strong focusing conditions using a KTP crystal with aperiodic poling.

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References and links

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1. Introduction

Second harmonic generation (SHG) is an attractive means of producing laser light in the ultraviolet and visible spectral ranges that cannot be covered readily by tunable solid-state lasers. This technique has become increasingly viable through the combination of new frequency doubling schemes and novel nonlinear materials. For instance, it was demonstrated that efficient SHG is possible by extracavity single-pass frequency doubling of a focused femtosecond laser beam in a "thick" nonlinear crystal [1]. The case of a "thick" nonlinear crystal is applicable when the length of a nonlinear crystal considerably exceeds a walk-off length over which two initially temporally overlapped pulses at different wavelengths become separated by a time equal to the fundamental pulse duration. Such a scheme has the advantage of simplicity when compared with alternative intracavity frequency doubling and avoids the drawbacks associated with the "green problem" [2]. Additionally, ultrashort second harmonic pulses can be generated under the conditions of strong focusing where the depth of focus can be considerably smaller than the length of a nonlinear crystal [3]. This type of scheme thus adds considerably to the versatility of the ultrafast lasers that are relevant to a range of applications that include time-resolved spectroscopy, studies in photobiology and photomedicine.

The quasi-phasematched (QPM) frequency conversion technique has become very well established since the electric field poling method was first demonstrated for LiNbO₃ (LN) [4, 5] and is currently the most attractive and practical scheme for nonlinear frequency conversion over broad spectral ranges. QPM has key advantages over birefringent phase matching that include a wide phase-matchable spectral range, accessible noncritical-phase matching and access to the largest nonlinear coefficients of the materials employed. To date, the most commonly used ferroelectrics for periodic poling are crystals from the LN or potassium titanyl phosphate (KTP) [6] families. It has also been shown that the periodically-poled bulk KTP crystal (pp-KTP) can be used efficiently for the frequency doubling of pulsed lasers having pulse durations as short as ~ 1 ps [7]. Conversion efficiencies in frequency doubling of up to 20% have been achieved in pp-KTP by employing an extracavity single pass configuration for a femtosecond Ti:sapphire laser for which sub-picosecond pulses were generated at around 425 nm [8]. It has also been reported for a diode pumped femtosecond Cr:LiSAF laser (fundamental wavelength of 860 nm) that frequency doubling in bulk aperiodically-poled and periodically-poled waveguide KTP crystals afforded SH conversion efficiencies of up to 12% and 37% respectively [9]. More recently, we demonstrated efficient generation of femtosecond pulses at 525 nm by extracavity frequency doubling of the output of a diode-pumped femtosecond Yb:KYW laser using a periodically poled LiTaO₃ crystal where the nonlinear conversion efficiency was observed to be 43% [10]. It is appreciated that the main limiting factor for SH conversion efficiency in the case of doubling of femtosecond laser pulses in "thick" nonlinear crystals is the presence of a significant group velocity mismatch (GVM) leading to temporal walk-off. Additionally, the spectral acceptance bandwidth of a nonlinear crystal can be less than the spectrum of femtosecond pulses. In these circumstances, it is vital to optimize the beam focusing conditions in the nonlinear crystals that are deployed.

In this paper, we report efficient frequency doubling of a diode-pumped femtosecond Yb:KYW laser output in an extracavity, single-pass configuration that incorporates either

aperiodically or periodically poled KTP crystals. Comparative and quantitative characterisations for both the periodic and aperiodic structures have been undertaken. The nonlinear conversion efficiency of 64% achieved in pp-KTP represents a highlight result as does the generation of green pulses (durations as short as 177 fs) using a KTP crystal with aperiodic poling.

2. Experimental set up

The pump source used in this work was a diode-pumped Yb:KYW laser which was passively mode locked using a semiconductor saturable absorber mirror. It produced femtosecond pulses of 130-140 fs duration in the 1040-1050 nm wavelength range with a pulse repetition frequency of 77.9 MHz and average output powers of 240-320 mW. The corresponding spectral width for the pulses was 9 nm which implies time-bandwidth product of 0.34. The output beam was characterised by a slightly elliptical spatial mode with a 1.12:1 ratio of ellipticity and the M^2 factor was measured to be 1.13 and 1.17 in the sagittal and tangential directions respectively. The 4.8mm-long pp-KTP and 2.7mm-long aperiodically poled KTP (app-KTP) nonlinear crystals of 1mm-thick were fabricated by the electrical poling technique and all were deployed in a type I ($e+e\rightarrow e$) first-order, quasi phase-matched SHG in which the d_{33} nonlinear coefficient was utilized. To maximize the effective nonlinear coefficient, the QPM order was chosen to be $m=1$ and the duty cycle, D , was 0.5. For this polarization the nonstationary length L_{nst} is around 200 μm , and the crystals are thus "thick". L_{nst} is the temporal walk-off characteristic length and is defined as $L_{nst} = t_p c / (n_{g2} - n_{g1})$, where t_p is the pulse length, c is the speed of light in vacuum and n_{g2} and n_{g1} are the group indices of the SH and the fundamental, respectively. The crystal facets were optically polished and antireflection-coated at both the fundamental and second harmonic wavelengths. The grating period for the periodically-poled crystal of 8.34 μm had been designed for QPM at 1040.5 nm at 50° C [11, 12]. The app-KTP had linearly chirped grating periods ranging from 8.24 μm to 8.44 μm over the lengths of the crystal sample to facilitate a phase-matched bandwidth of around 8 nm at the fundamental wavelength. The SHG data were taken with a set of focusing lenses having focal lengths covering the range of 63 mm to 4.5 mm and these corresponded to fundamental beam waist radii (w_0) inside the nonlinear crystals of 24 μm to 1.6 μm and confocal parameter ($b = 2\pi w_0^2 n / \lambda$, where n is the refractive index of the nonlinear crystal and λ is the fundamental wavelength) values from 6.3 mm to 28 μm .

3. Results and discussion

At the outset of these evaluations, the SHG efficiency of the nonlinear crystals was measured as a function of temperature for different L/b ratios, where L is length of the nonlinear crystal. Initially, the broadband SH temperature tunability was measured for the pp-KTP crystal when pumped at 1040 nm [Fig. 1(a)]. The peak of conversion efficiency curve shifts to lower temperatures with increase in the L/b parameter so that efficient SH generation was possible at room temperature with tight focusing. Such behaviour can be explained by the fact that in this condition a suitable angle is introduced between the wavevectors of the fundamental beam and the periodic structure (except for the region where L is approximately equal to b). In this case the period of the nonlinear structure can be substituted by an "effective" one that increases with tighter focusing, so that the QPM condition can be satisfied at a lower temperature of the nonlinear crystal for a given pump wavelength. It can be seen from Fig. 1(b) that for weak focusing ($b > L$) the temperature tuning characteristic for app-KTP is similar to those for pp-KTP but the peak SHG efficiency is now around 100° C because the pump used was centred at 1045 nm for this assessment. Under the conditions of tight focusing, where $b \ll L$, the interaction length over which the SHG process is most efficient reduces to approximately b so that different period groups in the app-KTP crystal can be selected by

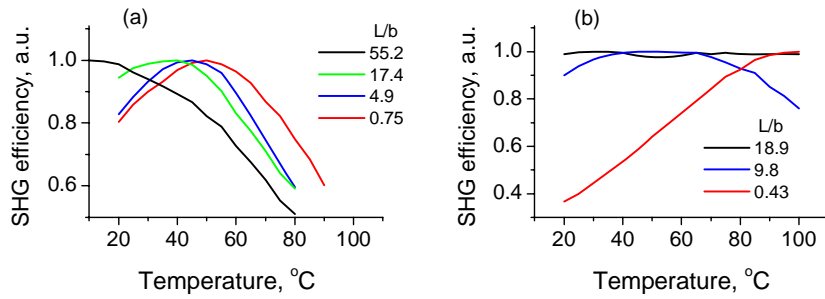


Fig. 1. Dependence of the SHG efficiency on temperature of the pp-KTP crystal (a) and app-KTP crystal (b) at different L/b parameters.

translating the focusing lens longitudinally. It can be seen from Fig. 1(b) that under these conditions of varying the pump light focus in the aperiodically poled nonlinear crystal and at $b \ll L$ negligible changes in the SHG efficiency are observed across the 10-100°C temperature latitude.

The maximum SH conversion efficiencies were observed in both crystals at L/b in the 5-10 range [Fig. 2(a)]. Specifically, 180 mW of green power was generated with a SHG efficiency of 60% from app-KTP crystals at $L/b=9.8$ [Fig. 2(b)]. SHG efficiencies as high as 64% were achieved with 145 mW of average green power when the pp-KTP crystal was deployed with $L/b=4.9$ [Fig. 2(b)]. Further increases in L/b led to an observed decrease in the SH conversion efficiency due to a reduction in the interaction length ($\sim b$) and, partly, the possible walk-off for the large angles involved with tight focusing. The input-output characteristics demonstrated significant pump depletion in both nonlinear crystals for all of the focusing conditions described in this paper. Using measurements of conversion efficiency in the loose-focus low power limit (cw source at 1040 nm) we calculated the effective nonlinear coefficient to be 8.6 pm/V for pp-KTP. This value is in a good agreement with the theoretical one of 10.75 pm/V ($d_{33}=16.9$ pm/V). The quality factor M^2 for the frequency-doubled output beam was measured to be around 1.2 for weak fundamental beam focusing

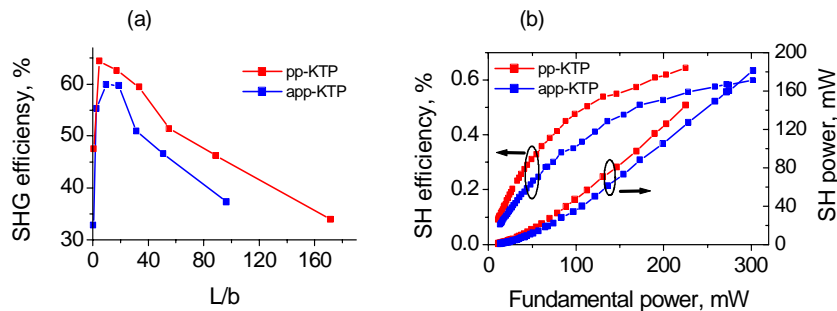


Fig. 2. (a). SH conversion efficiency versus L/b focusing ratio in 4.8mm-long pp-KTP and 2.7mm-long app-KTP; (b) Measured SH average output power and SH conversion efficiency versus incident fundamental power.

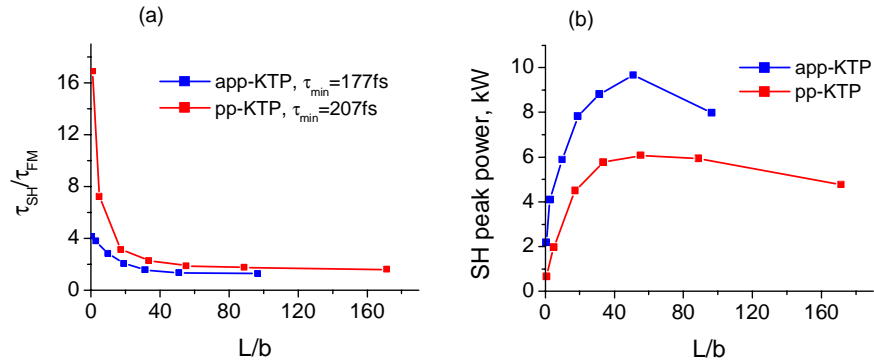


Fig. 3. (a). Ratio of the SH pulse duration to the fundamental pulse duration and (b) SH peak power as a function of focusing parameter L/b .

and 1.8 in the strong focusing conditions using either pp- or app-KTP crystals.

The temporal characteristics of the SH pulses were investigated using intensity autocorrelation technique and the duration of the frequency-doubled pulses in pp- and app-KTP crystals were deduced to be 2.2 ps and 0.58 ps respectively under the weakest focusing conditions, ($b>L$). Shorter duration green pulses were observed for tight focusing in both nonlinear crystals as a consequence of a reduced interaction length ($\sim b$). In Fig. 3(a) the ratio of second harmonic and fundamental pulse durations (τ_{SH}/τ_{FM}) is presented as a function of the focusing strength L/b . [Sech² pulse profiles assumed.] Green pulses as short as 207 fs were measured at 520 nm for the SHG process in pp-KTP at $L/b=171$. When an app-KTP crystal having a L/b ratio of 96 was employed, the SH pulse duration was shortened to 177 fs which is 40% longer than that of the fundamental laser pulse.

With regard to applications of SH femtosecond pulses it might be required to optimise the SHG laser system with one particular parameter in mind. For instance, it this could be the SHG efficiency, SH pulse duration or generated peak power. It has been shown in this study that by varying the L/b parameter we can achieve a maximum conversion efficiency at $L/b\sim 10$ and ultrashort SH pulses can be generated at large L/b ratios that are limited only by the nonlinear crystal input aperture. By way of illustration, the peak power of green pulses as a function of the L/b ratio is included as Fig. 3(b). Although the pp-KTP crystal was shown to have a higher conversion efficiency than the app-KTP, a greater SH peak power was available using the aperiodic crystal ($P_{peak}=9.7$ kW) because of the shorter pulse duration at a given L/b ratio. The maximum SH peak power was generated at $L/b\sim 50$ for both crystals. No green power degradation was observed during observational periods of hours at estimated maximum green power density inside the nonlinear crystal of around 60 GW/cm².

Examination of the SH output spectra profiles demonstrated a significant influence of the fundamental light focussing conditions for both pp- and app-KTP crystals at selected pump wavelengths and nonlinear crystal temperatures. In the case of the periodically-poled crystal, the SH spectrum was symmetric with a width of around 0.4 nm for weak focussing [see Fig. 4(a)]. The spectral width increases significantly at strong focusing due to the reduction in the effective crystal length. Additionally, spectral broadening arises predominantly to the longer wavelengths. The asymmetry of SH spectrum can be explained by considering the non-collinear QPM case. At $b\gg L$, the presence of angle between fundamental wave vector and the periodic structure wave vector means that the QPM conditions can be satisfied at wavelengths that are longer than those which correspond to QPM at zero angle between the wave vectors of the pump light and the periodic structure ($L\sim b$ region). With the app-KTP crystal, relatively broad and multi-structural SH spectra were observed in the weak focusing

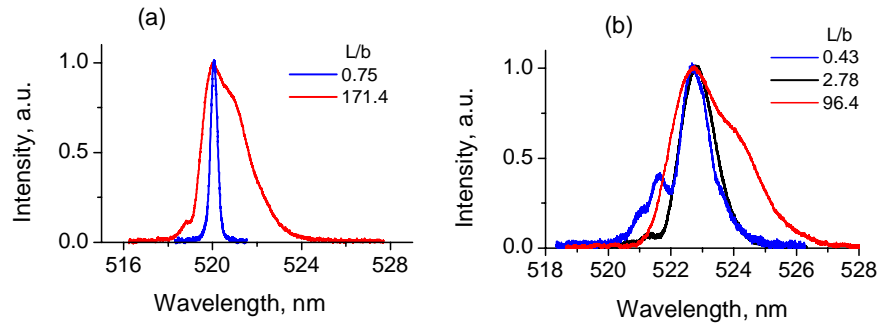


Fig. 4. Measured SH spectra at different L/b ratios for pp-KTP (a) and app-KTP (b).

condition because the entire period range that had been designed to facilitate a phase-matched bandwidth in the fundamental wavelength of around 8 nm could be utilized [see Fig. 4(b)]. With reductions in b the range of periods effectively employed in SHG is decreased. This gives rise to a narrower SH spectrum at $L/b=2.78$, compared to those at $L/b=0.43$. Further increases in L/b was seen to lead to a spectral broadening of the spectra for the green pulses due to the effects described above for pp-KTP. Thus, under strong focusing conditions SH spectra have similar bandwidths and shapes for both types of nonlinear crystals.

4. Conclusion

We can conclude by claiming that highly efficient frequency doubling of femtosecond pulses can be achieved using either pp-KTP or app-KTP nonlinear crystals. The SH conversion efficiencies of up to 60% and 64% that have been demonstrated in this work for app- and pp-KTP crystals, respectively, represent, to the best of our knowledge at least, the best performance yet reported for the frequency doubling of femtosecond laser pulses. Given the high optical-to-optical efficiency of our mode-locked ytterbium-based femtosecond laser, we can deduce the overall diode pump light-to-SH light conversion efficiency to be an impressive 18%. Efficient frequency doubling was achieved over a wide range of nonlinear crystal temperatures that include room temperature and the applicability of the extracavity “thick” nonlinear crystal regime has been demonstrated convincingly.