Efficient generation of green and UV light in a single PP-KTP waveguide pumped by a compact all-fiber system

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We present simultaneous efficient second- (SHG) and third-harmonic generation (THG) in a single periodically-poled KTP waveguided crystal pumped by a compact femtosecond Yb-based laser in a condition of exactly phase-matched frequency doubling and nonphase-matched sum-frequency mixing processes. Internal conversion efficiency as high as 33% for SHG (532 nm) and ~2% for the cascaded THG (355 nm) is reported. We believe this to be a clear experimental demonstration that strong third-harmonic can be generated in frequency doubling crystals through a nonphase-matched sum-frequency mixing process. © *2006 American Institute of Physics*.
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Sources of ultrashort pulses in the visible and UV spectral regions are important for a wide range of applications in ultrafast science and technology. Recent progress in biophotonics research such as microscopy, optical micromanipulation, and biomedical imaging continues to strengthen the need for compact, low-cost, visible, and UV sources that offer portability and practicality. Such laser-based sources have clear advantages over near-infrared lasers in allowing for stronger beam focusing, enhanced resolution in multidimensional imaging techniques[1] and high-resolution spectroscopy.[2] By using ultrashort-pulse lasers in preference to continuous-wave sources, it is possible to investigate ultrafast biological processes,[3] increase the resolution in microscopy,[4,5] and amplify weak signals in nonlinear and multiphoton techniques.[6–8] Imaging and display technology also draw useful benefits from ultrashort visible pulses, in breakthrough applications such as red/green/blue laser displays.[9] To date, frequency multiplication in nonlinear optical crystals has been the most efficient way to generate coherent visible and UV light. By employing small, robust crystals with periodically-poled structures to improve wavelength conversion efficiency[10,11] it has been possible to make frequency conversion the preferred candidate for out-of-the-lab visible and UV pulsed sources. The advent of fiber lasers provided a reliable and small footprint laser source[12] for use in a large selection of applications, including frequency conversion. When combined in a single configuration, a fiber laser and a suitable periodically poled crystal represent a practical and efficient source option for industrial use. In this paper, we report a periodically-poled KTP (PPKTP) crystal having a predesigned waveguide structure to implement a compact pulsed source for simultaneous generation of visible and UV light. A brief theoretical analysis is included to confirm that efficient frequency doublers can simultaneously yield efficient third harmonic. Several ways to improve the current performance are also suggested.

The PPKTP frequency-doubling crystal used in our work was fabricated by an ion-exchange and chemical poling technique.[13] This 12 mm long crystal was designed to provide optimized second-harmonic generation of 1065 nm incident radiation, at a temperature of 60 °C. The crystal is placed on a thermoelectric cooler/heater within a control loop that enabled the crystal temperature to be maintained to an accuracy of better than 0.1 °C. The pump light was focused in the crystal waveguide (cross-sectional area of $4\ \mu m \times 7\ \mu m$) without any special requirement concerning the Rayleigh range of the beam. To ensure maximum flexibility in operational wavelength, each of the embedded waveguides has been poled with a slightly different period ranging from 8.22 μm–8.72 μm. The duty factor D was 0.6. Therefore, tuning for the optimal conversion wavelength could be achieved either by tuning the temperature of the

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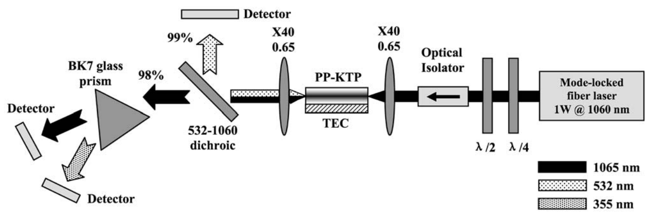

FIG. 1. Simplified layout of the experimental setup.

crystal or by changing the waveguide employed in second-harmonic generation (SHG).

The experimental setup for frequency doubling is shown in Fig. 1. The pump laser was built around a master oscillator-power amplifier (MOPA) architecture. The master source was a compact fiber laser mode-locked by a semiconductor saturable absorber mirror and its output signal power was boosted in a large mode area double-clad fiber amplifier pumped by a stack of 920 nm diode lasers. Driven at full power, the MOPA system could produce up to 1 W of average power with linearly polarized 3 ps pulses, at a repetition rate of 100 MHz. Figure 2(a) shows the optical spectrum of the laser operating at full power. A typical intensity autocorrelation trace is also provided as an inset. Self-phase modulation features are obvious in the spectrum, and any further increase in the average power was restricted by the onset of Raman effects.

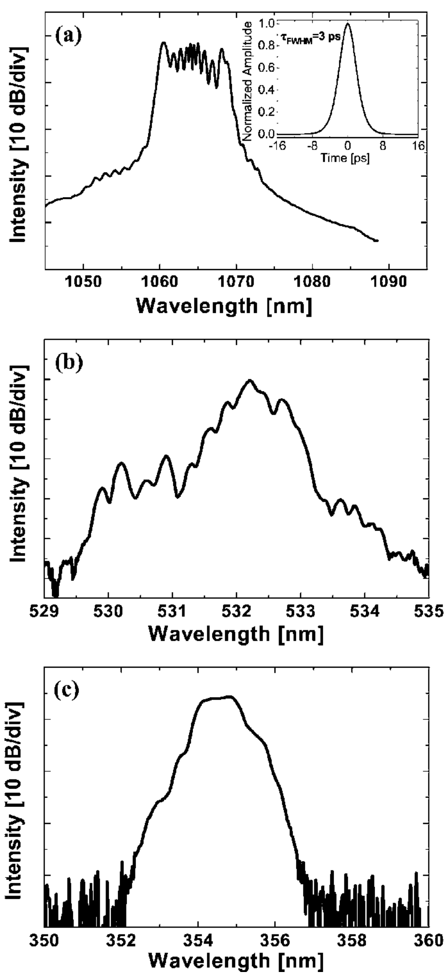

FIG. 2. (a) Optical spectrum of the fundamental radiation, inset shows the corresponding intensity autocorrelation trace ($\tau_{FWHM} \sim 3$ ps); (b) Optical spectrum of the SH radiation. (c) Optical spectrum of the TH radiation.

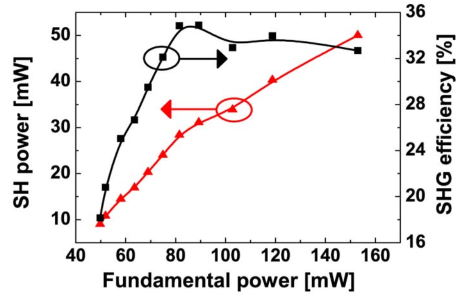

FIG. 3. (Color online) Generated SH average power and the internal efficiency of the PPKTP waveguide crystal as functions of the average power of the fundamental radiation.

When the laser beam was focused into the waveguide of the nonlinear crystal, intense nonlinear interactions give rise to a strong second-harmonic (SH) output that was collected by a microscope objective (Newport 40X) placed at the output of the crystal. To accurately measure its characteristics, the SH radiation was separated from the residual fundamental light by means of a dichroic mirror exhibiting 99% reflectivity at 532 nm, 98% transmission at 1060 nm, and ~95% transmission at 355 nm. Subsequent separation of the third-harmonic light from the fundamental was accomplished using a BK7—glass prism placed in the beam path.

The maximum measured second harmonic power was about 50 mW. Following collimation and separation from residual fundamental pump light and higher order harmonics, the SH signal was fed to a spectrometer and optical power meter. Figure 2(b) is a reproduction of the SHG optical spectrum. The conversion efficiency curve is plotted in Fig. 3. The saturation effects arising from strong two-photon absorption (TPA) (Ref. 14) and THG are evident in this plot. We measured a maximum SHG efficiency of 33%, where the main limiting factors in conversion efficiency are attributed to the limited spectral acceptance bandwidth of the nonlinear crystal and TPA.

The residual light transmitted through the dichroic mirror was directed through a BK7 glass prism that provides spectral dispersion. It was thus possible to separate the third harmonic beam component from the fundamental and to direct it to the input port of an optical spectrum analyzer. This produced the spectrum shown in Fig. 2(c), which exhibits a profile close to that of the second-harmonic signal. The bandwidth of the third-harmonic radiation is 1 nm which matches the input acceptance bandwidth of the nonlinear crystal. It is worth noting that, for the particular wavelength of the generated third-harmonic radiation, the transparency of the BK7 glass prism in Fig. 1 varies strongly with wavelength. However, owing to the narrow spectrum of the third-harmonic radiation, we do not expect this unwanted feature to cause any significant distortion in the measured spectral shape in Fig. 2(c).

Notably, the second- and third-harmonic radiations are generated *simultaneously* by nonlinear interactions that arise in the crystal. The power of the third-harmonic radiation was 3 mW, implying a conversion efficiency of 2%. Generation of third-harmonic radiation is attributed to a series of $\chi^{(2)}:\chi^{(2)}$ nonlinear cascaded processes (Ref. 15)—in our case the process of phase-matched SH generation followed by nonphase-matched sum frequency mixing (SFM) between the depleted fundamental and the strong SH light generated in the same waveguide.

To understand the mechanism of the generated TH signal, we introduce the quantity $M_{\mathrm{coh},m}=\pi/\Delta k_{2,m}$ ($\Delta k_{2,m}=k_3-k_2-k_1-2\pi m/\Lambda$) that we call quasi phase-matched (QPM) *coherence length* in analogy to the well known coherence length in volume nonlinear crystals. Using the Sellmeier equation formulation reported in literature,[16] we calculated the QPM coherence length for the processes of SFM in the PPKTP crystal for the relevant period of 8.8 μm that corresponds to first order QPM matching for the SHG process. We found that for $\lambda_{\mathrm{fund}}=1.065$ μm the SFM processes have QPM coherence lengths that correspond to different QPM orders of the SFM process equal to: $M_{\mathrm{coh},m}=\{1.5;2.3;4.7;71.0;4.2\}$ μm for $m=1$–5. For other m the QPM coherence length is even smaller. Thus one can expect rather efficient SFM processes and efficient generation of the TH signal with fourth-order QPM matching ($M_{\mathrm{coh},4}=71$ μm). The system of equations that describes the cascaded THG in single quadratic media with the plane wave and slow varying envelope approximations was taken from previous publications.[15] With the assumptions of exact phase matching for the SHG process, nondepletion of the fundamental wave due to SHG and THG, and nondepletion of SH wave due to THG, the system is simplified to:

$$\frac{dA_2}{dz}=-i\sigma_1 A_1^2; \quad \frac{dA_3}{dz}=-iA_1A_2\sum_{m(m\neq 0)}\sigma_{3,m}\exp(i\Delta k_{2,m}z), \tag{1}$$

where $\sigma_1=2\pi d_{\mathrm{eff},1}/\lambda_1 n_1 g_1$; $\sigma_{3,m}=2\pi d_{\mathrm{eff},3m}/\lambda_3 n_3 g_3$; with $d_{\mathrm{eff},1}=d_{zzz}2/\pi\sin(\pi D)$, $d_{\mathrm{eff},3m}=d_{zzz}2/m\pi\sin(m\pi D)$, and g_j—are the overlapping integrals. The duty factor D of the QPM structure used in our experiment is $D=0.6$, and therefore even order QPM matching is possible. The simplified solution of Eq. (1) for the squared third-harmonic amplitude when the small oscillating terms are neglected (except the term with $m=4$) gives

$$|A_3|^2\approx\left(\frac{1}{\pi}\sigma_1\sigma_3|A_1|^3 L M_{\mathrm{coh},4}\right)^2. \tag{2}$$

From Eq. (2), it is evident that despite the fact that the second step (SFM) is nonphase matched, the total cascaded process of THG behaves in the manner of a phase-matched third-order process. This means we do not expect periodic modulation of the efficiency of THG on the length of the media. TH intensity grows quadratically with nonlinear media length and has cubic dependence on the input intensity $I_3\propto I_1^3$. The longer the QPM coherence length $M_{\mathrm{coh},m}$ of the SFM process, the more efficient the THG becomes. Estimates derived from Eq. (2), assuming $g_1=g_2=0.5$, show that with similar conditions experienced in this work, one can expect a THG efficiency of around 2.7% at an input power of 150 mW. It should be noted that the calculated efficiency does not take into account the absorption at $\lambda_{3\omega}=0.355$ μm.[16] The exact value of the overlap integrals g_1, g_2 is unknown and the effect of group velocities mismatch has also been neglected in our analysis. We consider the measured experimental efficiency to be consistent with this estimation. We thus demonstrated that efficient TH signal can be obtained under the condition of an initial phase-matched SHG process with a subsequent nonphase-matched SFM process. This is in contrast to some previously published conclusions[17,18] that efficient THG with phase matched SH process in single quadratic crystal is not possible. However, in several other publications, theoretical models have been derived that are in agreement with our work (citations in Ref. 15), but they do not include experimental demonstrations of efficient THG as reported here. Previously reported THG efficiencies in experiments of this type in bulk KTP and KDP were less than 10^{-7} and thus impractical.[17,19]

In conclusion, we demonstrated simultaneous second- and third-harmonic generation in a PPKTP waveguided crystal in a condition of exactly phase matched frequency doubling and nonphase-matched sum-frequency mixing processes. The pump laser source was a compact, all fiber, high power mode-locked system, that provided a good quality beam and hence efficient light coupling into the crystal. The waveguide structure of the crystal allowed us to use simple, low focal length collimating optics. This system is a possible candidate for providing the portable visible and UV light sources of the near future. It is worth noting that current system performance could be improved by employing a narrow spectrum laser source achievable, for instance, by using a large mode area fiber amplifier to defeat nonlinear processes in the current fiber amplifier. Moreover, the crystal acceptance bandwidth can be improved by using an aperiodically poled crystal with synthesized grating profile optimized for the available laser source.

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