Widely tunable picosecond optical parametric generation and amplification in BiB₃O₆

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Abstract: Efficient generation of widely tunable picosecond pulses from the visible to near-infrared is demonstrated by optical parametric generation and amplification in BiB₃O₆. Pumped by the second harmonic of an amplified mode-locked Nd:YAG laser at 532 nm, also generated in BiB₃O₆, a signal and idler tuning range of 740-1893 nm has been achieved with angle tuning under type I (o→e+e) phase-matching in the optical yz-plane. With 40-ps pump pulses of 420-μJ energy, single-pass signal pulse energies of up to 48.6 μJ have been obtained at total OPA pump to signal and idler conversion efficiency as high as 30%. Significant temperature tuning under type I (o→e+e) noncritical interaction along the optical z-axis is also demonstrated, extending the signal tuning range from 740 nm down to 676 nm and idler tuning range from 1893 nm up to 2497 nm. Using second harmonic generation of the amplified signal pulses, also in BiB₃O₆, wavelength extension to 370-500 nm has been achieved at 24% conversion efficiency, providing 10-μJ pulses across the tuning range. Optical parametric generation and amplification in BiB₃O₆ under strong two-photon absorption pumped by 210-μJ pulses at 355 nm is also reported, providing amplified signal pulse energies of 14.2 μJ at OPA conversion efficiency as high as 21% and a spectral coverage across 450-1674 nm.

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References and links


1. Introduction

The development of new nonlinear crystals with improved optical characteristics is of vital interest for a wide range of frequency conversion devices and applications. Over the past decade a number of practical birefringent materials including β-BaB₂O₄ (BBO), LiB₃O₅ (LBO), and KTiOPO₄ (KTP) have been successfully developed and utilized in a variety of nonlinear optical applications. For frequency conversion of high energy pulses, BBO and LBO have been established as materials of choice, because of their high optical damage threshold, moderate nonlinear coefficients (dₑffective~1-2 pm/V), deep ultraviolet (UV) transparency, low cost and ready availability.

Bismuth triborate, BiB₃O₆ (BIBO), is a relatively new nonlinear material with unique optical properties for frequency conversion applications [1-3]. BIBO is a monoclinic crystal of space group C2. It offers an optical transparency from ~280 nm in the UV up to ~2.7 μm in the infrared (IR). While the UV transmission cutoff of BIBO is at a longer wavelength than BBO and LBO, it offers substantially larger effective nonlinearity (dₑffective~3.7 pm/V) [4], which is comparable to KTP. As a biaxial crystal, BIBO also exhibits very versatile phase-matching properties, large angular and spectral acceptance bandwidths, low spatial walkoff and broadband angle tuning at room temperature [3]. Such combination of properties makes BIBO an attractive new alternative for frequency conversion applications in the UV, visible and near-IR.

Since the first studies of the linear optical properties of BIBO [2], several frequency conversion experiments have been reported in this material. These include internal second harmonic generation (SHG) of continuous-wave (cw) radiation at 1.06 μm [5], single-pass SHG of high-intensity pulsed laser at 1.06 μm [6], Q-switched internal SHG at 1.06 μm [7], internal frequency-doubling of cw 946-nm Nd:YAG laser [8], photo-induced SHG in partially crystallized BIBO glass [9], and nanosecond optical parametric oscillation and harmonic generation [10,11]. Other demonstrations include efficient single-pass SHG and third harmonic generation (THG) of microjoule picosecond pulses [12], efficient SHG of high-repetition-rate picosecond and femtosecond pulses tunable in the near-UV and blue [4,13], a femtosecond optical parametric oscillator tunable across the visible [14] and femtosecond optical parametric generation and amplification at 1 kHz pumped by an amplified Ti:sapphire laser [15,16].

Here we report efficient parametric generation (OPG) and amplification (OPA) of microjoule picosecond pulses tunable from 676 nm to 2497 nm in BIBO using angle-tuned type I (β→e+e) phase-matching in the optical yz-plane as well as noncritical temperature-tuned type I (α→e+e) interaction along the optical z-axis, with an OPA conversion efficiency.
as high as 30%. We have also extended the demonstrated tuning range to 370-500 nm by SHG of the amplified signal pulses, also in BIBO, at a conversion efficiency of 24%. We further demonstrated tunable optical parametric generation and amplification in BIBO pumped by third harmonic of picosconed Nd:YAG laser at 355 nm under strong two-photon absorption.

2. Experimental setup

A schematic of the experimental configuration is shown in Fig. 1. The pump pulses at 532 nm were obtained by SHG of an amplified mode-locked Nd:YAG laser at 1.064 μm [12]. The laser delivers 35-ps pulses with energies up to 1 mJ at 25 Hz repetition rate in a TEM₀₀ spatial profile with a M²~1 and a beam diameter (full width at 1/e² level) of 2w₀=2 mm. Frequency-doubling was achieved in a uncoated 3×4×10 mm³ crystal of BIBO, cut for type I (e+e→o) phase-matching in the optical yz-plane at θ=169º (φ=90º), with a calculated effective nonlinearity deff~3.3 pm/V [3]. A maximum useful pump pulse energy of 420 μJ was available at 532 nm, with nearly diffraction-limited beam quality. The pump pulse duration at 532 nm, deduced from cross-correlation measurements between the fundamental and second harmonic in a 3.26-mm crystal of BIBO (φ=90º, θ=146º), was ~40 ps.

The BIBO crystal used for the OPG stage was 5×10×10 mm³ in dimension and was also cut for type I (o→e+e) phase-matching at θ=155º (φ=90º), with a calculated effective nonlinear coefficient deff~3.36 pm/V [3]. The pump pulses at 532 nm were focused to a beam waist radius of around 550 μm into the OPG crystal using a f=50 cm focal length lens and the generated superfluorescence after one pass of the OPG crystal was separated by a dichroic mirror from the residual 532 nm pump. Two lenses of focal lengths f=10 cm were used for collimation of the superfluorescence and focusing of residual pump pulses. The dichroic mirrors were >90% transmitting for signal wavelengths over 650-1000 nm and >95% reflecting for the pump. The mirrors used in the delay line were >99% reflecting for the pump light. As shown in the experimental setup, the polarization direction of pump light was vertical, while the polarization directions of generated idler and signal were horizontal.
The spectral bandwidth of superfluorescence signal beam generated in the OPG stage was typically about 60 nm. To improve the output bandwidth, we used the scheme of OPG followed by bandwidth narrowing using a diffraction grating before injection into the OPA stage, as shown in Fig. 1. The collimated suprefluorescence signal from the OPG crystal was directed onto an 1800 grooves/mm holographic grating (Optometrics Inc.) and the first-order diffracted beam was injected into the OPA crystal. Using this scheme, the bandwidth of amplified OPA signal spectrum at wavelengths near degeneracy and at maximum pump intensity was narrowed to less than 2.5 nm, measurement restricted by the 0.3-nm resolution of the spectrometer (TRIAX 190, Jobin Yvon Inc.) used in our experiment. When the grating was replaced by a highly reflecting mirror at the signal wavelength, the bandwidth of the OPA output signal spectrum was increased to typically tens of nanometers. In addition, since only the central portion of the spatial seed beam which is overlapped with the pump can be amplified in the OPA stage, the separation between the OPG and OPA crystals also acts as a spatial and spectral filter. We used a delay line for temporal synchronization of the residual pump and generated superfluorescence seed pulses for optimum conversion in the OPA stage and a dichroic mirror was used to recombine the pump and superfluorescence pulses before amplification. The BIBO crystal used for OPA stage was a 5×10×10 mm$^3$ sample, cut for type I ($o\rightarrow e+e$) phase-matching at $\theta=175^\circ$ ($\varphi=90^\circ$). The waist radius of the 532 nm pump beam in OPA crystal was about 300 $\mu$m. Both crystals used in the OPG and OPA stage were uncoated. All BIBO crystals used in our experiments were grown by the top-seeded method.

3. Experimental results

Angular wavelength tuning was achieved by synchronous rotation of the OPG and OPA crystals and the grating at room temperature. The measured tuning data together with the calculated curve using the Sellmeier equation in Ref. 2 under collinear phase-matching condition are depicted in Fig. 2. Also shown is the corresponding variation in the effective nonlinear coefficient across the tuning range, calculated from Ref. 3. Taking account of the ±1 nm measurement accuracy of the available wavelength meter (WaveMate-P, Coherent, Inc.), reasonable agreement between experiment and theory is evident. As can be seen from the figure, the shortest signal wavelength under angle tuning at room temperature is ~740 nm. However, we were able to extend the output signal to significantly shorter wavelengths down to ~676 nm using temperature tuning under type I ($o\rightarrow e+e$) noncritical phase-matching along the optical z-axis in the same crystal. This is shown in Fig. 3, where signal tuning from 740 nm to 676 nm (corresponding to an idler tuning from 1893 nm to 2497 nm) was demonstrated by varying the temperature of the OPA crystal from 25 $^\circ$C to 200 $^\circ$C. The solid lines represent the calculated temperature tuning range using the thermo-optic dispersion equation of BIBO [17]. From the plot, good agreement between the experimental data and theoretical calculation is evident.
Fig. 2. Angular wavelength tuning range and corresponding effective nonlinear coefficient for collinear type I \((o\to e+e)\) phase-matching in the optical \(yz\)-plane of BIBO as functions of internal crystal angle, \(\theta\).

Fig. 3. Signal and idler tuning range as functions of temperature under type I \((o\to e+e)\) noncritical phase-matching in BIBO along the optical \(z\)-axis.

In order to obtain the highest OPG-OPA conversion efficiency at different output wavelengths, the narrowband superfluorescence seed wavelength from the OPG after the grating was tuned to the wavelength with the maximum parametric gain in the OPA stage. Otherwise, the output pulses from OPA displayed a double-peak spectrum, with one component at the input seed wavelength and the other near the maximum of parametric gain. In Fig. 4, the maximum output pulse energy and conversion efficiency in the signal and idler as functions of the OPA output wavelength are shown. The output energy data have not been corrected for the reflection losses at the crystal faces. The central part of data separated by the vertical lines are obtained from the angular tuning under type I \((o\to e+e)\) collinear phase-matching in the optical \(yz\) plane, the rest correspond to temperature tuning under type I \((o\to e+e)\) noncritical phase-matching along the optical \(z\)-axis. As evident from the plot, in OPA the maximum pump-to-signal conversion efficiency reached about 22\% at 790 nm, with the maximum pump to signal and idler conversion of 30\%. The efficiency in our experiment is larger than the maximum pump-to-signal efficiency of 13\% obtained with 9-mm-length...
BBO [18] and the maximum pump to signal and idler conversion efficiency of 24% obtained with 10-mm-length LBO [19] in an OPA pumped with picosecond pulses at 532 nm. Across the entire angular tuning range from 740 nm to 1892 nm, the single-pass pump to signal and idler conversion efficiencies in excess of 12.7% with maximum value up to 30% and maximum signal pulse energies of up to 48.6 μJ at 790 nm were obtained for the maximum pump energy of 220 μJ in the OPA crystal. We also did not observe any saturation in efficiency at pump intensities up to the maximum value, implying the possibility of further energy scaling of output. The increased efficiency in our experiment is a result of the high effective nonlinearity of BIBO compared with BBO and LBO [3, 15, 20]. We also recorded the stability of OPG-OPA output pulse energy. The standard deviation in the output signal energy of 1000 pulses was around ±1.6%, which was close to but larger than the standard deviation of pump pulse energy of ±0.8%. This is to be expected, given the nonlinear nature of the process and the relatively high pump pulse intensities involved here.

Temporal characterization of the OPA output signal pulses were performed using non-collinear cross-correlation between the Fresnel-reflected pump pulses from the first face of the OPG crystal and the OPA output signal pulses in a 2-mm-length BBO crystal (θ=20º), and recording the generated pulse energy as a function of delay between the two pulses. A typical background-free cross-correlation of the output signal near 800 nm, at the maximum output pulse energy, is shown in Fig. 5. The de-convolution of the cross-correlation result in Fig. 5 with that of the pump pulse, and assuming a Gaussian temporal profile, leads to a signal pulse duration of 18 ps.

Fig. 4. Output pulse energy and conversion efficiency of the BIBO OPA as functions of wavelength. The filled squares and circles represent signal and idler output pulse energies, respectively. The open squares and circles represent the pump-to-signal and pump-to-idler conversion efficiency, respectively.
To extend the OPG-OPA tuning range to shorter wavelengths in the visible and near-UV, we used a third crystal of BIBO for SHG of the amplified signal pulses. The BIBO crystal was 3×6×4.6 mm³ in dimension, and cut for type I (e⁺e⁻→e) SHG at θ=153° (φ=90°). The crystal faces were antireflection-coated over 790-850 nm and 395-425 nm. The generated signal pulses from the OPA were directed and focused into the SHG crystal using a 150 mm focal length lens. The scheme provided effective SHG of amplified signal pulses from 370 nm to 500 nm by synchronous rotation of the SHG crystal angle with the OPA output wavelength at room temperature. In Fig. 6, the maximum generated output pulse energy as a function of the wavelength is shown. The maximum energy in the second harmonic of the amplified signal pulses reached 10 μJ at 420 nm, corresponding to a conversion efficiency of 24% from the OPA to SHG. The generated SH tuning range with the maximum conversion efficiency is consistent with the theoretical analysis in Ref. 3. More detail discussion about this is out of the scope of the paper. Combining the SHG approach and OPA angular and temperature tuning, a broad spectral range from ~370 nm in the UV up to ~2497 nm in the infrared is achieved, which almost covers the optical transparency of BIBO from 286 nm to 2700 nm [2, 15].
We also investigated optical parametric generation and amplification in BIBO pumped by third harmonic of the Nd:YAG laser at 355 nm. In BIBO, with a UV absorption edge near 280 nm, two-photon absorption (TPA) can be a significant limiting factor in the attainment of the highest conversion efficiency in any frequency conversion processes involving visible and UV pulses, particularly in the presence of high pumping intensities [11,12]. In these experiments, we used a double-pass pumping configuration in a single BIBO crystal, as shown in Fig. 7. The third harmonic pump pulses at 355 nm were generated in two BIBO crystals using a scheme similar to that described in Ref. 12. The uncoated BIBO crystal was 5×10×10 mm³ in dimension, and cut for type I (o→e+e) collinear phase-matching in the optical yz-plane at θ=155º (φ=90º). As shown in the experimental setup, the polarization direction of pump light was vertical, while the polarization directions of generated idler and signal were horizontal. The maximum available pump pulse energy at 355 nm was 210 μJ in pulses with duration of 35 ps. After depletion in the first pass of the crystal for OPG, the energy of residual pump pulse in BIBO at 355 nm was 87.8 μJ in the second pass for amplification. In the second pass, the pump beam was focused to a waist radius of 380 μm in the crystal for the amplification of generated signal in the first pass with an appropriate delay. A wavelength tuning range of 450-1674 nm was achieved with the maximum pump to signal and idler conversion efficiency of 21% in amplification stage, providing amplified signal pulse energies of 14.2 μJ at 470 nm. This output energy has not been corrected for the reflection losses at the uncoated crystal faces.
To study the effect of TPA upon optical parametric process in our experiment, the pump-to-signal conversion efficiency with and without taking account of TPA loss in amplification stage was determined. The results at a signal wavelength of ~480 nm are shown in Fig. 8. The TPA coefficient, $\beta_o$, used in our calculation is 0.71 cm/GW [12]. As shown in the figure, at the input pump pulse energy of 42 $\mu$J (peak intensity of 0.5 GW/cm$^2$), corresponding to the onset of saturation in actual measured conversion efficiency (without taking into account of TPA), the calculated TPA loss at 355 nm in 10-mm BIBO crystal is ~10%. At the maximum pump pulse energy of 87.8 $\mu$J (peak intensity of 1.04 GW/cm$^2$), the calculated TPA loss at 355 nm in 10-mm BIBO crystal is ~18.7%. Comparing the pump-to-signal conversion efficiency between two curves in the Fig. 8, we can see that the saturation in the actual measured conversion efficiency may be mainly due to the strong two-photon absorption. But under the pump intensity in our OPG-OPA experiment pumped at 532 nm, we did not observe any significant TPA at 532 nm.
4. Summary and conclusion

In conclusion, we have demonstrated efficient generation of widely tunable picosecond pulses with optical parametric generation and amplification in BIBO pumped with SHG of an amplified mode-locked Nd:YAG laser, providing a broad tunable range from 676 nm to 2497 nm and total OPA conversion efficiency as high as 30%. The wavelength of the output radiation was effectively extended to ultraviolet band covering from 370 nm to 500 nm with a cascade crystal BIBO by SHG. We have also demonstrated tunable optical parametric generator and amplifier system based on BIBO pumped by third harmonic of the Nd:YAG laser at 355 nm in the presence of strong two-photon absorption, providing a spectral coverage across 450-1674 nm. The experiments demonstrate that BIBO is a highly promising material candidate for high-intensity picosecond OPG-OPA systems operating across the visible to near-infrared. Further work is in progress to study practical configurations with BIBO for output energy scaling to millijoule picosecond pulses and improving the overall system conversion efficiency by using larger aperture crystals and higher pump pulse energies.

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