

Communication

Green-Yellow-Orange-Red Spectral Range with Sum-Frequency Generation Using BIBO Crystal Pumped with an Optical Parametric Amplifier

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Abstract: In this letter, we show the broadband sum-frequency generation (SFG) in the Green-Yellow-Orange-Red spectral range using bismuth triborate, BiB_3O_6 crystal (BIBO) as a nonlinear material. We perform a noncollinear phase-matching configuration within the BIBO crystal using the remaining light behind the second harmonic generation stage and the infrared idler of an optical parametric amplifier (OPA). The obtained mixing radiation of ultrafast light sources to generate femtosecond pulses across the 520.5 to 742.5 nm region is observed. The SFG spectrum from a single-pass cross-correlation intensity over such visible range is shown. The SFG wavelengths as a function of the tunable wavelength idler OPA agree with the expectations of the parametric conversion condition and open the door to practical multi-beam or multi-color sum-frequency generators.

Keywords: sum-frequency generation; femtosecond pulses; noncollinear phase-matching

1. Introduction

The growing field of ultrafast light sources still requires multi-beam or multi-color systems motivated by scientific, medical, and industrial applications [1–4]. Recently, wavelength tunability and intense pulse are needed for industrial manufacturing [5,6], scientific research of medical device fabrication [7], spectroscopy techniques to analyze surfaces and interfaces [8], biological imaging with multiphoton microscopy methodologies [9], time-resolved experiments [10], and many other fundamental applications of modern science. Multi-beam and access to the new spectral region of femtosecond pulses are possible with parametric frequency conversion, ergo ultrafast optical parametric oscillator (OPO) [11], intracavity sum-frequency generation (SFG) of pump and signal in a synchronously pumped femtosecond tunable OPO [12], optical parametric amplifiers (OPA) [13], second and third harmonic generators [14–16] or, as we show here, sum-frequency generators.

Novel ultrafast systems constitute powerful devices for the generation of widely tunable coherent radiation with nonlinear materials. The progress of birefringent crystals as nonlinear material enables frequency conversion for the light generation in the new spectral region [17]. A nonlinear optical material such as bismuth triborate, BiB_3O_6 (BIBO) has shown exceptional nonlinear coefficients ($d_{\text{eff}} = 3.2 \text{ pm/V}$) for wavelength generation between 290 and more than 2500 nm [18]. Such a BIBO crystal has demonstrated the efficient tunable parametric generation of femtosecond pulses compared with other birefringent materials [19,20]. With a single BIBO crystal, the femtosecond optical parametric oscillator covers a visible range across 480–710 nm at room temperature operation [21]. Also, sum-frequency generation delivering visible pulses from 412 to 500 nm with a noncollinear

optical parametric oscillator (NOPO) was shown as an interesting configuration, operated with an output coupler with a simultaneous emission of infrared (IR) and visible (VIS) light [22]. In the case of amplified robust systems, such as noncollinear and collinear optical parametric amplifiers, are useful as light sources for ultrafast spectroscopy and many other fundamental and real-world applications in modern science [23]. The operation of noncollinear optical parametric amplifiers (NOPA) systems in continuous tuning between 550 and 700 nm and near-infrared are also demonstrated [24]. In the case of collinear optical parametric amplifiers (OPA) systems, see, e.g., a second harmonic generator (SHG) of 520 nm from femtosecond pulses of 1040 nm output of Ytterbium-doped Potassium-Gadolinium Tungstate (Yb:KGW) laser (Spirit-8-OPA, Spectra-Physics), the SHG stage is used as a pump of such OPA to generate in the infrared range signal (630–1020 nm) and idler (1040–2600 nm). Such an OPA system requires nonlinear conversion accessories to cover in the visible region, and the remaining 1040 nm light behind the SHG stage can still be used.

In this letter, we experimentally expose the BIBO crystal to generate the sum of the optical frequencies under two synchronous beams. One beam from the remaining 1040 nm of SpiritOne-8 Spectra-Physics laser, and the second from an idler optical parametric amplifier. Such beams were leading to a parametric output with higher frequency in the visible range. This procedure allows the nonlinear mixing radiation of ultrafast light sources to generate high-repetition-rate femtosecond pulses across the 520.5 to 742.5 nm region with a noncollinear configuration. Access to higher frequencies is possible with harmonic generators. Indeed, the collinear optical parametric amplifier systems require nonlinear optical harmonics to cover the UV-visible range [25]. The nonlinear frequency conversion presented here offers several advantages as it is obtained by the remaining fundamental 1040 nm pump light, allowing the single-pass noncollinear phase-matching condition on a broad spectrum, while at the same time providing a viable tool of tunable coherent light.

2. Materials and Methods

We perform a tunable visible light with noncollinear phase-matching configuration [26–28] that allows parametric frequency conversion with the help of BIBO as a nonlinear material. BIBO crystal was mounted on a rotation stage (RS) for changing the phase-matching angle. The two beams coming from different arms overlap with a fixed angle of $\alpha = 14^\circ$ within a BIBO crystal. The remaining 1040 nm and the IR idler beams are focused and crossed inside a BIBO crystal with an orientation condition to the SFG with noncollinear phase-matching. In our case, both focused beam waist radii are close to 50 micrometers. The sum-frequency generation of the pulses is achieved in a single pass of 500 μm crystal. The crystal is a type I interaction in the yz plane ($\varphi = 90^\circ$) at an internal angle of $\theta \approx 161^\circ$ at normal incidence.

The setup for the parametric frequency generation is shown in Figure 1. For our sum-frequency generation experiments, we used SpiritOne-8 and SpiritOPA systems emitting a 20 kHz train of <400 fs pulses.

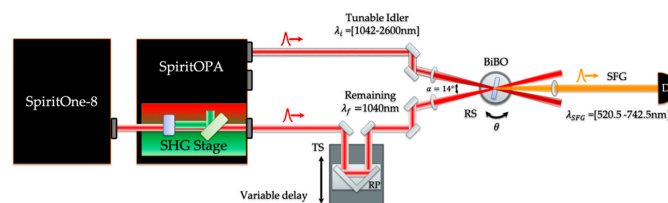


Figure 1. Schematic view of the experimental setup used for measuring the sum-frequency generation (SFG) across the 520.5 to 742.5 nm region from two femtosecond pulses superimposed within a BIBO crystal. The BIBO crystal is mounted on a rotation stage (RS) in a noncollinear phase-matching configuration with a fixed angle $\alpha = 14^\circ$. The variable delay line is used to temporally overlap the remaining pulse ($\lambda_f = 1040$ nm) behind the second harmonic generation (SHG) stage of SpiritOne-8 laser and the λ_i pulse from the tunable range 1042–2600 nm of the automated collinear optical parametric amplifier (SpiritOPA) with the help of a translation stage (TS) and a retroreflector prism (RP).

3. Results

The present study demonstrates the functionality of a BIBO crystal in combination with an optical parametric amplifier (OPA) to generate high-repetition-rate femtosecond pulses across the 520.5 to 742.5 nm region. The IR idler source is a computer controller tunable range 1042–2600 nm from the automated collinear SpiritOPA. Such an IR idler pulse of SpiritOPA is tuned to pump simultaneously with the remaining 1040 nm pulse. Both pulses overlap in the crystal with different focal lenses to ensure the maximum output power. Light emerged at the focal plane as a sum-frequency generation that it is produced in the middle of the two beams. This shows that SFG will be separated from the IR light beams because the proper phase-matching and the intensity generation is measured (Figure 2c). The focal plane of this sum-frequency generated radiation is imaged in turn by a focal lens onto the input plane of a detector. Detector D can be a power meter to measure the average power or a high-resolution spectrometer to measure the resultant SFG spectrum.

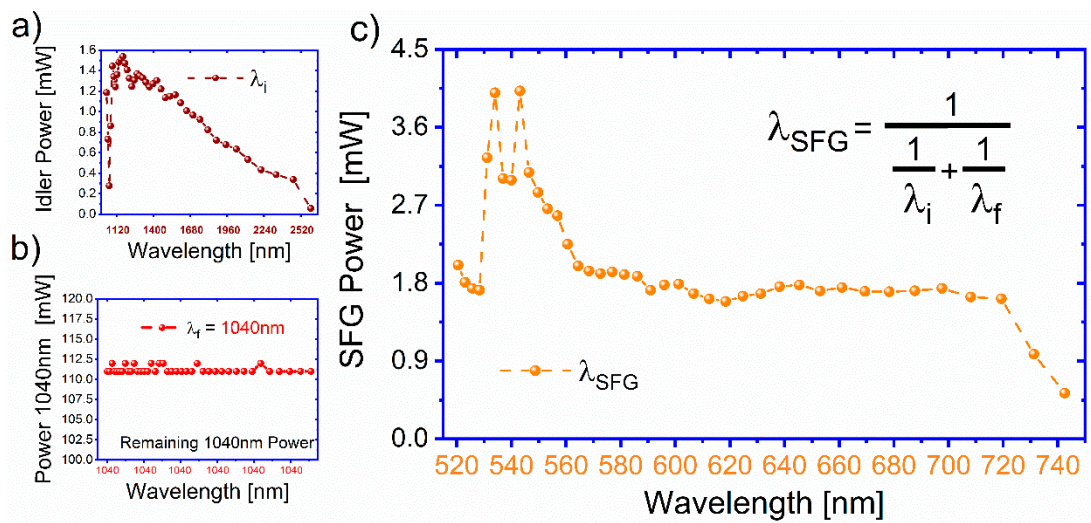


Figure 2. SFG average power using two infrared pump beams. The SFG measurement over a set of wavelengths that employs a noncollinear phase-matching configuration, which is directly pumped from two femtosecond pulses superimposed within a BIBO crystal. (a) One pulse is from the tunable range 1042–2600nm (wine line and average power values), (b) the second pulse is the remaining 1040 nm light (red line and average power values). (c) The resultant SFG as a function of the wavelength range of 520.5 to 742.5 nm. The orange dashed lines are only a guide for the eye.

We have performed a systematic measurement of the output SFG power with two infrared pump beams. One pulse is the remaining 1040 nm light, and the second pulse is the tunable idler OPA pump. The resultant SFG wavelength corresponds to the wavelength set as a function of the tunable OPA idler. The tunable idler OPA power is shown in Figure 2a (wine line and average power values). The remaining 1040 nm power that changes behind the SHG stage is <1% over a set of 1040 nm power measurements, as shown in Figure 2b (red line and average power values).

The following expression gives a typical SFG wavelength [23,29]:

$$\lambda_{SFG} = \frac{1}{\frac{1}{\lambda_f} + \frac{1}{\lambda_i}} \quad (1)$$

where $\lambda_f = 1040$ nm and λ_i is the tunable IR idler. To acquire the power of maximum in a SFG using pulses with different values of λ_i , the crystal was rotated to change the internal angle of the crystal with the gyration of the crystal holder for noncollinear phase-matching. Here, we achieved the wavelength range of 520.5 to 742.5 nm of such SFG from single-pass cross-correlation maximums, as shown in Figure 2c (orange line and average power values). Figure 3a shows the internal angle under

phase-matching of the BIBO crystal and conversion efficiency as a function of the SFG wavelength. We can extend the generation of light and the covered spectrum to visible with an angular wavelength tuning by rotation of the BIBO crystal and by varying the OPA idler.

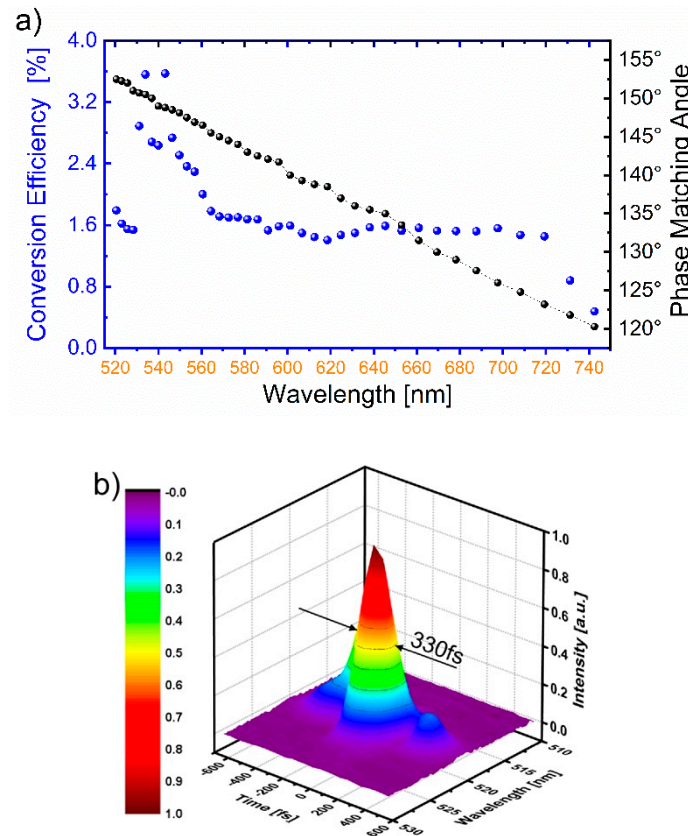


Figure 3. Tunable SFG by rotation of the BIBO crystal and by varying the optical delay line. (a) The internal angle under phase-matching (black circles) and conversion efficiency (blue circles) as a function of the SFG wavelength from 520.5 to 742.5 nm by changing the internal angle of the crystal between 152.5 to 120.3°. (b) Cross-correlation trace obtained by SFG with a reference pulse at 1040 nm and the idler at 1042 nm. A pulse duration of 330 fs is obtained.

The typical sum generation is presented in this work for reproducibility, a motorized rotation is used for controlled rotation of the crystal. A second motorized translation stage and a mounted retroreflector prism are performed for a controlled path length of a variable delay line. Such a pump pulse with a variable delay line is used for improving the temporal pulse overlap superimposed in the crystal because of group-velocity matching. Adjustment of the optical delay line is needed because changes in the angle of phase-matching produce small changes in the length of the crystal. With the help of the optical delay, a cross-correlation trace of sum-frequency generation is shown in Figure 3b from the 1042 nm pump of the OPA idler and the remaining 1040 nm light. The pulse duration of 330 fs is obtained with the cross-correlation intensity profile.

The normalized spectrum of SFG from the single-pass cross-correlation intensity over such a visible range is shown in Figure 4. In this measurement, light that emerged as a sum-frequency generation is coupled to a high-resolution spectrometer (HR4000) with an optical diffusor.

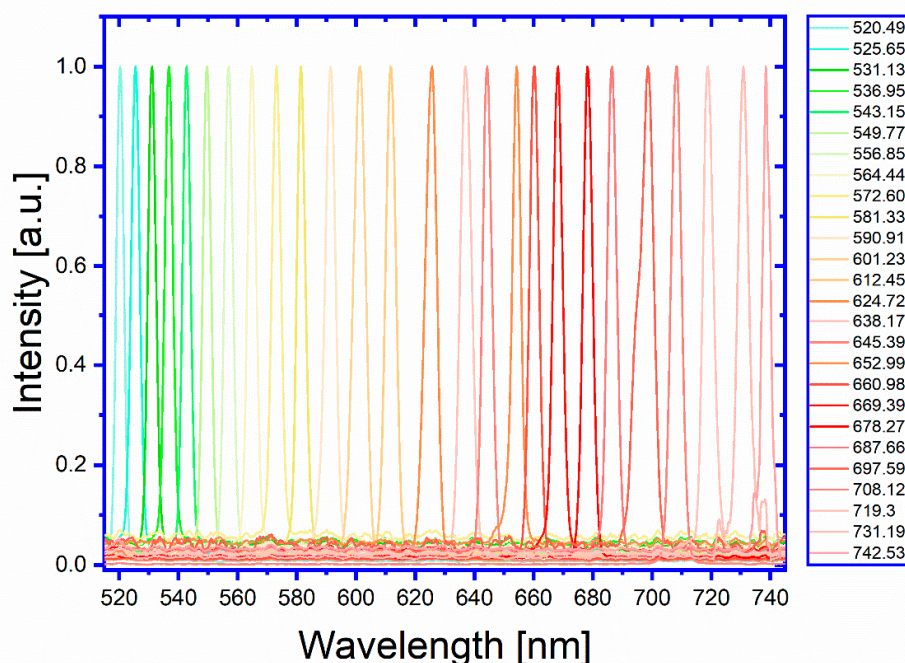


Figure 4. The resultant SFG spectrum. The spectrum shows the demonstrated tuning range of sum-frequency generation from 520.5 to 742.5 nm with normalized cross-correlation intensity measurements using a spectrometer HR4000 (Ocean Optics) as a detector.

4. Discussion

The nonlinear frequency conversion as a sum-frequency generator presented here offers several advantages as it is achieved with the remaining fundamental 1040 nm pump light, allowing single-pass noncollinear phase-matching conditions on a broad spectrum, while at the same time providing a viable tool of tunable coherent light. The obtained mixing radiation of ultrafast light sources to generate femtosecond pulses across the 520.5 to 742.5 nm region is observed from Figure 2c with the help of BIBO as a nonlinear material. It is important to note that the maximum SFG power corresponds to the maximum idler pump average power. The smallest power value close to 742 nm in the tuning range is limited by the infrared absorption cutoff on a BIBO crystal. Recently, Ghotbi et al. showed that BIBO exhibits an absorption spectrum feature below the 2.7 μm wavelength and transparency of the used BIBO in combination with idler absorption limits the spectral tunability [30]. In addition, the BIBO crystal has shown the possibility to efficient widely tunable optical parametric generation (OPG) and amplification (OPA) of picosecond and femtosecond pulses pumped by second and third harmonics of Nd:YAG laser and also extended the tuning range by pumping the amplified signal in BIBO with a second harmonic generation range from 370 to 500 nm [31]. The frequency conversion with noncollinear phase-matching is an effective scheme that provides tuning coverage across the transparency of the BIBO crystal and the corresponding magnitude of the significant nonlinear coefficient as a function of pumped wavelengths [32]. In our SFG configuration, the Green-Yellow-Orange-Red spectral range can be generated, and a tunable wavelength system may be implemented as a conversion accessory of an optical parametric amplifier idler with the help of the remaining 1040 nm light. The configuration can provide both visible and near-infrared outputs pulses, and it is possible to extend the tuning range by pumping the amplified idler in such a BIBO crystal with the second harmonic generation range discussed in detail in the previous literature [17–21]. Figure 4 shows the experimentally measured spectral range of the generated visible sum-frequency generation, as observed with a high-resolution spectrometer. It is also important to note the symmetric feature of a 3.1-nm spectral bandwidth found near 520.5 nm, 3.68 near 572.6 nm, and 4.93 near 601 nm. It can be shown from Figure 4 that the changes in the bandwidth of such spectral range may be caused by a length detuning of the optimal

phase-matching of the angle crystal and the adjustment of path length to improve the sum-frequency generation of such overlap. The maximum conversion efficiency is more than 3% in a single pass collinear configuration presented here with a BIBO crystal of length of 0.5 mm. A higher SFG output power and efficiency may be obtained using longer crystals, as found in [18]. Figure 3b illustrates the cross-correlation intensity profile of the sum-frequency generation pulse at 520.5 nm. The measured pulse duration is 330 fs, corresponding to the cross-correlation trace of the idler pulses obtained by sum-frequency generation with a 1040 nm pulse.

Our configuration with the remaining 1040 nm light has shown to be an alternative conversion accessory in both scientific and industrial work to extend the spectral range to Green-Yellow-Orange-Red. Such an SFG output is emitted simultaneously with the IR pumps, so the noncollinear simultaneous emission of 1040 nm, idler, and visible range makes it well suited for ultrafast spectroscopy/microscopy [22]. Characterization of the materials is also a possible application measuring two pulses simultaneously as the implementation in [33]. Our nonlinear optical results open the door to high-resolution nonlinear spectroscopy/microscopy applications, as it offers the possibility of selective wavelength in a single-pass noncollinear configuration.

5. Conclusions

In conclusion, we have shown a broadband sum-frequency generator using the remaining fundamental 1040 nm light of a femtosecond laser source and IR idler from the optical parametric amplifier. The sum-frequency generation into a visible range is possible with noncollinear phase-matching in a BIBO crystal. We provide wide tuning across the Green-Yellow-Orange-Red spectral range. The direct separation of three interacting waves is demonstrated with a simple cross-correlation configuration. Such configuration might be useful to perform practical multi-beam or multi-color experiments to scientific, medical, and industrial applications.

Author Contributions: All authors contributed equally to this work. J.L.D.-J. and R.Q.-T. conceived the project. Conceptualization, J.L.D.-J. and R.Q.-T., designed and implemented the experimental setup; methodology, J.L.D.-J. and R.Q.-T. contributed extensively to the planning, discussion and writing up of this work; validation, J.L.D.-J. and R.Q.-T.; formal analysis, J.L.D.-J. and R.Q.-T.; investigation, J.L.D.-J. and R.Q.-T.; resources, J.L.D.-J. and R.Q.-T.; data curation, J.L.D.-J. and R.Q.-T.; writing—original draft preparation, J.L.D.-J.; writing—review and editing, J.L.D.-J. and R.Q.-T.; visualization, J.L.D.-J. and R.Q.-T.; supervision, J.L.D.-J. and R.Q.-T.; project administration, J.L.D.-J. and R.Q.-T.; funding acquisition, J.L.D.-J. and R.Q.-T. All authors have read and agreed to the published version of the manuscript.

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