Sum and difference frequency generation of white light continuum with the ps pulses of Nd$^{+3}$:YAG laser in a thick BBO crystal

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Abstract

The white light continuum (WLC) generated in water/D$_2$O mixture by pumping with the fundamental of ps Nd$^{+3}$:YAG laser has been used as a variable frequency source for the sum frequency generation as well as for its amplification. 35 ps long pulses with 8 mJ energy at 1064 nm were mixed collinearly with the WLC generated by the same laser beam in a 20 mm thick BBO crystal. The obtained tunable output has been identified as the sum frequency between the fundamental and a portion of the WLC with the required phase matching. Theoretical simulations are also given along with a few initial experiments to use this combination for the difference frequency generation (optical parametric amplification) under non-collinear geometry.

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Keywords: Nonlinear optics; Sum frequency generation; Optical parametric amplification

1. Introduction

Ultrashort pulses have attracted a considerable attention due to their numerous applications in higher order harmonic generation and generation of soft X-rays [1–4]. During the last decade, the optical parametric amplification has been identified as one of the best techniques for the generation of the tunable pulses using chirped pulses [5–11]. Sum frequency generation has recently been used to extend the tunability range in femtosecond (fs) regime [12].

Optical parametric amplifiers (OPAs) have higher gain per unit length than the laser amplifier. In fs time scale, it is easier to handle both, the higher order phase distortion due to the material dispersion and nonlinear phase distortion due to self-phase modulation. In general, OPA utilizes instantaneous nonlinear interaction of spatially and temporally overlapped pump and the signal pulses. Depending upon the type of the crystal (Type I or II), interacting wavelengths and the pump intensity, gains of the order of $10^8$ can be achieved with the OPA [13]. Non-collinear optical parametric amplifier (NOPA) geometry on the other hand, aims to obtain broad amplification bandwidths by broadband phase matching, and consequently, the necessary spectral width to support extremely short pulses of fs time durations [14].

Recently an OPA pumped by nanosecond (ns) pulses has been shown to amplify the chirped fs pulses in a single pass scheme [15]. In picosecond (ps) time scale, Raman shifting of lasers is used for selective tuning [16]. Various other pump and seed combination have also been reported for optical parametric generation [17–19]. However, by using the white light continuum (WLC), enhancing the tunability range of the ps pulsed Nd$^{+3}$:YAG lasers does not seem to have been investigated in the past. In view of the above, in the present paper we report (i). tunable output generation by using the sum frequency of WLC and the fundamental of Nd$^{+3}$:YAG laser and (ii). the prospects of the amplification of WLC generated by ps pulses under...
non-collinear geometry by pumping it directly with the second and the third harmonics of the ps Nd$^{3+}$:YAG laser.

The paper is organized as follows. Section 2 gives the details of the experimental set up used in the study. Section 3 (Results and discussion) has been divided into three parts. The first part gives the method of generation of WLC in water/D$_2$O mixture. The second part gives the analysis of the experimental results on sum frequency generation between the fundamental of the ps laser and the obtained WLC. In third part, the non-collinear optical parametric amplification scheme is detailed under two headings containing theoretical simulations and experiments. Conclusions are given in Section 4.

2. Experimental

The experimental set up used in this study is shown in Fig. 1. A fraction of the output from the fundamental of Nd$^{3+}$:YAG laser (1064 nm, 8 mJ, 35 ps) was converted into single filament WLC by focusing it with a 1000 mm lens into a 100 mm long glass cell containing the water/D$_2$O mixture [20]. For sum frequency generation, the WLC and the fundamental were focused on the BBO crystal.

The second harmonic of the laser (532 nm, 35 ps, 2 mJ) was also used as the pump for the difference frequency generation as well as for the experiments on amplification in a non-collinear geometry. After passing through the delay line, the pump beam was focused by a concave mirror of 500 mm focal length into a 20 mm thick BBO (Type I) crystal cut at 21.9°. The irradiance of the pump beam was estimated to be 14.5 TW/cm$^2$ at the focus of the mirror, but the amplification of WLC was achieved at the irradiance value of 7.3 GW/cm$^2$ by keeping the crystal surface ~30 mm far from the pump focus to minimize the burning problems. For calibration purpose, the optical path was adjusted by the sum frequency generation of a tiny portion of 1064 nm with the pump (532 nm) by using a suitable BBO crystal. A spectrometer (Ocean optics, HR 2000) was used to record the spectra. A photodiode (Becker & Hickl, model PDI 400) was used to measure the efficiency of the amplification.

3. Results and discussion

3.1. WLC generation

As described above, the WLC was obtained by focusing the fundamental laser beam from the ps laser into a glass cell containing the water/D$_2$O mixture. The quality of the generated WLC was improved by varying the spot size of the laser beam inside the glass cell by moving the cell along its axis in the focal plane of the lens. The spectral and the beam shape fluctuations of the WLC were within 10% when pump energy was kept above its threshold value. Fig. 2 shows a typical spectrum and a photograph of WLC obtained under ps pumping. It is seen that there are a few major peaks with an underlying broad background. These peaks are due to the OH/OD vibrations and include the overtones of the stimulated Raman scattering frequency [21]. The WLC so obtained was used as the seed (signal beam) in the setup shown in Fig. 1.

3.2. Sum frequency generation of WLC and 1064 nm

In order to look at the sum frequency generation of WLC with 1064 nm, the path followed by the 532 nm was blocked in Fig. 1. The fundamental beam (by removing the IR filters) along with WLC (seed) was focused collinearly on the BBO crystal. The phase matching angle of
the BBO was 21.9°. In this geometry, the temporal and spatial mismatch between the two frequencies is guaranteed within the duration of the ps pulse. Upon tilting the BBO, radiation in the blue-green region was observed. The spectra of obtained signals are shown in Fig. 3. It was noticed that (i) the output wavelength depends upon the BBO tilt from its optical axis, (ii) the full width at half maximum (FWHM) of some of the spectra were remarkably broad, and (iii) the radiation was observed on both sides of the crystal due to internal reflection from its surfaces.

Various possibilities such as the sum frequency generation, the optical parametric oscillation in a cavity with the length matched with that of the BBO crystal can be proposed to explain this effect. In addition, the generation of the higher harmonics in the first few mm of the long crystal that act as the pump for the WLC (giving rise to the SFG with 1064 nm) can be attributed as follows. The broad 810 nm band of the WLC when mixed in the 21.9° crystal up to a maximum tilt of 4° from the optical axis is suitable for its sum frequency generation (SFG) with 1064 nm. This is due to the fact that the phase matching angle of BBO crystal for the obtained wavelength range for the SFG varies from 23° to 26°. With the available area of cross section (4 × 4 mm²) of the long BBO crystal (20 mm) used in the study, the SFG of the other strong components of the WLC (such as those centered at 550 nm or 650 nm) were not accessible.

The broader spectrum obtained at 482 nm for instance, may indicate towards obtaining the shorter pulsewidths after suitable compression. The pulse width (∆t) for the measured spectrum can be estimated by using the relation

\[ ∆t = 0.441λ^2/cΔλ; \]

where \( λ \) is the peak wavelength and \( c \) is the speed of light. However, the group velocity dispersion experienced by pulses while propagating through the optics (water/D₂O cell, and the length of the BBO crystal) will have to be taken into account before giving any final value. In addition, the resolution of the spectrometer (~5 nm) is very close to the obtained value of ∆λ. Table 1 gives the peak positions, the FWHM of the obtained spectra under this geometry, and the corresponding estimated pulse widths that may be obtained after suitable compression.

### 3.3. Prospects for optical parametric amplification

In optical parametric amplification (OPA), a high frequency pump beam at frequency (\( ω_p \)) amplifies a lower frequency signal beam (\( ω_s \)), in addition to the generation of a third idler beam (\( ω_id \) with \( ω_id < ω_s < ω_p \)), according to the phase matching condition

\[ h\bar{k}_p = h\bar{k}_s + h\bar{k}_id, \]

where \( k_p \), \( k_s \), and \( k_id \) are the wave vectors of the pump, the signal and the idler beams, respectively. In this process, the photon of the short-wavelength pump beam is destroyed to create a signal and an idler photon in an active medium, according to the energy conservation:

\[ ω_p = ω_s + ω_id. \]

For efficient conversion, the phase velocities of the pump, signal and the idler beams are matched by proper orientation of birefringent crystal.

In the fs regime, the phase matching does not ensure the matching of the group velocities of the three beams. As a result, three pulses propagate with three different speeds in the crystal. Also under this condition, the phase matching bandwidth is fixed as the signal and the idler velocities are fixed. Under such circumstances, by using a non-collinear geometry, the pump and the signal beams can be sent at a non-collinear angle (\( ζ \)) on to the crystal and the idler is emitted at another angle \( Ω \) with respect to the signal beam as shown in the Fig. 4. The broadband phase matching is obtained when the following condition for the group velocities of signal (\( V_{gs}(s) \)) and idler (\( V_{g(id)} \)) are fulfilled [22]:

\[ V_{g(s)} = V_{g(id)} \cos Ω. \]

### Table 1

The peak wavelengths and FWHMa for the obtained spectra (Fig. 3) under collinear geometry

<table>
<thead>
<tr>
<th>No.</th>
<th>Peak wavelength (nm) ±2 nm</th>
<th>FWHM ±1 nm</th>
<th>Corresponding curve in Fig. 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>445</td>
<td>10</td>
<td>a</td>
</tr>
<tr>
<td>2</td>
<td>460</td>
<td>8</td>
<td>b</td>
</tr>
<tr>
<td>3</td>
<td>463</td>
<td>7</td>
<td>c</td>
</tr>
<tr>
<td>4</td>
<td>482</td>
<td>13</td>
<td>d</td>
</tr>
<tr>
<td>5</td>
<td>490</td>
<td>8</td>
<td>e</td>
</tr>
<tr>
<td>6</td>
<td>502</td>
<td>11</td>
<td>f</td>
</tr>
</tbody>
</table>

a FWHM of the pump pulse (1064 nm) is 5 nm.
However, for the present case, the long BBO crystal and the pump pulse duration of 35 ps, the group velocity mismatch does not matter. In ps or ns times scales, the non-collinear geometry simply helps to separate the signal from the pump and the idler frequencies; it thus eliminates the need of dispersing elements to separate the beams [23].

3.3.1. Theoretical simulations for two pump wavelengths: retracing behaviour and broad band phase matching

Fig. 5 gives the typical theoretical simulations [24] for the two pump wavelengths at non-collinear angles for BBO (type I) crystal. These simulations provide a preliminary estimate of the phase matching angles for the amplification of the required signal wavelengths. A corresponding idler wavelength is also generated. As seen from the figure, the two wavelength curves meet at a single point in both the simulations. This point is known as the degeneracy point at which the signal and the idler wavelengths are common for a given pump. In addition to this, for several nonlinear optical crystals, another phenomenon called the ‘retracing behaviour’ is observed due to their dispersion properties. This retracing behaviour is also seen in the both the simulations for the selected non-collinear angles in the form of bends: (i) near 490 nm and 2500 nm in panel A and (ii) near 650 nm, 900 nm, 1200 nm and 2000 nm for panel B. This indicates that for example in panel A, in principle, if two signal frequencies are available within such regions, both of them may be amplified, besides generating two idler frequencies. This kind of behaviour has been actually observed by Liu et al. [25]. The retracing behaviour is also observed in temperature tuning of lithium borate by Zhao et al. [26]. The inset of panel B indicates the possibility of broad band phase matching near the angles of 22.7º.

3.3.2. Experiments on difference frequency generation

Towards experimental verification of these simulations, we planned experiments for the difference frequency generation. In Fig. 1, the temporal and the spatial overlap between the pump and the signal beam paths was achieved by self-diffraction from a dye film [27]. The alignment of the set up was cross checked by the sum frequency generation of 532 nm and 1064 nm in a BBO Type II crystal cut at an angle of 31.3º. Further, the difference frequency generation between the fundamental of Nd³⁺:YAG laser and 532 nm was achieved. For this purpose a 20 mm thick BBO crystal (Type I, 21.9º) was used. Amplification factors upto ~210 were obtained under this geometry for 1064 nm (Table 2). This

Table 2

<table>
<thead>
<tr>
<th>Signal energy (nJ)</th>
<th>Amplification factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.8</td>
<td>40</td>
</tr>
<tr>
<td>8</td>
<td>80</td>
</tr>
<tr>
<td>200</td>
<td>210</td>
</tr>
</tbody>
</table>

Fig. 5. Tunings curve for phase matching in a non-collinear geometry for signal and idler wavelengths when pumped with 355 nm (a), and 532 nm (b). The degeneracy points are observed at 532 nm (panel A) and 1064 nm (Panel B) in addition to the retracing behaviour in signal and idler arms in both the cases. The inset of Panel B indicates the possibility of broad band phase matching near the angles of 22.7º.

Table 2

Obtained amplification factors for the 1064 nm (signal) when pumped with 532 nm (energy = 1.5 mJ) under non-collinear geometry
confirmed the alignment of the required setup for the non-collinear geometry of parametric amplification. Under the collinear geometry, amplification has been reported for the WLC obtained by pumping the 1064 nm earlier [28].

In another experiment, the WLC was passed through a filter to block the fundamental and was focused on the BBO crystal along with the pump (532 nm). Fig. 6 gives a typical spectrum with a peak at 630 nm obtained under this geometry. Since the BBO crystal had a thickness of 20 mm with a cross section of $4 \times 4$ mm$^2$, the phase matching angle is relatively well defined. The phase matched output signal and the idler frequencies are governed by the angular tuning curves shown in Fig. 5 (panel B). It can also be seen that for a non-collinear angle of 2–2.5°, the crystal angles of 22° to 23° are suitable for the amplified wavelength. The amplified signal was weak, but several tens of times stronger than the WLC. Except another spectrum peaked at 628 nm, we were unable to observe any other wavelengths within the limits of the tilts restricted by the unblocked paths of the pump and WLC in to the BBO. Further increase in the pump energies resulted in the burning of the BBO crystals under ps pumping. Larger frequency tuning of the amplification may be achieved by choosing another BBO crystal suitable for other portions of the WLC accompanied by the subsequent adjustment of the delay in the pump beam. The spectral width ($\Delta \lambda$) of the amplified spectrum was 10 nm, as recorded by the spectrometer of resolution of ~5 nm.

4. Conclusions

Generation of tunable wavelengths in the collinear geometry due to the sum frequency generation between the WLC and the 1064 nm is experimentally demonstrated here. Single-stage amplification of the fundamental (1064 nm) of a ps Nd$^{3+}$:YAG laser in a non-collinear geometry has been obtained. The amplification has also been achieved for the WLC obtained by pumping the water/D$_2$O mixture by ps pulses at 1064 nm. These results can open avenues for further research to obtain high power tunable pulses in the ps time scales.

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